Theoretical investigation of dielectronic recombination of Sn^{12+} ions

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Theoretical calculations have been made for the dielectronic recombination (DR) rate coefficients of Sn^{12+} ion using a relativistic flexible atomic code with configuration interaction. Comparison of the rate coefficients for $4s$, $4p$, and $4d$ subshell excitation shows that while the $4p$ subshell excitation dominates over the whole temperature region, $4d$ subshell excitation at low temperature and $4s$ subshell excitation at high temperature cannot be neglected. In order to facilitate simple applications, the calculated DR rate coefficients are fitted to an empirical formula. The total DR rate coefficient is greater than either the radiative recombination or three-body recombination coefficients for electron temperatures greater than 1 eV. Therefore, DR can strongly influence the ionization balance of laser-produced tin plasmas.

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I. INTRODUCTION

Dielectronic recombination (DR) is a basic atomic process in electron-ion collisions. Accurate DR cross sections and rate coefficients are essential for studying the ionization balance of highly ionized ions in hot plasmas [1,2] and for calculating populations of lasing levels in research on x-ray lasers [3]. Furthermore, the dielectronic satellite (DS) decay coming from DR processes can give a significant contribution to both the apparent width and intensity of a resonance line [4,5] in a plasma. In recent years, extreme ultraviolet (EUV) light sources have been extensively pursued for next-generation lithography. Currently, the best candidate for such light sources arise from intra N shell ($n = 4$ shell) transitions or inter N - O shell transitions of tin (Sn) or xenon (Xe) highly charged ions, respectively [6]. Laser-produced plasmas of multicharged tin ions are regarded as one of the hopeful candidates for such EUV light sources [7–11]. Observations have shown that very bright emission lines from Sn^{8+} to Sn^{12+} ions lie just within the region of interest [12,13]. The DR rate coefficients for Xe ions have been calculated by Song and Kato [14] and Safronova *et al.* [15]. But to our knowledge, very little work has been reported on dielectronic recombination rate coefficients and dielectronic satellites for tin ions until now.

In order to consider the influence of DR on the ionization balance of tin ions and the contributions of DS to their spectra, as an example, Fu *et al.* [16] performed the first calculation on the DR rate coefficient and dielectronic satellite radiation from $4d^3 4fnl \to 4d^4 nl$ ($n = 4 - 15$, $l = 0 - 6$) of Sn^{10+} ions using the flexible atomic code (FAC) [17]. They found that the total DR rate coefficient has its maximum value between 10 and 100 eV and is greater than the recombination radiation (RR) and three-body recombination (TBR) rate coefficients (the number density of free electrons was assumed to be 10^{21} cm^{-3}) for the case of the electron temperature $T_e > 1$ eV. These results mean that DR can strongly influence the ionization balance in a laser-produced plasma of multicharged tin ions for the Sn^{10+} case. For the next step, in this paper we investigate the influence of configuration interaction (CI) and different excited electrons on the DR of Sn^{12+} ions using FAC. We calculate DR rate coefficients and discuss the effects of configuration interaction. We compare the rate coefficients from $4s$, $4p$, $4d$ subshell excitations, and further, we fit an empirical formula to the numerical DR rate coefficients. We also compare the rate coefficients of DR with those for RR and TBR.

II. THEORETICAL METHOD

In the isolated-resonance approximation, the DR rate coefficient from an initial state i into a final state k through an intermediate doubly excited state, for electrons in the Maxwellian distribution, can be expressed as

$$\alpha_{DR}(i,j,k;T_e) = \left(\frac{h^2}{2\pi m_e kT_e}\right)^{3/2} \frac{1}{2g_i} Q(i,j,k)\exp\left(-\frac{E_{ji}}{kT_e}\right), \quad (1)$$

where T_e is the electron temperature, m_e is the electron mass, h is Plank's constant, k is Boltzmann's constant, g_i is the statistical weight of the state i, and E_{ji} is the resonance energy. $Q(i,j,k)$ is the so-called satellite line intensity factor and is defined in terms of the radiative decay rate A^r_{jk} and the Auger decay rate A^a_{ji} by

$$Q(i,j,k) = \frac{g_j A^a_{ji} A^r_{jk}}{\sum_{k'} A^r_{jk'} + \sum_{i'} A^a_{ji'}}, \quad (2)$$

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where g_j is the statistical weight of the state j, the summation over i' runs through all the possible Auger final states, and the summation over k' runs through all the possible radiative final states. The radiative decay rate is given by

$$A^r_{jk} = \frac{4e^2\omega_p}{3\hbar c^3 g_j}|\langle\Psi_k|T^{(t)}|\Psi_j\rangle|^2, \qquad (3)$$

where Ψ_j and Ψ_k are atomic state functions (ASFs) for the states j and k, respectively, ω_p is the photon energy, and $T^{(t)}$ is a multipole radiative tensor operator. The Auger decay rate is given by

$$A^a_{ji} = \frac{2\pi}{\hbar}\left|\langle\Psi_i\varepsilon_j|\sum_{p<q}\frac{1}{r_{pq}}|\Psi_j\rangle\right|^2, \qquad (4)$$

where $\Psi_i\varepsilon_j$ is the ASF of the combined system of a bound state i and a free electron with energy ε_j.

In a practical calculation, the ASF is a linear combination of the configuration state functions (CSFs). The CSFs are antisymmetrized products of a common basis set of orthonormal orbitals. The basis sets are optimized with respect to the Dirac-Coulomb Hamiltonian. The contributions from $n = 4 - 15$ are evaluated directly, and the results are extrapolated up to $n = 100$ by the relevant scaling law $A^a \propto n^{-3}$ [18].

For comparison, the RR rate coefficient α_{RR} and TBR rate coefficient α_{3b}, which are also two important recombination processes in plasmas, are estimated simply by using formulas given by Colombant and Tonon [19]. From Eq. (5) of Ref. [19], we have

$$\alpha_{RR} = 5.2\times10^{-14}(\phi_Z/T_e)^{1/2}Z[0.429 + \tfrac{1}{2}\log_{10}(\phi_Z/T_e) + 0.469(T_e/\phi_Z)^{1/2}], \qquad (5)$$

and from Eq. (6) of Ref. [19] we have

$$\alpha_{3b} = 2.97\times10^{-27}\xi_Z/T_e\phi_Z^2(4.88 + T_e/\phi_Z), \qquad (6)$$

where ϕ_Z is the ionization potential and ξ_Z is the number of electrons in the outermost shell corresponding to the state of charge Z.

III. RESULTS AND DISCUSSION

A. Contributions of inner-shell electron excitation

The ground state of Sn^{12+} is [Ne] $3s^23p^63d^{10}4s^24p^64d^2$. In its DR processes, both $3l$ with $l = 0 - 2$ and $4l$ with $l = 0 - 2$ can be excited. Figure 1 shows the DR rate coefficients for intermediate doubly excited states $[3l^{18}4s^24p^6]^{-1}4d^37l'$ and $3l^{18}4s^24p^64d4f7l'$ ($l' = 0 - 6$). Each curve gives the sum of coefficients for all the permitted l' orbitals. Regarding the $3l$ shell excitations, it can be seen that DR rate coefficients for $3d$ subshell excitation are the largest and that the DR rate coefficients decrease with decreasing l quantum number. This result is similar to those for Co-like Pd ions obtained by Zhang *et al.* [20]. As the temperature increases, the DR rate coefficients for $3l$ shell excitation increase and gradually overlap with the DR rate coefficients for $4l$ shell excitation. The DR rate coefficient for $3d$ excitation exceeds that for $4d$ subshell excitation at $T_e = 550$ eV. This overtaking is because the DR rate coefficients depend mainly on $\exp(-E_{ji}/kT_e)$ in Eq. (1). With the increase of E_{ji}, the factor $\exp(-E_{ji}/kT_e)$ causes the curve to decline gradually. The peak positions of $3l$ are higher in energy than those of $4l$, since the energies for $3l$ shell excitations are higher than those of $4l$. For electronic temperature less than 100 eV, which corresponds to the regime employed in plasmas for EUV light sources, the DR rate coefficients for $4l$ shell excitation are much greater than for $3l$; contributions from $3l$ shell excitation can thus be neglected.

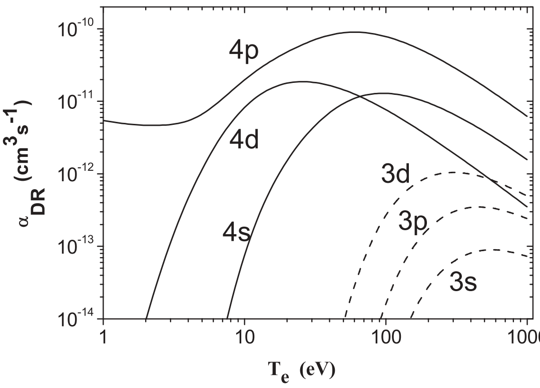

FIG. 1. DR rate coefficients for intermediate doubly excited states $[3l^{18}4s^24p^6]^{-1}4d^37l'$ and $3l^{18}4s^24p^64d4f7l'$. The notations $3s$, $3p$, and $3d$ on the broken curves stand for the respective excited M-shell orbitals. The notations $4s$, $4p$, and $4d$ on the solid curves stand for the respective excited N-shell orbitals. Each curve gives the sum of coefficients for all the permitted l' orbitals.

For $4l$ shell excitations, the DR rate coefficients for inner-shell $4p$ electron excitation are the largest, but $4s$ as well as $4d$ subshell excitations cannot be neglected. The largest contribution for $4s$ is 13% of the total DR rate coefficient and for $4d$ 30% of the total at $T_e < 100$ eV. This is in contrast to the case of $3l$ subshell excitations. And furthermore, Sn^{11+} $4s^24p^54d^34f$ and $4s^24p^64d4f5l$ states have several levels just above the autoionization threshold. These states strongly enhance and even dominate the whole DR process at low temperature.

For $4p$ subshell excitons, the doubly excited states $4p^54d^24lnl'$ or $4p^54d^25lnl'$ can be formed. Figure 2 shows the DR rate coefficients for $n = 7$ and $l' = 0 - 6$. It can be seen that the DR rate coefficients for $4p^54d^24l7l'$ give a significant contribution. DR rate coefficients for $4p^54d^25p7l'$

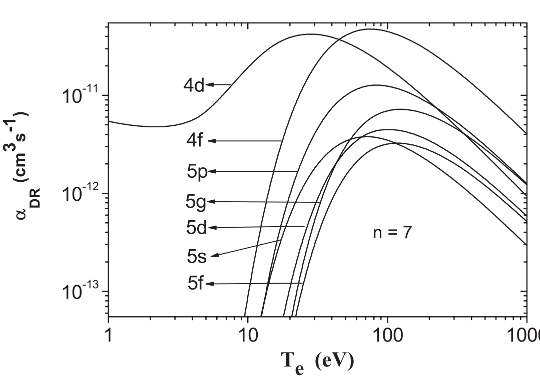

FIG. 2. DR rate coefficients for $4p$ excitation for the intermediate doubly states $4p^{-1}nl7l'$ with $nl = 4d$, $4f$, and $5s - 5g$. Each curve gives the sum of coefficients for all the permitted l' orbitals.

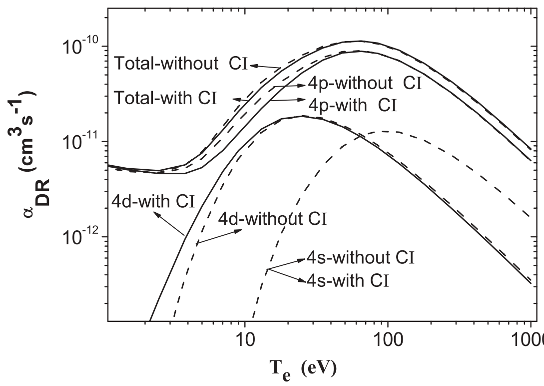

FIG. 3. Effects of configuration interaction (CI) on DR rate coefficients for intermediate doubly excited states $[4s^24p^64d^2]^{-1}nl7l'$. Solid curve: calculation with the inclusion of CI effects. Broken curve: calculation without the inclusion of CI effects. The notations $4s$, $4p$, and $4d$ on the curves denote the orbitals which are excited. The notation "Total" stands for the sums of the coefficients of $4s$, $4p$, and $4d$ excitation resonances.

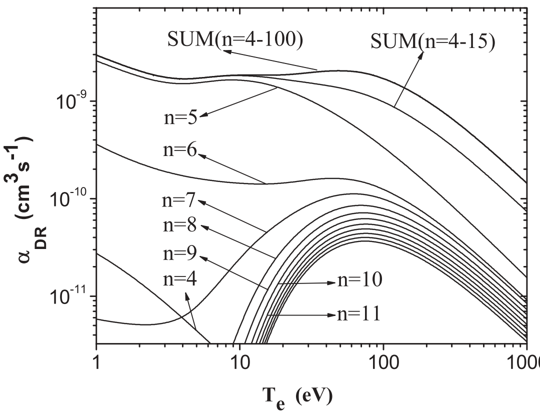

FIG. 4. The DR rate coefficients where an incident electron is captured to the orbitals with principal quantum number ranging from $n = 4-11$, which are indicated on the individual curves. The tags SUM ($n = 4-15$) and SUM ($n = 4-15$) stand for their sums over $n = 4-15$ and over $n = 4 - 100$, respectively.

are largest for the doubly excited state $4p^54d^25l7l'$ in the temperature range $T_e < 100$ eV. In our further calculations, the contributions from $4p^54d^25snl'$, $4p^54d^25dnl'$, $4p^54d^25fnl'$, and $4p^54d^25gnl'$ are neglected. For $4s$, $4d$ subshell excitons, a similar result can be found.

B. Influence of configuration interaction

In general, configuration interaction (CI) can impact the energy levels and the transition rates. O'Sullivan and Faukner [7] have pointed out that there are strong interactions between $4p^64d^{N-1}4f^1$ and $4p^54d^{N+1}$ configurations in multiply charged tin ions, and Koike *et al.* [6] subsequently verified this effect, where N stands for the electron occupation number in the corresponding ground-state ions. Here we study this CI effect on the DR rate coefficients. Figure 3 compares the results with CI and without CI for the DR for intermediate doubly excited states $[4s^24p^64d^2]^{-1}nl7l'$ (here $nl = 4d$, $4f$, and $5p$, l' include all the possible orbitals). Our results show that the DR rate coefficients for $4d$ subshell excitations with CI are larger than those without CI in the lower temperature region, but for $4p$ subshell excitation, the behavior is reversed. There is little influence on the DR rate coefficients for $4s$ subshell excitations with CI. We find that the CI effect can cause up to 12% difference in total DR rate coefficient at $T_e = 12$ eV. It is concluded that the contributions from CI for $4p$ and $4d$ subshell excitation cannot be neglected at low temperatures.

C. The total DR rate coefficients

In the present calculations, the ground configuration state Sn^{12+}[Ni] $4s^24p^64d^2$ includes nine levels, and the intermediate doubly excited states result from $4s$, $4p$, and $4d$ subshell excitations. The DR processes of Sn^{12+} ions can be expressed as

$$\begin{aligned}4s^24p^64d^2 + e^- &\rightarrow (4s^24p^64d^2)^{-1}nln'l'\\ &\rightarrow (4s^24p^64d^2)^{-1}n''l''n'l' + h\nu, \quad (7)\end{aligned}$$

here, $nl = 4d,4f,5p$, and $n' < 16$, $n'' < 5$, $l' < 10$, $l'' < 3$. After finishing separate calculations on DR rate coefficients for the nine ground configuration levels, we obtained the DR rate coefficients by averaging the nine separate rate coefficients over their statistical weights. In addition to the level-to-level rate coefficients, we obtained the configuration-averaged and total DR rate coefficients by averaging initial states and summing up over the final states.

Figure 4 shows the contributions of DR rate coefficients where a free electron is captured to different n shells. It can be seen that the DR for $n = 5$ gives an important contribution. At low temperature, the DR rate coefficients from $n = 5$ are very large and decrease with increasing electron temperature in the range $1 - 4$ eV. This means there are some DR processes with small resonance energies. As a function of the electron temperature, the DR rate coefficients depend mainly on $T_e^{-(3/2)}$ and $\exp(-E_{ji}/kT_e)$ in Eq. (1), and for the same T_e, a smaller E_{ji} gives larger DR rate if other quantities are the same in Eq. (1). Although the contributions from $n > 5$ decrease as n increases, their sum exceeds the contributions from $n = 5$, and the contributions from $n > 15$ are important and should not

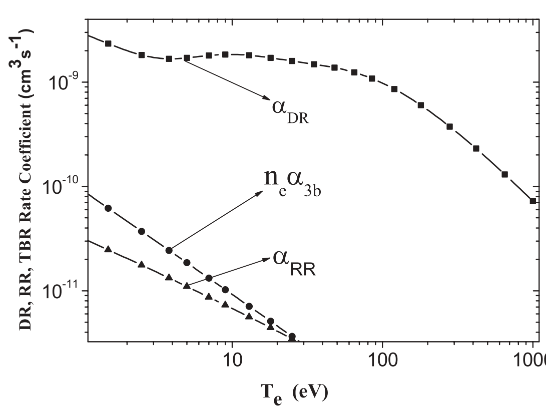

FIG. 5. DR, RR, and TBR rate coefficients of Sn^{12+} ions α_{DR}, α_{RR}, $n_e\alpha_{3b}$, where n_e is the number density of free electrons which are assumed as 10^{21} cm^{-3}. α_{DR} ($n = 4 - 100$) is the sum of DR rate coefficients from $n = 4 - 100$.

TABLE I. Fitting DR rate coefficients for Sn^{12+} ions using equation (8). Numbers in brackets denote powers of 10.

n	c_1	E_1	c_2	E_2	c_3	E_3	c_4	E_4	c_5	E_5
4 − 100	2.515[−7]	1.995[+1]	7.353[−9]	9.159[−1]	3.252[−6]	1.458[+2]	3.367[−8]	7.044[0]	1.451[−6]	7.206[+1]
4 − 15	5.682[−7]	6.920[+1]	3.184[−8]	6.876[0]	7.271[−9]	9.6[−1]	2.416[−7]	1.95[+1]	1.71[−6]	1.333[+2]
4	5.734[−11]	7.343[−1]								
5	6.423[−9]	9.241[−1]	2.566[−8]	6.683[0]	7.273[−8]	1.349[+2]	2.177[−7]	1.887[+1]	1.932[−7]	5.815[+1]
6	2.277[−7]	1.167[+2]	3.587[−9]	6.617[0]	7.252[−10]	7.131[−1]	1.471[−8]	2.104[+1]	7.081[−8]	5.855[+1]
7	6.755[−8]	7.234[+1]	1.690[−8]	3.364[+1]	2.395[−11]	1.420[0]	3.685[−10]	1.039[+1]	2.105[−7]	1.338[+2]
8	1.172[−7]	1.602[+2]	6.756[−10]	3.219[+1]	1.191[−9]	5.500[+1]	1.086[−7]	1.081[+2]	3.204[−8]	5.500[+1]
9	5.528[−8]	1.386[+2]	4.655[−9]	4.912[+1]	5.094[−8]	1.819[+2]	4.117[−8]	6.814[+1]	7.931[−8]	1.248[+2]
10	2.082[−8]	6.205[+1]	−4.232[−10]	8.610[+1]	1.390[−7]	1.520[+2]	3.365[−9]	3.196[+2]	4.496[−8]	8.866[+1]
11	4.270[−9]	1.339[+2]	4.041[−8]	7.148[+1]	1.809[−8]	2.122[+2]	1.126[−7]	1.369[+2]	9.857[−9]	2.057[+2]
12	2.943[−8]	9.617[+1]	9.367[−8]	1.521[+2]	1.474[−8]	7.783[+1]	1.389[−8]	6.723[+1]	1.460[−8]	2.066[+2]
13	4.505[−8]	1.232[+2]	3.669[−8]	7.805[+1]	1.230[−8]	1.798[+2]	1.705[−9]	5.944[+1]	5.338[−8]	1.704[+2]
14	2.108[−8]	8.775[+1]	3.373[−8]	1.235[+2]	4.746[−9]	1.317[+2]	1.852[−8]	7.281[+1]	5.673[−8]	1.744[+2]
15	2.122[−8]	1.329[+2]	2.411[−8]	7.402[+1]	4.708[−8]	1.774[+2]	1.619[−8]	1.452[+2]	2.623[−8]	9.825[+1]

be neglected at intermediate and high temperature. The values of the DR rate coefficients for $n = 6 - 15$ are at a maximum between 40 and 100 eV. In the temperature range $5 - 80$ eV, there exists a higher value for total DR rate coefficients, and thereafter, the DR rate coefficients drop quickly with increasing electron temperature.

In order to facilitate applications, one can use the approximation given in Eq. (8):

$$\alpha_{DR}(kT_e) = kT_e^{-3/2} \sum_i c_i \exp\left(-\frac{E_i}{kT_e}\right), \tag{8}$$

where c_i (cm^3 s^{-1}) and E_i are the fitting coefficients. Using the parameters listed in Table I, we can reproduce the present calculated DR rate coefficients to within 1%.

In Fig. 5 the DR, RR, and TBR rate coefficients are shown. Here the number density of the free electrons is assumed as 10^{21} cm^{-3}, which is the critical density for Nd:yttrium aluminum garnet (YAG) laser irradiation. It can be seen that the DR rate coefficients are greater than the RR rate coefficients and TBR rate coefficients for $T_e > 1$ eV. Clearly, DR will predominate the ionization balance.

IV. SUMMARY

Based on the relativistic atomic code, FAC, theoretical calculations have been made for the dielectronic recombination rate coefficients of Sn^{12+} ions. Investigation of the CI effect shows that contributions from CI for $4p$ and $4d$ subshell excitation cannot be neglected. Comparison of the rate coefficients from $4s$, $4p$, and $4d$ subshell excitations shows the $4p$ subshell excitation dominates in the $1 - 1000$ eV temperature region, but the DR rate coefficients from $4d$ and $4s$ subshells must be included. DR for $n = 5$ gives an important contribution, but the contributions from $n > 5$ still should not be neglected at intermediate and high temperature. In the temperature range $5 - 80$ eV, there exists a higher contribution for total DR rate coefficients. In addition, the total DR rate coefficient is greater than either the RR or TBR rate coefficients when the number density of free electrons 10^{21} cm^{-3} for the case of electron temperature greater than 1 eV. Therefore, the DR process can strongly influence the ionization balance of laser-produced multicharged tin ions. These results should be useful for further simulations and analyses of EUV spectra in laser-produced tin plasmas.

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[1] P. Mazzotta, G. Mazzitelli, S. Colafrancesco, and N. Vittorio, Astron. Astrophys. Suppl. Ser. **133**, 403 (1998).

[2] S. M. V. Aldrovandi and D. Pequignot, Astron. Astrophys. **25**, 137 (1973).

[3] A. Lampert, A. Wolf, D. Habs, J. Kenntner, G. Kilgus, D. Schwalm, M. S. Pindzola, and N. R. Badnell, Phys. Rev. A **53**, 1413 (1996).

[4] Y. B. Fu, C. Z. Dong, and D. H. Zhang, Chin. Phys. **15**, 2583 (2006).

[5] J. G. Wang, T. Q. Chang, and C. Z. Dong, Chin. Phys. Lett. **15**, 101 (1998).

[6] F. Koike, S. Fritzsche, K. Nishihara, A. Sasaki, T. Kagawa, T. Nishikawa, K. Fujima, T. Kawamura, and H. Furukawa, J. Plasma Fusion Res. **7**, 253 (2006).

[7] G. O'Sullivan and R. Faulkner, Opt. Eng. **33**, 3978 (1994).

[8] A. Sasaki, K. Nishihara, F. Koike, T. Kagawa, T. Nishikawa, K. Fujima, T. Kawamura, and H. Furukawa, IEEE J. Quantum Electron. **10**, 1307 (2004).

[9] R. C. Spitzer, T. J. Orzechowski, D. W. Phillion, R. L. Kauffman, and C. Cerjan, J. Appl. Phys. **79**, 2251 (1996).
[10] P. Jansson, B. Hansson, O. Hemberg, M. Otendal, and A. Holmberg, Appl. Phys. Lett. **84**, 2256 (2004).
[11] T. Aota and T. Tomie, Phys. Rev. Lett. **94**, 015004 (2005).
[12] J. White, P. Hayden, P. Dunne, A. Cummings, N. Murphy, P. Sheridan, and G. O'Sullivan, J. Appl. Phys. **98**, 113301 (2005).
[13] S. S. Churilov and A. N. Ryabtsev, Phys. Scr. **73**, 614 (2006)
[14] M. Y. Song, T. Kato, and J. Korean Phys. Soc. **49**, S374 (2006).
[15] U. I. Safronova, R. Bista, R. Bruch, and Y. Ralchenko, J. Phys. B **42**, 015001 (2009).
[16] Y. B. Fu, C. Z. Dong, M. G. Su, and G. O'Sullivan, Chin. Phys. Lett. **25**, 972 (2008).
[17] M. F. Gu, Astrophys. J. **590**, 1131 (2003).
[18] U. Safronova, T. Kato, and M. Ohira, J. Quant. Spectrosc. Radiat. Transf. **58**, 193 (1997).
[19] D. Colombant and G. F. Tonon, J. Appl. Phys. **44**, 3524 (1973).
[20] H. Zhang, Y. M. Li, J. Yan, and J. G. Wang, Phys. Rev. A **71**, 042705 (2005).