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Projectile and Target Dependence of the $K\alpha$ Satellite Structure*

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Proton, α-particle, and oxygen-ion beams are used to induce $K\alpha$ x-ray spectra in the elements Ca, Sc, Ti, V, Cr, and Mn. The target dependence of the satellite structure for $Z = 20$ to $Z = 25$ is studied. The relative intensity ratios for the production of one K-shell and multiple L-shell vacancies are measured. These ratios are compared with the predictions of the binary-encounter approximation (BEA) and the semiclassical approximation (SCA) for multiple ionization. The data strongly reflect the statistical approach used in the theories. The BEA theory agrees well with the proton and α-particle data but does not give good agreement with the oxygen data.

I. INTRODUCTION

Recently, there has been much interest in inner-shell ionization by heavy-ion bombardment. In the 1-MeV/amu range, the cross section for K-shell ionization has been predicted with some success by the modified Born approximation or by the binary-encounter model.[1-4] In recent experiments the x rays resulting from inner-shell ionization have been resolved into a number of satellite peaks,[5-8] which are due to single K-shell, multiple L-shell ionization. The intensities of these satellite peaks have been predicted in the semiclassical-approximation (SCA) formulation.[9] The results are expressed as an integral over the impact parameter of a product of single-ionization probabilities. The prediction of the peak intensities has also been done in the same manner using the binary-encounter approximation (BEA) to obtain values of the single-ionization probabilities.[10] In this paper a study is made of the dependence of the relative intensity of the multiple ionization produced as a function of the target for three projectiles. The $K\alpha$ satellite lines produced by proton, α-particle, and oxygen beams on targets of Ca through Mn are measured in high resolution. The measured intensities are examined to test the assumption that the multiple-ionization intensities can be expressed as a product of single-ionization probabilities.[11] In addition, the data are compared with the particular predictions of the SCA and BEA approximations.

II. EXPERIMENT

In the experiment the 0.8-MeV proton and 3.2-MeV α particles were produced by the University of Texas, model KN particle accelerator. The current of each beam was about 1 μA. The O^{5+} beam was produced by the University of Texas model EN tandem Van de Graaff accelerator. The beam energy for the Ca, Sc, Ti, V, Cr, and Mn targets was 30 MeV with a current between 10 and 100 nA. The targets of the various elements used were either foils or metal plates. Their thicknesses ranged from 0.5 to 800 mils. All targets were thick enough to stop the beam, and each was placed at an angle of 45° with the beam.

To analyze the spectrum a Bragg crystal spectrometer equipped with a LiF crystal was used. It was placed perpendicular to the beam at a 45° angle to the target. A flow proportional counter was used to detect the x rays. The pulses from the detector after amplification were sent through a single-channel analyzer. An energy gate was set to minimize the background in each spectrum and the data were collected in a PDP-7 computer which stepped the spectrometer. The counting time for each spectrometer setting was controlled by a current integrator. In this way effects due to beam fluctuations were minimized, and the same amount of charge struck the target for each setting.

In Fig. 1 the $K\alpha$ spectra are shown for oxygen bombarded on each element. The peaks are labeled according to the primary L-shell electron configuration of the initial state. The proton- and α-induced spectra are not shown here. The resolution ranges from about 7-eV full width at half-maximum (FWHM) for calcium to about 34-eV FWHM for manganese. The relative intensities are determined by fitting each peak to a Gaussian distribution and calculating the area under each curve. The uncorrected intensities are normalized to the total intensity of the $K\alpha$ spectrum. These are listed in Table I. The labeling of the columns, KL^n, corresponds to the number of vacancies in the K and L shell (e.g., KL^2 designates one K-

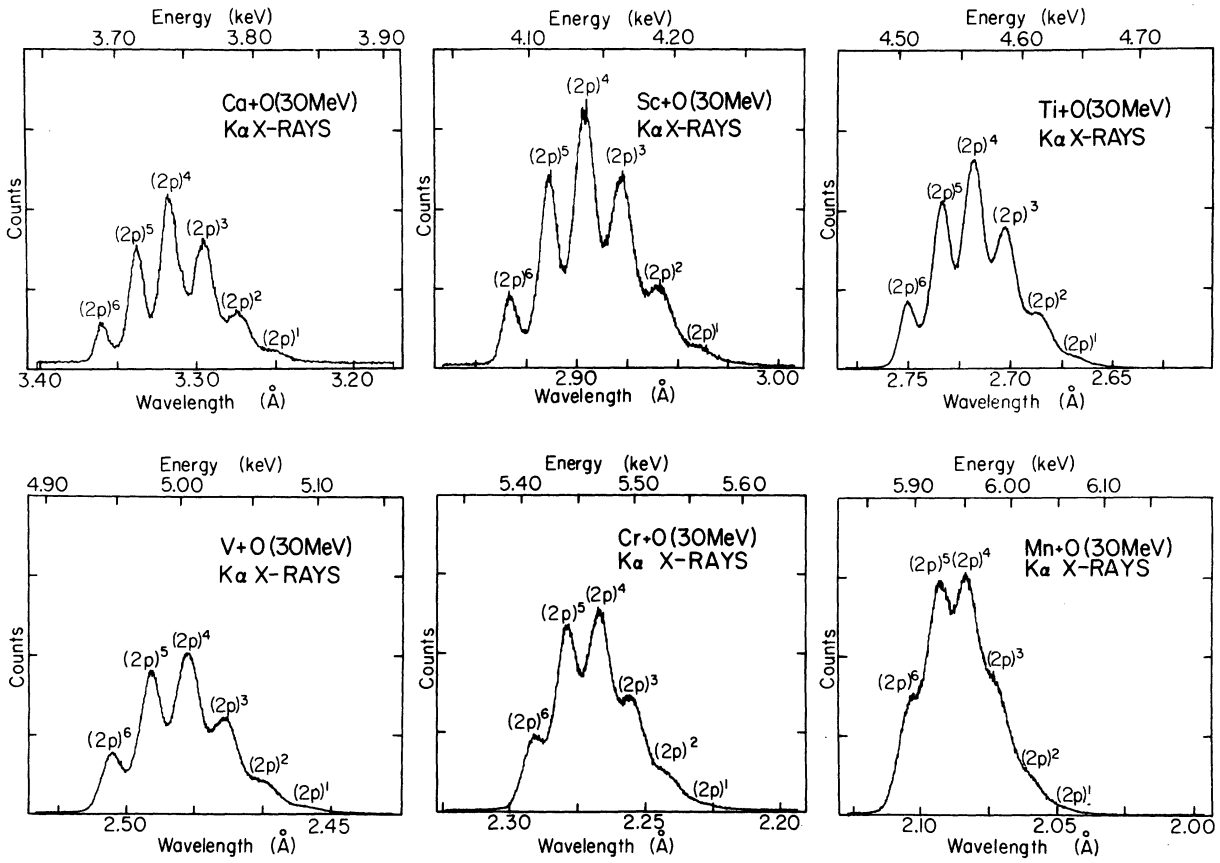

FIG. 1. $K\alpha$ spectra for calcium through manganese. The peaks are labeled according to the primary L-shell electron configuration of the initial state. The decreasing resolution is due to the smaller Bragg angle.

shell vacancy and two L-shell vacancies).

To correct the thick-target yields, data were taken at energies above and below each set of data points. The data were then normalized to the cross sections measured by Burch *et al.* before corrections were made.[12] Each peak was then corrected independently using the formula given by Merzbacher and Lewis.[13] The stopping powers were obtained by interpolation of the tables by Northcliffe and Schilling.[14] The mass attenuation coefficients were taken from the tables calculated by Dewey *et al.*[15]

To obtain a $K\alpha$ fluorescence yield, the statistical approach described by Larkins is used.[16] The individual rates are those calculated by Walters and Bhalla.[17] To apply this method exactly, the precise defect configurations should be known. In this experiment only the occupation of the L shell could be measured; thus the calculations are done assuming a full M shell. The M-shell rates are small compared with the L-shell rates in the normal configuration. The M-shell rates become increasingly more important with the increase of L-shell vacancies, and therefore the corresponding fluorescence yields are more sensitive to the M-shell population. Also in performing the calculations it is assumed that all of the L-shell vacancies occur in the $L_{\mathrm{II,III}}$ subshell. In the normal configuration the rates of the Coster-Kronig transitions, which transfer L_{I} vacancies to the $L_{\mathrm{II,III}}$ subshell, are much larger than the rates of the transitions involving the K-shell vacancy.[18] For a large number of L-shell vacancies, the rates may be altered in such a way that this may not be the case.[19] If this is true, then this may be another source of error in the fluorescence yield.

In the second part of Table I, the relative ionization cross sections R_{nL} are given. R_{nL} is the ratio of the cross section, $\sigma_{K,nL}$, for the configuration with one K-shell and n L-shell vacancies divided by the total K-shell cross section σ_K. The ratios are normalized to one for each case studied. The errors due to counting statistics and to the fitting of the peaks are less than 5%. The extraction of cross sections from thick-target yields normally introduces large errors in the final results. When applied to this data it was found that the total cross sections changed greatly, but the ratios R_{nL} were

never altered by more than 10% from the uncorrected ratios. Using this value as an estimate of the error due to thick-target corrections, the total error in R_{nL} is evaluated as 12%. This does not include any error arising from the calculations of the fluorescence yields.

III. DISCUSSION

For the case of single ionization of the Ith shell which contains N electrons, the ionization cross section σ_I can be written, neglecting correlation effects, as an integral over the impact parameter b of the probability of ionization per electron of the Ith shell, $P_I(b)$. The expression is

$$\sigma_I = N\int_0^\infty 2\pi b P_I(b)\,db. \quad (1)$$

This expression has been extended to calculate the cross section for multiple ionization.[9,10] Assuming that the probability for ionizing each subshell of the L shell is the same, the expression for single K-shell, n L-shell ionization cross section $\sigma_{1K,nL}$ is

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

where $n = 0, 1, \ldots, 8$. A factor of $1 - P_K(b)$ is approximated as one because $P_K(b)$ is small. The relationship between $P_K(b)$ and $P_L(b)$ can be seen in Fig. 2. In the region where $P_K(b)$ is nonzero, $P_L(b)$ is almost constant and can be approximated

TABLE I. Uncorrected and corrected relative intensity ratios.

Incident projectile	Element	Relative $K\alpha$ x-ray intensities (uncorrected) K	KL	KL^2	KL^3	KL^4	KL^5
0.8-MeV p	Ca	0.907	0.093				
	Sc	0.916	0.084				
	Ti	0.925	0.075				
	V	0.955	0.045				
	Cr	0.968	0.032				
	Mn	0.977	0.023				
3.2-MeV α	Ca	0.685	0.277	0.039			
	Sc	0.720	0.242	0.038			
	Ti	0.769	0.201	0.030			
	V	0.807	0.181	0.012			
	Cr	0.860	0.134	0.006			
	Mn	0.920	0.080				
30-MeV O	Ca	0.058	0.195	0.323	0.274	0.112	0.040
	Sc	0.065	0.207	0.315	0.264	0.114	0.034
	Ti	0.083	0.240	0.323	0.224	0.107	0.022
	V	0.106	0.264	0.331	0.200	0.040	0.020
	Cr	0.112	0.278	0.333	0.192	0.071	0.014
	Mn	0.160	0.267	0.332	0.153	0.071	0.017
		Relative ionization intensities R_{0L}	R_{1L}	R_{2L}	R_{3L}	R_{4L}	R_{5L}
0.8-MeV p	Ca	0.913	0.087				
	Sc	0.921	0.080				
	Ti	0.929	0.071				
	V	0.958	0.042				
	Cr	0.969	0.031				
	Mn	0.978	0.022				
3.2-MeV α	Ca	0.701	0.264	0.035			
	Sc	0.734	0.231	0.035			
	Ti	0.779	0.193	0.028			
	V	0.816	0.173	0.011			
	Cr	0.865	0.129	0.006			
	Mn	0.923	0.077	...			
30-MeV O	Ca	0.066	0.205	0.313	0.254	0.103	0.060
	Sc	0.079	0.223	0.294	0.228	0.126	0.050
	Ti	0.091	0.232	0.307	0.207	0.129	0.035
	V	0.110	0.262	0.300	0.212	0.087	0.029
	Cr	0.122	0.257	0.329	0.196	0.071	0.026
	Mn	0.150	0.250	0.324	0.159	0.076	0.042

by its value at $b=0$.[11] In the case shown in Fig. 2, $P_L(b)$ changes by less than 1% over the values of the impact parameter for which $P_K(b)$ is nonzero. This allows the factors containing P_L to be removed from the integral sign of Eq. (2). The new expression is

$$\sigma_{1K,nL} \cong \binom{8}{n} P_L(0)^n [1-P_L(0)]^{8-n} 2\int 2\pi b P_K(b)\, db$$

$$= \binom{8}{n} P_L^n(0)^n [1-P_L(0)]^{8-n} \sigma_K. \qquad (3)$$

Equation (3) gives an expression which predicts the values R_{nL}, and this prediction depends upon only one number, $P_L(0)$. If the primary cause of the vacancies is Coulomb ionization and if the statistical formulation is correct, then the data should reflect this statistical behavior.

To test the data for this behavior the experimental values R_{nL} are fitted with the binomial distribution as given by Eq. (3) by varying $P_L(0)$ until the sum

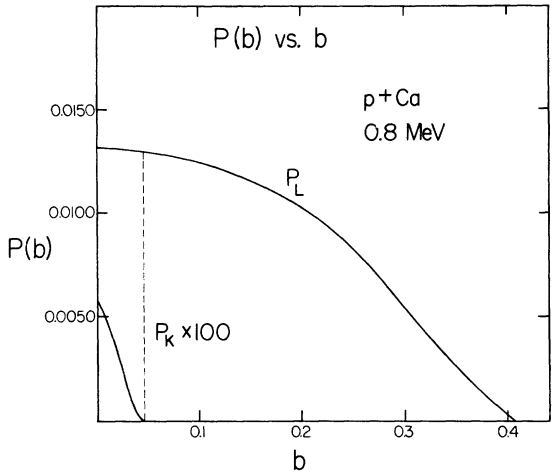

FIG. 2. $P_K(b)$ and $P_L(b)$ as a function of b for protons on calcium at 0.8 MeV. The calculations are done using the BEA theory. (See Ref. 10). The value of $P_L(b)$ is almost constant in the region where $P_K(b)$ is nonzero.

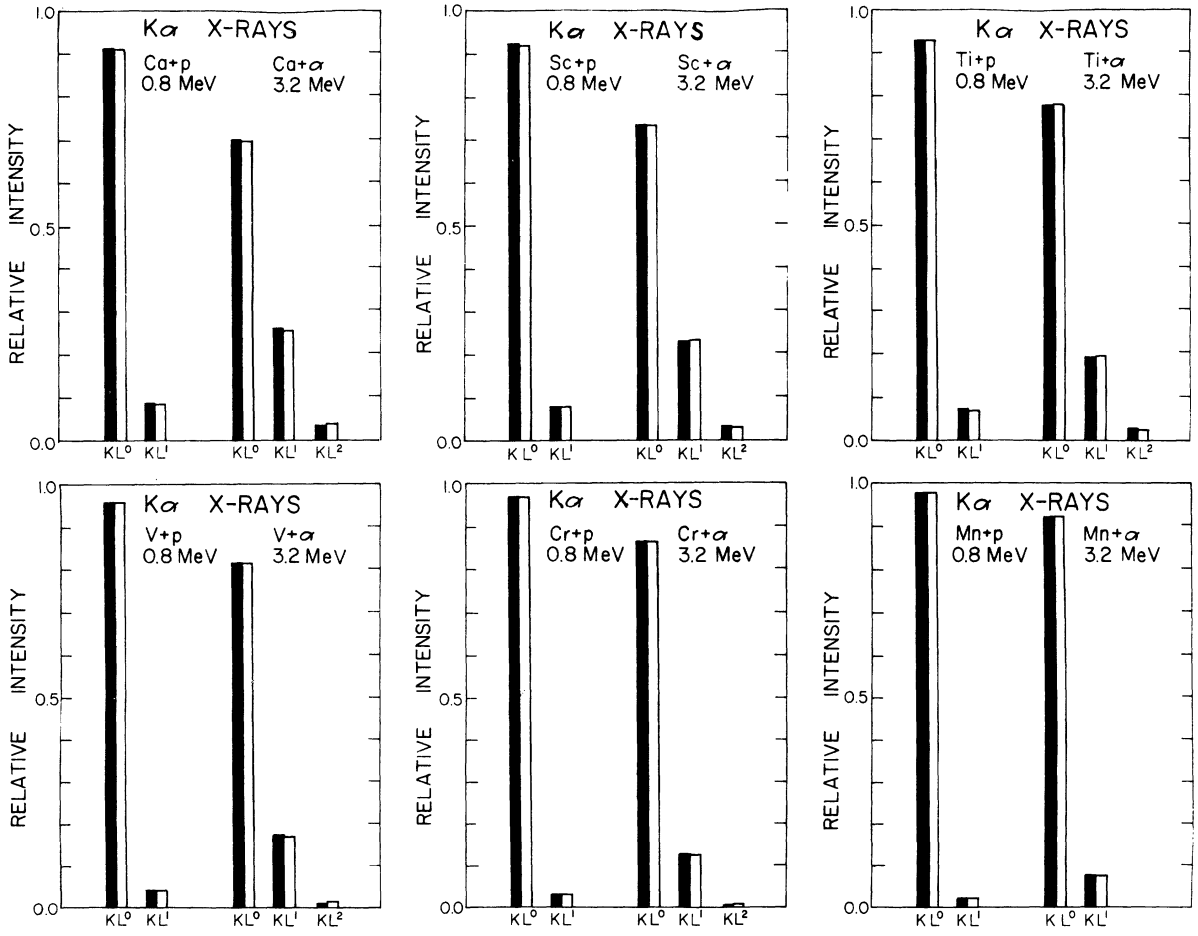

FIG. 3. Relative intensity ratios for protons and α-particle projectile at 0.8 MeV/amu. The solid bars are the measured ratios from Table I. The light bars are the values obtained by fitting the data to a binomial distribution. The final values of the fitting parameter are given in Table II.

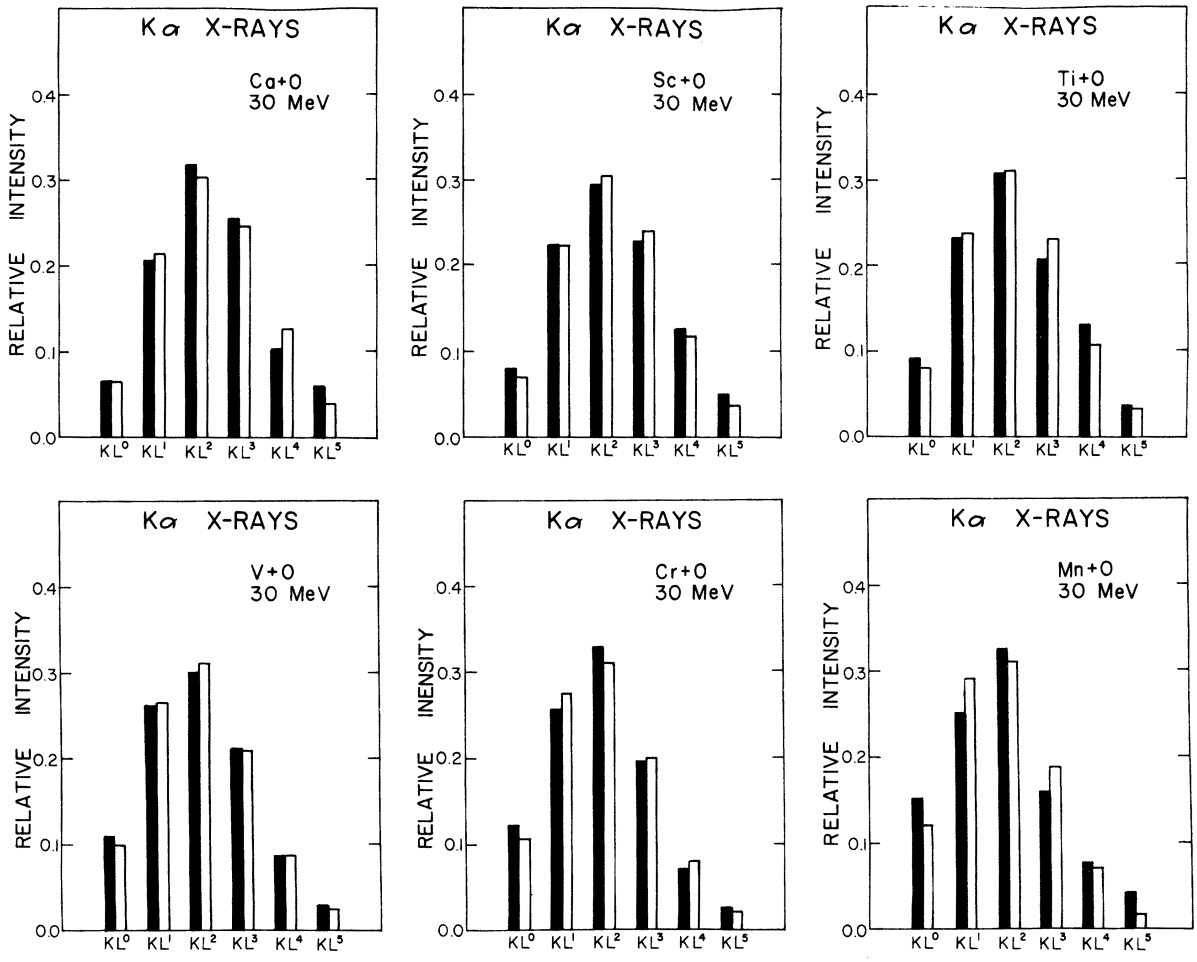

FIG. 4. Relative intensity ratios for oxygen projectile at 30 MeV. The solid bars are the measured ratios from Table I. The light bars are the values obtained by fitting the data to a binomial distribution. The final values of the fitting parameter are given in Table II.

of the squares of the differences is a minimum. The results of the fits are shown in Figs. 3 and 4. The set of bars marked KL^n correspond to the values of R_{nL}. The solid bars are the data given in Table I. The light bars are the values predicted by the fitting procedure. From the figures it can be seen that the data strongly resemble the statistical behavior predicted in Eq. (3).

In Table II the values of the parameter found by the fitting procedure are given. These are labeled as experimental values since they have been determined from data. The quoted errors are the fitting errors only. Also given in Table II are values of $P_L(0)$ as calculated in the BEA and SCA theories. The BEA values, in all cases, are at least in qualitative agreement with the experimental values. The proton and α-particle data agree well with the BEA values, but in all cases, the BEA theory overpredicts the amount of multiple ionization. The SCA calculations are done using harmonic-oscillator wave functions in the dipole approximation by Wu *et al.*[20] The use of these wave functions makes the calculations much simpler, but one should not expect them to give good quantitative agreement. Calculations using hydrogenic wave functions[9] give a value which is over four times smaller than the harmonic-oscillator calculations for protons on copper at 1 MeV. This would bring the SCA calculations into better agreement with the experimental values. As can be seen in Table II, the calculations, even using the crude model, give qualitative agreement with the experimentally extracted numbers. These are all greater than the experimental $P_L(0)$ so that, if the calculations are performed using more realistic wave functions, better agreement is expected.

The integral expressions, Eq. (2), can be evaluated directly and compared with the measured

TABLE II. Measured and calculated probability.

Element	$P_L(0)$ Expt.	$P_L(0)$ BEA	$P_L(0)$ SCA [a]
	Protons		
Ca	0.0115 ± 0.0003	0.0134	0.0199
Sc	0.0105 ± 0.0003	0.0110	0.0210
Ti	0.0093 ± 0.0002	0.0093	0.0203
V	0.0054 ± 0.0001	0.0079	0.0195
Cr	0.0040 ± 0.0001	0.0067	0.0193
Mn	0.0028 ± 0.0001	0.0057	0.0189
	α particles		
Ca	0.0440 ± 0.0010	0.0537	0.0795
Sc	0.0380 ± 0.0003	0.0438	0.0840
Ti	0.0305 ± 0.0006	0.0372	0.0811
V	0.0254 ± 0.0005	0.0300	0.0782
Cr	0.0181 ± 0.0002	0.0269	0.0772
Mn	0.0101 ± 0.0002	0.0228	0.0758
	Oxygen		
Ca	0.289 ± 0.007	0.918	0.494
Sc	0.282 ± 0.005	0.803	0.509
Ti	0.271 ± 0.007	0.715	0.522
V	0.251 ± 0.003	0.633	0.545
Cr	0.244 ± 0.006	0.558	0.546
Mn	0.233 ± 0.012	0.493	0.542

[a] Calculations are done using harmonic-oscillator wave functions (see Ref. 20).

R_{nL}. These calculations have been done using the BEA theory[10] and are shown in comparison with the experimental values in Figs. 5 and 6. These figures reflect the same relationship between the data and the experiment as $P_L(0)$ does. For the proton and α-particle data the BEA theory agrees well with the experiment, but it consistently overpredicts the amount of multiple ionization. For the oxygen data, the BEA theory greatly overestimates the amount of multiple ionization. Part of this effect may be due to the screening of the nuclear charge by some bound electrons. The equilibrium charge[21] of oxygen at 30 MeV is approximately +7. Because the BEA theory obeys a z^2-scaling law, this would reduce the values of $P_L(0)$ by a factor of one-fourth. The theoretical values would still overpredict the amount of multiple ionization. The discrepancy probably lies in the calculation of $P(b)$ itself. A small error in $P(b)$ is greatly magnified in the cross sections for large amounts of multiple ionization. For heavy ions like oxygen, where multiple ionization dominates, the cross sections

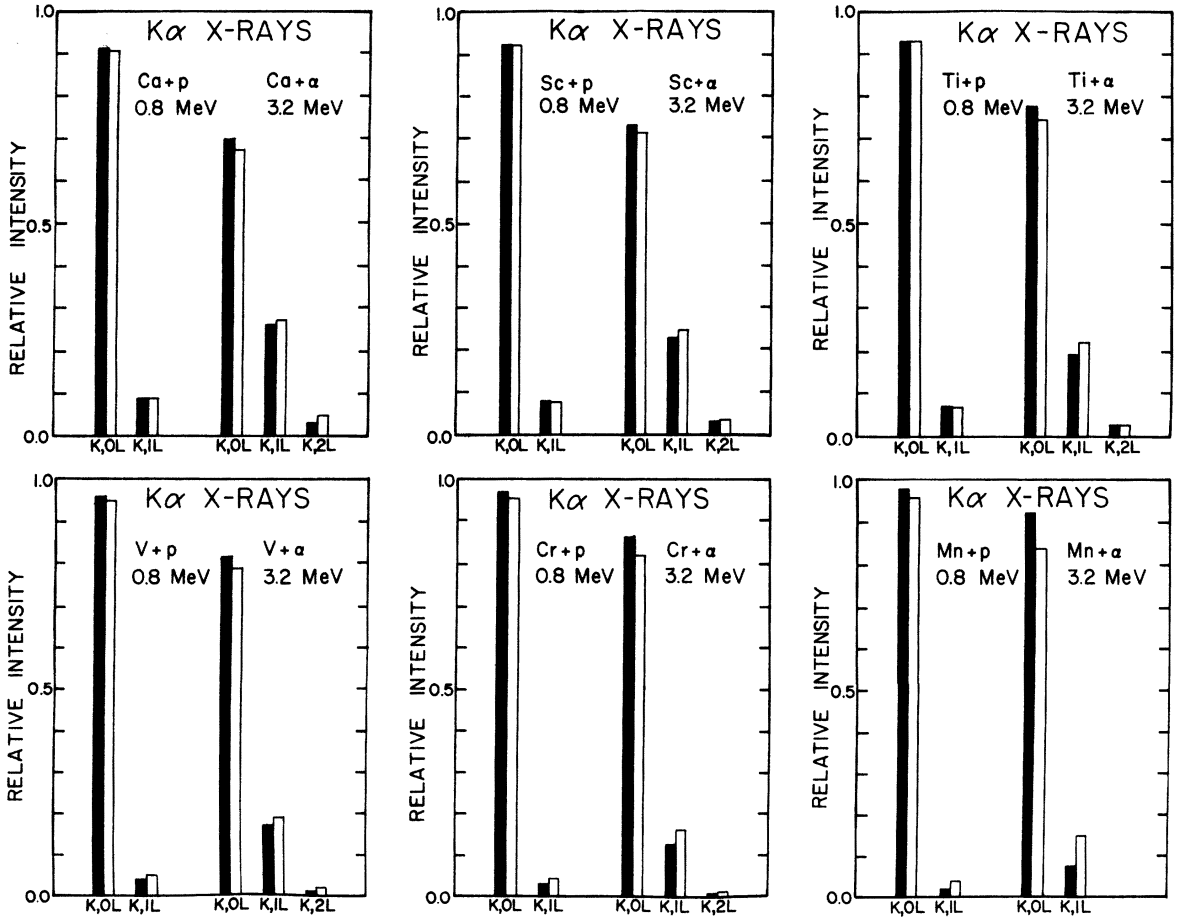

FIG. 5. Intensity ratios predicted by the BEA theory compared with the measured ratios for proton and α-particle projectile. The solid bars are the measured ratios. The light bars are the theoretical values calculated using Ref. 10.

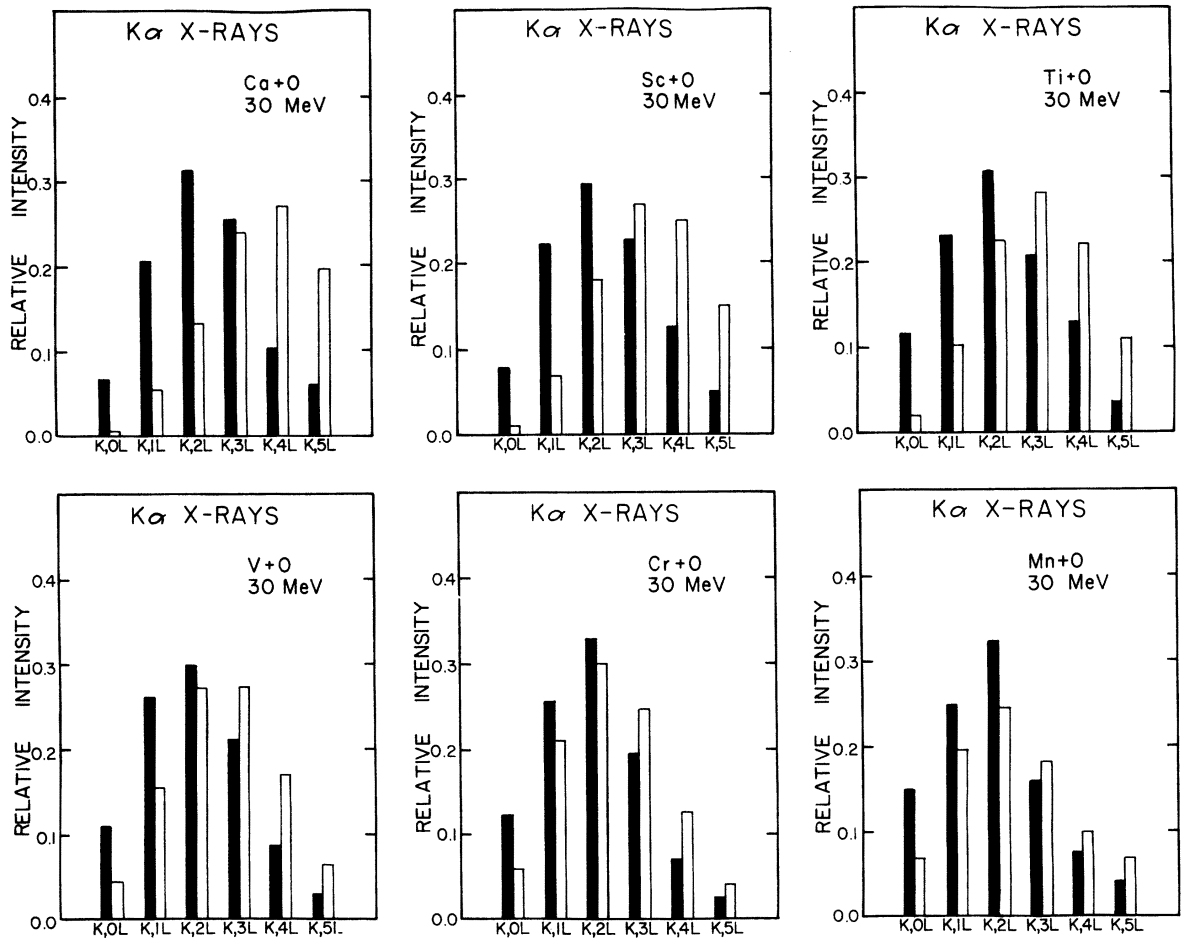

FIG. 6. Intensity ratios predicted by the BEA theory compared with the measured ratios for oxygen projectile. The solid bars are the measured ratios. The light bars are the theoretical values calculated using Ref. 10.

mostly consist of terms involving an integral of a product of $P(b)$. Thus small errors in $P(b)$ are multiplied together yielding large errors in the results. This trend can definitely be seen by comparing the three sets of data.

In summary, the statistical approach used in predicting multiple Coulomb ionization is well supported by the data. No comparison can be drawn between the BEA and SCA theories until better SCA calculations are available, but both give qualitative agreement with $P_L(0)$, which is a good estimate of the amount of multiple ionization. The failure of the BEA theory to agree with the oxygen data may be in part due to the deviation of the oxygen ion from a bare nuclear charge. More probably, the difference is the systematic overprediction of multiple ionization by the BEA theory, itself. Since the SCA calculations are first-order perturbation theory, it is expected that the results would not be valid for large ionization probabilities. No such restrictions exist for the BEA calculations. However, for heavy ions the calculations give unphysical predictions for the ionization probabilities. Despite the failure of the BEA for large z, it is shown in this paper that the predictions are in good agreement for protons and α particles at 0.8 MeV/amu.

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