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CLUSTER APPROACH TO DESCRIPTION
OF FISSION MODES*

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1 Introduction

Recent intensive investigations essentially enriched the knowledge about manifestation of the cluster degrees of freedom in light α -cluster nuclei. It is reasonable to speak about the nuclear molecule as the object of study [1, 2]. The investigations of the cluster degrees of freedom in heavy nuclei are still conjugated with large experimental and theoretical difficulties. However, some experimental data on fission of both the preactinide and actinide nuclei can be naturally treated in cluster terms. The strongly bounded fragments (with almost closed shells) take there the role of clusters. The well-known example of such a kind is the dependence of the multiplicity of neutron emission on the mass of the fission fragment (FF) in low-energy fission (Terrell curve [3]). For the FF that is close to the shell-nuclei in the vicinity of the mass numbers $A_H \approx 130$ and $A_L \approx 82$, the neutron multiplicity is equal to zero within the error bars (A_H and A_L are the mass numbers of the heavy and light FF, respectively). This means that in any configuration of the fissioning system such FF have not enough excitation energy for the neutron emission. As a result, we can assume that the preformation of these FF occurs at early stages of fission. These cluster-like prefragments remain unchangeable in the further evolution of the fissioning system. Based on the study of the neutron emission in the spontaneous ^{252}Cf fission [4, 5, 6], the indication of the validity of the above assumption was obtained for the fragments with the mass numbers $A_H \sim 130$ and $A_L \sim 80, 90$. The next example concerns the fission of the nuclei lighter than Th. As was established in [7], the asymmetric mass components with $A_H \sim 134$ in the FF mass distribution appear only for the fissioning nuclei with $A \leq 200$ and/or $Z \approx 80$. The vanishing of the asymmetric mass components for the heavier fission nuclei believes to be caused by the shell effects in the fragments. The heavy fragment is formed under the effect of the spherical shells with $Z_H = 50$ and $N_H = 82$. The mass of the light fragment is defined by the mass of the nucleus with the closed either spherical shell ($Z_L = 28$, $N_L = 50$) or deformed shell ($Z_L = 30$, $N_L = 44$) [8, 9]. Therefore, the sum of the FF masses is approximately equal to the borderline mass value discussed.

The strong similarity between the symmetric high-energy mode of the Fm-No fission with extremely small mass variance of the FF and the asymmetric mode $a1$ in the fission of the nuclei in the vicinity of $A = 210$ is noted in [9]. The symmetric fission mode assumed to be resulted from the preformation of two double magic ^{132}Sn nuclei. For the nuclei with $A \approx 210$, the configuration consisting from the two spherical fragments can be obtained with the replacement of one ^{132}Sn nucleus in the previous case by the double magic ^{78}Ni nucleus.

The presented qualitative picture is supported by the calculations based on the Woods-Saxon-Strutinsky model involving many deformation degrees of freedom [10]. It was shown that the density distribution and the shape of the fission nucleus at large elongation (in third minimum) look like a di-nucleus. Such a clusterization is a dramatic manifestation of the shell-structure role in the fission of heavy nuclei.

Besides the cluster aspects connected with the formation of the FF, there are indications of the light-cluster formation in the contact region between heavy nuclei [11].

The recent experimental data presumably indicating the clusterization of the fissioning $^{234}\text{U}^*$ nucleus were presented in Ref. [12]. The Ref. [12] deals with the fine structures of the FF mass (M) - total kinetic energy (TKE) distributions. By notation, the local areas of the two-dimensional distribution with increased yields of the FF, as compared to the ones supplied by the smoothed global distribution, are treated as the fine structure (see Fig. 1). The fine structures in the form of the ridges having a small slope to the energy axis in the $TKE-M$ distribution of $^{234}\text{U}^*$ were demonstrated in Refs. [13, 14]. The conclusion about their origination, mainly from the proton odd-even effect, was made. This effect is the strongest one, however it is not alone in producing the fine structure. For the extraction of the structure shown in Fig. 2, the total $TKE-M$ distribution in the cuts was smoothed at $M = \text{const}$. The smoothed distribution was subtracted from the initial one. This procedure has a minimal sensitivity to the structures produced by the proton odd-even effect because the direction of the cuts, along which the smoothing occurs, is approximately

parallel to the direction of the ridges produced by the odd-even effect. Using this procedure, the fine structure which has different nature as compared to structure produced by the odd-even effect was subtracted. We will try to interpret this structure in the framework of the cluster model presented below.

2 Cluster-like configurations in the fissioning nucleus

The phenomenological cluster concept of the low-excited heavy-nuclei fission being the development of the earlier models [15, 16] was suggested in [17, 18] to take into consideration the new experimental and theoretical results obtained from several fields. The fields considered are: the specific manifestation of the shell effects in the nuclear fission, the data on the clusterization in α -cluster systems, the calculations indicating the multimode character of the fission, as well as the theoretical models of the cluster decay. The integration of these questions is based on the assumption about the generality of the physical mechanisms determining the behavior of all nuclear systems where the cluster degrees of freedom are important. The paper at hand aims to provide a quantitative verification of the concept mentioned.

Referring to Ref. [18] let us assume that, for some elongations, the fissioning system is transformed to the one consisting of two large clusters connected by a neck. The large clusters are assumed to be the strongly bounded shell nuclei, for example, ^{132}Sn and ^{82}Ge . The nucleons in the neck are also joined into the clusters among which the α -particles are preferable. With the descent from the fission barrier, the large clusters retain their individual properties, and the elongation of the system is caused by the elongation of the neck. The elongation of the clusterized neck means the rearrangement of α -particles between the two large clusters. The different neck scissions are responsible for the formation of the different FF. The fission mode is thus determined by the large clusters. The preformation of different pairs of the large clusters is the reason of the multimodality of the nuclear fission. This

paper is aimed to estimate quantitatively the characteristics of the fissioning system within our approach based on the two key points: the preformation of two large clusters gives rise to the fission mode; the neck between the large clusters clusterises predominantly on α -particles.

Based on the experimental indications mentioned above, the probable mode in the thermal neutrons that induced the fission of ^{233}U and ^{235}U was chosen for particular calculations. In this mode, the fissioning system represents two large clusters ^{132}Sn , Ge and five α -particles (^{20}Ne) between them. The configurations investigated are schematically presented in Table 1. All configurations with labels from 1 to 11d are constructed using the following rule: the distance between the nearest edges of the large clusters is a multiple of the α -particle diameter; at fixed elongation of the system the α -particles, which are not included in the chain connecting the large clusters, can be in the positions to maximize the attractive forces raised by the nuclear interaction. The purpose of the comprehensive consideration of all possible configurations was out of this paper. We would like only to demonstrate the validity of the assumption on the clusterization that takes place in the fissioning nucleus in the description of the experimental data.

For the given cluster configuration, the potential energy of the system and total kinetic energy of the fragments are calculated as a function of location of the scission point along the neck. The potential energy of the cluster configuration consisting of n clusters is determined as follows

$$V_{int} = \sum_{i>j}^n U_{ij}(\mathbf{r}_i - \mathbf{r}_j) + \sum_{i=1}^n B_i - B_0, \quad (1)$$

where $U_{ij}(\mathbf{r}_i - \mathbf{r}_j)$ is the potential energy of the interaction between the "i" and "j" clusters, B_i is the binding energy of the cluster "i", B_0 is the binding energy of the fissioning nucleus in its ground state. The origin of the coordinates for V_{int} coincides with the energy of the ground state of the fissioning nucleus. The cluster-cluster potential depends on the Coulomb and nuclear interactions between the "i"

Table1: TKE for some configurations of the fissioning system $^{236}\text{U}^*$

label	Scission configuration	TKE MeV	label	Scission configuration	TKE MeV	label	Scission configuration	TKE MeV
1		196	5c		141	10a		151
			5d		138	10b		149
			5a'		130	10c		137
			5b'		135	10d		132
			5c'		138	11a		153
			5d'		143	11b		139
			6a		135	11c		136
			6b		132	11d		130
			6c		126			
			6d		122			
6e		121						
6f		117						
7a		169						
7b		168						
7c		150						
8a		172						
8b		157						
8c		149						
9a		149	13a		193			
9b		147	13b		168			
9c		142	3'a		182			
9d		132	14a		164			
2a		209	14b		145			
2a'		185						
3a		193						
3b		190						
3a'		171						
3b'		178						
4a		166						
4b		165						
4c		160						
4a'		147						
4b'		154						
4c'		158						
5a		148						
5b		145						

and "j" nuclei:

$$U_{ij}(r_{ij}) = U_{ij}^N + U_{ij}^C \quad (2)$$

The Coulomb potential U_{ij}^C can be easily calculated in accordance with Ref. [19]. For calculation of the nuclear interaction U_{ij}^N , the double folding method is used:

$$U_{ij}^N(r_{ij}) = \int \rho_i(\mathbf{r}_i) \rho_j(\mathbf{R} - \mathbf{r}_j) \mathcal{F}(\mathbf{r}_i - \mathbf{r}_j) d\mathbf{r}_i d\mathbf{r}_j \quad (3)$$

Here ρ_i and ρ_j are the nucleon densities of the interacting nuclei. The density dependent nucleon-nucleon interaction is taken as

$$\mathcal{F}(\mathbf{r}_i - \mathbf{r}_j) = C \left[F_{in} \frac{\rho_0(\mathbf{r}_i)}{\rho_{00}} + F_{ex} \left(1 - \frac{\rho_0(\mathbf{r}_i)}{\rho_{00}} \right) \right] \delta(\mathbf{r}_i - \mathbf{r}_j) \quad (4)$$

where N and Z are the neutron and proton numbers of the nucleus,

$$F_{in,ex} = f_{in,ex} + f'_{in,ex} \frac{N_i - Z_i}{A_i} \frac{N_j - Z_j}{A_j}$$

Interaction (4) is well-known in the theory of finite Fermi-systems [20], giving a good description of a large set of the experimental data. In (4), $\rho_0 = \rho_i + \rho_j$. This approximation is good for a small overlap of the nuclei. The nuclei overlap slightly in cluster configurations discussed. The following set of parameters was used in our calculations: $C = 300 \text{ MeV fm}^3$, $\rho_{00} = 0.17 \text{ fm}^{-3}$, $f_{in} = 0.09$, $f_{ex} = -2.59$, $f'_{in} = 0.42$ and $f'_{ex} = 0.54$.

For the nuclei with $A > 16$, the nuclear density was taken as follows:

$$\rho_i = \frac{\rho_{00}}{1 + \exp[(r - R_i)/a_0]}$$

where $R_i = r_0 A_i^{1/3}$ is the radius of the nucleus "i". For light nuclei, the following functional dependence for the density is more realistic

$$\rho_i(r_i) = A_i \left(\frac{\gamma^2}{\pi} \right)^{3/2} e^{-\gamma^2 r_i^2}$$

In our calculations, we used the following values of the parameters: $r_0 = 1.12 \text{ fm}$ and $a_0 = 0.54 \text{ fm}$. The value of the parameter γ was varied

from 0.5 fm^{-1} for ^{16}O up to 0.671 fm^{-1} for ^4He . For calculation of the interaction between the two deformed clusters ($R_i = R_{0i}(1 + \beta_i Y_{20})$), we used the results of Ref. [21]. The values of the deformation parameters β_i were taken from the tables of [22]. We should note that this method of calculation of the nucleus-nucleus potential was successfully used in determining the potential energy of the dinuclear systems formed in the deep-inelastic collisions of heavy ions [23]. Due to the density dependence of the nucleon-nucleon interaction, the potential $U_{ij}(r_{ij})$ has a repulsive core. For the majority of the pairs of the interacting nuclei, there is a pocket in the $U_{ij}(r_{ij})$ dependence on r_{ij} with a bottom corresponding to the intercenter distance $r_{ij} = R_i + R_j + 0.5 \text{ fm}$. In our calculations of the potential energies of cluster configurations, the distance between touching clusters corresponds to the bottom of the potential pocket. The binding energies B_i and B_0 were taken from [22]. For illustration, the calculated interaction potentials in the α -particle- ^{82}Ge and ^{82}Ge - ^{132}Sn system are depicted in Fig. 3.

The calculation of TKE as a function of the scission point along the neck was performed as follows. If the fissioning system is disintegrated into the A and B FF, then

$$TKE = \sum_{\substack{i \in A \\ j \in B}} U_{ij},$$

where the values of U_{ij} are calculated at a moment of the scission. It should be stressed that the scission criteria is not considered in the present model. Therefore, the dependencies of V_{int} and TKE on the FF mass can be calculated, however the model is not able to predict the yields of the FF.

3 Results and discussion

The important characteristics of the fissioning systems ^{233}U and ^{235}U were calculated according to the model described above. First of all, the total interaction energy for the sequence of the elongating configurations was evaluated. Beside this, the total kinetic energy values were calculated (see Table 1) for different places of the scission (labeled

by vertical lines) in each configuration. The calculated TKE values were compared with the experimental data on the fine structure of the TKE - M distributions of the $^{234}\text{U}^*$ FF (Fig. 4) that were measured using the time-of-flight spectrometer at the reactor of Moscow Engineering Physics Institute [24]. The complete analysis of the fine structures observed (the lines marked by letters A , B and structures F in Fig. 2) is out of the frame of this work and will be given in the forthcoming publications. As of now we note that the TKE values (Table 1) for configurations with three α -particles between large clusters are well grouped near the line of the fine structure labeled by letter "a". For configurations with four α -particles between large clusters, the TKE values lie slightly below the line "b" of the fine structure. The TKE values for configurations with a neck formed from the chain of five α -particles can not be compared with the experimental data because the statistics is too little in this region of the spectrum. The strong manifestation of the fissioning system configurations, in which the distance between the large clusters is a multiple of the α -particle diameter, is seen in Fig. 4 and requires a discussion. There are some reasons for the preferential production of the FF from the decay of these configurations. For a given system elongation, the general number of configurations, when not all α -particles are responsible for the integrity of the system, is essentially more (for example, the configurations 4, 7, and 8) than one in the case when all α -particles supply this integrity (the configuration 3'a). There are no "free" α -particles in the configuration 3'a to organize the other configurations by the rearrangements of alpha-particles along the alpha-chain. Therefore, the statistical weight can be more for the configurations with the intercluster distance proportional to the α -particle diameter. For these states, the scission probability can be larger as compared to the state without neck clusterization due to the multitude of the points preferable for the scission.

The subsequent comment concerns the discrete character of the lines "a" and "b" in the fine structure (Fig. 4). In our model, the scission in the neck gives the FF with masses differ on four amu , i.e. this scissions do not give continuing sequence of fragments masses ob-

served in experiment. As compared with our model, the intermediate values of the mass can be obtained, if one assumes, the small uncertainty in the mass numbers of the large clusters and/or small surplus of neutrons in the neck reference to its pure α -particle composition (the change of ^{20}Ne to ^{22}Ne is enough for this purpose). This surplus of neutrons can be caused by the well known demand for the Z/N ratio to be close to that in the parent nucleus. Our calculations demonstrated that the V_{int} and TKE values for the $^{82}\text{Ge}-5\alpha-^{132}\text{Sn}$ and $^{84}\text{Ge}-5\alpha-^{132}\text{Sn}$ modes differ by few hundred keV. This is why the continuity of the mass number is supplied without the appreciable change in the form of the discussed line of the fine structure.

As was noted above, for each cluster configuration of the fissioning system, the total energy of interaction of the clusters was calculated. The calculated values of V_{int} were compared with the well-known calculations of Ref. [25]. In Fig. 5, the dependencies of the energy of the fissioning ^{236}U nucleus are presented for different fission modes as a function of half-length of the fissioning nuclear system (see Ref. [25]). The points connected by the solid line correspond to the V_{int} values calculated by us for the configurations listed in Table 1. There is a good agreement (Fig. 2) between our results and the ones of Ref. [25] if to keep in mind that the valley image onto the potential energy surface is influenced by the shape parametrization used [26].

The agreement of the calculations performed using the models with the clusterized and unclusterized necks proves the shapes of the fission nucleus to be roughly similar in the both cases. However, only the cluster model provides the explanation of the periodic structure (the lines a and b in Fig. 4) linking it with the configurations in which the length of the neck is a multiple of the α -particle diameter. Therefore, the model with the clusterized neck seems to be not only the simplest algorithm of calculation but also a mechanism actually taking place in nature. One more conclusion can be derived from the comparison in question. A small difference between the predictions of the two models means that both phases – clusterized and unclusterized – can coexist in the neck of the fission nucleus.

Some comparisons with the results of the known calculations [27,

28] can be done if we estimate the barriers for the FF separation. By definition, the separation barrier is due to the dependence of the interaction energy between the fragments A and B on the distance s between their nearest edges.

$$V(s) = \sum_{\substack{i \in A \\ j \in B}} U_{ij}(s).$$

If the fragments, between which the scission occurs, touch each other, then $s = 0$. Let us assume that on every stage of the FF acceleration their shapes remain to be constant. The FF separation barriers for several configurations of the fissioning system are shown in Fig. 6. The labels of the curves correspond to the configurations given in Table 1.

The separation barriers for the compact initial configurations are presented in Fig. 6a. The FF for such configurations are characterized by the TKE close to the Q -value of the fission, i.e. the so called cold fission is realized from the compact configurations considered. The fission from the most compact configurations can be prohibited, as the TKE would exceed the Q -value (see, for example, configurations 12a, 2a and curve 2a). At the same time the TKE values close, but less than Q , can be reproduced by means of rearrangement of the α -particles in the neck (configuration 13) or elongation of the system (configuration 3a). The existence of the barrier for the FF separation in the compact configurations of the fissioning system and, following to this point, the tunnel mechanism of cold fragmentation are in agreement with the concept of Ref. [27]. The energy corresponding to the bottom of the potential pocket of the fragment-fragment potential was taken as the TKE in our calculations. The zero vibrations, which increase slightly the TKE and penetrability of the barrier of the FF separation, were not taken into account. It means that the TKE calculated without consideration of the zero vibrations can be considered as a low limit of the TKE values. However, the omitted such a dynamical effect as the nucleon exchange between the neighboring clusters can compensate in part the contribution from the zero vibrations. This effect results in increasing the depth of the potential pocket in the fragment-fragment potential [29]. For large elongation of

the fissioning system, the depth of the pocket in the fragment-fragment potential is small. Therefore, calculation of the TKE value contains less uncertainties with increasing the system elongation.

Rather interesting result follows from the calculation performed for the system elongation corresponding to the most probable TKE values and mass of the light fragment (Fig. 6b, curve 4c). For FF separation, the barrier practically vanishes. This fact correlates also with the conclusion of Ref. [28] about the disappearance of the barrier between the fission and fusion valleys at some stage of elongation of the nucleus. The absence of the barrier contains a purely clustering aspect: the clusterization of a light nucleus occurs at the excitation energies close to the disintegration threshold of this nucleus (see the phenomenological Ikeda rule [30]). Probably, the condition close to this requirement is realized in the neck being equivalent to the ^{20}Ne nucleus. Either disappearance or small height of the barrier for the FF separation mean the instability that is favorable for the clusterization.

As was assumed in Ref. [18], during the evolution, the fissioning system really passes the phase where the nucleons in the neck between the large clusters form the ^{20}Ne nucleus in its ground state. The values of V_{int} , the elongation of the system, and the TKE value obtained for the configuration 14 in Table 1 (the point labeled by ^{20}Ne in Fig. 5) seem to be close to the corresponding values for the configurations 4 and 4'.

The V_{int} and TKE values for configurations 7 and 8, containing the pyramid formed by the four touching α -particles, are close to the appropriate values obtained in the case of replacement of this pyramid by the deformed ^{16}O nucleus [22]. The similar result is obtained in the replacement of the pyramid formed by three α -particles in the configurations 9-12 by the ^{12}C nucleus in the ground state. For the configurations with the ^{16}O and ^{12}C nuclei in the neck, the values of V_{int} are marked by asterisk in Fig. 5. Thus, our calculations confirm a hypothesis of Ref. [18] that some symmetry should be observed between the prolate and oblate shapes of the neck nucleus (^{20}Ne): at some excitation (deformation) the clusterization occurs in both cases. When the distance between the large clusters increases, the nucleons

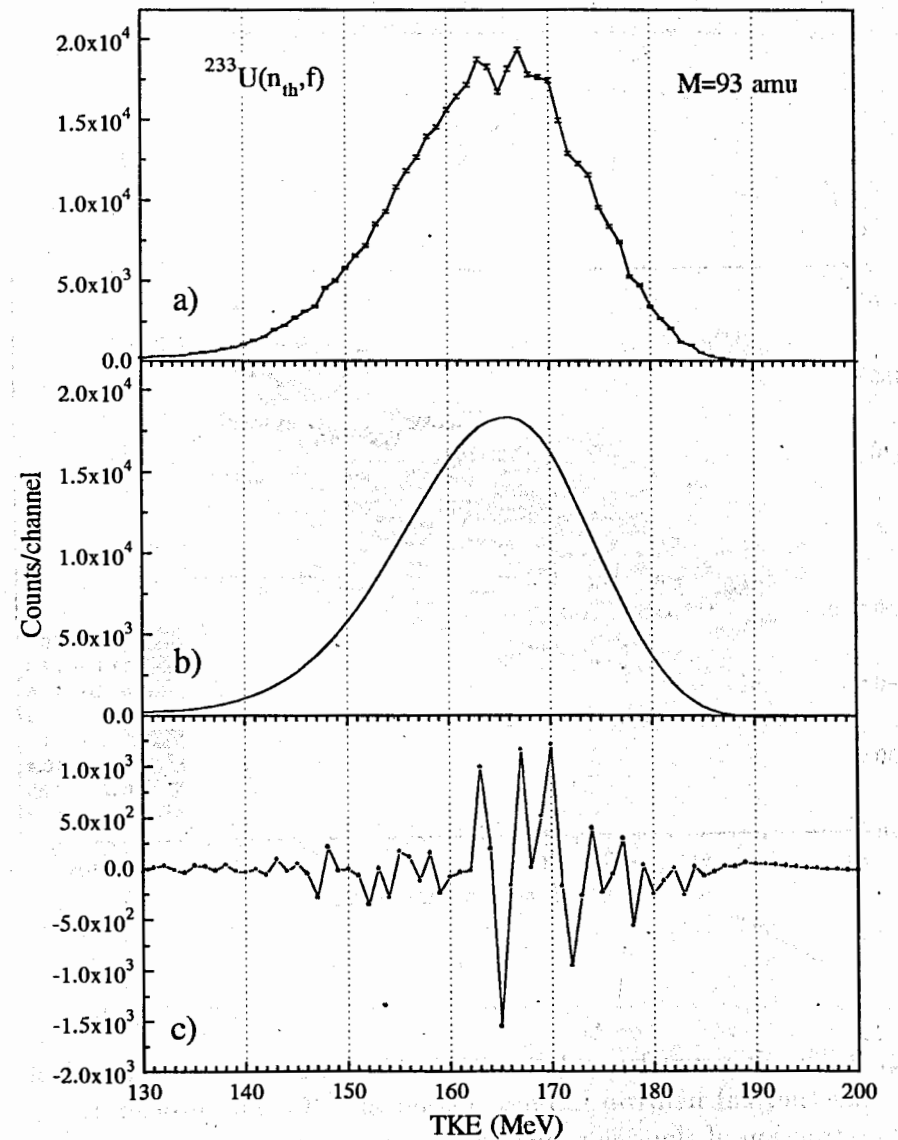


Fig. 1 Schematic illustration of the procedure for the fine structure extraction; a - initial cut of TKE - M distribution along $M = const$; b - the same cut after the smoothing; c - the result of subtraction the smoothed cut from the initial one.

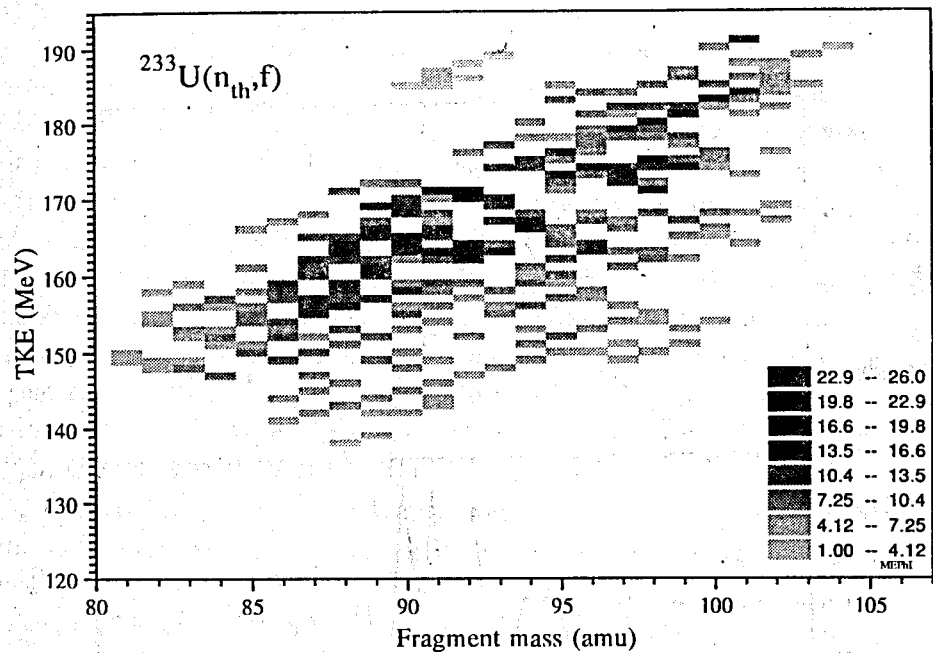


Fig. 2 The fine structure of the $TKE-M$ distribution for the FF of the thermal neutron induced fission of ^{233}U . The procedure for extraction of the fine structure is given in the text.

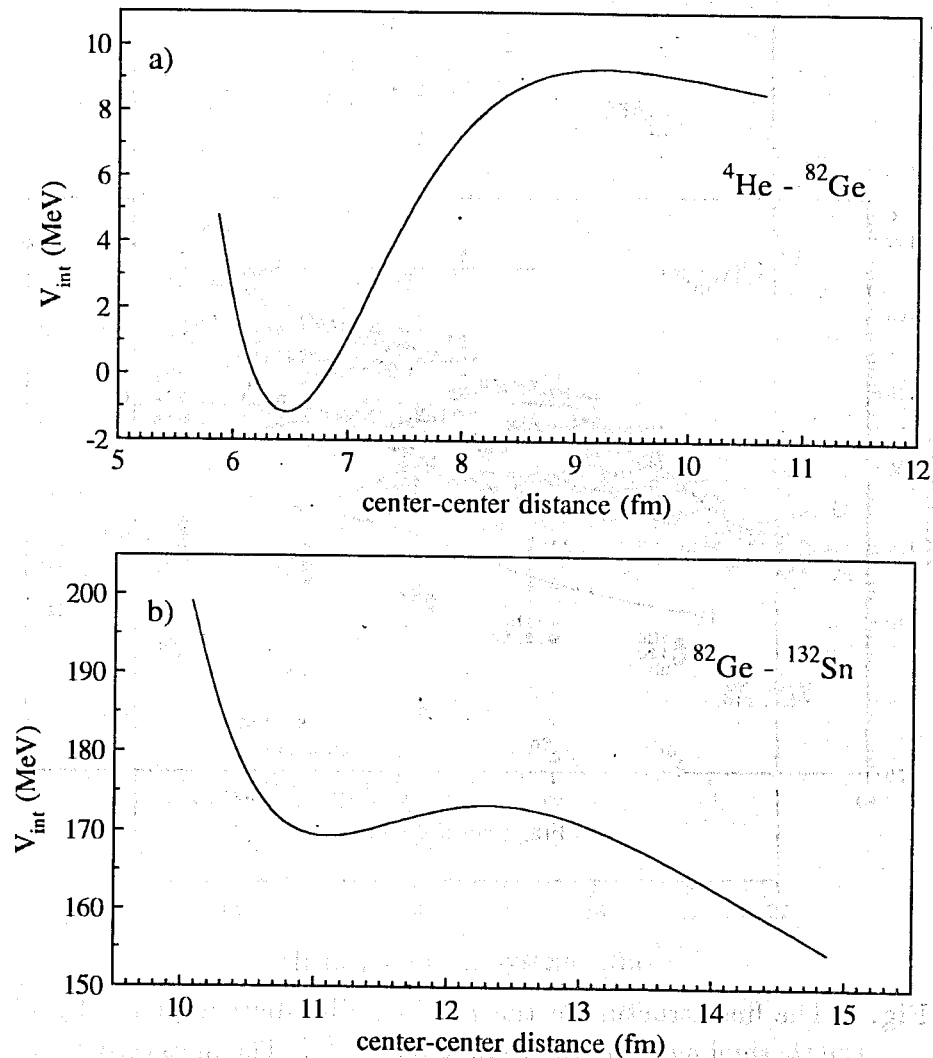


Fig. 3 The calculated interaction potentials for the α -particle- ^{82}Ge (a) and ^{82}Ge - ^{132}Sn (b) systems.

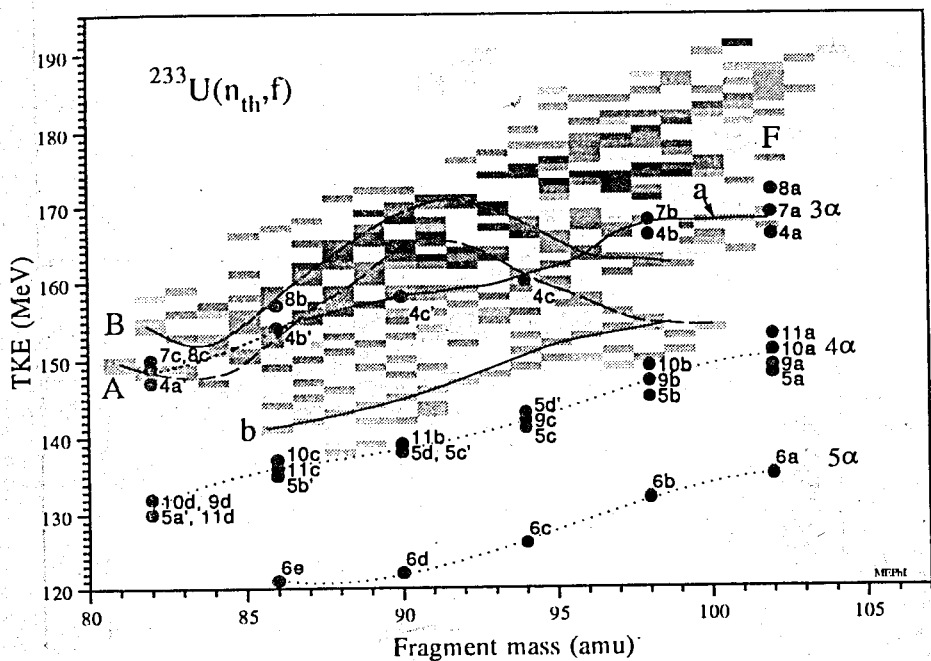


Fig. 4 The fine structure of the $TKE-M$ distribution for the FF of the thermal neutron induced fission of ^{233}U . The lines connecting the points of fine structure are drawn to guide the eye. The solid points correspond to the configurations presented in Table 1. For details, see the text.

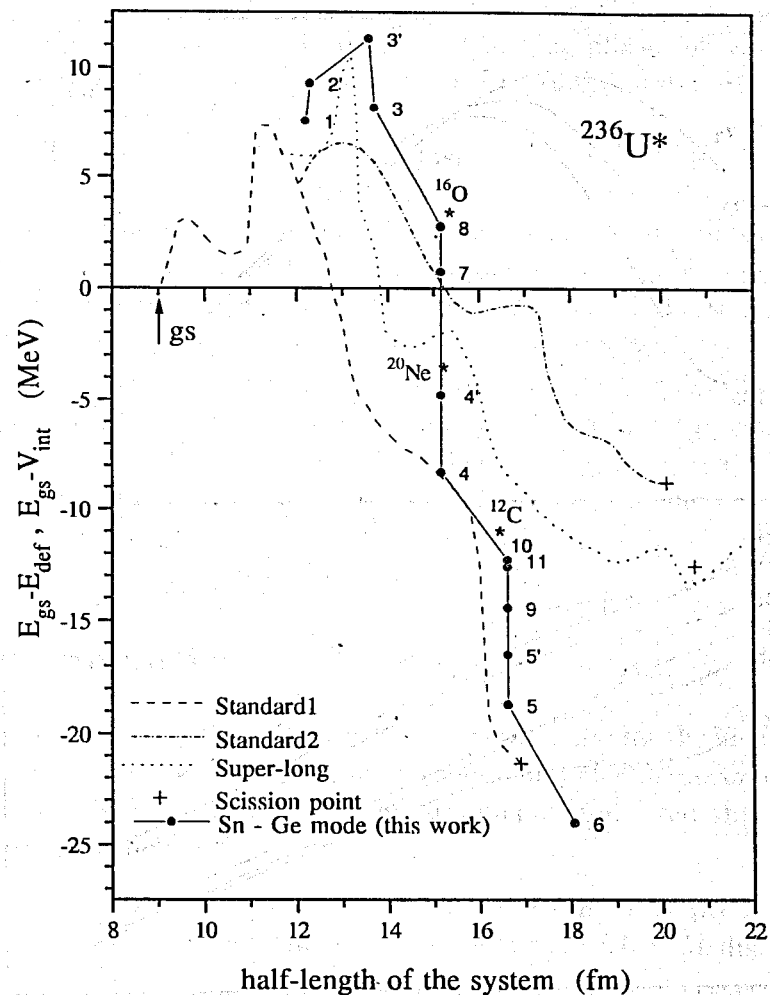


Fig. 5 The potential energy of the fissioning $^{234}\text{U}^*$ nucleus as a function of half-length of the system. The curves labeled as Standard 1, Standard 2, and Super-long are taken from Ref. [25]. E_{gs} is the energy of the ground state.

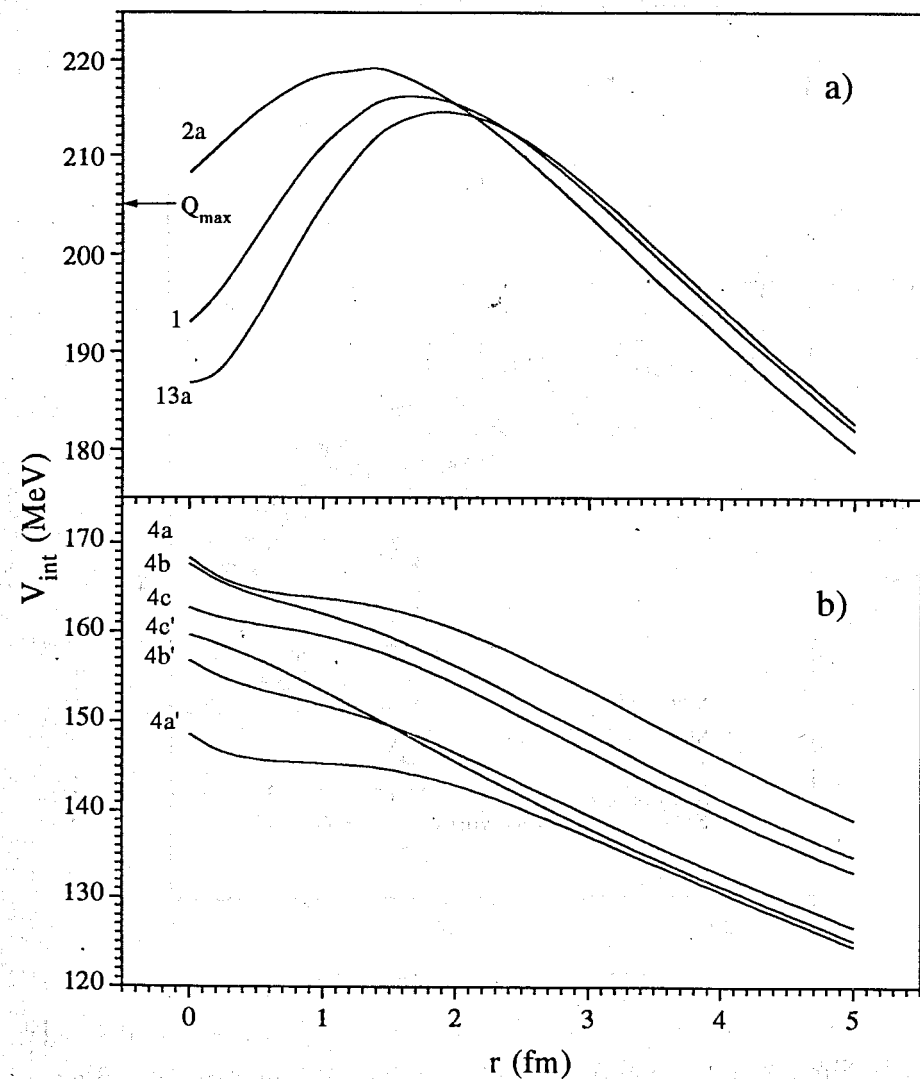


Fig. 6 The FF separation barriers for compact precission configurations of the fissioning system (a) and for elongations close to the most probable ones (b). The numbers near the curves indicate the configurations from Table 1.

in the neck consistently pass the following phases of association: α -clusters; α -clusters + ^{16}O ; ^{20}Ne in its ground state; $^{12}\text{C} + \alpha$ -clusters; α -clusters (see Fig. 5).

The existence of the light clusters ^{12}C and ^{16}O in the neck is an argument for the benefit of the rule taken in our calculations: to choose the probable points of the neck scission only between two any clusters. Otherwise, the scission takes place across the light nucleus, that is energetically less preferable. The idea of the clusterization of the neck can appear naturally from the other physical supposition. When the distance between the large clusters increases, the nucleon density between the large clusters decreases and the conditions for the so called percolation in the neck [31] are created. The actual percolation process has a random character, therefore, it is possible to expect, as was mentioned above, a coexistence of the unclusterized and clusterized phases in the neck.

4 Summary

Using our approach we obtained the verification for the bicluster mechanism of formation of the fission mode and clusterization of the neck of the fissioning nucleus. The arguments supported this conclusion are the following:

- For the $^{234}\text{U}^*$ nucleus the calculated TKE values are in agreement with some elements of the fine structure $TKE-M$ distribution of the FF.
- The obtained dependence of the potential energy of the fissioning nucleus $^{236}\text{U}^*$ on its elongation agrees with the previous calculations [25].
- The tendency similar to that shown in the calculations of Ref. [27, 28] for the height of the barriers for the FF separation as a function of the fissioning system elongation is observed.

The agreement of the calculations performed using the models with the clusterized and unclusterized necks proves the shapes of the fission

nucleus to be roughly similar in both cases. However, only the cluster model proves an explanation of the periodic structure (lines a and b in Fig. 4) linking it with the configurations where the length of the neck is a multiple of the α -particle diameter. Therefore, the model with the clusterized neck seems to be not only the simple algorithm for calculation but a mechanism actually taking place in nature. A small difference between the predictions of the two models means that both phases - clusterized and unclusterized - can coexist in the neck of the fissioning nucleus.

In our opinion, the most strong point that proves our model to be valid is the following. The correlation was established between clusterization of the fissioning nucleus and the fine structure in mass-energy distribution of the FF which is completely different from that produced by the proton odd-even effect.

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