Emission-angle dependence of fission fragment spin

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The average spin of fission fragment at specific mass asymmetry has been obtained as a function of emission angles in the ^{238}U ($\alpha_{39.1\ \mathrm{MeV}}$,$f$) system from radiochemically determined independent isomeric yield ratios of ^{132}I. The fragment average spin is seen to decrease from $11.2\hbar$ to $6.5\hbar$ with change in emission angle from 90° to 20°, due to the angle-dependent tilting component over and above the statistical wriggling, bending, and twisting components as per the collective mode model. Effects of entrance channel parameters and multichance fission are also discussed.

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I. INTRODUCTION

Angular momentum effects in heavy-ion reactions and fission are topics of recent interest. Several experimental [1–5] and theoretical studies [6,7] show that angular distributions and angular momenta of reaction/fission products depend upon various aspects. These aspects include the initial energy and orbital momentum, fragment mass asymmetry, interplay of various collective rotational degrees, and possibly dynamics. In-depth understanding of angular momentum effects, however, remains far from complete since experimental information is usually averaged over several degrees of freedom. The commonly used techniques [1–4,7] of prompt gamma angular correlation or multiplicity (N_γ) measurements generally provide estimates of fragment mass and/or charge averaged angular momentum, summed over emission angles. Additionally, uncertainty in N_γ measurements arises [1,4,7,8] on account of the statistical gamma rays, gamma multipolarities, and spin carried away by neutrons. A somewhat limited radiochemical method provides estimates of fission fragments spin free from some of the averaging effects in specific instances.

In the present work the average spin (J_{av}) of a specific fragment has been deduced as a function of emission angles from radiochemically determined independent isomeric yield (IIY) ratios of ^{132}I in the ^{238}U ($\alpha_{39.1}$ MeV,f) system. The present and earlier observations on angle-averaged J_{av} of ^{132}I at various excitation energies in the same system [9] have been interpreted in terms of the existing models [6,7,10].

II. EXPERIMENTAL

Electrodeposited U targets (150 μg/cm^2) on 25 μm Al-backing foils were irradiated for appropriate times using 40±0.4 MeV collimated (5 mm), external alpha particle beam at the 88-In. Variable Energy Cyclotron, Calcutta. Typical integral beam current was 2–3 μA h. The targets were kept at 45° inclination with respect to the alpha beam to minimize attenuation of emergent fragments at various angles. The recoiling fragments were collected on 25-μm-thick Al-catcher foils placed in cylindrical geometry at six different positions (angles) covering emergence angles from 90° to 20° with respect to the beam direction. Fission product ^{132}Te and ^{132}I^{m,g} were assayed by off-line gamma spectrometry at a fixed geometry on an efficiency-calibrated 60-cm^3 HPGe detector coupled to a 4K MCA. Resolution of the detector system was 2.0 keV at 1332 keV. The gamma lines followed were 228.2 keV of ^{132}Te and 772.7 keV composite line for ^{132}I^m and ^{132}I^g. From the observed activities as a function of time the IIY ratios of ^{132}I^m and ^{132}I^g were evaluated [5,9] after corrections for precursor (^{132}Te) contribution as follows. In each irradiation at each angle of observation, the area A under the 772 keV photopeak was related [5] to the cumulative yield of the precursor (Y_{Te}) and independent yields of the isomers (Y_m and Y_g) as a function of time as

$$\frac{A}{R} = Y_{\mathrm{Te}}F_P + Y_m F_M + Y_g F_G \ , \tag{1}$$

where F_P, F_M, and F_G are the respective time functions dependent on decay constants, branching fractions, and gamma abundances (for m and g states) apart from irradiation and cooling times. R is a constant including the fission rate and collection efficiency. Y_{Te} was deduced from the ^{132}Te activity itself in the same foil. Thus, in each case after correcting the observed activity A for precursor contribution, Y_m and Y_g were deduced by least-squares analysis to evaluate the independent isomeric yield ratio $[Y = Y_m/(Y_m+Y_g)]$.

The isobaric chain decay data systematics used are

$$
\xrightarrow{\beta^-} {}^{132}\mathrm{Te}(78.2\ \mathrm{h},\ 0+) \xrightarrow{\beta^-}
\begin{array}{c}
{}^{132}\mathrm{I}^m(83.4\ \mathrm{min},\ 8-) \xrightarrow{\beta^-} \\
\downarrow \%\mathrm{IT}\ 87.0 \\
{}^{132}\mathrm{I}^g(2.28\ \mathrm{h},\ 4+) \xrightarrow{\beta^-}
\end{array}
\left| \begin{array}{l} {}^{132}\mathrm{Xe} \\ (\mathrm{stable}) \end{array} \right.
$$

TABLE I. Independent isomeric yield ratios and J_{av} for ^{132}I as a function of emission angles.

$\theta_{c.m.}$ (deg)	$\Delta\theta$ (deg)	$Y_m/(Y_m+Y_g)$	$N(\theta)-I$	$N(\theta)-$Te	J_{av} ($\hbar$)
84.47	10.90	0.81±0.01	0.21±0.01	0.22±0.01	11.2±0.4
64.18	8.98	0.80±0.02	0.26±0.02	0.26±0.01	10.8±0.4
48.95	6.31	0.70±0.04	0.33±0.01	0.32±0.02	8.3±0.8
38.48	4.30	0.62±0.03	0.38±0.03	0.36±0.02	7.2±0.4
31.29	3.44	0.55±0.03	0.40±0.06	0.39±0.03	6.5±0.3
25.70	8.61	0.56±0.02	0.41±0.01	0.42±0.04	6.5±0.3

Table I shows the independent isomeric yield (IIY) ratios at various emission angles. Error limits quoted on the IIY ratios at different angles are essentially the precisions from five sets of measurements. Other sources of error in each single measurement include counting statistics (~5%), least-squares fitting, and detector efficiencies besides negligible errors in decay-scheme data. Error in a single measurement was estimated to be around 10%. The results were checked for any inconsistencies from activity correlations as follows. Table I shows the normalized activity per unit solid angle at each angle or $N(\theta)$ values for ^{132}Te and ^{132}I^{m+g} deduced from individual gamma lines. The closeness of the $N(\theta)$ values for ^{132}Te and total ^{132}I indicates similar regular anisotropy for these isobars as is expected. A further check was made from total ^{132}I/^{132}Te activity ratios at various angles. The observed ^{132}I/^{132}Te ratio of 0.44±0.05 is in reasonable agreement with the expected [12] from charge distribution systematics (0.42) in the present fissioning system.

III. RESULTS

The fragment angular momentum is generally deduced by comparing the probability ratio of the final isomeric spin states evaluated employing a statistical model with the experimentally obtained IIY ratio for appropriately chosen fragment initial J_{av} values. The statistical model calculates spin distributions during and after fragment deexcitation by neutron and cascade gamma (dipole and quadrupole) emission. The code GROGI2 [13] was used to deduce the fragment J_{av}. The code requires fragment average excitation energy (E_{av}), neutron transmission coefficients [$T_1(E_n)$], spin cutoff factors for neutron (σn) and gamma ($\sigma\tau$), experimental level-density parameters, and gamma-emission widths for the concerned isotopes as major inputs. The code takes into account neutron and gamma (dipole and quadrupole) competitive emissions and population of the yrast levels. GROGI2 provides spin distributions at each stage of deexcitation for the chosen fragment J_{av} at a fixed initial excitation energy (E_{av}). The E_{av} value at fragment mass ~134 in the fissioning system ^{242}Pu was estimated to be ~17 MeV on the basis of the usual prescription [14] as

$$E_{av}=[\nu(S_n+E_n)+E_\tau]_{th,A}+(E_{ex}-E_{th})A/A_{FN}\ , \quad (2)$$

with $E_n=1.75+0.65\sqrt{\nu_A}$ and $E_\gamma=1.1\nu_A+1.75$ based on empirical correlations with S_n as neutron separation energy for ^{134}I and ν_A, E_n, and E_γ as neutron multiplicity, neutron energy, and gamma-energy respectively at mass A in thermal fission of the same nucleus ^{242}Pu ($={}^{241}$Pu$+n_{th}$) at excitation energy E_{th}. E_{ex} is the total excitation energy in the present case (^{238}U$+\alpha={}^{242}$Pu with $A_{FN}=242$).

Optical model neutron transmission coefficients of Auerback and Perey were used. The σ values for neutron and gamma emissions were semiempirically calculated for each nuclei at different stages of deexcitation and were found to be typically $(5.0\pm1.5)\hbar$. Errors in J_{av} owing to variations in the E_{av}, $T_1(E_n)$, and σn values are insignificant as spin carried away by neutrons is insignificant. Inputs for gamma emission are largely empirical and the overall error in J_{av} is usually $<\pm0.5\hbar$. The major source of error is actually the scatter in the IIY ratios. Table 1 shows the J_{av} of fragment ^{134}I at six emission angles, based on the code GROGI2. Errors on the J_{av} values are due to the precisional scatter on the IIY ratios.

In medium-energy fission the occurrence of multichance fission (MCF) might influence our observations. Hence MCF contributions from plutonium isotopes 242, 241, 240, and 239 were calculated on the basis of the ratio of the widths for neutron emission and fission (Γ_n/Γ_f) for each nuclide. The ratio according to the statistical model is

$$\frac{\Gamma_n}{\Gamma_f}=\frac{2A^{2/3}}{C}\int_0^E eP(E-S_n-e)de \Big/ \int_0^{E-B}P(E-e)de \quad (3)$$

where A is the mass of the fissioning nucleus, C is a constant (~13.85 MeV), and $P(E)$ are the relevant level densities dependent on S_n, B_f (fission barrier), and the level-density parameter a_n as well as the ratio a_f/a_n as input parameters.

Calculations were carried out for both spin-independent [14] and spin-dependent [7] prescriptions and the results are shown in Table II along with the inputs [15]. In the spin-dependent case spin changes during particle evaporations are considered [7] while the spin-independent ratios were calculated with constant temperature level density as well as with various a_f/a_n ratios. The spin-independent ratios for a constant-temperature level density and $a_f/a_n=1.0$ ratio are seen to be in close agreement with the spin-dependent ratios. It is also seen that fission of ^{242}Pu and ^{240}Pu dominates the total fission width with nearly the same weight. The

TABLE II. Multichance fission evaluation.

Nuclide	^{242}Pu	^{241}Pu	^{240}Pu	^{239}Pu
E (MeV)	33.3	24.9	17.8	9.6
$\langle I \rangle$ ($\hbar$)	14.6	12.5	11.6	11.0
$T>B$ (MeV)	0.98	0.83	0.68	0.38
K_0 ($\hbar^2$)	124.0	103.0	84.0	46.0
%F(const T)	41.7	10.4	41.1	6.8
%F($a_f/a_n=1$)	43.3	9.1	42.9	4.7
%F(spin dependent)	45.3	10.5	43.5	0.7

MCF effect on the angle dependence of J_{av} will be discussed later.

IV. DISCUSSION

Table I shows that in ^{238}U($\alpha_{39.1\ \mathrm{MeV}}$,$f$), J_{av} for a specific fragment (^{134}I) varies from $11.2\hbar$ to $6.5\hbar$ with the change in emission angles from 90° to 20° in contrast to a small change of $\sim 5\%$ for mass-averaged fragments in the ^{238}U($\alpha_{42.8\ \mathrm{MeV}}$,$f$) system as obtained by Schmitt *et al.* [1] Thus the fragment J_{av} changes with emission angles although the magnitude of the change is much higher for a specific (mass-asymmetric) fragment. Schmitt *et al.* [1] showed that the observed emission angle dependence of J_{av} is a manifestation of the tilting mode of rotation characterized by projection (K) of the total spin (I) on the fission axis ($0<K<I$). On the other hand, a strong dependence of J_{av} on the fragment's mass arises through moments of inertia or spin cutoff factors [1,2,6,7]. Dayras *et al.* [4] showed the dependence of the gamma multiplicity on the fragment Z in a heavy-ion reaction and the effects of particle evaporation. Moretto *et al.* [6] also showed that the exit channel angular momentum varies along the mass-asymmetry coordinate even in the case of statistical equilibration.

A. Evaluation of fragment (^{134}I) average spin as a function of emission angles in first change fission

The emission angle-dependent average spin $J_{av}(\theta)$ for asymmetric, spherical fragment (^{134}I) was evaluated in first chance fission (^{242}Pu*) according to the collective mode model (CMM) of Schmitt and Pacheco [10]. We considered a statistically equilibrated configuration involving a K-dependent aligned projection and all the statistical rotational degrees for the calculations. Total equilibration of the rotational degrees was envisaged since even the tilting mode with the largest relaxation time compared to the wriggling, bending, and twisting modes equilibrates in compound nucleus fission [2]. In the present evaluations, the K distribution with the emission angle (θ) was calculated as prescribed by Schmitt and Tirion [16], after summing over the normalized initial total spin (I) distribution, $\sigma(E,I)$,

$$\langle K^2 \rangle = \sum_{I=0}^{\infty} \sigma(E,I) 0.5(I+0.5)^2 + \sin^2\theta[1-I_1(X)/I_0(X)] \ , \qquad (4)$$

$$X=(I+0.5)^2 \sin^2\theta/4K_0^2 \ ,$$

where I_0 and I_1 are the modified Bessel functions of zeroth and first order, respectively; $\sigma(E,I)$ was evaluated using the Hafner code [17] and appropriate transmission coefficients. K_0^2 is the variance of K distribution that governs the change of $\langle K^2 \rangle$ with the emission angle. K_0^2 was calculated using typical above-the-barrier temperature ($T>B$) and moments of inertia for fragments (I_1 and I_2) of spherical shapes as

$$K_0^2=[I_\perp I_\parallel/(I_\perp - I_\parallel)]T/h^2 \qquad (5)$$

for $I_\perp = I_1+I_2+I_r$, and $I_\parallel = I_1+I_2$.

The calculated K_0^2 for the first chance fissioning system ^{242}Pu* is 118.6 at mass 134 for a temperature ($T>B$) of 0.98 MeV and was used in the evaluation of fragment J_{av}. The calculated K_0^2 is also quite close to the reported [18] average and first chance fission K_0^2 values (112 and 119, respectively) based on experimental gross anisotropy in the same fissioning system. The $J_{av}(\theta)$ at mass 134 were calculated after summing up the spin contributions due to various rotational degrees as shown by Moretto and Schmitt [1,6] and then averaging as

$$J_{av} = \langle |J_{tot}| \rangle = \int_{-\infty}^{+\infty} |J| P(J) dJ \ , \qquad (6a)$$

where

$$P(J) = (1/\sqrt{2\pi}\sigma)\exp[-(J-J_{tot})^2/2\sigma^2] \ , \qquad (6b)$$

$$J_{tot} = [\langle J_{tl} \rangle^2 + \langle J_w \rangle^2 + \langle J_b \rangle^2 + \langle J_{tw} \rangle^2)]^{1/2} \ . \qquad (6c)$$

The average spins due to the individual rotational degrees were calculated following CMM [10] as follows. The angle-dependent tilting term ($\langle J_{tl} \rangle$) is given by [10]

$$\langle J_{tl} \rangle = R_I + (\langle Q^2 \rangle / 2R_I)[a_1^2 - (I_1/I_\perp)^2(a_1+a_2)^2] \ , \qquad (7)$$

where R_I is the rigid-rotation component [6,10] and $a_{1,2}$ are the tilting eigenvector components for the spherical fragments [10],

$$R_I = (I_1/I_\perp)(I^2-K^2)^{1/2}, \quad a_1 = I_1[I_\perp/I_r(I_1+I_2)]^{1/2} \ .$$

The angle (θ) dependence arises from the $\langle Q^2 \rangle$ term,

$$\langle Q^2 \rangle = \langle K^2 \rangle_\theta I_r / I_\perp (I_1+I_2) \ . \qquad (8)$$

The wriggling ($\langle J_w \rangle$), bending ($\langle J_b \rangle$), and twisting ($\langle J_{tw} \rangle$) mode contributions are all angle independent since statistical in nature. These contributions were evaluated using the detailed expressions [Eqs. (20) and (33)] of Schmitt and Pacheco [10],

TABLE III. Comparison of experimental and theoretical J_{av} for ^{132}I in the ^{238}U(α,f) system. At constant excitation energy: angle differential for $T>B=0.98$ MeV and $\langle I\rangle=14.6\hbar$.

$\theta_{c.m.}$ (deg)	K^2 ($\hbar^2$)	J_{av} expt. ($\hbar$)	J_{av} ($\hbar$) in $A=121$	^{242}Pu 134	J_{av} ($\hbar$): $K_0^2=65\hbar^2$ 134
84.47	77.5	11.2±0.4	10.5	11.5	10.9
64.18	68.0	10.8±0.4	10.1	10.8	10.4
48.95	52.8	8.3±0.8	9.4	9.9	9.7
38.48	38.5	7.2±0.4	9.1	9.2	9.2
31.29	27.9	6.5±0.3	8.8	8.8	8.8
25.70	19.8	6.5±0.3	8.7	8.5	8.5

$$\langle J_1\rangle y=R_I+X/2-0.5(X+R_I/2)\exp(-R_I/2X)$$
$$+(2\pi T)^{1/2}(a_{1y}/2)(1+R_I/4X)$$
$$\times\text{erfc}[(R_I/2X)^{1/2}]\ , \qquad (9)$$

where $X=a_{1y}^2T/R_I$, $y=w$ for wriggling and b for bending, a_y are the corresponding eigenvectors, and T is the temperature. For the twisting mode, for low values of the rigid-rotation component (R_I) (as in the present case) the contribution is [10]

$$\langle J_1\rangle_{tw}=[(2T/\pi)I_1I_2/(I_1+I_2)]^{1/2}\ . \qquad (10)$$

The width parameter σ in Eq. (6b) is dependent on the statistical modes since the variance for the tilting mode is zero [10]. σ was evaluated using the statistical mode eigenvectors as

$$\sigma^2=[a_{1w}^2+a_{1b}^2+a_{1tw}^2(1-2/\pi)]T\ . \qquad (11)$$

The σ values were 5.92$\hbar$ and 6.62$\hbar$ for the asymmetric and symmetric mass splits.

The calculated fragment J_{av} and their angular variation for mass 134 based on CMM [10] shows a general agreement with the experimental data as shown in Table III. These observations confirm the influence of mass-dependent aligned (K-dependent) spin component due to the tilting mode. Statistical equilibration of the rotational degrees is also apparent. To examine the effect of mass asymmetry further, J_{av} as a function of angle was also calculated at symmetric mass 121 as shown in Table III. The K_0^2 value for the symmetric split in first chance fission was obtained [19,20] as ≈ 124. It was seen that for the symmetric mass (121), variation of J_{av} with emission angle is less pronounced compared to the asymmetric mass 134, due to the higher contribution from the statistical components. The statistical components will be more predominant with deformation.

The effect of mass asymmetry on the angular variation of fragment J_{av} was also indicated by Schmitt *et al.* [1] through the dependence of J_{av} on the fragment's moment of inertia. Back *et al.* [2] also showed the dependence of fragment J_{av} on mass asymmetry. It was further shown by Back *et al.* [2] that the aligned and the statistical components vary with mass asymmetry in an opposite manner, i.e., while the aligned component increases with higher mass asymmetry, the statistical components decrease. In medium-energy fission yields of the symmetric (or deformed region) and asymmetric fragments are comparable. The asymmetric fragments show a stronger dependence on the tilting (aligned) mode while the symmetric fragments show a stronger dependence on the statistical modes. Therefore, N_γ measurements [1] for mass-averaged fragments show only a small variation in J_{av} with emission angle due to (a) mass-averaging effects on the angular dependence of the tilting mode and (b) the mixing of the different nature of dependence of the (angle-dependent) aligned and statistical (angle-independent) spin components on mass asymmetry.

Calculations were also carried out for mass 134 in the ^{238}U(α,f) system at alpha energies 25.2, 27.0, 33.1, 39.1, and 44.2 MeV for angle-averaged fragment J_{av} to compare with the experimental data [9] as given in Table IV. This table also shows a general agreement between the experimental and calculated J_{av} values despite variations in the entrance channel conditions. These observations confirm the influence of the statistical rotational modes with increasing excitation energy. Statistical equilibration of the collective rotational degrees is also apparent from the general agreements between experimental and calculated J_{av} values.

TABLE IV. Comparison of experimental and theoretical J_{av} for ^{132}I in the ^{238}U(α,f) system. At variant excitation energy: angle integral.

E_α (MeV)	$T>B$ (MeV)	$\langle I\rangle$ ($\hbar$)	$\langle K^2\rangle$ ($\hbar^2$)	J_{av} expt. ($\hbar$)	J_{av} in ^{242}Pu ($\hbar$)
25.2	0.71	7.4	13.9	6.9±0.5	6.1
27.0	0.75	8.6	18.0	7.5±0.6	6.7
33.1	0.88	12.0	31.6	9.7±0.8	8.4
39.1	0.98	14.6	43.5	9.7±0.5	9.7
44.2	1.07	16.4	52.8	10.1±0.8	10.8

B. Effects of multichance fission

Angle dependence of fragment spin as well as fragment angular distribution is due to the tilting mode governed by the parameter K_0^2, which in turn is dependent on temperature and shape parameters of the fissioning nuclei. Multichance fission effects are thus expected on both these experimental observables as higher chance fissioning nuclei have lower K_0^2. Mass-asymmetry dependence of angular distribution in fission is observed [21,22] and attributed to chance fission effects. In the present work the variation of $N(\theta)$ (Table I) qualitatively indicates a higher anisotropy for mass 132 compared to the reported [18] gross anisotropy (~ 1.52) in the same fissioning system. The $N(\theta)$ data are not directly usable to deduce the angular anisotropy and K_0^2 around mass 132 as these data are based on relative normalization of the decay-resolved activities in different catcher foils over a limited range of angles. Therefore, separate, detailed experiments on the mass-resolved angular distribution of fission products were carried out as described elsewhere [21,22]. The angular anisotropy for fission products around mass 132 was seen to be ~ 1.78 indicating a K_0^2 value of $65\hbar^2$ in the $^{238}U(\alpha_{39\ MeV},f)$ system.

Evaluation of the fragment (^{134}I) $J_{av}(\theta)$ for the observed K_0^2 value of 65, i.e., in a chance-fission-averaged system was also carried out using an average total spin of the system instead of the initial spin distribution in Eq. (4) and the average temperature of 0.83 MeV. These results are also given in Table III for comparison. Table III shows that the calculated J_{av} values and their variations with emission angles (θ) in both chance-fission-averaged case and in first chance fission are quite comparable to the experimental observations. Therefore in the total fissioning system (chance averaged) calculation with $K_0^2=65\hbar^2$, appropriate for the experimental angular anisotropy, is adequate for fragment J_{av} and its angular variation at mass 134. The experiment shows a slightly higher variation of $J_{av}(\theta)$. The calculations might yield more closely comparable results with the experimental observations if fragment deformation is considered [Eqs. (7) and (8)].

Apart from direct calculations of fragment J_{av} in a chance-fission-averaged system, the effect of higher chance fission on first chance fission results is also visualizable as follows. The deviation of the observed K_0^2 (65) at mass 132 from the calculated value of 118.6 based on first change fission is due to the significant extent (Table II) of third chance fission (^{240}Pu) occurring with a lower total spin ($\sim 11\hbar$), temperature (0.68 MeV), and therefore with a lower K_0^2 value. As a consequence, in third chance fission contributions due to the tilting as well as the statistical components would be leading less to a smaller angular variation of J_{av} as well as lower J_{av} values ($\sim 2\hbar$ to $1.5\hbar$) compared to first chance fission. Further, third chance fission also results in more forward-peaked fragments with lower spin due to low K_0^2 (higher anisotropy). Thus at lower angles, asymmetric fragments with spin less than expected from first chance fission will be observed with higher probability. Therefore for a particular asymmetric fragment, produced from both chance fissions, the spin difference at $\theta=90°$ and 0° will be more than expected from first chance fission only. For the symmetric fragments resulting essentially from more energetic first chance fission due to a higher fission barrier, such chance-fission-induced enhancement in the variation of $J_{av}(\theta)$, is not expected. This aspect is usually confirmed by the near-isotropic angular distribution (high K_0^2 values) [21,22].

To conclude, the present studies show that (a) specific fission fragment spin strongly depends on the angle of emission due to the correlation of the angular variation of the tilting mode with mass asymmetry, (b) collective rotational degrees are in statistical equilibrium, and (c) mass-averaging and multichance fission effects are significant.

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