Predominant Time Scales in Fission Processes in Reactions of S, Ti and Ni with W: Zeptosecond versus Attosecond

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The inhibition of fusion by quasifission is crucial in limiting the formation of superheavy elements in collisions of heavy nuclei. Time scales of $\sim 10^{-18}$ s inferred for fissionlike events from recent crystal blocking measurements were interpreted to show either that quasifission itself is slower than previously believed, or that the fraction of slow fusion-fission is higher than expected. New measurements of mass-angle distributions for ^{48}Ti and ^{64}Ni bombarding W targets show that in these reactions quasifission is the dominant process, typically occurring before the system formed after contact has made a single rotation, corresponding to time scales of $\leq 10^{-20}$ s.

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Amongst the 118 known elements, about 90 are found naturally on Earth. The heaviest elements [1], known as superheavy elements (SHE), can only be synthesized by the fusion of two heavy nuclei, using heavy-ion accelerators. For the SHE to be formed, the two nuclei that come into contact must evolve from a dinuclear shape to a compact equilibrated excited nucleus called a compound nucleus (CN). However, the large Coulomb energy often causes premature breakup before a compact CN is formed, a process known as quasifission, which inhibits the formation of SHE. Even if a CN is formed, the SHE yield is expected to be severely suppressed by fission of the CN itself, before it reaches its ground state.

Understanding the dynamics of, and competition between, quasifission and fusion will lead to more reliable predictions of opportunities to form a wider range of SHE isotopes in nuclear fusion reactions. The presence of fusion-fission is a signature that fusion has occurred, and a CN has been formed. However, the characteristics of the products of fusion-fission and quasifission show considerable overlap. Thus it is difficult to unambiguously separate quasifission and fusion-fission, despite ingenious fitting procedures [2]. Quasifission has generally been understood [3] to occur on short time scales of $\sim 10^{-20}$ s. Fusion-fission typically occurs on longer time scales, from $\sim 10^{-20}$ s to $\sim 10^{-16}$ s. Measurement of fission times can thus give a definitive signature of fusion-fission. Three main methods have been used to infer time scales, each with a different range of sensitivity.

(i) The neutron-clock method [4] counts the neutrons emitted before breakup into two fragments (scission). In principle, it is sensitive to time scales from 10^{-22} s to 10^{-16} s. However, accurate interpretation of the measurements in principle requires *a priori* knowledge of the dynamics, as well as neutron evaporation lifetimes [4]. Different assumptions [4,5] can lead to a factor of 10 differences ($\sim 10^{-20}$ s to $\sim 10^{-19}$ s) in deduced quasifission time scales [4,5], though neutron kinetic energies favor the shorter times [6].

(ii) The mass-angle distribution of the fragments measures the sticking time of the dinuclear system [3,7]. It can provide an almost model-independent estimate of quasifission time scales. However, if the system forms a compact CN, or rotates more than once, the fragment mass can no longer be correlated with angle, limiting sensitivity to times below $\sim 10^{-20}$ s.

(iii) The crystal blocking method measures the angular distribution of fission fragments [8] with respect to a major crystal axis of the target. Fragments emitted in this direction are deflected away (blocked) by the row of atoms, unless the CN has recoiled far enough from the lattice site where it was formed. This method is insensitive to short times, but gives access to longer time scales, in the range 10^{-18} s to 10^{-16} s [9]. It is thus sensitive to time scales associated with fusion-fission, and can indicate the presence of fusion-fission amongst the predominant quasifission events [10].

The crystal blocking method has recently been applied to fissionlike events from reactions forming very heavy elements [9–11]. Measurements showed some filling of the blocking dip, indicating prescission times of $\sim 10^{-18}$ s. However, very different interpretations of these times have been given. For the measurements of Refs. [9,11], the data were consistent with a single fission lifetime of $\sim 10^{-18}$ s, increasing slightly with increasing atomic number (Z) of the combined system (up to $Z = 106$) [11]. Since substantial quasifission is expected in these reactions, it was argued [11] that this time must be characteristic of the quasifission. In contrast, the measurements of Ref. [10] for still heavier systems ($Z = 120$, 124) were interpreted as showing a component (in the range 10%–20%) of very mass-asymmetric fusion-fission with times much longer than 10^{-18} s [10], attributed to shell-enhanced stability of superheavy elements around $Z = 120$.

According to previous expectations [3,12], the quasifission probability should increase with increasing charge of both the projectile and the dinucleus. Since the time scale of quasifission is expected to be shorter than for fusion-fission, the mean fission time should decrease with increasing Z, contrary to the crystal blocking results. The blocking results suggest significant deficiencies in the understanding of reactions forming very heavy elements. It is important to address this problem given the current worldwide efforts directed at producing SHE [1].

This Letter presents measurements of mass-angle distributions (MAD) for reactions of ^{34}S, ^{48}Ti, and ^{58}Ni with ^{184,186}W. These reactions are essentially identical to those used in the crystal blocking measurements of Ref. [9]. The measurements give a complete picture of the evolution of the combined system in the first 10^{-20} s. We present unambiguous evidence of the dominance of quasifission, and show that its time scale decreases with increasing mass of the combined system.

The experiments were performed at the Heavy Ion Accelerator Facility at the Australian National University. Pulsed beams of ^{34}S (149–189 MeV), ^{48}Ti (220–260 MeV), and ^{64}Ni (310–341 MeV) were provided by the 14UD electrostatic accelerator, and superconducting Linac (^{64}Ni). They bombarded isotopically enriched targets of ^{184,186}W, $\sim 50\ \mu\text{g/cm}^2$ in thickness, on $\sim 15\ \mu\text{g/cm}^2$ $^{\text{nat}}$C backings. Binary reaction products were detected in coincidence using two position-sensitive multiwire proportional counters, each with an area of $284 \times 357\ \text{mm}^2$. For the ^{48}Ti and ^{64}Ni induced reactions one counter covered laboratory scattering angles $5° < \theta < 80°$ and the other $50° < \theta < 125°$. The reactions with ^{34}S employed a slightly different geometrical setup, where the first counter covered $4° < \theta < 67°$ and the second $81° < \theta < 167°$.

The position information, together with either the measured time-of-flight [13] or the time difference between two coincident fission fragments [14], allowed the fragment velocities to be determined. Correcting for energy loss in the target, the mass ratio M_R of fragment mass to CN mass was determined, allowing the deduction of the center-of-mass scattering angle $\theta_{\text{c.m.}}$, and thus the mass-angle distributions.

The relationship of the MAD to the lifetime of the system before scission is illustrated schematically in Fig. 1(a). The projectile nucleus (red) is incident from above, and sticks to the larger target nucleus (blue) to form a dinucleus, which rotates as mass is transferred between the two parts. In quasifission, on average mass flow occurs from the heavy to the light partner, mass symmetry being approached asymptotically with an expected [3] time dependence $1 - \exp(t/\tau)$, where τ is the mass-equilibration time constant. If scission occurs very soon after initial contact then little mass is exchanged, and a projectilelike fragment is ejected at a backward angle, [Fig. 1(a), I] with a corresponding targetlike fragment at a forward angle. An increase of the lifetime of the dinuclear system results in larger rotation angles [θ_S in Fig. 1(a)] and more mass exchange [3] as shown in Fig. 1(a), II. A still longer time takes the system to point III. This evolution is illustrated on the MAD shown in Fig. 1(b), where the configurations (I, II, III) correspond to those sketched in Fig. 1(a). A long lifetime, where the system rotates more than one revolution, will destroy the correlation between the mass ratio and fragment emission angle, resulting in symmetric mass splits on average, independent of angle.

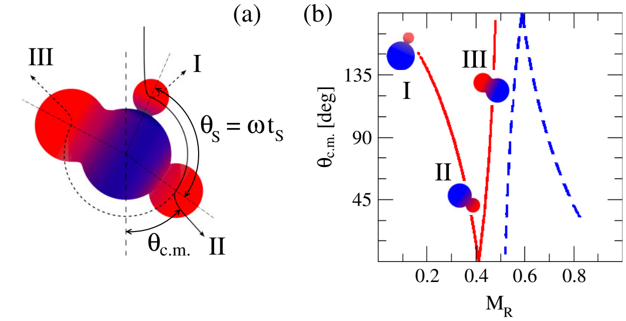

FIG. 1 (color online). Schematic illustration of the evolution of a dinuclear system. (a) Three different quasifission outcomes (I, II, and III) depend on the sticking time (t_S) and rotation speed (ω). (b) Corresponding MAD, illustrating the correlation between emission angle ($\theta_{\text{c.m.}}$) and mass ratios (M_R). The solid (red) line shows the correlated mass and angle evolution of the projectilelike fragment and the dashed (blue) line that of the targetlike fragment.

Mass-angle distributions determined following Ref. [14] for each of the three reactions, at the laboratory beam energies (E) indicated, are shown in the top panels of Fig. 2. The azimuthal coverage of the back (trigger) counter was essentially 90° for all θ; thus, the number of events observed at any $\theta_{\text{c.m.}}$ is proportional to $d\sigma/d\theta_{\text{c.m.}}$. In the MAD we see fissionlike events, more or less spread around $M_R = 0.5$, and also intense bands on either side corresponding to elastic scattering. For the ^{34}S reaction, the grey shaded region around $\theta_{\text{c.m.}} = 90°$ shows where the detector geometry gave no coverage. The panels below show the M_R projections for $45° \leq \theta_{\text{c.m.}} \leq 135°$. They show marked differences between the three reactions. The ^{64}Ni reaction gives a minimum in yield at symmetry ($M_R = 0.5$), ^{48}Ti a broad peak at symmetry, while ^{34}S gives a narrower peak at symmetry. The M_R spectra are in qualitative agreement with many other measurements with similar projectiles [3,4,6,7,15–19]. A conventional interpretation would imply a transition from a reaction mechanism dominated by the deep-inelastic process for ^{64}Ni to one dominated by fusion-fission for ^{34}S. This would give a corresponding large increase in average reaction time. However, the conclusions of Ref. [9] from analysis of crystal blocking measurements, for essentially the same reactions, were (i) that all three reactions have similar (long) mean lifetimes of $\sim 10^{-18}$ s, and (ii) these times are truly characteristic of the processes giving rise to the fissionlike events. These time scales would correspond to

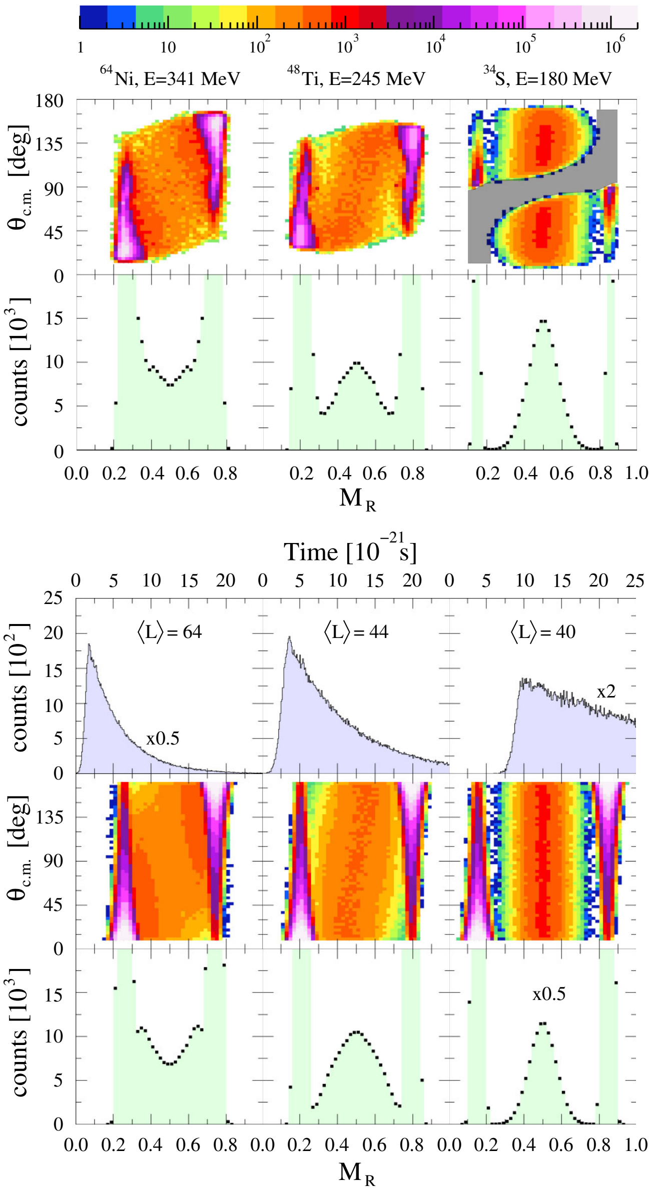

FIG. 2 (color online). The upper panels show the experimental MAD and corresponding projection onto M_R for ^{64}Ni + ^{184}W, ^{48}Ti + ^{186}W, and ^{34}S + ^{186}W (see text). The lower panels show simulated MAD for same reactions and energies, with the M_R spectra at the bottom, and the sticking time distributions above. The capture mean angular momenta $\langle L \rangle$ from CCFULL used in the simulations are also given. These result in good agreement between the simulations and the measurements (top panels).

hundreds of rotations before scission, giving mass distributions independent of angle. Therefore, we must now consider the features of our measured MAD, with reference to Fig. 1, which give access to reaction time information.

For the ^{64}Ni reaction, as the quasifission events move towards $M_R = 0.5$, they also move to more forward (and backward) angles. These correspond to scission of the dinucleus between points I and II of Fig. 1, and are therefore associated with rotations of significantly less than half a turn, and thus short sticking times. For the ^{48}Ti reaction, the correlation is reversed, fissionlike events furthest from $M_R = 0.5$ being found at the more forward and backward angles. These correspond to events between points II and III in Fig. 1, and thus to significantly longer time scales. Finally, for the ^{34}S reaction, the fissionlike events are essentially independent of angle, corresponding to scission at even longer times, beyond point III in Fig. 1.

For ^{48}Ti, it is clear that most of the fissionlike events show a correlation of mass with angle, and thus do not originate from fusion-fission. For ^{64}Ni the fraction of fusion-fission must be even less. To obtain a quantitative determination of the reaction time scales for the dominant quasifission process, a classical Monte Carlo model has been developed to calculate quasifission MAD.

The key ingredients of the model are the distribution of sticking times of the system, and the time scale for mass equilibration. The latter follows Tōke *et al.*, [3], with mass-equilibration time const $= 5.2 \times 10^{-21}$ s. The mass splits of individual events are randomized about the average M_R [as represented by the (red and blue) curves in Fig. 1(b)]. The spread (sigma) was varied from $M_R = 0.025$ at the initial mass split (compatible with deep-inelastic collision mass widths), to 0.07 at symmetry (compatible with fusion-fission). The conversion of sticking time to observed scattering angle is achieved using (i) angular momentum distributions for capture [20] calculated using the coupled-channel code CCFULL [21], (ii) an estimate of the average moment of inertia of the dinuclear system [7], and (iii) classical Coulomb trajectories for the incoming and outgoing nuclei. Uncertainties in the predicted mass-split evolution arise only from uncertainties in the Tōke parametrization. Uncertainties in the angle evolution arise from both the input angular momentum distributions and the moments of inertia. The time scales required to produce MAD matching the experiments depend linearly on these inputs. On this basis, the predictions are estimated to have systematic uncertainties significantly less than 50%, with smaller relative uncertainties.

The quasifission sticking time distributions were parametrized using a half Gaussian followed by an exponential decay. The average and width of the Gaussian, and the decay time, were individually adjusted to reproduce the measured MAD. For simplicity it was assumed that the parameters defining the Gaussian are independent of angular momentum. For visual consistency, elastic scattering was included with a mass width corresponding to the experimental resolution. Using the time distributions shown in Fig. 2, the simulated MAD (bottom panels) reproduce the experimental MAD (top panels in Fig. 2) quite well. The M_R spectra for $45° \leq \theta_{c.m.} \leq 135°$, shown in the lowest panels of Fig. 2, also agree with experiment.

The experimental mass-angle distribution for ^{64}Ni is reproduced using a Gaussian peak time of 1.8×10^{-21} s, with a subsequent decay time of 4.2×10^{-21} s. For ^{48}Ti the peak time is 3.6×10^{-21} s, and the decay time

8.1×10^{-21} s. Although similar MAD can be obtained by complementary adjustment of the peak time and decay time, the mean scission time for quasifission in the model is quite well defined for these reactions, at 5×10^{-21} s for ^{64}Ni, and 10×10^{-21} s for ^{48}Ti. For ^{34}S, the peak time is at least 10×10^{-21} s, while the decay time is not defined, since there is essentially no mass-angle correlation for such long times, so it can only be concluded that the scission time scale is significantly longer than 10×10^{-21} s.

The MAD simulations include only quasifission. A component corresponding to fusion-fission would also be expected. Following systematics for reactions with ^{16}O, it would comprise a Gaussian peak of sigma $\sim$0.07 at $M_R = 0.5$, independent of angle. The competition between fusion and quasifission has previously been investigated [22] for reactions forming isotopes of Th ($Z = 90$). From heavy element cross sections (only from the lower angular momenta) it was shown that the fraction of fusion is $\leq 10\%$ for projectiles heavier than Ca. Both the ^{48}Ti and ^{64}Ni reactions form heavier elements ($Z = 96$ and $Z = 102$, respectively), and fission competes at all angular momenta. Thus much less than 10% of fusion-fission would be expected. Inclusion in the simulations of a small fraction of long lifetime fission slightly shortens the required quasifission decay time; thus, the average quasifission times will be somewhat shorter than those quoted above. This only reinforces the major conclusions from this work, discussed below.

The extracted mean scission times of $\leq 10^{-20}$ s for quasifission in the reactions of ^{48}Ti and ^{64}Ni with isotopes of W must be compared with the mean scission times determined using the crystal blocking method for almost identical reactions [9,11] and beam energies. For the ^{48}Ti + $^{\text{nat}}$W reaction this was 1.0×10^{-18} s; for ^{58}Ni + $^{\text{nat}}$W it was 1.3×10^{-18} s. The blocking measurement selected a (rather wide [11]) subset of all fissionlike events, but this cannot be sufficient to explain a difference in deduced time scales of at least 2 orders of magnitude for these reactions. Even without the quantitative times from the simulations, it is clear from the MAD measurements alone that the dinuclei formed in the Ti and Ni reactions typically rotate less than one turn before scission, which is in quite good agreement with recent theoretical calculations [23,24].

This huge discrepancy in time scales between the MAD measurements and crystal blocking measurements raises questions about the analysis of crystal blocking data in reactions forming heavy nuclei. A first step to address this might be a more detailed presentation of the recoil effect of neutron evaporation from the fission fragments, which can significantly affect the shape of the crystal blocking dip [10,11]. An experimental avenue could consist of making blocking measurements for fissionlike mass splits and angles for which the MAD method has proven the time scale to be short. In this case, the blocking method should show no long-lifetime component.

In summary, we have measured mass-angle distributions, providing qualitative and quantitative evidence that the time scales of the dominant quasifission process for the reactions ^{64}Ni + ^{184}W and ^{48}Ti + ^{186}W are both $\leq 10^{-20}$ s; that for ^{64}Ni is $\sim$ half that for ^{48}Ti. Fission in the ^{34}S + ^{186}W reaction is clearly much slower, with a lifetime significantly longer than 10^{-20} s.

It is necessary to resolve the discrepancies in time scales highlighted in the Letter, as the blocking method, in principle, provides a unique method to investigate the presence of fusion-fission (and thus of fusion itself) among the predominant quasifission events in reactions aiming to form superheavy elements. The resultant better understanding of the reaction mechanisms, and time scales, should contribute significantly to the goal of forming and then investigating the properties of more superheavy elements and isotopes.

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