Dominance of the Breit Interaction in the X-Ray Emission of Highly Charged Ions Following Dielectronic Recombination

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The Breit interaction typically appears as a—more or less small—correction to the Coulomb repulsion acting among the electrons. We here propose two x-ray measurements on the angular distribution and linear polarization of the $1s2s^22p_{1/2}\, J=1 \rightarrow 1s^22s^2\, J=0$ electric-dipole radiation of high-Z, beryllium-like ions, following the resonant electron capture into initially lithium-like ions, for which the Breit interaction strongly dominates the Coulomb repulsion and leads to a qualitative change in the expected x-ray emission pattern. The proposed measurements are feasible with present-day x-ray detectors and may serve a stringent test on relativistic corrections to the electron-electron interaction in the presence of strong fields.

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The importance of relativistic effects on the spectra and properties of most elements from the periodic table has been known for many decades. Just a few years after Dirac had derived his famous wave equation for massive spin-1/2 particles in 1928 [1], Gregory Breit worked out the theory of two-electron interactions to calculate the fine-structure of atomic helium [2], a work which was found "fundamental to atomic physics and modern quantum electrodynamics" [3]. Since this seminal work, there has been continuous interest in further exploring and analyzing the relativistic contributions to the electron-electron (e-e) interaction, or the Breit interaction, in a large variety of nuclear, atomic, and molecular processes. Today, the full Breit interaction or at least parts of it are incorporated not only in (most) atomic computations [4,5] but also in hadron physics [6], heavy-ion collisions [7], atomic and molecular scattering [8], or even modern quantum chemistry [9].

While there were the earlier "beliefs" to apply the Breit interaction only in first-order perturbation theory [10], it can be derived rigorously within the framework of quantum electrodynamics (QED) as the retardation in the exchange of a virtual photon between the (bound) electrons [11,12]. Since the late 1980s, therefore, the Breit interaction has been utilized in a systematic fashion and tested to a very high accuracy, especially in two-electron QED calculations of helium- and lithium-like ions [13,14]. Although these computations are typically in excellent agreement with experiments [15,16], more often than not the Breit interaction gives rise to only a moderate shift in the binding energies of the electrons. Apart from energy shifts (in various photon spectra), the effects of the Breit interaction have been explored also for (spin-)forbidden transitions [17,18], the autoionization [19] and dielectronic recombination of highly charged ions [20,21], their radiative stabilization [22,23], or even the electron-impact ionization [24] for which an almost 50% increase was found for hydrogen-like uranium. In all these examples, however, the Breit interaction occurs as a "correction" to the (usually dominant) Coulomb repulsion, and great spectroscopic effort is often required then to separate its influence from other one- and many-particle effects in the observed spectra.

A remarkably strong (almost 100%) effect of the Breit interaction was recently seen experimentally by Nakamura and coworkers [25] for the dielectronic recombination of initially lithium-like ions. This large effect on the dielectronic recombination rate arises from the selective excitation of the $1s2s^22p_{1/2}\, J=1$ resonance that occurs well isolated with regard to other inner-shell excitations for all medium and heavy elements. In this Letter, we here propose x-ray measurements on the angular distribution and linear polarization of the $1s2s^22p_{1/2}\, J=1 \rightarrow 1s^22s^2\, J=0$ electric-dipole (E1) emission. For the characteristics of this line, it is shown below that the Breit interaction dominates the Coulomb repulsion among the electrons and leads to a qualitative change in the expected x-ray emission pattern for ions with $Z \gtrsim 73$. The proposed measurements are feasible with present-day x-ray detectors: While experiments on (i) the angular distribution of the $1s2s^22p_{1/2}\, J=1 \rightarrow 1s^22s^2\, J=0$ x-ray line are suitable especially for storage rings, (ii) the linear polarization of the emitted line can be observed at both storage rings and electron-beam ions traps, since it requires a polarization measurement at only one observation angle, and preferentially for $\theta = 90°$. An angle and polarization resolved measurement of the $1s2s^22p_{1/2}\, J=1 \rightarrow 1s^22s^2\, J=0$ x-ray line, following the dielectronic recombination of initially lithium-like ions, may therefore provide a stringent test of how relativistic corrections to the e-e interaction act in a strong electromagnetic field.

Theoretical background.—To understand the role of the Breit interaction on the x-ray emission of inner-shell excited ions, let us start from an isolated resonance that is formed by electron capture and (subsequently) decays under photon or electron emission. For a sufficiently long lifetime of this resonance, we can describe its formation and decay independently as a two-step process and assign to this resonance the intermediate state $|\alpha_d J_d\rangle$ with (total) angular momentum J_d, and where α_d represents all additional quantum numbers that are needed for its unique specification. If this intermediate state is formed in a collision of a beam of ions with unpolarized atoms or electrons, the magnetic sublevel population of $|\alpha_d J_d\rangle$ will in general not be distributed statistically but becomes aligned along the beam axis and with an equal population only for magnetic sublevels with the same modulus $|M_d|$.

Using the density-matrix theory [26], the magnetic sublevel population of excited ions and atoms is described most naturally in terms of the alignment parameters $\mathcal{A}_k(\alpha_d J_d)$. For the resonant capture of an (unpolarized) electron, these parameters can be expressed in the form [23]

$$\mathcal{A}_k(\alpha_d J_d) = \frac{N}{8\pi[J_0]} \sum_{ll'jj'} (-1)^{J_d+J_0-1/2}[l, l', j, j']^{1/2} \langle \alpha_d J_d \| V \| \alpha_0 J_0, lj{:}J_d\rangle \langle \alpha_d J_d \| V \| \alpha_0 J_0, l'j'{:}J_d\rangle^* \langle l0l'0|k0\rangle \times \begin{Bmatrix} j & l & 1/2 \\ l' & j' & k \end{Bmatrix} \begin{Bmatrix} j & J_d & J_0 \\ J_d & j' & k \end{Bmatrix}, \quad (1)$$

where N denotes a normalization constant, $\langle l0l'0|k0\rangle$, a Clebsch-Gordan coefficient, $[a, b, \ldots] \equiv (2a+1)\times(2b+1)\ldots$ and where the standard notation for the Wigner 6-j symbols has been utilized. The reduced matrix elements $\langle \alpha_d J_d \| V \| \alpha_0 J_0, lj{:}J_d\rangle$ refer moreover to the e-e interaction that leads to the formation of the resonance $|\alpha_d J_d\rangle$ in course of the collision, starting with an ion in the (initial) state $|\alpha_0 J_0\rangle$ and a free electron with orbital and total angular momenta, l and j, respectively. In the relativistic theory, as appropriate for medium and heavy elements, the (frequency-dependent) e-e interaction [4]

$$V = V^C + V^B = \sum_{i<j}\left(\frac{1}{r_{ij}} - (\boldsymbol{\alpha}_i \cdot \boldsymbol{\alpha}_j)\frac{\cos(\omega r_{ij})}{r_{ij}} + (\boldsymbol{\alpha}_i \cdot \boldsymbol{\nabla}_i)(\boldsymbol{\alpha}_j \cdot \boldsymbol{\nabla}_j)\frac{\cos(\omega r_{ij}) - 1}{\omega^2 r_{ij}}\right) \quad (2)$$

contains both, the instantaneous Coloumb repulsion (first term) and the Breit interaction, i.e., the magnetic and retardation contributions (second and third term). In this expression, moreover, ω is the frequency of the virtual photon and $\boldsymbol{\alpha}_i$ the vector of Dirac matrices as associated with the ith particle.

The alignment parameters (1) can be expressed also in terms of the partial cross sections σ_{M_d} for the population of the magnetic substates $|\alpha_d J_d M_d\rangle$. For example, the second-order parameter

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

fully describes the population of all excited ions with total angular momentum $J_d = 1$. The subsequent decay of these ions then lead to the emission of one (or several) photons, until their ground state is reached. Therefore, the angular and polarization properties of the characteristic photons are closely related to the parameter $\mathcal{A}_2$. A well-known example is the angular distribution

$$W(\theta) \propto 1 + \frac{\mathcal{A}_2}{\sqrt{2}} P_2(\cos\theta), \quad (4)$$

of every $|\alpha_d J_d = 1\rangle \to |\alpha_f J_f = 0\rangle$ E1 line in the projectile (rest) frame of the ion, and where P_2 denotes the Legendre polynomial and θ the angle between the momenta of the (incoming) electron and the emitted photon. A slightly more complex expression can be derived also for the angular distribution of the linear polarization of such $|1\rangle \to |0\rangle$ transitions [26], but this simplifies considerably for particular angles of the photon emission. If, for instance, the polarization is observed perpendicular to the electron or ion beam, the degree of polarization becomes

$$P_L = \frac{-3\sqrt{2}\mathcal{A}_2}{4 - \sqrt{2}\mathcal{A}_2}, \quad (5)$$

and can be easily determined experimentally by measuring the intensities of the light that is linearly polarized in parallel and perpendicular to the reaction plane: $P_L = (I_{0^\circ} - I_{90^\circ})/(I_{0^\circ} + I_{90^\circ})$.

Results and discussion.—Formulas (4) and (5) can be applied especially to the $1s2s^2 2p_{1/2}\, J = 1$ resonance state of beryllium-like (heavy) ions following the dielectronic recombination of (initially) lithium-like projectiles. For these $J = 1$ resonances, only the single alignment parameter $\mathcal{A}_2$ is nonzero due to the coupling of the angular momenta and can be calculated by using the (reduced) amplitudes $\langle \alpha_d J_d \| V \| \alpha_0 J_0, lj{:}J_d\rangle$ in Eq. (1). Moreover, since this resonance is well separated by several (hundred) eV from all other resonances with a $1s$ hole for the medium and heavy elements along the beryllium isoelectronic sequence [27], its formation and decay can be investigated independently. In the present work, we have applied multiconfiguration Dirac-Fock wave functions [23,28] to evaluate the transition amplitudes and alignment parameters from Eq. (1) in different approximations and with an accuracy of about 5%–10% in order to explore the effects of the (frequency-dependent) Breit interaction upon the angular distribution and linear polarization of the emitted photons.

Table I displays the alignment parameter $\mathcal{A}_2$ of the $1s2s^22p_{1/2}\,J=1$ resonance for six ions with $53 \le Z \le 92$; apart from the Coulomb repulsion, the full e-e interaction "Coulomb + Breit" has been taken into account in the computations. While the Coulomb repulsion alone results in a large positive alignment of all ions, and this is only slightly reduced from 0.699 for $Z=53$ to 0.468 for $Z=92$, a remarkable decrease of the alignment is obtained for the full e-e interaction in the transition amplitudes, changing its sign at about $Z \approx 73$. In fact, the value of $\mathcal{A}_2 = 0.699$ for $Z=53$ corresponds to an almost 100% population of the $M_J = \pm 1$ sublevels [cf. Eq. (3)] which can be understood in the nonrelativistic limit to the dielectronic recombination. In this limit, the magnetic sublevels of the $^3P_1$ term can be expressed as (antisymmetrized) products of some spatial times a spin part of the overall wave functions: $|1s2s^22p\,^3P_1, M_J\rangle = \sum_{M_L M_S} |L=1, M_L\rangle |S=1, M_S\rangle \langle L, M_L, S, M_S | J=1, M_J\rangle$. Since in a "spin-independent" collision, the (orbital) angular momentum is transferred perpendicular to the collision (i.e., the quantization) axis, only substates with $M_L = 0$ will be populated. In this limit, therefore, only the term with $M_L = 0$ occurs in the summation above, and this implies $M_J = \pm 1$ since, for $M_J = 0$, the Clebsch-Gordan coefficient vanishes identically ($\langle 1010|10\rangle = 0$). With increasing nuclear charge, the spin-dependent terms in the e-e interaction become more important, and for $Z \gtrsim 73$, in fact, the population of the $M_J = 0$ sublevel already dominates as seen from the negative value of the alignment for the $1s2s^22p_{1/2}\,J=1$ resonance.

The large differences in the alignment $\mathcal{A}_2$ for calculations with and without the Breit interaction in the transition amplitudes give rise also to a very different angular distribution of the $1s2s^22p_{1/2}\,J=1 \to 1s^22s^2\,J=0$ x-ray photons as shown in Fig. 1. While for only the Coulomb interaction, the photons are dominantly emitted along the beam axis under $\theta = 0$ and 180 degrees, a qualitative change in the emission pattern occurs if, in addition, the Breit interaction is taken into account. To the best of our knowledge, this is the first physics case where the incorporation of the Breit terms leads to a completely different behavior in comparison to what is expected from the Dirac-Coulomb theory for the many-electron ions. For an increasing nuclear charge of the ions, the angular distribution of the $1s2s^22p_{1/2}\,J=1 \to 1s^22s^2\,J=0$ emission is predicted to become first isotropic for $Z \approx 73$ and then more and more pronounced perpendicular to the ion beam. For beryllium-like bismuth and beyond (cf. right panel of Fig. 1 for $Z \gtrsim 83$), a perpendicular photon emission is clearly favored. Therefore, because the $1s2s^22p_{1/2}\,J=1 \to 1s^22s^2\,J=0$ line dominates in the x-ray spectrum by at least four orders over other spin-forbidden transitions of the $1s2s^22p_{1/2}\,J=1$ [29], it provides an ideal candidate to explore the details of the Breit interaction in relativistic ion-electron collisions.

A similar remarkable effect of the Breit interaction is predicted also for the linear polarization of the photons. Since the degree of this polarization is determined uniquely by the parameter $\mathcal{A}_2$, it provides an alternative access to the population of the magnetic sublevels. For a perpendicular photon emission in the projectile frame, Fig. 2 displays the degree of linear polarization as a function of the nuclear charge. For only the Coulomb repulsion, the photons are (linearly) polarized perpendicular to the reaction plane ($P \gtrsim -1$), almost independent of the charge Z. Apart from a strong depolarization effect on the photon emission due to the Breit terms in the e-e interaction, a qualitative change occurs again for $Z \approx 73$: While the $1s2s^22p_{1/2}\,J=1 \to 1s^22s^2\,J=0$ line gets first unpolarized at this particular charge, it becomes rapidly linearly

TABLE I. Alignment parameter $\mathcal{A}_2$ of the $1s2s^22p_{1/2}\,J=1$ resonance following the electron capture into (initially) lithium-like heavy ions with different nuclear charge. Calculations with only the Coulomb repulsion in the transition amplitudes are compared with a full treatment of the e-e interaction (Coulomb + Breit).

Nuclear charge Z	Coulomb	Coulomb + Breit
53	0.699	0.427
60	0.684	0.270
67	0.652	0.112
74	0.612	−0.032
83	0.528	−0.181
92	0.468	−0.314

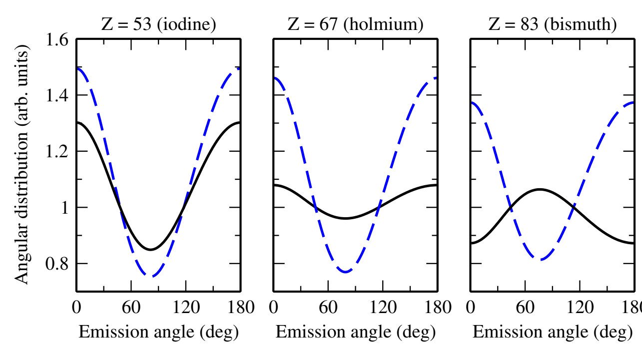

FIG. 1 (color online). Angular distribution of the $1s2s^22p_{1/2}\,J=1 \to 1s^22s^2\,J=0$ dipole emission for beryllium-like iodine (left panel), holmium (middle), and bismuth ions (right), following the resonant electron capture into (initially) lithium-like projectiles. Calculations with only the Coulomb repulsion in the transition amplitudes (blue dashed lines) are compared with a full account of the e-e interaction (Coulomb + Breit; black solid line). The computations were done for the laboratory frame in which the electrons are at rest.

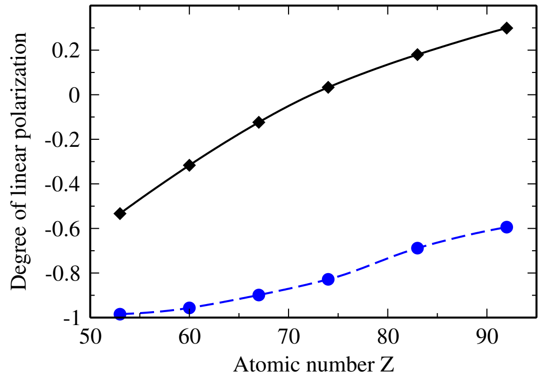

FIG. 2 (color online). Degree of linear polarization of the $1s2s^22p_{1/2}\,J=1 \rightarrow 1s^22s^2\,J=0$ emission as observed perpendicular to the ion beam, $\theta = 90°$, within the rest frame of the ions (projectile frame). Calculations with only the Coulomb repulsion in the transition amplitudes (blue dashed line) are compared with the full Coulomb + Breit interaction (black solid line).

polarized within the reaction plane if the nuclear charge is further increased beyond $Z = 80$.

The proposed measurements on the $1s2s^22p_{1/2}\,J = 1 \rightarrow 1s^22s^2\,J=0$ x-ray line are feasible with the present-day facilities, both at heavy-ion storage rings and electron-beam ions trap devices. At the GSI storage ring ESR, for example, accurate measurements on the linear polarization of x rays emitted following the radiative electron capture into bare uranium ions have been performed recently in very good agreement with earlier predictions [30,31]. At most ion traps, in contrast, the observation of the x-ray radiation is possible only in perpendicular to the electron beam. As seen from Fig. 2, however, a polarization-sensitive measurement under $\theta = 90°$ (in the rest frame of the ions) will allow a simple test of the predictions for high-Z, few-electron ions.

In conclusion, the influence of the Breit interaction on the angular distribution and linear polarization of the x-ray emission from highly charged ions has been analyzed, following the dielectronic recombination of (initially) lithium-like ions. For the radiative stabilization of the $1s2s^22p_{1/2}\,J=1$ resonance, in particular, a qualitative change in the emission pattern is predicted for $Z \approx 73$ if, in addition to the Coulomb repulsion, the full e-e interaction "Coulomb + Breit" is taken into account. Both the formation as well as the E1 decay of the $1s2s^22p_{1/2}\,J=1$ can be well resolved with present-day (position-sensitive) x-ray detectors with a resolution of $\lesssim 50$ eV and, hence, provide a very clean and promising route for studying the e-e interaction in relativistic collisions and the presence of strong fields.

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