Molecular-Orbital Study of Late-Fission Times in Deep-Inelastic ^{238}U + ^{238}U Collisions

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The U-like K-vacancy production probability in 7.5-MeV/nucleon deep-inelastic ^{238}U + ^{238}U collisions has been measured for no-fission, single-fission, and double-fission exit channels. The results are interpreted using quasimolecular orbital correlations for diatomic and triatomic configurations. This leads to the determination of lower limits for the time scale for late fission of U-like products (≥ 8 and ≥ 4 as) and their probability ($\geq 77\%$ and $\geq 52\%$), at mean initial-excitation energies of ~ 40 and ~ 105 MeV, respectively.

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The time scale of fission induced by heavy-ion nuclear reactions has received increasing attention in recent years [1-3]. In the 10-100-MeV initial-excitation energy range, fission times of the order of 0.1 as ($=10^{-19}$ s) or shorter are typical [1,2]. On the other hand, crystal blocking experiments indicate that the distribution of fission times can extend to much larger times and that an appreciable fraction of fission events can lie in this "late-fission" tail [3].

In this Letter, we propose using atomic K-shell vacancy production and decay in a 7.5-MeV/nucleon ^{238}U + ^{238}U deep-inelastic reaction as a clock to examine the late-fission fraction of the U-like reaction products and we report the first results using this method. The method is related to that proposed by Gugelot [4] and, although not as detailed as the other methods, has the advantage that the information is obtained simply and is essentially free of nuclear models. We first discuss the experimental findings, then provide an interpretation using molecular-orbital (MO) correlation diagrams for two- and three-body atomic systems [5-7].

Figure 1 sketches the experimental arrangement. A beam of 7.5-MeV/nucleon ^{238}U ions from the Lawrence Berkeley Laboratory SuperHILAC bombarded ^{238}U targets, 640 to 750 μg/cm^2 thick, set perpendicular to the beam. Scattered particles were detected on one side of the beam in a radial ionization chamber (RIC) [8] which was centered at a laboratory angle $\Theta = 40°$ and subtended azimuthal and polar angular ranges $\Delta\Theta = \pm 14.5°$ and $\Delta\phi = \pm 7.5°$, respectively. The collision partners were detected on the opposite side of the beam in a time-of-flight arm of two parallel-plate avalanche counters (PPAC), separated by 27 cm, and centered at $\Theta = -45°$, with $\Delta\Theta = \pm 20°$, $\Delta\phi = \pm 20°$. In the RIC, particles were identified by a ΔE-ΔE-E detection system and in the PPACs by ΔE, time-of-flight, and multiple-hit information. Each system could clearly distinguish fission products from unfissioned particles. Events were recorded for (1) both U-like reaction products surviving fission ("two-body" events), (2) one reaction product fissioning ("three-body" events), and (3) both reaction products fissioning ("four-body" events). Neutrons and light charged particles were not detected. X-ray spectra in coincidence with each type of particle event were taken in a 1000-mm^2, 8-mm-thick Ge (planar) detector. A Ge(Li) and a NaI detector served as gamma-ray detectors for the calculation of the x-ray intensity produced by internal conversion (IC) [9]. The Q value or total kinetic energy loss (TKEL $= -Q$) of each two- and three-body event was determined from the energy and angle of the unfissioned reaction product detected in the RIC. For four-body events, only overall spectra could be obtained. The angular and energy acceptances of our detectors spanned essentially the entire relevant Wilczynski plot for this reaction [10].

As sample coincident x-ray spectra, in Fig. 2 we show

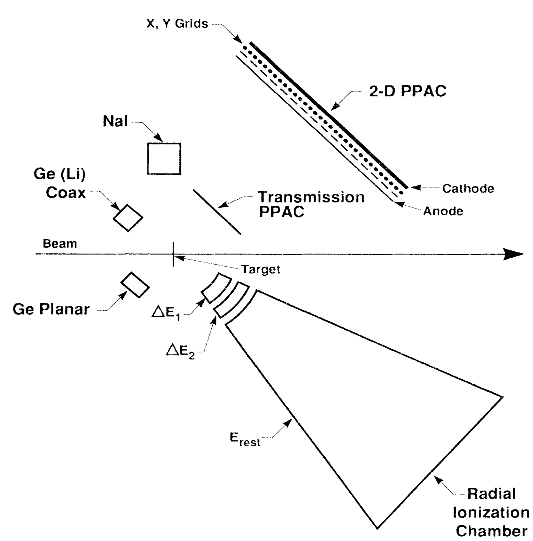

FIG. 1. Schematic of the experimental setup. See text for details.

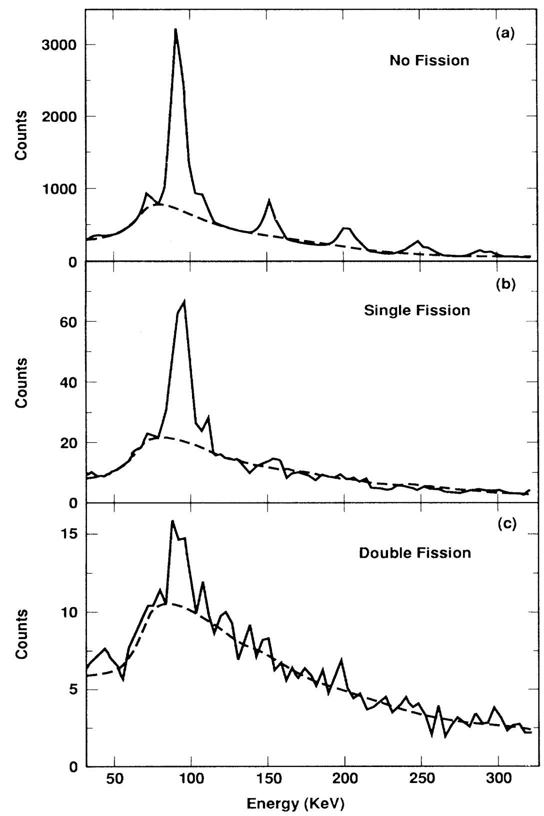

FIG. 2. Typical overall x-ray spectra, taken by the Ge planar detector (Fig. 1), in coincidence with (a) two-, (b) three-, and (c) four-body particle events. The continuum gamma-ray backgrounds are shown by dashed lines.

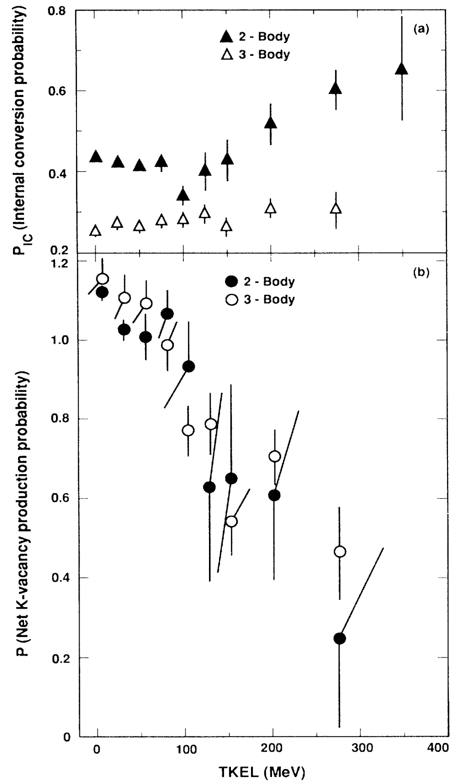

FIG. 3. (a) Internal conversion (IC) K-vacancy contributions for two- and three-body events as a function of the total kinetic energy loss (TKEL $=-Q$). (b) Net U-like K vacancies per reaction for two-body (●) and three-body (○) events as a function of the total kinetic energy loss, corrected for IC contributions.

the spectra summed over the entire detected TKEL range. In the two-body spectrum (a), one sees U K x rays and Coulomb excited gamma rays from ^{238}U, both Doppler broadened [11]. Spectra (not shown) gated by TKEL bins indicate only U K x rays, which is consistent with the finding [10] that any higher-Z product dominantly fissions. The three- and four-body x-ray peaks in (b) and (c) are 10 to 15 keV wide and could contain x rays from elements as low as $Z\simeq 86$. For simplicity, we call all these x rays "U-like." After subtracting the continuum gamma-ray background under each x-ray peak, as well as an interfering ^{238}U 104-keV gamma-ray peak [9], dividing by the total number of two-, three-, or four-body events, and taking into account the x-ray detector solid angle and efficiency, the number of U-like x rays per reaction was determined. From this, the number of IC-produced U-like x rays was subtracted, as described in Ref. [9] [see Fig. 3(a)]. We found the following net K-vacancy yields per reaction, averaged over the entire TKEL range, for two-, three-, and four-body decays, respectively (the U fluorescence yield $\omega_K=0.98$): $\bar{P}_2=1.08\pm 0.03$ (0.40), $\bar{P}_3=0.94\pm 0.06$ (0.27), and $\bar{P}_4=0.36\pm 0.08$ (0). The numbers in parentheses give the subtracted IC yields. Nuclear transitions which have a non-negligible IC coefficient are too slow, compared to fission, to yield any U-like K x rays. This gives rise to a reduced IC x-ray background for $\bar{P}_3$ and zero background for $\bar{P}_4$. Each maximum possible yield is 4, because each U-like product can have up to two K vacancies. Figure 3(b) shows the net K vacancy yields per reaction, $P_2(Q)$ and $P_3(Q)$, for two- and three-body events in individual Q-value bins. Calling $S_i(Q)$ the number of i-body particle singles per unit Q value, the relation between $P_i(Q)$ and $\bar{P}_i$ is $\bar{P}_i=\int P_i S_i\, dQ/\int S_i\, dQ \simeq P_i(\bar{Q}_i)$, if $S_i(Q)$ peaks sharply at $\bar{Q}_i$ (see below).

The most important result of this work is the observation that (1) $P_3(Q)\simeq P_2(Q)$ and (2) $\bar{P}_4>0$, i.e., U-like K x rays are observed even if both partners fission. From this, we conclude that *there is an appreciable fraction of fission events for which the observed fission time scale is at least of the order of the U K vacancy lifetime* $\tau_K=7$ *as* [12,13]. The simplicity of this albeit qualitative conclusion may be contrasted with the rather elaborate calculations typically necessary to extract fission time information from the other methods [1–3].

To understand our results quantitatively, we examine first the electronic and then, briefly, the nuclear processes. Since the relative velocities between the nuclei are much smaller than the Bohr velocity of the K electron(s) attached to the nuclei [14], the electronic processes are nearly adiabatic [15] and, in a collision, the lowest electronic levels follow (from left to right) the MO diagrams in Fig. 4. Figure 4(a) gives the MO correlation for U+U [6]. At 7.5 MeV/nucleon, ~28% of the final U $1s$ vacancies are produced in the $1s_\sigma$ MO and the rest in the $2p_\sigma$ MO [16]. In two-body events, each U-like product carries 50% of the total vacancies. In three-body events, the same MO diagram applies, if fission is "late." To give a quantitative meaning to this time, we note that observation of a U-like K x ray requires the $1s_\sigma$ and $2p_\sigma$ MO levels to lie within ~10 keV of the U $1s$ binding energy B_K(U). This necessitates a minimum separation $R_{\rm min}$ between the two U-like nuclei of $\sim 10^4$ fm [6]. Hence, for later discussion, we define the following absolute times ($t=0$ at nuclear contact between the ^{238}U projectile and ^{238}U target): t_1, the deep-inelastic sticking time; t_2, the time for the two U-like products to reach $R_{\rm min}$; $t_{\rm sc}$, the time at which a U-like product scissions; and t_3, the time at which the fission products, e.g., Pd+Pd for symmetric fission, reach a relative separation such that *their* $1s_\sigma$ MO energy still lies within ~10 keV of B_K(U) [5]. With these definitions, emission of a U-like K x ray followed by late fission occurs in the interval between t_2 and t_3. From the Q dependence of P_2 one

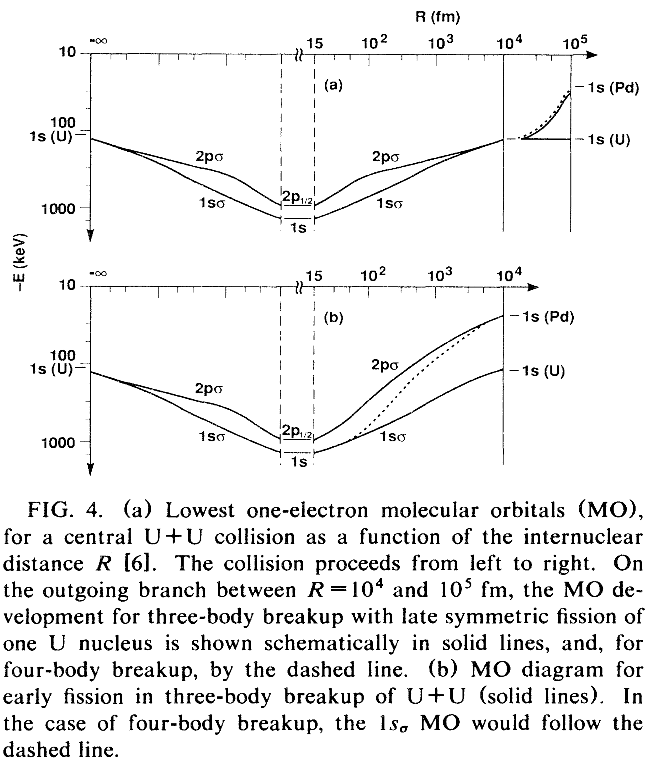

FIG. 4. (a) Lowest one-electron molecular orbitals (MO), for a central U+U collision as a function of the internuclear distance R [6]. The collision proceeds from left to right. On the outgoing branch between $R=10^4$ and 10^5 fm, the MO development for three-body breakup with late symmetric fission of one U nucleus is shown schematically in solid lines, and, for four-body breakup, by the dashed line. (b) MO diagram for early fission in three-body breakup of U+U (solid lines). In the case of four-body breakup, the $1s_\sigma$ MO would follow the dashed line.

finds $t_1 \simeq 10^{-3}$ as [9]; for repelling charged spheres, $t_2 - t_1 \simeq 0.3$ as, $t_3 - t_{\rm sc} \simeq 0.05$ as.

The sketch in Fig. 4(a) between $R=10^4$ and 10^5 fm illustrates schematically what happens in late three-body events after the time $t_{\rm sc}$: one U-like product remains unfissioned, the other may scission into two Pd nuclei. In late four-body events, both U-like products scission, as indicated by the dashed line. Figure 4(b) applies in very "early" three-body events ($t_{\rm sc} \ll t_2$). The diagram is a relativistic modification of a MO diagram for Be^{3+} breaking up into $(He+2H)^{3+}$, examined by Dowek *et al.* [7]. Here, the unfissioned U-like product receives $1s$ vacancies only from the $1s_\sigma$ MO. For four-body events with early fission of both U-like products, the $1s_\sigma$ MO would follow the dashed line to the Pd $1s$ state, and *no U-like K x rays could be produced.*

In the present system the late-fission fraction must be large: For early fission only, one expects $\bar{P}_4 = 0$, contrary to our result $\bar{P}_4 \simeq 0.36$, and $P_3(Q) \simeq 0.28\, P_2(Q)$, contrary to the result $P_3 \simeq P_2$ [Fig. 3(b)]. To extract more quantitative information on the fraction of late fissions, we use a simple model in connection with Fig. 4. Assume during the collision, $\nu(Q)$ vacancies per reaction are made in two MOs. Call $f(Q)$ the probability that a late-fissioning product emits a U-like K x ray. If a fraction $\chi(Q)$ of fissions are late and $1-\chi$ are early, the theoretical number of U-like K x rays per reaction, averaged over Q and assuming $\omega_K = 1$, is

$$\bar{P}_2 = \bar{\nu}, \tag{1}$$

$$\bar{P}_3 = [\bar{\chi}(1+\bar{f})/2 + 0.28(1-\bar{\chi})]\bar{\nu}, \tag{2}$$

$$\bar{P}_4 = (0.56 + 0.44\bar{\chi})\bar{\chi}\bar{f}\bar{\nu}, \tag{3}$$

where it is assumed that $\overline{\chi f \nu} \simeq \bar{\chi}\bar{f}\bar{\nu}$, etc., and where in $\bar{P}_3$ late and early fission of one partner, and in $\bar{P}_4$ late-late, late-early, and early-early fission of both partners are taken into account.

Before comparing these expressions with experiment, we note that S_2, S_3, and S_4 peak, at quite different Q values, so that two-, three-, and four-body events may have differing $\bar{\nu}$, $\bar{f}$, and $\bar{\chi}$ values. From our data, we find mean Q values $\bar{Q}_i = \int Q S_i\, dQ / \int S_i\, dQ$ for $i=2$ and 3: $-\bar{Q}_2 \simeq 8$ MeV and $-\bar{Q}_3 \simeq 80$ MeV. For four-body events, where the Q value could not be determined, a simple statistical-model calculation [17], considering only neutron emission and fission, and adjusted to fit $S_2(Q)$ and $S_3(Q)$, yields $-\bar{Q}_4 \simeq 210$ MeV.

Although, with $\bar{\nu}$, $\bar{f}$, and $\bar{\chi}$ differing, one cannot solve Eqs. (1)-(3) simultaneously, the conditions $\bar{f} \le 1$ and $\bar{\chi} \le 1$ still provide useful limits for the three- and four-body decay modes (indicated by subscripts). To obtain these limits, we assume on the basis of Eq. (1) that $\bar{\nu}_3 \simeq P_2(\bar{Q}_3) = 1.0 \pm 0.1$ and $\bar{\nu}_4 \simeq P_2(\bar{Q}_4) = 0.5 \pm 0.2$. Substituting the experimental values for $\bar{P}_3$ and $\bar{P}_4$, we find the results listed in columns 1 to 3 of Table I. In column 1, $\bar{E}_{\rm in}$ is the mean initial-excitation energy of one U-like product, equal to $-\bar{Q}/2$, since in a symmetric sys-

TABLE I. Late-fission fractions of U-like products.

$\bar{E}_{in}$ (MeV)	$\bar{\chi}$	$\bar{f}$	t_{sc} (as)
~105	≥0.52	≥0.41	≥4
~40	≥0.77	≥0.66	≥8

tem the total initial-excitation energy divides equally on the average.

In our model, f is the probability of U-like K x-ray emission between t_2 and t_3 by a fissioning channel. Lacking a proper dynamical calculation for the actual electronic and nuclear processes [18], we assume that this x-ray emission probability falls off exponentially with time. This yields $f \simeq 1-\exp(-t_{sc}/\tau_K)$, where we have used *a priori* the fact that in the present case, $t_3-t_2 \simeq t_{sc}$. Substitution of the limits for $\bar{f}_3$ and $\bar{f}_4$ gives the results listed in column 4 of Table I.

These lower limits on the times t_{sc} and on the fractions χ of late-fissioning U-like products must be confronted by any theoretical calculation of fission time distributions. The cooling of highly excited nuclei by neutron evaporation before fission [19] and the slowing down of the fission evolution by dissipative effects between the saddle point and scission [20,21] no doubt come into play here. But, to explain the large fraction of long fission times found here, one may have to adduce other proposals [22–24].

We conclude that the present method yields in a simple manner new information about the probability of late fission in highly excited U-like products, essentially independent of nuclear-model assumptions.

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