Fission cross section and fragment angular distribution in gold fission induced by 55 MeV alpha particles using solid state nuclear track detectors

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Abstract. The angular distribution of fission fragments in alpha induced fission has been studied at an incident energy of 55 MeV in ¹⁹⁷Au target. The relative differential fission cross sections were measured at different angles between 10° and 170° and the resulting angular distributions fitted by least squares method with Legendre polynomials. In the present work, a correction for the self-scattering and self-absorption of fission fragments in the target itself was applied and a target of 3 mg/cm² was used to get good statistics. The anisotropy for 55 MeV alpha induced fission of gold was $2\cdot83 \pm 0.43$ and the fission cross section calculated by integrating the measured angular distributions over all the solid angles was $5\cdot2 \pm 1.0$ mb, confirming the value of $4\cdot0 \pm 0.05$ mb reported by Burnett *et al* but contrary to the high value of 10 ± 3 mb reported by Ralarosy *et al*.

Keywords. Angular distribution; fission cross section; anisotropy; alpha particle; nuclear track detector.

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1. Introduction

In 1955, Bohr offered an explanation for the anisotropy in the angular distribution of fission fragments which had first been observed by Winhold *et al* (1952). The interpretation of the observed angular distributions and the important inferences drawn from them, are all primarily based on a model proposed by Bohr. The underlying idea of the model is that the stretched fissioning nucleus, in passing over the saddle point exhibits quantum states similar to those observed in the permanently deformed nuclei, except that the states of the saddle point nucleus are expected to be quasistationary since the nucleus spends a very small time at the saddle point.

A number of studies in angular distribution have been undertaken by many using a variety of targets and projectiles over wide energy ranges (Vandenbosch and Huizenga 1973). Most of these measurements involve experimental set-ups with fission chambers and solid state counters coupled with elaborate electronic circuitry and occasionally radiochemical techniques (Cohen *et al* 1954). Now, solid state nuclear track detectors (SSNTDs) are used for such measurements, especially for fission studies involving target nuclides of relatively low Z and (compared t — ie actinides) with low value of fission cross sections.

Previous measurements of fission cross sections carried out by different authors (Burnett et al 1964; Ralarosy et al 1973), showed that wide discrepancies existed in the

alpha induced fission cross section of gold by a factor of 2. The results of Ralarosy also indicated a large error (30%) in the fission cross section measured at 55 MeV by the well-known sandwich technique using SSNTDs. Since a very thin target (160 μ g/cm²) had to be used in this technique, the counting value was very poor and hence the large error. The aim of the present work is to measure the fission cross section at 55 MeV using a different method and with a thick target of gold (3 mg/cm^2) to secure good value. An appropriate correction was applied for self-absorption effects in the target (Jain 1990). According to this correction, the track density measured is related to the mass m, of the fissile material by $T = Cme^{-\mu m}$. C and μ are constants, of which, C is the track density for unit mass for an infinitely thin target $(m \rightarrow 0)$ while $e^{-\mu m}$ represents the deviation of the track density (T) vs. mass m from a linear relation to an exponential one. The angular distribution of the fission fragments was studied at fixed interval between 10° and 170° using lexan plastics and the fission cross section was obtained by integrating the observed differential cross sections. Incidentally from the study of the anisotropy of angular distribution, some inferences were drawn on the relative orbital angular momentum shared by the fission fragments.

2. Experimental details

2.1 General

The energetic alpha particles were obtained from the Variable Energy Cyclotron (VEC) at Calcutta, India. The collimating system restricts the diameter of the beam at the target to less than 2 mm. The beam current on the target was of the order of 50 nA. The total number of alpha particles striking the target was measured with a Faraday cup (FC) equipped with a secondary electron suppression device. The connections of FC were brought out and fed to an integrator.

Figure 1 shows the target and detector assembly used in the bombardment of the fissile targets. The target (gold) placed at the centre of the cylindrical tube was struck by a collimated beam from the cyclotron. The beam direction was perpendicular to the cylinder axis. This system was kept in vacuum in the scattering chamber (Baliga and Bhattacharya 1986). The fragments emitted from the target embedded themselves in



EXPOSURE APPARATUS

Figure 1. A schematic drawing of experimental arrangement for measuring fission fragment angular distributions.

Fission cross section...

foils (Lexan) were arranged around the cylinder. The detector foils (Lexan) were arranged at various angles $(10^{\circ}, 20^{\circ}...170^{\circ})$. The area over which fission tracks registered in each foil was $1.65 \text{ cm} \times 0.2 \text{ cm} = 0.33 \text{ cm}^2$, while the Lexan foils were located at a distance of 4.9 cm from the fissionable target. Consequently the uncertainty in angular position of a detector is less than $\pm 0.5^{\circ}$. The etching process of the Lexan foils was done (Singh *et al* 1990).

2.2 Target preparation

Target of gold was prepared by evaporating in vacuum the natural element in VEC target 1ab, using gold of 100% purity. A self-supporting gold foil of thickness 3 mg/cm² was made.

2.3 Experimental observation

The fission fragment track densities were measured for gold target for angles between 10° and 170° in the laboratory system. Data collected for fissioning nucleus were converted to centre-of-mass coordinates assuming (1) full momentum transfer of the incident charged particle to the compound nucleus, (2) equal kinetic energy for all fission fragments and (3) symmetric fragment mass distribution. These three conditions are more or less generally satisfied in charged particle induced fission of heavy elements at medium energies as those employed in the present investigation. The kinetic energy release in the centre-of-mass system was estimated from the relation (Terrell 1959).

$$E_{\kappa} = 0.121 \ Z^2 / A^{1/3} \, \text{MeV}$$

where E_K represents the average total kinetic energy of the fission fragments before neutron emission and Z and A are the atomic and mass number respectively, of the compound nucleus. The analysis of the data essentially consists of two parts: 1) Least square fitting of the centre-of-mass angular distributions by a series of Legendre polynomials, to draw inferences about the relative orbital angular momentum of the fission fragments and 2) measurement of the total fission cross section by integrating the differential cross sections. The result is then compared with previous data to resolve discrepancy.

The most serious difficulty in obtaining angular distributions to the desired accuracy concerns with the self-scattering and self-absorption of fission target itself, specially for those fragments emitted at larger angles with respect to the target normal. The overall result of this effect is to depress the differential cross section near 90° and thereby to enhance the apparent symmetry of angular distribution. In the present work, the correction for this effect was found to be appreciable for the gold target of thickness 3 mg/cm^2 (Jain 1990). Figure 2 shows the fission fragment angular distributions for gold target bombarded with 55 MeV alpha particles in the laboratory system (a) without correction and (b) after correction.

The laboratory counting rates and angles were converted to the corresponding centre-of-mass values as mentioned earlier. The resulting angular distributions $W(\theta)$ for the gold target plotted in the centre-of-mass system is shown in figure 3. In figure 3, it can be seen that the points measured in the backward and forward angles describe nearly the same curve showing the basic fore-and-aft symmetry and providing, in a direct way, a justification for the assumptions used in the transformation.



3. Results and discussion

3.1 Fission fragment angular distributions

The measured track densities in the laboratory $W_L(\theta)$ are converted into laboratory differential cross section $(d\sigma/d\Omega)_L$ using the formula

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{L} = \frac{W_{L}(\theta)}{\Omega_{L}\phi N} \tag{2}$$

where Ω_L is the laboratory solid angle subtended by the unit area of the detector over which the track density $W_L(\theta)$ is measured, ϕ is the incident alpha particle flux and N is the number of target nuclei per unit area. The laboratory differential cross sections are converted into centre-of-mass differential cross sections $d\sigma/d\Omega$, using the relevant transformation equations (Jain 1990).

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right) = \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_L \frac{1 + \gamma \cos\theta}{(1 + \gamma^2 + 2\gamma\cos\theta)^{3/2}}.$$
(3)

The symbols are designed in Appendix A.

The relative differential fission cross section $(d\sigma(\theta)/d\Omega)/(d\sigma(90^\circ)/d\Omega)$ or angular anisotropy $W(\theta)/W(90^\circ)$ as a function of angle θ was deduced for gold target (figure 4). A general expression of the type

$$\frac{W(\theta)}{W(90^{\circ})} = 1 + \sum_{l=2, \text{ even}} A_1 [P_1(\cos \theta) - P_1(\cos 90^{\circ})]$$
(4)

was used for fitting the observed anisotropies.

The choice of this particular form, as compared to other equivalent form $W(\theta)/W(90^\circ) = a_0 + \sum_{l=2, \text{ even}} a_1 P_1(\cos \theta)$ is motivated by the fact that the anisotropy $W(\theta)/W/(90^\circ)$, in (4) is normalized to unity for $\theta = 90^\circ$.

A linear, weighted, least square fit analysis was carried out taking into account the experimental errors (shown in Appendix B). The solid line in figure 4 is the best fit to the experimental data obtained using Legendre polynomials with terms up to $P_6(\cos \theta)$ (or A_6) with coefficients as tabulated in table 1. Coefficients higher than A_6 were found to be statistically not significant and hence not included in the table 1.

The dashed line in figure 4 indicate the $(\sin \theta)^{-1}$ variation of the anisotropy as expected from a classical model in which all the angular momentum brought in by the incident particle is delivered to the fission fragments and appears as their relative orbital angular momentum. Mathematically $(\sin \theta)^{-1}$ variation can be expressed in terms of Legendre polynomials as

$$\frac{W(\theta)}{W(90^{\circ})} = \frac{1}{\sin\theta} = 1 + 1.25P_2 + 1.27P_4 + 1.27P_6 + \dots$$
(5)

Comparing these coefficients with the experimentally observed coefficients listed in table 1, one can see that higher angular momentum components in the experimentally observed distributions drop off rapidly as compared to those in $(\sin \theta)^{-1}$. This is an indication that the observed relative orbital angular momentum of the fission



Figure 4. Relative differential fission cross section of gold as a function of the centre-of-mass angle in degrees.

Table 1. Coefficients of Legendre polynomial terms resultingfrom least squares fit of centre-of-mass angular distributions for ^{197}Au .

Target	Energy (Lab)	A ₂	A4	A ₆
¹⁹⁷ Au	55 MeV	1·28 ± 0·09	-0.19 ± 0.10	0·09 <u>+</u> 0·13

fragments is much smaller than the actual angular momentum brought in by the incident particle. The observation from table 1 that the anisotropy of the angular distributions could be fitted by three Legendre polynomials coefficients up to A_6 , is an indication that the relative orbital angular momentum of the fission fragments is the average $3\hbar$, while the average angular momentum brought in by the incident alpha particle is of the order of 20 \hbar . The difference between the two values is dissipated into the formation of high spin states of the fission fragments as well as into collective rotational degrees of freedom such as rolling friction in some cases.

3.2 Fission cross section

The integral cross-section for fission at a given energy of the projectile, σ_f can be determined by integration, over solid angle, of the fission fragment cross section

$$\sigma_f = \int_0^{2\pi} \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right) \mathrm{d}\Omega \tag{6}$$

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Target	Thickness	Energy	Fission cross section	Detector used	References
¹⁹⁷ Au	3 mg/cm ²	55 MeV	$5\cdot2\pm1\cdot2$ mb	SSNTD Lexan	Present
¹⁹⁷ Au	_	55 MeV	$4.0 \pm 0.05 \mathrm{mb}$	SSNTD Mica	Burnett et al (1964)
¹⁹⁷ Au	$163 \mu \mathrm{g/cm^2}$	55 MeV	10-0 ± 3-0 mb	SSNTD Lexan	Ralarosy et al (1973)

Table 2. Fission cross sections for alpha particle induced fission of gold.

$$\sigma_f = \frac{W(90^\circ)}{\phi N\Omega} \int_0^{2\pi} \frac{W(\theta)}{W(90^\circ)} d\Omega$$
$$= \frac{2\pi W(90^\circ)}{\phi N\Omega} \int_0^{\pi} \frac{W(\theta)}{W(90^\circ)} \sin \theta \, d\theta \tag{7}$$

where ϕ is the flux of bombarding particles, N is the number of target per cm², $W(90^{\circ})$ is the number of tracks detected at 90° in the centre-of-mass coordinate system, Ω is the solid angle subtended by the detector, $W(\theta)/W(90^{\circ})$ is the centre-of-mass anisotropy for the particular energy of the charged particle involved, and the integration is made over 2π sr.

The experimentally measured cross section is listed in table 2 for gold target. The error associated with the (α, f) cross section is estimated to be about $\pm 20\%$ for the gold target. The large error, associated with the $^{197}Au(\alpha, f)$ reaction cross section arises primarily due to its target thickness (3 mg/cm^2) . Measurements of some of these fission cross sections have been made by different methods by various authors. The results of these measurements are listed in table 2, for comparison with present data. The present result serves to confirm the experimental result of Burnett *et al* for alpha induced fission of gold. However, while Burnett *et al* have quoted 1 to 2% error in their result, our value has a much smaller error than that reported by Ralarosy *et al*.

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Appendix A

Conversion of laboratory variables into centre-of-mass variables

We consider the fission reaction and define the following symbols: 1 = projectile, 2 = target, 3 = first fission fragment, 4 = second fission fragment, $m_i = \text{mass of the } i\text{th}$ body (i = 1, 2, 3, 4).

 $\mu = m_1 m_2/(m_1 + m_2)$ = reduced mass of the projectile target system; T_i = laboratory kinetic energy of the *i*th body; $Q = T_3 + T_4 - T_1 - T_2$ = the Q value of the reaction; $K = \mu T_i/m_1$ = the kinetic energy of relative motion of the projectile target system in the CM frame. $\gamma = [m_1 m_3 K/m_2 m_4 (K + Q)]^{\frac{1}{2}}$ = an important parameter, and θ_L (or θ) = emission angle of the fragment 3 in the laboratory (or CM) frame. The transformation from angle θ_L to θ for $\gamma < 1$ is affected by

$$\cos\theta = (\cos\theta_L) \left(1 - \gamma^2 \sin^2\theta_L\right)^{1/2} - \gamma \sin^2\theta_L.$$
(A1)

The conversion of the differential cross section from the laboratory frame (suffix L) to the CM frame (no suffix) is done through

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_L \frac{1 + \gamma \cos\theta}{(1 + \gamma^2 + 2\gamma \cos\theta)^{3/2}}.$$
 (A2)

In applying the above formula in the conversion of laboratory observables into centre-of-mass observables, the following approximations are usually made:

(a) Although the fission fragment masses are distributed over a wide range, only the rate of the most abundant pair is considered. (b) In charged particle induced fission, say by alpha particles of 50–60 MeV, the fragment mass distribution is taken to be symmetric.

Appendix B

Procedure for linear, weighted, least square Legendre polynomial fit

In this appendix we describe the minor steps in carrying out a weighted least square fit of the form

$$Y(\theta) \equiv \frac{W(\theta)}{W(90^\circ)} = A_0 + \sum_{l=2}^{L} A_l P_l(\cos\theta), \ l = \text{even}$$
(B1)

to the anisotropy data, normalized accounting to

$$Y(90^{\circ}) \equiv 1 = A_0 + \sum_{l=2}^{L} A_l P_l(0)$$
(B2)

1) First, we define our notations. Let n = number of independent data points, $W(\theta_i) =$ experimental counts at angle $\theta_i (1 \le i \le n)$, $\langle (\delta W(\theta_i))^2 \rangle = W(\theta_i) =$ variance i.e. error squared of this observation, $Y_i = W(\theta_i)/W(90^\circ) =$ experimental value of the anisotropy, $\langle (\delta Y_i)^2 \rangle = (Y_i + Y_i^2)/W(90^\circ) =$ variance of the anisotropy at the *i*th point excluding $\theta = 90^\circ$ angle, and $f_i(\theta_i) = P_i(\cos \theta_i) - P_i(0) =$ a set of useful function defined for $2 \le l \le L$.

2) Next, the weighted chi-square is set up through

$$\chi^{2} = \sum_{i=1}^{n} \frac{1}{\langle (\delta Y_{i})^{2} \rangle} \left[1 + \sum_{i'=2}^{L} A_{i'} f_{i'}(\theta_{i}) - Y_{i} \right]^{2}$$
(B3)

minimization of chi-square with respect to the parameter $A_l (l = 2, 3, ..., L)$ requires

$$\frac{\delta\chi^2}{\delta A_1} \equiv \sum_{i=1}^n \frac{2f_i(\theta_i)}{\langle (\delta Y_i)^2 \rangle} \left[\sum_{i'=2}^L A_{i'} f_{i'}(\theta_i) - (Y_i - 1) \right] = 0.$$
(B4)

This set of simultaneous normal equations is written conveniently in matrix form as

$$[C][A] = [D] \tag{B5}$$

where C is a square matrix, and A, D are column vectors with typical elements denoted by $C_{ll'}$, A_l , D_l such that

$$C_{ll'} = \sum_{i=1}^{n} \frac{f_l(\theta_i) f_{l'}(\theta_i)}{\langle (\delta Y_i)^2 \rangle}, \quad D_l = \sum_{i=1}^{n} \frac{f_l(\theta_i) (Y_i - 1)}{\langle (\delta Y_i)^2 \rangle}$$
(B6)

(iii) Finally, (B5) can be solved by matrix inversion to get the best values of the unknown parameters A_i appearing in (B1)) as

$$[A] = [E][D] \text{ with } [E] \equiv [C]^{-1}, \tag{B7}$$

The elements of the error matrix E also yield the variances and covariance of A_i and $(A_i, A_{i'})$ in the form

$$\langle (\delta A_l)^2 \rangle = E_{ll}; \langle (\delta A_l) (\delta A_{l'}) \rangle = E_{ll'}$$
(B8)

The parameter A_0 (B1) and its variance are computed from

$$A_{0} = 1 - \sum_{l=2}^{L} A_{l} P_{l}(0)$$

$$\langle (\delta A_{0})^{2} \rangle = \sum_{l}^{L} \sum_{l'}^{L} E_{ll'} P_{l}(0) P_{l'}(0).$$
 (B9)

From (B9) we can directly calculate the desired fission cross section σ_f and its statistical form

$$\sigma_f = \alpha \int_0^{\pi} d\theta_y \sin \theta \ Y(\theta_y) = 2\alpha A_0$$

$$\delta \sigma_f = 2\alpha \sqrt{\langle (\delta A_0)^2 \rangle}$$
(B10)

where $\alpha = 2\pi W(90^{\circ})/(\phi N\Omega)$ as described in (7), and θ_{y} is the angle expressed in radians.

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