Nuclear Reaction Times in the Deep-Inelastic U+U Collision Deduced from *K*-Shell Ionization Probabilities

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The *K*-shell ionization probability P_K of the uraniumlike products has been measured in the deep-inelastic reaction U+U at a beam energy of 7.5 MeV/u as a function of the total kinetic energy loss $-Q$. P_K was determined for Q values down to -190 MeV. After the subtraction of the ionization induced by internal conversion of γ rays, a strongly Q-dependent P_K is found in qualitative agreement with theoretical predictions. From the data we infer a nuclear reaction time of approximately 10^{-21} s at $Q=-100$ MeV.

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In 1960, it was suggested that the bremsstrahlung spectrum produced in a nucleus-nucleus collision can be influenced by a time delay due to a nuclear reaction.[1] In a similar way, such a time delay can also influence the probability of ionization of an inner-shell electron during the collision.[2] Both effects are caused by interference between the ionization (or bremsstrahlung) amplitudes on the way into the collision and on the way out of the collision. The expected change of the shape of the bremsstrahlung spectrum and the change of the ionization probability have been observed experimentally in the resonant elastic scattering of protons by various nuclei.[3-6] It has been suggested by Anholt[7] and by Müller[8] that a similar effect should exist for the ionization of the atomic *K* shell in deep-inelastic collisions of heavy nuclei. Simple semiclassical models of the nuclear reaction mechanism in such collisions predict that the time delay should increase with increasing energy dissipation, i.e., the longer the nuclei stick together the larger the total kinetic energy loss (TKEL) in the collision.[9] Hence a measurement of the ionization probability P_K as a function of TKEL is equivalent to a measurement of P_K as a function of the sticking time T. A comparison of theoretical predictions and experimental results should then allow a direct determination of T.

In this Letter we report first measurements of the *K*-shell ionization probability of uranium in the deep-inelastic reaction U+U at 7.5 MeV/u as a function of TKEL. This reaction was chosen because a strong variation of P_K with TKEL is predicted by theory. Also, the inner-shell ionization probability due to the time-varying Coulomb field during the collision should be larger than the ionization probability due to internal conversion (IC) of γ rays from the deexcitation of the highly excited reaction products.[10] The experiment was done at Gesellschaft für Schwerionenforschung in Darmstadt. The experimental setup is shown in Fig. 1. The U beam was directed onto a 500-μg/cm^2 U target. The reaction products were measured with two position-sensitive parallel-plate avalanche counters (PPAC). The position of each particle was read out, both with respect to the left and the right edge of the counter in order to recognize double hits, i.e., events in which two reaction products hit the detector simultaneously. A ΔE counter in front of the PPAC allowed discrimination between fission fragments and nonfissioned heavy reaction products. The x rays were detected with two planar Ge detectors, and the γ rays with a coaxial Ge(Li) and a NaI detector. The energy scale and the photon detector efficiencies were determined before and after the run with calibrated γ sources at the target position.

The theoretical calculations of the process are based on the following assumptions: (1) The nuclear reaction can be described by classical trajectories which are interrupted for a time T during which the nuclei stick together. (2) The ionization

amplitude during the nuclear sticking is zero because the Coulomb field experienced by the atomic electrons is constant during this time. (3) The atomic electronic states can be described by molecular orbitals around the two charge centers evolving from the separated-atom (SA) states to the united-atom (UA) states as the nuclei collide. In the system investigated here, ionization of the $1s\sigma$ and $2p\sigma$ molecular orbitals had to be considered because both of these orbitals correlate to the SA K shell. The ionization amplitude can be determined as[7]

$$a_{if}(\epsilon) = -i(i/\hbar)\int_{-\infty}^{\infty} dt\, M(t,\epsilon)\exp[i\int_{-\infty}^{t} dt'\omega(t')], \quad (1)$$

where $\hbar\omega(t) = E(t) + \epsilon$, $E(t)$ is the electron binding energy, ϵ is the kinetic energy of the ionized electron, and $M(t,\epsilon)$ is the matrix element for ionization. E varies as the two nuclei approach, going from the SA binding energy to the UA binding energy. The integral can be broken up into the following parts: $-\infty \rightarrow 0$, $0 \rightarrow T$, and $T \rightarrow \infty$. One can then show that

$$a_{if} = a_{\text{in}} + a_{\text{out}} e^{i\omega(0)T}, \quad (2)$$

where a_{in} corresponds to the integral $-\infty$ to 0 and a_{out} to 0 to $+\infty$ for the integrand in Eq. (1) with $T = 0$. The ionization probability is $P_K = \int d\epsilon \times |a_{if}(\epsilon)|^2$. For $\omega(0)T$ of the order of unity one can expect a pronounced interference effect even after integrating over all final electron energies ϵ. Present calculations of $M(t,\epsilon)$ consider only radial coupling from the relativistic $1s\sigma$ and $2p\sigma$ molecular orbitals to the continuum.[8] Rotational couplings such as that between the $2p\sigma$ and the $2p\pi$ orbitals have yet to be included.[11] It should be noted that the energy loss in the collision reduces the c.m. velocity of the outgoing particles and thereby reduces P_K. Calculations show that for U+U this effect alone reduces P_K by less than 10%.[8]

The experimental data were sorted by determining the x-ray spectra for different Q-value bins (TKEL $= -Q$). The Q values were calculated from the scattering angles of the reaction products under the assumption that two-body events occur. Events where one or both of the emerging reaction products had fissioned were discarded. The x-ray spectra show that no measurable amount of reaction products heavier than U survived, i.e., only U-like reaction products were found. Figure 2(a) shows the relative cross sections for two U-like reaction products surviving fission as a function of $-Q$. For comparison, U production probabilities calculated

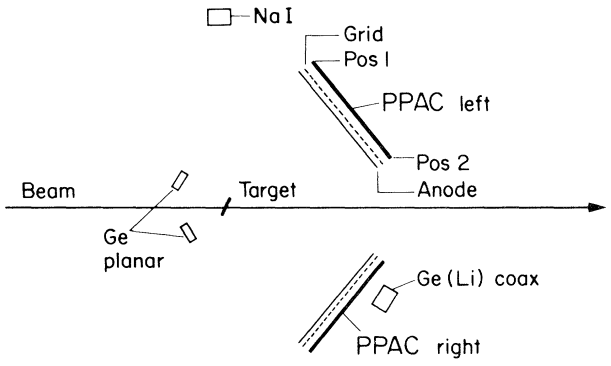

FIG. 1. Experimental setup for the present experiment. PPAC, position-sensitive parallel-plate avalanche counter.

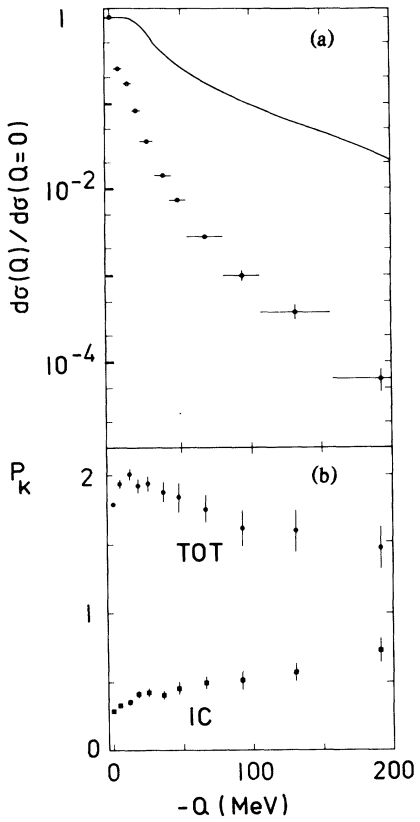

FIG. 2. (a) Relative cross sections for deep-inelastic reactions with unfissioned reaction products as a function of $-Q$. The curve is the U production probability computed in Ref. 9. (b) $P_K(Q)$ as determined from the raw data (circles). The squares give the IC contribution $P_{K,\text{at}}(Q)$ determined from the observed γ spectra.

by Riedel and Nörenberg[9] are shown, but they do not take into account fission of the primary reaction products. The ratio between the calculated curve and our data shows a high fission probability at high-$(-Q)$ values.[12,12] Nevertheless, we have found U nuclei surviving fission up to $-Q=190$ MeV. Since the cross section falls off very rapidly with $-Q$, the center of gravity of the Q bins is shifted towards smaller $-Q$ values.

The ionization probability obtained by dividing the number of U x rays (corrected for detector efficiency and solid angle) by the number of particle-particle coincidences with the same Q value is shown in Fig. 2(b). A considerable fraction of the observed K x rays must be attributed to internal conversion (IC) of γ rays from the highly excited reaction products [also shown in Fig. 2(b)]. In order to obtain the IC contribution, we first determined the IC x-ray intensity for quasielastic events where only Coulomb excitation is important; in this calculation, we chose the elastic scattering events in the angular range $34.5° \pm 2.5°$ using the appropriate conversion coefficients. Noting that the overall shape of the γ spectra is roughly the same at $Q=0$ and at lower Q values, except for the fact that the spectrum becomes continuous, we assumed that the amount of IC is proportional to the area of the γ spectrum above the K-absorption edge of U: $P_{K,\mathrm{IC}}=cA$, where c has been determined at $Q=0$, $\theta_{\mathrm{lab}}=34.5°$. This simple relation assumes that the mean conversion coefficient of the γ rays is independent of Q, an assumption which should be reasonable since the multipolarity of the dominant γ rays is $E2$.[14]

Subtracting $P_{K,\mathrm{IC}}$ from P_K, we obtain the atomic ionization probability $P_{K,\mathrm{at}}$ shown in Fig. 3. We observe a pronounced decline in $P_{K,\mathrm{at}}$ with decreasing Q, which by itself indicates that the sticking-time effect on P_K is large. The predictions of Müller *et al.*[8] are also shown in the figure. The calculations give P_K as a function of the time delay T due to the nuclear reaction. The theoretical curve has been multiplied by a factor of 0.88 in order to normalize $P_K(T=0)$ to the experimental $P_K(Q=0)$. Comparing $P_{K,\mathrm{at}}(Q)$ and $P_K(T)$ we can determine the reaction time T as a function of Q. The result is shown in Fig. 4 together with delay times calculated by Schmidt, Toneev, and Wolschin[15] and Wolschin[16] for the same reaction. Our results indicate delay times of the same general order as predicted. It is very pleasing to note that the atomic physics results confirm the predictions of the theory of deep-inelastic nuclear reactions. In a new experiment, we will try to determine P_K even if one of the reaction products undergoes fission. In this case we expect an x-ray yield a factor of 2 lower, because only the unfissioned atom can emit an x ray characteristic of a U-like nucleus, as a result of the fact that fission is many orders of magnitude faster than an x-ray transition.

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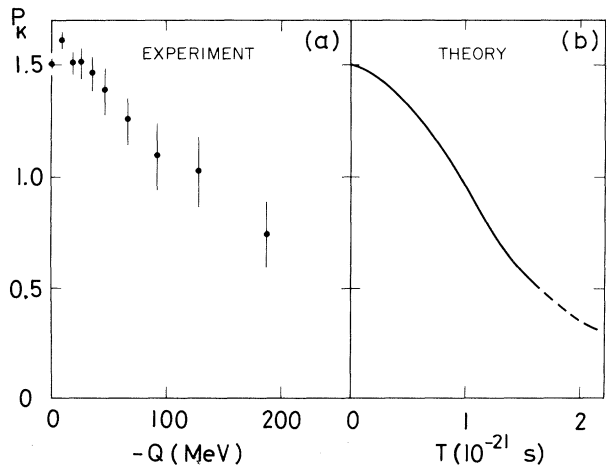

FIG. 3. (a) Probability of direct ionization $P_{K,\mathrm{at}}(Q)$ and (b) theoretical $P_{K,\mathrm{at}}(T)$ scaled from Ref. 8.

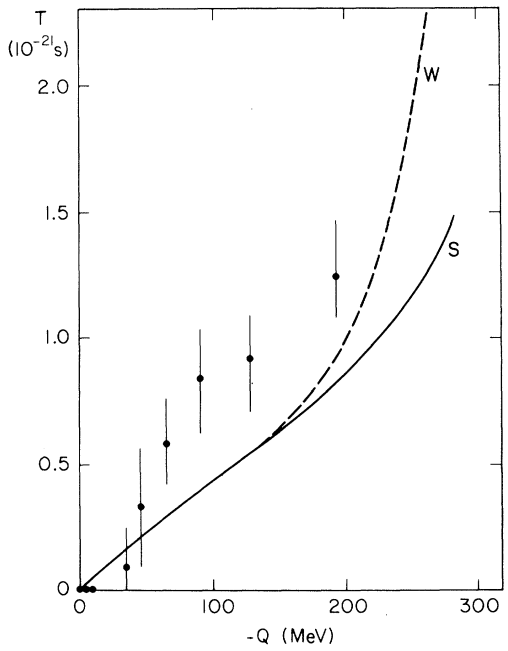

FIG. 4. Semiempirical relation between T and Q inferred from Fig. 3 and as calculated in Ref. 16 (W) and Ref. 15 (S).

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