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INVESTIGATION OF PHOTONUCLEAR REACTIONS LEADING TO SPONTANEOUSLY FISSIONING ISOMERS

ААБОРАТОРИЯ ЯДЕРНЫХ РЕАНЦИЙ

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#### INTRODUCTION.

Nuclei in excited states with very high probability of spontaneous fission and strongly forbidden radiative transitions are called spontaneously fissioning isomers.

The experimental evidance currently available testifies that such isomeric states are characteristic of heavy nuclei.

The study of spontaneously fissioning isomers consists in accumulating experimental data on the properties of metastable states and the characteristics of different nuclear reactions leading to these states. An analysis and systematisation of such data, based on theoretical predictions, allow one to draw conclusions about the nature of the phenomenon.

## I. DISCOVERY OF FISSIONING ISOMERS AND THE MAIN STAGES OF THEIR STUDY.

Spontaneously fissioning isomera were first observed in 1961 at the JINR Laboratory of Nuclear Reactions (Dubna, USSR).At that time the 3-m cyclotron of this Laboratory produced intense heavy ion beams and experiments on the synthesis of the spontaneously fissioning isotopss of transuranic elements were started. In the bombardment of an 238U target by 160 ions an unknown epontaneously fissioning activity with a half-life of 0.014 sec was observed 1). The observation of this activity was extremely unexpected since the known isotopes with halflives exceeding I min and with a weak spontaneous fission branching were known to be produced only in the reaction 2360+160. Therefore, the only possible interpretation of the effect observed could be that an isomeric state of some known isotope underwant fiasion in that case. By using cross bombardments ( the appropriate targets were bombarded by alpha particlea and deuterons<sup>2)</sup> as well as by neutrons<sup>3)</sup>) it has been shown that the isomeric state of the 242 Am macleus undergoes spontaneous fission with a half-life of 0.014 sec. The estimates made on the basis of the systematics of spontaneous fission lifetimes suggest that the spontaneous fission half-life of this nucleus in the ground state lies between 1012 and 1014 years. Consequently, the spontaneous fission probability of

the isomeric state is about a factor of 10<sup>21</sup> higher than that for the ground state.

The subsequent experiments using the cyclotron of the JINR Laboratory of Nuclear Reactions resulted, in 1964, in the observation of another spontaneously fissioning activity with a helf-life of 0.00I see in the reaction  $^{242}\text{Pu+IIB}$  (ref.<sup>[0]</sup>). The identification of this activity made at the Berkeley Radiation Laboratory showed that such a lifetime was characteristic of the two spontaneously fissioning isomers,  $^{240}\text{Am}$  and  $^{244}\text{Am}$  ref.5).

At that time a new concept of sponteneous fission of muclei in isomeric states became firmly established in nuclear physics. The studies of this phenomenon were initiated in many laboratories throughout the world.

In 1985-87, physical experiments were primarily simed at the determination of the main properties of the known americium isomers, their excitation energies and spins.

Information about these characteristics is generally gained from the spectra of gamma-rays, conversion electrons or alphe perticles emitted from the excited states. However, up to now spontaneous fission remains the only observable decay mode of fissioning isomers. In principle, the fission process can yield some data on the characteristics of isomeric states. For instance, the excitation energy of an isomeric state should be associated with the mean number of fission neutrons. The angular distribution of the fragments of polarized nuclei is determined by the spin of the initial state. However, it is rather difficult to extract this kind of information from the fiseion process experimentally, especially as the production cross section for all known spontaneously fissioning isomers is by a factor of 104-106 less than the induced fiscion cross section. As a result, at that period of time the studies of spontaneously fissioning isomers were restricted to the investigation of different reactions leading to the formation of isomeric states.

In 1965, at the Niels Bohr Institute in Copenhagen measurements were made of the threshold of the reaction

 $^{241}$ Fu(p,2n), which led to the spontaneously fissioning isomer,  $^{240}$ Am (ref.<sup>6</sup>). This threshold turned out to be 3.15 MeV higher than the threshold of the reaction producing the  $^{240}$ Am nucleus in the ground state. The difference between the thresholds was interpreted as the energy of the isomeric state. It was shown subsequently  $^{7)}$  that the measured excitation functions were treated not correctly enough in ref.<sup>6</sup>), which resulted in the higher threshold value ( a detailed consideration of this will be given below). Nevertheless, the subsequent measurements indicated that the energy of the isomeric state was rather high ( up to 2.5 MeV).

In 1965-66, at the Institute of Atomic Physics in Bucharest and at the JINR Laboratory of Nuclear Reactions at Dubna measurements of isomer ratios were carried out ( the ratios of the production oross sections for the nucleus in an isomoric and in the ground states,  $\frac{G}{G_0}$ ) in different reactions leading to the  $\frac{242}{4}$  m spontaneously fissioning isomer 8,9. A large set of bombarding particles ranging from low energy neutrons to  $^{\rm IIB}$  ions, was used in these measurements. It has been found that the isomer ratio remains practically constant and equals 4 x 10<sup>-4</sup> as the momentum imparted to the nucleus changes from (2-3)h ( I MeV neutrons) to (I5-20)h (60 MeV IIB ions). Such a behaviour of the isomer ratios was the basis for the hypothesis that the value of the isomeric state spin should be small.

In 1966-67, at the Institute of Atomic Physics in Bucharest detailed studies of the excitation function of the reaction leading to the <sup>242</sup>An spontaneously fissioning isomer in radiative neutron capture were carried out. As the energy of the isomeric state was 2.5-3.0 MeV and the spin was low, the spontaneously fissioning isomer of <sup>242</sup>An could be expected to be formed at low neutron energies ( at an excitation energy of about 5.5 MeV and with the compound nucleus spin of 2 or 3), and the cross section for the reaction (n, f) would not very substantially with neutron energy. The measurements IO) have however shown the threshold character of the reaction,

i.e., a sharp increase in the excitation energy at a neutron energy ranging from 0.3 to 1.0 MeV, followed by a smooth decrease. Later on it has been shown that a similar shape of the excitation function is also characteristic of the reaction producing  $^{244}$  m in radiative neutron capture<sup>II</sup>). The initial part of the excitation function (at energies lower than 1.5 MeV) was analogous to the energy dependence of the cross section for fission of the  $^{241}$  Am and  $^{243}$  Am isotopes by neutroms.

The first studies were restricted to americium spontaneously fissioning isomere ( $^{240}Am$ ,  $^{242}Am$ ,  $^{244}Am$ ) with relatively long lifetimes. Since those isomers were odd-odd nuclei, it was important to establish whether or not analogous isomeric states existed in nuclei with even numbers of protons and neutrons. On the basis of the systematics of the spontaneous fission half-lives of nuclei in the ground state, one could assume that the isomeric lifetime for nuclei with an even number of nucleons would be shorter than that in the case of d americium isomers. Therefore; a search for fissioning isomere with half-lives of  $10^{-9} - 10^{-9}$  sec was of great interest.

To carry out searches for spontaneously fissioning isomera with lifetimes shorter than I  $\mu$ sec, a special technique was developed at Dubna based on measurements of the distance between the target and the decay point, i.e., of the flight path of the recoil nucleus. This technique was used to observe an about 10<sup>-7</sup> sec spontaneously fissioning isomer<sup>12</sup>. Subsequently this technique and its variants found extensive applications at the Niels Bohr Institute, where in the course of 1968-1970 a large number of new spontaneously fissioning isomers with lifetimes ranging from 10<sup>-9</sup> sec to 10<sup>-4</sup> sac were produced in the isotopes of U, Fu, Am and Cm<sup>13</sup>,<sup>14</sup>.

The existence of spontaneously fissioning isomers with different lifetimes in a wide range of nuclei has made it possible to extend substantially the studies of the properties of isomeric states. For the time being, studies along this line are under way in a number of scientific research centres in the USA (Los Alamos, Seattle, etc.) and

in the Federal Republic of Germany (Heidelberg)<sup>15-18)</sup>. Experiments carried out with light charged particles (p,d, <sup>4</sup>He) provided a large amount of information on the characteristics of nuclear reactions involving isomers (cross sections, isomer ratios and the isomer-to-prompt fission ratio) and permitted the establishment of some properties of the spontaneously fissioning isomers,

### II. THE MAIN PROPERTIES OF SPONTANEOUSLY FISSIONING ISOMERS.

Spontaneously fissioning isomers are observed in nucles ranging from uranium to berkelium. Searches for analogous isomeric states in the lighter nuclei have produced negative results<sup>19-21)</sup>. One can assume that the existence of fiesion isomers is specific for nuclei with comperatively low fission barriers, Fig. I shows the variation of the fission isomer half-lives with proton and neutron numbers in the nucleus. For comparison, a similar dependence is shown for the ground states of the same nuclei. One can see from fig.I that in the case of isomeric states the dependence of the half-life on neutron number has a parabolic shape for all nuclei. This dependence is characteristic of nuclei with both even and odd numbers of protons and neutrons, while for the ground states the dependence of the same shape is observed only for eveneven nuclei. As one nucleon is added to an even-even nucleus. the half-life increases by a factor of 103-104. The same increase is also observed when another nucleon is added and an odd-odd nucleus is formed. In a number of even-even Pu isotopes 22) two isomeric states are observed, which decay by spontaneous fission, the lower states having shorter halflives. In the case of odd nuclei, a couple of isomeric states (with half-lives of IOO and 900 nsec) were observed only in one nucleus, 237 Pu (refs. 13,14,23)).

The only observable decay mode of isomeric states is apontaneous fissica. Attempts to observe alpha and gamma radiation from isomeric states have been no success. The upper limits were obtained to be 0.01 of an alpha particle per

spontaneous fission event of the  ${}^{241}Pu$ ,  ${}^{240}An$  and  ${}^{242}An$  isomers<sup>24)</sup> and IO genma-rays for isomers in  ${}^{236}U$  (ref.<sup>25)</sup>) and in  ${}^{242}An$  (ref.<sup>26)</sup>).

The energy spectra of fission fragments from the isomeric states does not show any difference from the analogous induced fission spectrum at a low excitation energy 27,28.

The isomeric state energies determined from measurements of the thresholds of the production reactions for these states are about 2.5 MeV. No systematic differences are observed in isomeric state energies between nuclei with even and odd numbers of protons and neutrons or with changing 2 and A. In those Pu isotopes that have a couple of isomeric states have an energy of 3.5 MeV I3,17).

The independence of the isomer ratio of the momentum imparted to the nucleus, observed in the 242Am nucleus. indicates that, as it has been already noted, the spin of the isomeric state is low. The same situation occurs also for the majority of other spontaneously fissioning isomers. The experimental ratios of the isomer cross section to the induced fission one for the Pu, Am and Cm isotopes at the same excitation energy but with different projectiles (protons, deuterons, alpha particles) turn out to be close 1?). At the same time, in nuclei with a couple of isomeric states observed, the higher-lying state may have high spin. This is suggested by the projectile energy dependence of the ratio. of the production cross section for the <sup>238</sup>Pu nucleus in the second and first isomeric states 22) ( this ratio increases with increasing projectile energy), as well as by the angular distribution of fission fragments from the isomeric state ( in the case of the 236Pu isomer with a half-life of 35 need and an excitation energy of about 3.5 NeV a large anisotropy is observed in the angular distribution of the fission fragments 29). The lifetime measurement for the high-lying icomeric states yielded a value considerably larger than the lifetime of the low-lying isomeric state. This can be explained assuming the fission barrier for this state to be higher or wider compared with that for the lew-lying isome ic state.

The manifestation of the high spin in experiment allows one to assume that the second isomeric state is a two-quasiparticle one, and as a result of the decoupling of a pair of nucleons, a certain amount of energy is added to the fission barrier.

It has also been shown experimentally that though the energy of the isomeric state is 2.5-3.5 MeV, the formation of the nucleus in an isomeric state takes place preferentially at the excitation of the levels lying in energy not lower than at 5.5-6.0 Mey 10,11). The excitation energy dependence of the production cross section for the spontaneously fissioning isomer displays the same threshold shape as that of the induced fission cross section. This suggests a correlation between the processes of isomer production and induced fission. A similar correlation also exists at the radiative capture of thermal neutrons 30). The 24IAm nucleus undergoes fission by thermal neutron capture and at radietive neutron capture the <sup>242</sup>Am spontaneously fissioning isomer is produced. The 243 Am nucleus does not undergo fission by thermal neutron capture and no corresponding isomer (244Am) is formed in this case with a noticeable cross section.

### III. SPONTANHOUSLY FISSIONING ISOMERS AND FISSION BARRIER STRUCTURE.

The mentioned properties of spontaneously fissioning isomers cannot be explained in the framework of the commonly used soncept of an isomeric state. In fact, though the sharp increases in spontaneous fission probability can be explained by a high energy and low spin, the strong forbiddenness for gamme-transitions from the isomeric state remains still unclear. The correlation between the isomer production from section and that of induced fission is also difficult to understand. This correlation seems to be associated with the existence of a certain between the isomeric state from the main system of muclear levels.

One could assume that the unusual properties of spontaneously fissioning isomere suggest the existence of a new

kind of nuclear isomeries. In 1964, G.N.Flerov and A.Bohr advanced the hypothesis that the nucleus in its isomeric state is characterized by extraordinarily large equilibrium deformation 3J/x. This hypothesis was confirmed by the theoretical calculations carried out by V.M.Strutinsky 33,34. In these calculations the nuclear potential energy was regarded as the sum

 $V = V_{LDM} + \delta \qquad , \qquad (1)$ 

where V<sub>true</sub> is the part of the potential energy, described by the liquid drop model, and  $\delta$  is the shell correction. The first term of this sum varies smoothly as one goes from one nucleus to another, while the shell correction strongly depends upon the level density in the vicinity of the Fermi boundary and changes noticeably with elight variations in proton and neutron numbers. It follows from these calculations that the dependence of the potential energy on nuclear deformation ( in the region of positive deformations) has the shape of a curve with a couple of minima and a couple of maxime rether than the shape of a parabola (fig.2). As a result, the concept of a two-humped fission barrier of nuclei has appeared. The model suggested by V.N. Strutinsky provides at least a qualitative explanation of all main properties of fissioning isomers 35). One of the minima, the deeper one, is associated with the ground state, while the other, corresponding to larger deformation, is related to the isomeric state. For a number of nuclei the second minimum is deep enough to contain a system of quasistationary states, the lowest of which being isomeric. The forbiddenness for gamma-transitions is associated with a large difference in deformations and the

The is noteworthy that the occurrence of isomeric levels in muclei at changes in their equilibrium shapes was already discussed in the paper by Hill and Wheeler  $\frac{32}{2}$ , in which they considered the transition of the nucleus from the oblate shape to the prolate one.

presence of the inner barrier separating the isomeric and the ground states. The relatively small height and width of the outer barrier contribute to the enhancement of the spontaneous fission probability. The correlation between the isomer production and induced fission cross sections is explained by the fact that both processes pass through the common stage of the inner barrier, and thereafter the nucleus either penstrates the outer barrier and undergoes fission, or remains in the isomeric state after the emission of gamma-rays.

The two-humped fission barrier explains a number of other phenomena, which could not be understood in terms of the previous concept of the fission barrier. These phenomera include the modulation of fission resonances  $^{36}$ , the appearance of wide fission resonances in the subbarrier region in the reactions  $(n,f)^{37}$  and  $(d_{n}pf)^{38}$ . The cause of these phenomena is the existence of levels with large fission widths in the second potential well. In the case of a resonance between levels in the first and the second potential wells the fission barrier penetrability and, consequently, the fission width increases sharply.

In experiments with gamma-rays  $^{39}$  the channel effects occurring in photofission of plutonium isotopes in the vicinity of the fission barrier B<sub>2</sub> were studied. The angular anisotropy reached its maximum value at an energy I MeV lower than B<sub>2</sub>, while in the framework of A<sub>2</sub>Bohr's concepts  $^{40}$  this maximum should lie above B<sub>2</sub>. This fact can be explained using the model of V.M.Strutinsky  $^{35}$  under the assumption that in the fission process only the spectrum of cr unelo above the outher, lower barrier shows up. The inner, high barrier determines the fission barrier observed experimentally.

Thus, the idea of a two-humped barrier proved very fruitful in explaining many physical phenomena associated with fission. However, there have been long no direct experimental evidence for the existence of quasistationary states in the nucleus at abnormally large deformation.

Recently the development of a new experimental technique has been started for the studies of the properties of spontaneously fissioning isomers, namely for the study of gamma-ray

and conversion electron energy spectra closely related to isomeric state population  $^{41,42)}$ . The recent publication of German physicists  $^{43)}$  describes the observation of conversion electrons emitted at the transition of the  $^{240}$ Pu nucleus to the isomeric level in the second potential well. The conversion lines in the electron spectrum are interpreted as components of the E2-transitions witk'n the K=0<sup>+</sup> rotational band, These transitions are characterized by abnormally large momenta of inertia, which indicates a large deformation of the nucleus in isomeric state.

#### IV. GAMMA-RAY EXPERIMENTS.

The studies of spontaneously fissioning nuclei offer unique possibilities of gaining information on the uncommon properties of heavy nuclei at very large deformations.

Firstly, in this case a spectroscopic approach to this problem is possible, which can be realised experimentally. As is known, the existence of a two-humped fission barrier leads to the occurrence in nuclei of two independent level systems separated by the inner barrier (fig.2). The isomeric state formation is associated with rediative transitions of the nucleus in the second potential well, end its decay in some nuclei (U, Np) may apparently proceed through transitions of the nucleus into the first potential well <sup>44</sup>. Thus, determination of the characteristics of gama-radiation and conversion electrons emitted as a result of isomeric state production and decay may yield some data on both the levele in the second potential well and radiative transitions between nuclear levels with lergely different deformation.

Secondly, the studies of spontaneously fissioning isomers allow to obtain semiempirically some data on the parameters of the two-humped fission barrier, i.e., determine the fission barrier heights and the depth of the potential well between them. The combined use of the nuclear reaction method and the two-humped fission berrier model has resulted in successful understanding of the properties of spontaneously fissioning isomere. The studies have extended from heavy-ion-induced reactions to reactions with lighter particles.

The experiments showed that the cross section for the formation of an isomeric state in the nucleus increases as the mass and charge of bombarding particles decrease. This fact is explained by reduction of the number of the channels of competing reactions. In this connection the use of photonuclear reactions is of interest since the absence of the Goulomb barrier and the binding energy make it possible to produce compound nuclei with low excitation energy immediately after gamma-ray absorption. This circumstance minimizes the number of intermediate stages of the process leading to the population of the isomeric layel.

Below we shall describe our gamma-ray experiments, which have been aimed at the production of Pu and Am spontaneously fissioning isomers. The measured results, their treatment and an analysis of the data obtained are given in the present paper. The parameters of the two-humped fission barrier for the nuclei are also listed.

### IV.I. Reaction characteristics. Experimental technique.

In our experiments we used gamma-rays of moderate energies. The dipole absorption of gamma-rays mainly occurs at gamma-ray energies between IO and 20 MeV ( unless this is suppressed anyhow) and thus the nucleus receives angular momentum I<sup>-</sup>. The interaction between gamma-rays and nuclei is characterized by relatively small cross section magnitudes. Though the photoabsorption cross section displays the so-called giant resonance (at  $E_{g} \approx 13$  MeV for heavy nuclei)  $^{45}$ , the absolute value of the cross section is only a few hundred millibarn in the maximum. However, modern electron accelerators permit the production of sufficiently intense gamma-ray fluxes, and photonuclear reactions may proceed with noticeable yields.

The main decay modes of an excited nucleus in the region of heavy nuclei are neutron emission and induced fiesion. Other possible forms of de-excitation are proton emission and inelestic scattering of gamma-rays, but the latter reaction occurs in the usage of heavy nuclei with rather low probability. In the studies of spontaneously fissioning iscasers the type of reaction is easily identified on the basis of the half-life

of the nescent isomer.

We studied the formation of spontaneously fiscioning isomers in the reactions  $(\gamma, n)$  and  $(\gamma', p')$ . The electron bremsstrahlung served as the source of gamma-rays. All the measurements were carried out in the external electron beam from the I7-orbit microtron of the Institute for Physical Problems of the USSR Academy of Sciences <sup>46</sup>.

Fig. 3 shows the experimental set-up. The electron beam was transported from the accelerator chamber through an electron guide about I m in length and, having passed through a thin aluminium window (0.2 mm), bombarded a Imm tungsten target. This thickness of the target (0.3 of the radiation length) permits the maximum yield of forward gamma-radiation. To provide full absorption of electrons, an aluminium screen 15 mm thick is placed behind the tungsten target. In some cases, to avoid neutron production in the stopping target proper, the latter was made of aluminium 3om thick, in which the neutron binding energy is rather high, IS MeV. The beam focussing on the target was performed by doublet quadrupole lenses. the effective size of the beam at the target being about 5 mm. The beam was monitored by the current on the target with an accuracy of 25. The average electron current in our experiments varied from a few microemperes to several tens of microssperes depending on electron energy and the type of experiments under way.

The electron energy in the micretron can be varied over a wide range by chan\_ing the strength of the main magnetic field or the orbit number. Irrespective of the electron energy value, the energy spread of beam electrons was about 50 keV.

The microtron was operated under pulsed operating conditions. The current pulses generally used were  $1-2\mu \sec$ long at a frequency rate of 400-600 Hs, consequently, the interval between pulses was equal to 1.5-2.5essec. During the pulse of electrons isomeric nuclei wers accumulated in the irradiated target, while between the pulses their decay was detected.

In all our experiments a multi-filament spark counter<sup>47</sup>) served as a fission fragment detector (fig.4). The main components of the counter are anode having the shape of a grid of tungsten filaments 0.Imm in diameter tightened on a metal ring, and a flat copper cathods with a well polished and chroms-plated surface. The distance between the electrodes was 2.0mm, and the grid spacing of the anode was 4mm. The target was placed at a distance of 3-4mm from the anode. All the components were placed in a vacuum chamber evacuated to a pressure of  $10^{-2}$ mmHg before filling with a working gas. A mixture of N<sub>2</sub> (1.5%) and He(98.5%) was used as a filler at a pressure of I atm.

The spark counter filled with this mixture provided practically complete discrimination of alpha particles against fission fragments even in the case of Am targets, which are very alpha active. The detection efficiency for fission fragments was about 30%. The rise-time of fission fragment pulses was 50 nsec. The counter time resolution was about 100 nsec.

One of the spark counter disadvantages is its long dead time that is determined by the voltage rise-time at the counter electrodes after the break-down of the interelectrode gap and is equal to 0.5 msec in this case. In addition, when opereted on a gamma-ray beam, the spark counter schibited another peculiarity, which should be taken into account in performing the experiments. This peculiarity consisted in the fact that the intense game radiation led to a break-down during the electron pulse, which was identical to the break-down due to fragment penstration. Because of the long dead time, the detector sensitivity was restored only in 0.5 msec. However, this circumstance did not prevent us from carrying out measurements for the 240 Am and 242 Am fissioning isomers since these have relatively long half-lives, 0.9 meet and 14 meet, respectively. However, in order to eliminate break-downs due to gamma-ray pulses (and prompt fission fragments), in the studies of isomore with lifetimes of a few microseconds (239 Pu and 241 Pu) we operated the counter under pulsed operating conditions.

The pulsed operating regime of the counter was achieved by means of a rather simple electronic circuit shown in fig.4.

During the current pulse the voltage at the counter electrodes sharply decreased, but after the passage of the gamma-ray pulse the voltage was completely restored.and one could detect the fragments of delayed fission.

The spontaneous fission fragments from target nuclei and elow neutrons resulting from the moderation of fast neutrons formed in photonuclear reactions were the source of the background during the experiment.

The slow neutrons induced fission of target nuclei between the electron pulses, thus being capable of immitating isomer decay. In order to provide shielding against slow neutrons, the counter was covered by a cadmium layer 0.8mm thick and a 5mm boron layer,

The background of spontaneous fission and slow-neutron induced fission limited the minimum gamma-ray energy, at which the yield of fissioning isomers was still noticeable.

The signals from the spark counter were fed into a 512channel analyzer with a channel width of  $0.1-4.0\,\mu$ sec, operated in the time regime. The start-up of the analyzer was synchronous with the microtron current pulse, which was achieved by means of a special generator.

While the energy dependence of the yield of prompt fission frequents was measured, the spark counter was removed from the stopping target by 50-70 cm. This was done to eliminate break-downs from gamma-ray pulses. The current decreased to I $\mu$ A. Under these conditions one could determine the ratio of the yields of delayed and prompt fission fragments. To control the counter efficiency, its counts were regularly calibrated at a fired electron energy (generally at I2 or I3 MeV). The tackground level was also checked systematically.

#### IV. MEASURED RESULTS AND THEIR ANALYSIS

The objective of the experiments was to produce Pu and An fissioning isomers in the  $(\gamma, n)$  and  $(\gamma, \beta')$  reactions and determine the dependence of the isomer yield upon the genmaray energy. The bremsstrahlung limiting energy  $E_{\beta}$  was varied over the range of 7-16 MeV.

Table I gives the target characteristics and the halflives of the mascent isomers.

Target	Enrichment, %	Target thickness mg/cm <sup>2</sup>	Reaction type	Neutron binding energy 52	Leomer )	Isomer half- life (µsec)
239 <sub>Pu</sub>	95	0.4	(8,8)	5,59	239 <sub>Pu</sub>	8.0
240 <sub>Pu</sub>	80	0.7	(J,n)	6.45	<sup>239</sup> Pu	- " -
<sup>242</sup> Pu	99	0.9	(ď,n)	6.26	241 <sub>Pu</sub>	23.0
241 <sub>Am</sub>	98	0.4	( <sub>4</sub> ',n)	6.84	240 <sub>Am</sub>	900
243 <sub>Am</sub>	98	0.4	( a, b)	6.23	242 <sub>Am</sub>	14000
243 <sub>Am</sub>	98	0.4	(8,8)	- " -	243 <sub>Am</sub>	6.5

TABLE I

The isomers were identified by their known half-lives (figs. 5a and 6a). In the  $^{243}Am(f, n)^{242mf}Am$  reaction no decrease in fragment activity was observed since  $T_{\rm I/2}$  (I4 msec) for the  $^{242}Am$  isomer was substantially larger than the time interval between microtron pulses (2.5 msec). The identification of the  $^{242}Am$  fissioning isomer was based on the following facts. All the reactions proceeded under the same conditions,  $T_{\rm I/2} \gg 2.5$  msec was observed and the yield curve for the 242mf Am fragments as a function of gamma ray energy had a shape analogous to that for other isomers.

a) Reaction integral yields. The direct result of the experiments was knowledge of the number of fission events N<sub>f</sub> (delayed or prompt fission) detected at a given electron energy  $E_0$ , related to the unit electron current J and to the measurement time t, i.e., the integral yields of the reactions were measured to be  $\gamma(E_0) = \frac{M_f - M}{Tt}$ ,

where N is the number of the background events.

Fig. 5b shows the measured isomeric yields  $X_i$  in the ( $\gamma$ ,n) reaction, while fig. 6b gives the dependence of the

 $^{239mf}$ Pu isomer yield on the bremsstrahlung limiting energy in the ( $\chi, \chi'$ ) reaction. Analogous results have been obtained for  $^{243mf}$ Am. The statistical errors involved in the measurements are also shown in these figures.

In the experiments the corresponding yields  $Y_{f}$  of prompt fission fragments have also been measured.

Although photonuclear reactions are in essence the simplest ones, the derivation of complete quantitative information from studying them encounters considerable difficulties. The matter is that retardation of fast electrons in a substance is accompanied by the production of photons with energies ranging from 0 to a maximum energy  $\mathbf{E}_0$  equal to the kinetic energy of incident electrons. Thus, the yield  $Y(\mathbf{E}_0)$  of any reaction, to be measured experimentally, is determined by the integral relation

integral relation  $Y(E_o) = \pi \int G(E_f) \phi(E_f, E_o) dE_f \qquad (2)$ 

where  $\mathcal{T}(\mathcal{E}_{i})$  is the reaction cross section;  $\mathcal{P}(\mathcal{E}_{i};\mathcal{E}_{i})$  is the spectrum of electrons with the kinotic energy  $\mathbf{E}_{0}$ ; A is the coefficient associated with the experimental geometry and the quantity of the target substance.

On the basis of the measurements we know the  $Y_i$  values at some n-points with a certain error (figs. 5 and 6c). These  $Y_i$  values are to be used to re-establish the  $\mathcal{G}$  ( $Y_i$ ) function, i.e., determine the excitation function of the reaction.

b) Reaction cross sections. In mathematics the equation of type (2) is called the Volterra integral equation, which is a particular case of the Fredholm equation. Momerous papers are devoted to the methods of solving this kind of problems, mainly by using computers. In particular, a large number of these papers deal with photonuclear reactions  $^{45,53}$ .

Although the solution of eq.(2) can be reduced to the solution of the system of elgebraic equations of the m-th order ( n is the number of Y measurements), this straightforward mathod is frequently unsuitable because it neglects the statistical character of the results obtained, and often leads to entirely incorrect solutions. Generally speaking, the Fredholm equation serves as a typical example of an incorrectly

posed problem. The essence of this incorrectness consists in thet relatively small changes in ' s initial data lead to substantial changes in the results of the problem solution.

For spontaneously fissioning isomers, such calculations require high accuracy in the measurements of the yields  $Y_i$ . As a rule, the preset accuracy of the determination of the

 $\mathcal{G}(\mathbf{E}_{\mathbf{X}})$  cross sections requires that an accuracy in measuring the Y; (E,) yield curve be by an order of magnitude higher. As the number of the detected delayed fission events is rather small due to the very small reaction yield Y. ( at 12-15 MeV the statistical error is usually 3-5%), there is no firm confidence in the correctness of the cross section re-establishment by this method. Nevertheless, we have carried out c. lculations for the cross sections using two different methods <sup>54</sup>). Both methods are based on the statistics] approsch but the solutions of integral equation (2) using a computer were different. The scheme of the calculation was as follown First, the G/ cross section was calculated from the crect solution of a algebraic equations. The resulting values were inserted in the initial integral equation and the calculated Y; values were compared with the experimental ones. Unless the results of the comparison satisfied the chosen statistical criterion ( it is generally analogous to the well known statistics) parameter,  $\chi^2$ ), the cross section was "smoothed" over the first or second derivative. The procedure was repeated until the necessary agreement was obtained.

This method of solution is called the "regularization" method, 55).

The calculated results are listed in figs. So and 6c. Fig. So shows that for the  $(\gamma, n)$  reaction all the cross sections display the characteristic feature, namely the prenence of the  $G_i$  maximum approximately at an energy E, which is 2-2.5 MeV higher than the reaction threshold.

The accurrence of this maximum is quite clear. After neutre emission, the nucleus still remains in an excited state end, due to gamme rediction, descends to the lower isometric state in the second potential well. However, when the energy of this excited state is close to the height of

the outer barrier, the fission width increases sharply and the production cross section for isomeric states becomes lower. Thus, in the excitation function maximum, the effective height of the outer barrier, i.e., the excitation energy at which the radiative width is close to the fission one, shows up.

Table 2 gives some data characterizing the measured excitation functions ( $\chi$ , n), i.e., the reaction thresholds ( $E_{\rm th}$ ), the positions of the excitation energy maxime ( $E_{\rm max}$ ) and the values of the cross section ( $G_{\rm i}$ ) in the maximum. The reaction thresholds were determined by extrapolating the excitation function to the zero value of the cross section ( these extrapolation methods will be dealt with below). The delayed-to-prompt fission ratios are also given in the table. For the calculation of these ratios the energy dependences of the fission cross section, shown in fig.7, have been used. For the  $^{242}$ Pu and  $^{243}$ Am isotopes, these dependences were obtained by using the experimental apparatus described above and a technique based on an analysis of the integral yields. For the  $^{239}$ Pu and  $^{241}$ Am isotopes, the data from raf.  $^{53}$ ) were used.

It may be seen from table 2 that the ratios are of the order of  $10^{-3}$ , i.e., by a factor of  $10^2$  higher than the analogous ratios measured for  $^{238}$ U and  $^{239}$ Pu at the gamma-ray energy of 55 MeV <sup>56</sup>). This difference between the ratios

We is apparently explained by the fact that at this high limiting energy of gamma-rays only a small fraction of bremsstrahlung contributes to the formation of fissioning iscmers.

Using the experimental ratios  $\frac{5}{5}$ , and the known values of  $\frac{5}{7}$ , on can obtain the ratios of production cross sections for a nucleus in an isomeric and in the ground state. It is evident from the table that these ratios are practically the same as those in reactions with charged particles 9.

Reaction	E <sub>th</sub> , Mev	E <sub>max</sub> , Mev	G <sub>2</sub> ba <b>rn</b>	रा0-3 इ	<u>G</u> 5, x10 <sup>-3</sup>
240 <sub>Pu</sub> (y,n) <sup>239mf</sup> Pu	10.0+0.3	11.5 <u>+</u> 0.2	170 <u>+</u> 60	1.5±0.5	0.7+0.3
<sup>242</sup> Pu(y,n) <sup>24Imf</sup> Pu	8.95 <u>+</u> 0.3	10.5 <u>+</u> 0.2	200 <u>+</u> 60	2.5±0.5	0.9 <u>+</u> 0.3
241 <sub>AB(y,n)</sub> 240mf <sub>Am</sub>	9.7 <u>+</u> 0.3	11.2 <u>+</u> 0.2	150 <u>+</u> 50	1.0 <u>+</u> 0,3	0.5+0.2
243 Am (J;n) 242mf Am	8.7 <u>+</u> 0.3	10.6 <u>+</u> 0.2	130 <u>+</u> 50	1.3 <u>+</u> 0.4	0.4+0.15

TABLE 2

#### V. FISSION BARRIER STRUCTURE

Our measurements of the excitation functions of reactions leading to fissioning isomers permit the determination of a number of parameters of the two-humped fission barrier, using the Strutinsky model.

V.I. Mechanism of fission isomer production and of nuclear fission in terms of the two-humped fission barrier model.

In the two-humped fission barrier model, the processes of fission and fission isomer production may be regarded as including a number of successive stages. Once a particle or gamma-ray is captured, a compound nucleus characterized by a strong interaction between one-particle and collective degrees of freedom is formed. In case an adequate amount of excitation energy is imparted to the collective degree of freedom, the nucleus may become strongly deformed, At deformation corresponding to the second potential well, a large portion of this energy may again be converted into thermal energy. This energy re-distribution is explained by that the deformation energy becomes small due to the existence of the second potential well, and the remaining energy, as a result of the strong interaction, is imparted to one-particle degrees of freedom. The nucleus in this state can be regarded as a compound nucleus at large deformation. Such a nucleus can

undergo fission and emit either a gamma-ray or a neutron. In the latter case its excitation energy may decrease to such an extent that it will be unable either to return to the first potential well or undergo fission, and remain in the second potential well. After the gamma-ray cascade, the nucleus will descend to the lower (isomeric) state.

Similarly to the common nuclear states, the cross sections for all of these processes (fission, neutron and gamma-ray emission) for highly deformed nuclei can be expressed via the reduced widths.

With this reaction mechanism, the fission and production cross sections for the isomer are determined by the following equations 58,59)

$$G_{f} = G_{c} \frac{\overline{f_{f}}}{\overline{f_{i}} + \overline{f_{ef}}} \frac{\overline{f_{e}}}{\overline{f_{i}^{2} + \overline{f_{ef}}} + \overline{f_{e2}}}, \qquad (3)$$

$$G_{i}^{c} = G_{c} \frac{\overline{f_{i}^{c}}}{\overline{f_{i}^{a} + f_{df}}} \cdot \frac{f_{d2}}{\overline{f_{k}^{a} + f_{k}^{a} + f_{k}^{a} + f_{d2}^{a}}}$$
(4)

where  $f_{2}$  and  $\bar{f}_{2}$  are the reduced widths for the transition through the inner barrier from the first potential well to the second one and reversely, respectively;

 $\overline{f_2}$  is the reduced width for the transition through the outer barrier ( the fission width for states in the second potential well);  $\overline{f_{ef}}$  and  $\overline{f_{e2}}$  are the reduced widths i r the emission of a particle from states in the first and second potential wells. In heavy nuclei, only neutron and gamma-ray emission practically occur, therefore,  $\overline{f_{ef}} = \overline{f_n} + \overline{f_r}$ , where  $\overline{f_n}$  and  $\overline{f_{ef}}$  are the neutron and radiative widths, respectively.

Expressions for these widths are of the following forms

$$\vec{f}_{1} = \frac{N_{h}}{2\pi \rho_{f}(E^{*})} , \qquad (r)$$

$$\overline{f_2} = \frac{N_A}{2\pi \rho_F(E^* - E_i)} , \qquad (6)$$

$$\vec{f_2} = \frac{N_B}{2\pi P_B(E^* - E_l)}, \qquad (7)$$

$$\int_{DI} = \frac{2mR^2}{\pi \hbar^2} \cdot \frac{1}{P_I(E^*)} \int_{\mathcal{F}}^{E^* B_n} \mathcal{E} P_I(E^* - B_n - E) dE , \qquad (B)$$

$$\int_{n_2} = \frac{2mR^2}{\pi\pi^2} \cdot \frac{1}{\beta_{ij}(E^* - E_i)} \int_{0}^{E^* - B_n - E_i} \int_{0}^{E^* - B_n - E_i - E} dE, \quad (9)$$

$$\int_{S_1} \sim \frac{1}{P_1(E^*)} \int_{S_1} P_1(E^*-E)E^2 dE$$
, (10)

$$\int_{\mathcal{H}^2} \sim \frac{1}{\rho_{\tilde{f}}(E^*-E)} \int_{\mathcal{P}_{\tilde{f}}} (E^*-E_i-E) E^3 dE, \qquad (II)$$

where  $\mathcal{N}_A$  and  $\mathcal{N}_S$  are the numbers of open channels in the inner and outer barriers, respectively;  $\mathcal{F}_I(\mathcal{E}^*)$  and  $\mathcal{F}_S(\mathcal{E}^*-\mathcal{E}_i)$ are the level densities at the excitation energy  $\mathbb{E}^*$  in the first and second potential wells (in the second potential well the excitation energy is lower by the magnitude of the energy in the second minimum  $\mathbb{E}_i$  than in the first potential well);  $\mathcal{B}_A$  is the neutron binding energy,  $\mathcal{E}$  is the kinetic energy of the emitted neutron, or the gamma-ray energy;  $\mathcal{M}_i$ ,  $\mathcal{R}_i$  are the compound nucleus mass and radius, respectively.

# V.2. Determination of the parameters of the two-humped fission barrier.

If fission is considered to be a one-dimensional picture, the two-humped fission barrier can be described by six parameters. These parameters characterize the height and curvature of the inner ( $\mathcal{E}_A$  and  $\hbar\omega_A$ ) and the outer ( $\mathcal{E}_B$  and  $\hbar\omega_A$ ) barriers, as well as the energy and curvature of the second minimum ( $\mathcal{E}_i$ ,  $\hbar\omega_i$ ). It is even from eqs.(5-7) that the reduced widths for the transition of a nucleus through the inner end outer barriers are determined by the parameters of these barriers. Since the cross sections for the fission process and fission isomer production are expressed by the reduced widths, the measurement of these cross sections allows to extract some information about the parameters of the two-humped fission barrier. However, a large number of parameters leads to the measured cross section to satisfy a few different sets of these parameters. Therefore, it is desirable to use such reactions or those aspects of them, which are meinly dependent on one parameter only and practically independent of the others.

In this respect photonuclear reactions leading to spontaneously fissioning isomore are of certain interest since they permit determination of the majority of the twohumped fission barrier parameters. Fig. 8 shows the excitation energy dependences of the cross sections for the  $(\mathcal{J}, \mathcal{J}')$  and  $(\mathcal{J}, n)$  reactions, calculated by formulae (3,4), as well as the isomer-to-induced fission ratios in the  $(\mathcal{J}, \mathcal{J}')$  reaction. The characteristic discontinuities in the curves describing these dependences permit unablguous determination of the heights of the outer and inner terriers for the final nucleus.

One can judge of the outer barrier height with respect to the bottom of the second potential well for the initial nucleus by the experimental values of the isomer-toinduced fission ratios. As is seen from eqs.(3,4), these ratios are of the following forms

 $\begin{aligned} G'_{i}G_{f} &= \int_{\delta^{2}}^{\delta^{2}} f_{2}^{*} & \text{for the } (\beta, \beta') \text{ reaction,} \quad (12) \\ G'_{i}G_{f} &= \int_{\delta^{2}}^{\delta^{2}} f_{2}^{*} & \text{for the } (\beta, n) \text{ reaction.} \quad (13) \end{aligned}$ 

The values of  $\vec{l_2}$  or  $\vec{l_1}$  obtained from these expressions permit determination of the number of open channels above the outer barrier at a given excitation energy and hence of the outer barrier height. However, the latter determined in this way will be less definite than the height value obtained from the excitation function shape (figs. 5,6,8). Indeed, in order to determine the fission barrier height, one requires knowledge of the energy interval between the barrier height and the excitation energy. For the determination of this energy interval from the number of open fission channels it is necessary to employ the model concepts of the excitation energy dependence of the level density. This dependence was chosen to be of the form:

 $f_{2}(E_{*}^{*}|) = \frac{f}{T} e^{\frac{f^{2} - f_{2}}{T}} \frac{2\tilde{I}^{*}}{2\sigma^{*}} e^{-\frac{\tilde{I}^{*} + f_{2}}{2\sigma^{*}}} \text{ at } E^{*} < B_{x} , \qquad (14)$ 

$$P_{2}[F_{*}^{*}] = \frac{\sqrt{F}}{R2A^{4}} \frac{e^{2\sqrt{A}t}}{2\sqrt{F}} \frac{2^{\frac{1}{2}}+t}{2\sqrt{F}} e^{-\frac{(t^{-\frac{5}{2}})^{2}}{2G^{2}}} \text{ ot } B^{*} > B_{*} , \qquad (15)$$

where the parameters  $\mathcal{A}$  and  $\mathcal{G}'$  are chosen from the comparison with experimental data (mainly from the density of neutron resonances). The parameters T and E. are chosen from the condition that the dependence  $\rho_{\ell}(\mathcal{E}^*, \overline{I})$  smoothly transform into the dependence  $\rho_{\ell}(\mathcal{E}^*, \overline{I})$  at  $\mathbf{E} = \mathbf{E}_{\mathbf{X}}$ :

 $E_{\chi} = 3,13 + P(Z) + P(N)$  MeV ,  $U = E^{*} - P(Z) - P(N)$ , where P(Z) and P(N) are the decoupling energy for the pairs of protons and neutrons. Fig. 9 shows the excitation energy dependences of the level density, calculated by eqs.(14) and (15).

Using the model concepts considered above and the measured excitation functions, we have obtained the numerical values of the two-humped fission barrier parameters for Pu and Am isotopes.

It is seen from comparing figs. 6c and 8a that a large part of the experimentally measured excitation function for the  $^{239}\text{Pu}(\ell_{,,\ell'})^{239\text{mf}}\text{Pu}$  reaction lies above the inner barrier. There are only indications of a decrease in the cross section at an excitation energy of 6 MeV. Hence one can make merely a rough estimation of the inner barrier height to be 6-7 MeV. The same estimation can be made for the height of the inner barrier for the  $^{243}\text{Am}$  isomer.

Some data on the outer barrier for the <sup>239</sup>Pu nucleus can be obtained from the measured ratio  $\frac{5}{20}r_{\rm p}$ , which is equal to 10<sup>-4</sup> at an excitation energy of 7 MeV. It is experimentally known that the radiative width slightly changes with excitation energy and equals about 0.03 eV at the given energy. Thus, from eq.(12) one can derive the reduced fission width for the states in the second potential well,  $\frac{7}{2} \sim 100$  eV. For the <sup>239</sup>Pu nucleus <sup>62)</sup>, it is possible to obtain the

For the <sup>23</sup> Pu nucleus <sup>62</sup>, it is possible to obtain the value of the level density in the second potential well at an excitation energy of 7 MeV ( $\rho_f \sim 10^3$  I/MeV) by extrapolating the experimentally observed modulation of subberrier fission resonances. The known values of  $\bar{\ell}_c$  and  $\rho_f$  enable one to

determine from eq.(7) the number of open channels N<sub>d</sub> under the outer barrier. The calculations yielded the value ~ IO. This corresponds to the excitation energy of about I.5 MeV with respect to the peak of the outer barrier and leads to the value  $E_{\rm d} \approx 5.^{-1}$  MeV with an error of 0.3 MeV.

More complete and more definite data on the parameters of the two-humped fission barrier can be obtained from the measured excitation functions of the ( X.n ) reactions, The excitation functions presented in fig. 5c make it possible to determine the heights of the outer barriers of the mentioned nuclei ( according to the position of the excitation function maximum). However, it should be noted that in this case some effective height of the barrier, at which the fission and radia. tive widths are equal, rather than the real one. is determined. The real barrier height exceeds the effective one by 0.7 MeV for odd-odd nuclei (240Am, 242Am) and by 0.8 MeV for even-odd nuclei (239 pu. 241 pu). In calculating this difference, use was made of the level density obtained from the observed subbarrier resonances and the barrier curvature parameter equal to 0.4 MeV for odd-odd nuclei and to 0.5 MeV for even-odd nuclei 63). The heights of the outer barrier, obtained in this manner with an error of 0.2 MeV are shown in table 3.

Unlike the outer barrier height, the energy of the second minimum ( or the energy of the isomeric state) cannot be determined from the excitation function shape so unambiguously. Indeed, due to the low isomer cross section, the last excitation function experimental points are-1 MeV higher than the supposed threshold. "herefore, to determine the threshold ( with an accuracy of 0.I-0.2 MeV), extrapolation of the experimental excitation function to the region of lower energies is necessary. Since the behaviour of the excitation function in the vicinity of the threshold is unknown, in the extrapolation the calculated excitation function passing through the experimental points is used. It is natural that the threshold walue depends in this case on the choice of a model that defines the excitation energy dependence of the cross section.

In the statistical model of a nucleus <sup>64)</sup> the excitation function for the reaction with the evaporation of one

neutron is determined by the following relation that well describes the experimental data

$$\mathbf{G}_{s}^{\prime} = \mathbf{G}_{c} \left( \mathbf{1} - e^{-\frac{\mathbf{A}^{E}}{T}} \right), \qquad (16)$$

where  $G_c$  is the compound nucleus cross section;

 $\Delta \leq$  is the excitation energy counted off from the reaction threshold;  $\mathcal{T}$  is the nuclear temperature. It can however be expected that, owing to the complex mechanism of reactions leading to spontaneously fissioning isomers (initially large nuclear deformation followed by the emission of a neutron), the excitation function will not be described by such a simple expression. A number of formulae have been suggested to describe the excitation energy dependence of the isomer production cross section. R.Vendenbosch <sup>65)</sup> has derived the following equation

$$G_{i} = G_{c} \left[ 1 - \left( 1 + \frac{A_{c}}{F} \right) e^{-\frac{A_{c}}{F}} \right], \qquad (17)$$

which i. based on the assumption that the evaporation of a neutron from the compound nucleus leads to the residual nucleus to appear either in the first or in the second potential well, between whose levels transitions are forbidden.

With the described multi-stage mechanism of isomeric state population and fission, the isomer production cross section can be written, using eqs.(3) and (4), in the form as follows

$$G_i = G_f \cdot \frac{I_{R_i}}{I_e} \cdot$$
(18)

The energy dependence of the fission cross section is well known experimentally ( in the energy range measured, the fission cross section varies very little). Therefore, the behaviour of the excitation function of the reaction leading to the formation of spontaneously fissioning isomers is determined by the ratio  $\sqrt{n_{f_{e_{e}}}}$  as a function of energy. The energy dependence of the reduced neutron and fission widths has been calculated using eqs.(7) and (9) and the excitation energy dependence of the lawel density, shown in fig. 9. In calculating  $\sqrt{n_{2}}$  it was taken into account that the density parameter of the lawels q at the barrier is by 20% larger

then that in the potential well  $(a = I_{,2} a_{,2})^{6I}$ . The height of the outer barrier relative to the bottom of the second potential well was chosen to be equal to 3 MeV ( the calculations showed that the outer barrier height strongly affects the absolute value of  $\frac{f_{1}}{f_{5}}$ , but this influence on its energy dependence is weaker). Fige Io shows the excitation functions for the reaction producing spontaneously fissioning isomers. calculated using formulae (16-18). It is seen from this figure that the excitation functions calculated using different forsulse differ to a noticeable extent. Consequently, their use in the treatment of experimental data leads, as has already Even noted above, to different values of the reaction threshold. This difference grows with using different values of the T and Q parameters that are components of eqs. (14) and (15). For instance, with changes in the parameter T within the limits of 0.5-I.0 MeV or in the parameter a within the limits of 20-35 I/MeV ( the experimental values of these parameters lis in these particular ranges), the reaction threshold values may differ by 0.8-0.4 MeV. This dependence on the parameters that are unknown for highly deformed states leads to an error of 0.3 MeV in the determination of the isomeric state energy.

Table 3 shows the isomeric state energies obtained using the excitation function calculated by formula (18). In determining the threshold the experimental and calculated energy dependences of the reaction cross sections and the yields were compared. In the case of yields, the calculated cross sections were integrated over the spectrum of electron bramestrahlung<sup>66</sup>) using eq.(2).

The isomer-to-prompt fission ratio  $\left(\frac{S_{1}}{C_{2}}-\frac{S_{2}}{C_{2}}\right)$  in the excitation function maximum permits determination of the height of the outer barrier for the initial nucleus with respect to the bottom of the second potential well ( $\mathbf{E}_{p}-\mathbf{E}_{i}$ ). Fig. II shows the calculated dependence of  $C_{1}/c_{p}$  on the outer barrier energy. The dependence of the level density on excitation energy, shown in fig. 9, was used to calculate the  $C_{1}$  and  $C_{2}$  values. From the comparison of the measured values

of  $\frac{6}{16}$ , with the calculated ratios  $\frac{6}{16}$ , shown in fig. II, one can obtain the values of ( $E_g - E_\ell$ ), which are also presented in table 3. The error in determining the difference ( $E_g - E_\ell$ ) is 0.3 MeV, due to an uncertainty in the parameters that are components of the expressions for the level density.

Nucleus	The (g,n) reactions			The $(\gamma, \gamma')$ reactions		
	E <sub>i</sub> , NoV	E <sub>B</sub> ⊷E <sub>i</sub> , MeV	≊ <sub>B</sub> ; Me¥	E <sub>B</sub> , NeV	Е <sub>д</sub> , МеV	
239 <sub>Pu</sub>	3.5 <u>+</u> 0.3	2.3±0.3	5.8 <u>+</u> 0.2	5.5 <u>+</u> 0.3	6.5+0.5	
240 <sub>Pu</sub>		2.6 <u>+</u> 0.3	l			
24I <sub>Pu</sub>	2.7±J.3	2 <b>.4</b> ±0.3	5.1 <u>+</u> 0.2			
<sup>242</sup> Pu		2 <b>.6<u>+</u>0.</b> 3				
240 <sub>AR</sub>	3.0 <u>+</u> 0.3	2 <b>.</b> 3 <u>+</u> 0.3	5.2±0.2			
241 <sub>Am</sub>		2 <b>.5<u>+</u>0.3</b>				
242	2.4 <u>+</u> 0.3	2 <b>.7<u>+</u>0.3</b>	5.1 <u>+</u> 0.2			
243 <sub>AR</sub>		2.2 <u>+</u> 0.3		5,5 <u>+</u> 0,3	6.5 <u>+</u> 0.5	

TABLE 3

# V.3. Comparison with other data and with theoretical calculations.

It is of interest to compare the parameters of the two-humped fission barrier listed in table 3 with the analogous parameters derived otherwise, and also with the results of theoretical calculations.

In ref.<sup>67)</sup> the  $E_s$  and  $E_i$  values for Pu, Cm and Bk isotopes are obtained from an analysis of the excitation functions for reactions involving the evaporation of two or three neutrons, leading to the formation of spontaneously fissioning nuclei. In this analysis the mentioned concept of the mechanism of reactions leading to spontaneously fissioning isomers was used. However, in contrast to our excitation function calculations based on the statistical properties of nuclei, the authors of ref.<sup>67)</sup> employed the specific spectra of one-particle states at different deformations.

Information on the two-humped fission barrier paramaters can also be obtained from the studies of the induced fission of nuclei. As has been noted above, the modulation of subbarrier fission resonances, observed in a number of cases, is explained by the existence of quasistationary states in the second potential well. From the known average spacings between the levels in both potential wells, the fission cross section on excitation energy, it is possible to determine the  $E_A$ ,  $E_A$  and  $E_1$  parameters. The data on these parameters, obtained on this basis, are listed in ref.<sup>63)</sup>.

The values of the  $\mathbf{I}_{s}$  and  $\mathbf{I}_{s}$  parameters were elso determined in a study of the (d, n?) reaction <sup>36)</sup>( the fission probability was measured as a function of nuclear excitation energy) and of the ( $\gamma_{s}$ f) reaction <sup>66)</sup> ( the fission cross section and the angular distribution of fission fragments were measured as a function of the gamma-rey energy).

Figs. I2 and I3 show the dependence of the  $\mathbf{E}_A$ ,  $\mathbf{E}_B$ and  $\mathbf{E}_i$  values for Pu and Am isotopes on mass number. It is seen that good agreement exists between the values obtained in different ways.

A large difference (~0.8 MeV) in the  $E_i$  values for the <sup>239</sup>Pu isotope, revealed in the measurements of the thresholds of reactions with gamma-rays and charged particles, attracts attention. This difference reaching beyond the limits of errors means to be due to the different initial states of the <sup>240</sup>Pu compound nucleus and to the level structure in the second potential well of the <sup>239</sup>Pu nucleus. In fact, the spin of the <sup>240</sup>Pu compound nucleus at photon capture is equal to I<sup>\*</sup>, therefore, at the evaporation of a neutron low-spin levels will be populated unlike the case of alpha particles, where the initial state of <sup>240</sup>Pu is characterised by a wide angular momentum spectrum. If one assumes now that the isceneric state of <sup>239mf</sup>Pu has a sufficiently high spin (> 5/2  $\hbar$ ) and

the low-spin levels are rather high-lying (above 0.5 MeV), an extrapolation of the measured excitation function for the ( $\zeta_1$ ,n) reaction will lead to the lowest state with low spin ( $\langle 5/2 \ h \rangle$ ) rather than to an isomeric state. Figs. I2 and I3 also show the  $E_A$ ,  $E_B$  and  $E_B$  parameters calculated by the shell correction method 57-69). Good agreement with the experimental data is clearly seen at  $A \leq 242$ . It is however noteworthy that the calculated barrier parameters increase with mass number (with moving toward the subshell containing I52 neutrons), while the experimental values do not exhibit this dependence.

These investigations of Pu and Am fissioning isomers produced in photonuclear reactions indicate that gamma-rays are an effective means of studying the properties of heavy nuclei at anomalously high deformations. Further development of these investigations will allow to obtain information about the fission barrier shape for a wide range of nuclei, about the transitions between the levels of different potential wells, and about the probabilities of different decay modes for quesistationary states in the second potential well.

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#### REFERENCES

- S.M.Polikanov, V.A.Druin, V.A.Karnaukhov, V.L.Mikheev, A.A.Pleve, N.K.Skobelev, V.G.Subbotin, G.M.Ter-Akopyan, V.A.Fomichev, Zh. Eksp. Teor. Fiz. <u>42</u>, 1464(1962)
- G.N.Flerov, S.M.Polikanov, K.A.Gavrilov, V.L.Mikheev, V.P.Ferelygin, Zh. Eksp.Teor.Fiz. <u>45</u>, 1396(1963)
- 3) A.F.Linev, B.N.Markov, A.A.Pleve, S.M.Polikanov, Nucl.Phys. <u>63</u>, 173(1965)
- Yu.V.Lobanov, V.I.Kuznetsov, V.P.Perelygin, S.M.Polikanov, Yu.Ts.Ogenessian, Yad.Fiz. <u>1</u>, 67(1965)
- 5) A.Ghiorso, private communication, Berkeley (1965)
- S.Ejørnholm, J.Borggreen, L.Westgaard, V.A.Karnaukhov, Nucl.Phys. <u>A95</u>, 513(1967)
- 7) S.Jägare, Phys. Lett. 32B, 571(970)
- 8) G.N.Flerov, A.A.Pleve, S.M.Polikanov, E.Ivanov, M.Martalogu, D.Foenaru, N.Vilcov, Physics and Chemistry of Fission, <u>1</u>, 307, IAEA, Vienna(1965)
- 9) G.N.Flerov, Yu.P.Gangreky, B.N.Markov, A.A.Pleve, S.M.Polikanov, H.Yungclaussen, Yad.Fiz. <u>6</u>, 17(1987)
- IO) G.N.Flerov, A.A.Pleve, S.M.Polikanov, S.P.Tretyakova, I.Boca, M.Sezon, I.Vilcov, N.Vilcov, Nucl. Phys. <u>AIO2</u>, 443(1967)
- II) I.Boca, N.Martalogu, M.Sezon, I.Vilcov, N.Vilcov, G.N.Flerov, A.A.Pleve, S.M.Pclikanov, S.P.Tretyakova, Nucl.Phys. <u>A134</u>, 541(1969)
- I2) Yu.P.Gangreky, B.N.Markov, S.M.Folikanov, H.Yungclaussen, Zh. Eksp.Teor.Fiz. (Letters), <u>4</u>, 429(1966)
- I3) N.L.Lark, G.Sletten, J.Pedersen, S.Bjørnholm, Nucl.Phys. <u>A139</u>, 481(1969)
- 14) S.M. Polikanov, G.Sletten, Nucl. Phys. A151, 656(1970)
- IS) R.Vandenbosch, K.L.Wolf, Second IAEA Symposium on Physics and Chemistry of Fission, Vienna, 439(1969)
- 16) V.Meteg, R.Repnow, P.von Brentano, J.D.Fox, Z.Physik, 226, 1(1969)

- 17) H.C.Britt, S.C.Burnett, B.H.Erkkila, J.E.Linn, W.E.Stein, Phys. Rev. <u>C4</u>, 1444(1971)
- 18) K.L.Wolf, J.P.Unik, Phys.Lett. 38B, 405(1972)
- 19) G.N.Flerov, Yu.P.Gangraky, B.N.Markov, N.K.Khanh, D.D.Pulatov, N.H.Shadieva, JINR, R7-5018, Dubna(1970)
- 20) S.Bjørnholm, J.Borggreen, E.Hyde, Nucl. Phys. <u>A155</u>, 561(1970)
- 21) J.M.Alexander, R.Bimbot, Phys.Rev. <u>C5</u>, 799(1972)
- 22) P.Limkilde, G.Sletten, Nucl. Phys. A199, 504(1973)
- 23) P.A.Russo, R.Vandenbosch, M.Mehta, J.R.Tesmer, K.L.Wolf, Phys.Rev. <u>C3</u>, 1595(1971)
- 24) A.G.Belov, Yu.F.Gengreky, B.Dalheuren, N.K.Khanh, A.M.Kucher, Yad.Fis. <u>17</u>, 942(1973)
- 25) A.G.Belov, Yu.P.Gangrsky, B.Dalhsuren, A.M.Kucher, Yad.Fiz. <u>14</u>, 685(1971)
- 26) J.Kantels, private communication, Copenhagen (1967)
- 27) B.H.Erkkila, R.B.Leachman, Nucl. Phys. A108, 689(1968)
- 28) R.L.Fergusson, F.Plasil, G.D.Alam, H.W.Schmitt, Phys.Lett. <u>31B</u>, 526(1970)
- 29) Yu.P.Gangraky, N.K.Khanh, D.D.Pulatov, F.Z.Hien, JINR, R7-6466, Dubma(1972)
- 30) B.Dalhsuren, G.N.Flerov, Yu.P.Gangrsky, Yu.A.Lazarev, B.N.Markov, N.K.Khanh, Nucl.Phys. <u>A148</u>, 492(1970)
- 31) G.N.Flerov, V.A.Druin, JINR, R-2539, Dubna (1966)
- 32) D.L.Hill, J.A.Wheeler, Phys.Rev. 89, 1102(1953)
- 33) V.M.Strutinsky, Nucl. Phys. A95, 420(1967)
- 34) V.M.Strutinsky, Nucl. Phys. AI22, I(1968)
- V.M.Strutinsky and S.Bjørnholm, Nucl.Structure Dubna Symposium, IAEA, 431(1968)
- 36) E.Migneco, J.P.Theobald, Nucl. Phys. AII2, 603(1958)
- 37) B.M.Gokhberg, G.A.Otroschenko, V.A.Shigin, Sov.Phys.Dokl. 4, 1074(1960)
- 38) B.B.Back, J.P.Bondorf, G.A.Otroschenko, J.Pedersen, B.Rasmussen, Nucl. Phys. <u>A165</u>, 449(1971)
- 39) N.S.Rebotnov, G.N.Smirenkin, A.S.Soldatov, L.N.Usachev, S.P.Kapitsa, Yu.M.Tsypenyuk, Yad.Fis. <u>11</u>, 506(1970)
- 40) A.Bohr, FUAE, 15, 175(1968)

- 41) Yu.F.Gengrsky, B.Dalhsuren, B.N.Markov, T.Nagy, A.Lejtai, I.Winnai, L.Jeki, G.Kluge, JINR, R15-6147, Dubna(1971)
- 42) E.Konecny, H.J.Specht, J.Weber, H.Weigman, H.L.Fergusson, P.Osterman, M.Wald-Schmidt, A.Siegert, Nucl. Phys. <u>A187</u>, 426(1972)
- 43) H.J.Specht, J.Weber, E.Konecny, D.Heunsmann, Phys.Lett. <u>41B</u>, 43(1972)
- 44) K.L.Wolf, J.P.Unik, Phys.Lett. 43B, 25(1973)
- 45) L.Katz, A.P.Baerg, F.Brown, PUAE, 15, 188(1958)
- 46) S.P.Kapitsa, V.N.Melekhin, B.S.Zakirov, L.M.Zikin, E.A.Lukyanenko, Yu.M.Tsypenyuk, Pribori i technika exp. No.I, 13(1969)
- 47) Yu.P.Gengreky, B.Belhsuren, Yu.A.Lazerev, B.N.Markov, N.K.Khanh, Pribori i technika eksp. No.2, 63(1970)
- 48) Yu.P.Gangreky, B.N.Markov, Yu.M.Tsypenyuk, Yed.Fiz. <u>11</u>, 54(1970)
- 49) Yu.P.Gengrsky, B.N.Markov, Yu.M.Tsypenyuk, Phys.Lett. <u>32B</u>, 182(1970)
- 50) Yu.P.Gengrsky, B.N.Markov, I.F.Kharisov, Yu.M.Tsypenyuk, Yad.Fiz. <u>16</u>, 271(1972)
- 51) Yu.P.Gengrsky, B.N.Markov, I.F.Kharisov, Yu.M.Tsypenyuk, Zh. Eksp. Teor. Fiz.(Letters) <u>14</u>, 370(1971)
- 52) B.Foreman, G.Seaborg, J.Inorg. & Mucl.Chem. 7, 305(1958)
- 53) A.S.Penfold, J.E.Leis, Phys.Rev. 114, 1332(1959)
- 54) B.C.Cook, Nucl.Instr.dleth. 24, 256(1963) V.G.Ivanchenko, Trudy FIAN, 69, 151(1972)
- 55) A.G.Tikhonov, Doklady AN SSSR, No.3, I5I(1963)
- 56) B.Tamain, B.Pfeiffer, H.Wollnik, E.Konecny, Nucl. Phys. <u>A173</u>, 485(1971)
- 57) R.Vandenbosch, J.R.Huisenga, FUAE, 15, 284(1958)
- 58) H.Jungelaussen, Yad.Fis. 7, 89(1968)
- 59) S.JEgare, Nucl. Phys. A137, 241(1969)
- 60) A.Gilbert, A.C.W.Cameron, Can.J.Phys. 43, 1446(1965)
- 61) T.Sikkeland, J.E.Clarkson, N.H.Steiger-Shafrir, V.E.Viola, Phys. Rev. <u>CS</u>, 329(1971)

- 62) M.G.Silbert, LASL Report LA-4108-MS(1969)
- 63) H.Weigman, J.P. Theobald, Nucl. Phys. A187, 305(1972)
- 64) J.D.Jackson, Can.J.Phys. 34, 767(1956)
- 65) R.Vandenbosch, Phys.Rev. C5, 1428(1972)
- 66) A.V.Ignatyuk, N.S.Rabotnov, G.N.Smirenkin, A.S.Soldatov, Yu.M.Tsypenyuk, Zh.Eksp.Teor.Fiz. <u>61</u>, 1284(1971)
- 67) H.C.Britt, M.Bolsterly, J.R.Nix, J.L.Norton, Phys.Rev. <u>C7</u>, 805(1973)
- 68) H.C.Pauli, T.Lederberger, Nucl. Phys. A175, 545(1971)
- 69) M.Bolsterly, E.O.Fiset, J.R.Nix, J.L.Norton, Phys.Rev. <u>C5</u>, 1050(1972)

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Fig. I.

Systematics of spontaneous fission helf-lives for nuclei from U to Bk. The upper part is for

the ground states, while the lower one is for isomeric states.

- - even-even nuclei;
- even-odd, odd-even nuclei;
- odd-odd nuclei.



Fig. 2. One-dimensional fission barrier. The solid curve corresponds to a two-humped fission barrier; the dashed curve is the fission barrier for the liquid-drop model.



## Fig. 3.Schematic diagram of the experimental set-up; (I)microtron,

- (2) quadrupole lens doublet,
- (3) stopping target,
- (4) electron absorber,
- (5) spark counter,
- (6) electron current meter.



Fig. 4. Schematic diagram of the spark counter and the basic electronic circuit of the pulsed counter control: (I) target, (2) counter anode, (3) cathode, (4) counter tank, (5) protecting screen.







Fig. 6.

The decay curve for the  $^{239}Pu$ fission isomer (a), the measured yield as a function of the gamma-ray energy (b) and the excitation function of the reaction  $^{239}Pu(\textbf{y},\textbf{y}')^{239m}Pu$  (c).



Fig. 7. The photofizzion cross section as a function of photon energy.



Fig. 8.

The calculated cross sections for fission isomer formation in the reactions  $(\forall, \forall') - (a)$ ,  $(\forall, n') - (c)$ and the isomer-to-indiced fission ratios -(b); as a function of the gamme-ray energy (E<sub>X</sub>).



Fig. 9.

The compound nucleus level density  $(\rho)$  as a function of the excitation energy  $E^*$ ) for Pu and Am isotopes.

#### Fig. IO.

The calculated fission isomer excitation functions ( $\frac{5'/C_c}{c}$  is the probability of fission isomer formation,  $\mathbf{E}^{\mathbf{E}}$  is the excitation energy relative to the reaction threshold). Numbers 1,2 and 3 refer to calculations by eqs.(16,17,18), respectively.

Fig. II.

The calculated ratios  $\mathcal{T}_{f_f}$ as a function of the difference between the neutron binding energy and the outer barrier height relative to the isomer state  $(B_n - B_f)$  for Pu and Am isotopes.



Fig. I2.

The experimental and calculated values of the fission barrier perameters (E<sub>4</sub>, E<sub>B</sub>, E<sub>i</sub>) as a function of mass number (A) for Pu isotopes. o - our results, = - the values obtained from reactions producing fission isomers by charged particles 67).  $\mathbf{v}$  - the values from the (n,f) and (d,pf) reactions 38,63),  $\blacktriangle$  - the values from the ( $\gamma_{gf}$ ) reactions<sup>66)</sup>. - calculations of M.Bolsterly et al.<sup>69)</sup> - calculations of H. Fauli and T.Lederberger<sup>68)</sup>



Fig. 13.

The experimental and calculated values of fission barrier parameters as a function of mass number for Am isotopes. The notation is the same as that in fig. 12.