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L-subshell-ionization cross sections of tungsten by electron impact near the threshold region

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L-subshell-ionization cross sections σ_3/σ_2 and σ_2/σ_1 for L_3, L_2, and L_1 levels for tungsten have been measured by electron impact at incident electron energies from about one to three times the ionization threshold energy of the L_1 subshell. Ionization cross sections for the L_1, L_2, and L_3 levels were deduced from the measured intensity radiations $L\beta_3$, $L\beta_1$, and $L\beta_2$ and using known values for the fluorescence yields, radiative transition probabilities, and Coster-Kronig parameters. The x-ray spectra were obtained with a curved crystal spectrometer (50 cm radius) and recorded on photographic film. Comparison is made with experimental measurements of Chu-Nan Chang and theoretical calculations of Gryzinski and McGuire. A good agreement is achieved for electron-beam energies $E_0 \geq 1.25$ times the energy E_1 of L_1-level ionization.

I. INTRODUCTION

This paper is concerned with cross sections for the removal of L_1, L_2, and L_3 electrons by electron impact. In the past years considerable effort has been made to study inner-shell vacancies production. Much of this work has been devoted to studies of heavy-particle collisions. As a result, our understanding of inner-shell ionization and excitation processes by heavy ion and proton impact is now well developed and the theoretical models are largely accepted. However, there are very few data on ionization cross sections by electron impact. Theoretical treatments of inner-shell ionization by electrons have been performed by Burhop,[1] Peach,[2] Omidvar *et al.*,[3] McGuire,[4] Worthington and Tomlin,[5] and Gryzinski.[6] Burhop's calculation is for silver and mercury, whereas Peach and Omidvar *et al.* have carried out calculations for low-Z numbers and mostly for outer shells. McGuire has done quite extensive calculations for a large variety of Z numbers and of inner shells. He presents his calculations in a scaled form $f(E_0/E_i)$; for sufficiently large Z numbers he considered a nonclassical scaling law of the type

$$\sigma_i E_i^{\alpha} = f(E_0/E_i) \ . \tag{1}$$

The symbols E_0, E_i, and σ_i denote, respectively, the incident electron energy, the ionization energy for the i shell, and the cross section of a given i shell. The values of $f(E_0/E_i)$ for electrons $2s$ and $2p$ correspond to $E_0 \geq 1.25E_i$.

Based on the first Born approximation, Bethe[7] expressed the cross section of the i shell by the following relation:

$$\sigma_i E_i^2 U_i = 6.51\times 10^{-20} a_i b_i \ln(c_i U_i) \ \text{cm}^2\ \text{keV}^2 \ , \tag{2}$$

where $U_i = E_0/E_i$, a_i, c_i are the Bethe parameters and b_i denotes the number of electrons in the i shell. Since this equation is based on the first Born approximation, its validity is limited to the region of $U_i \gg 1$.

Worthington and Tomlin modified the logarithmic term to give a plausible representation of the ionization cross section near the threshold. They obtained the following equation:

$$\sigma_i E_i^2 U_i = 6.51\times 10^{-20} a_i b_i \ln\left[\frac{4U_i}{1.65+2.35\exp(1-U_i)}\right] \text{cm}^2\ \text{keV}^2 \ . \tag{3}$$

The attainment of agreement between observed and Worthington and Tomlin cross sections depends on the choice of the parameter a_i.

The classical theory of inner-shell ionization that has appeared to be the most successful is that of Gryzinski. In this binary collision model the main idea is that the dominant interaction producing the transition is a direct energy exchange between the incident charged particle and the bound electron. The collision is considered as instantaneous and viewed as the collision of the incident particle of momentum k_1, with a free electron of momentum k_2. With these assumptions Gryzinski obtained the following expression:

$$\sigma_i = b_i(\sigma_0/E_i^2) g_i(x) \ , \tag{4}$$

where

$$g_i(x) = \frac{1}{x}\left[\frac{x-1}{x+1}\right]^{3/2}\left[1+\frac{2}{3}\left[1-\frac{1}{2x}\right] \times \ln[2.7+(x-1)^{1/2}]\right] ,$$

TABLE I. (a) Tungsten atomic yields and Coster-Kronig parameters from Krause (Ref. 17). (b) Tungsten radiative probability transitions from Scofield (Ref. 16) and shake-off probabilities from Parente *et al.* (Ref. 20).

(a)

ω_1	ω_2	ω_3	f_{12}	f_{13}	f_{23}
0.147	0.270	0.255	0.17	0.28	0.133

(b)

$p^R_{L_1}$	$p^R_{L_2}$	$p^R_{L_3}$	$p^R_{L_1\to M_3}$	$p^R_{L_2\to M_4}$	$p^R_{L_3\to N_5}$	P_1
0.805	1.397	1.244	0.330	1.138	0.1593	0.339

$$\sigma_0=6.56\times10^{-14}\ \text{cm}^2\ ,$$

$$x=E_0/E_i\ .$$

We can see that there is no general theory applicable to all elements and all energies of the electron beam. Experiments on inner-shell ionization by electronic impact have mostly been restricted to K-shell ionization; very few results are available for the L shell. To our knowledge, L-shell-ionization cross sections by electron impact have been measured for heavy elements by Huizinga,[8] Salem and Moreland,[9] and Shima *et al.*,[10] using electrons with energies up to several times the L ionization energy.

Measurements have also been made using low-energy electrons for tungsten by Chu-Nan Chang[11] and by Salgueiro *et al.*[12] and for gold by Ramos *et al.*[13] L x-ray cross sections using relativistic energy electrons have been measured by Middleman *et al.*[14] for Tm, Ta, Au, and Bi and by Park *et al.*[15] for elements ranging from Ba to Bi.

II. DETERMINATION OF CROSS SECTIONS

The ionization cross section σ_i for a given L_i subshell ($i=1,2,3$) may be obtained from the intensities of a given radiative x-ray transition if we know the fluorescence yields ω_i, the Coster-Kronig transition parameters f_{jk} ($j=1,2$: $k=2,3$; $j\neq k$), the shake-off probabilities P_i, and the radiative probability transitions p^R to the L subshell. The L-subshell radiative transitions are very small and have been neglected (Scofield[16]). To evaluate the transition cross sections, the $L\beta_3$, $L\beta_1$, and $L\beta_2$ radiations, corresponding, respectively, to the transitions $L_1\to M_3$, $L_2\to M_4$, and $L_3\to N_5$, have been used. The cross sections are obtained from

$$\sigma_1=\frac{F_{L\beta_3}}{\omega_1}\frac{p^R_{L_1}}{p^R_{L_1\to M_3}}\ , \tag{5}$$

$$\sigma_2=\frac{F_{L\beta_1}-\sigma_1 f_{12}\omega_2\left[(1-P_1)+\dfrac{P_1}{1-f_{23}}\right]\dfrac{p^R_{L_2\to M_4}}{p^R_{L_2}}}{\omega_2\dfrac{p^R_{L_2\to M_4}}{p^R_{L_2}}}\ , \tag{6}$$

$$\sigma_3=\frac{F_{L\beta_2}-\omega_3[\sigma_1 f_{13}+\sigma_2 f_{23}+\sigma_1 f_{12} f_{23}(1-P_1)]\dfrac{p^R_{L_3\to N_5}}{p^R_{L_3}}}{\omega_3\dfrac{p^R_{L_3\to N_5}}{p^R_{L_3}}}\ , \tag{7}$$

where $F_{L\beta_1}$, $F_{L\beta_2}$, and $F_{L\beta_3}$ are the number of x-ray photons of $L\beta_1$, $L\beta_2$, and $L\beta_3$ radiations, respectively, obtained from the corresponding intensities measured in the present work. The number of photons $F\beta_i$ considered include the diagram lines and the satellite lines (double and triple ionized atoms).

From Eqs. (5), (6), and (7) we obtained

$$\frac{\sigma_3}{\sigma_2}=\frac{\dfrac{F_{L\beta_2}}{F_{L\beta_1}}\omega_2\dfrac{p^R_{L_2\to M_4}}{p^R_{L_2}}-f_{23}\omega_3\dfrac{p^R_{L_3\to N_5}}{p^R_{L_3}}+A-B}{\omega_3\dfrac{p^R_{L_3\to N_5}}{p^R_{L_3}}}\ , \tag{8}$$

where

$$A=\frac{F_{L\beta_2}}{F_{L\beta_1}}\frac{\sigma_1}{\sigma_2}\frac{p^R_{L_2\to M_4}}{p^R_{L_2}}f_{12}\omega_2\left[(1-P_1)+\frac{P_1}{1-f_{23}}\right]$$

TABLE II. Relative intensity ratios of the radiations $L\beta_2$, $L\beta_1$ and $L\beta_1$, $L\beta_3$ and corresponding ratios of the number of x-ray photons, for energies E_0 of an electron beam, from 12.5 to 40 keV.

E_0 (keV)	$\frac{I_{L\beta_2}}{I_{L\beta_1}}$	$\frac{I_{L\beta_1}}{I_{L\beta_3}}$	$\frac{F_{L\beta_2}}{F_{L\beta_1}}$	$\frac{F_{L\beta_1}}{F_{L\beta_3}}$
12.5	1.33±0.11	25.4±3.2	1.29	25.8
12.7	1.19±0.11	18.8±2.4	1.15	19.1
12.8	1.13±0.10	15.4±1.9	1.10	15.6
12.9	1.06±0.10	12.7±1.6	1.03	12.9
13	0.99±0.09	11.4±1.1	0.96	11.6
13.5	0.86±0.08	9.5±1.2	0.83	9.6
14	0.77±0.07	8.2±0.3	0.75	8.3
14.5	0.70±0.07	7.2±0.45	0.68	7.3
15	0.64±0.06	6.6±0.5	0.62	6.7
16	0.59±0.06	6.4±0.4	0.57	6.5
17	0.59±0.06	5.5±0.3	0.57	5.6
20	0.51±0.05	5.2±0.9	0.495	5.3
25	0.49±0.05	4.8±0.4	0.476	4.87
30	0.48±0.04	4.8±0.4	0.466	4.87
35	0.47±0.04	4.8±0.4	0.456	4.87
40	0.46±0.04	4.8±0.4	0.447	4.87

TABLE III. Tungsten cross-section ratios of L_3/L_2-subshell (σ_3/σ_2) results of Gryzinski, McGuire, Chu-Nan Chang, and those of the present work, for incident electrons of energy E_0.

E_0 (keV)	Gryzinski	McGuire	Chu-Nan Chang	Present work
12.5	9.6			8.7
12.7	8.1			7.9
12.8	7.6			7.5
12.9	7.2			6.9
13	6.8			6.6
13.5	5.6			5.8
14	4.9			5.2
14.5	4.4	4.5		4.7
15	4.1	4.2		4.3
16	3.7	3.8		4.0
17	3.5	3.8	3.4	4.0
20	3.1	3.2	3.8	3.4
23			3.4	
25	2.8	2.9		3.3
26			3.2	
29			3.5	
30	2.7	2.7		3.2
35	2.6	2.7	3.0	3.1
40	2.6	2.6	2.8	3.1

and

$$B=\frac{\sigma_1}{\sigma_2}\frac{p^R_{L_3\to N_5}}{p^R_{L_3}}\omega_3[f_{13}+f_{12}f_{23}(1-P_1)]\ ,$$

$$\frac{\sigma_2}{\sigma_1}=\frac{\frac{F_{L\beta_1}}{F_{L\beta_3}}\omega_1\frac{p^R_{L_1\to M_3}}{p^R_{L_1}}-f_{12}\omega_2\left\{(1-P_1)+\frac{P_1}{1-f_{23}}\right\}\frac{p^R_{L_2\to M_4}}{p^R_{L_2}}}{\omega_2\frac{p^R_{L_2\to M_4}}{p^R_{L_2}}}\ . \qquad (9)$$

Values of ω_i and f_{ij} are from Krause;[17] $p^R_{L_i}$, $p^R_{L_1\to M_3}$, $p^R_{L_2\to M_4}$, $p^R_{L_3\to N_5}$ are from Scofield.[16] These values are displayed in Table I. We assumed, as was proved by Kinsey[18] and adopted by Ross[19] *et al.*, that the radiative probability transitions are the same for atoms with one, two, or three vacancies. The shake-off probability P_1 (Table I) is obtained from Parente *et al.*[20] As far as we know this is the first time that the shake-off probabilities have been considered.

III. EXPERIMENTAL PROCEDURE

Spectra were obtained with an x-ray tube with a beryllium window, 1 mm in thickness, and an anode of tungsten, by means of electron-beam bombardment. The electron-beam energies are measured with a very accurate digital voltmeter. The radiations were analyzed with a curved crystal spectrometer, having a quartz crystal [$d=1811$ Å, $(11\bar{2}2)$ planes], 2 mm in thickness, bent to a cylinder of 50-cm radius.

The energies of the $L\beta_1$, $L\beta_2$, and $L\beta_3$ lines of tungsten are 9.672 35, 9.9615, and 9.8188 keV; according to the different energies of the lines, corrections for different absorption on air, beryllium window, crystal,

TABLE IV. Tungsten cross-section ratios of L_2/L_1-subshell (σ_2/σ_1) results of Gryzinski, McGuire, Chu-Nan Chang, and those of the present work, for incident electrons of energy E_0.

E_0 (keV)	Gryzinski	McGuire	Chu-Nang Chang	Present work
12.5	4.1			6.9
12.7	3.0			5.0
12.8	2.7			4.0
12.9	2.5			3.3
13	2.3			3.0
13.5	1.8			2.4
14	1.6			2.1
14.5	1.5			1.8
15	1.4		1.4	1.6
16	1.3	1.2		1.5
17	1.3	1.1	1.4	1.3
20	1.2	1.2	1.2	1.2
23			1.1	
25	1.1	1.2		1.1
26			1.3	
29			1.2	
30	1.1	1.2		1.1
35	1.1	1.3		1.1
40	1.1	1.3		1.1

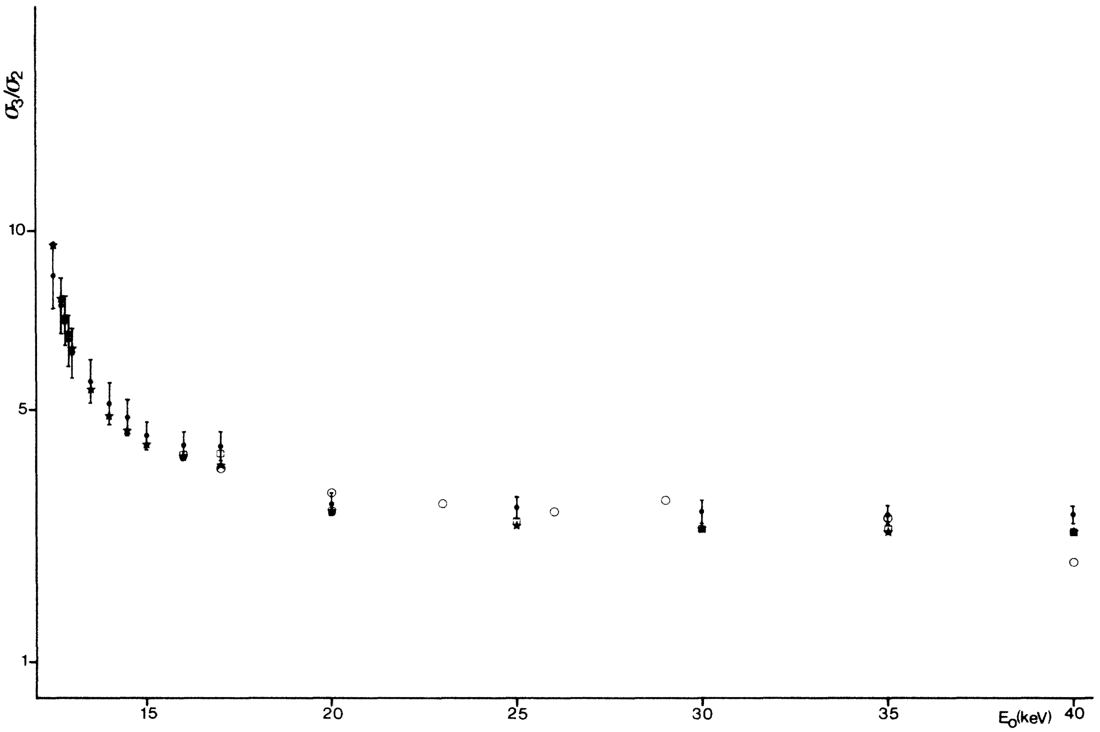

FIG. 1. Comparison of cross-section ratios σ_3/σ_2 for tungsten as a function of the energy of the incident electrons. * and □, theoretical results of Gryzinski and McGuire, respectively; ○ and ⧳, experimental results of Chu-Nan Chang and of the present work, respectively.

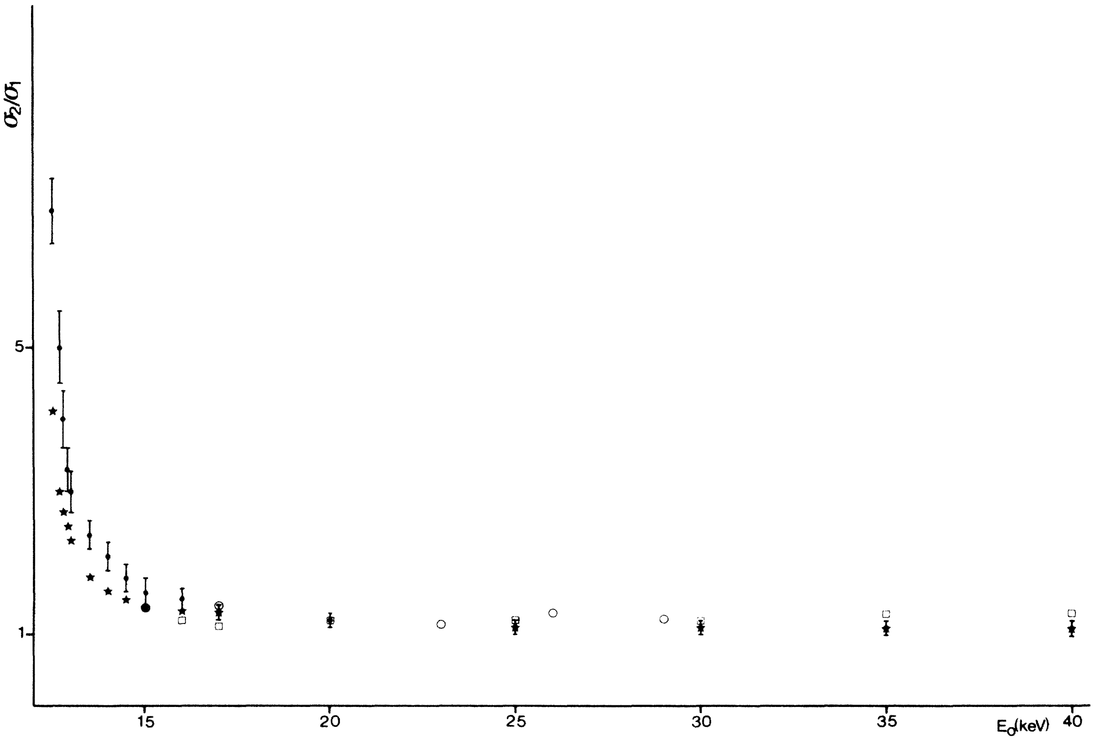

FIG. 2. Comparison of cross-section ratios σ_2/σ_1 for tungsten as a function of the energy of the incident electron. * and □, theoretical results of Gryzinski and McGuire, respectively; ○ and ⧳, experimental results of Chu-Nan Chang and of the present work, respectively.

and self-absorption on the anode itself have been made.

The spectra were recorded on photographic Kodirex film, single coated, and analyzed by means of a Joyce-Loebl microdensitometer; a resolution of 0.16% was achieved. The density range was determined by a previous calibration curve of the film.

IV. EXPERIMENTAL RESULTS AND CONCLUSIONS

In Table II we present our experimental results for the ratios $I_{L\beta_2}/I_{L\beta_1}$, $I_{L\beta_1}/I_{L\beta_3}$, and the corresponding values of $F_{L\beta_2}/F_{L\beta_1}$, $F_{L\beta_1}/F_{L\beta_3}$, of tungsten for different energy values. The present values of σ_3/σ_2 and σ_2/σ_1 are compared with the theoretical values of McGuire and Gryzinski and with the experimental values of Chu-Nan Chang. The errors that affect the present results are estimated to be 10% due to the experimental errors of intensity measurements and to the uncertainty in the theoretical parameters used at present. We display in Table III and Fig. 1 the relative ionization cross sections σ_3/σ_2. This ratio varies slowly with the energy of the projectile and there is a good agreement between theoretical and experimental values, according to the results previously reported (Salgueiro *et al.*[12]).

In Table IV and Fig. 2, σ_2/σ_1 ratios are shown as a function of the energy of the electron beam. The values obtained by Gryzinski do not agree with our experimental results for energies of the electron beam near the threshold of the L_1 level. This is not surprising if one realizes that the calculations of Gryzinski are in a classical approximation.

We can see that as the energy increases a good agreement is achieved. The results of McGuire are consistent with experimental values obtained by Chu-Nan Chang and the present work. These results are also expected if we consider the fact that the calculations of McGuire were made for values of energy E_0 greater than or equal to $1.25E_i$. As far as we know the ratio σ_2/σ_1 has not been determined before for energies just above the L_1 threshold. We conclude that more theoretical calculations and experimental values are needed in the low-energy region for full exploitation of the ionization cross sections.

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