Relativistic effects on resonant interactions between electrons and highly charged ions

Nobuyuki NAKAMURA\textsuperscript{1}, Anthony P. KAVANAGH\textsuperscript{2}, Hirofumi WATANABE\textsuperscript{3}, Hiroyuki A. SAKAUE\textsuperscript{4}, Yueming LI\textsuperscript{5}, Daiji KATO\textsuperscript{4}, Fred J. CURRELL\textsuperscript{2}, and Shunsuke OHTANI\textsuperscript{1}

\textsuperscript{1} Inst. for Laser Science, The Univ. of Electro-Communications, Tokyo 182-8585, JAPAN
\textsuperscript{2} Queen’s University Belfast, Belfast BT7 1NN, United Kingdom
\textsuperscript{3} CREST, Japan Science and Technology Agency
\textsuperscript{4} National Institute for Fusion Science, Toki, Gifu 509-5292, JAPAN
\textsuperscript{5} Institute of Applied Physics and Computational Mathematics, P.O.Box 8009, Beijing 100088

E-mail: n.nakamu@ils.uec.ac.jp

Abstract. We report measurements of resonant processes in electron collisions with very highly charged heavy ions made using an electron beam ion trap. By measuring the ion abundance ratio in the trap at the equilibrium condition as a function of electron energy, we have observed resonant processes such as dielectronic recombination and resonant excitation double autoionization very clearly. Remarkable relativistic effects due to the generalized Breit interaction have been clearly shown in dielectronic recombination for highly charged heavy ions.

1. Introduction

Resonant processes in electron-ion collisions are very important because they often have very large cross sections compared to non-resonant processes and thus can significantly affect the rates of the fundamental reactions in hot plasmas. In the interactions with highly charged ions (HCIs), a free electron can be captured with a large probability and simultaneously a bound electron can be excited to form a doubly excited state resonantly. This unstable intermediate state may decay by photon emission: Dielectronic Recombination (DR), or by emission of two electrons due to successive autoionization: Resonant Excitation/ Double Autoionization (REDA). For example, for Li-like ions, those reactions are represented as follows.

\[
e^{-} + 1s^22s \rightarrow 1s2snln'l' \rightarrow \begin{cases} 1s^22s^2 + h\nu + h\nu' + \cdots (DR) \\ 1s2s2l'' + e^- \rightarrow 1s^2 + e^- + e^- (REDA). \end{cases}
\]

In this paper, we present observations of these resonant processes using the Tokyo-EBIT (electron beam ion trap) \cite{1}). An EBIT \cite{2} is suitable for studying such collision processes of HCIs with electrons because it has a mono-energetic and unidirectional electron beam interacting with trapped HCIs. In previous studies, resonant processes have usually been observed by measuring enhancement of X-ray emission due to the resonant processes. On the other hand in the present study, the resonant processes have been observed by measuring the charge abundance ratio at the equilibrium condition, which changes drastically with the resonant processes, as a...
function of electron energy. The abundance ratio was obtained by measuring the intensity of ions extracted from the EBIT. Compared to the X-ray measurements where the observation solid angle is typically of the order of $10^{-3}$, the ion measurements have much higher efficiency since the total efficiency (extraction, transport, and detection) of the order of $10^{-1}$.

In the next section the principle and procedure of the present experimental method are described. The results for DR of highly charged iodine and bismuth ions and for REDA of Li-like iodine are given in Sec. 3, which is followed by a brief summary.

2. Experimental method

The present experimental method is similar to that used by Ali et al. [3], in which they used an electron beam ion source in Kansas to study DR processes for He-like argon. In the present study, a high-energy EBIT in Tokyo [4, 1] was used to study the resonant processes of very highly charged heavy ions. The element of interest was introduced into the EBIT through a gas injector [5] or an effusion cell [6]. The injection was done continuously. Elements whose natural abundance is dominated by one single stable isotope, e.g. iodine (atomic number $Z = 53$) and bismuth ($Z = 83$) were used so that the charge state of extracted ions can be clearly resolved with an analyzing magnet.

In the trap region of the EBIT, the charge state of trapped ions increases through electron impact ionization. On the other hand, recombination of a free electron and charge exchange collision with residual gas can reduce the charge state. Thus, when the rates for ion escape from the trap and multiple charge change processes (such as double ionization, multiple charge exchange) are negligibly small, the ionization balance at the equilibrium condition is determined by the rates for ionization, recombination and charge exchange. As a result, the ion abundance ratio for ions with adjacent charge-states $q$ and $q-1$ can be expressed in terms of cross sections for relevant collision processes [7]:

\[
\frac{n_{q-1}}{n_q} = \frac{\sigma_q^{DR} + \sigma_q^{RR} + (e/j)n_0\sigma_q^{CX}\bar{v}_i}{\sigma_{q-1}^{ion}}, \tag{1}
\]

where $\sigma_q^{DR}$ and $\sigma_q^{RR}$ are the dielectronic and radiative recombination (RR) cross sections for the ion with a charge state of $q$, $\sigma_{q-1}^{ion}$ the electron impact ionization cross section for the ion with $q-1$, $\sigma_q^{CX}$ the charge exchange cross section for the collision with a residual gas molecule or atom whose density is $n_0$, $\bar{v}_i$ the mean velocity of the ions, $e$ the unit charge, and $j$ the electron current density.

When there is no resonant process, i.e. when $\sigma_q^{DR} = 0$, the ratio $n_{q-1}/n_q$ varies slowly with electron energy. However, when the electron energy coincides (within a beam energy width of about 50 eV) with a DR resonance, the abundance ratio changes drastically, by an amount $\sigma_q^{DR}/\sigma_{q-1}^{ion}$. Thus in the electron energy dependence of the ion abundance ratio $n_{q-1}/n_q$, sharp structures due to DR processes for the ion with charge $q$ should appear on a smooth background.

It has to be noted that the assumption used to derive Eq.(1) is not always valid. For example, the rate for ion escape is generally small but not negligible for some cases. In addition, for very highly charged ions, cross sections for multiple charge exchange become comparable to that for single charge exchange. By taking the effects of ion escape and multiple charge exchange into account, Eq.(1) is modified as follows,

\[
\frac{n_{q-1}}{n_q} = \frac{\sigma_q^{DR} + \sigma_q^{RR} + (e/j)\sum_{i=q}^{q_{max}}(n_i/n_q)(n_0\sigma_i^{CX(i-q+1)}\bar{v}_i + e_i)}{\sigma_{q-1}^{ion}}, \tag{2}
\]

where $\sigma_i^{CX(i)}$ is the total cross section for more than $i-1$ electron capture from neutrals, $\epsilon$ the ion escape rate, and $q_{max}$ the maximum charge state in the trap. Through the third term
in Eq.(2), additional structures can appear on the background because the factor \( \frac{n_i}{n_q} \) can rapidly change with energy through DR for the ion with a charge state of \( i \), which is not of interest. For example, at the DR resonant energy for the charge state \( i \), the population of the ion with the charge state \( i \) decreases and thus \( \frac{n_i}{n_q} \) also decreases. As a result, the smooth background may show dips due to the strong population variation of higher charge state ions. This additional dip structure does not appear for the highest charge state in the trap and becomes larger for lower charge state ions because, as seen in Eq.(2), the effect is summed for the charge state \( q \) to \( q_{\text{max}} \). This additional dip structure can be corrected by the method discribed in Ref. [7]; however, no correction was needed for the data presented in this paper because it was confirmed that the effect was negligibly small.

Not only the resonant process in recombination, i.e. DR, but also that in ionization, i.e. REDA, can also make a sharp structure on the smooth background. By taking REDA into account, Eq.(1) is modified as follows,

\[
\frac{n_{q-1}}{n_q} = \frac{\sigma_q^{DR} + \sigma_q^{RR} + (e/j)n_0\sigma_q^{CX} \bar{v}_i}{\sigma_{q-1}^{\text{ion}} + \sigma_{q-1}^{REDA}},
\]

where \( \sigma_{q-1}^{REDA} \) the REDA cross section whereas \( \sigma_{q-1}^{\text{ion}} \) is the non-resonant contribution to the ionization cross section, i.e. direct ionization and excitation autoionization. Here the term arising from multiple charge exchange and ion escape is excluded since we discuss REDA only for the highest charge state in the trap in the present paper. As understood from Eq.(3), REDA can make a dip on the smooth background while DR can make a peak. Since the resonance energy of REDA for the ion with \( q - 1 \) and that of DR for the ion with \( q \) are different, the dip and the peak can be resolved if the electron energy resolution is smaller than the energy separation between those resonances.

In the present study, the ion abundance ratio was obtained by measuring the intensity of ions extracted from the EBIT. Figure 1 shows the diagram for the present experimental setup and procedure. The ions escaping from the trap were extracted into an HCI beam line [8], and detected after the charge separation with an analysing magnet. Several charge states were detected at the same time by using a position sensitive detector. The intensity ratio of the extracted ions would be different from the ion abundance ratio in the trap. However, we assumed that the total efficiency (extraction, transport, and detection) should be practically the same for
the ions with adjacent charge states. Actually we compared the charge abundance obtained from x-ray observation of trapped ions to that obtained from the observed ion intensity, and found that there was no significant difference between them [7]. Thus, the intensity ratio between adjacent charge states is considered to give the ion density ratio inside the trap.

In order to measure the ratio as a function of electron energy, the voltage at the electron gun was scanned in a stepwise fashion. For each step, ion counting was started 2 sec after the electron energy was changed to ensure that the equilibrium condition had been established, and continued for 8 sec. The voltage of the ion trap region was fixed (typically +3 kV) throughout the experiment. Both the ion signal and the electron-energy value when the ion signal was detected were recorded to a PC in list mode.

Figure 2 shows the typical charge state spectrum obtained for bismuth. As seen in the figure, ions with very high charge state can be efficiently extracted from the Tokyo-EBIT, which enables us to study resonant processes for very highly charged heavy ions.

3. Results

3.1. resonance in recombination: dielectronic recombination

Figure 3 shows the intensity ratio of extracted Li-like to He-like iodine obtained as a function of electron beam energy. A background has been subtracted, corresponding to \(\left(\sigma_{q}^{RR} + \left(e/j\right)n_{0}\sigma_{q}^{CX} \tilde{v}_{1}\right)/\sigma_{q}^{ ion} \) in Eq. (1). This background was obtained by fitting a second-order polynomial function to the non-resonance region. Thus the structures in the figure correspond to \(\sigma_{q}^{ DR}/\sigma_{q}^{ ion} \), the DR cross section for He-like iodine scaled with the ionization cross section of Li-like iodine. As seen in the figure, the \(K\eta'\) DR series has been clearly observed.

The present method is useful for a wide range of \(Z\) and for various charge states including both open- and closed shell systems. For example, Fig. 4 (a) shows the intensity ratio of extracted Be-like to Li-like bismuth as a function of electron beam energy. Again a smooth background was subtracted. Similarly to the iodine data (Fig. 3), the structures in Fig. 4 (a) correspond to the DR cross section for Li-like bismuth scaled with the ionization cross section of Be-like.

**Figure 3.** Intensity ratio of Li-like to He-like iodine extracted from the EBIT. Structures correspond to the \(K\eta'\) DR resonances of He-like iodine. Inset is a closeup view for the \(KLL\) DR resonance region.

**Figure 4.** (a) Intensity ratio of Be-like to Li-like bismuth extracted from the EBIT. Structures correspond to the \(KLL\) DR resonances of Li-like bismuth. (b) Theoretical DR cross section calculated with inclusion of the GBI, and (c) without the GBI.
bismuth, which is considered to be almost constant in this narrow electron energy range. On the other hand, Fig. 4 (b) and (c) show the theoretical DR cross sections; the former was obtained by the fully relativistic calculation including the generalized Breit interaction (GBI) while the latter was obtained by the fully relativistic calculation but without including the GBI. It is found that the peak at the lowest energy is strongly affected by the GBI and that the calculated cross section without the GBI fails to reproduce the experimental result whereas that with the GBI reproduces the experiment well. The present result clearly shows the importance of the GBI, which is the lowest order of the quantum electrodynamics effects in electron - ion collisions, in DR processes of highly charged heavy ions.

The theoretical cross section with the GBI still differs from the experiment. For example, the higher energy side of the $KL_{12}L_3$ resonances seems to have a tail for the experimental data whereas the theoretical result seems to have no tail. This is considered to be due to the interference between RR and DR. Their interference is generally weak for light and mid-$Z$ ions because the cross section of RR is much smaller than that of DR. However, for highly charged heavy ions, since the RR cross section, which increases rapidly with $Z$, becomes comparable to the DR cross section, which decreases as $Z$ increases, the interference can be so significant that the resonant feature shows the Fano-type profile [9]. However since several peaks overlap with each other, the Fano profile can not be recognised clearly in Fig. 4 (a).

Fig. 5 shows the intensity ratio of extracted B-like to Be-like bismuth, which corresponds to the DR cross section for Be-like bismuth scaled with the ionization cross section of B-like bismuth. As seen in the figure, Fano-type asymmetric profiles are clearly confirmed. In the figure, the preliminary results of fitting to Fano profiles are also shown. Interference between RR and DR was also observed through X-ray measurements for Hg ($Z = 80$) [10], Bi ($Z = 83$) [11], and U ($Z = 92$) [12]. Compared to those X-ray measurements, the present method has the advantage that the contribution from different charge states can be completely removed and that higher

---

Figure 5. Clear asymmetric profiles appeared in the intensity ratio between B-like and Be-like bismuth. Fano profiles fitted to the data are also shown with inclusion of the experimental width.

Figure 6. (a) Intensity ratio of He-like to Li-like iodine as a function of electron beam energy. Small positive peaks at around 30.2 and 30.35 keV correspond to the contribution from resonant excitation double autoionization. (b) Theoretical result obtained from the calculated cross sections for direct and resonant ionization of the Li-like ion, dielectronic recombination of the He-like ion, and the background fitted to the experimental data in the non-resonant regions.
statistical quality data can be obtained because the present method has much higher efficiency. The X-ray measurements are usually done at an observation angle of 90 degree with respect to the electron beam so that it gives differential cross sections while the present method gives the integrated cross sections. The comparison between the X-ray and extracted ion measurements is thus important to investigate the dependence of the degree of interference on the angle of photon emission. This effect will be further investigated and published elsewhere.

3.2. resonance in ionization: resonant excitation double autoionization

Figure 6 (a) shows the intensity ratio of extracted He-like to Li-like iodine for the KMM resonance region. A smooth background was subtracted. It should be noted that the ratio of higher charge state with respect to the lower charge state, i.e. the reciprocal of Eq.( 3), is plotted in order to emphasize the ionization here; thus DR should make a dip while REDA should make a peak in this plot. Accordingly, the peaks at around 30.2 and 30.35 keV are considered to be the contributions from REDA of Li-like iodine, whereas the dip structures are the DR resonances of He-like iodine. The present experiment is similar to our previous study [13], but the present data was obtained with a higher resolution by reducing the electron beam current. The resolution, which was determined by the electron beam energy width, was about 27 eV for the present study while it was about 50 eV for the previous study. Owing to the higher resolution, we could confirm the peak around 30.35 keV, which had been hidden in the tails of the DR structures on both sides in our previous study. REDA cross sections are generally small for highly charged heavy ions due to the large fluorescence yield of the resonant states. However the high efficiency of the present method enabled us to observe it very clearly.

Figure 6 (b) shows the theoretical results obtained from the calculated values for $\sigma^{REDA}_{Li}$, $\sigma^{ion}_{Li}$, and $\sigma^{REDA}_{He}$. The background corresponding to $\sigma_{RR}^q + (e/j)n_{0}\sigma_{CX}^q v_i$ in Eq.( 3) was estimated by comparison with the experiment and subtracted. As seen in the figure, overall agreement is found between the experiment and the calculation.

4. Summary

By measuring the charge abundance ratio at the equilibrium condition in an electron beam ion trap, we have observed resonant processes such as dielectronic recombination (DR) and resonant excitation double autoionization (REDA). The present method has been confirmed to have much higher efficiency compared to previous X-ray measurements. Accordingly we have successfully observed:-

* charge-state resolved DR resonant features,
* clear Fano profiles arising from the interference between DR and radiative recombination,
* clear evidence for the strong effect of the generalized Breit interaction on DR resonances,
* resonant processes in ionization processes (resonant excitation double autoionization).

The present method in combination with the injection method using an effusion cell has been proven to be very useful to study resonant processes of high charge states of various elements.

5. References

[1] N. Nakamura, J. Asada, F. J. Currell, T. Fukami, K. Motohashi, T. Nagata, E. Nojikawa, S. Ohtani, K. Okazaki, M. Sakurai, H. Shiraishi, S. Tsurubuchi, and H. Watanabe, Phys. Scr. T73, 362 (1997).
[2] R. E. Marrs, M. A. Levine, D. A. Knapp, and J. R. Henderson, Phys. Rev. Lett. 60, 1715 (1988).
[3] R. Ali, C. P. Bhalla, C. L. Cocke, M. Schulz, and M. Stockli, Phys. Rev. A 44, 223 (1991).
[4] F. J. Currell, J. Asada, K. Ishii, A. Minoh, K. Motohashi, N. Nakamura, K. Nishizawa, S. Ohtani, K. Okazaki, M. Sakurai, H. Shiraishi, S. Tsurubuchi, and H. Watanabe, J. Phys. Soc. Jpn. 65, 3186 (1996).
[5] N. Nakamura, T. Kinugawa, H. Shimizu, H. Watanabe, S. Ito, S. Ohtani, C. Yamada, K. Okazaki, and M. Sakurai, Rev. Sci. Instrum. 71, 684 (2000).
[6] C. Yamada, K. Nagata, N. Nakamura, S. Ohtani, S. Takahashi, T. Tobiyama, M. Tona, M. Sakurai, A. P. Kavanagh, and F. J. Currell, Rev. Sci. Instrum. 77, 066110 (2006).
[7] H. Watanabe, H. Tobiyama, A. P. Kavanagh, Y. M. Li, N. Nakamura, H. A. Sakaue, F. J. Currell, and S. Ohtani, Phys. Rev. A 75, 012702 (2007).
[8] N. Nakamura, F. J. Currell, D. Kato, A. P. Kavanagh, Y. M. Li, S. Ohtani, H. A. Sakaue, M. Sakurai, J. Sun, S. Takahashi, M. Tona, H. Watanabe, C. Yamada, and N. Yoshiyasu, Can. J. Phys. in press (2007).
[9] U. Fano, Phys. Rev. 124, 1866 (1961).
[10] A. J. G. Martínez, J. R. C. López-Urrutia, J. Braun, G. Brenner, H. Bruhns, A. Lapierre, V. Mironov, R. S. Orts, H. Tawara, M. Trinczek, J. Ullrich, and J. H. Scofield, Phys. Rev. Lett. 94, 203201 (2005).
[11] H. Tobiyama, H. Nohara, A. P. Kavanagh, N. Nakamura, H. Watanabe, H. A. Sakaue, Y. Li, D. Kato, F. J. Currell, C. Yamada, and S. Ohtani, J. Phys.: Conf. Ser. 58, 239 (2007).
[12] D. A. Knapp, P. Beiersdorfer, M. H. Chen, J. H. Scofield, and D. Schneider, Phys. Rev. Lett. 74, 54 (1994).
[13] N. Nakamura, H. Tobiyama, H. Nohara, D. Kato, H. Watanabe, F. J. Currell, and S. Ohtani, Phys. Rev. A 73, 020705 (2006).