Peculiarities of clusters formation in true ternary fission of ^{252}Cf and ^{236}U*

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(Dated: Today)

Abstract

The existence of a new type of cluster decay called "collinear cluster tri-partition" (CCT) is discussed by an analysis of the landscape of the potential energy surface (PES). The total energy of the ternary system is found as a sum the binding energies of fragments and nucleus-nucleus interaction between them. The pre-scission state of the ternary system is assumed to be arranged as a chain of the three fragments along a straight line. Minima and valleys of the PES are determined by variation of the proton and neutron distributions between them. Pre-scission prompt emission of neutrons is assumed and PES is calculated for the cases of emission of 2—4 neutrons. The presence of the valley corresponding to the formation of the isotopes of Sn with masses A=130—136 is inherent for all PES calculated for CCT for spontaneous fission of ^{252}Cf and fission induced by neutrons of ^{235}U. There are local minima indicating the formation of Ca, Fe, Ni, Ge and Se isotopes having magic proton or/and neutron numbers, such as 20, 28, and 50. The analysis shows that the experimentally observed ^{68}Ni is formed as the edge fragment of the ternary system connecting to Sn by Si and Ca isotopes at fission of ^{236}U and ^{252}Cf, respectively.

PACS numbers:

I. INTRODUCTION

The role of the nuclear shell structure in the formation of fission products appears in the observed asymmetric mass distributions depending on the excitation energy of the system undergoing in fission. In Refs.[1, 2] the yields of very asymmetric Ni and even Fe isotopes in the cold fission of ^{236}U analyzed as a fission channel around magic proton number Z =28 with the mass number 70 are discussed. The yield of the very asymmetric products was not observed in coincidence with conjugate heavy product and, therefore, the authors of the mentioned paper did not consider the possibility of those fission fragments as CCT products. This scenario of the yield of the very asymmetric products of the Ni isotopes with A=68 and 70 was studied in the $^{235}\text{U}(n_{\text{th}}, f)$ reaction and at spontaneous fission of ^{252}Cf by the FOBOS collaboration [3–7].

The observation of two and more nuclear fission products in the fission of ^{235}U with thermal neutrons and in the spontaneous fission of ^{252}Cf has opened a new area of study in the nuclear reactions. This phenomenon is connected with the appearance of cluster states in nuclear reactions and it is the manifestation of the shell structure which is responsible for the production of isotopes with magic numbers of neutrons and protons. When a massive nucleus loses its stability and goes to fission, first of all clusters are formed as future fragments having the neutron or/and proton number nearby the magic numbers 28, 50, 82 and 126. In the case of ternary fission one observes fragments with the charge number 28 and 50. This kind of ternary fission is called as the true ternary fission, which produces the fragments with comparable masses in difference from emission of alpha-particles or light charged nuclei $A < 16$ accompanying binary fission [8].

The probability of multicluster fission of the U, Pu and Cf isotopes are less than one percent of the corresponding cross sections of binary fission. The cross section of CCT is comparable with one of the well-known ternary fission with the emission of an alpha-particle [9, 10]. The emission of the light charged particles from the neck region on the plane perpendicular to the fission axis is main characteristic of the ternary fission. Although the light charged particles can be emitted in direction close to the momentum of the fission fragments [9]. The probability of the yield of the light charged particles heavier than alpha-particle decreases by increasing their charge and mass numbers. The observed yield of the group of the neutron rich isotopes of Ni and Ge in coincidence with the heavy fragment with mass numbers A=136—140 is an unexpected phenomenon. Therefore, theoretical interpretation of these processes is required for a full understanding of the mechanism. The ternary fission fragmentation of ^{252}Cf for all possible third fragments using the recently proposed three-cluster model [11] was studied in ref.[12]. The authors concluded that the theoretical relative yields imply the emission of the ^{14}C, ^{34,36,38}Si, ^{46,48}Ar, and ^{48,50}Ca as the most probably third particle in the spontaneous ternary fission of ^{252}Cf.

The results being discussed in this paper are based on two different experiments with binary coincidences of fission fragments and measurements of the masses and energies of the two fragments [4]. In two other experiments [7] for the study of spontaneous ternary fission of ^{252}Cf, events in coincidence with neutrons are reported. The prompt emission of neutrons from the neck region (scission neutron source) is inherent to the spontaneous fission of the actinides [13]. Authors of Ref.[7] reported about the two registered products of CCT in coincidence with the neutron multiplicity emitted from the neck region of fissioning nucleus. The "neutron belt" was assembled in a plane perpendicular to the symmetry axis of the spectrometer, which serves as the mean fission axis at the same time. The third fragment of CCT was missed by registration setup.

The relatively high yield of the CCT-effect (more than 10^{-3}/binary fission) is likely due to the collective motion through very elongated (hyper-deformed) pre-scission shapes and a large phase space covering a larger number of mass partitions with high Q-values [14]. The formation of the third cluster occurs in the neck region between the main binary fragments during the pre-scission stage of the splitting. The fact that the formation of binary fragments is the main channel is seen from all PES figures as a wide and deep valley at $Z_3 < 2$, where Z_3 is a charge number of the middle cluster.

The case of alpha-cluster formation has been well studied both experimentally and theoretically. But the CCT process is needed to be studied in detail taking into account conditions leading to the formation of ternary system with comparable masses and dynamics of the rupture of two necks connecting border fragments to the middle nucleus.

The aim of the calculation is to explain the possibility of the population of the Ni isotopes as a fragment in ternary system. The relatively large cross section of the yield of fission products with the given charge and mass numbers is the consequence of the population of the

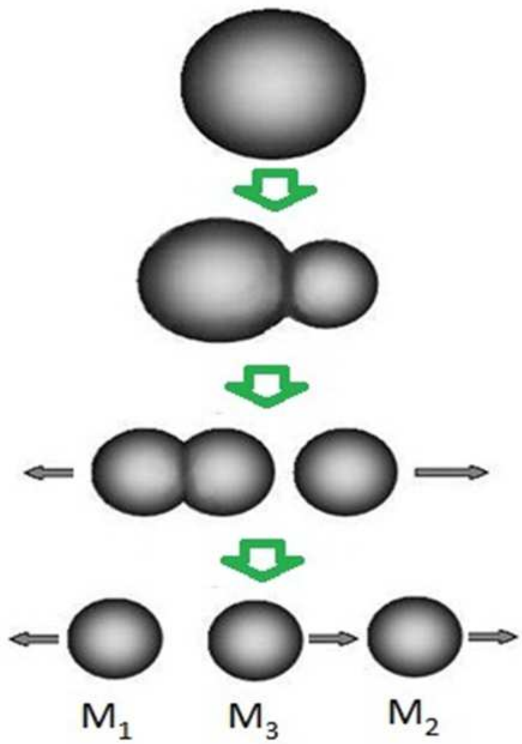

FIG. 1: (Color online) The sketch of the collinear cluster tripartition as two sequential fission process.

states corresponding the minima of PES. This is a necessary condition and the cluster can be emitted from the system if it is able to overcome the pre-scission barrier of the nucleus-nucleus interactions connecting the ternary system. Therefore, at first, it is important to analyze the PES landscape calculated for the considered fissioning system.

II. OUTLINE OF THEORETICAL APPROACH

The calculations are performed basing on an assumption that the third cluster has appeared in the neck region of the binary fragments due to fluctuation of the proton and neutron transfer between them during descent from the saddle point before scission point. The difference between the total energy of the ternary system and fissioning nucleus is used as the PES which shows an effect of the nuclear shell structure on the nascent fission products. The PES is found as a sum the energy balance of the interacting fragments and nucleus-nucleus interaction between them

$$
\begin{aligned}
& U(R_1, R_2, Z_1, Z_3, A_1, A_3) = V_1(R_1, Z_1, Z_3, A_1, A_3) \\
& +V_2(R_2, Z_2, Z_3, A_2, A_3) + V_{12}^{(Coul)}(Z_1, Z_2, R_1 + R_2) \\
& +Q_{ggg}. \qquad (1)
\end{aligned}
$$

Here $Q_{\rm ggg} = B_1 + B_2 + B_3 - B_{\rm CN}$ is the balance of the fragments binding energy of at the ternary fission; $V_1 \equiv V_{13}$ and $V_2 \equiv V_{23}$ are the nucleus-nucleus interaction of the middle cluster "3" (A and Z are its mass and charge numbers, respectively) with the left "1" (A_1 and Z_1) and right "2" (A_2 and Z_2) fragments of the ternary system; $V_{12}^{(Coul)}$ is the Coulomb interaction between two border fragments "1" and "2" which are separated by distance $R_1 + R_2$, where R_1 and R_2 are the distances between the middle cluster and two clusters placed on the left and right sides, respectively. The interaction potentials V_1 and V_2 consist of the Coulomb and nuclear parts:

$$
\begin{aligned}
& V_i((R_i, Z_i, Z_3, A_i, A_3) = V_i^{(Coul)}(Z_i, Z, R_i) \\
& +V_i^{(nucl)}(Z_i, A_i, Z_3, A_3, R_i), \quad \text{where} \quad i = 1, 2. \qquad (2)
\end{aligned}
$$

The nuclear interaction calculated by the double folding procedure with the effective nucleon-nucleon forces $f_{\rm eff}$ depending on nucleon density distribution [15]. The nucleon density of fragments is used as the Fermi distribution with the parameters r_0 = 1.15–1.18 fm, ρ_0 = 0.17 fm^{-3}, a = 0.54 fm. The details of the method can be found in the Appendix of Ref. [16]. The Coulomb interaction is determined by the Wong formula [17].

The nucleus-nucleus interaction potential between middle nuclei and left nuclei "1", $V_1(R)$, depends on the Coulomb interaction between "1" and "2" nuclei and vice-versa, the last interaction affects $V_2(R)$. The effect of the third fragment is important in the case of the short time between ruptures of two necks because the depth of the potential well will be smaller, *i.e.* the barrier against fission will be smaller. Consequently the probability of the ternary fission increases. The values of the binding energies of all considered possible fragments as constituents of ternary system, obtained from the table of masses by Audi *et al* [18]. The procedure of calculations of PES by Eq. 1 has been made by following steps: i) We find positions $R_{\rm m1}$ and $R_{\rm m2}$ of the edge fragments ${}^{A_1}Z_1$ and ${}^{A_2}Z_2$ relative to the middle cluster ${}^{A_3}Z_3$, respectively, providing the minimum value of U by variations of values of R_1

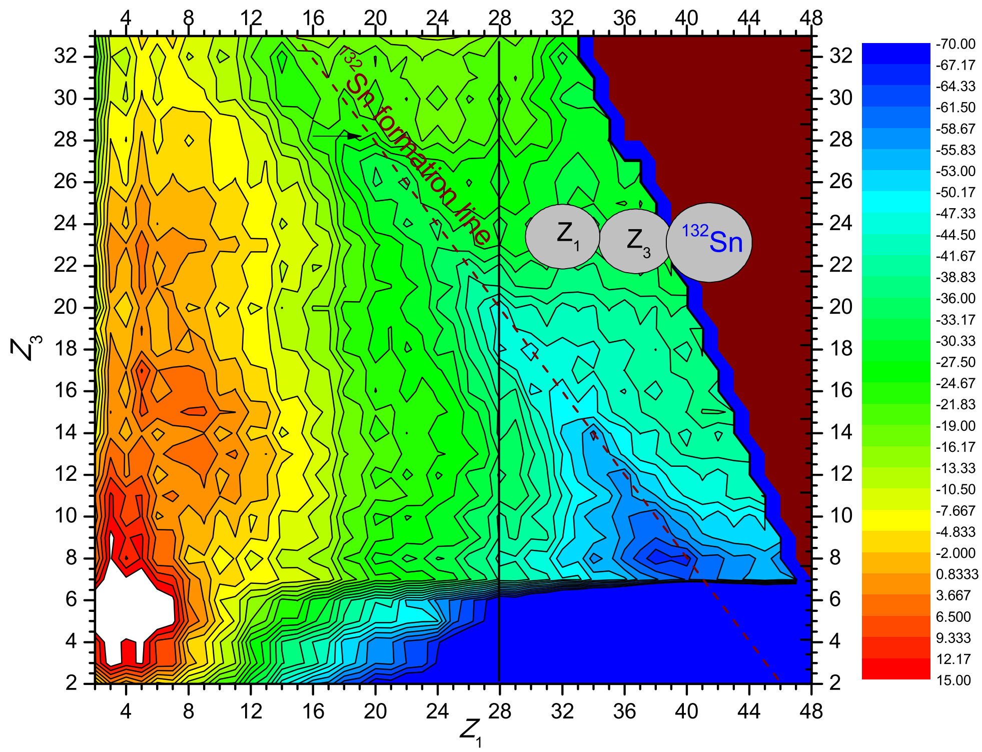

FIG. 2: (Color online) Potential energy surface calculated for the pre-scission state of the ternary system formed after emission 2 neutrons from ^{252}Cf as a function of the charge numbers of middle cluster and left edge fragment.

and R_2. ii) The cluster mass number A_3 for each charge number Z_3 is changed from the minimum value $A_3 = 2Z_3$ up to $A_3^{\max} = 2.6Z_3$ leading to a strong increase of U. iii) The charge number of the right edge fragment Z_2 is found from the conservation law for the proton numbers $Z_2 = Z_{\rm tot} - Z_3 - Z_1$. iv) The neutron distribution between constituents of ternary system at the given charge distribution. In order to find the minimum of U as a function of A_1 for the given mass and charge numbers $^{A_3}Z_3$ of the middle cluster and charge numbers Z_1 and Z_2, we vary A_1 from A_1=$2Z_1$ up to $A_1^{\max}$ corresponding to the strong increase of PES. The mass number of the right edge fragment A_2 is found from $A_2 = A_{\rm tot} - A_3^{\min} - A_1$. The above mentioned $Z_{\rm tot}$ and $A_{\rm tot}$ are the total charge and mass numbers of the fissioning system.

The results of calculations $U(R_{\rm m1}, R_{\rm m2}, Z_3, Z_1, A_3, A_1)$ can be presented as a matrix with the size $(\Delta Z_3, \Delta Z_1)$, where ΔZ_3 and ΔZ_1 are the interval of variation of the Z_3 and Z_1, respectively.

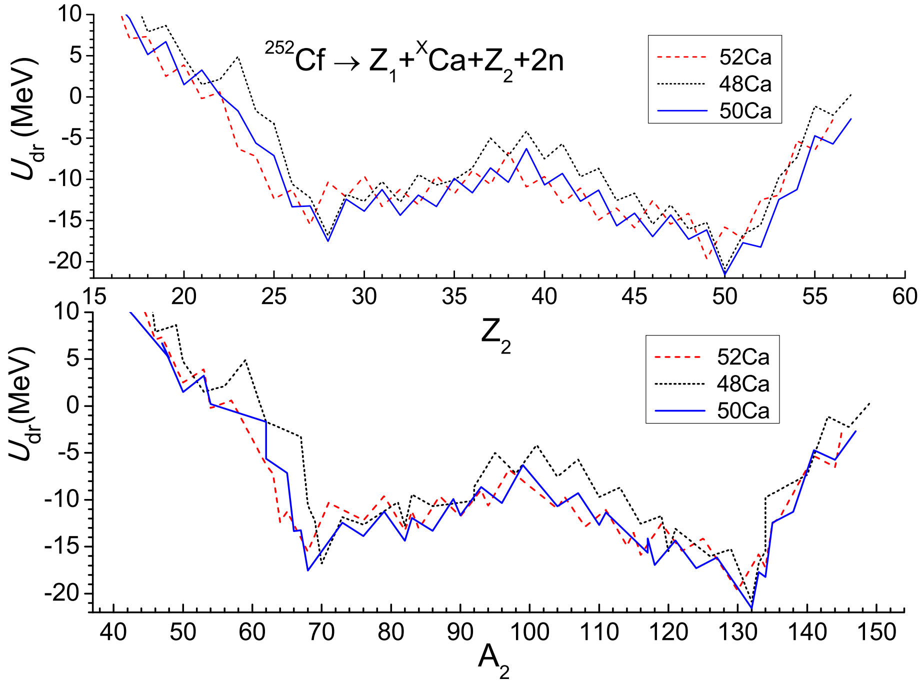

FIG. 3: (Color online) Comparison of the driving potentials calculated for the pre-scission state of the collinear ternary system $Z_1+{}^{A}\mathrm{Ca}+Z_2$ formed in the spontaneous fission of ^{252}Cf as function of Z_2 (upper panel) and as function of A_2 (lower panel)

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Probably the constituents are not in their ground state after formation and before their escape from the ternary system, but a procedure of calculation by consideration mass and charge numbers as variables, changing in the wide range of values will be very labor-consuming.

Therefore, shell effects in the binding energies do not depend on their deformation.

III. RESULTS OF CALCULATION

Results of the PES for the ternary fission of ^{252}Cf are presented by a contour map in Fig. 2 as a function of the charge numbers of the middle cluster Z_3 and one, Z_1, the right edge fragment. It can be seen a valley corresponding to the formation of the cluster ^{132}Sn for different values of Z_3 and Z_1. This fact reflects the long tail in the mass-mass distributions of the experimentally registered products which is parallel to the M_1 and M_2 axes (see Fig.4

in Ref. [4] and Fig. 5a in Ref. [7]). Those tails demonstrate the persistence of shell structure in the double magic nucleus ^{132}Sn in the formation of the fission fragments. The vertical line at Z_1=28 shows local minima corresponding to the formation of Ni isotopes as fragments of the ternary system. This line crosses the line on valley of formation ^{132}Sn isotope at Z_3=20 where there is a local minimum. The probability of formation of the cluster configurations ^{132}Sn+^{50}Ca+^{68}Ni after emission of two neutrons is large because the proton or neutron numbers of the three fragments are equal to the magic numbers wether 28, 50 and 82. The dependence of the driving potential $U_{\rm dr}$ extracted from PES $U(R_{\rm m1}, R_{\rm m2}, Z_1, Z_3, A_1, A_3)$ at $R_1 = R_{\rm m1}$ and $R_2 = R_{\rm m2}$ on the charge and mass numbers of the right edge fragment is shown in the upper and bottom figures of Fig. 3, respectively. The comparison of results obtained for the mass numbers 48 (dotted curve), 50 (solid curve) and 52 (dashed curve) of Ca being the middle cluster in Fig. 3, demonstrates that minimal values of driving potential corresponds to the formation of the ^{68}Ni isotope, which was observed with sufficiently large probability (see Refs. [4, 7]), when ^{50}Ca is formed as a middle cluster and ^{132}Sn is the right edge fragment. On the contour map of the PES there are local minima showing the favored population of ^{132}Sn+^{38}S+^{82}Ge, ^{132}Sn+^{36}Si+^{84}Se, ^{150}Ba+^{22}O+^{80}Ge, and others. We found that the middle cluster is more neutron rich than edge fragments. A much smaller energy minimum in the PES (by 10 MeV) for the alternative configuration, the ^{132}Sn+^{72}Ni+^{48}Ca channel, gives for this reaction a much smaller probability, the difference is due to the changed Coulomb repulsion forces. This effect is observed in the yields observed in the experiment [4].

In Fig. 4 obtained from Ref. [7] the events of yields of two products in the spontaneous fission of ^{252}Cf registered in coincidence are presented. Intense yield of the fission products with A_1 =68—94, A_1 =50—60, and A_2 =128—146 registered in coincidence is observed. Obviously we see the sufficient influence of the shell effects in nuclear matter in formation of the ternary fission products.

The results calculated for ^{252}Cf and presented in Fig. 5 show the valley of minimum values of the PES in the mass number regions A_1 =70—100 and A_2 =124—144. The population of the mass distribution of the edge fragments in the corresponding ranges of A_1 and A_2 should dominate in the ternary fission of ^{252}Cf . The fission probability from these pre-scission states depends on the value of the pre-scission barriers $B_{\rm sc1}$ and $B_{\rm sc2}$ which is determined by the depth of the potential well. The splitting probability of ^{132}Sn from

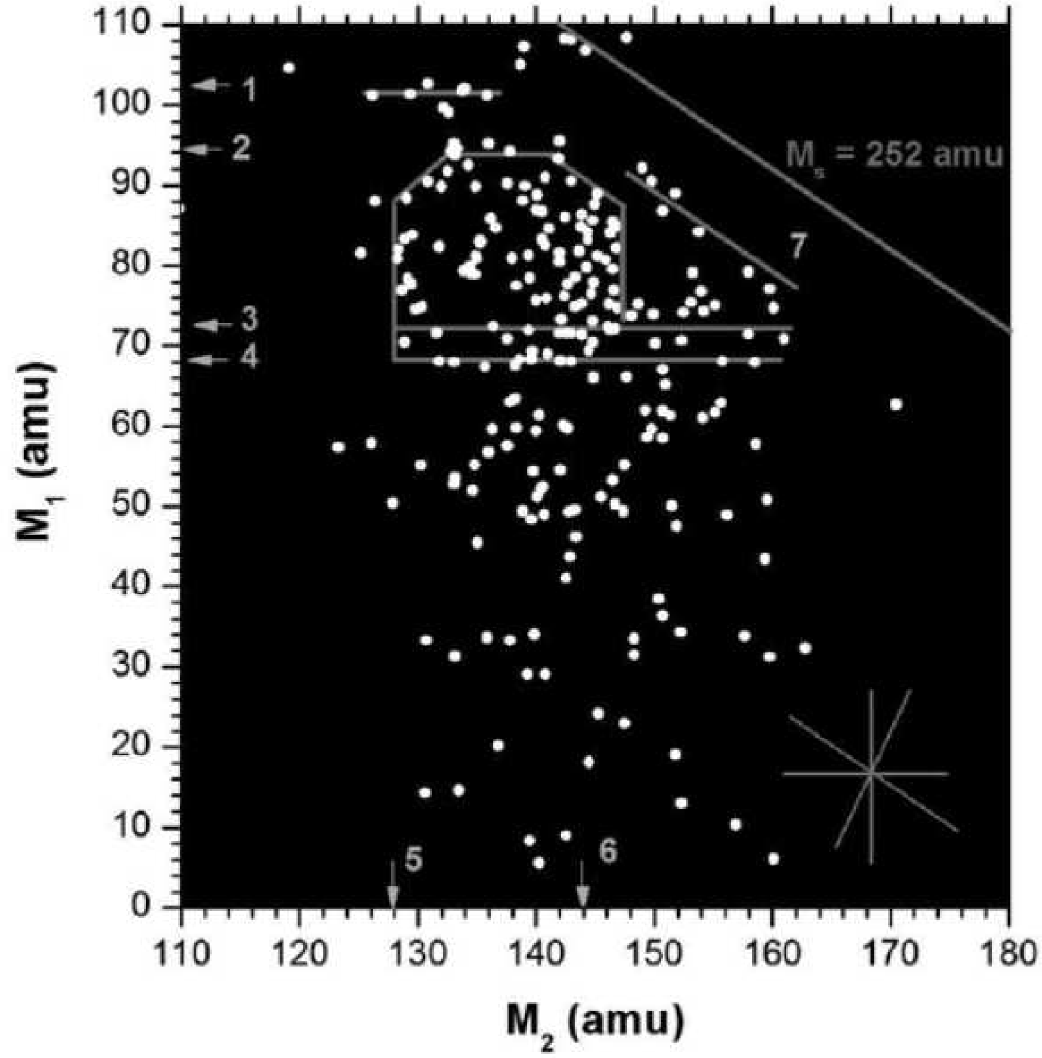

FIG. 4: The mass - mass distribution of the fission-fragments of the spontaneous fission of ^{252}Cf gated by 2n emission. Arrows with numbers 16 mark the positions of masses of magic nuclei, a line numbered 7 points to events with the loss of a ^{14}C nucleus. The main intensity is with masses for the third fragments from 3620. (Copy of Figure 10 from Ref.[7])

the other part of system is determined by the depth of the potential well, which can be considered as pre-scission barrier $B_{\rm sc}$. The value of $B_{\rm sc}$ depends on the charge distribution between fragments of the ternary system. For the collinear configuration of the ternary system, we have two necks in the connected system and, consequently, we have two barriers $B_{\rm sc1}$ and $B_{\rm sc2}$ for separation of the left and right edge fragments. The answer to the question, if these necks are unstable to rupture how the order of their scission depends on the relation

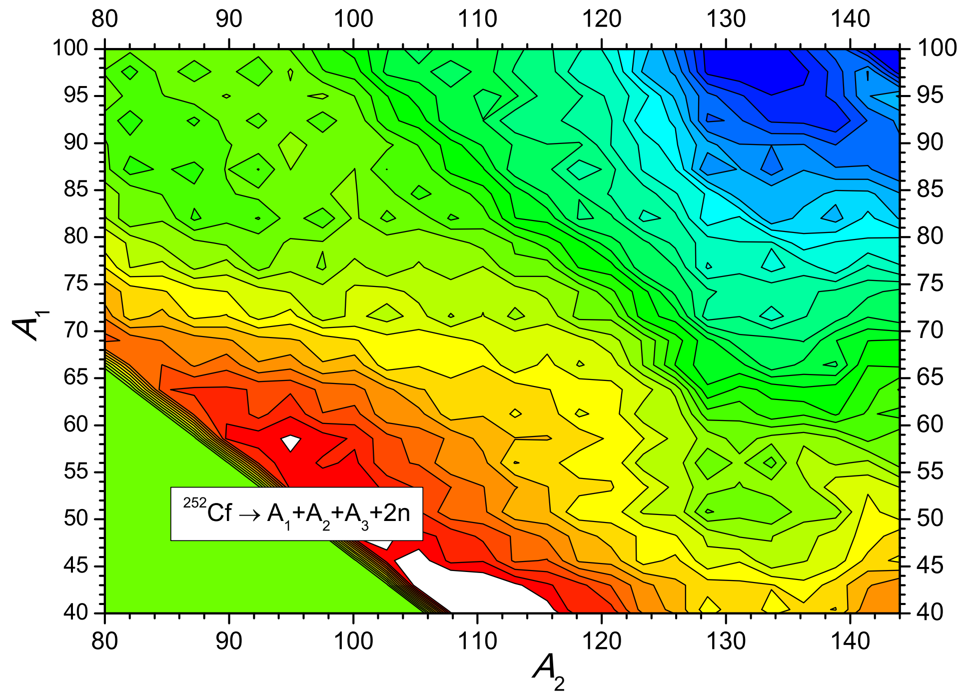

FIG. 5: (Color online) Potential energy surface calculated for the pre-scission state of the ternary system formed after emission 2 neutrons from ^{252}Cf as a function of the mass numbers of the left and left edge fragments.

between $B_{\rm sc1}$ and $B_{\rm sc2}$. The dependence of the values of $B_{\rm sc1}$ and $B_{\rm sc2}$ on the charge number Z_1, for the case that the middle cluster is ^{34}Si at CCT of ^{236}U, is shown in upper figure of Fig. 6. The bottom part figure of Fig. 6 presents the driving potential of the ternary system Z_1+^{34}Si+Z_2 being formed at CCT of ^{236}U. The presented results for the driving potential allow us to conclude, that 1) formation of the ^{68}Ni isotope in coincidence with ^{132}Sn occurs with large probability, because this configuration of the collinear ternary system has lower potential energy; the rupture of the neck connecting ^{132}Sn to ^{34}Si+^{68}Ni system occurs more easy than the rupture of the neck connecting ^{68}Ni to the ^{34}Si+^{132}Sn system. Then breaks down the ^{34}Si+^{68}Ni system in the field of ^{132}Sn. The similar situation takes place in case of CCT in the spontaneous fission of ^{252}Cf, where ^{68}Ni is formed together with ^{132}Sn + ^{50}Ca.

IV. SUMMARY

A possibility of the formation of ^{68}Ni isotope in the spontaneous fission of ^{252}Cf and ^{235}U(n_{th},f) reaction, analyzed by calculation of the potential energy surface for the collinear ternary system is obtained. It is found as sum of the energy balance of fission into three

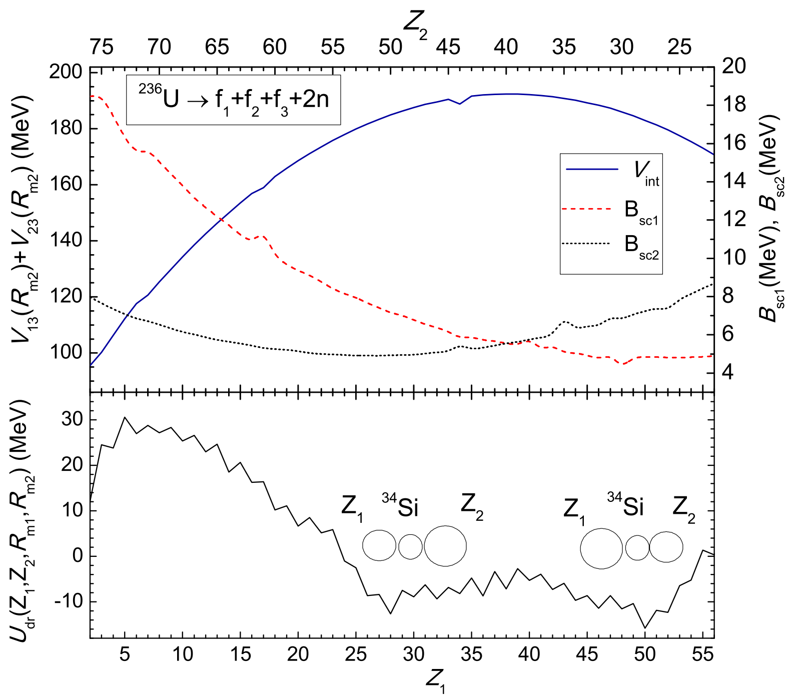

FIG. 6: (Color online) The nucleus-nucleus interaction potential (solid line, left axis), pre-scission barriers (right axis) B_{sc1} and B_{sc2} for the separation of the fragments Z_1 and Z_2 in the collinear ternary system fragment as a function of Z_1 and Z_2 (upper figure); Driving potential for the collinear ternary system as a function of Z_1 when middle cluster is ^{34}Si corresponding to the minimum value of PES (bottom figure) for CCT in the ^{235}U$(n_{\rm th}, f)$ reaction.

fragments, nucleus-nucleus interaction between neighbour fragments and the Coulomb interaction between the edge fragments. Mass and charge distributions of the collinear ternary system is studied by calculation and analysis of the potential energy surface as a function of the distances R_1 and R_2 connecting the edge fragments Z_1 and Z_2 to the middle cluster Z_3. This procedure has been done changing the charge numbers Z_1, Z_2 and Z_3 and corresponding mass numbers in the wide range of their values. As a result we determine the pre-scission state of the ternary system. The landscape of the potential energy surface presented as function of fragments Z_1 and Z_3 showed well pronounced deep valley corresponding to the formation of ^{132}Sn with different pair of fragments with Z_1 and Z_3. Minima and valleys can be seen on the contour map of landscape. There are local minima indicating of formation of Ca, Fe, Ni, Ge and Se isotopes having magic proton or/and neutron numbers, such as 20, 28, and 50. The analysis shows that the experimentally observed ^{68}Ni is formed as the edge fragment of the ternary system connected to the formation of Sn by Si and Ca isotopes at fission of ^{236}U and ^{252}Cf, respectively.

V. ACKNOWLEDGMENTS

The authors are grateful to Drs. D.V. Kamanin and Yu.V. Pyatkov for valuable discussions. A.K. Nasirov is grateful to the Russian Foundation for Basic Research for the partial financial support of this work.

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