Angular correlations in the two-photon decay of hydrogenlike ions: Relativistic Green's-function approach

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The angular correlations in the two-photon decay of hydrogenlike ions are studied within the framework of second-order perturbation theory, based on Dirac's equation. Particular attention has been paid to the effects which arise from the higher (nondipole) terms in the expansion of the electron-photon interaction. It is shown that the photon-photon angular correlation function, which is found symmetric with respect to the angle $\theta = 90°$ in the electric dipole approximation, becomes asymmetric because of the nondipole contributions, and that this effect is enhanced as the nuclear charge Z increases. Detailed computations on the photon-photon angular distribution have been carried out for the $2s_{1/2} \rightarrow 1s_{1/2}$ and $3d_{5/2} \rightarrow 1s_{1/2}$ transitions in neutral hydrogen (H) as well as for hydrogenlike xenon (Xe^{53+}) and uranium (U^{91+}) ions, and are compared with previous nonrelativistic results by Au [Phys. Rev. A **14**, 531 (1976)].

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I. INTRODUCTION

Since the early days of quantum mechanics, the two-photon transitions of (hydrogenlike) atoms has been the subject of studies both in theory [1–7] and experiment [8–10]. While, however, most investigations in the past dealt with the decay of neutral hydrogen and the low-Z ions, much of today's interest is focused also on the *high-Z* region in which the two-photon transitions serve as a sensitive probe of relativistic as well as quantum electrodynamical (QED) effects in strong electric fields. To explore and analyze these effects, a large number of theoretical case studies have been carried out in particular for the total decay rates and the photon energy distributions, based on Dirac's equation [5–7,11]. Less attention, in contrast, has been paid previously to the effects of relativity on the photon-photon *angular* correlation functions. Using a nonrelativistic approach, a first step towards such an angle-resolved analysis was performed by Au [4] almost 30 years ago, who calculated the angular correlations for the two-photon decay of the metastable $2s_{1/2}$ state in heavy hydrogenlike ions. Apart from the leading electric-dipole term, Au hereby incorporated also the higher multipoles in electron-photon interaction and included some semi-relativistic adjustments in his final results. For the $2s_{1/2} \rightarrow 1s_{1/2}$ two-photon decay especially, these nondipole effects were found to result in an asymmetrical shape of the photon angular distribution as function of the angle θ between the emitted photons, and when compared with the more familiar and symmetric (electric-dipole) form $1+\cos^2\theta$ [3,12]. A similar asymmetry in the photon-photon angular correlations were obtained later also in the *relativistic* computations by Mu and Crasemann [13] and Tong *et al.* [14], respectively, who studied the two-photon decay of a K-shell vacancy for molybdenum ($Z=42$) and silver ($Z=47$). But although the nondipole effects gave rise again in these case studies to a slight asymmetry in the angular correlation function, these effects are usually small for most neutral, medium-Z elements and suppressed, in addition, by the screening of the nuclear charge due to electron-electron interaction. Exploring the photon-photon angular correlations along the hydrogen isoelectronic sequence, in fact, provides a unique test bay for investigating the relativistic and nondipole effects in simple atomic systems.

In this contribution, second-order perturbation theory is applied to analyze the photon-photon angular correlation function for the two-photon decay of hydrogenlike ions. Starting from Dirac's equation, emphasis has been placed on the (relativistic) contraction of the wave functions as well as the influence of the higher multipoles in the expansion of the photon field. Most natural, these multipole effects beyond the electric-dipole approximation are studied by means of a (multipole) expansion of the two-photon transition amplitude and lead to a whole series of different decay amplitudes, shortly denoted by E1E1, E1M2, M1M1, E2M2, ..., respectively. Following the definition of the angle-differential emission rates in Sec. II A therefore we must first express the second-order amplitude in terms of the Coulomb-Green's-function in Sec. II B before the standard multipole expansion of the photon field can be applied (cf. Sec. II C). The complete expansion of both the Green's function and the photon field are utilized later in Sec. III to calculate the photon-photon correlation functions for the $2s_{1/2} \rightarrow 1s_{1/2}$ and $3d_{5/2} \rightarrow 1s_{1/2}$ decay for neutral hydrogen (H) as well as hydrogenlike xenon (Xe^{53+}) and uranium (U^{91+}) ions. From the comparison of our (relativistic) calculations with the nonrelativistic results by Au [4], we are able to extract the effects of relativity and of the higher multipoles in the expansion of the photon field as the nuclear charge Z is increased. Although both, the relativistic and nonrelativistic theory, predict a similar shift, at least *qualitatively*, for the photon-photon angular correlation function owing to the nondipole contributions, the nonrelativistic approximation by Au [4] overestimates the effects of the higher multipoles by more than 13% for the hydrogenlike uranium U^{91+}. Finally, a brief summary is given in Sec. IV.

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II. THEORY

A. Differential emission rate

Not much needs to be said here about the basic (second-order) formalism for studying the two-photon transitions of the hydrogenlike ions, which has been utilized elsewhere within both, the nonrelativistic [3] and relativistic framework [5,7]. Instead, we restrict ourselves to a short account on the angle-angle correlation function for the emitted photons. For angle-resolved studies, it is convenient to start from the *helicity* representation of the photon field, i.e., to describe the photons in terms of the wave vectors $\boldsymbol{k}_{1,2}$ and their spin projection onto the direction of propagation $\lambda_{1,2}=\pm 1$. In such a representation of the photon states, the wave vectors $\boldsymbol{k}_i = (k_i, \theta_i, \phi_i)$, $i=1, 2$ define not only the direction of the emitted photons $\hat{\boldsymbol{k}}_i \equiv (\theta_i, \phi_i)$ but also their energy $E_{\gamma_i} = \hbar c k_i$ which are related by

$$E_i - E_f = E_{\gamma_1} + E_{\gamma_2}, \tag{1}$$

to the total energies E_i and E_f of the initial $|n_i j_i\rangle$ and the final $|n_f j_f\rangle$ (one-electron) states, respectively. Moreover, since the conservation of energy (1) permits only one of the photon energies to be independent, the *complete* information on the energy and angular distributions of the two decay photons may be obtained simply from the (triple-differential) cross section

$$\frac{d^3W}{dE_1 d\Omega_1 d\Omega_2} = \frac{E_{\gamma_1}E_{\gamma_2}}{(2\pi)^3 c^2}\frac{1}{2j_i+1}\sum_{\mu_i\mu_f\lambda_1\lambda_2}|M_{fi}(\mu_f,\mu_i,\lambda_1,\lambda_2)|^2, \tag{2}$$

which is *double* differential in the emission angles and *single* differential in the energy of one of the photons. In Eq. (2), in addition, we have assumed that the excited ions are initially unpolarized and that neither the polarization of the photons $\lambda_{1,2}$ is observed nor that of the residual ion μ_f.

For any further analysis of the energy or angular distribution (2) of the emitted photons, we need to evaluate and simplify the bound-bound transition amplitude $M_{fi}(\mu_f,\mu_i,\lambda_1,\lambda_2)$ which, in second-order perturbation theory, is given by [5]

$$\begin{aligned} M_{fi}(\mu_f,\mu_i,\lambda_1,\lambda_2) &= \sum\!\!\!\!\!\!\int_\nu \frac{\langle\psi_{n_f j_f\mu_f}|\boldsymbol{\alpha}\mathbf{u}^*_{\lambda_1}e^{-i\boldsymbol{k}_1\cdot\boldsymbol{r}}|\psi_\nu\rangle\langle\psi_\nu|\boldsymbol{\alpha}\mathbf{u}^*_{\lambda_2}e^{-i\boldsymbol{k}_2\cdot\boldsymbol{r}}|\psi_{n_i j_i\mu_i}\rangle}{E_\nu - E_i + E_{\gamma_2}} \\ &+ \sum\!\!\!\!\!\!\int_\nu \frac{\langle\psi_{n_f j_f\mu_f}|\boldsymbol{\alpha}\mathbf{u}^*_{\lambda_2}e^{-i\boldsymbol{k}_2\cdot\boldsymbol{r}}|\psi_\nu\rangle\langle\psi_\nu|\boldsymbol{\alpha}\mathbf{u}^*_{\lambda_1}e^{-i\boldsymbol{k}_1\cdot\boldsymbol{r}}|\psi_{n_i j_i\mu_i}\rangle}{E_\nu - E_i + E_{\gamma_1}}. \end{aligned} \tag{3}$$

Here, the transition operator $\alpha\mathbf{u}_{\lambda_i}e^{i\boldsymbol{k}_i\cdot\boldsymbol{r}}$ describes the (relativistic) electron-photon interaction together with the unit vectors $\mathbf{u}_{\lambda_i}$ in order to denote the polarization of the individual photons. As indicated in formula (3), the summation over the intermediate states runs over the complete one-particle spectrum (ψ_ν, E_ν), including a summation over discrete part of the spectrum as well as the integration over the continuum. During the last decade, the *relativistic* form of the transition amplitude (3) has been used widely for studying the two-photon decay in high-Z hydrogenlike ions [5,7]. In such a relativistic description of the hydrogenlike ions, the initial (one-electron) states $\psi_{n_i j_i\mu_i}(\boldsymbol{r}) = \langle\boldsymbol{r}|n_i j_i \mu_i\rangle$ and final states $\psi_{n_f j_f\mu_f}(\boldsymbol{r}) = \langle\boldsymbol{r}|n_f j_f \mu_f\rangle$ are the analytically well-known solutions of the Dirac Hamiltonian for a singly bound electron [15].

In practice, of course, the *summation* over the complete spectrum is difficult to be performed explicitly. Apart from a limited summation over just a few intermediate states which are near in energy therefore a number of alternative methods have been proposed for calculating the second-order amplitudes (3). In the *discrete-basis-set* method, for instance, a finite set of pseudostates is determined variationally and utilized for carrying out the summation [6]. This method has been widely used during the past years in order to explore the two-photon decay of the metastable $2s_{1/2}$ state in heavy hydrogenlike ions [5,7]. An alternative approach is given, if the transition amplitude (3) is first expressed by means of Green's functions, which help avoid the direct summation. In the following section, we make use of this Green's-function method to evaluate the two-photon transition amplitude (3). For studying hydrogenlike ions, the great advantage of this method is that the Coulomb-Green's functions are known analytically, both within the nonrelativistic as well as relativistic theory [16].

B. Green's-function approach

Following Morse and Feshbach [17], we may introduce the formal solution of the Green's function for some Hamiltonian $\hat{\boldsymbol{H}}$, i.e., for the equation $(\hat{\boldsymbol{H}}-E)G_E(\boldsymbol{r},\boldsymbol{r}')=\delta(\boldsymbol{r}-\boldsymbol{r}')$, by

$$G_E(\boldsymbol{r},\boldsymbol{r}') = \sum\!\!\!\!\!\!\int_\nu \frac{|\psi_\nu(\boldsymbol{r})\rangle\langle\psi_\nu(\boldsymbol{r}')|}{E_\nu - E}, \tag{4}$$

which includes a summation (integration) over the complete spectrum of the Hamiltonian as discussed above. Using this ansatz therefore we can replace the summation over the intermediate states in Eq. (3) and rewrite the two-photon transition amplitude in the form

$$\begin{aligned} M_{fi}(\mu_f,\mu_i,\lambda_1,\lambda_2) &= \Big\langle\psi_{n_f j_f\mu_f}(\boldsymbol{r})\Big|\boldsymbol{\alpha}\mathbf{u}^*_{\lambda_1}e^{-i\boldsymbol{k}_1\cdot\boldsymbol{r}}G_{E_i-E_{\gamma_2}}(\boldsymbol{r},\boldsymbol{r}') \\ &\quad\times\boldsymbol{\alpha}\mathbf{u}^*_{\lambda_2}e^{-i\boldsymbol{k}_2\cdot\boldsymbol{r}'}\Big|\psi_{n_i\kappa_i\mu_i}(\boldsymbol{r}')\Big\rangle \\ &\quad+\Big\langle\psi_{n_f j_f\mu_f}(\boldsymbol{r})\Big|\boldsymbol{\alpha}\mathbf{u}^*_{\lambda_2}e^{-i\boldsymbol{k}_2\cdot\boldsymbol{r}}G_{E_i-E_{\gamma_1}}(\boldsymbol{r},\boldsymbol{r}') \\ &\quad\times\boldsymbol{\alpha}\mathbf{u}^*_{\lambda_1}e^{-i\boldsymbol{k}_1\cdot\boldsymbol{r}'}\Big|\psi_{n_i\kappa_i\mu_i}(\boldsymbol{r}')\Big\rangle, \end{aligned} \tag{5}$$

including the integration over both, $\boldsymbol{r}$ and $\boldsymbol{r}'$, respectively. Although, from a mathematical viewpoint, this form is equivalent to the second-order amplitude (3) and (5) appears to be more convenient for the calculation of these amplitudes if the Green's function (4) is known. For the Dirac Hamiltonian and a pure Coulomb potential, i.e., for the hydrogenlike ions, the *radial-angular* representation of the (relativistic) Coulomb-Green's function is given by [16,18]

$$G_E(\boldsymbol{r},\boldsymbol{r}') = \frac{1}{rr'}\sum_{\kappa m}\begin{pmatrix} g^{LL}_{E\kappa}(r,r')\Omega_{\kappa m}(\hat{\boldsymbol{r}})\Omega^\dagger_{\kappa m}(\hat{\boldsymbol{r}}') & -ig^{LS}_{E\kappa}(r,r')\Omega_{\kappa m}(\hat{\boldsymbol{r}})\Omega^\dagger_{-\kappa m}(\hat{\boldsymbol{r}}') \\ ig^{SL}_{E\kappa}(r,r')\Omega_{-\kappa m}(\hat{\boldsymbol{r}})\Omega^\dagger_{\kappa m}(\hat{\boldsymbol{r}}') & g^{SS}_{E\kappa}(r,r')\Omega_{-\kappa m}(\hat{\boldsymbol{r}})\Omega^\dagger_{-\kappa m}(\hat{\boldsymbol{r}}')\end{pmatrix}, \tag{6}$$

where $\Omega_{\kappa m}(\hat{\boldsymbol{r}})$ denote a Dirac spinor [15], and where the *radial* Green's function is given in terms of the four components $g^{TT'}_{E\kappa}(r,r')$ with $T=L,S$ referring to the *large* and *small* components of the associated (relativistic) wave functions. For the sake of brevity, here we will not display the radial components $g^{TT'}_{E\kappa}(r,r')$ explicitly but just recall that they can be expressed in terms of the (special) Whittaker functions of the first and second kind [16,18]. In the computations below, we have used the GREENS library [18] in order to obtain the energy and angular distributions for the emitted photons. This code was developed by us originally for studying the cross sections and polarization phenomena in the two-photon ionization of the hydrogenlike ions [19].

C. Multipole decomposition of the photon fields

Making use of Eq. (5), the computation of the two-photon amplitudes requires a six-*dimensional* integration over $d^3\boldsymbol{r} = r^2 dr \sin\theta d\theta d\phi$ and $d^3\boldsymbol{r}' = r'^2 dr' \sin\theta' d\theta' d\phi'$, respectively. As usual in atomic physics, however, these integrals can be further simplified by applying the techniques of Racah's algebra if all the operators are represented in terms of spherical tensors and if the *radial-angular* representation of the wave and Green's functions are used [18]. For the interaction of electrons with the radiation field, the spherical tensor components are obtained from a multipole expansion of the photon operator [20],

$$\mathbf{u}_{\lambda_{1,2}} e^{i\boldsymbol{k}_{1,2}\cdot\boldsymbol{r}} = \sqrt{2\pi}\sum_{L=1}^{\infty}\sum_{M} i^L[L]^{1/2}\left(\boldsymbol{A}^{(m)}_{LM} + i\lambda_{1,2}\boldsymbol{A}^{(e)}_{LM}\right) \times D^L_{M\lambda_{1,2}}(\hat{\boldsymbol{k}}_{1,2}\to\hat{\boldsymbol{z}}), \tag{7}$$

where $[L]=(2L+1)$ and the standard notation $\boldsymbol{A}^{(e,m)}_{LM}$ is used to denote the electric and magnetic multipole fields, respectively. Each of these multipoles can be expressed in terms of the spherical Bessel functions $j_L(kr)$ and the vector spherical harmonics $\boldsymbol{T}^M_{L,\Lambda}$ of rank L as [20]

$$\boldsymbol{A}^{(m)}_{LM} = j_L(kr)\boldsymbol{T}^M_{L,L},$$

$$\boldsymbol{A}^{(e)}_{LM} = j_{L-1}(kr)\sqrt{\frac{L+1}{2L+1}}\boldsymbol{T}^M_{L,L-1} - j_{L+1}(kr)\sqrt{\frac{L}{2L+1}}\boldsymbol{T}^M_{L,L+1}. \tag{8}$$

As seen from Eq. (7), the angular dependence of the photon emission results from the the Wigner (rotation) matrices $D^L_{M\lambda_{1,2}}(\hat{\boldsymbol{k}}_{1,2}\to\hat{\boldsymbol{z}})$ which transform the multipole fields with the original quantization axis along the photon propagation $\boldsymbol{k}_{1,2}$ into the fields with quantization axis along the $\hat{\boldsymbol{z}}$ direction.

The proper choice of a *common* quantization axis ($\hat{\boldsymbol{z}}$ axis) is important for the evaluation of the angular integrals in the transition amplitude (5), cf. the discussion in Refs. [19,21]. Since for the decay of unpolarized (as well as unaligned) hydrogenlike ions, there is *no* direction preferred for the overall system, we adopted the $\hat{z}$ axis along the momentum of the "first" photon: $\hat{\boldsymbol{z}}\parallel\hat{\boldsymbol{k}}_1$. Making use of the expressions (7) and (8) for such a choice of the quantization axis, we can rewrite the two-photon transition amplitude (5) as a sum over the electric and magnetic multipole components,

$$\begin{aligned} M_{fi}&(\mu_f,\mu_i,\lambda_1,\lambda_2) \\ &= 2\pi\sum_{L_1\Lambda_1}\sum_{L_2M_2\Lambda_2} i^{-L_1-L_2}[L_1,L_2]^{1/2}D^{L*}_{M_2\lambda_2}(\hat{\boldsymbol{k}}_2)\xi^{\lambda_1 *}_{L_1\Lambda_1}\xi^{\lambda_2 *}_{L_2\Lambda_2} \\ &\quad\times\Big[\langle\psi_{n_fj_f\mu_f}(\boldsymbol{r})|\boldsymbol{\alpha}j_{\Lambda_1}(k_1r)\boldsymbol{T}^{\lambda_1 *}_{L_1\Lambda_1}G_{E_i-E_{\gamma_2}}(\boldsymbol{r},\boldsymbol{r}') \\ &\quad\times\boldsymbol{\alpha}j_{\Lambda_2}(k_2r')\boldsymbol{T}^{M_2 *}_{L_2\Lambda_2}|\psi_{n_i\kappa_i\mu_i}(\boldsymbol{r}')\rangle \\ &\quad+\langle\psi_{n_fj_f\mu_f}(\boldsymbol{r})|\boldsymbol{\alpha}j_{\Lambda_2}(k_2r)\boldsymbol{T}^{M_2 *}_{L_2\Lambda_2}G_{E_i-E_{\gamma_1}}(\boldsymbol{r},\boldsymbol{r}') \\ &\quad\times\boldsymbol{\alpha}j_{\Lambda_1}(k_1r')\boldsymbol{T}^{\lambda_1 *}_{L_1\Lambda_1}|\psi_{n_i\kappa_i\mu_i}(\boldsymbol{r}')\rangle\Big], \end{aligned} \tag{9}$$

where the coefficients $\xi^{\lambda}_{L\Lambda}$ are given by [19]

$$\xi^{\lambda}_{L\Lambda} = \begin{cases} 1 & \text{if } \Lambda = L, \\ i\lambda\sqrt{\dfrac{L+1}{2L+1}} & \text{if } \Lambda = L-1, \\ -i\lambda\sqrt{\dfrac{L}{2L+1}} & \text{if } \Lambda = L+1. \end{cases} \tag{10}$$

As expected from expansion (7) of the photon operator, the transition amplitude (9) now contains an (infinite) summation over products of the different multipoles $E1E1, E1M2, M1M1, E2M2,\ldots$, which are characterized by the combination of the summation indices L, L', Λ, Λ' or, equivalently, by the symmetry of the vector spherical harmonics in the overall expansion. Each matrix element in the summation (9) still represents a six-dimensional integral over $\boldsymbol{r}$ and $\boldsymbol{r}'$, respectively, along with a summation over the spinor components of the wave and Green's functions. To separate the radial part from the spin-angular parts of these matrix elements, we further need to apply the radial-angular representation (6) of the Coulomb-Green's functions together with the known (two-component) representation of the Dirac wave functions in a Coulomb field [15]. After some algebra, we then obtain for the transition amplitude

$$M_{f_i}(\mu_f,\mu_i,\lambda_1,\lambda_2) = 2\pi \sum_{L_2 M_2} D^{L_2 *}_{M_2 \lambda_2}(\hat{\boldsymbol{k}}_2) \sum_{L_1 \Lambda_1 \Lambda_2} i^{-L_1 - L_2}[L_1,L_2]^{1/2} \xi^{\lambda_1 *}_{L_1 \Lambda_1} \xi^{\lambda_2 *}_{L_2 \Lambda_2} \sum_{\kappa m T T'} P^T P^{T'} \times \Big[U^{TT'}_{\Lambda_1 \Lambda_2}(\kappa_f,\kappa,\kappa_i;k_1,k_2;E_{\gamma_2}) \langle \kappa_f l_f^{\overline{T}} \mu_f | \boldsymbol{\sigma T}^{\lambda_1 *}_{L_1 \Lambda_1} | \kappa l^T m \rangle \langle \kappa l^{T'} m | \boldsymbol{\sigma T}^{M_2 *}_{L_2 \Lambda_2} | \kappa_i l_i^{\overline{T'}} \mu_i \rangle + U^{TT'}_{\Lambda_2 \Lambda_1}(\kappa_f,\kappa,\kappa_i;k_2,k_1;E_{\gamma_1}) \times \langle \kappa_f l_f^{\overline{T}} \mu_f | \boldsymbol{\sigma T}^{M_2 *}_{L_2 \Lambda_2} | \kappa l^T m \rangle \langle \kappa l^{T'} m | \boldsymbol{\sigma T}^{\lambda_1 *}_{L_1 \Lambda_1} | \kappa_i l_i^{\overline{T'}} \mu_i \rangle \Big], \tag{11}$$

where, again, $T=L,S$ is used to denote the large and small components, for which factor P^T is defined as $P^L=1$ and $P^S=-1$, and where $\overline{T}$ refers to the conjugate of T, i.e., $\overline{T}=L$ for $T=S$ and vice versa. Equation (11) displays the general form of the (relativistic) transition amplitude for the two-photon excitation or decay of hydrogenlike ions. The *angular* part of this amplitude is determined by the matrix elements of the rank L spherical tensors $\boldsymbol{\sigma T}^M_{L\Lambda}=[Y_\Lambda \otimes \sigma]^M_L$ which have been discussed previously [19,21]. In contrast to the angular part which can be evaluated algebraically, however, the *radial* integrals,

$$U^{TT'}_{\Lambda_1 \Lambda_2}(\kappa_f,\kappa,\kappa_i;k_1,k_2;E_\gamma) = \int g^{\overline{T}}_{n_f \kappa_f}(r) j_{\Lambda_1}(k_1 r) g^{TT'}_{E_b + E_\gamma \kappa}(r,r') j_{\Lambda_2}(k_2 r') g^{\overline{T'}}_{n_i \kappa_i}(r') dr\, dr', \tag{12}$$

in Eq. (11) have to be computed *numerically.* To this end, we first generate the individual components of the (radial) wave functions $g^T_{n_{i,f}\kappa_{i,f}}(r)$ and the Green's functions $g^{TT'}_{E\kappa}(r,r')$ and then carry out the two-dimensional integration over r and r'. In the present work, all the required functions and radial integrals were calculated by using the GREENS library [18].

III. RESULTS AND DISCUSSION

The two-photon transition amplitude (11) formally includes the summation over all the products of multipoles E1E1, E1M1, E1E2,... . Although still infinite, the number of *allowed* (i.e., nonzero) terms in the fourfold summation of Eq. (11) is restricted to certain combinations of the indices L_1, L_2, and Λ_1, Λ_2, respectively, owing to the parity and angular-momentum selection rules as obtained from the angular parts of the matrix elements [21]. Here, we shall not discuss these selection rules which follow very similar lines as the one-photon transitions, if the *symmetry* of the imaginary intermediate states is taken into account. In practice, there is usually one "leading" term in the expansion which dominates the two-photon transitions for any given pair of initial and final bound states. For example, the $2s_{1/2}$ level of the hydrogenlike ions decays into the $1s_{1/2}$ ground state primarily by the emission of two electric-dipole (E1E1) photons, while all the higher multipoles contribute with less than 0.5% to the total decay rate [5,7]. In contrast to the total rates, a more significant effect of the higher multipoles (sometimes called the *nondipole* effects), are expected for the *angular* distributions of the emitted photons. For heavy hydrogenlike ions, for instance, it was shown recently that the magnetic-quadrupole (M2) contribution shifts the effective anisotropy parameters by up to 30%, when compared with a pure electric-dipole approximation [22].

In this contribution, we make use of Eqs. (2) and (11) in order to explore the nondipole effects on the angular distributions for the two-photon decay of hydrogenlike ions. This first requires, or course, to define the *geometry* under which the emission of the two photons is considered. As described in Sec. II C, we adopt the quantization axis (z axis) along the momentum of the first photon $\hat{\boldsymbol{k}}_1$, so that the *angular correlation* between the two photons is characterized by the (polar) angle $\theta_2 \equiv \theta$ of the second-photon momentum with respect to this axis. Apart from the angle θ, the photon-photon correlation function also depends on the energies E_1 and E_2 of the photons. Instead of using these (absolute) energies, however, it is often more convenient to characterize the photon energy distribution in term of the dimensionless variable $x=E_{\gamma_1}/(E_{\gamma_1}+E_{\gamma_2})=E_{\gamma_1}/(E_i-E_f)$ which gives the fraction of

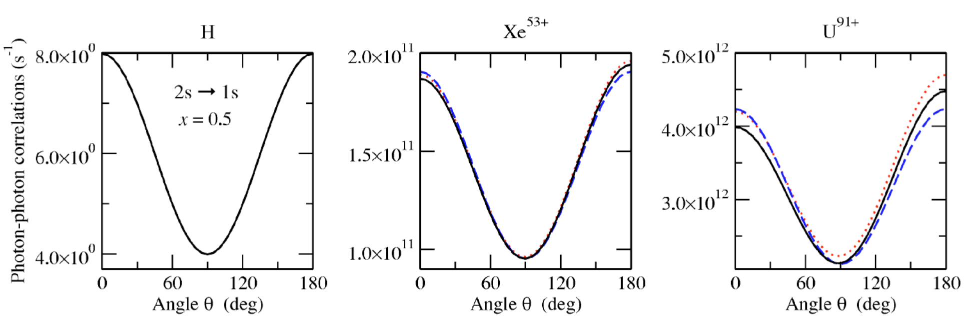

FIG. 1. Photon-photon angular correlations (13) in the $2s_{1/2} \rightarrow 1s_{1/2}$ two-photon) decay of the hydrogenlike ions. Results are presented for the exact relativistic theory (—), nonrelativistic approximation (---) by Au 4 as well as the relativistic electric dipole (--) approach for the relative photon energy $x=0.5$.

TABLE I. Intensity ratio $dW/dxd\cos\theta(\theta=180°)/dW/dxd\cos\theta(\theta=0°)$ for the $2s_{1/2}\rightarrow 1s_{1/2}$ two-photon decay in the hydrogenlike ions, taken at the relative photon energy $x=0.5$. Results from the exact relativistic computations using Eq. (11) are compared with the nonrelativistic approach by Au [4] and the (trivial) electric dipole approximation $1+\cos^2\theta$.

	H	Xe^{53+}	U^{91+}
Rel. exact	1.000	1.038	1.124
Nonrel. exact	1.000	1.079	1.281
Electric dipole	1.000	1.000	1.000

the energy as carried away by one of the two photons [5]. Using this variable, the angle- and energy-differential cross section (2) can be written as

$$\frac{d^2W}{dxd\cos\theta}(x,\theta)=8\pi^2(E_i-E_f)\frac{d^3W}{dE_{\gamma_1}d\Omega_1 d\Omega_2}, \qquad (13)$$

which is the most appropriate form for studying the photon-photon angular correlations. In this expression, the factor $8\pi^2$ arises from the integration over the solid angle $d\Omega_1=\sin\theta_1 d\theta_1 d\phi_1$ of the first photon as well as the integration over the aximuthal angle $d\phi_2$ of the second photon.

In Fig. 1, we display the photon-photon angular correlation function (13) for the $2s_{1/2}\rightarrow 1s_{1/2}$ (two-photon) decay of neutral hydrogen H, hydrogenlike xenon Xe^{53+} and uranium U^{91+} ions, taken at the (relative) photon energy $x=0.5$. To explore the nondipole effects in the angular correlation of the two photons, computations were performed within both the exact relativistic theory (solid line), which includes the summation over all multipole components in the amplitude (11), as well as the electric dipole approximation (dashed line) obtained by restricting the summation to $L=L'=1$ and $\Lambda,\Lambda'=0$, 2, respectively. Figure 1 also shows the results by Au [4] (short-dashed line) who incorporated some of the higher multipoles within his nonrelativistic approach. For neutral hydrogen, the nonrelativistic and relativistic results basically coincide and are well described by the angular distribution $1+\cos^2\theta$ [3,4]. That is, the angular correlation between the two photons is symmetric with respect to the angle $\theta=90°$ for the low-Z ions, a behavior which remains preserved along the isoelectronic sequence, if the (relativistic) dipole approximation (E1E1) is applied. In an *exact* calculation, in contrast, the photon emission of the second electron occurs predominantly into the backward direction (i.e., for $\theta>90°$) if taken relative to the first photon. Therefore our computations show that the nondipole terms in the electron-photon interaction give rise to an *asymmetric* shift in the photon-photon angular correlations which becomes larger as the nuclear charge Z is increased. Including the higher multipoles into the photon-photon correlation function, an asymmetry is found both within the nonrelativistic and relativistic theory. As seen from Table I, however, there are differences in the size of the asymmetry which become pronounced, in particular, for the parallel ($\theta=90°$) and back-to-back ($\theta=180°$) photon emission, and if considered for high-Z ions. For hydrogenlike uranium U^{91+} ions, for instance, the nonrelativistic calculation by Au [4] overestimates the effects of the higher multipoles by more than 13% when compared with the exact relativistic treatment.

In Fig. 1, the photon-photon angular correlations is calculated for the relative energy $x=0.5$ which refers to an equal energy of the two photons: $E_{\gamma_1}=E_{\gamma_2}$. While an equal energy sharing is the most likely one for the $2s_{1/2}\rightarrow 1s_{1/2}$ decay of hydrogenlike ions [5,7], photons with different energies $E_{\gamma_1}\neq E_{\gamma_2}$ have been observed in a number of experiments [10], as long as the total energy remains conserved in Eq. (1). Therefore a series of computations have been carried out for a number of (relative) energies x. For example, Fig. 2 displays the energy dependence of the differential decay rate (13) for hydrogenlike uranium U^{91+} ions and for the three relative photon energies $x=0.1$, 0.3, and 0.5, respectively. In this figure, again, we present the results from our relativistic exact and electric-dipole approximation as well as from the nonrelativistic computations by Au [4]. As seen from Fig. 2, the deviations of Au's nonrelativistic approach increase for small values of x. Comparing the two relativistic approximations, in contrast, the effects of the higher multipoles are largest for the emission of two photons with equal energy (i.e., for $x=0.5$).

Until now, we always considered the $2s_{1/2}\rightarrow 1s_{1/2}$ two-photon decay of the hydrogenlike ions. Apart from this experimentally well established transition, a two-photon emission has been observed also for the $3d_{5/2}\rightarrow 1s_{1/2}$ decay [10,23]. Similar as for the $2s_{1/2}$ level, the $3d_{5/2}$ dominantly decays along the $E1E1$ dipole-dipole mode. In the electric-dipole approximation, we therefore expect an angular correlation function of the form $1+\beta\cos^2\theta$, which is symmetric

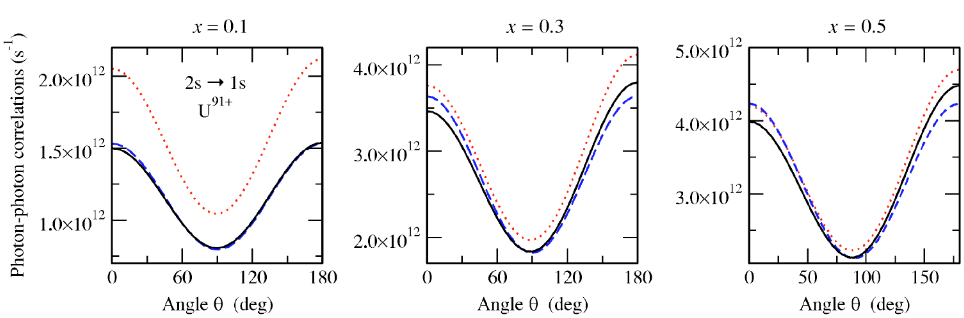

FIG. 2. Photon-photon angular correlations (13) in the $2s_{1/2}\rightarrow 1s_{1/2}$ (two-photon) decay of the hydrogenlike uranium U^{91+}. The correlation functions are shown for the three relative photon energies $x=0.1$, 0.3, and 0.5. See Fig. 1 for further details.

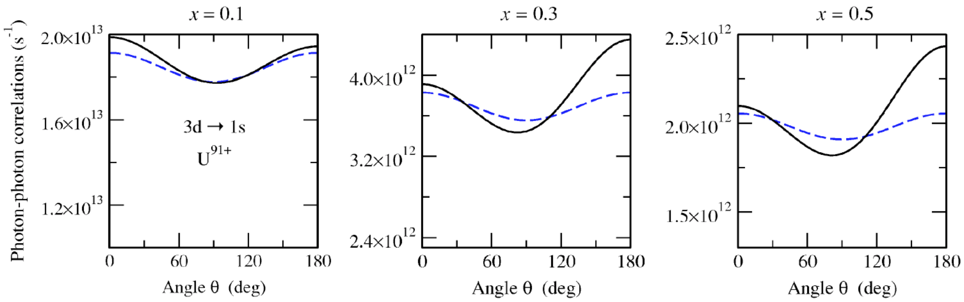

FIG. 3. Photon-photon angular correlations (13) in the $3d_{5/2} \rightarrow 1s_{1/2}$ (two-photon) decay of the hydrogenlike uranium U^{91+}. The correlation functions are shown for the three relative photon energies $x=0.1$, 0.3, and 0.5. See Fig. 1 for further details.

with respect to the angle $\theta=90°$ and where the anisotropy parameter $\beta=1/13$ [14,24], cf. Fig. 3. In contrast to the $2s_{1/2}$ decay, however, a much stronger effect arises here for the $3d_{5/2} \rightarrow 1s_{1/2}$ transition because of the higher multipoles. For the two-photon decay of the hydrogenlike uranium ions U^{91+}, for instance, the back-to-back photon emission is increased by almost 20% if the higher multipoles are taken into account.

IV. SUMMARY AND OUTLOOK

In conclusion, the *angular correlation functions* in the two-photon decay of hydrogenlike ions have been studied within the framework of second-order perturbation theory, based on Dirac's relativistic equation. Summation over the intermediate ion states, which occur in this framework, has been performed by means of the *relativistic* Coulomb-Green's functions. In addition, a complete expansion of the radiation field in terms of its multipole components has been made in order to incorporate the nondipole effects. Detailed calculations are performed for the photon angular distribution as function of the angle between the photons for the two-photon decay of the $2s_{1/2}$ and $3d_{5/2}$ levels and are shown for the decay of neutral hydrogen as well as for hydrogenlike Xe^{53+} and U^{91+} ions. As seen from the results obtained [cf. Fig. 1–3], the photon-photon correlation functions are much more sensitive to the contribution of the higher multipoles than found for the total rates. The higher multipoles of the radiation field typically result in an asymmetric shift of the photon-photon correlation function which is enhanced as the nuclear charge Z increases.

Apart from the higher-multipole contributions, we have also explored how the *relativistic contraction* of the electronic wave functions affects the angular correlations of the emitted photons. From the comparison of our (relativistic) computations with the *nonrelativistic* results by Au [4], we found that the contraction of the wave functions towards the nucleus often lowers the angular-differential and hence also the total cross sections in the high-Z domain. Overall, there is a partial cancellation between the relativistic and the nondipole terms in the two-photon amplitudes which decreases slightly also the asymmetrical shift in the photon-photon angular correlations.

In the present work, we have restricted our computations on the two-photon transitions to the *hydrogenlike* ions. For these ions, our results show that the Green's function approach provide a reliable and efficient access to the (second-order) amplitudes and hence also to various related properties such as the total decay rates, angular- and energy-differential cross sections, and to several others. In the future therefore we plan to extend this approach for studying the two-photon decay of many-electron systems. A first case study on the (two-photon) decay of a single K-shell vacancy in neutral heavy atoms is currently under way.

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