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ABSOLUTE CROSS-SECTIONS OF *s*- AND *d*-IONIZATIONS OF In$^+$ IONS AT THEIR COLLISIONS WITH SLOW ELECTRONS

Effective cross-sections of s- and d-ionization of In$^+$ ions by electron impact have been studied using a spectroscopic technique, when the ion and slow-electron beams cross each other at the right angle. A method of determination of the partial cross-sections for the ion ionization by electron impact is described. The absolute values of s- and d-ionization cross-sections for In$^+$ ions were found to equal 2.1×10^{-17} and 5.2×10^{-17} cm^2, respectively. The results obtained testify to the important role of the excitation-autoionization process that makes a substantial contribution to the electron-impact ionization cross-section of ions.

Keywords: indium, electron-ion collisions, ionization with excitation, partial cross-sections of ionization, excitation-autoionization.

1. Introduction

Electron-impact ionization is one of the most fundamental atomic collision processes. Its importance for all kinds of plasma stimulates experimental and theoretical studies, which gained a new impetus in connection with researches dealing with controlled thermonuclear fusion. Ionization cross-sections govern the evolution of basic elementary processes in many devices, such as magnetohydrodynamic generators, various gas-filled and electrovacuum devices, optical quantum generators, and so forth. Data concerning the ionization of ions by electrons find their application to the researches of processes in the upper atmosphere and in space researches.

Nowadays, the process of ionization of positive ions by electron impact has been studied for a number of elements [1, 2]. Intense researches during last decades not only provided a significant body of knowledge concerning atomic constants, but also gave rise to a new understanding of ionization mechanisms at the physical level [3]. In the corresponding experiments, the mass-spectrometry method of research was used, as a rule, to determine the total ionization cross-sections of ions, which are connected with the removal of electrons from both the external and internal atomic shells. However, this method does not allow one to directly determine the ionization cross-section for every shell separately (partial ionization cross-sections).

An essential issue in the course of electron removal from the internal ionic subshells is the formation of ions with various charges in excited states. In some cases, those states are stable with respect to the autoionization and the decay radiatively. Therefore, researches dealing with the excitation of spectral lines emitted at that enable us to determine the effective cross-sections of electron removal from the corresponding shells and, hence, to evaluate the partial cross-sections of ionization. Required data can be obtained using the spectroscopic method, while studying the process of ionization with the excitation. Earlier, we carried out researches of the *s*-ionization of K$^+$, Rb$^+$, and Cs$^+$ ions [4], as well as the *s*- and *d*-ionizations of a Tl$^+$ ion [5, 6], using electron impact.

In this work, we report results concerning the absolute cross-sections of the *s*- and *d*-ionizations of In$^+$ ions at the electron impact obtained on the basis of experimentally measured emission cross-sections for the excitation of $4d^{10}5p^2\ P_{1/2,3/2}$ levels in In^{2+} ions as a result of the *s*-ionization-with-excitation process,

$$\begin{array}{r} e+\mathrm{In}^+(4d^{10}5s^2)\ ^1S_0 \to \mathrm{In}^{2+}(4d^{10}5p)\ ^2P_{1/2,3/2}+2e \\ \downarrow \qquad\qquad \\ \mathrm{In}^{2+}(4d^{10}5s)\ ^2S_{1/2}+h\nu_{1,2} \end{array} \tag{1}$$

($\lambda_1 = 174.9$ nm, $\lambda_2 = 162.6$ nm), and for the excitation of $4d^95s^2\ ^2D_{3/2,5/2}$ levels in the same ions as a result of the d-ionization-with-excitation process,

$$e + \mathrm{In}^+(4d^{10}5s^2)\ ^1S_0 \rightarrow \mathrm{In}^{2+}(4d^95s^2)\ ^2D_{3/2,5/2} + 2e \\ \downarrow \qquad (2) \\ \mathrm{In}^{2+}(4d^{10}5p)\ ^2P_{1/2,3/2} + h\nu_{3,4}$$

($\lambda_3 = 153.3$ nm, $\lambda_4 = 185.0$ nm).

2. Experimental Part

The researches were carried out under conditions, when the electron and ionic beams crossed each other at an angle of 90°. The studied emitted radiation was observed in the direction perpendicular to the plane of crossing beams. The basic units of the experimental installation were described in detail in a number of works [7–9]. Therefore, we will consider only those aspects of the experimental technique that are specific to the investigations concerned.

The generation of an indium ion beam, the intensity of which is stable in time, is a complicated task, which results from the physico-chemical properties of the examined metal. In particular, the required pressure of a saturated vapor of indium atoms in the source ($10^{-1} \div 1$ Pa) is achieved at temperatures of $900 \div 1000$ °C, which is much higher than the indium melting point (156 °C). However, an environment with vapor of indium atoms is chemically aggressive at high temperatures, which gives rise to a destruction of some constructional elements of the ionic source and to the intense formation of its liquid phase on ceramic insulators. Taking the aforesaid into account, we developed and fabricated an ionic source [9] that can generate a stable beam of single-charged positive indium ions in the ground state. The ion energy was 800 eV, and the beam current equaled 1.6×10^{-6} A.

The ion beam was crossed with a beam of electrons generated by a three-anode electron gun. Within an energy interval of $20 \div 120$ eV, the current in the electron beam amounted to $(2.0 \div 2.4) \times 10^{-4}$ A, and the energy spread (monoenergeticity) of electrons was 1.5 eV. The calibration accuracy of the energy scale was ± 0.05 eV. Vacuum in the collision chamber was maintained at the pressure level $P \approx 5 \times 10^{-6}$ Pa.

Emitted radiation passed from the collision chamber into a vacuum monochromator fabricated following the Seya–Namioka scheme. Its inverse linear dispersion amounted to $d\lambda/dl \approx 1.7$ nm/mm. A "solar-blind" photoelectronic multiplier FEU-142 cooled by water was used as a radiation detector. The signal-to-background ratio amounted to $1/10 \div 1/20$. The root-mean-square error of relative measurements did not exceed 20%.

3. Measurements of Energy Dependences of Relative Emission Cross-Sections of Electron-Impact Ionization with Excitation

The research of the electron excitation of spectral transitions in In^{2+} ion owing to the s- and d-ionizations of In^+ ion by electron impact included the measurements of the optical functions of electron excitation $f(E)$ for spectral lines with wavelengths of 174.9 and 162.6 nm (for In^{2+}), and 153.3 and 185.0 nm (for In^+), respectively. For this purpose, we scanned the electron energy E and measured the amplitude of a useful signal produced by the examined process at every specific energy during equal time intervals. At every point, the useful signal was normalized by the total electron current. Note that the experimentally measured excitation function, $f_{\exp}(E)$, differs from the real one, $f_{\mathrm{real}}(E)$. This difference follows from the fact that electrons in the beam are characterized by the distribution function over the energy, $g(E)$, so that the function $f_{\exp}(E)$ is actually a convolution of two functions, $g(E)$ and $f_{\mathrm{real}}(E)$. The discrepancy between $f_{\exp}(E)$ and $f_{\mathrm{real}}(E)$ decreases if highly monochromatic electron beams are used. However, various methods of electron monochromatization considerably reduce the magnitude of electron current and therefore can be used only while studying the most intense spectral transitions.

The electron excitation functions measured by us for the transitions $4d^{10}5p\ ^2P_{1/2,3/2} \rightarrow 4d^{10}5s\ ^2S_{1/2}$ in In^+ ion (1) and the transitions $4d^95s^2\ ^2D_{5/2,3/2} \rightarrow$ $\rightarrow 4d^{10}5p\ ^2P_{1/2,3/2}$ in In^{2+} ion (2) that occur when slow electrons collide with In^+ ions, as well as the corresponding cross-sections (in relative units), were reported in works [8,10]. In Fig. 1, the sums of optical excitation functions for the λ174.9-nm and λ162.6-nm spectral lines (curve *1*) and for the λ153.3-nm and λ185.0-nm ones (curve *2*) are depicted, which correspond to the cross-sections of the s- and d-ionizations of In$^+$ ion, respectively. One can see that both examined curves reveal a structure. Optical excitation functions are known to contain information on both the direct process and all additional processes that are probable at this electron energy. This is these additional processes that manifest themselves in the analyzed curves in the form of such peculiarities as maxima and minima. The analysis of the results obtained showed that the energies of ionization states of indium (in both the atomic and ionic forms) are located within the energy interval of those peculiarities [11–14]. In our opinion, the nature of maxima and minima in the curves concerned is associated with the excitation and the following electron decay (directly or through cascade transitions) of autoionization states onto the levels of $4d^{10}5p$ and $4d^9 5s^2$ configurations of In$^+$ ion (the so-called excitation-autoionization process).

4. Determination of Absolute Cross-Section Values

According to work [15], the effective excitation cross-section for the analyzed optical transition with the wavelength λ at electron-ion collisions can be determined using the formula

$$\sigma_\lambda = \frac{e^2 v_i v_e \Phi_\lambda}{I_i I_e (v_i^2 + v_e^2)^{1/2}} F, \qquad (3)$$

where Φ_λ is the number of photons with the wavelength λ that are emitted from a unit volume per unit time owing to the collision between electrons and ions; I_i and I_e are the ion and electron current densities, respectively; v_i and v_e are the average velocities of ions and electrons, respectively, in the beams; e is the electron charge; and F is a coefficient that characterizes the uniformity of current density distributions over the beam cross-sections (the form factor). The latter is determined by the formula

$$F = \frac{\int i(z)dz \int j(z)dz}{\int i(z)j(z)dz}, \qquad (4)$$

where $i(z)$ and $j(z)$ are the dependences of the electron and ion currents, respectively, on the beam height. If the geometry of crossing beams is selected properly, the form factor F falls within the interval from 0.9 to 1.

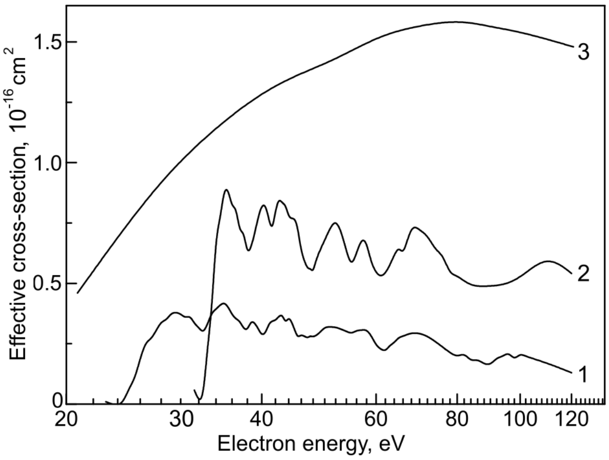

Fig. 1. Absolute cross-sections of *s*- (*1*), *d*- (*2*), and total (*3*) ionizations [20] of In$^+$ ion by electron impact

Let the number of registered photons per unit time (the useful signal) be written down as

$$C = \Phi_\lambda \eta, \qquad (5)$$

where η is the spectral sensitivity of the experimental equipment, and the relative velocity of particles in the beams (in the plane xy) as

$$V = v_i v_e \sin\theta / (v_i^2 + v_e^2 - 2 v_i v_e \cos\theta)^{1/2}. \qquad (6)$$

Then, taking into account that the beams intersect at the right angle ($\theta = 90$ °C), expression (3) for the effective cross-section of spectral line excitation by electron impact reads

$$\sigma_\lambda = \frac{C}{\eta} \frac{e^2}{I_i I_e} V F, \qquad (7)$$

where C/η is the rate of photon generation. Note that all the quantities in formula (7) can be directly measured in experiment. This is one of the advantages of the crossing-beam method.

In practice, the most complicated task consists in determining the spectral sensitivity η of a detecting system. As usual for this purpose, the intensity of studied radiation is compared with that produced by an absolute standard. However, the spectral lines analyzed by us fall within the wavelength interval of $100 \div 200$ nm, i.e. into the vacuum ultra-violet spectral range, the most precise calibrating source for which is synchrotron radiation. However, we had no

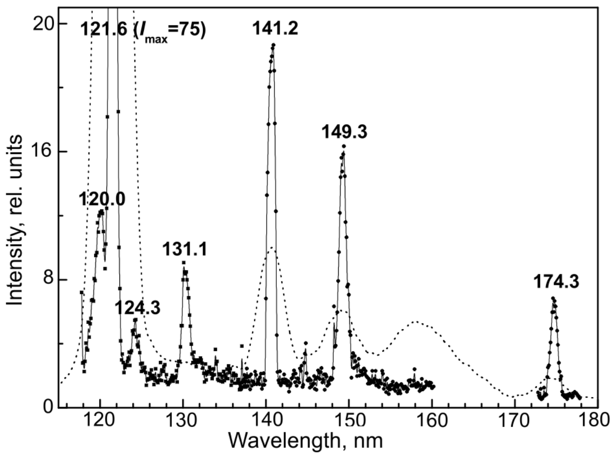

Fig. 2. Radiation emission spectrum of the residual gas at an electron energy of 100 eV, a current of 4×10^{-3}A, and pressures in the collision chamber of 10^{-5} (solid curve) and 10^{-3} Pa (dotted curve)

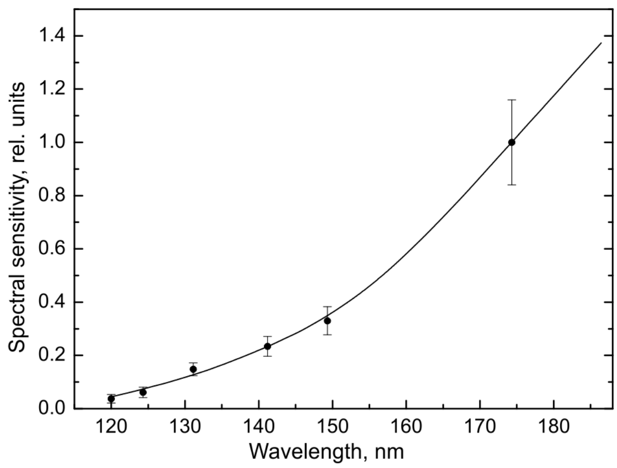

Fig. 3. Relative spectral sensitivity of the radiation detecting system

opportunity to use this source. An alternative technique for the determination of the installation spectral sensitivity, which has proven itself well in practice, is the application of one of the residual gases excited by electron impact as a calibrating source of radiation. Its essence consists in that, as one can see from formula (7), if the measurement conditions are invariant, the spectral sensitivity η_λ is a function of only the effective excitation cross-section σ of a spectral line. Therefore, knowing the electron excitation cross-sections for some spectral lines of the given gas, it is possible to determine the relative spectral sensitivity of a detecting system. In our case, the discrete values of the function $\eta_\lambda = f(\lambda)$ were determined using the formula

$$\eta_{\lambda_i} = \frac{C_i \sigma_1}{C_1 \sigma_i}\, \eta_{\lambda_1}, \tag{8}$$

where η_{λ_i} and η_{λ_1} are the sensitivity values for two spectral lines with the wavelengths λ_i and λ_1, respectively; C_i and C_1 are the corresponding signals measured at those wavelengths; and σ_i and σ_1 are the corresponding effective excitation cross-sections. Subscript 1 denotes the spectral line, for which η_λ is adopted to equal unity, whereas subscript i has as many values as the number of lines with known excitation cross-sections.

In this work, the relative spectral sensitivity of the radiation detecting system was determined from the electron-impact excitation cross-sections of hydrogen and nitrogen spectral lines. For this purpose, we studied the spectrum of electron excitation of residual gases in the wavelength interval of $100 \div 200$ nm, at an electron energy of 100 eV, and a current of 4×10^{-3} A, whereas the pressure in the collision chamber was maintained to be about 10^{-3} or 10^{-5} Pa (Fig. 2). The measured spectral lines were identified according to literature data [16]. The line of atomic hydrogen with $\lambda = 121.6$ nm, which was observed in both cases, turned out to be the most intense in the studied spectra. The other five lines were identified as the lines belonging to atomic nitrogen. Note that, in the case of higher vacuum, the intensity of lines decreased by an order of magnitude, which resulted in a reduction of the background signal. Therefore, the research of the energy dependences of the electron excitation for the spectral lines emitted by In^{2+} ions was carried out at a vacuum not more than 10^{-6} Pa. The dependence of the relative spectral sensitivity η_λ of the radiation detecting system on the wavelength, which was determined in such a manner, is exhibited in Fig. 3.

On the basis of determined η_λ-values and formula (8), it is possible to find the effective excitation cross-section

$$\sigma_i = \frac{\eta_{\lambda_1} C_i}{\eta_{\lambda_i} C_1}\, \sigma_1. \tag{9}$$

As the reference line marked by subscript 1, the line at the wavelength λ158.6 nm, which corresponds to the resonance transition $5s5p\ {}^1P_1^0 \rightarrow 5s^2\ {}^1S_0$

of In$^+$ ion, was chosen. The absolute value of effective electron-impact excitation cross-section of this line was obtained by normalizing the experimental data measured at an energy of 300 eV by the results of calculations carried out in the framework of the semirelativistic distorted-wave method, by using the Los-Alamos code [17]. The cross-section values determined in such a way were equal to 3.3×10^{-16} cm^2 for an energy of 300 eV and to 5.0×10^{-16} cm^2 at 100 eV. It should be noted that the first indicated value turned out twice as large as that obtained by normalizing the experimental data by the result of calculations according to the semiempirical Van Regemorter formula [18] (see our earlier work [8]). In our opinion, the normalization by the result of semirelativistic calculation is more correct. This conclusion is confirmed by the data of work [19], in which the results of calculations of the electron-impact excitation cross-sections of metal ions carried out in the framework of the semirelativistic distorted-wave method with the use of the Los-Alamos code agree well with both experimental data and the results of calculations carried out using other methods, in particular, the R-matrix one.

The absolute values $\sigma(E)$ of In$^+$ ion s- and d-ionization cross-sections obtained in this work are quoted in Table. The determination error for the absolute values of effective cross-sections of the analyzed lines for In^{2+} ion did not exceed 50%, being mainly a result of the errors made at relative measurements (20%) and the determination of a relative spectral sensitivity (16%), as well as of calculation accuracy (10%).

The obtained $\sigma(E)$-values of about 10^{-17} cm^2, which are comparable with the total ionization cross-section of In$^+$ ion studied in work [20] using the mass-spectrometry method, testify to rather a high efficiency of both the processes of s- (the cross-section σ^s) and d-ionizations (the cross-section σ^d) of In$^+$ ions by electron impact. A comparison of values obtained for the cross-sections of s- (curve *1*) and d- (curve *2*) ionizations, as well as for the total ionization cross-section of In$^+$ ion (curve *3*), which are shown in Fig. 1, evidences that, in the course of ionization of this ion by electron impact, not less than 50% of In^{2+} ions appear in excited states. This circumstance can be explained by the fact that an important role in the process concerned is played by indirect processes connected with ionization phenomena, in particular, the excitation-autoionization process, which can insert a sound contribution to the direct process. It is known that a distinctive feature of the ion electron excitation cross-section is the ultimate (mainly, maximum) value at the excitation threshold. Therefore, the excitation of autoionization states followed by their autoionization may result in a drastic jump of the ionization cross-section value and change it even by an order of magnitude. From the analysis of the results obtained, it follows that the dominant role in the studied processes is played by ionization states emerging owing to the excitation of electrons from the subvalence $4d^{10}$ shell, which means that the role of electron-electron correlations for such multielectron systems as In$^+$ ions is considerable.

Absolute cross-sections of s- and d-ionizations of In$^+$ ion by electron impact (10^{-17} cm^2)

No.	Transition	λ, nm	σ	σ^s	σ^d
1	$4d^{10}5p\ ^2P_{1/2} \to 4d^{10}5s\ ^2S_{1/2}$	174.9	0.7	2.1	
2	$4d^{10}5p\ ^2P_{3/2} \to 4d^{10}5s\ ^2S_{1/2}$	162.6	1.4		
3	$4d^95s^2\ ^2D_{5/2} \to 4d^{10}5p\ ^2P^o_{3/2}$	185.0	2.0		5.2
4	$4d^95s^2\ ^2D_{3/2} \to 4d^{10}5p\ ^2P^o_{1/2}$	153.3	3.2		

5. Conclusions

The energy dependences of the effective cross-sections for the excitation of spectral lines λ174.9 nm, λ162.6 nm and λ153.3 nm, λ185.0 nm corresponding to the transitions $4d^{10}5p\ ^2P \to 4d^{10}5s\ ^2S$ and $4d^95s^2\ ^2D \to 4d^{10}5p\ ^2P$ of In^{2+} ions owing to the s- and d- ionizations of In$^+$ ion by electron impact are studied. The analyzed curves reveal a structure, which can be explained by a contribution of the ionization with the excitation of an additional process – namely, the excitation and the electron decay of autoionization states (excitation-autoionization) – to the direct process. The absolute effective cross-sections of the s- and d-ionizations of In$^+$ ion by electron impact were found to equal 10^{-17} cm^2 by the order of magnitude. For this purpose, the relative spectral sensitivity of the radiation detecting system in the wavelength interval of $100 \div 200$ nm and the effective electron-impact excitation cross-section for the resonance line λ158.6 nm ($5s5p\ ^1P_1 \to 5s^2\ ^1S_0$) of In$^+$ ion were determined, the latter by normalizing the experimental data measured at an energy of 300 eV by the results of calculations carried out in the framework of the semirelativistic distorted-wave method with the help of the Los-Alamos code. Up to 50% of In^{2+} ions were shown to emerge in excited states owing to the ionization of In^{+} ions by electron impact.

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АБСОЛЮТНІ ПЕРЕРІЗИ *s*- І *d*-ІОНІЗАЦІЇ ІОНА In^{+} ПРИ ЗІТКНЕННЯХ З ПОВІЛЬНИМИ ЕЛЕКТРОНАМИ

Р е з ю м е

Спектроскопічним методом в умовах пучків іонів та повільних електронів, що перетинаються під кутом 90°, досліджено ефективні перерізи *s*- і *d*-іонізації іона In^{+}. Описано методику визначення абсолютних величин парціальних перерізів іонізації іонів електронним ударом. Визначено абсолютні перерізи *s*- і *d*-іонізації іона In^{+} електронним ударом, які при енергії електронів 100 еВ дорівнюють $2{,}1 \cdot 10^{-17}$ см^2 та $5{,}2 \cdot 10^{-17}$ см^2, відповідно. Отримані дані свідчать про важливу роль процесу збудження-автоіонізації, який дає суттєвий внесок у переріз іонізації іонів електронним ударом.