Evidence for Strong Breit Interaction in Dielectronic Recombination of Highly Charged Heavy Ions

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Resonant strengths have been measured for dielectronic recombination of Li-like iodine, holmium, and bismuth using an electron beam ion trap. By observing the atomic number dependence of the state-resolved resonant strength, clear experimental evidence has been obtained that the importance of the generalized Breit interaction (GBI) effect on dielectronic recombination increases as the atomic number increases. In particular, it has been shown that the GBI effect is exceptionally strong for the recombination through the resonant state $[1s2s^22p_{1/2}]_1$.

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The Quantum electrodynamic (QED) effects are quite important for highly charged heavy ions. One of the most important QED effects is the shift in the energy level. To date, many experimental and theoretical studies have shown the importance of the QED effects on the energy level (see, for example, the review by Plunien and Soff [1] and references therein). Recently, there have been a few excellent studies in which higher-order QED is tested [2]. In their studies, the transition energies of few-electron heavy ions were measured very precisely, and the small QED contribution on energy levels was investigated. On the other hand, the QED effects are also important for the collision processes of electrons and highly charged ions [3–6]. The lowest order QED effect in the electron collision processes can be described through the so-called generalized Breit interaction (GBI) [7], in which the retardation in the exchange of a single virtual photon between the free electron and the orbital electron in the ion is included. The importance of the GBI has already been confirmed for several processes in the collisions between electrons and highly charged ions. For example, for ionization, it was experimentally found that the electron impact ionization cross section for H-like uranium enhanced by about 50% due to the GBI [5,6]. For excitation, Widmann *et al.* [8] showed in their high-resolution spectroscopic measurements of the *K* x-ray spectra of hydrogenlike Xe^{53+} and heliumlike Xe^{52+} that the GBI plays an important role in the collisional excitation process. The GBI is important also for recombination processes, such as dielectronic recombination (DR) and resonant transfer and excitation (RTE). Zimmerer *et al.* [9] calculated DR cross section for H-like uranium using a distorted wave theory with and without the GBI correction, and showed the importance of the GBI effect. Badnell and Pindzola [3] used multiconfiguration Dirac-Fock wave functions with a Breit correction in order to calculate the DR cross section

for H-like bismuth, and also showed the importance of the GBI. There have been some experimental [10,11] and theoretical [12] studies which showed the importance of the GBI in RTE in which the collision partner is not a simple free electron but an electron bound to a target. In RTE measurements, the resolution is limited by the Compton profile of the target electron so that state-resolved measurements have not been reported. Despite the possibility of DR measurements augmenting the existing findings of RTE measurements through improved state-selectivity, there have been no experimental measurements reported which confirm the importance of the GBI effect on DR with an unbound electron although some theoretical studies [3] exist. This is because the GBI effect, which is one of the QED effects, becomes observable only for the processes involving deep inner-shell electrons in heavy ions, e.g., Knn' DR (the processes in which an electron is excited from the *K* shell to the *n* shell while the incident electron is captured into the n' shell) for few-electron heavy ions, for which it is difficult to perform experiments. Up to now, DR processes involving *K* shell electrons in few-electron heavy ions have been observed through x-ray observation with an electron beam ion trap (EBIT) [13]. For example, *KLL* DR resonances have been observed for mercury (atomic number $Z = 80$) [14], bismuth ($Z = 83$) [15], and uranium ($Z = 92$) [16]. In such x-ray measurements, however, it is difficult to resolve charge states mixed in the EBIT completely and to get enough statistics so that the GBI effect on DR has never been discussed. DR is a combination of dielectronic resonant capture which is the inverse process of Auger decay and radiative decay of doubly excited states created by the dielectronic resonant capture. The GBI effect on the Auger rate itself has been investigated through Auger electron spectroscopy of highly charged light ions such as Na-like Ar [17,18]. However, such measurement is practically impossible for

heavy ions because the branching ratio for Auger decays becomes quite small for heavy ions so that the Z dependence has never been investigated.

In this Letter, we present DR measurements for Li-like iodine, holmium, and bismuth ions by an alternative effective method, where the charge abundance ratio in an EBIT when the charge balance is at equilibrium is measured. By investigating the behavior of the DR resonant strength as a function of Z, we have clearly showed that the importance of the GBI rapidly increases with Z. Very recently, KLL DR resonances for H-like uranium have been observed with the heavy ion storage ring ESR of the Gesellschaft für Schwerionenforschung (GSI) [19] in a complimentary study. These results are still undergoing analysis, but it is expected that they too will show the effect of the GBI. The distinctive feature of our study is that it shows the strong Z dependence of the GBI effect and, in particular, the state-selective nature of this dependence.

The experimental method and procedure used in the present study are similar to those used by Ali *et al.* [20] and described in detail in our previous papers [21,24]; thus, they will be briefly described here. In the present study, an EBIT in Tokyo [22] was used. In order to investigate the Z dependence of the GBI effect, iodine ($Z = 53$), holmium ($Z = 67$), and bismuth ($Z = 83$) were studied. Iodine was introduced into the EBIT through a gas injector while holmium and bismuth were introduced with an effusion cell [23]. The injection was done continuously for all the elements used.

In the trap region of the EBIT, highly charged ions are produced through successive ionization by energetic electron beam impact. At the equilibrium, the ionization balance for trapped ions is determined by the rates for ionization and recombination processes. Thus, the ion abundance ratio for the adjacent charge-states q and $q - 1$ can be expressed in terms of cross sections for relevant collision processes [24],

$$\frac{n_{q-1}}{n_q} = \frac{\sigma_q^{\mathrm{DR}} + \sigma_q^{\mathrm{RR}} + \langle \sigma_q^{\mathrm{CX}} \rangle}{\sigma_{q-1}^{\mathrm{ion}}}, \tag{1}$$

where σ_q^{DR} and σ_q^{RR} are the dielectronic and radiative recombination cross sections for the ion with a charge state q, $\sigma_{q-1}^{\mathrm{ion}}$ the electron impact ionization cross section for the ion with $q - 1$, and $\langle \sigma_q^{\mathrm{CX}} \rangle$ the effective charge exchange cross section [6] for collisions with the residual gas.

When there is no resonant process, i.e., when $\sigma_q^{\mathrm{DR}} = 0$, the ratio n_{q-1}/n_q varies slowly with electron energy. However, when the interaction energy of electrons coincides (within the beam energy width of about 50 eV) with a DR resonant energy, the abundance ratio changes drastically by an amount of $\sigma_q^{\mathrm{DR}}/\sigma_{q-1}^{\mathrm{ion}}$. Across a narrow electron energy region where the ionization cross section $\sigma_{q-1}^{\mathrm{ion}}$ can be regarded as a constant, the enhancement in the abundance ratio can thus be considered to be due to the resonant structure of the DR cross section which has strong energy

dependence. Thus, the normalized DR cross section [σ_q^{DR}], which is defined as $\sigma_q^{\mathrm{DR}}/\sigma_{q-1}^{\mathrm{ion}}$, is derived from the experimental abundance ratio by subtracting the slowly varying background $b(E_e)$ which corresponds to $(\sigma_q^{\mathrm{RR}} + \langle \sigma_q^{\mathrm{CX}} \rangle)/\sigma_{q-1}^{\mathrm{ion}}$;

$$[\sigma_q^{\mathrm{DR}}] \equiv \frac{\sigma_q^{\mathrm{DR}}}{\sigma_{q-1}^{\mathrm{ion}}} = \frac{n_{q-1}}{n_q} - b(E_e). \tag{2}$$

In this study, the ion abundance ratio was obtained by measuring the intensity of ions extracted from the EBIT. Ions escaping from the trap were extracted into a beam line, and detected after the charge separation with an analyzing magnet. Several charge states were detected at the same time by using a position sensitive detector. It was assumed that the total efficiency (escape, transport and detection) was the same for the adjacent charge state ions. This assumption was confirmed experimentally by comparing the charge state abundance obtained from the x-ray observation of trapped ions with that obtained from extracted-ion observation [24]. The electron energy was stepwise scanned by controlling the voltage at the electron gun. For each step, counting of the ions was started 2 sec after the electron energy was changed to ensure the equilibrium condition had been established, and continued for 8 sec. The voltage of the ion trap region was fixed to $+3$ kV throughout the experiment.

For comparison with the experimental results, we calculated DR resonant strengths in the isolated resonance approximation. Although the interaction between resonances is important [25,26], in this Letter, we concentrated on the $KL_{12}L_{12}$ resonances for which the separation between resonances is so large that the isolated resonance approximation is well validated. Atomic structures of target ions were calculated by using the GRASP II code [27]. Continuum wave functions of the incident electrons were obtained by solving the Dirac equation in the distorted-wave approximation. The distorted-wave potentials were given by self-consistent potentials from GRASP II calculations. There was little difference in the potentials between the target-ion states and the recombined states. The Auger decay rates were calculated by using a recently developed program code [28]. In the program code, the rates of Auger decay from the autoionization state Ψ_j to the final continuum state Ψ_i are given by $A_{ji}^a = 2\pi |\langle \Psi_j | \sum_{p < q} V_{pq} | \Psi_i \rangle|^2$, where inter-electron interaction V_{pq} includes the GBI as well as the Coulomb interaction. Here, the Hartree atomic units are used unless otherwise stated. V_{pq} is expressed by, with the inter-electron distance r_{pq} and the wave number of the virtual photon ω,

$$V_{pq} = \frac{1}{r_{pq}} - \left[\vec{\alpha}_p \cdot \vec{\alpha}_q \frac{e^{i\omega r_{pq}}}{r_{pq}} + (\vec{\alpha}_p \cdot \vec{\nabla}_p)(\vec{\alpha}_q \cdot \vec{\nabla}_q) \right.$$
$$\left. \times \frac{e^{i\omega r_{pq}} - 1}{\omega^2 r_{pq}} \right] \tag{3}$$
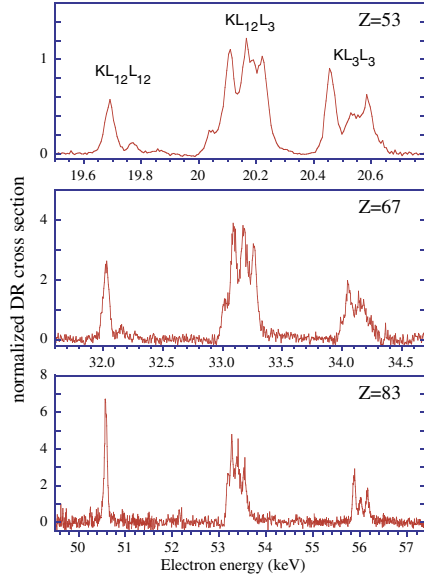
FIG. 1 (color online).   Dielectronic recombination cross sections of Li-like ions normalized to the ionization cross sections of Be-like ions for iodine ($Z = 53$), holmium ($Z = 67$), and bismuth ($Z = 83$). The horizontal energy scale was calibrated with the theoretical resonance energy.

where $\vec{\alpha}$s are the Dirac matrices. The imaginary component of the GBI operator, which is higher order in terms of the wave number and usually discarded, is taken into account in the present code. In DR calculations, all possible decay paths were taken into account, including radiative decay to lower doubly excited states and autoionizing decay to excited states. Magnetic dipole ($M1$) and electric quadrupole ($E2$) transitions were included in the radiative decay rates. The $E2$ transition was negligibly weak in general, while the $M1$ transition had a considerable contribution to the total decay rate.

Figure 1 shows the normalized DR cross section defined in Eq. (2) as a function of electron beam energy across the KLL DR resonance region. It is noted that escape and multiple charge transfer, which are not included in Eq. (1), can affect the abundance ratio and then the normalized cross section. These effects can make a dip at the DR resonance energy for the He-like ion [24]. For the iodine data, these effects were corrected as was done in our previous study [24]. For holmium and bismuth, no correction was applied because those effects were negligibly small (as confirmed from the fact that there is no dip in the data) due to the small abundance of the He-like ions compared with that of the Li-like and Be-like ions. For all the elements studied, three manifolds were observed, which correspond to $KL_{12}L_{12}$, $KL_{12}L_3$, and KL_3L_3 DR resonances (L_{12} denotes the $2s_{1/2}$ and $2p_{1/2}$ levels and L_3 the $2p_{3/2}$ level). As seen in Fig. 1, it was found that the resonance at the lowest energy side has strong Z-dependence; i.e., its relative intensity with respect to that of the other resonances rapidly increases as Z increases. To see this in detail, the close-up view of the
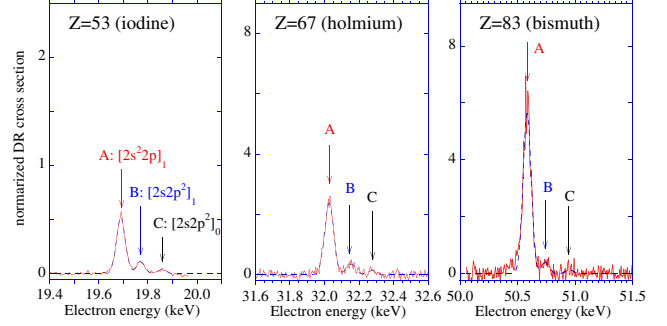

FIG. 2 (color online).   Similar to Fig. 1, but for the $KL_{12}L_{12}$ region. The solid line shows experimental results and the broken line the Gaussian functions fitted to the experimental data.

normalized DR cross section for the $KL_{12}L_{12}$ region is shown in Fig. 2. For iodine and holmium, the same data were used for both Fig. 1 and 2, while for bismuth, higher statistical quality data obtained by longer accumulation over a narrower region was used for Fig. 2. For each ion, three peaks were found and identified as shown in the figure. Owing to the narrow electron beam energy spread ($\sim$50 eV) in the EBIT compared to the energy separation of the resonant levels, these levels could be clearly resolved. As clearly seen in the figure, the DR cross section for the $2s^22p_{1/2}$ resonance (labeled A) rapidly increases with Z with respect to that of $[2s2p^2_{1/2}]_1$ (B) and $[2s2p^2_{1/2}]_0$ (C). Here, we drop the $1s$ from the notation.

Figure 3 shows the resonant strength ratio between the $[2s^22p_{1/2}]_1$ (A) and $[2s2p^2_{1/2}]_1$ (B) resonances, where the resonant strength is defined as the energy-integrated DR cross section. The ratio was obtained from the area of the Gaussian function fitted to each peak in Fig. 2. Since the
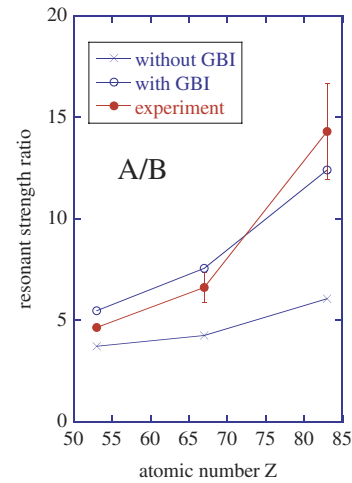

FIG. 3 (color online).   Resonant strength ratios between the $[2s^22p_{1/2}]_1$ (A) and $[2s2p^2_{1/2}]_1$ (B) resonant states. Closed circles represent the present experiments while open circles and crosses theoretical values with and without the generalized Breit interaction, respectively. The error bars represent only the statistical component. All systematic errors are considered to be cancelled out by taking ratios.

TABLE I.   Theoretical Auger rates and resonant strengths for the $[2s^2 2p_{1/2}]_1$ (A), $[2s2p_{1/2}^2]_1$ (B), and $[2s2p_{1/2}^2]_0$ (C) of bismuth.

		Auger rates 10^{14} (s^{-1})	resonant strength 10^{-20} ($cm^2 \, eV$)
$[2s^2 2p_{1/2}]_1$ (A)	with GBI	4.1	2.9
	w/o GBI	1.9	1.4
$[2s2p_{1/2}^2]_1$ (B)	with GBI	0.33	0.24
	w/o GBI	0.34	0.25
$[2s2p_{1/2}^2]_0$ (C)	with GBI	0.36	0.088
	w/o GBI	0.35	0.086

vertical axis of Fig. 2 is the normalized cross section defined by Eq. (2), the integrated area of the peak gives the resonant strength divided by the ionization cross section, i.e., $\Omega_q / \sigma_{q-1}^{ion}$, where Ω_q denotes the DR resonant strength. Thus, the area ratio between two resonances in Fig. 2, i.e., the vertical axis of Fig. 3, gives the resonant strength ratio Ω_q^A / Ω_q^B for the two resonances *A* and *B*. Here, we consider the ratio Ω_q^A / Ω_q^B to investigate the *Z*-dependent characters of the wave functions with the same electron-configuration in the $KL_{12}L_{12}$ resonances. It is clearly seen in Fig. 3 that the experimental ratio shows remarkable increase with *Z*.

In order to investigate the origin of this strong *Z* dependence, we compared the experimental results with two different theoretical values. One of them, which is shown by crosses in Fig. 3, was obtained with fully relativistic calculations but without GBI. Disagreement between the experiment and the theory is quite large especially for bismuth in spite of a relativistic wave function being used in the calculation. On the other hand, another theoretical result obtained by including the GBI is shown by open circles in the same figure. As clearly seen in the figure, the GBI modifies the resonant strength ratio by a large amount, and the agreement with the experiment becomes good. Therefore, it is concluded that the GBI effect is responsible for the strong *Z* dependence observed in the present experiment. The absolute values for the resonant strengths and the Auger decay rates for the corresponding states of bismuth are listed in Table I. As seen in the table, the GBI effect on the Auger rate and the resonant strength is almost negligible for the $[2s2p_{1/2}^2]_1$ (B) and $[2s2p_{1/2}^2]_0$ (C) resonances. On the other hand, for the $[2s^2 2p_{1/2}]_1$ (A) resonance, the GBI is found to enhance the resonant strength by a factor of about 2 as a result of the enhancement of the Auger rate. As shown in this study, the *Z*-dependence of DR resonant strength is quite different among resonant levels.

In summary, we have measured the resonant strength for *KLL* DR processes in the collisions between electrons and Li-like iodine, holmium, and bismuth ions. State-resolved measurements have clearly shown that the GBI effect on

DR resonant strength has strong state-selectivity and is quite strong exceptionally for the $[2s^2 2p_{1/2}]_1$ resonance among resonances within the $KL_{12}L_{12}$ manifold.

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