Experimental Signatures of Quasifission

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Abstract. Three decades ago, it was first recognized that the observation of fission fragments in heavy-ion induced reactions does not necessarily mean that they originate from the fission decay of a compound nucleus formed by the fusion of the projectile and the target nuclei. This conclusion was based on several different observations. First, it was recognized that the fission cross section exceeded the upper bound imposed by the existence of a stabilizing pocket in the ion-ion potential; thus some of the fission cross section must originate from partial waves that do not proceed through a compound nucleus and, in addition, that the fission mass distribution in these cases was observed to be wider than expected on the basis of a compound nucleus model. Concurrently, it was noted that the fission fragment anisotropy in heavy-ion induced fission substantially exceed expectations based on the transition state model. Subsequent studies of the two-dimensional mass-angle distribution of fission fragments clearly demonstrated that these fragments are the result of a dynamic process, in which the system evolves toward mass symmetry on a time scale that is commensurate with the rotational period of the complex. This process is now referred to as “quasi-fission” although the terms “fission without a barrier” and fast fission were also used. Recently, much progress in the theoretical description of this process has been achieved and further precise experiments have been conducted, which provide further constraints on our understanding of these complex processes that also play a critical role in attempts to synthesize heavy and super-heavy nuclei via heavy-ion fusion processes. In this talk I will discuss some of the history and recent developments in the study of the quasi-fission process.

1. A little history
The history of fission research goes back more than seventy years with the discovery of this new, unexpected and very spectacular process, in which an atomic divides into two almost equal size parts or fragments. The discovery is attributed to Hahn and Strassmann [1]. Science historians would, however, argue that also Lise Meitner made decisive contributions to this work, but that she was not given due credit because of the societal and political situation in Germany at the time. In retrospect, earlier hints of the fission process were probably missed in work by Enrico Fermi at Rome [2], who were disturbed by some large “noise” pulses in his Geiger-Müller tube when bombarding uranium with slow neutrons, but as experimenters can testify, aluminum foil can very often get rid of such large electronic noise, and this method also worked for Fermi when he inserted a this Al foil between the uranium and the detector. With the benefit of hindsight it is evident that the large “noise” pulses were probably caused by fission fragments reaching his detector. However, Ida Noddack [3] did suggest the revolutionary idea that the observation of reaction products with barium-like chemistry could indeed be Ba that was created by splitting the uranium nucleus into two large, almost equal fragments, but...
the idea was too outrageous to get serious consideration at the time and her suggestion was essentially ignored.

When Hahn and Strassmann again observed Ba products in neutron irradiated uranium [1], and the news of the results reached Lise Meitner and her nephew Otto Frisch, who were both in Sweden at the time, they quickly understood the implications of the experimental observation and concluded that what was seen was a process in which the uranium nucleus splits in half by neutron bombardment. Meitner and Frisch quickly published a letter in Nature [4] giving this explanation of the Berlin group’s results. In addition, within a couple of weeks of learning about the Hahn and Strassmann results, Otto Frisch was able to confirm the results [5] when he repeated the experiment in Stockholm. In the meantime, Niels Bohr learned about the new process from Lise Meitner over the Christmas holidays in 1938, just in time before he departed for a longer trip to the United States in January 1939. During his stay at Princeton University he devoted his time to understand and describe the fission process in detail in collaboration with John A. Wheeler. This collaboration resulted in the famous Physical Review article published in June 1939 [6], only seven months after the discovery, that gives a detailed exposition and understanding of this newly discovered process - a remarkable feat. Figure 1 shows the potential energy surface for $^{235}\text{U}$ and the fission barrier that they calculated for this system.

![Figure 1](image.png)

**Figure 1.** In this figure from Ref. [6] the potential energy surface obtained in the liquid drop model is shown along with the resulting fission barrier.

If is difficult to overestimate the consequences of the discovery of fission. Of course, it has led to a long and very fruitful research field in nuclear physics studies that we all love and which is continuing at this symposium. But it also launched nuclear physics into the public eye because of the far-reaching implications for the nuclear chain reaction in electric power production and nuclear explosions. Hardly a day goes by that nuclear fission is not the center focus of a news story in terms of either of these applications, even more than seventy years hence. But in some way, I believe that the whole field of nuclear physics, no matter how removed from studies of the fission process, has benefitted from this discovery in terms of justifying the continued study of this field, also the branches that do not have applications that are of interest for society as a whole.

After this brief diversion, I should get back on track and discuss the topic of “quasi-fission” and the experimental signatures of this process as the title of my talk promises. In the following, I will therefore touch on some of the early indications that the fission fragments seen in heavy-ion
bombardments originate from a slightly different process, although several properties do justify the “fission” part of the name.

2. Large fission cross sections and wide mass distributions

One of the first indications that fission fragments can come from non-equilibrium fission were seen in the work of Heusch et al. in the reaction of $^{132}$Xe+$^{56}$Fe at 5.73 MeV/u [7]. They found a fission-like cross section of 1040 mb, which corresponds to a maximum compound nuclear spin of 120 $\hbar$ (in the sharp cutoff model). This agrees well with fusion models [8], [9] assuming that fission exhausts the total fusion cross section. But they also realized that not all of these reactions could come from an equilibrated compound nucleus since the fission barrier disappears for spins larger than about 72 $\hbar$ according to the Rotating Liquid Drop Model [10]. Therefore, the fission-like processes originating from total angular momenta in the range 72 $\hbar < l < 120 \hbar$ must be characterized as “fission without a barrier”.

"The strongly damped component which constitutes the major part of the reaction cross section exhibits characteristics of a fusion-fission reaction with typical fission fragment kinetic energies and $1/\sin \theta$ angular distributions... We tentatively conclude that an essential part of the fully damped cross section originates from partial waves for which the compound nucleus has no fission barrier”.

Figure 2. Systematics of the width of fission fragment mass distribution obtained in different systems. The data are shown as a function of the difference between the critical angular momentum for fusion, $l_{\text{crit}}$, and the angular momentum for which the fission barrier vanishes $l_{Bf=0}$, such that the “fission without a barrier” contributes for positive values of the abscissa.

In addition it was noticed that the width of the mass distribution of fission fragments increased strongly for the partial waves where the fission barrier vanishes. Lebrun et al. [11] studied fission mass distributions of $^{20}$Ne+$^{nat}$Re and $^{40}$Ar+$^{165}$Ho reactions, each at two bombarding energies, and they found that the width of the fragment mass distributions increased noticeably with angular momentum and that it appeared to widen considerably above the partial wave for which the fission barrier vanishes. Later, Borderie et al. [12] reviewed a larger data set and found this conclusion fully supported and concluded that “fast fission” occurs for $l$-values, where the fission barrier vanishes, see Fig. 2.

Further evidence was found by Shen et al. [13] who analyzed the mass width of fission-like reaction products seen in U induced reactions and showed that, while the expected width was
seen for reactions with $^{16}$O and $^{24}$Mg, heavier targets lead to wider mass distributions. The width of the mass distribution may be estimated in a thermal model assuming a quadratic mass asymmetry potential

$$U(A) = \frac{1}{2} k (A - A_s)^2,$$

where $A_s$ is the symmetric mass, and $k$ is the stiffness coefficient, which based on theoretical liquid drop model estimates [14] is about $k = 0.0035$ MeV/u$^2$. At a temperature $T$ one obtains a variance of the mass distribution of

$$\sigma_A^2 = \frac{T}{k} \frac{8.5E^+}{A},$$

where $E^+$ is the nuclear excitation energy at scission given by $E^+ = E_{exc} + Q_{sym} - E_K - E_{def} - E_{rot}$. Here $E_{exc}$ is the excitation energy of the compound system, $Q_{sym}$ is the Q-value for symmetric fission, $E_K$ is the total kinetic energy estimated from the Viola systematics [15], $E_{def}$ accounts for the fragment deformation energy taken to be 12 MeV, and $E_{rot}$ is the rotational energy at scission. One thus expects only a weak (fourth root) increase in the mass width as a function of excitation energy. This dependence is found to agree well with data, as demonstrated for the reactions $^{238}$U+$^{16}$O, $^{26}$Mg [13] and for $^{32}$S+$^{182}$W and $^{48}$Ti+$^{166}$Er [16], see Fig. 3 left panel, but a somewhat larger stiffness coefficient of $k = 0.0048$ MeV/u is needed to reproduce the data.

Figure 3. Left panel: The standard deviation of fission fragment mass distributions are shown as a function of the available excitation energy at the scission point, $E^+$ for reactions of $^{32}$S+$^{182}$W, $^{48}$Ti+$^{166}$Er, and $^{60}$Ni+$^{154}$Sm, all leading to the compound system $^{214}$Th, from [16] (solid symbols) and $^{16}$O and $^{12}$C$^{+208}$Pb, [17] (open circles). The solid curve represents Eq.2 using a value of $k = 0.0048$ MeV/u$^2$ obtained by fitting the $^{16}$O and $^{12}$C$^{+208}$Pb data. Right panel: The excitation energy dependence has been removed and the data are shown as a function of the variance of the spin distribution.

One observes that the $^{60}$Ni reaction, also leading to the $^{214}$Th compound system, has substantially wider fission mass distributions. However, since a heavier projectile is used in this case, one may argue that this increase is caused by probing larger angular momenta in the fission process. To test this hypothesis one may remove the excitation energy dependence
by dividing with \((E^+)^{1/4}\) and display the results as a function of the mean squared angular momentum, \(\langle I^2 \rangle\), as illustrated in the right panel of Fig. 3. Using this analysis, it becomes evident that the main factor determining the large mass widths seen for \(^{60}\text{Ni}+^{154}\text{Sm}\) is most likely associated with the heavier projectile mass used in this system.

The conclusion from this analysis appears to be that there is good agreement with expectations based on thermal excitations in the mass asymmetry mode at scission for reactions induced by relatively light ions. For heavier projectiles, this thermal model fails and the observed large mass widths point to a dynamical origin of this effect, one that we now associate with the quasi-fission process.

3. Fission angular distributions

The angular distributions of fission fragments from excited nuclei is expected to be determined at the transition state in the process, namely the saddle point, where the orientation of the nuclear symmetry axis is given by statistical arguments following Halpern and Strutinsky \([18]\). In this formulation, the angular distribution may be written

\[
W(\theta) = \sum_I (2I + 1) T_I \sum_K \rho_I(K) |d_{0K}(\theta)|^2,
\]

(3)

where \(I\) is the spin of the nucleus, \(K\) its projection onto the symmetry axis, \(T_I\) is the transmission coefficient for fusion with spin \(I\), \(\rho_I(K)\) is the probability of spin projection \(K\) for total spin \(I\), and \(d_{0K}\) is the \(d\)-function. The level density at the saddle point may be written

\[
\rho(E, I, K) \propto e^{(E-B_I-E_{\text{rot}})/T}
\]

where the rotational energy, \(E_{\text{rot}}\) is

\[
E_{\text{rot}} = \frac{\hbar^2}{2J_\parallel} K^2 + \frac{\hbar^2}{2J_\perp} (I(I + 1) - K^2) = \frac{\hbar^2}{2J_\parallel} I(I + 1) + \frac{\hbar^2}{2} K^2 \left( \frac{1}{J_\parallel} - \frac{1}{J_\perp} \right),
\]

(4)

such that

\[
\rho(K) \propto e^{-K^2/2K_0^2},
\]

(5)

where

\[
K_0^2 = \frac{T}{\hbar^2} J_{\text{eff}}; \frac{1}{J_{\text{eff}}} = \frac{1}{J_\parallel} - \frac{1}{J_\perp}.
\]

(6)

Measurements of the fission anisotropy therefore gives information about the moments of inertia \(i.e\). the deformation of the saddle point configuration. To a good approximation, the fragment anisotropy, \(W(0^\circ)/W(90^\circ)\), is related to the variance of the \(K\)-distribution, \(K_0^2\), and the mean square of the spin distribution according to

\[
\frac{W(0^\circ)}{W(90^\circ)} \approx 1 + \frac{\langle I^2 \rangle}{4K_0^2}.
\]

(7)

The Halpern-Strutinsky model is based on the assumption that the angular distribution is determined at the saddle point, but in the early days it was also proposed that the scission point could take this role. However, a systematic set of data obtained by Reising \(et\ al\). [19] settled this question, at least for light-ion induced reactions. They used 42.3 MeV \(\alpha\)-particles to bombard a wide range of targets from \(^{197}\text{Au}\) to \(^{249}\text{Cf}\) and derived transition state shapes using the Halpern-Strutinsky model. The results, represented in terms of the parameter \(J_0/J_{\text{eff}}\), where \(J_0\) is the moment of inertia corresponding to the spherical shape, are given in Fig. 4 (open circles) as a function of the fissility parameter, \(x\). The solid curve represents the expectation on the basis of the liquid drop model, namely that the deformation of the saddle point increases with \(x\) up to the Businaro-Gallone point at about \(x=0.7\) whereafter it assumes less elongated shapes until it
Figure 4. The saddle point shape, given by $J_0/J_{\text{eff}}$ as predicted by the liquid drop model, is shown as the solid curve as a function of the fissility, $x$ and compared with experimental data. 

reaches sphericity at $x=1$. Here, at the spherical shape, the repulsive Coulomb forces are exactly balanced by the nuclear attractive forces represented by the surface tension in the liquid drop model and the fission barrier vanishes. It is clear that the equilibrium shape, that is reflected in the fission angular distributions, coincide with the saddle point as predicted by the liquid drop model and not at the scission shape represented by a dashed line for $x > 0.7$.

It is, however, interesting to ask what the fission anisotropy would be for systems with even higher fissility, $x=1$ or above. Taken at face value, the $K$-distribution should become flat such that no fission direction relative to the spin axis would be preferred resulting in a flat fission

Figure 5. The angular distributions for 218 MeV $^{32}$S-induced fission on targets of $^{197}$Au, $^{232}$Th, $^{238}$U, and $^{248}$Cm [20] are shown in the left part of the figure. Solid curves are the optimal fits to the data using Eq. 3 whereas the dashed curves are expected from the liquid drop model saddle shapes. Data for the $^{16}$O+$^{209}$Bi [21] system are shown in the right panels.
angular distribution. On the other hand, it seems unlikely that the system would not be able to re-adjust its orientation to minimize its rotational energy during the descent from saddle to scission, which for such heavy systems corresponds to a substantial change in the nuclear shape. In order to address this question, we studied four reactions using $^{32}$S beams at 218 MeV on targets of $^{197}$Au, $^{232}$Th, $^{238}$U, and $^{248}$Cm with fissilities of $x=0.817$, 0.899, 0.914, and 0.948, respectively, thus approaching the critical value of $x=1$ [20]. The resulting angular distributions are shown in Fig.5. Moreover, recent studies of anisotropies at near and below barrier energies in the $^{32}$S+$^{232}$Th system by Hinde et al. [22] also lead to the conclusion that this system bypasses the compound nucleus as well as studies of the $^{32}$S+$^{184}$W reaction by Zhang et al. [23], which find a substantial quasifission contribution.

In these $^{32}$S-induced reaction we clearly see a strong deviation from the naive expectation based on the saddle point model (dashed curves), whereas $^{16}$O-induced fission on a $^{209}$Bi target [21], a system with a slightly lower fissility of $x=0.774$, appears to agree well with the theory.

It should be mentioned here that at the time these data were originally published, alternate explanations for the enhanced anisotropy in some heavy-ion induced reactions were put forward. These explanations were focused on a proposed failure of the saddle point model and suggested that the angular distributions were determined closer to or at the scission point [24, 25]. Although models of this type are able to explain a small subset of the data, it was shown [26] that scission-type models are not able to reproduce the overall systematic trend of a larger set of data.

To investigate the role of angular momentum, we carried out experiments that compare the $^{16}$O+$^{238}$U and $^{32}$S+$^{208}$Pb reactions [27], which lead to compound systems with almost identical fissility of $x=0.84$ and they should therefore have the same saddle point properties. The results are given in Fig. 6 where the angular distributions are shown on the left at two energies for each system, such that a similar range of angular momenta are populated. One observes that while the $^{16}$O+$^{238}$U data are quite well represented by the liquid drop model predictions for the saddle point shape, this is not the case for $^{32}$S+$^{208}$Pb. Here, the observed fission anisotropies are substantially larger than expected, even at beam energies that populate the same range of angular moments. This is also illustrated in the right side of Fig. 6 where the values of $J_0/J_{eff}$, derived from the fission angular distributions, are shown as a function of the mean square of the spin distribution of the fissioning system, $\langle I^2 \rangle$ in comparison with the rotating liquid drop model expectation. One again observes that the $^{32}$S induced reaction lead to values of $J_0/J_{eff}$ that are substantially larger than expected from theory [10].

From these examples, and numerous others, the following conclusion can be drawn: 1) For fission induced by light ions and heavy ions up to perhaps mass 20-24, the saddle point model gives a good account of the observed fission angular distributions, but for heavier projectiles one finds that the experimental anisotropies are too large. 2) Comparisons of systems populating compound nuclei with essentially identical fission properties (fissility) and angular momenta show that the deviation from the saddle point model prediction appears to be associated exclusively with the smaller mass asymmetry in the entrance channel. This last observation excludes any explanation that involves the formation of an equilibrated compound system as an intermediate step in the process, and one is forced to conclude that the systems with a heavy projectile proceed through a dynamical process that fails to go through the compound nucleus formation. This conclusion is therefore in full agreement with those obtained form the analysis of fission cross sections and the widths of mass distributions mentioned above. Although these conclusions are quite solid, one must keep in mind that they are, to some extent, based on comparisons with theoretical models. In the following, I will therefore discuss other experimental evidence that is model independent and show that the quasifission process plays an large role in the processes that occur in heavy-ion bombardments.
Figure 6. Fission angular distributions (left panel) and the saddle point shapes $J_0/J_{\text{eff}}$ (right panel) derived from the data for the reactions $^{16}\text{O}+^{238}\text{U}$ and $^{32}\text{S}+^{208}\text{Pb}$ are shown and compared to rotating liquid drop model predictions (dashed curves).

4. Total kinetic energy

Much of the early work on quasifission was derived from experiments performed at GSI in the early and middle 1980’s [28, 29, 30, 13, 31]. The availability of Pb and U beams at energies extending up to almost twice above Coulomb barrier allowed for measurements in inverse kinematics where both fragments were detected in forward angles and fragment mass,

Figure 7. Left panel: The total kinetic energy of fission fragments from a wide range of nuclei are shown as a function of $Z^2/A^{1/3}$ of the fissioning system. The solid symbols are from systems studied in Refs. [29, 13]. Right panel: Cross section contours of the total kinetic energy are shown as a function of fragment mass for $^{238}\text{U}+^{27}\text{Al}$ (upper panel) and $^{238}\text{U}+^{48}\text{Ca}$ (lower panel) at $E_{\text{beam}} = 6$ MeV/u [29]. The dashed curves represent the Coulomb repulsion between the fragments at scission consistent with the Viola systematics at symmetry.
total kinetic energy and scattering angle could be derived from the recorded position and time of arrival of the fragments in large-area fast gas-filled detectors. These measurements provided an extended data set which I would like to discuss here.

First, let’s examine the total kinetic energy of the fission-like fragments seen in these studies. Viola et al. [15] have performed a comprehensive study of fission fragment kinetic energy for a wide range of systems and found that data for symmetric mass splits may be described by a simple analytic expression that essentially reflect the Coulomb repulsion between the deformed fragments at scission. In the GSI studies, this data set was extended substantially toward heavier systems and found to agree well with the Viola systematics. For the heavier targets used in these experiments, the main contribution to the fission-like fragments clearly come from quasifission. This is illustrated in the left panel of Fig. 7 where the open symbols were from Viola’s previous systematics and the solid points were obtained in the GSI work, mostly representing quasifission. Also the fragment mass dependence of the total kinetic energy is well described on the basis of the Coulomb repulsion, both for mostly fusion-fission reactions in $^{238}\text{U}+^{27}\text{Al}$ (right top panel in Fig. 7 as well as for the wide fragment mass distribution seen in $^{238}\text{U}+^{48}\text{Ca}$ (right, lower panel). It is therefore important to realize that the total kinetic energy does not separate compound fission and quasifission.

It is also interesting to examine the total kinetic energy as a function of the excitation energy of the system. This is done in Fig. 8 where the total kinetic energy, $E_K$, is shown as a function of the excitation energy, $E^+$, at the scission point (see Eq. 2 for definition) for the reactions studied with $^{238}\text{U}$ beams in Ref. [13]. It is interesting to note that the increase in total available energy associated with increased beam energy is not reflected in the total kinetic of the final fragments indicating that the motion toward scission is almost completely damped, or in other words, that the pre-scission kinetic energy appears to be small. A further analysis of the pre-scission kinetic energy may be found in Ref. [13].

Figure 8. The average kinetic energy for symmetric mass division observed in $^{238}\text{U}$ bombardment of various targets is given as a function of the excitation energy at the scission point, $E^+$ [13].
5. Mass-angle correlations

Niels Bohr’s [32] compound nucleus model is based on the assumption that formation and decay of the intermediate system are decoupled and that only certain quantities, such as total angular momentum, linear momentum and energy are conserved during the process. In fission decays of the compound nucleus it is thus required that there is no preference for forward or backward emission of fragments, because any information about the initial direction and mass asymmetry is lost during the intermediate compound nucleus step. Experimentally this means that, in a two-body exit channel, such as fission, the mass distribution must be symmetric (about $A_s$) for any center of mass angle and that the angular distributions must be forward-backward symmetric (about $\theta_{cm}=90^\circ$) for any fragment mass. Note that for two-body exit channels, one of these requirements implies the other. These conditions may also be expressed as,

$$\sigma_f(A, \theta_{cm}) = \sigma_f(A, \pi - \theta_{cm}) = \sigma_f(A_{cn} - A, \theta_{cm}) = \sigma_f(A_{cn} - A, \pi - \theta_{cm}),$$

(8)

where $A_f$ is the fragment mass, $A_{cn}$ is the total mass, and $\theta_{cm}$ is the emission angle in the center-of-mass system. In contrast, for any two-body exit channel one has the less stringent requirement

$$\sigma_f(A, \theta_{cm}) = \sigma_f(A_{cn} - A, \pi - \theta_{cm}).$$

(9)

Figure 9. Double differential cross section contours are shown for $^{238}\text{U} + ^{16}\text{O}$, $^{238}\text{U} + ^{26}\text{Mg}$, and $^{238}\text{U} + ^{32}\text{S}$ in the left, middle and right columns, respectively. The top, middle, and bottom rows correspond to $^{238}\text{U}$-beam energies of, 7.5, 6.7, and 5.4 MeV/u, respectively [13].
In Fig. 9 the complete mass-angle distributions for reactions of \(\text{^{238}U + ^{16}O}\), \(\text{^{26}Mg}\), and \(\text{^{32}S}\) are shown [13]. Although very symmetric cross section distribution are seen at all three energies for the \(\text{^{238}U + ^{16}O}\) and probably also for \(\text{^{238}U + ^{26}Mg}\) it is clear that the \(\text{^{238}U + ^{32}S}\) mass-angle distribution is skewed. At the lowest energy, two different fission components may even be visible. One may also note that the center-of mass energy that we used in the fission anisotropy study of \(\text{^{32}S + ^{238}U}\) [20] (\(E_{\text{cm}} = 192\) MeV) is only about 10\% lower than the one corresponding to 7.5 MeV/u \(\text{^{238}U}\) beams. Mass-angle distributions for \(\text{^{32}S + ^{232}Th}\) has also recently been measured at beam energies between \(E_{\text{beam}} = 143-168\) MeV at Australian National University [22]. In this work was was concluded that two different quasifission components contribute to the cross section, possibly associated with multi-turn rotation of the intermediate system.

For even heavier targets, it becomes even more clear that a fast, dynamical process is responsible for the fission-like products as shown in Fig. 10. Although these distributions clearly show that the main part of the fission-life cross section comes from quasi-fission, one cannot rule out that a fraction of the cross section leads to the formation of a compound nucleus which may decay by fission or particle-gamma emission.

Let me emphasize that many studies of fission fragment mass measurements fail to expose the asymmetry in the mass-angle correlation by concentrating on the angular region close to \(\theta_{\text{cm}} = 90^\circ\). Likewise, many fission angular distribution studies do not measure the fragment masses. Because of the two-body nature of quasi-fission, such measurements will always result in mass-symmetric distributions and thus miss this important clue that a short time-scale process.

![Figure 10. Same as Fig. 9 but for \(\text{^{35}Cl, ^{40}Ca, and ^{nat}Zn}\) targets [13].](image)
is observed.

6. Quasi-fission and synthesis of super-heavy elements
In recent years, incontrovertible evidence for the synthesis of heavy elements formed via the so-called “hot fusion” reaction has been found at Dubna and elsewhere [33]. The formation of these elements occurs by several successive small branches of neutron evaporation in competition with fission. So it is clear that some fusion-fission cross section must be present underneath the often much larger quasi-fission branch. From the fission measurements, one cannot obtain accurate estimates of this fusion-fission branch, but it is possible to set upper limits under the assumption that it follows the expected behavior in terms of width of the mass distribution and angular anisotropy. One such analysis was carried out by us for reactions leading to the $^{214}$Th system [34] using three different entrance channels. For the most mass symmetric system $^{60}$Ni+$^{154}$Sm we found that only of the order of 10-15% of the fission-like cross section could come from fusion-fission - however, it could also be substantially smaller. Data analysis of this type has not often been employed, but it may help provide some limits on fission barrier heights in systems populated in the “hot fusion” reactions that are successfully being used for the synthesis of the heaviest elements.

7. Time-scale of quasifission processes
The most striking signature that distinguishes quasifission from normal fusion-fission is the asymmetry in the mass-angle distributions as discussed above. This asymmetry arises because the intermediate system rotates only a finite angle and the mass drift toward symmetry is disrupted when re-separation occurs. In principle, these distributions therefore carry information about the time scale of the process under assumptions about the angular momenta involved and the moment of inertia of the rotating complex. In our work on the GSI data [29], we developed a simple prescription for such an analysis in which the rotation angle $\Delta \theta$ of the intermediate system is derived from the mass-angle distributions using the relation

$$\Delta \theta = \pi - \theta_i - \theta_f - \theta,$$

where $\theta$ is the observed scattering angle and $\theta_i$ and $\theta_f$ are half of the calculated Coulomb deflection angle in the entrance and exit channels. The reaction time may be written as

$$t_{reac} = \Delta \theta / \omega,$$

where $\omega = l/J$ is the angular velocity, $l$ is the angular momentum of the complex and $J$ its moment of inertia. Of course, although $l$ is conserved, one expects that both $\omega$ and $J$ varies during the rotation because of shape changes caused mainly by the mass transfer from the light to the heavy reaction partner. In the spirit of simplicity, we take two different estimates for $J$, either an average of the entrance and exit channel rigid moment of inertia assuming touching rigid spheres, $J = 0.5(J_{in} + J_{out})$ or a value of $J = 1.4 J_{sph}$ i.e. 40% larger than the rigid sphere value for the total system. In order to perform this analysis, the observed quasifission cross section was divided into three equal size bins and for each bin the rotation angle $\Delta \theta$ and the associated average mass and angular momentum of the system was evaluated as illustrated in Fig. 11 (see Ref. [29] for details).

For each bin, one may compute the average mass transfer $\Delta A$ toward symmetry and normalize relative to the maximum drift reaching symmetry, $\Delta A_{max} = |A_p - A_s|$, where $A_p$ is the projectile mass, see Fig 11. Using the data from 6.0 MeV $^{238}$U induced reactions of Ref. [29] to extract the quantities $\Delta A/\Delta A_{max}$ and $(l)$ and correlating them with the angle of rotation, $\Delta \theta$, and
Figure 11. This figure illustrates the correspondence between mass drift toward symmetry and the angle of rotation of the intermediate complex in quasifission. The final emission angles, $\theta_1, \theta_2, \theta_3$ for the three different mass regions are shown. [29].

reaction time, $t_{\text{reac}}$, we see that a tight correlation is obtained for $\Delta A/\Delta A_{\text{max}}$ plotted vs. $t_{\text{reac}}$. This dependence is well described as an exponential mass drift toward symmetry given by

$$\frac{\Delta A}{\Delta A_{\text{max}}} = 1 - \exp[-(t - t_0)/\tau],$$

(12)

where $t_0$ is a short time offset of about $t_0 \sim (1 - 2) \times 10^{-21}$ s before the mass drift starts - maybe to allow the connecting window between the two reaction partners to open, and $\tau$ is a characteristic mass drift time of $\tau = 5.3 \times 10^{-21}$ s, which appears to fit the data.

This analysis method was also applied to the larger data set from GSI [13] where it was shown that the mass relaxation time of $\tau \approx 5 \times 10^{-21}$ sec also holds for a wide range of beam energies, $E_{\text{beam}}=4.6-7.5$ MeV/u and therefore appears to be temperature independent.

Theoretically, it is of interest to ask whether this mass drift rate is consistent with theoretical estimates. Such estimates may be based on the assumption of an over-damped motion by balancing the driving force $F_c = dU/d\langle A \rangle = -k(\langle A \rangle - A_s)$ in the mass-asymmetry potential, Eq. 1 with dissipative force, $F_d = m^{-1}d\langle A \rangle/dt$, that acts in the opposite direction. Solving for the time dependence one finds the observed dependence given in Eq. 12, with

$$\tau = \frac{1}{mk},$$

(13)

where $m$ is the mobility and $k$ is the restoring force constant. A theoretical estimate of the mobility may be obtained on the basis of the one-body dissipation mechanism, which arises from particle collisions with the moving nuclear surface [35]. In the application to quasi-fission, the moving surfaces are represented by the increase of the lighter fragment and the shrinking the heavier partner that occurs during the mass transfer phase. Applying this mechanism to the simple geometry of two touching spheres of un-equal size one arrives at the following expression for the inverse mobility [36]

$$\frac{1}{m} = \frac{3}{8} \left( \frac{\pi}{3} \right)^{1/3} \left[ \frac{1}{A_1^{2/3}} + \frac{1}{A_2^{2/3}} \right] \hbar$$

(14)
Figure 12. The correlations between the normalized mass drift, angle of rotation, angular momentum, and reaction times for quasifission products formed by 6 MeV/u and 5.4 MeV/u indicated in figure) $^{238}$U-beams on various targets are shown [29].

or numerically

$$\frac{1}{m} = 2.5 \times 10^{-22} \left[ \frac{1}{A_1^{1/3}} + \frac{1}{A_2^{2/3}} \right] \text{[MeVsec]}. \quad (15)$$

Using a mass asymmetry restoring force coefficient of $k=0.04$ [MeV/u$^2$] this results in a characteristic mass relaxation time of $\tau=4.6 \times 10^{-21}$ sec for the $^{48}$Ca+$^{238}$U system in the entrance channel and $\tau=6.4 \times 10^{-21}$ sec at mass symmetry, which is a very good agreement with the experimental value of $\tau_{exp}=5.3 \times 10^{-21}$ sec. Here it should be noted that the dissipation associated with particles crossing the neck that is formed between the two interacting partners has been ignored; see Ref. [13] for further discussion of the validity of this assumption.

From the study of the mass drift and rotation of the intermediate complex in quasifission, we thus find a surprisingly good agreement with the expected energy dissipation strength and lack of temperature dependence predicted by the one-body wall dissipation mechanism. It is, however, clear that this analysis of the data is rather crude and prone to some uncertainty in the derived reaction times. Maybe a direct comparison between the observed mass-angle distribution with those predicted by modern reaction mechanism calculations [37] could provide more stringent tests of the assumed dissipation mechanism.
8. Summary
Even after seventy years of experimental study and theoretical developments, the fission process continues to reveal new and intriguing properties. Both normal compound fission and the more dynamical quasi-fission reaction probe many aspects of nuclear physics from the nuclear structure properties, that gives rise to the shell structure that modify the potential energy landscape, as well as many aspects of the dynamics associated with the drastic re-arrangement of the nuclear shape. In this talk, I have attempted to review the early studies of quasi-fission with emphasis on the experimental signatures for the process. Although much of the early thinking were guided by theories developed by W. Swiatecki and Bjørnholm [38], W. Nörenberg [39], H. Feldmeier [40] and others, I have not attempted to review the theoretical work in this talk but concentrated on the experimental signatures.

We have seen that there are a number of ways, in which quasi-fission distinguishes itself from the standard fusion-fission process. These are all associated with the shorter time scales of the quasifission process and the fact that the intermediate shapes, that the system assumes during the process, do not include the equilibrated compound nucleus captured behind a fission barrier. As we have seen, this has consequences for the fission anisotropies, the width of the mass distributions and most importantly, the mass-angle distributions that directly reveal the short time-scale, dynamical nature of the quasifission process. Indeed, for the heavier systems this feature allows us to study the time evolution of the mass transfer and compare the time scales with simple theoretical estimates, but much work remains to be done experimentally to study the process in greater detail and to develop even more sophisticated theoretical models describing the process.

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