Projectile Charge and Velocity Dependence of L-Subshell Ionization Cross Section Ratios for Heavy Elements*

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L-subshell ionization cross-section ratios for heavy elements induced by protons, α's, carbon ions, and oxygen ions in the range 0.5 to 6.0 MeV/amu have been investigated. The resulting ratios of $\sigma_{L_1}/\sigma_{L_2}$ and $\sigma_{L_3}/\sigma_{L_2}$ show large deviations from the plane-wave-Born-approximation, semiclassical-approximation, and binary-encounter-approximation predictions below 2 MeV/amu. These deviations are more pronounced for higher projectile charges.

In their study of L-subshell ionization cross sections for gold bombarded by protons and helium ions, Datz *et al.*[1] observed deviations of the low-energy helium-ion results for the L_3 to L_2 cross-section ratio, from the theoretical predictions of the plane-wave Born approximation (PWBA).[2] They suggested these discrepancies might be due to the effects of multiple ionization. Recently, Chang, Morgan, and Blatt[3] also reported a similar effect for α's on platinum and gold, but not for other elements in the region between $Z = 73$ and 83. Chang, Morgan, and Blatt[3] argued against the multiple-ionization mechanism since this should affect all elements in this narrow range of atomic number in a similar fashion. Instead, they suggested it was an electron-configuration effect due to the presence of single 6s electrons in Pt and Au. We report here measurements of $\sigma_{L_3}/\sigma_{L_2}$ and also $\sigma_{L_1}/\sigma_{L_2}$ ratios in heavy elements at low bombarding energies using different projectiles (^{1}H, ^{4}He, ^{12}C, and ^{16}O).

Beams of protons, α particles, carbon ions, and oxygen ions in the range of 0.5 to 6.0 MeV/amu were produced by the University of Minnesota MP tandem Van de Graaff accelerator. X rays emitted from the thin targets (typically ~75 μg/cm^2) were detected using a Si(Li) x-ray spectrometer having a resolution of 180 eV at 6.4 keV. L-subshell ionization cross sections for the target atoms were extracted from the observed $L_{\alpha_{1,2}}$, L_{γ_1}, and $L_{\gamma_{4,4'}}$ x-ray transitions. Details of the experimental arrangement and data analysis are discussed elsewhere.[4]

The resulting ratios of L_3 to L_2 ionization cross sections, $\sigma_{L_3}/\sigma_{L_2}$, for ^{1}H, ^{4}He, ^{12}C, and ^{16}O on uranium, are plotted as a function of bombarding energy per atomic mass unit in Fig. 1. The error bars represent typical uncertainties due to the statistical errors in the number of detected x rays and errors in the solid angle and efficiency of the x-ray detector. They do not include uncertainties in the fluorescence yields, the Coster-Kronig transition probabilities, and the radiative transition rates. Theoretical predictions using the PWBA,[2] the semiclassical approximation (SCA),[5] and the binary-encounter approximation (BEA)[6,7] are also plotted for comparison. The PWBA curves were obtained using the tables of Choi, Merzbacher, and Khandelwal.[8] The SCA curves were taken from the recent calculations of Hansteen, Johnsen, and Kocbach.[9] The BEA curves were computed using Hansen's code BEACON.[10] The results of all three models are very

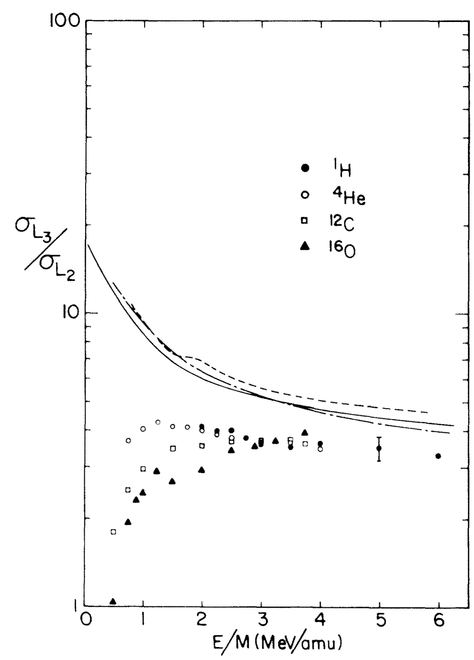

FIG. 1. Ratios of L_3- to L_2-subshell ionization cross sections for uranium resulting from proton, α particle, carbon ion, and oxygen ion bombardment as a function of projectile energy per atomic mass unit. The error bars are discussed in the text. Theoretical predictions using PWBA (solid curve), SCA (dashed curve), and BEA (dashed-dotted curve) are also plotted.

similar, with steeply increasing $\sigma_{L_3}/\sigma_{L_2}$ ratios as the projectile velocity decreases, particularly below 2 MeV/amu. The experimental data show completely different features in this low-energy range, e.g., the α ratios decrease below ~1.25 MeV/amu. The deviations of the experimental $\sigma_{L_3}/\sigma_{L_2}$ ratios from the theoretical predictions become more pronounced as the projectile energy decreases from 2.5 to 0.5 MeV/amu and as the projectile charge increases from two to eight. Figure 2 shows the results for $\sigma_{L_1}/\sigma_{L_2}$ ratios compared with theoretical calculations. Both PWBA and SCA predict that the ratios of $\sigma_{L_1}/\sigma_{L_2}$ have a minimum near 2.7 MeV/amu. This is due to the $2s_{1/2}$ state having a node in the radial wave function both in coordinate space and in momentum space. The results from BEA also show a minimum but at a higher bombarding energy. Deviations from the theoretical prediction are also seen in this case, at low energies, and also become more pronounced with increasing projectile charge. The extent of the departure from the theoretical values is approximately the same for the $\sigma_{L_3}/\sigma_{L_2}$ and $\sigma_{L_1}/\sigma_{L_2}$ ratios for a given projectile and velocity.

In order to examine whether these deviations exist for other heavy elements, we did similar measurements using ^{16}O on Bi, Au, and Ta. The resulting ratios of $\sigma_{L_3}/\sigma_{L_2}$ plotted versus the velocity ratios of the projectile (^{16}O) and of the target electrons, $V_p/\overline{V}_{2,3}$ ($\overline{V}_{2,3}$ are the average velocities of $2p_{1/2}$ and $2p_{3/2}$ electrons in the target elements) are plotted in Fig. 3. The curves are PWBA calculations for various target elements. All experimental data have a tendency to fall onto a common curve which deviates from the theoretical predictions at low velocity ratios. This implies that the same behavior, or ionization mechanism, occurs for a number of elements in the range of atomic number 73 to 92.

In considering effects of multiple inner-shell ionization, we have compared experimental x-ray ratios for specific transitions from different

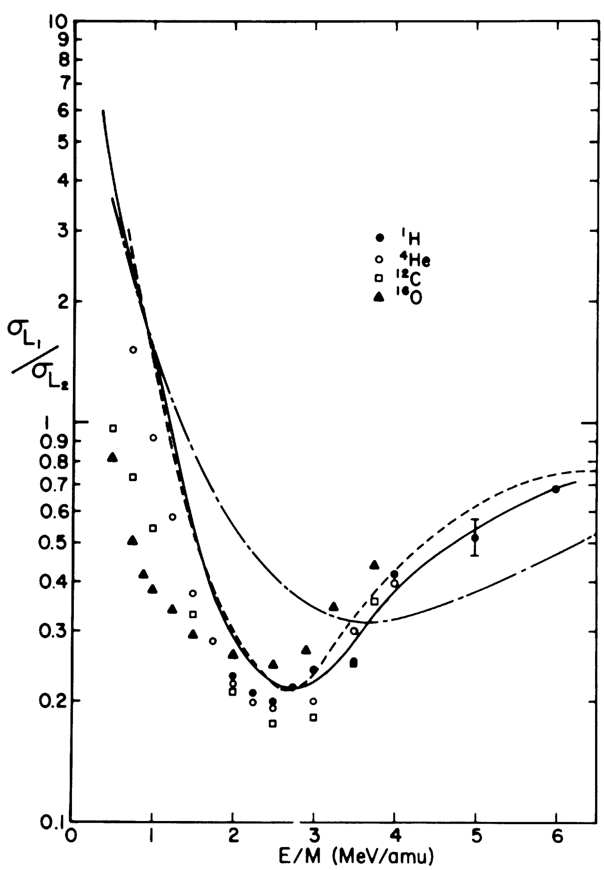

FIG. 2. Ratios of L_1- to L_2-subshell ionization cross sections for uranium. (See Fig. 1 caption for details.)

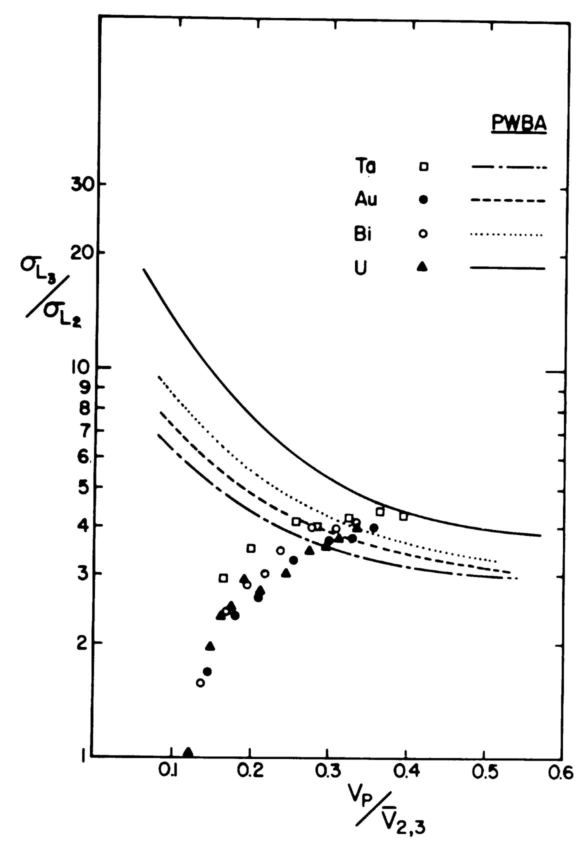

FIG. 3. Ratios of $\sigma_{L_3}/\sigma_{L_2}$ for ^{16}O on Ta, Au, Bi, and U as a function of the velocity ratios of the projectile (V_p) and the target electrons ($\overline{V}_{2,3}$). ($\overline{V}_{2,3}$ are the average velocities of $2p_{1/2}$ and $2p_{3/2}$ electrons in the target elements.) The curves are PWBA calculations for various target elements.

higher shells (e.g., M, N, O ...) to the same L subshells in uranium, with theoretical predictions, based on single ionization in uranium, by Scofield[11] using relativistic Hartree-Fock wave functions for the initial and final electronic states. Within the experimental uncertainties, the ratios of $L_l(L_3-M_1)/L_{\alpha_{1,2}}(L_3-M_{4,5})$, $L_{\beta_{2,15}}(L_3-N_{4,5})/L_{\alpha_{1,2}}(L_3-M_{4,5})$, and $L_{\beta_1}(L_2-M_4)/L_{\gamma_1}(L_2-N_4)$ are constant and agree well with theory. This indicates that simultaneous L-plus-M−, or L-plus-N−shell multiple-ionization effects are small. L-plus−outer-shell (e.g., the O shell) multiple ionizations are observed in the $L_{\beta_{9,10}}(L_1-M_{4,5})/L_{\gamma_{4,4'}}(L_1-O_{2,3})$ ratios, but they do not correspond to the observed deviations for $\sigma_{L_1}/\sigma_{L_2}$. We observed no significant energy shifts or peak broadening of the x-ray spectra, which suggests that the effect of contributions from double L-shell ionization are probably small also.

In summary, we found large deviations for both $\sigma_{L_3}/\sigma_{L_2}$ and $\sigma_{L_1}/\sigma_{L_2}$ ratios from theory below 2 MeV/amu which are dependent on the projectile charge, in the range $Z=73$ to 92. Our results do not support explanations based either on the simultaneous multiple inner-shell ionization,[1] or on the correlation of initial outer-electron configurations of the atoms.[3] We have examined the corrections introduced into the theoretical predictions by binding and Coulomb-deflection effects[12] and these appear to be incapable of explaining the present results. This is not surprising since such corrections would be expected to influence σ_{L_3}, σ_{L_2}, and σ_{L_1} in a similar manner and hence the ratios should be relatively unaffected. Furthermore, the persistence of the deviations observed for protons and α's, where binding and Coulomb-deflection corrections are small, suggests these effects are not the source of the present discrepancies. However, relativistic effects for low-energy projectiles on high-Z elements are expected to be sizeable; and relativistic PWBA calculations with the binding and Coulomb corrections may reduce the discrepancy between theory and experiment. This possibility has not yet been examined in any detail.

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