Experimental Signatures of Quasifission Reactions

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The features of fission-fragment angular distributions are shown to be sensitive to the occurrence of nonequilibrium processes such as quasifission. This conclusion is based on a quantitative analysis of the angular distributions of near-symmetric masses produced in ${}^{16}\text{O} + {}^{238}\text{U}$ and ${}^{32}\text{S} + {}^{208}\text{Pb}$ reactions. The results are discussed in terms of the "extra-push" model of Swiatecki.

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Fissionlike fragments have been observed in reactions for which the composite system has no barrier against fission.^{1,2} In view of the fact that a long-lived compound nucleus has clearly not been formed in this case, the mechanism responsible for these fragments has been termed "fast fission." A similar nonequilibrium fission process has been predicted to occur in some heavy-ion reactions even when the compound nucleus is expected to have a well-developed fission barrier. This latter process, called "quasifission," has been discussed in the dynamical model of heavy-ion collisions of Swiatecki³ and is predicted to occur as a result of dissipation and mass transfer in the entrance channel, which prevent the formation of an equilibrated compound nucleus inside the fission barrier, but which lead rather to a more deformed composite system which subsequently decays into fissionlike fragments.³⁻⁶

Although quasifission reactions are well defined in Swiatecki's extra-push model,³ previously used experimental signatures for this process have been only indirectly related to this definition either through the angular momenta contributing to the reaction or through the time scale of the reaction. We have found that a quantitative analysis of fragment angular distributions leads to a clear and very sensitive discrimination between quasifission and compound-nucleus fission reactions. This method relies on the fact that the fragments observed in quasifission reactions do not, by definition, result from fission decay over the compound-nucleus fission barrier. The angular distribution in normal compound-nucleus fission depends on the nuclear shape at the fission saddle point in such a way that the anisotropy increases monotonically with deformation. Abnormally large fission-fragment anisotropies may, therefore, be taken to indicate that a large fraction of the fissionlike fragments stem from quasifission reactions which originate from extended shapes outside the fission barrier.

The reactions ${}^{16}O + {}^{238}U$ and ${}^{32}S + {}^{208}Pb$ were chosen for this study as the compound systems. ²⁵⁴Fm and ²⁴⁰Cf, have essentially identical fission barrier shapes according to the rotating liquid drop model,⁷ the fissility parameter being x= 0.84 in both cases. Beams of 16 O at 90, 110, 130, and 148 MeV and ³²S beams of 185, 198, 205, 219, and 225 MeV from the Argonne National Laboratory superconducting linac booster were used to bombard targets of ²³⁸U and ²⁰⁸Pb, respectively. Reaction products with masses close to those expected for symmetric fission were detected in a 50-mm² solid-state telescope (8- μ m-thick ΔE) over the angular range of $\theta_{1ab} = 7^{\circ} - 70^{\circ}$ and in two 100-mm² Si detectors over the range $\theta_{1ab} = 70^{\circ}$ -174°. Fragment mass determinations were made by measuring the time of flight over the 18 cm distance between the target and the detectors. A time resolution of ~ 200 ps was achieved with use of the time structure of the beam. Absolute cross sections were obtained by normalizing to Rutherford scattering observed in a monitor detector positioned at $\theta_{1ab} = 15^{\circ}$.

The fragment angular distributions, some of which are presented in Fig. 1, have been analyzed within standard fission theory. In this theory,⁸ fission fragments are assumed to be emitted along the direction of the nuclear symmetry axis at the fission saddle point. This direction is described by symmetric-top wave functions \mathfrak{D}_{MK}^{I} in terms of the total spin *I*, projection *M* on the beam axis, and the projection *K* on the nuclear symmetry axis. For reactions with spin-zero target and projectile, M = 0 and summing over *K* and *I* leads to the expression for fission angular distribution:

$$W(\theta) = \sum_{I=0}^{\infty} \sigma(I) \sum_{K=-I}^{+I} \frac{1}{2} (2I+1) \rho(K) |\mathbf{D}_{0K}^{I}(\theta)|^{2}.$$
(1)



FIG. 1. Fragment angular distributions for the ¹⁶O + ²³⁸U and ³²S + ²⁰⁸Pb reactions at two beam energies for which the mean angular momenta of the two systems are nearly identical (see Table I). The solid curves represent fits to the data from which the total fission cross sections and the saddle-point deformations are deduced. The dashed curves represent angular distributions expected for the normal fission decay of ²⁵⁴Fm and ²⁴⁰Cf over the fission barrier predicted by the rotating liquid drop model (Ref. 7).

This equation applies only to cases where the competing decay modes are negligible, as is the case for the present systems. The functional form of the spin-dependent cross section $\sigma(I)$ is given by $\sigma(I) = \pi \lambda^2 (2I + 1)T(I)$, where the dependence of T(I) on I was taken from transmission coefficients derived from an optical-model calculation, which reproduce the elastic scattering data. The optical-model parameters used were as follows: for ${}^{16}\text{O} + {}^{238}\text{U}$; V = 42.9 MeV, W = 66.9

MeV, $r_0 = r_0' = 1.22$ fm, a = a' = 0.6 fm; and for ${}^{32}\text{S} + {}^{208}\text{Pb}$, V = 40 MeV, W = 40 MeV, $r_0 = r_0' = 1.17$ fm, a = a' = 0.54 fm. The value of *I* for which T(I) = 0.5 was adjusted to reproduce the measured fission cross section under the assumption that the fission cross section is associated with the lower angular momenta and that the high-angularmomentum tail has the same shape as that for the total reaction cross section. Other reasonable choices of the functional form of $\sigma(I)$ do not influence the qualitative conclusions of the analysis. The distribution of *K* values is assumed to be Gaussian,⁹ the variance K_0^2 being related to the nuclear shape at the fission saddle point by the relation

$$K_0^2 = (T/\hbar^2) \boldsymbol{y}_{eff},$$
 (2)

where

$$\boldsymbol{\vartheta}_{\mathrm{eff}}^{-1} = \boldsymbol{\vartheta}_{\parallel}^{-1} - \boldsymbol{\vartheta}_{\perp}^{-1}, \qquad (3)$$

T is the nuclear temperature at the fission saddle point, and $\boldsymbol{y}_{\parallel}$ and \boldsymbol{y}_{\perp} are moments of inertia for rotations parallel to and perpendicular to the nuclear symmetry axis, respectively.

The measured angular distributions were fitted by varying the parameter K_0^2 . The resulting fits are shown as solid curves in Fig. 1 and the parameters are listed in Table I. The values of g_0/g_{eff} (g_0 is the rigid-sphere moment of inertia) deduced from these data are plotted in Fig. 2 as a function of the mean square spin $\langle I^2 \rangle$ and compared with the predictions of the rotating liquid drop model (RLDM).⁷ For reactions proceeding through an equilibrated compound nucleus, we expect good agreement with the RLDM predictions since this model has been shown to work in numerous experiments¹⁰⁻¹³ with ¹⁶O or lighter projectiles. The results for the ¹⁶O + ²³⁸U reaction

Reaction	E beam	σ _{sym} (mb)	$\sigma_{\!R}^{ m OM}$ (mb)	W (0°)/W(90°)	$\langle I \rangle$ (\hbar)	$\langle I^2 angle \ \langle \hbar^2 angle$	T (MeV) ^a	g ₀ / g _{eff}
¹⁶ O + ²³⁸ U	90	185	440	1.87	17	400	1.15	1.47
	110	1030	1340	2.22	32	1225	1.36	0.74
	130	1640	1960	2.24	45	2310	1.52	0.44
	148	1710	2370	2.50	53	3240	1.65	0.42
³² S + ²⁰⁸ Pb	185	190	470	3.24	32	1320	1.23	1.30
	198	375	770	3.59	39	1910	1.35	1.10
	205	465	910	3.76	42	2200	1.41	1.06
	219	700	1170	4.01	52	319 0	1.51	0.86
	225	780	1270	4.14	55	3590	1.55	0.83

TABLE I. Parameters used in angular distribution calculations.

^aNuclear temperature at the fission saddle point.



FIG. 2. Values of $\mathcal{G}_0/\mathcal{G}_{eff}$ plotted as a function of the mean squared spin of the system, for the reactions ${}^{16}\text{O} + {}^{238}\text{U}$ and ${}^{32}\text{S} + {}^{208}\text{Pb}$. The dashed curve represents the theoretical prediction of the saddle-point shape based on the rotating liquid drop model (Ref. 7) which applies to both systems.

are in good agreement with the RLDM predictions, indicating that this reaction does indeed proceed via an equilibrated compound nucleus formed inside the fission saddle point. The predicted contraction of the fission saddle point with increasing angular momenta is observed in the data as decreasing values of θ_0/θ_{eff} . The large deviation of the lowest energy point is most likely associated with an inadequate description of the spin distribution in near-barrier heavy-ion fusion.

If an equilibrated ²⁴⁰Cf compound nucleus had been formed in the ${}^{32}S + {}^{208}Pb$ reaction we would expect angular distributions which are in agreement with the RLDM predictions as in the ¹⁶O +²³⁸U case. The ³²S + ²⁰⁸Pb data are, however, in clear disagreement with this expectation, suggesting that a major fraction of this cross section is associated with reactions which fail to produce the ²⁴⁰Cf compound system. Such processes clearly match Swiatecki's definition of quasifission reactions and we therefore conclude that a substantial fraction of the near-symmetric fragments observed in the ${}^{32}S + {}^{208}Pb$ reaction are associated with the quasifission process. We note here the sensitivity of fission angular distributions in distinguishing fusion-fission and quasifission processes since the fragments produced in the ³²S + ²⁰⁸Pb reaction cannot be distinguished from normal fission fragments on the basis of their masses or energies alone.

Although the present results for ${}^{32}S + {}^{208}Pb$, which indicate a strong inhibition of compoundnucleus formation, are in qualitative agreement with the extra-extra-push model,⁶ a quantitative analysis using this model predicts that the passage over the true saddle point to fusion at low angular momenta is inhibited only for projectiles such as ⁵⁶Fe and heavier. The parameters in the model, which lead to this conclusion, were, however, determined from an analysis of experimental data¹⁴ under the assumption that it is the angular dependence of the fragment mass distributions which distinguishes between compound fission and quasifission. The former process leads to angle-independent mass distributions. whereas the quasifission process was assumed to have angle-dependent fragment masses. This assumption requires, however, that the mass equilibration time and the rotational period both be shorter than or equal to the reaction time. The present data show that this is not the case, the $^{32}\mathrm{S}+^{208}\mathrm{Pb}$ reaction showing angle-independent mass distributions while having a manifestly nonequilibrium behavior of the fragment angular distribution.

These results can be reconciled in a quantitative way with the extra-extra-push model by changing the parameter values obtained in the analysis of Ref. 6. In particular, reducing the value of the parameter x_{cliff} from 0.84 to 0.78 produces the required inhibition of compoundnucleus formation without upsetting the agreement with the cross-section data of Ref. 14. Such a modification does cause some difficulty in accounting for the observation¹⁵⁻¹⁸ of evaporation residues in ${}^{40}Ar + {}^{208}Pb$, ${}^{50}Ti + {}^{208}Pb$, ${}^{54}Cr + {}^{209}Bi$, and most recently ⁵⁶Fe + ²⁰⁹Bi. These reactions have, however, all been carried out at energies in the vicinity of the Coulomb barrier where dynamic distortions,¹⁹ zero-point motions,²⁰ and quantum mechanical tunneling effects are known to be of major importance. None of these effects are included in the extra-push model and these results therefore emphasize the need for even more sophisticated models to provide an understanding of the complexity of heavy-ion reactions. This adjustment of the x_{cliff} parameter does, however, significantly reduce the predicted possibilities⁶ for producing super-heavy elements in the Z=112-116 range by heavy-ion fusion reactions although the recent observation^{17,18} of elements 107 and 109 underscores the importance of special dynamical and quantum mechanical effects in the near-barrier fusion reactions.

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Identification of E 2 Strength Distribution in 65 Cu by the (e, p_0) Reaction

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Double-differential cross sections for the reaction 65 Cu(e, p_0)⁶⁴Ni_{g.s.} were measured at eleven laboratory angles ranging from 42° to 138° with incident electron energies from 13 to 28 MeV. These have been decomposed into E1 and E2 components by use of a resonance model. Besides the large E1 cross section, the E2 strength is clearly separated at $E_x = 14.9$ MeV with the width of 5.1 MeV corresponding to the isoscalar giant quadrupole resonance.

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Giant multipole resonances (GMR) other than E1 have been studied by various projectiles. Up to now most of the experiments have been carried out only for inclusive reactions. With such reactions, it is difficult to determine in a model-independent way the multipolarities of the GMR. Successful completion of (e, e'p) experiments concerning the determination of the multipolarities of the GMR is expected.¹ Yet at this stage no results have been reported. Concerning exclusive

reactions, several authors have already reported on the angular distributions of the emitted particles through electrodisintegration. The ¹⁶O(e, p_0) angular distribution has been measured by Schoch *et al.*,² which showed that the contributions due to the spin current are important in the angular distribution analysis. Their interest, however, concerns excitation energy region much higher than that of the GMR. Skopik, Asai, and Murphy³ have measured ⁵⁶Fe(e, α) angular dis-