Anomalous Behavior of the Proton-Induced Fission Cross Sections of ^{235}U and ^{238}U at Extreme Sub-barrier Energies

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(Received 30 April 1986; revised manuscript received 28 January 1987)

The proton-induced fission cross sections for ^{235}U and ^{238}U targets have been measured for $E_p = 1-4$ MeV for the first time. Compared with the measurements at higher energies, the present results indicate a pronounced change in the slope of the excitation function below 4 MeV such that standard optical-model estimates fall far short of the observed cross sections. No plausible explanation in terms of fission of either the target or the compound nucleus can be found.

PACS numbers: 25.85.Ge

In recent years considerable attention has been focused on the behavior of heavy-ion reaction cross sections in the sub-barrier region where significant enhancement over predictions which use a static barrier has been observed.[1] On the other hand, for very light projectiles on heavy targets, such an enhancement is not expected. Experimental measurements of the proton-induced fission cross sections of ^{235}U and ^{238}U in the sub-barrier region published so far do lie within the limits set by standard optical-model calculations.[2-5] To investigate this aspect at still lower proton energies, we have used a technique which enables measurement of very low (subnanobarn) fission cross sections.

As shown schematically in Fig. 1, a uranium target, bombarded by a collimated proton beam from a Van de Graaff accelerator, was fronted by an annular Lexan polycarbonnate detector of thickness 120 μm (labeled A). This solid-state track recorder (SSTR) registered back-angle fission fragments from proton-induced fission as well as background fission events (i.e., neutron-induced fission, spontaneous fission, etc.) from the target. The geometrical detection efficiency of the detector was about 35%. The beam was stopped in 2-mm-thick aluminum, immediately after which, a second uranium target faced another SSTR (B) in an identical geometry. Therefore fission events seen by SSTR B were expected to give a reasonably accurate measure of the background fission events in SSTR A. The assembly also served as a Faraday cup to measure the total number of protons seen by the target. A negative potential was applied on the collimator to suppress ionization electrons. The assembly was cooled by contact with a copper cold finger dipped in liquid nitrogen. The cooling was required to prevent heating of the SSTR's and a possible annealing of fission tracks. For the same reason it was necessary to limit the beam current during the course of the experiment to 1 μA.

Both of the SSTR's were etched by a standard procedure[6] and scanned under a microscope. The fission tracks, through their characteristic length, shape, and dip angle, could be easily distinguished from any other surface irregularities. It was this ability of the SSTR to record fission tracks distinctly without being affected by the presence of a very high rate of incidence ($> 10^8$ sec^{-1}) of scattered protons which enabled measurement of fission cross sections at the subnanobarn level.

The results of these measurements are detailed in Table I. In every case the background rate of the proton target as measured by SSTR B is found to be very small compared to the total fission rate measured by SSTR A.

Possible sources of background contribution to the fission counts in SSTR A are as follow: (a) Fission caused in the target by neutrons. (b) Fissionlike tracks produced in SSTR A because of the high rate of incidence of scattered protons. (c) Fission caused in fissionable impurities located in the vicinity of SSTR A

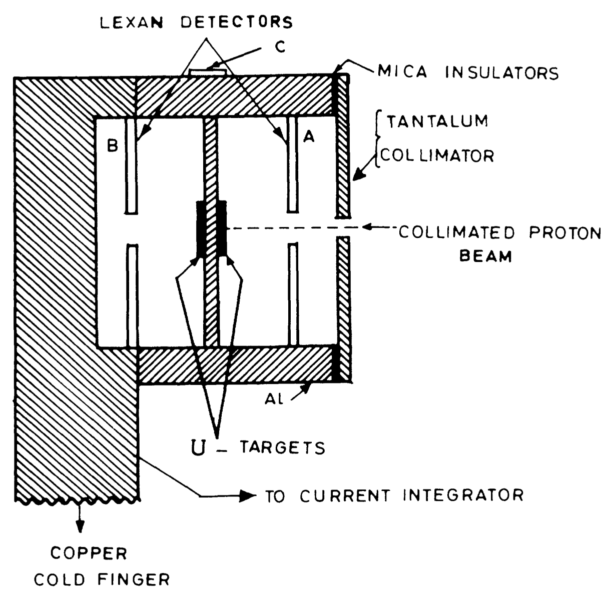

FIG. 1. Schematic diagram of the experimental arrangement for proton-induced–fission cross-section measurements.

TABLE I. Numerical data obtained in the experiment from which the proton-induced–fission cross sections (σ_f) were calculated. E_p is the beam energy, N_A and N_B are the numbers of fission tracks counted in detectors A and B, respectively, and N_p is the total number of protons seen by the target. The numbers in parentheses are the counts estimated from fission caused by neutrons produced through (p,n) reactions in impurities in the aluminum stopper.

Target[a]	E_p (MeV)	N_A	N_B	N_p (10^{16})	σ_f[b] (nb)
^{235}U	0.5	0 (0)	0 (0)	30.0	...
	1.0	7 (0)	0 (0)	6.2	0.3
	1.5	22 (0)	0 (0)	6.2	0.9
	2.0	55 (3)	4 (2)	6.5	1.8
	2.5	79 (18)	4 (13)	6.4	2.7
	3.0	215 (35)	19 (26)	6.3	7.0
	3.5	201 (26)	18 (19)	3.1	13.2
	4.0	1171 (78)	103 (62)	6.2	38.7
Masked	2.5	3	2	5.0	...
^{238}U	0.5	0 (0)	0 (0)	17.8	...
	1.0	13 (0)	0 (0)	5.9	0.5
	1.5	35 (0)	1 (0)	7.7	1.0
	2.0	25 (0)	6 (0)	8.4	0.3
	2.5	23 (0)	3 (0)	10.5	0.3
	3.0	54 (2)	5 (2)	5.0	2.0
	3.5	32 (10)	7 (7)	3.7	1.1
	4.0	369 (28)	16 (24)	5.0	24.7
	4.3	596 (27)	18 (20)	2.8	72.3
^{209}Bi	2.5	0	12[c]	4.9	...
^{232}Th	2.0	1	0	5.8	...
	3.0	2	0	6.5	...

[a]The ^{238}U targets were of natural uranium while ^{235}U targets had 94% enrichment. Thickness of target facing SSTR A was about 460 $\mu g\ cm^{-2}$ for ^{235}U, 380 $\mu g\ cm^{-2}$ for ^{238}U, and 680 $\mu g\ cm^{-2}$ for ^{232}Th. Thickness of target facing SSTR B was about 420 $\mu g\ cm^{-2}$ for ^{235}U, 250 $\mu g\ cm^{-2}$ for ^{238}U, and 280 $\mu g\ cm^{-2}$ for ^{232}Th. Thickness of ^{209}Bi was about 2.5 mg cm^{-2}. All targets were 1 cm in diameter.

[b]Fission cross sections were calculated from total fissions, N_p, and target thickness. Total fissions is N_A corrected for fission background (obtained through N_B) and detection solid angle.

[c]This count is for background ^{238}U target facing SSTR B.

by neutrons or through spontaneous fission. (d) Spontaneous fission in the target and the natural background of fission tracks in the detector. We shall now discuss how contributions from these various sources of background were assessed.

Fission events may be induced in the targets by neutrons from (p,n) reactions in the trace-level impurities encountered by the proton beam either upstream or in the aluminum stopper. The background at a position well removed from the area where the beam stopped was measured by our locating a target-detector sandwich at position C indicated in Fig. 1. The track count here was found to be near zero for both ^{235}U and ^{238}U, an indication that the source of background was the region traversed by the beam in the aluminum stopper. To estimate the number of fission tracks recorded in the detectors from these neutrons, it was necessary to have a quantitative estimate of various impurities in the stopper. Impurity levels of elements with $Z > 21$ could be estimated by use of the x-ray fluorescence technique. The significant elements were Fe, Mn, Cu, and Zn with concentrations of 250, 20, 10, and 40 ppm, respectively. By means of spectroscopic techniques impurity levels of very light elements were also determined. Lithium, which has a very high (p,n) cross section, was found to be present at the 5-ppm level. As can be seen from Table I the counts in SSTR B are well accounted for by the calculations which take into account the (p,n) cross sections of the various impurities, the energy and angular distribution of the neutrons, and the (n,f) cross sections of ^{235}U and ^{238}U. The calculated background contribution in SSTR A from neutron-induced fission is very small relative to the observed number of tracks in every case.

To determine whether scattered protons incident on the SSTR A could form tracks similar to fission tracks, an irradiation was carried out with a bismuth target facing SSTR A and the background ^{238}U target facing SSTR B. No fission tracks were observed in SSTR A (Table I) although the total number of incident protons was comparable to those used in the uranium runs. The counts in SSTR B indicate the same level of neutron background to be present in this experiment also. Therefore the absence of counts in SSTR A rules out the possibility of uranium impurities in the material close to the detectors being responsible for any of the detected fission tracks in the uranium runs either because of neutron-induced fission or because of spontaneous fission.

To determine the contribution from spontaneous fission and the natural background of fission tracks in the detector, SSTR samples were exposed to the targets in the experimental setup without the beam on for several hours and counted after processing. No fission tracks were observed in any of them.

It was noted during scanning that most of the tracks in SSTR A were oriented in a radial manner, indicating that they arose from the beam spot on the target. As an experimental check on the ratio of the contributions in the two detectors due to fissions not directly associated with the incident protons, irradiations were carried out after masking of the central portion of the targets (fraction of area masked $\approx 25\%$) with aluminum foil of thickness 6.7 mg cm^{-2}. This mask stopped fission fragments arising from the beam-spot area and allowed fission events from the unexposed portions of the targets to be detected. Analysis of the results, after taking into account target strengths, showed that background fission rates in the two targets are similar and very small compared to the fission rate in the proton target in the normal (unmasked) runs.

In Table I we also give the results of a few low-energy fission measurements done with ^{232}Th targets. Although these represent new measurements, they were done more in the spirit of doing a control for the uranium-target measurements. The cross sections obtained for ^{232}Th are far below those obtained for the uranium targets in the same energy region although the background-producing aspects are very similar in the two measurements.

The data of the uranium runs were analyzed by our ascribing all background counts to fission caused by neutrons produced in the aluminum stopper. The relevant data for these calculations are given in Table I. The present results for ^{235}U and ^{238}U, together with those of others[4,5] at higher energies, are shown in Fig. 2. The errors shown were obtained by our compounding the statistical error with an estimated 20% error due to uncertainties in detection efficiency, number of incident protons, and target strength. The present measurements at 4 MeV agree with the extrapolated trends of the measurements of Boyce *et al.*[5] and Kononov, Poletaev, and D'yachenko.[4] However, for lower energies there is a pronounced change in the trend of the excitation function so that the measured cross sections vary rather slowly with energy. From measurements at 0.5 MeV it appears that below 1 MeV the cross section again falls rapidly. No other measurements of fission cross sections at these energies have been reported.

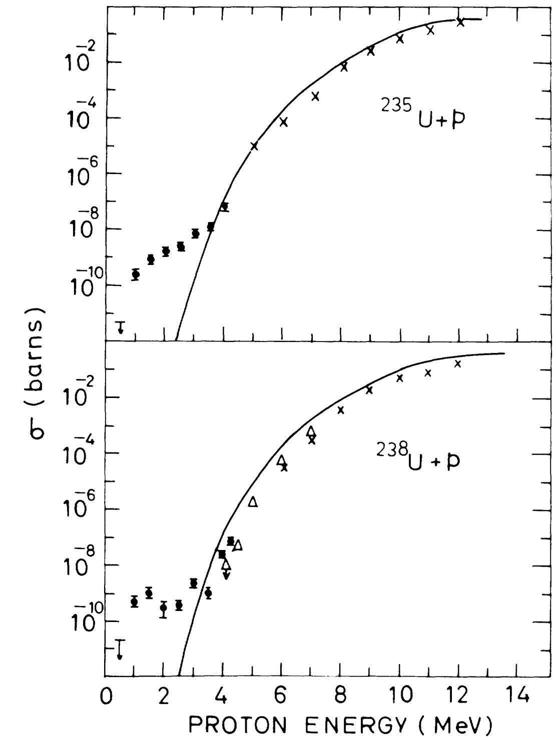

FIG. 2. Proton-induced–fission cross sections for ^{235}U and ^{238}U targets measured by Boyce *et al.* (Ref. 5) (triangles), Kononov, Poletaev, and D'yachenko (Ref. 4) (crosses), and the present authors (points with error bars). The continuous curve represents the reaction cross section calculated from an optical model with parameters given in the text.

For comparison, we have carried out optical-model calculations of the reaction cross section with the following set of parameters[7]: Coulomb radius parameter = 1.3 fm, imaginary potential strength = 5.0 MeV, radius parameter of imaginary potential = 1.355 fm, diffuseness parameter of imaginary potential = 0.635 fm, real potential strength = 50 MeV, radius parameter of real potential = 1.355 fm, and diffuseness parameter of real potential = 0.635 fm. It is clear that the calculated values cannot account for the observed large change in slope below 4 MeV for both ^{235}U and ^{238}U targets. Calculations were also performed with (a) surface-peaked imaginary potential, (b) energy-dependent potentials, and (c) variations of the shape parameters within prescribed limits.[8,9] None of these variations could account for the large observed cross sections in the region below 4 MeV.

The classical distance of closest approach is so large (about 132 fm for $E_p = 1$ MeV) that only Coulomb excitation of very low-lying levels in the target is likely to take place. It is observed that the slope of the measured cross section below 4 MeV is in qualitative agreement with the slope of the Coulomb excitation function for low-lying levels of the target nucleus.[10] However, these states will be faced by a formidable fission barrier and will not have the penetration probability required to explain the observed cross sections. Moreover, an estimate of the cross section for target-state fission from an extrapolation of the low-energy photofission data[11] of ^{238}U gives values which are orders of magnitude lower than the experimentally observed cross sections.

If the observed fission cross section below 4 MeV is due to fission following compound-nucleus formation, the process would have to be seen as a resonant capture of the incident proton. Because of the absence of similar structure in the excitation function at higher energies, the data do not support a picture of compound-nuclear resonance. The possibility of some sort of shape resonance is difficult to visualize in the present system at the extreme sub-barrier energies involved. It is also worth noting here that, unlike the uranium case, the measured low-energy fission cross sections for ^{232}Th are in line with their optical-model estimates.

The present observation of unexpectedly large proton-induced-fission cross sections for ^{235}U and ^{238}U at extreme sub-barrier energies has no ready explanation and therefore is of special theoretical interest. More insight can be gained if measurements of the cross sections for fission and other reaction channels in the extreme sub-barrier region for a range of heavy targets and light projectiles are systematically carried out. Some of this work has already been initiated by us.

We wish to thank Dr. S. S. Kapoor and Dr. M. A. Eswaran for useful discussions and Mr. M. Lal for carrying out the elemental analysis of impurities.

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