

## Entrance-Channel Dependence of Fission-Fragment Anisotropies: A Direct Experimental Signature of Fission before Equilibration

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Fission-fragment angular distributions in reactions of  $^{10}\text{B}$ ,  $^{12}\text{C}$ ,  $^{16}\text{O} + ^{232}\text{Th}$  and  $^{237}\text{Np}$ , and  $^{19}\text{F} + ^{237}\text{Np}$  have been measured. While the measured anisotropies in B- and C-induced fission are found to be in agreement with the predictions of the standard Halpern-Strutinsky theory, they are anomalously large in the case of O- and F-induced fission. Such a discontinuous behavior in angular anisotropy with respect to the entrance-channel mass-to-charge asymmetry provides an experimental verification of the predictions of the preequilibrium fission model proposed earlier to explain the anomalous fragment angular distributions.

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It is now well established<sup>1,2</sup> that in several cases of heavy-ion-induced fusion-fission reactions, the fission-fragment angular distributions exhibit much larger anisotropies than predicted by the standard Halpern-Strutinsky theory. Several explanations have been put forward to interpret these anomalous angular distributions. In one approach,<sup>3-5</sup> it has been proposed that the standard saddle-point statistical model<sup>6</sup> (SSPSM) breaks down at high spin and/or large values of  $Z^2/A$  of the compound nucleus (CN), and the angular distribution of fission fragments is governed by an effective transition state different from the fission saddle point. In another approach, anomalous angular distributions are ascribed to the possible emergence of new reaction channels<sup>7</sup> such as quasifission and fast fission, which result in fissionlike fragments without going through well-defined saddle points. In a recent work,<sup>8</sup> anomalous fragment angular distributions observed in sub-barrier fusion reactions have been analyzed on the basis of SSPSM to deduce the value of the mean-square spin  $\langle I^2 \rangle$  of the fissioning nucleus. The analysis gave considerably larger values of  $\langle I^2 \rangle$  than those calculated by standard reaction models, which may imply the presence of reaction mechanisms such as channel coupling at sub-barrier energies.

In an earlier work,<sup>9</sup> we had shown from the analysis of the anomalous fragment angular distributions in heavy-ion-induced fission reactions that, apart from CN fission, there is an additional component of fissionlike events of the composite system which has equilibrated in all degrees of freedom except the  $K$  degree of freedom. Since the variance  $\sigma_K^2$  of the unequilibrated  $K$  distribution of these non-compound-nuclear (NCN) events is small due to the memory of the entrance-channel reaction plane ( $K=0$ ), they give rise to highly anisotropic fragment angular distributions. Consequently, any admixture of such events leads to observed fragment anisotropies larger than that given by the standard theory for CN fission. It was pointed out that these NCN events consist of not only those arising from quasifission or fast fission,

but also from reaction trajectories leading to CN formation but reseparating prematurely in those systems where the fission barrier is comparable to the temperature. It was also suggested<sup>10</sup> that a characteristic signature of fission before full  $K$  equilibration will be an entrance-channel dependence of the fragment anisotropies for target-projectile combinations across the Businaro-Gallone ridge in the mass-to-charge asymmetry degree of freedom. To look for any such entrance-channel dependence of fragment anisotropies, we have carried out measurements of fragment angular distributions in fission induced by boron, carbon, and oxygen ions on thorium and neptunium targets and by fluorine ions on a neptunium target. To avoid possible uncertainties in the calculation of  $\langle I^2 \rangle$  arising from anomalous reaction mechanisms such as channel coupling at sub-barrier energies, the above measurements were carried out at above-barrier energies. The results of the present measurements indicate an entrance-channel dependence of the fragment anisotropies consistent with the expectations of the above "preequilibrium" fission model.

The experiments were performed using the heavy-ion beams from the Bhabha Atomic Research Centre-Tata Institute of Fundamental Research 14UD Pelletron accelerator. The  $^{232}\text{Th}$  and  $^{237}\text{Np}$  targets were about  $300 \mu\text{g}/\text{cm}^2$  thick deposited on aluminium backing. Two silicon-surface-barrier detectors of about  $30 \mu\text{m}$  thickness were used to detect the fission fragments. The elastic- and inelastic-scattering events of the projectile ions were eliminated by taking anticoincidence with veto detectors mounted behind the fission detectors. The angular distribution measurements were carried out over the laboratory angular range of  $80^\circ$ – $170^\circ$ . The relative solid angles of the two detectors were determined by counting the  $\alpha$  particles from the  $^{237}\text{Np}$  target and by taking data in both the detectors at overlapping angles. The absolute fission cross sections were measured by normalization to Rutherford scattering with a monitor detector mounted at a forward angle. The measured

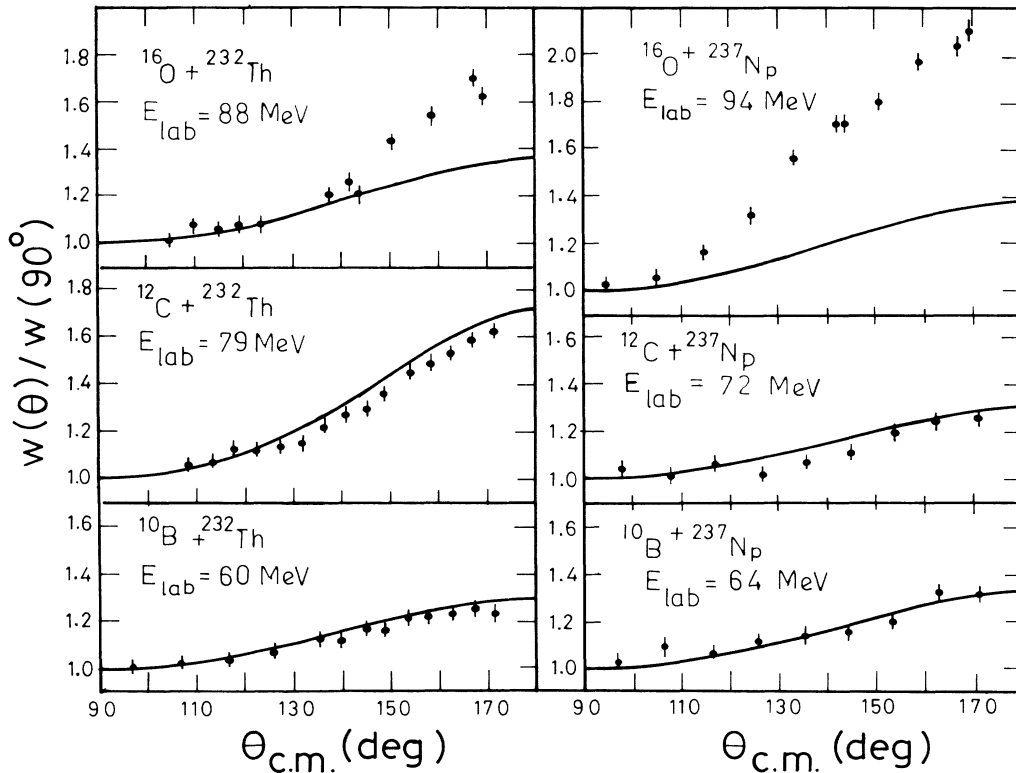


FIG. 1. Measured fragment angular distributions for  $^{10}\text{B}$ ,  $^{12}\text{C}$ ,  $^{16}\text{O} + ^{232}\text{Th}$  and  $^{237}\text{Np}$  systems at a few typical bombarding energies. Results of the SSPSM calculations are shown by the solid lines.

fragment angular distributions were transformed to the center-of-mass system assuming symmetric mass division and using the Viola systematics<sup>11</sup> for total fragment kinetic energy. Figure 1 shows the fragment angular distributions for a few typical cases of target-projectile systems and bombarding energies. Figure 2 shows the experimental anisotropies deduced by fitting Legendre polynomials with the observed angular distributions for all cases measured in the present work along with the available data for the  $^{16}\text{O} + ^{232}\text{Th}$  system.<sup>8,12</sup> The results of calculations based on the SSPSM are also shown in Figs. 1 and 2. The fragment angular distributions were calculated using the effective moment of inertia  $J_{\text{eff}}$  derived from the measured systematics of  $\alpha$ -induced fission anisotropies<sup>13</sup> rather than that calculated from the rotating-liquid-drop model<sup>14</sup> (RLDM). A small correction for the dependence of  $J_{\text{eff}}$  on the spin of the compound nucleus was, however, suitably incorporated. This procedure was adopted to remove uncertainties due to the choice of parameters which enter the RLDM calculations. The spin distributions were calculated in the framework of the models of Esbensen<sup>15</sup> and Wong<sup>16</sup> by adjusting the barrier fluctuation parameters to fit the fusion cross sections measured for the various systems. The agreement between the experimental and calculated fusion cross sections was quite good for all the target-projectile systems as seen from Fig. 3.

It can be seen from Fig. 2 that the experimental anisotropies are in general agreement with the predictions of the SSPSM for the cases of  $^{10}\text{B} + ^{232}\text{Th}$ ,  $^{12}\text{C} + ^{232}\text{Th}$ ,  $^{10}\text{B} + ^{237}\text{Np}$ , and  $^{12}\text{C} + ^{237}\text{Np}$  projectile-target systems. For  $^{16}\text{O} + ^{232}\text{Th}$ ,  $^{16}\text{O} + ^{237}\text{Np}$ , and  $^{19}\text{F} + ^{237}\text{Np}$  systems, the calculated anisotropies are in major disagreement with the experimental data. The values of the mass asymmetry  $\alpha = (A_T - A_P)/(A_T + A_P)$  for the different systems are given in Fig. 2. The above results clearly show that the anomalously large fragment anisotropies arise only for the target-projectile combinations whose mass asymmetry values are smaller than the Businaro-Gallone (BG) critical asymmetry,<sup>17</sup> which is calculated to be about 0.9 for the range of composite nuclei studied in the present work. The anomalous behavior is unlikely due to any compound-nucleus effects, since some of the above target-projectile combinations lead to similar compound nuclei, but give different anisotropies depending on the entrance-channel mass asymmetry. The recent results of Kailas *et al.*<sup>18</sup> for the  $^9\text{Be} + ^{232}\text{Th}$  ( $\alpha = 0.925$ ),  $^9\text{Be} + ^{235}\text{U}$  ( $\alpha = 0.926$ ) systems and those of Zhang *et al.*<sup>19</sup> for the  $^{19}\text{F} + ^{232}\text{Th}$  system ( $\alpha = 0.848$ ) also fall into the same pattern as above with the entrance-channel asymmetries for these systems on either side of the BG value and the measured anisotropies in agreement with the SSPSM calculations in the case of Be-induced fission and anomalous in the case of F-induced fission. It may

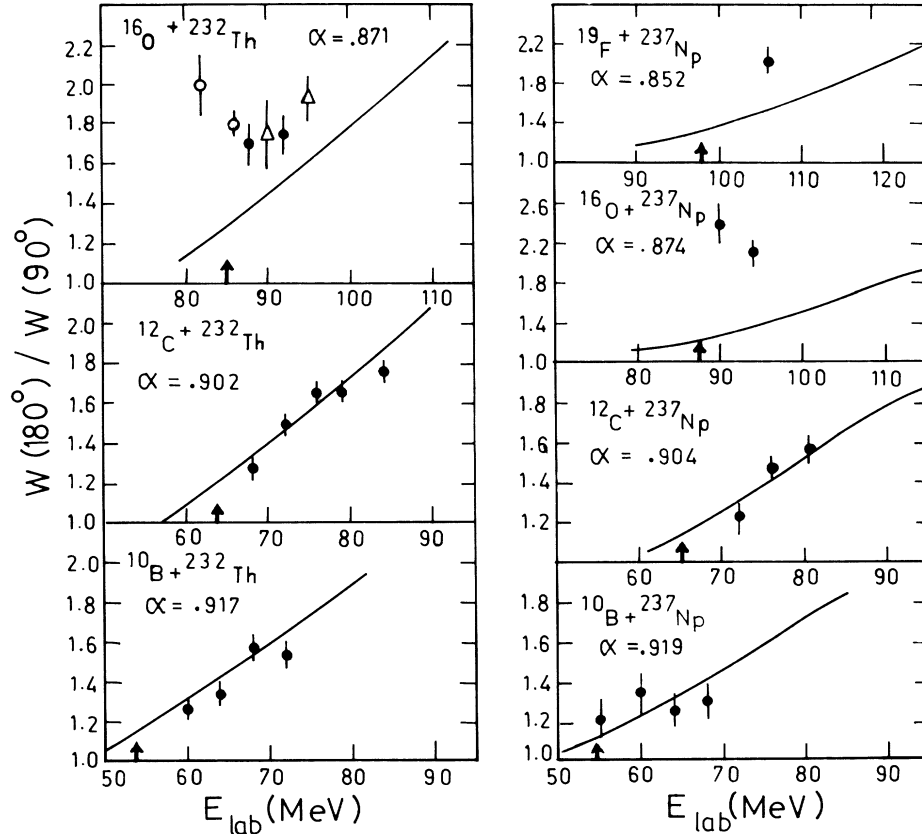


FIG. 2. Experimental and calculated fission-fragment anisotropies as a function of bombarding energy for various target-projectile systems. Open circles are from Ref. 8 and triangles are from Ref. 12. The arrows correspond to the calculated fusion barriers for various systems.

also be pointed out that while anomalous anisotropies have been reported<sup>2</sup> for the  $^{12}\text{C} + ^{236}\text{U}$  ( $\alpha = 0.903$ ) system, these measurements were at sub-barrier energies, where possible reaction mechanisms such as channel coupling may also cause larger than expected  $\langle I^2 \rangle$ , and hence anomalous angular distributions.

The present results of a discontinuous behavior in angular anisotropy with respect to entrance-channel mass-to-charge asymmetry are consistent with the predictions of the preequilibrium fission model and can be understood in the following manner. When the entrance-channel mass asymmetry in a heavy-ion fusion reaction is varied, a qualitative change in the fusion path is expected to take place across a critical mass-to-charge asymmetry called the Businaro-Gallone critical asymmetry ( $A_{\text{crit}}^{\text{BG}}$ ). When the entrance-channel asymmetry is smaller than  $A_{\text{crit}}^{\text{BG}}$ , the dinuclear system, after capture inside the conditional saddle point, relaxes in mass asymmetry and elongation, passes over the unconditional fission saddle point, and moves towards the spherical compound nucleus. Thermal diffusion during this phase can result in a re-separation of the mass-equilibrated fragments over the barrier leading to the occurrence of preequilibrium fission events. On the other hand, if the

entrance-channel mass asymmetry is larger than  $A_{\text{crit}}^{\text{BG}}$ , the system, after capture inside the conditional saddle point, experiences a driving force towards larger asymmetries and smaller elongation, thus leading to the formation of the compound nucleus in a relatively shorter time scale. Even if the system re-separates before the formation of the compound nucleus, the asymmetry of the resultant products will be close to or larger than the entrance-channel asymmetry and not fissionlike. Consequently, in these cases, preequilibrium fission events will not take place. With the systematics deduced in our earlier analysis,<sup>9</sup> the expected fraction of preequilibrium fission in the present systems can be as much as 30% of the CN fission events, for  $B_f/T \sim 2$ . Since the preequilibrium fission events have large anisotropies, an admixture of these events will make the observed anisotropies anomalous as compared to the SSPSM calculations for compound-nuclear fission. The anomalous behavior will be more pronounced at lower bombarding energies, where the CN anisotropies are small, than at higher bombarding energies. While this feature is seen in the  $^{16}\text{O} + ^{232}\text{Th}$  case for which data are available over a large energy range, a quantitative interpretation of this would require further investigations of the effects of oth-

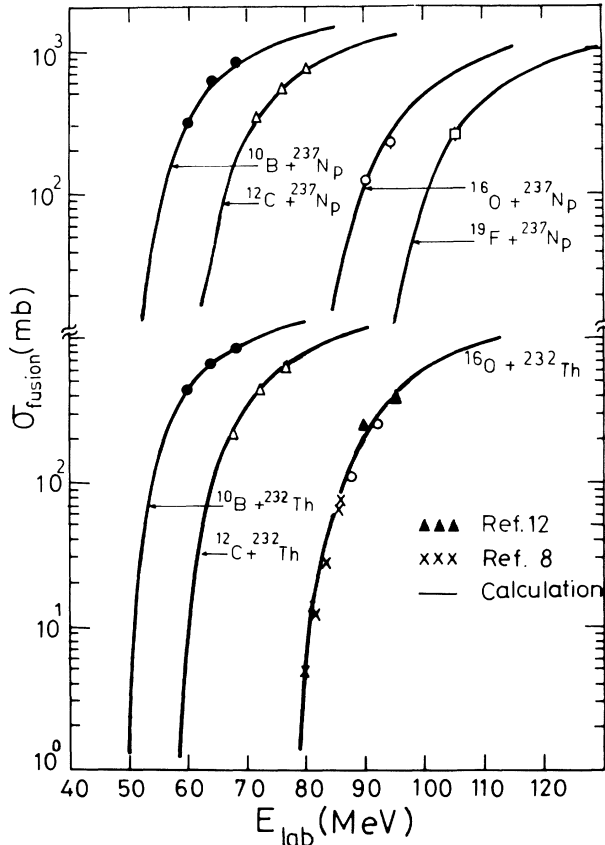


FIG. 3. Comparison between the measured and calculated fusion excitation functions for various target-projectile systems.

er possible mechanisms such as channel coupling and transfer-induced fission.

In conclusion, it is shown that anomalous fragment anisotropies appear only for systems with entrance-channel asymmetries smaller than the BG critical mass asymmetry. The anomalous angular distributions can be understood on the basis that the observed fission events in heavy-ion-induced fission reactions, in general, consist of

a mixture of CN fission and preequilibrium fission events.

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