# Observation of dielectronic recombination through two-electron–one-photon correlative stabilization in an electron-beam ion trap

Y. Zou*
*Applied Ion Beam Physics Laboratory, Fudan University, Shanghai 200433, People's Republic of China*

J. R. Crespo López-Urrutia and J. Ullrich
*Max-Planck-Institut für Kernphysik, Heidelberg, Germany*
(Received 3 October 2002; published 7 April 2003)

Dielectronic recombination (DR) for He-like $Ar^{16+}$ through both one-electron–one-photon and two-electron–one-photon (TEOP) stabilizations of Li-like states was studied with an electron-beam ion trap (EBIT). It turned out that this is an excellent method to investigate TEOP transitions. Its advantages are a high branching ratio for the TEOP transition and clean conditions under which spectator electrons are controlled. Further, state- and configuration-resolved KLL DR cross sections were obtained due to the unsurpassed electron energy resolution achieved in the EBIT in the energy range around 2 keV.

DOI: 10.1103/PhysRevA.67.042703 PACS number(s): 34.80.Lx

Simultaneous multielectron transitions resulting in single-photon emission were predicted by Heisenberg [1] in 1925. The first experimental observation of two-electron–one-photon transitions (TEOP) took place 50 years later in heavy-ion collision experiments by Wölfli *et al.* [2]. Since then, they have been studied in various experiments [3–11], where mostly double inner-shell $K$ vacancies were produced. In the subsequent decay, the branching ratio of the TEOP transition $K_{\alpha\alpha}$, in which two $L$ shell electrons jump into the $K$ shell and release one photon, to the single-electron hypersatellite transition $K_\alpha^h$ was measured, as well as the energies of the emitted photons.

As was already pointed out by Condon [12], ''multielectron jumps occur because of the fact that spectral terms may not be precisely labeled by means of electron configurations.'' This means that multielectron atomic states cannot be perfectly described by using single-electron configurations but, instead, a summation over a group of configurations is needed. Thus, predictions of TEOP transitions are very sensitive to the multielectron wave functions used and, therefore, experimental studies of this process supply stringent tests of the different theoretical models. In addition, these transitions can provide sensitive diagnostic means for hot plasmas [13].

However, $K_{\alpha\alpha}$ is a much slower transition compared to $K_\alpha^h$, hence its branching ratio is extremely small, e.g., $10^{-4}$ for $Z=26$, which makes measurements very difficult. Moreover, the uncertain number of accompanying outer-shell vacancies created with the double $K$ holes in most of the abovementioned experiments disturbs the comparison between experimental results and theoretical predictions.

In this paper, we show an excellent method to investigate TEOP transitions with a very high branching ratio, which simplifies significantly the measurement. Instead of observing the $K_{\alpha\alpha}$ TEOP transitions, we measure the TEOP transitions between the $1s^22p$ and $1s2s^2$ states. As there are no other competing strong radiative processes, the branching ratio of the TEOP transition is extremely high. At the same time, these experiments ensure unprecedented clean conditions where spectator electrons in the ions can be completely controlled. This is because the formation of the initial state of the TEOP transitions proceeds via DR resonances which are well separated for different charge states due to the high electron energy resolution achieved in the present measurement. Specifically in this work, we studied the correlative stabilization transition of $1s^22p\ ^2P_{1/2,3/2}$-$1s2s^2\ ^2S_{1/2}$ following the KLL dielectronic recombination (DR) resonance of He-like Ar ions in the EBIT.

The DR process is important in hot plasmas and is decisive for the charge state balance in the plasma. Studies of DR processes in He-like ions have been performed for several elements [14–22] by using EBITs, EBISs, or heavy-ion storage rings. So far, state- or configuration-resolved high electron energy resolution DR experiments have only been performed at heavy-ion storage rings [22–25], which have been the primary tools at low center of mass interaction energy, i.e., below a few hundred eV. Due to the low interaction energy, storage rings are mostly used to investigate $\Delta n=0$ DR processes [24,25] and $\Delta n>0$ DR in light ions [22,23]. For heavier ions, where $\Delta n>0$, DR needs higher collision energies and at present EBITs or EBISs are more suitable tools, although the electron-beam energy resolving power has been relatively low in all reported measurements to date. As a compensation, EBIT studies have an important advantage over current storage-ring experiments since emitted photons are observed. This opens up possibilities to study stabilization pathways in detail. In the work of Beiersdorfer *et al.* [18], for example, a high-resolving power of the photon energy was used for DR studies, thereby compensating for the lower resolution of the electron-beam energy. In the work of Ali *et al.* [14,15], DR cross sections, as well as differential cross sections of He-like Ar ions were studied by using an EBIS. In their work, the DR cross sections were obtained by studying the charge state fractions of extracted ions from the EBIT, and the differential cross sections were from fast electron energy scanning x-ray measurements performed at 0°.

*Electronic address: zouym@fudan.edu.cn

 

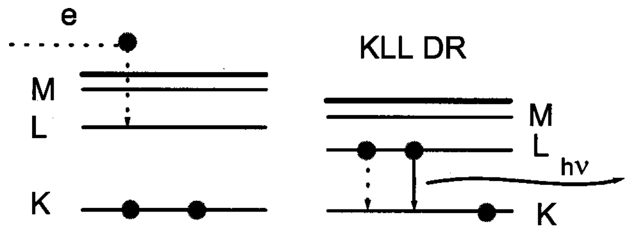


FIG. 1. Illustration of KLL dielectronic recombination for a He-like ion. Both electrons in the doubly excited intermediate state have the possibility to relax into the $1s$ shell.

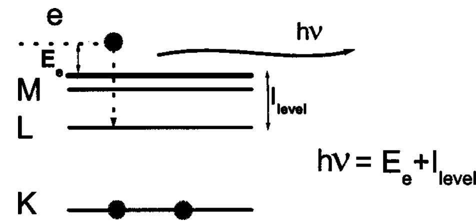


FIG. 2. Illustration of $n=2$ radiative recombination for a He-like ion.

In this paper, we report resolved DR cross sections of He-like Ar to the Li-like specific state $1s2s^2\,{}^2S_{1/2}$ and specific configurations of $1s2s2p$ and $1s2p^2$ at a similar resolving power as those reported at storage rings, but using an EBIT at much higher interaction energy, i.e., above 2 keV. The experimental method was basically same as that of Ali *et al.* [14,15] in their x-ray measurements. The differences are following: In this work, the x rays were detected at 90° with solid angle ($8.7\times10^{-4}4\pi$ sr) of three orders of magnitude higher than theirs, the electron-beam energy resolution is five times higher as compared to their work, and finally, the measurements were performed under a steady-state EBIT running mode instead of fast scanning mode employed in their x-ray measurements.

The DR process is resonant, in which a free electron is captured by an ion and, at the same time, another bound electron is excited, thereby forming an intermediate doubly excited state. Up to this point, this is just the time-reversed Auger process. If this intermediate state is stabilized by the emission of one or more photons, then the DR process is complete:

$$e^- + \mathrm{A}^{q+} \rightarrow (\mathrm{A}^{(q-1)+})^{**} \rightarrow \mathrm{A}^{(q-1)+} + h\nu$$

$$\downarrow$$

$$(\mathrm{A}^{(q-1)+})^{*} + h\nu'.$$

On the other hand, the intermediate state usually has a large probability to decay through Auger processes, in which case the DR was not "successful" because no recombination finally took place; instead only resonant scattering of the electron occurred. In this work, we are interested in KLL DR processes of He-like Ar ions as shown in Fig. 1. A free electron with energy $E_e$ is recombined with a ground-state He-like ion, forming intermediate Li-like $1s2l2l'$ states, specifically the $1s2s^2$, $1s2s2p$, or $1s2p^2$. These intermediate states are then stabilized through $K_\alpha$ transitions: $1s^22p$-$1s2s^2$, $1s^22s$-$1s2s2p$, and $1s^22p$-$1s2p^2$. Among them, $1s^22p$-$1s2s^2$ is the TEOP transition of interest, which actually contains two transitions to the different fine-structure states ${}^2P_{1/2,3/2}$.

In DR studies, an important accompanying process is a radiative recombination (RR) that can occur at any given electron energy:

$$e^- + \mathrm{A}^{q+} \rightarrow \mathrm{A}^{(q-1)+} + h\nu.$$

In this process, a free electron is recombined with an ion releasing a photon in order to maintain energy conservation (see Fig. 2). RR is not a resonant process with respect to the electron energy and, hence, it is associated with a continuous spectral feature: the photon energy is equal to the sum of the electron kinetic energy and the ionization energy of the final electronic state.

In this work, we measured x-ray intensities as a function of the x-ray energy and the electron energy, as shown in the scatter plot in Fig. 3. The TEOP transition energy and the resonant electron-beam energy for the DR process to the specific intermediate state $1s2s^2\,{}^2S_{1/2}$ were determined simultaneously. Using a proper normalization, the cross sections for this process and for the other KLL DR of He-like Ar ions were obtained, and the correlative transition rate was extracted.

The experiment was carried out at the electron-beam ion trap FreEBIT of the University of Freiburg [26] (now at the MPI-K in Heidelberg). The x-ray photons were measured using a high-purity germanium detector with a resolution of about 150 eV. The electron-beam energy was scanned from 1.5 to 4.0 keV. By using very low beam currents (2.7–5.7 mA), the energy spread of the electron beam was reduced to 8-eV FWHM (full width at half maximum), which gives a

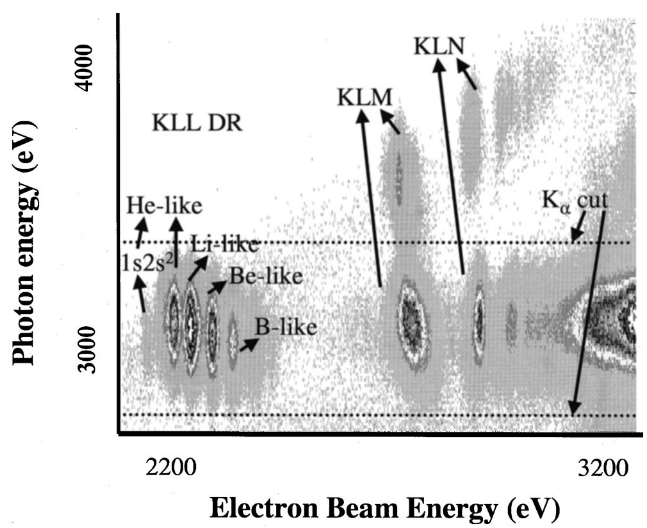


FIG. 3. Scatter plot of the x-ray intensity vs the electron-beam energy ($X$ axis) and x-ray energy ($Y$ axis). The first group of spots is the KLL DR resonances to the He-like, Li-like, Be-like, and B-like Ar ions. The KLM, KLN, and other DR events can be seen in the figure. Taking the "$K_\alpha$ cut" produces the spectrum shown in Fig. 4.

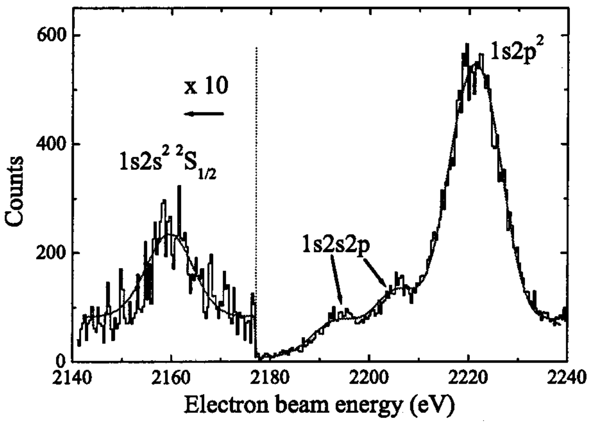


FIG. 4. A portion of the "$K_\alpha$ cut" projection (horizontal) from the Fig. 3 scatter plot is shown, highlighting the He-like part of the KLL DR events' x-ray intensity as a function of the electron-beam energy.

resolving power of $E/(\mathrm{FWHM}) = 270$, corresponding to a resolution of 0.3%. This allows us to resolve the He-like Ar DR resonance to the $1s2s^2\,^2S_{1/2}$ state from those to the $1s2s2p$ and $1s2p^2$ configurations, as illustrated in Fig. 4, showing a part of the $K_\alpha$ x-ray cut (the horizontal cut) from the scatter plot (see Fig. 3). Due to the low electron-beam currents used here, charge exchange between the highly charged ions and residual gas atoms was very competitive, making the lower charge states, i.e., Li-like, Be-like, etc., rather pronounced. The measurement was repeated 11 times, at electron-beam currents in the range between 2.7 and 5.7 mA, to ensure that the statistical uncertainty of the intensity of the weakest peak was less than 2% after averaging over all the datasets. Figure 4 shows one individual measurement. The absolute resonance energy was determined after applying a correction for the space charge of the electron beam, which was found to be linearly dependent on the electron-beam current, as expected. By varying the electron-beam current from 2.7 to 50 mA and extrapolating to zero current, the resonance energy can be determined with an estimated error of 1.1 eV.

The EBIT was run in a "steady-state mode" for this experiment, which means that the electron-beam energy was varied so slowly that the ion charge distribution at each energy is essentially identical to that of the static case. Hence, the rate equations for He-like, Li-like, and Be-like Ar ions for the electron-beam energy at the He-like KLL DR resonance are as follows:

$$\frac{dn_{16}}{dt} = (n_{15}\sigma_{I15} - n_{16}\sigma_{DR} - n_{16}\sigma_{RR16})j_e/e - n_{16}n_0\sigma_{C16}v,$$

$$\frac{dn_{15}}{dt} = (n_{14}\sigma_{I14} - n_{15}\sigma_{RR15} - n_{15}\sigma_{I15} + n_{16}\sigma_{DR} + n_{16}\sigma_{RR})j_e/e - n_{15}n_0\sigma_{C15}v + n_{16}n_0\sigma^1_{C16}v,$$

$$\frac{dn_{14}}{dt} = (n_{13}\sigma_{I13} + n_{15}\sigma_{RR15} - n_{14}\sigma_{RR14} - n_{14}\sigma_{I14})j_e/e - n_{14}n_0\sigma_{C14}v + n_{15}n_0\sigma^1_{C15}v + n_{16}n_0\sigma^2_{C16}v.$$

Here, $n_q$ is the density of $\mathrm{Ar}^{q+}$, $\sigma_{Iq}$ is the electron-impact ionization cross section for $\mathrm{Ar}^{q+}$, $\sigma^r_{Cq}$ is the cross section for the capture of $r$ electrons by $\mathrm{Ar}^{q+}$ (with an average velocity of $v$) from the residual gas of density $n_0$, $\sigma_{Cq}$ is the capture cross section for any number of electrons by $\mathrm{Ar}^{q+}$, $\sigma_{RRq}$ is the RR cross section of $\mathrm{Ar}^{q+}$, $\sigma_{DR}$ is the DR cross section for He-like Ar ions, and $j_e/e$ is the electron-beam current density normalized to $e$, the electronic charge. For highly charged ions, the multielectron capture cross section is not negligible compared to single-electron capture. In this work, we included double-electron capture in the rate equations. For steady state, we obtain

$$\frac{dn_{15}}{dn_{16}} = \sigma_{DR}/\sigma_{I15} + \sigma_{RR}/\sigma_{I15} + en_0v\,\sigma_{C16}/(j_e\sigma_{I15}) = \sigma_{DR}/\sigma_{I15} + (n_{15}/n_{16})_0,$$

where $(n_{15}/n_{16})_0$ is the off-resonance density ratio. By assuming that $n_{15}+n_{16}=n$ stays constant during the He-like DR resonance, the DR event count rate will be

$$R_e(E_e) = N(\epsilon, j_e/e, n)W(\theta)\sigma_{DR}(E_e)/(\sigma_{DR}(E_e)\sigma_{I15} + (n_{15}/n_{16})_0 + 1).$$

$N(\epsilon, e, j_e/e, n)$ is a coefficient including the detection efficiency, the electron current density, and the total number density of He-like and Li-like Ar ions; $W(\theta)$, is the angular variation of the photon emission caused by polarization of the DR stabilizing radiation. We assume a Gaussian shape for $\sigma_{DR}(E_e)$, as $\sigma_{DR}$ has to be folded with the electron-beam energy $E_e$ resolution. The assumption that $n_{15}+n_{16}=n$ remains constant introduces less than 10% uncertainty in the cross section obtained at the strongest resonance, and even smaller values at weaker resonances. The other main error source arises from the statistical uncertainty which contributes less than 2%, even for the weakest resonance. The results are listed in Table I. The ionization cross section, $\sigma_{I15}$, of Li-like Ar was taken from Younger's work [27] and the other cross sections used for error estimates are from Selberg *et al.* [28] and Younger [29].

The cross sections obtained for the DR processes starting from ground-state He-like Ar ions to produce Li-like Ar $1s2s^2$, $1s2s2p$, and $1s2p^2$ configurations are listed in Table I, along with the resonant electron energy of the DR from $1s^2\,^1S_0$ to $1s2s^2\,^2S_{1/2}$. Since the resonance energies of the DR to $1s2p^2\,^4P$ overlap with those of $1s2s2p$, the cross section for $1s2s2p$ in this work is mixed with a weak component from the contribution of $1s2p^2\,^4P$. This introduces an uncertainty of roughly 3%, which was estimated using the transition rates from Bhalla and Tunnell's work [30]. The rate of the two-electron–one-photon transition

TABLE I. This table presents the cross sections $\sigma$ for the DR processes from ground-state He-like Ar ions to Li-like $1s2s^2$, $1s2s2p$, and $1s2p^2$ configurations and their summation $\Sigma\sigma$ (KLL), the resonant electron energy $Ee$ for the DR from $1s^2\,{}^1S_0$ to $1s2s^2\,{}^2S_{1/2}$, and the rate $A$ of the two-electron–one-photon transitions, $1s^22p\,{}^2P_{1/2,3/2}$–$1s2s^2\,{}^2S_{1/2}$, together with their photon energies $E_x$. The theoretical results were obtained by using the Auger-electron energies, branching ratios, x-ray fluorescence yields, and lifetimes from the work of Bhalla and Tunnell [30].

| | Expt. This work | Others | Theor. |
|---|---|---|---|
| $\sigma(1s2s^2\,{}^2S_{1/2})$ ($10^{-20}$ cm$^2$ eV) | $1.05\pm0.1$ | | 0.974 |
| | 0.992 [a] | | |
| $\sigma(1s2s2p)$ ($10^{-20}$ cm$^2$ eV) | $12.8\pm1.3$ | | 13.1 |
| $\sigma(1s2p^2)$ ($10^{-20}$ cm$^2$ eV) | $49.5\pm6.0$ | | 50.1 |
| $\Sigma\sigma$ (KLL) ($10^{-20}$ cm$^2$ eV) | $63.4\pm7.5$ | 59.4 [b] | 64.2 |
| $\sigma(1s2s^2\,{}^2S_{1/2})/\Sigma\sigma$ (KLL) | $0.017\pm0.002$ | | 0.0152 |
| $E_e(1s2s^2\,{}^2S_{1/2})$ (eV) | $2159.7\pm1.1$ | 2160.6 [c] | 2162.5 |
| $A$ ($10^{12}$ s$^{-1}$) | $4.76\pm0.57$ | | 4.42 |
| | 4.49 [a] | | |
| $E_x({}^2P_{1/2}\text{-}{}^2S_{1/2})$ (eV) | $3044\pm6$ | 3047.1 [c] | 3049.6 |
| $E_x({}^2P_{3/2}\text{-}{}^2S_{1/2})$ (eV) | $3041\pm6$ | 3044.0 [c] | 3046.4 |

[a]Obtained using the $\sigma(1s2s^2\,{}^1S_0)/\Sigma\sigma$ (KLL) from this experiment and normalized to the total cross section from Ali *et al.* [15].
[b]Experimental result with error of 9%, from Ali *et al.* [15].
[c]Obtained using the level energies from the NIST database.

$1s^22p\,{}^2P_{1/2,3/2}$–$1s2s^2\,{}^2S_{1/2}$ was extracted from the DR cross section of the $1s^2\,{}^1S_0$ to $1s2s^2\,{}^2S_{1/2}$ state according to the following relation:

$$A(1s^22p\,{}^2P_{1/2,3/2}-1s2s^2\,{}^2S_{1/2}) = \sigma_{DR}(1s2s^2\,{}^2S_{1/2})2mE_eg_i/(g_s\pi^2\hbar^3R_a),$$

where $m$ is the electron mass, $R_a$ is the branching ratio for Auger decay to the total decay [30], and $g_i$ and $g_s$ denote the statistical weight of the initial and intermediate states, respectively. In this work, the x-ray detector resolution is not high enough to resolve the transitions of ${}^2P_{1/2}$-${}^2S_{1/2}$ and ${}^2P_{3/2}$-${}^2S_{1/2}$. Therefore, the corresponding rate reflects the sum of both transitions and is also listed in Table I, together with the x-ray energies for the transitions with uncertainties of 0.2%.

As can be seen in Table I, the total KLL DR cross section obtained in this work $(63.4\pm7.5)\times10^{-20}$ cm$^2$ eV, agrees with the EBIS result of $59.4\times10^{-20}$ cm$^2$ eV, of Ali *et al.* [15]. The DR cross sections for the $1s2s^2\,{}^2S_{1/2}$ state and the $1s2s2p$ and $1s2p^2$ configurations are the first obtained experimental results—no other experimental values are available for comparison. All the cross sections obtained here agree well with the theoretical predictions of Bhalla and Tunnel [30].

In the present measurements, the initial state of the TEOP transition is formed by a well-defined DR resonance from a well-resolved charge state, so that no spectator electrons are involved in the TEOP transitions. Under such clean conditions, we can confidently compare our TEOP results with the available theoretical calculations for Li-like Ar ions, and we find that they coincide with the Hartree-Fock prediction [30] within the experimental uncertainty. All of the TEOP relevant energies, i.e., the electron resonant energy of the DR process to $1s2s^2\,{}^2S_{1/2}$, and the TEOP transition x-ray energies are compared also to those from the NIST database, and are found to be in good agreement.

In summary, we observed DR through both one-electron–one-photon and two-electron–one-photon stabilizations in Li-like Ar ions and obtained experimental DR cross sections from He-like Ar $1s^2\,{}^1S_0$ to the Li-like specific state $1s2s^2\,{}^2S_{1/2}$ and specific configurations $1s2s2p$ and $1s2p^2$. Both were made possible by the excellent electron energy resolution achieved at energies beyond the reach of current storage-ring experiments. This method to investigate two-electron–one-photon processes has the advantage of a high branching ratio for this type of transition and a well-defined initial electronic configuration.

We gratefully acknowledge support from the Leibniz-Programm as well as from the separate funding contract (Contract No. ULL 166/2-1) of the Deutsche Forschungsgemeinschaft, from the Max-Planck-Institut für Kernphysik in Heidelberg, from the Deutsche Hochschulbauförderung, and from the University of Freiburg. One of the authors (Y.Z.) is grateful to the group of J. Ullrich of the Max-Planck-Institut für Kernphysik in Heidelberg for their hospitality, and was partly supported by the outstanding young scientist program from the Natural Science Foundation of China (Grant No. 0125520) and by the outstanding young professor project of the Chinese Educational Ministry.

[1] W. Heisenberg, Z. Phys. **32**, 841 (1925).
[2] W. Wölfli *et al.*, Phys. Rev. Lett. **35**, 656 (1975).
[3] C. Stoller *et al.*, Phys. Rev. A **15**, 990 (1977).
[4] T.P. Hoogkamer, P. Woerlee, F.W. Saris, and M. Gavrila, J. Phys. B **9**, L145 (1976).
[5] J. Volpp, R. Schuch, G. Nolte, and H. Schmidt-Böcking, J. Phys. B **12**, L325 (1979).
[6] R. Schuch, G. Gaukler, and H. Schmidt-Böcking, Z. Phys. A **290**, 19 (1979).
[7] A.R. Knudson, K.W. Hill, P.G. Burkhallter, and D.J. Nagel, Phys. Rev. Lett. **37**, 679 (1976).
[8] Y. Isozumi, Phys. Rev. A **22**, 1948 (1980).
[9] M.S.A.L. Al-Ghazi, J. Birchall, and J.S.C. Mckee, Phys. Rev. A **25**, 3072 (1982).
[10] S.I. Salmem, A. Kumar, and B.L. Scott, Phys. Rev. A **29**, 2634 (1984).
[11] Auerhammer, H. Genz, A. Humar, and A. Richter, Phys. Rev. A **38**, 688 (1988).
[12] E.U. Condon, Phys. Rev. **36**, 1121 (1930).
[13] F.B. Rosmej *et al.*, Phys. Rev. A **63**, 032716 (2001).

[14] R. Ali, C.P. Bhalla, C.L. Cocke, and M. Stockli, Phys. Rev. Lett. **64**, 633 (1990).
[15] R. Ali *et al.*, Phys. Rev. A **44**, 223 (1991).
[16] A.J. Smith *et al.*, Phys. Rev. A **54**, 462 (1996).
[17] A.J. Smith *et al.*, Phys. Rev. A **62**, 052717 (2000).
[18] P. Beiersdorfer *et al.*, Phys. Rev. A **46**, 3812 (1992).
[19] D.A. Knapp *et al.*, Phys. Rev. A **47**, 2039 (1993).
[20] T. Fuchs *et al.*, Phys. Rev. A **58**, 4518 (1998).
[21] H. Watanabe *et al.*, J. Phys. B **34**, 5095 (2001).
[22] S. Mannervik *et al.*, Phys. Rev. A **55**, 1810 (1997).
[23] G. Kilgus *et al.*, Phys. Rev. Lett. **64**, 737 (1990).
[24] W. Spies *et al.*, Nucl. Instrum. Methods Phys. Res. B **98**, 158 (1992).
[25] J. Linkemann *et al.*, Nucl. Instrum. Methods Phys. Res. B **98**, 154 (1992).
[26] J.R. Crespo *et al.*, Phys. Scr., T **T80**, 502 (1999).
[27] S.M. Younger, J. Quant. Spectrosc. Radiat. Transf. **26**, 329 (1981).
[28] N. Selberg, C. Biedermann, and H. Cederquist, Phys. Rev. A **54**, 4127 (1996).
[29] S.M. Younger, Phys. Rev. A **24**, 1278 (1981).
[30] C.P. Bhalla and T.W. Tunnell, J. Quant. Spectrosc. Radiat. Transf. **32**, 141 (1984).