Resonances in Electron-Impact Single, Double, and Triple Ionization of Heavy Metal Ions

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We report narrow dielectronic-capture resonances decaying by double autoionization, observed for the first time in electron-impact ionization of ions. We also observe corresponding peaks in double and triple ionization leading us to postulate new processes where dielectronic capture is followed by triple or quadruple autoionization, respectively.

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Photoabsorption and electron-scattering processes in heavy atoms and ions offer an attractive opportunity to study atomic physics in a region where the atomic structure is dominated by electron-electron interactions. Experiments and calculations of photoionization involving electrons of the closed $4d$ subshell in atoms with atomic numbers close to $Z=56$ (Ba) have revealed dramatic qualitative changes in photoabsorption spectra due to profound term dependence of atomic orbitals for certain electron configurations.[1] Also, in some previous measurements of electron-impact double ionization of heavy ions, large anomalous features at electron energies less than 2 times the threshold energy were found.[2] Recently, some of these features were interpreted as giant scattering resonances appearing in the scattered-electron channel.[3] LaGattuta and Hahn[4] postulated a different type resonance process which they named "resonant-excitation double autoionization" or REDA which, for atomic structures similar to those studied in this paper, can be represented as

$$e+A^{q+}(4d^{10}5l^{\nu})\rightarrow A^{(q-1)+}(4d^{9}5l^{\nu}n'l'n''l'')\rightarrow A^{(q+1)+}(4d^{10}5l^{\nu-1})+e+e, \quad (1)$$

where it is assumed that the decay of the intermediate doubly excited state takes place by sequential Auger processes. One sees from Eq. (1) that the process is a resonant dielectronic capture followed by Auger ejection, and another term used in the literature is "resonant-recombination double autoionization." Contributions to ionization from such a process have been inferred from experimental data (e.g., Crandall *et al.*,[5] Henry and Mesezane,[5] and Griffin *et al.*[6]) from a filling in ("snow on the stairs") of the cross section for ionization below excitation-autoionization thresholds.

Here we report the observation of narrow peaks in electron-impact single-, double-, and triple-ionization cross sections for various ions with atomic numbers $Z=55$, 56, 57, and 58. Most of these peaks were found on the "rising side" of what one may hypothesize are giant resonances.[3] Apparently, they arise from identical states populated during the collision and then branch into single, double, and triple net ionization. Our interpretation is that we have for the first time measured REDA contributions to net single ionization of ions. Furthermore, our data suggest that presence of new processes, contributing to net double and triple ionization, namely, resonant capture followed by triple (RETA) or even quadruple (REQA) autoionization. These contributions may enhance a cross section by as much as 30%.

Our experimental methods for the measurement of absolute cross sections for electron-impact ionization of ions have been described in detail.[7] We have extended the technique by implementing a fast energy-scanning option for measurements of fine details in cross-section curves. For this purpose we leave the electron gun in a fixed position with optimum overlap of the electron and ion beams and vary the electron energy with simultaneous registration of signal plus background, ion current, and electron current. One scan spans typically over 10 eV with 256 discrete intermediate energies and takes, e.g., 17.5 s. By repeating scans hundreds of times we average out possible fluctuations in the form factor, measurements of beam currents, and counting rates, and in other sources of data scatter. The space-charge broadening of the electron energy distribution is offset by our filling the space-charge well with slow ions from Kr gas fed into the system for the purpose. This gives an electron energy spread of 0.4 eV or better as evidenced by the structures observed.

Figure 1 presents two examples of measured cross sections for Xe-like ions, showing some of the features mentioned above. Data[8] for Cs^{+} were chosen, since it has

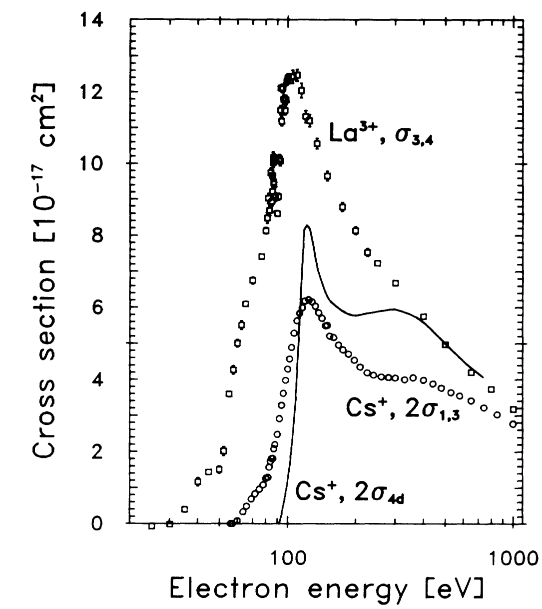

FIG. 1. Measured cross sections for single ionization of La^{3+} ions (squares) and double ionization of Cs^{1+} ions (circles; the Cs^{1+} data were multiplied by a factor of 2 for ease of display). Also shown is Younger's (Ref. 3) theoretical cross section σ_{4d} (solid line, also multiplied by 2) for ionization of the $4d$ subshell of Cs^{1+} ions with subsequent autoionization.

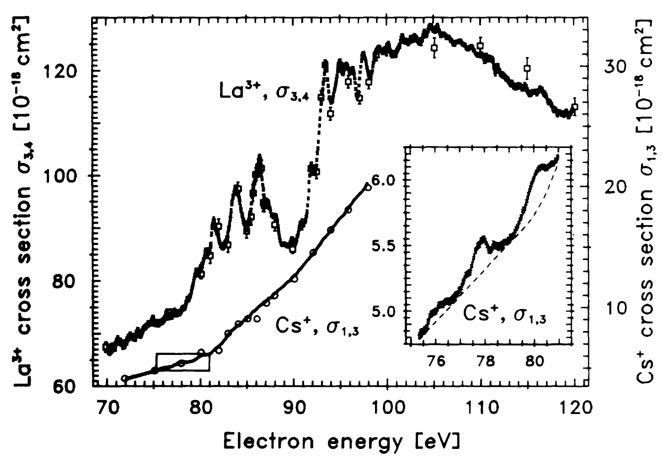

FIG. 2. Scan measurements of single ionization of La^{3+} ions (upper data set, left-hand scale) and double ionization of Cs^{1+} ions (lower data set, right-hand scale). The scan data were normalized to absolute cross sections from Fig. 1 (circles for Cs^{1+}, squares for La^{3+}) and smoothed over bins of five adjacent energies. Statistical uncertainties are indicated. Inset: Cs^{1+} double-ionization data in the energy range 75 to 82 eV. The dashed curve is assumed to approximate the nonresonant part of $\sigma_{1,3}$.

previously been fully identified[3] with the giant-resonance phenomenon, and we wanted to demonstrate the new features for this ion as well as the many others observed. To get the curves for both La^{3+} and Cs^{+} on the same plot, the cross sections for Cs^{+} have been multiplied by 2 for both our experimental data (circles) and the theory (for ionization of $4d$ electrons followed by autoionization) of Younger.[3] The theoretical giant-resonance feature occurs above the threshold for ejection of a $4d$ electron which is at 92.1 eV. In contrast, the experimental onset (see also Fig. 2) of the resonance peak is at about 80 eV suggesting additional contributions from excitation-double-autoionization (EDA) channels, e.g., through a $4d^9 4f$ intermediate state.

The cross section for single ionization of La^{3+} ions (squares) also exhibits a peak (at about 105 eV) with a shape resembling the giant resonance in Cs^{+}. However, ionization threshold from the $4d$ subshell are expected[9] now between 137 and 140 eV. Moreover, ionization of the $4d$ subshell would contribute nearly exclusively to double ionization due to subsequent Auger processes. Hence, we suggest that the large feature may possibly be a giant-resonance phenomenon in the excitation-autoionization channel of La^{3+} through states $4d^9 5s^2 5p^6 4f$ with excitation energies[9] between 97 and 130 eV.

Our looking closely at La^{3+} in the energy range from 70 to 120 eV by the fast-scanning technique reveals that the region abounds with structure in the ionization cross section as shown in Fig. 2 (upper data set, left-hand scale). Because of the complex electronic structures of rare-earth ions it is not readily possible to assign doubly excited states of La^{2+} ions to the observed resonances; however, we cannot think of a process other than REDA to produce such narrow features as those observed here.

The result of scan measurements for Cs^{+} in the vicinity of the expected EDA threshold at about 80 eV is also shown in Fig. 2 (lower data set, right-hand scale, and inset). The data indicate a change in slope at 90 eV, which is probably due to the onset of the predicted $4d$ giant-resonance ionization. At and below the suggested EDA threshold of 80 eV, we again found narrow peaks [see inset and Fig. 3(b)] which, following the interpretation of the La^{3+} results, are due to RETA. This hypothesis is supported by the fact that we also observed resonances with the same widths and positions in single ionization of Cs^{+} ions [Fig. 3(a)]. Thus, the REDA and RETA observed in these processes obviously result from identical states populated during the collision and then branching into net single or net double ionization.

Figure 3 presents scan measurements for single and double ionization of Cs^{+}, Ba^{2+} (both Xe-like), and La^{2+} (Cs-like) ions. For ease of display of resonant contributions to the cross sections we subtracted from the original measurements smooth curves (see dashed curve in inset of Fig. 2) resulting from spline fits through local minima in the measured cross sections. The fitted curve in each particular case (though somewhat arbitrarily chosen) is assumed to approximate the nonresonant por-

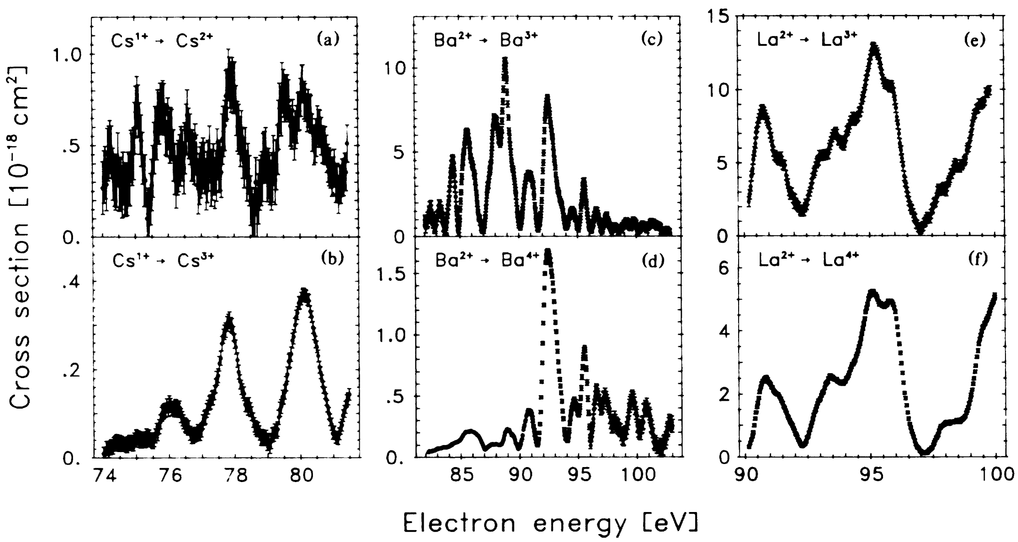

FIG. 3. Scan measurements of single (upper row) and double (lower row) ionization of ions (a),(b) Cs^{1+}, (c),(d) Ba^{2+}, and (e),(f), La^{2+}, after subtraction of approximated nonresonant contributions from the original data. The resonance cross sections displayed, e.g., for Cs^{1+} double ionization, (b), were obtained by subtraction of a smooth curve (see inset of Fig. 2) from the measured scan data. All data displayed in this figure were smoothed over bins of nine adjacent energies. Statistical uncertainties are indicated.

tion of the cross section. Because of high "background" arising from nonresonant ionization processes the statistical uncertainties in some of the displayed data are comparatively high. Note that double ionization of La^{3+} requires about 120 eV and hence the resonances showing up in single ionization (Fig. 2) cannot contribute here. The sharp features at 95 eV observed in single and double ionization of La^{2+} are probably due to an intermediate recombined $4d^9 5s^2 5p^6 5d^3$ state which can only lose three electrons in sequential autoionizations if at least one step involves a multielectron ejection, i.e., simultaneous emission of more than one electron.

We have also found peaks at identical energies in double and triple ionization of Ce^{1+} and Ce^{2+} ions. The subtracted (as for Fig. 3) resonance contributions are shown in Fig. 4. Again we have to conclude that the peaks observed are due to identical intermediate excited states, this time branching into net double and even triple ionization. The resonance positions can be associated with excitation of $4p$ rather than $4d$ electrons, as in the cases discussed so far.

In summary, we have made comprehensive measurements on single and multiple ionization of heavy metal ions and resolved numerous individual resonances in the cross sections. The only explanation presently possible for the resonances in single ionization is that they are due to resonant recombination and double autoionization (REDA). We see branching of resonant states into net double and net triple ionization and thus establish new mechanisms (RETA and REQA) contributing to multiple ionization of ions.

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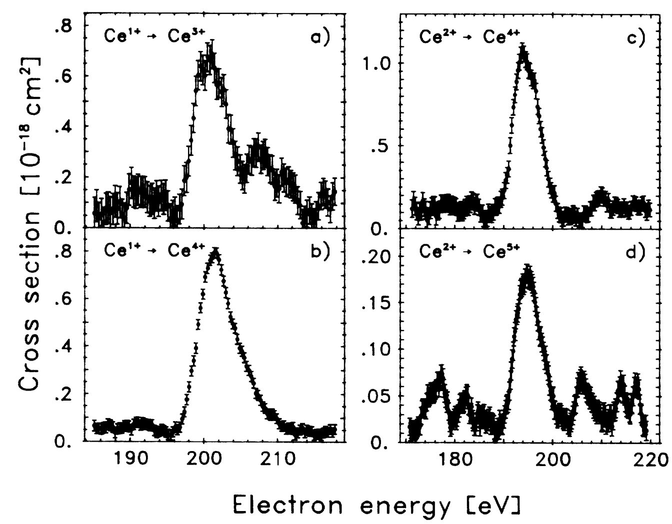

FIG. 4. Scan measurements of double (upper set) and triple (lower set) ionization of ions Ce^{1+} and Ce^{2+}, after subtraction of approximated nonresonant contributions from the original data. All data displayed in this figure were smoothed over bins of nine adjacent energies. Statistical uncertainties are indicated.

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