Two center shell model description of superheavy element synthesis, fission and cluster decay

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Abstract. A two center shell model provides the possibility to characterize precisely scission configurations. Therefore, low energy disintegration processes could be described within such models. A Woods-Saxon two center shell model, recently elaborated, was used to investigate the cluster decay, the low energy fission and the synthesis of superheavy elements.

1. Introduction
The basic idea of the macroscopic-microscopic approach is that a macroscopic model, as the liquid drop one, describes quantitatively the smooth trends of the potential energy with respect to the particle number and deformation whereas a microscopic approach such as the shell model describes local fluctuations. The combined macroscopic-microscopic method should reproduce both smooth trends and local fluctuations. In this paper, the reliability of the latter approach is tested for cluster decay \cite{1, 2, 3, 4, 5, 6}, cold fission \cite{7, 8} and superheavy element synthesis \cite{9, 10, 11, 12} treated as cold rearrangement processes.

2. Two center shell model
In the macroscopic-microscopic method, the whole system is characterized by some collective coordinates that approximately determine the behavior of many other intrinsic variables. The basic ingredient in such an analysis is the shape parametrization that depends on several macroscopic degrees of freedom. The generalized coordinates associated with these degrees of freedom vary in time leading to a split of the nuclear system into two separate fragments. The macroscopic deformation energy is calculated within the liquid drop model. A microscopic correction is then evaluated using the Strutinsky procedure. The basic ingredient is the parametrization of the shape of the nuclei. In the following, an axial symmetric nuclear shape surface during the deformation process from one initial nucleus to the separated fragments is obtained by smoothly joining two spheroids of semi-axis $a_i$ and $b_i$ ($i = 1, 2$) with a neck surface generated by the rotation of a circle around the axis of symmetry. By imposing the condition of volume conservation we are left with five independent generalized coordinates $q_j$ ($j = 1, 5$) that can be associated with five degrees of freedom: the elongation $R$ given by the distance...
between the centers of the spheroids; the necking parameter \( C = S/R_3 \) related to the curvature of the neck, the eccentricities \( \epsilon_i \) associated with the deformations of the nascent fragments and the mass asymmetry parameter \( \eta = a_1/a_2 \). A shape consistent two-center shell model with a Woods-Saxon potential was developed recently [13]. The Blomqvist-Wahlborn parametrization is adopted because it provides the same radius constant \( r_0 \) for the mean field and the pairing field. That ensures a consistency of the shapes of the two fields at hyperdeformations, i.e., two tangent ellipsoids. The Hamiltonian is obtained by adding the spin-orbit and the Coulomb terms to the Woods-Saxon potential. The eigenvalues are obtained by diagonalization of the Hamiltonian in the semi-symmetric harmonic two center basis [14, 15]. This model was extensively used in estimating the dissipated during disintegration processes [16, 17, 18, 19].

3. Cluster decay

The theoretical predictions [1, 2, 3, 4, 5, 6] for spontaneous emission of fragments with intermediate masses between fission products and alpha decay were confirmed experimentally in 1984 [20, 21, 22, 23]. Since that time, the exotic decay was investigated intensively. As evidenced in Ref. [24], unified approaches of binary disintegrations in a wide range of mass asymmetries as many body theories were used to reproduce the half-lives or to predict new decay modes [25]. All these cluster decay descriptions were made on a phenomenological basis in terms of experimental \( Q \)-values. The \( Q \) value gives a reference to the external barrier. From a theoretical point of view, the very good agreement obtained within the phenomenological models provides us three hints: the internal barrier obtained in the overlap region must be small, the effective mass of the reaction is well approximated by the reduced mass and the external barrier is well approximated within the Coulomb interaction.

Recently, the macroscopic-microscopic approach was developed to treat in a unitary manner the cluster decay and the cold fission process. The fine structure was explained within such models [26, 27, 28]. A magic valley in the \(^{232}\text{U}\) and \(^{238}\text{Pu}\) potential energy surfaces were evidenced [29, 30, 31, 32]. This valley belongs to a mass asymmetry consistent with the formation of the \(^{208}\text{Pb}\) daughter and has its origin in the shell effects fluctuations. At the same time, a similar valley was obtained within the Hartree-Fock-Bogoliubov approximation [33, 34]. It is worth to mention that two valleys in the macroscopic-microscopic potential landscape were identified for the first time in Ref. [35]. One of these valleys corresponds to fission, while the second one, called fusion valley in Ref. [35] is related to a mass asymmetry compatible with the existence of a Pb daughter in a composite system. Our valley ressort from a calculation of the least action trajectory, by minimizing numerically the action integral from the ground state of the parent up to the exit point of the barrier. The method was initiated in Ref. [36] and was extensively used to describe the fission process [37, 38, 39, 40]. The results obtained for the \(^{32}\text{Si}\) emission from \(^{238}\text{Pu}\) are reproduced in the following, but the same qualitative behavior was obtained also for \(^{24}\text{Ne}\) emission from \(^{232}\text{U}\). As a result of the minimization, the family of nuclear shapes for the cluster decay along the least action trajectory is plotted in Fig. 1. This figure reveals the fact that at a distance of 7 fm between the nascent fragments, the emitted cluster is clearly formed. A piece of matter of a volume amounting that of the emitted fragment begins to be separated in the compound system by a neck.

The neutron and proton single-particle diagrams are calculated along the minimal action trajectory, from the ground state of the parent nucleus and beyond the formation of two separated fragments. The neutron level scheme is plotted in Fig. 3. In Fig. 3, at \( R \approx 0 \), the parent nucleus is in a spherical configuration. For small deformations, the system evolves in a way similar to a Nilsson diagram for prolate deformations. In the left side of Fig. 3, the orbitals of the parent spherical nucleus are labeled by their spectroscopic notations. The levels of the emitted fragment can be identified and are plotted by thick lines. Both fragments are spherical after scission and their levels are bunched in shells. The levels of the light fragment are
Figure 1. Family of shapes along the minimal action trajectory for cluster decay. The distance between the centers of the two fragments $R$ in fm is marked for each shape.

Figure 2. Upper part: potential energy surface $V$ as function of the elongation $R$ and the mass asymmetry $\eta = a_1/a_2$. Lower part: contour plot of the potential energy surface. The step between two equipotential curves is 4 MeV. The variations of the coordinates $\epsilon_1$, $\epsilon_2$, and $C$ follow the least action path as function of $R$. The least action trajectory is plotted with a thick curve.

labeled by their spectroscopic notations. The shell effects reflect the large scale non-uniformity in the energy distribution of the individual particle states. For the nuclear binding the level distribution around the Fermi energy is of special importance. The nucleus is expected to be more bound if the level density around the Fermi energy is smaller. In Fig. 3 it is clear that a shell closure begins to appear at $R \approx 6-7$ fm given by the gap that is formed around the $N=126$ daughter level. A similar phenomenon appears also in the case of the proton level scheme. This rearrangement of levels is translated in fluctuations of the shell effect energy values. The sum of shell and pairing corrections $\delta U$ are displayed in Fig. 4. The ground state is stabilized at $R \approx 4$ fm by these corrections. Notice that beyond $R \approx 7$ fm, the total shell effect monotonically decreases up to the asymptotic final value. According to Fig. 1, at this elongation (7 fm) the emitted nucleus starts its preformation, that is the shape becomes necked in the median region of the nuclear shape.

The macroscopic-microscopic potential is plotted in the upper panel of Fig. 5 by a full line.
Figure 4. Total shell effects with pairing corrections $\delta U$ as function of the internuclear distance $R$ are plotted with a thick line. The partial contribution of shell effects alone is plotted with a dashed line while the pairing correction is represented with a dot dashed curve.

Figure 5. Upper panel: the macroscopic-microscopic potential barrier $V$ for the $^{32}$Si emission as a function of the elongation $R$ is plotted by a thick curve. The thin curve represent the Coulomb energy $Z_1Z_2e^2/R - Q$ relative to the experimental $Q$-value of the process. The dashed line represents the barrier given by the liquid drop model, without shell effects. Lower panel: the effective mass along the minimal action trajectory calculated within the GOA (full curve), the semi-adiabatic (dashed curve) and the cranking model (dot-dashed curve).

The liquid drop energy is plotted by a dashed line. The Coulomb interaction corrected to the experimental $Q$-value, i.e. the phenomenological quantity $V_{COU} = Z_1Z_2e^2/R - Q$ is also plotted in order to test the validity of the model. In cluster decay, the major contribution of the action integral comes from the external region. The external barrier is very sensitive to the theoretical $Q$-value. In this way, it is possible to compare the external part of the barrier for the two cases and to estimate the deviation from the $Q$-value. The macroscopic-microscopic theoretical and the phenomenological values agree well enough in the external part of the barrier, the difference being less than 2 MeV. In the lower part of Fig. 5 the effective mass along the minimal action trajectory is plotted for three models [41, 42, 43]: cranking, Gaussian overlap approach (GOA) and semi-adiabatic formula. The three inertia exhibit a similar shell structure. The cranking model gives the larger values, the GOA the smaller ones, while the semi-adiabatic model has always intermediate values. The peak at $R \approx 4$ fm is especially due to the sudden variation of the asymmetry parameter $a_1/a_2$ as function of $R$. After the scission, the inertia reaches the values of the reduced mass.

Using the calculated values of the deformation energy and the inertia, the half-life of $^{32}$Si emission was estimated by using a semi-empirical formula $T_{1/2} = 0.72 \times 10^{-21} P^{-1}$ [s] where $P$ is the WKB penetrability. We used the three above mentioned models for the inertia. The values of $\log(T_{1/2}[s])$ are 43.40, 26.73 and 31.89. They were obtained for the cranking model, the GOA and the semi-adiabatic formula, respectively. The experimental value is 25.30. Thus, a reasonable agreement is obtained when the inertia is calculated within the GOA.

We used this model to predict the best candidates for the cluster emission processes from
Table 1. Half-lives predictions for cluster decay.

| Reaction               | log(\(T[s]\)) Experiment [44] | log(\(T[s]\)) Theory | log(\(T[s]\)) Phenomenological [25] |
|------------------------|---------------------------------|-----------------------|-------------------------------------|
| \(^{24}\)Ne from \(^{232}\)U| 21.06                           | 22.41                 | 20.40                               |
| \(^{28}\)Mg from \(^{232}\)U| 23.13                           | 24.50                 |                                     |
| \(^{22}\)Ne from \(^{232}\)U| 25.84                           | 26.70                 |                                     |
| \(^{32}\)Si from \(^{238}\)Pu| 25.3                            | 26.76                 | 23.70                               |
| \(^{28}\)Mg from \(^{238}\)Pu| 27.68                           | 24.80                 |                                     |
| \(^{30}\)Mg from \(^{238}\)Pu| 28.56                           | 24.40                 |                                     |

\(^{232}\)U and \(^{238}\)Pu. The calculated half-lives are presented in table 1 and compared with phenomenological values. The theoretical estimations agree within two orders of magnitude with experimental data. These results represent a contribution towards an unitary treatment of fission and heavy ion emission.

4. Fission

Almost two decades ago, systematic measurements were performed to determine cold fission yields of \(^{252}\)Cf [45, 46]. Our aim is to analyze these data and to show that the cold fission of \(^{252}\)Cf is strongly connected with the cold valley of the double magic isotope \(^{132}\)Sn, although the experimental cold yields have a maximum corresponding to a different charge number.

\[ \text{Figure 6. (a) Adiabatic fission barrier Figure 7. Deformation energy minimized as function of the elongation } R \text{ along the statically with respect the eccentricity of the minimal action path. (b) Cranking inertia as second fragment } \varepsilon_2 \text{ and the mass asymmetry function of } R. \text{ (c) The estimated } A_2 \text{ during parameter } \eta \text{ as function of } R \text{ and } A_2 \text{ in the second saddle region.} \]

\[ \text{Figure 8. Experimental yields in arbitrary units (dashed line), compared with renormalized theoretical penetrabilities calculated within the microscopic-macroscopic model (solid line) with respect to } A_2. \]
In the past the cold fission of $^{252}$Cf was investigated within the double folding potential method [7, 8] emphasizing the role of higher deformations. In Ref [47], based on a macroscopic model by determining the tip distances for the exit point from the barrier for ground state deformed fragments, it was predicted that the major contribution in the yield distribution corresponds to the light fission fragment $A_2 \approx 100$, contradicting experimental data showing a peak at $A_2 = 107$. The root of this discrepancy can be understood in the following.

We extend the analysis performed in Ref. [48] to a more reliable microscopic approach to estimate the fission barrier, based on a new version of the Super Asymmetric Two Center Shell Model [13]. This approach was already used to describe the fusion/fission of some superheavy elements [49, 50] and of the dynamical effects in fission [13, 17].

For comparison with experimental data, the maximal values of the independent yields for a maximum excitation energy of 7 MeV were selected from Ref. [45]. The selected channels address binary partitions characterized by the following light fragments: $^{95}$Rb, $^{96}$Rb, $^{97}$Sr, $^{98}$Sr, $^{99}$Y, $^{100}$Y, $^{101}$Zr, $^{102}$Zr, $^{103}$Nb, $^{104}$Nb, $^{105}$Mo, $^{106}$Mo, $^{107}$Tc, $^{108}$Tc, $^{109}$Mo, $^{110}$Tc, $^{111}$Ru, $^{112}$Ru, $^{114}$Rh, $^{115}$Rh, $^{116}$Pd, $^{118}$Rh, $^{119}$Pd, $^{120}$Ag, $^{121}$Cd and $^{122}$Ag.

The adiabatic barrier in the multidimensional configuration space is determined by using the least action principle. In Fig. 6, the fission barrier $V$, the effective mass $M$ and the estimated mass number $A_2$ are plotted along the fission path. The inertia is computed within the cranking approximation [42]. The second barrier top is located at $R=11$ fm and corresponds to a mass $A_2 \approx 120$. For a constant charge density, this ratio of the mass asymmetry addresses a heavy fragment with $A_1 - Z_1 = 81$ and $Z_1 = 51$, these values being close to magic numbers. Thus, the second saddle point corresponds to a partition that includes a double magic fragment. However, microscopic approaches to fission [51, 52] established that the second saddle point is asymmetrical with a value compatible with the observed mass ratio of the fragment distribution. Therefore, the dynamical saddle configuration obtained within our model is checked by minimizing statically the deformation energy around $R=11$ fm. The eccentricity $\varepsilon_1$ and the neck parameter $C$ are kept constant. The detailed region is displayed in Fig. 7 confirming that the saddle point is located at $A_2 \approx 120$ and $R \approx 11$ fm.

Now we are interested in determining the fission barriers that address all the analyzed partitions. The asymptotic deformations of the two fragments are taken from the literature [53]. Thus, the shapes of the initial nucleus up to the second barrier and those of the final fragments are known. In order to avoid a complicated determination of the minimal action path for each partition, a linear variation from initial values of the generalized coordinates $\varepsilon_i$ ($i=1,2$) and $a_1/a_2$ is postulated starting from the saddle of the second barrier configuration up to the final ones, characterizing the fragments at the end of the fission process.

With these ingredients, the calculated penetrabilities at zero excitation energies through the barrier are compared to experimental yields in Fig. 8. We implicitly assume that the penetration of the inner barrier is the same for all partitions, so that differences in the barrier transmission between channels are induced only by the external barrier.

We obtain a very good agreement between the theoretical penetrability distribution and the experimental yields for $A_2 < 110$. The maximum theoretical value is at $A_2=107$, while the maximum experimental yield is at the same value. A sudden drop of theoretical penetrabilities is theoretically obtained for channels with $A_2 > 110$. As previously noticed, this behavior is connected with the ground state shapes of fragments that become oblate for these channels. Perhaps these oblate shapes are not the best configurations during the penetration of the barrier, and the final ground state oblate configurations are obtained only after the exit from the fission barrier.

Concluding, we computed the cold fission path in the potential energy surface of $^{252}$Cf by using the two center shell model, based on the idea of the cold rearrangements of nucleons during the cold fission process. We obtained a satisfactory agreement with experimental yields, by
considering variable mass and charge asymmetry beyond the first barrier of the potential surface. We can see that the mass asymmetry changed from a symmetric to the asymmetric configuration in the vicinity of the second barrier, due to the influence of the magic numbers $Z=50$ and $N=82$, i.e. $^{120}_{47}\text{Ag}+^{132}_{51}\text{Sb}$ partition. It was shown that a good agreement with experimental data can be obtained only if the fission path proceeds through this saddle configuration. Thus, the cold fission process of $^{252}\text{Cf}$ can be called Sn-like radioactivity, similar with the Pb-like radioactivity, corresponding to various heavy cluster emission processes. We call all these processes shortly $\kappa$ (cluster) decays. The peak in the final distribution corresponds to $^{107}\text{Mo}$, due to the mass asymmetry degree of freedom, allowing a lower barrier from Sn to this nucleus.

5. Superheavy element synthesis and alpha decay

![Figure 9. Fragmentation potential for the synthesis of the element $^{296}_{116}$](image)

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![Figure 10. Potential energy for the entrance channel $^{248}\text{Cm}+^{48}\text{Ca}$ for the superheavy element synthesis.](image)

Figure 10. Potential energy for the entrance channel $^{248}\text{Cm}+^{48}\text{Ca}$ for the superheavy element synthesis.

The synthesis of superheavy elements beyond $Z = 104$, suggested by Flerov [54], was predicted within the so-called fragmentation theory in Ref. [9] by using the cold valleys in the potential energy surface between different combinations, giving the same compound nucleus. Soon it was shown in Refs. [12, 10, 11] that the most favorable combinations with $Z \geq 104$ are connected with the so-called Pb potential valley, i.e. the same valley of the heavy cluster emission [3].

Due to the double magicity of $^{48}\text{Ca}$, similar with $^{208}\text{Pb}$, in Ref. [10] it was proposed $^{48}\text{Ca}$ as a projectile on various transuranium targets. Indeed, the production of many superheavy elements with $Z \leq 118$ (corresponding to the last stable element Cf) during last three decades was mainly based on this idea [55, 56, 57, 58, 59, 60, 61, 62]. Another possibility was envisaged by using a Ni beam [63]. It was found that the fast fission channel is emphasized. Therefore, we decided to investigate the reaction $^{248}\text{Cm}+^{48}\text{Ca}$. The first step is to estimate the fragmentation potential with respect the internuclear distance for the projectile target optimum combinations. We computed the minimal value of the energy of all possible binary systems at the touching configuration that give rise to the $^{296}_{116}$ superheavy element. The two interacting nuclei were considered at their ground state deformations. Afterwards, we computed the fragmentation
potential for the compound nucleus. The results are plotted in Fig. 9 as function of the distance between the centers of the fragments and the mass asymmetry. In the lower panel, a straight curve located between $R=10-14$ fm gives the location of the touching configuration for all possible partitions. It is possible to discern several minimums: the ground state minimum of the nearly spherical compound nucleus ($A_1 - A_2 / (A_1 + A_2) \approx 0$ and $R \approx 0$ fm), an isomeric minimum for fission ($A_1 - A_2 / (A_1 + A_2) \approx 0.3$ and $R \approx 7$ fm) and a molecular minimum for alpha decay ($A_1 - A_2 / (A_1 + A_2) \approx 1$ and $R \approx 6$ fm). Several possible trajectories related to the formation of the superheavy element, the quasifission, the fission and the $\alpha$-decay are also represented with full lines. These trajectories are plotted with dashed lines in the upper panel. It is interesting to note that the $^{48}$Ca proceeds through a path that follows the minimal values of the potential energies up to the touching. When the two nuclei start to overlap, the trajectory shifts to smaller mass asymmetries to arrive in a pronounced valley. This valley starts from the ground state and reaches the molecular minimum for alpha decay. In Fig. 10, a detailed plot of the fragmentation potential in the region of the entrance channel $^{248}$Cm+$^{48}$Ca is displayed. It can be observed that, due to shell effects, the maximal values of the fragmentation potential exhibits a structure that resemble to a serie of crenels. To form a superheavy element, the trajectory must proceed through a minimal value in this region, and the mass asymmetry changes towards lower values.

**Figure 11.** Family of shapes for the alpha-decay.

**Figure 12.** (a) Potential energy for the alpha emission along the alpha valley as function of the elongation $R$. (b) Effective mass.

**Figure 13.** Shell effects as function of $R$. 
The synthesis of superheavy elements and the fission was treated extensively in the literature within Hartree-Fock methods [64] and macroscopic-microscopic ones [65, 66]. It can be deduced from the potential landscape that the fission from the ground state of the compound nucleus proceeds through the deeper valley that belongs to Sn. The fission with Pb is obtained in peripheral reactions associated to quasi-fission. However, only few attempts exist for alpha-decay as superasymmetric fission [67, 68]. Most approaches compute the preformation probabilities and the penetration of the Coulomb barrier [69, 70, 71, 72, 73]. Therefore, we will focus on the alpha-decay valley. The shapes along the path denoted with the symbol $\alpha$ in Fig. 9 are plotted in Fig. 11. In the mentioned molecular minimum at $R \approx 6$ fm, an alpha particle begins to be formed on the surface of the daughter spherical nucleus. The alpha-decay barrier and the effective mass along the trajectory represented in Fig. 9 are displayed in Fig. 12. The origin of the molecular minimum can be understand if the shell effects are investigated. In Fig. 13 the Strutinsky corrections are plotted. A minimal value is obtained for the spherical parent nucleus. The values of the shell effects increase with the deformation. Once the alpha particle is formed on the surface, the nuclear system feels again a strong shell effect that is due to the spherical daughter, leading to a new energy minimum. Therefore, a molecular configuration is obtained. For larger $R$ internuclear distances, the macroscopic part of the total energy increases and a barrier is created. The $Q_\alpha$-value obtained with our model in 9.95 MeV. We computed phenomenologically the partial half-lifes and compared them with known experimental values obtained for other superheavy isotopes in Table 2. The predicted values for $^{296}\text{116}$ are well integrated in the systematics.

Table 2. Half-lives and $Q$-values of superheavy alpha decay.

| $Q_\alpha$ (MeV) | $T_\alpha$ (ms) |
|------------------|----------------|
| 11               | 15             |
| 10.89            | 6.3            |
| 10.80            | 18             |
| 10.67            | 53             |
| 9.95             | 206            |

The values of the shell effects increase with the deformation. Once the alpha particle is formed on the surface, the nuclear system feels again a strong shell effect that is due to the spherical daughter, leading to a new energy minimum. Therefore, a molecular configuration is obtained. For larger $R$ internuclear distances, the macroscopic part of the total energy increases and a barrier is created. The $Q_\alpha$-value obtained with our model in 9.95 MeV. We computed phenomenologically the partial half-lifes and compared them with known experimental values obtained for other superheavy isotopes in Table 2. The predicted values for $^{296}\text{116}$ are well integrated in the systematics.

Figure 14. Selected trajectories between the isomeric minimum and the scission as function of the elongation and the mass asymmetry.

Figure 15. Deformation energy $V$ and effective mass $B$ in the cranking approximation for the trajectories plotted in Fig. 14.
In Ref. [74] the experimental mass distribution obtained in the reaction $^{48}\text{Ca}+^{248}\text{Cm}$ was measured at an excitation energy of 33 MeV. So, we check if the model can bring some information in this context. We constructed the possible paths that start from the isomeric minimum and connect the touching configurations plotted in Fig. 9. The paths are displayed in Fig. 14. We calculated the potential energy and the effective mass using the cranking approximation for these trajectories. Calculating the barrier, we take into account the fact that the shell effects vanish following an exponential dependence $\delta E \exp(-T^2/T_0^2)$. It is known that $T_0=1.5$ MeV and $T \approx 1$ MeV for an excitation energy of 33 MeV. The driving potentials and the effective masses are plotted in Fig. 15. The normalized yields for all these partitions were determined using the WKB method. The result is plotted in Fig. 16. So, the model evidence that a large yield in the mass distribution can be obtained around $A_2 \approx 125$ if the system reaches the isomeric configuration. A such peak was found also in the experimental data [74].

Acknowledgments

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