Sharp change-over from compound nuclear fission to quasifission

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Fission fragment mass distributions have been measured from the decay of the compound nucleus ^{246}Bk that has been populated via two entrance channels. These entrance channels have a slight difference in their mass asymmetries that puts them on either side of the Businaro Gallone mass asymmetry parameter. Both target nuclei were deformed. Near the Coulomb barrier, at similar excitation energies, the width of the fission fragment mass distribution was found to be drastically different for the ^{14}N + ^{232}Th reaction compared to the ^{11}B + ^{235}U reaction. The entrance channel mass asymmetry was found to affect the fusion process sharply.

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In recent years considerable interest has been observed in studying the fusion of two heavy ions to reach the shore of an island of stability in the super heavy nuclei with atomic numbers (Z) around 114–126 [1,2]. In fusion reactions leading to the formation of a super heavy nucleus, the projectile and the target must overcome the large Coulomb repulsion and reach an attractive potential pocket due to strong nuclear forces. The fused dinuclear system may then reach a compact, heavy mononuclear configuration, which equilibrates in all degrees of freedom to a compound nucleus (CN). The equilibrated compound nucleus is formed at high excitation energies. Because of its very high fissility, it often reaches an unconditional mass-symmetric saddle through shape oscillations and subsequently undergoes binary fission. The system may also deexcite through light-particle evaporation and end up as an evaporation residue (ER) that is stable (or has a long lifetime). The deexcitation of the equilibrated compound nucleus is expected to be completely governed by statistical laws and should have no memory of its process of formation. The formation of compound nucleus (and subsequently the ER) is the key for the formation of super heavy nuclei. This, therefore, necessitates detailed understanding of the roles played by fission and fission-like processes in general and the fusion inhibition due to quasifission processes in particular.

Quasifission is a serious competitor for the formation of a compound nucleus (and, subsequently, the formation of an ER). Quasifission is a fission-like process that precedes the formation of a compact mononuclear system. In heavy systems (typically, $Z_t \cdot Z_p \gtrsim 1600$, where Z_t and Z_p are the target and projectile atomic numbers), the compactness of the exit channel configuration may prevent the formation of a mononucleus leading instead to quasifission. However, also for the fusion of much lighter systems ($Z_t \cdot Z_p \sim 800$), the existence of quasifission as exit channel has recently been demonstrated [3]. This may be explained macroscopically on the basis of the dynamics of the mass flow in the dinuclear system in the pathway to fusion that depends on the value of the entrance channel mass asymmetry (defined as $\alpha = (A_t - A_p)/(A_t + A_p)$, where A_t and A_p are the target and projectile mass numbers) and its magnitude with respect to the value of the Businaro-Gallone mass asymmetry (α_{BG}). Basically, α_{BG} separates the mass symmetric liquid-drop fission barrier from the asymmetric one [4]. For systems with α higher than α_{BG}, mass flows from the projectile to the target, leading to increasing mass asymmetry, and thus establishes quickly a mononuclear, compact shape. On the other hand, for more symmetric mass pairs with α lower than α_{BG}, mass flows in the direction of more symmetric dinuclear system, which, before evolving to a compact mononuclear shape and equilibrating to a compound nucleus, may pass over a mass-asymmetric saddle shape leading to quasifission. It may be mentioned here that quasifission is different from the fast-fission or precompound fission reactions [5] that follow the formation of a compact mononuclear system. Since quasifission occurs before the target and projectile fuse to form a compound nucleus, it hinders the formation of the ER. It is a prominent reaction channel at low excitation energies, just above the fusion threshold, where the ER formation also maximizes. Therefore, for a proper choice of target and projectile in a reaction aiming at the synthesis of a super heavy element, it is important to study the reaction mechanism of quasifission at beam energies close to the Coulomb barrier for different target-projectile combinations of varying entrance channel mass asymmetry that lead to the same fused system.

Recently Hinde *et al.* [6] studied the production cross section of ^{220}Th and observed that it is not independent of the entrance channel mass asymmetry. They made a comparison of the ER formation from their own experiment on ^{16}O + ^{204}Pb with other experiments on ^{40}Ar + ^{180}Hf, ^{82}Se + ^{138}Ba, and ^{124}Sn + ^{96}Zr, all of which led to the same compound nucleus

^{220}Th. The ER yield was found to be maximum for the most asymmetric entrant mass pair (^{16}O + ^{204}Pb), whereas the yields were less, at least by one order of magnitude, for more symmetric entrance channels. This observation brings out the importance of the role of mass flow as a function of α, the entrance channel mass asymmetry, on the ultimate evolution (CN formation or quasifission) of the dinuclear system. Moreover, the target deformation, in the reactions mentioned above, was different. This might also have played a role in the evolutionary fate of the dinuclear system. In the reactions reported in Ref. [6], ^{16}O + ^{204}Pb is more likely to result in fusion and the formation of an ER than the other three systems with more symmetric entrance channel mass pairs. However, it is interesting to note that the observed [6] hindrance of ER formation was not found to vary systematically with mass asymmetry. Microscopic effects [7] are suspected to affect substantially the ER production in conjunction with the macroscopic effect of the direction of the mass flow toward symmetric fragments which drives the quasifission reaction mechanism.

We have established in our previous communications [8,9] that the variation of the width of the fragment mass distribution with excitation energies is a promising probe for studying quasifission, particularly at energies close to the Coulomb barrier. Because the statistical fission of the compound nucleus proceeds through shape changes over a mass symmetric unconditional fission barrier, the fission fragment mass distribution is symmetric around the average mass of the target and the projectile. In such a case, the width (or standard deviation σ_m) of the mass distribution is a smoothly varying function of the excitation energy. However, because quasifission proceeds through a binary-fission-like reaction mechanism over a mass asymmetric fission barrier, the fragment mass distribution is expected to be mass asymmetric. For a mixture of statistical and quasifission, the mass distribution may still be peaked around the average of the projectile and the target mass, but the width of the mass distribution may get larger. Therefore, if the proportion of the quasifission reaction increases with change in the excitation energy, there will be an increase in the width of the mass distribution. Such an increase in the width of fragment mass distribution with a decrease in energy has been reported recently around the Coulomb barrier [10]. We have observed similar anomalous increases in width of mass distribution with decreases in beam energy in fusion of systems with the deformed ^{232}Th as target [9]. Because the target ^{232}Th is deformed, more emphasis on the postulated nuclear orientation dependent quasifission [11] was given in explaining the observed anomalous changes in the widths of the fission fragment mass distributions.

Hence, from the above experiments, it is suggested that there may be two factors that determine the quasifission reaction: (i) nuclear orientation dependence, particularly for deformed target and/or projectile, and (ii) direction of mass flow toward increased mass symmetry. It is of interest to assess which of these above two effects plays a dominant role. In the present article, we report clear evidence of the importance of the effect of mass flow over that of the dependence on the relative nuclear orientation on quasifission, for different deformed target and projectile combinations.

In the present experiment we chose the systems ^{11}B + ^{235}U and ^{14}N + ^{232}Th, with entrance channel mass asymmetry just larger ($\alpha = 0.911$) and smaller ($\alpha = 0.886$), respectively, than the mass asymmetry for the Businaro-Gallone ridge ($\alpha_{BG} = 0.893$) of the compound nucleus ^{246}Bk. However, the targets ^{235}U ($\beta_2 = 0.215$) and ^{232}Th ($\beta_2 = 0.207$) have similar ground state deformations so that the probabilities of the dinuclear systems forming an elongated shape (which is possible for the projectile hitting the tip of the prolate target) are expected to be similar, because the separation between the centers of the target and the projectile for an end-on collision varies only by about 0.13 fermi between the two cases. The probabilities for quasifission depending upon the orientation of nuclear axes are expected to be similar for both the systems. However, the probability of quasifission depending upon the direction of mass flow toward a symmetric dinuclear system would act in tandem for the ^{14}N + ^{232}Th ($\alpha < \alpha_{BG}$) system but in opposition for the ^{11}B + ^{235}U ($\alpha > \alpha_{BG}$) system. So we would expect quasifission in both the systems if the orientation dependent quasifission is the dominant reaction mechanism. But, we would expect quasifission only in ^{14}N + ^{232}Th and not in ^{11}B + ^{235}U if the direction of mass flow drives the system to quasifission. In the above scenario, we should be able to throw light on the relative importance of mass flow and orientation dependent quasifission mechanisms, as we attempt to form an identical compound nucleus ^{246}Bk with the same excitation energy and a similar angular momentum in both the reactions. We have also used the reaction of ^{14}N + ^{197}Au as a reference reaction, which is expected to be almost pure fusion-fission reaction at the excitation energies covered in the experiment.

The experiment involved measurement of the mass distribution of the fission fragments close to and above the Coulomb barrier. The beam energies were judiciously chosen to populate the ^{246}Bk nucleus at similar excitation energies. The experiment was carried out using pulsed ^{14}N and ^{11}B beams of width about 1.1 ns, with a pulse separation of 250 ns, from the 15UD Pelletron at the Inter University Accelerator Centre (IUAC), New Delhi. Targets of ^{197}Au (self supporting), ^{232}Th (on 200 μg/cm^2 Al backing), and ^{235}U (on 300 μg/cm^2 Ni backing) of thickness 500 μg/cm^2 were used. Targets were placed at an angle of 45° to the beam. Fission fragments were detected with two large area position sensitive MWPC [12]. The detectors were placed at 56 and 30 cm from the target. The operating pressures of the detectors were maintained at about 3 torr of isobutane gas. We measured the flight time of the fragments, the coordinates of the impact points of the fragments on the detector (θ, ϕ), and the energy loss in the gas detectors. From these measurements, we extracted the masses of the correlated fission events and the transferred momentum to the fissioning system.

In the range of the energies of our measurements, it was not expected that the fast-fission reaction would contribute in any significant way. The preequilibrium fission (which is characterized by nonequilibration of the K degree of freedom showing more fragments along the beam-axis than perpendicular to it [13]) does not change the mass distribution of fission fragments because the mass degree of freedom is expected to equilibrate and the system would pass over a mass symmetric unconditional fission barrier. Hence the

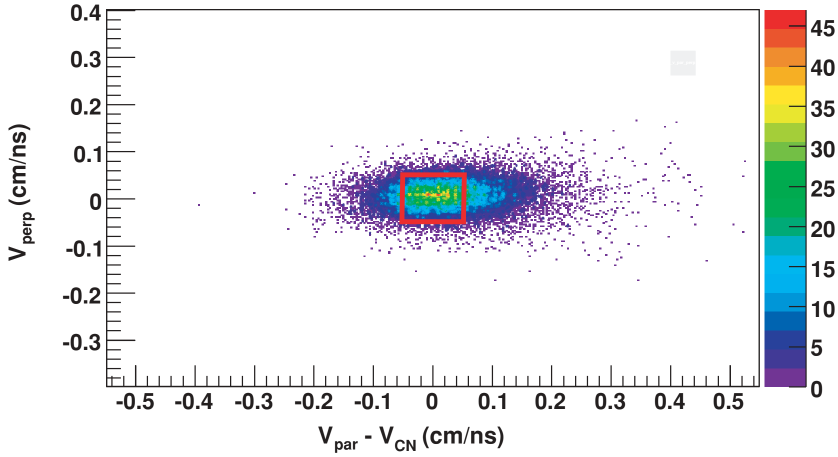

FIG. 1. (Color online) Measured distributions of velocity of the fissioning nuclei formed in the reaction $^{14}N + ^{232}Th$ at $E_{c.m.} = 77.3$ MeV. The (red) rectangle indicates the gate used to select the fusion-fission events for mass determination.

observed mass distribution of fragments would be a mixture of two possible reaction mechanisms—one of compound nuclear statistical fission (with small possible admixture of K nonequilibrated events) over a symmetric mass unconditional fission barrier and the other from quasifission reactions with fission over a asymmetric mass saddle point, before reaching a compact shape and forming a compound nucleus. In both the above reaction mechanisms, the incident projectile momentum would be completely transferred to the fused dinuclear system and the radial motion of the nucleons would be completely damped.

However, for excitation energies close to the Coulomb barrier, it is well known that fission fragments are also produced in an incomplete fusion-fission reaction, in which only a part of the projectile fuses with the target and the incompletely fused binary system equilibrates and undergoes statistical fission reaction. Hence such a reaction mechanism, known as transfer fission (TF), needs to be separated experimentally from the compound nuclear and quasifission reactions. The fission fragments from complete fusion events followed by fission were exclusively selected from the correlation of the velocities of the fissioning system (V_{par}) in the beam direction relative to the recoil of the fused system and the velocity perpendicular to the reaction plane (V_{perp}), as well as the correlation of the polar and azimuthal angles of the fragments (θ, ϕ) with respect to the beam axis [12,14]. For the complete fusion-fission (FF) process, the events were centered in the scatter plot with velocities ($V_{\mathrm{par}} - V_{\mathrm{CN}}$), $V_{\mathrm{perp}} = 0, 0$. The scatter in the velocities was essentially due to neutron evaporation from the fragments, as shown in Fig. 1, in a typical measurement of the distributions of the complementary fission events for the system $^{14}N + ^{232}Th$ at $E_{\mathrm{cm}} = 77.3$ MeV. The events corresponding to the incomplete fusion-fission events, called transfer fission (TF), are scattered around nonzero $V_{\mathrm{par}} - V_{\mathrm{CN}}$, V_{perp} values. The events due to the above two reaction channels (i.e., FF and TF) are also shown in a plot of the distribution of the polar folding angle in Fig. 2. Figure 2(a) shows that the measured folding angle distribution of FF events is peaked around 165° consistent with the expected value for complete transfer of momentum of the projectile. The events for incomplete fusion (i.e., TF), are peaked around a smaller folding angle as the ejectile moves in the backward direction. Figure 2(b) shows the same distributions for the events that are with in the rectangular gate in the $V_{\mathrm{par}} - V_{\mathrm{CN}}$, V_{perp} plot as shown in Fig. 1. It can be seen that the contribution of the incomplete fusion events (i.e., TF events) are drastically reduced and are estimated to be only about 1% of the fusion-fission (FF) events. The fission fragments are well separated from elastic and quasielastic reaction channels, both from the time correlation and energy loss spectra in the detectors. The masses were determined from the difference of the time-of-flights, polar and azimuthal angles, momentum, and the recoil velocities for each event within the gate on $V_{\mathrm{par}} - V_{\mathrm{CN}}$, V_{perp} as described. The experimental arrangements and the data analysis procedure were described in detail in earlier reports

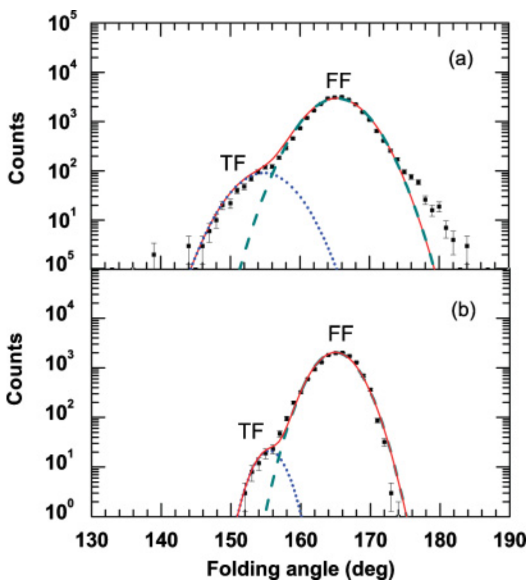

FIG. 2. (Color online) (a) Measured folding angle distributions of all fission fragments in the reaction $^{14}N + ^{232}Th$ at $E_{c.m.} = 77.3$ MeV. Solid squares are the experimental points, (green) dashed and (blue) dotted lines represent the Gaussian fitting of the fusion fission (FF) and transfer induced fission (TF) events, respectively. The sum of the two Gaussian fittings is shown by the solid (red) line. (b) Folding angle distributions of the fission fragments that are only with in the rectangular gate of Fig 1.

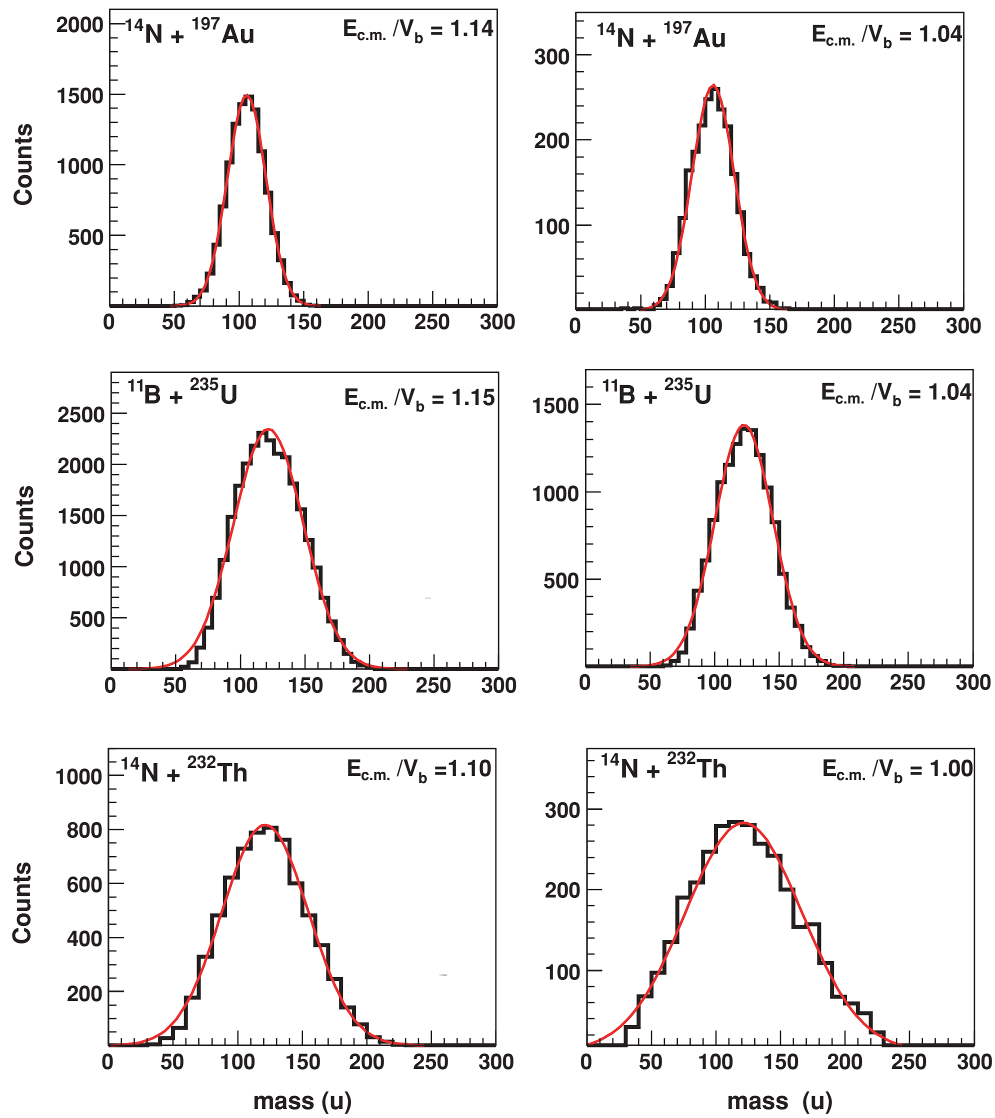

FIG. 3. (Color online) Measured mass distributions for the reactions $^{14}N + {}^{197}Au$, $^{11}B + {}^{235}U$, and $^{14}N + {}^{232}Th$ near and above the Coulomb barrier. The Gaussian fits are shown by (red) solid lines.

[9,12]. It may be noted that determination of mass from the difference of fragment flight times eliminates the uncertainty due to the time structure of the beam.

Representative mass distributions, near and above the Coulomb barrier energies, are shown in Fig. 3 for $^{14}N + {}^{197}Au$, $^{11}B + {}^{235}U$, and $^{14}N + {}^{232}Th$ systems. It can be observed that measured mass distributions are well fitted with single Gaussian distribution at all energies. The variations of the standard deviations (σ_m) of the fitted Gaussian to the experimental masses as a function of $E_{\text{c.m.}}/V_b$, where $E_{\text{c.m.}}$ is the beam energy in center of mass system and V_b is the Coulomb barrier, are shown in Fig. 4 for all three measured systems. It is seen that for $^{14}N + {}^{197}Au$ and $^{11}B + {}^{235}U$ reactions the variation of σ_m is smooth across the Coulomb barrier. This smooth variation of σ_m for the above two systems is in complete agreement with the expectation from a statistical fission reaction. However, a significant difference in the trend of the variation of σ_m is observed for the $^{14}N + {}^{232}Th$ system in which σ_m increases with decreasing energy. Careful rejection of any contribution due to incomplete fusion events attributes this increasing trend of width of the distribution with lowering energies to events with complete fusion of the target and the projectile.

The variation of standard deviation of the mass distributions with excitation energy for two reactions $^{14}N + {}^{232}Th$ and $^{11}B + {}^{235}U$ forming the same composite system ^{246}Bk is shown in Fig. 5. The solid (red) line in the figure shows the calculated variation from statistical theory [15] following the relation $\sigma_m^2 = \frac{1}{k}\sqrt{\frac{E^\dagger}{a}}$, where $E^\dagger$ is the excitation energy at the scission point and a is the nuclear level density parameter. Here, k is related to the stiffness of the potential energy landscape at the top of the barrier. A value of $k = 0.0033$ MeV/u^2 fitted the $^{11}B + {}^{235}U$ data well and is consistent with the comprehensive compilation of the data presented in Refs. [17] and [16]. It is interesting to note that for the $^{14}N + {}^{232}Th$ reactions, not only is there a sudden jump of mass widths (or σ_m) near the

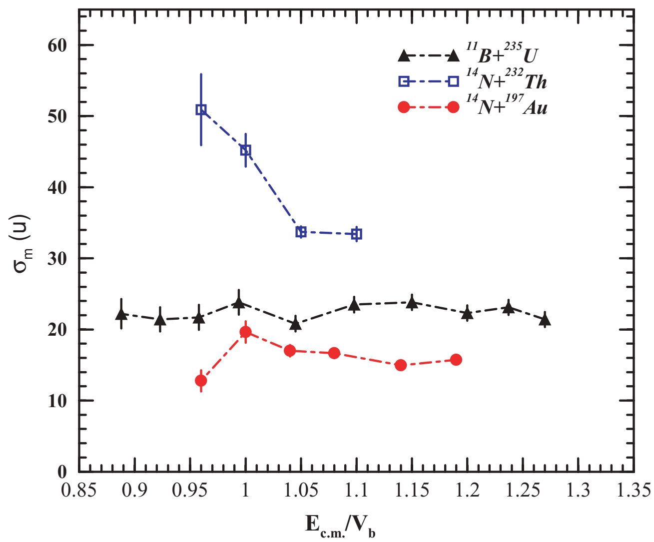

FIG. 4. (Color online) Variation of the standard deviation σ_m to the fitted Gaussian of the fission fragment mass distribution as a function of $E_{c.m.}/V_b$.

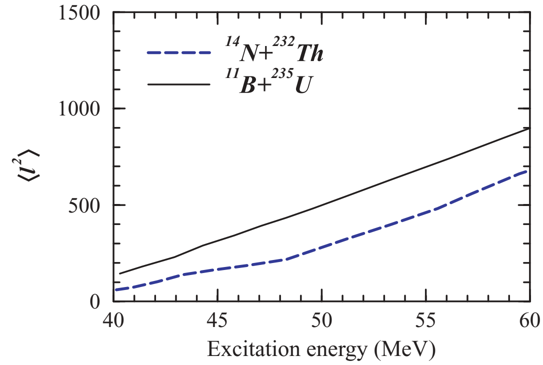

FIG. 6. (Color online) Variation of mean square angular momentum with excitation energy for the nucleus ^{246}Bk, populated by the two different entrance channels.

Coulomb barrier but also the magnitude is higher than that of the $^{11}B + ^{235}U$ reactions over the entire range of excitation energies.

The width of fission fragment mass distribution mainly depends on the excitation energy. However, it has weaker linear dependence on the mean square angular momentum $\langle l^2 \rangle$ brought in by the projectile [18]. In Fig. 6 we show the deduced [19] variation of $\langle l^2 \rangle$ with excitation energy for the two systems forming the same composite system ^{246}Bk. It can be seen that average mean square angular momentum value for the system $^{11}B + ^{235}U$ is always higher than that for $^{14}N + ^{232}Th$ in the range of our measured excitation energies. So, even if we correct the predicted widths of the mass distribution for the possible contribution for $\langle l^2 \rangle$, the correction for the $^{14}N + ^{232}Th$ system would be smaller compared to that for $^{11}B + ^{235}U$ and would not explain the observed sharp increase of the mass width for $^{14}N + ^{232}Th$ around the Coulomb barrier as shown in Fig. 5.

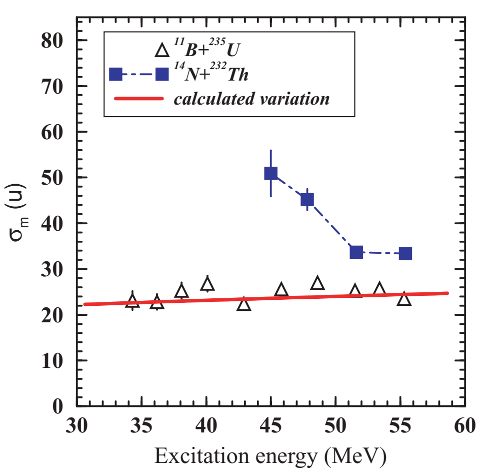

FIG. 5. (Color online) Measured variation of σ_m with excitation energy for the two reactions forming the same composite system ^{246}Bk. The calculated variation is shown by the solid (red) line.

We have observed that, for the fusion of two heavy nuclei forming the compound nucleus ^{246}Bk through the reactions $^{11}B + ^{235}U$ and $^{14}N + ^{232}Th$ at a similar excitation energy, the fused system behaves differently. For the system $^{11}B + ^{235}U$ ($\alpha > \alpha_{BG}$), the width of the mass distribution follows the trend expected for a statistical fission from an equilibrated compound nucleus. Because the mass flow in this system is toward increased mass asymmetry, the dinuclear system is expected to evolve quickly to a mononuclear system and reach equilibration. The absence of any anomalous increase in the width of the mass distribution also shows that the effect of nuclear orientation is not significantly present even for the events in which the projectile collides with the tip of the deformed target. But for the system $^{14}N + ^{232}Th$ ($\alpha < \alpha_{BG}$) near the Coulomb barrier, the width of the mass distribution begins to rise, contrary to the expectation for fission from a compound nucleus. Because we have already observed that the effect of nuclear orientation is insignificant for the initial configuration, which is similar to that of $^{11}B + ^{235}U$, we conclude that the direction of mass flow, which is toward increased mass symmetry, triggers the evolution of the dinuclear system toward a quasifission reaction that results in the increase of the width of the mass distribution in this system.

Therefore, in the fusion of the ^{246}Bk, in addition to the effect of deformation, the entrance channel mass asymmetry plays a crucial role in the reaction mechanism, particularly in the energies close to the Coulomb barrier. For the systems $^{11}B + ^{235}U$ to $^{14}N + ^{232}Th$, the entrance channel mass asymmetry changes from just above to just below that for the Businaro-Gallone ridge. This small change effectively reverses the flow of mass in fusing the target and the projectile from increasing mass asymmetry to increasing mass symmetry. The reversal of the mass flow results in a completely different reaction mechanism from a predominant formation of compound nucleus in the $^{11}B + ^{235}U$ reaction to reseparation of the composite in

$^{14}N + ^{232}Th$ over a mass asymmetric saddle in a quasifission reaction. This sharp macroscopic effect of mass flow determining the compound nuclear fusion-fission or the quasifission reaction for a small change in the entrance channel mass asymmetry shows the importance of mass flow during fusion of ^{246}Bk. For the synthesis of super heavy nuclei, the entrance channel would require more symmetric target and projectile masses than in the fusion of ^{246}Bk. Our measurements indicate that the sharp change-over of the reaction mechanism from fusion to quasifission with the reversal of mass flow across the Businaro Gallone ridge would be a crucial factor in the synthesis of super heavy nuclei.

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[1] Yu. Ts. Oganessian *et al.*, Phys. Rev. C **74**, 044602 (2006).
[2] M. G. Itkis *et al.*, Nucl. Phys. **A734**, 136 (2004).
[3] A. C. Berriman *et al.*, Nature (London) **413**, 144 (2001).
[4] U. L. Businaro and S. Gallone, Nuovo Cimento **5**, 315 (1957).
[5] S. Kailas, Phys. Rep. **284**, 381 (1997).
[6] D. J. Hinde, M. Dasgupta, and A. Mukherjee, Phys. Rev. Lett. **89**, 282701 (2002).
[7] P. Moller and A. J. Sierk, Nature (London) **422**, 485 (2003).
[8] T. K. Ghosh, S. Pal, T. Sinha, S. Chattopadhyay, P. Bhattacharya, D. C. Biswas, and K. S. Golda, Phys. Rev. C **70**, 011604(R) (2004).
[9] T. K. Ghosh *et al.*, Phys. Lett. **B627**, 26 (2005).
[10] R. Rafiei *et al.*, Phys. Rev. C **77**, 024606 (2008).
[11] D. J. Hinde *et al.*, Phys. Rev. Lett. **74**, 1295 (1995).
[12] T. K. Ghosh *et al.*, Nucl. Instrum. Methods Phys. Res. A **540**, 285 (2005).
[13] V. S. Ramamurthy and S. S. Kapoor, Phys. Rev. Lett. **54**, 178 (1985).
[14] D. J. Hinde, M. Dasgupta, J. R. Leigh, J. C. Mein, C. R. Morton, J. O. Newton, and H. Timmers, Phys. Rev. C **53**, 1290 (1996).
[15] B. B. Back *et al.*, Phys. Rev. C **53**, 1734 (1996).
[16] A. Ya. Rusanov, M. G. Itkis, and V. N. Okolovich, Phys. At. Nucl. **60**, 683 (1977).
[17] M. G. Itkis *et al.*, Yad. Fiz. **52**, 23 (1990).
[18] G. N. Knyazheva *et al.*, Phys. Rev. C **75**, 064602 (2007).
[19] B. R. Behera *et al.*, Phys. Rev. C **64**, 041602(R) (2001).