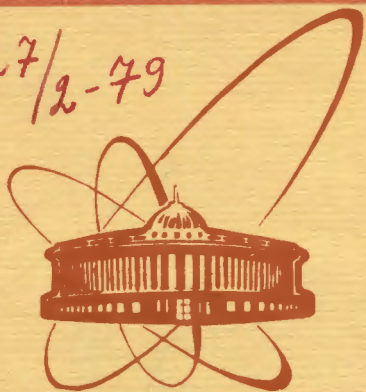


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**A STUDY  
OF SOME FISSION CHARACTERISTICS  
IN THE  $\alpha$ -PARTICLE-INDUCED FISSION  
OF TRANSURANIUM NUCLEI**

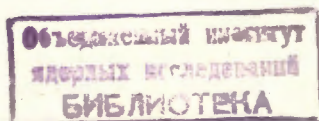
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A STUDY  
OF SOME FISSION CHARACTERISTICS  
IN THE  $\alpha$  -PARTICLE-INDUCED FISSION  
OF TRANSURANIUM NUCLEI

*Submitted to ЯФ*



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Дмитриев В.Д. и др.

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Изучение некоторых характеристик деления  
ядер трансурановых элементов  $\alpha$ -частицами

С использованием корреляционной методики измерения энергий двух фрагментов изучались массовые распределения осколков, образующихся при делении ядер  $^{237}\text{Np}$ ,  $^{238}\text{U}$ ,  $^{242}\text{Pu}$ ,  $^{243}\text{Am}$  ионами  $^4\text{He}$  в широком диапазоне энергий. Наблюдалось повышение выхода осколков симметричного деления составного ядра  $^{247}\text{Bk}$  при энергиях возбуждения  $E^* > 19$  МэВ, что интерпретируется как увеличение вероятности симметричного деления по мере приближения к ядрам с  $Z = 100$ . Приводятся данные по зависимостям полных кинетических энергий от массы осколков. Проводится также статистический анализ полученных результатов с целью определения разности потенциальных энергий симметричной и асимметричной деформаций.

Работа выполнена в Лаборатории ядерных реакций ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1979

Dmitriev V.D. et al.

E7 - 12150

A Study of Some Fission Characteristics in the  
 $\alpha$ -Particle-Induced Fission of Transuranium Nuclei

By using the angular correlation method consisting of measuring the kinetic energies of paired fragments, the mass distributions of fission fragments formed in the  $\alpha$ -particle-induced fission of  $^{237}\text{Np}$ ,  $^{238}\text{U}$ ,  $^{242}\text{Pu}$ , and  $^{243}\text{Am}$  were studied in a wide range of bombarding energies. An increase in the yield of symmetric fragments has been observed for the compound nucleus  $^{247}\text{Bk}$  at excitation energies  $E^* > 19$  MeV. This is interpreted as being due to an enhanced probability of symmetric fission as one approaches nuclei with  $Z = 100$ . Data on the mass dependence of the total kinetic energy are also presented. A statistical analysis of the obtained data is made with the purpose of finding the potential energy difference between the symmetric and asymmetric deformations.

The investigation has been performed at the Laboratory of Nuclear Reactions, JINR.

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The practical use of the fission process and the possibility of producing fission fragments in unusual states (e.g., neutron-rich nuclei, strongly deformed nuclei with high angular momenta) have made the study of fission one of the most intensively developing trends of nuclear physics. The study of the fission characteristics of nuclei gives valuable information about the properties of nuclear matter. Important results have lately been obtained in experiments to study the fission barriers of heavy nuclei. The existence of the so-called double-humped barrier determined by the shell corrections to the liquid drop fission barrier, has been confirmed experimentally. The knowledge of the fission barrier heights for heavy and superheavy elements allows one to predict their properties including their stability against spontaneous fission. The question of nuclear stability is, in principle, very important for the synthesis and search of new superheavy elements, whose stability is entirely determined by the shell term in the fission barrier. Information about the fission barrier structure is usually obtained by studying the energy dependence of the fission cross section, which in turn can be obtained from the mass distribution of the fission fragments. The mass distributions are usually the best known characteristics of fissioning nuclei. The asymmetric form of the mass distributions of fragments from spontaneous fission, involving an increase of the yields of fragments in the mass region  $A_f^1 \sim 140$  and in the region of the complementary fragment mass ( $A_f^2 = A_{C.N.} - A_f^1$ ), remains also in fission from low excited states. These "two-humped" mass distributions are interpreted as being due to the influence of closed shells on the yields of fission fragments. An exception

is the mass distribution of fragments from the spontaneous fission of the fermium isotopes  $^{258}, ^{259}\text{Fm}$  which fission mostly symmetrically. This fact is explained by a great probability of fission to two fragments of equal masses in the region of  $Z=50$  and  $N=82$ <sup>17</sup>. Another exception is the nucleus  $^{226}\text{Ra}$ , for which the symmetric fission contribution is quite significant even at low excitation energies, which leads to the appearance of a third peak in the mass distribution in the vicinity  $A_{C.N.}/2$  (e.g.  $^{72}$ ). The interpretation of the character of the fission fragment mass distributions and their change with excitation energy is based on the assumption of the existence of two modes of fission, viz. symmetric and asymmetric ones. However, at present there is no common opinion concerning the reasons why the nuclei undergo one or the other mode of fission.

At present there exist two popular viewpoints concerning the nature of symmetric and asymmetric fission. One is based on the assumption of a difference in the barriers for the two types of fission. Calculations of the nuclear potential energy, taking the Strutinsky shell correction into account, show that on the outer barrier there is a great instability with respect to octupole deformations, leading to a decrease of the asymmetric fission barrier.

Another interpretation assumes that the fission fragment mass distribution is shaped after passing the saddle point on the descent from the barrier to the scission point and is determined by the final states, namely, the level densities in the fragments at scission.

The presently available experimental results do not allow one to make an unambiguous conclusion about the mechanism of the formation of the fragment mass distributions at low excitation energies.

The purpose of the present work was to measure the mass distributions of the fission fragments formed in the  $\alpha$ -particle-induced fission of  $^{237}\text{Np}$ ,  $^{238}\text{U}$ ,  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  in the energy range of 24-36 MeV, so as to be able to study the dependence of the symmetric and asymmetric types of fission on excitation energy.

## EXPERIMENTAL PROCEDURE

The mass distributions of the reaction products were obtained by the angular correlation method consisting of simultaneously measuring the energies of paired fragments. This method allows one to kinematically separate the processes corresponding to a full angular momentum transfer from the bombarding particle (complete fusion reactions). To detect the fission fragments use was made of two surface-barrier Si(Au) detectors, whose energy resolution was not worse than 5%. The relative position of these two detectors and their angular apertures were calculated on the basis of the reaction kinematics so as to permit detection of all fission fragments satisfying the condition  $0.2 \leq A_1^1/A_1^2 \leq 3.0$ . This condition corresponds to the angular apertures of the first and second detectors  $2^\circ$  and  $10^\circ$ , respectively. The calibration of the detectors was performed using beams of the  $^{40}\text{Ar}$ ,  $^{84}\text{Kr}$  and  $^{136}\text{Xe}$  ions scattered elastically onto a thin  $^{197}\text{Au}$  target (evaporated onto a carbon backing  $30 \mu\text{g}/\text{cm}^2$  thick) at different angles. This provided the possibility of using the  $^{197}\text{Au}$  recoil nuclei for calibration. Under the assumption of a two-body mechanism of the compound nucleus decay, the fission fragment masses were calculated from the measured kinetic energies using the conservation of energy and momentum laws. A more detailed description of the processing of the experimental data has been given in ref.<sup>13</sup>. In our case the accuracy of the fission fragment mass determination was  $\pm 2.5$  a.m.u.

The experiments were carried out on an external  $\alpha$ -particle beam of the U-200 cyclotron of the JINR Laboratory of Nuclear Reactions. The variation of the  $\alpha$ -particle energy was performed by means of the beam extraction from different acceleration radii without worsening the energy resolution, which was about 1%. The  $\alpha$ -particle energies and the energy resolution of the beam were measured with the help of a Si(Au) detector located at  $20^\circ$  with respect to the beam direction and used also as a projectile monitor. The focusing of the beam on the target was achieved with the help of a collimator made of three carbon diaphragms  $2 \times 9$ ,  $2 \times 9$  and

3x9 mm<sup>2</sup>, respectively, located at a distance of 10 cm from each other. The last diaphragm was at a distance of 10 cm from the target.

### EXPERIMENTAL DATA ON MASS DISTRIBUTIONS

Figures 1 and 2 show contour diagrams of the total c.m. kinetic energies (TKE) of fission fragments as func-

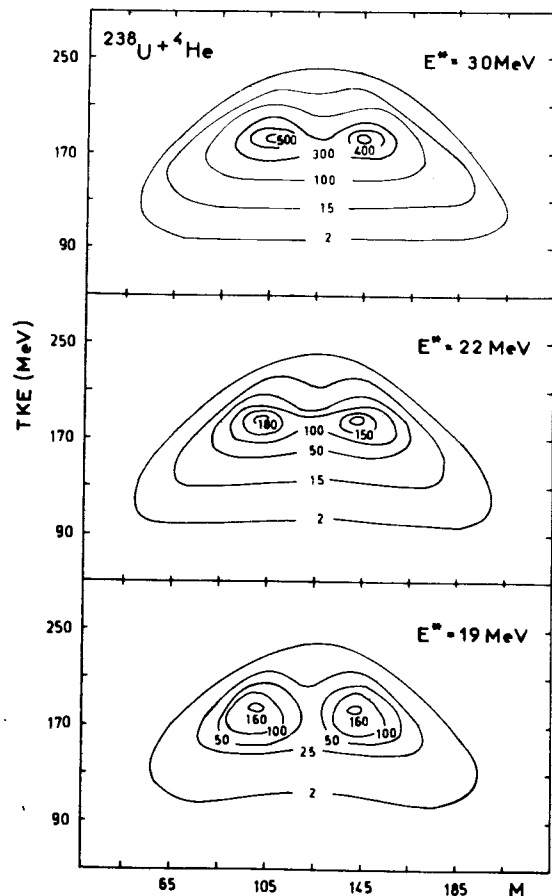


Fig. 1. Total kinetic energy vs fragment mass contour diagrams for the reaction  $^{238}\text{U}(^4\text{He},f)$ , at three different values of the compound nucleus excitation energy.

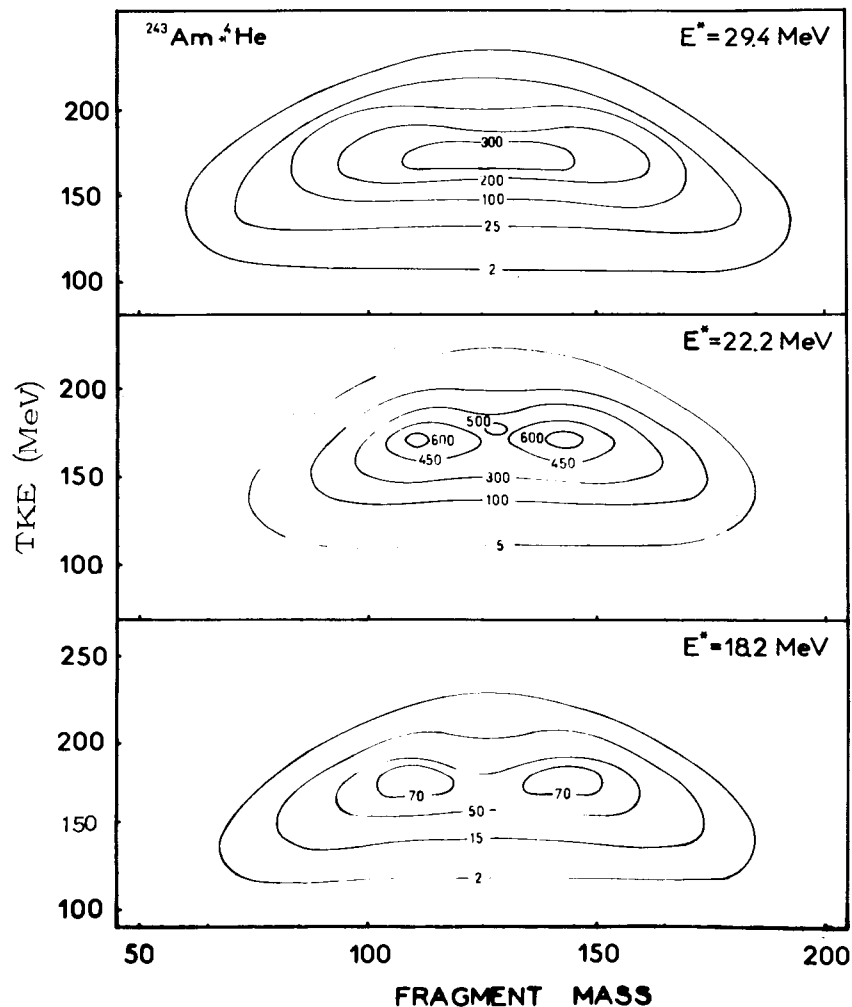


Fig. 2. Same as in fig. 1, for the reaction  $^{243}\text{Am}(^4\text{He},f)$ .

tions of their mass numbers for two reactions for different energies of the bombarding  $\alpha$ -particles. At minimum excitation energies, i.e., at  $\alpha$ -particle energies close to the Coulomb barrier of the reaction, two peaks manifest themselves distinctly in all cases, a fact being due to the influence of nuclear shells. As the excitation energy

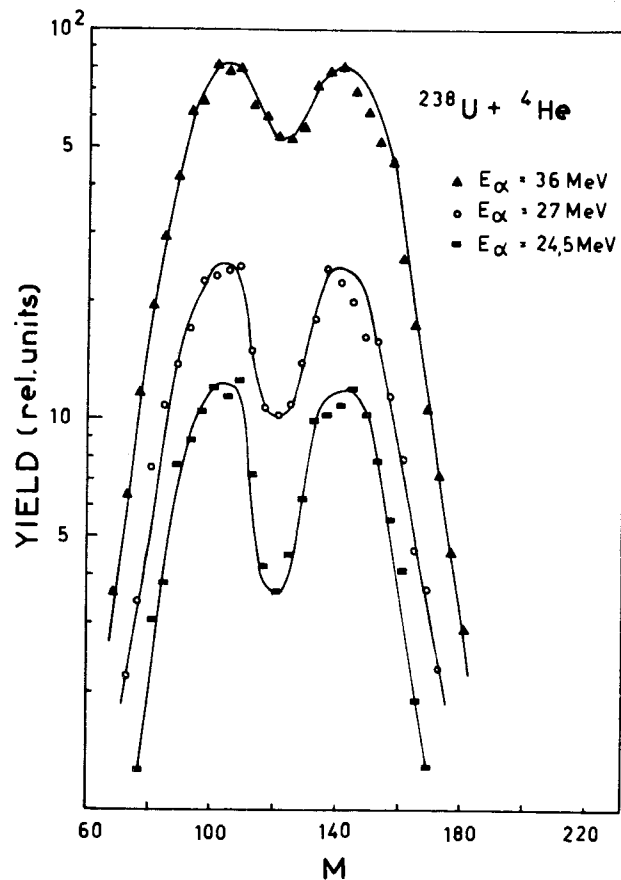


Fig. 3. Mass distributions of fission fragments formed in the reaction  $^{238}\text{U}(^4\text{He}, f)$  at three different values of the bombarding energy.

of the compound nucleus increases, an increase of the contribution of symmetric fission is observed in all cases. In particular, in the case of the fission of the compound nucleus  $^{247}\text{Bk}$  a peak corresponding to symmetric fission is distinctly seen already at an energy  $E_\alpha = 25.4 \text{ MeV}$  and remains as the excitation energy increases up to  $E^* \leq 30 \text{ MeV}$ .

Figures 3-5 show the fission fragment mass distributions for four reactions. From fig. 5 one can see the

contribution from the symmetric fission of the nucleus  $^{247}\text{Bk}$ . In general, the behaviour of the fission fragment mass distributions in the reaction  $^{243}\text{Am}(^4\text{He}, f)$  reminds qualitatively of the picture observed earlier in the fission of  $^{226}\text{Ra}$ , induced by different particles, at various excitation energies.

Similar regularities manifest themselves in the plots of the average total kinetic energy as a function of mass, shown in fig. 6. It is noteworthy that the experimentally measured value of the total kinetic energy is somewhat lower than the true one as a result of neutron evaporation, which may change the initial direction of the velocity of the fragments. The true value of the total kinetic energy of the fragments can be obtained using the relation

$$\text{TKE} = \text{TKE}' \left( 1 + \frac{\bar{v}_3 A_4}{A_3 A_{\text{CN}}} + \frac{\bar{v}_4 A_3}{A_4 A_{\text{CN}}} \right),$$

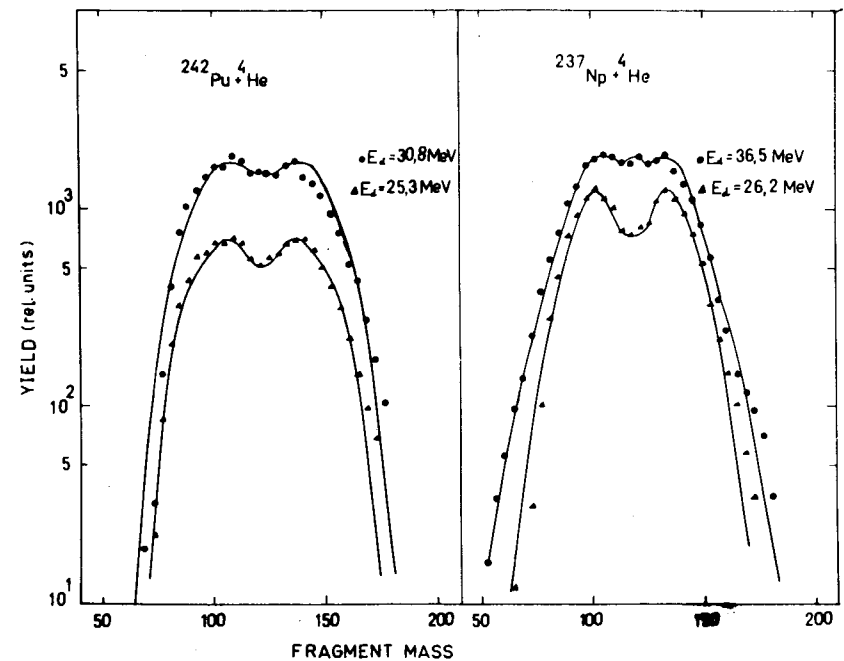


Fig. 4. Mass distributions of fission fragments formed in the reactions  $^{237}\text{Np}(^4\text{He}, f)$  and  $^{242}\text{Pu}(^4\text{He}, f)$ .

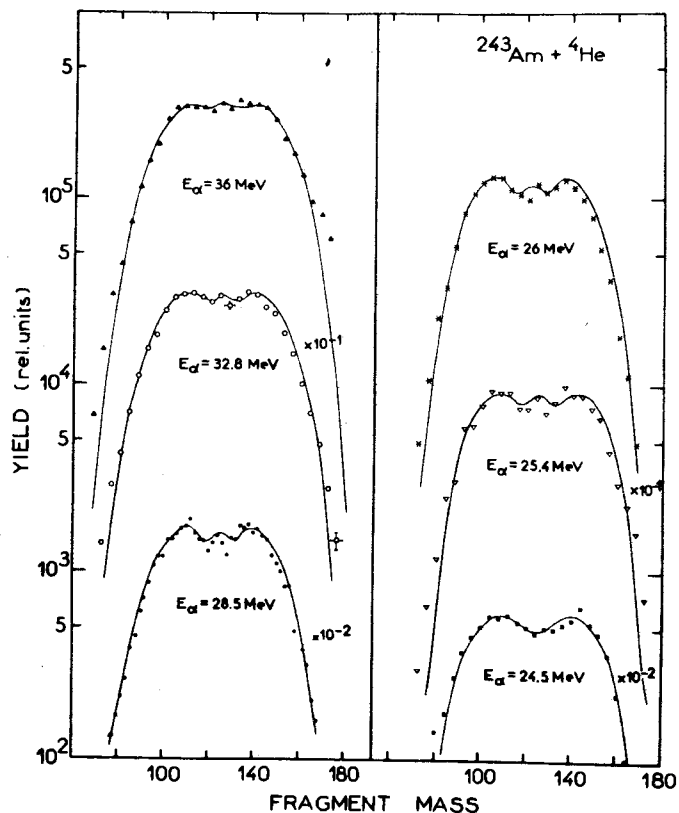


Fig. 5. Mass distributions of fission fragments formed in the reaction  $^{243}\text{Am} + ^4\text{He}$ , f) at six different values of the bombarding energy. The full lines drawn through the experimental points are calculated using the technique of ref. /4/.

where  $A_3$ ,  $A_4$ ,  $A_{CN}$  are the masses of the complementary fragments and the compound nucleus, respectively;  $\bar{\nu}_3$  and  $\bar{\nu}_4$  are the average numbers of neutrons evaporated by the respective fission fragments; and  $\text{TKE}'$  is the experimentally measured value of the total kinetic energy of the fragments. If one assumes that the neutrons are evaporated by the fragments, the average number of neutrons may be estimated from the energy balance of the reaction using the method described in ref. /3/. The values of the

total kinetic energy (TKE) obtained in this manner are presented in fig. 6. In the case of fission of the compound nucleus  $^{242}\text{Pu}$  the maximum value of TKE corresponds to asymmetric fission, when the mass of one fragment lies in the vicinity of  $A_f \sim 140$ . As the excitation energy increases, the TKE of fragments from asymmetric fission somewhat decreases, while no excitation energy dependence of the TKE of fragments from symmetric fission is observed. In the fission of the compound nucleus  $^{247}\text{Bk}$  the TKE values for fragments  $A_f \approx A_{C.N.}/2$  and  $A_f \approx 140$  show practically no difference and their changes with excitation energy are similar, too. As is known the fission fragment total kinetic energy, determined by the Coulomb repulsion, depends on the rigidity of the fragments with

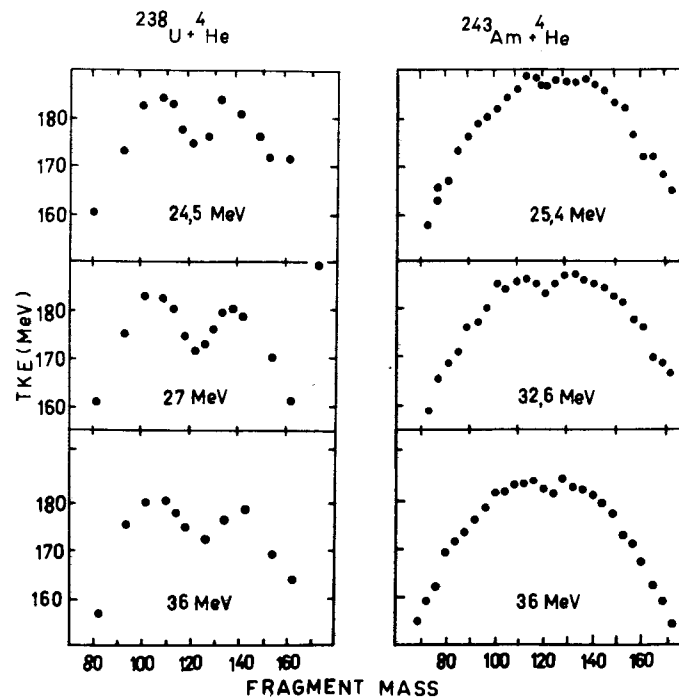


Fig. 6. Average TKE vs fragment mass for the fission of the compound nuclei  $^{242}\text{Pu}$  and  $^{247}\text{Bk}$  at different values of the bombarding energy.

respect to deformation, which, in turn, as is well known, depends on the degree of filling the nucleon shells. Thus, within the framework of the shell representation, the obtained result may be interpreted as due to a decreasing influence of nuclear shells, as excitation energy increases, on the fragment rigidity in the region of masses close to the magic ones. Within this representation the obtained results are very likely to be indicative of the fact that in the symmetric fission of  $^{247}\text{Bk}$  the influence of the closed  $Z=50$  shell manifests itself to the same extent as the  $N=82$  shell does for asymmetric fission.

### ANALYSIS OF EXPERIMENTAL RESULTS

In order to estimate the contributions from the symmetric and asymmetric modes of fission to the mass distributions, use was made of the technique to describe the mass distributions of spontaneous fission fragments, proposed in ref.<sup>/4/</sup>. Accordingly, the mass distribution of the fission fragments can be described with a high degree of accuracy by means of five Gaussians:

$$Y(A_f) = \frac{1}{\sqrt{2\pi}} \sum_{i=1}^5 N_i \sigma_i^{-1} \exp \left\{ -\frac{(A_f - A_i)^2}{2\sigma_i^2} \right\}.$$

The parameters of the Gaussians were obtained by a best fit to the experimental points. In addition, it was assumed that one half of the mass distribution was the mirror reflection of the other half. Fig. 7 shows an example of such an expansion of the mass distribution with the help of five Gaussians for the double- and triple- humped cases. The mass distributions calculated in this manner are shown by full curves in figs. 3-5. Fig. 8 shows the energy dependence of symmetric and asymmetric fission cross sections for the compound nucleus  $^{247}\text{Bk}$ , obtained with the help of the described procedure. In the same figure one can also see the energy dependence of the ratio of these cross sections. It is clear that as the excitation energy of the compound nucleus increases, the contribu-

tion from symmetric fission grows faster than that of the asymmetric one.

An analysis of the ratio of the symmetric to asymmetric fission probabilities was carried out in terms of the statistical model, according to which the ratio  $\sigma_f^s / \sigma_f^a = W_f^s / W_f^a$  can be expressed by the equation

$$W_f^s / W_f^a = C \exp \left\{ 2 \sqrt{a_s} (\overline{E^*} - E_s) - 2 \sqrt{a_a} (\overline{E^*} - E_a) \right\},$$

where  $C \sim 1$ , depending weakly on energy;  $\overline{E^*}$  is the nuclear excitation energy averaged over the evaporation cascade;  $a_s, E_s, a_a, E_a$  are the level density parameters of the nucleus and potential energies for symmetric and asymmetric types of fission, respectively.

The calculations showed that the ratio  $W_f^s / W_f^a$  is weakly dependent on the absolute values of the parameters  $a_s, a_a, E_s, E_a$ , but is very sensitive to the difference  $(E_s - E_a)$  and, also, to the character of the energy dependence of the level density parameters. The values of the potential energy difference for symmetric and asymmetric types of fission were calculated under the assumption that the level density parameters did not depend on excitation energy and provided that  $a_a = a_s = 1.1A/8$ . The values obtained are presented in the table.

As can be seen from the table, the quantity  $(E_s - E_a)$  exhibits no energy dependence, whereas it tends to decrease with an increase in the mass of the fissioning nucleus, thus enhancing the probability of symmetric fission. In the analysis of the mass distributions and in the calculations of the potential energy differences, made in the statistical approach, for symmetric and asymmetric deformations, we naturally did not consider the question as to where the mass distributions have been formed. If the mass distribution is formed at the scission point, then  $(E_s - E_a)$  is directly connected with the difference in the potential energy surfaces of the nucleus at this point<sup>/5-8/</sup> taking excitation energy into account. If, however, the mass asymmetry is determined at smaller deformations (at  $\beta \sim 0.8-1$ ), i.e., in the region of the second barrier<sup>/9,10/</sup>, then the relative probability of symmetric and asymmetric fission is actually determined by the probability of one or another fission configuration pene-



T a b l e

Compound nucleus	E* MeV	E* MeV	E <sub>s</sub> - E <sub>a</sub> MeV	E <sub>B<sub>s</sub></sub> - E <sub>B<sub>a</sub></sub> *	
				K=1.78 MeV	K=2.53 MeV
<sup>241</sup> Am	20.0	19.6	2.7		
	30.2	29.6	3.1		
<sup>242</sup> Pu	19.1	18.8	2.0	1.9	3.2
	21.6	21.0	1.8		
	30.4	29.3	2.0		
<sup>246</sup> Cm	19.4	19.1	1.8	1.2	1.6
	24.8	24.1	1.6		
	29.6	28.2	1.5		
<sup>247</sup> Bk	18.2	17.9	1.7		
	19.2	18.9	1.5		
	19.8	19.6	1.7		
	22.2	21.7	1.6		
	26.4	25.0	1.6		

trating to outer barrier. In the statistical approach these probabilities can be described with the help of level densities counted from different levels of the nuclear potential energy in the region of the second barrier. If this consideration is valid, the difference (E<sub>s</sub> - E<sub>a</sub>), as seen from the table, amounts to 1.5-3 MeV. It must be pointed out that a similar effect has theoretically been predicted by Möller<sup>9/</sup>.

Thus, the experimental study of the structure of the mass distribution of fragments, formed in the α-particle-

\* The quantities (E<sub>B<sub>s</sub></sub> - E<sub>B<sub>a</sub></sub>) were obtained in theoretical calculations by Möller<sup>9/</sup>, made for the ground states of even-even nuclei, for different values of the surface symmetry constant.

induced fission of heavy nuclei, confirms that the shell effects play an important role in the formation of the fragment mass distributions of the investigated nuclei at excitation energies up to E\* ~ 30 MeV.

In the case of fission of the compound nucleus <sup>247</sup>Bk the yields of the fragments corresponding to the symmetric and asymmetric modes of fission are almost equal at excitation energies E\* > 19 MeV and a quasitriple mass distribution is observed, which is interpreted as an in-

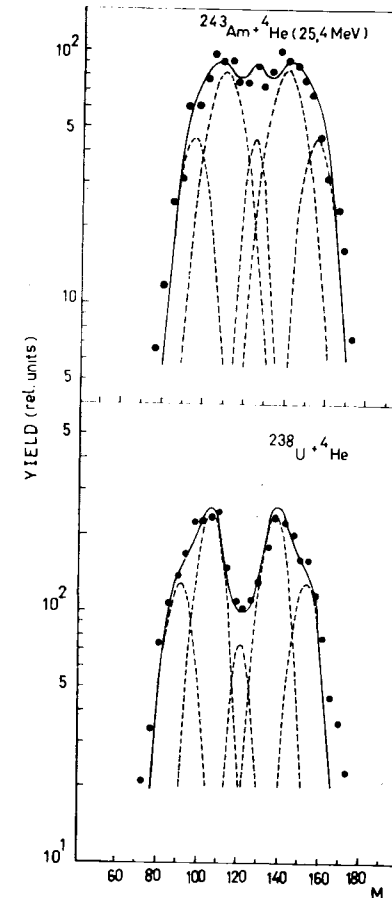


Fig. 7. An example of the expansion of mass distributions with the help of five Gaussians.

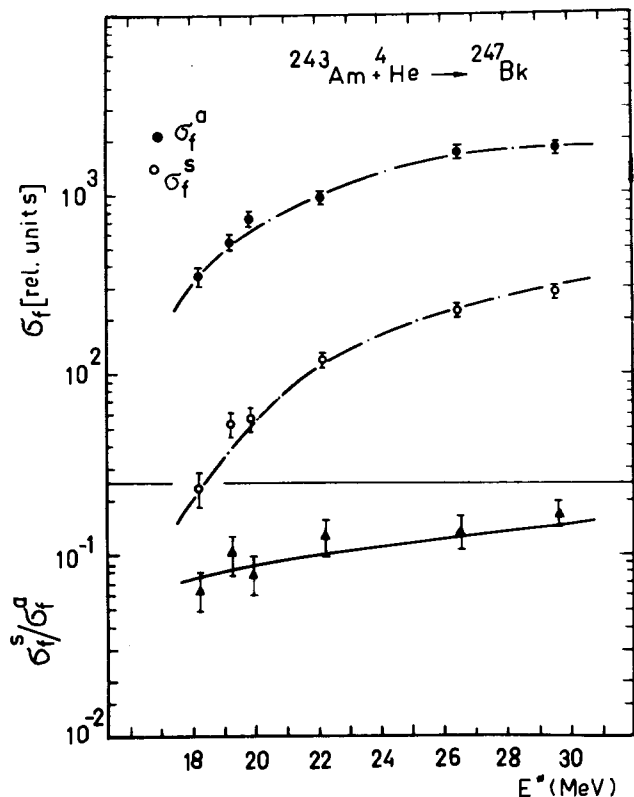


Fig. 8. The probabilities of symmetric and asymmetric modes of fission and their ratio as a function of excitation energy for the reaction  $^{243}\text{Am}(^4\text{He}, f)$ . In the lower part of the figure the full line corresponds to calculations made with  $(E_s - E_a) = 1.6 \text{ MeV}$  and  $a_s/a_a = 1$ .

crease of the symmetric fission probability as one approaches nuclei with  $Z = 100$ .

The total kinetic energy of fragments from the asymmetric fission of  $^{242}\text{Pu}$  is about 10 MeV higher than the TKE of the symmetric fission fragments, which is in agreement with previously obtained results and data from measurements of the kinetic energies of spontaneous fission fragments. However, in the case of fission of the compound nucleus  $^{247}\text{Bk}$  the total kinetic energies of fission frag-

ments from the symmetric and asymmetric modes of fission only slightly differ. One can assume that the increase of the average total kinetic energy of nearly symmetric fragments is due to the  $Z = 50$  shell, which, in the case of fission of the heavier nuclei (e.g., fermium isotopes) significantly changes both the mass distribution and the TKE.

The statistical analysis of the relative probabilities of the symmetric and asymmetric modes of fission and their change with excitation energy made under the assumption that the mass distribution is determined by the landscape of the nuclear potential energy surface has shown that the quantity  $(E_s - E_a)$  lies between 1.5 and 3 MeV and decreases with an increase of the mass of the fissioning nucleus. If such a tendency remains in the region of the heavier nuclei, this should result in enhanced yields of symmetric fission fragments. This has been confirmed by the studies of mass distributions of fragments from the spontaneous fission of  $^{258,259}\text{Fm}^{1/2}$ , where symmetric fission dominates.

In conclusion the authors wish to express their sincere thanks to Professor G.N. Flerov for interesting discussions and valuable comments in the course of performing the present investigations. Thanks are also due to H.Sodan, M.G.Itkis, V.V.Pashkevich, V.A.Rubchenya and Yu.A.Selitsky for helpful discussions. The authors are also grateful to the U-200 cyclotron crew for providing the monochromatic  $\alpha$ -particle beam of required parameters. The assistance of T.S.Salamatina, A.M.Rodin and Z.D.Pokrovskaya in acquiring and processing the data is also gratefully acknowledged.

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