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Energy Dependence of Multiple Inner-Shell Ionization of Al by Alpha-Particle Bombardment*

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The excitation of states in Al with single K-plus multiple L-shell vacancies formed by collisions with 0.4–3.0-MeV helium ions is studied by observing the x-ray decay associated with filling of the K-shell vacancy. The observed K x rays come from initial states with one K-shell and 0, 1, 2, and 3 L-shell vacancies. The absolute cross section for each of these transitions is determined and compared to a calculation using multiple Coulomb ionization. An important result of this experiment is the observation that the ratio of double to single ionization exhibits a maximum at a bombarding energy near the maximum for L-shell ionization.

I. INTRODUCTION

The application of high-resolution techniques to the study of x-rays produced in high-energy heavy ion-atom collisions has created an increasing interest in multiple inner-shell ionization phenomena.[1-9] An important aspect of the high-resolution studies has been the attempt to obtain the relative intensities of the satellites and to deduce relative ionization ratios which can be compared with different ionization theories.[10-12] Until now only relative satellite intensities have been reported because of the inherent nature of high-resolution crystal spectrometers. In this paper absolute cross sections of single K-shell, multiple L-shell ionization are reported. The results of high-resolution measurements on a thick target are combined with poor-resolution measurements of absolute x-ray yields as done by Basbas, Brandt, and Laubert.[13]

The bombardment of 0.4 to 3.0-MeV α particles on Al was chosen for two reasons. First, the poor resolution data available, as mentioned above,[13] makes the extraction of absolute cross sections possible. Second, Knudson *et al.*[10] have recently done a similar undertaking at higher energy. An extension of this investigation to lower energies yields especially fruitful information, since it includes data over the range for which the L-shell cross section is maximum. The prediction of this maximum should be of great theoretical importance in the test of any ionization theory. An overlap of the experiment done by Knudson *et al.* is given so that comparison between the two experiments can be made and a more complete picture obtained.

II. EXPERIMENT AND ANALYSIS

The experiment was performed with the University of Texas model KN Van de Graaff accelerator using α-particle beams of 17 energies between 0.4 and 3.0 MeV. The x-ray spectra were taken with a PDP-7 computer-controlled crystal spectrometer, as described elsewhere.[6-9] The energy resolution for a typical Al $K\alpha$ spectrum, as shown in Fig. 1, is 2.5 eV.

The x-ray spectra were taken with thick Al targets; therefore, in order to obtain correct x-ray yields, each x-ray line was corrected for beam-energy loss and self-absorption by the relation[14]

$$\sigma(E)=\left(\frac{4\pi}{N_t}\right)\left(\frac{dY}{dE}\ \frac{dE}{dx}\right)+\mu Y,$$

where Y is the x-ray yield per incident α particle, μ is the absorption coefficient in Al for each x-ray line, and N_t is the number of target atoms per cc. The values of μ used are 406, 397, 387, and 376 cm^2/g for the decay of states with defect or electron-hole configurations $(1K, nL)$ for $n=0,1,2,$ and 3, respectively.[15] The absolute experimental yields were obtained by normalizing to the data in Ref. 13. The raw yield $Y(1K, nL)$ of each x-ray line and the yield, Y(total), to which it is normalized, is given in Table I. $Y(1K, nL)$ was obtained by multiplying Y(total) by the ratio of the observed $K\alpha$ intensity for each configuration to the total observed $K\alpha$ intensity at each energy. The corresponding experimental x-ray-production cross sections, σ_x(total) and $\sigma_x(1K, nL)$, obtained are presented in Table II. The errors due to counting

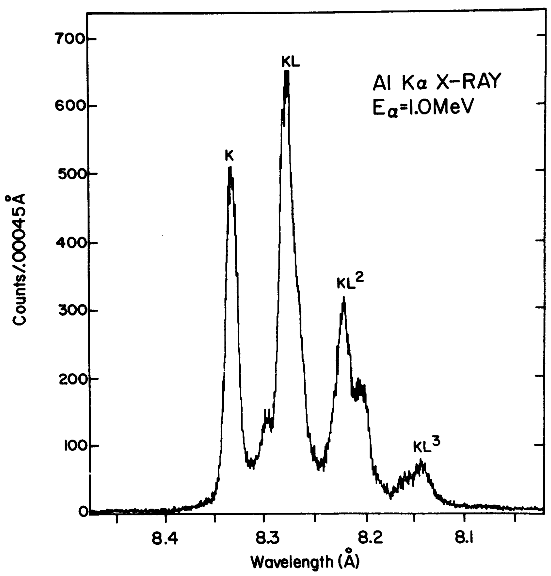

FIG. 1. Al $K\alpha$ x-ray spectrum produced by 1.0-MeV α particles. The peaks are labeled according to the number of K- and L-shell vacancies (e.g., KL^2 refers to a $K\alpha$ transition in an atom with one K-shell hole and two L-shell holes).

statistics are less than 1% in nearly all cases, and the quoted total error in the yields in Ref. 13 due to counting statistics is 3%. Their final total error is quoted as 25%. No attempt was made to estimate systematic errors due to corrections in this analysis.

To obtain an ionization cross section, the x-ray-production cross section of each configuration must be divided by the corresponding $K\alpha$ fluorescence yield for that particular configuration. Detailed calculations of fluorescence yields for defect configurations have recently been reported by Bhalla and Hein[16] for the case of neon. The calculations as outlined in that paper have been performed for some of the defect configurations of Al.[17] The values of the $K\alpha$ fluorescence yields $\omega_{K,nL}$ used are 0.041, 0.042, 0.044, and 0.045, corresponding to the electron-hole configurations $(1s)^1(2p)^n$ with $n=0, 1, 2,$ and 3 for Al. Ionization cross sections, $\sigma_{1K,nL} \equiv \sigma_x(1K, nL)/\omega_{K,nL}$, are given in Table III as percentages of the total Al K-shell ionization cross section $\sigma_{1K,nL}/\sigma_{\text{tot}}$ along with the total K-shell ionization cross section σ_{tot}. The ionization cross sections are also displayed in Fig. 2.

In interpreting these ionization cross sections two implicit assumptions have been made. (i) The K-shell hole is filled before any of the L-shell holes are filled. (ii) At the time of the $K\alpha$ transition all of the L-shell vacancies are in the $2p$ subshell. Assumption (i) asserts that the number of holes in the L-shell at the time of decay is the same as produced in the collision. No calculations have been made of the rates for filling L-shell vacancies when there is a K-shell hole. Walters and Bhalla[18] have calculated the rate for filling a $2p$ vacancy without any other holes present. For aluminum, this rate is less than 1% of the rate for a normal $K\alpha$ decay. The fluorescence yield used in obtaining the ionization cross section from x-ray production cross section is dependent on whether the L-shell holes are in the L_{I} subshell or in the $L_{\text{II,III}}$ subshell. The fluorescence yields used in the calculations are for all of the holes being in the $L_{\text{II,III}}$ subshell. The normal

TABLE I. Measured thick target Al K x-ray yield, Y, per incident α particle.

$E\alpha$ (MeV)	Y(Total) [a]	$Y(1K, 0L)$	$Y(1K, 1L)$	$Y(1K, 2L)$	$Y(1K, 3L)$
3.0	2.92×10^{-2}	9.98×10^{-3}	1.377×10^{-2}	4.86×10^{-3}	5.98×10^{-4}
2.8	2.30×10^{-2}	7.10×10^{-3}	1.087×10^{-2}	4.41×10^{-3}	6.10×10^{-4}
2.6	1.70×10^{-2}	5.04×10^{-3}	8.29×10^{-3}	3.18×10^{-3}	4.88×10^{-4}
2.4	1.25×10^{-2}	3.474×10^{-3}	5.868×10^{-3}	2.751×10^{-3}	4.06×10^{-4}
2.2	9.06×10^{-3}	2.44×10^{-3}	4.20×10^{-3}	2.020×10^{-3}	3.18×10^{-4}
2.0	6.23×10^{-3}	1.551×10^{-3}	3.012×10^{-3}	1.436×10^{-3}	2.296×10^{-4}
1.8	4.05×10^{-3}	9.35×10^{-4}	1.866×10^{-3}	1.060×10^{-3}	1.880×10^{-4}
1.6	2.48×10^{-3}	5.36×10^{-4}	1.160×10^{-3}	6.65×10^{-4}	1.185×10^{-4}
1.4	1.399×10^{-3}	2.886×10^{-4}	6.51×10^{-4}	3.87×10^{-4}	7.24×10^{-5}
1.2	6.97×10^{-4}	1.508×10^{-4}	3.282×10^{-4}	1.810×10^{-4}	3.69×10^{-5}
1.0	2.93×10^{-4}	6.56×10^{-5}	1.341×10^{-4}	7.83×10^{-5}	1.496×10^{-5}
0.9	1.748×10^{-4}	4.203×10^{-5}	8.07×10^{-5}	4.40×10^{-5}	7.99×10^{-6}
0.8	9.85×10^{-5}	2.556×10^{-5}	4.555×10^{-5}	2.339×10^{-5}	3.99×10^{-6}
0.7	5.24×10^{-5}	1.562×10^{-5}	2.349×10^{-5}	1.150×10^{-5}	1.78×10^{-6}
0.6	2.59×10^{-5}	8.62×10^{-6}	1.200×10^{-5}	4.61×10^{-6}	6.55×10^{-7}
0.5	1.182×10^{-5}	4.818×10^{-6}	5.23×10^{-6}	1.676×10^{-6}	9.15×10^{-8}
0.4	4.70×10^{-6}	2.295×10^{-6}	1.778×10^{-6}	6.26×10^{-7}	···

[a] Total yield normalized to the data of Basbas, Brandt, and Laubert.

TABLE II. Al $K\alpha$ x-ray production cross sections, σ_x, in barns. (Corrected for thick target.)

$E\alpha$ (MeV)	σ_x(Total)	$\sigma_x(1K,0L)$	$\sigma_x(1K,1L)$	$\sigma_x(1K,2L)$	$\sigma_x(1K,3L)$
3.0	1667	648	762	225	32.3
2.8	1478	549	683	211	35.2
2.6	1265	431	600	206	28.3
2.4	1029	326	499	181	22.5
2.2	790	238	382	152	18.1
2.0	600	170	288	125	16.8
1.8	455	119	215	103	17.4
1.6	334	80.0	156	83.1	14.9
1.4	233	51.0	109	62.6	10.7
1.2	148	30.2	68.8	41.5	7.08
1.0	81.1	16.4	37.9	22.7	4.10
0.9	55.4	11.4	25.9	15.1	2.95
0.8	35.6	7.84	16.7	9.29	1.80
0.7	21.3	5.11	9.99	5.16	1.00
0.6	11.7	3.18	5.46	2.59	0.46
0.5	5.86	1.89	2.70	1.14	0.12
0.4	2.7	1.05	1.19	.43	···

Coster-Kronig transition, which transfers L_{I} subshell holes to the $L_{\mathrm{II,III}}$ subshell, has a faster rate than the $K\alpha$ transition rate. It has been discussed previously[11] that Coster-Kronig transitions may not be energetically possible when there is also a K-shell hole. This would "freeze" the holes in the L_{I} subshell and be a possible source of error for the ionization cross sections given. This assumption does not affect the x-ray production cross sections as given in Table II.

III. DISCUSSION

Recently, two theories of Coulomb ionization have been modified so as to include multiple-ionization phenomena.[19,20] Both express the single K-shell, multiple L-shell cross section at an incident ion energy E, in terms of $P_K(E,b)$ and $P_L(E,b)$, and the probabilities for producing K-shell and L-shell ionization respectively at an impact parameter b, integrated over all values

TABLE III. Al K-shell ionization total cross sections, σ_{tot}, and ratios of the ionization cross section for each configuration to the total ionization cross section, $\sigma_{1K,nL}/\sigma_{\mathrm{tot}}$.

$E\alpha$ (MeV)	$\sigma_{\mathrm{tot}}\times 10^{-2}$ (b)	$\sigma_{1K,0L}/\sigma_{\mathrm{tot}}$	$\sigma_{1K,1L}/\sigma_{\mathrm{tot}}$	$\sigma_{1K,2L}/\sigma_{\mathrm{tot}}$	$\sigma_{1K,3L}/\sigma_{\mathrm{tot}}$
3.0	397	0.397	0.456	0.129	0.018
2.8	352	0.380	0.462	0.136	0.022
2.6	301	0.349	0.474	0.155	0.021
2.4	244	0.325	0.486	0.168	0.020
2.2	187	0.309	0.485	0.184	0.021
2.0	142	0.292	0.482	0.200	0.026
1.8	107	0.270	0.476	0.218	0.036
1.6	78.8	0.247	0.471	0.239	0.042
1.4	55.0	0.226	0.472	0.259	0.043
1.2	34.7	0.212	0.471	0.271	0.045
1.0	19.1	0.210	0.472	0.270	0.048
0.9	13.0	0.213	0.473	0.263	0.050
0.8	8.40	0.227	0.474	0.251	0.048
0.7	5.02	0.249	0.474	0.233	0.044
0.6	2.77	0.280	0.470	0.213	0.037
0.5	1.39	0.332	0.463	0.186	0.019
0.4	0.64	0.402	0.444	0.153	···

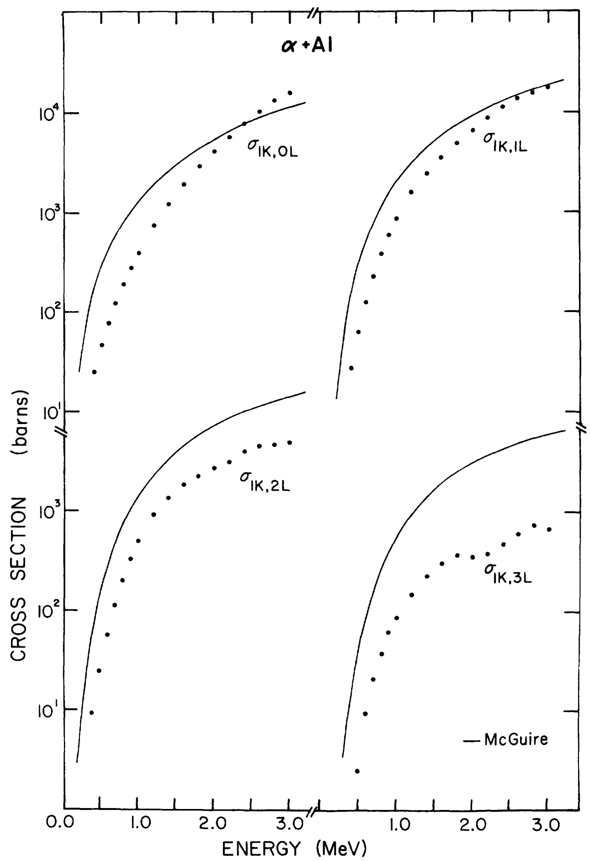

FIG. 2. Multiple ionization cross sections from α + Al for the electron-hole configurations $1K,nL$ with $n = 0$, 1, 2, 3. The solid curves are the predicted cross sections calculated in the modified BEA approximation.

of b. The differences in the two models arise from the approximations used. For the calculations in Ref. 20, the classical binary-encounter approximation (BEA) is formulated to calculate $P_L(E_i, b)$ [see Eq. (15) of Ref. 20]. The cross sections for single K-shell multiple L-shell ionization is then assumed to be given by a binomial distribution in these probabilities. This assumes that the electrons are not correlated in the ionization process. This assumption then leads to the following expression for the cross section [Eq. (24), Ref. 20].

$$\sigma_{1K,nL}(E_i) = 2\int_0^\infty 2\pi b\, P_K(E_i, b)\binom{8}{n} \times P_L^n(E_i, b)[1 - P_L(E_i, b)]^{8-n}\, db. \quad (1)$$

The quantities $\binom{8}{n}$ are the binomial coefficients. The cross sections as calculated from these formulae for α + Al are shown in Fig. 2 along with the measured results. From the energy of the x-ray transition it is not possible to distinguish $2s$ from $2p$ vacancies as discussed earlier; therefore all the L-shell electrons are treated equally in the calculation. This is done by assuming that all the eight electrons have a binding energy equal to the weighted average binding energy. This assumption does not affect the calculated values of $\sigma_{1K,0L}$, but does affect the calculated values of $\sigma_{1K,nL}$ for $n > 0$. It is seen that, while the data and the theoretical curves do not agree exactly, they do span the same orders of magnitude and have the same approximate shape. The calculations by Hansteen and Mosebekk,[19] use the semiclassical approximation (SCA) and are not available for

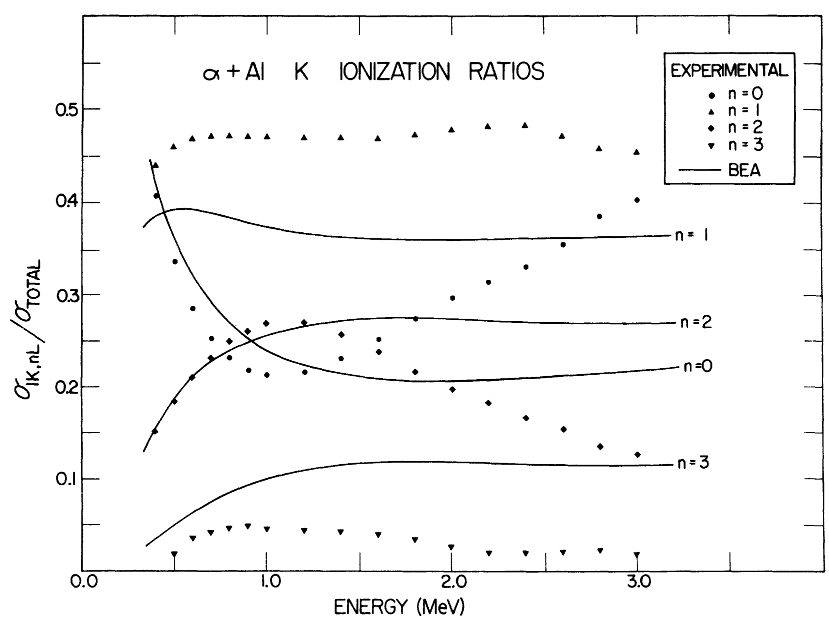

FIG. 3. Measured ratios of $\sigma_{1K,nL}/\sigma_{tot}$ for $n = 0$, 1, 2, 3. The solid curves are the ratios calculated in the modified BEA theory.

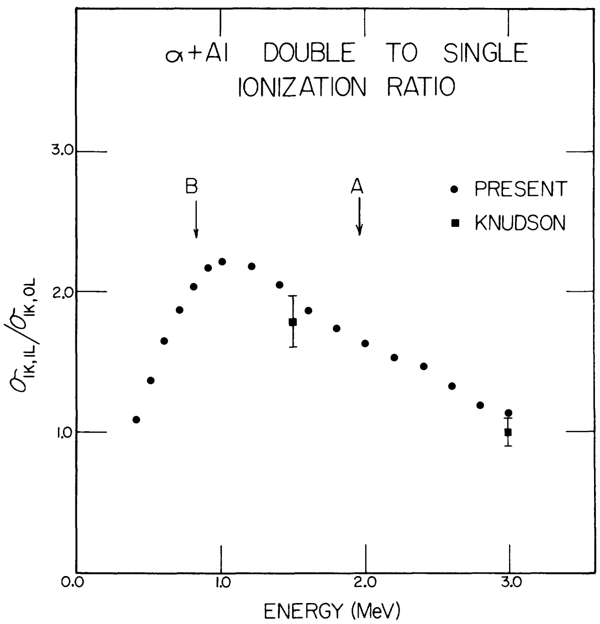

FIG. 4. Ratio $\sigma_{1K,1L}/\sigma_{1K,0L}$ exhibiting the peaking for maximum multiple ionization. Included is the data by Knudson *et al.* (Ref. 10) in this energy range. A is the position of the predicted maximum in the modified BEA theory. For reference the position of the maximum predicted for single L-shell excitation in the BEA theory is designated at B.

comparison with the α + Al case discussed here.

A linear plot of the ionization cross-section ratios, $\sigma_{1K,nL}/\sigma_{tot}$, given in Fig. 3 show finer details in the experimental results. For example, the ratio $\sigma_{1K,0L}/\sigma_{tot}$ exhibits a pronounced minimum at 1.0 MeV, whereas the ratios $\sigma_{1K,2L}/\sigma_{tot}$ and $\sigma_{1K,3L}/\sigma_{tot}$ exhibit pronounced maxima at that same energy. These observed extrema have not been previously reported in the literature and will serve as a crucial feature to be explained by any ionization theory. It is also interesting that the amount of triple ionization exceeds the amount of single ionization in the small energy range of 0.75 to 1.5 MeV. In the entire energy range reported here, double ionization exceeds single ionization, whereas at the higher energies reported by Knudson *et al.*[10] single ionization begins to dominate.

The results of BEA calculations are also presented in Fig. 3. In the high-energy region of these curves the ratios are essentially constant, implying little change in the amount of multiple L-shell ionization. In comparison to the theory the data in this energy region show a marked increase in the $\sigma_{1K,0L}/\sigma_{tot}$ curve and a decrease in the $\sigma_{1K,2L}/\sigma_{tot}$ and $\sigma_{1K,3L}/\sigma_{tot}$ curves. These features in the data indicate that the relative amount of multiple ionization is decreasing. The theory almost everywhere overestimates the amount of multiple ionization. This is seen, most contrastingly, in the overestimation of the $\sigma_{1K,3L}/\sigma_{tot}$ and $\sigma_{1K,2L}/\sigma_{tot}$ curves.

Probably the most important discrepancy between experiment and theory is the positioning of the extrema of the curves. The position of the extrema is the energy at which the degree of multiple L-shell ionization is the greatest. The modified BEA theory predicts maximum multiple ionization to occur at about $E_\alpha = 1.85$ MeV. To better see this effect in the data, $\sigma_{1K,1L}/\sigma_{1K,0L}$ is plotted in Fig. 4. This maximum is seen to come at $E_\alpha = 1.0$ MeV, 850 keV below the prediction. Also given in Fig. 4 for comparison are the data of Knudson *et al.*[10]

In summary, the energy dependence of the x-ray production cross sections for single K-shell, multiple L-shell ionization have been measured for the first time. The ionization cross sections have been obtained from the x-ray production cross sections by treating the $2s$ and $2p$ electrons as identical and using the calculated fluorescence yields $\omega_{1K,nL}$ for the electron-hole configurations $(1s)(2p)^n$. These cross sections are then compared to a modified BEA calculation. The gross features of the data are explained fairly well; however, under close scrutiny several areas of disagreement are observed. The data exhibits a fairly narrow pronounced maximum in the ratio $\sigma_{1K,1L}/\sigma_{1K,0L}$, whereas the theory predicts a broad maximum at a much higher energy. A few possible reasons for the discrepancy between the observed and calculated cross sections are (i) that the modified BEA theory fails to accurately describe multiple ionization; (ii) no electron correlation effects are included in the present calculations; (iii) the correct binding energies to be used in the calculations are subject to interpretation. The different L subshells have different binding energies. To simplify this problem a weighted average of the L-subshell binding energies is used as suggested in Refs. 19 and 20. Also the interpretation of the correct binding energy is complicated by the effect of the incident projectile on the bound electrons and the ionization state of the target; and (iv) the $K\alpha$ fluorescence yields are sensitive to the position of the vacancies in the L shell.

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Procedure for Computing Cross Sections for Single and Multiple Ionization of Atoms in the Binary-Encounter Approximation by the Impact of Heavy Charged Particles*

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A procedure is developed for computing cross sections for the multiple ionization of atoms by the impact of protons or other fully stripped nuclei. The ionization probability, as a function of energy and impact parameter, $P(E,b)$, is computed at several beam energies in the binary-encounter approximation for a ground-state hydrogenic electron scattered by an incident proton. Scaling laws are given which may be used to extend these results to other projectiles, other targets, and other hydrogenlike filled atomic shells. It is shown that $P(E,O) = \langle \sigma(E,r)/2\pi r^2 \rangle$ for isotropic, but otherwise arbitrary, electron-density distributions. A formulation for multiple-ionization cross sections is developed in terms of the single-electron probabilities $P(E,b)$ for each atomic shell, assuming that both the electrons and the shells are mutually independent. Numerical calculations are compared to recent predictions in the semiclassical Coulomb approximation and to recent satellite and hypersatellite x-ray data. The discrepancies are generally within those resulting from uncertainties of 30–200% in the single-ionization cross sections, when the ionization probability is much less than one. Then, approximating $P(Eb)$ vs b as a step function, the multiple-ionization cross sections are reduced to simple combinations of single-ionization cross sections. These single-ionization cross sections may be evaluated in the binary-encounter approximation by applying scaling laws to the usual universal curve that we tabulate. Multiple-ionization cross sections may thus be estimated without the aid of a computer.

I. INTRODUCTION

In the past several years there has been a resurgence of interest in ionization phenomena, in part owing to advancing experimental technique and in part owing to applications in other fields. Considerable progress has been made in developing approximate calculations of cross sections for the single ionization of atoms by the impact of charged particles, even though exact calculations have not been done. The approximate calculations are simple and may be applied to reasonably complex systems. Recently there have been observations of x-ray satellite[1] and hypersatellite[2] transitions corresponding to multiple ionization of atoms and molecules. Interpretations[3] and calculations[4] which have described the atomic data have quickly followed.

In this paper, simple formulas and tables are compiled which may be easily used to compute cross sections for the single ionization of atoms by the impact of charged particles in the binary-