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Excitation-autoionization of the $5p^6$ subshell in Ba atoms[⋆]

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Abstract. The excitation-autoionization cross section for the $5p^6$ subshell in Ba atoms was determined in an incident electron energy range from the lowest autoionization threshold at 15.61 eV up to 600 eV. The data were obtained by measuring the total intensity of ejected-electron spectra arising from the decay of the $5p^5n_1l_1n_2l_2n_3l_3$ autoionizing states. The energy behavior of the cross section is characterized by the presence of a strong resonance structure in the near-threshold energy region of 15.6–22.7 eV as well as a broad maximum around 80 eV. The cross section reaches its maximum value of $(3.2 \pm 1.0)\times10^{-16}$ cm^2 at 17.4 eV, which makes out a contribution of up to 25% to the total single-ionization cross section of Ba atoms. The role of particular configurations in formation of the cross section is considered on the basis of the spectroscopic classification and excitation dynamics of the $5p^5n_1l_1n_2l_2n_3l_3$ autoionizing states.

1 Introduction

Excitation-autoionization (EA) is an important indirect ionization process which can significantly contribute to the electron-impact ionization of atoms and ions. The role of EA becomes particularly remarkable when inner closed electron subshells get involved [1,2]. In alkaline earth atoms, namely in Ca and Sr, indirect ionization through excitation of the outer p^6 subshell provides the main contribution to the total ionization cross section [3–5]. In Mg atoms, however, only a contribution from the double-excited states $2p^6n_1l_1n_2l_2$ was observed [6]. The single ionization cross section of Ba atoms differs from that for all other alkali-earth atoms by the presence of two strong maxima at approximately 9 and 22 eV [3–5,7]. To the present, no direct investigation of the origin of these features has been performed and the general role of the $5p^6$ EA processes in the total ionization of Ba atoms by electron impact remains unclear.

Until recently, experimental data about EA processes mostly confined to observing step features in ionization cross sections above the excitation threshold of inner subshells [8,9]. The integral nature of such data neither provides information on absolute value of EA cross sections nor sheds light on the role of particular configurations or groups of levels involved in a given EA process. Meanwhile, the direct studies of EA processes can be performed by measuring the spectra of ejected electrons produced within the decay processes of autoionizing states (AIS). The intensity I_{if} of a line arising from the decay of an atomic AIS i with formation of the ion in a state f, is related to the excitation cross section σ_i of this AIS by the following expression [10,11]:

$$I_{if} \sim \frac{\sigma_i}{4\pi} B_{if} C_K, \qquad (1)$$

where B_{if} is the Auger decay branching ratio for AIS i and C_K is the asymmetry parameter characterizing the angular distribution of ejected electrons.

Since the accurate spectroscopic identification of the lines unambiguously determines the decay channels of the atomic AIS, then, according to expression (1), the cross section $\sigma_i(E)$ is determined as the sum of normalized intensities of lines reflecting all Auger decay channels of the state i in ejected-electron spectrum measured at the impact-energy value E. In this case, the EA cross section for a given subshell is determined as the sum of the cross sections σ_i of all AIS belonging to this subshell and possessing the excitation thresholds lower than E. Thus, having a set of ejected-electron spectra measured over a broad impact energy range and performing spectroscopic identification of the observed lines, it is possible to obtain, with an acceptable accuracy, the energy dependence of the EA cross section (autoionization function) for a certain

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atomic subshell. Such data allows to consider in detail the role of particular configurations or groups of levels in the EA contribution under study. The described method was successfully employed earlier for obtaining the autoionization functions for the $1s^2$ subshell in Li [12] and for np^6 subshells in Na ($n = 2$), K ($n = 3$), Rb ($n = 4$) and Cs ($n = 5$) (see [13] and references therein).

The present work, being a continuation of our investigations of the electron impact excitation of the $5p^6$ subshell in Ba atoms [11], reports on studies of EA contribution of the $5p^6$ subshell to the single ionization cross section of Ba atoms by electron impact. It is organized as follows. In Section 2, we briefly present the experimental procedure and some details of the theoretical approach. Section 3 gives a discussion of the experimental and calculated EA cross sections as well as a comparison with the known ionization data. Some general conclusions are drawn in Section 4.

2 Experimental and computational aspects

The measurements of the ejected-electron spectra of Ba atoms were carried out employing the electron spectrometer setup and the method both described in detail earlier [11,14]. The former consists of a source of the incident electron beam, an electron energy analyzer (of 127° electrostatic type), and an atomic beam source. To minimize the influence of the anisotropy of the angular distribution of ejected electrons, the measurements were carried out at a "magic" observation angle of 54.7° [10]. The incident and ejected-electron energy resolutions (FWHM) were about 0.4 eV and 0.07 eV, respectively. Below 21.5 eV impact energy the increment step was 0.1 eV.

The excitation dynamics of lines is demonstrated by the spectra shown in Figure 1. As can be seen, the total intensity of the spectrum at 19.4 eV is determined by the two groups of lines 1–13 and 22–38. According to the spectroscopic classification of ejected-electron spectra of Ba atoms [11], most of these lines correspond to the decay of the $5p^5n_1l_1n_2l_2n_3l_3$ atomic AIS formed by the dipole-forbidden transitions from the ground state of the atom. The spectrum at 80 eV has a more complex structure, since it contains, besides the atomic lines 1–63, also the lines I_1–I_6 reflecting the decay of the ionic AIS $5p^5n_1l_1n_2l_2$ with excitation thresholds above 21 eV. Lines 23, 28, 39, 40, 49, 53, 60 correspond to the decay of dipole-allowed atomic AIS [11]. By measuring the intensities of lines I_{if} (1) in spectra at different impact energies, the excitation functions for some low-lying AIS were obtained in the whole impact energy range under study. The relative experimental data were put on the absolute scale by normalizing the excitation function of the $5p^55d6s^2$ $^3D_1$ state to the calculated cross section at 600 eV.

By employing the method [13,15] described in Section 1, the $5p^6$ EA cross section of Ba atoms was determined as the normalized total intensities of ejected-electron spectra measured at different impact energies. However, one feature of the use of this method for barium should be emphasized. The fact is that due to the low ionization

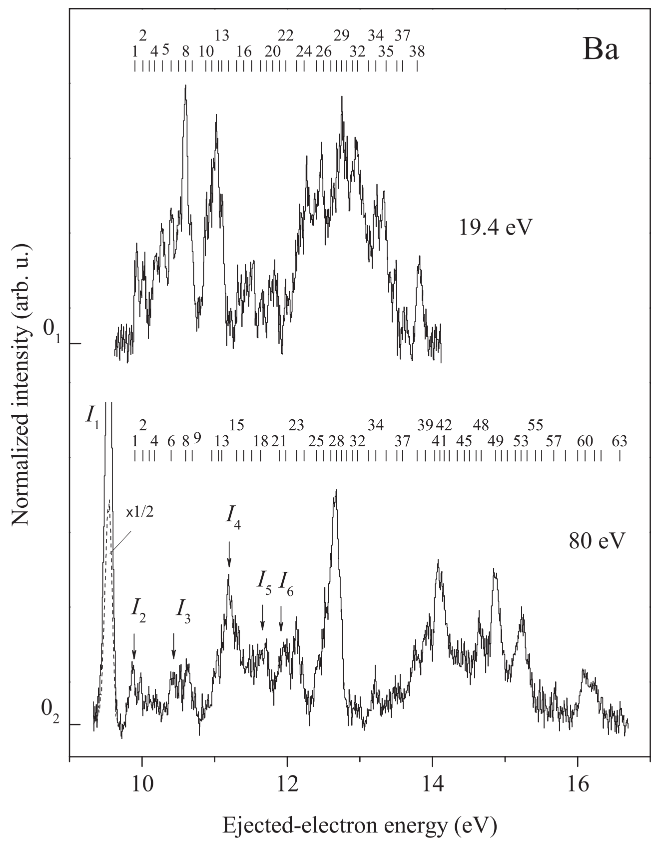

Fig. 1. Ejected-electron spectra of Ba atoms arising from the decay of the $5p^5n_1l_1n_2l_2n_3l_3$ AIS measured at 19.4 and 80 eV impact energies. The spectra are presented with a subtracted continuous background approximated by a polynomial function. Bars on top mark the energy position of ejected-electron lines considered in the present work. The indexation is given according to [11].

threshold of the $5p^6$ subshell at 21 eV [16] an additional decay channel opens for the $5p^5n_1l_1n_2l_2n_3l_3$ high-lying atomic AIS, namely

$$\mathrm{Ba}^*(5p^5n_1l_1n_2l_2n_3l_3) \rightarrow \mathrm{Ba}^{+*}(5p^5n_1l_1n_2l_2) + e_{ejc}. \quad (2)$$

As was shown earlier [11], just this decay channel is responsible for the low intensity or even total absence, in ejected-electron spectra, of the lines associated with the decay of atomic AIS with excitation thresholds above 22 eV. Thus, the EA cross section obtained from the spectra at impact-energy values beyond 22 eV will be lower than the total EA cross section of the $5p^6$ subshell. Since the subsequent decay of the ion states $5p^5n_1l_1n_2l_2$ contributes to the cross section for double ionization of Ba atoms [17], the efficiency of decay channel (2) can be estimated by analyzing the behavior of this cross section in the energy region above 22 eV. The comparative analysis of the total and double ionization cross sections [7] has shown that in the energy range 21–30 eV the fraction of the decay channel (2) in the total $5p^6$ EA cross section should not exceed 10%.

Taking into account the statistical uncertainty of the line intensity measurements and in the fluctuations of the experimental conditions, the total relative uncertainty in determining the EA cross section did not exceed 30% in the region up to 7 eV above the excitation threshold of the $5p^6$ shell (15.61 eV [11]) and 25% at higher impact energies. The obtained cross section in relative units was put on an absolute scale by normalizing to the excitation cross section of the state $5p^5 5d6s^2\ {}^3\mathrm{D}_1$ (line 28) calculated at 600 eV impact.

The theoretical EA cross section of the $5p^6$ subshell was obtained as a sum of calculated excitation cross sections of 58 atomic $5p^5 n_1 l_1 n_2 l_2 n_3 l_3$ AIS with excitation thresholds below 22 eV. The cross sections were calculated in relativistic distorted wave approximation (RDW) by using radial wave functions obtained in the standard Dirac-Fock-Slater method. The Flexible Atomic Code [18] and the relativistic jjJ coupling scheme of angular momenta were used. The detailed description of calculations, including details on the decay channels and the spectroscopic classification for all mentioned states has been presented earlier [11].

3 Results and discussion

3.1 $5p^6$ excitation-autoionization

Figure 2a shows the EA cross section σ_{EA} for the $5p^6$ subshell in the impact energy range 10–600 eV. As can be seen, the behavior of the cross section has a pronounced resonance character at low impact energies. This part of the cross section is shown in the inset, where the maxima a, b, c are clearly visible at 17.4, 19.6 and 21.4 eV, respectively. The cross section reaches its maximum value of $(3.2 \pm 1.0) \times 10^{-16}\,\mathrm{cm}^2$ at 17.4 eV impact energy (maximum a). A sharp decrease in the cross section above 22 eV is due to the opening of an additional decay channel (2) for high-lying atomic AIS. Above 40 eV, the cross section increases slowly, forming a broad maximum around 80 eV. With further increase in impact energy, the cross section decreases monotonically to the value of $(0.4 \pm 0.1) \times 10^{-16}\,\mathrm{cm}^2$ at 600 eV.

By definition, each point in the EA cross section is the sum of excitation cross sections for individual atomic AIS at given impact-energy value E. Thus, the energy dependence $\sigma_{\mathrm{EA}}(E)$ is, in fact, the resultant excitation function of the whole set of the AIS observed in ejected-electron spectra. Therefore, for its analysis, it is first necessary to consider the excitation cross sections of individual states.

In Figures 2b and 2c, the most typical experimental and calculated excitation functions for dipole-forbidden (Fig. 2b) and dipole-allowed (Fig. 2c) triplet states from the $5p^5 5d6s^2$ configuration are shown. In all experimental cross sections, the main maxima are located at low impact energies, which fully corresponds to the spin-exchange character of the excitation of these states. The presence of the “fine” structure just above the excitation thresholds of the states (see inserts) points out the effective formation of negative-ion resonances. Note that our preliminary data on excitation functions for triplet and singlet states

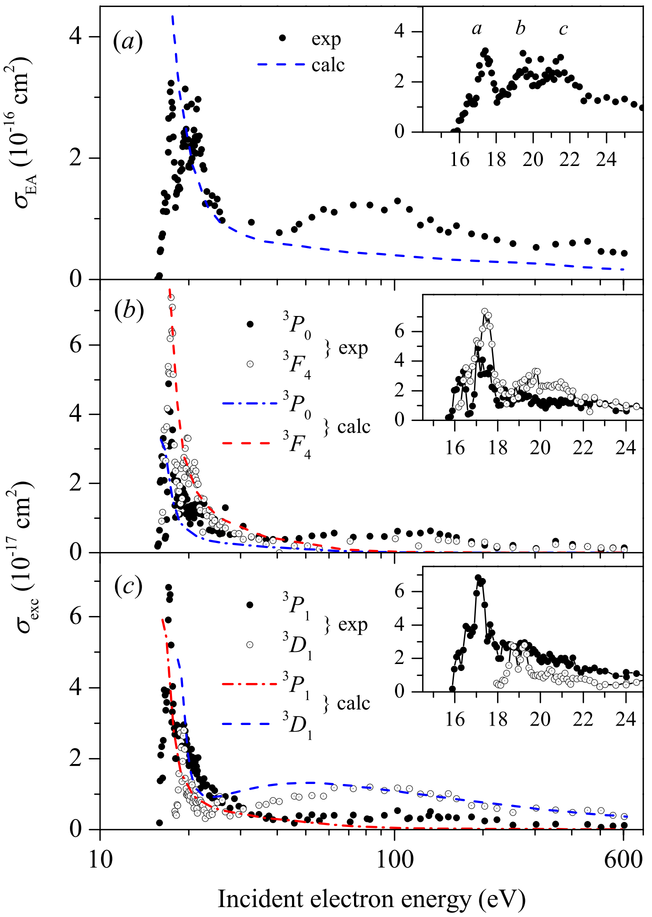

Fig. 2. The experimental and calculated EA cross section σ_{EA} (a) and excitation cross sections σ_{exc} of dipole-forbidden (b) and dipole-allowed (c) AIS from the $5p^5 5d6s^2$ configuration of Ba atoms. Inserts show the near-threshold parts of the experimental cross sections.

in $5p^5 5d^2 6s$, $5p^5 6s^2 6p$ and other high-lying configurations (to be published in a separate paper) show a similar excitation behavior. Thus, the observed resonance shape of the measured EA cross section at low impact energies reflects the purely correlation nature of the $5p^6$ excitation process in barium, namely, through the spin-exchange interaction and the formation of negative-ion resonances. The dominance of the maxima a–c in the entire investigated impact-energy range shows that the latter process is the main excitation channel for the $5p^6$ subshell. It should be noted here that a similar pattern of excitation was previously found for the $5p^6$ subshell in the cesium atom [19].

In order to find the role of particular configurations or groups of states in formation of the EA cross section, it is necessary to consider the energy structure and spectroscopic classification of the $5p^6$-core excited states in barium [11]. As can be seen from the inset in Figure 2a, the near-threshold maxima a–c are located within the energy region 15.6–21.8 eV where the $5p^5 n_1 l_1 n_2 l_2 n_3 l_3\ LSJ$ AIS are known in Ba atoms (see Tab. 3 in [11]). As follows from the spectroscopic classification of these states and from the analysis of the excitation dynamics of ejected-electron spectra in the entire impact-energy region under study, the maximum a of the EA cross section is formed by the resonances present in electron impact excitation of 25 states from $5p^55d6s^2$, $5p^55d^26s$ and $5p^56s^26p$ configurations which are located between 15.6 and 18.0 eV. The origin of the maximum b is due to the resonance excitation of 19 states from configurations $5p^55d^26s$, $6p$, $6d$, $5p^55d6s6p$, $7s$, $5p^55d6s^2$ and $5p^56s^26p$, $7s$ lying between 18.1 and 20.1 eV. The maximum c is a superposition of near-threshold resonances in the electron-impact excitation of 14 states from $5p^55d^26s$, $6d$, $7d$, $5g$, $5p^55d6s6d$, $7d$, $5f$ and $5p^56s^27d$ configurations located between 20.2 and 21.8 eV. Finally, the broad maximum around 80 eV consists mainly of contributions from the decay of dipole-allowed states in $5p^55d6s^2$, $5p^55d^26s$ and $5p^55d6s7s$, $7d$ configurations.

A comparison of the calculated and experimental cross sections shown in Figures 2a–2c shows a noticeable discrepancy between them in the whole impact energy range under study. At low impact energies, the reason for this is the use of the DW approximation in the calculations. It usually gives too large cross sections at low impact energies when channel coupling becomes more important. Meanwhile, the importance of taking close-coupling effects into account for barium has previously been well demonstrated in calculations of the electron-impact excitation of the valence $6s^2$ shell [20,21]. Undoubtedly, much stronger influence of these effects one should expect when the $5p^6$ subshell is excited.

A noticeable difference in the experimental and calculated EA cross sections observed above 40 eV shows that the theory underestimates the excitation efficiency of both dipole-allowed and dipole-forbidden states at high impact energies. It is currently difficult to say anything specifically about the reason for this situation, since the analysis of all our data on the measured excitation cross sections has not yet been completed. It is also necessary to check the sensitivity to the one-electron basis using for calculation of the energy spectrum and the scattering cross sections. We are planning additional calculations with more extended basis what perhaps help us to clarify the situation. In this regard, it is interesting also to note that a similar discrepancy between experimental and calculated excitation cross sections at medium and high impact energies was observed earlier in the simpler case of electron excitation of the $4p^6$ shell in krypton [22].

3.2 Autoionization and ionization

In Figure 3, the obtained EA cross section is shown together with the experimental single ionization cross sections of Ba atoms [3,7]. Note that the relative data [3] were normalized at the first maximum to the absolute data [7]. Both ionization cross sections show two prominent maxima at around 9 eV and 22 eV impact energies. The position and shape of the high-energy maximum correlate well, both in shape and magnitude, with the EA cross section. In particular, two autoionization thresholds in the EA cross section at 15.61 and 18.4 eV (see dashed lines) associated with the excitation of the lowest and most

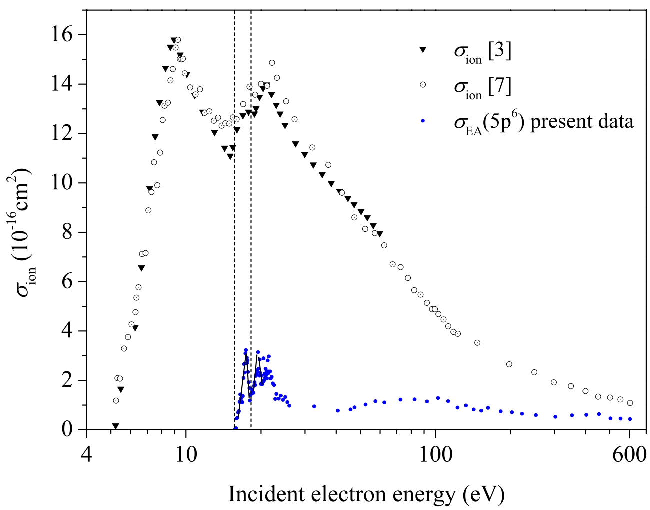

Fig. 3. The single ionization $\sigma_{\rm ion}$ cross sections [3,7] and the EA cross section $\sigma_{\rm EA}(5p^6)$ (present data) of Ba atoms. Vertical dashed lines mark the $5p^6$ autoionization thresholds at 15.61 eV and 18.4 eV [11].

efficiently excited configurations $5p^55d6s^2$, $5p^55d^26s$ and $5p^56s^26p$, respectively, can be observed in both ionization data too. The scatter of data [7] between 21 and 23 eV and subsequent slow decrease of the cross section reflect the opening of the additional decay channel (2) for high-lying atomic AIS with formation of ionic states in $5p^55d6s$ and $5p^55d^2$ configurations [23,24]. Therefore, the maximum of the single ionization cross section at 22 eV is attributed exclusively to the EA processes involving excitation of the $5p^6$ subshell. Comparing the cross sections also shows that in the impact-energy range 15.6–22.7 eV the relative $5p^6$ EA contribution to the single ionization cross section of Ba atoms is at least 25%.

4 Summary

We have presented the excitation-autoionization cross section for the $5p^6$ subshell in Ba atoms in the electron impact energy range from the lowest autoionization threshold at 15.61 eV up to 600 eV. The data were obtained by measuring the normalized total intensities of ejected-electron spectra arising from the decay of the $5p^5n_1l_1n_2l_2n_3l_3$ autoionizing states. The role of particular autoionizing configurations in formation of the excitation-autoionization cross section is considered. The strong resonant excitation of the lowest states in $5p^55d6s^2$, $5p^55d^26s$, $6p$, $6d$, $7d$, $5g$, $5p^56s^26p$, $7s$, $7d$ and $5p^55d6s6p$, $6d$, $7s$, $7d$, $5f$ configurations determines the similar resonant behavior of the measured cross section at low impact energies. Its shape and magnitude at high impact energies are determined by the total contribution from dipole-allowed states in $5p^55d6s^2$, $5p^55d^26s$ and $5p^55d6s7s$, $7d$ configurations.

The obtained data establish the origin of the well known maximum of the single ionization cross section of Ba atoms seen at 22 eV impact energy by attributing it exclusively to the EA processes in the $5p^6$ subshell. Still unknown origin of the resonance-like maximum of the ionization cross section at 9 eV impact energy calls for further research of the EA processes in Ba atoms involving excitation of the $6s^2$ subshell.

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Author contribution statement

Theoretical calculations were performed by Viktoria Roman and Alicia Kupliauskienė. Vladimir Borovik, Ivan Shafranyosh and Oleksandr Borovik performed measurements and processed the data. All authors contributed to the discussion of the results, writing and editing the manuscript.

References

1. K.J. Nygaard, Phys. Rev. A. **11**, 1475 (1975)
2. D. Mitnik, P. Mandelbaum, J.L. Schwob, A. Bar-Shalom, J. Oreg, W.H. Goldstein, Phys. Rev. A **50**, 4911 (1994)
3. S. Okudaira, J. Phys. Soc. Jpn. **29**, 409 (1970)
4. Y. Okuno, J. Phys. Soc. Jpn. **31**, 1189 (1971)
5. L.A. Veinshtein, V.I. Ochkur, V.I. Rakhovskii, A.M. Stepanov, Sov. Phys. JETP. **34**, 271 (1972)
6. Y. Kaneko, J. Phys. Soc. Jpn. **16**, 2288 (1961)
7. J.-M. Dettmann, F. Karstensen, J. Phys. B: At. Mol. Phys. **15**, 287 (1982)
8. G.H. Dunn, in *Electron Impact Ionization*, edited by T.D. Mark, G.H. Dunn (Springer-Verlag, New York, 1985), p. 277
9. D.L. Moores, K.J. Reed, Adv. At. Mol. Opt. Phys. **34**, 301 (1994)
10. E.G. Berezhko, N.M. Kabachnik, J. Phys. B: At. Mol. Phys. **10**, 2467 (1977)
11. V. Hrytsko, G. Kerevicius, A. Kupliauskiene, A. Borovik, J. Phys. B: At. Mol. Opt. Phys. **49**, 145201 (2016)
12. A.A. Borovik, Opt. Spectr. **109**, 319 (2010)
13. A. Borovik, A. Kupliauskiene, O. Zatsarinny, J. Phys. B: At. Mol. Opt. Phys. **46**, 215201 (2013)
14. A.A. Borovik, A.N. Grum-Grzhimailo, K. Bartschat, O. Zatsarinny, J. Phys. B: At. Mol. Opt. Phys. **38**, 1081 (2005)
15. A. Borovik, V. Roman, A. Kupliauskiene, J. Phys. B: At. Mol. Opt. Phys. **45**, 045204 (2012)
16. D. Rassi, K.J. Ross, J. Phys. B: At. Mol. Phys. **13**, 4683 (1980)
17. B. Peart, J.G. Stevenson, K.T. Dolder, J. Phys. B: At. Mol. Phys. **6**, 146 (1973)
18. M.F. Gu, Can. J. Phys. **86**, 675 (2008)
19. A. Borovik, O. Zatsarinny, K. Bartschat, J. Phys. B: At. Mol. Opt. Phys. **42**, 044010 (2009)
20. D.V. Fursa, S. Trajmar, I. Bray, I. Kanik, G. Csanak, R.E.H. Clark, J. Abdallah Jr., Phys. Rev. A. **60**, 4590 (1999)
21. O.I. Zatsarinnyi, L.A. Bandurina, V.F. Gedeon, Opt. Spectr. **97**, 499 (2004)
22. A. Dasgupta, K. Bartschat, D. Vaid, A.N. Grum-Grzhimailo, D.H. Madison, M. Blaha, J.L. Giuliani, Phys. Rev. A. **64**, 052710 (2001)
23. S.J. Rose, I.P. Grant, J.P. Connerade, Phil. Trans. R. Soc. A **290**, 527 (1980)
24. J. Nienhaus, O.I. Zatsarinny, W. Mehlhorn, Phys. Essay **13**, 307 (2000)