Photon pairs with tailor-made entanglement obtained from the two-photon decay of atomic hydrogen

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From the work by W. Perrie *et al.* [Phys. Rev. Lett. **54**, 1790 (1985)], it is known that the photon pairs that are emitted in the $2s_{1/2} \rightarrow 1s_{1/2}$ (two-photon) decay of atomic hydrogen are quantum mechanically correlated, i.e., entangled. However, less information is available about the degree of polarization entanglement between the two photons if an arbitrary geometry is considered for collecting the photons. In this paper, we study the effect of the decay geometry on the degree of polarization entanglement between the two emitted photons. Results are shown for the $2s_{1/2} \rightarrow 1s_{1/2}$ and $3d_{5/2} \rightarrow 1s_{1/2}$ two-photon transitions of atomic hydrogen. The outlined theory is general and can be applied also to heavier elements. To demonstrate the influence of relativistic and multipole effects, results are also shown for the $3d_{5/2} \rightarrow 1s_{1/2}$ transition of hydrogenlike uranium.

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I. INTRODUCTION

For a long time, nonlocality and entanglement have been known as puzzling features of quantum mechanics, incompatible with our "classical view" that all phenomena in nature should be local and realistic. Since the pioneering work of Bell [1], therefore, many experiments have aimed to test and understand these counterintuitive implications of quantum theory [2–4]. Only rather recently, however, the entanglement of composite quantum systems has been found an important resource for establishing novel quantum information protocols, such as quantum cryptography [5], teleportation [6], or for developing quantum algorithms [7,8] that may outperform any classical information processing in the future.

Due to this new interest in quantum entanglement, an active research program has been initiated during the last decade in order to explore physical systems that are suitable for producing and controlling entanglement. Historically, among the first processes available for entanglement studies were photon pairs that can be generated, for example, via cascaded two-photon emission as it was done in the first photon entanglement experiments [3].

In this work, we (re)consider a similar process that is also known to produce entangled photon pairs: The two-photon decay of metastable atoms and ions and, especially, atomic hydrogen with its well-known $2s_{1/2} \rightarrow 1s_{1/2}$ and $3d_{5/2} \rightarrow 1s_{1/2}$ $2E1$ decay as perhaps the simplest of these systems. So far, most of the studies of this process were focusing on the total decay rates [9,10], the energy distributions between the photons [11,10,12], and the angular correlation [13,14]. Later, atomic hydrogen (deuterium) has also been used to study the polarization correlation between the emitted photons and to test Bell's inequality [15–17]; but although the generation of quantum mechanically correlated photons has been demonstrated in these experiments, the geometrical properties of the emitted photons' polarization entanglement have not been analyzed in detail so far. Moreover, one should note that although the violation of Bell's inequality can serve as an indicator for entanglement [18], it is usually not suitable to *quantify* the amount of entanglement. This is due to the fact that, in general, nonlocality (in the sense of violating Bell's inequality) is not equivalent to entanglement (in the sense of nonseparability) as there exist entangled quantum states which do not violate any Bell-type inequality [19]. Therefore, in the following analysis, we will restrict ourselves to the widely accepted concurrence measure of entanglement which is strictly greater than zero for *all* entangled two-qubit states [20].

Our analysis of the photon-photon entanglement is based on the density matrix of the "atom+photon pair" that is described in Sec. II. There, we give a brief overview of the relativistic Green's function approach to calculate the final two-photon density matrix. This approach is general enough to be applied also to heavier systems. We also provide a simplified formula for the nonrelativistic dipole approximation. In Sec. III, we present the results for the $2s_{1/2} \rightarrow 1s_{1/2}$ and $3d_{5/2} \rightarrow 1s_{1/2}$ decay of atomic hydrogen with different geometries and initial sublevel populations. In order to illustrate the influence of relativistic effects we compare these results to the case of hydrogenlike uranium. Finally, a brief summary is given in Sec. IV.

II. THEORY

A. General bound-bound transition amplitude

The two-photon decay of atoms and ions is naturally described in the framework of second-order perturbation theory and the relativistic Dirac equation. Within this framework, the two-photon transition amplitude is given by [21,22]

$$M(\mu_f, \mu_i, \lambda_1, \lambda_2) = \sum\!\!\!\!\!\!\int_\nu \frac{\langle \psi_f | \mathbf{A}_1^* | \psi_\nu \rangle \langle \psi_\nu | \mathbf{A}_2^* | \psi_i \rangle}{E_\nu - E_i + E_{\gamma_2}} + \sum\!\!\!\!\!\!\int_\nu \frac{\langle \psi_f | \mathbf{A}_2^* | \psi_\nu \rangle \langle \psi_\nu | \mathbf{A}_1^* | \psi_i \rangle}{E_\nu - E_i + E_{\gamma_1}}, \quad (1)$$

where E_i and E_f are the eigenvalues, and $\psi_i(\boldsymbol{r}) \equiv \psi_{n_i j_i \mu_i}(\boldsymbol{r})$ and $\psi_f(\boldsymbol{r}) \equiv \psi_{n_f j_f \mu_f}(\boldsymbol{r})$ are the well-known solutions of the Dirac Hamiltonian for a single electron, if bound to the nucleus. Because of energy conservation, the energies E_i and E_f are related to the energies $E_{\gamma_{1,2}}$ of the emitted photons by $E_i - E_f = E_{\gamma_1} + E_{\gamma_2}$. From this one can define the energy sharing $x = E_{\gamma_1}/(E_{\gamma_1} + E_{\gamma_2})$ between the emitted photons. For photons propagating with wave vector $\boldsymbol{k}_i$ $(i=1,2)$ and unit polarization vector $\boldsymbol{u}_{\lambda_i}$ $(\boldsymbol{k}_i \cdot \boldsymbol{u}_{\lambda_i} = 0)$, moreover, the electron-photon interaction operator $\boldsymbol{A}_i$ in the transition amplitude (1) can be written in velocity gauge as

$$\mathbf{A}_i = \boldsymbol{\alpha} \boldsymbol{u}_{\lambda_i} \mathrm{e}^{i \boldsymbol{k}_i \boldsymbol{r}}, \tag{2}$$

where $\lambda_i = \pm 1$ denotes the *helicity*, i.e., the spin projection of the photon upon the direction $\boldsymbol{k}$ of propagation. As usual, the helicity ± 1 corresponds to a circular polarization $\sigma^{\pm}$ of the emitted photons.

As seen from Eq. (1), the evaluation of the second-order transition amplitude requires the summation over the "complete" spectrum of the system, including a summation over the discrete part of the spectrum as well as the integration over the continuum. For atomic hydrogen (or hydrogenlike ions), this summation can be carried out either explicitly [21] or by using the Green's function of the atom [14,23,24]:

$$G_E(\boldsymbol{r},\boldsymbol{r}') = \sum\!\!\!\!\!\!\int_{\nu} \frac{|\psi_\nu\rangle\langle\psi_\nu|}{E_\nu - E}. \tag{3}$$

Substituting Eq. (3) into Eq. (1) and making use of Eq. (2), we can rewrite the two-photon transition amplitude in the form

$$\begin{aligned} M_{fi}(\mu_f,\mu_i,\lambda_1,\lambda_2) &= \langle \psi_{n_f j_f \mu_f}(\boldsymbol{r}) | \boldsymbol{\alpha} \cdot \boldsymbol{u}^*_{\lambda_1} e^{-i\boldsymbol{k}_1 \cdot \boldsymbol{r}} G_{E_i - E_{\gamma_2}}(\boldsymbol{r},\boldsymbol{r}') \boldsymbol{\alpha} \cdot \boldsymbol{u}^*_{\lambda_2} \\ &\times e^{-i\boldsymbol{k}_2 \cdot \boldsymbol{r}'} | \psi_{n_i j_i \mu_i}(\boldsymbol{r}') \rangle + \langle \psi_{n_f j_f \mu_f}(\boldsymbol{r}) | \boldsymbol{\alpha} \cdot \boldsymbol{u}^*_{\lambda_2} e^{-i\boldsymbol{k}_2 \cdot \boldsymbol{r}} \\ &\times G_{E_i - E_{\gamma_1}}(\boldsymbol{r},\boldsymbol{r}') \boldsymbol{\alpha} \cdot \boldsymbol{u}^*_{\lambda_1} e^{-i\boldsymbol{k}_1 \cdot \boldsymbol{r}'} | \psi_{n_i j_i \mu_i}(\boldsymbol{r}') \rangle, \end{aligned} \tag{4}$$

which appears to be convenient for studying the properties of the two-photon decay as the Coulomb-Green's function is known analytically for both the relativistic and the nonrelativistic case [23]. For example, the relativistic Coulomb-Green's function takes the form

$$G_E(\boldsymbol{r},\boldsymbol{r}') = \frac{1}{r r'} \sum_{\kappa m} \begin{pmatrix} g^{LL}_{E\kappa}(\boldsymbol{r},\boldsymbol{r}') \Omega_{\kappa m}(\hat{\boldsymbol{r}}) \Omega^\dagger_{\kappa m}(\hat{\boldsymbol{r}}') & -i g^{LS}_{E\kappa}(\boldsymbol{r},\boldsymbol{r}') \Omega_{\kappa m}(\hat{\boldsymbol{r}}) \Omega^\dagger_{-\kappa m}(\hat{\boldsymbol{r}}') \\ i g^{SL}_{E\kappa}(\boldsymbol{r},\boldsymbol{r}') \Omega_{-\kappa m}(\hat{\boldsymbol{r}}) \Omega^\dagger_{\kappa m}(\hat{\boldsymbol{r}}') & g^{SS}_{E\kappa}(\boldsymbol{r},\boldsymbol{r}') \Omega_{-\kappa m}(\hat{\boldsymbol{r}}) \Omega^\dagger_{-\kappa m}(\hat{\boldsymbol{r}}') \end{pmatrix}, \tag{5}$$

where the angular part is written in terms of the Dirac spinors $\Omega_{\kappa m}(\hat{\boldsymbol{r}})$, while the radial part is given by 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 [23].

Together with a multipole decomposition of the photon fields, this radial-angular representation of the Green's function allows us to use Racah's algebra to carry out the angular momentum integrations that are required to evaluate the transition amplitudes (4) (see [14] and references therein for more details). Moreover, note that in Eq. (5), the summation over the relativistic angular momentum quantum number κ is restricted to only a few values due to the selection rules for a given bound-bound transition.

In the following, we shall employ the second-order amplitude (4) in order to analyze in detail the correlated spin states of the two emitted photons. Most naturally, this analysis can be performed by means of the density matrix that is associated to the spin-polarization of the emitted photons. However, since the application of this formalism to the two-photon decay has been discussed elsewhere [24], let us note only that the final-state (reduced) density matrix of the two emitted photons in the helicity representation is given by

$$\begin{aligned} \langle \boldsymbol{k}_1 \lambda_1, \boldsymbol{k}_2 \lambda_2 | \hat{\rho}_f | \boldsymbol{k}_1 \lambda_1', \boldsymbol{k}_2 \lambda_2' \rangle &= \sum_{\mu_i, \mu_i', \mu_f} \langle n_i j_i \mu_i | \hat{\rho}_i | n_i j_i \mu_i' \rangle M_{fi}(\mu_f,\mu_i,\lambda_1,\lambda_2) \\ &\times M^*_{fi}(\mu_f,\mu_i',\lambda_1',\lambda_2'), \end{aligned} \tag{6}$$

where the partial trace has to be taken over the unobserved (final) state of the atom and $\langle n_i j_i \mu_i | \hat{\rho}_i | n_i j_i \mu_i' \rangle$ denotes some generic initial-state density matrix. In the following, this general formulation of the bound-bound transition will allow us to study the effect of different initial populations of the excited state. The theory developed so far applies for the two-photon decay of any atom, if the energies and wave functions in the amplitude (1) are adopted properly for many-electron systems.

B. Nonrelativistic electric dipole approximation

In the previous section, we have outlined a rather general, relativistic theory that is suitable also for the two-photon decay of high-Z systems. However, due to the fundamental role of hydrogen and its experimental accessibility we present also a formula for the final two-photon state in the electric dipole approximation where we assume a two-step $2E1$ transition with only a single intermediate state. With the additional restriction to an initially unpolarized atom (or ion)

and an equal energy sharing $x=0.5$ between the two emitted photons we arrive at the following density matrix for the final two-photon state [28]:

$$\langle \boldsymbol{k}_1\lambda_1,\boldsymbol{k}_2\lambda_2|\hat{\rho}_f|\boldsymbol{k}_1\lambda_1',\boldsymbol{k}_2\lambda_2'\rangle = C\lambda_1\lambda_2\lambda_1'\lambda_2' \sum_{L,\mu_1,\mu_2} D^L_{\mu_1\mu_2}(\boldsymbol{k}) \times\langle 1\lambda_1 1-\lambda_1'|L\mu_1\rangle\langle 1-\lambda_2 1\lambda_2'|L\mu_2\rangle \times\left(\begin{Bmatrix} j_\nu & j_f & 1\\ j_i & j_\nu & 1\\ 1 & 1 & L\end{Bmatrix}+\begin{Bmatrix} 1 & 1 & L\\ j_\nu & j_\nu & j_i\end{Bmatrix}\begin{Bmatrix} 1 & 1 & L\\ j_\nu & j_\nu & j_f\end{Bmatrix}\right), \tag{7}$$

where C is a normalization constant, $\boldsymbol{k}=(\psi,\theta,\phi)$ the direction of the second photon relative to the first photon, j_i (j_f) are the angular momentum quantum numbers of the initial (final) state, and j_ν corresponds to the intermediate atomic state in the two-step decay process. Our approach of taking into account only $p_{3/2}$ intermediate states is justified for the hydrogenic $3d_{5/2}\rightarrow 1s_{1/2}$ but also for the $2s_{1/2}\rightarrow 1s_{1/2}$ transition, this approximation is valid as the intermediate states $p_{1/2}$ and $p_{3/2}$ are degenerate in the nonrelativistic limit. Also note that in Eq. (7) the final two-photon density matrix does not depend explicitly on the radial integrals any more as they can be absorbed into the normalization constant C. Hence Eq. (7) depends only on the decay geometry.

With the help of the simplified two-photon density matrix (7), one can easily reproduce the well-known $1+\cos^2(\theta)$ [9,13] and $1+1/13\cos^2(\theta)$ [25,26] shapes of the angular correlation for the hydrogenic $2s\rightarrow 1s$ and $3d\rightarrow 1s$ decay, respectively. Additionally, for the case of back-to-back emission ($\theta=180°$) during the $2s\rightarrow 1s$ decay, our approximate formula is also in agreement with the quantum mechanical prediction of $\frac{1}{4}[1+\cos(2\varphi)]$ for the linear polarization correlation as reported in [17,27]. Here, φ denotes the relative alignment of the two linear polarizers.

C. Measure of entanglement

Apart from performing angle-resolved studies [24], the helicity representation of the reduced density matrix (6) or (7) is convenient also for obtaining the polarization entanglement between the two photons. For this purpose, of course, we require a proper measure for the entanglement of the "two-qubit" system that is given by the spin-states of the photon pair. Although, for general N-qubit systems, the quantification of entanglement is still a great challenge for current research (see, e.g., Ref. [29] for a recent review on entanglement measures), Wootter's concurrence [20] can be applied for any two-qubit systems, such as a photon pairs, and has been widely used in the literature in order to determine their degree of entanglement. For any two-qubit state $\hat{\rho}$, either pure or mixed, the concurrence is defined as

$$\mathcal{C}(\hat{\rho})=\max(0,\sqrt{e_1}-\sqrt{e_2}-\sqrt{e_3}-\sqrt{e_4}), \tag{8}$$

where $\sqrt{e_i}$ are the square roots of the eigenvalues of the matrix $\hat{\rho}(\hat{\sigma}_2^{(1)}\otimes\hat{\sigma}_2^{(2)})\hat{\rho}^*(\hat{\sigma}_2^{(1)}\otimes\hat{\sigma}_2^{(2)})$ in descending order, $\hat{\rho}^*$ is the complex conjugate of $\hat{\rho}$, and $\hat{\sigma}_2^{(1,2)}$ are the Pauli matrices acting on the first and the second qubit, respectively.

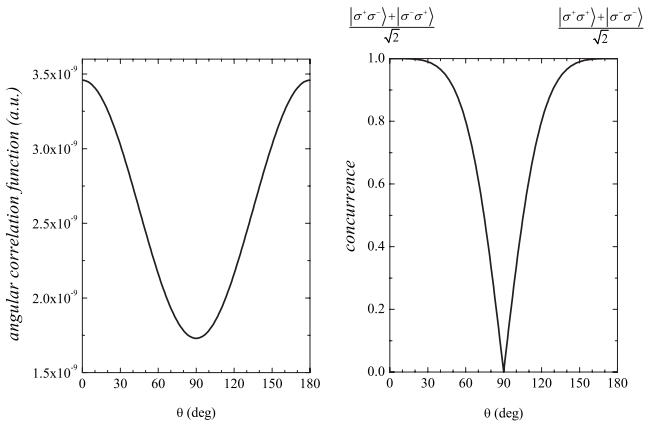

FIG. 1. Two-photon angular correlation function (left) and polarization entanglement in the helicity basis (right) as functions of the opening angle θ between two photons emitted in the $2s_{1/2}\rightarrow 1s_{1/2}$ decay of unpolarized hydrogen. Calculations are presented for the equal energy sharing $x=0.5$ between the photons.

It is, however, important to note that the concurrence values that we calculate in this way are conditioned on the simultaneous detection of the two photons in well-defined directions. This corresponds to a postselected analysis of the entanglement as it is common in parametric down-conversion experiments.

III. RESULTS AND DISCUSSION

A. Nonrelativistic regime

Having available the reduced density matrix for the spins of the two photons, we are now prepared to analyze the angular and entanglement properties of the photon pairs in the $2s_{1/2}\rightarrow 1s_{1/2}$ decay of one electron atoms. For the sake of convenience, let us start from the angular correlation function $W(\boldsymbol{n}_1,\boldsymbol{n}_2)$ which has been investigated in great detail, both in the framework of the nonrelativistic [13] and relativistic theory [14]. For low-Z ions, this correlation function follows a simple (and symmetric) $1+\cos^2\theta$ behavior (cf. left panel of Fig. 1), where θ is the (opening) angle between the two photons, and implies that the photons are preferably emitted either in parallel ($\theta=0°$) or back-to-back ($\theta=180°$) geometry, while the emission under 90° is suppressed by about a factor of 2.

Similar to the angular correlation, the concurrence (8) is symmetric also with respect to $\theta=90°$ and has its minimum at this angle. However, while the (angle-dependent) probability to find a photon pair is reduced just by a factor of 2, the concurrence changes from $\mathcal{C}=1$ for the maximally entangled (Bell) state $|\Psi^+\rangle=(|\sigma^+\sigma^-\rangle+|\sigma^-\sigma^+\rangle)/\sqrt{2}$ at $\theta=0°$ down to zero at $\theta=90°$ and back to a maximum entanglement for the state $|\Phi^+\rangle=(|\sigma^+\sigma^+\rangle+|\sigma^-\sigma^-\rangle)/\sqrt{2}$, if the photons are emitted at $\theta=180°$. For $\theta=0°$ and 180°, this behavior can be understood quite easily (within the helicity basis) due to the conservation of the spin projection along the axis of propagation and has

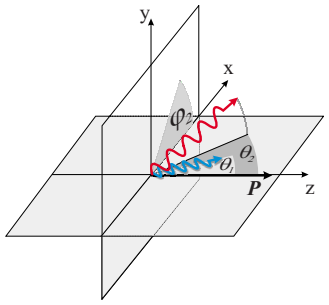

FIG. 2. (Color online) Geometric setup for the two-photon decay of a polarized atom. The polarization axis ***P*** defines the quantization axis. The first photon fixes the reaction plane (x-z plane) and requires only one angle, θ_1. θ_2 and φ_2 describe the direction of the second photon.

been utilized before in studying the violation of Bell's inequalities by means of the two-photon decay of atomic deuterium [15]. For $\theta=90°$, a more elaborate analysis is required in order to explain the vanishing entanglement, although this result might be expected in the nonrelativistic limit from the well-known fact that, in atomic and nuclear physics, no correlation occurs between the polarization states of two photons which are emitted under $\theta=90°$ in a $J=0\rightarrow J=1\rightarrow J=0$ radiative cascade [28].

Figure 1 applies for the case that the atoms in the excited $2s_{1/2}$ state are *unpolarized*. Using modern laser techniques, most (valence-shell excited) atoms can be polarized in a given direction and with a predetermined degree of polarization. Therefore we shall analyze next how the angular distribution and entanglement is affected if we consider the decay of initially polarized atoms. In contrast to the unpolarized case, three angles are required in order to fully describe the photon emission. If we define the reaction plane (x-z plane) to be spanned by the directions of the initial polarization (quantization axis) and the propagation of the first photon [cf. Fig. 2], the (polar) angle θ_1 is sufficient to characterize the first photon, and the two angles θ_2 and φ_2 for the second one.

For an initially polarized atom, Fig. 3 displays the angular correlation and polarization entanglement as a function of the (polar) angle θ_2. Calculations have been done for the emission of the first photon under the angles $\theta_1=0°$, 30°, and 60° (with respect to polarization axis of the atoms) and for different angles φ_2. For $\theta_1=0°$ (left column), the results for the angular correlation and concurrence agree with the unpolarized case in Fig. 1 and do not depend on the second angle φ_2 since the (initial) axial symmetry of the overall system must be preserved if the first photon is emitted along the quantization axis. This situation changes for a photon emission under some angle, say $\theta_1=30°$ or $\theta_1=60°$, and leads to a shift in the graph of the concurrence to the right, i.e., towards higher values of the angle θ_2, while the principal shape of the concurrence does not change. However, both the angular correlation and the (maximum degree of) entanglement decrease if θ_1, the angle between the polarization axis

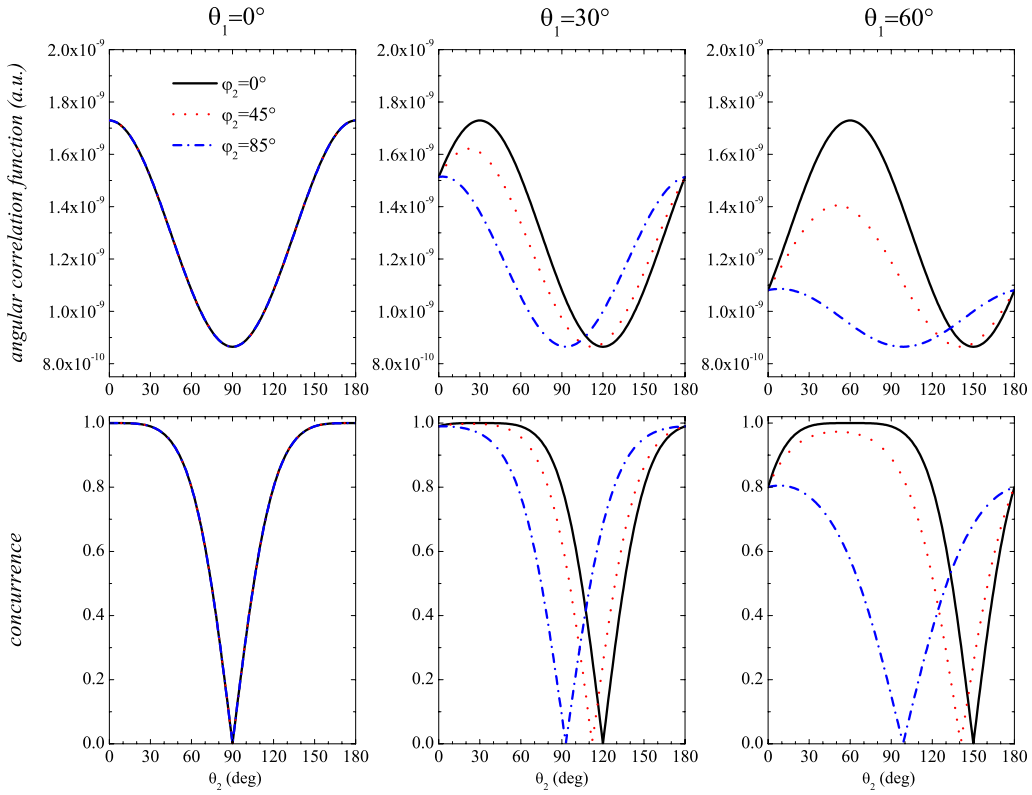

FIG. 3. (Color online) Two-photon angular correlation (upper panel) and polarization entanglement (lower panel) as functions of the angle θ_2 of the second photon for the $2s_{1/2}\rightarrow 1s_{1/2}$ decay of initially polarized hydrogen. Results are shown for different combinations of the angles θ_1 and φ_2.

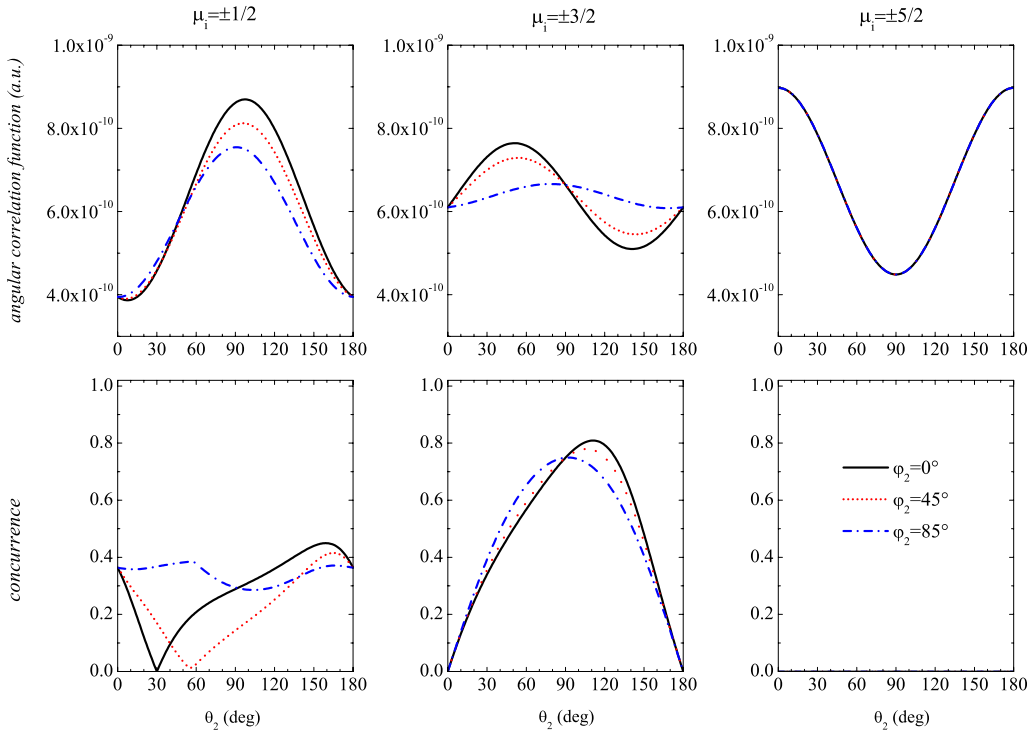

FIG. 4. (Color online) Two-photon angular correlation (upper panel) and polarization entanglement (lower panel) as functions of the emission angle θ_2 of the second photon for the $3d_{5/2} \rightarrow 1s_{1/2}$ decay of polarized hydrogen. Results are shown for different initial populations of the $|3d, \mu_i\rangle$ sublevels, an equal energy sharing $x=0.5$ and for a fixed angle $\theta_1=60°$.

of the atom and the first photon, is enlarged. Most clearly, this is seen below for $\theta_1=60°$ and $\varphi_2=85°$ in the right column of Fig. 3.

For polarized ions in the $2s_{1/2}$ state, all the effects are purely geometric and, in fact, very similar to the case of initially unpolarized atoms: The two photons are completely polarization entangled only if they are emitted in parallel or antiparallel directions, i.e., for $\theta_2=\theta_1$ and $\theta_2=180°-\theta_1$ while the entanglement vanishes for all geometries where the photons are emitted perpendicularly. Qualitatively, the angular correlation function shows a very similar behavior so that maximum/minimum entanglement always coincides with maximum/minimum intensity. The similarity between the decay of the polarized and unpolarized $2s_{1/2}$ state can be understood in the nonrelativistic limit if the effects of the spin-orbit interaction are neglected. In this simple approach, the $2s_{1/2} \rightarrow 1s_{1/2}$ decay can be viewed as a transition between two *pure* $2s$ ($l=0$) and $1s$ ($l=0$) states, and where the polarization correlation of the photon pair is not affected by the spin of the electron.

Apart from the well-studied $2s_{1/2} \rightarrow 1s_{1/2}$ decay of hydrogen, the $3d_{5/2} \rightarrow 1s_{1/2}$ two-photon transition has also been observed previously [30]. Compared to the dominant $3d_{5/2} \rightarrow 2p_{3/2} \rightarrow 1s_{1/2}$ cascade decay of two subsequently emitted photons, the (simultaneous) two-photon transition is then suppressed by about six orders of magnitude [10]. Nevertheless, this weak two-photon decay might be utilized for an equal energy sharing between the two photons. For this decay, Fig. 4 displays the angular correlation and polarization entanglement, assuming different initial populations of the $3d_{5/2}$ (sub-) levels. For initially unpolarized atoms, no entanglement is found, independent of the opening angle between the two photons. As was argued in Ref. [31] for general two-qubit systems, classical correlations compete with quantum mechanical correlations and may rule out entanglement, if the mixedness of the state becomes sufficiently strong. For unpolarized hydrogen in the $3d_{5/2}$ level, indeed, the complete mixture in the excited state gives rise to a highly mixed two-photon state even in the nonrelativistic framework.

In addition, there is also no photon-photon entanglement for the decay of the $\mu_i=\pm 5/2$ sublevels as seen from the right column in Fig. 4. For these two sublevels, the $|\Delta\mu|=2$ (two-photon) transition always implies a pure *product* state of the photon pair, independent of their geometry. However, a remarkable angular dependence of the photon-photon entanglement is found for the decay of the initial $\mu_i=\pm 3/2$ and $\pm 1/2$ substates. In these cases, the classical correlations in the mixed final state (of the two photons) tend to decrease the maximum attainable entanglement but lead to a *zero* concurrence only at selected angles. In Fig. 4, the concurrence is displayed for the decay of the $\mu_i=\pm 1/2$ substates as a function of θ_2, and for a fixed polar angle $\theta_1=60°$ of the "first" photon. When compared with the $\mu_i=\pm 3/2$ states, a notably lower degree of entanglement is found for the $\mu_i=\pm 1/2$ substates, a behavior which is consistent again with the typically larger degree of "mixedness" in the $\mu_i=1/2$ case if, for instance, measured in terms of the linear entropy $S_L(\rho)=\frac{4}{3}[1-\mathrm{Tr}(\rho^2)]$ which takes values in the range 0 to 1.

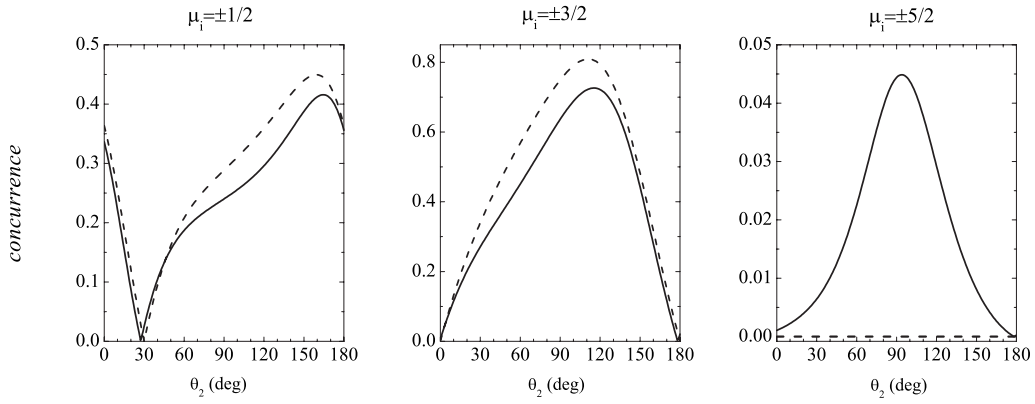

FIG. 5. Polarization entanglement in the $3d_{5/2} \rightarrow 1s_{1/2}$ decay of hydrogen (dashed line) and hydrogenlike uranium (solid line). Results are shown for different initial populations of the $|3d, \mu_i\rangle$ sublevels, an equal energy sharing $x=0.5$, and for fixed angles $\theta_1=60°$ and $\varphi_2=0°$.

B. Relativistic regime

So far, our discussion has been focused on the (nonrelativistic) case of hydrogen where the simplified density matrix (7) provides a very good description of the final two-photon system (when the initial atom is unpolarized). However, the theory in Sec. II A includes also relativistic and multipole effects so that it is suitable for the study of heavier systems. In a previous study on the spin entanglement between the photoion and the emitted electron during the photoionization of hydrogenlike systems [32], it was found that these relativistic effects can have a significant influence on the final-state entanglement.

Hence in order to obtain more insight also into the relativistic and multipole effects on the polarization entanglement in the two-photon decay, we have done exemplary computations for the $3d_{5/2} \rightarrow 1s_{1/2}$ decay of hydrogenlike uranium with different initial populations of the sublevels $|3d, \mu_i\rangle$. For comparison, the results for the concurrence measure are shown in Fig. 5 together with the ones obtained for hydrogen. From the curves for $\mu_i = \pm 1/2$ and $\pm 3/2$ (left and center) one can see that the results for uranium (solid lines) show a qualitatively similar behavior compared to the hydrogen curves (dashed lines). In particular, the positions of the zeros and maximum values agree within only a few degrees. On the other hand, one can see that for uranium the attained concurrence is notably lower than for hydrogen. For example, for $\mu_i = \pm 1/2$ and $\theta_2 = 70° - 160°$ the relativistic and multipole effects in uranium lead to a decrease of the concurrence by approximately 15% compared to hydrogen; and similarly, for $\mu_i = \pm 3/2$ and $\theta_2 = 40° - 120°$ the concurrence values for uranium are up to 20% lower than for hydrogen. In contrast to the $\mu_i = \pm 1/2, \pm 3/2$ cases, the situation is qualitatively different for $\mu_i = \pm 5/2$ (Fig. 5, right). While for hydrogen there is no polarization entanglement at all, for uranium we find a rather sharply peaked, slightly asymmetric curve with a maximum concurrence value of $\mathcal{C} = 0.045$ at $\theta_2 = 90°$. These examples clearly demonstrate that relativistic and multipole effects can influence the polarization entanglement in a nontrivial way.

IV. SUMMARY

In summary, we have investigated the entanglement between the spin states of the two photons emitted in the $2s_{1/2} \rightarrow 1s_{1/2}$ and $3d_{5/2} \rightarrow 1s_{1/2}$ decay of hydrogenlike systems. From the analysis above, based on Dirac's equation and second-order perturbation theory, a strong variation in the (degree of) entanglement is found, as a function of the geometry of the photon detection as well as the initial population of the ionic sublevels in the excited state (atomic polarization). While the results for the nonrelativistic dipole approximation reflect only the dependence on the geometry there can be also a significant change of the polarization entanglement due to the relativistic and multipole effects as we have shown for the $3d_{5/2} \rightarrow 1s_{1/2}$ decay of hydrogenlike uranium. It can be expected that such nonrelativistic effects will play a role also for the violation of the Bell inequality. Based on the general theory presented in the current work, this question will be investigated elsewhere.

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