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SPONTANEOUSLY FISSIONING ISOMERS

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I. INTRODUCTION

AFTER K. A. Petrzhak and G. N. Flerov discovered in 1940 the spontaneous fission of nuclei,[1] a large number of isotopes of transuranium elements, for which this type of decay is very important, have been synthesized. It has been established that on going over from uranium to elements with larger atomic numbers the probability of spontaneous fission increases, and for certain isotopes, as for example Fm^{256} or the isotope of the element 104—kurchatovium with a mass of 260, this mode of radioactive decay becomes the main one.[2-4]

A study of laws governing spontaneous fission has shown that one of the basic parameters determining its probability is the parameter Z^2/A where Z is the charge of the nucleus and A is the atomic weight.

The appearance of this parameter is connected with the development of the liquid-drop model of fission proposed independently by Ya. I. Frenkel,[5] and N. Bohr and J. Wheeler.[6] According to the liquid-drop model the fissionability of a nucleus is the larger, the larger the parameter Z^2/A, and this is reflected in the fact that on going from thorium to kurchatovium the probability of spontaneous fission increases on the average by a factor of about 10^{29} (Fig. 1).

During the past decade our views concerning the fission process have undergone considerable change. It has been established that the treatment of nuclear fission as a process of separation of an electrically charged drop is very approximate. The role of the internal structure of the nucleus during fission, and in particular spontaneous fission, turned out to be very large, and in a number of cases it even turned out to be decisive. For example, the experimentally observed fluctuations of the periods of spontaneous fission of nuclei can by no means be understood within the framework of the liquid-drop model. In fact, the curve describing the dependences of the periods of spontaneous fission on Z^2/A (see Fig. 1) has a maximum for all elements, i.e., the heavy isotopes with lower values of Z^2/A undergo spontaneous fission more readily than the light ones. At present it is clear that this is due to the influence of one-nucleon states on the fission barrier of nuclei. The influence of the internal structure of the nucleus manifested itself particularly strongly in the effect of the extremely sharp increase of the probability of the spontaneous fission of nuclei in anomalous isomer states observed in 1962 at the Nuclear Reactions Laboratory of JINR.[7] Detailed investigations of this effect carried out in recent years make it possible now to discuss certain new properties of the nuclei of heavy elements.

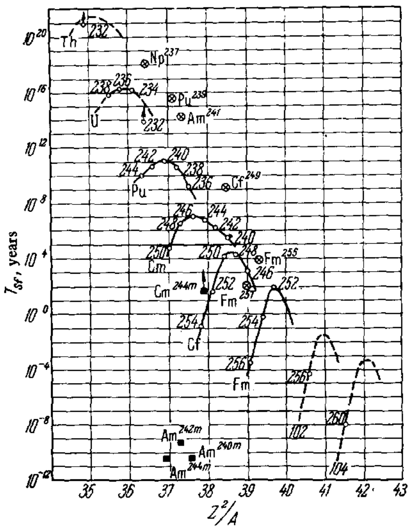

FIG. 1. Dependence of the spontaneous-fission half-life of nuclei T_{sf} on the parameter Z^2/A. The probability of spontaneous fission of a nucleus per unit time, λ, is given by the formula $\lambda = 0.7/T_{sf}$.

II: OBSERVATION OF THE SPONTANEOUS FISSION OF NUCLEI IN THE ISOMER STATE

The beginnings of the investigations that led to the discovery of spontaneous fission of nuclei in the isomer state date from 1961 when intense beams of heavy ions were obtained on the cyclotron of the Nuclear Reactions Laboratory at the JINR and when experiments were started to synthesize spontaneously fissioning isomers of transuranium elements. At the time no data were available concerning the spontaneous fission of isotopes of elements beyond fermium (Z = 100), but it was expected that their lifetimes would be short. Apparatus was therefore developed which made it possible to detect the spontaneous fission of nuclei with a half-life of more than 0.001 sec.

Figure 2 is a schematic diagram of such an experimental setup. The nuclei of the heavy elements produced when the target was bombarded with heavy ions acquired a sufficiently large momentum and were ejected from the target. Falling upon a moving collector they were stopped and moved together with the collector towards detectors of fission fragments. In the first experiments the fission fragments were detected with the aid of two ionization chambers. From a knowledge of the ratio of the counts in the chambers and the rate of revolution of the collector of the recoil nuclei, it is possible to estimate the period of spontaneous fission of the nuclei.

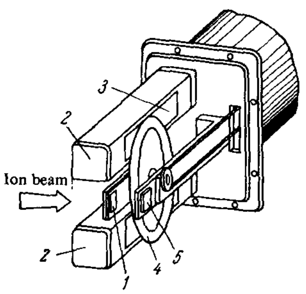

FIG. 2. Schematic diagram of the experimental setup for observing the spontaneous fission of nuclei with a short lifetime. 1—target; 2—ionization chambers; 3—window of ionization chamber; 4—aluminum disc collector of nuclei of transuranium elements; 5—collector of the ion current.

Since no external beam of ions was available at the time, the experimental setup was placed between the dees inside the cyclotron chamber.

The first stage in the investigations consisted of background experiments, namely irradiation in which no isotopes that decayed in a short time by means of spontaneous fission were produced. In fact, when a U^{238} target was bombarded with O^{16} ions having an energy of about 100 MeV, an excited fermium nucleus with mass 254 was produced. The principal decay process of this excited nucleus is the evaporation of 4-6 neutrons. This is accompanied by the production of the well-known isotopes of fermium with masses no less than 248 whose α decay rates are longer than 30 sec, whereas their probability of spontaneous fission is smaller by a factor of thousands than the probability of α decay. One must bear in mind that in addition to the neutron evaporation more complex reactions accompanied by the emission of charged particles (α particles, protons, etc.) are also possible. However, in this case known isotopes would be produced which live longer than the above-mentioned fermium isotopes and which do not decay by spontaneous fission.

In the light of this, it was most unexpected that in bombarding U^{238} with O^{16} ions some unknown isotope was produced which decayed by fission with a half-life close to 0.014 sec.[7]

The same isotope was also produced when U^{238} was bombarded with Ne^{20} and Ne^{22} ions. The effect was small—in the first experiments approximately one fission event was recorded per hour. However, the efficient operation of the electronic circuitry made it possible to carry out measurements even with such a small number of events. The results of the experiments with ionization chambers were subsequently confirmed with the aid of photographic emulsions.[8] The photographic emulsion method was also used for a more accurate determination of the half-life.[9] The measured life-time of the fissioning 0.014-sec isotope turned out to be much shorter than the half-life of all isotopes which could be produced in the bombardment of U^{238} with O^{16} ions.

Data on the Cf-Fm isotopes obtained in reactions proceeding through the formation of a compound nucleus followed by the evaporation of neutrons, protons, or α particles are presented in Table I. It is seen that the half-lives of these isotopes are much longer than 0.014 sec.

Table I does not include data on Np-Cm isotopes produced as a result of transfer reactions of several neutrons from O^{16} to U^{238}.

The half-lives of the isotopes of these elements are also long, and the probability of spontaneous fission is extremely small. Therefore the only explanation of the observed effect was that some known isotope in the isomer state undergoes fission.

The fact that the half-life of this isomer turned out to be very short, complicated considerably its identification. Both the methods of chemical separation of the elements and the mass-separator methods turned out to be inconvenient for investigations of an isotope with a life-time of 0.014 sec. The only possible way of identification of the isomer consisted in the use of the method of crossing reactions. Fortunately, it turned out that the observed isomer can be synthesized not only by using reactions involving multiply charged ions, but also by bombardment of the appropriate target with α particles, deuterons, and neutrons.[10-15] Moreover, in the case of deuterons and neutrons the cross section for the production of the fissioning isomer turned out to be largest. The aggregate of all the data on the yield of the fissioning isomer with $T_{1/2} = 0.014$ sec made it possible to arrive rather quickly at the conclusion that the observed decay is of the isomer Am^{242mf}.* The simplest reaction which leads to the production of this isomer is the $Am^{241}(n\gamma)Am^{242mf}$ reaction with a neutron energy of 1-3 MeV, concerning which preliminary data were obtained in [16].

Data on the simplest reactions leading to the production of the Am^{242mf} isomer are presented in Table II.

The Am^{242} isotope has been synthesized long ago and it was established that there are two states of this isotope.[17] In the ground state Am^{242} decays by β^- decay and K capture in the course of 16 hours. A 48-keV isomer state of Am^{242} is also known which decays mainly by means of a gamma transition to the ground state in the course of 152 years. The spontaneous fission period of this isomer which turned out to be $(8 \pm 3) \times 10^{11}$ years has been measured very recently.[18] One may expect that the period of spontaneous fission of Am^{242} in the ground state will be close to that value. Assuming that in our case we have observed the spontaneous fission of the previously unknown Am^{242} isomer, we reach the conclusion that compared with the ground state the probability of spontaneous fission in the isomer state is larger by a factor of approximately 10^{21}.

On the face of it, it may appear that the hypothesis of a "delayed fission" mechanism, according to which the isomer undergoes β or α decay in the course of 0.014 sec and the daughter nucleus has an excitation energy close to the fission barrier, i.e., 4-5 MeV, is just as simple.

The simplest estimates made for a superallowed β transition indicate that the excitation energy of the isomer should be close to 9 MeV, which is quite improbable.

The same is also true for α decay. Indeed, in order that the period of the α decay of Am^{242mf} be close to 0.014 sec, it is necessary for the α-decay energy to be

*Below we denote for simplicity isomers that decay by fission by the symbol mf.

Table I

Reaction	Isotope	Half-life	Principal decay mode	Period of spontaneous fission
$U^{238}(O^{16}, 3n)$	Fm^{251}	7 hour	E. C.: $\alpha(\sim 1\%)$	
$U^{238}(O^{16}, 4n)$	Fm^{250}	30 min	α	
$U^{238}(O^{16}, 5n)$	Fm^{249}	150 sec	α	
$U^{238}(O^{16}, 6n)$	Fm^{248}	0.6 min	α	
$U^{238}(O^{16}, p2n)$	Es^{251}	36 hour	E. C.: $\alpha(\sim 0.53\%)$	
$U^{238}(O^{16}, p3n)$	Es^{250}	8 hour	E. C.	
$U^{238}(O^{16}, p4n)$	Es^{249}	2 hour	E. C.: $\alpha(\sim 0.13\%)$	
$U^{238}(O^{16}, \alpha 2n)$	Cf^{248}	350 days	α	$1.5 \cdot 10^4$ years
$U^{238}(O^{16}, \alpha 3n)$	Cf^{247}	2.45 hour	E. C.	
$U^{238}(O^{16}, \alpha 4n)$	Cf^{246}	36 hour	α	2100 years
$U^{238}(O^{16}, \alpha 5n)$	Cf^{245}	44 min	E. C.: $\alpha(30\%)$	

greater than 8 MeV. Taking into account the fact that after the α decay the daughter nucleus should be excited up to 4–5 MeV, we again arrive at the conclusion that the excitation energy of the isomer must be very large (~ 7 MeV).

Thus the assumption that spontaneous fission of an isomer of Am^{242} was observed appeared to be the most sensible, even after the first experiments.

This assumption, simple in itself, leads under more careful examination to rather considerable difficulties. If the energy of the isomer state is low, then it is difficult to comprehend the strong increase in the fission probability. If, on the other hand, the energy is high, then the difficulty consists in the necessity of explaining the reason for the suppression of the gamma transitions from the isomer level of Am^{242}—a nucleus with an odd number of protons and neutrons for which the number of levels is large even in the region of 0.5–2.0 MeV.

After the discovery of the fissioning isomer Am^{242mf}, experiments were set up in the search of new fissioning nuclei of the same type.

The success of subsequent investigations was facilitated to a considerable extent by the discovery in 1962 of a new type of detector of fission fragments—the dielectric detector.[19-21] The principle of the operation of these detectors is based on the fact that in irradiating such materials as mica and glass the structure of the substance is disturbed much more strongly at points hit by fission fragments than at points hit by lighter particles. As a result of this, etching of material bombarded with fission fragments with hydrofluoric acid produces on its surface tracks that are visible in the microscope. An enormous advantage of these detectors is the practically complete absence of background. Using such detectors one is able to observe the decay of nuclei produced in reactions with heavy ions having very small cross sections. For example, such detectors made it possible to observe a number of additional isotopes which decay by fission and which are apparently in the isomer state.

The principle of operation of the experimental apparatus employed in subsequent investigations was practically no different from that of the apparatus employed in the first experiments. As an example we show in Fig. 3 a diagram of a setup in which the collection and transport of recoil nuclei emitted from the irradiated target is carried out by means of an infinite conveyer ribbon, and the fission fragments are registered by glass or mica detectors.

It was observed rather soon that when Pu^{242} is bombarded with B^{11} ions an isotope is produced which decays by fission after ~ 0.001 sec.[22] Considerations analogous to those used in the analysis of data on the Am^{242mf} isomer led to the belief that one was again observing the spontaneous fission of some isomer. This isomer, discovered in Dubna, was subsequently identified at the Lawrence Radiation Laboratory of the University of California. Surprisingly it turned out that as a matter of fact the isomers Am^{240mf} and Am^{244mf} undergo fission with close half-lives.[23]

Experimental investigations of Am^{240mf} showed that, just as Am^{242mf}, it is produced with the largest probability in reactions with neutrons.[24]

A fissioning isomer with a half-life close to 3.5 sec was observed when U^{238} was bombarded with O^{16} ions.[25] Regarding this isomer, it is only possible to say that its atomic number $Z \leq 100$ and its mass $M \leq 251$.

During the past two years efforts were directed, on the one hand, towards a search of comparatively long-lived isomers, and, on the other, towards a search of

Table II

Reaction	Energy of bombarding particle, MeV	Reaction cross section, cm²
$U^{238}(B^{11}, \alpha 3n)Am^{242mf}$	60	$6 \cdot 10^{-32}$
$U^{238}(B^{10}, \alpha 2n)Am^{242mf}$	60	$\sim 4 \cdot 10^{-33}$
$Am^{243}(\alpha, \alpha n)Am^{242mf}$	40	10^{-31}
$Pu^{239}(\alpha, p)Am^{242mf}$	40	$4 \cdot 10^{-32}$
$Pu^{242}(d, 2n)Am^{242mf}$	12	$3 \cdot 10^{-30}$
$Am^{243}(n, 2n)Am^{242mf}$	14	$1.5 \cdot 10^{-28}$
$Am^{241}(n, \gamma)Am^{242mf}$	1–3	$\sim 10^{-29}$

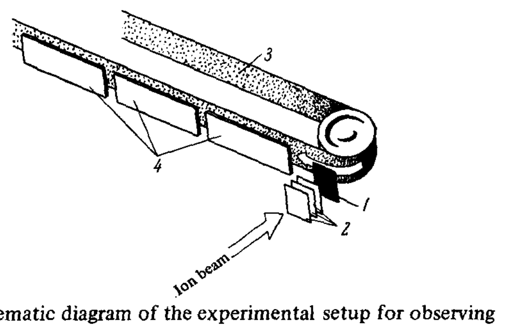

FIG. 3. Schematic diagram of the experimental setup for observing the spontaneous fission of nuclei with short lifetimes with a collector of recoil nuclei—a ribbon conveyer. 1—target; 2—filters for changing the ion energy; 3—ribbon collector of transuranium elements; 4—glass detectors registering the fission fragments of the nuclei.

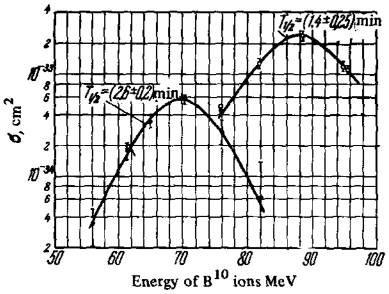

FIG. 4. Dependence of the production cross section of fissioning isotopes with $T_{1/2}$ = 2.6 min (Am^{234}) and 1.4 min (Am^{232}) on bombarding Th^{230} with B^{10} ions on the energy of the B^{10}.

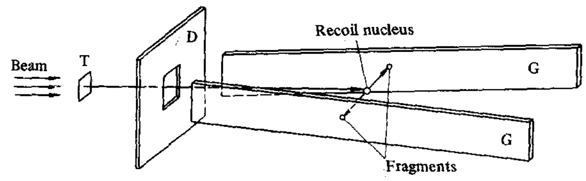

FIG. 5. Schematic diagram of the experiment for detecting the spontaneous fission of nuclei decaying in flight. T–target; D–diaphragm transmitting to the detectors of fission fragments nuclei produced in the evaporation of neutrons from an excited compound nucleus; G–glass detectors detecting fission fragments of nuclei.

isomers which are so short-lived that they decay in flight, scarcely having left the irradiated target. Spontaneously fissioning isotopes decaying after 1 min and 2.6 min were synthesized when a Bi^{209} target was bombarded with Ne^{22} ions and U^{233} and Th^{230} targets were bombarded with B^{11} and B^{10} ions respectively.[26–28] These isotopes were identified by measuring their excitation curves, although one might hope that in the future the mass-separator method will be employed in investigations of these isotopes.

When a multiply-charged ion coalesces with a nucleus of the bombarded target, an excited compound nucleus is produced. Fission or evaporation of a number of neutrons are the principal modes of decay of such a nucleus in the case of heavy elements. The average number of neutrons emitted from the nucleus changes in accordance with the change of the initial excitation energy. The dependence of the production cross section of each isotope in neutron evaporation on the ion energy has a characteristic peaked shape.

Figure 4 shows the experimentally obtained dependence of the production cross section of the isotope fissioning in 2.6 min on the energy of the B^{10} ions. The analysis of the curve shown in Fig. 4 led the authors to the conclusion that it is the Am^{234} isotopes that undergo fission.

In the case of the isotope with $T_{1/2}$ = 1.0 min the authors of the paper voiced the assumption that it is the Np^{228} isotope which undergoes fission.

A fissioning isotope decaying in 1.4 min has been obtained[28] rather recently when Th^{230} was bombarded with B^{10} ions. The experimental data show (see Fig. 4) that Am^{232} produced in the reaction $Th^{230}(B^{10}, 8n)Am^{232}$ undergoes decay.

No experimental data about the properties of the isotopes Np^{228}, Am^{232}, and Am^{234} were available until recently. A rough extrapolation of data on the spontaneous fission periods of uranium and plutonium isotopes to the masses 228, 232, and 234 shows that their spontaneous fission periods should be no less than 10^8 years for U^{228} and 10^2 years for Pu^{232} and Pu^{234}. Such an estimate does not contradict the calculated data of V. E. Viola, Jr., and B. D. Wilkins[29] for the fission barriers of U^{228}, Pu^{232}, and Pu^{234}.

There are no grounds for expecting that neighboring odd-odd nuclei should have shorter periods of spontaneous fission. Therefore one can, apparently, completely exclude the assumption that one is observing spontaneous fission on Np^{228}, Am^{232}, and Am^{234} in the ground state. It is most likely that the isotopes Np^{228}, Am^{232} and Am^{234} are in the isomer state from which the decay occurs by fission.

At the same time the data are, at present, insufficient to exclude also the hypothesis of the occurrence of delayed fission of U^{228}, Pu^{232}, and Pu^{234} produced in the excited state with an energy close to their fission barrier after β decay of Np^{228}, Am^{232}, and Am^{234}.

In 1966 a fissioning isomer with a lifetime of about 10^{-7} sec was successfully observed in Dubna.[30] In the search for fissioning isomers with such lifetimes, apparatus was employed which made it possible to observe the fission of nuclei that decay in flight (Fig. 5).

Nuclei emitted from the target after a nuclear reaction due to a multiply-charged ion pass between two sufficiently long glass detectors. If the period of decay is close to the time of flight of the nuclei along the detector, then there is an appreciable probability of registering the fission fragments. The geometry of the experiment was such that nuclei obtained in the evaporation of neutrons from compound nuclei were registered most efficiently.

The distribution of tracks along the length of the detectors allowed one to make a rough estimate of the half-life of the isotope. The isotope fissioning in a time $T_{1/2} = 10^{-7}$ sec was only observed when U^{238} was bombarded with C^{12} ions.[30] The dependence of the production cross section of this isotope on the energy of the C^{12} ions is shown in Fig. 6. The $\sigma = f(E)$ curve has a shape characteristic for the evaporation reaction of four neutrons, i.e., it is the previously well investigated Cf^{246} isotope which undergoes fission; in the ground state this isotope emits α particles with $T_{1/2}$ = 36 hours. The spontaneous fission period of this isotope in the ground state is 2×10^3 years.

In this case one is dealing with an isomer for which the probability of spontaneous fission increases by about a factor of 10^{17} compared with the ground state.

Simultaneously with the search for new fissioning isomers and directly after the observation of Am^{242mf},

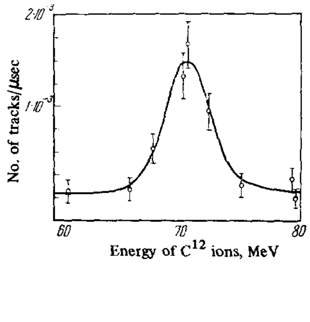

FIG. 6. Dependence of the production cross section of the isotope fissioning with $T_{1/2} = 10^{-7}$ sec when bombarding U^{238} with C^{12} ions on the energy of the C^{12}.

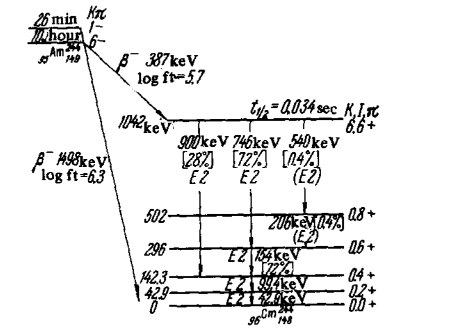

FIG. 7. Decay scheme of the Cm^{244m} isomer. Electromagnetic transitions from the isomer state to the levels of the rotational band are strongly forbidden on account of the large difference in the value of the projection of the spin on the symmetry axis of the nucleus.

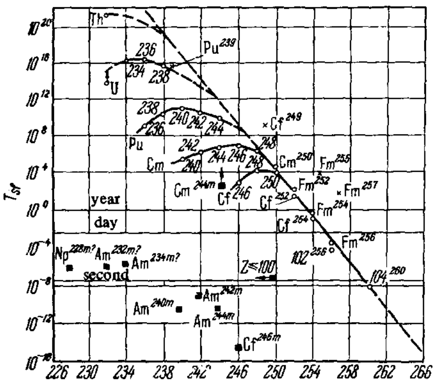

FIG. 8. Dependence of the half-life for the spontaneous fission of the nuclei T_{sf} on the mass of the nuclei: ■—isomers; ○—even-even nuclei; X—nuclei with an odd number of protons or neutrons.

attempts were made to observe spontaneous fission of already known isomers.

R. Vandenbosch and his co-workers[31] investigated the two-quasiparticle isomer Cm^{244m} whose decay scheme is shown in Fig. 7. As a result of the experiments which were carried out, it was shown that the period of spontaneous fission of the isomer Cm^{244m} is not less than 130 years. The period of spontaneous fission of Cm^{244} in the ground state is 1.3×10^7 years, i.e., in the case of the Cm^{244m} isomer if there is an increase in the rate of the spontaneous fission of the isomer, then it is by no more than a factor of 10^5.

The data on the spontaneous fission of the isomers of transuranium elements (including Np^{228}, Am^{232}, and Am^{234}) are presented in Fig. 8 and in Table III.*

III. THE DEVELOPMENT OF THE IDEAS CONCERNING FISSIONING ISOMERS IN THE TRANSURANIUM ELEMENTS

As has been seen above, the discovery of fissioning isomers occurred to some extent accidentally. Before the appearance of the first experimental work there was no theoretical work in which the possibility of spontaneous fission of nuclei in the excited state was considered.

Speaking of isomer isotopes of transuranium elements, one should mention that eight cases of isomerism were known in this region before the fissioning isomers were observed (Table IV). In all instances one can readily not only understand the reason for the retardation of the gamma transitions from the metastable states but also explain the structure of these states.

Even-odd and odd-odd nuclei have isomer states with energies amounting usually to several dozen keV. For even-odd nuclei the forbiddenness of the gamma transition is due to the difference in the value of the spin of the states of the odd nucleon. The low-energy states of odd-odd nuclei are as a rule connected with two configurations of the odd nuclei. Very often states are realized which correspond to parallel and antiparallel orientations of the spins of both particles with a projection of the spin on the axis of the nucleus $K = (\Omega_p \pm \Omega_n)$ where Ω_p and Ω_n are the spins of the odd protons and neutrons.

When $(\Omega_p + \Omega_n)$ and $(\Omega_p - \Omega_n)$ differ from each other by several units, it is possible that one of the states will be an isomer state.

Even-even nuclei also have isomer states. However, the energy of their isomer levels is close to 1 MeV since one of the nucleon pairs has to be broken apart for their production. For instance, in the case of Cm^{244} which we have already mentioned the isomer state was produced when a neutron pair was broken apart. As is seen from the cited decay scheme (see Fig. 7), the gamma transition from the metastable state I, $K\pi = 6, 6^+$ takes place only to rotational levels with K = 0 (K is the spin projection on the symmetry axis of the nucleus). At the same time, there appears additional forbiddenness for the gamma transition because of the considerable difference in the value of K.

In recent years a number of papers have been published in which ideas are expressed concerning the possible existence of new types of isomer states. At the same time, assumptions were made that an increase in the probability of spontaneous fission may occur in some of these states.

Table III

Isomer	Period of spontaneous fission in the isomer state	Period of spontaneous fission in the ground state	Enhancement factor of spontaneous fission in the isomer state
Np^{228mf}	1 min	$> 10^8$ years	
Am^{232mf}	1.4 min	$> 10^2$ years	
Am^{234mf}	2.5 min	$> 10^4$ years	
Am^{240mf}	0.0008 sec	$\sim 10^{14}$ years	$\sim 10^{22}$
Am^{242mf}	0.014 sec	$\sim 10^{14}$ years	$\sim 10^{21}$
Am^{244mf}	0.001 sec	$\sim 10^{14}$ years	$\sim 10^{22}$
Cf^{246mf}	10^{-7} sec	$\sim 2 \cdot 10^3$ years	$\sim 10^{17}$
$Z \leqslant 10.0\ M \leqslant 251$	3.5 sec		
Cm^{244m}	> 130 years	$1.3 \cdot 10^7$ years	$< 10^5$

Table IV

Isomer	$T_{1/2}$	Energy of isomer	Spin of isomer, ħ	Radiation in isomer decay
$_{93}Np^{236}$	5000 years		6	β
$_{93}Np^{240}$	60 min		5	β, γ
$_{94}Pu^{237}$	0.18 sec	245 keV	1/2	
$_{95}Am^{242}$	152 days	48.6 keV	5	γ, α
$_{95}Am^{244}$	26 min	69 keV	1	β
$_{96}Cm^{244}$	0.034 sec	1042 keV	6	γ
$_{97}Bk^{248}$	9 years			
$_{99}Es^{254}$	38.5 hours			

*The isomer Am^{238} which decays by fission with a half-life of 60 μsec has recently been observed at the Niels Bohr Institute in Copenhagen.

L. K. Peker[32] expressed the hypothesis of possible isomer states of the odd-odd isotopes Np^{240}, Am^{242}, and Am^{244} which can be produced when two pairs of nucleons are broken apart (six-quasiparticle states). Such states should be characterized by an excitation energy close to 2 MeV and the spin projection K on the symmetry axis should have a rather large value ($K \gtrsim 23$). No quantitative estimates of the times of radiative transitions from these states have so far been made. One also cannot say anything about the spontaneous fission, since the fission barrier of americium and neptunium nuclei in such states is not known.

L. A. Sliv and Yu. I. Kharitonov[33] considered the role of residual np interactions in heavy nuclei. Allowance for these interactions should lead to the appearance of an extremum on the curve depicting the dependence of the excitation energy of the nucleus on the spin. States with large values of the spin produced as a result of the formation of nucleons may turn out to be metastable.[34] Table V presents data on the expected values of the spin and excitation energy for isomers of this nature. The question of the spontaneous fission of nuclei in such states remains open.

A very interesting hypothesis of a perfectly new type of isomer states—vortex isomers—is due to Ya. B. Zel-'dovich.[35] According to this hypothesis it is possible for nuclear matter which is a superfluid liquid to exist as a drop of this liquid, i.e., a nucleus with a quantum vortex along the axis of the drop. The total angular momentum of the nucleus in such a state, produced by the bosons, is $n\hbar = Z\hbar/2$, where n is the number of bosons whose role is played by α particles and Z is the charge of the nucleus. Figure 9 shows the expected dependence of the minimum energy of the nucleus E_m on its spin. As is seen from Fig. 9, the vortex state with $I = I_0 = Z\hbar/2$ can be considered an isomer state, a transition from which can take place with a decrease of the spin by a quantity no smaller than αI_0. A change in the equilibrium shape of the nucleus compared with the shape in the ground state is also characteristic of such isomer states.

It is of great interest to search for vortex isomers in reactions with heavy ions, since in this case there is a large probability for the production of compound nuclei with a large value of the spin. One would think that the probability of spontaneous fission of vortex isomers will differ from the fission probability in the ground state. However, at present it is of course difficult to say what this difference will be.

After the appearance of the experimental data on fis-

Table V

Nucleus	Configuration	Spin, ħ	Excitation energy, MeV
Bi^{210}	$g_{9/2}$, $h_{9/2}$	9	~0.25
Bi^{211}	$g_{9/2}$, $h_{9/2}$	25/2	~0.8
Po^{211}	$g_{9/2}$, $h^2_{9/2}$	25/2	~1.2
Po^{212}	$g^2_{9/2}$, $h^2_{9/2}$	16	~2,9
Po^{214}	$g^4_{9/2}$, $h^2_{9/2}$	20	~4.0
At^{214}	$g^3_{9/2}$, $h^3_{9/2}$	21	~3.5
Rn^{216}	$g^4_{9/2}$, $h^4_{9/2}$	24	~5,0
Ra }	$i^2_{11/2}$, $f^2_{7/2}$	16	~3.0–4.0
Th }	$i^2_{11/2}$, $i^2_{13/2}$	22	
U }	$g^2_{7/2}$, $i^2_{13/2}$	18	~3,0–4.0
Pu }	$j^2_{15/2}$, $f^2_{5/2}$		
Cm }			

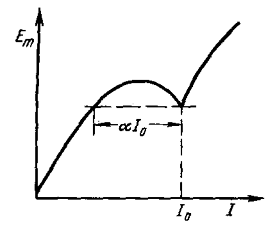

FIG. 9. Dependence of the minimum energy of the nucleus on its spin. I_0—the spin of the isomer state.

sioning isomers, it was assumed in a series of papers that the nuclei in the observed states have a shape which facilitates the process of tunneling through the fission barrier.

The hypothesis of possible nucleon configurations for which the shape of the nucleus in the isomer state will differ from the shape in the ground state in such a way that the probability of spontaneous fission of the isomer increases sharply, was considered by G. N. Flerov.[36] In particular, it was not excluded that the production of isomer states with enhanced fissionability of the nuclei is preceded by the production of anomalous excited states. This hypothesis is rather general in its nature, and is not readily amenable to quantitative calculations.

In analyzing data on the spontaneous fission of nuclei it is most important to know the fission barrier. Without knowing it, one essentially must not speak about calculating the probability of spontaneous fission.

All calculations of the fission barrier of nuclei carried out several years ago were related to the use of the liquid-drop model of the nucleus. Such an approach enabled one to explain successfully many experimental facts related in the first instance with fission of highly excited nuclei. At the same time, in the case of spontaneous fission the experimental data indicate that it is essential to take into account certain properties of nuclei that are not described by the liquid-drop model. The facts which altogether do not fit into the framework of the liquid-drop model are: the increase in the spontaneous-fission periods of odd-even nuclei compared with even-even nuclei and the very strong fluctuations of the periods for even-even nuclei.

Nowadays it is clear that in calculating the fission barrier it is essential to take into account corrections connected with the effect of one-nucleon states of the nucleus. In introducing such corrections one must use the so-called Nilsson diagrams describing the dependence of the energy of one-nucleon states on the deformation of the nucleus (Fig. 10). Nilsson diagrams are based on a calculation of nucleon states in the field of an anisotropic harmonic oscillator. It is of course clear that such a choice of potential should be considered as an approximation.

Particularly serious doubts as to the correctness of the behavior of the levels on the Nilsson diagrams arise for large deformations. The first attempt to take into account the effect of one-nucleon states on the fission barrier and correspondingly on the probability of spontaneous fission was made in the work of Johanssen[37] who estimated the difference in the barriers of neighboring nuclei due to the difference in the levels on which the nucleons are located on the Fermi surface. For even-even nuclei it was assumed that on increasing the deformation pairs of nucleons can pass from one level to another when these cross. At the same time, it was

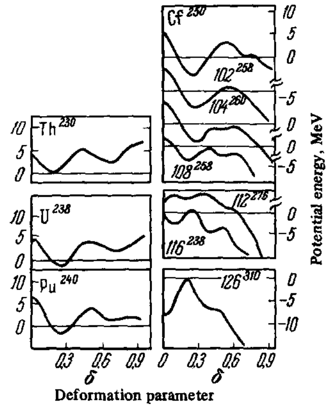

FIG. 11. Fission barriers of nuclei, calculated with account of shell effects in the work of V. M. Strutinskiĭ.[39]

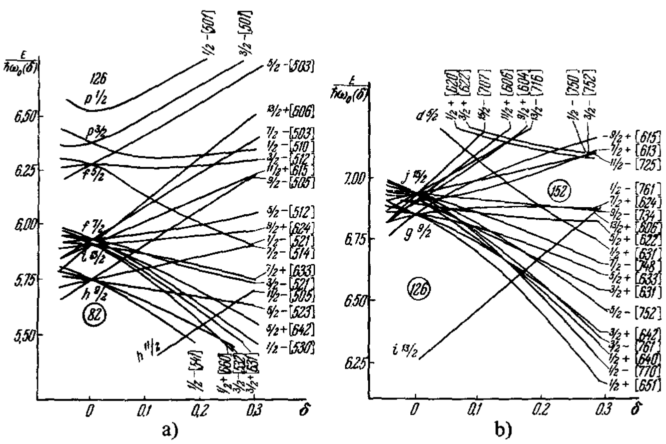

FIG. 10. a) Proton levels for Z > 82. b) Neutron levels for N > 126. The spin and parity of the level are indicated before the brackets. The numbers in the brackets denote $Nn_Z\lambda$ where N is the total number of oscillator quanta, n_Z is the number of oscillator quanta along the symmetry axis of the nucleus Z, λ is the projection of the angular momentum on the symmetry axis of the nucleus, δ is the deformation parameter that determines the eccentricity of the nucleus which has an ellipsoidal shape, E is the energy of the levels in units of $\hbar\omega_0(\delta)$ where $\hbar\omega_0(\delta)$ is the distance between the shells.

assumed that when it is deformed a nucleus changes its configuration in such a way that its total energy be at a minimum.

If there is only one nucleon on a given level, which is the case for odd nuclei, the picture changes. Now the odd nucleon can no longer pass from level to level during deformation because of spin and parity conservation of the nucleus.

Thus in the deformation of even-even nuclei the transition of nucleon pairs to levels which are being lowered can yield an energy gain and a corresponding lowering of the fission barrier; for odd nuclei such an effect is impossible. As a result of this even-even nuclei have a lower fission barrier.

The gain in the total energy of the nucleus due to the transition of nucleon pairs to levels which are being lowered depends on the location of the Fermi level which differs for different isotopes; this explains the observed fluctuations of the periods of spontaneous fission of even-even nuclei.

The model proposed by Johansson is rather crude since it does not take into account, for example, such appreciable effects as nucleon pairing. It is therefore natural that the predictions of the properties of heavy nuclei made on its basis are not borne out by experiments. However, the model does nevertheless reflect in a qualitative fashion correctly the effect of one-nucleon states on the fission barrier.

Recent work by V. M. Strutinskiĭ[38, 39] and also by W. D. Myers and W. J. Swiatecki[40] considered in detail the effect of the shell structure on the fission barrier of nuclei. Strutinskiĭ showed that for heavy nuclei the shell correction to the fission barrier calculated according to the liquid-drop model amounts to about 3 MeV, i.e., half of the fission barrier. Figure 11 taken from [39] depicts the fission barriers of various isotopes calculated by Strutinskiĭ. It is seen that the shape of the fission barrier changes strongly from nucleus to nucleus. Special attention should be drawn to the two-hump nature of the fission barrier of certain nuclei. Strutinskiĭ notes that the appearance of an additional minimum can give rise to an isomer state. Such states will be characterized both by a rather high energy and by a large deformation. The difference in the deformation of the ground and metastable states can lead to a strong hindrance of the gamma transitions. All of Strutinskiĭ's calculations were carried out for even-even isotopes. Obviously the appearance of a minimum at the fission barrier is also possible for certain odd isotopes; however, in this case the calculations are more complicated.

One hypothesis concerning "shape isomerism" was proposed by A. L. Malov, S. M. Polikanov, and V. G. Solov'ev.[41] The authors showed that it is possible for two-quasiparticle states with $K\pi = 12^-$ to exist (π is the parity of the nucleus), for which the equilibrium deformation parameter δ * is close to 0.32 whereas for the ground state $\delta \approx 0.24$. The energy of isomer states $K\pi = 12^-$ for odd-odd nuclei is close to 1.0–2.5 MeV. Many isotopes of Np, Am, Bk, and Es (Table VI) should have similar isomer states.

One of the questions which appears in the analysis of the possibility of the spontaneous fission of such isomers is the question whether the projection of the total spin K on the axis of symmetry of the nucleus is conserved in the tunneling process of fission.

Assuming that only the total spin and the parity are conserved in the fission of isomers with spin 12 and ad-

Table VI. Isomers in odd-odd nuclei with $K\pi = 12^-$ (K is the projection of the spin on the symmetry axis of a nucleus, π is the parity of the state)

Nucleus	Energy of isomer state, MeV	Deformation parameter of the nucleus in the isomer state, δ
Es^{248}	2.5–3.5	0.32–0.33
Es^{244}		0.32–0.33
Bk^{246}	2.5–3.0	0.32–0.33
Bk^{244}		0.32–0.33
Bk^{242}		0.31–0.32
Bk^{240}		0.31
Am^{246}	1.5–2.5	0.32–0.34
Am^{244}		0.32–0.33
Am^{242}		0.32
Am^{240}		0.32
Am^{238}		0.31
Np^{244}	1.0–2.5	0.32–0.33
Np^{242}		0.32–0.33
Np^{240}		0.31–0.32
Np^{238}		0.31
Pa^{236}	1.3–2.2	0.30

*δ is a parameter which defines the eccentricity of the nucleus which has the shape of an ellipsoid of revolution.

mitting the possibility of transition to the rotational levels with I = 12 and K≠ 12, one can expect an appreciable lowering of the barrier for these states.

Along with the effect of barrier narrowing, this will lead to a very strong increase in the probability of spontaneous fission.

An interesting effect which can lead to a sharp increase in the probability of spontaneous fission of a nucleus has been considered in the work of D. F. Zaretskiĭ and M. G. Urin.[42, 43] They developed the ideas about the effect of the mass coefficient on the fission process.

From the expression for the penetrability through a fission barrier

$$\lambda_f \sim \frac{\omega}{2\pi} \exp\left[-2\int_{\delta_0}^{\delta_f} d\delta \sqrt{2B[W(\delta)-E]}\right],$$

where ω is the frequency of quadrupole oscillations, δ is the deformation parameter of the nucleus, B is the mass coefficient, W(δ) is the potential energy of deformation, and E is the energy of the nucleus, it is seen that the probability of spontaneous fission depends strongly on the value of the mass coefficient B. Zaretskiĭ and Urin expressed the hypothesis that isomer states may exist which will be characterized by a complete disappearance of pair correlations. Their calculations show that this will be accompanied by a sharp decrease in the mass coefficient B which will approach the hydrodynamic limit. This will correspondingly cause a very strong increase in the probability of spontaneous fission (according to the estimates of Zaretskiĭ and Urin by a factor of about 10^{16}).

Thus, on the one hand, there has now appeared a hypothesis concerning the possible existence of new types of isomer states and, on the other, our ideas concerning the fission barrier have become considerably more profound.

It should, however, be noted that the considerations that have been developed are to an appreciable extent of a qualitative nature and further comparison with experimental data requires a quantitative treatment.

IV. INVESTIGATION OF THE PROPERTIES OF FISSIONING ISOMERS

In order to understand the nature of the observed effect, it is very important to determine in the first instance the energy and spin of the isomer states. The usual method for determining these quantities is an investigation of the gamma-ray and conversion-electron spectra, as well as that of α particles if α decay occurs.

Unfortunately, however, no forms of decay of the investigated isomers other than fission have so far been observed.

The experiments of R. Leachman et al.[44] have shown that the probability of α decay of the isomer $Am^{242\,mf}$ is smaller by at least a factor of ten than the probability of spontaneous fission. These results, by the way, made it possible to exclude completely the previously discussed mechanism of delayed fission following α decay.

A search for gamma radiation accompanying the decay of $Am^{242\,mf}$ was carried out by R. Diamond and F. Stevens. As a result of these experiments it was only established that there are less than a hundred gamma per fission.

In connection with the failure of the experiments to observe competing modes of decay, a series of experiments was set up to investigate the laws governing the production of fissioning isomers in nuclear reactions.

One of the directions of the investigations was the measurement of the so-called isomer ratios. The isomer ratio is the ratio of the production cross sections in the isomer and ground state. This quantity depends strongly both on the energy and on the spin of the level, as well as on the type of reaction used to obtain the isomer. For example, in the case of a state with a large spin it is more easily obtained in reactions with heavy ions than by bombarding the target with neutrons or protons. It is accordingly found that in reactions with light particles it is difficult to obtain a compound nucleus with large values of the spin.

The first data on the isomer ratio of Am^{242} synthesized in the Pu^{242}(d, 2n)Am^{242} reaction were obtained in experiments carried out on the cyclotron of the Atomic Physics Institute in Bucharest.[15] The value of the isomer ratio turned out to be small (3×10^{-4}) and depended weakly on the deuteron energy.

An analysis of these data has recently been carried out by V. P. Zommer and A. I. Prokof'ev[46] who calculated on the basis of the statistical approximation the dependence of the isomer ratio on the deuteron energy. In calculating the transition probability between different states no account was taken of the possible effect of the spin.

According to the calculations of Zommer and Prokof'ev there should be a rather strong dependence of the magnitude of the isomer ratio on the energy of the level (Table VII). Experimental data on the Pu^{242}(d, 2n)Am^{242} reaction and results of calculations with the assumption that the energy of the isomer level of $Am^{242\,mf}$ is 2.8 MeV are shown in Fig. 12. It is seen that there is rather good agreement between the calculations and the experimental data. In carrying out the calculations it was assumed that fission is the principal mode of decay of the $Am^{242\,mf}$ isomer. In addition to the Pu^{242}(d, 2n)Am^{242} reaction whose relative probability is rather large, other nuclear reactions were also investigated.

A comparison of the obtained data on the value of the isomer ratio for various reactions gives some idea of the value of the spin. Indeed, let us compare two reactions that take place with the production of a compound

Table VII

Energy of level, MeV	1.0	1.5	2.0	2.5	2.8	3.0
Isomer ratio	$9.3 \cdot 10^{-2}$	$2.2 \cdot 10^{-2}$	$4.8 \cdot 10^{-3}$	$9.9 \cdot 10^{-4}$	$4.2 \cdot 10^{-4}$	$2.3 \cdot 10^{-4}$

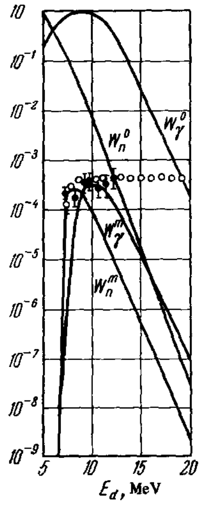

FIG. 12. Dependence of the isomer ratio on the deuteron energy. W_n^m and W_γ^m are the probabilities of production of the isomer state after emission of neutrons or gamma rays. W_n^0 and W_γ^0 are the transition probabilities to the ground state after emission of neutrons or gamma rays. ●—experimental results[15]; ○—theoretical data of[46].

nucleus and lead to the production of the isomer

$$U^{238} + B^{11} \rightarrow \alpha + 3n + Am^{242}$$

and

$$Pu^{242} + d \rightarrow Am^{242} + 2n.$$

The spin distribution of compound nuclei produced in these reactions will differ considerably (Fig. 13), since it depends on the mass and energy of the bombarding particle. If the spin of the isomer is large, one would expect that the probability of its production will be considerably larger for the U^{238} + B^{11} reaction. However, it turned out in fact that the value of the isomer ratio is practically the same for both reactions.[47] Table VIII presents experimental data on the value of the isomer ratio for various reactions leading to the production of the fissioning $Am^{242\,mf}$ isomer, and Fig. 14 depicts graphically the dependence of the isomer ratio on the average value of the spin of the compound nucleus. Data on the isomer ratio in the case of the known two-quasiparticle isomer Au^{196m} ($K\pi$ = 12^-, E = 600 keV) are indicated for comparison both in the table and on the figure. It is seen that the $Am^{242\,mf}$ isomer is produced with approximately equal probability in different reactions, whereas $Au^{196\,m}$ is produced with considerably more efficiency in the case where the compound nuclei have the larger spin.

These results are best interpreted if we assume that the spin of the fissioning $Am^{242\,mf}$ isomer is small (does not exceed several units of ħ). The best confirmation of the conclusion that the spin of $Am^{242\,mf}$ is small was recently obtained in experiments carried out on the cyclotron of the Atomic Physics Institute at Bucharest[16] where the production of $Am^{242\,mf}$ was observed in the reaction $Am^{241}(n, \gamma)Am^{242\,mf}$ with 1–3 MeV neutrons. The neutrons were obtained from the p + Li reaction.

The production cross section of $Am^{242\,mf}$ turned out to be close to 10^{-29} cm^2. The radiative capture cross section of 1–3 MeV neutrons by the Am^{241} isotope is unknown. According to rough estimates it should be 50–100 mb. This means that the isomer ratio of the investigated reaction is close to 10^{-4}, i.e., it does not differ considerably from that for 14-MeV neutrons.

In the radiative capture of 1–3 MeV neutrons the probability of the production of states of the compound nucleus Am^{242} with a spin larger than 6ħ is very small.

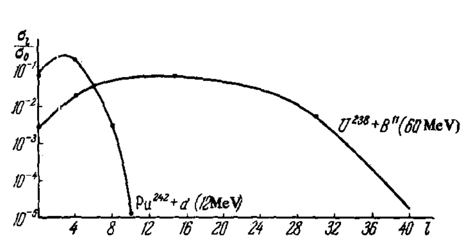

FIG. 13. Spin distribution of compound nuclei produced in the reactions Pu^{242} + d (19 MeV) and U^{238} + B^{11} (60 MeV).

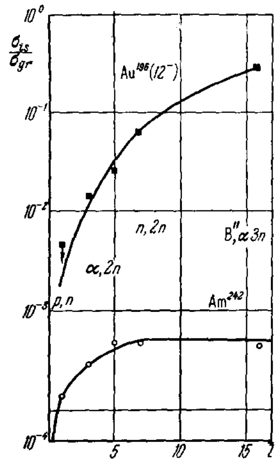

FIG. 14. Dependence of the isomer ratio for Am^{242} and Au^{196} on the average value of the spin of the compound nucleus.

If the spin of the $Am^{242\,mf}$ isomer is larger than 6ħ, then the isomer will only be produced after a gamma-ray cascade and therefore the probability of its production in the (n, γ) reaction should be appreciably smaller than in the (n, 2n) reaction; this is in fact not the case.

The results of experiments investigating the $Am^{241}(n, \gamma)Am^{242\,mf}$ reaction allow one to assume that the spin of $Am^{242\,mf}$ is small. The conclusion concerning the small value of the spin of $Am^{242\,mf}$ compels one to recall again the calculations of Zommer and Prokof'ev which are valid in this instance and which indicate that the energy of the isomer state should be close to 3 MeV.

The first experimental data concerning the energy of the isomer level were obtained for the Am^{240} isotope. In order to determine the energy of the level, measurements were made of the threshold of the $Pu^{241}(p, 2n)Am^{240\,mf}$ reaction leading to the production of a fissioning isomer with a lifetime of ~1 msec. These investigations were carried out on the electrostatic tandem generator of the Niels Bohr Institute in Copenhagen by S. Bjørnholm et al.[48] The exceptionally high stability of the proton beam along with the good en-

Table VIII

Reaction	Particle energy	$\bar{I}$	σ_{is}/σ_{gr}	
			$_{95}Am^{242}$	$_{79}Au^{196}$
n, γ	1/40 eV	0	$< 5 \cdot 10^{-7}$	
p, n	7 MeV 11 MeV	1 2	$1.5 \cdot 10^{-4}$	
d, 2n	10 MeV 14 MeV	3 5	$3 \cdot 10^{-4}$ $5 \cdot 10^{-4}$	0.015 0.025
n, 2n	14 MeV	7	$5 \cdot 10^{-4}$	0.065
$B^{11}\alpha 3n$	65 MeV	15	$5 \cdot 10^{-4}$	0.4

ergy resolution made it possible to carry out precise measurements. The obtained dependence of the cross section of the $Pu^{241}(p, 2n)Am^{240\,mf}$ reaction on the proton energy normalized to the induced fission cross section is shown on Fig. 15. It turned out that the shape of the curve is in good agreement with the formula

$$\sigma \approx 1-\left(1+\frac{\Delta E}{T}\right)e^{-\frac{\Delta E}{T}}$$

(where ΔE is the excitation energy relative to the reaction threshold and T is the temperature of the compound nucleus) obtained on the basis of calculations by Jackson who calculated the dependence of the probability of evaporation of various numbers of neutrons on the excitation energy of the nucleus.[49]

The results of the work investigating the $Pu^{241}(p, 2n)Am^{242\,mf}$ reaction showed that the threshold of this reaction is 3.05 MeV higher for the isomer than for the ground state. Assuming that the difference in the thresholds is equal to the energy of the metastable state of $Am^{242\,mf}$, the authors of [48] estimated it to be $E_i = 3.05 \pm 0.25$ MeV. The error is basically determined by the inexact knowledge of the mass of Am^{240}. The experimental error of determining the reaction threshold amounts of ~0.1 MeV.

The energy of the isomer state of $Am^{242\,mf}$ has recently been determined in a similar manner at the Atomic Physics Institute at Bucharest.[16] The $Am^{243}(n, 2n)Am^{242\,mf}$ reaction was studied in these experiments. Neutrons with an energy of up to 16 MeV were obtained from the d + d reaction. The deuterons were accelerated on a cyclotron. The resulting dependence of the cross section of the $Am^{243}(n, 2n)Am^{242\,mf}$ reaction on the neutron energy is shown in Fig. 16. The calculated curve is in good agreement with the experimental points for $E_i = 2.9$ MeV. The results of the latter show that for $Am^{242\,mf}$ the energy of the isomer state is $E_i = 2.9 \pm 0.4$ MeV.

Of course, the conclusion that the difference in the thresholds of the reactions leading to the production of the ground and isomer states is equal to the energy of the isomer state cannot be considered to be perfectly indisputable.

An analysis of the available experimental data on the thresholds of the (p, 2n) reactions was carried out in [48]. It turned out that for all the investigated reactions the production thresholds of nuclei in the excited state exceed the thresholds for the ground state exactly by the amount of the excitation energy. It can, nevertheless, not be entirely excluded that the energy of the isomer states is less than 3 MeV, and that by virtue of some unknown features of the structure of these states a direct transition to these states in the decay of an excited nucleus is strongly forbidden.

It can then be assumed that the transition must initially proceed to some level with an energy of ~3 MeV with a subsequent radiative transition to the isomer state.

So far no data are available which would clarify this question, and the conclusion of the authors of [48, 16] that the difference in the threshold of the reactions is equal to the energy of the isomer state can therefore be accepted as a working hypothesis. The question arises how one should understand the reason for the strong

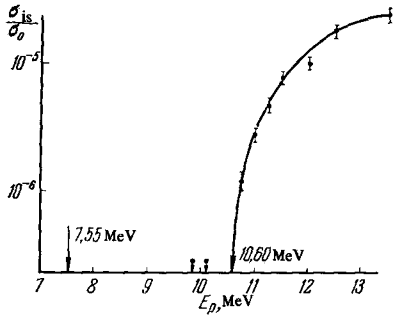

FIG. 15. Dependence of the cross section of the reaction $Pu^{241}(p, 2n)Am^{240\,mf}$ on the proton energy.

forbiddenness of radiative transitions to the ground state from a state with an energy of ~3 MeV and small spin. At present the only possible explanation is that the forbiddenness is due to the considerable difference in the deformation of the nucleus in the ground and isomer state.

Thus the results of the experimental investigations give grounds for assuming that a new form of isomer states of heavy nuclei noted for their large deformation, for which the penetrability of the fission barrier is greatly enhanced, has been observed. Undoubtedly this interpretation of the results of the investigations must be treated with caution. The conclusion drawn is apparently the only one which can be drawn on the basis of present-day ideas about the properties of heavy nuclei. An essential change in these ideas could in principle lead to other conclusions concerning the nature of fissioning isomers.

Speaking of further investigations in this field, one should draw special attention to experiments which make it possible to observe competing modes of decay of fissioning isomers (gamma and alpha decay). Although the first experiments conducted in this direction were unsuccessful, one might hope that an increase in the sensitivity of the measurement apparatus will open up possibilities for renewed searches. It is also very important to continue work synthesizing new fissioning isomers, and, in particular, to understand the preferential production of isomers of odd-odd nuclei. This can be explained as being due to two reasons:

1) Even-even and odd-even nuclei also have similar

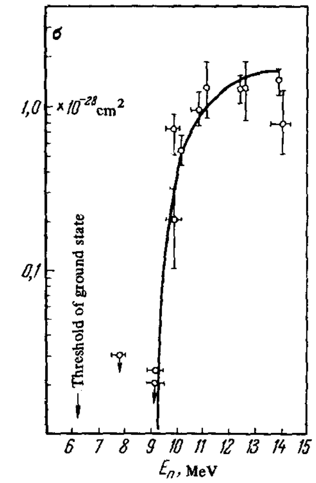

FIG. 16. Dependence of the cross section of the reaction $Am^{243}(n, 2n)Am^{242\,mf}$ on the neutron energy.

isomer states; however, the decay time from these states via fission is as a rule very short, and the apparatus used so far made it possible to observe only the Cf^{246mf} isomer.

2) Two odd nucleons facilitate the production of isomer states.

The probability of isomerism for even-even and odd-even nuclei turns out therefore to be considerably smaller.

Experimental investigations of the fission process of the discovered isomers are also of definite interest. It cannot be excluded that peculiarities of the structures of isomers may influence the nature of their fission; it may, therefore, be that by studying the peculiarities of the fission process of isomers one will succeed in obtaining information about the nature of the fissioning isomers.

Experimental and theoretical investigations of fissioning isomers are at present being continued, and there is no doubt that they will enrich our knowledge of the properties of heavy nuclei.

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