Impact-Parameter Dependence of the Probabilty for Copper K-Shell Ionization by Oxygen Projectiles*

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The experimental impact-parameter dependence of the probability for copper K-shell ionization by oxygen projectiles with energies between 25 and 43 MeV is presented. The shape of the experimental curve at 43 MeV isfound to be quite similar to one calculated in the semiclassical Coulomb approximation.

The description of the ionization of the inner shell of an atom in violent collisions with fast projectile ions as a direct Coulomb ejection process has enjoyed considerable success in cases where the point-charge character of the projectile dominates. Plane-wave Born-approximation Coulomb ionization calculations[1] and classical binary-encounter (BEA) calculations[2] are in remarkably good agreement with cross sections for K-shell ionization by light projectiles such as protons and α particles. The agreement between theory and experiment becomes even more impressive when binding and Coulomb deflection effects are included,[3] apparently extending the appropriateness of the Coulomb excitation description to include ionization by heavy ions, so long as the projectile charge (Z) is much less than that of the target.

If the de Broglie wavelength of the projectile is small compared to the dimensions of the orbit from which an electron is to be ejected, the ionization of the inner shell of a target by a fast projectile may be treated as if the incident ion were a classical one following a well-defined orbit during the encounter. Indeed, the classical binary-encounter ionization calculation, when done in the impact-parameter formalism[4] (as opposed to the total cross-section calculation of Ref. 2), and the semiclassical Coulomb approximation (SCA)[5] are based on the validity of this picture. Experimental information on the variation with impact parameter (b) of the ionization cross section provides a more detailed examination of the excitation mechanism than does a total cross-section measurement. In the case of heavy-ion projectiles such measurements are further of great value in attempts to understand multiple ionization processes. Experimentally, Stein *et al.*[6] have reported the impact-parameter dependence of the probability for ionization of the L shells of Te and I in Te-I collisions, although this situation of nearly matching target and projectile nuclear charge is one in which a molecular promotion scheme is likely to be the dominant excitation mechanism and is quite different from that encountered in the present experiment. Laegsgaard, Andersen, and Feldman[7] have reported experimental values for the K-shell ionization probabilities of Ag and Se by 1–2-MeV protons as a function of b and find good agreement with SCA predictions. Brandt, Jones, and Kraner[8] have confirmed this result with measurements of ionization probabilities for 0.3–3-MeV protons on Al, Ca, Ni, and Ag. It is the purpose of this paper to report the experimental probabilities as a function of impact parameter for ionization of the copper K shell by oxygen ions and to explore to what extent the success of the Coulomb mechanism for the case of light projectile impact is reproduced when the projectile is a heavy ion whose nuclear charge is much less than that of the target.

Thin (15–46 $\mu g/cm^2$) Cu foils, backed by 5 $\mu g/cm^2$ of carbon, were bombarded by oxygen beams of 25, 35, and 43 MeV (charge states 4^+, 6^+, and 7^+, respectively) from the Kansas State University tandem Van de Graaff. A Si(Li) detector (resolution 200 eV at 5.9 keV) located 1.5 cm from the target detected the copper K x rays at 90° to the beam, while a surface-barrier detector located 85 cm downstream detected in coincidence the scattered oxygen ions. The beam was collimated by a 1-mm aperture located 3 cm before the foil. Several collimators were used on the surface-barrier detector, ranging in dimension from 1×1.5 mm^2 for the smaller angles to 2×8 mm^2 for the larger angles, the smaller dimension being measured in the direction of the oxygen-ion deflection. This detector was mounted on a micrometer head so that it could be scanned a distance of 2.5 cm perpendicular to the beam direction, corresponding to a range in angular deflection of the oxygen ion of 0° to 1.7°. The impact parameter characterizing the Rutherford scattering of bare nuclei is related to the laboratory deflection angle, θ, of the projectile, for small θ, by $b = r_0/\theta$, where $r_0 = Z_1 Z_2 e^2/E$, Z_1 and Z_2 are the projectile and target nuclear charges, respectively, and E is the laboratory energy of the incident particle. At our energies the relationship between b and θ calculated using a screened Coulomb potential[9] differs from that given by the simple expression above by at most 4% at an impact parameter of 0.03 Å. Experimental measurements of the differential scattering cross section showed that for $b < 0.03$ Å ($\theta > 0.15°$ at 43 MeV) the scattering indeed was Rutherford over three orders of magnitude decrease in cross section. That single scattering events were primarily responsible for the angular deflections greater than 0.15° was verified by noting that neither the ratio of single Cu K x rays to scattered oxygen ions nor the measured K-shell ionization probabilities changed when the copper foil thickness was increased from 15 to 46 μg/cm^2. Scattering due to carbon and oxygen in the target was measured using Rutherford scattering at larger angles where energy resolution of ions scattered by the different target masses was possible, and a corresponding correction, never exceeding 10%, was made to the small-angle data.

The $K\alpha$ and $K\beta$ x rays of Cu recorded with no coincidence requirement (hereafter referred to as "singles" x rays) were shifted to energies higher than their nominal characteristic ones, due to multiple ionization of the Cu L and M shells. Both the shifts and the $K\alpha/K\beta$ intensity ratios were observed to be in agreement with those given by Richard.[10] The x-ray spectrum taken in coincidence with scattered oxygen ions displayed the same shifts, with a precision near 20 eV, and the same $K\alpha/K\beta$ intensity ratio as did the singles spectrum. This result is not surprising, since our coincidence requirement selects sufficiently penetrating collisions that one does not expect a large variation of L- and M-shell ionization with variation of the impact parameter over a range already much smaller than the radii of these shells. This observation gives us confidence that the fluorescence yield for the copper K x ray (ω_k), is not strongly b dependent.

The probability per electron for K-shell ionization at a fixed impact parameter, $P(b)$, was found by measuring the ratio of the number of

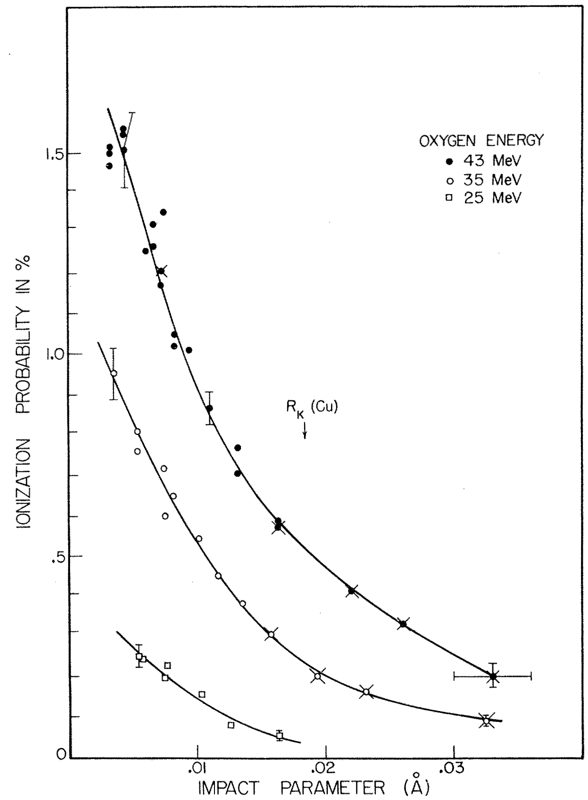

FIG. 1. Experimental probability per K electron for the ionization of the K shell of copper by oxygen projectiles as a function of impact parameter. Data points with and without crosses were taken using 15- and 46-μg/cm^2 copper targets, respectively. The absolute scale is uncertain to ±50%.

coincident Cu K x rays to twice the number of scattered oxygen ions and dividing by the efficiency of the x-ray detector and by the appropriate fluorescence yield. Our results are shown in Fig. 1. For simplicity we have chosen to take the atomic fluorescence yield ($\omega_k = 0.44$)[11] in calculating $P(b)$, although the correct value is dependent upon the state of ionization of the emitting Cu ion.

Figure 2 shows a plot versus b of the "radial" distribution of the cross section, $bP(b)$. We have calculated total cross sections from $\sigma = 2 \times 2\pi \int bP(b)\,db$ and display the results in Table I. Extrapolations of $P(b)$ beyond the range of our data were needed for this calculation, and are shown as solid curves in Fig. 2. We show also the cross sections deduced from our measured singles spectra using the assumed Rutherford scattering of the oxygen ions at 1° as a measure of the product of the number of beam particles

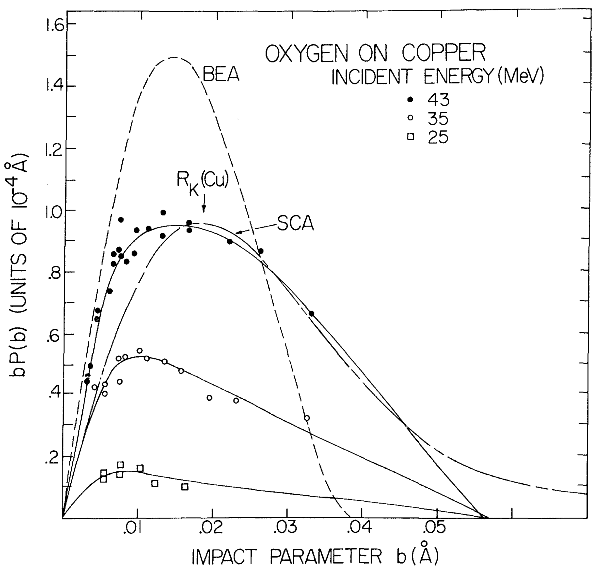

FIG. 2. Plot of $bP(b)$ versus b. The solid curves were used to calculate total ionization cross sections given in Table I. Theoretical curves (for 43 MeV, normalized to the total experimental cross section) are from Ref. 13 (BEA) and made using approximate expressions given in Ref. 8 (SCA).

and the target thickness. Included for comparison are the total σ's calculated for Coulomb ionization under the classical binary approximation.[4] We estimate that our absolute cross-section scale may be in error by as much as 50%, although our overall internal consistency is much better than this. The ratio of our total x-ray production cross sections to those of Sachtleben, Duggan, and Chaturvedi[12] is 0.6, to those of Richard,[10] 0.3; the reason for the discrepancy among the three results is not at present understood.

Theoretical curves for $bP(b)$ at 43 MeV shown in Fig. 2 are from an SCA calculation carried out using the approximations given by Brandt, Jones, and Kraner[8] (dash-dotted curve) and a BEA calculation made by McGuire[13] using a coordinate-space density function for the initial electron distribution (dashed curve). In order to emphasize comparison between theoretical and experimental *shapes* for $P(b)$ we have normalized the theoretical curves to give the experimental total cross sections. Since disagreement between theoretical and experimental total cross sections becomes worse at lower energy, the shape comparison is made only for the 43-MeV data. Although the formulas used for the SCA

TABLE I. Theoretical and experimental cross sections for copper K-shell ionization by oxygen.

Oxygen energy (MeV)	Ionization cross section (10^{-21} cm^2) This experiment [a] $4\pi\int bP(b)db$	σ_K (singles)	Theory σ_K (BEA)[b]
25	0.48	0.44	3.2
35	1.96	2.31	7.0
43	4.24	4.91	10.9

[a] ω_k taken as 0.44.

[b] Ref. 4. The BEA and SCA total cross sections are nearly equal between 25 and 43 MeV.

calculation are approximate ones which already begin to break down for 2.7 MeV/amu, the shape of the SCA curve is remarkably good. On the basis of comparisons of predictions from the approximate formulas with the published full SCA calculations of Hansteen and Mosebekk[14] at slightly higher projectile velocity, we expect that the full calculation would give even better agreement. The description of the data by the BEA curve appears to give insufficient weight to large b.

There are clearly features of the K-shell ionization by low-Z ions of higher-Z targets which do not find immediate explanation in the framework of direct Coulomb ionization, perhaps the most perplexing of these being the dependence of the cross section on the charge state of the projectile.[15,16] Indeed, in the present experiment the projectile charge-state appropriate to the collision is presumably primarily characteristic of the equilibrium state of the ion in the solid. At least part of the discrepancy between experimental and theoretical cross sections may be due to screening of the nuclear charge of the projectile, an effect which would be larger at lower projectile energy. Our results nevertheless indicate that even for heavy ions, so long as the projectile Z is much less than that of the target, the impact-parameter dependence of the cross section predicted on the basis of a Coulomb-ionization mechanism is very close to the correct one.

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Optically Detected Electron Spin Locking and Rotary Echo Trains in Molecular Excited States

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Electron spin locking, rotary echoes, and rotary echo trains have been detected for molecules in their excited triplet states by monitoring the microwave-induced modulation of the phosphorescence. The preliminary results of these studies suggest that nuclear and electron spins in ground and excited states, acoustic phenomena in molecular solids, energy transfer, spin-exchange, nuclear polarization, and a wide variety of additional processes can be optically detected, measuring as few as 10^4 spins.

Although optical detection of spin resonance in excited states has developed as a direct outgrowth of the original experiments of Brossel and Bitter[1] and Kastler,[2] the systems that one has been able to investigate have been limited, for the most part, to excited states that exhibit fluorescence as in gaseous atomic states[3] or phosphorescence as is found, for example, in the spin-forbidden transitions associated with ions[4] or excited triplet states.[5] What would be particularly useful would be a technique utilizing the fluorescence or phosphorescence from these excited states to detect optically electron or nuclear spins in nonradiative excited states or in ground states or to study with the sensitivity of optical detection other phenomena associated with solids such as their acoustic properties,[4,6] the dynamics of energy transfer, or electron and nuclear spin diffusion.

In developing such techniques, one can take advantage of the spin distributions in many of these spin systems. This is particularly true for triplet states in zero field where optical excitation into the singlet manifold followed by selective intersystem crossing into the individual zero-field triplet spin sublevels results in a highly aligned spin state.[7] A prerequisite for utilizing changes in the spin alignment of the excited states to detect optically other radiative or nonradiative processes is that the effective relaxation time associated with the excited triplet state spins be long compared to the time required to contact the electron spins to another spin reservoir or long compared to the correlation time of the spins with other phenomena that can affect relaxation. Unfortunately the electron spin relaxation time T_2 in excited triplet states is only on the order of microseconds because of fluctuating local fields due to nuclei in the lattice on molecules adjacent to the excited state. We wish