Strong Relativistic Effects and Natural Linewidths Observed in Dielectronic Recombination of Lithiumlike Carbon

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The dielectronic recombination spectrum of C^{3+} was recorded for low collision energies. The spectrum of the $1s^2 2p4\ell\ ^{1,3}L$ resonances in the range 0–0.6 eV was analyzed in detail. It was found that the strongest peaks in the spectrum must be explained by dielectronic recombination mediated by relativistic effects, a process forbidden in pure LS coupling. For strongly autoionizing states the natural linewidths were observed. The spectrum was assigned in detail by the aid of very accurate relativistic many-body perturbation calculations. [S0031-9007(98)06600-9]

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Few-electron systems of light elements are usually very well described in terms of LS coupling. Selection rules for atomic processes like radiative transitions, photoionization, and autoionization can be expressed in terms of L and S. For instance, in an electric dipole transition $\Delta S = 0$, $\Delta L = \pm 1$ or 0 while parity is changed, and for Coulombic autoionization S, L, and parity are all conserved. It is well known that intercombination transitions, i.e., spin-forbidden radiative transitions ($\Delta S \neq 0$), are extremely weak for light elements. The extent to which such selection rules are obeyed indicates in some sense the validity of LS coupling.

Dielectronic recombination is a process in which an electron is recombined with an ion through interaction with the electrons of the ion. The first step of the process is the reversal of autoionization; i.e., a free electron interacts with the bound electrons to form a doubly excited state. This state can decay by autoionization, but it can also relax by a radiative transition to a bound state. In the latter case, dielectronic recombination is completed.

In the present work, dielectronic recombination (DR) has been used as a tool to gain spectroscopic information on autoionizing states of the displaced system of berylliumlike carbon. For a Be-like system there are two ionization limits present: $1s^2 2s\ ^2S$ and $1s^2 2p\ ^2P$. In Be-like carbon (i.e., C^{2+}), these two thresholds are separated by 8 eV. Separate Rydberg series run toward these limits. Consequently there will exist levels of the type $1s^2 2pn\ell\ ^{1,3}L$ which are energetically degenerate with continuum states $1s^2 2s\varepsilon\ell\ ^{1,3}L$. If the states of the former type have the same L, S, and parity as some continuum state, nonrelativistically allowed autoionization will occur through electron-electron interaction. On the other hand, if no such symmetry exists, the state will be stable against such autoionization. For an experimental investigation utilizing dielectronic recombination, resonances of certain LS symmetries will be observed in the energy range 0–8 eV of the spectrum.

The experiment was conducted in the ion storage ring CRYRING [1] at the Manne Siegbahn Laboratory in Stockholm. In the electron beam ion source CRYSIS, C^{3+} ions were produced, which were accelerated to an energy of 6 MeV/amu. The ions were then cooled to a narrow velocity distribution in the electron cooler by merging the ion beam with a beam of cold electrons having the same average velocity as the ions. After preparing the ion beam in this way, the kinetic energy of the electrons relative to the ions was scanned in order to induce dielectronic recombination. By gradually detuning the acceleration voltage of the electron gun up to 360 V away from the cooling voltage, the relative energy in the center-of-mass system was varied from 0 to 8 eV, while the number of charge-changed ions (C^{2+}) was detected. The beam current of the stored ions was 1 μA and the storage lifetime was about 40 s. More experimental details can be found in Refs. [2,3].

The resolution obtained in the DR spectrum depends on the energy distribution (the "temperature") of the electron beam. The velocity distribution of the electrons consists of two components, the transversal and the longitudinal. Because of the kinematic compression obtained in a fast beam, the longitudinal component is usually small. At low relative energy, the transversal component limits the resolution. Danared *et al.* [4] introduced adiabatic expansion of the electron beam to reduce the transversal temperature by a factor of 10, resulting in a measured $kT_\perp$ of 10 meV. The reduced transversal temperature is hence extremely valuable in the present experiment where many resonances lie below 1 eV.

As mentioned above, Be-like systems have an interesting structure with both ordinary and displaced terms. In C^{2+} the displaced terms ($1s^2 2pn\ell\ ^{1,3}L$) pass the $1s^2 2s\ ^2S$ limit for the principal quantum number $n = 4$. For $n = 4$ there are four displaced configurations ($2p4s$, $2p4p$, $2p4d$, and $2p4f$), which form 20 LS terms. Nine of these lie below the $1s^2 2s\ ^2S$ limit, while eleven lie above; see Fig. 1. Among the latter terms, there are three with

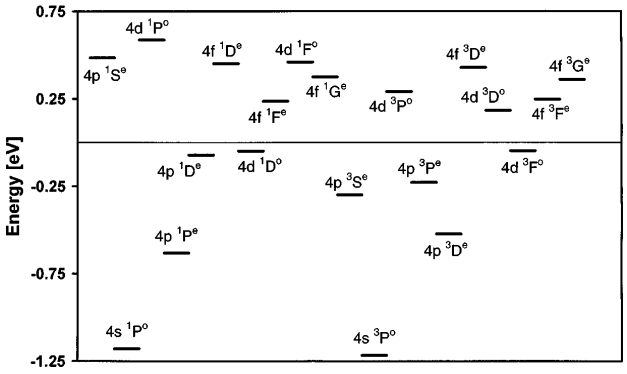

FIG. 1. Partial term diagram showing all LS terms of the $1s^2 2p4\ell$ configurations (i.e., the "displaced" terms. Eleven of the twenty terms based on these configurations lie above the first ionization threshold ($1s^2 2s\ ^2S$) and could possibly contribute to dielectronic recombination.

combinations of L, S, and parity which do not coincide with the energetically degenerate continuum states $1s^2 2s\varepsilon\ell\ ^{1,3}L$ ($L = \ell$). These states are consequently expected to have negligible autoionization rates.

The lowest part of the experimental DR spectrum is shown in Fig. 2(a). As can be seen there are two prominent peaks, at about 0.17 and 0.24 eV, respectively. The

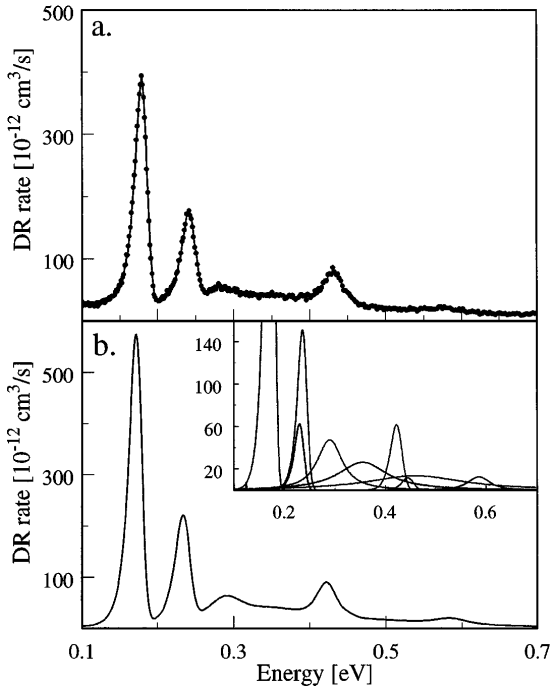

FIG. 2. In part (a) the experimental DR spectrum of C^{3+} in the range 0.1–0.7 eV is shown. A line is drawn between the data points to guide the eye. In part (b) the calculated spectrum is given. The theoretical spectrum includes the calculated widths, and the spectrum has also been folded with the experimental response function. The inset shows the contribution from each individual resonance included to form the total theoretical spectrum.

shape of these peaks is very well described with the expected *flattened electron temperature distribution* [3], which gives rise to an asymmetric shape. In fact, the first peak in the spectrum is, so far, the best available probe of the transverse electron temperature of the electron cooler in CRYRING. The spectral features for higher energies were more difficult to explain, in particular, the plateau-shaped part between 0.24 and 0.45 eV. Several experimental spectra were recorded, also with varying electron density, but the plateau was completely reproducible. In order to explain the plateau by "ordinary" DR resonances, dominated by the experimental width, more peaks are required than available from the possible terms in this region. Preliminary calculations by the Cowan code [5] (accurate within 0.05–0.1 eV in this case) did not explain either the first two peaks or the plateau.

More accurate calculations were performed by a method that combines relativistic many-body perturbation theory in all-order formulation (Lindroth and Hvarfner [6]) with complex rotation. This method has been shown to give very accurate results (see, e.g., Zong *et al.* [7] and references therein). From the calculations we obtained energies, widths, transition probabilities for both radiative decay and autoionization, as well as the resonance strength (i.e., the cross section for dielectronic recombination integrated over the line profile) (Table I). The high accuracy of the present calculations was confirmed by computing the $2s$-$2p_{1/2}$ and $2s$-$2p_{3/2}$ splittings in the lithiumlike system to be 7.9950 and 8.0085 eV, respectively. The corresponding experimental values, obtained by Bockasten *et al.* [8], are 7.9950 and 8.0083 eV.

The calculations revealed *two remarkable facts*. First, the two terms $2p4d\ ^3P$ and $2p4f\ ^3G$, which lie in the plateau region of the spectrum, have *very large natural widths*, 52 and 115 meV, respectively. Second, the "nonautoionizing" terms $2p4d\ ^3D$ and $2p4f\ ^{1,3}F$, not expected to contribute to dielectronic recombination due to negligible overlap with the continuum, were quite surprisingly found to be the *strongest* DR resonances. Since the autoionization of these states is very weak, the natural width is not observed but only the experimental response function.

The strength of a dielectronic resonance can be expressed as

$$S_d = \frac{\hbar\pi^2}{k^2}\,\frac{g_d}{g_i}\,\frac{A^a_{i\to d}\sum_s A^{\rm rad}_{d\to s}}{A^a_{i\to d} + \sum_s A^{\rm rad}_{d\to s}},$$

where the multiplicity of the intermediate doubly excited state is given by g_d and that of the initial state by g_i. The electron wave number is $k = p/\hbar$. The transition rate for population of the doubly excited state is given by $A^a_{i\to d}$ (i.e., the autoionization rate) and the rate for a radiative transition from the doubly excited state d to a lower state s is given by $A^{\rm rad}_{d\to s}$.

The equation shows that the strength is determined by the weakest of the decay modes. Often autoionization is

TABLE I. The calculated values of the energies (relative the $1s^2 2s\ ^2S$ threshold) and the widths of the resonances discussed in this work. Every fine structure level is given separately, since the widths, in the LS-forbidden cases, are strongly J dependent. The calculated strength (i.e., the integrated cross section) is also given.

Term	Level J	Energy (eV)	Width (meV)	Strength (10^{-20} eV cm^2)
$2p4d\ ^3D$	1	0.176	0.09	10.5
	2	0.177	0.18	17.7
	3	0.180	0.08	22.8
$2p4f\ ^1F$	3	0.236	0.10	5.4
$2p4f\ ^3F$	2	0.240	0.001	1.7
	3	0.242	0.25	5.0
	4	0.243	0.33	6.4
$2p4d\ ^4P$	0	0.292	52	1.3
	1	0.289	52	4.0
	2	0.285	52	6.8
$2p4f\ ^3G$	3	0.351	115	3.3
	4	0.353	115	4.2
	5	0.360	115	5.1
$2p4f\ ^1G$	4	0.375	115	2.2
$2p4f\ ^3D$	1	0.433	1.01	1.0
	2	0.430	1.01	1.6
	3	0.426	1.01	2.3
$2p4f\ ^1D$	2	0.452	0.22	0.9
$2p4d\ ^1F$	3	0.460	236	11.3
$2p4p\ ^1S$	0	0.485	221	0.2
$2p4d\ ^1P$	1	0.586	46	2.0

orders of magnitudes stronger than the radiative decay. In the present case, the calculated autoionization rate for the $2p4f\ ^3G$ is, for example, 7 orders of magnitude larger than the radiative decay rate. The DR rate can, however, be very large even if the autoionization rate is very low as long as it is larger than the radiative decay. From the formula above it is also seen that the resonance strength increases for low energies.

The two resonances observed at the lowest energies correspond to the $2p4d\ ^3D$ and the $2p4f\ ^3F, ^1F$ states (the latter terms cannot be experimentally resolved). As mentioned above, Coulomb interaction alone cannot mediate this recombination. It is only possible in the additional presence of the spin-orbit interaction, i.e., when only the total angular momentum J could be considered good. The observation does not imply that there is a breakdown in the LS-coupling approximation for this particular system. The fully relativistic calculation is performed in jj-coupling, but a recoupling of the state vectors shows that the LS mixing is very small. The $2p4d\ ^3D_3$, for example, consists to 99.95% of ^3D and only by totally 0.05% for ^3F (0.016%) and ^1F (0.032%), where only the ^1F term opens the way for dielectronic recombination since the ^3F state is below the threshold. For the $2p4d\ ^3D$ term, the LS mixing leads to autoionization rates for the three fine structure components ($J = 1, 2, 3$) of 134, 224, and 56 ns^{-1}, respectively. These rates are 3 to 4 orders of magnitude lower than for $2p4d\ ^3P$ and $2p4d\ ^1F$, but still much higher than the radiative decay rate of 5 ns^{-1}.

In Fig. 2 the experimental data are compared to the theoretical results. As can be seen, very good agreement is obtained. The calculated spectrum has been folded with the experimental response function. Every individual resonance that composes the total spectrum is plotted in the inset of Fig. 2(b). It is clearly seen that only a few broadened lines are needed to create the plateau-shaped region between 0.3 and 0.4 eV. We have also used the theoretical values as initial values for peak fitting by the commercial program PEAKFIT. For the lines predicted narrow, the analytical expression for a DR profile [3] was used, while for wide lines the experimental response function was approximated by a Gaussian line profile folded by a Lorentzian. Because of the number of unresolved peaks of different widths in the spectrum, it is, however, not possible to extract accurate values for every spectral line on experimental grounds. However, a very interesting conclusion can be drawn: the shape of the observed spectrum can only be explained, and very well described, *by strong relativistic effects and large natural line widths*. For the isolated spectral features, experimental energies could be determined: 0.182 eV for $2p4d\ ^3D$, 0.244 eV for the unresolved $2p4f\ ^{1,3}F$, 0.438 eV for $2p4f\ ^3D$, and 0.578 eV for $2p4d\ ^1P$. The scatter between different measurements is about 2 meV. By inclusion of the uncertainty for the absolute energy scale, we estimate the final experimental uncertainty to be 5 meV. The shape of the isolated DR profiles is excellently reproduced by the analytic function, indicating that no significant interference from radiative recombination is present.

These observations are indeed remarkable. Until now, studies by dielectronic recombination have not been done with sufficient resolution to clearly reveal the natural linewidths. The present observation was made possible by improved instrumental resolution in combination with very high autoionization rates for certain states. To the best of our knowledge, experimental evidence that *very strong contribution to dielectronic recombination could occur through relativistic effects* has not been presented previously. The two strongest peaks in the observed spectrum, which are caused by such effects, were observed by Andersen *et al.* [9] in a single pass experiment, but the resolution was not sufficient to permit identification of individual states. A similar spectral feature observed in an Auger spectrum has also been reported by Stolterfoht [10].

In a theoretical paper by Griffin *et al.* [11], the importance of using intermediate coupling instead of LS coupling for lithiumlike ions was pointed out. They found that the DR cross section increased substantially due to slight mixing between different LS terms. This is the same effect as reported in the present Letter, but here

we analyze selectively individual LS terms in detail, both theoretically and experimentally.

As discussed above, the autoionization rate may vary many orders of magnitude without loss of the DR rate, as long as the autoionization rate is larger than the radiative decay rate. A phenomenon related to the present observation has been reported earlier. In a beam-foil study of doubly excited states in neutral lithium by Mannervik and Cederquist [12], it was observed that the radiative lifetimes for quartet states above the $1s2s\ ^3S$ threshold was substantially shorter than nonrelativistic calculations predicted. For quartets below the threshold, however, the agreement was excellent. The effect was explained by relativistic autoionization (spin-induced autoionization) which could be amplified for states just above the limit, for which low-energy electrons could be emitted. Similar observations have also been reported in lifetime measurements in neutral beryllium by Ellis *et al.* [13] and in Na-like S^{5+} and Cl^{6+} Bengtsson *et al.* [14].

The present observation and the previous beam-foil lifetime measurements are two sides of the same coin. A very weak departure from *LS* coupling through relativistic interaction manifests itself in a strong observable effect.

The present calculations have high accuracy as can be seen in Fig. 2. The calculated rate is, however, larger than the experimental one. For the two lowest peaks the discrepancy is about 50%, but it is smaller for peaks at higher energies (see Fig. 2). A similar observation was made by Griffin *et al.* [15] in comparison with the data of Andersen *et al.* [9]. We have checked for excitation energies and radiative transition rates, for which the collisional strength should be most sensitive, but it is not clear whether the discrepancy is due to an experimental effect or due to an additional effect not included in the theoretical treatment. For the energies, the agreement is better than 10 meV for the resolved lines for which peak fitting could be used to obtain line positions. We note that Bockasten [16] observed a number of terms above the $1s^22s\ ^2S$ limit which "should not be affected by autoionization," among them $1s^22p4d\ ^3D_3$, for which he gave 0.1839 eV.

We conclude that the present work reveals that relativistic effects could be of major importance in dielectronic recombination even if the atomic system is described with a very high accuracy in (nonrelativistic) *LS* coupling. In particular, for collisions at low energies, it is not sufficient to perform DR calculations only in the *LS* coupling approximation. It is also shown that with the narrow experimental linewidth that can be obtained at the electron cooler of CRYRING, it is possible to resolve natural linewidth of autoionizing doubly excited states.

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[1] K. Abrahamsson *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **79**, 269 (1993).
[2] D. R. DeWitt *et al.*, Phys. Rev. A **50**, 1257 (1994).
[3] D. R. DeWitt *et al.*, Phys. Rev. A **53**, 2327 (1996).
[4] H. Danared *et al.*, Phys. Rev. Lett. **72**, 3775 (1994).
[5] R. D. Cowan, *The Theory of Atomic Structure and Spectra* (University of California Press, Berkeley, 1981).
[6] E. Lindroth and J. Hvarfner, Phys. Rev. **45**, 2771 (1991).
[7] W. Zong, R. Schuch, E. Lindroth, H. Gao, D. R. DeWitt, S. Asp, and H. Danared, Phys. Rev. A **56**, 386 (1997).
[8] K. Bockasten, R. Hallin, and T. P. Hughes, Proc. Phys. Soc. **81**, 522 (1963).
[9] L. H. Andersen, J. Bolko, and P. Kvistgaard, Phys. Rev. A **41**, 1293 (1990).
[10] N. Stolterfoht, Phys. Rep. **146**, 315 (1987).
[11] D. C. Griffin, M. S. Pindzola, and C. Bottcher, Phys. Rev. A **31**, 568 (1985).
[12] S. Mannervik and H. Cederquist, J. Phys. B **19**, L297 (1986).
[13] D. G. Ellis, I. Martinson, and M. Westerlind, Phys. Scr. **49**, 561 (1994).
[14] P. Bengtsson *et al.*, Phys. Scr. **49**, 303 (1994).
[15] D. C. Griffin, M. S. Pindzola, and P. Krylstedt, Phys. Rev. A **40**, 6699 (1989).
[16] K. Bockasten, Arkiv Fysik **9**, 457 (1955).