Experimental Demonstration of the Breit Interaction which Dominates the Angular Distribution of X-ray Emission in Dielectronic Recombination

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We report the experimentally determined angular distribution of the $[1s2s^22p_{1/2}]_1 \rightarrow [1s^22s^2]_0$ transition in dielectronic recombination of Li-like Au. Recently, Fritzsche *et al.* [Phys. Rev. Lett. **103,** 113001 (2009)] predicted that the Breit interaction plays a dominant role in the angular distribution of this transition. However, the predicted phenomenon has not yet been observed experimentally due to technical difficulties in conventional methods. To overcome the difficulties, we combine two different measurements with an electron beam ion trap (EBIT) to obtain the x-ray angular distribution. One is the x-ray measurement at 90° and another is the integral resonant strength measurement through the ion charge abundance in the EBIT. Our measurements agree well with the theoretical prediction and confirm the dominance of the Breit interaction.

The Breit interaction was first introduced by G. Breit around 1930 to describe relativistic effects in the electron-electron interaction [1]. It includes magnetic interactions and retardation in the exchange of a single virtual photon between the electrons. Results presented in a series of his papers successfully explained the fine structure of atomic helium. The Breit interaction affects not only the energy structure but also the dynamics; that is, the Breit interaction between the incident electron and the electrons in the target should also be considered for electron collisions with atoms or ions. In general, the magnetic interactions and the retardation effects are so small that they are usually treated as minor corrections to the major term (i.e., the Coulomb interaction). However, the Breit interaction often contributes significantly to dynamics involving a highly charged ion (HCI). For example, our previous study [2] showed that the Breit interaction can enhance dielectronic recombination (DR) resonant strengths by almost 100%. Such a significant contribution from the Breit interaction in electron-HCI collisions has also been studied for ionization [3,4], excitation [5], and resonant transfer excitation (RTE) [6,7].

Recently, Fritzsche *et al.* [8] made a theoretical study of the effects of the Breit interaction on the angular distribution of x rays emitted from the DR, which is the combination of dielectronic capture (DC) and successive radiative stabilization (RS),

$$e^- + \mathrm{A}^{q+} \xrightarrow{\mathrm{DC}} \mathrm{A}^{(q-1)+**} \xrightarrow{\mathrm{RS}} \mathrm{A}^{(q-1)+} + h\nu. \quad (1)$$
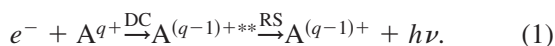
The angular distribution of the electric dipole emission can be expressed by the angular distribution coefficient $W(\theta)$ [9] as

$$W(\theta) = 1 + \beta P_2(\cos\theta), \quad (2)$$

where θ denotes the angle of emission with respect to the incident direction of the electron, P_2 is the Legendre polynomial, and β is the anisotropy parameter, which is related to the alignment parameter $\mathcal{A}_2$ [8,10] by $\beta = \mathcal{A}_2/\sqrt{2}$ for the $J = 1 \rightarrow 0$ transition. $\mathcal{A}_2$ is determined by the magnetic sublevel distribution of the intermediate state $\mathrm{A}^{(q-1)+**}$ produced by DC, and for $J = 1$ intermediate states, is expressed as

$$\mathcal{A}_2 = \sqrt{2}\frac{\sigma_{\pm 1} - \sigma_0}{2\sigma_{\pm 1} + \sigma_0}, \quad (3)$$

where σ_{M_J} denotes the population of the magnetic sublevel with the magnetic quantum number M_J. Fritzsche *et al.* [8] found that the Breit interaction dominates the alignment parameter, and thus, brings about a qualitative change in the angular distribution. According to their result, the angular distribution of the $[1s2s^22p_{1/2}]_1 \rightarrow [1s^22s^2]_0$ transition following DC into initially Li-like heavy ions is maximum at $\theta = 90°$ when the Breit interaction is considered. However, it is minimum at 90° when the Coulomb interaction is considered. They thus proposed to measure the angular distribution (or the polarization) of this DR x ray or the alignment parameter of the intermediate state to make a sensitive study of the effect of the Breit interaction on the DR processes of HCIs.

An electron beam ion trap (EBIT) [11] is a useful tool to study the polarization of emission from HCIs excited by an electron beam [12,13]. For example, Shlyaptseva *et al.* [14] measured the polarization-dependent spectra of x rays emitted by the DR of Be-like Fe. They used the

polarization sensitivity of a Bragg crystal spectrometer to prove the polarization properties. Bragg crystal spectrometers have also been used to measure the polarization of Lyman-α x rays. The energy dependence of the Ly-α polarization was measured for H-like Ar [15], Ti [16], and Fe [15], and their compilation recently proved the non-negligible contribution of the Breit interaction [17]. However, a Bragg crystal spectrometer is practically unsuitable for hard x rays, such as the lines of interest in this study (i.e., the K x rays of heavy ions). To observe such hard x rays with a Bragg spectrometer, one must use higher-order reflection, but its efficiency is practically inapplicable to the weak radiation from an EBIT. Recently, a novel polarimeter usable for hard x rays has been developed [18], but it is a state-of-the-art instrument and not commercially available at present.

Although the observation angle for EBIT experiments is usually limited to 90° with respect to the electron beam, a storage-ring device makes it possible to observe the angular distribution. For example, Ma *et al.* [7] measured the angular distribution of the RTE x rays emitted in collisions of H-like U with molecular hydrogen. Their result agrees well with theoretical calculations that include the Breit interaction. However, insufficient resolution caused by the Compton profile usually precludes proving the effect of the Breit interaction on state-resolved resonant processes.

In this Letter, we present the experimental determination of the angular distribution for the transition proposed by Fritzsche *et al.* using an EBIT in Tokyo. As an alternative to the direct angular distribution measurement, the alignment parameter is obtained from the combination of a differential x-ray measurement at 90° and an integral-resonant-strength measurement through the ion abundance inside the EBIT.

The present experimental method and procedure are similar to those used in our previous studies, both for the x-ray measurement [19,20] and for the ion-abundance measurements [2,21]; thus, they will be only briefly described here. For both the experiments, the Tokyo-EBIT [22,23] was used. To study the Breit-interaction effect on heavy ions, Au (atomic number $Z = 79$) was injected into the EBIT through an effusion cell [24]. It is also important to study the radiative processes of highly charged Au ions for the diagnostics and the control of indirect-drive inertially confined fusion plasmas [25].

For the x-ray measurement, the electron beam energy was scanned over the $KL_{12}L_{12}$ DR resonance region of He- to Be-like Au, where L_{12} denotes $2s$ and $2p_{1/2}$ levels. The scanning pattern was a triangle form from 44.8 to 46.3 keV with a scanning rate of 20 ms/scan. A high-purity germanium detector placed at 90° with respect to the electron beam was used to detect the x-ray emission from the intermediate states produced by the DC. Figure 1 shows the typical spectra as a function of electron beam energy.
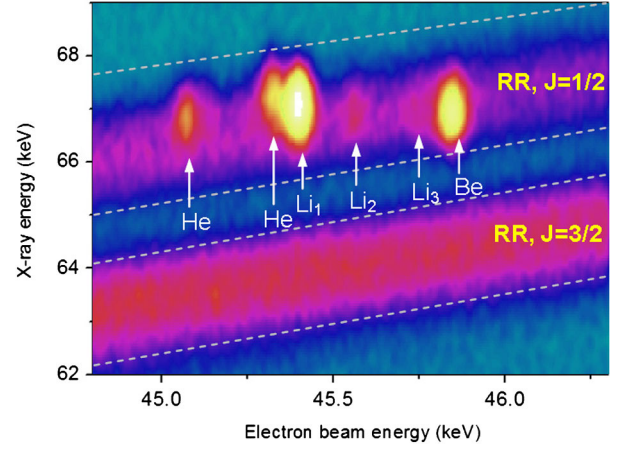

FIG. 1 (color online).   X-ray spectra from the $KL_{12}L_{12}$ DR of highly charged Au ions. X-ray counts are plotted as a brightness of color as functions of electron beam energy (horizontal axis) and x-ray energy (vertical axis). Bright spots correspond to x-ray enhancement due to the DR. The charge state responsible for each spot is indicated by labels. For the Li-like ion, which is of interest in the present study, the intermediate and final states are listed in Table I.

The x-ray energy was calibrated using the radiation from radio isotopes, whereas the electron energy was calibrated to the theoretical DR resonant energies. The two diagonal lines in the figure correspond to x rays emitted by radiative recombination (RR) into the L_{12} ($J = 1/2$) and L_3 ($J = 3/2$) orbitals. On the RR line for $J = 1/2$, several bright spots were observed, which correspond to x rays emitted by $KL_{12}L_{12}$ DR. Each spot is assigned, as indicated in Fig. 1, from comparisons with previous measurements [20,26,27] and theoretical calculations.

For the ion-abundance measurement, the electron-energy scan was done quasistatically to obtain the relative DR cross section $[\sigma_{Li}^{DR}]$ of Li-like Au from the ion-abundance ratio between Be-like and Li-like Au (n_{Be}/n_{Li}) using the following formula (valid only at equilibrium):

$$[\sigma_{Li}^{DR}] = \frac{n_{Be}}{n_{Li}} - B(E_e), \tag{4}$$

where $B(E_e)$ denotes the slowly varying background when the ion ratio n_{Be}/n_{Li} is plotted as a function of electron energy (see Ref. [2] for details). The abundance ratio was obtained by measuring the intensity of ions extracted from the EBIT. The electron energy was stepwise scanned, and we started counting the ions 2 s after the electron energy was changed (to ensure that charge equilibrium was established) and continued for 8 s. For both the x-ray and ion-abundance measurements, the electron beam current was 50 mA and the electron beam energy width was about 65 eV at full width at half maximum.

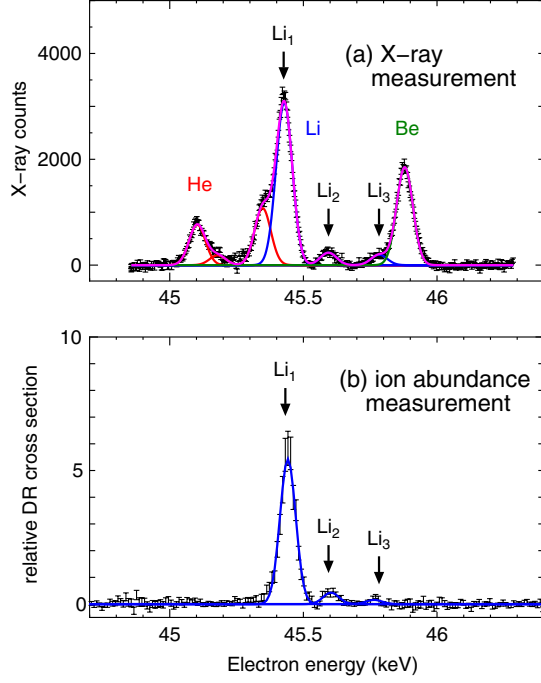Figure 2 shows the DR spectra obtained by (a) the x-ray measurement and (b) the ion-abundance measurement.

FIG. 2 (color online).   (a) K x-ray intensity plotted as a function of electron beam energy for the $KL_{12}L_{12}$ resonance region of highly charged Au ions. The background corresponding to radiative recombination is subtracted. The labels represent the charge state responsible for the DR process that led to x-ray enhancement. (b) Relative DR cross section [Eq. (4)] of Li-like Au obtained from ion-abundance measurement. The Gaussian functions fit to the experimental data are also shown by solid lines for both (a) and (b).

Figure 2(a) was obtained by projecting the x-ray counts given in Fig. 1 onto the electron-energy axis only for the $J = 1/2$ RR line. The background corresponding to the RR contribution is subtracted. Among the several DR peaks observed, the three peaks labeled Li_1 to Li_3 correspond to the DR of Li-like Au. The intermediate state $|d\rangle$ and final state $|f\rangle$ for Li_1 and Li_2, which are used to obtain the alignment parameter, are listed in Table I. The line proposed by Fritzsche *et al.* [8] corresponds to the transition from the intermediate state for Li_1 to the ground state; thus, the goal of this study is to determine the alignment parameter $\mathcal{A}_2$ for Li_1. We can obtain $\mathcal{A}_2$ by separately normalizing (as derived in the following) the x-ray and ion-abundance measurements to a reference transition, Li_2.

TABLE I.   Initial and final states for the $KL_{12}L_{12}$ DR shown in Figs. 1 and 2. Branching ratios calculated by using the Flexible Atomic Code [28] are also listed.

Label	Intermediate state $|d\rangle$	Final state $|f\rangle$	Branching ratio
Li_1	$[1s2s^2 2p_{1/2}]_1$	f_1: $[1s^2 2s^2]_0$	0.98
		f_2: $[1s^2 2p_{1/2}^2]_0$	0.02
Li_2	$[1s2s2p_{1/2}^2]_1$	f_3: $[1s^2 2s2p_{1/2}]_0$	0.34
		f_4: $[1s^2 2s2p_{1/2}]_1$	0.66

In the x-ray measurement, the intensity $I^{\text{X-ray}}$ (the area of each peak) should be proportional to the product of the DR resonant strength S^{DR} and the angular distribution coefficient W at $\theta = 90°$, which in turn is determined as follows by the total angular momentum of the intermediate and final states [8,10]:

$$W_d^{J_d \to J_f} = \begin{cases} 1 - \mathcal{A}_2^d/2\sqrt{2} & (J_d = 1, J_f = 0) \\ 1 + \mathcal{A}_2^d/4\sqrt{2} & (J_d = 1, J_f = 1) \end{cases}, \quad (5)$$

where $\mathcal{A}_2^d$ represents $\mathcal{A}_2$ for the intermediate state $|d\rangle$. Since the x-ray resolution is insufficient to resolve x rays from different final states, the contribution from all the final states should be summed to describe the intensity. Consequently, the following relationship is obtained using the resonant strength for DC (S^{DC}) and the branching ratio (B):

$$I_d^{\text{x ray}} \propto \sum_f S_{df}^{\text{DR}} W_d^{J_d \to J_f} = \sum_f S_d^{\text{DC}} B_f W_d^{J_d \to J_f}. \quad (6)$$

The ratio between the x-ray intensities of Li_1 and Li_2 is thus given by the following formula:

$$R_{Li_1/Li_2}^{\text{x ray}} \equiv \frac{I_{Li_1}^{\text{x ray}}}{I_{Li_2}^{\text{x ray}}} = \frac{S_{Li_1}^{\text{DC}} B_{f_1} W_{Li_1}^{1 \to 0} + S_{Li_1}^{\text{DC}} B_{f_2} W_{Li_1}^{1 \to 0}}{S_{Li_2}^{\text{DC}} B_{f_3} W_{Li_2}^{1 \to 0} + S_{Li_2}^{\text{DC}} B_{f_4} W_{Li_2}^{1 \to 1}}$$
$$= \frac{S_{Li_1}^{\text{DC}} W_{Li_1}^{1 \to 0}}{S_{Li_2}^{\text{DC}} (B_{f_3} W_{Li_2}^{1 \to 0} + B_{f_4} W_{Li_2}^{1 \to 1})}. \quad (7)$$

B_{f_3} and B_{f_4}, which are the branching ratios to the same electronic configuration but with different total angular momentums, should be $1/3$ and $2/3$, respectively, for the single configuration in the jj coupling scheme. As listed in Table I, we confirm that they are close to the jj coupling limit values for Au. The quantity in parentheses is thus unity regardless of $\mathcal{A}_2^{Li_2}$;

$$\frac{1}{3}\left(1 - \frac{\mathcal{A}_2^{Li_2}}{2\sqrt{2}}\right) + \frac{2}{3}\left(1 + \frac{\mathcal{A}_2^{Li_2}}{4\sqrt{2}}\right) = 1. \quad (8)$$

Thus, Eq. (7) can be simplified to

$$R_{Li_1/Li_2}^{\text{x ray}} = S_{Li_1}^{\text{DC}} W_{Li_1}^{1 \to 0} / S_{Li_2}^{\text{DC}}. \quad (9)$$

On the other hand, the ratio of the peak area in the ion-abundance measurement [Fig. 2(b)] is simply given by the ratio between the DC resonant strengths,

$$R_{Li_1/Li_2}^{\text{ion}} = S_{Li_1}^{\text{DC}} / S_{Li_2}^{\text{DC}}. \quad (10)$$

TABLE II.   Experimental results. The error (1σ) listed in the table corresponds to the error of the least square fitting weighted by the statistical uncertainties.

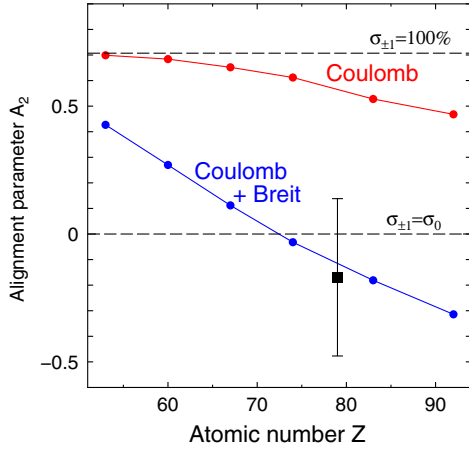| $R_{Li_1/Li_2}^{\text{X-ray}}$	$R_{Li_1/Li_2}^{\text{ion}}$	W_{Li_1}	$\mathcal{A}_2$
13.0 ± 0.9	12.3 ± 0.9	1.06 ± 0.11	-0.17 ± 0.31

FIG. 3 (color online).   Alignment parameter $\mathcal{A}_2$ for intermediate state $[1s2s^2 2p_{1/2}]_1$ in DR of Li-like ions. The present result for Au ($Z = 79$) is shown by the black square. Theoretical values taken from Ref. [8] are plotted as red dots (Coulomb interaction only) and blue dots (with Breit interaction).

Consequently, the angular distribution coefficient for Li$_1$ can be obtained by taking the ratio between Eqs. (9) and (10):

$$W_{\mathrm{Li}_1}^{1 \to 0} = R_{\mathrm{Li}_1/\mathrm{Li}_2}^{\mathrm{x\,ray}} / R_{\mathrm{Li}_1/\mathrm{Li}_2}^{\mathrm{ion}}. \qquad (11)$$

Finally, the alignment parameter $\mathcal{A}_2$ for Li$_1$ can be obtained from Eq. (5). Note that, to derive Eq. (11), we only assume that the branching ratios B_{f_3} and B_{f_4} are the ones obtained for the single configuration in the jj-coupling scheme. No other theoretical calculation is needed.

The experimental ratios are listed in Table II. The ratio $R_{\mathrm{Li}_1/\mathrm{Li}_2}^{\mathrm{X\text{-}ray}}$ was obtained by fitting Gaussian functions to the data shown in Fig. 2(a), which required about 200 h of data acquisition to obtain sufficient statistics. The ratio $R_{\mathrm{Li}_1/\mathrm{Li}_2}^{\mathrm{ion}}$ was obtained from two runs by fitting Gaussian functions to the data. A ratio of 12.6 ± 1.4 was obtained from the data shown in Fig. 2(b), which took about 7 h to acquire. From another run with 8 h accumulation, the ratio was 12.0 ± 1.3. The value listed in the table is the weighted average of the two runs. The table also lists W_{Li_1} and $\mathcal{A}_2$, which were determined from the experimental ratios. $\mathcal{A}_2$ is also plotted in Fig. 3 together with the theoretical values [8].

The present experimental result for Li-like Au indicates that the angular distribution of the emitted x ray has a maximum at 90° (negative $\mathcal{A}_2$ value), while the prediction that considers only the Coulomb interaction gives a minimum (positive $\mathcal{A}_2$ value) at 90°. As seen in Fig. 3, the theoretical result that includes the Breit interaction reproduces the experimental result quantitatively, whereas that with only the Coulomb interaction fails to do so. This is a clear demonstration that the Breit interaction dominates the angular distribution of x rays emitted in DR processes of highly charged heavy ions.

In summary, we have measured the alignment parameter of the intermediate state in the DR for Li-like Au with the Tokyo electron beam ion trap. By combining the x-ray measurement at 90° with respect to the electron beam and the relative resonant strength measurement through the ion abundance inside the trap, we obtained the angular distribution of the $[1s2s^2 2p_{1/2}]_1 \to [1s^2 2s^2]_0$ transition. The comparison with theory clearly reveals the dominance of the Breit interaction.

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