Parametrization of the angular correlation and degree of linear polarization in two-photon decays of hydrogenlike ions

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The spontaneous two-photon emission in hydrogenlike ions is investigated within the framework of second-order perturbation theory and Dirac's equation. Special attention is paid to the angular correlation of the emitted photons as well as to the degree of linear polarization of one of the two photons, if the second is just observed under arbitrary angles. Expressions for the angular correlation and the degree of linear polarization are expanded in powers of cosine functions of the two-photon opening angle, whose coefficients depend on the atomic number and the energy sharing of the emitted photons. The effects of including higher (electric and magnetic) multipoles upon the emitted photon pairs beyond the electric-dipole approximation are also discussed. Calculations of the coefficients are performed for the transitions $2s_{1/2} \to 1s_{1/2}$, $3d_{3/2} \to 1s_{1/2}$, and $3d_{5/2} \to 1s_{1/2}$, along the entire hydrogen isoelectronic sequence ($1 \leqslant Z \leqslant 100$).

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I. INTRODUCTION

Studies on the two-photon decay of atoms and ions have a long tradition, which can be traced back to the seminal works of Göppert-Mayer and of Breit and Teller [1,2]. These studies made the first prediction of nonlinear (second-order) processes in atomic physics and were originally motivated by astrophysics [3,4]. More recently, various theoretical investigations on such processes have aimed to test the standard model by measuring parity nonconservation (PNC) effects in both hydrogen- [5] and heliumlike ions [6–8]. Furthermore, fundamental effects of quantum theory have also been tested in experiments [9,10] and theoretically investigated [11], using two-photon decay channels. Recent technical advances in polarization and position-sensitive detectors have opened up the possibility of investigating angular and polarization properties of the radiation emitted in atomic decays [12–14]. For medium- and high-Z ions, in particular, measurements of two-photon angular and polarization properties are presently planned to be performed at GSI in Darmstadt in forthcoming years. To support these measurements and to extend previous investigations [15,16], we here present a relativistic study on the *angle correlation* and *degree of linear polarization* of the radiation emitted in (spontaneous) two-photon decays of hydrogenlike ions. We investigate both the angular dependence of the differential decay rate (referred to as angular correlation) as well as the degree of linear polarization of the "first" photon, if the "second" photon is observed under certain angles and if its polarization properties remain unobserved. Moreover, by using the symmetry properties of the second-order transition amplitude, the angular correlation and the degree of linear polarization of the emitted photons are written as an expansion in powers of $\cos\theta$, where θ is the (opening) angle between the direction of the two photons. Such a (relativistic) *parametrization* can be readily compared with other theoretical calculations and provides a theoretically well-justified fit model for future experiments that involve two-photon transitions. Up to now, there is no *fully* relativistic parametrization of such quantities for hydrogenlike ions available. A previous parametrization of the angular correlation performed by Au [17] was restricted to the $2s_{1/2} \to 1s_{1/2}$ transition as well as to a nonrelativistic framework. This paper is organized as follows: in Sec. II A we give a brief overview of the background theory involved in two-photon emission and introduce the (second-order) reduced amplitudes, which represent the building blocks of the theoretical calculation. Next, we define the angle correlation and degree of linear polarization functions together with its parametrization procedure in Secs. II B and II C. The numerical evaluation of the reduced amplitudes is described in Sec. II D. Section III contains the coefficients of the expansions for the $2s_{1/2} \to 1s_{1/2}$, $3d_{3/2} \to 1s_{1/2}$, and $3d_{5/2} \to 1s_{1/2}$ transitions. In order to describe more easily the relativistic behavior of these coefficients, we express them in terms of reduced two-photon matrix elements. In addition, the effect of higher-order multipoles other than the dominant electric dipole in these evaluations is also discussed. A brief summary is given in Sec. IV.

II. THEORY

A. Two-photon transition amplitude

The standard theoretical formalism for the description of two-photon decays in atoms or ions is based on the relativistic second-order perturbation theory and has recently been applied in Refs. [15,18,19]. In the present work, we therefore restrict

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ourselves to a compilation of those equations that are needed for the present analysis. The second-order transition amplitude for the emission of two photons with wave vectors $\boldsymbol{k}_\gamma$ ($\gamma = 1, 2$) and polarization vectors $\boldsymbol{u}_{\lambda_\gamma}$ ($\lambda_\gamma = \pm 1$), from an initial atomic state $|i\rangle = |n_i j_i m_i\rangle$ to a final atomic state $|f\rangle = |n_f j_f m_f\rangle$, with well-defined principal quantum number $n_{i,f}$, total angular momenta $j_{i,f}$, and their projections $m_{i,f}$, is given by [18]

$$\mathcal{M}_{fi}^{\boldsymbol{k}_1\boldsymbol{k}_2}(m_i, m_f, \lambda_1, \lambda_2) = \sum\!\!\!\!\!\!\int_{\nu} \left[\frac{\langle f|\,\hat{\mathcal{R}}^\dagger(\boldsymbol{k}_1,\lambda_1)\,|\nu\rangle\,\langle\nu|\,\hat{\mathcal{R}}^\dagger(\boldsymbol{k}_2,\lambda_2)\,|i\rangle}{E_\nu - E_i + \omega_2} + (1 \longleftrightarrow 2) \right]. \tag{1}$$

Here $\omega_{1,2}$ are the energies of the emitted photons and the transition operator $\hat{\mathcal{R}}(\boldsymbol{k}_\gamma, \lambda_\gamma)$ in Eq. (1) denotes the interaction between the electron and the electromagnetic radiation. Note that the index γ stands to the first ($\gamma = 1$) or second ($\gamma = 2$) photon. Moreover, the intermediate states $|\nu\rangle = |n_\nu j_\nu m_\nu\rangle$ in Eq. (1) form a complete set of states, including both discrete and positive or negative energy eigenvalues of the Dirac spectrum. The symbol $\sum\!\!\!\!\!\!\int_\nu$ stands for a summation over the discrete as well as an integration over the continuum part of this intermediate states.

In the so-called Coulomb gauge, which corresponds to the velocity form of the electron-photon interaction operator in the nonrelativistic limit, the transition operator reads

$$\hat{\mathcal{R}}(\boldsymbol{k}_\gamma, \lambda_\gamma) = \boldsymbol{\alpha} \cdot \boldsymbol{u}_{\lambda_\gamma} e^{i\boldsymbol{k}_\gamma \cdot \boldsymbol{r}}, \tag{2}$$

where $\boldsymbol{\alpha}$ is the usual vector of Dirac matrices.

Owing to the conservation of energy, the initial and final ionic energies E_i and E_f are related to the energies of the emitted photons $\omega_{1,2}$ by

$$\omega_t = E_i - E_f = \omega_1 + \omega_2, \tag{3}$$

where ω_t is the total energy of the transition. Instead of working with photon energies, it is therefore more convenient to describe the decay using the energy sharing parameter $y = \omega_1/\omega_t$.

The evaluation of the angular and polarization properties of the emitted radiation requires a spherical tensor decomposition of the photon fields contained in Eq. (1) into their electric and magnetic multipole components [20]. This decomposition reads

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

where $[L_\gamma] = 2L_\gamma + 1$, $k = |\boldsymbol{k}|$, and $D^{L_\gamma}_{M_\gamma\lambda}$ are the Wigner rotation matrices of rank L_γ and $\hat{a}^{p=0,1}_{L_\gamma M_\gamma}(k, \boldsymbol{r})$ refer to the magnetic ($p = 0$) and the electric ($p = 1$) multipole components, respectively. Thus, for example, the multipole component with $L_\gamma = 1$ and $p = 1$ is referred to as an electric-dipole ($E1$), while others are designated higher (magnetic and electric) multipoles.

Inserting Eqs. (2)–(4) into Eq. (1) and using the Wigner-Eckart theorem, we get the general expression for the second-order transition amplitude,

$$\begin{aligned} \mathcal{M}_{fi}^{\boldsymbol{k}_1\boldsymbol{k}_2}(m_i, m_f, \lambda_1, \lambda_2) = 2\pi \sum_{L_1 M_1 p_1} \sum_{L_2 M_2 p_2} (-i)^{L_1+L_2} [L_1, L_2]^{1/2} (-i\lambda_1)^{p_1} (-i\lambda_2)^{p_2} D^{L_1 *}_{M_1\lambda_1}(\hat{\boldsymbol{k}}_1) D^{L_2 *}_{M_2\lambda_2}(\hat{\boldsymbol{k}}_2) \\ \times \sum_{j_\nu m_\nu} \frac{1}{[j_i, j_\nu]^{1/2}} \Big[\langle j_f m_f L_1 M_1 | j_\nu m_\nu \rangle \langle j_\nu m_\nu L_2 M_2 | j_i m_i \rangle S^{j_\nu}_{L_1 p_1, L_2 p_2}(\omega_2) \\ + \langle j_f m_f L_2 M_2 | j_\nu m_\nu \rangle \langle j_\nu m_\nu L_1 M_1 | j_i m_i \rangle S^{j_\nu}_{L_2 p_2, L_1 p_1}(\omega_1) \Big], \end{aligned} \tag{5}$$

where the reduced amplitudes $S^{j_\nu}_{L_1 p_1, L_2 p_2}$ are defined as

$$S^{j_\nu}_{L_1 p_1, L_2 p_2}(\omega_2) = \sum\!\!\!\!\!\!\int_{n_\nu} \frac{\langle n_f j_f || \boldsymbol{\alpha} \hat{a}^{p_1\dagger}_{L_1}(k_1) || n_\nu j_\nu \rangle \langle n_\nu j_\nu || \boldsymbol{\alpha} \hat{a}^{p_2\dagger}_{L_2}(k_2) || n_i j_i \rangle}{E_\nu - E_i + \omega_2}. \tag{6}$$

The evaluation of the reduced matrix elements $\langle || \boldsymbol{\alpha} \hat{a}^{p\dagger}_L(k) || \rangle$ is discussed in Ref. [21].

The numerical evaluation of expression (6) is rather difficult and has been accomplished using different approaches, namely the Coulomb-Greens's function (both nonrelativistic [17,22,23] and relativistic cases [15,24,25]), and the finite basis set method [18,19]. The main difficulty of this evaluation concerns the summation over the infinite intermediate Dirac states $|n_\nu, j_\nu\rangle$, which includes both the discrete as well as the positive and negative continuum part of the spectrum. In this work, Eq. (6) is evaluated by exploiting both the relativistic Coulomb-Green's function and the finite basis set approaches. We outline the theory of these two approaches in Sec. II D. In the following sections, we define the angular correlation function and the degree of linear polarization of the radiation emitted in two-photon decays of hydrogenlike ions, which are the two observables of interest in the present work.

B. Angular correlation function

If we assume that the excited ions are initially unpolarized and that the spin states of the emitted photons remain unobserved during the measurement, the differential decay

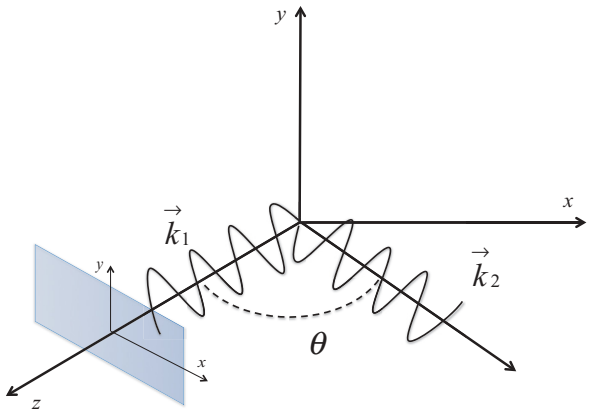

FIG. 1. (Color online) Geometry of the two-photon decay. The z axis is collinear with the first photon momentum vector $\boldsymbol{k_1}$. The plane zx contains the second photon momentum vector $\boldsymbol{k_2}$ The angle θ is the opening angle between the direction of the two photons.

rate can be written in atomic units as [18]

$$\frac{dW}{dy\,d\Omega_1\,d\Omega_2} = \omega_i^3 \frac{y(1-y)}{(2\pi)^3 c^2} \frac{1}{2j_i+1} \times \sum_{m_i,m_f\lambda_1,\lambda_2} \left|\mathcal{M}_{fi}^{\boldsymbol{k}_1\boldsymbol{k}_2}(m_f,m_i,\lambda_1,\lambda_2)\right|^2. \quad (7)$$

Here $\mathcal{M}_{fi}$ is given by Eq. (1). We define the angular correlation function $W^y(\theta)$ as the differential decay rate for a given opening angle θ between the two photons (see Fig. 1) and energy fraction y, irrespective of their polarizations.

Since there is no preferred direction for the decay of an unpolarized ion, it is generally more convenient to adopt the quantization (z) axis along the momentum of the first photon: $\boldsymbol{k}_1||z$ (see Fig. 1). This choice of quantization axis enables us to evaluate the angular correlation function by integrating Eq. (7) over the solid angle of the first photon ($d\Omega_1$) as well as over the azimuthal angle of the second photon ($d\phi_2$),

$$W^y(\theta) = 8\pi^2 \frac{dW}{dy\,d\Omega_1\,d\Omega_2}(\theta_1=0,\phi_1=0,\phi_2=0) = \frac{dW}{dy\,d\cos\theta}. \quad (8)$$

Thus the opening angle θ is the polar angle of the second photon θ_2. The factor $8\pi^2$ in Eq. (8) hereby arises from the integration over $d\Omega_1$ and $d\phi_2$. Due to the photon-photon permutation symmetry that characterizes the amplitude in Eq. (1) and corresponds to the exchange of the emitted photons, the analytic expression for $W^y(\theta)$ can only contain terms that are θ-even under the algebraic replacement of $\theta \to -\theta$. We can thus expand $W^y(\theta)$ in powers of $\cos\theta$,

$$W^y(\theta) = a_0\left(1+\sum_{i=1}^{\infty} a_i \cos^i\theta\right), \quad (9)$$

where the parameters a_i depend on the atomic number of the ion, the energy sharing parameter y, and the states involved in the decay. To make the procedure for obtaining the parameters a_n less cumbersome, the angular correlation was first expanded in orthogonal polynomials of $\cos(\theta)$. In the present case, we considered the Legendre polynomials [26]. Each coefficient l_m of this initial expansion was obtained by performing the scalar product [26] of the respective Legendre polynomial P_m with $W^y(\theta)$. This procedure simplifies the algebraic conversion of the various combinations of $\sin^i(\theta)\cos^j(\theta)$ (i and j being arbitrary integers) in Eq. (8) into powers of $\cos(\theta)$. Finally, each parameter a_n is given as a combination of the coefficients l_m. Note that only the parameter a_0 has units, which in S.I. are s^{-1}.

By integrating the parametrization (9) in dy and $d\cos\theta$, only the even parameters contribute to the total decay rate.

C. Degree of linear polarization

We used the spin-polarization density matrix to analyze the polarization properties of the emitted radiation, which has been applied several times to the two-photon decay rate [15,27]. The first Stokes parameter is given by [27]

$$P_1(\theta) = \frac{2}{\mathcal{N}}\mathrm{Re}\Big[\sum_{m_i,m_f,\lambda_2} \mathcal{M}_{fi}^{\boldsymbol{k}_1\boldsymbol{k}_2}(m_f,m_i,\lambda_1=1,\lambda_2) \times \mathcal{M}_{fi}^{\boldsymbol{k}_1\boldsymbol{k}_2*}(m_f,m_i,\lambda_1=-1,\lambda_2)\Big], \quad (10)$$

where $\mathcal{N}$ is a normalization coefficient given by

$$\mathcal{N} = \sum_{m_i,m_f\lambda_1,\lambda_2} \left|\mathcal{M}_{fi}^{\boldsymbol{k}_1\boldsymbol{k}_2}(m_f,m_i,\lambda_1,\lambda_2)\right|^2, \quad (11)$$

which assures that the trace of the spin-polarization density matrix is unitary. The derivation of Eq. (10) follows from the assumption that the polarization of only one (first) of the photons is observed. The normalization constant $\mathcal{N}$ is proportional to $W^y(\theta)$ defined in the previous section. Therefore, by employing Eq. (9) into Eq. (10) and using the symmetry properties of $\mathcal{M}_{fi}$, we can parametrize P_1 as

$$P_1(\theta) = \frac{b_0\left(1+\sum_{k=1}^{\infty} b_k\cos^k\theta\right)}{a_0\left(1+\sum_{i=1}^{\infty} a_i\cos^i\theta\right)}. \quad (12)$$

Like the case of a_i, parameters b_i depend on the ion atomic number and energy sharing. It should be noted that within the current geometry and since we assume that the atom or ion is unpolarized, the second Stokes parameter vanishes. Therefore, the degree of linear polarization, which we define as $P_L(\theta)$, is equal to the absolute value of the first Stokes parameter, i.e., $P_L = \sqrt{P_1^2+P_2^2} = |P_1|$ [27]. The procedure for obtaining the parameters a_n was also applied to the parameters b_k.

We find by analytical evaluation that the relations $b_1 = -b_3$ and $b_2 = -(1+b_4)$ are valid for the entire isoelectronic sequence, energy shares, or type of transition. By considering these relations, the degree of polarization $P_L(\theta)$ takes the form

$$P_L(\theta) = \left|\frac{b_0[\sin^2\theta(1+b_1\cos\theta - b_4\cos^2\theta)+\cdots]}{W^y(\theta)}\right|. \quad (13)$$

It can observed that by neglecting parameters b_i for $i>4$, the degree of polarization is zero for values of θ equal to 0° or 180°. This is expected since for these angles there is no unique plane of reaction defined by the direction of the photons, neither a unique coordinated system that defines the first photon polarization. Further relations between b_i with $i>4$ can thus be expected in order to have $P_L=0$ for these geometrical settings. Obtaining such relations goes beyond

the scope of this work since, as discussed in Sec. III, the contribution of these (higher) parameters to the degree of polarization can be neglected.

D. Computation of the *S* functions

The reduced amplitudes (6) were evaluated by making use of both the relativistic *Coulomb-Green's* function [15,24] and the *finite basis set* [19] approaches. The first approach is based on the formal solution of the Green's equation of the atomic Hamiltonian. This function contains the complete summation (integration) over the Dirac's spectrum that can therefore be used to replace the summation in Eq. (6). We note that the Green's function is known analytically both for the nonrelativistic Schrödinger-Coulomb as well as the relativistic Dirac-Coulomb Hamiltonian and it can be represented in terms of Laguerre polynomials [25]. The second approach (finite basis set) has been recently applied to two-photon decay [28] and two-photon absorption [8] in hydrogenlike ions as well as to Rayleigh scattering [29,30] in hydrogen. It is based on enforcing boundary conditions leading to a "discretization of the continuum," which enables one to replace the infinite sum over the bound states as well as the integration over the continuum by just a "finite summation" over a basis set [31].

In this work we use the *B-splines* basis set. Another basis set, *B-polynomials* [32], was used to check the results of the finite basis set method. By carrying out the calculations, we verified that both approaches yielded identical results for the values of the coefficients in Eqs. (9) and (12). Finally, to further assess the accuracy of the reduced matrix elements $S^{j_v}_{L_1 p_1, L_2 p_2}$, these elements were evaluated in both Coulomb and Lorentz gauges and tested in a total decay rate expression as performed in Ref. [19].

III. RESULTS AND DISCUSSION

Figure 2 displays the values of the coefficients a_i in the expansion (9) of the two-photon correlation function $W^y(\theta)$ for the two-photon $2s_{1/2} \rightarrow 1s_{1/2}$ transition as function of the nuclear charge Z. We considered only multipoles ($L < 4$) that give a visible contribution to the figure. Several different energy shares ($y = 0.1, 0.25, 0.5$) are displayed. We notice that the angular correlation function is well described, in the low-Z regime, by $W^y(\theta) \sim a_0(1 + \cos^2\theta)$, which corresponds to the dipole approximation [23]. This is in agreement also with the more general result of Yang [33], who predicts an angular correlation of the form $[1 + \beta \cos^2(\theta)]$ for dipole-dipole transitions. A deviation to this expression arises from a_1 and a_3, which leads to an asymmetry (with respect to $\theta = 90^\circ$) and to a "tilt" of the angular correlation function, i.e., a slight preference of a back-to-back emission of the two photons ($\theta = 180^\circ$), when compared with an emission into the same

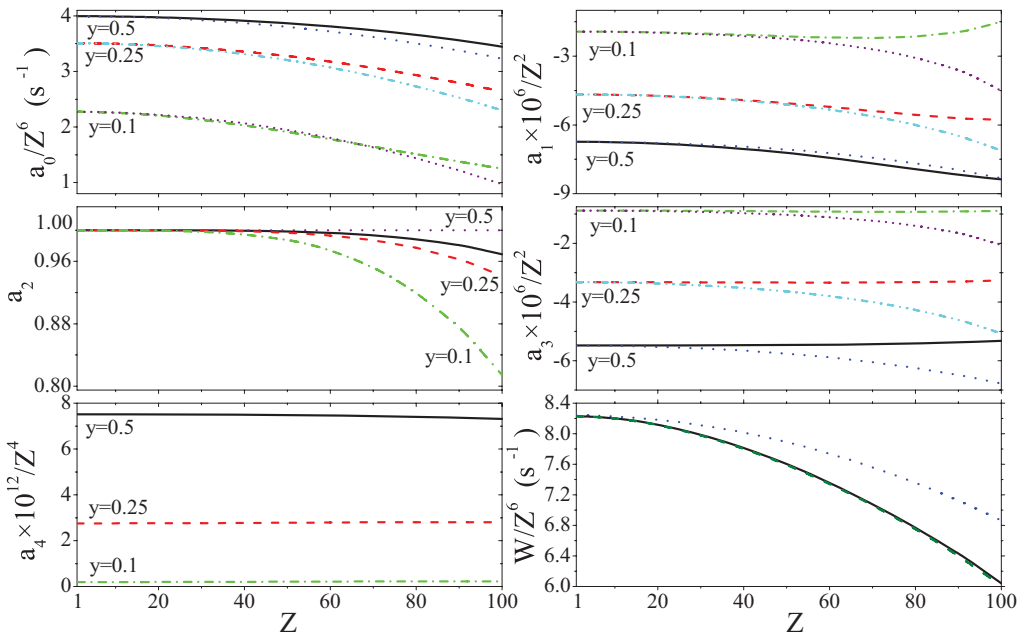

FIG. 2. (Color online) Values of the parameters a_i defined in Eq. (9), for the $2s_{1/2} \rightarrow 1s_{1/2}$ transition as a function of Z. The values obtained in this work, which correspond to different energy shares $y = 0.1$, 0.25, and 0.5, are represented by a black-solid, red-dashed, and green-dot-dashed curve, respectively. With exception of a_0 (given in s^{-1}), all parameters are dimensionless. The same energy shares values given by Au [17] are represented by a blue dotted line ($y = 0.5$), a cyan dot-dot-dashed line ($y = 0.25$), and by the purple short dashed line ($y = 0.1$). The parameters a_2 of Ref. [17] are always equal to one, as represented by the purple dotted line. On the bottom right plot, the blue dotted line refers to values of the total decay rate obtained from the parametrization of Ref. [17]. The black line represents the total decay values obtained from our current parametrization. The dark green dashed line contains the values provided by Refs. [18,19]. The level of precision of the plot does not allow one to distinguish differences between these references. Values of parameters b_i can be obtained from this figure using the relations $b_0 \approx -a_0$, $b_1 \approx a_3$, and $b_4 \approx -a_4$.

direction ($\theta = 0^\circ$). In order to quantify this deviation, we write the coefficients a_i directly in terms of the reduced amplitudes $S^{j_\nu}_{L_1 p_1, L_2 p_2}(\omega_2)$, defined in Eq. (6). By restricting ourselves to contributions of order $(\alpha Z)^2$ or lower, the angular correlation is given approximately by

$$W^y(\theta) \propto |\mathcal{S}_{E1}|^2 \left[\left(1 - 4 \left| \frac{\mathcal{D}_{E1}}{\mathcal{S}_{E1}} \right| \right) (1 + \cos^2\theta) - 4 \left| \frac{\mathcal{S}_{M1}}{\mathcal{S}_{E1}} \right| \cos\theta - 4 \left| \frac{\mathcal{S}_{E2}}{\mathcal{S}_{E1}} \right| \cos^3\theta \right], \quad (14)$$

where, for the sake of brevity, we introduced the notation

$$\mathcal{S}_{Lp} = S^{j_\nu}_{Lp,Lp}(\omega_2) + S^{j_\nu}_{Lp,Lp}(\omega_1) \quad (15)$$

and

$$\mathcal{D}_{Lp} = 2S^{j_\nu}_{Lp,Lp}(\omega_2) + 2S^{j_\nu}_{Lp,Lp}(\omega_1) + S^{j_\nu+1}_{Lp,Lp}(\omega_2) + S^{j_\nu+1}_{Lp,Lp}(\omega_1), \quad (16)$$

with $j_\nu = 1/2$ for the multipoles $2E1$ and $2M1$ and $j_\nu = 3/2$ for the $2E2$ case.

By expanding the wave functions and energies inside the terms $S^{1/2}_{2E1}(\omega)$ and $S^{3/2}_{2E1}(\omega)$ in powers of αZ (α being the fine structure constant), we find that the term $\mathcal{D}_{E1}$ would be equal to zero, if nonrelativistic wave functions and energies were employed. Furthermore, this term scales with the atomic number as $(\alpha Z)^2$. As can be observed from Eq. (14), the deviations of the angular correlation from the low-Z regime arise from both interference between the leading multipole $2E1$ and the next higher multipoles $2M1$ and $2E2$ as well as from relativistic effects.

The term $\mathcal{S}_{E1}$ is five orders of magnitude higher than the terms $\mathcal{S}_{M1}$ and $\mathcal{S}_{E2}$ for $Z = 1$. While the term $\mathcal{S}_{E1}$ scales as $(\alpha Z)^0$, the terms $\mathcal{S}_{M1}$ and $\mathcal{S}_{E2}$ scale as $(\alpha Z)^2$, which gives rise to the asymmetry of the angular correlation for higher-Z ions.

The scaling laws for the reduced amplitudes (6) were obtained from the expressions of the reduced matrix elements [21].

The expression (14) evaluated for the case of two emitted collinear photons ($\theta = 0$) has the same proportionality to the multipoles as a similar expression (18) obtained for the case of the absorption of two collinear photons [8].

Parameters a_2 and a_4 deviate from one and zero, respectively, for high-Z ions due to terms $S^{j}_{2M1}(\omega) \times S^{j'}_{2M1}(\omega')$, $S^{j}_{2E2}(\omega) \times S^{j'}_{2E2}(\omega')$, or cross multipoles not listed in Eq. (14), which scale as $(\alpha Z)^4$. These terms, along with similar ones in a_0, give the contribution of the multipoles $M1M1$, $E2E2$, $E1M2$, and $M2E1$ to the total decay rate. Parameters a_i with $i > 4$ depend on even higher multipoles and can be neglected even for $Z = 100$. The parameter a_0 and W shown in Fig. 2 scales as $(\alpha Z)^6$ due to the energy transition w_t in Eq. (7), which scales as $(\alpha Z)^2$.

The asymmetry of the angular correlation function was theoretically first studied by Au [17] using a nonrelativistic approach with the inclusion of higher (nondipole) order multipoles. We show in Fig. 2, for comparison purposes, the parameters obtained by Au for the $2s_{1/2} \rightarrow 1s_{1/2}$ transition and $y = 0.1, 0.25, 0.5$. For low-Z ions, we find good agreement between our results and Au's results in all parameters [34]. On the other hand, the agreement between the two data sets becomes worse for increasing values of Z. As can be observed from Eq. (14), the first correction to the term a_0 comes from relativistic effects, i.e., from the term $\mathcal{D}_{E1}$. In our work the full extent of these effects was taken into account using the Dirac theory, while in Ref. [17] nonrelativistic wave functions were used with a relativistic correction to the $2E1$ multipole.

To further assess our parameters a_0 and a_2, we evaluated the total decay rate by integrating our angular correlation function in dy and $d\cos\theta$ and compared it with Refs. [18,19]. The bottom right panel of Fig. 2 shows a plot of the total decay rate for several values of Z and values from other references. The difference between the values of Ref. [18] and Ref. [19] is so small that both references can be represented by a single line. The plot in Fig. 2 highlights that our results are in good agreement with the above two references. However, this is not the case for the Au values of the decay rate, which have a maximum discrepancy of 12% for $Z = 100$.

We observed good agreement between our results of the parameters a_1 and the respective results of Ref. [17] for $y = 0.5$, and this would seem to indicate a good agreement of the $2M1$ multipole contribution [see Eq. (14)]. With decreasing energy sharing values the agreement degrades, the maximum difference being at $y = 0.1$ and $Z = 100$ of 60%. On the other hand, for the parameters a_3, there is some disagreement in the $2E2$ multipole with high-Z ions at all energy shares, the maximum difference being of 35% for $y = 0.25$ and $Z = 100$. There were similar discrepancy values for the a_2 parameter and the maximum difference was 20% for $Z = 100$ and $y = 0.1$. In the parametrization provided by Ref. [17], the a_2 is equal to one for all energy shares and atomic numbers.

The asymmetry of the correlation function due to the relativistic and nondipole contribution was also investigated by Surzhykov *et al.* [15], who carried out a relativistic evaluation of all relevant multipoles. Table I shows values for the intensity ratio between the angular correlation under 0° and 180° obtained in the present work, as well as by Au [17] and Surzhykov *et al.* [15]. As can be observed, good agreement is found for the values presented by Surzhykov *et al.* [15].

Overall, we noticed that deviations from the formula $W^y(\theta) = a_0(1 + \cos^2\theta)$ start playing a role, reaching a size of some percent from hydrogenlike Tin ion onwards ($Z \gtrsim 50$).

As with the angular correlation function, the nondipole and relativistic effects on the degree of linear polarization function (13) can also be estimated by restricting its evaluation to contributions of order $(\alpha Z)^2$ or lower. The result of this

TABLE I. Intensity ratio between the angular correlation between 0° and 180° for several values of the atomic number and equal energy sharing ($y = 0.5$). Comparison between the values obtained in this work and other theoretical values.

	$W^{0.5}(180^\circ)/W^{0.5}(0^\circ)$		
	$Z = 1$	$Z = 54$	$Z = 92$
Surzhykov *et al.* [15]	1.000	1.038	1.124
Au [17]	1.000	1.038	1.131
This work	1.000	1.038	1.123

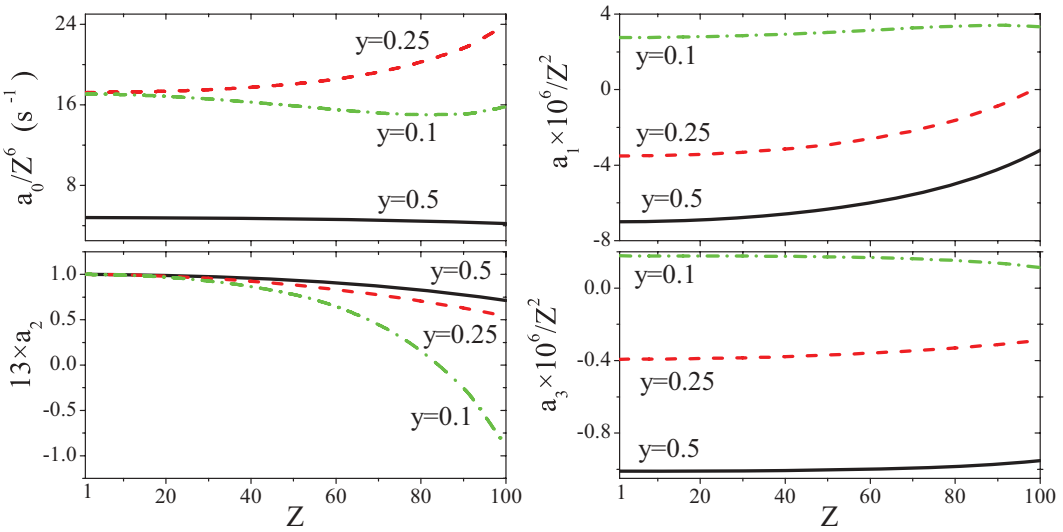

FIG. 3. (Color online) Values of the parameters a_i defined in Eq. (9), for the $2d_{3/2} \rightarrow 1s_{1/2}$ transition as function of Z. The different energy shares $y = 0.1, 0.25, 0.5$ are represented by a black-solid, red-dashed, and green-dot-dashed curve, respectively. With exception of a_0 (given in s^{-1}), all parameters are dimensionless. Values for parameters b_i can be obtained from this figure (within its resolution) using the relations $b_0 \approx -a_0/13$ and $b_1 \approx 13a_3$.

procedure is given by

$$P_L(\theta) \approx \left| \frac{-\left(1 - 4\left|\frac{\mathcal{D}_{E1}}{\mathcal{S}_{E1}}\right|\right) \sin^2\theta \left(1 - 4\left|\frac{\mathcal{S}_{E2}}{\mathcal{S}_{E1}}\right| \cos\theta\right)}{\left(1 - 4\left|\frac{\mathcal{D}_{E1}}{\mathcal{S}_{E1}}\right|\right)\left(1 + \cos^2\theta\right) - 4\left|\frac{\mathcal{S}_{M1}}{\mathcal{S}_{E1}}\right| \cos\theta - 4\left|\frac{\mathcal{S}_{E2}}{\mathcal{S}_{E1}}\right| \cos^3\theta} \right| . \tag{17}$$

The numerator of the degree of linear polarization (12) in $2s_{1/2} \rightarrow 1s_{1/2}$ transitions can be well described by $a_0 \sin^2\theta$, so that $P_L(\theta) = \sin^2\theta/(1 + \cos^2\theta)$ [35], as long as the atomic number is relatively small.

Equation (17) highlights that $a_0 = -b_0$ and $a_3 = b_1$ if multipoles lower than three were employed and also in this case b_0 and b_1 could be obtained from Fig. 2. The maximum degree of polarization corresponds to $\theta = 90^\circ$ [$P_L(90^\circ) = |b_0/a_0|$]. This maximum is not much affected by nondipole and relativistic effects with explicit values of 1 for $y = 0.5$ and 0.9 for $y = 0.1$. As in the case of a_4, parameters b_4 depend on multipole contributions of order $(\alpha Z)^4$, which are not shown

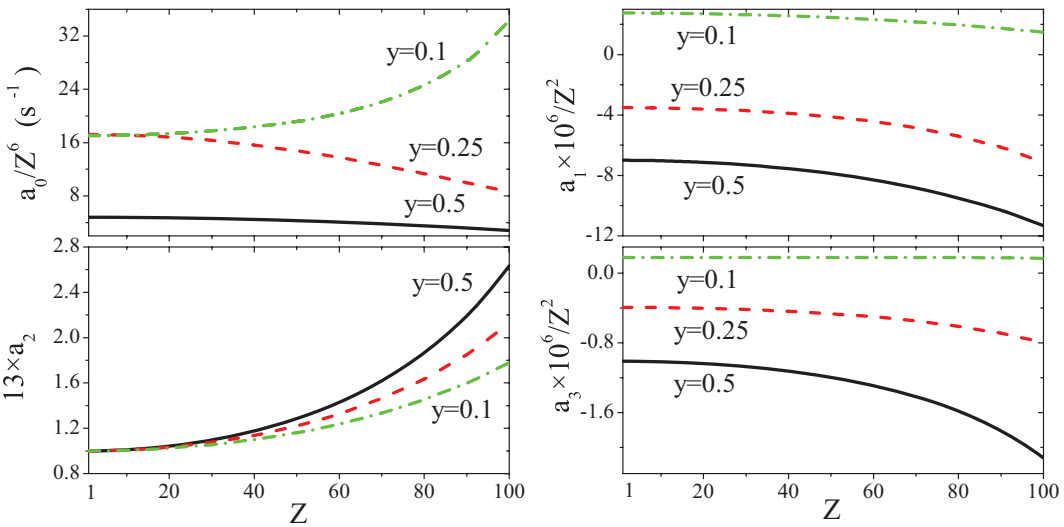

FIG. 4. (Color online) Values of the parameters a_i defined in Eq. (9), for the $2d_{5/2} \rightarrow 1s_{1/2}$ transition as a function of Z. The different energy shares $y = 0.1, 0.25, 0.5$ are represented by a black-solid, red-dashed, and green-dot-dashed curve, respectively. With exception of a_0 (given in s^{-1}), all parameters are dimensionless.

TABLE II. Values of the parameters a_i and b_i defined in Eqs. (9) and (12), for the transition $2s_{1/2} \rightarrow 1s_{1/2}$. The energy sharing is fixed at $y = 0.5$. All parameters are dimensionless, with the exception of a_0 and b_0, which are given in s^{-1}.

Z	a_0/Z^6	$a_1 \times 10^6/Z^2$	a_2	$a_3 \times 10^6/Z^2$	$a_4 \times 10^{12}/Z^4$	b_0/Z^6	$b_1 \times 10^6/Z^2$	$b_4 \times 10^{12}/Z^4$
1	3.9942	−6.7348	1.0000	−5.4827	7.5149	−3.9942	−5.4827	−7.5149
4	3.9935	−6.7378	1.0000	−5.4826	7.5147	−3.9935	−5.4826	−7.5147
8	3.9911	−6.7472	1.0000	−5.4824	7.5142	−3.9911	−5.4824	−7.5142
12	3.9871	−6.7630	1.0000	−5.4820	7.5132	−3.9871	−5.4821	−7.5132
16	3.9816	−6.7851	1.0000	−5.4815	7.5119	−3.9816	−5.4816	−7.5119
20	3.9744	−6.8135	1.0000	−5.4808	7.5101	−3.9744	−5.4809	−7.5102
30	3.9494	−6.9118	0.9998	−5.4781	7.5037	−3.9491	−5.4788	−7.5040
40	3.9142	−7.0488	0.9993	−5.4736	7.4941	−3.9132	−5.4757	−7.4951
50	3.8683	−7.2232	0.9984	−5.4664	7.4807	−3.8658	−5.4716	−7.4830
54	3.8468	−7.3029	0.9978	−5.4625	7.4740	−3.8434	−5.4697	−7.4771
60	3.8111	−7.4323	0.9966	−5.4553	7.4625	−3.8058	−5.4665	−7.4672
70	3.7419	−7.6704	0.9935	−5.4385	7.4381	−3.7319	−5.4602	−7.4466
80	3.6591	−7.9268	0.9885	−5.4136	7.4063	−3.6418	−5.4528	−7.4202
90	3.5614	−8.1788	0.9808	−5.3766	7.3640	−3.5332	−5.4436	−7.3849
92	3.5402	−8.2248	0.9788	−5.3671	7.3535	−3.5093	−5.4414	−7.3759
100	3.4474	−8.3798	0.9692	−5.3215	7.3078	−3.4036	−5.4321	−7.3363

in Eq. (17). We found that $a_4 \approx -b_4$ for all the isoelectronic sequence. Moreover, like in the case of a_i, parameters b_i with $i > 4$ depend on higher multipoles and therefore can be neglected.

As shown in Fig. 2, the deviations from both the angular correlation and the degree of linear polarization present a nontrivial dependence on the energy sharing that characterizes the decay: $a_{i=0,2,4}$ are greater in magnitude for higher energy shares, while $a_{i=1,3}$ and $b_{i=0,1,4}$ are smaller with increasing values of energy shares.

Other transitions in which a two-photon emission is observed are the $3d_{3/2} \rightarrow 1s_{1/2}$ and $3d_{5/2} \rightarrow 1s_{1/2}$ [36]. Unlike the $2s_{1/2} \rightarrow 1s_{1/2}$ transition that we studied above, these transitions are resonant and exhibit peaks for certain values of y corresponding to the $3d_{3/2,5/2} \rightarrow 2p_{3/2,1/2} \rightarrow 1s_{1/2}$ $2E1$ decay channels [28]. In the nonrelativistic limit, these resonances have a single value of $y_r = 5/32 \approx 0.156$. Figures 3 and 4 report the coefficients a_i and b_i as obtained for the two-photon transitions $3d_{3/2} \rightarrow 1s_{1/2}$ and $3d_{5/2} \rightarrow 1s_{1/2}$, respectively. The values of $y = 0.1$ and 0.25 were chosen to be outside the region of the resonant peak, thus avoiding its influence on the angular correlation function over all values of Z (see Fig. 5 of Ref. [28]).

Similar to what is observed on Fig. 1, deviations from the nonrelativistic formula $W^y(\theta) \approx a_0(1 + \cos^2\theta/13)$ [22] are of the order of some percent from $Z \approx 50$ onwards. Also in the lower Z regime the degree of linear polarization is given by $P_L(\theta) = \sin^2\theta/(13 + \cos^2\theta)$. As for the a_i case, parameters b_i of the $3d_{3/2} \rightarrow 1s_{1/2}$ transition can be obtained from Fig. 3 using the following relations $b_0 \approx -a_0/13$ and $b_1 \approx 13a_3$. Like the case of the $2s_{1/2} \rightarrow 1s_{1/2}$ transition, relativistic and nondipole effects do not give a sizable contribution to the

TABLE III. Same as Table II but for $3d_{3/2} \rightarrow 1s_{1/2}$ transition.

Z	a_0/Z^6	$a_1 \times 10^6/Z^2$	$13 \times a_2$	$a_3 \times 10^6/Z^2$	$a_4 \times 10^{12}/Z^4$	$13 \times b_0/Z^6$	$b_1 \times 10^6/(13Z^2)$	$b_4 \times 10^{12}/(13Z^4)$
1	4.7852	−6.9947	1.0000	−1.0110	3.3220	−4.7852	−1.0110	−3.3220
4	4.7846	−6.9911	0.9996	−1.0110	3.3228	−4.7839	−1.0112	−3.3233
8	4.7824	−6.9795	0.9984	−1.0108	3.3254	−4.7797	−1.0118	−3.3273
12	4.7788	−6.9600	0.9963	−1.0106	3.3297	−4.7726	−1.0128	−3.3340
16	4.7738	−6.9326	0.9934	−1.0103	3.3358	−4.7628	−1.0143	−3.3435
20	4.7673	−6.8969	0.9897	−1.0099	3.3438	−4.7501	−1.0162	−3.3559
30	4.7443	−6.7704	0.9768	−1.0084	3.3721	−4.7058	−1.0228	−3.3997
40	4.7111	−6.5852	0.9587	−1.0062	3.4139	−4.6431	−1.0324	−3.4638
50	4.6666	−6.3323	0.9352	−1.0031	3.4713	−4.5612	−1.0454	−3.5513
54	4.6453	−6.2092	0.9243	−1.0016	3.4994	−4.5229	−1.0516	−3.5939
60	4.6090	−5.9978	0.9061	−0.9988	3.5479	−4.4590	−1.0623	−3.6671
70	4.5365	−5.5608	0.8708	−0.9928	3.6490	−4.3353	−1.0838	−3.8179
80	4.4453	−4.9896	0.8282	−0.9844	3.7829	−4.1876	−1.1113	−4.0148
90	4.3327	−4.2339	0.7762	−0.9721	3.9615	−4.0146	−1.1461	−4.2737
92	4.3080	−4.0543	0.7645	−0.9688	4.0037	−3.9775	−1.1541	−4.3343
100	4.1957	−3.2113	0.7113	−0.9530	4.2046	−3.8159	−1.1905	−4.6199

TABLE IV. Same as Table II but for $3d_{5/2} \rightarrow 1s_{1/2}$ transition.

Z	a_0/Z^6	$a_1 \times 10^6/Z^2$	$13 \times a_2$	$a_3 \times 10^6/Z^2$	$a_4 \times 10^{12}/Z^4$	$13 \times b_0/Z^6$	$b_1 \times 10^6/(13Z^2)$	$b_4 \times 10^{12}/(13Z^4)$
1	4.7851	−6.9953	1.0001	−1.0111	3.3221	−4.7852	−1.0110	−3.3219
4	4.7820	−7.0002	1.0016	−1.0121	3.3243	−4.7848	−1.0111	−3.3224
8	4.7722	−7.0158	1.0066	−1.0153	3.3317	−4.7832	−1.0113	−3.3240
12	4.7558	−7.0420	1.0149	−1.0207	3.3439	−4.7805	−1.0117	−3.3266
16	4.7329	−7.0788	1.0266	−1.0283	3.3612	−4.7766	−1.0123	−3.3305
20	4.7036	−7.1265	1.0418	−1.0381	3.3837	−4.7712	−1.0132	−3.3356
30	4.6018	−7.2946	1.0960	−1.0733	3.4639	−4.7503	−1.0165	−3.3553
40	4.4601	−7.5374	1.1756	−1.1251	3.5822	−4.7148	−1.0223	−3.3877
50	4.2790	−7.8629	1.2846	−1.1964	3.7458	−4.6583	−1.0317	−3.4382
54	4.1957	−8.0187	1.3378	−1.2313	3.8261	−4.6282	−1.0369	−3.4651
60	4.0593	−8.2827	1.4294	−1.2916	3.9653	−4.5729	−1.0465	−3.5145
70	3.8024	−8.8127	1.6188	−1.4175	4.2570	−4.4493	−1.0689	−3.6274
80	3.5091	−9.4751	1.8665	−1.5844	4.6467	−4.2759	−1.1022	−3.7938
90	3.1821	−10.2988	2.1932	−1.8090	5.1749	−4.0418	−1.1513	−4.0390
92	3.1134	−10.4854	2.2706	−1.8629	5.3018	−3.9874	−1.1635	−4.1003
100	2.8250	−11.3217	2.6331	−2.1191	5.9100	−3.7368	−1.2236	−4.4060

maximum degree of polarization [$P_L(90^\circ) = 1/13$] of the first photon emitted by the $3d_{3/2} \rightarrow 1s_{1/2}$ transition. However, this is not the case of the $3d_{5/2} \rightarrow 1s_{1/2}$ transition, where $P_L(90^\circ)$ increases by 20% from neutral hydrogen to H-like uranium for $y = 0.5$.

Tables II–IV in the Appendix provide the values of the parameters a_i and b_i with $i < 5$ for the transitions considered in this work for a few atomic numbers and for the case of equal energy sharing. The values of parameters not shown in the Figs. 3 and 4 can be found in these tables.

The angular correlation and the degree of linear polarization expressed as Eqs. (9) and (12), together with the parameters in Figs. 2–4, provide a theoretically well-justified fit model for any future experiments that involve two-photon polarization, for example, the parity nonconservation mixing coefficient measurement [7].

IV. SUMMARY

We analyzed the angular correlation and the degree of linear polarization for the radiation emitted in two-photon decay of hydrogenlike ions for the transitions $2s_{1/2} \rightarrow 1s_{1/2}$, $3d_{3/2} \rightarrow 1s_{1/2}$, and $3d_{5/2} \rightarrow 1s_{1/2}$. These two physical quantities were expanded in powers of $\cos\theta$ and the coefficients were plotted for the entire isoelectronic sequence ($1 \leqslant Z \leqslant 100$). By restricting ourselves to the first two multipoles of the photon field expansions, these coefficients were written in terms of the reduced amplitude of the process. Overall, we have shown that the coefficients that deviate from nonrelativistic formulas begin to be some percent from approximately hydrogenlike tin ion onwards ($Z \gtrsim 50$).

When available, a comparison with previous works was performed in order to verify these present results.

The parametrization of the angular correlation and the degree of linear polarization presented in this work could be exploited in future experiments aimed at measuring the angular and polarization properties of the radiation emitted in two-photon decay of atoms or ions.

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APPENDIX: TABLES OF THE PARAMETERS

The values for the parameters a_i and b_i are shown, for a few atomic numbers and equal energy sharing. b_2 and b_3 are not shown since the relations $b_2 = -(1 + b_4)$ and $b_3 = -b_1$ hold true.

The considered transitions are $2s_{1/2} \rightarrow 1s_{1/2}$, $3d_{3/2} \rightarrow 1s_{1/2}$, and $3d_{5/2} \rightarrow 1s_{1/2}$, which are displayed in Tables II, III, and IV, respectively.

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