Angular correlations in the two-photon decay of heliumlike heavy ions

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The two-photon decay of heavy, helium-like ions is investigated based on second-order perturbation theory and Dirac's relativistic equation. Special attention has been paid to the angular emission of the two photons (i.e., how the angular correlation function depends on the shell structure of the ions in their initial and final states). Moreover, the effects from the (electric and magnetic) nondipole terms in the expansion of the electron-photon interaction are discussed. Detailed calculations have been carried out for the two-photon decay of the $1s2s\,^1S_0$, $1s2s\,^3S_1$, and $1s2p\,^3P_0$ states of helium-like Xe^{52+}, Au^{77+}, and U^{90+} ions.

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I. INTRODUCTION

Owing to the recent advances in heavy-ion accelerator and trap facilities as well as in detection techniques, new possibilities arise to study the electronic structure of simple atomic systems in strong (nuclear) Coulomb fields. Relativistic, quantum electrodynamics (QED), or even parity nonconservation (PNC) effects, which are difficult to isolate in neutral atoms, often become enhanced in high-Z, few-electron ions. In order to improve our understanding of these fundamental interactions, a number of experiments have been recently carried out on the characteristic photon emission from heavy ions [1–3]. Apart from the one-photon bound-bound transitions, the two-photon decays of metastable ionic states have also attracted much interest since the analysis of its properties may reveal unique information about the complete spectrum of the ion, including negative energy (positron) solutions of Dirac's equation. Until now, however, most two-photon studies were focused on measuring the total and energy-differential decay rates [4–10] which were found in good agreement with theoretical predictions, based on relativistic calculations [11–20]. In contrast, much less attention has been previously paid to the angular and polarization correlations between the emitted photons. The first two-photon correlation studies with heavy ions are likely to be carried out at the GSI facility in Darmstadt, where significant progress has been recently made in development of solid-state, position-sensitive x-ray detectors [21,22]. By means of these detectors, a detailed analysis of the angular and polarization properties of two-photon emission will become possible and will provide more insights into the electronic structure of heavy, few-electron ions.

Despite the recent interest in two-photon coincidence studies, not much theoretical work has been done so far to explore the photon-photon correlations in the decay of heavy atomic systems. While some predictions are available for the hydrogen-like [23,24] and neutral atoms [14], no systematic angular (and polarization) analysis was performed for the helium-like ions which are the most suitable candidates for two-photon investigations in high-Z domain. In the present work, therefore, we apply the second-order perturbation theory based on Dirac's equation to investigate the γ-γ angular correlations in the decay of two-electron systems. The basic relations of such a (relativistic) perturbation approach will be summarized in Sec. II. In particular, here we introduce the second-order transition amplitude that describes a bound-state transition under the simultaneous emission of two photons. The evaluation of these (many-body) amplitudes within the framework of the independent particle model (IPM) is thereafter discussed in Sec. II B. Within this approximation that is particularly justified for the high-Z regime [25–27], the photon-photon correlation function from Sec. II C can be traced back to the one-electron matrix elements. This reduction enables us to implement the well-established Green's function as well as finite basis set methods [28–30] and to calculate the correlation functions for the $1s2s\,^1S_0 \to 1s^2\,^1S_0$, $1s2s\,^3S_1 \to 1s^2\,^1S_0$, and $1s2p\,^3P_0 \to 1s^2\,^1S_0$ transitions in helium-like Xe^{52+}, Au^{77+}, and U^{90+} ions. Results from these computations are displayed in Sec. III and indicate a strong dependence of the photon emission pattern on the symmetry and parity of initial and final ionic states. Moreover, the significant effects that arise due the higher-multipole terms in the expansion of the electron-photon interaction are also discussed in the context of angular correlation studies. Finally, a brief summary is given in Sec. IV.

II. THEORETICAL BACKGROUND

A. Second-order transition amplitude

Since the second-order perturbation theory has been frequently applied in studying two-photon decay, here we may restrict ourselves to a short compilation of the basic formulas relevant for our analysis and refer for all further details to the literature [11,12,14,16,18,19,24–26]. Within the relativistic framework, the second-order transition amplitude for the emission of two photons with wave vectors $\boldsymbol{k}_i$ ($i = 1, 2$) and polarization vectors $\boldsymbol{u}_{\lambda_i}$ ($\lambda = \pm 1$) is given by

$$\mathcal{M}_{fi}(M_f, M_i, \lambda_1, \lambda_2) = \sum_{\gamma_\nu J_\nu M_\nu} \frac{\langle \gamma_f J_f M_f | \hat{\mathcal{R}}^\dagger(\boldsymbol{k}_1, \boldsymbol{u}_{\lambda_1}) | \gamma_\nu J_\nu M_\nu \rangle \langle \gamma_\nu J_\nu M_\nu | \hat{\mathcal{R}}^\dagger(\boldsymbol{k}_2, \boldsymbol{u}_{\lambda_2}) | \gamma_i J_i M_i \rangle}{E_\nu - E_i + \omega_2} + \sum_{\gamma_\nu J_\nu M_\nu} \frac{\langle \gamma_f J_f M_f | \hat{\mathcal{R}}^\dagger(\boldsymbol{k}_2, \boldsymbol{u}_{\lambda_2}) | \gamma_\nu J_\nu M_\nu \rangle \langle \gamma_\nu J_\nu M_\nu | \hat{\mathcal{R}}^\dagger(\boldsymbol{k}_1, \boldsymbol{u}_{\lambda_1}) | \gamma_i J_i M_i \rangle}{E_\nu - E_i + \omega_1}, \quad (1)$$

where $|\gamma_i J_i M_i\rangle$ and $|\gamma_f J_f M_f\rangle$ denote the (many-electron) states with well-defined total angular momenta $J_{i,f}$ and their projections $M_{i,f}$ of the ions just before and after their decay, and $\gamma_{i,f}$ all the additional quantum numbers as necessary for a unique specification. The energies of these states, E_i and E_f, are related to the energies $\omega_{1,2} = ck_{1,2}$ of the emitted photons by the energy conservation condition,

$$E_i - E_f = \hbar\omega_1 + \hbar\omega_2. \quad (2)$$

Using this relation, it is convenient to define the so-called energy sharing parameter $y = \omega_1/(\omega_1 + \omega_2)$ (i.e., the fraction of the energy which is carried away by the first photon).

In Eq. (1), moreover, $\hat{\mathcal{R}}$ is the transition operator that describes the interaction of the electrons with the electromagnetic radiation. In velocity (Coulomb) gauge for the coupling of the radiation field this operator can be written as a sum of one-particle operators:

$$\hat{\mathcal{R}}(\boldsymbol{k}, \boldsymbol{u}_\lambda) = \sum_m \boldsymbol{\alpha}_m \mathcal{A}_{\lambda,m} = \sum_m \boldsymbol{\alpha}_m \boldsymbol{u}_\lambda e^{i\boldsymbol{k}\cdot\boldsymbol{r}_m}, \quad (3)$$

where $\boldsymbol{\alpha}_m = (\alpha_{x,m}, \alpha_{y,m}, \alpha_{z,m})$ denotes the vector of the Dirac matrices for the m-th particle and $\mathcal{A}_{\lambda,m}$ the vector potential of the radiation field. To further simplify the second-order transition amplitude (1) for practical computations, it is convenient to decompose the potential $\mathcal{A}_{\lambda,m}$ into spherical tensors (i.e., into its electric and magnetic multipole components). For the emission of the photon in the direction $\hat{\boldsymbol{k}} = (\theta, \phi)$ with respect to the quantization (z) axis such a decomposition reads [31]

$$\boldsymbol{u}_\lambda e^{i\boldsymbol{k}\cdot\boldsymbol{r}} = \sqrt{2\pi} \sum_{L=1}^{\infty} \sum_{M=-L}^{L} \sum_{p=0,1} i^L [L]^{1/2} (i\lambda)^p \hat{a}^p_{LM}(k) D^L_{M\lambda}(\hat{\boldsymbol{k}}), \quad (4)$$

where $[L] \equiv 2L + 1$, $D^L_{M\lambda}$ is the Wigner rotation matrix of rank L and $\hat{a}^{p=0,1}_{LM}(k)$ refer to magnetic ($p = 0$) and electric ($p = 1$) multipoles, respectively.

The multipole decomposition of the photon field in terms of its irreducible components with well-defined transformation properties enables us to simplify the second-order amplitude by employing the techniques from Racah's algebra. Inserting Eqs. (3) and (4) into the matrix element (1) and by making use of the Wigner-Eckart theorem, we obtain

$$\mathcal{M}_{fi}(M_f, M_i, \lambda_1, \lambda_2) = 2\pi \sum_{L_1 M_1 p_1} \sum_{L_2 M_2 p_2} (-i)^{L_1+L_2} [L_1, L_2]^{1/2} (-i\lambda_1)^{p_1} (-i\lambda_2)^{p_2} D^{L_1 *}_{M_1\lambda_1}(\hat{k}_1) D^{L_2 *}_{M_2\lambda_2}(\hat{k}_2) \times \sum_{J_\nu M_\nu} \frac{1}{[J_i, J_\nu]^{1/2}} \Big[\langle J_f M_f L_1 M_1 | J_\nu M_\nu \rangle \langle J_\nu M_\nu L_2 M_2 | J_i M_i \rangle S^{J_\nu}_{L_1 p_1, L_2 p_2}(\omega_2) + \langle J_f M_f L_2 M_2 | J_\nu M_\nu \rangle \langle J_\nu M_\nu L_1 M_1 | J_i M_i \rangle S^{J_\nu}_{L_2 p_2, L_1 p_1}(\omega_1) \Big], \quad (5)$$

where the second-order reduced transition amplitude is given by

$$S^{J_\nu}_{L_1 p_1, L_2 p_2}(\omega_2) = \sum_{\gamma_\nu} \frac{\langle \gamma_f J_f || \sum_m \boldsymbol{\alpha}_m \hat{a}^{p_1 \dagger}_{L_1,m}(k_1) || \gamma_\nu J_\nu \rangle \langle \gamma_\nu J_\nu || \sum_m \boldsymbol{\alpha}_m \hat{a}^{p_2 \dagger}_{L_2,m}(k_2) || \gamma_i J_i \rangle}{E_\nu - E_i + \omega_2}. \quad (6)$$

Here, the summation over the intermediate states formally runs over the complete spectrum of the ions, including a summation over the discrete part of the spectrum as well as the integration over the positive- and negative-energy continua. In practice, such a summation is not a simple task especially when performed over the many-electron states $|\gamma_\nu J_\nu\rangle$. In the next section, therefore, we shall employ the independent particle model in order to express the reduced matrix elements (6) for many-electron ions in terms of their one-electron analogs.

B. Evaluation of the reduced transition amplitudes

As seen from Eqs. (5) and (6), one has first to generate a complete set of many-electron states $|\gamma J\rangle$ in order to calculate the second-order transition amplitude M_{fi}. A number of approximate methods, such as multiconfiguration Dirac-Fock (MCDF) [32,33] and configuration interaction (CI) [15], are known in atomic structure theory for constructing these states. Moreover, the systematic perturbative QED approach in combination with the CI method turned out to be most appropriate for describing both transition probabilities [34,35] and transition energies [36] in highly charged ions. In the high-Z domain, however, the structure of few-electron ions can already be reasonably well understood within the independent particle model (IPM). This model is well justified for heavy species especially, since the interelectronic effects scale with $1/Z$ and, hence, are much weaker than the electron-nucleus interaction [25,26,37]. Within the IPM, which takes the Pauli principle into account, the many-electron wave functions are approximated by means of Slater determinants, built from one-particle orbitals. For this particular choice of the many-electron function, all the (first- and the second-order) matrix elements can be easily decomposed into the corresponding single-electron amplitudes.

For a helium-like system, the decomposition of the reduced amplitude (6) reads

$$S^{J_\nu}_{L_1p_1,L_2p_2}(\omega_2) = -\delta_{J_\nu L_1}[J_i, J_\nu]^{1/2}\sum_{j_\nu}(-1)^{J_i+J_\nu+L_2} \times \begin{Bmatrix} j_\nu & j_0 & J_\nu \\ J_i & L_2 & j_i \end{Bmatrix} S^{j_\nu}_{L_1p_1,L_2p_2}(\omega_2), \quad (7)$$

where the one-electron matrix elements of the (electric and magnetic) multipole field operators are given by

$$S^{j_\nu}_{L_1p_1,L_2p_2}(\omega_2) = \sum_{n_\nu} \frac{\langle n_0 j_0||\boldsymbol{\alpha}\hat{a}^{p_1\dagger}_{L_1}(k_1)||n_\nu j_\nu\rangle\langle n_\nu j_\nu||\boldsymbol{\alpha}\hat{a}^{p_2\dagger}_{L_2}(k_2)||n_i j_i\rangle}{E_\nu - E_i + \omega_2}. \quad (8)$$

We assume here that the "spectator" electron, being in hydrogenic state $|n_0 j_0\rangle$, stays passive in the decay process. Moreover, $|n_i j_i\rangle$, $|n_\nu j_\nu\rangle$, and $|n_f j_f\rangle = |n_0 j_0\rangle$ denote the initial, intermediate, and final states of the "active" electron, correspondingly. The great advantage of formula (7) is that it helps us to immediately evaluate the many-electron transition amplitude (6) in terms of the (one-particle) functions $S^{j_\nu}_{L_1p_1,L_2p_2}(\omega_2)$. The summation over the complete one-particle spectrum that occurs in these functions can be performed by means of various methods. In the present work, we make use of (i) the relativistic Coulomb-Green's function [24,38,39] and (ii) a B-spline discrete basis set [16,20,28–30] to evaluate all the second-order transition amplitudes. Indeed, both approaches yield almost identical results for the angular correlation functions in the two-photon decay of heavy helium-like ions.

C. Differential decay rate

Equation (5) displays the general form of the relativistic transition amplitude for the two-photon decay of many-electron ions. Such an amplitude represents the "building block" for studying various properties of the emitted radiation. For instance, the differential two-photon decay rate can be written in terms of (squared) transition amplitudes as

$$\frac{\mathrm{d}w}{\mathrm{d}\omega_1\mathrm{d}\Omega_1\mathrm{d}\Omega_2} = \frac{\omega_1\omega_2}{(2\pi)^3c^2}\frac{1}{2J_i+1} \times \sum_{M_iM_f}\sum_{\lambda_1\lambda_2}|\mathcal{M}_{fi}(M_f, M_i, \lambda_1, \lambda_2)|^2, \quad (9)$$

if we assume that the excited ions are initially unpolarized and that the spin states of the emitted photons remain unobserved in a particular measurement. As seen from expression (9), the two-photon rate is single differential—owing to the conservation law (2)—in the energy of one of the photons but double differential in the emission angles. Accordingly, its further evaluation requires one to determine the geometry under which the photon emission is considered. Since no particular direction is preferred for the decay of an unpolarized (as well as unaligned) ion, it is convenient to adopt the quantization (z) axis along the momentum of the "first" photon: $\boldsymbol{k}_1||z$. Such a choice of the quantization axis allows us to simplify the rate (9) and to define the angular correlation function:

$$W_{2\gamma}(\theta, y) = 8\pi^2(E_i - E_f) \times \frac{\mathrm{d}w}{\mathrm{d}\omega_1\mathrm{d}\Omega_1\mathrm{d}\Omega_2}(\theta_1 = 0, \phi_1 = 0, \phi_2 = 0), \quad (10)$$

which is characterized (apart from the relative energy y) by the single polar angle $\theta = \theta_2$ of the "second" photon momentum with respect to this axis. In this expression, moreover, the factor $8\pi^2$ arises from the integration over the solid angle $d\Omega_1 = \sin\theta_1 \mathrm{d}\theta_1 \mathrm{d}\phi_1$ of the first photon as well as the integration over the azimuthal angle $\mathrm{d}\phi_2$ of the second photon. In the next section, we shall investigate the dependence of the function $W_{2\gamma}(\theta, y)$ on this opening angle θ for various bound-bound transitions and for a range of (relative) photon energies.

III. RESULTS AND DISCUSSION

With the formalism developed above, we are ready now to analyze the angular correlations in the two-photon decay of helium-like heavy ions. In experiments nowadays, the excited states of these ions can be efficiently populated in relativistic ion-atom collisions. For example, the formation of the metastable $1s2s\ ^1S_0$ state during the inner-shell impact ionization of (initially) lithium-like heavy ions has been studied recently at the GSI storage ring in Darmstadt [40]. The radiative deexcitation of this state can proceed only via the two-photon transition $1s2s\,^1S_0 \rightarrow 1s^2\,^1S_0$ since a single-photon decay to the $1s^2\,^1S_0$ ground state is strictly forbidden by the conservation of angular momentum. Figure 1 displays the photon-photon angular correlation function for this experimentally easily accessible decay of helium-like Xe^{52+}, Au^{77+}, and U^{90+} ions and for the two energy sharing parameters $y = 0.1$ (upper panel) and $y = 0.5$ (lower panel). Moreover, because the radiative transitions in high-Z ions are known to be affected by the higher terms of the electron–photon interaction (3), calculations were performed within both, the exact relativistic theory (solid line) to include all allowed multipole components $(p_1 L_1, p_2 L_2)$ in the amplitude (5) as well as the electric dipole approximation (dashed line), if only a single term with $L_1 = L_2 = 1$ and $p_1 = p_2 = 1$ is taken into account. In the dipole 2E1 approach, as expected, the angular distribution is well described by the formula $1 + \cos^2\theta$ that predicts a symmetric—with respect to the opening angle $\theta = 90^\circ$—emission pattern of two photons. Within the exact relativistic theory, in contrast, an asymmetric shift in the angular correlation function is obtained. As can be deduced from Eqs. (7)–(10), this shift arises from the interference between the leading 2E1 decay channel and higher multipole terms in the electron-photon interaction:

$$W_{2\gamma}(\theta, y) \propto (1+\cos^2\theta)+4\frac{\mathcal{S}_{M1}}{\mathcal{S}_{E1}}\cos\theta+\frac{20}{5}\frac{\mathcal{S}_{E2}}{\mathcal{S}_{E1}}\cos^3\theta+\cdots, \tag{11}$$

where, for the sake of brevity, we have introduced the notation $\mathcal{S}_{Lp} = S^{J_\nu=L}_{Lp,Lp}(\omega_1) + S^{J_\nu=L}_{Lp,Lp}(\omega_2)$. For high-$Z$ domain, the photon emission occurs predominantly in the backward directions if the nondipole terms are taken into account; an effect which becomes more pronounced for the equal energy sharing (cf. bottom panel of Fig. 1). Including the higher multipoles into the photon-photon correlation function, a similar asymmetry was found in the past for the $2s_{2/1} \rightarrow 1s_{1/2}$ decay in hydrogen-like heavy ions both within the nonrelativistic [23] and relativistic [24] theory.

Apart from the singlet $1s2s\,^1S_0$, the formation of the triplet $1s2s\,^3S_1$ state has been also observed in recent collision experiments at the GSI storage ring [9,40]. Although much weaker in intensity (owing to the dominant M1 transition), the two-photon decay of this $1s2s\,^3S_1$ state has attracted recent interest and might provide an important testing ground for symmetry violations of Bose particles [26,41]. The angular correlation between the photons emitted in this $1s2s\,^3S_1 \rightarrow 1s^2\,^1S_0$ (two-photon) decay is displayed in Fig. 2, by comparing again the results from the exact relativistic theory with the 2E1 dipole approximation. As seen from the figure, the photon-photon correlation functions for the $2\,^3S_1 \rightarrow 1\,^1S_0$ transition is much more sensitive with regard to higher multipoles in the electron-photon interaction than obtained for the $2\,^1S_0 \rightarrow 1\,^1S_0$ decay. The strongest nondipole effect can be observed for the equal energy sharing ($y = 0.5$), where the two-photon emission is strictly forbidden within the electric dipole approximation. This suppression of the 2E1 decay is a direct consequence of the exchange symmetry of photons as required by the Bose-Einstein statistics and, hence, a particular case of the Landau-Yang theorem that forbids the decay of vector particles into two photons (cf. Refs. [26,41–43] for further details). In contrast to the 2E1 channel, the E1M2 $2\,^3S_1 \rightarrow 1\,^1S_0$ transition can proceed even if the energies of the two photons are equal. This transition as well as higher multipole terms give rise to a strongly anisotropic correlation function that vanishes for the parallel and back-to-back photon emission and has a maximum at $\theta = 90^\circ$.

Large effects due to the higher multipole contributions to the $1s2s\,^3S_1 \rightarrow 1s^2\,^1S_0$ two-photon transition can be observed not only for the case of equal energy sharing ($y = 0.5$). For the relative energy $y = 0.1$, for example, the photon-photon angular correlation function is found symmetric with regard to $\theta = 90^\circ$ in the electric dipole (2E1) approximation but becomes asymmetric in an exact relativistic theory. In contrast to the decay of the $2\,^1S_0$ state, however, a predominant parallel emission of both photons appears to be more likely if the higher multipoles are taken into account. For the

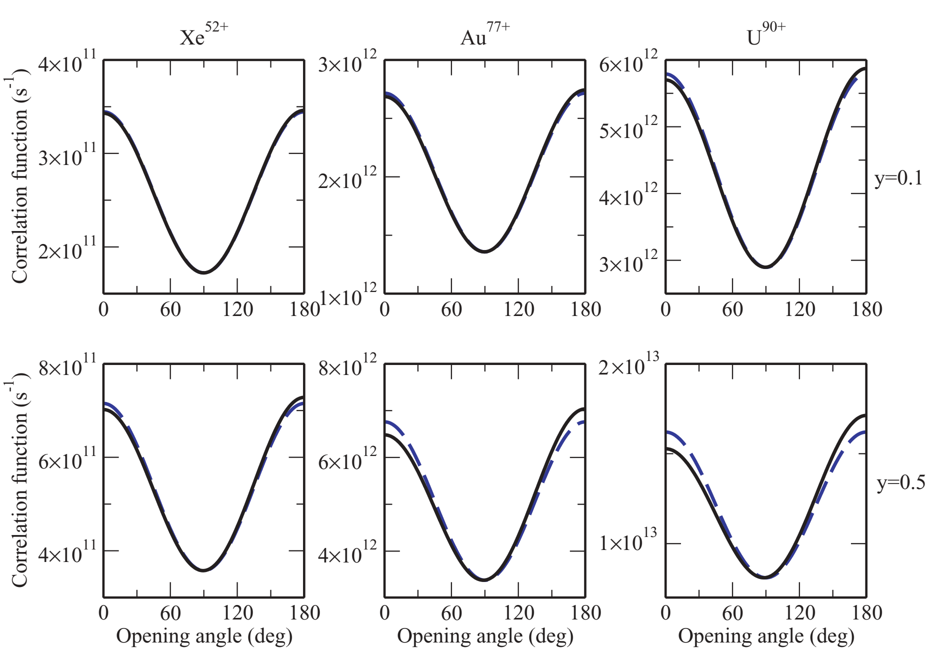

FIG. 1. (Color online) Angular correlation function (10) for the $1s2s\,^1S_0 \rightarrow 1s^2\,^1S_0$ two-photon decay of helium-like xenon, gold, and uranium ions. Calculations obtained within the electric dipole 2E1 approximation (dashed line) are compared with those including all the allowed multipoles (solid line). Results are presented for the relative photon energies $y = 0.1$ (upper panel) and 0.5 (lower panel).

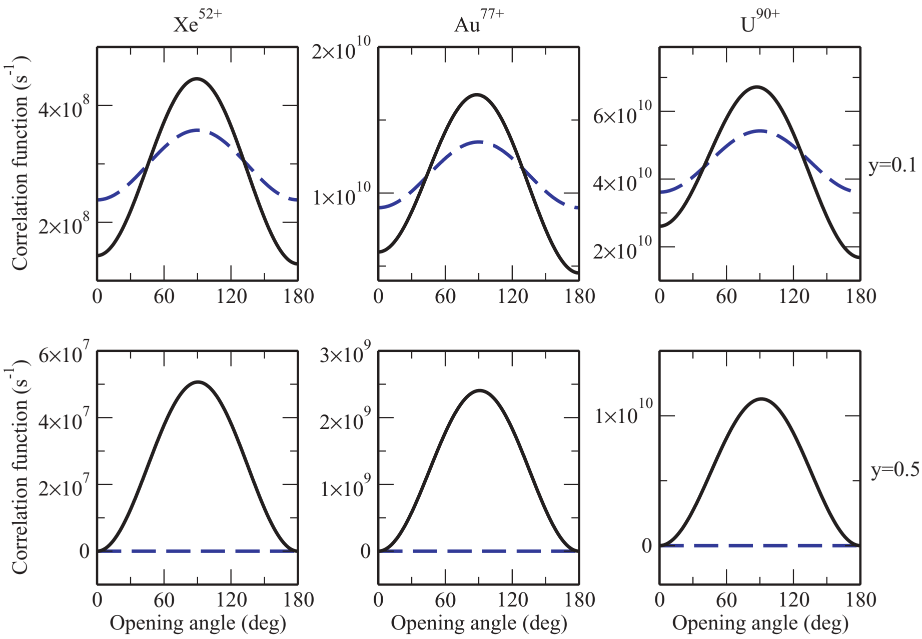

FIG. 2. (Color online) Angular correlation function (10) for the $1s2s\,^3S_1 \to 1s^2\,^1S_0$ two-photon decay of helium-like xenon, gold, and uranium ions. Calculations obtained within the electric dipole 2E1 approximation (dashed line) are compared with those including all of the allowed multipoles (solid line). Results are presented for the relative photon energies $y = 0.1$ (upper panel) and 0.5 (lower panel).

$2\,^3S_1 \to 1\,^1S_0$ two-photon decay of helium-like uranium U^{90+}, for example, the intensity ratio $W_{2\gamma}(\theta = 0^\circ, y = 0.1)/W_{2\gamma}(\theta = 180^\circ, y = 0.1)$ increases from unity within the electric dipole approximation to almost 1.6 in the exact relativistic treatment.

Until now we have discussed the photon-photon correlations in the decay of $1s2s$ (singlet and triplet) helium-like states. Besides these well-studied transitions, recent theoretical interest has been focused also on the $1s2p\,^3P_0 \to 1s^2\,^1S_0$ two-photon decay whose properties are expected to be sensitive to (parity violating) PNC phenomena in atomic systems [27]. Future investigations on such subtle parity nonconservation effects will require first detailed knowledge on the angle (and polarization) properties of two-photon emission as well as the role of nondipole contributions. The angular correlation function (10) for the $2\,^3P_0 \to 1\,^1S_0$ transition is displayed in Fig. 3, again, for two relative photon energies $y = 0.1$ and 0.5 and for the nuclear charges $Z = 54$, 79, and 92. Calculations have been performed both within the exact theory and the ("electric and magnetic") dipole approximation which accounts for the leading E1M1-M1E1 decay channel. As seen from the figure, the emission pattern strongly depends on the energy sharing between the photons. If, for example, one of the photons is more energetic than the second one their parallel emission becomes dominant (cf. upper panel of Fig. 3). In contrast, photons with equal energies (i.e., when $y = 0.5$) are more likely to be emitted back-to-back while the differential rate (9) vanishes identically for $\theta = 0^\circ$. Such a behavior of the photon-photon angular correlation function is caused by the interference between two pathways which

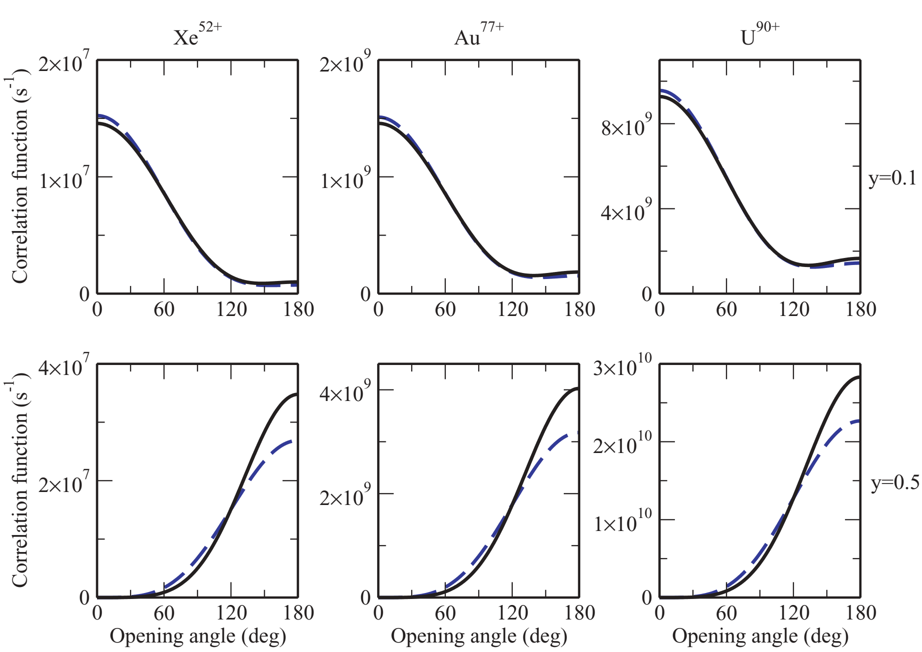

FIG. 3. (Color online) Angular correlation function (10) for the $1s2p\,^3P_0 \to 1s^2\,^1S_0$ two-photon decay of helium-like xenon, gold, and uranium ions. Calculations obtained within the dipole E1M1 approximation (dashed line) are compared with those including all of the allowed multipoles (solid line). Results are presented for the relative photon energies $y = 0.1$ (upper panel) and 0.5 (lower panel).

appear for each multipole component of the $2\,^3P_0 \rightarrow 1\,^1S_0$ transition. For instance, the leading E1M1-M1E1 decay may proceed either via intermediate $n\,^3S_1$ or $n\,^3P_1$ states, thus giving rise to a "double–slit" picture that becomes most pronounced for the equal energy sharing. Simple analytical expression for the angular correlation function which accounts for such a Young-type interference effect can be obtained from Eqs. (7)–(9) as

$$W_{2\gamma}(\theta, y) \propto \sin^4\theta/2|\mathcal{S}_{E1M1}|^2\left(1 + 2(1+2\cos\theta)\frac{\mathcal{S}_{E2M2}}{\mathcal{S}_{E1M1}}\right) + \cos^4\theta/2|\mathcal{D}_{E1M1}|^2\left(1 - 2(1-2\cos\theta)\frac{\mathcal{D}_{E2M2}}{\mathcal{D}_{E1M1}}\right) + \cdots, \quad (12)$$

where, similar as before, we denote $\mathcal{S}_{Lp_1,Lp_2} = S^{J_\nu=L}_{Lp_1,Lp_2}(\omega_1) + S^{J_\nu=L}_{Lp_1,Lp_2}(\omega_2) + S^{J_\nu=L}_{Lp_2,Lp_1}(\omega_2) + S^{J_\nu=L}_{Lp_2,Lp_1}(\omega_1)$ and $\mathcal{D}_{Lp_1,Lp_2} = S^{J_\nu=L}_{Lp_1,Lp_2}(\omega_1) - S^{J_\nu=L}_{Lp_1,Lp_2}(\omega_2) + S^{J_\nu=L}_{Lp_2,Lp_1}(\omega_2) - S^{J_\nu=L}_{Lp_2,Lp_1}(\omega_1)$. Obviously, if the energies of the two photons are equal, $\omega_1 = \omega_2$, the second term in Eq. (12) turns out to be zero and the photon emission is described by the $\sin^4\theta/2$ angular distribution modified by the nondipole terms in the expansion of electron-photon interaction. As seen from the lower panel of Fig. 3, the contribution from these terms becomes more pronounced for the back-to-back photon emission ($\theta = 180^\circ$) where they lead to about a 30% enhancement of the correlation function. It is interesting to note that such an enhancement remains almost constant along the helium isoelectronic sequence for $Z \geqslant 54$ due to similar ($\propto Z^{12}$) scaling of the E1M1 and E2M2 transition probabilities. Therefore, our calculations clearly indicate the importance of higher multipoles for analyzing the photon-photon correlations not only for high-Z domain but also for medium-Z ions.

IV. SUMMARY AND OUTLOOK

In summary, the two-photon decay of heavy, helium-like ions has been investigated within the framework of the relativistic second-order perturbation theory and the independent particle model. In this study, special emphasis was placed on the angular correlations between the emitted photons. A general expression for the photon-photon correlation function was derived that accounts for the complete expansion of the radiation field in terms of its multipole components. Based on solutions of Dirac's equation, this function has been calculated for the two-photon decay of the $1s2s\,^1S_0$, $1s2s\,^3S_1$, and $1s2p\,^3P_0$ states of helium-like Xe^{52+}, Au^{77+}, and U^{90+} ions. As seen from the results obtained, the photon emission pattern appears to be sensitive to the symmetry and parity of the particular excited state as well as to the higher multipole contributions to the electron-photon interaction. The strongest nondipole effects have been identified for the $1s2s\,^3S_1 \rightarrow 1\,^1S_0$ two-photon transition for which the 2E1 decay channel is forbidden owing to symmetrization properties of the system. For the other two transitions, $1s2s\,^1S_0 \rightarrow 1\,^1S_0$ and $1s2p\,^3P_0 \rightarrow 1\,^1S_0$, the higher multipoles of the radiation field typically result in a 10%–30% deviation of the photon-photon correlation function from the (analytical) predictions obtained within the dipole 2E1 approximation. This deviation becomes most apparent for the parallel and back-to-back photon emission and may be observed not only for high-Z but also for medium-Z ions.

The second-order perturbation approach based on the independent particle model, used in the present calculations, is appropriate for the analysis of forthcoming experimental studies on the two-photon transitions between the $^{2s+1}L_J$ excited and the ground states of helium-like, heavy ions. Besides these spontaneous decays, whose energies usually reach 100 keV, induced $J = 0 \rightarrow J = 0 + 2\gamma$ transitions between excited states are also likely to be explored at the GSI ion storage ring [44]. Having energies in the optical range (2–3 eV), these transitions may provide an alternative and very promising tool for studying the parity violation phenomena. Their theoretical analysis, however, requires a more systematic treatment of the electron-electron interaction effects. Based on the multiconfiguration Dirac-Fock approach and B-spline basis set method, investigations along this line are currently underway and will be reported elsewhere.

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