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Observation of resonant-excitation double autoionization in electron-I^{50+} collisions

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We present a clear observation of a resonant contribution to electron impact ionization of Li-like highly charged ions (I^{50+}). Li-like ions are the simplest in which resonant-excitation double autoionization (REDA) can occur. REDA was observed by measuring the ratio of the numbers of trapped ions of two neighbring charge states as a function of electron energy under equilibrium conditions using an electron beam ion trap. The resonant strength was determined by normalizing the experimental data to theoretical ionization cross sections at the nonresonant interaction energies. Good agreement was found between the observed and calculated resonant strengths for this observation of REDA in Li-like highly charged ions.

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Electron impact ionization (EI) is the most important primary process in plasmas, and necessarily its cross section is of overriding importance to model fusion, astrophysical, and other high-temperature plasmas. Although EI cross sections have been measured for various elements and various incoming electron energies, the measurements for highly charged ions (HCIs) have been performed only quite recently [1–5]. In particular, there are only a few reported observations of indirect processes such as excitation autoionization (EA), arising from direct inner-shell excitations contributing to EI processes, and resonant processes such as resonant-excitation double autoionization (REDA), involving dielectronic capture of an incident electron that is then followed by successive autoionization processes from the intermediate highly excited state [6]. The first observation of EA was made by Crandall *et al.* [7] for Li-like C, N, and O. They measured the EI cross sections for these ions as a function of electron energy and found steplike structures at threshold energies of K-shell excitations. These steps were designated as being due to EA. The same regions were studied by Hofmann *et al.* [8] in detail with higher resolution and higher counting statistics. On the steplike EA structures, they found many clear resonant structures that were identified as REDA.

The highest charge state for which EA and REDA were observed so far is Na-like Fe^{15+} [9] measured with a heavy ion storage ring for an interaction energy of less than 1 keV. After this measurement, no experimental result of REDA for higher charge state has been reported. To date, no experimental investigation has been reported for few-electron heavy ions, such as Li-like ions with $Z>10$ in spite of the simplicity of the system, whereas several theoretical calculations have been reported for, e.g., Li-like Xe^{51+} [10–12]. Several reasons can be given to account for this situation. First, for few-electron HCIs, since the fluorescence yield of doubly excited resonance states increases rapidly with Z, the branching ratio for REDA becomes very small with the dielectronic recombination (DR) branch becoming dominant. In addition, the dielectronic resonant capture cross section also decreases as the resonant energy (the energy difference between the initial state of the target ion and the capturing intermediate excited state) increases, so that the REDA cross section becomes consequently very small. Measurement of REDA is clearly very challenging for few-electron high-Z ions.

In addition to these physical reasons, there are also technical reasons why the crossed beam technique is practically impossible for HCIs with high Z and q. Also interaction energies higher than 1 keV are difficult to achieve with the present generation of storage rings. An experimental tool particularly suited to the measurement of EI cross sections for few-electron heavy HCIs is an electron beam ion trap (EBIT). The EI cross sections can be obtained by measuring the charge-state distribution of trapped HCIs interacting with the electron beam of an EBIT. For example, Marrs *et al.* [2,13] measured the EI cross sections for H-like heavy ions up to U^{91+} by measuring the ratio of the numbers of H-like and bare ions trapped through the observation of radiative recombination (RR) x rays for those ions at equilibrium. After correction for charge exchange, this ratio is proportional to the ratio of the RR cross section of bare ions to the EI cross section of H-like ions. Hence, the EI cross section can be obtained from a measurement of this ratio by normalizing it to a reliable theoretical RR cross section. However, this method cannot be easily applied to HCIs other than H-like ions because RR x rays cannot easily be resolved for lower charge states with usual solid-state detectors. In fact, Stöhlker *et al.* [14] did succeed in using a variant of the method used by Marrs *et al.* However, several recombination lines overlapped in the spectrum, so the sensitivity would not be sufficient to observe REDA. The absorber technique they additionally used to overcome this resolution limitation does not make a measurement at a fixed electron beam energy and so it also would not be suitable. Thus the method used by Marrs *et al.* or variants upon it do not seem suitable for measurements of REDA in Li-like ions.

In the present study, which has been carried out with the Tokyo-EBIT [15], we employed another method to observe the REDA process for Li-like I^{50+}. In this method, the ratio between numbers of trapped He-like I^{51+} and Li-like I^{50+}

ions, $n_{\mathrm{He}}/n_{\mathrm{Li}}$ (i.e., the ion number ratio), was measured at equilibrium by monitoring the extracted ions. Here n_{He} and n_{Li} are the number of He-like I^{51+} and Li-like I^{50+}, respectively. In an EBIT, an electron beam emitted from a cathode is accelerated toward an ion trap while being magnetically compressed by a superconducting magnet. The ion trap consists of three successive drift tubes (DTs), in which positive ions can be trapped axially by applying a positive bias to the two outer DTs and radially by the space-charge potential of the electron beam. The electron beam also successively ionizes the trapped ions, creating HCIs in the center of the trap. Iodine was injected into the trap in the form of a low density of $\mathrm{CH_3I}$, which was quickly ionized and fragmented by electron impact, the carbon and hydrogen ions acting as coolants for those of iodine.

In the present experiments, the REDA process represented below was studied for Li-like I^{50+},

$$e^- + \mathrm{I}^{50+}(1s^2 2s) \rightarrow \mathrm{I}^{49+}(1s2s3l3l') \rightarrow \mathrm{I}^{50+}(1s2s2l'') + e^- \rightarrow \mathrm{I}^{51+}(1s^2) + e^- + e^-. \quad (1)$$

These narrow resonance profiles appear on the slowly varying background signal of EI processes due to nonresonant EA and direct ionization.

The electron energy range studied is 29.5–32 keV. Since the electron beam is mono-energetic and the energy is below the ionization energy of He-like I^{51+}, there are no H-like and bare ions in the trap so that the rate equation for I^{51+} can be represented as

$$\frac{dn_{\mathrm{He}}}{dt} = \frac{j}{e}[\sigma_{\mathrm{Li}}^{\mathrm{ion}} n_{\mathrm{Li}} - \sigma_{\mathrm{He}}^{\mathrm{re}} n_{\mathrm{He}}] - \frac{n_{\mathrm{He}}}{\tau}, \quad (2)$$

where j the electron current density, $\sigma_{\mathrm{He}}^{\mathrm{re}}$ the recombination cross section for He-like I^{51+}, $\sigma_{\mathrm{Li}}^{\mathrm{ion}}$ the ionization cross section for Li-like I^{50+}, and τ^{-1} is a term including all other loss contributions, such as escape and charge exchange. Since the contribution from the double ionization of the Be-like ion is negligibly small, Eq. (2) includes all the important terms [16]. At equilibrium, it follows from Eq. (2) that

$$\frac{n_{\mathrm{He}}}{n_{\mathrm{Li}}} = \frac{\sigma_{\mathrm{Li}}^{\mathrm{ion}}}{e/j\tau + \sigma_{\mathrm{He}}^{\mathrm{re}}}. \quad (3)$$

Through this equation, ionization and recombination processes can be studied by measuring the ion number ratio [13].

The ion number ratio can be obtained by measuring the intensity of ions extracted from the EBIT [17,18]. The trapped ions are heated through successive long-range electron impacts so that finally they can escape from the trap. The ions which escape axially toward the electron collector side can be extracted into an HCI beam line [19]. In the present study, the efficiency of extraction and transmission in the beam line were assumed to be practically the same between adjacent charge states. Crespo Lopez-Urrutia *et al.* [20] measured the ion intensity ratio between extracted He-like and Li-like Kr, and found that $\alpha_{\mathrm{Li}}/\alpha_{\mathrm{He}}=0.90\pm0.15$ (α denotes the efficiency of extraction and transmission). Their measurement may support our assumption that the extracted ion intensity ratio gives the ion number ratio inside the trap.

The extracted He-like I^{51+} and Li-like I^{50+} were detected simultaneously with a position-sensitive detector placed just after the charge-analyzing magnet in the HCI beam line. Since natural iodine has only one isotope ($^{127}\mathrm{I}$), its charge states were clearly separated on the position-sensitive detector. The detection efficiency is considered to be practically the same for the He-like and Li-like ions. In order to examine the detection efficiency across the detector area, the measurement was repeated while using different positions on the detector, and no significant difference was observed.

The electron energy interacting with HCIs is determined by the potential difference between the electron gun and the central DT. The electron energy was scanned from 29.5 to 32 keV in steps of ~4 eV by controlling the voltage at the electron gun. For each step, counting of the ions was started 2 s after the electron energy was changed to ensure the charge equilibrium condition had been established, and continued for 8 s. It was confirmed that the charge-state distribution did not change during the acquisition period. The voltage of the central drift tube was fixed to +3 kV throughout the experiment while the final DT was set a further 50 V above this value, fixing the axial trapping voltage for the ions. The electron beam current was 55 mA, and the magnetic field at the trap was 4 T.

The present experimental procedure is similar to that used in the measurements of DR processes of He-like Ar by Ali *et al.* [18]. The present measurement, however, has been performed aiming at the observation of REDA, so that the following two points were mainly different from their experiment. (i) Heavy element (iodine) was selected as target because the resonant energy of REDA is well separated from that of DR. (ii) The leaky mode extraction [19] was used to obtain stable operational condition and high counting statistics, which is important to observe small contribution on the large background (arising from nonresonant ionization and recombination processes).

Figure 1 shows the intensity ratio of extracted He-like I^{51+} and Li-like I^{50+} as a function of electron energy. As seen in the figure, both positive and negative peaks exist on the slowly varying background. By extending Eq. (3) to include REDA and DR contributions, the ion number ratio can be expressed by the following formula in general:

$$\frac{n_{\mathrm{He}}}{n_{\mathrm{Li}}} = \frac{\sigma_{\mathrm{Li}}^{\mathrm{ion}} + \sigma_{\mathrm{Li}}^{\mathrm{REDA}}}{e/j\tau + \sigma_{\mathrm{He}}^{\mathrm{RR}} + \sigma_{\mathrm{He}}^{\mathrm{DR}}}, \quad (4)$$

where $\sigma_{\mathrm{Li}}^{\mathrm{ion}}$ is the (nonresonant) ionization cross section of the Li-like ion including direct and indirect (EA) processes, $\sigma_{\mathrm{Li}}^{\mathrm{REDA}}$ the REDA cross section, and $\sigma_{\mathrm{He}}^{\mathrm{RR}}$ and $\sigma_{\mathrm{He}}^{\mathrm{DR}}$ are the RR and DR cross section for the He-like ion, respectively. As is clear from the Eq. (4), DR processes for the He-like ion should affect the ratio $n_{\mathrm{He}}/n_{\mathrm{Li}}$. Actually, the negative peaks in Fig. 1 correspond to the KMM DR for He-like I^{51+}. In contrast, the positive peak is due to the REDA for Li-like I^{50+}. Another possible mechanism that can make a positive "peak" is the Fano line profile arising from the interference between RR and DR [21,22]. However, since the natural width of the KMM resonance is estimated to be less than 1 eV, it is practically impossible to observe because the

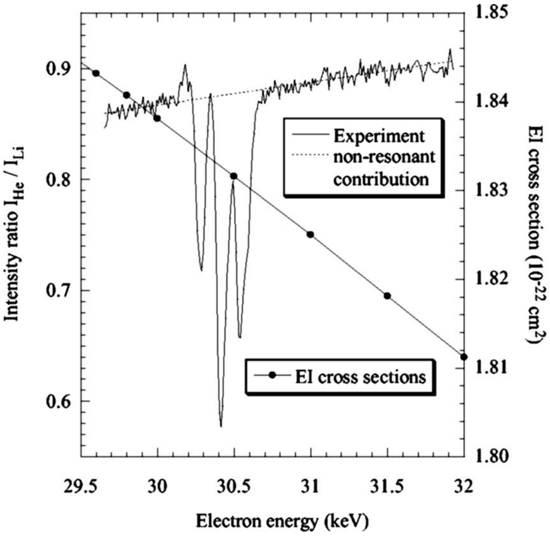

FIG. 1. (Solid line) Intensity ratio between He-like I^{51+} and Li-like I^{50+} extracted from the Tokyo-EBIT as a function of electron energy. The electron energy was calibrated with the DR resonant energy calculated by using the HULLAC code. (Dashed line) Nonresonant contribution to the intensity ratio obtained by fitting the experimental data for the off-resonance region to a second-order polynomial function. (Closed circles) Theoretical EI cross sections for Li-like I^{50+} used for obtaining the resonant strength of REDA (see text), in which the EA contributions were included but the REDA contributions were not included.

present electron energy resolution is about 50 eV. Therefore, the positive peak in the present measurement is unambiguous evidence of REDA, a process that has never been observed previously for such a heavy HCI.

Away from any resonances the ion number ratio can be expressed as

$$\left(\frac{n_{\mathrm{He}}}{n_{\mathrm{Li}}}\right)_{\mathrm{NR}} = \frac{\sigma_{\mathrm{Li}}^{\mathrm{ion}}}{e/j\tau + \sigma_{\mathrm{He}}^{\mathrm{RR}}}. \tag{5}$$

This contribution can be obtained by fitting the experimental data for the off-resonance region. The result of the fitting to a second order polynomial function is shown by the dotted line in Fig. 1. By using $(n_{\mathrm{He}}/n_{\mathrm{Li}})_{\mathrm{NR}}$, the Eq. (4) can be modified to

$$\sigma_{\mathrm{Li}}^{\mathrm{REDA}} = \sigma_{\mathrm{Li}}^{\mathrm{ion}}\left[\frac{(n_{\mathrm{He}}/n_{\mathrm{Li}})}{(n_{\mathrm{He}}/n_{\mathrm{Li}})_{\mathrm{NR}}} - 1\right], \tag{6}$$

outside of the He-like DR resonance region. Thus, making use of theoretical values for $\sigma_{\mathrm{Li}}^{\mathrm{ion}}$, the REDA cross section $\sigma_{\mathrm{Li}}^{\mathrm{REDA}}$ can be derived from the measurement. In the present study, the HULLAC code [23] was used to calculate $\sigma_{\mathrm{Li}}^{\mathrm{ion}}$, which is shown in Fig. 1. In the calculation, EA was taken into account as well as direct ionization. In the HULLAC code, the distorted wave approximation is used to calculate the ionization cross sections. Based on previous comparisons between calculated and measured direct electron impact ionization cross sections in few-electron highly charged ions [5,24,25], the uncertainty in the distorted wave calculation is considered to be much smaller than the present statistical uncertainty, which is about 12% as described in Table I.

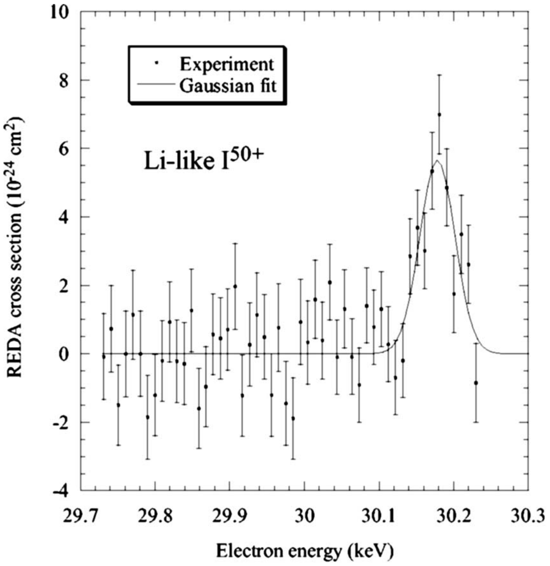

FIG. 2. REDA cross section obtained from the present experiment. The solid line is the Gaussian function obtained by the least-squares fitting to the experimental data.

Figure 2 shows the cross section for the REDA contribution at $E_e \simeq 30.2$ keV. It is noted that, according to the theoretical estimation, there is no strong KMM DR resonance for He-like I^{51+} in this region. The solid line in Fig. 2 is the Gaussian function obtained by least-squares fitting to the experimental data. According to Badnell and Pindzola [12], interfering effects between REDA and EA are generally small for highly charged ions. Thus, the resonant strength of the REDA process can be obtained by integrating the area under this peak. The result is listed in Table I as well as the theoretical values obtained by using the HULLAC code. In the calculation of branching ratios, $M2$ transitions were included although the Breit interaction was not included. We checked the effect of the Breit interaction by using the branching ratio

TABLE I. Resonant strength for resonant-excitation double autoionization in Li-like I^{50+}. The experimental error represents only the statistical one.

Present	Theory			
Resonant strength (10^{-22} cm^2 eV)	State	Resonant energy (eV)	Resonant strength (10^{-22} cm^2 eV)	Sum (10^{-22} cm^2 eV)
3.4±0.4	$(1s2s3s^2)_{J=1}$	30188	2.61	3.27
	$(1s2s3s3p)_{J=0}$	30234	0.25	
	$(1s2s3s3p)_{J=1}$	30235	0.41	

of $(1s2s2p_{3/2})_{J=5/2}$ estimated by interpolating those for other Z calculated by Chen and Reed [10], and found that the effect on the resonant strength is negligibly small for the states listed in the table. The experimental error listed in the table corresponds to the error of the least-squares fitting weighted by the statistical uncertainties. Since the electron energy resolution of the present study is not so high, several resonances can be superimposed. Further details about the resonant fine structure can be expected in higher-resolution measurements which will be performed in the near future by decreasing the electron beam current, although a longer accumulation time will be required to get sufficient statistics. The measurements will be extended also for HCIs with higher Z, for which the separation between the resonant states becomes larger so that they will be clearly resolved even with high electron beam current.

As well as the most preferable resonant state $(1s2s3s^2)_{J=1}$, other possible states that may contribute to the experimental peak are listed in the Table I. By considering the total contribution from these states, the agreement between the experiment and the theory is good and the measurement indicates that contributions from the $(1s2s3s3p)_{J=0}$ and $(1s2s3s3p)_{J=1}$ states should be included to account for the observed resonant strength. It should be noted that for Li-like ions, the experimental result for an individual resonant state can be compared directly to the theory as listed in the table because a few resonant states exist on the region where the EA contribution is flat. This is in contrast to the case of Na-like ions where many resonant states exist on a rapidly varying EA contribution [9], so that only superimposed results can be compared.

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