

URANIUM AND THORIUM FISSION INDUCED BY SUB-BARRIER DEUTERONS

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Submitted to JETP editor November 12, 1962

J. Exptl. Theoret. Phys. (U.S.S.R.) 44, 1445-1449 (May, 1963)

The fission cross sections of Th^{232} , U^{238} , U^{235} , and U^{233} bombarded with 5.8–6.6 MeV deuterons are measured with a semiconductor detector. For 6.6-MeV deuterons the cross sections are respectively 1.5×10^{-28} , 1.6×10^{-28} , 7.5×10^{-28} , and 12×10^{-28} cm² with $\pm 10\%$ accuracy. An investigation of the fragment kinetic energy distributions and an analysis of the fission cross sections indicate that Th^{232} and U^{238} undergo fission mainly following deuteron capture, but that at least 70% of the U^{235} and U^{233} fission events are preceded by stripping.

INTRODUCTION

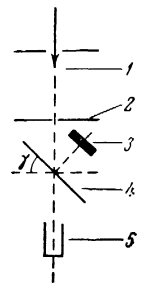
WHEN certain aspects of nuclear fission are investigated it is useful to induce fission by means of accelerated deuterons, thus producing nuclei having a high excitation energy that varies smoothly over a broad energy range. It then also becomes possible to investigate the fission of nuclei having a nucleonic composition that is practically unobtainable when the available substances are bombarded with neutrons. However, deuteron bombardment is associated with a certain ambiguity; because of its small binding energy and relatively large size a deuteron can either fuse completely with a nucleus or it can split and transfer only a single nucleon to the nucleus. In the case of U^{238} fission induced by deuterons having energy approximately equal to the Coulomb barrier or higher, the ambiguity of the reaction has been investigated experimentally by Sugihara et al.^[1] and by Nicholson and Halpern.^[2]

In the present work we determined the absolute cross sections for the fission of U^{233} , U^{235} , U^{238} , and Th^{232} induced by 5.8–6.6-MeV deuterons, and ascertained the mechanism of the sub-barrier interaction resulting in the fission of the given nuclei.

EXPERIMENTAL TECHNIQUE

The experimental arrangement is shown schematically in Fig. 1. A target consisting of the fissionable material distributed uniformly on a thin backing was bombarded with a 5×2 -mm collimated beam. Fission fragments were registered with a surface-barrier semiconductor detector of 25 mm² working area, made of n-type silicon having resistivity of the order 150 Ω -cm; the amplitude resolution of the detector was 2% in recording the

FIG. 1. Experimental arrangement. 1 – deuteron beam direction, 2 – diaphragm, 3 – semiconductor detector, 4 – target bearing fissionable material, 5 – Faraday cylinder.



spectrum of Am^{241} α particles. Measurements were performed with a small (4 V) bias on the detector, so that only the fragment ranges would fit within the registering layer and scattered deuterons would lose a minimal fraction of their energy. The geometric efficiency of fission fragment registration was found to be 4% on the basis of the efficiency for counting α particles from the portion of the target penetrated by deuterons. In calculating fission cross sections no correction was introduced for angular anisotropy in the fragment distribution, because according to results given in^[3] 6–7-MeV deuterons should induce relatively small anisotropy. The deuteron current at the target was of the order 0.1 μA . The total deuteron flux during the entire exposure time was determined with sufficiently high accuracy by means of a current integrator.^[4] The target backing was aluminum foil 5 μ thick. A uniform layer of fissionable material was deposited by vacuum evaporation of the low-melting compounds UF_4 and ThF_4 .^[5] The U^{233} , U^{235} , U^{238} , and Th^{232} layer thicknesses were 6, 176, 240, and 248 $\mu\text{g}/\text{cm}^2$, respectively. Simultaneously with the registration of fission events, pulses were fed to a 128-channel AMA-3 pulse-height analyzer for the determination of the fragment energy spectra. The cyclotron-accelerated deuteron energy was determined to within 0.1 MeV. Aluminum foils of suitable

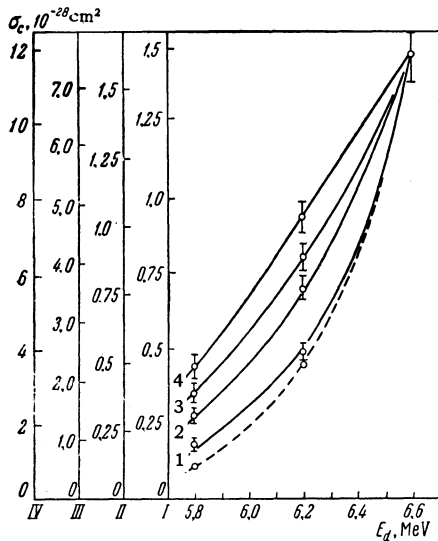


FIG. 2. Fission cross sections as functions of the deuteron energy E_d . The continuous experimental curves pertain to: 1 - Th^{232} , 2 - U^{238} , 3 - U^{235} , 4 - U^{233} . The dashed curve represents the calculated cross section for deuteron capture by thorium^[7] with $R_n = 1.5 \times A^{1/3} \times 10^{-13}$ cm normalized at the point $E_d = 6.6$ MeV, where $\sigma_c = 2.4 \times 10^{-28}$ cm².

thicknesses were used to slow the deuterons to the energies 6.2 and 5.8 MeV. The fissions induced by the background neutrons did not exceed 20%.

RESULTS AND DISCUSSION

The experimental fission cross sections are shown in Fig. 2. The total cross section for the fission of heavy nuclei by deuterons can be represented as the sum of the fissions corresponding to the complete capture of deuterons and those corresponding to stripping (with neutron capture). In the present case the deuteron energy is considerably below the Coulomb barrier, so that the contribution of the (d, n) reaction can be neglected; therefore

$$\sigma_f = \sigma_c P_{d,i} + \sigma_{d,p} P(E_x \geq B_f) P_{d,pf}, \quad (1)$$

where σ_c and $\sigma_{d,p}$ are the cross sections for deuteron capture and for the (d, p) reaction; $P_{d,f}$ and $P_{d,pf}$ are the fission probabilities of excited nuclei depending on the width ratio Γ_n/Γ_f of neutron emission and fission; and $P(E_x \geq B_f)$ is the probability that nuclear excitation following a (d, p) reaction lies above the fission threshold B_f .

The small deuteron energy range did not permit any appreciable variation of the width ratio Γ_n/Γ_f , since the excitation of the compound nucleus varied relatively little but at the same time was sufficiently large to exclude resonance effects. Thus the varia-

tion of the cross section σ_f with deuteron energy was determined mainly by σ_c and $\sigma_{d,p}$ with $P(E_x \geq B_f)$.

Depending upon whether the interaction between deuterons and target nuclei involved capture or stripping, nuclei having different excitation levels are formed. In the case of heavy nuclei the capture of 6.6-MeV deuterons results in the excitation

$$E_x = E_d + \epsilon_n + \epsilon_p - \epsilon_d = 15 - 16 \text{ MeV}, \quad (2)$$

whereas stripping results in the excitation

$$E_x = E_d + \epsilon_n - E_p - \epsilon_d \leq 8 \text{ MeV}, \quad (3)$$

where ϵ_n and ϵ_p are the neutron and proton binding energies in the nucleus, ϵ_d is the deuteron binding energy, and E_p is the kinetic energy of a proton from stripping.

A nucleus formed as the result of stripping can split only if its excitation, given by (3), is above the fission threshold. The proton energy is therefore subject to the limit

$$E_p \leq E_d + \epsilon_n - \epsilon_d - B_f. \quad (4)$$

As a result of differences in the fission thresholds^[6] and neutron binding energies, for $E_d = 6.6$ MeV the maximum energies of protons from stripping for Th^{232} , U^{238} , U^{235} , and U^{233} vary considerably, being 2.8, 2.9, 4.8, and 5.1 MeV, respectively. E_p is dependent on the distance between the proton and the nucleus, i.e., on the required deuteron stretching at the instant of stripping. An approximate calculation, performed by integrating the probability of deuteron stretching by an undistorted nuclear field, from that corresponding to $E_{p \max}$ up to infinity, indicates that the relative probability of fission following stripping is many times smaller for Th^{232} and U^{233} than for U^{235} and U^{238} . For this reason the reactions involved in deuteron-induced fission of these nuclei can differ greatly.

The contributions of the two possible fission-inducing reactions can be evaluated on the basis of the difference between the excitation energies (2) and (3). The peak-to-valley ratios in the fission-fragment kinetic energy spectra must differ considerably. We recorded the energy spectra of fragments from fission induced by 6.6-MeV deuterons, thermal neutrons, and neutrons from the reaction $\text{Be}^9(d, n)\text{B}^{10}$. The experimental peak-to-valley ratios for light fragments are given in the accompanying table. Neutrons emitted as a result of the deuteron bombardment of a thick beryllium target exhibit a continuous spectrum. In the bombardment of Th^{232} and U^{238} , which have a neutron-energy fission threshold of the order

Peak-to-valley ratio of the light fragment energy in the fission fragment spectra for different excitation energies.

Nucleus	Bombarding particles		
	Neutrons		Deuterons
	Thermal	From the reaction $\text{Be}^9(d,n)\text{B}^{10}$	
Th^{232}	—	7.3	4.7
U^{238}	—	3.7	2.7
U^{235}	7.3	4.2	4.9

1 MeV, the mean excitation is intermediate between the excitations associated with stripping and deuteron capture. The relatively high valleys in the Th^{232} and U^{238} spectra thus indicate fission following deuteron capture. Although the U^{235} target was shielded by cadmium, a large number of slow epithermal neutrons would lower considerably the mean excitation of U^{235} bombarded with beryllium neutrons. The agreement of the energy spectra for fission by deuterons and neutrons indicates that U^{235} bombarded with deuterons undergoes fission with lower excitation than Th^{232} and U^{238} ; this means that stripping makes a large contribution.

In Fig. 2 we compare the experimental cross sections σ_f with the energy dependence of the deuteron-capture cross section calculated for a sharply bounded nuclear model. The calculated curve for Th^{232} is based on [7]. All points corresponding to 6.6-MeV deuterons are superimposed to permit more convenient comparison. Figure 2 shows that the cross sections of the different nuclei decreased differently with diminishing E_d . The stripping reaction, which is important for U^{233} and U^{235} , can occur with greater distances between the nuclear center and deuteron charge than in the case of capture; therefore with decreasing deuteron energy the fission cross section associated with stripping must decrease more slowly. Different degrees of nuclear deformation can also have some effect on the steepness of the curve.

More definite conclusions regarding the character of the reaction leading to fission can be reached by comparing the absolute fission cross sections for a given deuteron energy such as 6.6 MeV. The cross sections for the formation of compound nuclei from Th^{232} and U^{238} must be very close in magnitude; the ratio of the fission cross sections must depend mainly on the competition between neutron emission and fission (Γ_n/Γ_f). According to the data in [8], the ratios Γ_n/Γ_f for Th^{233} and U^{239} differ by a factor 4, and for Pa^{234} and Np^{240} by about 1.5. These values

were used to calculate the fission probabilities taking account of the possibility of fission following neutron emission. The experimental fission cross section ratio is in good agreement with the value corresponding to the capture reaction. On the other hand, if U^{235} fission should also occur only after deuteron capture its cross section σ_f in accordance with a different value of Γ_n/Γ_f would differ from the U^{238} cross section by a factor of not more than 1.2. The experimental cross sections differ by a factor 5; this indicates that at least 70% of the U^{235} fission events occur following stripping reactions. Following neutron capture the fissionability of U^{233} is 1.2 times greater than that of U^{235} ; this agrees with the experimental cross section ratio when we consider that the maximum possible energy of protons from stripping is greater for U^{233} than for U^{235} .

The fragment-energy spectra, the dependence of the cross section σ_f on deuteron energy, and the comparison of absolute fission cross sections furnish evidence of differences in the reaction mechanism resulting in fission by 6.6-MeV deuterons. While the fission of Th^{232} or U^{238} is induced practically entirely by deuteron capture, the fission of U^{235} or U^{233} is preceded mainly by stripping. With increasing deuteron energy this diversity must diminish because the difference of the Coulomb barriers and of the maximum proton energies following stripping will become less important.

The authors are indebted to S. A. Karamyan for assistance.

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