# Dielectronic recombination from the ground state of heliumlike carbon ions

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The cross section for dielectronic recombination of  $C^{4+}$  ions in the  $1s^2$  ground state has been measured with an energy resolution of  $\approx 2$  eV at the heavy-ion storage ring TSR (at Heidelberg) up to the  $1s2s(^{1,3}S)$  and  $1s2p(^{1,3}P)$  excitation thresholds just below 310 eV. Resonance energies and absolute cross sections are found to be well reproduced by theoretical calculations except in a region extending  $\approx 5$  eV below the almost degenerate excitation thresholds  $1s2s(^{1}S)$  and  $1s2p(^{3}P)$ . These discrepancies imply that theory overestimates the cross section, energy-integrated over all dielectronic resonances, by  $\approx 10\%$ .

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#### I. INTRODUCTION

Measurements of the cross section for dielectronic recombination (DR) [1, 2] between electrons and multiply charged ions have reached high accuracy [3] suitable for a detailed comparison with theoretical predictions. In particular, experiments with merged electron and ion beams employing fast (multi-MeV) ions in a single-pass arrangement [4] or in an ion storage ring [5] were shown to yield absolute cross sections with small systematic errors at a good energy resolution. Few-electron systems continue to be a testing ground [6] for theoretical approaches to DR, such as extensive calculations of the properties of doubly excited atomic states [7–9], including also relativistic and QED effects [10], or detailed treatments of the recombination process itself, for example, in the case of overlapping and interacting dielectronic resonances [11].

Dielectronic recombination of heliumlike ions results from the resonant capture of a free electron into a lithiumlike doubly excited state, dominantly  $1s2s(^{1,3}S)nl'$  or  $1s2p(^{1,3}P)nl'$ , where n and l (l' for the "outer" electron) denote the principal and the angular momentum quantum number, respectively. The recombination is completed by the spontaneous radiative decay of the doubly excited state to a singly excited three-electron state. Since by far the highest radiative decay rate is associated with the  $1s2p(^{1}P)-1s^{2}$  transition of the core, the strongest DR resonances are those of the series  $1s2p(^{1}P)nl'$ . Studies of DR employing merged beams of electrons and ions in heliumlike configurations were limited so far to ions in the metastable excited  $1s2s(^{1,3}S)$ states [12, 13] and gave access only to Rydberg resonances. In particular, for metastable  $C^{4+}$  ions [12] the observed DR resonances at center-of-mass (c.m.) energies

between 0 and 9 eV are due to the states  $1s2p(^{1}P)nl'$   $(n \ge 5)$ ,  $1s2s(^{1}S)nl'$   $(n \ge 7)$ , and  $1s2p(^{3}P)nl'$   $(n \ge 7)$ .

In the present paper, results are presented for the DR cross section of  $C^{4+}$  ions in the  $1s^2$  ground state, measured with a stored ion beam and electrons at c.m. energies between 230 and 310 eV. This energy range includes the intrashell resonances 1s2l2l' as well as all Rydberg resonances converging to the excitation thresholds  $1s2s(^{1,3}S)$  and  $1s2p(^{1,3}P)$ . At a c.m. energy spread of  $\approx 2\,\mathrm{eV}$ , energies and cross sections can be determined for individual terms of the 1s2l2l' configuration. The observation of the Rydberg resonances using ground-state ions is of particular interest in view of the discrepancies with theory observed in the earlier experiments [13] with metastable heliumlike ions.

For heavier elements, DR of heliumlike ions has been investigated also in an electron-beam ion trap, detecting emitted x rays [14] (Ni<sup>26+</sup>, Mo<sup>40+</sup>), and in an electron-beam ion source, observing x rays and charge-state abundances [15, 16] (Ar<sup>16+</sup>, Ne<sup>8+</sup>). The variation of the electron-beam energy yielded the DR cross section with a typical energy resolution of 40 eV. In comparison to these measurements, the advantages of the present experiment with merged beams are a considerably better energy resolution, a well-defined initial charge state of the ions, and the possibility to investigate also ions of lower nuclear charge Z, where the relative influence of the electron-electron interaction is stronger.

Theoretical DR cross sections for the ground and excited states of C<sup>4+</sup> were presented by Badnell, Pindzola, and Griffin [9]. We have carried out further calculations which improve on those results a little. Instead of using a Thomas-Fermi model potential to generate our radial functions, we used Hartree exchange potentials [17],

which in turn were generated using Slater-type orbitals [18]. This resulted in a 10-15% increase in the ground-state cross section, depending on the intermediate state. Configuration mixing within the three-electron complex was included by Badnell, Pindzola, and Griffin [9] for the 1s2l2l' resonances only. Taking into account such mixing also for the higher doubly excited states significantly increased the integrated cross section of the 1s2l3l' resonances by allowing the configurations 1s2s3s and 1s2s3d to stabilize through mixing with 1s2p3p. The effect of such mixing becomes negligible for  $n \geq 4$ .

## II. EXPERIMENT

The measurements have been performed in the heavyion Test Storage Ring (TSR) [19] at the Max-Planck-Institut für Kernphysik in Heidelberg. The coasting  $C^{4+}$ ion beam of typically  $60 \,\mu\text{A}$  (1.3 ×  $10^8$  stored ions) at an energy of 71.7 MeV and an electron beam providing an electron density up to  $6 \times 10^7$  cm<sup>-3</sup> were overlapping on a length of 1.5 m (2.7% of the storage-ring circumference) in the electron-cooling device of the TSR. After multiturn injection from a tandem accelerator, the ions underwent electron cooling for a few seconds at an electron velocity equal to the mean initial ion velocity (electron energy 3.28 keV). Then, leaving the mean ion velocity essentially unchanged, the electron acceleration voltage was stepped to beam energies of 5.5-6.2 keV, corresponding to the c.m. energy range of the dielectronic resonances. At a mean storage lifetime of 11s, determined by the ionization of C<sup>4+</sup> ions in residual-gas collisions, a period of  $\approx 12\,\mathrm{s}$  was used for DR measurements after each injection. During the measurement, the ion-beam size was  $\approx 3 \,\mathrm{mm}$  and the full width at half maximum (FWHM) of the ion velocity distribution was  $4 \times 10^{-4}$  of the beam velocity.

The experimental setup, the measuring technique, and the data analysis were described in detail elsewhere [20]. Recombined ions were separated from the circulating beam by the first downstream dipole magnet and detected on a multichannel plate with an efficiency of  $(95 \pm 5)\%$ . The count rate was recorded as a function of the electron energy using an energy-modulation technique which allowed us to determine the background due to electron capture of the ions in the residual gas and to eliminate background fluctuations. The typical signal count rate at the strongest DR resonance was  $1.5 \times 10^4 \,\mathrm{s}^{-1}$  with a background of about the same magnitude. From the background-subtracted count rate, we determined the rate coefficient by normalizing to the number of ions in the interaction region and the electron density. The average c.m. energy was derived [20] from the frequency spectrum of the ion-beam Schottky noise, which yielded the ion velocity, and the electron acceleration voltage, which yielded the electron velocity after correcting for the space charge of the electron beam. Finally, to obtain the experimental cross section, the rate coefficient was simply divided by the average c.m. velocity since the relative energy spread of the electrons was small (FWHM  $< 10^{-2}$  of the c.m. energy). The estimated systematic error is  $\pm 1\,\mathrm{eV}$  for the absolute c.m.

energy and 20% for the absolute cross-section scale. The errors in comparing the energies and cross sections for different resonances observed in this experiment are a factor of 4–5 smaller ( $\pm 0.2 \,\mathrm{eV}$  and  $\lesssim 5\%$ , respectively).

Having passed the stripper foils of the tandem accelerator, a substantial fraction of the ions entered the ring in the metastable excited states  $1s2s(^{1,3}S)$  with mean lifetimes of 8 µs and 20 ms, respectively. Since the measurement started a few seconds after injection, the initial population of metastable states certainly had decayed. The repopulation rate of the metastable levels by collisions of  $C^{4+}$  ions with residual-gas molecules should be of the order of the loss rate of stored ions, determined by ionization processes in the same collisions. As the storage time was much longer than the given lifetimes of the metastable states, the average population of metastable states in the beam is expected to be negligible. In a scan of the c.m. energy range of 0-4 eV, where the strong DR resonances of metastable C<sup>4+</sup> ions are expected [12], only the smooth decrease of the nonresonant radiative recombination rate without additional peaks due to DR resonances was observed, in agreement with our estimates.

## III. RESULTS AND DISCUSSION

An overview of the measured spectrum is given in Fig. 1, where also the excitation thresholds of the  $C^{4+}$  ion and the Rydberg series of lithiumlike doubly excited states converging to these thresholds are indicated. The dominant resonances coincide with the levels of the channel  $1s2p(^1P)nl'$ , as one can see in particular for n=3,4, and 5. For n=3, also resonances appear that can be clearly attributed to the other channels; however, their

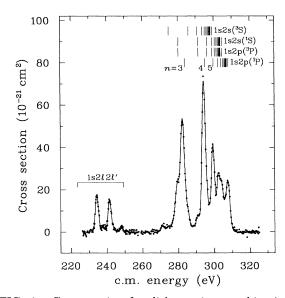


FIG. 1. Cross section for dielectronic recombination of ground-state  $C^{4+}$  ions due to resonances below the 1s2l excitation thresholds. The full line shows a Gaussian fit to the measured data. The approximate resonance positions indicated at the top were obtained from the excitation energies [21] of the  $C^{4+}$  core subtracting the binding energy of the outer electron estimated by  $(4/n)^2$  Ry.

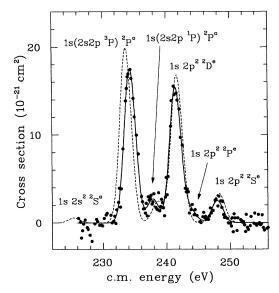


FIG. 2. Measured DR cross section due to the 1s2l2l' resonances with fitted Gaussian functions (full line) and the theoretical cross section convoluted with the experimental energy distribution (see text).

relative contributions seem to decrease rapidly with increasing n, as can be seen by comparing the resonances for n=3 and 4. The intrashell resonances 1s2l2l' yield a relatively small contribution to the total DR cross section and their structure closely resembles that of two-electron doubly excited states [5] with the 1s electron as a spectator. For the Rydberg resonances  $1s2p(^1P)nl'$  with  $n\geq 5$ , the cross section is reduced considerably by autoionization into the other channels with lower excitation thresholds.

The region of the 1s2l2l' resonances is shown in detail in Fig. 2. The peaks have been fitted by a sum of Gaussian functions, taking into account five of the six doublet terms. The  $1s2s^2\,^2S^e$  resonance, for which only a small integrated cross section is predicted, was not observed within the energy range scanned in our measurement. The fit yields a value of  $2.1\,\mathrm{eV}$  (FWHM) for the common width of all lines. The observed linewidth is much

larger than the natural width of the resonances and their fine-structure splitting and thus represents the electron energy spread in the c.m. frame. The corresponding longitudinal thermal energy of the electrons in the comoving frame of the electron beam amounts to  $1.7 \times 10^{-3} \, \mathrm{eV}$ . Also shown in Fig. 2 is the theoretical DR cross section convoluted with the c.m. electron energy distribution [20] for the given thermal energy, which is in good agreement with the measurement.

A detailed comparison between the experimental and the theoretical resonance positions and integrated cross sections is presented in Table I. To emphasize the relation between the three-electron doubly excited states and the corresponding states for a two-electron system, the coupling between the n=2 electrons is included in the term designations. The coupling of the 1s electron with the two-electron triplet states gives rise also to quartet terms for which, however, both the formation by electron collisions with ions in the  ${}^{1}S_{0}$  ground state and the stabilization via radiative transitions to  $1s^2nl$  states are strongly forbidden so that they can be neglected for the DR process. The presence of the 1s spectator electron manifests itself most clearly in the energies of the states 2s2p  $^{1}P$  and  $2p^{2}$   $^{3}P$ , which exchange their positions relative to the  $2p^2$  D state in comparison to the pure twoelectron states observed in DR of hydrogenlike ions [5]. The theoretical integrated cross sections listed in Table I agree with the experimental results within the estimated systematic error. For the integrated cross sections of the 1s2l2l' resonances the present theory essentially agrees with recent theoretical results [22] from Z expansions including relativistic effects and configuration interaction (see also Ref. [23]), and from single-configuration nonrelativistic calculations [8]. As also observed for the lowlying DR resonances of hydrogenlike ions [5], the measured and the calculated energy differences between the various resonances show slight discrepancies. These deviations have negligible influence on the DR rate coefficient for a thermal energy distribution.

Details of the Rydberg resonances are shown in Fig. 3. The theory, shown by a dashed line, is in good agreement with the data for the n=3, 4, and 5 peaks as well as for the Rydberg limit peak of the  $1s2p(^1P)$  series. However, as discussed below, there is some disagreement

TABLE I. Observed and predicted resonance energies and energy-integrated cross sections for dielectronic recombination of  $C^{4+}$  ions via the 1s2l2l' doubly excited states.

	Resonance energy (eV)			Integrated cross section (10 <sup>-21</sup> cm <sup>2</sup> eV)			
$\operatorname{Term}$	Expt.ª	Theory <sup>b</sup>	Theory	Expt.a	Theory <sup>b</sup>	Theory	Theory
$1s(2s^2 {}^1S) {}^2S^e$		225.4	227.1		1.23	0.81	
$1s(2s2p^{3}P)^{2}P^{o}$	234.3(1)	233.6	235.5	39.3(9)	44.13	39.18	39.05
$1s(2s2p^{1}P)^{2}P^{o}$	237.8(3)	238.6	239.0	6.1(6)	5.36	4.82	1.32
$1s(2p^{2}  ^{1}D)^{2}D^{e}$	241.4(1)	241.6	242.3	34.5(8)	37.85	40.41	42.18
$1s(2p^2 {\ }^3P)^{\ 2}P^e$	243.8(8)		243.2	3.1(6)		1.25	
$1s(2p^2  {}^1S)  {}^2S^e$	247.9(2)	248.2	248.5	5.6(5)	7.58	6.71	8.41

<sup>&</sup>lt;sup>a</sup>Statistical errors only.

<sup>&</sup>lt;sup>b</sup>This work.

<sup>&</sup>lt;sup>c</sup>Reference [22].

<sup>&</sup>lt;sup>d</sup>Reference [8].

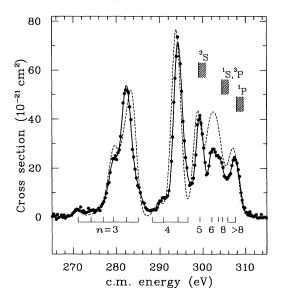


FIG. 3. Measured DR cross section due to the Rydberg resonances 1s2lnl' with fitted Gaussian functions (full line) and the convoluted theoretical cross section (dashed line). The labels below the curves indicate the line positions of the fitted Gaussian functions and their assignment to principal quantum numbers n to yield the results listed in Table II. The 1s2l excitation thresholds [21] are indicated at the top.

in the energy range between 300 and 308 eV, mainly for the resonances n = 6-8 of the  $1s2p(^{1}P)$  series, which appear as a single unresolved peak. The energy positions of Gaussian lines used to fit the data are indicated by vertical bars. For n = 3 and 4, the positions and heights of the indicated Gaussian components with a common width were varied in order to fit the data and to extract the energy-integrated cross section for each group of resonances. The n = 5 and 6 peaks were each fitted by a single Gaussian function of variable position, width, and height. For n = 7 and 8 the energies were fixed to the calculated Rydberg-level energies relative to the  $2^{1}P$  excitation threshold [21] and the widths were locked to that of the n=6 peak. The energy-integrated cross sections assigned to the states with different n are listed in Table II. The present theory reproduces within 10% the experimental results up to n = 5, whereas for n=3-5 the earlier single-configuration, nonrelativistic calculations [8] strongly underestimate the cross section. On the other hand, for the states with n = 6 and 7 even the present improved theory predicts much higher cross sections than observed.

To understand the origin of this discrepancy it is useful to consider more specifically the autoionization of the doubly excited states  $1s2p(^1P)nl'$  into the continuum channels opening up at the thresholds indicated in Fig. 3. The states with n=2-4 are closed to all continuum channels except the  $1^1S$  entrance channel. Theoretically, we find that n=5 is partially open to the  $2^3S$  continuum (5s, 5p closed, 5d, etc. open) and the good agreement with the experimental integrated cross section for n=5 appears to confirm this conclusion. A

TABLE II. Energy-integrated cross section  $(10^{-21} \, \text{cm}^2 \, \text{eV})$  for dielectronic recombination of ground-state C<sup>4+</sup> ions via resonances 1s2lnl'.

$\frac{}{n}$	Expt. <sup>a</sup>	Theory <sup>b</sup>	Theory	Theory
2	89(2)	96.2	102.9	93.2
3	260(4)	274.9	188.6	282.8
4	227(4)	223.0	163.3	
5	97(2)	110.2	57.5	
6	47(2)	81.8		
7	33(2)	69.0		
8	21(4)	18.6		
$\geq 9$	75(4)	90.8		
		Totals		
$\geq 6$	176(6)	260.2	272.1	
$\geq 2$	849(8)	964.5	784.4	

<sup>&</sup>lt;sup>a</sup>Statistical errors only.

similar problem arises for the n = 8 states, which are completely closed to the  $2^{3}P$  but only partially closed to the  $2^{1}S$  continuum. It is not possible to determine theoretically which of the  $1s2p(^{1}P)8p$  and 8d terms are open and which are closed (8s is almost certainly closed and 8f, 8g, etc. open). In the original work of Badnell, Pindzola, and Griffin [9] terms up to  $1s2p(^{1}P)8d^{-2}F^{o}$ came out as bound and this dominated DR of ions in the  $2^{3}S$  metastable state; hence, very little cross section was lost to the  $2^{1}S$  continuum (none in fact was shown in Fig. 2 of Ref. [9]). The relative height of the n=7 and 8 peaks agreed with the experimental results of Andersen, Bolko, and Kvistgaard [12], see Fig. 5 of Ref. [9], and no thought was given to reducing the n = 8 peak by opening up the 8p and 8d states to the  $2^{1}S$  continuum. Further experiments [13] on DR of metastable heliumlike ions show a consistent underestimation of the calculated cross section just below the almost degenerate  $2^{1}S$  and 2<sup>3</sup>P limits, apparently related to the radiative stabilization rates of the high Rydberg states  $1s2p(^3P)nl'$  and  $1s2p(^{1}S)nl'$ . There is no reason why the DR cross section in this energy range should not be underestimated by theory also for metastable  $C^{4+}$  ions. This, together with the fit results for n = 8 listed in Table II, seems to indicate that the 8p and 8d states are in fact open to the  $2^{1}S$  continuum.

Apart from these considerations, which helped us to improve the agreement for the n=8 integrated cross section, theory still overestimates the height of the peak composed of n=6 and 7. The reason for this disagreement is not clear. It would appear that the intermediate states  $1s2p(^1P)6l'$  and  $1s2p(^1P)7l'$  that are strongly coupled to the  $1^1S$  continuum are not coupled strongly enough to the  $2^3S$  continuum and that hence the reduction of the DR cross section by autoionization to this continuum is underestimated. For n=6 and 7, we find that the DR of ground-state ions is dominated by the  $1s2p(^1P)np$   $^2D^e$  term (60%) because it autoion-

<sup>&</sup>lt;sup>b</sup>This work.

<sup>&</sup>lt;sup>c</sup>Reference [8].

<sup>&</sup>lt;sup>d</sup>Z expansion; Ref. [22] (n=2) and Ref. [23] (n=3).

izes weakly to  $2^3S$ . A number of other terms populated equally strongly autoionize faster by up to a factor of 50. As a check on our perturbative autoionization rates we carried out a five-term close-coupling calculation using the R-matrix method [24]. Using quantum-defect theory, the threshold partial collision strengths were extrapolated down below threshold and compared with the n=6autoionization rates, yielding a difference of < 20%. In a model calculation in which the  $1s2p(^{1}P)np^{2}D^{e}$  autoionization rate to the  $2^3S$  continuum was increased by a factor of 20, integrated cross sections close to the experimental values were obtained for n = 6 and 7. We note that such an increase does not destroy agreement between theory and experiment for DR from the metastable ions [9, 12], but just reduces the fraction of metastable  $2^3S$ ions in the beam, required to interpret the experiment of Ref. [12], from 7% to 5%. The problem does not persist beyond n = 7 since the autoionization into the other continua then open dominates over that into the  $2^3S$ continuum.

As can be seen from the sum over states with n > 6in Table II, the predicted contribution of high Rydberg levels to the energy-integrated DR cross section is  $\approx 48\%$ higher than the experimental result. The earlier calculations [8] yield a similar result for these levels. For a thermal electron energy distribution of sufficiently high temperature ( $\gtrsim 1 \text{ keV}$ ), the DR rate coefficient will be approximately proportional to the energy-integrated cross section of all resonances. Hence, to illustrate the discrepancy between theory and experiment for the resulting DR rate coefficient in a hot plasma, we list in Table II also the sum over all observed resonances, i.e.,  $n \geq 2$ . The present theory overestimates the total integrated cross section by 14%, of which  $\approx 10\%$  are due to the Rydberg-state contribution  $(n \geq 6)$ . The earlier single-configuration, nonrelativistic calculations [8] underestimate the total integrated cross section by 8% and the contribution of the lower states up to n = 5 by  $\approx 24\%$ . This shows that the discrepancies revealed by this experiment are significant for the theoretical prediction of thermal DR rate coefficients if one aims at results accurate within 10% or less.

#### IV. CONCLUSION

We have measured as a function of the c.m. energy the cross section for dielectronic recombination of heliumlike  $C^{4+}$  ions in the  $1s^2$  ground state via the resonances 1s2lnl'. For  $n \leq 5$  the results are well reproduced by a theoretical calculation if relativistic effects and configuration mixing in the 1s2l3l' doubly excited states are included. The recombination cross section for higher states, in particular  $1s2p(^{1}P)$  6l' and  $1s2p(^{1}P)$  7l', is considerably lower than predicted by theory; apparently the autoionization rate to the 1s2s  $^{1}S$  continuum is strongly underestimated for the resonances dominating the DR cross section due to these levels. A better understanding of the mechanism leading to such discrepancies is highly desirable and may improve the theoretical description of dielectronic recombination also in other, more complex systems, where interlacing Rydberg series converging to closely spaced excitation thresholds of the core are observed frequently.

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