INTEGRAL CROSS SECTIONS OF IONIZATION OF L-SUBSHELLS OF TUNGSTEN ATOMS UNDER ELECTRON BOMBARDMENT NEAR ENERGETIC THRESHOLD

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The relative intensities $h = I(\beta_1)/I(\beta_3)$ and $g = I(\beta_{2,15})/I(\beta_3)$ of $L\beta_1$, $L\beta_{2,15}$, and $L\beta_3$ lines of the X-ray emission $L\beta$ spectrum of tungsten atoms W are experimentally studied under electron bombardment in a range of accelerating voltages $U = 13 \div 35$ kV. On the basis of the derived experimental data, the ratios of the integral cross sections σ_2/σ_1 and σ_3/σ_1 of ionization of L_1, L_2, and L_3 subshells are determined. For the account of the electron beam energy reduction under deepening in the substance of a specimen, the effective energy of electrons was used. It is revealed that the distinctions between the results of calculation in the classical model of binary collisions (MBC), those by the semiempiric Bethe formula with parameters derived for the K-shell ionization (BK), and the experimental values of σ_2/σ_1 and σ_3/σ_1 do not fall outside the limits of experimental errors ($8 \div 10$ % for σ_2/σ_1 and $13 \div 15$ % for σ_3/σ_1) in the region $U \geq 17$ kV. In the near-threshold region $U < 16$ kV, a good agreement between experimental data and theoretical results based on the MBC is preserved, whereas the differences between BK-calculations and experiment grow essentially, by reaching 30% for σ_2/σ_1 and 45% for σ_3/σ_1 at $U = 13$ kV. The parameters in the semiempiric Bethe formula are determined and can be used in calculations of the integral cross sections of ionization of L_1, L_2, and L_3 subshells.

The integral cross section of ionization (ICSI) of an electron shell is an important characteristic of the process of interaction of an atom with an incident particle. In particular, of specific meaning under electron bombardment are ICSI of inner K- and L-shells, because the correctness of their determination influences directly the exactness of results of a quantitative X-ray electron-probe microanalysis (the correction for the atomic number) [1].

However, up to now there is no single approach as for the calculation of ICSI of K-shells and L_i-subshells ($i = 1 \div 3$) of atoms and ions in the region of intermediate ($1.25 < \varepsilon < 4$) and especially near-threshold ($\varepsilon < 1.25$) energies of incident electrons ($\varepsilon = E/E_i$ is the excess of the energy of an incident electron E above the ionization potential E_i of the relevant shell or subshell). Indeed, sequential quantum-mechanical calculations of ICSI by the methods of convergent strong coupling (CSC) and R-matrix consider, in fact, only the processes of ionization and excitation of the electron shells of atoms of light elements with $Z < 15$ (see, e.g., [2—4]). Instead of that, for $Z > 15$, the calculations based on perturbation theory, namely on the first Born approximation, are well known. In particular, quite successful are the calculations of ICSI of inner electron shells of many multicharged ions in the region of intermediate and high energies of incident electrons with the use of distorted plane waves in this approximation. But, up to now, the plane-wave Born approximation (PWBA) remains to be widely used. The attractiveness of namely this approach consists in that it gives a rather simple analytic expression for the determination of ICSI (the Bethe formula [6] and its modifications [7, 8]) whose main parameters are the number of electrons n_i in the i-shell (subshell) and the energy excess ε. However, this model is valid only for great $\varepsilon \gg 1$ (as a rule, it is sufficient to consider $\varepsilon > 4$ [9]). Therefore, to use the Bethe formula for intermediate and especially near-threshold energies of incident electrons, one modifies it by correcting functions which ensure the agreement of calculations with the available experimental data on ICSI in the region $\varepsilon < 4$ by preserving the Bethe asymptotics at $\varepsilon \gg 1$. Such a semiempiric approach as for modifying the Bethe formula is realized in many works (e.g., [10—13] and recent ones [14—16]). However, the explicit form of correcting functions in the whole region of excesses $\varepsilon > 1$ is selected only for K-shell, and the attempts to define these functions for L_i-subshells are restricted by the region $\varepsilon > 1.25$ [9]. Thus, the form of correcting functions in the Bethe formula is unknown, in fact, in the near-threshold region $\varepsilon < 1.25$ for L_i-subshells. It should be mentioned that the classical MBC [17] is quite widely used for the description of the process of ionization of atoms by electron impact, which allows one to derive an analytic formula for ICSI also in the form of a function of n_i and ε. It is assumed that the MBC and PWBA are correct only for great excesses $\varepsilon \gg 1$.

The process of near-threshold ionization of K- and L-shells looks to be especially complicated for the atoms of heavy elements $Z > 70$ with unfilled outer electron shells. In this case, the calculations of ICSI by the methods of CSC or R-matrix are hardly possible in the general case due to the necessity to consider a huge amount of open and closed decay channels. Thus, the problem of choice of the most efficient parameters for a semiempiric description of ICSI for this group of elements is urgent. Its solution for ICSI of L_i-subshells can be derived if one uses the experimental data on the ratios of integral cross sections σ_2/σ_1 and σ_3/σ_1 or, in particular, the relative intensities of $L\beta_1$, $L\beta_{2,15}$, and $L\beta_3$ lines of the X-ray emission $L\beta$ spectrum [18]. Though this method does not allow one to determine the absolute values of ICSI of L_i-subshells, it is rather sensitive for the elucidation of the problem of correctness of various models of calculation of ICSI. Especially this concerns the near-threshold region of excesses of ε, because the significant increase in the ratios σ_2/σ_1 and σ_3/σ_1 under approaching the thresholds enables one to more clearly reveal the distinctions between the theoretical ratios in various approximations and experimental ones. Therefore, we carried out the experimental study of the ratios of cross sections of ionization σ_2/σ_1 and σ_3/σ_1 of L_1, L_2, and L_3 subshells by electron impact in the range of accelerating voltages $U = 13 \div 35$ kV ($1 < \varepsilon < 3$) and compared these ratios with the values calculated within various models.

Experimental Procedure

The X-ray emission $L\beta$-spectra of W under electron bombardment were obtained by using a Bragg spectrometer with a planar monocrystal of quartz in the third order of reflection from planes $(11\bar{2}0)$. As a source of X-rays, we took an IRIS-M module with an X-ray tube BSV-29W. The spectra were registered in the mode of stepwise scanning (with a step $\Delta\theta = 0.01 \div 0.03°$ on the energy scale $\Delta E = 2 \div 4$ eV) with a prescribed number of stored impulses N. The necessity to hold the acceptable duration of the experiment under near-threshold excitation ($U = 13 \div 14$ kV) forced us to restrict ourselves by $N = 10^4$ impulses, which corresponds to a relative error $\delta N = 1$ %; for higher voltage — by $N = 4 \cdot 10^4$ impulses, $\delta N = 0.5$ %. The reciprocal linear dispersion of a spectrometer in the region of $L\beta_1$-line was 38 eV/mm. Thus, the absolute error of determination of the energy was at most 4 eV for the inlet width of a detector $d = 0.1$ mm. By processing the spectrograms, we introduced the corrections for the dispersion of the installation under transition from the angle scale to the energy one, angle dependence of the reflection coefficient of a crystal-analyzer, and differences in the adsorption of $L\beta_1$, $L\beta_{2,15}$, and $L\beta_3$ lines of W in the substance of the X-ray tube anode (the correction by Philibert [1]), air, and the Be window of the tube. The relative intensities of $L\beta_1$, $L\beta_{2,15}$, and $L\beta_3$ lines of W were defined as the ratios of the areas under the experimental contours of these lines. The common relative error of determination of the relative intensities of $L\beta_1$, $L\beta_{2,15}$, and $L\beta_3$ lines of W was 3—4% under the given experimental conditions.

Discussion of Results

Below, we present the relations between the experimentally determined relative intensities of $L\beta_1$, $L\beta_{2,15}$, and $L\beta_3$ lines and the quantities σ_2/σ_1 and σ_3/σ_1. We took into account that the processing of spectrograms did not involved multiple-ionization satellites which correspond to the radiative transitions in LM-, LX-, LMX-, and LXY-ionized atoms (X, Y = N, O, ...). Then, to within a constant common factor, the intensities of $L\beta_1$, $L\beta_{2,15}$, and $L\beta_3$ lines together with satellites (in terms of the photon numbers) are as follows, with all corrections included:

$$I(\beta_{3+S}) = \sigma_1 \frac{\Gamma^R(\beta_3)}{\Gamma_1}, \tag{1}$$

$$I(\beta_{1+S}) = (\sigma_2 + \sigma_1 f_{12}) \frac{\Gamma^R(\beta_1)}{\Gamma_2}, \tag{2}$$

$$I(\beta_{2,15+S}) = [\sigma_3 + \sigma_2 f_{23} + \sigma_1 (f_{13} + f_{12} f_{23})] \frac{\Gamma^R(\beta_{2,15})}{\Gamma_3}. \tag{3}$$

Here, $\Gamma^R(\beta_3)$, $\Gamma^R(\beta_1)$, and $\Gamma^R(\beta_{2,15})$ are the partial widths of L_1-, L_2-, and L_3-levels corresponding to the radiative transitions $L_1 - M_3(L\beta_3)$, $L_2 - M_4(L\beta_1)$, and $L_3 - N_{4,5}(L\beta_{2,15})$; f_{12} is the yield of Coster—Kronig (CK) transitions of the type $L_1 - L_2$X; f_{13} is the yield of CK-transitions $L_1 - L_3M_5$ and $L_1 - L_3$X; f_{23} is the yield of CK-transitions $L_2 - L_3$X; and Γ_1, Γ_2, and Γ_3 are the total widths of L_1-, L_2-, and L_3-levels, respectively. It is taken into account in (1)—(3) that the initial vacancy in L_1-subshell is formed only to the expense of the direct ionization by electron impact, whereas that in L_2-subshell is formed due to both the direct ionization and autoionization via CK-transitions $L_1 - L_2$X (in the last case, the created two-vacancy states are the initial ones for L_2X-satellites which are imposed on the diagram $L\beta_1$-line). The generation of vacancies in L_3-subshell via direct ionization is supplemented by CK-transitions $L_1 - L_3M_5$ and CK-cascade $L_1 - L_2$X and L_2X $- L_3$XY. As in the case of $L\beta_1$-line, L_3M_5, L_2X, and L_3XY-states are initial for the relevant satellites which are registered along with the line $L\beta_{2,15}$. Solving the system of equations (1)—(3), we get

$$\frac{\sigma_2}{\sigma_1} = h\frac{\Gamma_2\Gamma^R(\beta_3)}{\Gamma_1\Gamma^R(\beta_1)} - f_{12}, \tag{4}$$

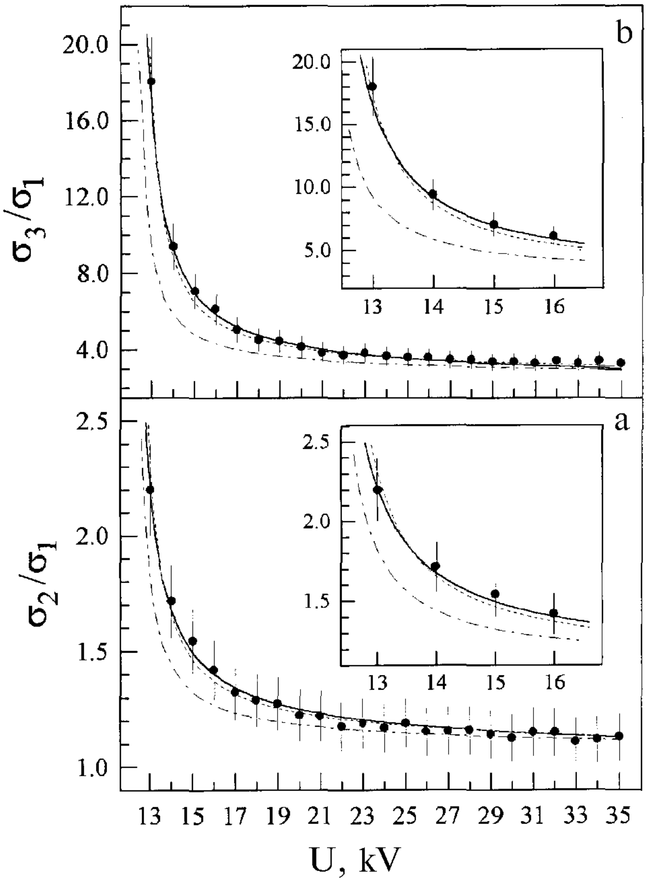
$$\frac{\sigma_3}{\sigma_1} = g\frac{\Gamma_3\Gamma^R(\beta_3)}{\Gamma_1\Gamma^R(\beta_{2,15})} - \frac{\sigma_2}{\sigma_1}f_{23} - f_{13} - f_{12}f_{23}, \tag{5}$$

where $h = \frac{I(\beta_1+s)}{I(\beta_3+s)}$; and $g = \frac{I(\beta_{2,15}+s)}{I(\beta_3+s)}$.

In the calculations by formulas (4) and (5), we used the well-justified values of the radiative widths of lines $\Gamma^R(\beta_i)$, widths L_2 and L_3 of levels Γ_2 and Γ_3, and yields f_{ij}. In particular, we took $\Gamma^R(\beta_1) = 1.138$ eV, $\Gamma^R(\beta_{2,15}) = 0.177$ eV, $\Gamma^R(\beta_1) = 0.33$ eV [19], $\Gamma_2 = 4.82$ eV, $\Gamma_3 = 4.81$ eV, and $f_{23} = 0.139$ [20]. Recently, we have derived a corrected value of the width of L_1-level and yields f_{12} and f_{13} for atoms W with regard for the existence of CK-transitions $L_1 - L_3M_5$: $\Gamma_1 = 7.12$ eV, $f_{12} = 0.156$, and $f_{13} = 0.457$ [18]. In view of the accuracy of determination of these values, the relative errors of calculations of σ_2/σ_1 and σ_3/σ_1 are equal to, respectively, $8 \div 10$ % by (4) and $13 \div 15$ % by (5).

Relative intensities of emission $L\beta_1$, $L\beta_{2,15}$, and $L\beta_3$ lines of W atoms and effective energies of bombarding electrons at some values of accelerating voltages

U, kV	$E_{\text{eff}}(L_1)$, keV	$E_{\text{eff}}(L_2)$, keV	$E_{\text{eff}}(L_3)$, keV	h	g
13	12.55	12.27	11.56	11.9 ± 0.4	14.8 ± 0.5
14	13.04	12.75	12.02	9.6 ± 0.3	7.97 ± 0.28
15	13.52	13.22	12.48	8.6 ± 0.3	6.00 ± 0.21
20	15.76	15.42	14.55	6.98 ± 0.24	3.78 ± 0.13
25	17.75	17.34	16.27	6.8 ± 0.24	3.35 ± 0.12
35	20.72	20.08	18.31	6.49 ± 0.23	3.10 ± 0.11



Ratios of the integral cross sections σ_2/σ_1 and σ_3/σ_1 of atoms W for various accelerating voltages. The designations are given in the text

It is worth of noting that we must consider a decrease in the energy of electrons of the beam at a deepening into the substance of a specimen while interpreting the results derived under electron bombardment of thick targets. One of the possible ways to solve this problem consists in the use of the effective electron energy E_{eff} which would be the monoenergetic equivalent of ionizing action of electrons of the beam in the process of diminution of their energy from $E_0 = eU$ (e is the electron charge) to E_i [21]. Such an approach justified in experiment [22, 23] is used in the present work: for any L_i-subshells, we calculated the effective energies E_{eff} whose values are given in the table for some accelerating voltages. We emphasize that, by virtue of the difference in the ionization potentials E_1, E_2, and E_3 for each L_1, L_2, and L_3 subshells at the same energy E_0 (E_1 = 12.103 keV, E_2 = 11.546 keV, and E_3 = 10.209 keV [24], respectively), the effective energies

$E_{\rm eff}(L_1)$, $E_{\rm eff}(L_2)$, and $E_{\rm eff}(L_3)$ differ remarkably one from another, especially at approaching the threshold.

The table shows the experimentally determined relative intensities h and g at some accelerating voltages, and the figure presents the ratios of cross sections σ_2/σ_1 and σ_3/σ_1 calculated by (4) and (5). The dashed line shows the result of calculations in the MBC [17], and the dash-dotted line corresponds to the semiempiric formula [15]:

$$\sigma_i = n_i \pi a_0^2 \left(\frac{\rm Ry}{E_i}\right)^{G(\varepsilon)} \left(a + \frac{b}{\varepsilon} + \frac{c}{\varepsilon^2}\right)\frac{\ln(\varepsilon)}{\varepsilon}, \tag{6}$$

where

$$G(\varepsilon) = d + \frac{k}{\varepsilon} + \frac{f}{\varepsilon^2},$$

$a = 3.125$, $b = -4.172$, $c = 1.877$, $d = 2.0305$, $k = -0.316$, $f = 0.1545$, a_0 is the first Bohr radius, and ${\rm Ry} = 13.61$ eV. For each accelerating voltage U, we used the relevant effective energies $E_{\rm eff}(L_1)$, $E_{\rm eff}(L_2)$, and $E_{\rm eff}(L_3)$ and, with them, calculated the cross sections σ_1, σ_2, and σ_3 and their ratios σ_2/σ_1 and σ_3/σ_1.

Formula (6) was deduced in [14] for ICSI of K-shell by fitting the parameters a, b, c, d, k, and f by the method of least squares on the comparison of the calculated ICSI with a large amount of experimental data for neutral atoms of various elements. In view of the volume of the used experimental data and the accuracy of approximation, we may assert that formula (6) is the best generalization of analogous formulas given in works [10—14, 16]. We note that the ratios σ_2/σ_1 and σ_3/σ_1 calculated by the formulas from [10—12, 15, 16] coincide in the limits of 2—3%; therefore, the figure of our work gives only the results calculated by (6).

As seen from the figure, in the region of accelerating voltages $U \geq 17$ kV ($\varepsilon \geq 1.4$), the differences between the results of calculations [15—17] and experimental values of σ_2/σ_1 and σ_3/σ_1 do not go beyond the experimental errors. However, while approaching the ionization threshold of L_i-subshells, the differences between the results of calculations by the MBC and semiempiric formula (6) increase significantly by reaching 30% for σ_2/σ_1 and 45% for σ_3/σ_1 at $U = 13$ kV. It is unexpected that the ratios σ_3/σ_1 calculated by the MBC fit the experimental data better than those by (6) on decreasing the accelerating voltage. We noted earlier that the use of the MBC is justified at high excesses. At the same time, for K-shells of some elements (e.g., Be, C, Ag, and Gd) in the region $\varepsilon < 1.5$, the values of ICSI calculated by this model coincide in the limits of 15—20% with experimental ones [25, 26]. Thus, the question on the limits of applicability of the MBC is not clarified finally. Therefore, the good agreement of experimental and theoretical values of σ_2/σ_1 and σ_3/σ_1 for L_i-subshells of W which is obtained in this work indicates the necessity of an additional consideration of the possibility for the use of the MBC in the near-threshold region.

As for the semiempiric formula (6), the presented values of the parameters a, b, c, d, k, and f correspond, as mentioned above, to the best agreement with experimental values of ICSI for namely K-shell. At the same time, for L_i-subshells, the coefficients in correcting functions can vary [9, 11]. Therefore, we carried out the fitting of the parameters a, b, c, d, k, and f by minimizing the function $Z(a,b,c,d,k,f)$ which is the sum of the squares of deviations of the experimentally determined ratios σ_2/σ_1 from the ones calculated by (6):

$$Z(a,b,c,d,k,f) = \sum_{i=1}^{N}\left[\left(\frac{\sigma_2}{\sigma_1}\right)_{\rm exp} - \left(\frac{\sigma_2}{\sigma_1}\right)_{\rm calc}\right]^2, \tag{7}$$

where $N = 23$. The minimization was performed by the method of gradient descent with subdivision of a step [27]. We obtained the best result if the parameters d, k, and f of the exponential function are constant, and only a, b, and c are varied. We got: $a = 2.979$, $b = 4.422$, and $c = 1.527$. Then we calculated the ratios σ_2/σ_1 and σ_3/σ_1 with the new values of a, b, and c (the continuous lines in the figure). As seen, the agreement with experiment is better in this case, especially in the near-threshold region. To additionally verify the correctness of this result with the given parameters a, b, and c, we calculated the absolute values of ICSI σ_1, σ_2, and σ_3 and compared them with the scaling data [9]. We obtained that, for $\varepsilon > 1.25$, the differences between these data do not exceed 6—8%. (We note the experimental data of ICSI of L_i-subshells of W for the near-threshold region are unknown to us). Thus, the general semiempiric formula (6) can be recommended for the use in calculations of ICSI of L_i-subshells of atoms of heavy elements with $Z > 70$ in the whole range of excesses ε, including the near-threshold region, if the values of the parameters a, b, and c given in this work are used.

Thus, on the basis of the experimentally determined values of the relative intensities of $L\beta_1$, $L\beta_{2,15}$, and $L\beta_3$ lines of the X-ray emission $L\beta$-spectrum of W, we have found the ratios of ICSI for L_i-subshells σ_2/σ_1 and σ_3/σ_1 in the region of excesses $1 < \varepsilon < 3$. We have clarified that the dependence of the ratios σ_2/σ_1 and σ_3/σ_1 on the energy of bombarding electrons is quite well described in the approximation of classical binary collisions. We have determined the numerical values of parameters in the semiempiric Bethe formula which give the best agreement of the experimental ratios σ_2/σ_1 and σ_3/σ_1 with those calculated by the formula.

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ІНТЕГРАЛЬНІ ПЕРЕРІЗИ ІОНІЗАЦІЇ L-ПІДОБОЛОНОК АТОМІВ ВОЛЬФРАМУ ПРИ ЕЛЕКТРОННОМУ БОМБАРДУВАННІ БІЛЯ ЕНЕРГЕТИЧНОГО ПОРОГА

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Резюме

Експериментально досліджено відносні інтенсивності $L\beta_1$-, $L\beta_{2,15}$- та $L\beta_3$-ліній — $h = I(\beta_1)/I(\beta_3)$ та $g = I(\beta_{2,15})/I(\beta_3)$ — рентгенівського емісійного $L\beta$-спектра атомів вольфраму при електронному бомбардуванні зразка у діапазоні прискорюючих напруг $U = 13 \div 35$ кВ. За отриманими даними визначено відношення інтегральних перерізів іонізації (ІПІ) L_1-, L_2-, L_3-підоболонок — σ_2/σ_1 та σ_3/σ_1. Врахування зменшення енергії електронів пучка із заглибленням у речовину зразка було здійснено шляхом введення ефективної енергії електронів. Виявлено, що в області $U \geq 17$ кВ розбіжності між результатами розрахунків у моделі класичних бінарних співударів (КБС) та за півемпіричною формулою Бете з параметрами, отриманими для K-оболонки (БК), і експериментальними значеннями σ_2/σ_1 та σ_3/σ_1 не виходять за межі похибок експерименту (8 — 10 % для σ_2/σ_1 та 13 — 15 % для σ_3/σ_1). У підпороговій області $U < 16$ кВ зберігається таке ж добре узгодження між експериментальними даними та розрахунками у моделі КБС, тоді як розбіжність між БК-розрахунками та експериментальними даними істотно зростає, досягаючи 30% для σ_2/σ_1 та 45% для σ_3/σ_1 при $U = 13$ кВ. Визначено значення числових параметрів у напівемпіричній формулі Бете, які пропонуються для розрахунків інтегральних перерізів іонізації L_1, L_2, L_3-підоболонок.