Excitation of Al K and Hf M X-Ray Lines by 70–400-keV H^+, He^{++}, N^{++}, O^{++}, and Ar^{++} Ions

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The K shell of aluminum and the M shell of hafnium have been excited using H, He, N, O, and Ar ions in the energy range 70–400 keV. The appearance of an excitation mechanism is observed for N, O, and Ar projectiles, which results in ionization cross sections several orders of magnitude greater than those predicted by a direct-scattering interaction such as is assumed for H and He ions. The results are examined in the light of the electron promotion mechanism of molecular-orbital theory as suggested by Der *et al.* The similar increases in cross sections for the hafnium M shell and the aluminum K shell using incident ions heavier than He suggest that an electron promotion mechanism does not provide a satisfactory explanation of the anomalous cross sections. It is felt that other factors, such as polarization of the relevant atomic shells of both the target atom and the projectile at close impact distances, must be considered in terms of the direct-scattering theory.

The use of hydrogen and helium ions for the excitation of characteristic x rays in many elements has been extensively studied in recent years.[1-4] The main theoretical approach has been the use of the Born approximation with unperturbed atomic wave functions used for the electron to be ejected, and plane-wave functions for the incident proton or α particle. The failure of this treatment at low energies has resulted in the addition of terms to account for the deflection of the projectile in the Coulomb field of the nucleus and, recently,[4] the use of a perturbation calculation to account for the partial binding of the electron to the incident projectile. Thus far, only the K-shell interactions have been extensively studied theoretically. There are few L-shell and M-shell calculations available. Recently, Der *et al.*[5] have reported anomalously high x-ray production cross sections for the carbon K shell using several projectile ions heavier than H and He. They have interpreted these results as resulting from an auto-ionization process due to the interpenetration of the electronic shell structure of the projectile with that of the target atom. During this process, electrons would be promoted into higher shells at specific projectile target atom internuclear distances, subsequently leaving the target atom in an ionized state.

In the present experiments, H^+, He^{++}, N^{++}, O^{++}, and Ar^{++} ions in the energy range 70-400 keV have been used to excite x rays from the aluminum K shell and the hafnium M shell. The targets were selected so that atomic shells of comparable energy but varying electron density would be excited. The hafnium M-shell transitions occur between 6.544 and 9.686 Å with the most probable transition occurring at 7.539 Å. The aluminum K line is 8.339 Å.

The ions were obtained from an rf ion source using special research-grade gases and were accelerated by a 300-kV accelerator. The ion beam from the accelerator was focused by a quadrupole lens on the entrance slit of a 10-kg 16-in. radius-of-curvature analyzing magnet. The analyzed beam was collimated to $\frac{3}{16}$ in., and then entered the target chamber through a $\frac{1}{8}$-in. aperture. The high-purity 99.999% metal targets were mounted in an ultrahigh-vacuum target chamber pumped by a 4-in. Orb-Ion pump.[6] The chamber had a residual background of $1-2\times 10^{-9}$ Torr and was maintained at $5-7\times 10^{-8}$ Torr with the beam into the chamber. The target current was integrated using standard suppression techniques and an Ortec current digitizer. The thick targets were polished, mounted, and cleaned using normal methods relevant to ultrahigh-vacuum and surface physics work. The x rays were detected using a gas flow proportional counter with a 6-μ aluminized Mylar window. The transmission of each window was independently measured at each wavelength. The detector pulses were amplified and analyzed by a gated ND 2200 pulse-height analyzer. Each data point presented in this paper represents the mean of three data runs made at one-week intervals, during which time the target was removed, recleaned, and remounted.

The x-ray production cross section σ_x was calculated from target yield I_μ at each ion energy E by the equation

$$\sigma_x = (1/N)\, dI_\mu / dE\, S(E) + (1/N)\, \mu/\rho\, I_\mu \ , \qquad (1)$$

where $S(E)$ is the target stopping power for each particular ion, μ/ρ is the absorption coefficient of each target for its own x-ray line, and N is the number of target atoms per gram.[1] The stopping powers were obtained by using a combination of theoretical and experimental sources.[7-10] Those stopping powers not available as experimental values were calculated by using theoretical equations developed by Lindhard, Scharff, and Schiott,[7] and a technique for heavy ions developed by Steward and Wallace.[8] The experimental values were obtained from Warshaw and Allison[9] and from Northcliffe.[10]

The x-ray production cross section σ_x is related to the ionization cross section by

$$\sigma_I = 1\sigma_x / \bar{\omega}_n \quad , \tag{2}$$

where $\bar{\omega}_n$ is the average fluorescent efficiency of the nth atomic shell. The value $\bar{\omega}_K = 0.04$ for the aluminum K shell was taken from Fink *et al*.[11] This value is the mean of the two most consistent values of the three experimental values given. The value $\bar{\omega}_M = 0.013$ for the hafnium M shell was obtained by extrapolating the few fluorescent yields presently available for the M shell.[11]

The cross sections are plotted in Figs. 1 and 2 as a function of the incident ion energy per amu. The data for H^+ and He^{++} excitation of each target were taken as a comparison to illustrate the anomalously high excitation obtained by using the heavier ions. The uncertainty in the absolute value of the cross sections is estimated to be approximately 30%, due principally to the uncertainty in the val-

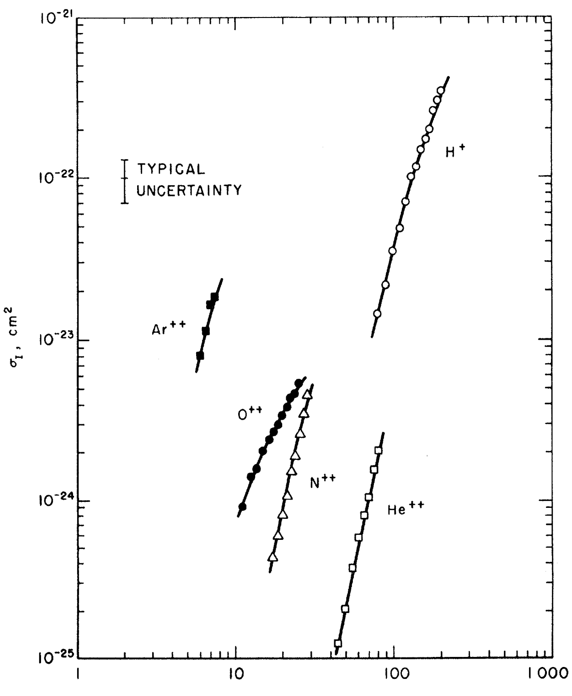

FIG. 1. Ionization cross sectons for the excitation of aluminum K-shell x rays by the impact of H^+, He^{++}, N^{++}, O^{++}, and Ar^{++} ions are plotted as a function of incident ion energy per amu.

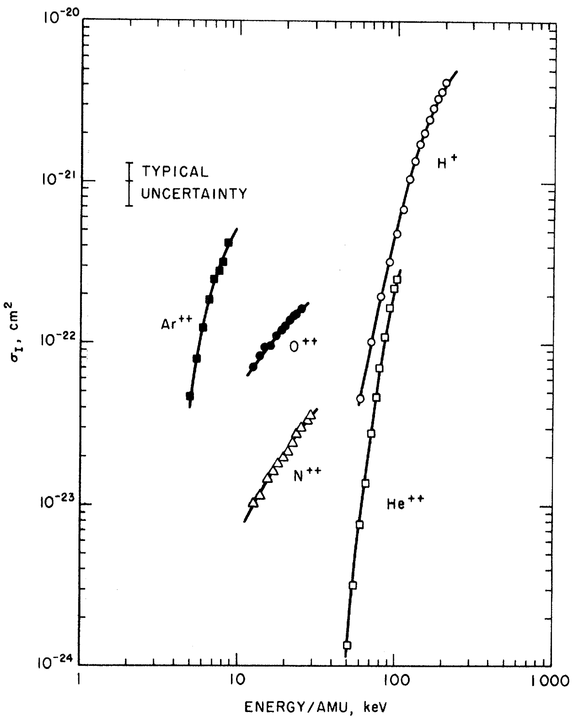

FIG. 2. Ionization cross sections for the excitation of hafnium M-shell x rays by the impact of H^+, He^{++}, N^{++}, O^{++}, and Ar^{++} ions are plotted as a function of incident ion energy per amu.

ues used for the stopping powers and the fluorescent yields. The internal uncertainty, i.e., the relative error in the experimental values, is less than 10%.

The data for H^+ and He^{++} excitation of the Al K shell agree with a direct-scattering theory that has been modified to include corrections for projectile deflection in the Coulomb field of the target nucleus and the partial binding of the aluminum K electrons to the incoming projectile.[4] Although there is little theoretical work available for ionization of the higher shells, it is still evident from Fig. 2 that the interaction mechanism leading to x-ray excitation in the hafnium M shell for H^+ and He^{++} is markedly different from that for N^{++}, O^{++}, and Ar^{++} excitation of the same. However, the close similarity between the behavior of the cross sections for both the Al K-and the Hf M-shell data presented here seems to indicate an interaction for the heavier ions other than the electron promotion mechanism of molecular-orbital theory as suggested by Der *et al*.[5] If one assumes that an approximate molecular system is formed by the incident-ion target atom, the internuclear distances required for K-shell crossings to occur are very small as compared with the internuclear distances required for

L- or M-shell crossings, i.e., the "promotion" of electrons to higher shells.[12] The incident ion spends a significantly larger part of its "interaction time" at impact distances within the critical range required for level crossings of the M electrons into higher shells, and considerably less time at impact distances with the critical range required for K-electron promotion. This leads us to expect that anomalies in the magnitude of the ionization cross section caused by an electron promotion type of mechanism would be significantly more pronounced in the M shell than in the K shell. This, however, seems not to be the case, as can be observed from Figs. 1 and 2 where the magnitude of the increases in yield for excitation of both the Al K and the Hf M shell appears to be quite similar.

In fact, a theoretical comparison of the heavy-ion yields with the yields obtained for hydrogen and helium ions suffers from a further breakdown of the Born approximation which is used as the starting point for present direct-scattering theoretical calculations. One of the conditions for the use of unperturbed atomic wave functions for the electrons to be ejected is that the polarization of the relevant electron orbits in the target atom and projectile can be neglected for low-Z incident ions.[13] Since this condition is not fulfilled in the present experiments, it is felt that the apparent anomalies in the ionization cross sections are a manifestation of the incomplete theoretical work presently available even for the light ions rather than being evidence for a totally new interaction mechanism. We suggest that factors such as distortion of the atomic shells due to polarization must be fully taken into account for high-Z incident projectiles in terms of the direct-scattering theory.

Further experimental work is presently being performed in which several other target-projectile systems were chosen so that the relevant shell radii are matched in order to optimize any level crossing effect.

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