Strong Polarization of a J = 1/2 to 1/2 Transition Arising from Unexpectedly Large Quantum Interference

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We experimentally show that the $1s^22s^22p_{1/2} - 1s2s^22p_{1/2}^2$ transition in Pb⁷⁷⁺ emitted in dielectronic recombination of Pb⁷⁸⁺ is strongly polarized, although it is an intrinsically unpolarized J = 1/2 to 1/2 transition. This unanticipated polarization is shown to be due to quantum interference with radiative recombination. The interference effect has been studied on an asymmetric resonance profile but has never been studied on polarization. In this Letter, we show that the effect on polarization can arise from a different cross term than that responsible for asymmetry, resulting in unexpectedly large polarization even for a nearly symmetric resonance suggesting a small interference.

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Introduction.—The polarization of recombination radiation in electron-ion collisions is crucial for diagnosing high-temperature plasmas, such as fusion, astrophysical, and laser-produced plasmas [1–4]. For the recombination of electrons with highly charged ions, x-ray polarization is also crucial for testing fundamental atomic physics involving strong relativistic and quantum electrodynamics effects [5]. In low-density plasmas, such as the solar corona, there are mainly two recombination processes in electron-ion collisions: nonresonant radiative recombination (RR) and resonant dielectronic recombination (DR)—

$$RR: e + A^{q+} \to A^{(q-1)+} + h\nu, \qquad (1)$$

DR:
$$e + A^{q+} \to A^{(q-1)+**} \to A^{(q-1)+} + h\nu$$
, (2)

where $A^{(q-1)+**}$ represents the inner-shell excited autoionizing state. The polarization or equivalent angular distribution of RR and DR x rays has been measured using an electron beam ion trap (EBIT) [6–10]; however, experimental studies are limited due to technical difficulties in measuring the degree of polarization of hard x rays for which crystal Bragg polarimeters [11] cannot be used.

When both the initial and final states are the same between RR and DR, they can make quantum interference. In atomic and molecular physics, the interference effects are found in almost all dynamical processes, such as multiphoton ionization [12,13], electron-atom collisions [14], atom-atom collisions, or molecular dissociations [15], and time-dependent strong field processes [16]. Attention has been paid to the so-called Fano or Feshbach resonances [17,18], which give rise to asymmetric line profiles due to an interference of a discrete state with a continuum state. These resonances are of broad interest in physics and, hence, have been investigated not only for atomic and molecular systems [19,20], but also for various systems from particle physics systems [21] to nanoscale mesoscopic systems [22,23]. The Fano resonances can also be actively used to control cold atom collisions [24] and extract relevant scattering information in nuclear scattering [25] and atomic collisions [26].

In previous experimental and theoretical studies [27–36], the interference between DR and RR involving highly charged heavy ions has been investigated mainly by focusing on the asymmetry in the Fano profile, because the degree of this asymmetry (usually represented by the so-called Fano shape parameter [17]) is a measure of the strength of the interference effect. However, most DR resonances have been confirmed to reveal a nearly symmetric profile, indicating a rather small interference effect. Therefore, the interference effect on the polarization of recombination x rays has also been considered insignificant and has never been investigated.

In this Letter, we present a polarization measurement for the following DR x ray of B-like Pb^{77+} emitted at 90° with respect to the incident electron direction:

$$e + 1s^2 2s^2 \rightarrow [1s2s^2 2p_{1/2}^2]_{1/2} \rightarrow [1s^2 2s^2 2p_{1/2}]_{1/2} + h\nu.$$
(3)

This DR x ray is emitted in a J = 1/2 to 1/2 transition; thus, it is intrinsically unpolarized. However, an unanticipated strong polarization is detected in the present experiment with an EBIT in Tokyo (Tokyo-EBIT) [37] and a recently developed Compton x-ray polarimeter [38]. Our theoretical study shows that the strong polarization is attributed to the interference between DR and RR, which have initial states with different total angular momenta. Although interference between different partial waves vanishes in the total cross section, it can appear in differential measurements. We show that the interference affects the polarization and profile in different manners; thus, the interference effect on the polarization can be unexpectedly large even for the resonance whose nearly symmetric resonance profile suggests a small interference effect.

Experiment.—The Tokyo-EBIT [37,39] was used for producing highly charged Pb ions. The EBIT has a Penning-like ion trap and a high-energy, high-density electron beam traveling through the trap. Highly charged ions are produced through successive electron impact ionization in the trap. A strong axial magnetic field produced by a superconducting magnet surrounding the trap is used for the radial confinement of the ion and for compressing the electron beam. The electron energy can be controlled by changing the potential of the center electrode of the trap with respect to that at the electron gun. To produce Pb ions, a vapor of Pb was injected through a side port of the EBIT using an effusion cell operated at 400 °C. Oxygen was introduced as a cooling gas from a gas injector at another side port.

X rays from trapped Pb ions emitted at 90° with respect to the electron beam were observed with the EBIT-CC [38], which is a recently developed hard x-ray Compton polarimeter based on a state-of-the-art detector for astronomical observations on board the satellite *Hitomi* [40–44]. It consists of Si detector layers and CdTe detector layers surrounding the Si layers. The Si detectors mainly act as scatterers, whereas the CdTe detectors as absorbers. Each layer is pixelized so that the scattered and absorbed positions of Compton scattered x rays and, hence, the scattered angle can be determined. From the azimuthal



FIG. 1. Two-dimensional plot of x-ray spectra from highly charged Pb ions as a function of the electron beam energy (bottom panel). The two diagonal lines correspond to RR into the j = 1/2 ($2s_{1/2}$ and $2p_{1/2}$) and j = 3/2 ($2p_{3/2}$) orbitals of He- to F-like Pb ions. The top panel shows the theoretical DR cross sections calculated with the flexible atomic code [45]. The arrows with "on" and "off" represent the energies where the polarization measurement was performed (see the text for details). The middle panel shows the experimental DR spectrum (black squares) obtained by integrating the two-dimensional data along the RR line for j = 1/2 and the fitted Gaussian functions (red, blue, and green lines). The experimental electron energy scale was normalized to the theoretical resonance energy for Li-like Pb.

angular distribution of the Compton scattered x rays, the degree of linear polarization of the incident x rays, which is defined by $P = (I_{\parallel} - I_{\perp})/(I_{\parallel} + I_{\perp})$, can be obtained, where I_{\parallel} and I_{\perp} represent the intensity of the x rays with an electric vector parallel and perpendicular to the quantization axis, respectively. A commercial pure Ge detector was placed at another observation port and used to monitor the x rays.

Figure 1 shows the x-ray spectra observed with the Ge detector to find the resonance energy before the polarization measurement with the EBIT-CC. It was obtained with the Ge detector while sweeping the electron beam energy over the $KL_{12}L_{12}$ resonance energy range. At several electron energies, the enhancement of x-ray intensity due to DR is confirmed on the diagonal lines corresponding to RR x rays into the j = 1/2 orbitals. The plot with the black squares in the middle panel shows the electron energy dependence of the x-ray intensity for the i = 1/2 diagonal line after subtracting the contributions of RR x rays and background. Gaussian profiles fitted to the data are also shown. From the comparison with a previous study [6] and the theoretical resonance strength calculated using the flexible atomic code [45] shown in the upper panel, they are assigned as DR into He- to Be-like ions. In this Letter, we are interested in the polarization of x rays emitted from the DR process of Be-like Pb indicated by the green curve. However, at the resonance energy, not only the objective DR x rays but also RR x rays from several charge state ions trapped in the EBIT were detected. The energy of the RR x rays for other charge states differs from that of the objective DR x rays, but it is practically impossible to resolve them due to a limited energy resolution. Thus, the observations of the RR and DR x rays (piling on the RR x rays) were alternatively performed by switching the electron beam energy between 49.89 and 49.72 keV shown by the "off" and "on" arrows in the top panel in Fig. 1, respectively. The energy switching was performed by the fast control of the potential at the trap region with a high-speed and highvoltage amplifier (Trek model 10/10) at a slew rate of $250 \text{ V/}\mu\text{s}$. In order not to change the charge abundance during the switching operation, the on and off periods were 3 and 7 ms, respectively, which are much shorter than the mean free time for recombining collisions under the present experimental condition. The data obtained within 0.5 ms after the potential change were not used, as the under- or overshooting of the potential at the trap was confirmed.

Results.—X-ray spectra obtained with the Ge detector for the on and off periods are shown in Fig. 2(a). The vertical scale is normalized by live time. The enhancement of the j = 1/2 peak (recombination x rays for the $2l_{1/2}$ orbitals) in the on spectrum corresponds to the DR into Be-like Pb [the process expressed in Eq. (3)]. The total observation time was about 230 h; thus, the experimental conditions such as the target density and coolant oxygen gas pressure could have changed during the long observation time. However, the fast energy switching with a millisecond timescale ensured the experimental conditions were the same between the on and off periods, as the fluctuations in the experimental conditions had a much larger time



FIG. 2. X-ray spectra obtained with (a) a Ge detector and (b) EBIT-CC. The red and black curves are the spectra for the on and off periods, respectively. Notably, the off spectrum is shifted by -170 eV (electron energy difference between the on and off periods). The blue curve in (b) represents the DR x-ray spectrum obtained by subtracting the off spectrum (shifted by -170 eV) from the on spectrum. The magenta solid line is the simulated spectrum for monoenergetic 73.7 keV x rays.

constant in the order of minutes to hours. In fact, the nearly complete agreement between the on and off spectra other than the j = 1/2 peak indicates the equal charge abundance for both periods.

Figure 2(b) shows the spectra of the Compton scattered x rays obtained with EBIT-CC simultaneously with the Ge detector measurements shown in Fig. 2(a). They were obtained by summing the energies deposited at the scattered and absorbed positions for the events where the incident x rays were scattered by the Si detectors and absorbed by the Si or CdTe detectors. Two peaks corresponding to the RR into n = 2 and 3 orbitals were observed. Because the detection efficiency drops rapidly with a decrease of the incident x-ray energy in this energy range, the relative intensity ratio between the n = 2 and 3 peaks differs from that in the Ge spectra. Unfortunately, due to the limited energy resolution, the i = 1/2 and 3/2peaks were not resolved in the EBIT-CC spectra. However, the enhancement due to the DR was clearly observed on the n = 2 peak. The enhanced component, which should correspond to the monoenergetic DR x rays arising from the process (3), is obtained by subtracting the off spectrum from the on spectrum, as shown by the blue curve in the figure. The magenta line represents the monoenergetic x-ray spectrum obtained with a Monte Carlo simulation [10,38]. The agreement between the enhanced spectrum (on-off) and the simulated spectrum supports that the enhanced component is the monoenergetic DR x rays.

The azimuthal angular distribution of the Compton scattered events for the enhanced DR x rays [corresponding to the "on-off" component in Fig. 2(b)] was obtained by subtracting the distribution for the off period from that for the on period after normalizing them with the measurement time. The event selection was made using the same parameters as those in our previous studies [10,38], except for the energy, which is 70-78 keV in this study. The modulation curve obtained by dividing the azimuthal angular distribution with the detector response function [10,38] is shown in Supplemental Material [46], confirming a distinct $\cos(2\phi)$ functional modulation indicating strong polarization. The degree of linear polarization of the DR x rays was obtained to be 0.327 with a statistical uncertainty of 0.034 using a likelihood analysis as in our previous studies [10,38]. Even if we assume 0.049 as the total uncertainty considering that the systematic uncertainty is 0.015 [38], the deviation from P = 0 expected in the absence of interference is as large as 6.7σ .

Discussion.—In previous studies [6–9], the polarization of DR x rays has been theoretically obtained without considering RR. However, the nonzero polarization of a J = 1/2 to 1/2 transition cannot be explained without interference. Thus, we performed a calculation including the interference with RR, whose effect on polarization has never been investigated as it has been considered to be small. Considering the electric dipole transition, the initial states for the recombination of a Be-like ion are $|\Psi_s\rangle = |1s^22s^2\epsilon s J = 1/2\rangle$ and $|\Psi_d\rangle = |1s^22s^2\epsilon d_{3/2} J = 3/2\rangle$, the autoionization state is $|\Psi_a\rangle = |1s2s^22p_{1/2}^2 J = 1/2\rangle$, and the final state is $|\Psi_g\rangle = |1s^22s^22p_{1/2} J = 1/2\rangle$. For the radiative recombination including the DR resonance in Be-like ions, the asymmetric parameter β , which determines the polarization and the angular distribution of radiation, is expressed as [48]

$$\beta = \frac{\sqrt{2} \operatorname{Re}(a^*b) + |b|^2}{|a|^2 + |b|^2},\tag{4}$$

where *a* and *b* denote the reduced transition matrix elements from the initial channels Ψ_s and Ψ_d , respectively:

$$a = \langle \Psi_g || T^k || \Psi_s \rangle + \frac{\langle \Psi_g || T^k || \Psi_a \rangle \langle \Psi_a || V_{ee} || \Psi_s \rangle}{E - E_r + i\Gamma/2}$$
$$= a_{\rm RR} e^{-i\delta_s} + a_{\rm DR} e^{-i\delta_s}, \tag{5}$$

$$b = \langle \Psi_g || T^k || \Psi_d \rangle = b_{\rm RR} e^{-i\delta_d}.$$
 (6)

 T^k denotes the transition dipole operator in velocity form [49,50], and V_{ee} denotes the electron-electron interaction, including the generalized Breit interaction. δ_s and δ_d denote the phase shifts of the continuum *s* and $d_{3/2}$ states, respectively. $a_{\rm RR}$ and $b_{\rm RR}$ denote the RR transition amplitudes, which are real numbers, whereas $a_{\rm DR}$ is a complex number, in general, which is imaginary at the resonant energy. We first calculated the DR resonant energy E_r and the lifetime Γ and then *a*, *b*, and β . Then, we obtained polarization of x rays emitted at 90° with respect to the electron beam as follows:

$$P = \frac{3\beta}{4+\beta}.\tag{7}$$

Notably, the initial state $|\Psi_s\rangle$ cannot generate any polarization, and only $|\Psi_d\rangle$ can give a nonzero degree of linear polarization due to an uneven m_l distribution. However, in the DR process of present interest, only $|\Psi_s\rangle$ is allowed as an initial state due to the parity and angular momentum conservation in the resonance dielectronic capture process. A nonzero polarization of the DR x rays is, thus, possible only from the cross term between the different partial waves, i.e., *s* and *d* waves, which cannot be assessed by the total cross section for which the cross term vanishes [26].

Figure 3(a) depicts the degree of linear polarization as a function of atomic number (Z). If we consider only the DR process without interference, the degree of linear polarization is exactly zero independent of Z, as shown by the black line in the figure. Meanwhile, if only the RR process is considered, $P \approx 0.6$ with little dependence on Z, as



FIG. 3. (a) Atomic number dependence of the degree of linear polarization for pure RR (magenta), pure DR (black), and total recombination including interference between DR and RR (blue). The red solid curve represents $P_{\text{int}}^{\text{DR}}$ defined in Eq. (8). $P_{\text{int}}^{\text{DR}}$ calculated by assuming $\Delta \delta = \delta_s - \delta_d = 0$ is plotted by the dashed red line, which overlaps with the black line at P = 0. The present experimental result is plotted by the red square. (b) Atomic number dependence of $|a_{\text{DR}}/b_{\text{RR}}|$ (red, left axis) and $\Delta \delta$ (blue, right axis).

shown by the magenta curve in the figure. The degree of linear polarization for the total recombination x rays, i.e., RR + DR, obtained by considering interference between the *s* and *d* waves is plotted by the blue curve in Fig. 3(a). To clarify the interference effect, $P_{\text{int}}^{\text{DR}}$ defined in the following formula is plotted by the red solid curve:

$$P_{\text{int}}^{\text{DR}} = \frac{(\sigma_{\parallel}^{\text{DR}+\text{RR}} - \sigma_{\parallel}^{\text{RR}}) - (\sigma_{\perp}^{\text{DR}+\text{RR}} - \sigma_{\perp}^{\text{RR}})}{(\sigma_{\parallel}^{\text{DR}+\text{RR}} - \sigma_{\parallel}^{\text{RR}}) + (\sigma_{\perp}^{\text{DR}+\text{RR}} - \sigma_{\perp}^{\text{RR}})}, \qquad (8)$$

where σ_{\parallel} and σ_{\perp} represent the differential cross sections for a photon parallel or perpendicular, respectively, to the electron beam direction when it is emitted at 90° with respect to the electron beam. The finite experimental electron beam width was considered in σ by convoluting a Gaussian profile with a 75 eV width. σ^{DR+RR} represents the cross sections for the total recombination including DR and RR with their interference, whereas σ^{RR} represents those for pure RR. If there was no interference effect [i.e., $\sigma^{\text{DR}+\text{RR}} = \sigma^{\text{DR}} + \sigma^{\text{RR}}$ in Eq. (8)], $P_{\text{int}}^{\text{DR}}$ equals exactly zero. Thus, the difference between the black line and red solid curve corresponds to the interference effect arising from the cross term between the s and d waves. As confirmed in the figure, the interference effect on the polarization is small at the lower Z region, but it rapidly increases with Z and reaches about 0.3 at $Z \sim 90$.

In general, DR is orders of magnitude stronger than RR. Thus, the interference between DR and RR was simply ignored in the previous theoretical studies [8,9,51,52]. In addition, in the present system, the DR resonance shows clear symmetric profiles as confirmed in the middle panel

in Fig. 1, suggesting that the interference effect can be ignored. However, if we ignore the RR term (i.e., b = 0), Eq. (4) shows that $\beta = 0$; thus, P = 0. At the resonant energy, if the difference of the phase shifts $\Delta \delta =$ $\delta_s - \delta_d = 0$, $P_{\text{int}}^{\text{DR}}$ defined in Eq. (8) is also zero, as confirmed by the red dashed line in Fig. 3(a). Therefore, the nonzero DR polarization originates from the interference between the DR and RR with the nonzero phase-shift difference. As shown in Fig. 3(b), the relative strength of DR (a_{DR}) with respect to RR (b_{RR}) decreases with increasing Z and becomes comparable in the higher Zregion. This is due to the synergy effect of the Z dependences of (i) the RR cross section, which increases with Z, and (ii) the resonance width, which becomes broader with Z by radiation damping. In addition, the phase-shift difference $\Delta \delta$ increases within π with Z. Thus, the polarization has a strong Z dependence.

The present experimental result for Pb (Z = 82) is plotted by the red square in Fig. 3(a), which should be compared with the red solid curve, as the experimental value was obtained by subtracting the RR contribution. As seen in the figure, a reasonable agreement is found between the experiment and theory, which reveals the large interference effect. Because the interference effect on the polarization is not peculiar to the present system, the effect can be examined in any DR process of any charge state, in principle. However, the polarization of DR x rays is generally not well known even without considering the interference with RR; thus, the effect is difficult to examine when the process is polarized regardless of the interference. Therefore, the present system is suitable to examine the effect, as the degree of polarization is exactly zero if there was no interference.

Finally, we discuss why the large interference effect on polarization exists for the resonance whose nearly symmetric resonance profile usually suggests a small interference effect. The differential recombination cross section at 90° is given by $[d\sigma(E)/d\Omega]_{90°} = \sigma_t(E)[1-\beta/2]/4\pi$, where σ_t is the total cross section [53]. The asymmetric parameter β defined in Eq. (4) is also energy dependent, and its cross term is proportional to $|a_{\rm DR}b_{\rm RR}|[(E-E_r)\cos(\Delta\delta)+$ $\Gamma \sin(\Delta \delta/2)]/[(E-E_r)^2 + \Gamma^2/4]$. However, when $\Delta \delta \approx$ $\pi/2$, as shown in Fig. 3(b), β is symmetric regarding the resonance energy even if it is not zero. Thus, the asymmetry in the differential cross section is mainly determined by σ_t , to which only the interference between the DR and RR with the same partial waves contributes. Consequently, a nearly symmetric resonance profile in the total or differential cross sections does not always mean a small interference effect on polarization, which can provide more information.

Conclusions.—In summary, using the Tokyo-EBIT and the EBIT-CC, a new hard x-ray Compton polarimeter, we experimentally found that the $[1s^22s^22p_{1/2}]_{1/2} - [1s2s^22p_{1/2}^2]_{1/2}$ x-ray transition in B-like Pb⁷⁷⁺ emitted in DR of Pb⁷⁸⁺ is strongly polarized, although it is an

intrinsically unpolarized J = 1/2 to 1/2 transition. Theoretically, we found that the quantum interference with RR, whose effect on the polarization of DR x rays has never been considered, can clearly explain the experimentally observed strong polarization. In particular, we found that only the cross term between different partial waves is responsible for the polarization, whereas that between the same partial waves mainly determines the resonance profile in the cross section; i.e., the polarization and the profile are both affected by the interference but in different manners through different cross terms. Thus, we have shown that, in general, the interference can make large modifications to the degree of polarization even for resonances whose nearly symmetric profile suggests a small interference effect. In addition, contrary to the present system, the interference between the same partial waves generally affects polarization. The previous studies [6-9] should, thus, be revisited by considering this missed interference effect.

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