Electron-Impact Double Ionization of I^{1+} and Xe^{q+} ($q=1,\ldots,4$) Ions: Role of $4d$ Electrons Like in Photoionization

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The cross sections for electron-impact double ionization of I^{1+} and Xe^{1+} ions (and to a lesser extent also of Xe^{2+}, Xe^{3+}, and Xe^{4+} ions) show a dominant resonancelike contribution which, both in shape and size, almost coincides with the partial $4d$-photoionization cross section of Xe atoms.

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Features and mechanisms of multiple ionization of ions by electron impact are widely unexplored. At present there is only a single publication on multiple ionization experiments with multiply charged ions.[1] In a series of crossed beam experiments with Xe^{q+} ($q=1, \ldots, 4$) ions we have found that multiple ionization can be as important as single ionization; e.g., the ratio of double to single ionization reaches about 70% for 700-eV electrons incident on Xe^{2+} ions and is probably increasing with the electron energy. Hence, multiple ionization processes should not be neglected in modeling and diagnostics of high-temperature plasmas particularly for high-Z elements.

In this Letter we report on direct measurements of absolute cross sections $\sigma_{q,q+2}$ for electron-impact double ionization of Xe^{q+} ($q=1,\ldots,4$) ions and I^{1+} ions. We demonstrate the importance of a two-step mechanism involving single ionization of the $4d$ subshell and subsequent autoionization. Unexpectedly, the cross section $\sigma_{q,q+2}{}^{4d}$ assigned to this indirect process nearly coincides, both in shape and in size, with the partial $4d$-shell photoionization cross section of Xe atoms. Our results suggest that electron correlation, term dependence, and collapse of wave functions which are extensively discussed[2] in the context of *photoionization* of Xe, Cs, and Ba atoms and ions have to be considered for *electron-impact ionization* of complex ions, too.

The experimental technique used for the present measurements was similar to that described previously.[3] In brief, ions extracted from an electron-beam ion source are charge-state analyzed and crossed perpendicularly by a high-intensity electron beam (up to 140 mA/cm^2 at 700-eV electron energy) with an interaction length of 6 cm. The ratio of signal to background was typically up to 200 at 700-eV impact energy. Two important improvements were achieved since our first experiments: (i) simultaneous measurement of parent and product ions; (ii) improved determination of background resulting mainly from stripping collisions by moving the operating electron gun out of the ion beam line instead of switching off the electron beam. Thus, the relative uncertainties of the present cross-section measurements could be reduced to about 2%–3%. The total uncertainty varies with electron energy and is about 16% at 100 eV and 10% at 600 eV. The main source of error is due to the determination of the electron current density which was obtained by a relative measurement of cross sections $\sigma_{1,2}$ for Ar^{1+} ions and normalization to data published by Woodruff, Hublet, and Harrison.[4]

Figure 1 shows measured cross sections $\sigma_{1,3}$ for electron-impact double ionization of Xe^{1+} and I^{1+} ions. Both cross sections are very similar in size and energy dependence and—what is more surprising—they both exhibit a prominent peak with a resonancelike shape around 100-eV electron energy. A similar structure was found in the cross section for single ionization of Xe^{3+} ions and was attributed to *excitation* of $4d$ electrons followed by autoionization.[5] In our case "humps" have onsets at the ionization thresholds of the $4d$ shells of Xe^{1+} and I^{1+}, respectively, and can therefore be explained by *ionization* of $4d$ electrons followed by autoionization, thus leading to the loss of two electrons instead of one. For further investigation of this feature we have also measured cross sections $\sigma_{2,4}$, $\sigma_{3,5}$, and $\sigma_{4,6}$ for electron-impact double ionization of Xe^{2+}, Xe^{3+}, and Xe^{4+} ions, respectively (see Fig. 2). Similar

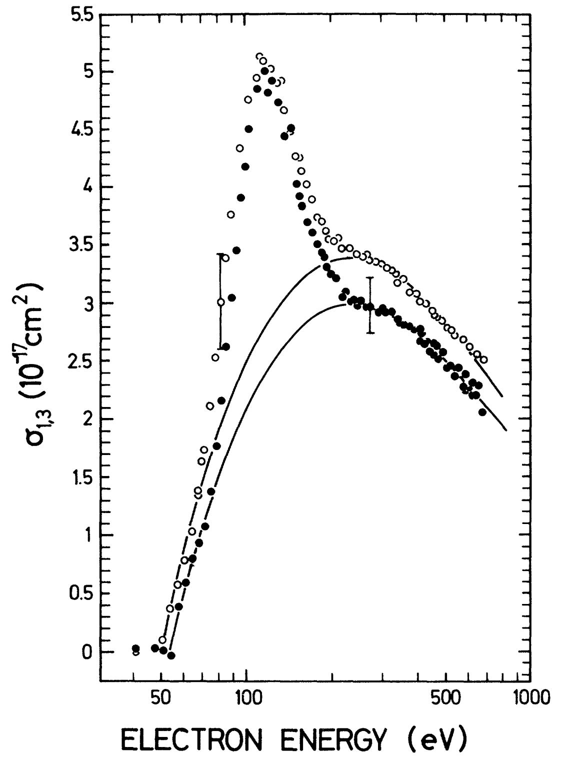

FIG. 1. Electron-impact double ionization cross sections $\sigma_{1,3}$ for Xe^{1+} (filled circles) and I^{1+} (open circles) ions. Typical total absolute uncertainties are indicated. The solid lines represent the direct "double-knockout" contribution which was obtained by a fitting procedure (see text) with the fitting function

$$\sigma = A[\ln(E/I)/EI]\{1-B\exp[-C(E/I-1)]\},$$

where E is the electron energy, I is the sum of the respective ionization potentials for the two outermost electrons, and A, B, and C are the fitting parameters.

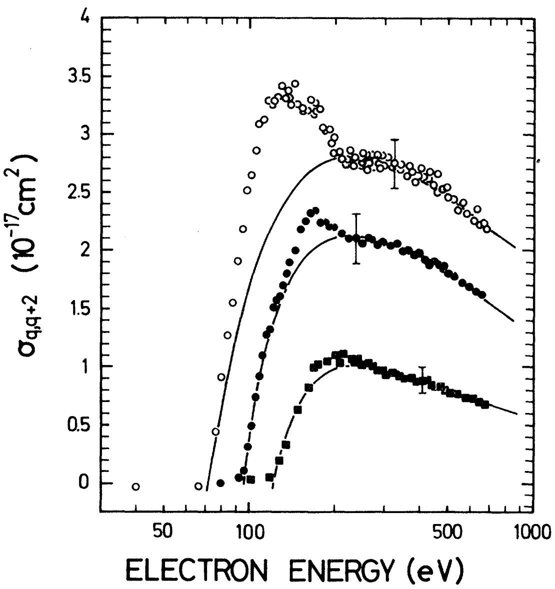

FIG. 2. Electron-impact double ionization cross sections $\sigma_{q,q+2}$ for Xe^{q+} ions: Xe^{2+}, open circles; Xe^{3+}, filled circles; Xe^{4+}, squares. Typical total absolute uncertainties are indicated. The solid lines represent the direct "double-knockout" contributions (see Fig. 1).

structures as in the case of Xe^{1+} and I^{1+} are observed. The onset of the peak is shifted with the increasing ionization energy of the $4d$ shell and its relative strength decreases with increasing ion charge state. These measurements confirm the assumption of $4d$-shell ionization-autoionization contributions to $\sigma_{q,q+2}$. A possible explanation for the observed decline of this contribution is the decreasing number of outer-shell electrons available for the subsequent autoionization process.

The thresholds of all measured cross sections $\sigma_{q,q+2}$ coincide with the minimum energy necessary to remove two electrons from the outermost $5p$ shell of the different parent ions. Thus, the cross sections can be split into two contributions: the direct ionization of two electrons from the O shell and the indirect $4d$-shell ionization-autoionization process. Since the two-step contribution is fairly localized around 100 eV we represent the direct process by a smooth fitting curve connecting the measured cross-section functions below and above the "hump." The partial cross section for the two-step process is obtained by subtracting the direct part—represented by the solid lines in Figs. 1 and 2—from the measured total cross sections $\sigma_{q,q+2}$. The resulting cross sections $\sigma_{q,q+2}^{4d}$ for the electron-impact $4d$-shell ionization-autoionization process is shown in Fig. 3 for Xe^{1+} and I^{1+} ions along with the partial photoionization cross section of the $4d$ shell of Xe atoms.

Apart from a small shift of about 14 eV which results from the difference in ionization potentials of $4d$ electrons in neutral Xe and Xe^+, I^+, respectively, an unexpected and remarkable agreement, in both absolute size and width, is revealed. In Fig. 3 we also include for comparison the cross section for ionization of a Xe^{1+}-$4d$ electron by electron impact as predicted by the Lotz formula[7] which is generally assumed to give a good representation of direct ionization. The discrepancy with what is expected and the close agreement with the photoionization data suggests

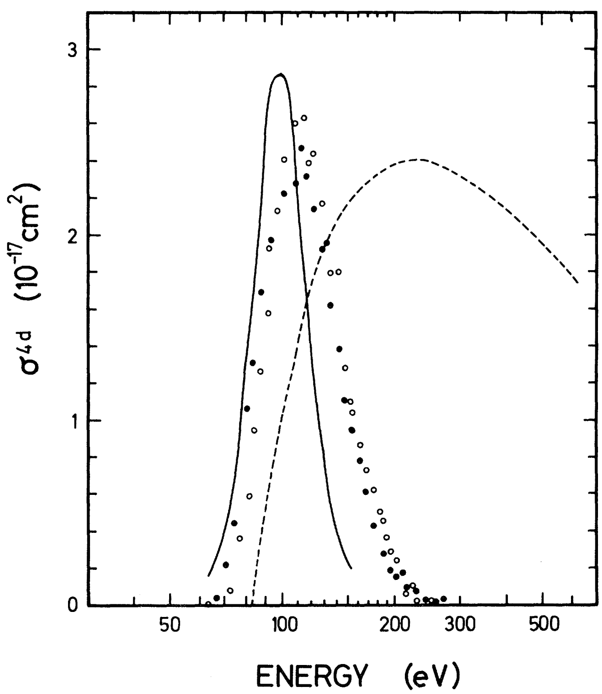

FIG. 3. Ionization of the $4d$ subshell: $4d$ photoionization cross sections for Xe atoms measured by Haensel *et al.* (Ref. 6) (solid line); electron-impact double ionization contribution of $4d$ electrons $\sigma_{1,3}^{4d}$ for Xe^{1+} (open circles) and I^{1+} (filled circles) ions obtained from Fig. 1 (see text); the dashed line is the electron-impact direct ionization cross section calculated by using the Lotz formula (Ref. 7).

that electron-impact ionization of $4d$ electrons is dominated by effects which also determine photoionization.

Distorted-wave calculations for many-electron ions have demonstrated a pronounced sensitivity of the ionization cross section to the description of the low-energy final-state electron.[8] The problem of one continuum electron outside a complex target has been extensively studied in photoionization work especially also in the case of opening a closed nd subshell.[9] Effects of collapsing wave functions, electron correlations, term dependence, and autoionization have been identified as determining the size and shape of photoionization cross sections. It appears obvious that atomic-structure effects also can dramatically change the expected cross sections for electron-impact ionization. Calculations must be done beyond the simplest average field (e.g., Herman-Skillman) model to predict whether the collision strength out of the $4d$ shell goes into autoionizing states (leading to single ionization) or into the continuum (leading to double ionization). Suitable theories are term-dependent Hartree-Fock or the random-phase approximation including exchange. Pindzola, Griffin, and Bottcher[10] have calculated excitation-autoionization in the cadmium isoelectronic sequence which exhibits strong target term dependence. Measurements of excitation-autoionization contributions to single ionization of ions probe term dependence in autoionizing levels, while studies of the present type probe term dependence in the continuum.[11]

Our measurements show that multiple ionization of complex ions may have large cross sections and, therefore, should not be neglected in plasma-modeling calculations. Two-step processes of $4d$-shell ionization followed by Auger decay may even dominate direct "double knockout" of outer-shell electrons. We have revealed a surprising agreement of the cross sections for electron impact and photoionization of the closed $4d$ shell which clearly demonstrates the interrelations between the two processes. Theoretical techniques already well developed for photoionization should be adapted to electron ionization. In this sense we are certainly at the beginning of stimulating theoretical and experimental work on electron impact ionization of complex ions.

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