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Projectile z Dependence of K X-Ray Emission Induced by Alpha Particles and Deutrons*

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Measurements of the yields of K x rays from Cl, Ca, Ti, Ni, and Cu targets and L x rays from Sn and Au targets produced by bombardment with α particles and deuterons of identical velocities have been performed at energies from 2.9 to 20 MeV/amu. The ratios of K x-ray yields for the Cl, Ca, Ti, Ni, and Cu targets all show substantial deviations from the ratios expected on the basis of the theoretically predicted z^2 dependence of the Born-approximation and binary-encounter descriptions. The deviations exhibit a "crossover" behavior in which the measured ratios are smaller than expected at low projectile velocities, rise above the expected value at higher velocities, and finally decrease toward the expected value at the highest velocities. The relationship between these deviations and similar deviations in stopping-power measurements has been investigated. Angular distributions of Ti K x rays and Sn K and L x rays were also measured and found to be isotropic to an accuracy of about 2%.

I. INTRODUCTION

Much interest has been stimulated lately in atomic inner-shell ionization produced by the passage of high-energy heavy charged particles through matter. The characteristics of K X-ray emission resulting from this process have been studied by Bissinger *et al.*[1] using 2–28-MeV protons and by Watson *et al.*[2] using 30–80-MeV α particles. The data obtained in these two experiments are satisfactorily accounted for by assuming the mechanism for the creation of inner-shell vacancies to be the ejection of electrons by direct scattering in the Coulomb field of the approaching projectile. Both the Born-approximation description presented by Merzbacher and Lewis[3] and the classical binary-encounter approach given by Garcia[4] reproduce the experimental x-ray production cross sections to within about 25% on the average. The experimental cross sections, however, are systematically found to fall below the theoretical excitation functions at low velocities, cross them at intermediate velocities, and rise above them at high velocities.

Both the Born-approximation theory and the binary-encounter theory contain the simple prediction that the ionization cross sections for two different projectiles incident at the same velocity on the same target are in the ratio of the square of the projectile atomic numbers. The projectiles in the above-mentioned experiments remain fully stripped of electrons during their passage through matter, and also the conditions for a plane-wave Born-approximation treatment of the process should be met quite well. Although the data of these experiments are in qualitative agreement with this prediction of a z^2 dependence, it is desirable to test the z dependence in this energy range more precisely than is allowed by the accuracy of the absolute cross sections given in Refs. 1 and 2, which is about 10%.

The main incentive for a detailed examination of the projectile z dependence at energies greater than approximately 2 MeV/amu stems from the fact that very large deviations from a z^2 dependence have been observed at lower velocities[5,6] and for heavy ions[7] where the conditions of fully stripped projectiles and applicability of the plane-wave Born approximation are not well met.

Measurements designed to check the z dependence at high velocities to an accuracy of less than 2% have recently been carried out.[8] In these experiments comparisons were made of the yields of x rays produced by beams of $^2H^+$ and $^4He^{++}$ selected by magnetic analysis so that the velocities incident on the target differed by only 0.640%, which is the difference in charge-to-mass ratio for the two projectiles. The results of this study showed the occurrence of significant deviations from a z^2 dependence for K x rays from Ti and Cu in the vicinity of 6.25 MeV/amu. In the present paper, we discuss the details of these experiments and report a considerable extension of the previous work both in projectile energy and in target atomic number.

II. EXPERIMENTAL METHODS

A. "Absolute" Measurements

Details of the beam transport system and target chamber used in the first type of measurements are shown in Fig. 1. We refer to these as "absolute" measurements since provision was made for directly counting the number of beam particles incident on the target. This task was accomplished with a plastic scintillator-photomultiplier system located downstream from the target in the direct path of the beam. The scintillators were 2.2-cm-diam cylinders of NE102, with thicknesses ranging from 0.4

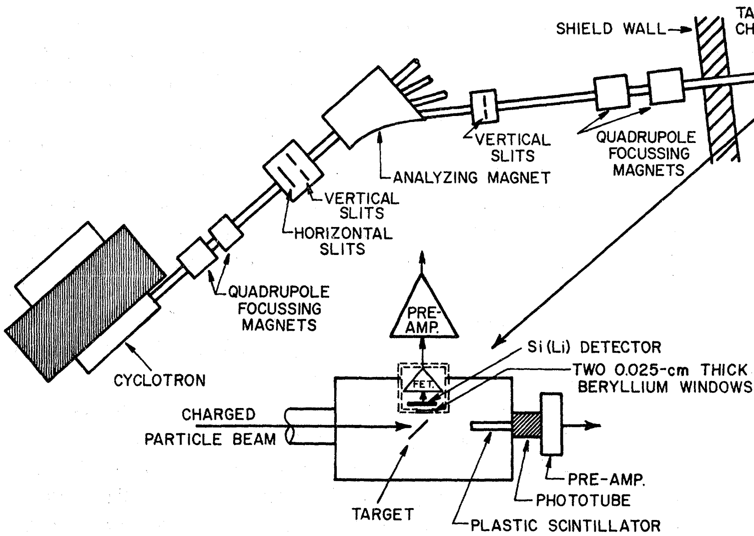

FIG. 1. Diagram of the beam transport system and target-chamber configuration.

to 1.3 cm and were located at distances varying from 9 to 15 cm with respect to the target. The photomultiplier used was an RCA 8575.

α-particle and deuteron beams having energies of 2.9, 4.5, 6.25, 7.0, 7.5, 12.5, and 20 MeV/amu were furnished by the Texas A&M variable-energy cyclotron. The targets, which were inclined at 45° with respect to the beam direction, were self-supporting metallic foils of Cu (6.35 mg/cm^2), Ni (3.60 mg/cm^2), Ti (4.36 mg/cm^2), and Au (14.7 mg/cm^2). (All target and target backing thicknesses referred to in this paper are effective thicknesses, which take into account the target inclination angle.)

X rays produced at the target by the α-particle or deuteron beams were detected in a Si(Li) spectrometer located at 90° with respect to the beam direction. During the course of these measurements, several spectrometers were used, all having sensitive areas of about 1 cm^2, but with target-spectrometer distances varying between 3.0 and 5.2 cm.

The electronic system used is shown in Fig. 2. Pulses from the scintillator, in order to be acceptable, had to fall in a narrow pulse-height range imposed by a fast differential discriminator. Pulses from the x-ray spectrometer were passed through the linear gate into a multichannel analyzer only when they were in time coincidence with an acceptable pulse from the scintillator. The selected scintillator pulses were also separately scaled to give the number of beam particles incident on the target. Significant features of the system are that beam contamination (discussed below) is prevented and x-ray events produced by beam particles that have been energy-degraded

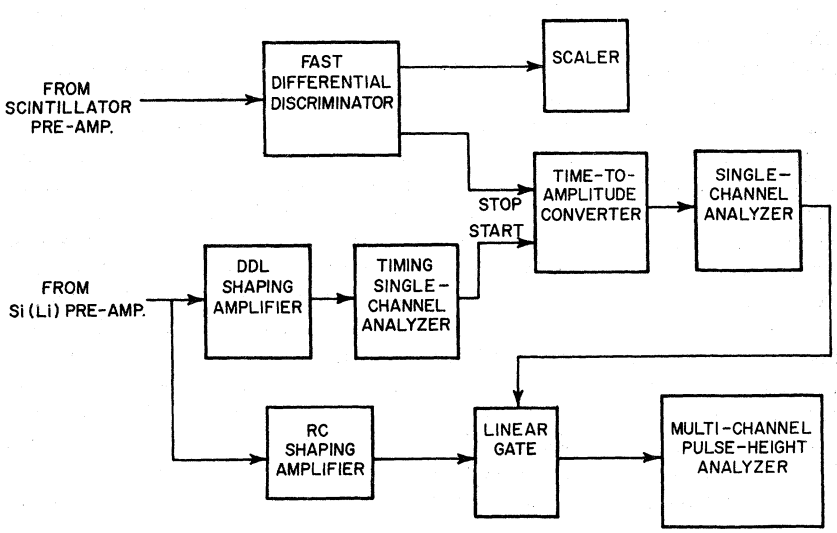

FIG. 2. Schematic diagram of the electronic system used for the "absolute" measurements.

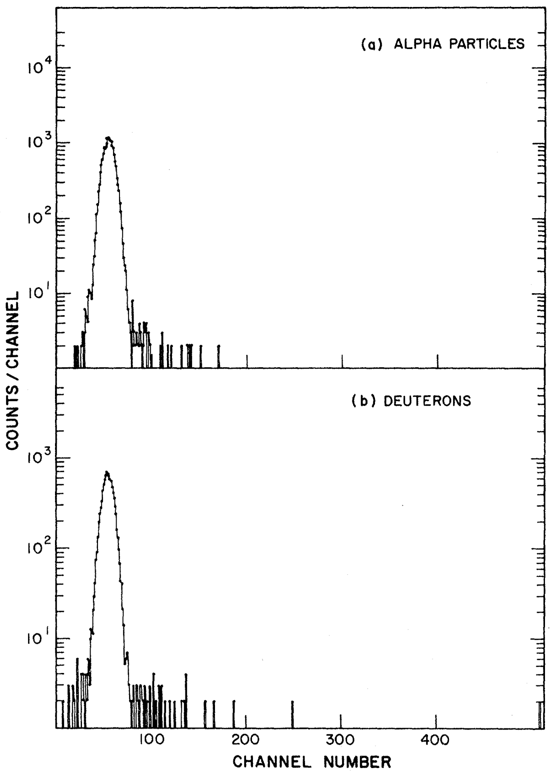

FIG. 3. Sample Ti K x-ray spectra obtained in the "absolute" measurements with (a) 30-MeV α particles and (b) 15-MeV deuterons.

due to slit edge scattering are eliminated, in addition to random background events. Sample K x-ray spectra resulting from the α particle and deuteron bombardments are shown in Fig. 3. The backgrounds were so small that there was little difficulty in extracting the number of counts in the $K_\alpha + K_\beta$ peak to an accuracy of at least 1%.

As indicated in the Introduction, the goal of the experiment was to measure the ratio of x rays produced by two nuclear projectiles having identical velocities but different nuclear charges. The choice of α particles and deuterons was based on the simplicity which it introduces into the measurement. Since the charge-to-mass ratio of these two nuclei differs by only 0.64%, their cyclotron resonance frequencies differ by the same amount if the magnetic field of the cyclotron remains unchanged. Furthermore, since the energy-analyzing and beam-focusing elements external to the cyclotron are magnetic devices, it is particularly simple to switch from one beam to another. Once the cyclotron and beam transport system are adjusted to produce a focused beam of one type at the target, all that is required is to change the ion source gas and shift the cyclotron rf by 0.64%. The two beam spots (as observed on a ZnS viewer) were indistinguishable from one another.

While the cyclotron resonances for α particles and deuterons are well resolved, their separation is not so large as to rule out *a priori* the possibility of a small contamination of one beam by the other. However, since the scintillator pulse heights produced by α particles and deuterons of the same velocity are not identical, the pulse-height requirements imposed by the fast differential discriminator effectively eliminated any such source of error in the absolute measurements. In addition, searches carried out at 6.25, 7.0, and 12.5 MeV/amu with a solid-state particle spectrometer placed in the path of the direct beam showed any contamination of one beam by the other to be less than 1 in 10^4. This null result was of importance for the "relative" measurements described in Sec. IIB, since those measurements did not discriminate against beam contamination.

Previous experience with the Texas A&M cyclotron has shown that H_2^+ is a frequent contaminant when beams of similar charge/mass ratio are being accelerated. (The cyclotron resonance of H_2^+ cannot be resolved from that of deuterons.) To eliminate this problem all measurements described in this paper were performed with a 200-μg/cm^2 Al foil placed over the analyzing magnet entrance slit to remove (by breakup) any H_2^+ ions in the beam.

The result of an absolute measurement on a given target x was expressed in terms of the quantity

$$A(x) = N_\alpha(x)/[4N_d(x)], \qquad (1)$$

where N_α and N_d are the measured number of x rays per α particle and deuteron, respectively.

B. "Relative" Measurements

The results which were obtained with Cu, Ni, and Ti targets indicated the desirability of extending the measurements to targets of smaller atomic number. Because of the rapid decrease of the x-ray detection efficiency with decreasing x-ray energy (due to absorption in the two 0.025-cm-thick Be windows) and the resulting decrease in counting rate, the previous absolute measurement technique did not seem well suited to lighter elements. Instead, a different method was used which measured α-induced to deuteron-induced x-ray ratios for a given target *relative* to the corresponding ratio for Ni. For these measurements the target chamber was modified by removal of the plastic scintillator and extension of the beam line to a shielded beam stop 2.35 m distant from the target. X rays from the target were detected as before but with a higher resolution spectrometer (~350 eV) of active area 30 mm^2 operated in a simple singles mode. In addition to the main target, a monitor Ni foil (0.13

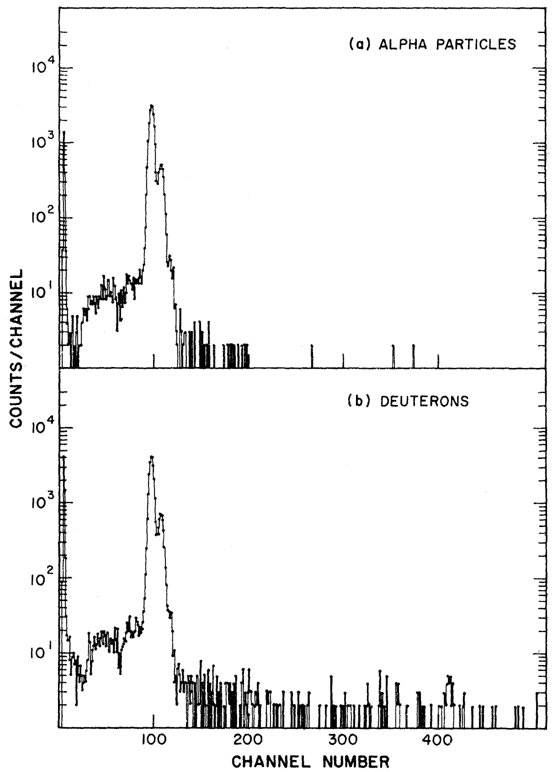

FIG. 4. Sample Ni K x-ray spectra obtained in the "relative" measurements with (a) 30-MeV α particles and (b) 15-MeV deuterons.

mg/cm^2) was placed in the path of the beam 46-cm upstream. The target-monitor foil separation was small enough so that acceptable beam spots could be obtained simultaneously at both locations. X rays produced at the monitor foil were detected and recorded by a second x-ray spectrometer and pulse-height analysis system, also operated in singles mode. For measurements obtained under these conditions, the x-ray counting rates were no longer limited by the plastic scintillator, so that the beam levels could be raised and the target-spectrometer distances could be correspondingly increased to about 12 times what they had been during the absolute measurements. This, of course had the beneficial effect of decreasing the sensitivity of the spectrometers' solid angles to small differences between the foci and the α-particle and deuteron beam spots.

The new elements which were bombarded in these relative measurements were Ca in the form of CaF_2 (4.7 mg/cm^2 on a 9.7-mg/cm^2 Al backing, and 0.38 mg/cm^2 on a 2.4-mg/cm^2 Al backing), Cl in the form of NaCl (0.22 mg/cm^2 on a 0.75-mg/cm^2 Mylar backing), and Sn (0.16 mg/cm^2 on a 0.75-mg/cm^2 Mylar backing). In addition, Ti (4.36 and 0.25 mg/cm^2) was also bombarded to provide a consistency check of the absolute Ti and Ni measurements. Sample x-ray spectra are shown in Fig. 4. One notices the higher backgrounds compared with those of Fig. 3, which were obtained in the coincidence experiments.

The value for relative measurements $R(x)$ on a given target x was calculated from the expression

$$R(x)=N_\alpha(x)\,N_d(\mathrm{Ni})/[N_d(x)\,N_\alpha(\mathrm{Ni})], \qquad (2)$$

where $N_j(x)$ and $N_j(\mathrm{Ni})$ refer to the number of x rays detected simultaneously by the main and monitor spectrometers, respectively, under bombardment by projectiles of type j. A relative measurement is, in principle, related to a corresponding absolute measurement by the equation

$$R(x)=A(x)/A(\mathrm{Ni}), \qquad (3)$$

where A is defined as in Eq. (1).

C. Angular-Distribution Measurements

To assist in the interpretation of the x-ray ratio measurements, an experiment was performed to measure the angular distribution of characteristic x rays induced by α-particle and deuteron bombardments. A cylindrical 28-cm-diam target chamber was employed and x rays originating at the target located in the center of the chamber emerged through a 0.6-cm-wide horizontal slot cut into one of the chamber's circular sides. To maintain the chamber's vacuum, a 1-mil Mylar foil covered the slot. The Mylar foil in its stretched condition was sufficiently uniform that differential attenuation effects on the x rays were negligible.

The x rays were detected with an x-ray spectrometer collimated to a sensitive area of 8 mm^2 and placed 16.5 cm from the target. The beam intensity was monitored by means of a second spectrometer whose position was left fixed at 90°. The angular distribution measurements used α-particle and deuteron beams having energies of 6.25 MeV/amu incident on targets of Ti (4.36 mg/cm^2) and Sn (3.89 mg/cm^2 on 2.4 mg/cm^2 Al).

III. DATA ANALYSIS

The number of x rays detected in each measurement was determined by integrating the area under the x-ray photopeak and subtracting an appropriate linear background. From these numbers, the preliminary absolute and relative ratios were calculated using Eqs. (1) and (2).

The projectiles striking the target are selected by magnetic analysis and hence they possess the same magnetic rigidity, but because of nuclear binding effects, the α-particle velocity is 0.64% greater than the corresponding deuteron velocity. What first appears to be a minor problem in the experimental technique can, in fact, be turned to advantage because α-particles lose velocity twice as fast as deuterons. Thus the larger initial α-particle velocity can lead to more nearly equal average velocities within the target than would otherwise be the case. In any event, the effect on the x-ray ratios of unequal average velocities can be corrected for by using either experimental excitation functions for α-particles[2] or protons,[1] or theoretical excitation functions provided by the plane-wave Born-approximation theory[3] and the binary-encounter model.[4] These functions are sufficiently accurate for this purpose since the resulting corrections to the measured α-particle-to-deuteron x-ray ratios are quite small. The largest correction was an increase of 2.6% for the 2.9 MeV/amu relative measurement on Ca, and typically the correction was less than 1%. All absolute and relative measurements were corrected for this effect.

Correction of the ratios for multiple scattering, which causes the total path length for a projectile to be longer than the simple geometrical thickness of the target, was unnecessary because this effect is nearly identical for particles of the same charge-to-mass ratio and velocity.

Dead-time loss corrections are of potential importance only for the absolute measurements in connection with the scintillator counting rate. For the rates which were used (typically 50 000/sec), dead-time losses were less than 1%, and their effect on the measured x-ray ratio was virtually eliminated by keeping the scintillator counting rates as nearly equal as possible for both halves of a ratio measurement.

The last type of correction to which attention was given concerns secondary effects arising from finite target thickness. In addition to x-rays resulting from vacancies due directly to the incident nuclear projectiles, vacancies can also be created by stopping electrons (δ rays) and bremsstrahlung which, in turn, are produced by the projectiles. In the limit of infinitely thin targets this secondary contribution becomes negligible in comparison with the direct contribution whose measurement was the objective of this study. The targets used were not particularly thin, however, and so consideration of this effect was warranted.

Even though secondary processes may contribute appreciably to the total x-ray production cross section, their effect on the α-to-deuteron x-ray ratio is expected to be much smaller. This is due to the fact that the stopping electron and bremsstrahlung cross sections have a theoretical dependence on the charge and velocity of the projectile which is the same as that of the primary cross section.

The effect of secondary x-ray production on the measured x-ray ratios may be examined by redefining the measured x-ray ratio [Eq. (1)] in terms of x-ray emission cross sections:

$$A=\frac{\sigma_\alpha{}^T}{4\sigma_d{}^T}=\frac{\sigma_\alpha+\delta_\alpha}{4(\sigma_d+\delta_d)}\ , \qquad (4)$$

where σ^T, σ, and δ are, respectively, total, primary, and secondary cross sections for x-ray production by α particles and deuterons. The primary x-ray ratio is given by

$$T=\sigma_\alpha/4\sigma_d\ . \qquad (5)$$

If it is assumed that $\delta_\alpha=4\delta_d$, then the fractional difference between the primary ratio and the measured ratio may be expressed as

$$\frac{T-A}{A}=\frac{(\delta_\alpha/\sigma_\alpha)(A-1)}{1-(\delta_\alpha/\sigma_\alpha)(A-1)}\ . \qquad (6)$$

The production of K x rays by secondary processes has previously been considered by Jarvis *et al.*[9] in connection with measurements of x-ray production by 160-MeV protons. These authors point out that above a certain target thickness the effective cross section for x-rays produced by secondary processes shows a saturation effect in the sense that it remains constant with further increase in target thickness. Thus, in the regime referred to by Jarvis as "infinite thickness," the ratio of the secondary to primary x-ray production cross sections is also independent of thickness. Most of the targets used in the present study qualify as "infinitely thick" so that the formula for this case can be applied. Table I shows a sample of total and secondary K x-ray cross sections for α particles incident on two of the targets used in these experiments. The values for the total cross sections were obtained from a "universal" curve of the type suggested by Garcia,[10] but constructed from the experimental data of Ref. 2 rather than from the theoretically calculated values given in Ref. 4 and the values for the secondary cross sections were calculated using the formulation of Jarvis *et al.*[9]

The largest calculated effect of secondary processes in the present work occurs where the mea-

TABLE I. Total and secondary K x-ray cross sections for α particles.

Element	Energy (MeV)	$\delta\alpha$ (b)	$\sigma\alpha^T$ (b)	$\delta\alpha/\sigma\alpha^T$
Cl	12	109	3.56×10^3	3.16×10^{-3}
	25	293	4.89×10^3	6.37×10^{-2}
	30	322	4.78×10^3	7.22×10^{-2}
	40	350	4.04×10^3	9.48×10^{-2}
Ti	12	4	1.41×10^3	2.84×10^{-3}
	25	92	2.98×10^3	3.19×10^{-3}
	30	118	3.29×10^3	3.72×10^{-2}
	40	155	3.56×10^3	4.55×10^{-2}

sured ratios are the largest, i.e., for Cl in the vicinity of 5 MeV/amu ($E_\alpha = 20$ MeV). From Eq. (6), the fractional difference between the primary and measured ratios for this case was found to be 1.4×10^{-2}, which is less than the experimental error. Thus, unless the actual dependence of the secondary cross section is grossly different from theory, secondary x rays have little effect on the measured ratio.

The following experiment was performed to check this conclusion: Two 2.54-cm-diam Ti foils (3.57 mg/cm^2) were positioned at 60° with respect to each other and aligned to intercept the charged particle beam as shown in Fig. 5. A 6.4-mm-diam hole in the center of the second Ti foil allowed the beam to pass through without interacting. Directly behind this foil was located a 104-μg/cm Ni foil to act as a monitor of the beam intensity. With a 3.2-mm-thick Al plate positioned so as to shield the x-ray detector from x rays emitted from the first Ti foil, the spectrum of x rays emitted during bombardment with 6.25-MeV/amu α particles was recorded. This measurement gave the number of Ti K x rays generated in the second foil by stopping electrons and bremsstrahlung produced in the first foil, relative to the number of Ni K x rays produced in the monitor foil by α particles. The Al shield was then removed and the number of Ti K x rays produced in the first foil by the α-particle beam was measured relative to the number of Ni K x rays produced in the monitor foil. Finally, the first Ti foil was removed and the number of Ti K x rays produced in the second Ti foil was measured relative to the number of Ni K x rays produced in the monitor foil. This information was used to correct the preceding measurements for Ti K x-ray production in the second Ti foil by α particles hitting near the edge of the hole. This entire set of measurements was then repeated using a beam of 6.25-MeV/amu deuterons. From the results of this experiment, it was estimated that the fractional difference between the primary and measured x-ray ratios for this case is less than 3×10^{-3}. The value calculated from Eq. (6) using the cross sections in Table I is 2.3×10^{-3}.

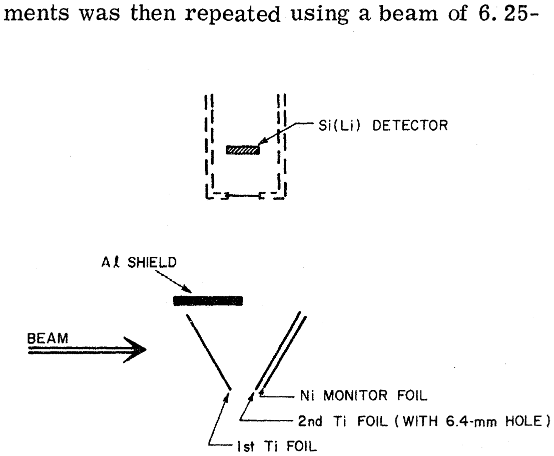

FIG. 5. Schematic diagram of the experimental arrangement used for the measurement of x-ray production by secondary processes.

In summary, we feel that all effects which could have an important influence on the measured values of the x-ray ratios have been considered, and that the deviations we have observed are not due to instrumental effects or deficiencies in experimental technique.

IV. RESULTS

A. X-Ray Yield Ratios

The principal results of this investigation, namely, the absolute measurements $A(x) = N_\alpha(x)/[4N_d(x)]$, and the relative measurements $A(x) = R(x)A(\mathrm{Ni})$ for each target x, are given in Table II. The errors listed for the absolute measurements represent standard deviations and include only counting statistics. The main contribution to the standard deviation errors given for the relative measurements were from uncertainties in background subtraction, since statistical errors were generally negligible. The entries in Table II include a smaller set of data published previously.[8]

In order to obtain the best values at each energy for the Ti and Ni x-ray ratios, the absolute and relative measurements were combined by the following least-squares technique. Let $A_i(\mathrm{Ti})$, $A_j(\mathrm{Ni})$, and $R_k(\mathrm{Ti})$ be representative values of absolute Ti, absolute Ni, and relative Ti measurements, respectively, all obtained at the same bombarding energy. Let $\overline{A}(\mathrm{Ti})$ and $\overline{A}(\mathrm{Ni})$ be the best values for the ratios for both targets. By the theory of least squares, the best values should minimize the expression

$$\sum_i w_i[A_i(\mathrm{Ti}) - \overline{A}(\mathrm{Ti})]^2 + \sum_j w_j[A_j(\mathrm{Ni}) - \overline{A}(\mathrm{Ni})]^2 + \sum_k w_k[R_k(\mathrm{Ti})\overline{A}(\mathrm{Ni}) - \overline{A}(\mathrm{Ti})]^2 .$$

The weights w are given by the inverse squares of the errors of the measurements. Applying the usual minimization procedure to this expression yields simple equations for each $\overline{A}$ in terms of the absolute and relative measurements. This procedure was used at each bombarding energy to obtain both $\overline{A}$ values as a function of energy. Since the elements Cu and Ni have atomic numbers which differ by only one unit, and since no significant difference could be seen in a comparison of the measurements for the two targets, it was decided to combine the Cu and Ni data for the least-squares fits.

The results of the fits are shown in Figs. 6 and 7, where smooth curves have been drawn through

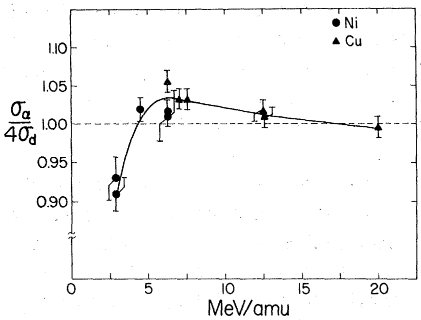

FIG. 6. "Absolute" x-ray ratios $A(x)$ for Ni and Cu as a function of projectile energy. The smooth curve has been drawn through the least-squares value at each of the energies measured.

the set of $\overline{A}$ values for Cu + Ni, and the set for Ti. The data from Table II have also been plotted on these figures for comparison. The relative Ti data have, of course, been transformed to equivalent absolute values by making use of the smooth curve for Ni in Fig. 6. We estimate that any point on the curves in Fig. 6 and 7 is accurate to 1.5%.

Corresponding results for Ca and Cl are shown in Fig. 8. The data are all relative measurements, so that the equivalent absolute values plotted in the figure are based on the Ni curve of Fig. 6. The few measurements of L x-ray ratios for Au and Sn targets listed in Table II are consistent with the z^2 prediction within experimental error.

B. Angular Distributions

The angular distributions of K x rays produced by 6.25-MeV/amu α particles and deuterons incident on Ti are shown in Fig. 9. The results have been corrected for x-ray absorption in the target by the method described in Ref. 2 using mass absorption coefficients from Ref. 11. The x-ray

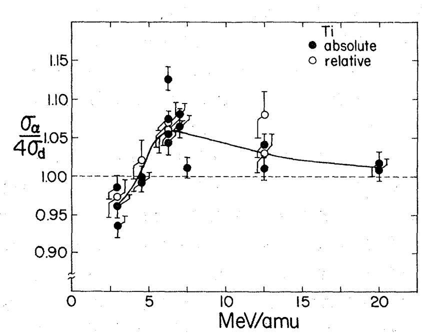

FIG. 7 "Absolute" x-ray ratios $A(x)$ for Ti as a function of projectile energy. The smooth curve has been drawn through the least-squares value at each of the energies measured.

TABLE II. X-ray yield ratios $A(x)$ determined from the "absolute" and "relative" measurements. All values have been corrected for the velocity difference discussed in Sec. III.

Energy (MeV/amu)	Absolute measurements Ti	Ni	Cu	Au(L)	Relative measurements Cl	Ca	Ti	Sn(L)
2.9	0.985 ± 0.016	0.909 ± 0.021			1.003 ± 0.04	1.031 ± 0.04	0.977 ± 0.04	
	0.961 ± 0.016	0.929 ± 0.028						
	0.934 ± 0.014							
4.5	0.993 ± 0.012	1.019 ± 0.015			1.277 ± 0.04	1.120 ± 0.04	1.021 ± 0.04	
	0.996 ± 0.017							
6.25	1.126 ± 0.015	1.011 ± 0.033	1.056 ± 0.014		1.264 ± 0.04	1.112 ± 0.04	1.068 ± 0.04	
	1.072 ± 0.012	1.014 ± 0.017					1.061 ± 0.04	
	1.043 ± 0.015							
	1.056 ± 0.016							
7.0	1.081 ± 0.013		1.033 ± 0.013					
	1.064 ± 0.014							
7.5	1.011 ± 0.013		1.032 ± 0.014	0.973 ± 0.016	0.995 ± 0.04	1.042 ± 0.04		1.047 ± 0.04
10.0					1.047 ± 0.04	1.048 ± 0.04		1.033 ± 0.04
12.5	1.040 ± 0.015		1.017 ± 0.014	1.022 ± 0.016	0.984 ± 0.04	1.004 ± 0.04	1.084 ± 0.04	
	1.009 ± 0.014		1.017 ± 0.013	0.980 ± 0.016			1.033 ± 0.04	
			1.008 ± 0.013	1.024 ± 0.016				
20.0	1.008 ± 0.014		0.996 ± 0.014	0.997 ± 0.016				
	1.016 ± 0.016							

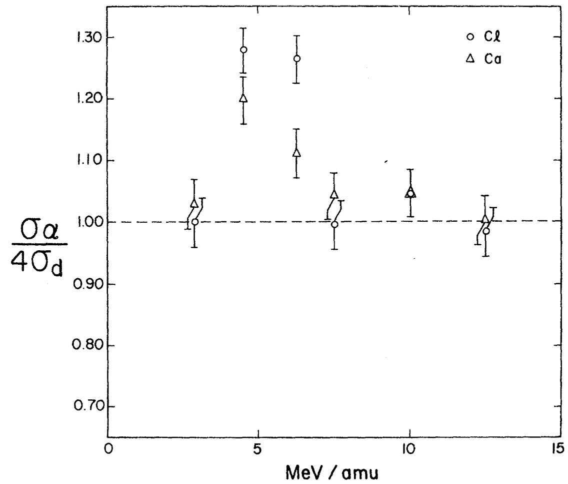

FIG. 8. "Absolute" x-ray ratios $A(x)$ for Cl and Ca vs projectile energy as derived from the "relative" ratios $R(x)$ and the least-squares curve for Ni.

yields have been arbitrarily normalized to unity at 90°. The mean deviation from isotropy is 1.1% for the α-particle measurements and 1.0% for the deuteron measurements.

In Fig. 10 is shown the angular distribution results for 6.25-MeV/amu α particles and deuterons incident on Sn. The x-ray spectrometer resolution made it possible to obtain separate angular distributions for $K\alpha$, $K\beta$, and L x rays. Again, each distribution has been normalized to unity at 90°, and target absorption corrections have been applied. The mean deviation from isotropy of the Sn $K\alpha$, $K\beta$, and L x rays was 2.1%, 3.6%, and 1.5% for the α-particle measurements and 1.7%, 2.5%, and 1.6% for the deuteron measurements, respectively.

It has been long been assumed that the charged particle induced emission of characteristic x rays is isotropic. A simple theoretical explanation can be constructed for the case of K emission on the basis of an angular momentum argument.[8] The only previous experimental evidence known to us for the correctness of this assumption is a 2% measurement of L x rays from Au induced by protons,[12] and 1.4% measurements of K x rays from Cu and In induced by 300- and 500-MeV electrons.[13] The present measurements provide additional experimental verification of the isotropy assumption. They also serve to remove an element of uncertainty regarding the interpretation of the ratio measurements. That is, the assumption of isotropy does seem to be confirmed so that a ratio measurement performed at 90° presumably gives the same value as the ratio of total x-ray yields.

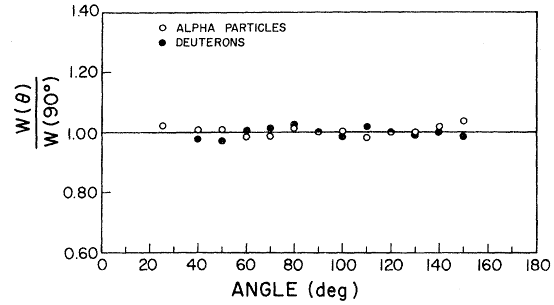

FIG. 9. Angular distribution of Ti K x rays produced by 6.25-MeV/amu α particles and deuterons.

V. DISCUSSION

A. Comparison With Previous Measurements

The general features of the data shown in Figs. 6–8 can be summarized as follows. The α-induced to deuteron-induced K x-ray ratios show substantial deviations from the theoretical z^2 prediction for all targets measured between Cl and Cu. All targets exhibit a "crossover" behavior in which the measured ratio is smaller than expected at low projectile velocities, rises above the expected value at higher velocities, and finally decreases toward the expected value at the highest velocities. The velocity for which the crossover occurs is larger for higher target atomic numbers. And finally, the magnitude of the maximum deviation above the crossover point decreases monotonically

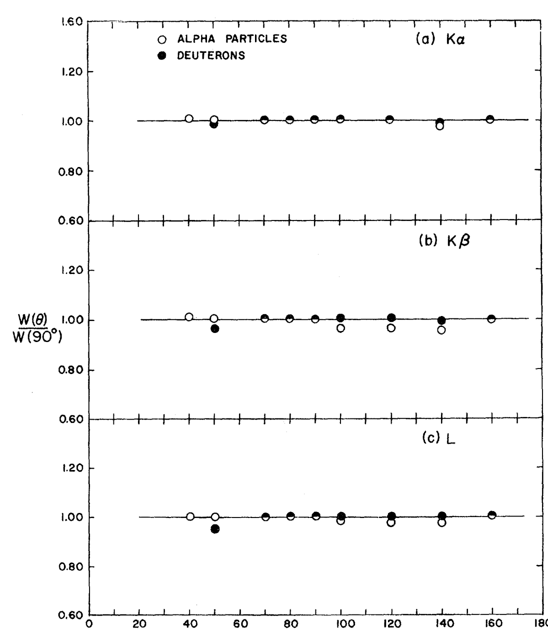

FIG. 10. Angular distribution of Sn (a) $K\alpha$ x rays, (b) $K\beta$ x rays, and (c) L x rays produced by 6.25-MeV/amu α particles and deuterons.

TABLE III. Experimental and theoretical values for the crossover point velocities.

Target	Experiment (MeV/amu)	Theory[a] (MeV/amu)
$_{13}$Al	0.5 ± 0.1[a]	0.51
$_{17}$Cl	2.9 ± 0.5[b]	0.99
$_{20}$Ca	2.7 ± 0.6[b]	1.40
$_{22}$Ti	4.4 ± 0.4[b]	1.75
$_{28}$Ni	4.4 ± 0.5[b]	3.00
$_{28}$Ni	3.7 ± 0.6[a]	3.00

[a] Reference 14. The errors given are based on the stated error of ± 2% for the ratio measurements.
[b] Present work.

with increasing target atomic number.

These measurements are closely related to measurements of the same type performed by Basbas *et al.*[14] on Al and Ni targets for a projectile velocity range which overlaps that of the present measurements. The latter data exhibit all the general features of our measurements and, in particular, the results for Ni, which was studied in both experiments, are in good agreement. Thus taken together the two experiments show a systematic behavior in the α-to-deuteron x-ray production ratio over a range of elements between Al and Cu.

The New York University group gives an outline of a theory[5,14] which attempts to make these results understandable. Expressed very briefly, two different effects are assumed to be dominant for velocities below and above the crossover point. In very slow collisions the reduction in cross section is attributed to an increase in the effective binding charge seen by the K electrons from Z to $(Z+z)$, where Z is the target atomic number and z is the projectile atomic number. In faster collisions, polarization of the K-shell orbit by the projectile is assumed, and an increase in the ratio is calculated.

In comparing the measurements with this theory, one can first note that the magnitude of the deviation above the crossover point is predicted to be inversely proportional to the target atomic number. The data show that the size of the effect does decrease between Cl and Cu but the case of Al is a striking exception to this trend; the maximum deviation observed for Al is about 10% compared with about 25% for Cl. It would clearly be helpful to have some experimental information for elements between these two.

A second comparison concerns the variation of the crossover point with target type. The simple prediction is that it should occur at a projectile velocity V_1 which satisfies the condition

$$V_1/V_2=I/(2Z_k^2R),$$

where V_2 is the Bohr velocity of the K electron, I is the binding energy of the electron, Z_k is the effective screened atomic number of the target, and R is the Rydberg constant 13.6 eV. Table III gives the comparison between experimental and predicted crossover points. It is apparent that significant differences from the prediction of this theory are observed.

B. Relationship to Stopping Power Measurements

Since electron excitation and ionization processes are the principal means by which charged particles lose energy in traversing matter, measurements of the yields of x rays resulting from these processes can provide information on the general theory of energy loss or stopping power. The most familiar result in this field is the Bethe stopping power formula

$$\frac{-dE}{dx}=\frac{4\pi e^4z^2N}{mV^2}\,B\,,$$

where ze is the charge of the projectile, V is its velocity, m is the electron mass, and N is the number of target atoms/cm^3. B is the stopping number and depends only on V and the target material. This result is based on the plane-wave Born approximation and contains within it a dependence on z and V which is analogous to the situation for x-ray production; the ratio of energy losses for two charged particles moving at the same velocity through the same medium is equal to the ratio of the squares of their nuclear charges. While the Bethe formula has been found to give a very good representation of an extensive body of experimental data, small discrepancies are known. For example, recent precise measurements by Andersen *et al.*[15] of the relative energy loss of hydrogen compared with helium projectiles in Al and Ta show the helium energy losses to be on the order of 1% greater than the factor of 4 predicted by the Bethe formula. Since this is reminiscent of the present x-ray yield deviations, it is of interest to seek a simple relationship between these two types of results.

This can be approached by expressing the stopping number B as a sum of terms B_i, where each B_i represents the contribution to B from the ith atomic shell. B_i is given by the equation

$$B_i=\int E\left(\frac{d\sigma_i}{dE}\right)dE\,,$$

where $d\sigma_i/dE$ is the (energy) differential cross section for transfer of an amount of energy E from the projectile to an electron in the ith shell, and the integral is over all possible energy transfers. In contrast, the quantity which is most closely related to the present x-ray measurements is the total ionization cross section for the ith shell,

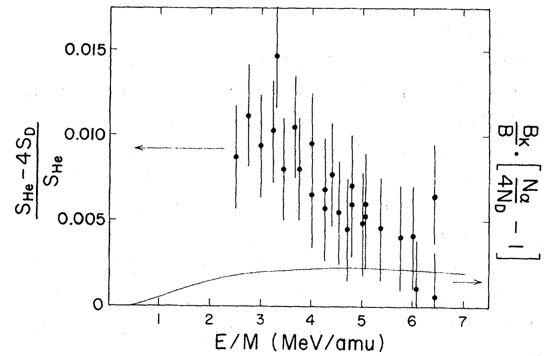

FIG. 11. Comparison of the energy-loss deviation from the work of Anderson *et al.* (Ref. 15) with the K x-ray yield deviation from the work of Basbas *et al.* (Ref. 14) for Al.

$$\sigma_i = \int \left(\frac{d\sigma_i}{dE}\right) dE .$$

From this comparison one sees that in order to establish a connection between the z^2 deviations observed in the x-ray and energy loss measurements one must know how the *differential* cross section deviates from the expected z^2 dependence as a function of energy transfer. Proceeding on the simple assumption that any such deviation is independent of energy transfer, it immediately follows that if a measurement of the K-shell total ionization cross section shows a z^2 deviation, then the same kind of deviation should appear in an energy-loss measurement, but reduced by approximately the factor B_k/B. In summary, if the x-ray yield deviations do directly reflect deviations in the corresponding ionization cross sections, then a direct comparison can be made between the energy-loss deviations of Andersen and the x-ray yield deviations observed in the present work and that of Basbas *et al.*[14] after applying the scaling factor B_k/B.

The most direct comparison can be made for Al, using the K x-ray measurements of Basbas *et al.* Figure 11 shows both Andersen's data and a smooth curve representing the x-ray yield deviations after multiplication by B_k/B. The values for B_k were those computed by Walske,[16] and the B values were obtained from the expression[15]

$$B = Z \ln(2mV^2/I),$$

whose accuracy is adequate for our purpose. Here Z is the target atomic number (13) and I is its average ionization potential (160 eV). In general, the two types of measurements do not agree, except at the highest energies. A possible explanation for the difference at lower energies might be associated with the existence of a z^2 deviation in the ionization cross section for the L shell. Even though there has been no experimental data to test this assumption, one can argue as follows: The K x-ray measurements on a variety of targets show z^2 deviations which persist up to projectile velocities which are comparable with the velocity of the bound K electron, and then diminish to zero at larger projectile velocities. If a similar condition governs the interaction with electrons in other atomic shells, then any such z^2 deviations would occur at smaller projectile energies than those for the K electrons.

A more limited comparison can also be made using the present x-ray measurements. For example, if we restrict our attention to measurements at 6.25 MeV/amu, for which the K x-ray deviations tend to be nearly maximum, then the products of the deviation times B_k/B give values in the range from 0.4% to 0.007% for Cl through Cu. In contrast, from Andersen's data on Al and Ta, one might estimate that the energy-loss deviations for elements between Cl and Cu would be quite similar, about 0.5%. These differences are qualitatively consistent with the explanation presented above in that deviations in the L-shell ionization cross sections would be expected to increase as the L electron velocity approaches the fixed projectile velocity i.e., as the target atomic number increases).

In conclusion, some degree of consistency between the energy-loss and K x-ray yield deviations can be perceived, but assumptions are required which go beyond what is experimentally known at this time.

C. Multiple-Ionization Considerations

If the fluorescence yield is independent of the excitation mode of an atom (in particular, whether by α-particle or deuteron bombardment), then the α-induced to deuteron-induced x-ray ratio is the same as the ionization cross-section ratio. Until now, we have not considered effects on the fluorescence yield resulting from possible differing initial states of excitation produced by the two projectiles. There is an increasing amount of evidence which demonstrates that the state of excitation is indeed dependent upon the characteristics of the projectile. For example, the K x-ray spectra resulting from nitrogen,[17] oxygen,[18] and fission fragment[19] bombardments of various targets show appreciable differences when compared with corresponding spectra induced by proton bombardments. These differences include x-ray energy shifts, $K\beta/K\alpha$ intensity ratio variations, and satellite structure. In many cases they can be quantitatively correlated with vacancies in higher shells in addition to the K shell, resulting from multiply ionizing collisions. Since fluorescence yields depend on the degree of ionization of the atom undergoing decay[20] one would not, in general, expect identical fluorescence yields for differing modes of production if multiple ionization is occurring.

McGuire and Mittleman[21] have performed an instructive calculation which investigates one possibility for a fluorescence yield variation resulting from a case of multiple ionization. As a trial model, they assume the total K x-ray emission rate includes contributions from both singly ionized (one K- shell vacancy) and doubly ionized (one K-shell and one L-shell vacancy) atomic states. They further assume that the Auger and radiative transition probabilities *per electron* are the same for both cases, so that the calculation concentrates solely on how average transition probabilities are affected by having one less electron available as a transition participant part of the time. Their result is that the average fluorescence yield F is related to the value F° corresponding to the case of only single K-shell ionization by

$$F = F^\circ [1 + F^\circ)\, \xi/6],$$

where ξ is the ratio of the double-ionization to the single-ionization total cross section. McGuire and Mittleman have estimated ξ for the bombarding particles, energies, and targets used in the present experiment by means of the classical Gryzinski method. Their approximate result is that

$$\xi = 60 z^2/Z^2,$$

where z and Z are atomic numbers of projectile and target, respectively. One is thus led to a non-negligible correction factor for the effect of fluorescence yield variation which must be applied to the experimental x-ray ratio in order to extract the ionization cross-section ratio. The correction factor gives a reduction in the x-ray yield ratio which varies from 10% to 2% for target elements between Cl and Cu.

There is some slight additional evidence, however, which suggests that the previous expression for ξ overestimates the double-to-single ionization ratio. Using the Hartree-Fock-Slater program of Herman and Skillman[22] we calculate energy shifts $\Delta E_\alpha \approx 30$ eV and $\Delta E_\beta \approx 69$ eV for iron $K\alpha$ and $K\beta$ x rays emitted from an ionic configuration $(1s^{-1})$ $(2p^{-1})$ compared with those associated with a single K-shell vacancy. Thus, in x rays which are induced by α-particle bombardment, one should observe a shift in the average x-ray energy of the order of $\Delta E\ \xi/(1+\xi)$, which is 7 and 16 eV for the multiply ionized $K\alpha$ and $K\beta$ components, respectively. The presence of additional vacancies would, of course, further increase the observable shifts. The experimental evidence[23] however is that no shift or line broadening is observable with 30- and 70-MeV α particles within an uncertainty of ± 7 eV.

It is possible that some variations in fluorescence yields may be affecting the present x-ray yield measurements. However, this effect would not appear to be the main cause of the observed deviations.

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Direct-Transition Features in Stripping Collisions of Heavy Neutral Atoms and Ions*

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Previously published experimental data on single-electron stripping for over 100 different neutral-neutral and almost 100 ion-neutral collision pairs are compared with Firsov's statistical ionization model. Reasonable agreement is found for a number of reactions, in particular for collisions between some rare-gas atoms. However, marked disagreement exists for many other collisions. From a model assuming a single-step transition of the ionized electron from the bound state into the continuum, a scaling law is developed semiempirically which is found able to reduce a large majority of these published data to within a factor of two of a general cross-section curve. Major disagreements with this model occur only in cases which are particularly well described by the Firsov model.

I. INTRODUCTION

Over the last 15 years, extensive experimental data have been accumulated on ionization and stripping in collisions of heavy atoms, molecules, and/or ions. However, possibly with the exception of inner-shell ionization, comparatively little understanding of these processes has developed. This is particularly true for single-electron stripping processes in collisions between heavy neutral atoms. In spite of a long list of experimental results,[1–46] only very few theoretical calculations of the corresponding cross sections have been published so far. Good *a priori* calculations only exist for the lightest collision partners H and/or He. If heavier collision partners are involved, it is generally assumed that the ionization process proceeds by means of a large number of crossings and pseudocrossings of the interatomic-potential curves through which the electronic wave function "diffuses" during the encounter. Then, ionization supposedly occurs by auto-ionization from states above the ionization threshold. Correspondingly, any theoretical treatment appears to be necessarily statistical in nature. So far, three independent approaches in this direction have been published by Mittleman and Wilets,[47] by Russek and co-workers[48] and by Firsov.[49] Of these, only Firsov's paper arrives at actual predications for total cross sections independent of unknown phenomenological parameters. Since its publication, Firsov's formula has been compared and found to agree reasonably well with a limited set of experimental data, mostly involving collisions of rare-gas particles, and therefore is relatively widely quoted and accepted.

In this paper, a much wider range of experimental results on the stripping of neutral-atom and ion projectiles in collisions with neutral target atoms is collected, involving many projectiles from hydrogen to uranium and target gases ranging from rare gases to some alkalies and some diatomic gases. Comparing this set of data with Firsov's formula, a reasonable agreement of experimental and theoretical cross-section values is found around the maxima of the cross-section curves. However, considerable disagreement is observed in many cases where cross sections have been measured at smaller velocities, with the experimental values dropping much faster than predicted at the smaller energies. The latter behavior seems to indicate that, in these cases, the ionization process is dominated by a single large-step transition of the electron, from the ground state either directly into the continuum or into a highly excited state, rather than by a gradual statistical excitation.

To investigate this point, a scaling law, more adequate for direct ground-state–to–continuum transitions, is developed semiempirically with the aim to reduce these cross sections to a general functional dependence. The corresponding derivations are based on the assumption that the electron transition occurs simply on account of the time-dependent perturbation of the projectile by the target atom or molecule. Because of a general lack of detailed knowledge, in addition a number of somewhat arbitrary assumptions are introduced