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Zn K- and Pb L-shell ionization by proton and ^{3}He-ion bombardment

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Absolute cross sections and excitation functions have been measured for Zn K-x-ray production by proton impact and for Pb L-x-ray production by proton and ^{3}He-ion impact over the bombarding energy range from 1.4 to 4.4 MeV using a Si(Li) detector. The total ionization cross sections for the Zn K shell and x-ray production cross sections for the Pb L shell obtained are compared with the binary-encounter approximation (BEA) and the plane-wave Born approximation calculations and the agreement is found to be good. X-ray production ratios, such as $L\alpha/L\beta$ and $L\alpha/L\gamma$, for Pb are also compared with the BEA calculations. The cross-over behavior of the Z_1 (projectile nuclear charge) dependence of the x-ray production cross sections was studied for the Pb L lines.

I. INTRODUCTION

A considerable amount of experimental work on innershell ionization has been published since Lewis, Simmon, and Merzbacher[1] reported the first modern measurements of K-x-ray production by proton impact. As early as 1958, Merzbacher and Lewis[2] had developed the plane-wave Born approximation (PWBA) for innershell ionization.[3] Calculations using the PWBA are generally in agreement with experimental values at higher energies. At lower energies, however, values calculated in the PWBA are higher than the experimental results. This is believed to be due partly to the Coulomb repulsion between the projectile and the target nucleus, and partly to an increase in the binding energy of the electron as a result of the close proximity of the projectile, both of which are neglected in the PWBA. These effects are taken into account in the theories of Bang and Hansteen[4] and of Brandt *et al.*,[5] which are in better agreement with the experimental results.

On the other hand, Garcia[6] has developed the binary-encounter approximation (BEA) for innershell ionization, which gives a simple scaling law: that is, the product of the ionization cross section and the square of binding energy U of the electron is constant for a given E/U, where E is the ion energy. The agreement with experiment is generally better in the BEA than in the PWBA over a broad range of projectile energies.

Experimental values of the ionization cross sections σ_i are usually derived from x-ray yield measurements, except for a few that have been deduced from Auger-electron measurements. In order to derive σ_i from an x-ray production cross section σ_x, a knowledge of the fluorescence yield ω is necessary. The fluorescence yield of the K shell, ω_K, is rather precisely known for many elements, except for those with very low Z.[7] However, the fluorescence yield of the L shell, ω_L, is still inaccurately known except for the very-high-Z elements. Furthermore, it has been established experimentally[8-10] and theoretically[11] that ω is different for different electron configurations at the time of x-ray emission; that is, ω is generally larger for multiply ionized states than for singly ionized states. This effect is remarkably large in heavy-ion impact ionization.[9,10] Therefore, precautions must be taken in choosing ω when we attempt to determine σ_i using σ_x. However, it is generally thought that the variation of ω is small for light-ion (e.g., proton and helium-ion) impact ionization.

As far as K-shell ionization is concerned, many measurements have been made and, generally, they are in good agreement with the theories of the BEA and PWBA. However, very few measurements of L-shell ionization for high-Z elements have been published. Recently, Shafroth *et al.*[12] reported a precise measurement of Au L-shell x-ray production by proton impact.

In the present paper, we present results of measurements of the absolute cross sections for the production of Zn K-shell x-rays and of Pb L-shell ($L\alpha$, $L\beta$, $L\gamma$, Ll, and $L\eta$) x rays by proton impact over the bombarding energy range from 1.4

to 4.4 MeV. The results of the Pb L-x-ray measurements are compared with those of Busch *et al.*,[13] which were published in the course of the present study. We also present the Z_1 dependences of the Pb L-shell ionization cross sections by proton and helium-ion impact, which, to our knowledge, have not been reported previously.

II. EXPERIMENTAL TECHNIQUES

An ion beam of either protons or ^{3}He ions accelerated by the 5-MeV electrostatic accelerator at Tohoku University entered the target chamber through a graphite slit 3 mm in diameter. After passing through a thin target foil, the beam was stopped at the end of a Faraday cage which was deep to prevent the background x-rays produced therein from reaching the x-ray detector. A self-supported thin target, prepared by the usual vacuum-evaporation technique, was positioned in the vacuum chamber at an angle of 45° with respect to the beam direction. X rays, produced in the target by ion impact, passed through a 10-μm Mylar window, a 15-μm aluminium absorber, and 2.2 cm of air before reaching an ORTEC Si(Li) x-ray detector with a 25-μm Be window, which was placed normal with respect to the beam direction. The energy resolution of the x-ray detector was 205 eV for 6.4-keV Fe $K\alpha$ x-rays and its absolute efficiency was determined with intensity-calibrated ^{57}Fe and ^{241}Am x-ray sources. The inside of the brass target chamber was covered with aquaduc-coated 5-mm-thick acrylite plates in order to reduce the background x-rays produced by scattered ions striking the chamber wall.

Assuming that the x-ray production is isotropic, the x-ray production cross section σ_x is calculated by

$$\sigma_x = (4\pi/\Delta\Omega)(Y/\epsilon n), \qquad (1)$$

where $\Delta\Omega$ is the solid angle subtended by the x-ray detector, Y is the x-ray yield per projectile corrected for the self-absorption in the target, the absorption of the Mylar window, the air, and the aluminium absorber, ϵ is the detection efficiency of the detector, and n is the number of target atoms per cm^2. The value of $\Delta\Omega$ was determined from the geometry to be 5.16×10^{-4} sr. The photoabsorption cross sections for the air and the target materials were taken from a compilation by Storm and Israel.[14] The absorption in the aluminium was determined experimentally. The corrections for the absorption of x rays in the target are rather complicated for the $L\beta$ and $L\gamma$ x-ray groups because the absorption edges of the L_2 and L_3 subshells, respectively, are found within the groups. However, since a thin target was used, the correction is small. For example, when we take the photoabsorption cross sections above and below the L_3-subshell absorption edge, where the jump in photoabsorption is greatest, the difference in absorption is only 1%. Therefore, the average values of photoabsorption cross sections in the energy range of these x-ray groups given by Storm and Israel[14] were used. n was obtained from measurements of the projectile ions scattered from the target. These ions were detected using a surface barrier detector placed at an angle of 136° with respect to the beam direction. From this measurement and the Rutherford scattering cross sections, the effective target thicknesses were determined to be 360 and 500 $\mu g/\text{cm}^2$ for Zn and Pb, respectively.

During the measurements, the counting rate of the x-ray detection system was normally kept below 100 cps to avoid a piling-up effect. Examples of the K x-ray spectra of Zn and the L x-ray spectra of Pb produced by proton impact are shown in Fig. 1. The spectra of Pb L x rays produced by helium-ion impact are very similar to that shown in Fig. 1(b). No significant difference between the spectra obtained by proton impact and those obtained by helium-ion impact was found. As is seen in Fig. 1(a), the $K\alpha$ and $K\beta$ peaks in the Zn x-ray spectrum are clearly resolved. Although five peaks ($L\alpha$, $L\beta$, $L\gamma$, Ll, and $L\eta$) are seen in the Pb spectra, $L\alpha$, $L\beta$, and $L\gamma$ are not single transitions, but comprise many x-ray lines which are not resolved owing to the limited energy resolution of the x-ray detector. The energy loss in passing through the target, calculated from the table of Northcliffe and Schilling,[15] is always less than 40 keV for protons, but is considerably larger for ^{3}He(160 keV for 1.4-MeV ^{3}He).

The errors in the present measurements are estimated to be about 15%, including those in the x-ray detection efficiency (10%), the x-ray absorption (5%), the solid angle (3%) and the target thickness (10%). The statistical errors are less than 2% for the Zn K, Pb $L\alpha$, and $L\beta$ cross sections, but varied from 5 to 25% for the weaker Pb L cross sections, since they are determined from weaker peaks.

III. RESULTS AND DISCUSSION

A. Zn K-shell ionization

1. *Total K-shell ionization cross section*

The total K-shell x-ray production cross section σ_x^K is given experimentally as the sum of the x-ray production cross sections for the $K\alpha$ and $K\beta$ transitions. The ionization cross sections σ_I^K are derived from the σ_x^K using the "most-probable" fluo-

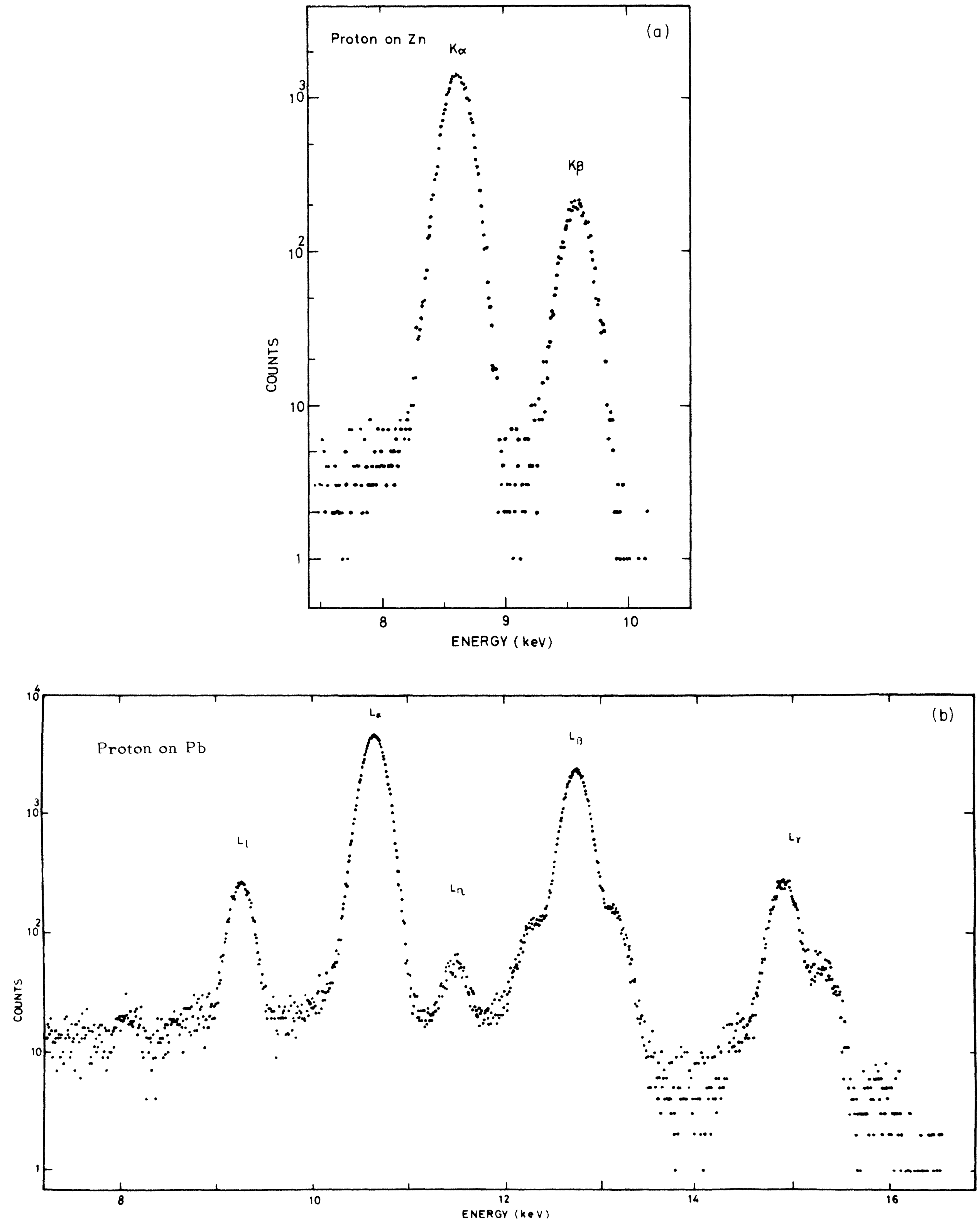

FIG. 1. Typical spectra of (a) Zn *K* and (b) Pb *L* x rays produced by proton impact, measured with a Si(Li) detector.

TABLE I. Fluorescence yields and Coster-Kronig yields used in the data analysis.

	Fluorescence yield		Coster-Kronig yield	
Zn	ω_K	0.479		
Pb	ω_{L1}	0.07	f_{12}	0.17
	ω_{L2}	0.363	f_{23}	0.156
	ω_{L3}	0.315	f_{13}	0.61

rescence yield given by Bambynek *et al.*[7] (see Table I). In Fig. 2, our experimental ionization cross sections are compared with theoretical calculations based on the BEA and the PWBA, and with the experimental results of Fahlenius and Jauho.[16] The low-energy data in the present experiment were obtained with a H_2^+ beam and plotted at equivalent proton energies. Although it is reported in some cases[17] that there is a difference between the ionization cross section for proton impact and that for H_2^+ impact at lower energies, no significant difference was observed in the present study. The data of Fahlenius and Jauho shown in

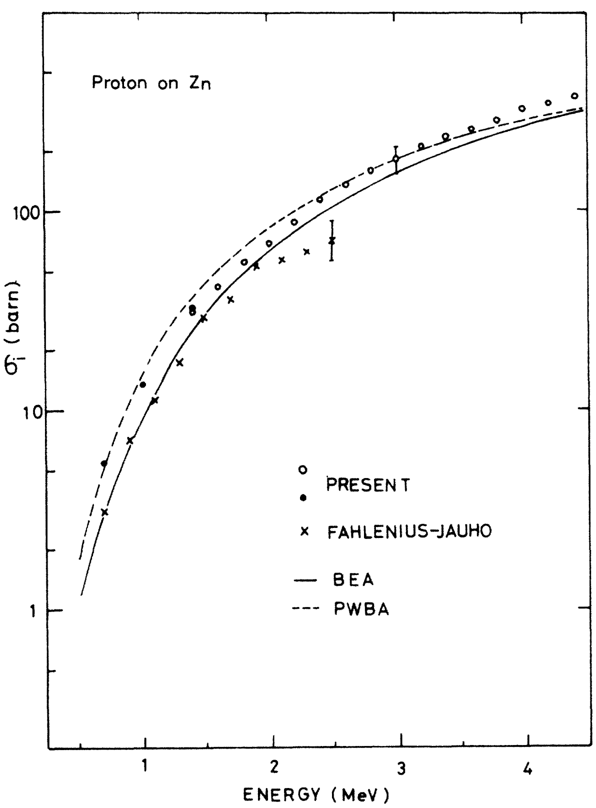

FIG. 2. K-shell ionization cross sections of Zn produced by proton impact. The results of Fahlenius and Jauho have been replotted using the most probable value of the fluorescence yield. The solid and dashed lines represent calculations based on the BEA and the PWBA.

Fig. 2 were plotted using the "most-probable" ω_K, which is different from the ω_K in their published results. The present data agree well with theoretical calculations, whereas the data of Fahlenius and Jauho tend to level off above 2 MeV.

2. $K\alpha/K\beta$ ratio

The $K\alpha$ and $K\beta$ lines are well separated in the present experiment as is shown in Fig. 1. From these spectra, a value of 7.04 for the ratio $K\alpha/K\beta$ was obtained after correction for the self-absorption in the target, for the absorption in the Mylar window, the aluminium absorber, and the air, and for the x-ray detection efficiency. The ratio $K\alpha/K\beta$ is constant to within the experimental errors and shows no energy dependence over the energy range investigated. A comparison of the ratio $K\alpha/K\beta$ obtained from various experiments[18,19] and from the theoretical calculation of Scofield[20] is given in Table II. The present value is reasonably close to that of Slivinsky and Ebert,[18] who used bremsstrahlung radiation as the excitation source, and that of Mistry and Quarles,[19] who employed electron bombardment. The latter value is close to that listed by Bambynek *et al.*[7] and Nelson *et al.*[21] However, the theoretical value of Scofield is still higher (10–20)% than the experimental results.

Richard *et al.*[22] have reported a reduction of the ratio $K\alpha/K\beta$ for Cu produced by oxygen-ion impact, as compared with that produced by proton impact, as a result of simultaneous outershell ionization. But this effect is expected to be generally small in the case of proton impact; that is, there should be little difference in the ratio $K\alpha/K\beta$ produced by photon, electron, or proton impact.

B. L-shell ionization of Pb

1. Total L-shell x-ray production cross section

Since the detector used in this experiment is not capable of resolving many x-ray lines resulting from transitions between various subshells in the $L\alpha$, $L\beta$, and $L\gamma$ groups, the individual subshell ionization cross sections could not be determined accurately. However, it is possible to calculate the

TABLE II. Comparison of the $K\alpha/K\beta$ ratio of Zn for various experiments and theory.

	$K\alpha/K\beta$	Excitation
Present	7.04	Proton
Slivinsky-Ebert	6.59	Bremsstrahlung
Mistry-Quarles	7.52	Electron
Bambynek *et al.*	7.41	"Most probable"
Nelson *et al.*	7.30	"Most probable"
Scofield	8.06	Theory

total x-ray production cross section using the fluorescence yields (ω_1, ω_2, ω_3), Coster-Kronig yields (f_{12}, f_{13}, f_{23}), and theoretical subshell ionization cross sections ($\sigma_i^{L1}, \sigma_i^{L2}, \sigma_i^{L3}$). The x-ray production cross sections for the L_1, L_2, and L_3 subshells are as follows:

$$L_1 \text{ shell: } \sigma_x^{L1} = \omega_1 \sigma_i^{L1},$$
$$L_2 \text{ shell: } \sigma_x^{L2} = \omega_2(\sigma_i^{L2} + f_{12}\sigma_i^{L1}),$$
$$L_3 \text{ shell: } \sigma_x^{L3} = \omega_3[\sigma_i^{L3} + f_{23}(\sigma_i^{L2} + f_{12}\sigma_i^{L1}) + f_{13}\sigma_i^{L1}]. \quad (2)$$

Thus, the total L-shell x-ray production cross section σ_x^L is given as the sum of the subshell x-ray production cross sections:

$$\sigma_x^L = \omega_1^{\text{eff}}\sigma_i^{L1} + \omega_2^{\text{eff}}\sigma_i^{L2} + \omega_3^{\text{eff}}\sigma^{L3}, \quad (3)$$

where

$$\omega_1^{\text{eff}} = \omega_1 + f_{12}\omega_2 + (f_{13} + f_{12}f_{23})\omega_3$$
$$\omega_2^{\text{eff}} = \omega_2 + f_{23}\omega_3 \quad (4)$$
$$\omega_3^{\text{eff}} = \omega_3.$$

The fluorescence yields and Coster-Kronig yields used are taken from the work of Bambynek *et al.*[7] and are given in Table I. The theoretical subshell ionization cross sections ($\sigma_i^{L1}, \sigma_i^{L2}, \sigma_i^{L3}$) can be calculated from the BEA or the PWBA.

On the other hand, the experimentally obtained total L-shell x-ray production cross section is as follows:

$$\sigma_x^L = \sigma_x^{L\alpha} + \sigma_x^{L\beta} + \sigma_x^{L\gamma} + \sigma_x^{Ll} + \sigma_x^{L\eta}. \quad (5)$$

Here $\sigma_x^{L\alpha}$, etc. are the measured x-ray production cross sections for the $L\alpha$, etc. transitions. Figure 3 gives a comparison of the experimental and calculated σ_x^L for proton impact. The agreement between the experimental results and the calculations based on the BEA are generally good. Also the data of Busch *et al.*,[13] which are in good agreement with the present results, have been plotted. Because the theoretical ionization cross sections for helium impact are less accurate, compared with those for proton impact, no attempt has been made to compare the calculated total L-shell x-ray production cross sections with the experimental results for helium impact.

2. Partial x-ray production cross sections for $L\alpha$, $L\beta$, $L\gamma$, Ll, and $L\eta$ transitions

Generally, in these experiments, measurements are made of x-rays of the $L\alpha$, $L\beta$, and $L\gamma$ groups with many unresolved peaks resulting from the various transitions between the subshells owing to the limited energy resolution of the x-ray detector. Therefore, it is convenient to compare experimental results of the x-ray production cross sections for these transitions with the calculated values. These partial x-ray production cross sections for the various groups can be calculated using the theoretical radiative transition rates for each case, as well as the florescence yields, Coster-Kronig yields, and theoretical ionization cross sections of the subshells.

Following a treatment very similar to the one given in Sec. III B 1, the following formulas are obtained for use in the calculation of these partial cross sections for the groups:

$$\sigma_x^{L\alpha} = [\sigma_i^{L1}(f_{13} + f_{12}f_{23}) + \sigma_i^{L2}f_{23} + \sigma_i^{L3}]\omega_3(\Gamma_{3\alpha}/\Gamma_3),$$
$$\sigma_x^{L\beta} = [\sigma_i^{L1}(f_{13} + f_{12}f_{23}) + \sigma_i^{L2}f_{23} + \sigma_i^{L3}]\omega_3(\Gamma_{3\beta}/\Gamma_3) + (\sigma_i^{L1}f_{12} + \sigma_i^{L2})\omega_2(\Gamma_{2\beta}/\Gamma_2) + \sigma_i^{L1}\omega_1(\Gamma_{1\beta}/\Gamma_1),$$
$$\sigma_x^{L\gamma} = (\sigma_i^{L1}f_{12} + \sigma_i^{L2})\omega_2(\Gamma_{2\gamma}/\Gamma_2) + \sigma_i^{L1}\omega_1(\Gamma_{1\gamma}/\Gamma_1),$$
$$\sigma_x^{Ll} = [\sigma_i^{L1}(f_{13} + f_{12}f_{23}) + \sigma_i^{L2}f_{23} + \sigma_i^{L3}]\omega_3(\Gamma_{3l}/\Gamma_3),$$
$$\sigma_x^{L\eta} = (\sigma_i^{L1}f_{12} + \sigma_i^{L2})\omega_2(\Gamma_{2\eta}/\Gamma_2). \quad (6)$$

Here, for example, Γ_3 is the theoretical total

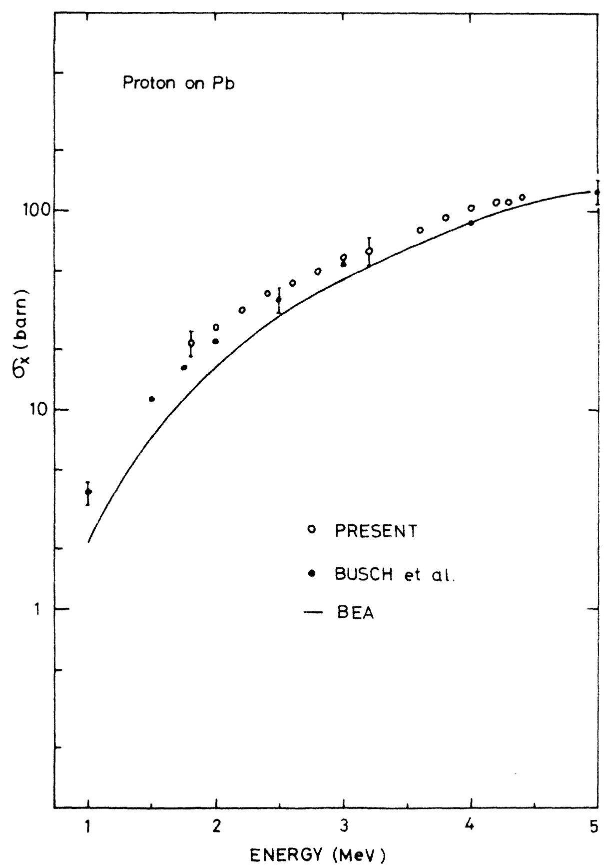

FIG. 3. Total Pb L-x-ray production cross sections produced by proton bombardment. The solid line represents a calculation based on the BEA (see text).

radiative transition rate of the L_3 shell, and $\Gamma_{3\alpha}$ is the sum of the radiative transition rates which contribute to the $L\alpha$ lines associated with hole filling in the L_3 shell: that is, $\Gamma_{3\alpha} = \Gamma_3(M_4 - L_3) + \Gamma_3(M_5 - L_3)$, where $\Gamma_3(M_4 - L_3)$ is the radiative transition rate from the M_4 shell to the L_3 shell. The radiative transition rates for many elements have been calculated by Scofield,[20] who applied the relativistic Hartree-Slater theory with a central potential and included the retardation effect.

In Fig. 4, the calculated σ_x for the groups are compared with the experimental values. The agreement between the experimental results and the BEA calculation is good at higher energies; however, the calculated values become systematically smaller than the experimental results at lower energies. However, for the reason mentioned above, no attempt has been made to compare the measured cross sections with the theory for helium impact. The present results for helium impact are shown in Fig. 5.

3. *X-ray production ratios for various groups*

As Shafroth *et al.*[12] have pointed out, the effect of Coulomb repulsion between the projectile and the target nucleus is canceled out, or at least reduced, in theoretical calculations of the x-ray production ratios (for example, $L\alpha/L\beta$). The effect of distortion of the wave function of the L-shell electron due to the incident particle is also reduced for calculations of these ratios. From an experimental point of view, the ratios eliminate many uncertainties, such as the inhomogeneity of the target thickness, and uncertainties in the geometry and in the ion-current measurement, and are only slightly affected by variations of the absorption of the window and the absorber and of the relative efficiency of the detector in the x-ray energy range studied. Therefore, the experimental values of these ratios provide a more rigorous test for the theoretical calculations.

These ratios are given in Fig. 6 as a function of

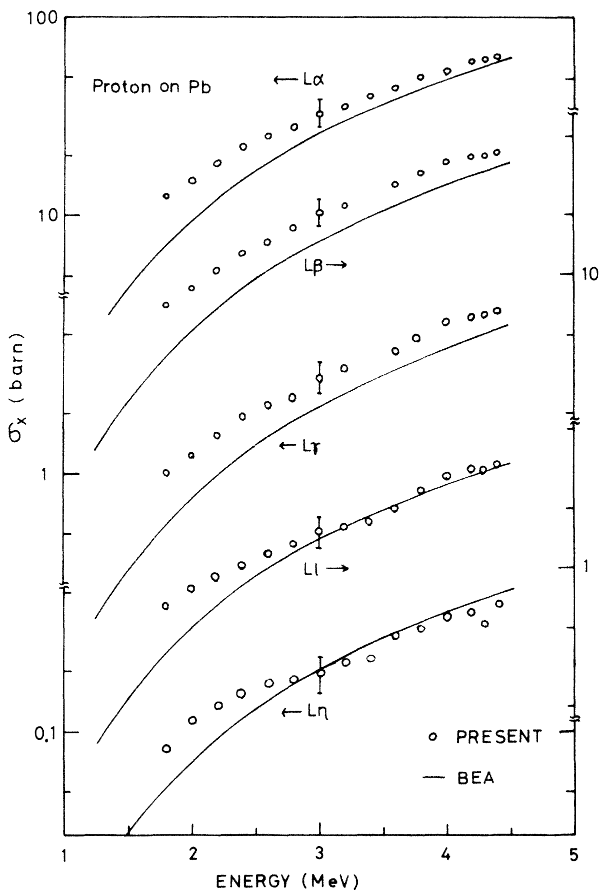

FIG. 4. Partial x-ray production cross sections of Pb $L\alpha$, $L\beta$, $L\gamma$, Ll, and $L\eta$ groups produced by proton impact. The solid lines represent a calculation based on the BEA (see text).

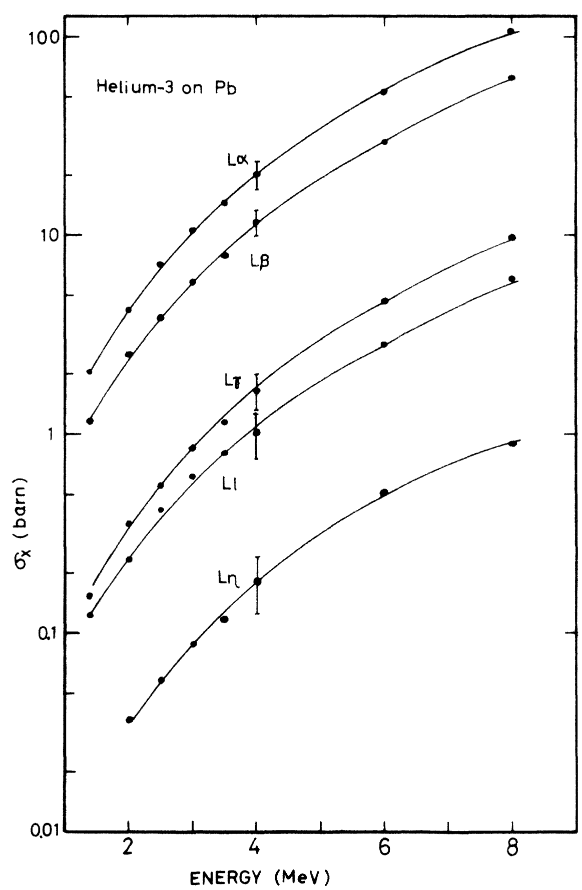

FIG. 5. Partial x-ray production cross sections of Pb $L\alpha$, $L\beta$, $L\gamma$, Ll, and $L\eta$ groups produced by ^{3}He bombardment. The solid lines are drawn to guide the eye.

proton energy. In this energy range, the measured ratios $L\alpha/L\beta$ and $L\alpha/L\gamma$ agree well with the calculated values based on the BEA at low energies, but deviate from them at higher energies. For the ratio $L\alpha/Ll$, the BEA gives a constant value of 18.8, because the initial vacancy is produced in the L_3 shell for both the $L\alpha$ and Ll transitions; however, the measured ratios vary with proton energy. (The probable value listed by Salem and Schultz[23] is 22.6 for Pb.) This characteristic was also found by Busch *et al.*[13] They attribute this to the difference in simultaneous outershell ionization in the $L\alpha$ and Ll transitions, which is neglected in the theory. Measured values of the ratio $L\alpha/L\eta$ are rather scattered, but are systematically larger than the results of the BEA calculation.

4. Z_1 dependence of Pb L-x-ray production

Both the BEA and the PWBA theories predict that the innershell ionization cross section is proportional to the square of the projectile nuclear charge (Z_1). This Z_1 dependence of the innershell ionization cross section has been experimentally investigated by Basbas *et al.*[24] and Lewis *et al.*[25] Both groups measured the ratio of K-x-ray production for helium impact to that for proton or deuteron impact, and their results deviate significantly from the theoretical value. The measured ratio is smaller than the calculated value at lower energies, rises above it at intermediate energies, and then converges toward it at higher energies. The so-called "crossover" behavior can be explained as follows: At low energies the ionization cross section decreases owing to Coulomb repulsion of the projectile particle and also owing to an increase in the effective nuclear charge as experienced by the K electron from Z_2 to Z_2+Z_1, where Z_2 and Z_1 are the target and projectile nuclear charge, respectively. On the other hand, the distortion of the K-shell electron orbit by the incident particle increases the cross section at higher energies. Therefore, the crossover point is expected to occur at intermediate projectile energies.

However, no experimental investigation has yet been reported on the Z_1 dependence of L-shell ionization, except for that of Ar L-shell ionization.[26] It is expected that the situation for L-shell ionization is more complicated than that for K-shell ionization. Applying the treatment used by Basbas *et al.*[24] for K-shell ionization, the crossover point for L-shell ionization is given as follows:

$$v_1/v_2 = 2U_L/RZ_L^2 , \qquad (7)$$

where v_1 and v_2 are the velocities of the projectile and the L-shell orbital electron of the target, re-

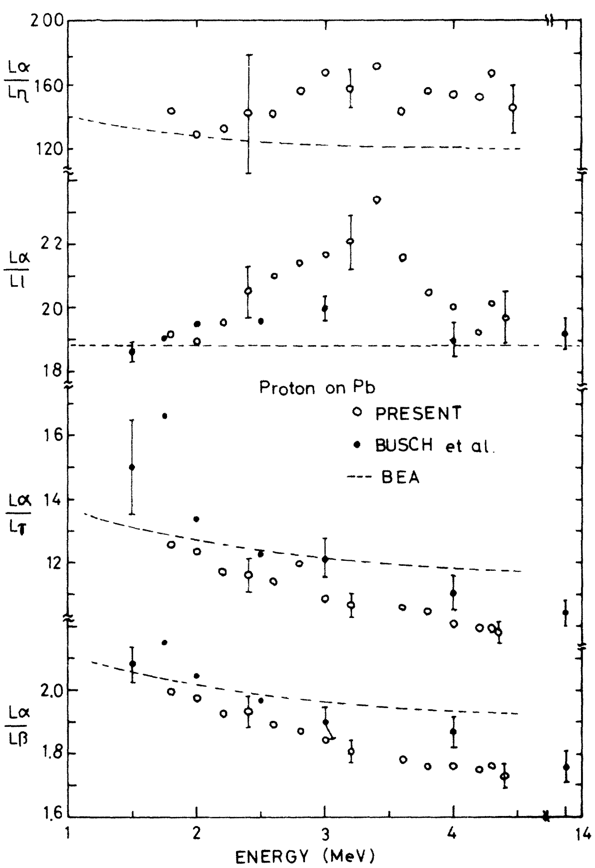

FIG. 6. Ratios of the $L\beta$, $L\gamma$, Ll, and $L\eta$ groups to the $L\alpha$ group for Pb produced by proton impact. The dashed lines represent a calculation based on the BEA (see text).

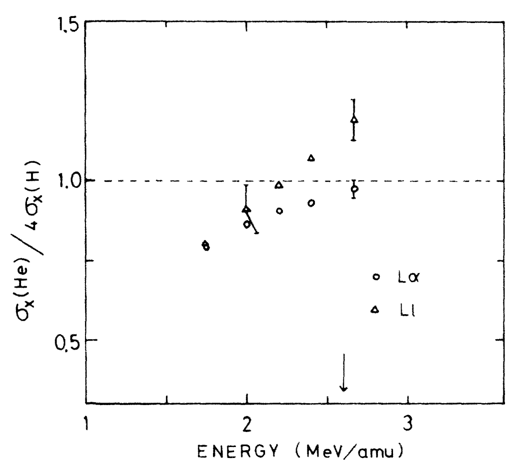

FIG. 7. Ratio of x-ray production of Pb L groups produced by proton and ^{3}He impact. The arrow shows the expected crossover point for the L_3 shell (see text).

spectively, U_L the binding energy of the L shell, R the Rydberg constant (13.6 eV), and Z_L the effective target nuclear charge ($Z_2 - 4.15$).

In Fig. 7 the measured value of the ratio $\sigma_x(\mathrm{He})/4\sigma_x(\mathrm{H})$ is given as a function of projectile energy per amu. As expected, at low energies the ratio is much smaller than unity but approaches unity between 2.5 and 3.0 MeV/amu, which is the region where the theories predict that the ratio should be unity. Although a wide range of projectile energies was not covered in the present experiment, it is expected that the ratio might be a little larger than unity at higher energies, and then decreases toward unity at even higher energies. In the present x-ray measurement, the individual subshell contributions could not be resolved. Therefore, it is difficult to choose a U_L for comparing the experimental and theoretical ratios. For example, all three subshells (L_1, L_2, and L_3 shells) contribute to the $L\beta$ and $L\gamma$ transitions. In the $L\alpha$ transition, for which the measurements are most accurate, however, only the L_3 shell is included as the initial vacancy. Therefore, we may compare the ratios of the $L\alpha$ lines obtained from the experiment and from calculations.

According to Eq. (7), the crossover point for Pb L_3-shell ionization is about 2.6 MeV/amu, which is rather close to the experimentally obtained value (see Fig. 7).[27] This means that Eq. (7), although based on general assumptions, gives a reasonable value of the crossover point even for L-shell ionization. It is hoped, however, that more systematic measurements of the Z_1 dependence of L-shell ionization will be made.

IV. CONCLUSIONS

The results of measurements of the Zn K and Pb L x-rays produced by proton impact over the energy range from 1.4 to 4.4 MeV were obtained. For Zn, the K-shell ionization cross sections are deduced from the measured x-ray production cross sections using the fluorescence yield. The results are in good agreement with calculations based on the BEA and PWBA. The ratio $K\alpha/K\beta$ is determined to be 7.04, independent of the incident energy. This result is close to the values obtained by other investigators employing x-ray and electron impact excitation, but is about 10% smaller than that calculated by Scofield.[20]

For the Pb L shell, instead of comparing the ionization cross sections, the total L and partial $L\alpha$, $L\beta$, $L\gamma$, Ll, and $L\eta$ x-ray production cross sections are compared, with those calculated by the BEA and with those measured by Busch *et al.*[13] The agreement is good to within the experimental uncertainties. The x-ray intensity ratios, $L\alpha/L\beta$, $L\alpha/L\gamma$, and $L\alpha/L\eta$, are close to the values calculated by the BEA at lower energies, but at higher energies a deviation occurs. In contrast to the calculations which give a constant value, the measured rations $L\alpha/Ll$ show a maximum in the vicinity of an impact energy of 3 MeV, which qualitatively agrees with the data of Busch *et al.*[13]

To test the Z_1 dependence of the L-shell ionization, the x-ray production was also measured for helium impact. At low energies, the measured ratios $\sigma_x(\mathrm{He})/4\sigma_x(\mathrm{H})$ for Pb L x-rays are smaller than unity, as is predicted by the BEA and PWBA theories, and become unity between 2.5 and 3 MeV/amu, which is in fairly good agreement with the calculated value, showing that the "crossover" behavior for L-shell ionization can be explained by the mechanisms suggested by Basbas *et al.*,[24] who successfully applied them to the K-shell ionization.

Finally, in contrast to the measurements of Au L x-ray production by Shafroth *et al.*,[12] no significant energy shifts of the Pb L x-ray groups were detected within the stability limit of our detection system.

Note added in proof. The revised calculation of L-shell ionization based on PWBA has been published by B. H. Choi, E. Merzbacher, and G. S. Kandelwal [At. Data 5, 291 (1973)]. The calculated values for Pb L-shell x-ray production are slightly higher than the experimental values over the energy range investigated in the present study. However, Professor W. Brandt kindly informed us that the calculated values with the binding and Coulomb deflection corrections fit nicely to the present values.

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