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Absolute photoionization cross sections of I^+ and I^{2+} in the $4d$ ionization region

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The absolute photoionization cross sections of I^+ and I^{2+} ions have been measured from 45 to 140 eV, covering the region of $4d$ ionization where a large maximum, also known as a ‘‘giant resonance,’’ occurs in the cross section. For both ions, the maximum cross section appears near 90 eV and is measured to be 23(3) Mb and 24(4) Mb, respectively. This is significantly larger than that previously reported for atomic iodine, and only slightly smaller than the values calculated using the random-phase approximation with exchange.

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Photoionization experiments of singly and multiply charged atomic ions are of considerable interest [1,2], motivated by fundamental and applied aspects. Information about photoionization is of importance for atomic theory to test the description of structural and dynamic effects along isonuclear or isoelectronic series, and to provide essential data for the modeling of astrophysical and plasma phenomena. Data reported for multiply charged ions have so far only been concerned with relative cross sections and identification of the many autoionizing lines resulting from excitation of inner-shell electrons [1,3,4], but absolute photoionization cross sections are important in guiding theoretical development.

This Rapid Communication reports a new step in multiply charged ion physics, providing an absolute photoionization cross-section measurement for a multiply charged ion I^{2+}. In addition, when combined with our absolute photoionization cross-section measurements for I^+, the long-standing controversy between experiment and theory concerning the oscillator strength of the $4d^{10}$ subshell in atomic iodine and its ions can be resolved. Very recently, attention was called to the significant difference, a factor of 3, between experimental data and calculations of the photoionization cross section for atomic I and its ions, I^- and I^{2+}, using the random-phase approximation with exchange over a broad energy region from 40 to 136 eV, a region that is dominated by the so-called $4d$ giant resonance. This intense resonance is related to the delayed onset of $4d \rightarrow \epsilon f$ transitions to the continuum, as a result of the centrifugal barrier that must be overcome by the escaping electron [6]. For the case of Xe, this peak was observed some time ago [7,8], and its cross section is well known [9,10].

Experimental values for the absolute photoionization cross section of atomic I are available from two independent studies. Photoemission measurements of the I $4d$ subshell in CH_3I [12] were normalized to an absolute photoabsorption measurement for CH_3I [13], and a maximum cross section of 11 Mb near 90 eV was obtained. A value of 6.5 Mb from a photoelectron spectroscopy study of laser produced I atoms has been reported [14], where the measurements were normalized to absorption data for I_2 [15]. These two experimental values clearly deviate significantly from the maximum cross section calculated very recently [5], where values close to 30 Mb for I^-, I, and I^{2+} were obtained, the latter exhibiting the smallest value of 27 Mb. The close correspondence between these calculations and earlier calculations [16,17] using an entirely different theoretical method emphasizes the significant deviation from experiment and calls for measurements of the cross section for the ions I^+ and I^{2+}, since reliable absolute photoionization cross sections can be obtained for these ions, in contrast to neutral iodine.

Relatively few absolute cross-section data exist for neutral atoms, due to difficulties in determining the target density. Apart from the noble gases, for which an absorption cell can be used, since the pressure can be measured sufficiently accurately, and isolated cases where heat pipes can be applied for metal vapors [18–20], the absolute cross sections for other atoms are inferred from various normalization procedures. Sum rules have been applied in the case of vapor discharge measurements [21], and calibration against a photoline of known cross section has been applied in an electron spectroscopy experiment [22], but such methods are prone to systematic errors. For ions, the target can be provided in the form of a beam and therefore the target density can be measured directly. Consequently, the absolute photoionization cross sections for ions are much more reliable. The main limitation is the rather low target-ion density, at least six orders of magnitude lower than that for neutral species. Thus much higher photon flux is needed, combined with a long interaction path length.

The experimental setup has been described in some detail before [23,24], so only brief details are given here. The experiments were performed using the merged-beam apparatus

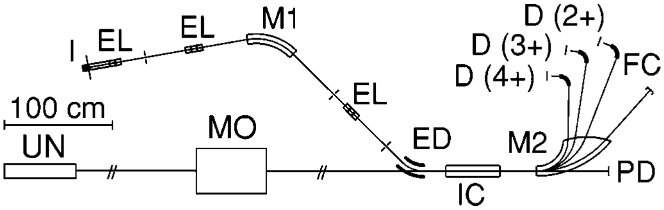

FIG. 1. Schematic drawing of the experimental setup. I, ion source; EI, Einzel lenses; M1 and M2, deflection magnets; ED, electrostatic deflector; IC, interaction chamber; D, detectors for 2+(3+), 3+(4+), and 4+ ions from photoionization of 1+(2+) ions; FC, Faraday cup for 1+(2+) ions; UN, undulator (not in scale); MC, Miyake monochromator (not in scale); PD, calibrated Al photodiode.

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shown in Fig. 1. A low-energy (2–4 keV) collimated ion beam of I^+ or I^{2+} ions was merged with a monochromatized photon beam from an undulator on the University of Aarhus storage ring ASTRID over a distance of about 50 cm. After the interaction region, the photoionization yields, the primary beam and the photon beam, were separated by a magnet; they were subsequently measured by particle detectors, a Faraday cup, and a calibrated photodiode, respectively. The I^+ and I^{2+} target ions were produced in a plasma-type ion source, using gaseous C_2H_5I; only their ground-state configurations, $4d^{10}5s^25p^4$ or $4d^{10}5s^25p^3$, respectively, both possessing three terms, are expected to have been populated significantly. In the case of the I^+ target beam, the yields of I^{2+}, I^{3+}, and I^{4+} ions were recorded, whereas I^{3+} and I^{4+} ions were recorded in the case of the I^{2+} target beam (corresponding to single, double and triple photoionization and single and double ionization, respectively). The absolute photoionization cross sections were determined from the count rates of the various product ion detectors, the current and velocity of the primary beam, the photodiode current, the interaction length and the effective beam size, together with the detector and photodiode efficiencies. The photodiode efficiency was obtained with a noble-gas ionization chamber containing 10–60 mtorr Ne (the data were extrapolated to zero pressure) and using Ne photoabsorption cross section [11]. The two-dimensional ion- and photon-beam profiles were determined by five sets of beam scanners. Below 85 eV, the photon beam was in general filtered for higher-order radiation by Al (45–70 eV) and Si (70–85 eV) foils, the only exception being the $h\nu + I^{2+}$ photoionization cross-section data between 50 and 85 eV. They were measured without foil and therefore are slightly influenced by second-order radiation; the resulting additional error due to this is expected to be less than 10%. Above 85 eV, the flux of higher-order radiation was insignificant.

The data were recorded with an accumulation time of 10–30 s/channel, a photon flux of $\sim 10^{13}$ photons/s, and an ion current of 120 nA for I^+ and 25 nA for I^{2+}. The photon-energy resolution was about 100 meV at 45 eV and increased with the photon energy, approximately as $E^{5/2}$, thus becoming 400 meV at 80 eV and 1.0 eV at 115 eV. The photon-energy scale was calibrated using autoionizing resonances in He and Kr, which were observed in a noble-gas ionization chamber; the required resonance energies were obtained from Refs. [25,26]. The background signal from extraneous processes was generally much smaller than the signal due to photoionization in the double-ionization channel, but the two signals were comparable in the single-ionization channel. The systematic uncertainty in the absolute cross-section measurements is estimated to be 15%, both for the continuum and for the resonance peaks.

Figure 2 shows the partial photoionization cross sections for I^+ ions leading to loss of one, two, or three electrons together with the total photoionization cross section in the energy interval 45 to 140 eV. The spectrum is dominated by the large $4d$-ionization peak. Production of I^{3+} ions is the main contributor to the total cross section, accounting for more than 90% of the total cross section, with a minor contribution from one-electron emission and hardly any three-electron emission. In the latter case, the appearance potential is $\sim$90 eV, whereas the additional increase at $\sim$130 eV can be attributed to $4p^{-1}$ transitions. The total cross section reaches its maximum near 90 eV and has a value of 23(3) Mb. The resonance structure seen near 47 eV is due to the excitation of $4d$ electrons to $5p$ orbitals, followed by excitation to $6p$ orbitals just above 60 eV [27].

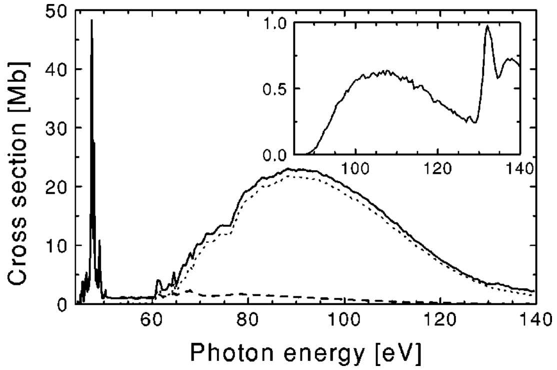

FIG. 2. Absolute cross section for photoionization of I^+ ions, leading to formation of I^{2+} (dashed line), I^{3+} (dotted line), and I^{4+} (insert, solid line) ions. In addition, the sum of the three cross sections is given (solid line).

Figure 3 shows the partial photoionization cross sections for I^{2+} leading to the formation of I^{3+} and I^{4+} together with

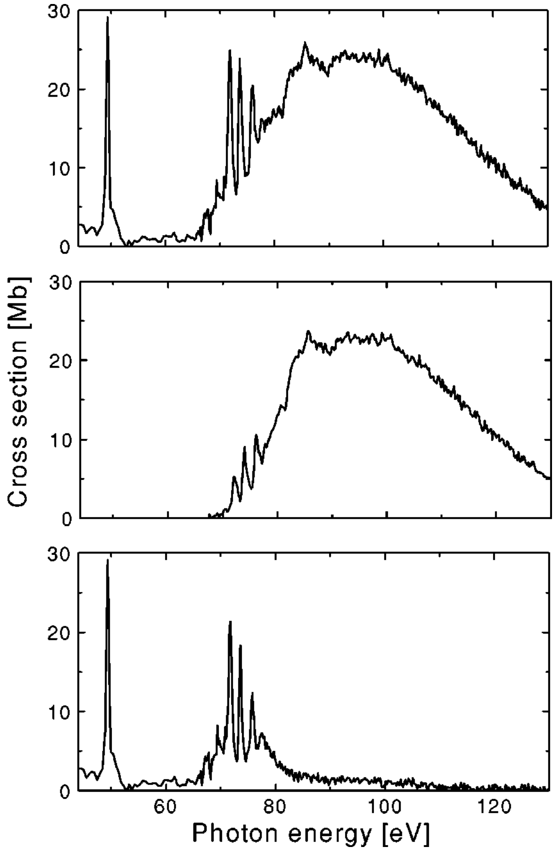

FIG. 3. Absolute cross section for photoionization of I^{2+} ions leading to I^{3+} (bottom) and I^{4+} (middle) and the sum of these two cross sections (top).

the sum of these two cross sections. The dominant process in the region around the cross-section maximum is again the emission of two electrons, which accounts for more than 90% of the total cross section. The maximum value appears at nearly the same energy for I^{2+} as for I^+ beams, and the cross section is also almost the same, determined to be 24(4) Mb. The photoionization spectra of I^{2+} ions exhibit a much more pronounced resonance structure in the 70–80-eV region than that observed for I^+. This structure, most noticeable in the I^{3+} product channel (loss of one electron), is due to $4d \rightarrow nf$ transitions, a process which becomes of increasing importance with the increasing charge state of the projectile ion. The change in the charge on the ion influences the effect of the centrifugal barrier, moving oscillator strength from the ϵf continuum into the discrete nf spectrum; this effect is well known and was first seen in the Ba, Ba^+, Ba^{2+} sequence [28]. In our experiment, we can see that the $4d \rightarrow nf$ transitions mainly result in the production of I^{3+} ions, especially for the lower n values, whereas ionization into the $4d^{-1}\epsilon f$ continuum primarily leads to production of I^{4+} via Auger decay. We note that our observations are also in good agreement with similar observations recently reported for multiply charged Xe ions [1]. The sharp structure in the double-photoionization cross-section data between 85 and 90 eV is expected to be artificial and is probably related to the energy dependence in the photodiode efficiency.

The absolute photoionization cross sections for I^+ and I^{2+} are the same within the experimental error of 15% for the $4d$ giant-resonance structure, as one would expect from the recent theoretical study [5]. The experimental cross sections reported here clearly deviate from those previously reported, and we attribute this to the normalization procedures used in the previous works. The measured cross section for I^{2+} appears to be slightly smaller than predicted from the random-phase approximation calculations [5]. This might be due to the neglect of relaxation effects. Calculations for photoionization of the I^- ion [29] have shown that taking relaxation effects into consideration reduced the calculated cross section by approximately 30%. Relaxation effects for I^+ and I^{2+} ions are calculated [30] to reduce random-phase approximation values by 10–15 %, leading to a very good agreement between theory and experiment.

In order to check our data, we have performed a similar photoionization cross-section determination for He^+ in the region of 54.4–140 eV. Because this cross section can be calculated very accurately (see, e.g., [31]), the measurement can be used to examine the accuracy of our experimental method. The obtained He^+ cross-section data differ from the theoretical ones by less than 10%, thus giving confidence in the cross sections we have measured for the iodine ions.

The present investigation has demonstrated the value of absolute photoionization cross-section measurements that do not have to rely on any normalization procedure. By changing the ion source used it will be possible to extend the present measurements to still higher charge states, with direct relevance to a number of astrophysically important issues.

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