

Chapter 1

Introduction

1.1 Historical outline

1.1.1 The discovery of the α -radioactivity

The study of nuclear fission is one of the earliest topics in nuclear physics which enflamed the minds of a lot of brilliant physicists. The importance of the discovery of radioactivity at the end of the XIX century had a capital importance in the acceptance of the atomic concept in the scientific community. In continuation to the discovery of the Röntgen rays by Wilhelm Conrad Röntgen, Antoine Henri Becquerel observed in 1896, the emission of a radiation from Uranium salts which caused the blackening of the photographic plate and the ionization of the air. A large number of studies were then triggered by this discovery. Pierre and Marie Curie studied systematically all the elements from the standpoint of radioactivity and discovered two new chemical elements which were called Polonium and Radium.

In 1899 Ernest Rutherford discovered that the *Uran emission* is formed from two components which differ in their penetration properties. The light absorbed radiation was called α -radiation whereas the penetrating one was called β -radiation. In 1900 Paul Villard discovered a third type of radioactivity, which was later called γ -radiation. The α -radiation was recognized from the beginning to be a current of energetic charged particles. The explanation of the nature of this radiation needed a longer time. In 1908 E. Rutherford and H. Geiger, discovered that the charge of the α particle is carrying 2 elementary charges and that its mass carries 4 mass units [1]. From here it was inferred that most likely the α -particles are formed from positively charged Helium ions. One year later, E. Rutherford and T. Royds were able to observe the optical emission spectrum of the neutralised α -particles which confirmed this assumption [2].

The α -particles produced in radioactive decay were, as mentioned above, very energetic, a fact which proved to be very convenient in using them as projectiles for atomic structure studies. The dispersion experiments of E. Rutherford led in 1911 to the present picture of atom structure [3]. According to this picture the atom consists from a positively charged nucleus, responsible for almost the entire mass, and a cloud of electrons found at a relatively large distance from the nucleus. As the radioactive decay showed, the nucleus was not an unvarying entity. Through emission of α particles the nuclear charge and thus the atomic number of the atom was decreased by two units. Contrary to that, through the β -decay the

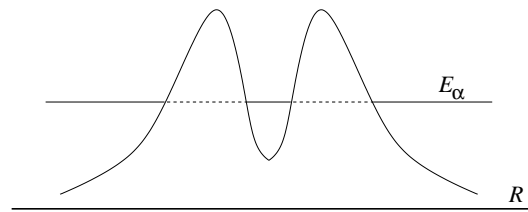


Figure 1.1: Naive representation of the potential energy of an α -particle inside a nucleus which undergoes α -decay.

charge of the nucleus was increased by one unit. Rutherford succeeded the first artificial atomic transmutation in 1919 by irradiating the Nitrogene with α particles[4]. As reaction products he obtained Oxigen and Hidrogen isotopes. The atomic number 7 became 8 and thus the fantastic old dream of alchemists, to transform elements, was in a peculiar way fulfilled. By extending the type of targets, Rutherford and collab. found every time Hydrogen when bombarding with α particles, a fact which immediately suggested that the Hydrogen must be a part of other nuclei. Rutherford named it "Proton".

It was soon realized that the α -particle is bound in the nucleus by the strong nuclear forces and outside the nucleus is repealed by the positive charge of the rest of the nucleus. If one represents the α -nucleus potential energy as a function of the reciprocal distance, then a volcano-like shape is obtained as sketched in Fig.1.1. Before the decay the α -particle is found inside the volcano and after decay outside. The height of the volcano wall is higher than the energy of the emitted α -particle. According to the laws of classical mechanics the α -particle cannot overcome the potential wall, but according to the quantum mechanics principles there is a certain probability that the α -particle is found outside the volcano. G.Gamow [5] and independently from him, R.W.Gurney and E.U.Condon [6], were able in 1928 to explain the α -decay process. It was stressed that the probability to tunnel the potential barrier is strongly dependent on the mass of the emitted particle, which means that for heavier nuclear fragments, as happens for cluster radioactivity and spontaneous fission, this probability is much smaller.

1.1.2 The discovery of the nuclear fission and its physical interpretation

In 1938 Hahn and Straßmann began a radiochemical analysis of the elements produced in neutron-U collisions. Among the products of neutron-U interactions they identified three isotopes of Ba, which have $Z = 56$, i.e. roughly half of that of U[7]. It was for the first time when a nuclear reaction produced changes in Z larger than 2. They showed that ^{232}Th is splitting in a similar manner under neutron shelling. They were also able to identify fragments of Sr ($Z=36$) and Yt ($Z=39$) and formation of noble gas elements. The nuclear fission was therefore discovered.

The interpretation of this phenomenon based on the Droplet Model was given shortly after by Meitner and Frisch[8]. The Droplet Model was developed in the preceeding years by G.Gamow, C.F. v. Weizsäcker, H.Bethe und N.Bohr and views the Nucleus as an electrically charged liquid drop. The attractive forces between the nucleons outweigh the opposite electric repulsion of the protons, such that the nucleus will be stabilized against small defor-

mations, in the same way as liquid drop through surface tension. In order to have a rough estimation of the kinetical energy acquired by the nuclear fragments through Coulomb repulsion, they used the Coulomb law for spherical charges:

$$E_{\text{kin}} = \frac{Z_1 Z_2 e^2}{d}$$

Z_1 and Z_2 are the nuclear charges of the two fragments, e the elementary charge and d the distance between the charges centers-of-mass. Since Hahn and Straßmann discovered that the heavier fragment is Barium, the complementary fragment in a binary splitting should be the noble gas Kr ($Z = 36$). The value of d was chosen to be of the order of fragment dimensions, approximately 1.5×10^{-15} m. Thus Meitner and Frisch evaluated for the kinetical energy of the fragments flying apart a total of 200 MeV. The description of the splitting process was given in a classical picture. The quantum tunneling effect was apparently not necessary in order to explain the results of Hahn and Straßman. Due to the large proton number in Uranium its nuclear building blocks are weakly bounded compared to those of resulting fragments.

Meitner and Frisch assumed that after the neutron capture in the isotope ^{238}U , the nucleus ^{239}U is splitting, based on the fact that in the nature the largest fraction of Uranium is made up of ^{238}U (99.275%) and only 0.720% from ^{235}U and 0.005% from ^{234}U . In fact only ^{235}U is fissioning through bombardment with slow neutrons.

After evaluating that the kinetical energy of each fragment has 100 MeV, a value which is 10 to 20 times larger than the highest values of the α -decay known at that time, Frisch conjectured that the fission products may be easily identify due to their high energies. With the help of a ionization chamber he succeeded to prove this [9].

Along with a large number of experimental studies triggered by the Hahn-Straßman discovery, the new phenomenon started to be debated by several theoreticians. The starting point was the interpretation of Meitner and Frisch. At first was studied the fissility of heavy nuclei as a function of mass and charge number. Meitner and Frisch assessed that a nucleus with more than 100 protons is unstable against fission. More precise estimations have been made by the C.F.v.Weizsäcker, E.Feenberg and J.Frenkel which independently arrived at a similar conclusion. But the best theory of nuclear fission given at that time was the one developed by Bohr and Wheeler [10]. Using the above mentioned Droplet Model of the nucleus they conjectured that from the incompressibility condition of the nuclear matter flow, the volume of the nucleus should remain constant for any kind of shape. Since for a spherical shape a nucleus has a minimal surface, then when the nucleus undergoes a deformation the surface increases and due to the surface tension the nucleus tends to regain its spherical form. However the Coulomb repulsion tries to increase further the deformation and to separate the protons. By virtue of the equivalence between mass and energy, expressed by the Einstein relation $E = mc^2$, the strength of the surface tension can be derived from the total mass of the nucleus. For light nuclei the surface tension is much stronger than the Coulomb repulsion. For very heavy nuclei, like Thorium or Uranium for example, small deformations can lead to predominance of the repulsive Coulomb forces compared to the surface tension and the nucleus is splitting. The highest point on the fission path was denoted by fission barrier. If the neutron captures a neutron, an excited state of the intermediary nucleus (compound nucleus) is attained. According to the Bohr's idea this intermediary nucleus will be described by

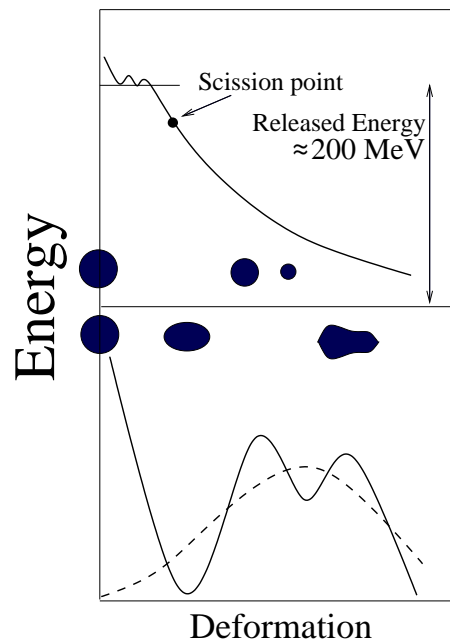


Figure 1.2: Potential energy of the fissioning nucleus in the region of Uranium as a function of the deformation. The lower panel is a magnification of the upper panel at the beginning of the curve, i.e. around the barrier threshold. The dashed curve corresponds to a Droplet Model calculation. The continuous curve takes into account shell effects.

means of the mathematical statistics, a concept which proved to be very fruitful in the theory of nuclear reactions. The nucleus is a very dense system such that the excitation energy is rapidly distributed between the different particles of the nucleus. Because each nucleon receives only a small fraction of the total excitation energy, the nucleus will stay a relatively long time in the excited state ($\approx 10^{-15}$ s) compared to the time necessary for the nucleon to cross the nucleus ($\approx 10^{-21}$ s). There is a chance that enough energy leaves the nucleus and concentrates on a single nucleon. It can also happen that the energy is relocated in the surface region and nuclear surface starts to oscillate, and if the excitation energy is large enough in order to surmount the barrier, then the nucleus is splitting. Bohr and Wheeler discussed in detail the formation of the compound nucleus, the magnitude of the fission threshold and other experimental findings on fission. However they described the deviation of the nucleus from the spherical shape only for very simple deformed shapes. More complicated shapes have been considered later, after World War II, using the first electronic computer (ENIAC), by Frank and Metropolis [11]. Their calculations, carried out in the frame of the Droplet Model, lead to the result that the fission of the nucleus in two identical fragments is the most favorable from an energetic point of view. This was in contradiction with the experimental results on the energy distribution of fission products in ^{235}U which indicates that, based on the momentum and energy conservation laws, the fissioning nucleus prefers the splitting in two fragments with different masses. The mass yields of the fission products indicated also this tendency to asymmetry [12]. It was clear at that time that the origin of the asymmetry in fission cannot be explained by the Droplet Model.

In 1948 Maria Goeppert-Mayer reported on experimental findings which indicated that

the nucleus possesses a shell structure similar to the electron shell of the atom. She showed that nuclei with 2, 8, 20, 28, 50, 82 or 126 neutrons or protons are particularly stable[13]. Fragments with neutron numbers between the magic numbers 50 and 82 should be favored in the fission of ^{235}U , with respect to fragments resulting from a symmetrical splitting.

A step forward in the theory was brought by V.M.Strutinsky who proposed a method to compute shell-corrections for strongly deformed nuclei[14]. Depending on the deformation the magic numbers can change compared to the ground state values. The fission barrier is changing dramatically compared to the one estimated in the Droplet Model. In fig.1.2 a schematic plot of the barrier in Uranium region is given, when one takes into account the shell-corrections according to Strutinsky (continuous curve) and when only the droplet part is considered (dashed curve). When shell-corrections are taken into account the nucleus is already deformed in its ground state and its potential energy is swinging between several maxima and minima with increasing deformation. At very high deformations the Coulomb repulsion is dominating and the nucleus is necking. At the scission point the two fragments are still in touch, but at large distances they are well separated. Thus the fission can be reached through tunnel effect from the first or second minimum. In case the nucleus is in its ground state, and therefore not excited, one deals with the spontaneous fission case. If the nucleus is splitting through tunnel effect, after he was excited on a state under the barrier, then one talks about fission below the threshold and if we decay from a state from the second minimum one deals with isomeric fission.

1.1.3 The discovery of nuclear molecules

In the early 1960s Bromley et al. measured the gamma radiation yields from the $^{12}\text{C}+^{12}\text{C}$ interaction at center-of-mass energies near 6 MeV and obtained sharp peaks in their bombarding energy dependence[15]. This result was intriguing because at 6 MeV center-of-mass energy, in the midst of the resonance spectrum, the classical distance of closest approach assuming pure Coulomb repulsion is greater than 8.5 fm, while the radius of a single ^{12}C nucleus, as determined at that time by electromagnetic or strong-interaction scattering experiments, is only about 3 fm! These resonances were requiring a strongly attractive nucleus-nucleus interaction at bombarding energies where the colliding nuclei were barely touching. Another puzzling feature of these Coulomb barrier resonances was their small total widths of approximately 100 keV, corresponding to a lifetime which exceeds the collision time by a factor of 10. The resonances were appearing at an excitation energy of approximately 20 MeV above the ground state of the compound nucleus, ^{24}Mg , where the level density is measured in hundreds of levels per MeV of excitation and therefore a natural question occurred: how can the resonance strengths remain sharply concentrated, and not be totally damped or dispersed into this near continuum?

An attempt to explain this resonant structure was given first time in [15]. It was suggested that the large widths are leading to an analogy with a metastable diatomic chemical molecule. A quasi-molecular interaction between the carbon nuclei was postulated as in Fig.1.3. The absorptive core implied that if the two carbon nuclei attain separation radii in this range, they coalesce and lead eventually to reaction products.

Vogt and McManus [17] suggested that the outer maximum results from deformation of the Carbon nuclei, while they are still bound together following a grazing collision effec-

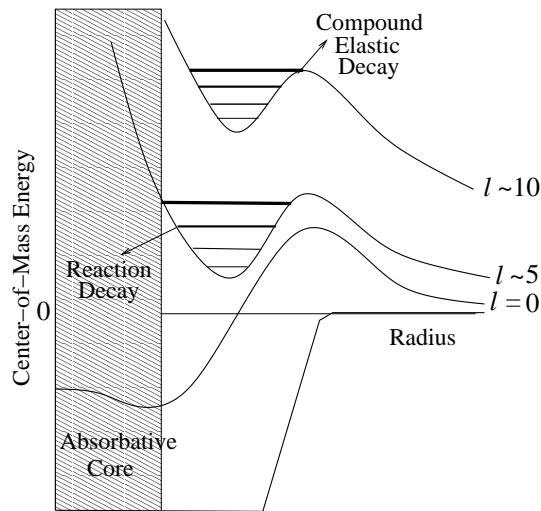


Figure 1.3: Schematic interaction potential for the C+C system for three representative orbital angular momenta as given by Bromley et al.[16].

tively by the nuclear interaction. On the other hand, Davis [18] has suggested that the outer potential maximum is simply the reflection of the ordinary optical model potential appropriate to the system when added to the Coulomb and centrifugal potentials. At first sight these suggestions appeared to be quite different because the equilibrium molecular separation as proposed by Vogt and McManus was in the range 7-10 fm, whereas as proposed by Davis it was ~ 5 fm. We know today that contrary to the assumption of these pioneering papers the Carbon nuclei are not spherical but well deformed, and that the deformability is not so large as advocated by Vogt and McManus. However the hypothesis of a quasi-molecular proved to be fruitful.

An essential step forward in the theoretical understanding of these resonances was done by Scheid, Greiner and Lemmer [19] who suggested that the experimentally observed intermediate structure in the cross section of elastic scattering is due to the quasibound molecular states while the gross structure originates from virtually bound molecular states of the nucleus-nucleus system. For that they introduced the *Double Resonance Mechanism* according to which the elastic and inelastic partial waves of the relative nucleus-nucleus motion resonate simultaneously with the corresponding virtual and quasibound molecular states in the potential, thus largely enhancing the transition strength between the elastic and a certain inelastic channel. The double resonance effect requires necessarily that the difference in energy and angular momentum between the virtual and quasibound states can be taken over by the intrinsic configuration of the nucleus-nucleus system.

It became clear in the last two decades that the resonant behavior observed in $^{12}\text{C}+^{12}\text{C}$, $^{12}\text{C}+^{16}\text{O}$, $^{12}\text{C}+^{13}\text{C}$, $^{16}\text{O}+^{16}\text{O}$, $^{16}\text{O}+^{24}\text{Mg}$, is not an isolated phenomenon occurring peculiarly only in a few systems and would disappear as the complexity of colliding nuclei is increased. Nowadays there is a wealth of experimental evidence that this behavior persists even in heavier nuclei, such as the $^{24}\text{Mg}+^{24}\text{Mg}$ [20], $^{28}\text{Si}+^{28}\text{Si}$ [21].

1.1.4 The discovery of superheavy elements

In the century following the discovery of radioactivity in U minerals by Bequerel, more than 30 new elements have been added to the periodic table of elements. The most recent nuclide charts [22] contain as the last entry the superheavy nucleus $^{292}116$ with a life-time of the order of ms.

In the first period (1896-1939) radioactive elements between Bi($Z=81$) and U($Z=92$) have been discovered using chemical methods. All of them were products of the primordial elements U and Th.

In the second period (1934-1955) elements were produced artificially. Neutron capture of the heaviest isotopes in the high neutron fluxes of nuclear reactors provided large quantities of the new elements. Each neutron captured by the target atom's nucleus underwent β -decay, changing into one proton and one electron and creating an element that had an additional proton compared to the target. At the end of this period researchers had produced the elements 93, 94, 99 and 100 in this way. In the same time they created elements 95, 96, 97, 98 and 101 by irradiating heavy nuclei with currents of α -particles, boosting thus the atomic numbers two steps at a time.

In the third period (1955-1974), a period characterized by the development of particle accelerators, the long-lived isotopes of the heaviest actinides produced in nuclear reactors were fused with high-intensity beams of isotopes of light elements B to O. In order to initiate the fusion it was necessary to collide the projectile and target with enough energy to overcome the electrostatic repulsive force. Consequently the compound nuclei, after fusion, were heated owing to excitation energies between 40 to 50 MeV. For this reason this production method was called *hot fusion* or *actinide-based fusion*. The heaviest element produced in this way was Seaborgium (Sg, $Z=106$) in 1974. The heat increased the likelihood that the new elements fission rather than relax into a stable state. Thus, above Sg the tendency to fission made this technique impossible to synthesise new elements.

In the last period, which began in 1974 and continues today (2003), closed-shell nuclei ^{208}Pb and ^{209}Bi were fused with medium-weight neutron-rich isotopes such as ^{54}Cr and ^{70}Zn especially in GSI and ^{208}Pb to ^{248}Cm with ^{48}Ca and ^{56}Fe in Dubna. Thus, in the early 1980s with the UNILAC facility in Darmstadt, it was possible to synthesize elements 107, 108 and 109, and after 1990, elements 111 and 112 were produced. In the last years the element 114 was produced in Dubna by irradiating the most exotic isotope of Pu, the one with $A=244$, with an intense stream of ions of ^{48}Ca . In 2000 the observation of the decay of the nuclide $^{292}116$, in the reaction $^{248}\text{Cm}+^{48}\text{Ca}$ was reported in FLNR-Dubna[23].

Starting with the middle of the seventies, using the theoretical frame of the two-center shell model (TCSM) [24, 25], the school of Frankfurt substantiated the necessity of bombarding the double magic lead nuclei with suitable projectiles [26]. It was shown that the shell structure of the two final fragments was visible deep inside the fusing nucleus, before the barrier was reached. The collective potential energy surfaces of heavy nuclei, as calculated in the framework of the TCSM, exhibit pronounced valleys which are promising doorways to the fusion of superheavy nuclei for certain projectile-target combinations. If projectile and target approach each other through those valleys, they get only minimally excited and the radial barrier which has to be overcome in order to fuse the nuclei is lowest compared to the neighbouring projectile-target combinations. In this way the optimal projectile-target com-

binations for the synthesis of superheavy elements could be predicted and this prompted the GSI group to follow this approach with the help of the SHIP mass-separator and to produce the new elements with $Z=106, 107, \dots, 112$.

1.1.5 The prediction of cluster radioactivity and its discovery

After the discovery of the α -decay process, it occurred naturally the question if nuclei may emit particle heavier than the α particle but lighter than the fission fragments.

The measurements of Rutherford and Robinson from 1914 [27] established that if such particles are emitted, then their number should not be larger than 10^{-4} compared to the number of Helium atoms.

The study of the heavy cluster emission in super-asymmetric fission was started in the middle of seventies in Dubna by the Romanian physicist A.Săndulescu and collaborators [28]. He discussed various mechanisms of decay and the possibility of observation of particles with masses intermediate between α and fission fragments. As the best candidates of cluster emitters were recommended the heavy isotopes of U.

The first experimental confirmation of the existence of cluster radioactivity was the observation of the decay of the nucleus ^{223}Ra with the emission of ^{14}C , as reported by two groups from England and Soviet Union [29, 30]. Each of these groups registered around ten events of emission of ^{14}C from the decay of ^{223}Ra with energy ≈ 30 MeV and the daughter nucleus (^{209}Pb) very close from the double magic values. These experiments proved that the probability of ^{14}C is 10 orders smaller than the probability of α emission. Naturally, the registration of these rare events in the large background of α -particles was a very difficult. Since then the ^{14}C decay of many other isotopes of Ra nuclei and many other heavier cluster-decays, has been observed, e.g. ^{20}O from ^{228}Th , $^{24,26}\text{Ne}$ from $^{230,232}\text{Th}$ and $^{232,234}\text{U}$, ^{23}F from ^{231}Pa , $^{28,30}\text{Mg}$ from ^{238}Pu , and $^{32,34}\text{Si}$ from ^{238}Pu and ^{241}Am have been observed.

It is also worthwhile to mention that the large majority of discovered emitters are even-even nuclei. To the date as uneven cluster is known only ^{23}F , emitted in the reaction $^{231}\text{Pa} \rightarrow ^{23}\text{F} + ^{208}\text{Pb}$. The heaviest cluster recorded until now is ^{34}Si observed in the reaction $^{242}\text{Cm} \rightarrow ^{34}\text{Si} + ^{208}\text{Pb}$ [31].

Except the heavy nuclei region there is known another cluster emitter, the neutron-deficient isotope ^{114}Ba which emits the cluster ^{12}C [32]. Similar to the heavy nuclei region where the daughter nucleus is the double magic ^{208}Pb or a neighbouring nucleus, the daughter of ^{114}Ba is the nucleus ^{102}Sn which is close to the double magic ^{100}Sn .

1.2 Actual problems in Cold Fission, Nuclear Molecules and Synthesis of Superheavy Elements

1.2.1 Experimental State-of-the-art in Cold Fission

In the binary nuclear fission of actinide nuclei the fragments are usually formed in highly-excited states which subsequently decay to their ground-states by emitting neutrons and gamma rays. However a small fraction of these fragmentations will attain a very high kinetic energy TKE which is very close to the corresponding binary decay energy Q . Since

in this case the fragments are formed with excitation energies close to their ground-states no neutrons are emitted. Milton and Fraser [33] were the first who noticed that some of the fission fragments are produced at such high kinetic energies that the emerging nuclei are formed nearly in their ground-state. Later on Signarbeux et al. [34] confirmed the previous interpretation by determining the mass distributions of the primary fragments for the highest values of the kinetic energy. They concluded that even before the scission takes place we deal with a superposition of two fragments in their ground state, from which the *cold fragmentation* term emerged. An interesting remark they made was that the odd-even fluctuations of Q due to nucleon pairing were not present also in the TKE_{max} values. In their view this smoothing of the odd-even effect was a consequence of a pair-broken from one of the fragments. The probability for neutronless fission is 0.0021 ± 0.0008 for ^{252}Cf .

In the last years the cold (neutronless) fission of many actinide nuclei into fragments with masses from ≈ 70 to ≈ 160 was an intensively studied phenomenon [35, 36, 37, 38, 39, 40, 41]. An important step in the understanding of the cold fission phenomenon was the observation that the final nuclei are generated in their ground states or some low excited states, which prompted some authors to relate these decays to the cluster radioactivity [42].

Since the fragments emitted in binary cold decays are produced with very low or even zero internal excitation energy, both fragments should have very compact shapes at the scission point and deformations close to those of their ground states [35, 43].

The first direct observation of cold (neutronless) binary fragmentations in the spontaneous fission of ^{252}Cf was made by using the multiple Ge-detector Compact Ball facility at Oak Ridge National Laboratory [39, 40], and more recently with the Gammasphere consisting of 72 detectors [41]. Using the triple-gamma coincidence technique, the correlations between the two fragments were observed unambiguously.

The Gammasphere and Eurogam facilities enable to identify this rare cold fission process using the triple γ -rays coincidence technique. Initially only few pairs of fragments were observed: ^{104}Zr - ^{148}Ce , ^{104}Mo - ^{148}Ba , ^{106}Mo - ^{146}Ba and ^{108}Mo - ^{144}Ba . More recent measurements evidenced a rich amount of combinations for even-even as well as for odd-odd splittings.

Very recently, the group of Tübingen reported some interesting results on the spontaneous decay of ^{252}Cf using a twin ionization chamber [44]. Two distinct mass regions of cold fission were observed: the first extending from the mass split 96/156 up to 114/138 and the second one comprising only a narrow mass range around the mass split 120/132.

The yields of the rotational states in binary cold fission were extracted from the intensities of γ -rays emitted in coincidence during the deexcitation of fragments for ^{104}Mo - ^{148}Ba and ^{106}Mo - ^{146}Ba [41]. It was shown that in cold fission, the angular momentum population is centered around the low-lying 2^+ and 4^+ states. The states higher than 6^+ are practically not populated, at variance with the strong ground-state band intensities, which are seen up to 14^+ for both nuclei, but as separate fragments. This proves the assumption concerning the cold rearrangement of nucleons during the cold fission.

1.2.2 Experimental State-of-the-art in Ternary Cold Fission

In the above mentioned experimental investigations of binary cold fragmentations, some indications of a third light fragment such as α , ^6He and ^{10}Be clusters [45, 46], were also

reported.

In a recent experiment [47] the cold fission decay of ^{252}Cf into three clusters was investigated. In coincidence measurements the three participants were identified as being ^{96}Sr , ^{10}Be and ^{146}Ba . In reanalyzing the data, two further systems were discovered, namely $^{90}\text{Y} + ^{142}\text{Cs}$ and $^{108}\text{Mo} + ^{134}\text{Te}$, all with ^{10}Be as the third particle. The data suggested that the transition from the first excited 2^+ state to the ground state in ^{10}Be was not Doppler broadened as one would expect if the system immediately separates into three clusters and the Be nucleus deexcites in flight. In addition it was observed that the transition energy of 3368 keV in the ^{10}Be nucleus interacting with ^{96}Sr and ^{146}Ba is probably by about 6 keV smaller than for the free ^{10}Be nucleus. The transition energy decreases further for the other two systems, being largest when both heavy clusters are spherical. The heuristic explanation was that the average shell model frequency in presence of the two heavy clusters is modified. The influence of both clusters leads to a softening of the ^{10}Be potential and thus to a somewhat smaller transition energy. The largest overlap, i.e. the strongest change in the average shell model frequency, of one heavy cluster with ^{10}Be is obtained for a spherical deformation. The interpretation of the above observation is the probable existence of a nuclear molecule with a half life larger than 10^{-13}sec [47]. Such large lifetimes would open up the possibility of a spectroscopy of giant nuclear molecules.

Independently of the experiment, there are some arguments that nuclear molecules of this type should exist: i) The cold fission of ^{252}Cf into three clusters is observed with an α -particle as the lightest nucleus [47]. ii) If an α particle can be emitted, there is no reason to believe that larger clusters cannot be emitted too. As an example serves the observation of heavy cluster radioactivity, though larger clusters are produced with a much smaller probability. iii) ^{10}Be consists of a core of two α -particles with two loosely bound neutrons [48] (and references therein), the latter being ideal to provide binding. iv) In theoretical calculations of molecular potentials, as we shall see in the course of the present work.

1.3 Structure of the work

Chapter 2 gives a presentation of standard methods which are dealing with the problem of quantum mechanical tunneling. In section 2.1 the stationary treatment of the barrier penetration is presented in the one dimensional case as well as in the two-dimensional case using the coupled channel formalism. In both cases we discuss the WKB approximation which provides formulas easy to evaluate.

Section 2.2 is dedicated to one-dimensional time-dependent methods. In sec.2.2.2 and app.B.2 we present in detail the numerical procedure to obtain the solution of the time-dependent Schrödinger equation and to compute the decay rate. As a study case we consider the α -decay of ^{212}Po .

In sect.2.3 we give an alternative way to derive transmission probabilities through barrier using the Feynman's path integral formalism.

The last section of chapter 2 deals with the problem of dissipation in quantum tunneling in the frame of Lindblad's theory of quantum open systems. An example is worked out for the neutronless spontaneous fission by approximating the barrier with two- and three-smoothly joined parabolic potentials.

These methods are used in chapter 6 to compute the penetrabilities(decay rates) and barrier crossing times in binary cold fission.

Chapter 3 reviews the most used methods to calculate the nuclear potential of fissioning or fusing nuclei. The macroscopic part of the potential is presented in 3.1 and formulas for the geometrical surface, Yukawa-plus-exponential and Coulomb (with and without finite-range) terms are given for an axial-symmetric nucleus. The influence of various multipolar deformations in Cassini parametrization on the macroscopic energy is discussed for the case of the fissioning nucleus ^{240}Pu .

The exposure continues with potentials used in heavy-ion reactions such as the double-folding potential (sec.3.3) or the proximity potential 3.4. Special attention is paid to the orientation dependence of the ion-ion potential when both target and projectile are deformed, a case less discussed in the literature.

In the last section of chapter 3 the self-consistent Hartree-Fock method is reviewed.

Chapter 4 gives an exhaustive derivation of the Hamiltonians of dinuclear and trinuclear molecules and their spectra in some cases of interest. The results of this chapter are used in the next chapter when studying the coupling of the fission and orientational degrees of freedom in cold fission.

In section 4.2 we present a soluble model for three nuclei. In order to obtain an analytical solution, several strong assumptions had to be made: i) the system is in a linear configuration, ii) the inclination angles of the nuclear symmetry axis to the axis which defines the linear orientation should be very small and iii) the light cluster has to be sandwiched between the two heavy ones. In 4.3.1 the mapping of the three-clusters geometrical picture to the algebraic one is given.

Chapter 5 gives a short introduction in the cluster radioactivity. In 5.2 this process is analyzed from the point of view of cold valleys concept. This part of the work is designed in order to make the connection with the later discussed phenomenon of cold fission. The dips in the driving potential of a sequence of actinides are identified with the most favorable cases of cluster decay.

Chapter 6 represent the gravitating point of the work. We apply the methods presented in the previous chapters to study the spontaneous cold fission. Several facets of this phenomenon are studied. The chapter is divided in two main parts. In the first part (sect.6.1) we expose the the cold binary fragmentation process in ^{252}Cf . To give a qualitative understanding of the process we display in 6.1.1 the driving potential of ^{252}Cf for different orientations and show that in order to obtain a qualitative agreement with the experiment the symmetry axes of the two fragments must be aligned (*pole – pole* configuration). Next we calculate for this configuration the mass-distribution yields and point that the experimental results can be understood only if the quadrupole together with octupole and especially hexadecupole fragments deformations are included in calculating the barriers. These calculations are performed under the assumption of vanishing excitation energy at scission. To get a hint on what happens when one switch-on the excitation energy we discuss in 6.1.2 the polarization of fragments at scission when one approaches the neutron-threshold in the excitation energy. It must be noted that all these calculations are performed for the case when the only dynamical variable is the elongation (fragment-fragment distance). The coupling of this predominant degree of freedom in fission with the rotational one is done in subsec.6.1.3.

The aim of subsec.6.1.4 is to investigate the formation of fragments angular momenta

in cold fission using the theoretical tools developed in 6.1.3. The scission configuration is pictured as a quasi-bound state of a giant molecule. In this model the angular momenta is carried by the small non-axial vibrations of the fissioning system which arise from the higher multipole components of the interaction potential. For the cold fission we consider only the contribution of the ground state of this vibrational spectrum, the first excited state being located at ≈ 5 MeV. In the case of pure cold fission ($E^* = 0$ MeV) the fragments deformations are taken to be those corresponding to the first minima in the deformation energy landscape. When the excitation energy increases, we recalculate the deformations by employing the LDM with a phenomenological receipt for the shell corrections.

The second part of chapter 6 deals with the cold ternary fission. The strategy adopted earlier to compute the mass-yields to the binary fission of ^{252}Cf is now adapted to the ^{10}Be -accompanied fission of the same actinide nucleus. In 6.2.2 we give an estimate of the possible shift of the first 2^+ of ^{10}Be when sandwiched at scission between the two heavier accompanying clusters.

Section 6.3 deals with the post-scission regime in ternary fission. Using classical trajectory calculations we estimate the final kinetic energies and asymptotic emission angle of the light particle accompanying the ternary cold fission.

The last chapter of the work discusses what in simple terms is the process inverse to the cold fission, i.e. the cold fusion. In sec.7.1 the concept of cold valley is again invoked to establish not only the most favorable projectile-combinations in the synthesis of superheavy elements, but also the most probable quasi-fission or cold spontaneous channels in the decay of these new elements. The problem of excitation of collective degrees of freedom for superheavy nuclei which are predicted by the relativistic mean-field to be double-magic or close to this number and spherical is considered in sec.7.2. Special attention is given to the possible anaharmonicities of the β -vibrational spectrum and to the features of giant electric resonances in superheavy nuclei with hollow-like structure.

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