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Interference Effects in the Quasimolecular *K* X-Ray Production Probability for 10-MeV Cl^{16+}-Ar Collisions

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The impact-parameter dependence of the quasimolecular K x rays emitted in collisions of 10-MeV Cl^{16+} with Ar has been measured. The coincidence spectra show a clear impact-parameter–dependent structure. The results are in good agreement with dynamical calculations which take into account the interference between the $1s\sigma$ decay amplitudes in the incoming and the outgoing halves of the trajectory.

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Quasimolecular x rays have been the subject of many studies over the past years,[1] not only for their intrinsic interest, but also because they potentially can be used to study transient molecular orbitals formed during ion-atom collisions. However, in practice this has been more difficult than expected partly because, as a result of the collision broadening,[2] the observed x-ray spectra exhibit a structureless shape[3-6] with a nearly exponential falloff essentially independent of the impact parameter.[6]

In the present experiment we have substantially and radically improved the experimental conditions used for the study of quasimolecular K x rays. These improvements have resulted in a strong impact-parameter–dependent structure in the noncharacteristic x-ray spectrum and have allowed the first comprehensive comparisons with theoretical calculations of transition probabilities. The essential and new improvement that has been made is the use of low-velocity hydrogenlike ions produced by the tandem acceleration-deceleration method[7] which made it possible for the first time to bring a K vacancy into the collision. As a result of the low velocity, the collision broadening (which is proportional to the velocity[2]) is reduced, and the adiabaticity condition[8] is better fulfilled. Furthermore, with decreasing velocity the collision time increases and thus the decay rate of $1s\sigma$ vacancies increases. In nearly symmetric collision systems, the presence of a K vacancy in the projectile gives a probability close to 0.5 for production of a vacancy in the $1s\sigma$ orbital. These conditions are expected to produce a considerable increase in the quasimolecular radiation cross section.[9]

Finally, another important effect is expected when a $1s\sigma$ vacancy is brought into the collision. The vacancy can decay with equal probability in the incoming or the outgoing half of the trajectory. Dynamical calculations[10-13] predict interference structures in the x-ray spectrum, which result from the coherent sum of the two corresponding amplitudes, and which could be observable through measurements as a function of impact parameter.

For convenience the experiment was carried out with a Cl^{16+} beam on Ar at 10 MeV. Although the K vacancy is in the lower-Z collision partner, the binding energy of K electrons in a H-like Cl ion is comparable to the binding energy of the K electrons in a neutral Ar atom. There is therefore a probability of ~ 50% to transfer the vacancy into the $1s\sigma$ orbital as a result of the K-vacancy sharing[14] in the incoming part of the trajectory.

The 10-MeV Cl^{16+} beam was obtained at the

Brookhaven National Laboratory from the dual MP tandem accelerator facility operated in a four-stage acceleration-deceleration mode[7] in the following way: Chlorine ions were accelerated in the two stages of the first MP tandem accelerator which was operated at + 8.4 MV at the terminal, and in the first stage of the second MP tandem accelerator which was operated at a negative high voltage of − 8.2 MV at the terminal. The last stage was then used to decelerate the Cl ions. By selecting charge states of 8^+ and 16^+ produced by the foil strippers in the first and the second accelerators, respectively, a Cl^{16+} beam at 10 MeV was obtained. The analyzed beam intensity varied between 3.9×10^8 and 2.3×10^9 particles/sec.

An Ar gas target cell with a three-stage differential pumping system was used in order to ensure single-collision conditions and to preserve the charge state of the incoming beam. The first and second stages were 20 and 30 mm long (in the direction of the beam) with pressures of 3×10^{-4} and 3×10^{-5} Torr, respectively. The pressure in the last stage was below 10^{-6} Torr. The scattered particles were detected simultaneously at sixteen angles with a position-sensitive parallel-plate avalanche counter, with a 2π azimuthal geometry.[15] The detector covered the impact-parameter range between 350 and 3200 fm. The uncertainty in the impact parameter due to the angular acceptance of the detector and to the gas target length is ~10%. The x rays were detected at 90° with respect to the beam direction with a 200-mm² Si(Li) detector which covered a solid angle of 4.5%. An Al absorber of 38 μm thickness was mounted in front of the Si(Li) detector to absorb the huge yield of projectile K x rays. Coincidences between scattered particles and x rays were recorded event by event. The true/random coincidence ratio was better than 20/1 for x-ray energies $E_x > 4.5$ keV.

The absolute probability of photon production for an impact parameter, b, was obtained from the yields of scattered particles, $N_p(b)$, and of true coincidences $N_x(b,E_x)$, by use of the relation

$$P(b,E_x) = \frac{N_x(b,E_x)\epsilon(E_x)}{N_p(b)\Delta\Omega_x},$$

where $\Delta\Omega_x$ is the x-ray detector solid angle, and $\epsilon(E_x)$ is the x-ray absorption factor. This relation assumes that the x rays are emitted isotropically and that the same target thickness is seen by the x-ray detector and the particle detector. This last assumption introduces a systematic error since the target length seen by the particle detector is larger than that seen by the x-ray detector. This geometrical factor is, however, difficult to calculate accurately because of the variation of the gas pressure along the interaction region. We estimate that the probabilities calculated with the above relation can be underestimated by a factor of ≲ 2.

Figure 1 shows the measured x-ray singles spectrum in 10-MeV Cl^{16+} + Ar. The main peak at 3.5 keV is dominated by radiative electron capture (REC) into the projectile K vacancy. The Cl K x rays are absorbed. The shoulder at $E_x \approx 6.5$ keV is due to pileup of REC pulses, and therefore it does not contribute to the coinicidence spectra. The two-electron–one-photon transitions of Cl and Ar, expected at ~ 5.6 and ~ 6.3 keV, respectively, cannot be observed in Fig. 1 because of their relatively low probability.[16]

The x-ray production probability as a function of photon energy is shown in Fig. 2 for various impact parameters. The error bars indicate the statistical errors only. For impact parameters below about 2250 fm, the conicidence spectra show a pronounced and broad maximum which moves toward higher x-ray energy as the impact parameter decreases. The peak moves from ~ 6 keV at 2600 fm to ~ 9 keV at 500 fm. The united-

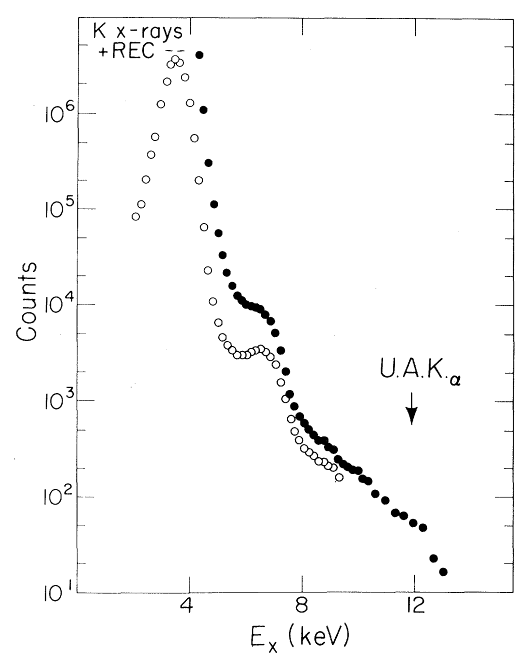

FIG. 1. Singles x-ray spectrum measured (open circles) in 10-MeV Cl^{16+} on Ar. The solid circles show the same spectrum after absorption correction.

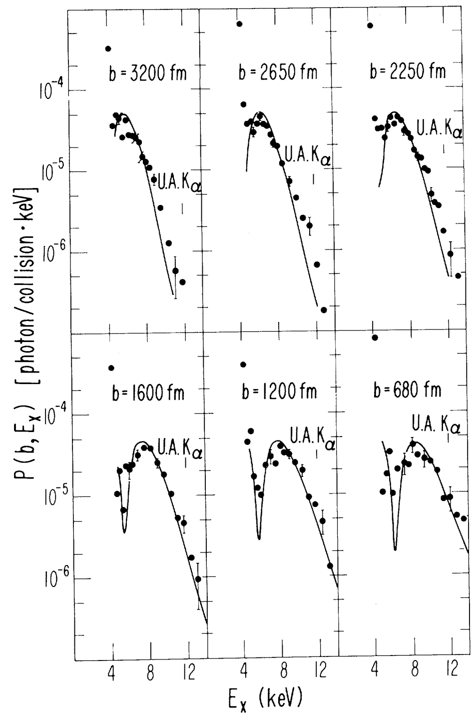

FIG. 2. X-ray production probability as a function of photon energy for various impact parameters. The arrows indicate the united-atom (U.A.) K_α transition energy. The solid lines are the results of dynamical calculations (Ref. 13).

atom K_α transition energy (indicated by the arrow in Fig. 1) is at 11.9 keV. The united-atom K-shell radius is roughly 1500 fm so that the onset of this shifting high-energy maximum corresponds to impact parameters which are small enough to form the united atom. These results differ markedly from those obtained in previous studies of quasimolecular K x-ray production probability (35-MeV Cl + NaCl,[6] 90-MeV Ni + Ni,[17] 143-MeV Nb + Mo[18]) which used projectiles with incident charge state $q > Z - 2$. In those studies the coincidence spectra were similar in shape to the singles x-ray spectrum and showed no structure and almost no dependence on the impact parameter.

Figure 3(a) presents the production probability integrated over E_x whereas Figs. 3(b)–3(d) show the impact-parameter dependence for three intervals of E_x. It is seen that the probability for low-energy photon emission is highest for collisions with large impact parameters, whereas the probability for emission of a high-energy photon comes essentially from the small–impact-parameter region.

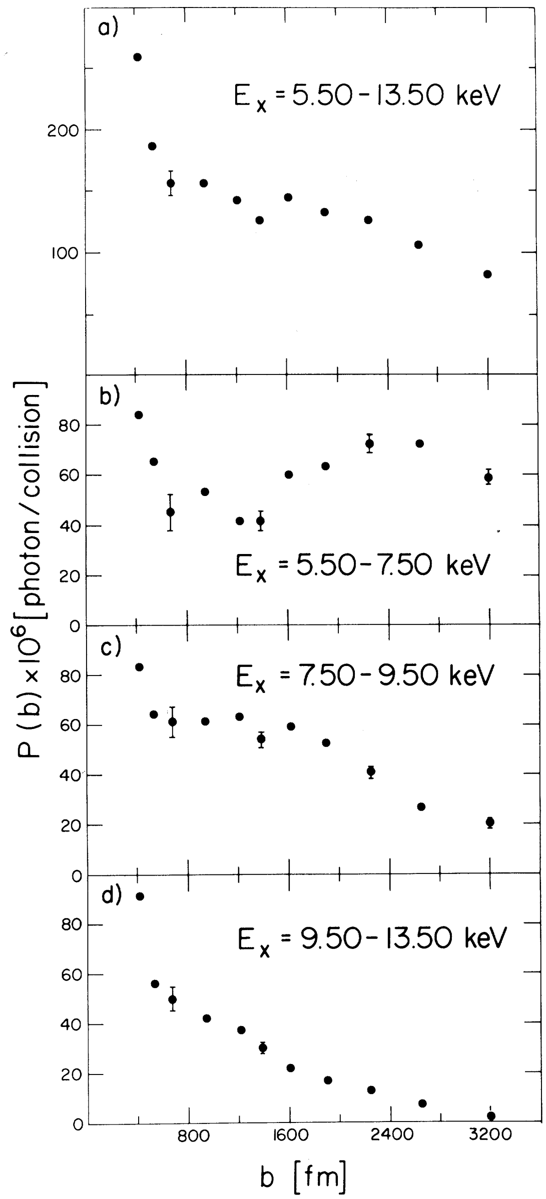

FIG. 3. (a) Total production probability of quasimolecular K x rays as a function of impact parameter. (b)–(d) Probability as a function of impact parameter for three intervals of photon energy.

The solid lines in Fig. 2 represent the results of dynamical calculations[13] which include transitions only from the $2p\sigma$ and the $2p\pi$ orbitals to the $1s\sigma$ orbital. The calculations gave the probabilities in arbitrary units; however, the same normalization factor was used for all impact parameters in Fig. 2. The calculations reproduce many of the observed features of the experiment. In particular, the width and position of the peak are well reproduced. The structure in the calculated curves arises from the interference between the $1s\sigma$ decay amplitude in the incoming part and the outgoing part of the trajectory. In the present system the calculated quasimolecular K x-ray spectrum goes through only one oscillation between 6 and 12 keV. The strong minimum predicted by the calculations is substantially less pronounced in the experiment. The folding of the theoretical curves with the experimental resolution for b does not produce a better agreement since the position of the minima moves very slowly with b as can be seen in Fig. 2 (for $b < 2000$ fm).

In summary, we have measured the impact-parameter dependence of the quasimolecular K x rays emitted in Cl-Ar collisions, using optimal experimental conditions, namely a H-like Cl projectile at a low incident energy of 10 MeV. We observed a pronounced structure in the radiation spectral shape which varies clearly with impact parameter. On the basis of a comparison with dynamical calculations we attribute the structure to the interference between the $1s\sigma$ decay amplitudes on the incoming part and the outgoing part of the trajectory. This is the first time that such structure has been observed in quasimolecular K x-ray spectra. The energy spectra of electrons emitted in 450-keV Kr + Kr collisions[19] show oscillations which were attributed to a similar origin, i.e., the interference between the decay amplitudes in the incoming and the outgoing parts of the trajectory. The emission of electrons was attributed[19] to the autoionization (which is analogous to the emission of molecular-orbital x-rays) of $4p\pi$ vacancies in the Kr-Kr quasimolecule.

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