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L-SUBSHELL IONIZATION BY PROTON AND
ALPHA-PARTICLE BOMBARDMENT OF Ta, Au, AND Bi

DISSERTATION

Presented in Partial Fulfillment of the Requirements for
the Degree Doctor of Philosophy in the Graduate School of
The Ohio State University

By

Chu-Nan Chang, B.S., M.S.

* * * * *

The Ohio State University

1974

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2. "L-Subshell Ionization By Proton and Alpha Particle Bombardment of Ta, Au, and Bi" C.N. Chang, J.F. Morgan, and S.L. Blatt, Bull. Am. Phys. Soc. 19, 568 (1974).
3. "Apparent Dependence of α -Particle-Induced L Subshell Ionization on Outer Electron Configurations" C.N. Chang, J.F. Morgan, and S.L. Blatt, submitted for publication (1974)

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CHAPTER I

INTRODUCTION

When a heavy charged particles passes through matter, it interacts with the constituent atoms of the matter. One consequence of this interaction is an energy loss by the charged particle. If the charged particle penetrates an atom deeply enough to be able to transfer energy to the inner-shell electrons of the atom, the electron is either removed to an outer unoccupied shell or to a continuum state of that atom. Characteristic X-rays, as well as Auger electrons, are then emitted. This charged particle-induced inner shell ionization phenomenon has been of interest for a long time for its fundamental importance, since the study of the inner-shell ionization provides a way of studying the behavior of the inner-shell electrons of an atom. It also provides a simple test of the mechanism of the energy loss of the charged particle which traverses matter. In addition to these, the inner shell ionization study itself has its own applications. The charged-particle-induced characteristic X-rays have recently been applied to the field of the trace element analysis.

Experimentally, one can measure the inner-shell ionization probability from the study of the emitted characteristic X-rays or Auger electrons. The selection rules for dipole radiation severely restrict the pairs of states which can be observed in X-ray emission studies. This makes X-ray spectra easier to interpret than Auger spectra. The inner-

shell ionization study in this work was confined to the study of the emission of characteristic X-rays.

If there is an electron vacancy created in the K shell, the vacancy can be filled by outer-shell electrons, such as L shell, M shell, etc., to give rise to K-series X-rays (K X-rays). The L-series X-rays (L X-rays) are emitted when an outer-shell electron drops to fill the L-shell vacancy. Following these, we have M-series X-rays, N-series X-rays, etc. The K series is the simplest one, since the K shell is a single shell. The L series is more complicated than the K series, since it has three subshells; the M series is even more complicated than L series; and so on. The inner shell ionization mechanisms would be easier to study for K shell ionization, via K-series X-rays. As a matter of fact, such studies have been done for the past 40 years. However, the L X-rays provide more information and finer checks of the adequacy of the use of hydrogenic wave functions in all model calculations (see Section 1.B.). The reason that the hydrogenic wave functions are commonly used in the model calculation is that they are simple and easier to handle, and the fact that, in the first order approximation, they describe the inner shell electrons of an atom quite well. In order to reveal the inadequacy of this "hydrogenic wave function approximation," one has to study the individual L-subshell ionizations. This is the main motivation for the present dissertation.

The L-series X-rays are shown in Fig. 1. The transition rate and the transition energies indicated in this figure are for tantalum L X-rays only. The notation for the atomic levels and the terminology of the X-ray lines used here are the same for the other elements. The notation for the atomic levels have been commonly accepted; there is no commonly

Fig. 1 Decay scheme of L x-rays for tantalum. The radiative emission rate of each subshell with a single electron vacancy is also indicated.

Ta L X-rays

X-ray emission
rates in $\frac{\text{eV}}{\hbar}$
(from Scofield)

0.0104
0.0069
0.311
0.245
0.060
0.080
0.0089
0.0116
0.0286
1.065
0.0070
0.196
0.00124
0.0036
0.043
0.096
0.843
0.148
0.0029

accepted terminology of the X-ray lines. One may find that a different terminology is used by different authors. We follow the terminology appearing in X-rays in Theory and Experiment, by Allison and Compton (1935). We can see that these X-ray lines have energies which fall into a few narrow groups, designated on the figure by the Greek subscripts (Rf. Fig. 6). The $O_{II} \rightarrow L_I$ X-ray line is unnamed, even though it exists in principle, and the transition rate can be calculated. These complicated L X-rays make the subshell ionization cross section measurement difficult, since the recently developed high resolution Si(Li) detector can only separate the ℓ , α , β , and γ components. As shown in Fig. 1, each component, except the α component, has contributions from at least two subshells. The α component can be used to obtain, directly, the L_{III} subshell ionization cross sections. The L_{II} and L_I ionization cross sections can only be obtained by the use of computer programs to resolve the L_γ complex. The X-ray spectrum obtained by use of a Si(Li) detector is described in Chapter II of this dissertation. The computer analysis is described in Chapter III.

In order to get a picture of the inner shell ionization mechanism, we describe three model calculations developed so far, in Section 1.A. In Section 1.B. of this chapter, the main goal of the L-subshell ionization study mentioned above will be described in detail.

1.A. Charged-Particle-Induced Ionization Mechanism

The processes of inner-shell ionization of an atom, induced by the impact of a heavy charged particle, have been probed both in experiment and theory for a long time. However, with the increasing application

of experimental techniques new to this area, the development of theoretical models was continuously spurred. Early in the century, Bohr^{1,2} investigated the interaction of a fast incoming heavy charged particle with atomic electrons, and developed a model for the stopping power of materials in terms of an impulse approximation, which regards the collision as producing sudden transfer of energy and momentum to atomic electrons. The Bohr theory gave the general structure of these cross sections correctly, but certain dynamical details remained "phenomenological" (i.e., not calculable within the scope of the theory), owing to the lack of quantum-mechanical theory at that time. Nevertheless, as we shall see, his work forms a basic frame for the development of the modern binary-encounter approximation (BEA)^{3,4} to evaluate the inner-shell ionization cross section induced by fast heavy charged particles. In 1930, Bethe⁵ established a quantum-mechanical theory based on the Born approximation, and thereby derived a number of important results^{6,7,8} concerning collision cross sections and the stopping power for fast particles. The gross features of the production of inner shell vacancies in an atom when it collides with protons and alpha particles then were understood on the basis of the plane wave Born approximation (PWBA) calculation made by Henneberg⁹ in 1933. Merzbacher and Lewis¹⁰ have reviewed developments in this field up to 1958.

1. A-1. Plane Wave Born Approximation

In PWBA, it is assumed that the incident and inelastically scattered particles can be described by plane waves. The interaction responsible for vacancy production is the Coulomb interaction between the bound electron and the projectile. The initial and final states of the atom

are described in terms of a transition from the electron's initial bound state to a state described by a continuum wave function with the other electrons remaining in their initial states. This problem is treated with the aid of a perturbation calculation that takes the impact particle as a swift simple charged particle, moving too fast to capture electrons appreciably. This provides the perturbation that causes a transition between the unperturbed levels of the target atom. Thus, the perturbation technique is appropriate here only if the probability for excitation and ionization remain small, and this can only be the case if the charge of the incident projectile, $Z_1 e$, is small and the speed large enough. A criterion for the validity of this perturbation theory is that the scattered wave emitted in the collision between the particle with velocity v and the atomic electron have small amplitude, and this is assured if¹⁰

$$Z_1 e^2 / \hbar v \ll 1.$$

The expression for the cross section is conveniently written as a differential, in terms of the momentum transfer $\hbar q$, for ionization into a given final state Ψ_f , as

$$d\sigma_{fi}(q) = 16\pi Z_1^2 \left(\frac{e^2}{\hbar v}\right)^2 \frac{dq}{q^3} \left| \int \exp(i\vec{q} \cdot \vec{r}) \Psi_f^*(\vec{r}) \Psi_i(\vec{r}) d\vec{r} \right|^2 \quad (1.1)$$

where $Z_1 e$, is the projectile charge, v its velocity, and $\vec{r}$ the coordinate of electron relative to the atomic nucleus. The total cross section is obtained when the expression is integrated over all momentum transfers compatible with the production of a final state f and summed over all permissible final states, i.e., integrated over all allowable kinetic

energies and directions of motion of the ejected electron.

In Merzbacher and Lewis¹⁰, a further approximation is used. The initial and final state of the ionized electron are described by hydrogenic wave functions with an effective charge Z_s , to account for the inner screening¹¹. The dimensionless parameters, Θ_s , η_s , are introduced for convenience in scaling, and defined as

$$\Theta_s = \frac{n^2 u_s}{Z_s^2}, \quad \eta_s = \frac{1}{Z_s^2} \left(\frac{\hbar v}{e^2} \right)^2 = \frac{mE}{MZ_s^2 \text{Rydberg}} \quad (1.2)$$

where n is the principal quantum number of the s th Shell, u_s is the actual binding energy (in Rydbergs), m and M are the mass of electron and projectile respectively, and E is the incident energy of the projectile in the laboratory. We note that Θ_s is proportional to the ratio of the true binding energy to that predicted by a hydrogenic wave function ($E_s = \frac{Z_s^2}{n^2}$), and has a value between 0 and 1, to account for the outer screening; and η_s is proportional to the ratio of the incident energy to u_s . In terms of these parameters, the total cross section of the s th shell ionization is given by

$$\sigma_s = \frac{8\pi Z_1^2 a_0^2}{Z_s^4 \eta_s} f_s(\Theta_s, \eta_s), \quad (1.3)$$

where a_0 is the Bohr radius, and the general expression for f_s is

$$f_s = \int_{W_{\min}}^{W_{\max}} dW \int_{Q_{\min}}^{\infty} \frac{dQ}{Q^2} |F_{ws}(Q)|^2.$$

Here we introduce the dimensionless quantities W and Q by the relations

$$WZ_s^2(\text{Rydberg}) = \epsilon_s; \quad Q = (a_0/Z_s) q^2,$$

where ϵ_s is the energy transferred to the atom, which is equal to the sum of the actual binding energy, u_s , of the s shell, and the actual kinetic energy of the ejected electron (from s shell) in the continuum, T , i.e. $\epsilon_s = u_s + T$. The function $F_{ws}(Q)$ is the "form factor" written in terms of the dimensionless variable Q . The form factor is defined as the integral parts of Eq. (1.1). For the K and L shells, the expressions for F_{ws} are given as quadratures by Eqs. (4.12), (4.14) and (4.15) in Merzbacher and Lewis¹⁰.

For each of the L subshells⁴⁷

$$\begin{aligned} |F_{w,L_1}(Q)|^2 &= |F_{w,2s}(Q)|^2 \\ |F_{w,L_2}(Q)|^2 &= 1/3 |F_{w,2p}(Q)|^2 \\ |F_{w,L_3}(Q)|^2 &= 2/3 |F_{w,2p}(Q)|^2 \end{aligned}$$

where

$$\begin{aligned} |F_{w,2s}(Q)|^2 &= A(Q,k) [Q^5 - (8/3 + 11/3k^2)Q^4 \\ &+ (41/24 + 6k^2 + 14/3k^4)Q^3 + (5/48 - 31/24k^2 - 10/3k^4 - 2k^6)Q^2 \\ &+ (47/3840 - 41/120k^4 - 2/3k^6 + 1/3k^8)Q \\ &+ (1/768 + 17/768k^2 + 7/48k^4 + 11/24k^6 + 2/3k^8 + 1/3k^{10})] , \end{aligned} \quad (1.4)$$

$$\begin{aligned} |F_{w,2p}(Q)|^2 &= A(Q,k) [9/4Q^4 - (3/4 + 3k^2)Q^3 \\ &+ (19/32 - 3/4k^2 - 1/2k^4)Q^2 \\ &+ (107/960 + 41/48k^2 + 113/60k^4 + k^6)Q \\ &+ (11/3072 + 3/64k^2 + 7/32k^4 + 5/12k^6 + 1/4k^8)] , \end{aligned} \quad (1.5)$$

with $W = 1/4 + k$, k is the wave number of the hydrogenic continuum wave functions of the ejected electron; and

$$A(Q,k) = \frac{2^4 Q \exp\{-(2/k) \arctan(k/Q - k + 1/4)\}}{[1 - e^{-2\pi/k}] [(Q - k^2 + 1/4)^2 + k^2]^5}.$$

As discussed by Lin et al.¹³ and Bang and Hansteen¹⁴ recently, when the proton energy is below 2 MeV and the alpha energy below 5 MeV for light elements ($Z \leq 30$), the experimental cross sections for the K shell ionization turn out to deviate greatly from the values of the PWBA calculations. A higher cross section is predicted by PWBA in the lower impact energy region. Bang and Hansteen¹⁴ proposed that the deviation may be in part due to the fact that the PWBA does not take into account the Coulomb repulsion between the impinging particle and the nucleus. This repulsion prevents particles of low energy from getting close to the nucleus and may thus be expected to give rise to a cross sections smaller than predicted by the above mentioned theory. As a matter of fact, several recent experiments¹⁵⁻¹⁸ have demonstrated that the repulsion effect is significant in the low energy region.

1. A-2. The Semiclassical Approach

Bang and Hansteen use a semiclassical approach (SCA) that describes the atomic Coulomb excitation in impact parameter form.^{14,19}

In order to apply a classical treatment of the charged projectile which is causing the Coulomb ionization, it must be possible to describe the projectile motion by a wave packet small compared with the distance of closest approach to the target nucleus.²⁰ Together with the condition of negligible spread of the wave packet during the collision, the

necessary and sufficient condition for a classical treatment of the projectile in the ionization processes is found to be¹⁹

$$\frac{2Z_1 Z_2 e^2}{\hbar v} \gg \frac{2\sin^2 \frac{\theta}{2}}{1 + \sin^2 \frac{\theta}{2}} , \quad (1.6)$$

where $Z_1 e$ is the charge of the target nucleus, and θ is the deflection angle of the impinging charged particle in the center of mass system.

Perturbation theory is also applied in this semiclassical calculation. Thus, as required in PWBA, the potential due to the projectile - target electron interaction must be represented as a small perturbation. A necessary condition for this is that the charge of the projectile is much smaller than that of the target nucleus,

$$Z_1 \ll Z_2 .$$

The theoretical values can be tested by comparing to the experimental values as follows:

$$\frac{d\sigma}{d\Omega} = \frac{d\sigma_R}{d\Omega} I_p , \quad (1.7)$$

where $\frac{d\sigma_R}{d\Omega}$ is Rutherford differential cross section, I_p is the ionization probability as a function of the impact parameter, p ,

$$I_p = \int_0^\infty \left(\frac{d\sigma}{dT} \right)_p dT; \quad (1.8)$$

and

$$\left(\frac{d\sigma}{dT} \right)_p \sim \frac{1}{\hbar^2} \left| \int_{-\infty}^{\infty} e^{i\omega t} \langle f | V(t) | i \rangle dt \right|^2 ,$$

which denotes the ionization probability per energy interval dT of the ejected electron as a function of the impact parameter. The frequency ω is connected to the energy loss of the incoming particle, ϵ_s as

$$\omega = \frac{T + u_s}{\hbar} \quad (1.9)$$

After introducing the energy and mass of the projectile, one finds

$$\left(\frac{d\sigma}{dT}\right)_p = 4\pi Z_1^2 \frac{M}{E} \frac{e^4}{\hbar^2} |M_p|^2, \quad (1.10)$$

with

$$|M_p|^2 = \left| \int_{-\infty}^{\infty} e^{i\omega t} \psi_f^*(\vec{r}) V(t) \psi_i(\vec{r}) dt \right|^2,$$

where $\psi_f^*(\vec{r})$ and $\psi_i(\vec{r})$ are electron wave functions of the final and initial state of the target atomic electron, $V(t) = \frac{1}{|\vec{r} - \vec{R}(t)|}$, and $\vec{r}$ here is the position vector of the electron and $\vec{R}(t)$ that of the incoming particle (Fig. 2). The evaluation of the matrix element M_p leads to a very complicated analytical and numerical calculation^{14,19}, especially when a hyperbolic path of the incident particle is required. An approximation of taking into account the effect of the Coulomb deflection (which deflects the path of the incident particle into a hyperbolic shape) of the projectile was adopted in the following way in Ref. 19: For each value of the impact parameter, the distance of closest approach is substituted in the calculations. This means that the coordinate system used in the derivation of Eq. (1.10), is tilted such that the impinging projectile is moving tangentially to the classical hyperbola at the point of closest approach to the atomic nucleus.¹⁴

Fig. 2 The classical path of the projectile in the Coulomb repulsive field of a nucleus.

This model has been tested either by the total cross section measurements for K and L shell ionization (cf Ref. 15, 16, and 21), or by differential cross sections measurements which, in turn, obtain I_p as a function of the impact parameters (cf. Ref. 17 and 18).

The Coulomb deflection effect considered in the semi-classical calculation indeed reduces the deviation of theoretical values from the experimental values. However, the "adiabatic effect" (or "binding change effect"), considered by Brant et al.²², must also be taken into account when the energy of the incoming particle is so low that the time for it passing through the atom is longer than the period of the inner bound electron precessing around the atomic nucleus. In this case, the binding energy of the electron is adiabatically changed due to the presence of the incident charged particle. The binding effect depresses the cross section at lower energies, since it increases the effective charge seen by the electron at the target nucleus and therefore decreases its average radius. The result, again, improves the agreement with experiment.

1.A-3. The Binary Encounter Approximation

Another viewpoint of the inner shell ionization mechanism is, as noted earlier the "impulse approximation," developed early in this century by Bohr. The dominant interaction producing the transition is a direct energy exchange between the incident charged particle and the bound electron. The role of the nucleus is then simply that of establishing the momentum distribution for the electron. The collision is thus viewed as a collision of the incident particle, of momentum K_1 , with a free electron of momentum K_2 . The so-called "binary encounter approximation" (BEA)

arises from this step. The cross section is then summed over all momentum exchange ΔK compatible with an energy exchange ΔE . This result is integrated over all allowed energy exchanges, and finally weighed by the distribution of electron momenta associated with the bound state. Let v and V be respectively the velocity of the incident particle and of a bound electron; N_i be the number of electrons having equivalent binding energy u_s ; $f(V)$ be the speed (momentum) distribution of the bound electrons. Then the ionization cross section can be expressed as the following:

$$\sigma_I(v) = N_i \int_0^\infty \sigma_i(v, V) f(V) dV \quad (1.11)$$

where

$$\sigma_i(v, V) = \int_{u_s}^E \frac{d\sigma}{dE} dE = \int^{\Delta E} \frac{d\sigma}{dE} dE$$

If hydrogenic velocity distributions are used in Eq. (1.11) these BEA results for a given inner shell or subshell have been shown to obey a scaling law,^{4,23} which states that the product of the binding energy squared and the cross section is a universal function of the incident energy expressed in units of the binding energy, i.e.,

$$u_s^2 \sigma_I = Z_1^2 f(E/\lambda u_s, \lambda) \quad (1.12)$$

where λ is the mass (in electron mass units) of the incident projectile. Thus a plot of $u_s^2 \sigma_I / Z_1^2$ Vs $E/\lambda u_s$ should yield the same results for all target atoms. (The separate dependence on λ in Eq. (1.12) is very weak when $\lambda \equiv \frac{M}{m} \gg 1$, and f is thus very nearly independent of λ for the cases of interest in the present studies.)

The effect of nuclear repulsion is also taken into account by making an approximate correction of the impact parameter and the kinetic energy of the projectile because of its motion in the repulsive field. These two corrections are incorporated into the cross section by the expression⁴

$$\sigma_I(E) = \sigma_I(E') \left[\frac{1}{2} + \frac{1}{2} \left(1 - 2 \frac{Z'_2/Z_2}{\pi E/u_s} \right)^{1/2} \right]^2 ,$$

where $E' = E - \frac{Z'_2 e^2}{\xi}$, $\xi = r_A = \frac{Z_2 e^2}{2u_s}$ (the "radius" of the subshell whose binding energy is u_s) and Z'_2 is the effective point charge at the origin which produces the repulsion. Since $Z'_2 \approx Z_2$ for large Z_2 , $Z'_2 = Z_2$ can be used in calculations.

The comparison between theories and experimental data has been discussed in some detail, with extensive presentation of results in graphical form, in a review article by Garcia, Fortner and Kavanagh²⁴. These authors review the Coulomb excitation aspect of inner ionization of an atom by a point charged particle up to 1972 in the first two parts of that article. Most of the comparison data are K shell ionization cross sections with proton energies ranging from 15 keV to 160 MeV, and targets from $Z_2 = 4$ to $Z_2 = 92$. They display the data in terms of a "universal" curve, plotting $u_s^2 \sigma_I / Z_1^2$ vs. $E/\lambda u_s$ as described by the scaling law, Eq. (1.12). All the data except the 160 MeV point form a reasonably compact curve above $E/\lambda u_s \approx 10^{-1}$. Below $E/\lambda u_s \approx 10^{-1}$, the data for targets with $15 < Z_2 < 70$ continue to define a universal function, but the data for lower Z_2 elements begin to spread in the direction of smaller cross sections. The behavior of the main group of the data seems to be

closer to PWBA for $E/\lambda u_s \leq 3 \times 10^{-1}$, and to be closer to the BEA below that. The reason may be because BEA has made the correction for nuclear repulsion; as shown by Bang and Hansteen,⁴ the Coulomb deflection of the projectile by the nucleus has effects which are important only for $E/\lambda u_s \leq 10^{-1}$. The further deviations below $E/\lambda u_s \approx 10^{-1}$, especially for lighter targets, may be due to the binding effect, as mentioned before (cf. Ref. 22), which also provides the first indication of "compound atom" formation that plays an important role in heavier-ion (other than protons, deuterons, or alphas) -atom collision. The mechanism of heavier ion-atom collision is proposed as Pauli-excitation rather than the mechanism of Coulomb excitation of lighter ion-atom collision. Only Coulomb excitations are considered in this dissertation. The deviation of the 160 MeV data may be due to the relativistic velocity of the high-energy projectile. Some attempts have been made^{25,26} at a theoretical formulation of the effects of special relativity associated with the bound electron. It seems no such efforts are available for a relativistic heavy projectile.

1.B. L-Subshells

Among the three main simple models, PWBA, BEA and SCA, the Born approximation provides the best simple theoretical framework for describing the direct Coulomb inner shell ionization induced by a charged particle. This is true not only because of the agreement mentioned above but because it can in principle (and in practice) be improved. The BEA is a simple model and gives simple expressions and scaling laws, but

Fig. 3 Hydrogenic momentum distribution of 1s and 2s
electrons with an equal binding energy.

is less flexible as a general framework. However, modified BEA calculation of L-subshell ionization cross sections by using the momentum distribution of each L-subshell electron was recently performed by Hanen³⁴ (CBEA).

As the cross section measurements seem to be of such high precision and the theoretical understanding of the collision mechanism so far advanced, it has become feasible to study finer detail of the process and gain new physical insights. On the theoretical side, some effects have been made to work out ionization cross sections in a distorted wave Born approximation (DWBA), using Coulomb wave functions²⁷. The evaluation of the integral of Eq. (1.1), the form factor, with fairly sophisticated Hartree-Fock-Slater type wave functions has become practical²⁷ with the advent of fast computing techniques. The improvements that arise from the use of better atomic wave functions in inner shell ionization processes are obviously expected to be more pronounced for L shell ionization than for collisions with K-shell electrons, especially the ionization of the $2s_{1/2}$ subshell. Senashenko et al.²⁸ used a non-relativistic Born approximation with Coulomb wave functions for initial and final states of the electrons to evaluate the momentum distributions of the electron in the 2s state and in the 1s state at an equal binding energy. They showed that the momentum distribution of the 2s electron, $P^2 |\phi_{2s}(P)|^2$, has larger values than that of the 1s electron, $P^2 |\phi_{1s}(P)|^2$ at $P > 3P_0 \frac{Z_2}{n}$, and at $P \approx P_0 \frac{Z_2}{n}$, $P^2 |\phi_{2s}(P)|^2$ is smaller than $P^2 |\phi_{1s}(P)|^2$ (See Fig. 3) (Here, $P_0 = \mu e^2 / \hbar$, μ is the reduced mass

of the atomic system, i.e., $\mu \approx m$ (the electron mass) for a heavy element, and n is the principal quantum number of the electron bound state.) A node exists in the wave function for the 2s state in the vicinity of $p = p_0 \frac{Z_2}{n}$. All these differences between the 2s and 1s wave functions make strikingly different projectile energy dependences for the ionization cross sections of 2s and 1s states²⁸. Since the radial dependence of the 2p state is very similar to the 1s state (which has no node in its wave function both in coordinate representation and momentum representation except at the origin), the study of the ionization of 2p and 2s states would be very instructive and provide a fairly direct measure of some non-trivial features of the atomic states, including the extent to which hydrogenic wave functions are capable of describing them.

In this dissertation, the effort has been made to measure the L subshell ionization cross sections of elements ranging from $Z_2 = 73$ (Ta) to $Z_2 = 83$ (Bi) induced by proton and alpha impact in the energy range where the 2s differs from the 2p most noticeably, and to study the mechanism of the charged particle ionization processes by comparing them with model calculations.

Thin foils ($\leq 100 \mu\text{g}/\text{cm}^2$) of tantalum, gold and bismuth of greater than 99.8% purity were used as targets. A Si(Li) detector system with an overall resolution of 250 eV at 5.9 keV was employed to detect the product L X-rays. A Gaussian fitting routine was used to resolve L_γ X-rays and to obtain $2s_{1/2}$ and $2p_{1/2}$ subshell ionization cross sections. The L_α X-ray was used to deduce $2p_{3/2}$ subshell ionization cross sections. Additional measurements were later taken for alphas on platinum, to further explore effects observed in the other targets.

CHAPTER II

EXPERIMENTAL METHOD

The L X-ray production cross sections of Ta, Au and Bi, induced by protons and alpha-particles were measured. As the studies concern the L-subshell ionization, the main task is to obtain a high resolution L X-ray spectrum under various bombarding energies of the incident particle. In addition to this high resolution L X-ray spectrum, the beam charge collection, the target thickness, and the absolute efficiency calibration of the Si(Li) detector are also important in the determination of the absolute L X-ray production cross sections. We will describe them in detail in the separate sections of this chapter. The general apparatus will be described first.

The Ohio State University 5.5 MV Van de Graaff accelerator was employed to give a proton beam from 1.0 MeV to 5.5 MeV and an alpha beam from 1.0 to 11.0 MeV in this experiment. The beam was momentum analyzed by a 90° bending magnet and was then switched to the 45° -south beamline by a switching magnet. Doubly-ionized alphas were used in the energy range from 5.0 MeV to 11.0 MeV. In minimizing the possible ion-source contamination of H_2^+ after the use of protons, ${}^4He^+$ was used first for about 10 hours for the lower energy portion of this experiment. However, a cross check was made at 5.0 MeV with both singly-ionized alphas and doubly-ionized alphas to assure that there was, indeed, little

hydrogen contamination in the α beam. The ${}^4\text{He}^{++}$ beam was found to be pure in the sense that the L_{α} X-ray yield was the same as that for the ${}^4\text{He}^{+}$ bombardment, within experimental uncertainties.

The target chamber contains a beam collimator and a particle detector used as a beam monitor (Fig. 4). This particle detector was also used to measure the target thickness when the combined factor of the solid angle of the detector and its angle relative to the beamline was found by a target of known thickness. The target was oriented at 45° relative to the beamline so that the upward-looking Si(Li) detector can see the beam spot.

Both the beam collimator and the collimator of the particle detector were designed to install in the existing chamber properly. As shown in Fig. 4, the collimators contain two tantalum slits. In the beam collimator, the entrance diaphragm has a $1/16''$ aperture and the exit diaphragm is $3/32''$. The latter is $2.75''$ away from the target. They are $1.5''$ apart and insulated from the chamber. The beamline is equipped with additional slits at the entrance port right after the switching magnet and a quartz annular collimator with a $1/4''$ aperture located about one foot before the target spot. The quartz serves the dual role as a collimator and a viewer for any uncollimated beam. When it is used in conjunction with a HVEC magnetic quadrupole, a well-focused beam spot, of about $1/8''$ diameter, could be obtained at the position of the entrance collimator in the chamber. The beam spot thus obtained at the target is believed to be $1/16''$. Two suppressor rings are installed, on the ends of the beam collimator, to suppress the scattered electrons from it so that this system can be used when a thick target is bombarded and it is necessary to use the chamber itself as a charge collector of the beam.

Fig. 4 Experimental apparatus around the chamber.

This also eliminates the secondary electron bombardment of the target, even though the influence of it to the cross section measurement is expected to be negligible.²⁹ The chamber is insulated by two delrin adaptors from the beamline and the Faraday cup. The electron suppression is accomplished by connecting the suppressor rings to the negative terminal of a 90-volt battery and the positive terminal to the beam collimator and ground (Fig. 4).

As also shown in Fig. 4 the particle detector, collimated by a 1/16" tantalum slit about 1/8" in front of the gold surface of the detector, was installed at 45° relative to the beamline. The other 3/32" tantalum slit which is one inch before the first one and 2.75" from the target spot, was installed along with the first in a cylindrical stainless steel holder to limit the background from the elastic scattering of the particles from elsewhere in the chamber.

In order to let the low energy photons (≤ 100 keV) get out of the chamber, two 3/8" diameter, 1 mil thick mylar windows were installed. Both of them are at 90° relative to the beamline: one is vertical to it, and the other is horizontal to it. We use the vertical one in this experiment. The other one may be used when coincidence measurements are performed (see Appendix A).

As mentioned at the beginning of this chapter, the L X-ray spectrum, the target thickness, the beam charge and the absolute efficiency (including the solid angle of the X-ray detector) are the four main measurable quantities for determining the absolute L X-ray production cross sections. The following sections will describe the measurement of the quantities in detail.

2.A. The L X-Ray Spectrum

A KeveX Si(Li) (Model # 3000) detector with a field effect transistor (FET) preamplifier (KeveX Model # 2000) was used to detect the photon events. The ORTEC 440A selectable active filter amplifier and the ORTEC 438 baseline restorer were used for pulse shaping (signal filtering) and dc baseline restoration. By properly adjusting the electronics, a well-resolved L X-ray spectrum (330 eV at 9.71 keV, the Au L_{α} X-ray) can be obtained.

The output of the KeveX 2000 preamplifier has a decay time of 50 μ sec. which matches the variable pole-zero cancellation network built in the ORTEC 440A selectable active filter amplifier so that the pole-zero can be properly adjusted to eliminate undershoot of the output signals from this pulse-shaping amplifier. The baseline restorer, by restoring the undershoot in a.c.-coupled amplifiers, eliminates the low amplitude noises which frequently come from ever-present sources of 60 cycles and reduces pileup distortion caused by pulses falling on the tail (long-term component) of previous pulses. The shaping time of the amplifier, 4 μ sec, and the active mode of baseline restoration, which provides a very much higher restoration rate and therefore a very much higher count rate capability for the same amount of pileup distortion than those of passive mode, were chosen to obtain an optimum high resolution spectrum. The output pulses from the amplifier are unipolar, to take advantage of the high restoration rate of the active mode of baseline restorer. The arrangement of this portion of the electronics is shown in Fig. 5.

Fig. 5 **Electronics block diagram for detecting X-rays and monitoring the elastic scattering of the incident charged particles.**

In order to keep the count rate under 400 counts/sec. to minimize the pileup distortion and to be within the scale of the current integrator which can register the beam current as low as 1 nA (in this lowest scale, the accuracy of the instrument is about 10% due to the leakage current of the instrument itself), the solid angle subtended by the source (the bombarded target spot) to the 30 mm² Si(Li) wafer was decreased by placing the detector farther from the target spot along the detector axis (decreasing the overall detection efficiency of the detector). Several one inch aluminum cylindrical adaptors were stacked for this purpose and also made the distance between the target spot and the Si(Li) detector adjustable. In this experiment, beam currents were kept between 5 and 40 nA in order not to require any change in the detecting system (the distance between the beryllium window of the Si(Li) detector is set about 6-9/32" away from the target) in the whole energy range of bombardment (proton, 1.0 to 5.5 MeV and alpha particle, 1.0 MeV to 11.0 MeV). Under these conditions, the count rate was able to be kept quite low (from 40 to 400 counts/sec. due to the rapid changing of the ionization cross section versus impinging energy of the charged particle) to eliminate any possible pulse pileup problems. The unchanged distance between the target spot and the detector gives easier efficiency calibration of the detector with known-activity sources (see section 2.D) and fewer uncertainties result from this efficiency measurement. Typical L X-ray spectra of this spectrometer (with measured resolution 250 eV at 5.9 keV) of tantalum, gold and bismuth are shown in Fig. 6. They are produced by 4.5-MeV proton bombardment with beam current $\approx$ 10 nA, and count rate $\approx$ 300 count/sec. The L X-rays are resolved by this spectrometer into four components: L_{ℓ} , L_{α} , L_{β} , and L_{γ} , as indicated in the figures.

Fig. 6 L X-ray spectrum of tantalum, gold and bismuth with
4.5 MeV incident protons.

Substructure in L_{β} is noticeable, but not resolved, and L_{γ} is visually resolvable into $L_{\gamma_1} + L_{\gamma_5}$ and $L_{\gamma_2} + L_{\gamma_3} + L_{\gamma_4}$. The L_{γ} X-rays were further resolved by a Gaussian fitting routine, and the counts of $L_{\gamma_1} + L_{\gamma_5}$ and $L_{\gamma_2} + L_{\gamma_3} + L_{\gamma_4}$ were obtained (see Chapter III for details). Small amounts of tantalum L X-rays are also observed in the gold and bismuth runs, as shown in Fig. 6. This is not due to the target contaminations, but, rather, probably comes from the tantalum target frame, with its 5/32" center hole. A test shows that a target frame without a target on it also produces tantalum L X-rays (at most 1 count/sec). This source of tantalum L X-rays will not have any effect on the measurement of L_{γ} X-rays of Au and Bi, since their peak positions are outside those of the tantalum L X-ray group. The contribution of these Ta L X-rays to the other components of the Au and Bi X-ray spectrum are negligibly small because even the largest component of Ta L X-rays, L_{α} , is so small and lies outside the L X-ray group of Au and Bi, and the components of Au and Bi are not exactly on the positions of the rest of the components of the Ta spectrum. The tantalum target would not have this problem, since a larger center hole (3/8") was used for its frame. The M X-rays also appear in the gold and bismuth spectra.

Tantalum L X-rays were obtained with proton energy from 1.0 to 5.25 MeV, in 250 keV steps, and with alpha energy from 1.5 to 10.5 MeV, in 500 keV steps. Gold L X-rays were obtained with proton energies from 1.0 to 5.5 MeV, in 250 keV steps, and with alpha energies from 1.0 to 11.0 MeV, in 500 keV steps. With the same Au target that was used in the alpha bombardments, a check has been made for proton bombardment in the energy range from 3.125 MeV to 1.125 MeV, in 250 keV steps, and 1.00 MeV

and 3.00 MeV points also were repeated. This check ensured that our data were experimentally repeatable. The systematic errors due to the use of different targets were also shown to be negligible in this check. Bismuth L X-rays were obtained with proton energies from 1.0 to 5.5 MeV, in 250 KeV steps; and alpha energies from 1.0 to 5.0 MeV, in 0.5 MeV steps.

2.B. Target Thickness

All the targets are thin foils ($\leq 100 \mu\text{g}/\text{cm}^2$). The tantalum targets are supplied by MicroMatter Company. They were evaporated on $15 \mu\text{g}/\text{cm}^2$ carbon foil backing. The gold and bismuth targets were made in an evaporator in the Van de Graaff Laboratory of the Ohio State University, by evaporating the subject material (the purity of gold is 99.999% and that of bismuth is 99.8%) on a $10 \mu\text{g}/\text{cm}^2$ thin carbon foil slide. It is then removed by floating it in distilled water. Several trials have been made to get the thinnest one which can be picked up by the target frame (9/16" x 11/16" tantalum with a 5/32" diameter hole in the center) without breaking the target. Several spare targets for each element were also prepared. As a matter of fact, different gold and bismuth targets were used in the proton and alpha experiments because the previous one was accidentally broken during the change of the target. Only one tantalum target was used to cover the proton and alpha bombardments. The tantalum target thus serves as a good reference in measuring the target thickness of gold and bismuth. A beam monitor, an ORTEC surface barrier semiconductor particle detector with warranted alpha resolution 16 keV (the FWHM of a 5.5 MeV thin Am^{241} alpha source line) was also used to evaluate the target thicknesses of gold and bismuth. The elastically-scattered particle spectrum, run simultaneously with the L

X-ray spectrum, was routed to the second 1024-channel group of the ADC. The first 1024-channel group was used for the L X-ray spectrum. Because the beam was kept at 5 to 40 nA, the count rate of the particles was within 600 counts/sec and a pileup gate did not have to be put in during the experiment. The electronic blocks are shown in Fig. 5. The proton particle spectrum obtained in this way, with a proton energy of 4.5 MeV, is given in Fig. 7 for tantalum, gold and bismuth. Carbon and oxygen scattering peaks are also seen in these spectra.

The elastic scattering yields of the incident particle should follow a $1/E^2$ law, where E is the incident particle energy, if the geometry of the detecting system remains unchanged during a given kind of particle bombardment. A plot of the products of E_p^2 and the elastic peak counts Y , which are normalized to the average values of $E_p^2 Y$ versus E_p is given in Fig. 8 for Ta, Au, and Bi. E_p is the incident proton energy, in the laboratory system. About $\pm 3\%$ observed uncertainty could be due to the uncertainty of the beam collection mentioned in the beginning of this chapter.

Four different particle detectors, located at 45° , 90° , 130° , and 150° relative to the incident beam line in an ORTEC scattering chamber, were used to measure the target thicknesses of the tantalum, and the gold target which was used in the alpha experiment. The bombarding particles were 3.0 MeV protons. The incident beam intensity was ≈ 80 to 100 nA. A pileup gate was used in this measurement for pileup rejection. The electronic blocks are given in Fig. 9. The solid angles of these particle detectors were obtained primarily by the known diameter of each detector collimator and the distance between it and the target.

Fig. 7.A-7 C The nuclear elastic scattering spectrum of 4.5 MeV protons on tantalum, gold, and bismuth targets.

Fig. 8 The product of the Rutherford scattering yields, Y , and the square of the incident energy of protons, E_p^2 , as a function of the incident energy of protons for Ta, Au, and Bi. $E_p^2 Y$ were normalized to their arithmetic average as indicated in the figure.

Fig. 9 Electronics block diagram for target thickness measurements.

Then they were checked by a gold target with six particle detectors in the chamber at six different positions to obtain a consistency between them. The target thicknesses thus obtained are believed within 2%. All these measurements were performed with the target tilted 17° relative to the beamline so that the 90° particle detector can not be blocked by the target frame. (The measured target thicknesses have been corrected for this orientation.)

By using the known thicknesses of gold and tantalum, a combined factor of the solid angle and the position of the monitor particle detector can be obtained. The monitor particle detector can be used to obtain the unknown thicknesses of the gold and bismuth targets which were broken during the change of targets in the proton experiment, since the monitor functioned in all the bombarding energies of protons. It was also turned on in the alpha experiment on Ta and Au at five incident energies, which were also used to check the target thickness. The monitor is very important for the low-melting-point element bismuth (m.p. 271.3° C). However, the larger energy deposit on a thicker Bi target during the alpha bombardment makes the particle detector measurements useless after several low energy alpha bombardments (1.0 to 5.0 MeV in 1.0 MeV steps) because of the deformation of the target at that time. The experiment was carried out at this point by rechecking the L_α X-ray at previous bombarding energies. The results showed that there were twenty percent or more deviations. Because of the importance of the lower energy part, we then used a much thinner Bi target to do the α experiment again. The energy-deposition rate was lower, but in order to get a good statistics, a longer time was consumed and consequently the

Table 1. Target thickness* for Ta, Au, and Bi used in this experiment.

Target	Incident Particle:	Proton	Alpha
Ta		111.1 ± 3.3 a)	111.1 ± 3.3 a)
Au		98.2 ± 5.9 b)	101.4 ± 3.0 a)
Bi		49.4 ± 2.9 b)	23.9 ± 1.4 c)

* The target thicknesses given here are actual thicknesses seen by the incident beam. The unit is $\mu\text{gm}/\text{cm}^2$.

a) Measured in ORTEC scattering chamber.

b) Measured in "X-ray chamber" and used thickness of Ta as a reference.

c) Measured in "X-ray chamber" and used thickness of Au as a reference.

target was still somewhat deformed. However, this time the particle detector was able to function through all the bombarding energies from 5.0 MeV to 1.50 MeV and served as a target thickness monitor. The results can be believed to $\leq 10\%$. The thickness of various targets used in this experiment are summarized in Table 1.

2.C. Beam Charge Collection

The number of charged particles bombarding the target were measured through a current integrator which is digitized by a scaler. The total charge collected for various runs varied from 12 μC to 300 μC , due to changing ionization cross sections and the various target thicknesses, to obtain good statistics of L_γ X-rays for each bombardment. The statistical uncertainties of L_{γ_1} and $L_{\gamma_{2,3}}$ were 2% to 15%.

As indicated by the elastic-scattering beam monitor (see 2.A), a 3% uncertainty was observed in the various beam currents from 5 nA to 40 nA. This uncertainty is included in the quoted target thicknesses.

2.D. The Absolute Efficiency Calibration of the Si(Li) Detector

Methods for efficiency calibration of a Si(Li) detector have been discussed elsewhere in detail^{30,32}. Here the Si(Li) detector was calibrated by putting sources of known activities at the target position in the chamber and with the chamber remaining in the same vacuum condition and the detector at the same geometry setting as that of the beam-on experiment. ^{241}Am , ^{57}Co , and ^{137}Cs , which were calibrated by IAEA to within 1%, were used to obtain 8 points from 13.9 keV to 59.5 keV.

A 5.898 keV point was obtained by using a ^{55}Fe source which was calibrated within 6.5% by the New England Nuclear Company (NEN).

In order to interpolate the points between 5.898 keV where some of our photon energies fall, and the higher energy calibration points, we fit our data through a cubic exponential expression of $\ln E$:

$$\epsilon(E) = \exp [A(\ln E)^3 + B(\ln E)^2 + C(\ln E) + D] ,$$

under the basis that the analytical expression of the efficiency of the detector is a complicated exponential function of photon energy E ,³⁰

$$\epsilon(E) = \Omega [1 - e^{-\mu_p(E)\chi_d}] e^{-\sum_i \mu_i(E,Z)\chi_i} ,$$

where Ω is the detector geometry factor, μ_i is the absorption coefficient for each substance through which X-ray photons must penetrate from the source to the detector, χ_i is the thickness of each absorber; μ_p is the absorption coefficient for photoelectric absorption in silicon and χ_d is the thickness of the depletion depth of silicon. The possible absorbers in the X-ray path include the 1 mil mylar window of the chamber, the 1 mil beryllium window of the detector, $\approx 6''$ of air between the chamber and the detector, the dead layer of the silicon, and the gold surface layer. The expression used, empirically, is found to fit quite well. In figure 10, we show the fits for various data which correspond to the different distances of the source to the beryllium window of the detector (5.88 cm and 7.72 cm) as well as the one we just described (the 6.28 inches one). The almost parallel tails of the fittings beyond ≈ 20 keV reveal the insignificance of the air absorption for those photon

Fig. 10 Absolute efficiency curve of Si(Li) detector.

energies. We also include the data of Watson et al.²⁹ in Fig.10, which was obtained in another laboratory and from another Si(Li) detector. It also shows the parallel tail in high photon energies. The different shape of the fitting curve in the low energy region is probably because of the different absorbers in the photon path (the chamber window and the dead layer of the detector, etc.)

In our data analysis, we used the fitting curve to find our detector efficiencies for the different photon energies. We list the efficiencies which we used in this work in Table 2. We also normalized our Au L_{α} X-ray yields (in barns) to those of the Los Alamos Scientific Laboratory group⁴⁸ to obtain the efficiency of our detector at the photon energy of the Au L_{α} X-ray. The efficiencies for other energies were then obtained by correcting the air absorption of each individual photon energy relative to the air absorption of the Au L_{α} X-ray. The efficiencies so obtained are also listed in Table 2 for comparison. The higher value, at Bi L_{γ} energies, of this normalization might be due to the lower intrinsic efficiency of our Si(Li) detector at that photon energy. (In this normalization and correction for air absorption, we assume that the intrinsic efficiency of our Si(Li) detector is the same for all the photon energies ranging from 8 keV to 17 keV).

We do not have any low energy data ($\approx$ 5 keV) to show how good the cubic exponential fit curve is, however, above 5 keV, this fitting provided a quick and accurate method (within 10%) to do the extrapolation and interpolation of the data.

TABLE 2 . Si(Li) Detector Efficiency

Photon Lines	Photon Energy (keV)	Detector Efficiency From Corrections for Air Absorption ($\times 10^{-4}$)	Detector Efficiency From Cubic Exponential Fitting ($\times 10^{-4}$)
TaL γ	10.9	1.36	1.36
AuL γ	13.4	1.45	1.42
BiL γ	15.3	1.51	1.39
TaL α	8.15	1.14	1.10
AuL α	9.71	(1.34)*	1.28
BiL α	10.84	1.36	1.36

* Obtained from normalizing our Au L α X-rays yields to those of Bearse et al.⁴⁸

CHAPTER III

DATA ANALYSIS AND ERRORS

We present in this chapter the method concerning the deduction of L-subshell ionization cross sections from the measured production cross sections for each component of the L X-rays. The related atomic parameters are carefully chosen from existing and published values. The numerical results of this work are presented, and finally, various possible sources of errors are surveyed and explained.

3.A. Data Analysis

The production cross sections for each component of the L X-rays (Fig. 6) can be easily obtained by the equation

$$\sigma_{Ly}^x = \frac{Y_{Ly}}{I\epsilon T} \quad (3.1)$$

where Y_{Ly} is the X-ray yield of one of the L X-ray components L_y for a given number of incident particles on the target, I (the subscript index y of L here refers to the particular component of L X-rays such as ℓ , α , β , γ , etc.); ϵ is the overall efficiency of the Si(Li) detector (including its solid angle and all absorbers in the path through which the photons travel from the target to the detector) and T the target thickness actually seen by the beam, obtained through Rutherford scattering measurements either in

an ORTEC scattering chamber or in the X-ray chamber, which has a beam monitor installed primarily at 45° relative to the beamline. The data from the ORTEC chamber were corrected for the 45° orientation of the target relative to the beamline. The details of the measurements of I , ϵ , and T were described in Chapter II.

In order to interpret the measured X-ray production cross sections one has to relate them to the theoretical total ionization cross sections. Because of the complicated decay scheme of the L X-rays (Fig. 1) and the difficulties involved in resolving all the L X-ray lines (especially the L_β group, which contains contributions from all L-subshells), one usually evaluates the individual X-ray production cross section of each resolved component and runs through the following analytic formula to relate the X-ray production cross sections to the ionization cross section.

$$\sigma_{L_\ell}^x = (\sigma_{L_1} f_{13} + \sigma_{L_1} f_{12} f_{23} + \sigma_{L_2} f_{23} + \sigma_{L_3}) \omega_3 F_{3\ell} \quad (3.2)$$

$$\sigma_{L_\alpha}^x = (\sigma_{L_1} f_{13} + \sigma_{L_1} f_{12} f_{23} + \sigma_{L_2} f_{23} + \sigma_{L_3}) \omega_3 F_{3\alpha} \quad (3.3)$$

$$\begin{aligned} \sigma_{L_\beta}^x &= \sigma_{L_1} \omega_1 F_{1\beta} + (\sigma_{L_1} f_{12} + \sigma_{L_2}) \omega_2 F_{2\beta} \\ &\quad + (\sigma_{L_1} f_{13} + \sigma_{L_1} f_{12} f_{23} + \sigma_{L_2} f_{23} + \sigma_{L_3}) \omega_3 F_{3\beta} \end{aligned} \quad (3.4)$$

$$\sigma_{L_\gamma}^x = \sigma_{L_1} \omega_1 F_{1\gamma} + (\sigma_{L_1} f_{12} + \sigma_{L_2}) \omega_2 F_{2\gamma} \quad (3.5)$$

where $\sigma_{L_\ell}^x$, $\sigma_{L_\alpha}^x$, $\sigma_{L_\beta}^x$, and $\sigma_{L_\gamma}^x$ are the X-ray production cross sections of the components L_ℓ , L_α , L_β , and L_γ ; σ_{L_1} , σ_{L_2} and σ_{L_3} are ionization cross sections of the subshells L_I , L_{II} , and L_{III} , respectively; ω_1 , ω_2 and ω_3 are their fluorescence yields; F_{ny} ($F_{3\ell}$, $F_{3\alpha}$, $F_{3\beta}$ --- etc.) are

the fraction of the radiation width of the subshell L_n (L_I , L_{II} and L_{III}) contained in the y^{th} spectral line, i.e.,

$$F_{ny} = \frac{\Gamma_{ny}}{\Gamma_n}$$

(for example, $F_{3\ell} = \frac{\Gamma_{3\ell}}{\Gamma_3}$).

Here, Γ_n is the total radiation width of L_n . In eq. 3.2-3.5, the parameters f_{12} , f_{23} and f_{13} are the so-called Coster-Kronig transition probabilities of $L_I \rightarrow L_{II}$, $L_{II} \rightarrow L_{III}$ and $L_I \rightarrow L_{III}$ respectively (the arrow indicates the direction of the electron vacancy transition between subshells).

Most of the previous works done on the L subshell studies^{33,35} used theoretical values of σ_{L_1} , σ_{L_2} and σ_{L_3} combined with known ω 's, F 's and f 's on the right hand side of Eq. (3.2) to Eq. (3.5) to evaluate the X-ray production cross section on the left hand side for comparisons. The advantage of this procedure is that it may get rid of the Coulomb deflection effect and some of the distortions of L-subshell electron wave functions due to the incoming charged particles, in cases where only ratios of the L X-ray production cross sections are to be compared. This serves as a comparison between experiment and theory for a particular correction in the interaction mechanism (cf. Ref. 7 which investigates the relativistic correction in PWBA). However, the actual features of the individual ionization cross sections may be hidden in this way. For the purpose of obtaining actual ionization cross sections from L X-ray measurements, we selected the L_α and L_γ X-rays to do a detailed analysis; the use of such an analysis is explained below.

The barely resolved L_γ x-rays have been computer-analyzed into four Gaussians with overall shape fits with χ^2 per degree of freedom of around 1.2 ± 0.5 . Four components of L_γ were obtained: L_{γ_5} and L_{γ_1} which relate to the L_{II} subshell, and $L_{\gamma_{2+3}}$ and L_{γ_4} , which relate to the L_I subshell. The Gaussian shape of the mono-energetic X-ray response from a Si(Li) detector can be justified by identifying the peak energy of each X-ray component in L_γ after the fitting. Examples of this fitting for Ta, Au and Bi are presented in Fig. 11. The peak energies are indicated by the arrows, and compare excellently with values measured with much higher resolution. (Other possible weak forbidden transitions are not indicated.) The widths of the Gaussians were selected and kept constant for each of the elements Ta, Au and Bi during the fitting in order to have a quick fit.

Eq. (3.5) can now be broken into

$$\sigma_{\gamma_{2+3}}^x + \sigma_{L\gamma_4}^x = \sigma_{L_1} \omega_1 F_1 \gamma_{2+3+4}, \text{ and} \quad (3.6)$$

$$\sigma_{L\gamma_5}^x + \sigma_{L\gamma_1}^x = (\sigma_{L_1} f_{12} + \sigma_{L_2}) \omega_2 F_2 \gamma_{5+1}. \quad (3.7)$$

The unresolvable L_{γ_6} in $L_{\gamma_{2+3}}$ (see Fig. 11) may raise a significant error in deducing σ_{L_1} from the uncorrected X-ray production cross section

$$\sigma_{L\gamma_{2+3+6}}^x + \sigma_{\gamma_4}^x \approx \sigma_{L_{2+3}}^x + \sigma_{\gamma_4}^x,$$

especially when σ_{L_2} is larger than σ_{L_1} . In order to eliminate this L_{γ_6} contribution, a calculation has been employed. That is to estimate σ_{L_2} from Eq. (3.7) by neglecting the small contribution of Coster-Kronig transitions, then put this estimated σ_{L_2} into Eq. (3.5) to evaluate σ_{L_1} .

Fig. 11 Gaussian fittings for L_{γ} X-rays of Ta, Au, and Bi.

The σ_{L_1} so obtained was run back to Eq. (3.7) to get a more correct value of σ_{L_2} . σ_{L_1} and σ_{L_2} now are considered good enough to make a correction of the $L_{\gamma_{2+3+4}}$ X-ray production cross section as follows:

$$\sigma_{L_{\gamma_{2+3}}}^x + \sigma_{L_{\gamma_4}}^x = (\sigma_{L_{\gamma_{2+3+6}}}^x + \sigma_{L_{\gamma_4}}^x) - (\sigma_{L_1} f_{12} + \sigma_{L_2}) \omega_2 F_{2\gamma_6} \quad (3.8)$$

Once this correction is made, Eq. (3.6) and Eq. (3.7) are used to obtain the final values of σ_{L_1} and σ_{L_2} . A consistent test can be made by putting the final σ_{L_2} into Eq. (3.5) to obtain σ_{L_1} and compare it with the σ_{L_1} just obtained previously. The calculations can be looped until consistency of σ_{L_1} is reached. In our data analysis, one loop is good enough to give a consistency within 0.01%.

σ_{L_3} was obtained through Eq. (3.3) by using the measured L_α X-ray production cross sections $\sigma_{L_\alpha}^x$, and σ_{L_1} , σ_{L_2} which were deduced as described in the last paragraph. A consistency check was also made (for Ta, Au, and Bi) for the L_β group through the comparison between the measured $\sigma_{L_\beta}^x$ and that calculated from Eq. (3.4) by using our deduced values of σ_{L_1} , σ_{L_2} and σ_{L_3} . These two sets of L_β X-ray yields of Ta, Au, and Bi are plotted in Fig. 12. The systematically higher value ($\approx 4\%$) of the measurements for L_β yields in Ta and Au than those calculated from Eq. (3.4) could be due to uncertainties in the efficiency calibration of the detector in the interpolated region (see 2.D). However, the agreement is still within 5% for all points, which include both proton bombardment and alpha bombardment.

Fig. 12A-12C Calculated and measured yields of L_β X-rays for Ta, Au, and Bi. The calculated values are obtained by using the deduced ionization cross sections σ_{L_1} , σ_{L_2} , and σ_{L_3} , assuming the same atomic parameters used in L_α and L_γ analysis. The statistical errors of the measured values are $\leq 2\%$.

3.B. Atomic Parameters and the Final Results

The uncertainties in converting X-ray production cross sections into ionization cross sections depends mostly on the accuracy of the fluorescence yields ω_1 , ω_2 , and ω_3 . A small effect also comes from the Coster-Kronig transition probabilities f_{12} , f_{23} , and f_{13} . The fraction of width, F_{ny} , used here is from the radiative emission rate calculated by Scofield.³⁶ The recently observed L_α/L_ℓ ratios either in a source experiment³⁷ or in a particle-induced experiment³⁵ give discrepancies from Scofield's calculation. In the source experiment ($55 \leq Z \leq 94$)³⁷, the theoretical ratios are found to be underestimated at low Z ($Z \leq 73$), while in the particle-induced experiments, the theoretical ratios are higher for Sm ($Z = 52$). The lower ratios of L_α/L_ℓ observed for Sm may be due to some multionization occurring in L_{III} and $M_{IV,V}$ for L_α . We thus checked our Ta data, which shows a good agreement with Scofield's value (Table 3). (The uncertainties assigned were due to the statistical error and 10% from other sources of error, mostly the efficiency calibration of the detector.) Other experimental values^{38,39} showed that Scofield's values are quite good. We evaluated some of the partial radiation widths F_{ny} from the experimental values available in Ref. 38 and tabulated in Table 4 to compare with Scofield's values. We found that there is a good agreement in theory and experiment. Therefore, in our converting process, we use Scofield's values for all the elements Ta, Au and Bi (Pb values in Scofield were used for Bi for the lack of Bi values there).

A review article of Bamlynek et al.⁴⁰ about fluorescence yields and Coster-Kronig transition probabilities which reviewed this field up to 1972, was strongly relied on to obtain a good value of fluorescence yields, and Coster-Kronig transition probabilities. We

TABLE 3. L_{α}/L_{β} Ratios for Ta. Scofield's Value = 21.84

E_p (MeV/a.m.u.)	L_{α}/L_{β} Proton Bomb.	E_{α} (MeV/a.m.u.)	L_{α}/L_{β} alpha bomb.
1.00	19.6 $\pm$ 2.6	0.375	24.5 $\pm$ 5.7
1.25	18.7 $\pm$ 2.4	0.500	19.3 $\pm$ 2.6
1.50	18.7 $\pm$ 2.4	0.625	19.9 $\pm$ 2.5
1.75	19.1 $\pm$ 2.3	0.750	18.8 $\pm$ 2.3
2.00	19.7 $\pm$ 2.3	0.875	18.6 $\pm$ 2.2
2.25	19.9 $\pm$ 2.3	1.000	19.6 $\pm$ 2.2
2.50	19.4 $\pm$ 2.3	1.125	19.9 $\pm$ 2.4
2.75	20.9 $\pm$ 2.3	1.250	20.2 $\pm$ 2.3
3.00	20.2 $\pm$ 2.2	1.375	20.0 $\pm$ 2.4
3.25	21.2 $\pm$ 2.2	1.50	19.5 $\pm$ 2.3
3.50	20.8 $\pm$ 2.2	1.625	20.2 $\pm$ 2.3
3.75	20.1 $\pm$ 2.2	1.75	19.7 $\pm$ 2.2
4.00	20.0 $\pm$ 2.2	1.875	20.4 $\pm$ 2.2
4.25	20.9 $\pm$ 2.2	2.00	20.0 $\pm$ 2.3
4.50	21.1 $\pm$ 2.2	2.175	21.0 $\pm$ 2.4
4.75	19.5 $\pm$ 2.2	2.750	20.2 $\pm$ 2.3
5.00	20.5 $\pm$ 2.2	2.375	21.5 $\pm$ 2.3
5.25	20.0 $\pm$ 2.2	2.50	20.5 $\pm$ 2.3
		2.625	20.4 $\pm$ 2.4

TABLE 4. Fraction of Radiation Width from (a) McGrary et al.³⁸ and (b) Scofield.³⁶

F_{ny}		$F_{3\alpha}$	$F_{2\gamma_{1+5}}$	$F_{1\gamma_{2+3+4}}$
Element				
Ta	a)	0.80 ± 0.02	0.150 ± 0.014	0.27 ± 0.04
	b)	0.807	0.156	0.217
Au	a)		0.164 ± 0.014	
	b)	0.787	0.163	0.227

tabulate both experimental and theoretical values in Table 5 for Ta, Au, and Bi. Here the "references" are listed in Ref. 40, and v_1 , v_2 and v_3 are Coster-Kronig-corrected fluorescence yields defined as

$$\sigma_L^X = v_1 \sigma_{L_1} + v_2 \sigma_{L_2} + v_3 \sigma_{L_3}$$

and have the relations

$$v_1 = \omega_1 + \omega_2 f_{12} + \omega_3 (f_{13} + f_{12} f_{23}) \quad (3.9)$$

$$v_2 = \omega_2 + \omega_3 f_{23} \quad (3.10)$$

$$v_3 = \omega_3 \quad (3.11)$$

The values we used in converting X-ray production cross section to ionization cross section are shown underlined. The tungsten theoretical values are tabulated to make up for the lack of Ta theoretical values. The value $\omega_1 = 0.096$, for Ta, was calculated by putting experimental values of $v_1 (= 0.218)$, $v_2 (= 0.303)$, $\omega_3 (= 0.228)$ and $f_{23} (= 0.148)$, and theoretical values of $f_{12} (= 0.160)$ and $f_{13} (= 0.324)$ into Eq. (3.9) and Eq. (3.10). This value was used instead of McGuire's value, $\omega_1 = 0.115$ to do the conversion. In Tables 6 and 7, we list all ionization cross sections we obtained in our laboratory and also include corresponding target thicknesses, and L_α X-ray production cross sections for future comparisons. Previous work with thick targets⁴³ and thin targets⁴⁸ are included for comparison.

TABLE 5. Fluorescence Yields and Coster-Konig Transition Probabilities of Ta, Au and Bi.

	ω_1	ω_2	ω_3	ν_1	ν_2	ϵ_{12}	f_{12}	f_{21}	References
Ta (Exp.)		0.25 ± 0.02	0.27 ± 0.01		0.31 ± 0.01			0.020 ± 0.04	Rao(1965b)
			0.25 ± 0.103		0.37 ± 0.06				Jopson(1964a)
			0.191	0.284	0.326				Kustner(1935)
	0.096^*	0.257 ± 0.013 (0.269)*	0.228 ± 0.013	0.218 ± 0.016	0.303 (0.303)	<0.14 (0.160)	<0.36 (0.324)	0.148 ± 0.010	Mohan(1970a)
			0.254 ± 0.025		0.303 ± 0.030		0.19		Price(1968) Ferreira(1965) Rao(1966)
				0.22 ± 0.01					
W (Theo.)	0.115	0.287	0.268			0.195	0.332	0.123	McGuire(1971a)
	0.138	0.271	0.253			0.160	0.324	0.117	Chen(1971a) and Craseman(1971)
Au	0.210*	0.236*	0.276	0.410	0.272	(0.083)	(0.644)	(0.132)	Kustner(1935)
	0.201*	0.211*				(0.25)	(0.51)	(0.22)	
			0.31 ± 0.04						Jopson(1964a)
	0.173*	0.253*	0.317 ± 0.025	(0.410)	0.395 ± 0.032	(0.083)	(0.644)	(0.132)	Price(1968)
	0.150*	0.325*	(0.317)	(0.410)	(0.395)	0.25 ± 0.13	0.51 ± 0.13	(0.22)	Paschke(1963)
							0.61 ± 0.07		Ferreira(1965)
	0.105	0.357	0.327			0.083	0.644	0.132	McGuire(1970a)
Bi			0.367						Kustner(1935)
			0.36						Risch(1958)
						0.19 ± 0.05	0.58 ± 0.05	$0.06^{+0.14}_{-0.04}$	Ross(1955)
						0.18 ± 0.02	0.58 ± 0.02	(0.16)	Freund and Fink(1969)
					0.51 ± 0.08 0.41 ± 0.039				Jopson(1964a) Price(1968)
	0.120	0.417	0.389			0.069	0.656	0.101	McGuire(1970a)

* Calculated values from existing data combined with other values from other sources which are indicated in parenthesis.

TABLE 6. L_{α} X-Ray and L-Subshell Ionization Cross Sections of Proton Bombardment.

1. Element and Its Thickness In $\mu\text{g}/\text{cm}^2$	2. Ep In MeV/a.m.u.	3. $\sigma_{L_{\alpha}}^x$ In Barns (This work) $\dagger$	4. $\sigma_{L_{\alpha}}^x$ In Barns (Previous work) $\ast$	5. σ_{L_1} In Barns	6. σ_{L_2} In Barns	7. σ_{L_3} In Barns (a)	8. σ_L In Barns (a)	9. σ_L In Barns (Previous work) $\dagger$
Ta (111.1)	1.00	5.74	4.86	2.65 ± 0.27	7.86 ± 0.24	28.6	39.1	
	1.25	9.89		3.46 ± 0.55	14.1 ± 0.4	49.6	67.2	
	1.50	15.4		6.07 ± 0.73	23.6 ± 0.7	76.8	106	84
	1.75	21.3		9.97 ± 0.90	32.8 ± 0.7	106	148	126
	2.00	28.6		21.1 ± 1.1	43.4 ± 0.9	138	203	192
	2.25	35.0	22	30.5 ± 1.5	52.3 ± 2.6	167	250	
	2.50	42.7		46.0 ± 1.4	63.4 ± 1.3	200	310	265
	2.75	50.6		58.4 ± 1.8	77.0 ± 1.5	235	371	
	3.00	60.0	49	77.9 ± 2.3	91.6 ± 0.9	276	445	370
	3.25	66.6		94.7 ± 1.9	103 ± 1	302	499	440
	3.50	72.6		108 ± 2	115 ± 1	327	549	495
	3.75	80.2		129 ± 3	126 ± 1	357	611	584
	4.00	88.1		148 ± 3	131 ± 1	390	669	630
	4.25	92.9		156 ± 3	143 ± 1	411	709	705
	4.50	100		178 ± 3	156 ± 2	439	773	
	4.75	106		192 ± 4	159 ± 2	462	813	
	5.00	115		215 ± 4	183 ± 2	498	896	
	5.25	116		225 ± 4	185 ± 2	497	907	
.....								
Au (98.2)	1.00	2.65	2.7	0.65 ± 0.09	2.48 ± 0.10	9.46	12.6	
	1.25	4.75		1.24 ± 0.12	4.91 ± 0.15	16.8	23.0	
	1.50	7.61		1.93 ± 0.14	8.30 ± 0.17	26.9	37.2	22
	1.75	10.8		2.62 ± 0.29	11.7 ± 0.4	38.4	52.7	63
	2.00	14.7		3.97 ± 0.36	16.0 ± 0.3	52.0	72.0	73

TABLE 6. (Continued)

1. Element and Its Thickness In $\mu\text{g}/\text{cm}^2$	2. E_p in MeV/a.m.u.	3. $\sigma_{L\alpha}^x$ In Barns (This work) $\dagger$	4. $\sigma_{L\alpha}^x$ In Barns (Previous work)*	5. σ_{L_1} In Barns	6. σ_{L_2} In Barns	7. σ_{L_3} In Barns (a)	8. σ_L In Barns (a)	9. σ_L In Barns (Previous work) $\dagger$
Au (98.2)	2.25	19.1	18	6.32 ± 0.38	20.9 ± 0.4	66.6	93.8	
	2.50	23.3		9.10 ± 0.46	26.4 ± 0.5	80.0	116	131
	2.75	28.3		12.2 ± 0.6	32.3 ± 0.7	96.3	141	
	3.00	33.2	32	18.2 ± 0.7	38.0 ± 0.5	110	166	203
	3.25	38.6		23.9 ± 0.8	44.2 ± 0.6	127	194	233
	3.50	43.4		28.9 ± 0.9	50.8 ± 0.7	141	220	163
	3.75	49.8		34.8 ± 0.9	56.0 ± 0.7	160	251	288
	4.00	55.0		40.4 ± 1.1	60.6 ± 0.7	175	276	318
	4.25	59.4		46.3 ± 1.1	66.5 ± 0.8	187	300	345
	4.50	64.7		51.2 ± 1.3	72.8 ± 0.9	203	327	
	4.75	70.6		60.6 ± 1.3	78.2 ± 0.9	219	358	
	5.00	74.7		67.8 ± 1.5	83.2 ± 0.9	230	379	
	5.25	79.7		71.4 ± 1.5	87.5 ± 1.0	245	403	
	5.50	85.7		80.2 ± 1.5	94.2 ± 0.9	261	435	
Au (101.4)	1.00	2.75	2.7	0.80 ± 0.06	2.42 ± 0.05	9.73	13.0	
	1.125	3.90		1.05 ± 0.08	3.49 ± 0.07	13.9	18.4	
	1.375	6.31		1.58 ± 0.13	6.02 ± 0.12	22.5	30.1	
	1.625	9.94		2.28 ± 0.16	10.3 ± 0.2	35.4	48.0	
	1.875	13.4		3.37 ± 0.20	14.6 ± 0.2	47.5	65.5	
	2.125	17.5		4.87 ± 0.29	18.8 ± 0.3	61.8	85.5	
	2.375	22.5		8.06 ± 0.40	24.7 ± 0.4	77.7	111	
	2.625	27.6		10.7 ± 0.5	31.6 ± 0.5	94.8	137	
	2.875	33.2	32	14.9 ± 0.7	36.6 ± 0.6	113	164	
	3.000	35.1		18.2 ± 0.7	38.4 ± 0.6	118	174	203
	3.125	38.1		21.8 ± 0.9	44.2 ± 0.7	126	192	

TABLE 6. (Continued)

1. Element and Its Thickness In $\mu\text{g}/\text{cm}^2$	2. E_p In MeV/a.m.u.	3. $\sigma_{L\alpha}^x$ In Barns (This work)†	4. $\sigma_{L\alpha}^x$ In Barns (Previous work)*	5. σ_{L_1} In Barns	6. σ_{L_2} In Barns	7. σ_{L_3} In Barns (a)	8. σ_L In Barns (a)	9. σ_L In Barns (Previous work)†
Bi (49.4)	1.00	1.38	1.6	0.63 ± 0.11	1.24 ± 0.09	4.02	5.89	
	1.25	2.83		0.69 ± 0.11	2.21 ± 0.12	8.67	11.6	
	1.50	4.53		1.06 ± 0.14	4.42 ± 0.17	13.8	19.3	
	1.75	6.64		1.61 ± 0.14	6.33 ± 0.21	20.2	28.2	
	2.00	9.15		2.33 ± 0.19	8.86 ± 0.22	27.8	38.9	
	2.25	12.0		3.23 ± 0.51	11.6 ± 0.3	36.3	51.1	
	2.50	15.1		4.65 ± 0.55	15.9 ± 0.5	45.1	65.6	
	2.75	18.3		5.94 ± 0.42	18.6 ± 0.5	54.5	79.0	
	3.00	21.9		8.14 ± 0.54	22.5 ± 0.6	64.7	95.4	
	3.25	25.4		12.1 ± 0.7	25.4 ± 0.6	73.1	111	
	3.50	29.4		14.8 ± 0.7	29.6 ± 0.7	84.1	129	
	3.75	33.0		16.0 ± 0.8	33.3 ± 0.7	95.0	144	
	4.00	36.8		20.9 ± 1.0	37.6 ± 0.8	104	162	
	4.25	41.4		23.1 ± 1.1	41.5 ± 0.9	117	182	
	4.50	45.5		30.3 ± 1.3	46.5 ± 0.9	125	202	
	5.00	52.0		34.5 ± 1.5	53.1 ± 1.1	143	231	
	5.25	55.7		36.2 ± 1.5	56.7 ± 1.1	154	247	
	5.50	59.3		41.8 ± 1.9	59.7 ± 1.2	162	263	

* Bearnse et al.⁴¹† Bernstein and Lewis⁴³‡ The statistical errors are 1-2% for energy below $E_p = 2.0$ MeV, and less than 1% for the rest of them.(a) The statistical error comes mainly from the statistical uncertainty of the $L\alpha$ x-ray yields, thus the statistical error is approximately equal to that of $L\alpha$ x-ray production cross section.

TABLE 7. $L\alpha$ X-Ray L-Subshell Ionization Cross Sections of Alpha Bombardment.

1. Element and Its Thickness In $\mu\text{g}/\text{cm}^2$	2. E α In MeV/a.m.u.	3. $\sigma_{L\alpha}^x/4$ In Barns (This work)*	4. $\sigma_{L\alpha}^x/4$ In Barns (Previous work)*	5. $\sigma_{L1}^x/4$ In Barns	6. $\sigma_{L2}^x/4$ In Barns	7. $\sigma_{L3}^x/4$ In Barns (a)	8. $\sigma_{L4}^x/4$ In Barns (a)	9. $\sigma_{L4}^x/4$ In Barns (Previous work)†
Ta (111.1) α^+ α^+	0.375	0.252		0.47 ± 0.04	0.29 ± 0.02	1.11	1.87	
	0.500	0.783		0.96 ± 0.07	1.06 ± 0.04	3.64	5.66	
	0.625	1.60		1.23 ± 0.09	2.24 ± 0.07	7.72	11.2	
	0.750	2.50		1.35 ± 0.11	3.36 ± 0.07	12.4	17.1	
	0.875	3.76		1.94 ± 0.16	5.21 ± 0.10	18.6	25.7	
	1.000	5.31		2.22 ± 0.13	7.27 ± 0.14	26.5	36.0	
	1.125	7.00		2.72 ± 0.22	9.55 ± 0.19	35.1	47.4	
	1.250	9.17		3.62 ± 0.25	13.4 ± 0.3	45.8	62.8	
	1.250	9.22		3.42 ± 0.24	13.4 ± 0.3	46.1	63.0	
	1.375	11.4		4.29 ± 0.39	16.4 ± 0.2	57.0	77.8	
	1.500	14.0		5.24 ± 0.31	20.1 ± 0.2	70.0	95.3	
	1.625	16.9		7.62 ± 0.38	25.2 ± 0.3	84.1	117	
	1.750	19.8		10.0 ± 0.4	29.9 ± 0.3	98.0	138	
	2.875	22.9		13.4 ± 0.4	34.0 ± 0.3	112	160	
	2.000	26.6		17.9 ± 0.5	40.2 ± 0.4	129	187	
	2.125	30.0		23.0 ± 0.9	46.4 ± 0.5	144	214	
	2.250	33.8		28.9 ± 0.9	49.6 ± 0.5	162	240	
	2.375	36.4		32.9 ± 1.0	54.9 ± 0.6	173	261	
	2.500	41.1		41.0 ± 1.2	62.9 ± 0.6	194	298	
	2.625	44.2		46.2 ± 1.9	67.0 ± 0.7	208	321	
Au (101.4)	0.250	0.202		0.043 ± 0.004	0.021 ± 0.002	0.043	0.11	
	0.375	0.110		0.15 ± 0.01	0.105 ± 0.006	0.30	0.56	
	0.500	0.229		0.36 ± 0.02	0.27 ± 0.01	0.86	1.49	
	0.625	0.669		0.62 ± 0.03	0.61 ± 0.02	2.05	3.29	
	0.750	1.12		0.61 ± 0.04	1.05 ± 0.03	3.75	5.41	
	0.875	1.68		0.85 ± 0.04	1.73 ± 0.04	5.66	8.24	
	1.000	2.52		0.86 ± 0.10	2.54 ± 0.08	8.80	12.2	
	1.125	3.52		0.91 ± 0.08	3.69 ± 0.11	12.5	17.1	

TABLE 7. (Continued)

1. Element and Its Thickness In $\mu\text{g}/\text{cm}^2$	2. Ex In MeV/a.m.u.	3. $\sigma_{\alpha/4}^*$ In Barns (This work)†	4. $\sigma_{\alpha/4}^*$ In Barns (Previous work)*	5. $\sigma_{L1/4}$ In Barns	6. $\sigma_{L2/4}$ In Barns	7. $\sigma_{L3/4}$ In Barns (a)	8. $\sigma_{L/4}$ In Barns (a)	9. $\sigma_{L/4}$ In Barns (Previous work)†
Au (101.4)	α^+	1.250	4.60	1.00 ± 0.08	4.64 ± 1.11	16.5	22.1	
	α^+	1.250	4.74	1.06 ± 0.07	4.98 ± 0.10	16.9	22.9	
		1.375	5.88	1.48 ± 1.10	6.31 ± 0.13	20.8	28.6	
		1.500	7.27	1.79 ± 0.11	7.77 ± 0.16	25.8	35.3	
		1.625	9.14	2.23 ± 0.13	9.93 ± 0.15	32.4	44.6	
		1.750	10.6	2.84 ± 0.14	11.7 ± 0.2	37.4	51.9	
		1.875	12.6	3.83 ± 0.15	13.8 ± 0.2	44.2	61.9	
		2.000	14.4	4.27 ± 0.30	16.0 ± 0.3	50.4	70.7	
		2.125	16.5	5.74 ± 0.40	17.9 ± 0.4	57.3	80.9	
		2.250	18.5	6.99 ± 0.35	20.5 ± 0.4	63.6	91.0	
		2.375	20.6	7.67 ± 0.38	23.3 ± 0.5	71.0	102	
		2.500	23.0	9.82 ± 0.49	25.9 ± 0.4	78.3	114	
		2.625	25.1	12.3 ± 0.5	27.3 ± 0.4	84.6	124	
		2.750	27.6	14.5 ± 0.6	30.5 ± 0.5	92.0	137	
Bi (23.9)		0.375	0.0566	0.10 ± 0.02	0.029 ± 0.004	0.12	0.25	
		0.500	0.203	0.18 ± 0.02	0.16 ± 0.01	0.53	0.87	
		0.625	0.431	0.27 ± 0.03	0.33 ± 0.01	1.25	1.85	
		0.750	0.757	0.43 ± 0.04	0.60 ± 0.02	2.15	3.18	
		0.875	1.19	0.46 ± 0.04	0.98 ± 0.04	3.53	4.97	
		1.000	1.76	0.56 ± 0.06	1.47 ± 0.06	5.29	7.33	
		1.125	2.39	0.69 ± 0.05	1.88 ± 0.08	7.25	9.82	
		1.250	3.11	0.88 ± 0.09	2.83 ± 0.11	9.41	13.1	

* Bearnse et al.⁴²† Bernstein and Lewis⁴³

‡ and (a) see remarks on Table 5.

3.C. Error Analysis

The standard counting errors for the L_{α} X-rays of Ta, Au, and Bi induced by proton bombardment are less than 1%; those for L_{γ} X-rays which relate to L_I ionization (i.e., $L_{\gamma_{2+3+4}}$) are generally less than 10%, and less than 6% for L_{γ} X-rays which relate to L_{II} ionization (i.e., $L_{\gamma_{1+5}}$), except where the incident proton energy is below 2.0 MeV. For these low energies, 10-15% standard errors were estimated for both of them. All the errors assigned in Table 6 and Table 7 are due to these statistics. Because the ionization cross section is a sharply decreasing function with decreasing velocity of the impinging particle, the standard error for the L X-rays from alpha bombardment is generally 2-4% higher than that of proton bombardment at an equal incident energy even though a reasonable adjustment of the charge collection was made for alpha bombardment. Aside from this uncertainty from the statistics of the counting process, there are several possible sources of uncertainty contributing to the ionization cross sections during the data reduction processes:

- (1) In the measurement of target thickness.
- (2) In the measurement of efficiency of the Si(Li) detector.
- (3) Self-absorption by the target atoms.
- (4) In assuming the angular distribution of the L X-rays to be isotropic.
- (5) In all atomic parameters, such as fluorescence yields (a correction for radiationless transitions - Auger emissions), Coster-Kronig transition probabilities (a correction for the transitions between L subshells themselves), and the radiative

transition rates (a correction for the branching ratios, or partial widths, of the measured L X-ray components). These parameters are either from theoretical calculations or from experimental values of other sources.

- (6) Coulomb excitations of the target nucleus and electron captures of the projectile.
- (7) K-shell ionization contributions.

The first four deal directly with the measurements of the yields of each component of the L X-rays, while the others refer to the conversion processes - converting the X-ray yields to vacancy-production probabilities.

3.C-1. Errors from the Measurement of the L X-ray Production Cross Sections

The uncertainties in (1) and (2) have been discussed in Chapter II in detail. They contribute an overall error of about 15%. As mentioned in Chapter II, the error arising in the charge collection of the incident beam was included in that of the target thickness measurement with an elastic-scattering beam monitor. Of course, the uncertainty in the measurement of the thickness of the reference targets ($\approx 2\%$) was also included.

The self-absorption of the target itself is very small for these thin foils of Ta, Au, and Bi ($\leq 100 \mu\text{g}/\text{cm}^2$). This can be estimated easily by following formula:

$$N_d = \int_0^T N(\tau) e^{-\mu\tau \tan \Theta} d\tau, \quad (3.12)$$

where N_d is the total number of photons to be detected by the Si(Li) detector; $N(\tau)$ is the number of photons emitted at the depth τ in the interval $d\tau$ (see Fig. 13); T is the target thickness actually seen by the incident beam; Θ is the angle of the target with respect to the beam direction; and μ the mass absorption coefficient of the target. Since the energy loss of the incident charged particle is very small [in our experimental energy range, the energy loss of the protons and alpha particles range from 2.7 keV (for 5.5 MeV protons) to 38.1 keV (for 1.0 MeV alpha-particles) when they pass through a $100 \mu\text{g}/\text{cm}^2$ tantalum foil], one can approximate $N(\tau)d\tau$ as

$$N(\tau)d\tau \approx \frac{N}{T} d\tau ,$$

where N is the total number of photons emitted. From this approximation, one easily obtains

$$N_d = N \frac{(1 - e^{-\mu T \tan \Theta})}{\mu T \tan \Theta} ;$$

at $\Theta = 45^\circ$,

$$N_d(\Theta = 45^\circ) = N \frac{1 - e^{-\mu T}}{\mu T}$$

The self-absorption correction factor $\frac{1}{\mu T} (1 - e^{-\mu T})$ was calculated for a Ta target ($T = 100 \mu\text{g}/\text{cm}^2$) and 10 keV photons as 0.988 (the value μT was obtained from Nuclear Data Tables, Vol. 7, No. 6, June, 1970). This approximately 1% correction was not made in our data analysis for it is small compared to the other sources of uncertainties.

The isotropic nature of X-rays induced by a swift charged particle

Fig. 13 The orientation of the target.

has been assumed for a long time, but so far as we know, only two previous measurements show the correctness of this assumption for K X-rays^{41,42} for Cu, Ti, and Sn, and two measurements for L X-rays for Sn and Au^{41,43}. However, one can always argue on the basis of angular momentum that there is no reason for this assumption to be incorrect; in fact, it is always true that for s states ($\ell = 0$) of the atom and E1 radiation, the isotropic property can be shown to be independent of the populations of the magnetic substates⁴⁴. For p states ($\ell = 1$), so long as the populations in magnetic substates are equal, there are also isotropic angular distributions. Another simple way of looking at it is that the time of interaction between the fast charged particle and the target atom is much shorter than the lifetime of the ionized state of that atom so that there is no specific quantum direction to be remembered when the high excited atom begins to decay to its ground state. One can easily do the experiment on the L_{α} X-ray angular distribution to check the validity of this isotropic assumption for the 2p state. The vertical construction of the Si(Li) detector in our laboratory prevented us from performing this measurement. Nevertheless, the ionization of the 2p subshell in our energy range dominates the L-shell ionization and from the previous measurements of L X-ray angular distributions mentioned above, it is safe to hold the isotropic assumption to be correct.

3.C-2. Atomic Parameter Contributions

The uncertainty in the Coster-Kronig transition probabilities contributes no more than 15 percent errors in the determination of the total ionization cross section of each subshell, even though these transition probabilities fluctuate between theories and experiments and between experiments themselves. Table 5 shows this fact; the biggest gap between theory and experiment occurs in f_{12} , which is important in the data analysis. We use theoretical values rather than experimental values since the experimental values either have a large error bar (Au) or no definite value has been determined (Ta). The experimental values for Bi for f_{12} , of Ross, Freund and Fink, are in good agreement with each other; however, we use theoretical values instead for ease in tracing the deduction of the ionization cross section of Bi and to compare it with those of Ta and Au. The theoretical values for Au and Bi are three times lower than those of experiment, but affect the determination of the L_{II} subshell ionization cross section (mainly from Eq. (3-7)) less than 15 percent, since the first term of Eq. (3-7) contribute at most $\approx 15\%$ of the $L_{\gamma_{1+5}}$ X-ray yields. The higher value of f_{12} makes σ_{L_2} smaller. We plot σ_{L_2} values for Au which were deduced by using both $f_{12} = 0.083$ and $f_{12} = 0.25$, in Fig. 14.

The value of σ_{L_1} does not involve any effect from Coster-Kronig transition probabilities. σ_{L_3} has a negligible effect from f_{12} , not only because $\sigma_{L_3} \gg \sigma_{L_1}$ in our energy range but also because the product of f_{12} and f_{23} is small (≈ 0.01). The effect of f_{13} and f_{23} will also be small, again because $\sigma_{L_3} > \sigma_{L_1}$ in the energy range we are interested in. Nevertheless, f_{13} and f_{23} have more experimental information and

Fig. 14 σ_{L_2} of Au which are deduced by using $f_{12} = 0.083$ (theory) and $f_{12} = 0.25$ (experiment) for proton bombardments.

are closer to the theoretical values than f_{12} . From a comparison of all existing values, either theoretical or experimental, the values of f_{13} and f_{23} we used in our data analysis could be believed to $\leq 15\%$ which makes the error contributions of f_{13} and f_{23} to the L_{III} ionization cross section σ_{L_3} at most 10% in the extreme case when $\sigma_{L_3} = \sigma_{L_2} = \sigma_{L_1}$. However, σ_{L_3} is, in general, four times larger than σ_{L_2} and seven times larger than σ_{L_1} , which reduces the error contributions of f_{13} and f_{23} to $\approx 1\%$.

The lack of experimental information on fluorescence yield ω_1 and ω_2 could give rise to errors in the ionization cross sections of the L_I and L_{II} subshells. Furthermore, the fluorescence yields depend both on the projectile ion and its impact energy. A factor of four difference in fluorescence yields for the Ar L shell, between H^+ and He^+ bombardment at low energy (≤ 200 keV/a.m.u.), were observed by Stolterfoht *et al.*⁴⁵ The change of fluorescence yield is primarily because of the different electron configurations created by the incident projectile ions (different degrees of multionization occur with different impact projectiles and varying energies). However, from our data in the overlap region of H^+ and ${}^4_2He^{++}$ bombardments, we did not observe any substantial effects of this kind. The fluorescence yields which we used in the deduction of L-subshell ionization cross sections are a kind which is related through a single ionization process, such as electrons, or photons, or from a nuclear disintegration (electron capture or internal conversion processes). The fluorescence yields for single ionization have been investigated in theory and in experiment for a long time (see Table 5). The lack of data in ω_1 and ω_2 is due to the experimental difficulties, which involve detecting Auger electrons or resolving the

L X-rays which decay from L_I or L_{II} subshells. When there is no experimental value available, we use the theoretical values instead as mentioned in the last section of this chapter. Under the assumption that the multi-ionization effect in our energy range of the bombarding particle is negligible (this seems to be justified by noticing that a feedback check of L_β X-rays is quite consistent as indicated in section 3.A), the fluorescence yields could give us no more than $\approx 10\%$ uncertainty, estimated by comparing the various values obtained in different laboratories and the better agreements between theory and experiment. The values of ω_1 and ω_2 in Table 5 which are marked with a star were calculated from Eq. (3.9) and Eq. (3.10) based mainly on the existing experimental values of ν_1 and ν_2 . Those values from other sources in the calculation are indicated in parentheses. The table indicates that the theoretical values give better agreement with more recent experimental values.

The radiative rates of L-subshell transitions were discussed in detail in section A. The use of Scofield's values should be good to 10%.

3.C-3. L-Subshell Vacancies Not Created by Means of a Direct Atomic Coulomb Excitation

The L subshell vacancies can also be created by dropping electrons from L subshells, mainly L_{II} and L_{III} , to fill K shell vacancies (K X-ray transitions) which were also created during the bombardment by the charged particle. However, a previous experiment⁴³ shows several orders of magnitude lower ionization cross sections for the K shell than that for the L shell. We also tried a L-K x-ray coincidence experiment (Appendix A), the negative outcome of which verified that a negligible

contribution arises from K shell ionizations.

Internal conversion of a nuclear transition creates inner shell electron vacancies, also. The contribution to the total X-ray yield depends on the comparative cross section for direct atomic excitation and Coulomb excitation of the nucleus, as well as the conversion coefficients for the excited nuclear states. This contribution was investigated for the Ta¹⁸¹ nucleus⁴⁹ and it was indicated that about 20% correction needed to be made for the K-shell ionization cross section of Ta. Because of the much larger cross section for direct atomic L-shell ionization, the correction for the effects of nuclear excitation is unnecessary in this case. It can further be justified from the small internal conversion coefficients for L subshells from the first and second excited states of the target nuclei Ta¹⁸¹, Au¹⁹⁷, and Bi²⁰⁹. There is little possibility of exciting Bi²⁰⁹ to its first excited state (910 keV) in our incident energy range, not only because its high energy, but also because its transition mode is an M1 transition. The internal conversion coefficient for L-subshells of the first two excited states of Ta and Au are listed in Table 8, based on the theoretical values of Sliv and Band, as listed in Appendix 5 of Ref. 44.

The last possibility of creating inner shell vacancies is from electron capture by the incident charged particle, especially when the incident energy is low. However, from the formula given by Nikolaev⁵⁰, the cross sections for electron capture by the incident charged particle from the L shells of Ta, Au, and Bi targets can be estimated to be very small ($\approx 10^{-9}$ barns for 2.0 MeV protons in collision with Ta). This indicates that no corrections for this electron capture process to

Table 8. Internal Conversion Coefficients of Ta^{181} , Au^{197} , and Bi^{209} for L_I , L_{II} , and L_{III} subshell electrons.

Isotope	1st Excited State and Its Transition Mode	2nd Excited State and Its Transition Mode	Internal Conversion Coefficients
Ta^{181}	6.3 KeV E1	136.25 KeV M1 + 16% E2	$L_I = 1.16 \times 10^{-1}$ $L_{II} = 3.96 \times 10^{-2}$ $L_{III} = 2.13 \times 10^{-2}$
Au^{197}	77.34 KeV M1 + 10% E2	268.8 KeV M1 + 30% E2	$L_I = 2.70$ $L_{II} = 0.86$ $L_{III} = 0.60$
Bi^{209}	9 10 KeV M1		$L_I = 5.07 \times 10^{-3}$ $L_{II} = 5.31 \times 10^{-4}$ $L_{III} = 3.31 \times 10^{-5}$

a) Not including the contribution of the first excited state, since the transition energy is too small to convert to the L shell electrons.

the L-subshell ionization cross sections from direct atomic Coulomb interaction is necessary.

3.C-4. Summary

All errors of the various sources which were mentioned above can be carried over to the final results of our absolute ionization cross sections. Following the method provided by P.R. Bevington, in Data Reduction and Error Analysis for the Physical Science (1969), we estimated that the uncertainties from the measurements of the absolute yields of the components of L_Y X-rays: $L_{Y_{1+5}}$ and $L_{Y_{2+3+4}}$ are $\approx 15-18\%$, and $\approx 15\%$ for the measurements of the absolute L_α X-ray yields; the errors from the conversion processes could amount to a maximum of $\approx 14\%$ for σ_{L_1} and σ_{L_3} , $\approx 21\%$ for σ_{L_2} . The absolute cross section of this work thus can be believed to 21% for σ_{L_3} , 28% for σ_{L_2} and 22% for σ_{L_1} . However, the relative uncertainties are close to the statistical errors, since the other factors such as target thickness, efficiency of the Si(Li) detector and all the atomic parameters affect the results uniformly, i.e., they would not change the shape of the charged particle energy dependence of the ionization cross sections.

CHAPTER IV

DISCUSSIONS AND CONCLUSIONS

In the following discussions, we first make comparisons between our data and PWBA, BEA, and SCA calculations. The "Coulomb deflections effect" and the "binding change effect" which are usually observed in K-shell ionization or total L-shell ionization studies were also observed in these L-subshell ionization studies. The "binding change effect" of the L_{II} and L_{III} subshells seems to be less than that of the L_I subshell in our energy range of studies. In addition to the two effects just mentioned above, other finer corrections, which are due to the use of particular initial and final wave functions of the ionized electron of the target atom, are discussed below. The inadequacy of the use of simple hydrogenic wave functions in the ionization cross section calculation is demonstrated.

In the course of examining the initial and final wave functions, of the ionized electron, we found an unusual behavior of the $\sigma_{L_3}/\sigma_{L_2}$ ratios for the Au target, compared to the others, at low-energy alpha bombardments. The outer-shell electron configurations appear to influence this behavior. This outer-shell electron configurations dependence was further examined by testing the ratio with a Pt target, which has a similar electron configuration of Au. Results support the apparent configuration dependence.

4.A. Discussion of the Results

The resulting L-subshell ionization cross sections for Ta, Au and Bi are plotted in Fig. 15, 16, and 17. Non-relativistic PWBA and CBEA are also included for comparisons. The choice of the units of the coordinates follows the PWBA theory, which predicts that at low bombarding energies, the cross sections following approximately the scaling law

$$(Z_L^4 \sigma_{L_n} \Theta_{L_n}) / Z_1^2 = f\left(\frac{\eta_L}{\Theta_{L_n}^2}\right)$$

where Z_1 is the nuclear charge number of the incident ions; Z_L is the "effective nuclear charge number"¹⁸ of the target atoms, whose atomic number is Z_2 in the L shell, given by

$$Z_L = Z_2 - 4.15.$$

L_n indicates the subshell L_1 , L_2 , and L_3 ; Θ_{L_n} is the screening parameter, which describes the electron binding energy in terms of an "ideal ionization potential of the L_n - subshell without outerscreening", i.e.

$$\Theta_{L_n} = \frac{4I_{L_n}^0}{Z_L^2 \text{Rydberg}}$$

here, $I_{L_n}^0$ is the observed ionization potential of the L_n subshell; η_L is the bombarding energy in units of the hydrogenic electron binding energy, i.e.

$$\eta_L = \left(\frac{v}{Z_L V_0}\right)^2$$

where v is the velocity of the impact ion and V_0 is that of Bohr electron,

Fig. 15 σ_{L_1} , σ_{L_2} , and σ_{L_3} for Ta, with PWBA and CBEA curves; σ_{L_1} , σ_{L_2} , and σ_{L_3} are drawn from the tabulated values of Table 6 and 7. The statistical errors are assigned in these tables.

Fig. 16 σ_{L_1} , σ_{L_2} , and σ_{L_3} for Au, with PWBA and CBEA curves; σ_{L_1} , σ_{L_2} , and σ_{L_3} are drawn from the tabulated values of Table 6 and 7. The statistical errors are assigned in these tables.

Fig. 17 σ_{L_1} , σ_{L_2} , and σ_{L_3} for Bi, with PWBA and CBEA curves; σ_{L_1} , σ_{L_2} , and σ_{L_3} are drawn from the tabulated values of Table 6 and 7. The statistical errors are assigned in these tables.

namely, $V_0 = \frac{e^2}{\hbar}$.

The theoretical curves for PWBA are obtained from the listed values of $f_{L_n}(\Theta_{L_n}, \eta_L)$ calculated by Choi et al.⁴⁷, and those of CBEA are obtained from scaling the cross sections which were calculated by Hansen³⁴, using a proper momentum distribution for each L subshell.

PWBA is seen to describe the observations much better than CBEA, especially the energy dependence of the shape of each ionization cross section, for the elements which we studied. The overestimated absolute values predicted by PWBA have been noticed for a long time in the study of K shell ionization when the incident energy of the charged particle is low, i.e., the criteria of PWBA are not matched. As mentioned in Chapter I, two main corrections have to be made: (1) the binding change effect and (2) the Coulomb deflection effect. The criteria of these effects to be significant are discussed in detail by Basbas et al.⁵¹ who use perturbation theory to do a series expansion of the ionization cross section in terms of the strength of the Coulomb interaction between the incident charged particle and the atomic nucleus, and the strength of ionization Coulomb interactions between the incident charged particle and the atomic electrons, and conclude as follows: (1) Whenever

$$v/V_k \lesssim \Theta_k/2 \text{ (i.e., } \eta_k/\Theta_k^2 \lesssim 1/4 \text{)}, \quad (4.1)$$

where $V_k = Z_k V_0$ is the mean velocity of K-shell electron, the binding change effect is significant for K shell ionization. This simply states that the K-shell orbit will be distorted (the assumption of the unperturbed initial state of the atomic electron PWBA is broken) by the incident charged particle if the electron response time $1/\omega_k$ is comparative to particle-electron-interaction time $\approx a_k/v$, where $a_k = a_0/Z_k$

and a_0 is the Bohr radius. (2) Whenever

$$q_0^{-1} \leq \frac{Z_1 Z_2 e^2}{M_\mu V^2} \approx Z_1 \left(\frac{m}{M_\mu}\right) \left(\frac{V_k}{V}\right)^2 a_k, \quad (4.2)$$

the Coulomb deflection of the incident charged particle in the repulsive field of the target atomic nucleus must be significant and affect the K-shell ionization cross sections. Here, q_0 is the minimum momentum transferred from the impacting ion to the bound electron; m is the electron mass; and M_μ the reduced mass of the point charges $Z_1 e$ and $Z_2 e$ which undergo Coulomb interactions. The quantity $\frac{Z_1 Z_2 e^2}{M_\mu V^2}$ is equal to one-half of the distance of closet approach in a head-on collision of the Coulomb interaction between the point charges $Z_1 e$ and $Z_2 e$. The quantity designated q_0^{-1} is called the optimum penetration distance of the incident charged particle, since when the impact parameter is $\approx q_0^{-1}$, the ionization probability is the greatest. Therefore, Eq. (4.2) simply states that the optimum penetration distance is small compared to the one-half distance of the head-on collision of these two point charged particles. Under this condition, one immediately sees that the incident charged particle can be strongly distorted by the repulsive Coulomb field of the target atomic nucleus. The plane wave approximation of the incident charged particle in PWBA theory is therefore no longer valid. Eq. (4.2) can be alternatively written as

$$\left(\frac{v}{V_k}\right)^3 \leq \left(\frac{\Theta_k}{2}\right) Z_1 \left(\frac{m}{M_\mu}\right) \quad (\text{i.e.} \quad \frac{\eta_k^3}{\Theta_k^2} \leq \frac{1}{4} Z_1^2 \left(\frac{m}{M_\mu}\right)^2),$$

if we put $q_0 \approx \frac{I_0}{\hbar v}$ combined with the definition of Θ_k . One also notices that $M_\mu \approx M_1$ (the projectile mass), if $M_2 \gg M_1$ which is the cause of our study.

Fig. 18 σ_{L_1} , σ_{L_2} , and σ_{L_3} for Au, with PWBA and SCA curves; σ_{L_1} , σ_{L_2} , and σ_{L_3} are drawn from the tabulated values of the Tables 6 and 7. The statistic errors are assigned in these tables.

Applying these criteria to the L-shell, we notice that our energy range ($\eta_L \cong 0.002 - 0.04$, and $\eta_L/\Theta_L^2 \leq 0.1$) falls where the requirements of having the corrections of the binding effect and the Coulomb deflection effect are satisfied. We thus compared our Au data to the calculations of SCA (Fig. 18). The semiclassical picture, which includes Coulomb deflection by considering the path of the projectile to be a hyperbola instead of a straight line, has better agreement with the data for L_{III} subshell ionization cross sections at incident energy/a.m.u. ≤ 2.0 MeV for both proton and alpha-particle bombardments. For L_{II} ionization, the proton data below 2.0 MeV follows SCA, as does the L_{III} ionization. Deviations from SCA are observed for alpha energies below 1.0 MeV/a.m.u. The plot of $\sigma_{L_3}/\sigma_{L_2}$ ratios versus particle energies exhibits this feature in a more pronounced way (Fig. 23). We will discuss this deviation later. There are no SCA calculations available for Ta and Bi. However, from comparisons given above, the Coulomb deflection effect is sizable and predictable when the incident particle energy is below 2.0 MeV/a.m.u. When the energy is above 2.0 MeV/a.m.u. the ionization cross sections of L_{III} and L_{II} subshells are not estimated in the SCA calculations.

So far there is no evidence that the ionization cross section of L_I is well predicted by either CBEA or SCA. The general shape of the L_I ionization follows PWBA reasonably well, in the sense that both data and predictions of PWBA tend to consist of two smooth increasing curves whose junction are around $\eta_L/\Theta_L^2 \approx 0.01$. These not only strongly reveal the fact that a quantum mechanical treatment is better than other approaches (e.g. SCA and CBEA) but also shows more clearly that there is a strong correlation between the incident energy of the charged particle and the momentum distribution of the bound electrons in the momentum

space: The momentum distribution of a $2s_{1/2}$ electron has a node at $p = \frac{Z_L}{2a_0} \hbar$ according to a calculation based on a non-relativistic hydrogenic wave function²⁸ (Fig. 3), while those of $2p_{1/2}$ and $2p_{3/2}$ electrons are similar to that of $1s_{1/2}$ electrons which is without nodes except at $p = 0$. The gross behavior of $2p_{1/2}$ and $2p_{3/2}$ ionization thus are pretty much similar to that of $1s_{1/2}$ (K-shell) ionizations. The different form of the initial wave function of $2s_{1/2}$ state from those of the p states produces the sharp kink in the energy dependence of the ionization cross sections of L_I subshell. Basing his considerations on PWBA and the node of the momentum distribution of a $2s_{1/2}$ electron at $p = \frac{Z_L}{2a_0} \hbar$, Merzbacher¹ roughly estimated that the sharp change in the energy dependence of the cross section would be expected in the neighborhood of $\eta_L/\theta_L^2 \approx 0.06$. Qualitatively, this is indeed shown by our data. The other fact revealing the correlations between the particle-energy dependence of the cross section and the electron-momentum distributions in the momentum representation is that the ionization cross sections for the L_I subshell are larger than those for the L_{II} subshell at an energy below $\eta_L/\theta_L^2 = 0.01$, which relates to the larger amplitude of the momentum distribution of $2s_{1/2}$ electrons than that of $2p_{1/2}$ electrons in the higher-momentum portion ($p \gtrsim 3p_0 \frac{Z_L}{n}$, see Fig. 3). The two peaks of the momentum distribution of $2s_{1/2}$ electron (see Fig. 3) may thus contribute to the two smooth increasing curves seen in the energy dependence of the ionization cross section.

It would be much easier to understand the correlations between the incident ion energy and the initial electron wave function from the semiclassical picture proposed by Bang and Hansteen¹⁴. By introducing

the impact parameter, b , Bang and Hansteen find that the ionization is most likely to occur at $b \approx q_0^{-1}$ (q_0^{-1} is thus called "optimum penetration distance"). If this optimum penetration distance corresponds to a place where the electron has the least amplitude in its momentum distribution (and according to the Bohr atomic picture, one can grossly estimate the place where the electron has the least chance to be, i.e., $r = \frac{\hbar}{p}$ and $p = \frac{Z_L \hbar}{2a_0}$), the ionization cross section must change around there.

Therefore, the location of the "kink" in the energy-dependent ionization cross section could be estimated as

$$\frac{1}{q_0} \approx \frac{2a_0}{Z_L} \quad (4.3)$$

This immediately gives $\eta_L \approx \Theta_L^2/2^4$, i.e. $\eta_L/\Theta_L^2 \approx 0.06$. All the above discussions show that when the incident energy is decreasing, the momentum of the electrons which are most likely to be ionized is increasing or vice versa. Apparently, in our energy range, the majority of the $2s_{1/2}$ electrons to be ionized fall in the vicinity of the node of the momentum distribution.

Qualitatively, PWBA describes the observations quite well.

Quantitatively, substantial deviations between data and theory begin at the kink. There is sizable evidence of the tendency of the upper smooth increasing curve to extend downwards to the low energy side of the position of the kink predicted by PWBA (i.e. the junction point of the two smooth curves in the data moves to the lower energy than that of PWBA). This simply indicates that the node position of the momentum distribution

of the $2s_{1/2}$ electron moves upwards to the higher momentum side than what was predicted by a non-relativistic hydrogenic wave function. This, in turn, implies that the non-relativistic hydrogenic wave function used in PWBA is not adequate. In fact, for a heavy atom, the relativistic effect of the bound electron has been found to be important^{26, 33} and use of Hartree-Fock-Slater wave functions may produce effects which are also sizable for L-shell electrons, especially in the region where there is a node⁵². From the comparison of hydrogenic and Hartree-Fock $\ell = 1$ radial eigenfunctions for mercury⁵², it is seen that the effect of using Hartree-Fock-Slater wavefunctions does exhibit the tendency to move the node upwards to the high-momentum side. In order to investigate these two effects, we compared the ratio $\sigma_{L_1}/\sigma_{L_2}$ of our data to those predicted by PWBA. The Coulomb deflection effect and binding effect, which are significant at the low energies as discussed at the beginning of this chapter, could be eliminated, since they affect each subshell in a similar manner. Fig. 19-21 show the charged-particle energy dependence of these ratios for Ta, Au, and Bi, respectively. The valleys of these ratios reflect the location of the node in the momentum distribution of the $2s_{1/2}$ electrons. Again these ratios are qualitatively described by non-relativistic PWBA quite well. The gradual shift of this valley in non-relativistic PWBA from that of the data in Ta($Z = 73$) to higher energies in Bi($Z = 83$) may be mainly due to the larger shift of the zero-amplitude momentum position of the non-relativistic hydrogenic momentum distribution of the $2s_{1/2}$ electron of higher Z elements toward the lower momentum position than that based on the actual wave functions (See Fig. 3)

Fig. 19 The $\sigma_{L_1}/\sigma_{L_2}$ ratio for Ta.

Fig. 20 The $\sigma_{L_1}/\sigma_{L_2}$ ratio for Au.

Fig. 21 The $\sigma_{L_1}/\sigma_{L_2}$ ratio for Bi.

The relativistic calculations were terminated at $\eta_L \approx 0.01$; no calculations so far have appeared for energies higher than that. However, the relativistic hydrogenic wave function indeed produces part of the necessary correction at $\eta \leq 0.01$, as indicated by many authors^{26,33,46}. We also show in Fig. 22, that the cross-over points of $\sigma_{L_1}(E)$ and $\sigma_{L_2}(E)$, and of $\sigma_{L_1}(E)$ and $\sigma_{L_3}(E)$, are much closer to the prediction of relativistic PWBA than that of non-relativistic PWBA. Nevertheless, the "Hartree-Fock-Slater effect" could also be important, since the valley of $\sigma_{L_1}/\sigma_{L_2}$ of our data indicates the exact place which corresponds to the position of the node based on Hartree-Fock-Slater wave functions rather than to the node of non-relativistic PWBA wave functions.

A Hartree-Fock-Slater type of calculation thus is very important and suggestive at this point to clarify which effect dominates the shift of the valley of the $\sigma_{L_1}/\sigma_{L_2}$ ratios. Relativistic calculations cannot be carried beyond $\eta_L \approx 0.01$ because of difficulties in divergence of the integration.

4.B. $\sigma_{L_3}/\sigma_{L_2}$ Ratios

This Hartree-Fock-Slater effect is negligibly small for σ_{L_2} and σ_{L_3} because the wave functions having no nodes, such as those of $2p_{1/2}$ and $2p_{3/2}$, and are relatively well described by a hydrogenic wave function. One expects that the energy dependence of the $\sigma_{L_3}/\sigma_{L_2}$ ratios can thus be predicted much better than the $\sigma_{L_1}/\sigma_{L_2}$ ratios by PWBA. Figure 23 shows these ratios. The vertical uniform displacement between data and theory may be partly due to the atomic parameters such as ω 's and F 's involved in the data analysis. The relativistic correction also tends

Fig. 22 Au L subshell ionization cross sections at low energies, with non-relativistic PWBA and relativistic PWBA curves.

Fig. 23 The $\sigma_{L_3}/\sigma_{L_2}$ ratio of Ta, Au, and Bi.

to lower the theoretical curve^{56,46}. It can also be seen in Fig. 22 that relatively larger corrections must be made for σ_{L_2} than for σ_{L_3} due to the relativistic effect at $\eta_L \leq 0.01$, which will move the theoretical curve of the $\sigma_{L_3}/\sigma_{L_2}$ ratio downwards toward the data. The dramatic drop of the ratio in the α data at lower energies has been seen by Datz et al.⁴⁶, who measured only Au. However, relatively little evidence of a corresponding drop in the α data in Ta and Bi is observed. The explanation of this dropping as due to a multionization effect⁴⁶ does not seem favorable, since the multionization effect should affect all elements in this narrow range of atomic number in a similar fashion. The agreement between the deduced L_β X-ray yields and the measured L_β X-ray yields (as shown in Fig. 12) shows that this multionization effect is negligibly small; the L_β X-ray line is a complex of the decays from all three L subshells and the calculated L_β X-ray yields result from the same atomic parameters used in deducing σ_{L_1} , σ_{L_2} , and σ_{L_3} from the L_α and L_γ X-rays (see Chapter III, section A).

Two explanations can be conceived in addition to a multionization effect, namely an outer electron configuration effect and a nuclear Coulomb excitation effect. The latter is unfavorable, as noted earlier, since the cross section for direct atomic ionization of the L shell is much larger than that of the nuclear Coulomb excitation. The former is a possible one, since the outer electron configurations of tantalum and bismuth are $6s^2 5d^3$ and $6s^2 6p^3$ respectively, while that of gold is $6s^1 5d^{10}$. In some way, perhaps the single 6s electron in gold might be affecting the L-subshell ionization results in a way which differs from what occurs with a complete 6s shell as is the case for the other two elements. However, the mechanism of the ionization process involving

this effect remains unknown and need further investigations (see Appendix B for details, including a preliminary test using a Pt target.)

4.C. Conclusions

As discussed above, the features of L-subshell ionization of a heavy atom by a light ion can generally be described through a Coulomb interaction in the framework of PWBA. Through the study of $2s_{1/2}$ ionizations finer details of the ionization process are revealed. These details suggest the necessity of further investigations, especially of the Hartree-Fock-Slater effect, which could contribute a sizable effect in the heavy elements, along with the relativistic effect. A non-relativistic PWBA calculation using Hartree-Fock-Slater wave functions to indicate the quantitative shift of the valley of the $\sigma_{L_1}/\sigma_{L_2}$ ratio would be instructive and illuminate the usage of light ions as a probe to investigate the characteristics of the electrons in the deep territory of an atom.

The features of the $\sigma_{L_3}/\sigma_{L_2}$ ratio at low energies of alpha-particle bombardment are also very interesting, and may end up producing new physical insight. However, more experimental information is needed before any conclusions can be made. Such information could include detailed high-resolution L X-ray spectra (to be sure that additional peaks are not interfering with the data-reduction assumptions made on the present study), and energy-dependence measurements on atoms around the closure of other s subshells, such as 5s (to see if the apparent dependence persists). We look forward to such measurements as a continuation of the work developed during the course of the present study.

APPENDIX A

K-L X-RAY ON-LINE COINCIDENCE MEASUREMENT

Whenever an electron vacancy is created in the K shell, an outer-shell electron will drop down to fill the vacancy and result in fluorescence K X-rays or Auger electron emission. Approximately 90% of the electrons come from the L shell; the L_{III} electrons give a K_{α_1} X-ray and the L_{II} electrons give K_{α_2} X-rays, competing with Auger electron emission. Thus K-shell transitions created electron vacancies in the L_{II} and L_{III} subshells, which in turn give rise to L X-rays. This cascade of transitions produces an enhancement of the L X-ray yield which does not arise from the direct ionization of L subshells, and hence could distort our results for these ionization cross sections. We therefore decided to look for such coincidences, in an attempt to measure this correction factor. Source K-L coincidence experiments have been made to measure the fluorescence yields of L_{II} and L_{III} (i.e. the values of ω_2 and ω_3)⁵³ and to study the multiple transition possibility from a K-L angular correlation experiment⁵⁴. On-line coincidence experiments have not been reported so far.

There are at least two things that can be determined from an on-line K-L coincident measurement: (1) the relative possibility of ionizing K shell electrons to that of L shell by the incident charged particle, (2) from the difference of the coincident and non-coincident of L X-ray spectrum, if any, to determine the energy dependence or ion

dependence of the fluorescence yields ω_2 and ω_3 .

Since the ionization cross section of any L subshell is much larger than the ionization cross section of the K shell, the L X-rays provide a huge random-coincidence background. The true coincidence events are therefore difficult to observe. A more sophisticated arrangement of the electronics is needed to suppress this background. However, a simple, conventional fast-slow coincidence circuit (Fig. A1) provides us the upper limit values of the K shell ionization compared to L shell ionization.

The coincidence electronics was lined-up by using a ≈ 2.0 mCi ^{57}Co source which has a mode of disintegration via electron capture (99.8%) to the 136.3 KeV level of ^{57}Fe (Fig. A2). The iron 6.4 K X-ray and 121.9 KeV γ -ray, which were recorded in the Si(Li) detector and a NaI(Tl) x-ray detector, respectively, provide a good coincidence reference. The time spectrum is shown in Fig. A3. The first spectrum is the time spectrum from a time-to-pulse-height converter (TAC) without any energy requirements; the second spectrum was routed by the slow overlap-coincidence signal of the gated iron K X-rays and 122 keV γ X-ray. The peak to background ratio is obviously improved for the routed time spectrum. The resolving time was found to be ≈ 20 nsec. In the actual on-line experiments, we bombarded an Au target with 3.0 MeV protons. A triangular target holder was used, in order to tilt the target in such a way that the low energy X-rays can get out of the two mylar windows perpendicular to the beam line as previously described (see Chap. II). K X-rays of Au were detected by the NaI(Tl) X-ray detector, which has

Fig. A1 Electronics block diagram for K - L X-ray coincidence measurement.

Fig. A2 ^{57}Co decay scheme.

Fig. A3 Time spectrum of the γ ray - K x-ray in coincidence. The spectrum in the first 128 channels is the time spectrum without routing signals, while the spectrum in the second 128 channels is the time spectrum which was routed by the slow overlap coincident pulse of the gated γ ray and K x-rays of ^{57}Fe .

overall resolution = 44.3% for 5.9 keV photons; and L X-rays were detected by the Si(Li) detector. The target-detector distance was adjusted such that optimum total counting rates for these two detectors were obtained. During this experiment, both the K X-rays and the L X-rays were gated with single-channel analyzers and input to the slow coincidence unit (ORTEC 418A) and the slow overlap-coincidence signal was used to route the time spectrum to limit some of the backgrounds. No evidence of true coincident events was observed.

The following records provide an estimate of the upper-limit of the K shell ionization probability relative to L shell ionization probability:

Count rate of the gated L X-rays of Si(Li) detector, $N_L = 78.9$ counts/sec.

Count rate of the gated K X-rays of NaI(Tl) X-ray detector, $N_K = 47.5$ counts/sec.

K-L X-ray slow overlap coincidence count rate $N_A = 4.89 \times 10^{-3}$ counts/sec.

The calculated random coincidence count rate is equal to

$$\begin{aligned} N_R &= N_L N_K (2\tau) = 78.9 \times 47.5 \times 2 \times 5.0 \times 10^{-6} \\ &= 3.745 \times 10^{-3} \text{ (counts/sec)} \end{aligned}$$

The resolving time used here is the width of the input logic pulse of the coincidence unit. The true coincident count rate, $N_T \leq N_A - N_R = 1.2 \times 10^{-3}$ counts/sec and $N_T/N_L \leq 1.5 \times 10^{-5}$. This indicates that the ratio of L X-rays due to K-shell ionization to L X-rays caused by direct L-shell ionization is of the order of magnitude 10^{-5} . After corrections for the fluorescence yields of K and L shell were made, this value is

comparable to the ratio of the K-shell ionization cross section to the L shell ionization cross section measured by Bernstein and Lewis as 1.37×10^{-4} at proton energy 2.5 MeV⁴³.

APPENDIX B

APPARENT DEPENDENCE OF α -PARTICLE-INDUCED L SUBSHELL IONIZATION ON OUTER ELECTRON CONFIGURATIONS

(This appendix, giving more details of the $\sigma_{L_3}/\sigma_{L_2}$ effect described in Chapter IV, is adapted from a paper submitted for publication by the present author, with J. F. Morgan and S. L. Blatt. A separate list of references follows.)

In the past several years a wealth of information has been published on K-shell ionization cross sections for positively charged particles. Recently data has begun to be available on the L-shell ionization cross sections. Although the earliest data^{1,2)} were for total L-shell cross sections, with the advent of higher resolution Si(Li) detectors, and the use of spectrum stripping techniques, it has become feasible to measure the ionization cross sections for the individual L-subshells. The first published work on L-shell cross section measured in this manner was the work of Datz et al.³⁾ who studied L-shell cross sections for protons and alpha particles incident on gold.

The most interesting of the results of Datz et al. was to be noted in the behavior observed in the ratio of the L_{III} to L_{II} cross sections. For low energy protons the $\sigma_{L_3}/\sigma_{L_2}$ ratio is in qualitatively good agreement with the plane-wave Born approximation (PWBA), following

the energy dependence of the PWBA quite well but differing in overall normalization by about a factor of 0.7. The behavior for low energy alpha particles was found, however, to be strikingly different. As the alpha particle energy is lowered, the $\sigma_{L_3}/\sigma_{L_2}$ ratio shows a dramatic drop from both the proton ratio at the same projectile velocity and the PWBA calculation (see Fig. B1). Datz et al. suggested that an explanation for this drop might be found in the phenomena of multiple ionization since low velocity alpha particles are known⁴⁾ to induce more multiple-ionization than are protons, and hence the use of published values of fluorescence yields and transition strengths may produce estimates which are badly in error for some of the observed transitions.

At the same time the results of Datz et al. became available, we were conducting a systematic study of L-subshell cross sections in heavy elements. The overall results of that study, as well as the full experimental details, will be published elsewhere.⁵⁾ In this letter we wish to report on our findings for the $\sigma_{L_3}/\sigma_{L_2}$ ratios in some of these events.

The experimental set up was conventional. Proton and alpha particle beams from the OSU-CN Van de Graaff were incident on thin foils of tantalum, gold and bismuth ($\leq 100 \mu\text{gm}/\text{cm}^2$). The L X-rays were detected, at 90° to the incident beam, with Si(Li) detector with an overall resolution of 250 eV at 5.9 keV. Target stability and beam integrations were monitored by observing the nuclear elastic scattering with a silicon surface barrier detector mounted at 45° to the beam. In all cases the elastic cross section was well described by the Rutherford formula, thus indicating that no target thickness changes or beam

Fig. B1 Ratio of the ionization cross sections for the σ_{L_3} and σ_{L_2} subshells induced by protons and alpha particles. The solid line gives the results from a PWBA calculation,

integration irregularities occurred during the experimental runs. The L_γ portion of the X-ray spectrum was fitted with a Gaussian peak-fitting program to separate the components of L_I and L_{II} transitions. These were then combined with published values of the fluorescence yields⁶⁾, Coster-Kronig transition probabilities⁶⁾ and the partial transition widths⁷⁾ of the particular radiative transition in each L subshell to yield the individual subshell ionization cross section. The L_{III} cross section was obtained from the L_α line

The results for targets of Ta, Au, and Bi are shown in Fig. B1, where the circles and squares are the proton data and the triangles are the alpha particle data. The gold data is in excellent agreement with the work of Datz et al. not only qualitatively, but also in the quantitative values of the $\sigma_{L_3}/\sigma_{L_2}$ ratio. The data for tantalum and bismuth, which bracket gold in both A and Z, are dramatically different, giving no evidence of the downturn for low alpha energies.

It is difficult to explain this result on the basis of multi-ionization which should affect all elements in this narrow range of atomic number in a similar fashion. However, such an explanation could be checked easily: we used our deduced L-subshell ionization cross sections, σ_{L_1} , σ_{L_2} , and σ_{L_3} , to calculate the expected yield of L_β X-rays (assuming the same atomic parameter used in deducing σ_{L_1} , σ_{L_2} , and σ_{L_3} from the L_α and L_γ x-rays). The calculated L_β yield agrees very well with the values actually observed. These results lead us to conclude there is little or no effect due to multi-ionization effect on any of these cross sections.

If the low energy portion of the $\sigma_{L_3}/\sigma_{L_2}$ ratio cannot be explained by multiple ionization what can be the cause? Two explanations were conceived. The first argues that the effect is due to the atomic structure of the atoms. The outer configuration of tantalum and bismuth are $6s^25d^3$ and $6s^26p^3$ respectively, while that of gold is $6s^15d^{10}$. In some way, perhaps the single 6s electron in gold might be affecting the L-subshell alpha-particle ionization results in a way which differs from what occurs with a complete 6s shell as is the case for the other two elements.

Another explanation is that the effect might be related to nuclear Coulomb excitation, a process whose contribution increases with lower projectile velocity, followed by de-excitation through internal conversion. In favorable cases this process is known to contribute substantially to observed X-ray yields; for example, there is a contribution of approximately 20% to yield of K X-rays for 2.0 MeV protons on a Ta^{181} target⁸⁾. The internal conversion coefficients for the first excited state of Au^{197} (77.3 keV) do show some difference in the probability of converting L_{II} electrons and L_{III} electrons⁹⁾. However, the nuclear Coulomb excitation cross sections is not comparable to the large ionization cross section for direct atomic excitation of the L subshells, so that this possibility of explaining the striking differences in the alpha induced cross section data does not seem favorable.

If the first explanation is correct, the low-energy drop off seen in Au should also be seen in platinum, whose outer shell structure ($6s^15d^9$) resembles that of gold, both having only a single 6s electron. Therefore similar measurements were made with α -particles on Pt. The

Fig. B2 Ratio of the σ_{L_3} to σ_{L_2} cross sections induced by
alpha particles on Pt.

$\sigma_{L_3}/\sigma_{L_2}$ results are shown in Fig. B2. As can be seen from the figure, the results show the same pronounced drop at low projectile velocities as previously observed in gold.

What mechanism can account for this effect, if it is indeed due to the closed or open character of the 6s shell? Several factors may enter. It has been shown¹⁰⁾ that the probability for low-energy ions to pick up outer-shell electrons is different when some of the subshells are not completely filled. This effect is expected to be larger for α -particle projectiles than for protons. Further, α -particles in different charge states may have different probabilities for causing inner-shell ionization. A complete explanation awaits additional experiments. However, the existence of a strong relationship between outer-shell configurations and inner shell configurations and inner-shell ionization seems to be the only clear way to describe the observations made thus far. Experiments to further clarify this effect are being planned.

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