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## Precise Test of the $z^2$ Dependence of X-Ray Emission Induced by $\alpha$ Particles and Deuterons\*

C. W. Lewis, J. B. Natowitz, and R. L. Watson

*Cyclotron Institute, Texas A & M University, College Station, Texas 77843*

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The ratios of x-ray yields from the irradiation of Ti, Cu, and Au targets with  $\alpha$  particles and deuterons of identical velocities have been determined between 6.25 and 20 MeV/amu. At the lowest velocities the data for  $K$  x rays from Ti and Cu indicate deviations as large as 12% from the theoretical ratios calculated according to the Born approximation. No significant deviation is observed for  $L$  x rays from Au.

Data have recently become available concerning  $K$ -electron-shell ionization by proton and  $\alpha$  bombardments between 2 and 28 MeV/amu<sup>1,2</sup> where the ionization cross section is deduced by measuring the number of  $K$  x rays emitted. At such velocities these projectiles remain totally stripped of electrons during their passage through matter, and also the conditions for a plane-wave Born-approximation treatment of the process should be quite well met. The resulting theory, as presented by Merzbacher and Lewis,<sup>3</sup> contains in it the simple prediction that the ionization cross sections for two different projectiles,  $a$  and  $b$ , incident at the same velocity on the same target are in the ratio  $(z_a/z_b)^2$  where  $z$  is the atomic number of the projectile. The data of Refs. 1 and 2 do seem to be in agreement with this prediction within the accuracy of the measurements, which is about 10%. This is in contrast to the situation with heavy-ion projectiles and lower velocities where the data<sup>4-6</sup> show very large deviations from a  $z^2$  dependence.

This paper presents results of measurements designed to check the  $z^2$  dependence at high velocities to an accuracy of less than 2%. The experiment compared the numbers of x rays produced by  $^2\text{H}^+$  and  $^4\text{He}^{++}$  beams selected by magnetic analysis so that the velocities incident on the target differed by only 0.640%, which is the

difference in charge/mass for the two projectiles. Beams having energies of 6.25, 7.0, 7.5, 12.5, and 20.0 MeV/amu were furnished by the Texas A & M variable-energy cyclotron. To switch from one beam to the other it was necessary only to change the cyclotron radio frequency by 0.64%, leaving all magnetic fields (cyclotron, analyzing magnet, focusing quadrupoles) unchanged. The two cyclotron resonances are well resolved; in particular it was experimentally determined that the contamination of one beam by the other was less than  $10^{-4}$ . Previous experience with the Texas A & M cyclotron has shown that  $\text{H}_2^+$  is a frequent contaminant when beams of particles with similar charge/mass are being used. Since the  $\text{H}_2^+$  and  $^2\text{H}^+$  resonances could not be resolved, a 200- $\mu\text{g}/\text{cm}^2$  aluminum foil was placed over the analyzing-magnet entrance slit to remove (by breakup) any  $\text{H}_2^+$  ions in the beam.

The number of beam particles incident on the target was determined by direct counting with a plastic scintillator which was placed behind the target in the path of the beam. The x rays were detected in a Si(Li) spectrometer (1- $\text{cm}^2$  area) placed at  $90^\circ$  with respect to the incident-beam direction at distances of 3.4 to 5.2 cm from the target. Self-supporting metallic foils of Ti, Cu, and Au were positioned at  $45^\circ$  to the incident beam, giving effective thicknesses of 4.36, 6.35,

and 14.7 mg/cm<sup>2</sup>, respectively. To reject events associated with background and with beam particles that were energy degraded due to slit-edge scattering, a narrow-energy "window" was placed on the pulses from the scintillator with a fast differential discriminator. Only those pulses satisfying the energy requirement were counted, and the x-ray detector pulse-height-analysis system was free to accept pulses only in coincidence with the selected scintillator pulses.

The results of the measurements of *K* x rays from Ti and Cu and *L* x rays from Au are given in Fig. 1. The average values of the velocities of the two beams in each target are slightly different due to the initial 0.64% difference and the differing rates of velocity loss in the target. The ratios have been corrected for this effect by using experimental excitation functions from Refs. 1 and 2. The correction was largest for the 12.5-MeV/amu bombardment of Au, resulting in a 1.0% decrease in the ratio. Corrections for multiple scattering and dead-time losses were considered and found to be completely negligible. The errors associated with the measured ratios represent standard deviations. They take into account counting statistics (1%) and the reproducibility of measurements. Under the assumption

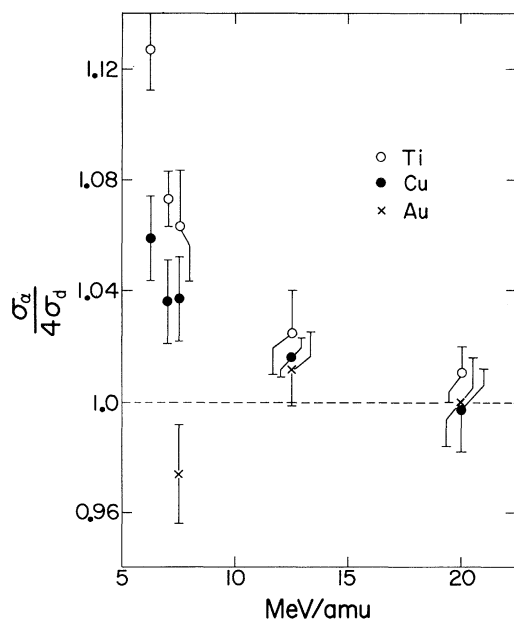


FIG. 1. Reduced cross-section ratio  $\sigma_\alpha/4\sigma_d$  as a function of projectile velocity for *K* x-ray emission from Ti and Cu and *L* x-ray emission from Au. The indicated errors are standard deviations. The dashed line shows the prediction of the Born-approximation theory.

that the reduced ratio of x-ray yields is the same as the reduced ratio of ionization cross sections,  $\sigma_\alpha/4\sigma_d$ , Fig. 1 shows that the experimental ratio for *L* x rays from Au does not differ significantly from the theoretical value of 1.0 over the velocity range of our measurements. In contrast, the ratios for *K* x-ray production from Ti and Cu show increasing deviation from theory with decreasing velocity. At 6.25 MeV/amu these deviations are 12% for Ti and 6% for Cu. For Ti, at this velocity, an independent experiment was performed to investigate the x-ray contribution from secondary effects (stopping electrons and bremsstrahlung). The results showed that less than 0.3% out of the total deviation of 12% could be attributed to these effects.

Several considerations can be discussed regarding the observed deviation. One can note first that if the x-ray angular distributions were different for  $\alpha$ - and deuteron-induced events, then a deviation from the theoretical ratio could exist at any given angle of x-ray emission, even if there were no deviation when averaged over all angles. It has long been assumed however that the emission is isotropic. Even though the only experimental evidence for this is a measurement<sup>7</sup> of *L* x rays from the bombardment of Au, we can find no reason to question this assumption, at least in the case of *K* x rays. For, to the extent that the emission of *K* x rays involves only the inner electrons, a vacancy in the *K* shell means that the *K* x ray is radiated from a system whose total angular momentum is  $\hbar/2$ , and in the case of electric-dipole emission, this leads to an isotropic angular distribution independent of substate populations.<sup>8</sup>

A second consideration relates to how well one might expect a plane-wave Born-approximation theory to work under the conditions of the experiment. This in turn can be broken into two parts: the validity of representing the projectile's motion by plane waves, and the justification of neglecting the polarization of the initial *K*-electron wave function by the incoming projectile. Concerning the use of plane waves (rather than free-state Coulomb wave functions), a measure of the accuracy of this approximation is provided by a calculation of the projectile's deflection angle when its impact parameter corresponds to a high probability for ionization to occur. For the velocities used this occurs for distances of the order of the Bohr radius for the electron,<sup>9</sup> which leads to classical deflection angles of only 0.06° and 0.1° for 6.25-MeV/amu projectiles incident

on Ti and Cu, respectively. Thus the use of plane waves would seem to be very well justified.

The question of possible distortion of the initial  $K$ -electron wave function by the incoming projectile is rather more difficult to answer. To begin with, we note that such an effect would be qualitatively consistent with the experimental results. For, the more tightly bound the electron, the more resistant it should be to such distortion. The  $K$  electron in Cu is more tightly bound than in Ti, and the deviation from the  $z^2$  dependence is observed to be less for Cu. The only quantitative estimate of distortion which is available at this time is that due to Brandt, Laubert, and Sellin.<sup>4</sup> Their approach considers the distortion effect from the point of view that the positive projectile provides additional binding of the  $K$  electron, and the effect of this on the ionization cross section is then calculated. If the binding correction function  $\epsilon$  from Ref. 4 is calculated for the 6.25-MeV/amu bombardments of Ti and Cu, and used to scale the ordinate and abscissa of the universal curve in Ref. 6, then one finds qualitative disagreement with the experimental results. That is, the binding correction predicts a *reduction* in  $\sigma_e/4\sigma_d$ , rather than the experimentally observed increase. It should be mentioned however that the expression for the function  $\epsilon$  was found by examining the form of the ionization cross section at very low bombarding energies, so that one should doubt the validity of applying it to the high energies of the present experiment. Brandt, Laubert, and Sellin<sup>4</sup> also introduce a correction factor to account for the Coulomb deflection of the projectile. Under the conditions of our experiment we find that this factor has a negligible effect on the ionization cross sections, in agreement with the conclusion of the previous paragraph.

An intriguing alternative interpretation of the experimental results is one that assumes the  $K$ -ionization process to be well described by the Merzbacher theory, but that the fluorescence yields are different for  $\alpha$ - and deuteron-produced  $K$ -shell vacancies. It has customarily been assumed that fluorescence yields are independent of the particular excitation process that creates the initial vacancy. However, the experiments of Richard *et al.*<sup>10</sup> in which one observes shifts in the energies of  $K\alpha$  and  $K\beta$  x rays, and changes in the relative  $K\alpha/K\beta$  yield, when the type of projectile is changed, clearly show that features of the x-ray emission processes are being influenced by the initial mode of excitation.

Before one could claim that a difference in fluorescence yields is demonstrated by the present experiment, a much more careful assessment of the electron wave-function distortion effect would have to be made.

Finally we would emphasize two points. Firstly, assuming the experimental x-ray yields directly reflect the ionization cross sections, the results show an unexpectedly large deviation from the  $z^2$ -dependence prediction of the Born-approximation theory. Secondly, this type of experiment is free from a variety of theoretical and experimental problems which are present in comparing absolute cross-section measurements with theory. Experimentally, one does not have to determine target uniformity or detector efficiency, making possible more accurate measurements. The theoretical simplification is that uncertainties related to the normalization of the emitted  $K$ -electron continuum wave function<sup>11</sup> and the approximation of the initial bound-electron wave function by a nonrelativistic hydrogenic wave function are largely eliminated since a ratio of cross sections is measured. It might also be noted that the particular choice of deuterons and  $\alpha$  particles as projectiles is a desirable one from the standpoint that the parameter relationships in the classical Coulomb scattering of projectiles with the same charge/mass are identical, except for the projectile-target reduced mass effect. This has the result of further decreasing the importance of precisely what is taken for the projectile wave functions. It seems apparent that an extension of this type of measurement in both bombarding energy range and choice of target can lead to a substantial increase in understanding of the ionization and x-ray emission processes.

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## Theory of Nearly Symmetric Excited-State-Excited-State Electron Capture in Ion-Atom Inelastic Scattering

Burke Ritchie\*

*Theoretical Studies Branch, Laboratory for Space Physics, National Aeronautics and Space Administration-Goddard Space Flight Center, Greenbelt, Maryland 20771*

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Formulas are derived, in the two-state approximation, for the cross sections for ion-atom atom excitation and excited-state ion capture for the case when the excited states are nearly degenerate. Under certain conditions the theory predicts oscillations in the total excitation and capture cross sections resulting from interference between the two degenerate final channels, in qualitative agreement with the measurements of van den Bos on H<sup>+</sup>-He excitation collisions and of Hughes *et al.* on capture into excited states of H.

Smith<sup>1</sup> first showed that the symmetric A<sup>+</sup>-A electron-capture cross section could be approximated<sup>2</sup> by

$$\sigma = \sigma_F - \pi^{3/2} b_0 \left| \frac{d^2 \Delta \eta(b)}{db^2} \right|_{b=b_0}^{-1/2} \cos[2\Delta \eta(b_0) - \pi/4], \quad (1a)$$

$$\Delta \eta(b) = \eta_g(b) - \eta_u(b), \quad (1b)$$

where  $\eta_g$  and  $\eta_u$  are the phase shifts for the elastic scattering in the ion-atom potentials *gerade* and *ungerade*, respectively, with respect to inversion through the molecular origin. Equation (1a) can be derived from the expression for the two-state symmetric capture cross section<sup>3</sup> when  $\Delta \eta$  passes through a maximum at a particular impact parameter, producing an oscillatory structure in the total cross section because the capture probability has remained nearly constant over a range of impact parameters.<sup>2</sup> Olson<sup>4</sup> then showed that the *inelastic* cross section for an asymmetric capture could also be analyzed in this form to predict oscillations, provided one first makes the Stueckelberg-Landau-Zener approximation,<sup>5</sup> which permits the expression of the cross section in terms of known analytic functions. Qualitatively<sup>4</sup> this point of view is related to an idea of Lichten,<sup>6</sup> who pointed out that the A<sup>+</sup>-B system approximately regains the g-u symmetry as the united-atom limit is approached. In this paper we point out that in the A<sup>+</sup>-B system the g-u symmetry is also approximately regained when A<sup>+</sup> excites B to a Rydberg state such that the excited electron sees effectively one unit of charge on B and (if A<sup>+</sup> is a proton) exactly one unit of charge on A<sup>+</sup>. Hence these excited states have approximate *gerade-ungerade* symmetry with respect to charge; asymmetry with respect to mass is not relevant in the Born-Oppenheimer separation of nuclear and electronic motion, which is usually invoked in moderately low-energy atomic collisions. This situation occurs in the excitation of He by protons to  $n \geq 3$  states, accompanied by capture into the  $n \geq 3$  states of the hydrogen atom [effectively, it also occurs for He excitations as low as  $n=2$ , since the energy defect between excitation to the He(2<sup>1</sup>S) state and capture into the H(2s) state is only about 0.01 Ry<sup>7</sup>], and is apparently responsible for an oscillatory structure which appears prominently in the total 1<sup>1</sup>S-3<sup>1</sup>P and 1<sup>1</sup>S-3<sup>1</sup>D direct excitation cross sections measured by van den Bos,<sup>8</sup> less prominently in the capture cross sections into H(3s) and H(4s),<sup>9</sup> and more prominently in the captures by protons on heavier noble-gas atoms.<sup>9</sup> These collisions occur above 1 keV, which is the limit at which the Born-Oppenheimer approximation begins to lose validity; however, center-of-mass effects on the motion of the excited electron will probably be small enough