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Alpha-induced fission of ²³⁵U at extreme sub-barrier energies

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The alpha-induced fission cross section for 235 U has been measured for $E_{\alpha}=3$, 4, and 12 MeV for the first time. Taken together with the already published data for protons at extreme subbarrier energies, these results suggest that the observed cross section below 5 MeV alpha energy may be due to fission of low-lying target states populated by Coulomb excitation. However, it is not clear how such states have the required penetrability of the fission barrier. An explanation of these results may lead to a better understanding of deep sub-barrier tunneling, especially with regard to the fission process.

We have recently published data on proton-induced fission of 235 U in the deep sub-barrier region, i.e., below 4.5 MeV.¹ It showed that the fission cross section does not fall off exponentially with decreasing energy as would be expected from an extrapolation of the higher-energy measurements but changes slope markedly around 4 MeV to give a shelflike behavior. As this was an inclusive measurement, the following mechanisms were experimentally indistinguishable:

Process I: (a) Proton penetrates Coulomb barrier, (b) compound nucleus (CN) is formed, (c) CN undergoes fission without hindrance as it has an excitation energy above its fission barrier.

Process II: (a) Proton scatters inelastically, (b) lowlying target states are populated by Coulomb excitation, (c) these states fission by penetration of the fission barrier.

It is clear that the cross sections observed below 4 MeV far exceed the limits set by optical model calculations of the reaction cross section. There is also no obvious way in which the change of slope of the data points in this region can be accounted for by process I. We, therefore, turn to process II. The distance of closest approach at the experimental energies is so large (≈ 130 fm for $E_p = 1$ MeV) that only Coulomb excitation of very low-lying states in the target is likely to take place. If the observed cross section is due to fission of these excited states, it should be possible to measure similar cross sections with low-energy alpha particles. The Q values for compound nucleus formation with protons and alphas are:

System	Q (MeV)		
$p + {}^{235}U$	+4.8		
$\alpha + {}^{235}U$	-5.2		

It is particularly interesting to carry out experiments in the region $E_{\alpha} < 5$ MeV where compound nucleus formation (process I) is energetically disallowed. We have carried out measurements at $E_{\alpha} = 3$ and 4 MeV with the 5.5 MeV van de Graaf accelerator at Bombay and at $E_{\alpha} = 12$, 12.2, and 27 MeV using the Variable Energy Cyclotron at Calcutta. In the Van de Graaf experiments particle- and energy-analyzed beams were used while in the cyclotron experiments the 12 and 23.3 MeV points were obtained by degrading an unanalyzed 27-MeV beam with gold degrader foils of appropriate thickness placed 1-cm upstream of the target. The incident alpha-particle energy spread full width at half maximum was estimated to be less than 2 MeV for the 12-MeV experiment.

The experimental method, shown schematically in Fig. 1, was the same as the one employed to study low-energy proton-induced fission.¹ The incident beam traversed a thin uranium target (target A) and stopped in the backing of 0.25 mm thickness. An annular plastic (lexan polycarbonate) track recorder (detector A) placed 3 mm upstream of the target detected fission events in target A with an efficiency of 35%. A second target (target B) was sandwiched back to back with the first target and faced another annular track detector (detector B) in a geometry similar to the first target-detector pair. The assembly housing the detectors and targets was cooled by contact with a copper cold finger dipped in liquid nitrogen.

The idea of the second target-detector pair was to obtain a reliable estimate of the experimental background which, in general, can be made up of the following types of events:

(i) Fissionlike tracks produced in the detector because



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

of backscattered alpha particles.

(ii) Spontaneous fission in the target and the natural background of fission tracks in the detector.

(iii) Fission caused in the target by beam-generated neutrons.

We now discuss how contributions from these various sources were assessed. To determine whether scattered projectiles incident on the detector could form tracks similar to fission tracks, an irradiation was carried out with a bismuth target. No fission tracks were observed in the detector although target currents and irradiation times were comparable to those used in uranium target runs.

To determine the contributions from spontaneous fission and natural background of fission tracks, the detector was exposed to the target without beam for a length of time comparable to the beam-on runs. No fission tracks were observed in this case either.

Fission events may be induced in the targets by neutrons from (α, n) reactions in low Z impurities encountered by the beam either upstream or in the target and backing. Target B, being close to target A, sees a similar flux and, therefore, events seen in detector B can serve as a reliable measure of the neutron-induced background in target A. For distant sources the neutron intensities on the two targets are similar. For neutrons produced in the target and backing, due to proximity effects, the neutron intensity on target A can be greater than on target B. If R_n is the ratio of background fission rates in the two targets, the alpha-particle-induced fissions detected in detector A is $(N_A - R_n N_B)$ where N_A and N_B are the fission events recorded in detectors A and B, respectively. It is necessary to make a reliable estimate of R_n to obtain the alpha-particle-induced cross sections from the measured data. This is done as follows.

With an aluminum-backed target the (α, n) channel in the bulk material is open at $E_{\alpha} = 4.3$ MeV, and events recorded in both detectors will be largely due to neutroninduced fission. The value of R_n can, therefore, be obtained from this run, i.e., $R_n = N_A/N_B$. This value of R_n (after a minor correction to take into account the different target thicknesses) can also be applied to the 3 and 4 MeV runs done with gold-backed targets, where neutrons are produced through (α, n) processes in light-element impurities in the target and its backing.

For the 27-MeV run, neutron production is mainly in the backing of target A but the large laboratory anisotropy of neutron emission will reduce R_n with respect to low energies. The 23.3 and 12 MeV runs were carried out by degrading the 27-MeV beam. The major portion of neutron production for these cases is from the degrader which is kept 1 cm upstream of target A. Because the two targets are only 0.5 mm apart, they see essentially the same neutron intensity from this source, i.e., R_n = ratio of target thicknesses. For the neutrons produced in target A and its backing, the large anisotropy of neutron emission gives a considerably larger neutron intensity on target B than on target A. Calculations reveal that a reasonable upper estimate of R_n for the 12-, 23.3-, and 27-MeV runs is just the ratio of target thicknesses. The events recorded in the detectors, R_n values used, and alpha-induced fission cross sections obtained in various runs are shown in Table I.

TABLE I. Numerical data obtained in the experiment from which the alpha-induced ²³⁵U fission cross section (σ_f) were calculated. E_a is the incident alpha energy, N_A is the number of fission tracks in the detector A facing projectile incident target, N_B is the corresponding number for detector B facing background target, and N_a is the total number of target-incident alphas. R_n is the estimated ratio of background fission rates in the two targets.

Beam ^a stops in	Ea (MeV)	NA	N _B	Ν _α (10 ¹⁶)	R _n	σ _f ^b (nb)		
Al	4.3	871	352	4.4	2.5			
Au	3.0	120	9	6.1	3.1	3.2		
Au	4.0	499	50	6,7	3.1	10.8		
Áu	12.0	629	247	1.3 <i>E</i> -2	1.3	5.0E3		
Au	23.0	7656	252	2.4 <i>E</i> -5	1.3	6.5E7		
Au	27.0	38923	856	1.5 <i>E</i> -5	1.3	5.4 <i>E</i> 8		

^aAll targets had 94% enrichment of ²³⁵U. Thicknesses of target facing detector A was 524 μ gm cm⁻² for the gold-backed targets and 420 μ gm cm⁻² for the aluminum-backed target. Thickness of target facing detector B was 398 μ gm cm⁻² in all cases and was aluminum backed. Diameter of the targets was 1 cm. All backings were 0.25 mm thick.

^bFission cross sections were calculated from alpha-induced fissions, N_{α} , and target thickness. Alpha-induced fissions are equal to $(N_A - R_n N_B)/\epsilon$, where ϵ is fission detection efficiency.

The cross sections are plotted in Fig. 2 together with the values obtained by other authors^{2,3} and optical model calculations of the reaction cross sections.

The measurements at 23.3 and 27 MeV are in agreement with other reported measurements while the 12-MeV point agrees with (within errors) the limit set by the calculated reaction cross section. However, the data for 3 and 4 MeV are in the subthreshold region for compound



FIG. 2. Alpha-induced fission cross sections for 235 U target measured by Colby, Shoaf, and Cobble (Ref. 2) (circles), Vandenbosch *et al.* (Ref. 3) (squares), and present authors (points with error bars showing statistical uncertainties only). Continuous curve represents the reaction cross section calculated from an optical model.

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nucleus formation, and are comparable to the protoninduced fission cross sections of 235 U measured in the deep sub-barrier region.¹ They may be due to fission of lowlying states populated by Coulomb excitation. As shown in Fig. 3, the slope of the measured cross section in the low-energy region is in qualitative agreement with the slope of the Coulomb excitation function calculated for a low-lying level of the target nucleus for both protons and alphas.⁴

The fission lifetime of the concerned states can be estimated by using the relation

 $\sigma(\alpha, f) = \sigma(\operatorname{coulex}) \tau(\gamma) / [\tau(\operatorname{fiss}) + \tau(\gamma)].$

Putting $\sigma(a, f) \approx nb$, $\tau(\gamma) \approx nsec$, $\sigma(coulex) \approx mb$, we obtain $\tau(fiss) \approx msec$. However, such small fission lifetimes of low-lying states are not consistent with the formidable fission barrier faced by them. For instance, the fission lifetime of the ground state is $\approx 10^{17}$ yrs.

One can consider the possibility that these states couple to collective levels in a speculative "scission well" and fission with a greater probability than the ground state. An estimate of this enhancement can be made by calculating the penetration of double humped barriers.⁵ This yields a maximum reduction factor of 10^{5} in the half-life whereas a reduction of the order of 10^{27} is required to be consistent with the results. Similarly, fission following a direct reaction such as pick up or stripping is extremely unlikely since the alpha particle is a tightly bound system. Fission of the target nucleus induced by the Coulombic field during interaction is also difficult to invoke for the alpha + ²³⁵U system at such low energies though such a process has been investigated in heavy-ion reactions.⁶

In conclusion, the unexpectedly large cross section for 235 U fission by alpha particles of energy 3 and 4 MeV tak-



FIG. 3. Calculated Coulomb excitation function $(E_2 \text{ only})$ of the 103-KeV state in ²³⁵U by alpha particles (dashed lines) and protons (continuous lines). Also shown are the measured fission cross sections induced by protons (points) and alphas (crosses).

en together with the results for protons at deep sub-barrier energies, seems to indicate fission of low-lying Coulomb excited target states. However, it is not clear how these states fission since their conventional fissionability is far below the observed value. These results, therefore, signal the emergence of some basic and unexpected behavior of the nucleus which may be of fundamental theoretical significance to deep sub-barrier tunneling in general and fission in particular.

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