Evidence of microscopic effects in fragment mass distribution in heavy ion induced fusion-fission reactions

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Abstract

Our measurements of variances ($\sigma_m^2$) in mass distributions of fission fragments from fusion-fission reactions of light projectiles (C, O and F) on deformed thorium targets exhibit a sharp anomalous increase with energy near the Coulomb barrier, in contrast to the smooth variation of $\sigma_m^2$ for the spherical bismuth target. This departure from expectation based on a statistical description is explained in terms of microscopic effects arising from the orientational dependence in the case of deformed thorium targets.

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The formation of a super heavy element through primary fusion of two nuclei is restricted by the subsequent evolution of the compound system dependent on its survival (with at most particle emission) as opposed to its fission. The nuclei must have enough kinetic energy to overcome the repulsive electrostatic energy in order to come within the range of the attractive nuclear forces in a touching configuration. The path the system takes in a complicated multidimensional potential energy landscape [1] governs the fusion of the two nuclei from a touching configuration to a composite system, equilibrated in all macroscopic degrees of freedom. As an example, depending upon the initial conditions of excitation, the entrant system of target and projectile can reach a fusion meadow in the energy landscape, equilibrate to a compound nucleus and cool down after the evaporation of a few particle and photon emission to a evaporation residue (ER), or the super-heavy compound nucleus could choose another path to undergo shape oscillations over an unconditional saddle to reach a fission valley.

The topography of the potential energy surface in the parameter space (involving the deformations of the two touching nuclei, their mass asymmetry, the separation between the two and the nature of the neck joining them) is far too complicated to enable us to determine theoretically the path taken by the system in its evolution. This is even more so, because of possible microscopic effects. Accordingly, it is of paramount importance to use experimental probes together with a phenomenological understanding to elucidate the route actually traversed by the system. Thus the observed angular anisotropy in the fission fragments (ratio of yields parallel and perpendicular to the beam direction), following statistical laws, on one hand and the measured cross sections for production of evaporation residues on the other, are generally taken to indicate that the system equilibrates to compound nucleus in the fusion meadow. However, recent interpretations [2,3] based on measurement by these two probes have led to contradictory conclusion regarding the path taken by the system vis a vis the fusion meadow or the fission valley or for that matter through an entirely different route over an asymmetric saddle. The present authors [4] have proposed that accurate measurements of mass distribution can be used as reliable tool to help pin down the route followed by the system to reach the fission valley. The present letter reports accurate measurements of fission fragment mass distributions as a function of the excitation energy close to the Coulomb barrier in several systems with different projectiles on a deformed as well as a spherical target and the phenomenological explanations of the observed variations of the width of mass distributions for different topographical routes the systems follow through the energy landscape. Our measurements for the first time clearly picks up the microscopic effects in determining the path the systems follow in reaching the fusion meadow or the fission valley.

The experiments were performed with judiciously chosen projectiles of $^{12}$C,
$^{16}$O and $^{19}$F on deformed $^{232}$Th and spherical $^{209}$Bi targets. Large deviations in the fragment anisotropy from the predictions of statistical theory [5] were reported for thorium target [6,7,8], while those for spherical bismuth target followed the statistical predictions [9,10]. For the spherical bismuth target, the entrant system is compact for any orientation and the expected mass flows are from target to projectile in all target-projectile systems [11]. However, the compactness in shape for the entrance channel changes quite appreciably as the impact point of the projectile changes from the equatorial to the polar regions of the prolate thorium nuclei, and the macroscopic effects of mass flow for carbon (projectile to target) is opposite to that of oxygen and fluorine nuclei (target to projectile) reacting with thorium target. In all the cases, the macroscopic effects only predict a smooth variation of the width of the fragment mass distributions with the excitation energies or the temperature of the equilibrated fused system [12]. So any departure of the smooth variation of the width of the mass distributions would be a likely signature of microscopic effects driving the systems through different pathways in the energy landscape.

Pulsed heavy ion beams from the 15UD Pelletron at Nuclear Science Centre (NSC), New Delhi, India, had been used in the experiments. The pulse width was about 0.8-1.5 ns with a pulse separation of 250 ns. The energy of the beams were varied typically in steps of 1-2 MeV, from a few MeV above the Coulomb barrier to a few MeV below it. The targets were either self-supporting $^{232}$Th of thickness 1.8 mg/cm$^2$ or a 500 µg/cm$^2$ thick self-supported $^{209}$Bi. Complementary fission fragments were detected with two large area (24 cm $\times$ 10 cm) X-Y position sensitive multi-wire proportional counters (MWPCs) [13]. The fission fragments were separated from elastic and quasi-elastic channels using time of flight of particles and the energy loss signal in the detectors. Folding angle technique was used to differentiate between fusion-fission (FF) and transfer fission (TF) channels, from a distribution of the events in $\theta - \phi$ correlations or an equivalent procedure of the correlation of the fissioning system velocities parallel and perpendicular ($V_{\text{par}} - V_{\text{perp}}$) to the reaction plane [14]. In Fig. 1, typical separation of fragments from exclusively FF reactions are shown for both procedures. The resulting fragment mass widths differ at most few percent and clearly do not have any impact on the final experimental results or conclusions drawn from it. The masses of the fission fragments were determined event by event from precise measurements of flight paths and flight time differences of complementary fission fragments. The estimated mass resolution for fission fragment was about 3 a.m.u. The details of experimental arrangement and data analysis and elimination of systematic errors were reported in reference [13,15].

The measured mass distributions in earlier reported cases of $^{19}$F, $^{16}$O + $^{232}$Th and $^{16}$O + $^{209}$Bi [4,15] and the presently measured case of $^{12}$C + $^{232}$Th and $^{19}$F + $^{209}$Bi at all energies are well fitted with single Gaussian distributions around the symmetric mass split for the target plus projectile systems. Typical
Fig. 1. Distributions of complementary fission fragments in $\theta$-$\phi$ (upper panel) and $V_{\text{par}}$-$V_{\text{perp}}$ (lower panel). The contour represents the gate used to select the fusion fission events.

Fig. 2. Mass distributions at two excitation energies for the system $^{19}$F + $^{232}$Th. The Gaussian fit are shown by the solid lines.

mass distributions for the system $^{19}$F + $^{232}$Th, at excitation energies of 49.4 MeV and 39.1 MeV, fitted with a Gaussian are shown in Fig. 2. The variation of the variance of the fission fragment mass distribution ($\sigma_m^2$) are shown by solid squares in Fig. 3 for $^{19}$F and $^{16}$O projectiles on the spherical $^{209}$Bi nuclei.

It has been observed that the mass variance ($\sigma_m^2$) shows a smooth variation (trend is shown by solid lines) with the excitation energy of the fused system across the Coulomb barrier. This is in qualitative agreement with the pre-
dictions of statistical theories. It is also noted that no significant departures are reported in the fragment angular anisotropy measurements as shown by the open symbols in the lower halves of the figures (predicted anisotropies from SSPM theory [5] shown by dashed lines) for the spherical target and projectile systems [9,4,10]. Thus for these target-projectile combinations, we conclude that the systems fused to an equilibrated compound nucleus in the fusion meadow for all excitation energies, and subsequently underwent shape changes to reach an unconditional mass symmetric saddle and fission. Predominantly macroscopic forces are assumed to govern the paths taken by the above systems.

![Graph of mass variance vs. excitation energy](image1.png)

**Fig. 3.** Mass variance ($\sigma_m^2$) as a function of excitation energy ($E^*$) for spherical bismuth target. The arrow points to excitation energy corresponding to Coulomb barrier. The solid lines show smooth variation of $\sigma_m^2$ with $E^*$. Reported fragment anisotropy A (open symbols) and SSPM predictions (dashed lines) are shown in lower halves.

The variances of mass distribution ($\sigma_m^2$) for reactions of different projectiles for the present as well as our earlier reports [15,4] on the deformed thorium target are shown in upper panel of Fig. 4 a-c, for $^{19}$F, $^{16}$O, $^{12}$C projectiles, respectively. In all three cases, as the excitation energy is decreased, the $\sigma_m^2$ values, shown by solid squares, decreased monotonically, but shows a sudden upward trend approximately at the Coulomb barrier energies. This is once again followed by a smooth decrease as energy is further lowered. The sudden increase in $\sigma_m^2$ values is most prominent ($\sim 50\%$) in case of $^{19}$F + $^{232}$Th and decreases to $\sim 15\%$ in $^{16}$O + $^{232}$Th and to $\sim 10\%$ in the $^{12}$C + $^{232}$Th system. It has been simulated and experimentally verified that sudden rise in $\sigma_m^2$ values could not be explained by any systematic error, e.g., loss of energy of fragments in target or mismatch of timing in two T.O.F. arms. The anomalous increase in angular anisotropy in all these systems [6,7,8,16,17,18] has been shown by open symbols in the lower panel of Fig. 4 d-f. It is interesting to note
that anomalous increase in width of the mass distribution were observed at almost the same beam energies at which anomalous enhancement in fragment angular anisotropy were reported.

Fig. 4. Variation of $\sigma^2_m$ (solid squares) with excitation energy for three systems. The dotted and dot-dashed curves are variation for normal and postulated quasi-fission modes, respectively. Calculated $\sigma^2_m$ (thin and thick solid lines) are shown for two critical angles ($\theta_c$). Reported anisotropy A (open symbols) and SSPM predictions (dashed lines) are shown in lower panels. The arrow points to excitation energy corresponding to Coulomb barrier.

Observation of a sudden rise in $\sigma^2_m$ values as the excitation energy is lowered may signify a mixture of two fission modes, one following the normal statistical prediction of fusion-fission path along zero left-right mass asymmetry ($\alpha$), and another following a different path in the energy landscape with zero or small mass asymmetry. The mixture of the two modes could give rise to wider mass distributions. Similar to the postulation of the orientation dependent quasi-fission [3], we postulate that for fusion-fission paths corresponding to the projectile orientations up to a critical angle ($\theta_c$) of impact on the polar region of prolate thorium, the width and energy slope of the symmetric mass distributions are different, as shown by dot-dashed curves in Fig. 4 a-c, compared to those for the normal statistical fusion-fission paths (dotted curves). The mass widths weighted by the fission cross sections (which are assumed to be very close to fusion cross section as the composite systems are of high fissility) from earlier measurements [6] are mixed for the two fusion-fission modes and shown by thick and thin continuous curves in Fig. 4 a-c for different critical polar angles separating the two fission modes, for all three systems. As can be seen from the reasonable agreement of the mixed $\sigma^2_m$ values with the observed
fission fragment mass widths, we can phenomenologically explain the observed increase in the widths of the mass distributions when energy is decreased. It is interesting to note that the fusion-fission process is clearly dominated by the normal process at above Coulomb barriers and the "anomalous" fission process is dominant at lower energies. However, experimental evidence suggests that the variations of mass distributions with excitation energies are similar for the both processes, probably dominated by macroscopic forces, but differing quantitatively due to microscopic effects.

Extensive calculations of the multidimensional potential energy surface have successfully explained spontaneous and low energy fission phenomena [19,20]. Calculated paths through the minimum energy valleys and over ridges in the potential surface showed that apart from the deformations and necking of the two nascent fragments, the left-right mass asymmetry also plays a crucial role. All the heavier than actinide nuclei show mass symmetric ($\alpha = 0$) and mass asymmetric ($\alpha \neq 0$) saddle shapes with a ridge separating the two down the scission path. The relative heights of the two saddles and the separating ridge governed the symmetric, asymmetric or a mixture of the two fission paths in specific cases. Recent extensions [21,22] of the five dimensional energy landscapes for fusion of $^{48}$Ca with $^{244}$Pu have been carried out. In addition to the calculated minimum energy path to reach the fusion meadow and the subsequent descent to the fission valley over a mass symmetric unconditional saddle corresponding to the fusion-fission (FF) path, at higher excitations, most of the paths may deviate through a mass symmetric saddle shape before fusion to re-separate in a quasi-fission (QF) reaction mode. In a very similar situation, in case of fusion of spherical projectiles with deformed $^{232}$Th nuclei, above the Coulomb barrier, the system follows a fusion-fission path over the mass symmetric unconditional saddle. But as the energy is decreased, these paths are progressively blocked and then the microscopic effects come into play. For the polar region of the deformed target, the system starts from an initial condition with varying deformation, separation and damping of radial motion. This results in the system finding a minimum energy path skirting the fusion meadow and over an almost mass symmetric saddle. In analogy to skiers coming down a mountain slope from different heights (initial energy), go over a peak (fusion barrier) to a meadow (fusion) and continuing to slide over a small hillock (unconditional fission barrier) to reach the valley below (scission) in the established route (FF), those who start just below or at the peak, the normal route is blocked. However, if mountainsides are different (microscopic effect due to deformation) and a ridge exists near the peak, some of the skiers can reach the ridge and follow it over a hilltop (conditional mass symmetric saddle) and reach almost the same spot at the valley in different route (QF). However, for a spherical target, the mountain sides are all similar and no ridges exist. The current experimental results strongly indicates the likely scenario described above and calls for detailed calculations of the energy diagrams for the motion of the nucleons through the dissipative system with
different initial conditions.

We have clearly established with the present string of precise measurements that widths of the mass distributions is a sensible tool to observe departure from the normal fusion-fission path in the fusion of heavy nuclei. The exact mechanisms for the departure from normal fusion-fission paths are not known accurately, although it has been stressed that effect of any admixture of transfer fission can be ruled out. However, macroscopic effects such as the direction of mass flow or the mass relaxation time being too prolonged may not be the cause. It has been established earlier from the experimental barrier distributions, the reaction cross sections in $^{19}$F, $^{16}$O, $^{12}$C + $^{232}$Th in near and below Coulomb barrier energies are mostly for impact of the projectiles on the polar regions of the thorium nuclei. Following the quantum mechanical effects favouring similar shapes in entrance and exit channels [1], we modify the simple postulation of the microscopic effects of the relative orientation of the projectile to the nuclear symmetry axes of the deformed target [3]. We assume that for the non-compact entrance channel shape, the impact of the projectile in the polar region of $^{232}$Th target drives the system to an almost mass symmetric saddle shape, rather than a compact equilibrated fused system. The observed fragment mass widths can be quantitatively explained under such assumptions. At sub-coulomb barrier energies, the cross sections for quasi fission channel increases with increased initial separation of the two nuclei in touchy condition, i.e., the effect increased with the mass of the composite system. So quasi-fission is more prominent in $^{19}$F + $^{232}$Th than $^{12}$C + $^{232}$Th. However in each system, as beam energy is increased, the reaction rapidly spread over all the nuclear surface and quasi-fission channels are overshadowed by normal fusion-fission. The above postulation is supported by the observation that for the spherical target $^{209}$Bi, where entrance channel compactness of shape is same for all relative target-projectile orientations, only normal fusion-fission paths, as characterized by the smooth variation of fragment mass widths with excitation energy, are observed. It is also worthwhile to note that effect of the anomalous mass widths increases with left-right mass symmetry in the entrance channel in case of $^{19}$F, $^{16}$O, $^{12}$C + $^{232}$Th system in consonance with our description. Our measurements indicate that higher entrance channel mass asymmetry and energies close to the Coulomb barrier are preferable to increase the probability of reaching the fusion meadow in synthesis of super-heavy elements in heavy ion reactions.

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