Evidence for Long Formation Times of Near-Barrier Fusion Reactions

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High energy γ rays from the decay of the giant dipole resonance built on highly excited states in ¹⁶⁴Yb at $E_{ex} = 49$ MeV formed in two different reactions have been measured. While standard statistical model calculations can describe the γ -ray spectrum from the ¹⁶O + ¹⁴⁸Sm reaction they fail to reproduce the γ -ray spectra from the more symmetric reaction ⁶⁴Ni + ¹⁰⁰Mo. Simple model calculations which include particle evaporation and γ -ray decay during the formation process suggest that the observed differences may be related to a long fusion time in the more symmetric reaction.

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It is well established that dissipative effects influence the reaction dynamics of heavy-ion collisions. Swiatecki [1] developed a dissipative collision model which shows that in certain projectile-target combinations it is not enough just to pass the one-dimensional potential-energy barrier in order to form a compound system, but an "extra push" is required. Once the reactants pass inside a saddle point in the complex multidimensional configuration space, fusion is considered to have occurred. It is generally assumed that the system equilibrates rapidly and the subsequent decay can be described within the framework of the statistical model. However, recent data have cast doubts on the validity of this assumption. Neutron multiplicity measurements of the fusion of nearly symmetric systems near the Coulomb barrier have raised the possibility that the reaction dynamics influences the decay of the compound nucleus [2]. This was demonstrated by comparing the fusion of a symmetric system to that of a very asymmetric reaction, one which agreed with standard statistical model calculations. A different probe which is well suited to study dynamical effects in the fusion process is γ rays from the decay of the giant dipole resonance (GDR) built on highly excited states. It has been shown that γ rays from the GDR are emitted predominantly in the earliest steps of the compound nucleus decay [3] and that the shape of their spectrum is sensitive to the deformation of the nuclear system. Recently, the γ ray of the GDR was applied to study the time scale of quasifission reactions [4]. In this Letter we will show, for the first time, clear evidence from highenergy γ -ray measurements for entrance channel dependent effects in heavy-ion fusion. We suggest that this effect occurs because the fusion in the almost symmetric reaction $^{64}Ni + ^{100}Mo$ is sufficiently slow that the assumption of a fully equilibrated system during the first stages of decay is not valid. The fusion of the reference (asymmetric) reaction ${}^{16}O + {}^{148}Sm$ is fast enough so that the equilibrium statistical model gives an adequate description of decay data. The underlying physics which we invoke to account for the delay of the fusion process is closely related to that responsible for a number of dissipative effects on heavy-ion reactions, and the fission process.

The experiments were performed with the HHIRF tandem accelerator at Oak Ridge National Laboratory. Isotopically enriched self-supporting 148 Sm (3.05 mg/cm²) and 100 Mo (450 μ g/cm²) targets were bombarded with 82 MeV ¹⁶O and 232 MeV ⁶⁴Ni, respectively, forming the compound nucleus ¹⁶⁴Yb at an excitation energy of $E_{\rm ex}$ = 49 MeV. High-energy γ rays were measured with 76 BaF₂ detectors arranged in four arrays of 19 crystals each, positioned at 21° , 63° (two), and 117° and at a distance from the target of 57 cm and 77 cm for the backward and forward angle arrays, respectively. Neutron and γ -ray identification was achieved using time of flight. The γ -ray multiplicity of the events was recorded using 55 NaI detectors of the spin spectrometer [5]. The γ -ray energy deposited in the individual BaF₂ detectors was summed within each array and the total spectrum is the sum of the spectra of the four arrays. The effect of pileup on the resulting spectra was carefully examined. Realistic Monte Carlo simulations indicated a negligible pileup effect. This was confirmed experimentally by a different analysis in which we summed pulse heights in only the nearest-neighbor crystals around detectors registering a high-energy γ -ray hit. This reduces the solid angle covered by "one detector" by at least two without degrading the high-energy γ -ray response. The resulting spectra showed no significant differences compared to the array-summed spectra. We conclude that pileup does not distort the spectra. Cosmic rays were rejected using the event structure in the BaF_2 detectors and the spin spectrometer. Contributions from target contaminations such as carbon and oxygen were measured in a separate reaction ($^{64}Ni + {}^{12}C$). The properly normalized yield, as determined by characteristic γ -ray lines moni-

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FIG. 1. Calculated spin distributions corresponding to the fold gate of 5–10 applied to the γ -ray spectra for the ¹⁶O (\blacksquare) and ⁶⁴Ni (\Diamond) induced reactions.

tored with two Ge detectors, was then subtracted from the spectra.

In order to compare the γ -ray spectra from the two reactions it is important that they are gated by the same range of angular momenta. This was accomplished by gating on a γ -ray coincidence fold between 5 and 10 in the spin spectrometer. The present compound system has been extensively studied [6] and the spin distribution has been measured. The evaporation code EVAP [7] was used to calculate the γ -ray multiplicity and the response of the present arrangement of the spin spectrometer was simulated with GEANT3 [8]. The extracted simulation parameters had to be adjusted slightly in order to reproduce the fold distributions from both reactions simultaneously. Figure 1 shows the selected spins of the two reactions $[^{16}O$ (solid squares) and ^{64}Ni (open diamonds)] calculated with the measured spin distributions and conditions of the present experiment (γ -ray fold = 5– 10, trigger γ rays > 8 MeV). This fold gate was selected in order to discriminate against deep inelastic contributions on the low spin side and to include spins close or even beyond the maximum spin for the ¹⁶O reaction. In both reactions the fold gate selects spins between $10\hbar$ and $30\hbar$. However, the distribution from the ⁶⁴Ni reaction has a tail extending beyond $40\hbar$.

The γ -ray spectra for the ¹⁶O (a) and ⁶⁴Ni (b) induced reactions are shown in Fig. 2 as histograms. The solid lines correspond to calculations using the statistical model code CASCADE [9]. The initial spin distributions were taken from the EVAP simulations shown in Fig. 1, and the level density parameter was chosen to be A/10. The calculated γ -ray spectra were folded with the response function generated with GEANT3. The GDR parameters extracted from a fit to the ¹⁶O data were $S_1 = 33\%$, $E_1 = 12.2 \pm 0.2$ MeV, $\Gamma_1 = 4.9 \pm 0.5$ MeV, $S_2 = 67\%$, $E_2 = 15.7\pm0.5$ MeV, and $\Gamma_2 = 6.9\pm1.0$ MeV, consistent with previous measurements in this mass region and excitation energy [10]. However, it is obvious that these parameters cannot reproduce the γ -ray spectrum in the ⁶⁴Ni-induced reaction. The absolute normal-



FIG. 2. Comparison of the γ -ray spectra from ¹⁶⁴Yb following the reaction ¹⁶O + ¹⁴⁸Sm (a) and ⁶⁴Ni + ¹⁰⁰Mo (b) and (c). The solid curves are calculations using the code CASCADE. Part (c) shows the calculation which included contributions from the formation (dashed) and the compound nucleus (dot-dashed) decay.

ization of the calculations was performed using the fusion cross section [6], beam current, and response of the detectors and has an estimated error of ~ 20%. We also investigated the effect of the relatively large initial excitation energy spread due to the target thickness. We included this spread as the initial excitation energy spread in CASCADE and observed no significant change of the spectral shape. The shape of the spin distribution does not change sufficiently as a function of the target-induced excitation energy [6] to lead to any significant effect on the mapping of our fold gate to angular momentum, or to influence the absolute normalization significantly compared to the quoted uncertainty.

Figure 3(a) shows the quality of the fit of the ¹⁶O data (solid squares) and the large discrepancy for the ⁶⁴Ni data (open diamonds) in a linearized plot. To create this plot, the data as well as the calculations were divided by the same spectrum calculated using a constant dipole γ -ray strength function specified above.

The observation of an apparent excess of total γ -ray emission in the more symmetric system is consistent with the previously reported deficiency of neutrons [2], and also with an experiment measuring an excess of sum γ ray energy [11]. As mentioned before a similar effect was observed recently in the γ -ray decay of neutron deficient Th isotopes, where the more symmetrically formed reaction could only be described by including contributions from quasifission reactions [4].

Before the observed differences can be attributed to different formation properties, other possibilities have to be excluded. The main concern is that the 64 Ni data contain contributions from mechanisms other than fusion-evaporation reactions. Measurements on similar systems



FIG. 3. Linearized plots of the γ -ray spectra from the ¹⁶O (**■**) and ⁶⁴Ni (\Diamond) induced reactions. The data as well as the calculations (solid lines) were divided by the same constant strength function. The calculation shown in (a) is for the ¹⁶O data; the calculation for the ⁶⁴Ni data is essentially identical. The total calculated spectrum in (b) is a sum of contributions from the formation (dashed) and the compound nucleus (dot-dashed) decay.

[12], as well as calculations using the code HICOL [13], which is based on Swiatecki's dissipative collision model, show that the deep inelastic cross section is approximately equal to the fusion cross section, so that it cannot be easily disregarded. However, according to HICOL, the fragment excitation energies produced in these deep inelastic reactions are extremely low (< 12 MeV). In addition the spin transfer to the reactants is also predicted to be very low (< $2.5\hbar$). HICOL reproduces results of other deep inelastic measurements quite well. Another simple estimate based on the total kinetic energy systematics and an excitation energy sharing proportional to the mass of the fragments yields 17 and 12 MeV for ¹⁰⁰Mo and ⁶⁴Ni, respectively. We calculated the γ -ray decay of reaction fragments with their properties using CASCADE. We assumed a uniform excitation energy distribution between 16 and 26 MeV and a triangular spin distribution between 0 and $10\hbar$. We found that the contributions to the γ -ray spectra are negligible (< 1% above 9 MeV). In addition, the slope of these calculated spectra—in agreement with experimental data [14]—is much steeper and certainly cannot account for the observed discrepancies. A calculation based on the HICOL results would yield an even smaller contribution. Another possible contaminant is fission. At the present excitation energy no fission has been observed [6,12], and the statistical model calculations predict extremely small cross sections ~ 0.2% [6]. Calculations were performed with a modified version of CASCADE that follows the decay of the fission fragments [4]. Even with a reduction of the fission barrier by a factor of 0.6, which yielded a fission cross section of 11% of the fusion cross section, the contributions of γ rays following fission above 9 MeV were < 1.5% which could not account for the discrepancies in the data. One difference that also has to be considered is the additional high spin contributions shown in Fig. 1 for the 64 Ni reaction. Recently it has been reported that the GDR parameters are a strong function of spin [15], so it might be conceivable that the change of the GDR parameters at high spins could account for the observed spectral shape. CASCADE calculations show, however, that it is not possible with two sets of GDR parameters for low and high spins to fit both data sets consistently. It was possible to fit the ⁶⁴Ni data with extremely large GDR widths, especially for the lower GDR component, $E_1 = 11.9\pm0.2$ MeV, Γ_1 = 8.5 ± 0.5 MeV, $E_2 = 16.0\pm0.5$ MeV, and $\Gamma_2 = 9.0\pm1.0$ MeV. The physical significance of such GDR parameters in this fit to the ⁶⁴Ni-induced data is not obvious, but they clearly highlight the differences between the ⁶⁴Ni and ¹⁶O data.

Finally, we explore the possibility that differences in the formation process of the two reactions could produce the different spectral shapes. According to the semiclassical dissipative collision code HICOL, the ¹⁶O-induced reaction reaches equilibrium quite rapidly ($\sim 10^{-21}$ s) in the shape and thermal energy degrees of freedom tracked by the code. The calculated approach to equilibrium is much slower ($\sim 10^{-20}$) for the ⁶⁴Ni reaction. This "formation time" for the ⁶⁴Ni + ¹⁰⁰Mo is, in fact, comparable to the mean time for neutron evaporation. If neutrons and γ rays are emitted during this prolonged formation time, a modification of the resulting emission spectra compared to those measured with ¹⁶O + ¹⁴⁸Sm is not surprising.

To establish a qualitative idea of this effect which might be expected, we have attempted to model this nonequilibrium decay picture using a stepwise application of the equilibrium statistical model in the form of the code CASCADE. According to calculations with HICOL, key parameters (thermal energy, composite system shape, etc.) of the statistical model are changing during the formation time. We treat this by dividing the formation time into time steps, using fixed statistical model parameters obtained from the appropriate time averaged HICOL result for each step, allowing decays to occur within the time step, and obtaining the input population distributions for each step from the results of the preceding steps. The final step is a conventional equilibrium statistical calculation with decay proceeding without a time cutoff. For the present purposes, the simplest (i.e., two-step) implementation of this scheme is adequate as an illustration. We take the mean excitation energy (30 MeV) and deformation parameters from the HICOL calculation. The GDR parameters derived from these HICOL results are $E_1 = 9.2$ MeV and $E_2 = 18.0$ MeV. The corresponding widths are estimated as $\Gamma_1 =$ 3.0 MeV and $\Gamma_2 = 6.0$ MeV. A level density parameter of A/15 was chosen, guided by level densities for superdeformed shapes [16]. The first (formation) step lasts for $\sim 2 \times 10^{-20}$ s. A rigorous extraction of the uncertainty in this formation time is difficult; we estimate a value of $^{+2}_{-1} \times 10^{-20}$ s. The resulting populations in excitation energy and spin were taken as input for the next (fully equilibrated) step, which was treated within the regular statistical model using the standard parameters mentioned earlier. Results of these calculations are shown in Figs. 2(c) and 3(b) in comparison with the ⁶⁴Ni data. The total γ -ray spectrum (solid) is a sum of two contributions: γ rays emitted during the formation time (dashed) and γ rays from the compound nuclei (dot-dashed). There are still some discrepancies between the data and the calculation including the formation time effect [Fig. 3(b)]. It should be remembered, however, that the present incorporation of the formation time in the statistical model is oversimplified, since the whole formation process is treated within only one step. In addition, some of the model parameters (for example, the level densities) are not even known under these circumstances and can only be estimated.

The main difference in the spectral shape between the regular statistical model and including the formation time effect is not additional γ rays from the formation phase, as can be seen in Fig. 2(c). The main effect depends on the fact that the branching ratio for emission of high-energy γ rays (i.e., E_{γ} near the GDR peak) is a strongly increasing function of increasing excitation energy. The reduced effective excitation energy during the formation time therefore results in a reduced high energy γ -ray yield during the initial step. The cooling of the compound nucleus due to particle evaporation during the formation stage results in a lower mean energy and, hence, a modification of the γ -ray spectrum resulting from the compound system after full equilibration. It is worth noting that those γ rays emitted during the formation stage sample a quite different dipole strength function from that of the compound system. From Fig. 2(c) it can be seen that this contribution could lead to the very large widths obtained from the simple statistical model analysis of the ⁶⁴Ni data described earlier. We suggest that this delay in fusion in the symmetric reaction channel arises from the same underlying physics responsible for the similar delay in fission, the extra-push effect seen in fusion excitation functions, and for effects in quasifission [4].

In conclusion, we observed large differences in the γ -ray spectrum following the decay of the compound nucleus ¹⁶⁴Yb when formed with two reactions. The observed

effects may be related to a long fusion time in the reaction 64 Ni + 100 Mo.

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