Cold Valleys in Cluster Radioactivity, Cold Fission and Formation of Heavy and Superheavy Nuclei

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Abstract

We present an investigation of the cluster radioactivity, cold spontaneous fission and fusion processes using the concept of cold valley. The minima in the calculated driving potential account in the great majority of cases for the observed cluster radioactivity channels of actinides, cold fission yields of ^{252}Cf and quasi-fission mass distributions in the synthesis of superheavy nuclei.

1 Introduction

The possibility to have a cluster process is related to its exotermicity:

$$Q = M(A,Z) - M(A_1,Z_1) - M(A_2,Z_2) > 0$$

Here, $M(A_i,Z_i)$ are the atomic masses. For the elements found in the second half of the periodic system, which have average binding energy per nucleon smaller than the lightest elements, this condition is fulfilled for a large range of nuclear decays. However in the great majority of cases the possibility of decay is hindered by the small barrier penetrability. This quantity reaches large values only in two cases: α-decay and spontaneous fission of heavy nuclei. An exception is given by the high penetrabilities of the daughter nuclei ^{14}C, ^{24}Ne, etc., when the complementary nucleus is close to the double magic ^{208}Pb, values which can be larger than those corresponding to the α-decay.

The phenomenon of cluster radioactivity was predicted at the end of 1970's decade by Săndulescu et al. [1] and confirmed a few years later by Rose and Jones [2].

The concept of *Cold Valley* was introduced in relation to the structure of minima in the so-callled driving potential. The driving potential is defined as the difference between the interaction potential and the decay energy of the reaction

$$V_{\rm driv} = V(R, Z_1, A_1, \{\beta^{(1)}\}_{2,3,4}, \{\beta^{(2)}\}_{2,3,4}, \omega_1, \omega_2) + B_1 + B_2 - B_{\rm CN} \quad (1)$$

In the above formula B_1, B_2 and B_{CN} are the binding energies of the cluster, daughter and mother nucleus. The driving potential is also depending on the charge Z_1, mass A_1, the distance between the centres-of-masses of the two nuclei R, mutual orientations $\omega_{1,2}$ and the quadrupole(β_2), octupole (β_3) and hexadecupole deformations (β_4) through the heavy ion potential V defined as

$$V(\boldsymbol{R}) = \sum_{\lambda_i \mu} V_{\lambda_1\ \lambda_2\ \lambda_3}^{\ \mu\ -\mu\ 0}(R) \cos\mu(\alpha_2 - \alpha_1) d^{\lambda_1}_{\mu 0}(\beta_1) d^{\ \lambda_2}_{-\mu 0}(\beta_2) \quad (2)$$

The *Ansatz* (1) corresponds to the s-channel, i.e. relative angular momentum $l = 0$.

A long time ago Săndulescu *et al.* investigated the occurence of fusion valleys as doorways to the synthesis of superheavy elements [3, 4]. The basic idea of this approach was to calculate the driving potential of a given compound nucleus for all possible projectile-target combinations as $\eta_Z = (Z_1 - Z_2)/(Z_1 + Z_2)$ at the touching point R_c, i.e. the point where the assumed spherical fragments are coming in contact and they interact only by means of the Coulomb force. The charges of the target and the projectile were determined by requiring for a fixed η that the potential $V(R, \eta, \eta_Z)$ attains a minimum in the η_Z direction, i.e. for every fixed mass pair (A_1, A_2) a single pair of charges is determined among all possible combinations. Next, minima of the potential in the two-dimensional (R, η) landscape were searched. From here it was inferred a criterion for cold fusion, i.e. the deep minima of the driving potential are corresponding to the projectile-target combinations where the compound nucleus has a minimum of excitation and will de-excite to the ground-state with the emission of a couple of neutrons. The occurence of the mass-asymmetry valleys is due to the shell effects. It was advocated in ref. [5], using the frame of the fragmentation theory, that due to the existence of different mass-asymmetry valleys for the same compound system, a new, highly asymmetric fission mode appears in which one of the fragments is close to the double magic nucleus ^{208}Pb. This new type of decay is nothing else than the cluster radioactivity.

Formation of cold fusion valleys is strongly dependent on which shape is assumed for the incoming projectile and target. For the case when one assumes spherical shapes, as actually was done in [3, 4], the most important valleys found are centered around the double magic nuclei ^{132}Sn (a weak asymmetric valley) and ^{208}Pb (a very asymmetric valley). The introduction of deformation modifies the valley structure of the driving potential. The valleys occuring when nuclei are assumed to be spherical are weakened or even washed out when the experimental or calculated ground state deformations of the nuclei are switched on. Instead, other valleys are substantiated, depending crucially on the mutual orientation of the nuclei as it was proved recently [6].

The possible formation of superheavy nuclei with $Z = 102-122$ in reactions with ^{22}Ne, ^{26}Mg, ^{48}Ca, ^{58}Fe and ^{86}Kr ions at energies near and below the Coulomb barrier has been studied at FLNR-JINR Dubna [7]. Along with other relevant quantities the mass distributions of fission fragments was determined. The nuclei ^{256}No, ^{286}112, ^{292}114 and ^{296}116 have been produced in reactions with ^{48}Ca at the same excitation energy E^*=33 MeV. The main pecularity of the data is that the quasi-fission mechanism is dominating in the case of the decay of the nuclei ^{286}112, ^{292}114 and ^{296}116. The mass distribution of quasi-fission products was found to be asymmetric with the peak of the lighter fragment located at $A_1 \approx 80$ for the superheavy nucleus ^{286}112, ≈ 84 for the superheavy nucleus ^{292}114 and ≈ 86 for ^{296}116. The authors also claimed that despite the dominating contribution of the quasi-fission process, in the symmetric region of fission fragment masses ($A_{CN}/2 \pm 20$) the process of compound nucleus(CN) fission is prevailing. Indeed, in the light fragment mass-distribution a small and weakly asymmetric bump occurs (see Fig.1 of [7]). For all three superheavy nuclei the light fragment mass is centered around $A_1 = 132 - 134$. The nucleus ^{296}116 presents another pecularity : an additional bump centered on $A_1 = A_{CN}/2$, i.e. a totally symmetric distribution of decay products.

2 Cluster Radioactivity

2.1 Cluster Radioactivity in Radium and actinides

The cold valleys in the case of CR are strongly dependent on the deformation of the clusters and daughter nuclei. As shown in [8] quadrupole and hexadecupole deformations, especially of the daughter could play an important role in reaching th experimental values of the decay rate.

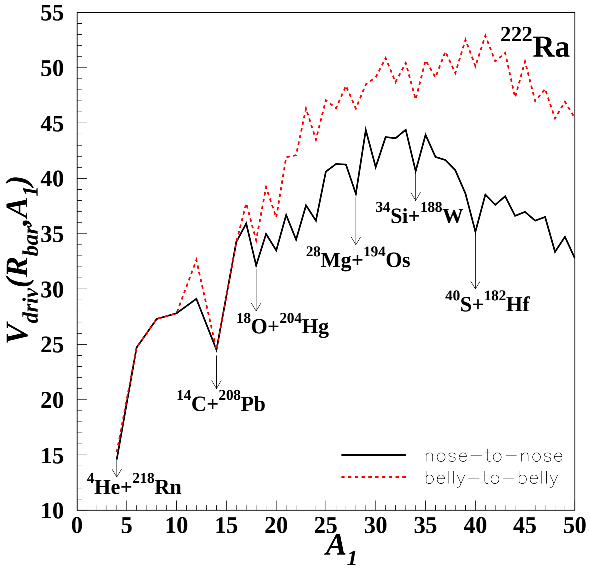

Figure 1: Comparison between the $p-p$ and the $e-e$ oriented driving potentials for the mother nucleus ^{222}Ra.

In the case of ^{222}Ra the most pronounced valley corresponds to the case ^{14}C+^{208}Pb, both nuclei being spherical (see Fig.1). However beyond the cluster mass number $A_1 = 16$ the deformation cause the deviation of the $e-e$ curve from the $p-p$ one. The existence of the valley corresponding to the emission of the cluster ^{14}C and the double-magic daughter nucleus ^{208}Pb is confirmed in experiment [9]. Simmilarly happens to the Radium isotopes 221, 223, 224 and 226.

At the other extreme of studieded cluster emitters, i.e. ^{242}Cm, we are inferring from Fig.2 that the cluster valleys are shifted to larger cluster masses. In the case of $p-p$ configuration the deepest valley corresponds to the splitting ^{40}S+^{202}Hg and the second important valley is the recently reported splitting ^{34}Si+^{208}Pb [10]. This last valley is less favourable in the frame of the cold valley picture of decay because of the sphericity of both cluster, ^{34}S, and daughter nucleus ^{208}Pb which rises sensitively the barrier compared to the case of the prolate (β_2 =0.254) cluster, ^{40}S, emitted at the equator of the oblate($\beta_2 \approx$-0.09) mother nucleus ^{202}Hg. As one can see from Fig.2 if the cluster ^{40}Si would be emitted at the pole of ^{202}Hg, with its symmetry axis perpendicular to the axis joining the centers of the two nuclei($e-e$ configuration), then the corresponding valley would lose completely in importance and the only observed cluster decay should be ^{34}Si+^{208}Pb. Of course that the spectroscopic factors may invert the importance of these two valleys by favouring the clusterization of the double magic ^{208}Pb. It is therefore necessary that the investigation of the cluster emission in ^{242}Cm looks for both splittings and not only of ^{34}Si+^{208}Pb.

The cold valley concept applied to cluster radioactivity is also giving a satisfactory explanation of the observed splittings for different isotopes of Th and U.

In the case of Th, where cluster radioactivity has been observed, we draw in Fig3 the driving potential in $p-p$ configuration for four of its even-even isotopes. For all 4 investigated isotopes we indicated the corresponding α-decay which is the predominant cluster emission mode. For ^{226}Th the first two deepest cluster valleys are given by the combinations ^{14}C+^{212}Po and ^{18}O+^{208}Pb, and are actually experimentally confirmed cluster emission channels. We notice here that ^{226}Th is the only isotope of Th where one observes the emission of ^{14}C. For ^{228}Th the deepest minimum is encountered for the splitting ^{20}O+^{208}Pb which is the only observed cluster emission channel up to now [11]. The splittings, ^{14}C+^{214}Po and ^{22}Ne+^{206}Hg, although not observed, are also corresponding to deep valeys, and we put a question mark(?) to stress

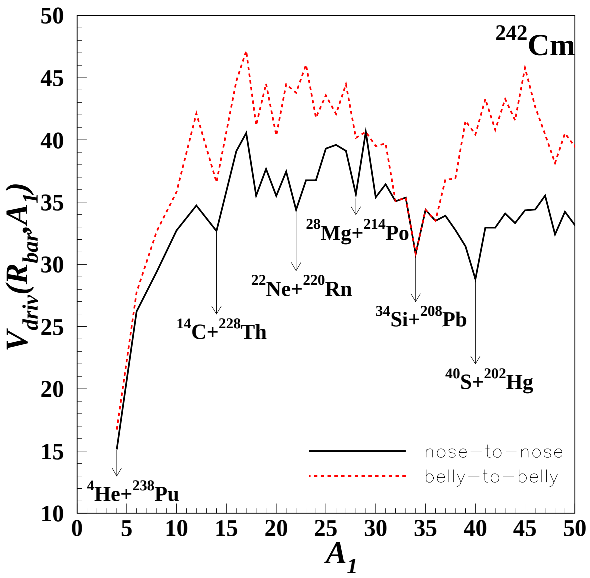

Figure 2: Same as in Fig.1 for ^{242}Cm.

that their possible observation in the future should not be excluded. Moving two neutrons further at ^{230}Th we find that the deepest minimum moved two cluster charge numbers further and corresponds to the observed splitting ^{24}Ne+^{206}Hg [12]. However the neighbouring splittings ^{20}O+^{210}Pb and ^{22}O+^{208}Pb have also close values of the minima but they were not recorded in experiment. Also the ^{14}C emission channel should be favorable for ^{230}Th according to the cold valley approach but no experimental evidence was provided to the date. The last Thorium's isotope, ^{232}Th, is the most rich in comparable deep valleys. If we leave aside the valley of ^{14}C+^{218}Po, then the deepest one is the experimentally confirmed splitting ^{24}Ne+^{208}Hg. Also observed is the neighbouring splitting ^{26}Ne+^{206}Hg. The other minima corresponding to the emission of ^{20,22}O and ^{28,30}Mg are not yet experimentally registred.

One may conclude that the reason of the decrease in importance of ^{14}C emission when moving from ^{226}Th to ^{232}Th is due to the corresponding daughter nucleus, i.e. Polonium. By adding more and more neutrons to Po, we get away from the neutron magic shell and most likely the spectroscopic factors are decreasing. The probability of reaching different cold valleys from the ground state (cluster preformation) should come into play.

If one cheks the deformations of the emitted clusters one finds that the deepest valleys for ^{228,230,232}Th are corresponding to high quadrupole and/or hexadecupole distortions which indicate a possible subcluster strucure, as is the case of ^{20}O+^{208}Pb, ^{24}Ne+^{206}Hg and ^{24}Ne+^{208}Hg.

It is also worthwile to notice for the case of Thorium that the cluster radioactivity cold valleys are grouped in "super-valleys" which are broader when moving to larger neutron numer of the mother nucles. For the emitter ^{228}Th we have a "super-valley" which ranges from cluster mass A_1=18 to 24 whereas for ^{232}Th it ranges A_1=18 to 30.

For Uranium we take as well 4 even-even isotopes which are known cluster emitters(see Fig.4). The mother nucleus ^{230}U has a very pronounced valley centered on the splitting ^{22}Ne+^{208}Pb, which is actually the only case of observed CR for this nucleus. The reason for obtaining such a deep minimum is due not only to the double magicity of the daughter nucleus, but also to the large cluster's quadrupole (β_2=0.326) and especially hexadecupole (β_4=0.225) deformations. For the next isotope, ^{232}U, the minimum shifts on the cluster ^{24}Ne, which is oblate(β_2 =-0.215), and in order to overcome a smaller barrier, it should be emitted with its symmetry axis perpendicular to the fission axis. Adding two more neutrons, we obtain two comparable minima for ^{24}Ne+^{210}Pb and ^{28}Mg+^{206}Hg. In between we have the combination ^{26}Ne+^{208}Pb which although is not giving a minimum in the driving potential it shares the same

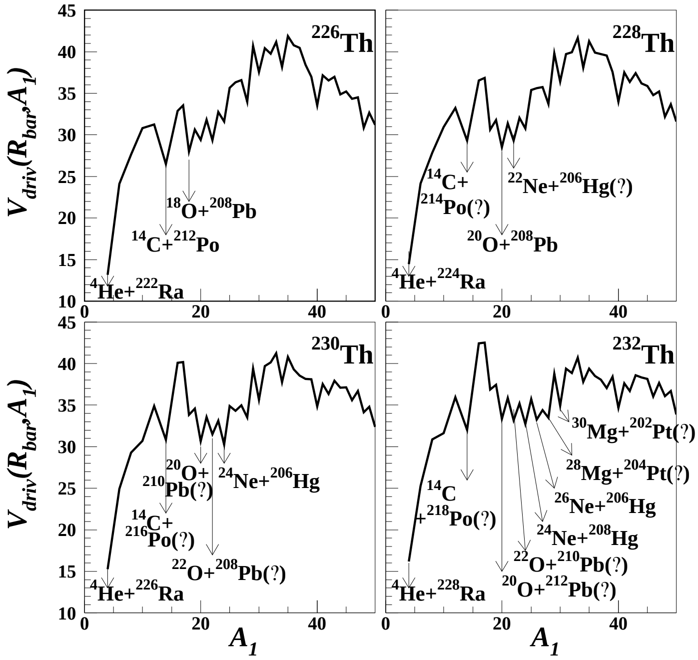

Figure 3: The driving potential of ^{226,228,230,232}Th in the $p-p$ configuration. The question mark (?) indicates that the given combination, although not observed in experiment, has a favorable position in the cold valley.

valley as ^{28}Mg+^{206}Hg. It is important to stress that the clusters of all these three splittings, which were also reported by experiment [13], have large quadrupole deformations and especially hexadecupole deformations. The last investigated isotope of Uranium, ^{236}U, has less pronouned minima, but like in the previous cases, two cluster decay channels were observed in experiment, corresponding to two deep minima, ^{28}Mg+^{208}Hg ^{30}Mg+^{206}Hg. In the first case we deal with a prolate cluster and in the second with an oblate cluster, both endowed also with a sensitive negative hexadecupole deformation.

Actually for the two isotopes ^{234,236}U we obtained that the deepest minima corresponds to ^{14}C because it enters in combination with Rn isotopes which are increasingly deformed. However the emission of ^{14}C was not observed in any isotope of Uranium which means that we deal with a hindrance in the preformation of such combinations, the main responsible, in our opinion, being the daughter nucleus which preferes to acquire a spherical or almost spherical shape, in the neighbourhood of the double-magic ^{208}Pb. Everything is going on as if the heavier(daughter) nucleus, is transferring nucleons, or even clusters, untill it reaches the valley containing spherical or almost spherical isotopes of Pb and Hg. In the same time the cluster is acquiring quadrupole and hexadecupole deformation mainly, having in almost cases a clear clusterized structure which indicates that it is possible that a sequencial transfer of alphas is taking place. For the case of the cluster ^{22}Ne we can enview for example the transfer of two alphas to the spherical ^{14}C which are leading to a large hexadecupole deformations with the two alphas arranged at the opposite ends of ^{14}C.

For the investigated isotopes of Uranium we should also remark, when increasing the number of neutrons, the gradual weakening of the cluster valley and the increase in importance of the minimum corresponding to the splitting ^{40}S+$^{A-40}$Os. However, we expect in this case like for ^{14}C a hindrance in the preformation.

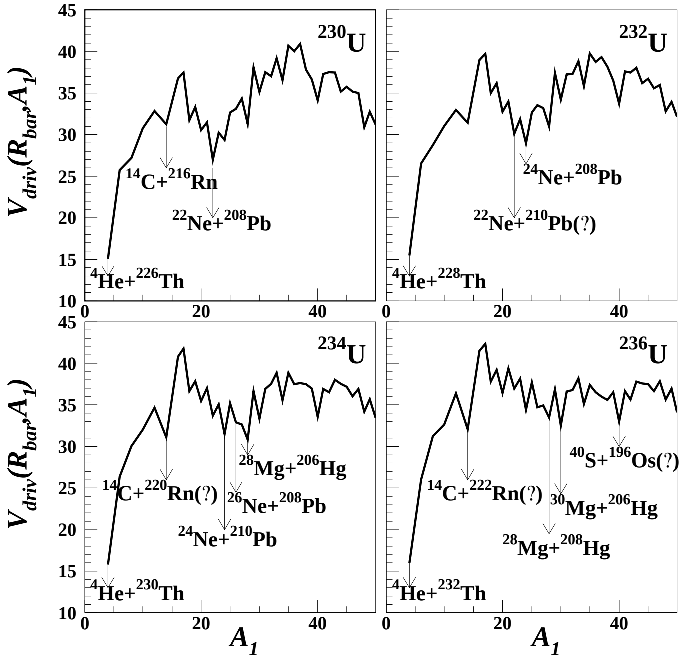

Figure 4: Same as in Fig.3 for ^{230,232,234,236}U.

3 Spontaneous Cold Fission of ^{252}Cf

In cold fission only one orientation is singled-out and this is the configuration in which the tips(poles) of the prolate deformed nuclei are aligned along the nuclei symmetry axes. For the pole-pole($p-p$) configuration ($\theta_{1,2}=0$, $\phi_{1,2}=0$) the driving potential $V_{\rm driv}$ is minimized along η_Z at $R=R_{\rm bar}$. At the other extreme the equator-equator ($e-e$) orientation is encountered when the bellys of the two fragments are aligned perpendicular to the common symmetry axes.

In Fig.5 a cut along the mass asymmetry coordinate of the driving potential is represented. The solid curve corresponds to the $p-p$ scenario whereas the dashed one to the $e-e$ one. The difference are tremendous. The Ca-valley (Pb is inside this valley) is very pronounced for the $e-e$ orientation and very shallow for the $p-p$ one. For fission the most relevant valley in the $e-e$ configuration is the quasi-symmetric one centered on a Sn isotope (other than the double magic ^{132}Sn) whereas for for the $p-p$ configuration the quasi-symmetric valley, this time centered on ^{132}Sn, is overtaken by a broader, deeper and more asymmetric valley which contains the splittings Sr+Nd, Zr+Ce and Mo+Ba.

In [15] we explained in a $p-p$ scenario the yields structure observed in the experiment [14]. In the region of small yields the fragments are spherical or nearly spherical (with a small prolate deformation) and have high-Q values. This is the region centered on Sn isotopes. In the region of higher yields the quadrupole and hexadecupole deformations especially are determining a drastic lowering of the barrier. These calculations are firmly excluding other orientations, such as $e-e$ for example.

4 Formation and Decay of Superheavy Elements

In Fig.6 a cut along the mass-asymmetry coordinate of the driving potential of the superheavy nucleus ^{112}286 is given. From the inspection of this figure we remark the differences in the driving potential when the target and the projectile are colliding with different orientations. The Ca-valley (with heavy partner U) is more pronounced when the fragments are coming in contact in $e-e$ or $e-c$ orientations. The valleys corresponding to the cluster radioactivity

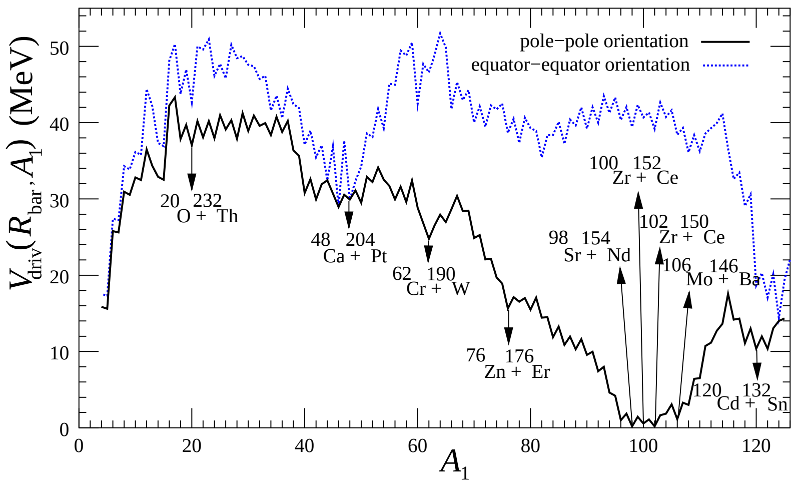

Figure 5: Driving potential of ^{252}Cf as a function of the projectile mass A_1 for the pole-pole orientation (continous line) and for the equator-equator orientation (dashed lines) at the barriers value of the R coordinate.

(superasymmetric valleys) are occuring with some differences: for $p-p$, the most pronounced are centered on ^{14}C, ^{22}Ne and ^{28}Mg, whereas for $e-c$ we remark ^{24}Ne, ^{30}Mg and ^{34}Si.

For $p-p$ oriented fragments the valley centered on ^{62}Cr is separated by a high barrier from what we call the *Pb*-valley, centered on ^{76}Zn and ^{80}Ge, and obviously for such configurations the tendency of the initial cold strong asymmetric system to move in the symmetric direction, before undergoing quasi-fission, is hindered.

The $p-e$ orientation presents features common to the $p-p$ but also to the $e-e$ and $e-c$ orientations. Similar to the $p-p$ case, the valley for Ca is less pronounced and the Mo-valley is broader and the target ^{96}Sr will give a minimum in the potential for this orientation.

For the $e-e$ orientation we deal with three important valleys: the Ca-valley, the *Pb*-valley, centered on ^{80}Ge and the Sn-valley centered mainly on ^{134}Te. As can be seen in Fig.7 this last valley is common also to the $p-p$ orientation and is assigned in the FLNR-Dubna experiment to events emerging from the fusion-fission process. On the other hand between the *Pb*-valley and the Sn-valley a huge barrier is showing up which determines an even stronger hindrance to symmetric quasi-fission compared to $p-p$ orientation. The *Pb*-valley, which is very pronounced, is centered on the same light fragment mass numbers as the mass distribution reported in [7] and assigned to quasi-fission.

Making the association $p-p$ *deepest valley* $\rightarrow$ *maximum cold fission yield*, which seems to be valide in the case of ^{252}Cf, we infer that the occurence of light fragments with masses around A_1=132-134 can be due also to the fission of the compound superheavy nucleus ^{286}112. On the other hand the quasi-fission mass distribution seems to be explained by means of the prevailance of an orientation close to the $e-e$ one. In the entrance channel we have a stable Ca-valley in the $e-e$ scenario. Since the closest valley from this one is the *Pb*-valley, then we expect the highest quasi-fission yields to correspond to a region around $A_1 \approx 80$. Since the excitation energy produced in the reaction ($E^* \approx 33$ MeV) is large enough such that a part of the flux moving in the symmetric region reaches the weak asymmetric valley of Sn and subsequently decay, we do not exclude that a part of the bump recorded in experiment, and assigned to the fusion-fission, is contaminated to a certain extent with quasi-fission events.

For the driving potential of the superheavy nucleus ^{292}114 (see Fig.7) we remark a similar structure of $p-p$ and $e-e$ valleys as for ^{286}112. However the minimum of the weak-asymmetric $p-p$ valley is displaced now to higher symmetry, i.e. on ^{140}Xe. The second asymmetric $p-p$ valley in importance is centered on ^{106}Mo (which contains also Zr isotopes) and arises only for $p-p$ orientations, and the third one corresponds to the deep Cr minimum (in combination with

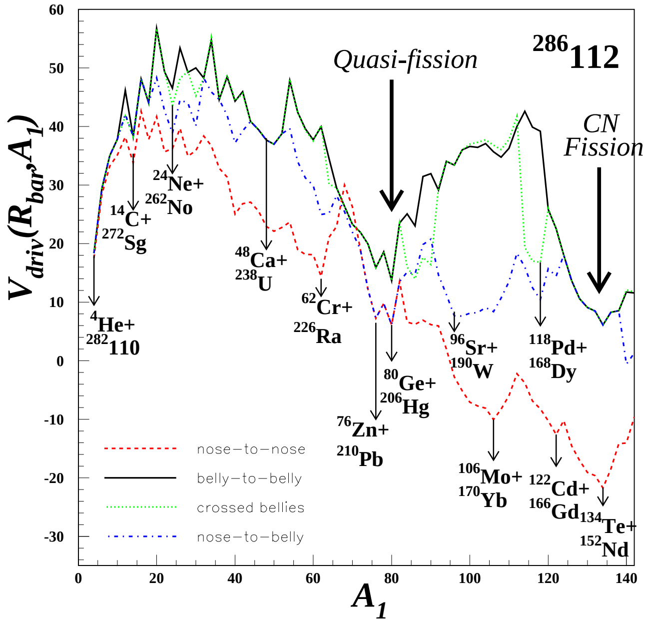

Figure 6: Cold Valleys of $^{286}112$ for different orientations of the target and the projectile. With thick arrows we indicated the regions were the experiment [7] claims quasi-fission and fusion-fission events.

U). The most important two valleys in the $e-e$ orientation are the Pb-like and is centered on the very deep minima ^{84}Ge+^{208}Pb and ^{86}Se+^{206}Hg and weak asymmetric valley centered on A_1=134 like in the previous case. One should note that the occurence of this deep valley is motivated by the transition to oblate deformations of the target as has been noted in [6] and also that it is displaced to higher projectile masses as happens also for the quasi-fission peak, which is centered on A_1=84 [7].

In the case of the superheavy nucleus $^{296}116$ the driving potential is qualitatively very similar to the one corresponding to $^{286}112$ (see Fig.8). There are however some differences. In the $e-e$ orientation the Pb-valley is centered this time around $A_1 \approx 86$ whereas the Sn-valley remains almost unchanged. Instead the deepest $p-p$ valley is now centered on the symmetric region and the fragmentations with light mass number 132-134 are contributing, at least for low excitation energy, with yields smaller than for $^{286}112$.

The additional bump recorded in experiment for $^{296}116$ and centered on the symmetric region is, in the light of our calculations, a good candidate for fusion-fission signature. However the weak asymmetric yields centered on $A_1 \approx$132-134 could be, like in the case of $^{286}112$, an admixture of quasi-fission and fusion-fission events.

5 Conclusions

The various predictions of cluster emission likelyhood in actindes made in this paper have to be completed also by a study of preformation factors. In order to determine the decay rates it is not enough to have the relative barrier heights. According to the empirical law of Blendowske and Walliser [16], the logarithm of the spectroscopic factor has a linear decrease with the mass of the emitted cluster. To the date the experimental and phenomenological theoretical models [17] are pointing to a deviation from this line beyond ^{34}Si. In any case the spectroscopic factor decrease sensitively with mass number and this is one of the reason why for mother nuclei larger that Cm, when the most favorable channels predicted by the cold valley are shifted beyond cluster mass number 40 are not observed, although they enter in combination with .

In the binary cold spontaneous fission the decaying system tends to choose an optimum path in which the emerging fragments are oriented pole-to-pole. Only for such a configuration the

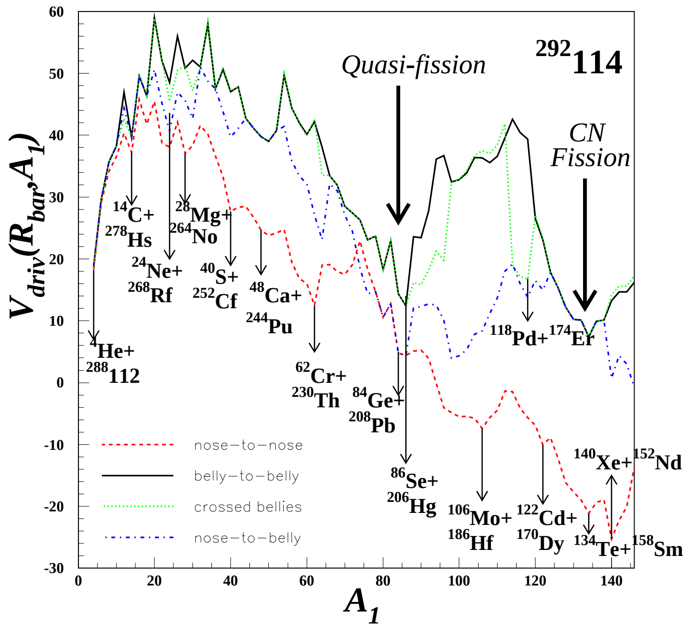

Figure 7: Same as in Fig.6 but for $^{292}114$.

mass distribution observed in experiment can be understand. Other orientations are apparently precluded in cold spontaneous fission and should be regarded as quasi-fission doorways in the synthesis of superheavy elements by cold fusion.

For the superheavy nuclei studied in this paper, the $p-p$ orientation produces no valley in the mass region where high quasi-fission yields are observed, confirming thus the conclusion of the authors of [7]. On the other hand the bumps observed in the weak asymmetric region with light fragment masses A_1 ≈132-134 are not necessarily entirely due to the decay of the CN, a contamination with quasi-fission fragments being possible according to our opinion.

We conclude that the inclusion of excitation energy around 33 MeV, as occurs in the synthesis reactions with ^{48}Ca, is not diminishing the importance of the main $p-p$ cold valley in favor of other valleys. Obviously, at a comparable excitation energy the fission mass distribution obtained in the experiment [7] does not follow the pattern of the $p-p$ valleys. Our investigation leads to the conclusion that the claimed fusion-fission events in the symmetric mass region are belonging, at least partially, to the quasi-fisson process, when configurations others than the $p-p$ one are important. Then, if nuclei like Mo and Zr would be recorded in the mass distribution yields, and actually a small bump of Mo is measured following the synthesis reaction with ^{48}Ca [18], this could indicate in our opinion the recording of compound nucleus fission events. The small bump observed in the mass-symmetric region for ^{296}116 is likely to be due to the fission of the compound nucleus.

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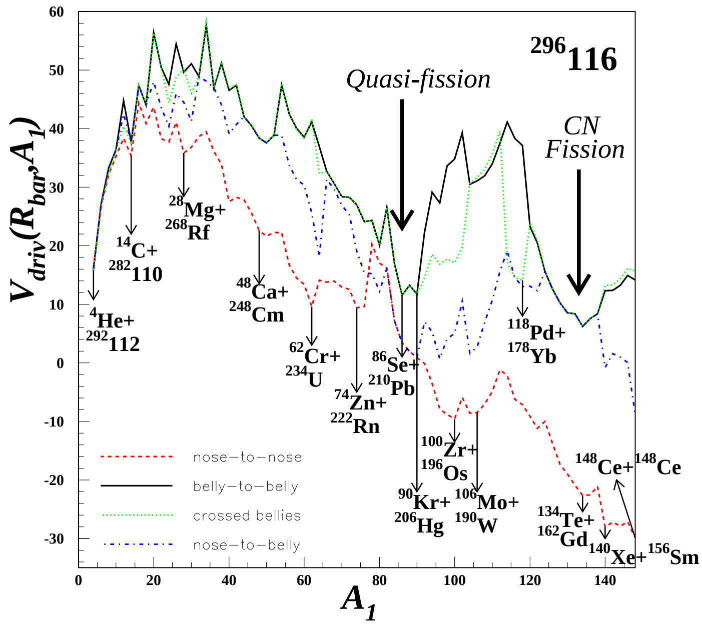

Figure 8: Same as in Fig.6 but for ^{296}116.

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