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Projectile Charge Dependence of K-Shell Ionization by Swift Light Nuclei*

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K-shell x-ray yields were measured for Al and Ni bombarded by H, D, He, and Li of kinetic energies between 1 and 7.5 MeV/amu. Ratios of K-shell ionization cross sections for projectiles of different atomic numbers Z_1, at equal velocities, show pronounced deviations from the Z_1^2 dependence predicted in the plane-wave Born approximation. The effect can be understood in terms of distortions of the K shell in the field of the ionizing particles.

This Letter reports ratios of cross sections for K-shell ionization of target atoms of atomic number Z_2 by different energetic charged particles of atomic number Z_1 and mass number M_1. The cross sections $\sigma_{2K}(Z_1)$ were measured for targets of Al ($Z_2=13$) and Ni ($Z_2=28$) under bombardment by beams of ${}_1^1$H, ${}_1^2$D, ${}_2^4$He, and ${}_3^7$Li. The new data confirm the prediction[1] that at high particle velocities the ratio $R_{2K}\equiv[\sigma_{2K}(Z_1')Z_1'^{-2}]/[\sigma_{2K}(Z_1)Z_1^{-2}]$, for $Z_1'>Z_1$, has values larger than 1. The results are shown in Fig. 1, together with earlier data measured at lower particle velocities.[1-4] The Ni data run smoothly into the ratios measured for Cu ($Z_2=29$) K shells by Lewis, Natowitz, and Watson.[5]

The significance of this new effect lies in the fact that it exhibits quantitatively the basic limitations of the plane-wave Born approximation (PWBA) in the theory of ionization cross sections. The origins of this effect can be traced to the finite charge of real ionizing particles.[4]

If $Z_1 \ll Z_2$, the projectiles act as bare point charges when ionizing K shells, and Coulomb excitation dominates.[6] One can suitably scale the cross sections for this process by introducing the wave-mechanical hard-sphere cross section $4\pi a_{2K}^2$ for each of the two electrons in the K shell of radius $a_{2K}\equiv\hbar^2/me^2Z_2$, weighted by the square of the strength of the Coulomb interaction of the electrons with the incoming particle relative to their interaction with the target nucleus, $(Z_1/Z_2)^2$:

$$\sigma_0 \equiv 8\pi a_{2K}^2 (Z_1/Z_2)^2. \tag{1}$$

In the PWBA one calculates, in effect, the scaled cross section $S_{2K}\equiv(\sigma_{2K}/\sigma_0)$ in the limit $Z_1 e\to 0$, with the result

$$S_{2K}^{\mathrm{PWBA}} = \lim_{\sigma_0\to 0}(\sigma_{2K}/\sigma_0) = \eta^{-1} f(\eta,\theta), \tag{2}$$

where $f(\eta,\theta)$ is a dimensionless function of $\eta \equiv v_1^2/v_{2K}^2$ independent of Z_1, $v_{2K}\equiv Z_2 e^2/\hbar$ being the K-shell electron orbital velocity. The parameter $\theta\equiv 2I_{2K}/Z_2^2$, where I_{2K} (in a.u.) is the observed ionization energy of the K shell, grows slowly with Z_2 from ~0.7 for light target elements to ~0.9 for heavy elements. The large range of cross sections under consideration is shown in the nomograph on the top scale of Fig. 1.[7]

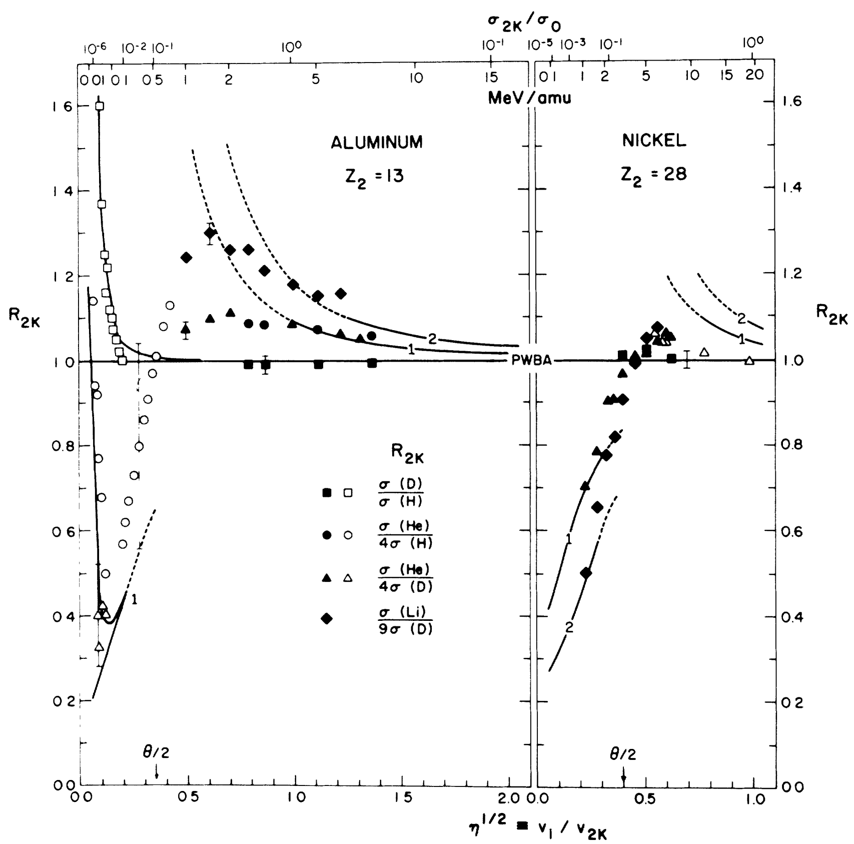

FIG. 1. Ratios R_{2K} of K-shell ionization cross sections as a function of $\eta^{1/2}$ for Al and Ni under bombardment by particles Z_1 and $Z_1' \geq Z_1$. The upper scales give the particle kinetic energy in MeV/amu and a nomograph of the ionization cross section for $Z_1=1$ in units of σ_0, Eq. (1), where $\sigma_0(\text{Al})=27$ kb and $\sigma_0(\text{Ni})=1.2$ kb. Solid symbols represent new data. On the Al plot, open symbols denote data taken from Refs. 1, 3, and 4; for clarity, the two highest points in Ref. 1 have been omitted. On the Ni plot, open symbols represent the data from Ref. 5 for Cu ($Z_2=29$). The new ratios for D/H are equal to 1 within experimental error. The curves depict theoretical Z_1 dependences for small and large η, extended by dashed lines to the value $\eta=\theta^2/4$ where the effect is expected to cancel.[1] The curves marked 1 and 2 represent the predicted dependences of the effect of $Z_1'-Z_1=1$ and 2, respectively.

The proportionality of the cross section to Z_1^2 is characteristic of the PWBA: One assumes the charge of the incoming particle to be so vanishingly small that, during the collision, the particle wave remains planar and the target electron wave function remains an unperturbed eigenstate of the atom. This theory predicts that, for different types of particles Z_1 and Z_1' impinging with equal velocity v_1 on given target atoms Z_2, the ratio $R_{2K} \equiv S_{2K}(Z_1')/S_{2K}(Z_1)$ is always unity.

For projectiles of finite positive charge, significant deviations of R_{2K} from unity are predicted to occur at low and high η.[1-3] Their origins can be understood qualitatively in terms of simple plausibility arguments. In very slow collisions, $v_1 \ll v_{2K}$, K-shell ionization occurs only for deep penetration into the target K shell. In such collisions, the binding charge seen by the electrons to be excited increases, effectively, from Z_2 to Z_2+Z_1. This decreases the cross section. A Taylor series expansion of $\sigma_{2K}^{\text{PWBA}}$ about the unperturbed binding energy, retaining only first-order terms in the change of the binding, results in a *subtractive* Z_1^3-dependent effect in the sense that for $Z_1/Z_2 \ll 1$, $\sigma_{2K}=\sigma_{2K}^{\text{PWBA}} - |O((Z_1/Z_2)^3)|$. By contrast, at particle velocities $v_1 \gtrsim v_{2K}$, K-shell ionizations occur over interaction distances of order $a_{2K}=Z_2^{-1}$ (a.u.) and larger. In terms of the K-shell polarizability $\alpha_{2K} \propto Z_2^{-4}$ (a.u.), the K-shell orbits are distorted in the field of the passing particle, $\mathcal{E} \lesssim Z_1e^2/a_{2K}^2$, by a relative amount $\delta/a_{2K}=\alpha_{2K}\mathcal{E}/ea_{2K} \propto Z_1/Z_2$ since $e\delta=\alpha_{2K}\mathcal{E}$. This shortens the effective interaction distance and thus increases the interaction strength between particle and electron from Z_1 to $Z_1(1-\delta/a_{2K})^{-1}$. When inserted in Eq. (1), an *additive* Z_1^3-dependent effect results in the sense that for $Z_1/Z_2 \ll 1$, $\sigma_{2K}=\sigma_{2K}^{\mathrm{PWBA}}+|O((Z_1/Z_2)^3)|$. One concludes that the ratio R_{2K} for different particles Z_1 and Z_1', where $Z_1<Z_1'\ll Z_2$, should show deviations proportional to $(Z_1'-Z_1)/Z_2$ from the value 1 predicted by the PWBA in the following ways[8]:

$$R_{2K}\equiv\frac{S_{2K}(Z_1')}{S_{2K}(Z_1)}=\begin{cases}1-|O((Z_1'-Z_1)/Z_2)| & \text{for } \eta\ll 1,\\ 1+|O((Z_1'-Z_1)/Z_2)| & \text{for } \eta\gtrsim 1.\end{cases} \tag{3}$$

Given these two domains of η, one expects the effect to change signs between the range of validity of the equations; in fact, it occurs at $\eta=\theta^2/4$.[1]

At very low velocities, R_{2K} depends, as well, upon the charge-to-mass ratio of the particles. This effect is well understood: The mass dependence of the Coulomb deflection of projectiles of equal velocity in the field of the target nucleus favors, through their deeper K-shell penetration, excitation by the projectiles with the lower value of Z_1e/M_1.[2,3]

The ionization cross sections are extracted from measurements of characteristic x-ray yields, with the assumption that the fluorescence yield is independent of Z_1.[9] Extensive series of such data on Al have shown the effect under discussion for $\eta<1$ and confirm in some detail the theoretical predictions of its magnitude.[1-4] In Fig. 1 we report the new experimental results for $\eta\gtrsim 1$ which, together with the earlier data, exhibit the entire η dependence of the effect anticipated by Eq. (3). Details of the data acquisition and the absolute cross sections will be published elsewhere.

Thin targets of Al (20 μg/cm^2) and Ni (180 μg/cm^2) were exposed at 45° to particle beams from the Brookhaven National Laboratory tandem Van de Graaff accelerator. The characteristic K-shell x rays from the target were viewed perpendicular to the beam direction by a Si(Li) x-ray detector. The transmitted beam current, typically 20–50 nA, was measured in a 10-m Faraday cup which also served as a beam dump. K-shell x-ray spectra with peak-to-background ratios of at least 50:1 were obtained. With the assumption that the fluorescence yield is Z_1 independent, the uncertainty in the ratio R_{2K} is ±2% for the present data. The earlier low-energy data have an uncertainty of ±30%.

The Al ($Z_2=13$) data show three distinct Z_1 dependences (Fig. 1). For $Z_1'=Z_1$ (D vs H), R_{2K} is unity within experimental accuracy at intermediate and high velocities, and thus tests the basis of our arguments. For $Z_1'-Z_1=1$ (He vs H or D), R_{2K} rises with increasing η from below unity to values above unity near $\eta=\theta^2/4$, passes through a maximum and slowly approaches unity from above. For $Z_1'-Z_1=2$ (Li vs H or D), R_{2K} follows the same trends over the velocity range studied. The deviations from unity near the maximum are somewhat larger than twice the deviations observed for $Z_1'-Z_1=1$. This could indicate that the experiment is sensitive to higher-order correction terms in just the η range where problems of convergence are expected.[1] Beyond the maximum, the deviations from unity are proportional to $Z_1'-Z_1$ and show asymptotically the predicted decline proportional to $\eta^{-3/2}$.

The rise of R_{2K} observed at the lowest particle velocities shows the influence of differences in the charge-to-mass ratios on the Coulomb deflection of the incoming projectiles. The isotope effect shown for the pair ^{2}D/^{1}H is similar to that of the pair ^{4}He/^{3}He reported earlier.[3] For the pair ^{4}He/^{2}D, of equal charge-to-mass ratio, the influence of the Coulomb deflection essentially cancels, and $R_{2K}<1$ even at very small η, as predicted.[2]

The Ni ($Z_2=28$) data in Fig. 1 exhibit the same trends. At low η they confirm the predicted Z_1 dependence of the R_{2K} decline. At $\eta>\theta^2/4$, the Ni data overlap with and are extended by recent Cu ($Z_2=29$) data.[5] The magnitude of the effect is reduced relative to that observed for Al ($Z_2=13$) as expected from its dependence on Z_2^{-1}.

In summary, experimental K-shell ionization cross sections for swift nuclei of atomic number $Z_1\ll Z_2$, as extracted from characteristic x-ray-yield measurements, show a new, distinct velocity-dependent effect proportional to Z_1^3. The magnitude of the effect, and its opposite sign in the two domains of particle velocities $v_1<v_{2K}$, and $v_1>v_{2K}$, confirm theoretical predictions given earlier.[1,2] The physical origins of the effect are traced to K-shell distortions in the field of the ionizing particle. We conclude that the plane-wave Born approximation is inadequate in significant and basic ways. The experiments point to the need for new approaches to the problem of Coulomb excitation of atoms with approximations that realistically and comprehensively incorporate, *ab initio*, the finite charge of physical projectiles.

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[1]G. Basbas, W. Brandt, and R. Laubert, Phys. Lett. 34A, 277 (1971).

[2]W. Brandt, R. Laubert, and I. Sellin, Phys. Lett. 21, 518 (1966), and Phys. Rev. 151, 56 (1966).

[3]W. Brandt and R. Laubert, Phys. Rev. 178, 225 (1969).

[4]G. Basbas, W. Brandt, and R. Laubert, to be published.

[5]C. W. Lewis, J. B. Natowitz, and R. L. Watson, Phys. Rev. Lett. 26, 481 (1971).

[6]W. Brandt and R. Laubert, Phys. Rev. Lett. 24, 1037 (1970).

[7]When $\eta \ll 1$, S_{2K} rises steeply with η, approximately proportional to $\eta^4\theta^{-9}$. In the range $\eta = 5\times10^{-3}$ to $\eta = 10$, $f(\eta, \theta)$ is known numerically [G. S. Khandelwal, B. H. Choi, and E. Merzbacher, At. Data 1, 103 (1969)]. Near $\eta = 1$, S_{2K} goes through a maximum of order unity and diminishes slowly as $\eta^{-1}\ln\eta$ when $\eta \gg 1$.

[8]It is interesting to note that the signs in Eq. (3) would be reversed if the incoming projectiles were of negative charge.

[9]Testing a conjecture by Lewis, Natowitz, and Watson (Ref. 5), we measured the relative $K\alpha/K\beta$ x-ray yields in Ni and found them to be constant to ±5% for all the projectiles in the particle energy range studied. By comparison, the measured Ni x-ray yield ratios vary over a factor 2 in this range. We take this as evidence that on the scale of the effect under investigation the fluorescence yield is independent of Z_1, at least for small Z_1, as indeed one should expect on theoretical grounds.

Final States in the Dissociative Excitation of Molecular Hydrogen*

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The dissociative excitation $H_2 + e \to H(2s) + H + e$ has been studied with a time-of-flight method. Measurements of the excitation function and the angular distribution of the metastable atoms show that the slow atoms arise from dissociation out of $e\,^3\Sigma_u^+$ and $B'\,^1\Sigma_u^+$ states and from predissociation out of the $d\,^3\Pi_u^+$ and $D\,^1\Pi_u^+$ states. The fast atoms arise from a previously unreported $^1\Pi_u$ state that corresponds to a separated-atom limit in which both atoms are in $n = 2$ states.

The dissociative excitation of molecular hydrogen can proceed via the process $H_2 + e \to H(2s) + H + e$ in which one of the product hydrogen atoms is in the metastable $2^2S_{1/2}$ state. Leventhal, Robiscoe, and Lea[1] used a time-of-flight (TOF) method to measure the energy distribution of the $H(2s)$ atoms and found that the dissociative excitation yielded metastable atoms in two distinct kinetic-energy ranges: ~0.3 eV ("slow") and ~4.7 eV ("fast"). Subsequent TOF experiments on the dissociation of H_2 have been reported by Clampitt and Newton[2] and also by Czarnik and Fairchild.[3] In the present Letter we report results obtained from a TOF experiment[4] that differs from the earlier work mainly in that the detector can view the angular distribution of the $H(2s)$ atoms with a resolution of 1 degree over a range of 60 to 120 degrees with respect to the electron beam. From measurements of the excitation function and the angular distribution of both the slow and fast metastable atoms we deduce the symmetry, multiplicity, and asymptotic energy of the final states of the dissociation process; our conclusions here differ in several significant aspects from those reported by the abovementioned workers. For example, we find that the fast $H(2s)$ atoms arise from a previously unreported $^1\Pi_u$ state that corresponds to a separated-atom limit in which both H atoms are in $n = 2$ levels.

Figure 1(a) shows a schematic of the dissociative excitation process. An electron collides with the H_2 molecule and excites it "vertically" in the Franck-Condon region. If the transition is to a point above the asymptotic limit of the excited state, (or if, as in predissociation, a normally bound final state mixes with another state that has a lower asymptotic energy) then the molecule dissociates. The excess of the excitation energy over the asymptotic energy of the final state is shared, equally, as kinetic energy by the outgoing hydrogen atoms. The