L- and M- shell ionization of various elements by fast-α-particle bombardment*

S. T. Thornton, R. H. McKnight, and R. R. Karlowicz
Department of Physics, University of Virginia, Charlottesville, Virginia 22901
(Received 19 September 1973)

L-shell total ionization cross sections have been measured for targets of Sn, Ag, Tb, Bi, and Au from bombardment of 1–5-MeV α particles. Similar M-shell x-ray cross sections have been obtained for Tb, Bi, and Au. The L-shell cross sections have been compared with theoretical calculations from the plane-wave Born approximation, the semiclassical approximation, and the binary-encounter approximation. The latter theory best reproduces the experimental data. A comparison of L-subshell transitions was also made between experiment and the three theories.

I. INTRODUCTION

In a continuing program to understand inner-shell excitation by fast heavy-ion bombardment, we have determined absolute ionization cross sections for L-shell vacancies resulting from 1–5-MeV α particles impinging on targets of Sn, Ag, Tb, Bi, and Au. In addition, we also have measured M-shell x-ray production cross sections over the same energy range for targets of Tb, Bi, and Au. There has recently been a great interest in measurements of this type owing in part to significant experimental advances in x-ray detection and heavy-ion acceleration and to various theoretical models used to understand the reaction mechanisms.

Except for much-lower- and higher-energy α-particle bombardment, almost no previous measurements have been published for L- and M-shell ionization by α particles.[1] There has been considerable work reported for proton bombardment.[1]

In the present measurement we have used a Si(Li) detector to detect x-rays resulting from L- and M-shell vacancies produced by the fast α particles as they pass through thin (and in some cases also thick) targets. The x-ray production cross sections are converted to total ionization cross sections for the L-shell by using tabulated fluorescence yields.

The justification for the present measurement is threefold. Firstly, the whole field of atomic ionization by fast heavy-ion bombardment is new and has produced many exciting and unexpected results. It is important to push the experimental data into unexplored regions. Secondly, the actual reaction mechanism is complex, and its study will require more data to fully understand its dependence on mass, charge, energy, etc. Universal ionization curves are actually based on a small amount of data, and the data are often inconsistent. Thirdly, there is a great need from applied scientists for these ionization cross sections. Technological applications for this type of experiment are manyfold in several areas of science.

II. EXPERIMENTAL PROCEDURE AND RESULTS

The x rays were detected with a Si(Li) detector located at 90° with respect to the α-particle direction. The nominal resolution was 175 eV [full width at half-maximum (FWHM)] for a 5.9-keV x ray. The α particles were accelerated by the University of Virginia 5.5-MV CN Van de Graaff accelerator.

The experimental procedure has been discussed in detail elsewhere.[2] Briefly, the thin targets were prepared by evaporating the material onto thin (20 $\mu g/cm^2$) carbon foils and were positioned at 45° with respect to the α-particle projection. At 43.6° a solid-state detector was placed to detect the elastically scattered α particles. This served to determine the absolute target thickness by comparing it with Coulomb scattering and also to check for target deterioration. Typical beam currents were 10 nA. Typical target thicknesses were 100–400 $\mu g/cm^2$. Thick-target measurements, in which the α particles are stopped in the target, were made for Au and Ag.

Careful attention was directed towards beam collimation, target deterioration, electronic losses, and beam charge collection and the data were corrected for all such factors. Self-absorption of the L-shell x rays in the thin targets was small, and corrections were applied for the thick-target data. The self-absorption for the lower-energy M-shell x rays was significant and was as large as 75% for the Au target. Uncertainties in the self-absorption corrections are large for the M-shell measurements, and the resulting uncertainties for the M-shell cross sections are estimated to be 60%. This large uncertainty and other uncertainties such as in the atomic fluorescence yield prevented us from quoting M-shell ionization cross sections. The L-shell x-ray cross sections were converted to ionization cross sections by using average atomic fluorescence yields from the tabulation by Bambynek *et al.*[3] The L-shell ionization cross sections are estimated to have uncertainties of 20%. The thick-target cross sections were determined by the method described by Bothe and Fränz.[4] The present thick-target measurements and the results of measurements involving heavier ions (neon) will be compared with similar thin-target measurements in a separate communication.

The cross sections are shown in Figs. 1–3. In Fig. 1 are shown L-shell ionization cross sections for Ag, Tb, and Au. The lines are theoretical calculations and will be discussed later. L-shell ionization cross sections for Sn and Bi are displayed in Fig. 2, and the M-shell x-ray production cross sections for Bi, Au, and Tb are shown in Fig. 3. For the Tb, Au, and Bi measurements the $L\alpha$, $L\beta$, and $L\gamma$ peaks were resolved and separate cross sections measured. The sum of these cross sections is presented in Figs. 1 and 2.

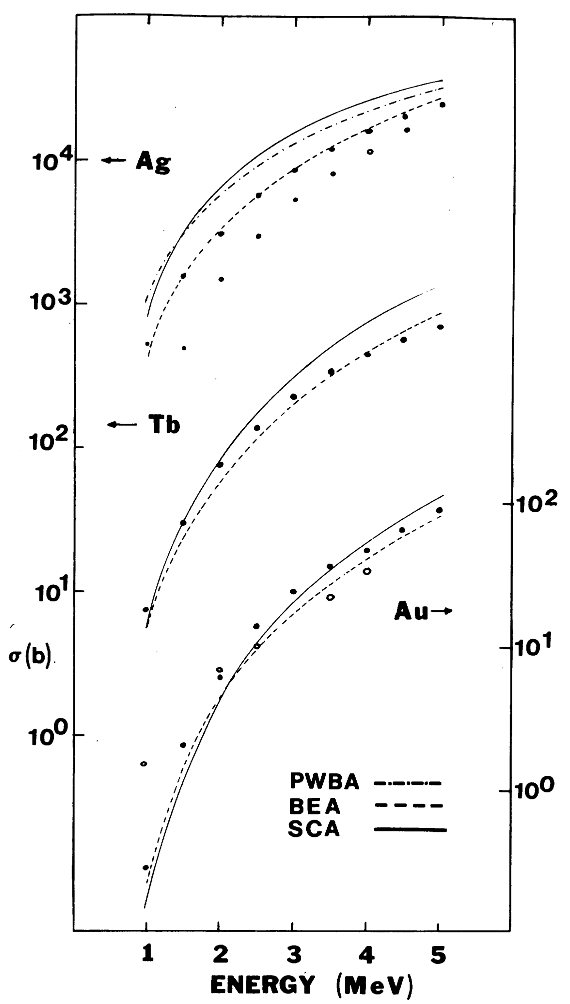

FIG. 1. Total L-shell ionization cross sections. Present results: thick target (○) and thin target (●).

III. DISCUSSION

Three theoretical calculations have been compared with the present data. These are the plane-wave Born approximation (PWBA),[5] semiclassical approximation (SCA),[6] and the binary-encounter approximation (BEA).[7] Each of these reaction theories will be discussed in turn.

A. Plane-wave Born approximation

The PWBA uses first-order perturbation theory to calculate the cross section for an inelastic collision between the heavy, charged projectile and the atomic electrons. A lucid review of the subject has been given by Merzbacher and Lewis.[5] Plane waves are used for the wave functions, and the distortion of the projectile wave function by the atomic electrons is neglected.

Merzbacher and Lewis list two conditions for the applicability of the PWBA. The first condition is that the velocity of the projectile be less than or comparable to the velocity of the orbital elec-

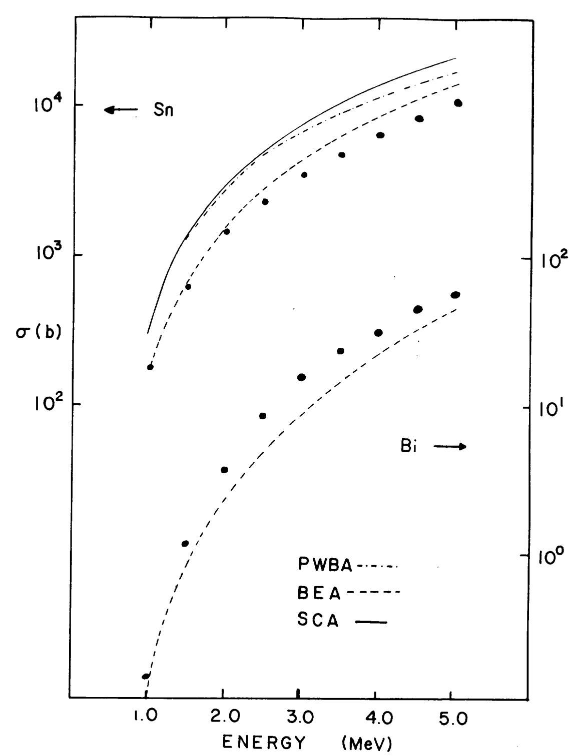

FIG. 2. Total L-shell ionization cross sections for Bi and Sn thin-target measurements.

tron. This can be expressed by the relation

$$(4m_e/m_1)E_1 \gtrsim U\,, \tag{1}$$

where m_e is the mass of the electron, m_1 and E_1 are the projectile mass and energy, respectively, and U is the binding energy of the electron. The second condition sets a lower limit to the projectile velocity,

$$Z_1e^2/hv_1 \ll 1, \tag{2}$$

where Z_1e and v_1 are the projectile charge and velocity, respectively.

Conditions (1) and (2) are obeyed for all the projectile energies and targets used in the present investigation. The worst case is for condition (2) for $E_\alpha = 1$ MeV, when the quantity in Eq. (2) is 0.6. Merzbacher and Lewis state that conditions (1) and (2) are satisfied for both proton and alpha projectiles between 100 keV and 5 MeV for targets with atomic numbers greater than 10. Thus it seems safe for the present investigation that the PWBA be applied. We have used the compilation of Khandelwal *et al.*[8] which assumes nonrelativistic effects. The results of the calculations are shown in Figs. 1 and 2.

B. Semiclassical approximation

The SCA has been proposed and discussed in a series of papers by Hansteen and co-workers.[6] The SCA is a semiclassical theory describing atomic Coulomb excitation by heavy, charged projectiles. It is based on a time-dependent perturbation treatment, and one of its most useful virtues is that the reaction is described in terms of impact parameters. Straight-line paths approximating the hyperbolic path are used to take account of Coulomb deflection. In this way adiabatic and diabatic collisions may be calculated. Bang and Hansteen[6] show that at high energies the SCA and PWBA theories produce equivalent ionization cross sections.

There are three conditions for the use of the SCA. The first is that the energy loss ΔE of the projectile be small,

$$\frac{\Delta E_1}{E_1} << 1\,. \tag{3}$$

The second condition is that

$$Z_2 >> Z_1\,, \tag{4}$$

where Z_2 is the charge of the target. The last condition is the most stringent,

$$\frac{2Z_1Z_2e^2}{hv_1} >> \frac{2\sin\frac{1}{2}\theta}{1+\sin\frac{1}{2}\theta}\,, \tag{5}$$

where θ is the projectile scattering angle. All three conditions are easily obeyed in the present experiment. The SCA is most useful in the adiabatic region where the projectile energies are too low for the PWBA to be applied.

We have obtained computer programs from Hansteen to calculate the L-shell ionization cross section. The SCA calculation is compared with the data in Figs. 1 and 2.

C. Binary-encounter approximation

The BEA assumes that the ionization is due to a direct energy exchange between the heavy, charged particle and an atomic electron. The basic theory has been discussed in various forms by several authors,[9] but Garcia[7] has applied it directly to interactions of the type considered in the present study. A cross section $d\sigma/d\Delta E$ for an energy exchange ΔE is first calculated. This cross section is then integrated over all possible energy exchanges from the electron binding energy up to the projectile energy. This result is then averaged over the bound-electron velocity distribution. Corrections are made due to the nuclear repulsion.

The primary condition for the applicability of the BEA is that the projectile energy be large enough. A reasonable criteria is that

$$B = \frac{E_1 m_p}{m_1 U} > 100\,, \tag{6}$$

where m_p is the mass of the proton, and the other symbols are as described earlier. The quantity B varies from 26 to 413 in the present study. The BEA will work best for the higher projectile energies on the low-Z targets. The worst case is

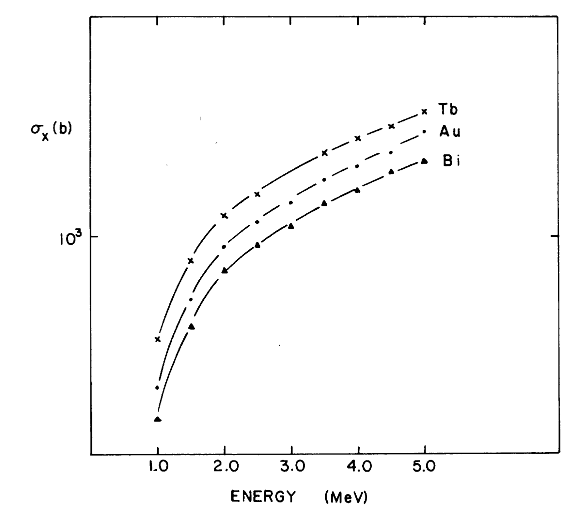

FIG. 3. M-shell x-ray production cross sections. Solid lines are drawn through the data points which are all from thin-target measurements.

for 1-MeV α's on Au. The comparison of the calculation with the data can be seen in Figs. 1 and 2.

D. Comparison of theoretical calculations

It is clear from Figs. 1 and 2 that the BEA provides the best agreement with the experimental results. The SCA and PWBA are consistently too high. We observed a similar result in our previous K-shell results.[2]

If multiple-ionization effects were important, our experimental results would be even lower. No multiple-ionization calculations were available to be applied to the present results. The SCA provides reasonable agreement for the heavier targets.

A universal curve[7] of $U^2\sigma_1/Z_1^2$ vs $m_pE/1000m_1U$ is shown in Fig. 4 for the L shell. The solid line is the BEA prediction. Good agreement is obtained for the L shell over a wide range of energies for the present measurements.

E. Subshell calculations

In order to understand differences between the theoretical results, we calculated cross sections for $L\alpha$, $L\beta$, and $L\gamma$ x rays by the method described in detail by Shafroth *et al.*[10] The radiative widths were obtained from Scofield[11] and the atomic fluorescence yields and Coster-Konig values were taken from the review of Bambynek *et al.*[3] The theoretical cross section σ_{L_i} for the ith subshell was obtained for the theories discussed previously.

The σ_{L_i} results for Ag are shown in Fig. 5. This result is typical for the other targets. The SCA σ_{L_1} cross section is higher than the BEA and PWBA at higher α energies and slightly lower for σ_{L_2} and σ_{L_3}. At the lower energies the three theoretical results are similar.

The ratios $\sigma_{L\alpha}/\sigma_{L\beta}$ and $\sigma_{L\alpha}/\sigma_{L\gamma}$ for the measurements are compared with the theoretical predictions in Fig. 6 for Au. The SCA is consistently high and does not follow the trend of the data. The PWBA curve is scaled from the proton on the Au re-

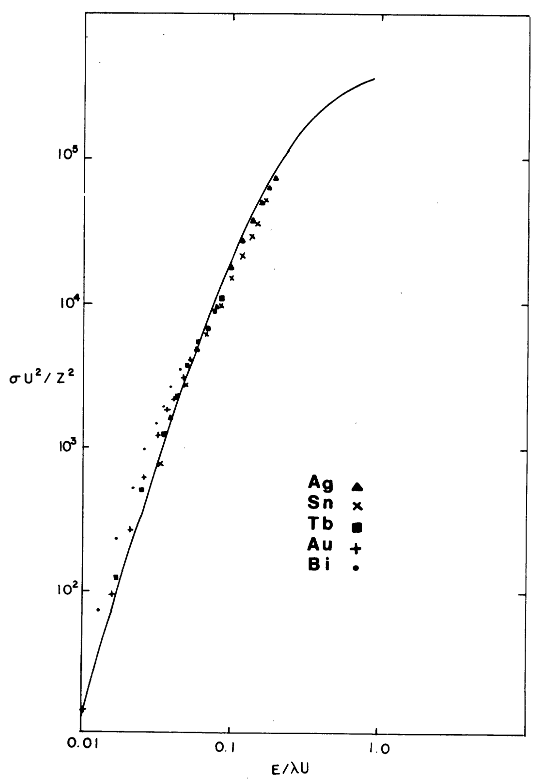

FIG. 4. Universal L-shell ionization curve summarizing present results. The solid line is a BEA calculation. The scaled cross-section units are 10^{-24} keV^2/cm^2. Other parameters are defined in the text (Sec. III C).

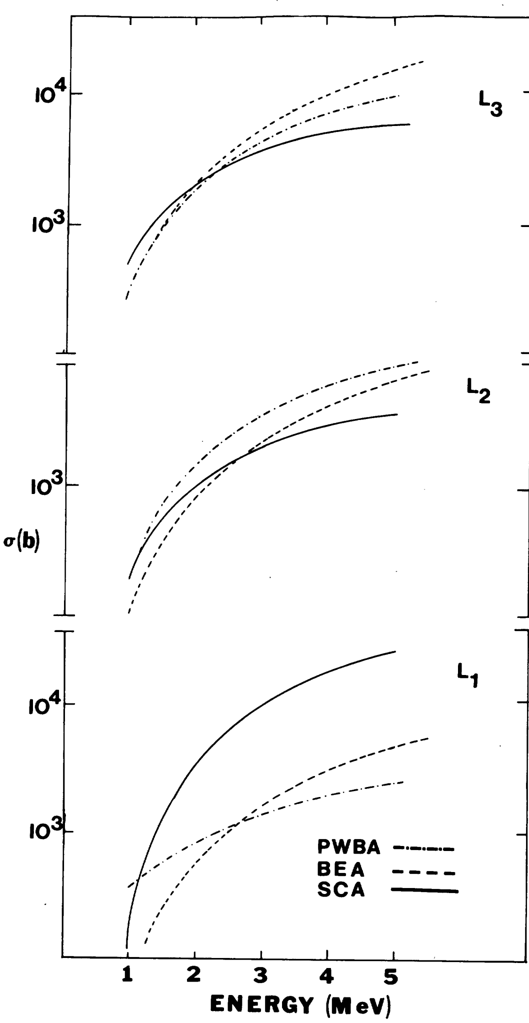

FIG. 5. Theoretical calculations for the ionization of individual subshells of Ag using the PWBA, SCA, and BEA formalism.

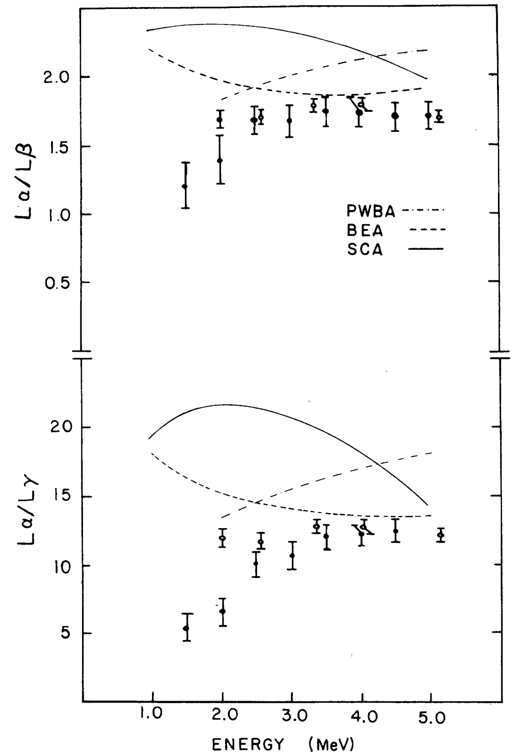

FIG. 6. $L\alpha/L\beta$ and $L\alpha/L\gamma$ ratios for Au. Present results are indicated by solid dots while open circles are data taken from Ref. 11 and are scaled to equal velocity. Calculations are discussed in the text.

sults of Shafroth *et al.*[10] and, although high, these results do follow the experimental trend. The *p*-Au data of Shafroth *et al.* were scaled for equal α-particle velocities and there is reasonable agreement between these data and our α-particle results. The present data extend to lower velocities into the adiabatic region where one would not expect the BEA to be valid and where it indeed does diverge. It is disappointing that the SCA does not provide a better description at low energies. The $\sigma_{L\alpha}$ cross section depends primarily on σ_{L_1} which is in disagreement badly with the BEA and PWBA. Hansen[12] has also noted this behavior for the SCA.

F. Thick- and thin-target comparison

Thick-target measurements were also taken for some of the present targets, and the results are compared with the thin-target data in Fig. 1. There can be great uncertainty in thick-target measurements because in order to convert to cross sections at the incident bombarding energy one needs to accurately know x-ray absorption cross sections and dE/dx incident particle energy losses in the target.

We consider the agreement shown in Fig. 1 well within the uncertainties presently obtainable in measuring thick-target yields. The details of the thick-target measurement and cross-section determination will be discussed elsewhere,[13] but the agreement between the thick and thin targets is gratifying.

IV. CONCLUSIONS

L-shell ionization and M-shell x-ray cross sections have been measured for 1–5-MeV α particles onto several targets. A comparison between the SCA, BEA, and PWBA calculations indicates that the BEA provides the best agreement with the experimental results. An investigation of the subshell cross sections indicates some possible problem with the σ_{L_1} prediction of the SCA.

Considering the limitations of the three theories, the BEA worked surprisingly well and the SCA results were disappointing, since many of the data were taken in the adiabatic energy region.

A comparison of thick- and thin-target cross sections indicated good agreement and provided support for the accuracy of the measurements.

ACKNOWLEDGMENTS

The authors acknowledge computer support from the University of Virginia Division of Academic Computing. We also thank Professor R. C. Ritter for assistance in early stages of this work. We wish to thank Professor J. M. Hansteen for providing a copy of his SCA program and Professor J. D. Garcia for comments concerning the BEA calculations.

---

*Supported in part by the University of Virginia Center for Advanced Studies (NSF) and the University Research Policy Council.

[1]J. D. Garcia, R. J. Fortner, and T. M. Kavanagh, Rev. Mod. Phys. 45, 111 (1973).
[2]R. H. McKnight, S. T. Thornton, and R. R. Karlowicz, Phys. Rev. A 9, 267 (1974).
[3]W. Bambynek, B. Craseman, R. W. Fink, H.-U. Freund, H. Mark, C. D. Swift, R. E. Price, and P. Venugopala Rao, Rev. Mod. Phys. 44, 716 (1972).
[4]W. Bothe and H. Fränz, Z. Phys. 52, 466 (1929); H. W. Lewis, B. E. Simmons, and E. Merzbacher, Phys. Rev. 91, 943 (1953).
[5]E. Merzbacher and H. W. Lewis, Handb. Phys. 34, 166 (1958).
[6]J. Bang and J. M. Hansteen, K. Dan. Vidensk. Selsk. Mat.-Fys. Medd. 31, No.13 (1959); J. M. Hansteen and O. P. Mosebekk, Phys. Lett. 29B, 281 (1969); J. M.

Hansteen and O. P. Mosebekk, Z. Phys. 234, 281 (1970).
[7]J. D. Garcia, Phys. Rev. A 1, 280 (1970); 1, 1402 (1970); 4, 955 (1971).
[8]G. S. Khandelwal, B. H. Choi, and E. Merzbacher, At. Data 1, 103 (1969).
[9]M. Gryzinski, Phys. Rev. 138, A305 (1965); 138, A322 (1965); 138, A336 (1965); E. Gerjuoy, Phys. Rev. 148, 54 (1966); J. D. Garcia, E. Gerjuoy, and Jean E. Welker, Phys. Rev. 165, 66 (1968).
[10]S. M. Shafroth, G. A. Bissinger, and A. W. Waltner, Phys. Rev. A 7, 566 (1973).
[11]J. H. Scofield, Phys. Rev. 179, 9 (1969).
[12]J. S. Hansen, Phys. Rev. A 8, 822 (1973).
[13]R. H. McKnight and S. T. Thornton (unpublished).