Краткие сообщения ОИЯИ №3(66)-94 *JINR Rapid Communications No.3(66)-94*
УДК 539.173

MULTIMODAL FISSION OF NEUTRON-DEFICIENT NUCLIDES OF Th AND Ac

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Investigations of low-energy fission in reactions $^{204,208}Pb$ and $^{203}Tl + {}^{16}O$ have been performed with the help of the double-arm time-of-flight fragment spectrometer on the U-400 cyclotron beam, FLNR, JINR. The goal is to obtain experimental information on multimodal structure of mass-energy fragment distributions in the field of *A* fissionable nuclei earlier unstudied. For the first time, with such purpose heavy ion-induced reactions have been used. In connection with the experimental data obtained the influence of nucleon nuclei composition on principal fission modes — symmetric and asymmetric ones (Y_s / Y_a) — are discussed.

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

Мультимодальное деление нейтронодефицитных нуклидов Th и Ac

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На пучке циклотрона У-400 ЛЯР им.Г.Н.Флерова (ОИЯИ) с помощью двухплечевого времяпролетного спектрометра осколков проведены исследования низкоэнергетического деления в реакциях $^{204,208}Pb$ и $^{203}Tl + {}^{16}O$ с целью получения экспериментальной информации о мультимодальном строении массово-энергетических распределений осколков в неизученной ранее области *A* делящихся ядер. Впервые с такой целью использованы реакции с тяжелыми ионами. В связи с полученными экспериментальными данными обсуждается влияние нуклонного состава ядер на соотношение основных мод деления — симметричной и асимметричной.

Работа выполнена в Лаборатории ядерных реакций им.Г.Н.Флерова, ОИЯИ.

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One of the important results obtained in the fission physics during the last several years is the experimental discovery of multimodal structure of fragment mass and energy distributions of low energy fission nuclei in the region of Pb [1] and of the spontaneous fission of nuclei in the vicinity of Fm [2] as well as the establishing of the genetic relation of this phenomenon with structural (shell) peculiarities of the deformation potential energy surface of a nucleus fissioning along the mass asymmetric coordinate [3].

The unstudied earlier region of transient nuclei between At and Th seems to be rather promising for further investigation of the formation mechanisms of mass, energy and charge distributions of fragments in general and of the multimodal fission in particular. Just in this region one can expect a sharp change in the properties of mass, energy and charge distributions of fission fragments depending on the nucleon composition and excitation energy, i.e., the transition from symmetric to asymmetric fission and related changes in the barrier heights ratio of mass-symmetric and mass-asymmetric fission modes, the length and duration of nucleus motion from the saddle point to the scission point. That is why the experimental data on the character and scale of these changes should be critical for testing the theoretical description of dynamic and static aspects of the fission process.

Unfortunately, the advance into the indicated region of nuclei is hampered by a serious experimental problem caused first of all by the absence of stable nuclides with Z = 84—87 which could be used as target nuclei in reactions with light charged particles and neutrons that are traditionally used for the investigation of the fission process at low energies $E^* \leq$ 20—30 MeV.

The present study attempts to apply for the discussed purpose reactions with heavy ions. Their research object is usually very neutron deficient nuclides. The main difficulty in the realization of the given possibility is related with the production of nuclei with an excitation energy $E^* \leq$ 20—30 MeV which corresponds to the energy of ions near and below the Coulomb barrier $E_i \leq B_c$ and consequently with a rapid decrease of the fission cross section at decreasing E_i (when decreasing E^*, one achieves a better separation of fission modes, which is especially important when one of them dominates, as is expected in our case). According to the idea, on which the cold synthesis [4] method is based, the smallest excitation energy on account of the Q-reaction is achieved in the case of using as a target the double magic nuclei near ^{208}Pb and ions of ^{12}C, ^{16}O.

In this connection we have chosen for the first experiments the reaction ^{208}Pb(^{16}O, f) for which the fission cross sections, distribution of ^{224}Th compound nuclei angular momenta and spectra of pre- and post-neutrons

[5,6] are already known. The two other selected reactions, $^{204}Pb(^{16}O, f)$ and $^{203}Tl(^{16}O, f)$ lead to the formation and fission of compound nuclei ^{220}Th and ^{219}Ac and along with that satisfy the criterion of minimum excitation energy.

Note that at present the GSI realized another opportunity of studying the low-energy fission of transient nuclei and the use of relativistic secondary beams of radioactive nuclei of the ^{222}Th, ^{225}Th type and of their fission in the process of electromagnetic dissociation on an active target [7].

In this research, measurements were performed with a two-arm time-of-flight spectrometer of fragments «DEMAS-2» on the extracted ion beam of the U-400 cyclotron of FLNR, JINR. The spectrometer consists of two wide-aperture position sensitive avalanche counters providing the registration of fragments within the solid angle of about 0.2 steradian for each arm and of start-up parallel-plate avalanche counters [8].

Masses and energies of fragments were deduced from a set of experimentally measured values $T_{1,2}$ of the time-of-flight and $X_{1,2}$-, $Y_{1,2}$ coordinates of reaction products entry into the detector. The two-body processes in this case were selected on the condition $\overline{\Theta}_1 + \overline{\Theta}_2 = 180° \pm 3°$, where $\overline{\Theta}_{1,2}$ are the divergence angles in the center of mass system.

The spectrometer was calibrated by means of a thin source of spontaneous fission fragments of ^{252}Cf and provided the time resolution of about 200 pico sec. and the mass resolution $\delta M/M \cong 2\%$ (the peak/valley ratio in the mass distribution of $^{252}Cf \cong 25$). In these experiments there were used targets of enriched isotopes of ^{204}Pb, ^{208}Pb and ^{203}Te with a thickness of $\cong$ 150—200 $\mu g/cm^2$ sputtered on a substrate of Al_2O_3 with a thickness of 30—50 $\mu g/cm^2$. The beam intensity and its energy spread were $\cong 1 \times 10^{11}$ pps and 0.5%, respectively.

The data on the energy characteristics of reactions investigated and on experimental values of the first momenta of fragments mass and energy distributions are presented in the Table, where E_i — the ion energy in the laboratory system of coordinates, E^* — the excitation energy of the complex compound nucleus, E^*_{sp} — the excitation energy of the fissioning nucleus in the saddle point, $\langle l^2 \rangle$ — the mean square of the transferred angular momentum [9]. E_K — the average kinetic energy of fragments, σ^2_E and σ^2_M — the fragments energy and mass dispersions. Note that the large

Table

Reaction	Comp. nucleus	E_i, MeV	E^*, MeV	E_f, MeV	E^*_{sp}, MeV	$\langle l^2 \rangle$, $\hbar^2$	$\langle E_k \rangle$, MeV	σ^2_{E}, MeV^2	σ^2_M, (amu)^2
$^{16}O + ^{208}Pb$	^{224}Th	85	32,4	7,2	25,2	320	164,3±1,0	122±6	185±8
		77	25,9		18,7	170	164,8±1,8	101±9	172±10
$^{16}O + ^{204}Pb$	^{220}Th	85	43,3	8,8	25,5		163,3±1,0	131±7	172±7
		77	27,8		19,0		163,7±2,2	107±11	157±13
$^{16}O + ^{203}Tl$	^{219}Ac	85	36,7	10,0	26,7		162,0±0,9	127±5	164±6

ion energy lies near the Coulomb barrier and the small one — by several MeV lower. Thus in the second case the fission cross section was 30—40 times smaller which determined the difference in the number of registered events (2—3)×10^4 for E_i = 85 MeV and (2—4)×10^3 for E_i = 78 MeV. Note also that at barrier energies ($E_i < B_{fus}$) a compound nucleus is produced with $\langle l^2 \rangle$ values typical for reactions with light charged particles which, at the above indicated energies E^*, occur higher the Coulomb barrier. This is of course a favourable circumstance for the comparative analysis of experimental data on low-energy fission.

Figures 1 and 2 present the results of measurements as complete mass distributions $Y(M)$, symmetrized with respect to $M = A/2$ and normalized

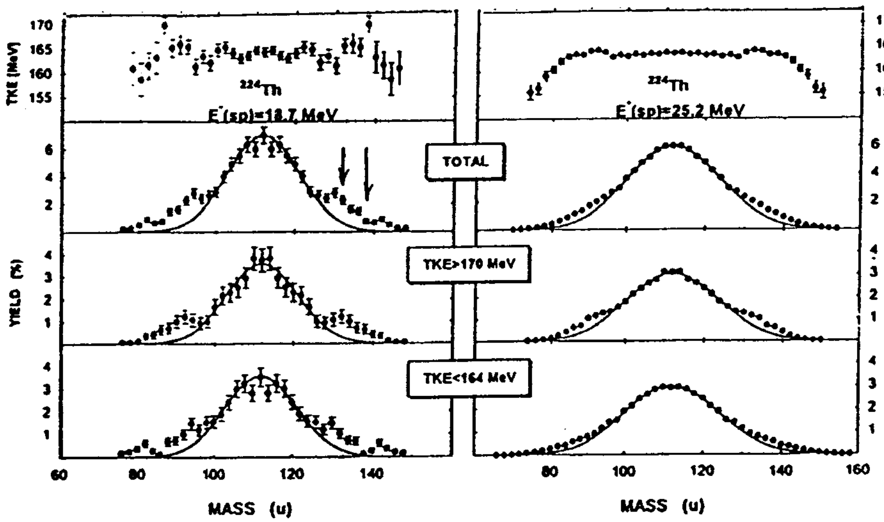

Fig.1 Fragment mass and energy distributions for ^{224}Th

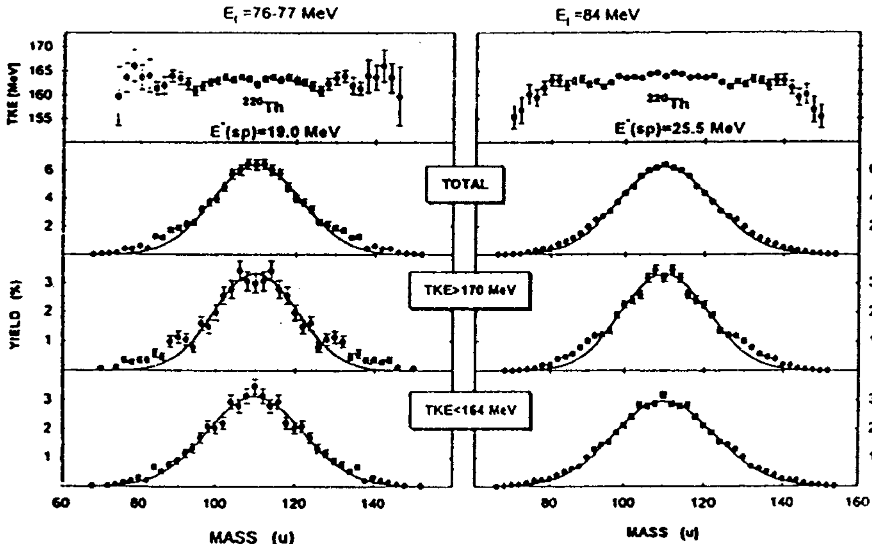

Fig.2. Fragment mass and energy distributions for ^{220}Th

to 200%, and average kinetic energies of paired fragments $\overline{E}_k(M)$ with masses M and A-M for ^{224}Th and ^{220}Th.

The curves in figs.1 and 2 present the results of the three-component analysis (according to the number of main fission modes, see above), presuming the Gaussian shape of partial distributions $Y_i(M)$. The indexes i denote: $i = s$ — mass-symmetric fission mode with an average mass of fragment $\overline{M} = A/2$, $i = a0$ and $a1$ — mass-asymmetric fission modes with $\overline{M}_{a0} = 139$ and $\overline{M}_{a1} = 132$, respectively.

Figure 3 presents exclusive yields of heavy fragments for the mass asymmetric nucleus of ^{224}Th: the broken curves show the components $Y_{a0}(M)$ and $Y_{a1}(M)$ taken separately, the solid ones —the sum $Y_a(M) = Y_{a0}(M) + Y_{a1}(M)$, experimental points present the difference between the observed total yields and the curves describing the symmetric mode distribution. The yield of $Y_a(M)$ increases noticeably with the cooling of the fissioning nucleus. The mass symmetric mode is dominating in the case of lighter nuclei. In full correspondence with subsequent outcome for the dependence of the $Y_i(M)$ spectrum on the excitation energy and the nucleon content of fissioning nuclei, there changes the height of the $\overline{E}_k(M)$ maximum in the part of figs.1,2, the apparition of which is explained by the

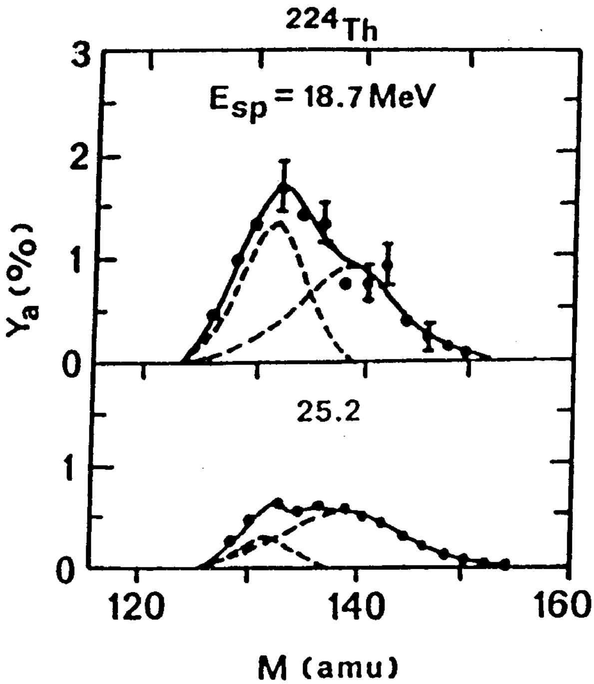

Fig.3. The yield of asymmetric modes Y_{a1} and Y_{a0} for ^{224}Th

ratio for separate fission modes $\overline{E}_k^{s} < \overline{E}_k^{a0} < \overline{E}_k^{a1}$ and that in accordance with this inequality at the discrimination of events with small fragment kinetic energies there takes place the enrichment of the mass distribution of fragments with asymmetric modes (at the fission of ^{224}Th it becomes trihumped) and, on the contrary, at the discrimination of events with large fragment kinetic energies the mass distribution gets free of them and becomes the same as in the symmetric mode.

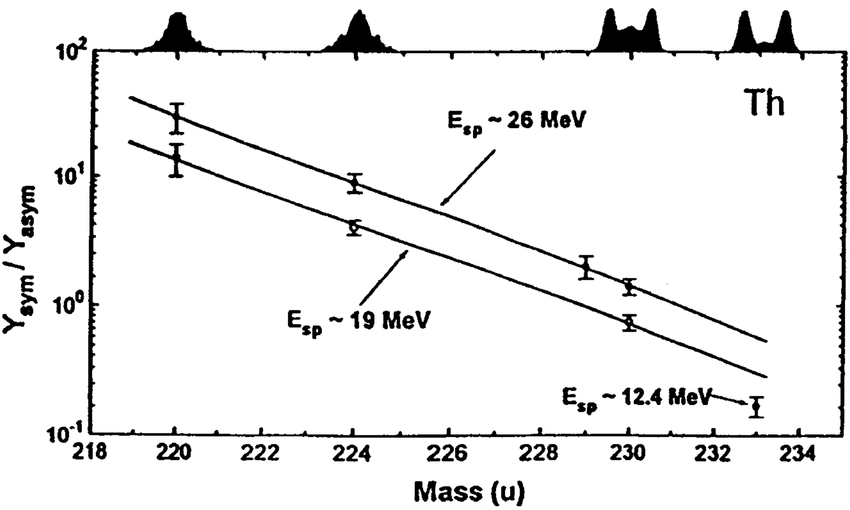

Fig.4. The ratio symmetric and asymmetric components as function mass number A. The data for ^{229}Th, ^{230}Th, ^{233}Th from Refs. 10, 11, 12

The statistical accuracy of the performed measurements, especially for the lower and more interesting energy ($\overline{E}^*_{sp} \cong 19$ MeV), is not sufficient for the deduction of numerous parameters, determining the yields $Y_i(M)$ and the exact characteristics of separate modes, first of all the average energies of fragments $\overline{E}^i_k$.

Nevertheless, even at this initial stage of investigations a rather favourable situation for the discussion of two interesting observables has built up. The first one is dependence of Y^t_a / Y^t_s on the number of neutrons N in the chain of Th isotopes. It turns out that for the reactions ^{3}He + ^{226}Ra → ^{229}Th [10] ^{4}He + ^{226}Ra → ^{230}Th and [10,11] there are experimental data on the mass distributions of fission fragments at close excitation energies E^*_{sp}. The totality of data on the ratio Y^t_a / Y^t_s deduced from the results of [10,11] and of this work is presented in fig.4. The significance of the scale of changes of $Y(M)$, corresponding to the values of Y^t_a / Y^t_s is explicitly demonstrated by means of upper part of Fig.4 which presents the mass distributions themselves for a lower average energy $\overline{E}^*_{sp} \cong 19$ MeV. The picture becomes still more impressive in case the things shown in Fig.4 are extrapolated to $A = 232$—233. It follows from the experimental data on the fission of ^{232}Th by γ-quanta and neutrons [12] that in the corresponding to their mass distributions there will be observed a substantial domination of the asymmetric component and they will have a dihumped shape which is traditional for the low energy fission of actinides.

The second most important factor determining the modal structure of the mass and energy distribution of the fragments is the experimental data about the kinetic energy spectrum of

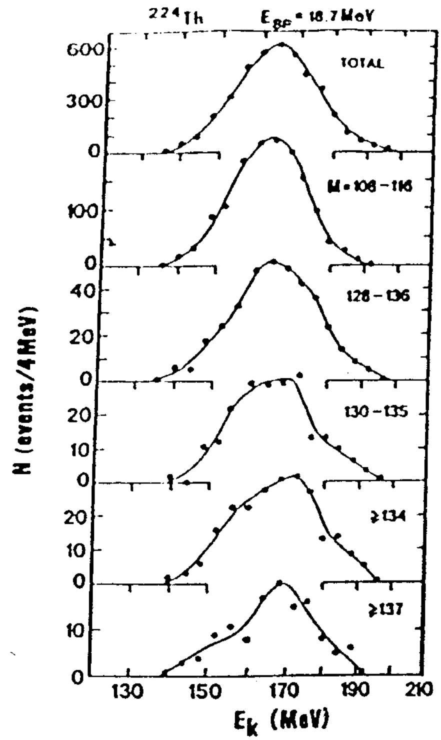

Fig.5.The fragment energy spectra ^{224}Th for different mass division

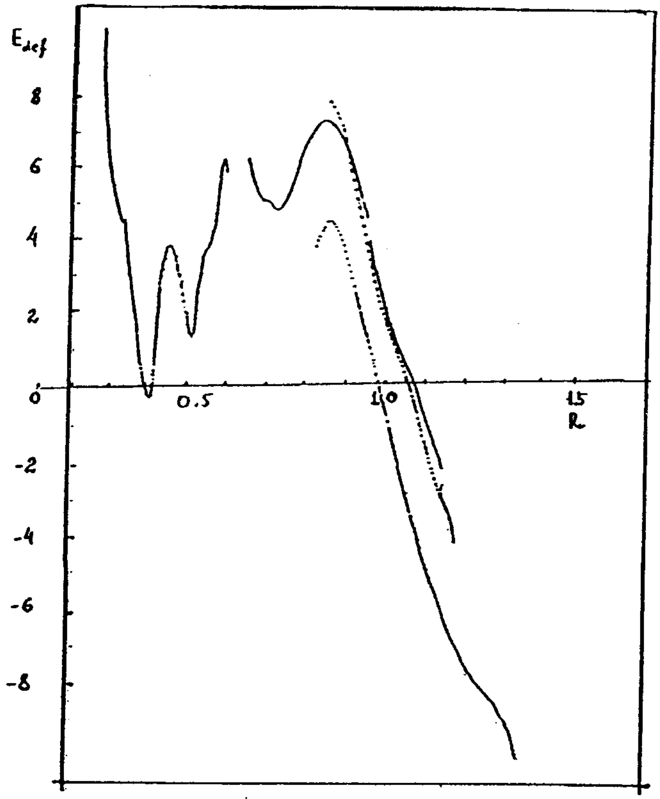

Fig.6. The calculations of deformation energy ^{224}Th as function of the distance between fragments

the fragments at different mass division, i.e., M_N / M_L. The presence in such spectra of several precisely defined groups of fragments with different kinetic energies for specific M_T / M_L ratios (e.g., two groups in the Fm range and three groups in the Pb range) proves this new concept in fission physics. In our case we have also analysed the kinetic energy spectra of ^{224}Th fission fragments having different mass, shown in Fig.5. The diagram demonstrates that despite the rather modest statistics we can observe in the $E_k(M)$ spectrum all the main regularities occurring in the Pb-Fm range, i.e., three groups of fragments corresponding to the three different configurations of fissile nuclei at the scission point — one elongated (symmetrical fission) and two more compact configurations, determined by higher kinetic energy values of the fragments. This previous result certainly requires further investigation and more statistics.

In this respect, the work performed by us within the micro-macro approach [3] to the calculation of the deformation energy is of great interest and also predicts possible existence of 3 division modes of the ^{224}Th

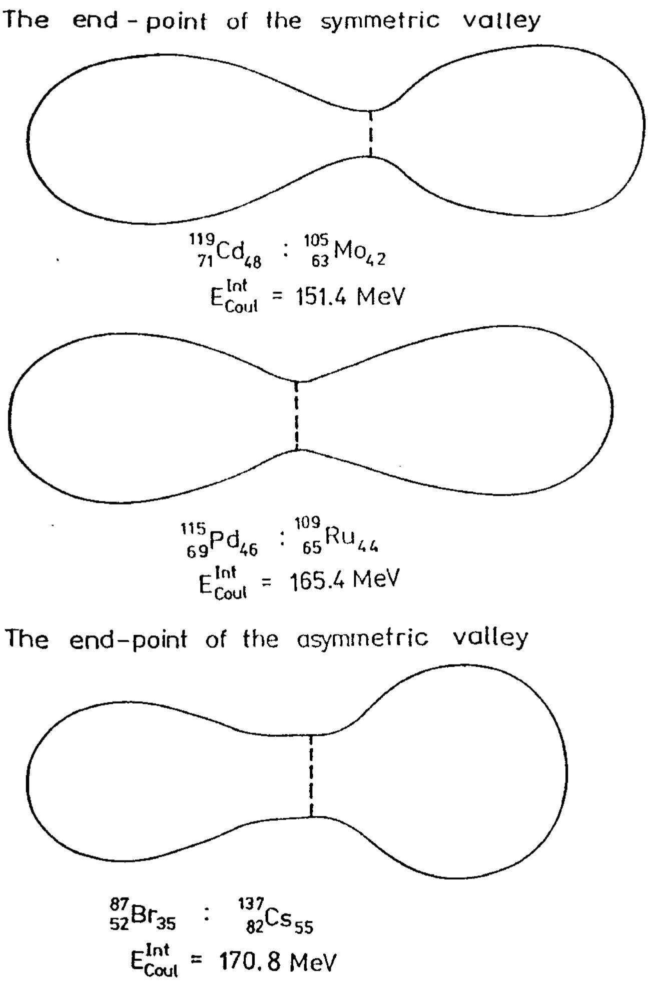

Fig.7

nucleus. Figures 6 and 7 show the results of these calculations as a dependence of the deformation energy on the distance between the fragment centres during the process of the nucleus movement from the ground state to the scission point and as different configurations of the fissile nucleus at the moment of rapture corresponding to different fission modes. The obtained kinetic energy E_k^i values for different fission modes of ^{224}Th on the whole correctly reflect the $E_k^s < E_k^{a0} < E_k^{a1}$ ratio observed in the experiment.

Conclusion

Mass and energy distributions of low-energy fission fragment ($E^* \cong$ 20—30 MeV) in the reactions ^{204,208}Pb and ^{203}Te + ^{16}O leading to the compound nuclei ^{220}Th, ^{224}Th and ^{219}Ac are investigated.

As shown by the experiment, fragment mass and energy distributions for the studied nuclei have a clearly defined multimodal structure completely fitting the theoretical predictions given in the work for this nuclei range.

Analysis of the experimental results obtained in these measurements has led to the revelation of a new regularity, namely, the fact that the ratio of yields from the mass-symmetric and mass-asymmetric modes of fission changes by about two orders of magnitude in going from ^{230}Th to ^{220}Th. This result indicates that (by varying only the number of neutrons in a fissioning nucleus, say, in Th (or Ac, Ra, Fr) we can investigate all possible transformations of the fission fragment mass and energy distributions, from the mass-symmetric ones typical for nuclei lighter than lead, and the three-humped ones observed in the case of Ra, and Ac, to the traditional mass-asymmetric distributions which characterise the actinide region. In addition this makes it possible to establish a quantitative relationship between these transformations and the structural features of the potential energy of surfaces and the dynamics of collective motion.

One of us (V.V.Pashkevich) thanks IN2P3 for grant that made possible his stay in Bordeaux.

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Received by Publishing Department
on July 20, 1994.