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Relativistic calculation of the two-photon decay rate of highly excited ionic states

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Based on quantum electrodynamics, we reexamine the two-photon decay of one-electron atoms. Special attention is paid to the calculation of the (two-photon) total decay rates which can be viewed as the imaginary part of the two-loop self-energy. We argue that our approach can be easily applied to the cases with a virtual state having an intermediate energy between the initial and the final state of the decay process leading, thus, to the resonance peaks in the two-photon energy distribution. In order to illustrate our approach, we obtain fully relativistic results, resolved into electric and magnetic multipole components, for the two-photon decay rates of the $3S_{1/2} \to 1S_{1/2}$ transition in neutral hydrogen as well as in various hydrogenlike ions.

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I. INTRODUCTION

Since the seminal work of Göppert-Mayer [1], two-photon decay rates of excited states in hydrogenlike atoms and ions have been the subject of intense experimental [2–5] and theoretical [6–13] studies. For many years, the investigations have dealt not only with the total decay rates but also with the energy and even angular distributions of the two emitted photons. By analyzing these (two-photon) properties, unique information has been obtained about the structural properties of one-electron systems including subtle relativistic effects as well as about the basic concepts of quantum physics such as, e.g., the entanglement.

Even though large experimental and theoretical efforts have been undertaken in the past to understand various aspects of the two-photon decay of hydrogenlike atoms, the analysis of this process still raises a number of unresolved problems. One of these problems, which currently attracts a lot of interest, concerns those two-photon transition from highly excited states to the ground state which pass through an intermediate state with a lower energy than the initial state of the two-photon process [9,10] and can alternatively decay to the ground-state via two (or more) sequential one-photon emissions. Such a transition leads to resonance peaks in the energy spectrum of the coherently emitted photons from the two-photon decays which are located at the energies corresponding to the (real) intermediate states. One of the most pronounced examples of such a situation is the $3S_{1/2} \to 1S_{1/2}$ two-photon $E1E1$ transition for which the differential (in energy) emission rate has singularities at energies corresponding to the $3S_{1/2} \to 2P_{1/2} \to 1S_{1/2}$ and $3S_{1/2} \to 2P_{3/2} \to 1S_{1/2}$ cascades. A proper treatment of these singularities is obviously required for computing total decay rates obtained after an integration over the energies of the coherently emitted photons in the direct two-photon decay $3S_{1/2} \to 1S_{1/2}$.

During the last two decades, the theoretical treatment of the resonances in the energy distribution of the emitted photons has been discussed in a number of places. In general, the decay rate Γ_i of an initial state $|i\rangle$ in a hydrogenlike atom is the sum of a one-photon decay rate $\Gamma_i^{(1)}$ and a two-photon contribution $\Gamma_i^{(2)}$,

$$\Gamma_i = \Gamma_i^{(1)} + \Gamma_i^{(2)}. \tag{1}$$

The expression for $\Gamma_i^{(2)}$ as originally derived in [1] is easily seen to involve an integral over the energies of the emitted photons, the sum of which has to be equal to the energy difference of the initial and final states, and a summation over all possible intermediate, virtual states. In order to avoid problems with nonintegrable singularities, the authors of [9] have attributed $\Gamma_i^{(2)}$ only to the so-called nonresonant intermediate transitions, in contrast to a summation over the complete intermediate-state spectrum. The nonresonant transitions are given by intermediate states of energy higher than the energy E_i of the initial state [9] (the "resonant" intermediate states, which are involved in the one-photon cascade processes, are explicitly excluded from the sum over intermediate states). Based on this assumption, the nonresonant contribution for the $3S_{1/2} \to 1S_{1/2}$ two-photon transition was calculated as $\Gamma_{3S}^{(2)} = 8.2196\ \mathrm{s}^{-1}$ for hydrogen. Later, this result has been also confirmed in the calculations by Florescu and co-workers [10] who used a different method for the summation over the "nonresonant" states.

Although the results presented in Refs. [9,10] are in mutual agreement, they are both based on the interpretation of the two-photon decay rate $\Gamma_i^{(2)}$ as a rate generated only by nonresonant intermediate levels. In our paper, we would like to propose an alternative way for the computation of the (two-photon) total decay rates which leads to a natural removal of the infinities otherwise introduced into the expression for the two-photon decay rate. We apply here a fully relativistic, quantum electrodynamical approach to reinvestigate the two-photon decay of highly excited states of hydrogenlike atoms, paying special attention to a careful handling of the resonances infinitesimally displaced from Feynman's photon integration contour (these singularities exactly correspond to the problematic "resonant" intermediate states). By making use of this approach, we obtain finite, physically

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sensible results for the decay rates of the two-photon $3S_{1/2} \to 1S_{1/2}$ transitions in neutral hydrogen as well as in various hydrogenlike ions. Apart of the leading, electric dipole ($E1E1$) transition, we also discuss the contributions from the higher multipole components to the total decay rate.

This paper is organized as follows: after a brief survey of the theoretical expressions used in our analysis (Sec. II), we proceed by discussing the method of evaluation (Sec. III) as well as the numerical results obtained for the differential as well as the total two-photon decay rates (Sec. IV). Conclusions are given in Sec. V.

II. THEORY

Within quantum electrodynamics, the (negative) imaginary part of the self-energy is just the $\Gamma/2$, where Γ is the decay width [14–16]. The one-photon decay $\Gamma^{(1)}$ rate is obtained from the imaginary part of the one-loop self-energy, while the two-loop self-energy gives rise to the two-photon decay rate $\Gamma^{(2)}$. Because the relativistic formulation of the two-loop self-energy problem has been discussed before in a number of places [17,18], we only mention here that by following a straightforward generalization of the standard procedure described for the nonrelativistic framework in Refs. [12,13,16], we obtain the following expression for the two-photon decay rate ($\hbar = c = \epsilon_0 = 1$):

$$\Gamma_i^{(2)} = \frac{\alpha^2}{\pi} \lim_{\epsilon \to 0} \mathrm{Re} \int_0^{\omega_{\max}} d\omega_1 \omega_1 \omega_2 \int d\Omega_1 d\Omega_2 S_{if}(\omega_1, \omega_2), \tag{2}$$

where $\omega_1 + \omega_2 = \omega_{\max} = E_i - E_f$ with the initial and final state energies E_i and E_f, respectively. S_{if} is given by

$$\begin{aligned} S_{if}(\omega_1, \omega_2) = \sum_\nu \Bigg(& \frac{\langle \psi_f | \mathbf{A}_1^* | \psi_\nu \rangle \langle \psi_\nu | \mathbf{A}_2^* | \psi_i \rangle}{E_i - E_\nu - \omega_2 + i\epsilon} \\ & + \frac{\langle \psi_f | \mathbf{A}_2^* | \psi_\nu \rangle \langle \psi_\nu | \mathbf{A}_1^* | \psi_i \rangle}{E_i - E_\nu - \omega_1 + i\epsilon} \Bigg) \\ & \times \sum_\rho \Bigg(\frac{\langle \psi_i | \mathbf{A}_1 | \psi_\rho \rangle \langle \psi_\rho | \mathbf{A}_2 | \psi_f \rangle}{E_i - E_\rho - \omega_1 + i\epsilon} \\ & + \frac{\langle \psi_i | \mathbf{A}_2 | \psi_\rho \rangle \langle \psi_\rho | \mathbf{A}_1 | \psi_f \rangle}{E_f - E_\rho - \omega_2 + i\epsilon} \Bigg), \end{aligned} \tag{3}$$

where in the second factor, the initial and the final state are exchanged, but the infinitesimal imaginary part in the denominators remains $+i\epsilon$ (i.e., does not change sign). We here manifestly assume that $\psi_i(\boldsymbol{r}) \equiv \psi_{n_i j_i \mu_i}(\boldsymbol{r})$ 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 in the standard representation, describing an electron bound to a pointlike nucleus with charge number Z. For photons propagating with wave vector $\boldsymbol{k}_i$ ($i = 1, 2$) and unit polarization vector $\mathbf{u}_{\lambda_i}$ ($\boldsymbol{k}_i \cdot \boldsymbol{u}_{\lambda_i} = 0$), moreover, the electron-photon interaction operator $\mathbf{A}_i$ in the transition amplitude (3) can be written in velocity gauge as

$$\mathbf{A}_i = A_0 \boldsymbol{\alpha} \cdot \boldsymbol{u}_{\lambda_i} e^{i \boldsymbol{k}_i \cdot \boldsymbol{r}}, \tag{4}$$

where A_0 is a normalization factor, $\boldsymbol{\alpha}$ are the standard Dirac matrices, and $\lambda_i = \pm 1$ denotes the *helicity*, i.e., the spin projection of the photon onto the direction of propagation $\boldsymbol{k}_i$. It is important to note that even though the electron-photon interaction operator (4) depends, of course, on the direction of the photon emission, one has to integrate over these directions in Eq. (2) in order to get the total decay rate.

III. EVALUATION

The summation over the intermediate states in the amplitude (3) runs over the *complete* one-particle spectrum $|\psi_\nu\rangle \equiv |\psi_{n_\nu j_\nu \mu_\nu}\rangle$, including a summation over the discrete part of the spectrum as well as an integration over the positive- and negative-energy continuum of the Dirac spectrum. One has to use the full Dirac-Coulomb Green function—which is not known in closed analytic form—in order to perform this calculation consistently. In recent years, the Green's function method [19] has been widely applied for the analysis of the total two-photon decay rates as well as the photon-photon angular correlation functions [20]. Various possibilities for the numerical implementation of the relativistic Green's function are known, among which we would like to mention (i) a well-known formulation in terms of Whittaker functions [21] and (ii) a Sturmian decomposition in terms of Laguerre polynomials as suggested by Hylton and Snyderman [22].

We use the latter representation and apply the techniques of Racah's algebra to all spherical tensors and to the standard radial-angular representation of the wave functions, and to the Dirac-Coulomb Green function. For the interaction of electrons with the radiation field, the spherical tensor components are obtained from the known standard multipole expansion of the photon operator (see, e.g., Eq. 5 of Ref. [23]),

$$\boldsymbol{u}_\lambda e^{i \boldsymbol{k} \cdot \boldsymbol{r}} = \sqrt{2\pi} \sum_{LMp} i^L (i\lambda)^p \sqrt{2L+1} \boldsymbol{A}_{LM}^{(p)} D_{M\lambda}^L(\boldsymbol{n}), \tag{5}$$

where $\boldsymbol{A}_{LM}^{(p)}$ denotes the electric ($p = 1$) and magnetic ($p = 0$) multipole fields, respectively.

IV. RESULTS

The great advantage of the multipole decomposition (5) is that it allows us to study the contributions to the total (two-photon) decay rate from the various *allowed* multipole combinations. We use the integrand in the integral over ω_1 in Eq. (2) as a measure of the differential decay rate (where we can set explicitly $\epsilon = 0$ for the differential rate). The energy distributions of the two photons emitted in the $3S_{1/2} \to 1S_{1/2}$ decay of neutral hydrogen and hydrogenlike ions are calculated as a function of the energy sharing parameter $x = \omega_1/(\omega_1 + \omega_2)$. For energy sharing in the range $0 < x < 1$, the contributions to the energy distribution from the $E1E1$ and $E1M2$ multipole combinations are displayed in Figs. 1 and 2, respectively. As seen from these figures, the photon energy distributions for both multipole combinations exhibit sharp resonance peaks. As already mentioned, this behavior is due to the fact that the summation in Eq. (3) includes also intermediate states $|\nu\rangle$ having an energy E_ν with $E_i > E_\nu > E_f$. However, the intermediate states contributing to the peaks are not only defined by the (symmetry of)

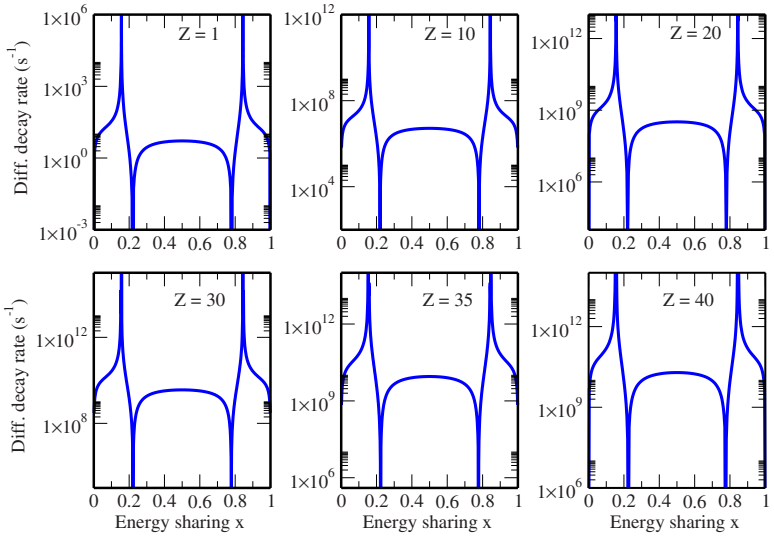

FIG. 1. (Color online) Differential decay rate $d\Gamma/dx$ for the dominant $E1E1$ component of the $3S \rightarrow 1S$ two-photon transitions in neutral hydrogen as well as in hydrogenlike ions with $Z=1$, 10, 20, 30, 35, and 40, where x is the energy sharing $x=\omega_1/(\omega_1+\omega_2)$. Relativistic wave functions are used for the initial, intermediate, and final states, but the electron-photon interaction has been restricted to electric dipole term ($E1E1$ term). The resonance peaks in the decay rate correspond to the resonant $3S_{1/2} \rightarrow 2P_{1/2} \rightarrow 1S_{1/2}$ and $3S_{1/2} \rightarrow 2P_{3/2} \rightarrow 1S_{1/2}$ decay processes.

the initial $|i\rangle$ and final $|f\rangle$ states but also by the multipole components of the radiation field involved in the two-photon process and are different for $E1E1$ as opposed to $E1M2$, and in addition, marked differences exist between the low-Z and the high-Z region. For example, the fine structure of the resonance in the $E1E1$ energy spectrum grows with Z (the contributing states are $2P_{1/2}$ and $2P_{3/2}$). By contrast, no splitting is observed—even for very heavy ions—for the $E1M2$ component of the $3S_{1/2} \rightarrow 1S_{1/2}$ decay. Only one intermediate state, namely $2P_{3/2}$, is allowed for $E1M2$.

Our treatment of the intermediate state resonance peaks in the integration over the photon energy is dictated by an accurate analysis of Eqs. (2) and (3) as obtained from the relativistic two-loop self-energy. The general structure of the contribution to $S_{if}(\omega_1, \omega_2)$ generated by virtual states with intermediate energies $E_i > E_\nu > E_f$ with resonance energy $\omega_r = E_i - E_\nu$ or $\omega_r = E_\nu - E_f$ is as follows:

$$S_{if}(\omega_1, \omega_2) \sim \frac{R_1}{\omega_r - \omega + i\epsilon} + \frac{R_2}{(\omega_r - \omega + i\epsilon)^2}. \tag{6}$$

The integration of the first term can be carried out using the Dirac prescription (see, for example, Ref. [12]). The second term of Eq. (6) can be treated using the formula

$$\lim_{\epsilon \rightarrow 0} \mathrm{Re} \int_0^1 d\omega \left(\frac{1}{\omega_r - \omega + i\epsilon} \right)^2 = \frac{1}{\omega_r(\omega_r - 1)}, \tag{7}$$

where we used an appropriate scaling of the photon energy integration variable in order to map the integration region to the interval (0, 1). It is important to note that Eq. (7) holds strictly for $0 < \omega_r < 1$, but the limit is not approached uniformly [12,13]; i.e., it would be forbidden to exchange the sequence of the limit $\epsilon \rightarrow 0$ with the integration over ω. As usual in quantum electrodynamic processes, all regulators have to be kept up until the very end of the calculation.

With these preparations, it is easy now to integrate over the photon energies (see Table I for the $3S_{1/2} \rightarrow 1S_{1/2}$ process). As seen from the table, the cross sections for the $E1E1$, $E1M2$, and $M1M1$ components scale with the nuclear charge as Z^6, Z^{10}, and Z^{10}, respectively. As expected, this scaling behavior is similar to the Z-scaling of the multipole components in the $2S_{1/2} \rightarrow 1S_{1/2}$ transition.

Furthermore, as seen from Table I and as implied by the nonuniform convergence of the integrals, the intermediate

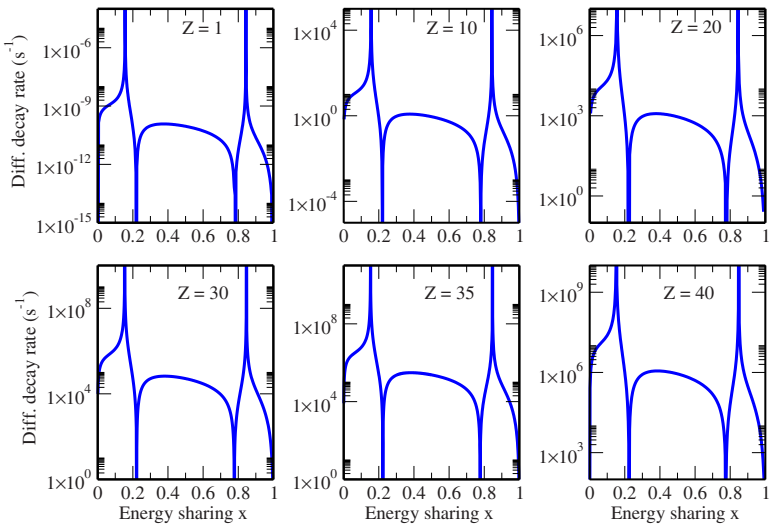

FIG. 2. (Color online) Differential decay rate $d\Gamma/dx$ for the $E1M2$ component of the $3S \rightarrow 1S$ two-photon transitions in neutral hydrogen as well as in hydrogenlike ions with $Z=1$, 10, 20, 30, 35, and 40 (x is the fraction of energy carried by the $E1$ photon). The $E1M2$ term is treated relativistically. The resonances in the decay rate exclusively correspond to the $3S_{1/2} \rightarrow 2P_{3/2} \rightarrow 1S_{1/2}$ cascade, the $2P_{1/2}$ state does not contribute.

TABLE I. Contributions from different combinations of multipoles to the integrated decay rate $\Gamma^{(2)}$, in units of radians per second. Relativistic calculations have been performed for different hydrogenlike ions.

Z	$E1E1$ (Z^{-6})	$E1M2$ $(Z^{-10}10^{10})$	$M1M1$ $(Z^{-10}10^{12})$
1	2.08	1.19	6.13
5	2.03	1.18	6.13
10	1.98	1.16	6.14
15	1.94	1.14	6.16
20	1.90	1.12	6.20
25	1.84	1.08	6.24
30	1.79	1.03	6.30
35	1.67	0.96	6.39
40	1.60	0.86	6.50

states with the energies E_ν lying between the energies of the initial and the final states give a *finite* contribution to the two-photon decay rate. For the electric dipole ($E1E1$) transition in a neutral hydrogen atom, e.g., a proper treatment of the intermediate $2P_{1/2}$ and $2P_{3/2}$ states leads to the decay rate of $\Gamma^{(2)}_{3S}=2.08\ \mathrm{s}^{-1}$ which is in agreement with the result of nonrelativistic calculations reported in Ref. [12]. However, when comparing our prediction with the theoretical data by Cresser and co-workers [9] a large discrepancy by about a factor of 4 is observed. The occurrence of the discrepancy is natural because the problematic virtual states with intermediate energies are treated differently in [9].

V. CONCLUSIONS

The two-photon decay of hydrogenlike ions has been reinvestigated within the framework of relativistic quantum electrodynamics. Starting from first principles of this theory, we treat the total (two-photon) decay rate as the imaginary part of the relativistic two-loop self-energy. The great advantage of this approach, which has its roots in field theory, is that it provides a simple and efficient route to handle the potentially problematic cases of those two-photon transitions from an excited into the ground state which pass intermediate states that can otherwise also be reached in one-photon cascades from the initial to the final states. We found that those states with energies that lie between the energy of the initial and the final states, contribute a finite correction to the total two-photon decay rate. Taking into account this correction, we calculate the rates for the $3S_{1/2}\to 1S_{1/2}$ two-photon decay of neutral hydrogen as well as hydrogenlike ions. Our results are in a good agreement with nonrelativistic calculations for low Z (see Refs. [12,13]) but show a significant deviation from the data by Cresser and co-workers [9].

Our quantum electrodynamics approach, as discussed in the present paper, opens a way for a *systematic* theoretical analysis of the simultaneous, coherent two-photon emission from one-electron (and many-electron) atomic systems, even in cases where problematic intermediate states with an energy between the initial and final states give rise to resonance peaks in the photon energy distributions. We stress here that a conceivable alternative approach to the removal of the formal infinities generated by the intermediate "cascade" states, which is based on the explicit removal of these states from the sums over ν and ρ in Eq. (2), gives rise to a number of conceptual problems, including gauge noninvariance with respect to length and velocity gauges [13]. Our approach is manifestly gauge invariant and also avoids problems connected with the identification of the infinitesimal parts $i\epsilon$ in the propagator denominators in Eq. (2) with partial or total decay rates of the intermediate states: the ϵ parameters are free parameters which approach zero after all other operations, including the integrations over the photon energies, have been performed. This operation leads to a finite result and corresponds, as explained in Ref. [13], to a *partial* removal of the problematic intermediate states from the sum over all virtual states involved in the two-photon process, albeit in a fully gauge-invariant manner.

In addition to its relevance for atomic physics, our approach may have a significant impact for astrophysical studies where a detailed knowledge of the (properties of) two-photon transitions is highly required for the analysis of cosmological hydrogen and helium recombination. The contribution of two-photon processes to the recombination history represents an issue which has recently attracted substantial theoretical interest [24].

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