# Observation of Trielectronic Recombination in Be-like Cl Ions

M. Schnell,[1] G. Gwinner,[1] N. R. Badnell,[2] M. E. Bannister,[3] S. Böhm,[4] J. Colgan,[5] S. Kieslich,[4] S. D. Loch,[5] D. Mitnik,[6] A. Müller,[4] M. S. Pindzola,[5] S. Schippers,[4] D. Schwalm,[1] W. Shi,[4] A. Wolf,[1] and S.-G. Zhou[1,7]

[1]*Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany*
[2]*Department of Physics, University of Strathclyde, Glasgow, G4 0NG, United Kingdom*
[3]*Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*
[4]*Institut für Kernphysik, Justus-Liebig-Universität, 35392 Giessen, Germany*
[5]*Department of Physics, Auburn University, Auburn, Alabama 36849, USA*
[6]*Departamento de Física, Universidad de Buenos Aires, FCEyN Buenos Aires (C1428EGA), Argentina*
[7]*School of Physics, Peking University, Beijing 100871, China*
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Recombination involving the core excitation of two electrons, which may be termed trielectronic recombination, has been experimentally identified for the first time. Using $Cl^{13+}$ ions circulating in the TSR heavy-ion storage ring, we have observed surprisingly strong low-energy trielectronic recombination resonances, comparable to the dielectronic process. At higher electron-ion collision energies, trielectronic recombination is suppressed due to the autoionization of the triply excited intermediate state into excited final states. The formation of the intermediate state depends sensitively on configuration mixing, making trielectronic recombination a challenge to atomic-structure calculations.

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Electron-ion recombination in dilute plasmas can proceed in two ways. First, one can have the capture of the free electron into a nonresonant bound state accompanied by the emission of a photon, referred to as radiative recombination. Second, the recombination can involve the excitation of the core electron configuration in the target ion and the capture of an electron into a resonant state, followed by radiative deexcitation into a state stable against autoionization. For this latter process only single electron excitation of the core has been experimentally observed; hence the process is referred to as dielectronic recombination (DR).

In many ions, DR is the dominating recombination pathway and, therefore, the most important cooling mechanism in many astrophysical plasmas. Experimenting for the first time with Be-like ions at the TSR electron-cooler facility, we report the observation of electronic recombination involving the excitation of two core electrons into a triply excited intermediate state, that is, trielectronic recombination (TR). TR resonances represent more complex features than DR resonances because radiative stabilization competes with additional autoionizing decay channels not present in DR.

Trielectronic recombination is expected to be significantly weaker than DR, and a suitable atomic system must be chosen for a successful identification. So far, a search for TR in He-like krypton using crystal channeling has resulted in an upper limit for the cross section, but no observation [1]. In experiments on electron-impact ionization of $Li^+$, the formation of triply excited intermediate states by trielectronic capture was seen, but no radiative stabilization [2]. An efficient mechanism for a double excitation of the core is the strong mixing between $s^2$ and $p^2$ configurations, for example, in He-like or Be-like ions. An important advantage of Be-like ions is that, due to the comparatively low $2s \rightarrow 2p$ excitation energy, triply excited states exist which can stabilize to a level below the ionization threshold of the B-like ion via the emission of a single photon. From our experimental point of view, $(\Delta N = 0)\, 2s^2 \rightarrow 2p^2$ excitations in Be-like ions are also more favorable compared to $(\Delta N = 1)\, 1s^2 \rightarrow 2s^2$ or $2p^2$ in He-like ions, as $\Delta N = 0$ DR/TR resonances appear at much lower electron-ion collision energies, where storage ring measurements have their best spectral resolution [3] ($N$ denotes the principal quantum number of a core electron, and $n$ refers to the captured Rydberg electron).

We have carried out measurements with several Be-like ions ($F^{5+}$, $Cl^{13+}$, $Fe^{22+}$, and $Cu^{25+}$). Here we focus on chlorine, where the TR lines are the most striking, but they were present in all cases. The Van-de-Graaff Tandem/LINAC facility of the Max-Planck-Institute for Nuclear Physics in Heidelberg was used to accelerate Cl ions to 4 MeV/nucleon. Subsequently they were injected into the TSR storage ring, yielding stored ion currents of up to several hundred $\mu$A and a beam lifetime of 90 s. After each injection, the ion beam was electron cooled by merging a magnetically guided beam of cold electrons with the ions over a distance of about 1.5 m. In the ensuing measurement period, the electron-cooler device was used as an electron target. Recombination spectra were recorded by shifting the velocity of the electron beam periodically, introducing longitudinal electron-ion collision energies ranging from 100 $\mu$eV to around 100 eV. Recombined B-like ions were separated from the stored beam by the next dipole bending magnet behind the interaction region. Because of their high kinetic energy, single ions could easily be detected with $> 95\%$

  

efficiency in a CsI scintillator. From the measured rate, the known electron density, and the measured ion current, an absolute recombination rate coefficent as a function of energy $\alpha(E)$ could be determined with a relative uncertainty of about 15% accuracy (details of the experimental procedure are found in [4,5]). The recombination rate coefficient is the rate per ion and per electron density and is related to the recombination cross section $\sigma$ by $\alpha = \langle v\sigma \rangle$, where $v$ is the relative electron-ion velocity, and the bracket indicates averaging over the experimental velocity spread.

The measured $Cl^{13+}$ recombination spectrum is shown in Fig. 1. Resonant electron capture (DR or TR) leads to series of peaks, extending downward in energy from their series limits in a regular pattern of resonances at energies $E_{\rm res}$, well approximated by

$$E_{\rm res} = E_{\rm exc} - \frac{RZ_{\rm eff}^2}{n^2}, \quad (1)$$

where $E_{\rm exc}$ is the core excitation energy, $R$ is the Rydberg constant, $Z_{\rm eff} = Z - 4$ is the charge of the target ion, and the electron is captured into a state $n$. Resonance positions based on Eq. (1) and empirical core excitation energies taken from the NIST database [6] are shown with the spectrum for both dielectronic ($2s^2 \to 2s2p$) and trielectronic ($2s^2 \to 2p^2$) $\Delta N = 0$ recombination. They account for all resonant recombination in $Cl^{13+}$ below 80 eV collision energy, as no $\Delta N > 0$ peaks ($E_{\rm exc} >$ 400 eV) fall into this range, and no sign of metastable

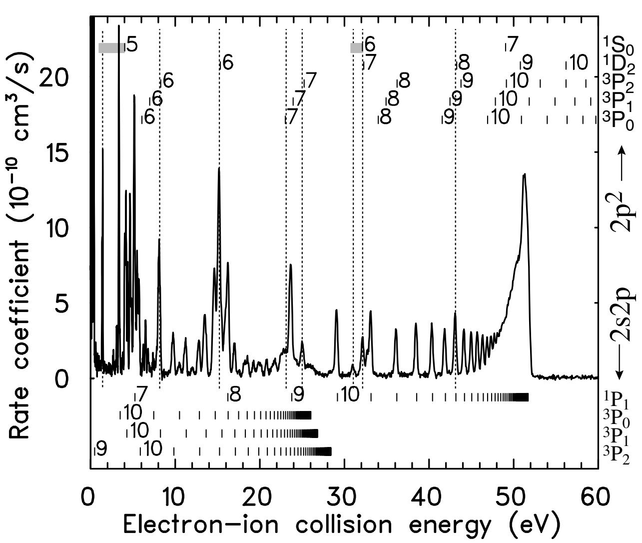


FIG. 1. Measured recombination spectrum of $Cl^{13+}$. Trielectronic resonances based on $2p^2$ core excitations are present and can be assigned to the five $2p^2$ series shown above the data (obvious examples are marked by dashed lines). The $2s2p$ DR series are at the bottom. In the case of the low-$n$ $^1S_0$ lines, where states of low angular momentum $l$ are shifted significantly with respect to high-$l$ ones, the calculated range of resonances is indicated by the grey bars.

$Cl^{13+}$ ions (e.g., $2s2p\,^3P_0$) was detected. DR is clearly dominating the spectrum. This is especially true for the singlet $2s^2 + e^- \to 2s2p\,(^1P)nl$ DR series anchored at 52 eV, which is prominently visible due to the pileup of unresolved high-$n$ Rydberg resonances. Mostly weaker resonances from triplet $2s^2 + e^- \to 2s2p(^3P_{0,1,2})nl$ DR are seen below $\approx 27$ eV. But, it is also apparent that some observed features (indicated by the dashed lines in Fig. 1) cannot be explained by DR. Some are additional freestanding resonances (e.g., at 1.5, 25, and 32 eV); others blend with DR lines, leading to enhanced peak heights (e.g., at 43 eV). The good alignment with predicted positions of $2s^2 \to 2p^2$ resonances suggests trielectronic recombination as their origin. Immediately, this assignment raises some issues: (i) none of the predicted TR resonances above the $^1P$ DR series limit at 52 eV are observed, despite the very low experimental background; (ii) triplet TR series are discernible only below the triplet DR series limit around 27 eV. The $2p^2(^3P)7l$ TR peaks are rather prominent, but the corresponding capture into $n = 8$ is already beyond the experimental sensitivity. Typical $\Delta N = 0$ DR spectra display a much smoother variation of resonance strengths with respect to $n$ (see, e.g., [7]). This behavior of the TR resonance strength along a Rydberg series is quite different from the situation in DR and reflects general decay properties of the triply excited intermediate states.

In DR (and TR), the recombination cross section via an intermediate state $s$ can be written as

$$\sigma^s_{\rm DR,TR} \propto A^a_{i\to s} \frac{\sum_f A^r_{s\to f}}{\sum A^r + \sum A^a}. \quad (2)$$

DR (TR) is proportional to the capture rate $A^a_{i\to s}$ (connected to the autoionization rate $A^a_{s\to i}$ via detailed balancing) from the initial state $i$ to the doubly (triply) excited state $s$. However, only a fraction of the ions—given by the second factor—stabilizes radiatively instead of autoionizing. Here $\sum_f A^r_{s\to f}$ is the radiative rate into all final states stable against autoionization, and $\sum A^r + \sum A^a$ is the total decay rate of the intermediate state $s$ via radiation and autoionization, respectively. For doubly excited intermediate states produced in $\Delta N = 0$ DR, we usually have the situation where the entrance channel is also the only relevant autoionizing *exit* channel (for exceptions due to fine structure, see, e.g., [7,8]). In contrast, a triply excited state can autoionize into the continuum of an *excited state*, e.g., $2s^2 + e^- \to 2p^2(^{2S+1}L)nl \to 2s2p(^{2S'+1}L') + e^-$. This is possible when the collision energy is above the limit of a DR series with appropriate symmetry. Hence, the data suggest that triply excited states based on a $2p^2\,(^3P)$ core with resonance energies above 27 eV decay predominantly via autoionization into $2s2p(^3P) + e^-$, preventing the completion of the recombination process. In much the same way, $2p^2(^1S,\,^1D)nl$ states are lost through autoionization into $2s2p(^1P) + e^-$

beyond 52 eV. So the spin multiplicity of the core state does not change in autoionization; i.e., $S' = S$. An important experimental consequence of this is that strong TR peaks can be observed only in regions where DR is present, and the highest possible energy resolution is required to resolve TR within the DR forest.

For a quantitative understanding, we have carried out theoretical calculations using the atomic-structure code AUTOSTRUCTURE [9]. In calculations for Be-like systems [10], the $2p^2$ configuration is routinely included to get the atomic-structure correct. Thus our calculation naturally returns both DR and TR resonance contributions. We modified our code to extract the resonance contributions attached to the $2s2p$ and $2p^2$ excited cores separately, to allow identification of TR resonances measured in the experiment. The results can be seen in Fig. 2. In the calculation the core energies have been shifted to their NIST values. Since there was no NIST value for the $2p^{2\,1}S_0$ core, a shift that gave the best agreement with the experimental positions of the $2p^{2\,1}S_0nl$ resonances at 1.5, 3.5, and 32 eV was chosen. As can be seen from Fig. 2, calculated and measured resonance positions agree well at all energies. The same holds for the TR resonance strengths at low energies, but theory increasingly underestimates the TR peak heights towards higher collision energies. The calculation of these individual TR peaks represents a challenge to existing theoretical calculations, due to the strong dependence on configuration mixing.

The steps in the TR spectrum discussed above can be quantified with the help of the calculation. The resonance strength $S_{LSj,n}$ for recombination via an intermediate state $2p^2(^{2S+1}L_j)n$ is given by the energy-integrated recombination cross section $\int \sigma_{LSj,n}(E)dE$ through this particular intermediate state. In Fig. 3 calculated values of $S_{LSj,n}$ for all TR series are plotted against the resonance energy. A drop by roughly 2 orders of magnitude is found as the TR resonance positions move above the DR series limits of the same multiplicity $2S+1$. This is consistent with the lack of experimental observation of triplet and singlet TR resonances above 27 and 52 eV, respectively. However, the current data cannot verify the size of the drops.

For application in astrophysics, the recombination rate coefficient has to be folded with a Maxwellian electron velocity distribution corresponding to the plasma temperature [5,11]. Some TR peaks are rather strong, and we find that they modify the Maxwellian-averaged rate coefficient $\bar{\alpha}(T_e)$ significantly at lower temperatures relevant to photoionized plasmas [11]. The contributions from the various processes to $\bar{\alpha}$ are shown in Fig. 4 as a

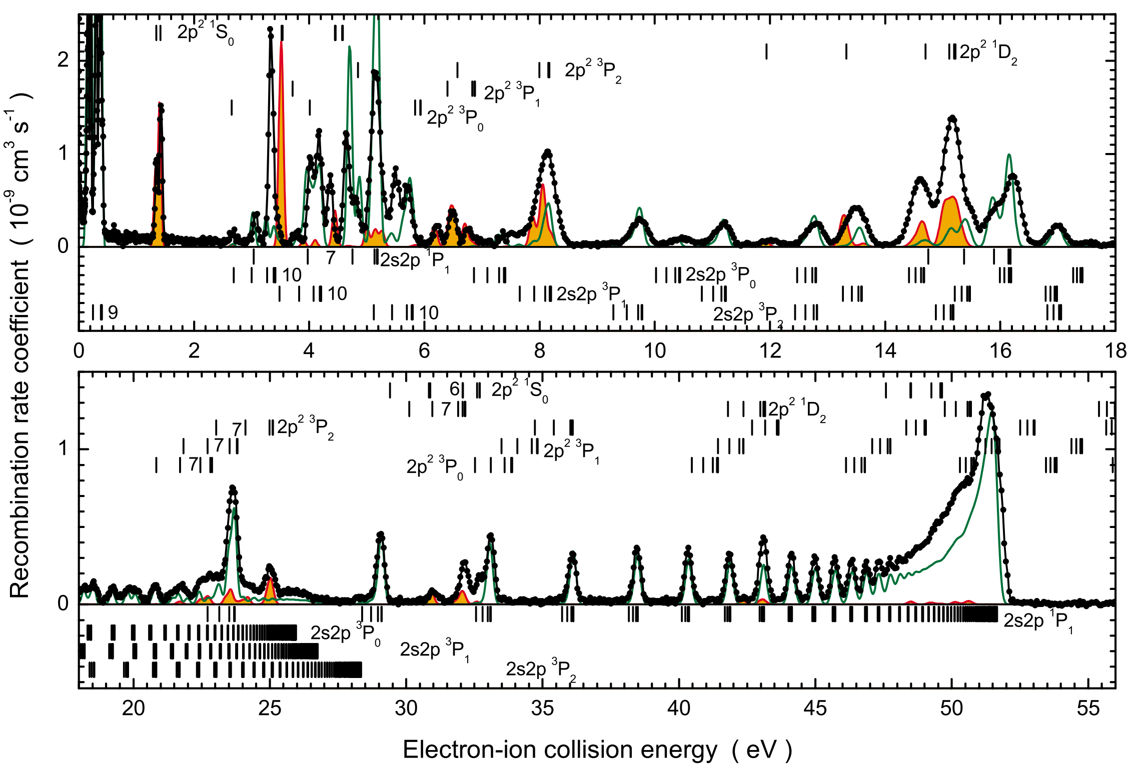


FIG. 2 (color online). Comparison of the measured recombination spectrum (black data points) with results from the AUTOSTRUCTURE calculation. The theoretical DR resonances are shown by the solid line, and TR resonances by the shaded areas. Energy positions for DR (TR) peaks are indicated below (above) the spectrum. The experimental energy axis has been scaled down by about 2% to achieve agreement with the known $^1P_1$ series limit at 52 eV. The disagreement of widths between 10 and 18 eV is due to a degraded instrument resolution.

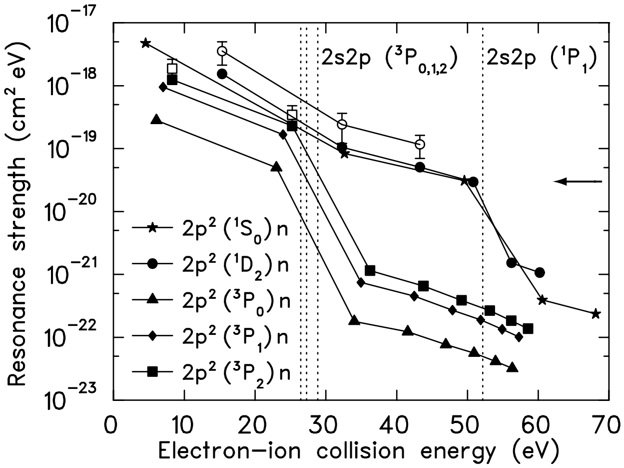


FIG. 3. Calculated TR resonance strengths plotted against the collision energy. Each point represents a sum over an $n$-manifold. The leftmost points correspond to $n = 6$, except for $^1S_0$, which starts with $n = 5$. The dashed vertical lines mark the DR series limits. The unfilled symbols show estimated measured strengths for a few select resonances (shapes identify the same series as filled symbols). This can be compared to the approximate experimental sensitivity indicated by the arrow.

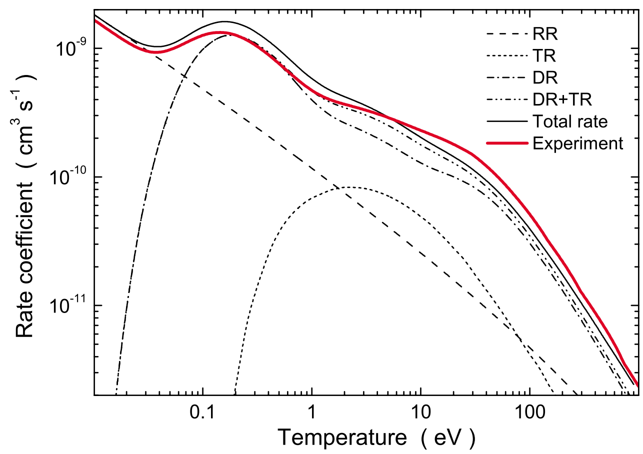


FIG. 4 (color online). Maxwellian-averaged recombination rate coefficient $\bar{\alpha}$ as a function of the electron temperature. $\Delta N > 0$ processes are not included but should not play a role at $T_e < 100$ eV. The calculation includes Rydberg states up to $n = 1000$. Field ionization in the magnets limits experiment to $n \lesssim 72$. For comparison with theory, a (small) calculated contribution of $n > 72$ states has been added to the experimental rate.

function of the electron temperature (as DR and TR cannot be disentangled reliably in parts of the experimental spectrum, we show theoretical rates). Everywhere between 1 and 100 eV, the TR contribution to the total rate is at least 10%; at $kT \approx 10$ eV it peaks at an estimated 20%–40%. The radiative stabilization of TR also produces additional line emission from the plasma and might be visible in astronomical observations. While the cosmic abundance of Cl is rather low, the situation in close-by, astrophysically more relevant elements such as Si, S, or Ar must be expected to be similar. Note that recombination onto long-lived metastable core states [e.g., $2s2p(^3P_0)$] could play a role in astrophysical environments, especially for nuclei with no magnetic moment; in the TSR these states seem to be quenched and are not considered in the calculation (see [12] for work on metastable Be-like ions).

In conclusion, we report the first observation of trielectronic recombination. In a domain of collision energies, where the intermediate triply excited state is stable against autoionization into excited final states, trielectronic resonances are found to be comparable in strength to their dielectronic counterparts. Above DR series limits, the strength of TR series of corresponding multiplicity drops dramatically and beyond observation as new autoionizing decay channels open up. Theory reproduces the TR spectrum and is used to isolate the TR contribution to the total rate. It is found that TR contributes up to 20%–40% of the total rate for plasmas with temperatures between 1 and 100 eV.

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