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L-SHELL IONIZATION MEASUREMENT OF TUNGSTEN BY ELECTRON IMPACT*

Peng Xiu-feng(彭秀峰), He Fu-qing(何福庆), Long Xian-guan(龙先灌),
An Zhu(安 竹), and Luo Zheng-ming(罗正明)
State Key Laboratory for Radiation Physics and Technology of Education Ministry of China,
Institute of Nuclear Science and Technology, Sichuan University, Chengdu 610064, China

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L-shell partial production cross sections of $L_{\alpha-}$, $L_{\beta-}$, $L_{\gamma-}$rays by electron impact were measured by observing the counts of X-ray from impacted thin tungsten target. Total production cross sections and mean ionization cross sections were deduced from these measured results. The electron beam energy range was from 11 to 36 keV. Tungsten was sputtered onto a carbon backing to reduce bremsstrahlung of the backing. The effect of electrons reflected by the backing has been corrected. Comparison with two theoretical calculations has performed. The experimental results agree rather well with the theoretical predications.

Keywords: electron impact, L-shell ionization, tungsten, cross section
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The determination of atomic inner shell ionization cross sections for electron impact is of importance in various basic and applied fields. Furthermore, this type of data is also important to see to which extent the existing theoretical models can describe the experimental data in the energy range from threshold to several tens keV. But reliable experimental values for ionization cross sections are quite scarce, especially for L and M shells. For tungsten the experiment studies of relative L-shell ionization by low-energy electron impact there were three reports, namely the measurement of L-subshell ionization cross section ratios (σ_{L3}/σ_{L2} and σ_{L2}/σ_{L1}) by Carvalho *et al.*(in the energy range from 12.5 to 40 keV),[1] the measurement of L-subshell ionization cross section ratio σ_{L3}/σ_{L2} by Salgueiro *et al.* (in the energy range of 11.59–11.98 keV),[2] and the determination of L-subshell ionization cross sections $\sigma_{L1}, \sigma_{L2}, \sigma_{L3}$ by Chang (in the energy range of 11–40 keV).[3] In the report of Chang a thick target was used and the relative ionization cross sections deduced from the measured X-ray production cross sections were finally scaled to the values of σ_{L1}, σ_{L2}, σ_{L3} calculated according to the McGuire scaling at E_e=40 keV.

Up to now not any report dealing with absolute measurement of the production cross sections σ_{L_α}, σ_{L_β} and σ_{L_γ} for tungsten in the energy range of the present experiment has been found to our knowledge. These cross sections have been measured by observing the number of characteristic L X-rays produced when thin tungsten target was impacted with an electron beam of known energy and intensity. A block diagram of the present experimental setup is described in Fig.1.

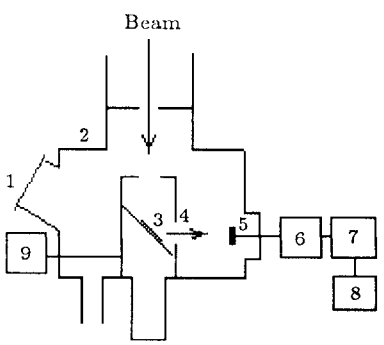


Fig.1. Block diagram of the equipment. 1.View window; 2.Chamber; 3.Target; 4.Faraday cup; 5.Si(Li) detector; 6.Preamplifier; 7.Main amplifier; 8.MCA; 9.Charge integrator.

A monoenergetic beam of electron whose energy varied from 11 to 36 keV was well collimated and then hit the target which was placed at 45° with respect to the direction of incident beam. The beam spot on the target was less than 3 mm in diameter. A Si(Li) detector, which with a horizontal dipstick was kept inside a vacuum chamber to reduce energy loss from air and placed about 10 cm from the center of the target, had 12 mm^2 active area, 3.5 mm effective

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depth and an energy resolution of 180 eV (full width at half maximum) at 5.9 keV for the Mn$K\alpha$ X-ray. The detector efficiency was calibrated with a set of standard sources with 3% uncertainty: ^{241}Am, ^{137}Cs and ^{54}Mn. A deep graphite Faraday cup connected with an ORTEC model 439 digital current integrator, was used to collect the charges on and through the target. The tungsten of 28.8 μg/cm^2 was sputtered onto a 63μg/cm^2 carbon backing and the target thickness was determined by direct weighing of a measured area, by using a balance with a precision of 10^{-6}g. The target was placed at the center of the chamber and was provided by China Institute of Atomic Energy. During experiment the gas pressure was less than 3×10^{-3} Pa in the chamber and the intensity of electron beam current was adjusted to be about 1 nA. Accelerating voltage can vary from 0 to 60 kV and was furnished by a special supply having a stability of 10^{-3}. A digital voltmeter was used for monitoring high voltage variance.

The production cross section $\sigma_{\mathrm{L}\alpha}$ is related to the observed L_α X-ray counts $N_{\mathrm{L}\alpha}$ by the formula

$$\sigma_{\mathrm{L}\alpha} = \frac{4\pi N_{\mathrm{L}\alpha}\cos\theta}{N_{\mathrm{e}} dn\varepsilon\Omega}, \tag{1}$$

where n (in 1/cm^3) and d (in cm) are the density and the thickness of the target respectively; N_{e} is the number of electrons hitting the target; ε the efficiency of the detector; Ω the solid angle subtended by the beam spot to the detector's effective area; θ the angle between the incident electron beam and the normal of the target surface. The expressions of $\sigma_{\mathrm{L}_\beta}$ and $\sigma_{\mathrm{L}_\gamma}$ are similar to the above expression. The mean ionization cross section $\overline{\sigma}_{\mathrm{L}}$ of the L-shell can be expressed as

$$\overline{\sigma}_{\mathrm{L}} = \frac{\sigma_{\mathrm{L}}}{\overline{\omega}_{\mathrm{L}}} = \frac{\sigma_{\mathrm{L}_\alpha} + \sigma_{\mathrm{L}_\beta} + \sigma_{\mathrm{L}_\gamma}}{\overline{\omega}_{\mathrm{L}}}, \tag{2}$$

where σ_{L} is the total production cross section and $\overline{\omega}_{\mathrm{L}}$ is the mean fluorescence yield of L-shell, which is defined[4] by

$$\overline{\omega}_{\mathrm{L}} = \frac{\omega_1^{\mathrm{eff}}/I_1 + \omega_2^{\mathrm{eff}}/I_2 + 2\omega_3^{\mathrm{eff}}/I_3}{1/I_1 + 1/I_2 + 2/I_3}, \tag{3a}$$

where I_i is the ionization energy of the ith subshell and

$$\omega_1^{\mathrm{eff}} = \omega_1 + f_{12}\omega_2 + (f_{13} + f_{12}f_{23})\omega_3, \tag{3b}$$

$$\omega_2^{\mathrm{eff}} = \omega_2 + f_{23}\omega_3, \tag{3c}$$

$$\omega_3^{\mathrm{eff}} = \omega_3, \tag{3d}$$

with ω_i being the fluorescence yield for the ith subshell and f_{ij} the Coster-Kronig transition probability between the ith and jth subshells. The value $\overline{\omega}_{\mathrm{L}}$ of the tungsten was estimated to be 0.27 from the above equations. The main sources of the total production cross section uncertainties come from inhomogeneity of the target thickness (<5%), the detector efficiency (<4%), the beam integration (<3%) and the counting statistical errors ((2–12)%). Without taking into account the errors of constants used in the cross section calculation, the overall uncertainty of the cross section was estimated to be less than 14%. The fluorescence yields, Coster-Kronig transition probabilities and ionization energies of L-subshell used in the evaluation procedure of the present study are given in Table 1.

Table 1. Fluorescence (ω_i)[a], Coster-Kronig transition coefficient (f_{ij})[a], ionization energy (I_i)[b] (in keV) and X-ray emission rates (Γ)[c] for W.

ω_1	ω_2	ω_3	f_{12}	f_{13}	f_{23}	I_1	I_2		I_3
0.147	0.270	0.255	0.170	0.280	0.133	12.099	11.542		10.205
$\Gamma_{\text{total-L}_1}$	$\Gamma_{\text{total-L}_2}$	$\Gamma_{\text{total-L}_3}$	$\Gamma_{\text{M4-L}_3}$	$\Gamma_{\text{M5-L}_3}$	$\Gamma_{\text{M4-L}_2}$	$\Gamma_{\text{M3-L}_1}$	$\Gamma_{\text{M2-L}_1}$	$\Gamma_{\text{N1-L}_3}$	$\Gamma_{\text{N1-L}_2}$
0.805	1.397	1.244	0.1018	0.898	1.138	0.330	0.2636	0.01119	0.00755
$\Gamma_{\text{N2-L}_1}$	$\Gamma_{\text{N3-L}_1}$	$\Gamma_{\text{N4-L}_2}$	$\Gamma_{\text{N5-L}_3}$	$\Gamma_{\text{O3-L}_1}$	$\Gamma_{\text{O4-L}_2}$	$\Gamma_{\text{O4-L}_3}$			
0.0647	0.0858	0.2118	0.1593	0.01288	0.00586	0.00468			

[a] taken from Ref.[5]; [b] taken from Ref.[6]; [c] taken from Ref.[7].

A typical X-ray spectrum for tungsten was shown in Fig.2. The partial production cross sections of L_α-, L_β-, L_γ-rays, the total L-shell production cross sections σ_{L} (not including L_1 X-rays), and the L-shell mean ionization cross sections $\overline{\sigma}_{\mathrm{L}}$ as functions of incident electron energy are plotted in Figs.3 and 4.

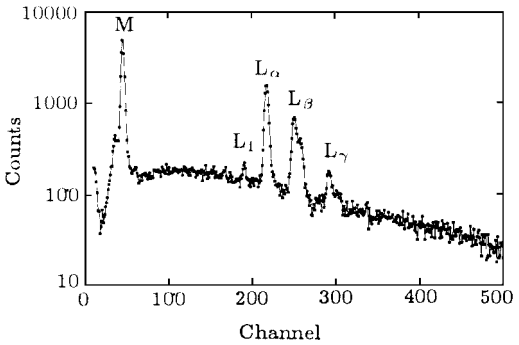


Fig.2. X-ray spectrum of tungsten at the electron energy of 30 keV.

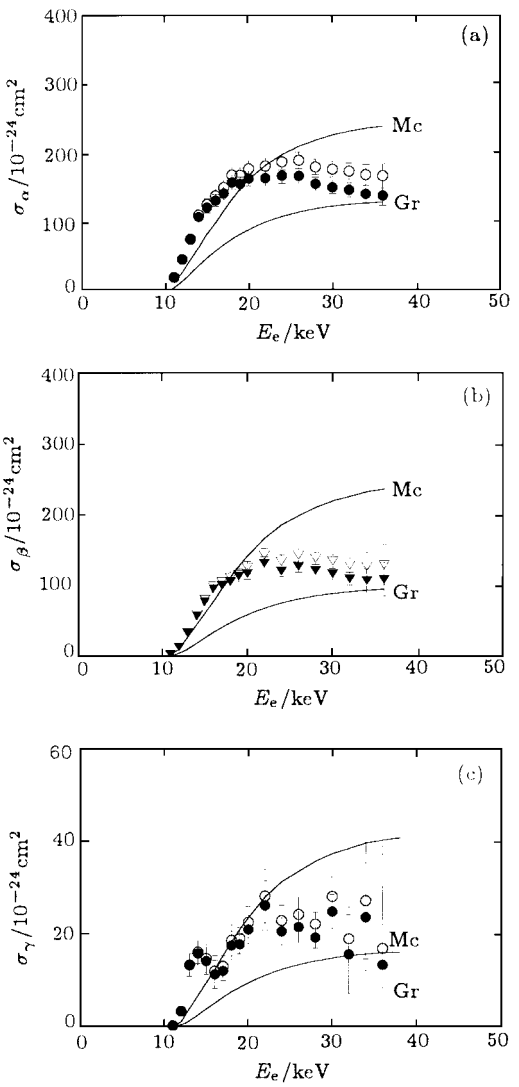


Fig.3. L-shell cross sections as functions of electron energy. (a) experimental values of $L_\alpha-$rays production cross section, (b) experimental values of $L_\beta-$rays production cross section, (c) experimental values of $L_\gamma-$rays production cross section; The symbol Mc indicats McGuire's theory, Gr Gryzinski's theory. The hollow symbols are experimental values and the filled symbols are corrected experimental values for the reflected electron effect.

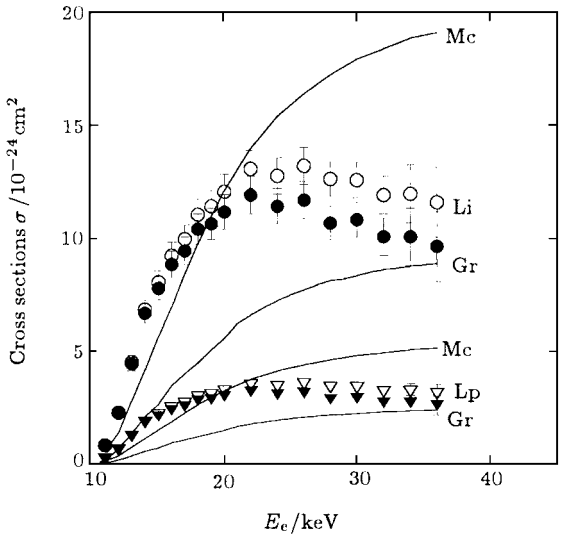


Fig.4. Total production cross sections (L_p) and ionization cross sections (L_i)of tungsten as functions of electron impact energy. The hollow symbols are experimental values and the filled symbols are corrected experimental values.

Because the preparation of a thin self-supporting target is difficult, a 63 μg/cm^2 carbon film was used as backing in the present experiment. But the electrons reflected from the backing film, when inversely passing through the tungsten target, may once again ionize tungsten atoms, resulting in a systematic overestimation of the measured cross sections. We have calculated the fraction of L-shell ionization events caused by the electrons reflected from the carbon backing, the resulting L-subshell production cross section can be written as

$$\sigma_{\mathrm{L}_i}(E) = \frac{4\pi N_{\mathrm{L}_i}\cos\theta}{N_e dn\varepsilon\Omega} - \cos\theta\int_0^E \Phi_{\mathrm{ref}}(E')\sigma_{\mathrm{L}_i}(E')\mathrm{d}E', \quad (4)$$

where i indicates α, β, γ and the second term is the fraction of L-subshell ionization events caused by the electrons reflected from the carbon backing, and $\Phi_{\mathrm{ref}}(E')$ is the energy spectrum of the electrons reflected from the backing surface, which can be acquired by using a so-called bipartition model for electron transport.[8] The symbol $\sigma_{\mathrm{L}_i}(E')$ is the production cross sections induced by the electrons with energy E'. Equation (4) can be resolved by iteration (see Ref.[9]).The corrected values of the measured cross section and corresponding errors are also presented in Figs.3 and 4.

In Figs.3 and 4 the measured cross section values for L lines from the present measurements are

compared with two different calculations: the so-called binary encounter approximation(BEA) theory of Gryzinski[10] and the theory of McGuire for atomic rearrangement.[11] For comparison with the experimental data, the L-subshell ionization cross sections σ_{L_1}, σ_{L_2}, σ_{L_3} deduced from the above two theories must be converted into the production cross sections by following formulae:[12]

$$\sigma_{L_\alpha} = \frac{\Gamma_{M4,5\text{-}L_2}}{\Gamma_{\text{total-}L_3}}\omega_3[\sigma_{L_3} + f_{23}\sigma_{L_2} + (f_{13} + f_{12}f_{23})\sigma_{L_1}], \tag{5}$$

$$\begin{aligned}\sigma_{L_\beta} =& \left(\frac{\Gamma_{N1.5\text{-}L_3} + \Gamma_{O4,5\text{-}L_3}}{\Gamma_{\text{total-}L_3}}\right) \\ & \cdot \omega_3[\sigma_{L_3} + f_{23}\sigma_{L_2} + (f_{13} + f_{12}f_{23})\sigma_{L_1}] \\ & + \frac{\Gamma_{M4\text{-}L_2}}{\Gamma_{\text{total-}L_2}}\omega_2(\sigma_{L_2} + f_{12}\sigma_{L_1}) + \frac{\Gamma_{M2,3\text{-}L_1}}{\Gamma_{\text{total-}L_1}}\omega_1\sigma_{L_1},\end{aligned} \tag{6}$$

$$\begin{aligned}\sigma_{L_\gamma} =& \left(\frac{\Gamma_{N1,4\text{-}L_2} + \Gamma_{O4\text{-}L_2}}{\Gamma_{\text{total-}L_2}}\right)\omega_2(f_{12}\sigma_{L_1} + \sigma_{L_2}) \\ & + \left(\frac{\Gamma_{N2,3\text{-}L_1} + \Gamma_{O2,3\text{-}L_1}}{\Gamma_{\text{total-}L_1}}\right)\omega_1\sigma_{L_1}.\end{aligned} \tag{7}$$

In Eq. (5) $\Gamma_{M4,5\text{-}L_3}$ and $\Gamma_{\text{total-}L_3}$ indicate the X-ray emission rates for L_α (M4,5-L_3 transition) and the total (M,N,O-L_3 transition) X-rays. In Eqs.(6) and (7) corresponding parameters have similar mean within Eq.(5). The emission rates used in theoretical calculation of the production cross section are also given in Table 1.

It can be seen that the shape of the energy dependence of cross sections measured in this work follows neither of the two theories. At lower energies (bellow 19, 18, 15 keV for σ_α, σ_β, σ_γ respectively) experimental data are higher than the two theory predictions and at higher energies the experimental results are gradually getting close to the predictions by BEA and in remainder energy ranges the present experimental results lie in between the two theory predictions. The above observed trend of experimental curves is roughly consistent with the results of Chang,[3] who measured the production cross sections for $L_{\alpha 1}$, $L_{\beta 1}$ and $L_{\beta 3}$ X-rays, then deduced the ionization cross sections σ_{L_1}, σ_{L_2}, σ_{L_3} and also compared these ionization cross sections with the predictions of Gryzinski and McGuire.

In conclusion, owing to the discrepancies between the experimental data and the theoretical results near threshold energy, it seems that none of available theories can successfully interpret the behaviors of L-shell ionization of the heavy elements by low-energy electron impact so far. Therefore, further investigation is needed both experimentally and theoretically for heavy elements at low-energy electron impact so as to get a better understanding of the inner-shell ionization processes for the heavy elements.

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