Time delay effects on K x-ray production probability in deep inelastic U + U and U + Pb nuclear reactions

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If an atomic collision is accompanied by a deep inelastic nuclear reaction with a time delay ΔT, a phase change which affects the ionization probability P_K of the target- and projectile-like products is introduced between the incident and outgoing ionization amplitudes. In a deep inelastic nuclear reaction, the nuclear reaction time ΔT is monotonically related to the total kinetic energy loss $(-Q)$ of the reaction products. Therefore a measurement of P_K as a function of Q then yields ΔT. The K-shell ionization probability P_K has been measured in the deep inelastic reactions U + U and U + Pb at a beam energy of 1785 MeV as a function of the total kinetic energy loss $-Q$. P_K was determined for Q values down to -190 MeV. After subtraction of the ionization induced by the 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 1×10^{-21} s at $Q=-100$ MeV. The observed reaction times agree fairly well with the predictions of the classical nuclear reaction models used to describe deep inelastic scattering processes.

I. INTRODUCTION

In several publications, it has been suggested that atomic effects observed in ion-atom collisions could be used to study the time structure of nuclear reaction processes. These ideas are based on the fact that the atomic inner-shell ionization probability could be affected by the nuclear reaction time due to time-dependent interference effects between the amplitudes describing the ionization of the inner-shell electrons during the approach and the separation of the two colliding nuclei. Experimentally, the predicted variation of the inner-shell ionization probability has been observed in the resonant elastic scattering of protons on several nuclei.[1–3]

Anholt[4] and Müller[5] have suggested that a similar effect could also exist in the ionization process of the atomic K shell in deep inelastic collisions of very heavy nuclei, like uranium. In this kind of elastic reactions, target and projectile are thought to form a nuclear complex for a time ΔT (typically of the order of magnitude of 10^{-21} s), in a process involving a large dissipation of kinetic energy and a considerable mass transfer. On the other hand, from the atomic point of view, the nuclei involved are the source of a time-dependent Coulomb potential which affects the behavior of the inner-shell electron and is responsible for most of the atomic excitation processes. In such collisions, it is also believed that the inner-shell electrons will follow the nuclear motion almost adiabatically into molecular orbitals (MO's). According to this model, the ionization probability of a K-shell electron of the projectile or target atom during the collision can be described in terms of electron ionization of the $1s\sigma$ and $2p\sigma$ molecular orbitals. Such an electronic behavior has been studied intensively by several groups[6–8] and by ourselves[9,10] over the last ten years, primarily looking at heavy-ion collisions below the Coulomb barrier where the change in the electromagnetic field (i.e., in the nuclear trajectories) is well known. Moreover, such a method provides a unique means of extending the experimentally accessible range of atomic physics far beyond the region of stable elements. The present paper reports of a series of experiments performed to investigate the behavior of K electrons in the deep inelastic reactions $^{238}U+{}^{238}U$ and $^{238}U+{}^{208}Pb$ at 1785 MeV, whose first results were published in Ref. 11. The resulting information is used to study the time evolution of these nuclear reactions in a time region of 10^{-21} s not accessible by any other time measuring methods. The experimental arrangement, the observed x- and γ-ray spectra, and the particle spectra are described in Sec. II. In Secs. III and IV of this paper we deal with the data analysis and the background problems which have to be solved in order to obtain the purely quasimolecular inner-shell ionization probability of interest here. The results are discussed in Sec. V and compared in Sec. VI with some theoretical predictions based on semiclassical nuclear friction models.

II. EXPERIMENT

A. Principles

Simple semiclassical models of nuclear-reaction mechanisms in heavy-ion deep inelastic collisions predict that the sticking-time interval ΔT of two nuclei increases with increasing total kinetic energy loss (TKEL) or decreasing Q value (TKEL equal to $-Q$). Hence, a measurement of the ionization probability P_K as a function of Q is equivalent to a measurement of P_K as a function of ΔT, meaning that a comparison between calculations of $P_K(t)$ for a delay time t based on the molecular model of electron ionization processes and the experimental results should allow determination of ΔT. According to Refs. 4 and 5, the interference pattern is prominent when $\omega(0)\Delta T \simeq 1$, where $\hbar\omega(0)$ is the binding energy for the MO's at the minimal distance reached in the collision between the two nuclei. For collisions of very heavy nuclei such as U + U at energies close to the Coulomb barrier, binding energies of the order of 1 MeV for both $1s\sigma$ and $2p\sigma$ quasimolecular levels are predicted by the time dependent molecular model, suggesting that sticking times of the order of 10^{-20}–10^{-21} s could be measured by this method.

The probability P_K for inner-shell ionization during the collision is determined by the intensity of the characteristic K x-ray radiation emitted by the separated reaction products, if the lifetime of the vacancies produced in the MO states is longer than the collision time. This condition is fulfilled for deep inelastic processes, because here the reaction time is always much shorter than the relatively slow electromagnetic decay of a vacancy. In order to perform such an experiment it is necessary to measure the characteristic x rays as well as the TKEL associated with each event. We chose to investigate the systems U + U and U + Pb, because of the expected high K-shell ionization probability (for both $1s\sigma$ and $2p\sigma$ orbitals) and the very high binding energy of the molecular levels.

B. Experimental arrangement

The experiment was performed at the UNILAC heavy-ion facility of the Gesellschaft für Schwerionenforschung (GSI), Darmstadt. The 7.5 MeV/nucleon ^{238}U beam (charge state 68 +) was directed onto a 500 μg/cm^2 ^{238}U (600 μg/cm^2 ^{208}Pb) target foil. In order to suppress systematic effects due to an eventual target evaporation, the uranium material was sandwiched between two layers of carbon (15 μg/cm^2 on the front side and 43 μg/cm^2 on the back side, respectively). The mean energy loss in the carbon and in the target itself led to an effective beam energy in the reaction of 7.42 MeV/nucleon (1766 MeV). The basic arrangement is shown in Fig. 1(a). The characteristic x rays, together with the γ rays from target and projectile, were registered in coincidence with the reaction products using four photon detectors, placed at different angles and each one covering a complementary photon energy range. The x rays (of about 100 keV) were detected with two planar high-purity Ge detectors (HP Ge) placed backwards at $+150°$ and $-150°$ with respect to the beam axis. Every detector was provided with absorbers of 2 mm Al and 1 mm Cu in order to reduce the lower-energy radiation and optimize the detector efficiency in the photon energy range between 70 and 120 keV, where the characteristic U and Pb K-shell radiation is expected. At these energies, the energy resolution was about 3 keV, which allowed us to distinguish between $K\alpha_1$, $K\alpha_2$, and $K\beta$ atomic transitions. A typical spectrum is given in Fig. 2 and is discussed in the next section.

The γ rays due to the nuclear deexcitation of the reaction products were measured in a NaI (7.6×7.6 cm) detector placed at 90° for photon energies up to 4 MeV. Finally, a coaxial Ge(Li) (80 cm^3) detector placed behind the right particle detector covered the photon energy spectrum below 2 MeV. The energy scale and the photon detector efficiency for each detector were determined before and after the run with calibrated γ sources (^{207}Bi, ^{54}Mn, ^{152}Eu) placed at the target position in the scattering chamber. Each photon detector was also provided with lead shields in order to suppress the radiation background produced in the collimating slits and in the Faraday cup. Both collimator and Faraday cup were placed far away from the target in order to reduce the solid angle of the photon-background sources as much as possible.

The nuclei emerging from the reaction were detected in the angular range from 17° to 58° (lab) with two position-sensitive parallel-plate avalanche counters (PPAC's) placed symmetrically to the beam axis. The position was

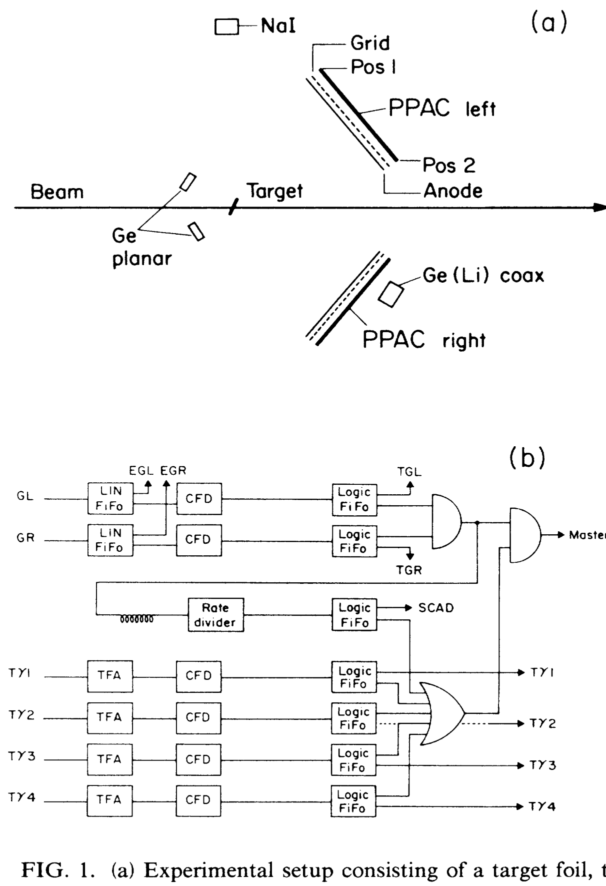

FIG. 1. (a) Experimental setup consisting of a target foil, two position-sensitive avalanche counters (PPAC's), two planar high-purity germanium photon detectors (HPGe) for the detection of the characteristic K x rays, and a NaI and a Ge(Li) detector for the measurement of the high energy γ rays. (b) Hardware trigger logic.

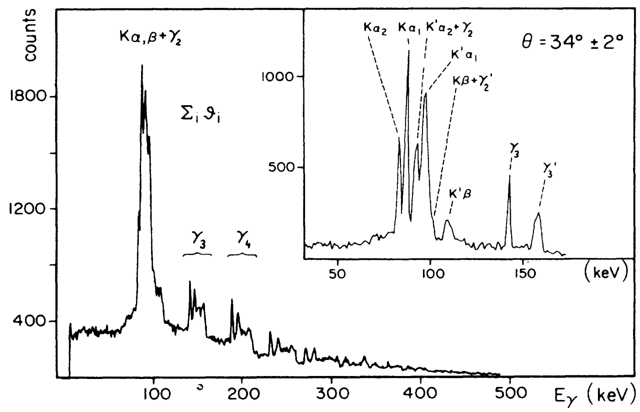

FIG. 2. Typical HPGe-detector photon spectrum for an elastic U + U collision. The label K indicates characteristic K x-ray emission, whereas γ_i stand for the Coulomb-excited lines. Each line is characterized by two energies, according to the Doppler shift $(\gamma_i, \gamma_{i'})$.

read out as the delay of the signal from the backplane (used as delay line) with respect to the prompt anode signal. The delay line was designed so that the delay time was proportional to the scattering angle. In addition, the delay time for each event was determined with respect to the left- and the right-hand sides of each particle detector. This information was used to distinguish between single and multiple events in the same detector. For one detector, the angular range was determined from the dimensions of the geometrical arrangement, whereas the 90° kinematical condition for the elastic scattering of identical particles, such as U + U, was used to calibrate the angles of the second detector. The detector angular resolution was about 0.4°. A two-track trigger (both PPAC's had to fire) imposed the condition that the boundary of the accepted binary events corresponded to the angles between 32° and 58° for elastic and quasielastic scattering.

For the particle identification, a ΔE counter was placed in front of the PPAC. Its signal allowed us to distinguish between fission fragments and unfissioned heavy-ion reaction products. The information from all detectors was interfaced by standard CAMAC electronics to a PDP11/45 computer system working in event mode storage with magnetic tape support [Fig. 1(b)]. A coincidence between the signals from the two avalanche counters and at least one of the photon detectors was required to accept an event. For each event, 29 parameters were stored; the data were processed off line. In addition to the two-particle one-photon events ($2T\gamma$), also the two-particle events ($2T$), prescaled by a factor of 10, were registered. The data presented here were obtained from about 3×10^6 registered $2T$ events.

C. X-ray spectra

Figure 2 shows the observed x-ray transition lines ($K\alpha_1$, $K\alpha_2$, K_β) emitted from target and projectile, which are relevant for the determination of the K-vacancy production probability, as well as the Coulomb-excited ^{238}U γ emission and the γ-ray continuum. The photons have a Doppler shift which depends on the angle between the emitting particle and the coplanar x-ray detector. For coincident events, this Doppler-shift signature allows one to distinguish between photons emitted from a reaction product hitting the left (with respect to the beam direction) or the right PPAC detector. Each line is split into two distinct peaks which can best be seen in the case of the Coulomb-excited lines emitted by the uranium nuclei. Due to the finite aperture of particle and photon detectors and the Dopper effect, the shape of the x-ray and γ lines measured in coincidence with the right PPAC is different from that of the spectra coincident with the left particle detector. Accordingly, the solid-angle efficiency in the laboratory system is not the same for both cases. Furthermore, shape and position of each photon peak strongly depend on the reaction kinematics, i.e., on the relative angle between the emitting particle momentum and the photon detector.

The coincident photon energy spectrum shown in Fig. 2 is typical for U + U collisions. The prominent x-ray lines at a laboratory energy of about 85 keV are the atomic K transitions from uranium. The upper inset displays the spectrum taken at a scattering angle $\theta_{\rm lab}=34°\pm2°$ (for the right detector). In the observed angular range between 32° and 58°, it can occur that the $K'\alpha$ characteristic x rays interfere with the $K\beta$ line and the γ_2 Coulomb line of ^{238}U with those of the $K\alpha$ and $K\beta$ lines. For this reason, the ionization probability was evaluated first by using the whole energy range of the K radiation. Afterwards, the results obtained in this way were corrected for all possible effects, including γ_2, which contribute to K-vacancy production. A polynomial fit was applied to the measured background, and its contribution subtracted from the x-ray lines. This background results from Compton scattering in the detector, from unresolved γ rays emitted by highly excited reaction products, from nuclear bremsstrahlung, and bremsstrahlung from secondary electrons. A data sorting for different kinematic regions (different scattering-angle and Q-value bins) was performed to verify that the fitting procedure was appropriate for all angle combinations.

D. PPAC spectra

As mentioned, the particle detection and identification system consisted of a position-sensitive avalanche counter (PPAC), provided with a ΔE discrimination in front. Figure 3 gives the angular distribution for (a) U + U and (b) U + Pb collisions. This plot is generated offline and represents the measured time differences between the two delayed signals which are produced by every event at both ends of the delay line of each particle detector. The time difference is directly proportional to the scattering angles in a given PPAC. In both cases, at least two different regions can be distinguished: the first one covers a lab scattering angle range between 17° and 32° and the second one the range between 32° and 58°. These angles refer to the right PPAC. In the first region, only three-or four-body reaction products from fragmentation processes are registered, because binary events are excluded here by the

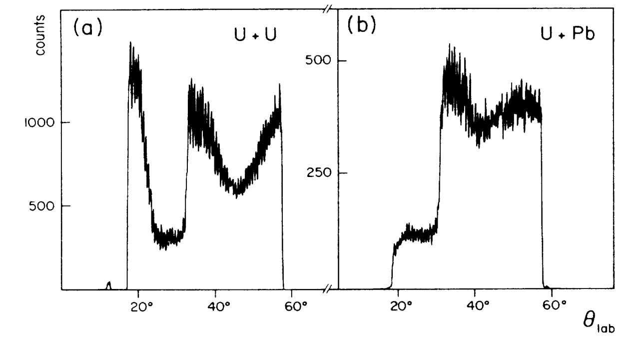

FIG. 3. Position spectrum of the left PPAC (raw data) for (a) U + U and (b) U + Pb.

fact that the angle in the left PPAC would be greater than 58° for such events. Nearly 80% of these multifragment events are in coincidence with events lying in the same angular region of the left PPAC and also in coincidence with fission fragments registered by both ΔE counters (i.e., four-body events). The region between 32° and 58° is dominated by the elastic and quasielastic binary channel. The symmetry about 45° for U + U reflects the fact that our setup does not allow one to distinguish between the recoiling target nucleus and the projectile. Our ΔE information was not sufficient to distinguish between U and Pb in the slightly asymmetric U + Pb case. In principle, it is possible to establish a unique center of mass scattering angle for each binary event using the information on the Doppler shift of the x rays emitted by the reaction products. However, the statistical accuracy of our data in the inelastic region was not good enough to apply this method here.

III. DATA EVALUATION

The data were processed by dividing the particle spectra into scattering-angle bins, each characterized by a mean scattering angle related to the right detector. Each bin contained elastic, slightly inelastic, as well as deep inelastic reaction products. Several cuts were applied to each event in order to identify and eliminate the fission product contribution. X-ray spectra coincident only with binary events were then generated for different Q-value bins of the nuclear reaction.

The TKEL was calculated from the scattering angle of the reaction products, using two-body kinematics and assuming that the detected particles have the same masses as the original target and projectile nuclei. This assumption is justified by the Z-distribution measurements performed by Hildebrand *et al.*[12] for U + U at the same energy. In that experiment, it was found that the Z distribution peaks sharply at $Z=92$ with no indication of any higher Z values. This result is supported by our own observation and proves that in U + U collisions the binary reaction products surviving fission are mainly uranium particles. The reason is obvious: In the case of an asymmetric fragmentation of the U + U complex, all the reaction products heavier than U do not survive the sequential fission process. Therefore, the highest probability for having a binary reaction not followed by a fission corresponds to two emerging U particles. In our x-ray spectra, also, no evidence for the existence of reaction products heavier than U has been found.

The situation is more complicated for U + Pb, because here the mass of the incoming U nucleus can be reduced and that of the Pb nucleus increased by mass transfer, a process which increases the chance for survival against fission. As a consequence, the observed mass spread for U + Pb is considerably larger than in the U + U case and accordingly leads to much larger uncertainties in the evaluations of the Q values. In addition, some uncertainties are introduced by the fact that we are not able to distinguish between projectile- and target-like products. For the calculation of TKEL, the nucleon evaporation has been taken into account, but was found to be small compared to our experimental uncertainty.

Figure 4 shows the observed ionization probability for U + U and U + Pb as a function of the Q value. These results were obtained by dividing the number of U and Pb x rays (corrected for detector efficiency, solid angle, and fluorescence yield) by the number of particle-particle coincidences measured at the corresponding Q values. Since each K level is occupied originally by two electrons, the maximum value allowed for $P_K(Q)$ is 4. Our experiment indicates that at $Q=0$ MeV about 50% of the K-shell electrons in the U + U system and only about 30% in the U + Pb system are ionized. In order to obtain the net atomic-ionization probability these raw data have to be

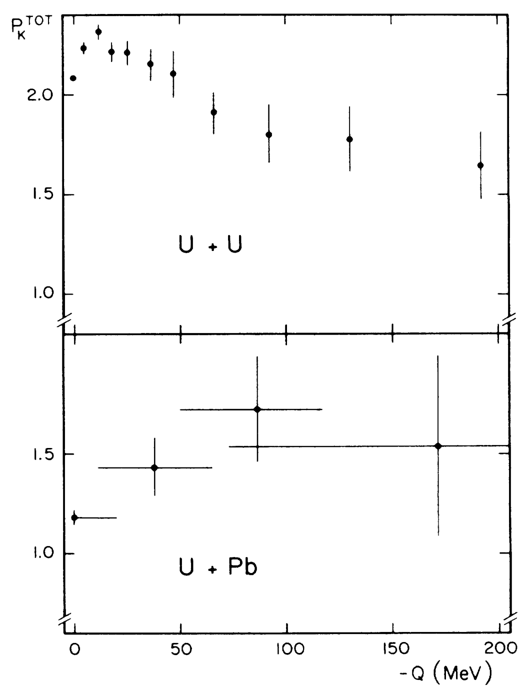

FIG. 4. Total ionization probability P_K^{tot} as a function of $-Q$ for U + U (upper part) and U + Pb (lower part). These raw data include contributions from mechanisms other than molecular excitation, such as internal conversion. Also, they are not yet corrected for fission effects.

corrected for all additional processes contributing to inner-shell ionization. This procedure is described in detail in the following section.

IV. CORRECTIONS

A. Internal conversion contribution

It is well known that an excited nucleus is able to interact directly with a bound inner-shell electron and to transfer its excitation energy to it, so that the electron is ejected in a one step process. The electromagnetic decay of the newly created hole in the atomic shell produces an Auger electron or a characteristic x ray. This so-called "internal conversion" (IC) process has been investigated in detail and the probabilities are well known. The number of inner-shell vacancies produced by internal conversion depends on the γ multiplicity, multipolarity, and the energy distribution of the γ rays emitted by the excited nucleus. The typical time scale involved in such an electromagnetic process is of the order of 10^{-12}–10^{-15} s, i.e., very large compared to the nuclear and atomic times of interest here, ranging between 10^{-21} and 10^{-23} s. Therefore the IC ionization adds itself incoherently to the atomic ionization probability and the number of characteristic x rays due to IC has to be subtracted from the observed K x-ray spectra.

To obtain a relationship between the IC produced x-ray and the observed γ-ray spectra, we investigated first the elastic and quasielastic channel, where the well known Coulomb-excitation process dominates. To study the U + U case, an angular range of 34°±2° was selected, i.e., the kinematic region corresponding to impact parameters where only Coulomb excitation of the nuclei occurs. It is well known that ^{238}U is a good rotator and that the deexcitation occurs in form of a γ cascade where $E2$ transitions dominate. The low-energy part of these spectra including the K x rays was measured with the HPGe detector and the high energy part with the Ge(Li) and NaI detectors. The first eight Coulomb-excited lines were observed with the HPGe detector, and twelve more with the other two detectors. Using the absorber and efficiency corrected intensity of these lines and the appropriate conversion coefficients, the amount of IC produced K x rays for $Q=0$ was first determined. Since the conversion coefficients decrease rapidly with increasing γ-ray energy, we found that only the low-energy part of the Coulomb-line spectrum is of importance.

The situation changes with increasing inelasticity of the collision. Here, the intensity of the Coulomb lines decreases continuously with decreasing Q value, until they are totally swamped by a continuous γ-ray spectrum, the dominant background in the deep inelastic region. Fortunately, the envelope of the Coulomb-excitation spectrum at $Q=0$ was found to have approximately the same shape as the continuous γ-ray background. This allowed us to assume that the IC contribution is simply proportional to the areas of the γ-ray spectra above the K-absorption edge for uranium. We determined this proportionality factor for all three photon detectors first at $Q=0$ and then for the Ge(Li) and NaI detectors for all Q values of interest, from the elastic down to the deep inelastic region. By means of these intensity ratios and using the known IC contribution for $Q=0$, we were finally able to evaluate the ionization probability $P_K^{\mathrm{IC}}(Q)$ for characteristic x-ray production due to internal conversion as a function of the Q value.

In this procedure we have assumed that the shapes of the γ-ray spectra are independent of the Q value, an assumption justified by both the Ge(Li) and NaI γ-ray spectra. Based on the results of earlier experiments,[13,14] showing that about 80–100 % of the γ-ray continuum results from $E2$ transitions, we have also assumed that the conversion coefficient is independent of the Q value.

The results of this evaluation for U + U and U + Pb are displayed in Fig. 5. In the U + U case the IC contribution increases first with decreasing Q value until about -70 MeV and then levels off to a more or less constant value. This behavior is in good agreement with measurements on other systems like Xe + Pb and Xe + Th.[15]

In the case of elastic U + Pb scattering, as expected, only Coulomb lines from excited uranium nuclei were observed. Assuming that only these lines contribute to the IC ionization probability at $Q=0$, we find a value which is indeed only half of that measured for U + U. The Pb nuclei contribute to the IC K x-ray production rate in the deep inelastic region only.

B. $\gamma_{1,2}$ contribution

The contributions of the lowest-lying Coulomb γ lines, γ_1 (44 keV) and γ_2 (104 keV) was considered separately. The γ_1 line can be converted in the L shell only, and,

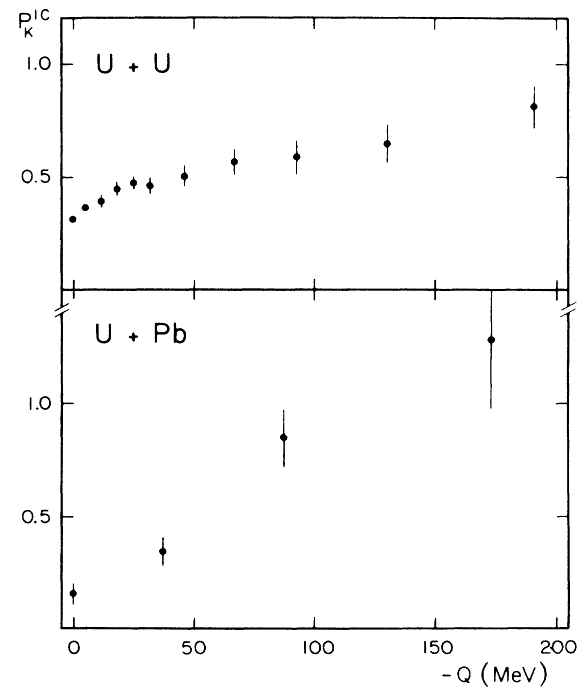

FIG. 5. Internal-conversion contribution $P_K^{\mathrm{IC}}(Q)$ to the ionization probability, as determined from the measured γ-ray spectra.

therefore, is of no importance here. The γ_2 line is Doppler broadened and partially superimposed on the uranium $K\alpha$ and $K\beta$ transition lines. Its contribution has been calculated by assuming that the higher states depopulate in the form of a cascade so that their intensity is determined by that of the upper lines, e.g., γ_3, γ_4, etc. It is found that the γ_2 transition contributes about 4% to the IC K x-ray intensity at $Q=0$. Its influence decreases with increasing inelasticity and was quantified by assuming a constant ratio to the higher lying γ lines. For Q values less than -50 MeV the γ_2 contribution can be neglected.

C. Fission rejection

Another important aspect of this experiment is the correct interpretation and evaluation of the particle spectra, particularly in view of the fission-fragment identification. An incomplete fission event rejection not only yields a wrong Q value for the reaction, but also a wrong value for P_K. This is due to the fact that the fission time of the excited U-like or Pb-like nuclei is much shorter ($\sim 10^{-21}$ s) than the lifetime of a K vacancy (10^{-17}–10^{-18} s). Therefore no characteristic projectile- or target-like x rays will be observed if fission occurs, and accordingly the value of P_K is reduced if these events are not properly suppressed.

The fission process associated with the deep inelastic scattering has been investigated recently, but so far the picture is still incomplete in this region. In our analysis we considered only the so-called sequential fission processes assuming that the target-projectile complex separates first and then one or both reaction products fission in a second step within about 10^{-20} s, i.e., at an internuclear distance of about 70–100 fm. So far, no faster sequential fission processes have been observed at our beam energy for such heavy systems. To identify such events we used the ΔE spectra as well as the position information delivered by our particle detector system.

Let us consider first events where only one fission fragment reaches a PPAC and the other is lost. In principle, our ΔE detectors were able to distinguish between intact reaction products and fission fragments. Nevertheless, it was not possible to reject all unwanted events using the ΔE information alone, because the total energy of the registered particles could not be determined in this experiment. The lack of this information prevented us from determining the complete kinematics of the fission process with sufficient accuracy and accordingly it was impossible to distinguish between fission events resulting from the quasielastic and deep inelastic region. Although this background contribution amounted to only about 2% in the elastic scattering region, it became important with increasing inelasticity. There, these events finally formed a peak below the unfissioned particle peak in the ΔE spectrum and partially overlapped with it. Except for the highest Q values, this effect was reduced to a negligible amount by applying restricting cuts to the ΔE spectrum. This procedure affects only the statistics but not the ionization probability. For the highest Q values the contribution of fission events was evaluated by extrapolating the ΔE fission peak into the unfissioned peak region. We

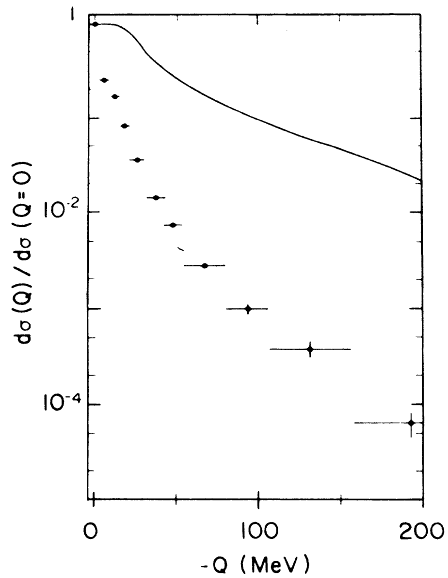

FIG. 6. Cross sections for deep inelastic processes with unfissioned reaction products normalized to the value at $Q=0$. The solid line is the production probability (ignoring fission) computed in Ref. 17.

found that due to the tail under the relevant part of the ΔE spectrum about 25% of all events at -190 MeV were associated with fission. At higher inelasticities this value increased to 100%.

In the case where both fission fragments reached the same detector simultaneously, the electronic pileup simulates an intact uranium-like event, because the energy loss for U in the ΔE counter is about twice that for fission fragments. These events were rejected by applying appropriate cuts on the particle multiplicity spectra (the sum of both time signals for each delay line). Finally, the amount of double hit events, where the relative scattering angle between the two fragments was smaller than the detector angle resolution, had to be determined. Assuming that the fission is isotropic in the center of mass of the decaying particle, it is found that their contribution at the position of the single events (in the multiplicity spectra) is not more than 5% in the worst case, i.e., at the highest values of the inelasticities considered here.[16]

Figure 6 shows the relative cross sections for two U-like reaction products surviving fission, as a function of $-Q$. For comparison, uranium production probabilities calculated by Riedel[17] are shown, which, however, do not take into account fission of the primary reaction products. At high Q values the ratio between the calculated curve and our data indicates a high fission probability, varying by about two orders of magnitude over the energy range from 0 to -200 MeV. However, it is interesting to note that U- and Pb-like nuclei have been found surviving fission up to excitaton energies of 190 MeV.

V. RESULTS

The final results for the atomic inner-shell probability P_K^{at} on U + U and U + Pb collison corrected for all nonatomic contributions $[P_K^{IC}(Q),\ P_{\gamma_2}(Q)]$ from P_K^{tot} are displayed in Fig. 7 (the open circles at high $-Q$ are corrected for the ΔE-fission tail contamination, discussed in the preceding section). P_K^{at} at $Q=0$ has been determined as an absolute value without normalizing to other experiments. Our results at $Q=0$ can be compared with other experiments. Molitoris *et al.*[10] measured P_K^{at} as a function of the impact parameter b of the scattering. In this experiment the problem of inner internal conversion was solved by using a γ-cascade suppressor. At the smallest impact parameters measured (7–10 fm) and for beam energies of 7.3 and 7.5 MeV/nucleon, they found $P_K=1.6\pm0.2$ and 1.8 ± 0.2, respectively. Calculations by Müller[5] predict for P_K a value of 1.71 at $Q=0$ and $b=0$ which agrees fairly well with our result $[P_K^{at}(Q=0)=1.68\pm0.05\pm0.12]$ and that of Ref. 10. The accuracy of our data, which is determined by statistics and by uncertainties in the background subtraction (first contribution) is also affected by systematic uncertainties in the determination of the γ-detector efficiencies, which can change the result by $\pm7\%$ (second contribution).

Figure 7 shows that for U + U $P_K^{at}(Q)$ drops by a factor of ~ 2 if Q decreases from 0 to -190 MeV. Since only a small fraction of this decrease results from the reduced kinetic energy in the outgoing channel ($\sim 10\%$ at -190 MeV according to Ref. 5), we have here clear evidence of a time delay effect. As it can be seen, this effect is much less pronounced in the U + Pb case (see lower part of Fig. 7). The latter, at first sight surprising, result can be qualitatively explained in the following way: Let us assume that the Q dependence of the time delay effect producing the interference pattern in P_K^{at} is the same in both cases, i.e., depends on the parameter $(\omega\,\Delta T)$ (see Sec. VI). From quasimolecular model calculations[18] it is known that the binding energy $\hbar\omega_{\rm united}$ for the $2p\sigma$ orbital in the united system U + Pb is about $(1/1.6)\hbar\omega$ that of U + U. Therefore the results for U + U can be compared with those for U + Pb simply by expanding the time scale for $P_K(\Delta T)$ (where ΔT is the nuclear time delay) by a factor 1.6 for the U + U case. This scaling law is also valid for any Q values because the nuclear friction models[19] predict a linear dependence between ΔT and $-Q$. The result of such a rescaling of the U + U data is displayed in the lower part of Fig. 7 (open triangles) together with the U + Pb data. The agreement is surprisingly good and confirms not only the validity of the U + Pb results but also the existence of the suggested time delay effect.

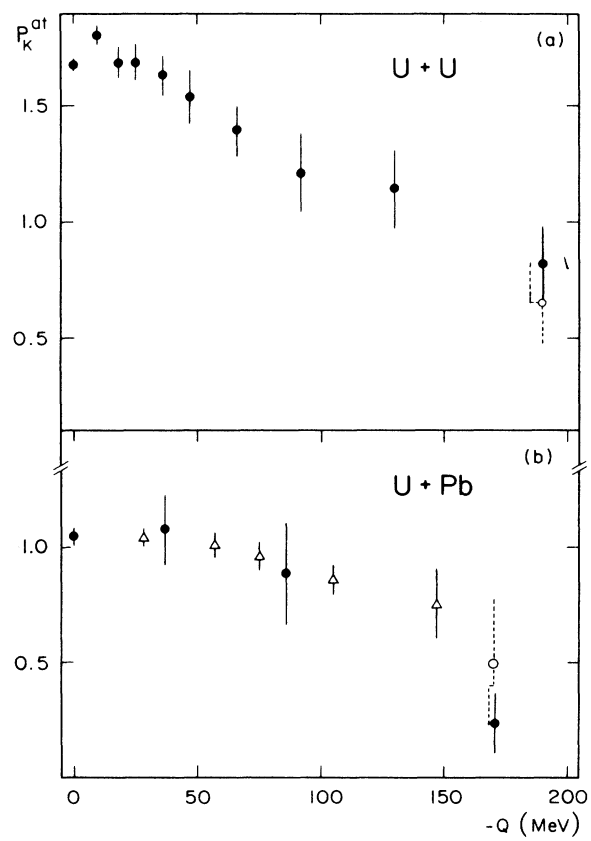

FIG. 7. Final values of $P_K^{at}(Q)$ for the reaction (a) U + U and (b) U + Pb. The open circles indicate data points corrected for fission and the open triangles correspond to U + U results rescaled for the comparison with U + Pb data.

VI. $P_K(Q)$: AN ATOMIC CLOCK FOR DEEP INELASTIC NUCLEAR PROCESSES

To understand the observed results for the atomic-ionization probability P_K^{at} as a function of Q, we have to compare them with the predicted behavior of $P_K(\Delta T)$. The theoretical calculations are all based on the semiclassical approximation (SCA) model, assuming that the nuclei are moving on classical trajectories whose time evolution (in the cms system) is interrupted for a time interval ΔT during which the two nuclei stick together. The electronic states are described by quasimolecular orbitals which evolve during the collision around the two charge centers from separated-atom (SA) states to united-atom (UA) states and back again to SA states. In particular, both the $1s\sigma$ and $2p\sigma$ molecular orbitals are correlated to the SA K shell. The inner-shell ionization probability due to the time-varying Coulomb field produced by the two nuclei is given by the following expression:[4]

$$P_K=\int_0^\infty d\epsilon \sum_{i,f} \|a_{if}\|^2 \ ,$$

where ϵ denotes the kinetic energy of the ionized electron and i the initial and f the final state of the system. The ionization amplitude a_{if} is defined as

$$a_{if}(\epsilon)=\int_{-\infty}^{+\infty} dt\, M(t,\epsilon)e^{i\int_{-\infty}^{t}\omega(t')dt'} \ ,$$

where $M(t,\epsilon)$ is the matrx element of the ionization process, $E(t)$ the MO binding energy, and

$$\hbar\omega=E(t)+\epsilon \ .$$

Here, $E(t)$ varies as the two nuclei approach, changing from the SA-binding to the UA-binding energy. The integral can be split into three parts by integrating first from $-\infty$ to 0, from 0 to the sticking time ΔT, and from ΔT to $+\infty$. The second part of the integral is zero since the

Coulomb field responsible for the ionization process remains constant during the sticking time. The remaining two terms can then be written in the following way:[2,3]

$$a_{if}(\epsilon)=a_{in}+a_{\rm out}e^{i\omega(0)\Delta T}\ ,$$

where $a_{\rm in}$ denotes the ionization amplitude in the input channel and $a_{\rm out}$ that in the outgoing channel. A time delay due to a nuclear sticking effect introduces simply an additional phase shift between the two ionization amplitudes. According to this formula, pronounced interference effects are expected to occur for $\omega(0)\Delta T=1$, even after integrating over all final electron energies ϵ.

Using the formalism developed for positron pair creation in heavy-ion collisions, Müller *et al.*[5,20] succeeded in calculating P_K as a function of the delay time ΔT. The left-hand part of Fig. 8 shows the experimental results for $P_K^{\rm at}(-Q)$ obtained for U + U, whereas the right-hand part gives the predicted $P_K(\Delta T)$. The calculations have been made for one fixed impact parameter $b\simeq 6.9$ fm only, but it is safe to assume that this impact parameter represents fairly well the angular range of $45°\pm 13°$ and the Q-value range covered in our experiment.

In order to establish a relationship between Q values and the atomic delay time, we had to normalize the absolute scale of $P_K(\Delta T)$ to the experimental data at $Q=\Delta T=0$. This normalization factor was found to be 0.97, i.e., remarkably close to 1. The similarity between the experimental Q value and the corresponding theoretical ΔT dependence is evident and leaves no doubt that the predicted interference effect between the inner-shell ionization amplitudes not only exists, but also that the predicted sticking times for deep inelastic processes are indeed of the right order of magnitude, as will be discussed in the following.

The relationship between Q value and reaction time established by the correlation between the experimental data for $P_K^{\rm at}(Q)$ and the predictions for $P_K(\Delta T)$ is displayed in Fig. 9. As can be seen, the sticking time is of the order of 10^{-21} s for Q values around 100 MeV. It should be noted that the sticking time ΔT as defined in the quasimolecular picture discussed above corresponds to the time interval during which the Coulomb field experienced by the inner-shell electrons during the collision remains constant in time. In nuclear physics, particularly in deep inelastic scattering, different delay definitions are used. For instance, Schmidt *et al.*[19] defined the interaction time ΔT as the difference between the interaction time for the reaction at a given Q value and that for the elastic channel, i.e., $\Delta T=\tau_{\rm int}-\tau_{\rm int}(Q=0)$. For comparison, the results of two calculations based on this definition and the classical nuclear friction model are also given in Fig. 9. The solid

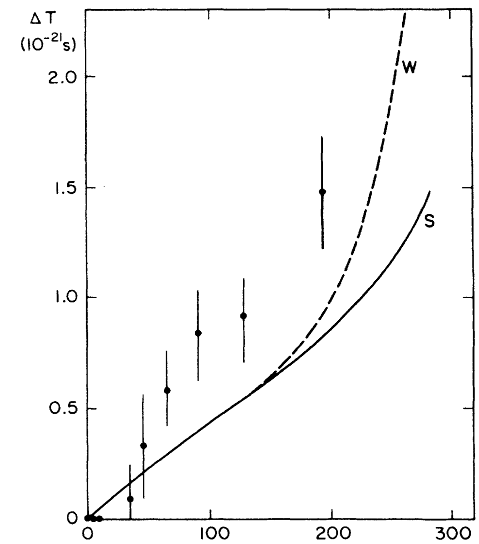

FIG. 9. Semiempirical relation between ΔT and Q for U + U as inferred from Fig. 8 and as calculated in Ref. 19 (S) and Ref. 21 (W).

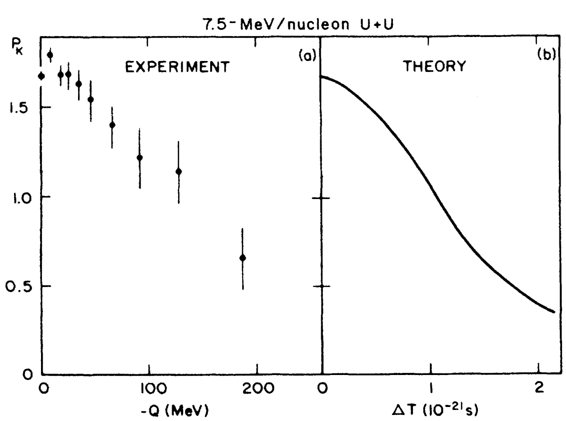

FIG. 8. (a) Experimental atomic-ionization probability $P_K^{\rm at}(Q)$ and (b) predicted $P_K(\Delta T)$ rescaled from Ref. 5.

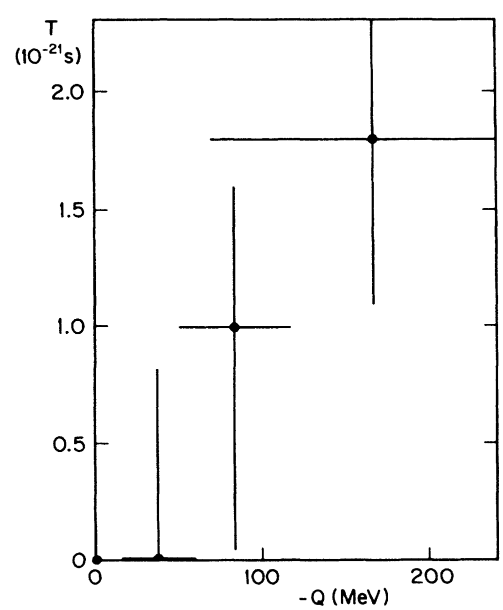

FIG. 10. Same as Fig. 9, but for the system U + Pb.

line indicates results of Ref. 19 and the dashed line those obtained by Wolschin *et al.*[21] The observed correlation between sticking time and Q value is surprisingly close to the theoretical predictions.

The same considerations are valid for the U + Pb system. The results are displayed in Fig. 10. Since the quasimolecular ionization probability $P_K(\Delta T)$ has not yet been calculated for U + Pb at 1766 MeV, we have rescaled its values from an existing calculation made by Müller[5] for Pb + U at 8.97 MeV/nucleon. The fact that we are using the results evaluated for the inverse process should not affect the interpretation, because the interference pattern depends only on the energy of the MO levels and the sticking time. Although the statistical errors are large here, these data suggest that the reaction times for U + Pb might be of the same order of magnitude as for U + U at the same Q value. Time delay calculations based on nuclear physics models are not yet available for the U + Pb system.

VII. CONCLUSIONS

The aim of this experiment was to study the deep inelastic reaction processes in high-energetic U + U and U + Pb collisions and to examine the possibility of using the predicted interference effect in MO inner-shell ionization to determine the nuclear-reaction or sticking time of this particular type of reaction. By measuring the K-shell ionization probability of the projectile- and target-like reaction products, P_K, as a function of the total kinetic energy loss (TKEL), or Q value, in a multiparameter experiment, we have established the existence of these interference effects and proved that time information can be deduced from it, provided the time dependence of P_K is known. This time dependence of the inner-shell ionization probability can be calculated by assuming that inner-shell molecular orbitals are formed during the highly energetic, but from the point of view of the inner-shell electrons quasiadiabatic, collision process. The results of these calculations do not depend strongly on the details of the nuclear reaction mechanism, except for the time delay it introduces.

Using these predictions and our experimental results, we found that the magnitude (typically 10^{-21} s for a TKEL of 100 MeV), as well as the Q dependence of the reaction time, agree quite well with the sticking times obtained from the classical nuclear friction model calculations.

The influence on atomic shells by nuclear reactions has been studied so far only in rather simple cases such as proton capture on Cd and elastic proton scattering on more or less well defined nuclear resonances.[1-3] We have shown here that the same method can be applied even to heavy nuclei interacting in a very complex way. The observed effects are large and well defined, provided colliding systems are chosen for which the condition $\omega(0)\Delta T \simeq 1$ is satisfied. This means that the inverse of the binding energy (in appropriate units) of the inner-shell electrons for the united-atom system must be of the same order of magnitude as the reaction time. Furthermore, it is required that the Coulomb-induced ionization process dominates the other excitation processes, such as γ-ray excitation or inner conversion. Finally, the target- and projectile-like reaction products should have a finite probability of surviving fission, so that the inner-shell electrons have enough time to readjust to the nuclear charge of the reaction products and to emit the characteristic K x rays. Of course, all of these conditions restrict the application of our new time measuring method to a very limited number of possibilities. In a more sophisticated experiment, performed most recently at the SUPERHILAC in Berkeley, we have studied the possibility of using three-body events to gain time information.[22] The results of this investigation[23] indicate that our method even works if one of the reaction products fissions immediately after the nuclear collison, meaning that at least the range of very deep inelasticities can also be studied with this new method.

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