

FORMATION OF A SPONTANEOUSLY FISSIONING ISOMER IN REACTIONS INVOLVING  
 $\alpha$  PARTICLES AND DEUTERONS

G. N. FLEROV, S. M. POLIKANOV, K. A. GAVRILOV, V. L. MIKHEEV, V. P. PERELYGIN, and  
A. A. PLEVE

Joint Institute for Nuclear Research

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A spontaneously fissioning isomer previously detected in experiments with accelerated oxygen and neon was observed by irradiating plutonium and americium with  $\alpha$  particles and deuterons. The spontaneously fissioning nucleus has  $Z \leq 95$  and  $A \leq 242$ .

WE have previously reported<sup>[1-3]</sup> on experiments of a spontaneously fissioning isomer, produced when  $U^{238}$  is bombarded with heavy ions. One of the main conclusions of these investigations was that for the observed isomer the probability of spontaneous fission is more than  $10^{10}$  times larger than the probability of spontaneous fission for the ground state of the nucleus.

For a more detailed investigation of the observed phenomenon it would be most important to identify accurately the obtained isomer and to obtain information concerning its spin. It is known, however, that reactions with heavy ions proceed via a large number of channels. They are consequently not very convenient for an exact identification of any particular isotope. We have therefore undertaken to obtain a spontaneously fissioning isomer in reactions with  $\alpha$  particles and deuterons.

The experiments were carried out with the extracted beam of the 150-centimeter cyclotron of the I. V. Kurchatov Atomic Energy Institute. The apparatus used in the experiments is analogous to that described previously<sup>[1]</sup>. As in the experiments with the heavy ions, the products of the nuclear reactions were knocked out of the target as a result of the momentum of the bombarding particles. Inasmuch as the range of the recoil nuclei from plutonium bombarded with deuterons of energy  $\sim 20$  MeV is estimated by Bryde et al<sup>[4]</sup> and by Winsberg and Alexander<sup>[5]</sup> at  $\sim 25 \mu\text{g}/\text{cm}^2$  of  $\text{PuO}_2$ , particular attention was paid to the target preparation procedure. The target material was first rid of impurities by an ion-exchange method. It was then dissolved in a mixture of ethyl alcohol and acetone with a small amount of nitrocellulose and deposited layer by layer on the substrate by a capillary method (15-20 layers). Each

layer was heated 15 minutes at  $500^\circ\text{C}$ . The amount of plutonium and americium on all the prepared targets was approximately  $60 \mu\text{g}/\text{cm}^2$ . The uranium target was  $500 \mu\text{g}/\text{cm}^2$  thick. The recoil nuclei were collected on an aluminum disc rotating at 850 rpm. The spontaneous fission fragments were registered with semiconductor detectors and photographic plates.

We used 39.6-MeV  $\alpha$  particles to bombard targets made of mixtures of  $\text{Pu}^{242}$ ,  $\text{Pu}^{241}$ , and  $\text{Pu}^{240}$ ,  $\text{Am}^{243}$  with  $\text{Am}^{241}$  admixtures,  $\text{Pu}^{239}$ , and  $U^{238}$ . Spontaneous-fission fragments with short lifetimes were registered in the experiments with the  $\text{Am}^{243,241}$ ,  $\text{Pu}^{242-240}$ , and  $\text{Pu}^{239}$  targets. The effect was not observed in the experiment with  $U^{238}$ . Data on the decay of spontaneously fissioning nuclei, obtained in experiments with  $\alpha$  particles, are shown in Fig. 1, in which only one half-life,  $12.6 \pm 1.1$  msec, can be separated.

The yield curves for spontaneously fissioning nuclei from bombarded plutonium and americium

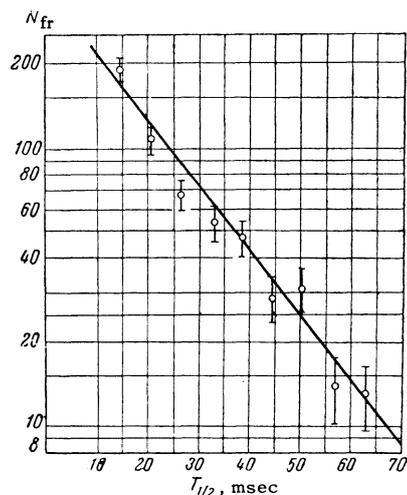


FIG. 1

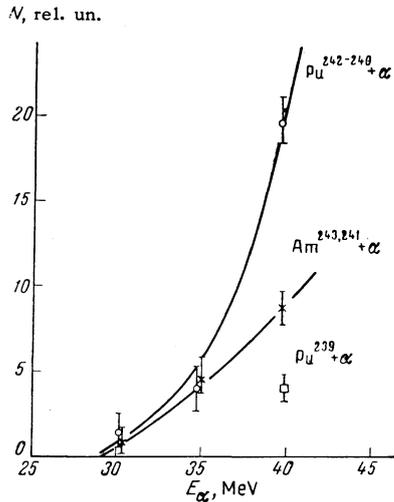


FIG. 2

targets are shown in Fig. 2. The  $\alpha$ -particle energy was varied with the aid of aluminum foils. Estimates of the cross section for the production of spontaneously fissioning nuclei from bombarded  $\text{Pu}^{239}$  yield a value  $4 \times 10^{-32} \text{ cm}^2$ , which should be regarded as a lower limit, since the angular distribution of the reaction products is unknown. In experiments with deuterons, targets of  $\text{Pu}^{242-240}$  and  $\text{Pu}^{239}$  were used. The yield of spontaneously fissioning nuclei from  $\text{Pu}^{242-240}$  bombarded with 19.8-MeV deuterons is double (per  $\mu\text{A}\cdot\text{hr}$ ) the yield from a similar target bombarded with  $\alpha$  particles of energy 39.6 MeV. When the energy of the deuterons was decreased to 16.2 MeV, no reduction in the yield of the spontaneously fissioning nuclei was observed. The effect was not observed in the experiment with  $\text{Pu}^{239}$ . Data on the decay of spontaneously fissioning nuclei, obtained in experiments with deuterons ( $\text{Pu}^{242-240} + d$ ), are shown in Fig. 3.

The half-lives obtained in experiments with  $\alpha$  particles ( $12.6 \pm 1.1 \text{ msec}$ ) and with deuterons ( $14 \pm 2 \text{ msec}$ ) are in good agreement with the value  $13.5 \pm 1.2 \text{ msec}$  obtained in experiments with heavy ions<sup>[6]</sup>. The agreement of the half lives indicates that one and the same spontaneously-fissioning isomer is synthesized in either the experiments with the heavy ions or in the experiments with the  $\alpha$  particles and deuterons. Since this isomer can experience, in addition to spontaneous fission, also  $\alpha$  decay with energy (in accordance with the systematics of<sup>[7]</sup>) exceeding 8 MeV, corresponding to a life time  $\sim 10^{-2} \text{ sec}$ , experiments were set up to observe the  $\alpha$ -decay branches. So far, however, only the upper limit of this effect was established,  $\sigma_{\alpha}/\sigma_{\text{fr}} < 50$ .

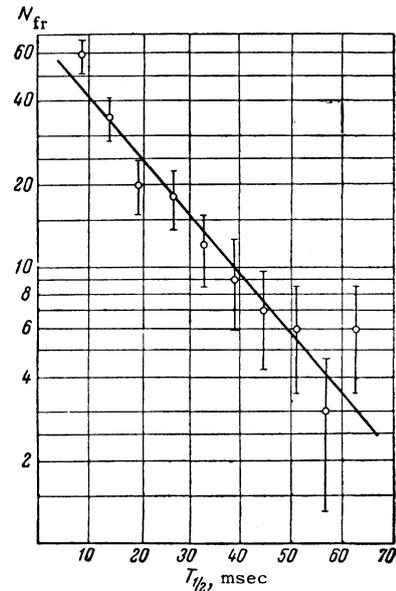


FIG. 3

From the experiments on deuteron bombardment of  $\text{Pu}^{242-240}$  it follows that the spontaneously fissioning isomer has an atomic number  $Z \leq 95$ , while from the experiments with  $\alpha$ -particle bombarded  $\text{Pu}^{239}$  it follows that the mass number of this isomer is  $A \leq 242$ . The energy balance of the different channels for the interaction between  $\alpha$  particles and  $\text{Am}^{243,241}$  shows that the most likely to experience spontaneous fission is an americium isotope, although plutonium isotopes cannot be fully excluded. As to the neptunium isotopes, they should be produced by reactions of the type  $(\alpha, \alpha' \text{He}^3)$ ,  $(\alpha, \text{He}^3 2p)$ ,  $(\alpha, 2\alpha')$  etc, for which the energy in our experiments is insufficient.

Also favoring the conclusion that the spontaneous fission is experienced by the americium isotope is the absence of the effect in experiments on  $\alpha$ -particle bombardment of  $\text{U}^{238}$ . The absence of the effect in deuteron bombardment of  $\text{Pu}^{239}$  indicates that we are apparently dealing with  $\text{Am}^{242,241}$  in the isomer state. The period of spontaneous fission of  $\text{Am}^{241}$  in the ground state is  $\sim 2 \times 10^{14} \text{ years}$ <sup>[8]</sup>, while the period of spontaneous fission of  $\text{Am}^{242}$  has not been measured, but in accordance with the systematics of<sup>[9]</sup> it does not differ strongly from that of  $\text{Am}^{241}$ . Thus, the probability of spontaneous fission of the isomer is increased at least  $10^{19}$  times even if other decay channels are taken into account. There are no experiments so far on the excitation energy that leads to such an increase in the probability of spontaneous fission. Estimates based on the expression for

the penetrability of the potential barrier in fission [<sup>10</sup>] yield a value  $\sim 2.5$  MeV.

The angular momentum of the nucleus produced when a 16-MeV deuteron coalesces with the plutonium nucleus apparently does not exceed  $(10-12)\hbar$ . It is possible, of course, for the angular momentum to increase as a result of the  $\gamma$ -ray cascade which precedes the formation of the isomer state, but the spin of this isomer apparently does not exceed  $16\hbar$ .

In conclusion the authors consider it their pleasant duty to thank the staff of the Cyclotron Laboratory of the I. V. Kurchatov Institute of Atomic Energy, headed by S. P. Kalinin and N. I. Venikov, for faultless operation of the accelerator and for continuous help with the work.

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