THE ROLE OF AUTOIONIZING STATES IN ELECTRON-IMPACT EXCITATION OF THE λ230.6 nm INTERCOMBINATION LINE OF AN INDIUM ION

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The electronic excitation function of the intercombination line λ 230.6 nm of an In$^+$ ion is first investigated by the spectroscopic method in the energy range from the threshold to 100 eV using ion and electron beams crossing at the right angle. It is established that the defining mechanism of the excitation of this line is a resonant excitation of the "electron + ion" system accompanied with the generation of atomic autoionizing states (AIS) and their following electron decay into the $^3P_1^0$ level of an In$^+$ ion. It is determined that, in the energy region of the spin-orbit splitting of excited levels, the dominant contribution to the resonance excitation is made by the Coster–Kronig process. It is discovered that, at the electron energies higher than the fivefold threshold, a decrease of the excitation function does not correspond to the law E^{-3} characteristic of intercombination transitions.

1. Introduction

The investigation of the phenomena accompanying inelastic collisions of electrons with ions represents a source of information about the mechanisms of their interaction and the processes of excitation, ionization, and recombination of ions. Data obtained in the course of these researches find a wide application for astrophysics, analytic engineering, laser physics, development of new technical devices and technologies. In the case of an In$^+$ ion, the interest in such data grows in view of using the $5\ ^3P_1^0 \rightarrow 5\ ^1S_0$ intercombination transition as a source of the optical frequency standard [1].

At present, the excitation of intercombination lines in electron-ion collisions is investigated for a finite number of singly charged ions due to their very weak intensity. In the case of heavier many-electron ions, the probability of the excitation of intercombination lines increases, which is associated with a significant rise of the role of relativistic, correlation, and resonance effects. In particular, the effective cross section for the excitation of the resonance intercombination transition of a Li$^+$ ion [2] reaches a maximum near the threshold and amounts to only $\sim 10^{-18}$ cm^2. Starting from the energy equal to the ionization potential of a Li$^+$ ion, the investigated energy dependence varies according to the law E^{-3} characteristic of intercombination transitions. The discovered resonance contribution of AIS to the effective excitation cross section was insignificant, which was also confirmed by the results of theoretical calculations [3]. However, according to our previous investigations [4, 5], in the case of a heavier many-electron Tl$^+$ ion, the effective cross section for the electronic excitation of the intercombination line (λ190.8 nm) appears to be of the same order of magnitude ($\sim 10^{-16}$ cm^2) as that of the excitation of the resonance line (λ132.2 nm), which is conditioned by the dominant contribution of resonance processes to the population of the $6^3P_1^0$ level. Investigating the energy dependence of the effective excitation cross section (the excitation function) of the intercombination line of a Tl$^+$ ion, we discovered complicated structural peculiarities (including those above the ionization potential of the ion), where the behavior of the excitation function is deviated from the law E^{-3}. Theoretical calculations of the collision strengths for the intercombination transitions of an Al$^+$ ion performed by the R-matrix method also confirm that the principal contribution to the collision strengths for such transitions is made by resonance processes at the expense of the electron decay of AIS, especially at low electron energies [6].

It's worth noting that, in the case of ions having a charge of higher multiplicity, an important part is played by relativistic interactions and correlation effects, the same way as in the inner shells of many-electron singly charged ions. This results in the fact that, along with dipole transitions, the probability of transitions of other multiplicities significantly increases; moreover, in the case of the excitation of transitions of such a kind, the contribution of resonance processes to the effective excitation cross section dominates as compared to the case of direct excitation [7—8].

A deeper understanding of the physics of spin-exchange processes requires systematical investigations of the electronic excitation of ions, especially within one group of the Mendeleev Periodic table of chemical elements. That's why the experimental investigation of the excitation of the intercombination line of an In^+ ion belonging to group III like a Tl^+ ion, is of significant scientific interest.

In the present paper, we discuss the results obtained in the course of spectroscopic investigations of the energy dependence of the effective cross section for the excitation of the intercombination line λ 230.6 nm of an In^+ ion in the energy range from the excitation threshold to 100 eV:

$$\begin{aligned}
&\mathrm{e} + \mathrm{In}^+(4d^{10}5s^2)^1S_0 \rightarrow \\
&\rightarrow \quad \mathrm{In}^{+*}(4d^{10}5s5p)^3P_1^0 + \mathrm{e}_1 \\
&\qquad \downarrow \\
&\qquad \mathrm{In}^+(4d^{10}5s^2)^1S_0 + h\nu(\lambda 230,6\ \text{нм})
\end{aligned} \tag{1}$$

It's worth noting that investigations of such a kind represent a very complicated experimental problem. In particular, it is conditioned by the following reasons: first, the saturation vapor pressure in an ion source necessary for obtaining a stable beam of In^+ ions is reached at high temperatures (800 — 1000 °C), while the melting temperature for indium amounts to 156 °C; secondly, at so high temperatures, indium metal is chemically aggressive, which results in the disruption of some structural elements of the ion source and the intensive formation of the liquid phase of the metal on ceramic insulators.

2. Experimental Apparatus

The investigations were performed on the experimental setup consisting of an ion source, ion-optical system, ninety-degree electrostatic ion selector, system for the vertical focusing of an ion beam, cooled atom trap, three-anode electron gun with a ribbon indirectly heated cathode, and electron and ion collectors. A detailed description of the setup is given in [9].

Specific peculiarities of the execution of experiments with indium metal put strict demands to the construction of the ion source and the ion-optical lens system as well as to the choice of the parameters of the experiment which are optimal for performing the precise investigations for a long time. In particular, the principal factor of the stable operation of the ion source lies in the choice of the way to obtain positive singly charged ions of the investigated element. In order to obtain a stable beam of In^+ ions, we considered two ways of their generation: in the discharge and on a heated surface. Since indium ions are generated in the lower metastable $^3P^0_{0,2}$ states with a high probability when the ion source is operating in the discharge mode, we have chosen the means of ionization of indium atoms on a heated tantalum surface, which gives a possibility to exclude the formation of the above-mentioned states. In order to obtain a stable beam of In^+ ions of the sufficient intensity, we designed various types of the ion source where positive ions were generated on a heated surface. The most effective construction of the ion source appeared to be the one with an ionizer in the form of a concave spherical tantalum surface heated with the help of a tungsten spiral.

Under the vacuum conditions of the order of $\sim 10^{-8}$ Torr, an ion beam (having a cross section of 2×2 mm^2, an energy of $E_i = 700$ eV, and a current of $I_i = (1.0\div 1.5)\times 10^{-6}$ A) crossed an electron beam (with a cross section of 1×8 mm^2, an energy of $E_e = (5\div 100)$ eV, a current of $I_e = (7\div 10)\times 10^{-5}$ A) at a right angle in the equipotential collision region.

The electron gun operated in the mode of longitudinal compression: electrons emitted by the heated surface of the oxide cathode were first accelerated in the gap "the cathode – the second anode" and then decelerated in the gap "the second anode – the third anode". Important characteristics of the electron gun are the dependence of the current on the accelerating voltage (the volt-ampere characteristic) and the electron energy distribution function.

In order to obtain reliable data upon investigating the electronic excitation of a spectral line, it is necessary to choose the operating parameters of the electron gun in such a way that the volt-ampere characteristic saturates at the energy corresponding to the excitation threshold of this line. For this purpose, we have performed a number of test measurements of the volt-ampere characteristics (for various adjusting parameters of the electron gun). Some of these characteristics are depicted in Fig. 1,*a*. In particular, in order to investigate the electronic excitation of the intercombination line of an In^+ ion, we have chosen the adjusting parameters of the electron gun that correspond to the volt-ampere characteristic given in Fig. 1,*a* (curve *2*). The electron energy distribution curve for such an adjustment of the electron gun is represented in Fig.1,*b*. The monoenergeticity of the electron beam was determined as the width of the electron energy distribution curve at its half-height. This curve was obtained by differentiating the dependence of the electron current on the retarding potential. As was established, the monoenergeticity in the beam is greatly influenced by, first, a potential sag in the interanode gaps and, secondly, the value of the electron current I_e. The smallest energy spread of electrons ($\Delta E_{1/2} \sim 0.3 \div 0.4$ eV) can be reached at low values of the potentials across the anodes of the electron gun and at the currents of the electron beam $I_e = 10 \div 30$ μA. However, under real experimental conditions, the registered signals from the investigated processes accompanying electron-ion collisions are superweak, which is connected, first of all, to the low ($10^6 \div 10^7$ cm^{-3}) concentrations of interacting particles. That's why, to obtain a useful signal of the required level, one needs higher values of I_e. This results, in turn, in worsening the monoenergeticity. The above-stated facts imply that, when adjusting the electron gun, it is necessary to find a reasonable compromise in choosing the magnitude of the electron current I_e so as to retain the required monoenergeticity of the electron beam. As one can see from Fig.1,*b*, the magnitude of the monoenergeticity of the electron beam in our investigations amounts to $\Delta E_{1/2} \sim 0.6$ eV.

One of the important stages of investigating the electronic excitation function of a spectral line lies in calibrating the energy scale. The accuracy of determining the electron energy depends on the contact potential difference, the decelerating influence of the space charge of the electron beam, and the accelerating action of the ion beam on electrons of the beam. While investigating the intercombination line of an In^+ ion, we determined the contact potential difference which was approximately equal to 2.1 eV as well as the magnitude of the decelerating influence exerted by the space charge of the electron beam, ~ 0.3 eV (at $I_e = 80$ μA), and that of the accelerating action of the ion beam, ~ 0.4 eV (at $I_i = 1$ μA). With regard for these factors, the calibration of the energy scale of electrons is performed to within ± 0.1 eV.

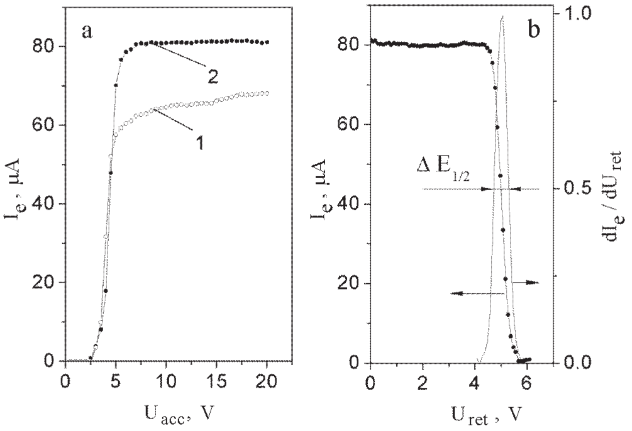

Fig. 1. Electron gun characteristics: *a* — the volt–ampere characteristics at the accelerating voltage $U_{\text{acc}} = 7$ V and the voltages across the first and second anodes $U_1 = 4$ V and $U_2 = 20$ V (curve *1*), $U_1 = 5$ V and $U_2 = 35$ V (curve *2*); *b* — the electron retardation curve (points) and electron energy distribution (solid line)

The investigations of the excitation function of the intercombination line of an In^+ ion by electron impact lay in measuring the relative intensity of radiation on the wavelength $\lambda 230.6$ nm at various values of the electron energy. The spectral separation of radiation was performed with the help of a seventy-degree vacuum monochromator constructed following the Seya—Namioka scheme with a concave toroidal grating (1200 groove/mm) and the reciprocal linear dispersion $d\lambda/dl \sim 1.7$ nm/mm. Radiation was detected with the help of a solar blind photoelectric multiplier. The data accumulation time at each experimental point was equal to 2000 s, while the magnitude of the useful signal — $1 \div 0.2$ pulse/s, with the relationship signal/background varying from 1/10 to 1/30. The measurements and the processing of experimental data were automatized using a computer.

It's also worth noting that a comparatively long lifetime of the excited ${}^3P_1^0$ level ($\tau = 4.4 \times 10^{-7}$ s [10]) results in a sufficient remove of the investigated radiation from the observation region, which requires a long time for the accumulation of data at every point.

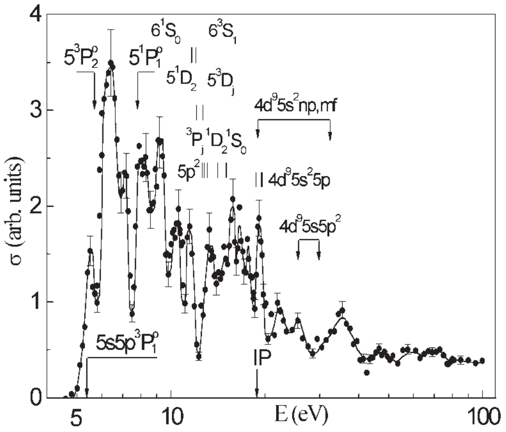

Fig. 2. Electronic excitation function of the resonance intercombination line λ230.6 nm ($5s5p\ ^3P_1^0 \rightarrow\ 5s^2\ ^1S_0$) of an In^+ ion

3. The Results and Their Discussion

An In^+ ion represents a many-electron atomic system with relativistic and correlation effects being essentially expressed. That's why, along with dipole transitions, spin-forbidden ones are also effectively excited. It is proved by the results which we obtained from precisely investigating the energy dependence of the effective cross section for the electronic excitation of the intercombination line of an In^+ ion on the wavelength λ230.6 nm ($5s5p\,^3P_1^0 \rightarrow 5s^2\,^1S_0$). The results of investigations are given in Fig. 2. Here, we also indicate the mean-square error of measurements (±10 %), the excitation threshold of the investigated line, and the ionization potential and the excited levels of an In^+ ion, to which the infinite series of AIS of an indium atom converge. As one can see from the figure, the excitation function of the intercombination line of an In^+ ion has clear structural features in the energy range from the excitation threshold to 40 eV. It is known that, in electron-ion collisions, the electron decay of AIS, which were created due to a resonant capture of the incoming electron by an ion with the simultaneous excitation of one of its bound electrons, results in a resonant excitation of an ion and additionally contributes to the effective cross sections for the direct excitation:

$$\mathrm{e} + \mathrm{In}^+(4d^{10}5s^2)^1S_0 \rightarrow$$

$$\begin{aligned} \rightarrow\ & \mathrm{In}^{**}(4d^{10}nln_1l_1n_2l_2, 4d^9nln_1l_1n_2l_2n_3l_3) \\ & \downarrow \\ & \mathrm{In}^{+*}(4d^{10}5s5p)^3P_1^0 + \mathrm{e}_1 \end{aligned} \qquad (2)$$

It's worth noting that, in this case, the resonant excitation dominates over the direct one, that's why it is impossible to determine the nature of the direct process. However, the obtained results testify to that a decrease of the energy dependence of the effective cross section for the excitation of the intercombination line of an In^+ ion (just as for the case of a Tl^+ ion) deviates from the law E^{-3} characteristic of spin-forbidden transitions.

Since an In^+ ion is characterized with a substantial splitting of energy levels, there appears a possibility for the effective electron decay of atomic AIS positioned in the energy range of level splitting without changing the principal and orbital quantum numbers, i.e. for the decay through the Coster–Kronig process. That's why the first maximum of the excitation function of the In^+ intercombination line can be most probably explained by the contribution of the AIS of the $5s5p(^3P_2^0)nl$ configuration [11], which lie in the energy interval (ΔE = 0.31 eV) between the $^3P_1^0$ and $^3P_2^0$ levels:

$$\mathrm{In}^{**}[5s5p(^3P_2^0)nl] \rightarrow \mathrm{In}^{+*}[5s5p^3P_1^0] + \mathrm{e}'. \qquad (3)$$

The second (dominant) maximum is caused by the contribution of the AIS of the $5s5p(^1P_1^0)nl$ configuration [11] lying in the energy region (ΔE = 2.44 eV) between the $^3P_1^0$ - and $^1P_1^0$ levels:

$$\mathrm{In}^{**}[5s5p(^1P_1^0)nl] \rightarrow \mathrm{In}^{+*}[5s5p^3P_1^0] + \mathrm{e}''. \qquad (4)$$

We established that the predominant contribution to the population of the $^3P_1^0$ level is made by the AIS of the $5s5p(^1P_1^0)np$ configuration with $n = 6\div8$.

The structure above the excitation threshold of the $5^1P_1^0$ level (7.81 eV) is conditioned by the contribution of the AIS that converge to both ordinary $5snl\ ^{1,3}L_j$ [12], displaced $5p^2$ ($^3P_{0,1,2}$, $^1D_2$, $^1S_0$) [12], and the Batler's $4d^95s^25p$ [12—14], $4d^95s5p^2$ [15] levels of an In^+ ion:

$$\mathrm{e} + \mathrm{In}^+(4d^{10}5s^2)\ ^1S_0 \rightarrow \begin{bmatrix} \mathrm{In}^{**}(4d^{10}5snln_1l_1) \\ \mathrm{In}^{**}(4d^{10}5p^2nl) \\ \mathrm{In}^{**}(4d^95snln_1l_1n_2l_2) \end{bmatrix} \rightarrow$$

$$\rightarrow \mathrm{In}^{+*}(4d^{10}5s5p)\ ^3P_1^0 + \mathrm{e}''. \qquad (5)$$

It's worth noting that, in this case, the most essential contribution to the effective cross section of the investigated line is also made by the AIS that decay through the Coster—Kronig process. As the indication of such a decay, one can cite the maxima of the excitation function lying between the $5s6s\,^3S_1$ and $5s6s\,^1S_0$ levels and those between $5p^2\,^3P_1^0$ and $5p^2\,^1D_2$ ones, respectively.

Starting from the energy of 11.64 eV, there arise new channels of populating the $^3P_1^0$ level at the expense of cascade transitions from ordinary excited, displaced, and Batler's levels. We should mention that, above the ionization potential of In$^+$, the excitation function of the intercombination line also reveals a structure. An especially clear maximum at an energy of 19 eV is most probably associated with the cascade-like populating of the $^3P_1^0$ level from the levels of the $4d^9 5s^2 5p$ configuration as well as with a decay of the AIS that converge to the former. A lack of data concerning the AIS of an indium atom complicates the interpretation of the discovered structure in the region above the ionization potential of an In$^+$ ion. However, there exist at least three processes responsible for its origin: the excitation of the internal electron of an ion and the formation of Batler's levels; the simultaneous excitation of the both valence electrons and the formation of displaced terms; and the decay of the AIS, whose convergence limit is presented by the indicated ion levels. As for a deviation of the decrease of the excitation function from the law E^{-3} characteristic of intercombination transitions, this fact is connected to the essential resonant contribution of atomic and ion AIS of indium, as well as to the configuration mixing of the $^3P_1^0$ and $^1P_1^0$ levels of an In$^+$ ion.

4. Conclusions

A modernization of the ion source and an improvement of the experimental technique gave a possibility to investigate, for the first time, the excitation of the resonant intercombination line λ230.6 nm ($5^3P_1^0 \to 5^1S_0$) of an In$^+$ ion by electron impact in the energy range from the threshold to 100 eV. The results of investigations demonstrate that this line is effectively excited in a wide energy interval at the expense of the essential contribution of AIS to populating the $5^3P_1^0$ level (resonant excitation), in particular, through the Coster–Kronig process. In the near-threshold range of electron energies, this process makes a dominant contribution to the effective cross section of the excitation of the line under study. The mechanism of excitation of the intercombination line of an In$^+$ ion is complicated by the simultaneous running of various processes: the direct excitation of the ion from the ground state to an excited one with a change of the spin, which is connected to the violation of the Russell—Saunders coupling due to a significant increase of the role of relativistic and correlation effects; the resonance excitation of an ion associated with the formation and decay of AIS; as well as cascade effects.

At present, theoretical calculations of the excitation of an In$^+$ ion by electron impact are absolutely absent in the literature, which does not give an opportunity to compare the experiment and theory for a more detailed analysis of the excitation of spin-forbidden transitions. The obtained experimental results can stimulate such calculations, in which both electron correlation and relativistic corrections will be simultaneously accounted for in order to adequately describe the electron scattering by In$^+$ ions.

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РОЛЬ АВТОІОНІЗАЦІЙНИХ СТАНІВ В ЕЛЕКТРОННОМУ ЗБУДЖЕННІ ІНТЕРКОМБІНАЦІЙНОЇ ЛІНІЇ λ230,6 нм ІОНА ІНДІЮ

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Резюме

Спектроскопічним методом в умовах іонного й електронного пучків, що перетинаються під прямим кутом, вперше досліджено функцію електронного збудження інтеркомбінаційної лінії λ230,6 нм іона In^{+} в енергетичному інтервалі від порога збудження до 100 еВ. Встановлено, що визначальним механізмом збудження цієї лінії є резонансне збудження системи “електрон+іон” з утворенням атомарних автоіонізаційних станів з наступним їх електронним розпадом на $^3P_1^0$-рівень іона In^{+}. Визначено, що в енергетичному інтервалі спін-орбітального розщеплення збуджених рівнів домінуючий внесок в резонансне збудження дає процес Костера—Кроніга. Виявлено, що при енергіях електронів, більших за п’ятикратне порогове значення, спад функції збудження не відповідає закону E^{-3}, характерному для інтеркомбінаційних переходів.