Dielectronic Capture with Subsequent Two-Electron Emission in Electron-Impact Ionization of C^{3+} Ions

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In electron-impact ionization of C^{3+} ions we found the first unambiguous experimental evidence for resonant-excitation auto-double-ionization processes. These involve capture of the projectile electron by the ion with simultaneous K-shell excitation and subsequent emission of two correlated electrons. The resonance strength measured for the $1s2s^22p\ ^3P$ contribution is $(2.0\pm0.4)\times10^{-20}\ \mathrm{cm}^2\ \mathrm{eV}$. In addition, we resolved individual thresholds of excitation autoionization via states $1s2s2l$ and identified other resonant contributions to net single ionization.

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Electron-impact ionization of ions is a fundamental process in hot plasmas. Intensive research during the last ten years has not only provided a substantial amount of cross-section data, but also revealed new physical insights into ionization mechanisms. Beside direct ejection of an electron from the ion, the most important indirect process contributing to single ionization is excitation of an inner-shell electron with subsequent autoionization.[1]

An exotic higher-order mechanism was predicted by LaGattuta and Hahn[2]: resonant excitation followed by double autoionization (REDA). This process involves capture of the projectile electron into a bound state of the one less charged ion with simultaneous excitation of an inner-shell electron. It is analogous to the first step of a dielectronic recombination process; however, the highly excited intermediate state does not decay by the emission of photons (as in dielectronic recombination) but by two *sequential* Auger processes emitting one electron each, thus leading to a net ionization of the ion. Another resonant process was first mentioned in a publication by Henry and Msezane[3]: resonant excitation with subsequent auto-double-ionization (READI). Again the first step is a dielectronic capture of the projectile electron, but now the intermediate state decays by *simultaneous* emission of two electrons. Recently, a theoretical effort was made to predict the possible contribution of READI to electron-impact ionization of ions along the lithium isoelectronic sequence.[4] The result was not promising to ever see READI in such an ion, for the maximum ratio of resonant versus direct single ionization of the $2s$ electron for ions with atomic numbers $Z\leq5$ was predicted to be between 0.6×10^{-3} and 6.5×10^{-3} for 2-eV energy width. That means the predicted READI resonances should sit on a "background" of direct ionization which is about 1000 times higher than the peak itself and explains why a serious attempt[5] to measure these resonances for O^{5+} ions had failed.

Stimulated by the very first recent observation of strong narrow resonances in the ionization of heavy metal ions,[6] we made an attempt to identify READI and REDA resonances in electron-impact ionization of Li-like C^{3+} ions. Our principal experimental setup has been described earlier.[7,8] The C^{3+} ions were produced from CH_4 in the Giessen electron-cyclotron-resonance ion source.[9] While we usually sweep the electron beam across the ion beam and thus obtain absolute cross sections,[7] we left the electron gun in a fixed position with optimum beam overlap and did fast electron-energy scans. The resulting relative measurements were calibrated against a number of absolute cross-section measurements which were taken by using our usual technique. The absolute cross sections thus obtained for the ionization of C^{3+} are in agreement with measurements of Crandall *et al.*[10] The uncertainty of the present measurements is about 0.15% statistical and at most 10% total. We scanned the energy range from 238 to 369 eV which covers all possible energies for resonant electron capture into C^{3+} ions. Cross sections ranging from 2.3 to $2.6\times10^{-18}\ \mathrm{cm}^2$ were measured at more than 2000 individual energies. Figure 1 shows the data smoothed over bins of seven adjacent energies. A wealth of structures is revealed arising from various K-shell excitation processes leading to a net ionization of C^{3+}. The most prominent contribution with a threshold at about 290 eV is due to direct excitation of the K shell with subsequent autoionization

$$C^{3+}(1s^22s)+e\rightarrow C^{3+}(1s2s2l)+e \\ \quad\hookrightarrow C^{4+}(1s^2)+e. \tag{1}$$

Also clearly visible are a number of resonance features which arise from dielectronic capture with subsequent two-electron emission:

$$C^{3+}(1s^22s)+e\rightarrow C^{2+}(1s2snln'l') \\ \quad\hookrightarrow C^{4+}(1s^2)+2e. \tag{2}$$

The energy range 238 to 258 eV contains resonances $1s2s2l2l'$ which can only contribute to the measured cross section through auto double ionization, i.e., emis-

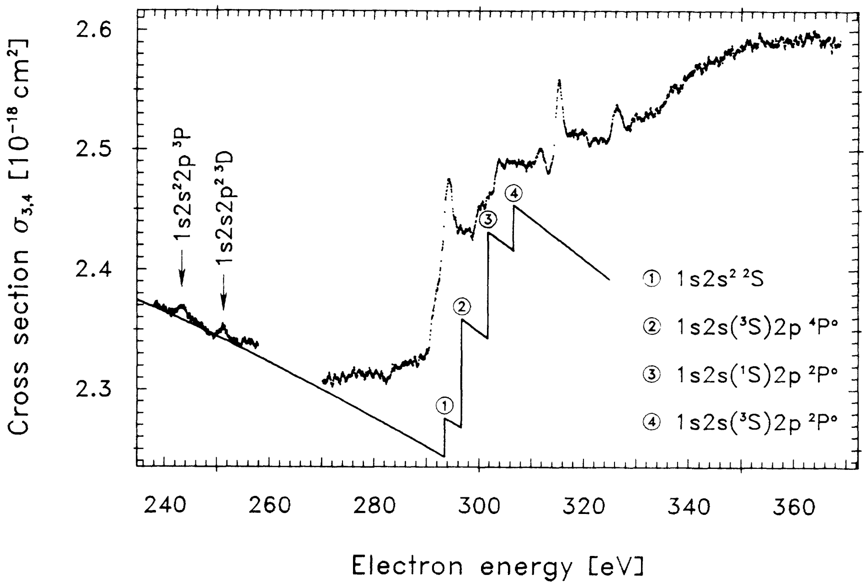

FIG. 1. Cross sections for electron-impact single ionization of C^{3+} ions. The scan data were smoothed over bins of seven adjacent points of the original measurement. For typical statistical uncertainties, see Fig. 2. The READI peaks are indicated. The solid line is the sum of the direct ionization [represented by $0.85\sigma_{2s}$; where σ_{2s} is Younger's calculation (Ref. 11)] and the excitation-autoionization contributions calculated by Henry (Ref. 12).

sion of correlated electrons because any single Auger process would lead to a bound state of a C^{3+} ion. In order to find the predicted small resonances, we scanned this range for more than 50 h. Figure 2 presents an enlarged plot of the experimental result. The two main resonances can be assigned to states $1s2s^22p\ ^3P$ and $1s2s2p^2\ ^3D$; the experimental energies (243.3 and 251.3 eV, respectively) agree well with calculations by Safronova and Lisina (243.0 and 251.2 eV).[13]

The straight line in Fig. 2 approximates Younger's calculated cross section σ_{2s} for direct ionization of the $2s$ electron.[11] It was multiplied by a factor 0.85 to fit our data and describes the "background" on top of which we observe the

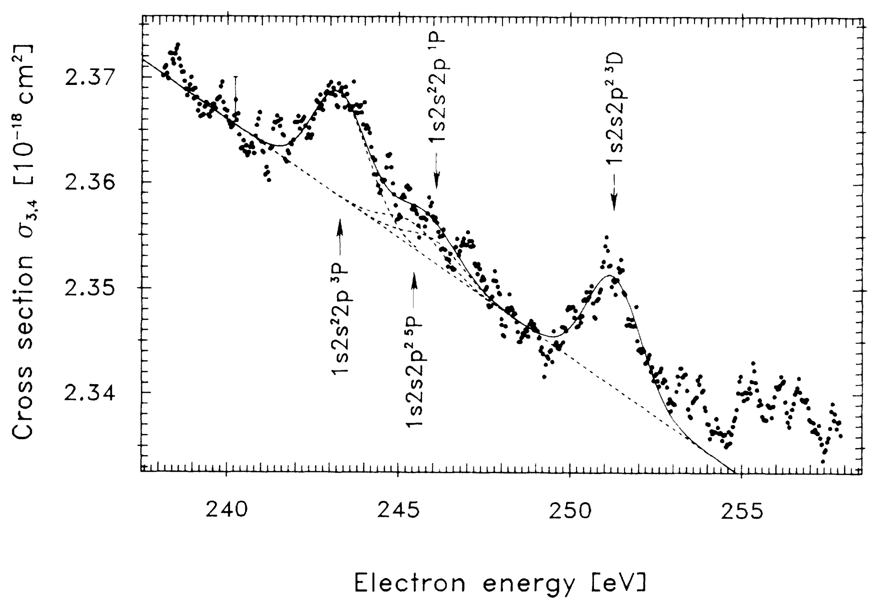

FIG. 2. Blow up of the low-energy part of Fig. 1. The statistical uncertainty is indicated. The assignment of resonant states to the peaks is based on calculated energy levels (Ref. 13). The straight dashed line approximates $0.85\sigma_{2s}$, the cross section for direct single ionization (Ref. 11). Added to this are resonant contributions represented by Gaussian distributions of 1.85-eV width.

various inner-shell contributions. We have fitted four Gaussian distributions (representing the assumed experimental energy spread) to the features rising from the background all with a width of 1.85 eV. The solid line represents the sum of the Gaussian distributions and $0.85\sigma_{2s}$. The experimental resonance strength $S=\int\sigma dE$ for the $1s2s^2 2p^3\ {}^3P$ contribution is $S=(2.0\pm0.4)\times10^{-20}$ cm^2 eV which has to be compared to the range of $S=0.58$ to 3.2×10^{-20} cm^2 eV predicted theoretically. The radiative decay of this resonance term can be ignored, because it is negligibly small.[4] There are no calculations for the other resonances assigned to the peaks in the data. The observed strength for the $1s2s2p^2\ {}^3D$ resonance is $S=(2.1\pm0.4)\times10^{-20}$ cm^2 eV. The shoulder appearing in our data around 246 eV can be due to contributions from $1s2s2p^2\ {}^5P$ and $1s2s^2 2p\ {}^1P$ states. Since the experimental resolution is not sufficient to separate these resonances, the evaluation of individual strengths becomes very uncertain. The total strength in the shoulder extracted from the data is $(9\pm5)\times10^{-21}$ cm^2 eV. According to theory[4] the contribution from the $1s2s^2 2p\ {}^1P$ state is negligible since its population by resonant recombination is about 3 orders of magnitude below that of the $1s2s^2\ {}^3P$ state.

The data seem to indicate additional (resonant) contributions at energies above 252 eV. Indeed there are more states with $1s2s2p^2$ configuration at energies up to about 258 eV which could influence the measured cross section.[13]

We did not do scans in the energy range between 258 and 270 eV because no resonances can be expected there. The next possible resonant states are $1s2s^2 3l$ states for which an estimate with Slater screening[14] predicts energies above 280 eV. Though the present data do not show any peaks in that region, the cross section rises above the normalized Younger cross section $0.85\sigma_{2s}$ already below the first threshold of direct K-shell excitation. An explanation is the accumulation of resonances $1s2s^2nl$ which can decay by auto double ionization and also of states $1s2s2pnl$ with $n\geq4$ which can additionally autoionize in two sequential steps by a Coster-Kronig transition and a subsequent Auger process. It is reasonable to assume a much higher probability for such contributions (double autoionization) than the one responsible for the energetically lower-lying resonances (auto double ionization).

There is an additional decay path for the $1s2s2lnl$ resonances if $n\gtrsim250$ which is opened by field ionization in the electron gun where electric fields as high as 5 V cm^{-1} might be present. Rydberg C^{3+} ions produced by autoionization of C^{2+} $(1s2s2lnl)$ and survival of the flight through the collision region can still be field ionized if $n\gtrsim100$ when these ions enter the analyzing magnet seeing a $\mathbf{v}\times\mathbf{B}$ field of up to 560 V cm^{-1}. Field ionization will happen already at the entrance to the magnetic field so that the resulting C^{4+} ions are collected by the ion detector.

With this in mind, one may expect a strong but energetically narrow resonance feature composed of contributions from an infinite number of states all with Rydberg quantum numbers greater than about 100 converging to a $1s2s2l$ excitation threshold. We suggest that the sizable resonance feature at 294 eV is due to this accumulation of Rydberg resonances.

From the data of Fig. 1, four steps can be distinguished related to excitation autoionization contributions via $1s2s2l$ states. These steps can be assigned to the excitation of ground-state C^{3+} ions to states $1s2s^2\ {}^2S$, $1s2s({}^3S)2p\ {}^4P^0$, $1s2s({}^1S)2p\ {}^2P^0$, and $1s2s({}^3S)2p\ {}^2P^0$.

There is another strong feature around 315 eV. The data below 310 eV and above 317 eV indicate at least one more excitation step in the cross-section function about 0.025×10^{-18} cm^2 high. There are two peaks in this energy range; one at 311.7 eV, the other at 315.2 eV with a valley in between, which is lower than expected from the course of the cross section below 310 eV. This makes the whole feature look like an interference pattern. Aside from the principal possibility for interference between amplitudes of different reaction paths leading to the observed ionization, one cannot easily assign a resonant process to the observed energy. All $1s2s2l$ excited C^{3+} states which can be populated by direct excitation of a K-shell electron are below 310 eV and hence also all resonances $1s2s2lnl'$, while all excited states $1s2snl$ with $n>3$ are around or above 236 eV.[12] Though, according to calculations with use of Slater screening, the lowest possible (REDA) resonances $1s2s3l3l'$ should be expected at energies above 320 eV, this simple approximation may not be good enough to rule out the appearance of these resonances at 315 eV. (By using that method, we assign a configuration $1s2s3l3l'$ to the peak at 326.3 eV and $1s2s3l4l'$ to the little bump barely showing up at about 330 eV.) In any case, the strong unpredicted features observed around 315 eV are surprising and require detailed theoretical analysis.

For the features under discussion, we suggest a process which involves capture of the free electron into a high Rydberg state with simultaneous excitation of a $1s$ electron to the $2p$ subshell *and* a simultaneous shakeup process[15] exciting the $2s$ electron to the $2p$ subshell. The series limit of such resonances would be a $1s2p^2\ {}^2S$ excitation which, according to Chen, has an energy of 313.4 eV.[16] Another explanation for the occurrence of $1s2p^2nl$ resonances is the strong mixing between $2s^2$ and $2p^2$ states.[17] Configuration interaction between $1s2s^2({}^2S)nl$ and $1s2p^2({}^2S)nl$ terms would transfer some $1s2s^2({}^2S)nl$ character to states assigned to $1s2p^2({}^2S)nl$, and thereby allow transitions from the $1s^2 2s$ ground state of C^{3+}. The measured cross section still increases with energy and appears to reach a maximum at about 360 eV. This energy is close to the highest series limit for resonant-excitation processes involving a K-shell electron (the ionization $1s^2 2s \rightarrow 1s2s$ has a threshold of 366.8 eV as calculated by use of Slater screening). Again it is possible that the twofold infinite series of Rydberg resonances $1s2ln'l'n''l''$ pile up below 366.6 eV and give rise to the observed cross-section increase.

In summary, we have demonstrated exciting new contributions to single ionization of multiply charged ions. The experimental technique used provides a new method for spectroscopy of intermediate states produced in electron-ion collisions. In particular, contributions of dielectronic capture with subsequent auto-double-ionization could be identified for the first time. Contributions from Rydberg states which are usually cut off by fields and hence are missing in dielectronic recombination measurements can be observed up to "infinite" n in electron-impact ionization experiments.

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