

Three-Particle Exclusive Measurements of the Reactions $^{238}\text{U} + ^{238}\text{U}$ and $^{238}\text{U} + ^{248}\text{Cm}$

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Kinematically complete experiments have been performed on the three-body exit channels in 7.5 MeV/amu $^{238}\text{U} + ^{238}\text{U}$ and $^{238}\text{U} + ^{248}\text{Cm}$. No evidence was found for an instantaneous three-body breakup. Instead, a two-step nature with minimum scission-to-scission times of 10^{-20} s has been derived even for intermediate nuclei $Z > 110$. However, the observed fission-fragment angular distributions in this region are incompatible with the usual saddle-point concept.

Our first exclusive investigations of the three-body exit channels in nuclear reactions between very heavy ions have focused on sequential fission as a probe for the magnitude and orientation of the spin transfer in deeply inelastic collisions.^{1,2} We report in this Letter an extension of such measurements to the heaviest available target-projectile combinations ^{238}U on ^{238}U and ^{248}Cm . These systems are of particular interest both because of their strong mutual Coulomb forces and their enhanced mass diffusion,³ enabling the study of fission phenomena for nuclei with $Z > 100$ in a region close to and exceeding the limits of the liquid-drop stability. The same experimental technique was used as in our previous work, based on kinematical coincidences with large area detectors. We find that, even for the largest mass transfers, the reaction may still be interpreted as a two-step mechanism with fission following a deeply inelastic collision. We find, however, fission-fragment angular distributions incompatible with the usual statistical distribution of K states at the liquid-drop saddle point.^{4,5} Again in the region of the heaviest elements, we also observe a considerable broadening of the fission-fragment mass distribution. It remains unclear, at present, whether indeed nonequilibrium processes are involved for nuclei produced outside their fission saddle point.

^{238}U and ^{248}Cm targets of 100–200 $\mu\text{g}/\text{cm}^2$ were bombarded with a ^{238}U beam of 7.5 MeV/amu from the UNILAC at Gesellschaft für Schwerionenforschung. The three-particle events were investigated as coincidences between a position-sensitive ΔE - E gas ionization chamber for the heavy nonfissioning fragment with $Z < 92$, and a large-area position-sensitive parallel-plate avalanche counter for the two fission fragments,

the latter detector achieving 4π efficiency for c.m. angles of the first reaction step between 45° and 110° due to kinematical focusing. More details of the arrangement and of the data analysis have been described previously.^{1,2} The strong background from four-body events resulting from double sequential fission was very effectively rejected by inspecting the coplanarity of the three observed velocity vectors in the c.m. system. As verified in Monte Carlo simulations, the probability of confusing a light transfer product of the first reaction step with a heavy fission fragment of the second step—a severe problem in one-particle inclusive measurements³—is negligibly small down to $Z \approx 70$ in the present exclusive experiments.

The distribution of the vector difference $v_R = |\vec{v}_1 - \vec{v}_2|$ of the fission-fragment laboratory velocities from $^{238}\text{U} + ^{238}\text{U}$, integrated over all other observables, is shown in Fig. 1 for two different bins for the charge Z of the fissioning nuclei (determined as the Z complement to the surviving lighter transfer product). As found before for lighter systems,^{1,2} the data clearly demonstrate the existence of intermediate fissioning nuclei as “resonances,” even for the largest mass transfers in the first reaction step. The centroids of the distributions, transformed into average total kinetic energy $\langle E_R \rangle = \frac{1}{2} \mu v_R^2$, where μ is the reduced mass of the two fragments, are plotted in Fig. 2(a) versus the atomic number Z of the fissioning nuclei; results⁶ from the lighter targets, ^{58}Ni and ^{90}Zr , have been included for comparison. All data points follow Viola systematics⁷ very well. Within our present accuracy of a few percent, they are also independent of the Q value, the c.m. angle of the first reaction step, and the orientation of the fission

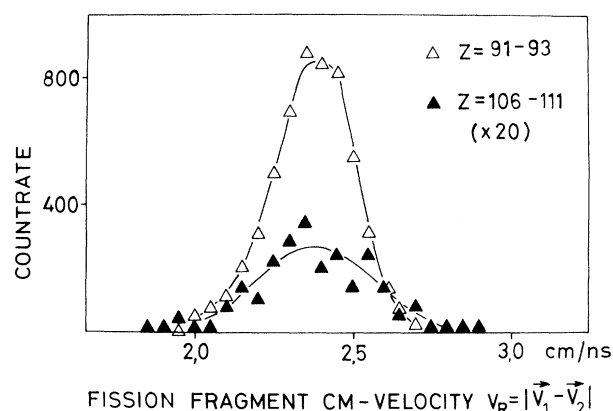


FIG. 1. Distribution of the vector difference $|\vec{v}_1 - \vec{v}_2|$ of the fragment laboratory velocities for two Z bins in the system $^{238}\text{U} + ^{238}\text{U}$, integrated over all other observations.

direction. Thus, an instantaneous three-body breakup of the collision complex can definitely be ruled out up to the heaviest elements produced. From simple Coulomb-trajectory calculations, the lack of detectable Coulomb distortions, which would have led to a correlation between $\langle E_R \rangle$ and the fission direction, establishes a lower limit on the distance of the nonfissioning partner from the fissioning nucleus at scission of 70–100 fm, corresponding to a minimum “scission-to-scission” time of $(3-4) \times 10^{-21}$ s.

The fission-fragment angular distributions are described in spherical coordinates in the rest frame of the fissioning nuclei.^{1,2} The normal to the reaction plane of the first step is chosen as the quantization axis ($\theta = 0^\circ$); the beam axis, lying in the reaction plane ($\theta = 90^\circ$) defines the zero direction of the azimuth angle φ . The fragment azimuthal distributions, integrated over all polar angles θ , are shown in Fig. 3 for a c.m. angular range of 80° – 100° of the first reaction step and cuts in Z corresponding to the heaviest elements. An essentially isotropic behavior with a $90^\circ/0^\circ$ ratio of 1.13 ± 0.22 for $\text{U} + \text{U}$ and 0.84 ± 0.21 for $\text{U} + \text{Cm}$ is found. For a really fast descent towards scission in the second step, anisotropies like an increased fragment intensity parallel to the c.m. recoil axis $\varphi \approx 90^\circ$ of the first step (approximately equivalent to the orientation of the internuclear axis of the primary collision complex at scission) should have been observed. In order to obtain isotropy, an average rotation $\geq \pi$ of the fissioning system before scission is required in a simple model, assuming a symmetrical triangular angular momentum distribution

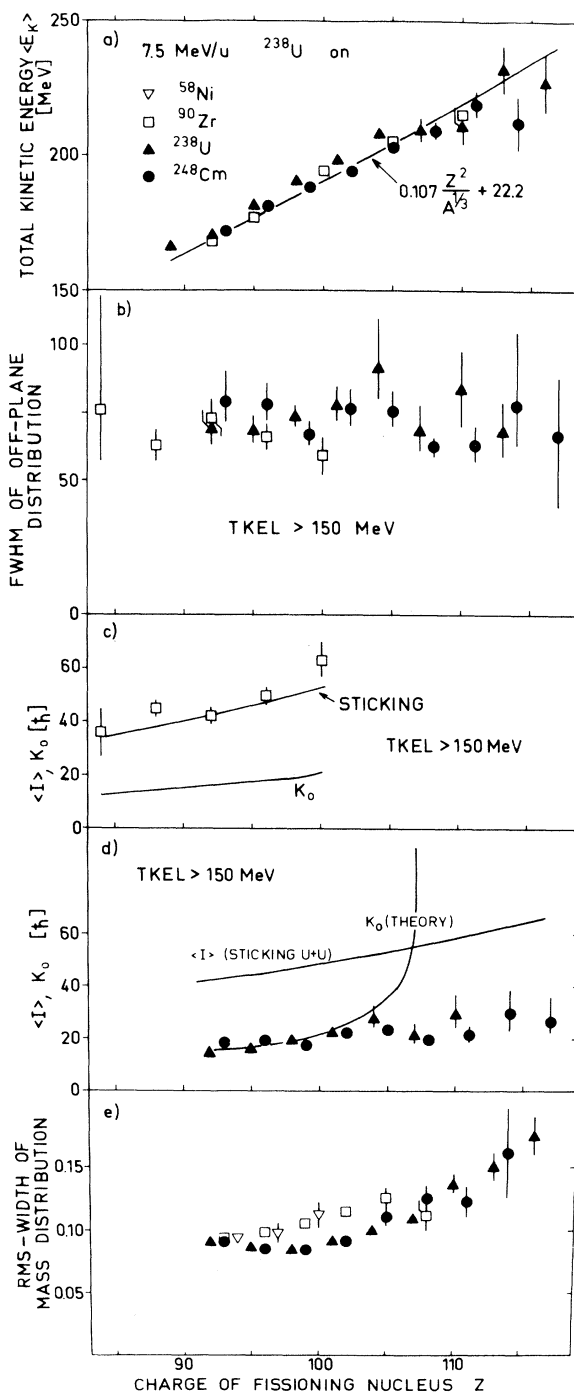


FIG. 2. (a) Average fragment total kinetic energy compared with the Viola systematics; (b) full width at half maximum of the out-of-plane fission fragment angular distributions; (c) average oriented spins $\langle I \rangle$ compared to the sticking-model dependence (the K_0 values used are also shown); (d) extrapolated oriented spins and deduced effective values for K_0 , compared to the liquid-drop-model prediction; (e) width of the fragment mass distributions, defined as rms of $A_1/(A_1 + A_2)$. All quantities are plotted vs the atomic number of the fissioning nucleus.

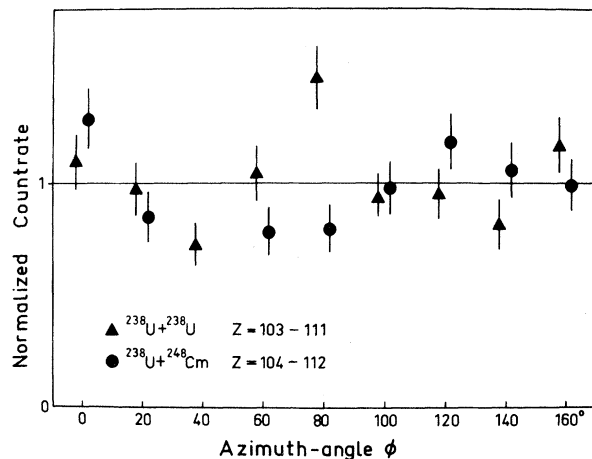


FIG. 3. Azimuthal angular distributions of fission fragments for a high- Z bin integrated over the polar angle. Systematic errors may double the indicated statistical errors.

starting at zero and taking account of both rotational directions because of identical particles in the entrance channel. The fluctuations in the initial direction stemming from the bending have been estimated to be negligible ($\langle \sigma_\phi^2 \rangle^{1/2} \approx 3^\circ$). With an average spin of $50\hbar$ (see discussion below), average rotation angles $\geq \pi$ for a fissioning mass-280 nucleus correspond to times of $\sim 12 \times 10^{-21}$ s with limits of 7×10^{-21} s for rotation of a rigid sphere, and 25×10^{-21} s for rotation of two mass-140 nuclei sticking together. These scission-to-scission times are considerably larger than the limit obtained above provided that the fluctuations in the time dependence of the shape changes down to scission are smaller than that to make these estimates meaningful at all. They are also larger than theoretical saddle-to-scission time estimates of $\sim 4 \times 10^{-21}$ s.⁸

As observed before in the sequential fission of lighter systems,^{1,2} the out-of-plane fragment angular distributions exhibit the strong concentration in the reaction plane expected for the decay of a system with considerable angular momentum aligned perpendicular to that plane. The half-width of these distributions as a function of the atomic number Z of the fissioning nuclei is shown in Fig. 2(b) for a cut in the total kinetic loss $-Q > 150$ MeV, selecting only deeply inelastic events. Rather constant values of $\sim 75^\circ$ are observed up to the heaviest elements. As in our previous work,^{1,2} averaged oriented spins $\langle I \rangle$ of the fissioning nuclei can be deduced from the polar distributions, relying on empirical values⁹

for the variance K_0 of the Gaussian distribution of the spin projection K on the symmetry axis of the fission saddle-point configuration, and assuming randomly oriented spin components $M_0 \approx 13\hbar$ as established for $^{208}\text{Pb} + ^{90}\text{Zr}$.^{1,2} The Z dependence of $\langle I \rangle$, derived for the deeply inelastic component in $^{238}\text{U} + ^{90}\text{Zr}$ [Fig. 2(c)],⁶ is consistent with a sticking-model behavior up to $Z \approx 100$ where empirical K_0 values (shown for comparison) exist.

In the region $Z > 100$, only accessible with ^{238}U and ^{248}Cm targets, empirical values for K_0 are not known. An extrapolation of the rotating-liquid-drop model⁵ yields a steep rise of K_0 with a complete loss of stability and a corresponding divergence of K_0 around $Z = 107$ [Fig. 2(d)], taking account of angular momenta $\sim 50\hbar$. This should have resulted in isotropic polar distributions. Conversely, any attempt to derive spins for $Z > 100$ on the basis of the observed constant angular widths would lead to an obviously unphysical divergence of $\langle I \rangle$. In Fig. 2(d), we have therefore reversed the method and plotted effective values of K_0 deduced from the widths, assuming the smooth sticking dependence of $\langle I \rangle$ established for $Z < 100$ to remain valid up to $Z \approx 115$ and incorporating the previous value of M_0 . The rather constant behavior of K_0 is in clear conflict with the usual concept of a statistical equilibrium at the saddle together with conservation of K beyond.⁴ We offer two alternative interpretations for this result:

(i) Retaining the notion of an equilibrated system before fission, K nonconservation on the passage from saddle to scission—conceivable in view of the high temperatures involved and the long path from a nearly spherical saddle shape for high- Z elements—may favor a drift towards lower K values due to the higher level density of low- K states for prolate shapes. It seems tempting to treat K in a transport formalism with microscopically derived transport coefficients.

(ii) In a nonequilibrated, relatively “fast” process the fissioning system, formed already beyond its saddle point, may be descending towards scission at the scission time of the first reaction step. The orientation of the fission direction would then not reflect properties of the fissioning nucleus, but average random spin components of the primary collision complex in the direction of the common separation axis.

Clearly, experimental data on fragment angular distributions in fusion-fission reactions for such

high- Z intermediate nuclei would be of great value in this context.

The observed fission-fragment mass distributions show the well-known asymmetric mass split in the quasielastic region with a rapid transition to symmetric fission for larger energy losses. The Z dependence of the rms width of these distributions, plotted in Fig. 2(e), exhibits a considerable increase in the region of $Z > 100$ up to the heaviest elements, much larger than expected¹⁰ for the slight increase in nuclear temperature, correlated with Z . A possibly related broadening in the mass distribution has also recently been observed in fusion-fission reactions of lighter elements with fission barriers reaching zero because of very high angular momenta.⁹ This apparent lack of dependence on the entrance channel suggests that the phenomenon reflects inherent properties of the liquid-drop model, i.e., a correlation between a decreasing stability against mass asymmetry and a loss of stability in the fission degree of freedom for nuclei with a vanishing fission barrier. Alternatively again, a relatively fast process may be involved, in which the shape of the observed mass distributions is influenced by an incomplete thermalization of the asymmetry mode, strongly excited in the first step of the reaction.¹¹

In summary, our investigations of the heaviest collision systems available have so far not given evidence for an instantaneous three-body breakup. Irrespective of the apparent two-step nature

of the observed fission phenomena with relatively long scission-to-scission times of $\geq 10^{-20}$ s, non-equilibrated systems may still be involved. Their clear identification has to be left to future experiments.

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Evidence for a Soft Nuclear-Matter Equation of State

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The entropy of the fireball formed in central collisions of heavy nuclei at center-of-mass kinetic energies of a few hundred MeV per nucleon is estimated from the ratio of deuterons to protons at large transverse momentum. The observed paucity of deuterons suggests that strong attractive forces are present in hot, dense nuclear matter, or that degrees of freedom beyond the nucleon and pion may already be realized at an excitation energy of 100 MeV per baryon.

One of the principal motivations for accelerating heavy-ion beams to relativistic energies is the hope of producing and studying matter at baryon densities greater than are found in atomic nuclei. However, information about the properties of the dense matter thus created is obscured by

the fact that the matter remains hot and dense only for a very short time, $< 10^{-22}$ sec, and our observations are limited to the products emitted as it disassembles. We present arguments that the ratio R_{dp} of deuterons to protons is established during the early stages of the fireball's existence,