

UNIVERSITÀ DEGLI STUDI DI CATANIA

Tales of clustering in the microphysics world

by

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Abstract

The works presented in this thesis unfold between the deeply intertwined realms of nuclear physics and chemistry: despite the ordinary perception which may lead to consider these two sciences as separated, the picture appears completely different when diving deep in the study of some peculiar characteristics and phenomena. Symmetries, interactions, reactions, excitation modes and the concept of molecular assemblies itself are, in fact, not exclusive of neither of the two frameworks.

These analogies are firstly presented through a detailed and organized parade following the historical steps in the development of several models for the description of the simplest atoms *and/or* nuclei, and of the interactions between their constituents. The cases in which a chemical approach has proved itself useful for the description of a nuclear phenomenon, and *vice versa*, are not rare, and will be broken down along the Sections in the first Chapter. These include the development of models for nuclear and chemical potentials, for intermolecular and inter-cluster interactions and for the study of clustering in light nuclei and chemical solutions. On the basis of these models and analogies, different investigation techniques are thus presented in Chapter two before entering the experimental heart of the matter.

Three Chapters are indeed dedicated to the experimental investigations conducted for three of the most important self-conjugate nuclei: ^{12}C , ^{16}O and ^{20}Ne . The understanding of the spectroscopy and of the occurrence of cluster configurations in their ground and excited states, is extremely important to unveil some sketchy features and to solve long-standing questions about crucial aspects of astrophysical and nuclear physics phenomena. The development of cluster structures in their *ground* states (for ^{12}C and ^{16}O) was studied through the applications of a so-called *crystallographic* model, while the features of some important excited states (such as the *Hoyle state* in ^{12}C) have been investigated through low-energy nuclear reactions, designed and performed with fine state-of-the-art set-ups and techniques.

All comes to an ending with a radiochemistry experiment representing the closure of a cycle, where nuclear physics phenomena (such as neutron radiative capture and nuclear recoil) are finally exploited to investigate the bizarre behaviors of radioactive atoms inside a solution, not describable through the exclusive application of ordinary chemistry approaches.

Acknowledgements

The acknowledgments will appear in the printed version of this Ph.D Thesis

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For/Dedicated to/To my...

Chapter 1

Forces and bindings in the microphysics world

In our universe, every entity, individual, object, and particle is tied to other ones by means of different types of *bonds*. Aside from any type of bond which can be classified in the "abstract" or "emotional" domain, in the long await for an unified theory, all those known to the mankind can nowadays be considered as derived from four fundamental forces: the *gravitational* attraction, the *electromagnetic* interaction, and the *strong* and *weak* nuclear interactions. Each one of these forces has its characteristic range, specific strength and response to the distance of the interacting partners. For example, the gravitational force, which inherently depends from the mass of an object, is always present and can only be observed as an *attractive* interaction, pulling the bodies together with an intensity inversely proportional to the square of their respective distance. On the other hand, the electromagnetic and the nuclear interactions can be attractive or repulsive depending on, respectively, the intrinsic nature of the interacting partners and their distance; the electromagnetic force is repulsive when the interacting bodies have the same nature, while it is attractive when objects of opposite nature are facing each other. The nuclear force, however, has an overall attractive nature, but its characteristics are strongly and non-linearly dependent on the distance of the two partners, in this resembling the *Lennard-Jones* forces typical of molecular interactions [1, 2]. Although the gravitational force is always present, it can be completely neglected while treating physical or chemical cases involving atoms, nuclei and molecules, since its intensity is several orders of magnitude smaller than that of the nuclear and electromagnetic forces; actually, the gravitational force mainly play a sizable and observable role in the study of the dynamic properties of macroscopic objects and in the astronomical study of the planets' motion. The electromagnetic interaction is the main actor in the definition of the stability of atoms and molecules and the responsible for the stability of these systems. The *strong* nuclear force is, as stated in the name itself, what makes an atomic nucleus stable, opposing the electrostatic repulsion between the protons, which would therefore make impossible for the nucleus to hold a stable structure. The *weak* nuclear force, on the other hand, acts between leptons and quarks, and allows to model and study the properties of some unstable nuclei and the basis of their radioactive decays. The aim of this Chapter is to discuss the analogies and differences between the chemical bonds, built on the electromagnetic interaction, and the nuclear forces present between the different components of an atomic nucleus, especially in the presence of stable clustered structures. Such analogies can be precious for the understanding of α clustering in nuclei,

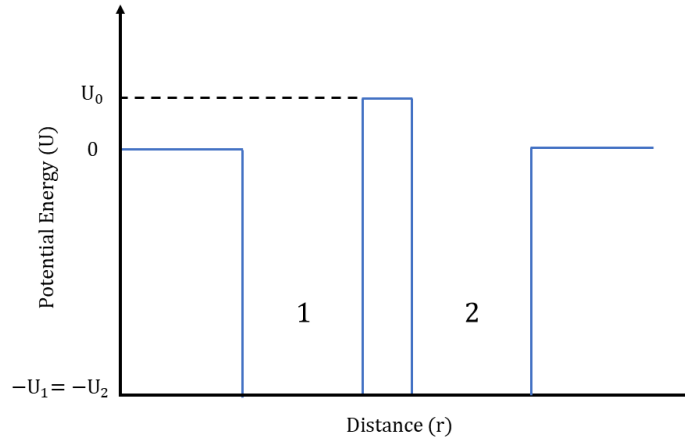


FIGURE 1.1: Sketch of two identical and close-lying potential wells, separated by a finite barrier of energy U_0 .

considering that, differently from the electromagnetic force, some important details on the behavior of nuclear force are today still shrouded in mystery.

1.1 Atomic interactions and molecule formation in the quantum world

Two roads are viable to understand how a system of interacting particles is quantum-mechanically composed: either one solves a Schrödinger equation with a custom, complicated, potential (not without any difficulties) or uses approximated methods. The latter may of course appear as limited, but allows to have a point of view general enough to describe, on different energetic and spatial scales, some phenomena that may seem different, like those related to *nucleons* and *molecules*, at least from a first glance. Several important phenomena, based on parallelisms and analogies between the molecular and the atomic worlds, will be discussed in the following Sections.

1.1.1 The double potential well

Among the approximate methods discussed in the above paragraph, a very useful (and general) one, which allows to model the behavior of two interacting particles, is that of the *double potential well*. In this case, two identical potentials with the same well depths $U_1 = U_2$ are separated by a barrier of a given energy U_0 , as sketched in Figure 1.1; of course, if U_0 were to be infinitely high, the energy states of a particle in one of the two wells would be stationary states of identical nature, and the wells would form two completely separated systems.

However, if one considers ψ_1^2 as the probability to find the particle in the first well, and ψ_2^2 as that of finding it in the second well, the tunnel effect would always allow the interchange of the particle between the two potential wells. In general, the possibility of having the particle in the first well would be equal to having it in the second one, and the actual finding probability can be written as $\psi^2 = \frac{1}{2}(\psi_1^2 + \psi_2^2)$. Note that it is possible to obtain the same result by elevating each one of the following functions (ψ_s ,

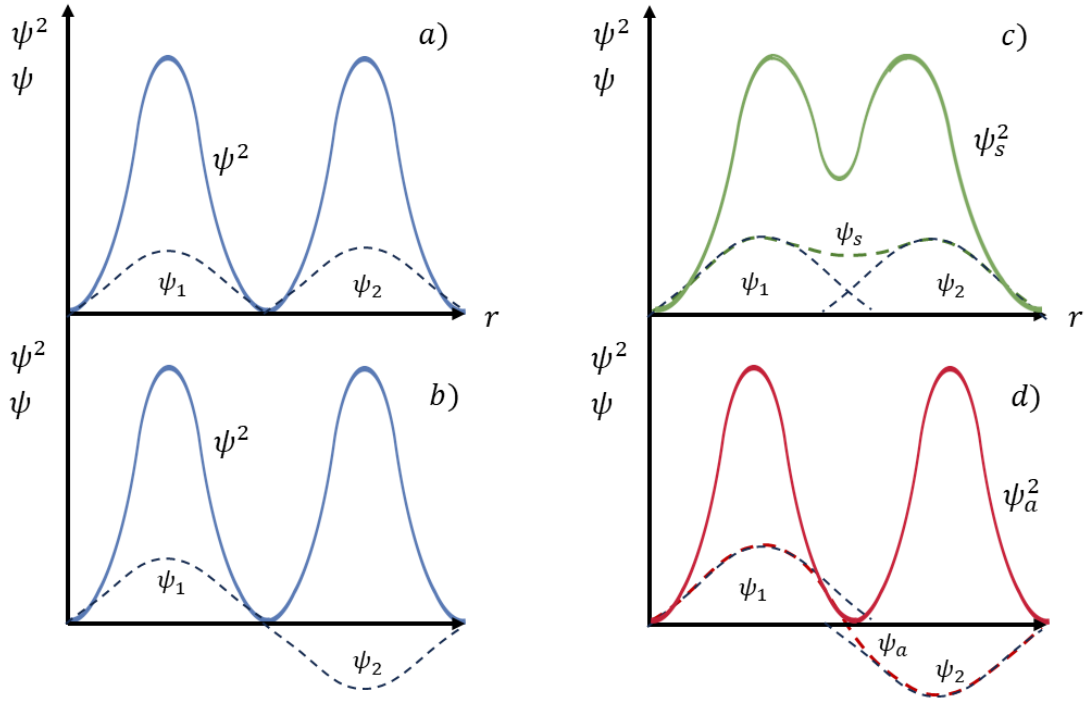


FIGURE 1.2: Left (a, b): wavefunction and probability of presence of distant particles; Right (c, d): Symmetric and asymmetric wavefunction of two particles, taking exchange phenomena into account.

the *symmetric* function of the system, and ψ_a the *anti-symmetric* one) to the second power:

$$\psi_s = \frac{1}{\sqrt{2}}(\psi_1 + \psi_2); \quad \psi_a = \frac{1}{\sqrt{2}}(\psi_1 - \psi_2) \quad (1.1)$$

under the condition that the mixed product $2\psi_1\psi_2$ is *negligible*. This is indeed verified if the distance between the two wells is large enough to have ψ_1 negligible in the second well and ψ_2 negligible in the first one. When the two wells are largely separated, the ψ_s and ψ_a functions will both correspond to the same E_i energy value, giving rise to degeneration (Figure 1.2, a) and b)).

However, this framework holds until the two wells could be considered as independent; this condition is broken if the distance between them is reduced. In this case, ψ_1 and ψ_2 overlap, and the squares of ψ_s and ψ_a would be *different* and would give two different energy values (E_s and E_a), removing the degeneration (Figure 1.2, c) and d)). This behavior is analogous to that of two identical sinusoidal oscillators, where their oscillatory frequencies and energies are identical as long as they are *separated*; if they couple and interact, the two oscillatory frequencies will not hold the common value anymore, but will move towards two different frequency values, one higher and one lower than the original common one [3]. A similar behavior occurs also for two coupled harmonic oscillator potentials, as nicely discussed in Ref. [4]. Upon the consideration of this analogy, it is possible to suggest the name of "*quantum resonance*" for the approximation which describes the ψ wavefunction of the particles as the $\psi_1 \pm \psi_2$ overlap, each of the two relative to one of the two components forming the total potential of the system.

The next Sections will show that this approximate method has a huge utility in the description of molecules in chemistry and clustered structures in nuclear physics.

1.1.2 Forces between atoms and the quantum valence theory

Some approximated methods for the description of atoms and molecules will be presented in the following, serving also as a guide for the solution of more complex problems concerning the atomic nuclei. First of all, it is necessary to state how the molecules are formed: the chemical combinations available through the interplay of different atoms in the *multifaceted dance* of the chemical bonding process have way more vary and numerous characteristics than those of the atoms themselves. After the formulation of the mass and volumes combination laws (respectively, by Avogadro and Gay-Lussac) laid the basis for an experimental description of the atomic theory, three questions motivated the research to find the *rationale* for the behavior of atoms during chemical bindings and "combinations", allowing to finally build the *Chemical Valence Theory*:

- *i*) Why, and how, do the atoms actually combine to form higher-scale aggregates, such as the oxygen molecules we breathe, the water we drink, or the cellulose strands composing the pages of this thesis? As it will be shown in the following, all the properties defining the actual outcome of the chemical behavior reside in the *nature* of the valence forces themselves.
- *ii*) It is easy to see that the number of atoms composing a given molecule is always the same. A water molecule, e.g., is always composed by one oxygen and two hydrogen atoms; any modification in this specific ratio would produce completely different (and absolutely not-drinkable...) compounds. However, on the other hand, molecules can aggregate together in a virtually infinite scale, forming liquid and solid masses under the influence of cohesive forces. Would this mean that one should look for some type of *saturation* rules for the maximum number of bonds one atom can form? Would it be possible to find an analogue situation between nucleons inside an atomic nucleus?
- *iii*) What is the reason behind the fact that atoms in molecules and crystals occupy strictly relative and fixed positions in space? Is this hinting for a precisely *oriented* nature of the chemical bonds?

In the chemical realm, the responsible of microphysics bindings is essentially the electromagnetic interaction, with its many facets. While molecules merge together to form liquid and solid phases through the means of weak interactions, mostly of electrostatic nature, atoms are held together in molecules through the action of the so-called chemical bonds; these are the result of the molecular system's search for a free energy minimum, given by the outbalance of the rise of entropy and the lowering of enthalpy due to the formation of the bonds themselves. Although all the bonds are based on the electromagnetic interaction, these can be of different types, with deeply diverse micro- and macroscopical characteristics. The most common bonds are thus classified as of *ionic*, *metallic* or *covalent* nature.

Ionic bonds are typical of inorganic salts, like *NaCl* (sodium chloride, i.e., table salt), and are the simplest expression of the role played by the electrostatic force in the making

of chemical bonds; salts, in fact, are chemical compounds found in the shape of crystals, three-dimensional moieties repeating themselves in an extremely regular way throughout the macroscopic structure. The constituents of the *unit cell* of a crystal are the ions, elements which are rich or deficient of one or more electrons: in the aforementioned case, $NaCl$, the cation (Na^+ , which has lost an electron) and the anion (Cl^- , who gained the electron) are particles with opposite charge, so the resulting Coulomb force will, of course, be attractive. Thanks to the highly electrostatic nature of the bond, the electron is formally completely transferred from the sodium atom to the chlorine one, respectively giving each one full positive and negative charge and the same electronic configuration as the neighboring noble gas: since the electron is donated and not actually shared, the bond is defined as *non-covalent*, and the resulting compound is ionic.

Metallic bonds are characteristic of metals and their elemental compounds and alloys. A metallic foil of pure gold is composed by only Au atoms, repeating themselves in the space in a regular way following the compound's specific lattice parameter, i.e., the distance between each atom in the unit cell. Given the electropositive character of metallic elements, they are keener to give up their external electrons to reach a more stable configuration. Because of this, a metallic sample is thus composed by atomic centers of the respective element fixed in space, bearing a positive charge equal to the number of electrons ceded to the rest of the structure. This explains why metals usually are excellent conductors of electricity: the valence electrons are not berth on their respective atom, but de-localized and free to roam throughout the whole structure.

However, to the aims of this thesis, the most important class of bonds is that of *covalent bonds*, where the two partners effectively *share* the binding electrons. This is the usual case for binding partners with similar electron affinities, i.e., elements like C and O , lying in the same area of the periodic table and, of course, of homopolar bonds between identical elements (gases like O_2 , H_2 , Cl_2), in which there is no reason for one of the atom to be more affine to electrons than the other one. The common covalent bond is formed when two covalent electrons are available, usually put in play by each of the two elements, but different types of situations, like the dative bond (where both the electrons are provided by one of the partners) are actually possible.

A *guideline* case is usually exploited to introduce the quantum mechanical features of the chemical bond: that of the H_2^+ ionic molecule. It is constituted by two protons with a shared electron ($p + p + e$), and it will come in handy later in this Chapter to draw some analogies with the ^9Be nucleus, where a single neutron is shared between two α clusters ($\alpha + \alpha + n$).

At the basis of the description of every quantum-mechanical bond is the so-called *Born-Oppenheimer approximation*, discussed by Max Born and Robert Oppenheimer in a work published on the german journal *Annalen der Physik* in 1927 [5]. In fact, problems arise when trying to compute the solution for the Schrödinger's equation of three body systems, like the simplest ionized molecule (H_2^+), for which an exact solution cannot be obtained. The aforementioned approximation treats the two interacting nuclei, which are way heavier and slower than the binding electron, as fixed in space, and only the electron motion is considered: in this way, it is possible to solve the Schrödinger equation for the wavefunction of the electron alone, which moves in the field generated by the nuclei. This is particularly valid for the ground states of molecules, where calculations prove that the electron's speed of movement is about a thousand times higher than that of the nuclei [6]. The first quantum-mechanical theory for the chemical bond is that of

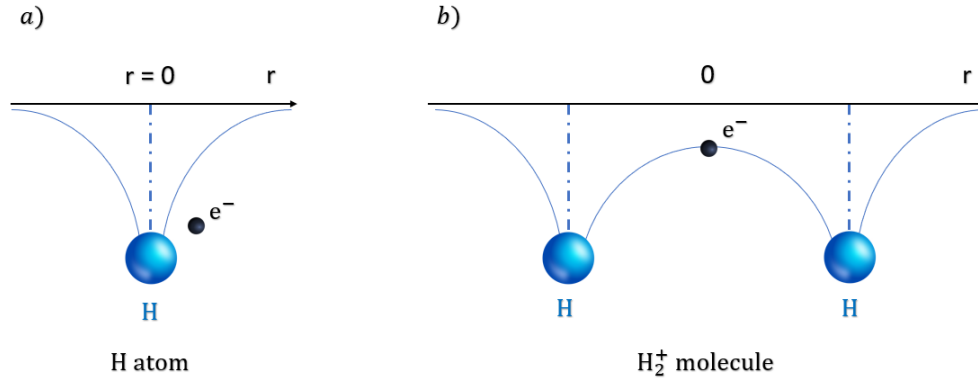


FIGURE 1.3: Schematic representation of the potential energy diagrams for a) the hydrogen atom and b) the H_2^+ ionized molecule.

the Valence Bond (VB), firstly proposed by G. N. Lewis in 1916 [7]; various features of this theory, like the concepts of orbital overlap, spin pairing, the onset of σ and π bonds, hybridization of atomic orbitals and so on, are extremely widely used in the study of a broad range of chemical compounds. All of these are also useful for the development of the Molecular Orbitals (MO) theory, presented in the following Section, for which it is easier to see resemblances and similarities with the world of nuclear physics.

1.1.3 Molecular Orbital method

The most important outcome of the Born-Oppenheimer approximation is the possibility to solve the Schrödinger equation for different values of the distance between the two interacting nuclei, r , and obtain the potential energy of the molecular system. The case of the H_2^+ ionized molecule is the simplest, not only because it only has one binding electron, but also for the fact that the bonding partners are two identical atoms, reducing the number of variables playing a role in the problem. This type of ionized molecule, which can be produced through electric discharges in H_2 gas, is interesting since it could help to understand the behavior of a single electron under the influence of two nuclei.

In a system like that of Figure 1.3, it is possible to write the Schrödinger equation of the single electron as

$$\Delta\psi + \frac{8\pi^2m_e}{h^2} \left[W + \frac{1}{4\pi\epsilon_0} \left(\frac{e^2}{r_{H_1}} + \frac{e^2}{r_{H_2}} - \frac{e^2}{r} \right) \right] \psi = 0 \quad (1.2)$$

This equation can be exactly solved and the minimum energy (W) found for the H_2^+ system at $r = 1.06 \text{ \AA}$ is of -2.64 eV (expressed with respect to the $r \rightarrow \infty$ value), meaning that the theory expressly foresees the existence of a stable H_2^+ molecule, with a positive dissociation energy. Unfortunately, this is not as easy for all the other molecules, for which it is not possible to find an exact solution of the Schrödinger equation; however, some methods like that of the Molecular Orbital may help to find approximate solutions for the energy levels of all the molecules.

In a molecule such as H_2^+ , the potential felt by the binding electron is a result of the superimposition of the potentials generated by the two nuclei, as reported in Figure 1.3; of course, it is possible to observe some peculiarities which differentiate the H_2^+

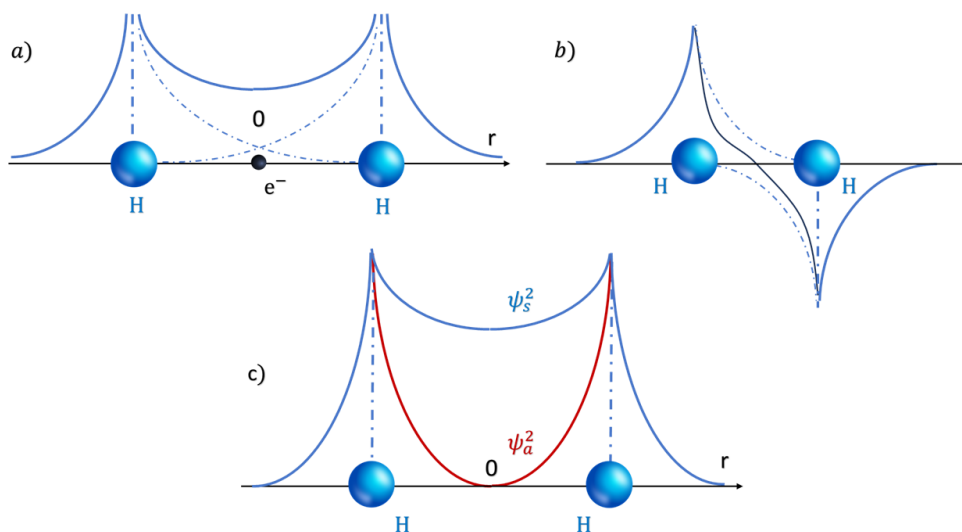


FIGURE 1.4: Possible wavefunctions for the stationary states of H_2^+ , given by a) the sum and b) the difference between the two ψ_{1s} wavefunctions.

potential with respect to that of the H atom - electron system. When the electron is found in the space between the two nuclei (Figure 1.3, b), it is subject to the attraction of both nuclei, and its potential energy is clearly lower, or higher in absolute value, with respect to that it has when part of an atom (Figure 1.3, a); this is directly related to an increase in the stability of the system. It is worth noting that the description of the system is highly dependent on the distance between the two potentials: when r is very large, the two potential wells are really distant, and each one of them is just analogue to that of a single hydrogen atom (Figure 1.3, a); on the other hand, when the distance is reduced to $r = 0$, the two-potentials system collapse to the same situation of a $+2e$ charge generated by a helium nucleus.

An approximated method to build this ionized molecule is based on the H^+ ion and the H atom, constituents of the H_2^+ molecule; starting from a large separation distance, it is possible to study the behavior of the nuclei and of the potentials while they are brought closer. In the initial framework, which one of the two is the "atom" and which is the "ion" depends from the position of the electron wavefunction ψ_{1s} ; however, for both the starting cases, when r has a finite value, the electron can tunnel back and forth through the potential barrier between the two nuclei, and cannot be formally assigned anymore to one nucleus or the other. The wavefunction of the stationary state is thus expressible through the sum or the difference between the two exponential wavefunctions representing the 1s state, as shown in Figure 1.4. But the most important feature is how the squares of these two functions give information about the binding probability: it is clear how the *symmetric* function corresponds to an increased probability of finding the electron in the central area between the two nuclei. From the comparison between this feature and what reported in Figure 1.2, it is possible to understand why the symmetric function is the one granting the formation of the chemical bond. In fact, it allows to accumulate electron charge density in the central region, necessary to the onset of the chemical bond, where the potential energy is lower than that of the corresponding separated atoms; a stable molecule is thus formed, and this type of bond is commonly referred to as a σ bond.

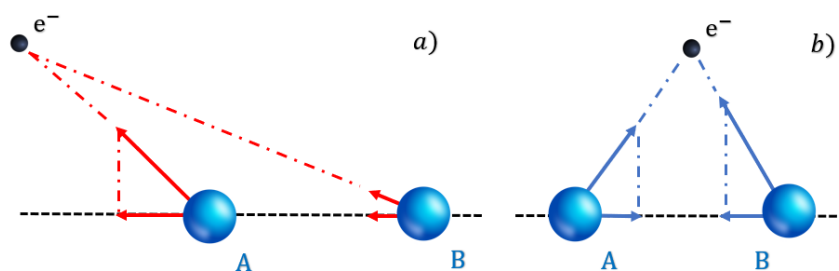


FIGURE 1.5: Resulting forces from the Coulomb interactions between one electron and the two partner nuclei: *a*) configuration yielding a separation of the atomic nuclei; *b*) onset of attractive forces, when the electron is found between the two nuclei.

When considering more complex orbitals (with respect to the *s* ones), their directional properties become crucial in the determination of the strength of the bond since, as mentioned earlier, it strongly depends from the actual superimposition of the orbitals of the bonding atoms; this would also strongly influence the geometry of the formed molecules. The type of bond formed in this case is a *covalent bond*, and its existence is based on some important requisites:

- Both partner atoms must contribute to the formation of the bond with one of their external ("valence") atomic orbitals, having similar energies.
- The electron affinity difference between the two atoms must not be high, otherwise an ionic bond would be formed. This means that a covalent bond is usually formed between atoms who sit in near spots in the periodic table.
- The conjunction direction of the partner atoms should allow the maximum superimposition between the orbitals.
- The two electrons needed for the formation of the bond can come one from each atom, or both from the same one, but they are still required to pair their spin in order to satisfy the *Pauli* exclusion principle.

As shown in Figure 1.5, it is worth noting that the force holding the atoms together in a chemical bond is still of electrostatic nature, born from the accumulation of negative charge in the inter-atomic region, due to the presence of the electrons. The *b*) side of the Figure shows how, when the electron is found between the nuclei, the components of the Coulomb force lying on the internuclear axis gives rise to *attractive* forces, which will lead to a rapprochement of the nuclei; on the other hand (*a*)), when the electron is found beyond one of the nuclei, the resulting components of the forces will push the nuclei farther.

Figure 1.6 reports the behavior of the potential energy of a hydrogen atom as a function of its distance from the fluorine atom in a hydrofluoric acid (HF) molecule; it can be noted that when the hydrogen approaches closer to the fluorine, the potential energy grows rapidly. This roughly represents the surface of the fluorine atom, and the repulsion experienced by the hydrogen atom is thus an atomic-scale analogue to the *contact* forces. But let's get some numbers into play: the horizontal line in the figure represents a hydrogen atom with a mechanical energy of -4×10^{-19} J, at one instant at rest in point C (at 1.6 Å from the center of the fluorine atom), where its kinetic energy is 0. However,

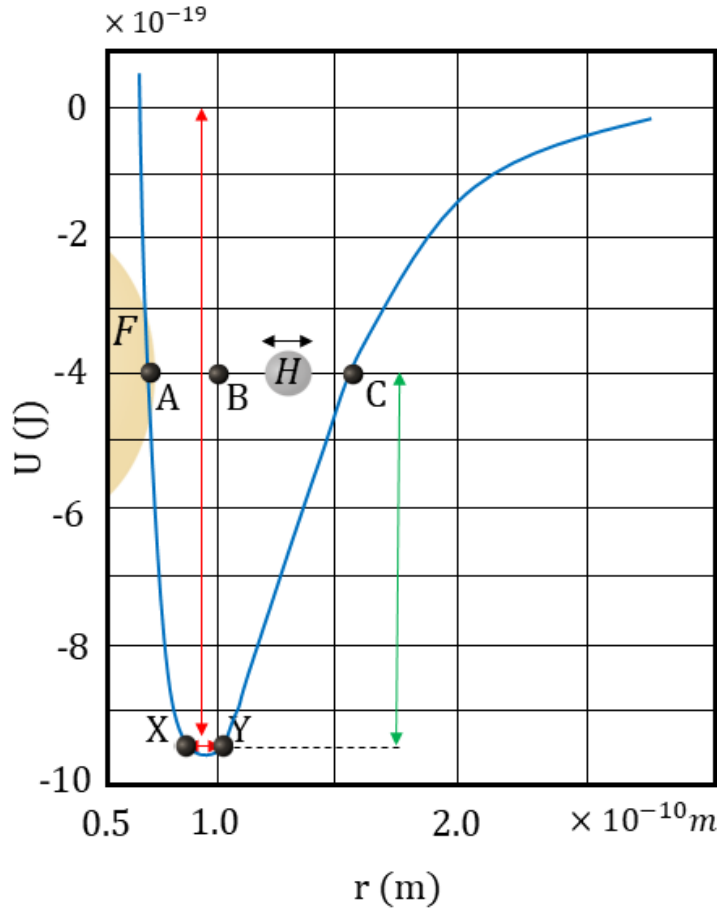


FIGURE 1.6: Potential energy curve for a bi-atomic hydrofluoric acid (HF) molecule as a function of the internuclear distance. The red vertical arrow represents the dissociation energy needed to separate the fluorine and hydrogen atom forming the molecule; the green arrow represents the energy of the hydrogen atom when it is located in *B*.

Adapted from [8].

when it is attracted from the fluorine atom, its motion begins, and in point *B* (at 1 Å from the fluorine center) its kinetic energy is 5.6×10^{-19} J. Following its approach to the F atom, the hydrogen starts to feel the contact with its most superficial states, whose repulsive force start to slow it down; it is in point *A* that the kinetic energy of the H atom becomes zero again, but just for one instant, before being pushed away by the fluorine repulsion, moving again towards *C*. The result is that the Hydrogen atom is now oscillating with a motion similar to that of a pendulum: this is the dynamical binding condition between hydrogen and fluorine in a hydrofluoric acid molecule.

All the negative energy values down to the bottom of the curve are *classically* allowed for the hydrogen atom, so that it can stay bond to the fluorine atom; however, in its quantum mechanical equilibrium configuration, the hydrogen atom is usually almost stationary, with an energy of -9.5×10^{-10} J represented between the *X* and *Y* points in Figure 1.6, where it oscillates. To dissociate a molecule which is in this state it is necessary to produce an energy high enough to allow the hydrogen atom to separate itself from the bond with the fluorine, i.e., bring it to at least $E = 0$ J. This is also the same energy which is generated when a hydrogen atom is captured by a fluorine atom to form the HF molecule [8].

This example is important to sketch the general properties of the potential energy curve for a bi-atomic molecule. In fact, a potential energy curve of the type of Figure 1.6 shows three characteristic features, typical of molecular bonding: *i*) when the inter-nuclear distance value approaches to $+\infty$, i.e., for very high distances between the nuclei, the potential energy tends to zero; *ii*) when the nuclei get too close, the interaction force becomes extremely repulsive and the potential energy value rises to $+\infty$; *iii*) the most important feature of all is the establishment of an *equilibrium bond length*, the R value corresponding to the minimum of the potential energy curve. The negative potential energy value registered at this distance, i.e., the minimum of the curve, is closely related to the *bond dissociation energy* (it differs only for the vibrational frequency of the harmonic oscillator which characterizes the bond, $-\frac{1}{2}\hbar\omega$, the so called *zero-point energy*), the amount of energy required to be transferred to the system to break the bond between the nuclei.

1.1.3.1 Correlation diagrams for the study of diatomic molecules

As mentioned, the study of the H_2^+ ionized molecule is useful to introduce the cases of more complex diatomic molecules and crystals; the two main perks of this system are the possibility to conduct an exact study of the electronic motion, and the absence of difficulties typical of systems with several electrons, which would require a more deep application of the Hartree-Fock method. The system consists of a single electron moving inside the field generated by the two protons at a fixed distance, R ; such a problem may be exactly solved through variables separation [9] in spheroidal coordinates based on elliptical and hyperbolic equipotential surfaces and ϕ , the angle formed by the electron and the internuclear axis. One of the most important properties of the wavefunction coming from the solution of the Schrödinger equation is its behavior with respect to the inversion operation about the center of the internuclear axis, demonstrable through fundamental symmetry considerations.

To discern symmetric (even) and asymmetric (odd) functions, on the basis of whether they will or will not change their sign after the inversion operation, it is possible to use the German terms "*gerade*" and "*ungerade*", respectively for *even* and *odd*. Depending on the component of the angular momentum of the wavefunction ($\ell_m = 0, 1, 2, \dots \rightarrow \sigma, \pi, \delta, \dots$) it will thus be possible to have $\sigma_g, \sigma_u, \pi_g, \pi_u, \dots$ functions. The inversion operation actually consists of a change of sign of the hyperbolic coordinate and a 180 degree phase shift of the ϕ angle: if the function is *gerade* (*even*) it will maintain the same sign, otherwise an *ungerade* (*odd*) would change its sign. These rules are important because they are valid not only for the H_2^+ molecule, but for all the homo-nuclear diatomic molecules, i.e., any molecule composed by two identical atoms (like C_2 , O_2 , $\text{N}_2, \dots$).

Figure 1.7 reports the behavior of the lowest levels of the H_2^+ molecule as a function of the distance between the two nuclei; this preliminary chart serves to introduce the matter on a first approximation, and does not include the potential energy coming from the Coulomb repulsion between the nuclei¹. The main aim, in this framework, is to underline the behavior of the different levels at very short and very large distances: in fact, for $R \rightarrow 0$, the system collapses to obtain a nucleus with two protons and one orbiting electron, i.e., an analogue of a singly ionized helium atom (He^+). An electron moving around a nucleus with charge Z , will have energetic levels given by $-Z^2/2n^2$,

¹To simplify the notations, this Section will make an intensive use of the atomic and Hartree units for distances and energy, respectively. $1 a_0 = 5.2918 \cdot 10^{-11} \text{ m}$; $1 \text{ Ha} = 4.3597 \cdot 10^{-18} \text{ J}$,

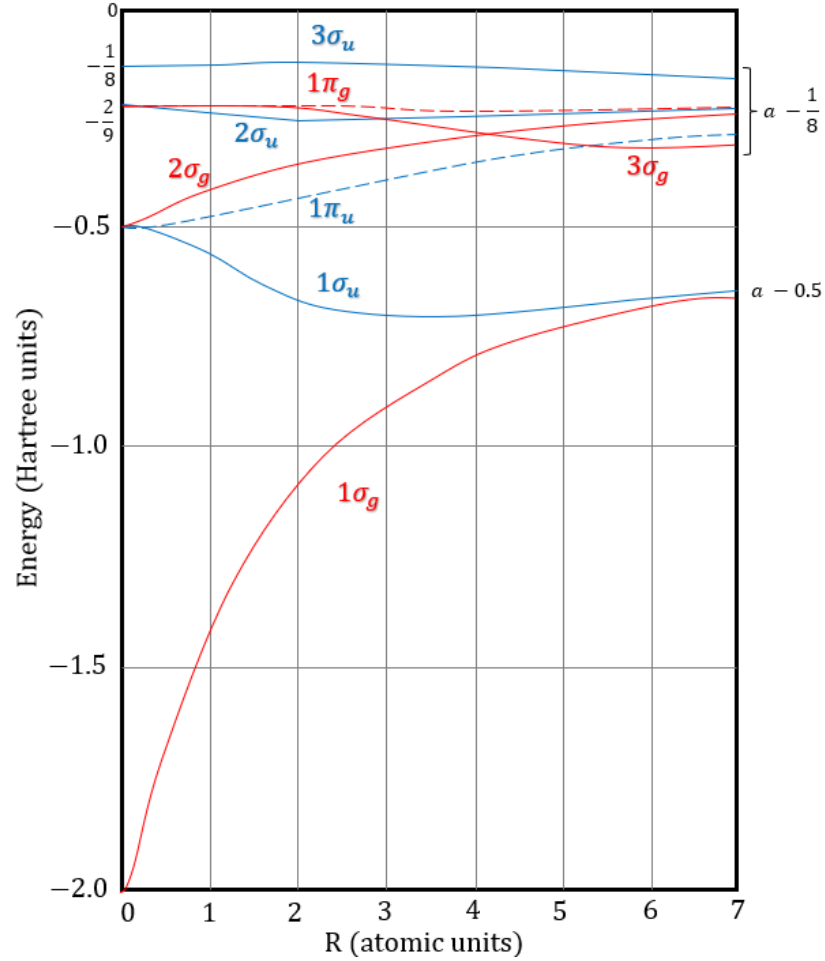


FIGURE 1.7: Lowest H_2^+ energetic levels as a function of the internuclear distance. The repulsion component due to the positive charge of the two nuclei is *not* taken into account in this chart. One Hartree (Ha), a typical unit used in molecular physics, is equal to $1 \text{ Ha} = 4.3597 \cdot 10^{-18} \text{ J}$. One atomic unit, i.e. the Bohr radius, is equal to $1 a_0 = 5.2918 \cdot 10^{-11} \text{ m}$. The a symbol on the vertical axis on the right indicates the value reached *asymptotically* by the curves.

where n is the principal quantum number: for $Z = 2$, one will have $E = -2.0 \text{ Ha}$ for $n = 1$, $E = -1/2$ for $n = 2$ and so on. At the other extreme, for large internuclear distances, it will follow that the problem is reduced to that of the Hydrogen atom, since the two protons are now separated beyond the range of the mutual interaction. In this case, the levels will converge to the energy values of $-1/2 \text{ Ha}$ and $-1/8 \text{ Ha}$, corresponding to the $n = 1, 2$ values in the separated atom and $n = 2$ value in the helium ion.

When studying the behavior of the various levels between the two limits, it is possible to notice how the $1\sigma_g$ state is the lowest for the He^+ ion, but degenerates to the value of -0.5 Ha with the $1\sigma_u$ level for the separated atoms case (the latter, tends to the $n = 2$ level of the He^+ ion for $R \rightarrow 0$). To rationalize this particular behavior, it is necessary to dive a little deeper in the study of the wavefunctions, which are shown in Figure 1.8 for separated and bound atoms. In these charts, the abscissa is related to the z coordinate, i.e., the distance between the two nuclei (where the origin point, 0, is the middle point of the internuclear axis).

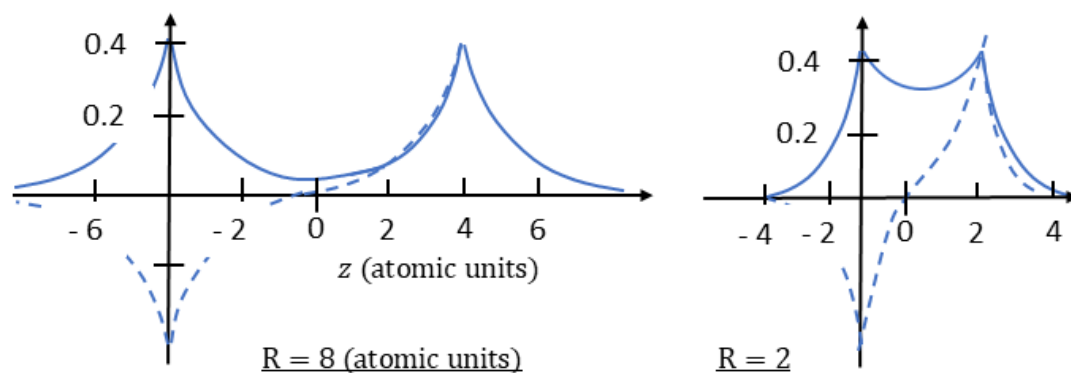


FIGURE 1.8: Normalized wavefunctions for the $1\sigma_g$ (solid line) and $1\sigma_u$ (dashed line) functions of the H_2^+ system for separated atoms case ($R = 8a_0$) and in the bond equilibrium condition ($R = 2a_0$).

The first of the two graphs ($R = 8a_0$) is referring to highly distanced nuclei, i.e., an unbound system of two protons, which will show a peak for the electron wavefunction in correspondence of the position of each one of them. These are of course related to the $1s$ atomic orbital wavefunctions of the single hydrogen atoms, which have simple $\exp(-r_a)$ and $\exp(-r_b)$ forms; the difference between the behavior of the $1\sigma_g$ and $1\sigma_u$ functions is that they are respectively added or subtracted to each other. The result of the combination of two or more atomic orbitals is a so-called *molecular orbital* (MO), as the ones reported in Figure 1.8; in this approximation, defined as *Linear Combination of Atomic Orbitals* (LCAO), the MOs of a molecule are considered as *linear combinations* of the single atoms' atomic orbitals.

However, this approximation holds better for high distance values between the two atoms, and loses reliability for lower values of R . Nonetheless, this method is still sufficient for intermediate distances, such as the $R = 2a_0$, reported in the right-hand part of Figure 1.8, and defined as the equilibrium distance for the onset of the chemical bond. Of course, the LCAO approximation breaks for $R \rightarrow 0$, where the MO degenerates to a single peak corresponding to the He^+ ion $1s$ peak, an $\exp(-2r)$ function well different from the $2 \cdot \exp(-r)$ one attainable from the LCAO method.

Figure 1.9 now represents the same H_2^+ lowest energetic levels as Figure 1.7, but with the inclusion of the potential deriving from the Coulomb repulsion at small distances. From this graphic is clear how the curve for the $1\sigma_g$ is characterized by an evident minimum, differently from what seen when neglecting the repulsive term of the potential energy; the minimum of the curve is found at a distance of $R \approx 2a_0$, with an energy of $-0.60 Ha$. This thus corresponds to the equilibrium distance, at which the chemical bond is developed; following the value of the curve for $R \rightarrow \infty$, it is possible to determine a *dissociation energy* (i.e., the work needed to bring the two atoms to infinite distance) of $0.10 Ha$. On the other hand, the $1\sigma_u$ is a repulsive level, since the potential energy of the system monotonically decreases with the increment of the internuclear distance: this means that, for any R value, no work is needed to separate the atoms, which naturally tend to move away.

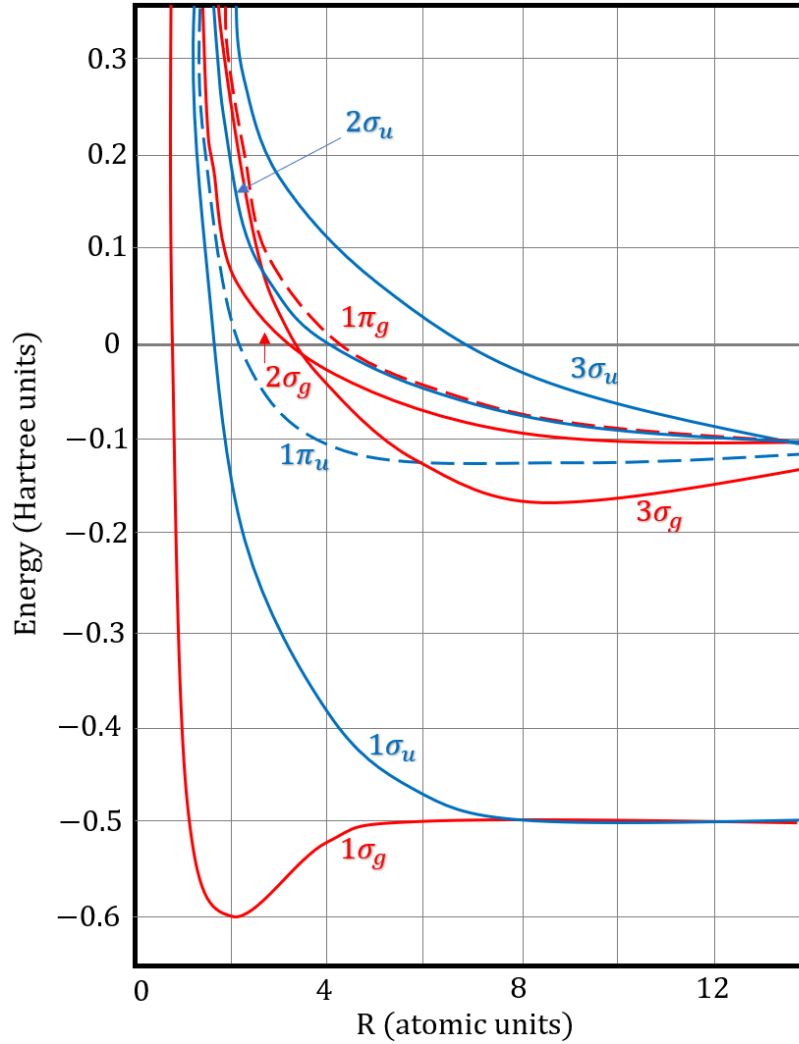


FIGURE 1.9: Representation of the lowest energetic levels for the H_2^+ ionized molecule as a function of the internuclear distance, with the inclusion of the potential energy imputable to the nuclear repulsion. One Hartree (Ha) is equal to $1 \text{ Ha} = 4.3597 \cdot 10^{-18} \text{ J}$. One atomic unit is equal to $1 a_0 = 5.2918 \cdot 10^{-11} \text{ m}$.

1.1.4 The covalent bond method (Heitler-London)

The most common type of chemical bond is, actually, the covalent bond, where two electrons are put in play to form it and to tie the partner atoms together. This means that a further theoretical effort is needed, moving from the simplest H_2^+ ionized molecule to the smallest complete molecular system: the hydrogen (H_2) molecule.

In this case, the Schrödinger equation describing the motion of the electrons in the H_2 molecule is similar to that of atoms with two electrons; on the other hand, the potential energy has a more complicated form, given that it already comprehends, between attractions and repulsion, six Coulomb terms. Although this system is usually solved with the Molecular Orbital method, an easier pathway is given by the *Heitler and London* Valence Bond method [10]. The first step of this approximation would consider the two H atoms as separated, each one with one of the electrons forming the pair (nucleus A with electron 1 and nucleus B with electron 2, both in their ground state with energy

E_0). The potential energies of the two atoms, built on the interaction with the electron at a given distance r , will be respectively defined as ke^2/r_{A1} and ke^2/r_{B2} and the wavefunction of the system given by:

$$\psi = \psi_{A1} \cdot \psi_{B2} \quad (1.3)$$

However, when the two nuclei are separated by a finite distance, the mutual repulsions between the two nuclei and between the two electrons must be taken into account, along with the attraction forces acting between one nucleus and the respective electron. The potential energy will thus be expressed as:

$$U = -\frac{e^2}{4\pi\epsilon_0} \left(\frac{1}{r_{A1}} + \frac{1}{r_{B2}} + \frac{1}{r_{A2}} + \frac{1}{r_{B1}} - \frac{1}{r} - \frac{1}{r_{12}} \right) \quad (1.4)$$

where the last four terms express the perturbation energy ΔE of the initial system: the attraction between the A nucleus and the second electron ($1/r_{A2}$), and the B nucleus and the first electron ($1/r_{B1}$), and the repulsions between the two nuclei ($1/r$) and the two electrons ($1/r_{12}$). Given that the two electrons are obviously not differentiated, the system will exhibit an exchange degeneracy; the wavefunctions of the unperturbed system will thus have the form $\psi_{\pm} = \psi_{A1} \cdot \psi_{B2} \pm \psi_{A2} \cdot \psi_{B1}$, and the perturbation energy is expressed through the following formula:

$$\bar{E}_{\pm} = \frac{\int \Delta E \psi \psi^* d\tau}{\int \psi \psi^* d\tau} \quad (1.5)$$

where the integration is extended to the electrons field. When using one of the expressions of $\psi_{\pm}$ in place of the general ψ and ψ^* , it is possible to proceed with the computation of the integral and develop an expanded expression for the numerator of Equation 1.5

$$\int \Delta E \cdot \psi_{A1}^2 \psi_{B2}^2 d\tau_1 d\tau_2 + \int \Delta E \cdot \psi_{A2}^2 \psi_{B1}^2 d\tau_1 d\tau_2 \pm 2 \int \Delta E \psi_{A1} \psi_{B2} \psi_{A2} \psi_{B1} d\tau_1 d\tau_2 \quad (1.6)$$

Upon the exchange of the two electrons, the physical meaning of the problem will not be altered, and the first two integrals are thus equal; these can be indicated with a common value " C ", while the last integral has a value which will be indicated with " J ". All of Equation 1.6 can thus be reduced to $2C \pm 2J$. Regarding the denominator, it is easily expanded as:

$$\int \psi_{A1}^2 d\tau_1 \psi_{B2}^2 d\tau_2 + \int \psi_{A2}^2 d\tau_1 \psi_{B1}^2 d\tau_2 \pm 2 \int \psi_{A1} \psi_{B2} \psi_{A2} \psi_{B1} d\tau_1 d\tau_2 \quad (1.7)$$

Following the postulates of quantum mechanics, if the hydrogen atoms wavefunctions are normalized, the first two terms are clearly equal to 1. The last term is however not null, differently from the He atom case, since the ψ_A and ψ_B functions are related to two different atoms and, thus, not orthogonal. If the value of the last integral is indicated as " S " (with values limited between 0, when the atoms are well separated, and 1, when $R = 0$ and $\psi_A = \psi_B$), Equation 1.5 could be re-written as:

$$E_{\pm} = \frac{C \pm J}{1 \pm S} \quad (1.8)$$

This is indicating that in the H_2 molecule, similarly to the two electrons in a He atom, two different values of energy are expected, different from the sum of the energies of the

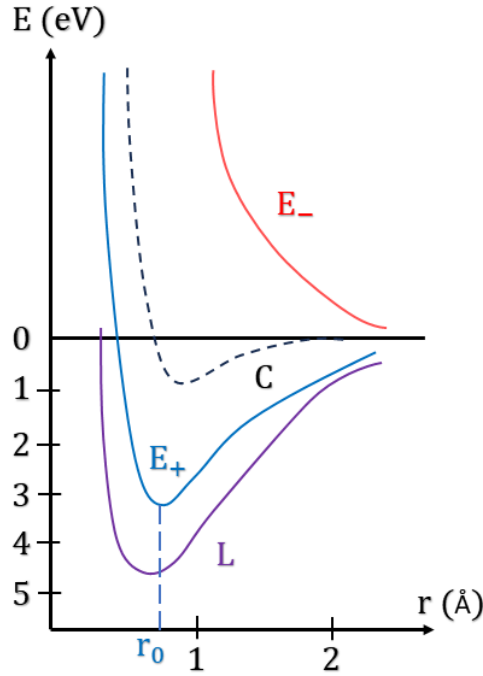


FIGURE 1.10: Evaluation of the binding energy in the H_2 molecule, with the separation of the different components of the wavefunction: (C) the value of the integrals at the numerator as a function of the distance r , related to the Coulomb interaction between the atoms; (E_+/E_-), whose sign is determined by the J term at the numerator: if J is positive, the E_- repulsive curve is obtained, while, for a negative J , it is possible to draw the E_+ attractive curve, showing a marked minimum. (L) values of the binding energy curve calculated through the *Morse formula*.

separated atoms (E_0). These are $E = 2E_0 + E_+$ and $E = 2E_0 + E_-$. However, the S term at the denominator is usually small, and thus negligible in the framework of a qualitative discussion of these formulas.

Figure 1.10 reports the behavior of the different components forming the binding energy curve in the H_2 molecule. It is evident that the responsible for the onset of the chemical bond is the E_+ curve, derived from the negative sign of the J term, the integral related to the electron exchange. In fact, this curve shows a pronounced minimum ($E = 3.2$ eV) in correspondence of $r_0 = 0.8$ Å. The experimental value for the equilibrium distance of H atoms in the H_2 molecule, derived from Raman spectroscopy analyses, is of 0.74 Å, in excellent agreement with the calculation, while the position of the energy minimum is smaller than the experimental value ($\simeq 4.5$ eV). The L curve in Figure 1.10 represents the potential energy curve obtained from the phenomenological *Morse formula* [10] that, avoiding the explicit (and complicated) calculation of the J term, can approximate in a very good way the binding energy values (4.7 eV for the H_2 case) and equilibrium distances of an homonuclear molecule. In any case, the most important feature observed in this Section is the fact that the formation of a stable bond in the H_2 molecule can only be explained through a specifically quantum-mechanical notion, i.e., the electronic exchange.

The fact that the chemical bond can only be formed as the result of an electronic exchange and of an enhancement of electronic charge density in the internuclear region

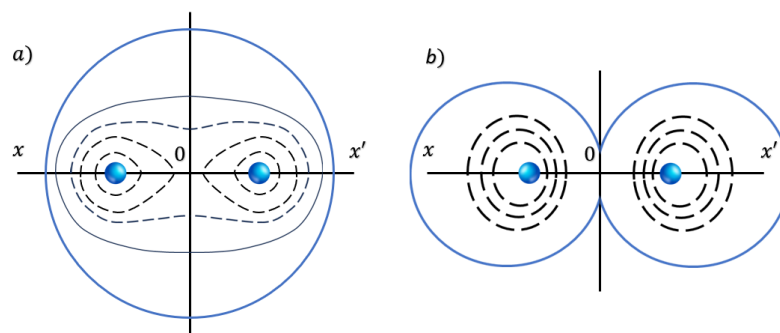


FIGURE 1.11: Equal distribution curves for the electronic density in the H_2 molecules around the two hydrogen atoms, in the a) bonding orbital and b) anti-bonding orbital.

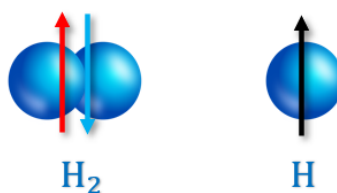


FIGURE 1.12: Schematic diagram explaining the saturation of the H_2 molecule.

is made clear from the diagrams in Figure 1.11. These curves are built by considering the probabilities of presence of the two electrons times the electron charge e , and are symmetrical with respect to the middle point of the internuclear axis. In this case, the a) diagram is related to the ψ_+ wavefunction, while b) is referring to the distribution of the ψ_- one. It is possible to see how for ψ_+ the negative charge density is accumulated between the two protons, while it is pushed in the external area for the ψ_- case, showing how this is a *repulsive* state. Therefore, for the H_2 molecule, the ψ_+ function will be indicative of a bonding orbital, while the ψ_- is representative of an anti-bonding orbital. Considering the Pauli principle and both the spacial and spin parts of the wavefunctions, the spins of the two electrons need to be *paired* to form a chemical bond. On the other hand, the anti-bonding configuration is linked to a parallel alignment of the electronic spins.

All the mechanics shown in the previous paragraphs are also accounting for the *saturation* of the valences of the hydrogen atoms in the H_2 molecule. As sketched in Figure 1.12, when a H atom approaches a H_2 molecule, their interaction is ruled by the relative orientations of the spins. Since only two values ($\pm\frac{1}{2}$) are allowed for the electronic spin, the spin of the electron of the roaming H atom will always be parallel with one of those forming the bond in H_2 , generating a repulsive interaction. On the other hand, the *exchange* between the electrons with the anti-parallel spin would lead to parallel spin in the H_2 molecule, being forbidden by the *Pauli* exclusion principle. This justifies the saturation and subsequent non-reactivity of the H_2 molecule and of noble gases.

1.1.4.1 Generalization to the spin-covalence method

Using the simplest H_2 molecule as a starting point, it is possible to draw a general rule stating that two atoms can bond when each one of them put a single unpaired

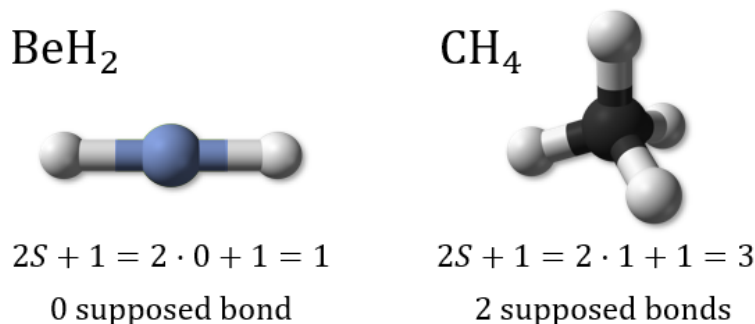


FIGURE 1.13: Number of supposed bonds for the beryllium (left) and carbon atoms (right), along with the deriving apparent exceptions to the covalence rule.

electron into play; when the two electrons pair their spins in the same molecular orbital, a *covalent bond* is formed. As a very straightforward rule of thumb, it is possible to say that the *covalence* of an atom, i.e., the number of atomic bonds it can form, is directly related to its number of unpaired electrons. By definition, an atom is found in its energetic ground states when all the spins of its *unpaired* electrons are parallel. Given that the *Spin multiplicity* is defined as $2S + 1$, where S is linked to the sum of all the electronic spins ($|\frac{1}{2}|$ each), an atom with 1 unpaired electron would have a spin multiplicity of $2S + 1 = 2 \cdot \frac{1}{2} + 1 = 2$; the covalence of the atom, i.e., the number of bonds it can form, can be easily computed as this number *minus* 1. Straight from this rule, it is clear how atoms where all the electrons are paired in spins would, in principle, not form any bond ($2 \cdot 0 + 1 = 1$), since their covalence would be 0: this is the case of all the so-called "*noble*" gases.

Figure 1.13 shows the first two apparent exceptions to the rule relating the number of unpaired electrons with the allowable number of bonds. Be, as an element, only has four electrons, while C has six; given that the ns orbitals can contain up to two electrons, and the np ones up to six, the Be atom shouldn't be able to form any chemical bond, while the carbon would be limited to 2. However, evidences of the existence of molecules such as BeH_2 and CH_4 deny these suppositions. To explain the formation of these molecules in a simple way, it is important to understand that the atoms could be excited to levels with higher energy than the ground state, e.g., through the promotion of their electrons to the next atomic orbital: in the Be case, which has a $1s^2 2s^2$ electronic configuration, one of the two electrons in the $2s$ subshell can be promoted to the $2p$ level with an energy of just 3 eV, easily attainable through the formation of the bond itself. In this case, the electronic configuration of the excited Be atom would be indicated as $1s^2 2s^1 2p^1$. As reported in the previous paragraph, this promotion would generate a triplet state, because the Be atom would now have *two* unpaired electrons, with parallel spins, ready to form two chemical bonds. For the carbon atom, the promotion of one electron from the $2s$ to the $2p$ level ($1s^2 2s^2 2p^2 \rightarrow 1s^2 2s^1 2p^3$) generates a quintuplet state with 4 unpaired electrons and, thus, makes it possible to form molecules with 4 chemical bonds. Another step, defined as *hybridization* of the s - p orbitals, allows the carbon atoms to form four identical bonds through hybrid sp orbitals, instead of forming three bonds with the $2p$ orbitals and one with the $2s$, which would result weaker and not spatially oriented. This mechanism also explains why the Boron atom can form 3 bonds (and not only one), and why atoms like Phosphorus and Sulfur have different valences with a two-units difference (respectively: 3, 5 and 2, 4, 6).

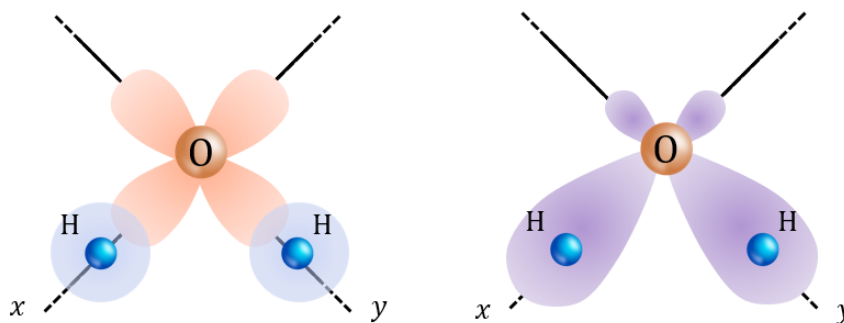


FIGURE 1.14: Schematic representation of the atomic and molecular orbitals in a water molecule.

1.1.4.2 The spatial character of the atomic bond: the oriented valence

Since the onset of a chemical bond is related to an accumulation of negative charge between the two nuclei, the formation of the bond will be favored proportionally to the magnitude of the overlap of the two electronic distributions of the partner atoms. To understand how the different spatial properties of the atomic orbitals may influence the characteristics of the chemical bonds in a molecule, it is enough to study the behavior of the elements of the first two periods of the Periodic Table; these atoms will only use their $1s$, $2s$ and $2p$ atomic orbitals to form chemical bonds. While the ns orbitals have a completely isotropic symmetry, the np orbitals keep the same dependence with respect to the distance from the center of the nucleus, but have completely different spatial properties.

Let's make an example based on the *water* molecule. The oxygen atom has six valence electrons, with a $2s^2 2p_z^2 2p_x^1 2p_y^1$ external configuration. Since the unpaired electrons are sitting in two $2p$ orbitals, each one of these can grant the maximum overlap with a $1s$ orbital of a H atom, given that the two hydrogens are lined up with the directions of the two p orbitals (x and y axes, Figure 1.14, left). At the same moment, if another H atom approaches along the z -axis (perpendicular to the page), it would experience the same repulsion described earlier, as an effect of the *Pauli* principle. The overlaps between the $2p$ orbitals of O and the $1s$ of the two H atoms give birth to the Molecular Orbitals pictorially shown in the right-hand scheme of Figure 1.14. Of course, this simple representation could lead to the idea that the HOH angle in the water molecule is of 90° ; this is actually not true, since the experimentally observed angle is of $\approx 104.5^\circ$. This can be understood if the water molecule is seen through a three-dimensional perspective, also including the electron pair in the $2p_z$ orbital. In a simple framework, following the so-called VSEPR (*valence shell electron pair repulsion*) theory [11], a non-bonding pair of electrons actually demands more space than that occupied by two electrons involved in a chemical bond. When a molecule is made up by three atoms (one central atom with two bonds), and no lone electron pairs are present, it will assume a linear configuration, as for the BeH_2 molecule in Figure 1.13, following the natural repulsion of the electrons forming the bonds; however, the presence of the additional electron pair on the oxygen atom results in a compression of the angle formed by the hydrogens, pushing it towards the 104.5° value.

This also accounts for the existence of an electric dipole momentum in the water molecule, and it would be really difficult to find other properties with such paramount

importance: the simple presence of a finite dipole momentum in the structure of the water molecule directly accounts for the existence of life itself, at least as it is known. The water molecule is thus a clear example of the oriented bond theory, and how the spatial distribution of the bonds and the electrons in a molecule, i.e., its *Stereochemistry*, are extremely important to describe everyday and fundamental phenomena. If the water molecule would not have a finite electric dipole momentum, it would be impossible for it to exist in the liquid phase at room or even colder temperatures; it would appear instead more similar to the lightest organic compounds, like methane or ethane (CH_4 and C_2H_6), which only become liquids at temperature lower than $\approx -166^\circ\text{C}$ and $\approx -89^\circ\text{C}$. At room temperature, in fact, these compounds are gases, since the absence of a net dipole momentum in the overall symmetry of the C-H bonds does not allow to establish intermolecular interactions strong enough to hold different molecules together in a condensed phase. The properties of the intermolecular interactions and their affinities to similar nuclear physics phenomena will be discussed in higher details in the next Section.

The spatial properties of the atomic and molecular orbitals are also important to describe the different type of covalent bonds that could be found in a molecular system. In fact, two partner atoms can bond with more than a single link (σ bond), forming the so-called double or triple bonds, where the π molecular orbitals make their appearance. To understand how the different σ and π orbitals are formed, it is necessary to retrieve the concept of orbital hybridization presented in the previous subsection. It has been mentioned that the hybridization of the $2s$ orbitals with all the three $2p_{x,y,z}$ orbitals forms four identical orbitals, with mixed $2p$ and $2s$ components, called sp^3 hybrid orbitals: the denomination comes from the mixing ratio of one s (s) to three p (p^3) orbitals. As a result, on the basis of the prescription of the VSEPR theory and the experimental evidences, the bonds formed through these orbitals will be distributed along a tetrahedral structure, like in diamond and saturated organic compounds [11, 12].

However, the tetrahedron is not the only structure that can be formed through the hybridization process; when it is done for one $2s$ orbital with only *two* of the $2p$ orbitals (e.g., $2p_x$ and $2p_y$), the resulting system is composed by three identical sp^2 orbitals, lying on the same plane with a 120° angle, and the fourth non-hybridized orbital ($2p_z$) perpendicular to their plane. This hybrid structure, for the carbon atoms, is fundamental to explain the structure and the properties of graphite, which is composed by thin layers with hexagonal patterns of sp^2 carbon atoms. Here, the atoms inside the same plane are held together with strong forces, typical of the chemical bonds, while the $2p_z$ orbitals weakly keep the different planes together through intermolecular forces, dependent from the surface of contact and the electrostatic properties of the atoms. This accounts for the different properties of graphite, from its flakiness, which allows pencils to write, to its ability to be used as a dry lubricant in high-tech environments [13].

The planar structure given by the sp^2 hybridization is also typical of different classes of organic compounds, the so-called "unsaturated" structures, like alkenes and aromatic compounds. An useful sketch of the ethylene molecule (C_2H_4) is given in Figure 1.15, and allows to understand the mechanism of the *double chemical bond*, which allows to maintain the four-fold valence for the carbon atoms in unsaturated molecules. The bond scheme here reported gives an explanation for the fact that the C–C bond is shorter, and thus stronger, in the ethylene molecule with respect to the saturated ethane molecule (respectively, 1.30 Å and 1.44 Å, and 3.6 and ≈ 6 eV [12]).

As already shown, the bonds resulting from the frontal ("direct") overlap of two orbitals of any type are called σ bonds. In the ethylene molecule of Figure 1.15, C_2H_4 , five

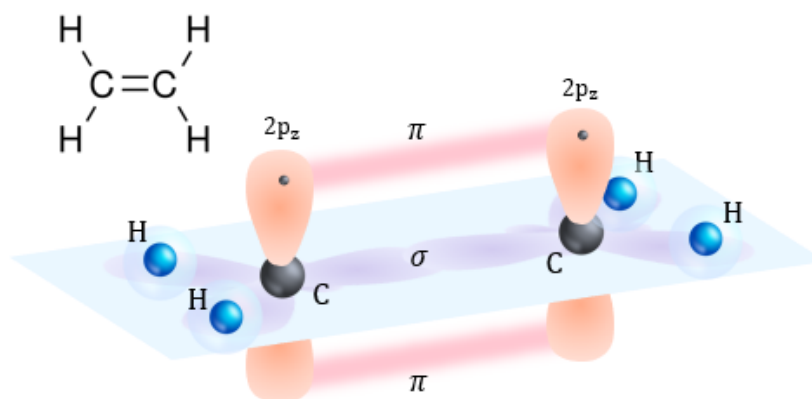


FIGURE 1.15: Schematic representation of the different bonds taking place in a C_2H_4 molecule: four σ bonds between the sp^2 hybrid orbitals of the C atoms and the $1s$ of the hydrogens, one σ between the remaining sp^2 orbitals of the C atoms, and one π bond born from the lateral overlap of the non-hybridized $2p_z$ orbitals, perpendicular to the plane described by the other bonds.

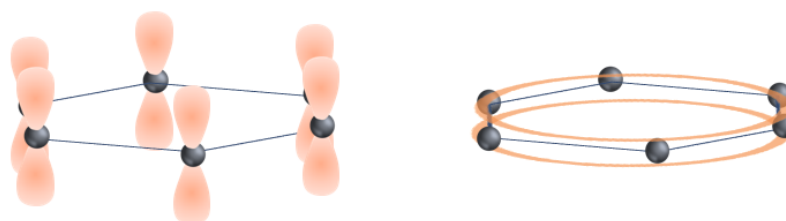


FIGURE 1.16: *Left*: the six $2p_z$ orbitals of each of the carbon atoms in the benzene (C_6H_6) molecule; *Right*: onset of the resonant molecular π orbital due to the lateral overlap of all the six $2p_z$ spare orbitals in the benzene ring.

different σ bonds are present: four between the C and H atoms, and one in the middle, formed by the two carbons. On the other hand, given the geometry of the system, the overlap of the two $2p_z$ orbitals could never happen along their natural direction: in this case, a *lateral* overlap is said to happen, and this type of bonds are called π bonds. A double bond is thus composed by one σ bond and one π bond acting between the same couple of partner atoms; this can of course be generalized to the triple bonds, where the orbital hybridization is of the sp type, two $2p$ orbitals remain orthogonal to the C–C axis, and two π bonds are thus formed in addition to the σ one.

One of the most important properties deriving from the formation of the π bonds, however, is the possibility to have a delocalization of the valence electrons, like in the case of the *benzene* molecule. Here, as reported in Figure 1.16, the six orthogonal $2p_z$ orbitals form a de-localized electronic cloud above the plane of the hexagonal ring, making this structure one of the most peculiar ones in the whole organic chemistry landscape. This derives from the fact that the six electrons coming from the various $2p_z$ orbitals can only form three double bonds, but it is impossible to determine the three couple of atoms that would effectively be doubly linked (1–2, 3–4 and 5–6 or 2–3, 4–5 and 6–1); the result is the adoption of a mixed structure, *resonating* instantaneously between the two possibilities, more precisely represented through the aforementioned electronic cloud distribution, also reported in the right-hand side of Figure 1.16. This

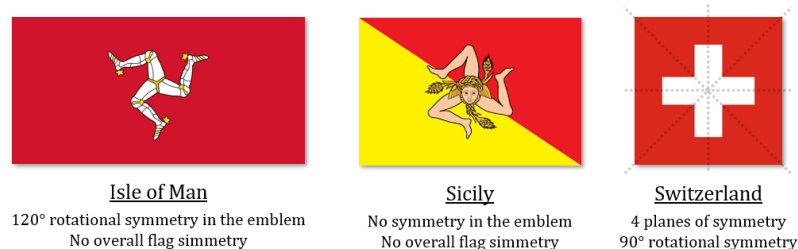


FIGURE 1.17: Elements of symmetry of different flags and emblems. The *triskeles* emblem in the center of the flag of Sicily would of course benefit of the same three-fold symmetry as that of the Isle of Man, if it were not for the presence of the *Gorgoneion*, the head in the center of the emblem. Last, a depiction of the symmetry planes in the most symmetric flag, the Swiss one.

is also crucial to explain different physical and chemical properties of all the different classes of aromatic compounds [14].

1.1.5 Symmetry in molecules and in their excited states

One of the main ways to rationalize some peculiar properties and characteristics of a given compound is the study of its structure: the disposition of the different elements, their ratios, the distance between the planes of a crystal. However, it is certainly not easy to draw conclusions from these types of analysis if there is no information to start with; to compare the experimental results (i.e., the x-ray or electron diffraction) with the theoretical calculations, it is necessary to have some basic hypotheses for the atomic distribution and the molecular symmetry. This is fundamental for the study of regular crystalline structures, both in molecules and, as it will be shown in Section 1.2.3.8, for the so-called self-conjugate atomic nuclei.

The Group Theory refers the study of molecular and crystalline structures to that of finite geometric figures, exploiting their multiple symmetry elements to reduce the number of variables and simplify the problem. The main assumption at the basis of this theory is the fact that the atoms are rightfully considered to have a spherical shape (the local differences in the electronic distributions are negligible for the comparison of two atoms of the same element); then, it is necessary to look for all types of *operations*, like rotations and reflections, or the inversion, which will produce a final state which is absolutely identical to the initial one. Each *operation* is then related to a specific *symmetry element*, like a *point*, a *line*, or a *plane*. For example, if one was looking at a square that was rotated by 90° when they were looking away, it would be impossible to state if the square was effectively rotated or not: this is a simple explanation of a symmetry operation. Obviously, this is not limited to chemistry, but it is a mathematical theory whose applications ranges from vexillography, art and concept design to anatomy itself (although more on the external than on the internal level). Figure 1.17 reports the different levels of symmetry and the respective elements that can be found in some flags, and how the overall symmetry of the system depends from the behavior of all its elements: for example, although the triskelion emblem on the Isle of Man flag is identical when rotated alone by 120°, this property is not extended to the whole flag, which has a rectangular shape, clearly not suitable with a 120° rotational symmetry. Nevertheless, this is not the case of the most symmetrical flag, the Swiss one: thanks to the square

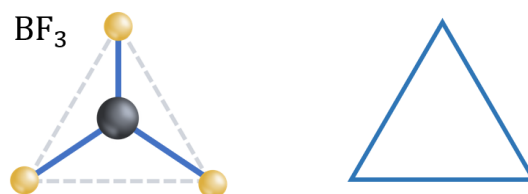


FIGURE 1.18: The structure of the Boron trifluoride (BF_3) molecule in comparison with an equilateral triangle.

shape of the vexillum, which follows the characteristics of the central white cross, the whole flag can benefit of four planes of symmetry and of the adaptability to the 90° rotation.

Several of these symmetry operations, if performed together leaving at least one common point unmodified, e.g., in a molecular structure, take the name of a *Point Group*; in the more extended framework of the description of crystals, it is also necessary to study the translations through space, and thus to treat more complex *Space groups*. From a chemical point of view, a molecule is defined from its composition, its point group and the position of its atoms on the symmetry elements. The study of the molecular symmetry is extremely important to understand the actual atomic orbitals which may get into play in the formation of a molecular orbital and those which will not, yielding several chances to simplify the problem of the Linear Combination of Atomic Orbitals and all the derived overlap calculations, necessary to study the formation of the Molecular Orbitals.

The only operation which is common to every molecular system is the *identity* (E) operation, equivalent to doing literally *nothing* to the object; this is included in the theory for technical reasons, and to grant that every system could have at least one symmetry element and the correspondent operation. However, among the most common operations, which can also serve as a bridge to draw analogies between the chemical and the nuclear physics world, it is possible to find the *rotation* and the *reflection*.

A **rotation** about an n -fold symmetry axis (C_n) is defined as a rotation of $360^\circ/n$ degrees; while the C_1 operation (a 360° rotation) is obviously equivalent to the E operation, the following C_2 , C_3 , C_4 ... operations are referring to 180° , 120° , 90° ... rotations, and so on. Although more complex combinations of symmetry operations may exist, also considering the distinction between clockwise and counter-clockwise rotations, it is better to limit their number to those useful to the purpose of this thesis. For example, an ammonia (NH_3) molecule has a three-fold C_3 symmetry axis, while a water molecule, H_2O , only benefits from the C_2 rotations; when a single system possesses more than one rotation axes, the highest-index one (n) will be called the **principal axis**. The benzene molecule in Figure 1.16, among several other symmetry operations, also has six C_2 axes; however, the principal one is the C_6 axis (corresponding to a 60° rotation), passing through the geometrical center of the hexagon.

There are then two different types of **reflections** about a *mirror plane*, and are indicated with the symbol σ . The mirror plane defining the reflection operation may contain the principal axis of the molecule, or it may be perpendicular to it; the corresponding reflections are thus respectively denoted as σ_v (vertical) and σ_h (horizontal). For example, a H_2O molecule has two *vertical* planes of symmetry, a NH_3 molecule three, while a benzene ring has a *horizontal* mirror plane, in addition to the other elements of

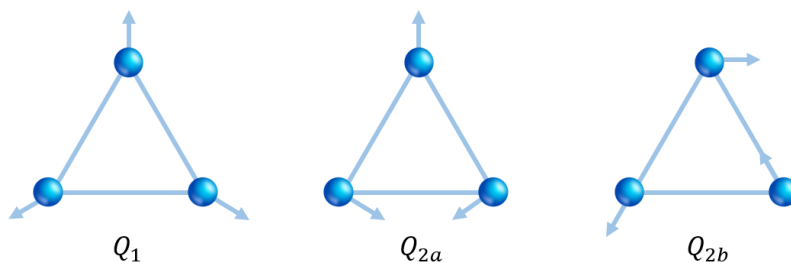


FIGURE 1.19: The structure and the vibrational modes of the H_3^+ molecular ion.

symmetry. Other notable symmetry operations, which will not be discussed in details here, are the **inversion**, the **reflections** about a *dihedral plane* (σ_d) and the ***n*-fold improper rotations**, composed of an *n*-fold rotation and a reflection about a mirror plane perpendicular to the axis of that rotation; the most important property of this composed operation is that the rotation and the reflection alone do not need to be symmetry operations of the molecular system, but the overall *improper rotation* itself will be.

The definition and classification of all the symmetry elements and the respective operation allows to group together molecules which possess the same symmetry elements; by doing so, methane (CH_4) and carbon tetrachloride (CCl_4), both having the same symmetry elements of a regular tetrahedron, are classified in the same group, while the H_2O molecule is put in another one. The common *Schoenflies* nomenclature system will be used to identify point groups in this work; however, since an extensive treatment of all the point groups and their classification [15] is far beyond the purpose of this thesis, any specific notion which may be needed during the treatment of symmetries in clustered atomic nuclei will be recalled on the spot.

One of the most important things to take into account is that the experimental observations can not be made on molecules alone, but on a multitude of them; it is thus important to infer the individual properties of the molecules from the study of the ensemble, and this is only possible when studying a structure which is regular in space and time, differently from the cases of gas and solutions. Nevertheless, it is crucial to identify the techniques which could help to determine the molecular symmetries in the best way. For example, polarized molecules have a permanent electric dipole momentum, represented with a vector; by definition, this can not exist if the molecule has a symmetry center and/or an axis with a symmetry plane perpendicular to it. Similarly, magnetic properties are not actually useful for the determination of molecular symmetry, since paramagnetism is a property of single atoms and diamagnetism is just a very general property of the molecule. One of the most common technique adopted for the study of the molecular symmetry is the vibrational spectroscopy: in this framework, a molecule can absorb the received infrared radiation if it has the same frequency of one of its vibrational modes, which are completely dependent from the molecular symmetry and structure. In this way, it is possible to obtain information on the presence of some particular types of bonds, and on their strength and orientation [15].

An useful example to exploit the role of symmetries in molecules is linked to the spectroscopic analysis of the H_3^+ molecule reported in Ref. [16]. This molecular ion, until that moment unknown in conventional chemistry due to its high protonation reactivity, was discovered by Thomson in 1911. Today, it is studied as one important constituent

in hydrogen plasmas, and as a major actor in several reactions in the field of interstellar chemistry [17, 18].

As usual, this five-body system (three nuclei and two electrons) is treated in a framework derived from the Born-Oppenheimer approximation, where the eigen-values coming from the Schrödinger equation are related to the motion of the heavier nuclei, while the perturbation Hamiltonian contains the derivatives of the electronic wavefunctions with respect to the nuclei themselves. As shown by several theoretical calculations [19, 20], this molecular ion can be described with an equilateral D_{3h} structure, in analogy with the description of the ^{12}C nucleus ground and Hoyle states in the framework of the Algebraic Cluster Model (ACM) (see Section 1.2.3.8).

Figure 1.19 depicts the first two vibrational modes of the H_3^+ system, taking into account the degeneration of the Q_2 mode; here, the Q_{2a} component is related to the typical *bending* mode of a triatomic molecule. The Q_1 *breathing mode* finds an analogue in the ^{12}C nucleus if its Hoyle state is considered as an equilateral triangular structure larger than that of the ground state. The Q_{2a} mode preserves a higher grade of symmetry with respect than the Q_{2b} stretching, an asymmetric mode which decreases the symmetry degree of the system; these two modes have the same frequency (i.e., are degenerate) only inside the framework of the harmonic approximation. However, beyond the harmonic approximation, this degeneracy is lifted by anharmonic effects such as the so-called *l*-type interaction, whose impact becomes evident when comparing theoretical and experimental spectra, as shown in Figure 1.20, reported from Ref. [16].

The spectra observed in Figure 1.20 correspond to the experimental measurement of the ν_2 fundamental band of the H_3^+ ion, obtained through high-resolution IR laser spectroscopy. The observed spectrum (bottom frame) has a complex fine structure, which is not reproducible with a simple harmonic model or with an effective hamiltonian neglecting the anharmonic interactions. A first theoretical simulation (top frame) considered the rotational levels associated to the ν_2 vibration, but did not include the *l*-type coupling, i.e., the anharmonic interaction taking place in the degenerate vibrational modes, where the *angular momentum* (l) can couple with the molecular rotation; in this way, the degeneration of levels with $l \pm 1$ is removed, generating the *l*-doubling in the spectrum. Neglecting this coupling, this simulation fails in the reproduction of the intensity, the density and the disposition of the experimentally observed spectral lines; specifically, the double structure which characterizes each rotational transition is completely missing.

At variance, the inclusion of the *l*-coupling between the degenerate vibrational levels allows an accurate description of the spectrum, as reported in the central frame of Figure 1.20. This coupling lifts the degeneration of the $l \pm 1$ components of the ν_2 vibration, generating an evident splitting of levels. This shows how the agreement between the observed spectrum and the theoretical calculations is highly enhanced when fully considering all the quantum mechanical implications due to the symmetries in the H_3^+ ion structure.

Section 1.2.3.8 will show that, also in the nuclear physics framework, the use of symmetry arguments is fundamental to unveil the structure of α clustered nuclei, by inspecting the occurrence of peculiar sequences of spin and parity values for excited states populating rotational and vibrational bands, including the presence of parity doubling of levels.

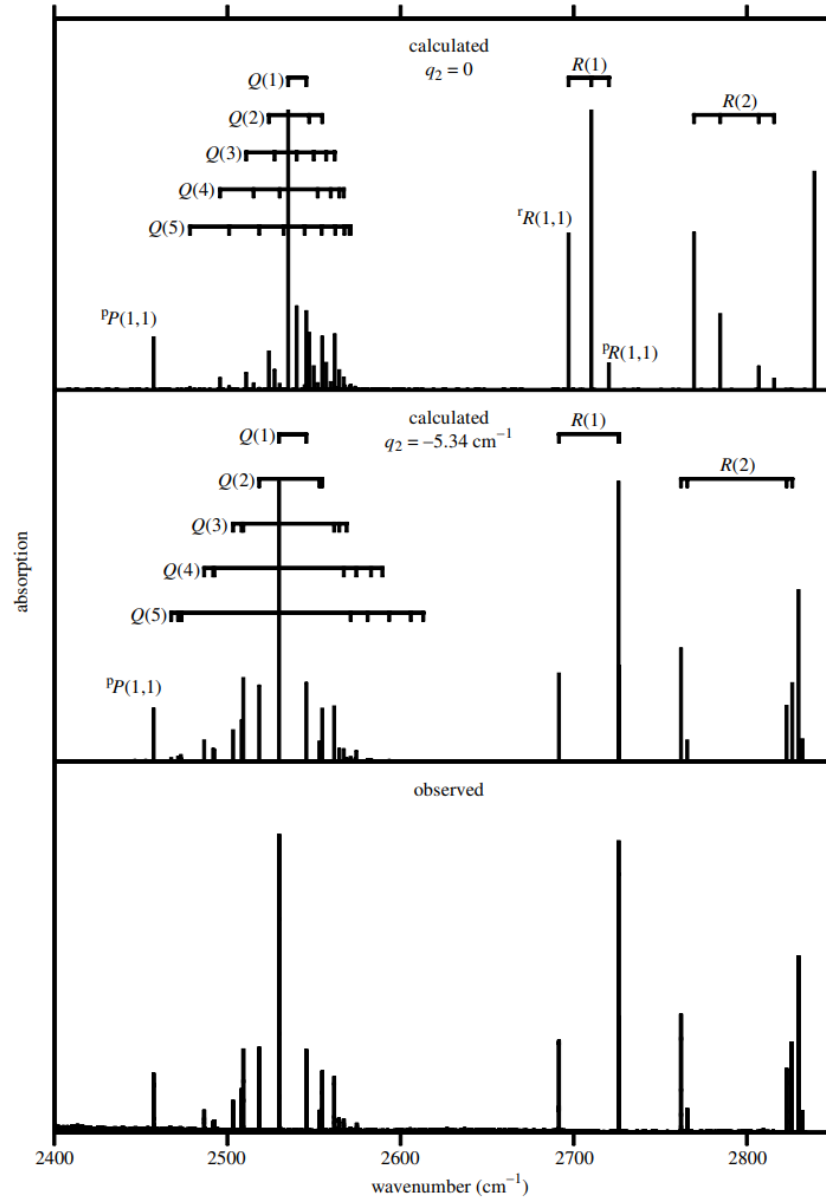


FIGURE 1.20: The figure reports the calculated and observed spectra of the fundamental ν_2 band of H_3^+ , showing the effects of ℓ -type doubling and resonance. The *central* spectrum is calculated with values of the parameters fitted to the observed spectrum, while, in the *top* diagram, the same parameter values are used, except that the q_2 and the higher-order ℓ -type doubling parameters are set equal to zero. *Bottom*: the same spectrum as observed by Ref. [21]. Figure taken from Ref. [16].

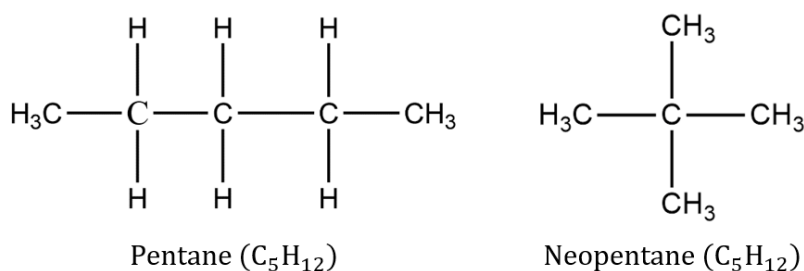


FIGURE 1.21: Planar projections of the pentane and neopentane molecules, isomers with 5 carbon and 12 hydrogen atoms.

1.1.6 Intermolecular interactions

Everything that was presented until this moment is mainly related to the interactions between subatomic particles (like protons and electrons in an atom) or between different atoms to form molecules. All of these concepts are important to rationalize the chemical properties of the compounds that make up the world, but they are not immediately related to the reasons behind the formation of *condensed* phases.

From the formation of liquid water to the ability of geckos to stick on almost completely smooth surfaces, the *intermolecular* forces are way more important than one may imagine. These types of interactions acts between certain regions of different molecules, depending from their three-dimensional chemical structure, the distribution of the electric charges and the deriving electric dipole and, of course, the temperature of the system.

The intertwining between molecular structures and symmetries and the properties of intermolecular forces is well larger than what could seem at a first thought. In general, these interactions are directly dependent from the three-dimensional structure of a molecule, but this aspect becomes even more evident in certain cases, i.e., for non-polar molecules. A molecule is non-polar if it does not have a permanent dipole momentum: this may happen because of its symmetry, or for molecules whose atoms have similar values of electronegativity. The simplest hydrocarbons, composed by only carbon and hydrogen, fall in the last category: the difference in the electronegative behavior of these two elements is pretty much negligible. This means that different molecules of a given hydrocarbon species have very few ways to attract each other and form a condensed phase: for this reason, the lightest species in the alkane series (C_nH_{2n+2}) are gaseous at room temperature.

The main possible way of interaction for non-polar molecules is, in fact, governed by the so-called *London* dispersion forces: these are derived from an instantaneous delocalization of the electronic charge density, which will form an *instantaneous electric dipole* in the molecule. After it is formed, this dipole will induce a charge delocalization in a nearby molecule, like in a normal dipole-induced dipole interaction, and so on; with this information, it is easy to forecast how this is strongly dependent from the available surface of interaction.

Figure 1.21 shows the difference in the structures of the pentane and neopentane molecules, two isomers with the same number and types of atoms, but with very different physical properties. The boiling point of pentane (Figure 1.21, left) is in fact of $\approx 36^\circ\text{C}$; on the other hand, that of neopentane (Figure 1.21, right) is almost thirty degrees lower

($\approx 9.5^\circ\text{C}$). This curious effect happens because the surface available for the action of the London dispersion forces is way higher for the linear structure of pentane. In fact, the pentane molecules can be packed in a better way with respect to its isomer, offering each other large portions of their surfaces, enhancing the interaction area and overall strength. This is also one of the reason why (other than for the molecular weight itself) the species in the alkane series have higher boiling and melting points proportionally to the length of the carbonious chain.

1.1.6.1 The Van der Waals forces

The aforementioned London dispersion forces are classified in the more general group of the *Van der Waals* forces, as defined by the homonymous physicist who draw a model for their behavior in 1873. This group of forces generally includes the following three sub-categories:

- The *Keesom* interactions, established between molecules with permanent electric dipoles. Although all the Van der Waals forces present anisotropic characters, the *Keesom* interactions are the only one which can actually result as attractive or repulsive as a function of the spatial orientation of the partners. The most intense interaction in this group is the Hydrogen bond, which will be presented in the next subsection.
- The *Debye* interactions, taking place when a molecule with a permanent dipole momentum induces a dipole in another system, since all atoms and molecules can generally be polarized.
- The *London* dispersion forces, which have been shown in the previous paragraph.

Other types of intermolecular interactions not included in any class of the Van der Waals forces are the ion-dipole interaction and the *halogen bond*; the last one will be discussed in more details, as it appears to be tied to some radiochemical aspects discussed in Chapter 6 and in the next subsection. The Van der Waals forces show again potential curves that are attractive for long ranges and repulsive at short ranges. The average energy of the different types of intermolecular interactions is well lower than that of the actual chemical bonds. While the energy of the latter is usually of the order of 100 – 1000 kJ/mol, that of intermolecular interactions is around tens of times lower, and only in some cases of hydrogen bonds can reach up to 50 – 100 kJ/mol.

However, *union makes strength*: although the intensities of the intermolecular interactions are nowhere near those of the actual chemical bonds, the high number of additive interactions can yield very important out-comings. When the intermolecular forces are capable of overcoming the natural thermal agitation of the molecules, they can be responsible for the formation of condensed phases or, for example, of several enzyme-substrate or inorganic catalysis interactions. For these reasons, these interactions have fundamental importance in scientific fields like nanotechnology, surface sciences, condensed matter physics, structural biology and supramolecular chemistry.

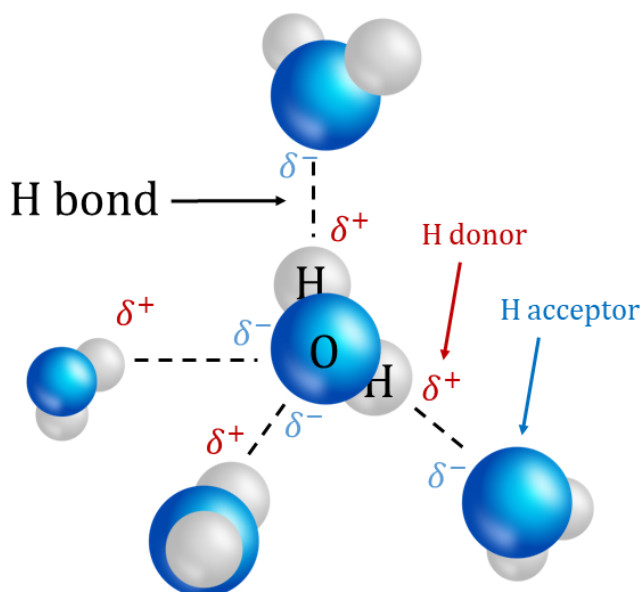


FIGURE 1.22: Schematic representation of the formation of hydrogen bonds between water molecules.

1.1.6.2 Hydrogen and halogen bonds

The hydrogen bond is a particular type of intermolecular interaction, which is usually distinguished from the others for its higher strength; in larger molecules, it can also manifest as an *intra*-molecular bond, if both the hydrogen donor and acceptor are part of the same molecular system. This type of bond can be formed when the H atom is bond and interacts with highly electronegative atoms, like **N**, **O** and **F**; the electronegative atom bond to the hydrogen is defined as the *Donor* (Dn) in the bond, while the other atom, bearing a lone pair of electrons (like the oxygens in the water molecules) will be the hydrogen bond *Acceptor* (Ac). Of course, an acceptor atom can accept as many hydrogen bonds as its number of electronic lone pairs (two in the case of the O in H₂O. Figure 1.22), allowing to form more complicate and regular structure; this is, actually, the reason why water ice has such a regular hexagonal structure and its density is lower than that of liquid water. Such a simple feature bears a lot of consequences: like the fact that ice floats on water, so lakes do not freeze completely, and the life underneath the surface can be preserved also in the hardest winters.

The hydrogen bond also accounts for the properties of materials like wool, hydrogels, and paper, but, most importantly, it is responsible for the high boiling point of water, at 100°C. The best way to understand the importance of hydrogen bonds is to put hydrogen sulfide (H₂S) into play; if one compares its structure to that of water, the two molecules are extremely similar. Same number of hydrogens, a central atom from the oxygen group, same linear bent structure (although with different angles and bond length); however, the boiling point of H₂S is $\approx 160^\circ\text{C}$ lower than that of water (-59.55°C). This large difference is mainly due to the presence of strong hydrogen bonds between water molecules, but absent in the H₂S system; sulfur is, in fact, less electronegative than oxygen, not suitable for the onset of significant hydrogen bonds, but only for weak dipole-dipole and London interactions.

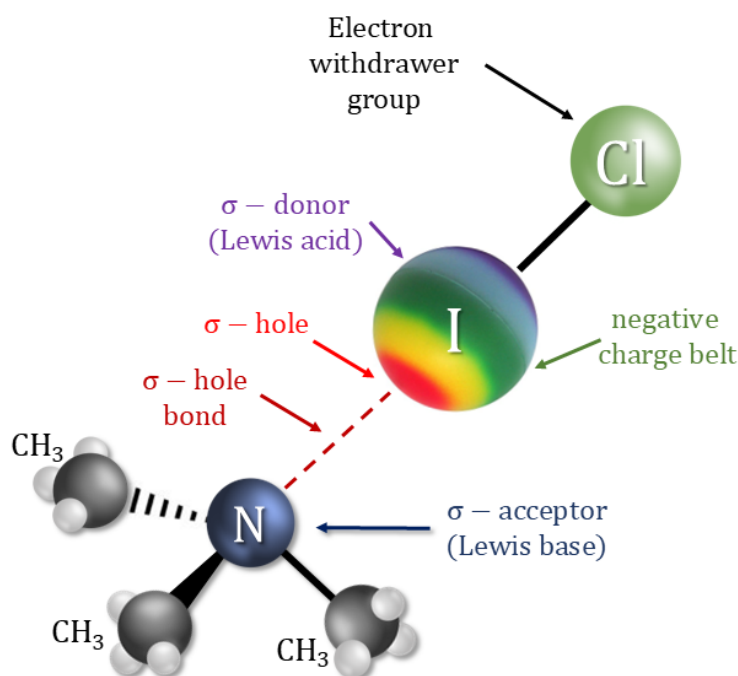


FIGURE 1.23: Schematic representation of the formation of *halogen bonds* between molecules. The σ -hole *skull-cap* in the Iodine electron cloud is indicated in red; it indicates a local depletion of negative charge able to trigger intermolecular bonds.

However, the hydrogen bonding is not a mere dipole-dipole electrostatic interaction; it is derived from a mixing of orbital interactions, charge transfers and quantum-mechanical delocalization of electrons, making it actually a resonance-assisted interaction. Nevertheless, not all types of hydrogen bonds are the same; some of them may be more defined and localized, such as those formed in ice or between the nucleotide bases in DNA strands, while those in liquid water or supramolecular assemblies are weaker and more delocalized, with intensities similar to the other Van der Waals forces.

As already mentioned, a similar type of interaction is the so-called halogen bonding: this is a weak intermolecular force, with a mechanism similar to the Lewis acid-base interaction. In this case, as reported in Figure 1.23, the halogen acts as a Lewis acid, since it is a big atom with a partial $\delta^+ - \delta^-$ charge separation, accepting electronic charge from the Lewis base on its δ^+ region [22]. Differently from the hydrogen bond, there is no need for the presence of other electronegative atoms, rather than the halogen itself; the halogen bond is thus usually formed (and studied) in alkyl-halide systems, like iodomethane. CH_3I is often used since the iodine atom is characterized by a large σ -hole on its cap, an electro-positive region which is accentuated in heavier and polarized halogens, like iodine.

In the last Chapter of this thesis, a peculiar role of intermolecular bonds in the formation of trimeric clusters of molecules (driven essentially by the coupling of hydrogen and halogen bonding, with a marked directionality) in organic chemistry environments will be presented. It is in fact possible that such molecular clustering effect could have a prominent role in the determination of radioisotope *retention* processes in hot atom

chemistry experiments, and therefore that nuclear chemistry experiments could represent a way to unveil molecular clustering effects that is complementary to the spectroscopic methods (like infrared, ultra-fast and resonance Raman spectroscopy) already in use [23–25].

1.2 The fall of simplicity: the Nuclear force

... into what pit thou seest
from what height fallen: so much the stronger proved
he with his thunder; and till then who knew
the force of those dire arms?

J. MILTON, *Paradise Lost*, Book 1, 91-94

The atomic nucleus entered the field of physics studies as a crucial component of the atomic model, but the fact that the study of the structure of atomic nuclei implied the knowledge of a new type of force, outside the natural everyday experiences, has always made its description a particularly overwhelming task. The investigation on the nuclear forces is, in fact, one of the scientific question on which the humanity has actually spent the most time on, with respect to any other type of research problem. Today, nuclear experiments provide elegant methods for the determination of nuclear properties and a solid demonstration of the power of quantum mechanics. It is possible to think about two approaches to perform the study of the nuclear structure phenomena in the "easiest" available way, and they will be presented in the following subsection, in analogy to the molecular systems reported in the beginning: in the *deuteron* system, where a neutron and a proton are bound together, or in *nucleon-nucleon* scattering experiments.

All the discussions in this Section will prove that nuclear physics is not an isolated subject, and it is also not self-sufficient; it rather is a crucial brick of a bigger structure, alongside with mathematics, chemistry, and other branches of physics, allowing to deepen the mankind's knowledge on the nature of matter at really profound levels, beyond the electronic structure of atoms and molecules. Anyway, what once seemed a simple model of matter soon reveals itself as a vast and intricate tapestry. As John Milton wrote in *Paradise Lost*, knowledge is both a burden and a power: the fall from simplicity and order leads not only to darkness, but also to a deeper insight. So too, the fall of simplicity experienced, at a first sight, in studying Nuclear Physics opens the way to richer, more fundamental, truths.

1.2.1 Some properties of the nuclear forces

The fact itself that stable nuclei are present in the world is a direct proof of the existence of some sort of nuclear interaction, since the Coulomb repulsion between the protons would otherwise make any nucleus unstable. The following subsection will review the different properties of the nuclear forces: this will be done through the study of the simplest bound system, the deuteron, and by the means of the n-p scattering and the exchange interactions. More complicated frameworks can be treated through suitable nuclear force potentials and with the study of three-body forces.

1.2.1.1 The deuteron

The deep knowledge that has been attained over the atomic structure and properties derives from a profound study of the simplest atom, the hydrogen ^1H , made up by one proton and one electron; in the same way, the most direct information for the nucleon-nucleon interaction has to come from the study of a system composed by *two*

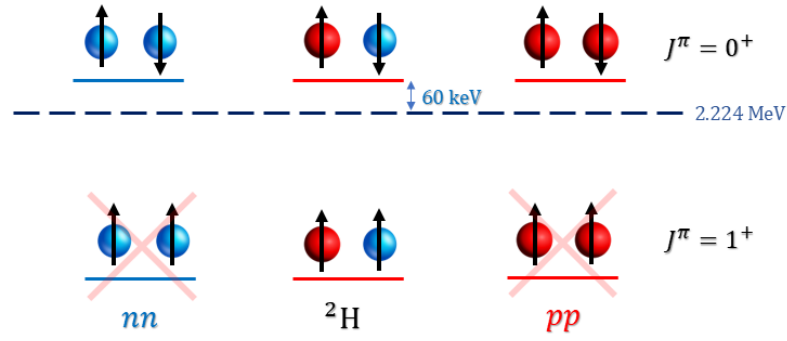


FIGURE 1.24: Schematic representation of the ground and first excited states of the three possible di-nucleon systems: the *di-neutron* (nn), the *deuteron nucleus* (np) and the *di-proton* (pp). Of these three, due to the Pauli's exclusion principle, the only bound ground state is that of the deuteron.

nucleons. The great part of the available information derives from nucleon-nucleon scattering experiments; but, luckily, a single bound state of the neutron-proton system exists. This is the *deuteron*, i.e., the nucleus of the heavy hydrogen atom (${}^2\text{H}$), and it allowed to obtain priceless information on the nucleon-nucleon potential nature, and some really preliminary leads on the understanding of the properties of heavier nuclei. The deuteron is, in fact, the only nucleon-nucleon available bound system: the other two, the di-proton (${}^2\text{He}$ nucleus) and the di-neutron (${}^2\text{n}$), as it will be later shown, are not bound (Figure 1.24). Just as a side-note, if the pp system were to exist as a bound state, the Sun would "quickly" burn *all* the hydrogen, transforming it in the mysterious *di-proton*, which could then maybe have a β^+ decay towards a deuteron state; this is of course not happening, since the stars live, instead, for very long times ($\simeq 8 \text{ Gy}$).

One of the most important properties of the nuclear force is that it is an extremely short-range interaction: it is so short-ranged that a nucleon in a large nucleus (let's say with more than ten nucleons) is not even interacting with all of its comrades, but only with the first neighbors. On the basis of this, the binding energy of the nucleus is thus varying proportionally to the mass number A , and not to $\frac{[A(A-1)]}{2}$, which would be the number of interacting pairs if the nuclear force had a range large enough to cover all the nucleus. The actual range of the nuclear interaction is thus of $a \approx 1.5 \times 10^{-15} \text{ m}$ [26].

The main assumption, crucial to simplify the study of the problem, is that the interaction is described by a central potential, exclusively dependent from the distance between the nucleons, and that the ground state has an angular momentum equal to zero (like in the hydrogen atom, it is a S -state). The simplest structure that can be assumed for its ground state is, in fact, a system in which the two nucleons are in an S -state of orbital motion and with parallel spins, a ${}^3S_1$ state with even parity; on the other hand, the ${}^1S_0$ (with antiparallel spins) is an unbound state. However, when trying to describe the scattering problem, the ${}^3S_1$ state alone is not enough: both the triplet and singlet states are needed for a successful description [27]. In the hydrogen atom, the interaction potential is known (since it is based on the Coulomb force), and the task is to find the various possible energetic levels. In the deuteron system, the binding energy is experimentally determined (by detection of the γ rays produced after the capture of slow neutrons from a hydrogen-containing target), and used to establish the boundary conditions on the potential shape. The binding energy is the amount of energy required

to separate the two nucleons, and it is equal to the difference between the rest energies of the neutron and the proton and that of the deuteron:

$$\varepsilon = (m_p + m_n)c^2 - m_d c^2 \simeq 2.224 \text{ MeV} \quad (1.9)$$

If the two nucleons are bound together, in the framework of the deuteron system, by a potential of range a , the distance separating the two must be of the same order of magnitude of the potential range itself; otherwise, this would not be able to hold them together. However, this poor knowledge on their position, on the basis of the Heisenberg's indetermination principle, imposes that their relative momentum must at least be of the order of $p \simeq \frac{\hbar}{a} \simeq 130 \text{ MeV}/c$; of course, their mutual kinetic energy would be $T = \frac{p^2}{2\mu} \simeq 20 \text{ MeV}$, where μ is the reduced mass of the system (half the mass of a nucleon). The binding energy ε is the difference between the *negative* attractive potential energy (V), keeping the particles bound together, and the kinetic energy (T), tied to their relative motion, which naturally tends to disturb the bonding condition. Since ε is well smaller than T (and V), in a first, rough approximation, it is possible to consider the potential as the usual *square well*, with radius a and depth V .

$$V(r) = \begin{cases} -V & \text{if } r \leq a \\ 0 & \text{if } r > a \end{cases} \quad (1.10)$$

The Schrödinger equation can thus be written as following, by indicating the eigenfunction as $\chi(r)$; then, to search for the energy eigenvalue $-\varepsilon$ for the nucleons moving in this potential, representing a state with angular momentum $\ell = 0$:

$$\left[-\frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial r^2} + V(r) \right] \chi(r) = -\varepsilon \chi(r) \quad (1.11)$$

Following classical quantum mechanics prescriptions, it is then possible to develop the solutions for this equation [26], under the imposition of the suitable boundary conditions, and obtain a relation between the depth (V) and the extension (a) of the spin-triplet and isospin-singlet well [26]:

$$a^2 V = \frac{\pi^2 \hbar^2}{4m_n} \rightarrow a = \frac{\pi \hbar}{2\sqrt{m_n V}} \quad (1.12)$$

where m_n is the mass of a nucleon, and $a^2 V$ is effectively related to the *volume* of the potential, independently from its shape; this condition is directly coming from experimental data proving that the binding energy of the deuteron is low. Now, apart from the extension of the potential a , it is possible to give an estimate of the root mean square radius of the deuteron *per se* from electron scattering experiments $r_d \approx 5 \text{ fm}$. But the wavefunction of the deuteron, in the $r > a$ region, is decreasing as $\exp(-Kr)$; from the Heisenberg principle, one can connect the mean radius of the deuteron with the external wavenumber as $r_d = 1/K = \frac{\hbar}{(m_n \varepsilon)^{\frac{1}{2}}}$. It is then possible to compare the mean radius of the deuteron and the extension of the nuclear potential:

$$\frac{r_d}{a} = \frac{1/K}{a} = \frac{2}{\pi} \left(\frac{V}{\varepsilon} \right)^{\frac{1}{2}} \quad (1.13)$$

Assuming as the range of nuclear forces the sum of the nucleon radii, one can roughly estimate $a \simeq 2.5 \text{ fm}$. The $\frac{r_d}{a}$ term is then well larger than 1, and this means that the

deuteron is a weakly bound nucleus, where the particles spend the large part of their time *beyond* the radius of the attractive potential, which holds them together. Furthermore, the previous equation would lead to a depth of the (real) potential well of $V \simeq 30$ MeV, much more than the binding energy of the deuteron: this is a further indication of the weak intensity of the bond, together with the experimental fact that the deuteron has no *bound* excited states.

1.2.1.2 Neutron-proton scattering and exchange interactions

Another powerful tool to draw some information about the basic properties of the nuclear force is the scattering of low-energy neutrons from proton (hydrogenated) targets; the low energy of the neutron beam allows to select events with angular momentum $\ell = 0$, and to use non-relativistic prescriptions. Of course, given the high mass difference between electrons and neutrons, the hydrogen target can be considered as analogue to a proton target: the collision between an electron and a neutron would be similar to that between a car and a tennis ball. The possibility to use neutron beams even at very low (thermal or sub-thermal) energies to induce nuclear scattering or reactions is granted by the absence of a Coulomb interaction between the proton and the neutron [27]. The n-p scattering is also the best option in terms of practical feasibility: the p-p scattering is hindered by the presence of the Coulomb barrier, while for the n-n scattering the main technical difficulty is related to the impossibility of having a target made of free neutrons. In both cases, the fact that the target and the projectile are identical particles leads to further quantum mechanical complications in the interpretation of data [28]. The scattering process takes the place of the binding interaction if the kinetic energy of the nucleons pair, $\frac{1}{2}(\hbar^2/\mu r^2)$, is higher, in absolute value, than the interaction potential V_0 , corresponding to the equilibrium distance. The only inelastic reaction that could take place in this energy range is the capture reaction $n + p \rightarrow d + \gamma$, which contribution is however lower than 1% of the scattering cross section, and can thus be disregarded for this Chapter's purposes. It is possible to treat this problem as a function of the binding energy of the deuteron. Let's re-define the potential as:

$$U(r) = \frac{2\mu}{\hbar^2} V(r) \quad (1.14)$$

Obviously, given the finite range of nuclear forces, it is assumed that $V(r)$ is null beyond a finite range r . Therefore, if the energy of the particles in the center of mass frame is $\hbar^2 k_1^2 / 2\mu$, the eigenvalue equation is satisfied by the χ_1 function:

$$\left[\frac{\partial^2}{\partial r^2} + k_1^2 - U(r) \right] \chi_1(r) = 0 \quad (1.15)$$

This relation is also valid for a different momentum or wavenumber (e.g., k_2) and the respective wavefunction (χ_2). Following a short sequence of mathematical steps [26], one can obtain:

$$\left[\chi_2 \frac{\partial \chi_1}{\partial r} - \chi_1 \frac{\partial \chi_2}{\partial r} \right]_0^R = (k_2^2 - k_1^2) \int_0^R \chi_1 \chi_2 dr \quad (1.16)$$

If $\phi_i(r)$ is the asymptotic form of the $\chi_i(r)$ function, valid for large values of r , on the same basis:

$$\left[\phi_2 \frac{\partial \phi_1}{\partial r} - \phi_1 \frac{\partial \phi_2}{\partial r} \right]_0^R = (k_2^2 - k_1^2) \int_0^R \phi_1 \phi_2 dr \quad (1.17)$$

because $\phi_i(r)$ satisfies the same equation as $\chi_i(r)$ if $V(r)$ is considered null for large values of r . Then, for very large ranges ($R \rightarrow \infty$), when $\phi_i(R) = \chi_i(R)$ and $\chi_i(0) = 0$, when subtracting Eq. 1.17 from Eq. 1.16, one obtains

$$\left[\phi_2 \frac{\partial \phi_1}{\partial r} - \phi_1 \frac{\partial \phi_2}{\partial r} \right]_{r=0} = (k_2^2 - k_1^2) \int_0^\infty (\chi_1 \chi_2 - \phi_1 \phi_2) dr \quad (1.18)$$

When k_2^2 is positive, the state is representing a *scattering state*, with

$$\phi_{k_1}^{(r)} = \phi_k^{(r)} \simeq \frac{\sin(kr + \delta)}{\sin \delta} \quad (1.19)$$

where the normalization condition is set as $\phi_k(0) = 1$. On the other hand, if k_2^2 is negative, the *state* is *bound*, with $k_2^2 = -K^2$, as set when talking about the deuteron system. In this case,

$$\phi_{K_2} = \phi_K = \exp[-Kr] \quad (1.20)$$

with the same normalization condition, $\phi_K(0) = 1$. Since the interaction potential has a short effective range, the asymptotic forms (ϕ) are equal to the respective exact wavefunctions in the almost whole integration interval of Eq. 1.18. Given that K is directly related to the binding energy ($K^2 = \frac{2\mu\varepsilon}{\hbar^2}$), the phase shift in the low-energy scattering of the *S*-wave, δ , can be determined as a function of the binding energy of the deuteron. For small values of k , in fact:

$$\delta \simeq -k/K \quad (1.21)$$

showing, under comparison with the phase shift in the scattering from a rigid sphere ($\delta = -ka$, where a is the radius of the sphere), that the radius of the deuteron ($1/K$) is equal to the radius of the equivalent rigid sphere, when treated in conditions of very low energies. The study of the phase shifts as a function of the energy is a convenient way to represent the experimental results and a starting point for the theoretical analysis of the potentials. On the basis of the same approximations, the cross section is related to the binding energy ε of the *triplet* system (the deuteron) and derived as follows:

$$\sigma_{triplet} = \frac{4\pi \sin^2 \delta}{k^2} \simeq \frac{4\pi \delta^2}{k^2} = \frac{4\pi}{K^2} = \frac{4\pi \hbar^2}{m_n \varepsilon} \quad (1.22)$$

When including the experimental values in this equation, it is possible to obtain a cross section of about 2.2 barns; however, if compared with the experimental data of ≈ 20.3 barns for the n-p scattering cross section in the region $1 < E_{kin} < 1000$ eV, the difference between the two values is pretty high. The solution of the dilemma was, once more, suggested by Eugene Wigner in 1935. This is to impute to the fact that nucleons, like electrons, are characterized by having a *spin* moment, which was not taken into account in this derivation; the inclusion of this feature allows to obtain an accurate theoretical description of the experimental scattering cross section. The phase shift δ is also related to the *scattering length* a of the given state ($\tan \delta \propto a$): this quantity can be geometrically seen as the tangent to the wavefunction calculated in the points where the nuclear well vanishes. A positive value for the scattering length a is then an indication of the existence of a *bound state* (as in the deuteron case), a system that can be obtained when the neutron and the proton collide with parallel spins, with *triplet* multiplicity ($^3S_1$). Nevertheless, a different scattering state, the singlet ($^1S_0$) state, is obtained when the two nucleons collide with opposed spins: this is a non-stable state of the deuteron (the "virtual deuteron" d^*), unbound, which would involve a different

(both in depth and width) potential well. Considering the statistical weights of the triplet and singlet contributions, the experimentally observed elastic scattering cross section is actually given by Equation 1.23:

$$\sigma = \frac{3}{4}\sigma_{triplet} + \frac{1}{4}\sigma_{singlet} \quad (1.23)$$

By using the value $\sigma_{triplet} \simeq 2.2$ barn obtained above and the $\sigma = 20.3$ barn experimental value, it is possible to deduce the *huge* value $\sigma_{singlet} \simeq 75$ barn. This is a clear indication of the fundamental fact that nuclear forces are strongly dependent on the coupling of spins of the interacting nuclei! Furthermore, the *unbound* nature of the singlet state of the deuteron, and then the sign of its scattering length, were proven almost a century ago, thanks to the use of targets made by hydrogen (H_2) molecules and to the fact that the relative spins of the two H nuclei can be parallel (ortho-hydrogen, $S_H = 1$) or anti-parallel (para-hydrogen, $S_H = 0$). In 1937, J. Schwinger and E. Teller, with a wonderful flight of the mind, suggested that a scattering experiment, performed with slow neutrons on an ultra-cold ($T < 20$ K) molecular hydrogen target, could have the power to discriminate the cross section on the ortho- and para-hydrogen phases of the systems, given that all the isomers of the latter (anti-parallel spins of the protons) would flip to the spin-aligned singlet system at temperatures lower than 20 K [29]. Some *virtuoso* mathematical procedures involving the scattering length of ortho- and para-hydrogen suggested that the scattering length of the singlet state (a_s) can be negative only if the cross section of the slow neutrons on the para-hydrogen is way lower than that on the ortho-hydrogen molecule ($\sigma_{para} \ll \sigma_{ortho}$); these σ were experimentally measured by Sutton et al. in 1947 [30], proving once and for all the *unbound* nature of the singlet state in two-nucleon systems. All this beautiful story underlines, once more, the clear evidence of an extremely *spin-dependent* nature of the nuclear interaction potential, differently from the cases of electromagnetic and gravitational forces.

This scheme is also explaining, in combination with the Pauli exclusion principle, why the deuteron is a bound nucleus, while the di-neutron and the di-proton have never been observed. Nevertheless, although one could think of looking for this explanation in some sort of favorable symmetrical n-p pairing, the fact that the triplet $^3S_1$ state represents the bound state of the two nucleons in the deuteron is an evidence that it is only possible for two *different* particles. If two neutrons or two protons were to have parallel spins to form a triplet state, this would be a clear violation of the Pauli's principle (Figure 1.24), on the other hand, when the spins of one proton and one neutron are anti-parallel, they form a singlet state ($^1S_0$) which, as seen before, is unbound. For this reason, the study of the deuteron system is the main source of information about the triplet state potential, while the nucleon-nucleon scattering is important to study unbound states and the singlet potential, and its different partner combinations prove the *charge independence* of the nuclear force.

1.2.1.3 Potential shapes for nuclear forces

Given the nature of the nuclear force, it is possible to expect some particular requirements for the potential representing it; first of all, it is necessary for it to be a scalar quantity, since it is an energy. On the basis of the fact that protons and neutrons are indistinguishable in the framework of this interaction, the potential must remain unchanged upon the exchange of the two partner particles. Finally, the *principle of*

time-reversal invariance must hold, i.e., the potential must stay the same even if the time flow direction is reversed; this principle is generally valid in various well-known scenarios in classical and quantum physics. These conditions pose some vetoes on the existence of some of the terms of the potential, like $\vec{r}_{12} \cdot \vec{p}_{12}$, $\vec{r}_{12} \cdot \vec{S}$ or $\vec{r}_{12} \cdot \vec{L}$, with all the symbols maintaining the same meaning as in the previous Sections [31]. The simplest available models for the study of the nuclear force are those based on static potentials, which cannot depend on the momentum $\vec{p}$ or on the orbital angular momentum $\vec{L}$, being only based on the distance $\vec{r}$ and the spin $\vec{S}$. Another property on which a static nucleon-nucleon potential can depend on is the *parity* of the wavefunction describing the system; since this depends on L being *even* or *odd*, the nuclear force will be different for *even* or *odd* L . From the different combinations of all these parameters, it is possible to draw a list of *six* possible static forces. Unfortunately, various nuclear scattering experiments proved that the nuclear force is not static at all, making it therefore useful to consider simple velocity-dependent forces or, better, *momentum-dependent potentials*. The most common approach to study the momentum dependence of the nuclear potential is to investigate the nuclear force at continuously increasing energies, typical of those of Fermi motion of nucleons in nuclei, but kept lower than few hundred MeVs (at higher energies, the production of pions (π) starts to arise). One of the simplest, and most important, terms to take into account when studying this type of dependence for the nuclear potential is the *spin-orbit coupling* or *interaction*, $\vec{L} \cdot \vec{S}$. The magnitude of this term depends on the relative angle between the two vectors and, of course, on the module of both $\vec{L}$ and $\vec{S}$; it vanishes when $\vec{S} = 0$. However, as discussed in [31], it is not possible to obtain even a semi-quantitative fit of the two-nucleon systems data without the inclusion of higher-order velocity dependent terms, like terms of the second-order with respect to $\vec{p}$ (i.e., terms containing two powers of $\vec{p}$ or $\vec{L}$, or one of each). Unfortunately, the possible number of such terms is extremely high, and it is very difficult to consider and determine their importance; the most common approach is to just adopt some of the simpler ones, to obtain a fit to the experimental data good enough for the specific purpose.

The short-range character of the nuclear force is to be imputed to a particular property of the exchange particle mediating it: it has a finite mass. While, e.g., the photon, which mediates the electromagnetic interaction, has zero mass and travels at the speed of light, the pion π (mediating the *effective* nuclear force) has a mass of $m_\pi c^2 \approx 140$ MeV. The short range is a consequence of the fact that, for the pion to be produced by one nucleon and transmitted to the other, a violation of the conservation of energy of $\Delta E \approx m_\pi c^2$ must happen; however, on the basis of the uncertainty principle, this can only last a short time: $\Delta t = \hbar/\Delta E$. During this narrow time window, the pion can only travel as far as:

$$r = c\Delta t = \frac{\hbar}{m_\pi c} \equiv \frac{1}{\mu} \quad (1.24)$$

i.e., about 1.4 fm. Given that this is also the De Broglie wavelength of 350 MeV neutrons, and it is not possible to go over this value of energy without encountering large experimental difficulties, this also pose a limitation on the scale of the information obtainable with nucleon scattering experiment. The *meson field theory*, just very briefly sketched, allows to write another form for the potential energy of the interaction between two nucleons, formulated by H. Yukawa in 1937, and therefore named "the Yukawa potential":

$$V = -\gamma^2 \frac{e^{-\mu r}}{r} \quad (1.25)$$

where μ is defined from Eq. 1.24, and γ is a constant which occupies an analogue role in meson theory as the electron charge e in the electromagnetic theory.

Bearing all the features (and most important, the limitations) exposed in the previous paragraphs it is possible to understand one of the crucial features of the nucleon-nucleon force, i.e., that it is characterized by a *repulsive core*. The potential, indeed, becomes enormously repulsive (tends to $+\infty$) at distances lower than 0.5 fm, as the exchanges of other (much more massive) mesons, different from π , start to take place.

On the light of the previously sketched phenomenology, one of the most comprehensive potentials to describe the nucleon-nucleon force is the so-called *Hamada-Johnston* [32] one. This formalism uses a total of twelve components, with some spin-orbit potentials in addition to the static potentials mentioned above, for *even* and *odd* L (referred to as $L - S$), and other second-order velocity dependent potentials of the form $[\vec{L} \cdot \vec{L} - (\vec{L} \cdot \vec{S})^2]$, also different for *even/odd* L and for $S = 1, 0$. This potential includes also a *tensor* term, needed to explain the presence of a (small) *quadrupole* momentum in the deuteron, with a consequent rupture of the spherical symmetry. Every one of these components is usually either repulsive (positive) or attractive (negative). It is actually necessary to understand that the effective potential experienced by a couple of interacting nucleons is somehow the sum of all the applicable potentials. For example, if their spins are parallel and the state has $L = 1$, only seven of the twelve potentials are active: four *central* terms ($S = 0$ and 1 both with even and odd L), the *tensor* and the $L - S$ terms with $S = 1$ and odd L , and last, the $L - L$ term with $S = 1$ and odd L . Of course, when the two interacting nucleons are protons, it is necessary to add another *small* term related to the Coulomb potential.

One could focus the attention on the term which, among the twelve different ones, appears to be most important in the ground state of the deuteron, i.e., the $S = 1$, even- L , central potential (third in the bottom row of Figure 1.25). It is of course interesting to compare it to the previously used approximation of a square-well potential (red-dashed line). Some very qualitative similarities can be drawn for the calculated wavefunction, especially if the square-well potential is considered to extend down to $r = 0$.

Other potentials are also discussed in the literature to describe the two nucleons force, fitting a large body of available experimental data [33, 34], on the basis of a more complex velocity dependence, and different prescriptions for the hard repulsive core. The qualitative similarities, but most of all the differences, between these potentials and the Hamada ones have been historically considered as a measure of the uncertainty of the knowledge of the nuclear force. It is worth noting that several famous *realistic shell model* approaches (as the one developed by Tom Kuo and Gerry Brown in 1966) made use of this type of potentials to describe the structure of the lighter-most nuclei [35].

1.2.1.4 Three-body forces

To complicate the treatment of the nuclear force, beside complex and non-central potentials, also arises the fact that this force can not once again be treated as the gravitational and electromagnetic force: i.e., it is not a *two-body* interaction. When three or more nucleons are present in mutual proximity, being the nuclear force based on meson exchange, it is not possible to consider the force acting on the first nucleon just as the

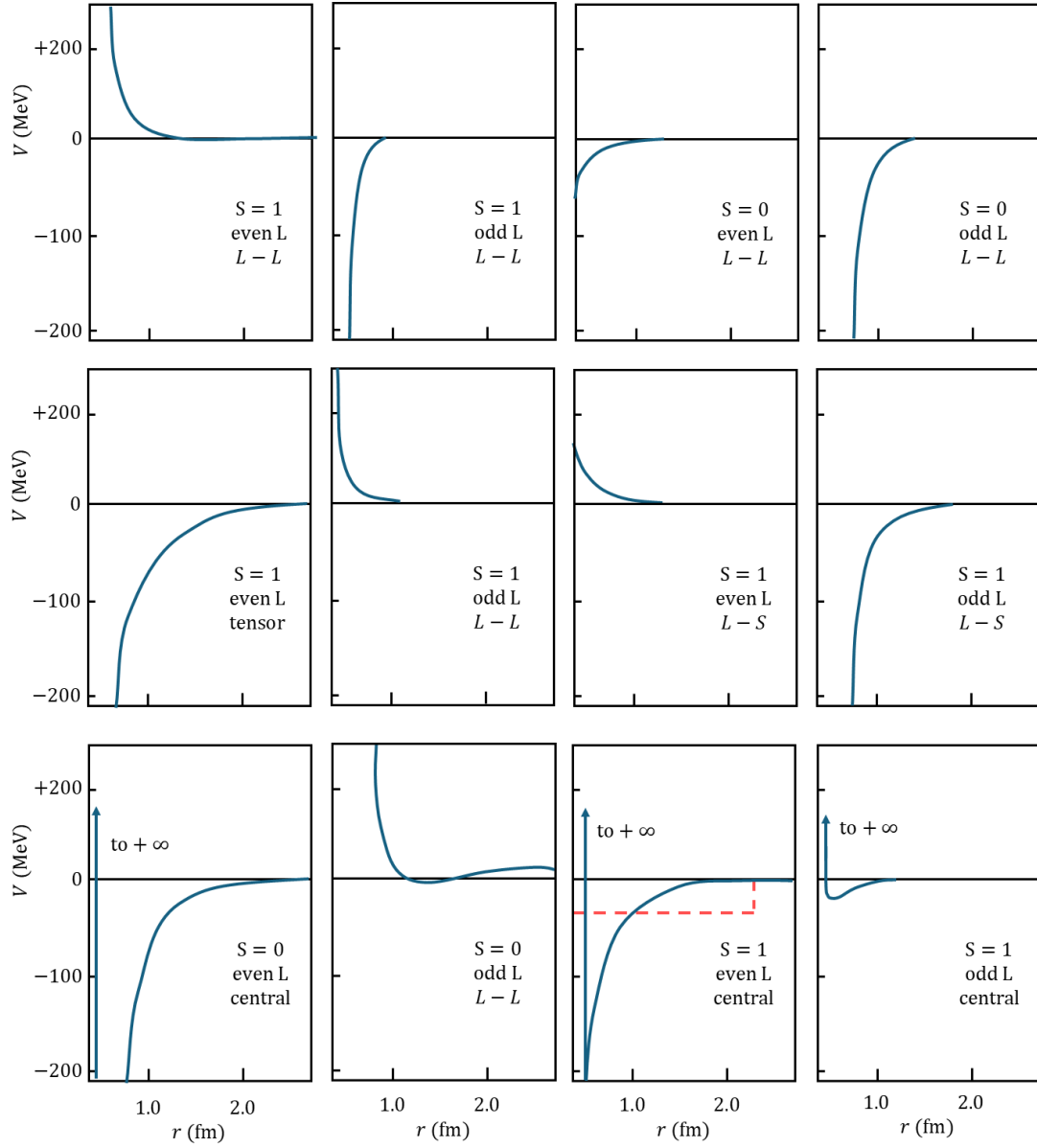


FIGURE 1.25: The twelve components of the Hamada-Johnston potential formalism. The 11th graph, third from the left in the bottom row, shows a comparison between the $S = 1$, even L central term (solid blue line) and the common square-well prescription (red dashed line).

sum of that existing with the second and that experienced with the third. Given that many mesons can be created simultaneously in these types of processes, it is possible to extrapolate the discussion from system of three to those of *many* nucleons, and thus study the *many-body forces*. This thus requires that the range of the force, defined in Eq. 1.24, would decrease proportionally to the increase of interacting bodies. A rough estimate sets the range of an n -body force as the range of the two body force, multiplied by a damping factor $1/(n - 1)$. If the range r , as in Eq. 1.24, is calculated as ≈ 1.4 fm, it will become 0.7, 0.47, 0.35 fm (and so on) for 3, 4 or 5 interacting particles.

It is possible to estimate the nucleons' volume and their mutual distance inside a nucleus, considering that the volume of a single nucleon is $\frac{1}{A} \cdot V_{nucleus} \simeq \frac{4}{3}\pi(1.07 \text{ fm})^3$: the radius of a nucleus is, on average, 1.07 fm, making the mean distance between the nucleons of ≈ 2.1 fm. Of course, they also can not approach each other close than 0.5 fm, because of the presence of the repulsive core of the nuclear potential. The value of the average distance is however higher than the range of the higher-order many-body forces, and it is therefore justifiable to expect their contribution to have a secondary importance. The main role in the determination of the nuclear structure characteristic is thus of the *two-body* forces, with a small, yet not negligible, contribution from the *three-body* forces, usually studied in systems of three (or more) nucleons. These are approached through a *tentative* description of their properties based only on the superposition of two-body forces; any discrepancy in this representation attempt would thus be an evidence of the presence of *three-body* forces. As for the case of two-body systems, the three-nucleons systems can be investigated via the study of bound states, like ^3H and ^3He , or by scattering of neutrons/protons from deuterons, an experimental approach which allows to obtain a large body of information (see, e.g., [36]).

The analyses of these data are however very difficult, since in classical and quantum mechanics there is no exact solution to the three-body problem; nevertheless, it seems possible to have at least a glimpse of the contribution given by the *three-nucleons* interaction in a three-body system, the triton ^3H , whose binding energy is experimentally known to be of 8.48 MeV. If it is possible to estimate the binding energy coming from the *two-body* interaction to ≈ 7 MeV, the residual ≈ 1.5 MeV should be implicitly coming from *three-body* forces, in agreement with estimates obtained with other approaches. It is thus possible to conclude that the contribution of the higher-order forces (mainly *three-body*) inside atomic nuclei is of around 20% of the magnitude of the potential generated by the *two-body* forces, decreasing to approximately 15% for heavier nuclei [31].

1.2.1.5 A short summary of the properties of the nuclear force

What discussed in the previous paragraphs of this Section may be helpful to summarize some of the most important and immediate properties of the nuclear interaction, here listed [37]:

- First of all, the two-body nuclear potential, at the basis of the description of the nuclear interaction, is described by two parameters: its *depth* (the "strength") and its *range*, the limit beyond which the potential starts to decrease to zero.
- The range of the potential, through studies on the deuteron system and nucleon-nucleon scattering experiments, is found to be *smaller* than the average nuclear

radius: this means that a nucleon inside a larger nucleus is usually not capable to interact with all the others, but only with its immediate neighbors.

- Neglecting the influence of the Coulomb repulsion between protons, it is possible to state that the nuclear interaction is *independent* from the *charge* of the partner nucleons: n-p, n-n and p-p interactions are of the same magnitude. However, not all the pairings are successful in the same way: a *spin-dependence* is required for the nuclear force, meaning that nucleons with parallel spins are more strongly attracted with respect to those with antiparallel spins, as seen with triplet and singlet states in nucleon scattering experiments and in the study of the ^2H system.
- (This sounds more like a *preview* than a *conclusion*): some sort of *saturation phenomena* must occur inside the atomic nucleus, especially when taking α particles into account. These will be discussed in the following Section, and it is possible to assume, beforehand, that the attraction between nucleons occupying the same orbital state is seriously greater than that between nucleons in different states, albeit the latter is certainly not null.

1.2.2 Complex nuclei and saturation of the nuclear forces

In order to grant a regularity in the description of the nuclear structure, and to avoid the collapse of all the different structures to just ultra-dense clusters with radius equal to the range of the nuclear force, one of its most important properties must be the eventuality to reach a *saturation*.

A *field theory* of the nuclear force allows to account for the properties of the two-body interaction and for the description of more complex nuclei, with a binding energy somehow proportional to the number of nucleons but, most importantly, with a *nuclear density* which is independent from it, as a consequence of the aforementioned saturation. It is useful to think of this property in analogy to condensed phases of ordinary matter: the density of water, in the same conditions of temperature and pressure, is practically no different in any way between a glass of sea water and the Pacific Ocean.

One can easily see that it is not possible to assume the presence of an attractive potential between all pairs of nucleons in a more complex nucleus, thus complicating the description of these systems; if one were to apply the same potential to every possible nucleon pair in a complex nucleus, the resulting forces would be of the so-called *ordinary* (or "Wigner") type. If this was true, the average potential energy *per nucleon* would be proportional to the number of nucleons lying in its close proximity, i.e., between the range of the nuclear force; as a consequence, the potential energy would increase proportionally to the density. When the latter increases, the volume available for each nucleon would clearly diminish, and the average kinetic energy increase. In the framework of ordinary forces, the increase of the kinetic energy term and the decrease of the nuclear force are of different magnitude, and this would lead to the point in which all the nucleons are concentrated within the range of the nuclear forces to each other [38]. The radii of all nuclei would then substantially collapse to the range of the interaction and the binding energy would be equal to the *square* of the number of nucleons.

What stated earlier is, of course, in absolute contradiction with all the known properties of the nuclei, whose radii and binding energies only increase proportionally to, respectively, $A^{1/3}$ and A . It is thus evident that some properties of the nuclear force

must be extraneous to those of ordinary forces: i.e., a nucleon must be attracted to only a *limited* number of other nucleons in one nucleus, with the nuclear interaction showing some sort of *saturation character*. The simplest (albeit not exhaustive) ways to account for the saturation phenomena could be to consider the presence of the *repulsive core* of the potential for short distances between the nucleons, or a decrease of the depth of the potential (becoming more shallow) in correspondence with an increase of the kinetic energy of the nucleons. However, these assumptions are too simple to hold in the complicated framework of the description of the nuclear interaction, and a more detailed explanation is needed. The always increasing (in quality and in energy) available amount of experimental data led the research towards a more complex description of the saturation phenomenon. Nowadays, the structure of the nucleons is described by using quarks and gluons, which may in principle account for the repulsive and attractive behavior of the potential, for respectively short and range-like distances, in analogy to the chemical and intermolecular forces present between molecules forming an ordinary liquid. Anyway, the full understanding of the behavior of nuclear forces originating from Quantum Chromo-dynamics is still an open (and intriguing) theoretical challenge [39].

One of the most followed theories regarding the saturation character of the nuclear forces was proposed in the '30s by Heisenberg, with the original suggestion of the concept of *exchange forces*, taking place between nucleons: in this framework, a neutron and a proton were considered to bond through the exchange of an electrical charge, with the proton transforming into a neutron and the neutron becoming a proton. Of course this is just a simplified idea, which does not really justify, e.g., the interaction between two protons (a simultaneous double exchange is rather unlikely to happen), but it was used at that time (and it was the first serious attempt made) to justify the saturation of nuclear forces in a way similar to the *valence saturation* seen in molecular chemistry and driven from an *exchange* process, as discussed in Section 1.1.4.

Finally, one of most wonderful proof of the existence of saturation phenomena of the nuclear force is the α particle, with its exceptional stability and the keystone role it plays in the determination of the structure and properties of a sizable part of the nuclide chart. These properties will be discussed in the following Sections.

1.2.2.1 Saturation of the α particle

To discuss about the saturation of the nuclear interaction, it is important to start from the beginning: the behavior of the binding energy of some nuclei as a function of the increasing number of bonds between the nucleons inside their structure. Let's analyze, following the suggestions by Wigner reported in Figure 1.26, the situations of the ^2H , ^3H , ^3He and ^4He nuclei, which respectively form 1, 3, 3 and 6 bonds. The simplest way to analyze the trend of the binding energies would be to treat it in the fashion of a chemical bond model. The Figure reports, in the various columns along the type of nuclei, the total binding energy, the scheme and number of bonds, and the binding energy per bond². The first, crucial, information which comes to the eye is the fact that the binding energy per bond is anything but constant; as a consequence, this makes a simple chemical-bond model not adapt, at a first sight, for the description of this phenomenon. Nevertheless, it will be shown how the analogy with the chemical model is

²It is assumed, as a first and rough approximation, that the bonds are independent and not influencing each other.

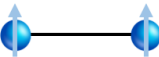
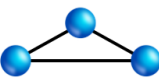
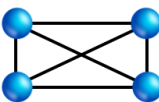
Nucleus	Binding Energy (MeV)	Bond Diagram	N. of bonds	Energy per bond (MeV)
${}^2\text{H}$	2.2		1	2.2
${}^3\text{H}, {}^3\text{He}$	8.48, 7.72		3	≈ 2.7
${}^4\text{He} (\alpha)$	28		6	4.7

FIGURE 1.26: Table showing the binding energy, the bond diagram and the energy per bond for different nuclei. Adapted from [37]. The arrows on the nucleons in the deuteron case indicate that the bound state is a spin *triplet*.

still useful to understand the concept of saturation and other peculiarities of the nuclear force within the effect of many-body forces.

Let's thus extend the concepts of binding processes, previously presented for the lightest nuclei, to those with mass larger than the α particle. Through the application of the simple bond framework, one could expect that the binding energy of a nucleus could increase proportionally to the number of bonds: given that, for a nucleon, the number of bonds is equal to $A(A-1)/2$, it would be reasonable to expect the binding energy per nucleon to be proportional to A for large atomic numbers, with a subsequent reduction of the nuclear volume (see also the discussion of Section 1.2.2). Of course, these conclusions are off the mark, and not in agreement with experimental data; the binding energy per nucleon is not an ever-crescent function, but rather a smooth curve with a maximum in proximity of the ${}^{56}\text{Fe}$ nucleus. For nuclei with Z higher than ≈ 15 , it is possible to consider the whole curve to be almost constant, fluctuating around an energy of $\approx 8 \text{ MeV/A}$. Exceptions to this property are the high rise for $3 < Z < 10$, and the slow increase and decrease respectively for approximately $Z < 30$ and $Z > 30$; it will be clear to see how the former can be imputed to the features described in Figure 1.26.

It is not difficult to imagine an explanation for both of the smooth deviations before and after $Z \approx 30$. The rise at the early part ($Z \simeq 10 - 30$) could be ascribed to the higher weight that surface effects have in the determination of the binding energy for lighter nuclei. In fact, a nucleon on the surface of the nucleus is surrounded by a lower number of neighbors with respect to those placed in the bulk: the contribution of the surface nucleons to the binding energy is thus *lower* than that of the bulk ones. Given that the surface to volume ratio decreases with increasing volumes, the surface effect is more prominent for lighter nuclei, which will in turn have lower binding energies.

On the other hand, the smooth decrease of the binding energy curve for higher mass nuclei can be considered as a consequence of the Coulomb repulsion between protons. It is known that the Coulomb energy (per proton) is proportional to the number of other protons inside the nucleus ($Z-1$). If the specific nuclear binding energy is actually independent from the atomic number, the Coulomb repulsion acting between all the

protons is instead directly proportional to it; having it the form of a repulsion, this will produce a *decrease* in the overall binding energy, increasing for higher Z .

Nevertheless, a priceless information which can be drawn from the curve of the binding energy per nucleon is that the constant value on which the curve is approximately stabilized beyond $Z \approx 10$, around 8 MeV/A, is practically equal to that of the α particle. This means that the average number of bonds formed by a given nucleon, even in heavier nuclei, is the same as in an α particle (i.e., ≈ 3), unveiling one of the main properties of *saturation* phenomena of the nuclear force: each nucleon effectively interacts, on the average, with just *three* neighboring nucleons. This property is similar to what presented in the previous Sections for chemistry, e.g., for the binding processes in liquids and solids and the polar chemical bonds, governed by the concept of the saturation of the chemical valence; in both cases the binding energy appears proportional to the *mass* of the material.

Without diving into the details of the field theory and of the internal structure of the nucleons, a model which will allow to qualitatively describe the interaction conditions and the saturation properties is the so-called *independent particle model*, where each given nucleon is considered as moving in the mean field generated by all the other nucleons. In few words, the wave function of the whole nucleus will be treated as an anti-symmetrized product of the various one-particle functions, assuming that each one-particle state only involves one orbital and spin state. This is, of course, a very simplifying assumption of this model, but it will be useful to the formulation of the saturation and other properties of atomic nuclei. The most important characteristic of the nuclear force to keep in mind when describing the saturation property is that the *nucleons are strongly attracted to each other only if they are in the same orbital state*; the high value of binding energy for the α particle is thus an indication of its saturated state. The special character of the α particle is then linked to the presence of the *Pauli exclusion principle*: only two neutrons and two protons can be found in the same orbital state at a given time, meaning that a strong attraction can occur between a maximum of *four nucleons* at a time, as for the α particle case.

The exclusion principle gives yet another argument for the exceptional stability of the α particle: in fact, ${}^4\text{He}$ is the only existing stable nucleus with $A = 4$, while ${}^4\text{H}$ and ${}^4\text{Li}$ do not exist, and their half-life is of the order of some tens of *yoctoseconds* ($\approx 10^{-23}$ s). These systems, respectively composed by one proton and three neutrons and three protons and one neutron, can not accommodate all the four nucleons in the same spatial state: only three out of four nucleons are found in the minimum energy state feeling a strong mutual attraction, while the fourth spare nucleon is just unbound to the others.

The next goal is to present the structure of the lightest nuclei, which are the most influenced by the aforementioned properties of saturation and nucleon symmetry of the nuclear force; their structure and properties are reviewed in the following, with particular attention given from the point of view of α clustering phenomena in comparison with the single particle model.

1.2.2.2 Structure of the lightest nuclei

In Nuclide Charts, in the same fashion as in Periodic Tables, the various nuclides are grouped on the basis of their number of neutrons (abscissa) and of protons (ordinate):

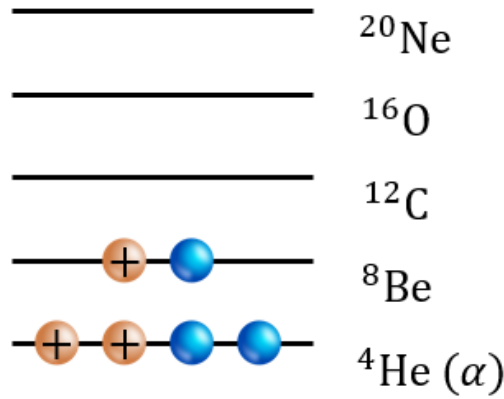


FIGURE 1.27: Schematic diagram (for the lighter nuclei) of one-particle orbital levels in nuclei; protons are indicated with the + symbols, other particles are neutrons. Each level is marked with the corresponding nucleus obtained when it is filled to saturation.

The figure reports the scheme obtained for the ${}^6\text{Li}$ nucleus.

in its early part, the stable nuclides are usually those lying along the bisector, i.e., the $N = Z$ line. Other regularities, which can often be interestingly linked to some nuclear properties, are also found in the distribution of the nuclides alongside the chart. A useful starting basis for the analysis of this problem may be again represented by using a Fermi-gas independent particle model. A sketch of a sequence of single-particle energy levels suggested by this model is reported in Figure 1.27. The energy of a nucleus depends on the number of nucleons in the top level and in the levels below. Given the distinction between protons and neutrons, as an obvious consequence of the Pauli principle, each level can be occupied by four nucleons at most. When studying the *ground states* of the various nuclei, it is implicitly assumed that all the levels beneath the last one are completely filled, i.e., no nucleon has been excited to an higher level and no holes are present in the inner orbital states. For example, Figure 1.27 shows the diagram which is obtained in the case of the ${}^6\text{Li}$ nucleus.

The first part of the nuclide chart, going from ${}^6\text{Li}$ to ${}^{17}\text{O}$, shows some greatly regular behaviors. ${}^6\text{Li}$, produced by the addition of a n-p couple to the α particle core, is a stable nucleus, while its *isobaric* cousins, ${}^6\text{He}$ and ${}^6\text{Be}$, are respectively unstable against β -emission and completely unstable. The instability of ${}^6\text{Be}$ is easily ascribable to the Coulomb repulsion existing between the two excess protons, but it is interesting to dive into the reasons for the differences between the stability of ${}^6\text{Li}$ and ${}^6\text{He}$: if the nuclear force is charge-independent, why does the addition of a n-p couple make a nucleus more stable than that obtained after the addition of two neutrons? Would it mean that the n-p interaction is stronger than the n-n one? Actually, as already observed in Section 1.2.1.2, the reason is to be looked for in the *spin-dependence* of the nuclear interaction.

It is important to remember that, due to the Pauli exclusion principle, two protons (or two neutrons) must have opposite spins if they are in the same orbital state; two different nucleons, like a proton and a neutron, may instead have parallel spins, and occupy a *triplet* state. Given that nucleons in triplet states are more bound than those in singlet states, as shown in the cases of deuteron and nucleon-nucleon scattering, a n-p pair in a half-occupied orbital state will produce a stronger attraction with respect to those existing between n-n and n-p pairs, explaining the ${}^6\text{Li}$ stability.

After ${}^6\text{Li}$, the addition of a neutron would produce ${}^7\text{Li}$ (the most abundant stable Li-isotope), while the addition of a proton makes ${}^7\text{Be}$, a relatively stable but radioactive isotope (with an half-life of 54 days); this relative stability is due to the fact that the Coulomb energy difference in this nucleus is higher than the n-H mass difference. On the other hand, nuclei like ${}^5\text{Li}$ or ${}^5\text{He}$ are extremely short lived: this is a direct consequence of the fact that the attraction felt by the 5th nucleon, either a proton or a neutron, towards the α core (${}^4\text{He}$) is too weak to hold the corresponding nucleus together.

Taking one step further and adding another proton to ${}^7\text{Li}$ to form ${}^8\text{Be}$, the high instability of this nucleus would of course appear surprising and contradictory, but it has to be imputed to the great stability of the two constituent α particles, in which the nucleus is spontaneously divided within a process quite similar to *fission*, driven by the competition of surface to Coulomb effects in presence of a strongly deformed shape of the nucleus. On the other hand, the addition of another neutron to ${}^8\text{Be}$ starts to fill the third level of Figure 1.27, and gives rise to the formation of the stable ${}^9\text{Be}$ nucleus. This is a proof of the presence of a "residual" attraction between nucleons in different states, i.e., those in the two α particles and the so-called "covalent" neutron, binding them together. It is worth noting the *weak* nature of the binding of ${}^9\text{Be}$: the one-neutron separation energy is just $S_n = 1.66$ MeV.

The analysis of the remaining nuclei up to ${}^{17}\text{O}$ gives the further information that there are no stable *odd-Z-odd-N* nucleus beyond ${}^{14}\text{N}$.

1.2.2.3 The need for nuclear models

The inherent complexity of the nuclear force and potential calls for the need of several different nuclear models, each with its peculiarities, suitable for the description of all the various facets of this not-easily understood interaction. The main complication is given by the fact that the nuclei are, of course, complex many-body systems, composed by protons and neutrons, in turn made up of *quarks* and *gluons*; consequently, an approach based on Quantum Chromodynamics can not currently be sought, due to the extremely divergent complexity of the problem.

For this reasons, starting from the discover of the atomic nucleus, several models have been developed to describe it as a system directly based on nucleons. In this framework, a *model* is an ensemble of simplifying mathematical or physical hypotheses, with whom it is possible to calculate the characteristics of nuclear systems within a certain grade of approximation. Given the non-additivity of the nuclear interaction, these also have to account for the increased complexity given by the extra-interactions between three or more bodies, as in the fashion of an *ab-initio* approach. Some two- and three-nucleon potentials (see, e.g, Refs. [40, 41]) achieved a reasonable approximated description of the structure of some classes of light nuclei, but even in this case, some important aspects are still poorly known (as, e.g., clustering properties of self-conjugate nuclei, structure of highly-lying excited states, etc.). Since a general and comprehensive theory for the description of all the nuclei is still missing, nuclear models are still fundamental to reproduce each some specific sets of properties, under the respective assumptions and simplifications. Like this, the different pieces of the puzzle aiming to describe the complex nuclear physics scenario are formed.

The first atomic and nuclear models have been developed at the beginning of the last century: two of these pioneer models are due to Nagaoka (*electron rings model*) and J.

J. Thomson (*plum pudding* or *charged cloud model*) [42, 43]. Each subsequent model was developed to contradict, or to better address, the problems and inefficiencies of the previous models, like Ernst Rutherford did in 1911 [44], proposing a model in which the positive charge of an atom is concentrated in a very *small* and *central* portion of its space ($\varnothing_{\text{nucleus}} \approx 10^{-14}$ cm), allowing to lay the foundations for modern nuclear and atomic physics.

One of the first models which was actually built for the specific description of the properties of nuclei [45] originates on the basis of Niels Bohr's concept of a compound nucleus reaction: similarly to transition states in chemical reactions, the projectile particle is captured by the target nucleus to form a *compound* transient system, rapidly sharing its energy with the nucleons of the target nucleus. Alongside the saturation behavior of the specific interaction, characteristics like low compressibility and well defined surface effects evidence the analogy between nuclear matter and liquids, making (e.g.) a *liquid drop model* useful to describe several properties of nuclear systems (Carl Friedrich von Weizsacker, 1935). In fact, the *nuclear matter* is usually considered as composed by nucleons behaving as free particles, like molecules in condensed phases of ordinary matter. This model, which approximates nuclei to charged liquid drops, is particularly adapt for quantitative studies of nuclear surface energy, nuclear masses and decays following nuclear reactions. However, when trying to predict the nuclear masses of the various nuclides through the liquid drop model prescriptions, several periodical and systematic deviations are observed, reflecting the so-called *shell-closure* effects (happening in correspondence of particular "*magic*" nucleon numbers), representing conditions of extraordinary nuclear stability, where the corresponding nuclei are characterized by masses well lower than those predicted by the model. These limitations can be overcome through the application of other models, like the independent particle model presented before and the *cluster model*, which will be the main actor under the spotlight of the remaining part of this first Chapter.

An overview of the most common models exploited in modern nuclear physics is listed in Table 1.3 and graphically sketched in Figure 1.28; in these frameworks, the models are divided in the two classes of *Strong Interaction Models* (SIM) and *Independent Particle Models* (IPM). Given the partial contradictory natures of these models, *Unified* or *Collective* theories have been introduced to try to merge these two approaches and take care of these discrepancies, and are often preferred for the description of modern experimental data. The main differences lay in the way in which the nucleus and its nucleons are originally described: in SIMs, the nucleus is treated as a structure of strongly coupled nucleons, while IPMs depict each nucleon as quasi-independent particles moving in the mean field generated through the average action of all the others.

Strong Models	Interaction	Fundamental hypothesis	Field of application
<i>Liquid drop</i>		The nucleus is assimilated to a liquid drop, where the nucleons behave similarly to molecules, and surface effects play an important role. Suitable for very large nuclei.	Calculation of binding energies, predictions of vibrational levels and fission processes.

Strong Interaction Models	Fundamental hypothesis	Field of application
<i>Collective hydrodynamics</i>	The nucleus is treated as a continuous fluid of nucleons, subject to hydrodynamical laws, to describe its collective motions, such as vibrations and rotations	Description of Giant resonances at low-to-medium energies and of collective flows of matter in relativistic collisions.
<i>Compound nucleus</i>	The compound nucleus, formed as a transient state in a nuclear reaction, has no memory of the way it is formed (entrance channel): the exit channel of the reaction gives spectroscopic and structural information of the transient state.	Measurements and predictions of the cross sections (σ) of the various reactions and analysis of nuclear resonances. Often coupled with the Optical Model.
<i>α particles or clusters</i>	α particles are considered as the fundamental units building the nuclei, with strong forces between their four inner nucleons and weak interactions between different α .	Description of α radioactivity of heavy nuclei. Structure of ground and excited states of self-conjugate nuclei (even-even like ^8Be , ^{12}C , ^{16}O). Molecular clustering in non self-conjugate nuclei.

Independent Particle Models	Fundamental hypothesis	Field of application
<i>Potential Well</i>	The nucleus is described through the use of a potential well, with defined range and depth.	Elastic scattering cross section of neutrons on heavy nuclei, pioneer version of the Optical Model.
<i>Fermi Gas</i>	Nuclei are seen like gases of non-interacting fermions, totally degenerated, where the eigen-functions of single nucleons are plane waves.	Behavior of nuclear matter. Description of thermodynamical properties observed in intermediate-energy heavy ion collisions. Multi-fragmentation phenomena.

Independent Particle Models	Fundamental hypothesis	Field of application
<i>Optical</i>	The nucleus is treated through a <i>complex</i> potential well, where the real part and the imaginary one respectively account for the scattering and absorption phenomena.	Elastic scattering angular distributions and absorption cross sections; it was often used to complement compound nucleus predictions. Improved versions of optical models considered the possibility of coupling of several reaction channels (<i>Coupled Channels</i> approximation).
<i>Shell</i>	The nucleons move like quasi-independent particles inside a spherical potential well (with suitable spin-orbit terms), following the experimental nuclear density distribution.	Levels generated by small single-nucleon excitations, properties of nuclear ground states as a function of the atomic number A , explaining the existence of magic numbers.
<i>Shell and single particle</i>	Suitable for nuclei with <i>odd</i> A , where the spare nucleon (usually the first one after a shell closure) strongly influences the properties of the nucleus.	Possibility to study the single-particle levels in nuclei with a spare nucleon external to full shells.
<i>Shell and many particles</i>	As the above, but with more than one spare nucleons, taking the interactions between the external and weakly bound nucleons into account.	Multi-particle levels for nuclei with more than one spare nucleon. Configuration mixing.
<i>Deformed shell model</i> (Nilsson)	A particular shell model built with a potential well showing a quadrupolar deformation (prolate or oblate). Spin-orbit coupling is still present.	Structure of ground state and low energy excited states of deformed nuclei.

Unified Models	Fundamental hypothesis	Field of application
<i>Unified</i>	Nucleons are almost independent, in a common, non-spherical and slowly variable potential well.	Study of levels due to the excitation of both single particles or collective rotational and vibrational modes (like the following models and the <i>liquid drop</i> one).
<i>Collective</i>	Specific case of the <i>Unified model</i> , which only takes the collective motions into account.	Collective vibrational and rotational levels.
<i>Spheroidal core</i>	Similar to the preceding, where the core deformation is due to the external nucleon cloud.	Rotational levels of deformed nuclei and collective excitations.

TABLE 1.3: Table representing the various nuclear models, their fundamental hypotheses and the main fields of application. Adapted from [28].

The main point is that there is no model which is completely right or completely off the mark. For example, the shell model is a refinement of the Fermi gas model and this is, in turn, completely opposed to the Liquid Drop model; nevertheless, each one was capable to justify and explain some particular nuclear properties, which the other models could not. Moreover, the Optical model is excellent for the description of elastic scattering cross section at medium-high energies, but not really suited to determine the cross section of resonant processes or to describe the shape of the emitted particle spectra, for which compound nucleus approaches are much more suitable; in this way, these two models complement each other in the description of nuclear reactions.

In the same fashion, signatures of clustering emerge naturally when considering advanced versions of the shell model, in the form of long range correlations between the nucleons in some peculiar investigated systems, like ^{19}F ; oddities such as the *cluster joke* related to this nucleus will be presented in following discussions. On the other hand, the spatial distribution and localization of the various clusters, a crucial aspect of the clustered description of nuclei, is hardly achievable within the framework of the shell model. For this reason, the next Sections are completely dedicated to the presentation and discussion of the so-called *nuclear cluster model*, especially from the point of view of the formation and organization of α -like clusters inside the nuclear volume, and how these influence the properties of self-conjugate nuclei *in primis*.

It would be now interesting to make a brief example on the usefulness of models for the computation of some quantities needed in the framework of nuclear physics studies. If one exploits the wave functions of a shell model with a simple harmonic oscillator potential, without any further correction, it is possible to determine, as a beautiful exercise, the density of the nuclear matter in the ground state of a self-conjugate nucleus, like ^{12}C . This may be compared with results coming from electron scattering experimental data (or, also, ^{12}C -hadron scattering on an optical model basis). In this framework,

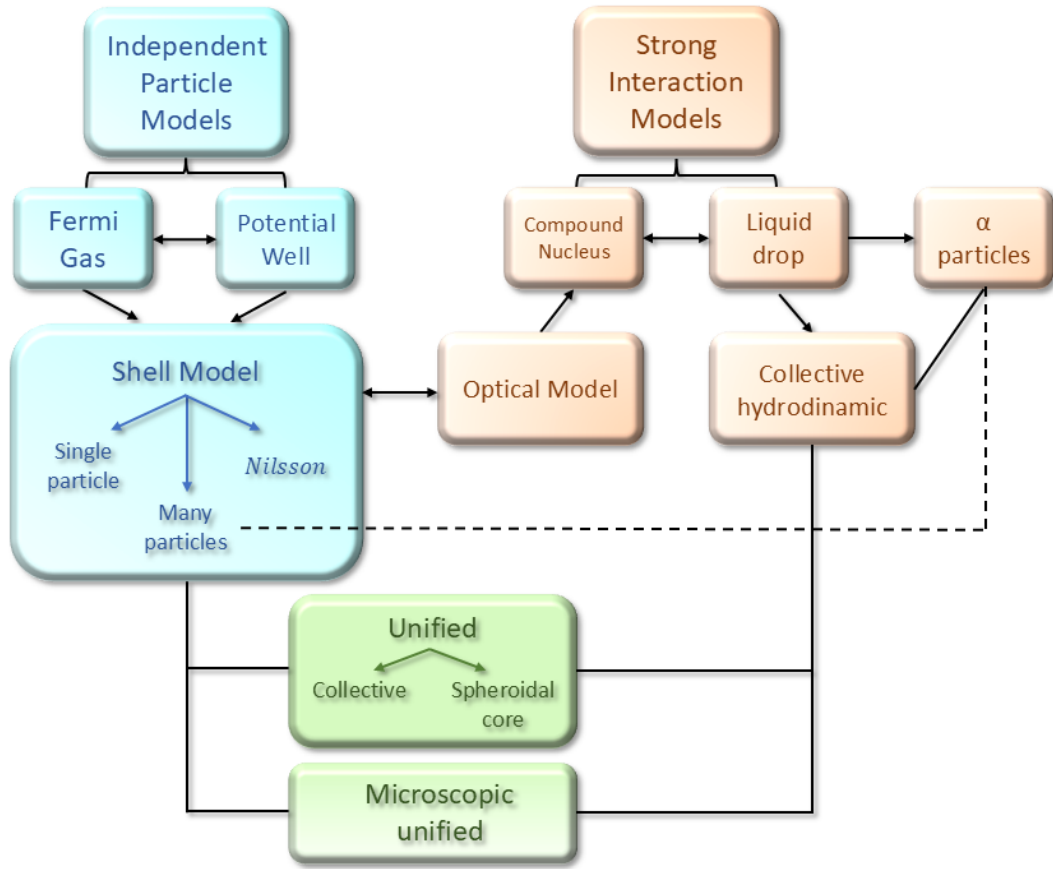


FIGURE 1.28: Conceptual map presenting the connections between the nuclear models listed in Table 1.3, divided in Independent Particle Models, Strong Interaction Models and Unified Models. Adapted from [28].

the insurgence of eventual discrepancies may underline important deviations with respect to the classically assumed spherical or spheroidal shapes due to the occurrence of clustering.

In the framework of a mean-field model casting the nucleus with a potential having an harmonic oscillator shape, the wavefunctions of the $1s_{1/2}$ and $1p_{3/2}$, $1p_{1/2}$ orbits can be written as follows [46, 47]:

$$\psi_{(1s_{1/2})} = \left(\frac{1}{\pi a^2} \right)^{3/4} \exp \left(-\frac{r^2}{2a^2} \right) \chi_{\pm} \quad (1.26)$$

$$\psi_{(1p_{3/2})} = 2 \left(\frac{4}{9\pi a^{10}} \right)^{1/4} r \exp \left(-\frac{r^2}{2a^2} \right) \begin{cases} \chi_+ Y_{1,1} & \text{if } m_j = 3/2 \\ \sqrt{\frac{2}{3}} \chi_+ Y_{1,0} + \sqrt{\frac{1}{3}} \chi_- Y_{1,1} & \text{if } m = 1/2 \\ \sqrt{\frac{1}{3}} \chi_+ Y_{1,-1} + \sqrt{\frac{2}{3}} \chi_- Y_{1,0} & \text{if } m = -1/2 \\ \chi_- Y_{1,-1} & \text{if } m_j = -3/2 \end{cases} \quad (1.27)$$

where the terms up to the exponential represent the spatial part of the wavefunction, while the χ terms are related to the *spin* contributions, and the $Y_{\ell,m}$ terms are the spherical harmonics functions. Taking into account the spin-orbit coupling, the degeneracy in

the $1p$ level is removed, and it splits into the $1p_{3/2}$ (Eq. 1.27) and $1p_{1/2}$ wavefunctions, with a similar form (see Ref. [47] for more details). The matter density in the nuclear system can be calculated as

$$\rho(\vec{r}) = \sum_{k=1}^A |\psi_k(\vec{r})|^2 \quad (1.28)$$

an equation valid in the approximation of *independent* nucleons, i.e., without any type of mutual correlations. Obviously, a further requirement is for the integral of the density on the whole volume to be equal to the atomic number of the nucleus ($\int \rho(\vec{r}) d\tau = A$). Before diving into the more complicated case of the ^{12}C nucleus, it would be good to understand the density calculation for one of its three supposed constituents, i.e., the α particle, the ^4He nucleus. In this case, Equation 1.29 takes the form

$$\rho(\vec{r}) = \sum_{k=1}^4 |\psi_k(\vec{r})|^2 \quad (1.29)$$

but all the four nucleons in the ^4He nucleus occupy the $1s_{1/2}$ orbit, and will thus have the same wave function reported in Eq. 1.26; the density will be

$$\rho(r) = 4 \cdot \left| \left(\frac{1}{\pi a^2} \right)^{3/4} \exp \left(-\frac{r^2}{2a^2} \right) \right|^2 \quad (1.30)$$

which is four times the same spatial wavefunction of one single nucleon, squared. This is

$$\rho(r) = 4 \cdot \left(\frac{1}{\pi a^2} \right)^{3/2} \exp \left(-\frac{r^2}{a^2} \right) \quad (1.31)$$

The calculation is of course way more complicated in the case of the ^{12}C , but its development is worth a shot for the sake of completeness. The ^{12}C nucleus is composed by 6 protons and 6 neutrons: in the harmonic oscillator orbital schemes (with spin orbit coupling), two particles (protons and neutrons) occupy the $1s_{1/2}$ level, while the other four lie in the $1p_{3/2}$, with different spin values ($+3/2, +1/2, -1/2, -3/2$). The density distribution will thus be composed of different parts:

- The density distribution of the ^4He nucleus, $\rho_{^4\text{He}}$
- A common part related to the four spatial functions, all equal in the $1p_{3/2}$ orbit, took 2 times to account for both protons and neutrons
- Everything related to the spin and spherical harmonics functions.

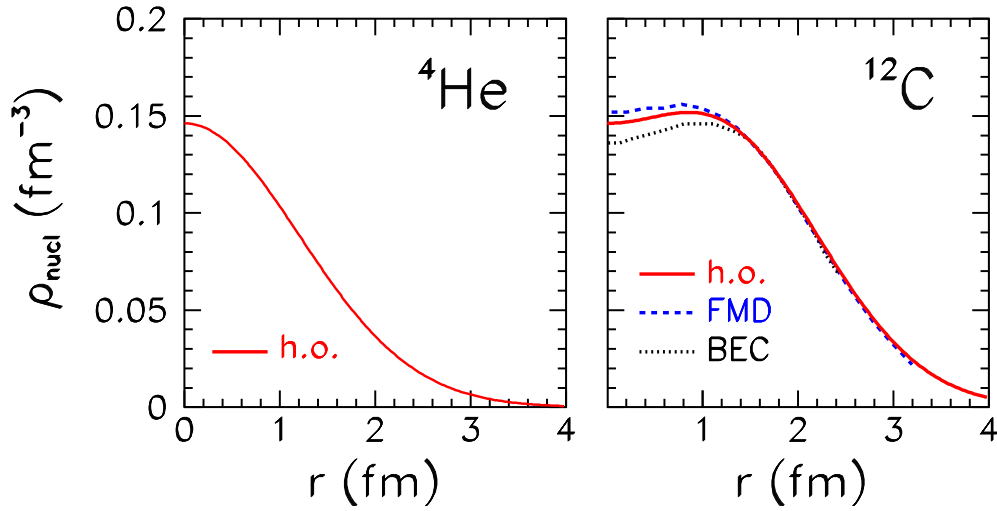


FIGURE 1.29: *Left panel:* Plot of the nuclear density distribution of the ${}^4\text{He}$ nucleus as a function of the distance r from the center, in the framework of the harmonic oscillator model of the nucleus (red solid line). In Equation 1.31, it was assumed $a = 1.7$ fm to obtain a central nuclear density of $\rho \simeq 0.146 \text{ fm}^{-3}$. *Right panel:* The same for the ${}^{12}\text{C}$ nucleus in its ground state, as obtained from Equation 1.39 (red solid line). For comparison, the predictions of *charge* density distributions obtained by analyzing electron scattering data within the Fermionic Molecular Dynamics (FMD, blue dashed line) and the Bose-Einstein Condensate (BEC, black dotted line) models are reported, simply multiplied by a factor of 2 to take into account the neutron-proton symmetry of this nucleus. FMD and BEC data are taken from Ref. [48].

The density distribution appears, therefore, as follows:

$$\begin{aligned} \rho(r) = \rho_{4\text{He}} + 2 \cdot & \left[4 \left(\frac{4}{9\pi a^{10}} \right)^{1/2} r^2 \exp \left(-\frac{r^2}{a^2} \right) \right] \\ & \cdot \left[\chi_+^2 Y_{1,1}^2 + \left(\frac{2}{3} Y_{1,0}^2 \chi_+^2 + \frac{1}{3} Y_{1,1}^2 \chi_-^2 + \frac{2\sqrt{2}}{3} Y_{1,0} Y_{1,1} \chi_+ \chi_- \right) \right. \\ & \quad \left. + \left(\frac{1}{3} Y_{1,-1}^2 \chi_+^2 + \frac{2}{3} Y_{1,0}^2 \chi_-^2 + \frac{2\sqrt{2}}{3} Y_{1,-1} Y_{1,0} \chi_+ \chi_- \right) \right. \\ & \quad \left. + \chi_-^2 Y_{1,-1}^2 \right] \quad (1.32) \end{aligned}$$

Some features allow to simplify the expression: due to their orthogonality, all the various products between the χ terms are equal to 1 or 0; it follows that:

$$\begin{aligned} \rho(r) = \rho_{4\text{He}} + 8 \cdot & \frac{2}{3} \left(\frac{1}{\pi a^{10}} \right)^{1/2} r^2 \exp \left(-\frac{r^2}{a^2} \right) \\ & \cdot \left(Y_{1,1}^2 + \frac{2}{3} Y_{1,0}^2 + \frac{1}{3} Y_{1,1}^2 + \frac{1}{3} Y_{1,-1}^2 + \frac{2}{3} Y_{1,0}^2 + Y_{1,-1}^2 \right) \quad (1.33) \end{aligned}$$

All the similar terms can be added to obtain

$$\begin{aligned}\rho(r) &= \rho_{\text{He}} + 8 \cdot \frac{2}{3} \frac{1}{\sqrt{\pi}a^5} r^2 \exp\left(-\frac{r^2}{a^2}\right) \cdot \left(\frac{4}{3}Y_{1,1}^2 + \frac{4}{3}Y_{1,0}^2 + \frac{4}{3}Y_{1,-1}^2\right) \\ &= \rho_{\text{He}} + 8 \cdot \frac{2}{3} \cdot \frac{4}{3} \frac{1}{\sqrt{\pi}a^5} r^2 \exp\left(-\frac{r^2}{a^2}\right) \cdot (Y_{1,1}^2 + Y_{1,0}^2 + Y_{1,-1}^2) \quad (1.34)\end{aligned}$$

One can now explicit the form of the various spherical harmonics

$$Y_{0,0} = \frac{1}{\sqrt{4\pi}}; \quad Y_{1,-1} = \sqrt{\frac{3}{8\pi}} e^{-i\phi} \sin \theta \quad (1.35)$$

$$Y_{1,-1} = \sqrt{\frac{3}{4\pi}} \cos \theta; \quad Y_{1,1} = \sqrt{-\frac{3}{8\pi}} e^{-i\phi} \sin \theta \quad (1.36)$$

bearing in mind that $|e^{i\phi}| = |e^{-i\phi}| = 1$. Then, the term with the addition of the spherical harmonics, inside the last parenthesis in Eq. 1.34 becomes:

$$Y_{1,1}^2 + Y_{1,0}^2 + Y_{1,-1}^2 = \frac{3}{8\pi} \sin^2 \theta + \frac{3}{4\pi} \cos^2 \theta + \frac{3}{8\pi} \sin^2 \theta = \frac{3}{4\pi} \sin^2 \theta + \frac{3}{4\pi} \cos^2 \theta = \frac{3}{4\pi} \quad (1.37)$$

If one inserts this result in Equation 1.34

$$\begin{aligned}\rho(r) &= \rho_{\text{He}} + \frac{8 \cdot 2 \cdot 4}{9} \cdot \frac{1}{\sqrt{\pi}a^5} r^2 \exp\left(-\frac{r^2}{a^2}\right) \cdot \frac{3}{4\pi} = \\ &= \rho_{\text{He}} + \frac{16}{3} \cdot \frac{1}{\pi^{3/2}a^5} r^2 \exp\left(-\frac{r^2}{a^2}\right) = \rho_{\text{He}} + 4 \cdot \frac{4}{3} \cdot \frac{1}{\pi^{3/2}a^3} \frac{r^2}{a^2} \exp\left(-\frac{r^2}{a^2}\right) = \\ &= 4 \left(\frac{1}{\pi a^2}\right)^{3/2} \exp\left(-\frac{r^2}{a^2}\right) + 4 \left(\frac{1}{\pi a^2}\right)^{3/2} \frac{4}{3} \frac{r^2}{a^2} \exp\left(-\frac{r^2}{a^2}\right) \quad (1.38)\end{aligned}$$

and finally:

$$\rho(r) = 4 \cdot \left(\frac{1}{\pi a^2}\right)^{3/2} \exp\left(-\frac{r^2}{a^2}\right) \left(1 + \frac{4}{3} \frac{r^2}{a^2}\right) \quad (1.39)$$

Furthermore, from the value of ρ_0 in correspondence of $r = 0$ (the *central* density, being $\simeq 0.146 \text{ fm}^{-3}$ for medium mass nuclei [31]), it is possible to derive the value of the parameter a :

$$\rho(r=0) = 4(\pi a^2)^{-3/2} \rightarrow \frac{0.146}{\text{fm}^3} = \frac{4}{(\pi a^2)^{3/2}} = \frac{4}{\pi^{3/2}} \cdot \frac{1}{a^3} \quad (1.40)$$

$$a = \left(\frac{4}{\pi^{3/2} \cdot 0.146} \text{fm}^3\right)^{1/3} \rightarrow a \simeq 1.7 \text{ fm} \quad (1.41)$$

Figure 1.29 reports the trend of the obtained nuclear densities for the ^4He and ^{12}C nuclei in their ground states as derived from the harmonic oscillator potential. Even if this model is very simplified, it is a useful tool to have an idea of the distribution of nuclear matter in self-conjugate nuclei. As an example, the present harmonic oscillator calculations (Equations 1.30 and 1.39) are compared with more refined predictions based on the Fermionic Molecular Dynamics and Bose-Einstein Condensate models, aiming at the description of the charge form factors derived from electron scattering experiments [48] (the *charge* density distributions reported in the literature were multiplied by a

factor of 2 to obtain the *nuclear* density distributions). The presence of clustering phenomena leads to a peculiar behavior of the nuclear matter distribution especially in the central region of the nucleus; for example, within the BEC model, it is possible to observe a sizable depletion of nuclear matter in such region. The extraction of $\rho(r)$ from experimental data of scattering and/or reaction processes is therefore a precious tool to unveil the structure of light nuclei.

1.2.3 α cluster structures in atomic nuclei

...As bees
in spring-time, when the Sun with Taurus rides,
pour forth their populous youth about the hive
in clusters; they among fresh dews and flowers
fly to and fro...
J. MILTON, *Paradise Lost*, Book 1, 768-772

The main aim of α cluster models of nuclei is to describe the clustering phenomena through the study of peculiar, and often symmetric, distributions of these particles between the boundaries described by the physical dimensions of the nucleus. One important thing to bear in mind is that the α -clusters inside the nuclei are not exactly like the common α particles (i.e. the ${}^4\text{He}$ nuclei), but rather *transient* structures. In the earliest works about this matter, the α clusters were considered as bodies interacting with the same van der Waals-like features as molecules in ordinary liquids [49]; most importantly, the basic assumption was that the forces between the α particles were additive. However, more detailed investigations showed how this can not be the case, unless if not in the presence of a sufficiently strong residual, non-exchange, attraction, with a range longer than the bulk of the force ("long-range" correlations) [49]. This assumption does not hold in general for all nuclei in all their states of excitation energies; therefore, the analogy with the van der Waals interactions must be managed with some care.

Nevertheless, another chemical analogy is still tempting, i.e., a comparison of the interaction between α particles with the chemical binding forces: one is based on the exchange of electrons between atoms, while the other relies on the transfer of nucleons between the α clusters themselves. The α clusters in nuclei are still transient units, but it is possible to legitimately treat them as "permanent", if the duration of the periods of the investigated properties, like rotations or vibrations of the nucleus, is shorter than the life-time of the α clusters. Some simple semi-classical calculations, considering the average transit time of an α particle inside a self-conjugate nucleus and the typical vibration and rotation times of low-lying excited states, confirm the rationality of this assumption [49, 50].

1.2.3.1 $\alpha - \alpha$ scattering and $\alpha - \alpha$ potential

One should certainly begin the discussions on α clustering from the analysis of the empirical evidences on the α particles interaction; these mainly come from experimental $\alpha + {}^4\text{He}$ scattering data obtained by Wheeler [51, 52], and from the structure of ${}^8\text{Be}$. In the cluster framework, the beryllium nucleus could be envisaged as composed by two α particles, analogue to an homopolar diatomic molecule. The ground state of the ${}^8\text{Be}$ nucleus is an unbound state with an energy of ≈ -0.92 MeV, so the scattering experiments

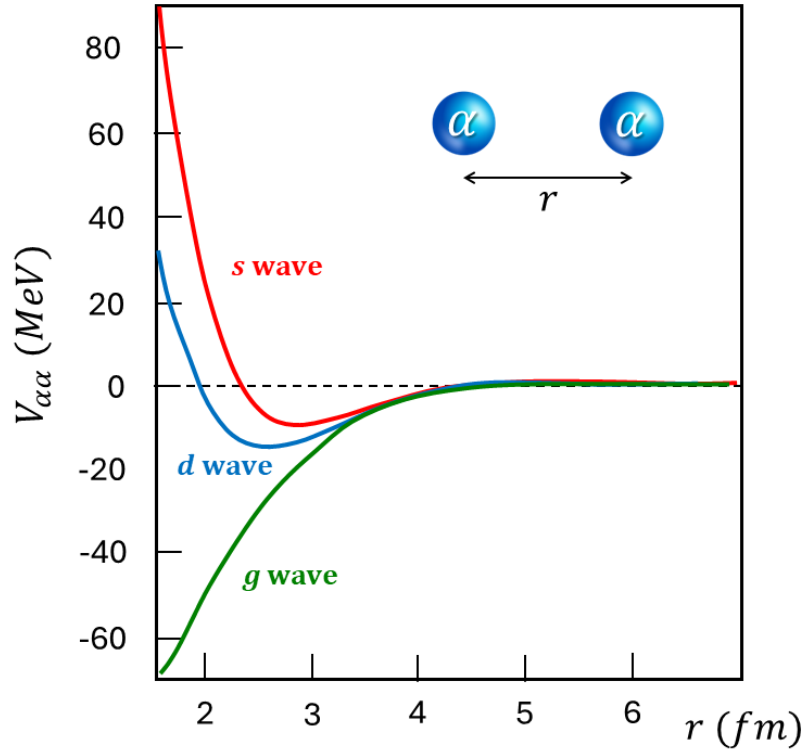


FIGURE 1.30: $\alpha - \alpha$ effective interaction potential derived from phase shift analysis of elastic scattering data by Ali and Bodmer [53]. Different colors indicate the shape of potential for different orbital angular momenta involved in the scattering process. The presence of Coulomb barrier was included in the potential, while the centrifugal barrier was not added to the potential. Note the striking similarity with the Morse potential discussed in molecular chemistry and shown in Figure 1.10

are usually able to study just the excited levels of ^8Be , the first one being about 3 MeV higher in energy. The inclusion of a polarization potential [53], corresponding to the van der Waals forces between molecules, makes the problem very similar to intermolecular interactions seen in chemistry, see Section 1.1.6. The treatment of the $\alpha - \alpha$ scattering data is also simplified by the fact that it deals with identical scattering partners, like in proton-proton collisions, with the advantageous difference that the α particles have no intrinsic angular momentum, high internal binding energy, and are Bosons, subject to the Bose-Einstein statistics; on the basis of this, only the even orbital momentum (ℓ) partial waves can contribute to the scattering cross section (i.e., mainly s, d and g partial waves).

Given the supposed important role of the α particle in the determination of several properties of more complex nuclei, the study of the $\alpha - \alpha$ interaction may thus be a keystone to the understanding of their structure and even on basic nucleon-nucleon forces. In this case, the ratio of the observed cross section to the purely classical Rutherford one can be written as $\frac{d\sigma/d\Omega_{obs}}{d\sigma/d\Omega_{Ruth}} = |\mathcal{R}|^2$, where $\mathcal{R}$ is a complex quantity obtained by the sum of different terms [51]:

$$\mathcal{R} = \mathcal{R}_c(\theta) + \mathcal{R}_c\left(\frac{\pi}{2} - \theta\right) + \sum_{\text{even } \ell} \mathcal{R}_\ell(\theta) \left(1 - e^{2i\delta_\ell}\right) \quad (1.42)$$

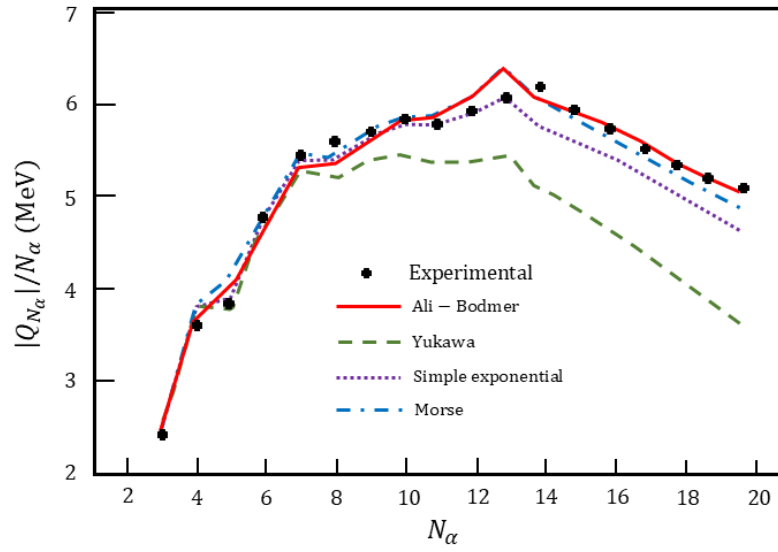


FIGURE 1.31: Decomposition energies into $N\alpha$ particles ($|Q_{N\alpha}|$), normalized to the number of α particles (N_α), as a function of N_α itself for various nuclei up to $A = 80$. Adapted from [56]

where the first two terms represent the pure Coulomb scattering, comprehensive of the mutual interference, while the sum on even ℓ is related to the "anomalous" contributions from the scattering deriving from the effective nuclear interaction between the two α particles, and δ_ℓ is the corresponding phase shift. These phase shifts can be obtained through the fitting of large bodies of low energy α scattering data, as done in the past, using $\ell = 0, 2, 4$ partial waves, enlightening the presence of some low-energy resonances with widths comparable to the single-particle (Wigner) limit (see Section 2.3), as reported in Refs. [53, 54].

Following the prescriptions in Refs. [53, 54], the form of the $\alpha - \alpha$ potential can be drawn through the energy dependence of the obtained phase shifts. The most interesting feature is the similarity between the forms of the real parts of these potentials, plotted against the $\alpha - \alpha$ distance (Figure 1.30), and the interaction potential existing between atoms in chemical frameworks (examples of these potentials have been discussed in Figure 1.10): all of these, albeit with different shapes, show a minimum energy for an ideal interaction distance and a strong repulsive core at short ranges (≈ 1.7 fm). Of course, the real parts with higher ℓ are more similar to the (chemical) molecular potential if the presence of the centrifugal barrier is neglected; on the other hand, when taking it into account, the repulsive part of the potential starts to become dominant and the depth of the potential pocket start to become shallower, leading to a complete disappearance for higher ℓ values (e.g., 6, 8).

The next step would be to use the two-body potentials obtained in this way to deepen the prescriptions of the α -cluster model, as done in Refs. [55, 56]. The ideas there presented are related to the possibility of the description of the decomposition energies into $N\alpha$ particles ($|Q_{N\alpha}|$) of light and medium-mass nuclei through the sum of the interaction energies of the constituent α particles.

In this hypothesis, the α particles, confined within the nuclear volume, are supposed

to occupy configurations of minimized energies, i.e., following quasi-crystalline distributions. As it will be shown several times in this thesis, these shapes range from the equilateral triangle configuration for ^{12}C , to a tetrahedron for ^{16}O , up to more complex forms, like the platonic icosahedron for ^{52}Fe , where a central α particle is surrounded by the other twelve forming the icosahedron. In Figure 1.31, the binding energy of the various nuclei (related to the sum of the various $\alpha - \alpha$ energies) is modeled through the application of different molecular or nuclear physics potentials: the Morse, Yukawa and Ali-Bodmer potentials and a simple exponential one. As it is shown in the Figure 1.31, the Ali-Bodmer and Morse molecular-like potentials are the best suited to describe the trends of the binding energy for the largest part of self-conjugate nuclei.

This is an indication of how crucial the role of the two-body potential is in the description of the $\alpha - \alpha$ scattering and in the framework of α clustering. Furthermore, it enforces the vision by which the α -cluster structure could be successfully treated through minimal geometric considerations and two-body models. Last, but not least, the difficulties encountered by the various potentials in describing experimental data in the region $A \approx 52 - 56$, seems to suggest a reduction of the α -clustering organization, possibly due to the presence of the shell closure for the magic number $Z = 28$, or to the rearrangement of clusters in geometrical shapes less symmetric than the platonic solids adopted in Refs.[55, 56].

However, as long as it would be really nice to treat the $\alpha - \alpha$ interaction as a complete analogue of the van der Waals forces between molecules, some differences in the features of the potential and in the intrinsic characteristics of exchange forces grant the $\alpha - \alpha$ interaction no additivity, in striking contrast with van der Waals-based attractions. This feature will be addressed in the next Section, to introduce the prescriptions for the calculation of binding energies in self-conjugate nuclei via the balance between kinetic and potential energy of the intervening clusters.

1.2.3.2 Cluster disintegration of self-conjugate nuclei

Following the fundamental works by Wefelmeier [57, 58] and Hafstad and Teller [59], it could be actually thought that the observed values of binding energies for the self-conjugate nuclei may be interpreted on the basis of a supposed van der Waals-like additivity of $\alpha - \alpha$ interactions. In this framework, the binding energies of the α -clustered nuclei would be calculated on a direct proportionality with respect to the number of $\alpha - \alpha$ bonds inside the nucleus. An empirical way to calculate the disintegration energy of a nucleus into all the constituent clusters ($E_{dis} = -Q_{N\alpha}$) is to compute the difference between its mass and the sum of those of the constituent α particles ($Q_{N\alpha} = m_{nucleus} - N \cdot m_{\alpha}$): all of the $Q_{N\alpha}$ values are *negative*, except for the ^8Be nucleus, which has $Q_{2\alpha} = +92 \text{ keV}$. In this way, all the lighter α -nuclei show a good proportional agreement between the experimental disintegration energies and the number of bonds, with the only exceptions of ^8Be , which is unbound, and for ^{20}Ne , if one considers all the $\alpha - \alpha$ bonds to be equivalent.

However, as shown in results by Margenau [60], this precise regularity cannot be ascribed to any type of *simple* explanation, but should actually arise from more complicated compensations between energy terms of different order of approximations, derived from the *non-additive* characteristics of the $\alpha - \alpha$ interactions [61]. As an example, in the triangular configuration of ^{12}C , one proton and a neutron from two α centers can overlap

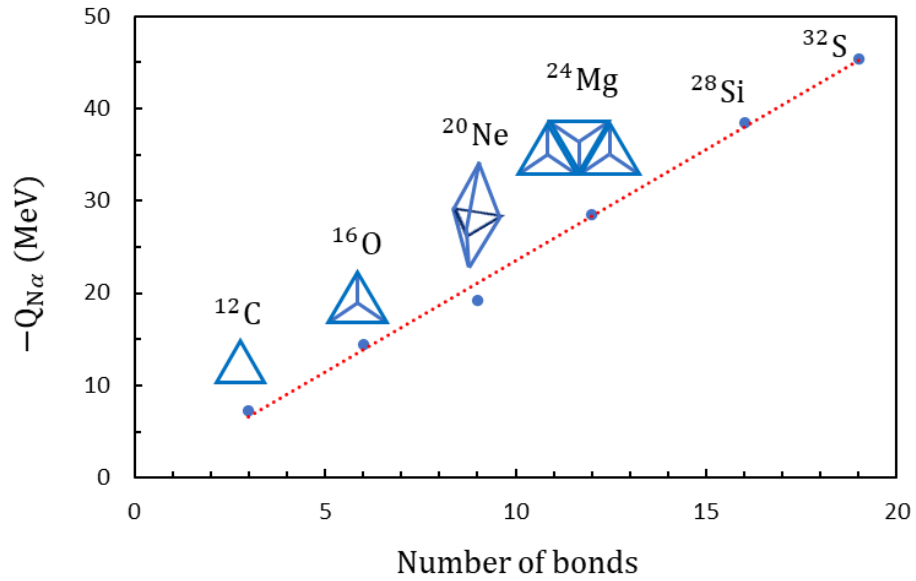


FIGURE 1.32: Schematic overview of the α cluster configurations of the self-conjugate nuclei up to $A = 32$, following the prescriptions of Wefelmeier, Wheeler and Hafstad-Teller. The dotted line represents the linear fit of the various values, and shows the linear dependence between the energy needed to disintegrate a self-conjugate nuclei into the constituent $N\alpha$ clusters and the number of $\alpha - \alpha$ bonds.

into an excited state of the third, a contribution which is of the same order of magnitude as the second order interactions between pairs of α particles, in turn compensating the first order repulsion, and generating the non-additive character of the interaction. Another degree of complexity is the fact that despite the number of adjacent pairs of α particles does not really increase with the increasing number of particles, the actual amount of many-body *bonds* does, and some partial compensations are thus possible.

As previously stated, one of the works which gave high popularity to the α -cluster model was the one published in 1937 by Wilfried Wefelmeier, a polyhedral researcher with two degrees, in Economics and Chemistry [57, 58]. A crucial assumption of this model is the transient nature of the α -clusters inside the nuclei, governed by a characteristic half-life; following the prescriptions of the model, the structures of the various nuclei are described through the adoption of regular geometries, minimizing the free energy. Upon this geometrical assumption, it should be possible to calculate the disintegration energies of self-conjugate nuclei and the weak residual interaction between the α particles; one could plot the dependence of the disintegration energies of the self-conjugate nuclei with respect to the number of $\alpha - \alpha$ bonds, as done in Figure 1.32, to find a surprisingly good linear agreement for nuclei in the $A = 8 - 32$ region (from 2 to 8 α particles). As mentioned, the only discrepancies are related to the unbound ^8Be nucleus (not shown) and to the ^{20}Ne , which shows a slightly underestimated value. All the nuclei are reported with geometrical schemes for the related $N\alpha$ configuration, and this is useful to explain the underestimation in ^{20}Ne . The scheme is in fact describing nine $\alpha - \alpha$ bonds, with a reduced effectivity for the longer (and thus, weaker) bond between the top and bottom α particles in the trigonal bi-pyramid: this feature is responsible of the reduced $E_{dis} = -Q_{N\alpha}$ value for the Neon nucleus. The slope of the fit line, i.e., the ratio between the $-Q_{N\alpha}$ and the respective number of bonds, has an average value of ≈ 2.45 MeV; this value can therefore represent the average $\alpha - \alpha$ binding energy in self-conjugate nuclei.

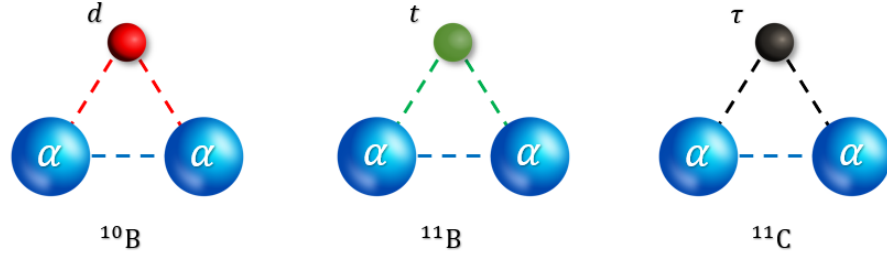


FIGURE 1.33: Conceptual sketch of the three systems formed by the addition of a smaller cluster (d, t, τ) to two interacting α particles.

A very interesting thing, but commonly overlooked in previous literature works, is the possibility to extend this argument also to *non self-conjugate* nuclei, which are treated on the basis of a N_α structure with the addition of a smaller cluster, like d , ${}^3\text{H}$ (t) or ${}^3\text{He}$ (τ). This concept is graphically reported in Figure 1.33 and discussed in the following, under the hypotheses that the $\alpha - \alpha$ bonds do not produce interferences with those formed with the other cluster, and that the spare particle occupies a symmetrical and central position in the structure of the *nuclear molecule*.

If one adds a smaller cluster to self-conjugate nuclei made by two or more interacting particles, several stable systems are produced; even the unbound ${}^8\text{Be}$ nucleus is made stable by the addition of a d, t or τ particle, respectively forming the stable ${}^{10}\text{B}$, ${}^{11}\text{B}$ and ${}^{11}\text{C}$ nuclei. The reason for the stabilization of the three-body system, that would also give a partial answer to the non-additivity concerns of Margenau, is to be looked for in the increase of the number of *Jacobi* coordinates of the nucleus upon the addition of the third particle. The 2α system has in fact a resulting binding energy given by the sum of the expectation values of the kinetic and potential energy, $\langle T \rangle + \langle V \rangle \approx 0$, making the nucleus unbound. However, in 3α systems (or, more generally, in three-body systems), the expectation values become $2\langle T \rangle$ and $3\langle V \rangle$, given that the kinetic energy only depends on *two* Jacobi coordinates, while the potential energy is related to *three* bonds. The negative V term is now larger than the T one, and the binding energy less than zero: the nucleus is bound and the non-additivity problems can be, at least qualitatively, overcome.

With respect to the schemes of Figure 1.33, the $\alpha - \alpha$ binding energy in the non self-conjugate ${}^{10}\text{B}$ nucleus will be related to the difference between the disintegration energy of the nucleus and the energy of the two $\alpha - d$ bonds; the latter can be obtained from the ${}^6\text{Li} \rightarrow \alpha + d$ disintegration energy, which is of 1.4743 MeV [62]. Similar procedures applies for the other non self-conjugate nuclei: in this way, it should in principle be possible to obtain a plot similar to that of Figure 1.32. As an example, the case of the ${}^{10}\text{B}$ nucleus requires to find its disintegration threshold as $|Q_{10B \rightarrow \alpha + \alpha + d}|$, being equal to

$$|Q_{10B \rightarrow \alpha + \alpha + d}| = \varepsilon_{\alpha\alpha} + 2\varepsilon_{\alpha d} \quad (1.43)$$

where $\varepsilon_{\alpha d}$ is the aforementioned ${}^6\text{Li}$ disintegration energy. A generalized formula which applies to all the not self-conjugate nuclei would be:

$$|Q_{N_\alpha + nX}| \equiv N_{\alpha\alpha} \cdot \varepsilon_{\alpha\alpha} + N_{\alpha X} \cdot \varepsilon_{\alpha X} \quad (1.44)$$

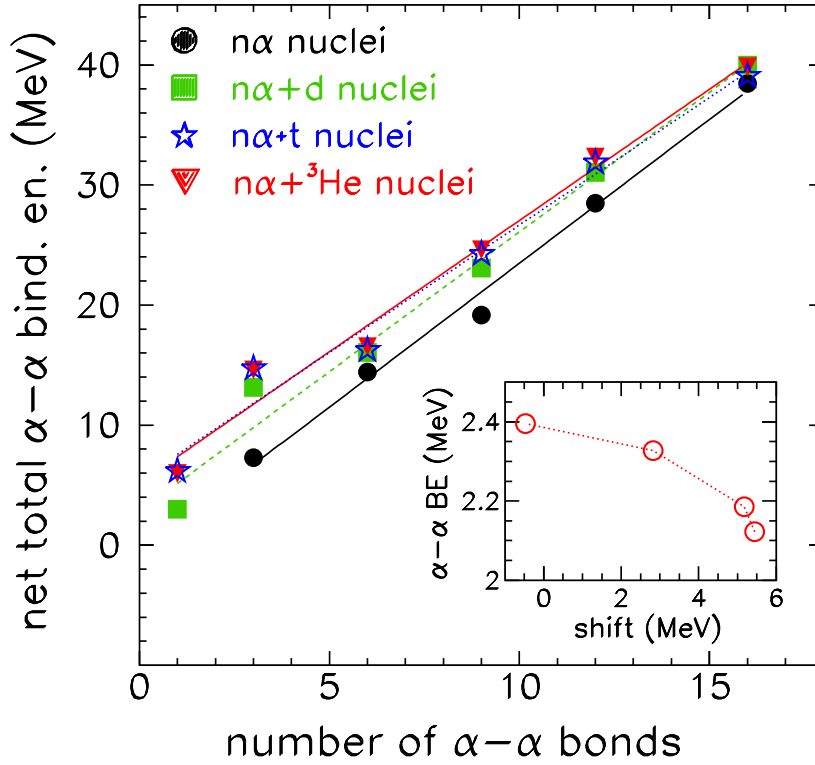


FIGURE 1.34: A modern version of the "Wefelmeier plot" reported in Figure 1.32, here drawn also for non self-conjugate nuclei made by alpha clusters and a deuteron or a triton or a ${}^3\text{He}$. The insert shows the behavior of the predicted $\alpha-\alpha$ binding energy as a function of the intercept (here defined as *shift energy*) obtained from the linear fit of the net total binding energy $\varepsilon_{\alpha\alpha}^{\text{tot}}$. The ideal case of clustering would correspond to vanishingly small shift values and nearly equal values of $\alpha-\alpha$ binding energies, independently from the combination of clusters used to assemble the nucleus.

where X is the generic smaller cluster, and $\varepsilon_{\alpha X}$ is related to the respective disintegration threshold. Reversing the formula:

$$\varepsilon_{\alpha\alpha}^{\text{tot}} \equiv N_{\alpha\alpha} \cdot \varepsilon_{\alpha\alpha} = |Q_{N_{\alpha}+nX}| - N_{\alpha X} \cdot \varepsilon_{\alpha X} \quad (1.45)$$

from which it is possible to calculate and plot the total energy due to $\alpha-\alpha$ bonds, $\varepsilon_{\alpha\alpha}^{\text{tot}}$, as a function of the number of $\alpha-\alpha$ bonds expected for the respective geometric cluster configuration: one bond for ${}^{10}\text{B}$, three for ${}^{14}\text{N}$ and so on. The $\varepsilon_{\alpha\alpha}^{\text{tot}}$ values are reported as a function of the number of $\alpha-\alpha$ bonds $N_{\alpha\alpha}$ in Figure 1.34, both for self-conjugate nuclei and for not self-conjugate nuclei (with the procedure above discussed). If the addition of the smaller particle was to destroy the cluster structure, the plot in Figure 1.34 would not show any type of correlation between the different nuclei, but it is there, and the $\varepsilon_{\alpha\alpha}$ values drawn from the slopes of the curves are actually similar to that of self-conjugate nuclei (black points and line).

Ideally, in a condition where simple two-body interactions are involved, without any interference between the bonds, it should be possible to expect to reach a constant $\varepsilon_{\alpha\alpha}$ value for all the inspected nuclei, regardless of the chemical composition of clusters.

Furthermore, the linear correlation between the $\varepsilon_{\alpha\alpha}^{tot}$ and the number of α bonds should be of the mathematical form $\varepsilon_{\alpha\alpha}^{tot} = N_{\alpha\alpha} \cdot \varepsilon_{\alpha\alpha}$, without any additional energy shift. This is indeed not exactly the case shown by the data: they are well described by a formula of the type $\varepsilon_{\alpha\alpha}^{tot} = N_{\alpha\alpha} \cdot \varepsilon_{\alpha\alpha} + E_{shift}$, and the trends of $\varepsilon_{\alpha\alpha}$ and E_{shift} are shown in the insert of Figure 1.34. It is interesting to note that the deviation from the ideal case is quite limited, underlining a substantially well founded assumption of clustering also in presence of a single *non-alpha* cluster in the nucleus. Further, a curious anti-correlation is seen between $\varepsilon_{\alpha\alpha}$ and E_{shift} , and this is reasonable: larger E_{shift} values are associated to reduced "clustering power" and this is compatible with a reduction of the average binding energy between a couple of α particles (and, obviously, with the energy conservation principle).

Up to now, the attention was only fixed to the masses (i.e. the most relevant of the *ground state* properties) of self-conjugate nuclei, but the development of α structures is crucial also to explain the occurrence of *special* excited states (as, e.g. the Hoyle state in ^{12}C) very far from the ground state, close to the many-cluster disintegration thresholds. This effect was described by Ikeda and collaborators in a pioneer work in 1968, with the publication of the so-called *Ikeda diagram* [63]. In this diagram, the idea that strongly clustered structure should appear in excited states of light nuclei at energies similar to those of some particular thresholds is pictorially translated, marking the disintegration in α particles or in self-conjugate nuclei lighter than the parent one. More extended versions of this diagram can, of course, include non self-conjugate nuclei and their respective $\alpha+n$ decompositions, with different number of neutrons, as suggested by Wolfram von Oertzen in 1999 [64].

A modern rendering of the original Ikeda diagram is reported in Figure 1.35, with all the relevant cluster thresholds for the self-conjugate nuclei up to ^{28}Si . For example, the occurrence of α cluster excited states in ^{20}Ne is expected at energies close to 4.8, 11.9 and 19.17 MeV, and indeed 0^+ states at 6.73, 7.19, 12.4 MeV with a sizable α structure were experimentally observed in α transfer reactions [65]. The situation is less clear in the $E_x > 14$ MeV region, and the work discussed in Chapter 5 will experimentally explore this region.

The intrinsic philosophy of the Ikeda diagram is indicating a very relevant role of clustering phenomena also in nuclear astrophysics: this is due to the fact that the presence of clustered states at energies really similar to those of some particular thresholds may have dramatic impacts on the rates of Helium (α) burning processes in stars, like for the case of the 3α fusion and the ^{12}C Hoyle state [66], or heavier burning pathways like the more violent C+C fusion, as recently discussed in Ref. [67].

1.2.3.3 Comparison of nuclear and molecular excitations: the collective model

Phenomena leading to the formation of α clusters are naturally associated to the occurrence of large *deformations* in the nucleus. These structures can therefore be characterized by significative vibrational and/or rotational excitation, in a fashion very similar to the one observed in molecules and already sketched in Section 1.1.5. Figure 1.36 reports the coupling of angular momenta for a nucleus having a symmetry axis (fixed in the nucleus reference frame) [68]. Another axis, $\hat{z}$, is instead fixed in space. Analogously to

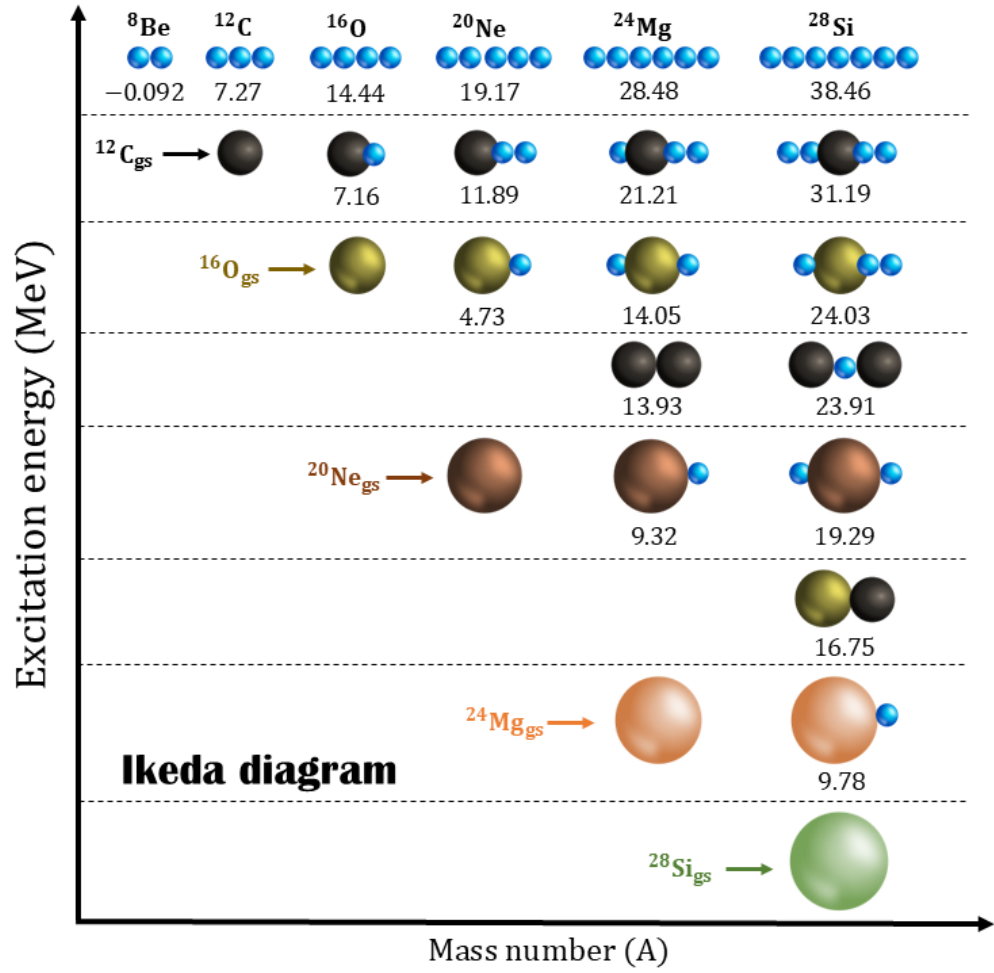


FIGURE 1.35: Ikeda diagram for the self-conjugate nuclei up to ^{28}Si . The number under every cluster disintegration represent the respective threshold energy, expressed in MeV.

the molecular case, there is no quantum mechanically allowed rotation about the symmetry axis in the lowest states³; at variance, if there are "loose nucleons" in outer (and non-completely filled) shells, the intrinsic motion of such particles takes places about the symmetry axis, with a component angular momentum $\hbar K = \sum_i \hbar K_i$, being K_i the component associated to a single nucleon. In the following, the collective angular momentum, perpendicular to the symmetry axis, will be indicated with $\vec{R}$, and the total angular momentum with $\vec{J}$. The component of $\vec{J}$ along the $\vec{z}$ axis will be $\hbar M$, while the component of $\vec{J}$ along the symmetry axis will be obviously $\hbar K$. In a semiclassical fashion, the absolute value of $\vec{R}$ is therefore given by:

$$|\vec{R}| = \hbar \sqrt{[J(J+1) - K^2]} \quad (1.46)$$

As seen for the molecular case, it is possible to make some simple investigations on such a complex coupling situation if one can assume that nuclear motion can be separated into: (i) a collective rotation; (ii) a vibration around the static equilibrium shape; (iii) an intrinsic motion of the loose nucleons in the non-spherical potential. Typically, the

³A beautiful experimental observation of such hindered rotation is related to the evolution with the temperature of the molar specific heat for diatomic molecules; see, e.g., [69]

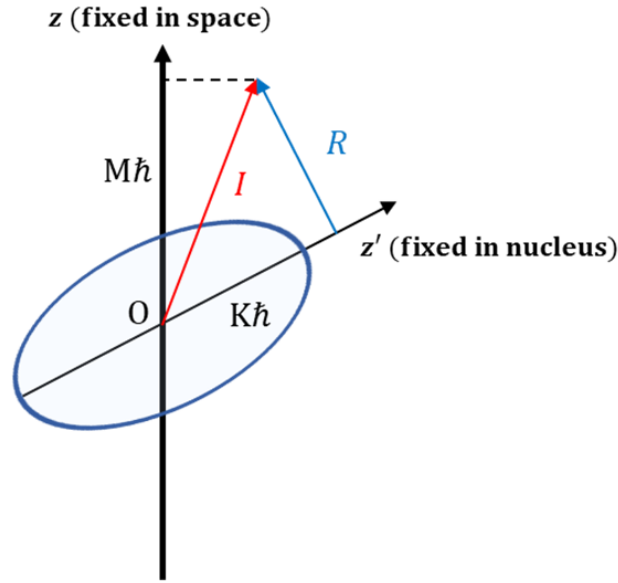


FIGURE 1.36: Vector scheme of the coupling of the angular momenta for a nucleus with a symmetry axis (z') fixed in the reference frame of the nucleus itself (*intrinsic* axis), with respect to an axis fixed in space (z).

excitation energy scales corresponding to each one of these motion increases going from (i) to (ii) to (iii); again, this is similar to what is observed in molecules.

Concerning the states due to intrinsic excitations, the motion of nucleons inside a deformed quadrupolar potential has been studied in details by Sven Nilsson in the '60s; the solution of the Schrödinger equation leads to a very peculiar sequence of single particle states, reported in Figure 1.37 (the Nilsson diagram). The most important thing to note is the (partial) removal of the degeneration of the ordinary (spherical) shell states: for example, while in the spherical shell model the $d_{5/2}$ shell can host protons, or neutrons, having projections of the total angular momentum $\pm\frac{5}{2}, \pm\frac{3}{2}, \pm\frac{1}{2}$, in the Nilsson model the $d_{5/2}$ shell splits into three different *deformed* (or Nilsson) shells: the $1/2^+, 3/2^+, 5/2^+$ ones, with different energies. Note that the $1/2, 3/2, 5/2$ numbers indicate the quantum number K_i of the single particle sitting in the specific orbital, while the plus or minus superscripts indicate the parity of the single particle wave function. In each Nilsson shell, it is possible to host *two* protons or two neutrons; for example, the $3/2^+$ shell can be filled by two protons (or, analogously, two neutrons) having two oppositely directed projections of angular momentum, i.e. $\pm 3/2$. A very interesting implication of this simple observation is that well deformed nuclei having even Z and even N fully occupy the Nilsson orbitals; this property will be later used to explain the insurgence of α clustering in the $A \approx 20$ region of the nuclide chart.

A brief discussion of the vibrational states will now follow: these are typically associated to nearly spherical nuclei, with few nucleons outside a *core* made by fully closed shells. The loose nucleons can trigger a collective oscillation of the whole nuclear surface, that is characterized by a certain number of vibrational quanta (the *phonons*), having energy $\hbar\omega$ and angular momentum $\hbar\ell$, according to the prediction of a quantum harmonic oscillator. One could therefore expect these vibrational excitations to be (approximately) *equally*

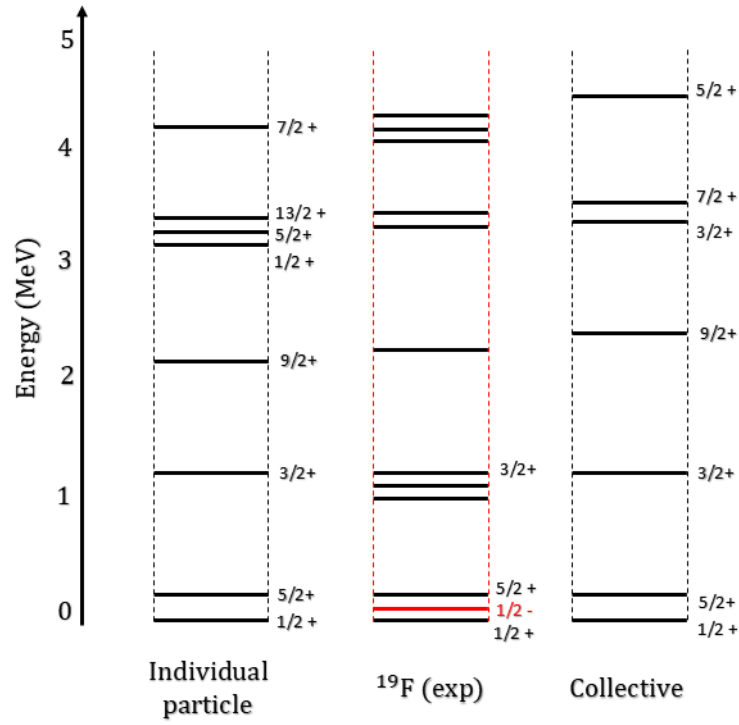


FIGURE 1.38: Level scheme of the ^{19}F nucleus as seen from experimental measurements (middle), and as predicted from the Individual particle (left) and Collective (right) models. The first excited state of the ^{19}F nucleus, at $E = 109.9$ keV, is highlighted in red. Adapted from [68]

intrinsic motion⁵.

As a very interesting application of the theory above discussed, let's consider the extremely special case of low energy excited states of ^{19}F . From nuclear magnetic resonance measurements, it was known from the '50s that the spin of this nucleus was $J = 1/2$, with positive parity. Since ^{19}F has one proton and two neutrons outside the ^{16}O doubly magic core, according to the spherical shell model, it is expected that the two extra neutrons would couple to give zero contribution to the spin of ^{19}F , and that the odd proton alone would determine the J^π of the entire nucleus. In particular, if one places this odd proton in the first available shell after the $A = 8$ gap (i.e. the $1d_{5/2}$), a $J^\pi = 5/2^+$ should be expected, totally different from the experimental observation. Why? Because $A \approx 20$ is a region with strong prolate *nuclear deformation*, and the single-particle orbitals to be used must follow the Nilsson diagram. Since for prolate deformation the lowermost Nilsson orbital after the $A = 8$ gap is the $1/2^+$ one, the mystery is solved.

After understanding the process, one could expect to unveil the nature of (almost) all the low-lying states in ^{19}F in terms of Nilsson single-particle excitations and/or collective rotations built on them. By looking at Figure 1.38 there are indeed several correspondences between the experimental findings and the predictions, and this is joyful. But

⁵For the special case of $K = 1/2$, this formula must be modified to take into account the sizable coupling of rotational and intrinsic motion; this effect is due to the *Coriolis forces* acting on a single particle moving into a rotating reference frame.

there is something strongly unexpected (and, at first sight, *discouraging*): it is *impossible* to describe (with the models here discussed) the nature of the first excited state, a *lovey-dovey* $1/2^-$ at only 109.9 keV of excitation energy⁶.

How can one explain its structure, then? It can be understood as a *joke* due to α clustering! Let's dive into the reasons for this exclamation. The filling of Nilsson orbitals for the ground state is quite straightforward: outside the $A = 8$ shell closure, there are two neutrons filling the $1/2^+$ Nilsson (neutron) orbital and the odd proton filling the $1/2^+$ Nilsson (proton) orbital; the odd proton determines the J^π of the nucleus. Now, the promotion of the odd proton to the next or to the next-to-next Nilsson orbitals would lead to a positive parity state, and the same would occur if some sort of collective rotation would be based on the ground state, in full disagreement with the negative parity of the *lovey-dovey*. But there is another possibility. One could imagine that a "frozen" proton from the *core* (in particular, from the more external $1/2^-$ Nilsson orbital) leaves the *paternal house* and makes the *jump* to reach the odd proton in the $1/2^+$ orbital. For sure, the nucleus can regain a bit of energy by pairing the protons in the $1/2^+$ orbital, but the energy gained is by far *smaller* than the energy needed to (i) break the pair in the $1/2^-$ orbital, and (ii) to shoot the proton above the $A = 8$ gap; it is impossible, on this simple reasoning, to imagine such a complicated mechanism describing a state of only 110 keV... However, the J^π of the state predicted in this way would be the right one! Is there some neglected hidden energy source, and could that counterbalance the huge energy pawn in the "jumping" of the frozen proton? Yes, there is, and it is due to the onset of a strong α clustering phenomenon. When the frozen proton reaches the odd one in the $1/2^+$ orbital, a *magic* (in all the senses) configuration of two protons and two neutrons is seen, where they are sitting into orbitals with more or less the same energy, with positive parity and $\pm 1/2$ angular momentum projections, and furthermore completely filling the orbitals. This situation is nearly identical to the formation of an α particle in the $1s_{1/2}$ shell, leading to the saturation of nuclear forces with the huge gain of binding energy that characterize the α structure (see Section 1.2.2.1). This very large energy gain due to α clustering counterbalances (and rewards) all the efforts made by the frozen proton to escape from the core: a *breath* (110 keV) is sufficient to make this *scheming* occur⁷! This is certainly one of the most *beautiful* manifestation of α clustering in a non self-conjugate nucleus, and this is a condition that is common to other $A \approx 20$ deformed nuclei⁸.

1.2.3.4 Rotational bands in α -clustered nuclei

The rotational properties and the derived bands in self-conjugate nuclei can be suitably introduced through the study of the ^8Be system, a quasi-bound nucleus just 92 keV

⁶This apparently *innocent* state is the culprit of several difficulties when facing off the nuclear reactions leading to the destruction of fluorine in stars. In fact, with a so low excitation energy, this state can be thermally excited in stars and the reactions can occur by light particles (p or α) colliding on this state, and not only on the ground state, of ^{19}F .

⁷It is interesting to note that 110 keV is truly a *tiny* quantity for a light nucleus: if nuclear forces would have been *very slightly different* from what they are, these 110 keVs could have been 50, or 10 or even... -50 keV, and the *lovey-dovey* could have been the *ground state* of ^{19}F ! This would have led to insurmountable difficulties in describing its nature with the shell or the collective model, and perhaps all the history of nuclear structure as it is currently known would have been different.

⁸This peculiar property has also an influence on the onset of incomplete fusion mechanism in heavy ion collisions at energies slightly above the barrier; this property was investigated through a phenomenological model in Ref. [70].

above the α -decay threshold, and with a very narrow decay width of just 6 eV (hindered by the Coulomb barrier). Its ground state is a 0^+ state, and it is the head of a rotational band which keeps going with the first 2^+ excited state at 3.03 MeV and the 4^+ state at 11.4 MeV. This band serves as a typical example of the 2α di-nuclear configuration of cluster structures. The $\alpha - \alpha$ potential has been already introduced in Section 1.2.3.1, and it is known to have two fundamental features: a strong repulsive behavior in the inner region, within the range of ≈ 1.8 fm (the so-called *structural repulsive core*), and an attractive nature for ranges up to the contact distance of around 3.5 fm.

Let's make some simple calculations. From Equation 1.47, since $K = 0$ and $J = 2$, the inertial parameter $\frac{\hbar^2}{2\mathcal{I}}$ is determined from the $2^+ \rightarrow 0^+$ energy jump ($\Delta E_x \simeq 3$ MeV):

$$\frac{\hbar^2}{2\mathcal{I}} \simeq \frac{\Delta E_x}{J(J+1)} = \frac{3 \text{ MeV}}{2 \cdot 3} = 0.5 \text{ MeV} \quad (1.48)$$

Now, assuming a dumbbell configuration with the two α particles in contact, and considering the radius of an α particle being $r_\alpha \simeq 2$ fm, from the Steiner theorem it is possible to calculate the theoretical moment of inertia:

$$\mathcal{I} = 2 \cdot \left[\frac{2}{5} m_\alpha r_\alpha^2 + m_\alpha r_\alpha^2 \right] \simeq 42300 \frac{\text{MeV fm}^2}{c^2} \quad (1.49)$$

a value that is in excellent agreement with the experimental inertial parameters; in fact:

$$\frac{\hbar^2}{2\mathcal{I}} = \frac{\hbar^2 c^2}{2\mathcal{I} c^2} = \frac{(197.3)^2 \text{ MeV}^2 \text{ fm}^2}{2 \cdot 42300 \frac{\text{MeV fm}^2}{c^2} c^2} = 0.46 \text{ MeV} \quad (1.50)$$

This is a further clue of the dimeric nature of ^8Be due to its α cluster nature. Similar values for the inertial parameter are found when considering the $4^+ \rightarrow 2^+$ or the $4^+ \rightarrow 0^+$ energy jumps.

The identification of di-nuclear structures in ^{16}O and ^{20}Ne nuclei, in addition to that of ^8Be , proved that molecular-like structures can also occur in a large region of nuclear many-body systems. Figure 1.39 shows two rotational bands with $K^\pi = 0^+$ and 0^- for both ^{16}O and ^{20}Ne . As it can be seen, the states with $J^\pi = 1^- \div 7^-$ in the negative parity band are located above the α -decay threshold energy, and can thus be observed as resonances in α -scattering experimental data. The analysis of the α -decay widths of the negative parity band states points out Γ_α values almost equal to the corresponding single-particle (Wigner) limit, i.e., that the probability of existence of α -clusters in the nuclear surface is almost unitary for all of these resonance states. This is a *smoking gun* proving that the intrinsic nature of the negative band states in these two self-conjugate nuclei are of *peanut-like* di-cluster structures: $\alpha + ^{12}\text{C}$ for ^{16}O and $\alpha + ^{16}\text{O}$ for ^{20}Ne . The moment of inertia obtained from the energy jumps of such rotational states are in agreement with the picture of two touching (asymmetric) clusters.

However, if the $K^\pi = 0^-$ band has a di-nuclear structure, one could expect the existence of a $K^\pi = 0^+$ band which forms, together with the former, some type of "parity doublet bands", corresponding to the *inversion doublet bands* typical of some chemical systems, like *ammonia* (NH_3) or similar [71], as shown in Figure 1.40.

This $K^\pi = 0^+$ band, with its di-nuclear structure, is shown in Figure 1.39, and it is

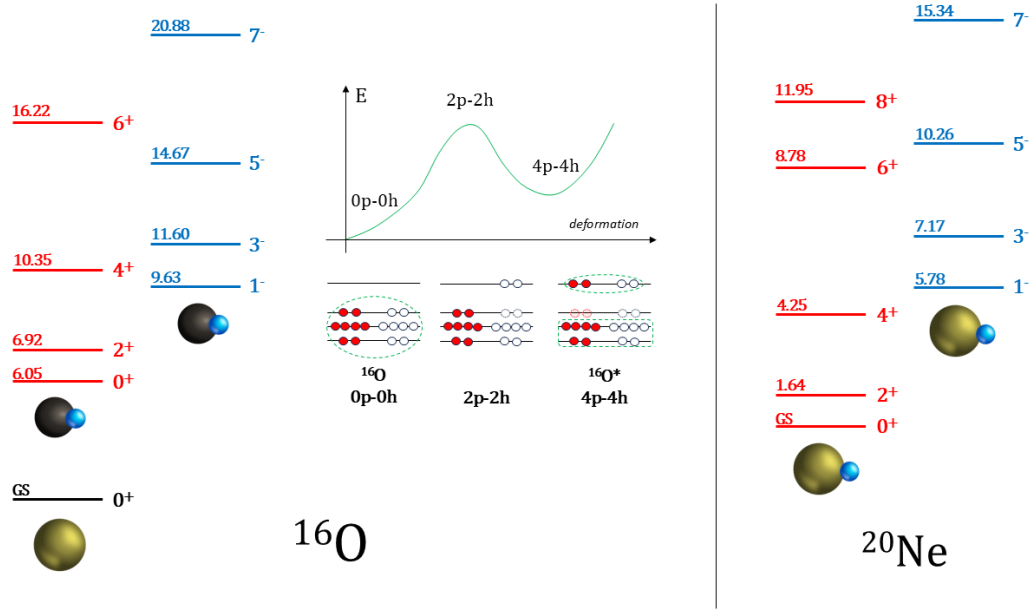


FIGURE 1.39: Rotational bands of the ^{16}O (left) and ^{20}Ne (right) nuclei explained in the fashion of the cluster model, through the representation of clustered $^{12}\text{C} + \alpha$ and $^{16}\text{O} + \alpha$ structures. The adoption of *peanut-like* structures allows to justify the level sequence of the rotational band better than the classical spheroidal models. *Middle*: Schematic representation of cluster formation in the 6.05 MeV ^{16}O state as a four particle-four hole ($4p - 4h$) excitation leading to a sizable structural change.

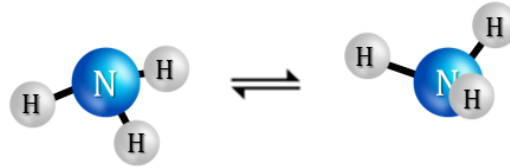


FIGURE 1.40: Pyramidal inversion of the ammonia (NH_3) molecule. This peculiar phenomenon gives rise to inversion doublet bands in this type of molecules.

the one starting with the first 6.05 MeV excited state in ^{16}O ⁹ and with the ground state in ^{20}Ne . This band, for the ^{16}O nucleus, can be assumed to be built on a *quartet* configuration [73], i.e. an excitation mode of the four α clusters. The model of the molecular-like structure is thus based on the strong internal coupling between the four nucleons in an α particle and the weak correlation between the four-particle cluster and the residual core nucleus (e.g., one α and the ^{12}C core in ^{16}O). These different excitation modes and the respective structural changes induced in the ^{16}O nucleus are represented in the central part of Figure 1.39. Here, the 6.05 MeV state, with a pronounced α structure, is seen as a result of a complex $4p - 4h$ excitation leading to a sizable energy re-gain because of the formation of an α -like structure in an excited orbital filled by four correlated nucleons.

⁹Because of the double shell closure in the ground state of ^{16}O , the resulting spherical configuration can not give quantum mechanically rise to a rotational band. A different story is expected for roto-vibrational bands when ^{16}O is assumed to have the shape of a *tetrahedron*; see Ref. [72] for further details based on symmetry arguments.

1.2.3.5 Quarted excitations in self-conjugate nuclei

Akito Arima, Vincent Gillet and Joseph Ginocchio proposed the so-called *Quartet Model* at the beginning of the 70s [73]: in this framework, the atomic nuclei were described as made by α particles lying in orbitals analogue to those of the harmonic oscillator, studying the transitions of *whole quartets* from one orbital to another, allowing in this way to describe the presence of 0^+ cluster states at high excitation energies. This model was applied with some success especially to the spectroscopy of ^{20}Ne , as already done in Refs. [74, 75] and within the experimental work discussed in Chapter 5 of this thesis.

The main aim of this model is thus to succeed in the description of the structure of low-spin, highly excited states of self-conjugate nuclei, where the shell model very often fails. The basic assumption of this model is the existence of *quartets*, strongly interacting structures of 2 protons and 2 neutrons occupying degenerate single-particle states (α -like particles). Another fundamental assumption is the fact that, in a self-conjugate nucleus, the separation energy of a nucleon is *larger* than that of an α particle: this means that a lower amount of energy is needed to make the nucleus emit an α particle rather than a single proton or neutron, reflecting the strong character of the nuclear interaction inside the α particle. For example, in ^{16}O , only 7.2 MeV are needed to emit an α particle, while more than double the energy is required for the emission of a *neutron*, an indication of how one nucleon in the nucleus interacts way more strongly with the three forming the emitted α particle than with the others in the nucleus. The interaction energy between nucleons inside the α particle is, in fact, approximately double than that existing with the others in the ^{12}C core.

This model describes the existence of excited states in self-conjugate nuclei through the particle-hole excitations of quartet structures, which are characterized by the mentioned internal strong bindings and weak mutual external interactions. This means that the first 0^+ excited states of self-conjugate nuclei are explained through the excitations of whole quartets rather than single nucleons, given their strong internal bonds; for nuclei up to ^{52}Fe , these quartets are limited to only some of the shells of a spherical harmonic oscillator, these being the $(0p)$, $(0d, 1s)$ and $(0f, 1p)$ shells. The interaction energies between the different shells can be calculated by different means: for example, those of single-step excitations $[(0p) - (0d, 1s)]$ and $[(0d, 1s) - (0f, 1p)]$ can be obtained by the determination of the position of the first 0^+ excited states of, respectively, ^{16}O and ^{40}Ca , known from the literature to have large α structures [73]. On the other hand, the two-level $[(0p) - (0f, 1p)]$ excitation is generally supposed to be small, given the extremely limited overlap of the relative wave functions of these shells.

Quartet-hole interactions are supposed to be constant in this framework, regardless of the mass number of the nucleus and the number of excited quartets. The common notation to indicate quartet configurations in the homonymous model is to consider the α particle as the core, and consequently use the $[xyz]$ notation to indicate the disposition of the quartets in the various aforementioned shells. The term x refers to the number of quartets in the $N = 1$ major shell, y is the number of those in the $N = 2$ major shell, while z refers to the quartets in the $N = 3$ shell; an example of this notation is given in Figure 1.41 for the ground state and the first excited state of the ^{16}O nucleus. Another way to compactly indicate the configuration is to note it as Q_p^x , $Q_{(sd)}^y$ and $Q_{(pf)}^z$. If one restricts the calculation to the first two orbits ($N = 1, 2$), the energy of a determined

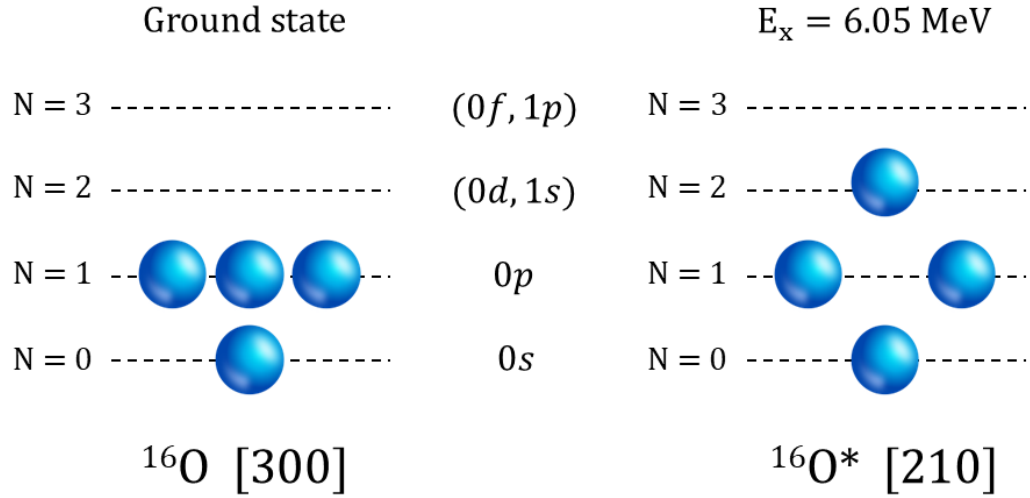


FIGURE 1.41: Example of quartet configurations for the ground state (left) and the first excited state (right) of the ^{16}O nucleus. Note: the quartet sitting in the $0s$ orbital is never indicated in the compact notation $[xyz]$, being considered as the *basic brick* of the model.

excited state will be given by the equation:

$$E^* = (Q_p^x Q_{(sd)}^y) = E_0(4x + 4, 2x + 2) + E_0(4y + 16, 2y + 8) - E_0[4(x + y) + 4, 2(x + y) + 2] - E_0(16, 8) + (3 - x)yV_{p,(sd)} \quad (1.51)$$

where the terms $E_0(A, Z)$ are the interaction energies of nuclei with A nucleons and Z protons, while $V_{p,(sd)}$ reflects the interaction between y quartets in the $N = 2$ level and $(3 - x)$ holes in the $N = 1$ level. This is an obvious consequence of the fact that every quartet promoted to the $N = 2$ level leaves a hole in the $N = 1$ level. The $V_{p,(sd)}$ quantity is determined by assuming the one-quartet one-hole excitation energy equal to 6.05 MeV, i.e., the first 0^+ state of ^{16}O , which will be discussed in detail in Chapter 4. The quartet model proved very useful to describe the scheme of excited states in the ^{20}Ne nucleus: as a prove, signals of the presence of $[220]$ and $[211]$ quartet states have been reported in the literature, respectively through α transfer [74] and compound nucleus [76] reactions. Chapter 5 also discusses an experiment on the $^{19}\text{F}(p, \alpha_i)^{16}\text{O}$ reactions, aimed to shed light on the structures of some particular ^{20}Ne excited states, and to solve long-standing discrepancies in previous literature data.

Modern and more evolute versions of the quartet model, which can for example include microscopical features like the arbitrary values of isospin and angular momentum for the quartets [77, 78], allowed to obtain much more accurate predictions about the spectroscopy of light and medium mass nuclei. Nevertheless, this is suitable not only for the self-conjugate ones: an excellent description of the low energy part of the level scheme has been obtained also for some non self-conjugate nuclei.

1.2.3.6 The LCNO: a linear combination method for the study of nuclear orbitals

The molecular orbital (MO) model is a powerful microscopic method to investigate clustering phenomena: in this framework, the nucleons are individually described as moving in an average single particle field generated by the system, whose shape and other properties contain information about the clustering characteristics of the nucleus itself [79]. The aim of the MO model is thus the preservation of the concept of single-particle mean field, even in presence of many centers due to the development of clustering in the nuclear structure. In addition, it can grant information about the motion of the valence nucleons moving outside the clusters, as in the case of non self-conjugate nuclei, which will be treated in the next Section. This is more of a general model, with three different variants most commonly used:

- The *two-center shell model* variant, generally used for *two-cluster* systems (e.g., isotopes of beryllium): in this framework, the single particle orbitals (the so-called *molecular orbitals*) are generated by the local average potential, which has a shape similar to the cluster configuration and density distribution of the system [80].
- The *LCNO method*, based on a parallel with the *LCAO* approach (*Linear Combination of Atomic Orbitals*) of molecular chemistry, with the difference that this is based on *Nuclear Orbitals*, hence LCNO [81, 82]. This approach regards the molecular orbitals as constructed by a linear combination of the nuclear orbitals around the centers of the clusters constituting the nucleus. This method will be the focus of the attention in the remaining part of this Section.
- The *self-consistent LCNO, or Hartree-Fock-LCNO* method, where the orbitals are further combined to obtain self-consistent resulting molecular orbitals [83]; as *spoiled* by the name, the coefficients of the linear combinations, in this case, are determined by the solution of the Hartree-Fock equation.

The aim is to go deeper in the study of the LCNO method, the second listed above. In this case, as in the two-center shell model, the form of the mean field is considered to be similar to the density distribution of the cluster structures inside the nucleus. Special attention is dedicated, as expressed in the Group Theory (see Section 1.1.5), to the symmetry aspects of the mean field which are mirroring those of the cluster distribution itself. This means all those symmetry operations leaving the cluster density distribution *and* the mean field invariant, gathered into a *group*. Therefore, the single particle wave functions, i.e., the nuclear orbitals, governed by the mean field, must belong to an irreducible representation of the group(s). In the LCNO method, the nuclear orbitals are linearly combined to obtain molecular orbitals which will be the basis states of the *irreducible representations* of the respective groups.

Let's exploit two cases to explain the LCNO method: the case in which the mean field is determined by an $\alpha - \alpha$ structure (like a *dumbbell* for gym training), and the one in which it is determined by a 3α structure (similar to an *equilateral triangle*). In both cases, to make the problem easier, the *nuclear orbitals* are treated as simple *harmonic oscillator* shell model wave functions (see, e.g., Section 1.2.2.3 for some explicit functional forms) developing around the cluster center.

$\alpha - \alpha$ **systems**: if one locates the two α centers in the positions $S/2$ and $-S/2$ along the z -axis (the inter-cluster axis), the nuclear orbitals up to the $0p$ one will be denoted as:

$$|0s, S/2\rangle ; |0p_i, S/2\rangle ; (i = x, y, z) ; \quad (1.52)$$

$$|0s, -S/2\rangle ; |0p_i, -S/2\rangle ; (i = x, y, z) ; \quad (1.53)$$

The following symmetry properties of the mean field would be required: (i) the invariance with respect to the rotation around the z -axis, $R_z(\theta) = \exp(-i\theta L_z)$; (ii) invariance to the reflection about the xz -plane, $\sigma_V(xz)$; (iii) invariance for the 180° rotation around the perpendicular y -axis, $R_y(\pi) = \exp(-i\pi L_y)$. It is also useful to underline that the combination of the operations (ii) and (iii) is correspondent to the *Parity* (P) operation, i.e., $P = \sigma_V(xz) \cdot R_y(\pi)$. Of course, the parity and rotation invariance are valid as long as the two clusters in the system are identical, like the two α s in the case of $\alpha - \alpha$ systems, while they must be disregarded when studying systems of two non-identical clusters (e.g., $\alpha + {}^{16}\text{O}$).

A combination of all the requirements listed above is a set of operation which, following the definitions given in Section 1.1.5, constitutes the group denoted as $D_{\infty h}$. The three sets of operators, $\{R_z(\theta)\}$, $\{1, P\}$ and $\{1, R_y(\pi)\}$, all form their respective groups, which are all *Abelian* (or *commutative*) and whose irreducible representations are one-dimensional. The basis states of the representations are the eigen-states of the respective operators P , $R_z(\theta)$ and $R_y(\pi)$; however, considering that the two latter operators do not commute, one can build the simultaneous eigen-states of $R_z(\theta)$ and P upon the *nuclear orbitals* of Equation 1.53. These are eight, of the following types:

$$f_1 = n_1 [|0s, S/2\rangle + |0s, -S/2\rangle] ; f_2 = n_2 [|0s, S/2\rangle - |0s, -S/2\rangle] ; \quad (1.54)$$

$$f_3 = n_3 [|0p_z, S/2\rangle - |0p_z, -S/2\rangle] ; f_4 = n_4 [|0p_z, S/2\rangle + |0p_z, -S/2\rangle] ; \quad (1.55)$$

$$f_5 = n_5 [|(0p_x + i0p_y), S/2\rangle - |(0p_x + i0p_y), -S/2\rangle] ; \quad (1.56)$$

$$f_6 = n_6 [|(0p_x + i0p_y), S/2\rangle + |(0p_x + i0p_y), -S/2\rangle] ; \quad (1.57)$$

$$f_7 = n_7 [|(0p_x - i0p_y), S/2\rangle - |(0p_x - i0p_y), -S/2\rangle] ; \quad (1.58)$$

$$f_8 = n_8 [|(0p_x - i0p_y), S/2\rangle + |(0p_x - i0p_y), -S/2\rangle] ; \quad (1.59)$$

where the f_{1-4} terms have $L_z = 0$, the $f_{5,6}$ have $L_z = +1$ and the last two, f_{7-8} have $L_z = -1$. Some of these functions (e.g., f_1 and f_3) have the same L_z value, and are thus not orthogonal: they need to be *orthogonalized* (for example, through the *Gram-Schmidt method* [84]), yielding orthonormal nuclear "molecular" orbitals denoted as φ_1 and φ_3 . Then, the many-body wave function of the $\alpha - \alpha$ system (${}^8\text{Be}$) is obtained by filling the two orbitals, φ_1 and φ_2 . Furthermore, as shown in the next Section, if one considers the structures of the ${}^{9,10}\text{Be}$ nuclei as *nuclear molecules* described by the 2α core plus the *valence* neutrons, their wave functions are obtained by accommodating the first 8 nucleons in the φ_1 and φ_2 orbits (to form the 2α core), and the remaining ones in the suitable valence orbitals, chosen between the remaining φ_{3-8} functions with increasing energy sequence [79].

3α system (equilateral triangular geometry): structures with three cluster centers can be described through the adoption of an equilateral triangle geometry, and all the derived symmetry properties. If one defines the three clusters as A, B and C , the *nuclear*

orbitals up to the $0p$ orbital can be denoted as

$$|0s, K\rangle; \quad |0p_i, K\rangle \quad (i = x, y, z) \text{ and } K = A, B, C \quad (1.60)$$

The mean field of the system must obey to the following symmetry properties: (i) invariance for $2\pi/3$ and $4\pi/3$ rotations around the z -axis ($R_z(2\pi/3)$ and $R_z(4\pi/3)$); (ii) invariance for the π rotation around the y -axis, $R_y(\pi)$ and (iii) the invariance with respect to the reflection about the xy -plane, defined as σ_h . When combining the last two operations, the resulting $\sigma_h R_y(\pi)$ is nothing more than the reflection about the yz -plane; to make a long story short, from all the combinations of the three requirements, it is possible to obtain a total of twelve operations (including the identity E). Following the prescriptions reported in Section 1.1.5, this gathering of operations forms the D_{3h} group, useful for the study of systems like the ^{12}C nucleus (see Chapter 3).

At the end of the day, in the LCNO approach, orthogonalized nuclear orbitals are linearly combined, similarly to what happens in the LCAO method, to obtain molecular-like orbitals. Albeit the LCNO method appears useful, as a first approximation, to obtain the actual form of the wave functions, it does not give their expression as a function of spin and parity eigen-states. These could, however, be obtained through a study based on the application of Peierls and Yoccoz theories on the projections of parity and angular momentum, as it will be shown in Section 1.2.3.8.

1.2.3.7 Nuclear molecules

An interesting aspect of the insurgence of clustering phenomena in self-conjugate nuclei is the possibility to treat them as geometrical distributions of α particles, as it happens during the study of molecular or crystal structures; however, another level of analogy between the nuclear and the chemical world is developed also when studying the non self-conjugate nuclei. In these cases, as already mentioned, the spare neutrons (i.e., those which are virtually not part of any α particle) can be treated as *covalent* particles, like the electrons binding atoms together in common chemical bonds.

As discussed in Ref. [85], interesting systems to study the insurgence of molecular states are nuclei with mass 9 and 10, especially beryllium and boron systems. In these cases, the nucleus is composed by two α particles and one or two excess nucleons. A sizable fraction of the following discussion will be dedicated to the example of the ^9Be nucleus, to the development of its excited levels scheme as a function of the two-centered dimer structure, and finally to the construction of more extended polymer structures by the addition of $\alpha + 2n$ units to the excited ^{10}Be states, used as a base.

The prototypal example for the study of two center states is thus the ^9Be nucleus, the only stable isotope of this element, where two α particles are bound together by a wanderer neutron. The first theoretical attempts to the description of binding in non self-conjugate nuclei was made by Hafstad and Teller [59], exploiting the situations of those isotopes having only one excess neutron with respect to the corresponding self-conjugate system, like ^5He , ^9Be , ^{13}C and ^{17}O . One of the most important outcomes of the study is how the binding energy of these nuclei is not only dependent on the $\alpha - \alpha$ interaction, but also on the additional degree of freedom introduced by the extra neutron(s). On this basis, while the study of the ^5He nucleus proved useful to describe the $\alpha - n$ interaction, the ^9Be is the key to understand the Hamiltonian contributions deriving from exchange interactions. As already mentioned in some previous discussions,

the ${}^9\text{Be}$ system can be seen as a nuclear analogue to the H_2^+ ionized molecule, where the two protons are bound by the exchange of an electron; in the same way, Hafstad and Teller gave the basic description of neutron-rich nuclei as systems in which the α clusters are stabilized through the exchange of the spare neutrons. Given the clear analogy with the molecular covalent binding process, these extra neutrons are often called *covalent* neutrons. However, the discussion of the ${}^9\text{Be}$ properties is a little complicated by the fact that, when taken on its own, neither of its sub-units ($\alpha - \alpha$ or $\alpha - n$), represents a bound nucleus, since ${}^5\text{He}$ and ${}^8\text{Be}$ are not stable nuclei; the final binding effect is, in fact, only given by the three-body interaction. In a milestone work published at the end of the '70s, Fonseca, Revai and Matveenko demonstrated that, despite the covalence neutron has a mass smaller but of similar scale to the cluster centers, the three-body problem can still be suitably studied within the Born-Oppenheimer approximation, with a sizable simplification in the study of the binding mechanism and of the spectroscopy of ${}^9\text{Be}$ [86].

Since the level schemes of ${}^{9,10}\text{Be}$ and ${}^{10}\text{B}$ are quite well established [62], the deriving information can be exploited to investigate the matter and shed light on the specific structure of polymeric nuclear structures. Also in the case of non-self conjugate nuclei, it is expected to observe sizable clustering phenomena in nuclear states near to the various thresholds predicted by the Ikeda model [64].

The ${}^9\text{Be}$ nucleus is obtained by binding the spare neutron to the ${}^8\text{Be}$ core, described through the $\alpha - \alpha$ potential discussed in Section 1.2.3.1. Figure 1.42 reports the orbital scheme (*correlation diagram*) of the two-center (2α) nuclear system, which is useful to understand the level sequence of the valence neutron in the ${}^9\text{Be}$ nucleus. As reported in the chart, and analogously to the molecular chemistry case described in Section 1.1.3.1, every level is subject to a four-fold characterization, on the basis of: its angular momentum ($\ell = s, p, d, f$), the projection of the spin ($\frac{1}{2}, \frac{3}{2}, \frac{5}{2}, \frac{7}{2}$), its parity ($\pi = +, -$) and its symmetry ($p = \textit{gerade}$ or *ungerade*), relative to the property of exchange of the identical cores around which the valence neutron roams. This correlation diagram allows to set some correspondences between the covalent states of the $2\alpha + n$ system, i.e the separated configuration at large distances, and the levels of the ${}^9\text{Be}$ nucleus, i.e. the united system formed at very small r values. For example, at large distances, the lowest state with positive parity has $J^\pi = 1/2^+$, $p = \textit{ungerade}$, and it is of *binding* σ nature (red dashed line). For the limit of the unite nucleus, with $r \rightarrow 0$, this state rises in energy and merges to the $d_{5/2}$ level. The overlap and the exchange of positions between the levels in the separate $2\alpha + n$ system and in the ${}^9\text{Be}$ nucleus is extremely important: while the aforementioned $1/2^+$ (u) level is the lowest at larger distances, the ground state of the ${}^9\text{Be}$ nucleus is actually the $3/2^-$ (g) state, a *binding* orbital of the π type, finally merging into the $p_{3/2}$ state in the limit of the united nucleus. All the other peculiarities in the level scheme of the system as a function of the distance can be inferred from the correlation diagram.

The insurgence of molecular-like structures in light nuclei was also clearly observed with Antisymmetrized Molecular Dynamics calculations; these models have the strong advantage of being able to reproduce both shell model-like states and cluster states in the same framework [88, 89]. As a guideline, it is useful to take inspiration from the orbital schemes and density distributions obtained with AMD calculations in Ref. [90] for the cluster structure in ${}^{10}\text{Be}$. These are, indeed, extremely useful to facilitate the approach of this matter considering the analogies with the oriented valence theory discussed in Section 1.1.4.2.

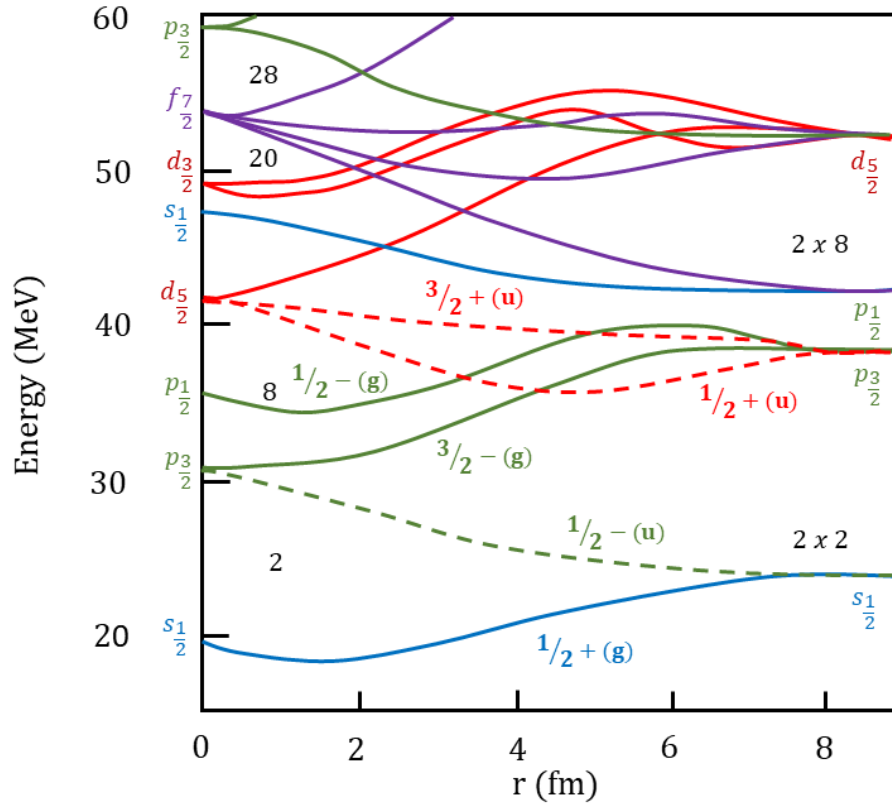


FIGURE 1.42: Correlation diagram showing the level sequence for the orbitals obtained through a two-center shell model approach, related to the Nilsson diagram for a deformed nucleus. For the s and p levels (on the basis of the right axis), solid lines represent *gerade* states, while dashed lines are *ungerade* levels. Adapted from Refs. [85, 87]. For a comparison, the reader is referred to the correlation diagram for the H_2^+ ionized molecule of Figure 1.9.

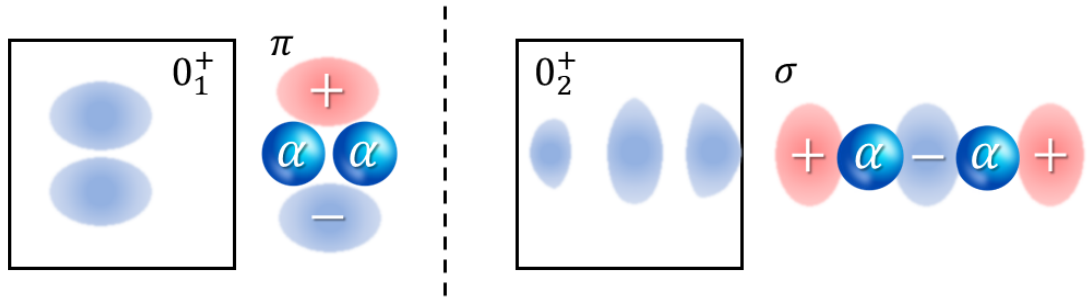


FIGURE 1.43: Density distributions of the single-particle wavefunctions in the first (left) and second (right) 0^+ excited states, envisaged as a $2\alpha + 2n$ structures, following AMD calculations. The spheres and cloud representations at the sides of the density calculations represent the distribution of the π and σ binding orbitals around the α cores. Adapted from [90].

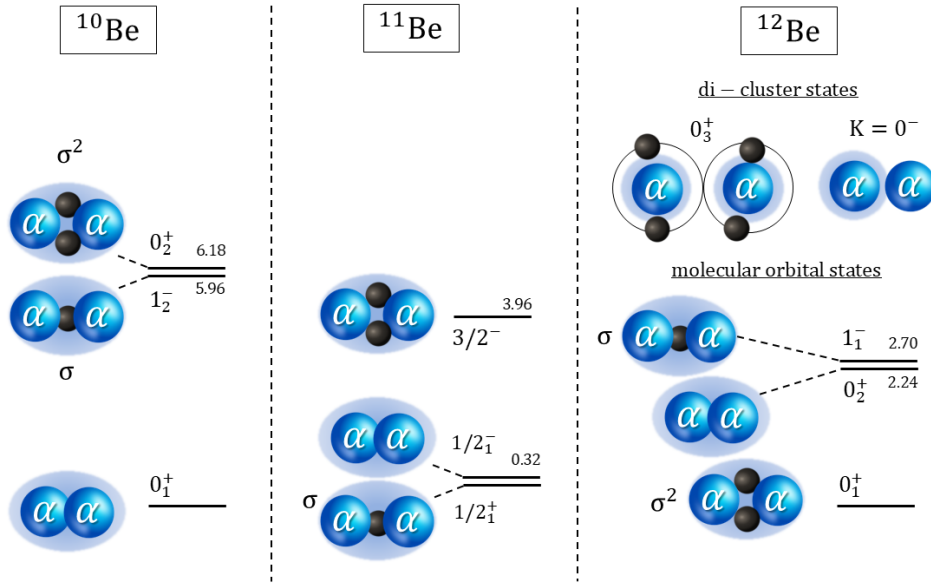


FIGURE 1.44: Schematic suggestion of cluster states in the level schemes of the $A = 10, 11, 12$ isotopes of Beryllium. The 2α cores and the valence neutrons in the σ orbitals are highlighted with black dots in the molecular orbital states; neutrons occupying the π orbitals are not explicitly highlighted for clarity reasons. Adapted from [90].

Like the molecular orbitals in "ordinary" molecules are formed by the combination of atomic orbitals, those in *nuclear* molecules (in 2α systems, in this specific case) are born from a linear combination of the neutron's p orbits around the two α -like particles. The overlap of these p orbits may give birth to different molecular orbitals, and this is nothing new, since it has been shown for different molecular systems in Section 1.1.4.2. The negative-parity offspring is the so-called " π orbital", schematically produced by a side overlap of the p orbitals, while their longitudinal superimposition sets up the positive parity σ orbital. Figure 1.43 shows how the σ orbitals has two nodes along the $\alpha - \alpha$ axis, gaining kinetic energy proportionally to the development of the 2α character of the nucleus, resulting in the presence of ground state configurations, for nuclei like $^{11,12}\text{Be}$, very different from those predicted by the shell model. In fact, on its basis, a nucleus with eight neutrons would be well bound and almost spherical, having a neutron magic number $N = 8$; however, this rule is broken by the strongly clustered 2α structure exhibited by the ^{12}Be nucleus, which is highly deformed and not stable.

In the past, beryllium isotopes have been extensively studied through the application of different theoretical models, like the *cluster* and the *molecular orbital* ones, which make an *a priori* assumption about the 2α configuration for these nuclei, or AMD calculations. The latter does not assume the existence of any type of cluster cores, only to actually confirm their presence in low-lying states of the various isotopes as the result of the calculations. Through the analysis of the single-particle wave functions, the AMD model was able to find associations between the orbits of the valence neutrons and the molecular orbitals developed around the 2α core. The results of these association, i.e., the suggested cluster states in ^{10}Be , ^{11}Be and ^{12}Be , are reported in Figure 1.44; the schemes also show the number of neutrons residing in the σ -like orbitals (which are dominated by positive parity components), alongside their respective excitation energies. Taking the ^{10}Be nucleus (left) as an example, the 0_1^+ , 1_2^- and 0_2^+ states can be tagged

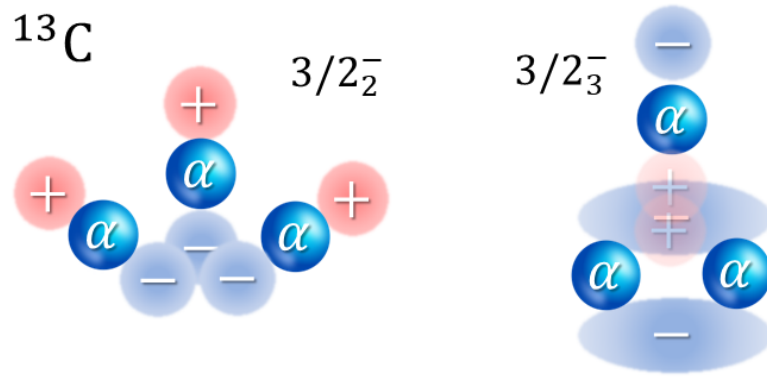


FIGURE 1.45: Sketched distributions of the single-particle molecular orbitals of the valence neutron around the 3α structure of the ^{13}C nucleus for its $3/2_2^-$ and $3/2_3^-$ states. Adapted from [92].

as π^2 , $\pi\sigma$ and σ^2 cluster configurations, where the superscript indicates the number of neutrons in each type of orbital. The spatial distributions of the single-particle orbits of the valence neutrons in the respective orbitals have been shown in more details in Figure 1.43, where the structures of the π - and σ -like orbitals are explicitly developed.

Similarly, the three states reported for the ^{11}Be nucleus (middle) respectively correspond to $\pi^2\sigma$, π^3 and $\pi\sigma^2$ configurations of three valence neutrons, while the four neutrons in the ^{12}Be nucleus (right) occupy $\pi^2\sigma^2$, π^4 and $\pi^3\sigma$ configurations. One interesting outcome of this discussion is that the development of the 2α cluster structure of these nuclei is strongly proportional to the number of valence neutrons in the σ orbitals: the more neutrons are in this type of molecular orbitals, the higher is the degree of 2α -clustering in the structure. The σ orbital is in fact decreasing its single-particle energy upon addition of valence neutrons, as a result of the kinetic energy gain with the 2α particles moving further apart; on the other hand, the addition of neutrons in the π orbitals, given their spatial characteristics, tends to soften the cluster properties of the structure. This is also at the basis of the breaking of the $N = 8$ magic number for the neutron shell ^{12}Be , and thus of its unstable nature [90]. These characteristics are furtherly exaggerated in highly-excited states of ^{12}Be , whose structures may even be described by $^6\text{He} + ^6\text{He}$ or $^8\text{He} + \alpha$ configurations.

The *icing on the cake* on the cluster configurations of neutron-rich beryllium isotopes comes anyway from a *household* observation on the possible rotational bands in which the excited states of such nuclei can be classified. For example, in Ref. [91] all these bands have been fitted with a formula of the type of Equation 1.47; despite few cases with lower inertial parameters, the largest part of such rotational bands have inertial parameters close to 0.5 MeV, i.e. similar to the ^8Be ground-state band, that is fully characterized by a dimeric cluster structure. This finding qualitatively confirms the covalent role played by the extra-neutron in binding together the two α centers.

As a concluding mention, similar discussions have also been reported on the $3\alpha + n$ structure of the ^{13}C nucleus by N. Furutachi and M. Kimura [92], who regard the structure of some states of this nucleus as a bent linear chain of three α particles, with one neutron having the binding role. This is mainly the case of the rotational band built on the $3/2_2^-$ state at 11.4 MeV, whose bent chain structure is particularly stable with respect to the bending motion of the three cluster. Differently from what happens

in some excited states of self-conjugate nuclei, like the Hoyle state of ^{12}C at 7.65 MeV, which were originally thought to be represented by a chain-like structure, but are now more commonly described as a diluted gas-like or a triangular 3α configurations [93, 94], this type of bent structure in ^{13}C is actually stabilized by the presence of the spare neutron. The molecular orbital distributions of the states in which the 3α character is more evident are pictorially described in Figure 1.45, on the basis of the respective wave function density distributions (see Ref. [92] for more details), showing the evidence of the bent-chain structure for the $3/2_2^-$ state (left) on which a stable rotational band is built. In this rotational band, in fact, the presence of the valence neutron gives rise to an almost pure π orbital, shortening the $\alpha - \alpha$ distance and making it orthogonal to the ground state band; this would hinder the bending motion of the clusters and enhancing the stability of the linear chain configuration. At variance, a much more compact configuration is seen for states having the $3/2_3^-$ state as band-head; for this state, the calculations shows that different 3α configurations are mixed, leading to a difficult interpretation based simply on a molecular-orbital framework (Figure 1.45, right panel).

Several other nuclear molecules built by covalent neutron(s) on $N\alpha$ clustered structures have been also suggested for other beryllium, carbon, oxygen and neon isotopes. A review of recent investigations on such nuclei can be found in Ref. [95].

1.2.3.8 Symmetries and excitations in self-conjugate nuclei

*Tyger, Tyger, burning bright
in the forests of the night
what immortal hand or eye
dare frame thy fearful
symmetry?*
W. BLAKE, *The Tyger*, 1-5

In recent times, a renewed interest was observed in the theoretical nuclear physics community towards the description of clustered nuclei, both in self- and non self-conjugate cases, starting from the *symmetries* characterizing the molecular-like alpha structures. Some interesting results are described, e.g., in Refs. [72, 94, 96–99] and references therein. This intensive use of symmetry to unveil the properties of the roto-vibrational excitation of a quantum system is another example of a strong analogy coming from the chemical realm: as discussed in Section 1.1.5, also in the case of atomic molecules (as the $^3\text{H}^+$), the interpretation of the various branches of Raman spectra was enormously simplified by symmetry considerations.

This Section will present a connection between the structure of the wavefunctions derived from the LCNO models (with the full advantage of considering a peculiar symmetric arrangement of the cluster centers) and the ordinary nuclear wavefunctions, expressed as eigenstates of parity and angular momentum. In other words, more close to the experimentalists' vantage points, it is interesting to understand which spin and parities could be observed in the roto-vibrational motion of a molecular structure characterized by a given symmetric arrangement. This procedure can be performed by using the *theory of projection* introduced in 1957 in a seminal work by Sir Rudolf Peierls and Jean Yoccoz [100]. All the details of this projection procedure are quite complicated, and will

be described in the following in a very schematic and simplified way, derived from Ref. [79].

Let's start by considering, as a prototype example, a *model* wavefunction ϕ coming from the LCNO model. ϕ has surely several symmetries coming from the spatial arrangement of clusters, but it is not in an eigenstate of J^π . Usually, the operators of a symmetry that is *broken* inside a nuclear model (as the LCNO) are represented by operators that are unitary and form a group (continuous or finite); such operators can be denoted as $U(\beta)$, where β is a symbol indicating the *group parameters*. For example, in the case of parity symmetry (that is typically broken by LCNO wavefunctions), the group is made by only two elements $\{1, P\}$, being P the parity operator. Another example: in the case of a rotational symmetry, the group is made by the rotation operators $R(\theta_1, \theta_2, \theta_3) = e^{-i\theta_1 J_x} e^{-i\theta_2 J_y} e^{-i\theta_3 J_z}$, being $\theta_1, \theta_2, \theta_3$ the Euler angles and J_x, J_y, J_z the angular momentum operators.

Because of the involved symmetry, $U(\beta)$ commutes with the total Hamiltonian H of the system: therefore, any wavefunction of the type $U(\beta)\phi$ has the same energy expectation value of ϕ :

$$\langle U(\beta)\phi | H | U(\beta)\phi \rangle = \langle \phi | H | \phi \rangle \quad (1.61)$$

The $U(\beta)\phi$ are then *degenerate* in energy, and this allows to build a *better* (in the sense of the purpose of finding states with given J^π values) wavefunction by a linear combination of the $U(\beta)\phi$. As a first step one can consider a functional space F spanned by $U(\beta)\phi$, i.e. the functional space made by all types of linear combinations of $U(\beta)\phi$. When diagonalizing the Hamiltonian H inside this functional space F , it is possible to obtain the best wavefunction achievable with the linear combination of $U(\beta)\phi$. If one indicates with ϕ_k an orthonormal basis wavefunction of the F space, the diagonalization of H in F can be performed by diagonalizing the matrix having the generic elements $\langle \phi_k | H | \phi_{k'} \rangle$. To solve this problem, one must consider the key point that the F space can be decomposed into the direct sum of smaller sub-spaces, each one being the self-space of some irreducible representation of the $\{U(\beta)\}$ group. As an example, when dealing with the rotational symmetry, the space F can be decomposed as:

$$\begin{aligned} F = \{ \phi_{J_1 M_1}, M_1 = -J_1 \div J_1 \} \oplus \\ \{ \phi_{J_2 M_2}, M_2 = -J_2 \div J_2 \} \oplus \\ \{ \phi_{J_3 M_3}, M_3 = -J_3 \div J_3 \} \oplus \dots \quad (1.62) \end{aligned}$$

In this way, the diagonalization of H in the F space is equivalent to the diagonalization of H within each sub-space of irreducible representation, and this means that the best wavefunction obtained from the diagonalization procedure would restore the symmetry that was originally lost in the *model* wavefunction. For example, in the case of rotational symmetry, the best wavefunction obtained in F with the above mentioned procedure would have a definite value of angular momentum J . The same obviously holds also in the case of parity symmetry.

Projection of parity. Let's assume that the model wavefunction ϕ is not in an eigenstate of parity. Since for the parity symmetry the group is composed by the elements $\{1, P\}$, one must minimize the energy by considering linear combinations of ϕ and $P\phi$ wavefunctions, a procedure equivalent to the diagonalization of the Hamiltonian in the

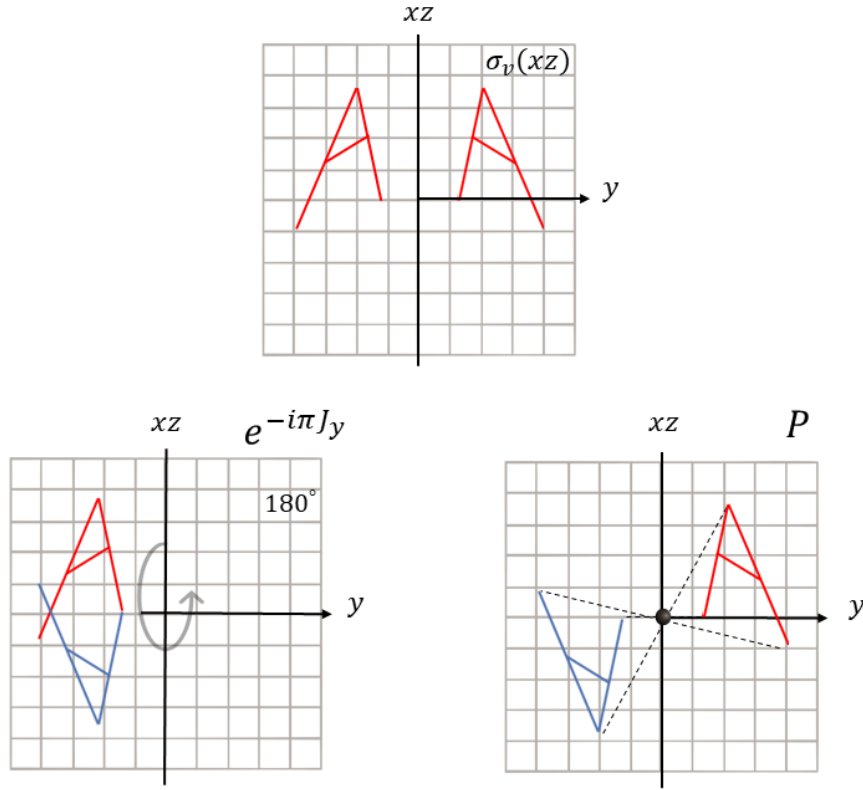


FIGURE 1.46: Visual representations of three different symmetry operations: (Top) reflection about the xz plane; (Bottom left): 180° rotation about the y axis; (Bottom right): the parity operation, i.e., an inversion of the coordinates of the object. To highlight the fact that the reflection operation is a combination of the last two, the initial and final conditions of the object are represented in red, while the intermediate state is reported in blue.

functional space F made by all types of linear combinations of ϕ and $P\phi$. With a simple algebraic trick, the F space can be also fully covered by the linear combinations of $(1 + P)\phi$ and $(1 - P)\phi$ wavefunctions, since:

$$c_1\phi + c_2P\phi = \frac{c_1 + c_2}{2}(1 + P)\phi + \frac{c_1 - c_2}{2}(1 - P)\phi = b_1(1 + P)\phi + b_2(1 - P)\phi \quad (1.63)$$

but this last expression has the enormous advantage that $(1 + P)\phi$ has *positive* parity and $(1 - P)\phi$ has *negative* parity. For this reason it is possible to denote:

$$\phi^{(\pm)} = n^{(\pm)}(1 \pm P)\phi \quad (1.64)$$

being $n^{(\pm)}$ a normalization constant. Now, according to the Peierls-Yoccoz procedure, one must diagonalize H within F , that is spanned by linear combination of $\phi^{(\pm)}$ functions. But, since a nuclear state has a definite parity, the matrix element $\langle \phi^{(+)} | H | \phi^{(-)} \rangle$ is always zero, and then the diagonalization does not mix the $\phi^{(+)}$ and $\phi^{(-)}$ functions (as obvious from their behavior). This means that the best wave functions obtained from the diagonalization procedure ($\phi^{(+)}$ and $\phi^{(-)}$) restore the parity symmetry originally lost in the ϕ model wavefunction. It can be didactically useful to demonstrate that the operators $\frac{1 \pm P}{2}$ are indeed *projection* operators (from which it descends the *name* to the Peierls-Yoccoz procedure): considering that $P^2 = 1$, it is

immediate to verify that $(\frac{1 \pm P}{2})^2 = \frac{1 \pm P}{2}$.

Projection of angular momentum. Also in this case, it is assumed that the model wavefunction ϕ is not an eigenstate of the angular momentum. A linear combination of $R(\theta_1, \theta_2, \theta_3)\phi$ is needed to minimize the energy by a diagonalization of the Hamiltonian within the F space made by all types of linear combinations of $R(\theta_1, \theta_2, \theta_3)\phi$. In this case, it is not simple to find the expression of a general wavefunction ψ of the space F that can be related to the rotation group parameters (i.e. the Euler angles). The use of the Wigner $D_{MK}^J(\theta_1, \theta_2, \theta_3)$ functions allows to solve this complicated problem [79, 101], leading to an expression of ψ as a linear combination of the type:

$$\psi = \sum_{JKM} c_{JKM} (P_{MK}^J \phi) \quad (1.65)$$

where the angular momentum projection operator P_{MK}^J is defined as:

$$P_{MK}^J = \int_{\text{all space}} \overline{D_{MK}^J} R(\theta_1, \theta_2, \theta_3) d\theta_1 d\theta_2 d\theta_3 \quad (1.66)$$

It is possible to demonstrate [79] that the projection operator P_{MK}^J is able to generate, from any *model* wavefunction ϕ , an eigen-state of angular momentum with magnitude J and third component M . By converse, this property leads to the fact that the diagonalization procedure involving $P_{MK}^J \phi$ as basis functions that generate the F space leads to a best wave function having a *definite* angular momentum value.

Effect of various symmetries on relevant quantum numbers. It is now possible to mix the information on parity and intrinsic angular momentum projection K of a nucleus by defining a wavefunction of the type:

$$\phi^{(\pm)}(K) \equiv \int_0^{2\pi} e^{iK\theta} e^{-iJ_z\theta} \phi^{(\pm)} d\theta \quad (1.67)$$

After several steps, that are omitted for brevity (see Ref. [79]), one can demonstrate that $\phi^{(\pm)}(K)$ are the eigen-functions of the J_z operator:

$$J_z \phi^{(\pm)}(K) = K \phi^{(\pm)}(K) \quad (1.68)$$

Starting from this result, it is possible to find some important limitations to the quantum numbers characterizing an excited state of a given nucleus obeying to a peculiar geometrical symmetry within a given model (as the LCNO, for example).

The first case that it is discussed is the **invariance for rotation of an angle $2\pi/N$ around the z-axis**. The model wavefunction would be of the type:

$$\phi = e^{-i\frac{2\pi}{N} J_z} \phi \quad (1.69)$$

This simple symmetry consideration on the *model* wavefunction would put strict constraints on the K values that a nuclear state can assume. In fact, by using the Equation

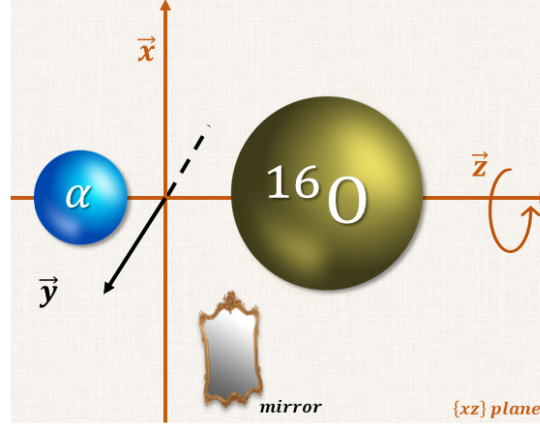


FIGURE 1.47: An example of symmetries involved in a di-cluster system as the $\alpha + {}^{16}\text{O}$ configuration in ${}^{20}\text{Ne}$.

1.69:

$$\begin{aligned} \phi^{(\pm)}(K) &= \int_0^{2\pi} e^{iK\theta} e^{-iJ_z\theta} \phi^{(\pm)} d\theta = \int_0^{2\pi} e^{iK\theta} e^{-iJ_z\theta} e^{-i\frac{2\pi}{N}J_z} \phi^{(\pm)} d\theta \\ &= \int_0^{2\pi} e^{iK\theta} e^{-iJ_z\theta} e^{-i\frac{2\pi}{N}K} \phi^{(\pm)} d\theta = e^{-i\frac{2\pi}{N}K} \int_0^{2\pi} e^{iK\theta} e^{-iJ_z\theta} \phi^{(\pm)} d\theta \quad (1.70) \end{aligned}$$

leading to the relation:

$$\phi^{(\pm)}(K) = e^{-i\frac{2\pi}{N}K} \phi^{(\pm)}(K) \quad (1.71)$$

This recursive relation implies that $\phi^{(\pm)}(K)$ is always *zero*, unless $e^{-i\frac{2\pi}{N}K} = 1$, that occurs for $K = 0, \pm N, \pm 2N, \dots$ values. This means that, in presence of this discrete symmetry, the angular momentum eigen-state wavefunction are different from zero only in the following case:

$$P_{MK}^J[\phi^{(\pm)}] \neq 0 \rightarrow K = 0, \pm N, \pm 2N, \dots \quad (1.72)$$

A simple generalization of the previous example involves the **axial symmetry around the z-axis**; in this case the model wavefunction has the form:

$$\phi = e^{-i\theta J_z} \phi \quad (1.73)$$

for any θ value. This would immediately remove the N dependence in the constraint on the K value, leading to:

$$P_{MK}^J[\phi^{(\pm)}] \neq 0 \rightarrow K = 0 \quad (1.74)$$

Now, a brief discussion of the last important case of symmetry, the **invariance for reflection on the (xz)-plane**, often indicated as $\sigma_v(xz)$. As immediately evident from Figure 1.46, this type of reflection can be decomposed into a rotation of 180° around the y-axis, followed by a *parity* operation. The symmetry of the model wavefunction would then assume the form:

$$\phi = \sigma_v(xz)\phi = e^{-i\pi J_y} P \phi \quad (1.75)$$

After some passages [79], it can be demonstrated that this leads to the equivalence:

$$P_{M-K}^J[\phi^{(\pm)}] = \pm(-1)^{J-K} P_{MK}^J[\phi^{(\pm)}] \quad (1.76)$$

that, in the important (symmetric) case $K = 0$ leads to:

$$P_{M0}^J[\phi^{(\pm)}] = \pm(-1)^J P_{M0}^J[\phi^{(\pm)}] \quad (1.77)$$

This recursive formula immediately imposes the following constraints on the quantum numbers of a nuclear state characterized by this reflection symmetry:

$$\begin{aligned} P_{M0}^J[\phi^{(+)}] \neq 0 &\rightarrow \text{even } J \\ P_{M0}^J[\phi^{(-)}] \neq 0 &\rightarrow \text{odd } J \end{aligned}$$

and this will introduce a strict correlation between the parity and angular momentum of the wavefunction. All the considerations here discussed will be of fundamental importance to predict the *sequence* of J^π states expected from excitations of clustered nuclei with a given symmetric arrangement of α particles, a precious information for the experimental research on clustering configuration of self-conjugate nuclei.

The case of di-cluster systems. Let's now consider a nuclear *dimer*, constituting a system made by two clusters of *different* type, as the $\alpha + {}^{16}\text{O}$ configuration in ${}^{20}\text{Ne}$. As shown in Figure 1.47, the model wavefunction ϕ is axially symmetric around the z -axis and is also invariant for reflection with respect to the (xz) plane. By a combination of the discussions on symmetries made in the previous Sections, it is evident that the projections $P_{MK}^J[\phi^{(\pm)}]$ are different from zero only for $K = 0$, and that with this constraint the parity must satisfy the rule $\pi = (-1)^J$. This means that the expected J^π values for ground or excited states nuclear molecules with such dumbbell symmetry are of *natural parity*, being:

$$\begin{aligned} J^\pi &= 0^+, 2^+, 4^+, 6^+ \dots \\ J^\pi &= 1^-, 3^-, 5^-, 7^- \dots \end{aligned}$$

as beautifully seen, experimentally, for the $K = 0$ positive and negative parity rotational bands already shown in Figure 1.39 for $\alpha + {}^{12}\text{C}$ configuration in ${}^{16}\text{O}$ and $\alpha + {}^{16}\text{O}$ configuration in ${}^{20}\text{Ne}$.

Finally, it is worth noting that for dimeric structures made by two *identical spinless* clusters, the intrinsic wave function must have a definite positive parity and therefore, in this special case, only $J^\pi = 0^+, 2^+, 4^+, 6^+ \dots$ states are allowed.

The case of systems with D_{3h} symmetry. The D_{3h} symmetries due to an equilateral triangle configuration of identical clusters have been already discussed in Section 1.2.3.6, in the context of the LCNO model. It is thus possible to derive the constraints due to the involved symmetry operations, sketched for convenience in Figure 1.48. The invariance of ϕ against a rotation of $\frac{2\pi}{3}$ around the z -axis leads to non-vanishing projections $P_{MK}^J[\phi^{(\pm)}]$ only for:

$$K = 0, \pm 3, \pm 6 \dots \quad (1.78)$$

Now, the invariance of ϕ against the rotation of 180° with respect to the y -axis can be treated with a technique similar to the cases discussed in the previous Section, leading to the recursive relation $P_{M-K}^J[\phi^{(\pm)}] = (-1)^{J-K} P_{MK}^J[\phi^{(\pm)}]$. For the $K = 0$ case (that is present in the list 1.78) this immediately leads to the need of $J = \text{even}$ values to have

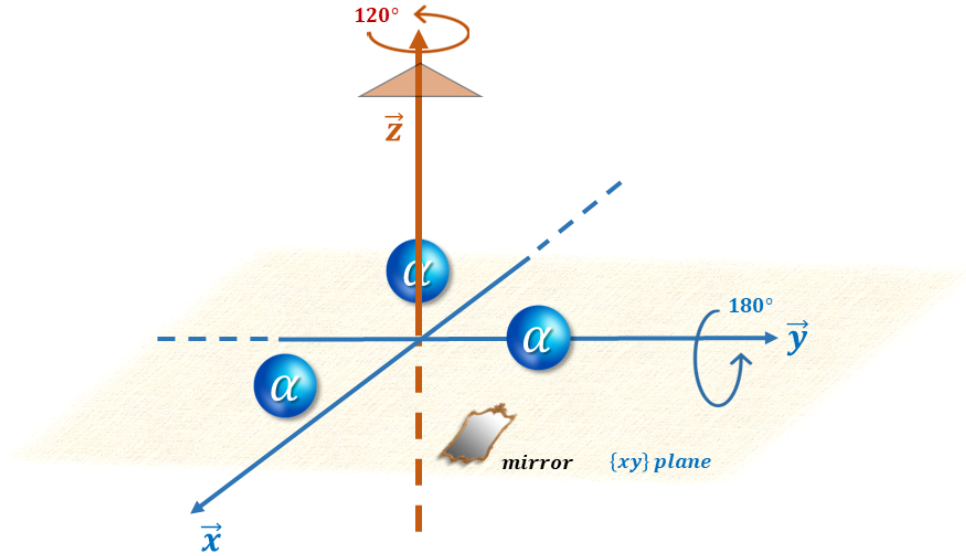


FIGURE 1.48: An example of symmetries involved in a trimeric system provided with a D_{3h} equilateral triangular symmetry, as recently suggested for the 3α configuration in ^{12}C .

a non-vanishing projection $P_{M0}^J[\phi(\pm)]$. Finally, the symmetry for reflection around the horizontal (xy) plane, $\sigma_h(xy)$, can be treated in a way similar to the $\sigma_v(xz)$ vertical case seen before, leading to non-vanishing projections $P_{MK}^J[\phi(\pm)]$ only for parity values $\pi = (-1)^K$. Putting all the constraints on the quantum number here discussed into combination, it is finally found that the only possible J^π values for ground and excited states coming from the excitation of an equilateral triangular structure are the following:

$$J^\pi = 0^+, 2^+, 4^+, 6^+ \dots$$

$$J^\pi = 3^-, 4^-, 5^-, 6^- \dots$$

$$J^\pi = 6^+, 7^+, 8^+, 9^+ \dots$$

By grouping these states together, and considering the energies expected by a classical rotator, it is possible to predict a very peculiar sequence of rotational levels of the type:

$$J^\pi = 0^+, 2^+, 3^-, 4^\pm, 5^-, \dots \quad (1.79)$$

The discovery of some of these members in the rotational bands built on ^{12}C ground and Hoyle states has indeed been suggested in Ref. [94] as the *smoking gun* proof of the equilateral triangular α cluster structure of such states. In such a case, the Algebraic Cluster Model (ACM) predictions, based on symmetry consideration and on the Group Theory, were used as a guideline for the interpretation of experimental data. It is worth noting that the experimental *hunt* towards the missing members of the rotational (and vibrational) states predicted in ^{12}C by the D_{3h} symmetry is still open and will lead to interesting consequences in the next decades.

1.3 Some final remarks

All the features presented in this first Chapter represent important analogies between the chemical and the physical world, as firstly suggested by George Gamow and Charles Critchfield in their textbook [38]: given the limited knowledge available about the nuclear force, it is useful to study the macroscopic properties of the nuclei through chemical-like approaches, exploiting the analogies and avoiding, at first impact, to build the structures of the nuclei starting from *ab-initio* methods. This allowed, even quantitatively, to understand (for example) the behavior of long-range residual correlations, responsible of the presence of highly pronounced nuclear deformations and/or clustering effects. Furthermore, it is not a coincidence that only nuclear physics has a so large gamma of models to describe its phenomenology (see Table 1.3): important details of the nuclear force are still unknown, but the exploitation of radioactive beams, allowing the study of nuclei far from the stability valley is of course enriching the phenomenology.

Another interesting analogy with the chemical framework is that of the intensive usage of symmetries: the regular geometric configurations of clusters impose some restrictions on the quantum numbers of nuclear states, so to allow to unveil important information on the nuclear structures starting from the sequences of rotational and vibrational states. It also has to be said that, in recent times, thanks to the enormous availabilities of computational power, new *ab-initio* models are developing, starting from complicate expressions of effective forces between the nucleons (like the *Argonne-V18*), aiming to the reproduction of the lightest nuclei spectroscopy. Albeit a good agreement with experimental data has been found, several aspects (like the presence of largely α -clustered high excitation energy states in self-conjugate nuclei) are still not fully understood, and this is another reason for which the experimental study of their characteristics becomes really interesting.

The main subject of this thesis is therefore based on new sets of experimental data aiming at investigate the structure of various self-conjugate nuclei (as ^{12}C , ^{16}O , ^{20}Ne). The different techniques used to treat and study these data will be reported, in details, in the next Chapter. As discussed in this Chapter, very strong connections with models and techniques coming from the chemical and molecular physics worlds will be exploited all along this work. Finally, *for the sake of symmetry*, the thesis will be completed by a Chapter describing the experimental results of a chemistry experiment in which the possible occurrence of clustering in atomic molecules is investigated by means of nuclear physics techniques.

Chapter 2

Some techniques for the study of clustering in nuclei and molecules

The main body of this thesis has a heavy experimental character; however, an introductory overview about the different techniques useful for the study of clustering is mandatory to have all the necessary instruments to analyze the experimental data. Through the application of different investigation techniques with physical and chemical origins, some open questions of clustering in the nuclear and molecular world will (hopefully) be addressed.

2.1 Coherence effects in quantum scattering

It has been shown how this world is often arranged in clustered structures, whose geometrical and symmetry properties often determine the structure and the dynamical evolution of the whole system. The most direct way to investigate the nature of these properties is thus to "have a look" at the structure itself; this is not only important in the field of materials chemistry, but also in the biochemical and biomedical fields. As every reader could recall, in fact, one of the most ground-breaking discoveries related to the observation of the structure of a macromolecule is due to R. Franklin and her student, R. Gosling [102], who produced the famous "Photo 51" in 1952, reporting the first x-ray diffraction pattern of the DNA molecule.

Such a technique for the deep study of the molecular structure is based on the *interference* properties of interacting waves. These can be destructive, when the peaks of the first wave coincide with the hollows of the second one, or constructive, when the two waves have the same phase and peaks and hollows of the first wave coincide with those of the second one. Through the study of constructive and destructive interference regions, having a suitable screen or detector, it is possible to find the pattern of low and high intensities of the signal. When an object is found in the path of a wave, this will interfere with it and modify its pattern, as a result of a phenomenon called "*diffraction*", which occurs when the size or a particular dimensional parameter of the obstacle is comparable with the wavelength of the radiation. On the basis of this, it is possible to study the structures of crystals and molecules by using radiations of a suitable wavelength λ .

2.1.1 X-ray diffraction of molecules

To give some insights on the history and the basis of the technique of X-ray diffraction for the study of crystal structures, it was developed across the end of the XIX and the beginning of the XX century, following the discovery of X-rays by W. Röntgen [103] in 1895. X-rays are electromagnetic radiations, usually generated by the bombardment of metal targets with highly energetic electrons, with a wavelength of around $\lambda \approx 10^{-10}$ m; the suggestion of the possible diffraction of X-rays after their passage through the lattice planes of a crystal was proposed by M. von Laue in 1912 [104]. This suggestion was based on the fact that the average distance between lattice planes in a crystal is of some ångström (Å) perfectly suitable for the diffraction of X-rays; the diffraction pattern, i.e., the alternation and the intensity of the interference minima and maxima will thus give information on the distribution, distance and orientation of the lattice planes in the crystal. The original von Laue's method was based on the use of a broad-band X-ray beam passing through a single crystal, with a screen which could be photographically impressed to record the diffraction pattern. The basis of the idea was the fact that a single crystal may of course not be suitably oriented for the diffraction of a specific wavelength of X-rays, but upon the utilization of a broad-band beam, it would have been possible to obtain diffraction for at least one of the λ s of the beam. A couple of years later, in 1914, Paul Scherrer and Peter Debye developed the so-called "*Debye-Scherrer powder method*" [105], which is exactly the opposite of von Laue's method: when using a monochromatic radiation, if the sample is a powder (and not a single crystal), some of the crystallites will be oriented in a suitable way to produce diffraction for the specific λ used. Data obtained from powder diffraction experiments are, moreover, useful to extract information about the phase diagram of a given substance, since different solid phases (allotropes) produce different diffraction patterns.

Through a simple application of the X-rays diffraction laws on a polyatomic molecules gas (therefore, characterized by casual spatial orientations) it is possible to immediately reach an analytic form for the intensity of the diffraction pattern, averaged over all the available molecular orientations. This intensity curve is usually referred to as *Wierl* equation, and is expressed as in Ref. [106]:

$$I(\theta) = N \left[\sum_i f_i^2 + 2 \sum_{i,j} f_i f_j \frac{\sin(\mu r_{ij})}{\mu r_{ij}} \right] \quad (2.1)$$

where N is the number of molecules, f_i represents the atomic form factor for the i -th encountered atom (and is a function of the scattering angle, θ , and of the charge distribution of the electronic shells), r_{ij} is the distance between the two atoms generating the matter wave interference, while $\mu = \frac{4\pi}{\lambda} \sin(\frac{\theta}{2})$ is called scattering vector and it is tied to the transferred momentum in the collision between a photon of the beam and a given atom. θ is the scattering angle in the laboratory frame, while λ is the wavelength of the used X-ray beam.

For the case of a gas of di-atomic homopolar molecules, the previous equation is strongly simplified, in the more familiar form [15, 107]:

$$I(\theta) = 2N f^2 \left[1 + \frac{\sin(\mu r)}{\mu r} \right] \quad (2.2)$$

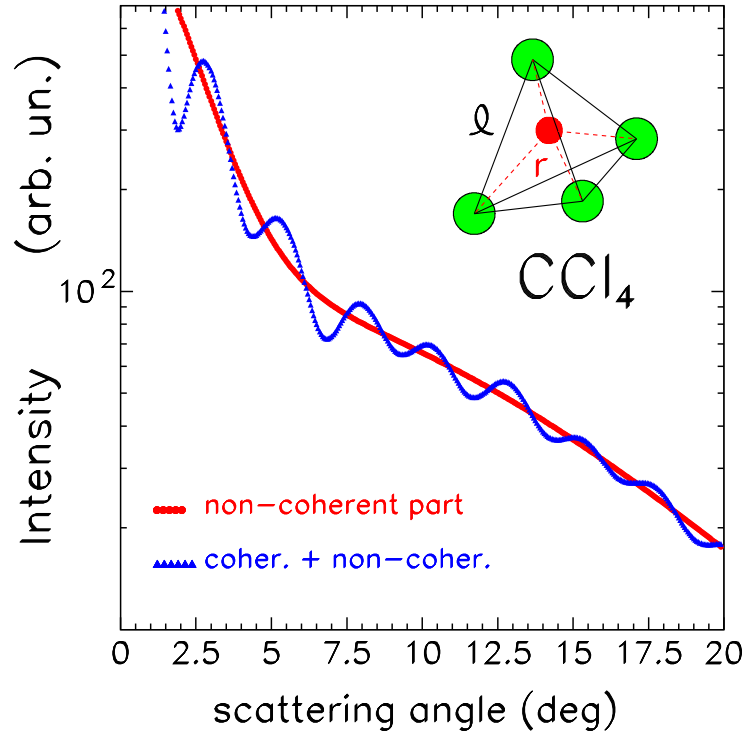


FIGURE 2.1: Calculation of the expected diffraction figure when a 100 keV x-ray beam impinges on a gas of carbon tetrachloride CCl_4 . Red circles: non-coherent part of scattering. Blue triangles: sum of coherent and non-coherent contributions.

A very instructive example is reported in the following: the diffraction of a 100 keV X-ray beam (with a wavelength of $\lambda = 0.123 \text{ \AA}$) on a polyatomic gas with a tetragonal structure, like carbon tetrachloride CCl_4 . The literature value of $r = 1.75 \text{ \AA}$ will be considered for the length of the C-Cl radial bond and $\ell = \sqrt{8/3} \cdot r \simeq 2.87 \text{ \AA}$ for the Cl-Cl bonds at the edges of the pyramid [12, 15]. The Wierl equation for the average molecular intensity thus becomes:

$$I(\theta) = f_C^2 + 4f_{Cl}^2 + 8f_C f_{Cl} \frac{\sin(\mu r)}{\mu r} + 12f_{Cl} f_{Cl} \frac{\sin(\mu \ell)}{\mu \ell} \quad (2.3)$$

where the atomic form factor behaviors, f_C and f_{Cl} , can be deduced from crystallographic tables in Ref. [108]; these are explicitly obtained through a theoretical adaptation of Thomas-Fermi atomic models, treated over a large database of experimental measurements of both atomic structure and scattering. From equation 2.3 it is possible to obtain the diffraction trend shown in Figure 2.1 (blue triangles); the expected trend in the absence of the diffraction-related coherent term is instead shown with red dots. The angular position and the relative amplitude of maxima and minima represents a characteristic pattern, strongly related to the interatomic distance in the molecule and to the specifically chosen geometry. The experimental data trends obtained by van der Grinten in 1933 [109] have proved to be in excellent agreement with the predictions of Equation 2.3, for the various adopted X-rays energies.

2.1.2 Electron diffraction of molecules

In more recent times, the structure of molecules has been studied through electron beam diffraction, with energies in the keV region; the first works around this matter are linked to the well-known Davisson and Germer studies (1927). A simple non-relativistic calculation about the De Broglie wavelength of an electron beam shows that $\lambda = \sqrt{\frac{h^2}{2m_e e \Delta V}} \simeq \sqrt{\frac{150}{\Delta V}} \text{ (\AA)}$, with the accelerating voltage expressed in volts. It is clear how just 150 Volts are enough to obtain wavelengths of the order of the $\text{\AA}$, while in the case of X-rays beams it is necessary to produce 12 kV electron beams to obtain a *brehmsstrahlung* of analogue wavelength [110]. This explains the great success in the exploitation of electron diffraction techniques for the study of molecular geometries, and the subsequent determination of the covalent bond lengths between atoms in molecules. The intensity of the diffraction figure is described through a formula of the same type of Equation 3.1, seen for the case of X-rays diffraction, but with the foresight of using the *electronic* form factors, dependent from both the electronic charge distributions and the nuclear charge, in place of the *atomic* ones.

2.1.3 Extension to the nuclear case

High-energy electron beams ($\approx 10^2 \text{ MeV}$) have wavelengths of the order of some femtometers (10^{-15} m), and have been widely exploited, after Robert Hofstadter pioneer studies [111], to investigate the form factors of nuclei and determine the trends of the nuclear charge densities. These electron scattering experiments are a powerful tool to probe the structure of spheric or *spheroidal* nuclei, but appear largely limited when trying to describe more exotic structures, like neutron halos or α -particles cluster.

Proton and neutron beams of suitable energies (several tens of MeV) could be quantum-mechanically represented by matter waves with $\lambda \approx 2\text{--}5 \text{ fm}$, and are thus exploitable for the study of nuclear diffraction phenomena. Historically, some analyses of this type have been conducted by Fernbach and collaborators [112] to characterize several nuclear radii, and constituted the basis on which the Optical Model for nuclear reactions was developed [113–115]. At the basis of the common Optical Model calculations is the assumption that the real and imaginary potentials, respectively describing the diffractive and *absorbent* part of the collision process, had spherical or spheroidal symmetry, thus not properly taking into account the possibility of formation of nuclear clusters in ground states.

On the other hand, differently from what happens in molecular physics, where the geometrical structure of the ground state is fixed by the quantum properties in play, in the nuclear case the formation of α particles clusters in the ground state is a dynamical process. This is, in fact, leading to the continuous formation and dissolution of the cluster themselves inside the mean nuclear field generated by all the nucleons. The fraction of time the system spends in a cluster configuration is tied to the spectroscopic factor of that precise configuration, and it typically is a small fraction (few percents, [116, 117]) with respect to the time the system spends in the spheroidal configuration. The two contributions, however, can manifest simultaneously in a scattering process: the matter wave could, indeed, encounter a common spheroidal nuclear obstacle, producing the usual diffractive phenomenon treatable with the Optical Model (or with more refined variants, like the Coupled Channels one [118]), or, for a limited fraction of events, encounter the target nucleus in its cluster configuration. In this case, the

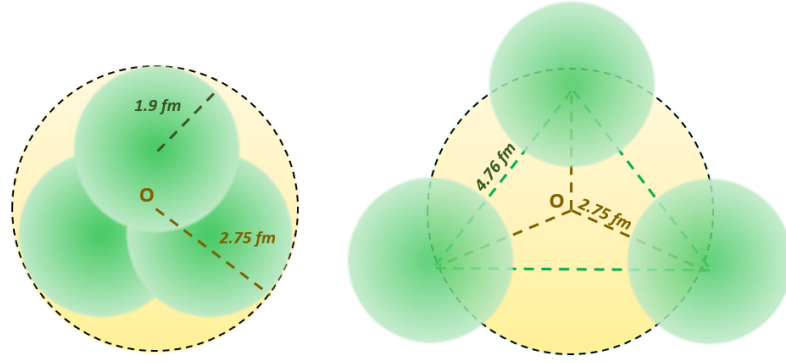


FIGURE 2.2: Packing of alpha particles that can be dynamically formed because of clustering. (left panel): tight packing (right panel): loose packing, with alpha particles forming at the surface of the ^{12}C nucleus.

Optical Model approach would be absolutely inadequate; instead, coherent scattering phenomena should be considered, held by a *Wierl-like* equation (3.1), already exploited in the molecular chemistry framework. In the nuclear case, however, the aforementioned atomic or electronic form factors should be swapped with the scattering amplitudes of the matter wave on the single cluster, i.e., an α particle. Both contributions, spheroidal and coherent, would come into play in the description of experimental data of proton-s/neutrons elastic scattering on clustered nuclei.

In this framework, it can be useful to calculate some geometric properties for some specific cases of self-conjugate nuclei. For the ^{12}C nucleus, one of the subject of this thesis, if $A = 12$ and $R_0 = 1.2$ fm are considered in the nuclear radii formula, $R = R_0 \cdot A^{1/3}$, the radius of the carbon nucleus would be $R \simeq 2.75$ fm. At the onset of α -cluster structure, the twelve nucleons playing inside the ^{12}C core could reorganize themselves to form three α particles, whose most symmetrical distribution would resemble an equilateral triangle. The (possible) relative position of the three cluster has been discussed in several theoretical works [119–121] by using various nuclear models (AMD, FMD, MCSM...); it is possible to derive some simple ideas on this by considering that a too-close spatial overlap of the matter distributions of clusters would lead to a sudden *dissolution* of their structure. This effect would occur, for example, by considering 3α particles internally tangent to a sphere of $R \simeq 2.75$ fm radius, considering that the average radius of an α particle is $R_\alpha \simeq 1.9$ fm. At variance, if the clusterization is considered to be essentially a *surface* phenomenon, it can be assumed that the centers of α clusters are placed symmetrically at the surface of a sphere of $R \simeq 2.75$ fm radius, leading to an estimate of the $\alpha - \alpha$ distance $\ell = R\sqrt{3} \approx 4.76$ fm. Within this assumption, essentially no overlap of nuclear matter between the different clusters should be observed.

In this schematic frame, a proton beam accelerated at energies between 30 and 80 MeV, would have the right associated de Broglie wavelengths of $\lambda \approx 3.5 - 5.7$ fm to unveil the presence of coherence effect due to the onset of clustering in the ^{12}C nucleus. These concepts, in the framework of suitable approximations which will be presented in the next Chapter, laid the basis for the development of a novel fitting method aimed to describe the $p+^{12}\text{C}$ elastic scattering cross section data, through the detailed coupling of optical model-like calculations and the coherent diffraction contribution given by the

α cluster structures. A similar preliminary analysis was also conducted on the (more limited) $p+^{16}\text{O}$ elastic scattering database reported in the literature.

2.2 Radiative branching ratio measurements

The decay process of a nuclear state is a *treasure trove* to probe the details of the wavefunctions of both the *parent* and the *daughter* nuclei. In fact, the time-dependent perturbation theory of Quantum Mechanics [4] allows to write the decay constant λ via the *second Fermi Golden Rule*:

$$\lambda = \frac{2\pi}{\hbar} < \varphi_f | H_{trans} | \varphi_i > \cdot \rho_f(E_{decay}) \quad (2.4)$$

Here, $\rho_f(E_{decay})$ is the density of final states accessible from the decay of the parent nucleus (and is "easily" calculable from phase space considerations), φ_i and φ_f are respectively the parent and daughter wavefunctions, and H_{trans} is the (perturbative) portion of the Hamiltonian that is responsible of the transition. It is evident that, if H_{trans} is known, the knowledge of λ from experimental data can *discriminate* among several (and often different) *theoretical* wavefunctions φ_i and φ_f obtained from different models: this is a fundamental tool to unveil the details of nuclear structure. All the types of nuclear decay (α , β , γ , fission...) can obviously be used in this framework.

Among them, the γ decay has the huge advantage that the transition operators are well known from the theory of electromagnetism [122]. As an interesting example for the cluster realm, the λ_γ decay constants of 2^+ and 4^+ excited states of ^8Be (expressed as transition strength $B(E2)$, as discussed later in this Section) were obtained from very refined radiative capture experiments performed by Datar and collaborators [123, 124]. These were compared with the cluster model and ab-initio calculations, and a nice agreement with the cluster model wavefunction was seen. However, there are some limitations in applying this methods to other nuclei. Since the excited states with the most interest to know the λ_γ are *unbound*, the most prominent decay is typically the *hadron* one (with the typical emission of a light nucleus as proton, a neutron, an α -particle etc), with branching ratios against *electromagnetic* decays often in the range $10^4 - 10^7$: the γ decay of an unbound state is an *extremely rare* process! Furthermore, the radiative capture cross sections on unbound states, which depend on the λ_γ , are typically *extremely* low (few hundreds of nb in the case of $\alpha + \alpha$ radiative capture of Refs. [123, 124]). If precise λ_γ (or $B(E2)$) values are needed to benchmark the outcomes of two different theories, very long-lasting (and refined) experiments are needed. In this thesis, an example of these investigations on the Hoyle state of ^{12}C will be discussed, for which the λ_γ value traditionally accepted in the literature was recently put into question by a new experiment [125]. It is worth noting that, in general, since the λ_γ enters in the theoretical expression of radiative capture cross sections, its knowledge is often a fundamental ingredient to determine the reaction rate of formation of new elements in stars [126, 127].

To introduce the concepts of determinations of decay constants λ_γ , half-lives $\tau_\gamma = \frac{1}{\lambda_\gamma}$, decay constants $\Gamma_\gamma = \frac{\hbar}{\tau_\gamma} = \hbar\lambda_\gamma$ and branching ratios, it is important to recall, beforehand, some classical features of oscillating assemblies of charges, generally referred to as

multipoles. It is possible to define an *electric* multipole moment as [128]:

$$Q_{l,m} = \int r^l Y_{l,m}^*(\theta, \phi) \rho d\tau \quad (2.5)$$

where Y_{lm}^* is the complex conjugate of the l - and m -order spherical harmonic function, ρ is the charge density and $d\tau$ is the volume element over which the integral of Q_{lm} is calculated. In this case, the integration region is the region of space occupied by the electrical charge, i.e., the whole atomic nucleus. On the basis of its order, the electric dipole moment has different components: for example, for $l = 1$, the three components $Q_{1,1}$, $Q_{1,0}$ and $Q_{1,-1}$ will be available, each one of them respectively related to the spherical harmonic with $l = 1$ and $m = 1, 0$ and -1 . The explicit forms of these components are reported in Ref. [128]. The textbook definition of the *dipole* moment is thus that of a vector with x components:

$$P_{Ex} = \int x \rho d\tau \quad (2.6)$$

yielding three components for the *dipole* moment. On the other hand, when going one step forward to the spherical harmonics of order $l = 2$, one obtains five linearly independent components of the so-called *quadrupole* moment, that often characterize the shape of ground and/or excited states in nuclei, see Section 1.2.3.3. Each of these five components has an expression similar to that of the $Q_{2,0}$ one, reported as an example:

$$Q_{2,0} = \sqrt{5/16\pi} \int r^2 (3 \cos^2 \theta - 1) \rho d\tau = \sqrt{5/16\pi} \int (3z^2 - r^2) \rho d\tau \quad (2.7)$$

where z is considered to be the symmetry axis of the nucleus. When the charge density of the nucleus has the symmetry of an ellipsoid of rotation around the symmetry of its z -axis, the other components of the quadrupole moment will be vanishing. In parallel to the electric multipole moments, it is possible to define, in a similar fashion, the magnetic multipoles; these are usually "intertwined" to the electric ones, and their expression is

$$M_{l,m} = - \int r^l Y_{l,m}^*(\theta, \phi) \cdot \frac{\text{div}(\vec{r} \times \vec{j})}{l+1} d\tau \quad (2.8)$$

where $\vec{j}$ is the electrical current density inside the system of charges. However, some simplifications may be made when applying the concepts of multipoles to the study of atomic nuclei: (i) a multipole moment of order l has an expectation value equal to *zero* for any nucleus with an angular momentum $J < l/2$; (ii) the electric dipole moment of any nucleus is equal to *zero*. Nevertheless, it is still possible to compute the radiated power of an oscillating multipole in a nuclear framework, given that the final quantum-mechanical formulas do not contain any electric or magnetic multipole moments, but only the *matrix* elements instead. Atomic nuclei, like the atoms themselves, despite not having any electric dipole moments, do emit electric dipole radiation, as predicted by the quantum theory of radiation and not describable through classical approaches.

As a result of the classical calculations of the power radiated from an oscillating system of charges, based upon the solution of Maxwell equations and the integration of the Poynting vector over the area surrounding the nucleus, it is possible to obtain the total

power radiated by an electric multipole Q_{lm} with angular frequency ω :

$$P_E(l, m) = \frac{2(l+1)e}{\epsilon_0 l [(2l+1)!!]^2} \left(\frac{\omega}{c}\right)^{2l+2} |Q_{l,m}|^2 \quad (2.9)$$

with an analogue expression for the case of the magnetic multipole. To give a more specific example, let's consider the case of a proton moving in a circular orbit of radius r : this will have a dipole moment equal to $\sqrt{3/8\pi}er$. Following the definition of Equation 2.9, the power irradiated when it moves with angular frequency ω_0 will lead to the familiar *Larmor formula*:

$$P_E(1, 1) = \frac{\omega_0^4 e^2 r^2}{6\pi\epsilon_0 c^3} = \frac{e^2 a^2}{6\pi\epsilon_0 c^3} \quad (2.10)$$

where $a = \omega_0^2 r$ is the acceleration of the proton in a circular motion. This radiated power is the reason of the failure of the classical atomic model in which the electron was pictured as simply orbiting around the nucleus: when the irradiation continues, the radius of the orbit should decrease constantly, until the collapse of the electron/proton to the center of the orbit. On the same basis as the Bohr model of the atom, it is however possible to assume that the proton moves in a harmonic potential, with its frequency staying constant; it will have a total mechanical energy of $E = m\omega_0^2 r^2$, so

$$\frac{dE}{dt} = -P_E(1, 1) = -\frac{\omega_0^2 e^2}{6\pi\epsilon_0 m c^3} E \quad (2.11)$$

After integration over t

$$E = E_0 e^{-\lambda t} \quad (2.12)$$

where the λ term is equal to

$$\lambda = \frac{\omega_0^2 e^2}{6\pi\epsilon_0 m c^3} = \frac{P_E(1, 1)}{E} \quad (2.13)$$

which is the constant regulating the classical exponential decay of the proton in a circular orbit; moreover, the angular momentum of the particle ($L = m\omega_0 r^2 = E_0/\omega_0$) fades away along with the energy decreasing to zero, carried by the radiation field.

The next step of this discussion will focus on the transition from the classical approach of the phenomenon to the quantum mechanical one, where the problem is solved, as indicated before, through the time-dependent perturbation theory, considering the electromagnetic interaction between the charges system and the quantized field inside a spherical volume. The results found in this approach are in agreement with the classical ones, but with two specific additional features, characteristic of quantum mechanical systems:

- The energy radiated by the particle or system is quantized as $E = \hbar\omega$. In this framework, the decay constant λ ¹, defined in Equation 2.13, represents the *number* of quanta emitted per unit time when the rate of energy emitted is defined through the classical expression. For electric and magnetic irradiation is thus possible to obtain

$$\lambda_E(l, m) = \frac{P_E(l, m)}{\hbar\omega} = \frac{2(l+1)}{\hbar\epsilon_0 l [(2l+1)!!]^2} \left(\frac{\omega}{c}\right)^{2l+1} |Q_{l,m}|^2 \quad (2.14)$$

¹Here, the γ subscript is omitted from the λ to unburden the notation, but the meaning is clear.

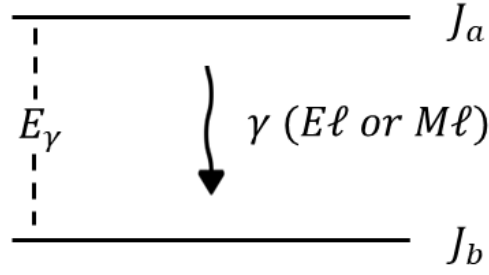


FIGURE 2.3: Electromagnetic transition involving the emission of a γ ray, between the states J_a and J_b .

$$\lambda_M(l, m) = \frac{P_M(l, m)}{\hbar\omega} = \frac{2(l+1)\mu_0}{\hbar l[(2l+1)!!]^2} \left(\frac{\omega}{c}\right)^{2l+1} |M_{l,m}|^2 \quad (2.15)$$

- The form of Equation 2.5 should be modified to replace the charge density ρ with a quantity more suitable to a quantum mechanical system, i.e., the unit charge multiplied for the probability density (ψ_a^2), considered for all the charged particles.
- Another important feature shown by the quantum theory of radiation is that the $Q_{l,m}$ term in Equation 2.14 is not *simply* the multipole moment element in the initial state (ψ_a) but, rather, the *multipole matrix element* between the initial and final states, ψ_a and ψ_b

$$Q_{l,m}(ab) = e \sum_{k=1}^z \int r_k^l Y_{l,m}^*(\theta_k \phi_k) \psi_b^* \psi_a d\tau \quad (2.16)$$

Similarly, for the magnetic multipole moment

$$M_{l,m}(ab) = -\frac{1}{l+1} \frac{e\hbar}{m_P} \sum_{k=1}^z \int r_k^l Y_{l,m}^*(\theta_k \phi_k) \text{div}(\psi_b^* \mathbf{L}_k \psi_a) d\tau \quad (2.17)$$

where $\hbar \mathbf{L}_k = -\hbar i \mathbf{r}_k \times \nabla_k$ is the angular momentum operator [128]. This is accounting only for the contribution to the magnetic field coming from the *orbital motion* of the protons, and an additional term is necessary to take the *intrinsic* magnetic moments into account. Given their quantum mechanical nature and the relation to electromagnetic transitions (Figure 2.3, for which the *natural width* Γ can be expressed as a function of the frequency or the energy $\Gamma = \hbar \Delta\omega_{1/2} = \hbar \lambda = \hbar/\tau$), these moments are subject to selection rules and *reduced transition rates*, which, for an electric transition of order l , can be defined as [128]

$$B(E\ell, J_a \rightarrow J_b) = \frac{1}{2J_a + 1} \sum_{m_{J_a}} \sum_{m_{J_b}} Q_{l,m}^2(ab) \quad (2.18)$$

taking into consideration the requirements for the value of $|m| = |m_{J_a} - m_{J_b}| \leq l$. The same expression holds for magnetic transitions, replacing $Q_{l,m}(ab)$ with $M_{l,m}(ab)$. The *decay constants* for the electric and magnetic transitions, λ_E and λ_M are then:

$$\lambda_E(l, J_a \rightarrow J_b) = \frac{2(l+1)}{\hbar \epsilon_0 l[(2l+1)!!]^2} \left(\frac{\omega}{c}\right)^{2l+1} B(E\ell, J_a \rightarrow J_b) \quad (2.19)$$

$$\lambda_M(l, J_a \rightarrow J_b) = \frac{2(l+1)\mu_0}{\hbar l[(2l+1)!!]^2} \left(\frac{\omega}{c}\right)^{2l+1} B(M\ell, J_a \rightarrow J_b) \quad (2.20)$$

Remembering that $\lambda = 1/\tau$ and $\Gamma\tau = \hbar$, hence $\Gamma = \hbar/\tau = \hbar\lambda$, from Equation 2.19, it is possible to obtain

$$\Gamma_{\gamma(E\ell)} = \frac{2(l+1)}{\epsilon_0 l[(2l+1)!!]^2} \left(\frac{\omega}{c}\right)^{2l+1} B(E\ell, J_a \rightarrow J_b) \quad (2.21)$$

As discussed in the introduction to this Section, stringent comparisons between the estimated transition rates with different models and the experimental values, would be very useful to obtain insights on nuclear structure. The transition probability (or rate) for a given type of multipole radiation can, in fact, be theoretically computed through Eqs. 2.19 and 2.20, but an accurate knowledge of the wavefunctions of the initial and final states is needed to know the multipole matrix elements. However, following the prescriptions reported by Weisskopf [129], to at least estimate the order of magnitude of these transitions, it is enough to assume simple shell-model wave functions, with the approximations listed here: (i) the wave function of the initial state is a single-particle function described by a spatial term and a spherical harmonic, $\psi_a = R_a(r)Y_{l,m}(\theta, \phi)$, i.e., the one describing an *odd* proton, while the other nucleons (assumed to be in closed shells) are not contributing to the matrix element; (ii) the final state is described by a single-particle proton wave function $\psi_b = R_b(r)/\sqrt{4\pi}$, i.e., that of an *S*-state; (iii) only to the aim of making a crude estimate, it is possible to assume the radial wave functions (R_a and R_b) to be constant inside the nucleus and vanishingly small outside. This is thus referring to a simple transition in which a proton is jumping from a state with orbital angular momentum ℓ to a final state with momentum equal to zero, without the other particles taking part in the transition. Given that both squares of the radial wave functions must be normalized to 1 inside the volume of the nucleus, one can insert the values of ψ_a and ψ_b listed above in Equation 2.16 and, since the spherical harmonic function is normalized, after the radial integration [128], it is possible to obtain the expression for $Q_{l,m}(ab)$

$$Q_{l,m}(ab) = \frac{e}{\sqrt{4\pi}} = \frac{3R^l}{l+3} \quad (2.22)$$

In the end, if this value is put in Equation 2.14 for the determination of λ_E , this becomes

$$\begin{aligned} \lambda_E^{Weiss.}(l) &= \frac{2(l+1)}{l[(2l+1)!!]^2} \left(\frac{3}{l+3}\right) \frac{e^2 R^{2l}}{4\pi\epsilon_0\hbar} \left(\frac{\omega}{c}\right)^{2l+1} = \\ &= \frac{4.4(l+1)10^{21}}{l[(2l+1)!!]^2} \left(\frac{3}{l+3}\right)^2 \left(\frac{E_\gamma}{197}\right)^{2l+1} R^{2l} \end{aligned} \quad (2.23)$$

where the transition energy $E_\gamma = \hbar\omega$ is expressed in MeV, and the nuclear radius R in fm. When $\lambda = \lambda^{Weiss.}$, this value is often indicated as one *Weisskopf units* (1 w.u.). Similar calculations, on the basis of Equation 2.15, can be done for the magnetic multipole transitions.

The Weisskopf estimates of the λ_γ or, equivalently, of the $B(E\ell)$, are powerful indicators of the *nature* of the nuclear states involved in the electromagnetic transition. For example, quadrupole transitions between states with large deformation dominated by a prominent collective nature are characterized by very large values of $B(E2)$, often exceeding 10 times the corresponding Weisskopf estimates.

In the cluster realm, the archetypal example is constituted by the $B(E2)$ value for the $4^+ \rightarrow 2^+$ transition in ^8Be measured by Datar *et al.* [123, 124]. This transition involves two states of the molecular band built on the super-deformed $\alpha - \alpha$ ground state of ^8Be . The $BR = \frac{\Gamma_\gamma}{\Gamma_\alpha}$ branching ratio is *tiny* ($BR \simeq 1.3 \cdot 10^{-7}$), but the obtained $\Gamma_\gamma \simeq 0.48$ eV value would correspond to $B(E2) \simeq 21 e^2 fm^4$ that means about 22 w.u., indicating the strong deformation of the nuclear molecule, in full agreement with cluster model calculations [130].

In the ^{12}C case, the situation is less simple [131]. While it is not difficult to obtain the $B(E2)$ value for the *bound* 4.44 MeV 2^+ state ($2_1^+ \rightarrow GS$ transition, $\Gamma_\gamma = 10.8$ meV, $B(E2) = 7.94 \pm 0.66 e^2 fm^4$, i.e. just 0.97 w.u. ²[132]), the situation is by far very complicated for the $0_2^+ \rightarrow 2_1^+$ γ -transitions starting from the *unbound* Hoyle state and going to the 4.44 MeV state³. A general consensus was obtained during the experimental campaigns of the '60s and '70s, leading to a value of $\Gamma_\gamma^{E2} \simeq 3.81 \pm 0.39$ meV [133], corresponding to $B(E2) \simeq 13.5 \pm 1.4 e^2 fm^4$. If one uses Equation 2.23 assuming a radius parameter $r_0 = 1.2$ fm and using the transition energy $E_\gamma(0_2^+ \rightarrow 2_1^+) \simeq 7.654 - 4.439 = 3.215$ MeV, it will be possible to obtain $\lambda_{Weiss.} \simeq 6.91 \text{ s}^{-1}$ and then $\Gamma_{Weiss.} \simeq 0.456 \cdot 10^{-3}$ eV; this would then lead to the very large value of 8.36 ± 0.86 w.u. for the transition probability. Such experimental value is more or less reasonably reproduced by theoretical models where the Hoyle state is characterized by a very well developed 3α triangular structure, while the ground state has a more ordinary spheroidal like structure [134]; this aspect will be deepened at the end of Chapter 3. As an example, recent Monte Carlo Shell Model calculations [121] predict a $B(E2) = 8.5 e^2 fm^4$, not too far from the experimental value. In a recent experimental work, however, the Γ_γ^{E2} value was increased up to $\Gamma_\gamma^{E2} \simeq 5.0 \pm 0.5$ meV [125], about 3σ far from the literature value; consequently, also the $B(E2)$ would be larger by a factor $\simeq 1.34$ than the previously accepted values, changing the agreement between the various models aiming to the microscopic description of the structure of the Hoyle state. This subject would be discussed in more details in Chapter 3 of the present thesis, where a new experiment on the radiative transition from the Hoyle state is reported.

2.2.1 Experimental techniques for measurements of radiative transitions from particle-unbound states

One of the most important ways to obtain information about the properties of a particular state is based on the experimental measurement of the branching ratio of the radiative decay Γ_{rad}/Γ , associated to electromagnetic transitions; if the total Γ is known from the literature, it is immediate to estimate the Γ_{rad} of the state with all the nuclear structure consequences discussed in the previous paragraph. The most common experimental techniques exploited for these type of measurements are the *associated-particle* and the *particle- $N\gamma$ multiple coincidence* techniques, which are respectively schematized in Figures 2.4 and 2.5.

²The $B(E2)$ values can be given, in absolute units, for an *exciting* transition $0_1^+ \rightarrow 2_1^+$ or for a *decay* transition $2_1^+ \rightarrow 0_1^+$. In a compact way, they are indicated respectively as $B(E2) \uparrow$ and $B(E2) \downarrow$. The two quantities are linked by a spin factor: $\frac{2J_i+1}{2J_f+1} = \frac{1}{5}$, in a way that $B(E2) \downarrow = \frac{1}{5} B(E2) \uparrow$ [132].

³In this discussion, the *pair emission* decay directly linking the Hoyle state with the ground state is being neglected, given that its relative branching ratio is of the order of 1% with respect to the γ cascade, see [126].

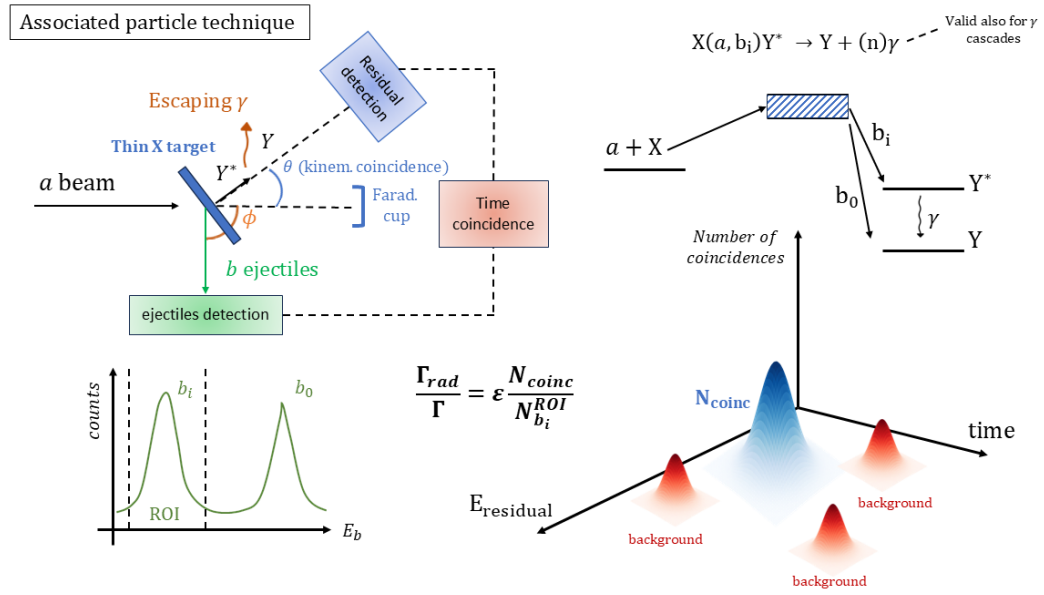


FIGURE 2.4: Schematic overview of an experimental design set-up based on the associated particle technique for a generic $X(a, b_i)Y^* \rightarrow Y + (n)\gamma$ reaction. The $(n)\gamma$ notation is used to generalize the process to single or cascade γ emission.

The associated particle technique is based on the detection of the particles (b_i) ejected after the collision between the *projectile* a and the *target* X , detected at a given angle (constrained by the kinematics of the collision), selected with the correct energy value, and registered in time coincidence with the detection of the residual Y nucleus. Kinematic constraints are also useful to unequivocally relate the energy and the angles of the ejectiles with those of the corresponding residual nuclei: the latter can be in their ground state, whose ejectiles are usually tagged as b_0 , or in a given i -th excited state, with the respective ejectiles noted as b_i . The number of counts of b_i particles in their specific detector will thus be equal to the number of residual Y^* nuclei formed in the i -th excited state going in the kinematical cone fixed by the coincidence geometry. The time coincidence measurement between the residual and the ejectile detection is useful because of the fact that the Y^* nucleus may decay before reaching the detector. As reported in detail in a dedicated experiment in Chapter 3, these type of measurements are suitable for the determination of radiative decay branching ratios, like in the case of the ^{12}C Hoyle state. For example, in the particular case of the ^{12}C Hoyle state, the excited carbon nuclei decay, most of the time, through α particle emission, but just ≈ 1 in 2500 times they decay through electromagnetic (γ or pairs) transitions; these pathways leave a ^{12}C in the ground state, whose kinematics is similar (but not identical, as will be shown in Chapter 3) to the other excited ^{12}C recoils, and is detected by the same apparatus at the *right* recoil angle expected from kinematics.

Since the recoil detector is of course sensitive to a number of other contaminant reactions, to avoid having to look for a needle in a haystack, it is necessary to tag the detected ground state carbons in a kinematic plus time-coincidence with the corresponding ejectile (defined as $b_i = b_2$ particles). These events can be represented through a two-dimensional histogram such as that shown in the right-hand side of Figure 2.4, where the event counts are reported as a function of the time-of-flight difference and the energy of the recoil ions

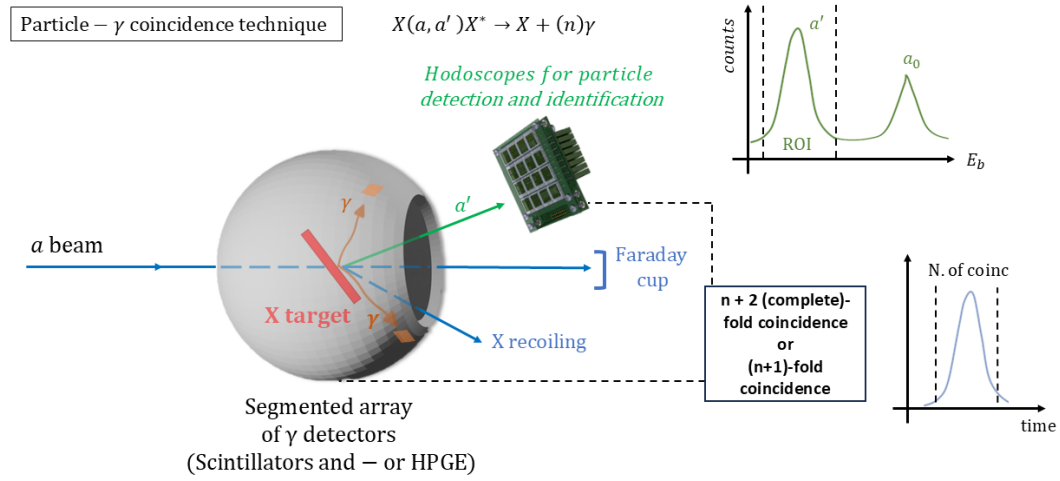


FIGURE 2.5: Sketched example of the typical set-up for a general particle– γ coincidence experiment.

[126]. There, the events lying beneath the area of the coincidence (blue) peak are the valid hits (the counts of interest) which are separated from the background (red) counts thanks to the refinement of the technique. As reported in the Figure, the branching ratio (Γ_{rad}/Γ) for the radiative decay of a particular excited state is thus given by the ratio between the total number of coincidence events (N_{coinc}) divided by the total number of ejectile counts in the region of interest (N_{bi}^{ROI}), with all the suited efficiency or loss corrections. A detailed breakdown of this technique for the study of the radiative decay branching ratio of the ^{12}C Hoyle state will be presented in Chapter 3.

The other technique, illustrated in Figure 2.5, is the particle– $N\gamma$ coincidence technique, and may allow to obtain information similar to that of the associated particle technique, e.g., on the radiative decay of the ^{12}C Hoyle state; in this case, this decay may be observed through the $\gamma - \gamma$ cascade happening via the 4.44 MeV excited state. The cascade is observed upon detection of the two γ rays with two detectors (if their angles are known) or, even better, with a segmented array of scintillators or HPGE detectors for γ rays; in the mean time, the other particle (the a' particle in the generalization of Figure 2.5, but, e.g., an inelastic proton in a $^{12}\text{C}(p, p')^{12}\text{C}$ process) is detected with a suitable detector, as a hodoscope which may grant the detection *and* identification of the particles. The energy of the detected inelastically scattered particle can be directly related to the excitation energy of the recoiling target nucleus; the number of triple coincidences (γ, γ and inelastic particle) allows to obtain the Γ_γ/Γ ratio.

These types of studies allowed to understand that the decay of the ^{12}C Hoyle state mainly proceeds through the pathway of the α particle channel, meaning that the partial width of this decay (Γ_α) is approximately equal to the total width Γ ($\approx 8.90 \pm 1.08 \text{ eV}$), while that of the γ and the radiative channel are of the order of few meV. The resonance strength (ω_γ) is thus completely determined by the α decay channel, as

$$\omega_\gamma = \Gamma_\alpha \Gamma_{rad} / \Gamma = \Gamma_{rad} \quad (2.24)$$

The triple- α process is a textbook example of a reaction whose rate determination has very important astrophysical and practical consequences. For example, the triple α process, burning helium nuclei in red giant stars, is fully depending on this quantity

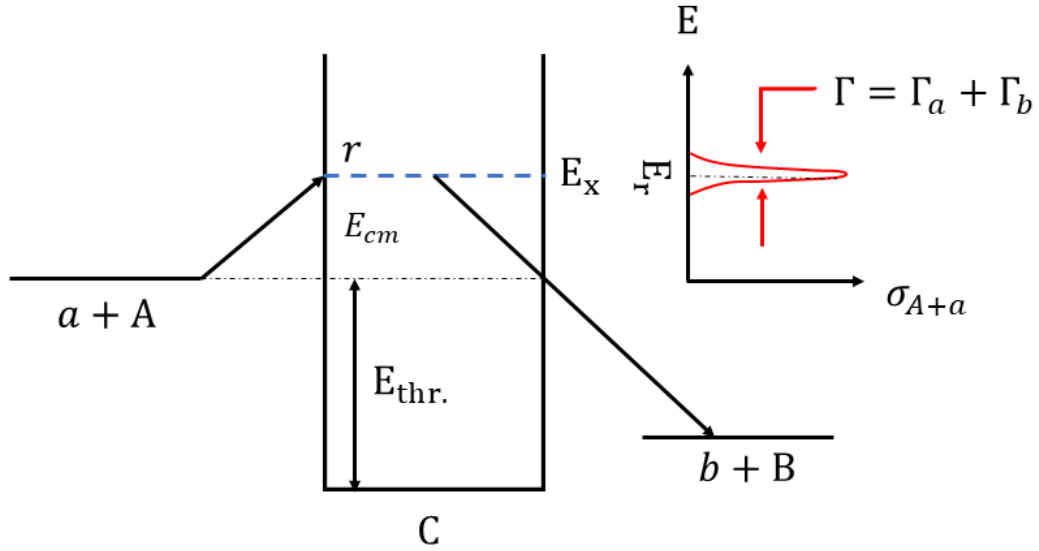


FIGURE 2.6: Outline of a compound nucleus mechanism populating a resonance at an energy E_r . The excited state in the compound nucleus is formed by an $a + A$ collision and decay in the $b + B$ channel. A schematic view of the occurrence of an *isolated* resonance in the $A(a,b)B$ integrated cross section is sketched on the right side of the panel.

[135] and even a small variation (within a few tens of percent) with respect to the commonly adopted value would lead to sizable changes both on the nucleosynthesis side and on questions linked to stellar evolution, as discussed in details in Ref. [136].

2.3 Compound Nucleus reactions and Nuclear Resonances

High-precision experiments involving the formation of a compound nucleus represent a powerful way to probe clustering in nuclei. The compound nucleus reaction theory was early introduced by Niels Bohr in 1936: according to the model, a low-energy incident particle is fully *absorbed* by the target nucleus. After several internal collisions with the other nucleons in the target, it will share a part of its momentum with the other nucleons of the target, leading to a change in the momentum distribution inside the nucleus. The binding energy and a part of the kinetic energy of the projectile will be converted into excitation energy of the system formed by the merging of target + projectile, that is then named the *compound nucleus* [31, 137].

To outline this model, let's consider a projectile a impinging on a target A : the *formation* of a compound system C will be observed at a given excitation energy. Such a unstable system will subsequently decay in any of the *open* channels, i.e. a particle decomposition (typically a couple) that is allowed by energy, angular momentum, parity and eventually isospin conservation rules.

To simplify the dissertation, it is assumed that an *isolated* resonance is formed in C , and that only two open channels are involved, a and b . The "b" label indicates the emission of an *ejectile* nucleus b and a *residual* nucleus B , as sketched in Fig. 2.6. From the quantum mechanics point of view, the *cross section* σ must depend on the properties of

the excited state of the compound nucleus through the logarithmic derivative f_l of the radial wave-function $u_l(r)$ at the nuclear boundary $r = R$ [127, 138].

The solution of the Schrödinger equation beyond the nuclear surface leads to the radial wave function, which assumes the form (see Refs.[127, 138, 139] for a complete derivation):

$$\begin{aligned} u_l(r) &= Au_l^+(r) + Bu_l^-(r) \\ &= Ae^{-i\delta_l}[G_l(r) + iF_l(r)] + Be^{-i\delta_l}[G_l(r) - iF_l(r)], \quad r > R \end{aligned} \quad (2.25)$$

where u_l^+ and u_l^- are the asymptotic limits of incoming (+) and outgoing (−) spherical waves at large distances from the reaction vertex, and δ_l is the Coulomb *phase shift*. Several simplifications occur for $l = 0$ neutrons: it is possible to express $F_l = krj_l(kr)$ and $G_l = kr\eta_l(kr)$, where $j_l(kr)$ and $\eta_l(kr)$ are the spherical Bessel and Neumann functions. For the more complicated scenario of a charged particle with a given orbital angular momentum ℓ , they respectively correspond to the *regular* and *irregular* Coulomb wave-functions. In this context the *real* quantities S_l and P_l (shift factor) and *penetration factor* are particularly important. They are fully determined by the *boundary conditions* of f_l outside the nucleus:

$$\begin{aligned} f_l &= R \left(\frac{1}{u_l^+(r)} \frac{du_l^+(r)}{dr} \right)_{r=R} \\ &= R \left[\frac{G_l(dG_l/dr) + F_l(dF_l/dr) + iG_l(dF_l/dr) - iF_l(dG_l/dr)}{F_l^2 + G_l^2} \right]_{r=R} \\ &\equiv S_l + iP_l \end{aligned} \quad (2.26)$$

being:

$$\begin{aligned} S_l &= R \left[\frac{F_l(dF_l/dr) + G_l(dG_l/dr)}{F_l^2 + G_l^2} \right]_{r=R} \\ P_l &= R \left(\frac{k}{F_l^2 + G_l^2} \right)_{r=R} \end{aligned} \quad (2.27)$$

These quantities depend on the wave-number k in the center of mass system, the channel radius R and the orbital angular momentum l ; the Coulomb functions for charged particles also depends on the Sommerfeld parameter (η) [138]. If one considers $l = 0$ neutrons for simplicity, being $F_0 = \sin(kr)$ and $G_0 = \cos(kr)$ [46], it is possible to obtain $P_0 = kr$ and $S_0 = 0$: the shift factor is vanishing, due to the absence of a barrier. The reaction cross section close to an isolated resonance is thus obtainable in terms of P_l and S_l (see, e.g., [138]), leading to the well-known *Breit-Wigner* formula:

$$\sigma = (2l + 1) \frac{\pi}{k^2} \frac{\Gamma_a \Gamma_b}{(E - E_r)^2 + \Gamma_r^2/4} \quad (2.28)$$

where Γ_a is the *partial width* for the entrance channel of the reaction, and Γ_b for the exit channel. The partial widths can be calculated as

$$\begin{aligned} \Gamma_a &\equiv -\frac{2[P_l(E)]_a}{(\partial f_l / \partial E)_{E_r^0}} = 2P_l^a(E)\gamma_a^2 \\ \Gamma_b &= 2P_l^b(E)\gamma_b^2 \end{aligned} \quad (2.29)$$

where $P_l^a(E)$ and $P_l^b(E)$ are the penetrability factors for the entrance and the exit channels, following the same notation as for the partial widths. The penetrability factor is important in the determination of the partial widths since a particle must penetrate the barrier to induce the reaction. Furthermore, γ_a^2 and γ_b^2 are the *reduced widths*, easily obtained via the inversion of Eqs. 2.29. These numbers make the σ dependent on the properties of the *inside* of the compound nucleus, and they are connected to the spectroscopic factors C^2S , as will be later shown.

The total width of the resonance would approximately correspond to the width of the observed Lorentzian-like function characterizing the cross section, in proximity of an isolated resonance. This is inversely proportional to the resonance lifetime, and corresponds to the sum of each partial width:

$$\Gamma_r = \Gamma_a + \Gamma_b \quad (2.30)$$

Another very important property of a resonant state is the *observed* resonance energy, E_r , defined as:

$$E_r = E_r^0 + \frac{S_l(E)}{(\partial f_l / \partial E)_{E_r^0, q=0}} = E_r^0 - S_l(E) \gamma_a^2 \quad (2.31)$$

indicating that E_r , observed in the peaking of the cross section for an isolated resonance, is actually shifted with respect to the *formal* energy E_r^0 , of a quantity $-S_l(E) \gamma_a^2$. This feature is directly related to the presence of a barrier in the entrance channel.

To understand the link between the nuclear reaction quantities discussed here and the nuclear structure of a state, a semi-classic example (see, e.g. [31]) would be extremely useful. A suitable candidate is one excited state of ^{11}Be , whose wave function Ψ_1 is that of the ground state of ^{10}Be with a further neutron in the $2s_{1/2}$ orbit, i.e.

$$\Psi_1(^{11}\text{Be}) = \Psi(^{11}\text{Be}_{gs}) \otimes \Psi(2s_{1/2}) \quad (2.32)$$

Since ^{10}Be is an even-even nucleus, its ground state will be 0^+ and thus the state of equation (2.32) would have $J^\pi = 1/2^+$. This is a highly excited state of ^{11}Be with an unbound neutron (given that, according to shell model predictions, the last bound neutron state would stay in the $1p_{1/2}$ orbital); this can be released in a nucleon-emitting process like:

$$^{11}\text{Be}^* \rightarrow ^{10}\text{Be}_{gs} + n(2s_{1/2}) \quad (2.33)$$

If one indicates with v_i the velocity of the neutron *inside* the ^{11}Be nucleus and with R the radius of the nucleus, the extra-neutron will reach the nuclear surface in a time of the order of R/v_i . When hitting the surface, it will feel a change in the potential which can reflect it again inside the nucleus. It is possible to demonstrate (on the basis of a simple wave mechanics calculation, see e.g. [31]) that, on average and in first approximation, it will be reflected a number of times equal to $v_i/4v_e$, where v_e is the velocity with which the outgoing neutron is eventually emitted [31]. The average time required for a neutron to be emitted is thus:

$$\tau_0 \approx \frac{R}{v_i} \frac{v_i}{4v_e} = \frac{R}{4v_e} \quad (2.34)$$

A more accurate description of the transmission coefficients leads to a slightly different expression:

$$\tau_0 \approx \frac{R}{3v_e} \rightarrow \Gamma_0 = 3 \frac{\hbar v_e}{R} \quad (2.35)$$

If one indicates the reduced mass of the considered binary channel decomposition with μ , and treats v_e as the relative velocity of the decay partners, it is possible to obtain $\mu v_e = \hbar k$, where k is the *external* wave number, related to the unbound particles. After simple substitutions:

$$\Gamma_0 = 3 \frac{\hbar v_e}{R} = \frac{3 \hbar^2 k}{\mu R} = \frac{3 \hbar^2 k R}{\mu R^2} = 2 \cdot k R \cdot \frac{3}{2} \frac{\hbar^2}{\mu R^2} \quad (2.36)$$

But for s -wave neutrons, the penetrability is just $P_0 = kR$, as already shown; based on the definition of Eq. (2.29), the following value for the reduced partial width can be considered as the so-called *single-particle limit* or *Wigner limit*:

$$\gamma_0^2 = \frac{3}{2} \frac{\hbar^2}{\mu R^2} \quad (2.37)$$

When taking into account all the possible configurations of the type of equation (2.32) leading to a $J^\pi = 1/2^+$ state, it is possible to write the expansion:

$$\begin{aligned} \Psi_i(^{11}\text{Be}^*) = & c_{i1} \Psi(^{10}\text{Be}_{gs}) \otimes \Psi(2s_{1/2}) + \\ & + c_{i2} \Psi(^{10}\text{Be}_{1-}) \otimes \Psi(1p_{1/2}) + c_{i3} \Psi(^{10}\text{Be}_{2+}) \otimes \Psi(1d_{5/2}) + \dots \end{aligned} \quad (2.38)$$

which represents a wave-function expansion of a generic $1/2^+$ state i of ^{11}Be in the $^{10}\text{Be} + n$ configurations; when thought from a dynamical point of view, one can consider the nucleus as spending a given quantity of time in each of them. The decay partial width of the i -th state through the emission of a neutron will be thus linked to the partial lifetime and the partial widths by:

$$\Gamma_i = \hbar \sum_k \lambda_{0k} c_{ik}^2 \quad (2.39)$$

where λ_{0k} is the decay rate of the k -th configuration (see equation 2.34) and the c_{ik}^2 factors are equivalent to the fraction of time spent by the nucleus in the k -th configuration. The sum of all these amplitudes gives:

$$\theta_i^2 = \sum_k |c_{ik}|^2 \quad (2.40)$$

corresponding to the *dimensionless* reduced partial widths, i.e. the ratio of the above-discussed decay partial widths to those at the Wigner limit $\theta_i^2 = \frac{\gamma_i^2}{\gamma_0^2}$. Given the nature of the factors c_{ik} defined in equation (2.38), the following *sum rule* is derived:

$$\sum_i \theta_i^2 = 1 \quad (2.41)$$

Then, for the generic i -th state decaying by neutron emission, $\theta_i^2 \leq 1$. When the structure of the state is completely dominated by one precise single-particle configuration, like in $^{10}\text{Be} + n$, it will have $\theta_i^2 \simeq 1$: this will mean that the decay partial width is *at the Wigner limit*. The experimental investigation of a nuclear state's partial width is therefore very useful to probe the occurrence of peculiar configurations. This is useful in the case of clustering phenomena, for which it is possible to expect that the partial

widths linked to the α emission or other cluster decompositions would be characterized by a significant fraction (typically $> 10\%$) of the Wigner limit.

The mentioned features are not exclusive to the case of $l = 0$ $X + n$ configurations, but can be generalized to all the possible channels, and their extension to the more complicated case of charged particles emission in an arbitrary l wave can be derived (see Ref. [31, 127] for more details). The following derivation is based on the hypothesis that the compound nucleus populates the resonance r and only one channel, c , is open. The corresponding wave function of this meta-stable state, contributing to the emission in the channel c , is composed by a linear combination of radial single-particle wave functions u_{rk}^c , each contributing to a particular configuration of the compound system in the decomposition c ⁴:

$$\Psi_r(\vec{R}) = \left[\sum_k c_{rk} u_{rk}^c(R) \right] \frac{Y_{lm}(\theta, \varphi)}{R} \quad (2.42)$$

The partial width Γ_c of the resonant state is defined as the probability flux of the particle through the only open channel c , and it can be calculated by integrating the quantum-mechanical current [46, 139], through a sphere of radius R , in the whole solid angle:

$$\Gamma_r^c = \hbar \int_{4\pi} R^2 j d\Omega = \hbar \int_{4\pi} R^2 \frac{\hbar}{2i\mu} \left(\Psi_r^* \frac{\partial \Psi_r}{\partial r} - \frac{\partial \Psi_r^*}{\partial r} \Psi_r \right) d\Omega \quad (2.43)$$

After some algebraic steps (see Ref.[127]):

$$\Gamma_r^c = \sum_k 2 \frac{\hbar^2}{\mu R^2} P_l^c(E) C^2 S \varphi_{ck}^2 \quad (2.44)$$

where:

$$C^2 S = c_{rk}^2 \quad (2.45)$$

$$\varphi_{ck}^2 = \frac{R}{2} |u_{rk}^c(R)|^2 \quad (2.46)$$

The $C^2 S$ quantities are the *spectroscopic factors*, i.e., the square of the *spectroscopic amplitudes* used in the linear combination of Eq. 2.42, and the φ_{ck}^2 quantities are the decay rates in the channel c from each configuration k .

The spectroscopic factor, tightly linked to the structure of a state, is a measure of the probability that a compound state r , with a cluster configuration c , can be described by the single-particle configuration k . From Eq. 2.44 the partial width of the decay of a state in a particular channel can be written as a product of three factors: (i) the probability of the nucleons arranging themselves in a configuration corresponding to the final state ($C^2 S$), (ii) the probability of the single particle being present at the boundary ($\propto |u_{rk}^c(R)|^2$), and (iii) the probability of the particle penetrating the centrifugal and Coulomb barrier ($P_l^c(E)$).

⁴Typically, a channel (decomposition c) involves the presence of the two particles b and Y . Each *particular configuration of a given decomposition* is found by solving the Schrödinger equation for a Hamiltonian describing the motion of particle b within the well generated by $b + Y$ in the single-particle approximation. This equation leads to several (discrete) solutions that will represent a *particular configuration* of the decomposition c ($\equiv b + Y$).

2.3.1 The R -matrix theory: basic elements

The R -matrix theory is frequently used in compound nucleus scenarios to draw spectroscopic properties, such as E_x , J , π , γ_c^2 , of given nuclear states starting from the experimental excitation functions and angular distributions; it is, therefore, a very useful tool to investigate clustering phenomena in light nuclei. More thorough treatments of the R -matrix theory can be found in Refs. [139, 140], while only a fast presentation will be given in the following paragraphs.

The R -matrix method was introduced by Wigner and subsequently refined with Eisenbud's aid [141, 142] during the '30s. The most important observables of nuclear reactions are described through several parameters, like the radius a of the *nuclear sphere*, the energy levels, and the reduced widths. These are typically treated as free parameters in best-fit procedures of experimental data, while any assumptions are made around the shape of the wave functions inside the *nuclear sphere*; only their properties at its surface are used. These are expressed in terms of the logarithmic derivatives of the wave functions at the boundaries, forming the matrix element. This makes the theory very general, since it does not rely on any detail about the potential energy function inside the nucleus, except the assumption regarding the formation of the compound nucleus itself. The R -matrix theory grants to extend the previously discussed Breit-Wigner formula (2.28) of the cross section to a more general form, including an arbitrary number of channels and resonances, also considering the mutual interference effects. The R -matrix links the value of the internal region wave function to its derivative at each entrance channel, and is defined, for an arbitrary number of channels c, c' , as:

$$\Re_{c,c'} \equiv \sum_r \frac{\gamma_{rc'} \gamma_{rc}}{E_r - E} \quad (2.47)$$

This definition (2.47) explicitly connects the elements of the R -matrix to the energy, being γ_{rc} and E_r energy-independent parameters. The poles of the R -matrix occur at each value E_r of energy and each of the $\Re_{c,c'}$ elements represent a real number. The position of the poles is moreover independent on the channels c and c' . In the framework of R -matrix, a channel is identified by the quantum numbers $\{\alpha(J_1 J_2)sl, JM\}$, where the $\alpha(J_1 J_2)$ represent a specific state of excitation α of given nuclei (1 and 2), with spins J_1 and J_2 , $\vec{s} = \vec{J}_1 + \vec{J}_2$ (channel spin), l (orbital angular momentum of their relative motion), $\vec{J}$ and M (the total spin and its component along a quantization axis). For an entrance channel made by a projectile and a target nucleus, one can set $\vec{J}_1 = \vec{j}_p$ and $\vec{J}_2 = \vec{j}_t$, following:

$$\vec{J} = \vec{l} + \vec{j}_p + \vec{j}_t \quad (2.48)$$

Given the degeneracy of these values, there are $(2l+1)(2j_p+1)(2j_t+1)$ different sets of spin orientations happening with the same probability. The cross section σ must then be multiplied for the statistical factor:

$$g(J) = \frac{2J+1}{(2l+1)(2j_p+1)(2j_t+1)} \quad (2.49)$$

The σ and the phase shifts can be derived by using the boundary conditions and the energy independent parameters γ_{rc} . In the case of an isolated resonance and an arbitrary number of channels, one can derive the so-called *generalized one-level Breit-Wigner*

formula for a generic reaction leading from the α to the α' channel:

$$\sigma(\alpha, \alpha') = \frac{\pi}{k^2} \frac{2J+1}{(2j_p+1)(2j_t+1)} \frac{(\sum_{ls} \Gamma_{rc})(\sum_{l's'} \Gamma_{rc'})}{(E - E_r - \Delta_r)^2 + \Gamma_r^2/4} \quad (2.50)$$

where $\Gamma_{rc} = 2P_c(E)\gamma_{rc}^2$ are the partial widths, $\Gamma_r(E) = \sum_c \Gamma_{rc}(E)$ is the total width of the resonance r and $\Delta_r(E) = \sum_c \Delta_{rc}(E)$ is the total level shift. The latter is the overlap of partial level shifts, already presented in the Breit-Wigner equation: $\Delta_{rc}(E) = -[S_c(E) - S_c(E_r)]\gamma_{rc}^2$, with both penetration and shift factors referred to the *interaction radius*. This is usually set as small as possible, so that the quantities of the resonance theory contain primarily information on the nuclear interaction, thus commonly taken equal to the smallest separation distance of the nuclear pair for the reaction to occur, i.e. the *channel radius* $R = r_0(A_t^{1/3} + A_p^{1/3})$, with $r_0 = 1.0$ - 1.5 fm. A simplification of equation 2.50 was introduced by Thomas in 1951, observing that since the shift factor $S_c(E)$ only has a weak dependence on the energy, it is expandable as follows:

$$E - E_r - \Delta_r \approx (E - E_r) \left[1 - \left(\frac{d\Delta_r}{dE} \right)_{E_r} \right] \quad (2.51)$$

By using the result of equation 2.51, the commonly used formulation of equation 2.50, reproducing the $\alpha \rightarrow \alpha'$ cross section for an isolated resonance r , becomes:

$$\sigma(\alpha, \alpha') = \frac{\pi}{k^2} \frac{2J+1}{(2j_p+1)(2j_t+1)} \frac{(\sum_{ls} \Gamma_{rc}^o)(\sum_{l's'} \Gamma_{rc'}^o)}{(E - E_r)^2 + \Gamma_r^o/4} \quad (2.52)$$

where the *formal widths* Γ_{rc} have been replaced by the *observed widths* Γ_{rc}^o :

$$\Gamma_{rc}^o \equiv \frac{\Gamma_{rc}}{1 - (d\Delta_r/dE)_{E_r}} = \frac{\Gamma_{rc}}{1 + \left(\sum_{c''} \gamma_{rc''}^2 \frac{dS_{c''}}{dE} \right)_{E_r}} \quad (2.53)$$

The difference between formal and observed width becomes much more important following the increase of the reduced width of the state.

Operatively, it is possible to use multi-channel multi-level R -matrix formulations like the one of Ref. [143] to find the set of parameters E_x , J , π , γ_c^2 reproducing the experimental trend of the cross section for each considered channel and process. In particular, the approach of [143] shows the sizable advantage of using, as starting parameters in the fit search, the observable values and not the formal ones, strongly simplifying the data analysis. Given that clustering phenomena are related to states with pronounced α -emission (Γ_α) partial widths, the study of compound nucleus reaction channels involving the emission of an α -particle, like *resonant elastic scattering* (RES) (α, α), *resonant inelastic scattering* (α, α'), (p, α) (as in Chapter 5 of this thesis) and (d, α) reactions, is crucial to determine the Γ_α partial width of a given excited state from best-fit procedures. A comparison of the γ_α^2 reduced widths with the single-particle (Wigner) limit is then a very important instrument to unveil the presence of states characterized by a pronounced α structure. In Chapter 5 this criterion will be intensively used to search for the existence of quartet excitations in ^{20}Ne .

2.3.2 Coupling of angular momenta for the analysis of angular distributions

Apart from the complete R -matrix theory, there is a different, and more handsome in a certain sense, way to determine (with some approximations, of course) the possible J^π value of a resonant state populated in a compound nucleus experiment. It is based on the angular correlation theory chiefly developed by the mathematical physicist Lawrence Biedenharn, and here briefly discussed, with some interesting examples that will pave the way to its subsequent application to the ${}^3\text{He}+{}^{13}\text{C}$ data discussed in Chapter 4. First of all, it is necessary to restrict the discussion to nuclear reactions induced by spinless particles: in this case j_i and j_f will indicate the spin of the target and residual nuclei, ℓ_i and ℓ_f the relative orbital angular momenta in the initial and final channels, and J will be the spin of the compound nucleus. It will be assumed that a single *isolated resonance* is excited in the experiment. The sequence of such values will be indicated by the symbol $j_i(\ell_i)J(\ell_f)j_f$. In this case, the angular correlation theory [144, 145] predicts that the angular distribution of the observed ejectiles assumes the form:

$$\frac{d\sigma}{d\Omega}(\theta_{cm}) = \frac{(-1)^{j_i-j_f}}{4\pi} \sum_n A_n(i)A_n(f)P_n[\cos(\theta_{cm})] \quad (2.54)$$

where $P_n[\cos(\theta_{cm})]$ are the Legendre polynomials, n runs from 0 to ∞ , and $A_n(i)$ and $A_n(f)$ are two coefficients taking into account the coupling of angular momenta in the various reaction channels:

$$\begin{aligned} A_n(i) &\equiv \sqrt{2J+1} \cdot \bar{Z}(\ell_i J \ell_i J; j_i n) \\ A_n(f) &\equiv \sqrt{2J+1} \cdot \bar{Z}(\ell_f J \ell_f J; j_f n) \end{aligned} \quad (2.55)$$

The $\bar{Z}$ are the so-called Huby coefficients, that are in turn related to the (more famous) Wigner $(\ell\ell'00|n0)$ and Racah $W(\ell J \ell' J'; jn)$ coefficients:

$$\bar{Z}(\ell J \ell' J'; jn) \equiv \sqrt{(2\ell+1)(2\ell'+1)(2J+1)(2J'+1)} \cdot (\ell\ell'00|n0) \cdot W(\ell J \ell' J'; jn) \quad (2.56)$$

A very important property of the Racah coefficients is that they are *non-zero* only for a limited number of n in the series, and in particular the n index of the sum must obey the inequality:

$$n \leq \min(2J, 2\ell_i, 2\ell_f) \quad (2.57)$$

Furthermore, the parity conservation, related to the properties of the Wigner coefficient, restricts each value assumed by the n index of the sum to be *even*. Finally, to summarize, the angular distribution of the detected particle in a two body nuclear reaction exciting a single resonant state in the compound nucleus with a spinless projectile and a spinless ejectile may be expressed as:

$$\frac{d\sigma}{d\Omega}(\theta_{cm}) = B_0 + B_2 P_2(\cos \theta_{cm}) + B_4 P_4(\cos \theta_{cm}) + \dots + B_{2L} P_{2L}(\cos \theta_{cm}) \quad (2.58)$$

where, for historical reasons related to the fact that in the inequality 2.57 above only angular momenta are involved, it is customary to indicate $n_{max} \equiv 2L$, and then $L = \min(J, \ell_i, \ell_f)$. The understanding of the *limit to the complexity* (i.e. the existence of a cut-off value) in the Legendre polynomial expansion of an angular distribution in a compound nucleus reaction represented a point of honor of Nuclear Physics towards the development of Advanced Quantum Mechanics, and it is historically interesting by

itself⁵. It is important to note that, if two partially overlapping resonances with *opposite parities* are involved, *odd* Legendre terms appears in the series (but with maximum order not exceeding the limitations discussed above).

If the colliding particles have a spin, the scheme discussed above can still be used, but with the care of considering the fictitious *channels spins* $\vec{s}_i = \vec{s}_{proj} + \vec{s}_{targ}$ for the entrance channel and $\vec{s}_f = \vec{s}_{eject} + \vec{s}_{resid}$ for the exit channel. The coupling of angular momenta then becomes:

$$s_i(\ell_i)J(\ell_f)s_f \quad (2.59)$$

and several possible values of channel spins can contribute to the formation of the same resonant state⁶. While the limitations to the complexity given by the inequality 2.57 are still the same, a modification occurs in the $A_n(i)$ and $A_n(f)$ coefficients of the Legendre expansion, always involving, however, the Racah coefficients. One then obtains $A_n(i) = F_n(\ell_i \ell'_i s_i J) \cdot a_n(\ell_i \ell'_i)$ and $A_n(f) = F_n(\ell_f \ell'_f s_f J) \cdot a_n(\ell_f \ell'_f)$ and, for the case of *pure* transition (i.e. with just one possible value of ℓ_i and ℓ_f) finally:

$$\begin{aligned} \frac{d\sigma}{d\Omega}(\theta_{cm}) &= \sum_n A_n(i)A_n(f)P_n[\cos(\theta_{cm})] \\ A_n(i) &= F_n(\ell_i s_i J) \cdot a_n(\ell_i \ell_i) \\ A_n(f) &= F_n(\ell_f s_f J) \cdot a_n(\ell_f \ell_f) \end{aligned} \quad (2.60)$$

where, with obvious notations:

$$\begin{aligned} a_n(\ell\ell) &\equiv \frac{2\ell(\ell+1)}{2\ell(\ell+1) - n(n+1)} \\ F_n(\ell s J) &\equiv (-1)^{s-J-1}(2\ell+1)\sqrt{2J+1} \cdot (\ell\ell 1 - 1|n0) \cdot W(JJ\ell\ell; ns) \end{aligned} \quad (2.61)$$

where the dependence of the $F_n(\ell s J)$ *nuclear reaction coefficients* on the Wigner and Racah coefficients is evident. It is trivial to calculate, by hand, the so called *spinless particle parameters* $a_n(\ell\ell)$, while it is more complicated to calculate the nuclear reaction coefficients (but still feasible with the use of suitable tables [101], or by using the 3-*j* and 6-*j* symbols of Wigner [46], that are easily correlated to the Wigner and Racah coefficients). However, to simplify the life of experimentalists, in Ref. [145] plenty of tables with ready-to-use $F_n(\ell s J)$ coefficients are reported for many values of angular momenta.

Now, let's make some important examples to fix the ideas here discussed. As a first case to demonstrate the (enormous) power of the theory derived above one can make a *by hand* calculation of the angular distribution expected for a $J = 1^-$ resonance in ^{20}Ne

⁵Brief (and edifying) story: (1941) Charles Critchfield and Edward Teller shot a *conjecture* on this limitation of complexity in angular distributions on a Physical Review article, just driven by their intuition and good sense. (1947) Elmer Eisner and Robert G. Sachs turned out to find a quantum mechanical demonstration of the conjecture based on symmetry arguments. (1948) Chen Ning Yang demonstrated the validity of the conjecture also for relativistic particles, a fundamental job to theoretically treat the β decay process. (1952) John Blatt and Lawrence Biedenharn nestled the (already proven) conjecture inside the full scheme of angular correlation theories, making full use of the properties of Wigner and Racah coefficients.

⁶In this unfortunate, but quite common, case, one will have to calculate the angular distribution pertaining to a given channel spin and sum it incoherently, with a weighting factor, to the angular distribution associated to the alternative channel spin option(s). Weighting factors must be determined experimentally from the data fitting.

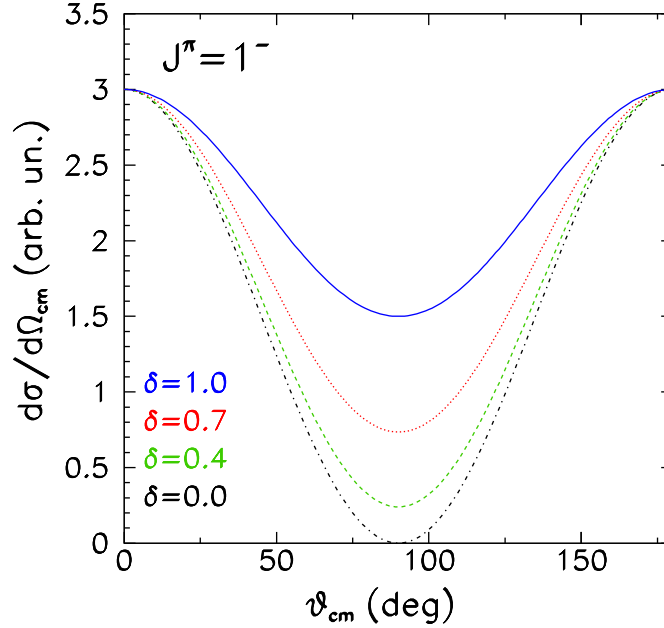


FIGURE 2.7: Trend of angular distribution expected for a $J^\pi = 1^-$ isolated resonance populated in the $^{19}\text{F}(\text{p},\alpha)^{16}\text{O}$ (see the text for the discussion of the coupling of angular momenta in the entrance and exit channels). The δ options are referred to different weights between the $s_i = 0, 1$ entrance channel spin options. In the R -matrix theory, such weights are connected to the ratios between the partial widths for the entrance channel in the $s_i = 0$ and $s_i = 1$ options.

compound nucleus populated by the $^{19}\text{F}(\text{p},\alpha_0)^{16}\text{O}$ that will be studied experimentally in Chapter 5. Since both proton and ^{19}F have $J^\pi = 1/2^+$, two different choices of channel spins will be possible for the entrance channel, i.e. $s_i = 0, 1$. In the exit channel, instead, $s_f = 0$. One must then consider two angular momentum coupling schemes: $s_i(\ell_i)J(\ell_f)s_f = 0(1)1(1)0$ and $s_i(\ell_i)J(\ell_f)s_f = 1(1)1(1)0$. In both cases, $n_{\max} = 2L = \min(2, 2, 2)$, therefore in both case the angular distribution should have the limiting form $1 + A_2(i)A_2(f)P_2[\cos(\theta_{cm})]$.

Let's calculate the $A_2(i)$ and $A_2(f)$ coefficients for both the cases. When $s_i = 0$, it follows $s_i(\ell_i)J(\ell_f)s_f = 0(1)1(1)0$ and $A_2(i) = a_2(\ell_i\ell_i) \cdot F_2(\ell_i s_i J) = a_2(11) \cdot F_2(101) = -2 \cdot \frac{\sqrt{2}}{2} = -\sqrt{2}$, where the value $F_2(101) = \frac{\sqrt{2}}{2}$ was taken from Table I(a) of Ref. [145]. *Simili modo*, for the exit channel: $A_2(f) = a_2(\ell_f\ell_f) \cdot F_2(\ell_f s_f J) = a_2(11) \cdot F_2(101) = -2 \cdot \frac{\sqrt{2}}{2} = -\sqrt{2}$. Then, for the channel spin *zero* branch, the result is $\frac{d\sigma}{d\Omega}(\theta_{cm}) = 1 + (-\sqrt{2}) \cdot (-\sqrt{2})P_2[\cos(\theta_{cm})] = 1 + 2P_2[\cos(\theta_{cm})]$.

When $s_i = 1$, consequently $s_i(\ell_i)J(\ell_f)s_f = 1(1)1(1)0$ and $A_2(i) = a_2(\ell_i\ell_i) \cdot F_2(\ell_i s_i J) = a_2(11) \cdot F_2(111) = -2 \cdot (-\frac{\sqrt{2}}{4}) = +\frac{\sqrt{2}}{2}$, where the value $F_2(111) = -\frac{\sqrt{2}}{4}$ was taken from Table I(a) of Ref. [145]. The exit channel coefficient $A_2(f) = -\sqrt{2}$ is the same as before, because the right semi-reaction has the same angular momentum coupling as the previous case. Then, for the channel spin *one* branch, $\frac{d\sigma}{d\Omega}(\theta_{cm}) = 1 + (+\frac{\sqrt{2}}{2}) \cdot (-\sqrt{2})P_2[\cos(\theta_{cm})] = 1 - P_2[\cos(\theta_{cm})]$.

The final result is a linear combination of the two cases; indicating with δ^2 the (positive) relative weight between the two channel spin cases, finally:

$$\frac{d\sigma}{d\Omega}(\theta_{cm}) = (1 + \delta^2) + (2 - \delta^2) \cdot P_2[\cos(\theta_{cm})] \quad (2.62)$$

and one can therefore expect that the expansion of experimental angular distribution data in Legendre series would have just the B_0 and B_2 as the only *non-vanishing* coefficients. Figure 2.7 shows the trend of the expected angular distributions as a function of several δ^2 values. The huge power of this approach in assigning the J^π of resonances in compound nucleus experiment is then evident. Obviously, these type of calculations can be generalized to all the possible angular momentum coupling for a generic reaction.

Let's make another example: in this case, the angular momenta inequality seen before will be used, i.e. $\ell_i \geq L, J \geq L, \ell_f \geq L$, together with other useful considerations based on conservation rules and symmetries present in a reaction driven by the strong force:

- Parity conservation rules;
- symmetry due to the presence of identical zero-spin bosons in one channel;
- Isospin conservation rules,

to derive a J^π assignment for a resonant state seen in a typical compound nucleus experiment.

The focus is on the ${}^7\text{Li}(p,\alpha){}^4\text{He}$ reaction data in the region of bombarding energies between $E_p \simeq 3.0 - 3.1$ MeV. Experimental excitation functions show a bump with a Breit-Wigner shape, associated to the resonant excitation of the 19.9 MeV state in ${}^8\text{Be}$ ($E_x = E_{cm} + S_p \simeq \frac{7}{8}E_p + 17.2 \text{ MeV} \simeq 19.9 \text{ MeV}$). The experimental angular distribution of the cross section in this region can be described excellently by including in the Legendre expansions only the B_0, B_2 and B_4 terms. This means that $L = 2$, and that $\ell_i \geq 2, J \geq 2, \ell_f \geq 2$. But in the exit channels there are two identical spin-zero bosons: this means that the exit channel spin is zero, and therefore $J = \ell_f$. Furthermore, only natural parity states are allowed: $\pi = (-1)^{\ell_f}$. But the identity of the bosons further restricts the parity of the wavefunction to be *positive*! This means that only $0^+, 2^+, 4^+, 6^+ \dots$ states in the compound nucleus are allowed. But the fact that $\ell_f = J \geq 2$ immediately discards the 0^+ case. Now, about the analysis of the entrance channel: the intrinsic spin and parities of the proton and ${}^7\text{Li}$ are respectively $1/2^+$ and $3/2^-$, leading to two possible values of channel spin (with negative channel parity): $s_i^\pi = 1^-$ or 2^- . This means that, because of the conservation of parity, $\ell_i = 1, 3, 5, 7 \dots$; however, since $\ell_i \geq 2$, the $\ell_i = 1$ case must be discarded. Therefore one has to choose between $\ell_f = 2$ or 4 or $6 \dots$ and $\ell_i = 3$ or 5 or $7 \dots$, with the constraint given by the limitation to the complexity of the angular distribution, i.e. that at least one of the three number ℓ_i, J, ℓ_f must be equal to $L = 2$. Then, the J^π of the 19.9 MeV state must be 2^+ . One can finally unveil the ℓ_i value: the coupling of the candidate value $\ell_i = 3$ with the entrance channel spin $s_i = 1, 2$ allows, for both cases, to form the $J = 2$ value needed.⁷

⁷In fact, being $\vec{J} = \vec{s}_i + \vec{\ell}_i$, from the addition theorem will follow that $|s_i - \ell_i| \leq J \leq s_i + \ell_i$, and this means $|1 - 3| \leq J \leq 1 + 3$ and $|2 - 3| \leq J \leq 2 + 3$ for the two channel spin options. Then $2 \leq J \leq 4$ and $1 \leq J \leq 5$, and in both cases the $J = 2$ value is allowed in the coupling. This is *not true* for the $\ell_i \geq 5$ options: for example, for $\ell_i = 5$ it would be $5 \leq J \leq 6$ and $3 \leq J \leq 7$.

When dealing with several resonances partially overlapping, it is possible to describe the experimental angular distributions with a Legendre polynomial expansion *truncated* to a suitable $2L$ value taking into account the strong *hindrance* of high order partial waves due to the tunneling of the centrifugal barrier. This is a situation commonly occurring in nuclear reactions at well sub-barrier energies (as in nuclear astrophysics scenarios); the presence of bumps in even B_i coefficients can be then used to *suggest* the J^π of the resonance, together with the consistent application of all the other conservation laws discussed above. In this (more complex) case, also the *odd* B_i terms can bear a lot of information: in fact, the presence of *bumps* or *dips* in odd-coefficients is associated to *interferences* between *two* neighboring *opposite-parity* states [145–147]. This property has often a paramount importance in the disambiguation of alternative J^π assignments.

An interesting application on this aspect to a complex case is discussed in Ref. [148], where clear interference effects were observed in the following reactions: $p+{}^9\text{Be} \rightarrow d+{}^8\text{Be}$ (with intrinsic spins $\frac{1}{2}^+ + \frac{3}{2}^- \rightarrow 1^+ + 0^+$), and $p+{}^9\text{Be} \rightarrow \alpha+{}^6\text{Li}$ (with intrinsic spins $\frac{1}{2}^+ + \frac{3}{2}^- \rightarrow 0^+ + 1^+$). These reactions can clearly populate *unnatural parity* states and *surely* involves $\ell_i \neq \ell_f$ (the *channel parities* are opposites, *negative* for $p+{}^9\text{Be}$ and *positive* for $d+{}^8\text{Be}$ or $\alpha+{}^6\text{Li}$ channels), but the interference rule was still thoroughly used in that work to deduce spectroscopic information on close-lying states in the ${}^{10}\text{B}$ compound nucleus.

2.4 Clustering in atomic molecules and molecular cages

In Chapter 1 it was discussed that also *atomic molecules* can interact by means of relatively weak forces, essentially of electrostatic or electrodynamic nature. Such forces, as the van der Waals, Keesom and London ones, have a behavior very similar to the $\alpha - \alpha$ effective forces, provided that adequate re-scalings are applied to the investigated system. Obviously, a large difference with the nuclear case is related to the fact that, in the chemical environment, sizable fractions (or even multiples) of Avogadro number of interacting elements (the molecules) are typically under investigation, allowing for the analysis of macroscopic quantities related to optical and/or thermodynamical properties of samples.

An interesting example of molecular clustering in soft matter, driven by a *variant* of the Keesom forces, has been recently investigated starting from the very peculiar distribution of the electronic cloud in a halogen atom bound inside an organic molecule. As discussed in Section 1.1.6.2, the presence of an electropositive σ -cap can trigger quite strong halogen bonds with other molecules, leading to a *dimeric* re-organization of the molecules inside the solution. But the occurrence of a *negative charge belt* in the equatorial region of the bound halogen can likewise trigger the formation of other intermolecular bonds, again based on Keesom interaction, with peculiar *geometrical arrangements* taking into account all the interactions between the local dipoles. An interesting example of this type of molecular clustering was discussed in the very recent work of Ref. [22], where the intermolecular bindings in several mixtures of Iodomethane (CH_3I) and Ethanol ($\text{C}_2\text{H}_5\text{OH}$) molecules were studied by means of Infrared (IR) and Raman spectroscopy, with the aim of highlighting the formation of oligomeric structures in the samples on the basis of *cooperative interactions* given by the superimposition of hydrogen bonds and halogen bonds.

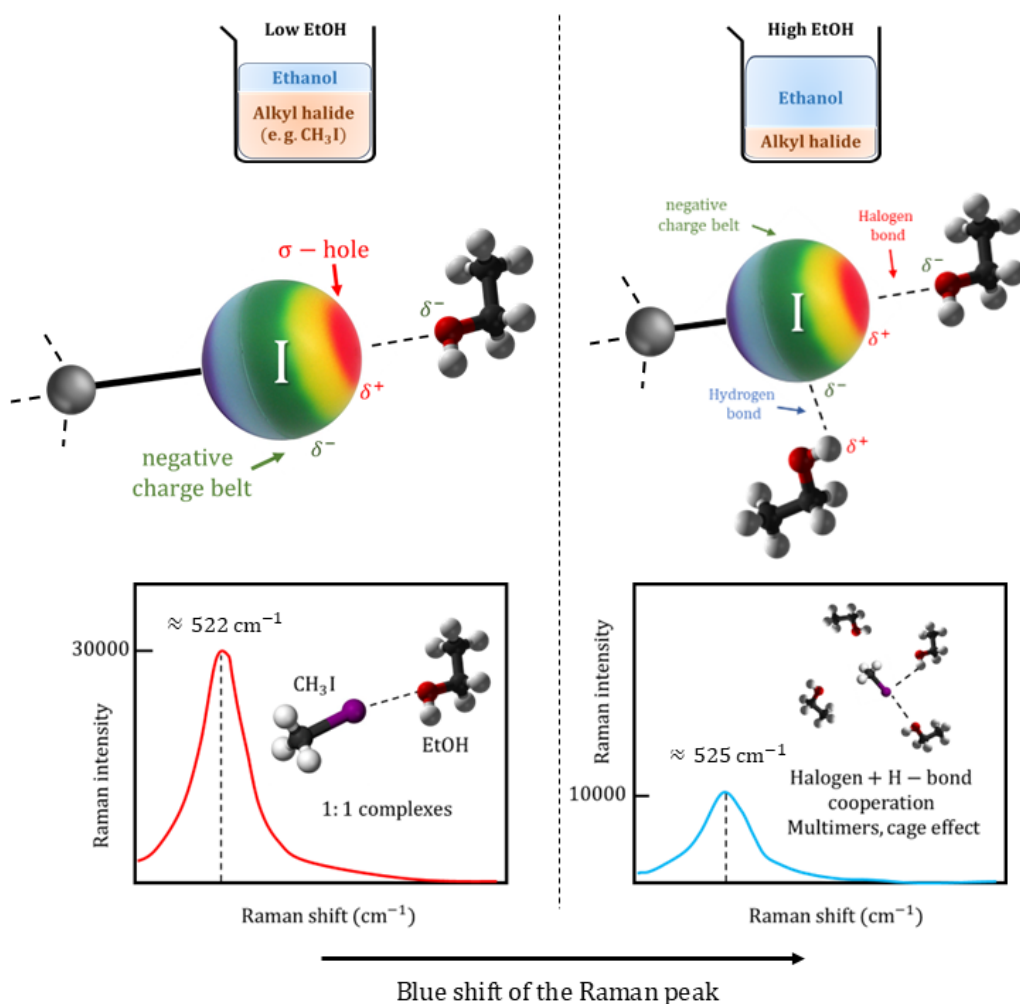


FIGURE 2.8: Schematic representation of the different interactions taking place between methyl iodide and ethanol molecules in mixtures with different concentrations of ethanol. The interaction behavior is monitored and reported through Raman spectroscopy: at low alcohol concentrations, the two species are interacting with either halogen or hydrogen bonds, and the Raman peak of the C-I bond in the CH_3I molecule is centered at around 522 cm^{-1} . For higher ethanol concentrations, the C-I peak is blue-shifted towards higher wavenumbers (now $\approx 525\text{ cm}^{-1}$), because of the onset of cooperation effects between the hydrogen and halogen bonds, and the consequent formation of multimer configurations which act like solvent cages. Re-built from data of Ref. [22]

In Ref. [22] it was observed that, when the composition of the mixture is at 1:1 ratio, molecular dimers between the two different molecular species (Iodomethane and Ethanol) are mainly formed, on the basis of a single interaction of hydrogen-bond or halogen-bond nature. On the other hand, following the increase of the ethanol concentration, the $\text{C}_2\text{H}_5\text{OH}$ molecules start to form a network built on *both* hydrogen and halogen bonds, which may host CH_3I molecules, with a strong *trimeric* structure leading to marked cooperative effect. This beautiful type of cooperation between halogen and hydrogen bonds is favored by the high polarizability of the iodine atom and by the increase of the σ -hole on the halogen in the CH_3I molecule, due to the electron withdrawing characteristics of the $-\text{CH}_3$ group. The qualitative behavior of the intermolecular bonds that leads to the clusterization of molecules is sketched in the second

row of Figure 2.8: it is possible to appreciate the *directionality* of the two competing bonds triggered by the non-uniform electronic distribution in the iodine atom.

The experimental evidence of this molecular re-arrangement was based on the fact that both the solvent and the hydrogen-bond frameworks act like a cage, modulating the vibrational frequencies of the C-I and O-H bonds. More specifically, the Near Infra Red (NIR) spectroscopic analyses show variations in the O-H overtone band, while the Raman spectra exhibit blue-shifts of the C-I band with increasing ethanol concentration (see Figure 2.8, third row). This last point is important to understand the observed phenomenon. The presence of a blue-shift in the Raman spectrum clearly indicates the *reduction* of the bond length (leading to an *increase* of the vibration frequency, and therefore to the blue-shift in the Raman spectrum). This (small, but non negligible) reduction can be attributed to the formation of a *cage* of molecules, interacting via the Keesom forces with the iodomethane, that would pack more tightly the (main) C-I bond. A similar shrinking effect is also seen in the C-H bonds in iodomethane. Other indications on the presence of this effect are suggested by detailed Molecular Dynamics (MD) calculations, taking into account all local dipole distributions in the molecules [22].

Optical spectroscopy, however, may not be the only way to study cage or solvent effects in solutions; given that the onset of the cages is favored in the presence of Ethanol and Alkyl Iodides mixtures, these can be formed also in radiochemical frameworks, like during the study of the Szilárd-Chalmers reaction in Ethyl Iodide-Ethanol mixtures, as shown in Chapter 6 of this thesis. A brief recall of the Szilárd-Chalmers process, one of the most beautiful links between chemistry and nuclear physics, will be given in the following.

The aim of this process, described in the original work from Leo Szilárd and Thomas A. Chalmers, [149] is to separate radioactive isotopes from original matrices of chemical compounds (usually organic molecules). First, the organic molecule is irradiated with thermal neutrons, for which the target nucleus (as the Iodine atom bound inside an alkyl-iodide molecule) can have a very high capture cross section (e.g., 6.2 barn for ^{127}I). The collision of the *thermal* neutron on the target nucleus is not able at all to break its bond with the rest of the molecule: its energy, of just ≈ 0.025 eV, is not enough to even tickle, e.g., the C-I bond of an alkyl-iodide molecule, whose energy is about 2.32 eV. However, the emission of one or more γ -rays from the formed excited ^{128}I produces a recoil energy higher than 100 eV [150–152], well larger than the binding energy of the molecule, freeing the target atom (Iodine) from its molecule. The radioactive atom will then move with kinetic energies (of the order of 100 eV) much larger than the thermal energy: it is called a *hot atom* [150] and it will lose energy by collisions with the surrounding atoms up to thermalization. The freed iodine atom can be extracted from the organic phase through a reducing aqueous solution, and a subsequent precipitation with a suitable salt, like silver nitrate (AgNO_3) in the case of iodine. To facilitate the collection of radioactive species, some tiny quantity of stable *carrier* (as bi-sublimated molecular I_2 in the case of Iodine) is added before the irradiation. A scheme of this very important radiochemistry process is reported in Figure 2.9.

Unfortunately, the yield of the radionuclide is not always unitary, and some *retention* effects may interfere with the extraction of the Iodine, as it will be discussed in detail in Chapter 6. The fast motion of the *hot atom* in the solution forms a lot of *reactive radical* species, which can recombine and react with the iodine atom in very fast ways [151,

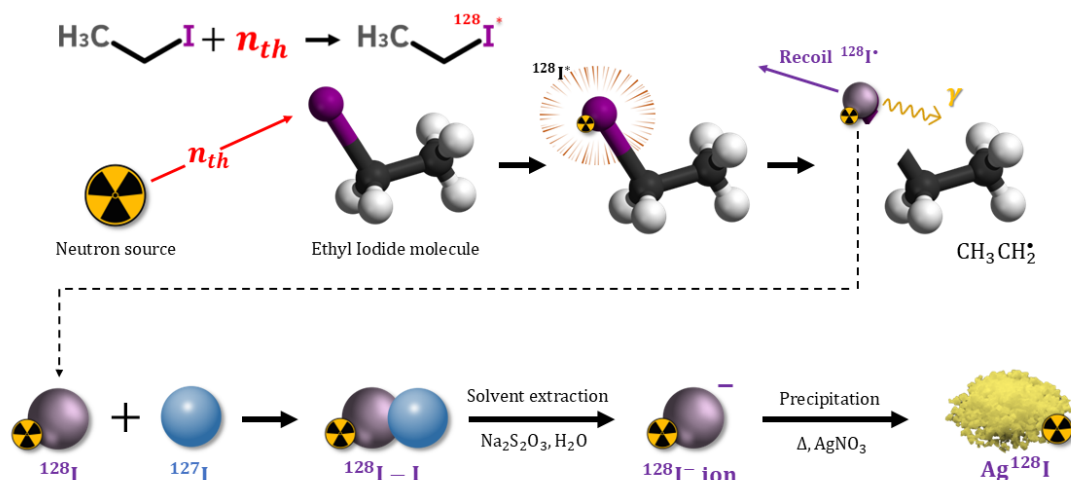


FIGURE 2.9: Scheme describing the Szilárd-Chalmers process. *Top row*: from the capture of the thermal neutron to the emission of the γ ray and the breaking of the C-I bond due to recoil energy of the ^{128}I atom. *Bottom row*: sequence of the chemical processes necessary to extract and precipitate the radioactive ^{128}I

[152]. In this framework, *cage effects*, firstly introduced by James Franck and Eugene Rabinowitch in 1934 [153], were often suspected, in radiochemical experiments, to play a role in the re-entering of the radioactive atoms inside a molecule of the irradiated sample, increasing the retention. The onset of *cage* regions is typical of chemical reactions involving the formation of radicals starting from non-radical precursors [154], as it occurs when fast "hot" atoms collide with other molecules of the samples.

To avoid this phenomenon, the action of *scavenger* molecules (like *Ethanol*, *Aniline*,...), [152] is typically used to *unravel* the *radical nest* [151] formed around the radioactive, hot atom. Clearly, since the formation of solvent networks or cages by intermolecular forces may interfere with the movement of the hot ^{128}I radioactive atom and on the formation of more intricate radical nests, it is extremely interesting to investigate if this type of molecular clustering has a role in the determination of retention effects. In a simple geometrical framework, in fact, a radical nest could be easily formed inside an ensemble of well clustered molecules, characterized by oligomeric structures, as discussed before starting from the observations of Ref. [22]. Chapter 6 will report data aiming at exploring molecular clustering in the framework of a nuclear chemistry experiment.

This is the final knot ending a cycle of parallelisms which started when some chemical approaches, like the crystallographic technique and the LCAO model were mirrored and applied to nuclear frameworks. Here, nuclear techniques like the neutron capture reaction leading to the emission of the hot atom by recoil and the nuclear detection of its decay products are instead used to study delicate molecular clustering phenomena, triggered by weak Keesom-like forces, which were in turn also paralleled to the interactions between different α clusters in nuclei. The remaining Chapters of this thesis will thus be dedicated to the presentation of all the works in which these parallelism, up to now only described in theoretical terms, were experimentally applied.

Chapter 3

Structure of ^{12}C in its ground and Hoyle states

The different isotopes of Carbon, the most important element for life as it is known in the Universe, are characterized by one main feature: the possibility to exhibit a structure based on three α clusters (see, e.g., Sect. 1.2.3.7). It has already been discussed, in Chapter 1, how the insurgence of clustering can modify the density distribution of matter with respect to an independent particle approach (see Fig. 1.29).

The most abundant isotope of carbon is ^{12}C , mainly forged in stars through a nuclear reaction involving the collision of three α particles. However, despite taking place in Helium-rich environments (like the insides of Red Giant stars), a simultaneous collision of three α particles, defined as the "direct" mechanism for the synthesis of ^{12}C through the three- α process, is highly suppressed by phase-space considerations. For this reason, an alternative *sequential* process was proposed by Salpeter and Opik [155], based on the collision between an α particle and an already-formed ^8Be nucleus. Nonetheless, the half-life of the ^8Be is extremely low ($\approx 10^{-16}\text{s}$); so, this mechanism alone is not sufficient to explain the abundance of ^{12}C in the Universe.

To justify the rate of formation of ^{12}C in Red Giant stars, some feature leading to a strong enhancement of the $^8\text{Be}+\alpha$ reaction cross section is strongly needed. A revolutionary idea, developed by Fred Hoyle in the '50s [156], was to suggest the existence of a state, in the ^{12}C level scheme, lying at an energy very close to that of the $^8\text{Be}+\alpha$ system, which could give rise to a strong *resonant process* in the cross section at colliding energies similar to thermal motion of the two nuclei in Red Giants environments (i.e., a few hundreds of keVs). The $^8\text{Be}+\alpha$ threshold in ^{12}C is of 7.367 MeV, being very low because of the occurrence of strong α clustering structure (see Sect. 1.2.3.2). The existence of a state such as the one predicted by Hoyle, a 0^+ state lying at $E_x = 7.654$ MeV, was indeed experimentally verified by W. A. Fowler just a few years later. This state, which is the second excited state in the ^{12}C level scheme, and the first 0^+ after the ground state itself, is nowadays referred to as the *Hoyle state* (for some historical note see, e.g., Ref. [95]). Because of its strongly clustered nature, the Hoyle state was often considered as the *archetype* of a nuclear molecule, and its theoretical knowledge still represents a challenge in modern nuclear physics [133, 157].

However, several models, starting from the pioneer one of Wefelmeier (Section 1.2.3) predicts that also the *ground state* of the ^{12}C nucleus can show some signals of a 3α

cluster configuration, seen as a transient disposition of the nucleons, in competition with the more likely spheroidal-like structure, predicted by shell and the Hartree-Fock model calculations.

Naturally, in a *dynamical vision* of the structure of the nucleus, depending on the particular state considered, the *weight* of the cluster configuration (i.e., the "time" percentage the nucleus spends in this structure) can deeply vary, as a function of the properties of the state. If the Hoyle state at 7.654 MeV is more commonly accepted to be described through a three α particles cluster structure [48, 93, 121], the situation is not as straightforward for the ^{12}C ground state, whose structure is mainly described by the spheroidal configuration [121, 158]. Indeed, in the '70s, different experiments reported some difficulties in the accurate description of the cross section of the $p + ^{12}\text{C}$ reaction through optical model approaches based only on the presence of a spheroidal structure [117].

The first part of this Section is therefore dedicated to a new analysis on the cluster structure of the *ground state* of ^{12}C by means of a comprehensive analysis of $p + ^{12}\text{C}$ scattering and reaction data at intermediate energies, with a complex model taking into account that the collision between the projectile and the ^{12}C nucleus can occur on a clustered structure, giving rise to marked coherent phenomena due to diffraction effects on the clusters (see Section 2.1.3). The second part of the Section will, on the other hand, be dedicated to the study of the very elusive *radiative decay* of the Hoyle state, that has been subject of a recent re-measurement in Ref. [125], leading to results quite far from the commonly accepted value, and consequently to a revision of the reduced transition probability for the $E2$ transition (see Section 2.2). Obviously, a change of the Γ_{rad} value is also directly impacting on the rate of the triple alpha process, with interesting consequences both in nucleosynthesis calculations of the helium-burning phase [135] and on stellar evolution, as discussed, e.g., in Ref. [136].

3.1 Clustering in ^{12}C ground state through a diffractive method

As the starting point of the analysis, in agreement with ACM calculations [94, 159], it was assumed that ^{12}C in the ground state can have an equilateral triangular structure, thus exhibiting the characteristics of the D_{3h} symmetry group. As discussed in Section 1.1.5, this structure would have three main symmetry elements: (i) a main tri-fold symmetry axis (C_3 element), perpendicular to the plane described by the three α particles; (ii) three two-fold secondary symmetry axes, each one passing through an α particle and orthogonal to the main axis; (iii) an horizontal symmetry plane, highlighting the triangular structure and cutting the α particles in half.

In the framework of the ACM, the Hoyle state is seen as a *breathing mode* of the ground state, i.e., a structure with a larger radius coming from the enhanced separation of the α particles. When adopting these structures, it is possible to predict the level schemes for the ^{12}C ground and Hoyle state bands, as discussed in details in Section 1.2.3.8 by using the Peierls-Yoccoz projection method [79, 100]. Figure 3.1 reports a schematic sequence of the levels predicted by this type of calculations based on equilateral triangular structures.

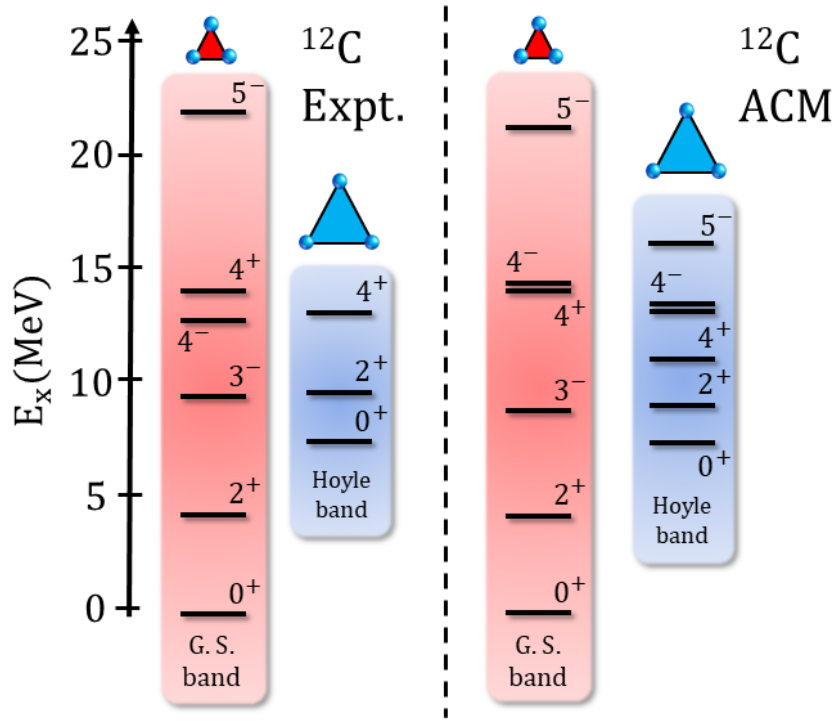


FIGURE 3.1: Comparison between the level sequences of ^{12}C ground and Hoyle rotational bands, as experimentally determined (*left*) and as predicted by the Algebraic Cluster Model (*right*), assuming an equilateral triangular structure for both states. The Hoyle state structure is depicted as a triangle with a large radius with respect to the ground state. Adapted from [94]

In this Chapter, the possible presence of such a symmetrically-arranged clustered structure in the ground state of ^{12}C was investigated by analyzing the data of angular distributions of $p+^{12}\text{C}$ elastic scattering events at intermediate bombarding energies. This study was carried on through the development of a simple model that considers the superposition of effects due to the scattering on a spheroidal-like component (dominant, and described with Coupled Channels calculations in the framework of spheroidal optical potentials [158]) and a cluster component in the target (treated in analogy with the molecular chemistry description of the *Debye-Scherrer* scattering of X-rays on molecules, as discussed in details in Section 2.1.1).

The present model is based on few fundamental assumptions, here listed in an overview:

- for the considered bombarding energy range of data ($p+^{12}\text{C}$ elastic scattering at $E_p = 30 - 80$ MeV) it is possible to neglect the insurgence of compound nucleus effects;
- the ^{12}C ground state is treated as dynamically oscillating between a more common spheroidal configuration of the nucleons and a more *exotic* and rare α -cluster organization as, sketched in Figure 3.2;
- as previously discussed in Section 2.1.3, in this energy domain, the de Broglie wavelengths of the proton beams here considered are of around 3.5 - 5.7 fm, i.e., of the same order of the $\alpha - \alpha$ cluster distance in the carbon nucleus, allowing to observe coherence effects due to the scattering of the waves on the α centers;

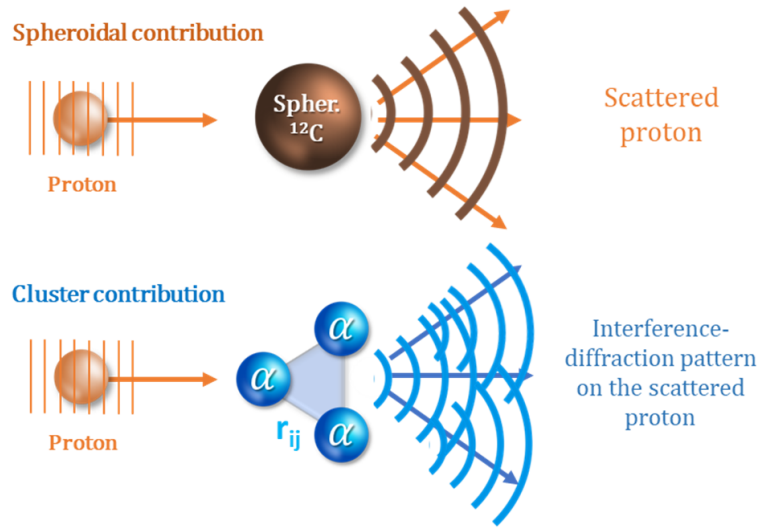


FIGURE 3.2: Schematic representation of the two different contributions expected to make up the description of the experimental data of $p+^{12}\text{C}$ elastic scattering, respectively due to the proton scattering on a spheroidal configuration of the ^{12}C nucleus, described through the Optical Model (*upper panel*), and on the clustered structure, producing coherence effects (*lower panel*).

- as reported in the introductory Chapters of this work (Section 1.2.3.1), the $\alpha - \alpha$ interaction inside the nucleus is extremely weak, and it is thus possible to consider each α cluster as a free and "unbound" scattering center;
- on the basis of calculations through the Algebraic Cluster Model (ACM) [159], it is possible to hypothesize an equilateral triangular α -clusters configuration;
- the angular distributions observed in the works available in the present database are supposed to be given by the superpositions of effects due to the proton scattering on the ^{12}C target nuclei in the spheroidal and in the triangular α -cluster configurations, respectively referred as *spheroidal* and *coherent* contributions;
- as a first-basis approximation, interference effects between the spheroidal and the coherent contributions have been neglected; the inclusion of such effects could be an interesting starting point for future analyses, together with the possibility to use, e.g., neutron scattering data (which would not suffer the complication given by the presence of the Coulomb barrier).

The scattering events imputable to the spheroidal configuration, i.e., the major contribution at any energy, were treated through an Optical Model approach, exploiting Coupled-Channel calculations on the basis of the prescriptions and the optical potential starting parameters reported in [158], that were quite similar to the widely adopted systematics reported by Becchetti and Greenless [160]. Taking into account the coupling of some reaction channels, it is necessary to not neglect the possible excitation of the ^{12}C target during the collision process induced by the proton, which could subsequently influence the elastic scattering cross section. The most important states to include in this *two-way* coupled channel excitation are the ground state, the 4.44 MeV 2^+ state and the 4^+ state at 14.08 MeV, since both of them are part of the ground state rotational

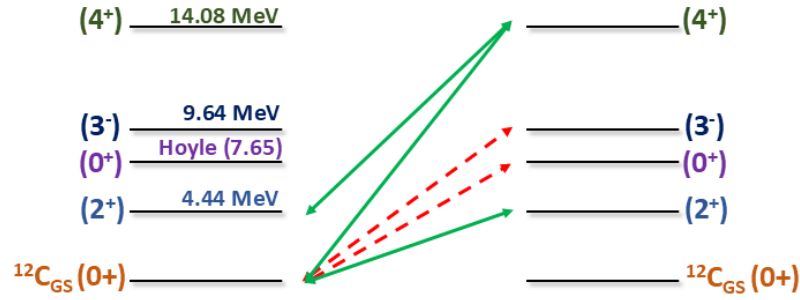


FIGURE 3.3: Allowed (*green solid arrows*) and neglected (*red dashed arrows*) couplings between the ground and the different excited states of the ^{12}C nucleus. The double arrow in each case indicates the two-way channel couplings.

band in ^{12}C . Couplings like those with the Hoyle state (0^+ at 7.654 MeV) and with the 3^- state at 9.64 MeV, related to the octupole and to the β bands, have been in principle neglected since, as already shown in the literature, they would have a quite small effect on the determination of the *elastic* scattering cross section (Figure 3.3) [158].

In the usual Optical Model approaches, both surface and volume terms can be simultaneously used to describe the optical potential generated by the target nucleus. However, a dedicated χ^2 analysis showed that the inclusion of a Wood-Saxon volume term led to no improvements in the fit, either with constant depth or with a typical linear energy dependence; therefore, it was decided to include only the surface term to reproduce the experimental data.

Concerning the evaluation of the contribution to the cross section given by coherence phenomena on the α clusters, the angular distribution $I(\theta)$ was computed through the *Wierl* equation, already discussed in Section 2.1.1:

$$I(\theta) \propto \sum_i |f_i|^2 + 2 \sum_{i,j} f_i \overline{f_j} \frac{\sin(\mu r_{ij})}{\mu r_{ij}} \quad (3.1)$$

In the framework of the onset of α -clustering phenomena in ^{12}C , it is possible to apply this equation adapting it to the equilateral triangle structure for the three α scattering centers in the carbon nucleus and to the wavelength of the matter wave associated with the proton beam. By doing so, the Wierl formula would appear as presented in Eq. 3.1, where r_{ij} is the distance between the i and j clusters (seen as unbound and free scattering centers), μ is dependent from k , the proton beam wave number, and has the expression $\mu = 2k \cdot \sin(\theta/2)$, θ is the scattering angle in the center of mass frame and f_i is the scattering amplitude relative to the i -th cluster, i.e., the scattering properties of a single α cluster took alone. The *complex conjugation* involving one of the scattering amplitudes is due to the fact that in the nuclear reaction theory these can be *complex* functions. Within the present model, the experimental elastic scattering angular distributions would then be described by the expression:

$$\left[\frac{d\sigma}{d\Omega}(\theta) \right]_{\text{exp}} = A \left[\frac{d\sigma}{d\Omega}(\theta) \right]_{\text{spher}} + B \left[\frac{d\sigma}{d\Omega}(\theta) \right]_{\text{clust}} \quad (3.2)$$

being A and B two constants representing the relative weights associated to the spheroidal and coherent contribution to the cross section, respectively. In the above expression,

$(\frac{d\sigma}{d\Omega})_{spher}(\theta)$ is estimated by means of Coupled-Channel calculations performed with the CHUCK code [161], while the $(\frac{d\sigma}{d\Omega})_{clust}(\theta)$ term is estimated by using equation (3.1). In addition, assuming that no further contribution is present, the unitary constraint $A + B = 1$ must be satisfied.

An estimate of the B/A ratio required to describe the data could also represent an useful indicator of the degree of clusterization in the ground state of the target [116]: the B and A parameters can be linked to the projections of the ^{12}C wave function respectively on a clustered and on a spheroidal configuration. Moreover, the ratio:

$$R = \frac{B (\frac{d\sigma}{d\Omega})_{clust}}{A (\frac{d\sigma}{d\Omega})_{spher} + B (\frac{d\sigma}{d\Omega})_{clust}} \quad (3.3)$$

can give an indication of the influence of the cluster component in the variation of the shape of experimental elastic scattering cross section, as discussed in Ref. [117].

3.1.1 Experimental data analysis and application of the model

The model hereby described has been applied to data coming from $p + ^{12}\text{C}$ elastic scattering experiments reported in the literature in the 20th century and collected in the EXFOR database [162] for bombarding proton energies of ≈ 30 to 80 MeV; in this way, the wavelength of the incident beam can lead to coherent scattering from a cluster structure in the ^{12}C nucleus. In details, the used data were taken from Refs. [117, 163–169]. In a similar way, data for the $p + ^4\text{He}$ elastic scattering have been collected from the same database, at six different energies between 31 and 85 MeV [170–173], to compute the $p + \alpha$ -like single center scattering amplitude, required in Equation 3.1, and fitted through the Optical Model approach. The results of the fits are reported in Figure 3.4; a multi-dimensional cubic spline interpolation of all the fit results has then been performed to estimate the values of the scattering amplitudes, $f_i(\theta)$, in the whole energy domain of interest, for all the scattering angles between 0 and 180 degrees.

Concerning the *spheroidal* component of Eq. 3.2, as a first step, Coupled-Channel calculations have been performed by using, as starting values for the optical potentials and the deformation parameters, the ones adopted in Ref. [158]. It was verified that the missing couplings to high energy states in ^{12}C can be very well described with very small variations (of a few percent) in the Optical Model parameters of Ref. [158]. In this way, a fully satisfactory reproduction of the former Coupled-Channel calculations obtained in the energy range $\simeq 30$ -40 MeV was obtained. By extrapolating such calculations to the entire 30-80 MeV energy range of presently published data, that were not available at the time of publication of Ref. [158], it is possible to observe a reasonable agreement, even if some sizable deviations, especially at angles larger than 40° , are still present.

It was therefore decided to improve the data description by performing a comprehensive fit of all the available $p + ^{12}\text{C}$ elastic scattering via a dedicated fit program that uses the CHUCK code to calculate the Couple-Channel contribution. The energy dependence of the amplitude of the real potential and the ground state deformation was fixed to the already-optimized values reported in Ref. [158], while the variation of the real radius and the real and imaginary diffuseness was allowed within $\pm 20\%$ of the values suggested in [158]. The imaginary radius and potential depth were varied within 1.0 – 1.8 fm and 1.0 – 6.0 MeV respectively. Such broader limits account for the large variability reported

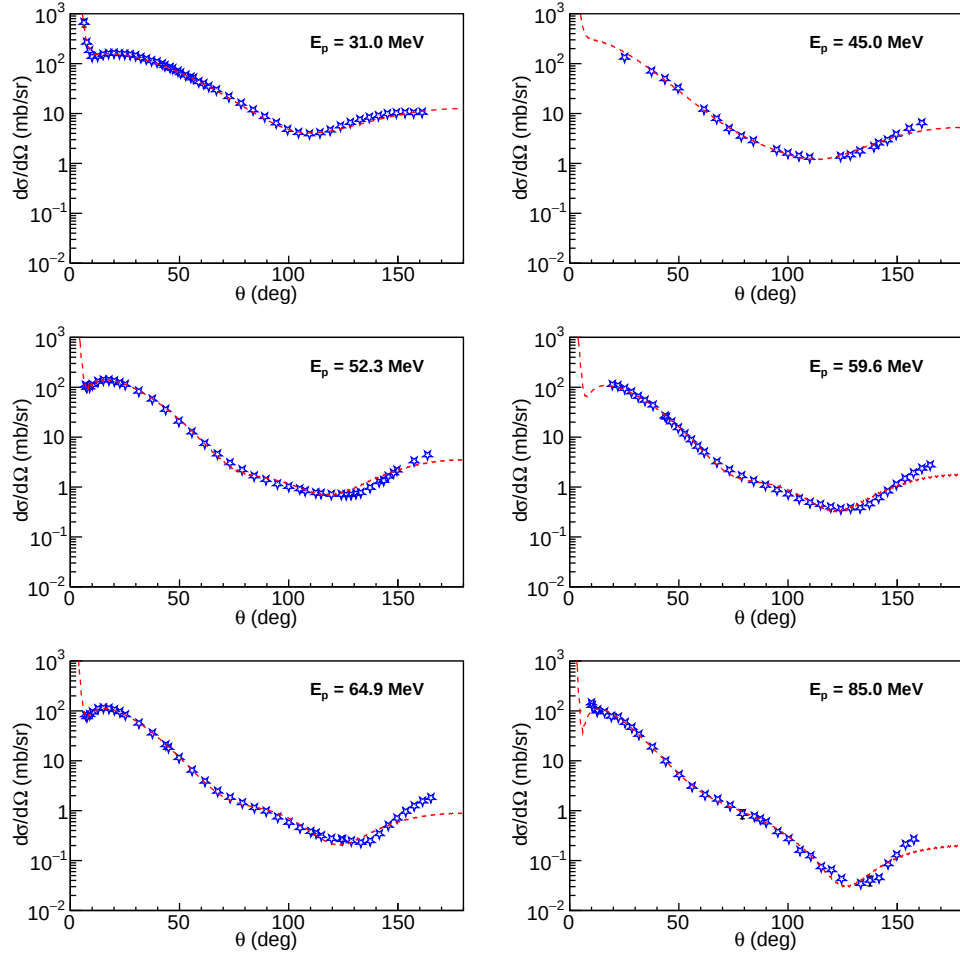


FIGURE 3.4: Angular distributions obtained for $p + {}^4\text{He}$ elastic scattering data in the $E_p = 31 - 85$ MeV region. *Red dashed lines*: the results of the global OM fit based on the entire data set. *Blue stars*: experimental data, Refs. [170] (31 MeV), [171] (45.0 - 64.9 MeV), [172] (85 MeV).

in several optical model compilations (see, e.g., [160, 174, 175]) and, most importantly, can allow a better fit of the present database, which spans a broader energy range (30-80 MeV).

Since both imaginary radius and potential depth strongly determine the behavior of the *reaction* cross sections as a function of energy, data of $p + {}^{12}\text{C}$ reaction cross sections from 30 to 120 MeV, taken from Ref. [176] were also included in the fit. A preliminary χ^2 analysis of the fitting of elastic scattering and reaction data allows to verify, once more, that the inclusion of a *volume term* in the imaginary potential does not lead to an improvement in the description of data. Furthermore, it was verified that the assumption of a linear decrease with energy of the depth of the surface term in the imaginary potential does not improve the description of elastic scattering data and it worsen the reproduction of the slope of the reaction cross section. For these reasons, and in agreement with Ref. [158], the imaginary potential used in the present analysis was represented by a derivative Woods-Saxon form factor with fixed depth.

The results of the Coupled-Channel fit are shown in Figures 3.6 and 3.7 for the elastic scattering data and in Figure 3.5 for reaction data (red dotted lines), together with data

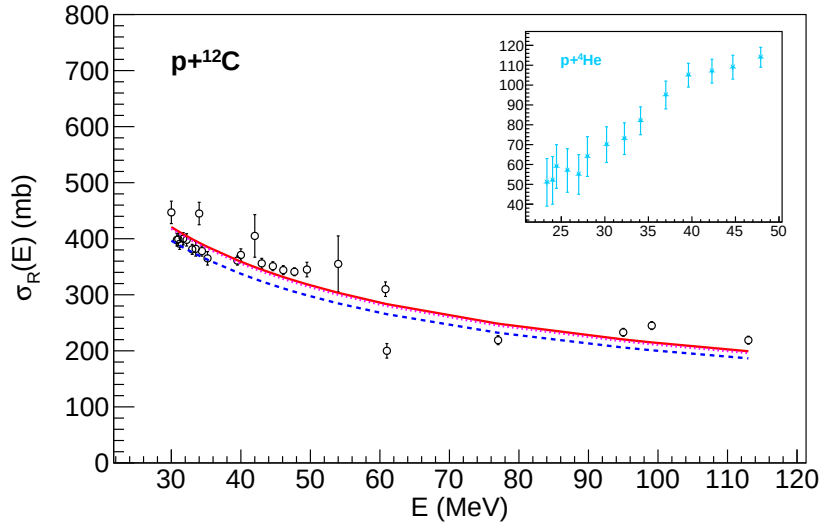


FIGURE 3.5: Plot of the excitation function for the $p + ^{12}\text{C}$ reaction cross section in the intermediate energies range. Experimental data (black circles) from Ref. [176]. Blue dashed line: Coupled-channel calculations based on the starting parameters from Ref. [158]; red dotted line reports CC calculations with the optimized parameters obtained in this work. Finally, the red solid line is referring to the CC calculation made with the parameters from the global fit and with the inclusion of the coherent contribution (see Table 5.1). In the insert, the $p + ^4\text{He}$ reaction cross section data coming from Ref. [176] are shown for comparison.

taken from the literature and the results obtained by using the parameters from Ref. [158] (cyan dashed line); blue dashed lines in Figures 3.6 and 3.7 represent the coherent contribution $B \left(\frac{d\sigma}{d\Omega} \right)_{clust}$. The simultaneously good reproduction of both the datasets gives support to the optimal parameters obtained in the fit and reported in Table 3.1. In particular, the joint presence of a shallow imaginary potential ($W_s = 2$ MeV) and a large imaginary radius ($r_w = 1.67$ fm) appears as the best option to reproduce the absolute values and the slope of the reaction cross section in a very broad energy domain ($\simeq 30$ -120 MeV).

The cumulative distribution of the point-by-point relative errors $\delta(\theta) = \frac{\sigma_{th}(\theta) - \sigma_{exp}(\theta)}{\sigma_{exp}(\theta)}$ for the present fit, alongside with that obtained adopting the starting parameters derived from Ref. [158], are shown in Figure 3.8. The first is the red dashed histogram, obtained with calculated $\delta(\theta)$ from the results of the Coupled-Channel fit. As shown in the figure, it is possible to see how the new Coupled-Channel fit is much narrower and, especially, symmetric if compared to the parallel distribution obtained using the literature parameters of Ref. [158] (green dashed histogram).

After this preliminary work, it was then verified if a comprehensive fit of all the experimental data (scattering + reactions) with a superposition of spheroidal and coherent components, as given by equations 3.1 and 3.2, could further improve the description of experimental data. In this case, the optical model parameters that can be varied during the fit procedure were the same as before, but in addition also the B and r_{ij} parameters of eq. (3.1) (related to the coherent part of the elastic scattering angular distribution) were included in the fit.

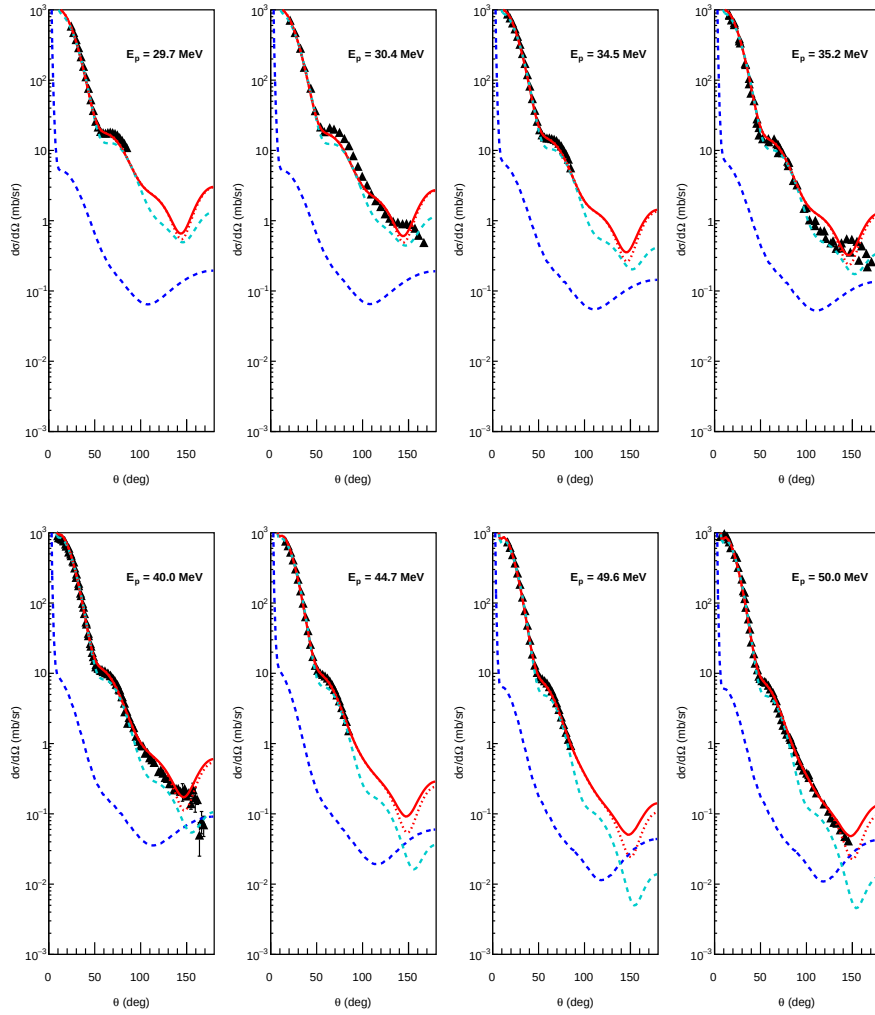


FIGURE 3.6: See caption in Figure 3.7.

	V_0	r_0	a_0	W_s	r_w	a_w	r_c
CC	$59 - 0.34 E_p$	1.06	0.65	2.0	1.67	0.66	1.2
CC + clust.	$59 - 0.34 E_p$	1.05	0.65	2.0	1.67	0.66	1.2

TABLE 3.1: Optical model parameters as used in the present work fits. The depths of the potentials are expressed in MeV, while the reduced radii and diffuseness are given in fm. The quadrupole deformation parameter used in the present analysis is $\beta_2 = -0.663$. The depth of the spin-orbit potential is fixed to 6 MeV.

The contribution due to the coherent part is found to be very small ($B = 0.43 \pm 0.18\%$); nevertheless, its introduction in the fit (and the magnifying effect due to the coherent sum of the three scattering amplitudes coming from the scattering on the α -like clusters) leads to an improvement in the description of data with respect to the case considering only the spheroidal part. In fact, the overall χ^2 is $\approx 10\%$ better when the coherent term is included, and also the $\delta(\theta)$ distribution obtained from the comprehensive fit (blue filled histogram in Fig. 3.8) is narrower ($\approx 5\%$ reduction of the FWHM) than the pure spheroidal case. However, the statistical error on the estimated B parameter coming

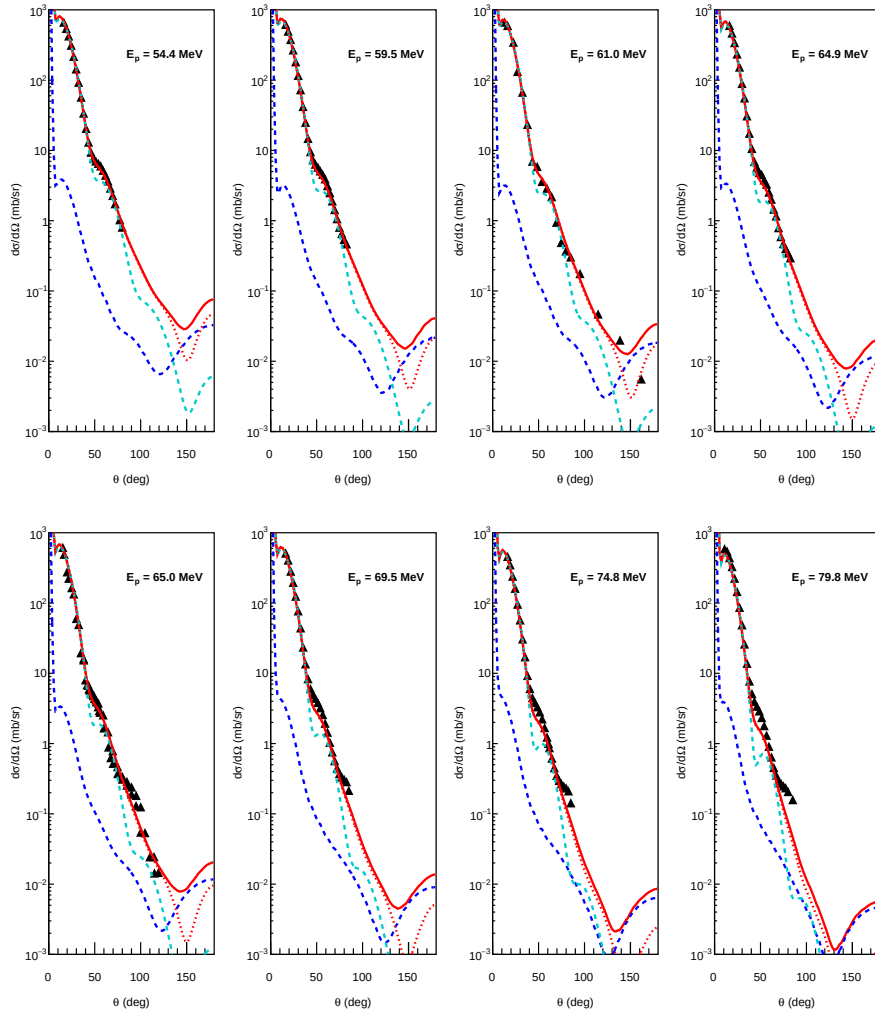


FIGURE 3.7: Angular distributions for $p + ^{12}\text{C}$ elastic scattering at several energies. The black triangles represents experimental data from the following works: Ref. [163] (29.7, 34.5, 44.7, 49.6, 54.4, 59.5, 64.9, 69.5, 79.8 MeV), ref. [164] (30.4 MeV), ref. [165] (35.2 MeV), refs. [117, 166] (40 MeV), ref. [167] (50 MeV), ref. [168] (61 MeV), ref. [169] (65 MeV). Cyan dashed line: Coupled channel calculations on the basis of Ref. [158] original parameters. Red dotted line: CC predictions using the optimized parameters from the spheroidal component fit. Blue dashed line: coherent contribution $B\sigma_{clust}(\theta)$ obtained from the global fit.

from the fit results is also compatible with only an *upper limit* of the presence of a cluster component. In fact, B is found to be compatible with zero within less than 3σ . From the present analysis, it is possible to conservatively suggest a cluster occurrence probability in the ground state of ^{12}C lower than 1.0% at 99.75% confidence level.

An interesting finding coming from the data analysis is related to the inter-cluster distance r_{ij} . The value obtained from the fit is 3.9 ± 0.5 fm, in agreement with previous estimates obtained with similar analysis techniques [117] and it is also in agreement with theoretical calculation based on the Monte Carlo Shell Model of Ref. [121]. Unfortunately, both because of the limited range of data and of the smallness of the cluster contribution found, the inter-cluster distance value here obtained is affected by a sizable

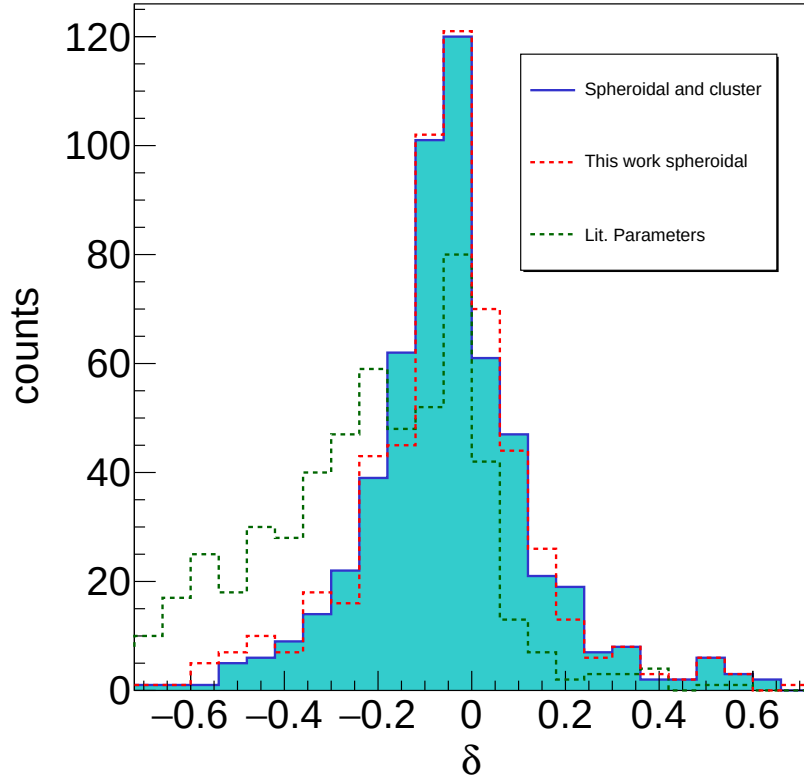


FIGURE 3.8: Normalized residual distribution: $\delta = \frac{\sigma_{th}(\theta) - \sigma_{exp}(\theta)}{\sigma_{exp}(\theta)}$. Green dashed line: Coupled Channels calculations with literature parameters [158]. Red dashed line: Coupled Channels calculations with optimized parameters obtained in the present work. Blue filled histogram: both spheroidal and cluster components were included.

uncertainty, not allowing to discriminate the predictions between several cluster model calculations [119–121].

The ratio of the cluster contribution versus the total differential cross section, Eq. (3.3), is found to have a moderately weak dependence on the energy. The energy- and angle-averaged ratio R (in the $20^\circ - 140^\circ$ angular domain) is found to be of the order of $1/10$. This estimate is close to the older estimate reported in [117].

The present upper limit of the B parameter can be qualitatively compared with theoretical calculations on the fraction of clustering component in the ground state of the ^{12}C wave-function. For example, Ref. [119] reported calculations performed with the Anti-symmetrized Molecular Dynamics (AMD) model. In these calculations, a quite similar cluster probability was found both for the ground and Hoyle states in ^{12}C , being of the order of 40%. More recently, a Monte Carlo Shell Model Calculation making use of Statistical Learning methods for classifying the nature and the behavior of the used basis states [121] pointed out a quite minor role of the α clustering component in the wave function of the ground state, of about 6%. In this context, the upper limit on the clustering component in the ^{12}C ground state deduced here (1%) is of the same order of magnitude of the estimate of Ref. [121], supporting the minor role played by α clustering in the ^{12}C ground state wave function.

One important outcome of the discussed analysis involves the planning of future experiments aimed to unveil the presence of clustering structures in elastic scattering processes. In this framework, the expected effect of the coherent term in modifying the trend of the elastic scattering angular distributions at intermediate bombarding energies was calculated. In performing such calculations, it was assumed that the spheroidal component in the $p+^{12}\text{C}$ elastic scattering is described by the differential cross section previously reported by De Leo et al. in Ref. [158], where a broad Coupled-Channel scheme including the ground state band, the β -band, and the octupole band, was used. Then, by using the Wierl equation (3.1), a percentage deviation (with respect to a pure spheroidal scattering) due to the presence of a 1% contribution of the coherent term (i.e. $B = 0.01$) in the linear combination of Eq. (3.2) was estimated. The trends of percentage deviations as a function of the scattering angle are displayed in Figure 3.9 in the angular region $\theta = 40^\circ\text{--}100^\circ$. In this region, the cross sections are still quite large (> 0.1 mb/sr) and can be reasonably measured even in experiments with tightly collimated detectors (solid angles $\Delta\Omega$ of the order of 0.1 msr, angular opening smaller than 1°), by using beams of 500 nA impinging on targets of $500\text{ }\mu\text{g}/\text{cm}^2$. In similar experimental conditions, it would be possible to expect ≈ 6000 counts per day in the most unfavorable case of 0.1 mb/sr, allowing to reach a *statistical* uncertainty around 1% with two days of beam time. Considering 15 beam energy changes in the 30-80 MeV region, that can be obtained, e.g., at INFN-LNS or at the GANIL cyclotrons, a total of 30 days of beam time would be required to perform this type of experiments, a quite challenging scenario. Furthermore, the calculations were restricted to angles smaller than 100° , also to avoid the complications due to possible interference effects induced by nucleon exchange or compound-like phenomena that could occur at the most backward angles.

The different line with different colors reported in Figure 3.9 indicate calculations performed by assuming several values of the inter-cluster distance, in the range 3.5 - 4.4 fm. The presence of the coherent term would lead to some sizable deviations (of the order of 4-8%) with respect to ordinary spheroidal calculations especially in the angular region around $50\text{--}60^\circ$. In addition, the angular region from 70° to 90° has the largest sensitivity to the inter-cluster distance. Dedicated measurements of the absolute differential cross section with a small angular step and a cumulative uncertainty better than 3% could therefore be useful to shed light on the existence and the magnitude of the coherent term due to clustering in the elastic scattering. The same conclusions can be drawn if the optimized Coupled Channels calculations obtained in the present work are used to describe the spheroidal component in the estimate of the percentage deviations.

Among the concluding remarks, it is interesting to underline the fact that future experiments could indeed be performed using neutron beams as matter waves for similar diffraction studies, given the simplification that would come from the absence of the Coulomb barrier. Obviously, this choice would face off several technical difficulties, being related both to the neutron beam preparation and to the detection of the scattered neutrons. The developments of almost monochromatic neutron beams at the RAON facility in South Korea or the use of time-tagged neutron beams at the n-TOF facility at CERN, coupled with recent successes achieved in fast neutron detection (see, e.g., [177–179]) with reasonably large efficiencies and quite good energy resolutions appear as potentially interesting points of reflection for the planning of further experiments.

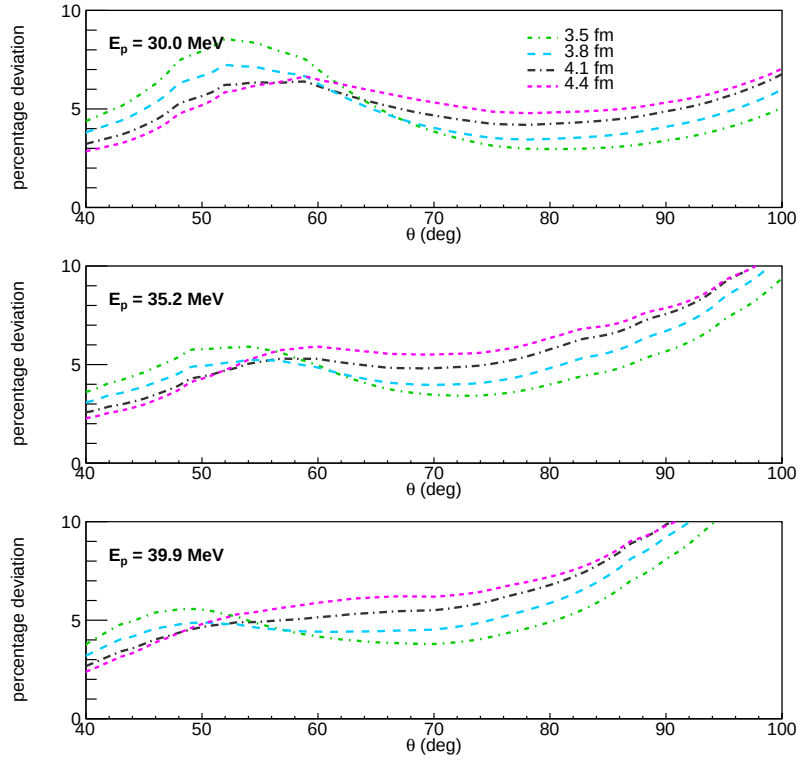


FIGURE 3.9: Computation of the expected percentage deviations for the differential cross sections in $p + ^{12}\text{C}$ elastic scattering events, taking into account a 1% contribution from the *coherent term*. This was calculated assuming different values of the inter-cluster distance, r_{ij} , as reported in the label in the top chart. The spheroidal contributions are taken from Coupled Channel calculations in Ref. [158].

3.2 The structure of the ^{12}C Hoyle state and its radiative decay

As reported in the introductory paragraph of this Chapter, the universally accepted mechanism for carbon formation in stars proceeds through the sequential excitation of the Hoyle state of ^{12}C , a 0^+ state at $E_x = 7.654$ MeV. Nevertheless, after this process, the ^{12}C nucleus is formed in a *metastable* state, not suitable to build compounds and life. To have the possibility to proceed on with the universe's history, it is thus necessary for it to *de-excite* and revert to its ground state. Unfortunately, this process is not as straightforward as it may seem, and does not always proceed through a radiative de-excitation, which would lead to a relaxed ^{12}C nucleus in its ground state: as discussed in Section 2.2, the *radiative decay* is, indeed, very rare. Extremely more common is the *particle decay*, from which the ^{12}C nucleus is separated *again* in the three constituent α particles, a dead end (see Figure 3.10). At the end of the process, the rate of formation of elemental carbon is strongly determined by the competition of these two decay modes, which are deeply tied to the Hoyle state nuclear properties (see Section 2.2).

In spite of the fact that the odds and the properties of the *particle decay* are well known and there is an overall consensus on them [180–182], a general agreement on the *branching ratio* of the radiative decay has yet to be officially and unambiguously reached. Recent measurements [125] have actually suggested an out-of-scale value for the radiative decay branching ratio, around 50% higher than the commonly accepted one,

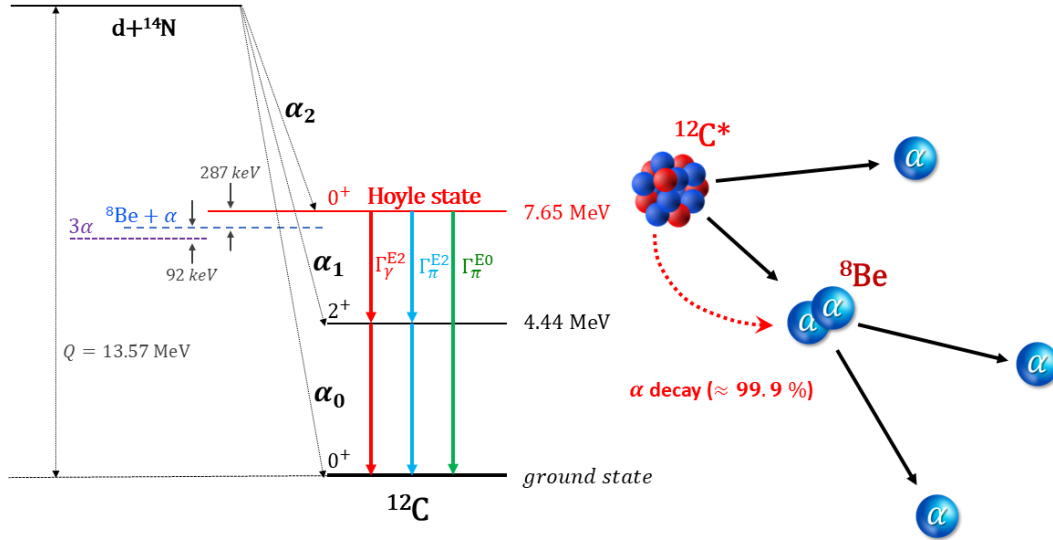


FIGURE 3.10: *Left*: depiction of the level scheme of ^{12}C and the mechanism of population of the Hoyle state through the $d+^{14}\text{N}$ reaction exploited in this experiment. *Right*: schematic representation of the particle decay of the Hoyle state, proceeding through the formation of a ^8Be nucleus.

result of several older experiments [183–186]. Such a higher estimation of the branching ratio value should not be overlooked, since, when coupled with recent pair probability emission [187] it would virtually imply a $\approx 34\%$ increase of the three- α process rate, possibly affecting the balance between carbon and oxygen in helium burning processes and, also, playing a role in determining the boundaries of the pair-instability mass gap for the formation of black holes [136]. In fact, higher productions of ^{12}C during the *three-alpha* process in red giant stars would affect the structure and the development of the stellar nucleus itself, leading to a lower probability for the star to enter the pair-instability mass gap, thus strongly influencing the rates and previsions for the formation of black holes. Obviously, as discussed in Section 2.2, a change in the decay probability via the $E2$ electromagnetic transition in the Hoyle state would change the interpretation of its cluster structure, having therefore an impact also on the nuclear structure side.

The disagreement between data coming from Ref. [125] and two other recent experiments [188, 189] called for an independent verification. A new measurement was even more necessary also considering the fact that a new experiment [190] detecting, for the first time, all the reaction products in a four-fold complete kinematics (see Section 2.2.1) reported a value even larger than that of ref. [125] but with a high uncertainty. All these outcomes led to the development of a new experiment, described in the next Sections, and aimed at measuring the radiative decay branching ratio of the Hoyle state with an innovative setup based on the associated particle technique (Section 2.2.1).

3.2.1 Charged particle spectroscopy and coincidence technique

To contribute to this thorny question, a new, almost background-free, measurement of the radiative decay branching ratio of the Hoyle state was performed, exploiting the associated particle technique through the $d+^{14}\text{N}$ nuclear reaction, producing Hoyle state-excited ^{12}C nuclei in a terrestrial laboratory. The branching ratio of the radiative decay

is thus directly calculated by the ratio of the number of events leading, after radiative emission, to ^{12}C nuclei in the ground state, with respect to the total number of produced Hoyle states measured in a detector in coincidence. This particle-particle coincidence experiment adopted different strategies to maximize the signal to background ratio and to identify, in the clearest way, the counts associated to the radiative decay of the Hoyle state. Among these strategies, for the first time in a charged-particle coincidence experiment, a careful topological study of the detection efficiency of coincident particles was performed along *two* dimensions to build a bi-dimensional efficiency map. In this way, it was possible to operate in a safe range of unitary coincidence efficiency, keeping under strict control one of the most critical sources of systematic uncertainties. The branching ratio value deduced from this experiment is in contrast with the higher one reported in [125] and in excellent agreement with the previous literature estimates [191].

The adopted reaction used to perform the experiment was the $^{14}\text{N}(\text{d}, \alpha_2)^{12}\text{C}$, exploiting the collision between a deuteron beam and the nitrogen contained in thin Melamine ($\text{C}_3\text{H}_6\text{N}_6$) targets, to produce excited ^{12}C nuclei; the subscript "2", referred to the α particle, is indeed indicating the emission of a carbon nucleus in its second excited (i.e. the Hoyle) state. This reaction is important because, at bombarding energies as low as the 2.7 MeV used in this framework, it is particularly selective with respect to low-lying excited states of ^{12}C [192]. A telescope of silicon detectors for charged particles was mounted in the backward hemisphere to measure the energy of the α particle emitted by the ^{16}O compound nucleus, and thus to tag the energy of the recoil excited $^{12}\text{C}^*$ nucleus, as reported by the graphical rendering of Figure 3.11. Through the analysis of this spectrum, it is possible to select the events for which the recoil $^{12}\text{C}^*$ nucleus is populated in the Hoyle state, by restricting the field to a narrow region around the expected α_2 energy. The main advantage of this reaction is its two-body nature and the locked kinematics that follows. The knowledge of the direction and the energy of the backward α particles imposes a strong constraint on the momentum vector of the associated recoil $^{12}\text{C}^*$. Thus, it is possible to accurately know the direction in which another charged particle detector, the *forward* one, should be placed to measure, with high efficiency and low background noise, the coincident recoil after the (very rare) radiative decay to the ground state. In the end, the radiative branching ratio is measured as the ratio of the successful α_2 – ^{12}C coincidences to the single α_2 particles measured by the backward detector, as discussed in the general protocols of associated particle technique in Section 2.2.1.

3.2.2 Experimental set-up

The experiment was performed at INFN - Laboratori Nazionali di Legnaro, using a highly collimated ^2H beam with an energy of 2.7 MeV and typical intensities of 10 - 40 nA; the value of the beam intensity was chosen low enough to minimize the presence of random coincidences. The backward and forward detectors used in the present experiment were both telescopes constituted by two stacked silicon detectors; for the forward telescope the first ΔE unit was a thin 14.8 μm Si detector, while the E_{res} unit was a thicker 500 μm silicon pad. In the backward unit, a 92.9 μm ΔE detector was mounted in front of a 500 μm Silicon pad. To reduce the background coming from unwanted contaminant reactions, both telescopes exploited the anti-coincidence technique, based on a logic condition between the signals produced by the two different stages [192, 193]. The α particles of interest, less penetrating than deuterons and protons, are stopped in the

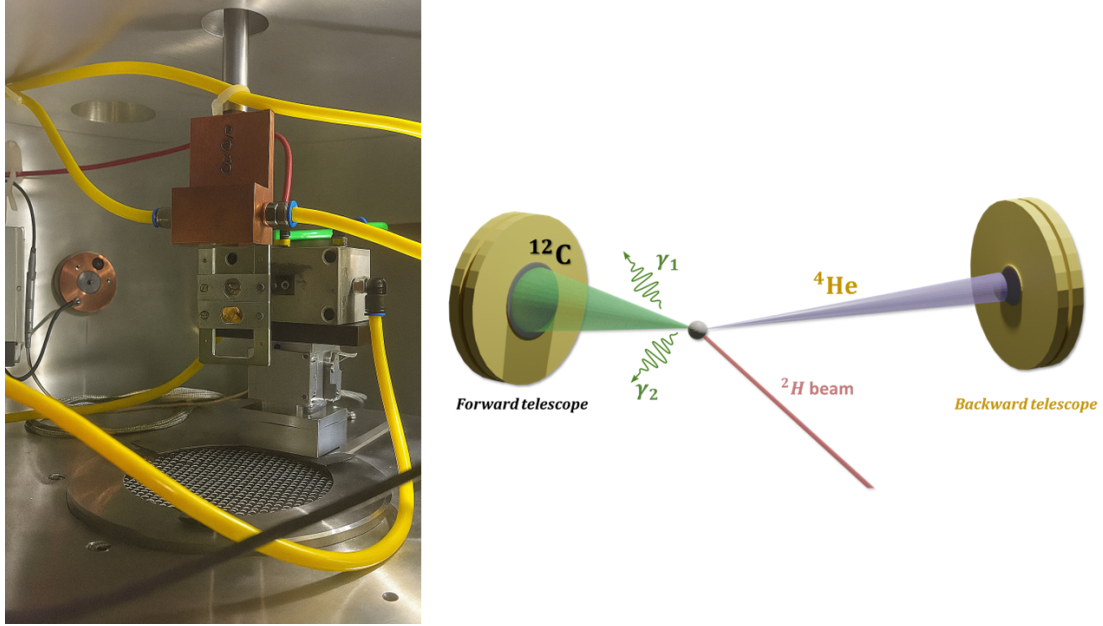


FIGURE 3.11: *Left panel:* Picture of a part of the experimental apparatus as seen from the inside of the reaction chamber. *Right panel:* Three-dimensional conceptual rendering of the reaction and detection processes. The kinematical cones of the tagging ^4He particle and the coincident ^{12}C de-excited nuclei are respectively shown in violet and green.

first stage of the detector, allowing to be detected in *stopping mode*, with an energy resolution better than the case of a *punch-through mode* (where there is the need of summing the energy deposited in the two detectors).

The electronic chain scheme is reported in Figure 3.12; during the course of experimental runs, the detection rate at the forward detector was lower than 1 kHz, while the measured α_2 rate at the forward detector was < 0.1 Hz; due to the width of the time coincidence window, ≈ 5 ns, the expected spurious coincidence rate was lower than 10^{-6} Hz. In this framework, the main contaminants are elastically scattered protons and deuterons from the (d,p) reaction, and their energy deposition in the forward detector, less than 1.3 MeV, is way lower than that expected for the ^{12}C recoils. Six circular collimators with 1 to 3 mm diameters, placed before and after the reaction chamber, were used to ensure the reproducibility of the beam position and of the reaction vertex; the beam current was constantly monitored, both on the collimators and on the Faraday cup, at the end of the line; all of this led to a beam spot on the target with a diameter smaller than 1 mm.

The targets were constituted of thin organic layers of Melamine, $\text{C}_3\text{H}_6\text{N}_6$, with an average thickness of $\approx 50 \mu\text{g}/\text{cm}^2$, deposited on very thin layers made of $\approx 10 \mu\text{g}/\text{cm}^2$ carbon foils. The backing was oriented towards the direction of the beam, and a thin film of few Angstrom of Au was deposited on the Melamine to improve its stability under bombardment and to allow a further energy separation of different reaction products because of the different energy loss. The targets were produced in two laboratories (INFN - Laboratori Nazionali del Sud, Catania, and INFN - Laboratori Nazionali di Legnaro, Padova) and had some different characteristics, especially in the morphology of the backing: a whole Appendix is dedicated on a collateral experiment studying the

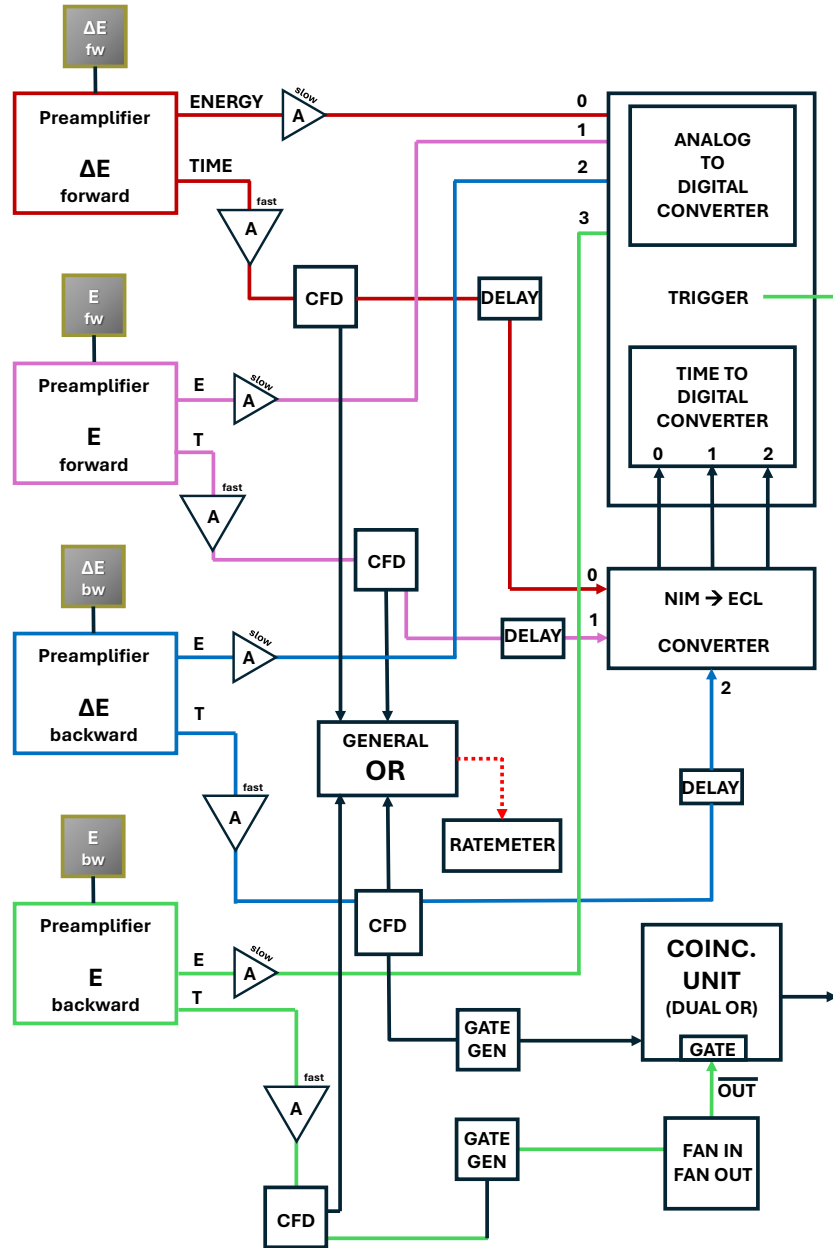


FIGURE 3.12: Scheme of the electronic chains used to connect the four detectors to the acquisition console and to trigger coincidences. Each detector was connected to its respective preamplifier, and its signal split in *time* and *energy* signals, respectively sent towards *fast* and *slow* amplifiers (indicated with triangles). The time signals are also sent to constant fraction discriminators (CFD), where they are once again split and sent to the *general OR* module and to the ADC.

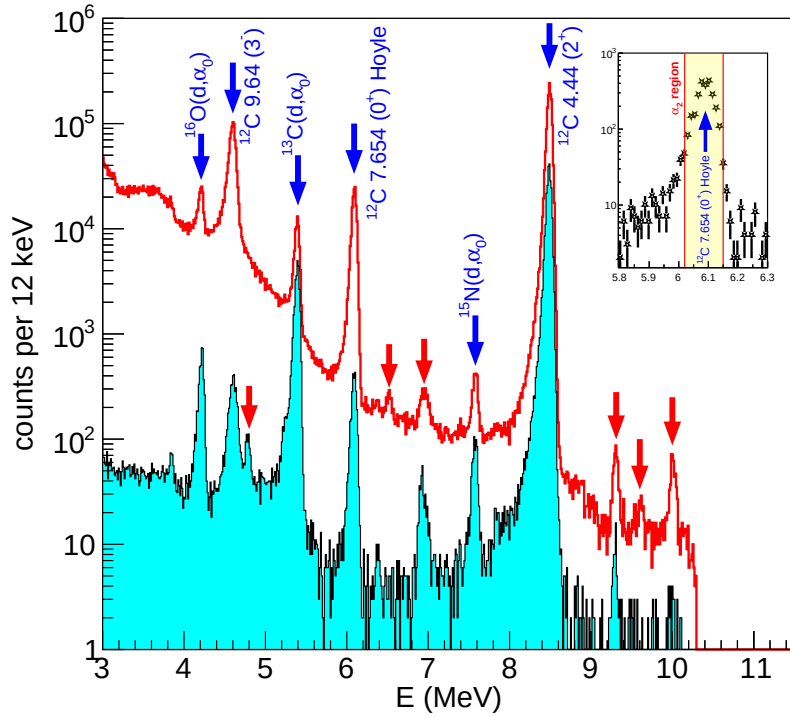


FIGURE 3.13: *Red line*: energy spectrum obtained with the backward telescope, used in anti-coincidence mode. *Blue arrows* indicate ejectile energies calculated for the expected nuclear reactions on the *main* target elements. *Red arrows* are the energies of reactions occurring on minor contaminants present in the target, such as ^7Li , ^9Be , ^{19}F . *Cyan-filled histogram*: the same as the blue spectrum, but reduced only to the events for which a coincident hit is present in the forward telescope. The background in the proximity of the Hoyle peak is strongly suppressed (by about two orders of magnitude) in the coincidence data. *Insert*: A zoomed view of the cyan-filled spectrum in proximity of the α_2 peak.

influence that these different morphologies had on the survival rate of nitrogen in the targets under the beam action.

The experimental set-up is reported in Figure 3.11: here, the backward telescope was placed at a fixed angle (centered at $\theta = 90^\circ$ with respect to the beamline), while the forward one was mounted on a movable support in the forward hemisphere, which could rotate around the target. The target was tilted at 45° with respect to the beam direction to minimize the energy loss of the ions in the Melamine layer while traveling towards the forward telescope.

The vertical position of the backward telescope was controlled with a micro-positioning device with an accuracy better than $1\ \mu\text{m}$, to maximize the detection efficiency with respect to the z -coordinate and the detection angle. The crucial point of the experiment is, in fact, the actual reconstruction of the α_2 - ^{12}C coincidence with an efficiency close to 100%; to do so, it was necessary to thoroughly calibrate the relative position of the two telescopes. Monte Carlo simulations of the radiative decay of the Hoyle state, following the $^{14}\text{N}(d, \alpha_2)^{12}\text{C}$ reaction, fully accounting for all effects due to the radiation-matter interactions and the beam optics, have been performed to estimate the required solid angle for the forward detector as a function of a given position of the backward

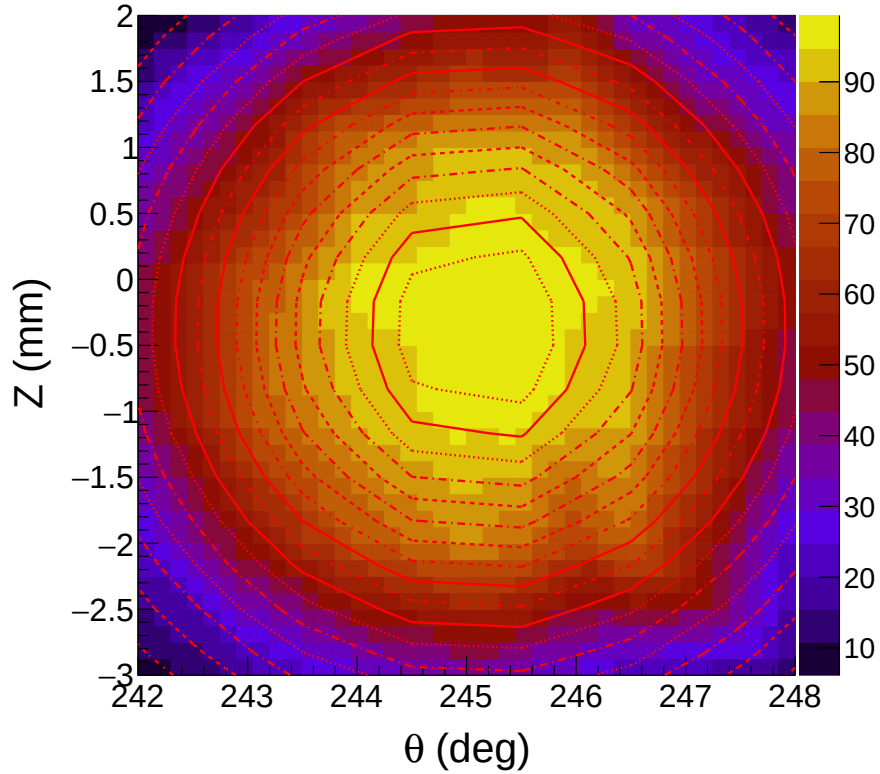


FIGURE 3.14: Bi-dimensional z - θ efficiency map for α_1 - ^{12}C coincidence events. The θ values are expressed in an arbitrary reaction chamber reference frame. Unitary coincidence efficiency is safely achieved in a broad z and θ domain. This plot is used to center the forward telescope with the $^{12}\text{C}^*$ (4.44 MeV) kinematic emission cone.

one. In a conservative fashion, the forward detectors were carefully placed in order to overabundantly cover the solid angles to detect the ^{12}C cone produced in the α_2 - ^{12}C coincidence.

The calibration of the relative angle between the two detectors was carefully done through the measurement of the coincidence rate at the two detectors based on the population of the ^{12}C first excited state, the 2^+ at 4.44 MeV, when the forward detector is positioned at $\theta = 68.3^\circ$, the expected direction of the $^{12}\text{C}^*$ recoil from binary kinematics. The corresponding α_1 particle at the backward detector is then energetically tagged by selection of the events with suitable energy, as reported in Figure 3.13 for the α_1 channel. This channel was chosen for the calibration since this excited state exclusively decay towards the ground state (with unitary branching ratio) with a radiative transition, thus the α_1 - ^{12}C coincidence rate has to be strictly equal to the α_1 detection rate in the backward telescope.

The result of this efficiency calibration is reported in Figure 3.14 in the form of a bi-dimensional efficiency map, where the horizontal axis reports the angular position of the forward detector (varied with 0.1° steps), and the vertical axis is related to the quote (z) of the backward detector, adjusted through the micro-positioning device. The map uses a color scale to better evidence the large central spot in which the detection efficiency is unitary, derived by the conservative placement of the detectors. As evident

from the Figure, the unitary efficiency spot has an opening of about 1 mm for the z variable and of about 1° for the detection angle. These values are well larger than the nominal accuracies of the z and θ placements reached in the experiment ($1\ \mu\text{m}$ and 0.05° respectively), allowing to safely assume the unitary efficiency condition during the whole experiment.

After the determination of the relative positioning of the two telescopes, the forward one has been rotated by $\Delta\theta = 3.5^\circ$ towards the beam direction to be centered with respect to the angle expected for the ^{12}C in coincidence with the α_2 particle; the error in this rotation has been computed through kinematical considerations. The upper-limit of out-of-plane deviations was instead verified through laser-scanning 3D reconstructions of the experimental chamber, and found to be $< 10^{-2}$ degrees, clearly not affecting the unitary efficiency.

Other sources of possible efficiency variations, such as small changes in the beam emittance on the target were constantly kept under control through the variations of the α_1 – ^{12}C coincidence rate measured in the place of the α_2 – ^{12}C coincidence and, furthermore, by checking the position of the (θ, z) efficiency center every 24 hours of beam time. It is thus possible to estimate the magnitude of systematic uncertainties in the overall efficiency of the present experiment to be lower than 2%; this is the first time in which such a bi-dimensional efficiency calibration is performed in a charged particle coincidence experiment, providing the keys to unambiguously probe the branching ratio of the radiative decay. Furthermore, it is worth noting that in similar associated particle experiments performed during the '70s, the unitary efficiency was never reached: for example, in the beautiful work by Mak and collaborators [185] the peak efficiency reached was of about the 80%.

3.2.3 Determination of the radiative decay branching ratio for the Hoyle state

The data analysis was restricted to the α_2 events selected in the highlighted area shown in the insert of Figure 3.13, in order to fix the region of interest for the determination of the *denominator* entering in the calculation of the branching ratio of radiative decay for the Hoyle state. After the imposition of the time-coincidence between a valid hit in the forward detector and in the backward telescope, the spectrum reported in Figure 3.13 is reduced to the coincidence spectrum indicated by the solid cyan filling, allowing to strongly exclude the unwanted background, coming from the aforementioned contaminant reactions. Therefore, for coincidence events around the α_2 energy region, it is possible to obtain the α_2 – ^{12}C coincidence rate through the analysis of the parallel energy spectrum of the forward detector, reported in Figure 3.15. In this case, the spectrum is divided in four regions:

- a low-energy continuum, given by the $\alpha_2 - \alpha$ coincidences, which is well described by Monte Carlo simulations (purple-line). This is a dominant contribution in the forward spectrum, since the main pathway for the Hoyle state decay is the α -particle emission. An important characteristic is its sharp fall around 1.8 MeV, due to a kinematic limit that can be easily calculated by energy and momentum

conservations¹. The Monte Carlo simulation here discussed takes into account both the kinematics of the in-flight decay, the geometry of the setup, the energy resolution of the detectors and the energy loss in the target.

- The $^{12}\text{C}-\alpha_2$ coincidence region of interest (yellow shadowed area), which lies in the 1.95-2.35 MeV region. In such region, where the true coincidences coming from the radiative decay of the Hoyle state are expected, a clear peak of events is seen, well reproduced by Monte Carlo simulations of the same type of the ones discussed in the previous point.
- Two small bumps of counts peaked at energies around 2.5 and 3.0 MeV, fully compatible respectively with (i) 2α particles simultaneously entering in the forward detector and coming from the ^8Be daughter of the Hoyle state, and (ii) leaky ^{12}C particles coming from the *tail* of the *plethora* of 4.44 MeV states populated in the reaction events. They are both in full agreement with their respective Monte Carlo simulations (azure and green histograms). Indeed, an accurate choice of the thickness of the target layer and of the gold deposition helps to increase the energy of these events outside of the energy region interested by the $^{12}\text{C}-\alpha_2$ coincidence events; this is allowed by the fact that low-energy heavy ^{12}C ions deposit way more energy, in matter, than the α particles, largely separating them in the energy axis.

All of these constraints on the data allowed to deduce the $^{12}\text{C}-\alpha_2$ coincidence rate by a careful energy selection. Upon the subtraction of the background (with an overall total of ≈ 2500 counts), it was possible to obtain a total of around 125000 α_2 counts tagged by the backward telescope working *in single mode*. It is thus possible to consider the systematic uncertainty on the α_2 event rate estimation negligible with respect to the one coming from the efficiency estimation. It was then possible to observe a total of 57 counts in the α_2 - ^{12}C coincidence region, with a background contribution of 4 counts, estimated by gating on equivalent-bin regions near to the α_2 peak. After the removal of the small background, a value for the radiative decay branching ratio of $\Gamma_{\text{rad}}/\Gamma_{\text{tot}} = 4.2(6) \cdot 10^{-4}$ was estimated, in very good agreement with the commonly accepted estimate of $4.19(11) \cdot 10^{-4}$ [191], while the recent estimate by [125] is more than 3σ higher than the present measurement. It is thus possible to suggest to exclude it from the weighted averages usually exploited for the determination of the radiative decay branching ratio of the Hoyle state in ^{12}C .

A summary of the literature results with the respective experimental error is reported in Figure 3.16; upon the exclusion of the outlier values [125, 190, 194], a weighted average value of $\Gamma_{\text{rad}}/\Gamma_{\text{tot}} = (4.11 \pm 0.10) \cdot 10^{-4}$ is proposed.

This result once again confirms the traditional value for the branching ratio and does not suggest any deep revision to the mechanism of the triple-alpha reaction with respect to that proposed by Caughlan and Fowler [195]. This has crucial astrophysical implications, since a modification of this process' rate was proposed as a possible explanation of the problem of the pair-instability mass gap for black holes, since the mass limits were

¹From binary kinematics, the ^{12}C in the Hoyle state would fly towards the forward detector with a velocity $v_{\text{rec}} \simeq 0.0211 c$. During its flight path, it would sequentially decay into a ^8Be nucleus and an α particle, with a Q -value of 0.287 MeV. Non-relativistic momentum and energy conservations would read $m_\alpha v_\alpha^{cm} = m_{s_{\text{Be}}} v_{s_{\text{Be}}}^{cm}$ and $Q = \frac{1}{2} m_\alpha (v_\alpha^{cm})^2 + \frac{1}{2} m_{s_{\text{Be}}} (v_{s_{\text{Be}}}^{cm})^2$. Now, assuming for simplicity that $m_{s_{\text{Be}}} \simeq 2m_\alpha$, it is possible to solve the system of the two equations to obtain $v_\alpha^{cm} \simeq 0.01 c$. When the two velocity vectors $\vec{v}_{\text{rec}}$ and $\vec{v}_\alpha^{cm}$ are collinear, the resulting velocity is maximal and would correspond to an α particle of kinetic energy of $\simeq 1.82$ MeV.

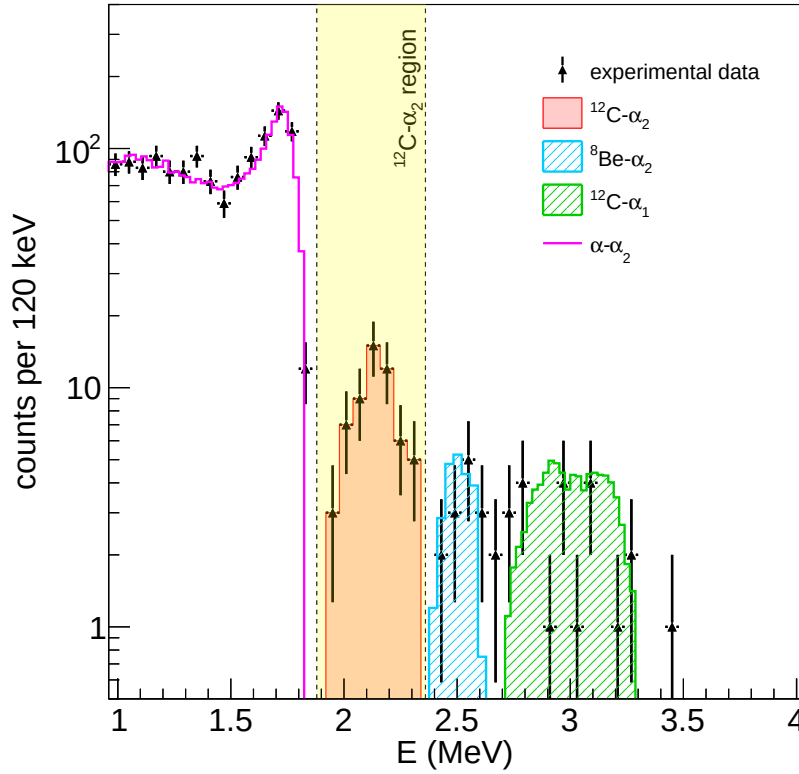


FIGURE 3.15: Forward energy spectrum in time-coincidence with the narrow energy region around the α_2 peak shown in the region of interest evidenced in the insert of Figure 3.13. The yellow area indicates the α_2 - ^{12}C region of interest where the genuine coincidence events are expected. The purple line is a Monte Carlo simulation of the expected contribution of α_2 - α coincidences. The azure and green lines represent, respectively, Monte Carlo simulations of the α_2 - ^8Be and α_1 - ^{12}C contributions.

indeed challenged by recent LIGO-Virgo observations made on intermediate mass black holes [196, 197]. On the basis of this, solutions to this puzzling problem should be therefore explored outside the nuclear structure domain. Accounting for the convection mechanisms in stars [198, 199], the clarification of the triple-alpha process reaction rate is extremely useful to pose stricter constraints on the $^{12}\text{C}(\alpha, \gamma)^{16}\text{O}$ reaction rate; when combined, the properties of these two fundamental reactions are crucial to define and explain the ratio of carbon and oxygen abundance in the Universe [135].

From the nuclear structure point of view, by using the known total width of the Hoyle state ($\Gamma = 9.3 \pm 0.9$ eV) and the pair emission width reported in the literature ($\Gamma_\pi = 6.2 \pm 0.2 \cdot 10^{-5}$ eV) [133] it is possible to estimate the Γ_{E2} value for the Hoyle state coming from the present average of the branching ratios: $\Gamma_{E2} = 3.76 \pm 0.38$ meV. This value is in very good agreement with literature estimates and can be compared with predictions coming from different theoretical models aiming to describe the nuclear structure of ^{12}C . An example of various model predictions is shown in the following Table, obtained by combining data coming from Refs. [79, 121, 134] and, where necessary, converted from $B(E2) \uparrow$ to $B(E2) \downarrow$ (see the discussion reported in Section 2.2).

It is evident that the improved experimental value calls for more refined predictions

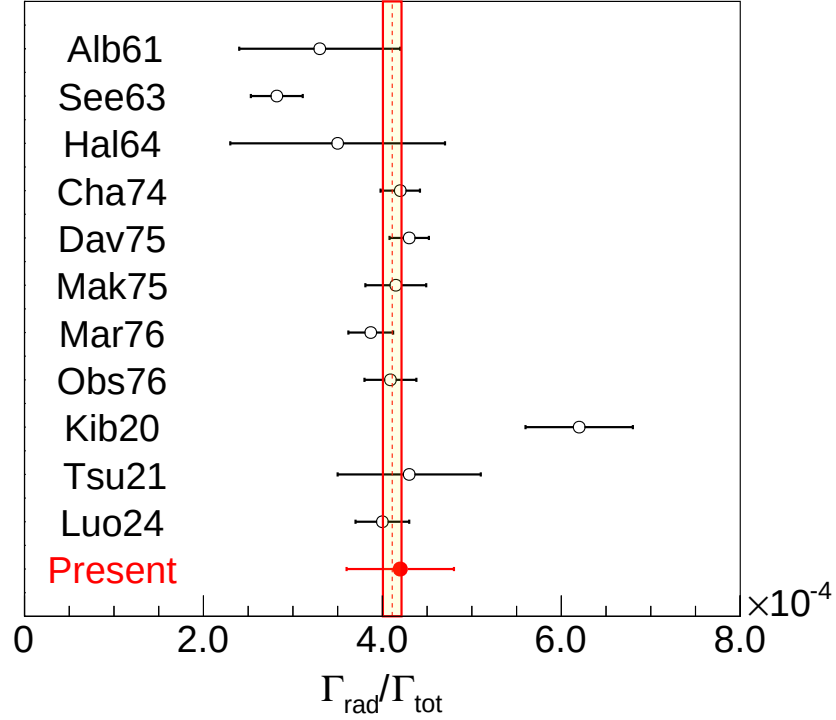


FIGURE 3.16: Radiative decay partial width $\left(\frac{\Gamma_{\text{rad}}}{\Gamma_{\text{tot}}}\right)$ of the ^{12}C Hoyle state as obtained from charged-particle coincidence experiments (Refs. [183–185, 188, 189, 200–202]) and γ -particle coincidence experiments (Refs. [125, 186, 203]), all reported in black, while the result from the present experiment is reported in red. The yellow central band indicates the new confidence interval, obtained through a weighted average of all available estimates, excluding the outlier values (Refs. [125, 202]).

Exp. $B(E2)$ ($\text{e}^2 \text{ fm}^4$)	NLEFT	FMD	GCM	MCSM
13.5 ± 1.4	12 ± 4	19	3.5	8.5

TABLE 3.2: Reduced transition strengths $B(E2)$ for the $0_2^+ \rightarrow 2_1^+$ electromagnetic transition leading to a gamma ray of 3.215 MeV. The present experimental data, coming from the global average of radiative branching ratios discussed in the text, is compared to predictions of *ab-initio* nuclear lattice effective field theory (NLEFT) [134], Fermionic Molecular Dynamics [120], Generator Coordinate Method (GCM) [79] and MonteCarlo Shell Model (MCSM) [121].

from theoretical models (a not easy work, considering the difficulties in determining the wavefunction of the Hoyle state because of the possible coexistence of shell and cluster effects, see e.g. the discussions in Ref. [121]). In any case, it is interesting to note that both NLEFT and MCSM predictions, that are closer to the experimental value, describes the structure of the Hoyle state as having a dominant triple α structure arranged to form an obtuse triangle. It is almost certain that in the next years further theoretical efforts will be reported in describing the now fully ascertained value of the reduced transition strength of the Hoyle state².

3.3 Some concluding remarks and perspectives

Despite the efforts performed by several experimental and theoretical groups in studying the structure of the ^{12}C in the light of α cluster formation, several aspects are still mysterious and need to be verified in the next future. Let's indicate some of these points as a possible *vademecum* for further investigations.

- The structure of the ^{12}C ground state still needs to be understood. Two different visions, interpreting this nucleus as a weakly deformed spheroid or as a clustered triangular α structure, can describe some part of the data coming from spectroscopic information and/or from the analysis of nuclear reactions. In this framework, new experiments with hadron or electromagnetic probes can shed further light on this aspect.
- From the experimental point of view, a strong improvement of several aspects of the spectroscopy of the Hoyle state have been reported in the last decade. Such a beautiful mess of data should now be subject to an intense campaign of comparisons with many theoretical model predictions, with the aim of unveiling the details of the cluster nature of this state. As a final consideration, at the state of the art, it is indeed a pity that the total width of the Hoyle state is known with a relative error of about 10%.
- In recent times, some suggestions on the possible presence of an Efimov state close to the Hoyle state (at about 7.458 MeV) have been discussed both theoretically and experimentally, but conclusive proofs on its existence are still missing. Given its importance both for fundamental quantum mechanical aspects and because of a potentially large astrophysical impact, new experiments on this aspect could be very impacting.
- Finally, the radiative widths of highly-lying excited states in ^{12}C , above the Hoyle state, can have an astrophysical influence in contexts where the triple α process occurs at large temperatures (above 2 GK). For example, recent measurements with the associated particle method (see Section 2.2.1) found a very small, but non-vanishing radiative branching ratio of $1.3 \pm 1.2 \cdot 10^{-6}$ for the 9.64 MeV 3^- state. New experiments could refine the very large uncertainties still affecting this quantity, even if this means to fight against huge backgrounds compared with the very rare expected decay events.

²Note added in proof: a very recent work [204] re-analyzed the data of Ref. [125] finding some inconsistencies; once removed, a value in agreement with the present value and the whole literature average is obtained.

Chapter 4

Cluster states of ^{16}O at high excitation energies

Immediately after ^{12}C , the next self-conjugate nucleus is ^{16}O ; as reported in the previous Chapter, the two nuclei are strongly tied to each other, not only for the fact that they compose organic systems and a vast gamma of life-building molecules, but also for the maintenance of the abundance equilibria in the interstellar matter, regulating crucial phenomena and cycles of stellar nucleosynthesis and black holes generation. Several covalence effects are observed in neutron-rich oxygen isotopes [205–207].

Recently, several theoretical works attempted to study the ^{16}O nucleus structure, hoping to find some connections or evidences of clustering effects, in its ground and excited states. Similarly to the case of the ^{12}C , a molecular-like structure with tetrahedral symmetry, made up of a pronounced 4α cluster configuration, was reported in [208] after Hartree-Fock calculations. Evidences of such symmetry, on which some ACM calculations have been based [96], have been found in the ground state band properties, while they are more difficult to find for other excited bands related to other vibration modes.

Being more complex than the preceding self-conjugate nuclei, the ^{16}O nucleus has more available cluster configurations, going from the simple $\alpha + ^{12}\text{C}$ decomposition to complicated tetrahedral or condensate structures, recently investigated inside a "container model" of the nucleus [209, 210]; the structure of low-lying states was instead investigated through the Skyrme model [211], finding a level scheme which roughly agrees with experimental data, pointing towards a tetrahedral configuration for ^{16}O *ground state*. Several models, such as the ACM-VAP (*Variations After Projection*) were exploited to predict the possible formation of linear chain states in ^{16}O states excited at high energies; states with this structure would belong to a rotational band whose 0^+ band-head lies at $E_x = 33$ MeV, and with the last state being a $J^\pi = 6^+$ state.

In the recent times, one of the hottest experimental fields was that of the search of a Hoyle-analogue state in the ^{16}O nucleus spectroscopy, i.e., a state which could exhibit properties similar to those of the 7.654 MeV 0^+ state in ^{12}C . According to the previsions given by Ikeda and his diagram, such a state should be located close to the 4α disintegration threshold, at 14.44 MeV of excitation energy; this is the region in which the largest part of the experimental studies were focused. One of the most probed candidates is a

0^+ state at ≈ 15.1 MeV, where several experiments [212–214] tried to draw information about its possible 4α structure, but without reaching any firm conclusions.

An "unconventional" (and very recent) way to search for clusterization effects in ^{16}O is the analysis of collective flow parameters deriving from heavy-ion collisions performed in the ultra-relativistic regime [215, 216], since it appears that these may be influenced by the occurrence of a tetrahedral α -cluster structure rather than an ordinary soft sphere. *Mutatis mutandis*, a probe coming from the study of nuclear reactions at intermediate energies will be discussed in the next Section, through the application of the crystallographic approach (Optical model + coherence phenomena), in the same fashion as for the ^{12}C case. Section 4.2 will instead discuss, on the light of the possible onset of α clustering effects at high excitation energies, the spectroscopy of highly-lying states in ^{16}O decaying into an α particle and leading to the emission of a ^{12}C residual in the ground state or in the Hoyle state.

4.1 A crystallographic approach to study α -clustering phenomena in ^{16}O ground state

The aim of this Section is to explore the possible presence of cluster structures in the *ground state* of the ^{16}O nucleus, on the basis of the same prescriptions and approximations presented in the ^{12}C case, through the presence of *coherent effects* in elastic scattering collisions, due to the diffraction on the α clusters. In this case, the oxygen nucleus is supposed to oscillate between the usual spherical structure of 16 mixed nucleons, described by the shell model, and a rather minor tetrahedral configuration made of 4 α clusters. The elastic scattering on such different structures would influence the behaviors of the elastic scattering angular distributions of protons impinging on the ^{16}O nucleus as a function of the two different configurations.

4.1.1 Details on the adopted model

As for the ^{12}C case (see Section 3.1.1), it is necessary to study, preliminarily, the literature data of $p + ^4\text{He}$ collisions to describe the effect of the scattering of protons on the single α cluster, (i.e., the ^4He nucleus) in order to determine the scattering amplitude $f(\theta)$ entering into the Wierl formula (Eq. 3.1) to describe the angular distributions of diffractive elastic scattering of matter waves on molecular-like targets. This procedure was performed in the same way as described in Section 3.1.1.

Then, to describe the elastic scattering of protons on the dominant spherical structure in a broad energy range of literature data, between 10 and 100 MeV of proton beam energy, several Optical Model simulations, based on three commonly adopted parameterizations, have been conducted. Differently from the ^{12}C case (see Section 3.1), in the present one, the more refined Coupled Channel calculations have not been used to describe the spherical elastic scattering contributions because of the limited availability, in the databases, of detailed data on $p+^{16}\text{O}$ elastic and inelastic scattering data [162]. These experimental limitations can be also due to the difficult manufacturing of oxygen-containing targets, leading to great difficulties in the planning of low-background and high resolution measurements.

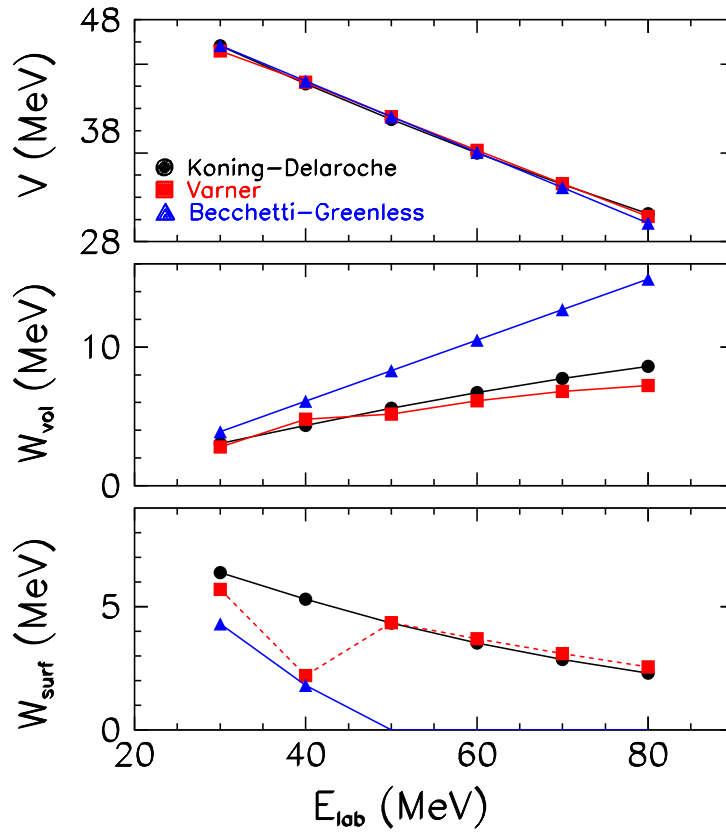


FIGURE 4.1: Evolution of the calculated depths of the real potential (V , *top panel*), the imaginary volume potential (W_{vol} , *central panel*) and the imaginary surface potential (W_{surf} , *bottom panel*) as a function of the bombarding energy in the $E_p = 30 - 80$ MeV region, on the basis of the three Optical Model global systematics here adopted: [217] (black dots), [175] (red squares) and [160] (blue triangles).

As already sketched in Sections 1.2.2.3 and 3.1.1, the Optical Model allows to treat the scattering and absorption phenomena of a matter wave on a target nucleus through the use of an optical potential constituted by a real term (V_0), accounting for the *scattered* part of the wave, and an imaginary term (iW) related to the wave portion absorbed by the target nucleus. The complete form of the optical potential is thus:

$$V = -(V_0 + iW) \quad (4.1)$$

where V_0 and W are real and positive functions of the distance from the center (the so called *form factors*) of the nucleus. Similarly to the ^{12}C case, the V_0 term has been treated by using a Wood-Saxon shape for the potential well. On the other hand, for the imaginary part of the potential, based on a phenomenological analysis of several elastic scattering data [115], different shapes have been taken into consideration other than the Wood-Saxon one, including gaussian shapes located near the surface, to consider the likely absorption of particles already when they approach the nuclear surface, due to the strength of the nuclear force. This potential is then added of the Coulomb potential term and, if the spin of the two reagents is not zero, of an additional term related to the spin-orbit coupling in nuclear forces.

In the specific case of the $p + ^{16}\text{O}$ elastic scattering treated in this Section, three different optical potential prescriptions have been historically reported in the literature for collisions at intermediate energies ($E_p \simeq 20 - 100$ MeV). These Optical Model parameters have been obtained through fits on wide databases of experimental data at various energies and on different targets, and will hereon be referred (from the names of their compilers) to *Koning-Delaroche* [217], *Varner* [175] and *Becchetti - Greenlees* [160]. All these parameterizations include both a volume (Saxon-Woods like) and a surface (gaussian-like) shape for the imaginary part. The trend of the depths of the real (volume) potential and of both the volume and surface parts of the imaginary potential are described, in Figure 4.1, with different colors and symbols for the three parameterizations, as a function of the bombarding energy. The Optical Model parameters are quite similar in absolute values and show similar energy trends in the three systematics here adopted; it is interesting to note that the surface part of the imaginary term in the Becchetti-Greenless systematics becomes zero at energies above $\simeq 50$ MeV; consequently, a large volume imaginary term is needed at high energies to reproduce the global datasets of elastic scattering and nuclear reactions [160].

Given the very limited range of available data, a further Optical Model fit, trying to improve the description of the experimental data by starting with the global Optical model potential, was avoided. At variance, it was assumed that the global Optical Model parameters here adopted from systematics would already represent an optimized condition in describing the spherical contribution, and therefore an improvement in the description of data can come by a different reaction scenario, i.e. the one involving the collisions on a cluster structure.

Even if this approximated procedure can give only a qualitative hint on the possible presence of transient tetrahedral configurations, it can still be useful as a guideline for further works both on the experimental and theoretical sides. On the same basis of the ^{12}C case (Chapter 3), the analysis of angular distributions with the sum of spherical and coherent terms can give an idea on the possible contribution coming from the scattering on an α clustered structure.

4.1.2 Application to the case of the ^{16}O nucleus

Given the similar dimensions of the ^{16}O and the ^{12}C nuclei, the energy range to get a proton wave of the suitable wavelength is similar, i.e., 30 - 100 MeV; the cross section data for the $p + ^{16}\text{O}$ elastic scattering have been taken from literature databases [164, 168, 218–220] at the closest energies available with respect to the $p + ^4\text{He}$ fits shown in Figure 3.4; in this way, the effect of the interpolation between the various intermediate energies can be reduced at a minimum. However, these datasets do not allow a direct and deep comparison with the performed simulations, since they are too sparse in angle, often limited to the forward hemisphere only, and affected by too high uncertainties on the determination of the absolute cross section (even higher than 10% at some points). For this reasons, these will be adopted to draw *qualitative* conclusions on the present analysis.

The scattering amplitude of the single α -cluster and the coherent contributions given by the regular distribution of the α particles have been computed in a way very similar to the ^{12}C case. In particular, by indicating with $f(\theta)$ the $p+\alpha$ scattering amplitude, the specific Wierl formula derived from Equation equation 3.1 for the scattering of a proton

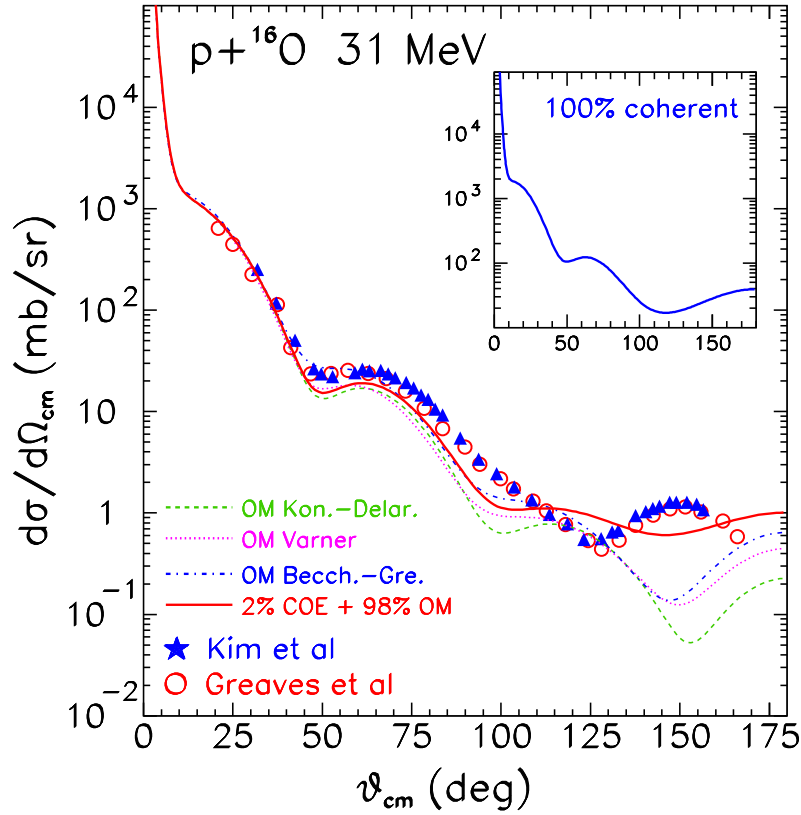


FIGURE 4.2: Angular distribution simulations at $E_p = 31$ MeV based on three different optical model parameterizations (dashed lines, Refs. [160, 175, 217]), and on a combination of the optical spherical contribution and the coherent one (red solid line). Superimposed to the curves are experimental data from Refs. [164, 220]. The insert shows the same angular distribution simulation obtained after a parameterization assuming 100% of the contribution coming from the coherent clustering term.

wave on a tetrahedral ^{16}O configuration would become:

$$\frac{d\sigma}{d\Omega_{clu}}(\theta) = 4|f(\theta)|^2 \cdot \left(1 + 3\frac{\sin \mu r_{ij}}{\mu r_{ij}}\right) \quad (4.2)$$

Also in this case, r_{ij} represents the expected distance between two single α -clusters in the ^{16}O nucleus, i.e., the edge of a tetrahedron inscribed inside a sphere, calculated through Euclidean geometry: $r_{ij} = \frac{2\sqrt{6}}{3}R$, with $R = r_0\sqrt{16}$ and $r_0 = 1.25$ fm. The required λ for the proton wave diffraction would thus be of around 4 – 6 fm, requiring the study of $p + ^{16}\text{O}$ elastic scattering experiments in the E_p range of 30 – 80 MeV. The interplay of the spherical (dominant) and the coherent (minor) contributions, without the inclusion of interference effects, is then calculated in the same way as for the ^{12}C case (Eq. 3.2), to account for different weights of the two contributions.

The main body of Figure 4.2 reports the angular distribution simulations of $p + ^{16}\text{O}$ elastic scattering in the spherical configuration, as obtained by the three different optical potentials parameterization here used, with different colors and dashing (Refs. [160, 175, 217], as shortened in the labels of the picture). The literature experimental data are superimposed to the simulations; they start to drift from the simulated trends (for a

100% spherical configuration) at $\theta \simeq 40^\circ$. Although these discrepancies may be reduced by adopting different optical parameters prescriptions, one should not exclude that they may be hinting for the presence of some clustering phenomena.

The simulation based on a 100% coherent contribution, shown in the insert of Figure 4.2, has in fact a completely different shape with respect to the spherical calculations, especially at larger angles, and seems to visually agree in a better way with the experimental data at medium-to-large angles. If one qualitatively combines the two contribution with a 98% contribution of spherical scattering and a 2% contribution of the coherent term coming from the α tetrahedral structure, it would be possible to obtain the red solid line shown in the main panel of Figure 4.2, giving a bit better agreement with experimental data. Similar findings are also seen for other data at energies different than 31 MeV, and this would support further efforts to investigate in a more quantitative way the possible presence of coherent effects in elastic scattering data, possibly profiting also of almost-monochromatic neutron beams at intermediate energies (see the discussion in Section 3.1.1). Of course, to perform this type of analysis on experimental data, it is necessary that some fundamental requirements (about angular steps and measurement uncertainties) are met; the uncertainties coming from the thickness measurements of an oxygen-containing target and the possible degradation of the oxygen content in the target under bombardment would transform this type of experiments in an out-and-out challenge.

4.2 The HELICA experiment to study highly excited states in ^{16}O

The spectroscopy of ^{16}O is particularly important, as this nucleus is both doubly magic and self-conjugate. Although the situation of cluster studies in neighboring nuclei (like ^{12}C and ^{20}Ne) appears to be more clear, it is not easy to say the same for the ^{16}O nucleus. This case is more puzzling: tetrahedral configurations (or even less usual ones like squares or kites [221]) have been suggested, in the framework of different theoretical predictions, for both the ground and low energy states of ^{16}O [211]; however, in some energy regions (like the 13 - 15 MeV one, near the 4α disintegration threshold), experimental results are contrasting [222], and a general consensus on the cluster properties of the states therein is still far.

One of the most unclear situations unfolds at energies higher than 19 MeV, where several high-spin states have been predicted, from a theoretical point of view, but with just limited correspondences with experimental data [72, 211]. An even more puzzling question is related to the occurrence (and the nature) of the low-spin states in the same energy region, given that the spectroscopic information available are only fragmentary [222]. As predicted by Arima, Gillet and Ginocchio in their *quartet model* (see Section 1.2.3.5), several strongly α -clustered 0^+ states should lie in this energy region (as sketched in the right panel of Figure 4.3, but no conclusive experimental evidences have been reported to date [192]. In this model, the clustered 0^+ states in self-conjugate nuclei are sequentially built by considering the promotion of α -like particles from one orbital to another one of an harmonic oscillator potential well, as discussed in details in Section 1.2.3.5. To complicate matters, other competing structures have been proposed in this energy region, related to the ^{16}O giant dipole resonance [223], along with a possible $J = 2$ isoscalar state near to the deuteron - nitrogen disintegration threshold, with a

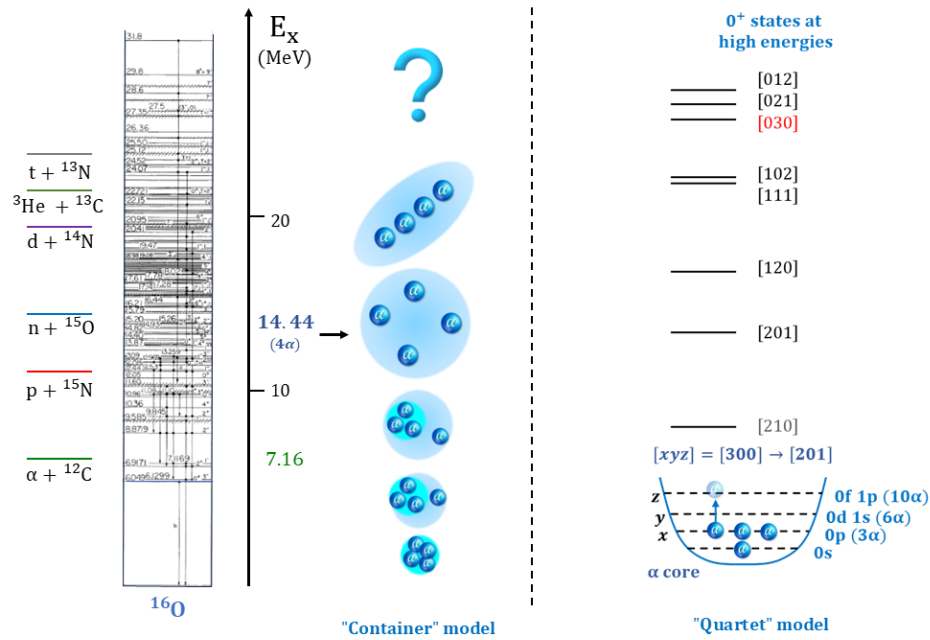


FIGURE 4.3: Pictorial representations of the possible evolution of the structure of the ^{16}O nucleus on the basis of the *container* model [210] (middle panel) and the *quartet* model [73] (right panel) predictions. On the left, the sequence of known levels and the position of several disintegration thresholds is reported (data taken from Ref. [222]).

pronounced $^{12}\text{C}+\text{d}+\text{d}$ structure [224]. Thorough spectroscopic investigations could help to find an answer to all these long-standing open questions.

Taking a look to the full nuclear structure framework, the ^{16}O case is in general particularly intriguing, since it has a lower level density than its neighboring even-even nuclei. Further, as usual for self-conjugate nuclei, the first emission threshold is not that of a single particle (proton or neutron, $\text{p} + ^{15}\text{N}$ or $\text{n} + ^{15}\text{O}$, both higher than 10 MeV); instead, the $\alpha + ^{12}\text{C}$ disintegration already happens at $E_x = 7.16$ MeV, hinting for the occurrence of clustering effects at nearby energies (and the presence of the 6.05 MeV second 0^+ state is a beautiful demonstration of such aspect, as discussed in Section 1.2.3.4). Recently, a microscopic model based on the so called *container* philosophy [210] suggested the occurrence of a continuous evolution of the structure of ^{16}O , starting from a compact structure for its ground state, towards the partial development of clustered configurations of the type $\alpha + ^{12}\text{C}$ near the α -threshold, the onset of gas-like scenarios at energies equal or higher than the 4α threshold at 14.44 MeV, and finally arriving to suggest the potential existence of a linear chain structure at about 20 MeV, as pictorially sketched in the middle panel of Figure 4.3. Unfortunately, only few theoretical predictions have been made about the possible existence of other 0^+ cluster states at still higher excitation energies. Following on Ichikawa predictions [225], it would look like that, on the basis of the Skyrme-cranked Hartree-Fock model, an extremely deformed linear-chain 0^+ state could exist at very high excitation energies of around 28-35 MeV.

From the experimental point of view, some groups [226, 227] performed particularly challenging measurements, by investigating 4α coincidence events produced from the decay of ^{16}O nuclei highly excited through nuclear reactions of different nature. Multi-particle correlations techniques were then applied to disentangle the different decay

channels involving the emission of α particles. One of these experiments was conducted by M. Freer and collaborators in 1995, where, through detailed correlation studies, it has been possible to identify various peaks related to different unbound states exit channels ($^8\text{Be} + ^8\text{Be}$, $\alpha + ^{12}\text{C}$, $\alpha_3 + ^{12}\text{C}_{9.63}$), but only reaching up to a maximum excitation energy of ≈ 22 MeV in ^{16}O . Another more recent attempt, performed by Charity and collaborators in 2019, reached up to about 27 MeV excitation energy, but demonstrated the difficulties in evidencing the occurrence of clear structures correlated to the $\alpha_2 + ^{12}\text{C}_{\text{Hoyle}}$ decay channel, which would represent a *smoking gun* to identify the occurrence of highly clustered states in the ^{16}O spectroscopy.

In this context, the aim of the HELICA experiment here discussed was the study of the occurrence of *low-spin* and *high-energy* states of the ^{16}O nucleus. To impose the right constraints for the selective study of these specific states, it can be useful to choose a reaction with high *separation energy* (S_x) and to perform it with very low center of mass energies: in this way, restrictions on the angular momentum of the entrance channel (related to the beam energy) can help to selectively populate low spin states. Reactions that fulfill these requirements are the $d + ^{14}\text{N}$ or the $^3\text{He} + ^{13}\text{C}$; specifically, the latter was chosen since, at low energies, it essentially proceeds by the formation of a very excited ^{16}O compound nucleus, avoiding the disturbing presence of direct effects that may affect the deuteron-induced reactions even at sub-barrier energies [228]. The $^3\text{He} + ^{13}\text{C}$ reaction, even at very low energy, allows to study the cross section of the *golden channel* leaving the residual ^{12}C nucleus in the Hoyle state. This would represent a direct, albeit not simple, way to access these four-body decay channel with a measurement *in single mode*.

Up to date, only few data are available for the $^3\text{He} + ^{13}\text{C}$ reaction at energies lower than 3 MeV, due to experimental difficulties especially related to (i) the necessity of discrimination of the emitted α particles; (ii) the high value of the Coulomb barrier ($U_c \approx 2.8$ MeV), strongly suppressing the cross section of under-barrier collisions. Holmgren et al. [229, 230] report few differential cross sections but only for the $\alpha_{0,1}$ channels (i.e. the ones leaving the ^{12}C residual nucleus in the ground state and in the 4.44 MeV state). On the other hand, Weller [231] investigated also the α_2 channel (with the ^{12}C nucleus left in the Hoyle state) but at energies of 5 to 8 MeV, well higher than the Coulomb barrier. One of the very few works reporting under barrier data for the α_2 (Hoyle) channel is discussed in Ref. [232], which however shows all the limitations encountered in the measurement of the α_2 particles with a single-stage detector. In fact, the protons coming from the nearby $^{13}\text{C}(^3\text{He}, p_{1,2})^{15}\text{N}$ reactions at forward angles produce peaks around 100 times larger than the neighboring α_2 peak. The paper, indeed, reports only two complete angular distributions for the $\alpha_{0,1,2}$ channels at energies lower than 2 MeV, showing some asymmetries with respect to 90° that are typical of interference effect between overlapping states with different parities (see the discussions in Section 2.3.2).

All the considerations here listed, along with the limited number of studies on low J states above 20 MeV, and considering the predictions of the quartet model [73] on the existence of some 0^+ states in the region between 23 and 26 MeV, motivated the design and the realization of the HELICA experiment, which will be discussed in the next Sections.

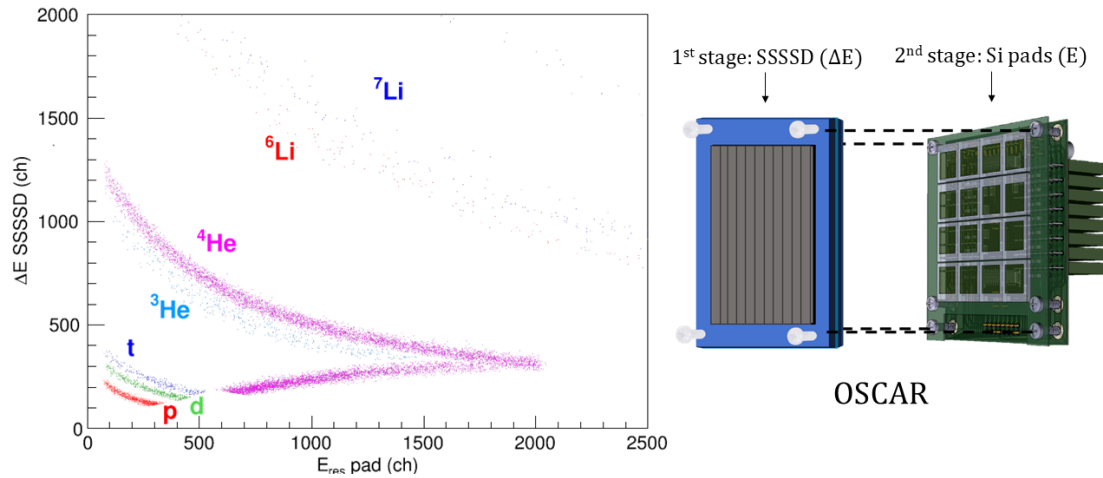


FIGURE 4.4: *Left*: $\Delta E - E$ identification matrix obtained for a pseudo-telescope of the OSCAR detector, as obtained from the commissioning experiment [234]. It is possible to see the clear separation of the various isotopes of elements with $Z = 1, 2$ and 3 . *Right*: scheme of the two stages of the OSCAR detector. See Ref. [234] for further details.

4.2.1 The experimental setup and the OSCAR detector

The experiment was performed at INFN - Laboratori Nazionali di Legnaro with $^3\text{He}^+$ beams delivered by the AN2000 accelerator, with energies between 1.4 and 2.2 MeV. The energy steps were of 20 keV, and the beam had a resolution better than 0.5 keV, a spot size of around 2 mm and a current ranging from $\simeq 70$ to 190 nA. The charge status of the beam when passing the target was studied in details through dedicated runs with and without the target; the findings were in excellent agreement with the tables reported in Ref. [233]. The target used was made by a self-supporting foil enriched in ^{13}C at a 99% level, with a thickness of $29 \pm 1 \mu\text{g}/\text{cm}^2$ determined by dedicated energy loss measurements of α particles emitted by a ^{241}Am source. Dead-time corrections were performed by injecting signals coming from a calibrated pulser with a fixed rate into the front-end electronics and the acquisition system.

The detection apparatus was composed by four OSCAR modules, arranged in the so-called HELICA configuration, as sketched in Figure 4.5: a very compact array, in which the detector hodoscopes, the target, the cooling system (with liquid circulation) and the 96 electronic channels are enclosed in a vacuum chamber of approximately 30 cm diameter. Despite the high density of materials in the chamber, the vacuum level registered during the experiment was of the order of 10^{-6} mbar. The system has a high granularity (with a $\Delta\theta \simeq 2^\circ$) given by the 128 $\Delta E - E$ pseudo-telescopes, and grants full coverage of polar angles; the telescopes have two stages of 20 and 500 μm thickness, which allow to obtain very low identification thresholds, of $\simeq 1.4$ AMeV (fundamental to discriminate the slow α_2 particles from other ejectiles) and simultaneously an excellent energetic resolution of 50 keV at 5.5 MeV of *identified* deuterons [95]. For helium isotopes, in the present experimental conditions, it was possible to reach an average identification threshold of around 6 MeV and an energy resolution of 60 keV.

Going a bit more in details, OSCAR (hOdoscope of Silicons for Correlations and Analysis of Reactions) is a modular detector composed by two independent stages, and its scheme

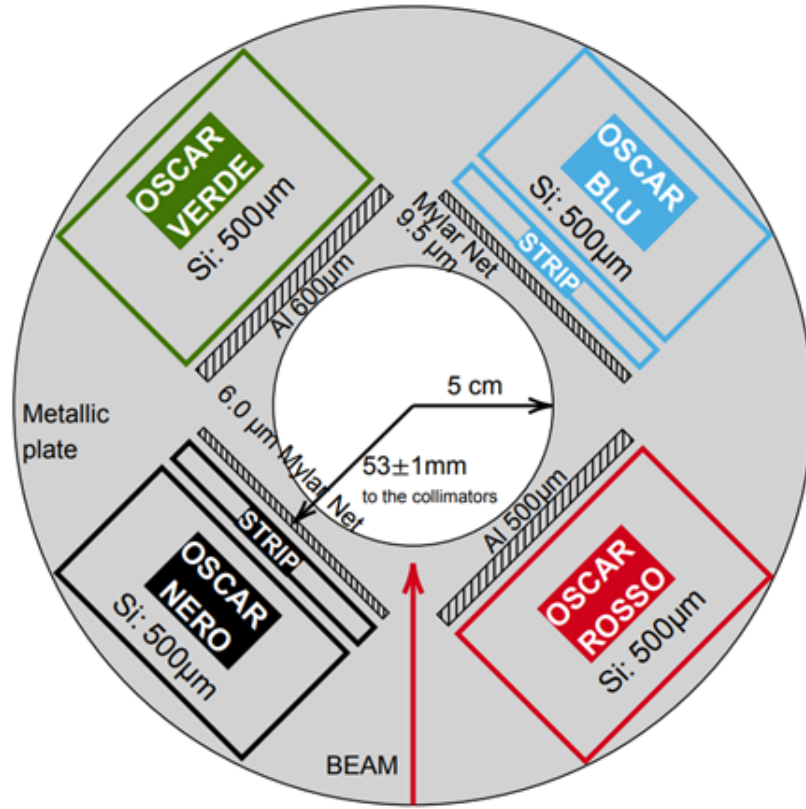


FIGURE 4.5: A schematic representation of the HELICA detection array composed by the four OSCAR units. The presence of layers of absorbers to stop selected categories of ejectiles has been evidenced with dashed rectangles.

is reported in the right-hand side of Figure 4.4. The first one is a Single Sided Silicon Strip Detector (SSSSD), made by 16 ultra-thin strips with a thickness of $20\ \mu\text{m}$; it is responsible for the low detection threshold ($\approx 0.5\ \text{AMeV}$), allowing to identify the slowest particles. The second stage is composed of 16 single ceramic-framed silicon pads detectors, with an active area of $1\ \text{cm}^2$ each and thickness varying, depending on the specific application, from 300 to $500\ \mu\text{m}$. From the overlap between the two stages of a single OSCAR module, it is possible to obtain $64\ \Delta E - E$ pseudo-telescopes, allowing a high angular resolution of 1° at 55° . However, one of the main perks of OSCAR is its high modularity, and the possibility to combine different units or stages to form custom set-up or to couple with other complex detection systems (as it was made in the past with the Advanced Gamma Tracking Array (AGATA) and the MAGNEX magnetic spectrometer).

The HELICA configuration, reported in Figure 4.5, is thus made of four OSCAR modules mounted one for each side of a square surrounding the target, centered at $\pm 45^\circ$, $\pm 135^\circ$ with respect to the beam direction. Of the four modules, two of them (OSCAR BLU, at $+45^\circ$, and OSCAR NERO, at -135°) were preceded by a thin Mylar foil and fully equipped with both the SSSSD and the pad stages, while the other two (OSCAR ROSSO, at $+135^\circ$, and OSCAR VERDE, at -45°), were mounted using only their second pad stage, with $500 - 600\ \mu\text{m}$ thick aluminum foils covering them, while the very thick aluminum foils were used to stop all the reaction ejectiles with the exception of high energy protons (well above $10\ \text{MeV}$ in the whole angular domain) emitted in the

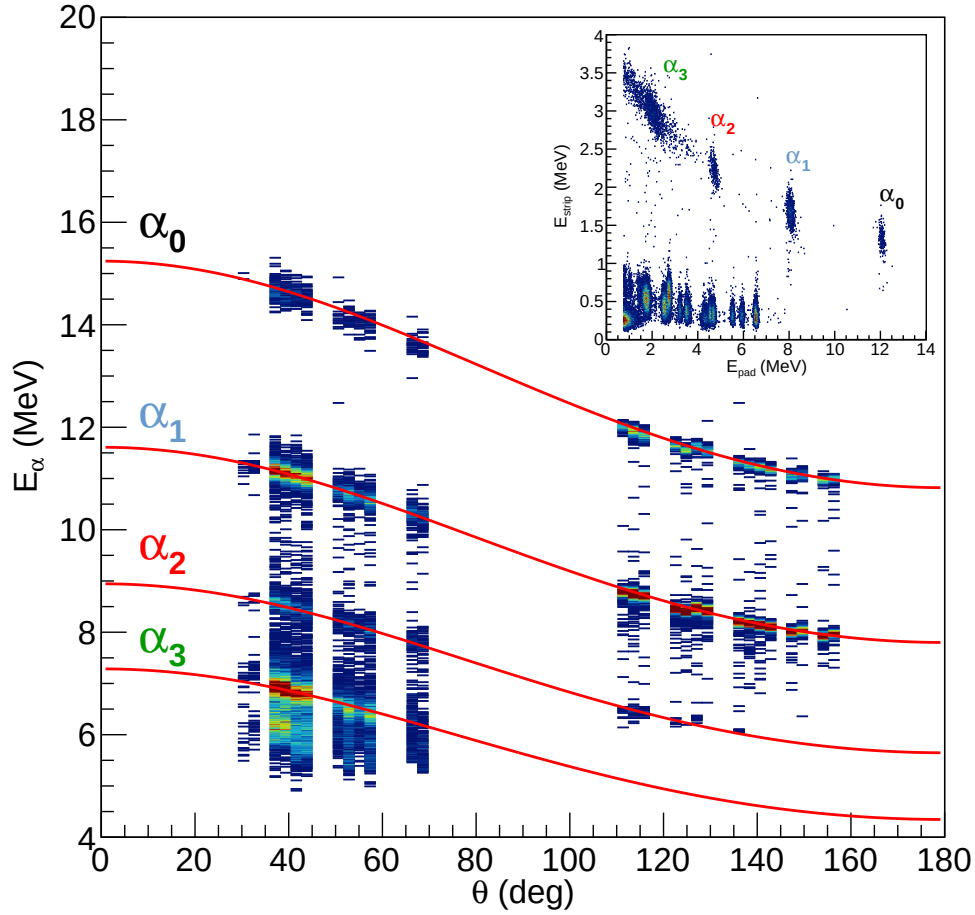


FIGURE 4.6: Kinematic loci of the identified families of α particles emitted in the $^3\text{He} + ^{13}\text{C}$ reactions studied at 2.02 MeV of bombarding energy. The red lines indicate the predicted kinematic loci for the α_i reactions populating the 0, 4.44, 7.65 (Hoyle) and 9.63 MeV states of the ^{12}C residual nucleus. The insert shows a typical $\Delta E - E$ identification matrix obtained from a given strip-pad combination of the hodoscope, in this case at $\theta_{lab} \approx 40^\circ$ and $E_{lab} \approx 1.52$ MeV.

$^3\text{He}(^{13}\text{C}, p_0)^{15}\text{N}$. Data coming from this reaction and analyzed through the OSCAR VERDE and OSCAR ROSSO detectors will be the subject of a specific analysis that will be performed in the future.

For the purpose of the present experiment, only data coming from the OSCAR BLU and NERO detectors were considered. The BLU (forward) detector covered angles in the range $\theta_{lab} \simeq 24 - 68^\circ$, while the NERO one (backward), centered at 135° , covered the $\theta_{lab} \simeq 111 - 156^\circ$. The Mylar foils preceding the detectors were used to stop the high flux of elastically scattered particles. The detection solid angles have been estimated through careful Monte-Carlo simulations, bench-marked with Rutherford scattering runs with protons and ^3He beams on Au targets. Accurate detector energy calibrations were performed at different energies and without stopper foils, with a mixed-nuclide α source and several proton elastic scattering points on Au and ^{12}C targets.

An example of a $\Delta E - E$ identification matrix obtained for a typical OSCAR pseudo-telescope at forward angles is reported in the insert of Figure 4.6. Several bumps associated to the proton and deuteron channels (due to the $(^3\text{He}, p)$ and $(^3\text{He}, d)$ reactions) can be easily recognized. However, the most interesting ones are the four bumps clearly distinguishable at higher energies, due to the $^{13}\text{C}(^3\text{He}, \alpha)^{12}\text{C}$ reactions, that are the subject of the present work. Dedicated cuts performed by hand on each α particle bumps were performed for each pseudo-telescope, without any summing procedure that could degrade the achieved identification.

The identification of the different decay channels in the studied reactions can be unambiguously confirmed through the analysis of the kinematic loci (E_α, θ) of the *well identified* α particles. They are shown in the main panel of Figure 4.6 for a reference bombarding energy. As evident, data are in excellent agreement with the different kinematic calculations (red lines) based on the α channels populating the residual ^{12}C nucleus in the 0, 4.44, 7.65 (Hoyle) and 9.63 MeV excited states. In the backward hemisphere, the identification of the α_2 particles is partially limited by threshold effects, which is of around 1.4 MeV per nucleon. The double check with the kinematic loci is crucial to discriminate the α particles which may come from ^{12}C contaminants on the target, since the slope of the kinematic line, dependent from the target mass, would be different.

The separation between the different loci (except for some background and contaminant counts coming from α s associated to the 10.84 MeV 1^- state in ^{12}C) and their excellent agreement with the kinematic calculation, in both the backward and forward hemispheres, are seen in the whole bombarding energy range of the experiment.

After the data reduction through the selection of the $^{13}\text{C}(^3\text{He}, \alpha_{0,1,2})^{12}\text{C}$ reaction events, the alpha particle yields have been transformed into absolute differential cross section data based on the measured integrated current, the areal density of the target, and the corrections for the dead time and the solid angles subtended by the detectors, all adding up to an estimated uncertainty of $\approx 8\%$. Some examples of obtained angular distributions are reported in Figure 4.7, and are found to be in reasonable agreement with previously reported observations [229, 232, 235].

4.2.2 J^π assignment of excited states in ^{16}O

The angular distribution data discussed in the previous Section and related to the α_0 and α_1 channels were decomposed in Legendre polynomials, truncated to the fourth-order, with B_i ($i = 0, \dots, 4$) indicating the coefficients of the expansion; the choice of the maximum order of the Legendre Polynomial fit was made by a critical χ^2 values analysis [236]. The present truncation would limit the entrance orbital angular momenta *mainly* to $\ell_{in} \leq 2$, i.e. the dominant terms in the present (well under barrier) scenario. Experimental data are satisfactorily reproduced by the Legendre polynomial fit, as indicated by the red solid lines of Figure 4.7; $\chi^2/\text{d.o.f.}$ values, determined by considering only statistical uncertainties, are in the range $\approx 0.8 - 3.5$. The data of the α_2 angular distributions were not included in this procedure of analysis mainly because of the possible presence of threshold effects at backward angles, that could bias the Legendre polynomial decomposition.

By using all the rules discussed in details in Section 2.3.2, it is possible to interpret the angular distributions obtained in the present experiment. If one firstly focuses the

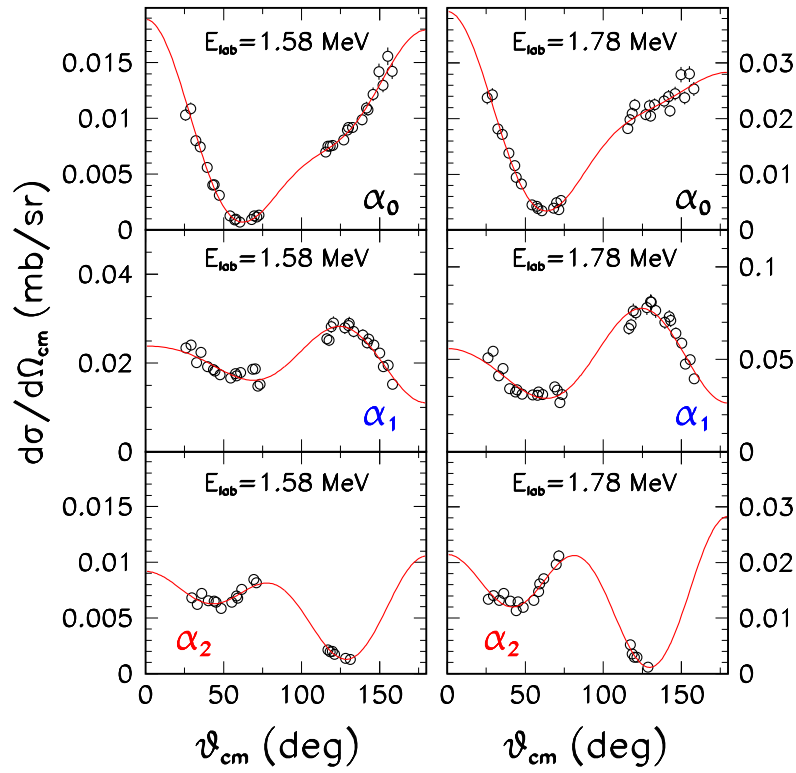


FIGURE 4.7: Angular distributions of the $^{13}\text{C}(^3\text{He}, \alpha_{0,1,2})^{12}\text{C}$ reactions at bombarding energies of 1.58 and 1.78 MeV. The error bars indicate the statistical uncertainties, while red solid lines are the Legendre polynomial fit of the experimental data.

attention to the $^{12}\text{C}_{gs}$ transition (α_0 channel), it is only possible to populate states of *natural parity* in the compound nucleus. Some considerations will be then generalized to the α_1 transition, leading to the 4.44 MeV 2^+ state in ^{12}C .

In all the cases here explored, the resonances observed in the experimental excitation functions are *broad* and *not isolated*, as it can be deduced from the quite smooth behavior of the integrated cross sections reported in Figure 4.10, that will be discussed in details later. It is then necessary to inspect (a) which is the maximal *even* term of the Legendre coefficients B_i showing anomalies (bumps or dips) in the unfolding of angular distributions as a function of energy, and (b) if there are interference terms (*odd* coefficients showing anomalies) *in between* two resonant contributions.

In the low energy domain ($E_{cm} \simeq 1.2 - 1.4$ MeV), it is possible to note a *bump* in the B_2 coefficient (red triangles) with a simultaneously small and almost flat B_4 term, as reported in Figure 4.8. The bump of the B_2 term can be better evidenced when this coefficient is normalized to the B_0 term, as reported in the insert of Figure 4.8, or when the B_2 coefficients are normalized for the Gamow factor. This anomaly in the Legendre coefficients can be attributed to the excitation of a state at 24.1 MeV, for which some limited spectroscopic observations have been reported in the literature [222]. Recalling the theory described in Section 2.3.2, this would imply a maximum value $L = 1$ and then:

$$\ell_i \geq 1 \quad J = \ell_f \geq 1 \quad (4.3)$$

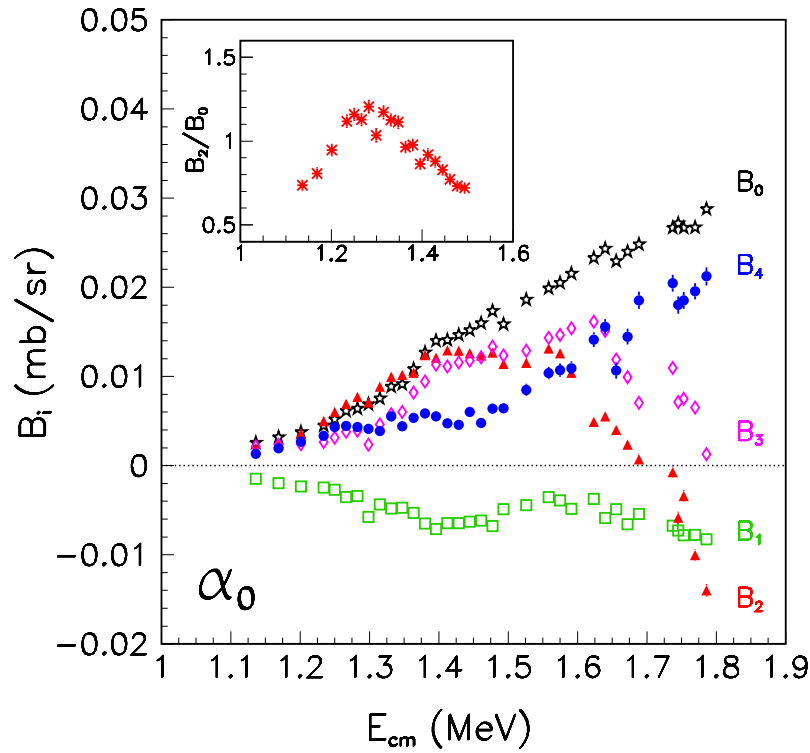


FIGURE 4.8: B_i coefficients distributions as functions of the c.m. energy, obtained from the Legendre polynomial expansion of the experimental angular distributions for the α_0 channel. The insert shows the energy trend of the specific B_2/B_0 ratio in the region between 1.1 and 1.5 MeV.

with at least one of the three values (ℓ_i, J, ℓ_f) being equal to 1. The channel spin of the entrance channel is not univocal, since it comes from the vector sum of the $1/2^+$ value of the ^3He projectile with the $1/2^-$ value of the ^{13}C of the target nucleus. At variance, the exit channel (α particle + ^{12}C in the ground state) has unambiguously *zero* channel spin, with positive channel parity. The entrance and exit channel spin will therefore be equal to:

$$s_i = (0^-, 1^-) \quad s_f = 0^+ \quad (4.4)$$

Moreover, the only allowed values for J^π are those with *natural parity*; this means that if ℓ_f is *even*, ℓ_i *must thus be odd* (and *vice versa*), since it has *negative channel parity*. Equation 4.3 then reads as:

$$\ell_i = 1, 2, 3, \dots \quad J = \ell_f = 1, 2, 3, \dots \quad (4.5)$$

However, since the experiment is performed at well under-barrier beam energies, this fact would *strongly suppress* all the $\ell_i \geq 3$ values; the previous inequalities would therefore give $\ell_i = 1, 2$ and $J = \ell_f = 1, 2, 3, \dots$. These values must be again selected, on the basis of the *possible* parity of the resonant state in the compound nucleus (CN). It is possible to distinguish between two cases: if the CN has *positive* parity, the values are restricted to $\ell_i = 1$ and $J = \ell_f = 2, 4, 6, \dots$. It will then follow that $J = \ell_f = 2$; the $J = \ell_f = 4, 6, \dots$

values must be discarded because it wouldn't be possible to obtain them from the sum:

$$\vec{J} = \vec{\ell}_i + \vec{s}_i \quad (4.6)$$

Following that, the triplet of allowed values of angular momenta are, respectively $(\ell_i, J, \ell_f) = (1, 2, 2)$ and the resonant state would have $J^\pi = 2^+$.

If the CN would have *negative* parity, it must be $\ell_i = 2$ and $J = \ell_f = 1, 3, 5, \dots$; but, since the value $L = 1$ *must* appear for at least one of the three elements (ℓ_i, J, ℓ_f) it will follow that $(\ell_i, J, \ell_f) = (2, 1, 1)$ and $J^\pi = 1^-$. The result is that the assignment of the J^π for the excited state at 24.1 MeV, is (still) *ambiguous* between the two values of 2^+ and 1^- .

In the high energy domain ($E_{cm} \simeq 1.6 - 1.9$ MeV), the value of the B_4 term highly increases, while the B_2 shows a pronounced dip. This behavior can reflect the occurrence of another broad state at excitation energy close to 24.5 MeV, reported sometimes in the literature [222]. It was a true shame that it was not possible to follow the decrease of the B_4 term above $E_{cm} = 1.9$ MeV because of reaching the maximum energy available at the AN2000 accelerator. Anyway, following the same procedures described above, and considering (as usual, given the low beam energy) $\ell_i < 3$, it is still possible to discriminate two cases depending on the parity of the compound nucleus. If the CN would have *negative* parity, it would follow that $\ell_i = 2, 4, 6, \dots$ and $J = \ell_f = 3, 5, 7, \dots$; but values of $\ell_i \geq 4$ can be neglected because of the hindrance due low energy of the beam. This will leave only $\ell_i = 2$, and thus $J^\pi = 3^-$, the only allowed value available from the coupling between $\vec{\ell}_i + \vec{s}_i = \vec{2} + \vec{1}$. The $\vec{\ell}_i + \vec{s}_i = \vec{2} + \vec{0}$ coupling, given by the alternative 0^- entrance channel spin choice, is discarded because it would lead to a $J^\pi = 2^-$, which is not acceptable due to the constraints on the *natural parity* of the states; furthermore, it does not appear in the series of required $J = \ell_f = 3, 5, 7, \dots$, exactly as the $J^\pi = 1^-$ that could come from the $\vec{\ell}_i + \vec{s}_i = \vec{2} + \vec{1}$ alternative coupling.

If the CN would have *positive* parity, the associated values would be $\ell_i = 3, 5, 7, \dots$ and $J = \ell_f = 2, 4, 6, \dots$. In this case, the problem would arise from the fact that ℓ_i must be lower than 3 because of the strong hindrance due to the barrier, giving no solutions to the problem. So, for the *high energy state* (24.5 MeV) of the ^{16}O compound nucleus, it is possible to suggest a $J^\pi = 3^-$ assignment.

Now, it would be interesting to try remove the ambiguity still present in the $J^\pi = 1^-, 2^+$ values suggested for the state at 24.1 MeV. In this respect, a very useful *hint* can be obtained through the presence of a bump in the *odd* B_3 term at intermediate energies in between the two states ($E_{cm} \simeq 1.3 - 1.7$ MeV); this peculiar behavior would indicate the interference between two close-lying *opposite parity* states. Considering that the high energy state is, very likely, a $J^\pi = 3^-$, this would suggest a *removal* of the ambiguity for the 24.1 MeV state, leading to a $J^\pi = 2^+$ assignment.

4.2.3 The case of the α_1 channel

The situation for the B_i coefficients, as a function of E_{cm} , for the α_1 channel is reported with different colors and symbols in Figure 4.9. It is possible to apply the same procedure of the angular distributions analysis seen before also to the α_1 channel, as a *further*

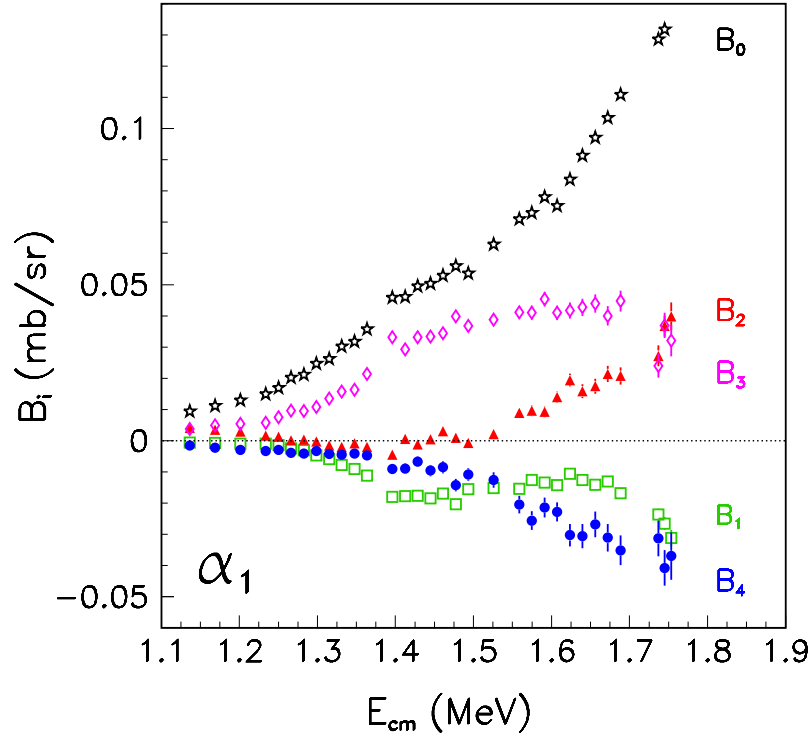


FIGURE 4.9: B_i coefficients distributions as functions of the c.m. energy, obtained from the Legendre polynomial expansion of the experimental angular distributions for the α_1 channel.

counter-proof of the previous assignments. Obviously, in this case, much more ambiguities could be present because of the *non-zero* channel spin of the outgoing channel, $s_f = 2^+$.

At *low energy* ($E_{cm} \simeq 1.2 - 1.4$ MeV), i.e. in the region of the first resonance at 24.1 MeV (previously suggested to have $J^\pi = 2^+$), it is possible to see how the dominant term is the B_0 one, with the presence of a *large* B_3 term which suggests the occurrence of *interferences* between close-lying states with opposite parities. Table 4.1 reports the *allowed couplings* between ℓ , s , and J for the α_1 channel, with the ℓ_i values truncated to 3 and the ℓ_f values truncated to 4; both *natural* and *unnatural* parity states in the CN have been included in the Table.

The rule discussed in the previous Section indicates that, in this case, it must necessarily be $\ell_i \geq 0$; $J \geq 0$; $\ell_f \geq 0$, with at least one of the (ℓ_i, J, ℓ_f) values being equal to *zero*. As shown in the Table, the $J^\pi = 2^+$ assignment previously made from the α_0 data is *compatible* with the observed trend of angular distributions in the α_1 data. In fact, in this case, $\ell_f = 0$. However, it is worth noting that the $J^\pi = 1^-$ assignment can not be discarded only based on the α_1 analysis: if $\ell_i = 0$, this value is allowed.

In the *high energy domain* ($E_{cm} \simeq 1.6 - 1.9$ MeV), the B_4 term shows a pronounced dip, and thus $L = 2$. It will follow that $\ell_i \geq 2$, $J \geq 2$, $\ell_f \geq 2$, with at least one of the (ℓ_i, J, ℓ_f) values being equal to *two*. From the α_0 analysis, it was suggested $J^\pi = 3^-$. In Table 4.1, the 3^- value is obtainable only in two ways: $(\ell_i, J, \ell_f) = (2, 3, (1, 3))$: one of

s_i	ℓ_i	J^π	ℓ_f	s_f
1^-	1	0^+	2	2^+
1^-	$\begin{pmatrix} 0 \\ 2 \end{pmatrix}$	1^-	$\begin{pmatrix} 1 \\ 3 \end{pmatrix}$	2^+
1^-	1	1^+	2	2^+
1^-	$\begin{pmatrix} 1 \\ 3 \end{pmatrix}$	2^+	(024)	2^+
1^-	2	2^-	$\begin{pmatrix} 1 \\ 3 \end{pmatrix}$	2^+
1^-	2	3^-	$\begin{pmatrix} 1 \\ 3 \end{pmatrix}$	2^+
1^-	3	3^+	$\begin{pmatrix} 2 \\ 4 \end{pmatrix}$	2^+
0^-	2	2^-	$\begin{pmatrix} 1 \\ 3 \end{pmatrix}$	2^+
0^-	3	3^+	$\begin{pmatrix} 2 \\ 4 \end{pmatrix}$	2^+

TABLE 4.1: s, ℓ, J couplings allowed for the α_1 channel (both *natural* and *unnatural* parity states are reported). *Note:* the maximum ℓ_i value is truncated to 3 and the maximum ℓ_f value is truncated to 4.

these options is actually *compatible* with the requirements given by the limiting value $L = 2$ value, supporting the previous assignment. Considering the presence of *large odd terms* (especially the B_3), between the regions of low and high energy where the two states at 24.1 and 24.5 MeV contribute to the cross section, a further confirmation of their *opposite parities* is obtained. In the end, the α_1 analysis gives some further strength to the J^π assignments performed in the previous Section for the two states at 24.1 and 24.5 MeV.

4.2.4 Angular distributions and integrated cross sections

The trends of the angular distributions and the Legendre polynomials analysis support the hypothesis of dominance of the Compound Nucleus mechanism, in agreement with the old findings of Ref. [232], obtained at slightly higher energies. By comparison of the present results to the literature database, it is possible to suggest a correspondence of the states here proposed with the ones reported in $^3\text{He} + ^{13}\text{C}$ systems at 24.1 MeV ($\Gamma \simeq 450$ keV) and 24.4 MeV ($\Gamma \simeq 250$ keV), for which there were no previously reported J^π assignments [222]. It is also likely that the 24.5 MeV state here found would correspond to the isoscalar 24.36 ± 0.07 MeV state seen in neutron decay of ^{16}O and tentatively reported to have $J^\pi = 2^+$ or 3^- [222].

Thanks to the Legendre polynomials decomposition, it was also possible to estimate the integrated cross sections (σ) for all the investigated reaction channels, and their evolution with energy (Figure 4.10: α_0 (black squares), α_1 (blue stars) and α_2 (red dots) channels). The angular distributions, crucial to estimate the value of the integrated cross section, often show a moderate *asymmetry* between the forward and backward hemispheres. However, strong forward (or backward) peaking effects, that are typical of more direct processes, are not observed in the whole energy domain here explored, as shown in the insert of Figure 4.10, where the evolution of the shapes of α_0 angular distributions is reported for each bombarded energy. This finding would give further credit to the dominance of the compound nucleus reaction mechanism.

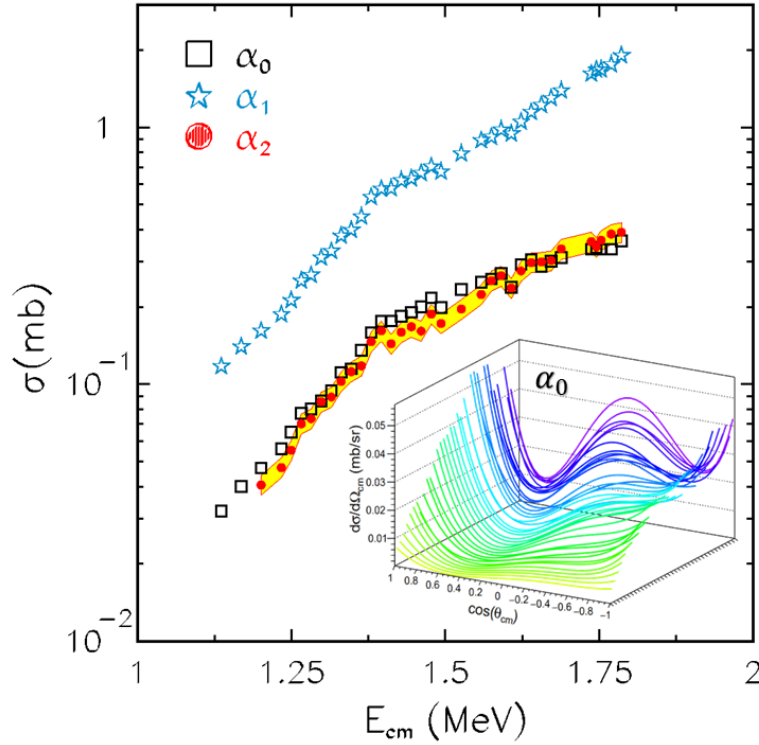


FIGURE 4.10: Integrated cross sections for the α_0 (black squares), α_1 (blue stars) and α_2 (red dots) reaction channels; the statistical uncertainties, reported in the error bars, are generally smaller than the size of the symbols. The non-statistical uncertainties amount to approximately 8% of the value for all the reaction channels. The yellow band in the α_2 data set represents the uncertainty due to the shape of the angular distributions in the backward hemisphere, derived from the possible presence of detection threshold effects. Inset: evolution of the α_0 angular distributions as a function of the bombarding energy; the increase in energy is represented from the evolution of the color scale, from yellow (1.4 MeV) to violet (2.2 MeV).

For the α_2 reaction channel, at backward angles, the emitted alpha particles can be partly cut by identification threshold effects for some bombarding energies, while no biases are observed at forward angles. Anyway, the experimental α_2 angular distributions have quite similar shapes in the whole bombarding energy range here explored. To avoid fluctuations in the reconstruction of the α_2 integrated cross section, the effect of the backward-to-forward asymmetry of angular distributions in the integrated cross section has been evaluated via an *average* correction factor obtained from the experimental data ($\langle \frac{\sigma_{back}}{\sigma_{forw}} \rangle \simeq 0.51$); similar factors are found also for the two angular distributions at $E_{lab} < 2$ MeV reported in Ref. [232]. The integrated cross section of the α_2 channel was then estimated by integrating the unbiased data at forward angles with the application of the correction factor. The yellow band reported in Figure 4.10 represents the effect of a $\pm 1\sigma$ variation in the correction factor. The maximum relative statistical uncertainties for the $^{13}\text{C}(^3\text{He}, \alpha_{0,1,2})^{12}\text{C}$ integrated cross sections are smaller than a few percent, leading to error bars smaller than the symbols of Figure 4.10. Non-statistical uncertainties amount to $\approx 8\%$ for all the reaction channels.

4.2.5 Branching ratio of the Hoyle and ground state transitions

The study of highly excited ^{16}O states at $E_x \geq 15$ MeV is very interesting, since their de-excitation can proceed through the emission of α particles to the ground or low-lying ^{12}C excited states. As discussed in the previous Sections, the present experiment is strongly tied to a compound nucleus scenario; in this framework, the cross section ratio between the α_2 and the α_0 channels ($\frac{\sigma_{\alpha_2}}{\sigma_{\alpha_0}}$) is linked to the branching ratio of the respective partial widths, $\frac{\Gamma_{\alpha_2}}{\Gamma_{\alpha_0}}$, which are connected to the reduced partial widths (γ_α^2) through the quantum penetrability (P_{ℓ_f}), as expressed in the equation $\Gamma_\alpha = 2 \cdot P_{\ell_f} \gamma_\alpha^2$ from Ref. [237]. Since both the α_0 and the α_2 transitions have a channel-spin equal to zero, the ℓ_f values of a ^{16}O state, with a given J^π , involved in these transitions must be fixed and identical. The partial width of a state is a crucial parameter in the investigation of its properties, since it is proportionally related to the probability that, in the respective parent ^{16}O state, the nucleons could be arranged in a configuration similar to that of the particles produced by its decay. The ^{12}C Hoyle state, as discussed in Chapter 3, is characterized by a structure with a strong α cluster components; therefore, oxygen states which emit α particles and decay into ^{12}C with a $\gamma_{\alpha_2}^2 > \gamma_{\alpha_0}^2$ ordering would be interesting candidates to show a pronounced α -clustered nature. Considering the dependence of the partial widths from the quantum penetrability, it is possible to express the branching ratio of the two decay ways as:

$$BR = \frac{P_{\ell_f}(\alpha_2) \gamma_{\alpha_2}^2}{P_{\ell_f}(\alpha_0) \gamma_{\alpha_0}^2} \quad (4.7)$$

In this framework, the penetrability for the two channels can be calculated through the regular and irregular Coulomb functions, for various $\ell = 0, 1, 2, 3$ values.

In Figure 4.11, the results for the $\frac{\sigma_{\alpha_2}}{\sigma_{\alpha_0}}$ branching ratio obtained in the present experiment are shown as a function of the excitation energy of the ^{16}O compound nucleus, alongside with those reported by previous literature experiments at higher energies [231, 232]. The values are in reasonable agreement within their mutual uncertainties. Data from the $^{14}\text{N}(d, \alpha_{0,2})^{12}\text{C}$ reactions [228] are also reported, at lower energies, since the kinematics and the Q -values of these reactions are very similar to the present ones, allowing to justify all the considerations on the involvement of low- ℓ partial waves in the entrance channel made before. A difference is however present, since a non-vanishing contribution could come from the direct reaction mechanism in the $^{14}\text{N}(d, \alpha_0)^{12}\text{C}$ channel [192, 228], which would enhance the cross section with respect to what happens in a pure compound nucleus scenario. An increase in the cross section of the α_0 channel would of course *lower* the apparent branching ratio, with respect to the effective one. For these reasons, the data coming from the (d, α_0) reaction should be treated as *lower limits* of the effective ^{16}O states $\frac{\Gamma_{\alpha_2}}{\Gamma_{\alpha_0}}$ branching ratios.

The overall $BR(E_x)$ trend is characterized by prominent peaks and dips, a typical behavior when the compound nucleus mechanism is involved, leading to the excitation of several states. The lines reported in Figure 4.11 are related to the penetrability ratios $\mathcal{R} = \frac{P_{\ell_f}(\alpha_2)}{P_{\ell_f}(\alpha_0)}$ calculated for four different orbital angular momenta in the outgoing channels ($\ell_f = 0, 1, 2, 3$, respectively in black, red, blue and green colors) by assuming a radius parameter $r_0 = 1.4$ fm.

Given that the branching ratio is the product between the penetrability and the reduced partial width ratios ($BR = \mathcal{R} \cdot \frac{\gamma_{\alpha_2}}{\gamma_{\alpha_0}}$), values of the experimental Branching Ratios which

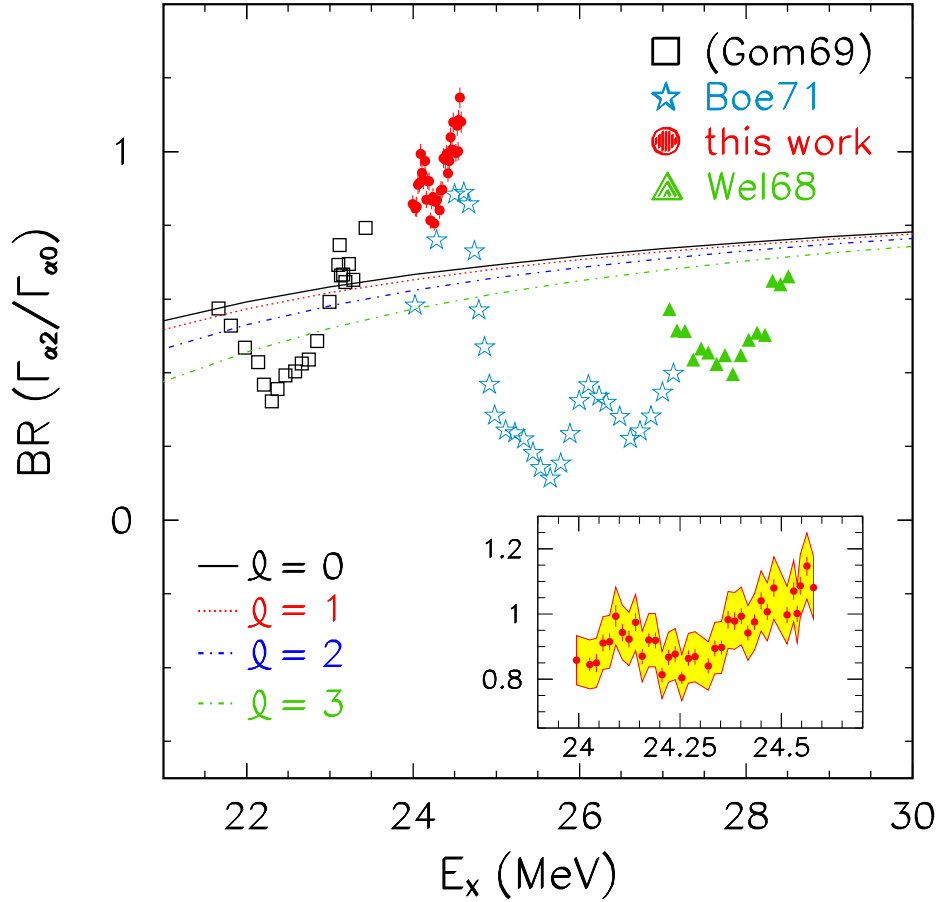


FIGURE 4.11: Branching ratio of the Hoyle state versus the ground state transition $\left(\frac{\Gamma_{\alpha_2}}{\Gamma_{\alpha_0}}\right)$, observed for some reactions populating highly excited ^{16}O compound nuclei as intermediate states. Experimental data: for $^3\text{He} + ^{13}\text{C}$ reactions, Ref. [231] (green triangles) and Ref. [232] (blue stars); $^2\text{H} + ^{14}\text{N}$ reaction, Ref. [228] (black squares). Literature uncertainties are not reported for clarity reasons. Data from this work are represented by red dots with error bars. The colored lines represents the penetrability ratios R for s -, p -, d - and f -waves in the final channel, respectively reported in black, red, blue and green. The insert shows a zoomed view of the present data, where the yellow band encapsulates the uncertainty due to the reconstruction of the angular distributions in the backward hemisphere.

are above the trends given by the $\mathcal{R}$ lines, would directly indicate states with $\gamma_{\alpha_2}^2 > \gamma_{\alpha_0}^2$, i.e. with a potential α cluster nature. This indeed happens for data coming from the present experiment and from Ref. [232], in the $E_x \approx 24 - 25$ MeV region, where the $\mathcal{R}$ lines are clearly exceeded. A similar situation is seen for data from Ref. [228] at $E_x \approx 23.5$ MeV for the $^{14}\text{N}(d, \alpha_{0,2})^{12}\text{C}$ data. Another aspect worth noting, in the light of interesting future explorations, is the increasing trend shown by the same data at the lowest energies ($E_x \leq 22.5$ MeV).

In the framework of the nowadays available models, the nature of these anomalies is not actually clear and calls for further deepening both from the experimental and theoretical sides. For example, given that the J^π of the two investigated states can

possibly be 2^+ and 3^- , a possible hypothesis could involve the rotational excitation of some quartet states [73, 238] with a developed 4α structure, for which the band-heads were proposed to exist at high excitation energies, larger than 23 MeV. Nonetheless, it is not easy to ascertain these hypotheses without new theoretical calculations, involving in details the coupling between the nuclear structure of the ^{16}O excited states and detailed penetrability calculations, also taking into account the anomalously broad, diluted and clustered distribution of matter in the Hoyle state.

4.3 Some concluding remarks

Since the beginning, the ^{16}O nucleus has been one of the main actor of the nuclear cluster physics theater: the possible presence of a tetrahedral structure, and its influence on the sequence of states produced by roto-vibrational excitation of this symmetric assembly, was discussed in details by Dennison [239] in a pioneer work in 1940. In this respect, and considering that many other subsequent theoretical works suggested the occurrence of tetrahedral structure even in the ground state, it seems very opportune to deepen the study, discussed in Section 4.1.2, on the presence of coherent effects regulated by the Wierl formula on elastic scattering of protons (or neutrons) on ^{16}O at intermediate energies. Several experimental difficulties must be anyway faced, starting from the use of suitable target containing ^{16}O ; in this context, the use of supersonic jet targets can be an interesting solution, even if the typical target thickness are quite low (of the order of a few $\mu\text{g}/\text{cm}^2$ maximum) and very long beam times are therefore needed. The use of neutrons as wave matters could of course further simplify the Optical Model parameterization of the spherical contribution to the elastic scattering.

Also the high energy part of the ^{16}O spectrum merits further investigations that could lead to surprising findings. The quite large branching ratios $\frac{\alpha+^{12}\text{C}_{\text{Hoyle}}}{\alpha+^{12}\text{C}_{\text{gs}}}$ of the α decays observed for states at about $E_x \approx 24.5$ MeV is a very appetizing finding to study the onset of very diluted α clustered states at very large excitation energies. Therefore, it seems very useful to continue the branching ratios analyses also in regions where data are still missing. For example, the region at $E_x \approx 23.5 - 24.0$ MeV is not covered by data coming from both the $^3\text{He}+^{13}\text{C}$ or $\text{d}+^{14}\text{N}$ reactions, and this is a pity, because the branching ratio data coming from the adjacent region at $E_x \approx 23.2$ MeV are still well higher than the calculations based on the barrier penetration model. In the same fashion, the extrapolation of data in the $E_x < 22$ MeV region could suggest an overcoming of the penetrability calculations that would call for further dedicated measurements. Finally, it is worth noting that $\text{p}+^{15}\text{N}$ reactions can also populate ^{16}O states α decaying into several states of ^{12}C (including the Hoyle state), and therefore can represent a further experimental way to study the trend of branching ratio also at lower excitation energy regions.

Chapter 5

Quartet states of ^{20}Ne at high excitation energies

After the ^{12}C and ^{16}O cases, the ^{20}Ne is another self-conjugate nucleus showing very peculiar aspects linked to the occurrence of α clustering. Differently from the ^{12}C (just weakly deformed) and ^{16}O (spherical, according to the shell model) ground states, the ^{20}Ne has a quite large quadrupolar deformation ($\simeq -0.23\text{ e b}$) that heavily affects several aspects of its spectroscopy. It was already discussed in Chapter 1 (Section 1.2.3.4) how the decomposition of the ^{20}Ne into two sub-clusters, both doubly magic nuclei (^4He and ^{16}O), would give rise to parity-doubled rotational bands characterized by large moment of inertia. Other models, from the historical ones of Wefelmeier and Hafstad-Teller, to the more recent Algebraic Cluster Model, considered the ground state of the ^{20}Ne nucleus as a bi-pyramidal 5α structure with D_{3h} symmetry [240]; in this way, it was possible to obtain reasonable descriptions of form factors, radii and electromagnetic transition parameters involved in this nucleus spectroscopy. As shown in [240], all the nine normal vibrational modes which can be predicted applying the bi-pyramid configuration and the D_{3h} symmetry, are reflected in the known experimental level scheme.

At large excitation energies, more complicated α cluster effects can influence the structure of ^{20}Ne . For example, the α decay of excited states above $E_x \simeq 11\text{ MeV}$ can occur leaving the ^{16}O residual nucleus into its highly-clustered 6.05 MeV second 0^+ state (see the discussions reported in Sections 1.2.3.4 and 1.2.3.5), and the analysis of the reduced partial widths (see Section 2.3) can represent a *mine* of information to understand the behavior of the wavefunction of such highly excited states in ^{20}Ne . In this Chapter, the focus of the attention will be on this peculiar aspect of the α cluster structure of ^{20}Ne , by searching for possible *quartet* excitations at excitation energies larger than 14 MeV (see Section 1.2.3.5), whose existence was suggested in Ref. [73], but never firmly established. To perform these investigations, $p+^{19}\text{F}$ nuclear reactions have been exploited to form ^{20}Ne excited states, and the data coming from the α decay of such states were analyzed through detailed R matrix fit to the experimental data. The details of the R matrix formalism were discussed in Section 2.3.1, and the Reader is referred to Sections 2.3 and 2.3.1 for aspects concerning the extraction of the reduced partial widths, the estimate of single particle partial widths (Wigner limits) and the calculation of channel radii.

5.1 The $^{19}\text{F}(\text{p}, \alpha)^{16}\text{O}$ reaction and the structure of ^{20}Ne

The long history of the $^{19}\text{F}(\text{p}, \alpha)^{16}\text{O}$ reactions begins in the first part of the 30's, when John Cockroft and Ernest Walton, both under the doctoral advisory of Ernest Rutherford, began their studies on the first nuclear reactions and transmutations, finding evidences of the emission of long range (i.e., quite energetic) α particles induced by impinging proton beams on fluorine targets. Some historical experimental works are described in Refs. [241–244] and some acute theoretical considerations on the experimental data were drawn in 1939 by Robert Oppenheimer and Julian Schwinger [245].

5.1.1 General aspects of proton + fluorine collisions

Protons+ ^{19}F collisions are very important because, at low bombarding energies, they are dominated by a compound nucleus mechanism, with the formation of the intermediate ^{20}Ne system, and among the open decay channels it is possible to have the $\alpha_i + ^{16}\text{O}$ ones. On the basis of the historical notation prescriptions [146, 246], that will be used coherently in the following analysis, the α_0 channel indicates a linked residual ^{16}O nucleus left in its ground state, while the α_π channel is related to the 6.049 MeV 0^+ state, which decays to the ground state by pair emission (hence, the π notation), and is characterized by a pronounced *four particles - four holes* ($4p - 4h$) configuration, linked to a strongly clustered structure formed by an α particle interacting with a ^{12}C core (see Section 1.2.3.4).

$^{19}\text{F}(\text{p}, \alpha)^{16}\text{O}$ reactions, performed in a wide range of bombarding energies ($\approx 0.5 - 10$ MeV) [146, 243, 244, 246–248], were widely used in the last 60 years to study the ^{20}Ne compound nucleus spectroscopy in the $E_x \simeq 13 - 18$ MeV region, but it is only in recent times that they began to be important also in the nuclear astrophysics field. In fact, the knowledge of its cross sections may help not only to shed light on the question of fluorine nucleosynthesis in Asymptotic Giant Branch (AGB) stars [249–251], but also on the determination of the balance between the $^{19}\text{F}(\text{p}, \alpha)^{16}\text{O}$ and $^{19}\text{F}(\text{p}, \gamma)^{20}\text{Ne}$ reaction rates, important to define escape pathways in the CNOF cycle [252–254]. This is important because the radiative capture reaction provides, in hydrogen-rich stellar environments, an important channel of fluorine destruction *and* ^{20}Ne formation, paving the road for the synthesis of heavier elements, up to Calcium, in more ancient stars [254].

However, the astrophysical field is not the only one that could strongly benefit from a deeper understanding of the properties of this reaction: for example, further investigations of the α_0 and α_π channels would be very important in the nuclear structure field, given that they could help to reveal the presence of cluster structures in the ^{20}Ne nucleus. These became very important at the energies above the proton separation energy ($S_p = 12.844$ MeV), considering that this threshold is quite close to the $2\alpha + ^{12}\text{C}$ cluster decay threshold (11.89 MeV), representing one of the region of interest reported in the Ikeda diagram [63] but, up to date, unfortunately still affected by the presence of ambiguities and/or missing information from the spectroscopical point of view [65].

$^{19}\text{F}(\text{p}, \alpha_{0,\pi})^{16}\text{O}$ reactions are especially powerful for the study of these states, since conservation rules on parity and angular momentum only allow the population of *natural parity* states in the compound nucleus. Furthermore, it is also easy to demonstrate that the angular momenta in the entrance channel, in the outgoing one, and in the resonant

state of the compound nucleus (respectively, ℓ_i, ℓ_f and J) must be *equal* [146]. $^{19}\text{F}(\text{p}, \alpha_{0,\pi})^{16}\text{O}$ reactions, if performed at low bombarding energies (where only low- ℓ_i partial waves are involved because of the presence of hindrance effects due to the centrifugal barrier) would thus be highly selective, favoring the population of low J states in the ^{20}Ne compound nucleus.

These selective features are extremely important to highlight the presence of a peculiar class of cluster states, i.e. the ones with $J^\pi = 0^+$, whose existence in the region of high excitation energies ($\approx 13 - 16$ MeV) was predicted many years ago from the quartet model (see Section 1.2.3.5) [73]. In this framework, the situations of the 14.47 and 14.92 MeV 0^+ states of ^{20}Ne are particularly interesting, because some previous works suggested their possible quartet nature [75, 76] while, at variance, an opposing interpretation was also proposed for the first state, based on which it could be seen as the $T = 1$ isologue of the 4.435 MeV state of the isobar ^{20}F nucleus [255]. Then, one of the aims of the experiment presented in this Chapter is to provide some clarifications on these questions.

Indeed, in the last years, new theoretical interpretations and experimental measurements of the $^{19}\text{F}(\text{p}, \alpha_0)^{16}\text{O}$ reaction have been produced [256–260], following the renewed interest in the reaction. However, it is still possible to find some data-poor regions in the α_π channel and some persisting discrepancies between data sets in the α_0 channel excitation functions, as discussed in details in Refs. [261, 262]. On the basis of this, other aims of the present work are:

- to provide new data in the poorly known 1.1 - 1.3 MeV center of mass energy region in the α_π channel [262];
- to try to solve the strong disagreement between the available data around $E_{cm} \simeq 1.7$ MeV in the α_0 channel. As a matter of fact, despite accurate normalization procedures (as described in Ref. [261]), the data from Ref. [263] still appears $\approx 50\%$ larger than those of Ref. [75]. Up to today, this stands as the main discrepancy in the available compilations of $^{19}\text{F}(\text{p}, \alpha_0)^{16}\text{O}$ data, preventing a solid spectroscopic analysis and the unambiguous determination of the partial widths for the 0^+ (and neighboring) states at 14.47 MeV in ^{20}Ne .

As an additional note, it is important to highlight the difficulty in the measurement of the $^{19}\text{F}(\text{p}, \alpha_\pi)^{16}\text{O}$ reaction cross section, since the state at 6.049 MeV in the residual ^{16}O lies dangerously close in energy to the subsequent 6.130 MeV (only ≈ 80 keV difference) γ -emitting state, which can easily overwhelm the signal coming from the pair-emitting state. An experimental setup aiming to resolve this doublet in the ejectile spectrum would thus need to have very high energy resolution and to be characterized by low-background.

The next Sections will be dedicated to the description of the experimental set-up, the presentation of the internal normalization procedure adopted to determine the absolute cross section for both the $\alpha_{0,\pi}$ channels, and to the inclusion of the newly-obtained data (together with several other scattering and reaction channels) inside a wide and comprehensive R -matrix fit, allowing to obtain a new set of reduced partial widths for the $E_x \approx 14 - 15$ MeV region of the ^{20}Ne level scheme and then to draw some new hypotheses on the structure of the pertaining states.

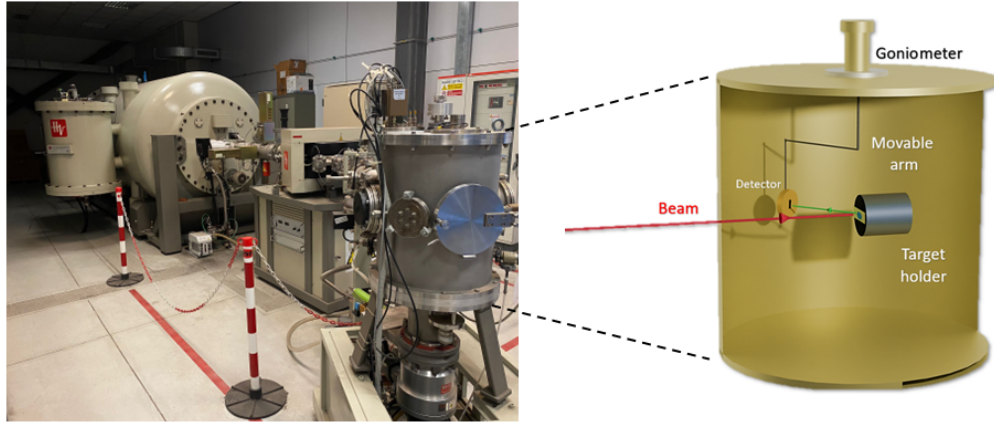


FIGURE 5.1: Picture of the experimental apparatus (accelerator and scattering chamber) and a zoom-in showing a three-dimensional render of the insides of the reaction chamber.

5.1.2 The experimental setup

The experimental measurements were performed at the Singletron accelerator of the Physics Department at the University of Catania, with energies of the proton beams spanning the windows between 1.13 – 1.30 MeV and 1.62 – 1.72 MeV; the beam energy stability was better than 1 keV, while an average intensity of ≈ 100 nA was used, with a maximum beam spot size of ≈ 2 mm². In order to build the excitation functions and the angular distributions of the cross sections, the energy of the beam was varied with steps of 10 or 20 keV, while the detector was set-up in six different angular positions, at 115°, 125°, 135°, 145°, 155° and 165°. The central polar angle was known with a one prime uncertainty, and the angular opening of the collimator on the detector was of $\approx 1^\circ$.

The used target was a calcium fluoride (CaF_2) film of 30 $\mu\text{g}/\text{cm}^2$ thickness deposited over a 10 $\mu\text{g}/\text{cm}^2$ carbon backing, both thicknesses measured with a 5% accuracy, before and after the evaporation, through the energy loss of 5.486 MeV α particles emitted by a 241-Am source. A high level of vacuum, of the order of 10^{-7} mbar (even better for some runs), was maintained in the chamber during the whole experiment, to prevent the deposition and the build-up of carbon impurities on the target. The experimental apparatus is represented in the picture and in the 3D rendering of the scattering chamber reported in Figure 5.1.

During the data-analysis, the energy of the accelerated beam was cross-checked through a careful calibration procedure, based on fine-step scanings of a well-known, benchmarked, anti-resonance trend shown by the $\text{p}+^{12}\text{C}$ elastic scattering differential cross section (DCS) at $\theta_{\text{lab}} = 165^\circ$ and $E_{\text{beam}} = 1.6 - 1.7$ MeV [264]. During this process, the $\text{p}+^{12}\text{C}$ cross section data were determined via two independent methods, i.e., an *internal normalization* of the $\text{p}+^{12}\text{C}$ events to the (Rutherford) elastic scattering yields associated to $\text{p}+^{40}\text{Ca}$ events, and *direct* estimates based on the integrated current measured on the whole chamber, used as a Faraday Cup. The results of the calibration procedure are reported in Figure 5.2 as open triangles and full dots, compared with the literature values of Ref. [264] (red line). The absolute cross section scale of the present calibration data was fixed to the one of Ref. [264]. The calibration points here obtained are in excellent

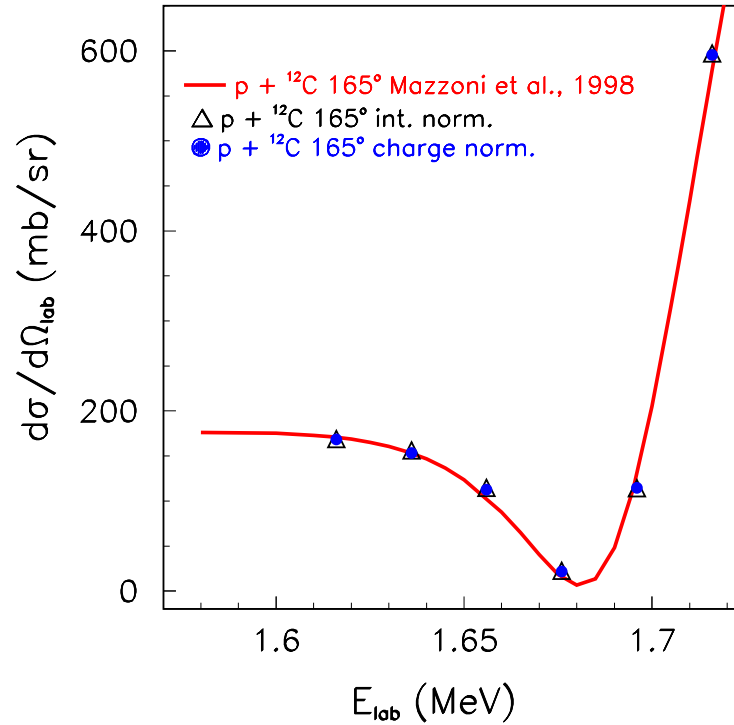


FIGURE 5.2: Verification of the beam energy calibration obtained by comparison of the present differential cross section data of $p+^{12}\text{C}$ elastic scattering at $\theta_{\text{lab}} = 165^\circ$ and analogous bench-marked experimental data reported in Ref. [264] (red line). The two different methods of analysis of the present data (internal normalization based on comparisons with the Rutherford scattering on ^{40}Ca atoms present in the target *versus* the use of the collected beam charge) are shown with different symbols. Statistical errors are smaller than the size of symbols.

agreement (discrepancies lower than 1 keV) with the reference data [264], underlining the reliability of the calibration.

The reaction ejectiles were detected with a surface barrier silicon detector, combined with a low-noise charge pre-amplifier and a spectroscopic amplifier, included in the front-end electronic chain; all the amplifier parameters were carefully tuned to enhance the experimental resolution, with particular care dedicated to the pile-up rejection, achieving a 0.3% energy resolution at 5 MeV. The energy response of the detector was calibrated through a triple-nuclide mixed α source and with proton elastic scattering points on the different elements composing the target, taking into account all the energy losses of the beam and the outgoing particles; it was thus possible to obtain an excellent linearity ($1 - R^2 \simeq 6 \times 10^{-6}$) in the whole calibration domain, between 1 and 6 MeV. During the collection of the beam current on the whole insulated chamber, used as a Faraday Cup, the charge losses due to possible escapes of backward-emitted electrons were neglected, considering the geometry of the beamline and of the chamber.

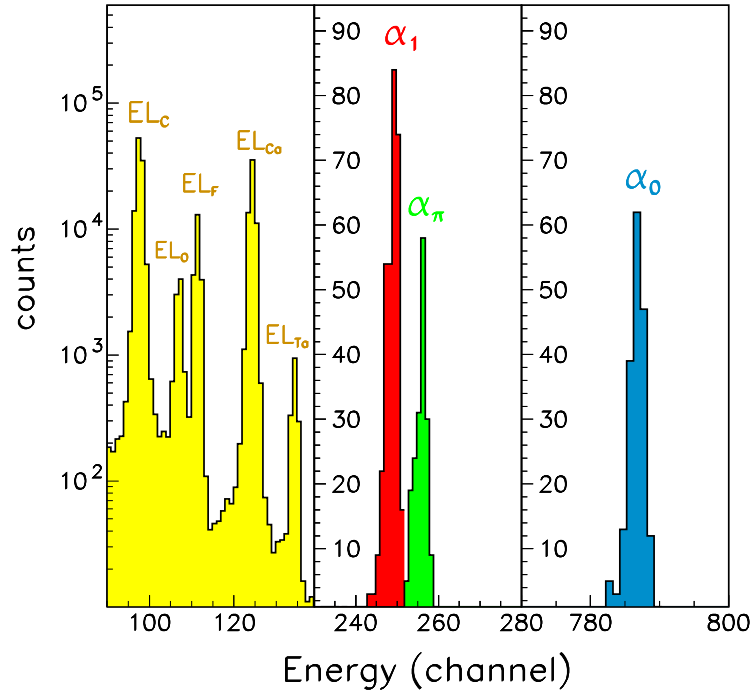


FIGURE 5.3: Energy spectra of the ejectiles emitted at $\theta = 165^\circ$ for $E_p = 1.2$ MeV protons on CaF_2 (on carbon backing) target. Horizontal and vertical scales are separated, as a function of the ejectile energy, for clarity reasons. Yellow: elastic peaks (C, O, F, Ca and Ta), logarithmic y-scale. The presence of Ta contaminants in the target is due to the tantalum crucible used to evaporate the CaF_2 layer, which has very high melting and boiling points. Peaks due to the α_1 , α_π and α_0 channels are respectively shown in red, green and blue, with a linear count scale.

5.1.3 Excitation functions and angular distributions of the $^{19}\text{F}(\text{p}, \alpha_\pi)^{16}\text{O}$ reactions

As shown from a typical ejectile energy spectrum, reported in Figure 5.3 for $E_{\text{beam}} = 1.2$ MeV and $\theta_{\text{lab}} = 165^\circ$, the obtained energy resolution for the α_π peaks is a $\approx 30\%$ better than the current state-of-the-art, represented by the data of Ref. [147]. As introduced in the previous Section, this is extremely important since it will allow to unambiguously separate the counts for the α_π events from those of the closely lying α_1 peak. The lower-energy region of the spectrum (in yellow) shows a very good separation of the elastic peaks: a multi-gaussian fit was performed to obtain the counts subtended by all the peaks, with the addition of a polynomial contribution to describe the (small) observed background.

As extensively reported in Refs. [265, 266] the possible variation of the CaF_2 target stoichiometry, i.e., of its calcium and fluorine elemental richness during the bombardment with the beam, must be taken into account, since a wrong evaluation of the ^{19}F content would obviously be misleading in the determination of the cross section. As a solution to this problem, the counts for the α_0 and α_π channels were always *internally normalized* to the elastic yield measured for the $\text{p} + ^{19}\text{F}$ elastic scattering in the same experimental conditions (θ and energy), as similarly done, for example, in [75, 147]. To obtain the

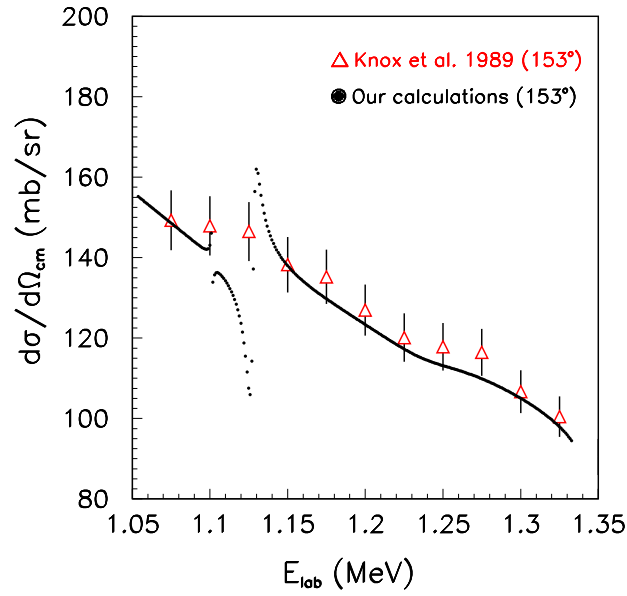


FIGURE 5.4: Comparison between the elastic scattering data by Knox and Harmon at 153° [267] (*triangles with error bars*) and the R matrix extrapolations at the same angle obtained by using the spectroscopic parameters of Ref. [262] (*black dots*).

elastic scattering data in the whole angular and energy domain needed for the internal normalization procedure, their absolute DCS have been extrapolated from the R Matrix fit performed in Ref. [262], which describes very well the experimental absolute $d\sigma/d\Omega$ data obtained from [147] at 135° and 145° . As a further check of the reliability of the extrapolations, the elastic scattering data taken from Ref. [267] at 153° , which have been recently bench-marked in the work of Ref. [268], have been compared to the R matrix extrapolations at the same angle. The agreement is excellent, within very few percent.

The expression used to obtain the DCS section for the α_0 and α_π channel, within the limits of thin target approximation and small solid angles of detections, is given by the following Equation:

$$\frac{d\sigma^{\alpha_{0,\pi}}}{d\Omega}(\theta, E) = \frac{N_{\alpha_{0,\pi}}}{N_{p,ela}} \cdot \frac{d\sigma^{p,ela}}{d\Omega} \quad (5.1)$$

where $N_{\alpha_{0,\pi}}$ are the counts under the $\alpha_{0,\pi}$ peaks and $N_{p,ela}$ are the counts under the elastic scattering peak of fluorine; the Equation 5.1 returns the DCS in the laboratory frame. This is then transformed in absolute DCS in the center of mass frame through the application of a relativistic Jacobian transformation. This method allows to determine the absolute DCS for the α_0 and the α_π channels independently from the possible fluorine content variation in the target.

Some DCS are reported, for both the α_0 and α_π channels, in Figure 5.5, at different polar angles and as a function of the bombarding energy. Error propagation principles were applied to estimate the statistical uncertainties of the measurements, including those on the count rate; moreover, a conservative 10% non-statistical uncertainty for the determination of the $p + ^{19}\text{F}$ elastic scattering DCS was assumed, considering the deviations observable between the bench-marked data sets used in [268].

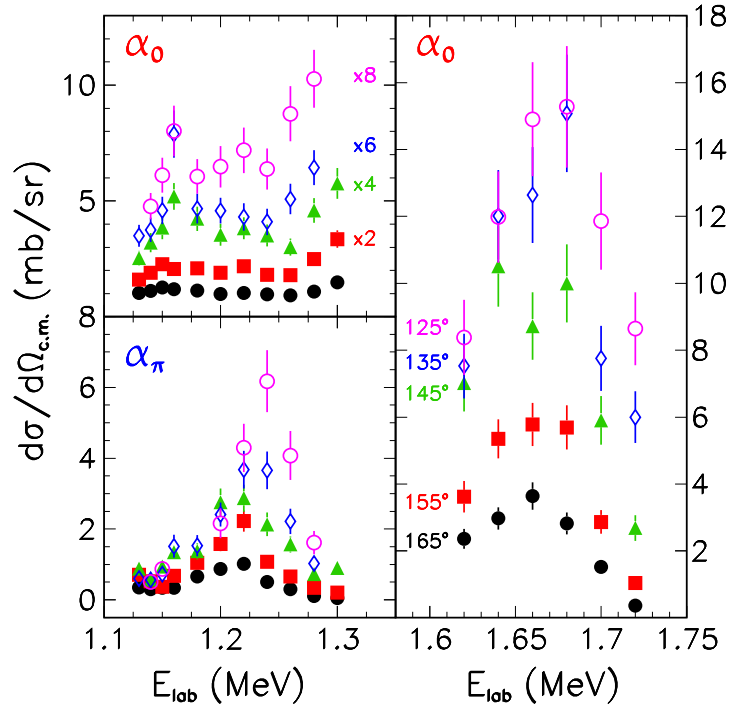


FIGURE 5.5: Excitation functions ($d\sigma/d\Omega_{cm}$) obtained from the present work, for the α_0 (Top Left panel: $E_{lab} \simeq 1.1 - 1.3$ MeV; Right panel: $E_{lab} \simeq 1.6 - 1.72$ MeV) and the α_π (Bottom Left panel: $E_{lab} \simeq 1.1 - 1.3$ MeV) channels, shown at different polar angles in the laboratory reference frame. Horizontal error bars are smaller than symbols sizes. The energy resolution is around 1 keV in the explored range. For clarity reasons, different scaling factors have been used for data at various polar angles, as indicated in the top left panel: these were, respectively, 2, 4, 6 and 8 for the 155°, 145°, 135° and 125° data.

In Figure 5.6, the excitation function $\frac{d\sigma}{d\Omega_{lab}}$ obtained at $\theta_{lab}=155^\circ$ has been compared to previous datasets obtained at $\theta_{lab}=150^\circ$ (Refs. [269, 270]). Data from the present work are in agreement, within the error bars, with the ones reported by Ref. [270]. As it was also suggested in Ref. [270], a scale factor of 2 was applied to data of Ref. [269] to match the two cross section scales. It is worth noting that a similar scaling factor was already used in the past to match the cross section scales of data from [256] and [269] at lower energies.

Since the present data were collected in the backward hemisphere, it is necessary to know the shape of angular distributions in the whole range of polar angles, in order to obtain the integrated cross sections for each energy. Such information can be derived from previous works (as Refs. [146] and [246]), in relative units or as Legendre-polynomial expansions, for both channels here investigated. The angular distribution shapes were then extrapolated at the energies of interest from Refs. [146, 246] through spline fits of B_i , the coefficients of Legendre polynomial expansions of the angular distributions, truncated to the fourth-order (i.e. considering the d -wave as the highest partial wave contributing to the cross section, as discussed in Refs. [147, 246, 262, 263]).

Then, the integrated cross sections were estimated, together with their uncertainties, for the $\alpha_{0,\pi}$ channels from the equation $\sigma(E) = A(E) \cdot 4\pi B_0(E)$. Here, $A(E)$ represents

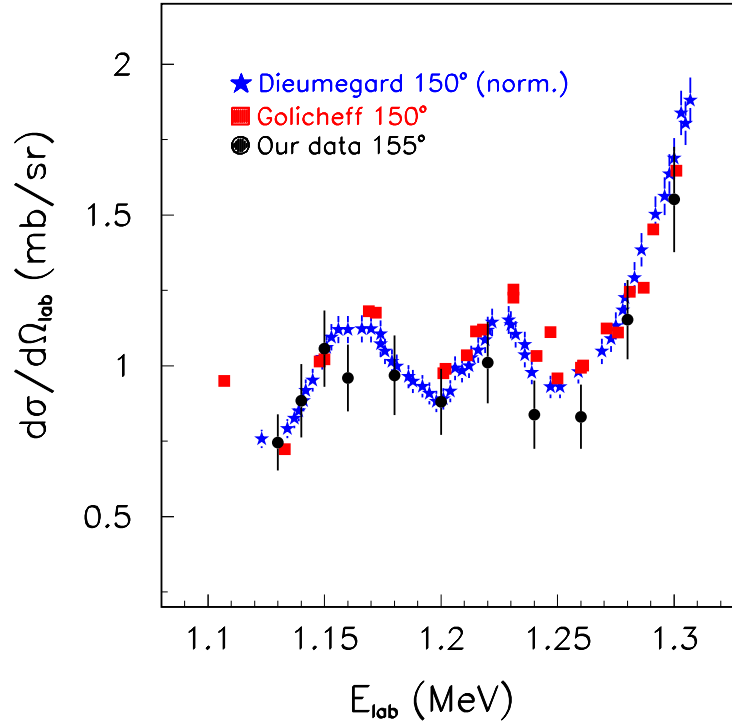


FIGURE 5.6: Excitation function $\frac{d\sigma}{d\Omega_{lab}}(E_{lab})$ at 155° obtained from the present work (black dots) and those from [270] (red squares; experimental uncertainty not indicated in the original work) and [269] (blue stars, normalized as discussed in the text).

a scaling parameter obtained by fitting this work's (absolute) angular distributions at backward angles to the overall angular distribution shapes (in relative units) extrapolated from Refs. [146, 246].

Some examples of these normalization fits can be seen in Figure 5.7. A good agreement between the present data and the trends previously reported in the literature (red and blue lines), is observed for both reaction channels.

Concerning the α_π channel, the integrated cross section data obtained in the present experiment are reported as black dots in Figure 5.8. Green triangles refer to data obtained at the INFN-LNL (Legnaro, Italy) [75] and TTT3 tandem (Naples, Italy) [271] accelerators during the '70s; the integrated cross sections coming from the second (previously unpublished) dataset, and all the assumptions made in treating them, were discussed in details Ref. [262]. A good agreement (within ≈ 2 standard deviations) between the two data sets is evident, with the exception of a slight energy shift, smaller than 10 keV, at $E_{cm} > 1.15$ MeV.

For what concerns the α_0 channel, the zone around $E_{cm} \approx 1.7$ MeV was the center of the attention in the second energy region explored in this experiment. In fact here, even after a careful normalization procedure, based on several existing data sets and already applied in Ref. [261], the data coming from the Cuzzocrea et al. (1980) [75] and Clarke and Paul [263] experiments are still evidently misaligned.

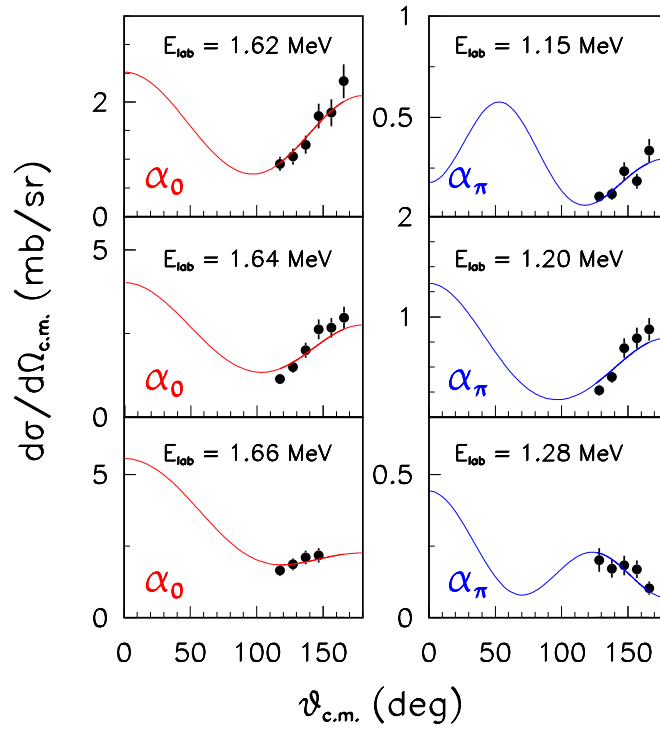


FIGURE 5.7: Angular distributions (*black dots*) of the $^{19}\text{F}(\text{p}, \alpha_0)^{16}\text{O}$ (*left panels*) and $^{19}\text{F}(\text{p}, \alpha_\pi)^{16}\text{O}^*$ (*right panels*) reactions investigated in the present work. Red and blue solid lines represent the shape of angular distributions reported in Refs. [146, 246], normalized to the present absolute cross sections data obtained in the backward hemisphere at some bombarding energies.

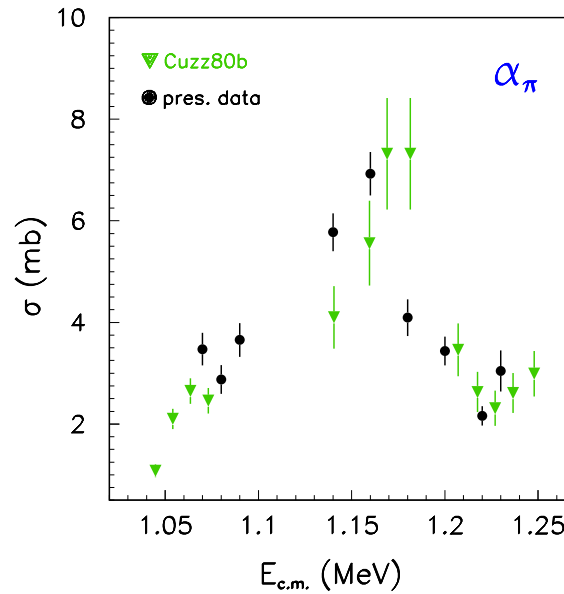


FIGURE 5.8: The experimental data obtained in the present work (*black dots*) in comparison with the measurements of Ref. [271] (*green downwards triangles*) already reported in Ref. [262].

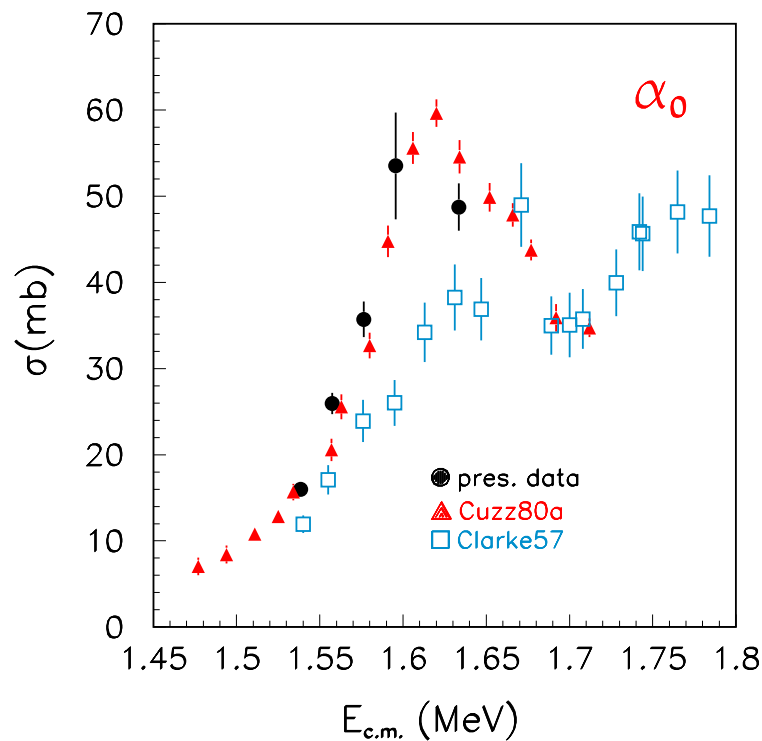


FIGURE 5.9: Comparison of the data obtained for the α_0 channel integrated cross section (σ) with previous literature data from [75] (*red triangles*) and [263] (*cyan squares*), as obtained from the global normalization of all the literature datasets made in Ref. [261].

In Figure 5.9, the data obtained from the present experiment (*black dots*) are reported along these two literature data sets (Ref. [75] as *red triangles*, Ref. [263] as *cyan squares*). As evident from the Figure, present data appear to be in much better agreement with Ref. [75] rather than with Ref. [263], even after the careful normalization of all the literature datasets performed in [261]. From a statistical point of view, the obtained data are in excellent agreement with the ones of Ref. [75] (within $\approx 1.7\sigma$); much larger discrepancies (between $\pm 3.5\sigma$) are obtained by comparing this work's data to the ones of Ref. [263] (as normalized in [261]). A possible origin of the difference between the two existing datasets may be linked to the different shapes of angular distributions (at $E_p \approx 1.7$ MeV) reported and/or to efficiency effects in evaluating the integrated current in Ref. [263].

5.1.4 The structure of ^{20}Ne at high excitation energies

To improve the spectroscopic knowledge of the excited states of the ^{20}Ne compound nucleus in the $E_x \simeq 13.7 - 14.8$ MeV region and, consequently, to draw some considerations on its structure, a comprehensive R -matrix fit was performed, including the α_0 and α_π data obtained in this measurement. The fitting procedure was applied to the same database of Ref. [262], with the addition of the integrated cross section data coming from the present work for the α_π channel in the $E_{cm} \simeq 1.07 - 1.24$ MeV region, and for the

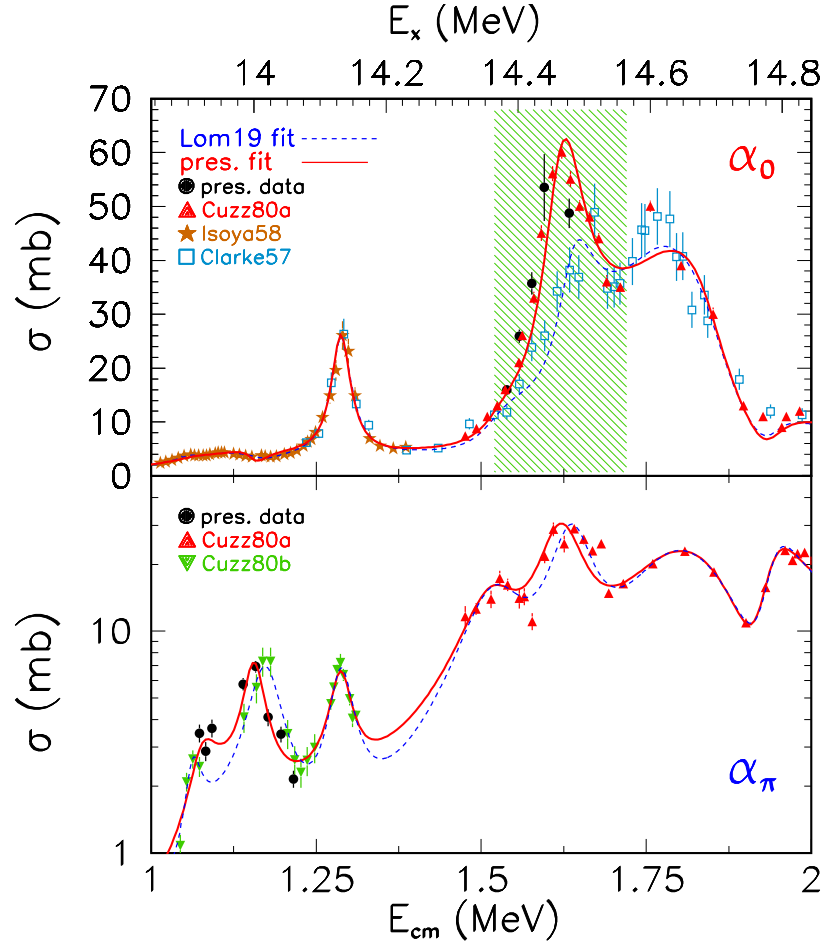


FIGURE 5.10: R -matrix fit to present data (red solid lines), together with other data previously reported in the literature. (*Upper panel*) Integrated cross section for the α_0 reaction channel. Black dots: present data. Literature data, as normalized in Ref. [261], are displayed as follows: red upwards triangles are data from Ref. [75]; brown stars are data from Ref. [146]; light blue boxes are data from Ref. [263]. Blue dashed line: R -matrix fit from Ref. [262]. Inside the green hatched box, only present data and data from Ref. [75] were included in the fit. (*Lower panel*) Integrated cross section for the α_π reaction channel. Green downwards triangles refer to data from Ref. [271], as reported in Ref. [262]. Blue dashed line: R -matrix fit from Ref. [262].

α_0 channel in the $E_{cm} \simeq 1.54 - 1.62$ MeV range. Furthermore, in the $E_{cm} \simeq 1.54 - 1.72$ MeV region (green hatched area in Figure 5.10, upper panel), the α_0 integrated cross section data by [75] were included in the fit, considering their coherence with the present measurements, while the data from Ref. [263] were excluded. The starting resonance parameters (E_x , total and partial width values) adopted in the fit were taken from Ref. [262]. J^π values were fixed to the ones of Ref. [262], that were in good agreement with the systematic reported in Ref. [65].

The results of the new R -matrix fit are shown in Figure 5.10 as red solid lines; blue dashed lines reports the previous fits of Ref. [262]. Only results for the α_0 and α_π integrated cross section for $E_{cm} = 1 - 2$ MeV are displayed, while the fits to the other included data (Ref. [262] database) give nearly the same results of Ref. [262] and are not shown for simplicity.

From the present R -matrix fit, it is possible to recognize a small shift in the energy of the peaks seen at $E_{cm} \simeq 1.07 - 1.15$ MeV in the α_π integrated cross section and, mainly, a strong change in the shape and amplitude of the large peak seen in the α_0 cross section at $E_{cm} \simeq 1.6$ MeV. Table 5.1 shows the new parameters (excitation energies, total and partial widths) obtained from the present fit for states in the excitation energy region $E_x \approx 13.8 - 14.7$ MeV; such values are compared with the corresponding ones reported in Ref. [262].

In general, the positions of excited states agree with the literature values within about 20 keV, and the total and partial widths are in most cases in agreement with the previous values within $\approx 10\%$, essentially confirming the results reported in Ref. [262]. Some interesting deviations are seen for the 13.93 MeV state, only involved in the α_0 and p_0 channels, for which the Γ_{α_0} partial width is $\approx 40\%$ larger than the previously reported values.

Concerning the 13.92 MeV 2^+ state, the new data of the α_π channel lead to a sizable change in the total and partial widths; the total width is almost doubled, reaching a value (53.4 keV) very close to the one reported in the compilation of Ref. [65] (48 keV); the Γ_{α_π} partial width is approximately two times the previously reported value of Ref. [262]. The Γ_{p_0} value is also a factor $\simeq 2$ larger than the one of Ref. [262], going towards a better agreement to the old value reported by Isoya (57 keV, Ref. [146]). Differences are also seen in the partial widths of the 14.02 MeV 1^- state, which shows a $\frac{\Gamma_{\alpha_\pi}}{\Gamma_{\alpha_0}}$ branching ratio close to unity; a similar branching was also reported by Ref. [146].

The most interesting information comes out for the 0^+ state at $E_x \simeq 14.47$ MeV. Ref. [76] indicates this state as the head for the [211] quartet band; Ref. [75] instead suggested the 14.92 MeV 0^+ state as the [211] band-head, assigning a possible [220] structure to the 14.47 MeV one. On the other hand, Ref. [248] indicates that this state could be the iso-analogue of the 4.315 MeV, $T = 1$, 0^+ state in ^{20}F , which does not show a quartet configuration.

In Figure 5.10 one can observe that this state is responsible for the pronounced peaks seen at $E_{cm} \simeq 1.61$ MeV in the integrated cross sections of both reaction channels: the conflict between data of [263] and [75] has therefore prevented a solid spectroscopic analysis. The present reduced widths, reported in Table 5.1, show a small p_0 branching ratio, $\frac{\Gamma_{p_0}}{\Gamma} \simeq 0.14$, in contrast with the very large value $\frac{\Gamma_{p_0}}{\Gamma} \simeq 0.79$ reported in Ref. [248]. The occurrence of a $\frac{\Gamma_{p_0}}{\Gamma}$ branching ratio much smaller than 0.79 was also reported in a recent fit involving only elastic scattering data (Ref. [268], $\frac{\Gamma_{p_0}}{\Gamma} \simeq 0.40$). The present Γ_{p_0} value leads to a spectroscopic factor ≈ 4 -5 times smaller than the one experimentally observed for the 4.315 MeV state in ^{20}F [272, 273], in disagreement with the hypothesis of an iso-analogue state.

Furthermore, as discussed in Refs. [75, 76], the presence of reduced partial widths $\gamma_{\alpha_\pi}^2 > \gamma_{\alpha_0}^2$ could point out the occurrence of a quartet state in ^{20}Ne of the [211] type. In this framework, it was firstly suggested in [76] that the 14.47 MeV state could have a quartet structure, because of their estimate of reduced partial widths: $\frac{\gamma_{\alpha_\pi}^2}{\gamma_{\alpha_0}^2} \approx 1.6$. In the present analysis, a smaller value of the ratio of the partial widths was found $\left(\frac{\gamma_{\alpha_\pi}^2}{\gamma_{\alpha_0}^2} \approx 1\right)$, not supporting the quartet structure previously suggested.

At variance, the present analysis and Ref. [262] indicate the possible [211] quartet nature of the 14.92 MeV state, for which the dimensionless reduced partial width of

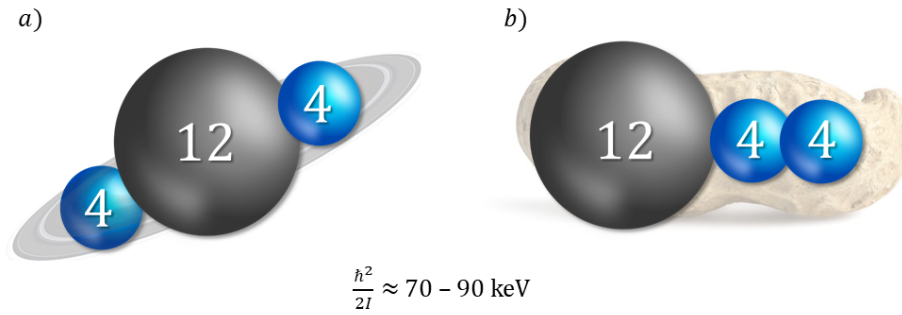


FIGURE 5.11: Pictorial representation of the two limiting configurations for the $^{12}\text{C} + 2\alpha$ cluster structure of the ^{20}Ne nucleus, resembling *a)* a planet like Saturn and *b)* a peanut.

the α_π channel is dominant over the p_0 and α_0 channels, in agreement with the old conclusions of Ref. [75].

To explain why a large reduced partial width on the α_π decay channel would signal a quartet state in ^{20}Ne , Figure 5.12 reports the lowermost possible quartet configurations. From a comparison with the quartet configurations in the daughter nucleus ^{16}O , shown in Figure 1.41, it is evident that a [211] configuration in ^{20}Ne can easily transform into a [210] configuration in ^{16}O by emitting the loose α particle sitting in the fp shell. Since the [210] configuration in ^{16}O fully characterizes the $\alpha + ^{12}\text{C}$ cluster structure of the 6.05 MeV 0^+ state, the presence of a large dimensionless reduced partial width of the α_π channel is a fingerprint of the existence of such quartet state at high excitation energies.

This finding suggests some interesting, even if simplified, speculations. The [211] quartet state should have, in fact, a pronounced $^{12}\text{C} + 2\alpha$ cluster structure [73, 75]. If the clusters are assumed as homogeneous spheres touching at their surfaces, it is possible to (roughly) estimate, with classical calculations based on the Steiner theorem, the inertial parameter of such configuration as $\frac{\hbar^2}{2I} \approx 70\text{-}90 \text{ keV}$, depending on the limiting cases assumed for the $^{12}\text{C} + 2\alpha$ structures as sketched in Figure 5.11.

Therefore, a possible 2^+ rotational excitation of the 14.92 MeV quartet state should be located at $E_x \approx 15.3 - 15.4 \text{ MeV}$. Indeed, both the present work and Ref. [262] reported the presence of 2^+ states in this energy window. In this context, the state at 15.419 MeV is a promising candidate: its reduced partial width for the α_π channel is well larger than the one for the α_0 ($\gamma_{\alpha_\pi}^2 \simeq 10 \text{ keV}$, $\gamma_{\alpha_0}^2 \simeq 2.7 \text{ keV}$), and both are much larger than the one for the p_0 channel (few tenths of keV), a pattern quite similar to the one observed for the 14.92 MeV state. These speculations strongly call for further experiments, including the ones studying the elusive $^8\text{Be} + ^{12}\text{C}$ cluster decay in ^{20}Ne [274].

5.2 Conclusions

This Chapter reported results on a new experiment aiming to study the $^{19}\text{F}(p, \alpha)^{16}\text{O}$ reaction at low bombarding energies, using very high-quality beams delivered by a Singletron accelerator, and state-of-the-art solid state detectors. This allowed to clearly separate the contributions coming from the ^{16}O doublet at 6.049-6.13 MeV. Particle yields were transformed into absolute differential cross sections by means of an accurate

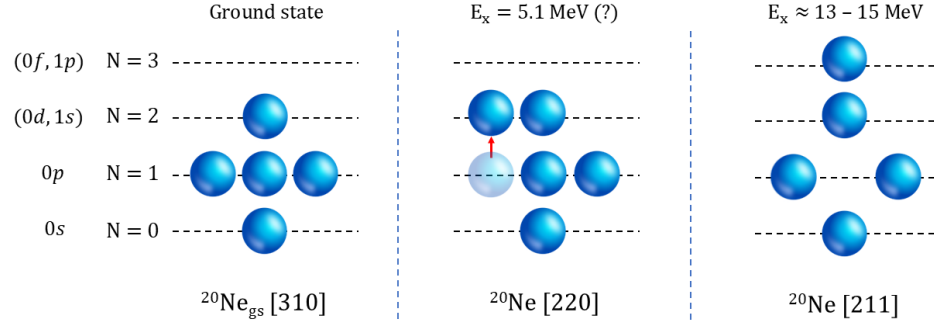


FIGURE 5.12: Quartet model scheme for the structures of the ground state (*left*), the [220] state predicted at 5.1 MeV (*middle*) and the [211] state at $E_x \approx 13 - 15 \text{ MeV}$. The Reader is also referred to Figure 1.41 for analogous quartet configurations in the daughter nucleus, ^{16}O . The [220] configuration, theoretically predicted to be at 5.1 MeV, was indeed suggested [74] to characterize the 7.20 MeV 0^+ state in ^{20}Ne .

J^π	$E_x^{[prev.]}$	E_x	$\Gamma_{cm}^{[prev.]}$	Γ_{cm}	$\Gamma_{\alpha_0}^{[prev.]}$	Γ_{α_0}	$\Gamma_{\alpha_\pi}^{[prev.]}$	Γ_{α_π}	$\Gamma_{p_0}^{[prev.]}$	Γ_{p_0}
...	...	...	...	...	...	...	...	...	...	...
1^-	13.89	13.89	293	174	162	157	17	16.4	447 eV	537 eV
2^+	13.910	13.927	29	53.4	8	12.7	21	40.6	21 eV	39 eV
0^+	13.912	13.932	251	351	251	351	0	0	381 eV	563 eV
1^-	14.02	13.992	61	49	6	23.5	55	25.4	189 eV	140 eV
2^+	14.131	14.131	39	36.5	33	32	6	4.1	405 eV	368 eV
1^-	14.351	14.352	151	127	42	25	108	101	1.0 keV	877 eV
0^+	14.466	14.450	96	99	64	69.5	25	16.2	7.0 keV	13.5 keV
1^-	14.596	14.596	287	315	212	230	65	75.5	9.8 keV	9.6 keV
0^+	14.653	14.653	160	148	143	133	11	8.1	6.5 keV	6.9 keV
...	...	...	...	...	...	...	...	...	...	...
2^+	15.419	15.419	60	60	29	29	29	29	63 eV	63 eV
...	...	...	...	...	...	...	...	...	...	...

TABLE 5.1: Spectroscopic parameters for states in ^{20}Ne determined from the present R -matrix fit, contributing in the energy region explored in the present experiment, i.e. $E_{cm} \simeq 1.0\text{-}1.7 \text{ MeV}$. Parameters for the 15.419 MeV state are also included because they are recalled in Sect. 4. Present values are compared with the previous ones from Ref. [262]. Excitation energy values E_x are given in MeV. The total (Γ_{cm}) width and α partial widths (Γ_{α_0} , Γ_{α_π}) are given in keV, while the proton partial widths Γ_{p_0} are expressed in the units indicated in the text.

internal normalization procedure based on the use of bench-marked elastic scattering data. Differential cross sections were then integrated in the whole angular domain by considering the trends of angular distributions reported in the literature.

This experiment had two main goals: to provide new data in the poorly known energy region of $E_{cm} \approx 1.1$ MeV for the α_π channel, and to investigate the discrepancy still persisting in the literature between the data of Refs. [75] and [263] in the α_0 reaction channel. Concerning the last point, the present data clearly agree with the ones of Ref. [75]; a possible explanation for the existing discrepancy between the datasets [75, 263] is also outlined.

The availability of new cross section data allows to refine the R -matrix fit of several reaction channels reported in Ref. [262]. As a result, nuclear structure information on natural-parity states in ^{20}Ne at $E_x \approx 13.9$ -14.7 MeV was updated. In particular, the contrasting interpretations for the structure of the 14.47 MeV 0^+ state were disentangled, and a quartet structure for the 14.92 MeV 0^+ appears to be supported by the data here obtained. Qualitative considerations suggest that the 2^+ state at 15.42 MeV, reported in [262], could represent a rotational excitation of the quartet state at 14.92 MeV. If this hypothesis were true, the 14.92 MeV 0^+ state would be characterized by a very large deformation, given by its possible $^{12}\text{C}+2\alpha$ structure. Further works, both from the experimental and theoretical side, can be very useful to shed light on this suggestion and to unveil the cluster nature of highly-lying states in the self-conjugate ^{20}Ne nucleus. Furthermore, among the possible new experiments that can be planned in the future, it seems particular intriguing to investigate the practically unknown excitation functions and angular distributions of the $^{19}\text{F}(p, ^8\text{Be})^{12}\text{C}$ reaction at low energies, allowing to have a direct access to a decay channel in ^{20}Ne characterized by a very exotic cluster decomposition.

Chapter 6

Clustering of polar molecules with hot atom chemistry

Up to now, the attention has been mainly focused on nuclear physics phenomena, whose description was often supported by models and techniques proper of molecular physical chemistry. These were, in fact, exploited to discuss clustering phenomena, since these appear to be so similar to what happens in molecular systems, aside from the scales of the assemblies and of the interactions. In this Chapter, nuclear physics will *return the favor* to molecular physical chemistry: its outstretched hand will in fact offer a raffinate nuclear mechanism to produce fast atoms. These, moving with high speeds inside their solution, may indeed generate interesting molecular-breaking phenomena, which may in turn be sensitive to the presence and the magnitude of some particular intermolecular interactions.

6.1 Hot atom chemistry: nuclear tools for molecular physics

From a chemical point of view, the behavior of the different isotopes of a certain element should be absolutely identical, given that the chemical properties of an atom are fundamentally dependent on its external electrons and not on the composition of the nucleus. For this reason, isotopes of various elements are nowadays widely used for a great range of applications, from isotopic dating, to food treatments and to the development of isotopic tracers and radio-therapies: the main point of strength of these applications is, indeed, the fact that the different isotopes are chemically and biologically indistinguishable, but perfectly recognizable with the right nuclear apparatuses.

However, some exceptions are always hidden in wider generalizations. The behavior of an atom could actually be influenced by unpredictable events happening in its *radioactive* nucleus, getting it ready for some *bizarre adventures* and behaviors, justified by the large¹ quantity of energy stored and released by nuclear phenomena. These effects are, indeed, some of the most beautiful things that may help to build a bridge between chemistry and nuclear physics.

One example of this interconnection is represented by the *Szilárd-Chalmers* effect, which consists in the separation of a radioactive isotope of a given atom from a parent molecule,

¹Large with respect to the usual atomic and molecular realm, i.e. from meV to some eV.

irradiated with thermal neutrons. The separation is the result of the recoil that the heavy atom experiences upon its de-excitation, generally proceeding through the emission of one or more energetic γ rays of several MeV [152, 275]. This effect, discovered by L. Szilárd and T. A. Chalmers in 1934 [149], proved itself very useful for the separation, preparation and enrichment of radioactive isotopes of various metallic and non-metallic elements [276, 277].

The first example, which led Szilárd and Chalmers to the discovery of the homonymous effect, concerned the irradiation of Ethyl Iodide with a neutron source: the ^{127}I nucleus of the molecule has a quite high capture cross section ($\sigma_{th} = 6.2$ barns [278]) for thermal neutrons. This makes it likely to capture a neutron emitted from the source, transforming into an excited, radioactive ^{128}I atom. The energy of the thermal neutron ($E_{th} \approx 0.025$ eV) is not enough to break any of the chemical bonds in the molecule (having energies of 2 – 4 eV). Nevertheless, following the neutron capture, the ^{128}I atom will be excited at an energy close to 6.28 MeV, which is the separation energy (S_n) of a neutron in the ^{128}I nucleus [62]. Considering, for simplicity, the de-excitation through the emission of a single γ ray, the subsequent recoil energy of the ^{128}I atom would be of around 160 eV, way higher than the C-I binding energy [150]². The ^{128}I atom is therefore no more bound to the Ethyl Iodide molecule, and moves freely in the solution; as a single $^{128}\text{I}^*$ atom, it is now chemically separable with respect to the target ^{127}I atoms bound in the Ethyl Iodide molecule. Upon extraction in water by a reducing agent and subsequent precipitation with Silver Nitrate (AgNO_3), it is possible to form the insoluble Silver Iodide (Ag^{128}I) salt, highly enriched with the radioactive isotope. A sketch of the phases involved in the Szilárd-Chalmers process was introduced in Section 2.4, Figure 2.9. Recent examples of the use of the Szilárd-Chalmers process in a large variety of radiochemical experiments are described, for example, in Refs. [279–286].

From the simple schematic picture described, one can expect that all the produced radioactive atoms can be extracted from the original matrix (often an *organic liquid*). However, it was early shown that a sizable fraction of such atoms is *retained* inside the matrix (*retention* effect), limiting the achievable enrichment. A pioneering model trying to explain this phenomenon was based on the work by Franck and Rabinowitch [153], and considered the formation of a *cage* of atoms and radicals able to entrap the radioactive atom, as discussed in Section 2.4.

Even if a large body of data was collected during the '30-'60s of the past century, it is still difficult to fully ascertain the sequence of reaction mechanisms that the fast-recoiling radioactive atom (*hot atom*) induces on the atoms of the solution, both before and after its thermalization [287]. Two very different theories have been proposed in the literature to explain the fate of the hot atom: the *billiard ball + epithermal* model by Libby [150, 288, 289] and the *random fragmentation* model by Willard [151]. Both models succeeded in explaining a part of the observed phenomenology, but are still inadequate to describe the entire process. Furthermore, the possible occurrence of direct *bimolecular substitutions* of the hot atom in a molecule of the original matrix has been also proposed as a possible mechanism to explain the retention [287]. The situation is made even more puzzling when considering that thermalized X^* radioactive atoms, during their diffusion into the surrounding matrix, can collide against a molecule containing a non-radioactive atom of the same element X . This could result, with a non-vanishing probability, in an *exchange* process that would enhance the retention [290–292]. This already intricate

²However, even in presence of a cascade involving three or four γ rays with similar energies, energies above 80 eV can be expected [151]

picture is further complicated by the effects due to the *dilution* of the target material with another compound. As an example, a different behavior in the retention mechanism was reported by using different organic solvents (e.g. benzene or cyclohexane solvents diluting a carbon tetrachloride target, in [293]), in contrast with the simple hypothesis of a lowering of the retention because of the unraveling of the *cage* due to dilution. Other sizable effects on the fraction of retained materials are observed when *scavenger* materials are added to the target solution prior to the neutron bombardment [152, 275, 276]; a typical example is the use of I_2 as scavenger in Ethyl-Iodide solutions. Also in this case, further complications arise from the dependence of the thermal exchange rate on the inverse of the I_2 concentration in the solution [292], giving rise to quick exchange processes at very low molecular Iodine concentrations, that complicate the description of the retention mechanisms. It is worth noting that dissolved Iodine would also act as a *carrier* for radioactive ^{128}I atoms, leading to an additional reaction mechanism in the already complex picture described above.

It is therefore not surprising that several physical and chemical aspects of the retention in the Szilárd-Chalmers effects are still mysterious and worth to be explored with new experiments. In this work, in particular, the attention was focused on the role played by the dilution of the Ethyl-Iodide target in the retention of ^{128}I radioactivity, with different concentrations of Ethanol. This investigation takes inspiration from the previous work reported in Ref. [294], where the retention of ^{128}I radioactivity in the organic phase was studied as a function of three different Ethanol concentrations (0%, 1% and 50% mol), and a non-monotonic trend was reported. Differently from Ref. [294], the irradiation was performed in the presence of a very low γ -ray field, strongly reducing the formation of Iodine by radiolysis of the target material [295].

The results of these investigations are discussed in the following Sections, where the effects of dilution on the retention phenomena of radioactive atoms produced in the Szilárd-Chalmers reaction are explored. After describing the experimental procedures adopted to quantitatively estimate the yield of free radioiodine produced by the neutron irradiation of different Ethyl Iodide-Ethanol solutions with a minimum level of gamma radiation, the non-monotonic behavior of the obtained results is discussed: this clearly shows the presence of an optimum Ethanol concentration able to maximize the radioisotope yield. Aside from the technical interest due to a "carrier-free" procedure that optimizes the activity concentration of radioiodine, some tentative suggestions on the *causes* leading to the observed trend of data could be linked to the onset of inter-molecular clustering phenomena, due to halogen-hydrogen bonds (see Section 2.4) and leading to a modification of the cage effect.

6.2 The *radiochemistry* experiment

The *radiochemistry* experiment performed at the INFN-LNS of Catania, and described in the next Sections, was a truly unconventional, beautiful and demanding experience involving several scientific aspects (from organic chemistry to electronics, from molecular physics to nuclear physics, from engineering to radio-protection). The difficulties encountered along the route towards the experiment, and overcome thanks to a strong team game, represented a very formative testbed for a series of possible subsequent investigations on a research field, the nuclear chemistry, that is nowadays not too much widespread in Italy and Europe.

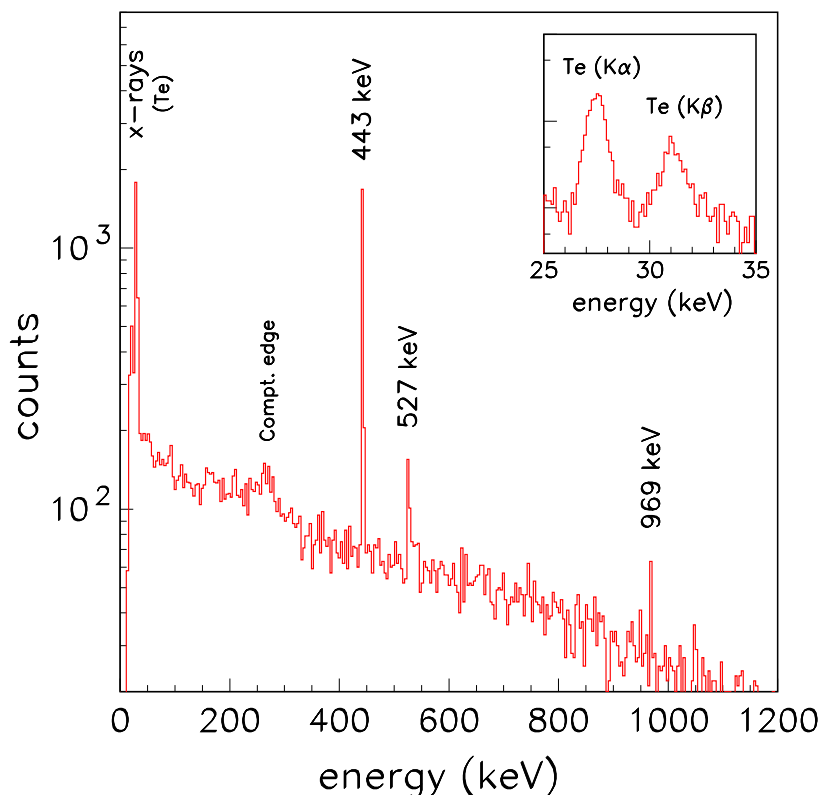


FIGURE 6.1: Gamma-ray spectrum of the irradiated sample as detected with a shielded HPGe station with thin entrance window. In the inset, the part of the spectrum due to the characteristic X-rays of ^{128}Te produced by electron capture of ^{128}I is shown

6.2.1 Preparation of the solutions and thermal neutrons irradiation

Six different samples were prepared and irradiated in the same geometrical and temporal conditions. The reagents were transparent Ethyl Iodide (Merck (Germany), stabilized with elemental silver) in a grade suitable for research purposes, and absolute Ethanol in analytical grade, ACS reagent. The samples were handled, from the storing to the extraction phase, in UV-light proof vessels (polypropylene) and environments, to avoid the photolysis of the molecules, leading to uncontrolled formation of molecular Iodine in the solutions. Gas-chromatographic analyses, made by the supplier, indicate the possible presence of Ethanol at a level smaller than 1% (a/a); other physical properties of the Ethyl Iodide (as density and refraction index) agree, within few ‰, with the standard values reported in the literature for pure Ethyl Iodide. The Ethyl Iodide was filtered with grade 42 ashless filter paper to minimize the possible traces of silver used as stabilizer. The properties of the sample solutions, prepared with different concentrations of Ethyl Iodide and Ethanol in 100 mL total volume, are summarized in Table 6.1.

All the solutions were irradiated for 90 minutes ($\simeq 3.6 t_{1/2}$ of the radioactive ^{128}I), under the same neutron flux conditions, with a $\simeq 33$ GBq Am-Be source. The samples were irradiated and treated one after the other, with a 30 minutes time shift to allow the separation and counting procedures. The irradiation was performed at INFN – Laboratori Nazionali del Sud (Catania, Italy). Polyethylene blocks were interposed between the source and the irradiated sample to thermalize the fast neutrons. With the adopted geometry, the total neutron flux at the irradiated sample spot was $\Phi_n \approx 1.75 \cdot 10^3$

Sample ID	vol(mL)	Et-I(mol%)	Et-OH(mol%)
I	100	100	0
II	100	99	1
III	100	95	5
IV	100	90	10
V	100	85	15
VI	100	50	50
VII	200	95	5

TABLE 6.1: Ethyl Iodide : Ethanol concentration ratios for the seven different samples used in the present experiment

cm^2s^{-1} . The fraction of thermal-to-fast neutrons was estimated by detailed Monte-Carlo simulations of the irradiation chamber volume and was about 2.4; epithermal neutrons yields were well smaller than thermal and fast neutrons ones. The γ -ray equivalent dose at the irradiation point was ≈ 2.4 mSv/h, a factor 10^{-3} – 10^{-4} smaller than the ones obtainable by using Ra-Be sources [294, 295]. The very low gamma dose would reduce the production of Iodine atoms (carriers) by radiolysis, allowing to genuinely highlight the effects due to Ethanol. All the samples were extracted after 15 ± 1 minutes from the exit from the irradiation chamber; this would allow a temporal comparison with the retention data from [294].

In a second experiment, performed about two hours after the end of the previously described operations, the irradiation of a seventh sample containing 200 mL of Ethyl Iodide (95 mol%) and Ethanol (5 mol%), was performed in identical conditions. 50 mL of this solution were then picked up four times from the bottle at different post-irradiation times (from 15 to 39 minutes after the exit from the irradiation chamber), in order to investigate the possible occurrence of exchange and/or recombination effects of *thermal* radioiodine diffusing in the solution [291, 292, 294].

Some control γ -ray spectra of the irradiated solutions and extracted radioiodine samples were obtained by using a shielded HPGe detection station; an example is reported in Figure 6.1. All the observed gamma and X-ray lines, also the fainter ones, are fully compatible with the decay of ^{128}I ; lines related to the activation of unwanted contaminants are absent or below the detection limit.

6.2.2 Liquid-Liquid extraction process

Each sample was treated through a multi-step extraction process [276, 296] in a separator funnel, inside a glove-box chamber. The liquid-liquid extraction was reiterated for two times with 100 mL of aqueous sodium thiosulphate solution ($\text{Na}_2\text{S}_2\text{O}_3$, 10% w/w) [294], shaken for 2 minutes and left to rest for 1 other minute each time; such operations were conducted as fast as possible because of the short half-life of ^{128}I (≈ 25 min). After the first extraction, the organic phase was collected and re-extracted with the same amount of the water solution, while the aqueous phase was stored for the successive activity measurement, to be merged with the aqueous fraction obtained from the second extraction. For the four sub-samples coming from the seventh sample, used for the study

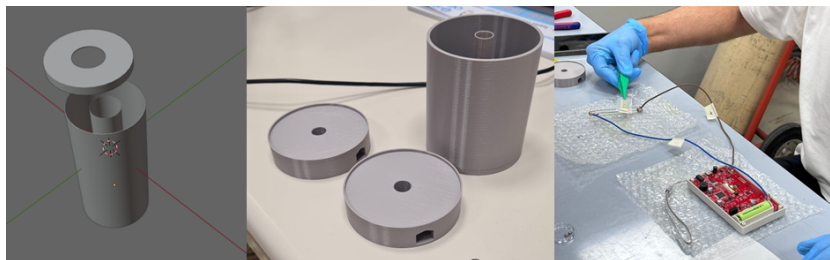


FIGURE 6.2: *Left*: 3D render design of the jacketed Geiger counters for liquids; *Middle*: the 3D-printed vessels, hosting the Geiger tube; *Right*: a phase of the assembly of the electronics and the tube inside the vessels.

of the retention as a function of the post-irradiation time, the duration of the (single) extraction procedure was halved; again, this expedient was needed because of the short lifetime of ^{128}I .

6.2.3 Detection system and activity measurements

The activities of the organic and the water phases were measured through two identical, custom-made jacketed Geiger-Muller counters for liquids (see Figure 6.2 and, e.g., [297]); the tubes were J321 Geiger tubes with an operation voltage of 400 V. The respective counts were monitored using a software which allowed to obtain a temporal series of the count rates through the whole measurement time (≈ 7 min for each one of the first six investigated samples) and to correct for the (negligibly small) dead time effect. The response of the two Geiger detectors was tested with a ^{137}Cs source, which emits a γ ray of energy (662 keV) similar to the ones of ^{128}I (443 and 527 keV); the observed count rates at the two detectors for several fixed geometries of irradiation were fully consistent (within the random fluctuations). The two measurements for the organic and aqueous phases were started at the same time instant. In the adopted experimental configurations, an almost total self-absorption of β -rays in the liquids and the vessels walls of detectors is expected by calculations exploiting the commonly used absorption formulas [297]. Self-absorption of γ rays in the different liquids (water, Ethanol, Ethyl Iodide and their solutions) was also calculated (starting from the data of [298]) to estimate the detection efficiency of the γ rays. The reported count rates were then corrected accordingly, even if in these experimental conditions the efficiencies are quite similar for both the detectors in the whole range of dilutions. The activity ratio between the aqueous and organic phases was then calculated, for all irradiated samples, from the observed count rates (background subtracted) registered by the detectors: R_{wat}^{net} and R_{org}^{net} . Dedicated background runs were performed for both counters before the measurement of every sample; the background count rates (≈ 20 cpm) were the same for the two detectors within the random fluctuations. The yield Y_{free} , and the fraction f_{free} of extracted free ^{128}I in the water solution were then calculated through the following formulas:

$$Y_{free} = \frac{R_{wat}^{net}}{R_{org}^{net}} \quad (6.1)$$

$$f_{free} = [1 + Y_{free}^{-1}]^{-1} \quad (6.2)$$

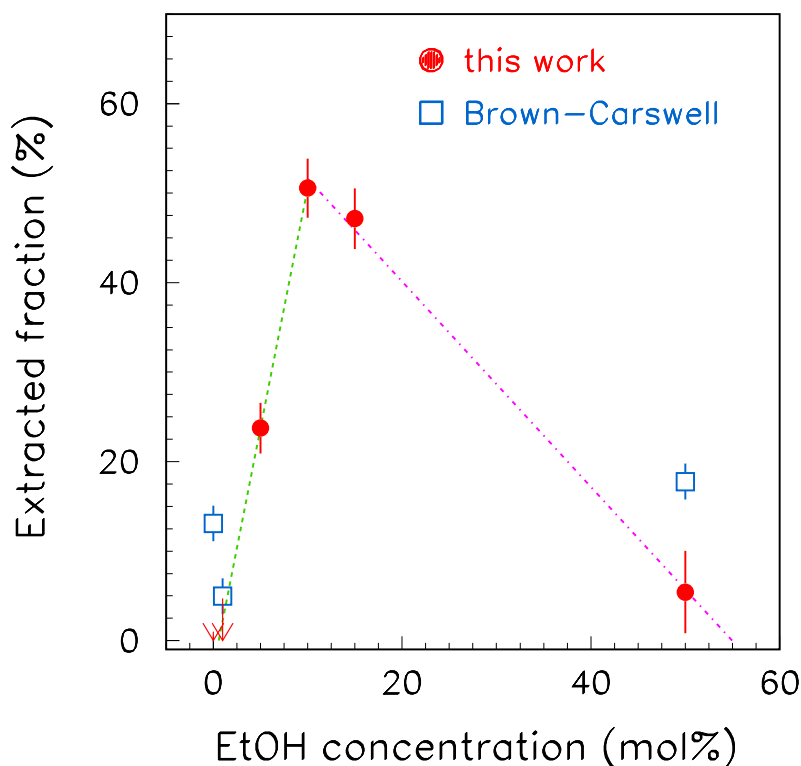


FIGURE 6.3: Effect of the Ethanol concentration (in mol%) on the extracted fraction f_{free} of ^{128}I . Red dots (or red arrows) indicate data (or upper limits of the extracted fraction) obtained from the present experiment, while data from Ref. [294], taken at a similar post-irradiation time, are reported as blue open squares. The dashed and dotted lines are used to guide the eye.

The uncertainties were calculated through statistical error propagation formulas, considering uncertainties on the activity and background measurements for both the organic and the water phases. When the fraction f_{free} turned out to be compatible with zero, its upper limit was estimated by using the statistical approach by Feldman and Cousins [299].

6.3 Extracted fraction of Radio-Iodine and retention

The extracted fraction (f_{free}) data obtained from the activity measurements in the present experiment are reported in Figure 6.3 as red dots with error bars; upper limits are reported as red arrows. In this Figure, data from Ref. [294] at a nearly identical post-irradiation time, have been included for three different Ethanol concentrations: 0, 1, and 50 mol% (blue open squares).

Some differences between the two data set can be easily recognized. For example, for pure Ethyl Iodide samples, the present data indicate a very large retention of the ^{128}I in the organic phase, while in the case of Ref. [294], a fraction $f_{free} \approx 13\%$ was obtained. This

difference could be explained by considering the strong enhancement of Ethyl Iodide-Iodine exchange processes triggered by very low concentrations of stable Iodine atoms [292, 300]. The fraction of Iodine atoms in the solution is indeed enhanced by radiolysis under the more intense γ -ray flux characterizing the Ra-Be source used in Ref. [294]. Free Iodine atoms could also act as a carrier to form ^{128}I - ^{127}I molecules available for subsequent aqueous extraction. In the present case, given the extremely faint γ dose, the presence of Iodine atoms due to radiolysis is extremely low: therefore, according to Ref. [292], exchange processes are expected to be largely enhanced, increasing the retention. Further, a smaller concentration of stable Iodine carriers would hinder the formation of ^{128}I - ^{127}I molecules. As a result of these processes, the ^{128}I atoms can be easily captured by radicals formed by the damping of kinetic energy in the recoiling nucleus [151]. In agreement with the present observations, vanishingly small f_{free} data were also reported by Alimarin and Svoboda [295] by using Po-Be sources with a very low γ dose.

Interestingly, despite the large uncertainty, the *difference* between f_{free} values observed at 50 mol% EtOH concentration and for pure Ethyl Iodide (5.4 %), is quite similar to that obtained in Ref. [294] ($\simeq 4.7\%$). In this light, the two findings seem coherent: once the exchange processes induced by the tiny fraction of Iodine produced by radiolysis are taken into account, and after having waited for a long post-irradiation time, the dilution effect due to large EtOH concentrations has only a residual influence in enhancing the free radionuclide yield; at variance, as reported in Ref. [294], the effect of large quantities of EtOH is very useful to enhance the f_{free} for very short post-irradiation times.

The most striking aspect of the experimental retention curve reported in Figure 6.3 is due to the presence of an optimum EtOH concentration (≈ 10 mol%) at which the free radio-halide yield shows a pronounced maximum ($f_{free} = 51\% \pm 3\%$). No similar effects seem to have been reported to date; the checks made on the detection systems would exclude the occurrence of some insidious systematic effect. Furthermore, a very similar f_{free} fraction was obtained by performing an independent subsequent experiment, with different Ethyl Iodide and Ethanol batches, where the same optimum EtOH concentration (10 mol%) was used. In the last case, the extraction of the radioiodine was performed via precipitation of AgI from the aqueous solution treated with KI and AgNO_3 [296]; γ radiation was detected by a shielded HPGe detector both for the whole irradiated sample at the exit of the irradiation chamber and for the AgI precipitate, allowing to estimate the f_{free} value (even with a large relative error, of $\approx 20\%$).

An explanation of the observed maximum in the f_{free} curve is difficult, surely because of the competition between several effects in the retention of the radioiodine, occurring at different time scales [152, 287, 301]. In this framework, it can be useful to monitor the organic retention as a function of the *post-irradiation* time: in the presence of *thermal exchange* effects with a long mean life (e.g., of the same order of the radioiodine half-life), a sizable variation, with time, of the organic retention should be clearly observed. These measurements were performed by using the seventh sample, and the obtained results are shown in Figure 6.4 for the Y_{free} value defined in Eq. 6.1. The four measurements are reported as open dots with error bars, and compared with the result obtained in the first part of the experiment (black star). The four points are compatible each other within the errors and their weighted average is fully compatible with the black star. The observed flat trend would therefore indicate that, in the present experiment, the mean life of the thermal exchange is surely well smaller than a few minutes. The concentration of I_2 molecules produced by radiolysis can be estimated, in a first approximation, by using the yields reported in Ref. [302] giving, in the present case, $[\text{I}_2] \approx 1.8 \cdot 10^{-9} \text{M}$. By

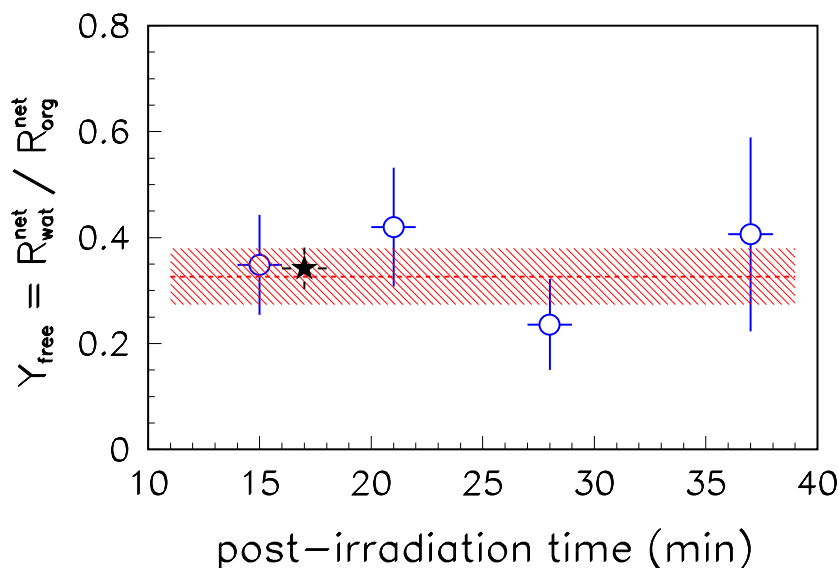


FIGURE 6.4: Trend of the $Y_{\text{free}} = R_{\text{wat}}^{\text{net}} / R_{\text{org}}^{\text{net}}$ radioiodine yield as a function of the post-irradiation time. Blue open dots: data from the seventh sample, made by a 95 mol% Ethyl Iodide + 5 mol% Ethanol solution. The dashed line indicates the weighted average of data, while the red band indicates the 1σ confidence region. The black star is the Y_{free} value obtained in the first part of the experiment for a solution with the same dilution. The horizontal position of the star was shifted forward by two minutes for clarity reasons.

using the second-order kinetics proposed by [292], it is possible to roughly estimate an half-time for the Iodine exchange of the order of ≈ 10 s, in qualitative agreement with the present findings and the ones of Ref. [303].

Since kinetics considerations are in agreement with previous theories and observations, the search for a possible explanation of the observed optimum of the yield is stimulated in a different direction. It is in principle possible that some unexpected contaminants in the reagents or in the vessels could have played a role in the peculiar rise and fall trend observed in the f_{free} data, and the present findings surely call for further independent investigations on such interesting aspects. In this framework, few repeated measurements were performed at small Ethanol concentrations, using different batches of materials, different vessels, different quantities of irradiated samples, and extraction techniques, leading to results similar to the ones discussed here within $\approx 20\%$. It is therefore possible that the mechanism leading to the observed data trend could have a more profound origin.

6.4 Possible modification of cage effects due to molecular clustering

To explain the curious findings observed in the previous Section, a possible interpretation of retention data is proposed, based on peculiarities that can occur in the *inter-molecular* binding of the molecules forming the reaction target (see Section 2.4): they could in fact cause a substantial change of the topology and the strength of the molecular *cage*

surrounding the slowed-down radioactive Iodine atom, leading to a modification in the observed retention.

It has been recently discussed in Ref. [22], as recalled in Section 2.4, that Methyl-Iodide diluted in Ethanol gives rise to the formation of trimeric molecular clusters because of the onset of *hydrogen-halogen* bonding. This phenomenon is mainly triggered by the peculiar charge distribution of the Iodine atom inside an alkyl-Iodide molecule: the presence of a σ -hole and of a pronounced *negative belt* of the charge distribution [304] would cooperatively lead to an attraction of the oxygen (δ^-) of an Ethanol molecule from the σ -hole side, and an attraction of a hydrogen (δ^+) of *another* Ethanol molecule from the negative belt side. Such spatial re-organization of the molecules in trimeric aggregates starts to be effective only in presence of *large* Ethanol excess ($> 30 - 40\%$) with respect to the *Methyl*-Iodide [22].

Interestingly, this finding seems to be qualitatively correlated with the present observations on the f_{free} curve: at small Ethanol fractions, the inter-molecular interaction in the solution would be essentially *binary* in nature, leading to cages with simple topologies and reduced strengths that could be easily unraveled by increasing the dilution. But when the solution would overcome a critical range of Ethanol fraction, the onset of cooperative effects leading to EtI-EtOH-EtI trimeric aggregates would change the properties of the molecular cage, leading to a large trapping probability of the produced radioiodine (both by exchange mechanisms or by recombination with surrounding radicals).

To support the above hypothesis, it would be important to perform fully quantum mechanical simulations of the structure and dynamics of the Ethyl-Iodide diluted with Ethanol: the charge displacements in Ethyl-Iodide can in fact be a bit different from the Methyl-Iodide molecule used in Ref. [22]. This aspect will be investigated in the future by performing detailed molecular dynamics simulations, analogue to the ones of Ref. [22]. Anyway, it is worth noting that simplified semi-classical electrostatic calculations made by using known dipole moments [305] and bond lengths [306, 307] of Methyl-Iodide and Ethyl-Iodide molecules would indicate a change of just $\approx \pm_{15\%}^{6\%}$ in the overall negative charge excess in the Iodine region when adding a further Ethyl group to the molecule. The two different values come from two different assumptions on the δ^+ localization in the molecule: fully on the carbon of the CH_3- group (-15%), or *shared* between the two carbons with a ratio inversely proportional to the distance from the iodine atom ($+6\%$). In both cases, it seems quite unlikely that such small relative variations of the charge excess would strongly modify the onset of halogen-hydrogen cooperative in Ethyl Iodide + Ethanol solutions if compared with Methyl Iodide + Ethanol ones. Furthermore, from the experimental chemistry side, this hypothesis stimulates to perform new Raman blue-shift analyses, of the same type of the ones in Ref. [22], also on Ethyl Iodide + Ethanol solutions with the same concentrations used in the present experiments.

If confirmed, these effect could represent an interesting cause for reflection to better understand, from a physical chemistry base, the effect played by the chemical bond on retention in Szilárd-Chalmers reactions, and could be extended, with further experiments, also to different combinations of Alkyl-Iodides and Alcohols.

6.4.1 *Epilogue: the radio-Iodine separation as radioactive AgI*

The last step of the series of radiochemistry measurements performed was linked to the *physical* separation of the radioactive isotope from the solution, which is done through



FIGURE 6.5: Experimental set-up inside the glove box in the "α laboratory" of radio-chemistry at INFN-LNS, Catania.

precipitation under the form of an insoluble salt, inside the controlled work environment reported in Figure 6.5. After the solvent extraction, which ends with the reduction of the Iodine atoms (^{128}I) to *Iodide ions* ($^{128}\text{I}^-$) under the action of the reducing agent $\text{Na}_2\text{S}_2\text{O}_3$, the aqueous phase is treated with silver nitrate (AgNO_3) and potassium Iodide (KI). The result is the immediate formation of silver Iodide, AgI , enriched in ^{128}I . A heating system and a pump for filtration under vacuum are actually useful to speed up the process: in the end, the radioactive Ag^{128}I is obtained under the form of a yellow powder deposited on the filter paper, as shown in Figure 6.6. Activity measurements performed with the HPGe detector confirmed that almost all the free radioiodine present in the solution was precipitated in the filter paper, underlining a very efficient way to extract radioactive material from the production matrix with a good level of enrichment.

6.5 Conclusions

This Chapter investigated some aspects of the retention of ^{128}I in liquid solutions of Ethyl-Iodide + Ethanol subject to the Szilárd-Chalmers reaction under thermal neutron bombardment. The irradiation was performed in a very low γ -ray field to minimize radiolysis production of unwanted Iodine atoms. The observed fraction of extracted Iodine f_{free} as a function of Ethanol concentration shows: (i) a threshold concentration (~ 1 mol%) under which the addition of Ethanol is ineffective in increasing the f_{free} ; (ii) a region where the increase of the Ethanol concentration corresponds to an increase of f_{free} , until an optimum value $f_{\text{free}} \simeq 51\%$ is reached; (iii) a region of large Ethanol dilution (up to 50 mol%) where the increase of the Ethanol concentration induces a drop of the observed f_{free} value. While the findings obtained in (i) and (ii) can be explained, respectively, by: (i) considering the catalytic effect of Ethanol in Iodine-Ethyl Iodide exchange effects reported by [291, 294] and (ii) invoking *dilution* effects on the radical cage formed by the hot atom, the findings at the item (iii) seem to be quite surprising and difficult to be explained within the currently adopted phenomenology on organic retention.



FIGURE 6.6: Radioiodine sample (in the form of yellow Ag^{128}I powder) as deposited on black filter paper at the end of the precipitation and of the whole extraction procedure.

Despite the care that was put in the handling of the reagents and the vessels used in the experiment, and the presence of some repeated measurements, it is still not possible to completely exclude the hypothesis that the drop of f_{free} with the Ethanol concentration could be attributed to some obscure effects of an unwanted contaminant. If this is not the case, an alternative and intriguing suggestion to explain the present findings could come from a potential modification of the cage effect due to the formation of trimeric molecular aggregations in the solution.

All the findings reported here call for further experimental investigations and underline, once more, the complexity of the physical and chemical mechanisms at the basis of the Szilárd-Chalmers process.

Conclusions

*Yes, I will be thy priest, and build a fane
In some untrodden region of my mind,
Where branched thoughts, new grown with pleasant pain,
Instead of pines shall murmur in the wind:
Far, far around shall those dark-cluster'd trees
Fledge the wild-ridg'd mountains, steep by steep;
And there by zephyrs, streams, and birds, and bees
The moss-lain Dryads shall be lull'd to sleep...*
J. KEATS, *Ode to Psyche*, 50-57

It is now the time to draw some conclusions about the work of this three-years PhD project, which has been presented in the pages of this thesis. Its character was surely as polyhedral as the investigated subjects, not focusing on a single reaction, nucleus or experiment, but rather on a series of experimental and theoretical investigations, both on the chemistry and physics field, and all the possible ways to correlate and intertwine them. Despite all the efforts and the dedication, every result is not actually marking an arrival point: it is instead a wonderful source of question marks, paving the road to new experimental and theoretical challenges necessary to address all the questions, both the new ones and those which still remain unsolved.

Let's take the case of the ^{12}C as a first example: the crystallographic approach has produced some interesting results, but its real *potential* is hindered by the quality and the type of the available experimental data. The possible usage of higher quality data, with larger angular covering, smaller angular steps and lower experimental uncertainty may help in the investigation inside the most sensitive regions highlighted in Figure 3.9. Similarly, parallel experiments could be performed exploiting *neutron* beams, since these are charge-less particles which would not resent from the Coulomb repulsion generated by the protons in the ^{12}C , strongly simplifying the treatment and the parameters to be tuned in the model. The general lack of data, together with the higher complexity of the scattering problem and of the structure itself, is also preventing a solid *crystallographic* analyses in the following self-conjugate nuclei, i.e., ^{16}O and ^{20}Ne . The results and the drawbacks of the oxygen case have already been presented in Section 4, but, to date, the method has not been applied to the neon nucleus; under the correct experimental circumstances and theoretical approximations (for example, about the different α distances in the trigonal by-pyramid structure), this could be an useful approach to highlight some clustering effects in the ground state of the ^{20}Ne nucleus.

About the investigation on the Hoyle state, it was successful to shed light on the recent discrepancies which arose about the determination of the Hoyle state radiative decay branching ratio, which have a huge impact both on nuclear astrophysics questions and

in the cluster description of its nuclear structure. In the same fashion, it could be interesting to intensively point the efforts towards a similar measurement of the Γ_{rad} value for the so-called *Efimov* state or to the 3^- state at 9.64 MeV. An experiment dedicated to the latter state would indeed be similar to the one reported in this thesis, but *a hundred times* harder: in fact, according to Tsumura et al. [188], the radiative decay branching ratio of this state is supposed to be of the order of 10^{-6} .

On the ^{16}O side, it would be interesting to analyze the presence of particular states whose branching ratios (as in Figure 4.11) may result higher than the calculated penetrability limits, hinting for the occurrence of possible cluster configurations. Other interesting aspects may be hidden in the study of the $d + ^{14}\text{N}$ reaction at lower energies, although this would be a more complex field, given the necessity to discriminate between the useful compound nucleus contributions and the contamination from the direct processes, which are always a danger when using deuteron beams.

To deepen the knowledge on the ^{20}Ne spectroscopy and the characteristics of some of its excited states, $p + ^{19}\text{F} \rightarrow ^8\text{Be} + ^{12}\text{C}$ reactions may be really interesting: the detection of the ^8Be nucleus as $\alpha - \alpha$ coincidence with the ^{12}C recoil may be useful to study and determine the partial decay width in this channel. This could allow to underline the existence of cluster states with pronounced $^{12}\text{C} + \alpha + \alpha$ configurations, with the *planetary* and *peanut*-like structures reported in Figure 5.11.

From the point of view of the radiochemistry experiments, the one reported in Chapter 6 is nothing but a starting point for a series of similar experiments and procedures, aiming to describe the Szilard-Chalmers reaction and hot atom processes in the frameworks of different families of alkyl halides (or similar) and alcohols, or with any other types of compounds suitable for the Szilard-Chalmers process. With the right prescriptions, even the most difficult measurements (e.g., those exploiting methanol instead of ethanol as a scavenger) may be performed, to unveil the influence of inter-molecular clustering effects on the topologies of cage effects and, consequently, on radionuclide retention.

What is reported in the appendixes is a further proof that, in any case, it is actually difficult to disentangle the chemistry from a nuclear physics experiment, or the physics from a chemical procedure. Chemical approaches and theories have proven useful not only for the "simple" realization of targets used in nuclear physics experiments, but also (and a lot, as reported in this thesis) in the description of phenomena intrinsically pertaining to the nuclear physics world, such as the occurrence and the regularities of cluster configurations in light self-conjugate nuclei. These are the reasons for which these two worlds, contrary to what often happens and to some dull cliches, should not be seen as distant or separated, if not for the energies and the distance scales put into play by the system. Even if, also in this case, some connections are still found...

Appendix A

Ion beam modification of organic layers by deuteron bombardment

Commonly, when performing fundamental or applied nuclear physics studies, a number of problems arise about the stoichiometric and/or areal density stability of the exploited targets, making these quantities important sources of uncertainty in the absolute estimation of cross sections. In connection with the work of this thesis, it was possible to accurately characterize the properties and the modifications observed in thin Melamine layers deposited on carbon backings, and subject to high power densities delivered by a low energy deuteron beam. The nitrogen content and the processes of its expulsion from the organic molecules have been investigated as a function of the fluence of the beam and of the flux of particles, in order to unveil possible properties related to the roughness of target surfaces.

A.1 Surface modifications due to swift ion beams

When swift particles of an ion beam hit the surface of a solid material, they can lead to ejection of material by sputtering or desorption (see, e.g. [308] and references therein). In insulating materials, the erosion by ion bombardment has also been connected with the formation of track damage [309], as a result of ion-induced electronic excitations [310]; the mechanism of energy transfer from the electronic degrees of freedom to the atomic motion leading to the displacement (or even to the ejection) of atoms from the insulator are still only partially understood [310, 311]. It is worth noting that below a *critical* thickness of insulating material, the ion-induced sputtering yields dramatically drop out, and the origin of this effect is still not well understood [311]. Furthermore, when the insulators are hydrocarbons, the C-H bond breaking due to the electronic excitation can lead to a strong rearrangement of the bonds of the bombarded atoms leading, for example, to the formation of new molecules (as H₂) that can be diffused out from the material [312, 313]. All these structural modifications due to the ion-induced sputtering or desorption of bombarded material, or more complex chemical modifications when dealing with polymeric layers [314], must be taken into account. In fact, they can have a sizable impact both in Ion Beam Analysis (IBA) protocols for the characterization of coatings and thin films and also in nuclear spectroscopy experiments, where accurate absolute reaction cross section measurements are needed.

As an example, Elastic Recoil Detection Analysis (ERDA) of hydrogen implanted in facing materials (typically graphite or beryllium) of fusion reactors has been proposed as a powerful tool to trace the particle balance in fusion reactors [315, 316], but provided that the de-trapping effects during ERDA are adequately taken into account [316, 317].

While processes leading to hydrogen desorption or emission from thin metallic or polymeric layers have been widely studied in the past, much fewer data have been reported on the emission of nitrogen atoms due to ion beam bombardment. For example, Ref. [318] investigated the desorption of Nitrogen atoms previously absorbed in thin amorphous carbon via the analysis of secondary electrons emitted in heavy ion collisions ([319, 320]). Interestingly, it was noted in Ref. [321] that the irradiation of polyamide ($\text{C}_{10}\text{H}_2\text{N}_2\text{O}_4$) layers with heavy ions at hundreds of keV did not lead to sizable nitrogen emission from the bombarded sample, while an overall density enhancement of the irradiated sample was observed.

Layers of organic or inorganic compounds containing nitrogen atoms have been widely used in the past for several purposes. Evaporated, deposited or implanted on a substrate (typically, thin carbon foils or other metals, see e.g. [322–324]), they have been often used as targets in nuclear reaction experiments involving ^{14}N or ^{15}N isotopes (see, e.g., [192, 193, 325–327]). Thick targets of sintered boron nitride were recently used as production target for the radiologically-important ^{11}C and ^{13}N positron-emitting radionuclides [328, 329]. Very thin silicon nitride targets were prepared with dedicated radio-frequency reactive sputtering techniques [330], and were used, for example, for a high precision experiment on the structure of the proton rich ^{16}F with low energy nuclear reactions [331], or for cross section measurements in ion beam analysis applications [332].

Indeed, ion beam analysis techniques have been widely used in the past to determine the quantity of nitrogen present in metals, semiconductors, polymers or biological samples (for a review, see, e.g. [333]). In this framework, changes of stoichiometry and/or nitrogen ejection under ionic bombardment must be considered in quantitative estimates, and the limited number of data on the destruction and/or desorption yields due to ion bombardment can represent a limit in quantitative analyses. A similar problem affects the absolute cross section determination in nuclear reactions on nitrogen-bearing targets; for example, it was early noted that the nitrogen quantity present in Melamine targets subject to deuteron bombardment was subject to a sizable reduction even with moderately low currents [192]. It appears that detailed measurements of the degradation of the nitrogen content in several organic compound under ionic bombardment are still fragmentary, thus preventing to have the full control of one of the potential source of systematic errors.

The present work discusses the results of a long-lasting experimental campaign aimed at measuring the evolution with time of the nitrogen content in Melamine layers evaporated on several carbon substrates with different morphological properties. These measurements were collateral to the experiment aimed to probe the properties of excited states in the ^{12}C nucleus [327] and reported in Chapter 3 of this thesis. The trend of data clearly indicates the occurrence of two effects: a *prompt* nitrogen emission, occurring at a quite low irradiation fluence, and a (much more *gradual*) *slow* emission, leading to a saturation scenario. Interestingly, the measured destruction cross section, leading to the release of nitrogen atoms from the target, seems correlated to the *morphology* of the carbon substrate, suggesting a possible role of surface irregularities in the re-capture of the emitted atoms. Furthermore, the prompt emission is larger at higher particle fluxes;

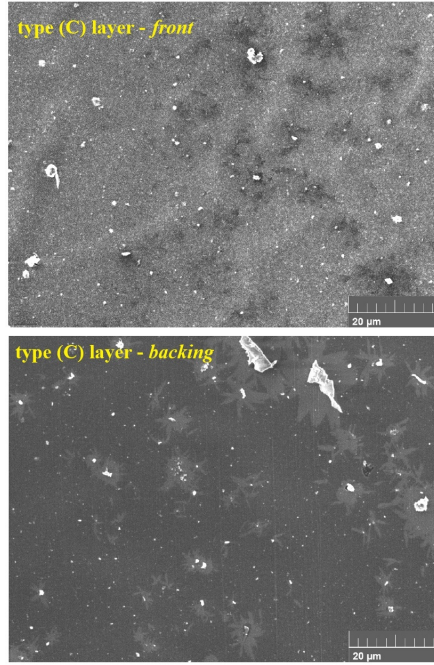


FIGURE A.1: Scanning Electron Microscope (SEM) pictures of a type (C) sample before the ionic bombardment. The SEM image of the (top) Melamine layer of the sample is shown in the upper panel. The SEM image of the carbon backing is shown in the lower panel.

the data also indicate the occurrence of a threshold value ($\simeq 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$), below which prompt emission becomes vanishingly small.

A.2 Experimental setup, sample properties and data taking

The experiment was performed by using deuterons beams of 2.7 MeV produced at the CN accelerator of the INFN - Laboratori Nazionali di Legnaro, Italy. Deuterons bombarded two series of samples made by a Melamine layer ($50 \pm 2 \mu\text{g}/\text{cm}^2$ areal density, determined by energy loss measurements) evaporated on thin carbon backings (having several thicknesses, ranging from 5 to $10 \mu\text{g}/\text{cm}^2$). Each series of samples was based onto two different types of backing: the first one, manufactured in Catania (Italy) and then indicated in the text as "type (C) layer", was obtained by floating thin carbon layers produced by the Yissum Research Development Company; the second one, manufactured in Legnaro (Italy) and then indicated in the text as "type (L) layer", was home-made by thermal evaporation of graphite rods, leading to a deposit on suitable glasses treated with a release agent, and then floated. Melamine layers were deposited, in both cases, by Physical Vapor Deposition (PVD) techniques, starting from the same sample of Melamine powder (Aldrich, $> 99\%$).

The two types of used backings show very different roughness, as obtained from dedicated measurements with a Scanning Electron Microscope (SEM). Figures A.1 and A.2 display two SEM pictures of the Melamine layer deposited on the carbon backings (upper panels)

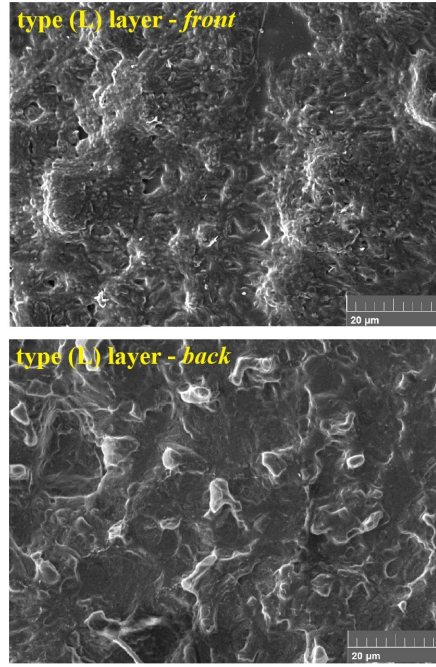


FIGURE A.2: Scanning Electron Microscope (SEM) pictures of a type (L) sample before the ionic bombardment. The SEM image of the (top) Melamine layer of the sample is shown in the upper panel. The SEM image of the *back side* of the target (where no Melamine was evaporated) is shown in the bottom panel.

and of the backing itself (lower panels). In the case of the type (C) sample, a nude backing was used to better appreciate the smoothness of the surface.

From Figure A.1 (lower panel) the very smooth behavior of the carbon layer is evident; according to the SEM, the maximum irregularities in the surface have a typical length well smaller than $2\ \mu\text{m}$. Consequently, the Melamine deposition appears in turn very homogeneous at the micrometer scale (upper panel). A completely different trend is observed for the type (L) samples (Figure A.2): a much more pronounced roughness is seen both in the front and the back sides of the sample, with irregular inhomogeneities having typical length of $\approx 4\ \mu\text{m}$. These two extremely different features would allow to investigate the effect of the roughness on the destruction of the organic material under ionic bombardment.

In this framework, a large variety of samples of both species were exposed to very long deuteron irradiation, by using the experimental setup sketched in the insert of Figure A.3. Deuteron beams delivered by the accelerator were driven inside a scattering chamber operated in vacuum (residual pressure of the order of 10^{-6} mbar); the beam was tightly shaped by a series of diaphragms and collimators to assure a beam spot on the target of $\simeq 1$ mm diameter, with an almost uniform particle flux distribution. The beam current was monitored by a custom-designed electrostatically-suppressed Faraday cup, followed by a pico-ammeter with a precision better than 1%. During each irradiation shift, the beam current was stable within 5% maximum; this figure can be adopted as maximum non-statistical uncertainty affecting the beam current evaluation. The beam current was varied in the ≈ 25 -55 nA range; considering a beam spot of 1 mm diameter and the energy loss in the sample, it is possible to estimate a power density deposition on the target of $\mathcal{I} \approx 150\ \text{W}/\text{m}^2$ for 10 nA beam current. The sample holder was made

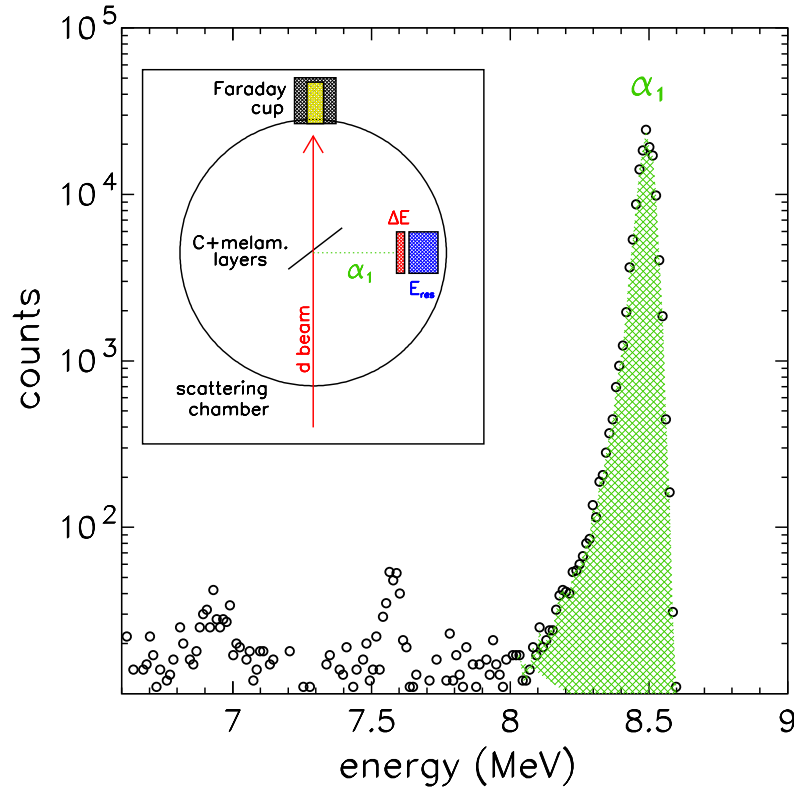


FIGURE A.3: Example of a spectrum taken during the bombardment of Melamine+carbon layers with deuterons of 2.7 MeV, showing the part of spectrum close to the $^{14}\text{N}(\text{d},\alpha_1)^{12}\text{C}$ reaction, leading to the population of the 4.44 MeV state in the ^{12}C residual nucleus (*green hatched area*). The insert reports the schematic layout of the experimental setup used to perform the present measurements.

by copper, and it was cooled in vacuum by a forced circulation of a liquid kept at -2°C . The relative angle between the beam and the surface of samples was of $\simeq 45^\circ$.

To trace the evolution of the nitrogen content in the samples, the signal coming from the $^{14}\text{N}(\text{d},\alpha_1)^{12}\text{C}$ reaction was exploited, as suggested in Ref. [332]. The significance of the α_1 notation is the same as defined in Section 3.2. This work takes into account data from a portion of the more complex setup described in Ref. [327], constituted by a solid-state telescope, made by a $93\text{ }\mu\text{m}$ ΔE detector, followed by a $500\text{ }\mu\text{m}$ E_{res} stage. The detector system was collimated by a circular diaphragm and subtended a solid angle of $\simeq 0.71\text{ msr}$.

The front-end electronic, based on NIM modules and VME-ADC of the FAIR acquisition system (with a dead time smaller than 1%), was built to operate the telescope in anti-coincidence mode, allowing to discard the interfering peaks due to high energy protons emitted in (d,p) reactions on nitrogen and other elements present in the samples. An example of energy spectrum obtained during the bombardment is reported in Figure A.3. The region of interest where the α_1 transition is expected is indicated as a green hatched area; the data exhibit a very low background (smaller than 0.4%) and a good energy resolution ($\simeq 60\text{ keV}$).

Five experimental runs with (C)-type samples and eight experimental runs with (L)-type samples were performed. The elapsed irradiation time was varied run per run to have

the possibility of following, in the most detailed way, the temporal evolution of nitrogen content in the bombarded layer. Typical elapsed times goes from a few tens of minutes up to several hundreds of minutes, depending on the type of the layer.

A.3 Nitrogen emission under deuteron irradiation

When Melamine layers are irradiated with swift ions, nitrogen atoms can be copiously emitted; electronic excitation can in fact lead to the breaking of the C-N amine bond in the molecule and the ejection of nitrogen. In fact, C-N bonds (≈ 66 kcal/mol) are well weaker than C-H paraffin and olefin bonds (≈ 99 kcal/mol), and the emission of nitrogen is expected to dominate the molecular destruction processes. To trace the *remaining* nitrogen content in Melamine layers subject to irradiation, the exploited analytical probe was the rate of reaction products (hereafter "*the N probe*", in particle per second, *pps*) falling in the α_1 energy region (green region of interest) of the spectrum in Figure A.3, as a function of the beam fluence ϕ hitting the target. The time sequence used to investigate the fluence dependence of the remaining nitrogen in the layer was sampled with time bins varying as a function of the elapsed time: typically, they were smaller (≈ 5 minutes) at the beginning of the sequence, becoming larger (≈ 10 minutes or more) at the end.

Figure A.4 reports two examples of fluence dependence of the N probe for a (C)- and a (L)-type Melamine layers; similar trends are observed for all the other studied layers. It is evident, in both cases, the exponential-like reduction of the N probe in the first part of the graphs; then, the data remain quite flat, pointing towards a sort of saturation value of the N probe at large fluence values. The different behavior of the two types of Melamine layer can be gathered at a first sight by noting the much slower decay of the N probe for the (L)-type Melamine layers, that are characterized by substrates with larger roughness (see Figure A.2). To quantitatively estimate some relevant properties observed for the N probe, $N(\phi)$, the experimental data were fitted with the following function:

$$N(\phi) = N_{\infty} + (N_0 - N_{\infty}) \cdot e^{-\sigma_{dest}\phi} \quad (\text{A.1})$$

that represents an exponentially-decaying behavior ($e^{-\sigma_{dest}\phi}$) summed with a saturation value N_{∞} . Here, ϕ indicates the beam fluence on the target, while σ_{dest} is the *destruction cross section*, that is linked to the microscopic probability of breaking the Melamine molecule inducing the emission of nitrogen atoms from the layer. The N_0 and N_{∞} parameters are respectively linked to the N probe values observed at a *vanishing fluence* and at a very large fluence, and their trend will be analyzed in the following Section.

Equation A.1 nicely reproduces the data in the whole fluence range here explored, as evident in Figure A.4; a similar agreement is observed also for the other samples here investigated. It is worth noting that the same functional form has been widely used in the previous literature to describe the surface layer destruction both on implanted or coating materials [308] and to describe the yield of secondary electrons associated to the surface erosion due to ion beam bombardment [320].

From the data fitting, it is possible to extract some relevant properties affecting the nitrogen content in the Melamine layers, such as the destruction cross section σ_{dest} and the N probe values at zero and very large fluence, N_0, N_{∞} . In estimating such quantities and their errors from the fit, the covariances of parameters were took into account.

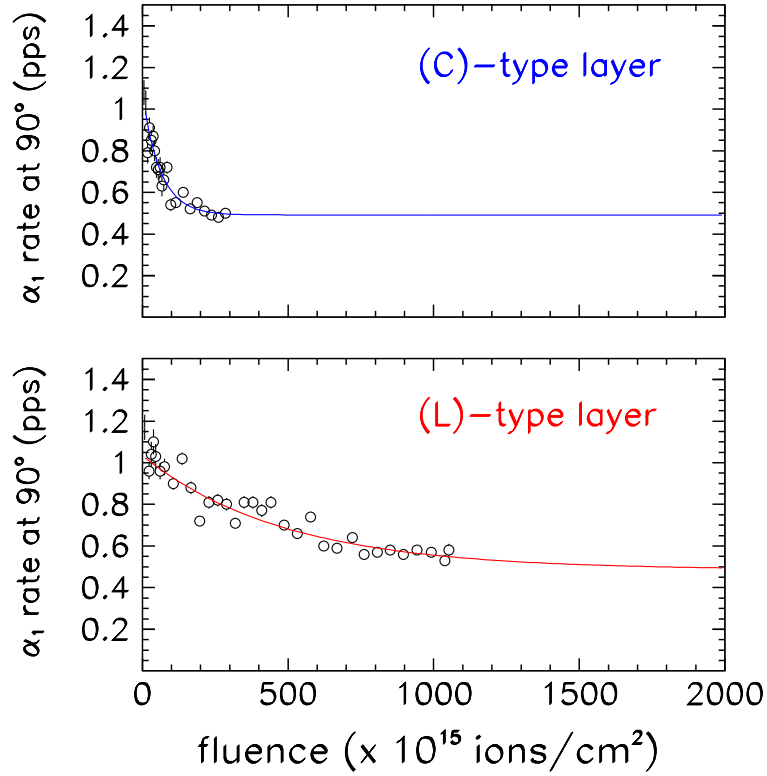


FIGURE A.4: Rates of particles (particle per second) detected at 90° in the region of interest of the α_1 transition (the N probe, $N(\phi)$, green area of Figure A.3), as a function of the beam fluence. *Upper panel*: an example of $N(\phi)$ evolution with the beam fluence for the irradiation of a (C)-type layer. *Lower panel*: an example of $N(\phi)$ evolution with the beam fluence for the irradiation of a (L)-type layer. Solid lines indicated the fit of data with a functional form given in Equation 1.

For example, the destruction cross section σ_{dest} seems correlated to the roughness of the carbon substrate used as a backing for the Melamine. In particular, for the smoother (C)-type layer of Figure A.4 (upper panel) $\sigma_{dest} = 1.6(2) \cdot 10^{-17} \text{ cm}^2$, a value almost one order of magnitude higher than the one of (L)-type layer (lower panel), $\sigma_{dest} = 0.21(2) \cdot 10^{-17} \text{ cm}^2$.

The marked difference of destruction cross sections in the two different types of layers is observed also for the other investigated samples. In particular, for the very smooth and almost uniform (C)-type samples, the σ_{dest} values are concentrated in the region $[1.2 - 2.2] \cdot 10^{-17} \text{ cm}^2$. This behavior can be appreciated by plotting the σ_{dest} values as frequency histogram (with a large binning due to the limited data samples); for data coming from (C)-type layers, the σ_{dest} values are reported as a blue line-hatched histogram in Figure A.5. It is worth noting that the systematics of nitrogen desorption cross sections derived in Ref. [318] as a function of different bombarding ions having different specific energy losses, would suggest a value of $\sigma_{desorp.} \approx 1.5 \cdot 10^{-17} \text{ cm}^2$, quite close to the presently obtained destruction cross section. This finding seems to indicate the common underlying nature of the two phenomena (destruction of nitrogen from a molecule and desorption of nitrogen atoms adsorbed inside a material), connected to the electronic energy loss in the material [310, 311].

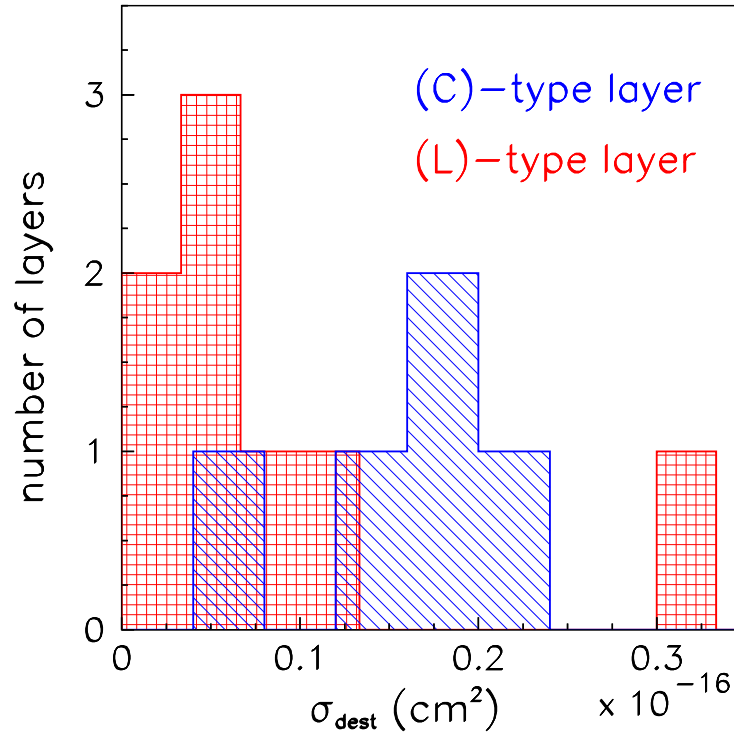


FIGURE A.5: Distribution of the nitrogen destruction cross sections σ_{dest} obtained from the fit of data related to (C)-type and (L)-type layers, indicated respectively as blue, line-hatched and red, square-hatched histograms.

A quite different behavior is instead observed for the (L)-type layers (red gridded histogram in Figure A.5); the distribution of σ_{dest} values is a bit broader, and its center is approximately five times smaller than the one characterizing the (C)-type layers. It is worth noting that, for both (C)- and (L)-type layers, the σ_{dest} values obtained do *not correlate* to the thickness of the used carbon backing ($R^2 \simeq 0.07$) or to the impact point of the beam on target ($R^2 \simeq 0.13$). The origin of this effect, that makes the (C)-type layers less suited to withstand longer irradiation periods, should be therefore traced elsewhere, and in particular to the roughness of the substrate.

The SEM pictures shown in Figures A.1,A.2 clearly indicate the different roughness of the carbon substrates and, consequently, of the Melamine coating. In a simple scenario, it is possible to hypothesize that the large roughness ($\approx 4 \mu\text{m}$ typical scale, in the frontal plane of the layer) of the (L)-type substrate could favor oblique ejection of the material (as suggested in Ref. [334]) and the consequent re-trapping of nitrogen atoms inside the Melamine layer modified by the ion beam bombardment, possibly forming new C-N bonds inside the material. It is also possible, as indicated in Ref. [334], that a pronounced irregular topology of the surface could favor the re-trapping of the ejected nitrogen atoms.

The present results stimulate to perform new analyses on Melamine layers covering substrates of different, and controlled, roughness; in this respect, for example, thin palladium layers could be used as suitable backings [335, 336] to carefully determine the dependence of the σ_{dest} from the surface roughness.

A.4 Properties of the *prompt* and *slow* components in Nitrogen emission

The fit of the nitrogen signals, coming from the α_1 spectra, allows to determine, run-per-run, the extrapolation at zero and very large fluence of the rate of observed α_1 particles due to the presence of remaining nitrogen in the bombarded layer (N_0 and N_∞ respectively). If one normalizes such quantities for the average mean current i_{med} of each run, it is possible to obtain normalized particle rates r_0^{norm} and r_∞^{norm} that, for a *fixed* value of the areal density of nitrogen atoms in the layer, would depend only on the *geometry* of the experimental setup and on the differential cross section (DCS) of the nuclear reaction $^{14}\text{N}(\text{d},\alpha_1)^{12}\text{C}$ at 2.7 MeV bombarding energy and $\theta_{lab} = 90^\circ$ (as discussed before, dead time corrections are vanishingly small). In particular, adopting the DCS value of ≈ 4.4 mb/sr by averaging the data reported in Refs. [228, 337] and considering the solid angle subtended by the detection system, it is possible to estimate a normalized particle rate $r^{norm} = 0.039(4)$ pps/nA for fresh Melamine layers of $50 \mu\text{g}/\text{cm}^2$ areal density. The uncertainty on r^{norm} takes into account the uncertainty on the DCS reported in the literature and the ones related to the geometry of the setup. Obviously, in absence of any degradation effects, the normalized values r_0^{norm} and r_∞^{norm} should be equal, within the uncertainties, to r_{max} . In general, this is indeed not the case for most of the samples analyzed in this work.

Figure A.6, reports the r_0^{norm} (upper panel) and r_∞^{norm} (lower panel) values obtained from the data fitting as a function of one of the most dominant quantities involved in molecular destruction of insulating layers, i.e. the particle flux impinging on the target. The red solid lines with hatched areas represent the $r^{norm} = 0.039(4)$ pps/nA limiting value expected for a fresh layer. The r_∞^{norm} (lower panel) values decrease as a function of the particle flux, quite irrespective of the different type of samples; this behavior could be qualitatively explained by considering, for example, the larger number of overlapping ion tracks that are cumulatively formed during the bombardment, leading to a larger depletion of nitrogen (even on the re-trapped atoms).

It is more intriguing to discuss the peculiar trend affecting the r_0^{norm} (upper panel): in principle, all the values should fall within the hatched area representing the $r^{norm}=0.039(4)$ limiting value, but this is indeed not the case for several layers investigated here. It is interesting to note that, regardless the type of layer, the r_0^{norm} values decrease with increasing particle flux, being very often well lower than the limiting value. This effect can be explained by considering the presence of a very fast emission mechanism (*prompt emission*) occurring in the early phases of the irradiation (within few minutes from the start, corresponding, in the present experimental conditions, to fluence values typically smaller than 10^{15} cm^{-2}). In this respect, it is worth noting that prompt ejection of hydrogen from polymeric materials subject to swift ion bombardment has been already reported in the literature [338], co-existing with the (most common) slow component. Following the suggestions of [338], prompt emission could be attributed to the simultaneous breaking of bonds in the organic molecules triggered directly by the shower of colliding ions.

The occurrence of prompt nitrogen emission from an organic sample would represent a very critical point both for quantitative analysis of organic material based on ion beam analysis, and for high precision measurements of absolute cross sections in nuclear physics. While it is possible to correct data *a posteriori* for the nitrogen ejected during

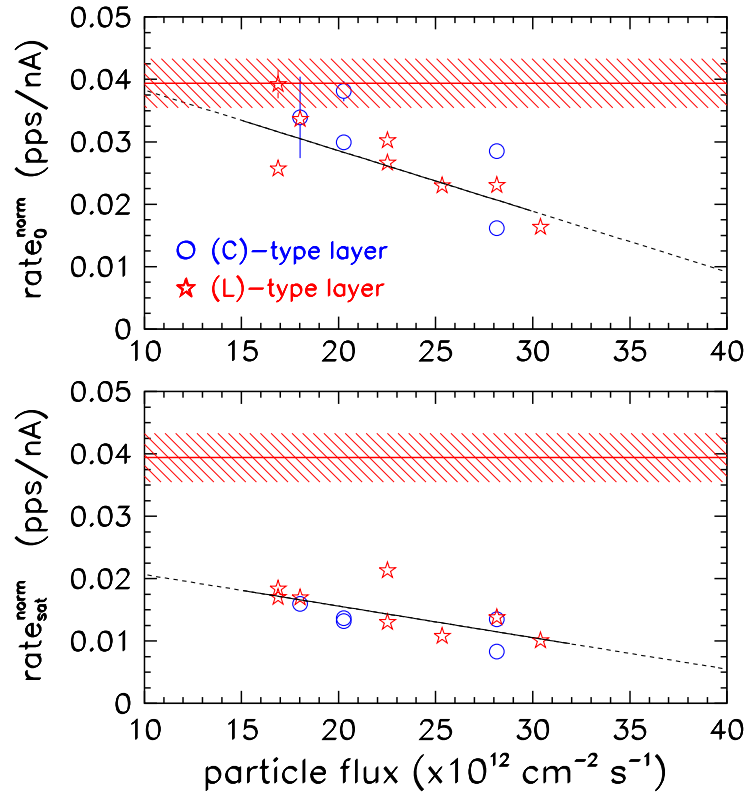


FIGURE A.6: Normalized initial rate r_0^{norm} (upper panel) and normalized saturation rate r_{∞}^{norm} (lower panel) of particles coming from α_1 transition as a function of the average beam particle flux. Blue circles: (C)-type samples. Red stars: (L)-type samples. The red line with hatched band represent the r^{norm} limiting value deduced from the present experimental geometry and by using the differential cross section for the $^{14}\text{N}(d,\alpha_1)^{12}\text{C}$ reaction reported in Refs. [228, 337]. Black solid lines, with dashed extrapolations, are the results of unweighted fits of all the data, including both (C)- and (L)-type samples.

the slow emission phase by using suitable functional form (as, e.g., the one of Equation A.1, the prompt emission is much more difficult to be corrected and can irreversibly bias the data. However, from the present data it is possible to derive the *boundaries* of particle flux at which the prompt component becomes vanishingly small, allowing to minimize an insidious source of systematic errors. In this context, on a first approximation basis, a linear fit of all the data (black line in Figure A.6, upper panel) was performed, and compared with the r^{norm} limiting value: the two lines crosses at a particle flux value $F \approx 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$, below which the prompt emission of nitrogen atoms is negligibly small, since the extrapolated r_0^{norm} values would agree with the r^{norm} limiting value. For highly focused deuteron beams at 2.7 MeV (with $\approx 1 \text{ mm}$ diameter) this particle flux would correspond to deuteron beam currents of approximately 10 nA.

It is interesting to observe that the slopes of the lines resulting from the fit of the r_0^{norm} and r_{∞}^{norm} data are quite different; this point would presumably indicate a different *nature* at the basis of the two mechanisms of emission. In this framework, it would be very interesting to extend this type of analysis by exploring different bombarding energy regions (e.g. in the 1-2 MeV and 3-5 MeV ranges) and also by changing the *nature* of the impinging particle, to strongly vary the specific energy loss.

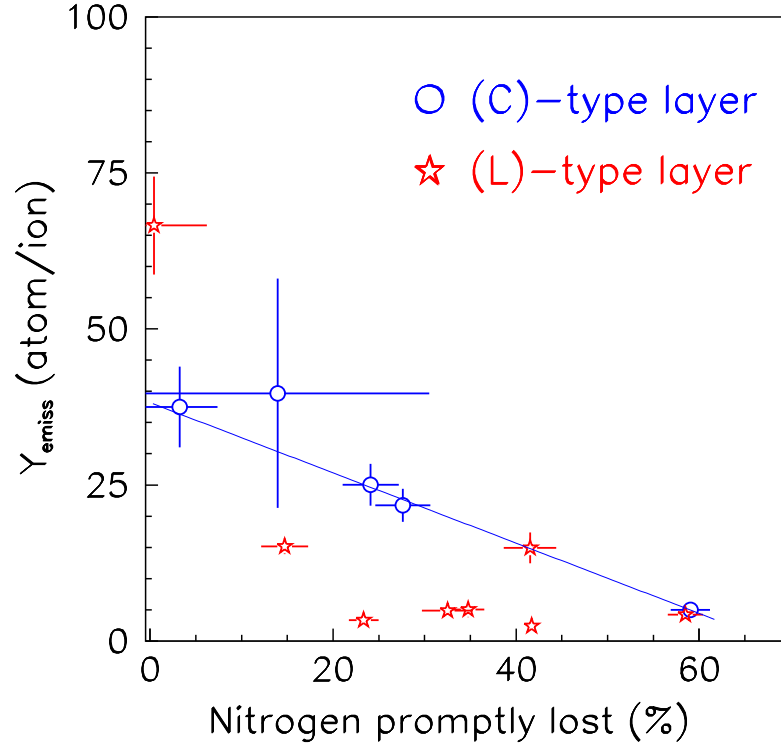


FIGURE A.7: Yield of nitrogen atoms at the beginning of the *slow* emission phase (Y_{miss}) as a function of the percentage of nitrogen lost during the *prompt* emission phase. Blue circle: (C)-type layer; red stars: (L)-type layer. The blue solid line represents a linear fit of (C)-type layer data only.

To investigate the effects due to the roughness of the Melamine layers on the prompt emission, the destruction cross section σ_{dest} was correlated with the percentage of nitrogen atoms lost during the *prompt* emission; the last quantity can be estimated by considering the difference between r^{norm} and r_0^{norm} , normalized to the r^{norm} limiting value. The plot correlating the two quantities is reported in Figure A.7, with different symbols and colors on the basis of the different type ((C)- or (L)-type) of the layer. For the data coming from the (L)-type layers, which are characterized by a greater roughness, no evident correlations are observed: with the exception of three points, the σ_{dest} value are highly clustered around $3 \cdot 10^{18} \text{ cm}^2$, irrespective of the variation (in the $\approx 20\text{-}60\%$ range) of the nitrogen lost by prompt emission. At variance, an interesting linear-like correlation ($R^2 \simeq 0.83$ for a weighted linear fit) is observed for the smooth (C)-type layers (blue open dots): the destruction cross sections linearly decrease when increasing the percentage of nitrogen atoms lost in the prompt emission. This effect could be qualitatively explained by considering the re-processing of the bombarded materials at the very beginning of the irradiation: large fractions of nitrogen atoms promptly lost could be in fact indicative of both a significative surface modification, with an artificial increase of the roughness of the bombarded surface, and a scenario in which a sizable part of the remaining nitrogen atoms were subject to the formation of new (and more tight) types of bonds with carbon atoms of the material bulk. In both cases, such a modification, occurring in the first phase of the collision, can lead to a systematic variation of the destruction cross section. Unfortunately, in the environment presently used, it was not possible to perform an infrared analysis on the irradiated samples to trace the evolution of the nature of the bonds formed during the bombardment; a new technical setup is

currently under development to allow this important characterization of samples that would better fix the peculiar behavior of Melamine in the very first phase of irradiation.

A.5 Summary and conclusions

In this Appendix, the degradation effects induced by ion beam bombardment of Melamine ($\text{C}_3\text{H}_3\text{N}_6$) samples coating very thin carbon layers have been investigated, exploiting deuteron beams of 2.7 MeV bombarding energy, with quite high fluxes, ranging in the $\approx 1.5 - 3.5 \cdot 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$ region. The two types of bombarded samples have a markedly different roughness, as clearly measured by the SEM analysis of the Melamine layers and of the carbon backings. The main investigated signal of degradation was related to the destruction of the Melamine molecule, giving rise to the emission of nitrogen atoms. The loss of nitrogen was continuously measured via the $^{14}\text{N}(\text{d}, \alpha_1)^{12}\text{C}^*$ reaction; in particular, the α particles of interest were detected by a telescope of solid state detectors, operated in anti-coincidence mode to minimize the background due to other reactions.

The rate of α particles obtained from the bombarded samples was monitored as a function of the fluence in a very broad range, and for all the samples investigated, the typical exponential-like decrease of nitrogen content in the Melamine, due to the continuous molecular destruction (a consequence of the strong ionization induced by the beam particles) was observed. The fit of data with a commonly adopted exponential-like function allowed to estimate the destruction cross section σ_{dest} for each irradiated sample. The σ_{dest} values were observed to be strongly correlated to the roughness of the samples, and in particular that, on the average, layers with a larger roughness are more resistant to the beam-induced nitrogen emission, therefore showing a smaller σ_{dest} . It is possible that the surface rugosity could favor a re-enter of nitrogen atoms in the material bulk, forming new types of molecular bonding with carbon atoms. Future plans to experimentally investigate this point with dedicated infrared spectroscopy of the samples after the bombardment phase are already under design.

The analysis of the rate curves evidenced the occurrence of a saturation value of the nitrogen content that is still present in the samples even at very large elapsed fluence; it is possible that this effect is due to the establishment of a dynamical equilibrium between the emitted and re-absorbed nitrogen atoms from the damaged sample. The data extrapolation to very low fluence values unveiled the existence of an interesting *prompt* emission mechanism, leading to a very rapid emission of nitrogen atoms at fluence values smaller than a few 10^{15} cm^{-2} . This experiment looked for the physical boundaries of such prompt emission, since it can be an uncontrolled source of systematic error when performing absolute nuclear cross section measurements or ion beam analyses involving organic materials, such as Melamine. It was found that prompt emission is negligibly small for particle flux values smaller than $\approx 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$, irrespective of the roughness of the samples. Finally, the inspection of the destruction cross sections as a function of the percentage of nitrogen atoms promptly emitted in the first phase of bombardment shows a linear-like correlation, valid only for the smoother samples; in particular, samples characterized by a large fraction of nitrogen lost in the prompt phase are characterized by smaller destruction cross sections, thus indicating a more robust behavior under subsequent (and long lasting) ion beam bombardment. This finding seems to indicate a dependence of the destruction cross section from the previous irradiation *story* of a given sample, possibly because of significative changes in the

topology of the bombarded organic layers even at moderately low fluence values. Further investigations with different types of bombarding particles and different beam energies are planned in the future to trace the evolution of the observed correlations as a function of the specific energy loss in the organic material.

Appendix B

Lanthanides and Actinide targets for heavy ion physics

The contacts between chemistry and nuclear physics become more and more close when approaching the last boxes of the Periodic Table. For example, when studying the properties of the heaviest elements (such as actinides), it is indeed necessary to take their radioactivity into account. To dive into the heart of the matter, these elements are necessary for the fabrication of targets (which, of course, happens through the application of refined chemical processes) for the production of superheavy elements through heavy-ion irradiation. This Appendix will describe a new procedure to obtain stable targets suitable for this type of investigations.

B.1 Temperature survivability of SiC-Ta-Pd targets for the production of superheavy elements

B.1.1 Introduction

The production of superheavy elements (SHEs) requires the utilization of actinoids targets fulfilling some precise requirements. First, the target needs to be a good thermal and electrical conductor, in order to dissipate the heat and the charge deposited by the impinging beam. It also needs to withstand vacuum atmospheres and mechanical stresses during the placement in the frame; most importantly, it must be chemically stable, inert with respect to oxidation and compatible with the backing material. Unfortunately, metallic late actinoid targets are difficult to produce due to the low quantities available. Molecular Plating (MP) is currently the method of choice used for preparing such targets, but the deposited actinoid films are thermally and electrically insulating. This might present a problem for irradiation with high-intensity heavy-ion beams for long periods of time. A class of compounds that allow to produce conductive actinoid films is that of Intermetallic Compounds (IMC), i.e., a metallic alloy of two or more elements with ordered solid-state structures. An intermetallic target is composed of a host and a target material and has a crystal lattice different from those of the initial reagents. Targets for SHEs production in form of IMCs have already been produced by adding a thermal treatment under hydrogen atmosphere, the so-called coupled reduction (CR), after MP, and have been successfully tested [339–341]. Unfortunately, the

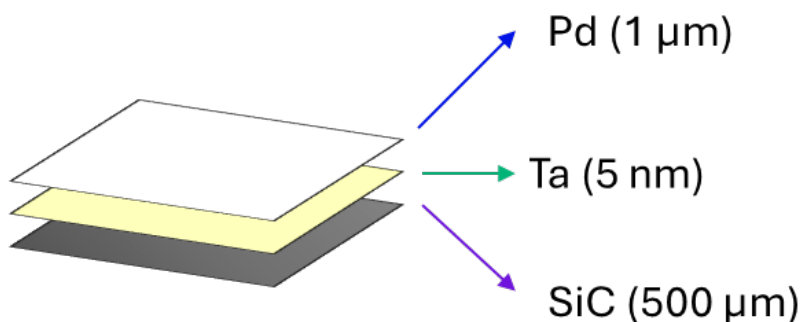


FIGURE B.1: Schematic representation of the layered target base exploited in this work for the production of Tb:Pd intermetallic targets.

tested targets appeared too thick for SHEs production. To confine the target material in a thinner layer and to make it more resistant to the production process, it is possible to embed it in a multilayered structure, composed of a silicon carbide (SiC) backing, a thin Ta seeding layer, and a Pd film on the top, it being the intermetallic host, as reported in Figure B.1. The aim of this study is to synthesize an intermetallic target of Pd hosting a lanthanoid (as homologue to late actinoids), to design the process and delineate a methodology for the production of actual actinoids targets in the future.

B.1.2 Materials and methods

The layered target is composed by a first 500 μm -thick layer of epitaxially-grown single-crystal 4H-SiC, used as high melting point and diffusion-resistant backing, a thin 5 nm Ta deposition (seeding layer) and, on top, a 1 μm thick Pd layer. Ta and Pd films were grown on SiC substrates using the DC magnetron sputtering (AJA International, Inc.). Immediately before deposition, the substrates were cleaned in-situ using an Ar^+ plasma generated by supplying 30 W RF power to the substrate holder in a 13.3 mbar Ar atmosphere. Next, a 5 nm-thick Ta layer was grown to promote the adhesion of the Pd layer, followed by 1 μm of Pd. Both materials were deposited at room temperature using a sputtering power of 100 W DC in a 4 mbar Ar partial pressure using Ta and Pd targets (99.95%, Kurt J. Lesker, Inc.). Before depositing the films, the Ta and Pd deposition rates (0.08 nm/s and 0.13 nm/s, respectively) were determined by growing reference films for 10 min and measuring the film thickness using low-angle X-ray reflectivity. Past works demonstrated that sputtered Pd films typically assume a (111) fcc polycrystalline texture when synthesized at room temperature [342–344].

The deposition of Tb was carried out by MP. An aliquot of 20 μL of a $\text{Tb}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ solution (15 mg in 200 μL 0.01 M HNO_3) was used to obtain a areal density of 0.67 mg/cm^2 Tb^{3+} ions on the multilayered substrate. In this way, the Tb^{3+} ions are deposited on the Pd surface as result of the application of a constant current density of approx. 1.27 mA/cm^2 between a Pt anode and a Cu cathode for 45 min in N,N-dimethylacetamide solvent (99%, Sigma-Aldrich). It is hypothesized that $\text{Tb}(\text{OH})_3$ species, insoluble in the organic solvent, are formed during the galvanostatic treatment, and then deposited on the surface of the multilayered substrate. However, the Tb^{3+} ions still need to be reduced to metallic Tb and to diffuse into the Pd layer; this has been done through CR, where, after three cycles of vacuum ($\sim 10^{-4}$ mbar) and He-flushing

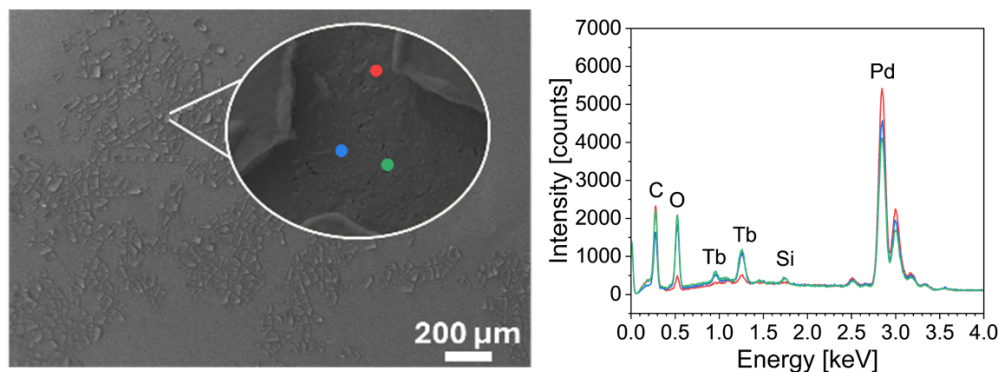


FIGURE B.2: SEM image of one Pd layer defoliated from the substrate, after a 30 min thermal treatment at $T = 400\text{ }^{\circ}\text{C}$. Point EDX spectra correspond to the points in the magnified area reported in the image.

(4 bar, 100 mL/min), the samples were treated at different temperatures (200, 400, 800 and 900°C) under H_2 flow (4 bar, 25 mL/min). To investigate the thermodynamical and kinetic dependence of the Tb diffusion into the Pd layer, other parameters such as the Tb concentration (0.33 and 0.67 mg/cm^2) and the heating time (30 min and 24 h dwell time at the maximum temperature, with a heating rate of 10°C/min) were varied. The samples, after each step, were characterized by Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray spectroscopy (EDX), using an acceleration voltage of 10 kV.

B.1.3 Results and discussion

The first observable result of the MP is a layered deposition of flakes containing Tb, C and O on the palladium layer, as confirmed by the SEM images and the EDX spectra (not shown here). During the thermal treatment, the various samples showed different responses as a function of the heating temperature: as expected, no effects were observed for a 200°C treatment, and the Pd layers were unstable at $T \geq 800\text{ }^{\circ}\text{C}$. The most interesting feature is observed at $T = 400^{\circ}\text{C}$, thus chosen as the working temperature, at which the Pd layer defoliates, forming a self-supporting target. Only a very thin Pd layer remains on the SiC-Ta backing, as confirmed by the EDX spectrum, in which Pd and Si peaks can still be seen. The defoliation happens to be extremely useful, since the Pd foil is resistant enough to be handled and to be mounted on a target frame and keeps the Tb deposition on it; the self-defoliation of the target is a favorable feature since it avoids an originally designed further step, aimed to mill the SiC-Ta layer from the backside.

The deposition and diffusion of the Tb atoms on the defoliated Pd layer, after the 30 min treatment at 400°C , has been investigated through SEM-EDX analysis, reported in Figure B.2. As it can be seen from the O, Tb and Pd peaks took in three different spots of the lower layer of the deposition, the diffusion is still not homogeneous, hinting that longer treatment times may be needed or saturation of Tb into the Pd bulk may be reached. On the other hand, when a sample was treated at the same temperature, but for a 24 h heating time, the defoliated Pd layer does not have the same quality and resistance as those obtained with a 30 min treatment. This is pointing towards the necessity of finding an optimal treatment time, longer than 30 min, to obtain a more

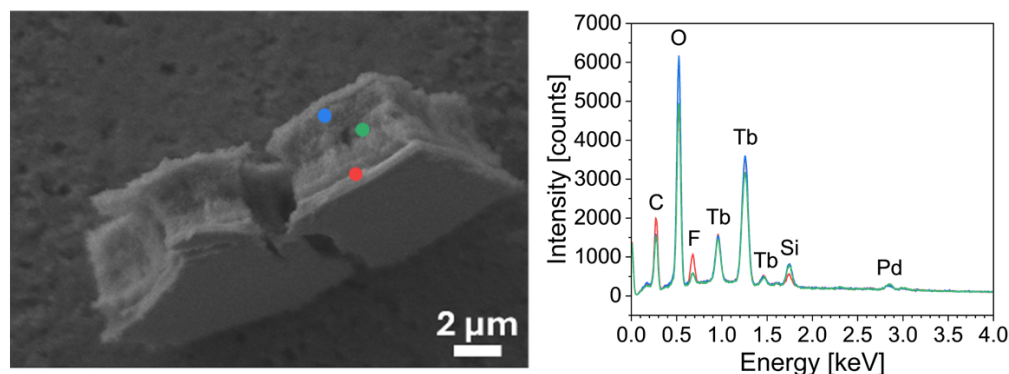


FIGURE B.3: SEM image of a flake of the Pd layer of Figure B.2, found lying on its side over the SiC backing with corresponding point EDX spectra. The total thickness of the layered deposition is of about 3 μm .

homogeneous diffusion, but shorter than 24 h, so that the target can resist it. On a side note, the diffusion of the Tb into the Pd lattice seems to make it sturdier and more resistant to high temperatures: when the sample was treated for longer times, it was possible to identify two fractions of the original defoliated Pd layer: pure Pd, more shattered and in some points still stuck on the SiC backing, and a more tough part, corresponding to the central disc on which the MP was performed.

During the SEM-EDX analysis of the target backing Figure B.3, a small flake coming from the lost Pd foil was found lying on its side. The study of the peaks related to the various layers allows to make some conjectures on what the distribution of the different elements in the final product at these operative conditions could be. The total thickness of the flake is around 3 μm , and the top layer appears to be mostly composed of Tb and O, showing only a weak Pd peak; the bulk shows similar Tb and Pd signals, while the bottom layer is obviously mostly Pd, but also shows the presence of Tb. Given the original thickness of the Pd layer of 1 μm (one third of what was now obtained), variations in the deposition concentration (lower Tb loadings) or technique (to cover the total area of the target instead of the central $d = 1$ cm disc) may be adopted in the future.

B.1.4 Outlooks and future perspectives

The intermetallic target samples produced by the MP+CR method seem to have some suitable characteristics for a future application with late Actinides for the productions of SHEs. Although some aspects of the production process (such as the thermal treatment time and the effective deposition method) still need to be tuned, the first results are promising. The next steps would include a more thorough characterization of the lanthanide diffusion into the Pd foil (e.g., by using an α -emitting isotope) and the improvement of the yield of the process, crucial when working with very limited quantities of Actinides.

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