JULY 1991

PHYSICAL REVIEW C NUCLEAR PHYSICS

THIRD SERIES, VOLUME 44, NUMBER 1

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Proton-induced fission of ²³⁸U at extreme sub-barrier energies

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(Received 14 January 1991)

Cross sections for proton-induced fission of 238 U have been measured at seven proton energies ranging from 3.0 to 4.45 MeV using a kinematic coincidence technique, and an upper limit established at an energy of 2.5 MeV. Contrary to recent findings of Ajitanand *et al.* [Phys. Rev. Lett. 58, 1520 (1987)], the present work indicates that the fission cross section decreases as expected at extreme subbarrier energies down to a level of ≈ 20 pb at 3 MeV.

Ajitanand et al. [1,2] have recently reported unexpectedly large cross sections for proton and α -induced fission of uranium targets, measured using a nuclear-track detector technique for identifying fission fragments. By this method, they have found that the fission cross section persists at a level of 0.1-1.0 nb down to beam energies of only a few MeV. In view of these results, we have undertaken to measure one of the reactions $(p + {}^{238}\text{U})$ using a different experimental technique, in which both fission fragments are detected in kinematic coincidence. Based on these measurements, we find that the fission cross section behaves as expected from simple barrier penetration and optical-model calculations such as those of Ref. [1]. These results are clearly at variance with the earlier measurements, which report a cross section 100 times larger at even lower proton energies.

Measuring these extremely small cross sections in a reasonable amount of time requires relatively large beam currents (300-900 nA) and, therefore, detectors which are almost totally insensitive to large numbers ($\approx 10^{7}/$ sec) of elastically scattered protons. Ajitanand *et al.* solved this problem by using Lexan polycarbonate track detectors [1]. These detectors are insensitive to protons and α particles but they do not provide any timing infor-

mation that would help to discriminate against background events.

We decided to carry out a real time experiment using two 20×20 cm² parallel-grid avalanche counters (PGAC's) [3] to detect both fission fragments in kinematic coincidence. In a prior test, these detectors were found to be insensitive to high proton rates when operated with 2 Torr isobutane gas as in the present experiment. The PGAC's provided timing information (the time resolution between the counters was typically 500 ps), x and y positions (with a resolution of 3 mm), and specific ionization. Binary fission events were identified by requiring three conditions: time coincidence, back-to-back emission of the fragments in the cm system, and anode signals consistent with strongly ionizing particles.

The two counters were used to detect coincident fission fragments produced in a 260 μ g/cm² U₃O₈ target which was bombarded by a collimated beam of protons from the Argonne Physics Division's 4.5 MV Dynamitron Accelerator. The PGAC's were mounted at 90° to the beam direction at a distance of 11.5 cm from the target, each subtending a solid angle of 1.8 sr. A silicon monitor detector was mounted 39 cm from the target at an angle of 165° with respect to the beam direction, in order to

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measure elastically scattered protons for normalization purposes. This detector subtended a solid angle of 5.4 $\times 10^{-5}$ sr. The beam current, which was between 300 and 900 nA in all runs, was monitored in a Faraday cup located 70 cm behind the target. The duration of the runs ranged from 2 h at 4.45 MeV to 14 h at 3 MeV. The beam was collimated by a set of 4-jaw slits located 110 cm in front of the target. A 350 $\mu g/cm^2 UF_4$ foil was mounted 2.5 cm above the target. This foil was simultaneously monitored, using the PGAC's, in order to determine the background resulting from neutrons produced in (p,n) reactions in the beamline and slits.

Spectra of the time difference $\Delta t = t_2 - t_1$ between the anode signals from the counters are shown on the lefthand side of Fig. 1 for events which satisfy the requirement of large anode signals. Events within a 25-ns window, indicated in Fig. 1 by dashed lines, are considered to be coincidences. Calculated mass distributions are shown on the right-hand side of Fig. 1 for coincidences which satisfy the additional requirement of back-to-back emission in the cm system. The mass spectra all exhibit the double-humped structure characteristic of actinide fission. This structure is also evident in the time spectra.

The background at long and short times in some of the runs is attributed to a known, weak contamination of 252 Cf on the walls of the scattering chamber. In these



FIG. 1. The measured difference in arrival time $(\Delta t = t_2 - t_1)$ at the two detectors is shown on the left-hand side, for events satisfying the requirement of large anode signals. Events falling within the time window indicated by the dashed lines are considered coincidences and accepted for further analysis. Derived fragment mass spectra are shown on the right-hand side for coincidences satisfying the additional requirement of back-to-back emission (see Fig. 2).

events, it is one fragment traversing both detectors which creates the apparent coincidence. The ^{252}Cf background is particularly noticeable at long times in the 4-MeV data due to the absence of an aluminum plate on the back of one of the detectors. In other runs the background counts correspond to fragments that passed through viewing holes in the aluminum plates behind either detector. In all cases, the ^{252}Cf events can be clearly separated from fission events in the target.

The mass distributions in Fig. 1 were derived from the measured time difference $\Delta t = t_2 - t_1$ by assuming that the fission occurred at rest in the laboratory frame and that the sum of the velocities of the fragments was a constant,

$$v_1 + v_2 \simeq v_s = 2.4 \text{ cm/ns}$$
. (1)

The latter had been found empirically to be a reasonable approximation, nearly independent of the individual masses [4]. These assumptions lead to a quadratic expression for the velocities,

$$v_1^2 + \left(\frac{s_1 + s_2}{\Delta t} - v_s\right)v_1 - \frac{v_s s_1}{\Delta t} = 0,$$
 (2)

where s_1 and s_2 are the distances from the target to the point of impact of the fragment on the detector. The fragment mass is obtained from conservation of momentum in the cm system, i.e.,

$$m_1 = \left(\frac{v_s - v_1}{v_s}\right) m_{\text{tot}} , \qquad (3)$$

where m_{tot} is the total mass of the p+U system. The effect of the center-of-mass motion is negligible and is ignored in the calculation. From the timing and position resolution of the detectors, this procedure is estimated to give a mass resolution of 3-4 u.

The back-to-back emission of the fragments was checked by calculating the quantities $(x_1+x_2)/2$ and $(y_1+y_2)/2$, where (x_1,y_1) and (x_2,y_2) refer to the points of detection in the two counters, the x direction in both being defined along the beam and the y direction along the vertical. Histograms of these average positions are shown in Fig. 2 for events which satisfy the coincidence and anode signal requirements. From back-to-back events, one expects to see an average position corresponding to the location of the beam spot on the target with some spreading due to the combined effects of the initial momentum of the fissioning system, neutron evaporation from the fragments, multiple scattering in the target, and the position resolution of the detectors. A well-defined average position is evident at the higher energies, and this allowed a window $(2.6 \times 2.4 \text{ cm}^2)$ to be placed as illustrated to define valid fission events. The number of events satisfying all three requirements (time coincidence, backto-back emission, and large anode signals) at each incident proton energy is listed in Table I.

The setup used in the present experiment provided for an efficient method of measuring the background contribution from neutron-induced fission. A secondary UF₄ foil was placed 2.5 cm above the target being bombarded by the proton beam. Although it was not exactly centered with respect to the detectors, neutron-induced fission

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FIG. 2. The average positions of detection in the two counters are shown at the different beam energies for events satisfying the requirements of time coincidence and large anode signals. Events falling within the indicated rectangular window are considered to exhibit back-to-back emission and accepted as true fission events.

events emerging from the foil would still be detected in kinematic coincidence with an efficiency of $\approx 80\%$. A window identical to those in Fig. 2 was placed around the location of the UF₄ foil. Only two events were observed to fall within this window. Both of these events occurred in the 4-MeV data and are visible in Fig. 2 as the points with the largest y coordinates. From their location, it seems more probable that these two events are actually associated with the primary target. As a result, the neutron back-

TABLE I. Fission counts and cross sections for $p + {}^{238}U$.

Energy (MeV)	N _{fis} (cts)	$\sigma_{\rm fis}$ (b)
2.50	0	$< 1.2 \times 10^{-10} a$
3.00	1	$(2.3 \pm 5.8) \times 10^{-11}$ a
3.25	5	$(1.9 \pm 0.9) \times 10^{-10}$
3.50	23	$(1.5 \pm 0.3) \times 10^{-09}$
3.75	22	$(4.1 \pm 0.9) \times 10^{-09}$
4.00	215	$(215 \pm 0.2) \times 10^{-08}$
4.25	108	$(6.9 \pm 0.7) \times 10^{-08}$
4.45	367	$(1.9 \pm 0.1) \times 10^{-07}$

^aThese uncertainties correspond to a 68%-confidence level according to the prescription of Schmidt *et al.* [7]. ground was neglected in calculating the cross section for proton-induced fission.

As indicated in Table I, only 1 fission event was observed at an incident-proton energy of 3.0 MeV. Because of the over determination of the properties of the event, i.e., collinearity time difference Δt and energy losses in the two detectors, the confidence level for this being a true fission event is estimated to be $\geq 90\%$. This is calculated from the probability that the event in the position window is accidental.

The cross sections were calculated assuming an isotropic angular distribution for the fission fragments. This is expected to be a reasonable assumption based on the data of Boyce et al. [5], who find an anisotropy of 5% at 10-MeV incident proton energies. The present data were also analyzed to produce angular distributions at each energy. All of these were found to be consistent with the assumption of isotropy to within statistical uncertainties. The resulting fission cross sections are listed for all incident energies in Table I and shown in Fig. 3 along with the data of Boyce et al. [5], Kononov et al. [6], and Ajitanand et al. [1]. At 2.5-MeV incident energy, no valid fission events were observed and hence the listed value represents an upper limit on the cross section at this energy. The uncertainties in all cases reflect statistical uncertainties in the number of fissions observed compounded with an estimated 3% uncertainty in the solid angle subtended by the PGAC's. The uncertainties at the lowest energies were calculated according to the prescription of Schmidt et al. [7] and represent a 68%-confidence level.



FIG. 3. The fission cross sections measured in the present work (solid circles) are compared to the results of Ajitanand *et al.* [1] (open circles), Boyce *et al.* [5] (plusses), and Kononov *et al.* [6] (open squares). The solid drawn curve represents the optical model estimate published in Ref. [1].

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The present measurements are in good agreement with all of the previous data at energies above 4 MeV, and with the data of Ajitanand *et al.* [1] at energies of 4 and 3.5 MeV. Below 3.5 MeV, the present measurements exhibit an exponential decline with decreasing proton energy, following the behavior expected from barrier penetration. We do not observe the enhancement seen in the data of Ref. [1]. The one event seen at 3 MeV indicates a cross section several orders of magnitude below that seen in the previous measurement. The fact that no events were seen at 2.5 MeV indicates an upper limit on the cross section a factor of 3 lower than the previous measurement.

In conclusion, we find no evidence of an enhancement in the proton-induced fission cross section for 238 U at the en-

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ergies measured. The data indicate an exponential decline with decreasing proton energy, in agreement with the expectation of barrier penetration and consistent with the optical model calculation of Ajitanand *et al.* [1].

The authors would like to thank the crew of the Dynamitron Accelerator for providing the proton beam for these measurements. This work was supported by the U.S. Department of Energy, Nuclear Physics Division, under Contract No. W-31-109-ENG-38 and by the Laboratory Graduate Thesis Program administered by the Argonne Division of Educational Programs with funding from the U.S. Department of Energy.

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