Negative-continuum effects on the two-photon decay rates of hydrogenlike ions

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Two-photon decay of hydrogenlike ions is studied within the framework of second-order perturbation theory, based on the relativistic Dirac's equation. Special attention is paid to the effects arising from the summation over the negative-energy (intermediate virtual) states that occur in such a framework. In order to investigate the role of these states, detailed calculations have been carried out for the $2s_{1/2} \to 1s_{1/2}$ and $2p_{1/2} \to 1s_{1/2}$ transitions in neutral hydrogen H as well as for hydrogenlike xenon Xe^{53+} and uranium U^{91+} ions. We found that for a correct evaluation of the total and energy-differential decay rates, summation over the negative-energy part of Dirac's spectrum should be properly taken into account both for high-*Z* and low-*Z* atomic systems.

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I. INTRODUCTION

Experimental and theoretical studies on the two-photon transitions in atomic systems have a long tradition. Following seminal works by Göppert-Mayer [1] and by Breit and Teller [2] a large number of investigations have been performed in the past which focused on the decay of metastable states of light neutral atoms and low-*Z* ions. These investigations have dealt not only with the total and energy-differential decay rates [3–5] but also with the angular distributions [6–9] and even polarization correlations between the two emitted photons [10–12]. Detailed analysis of these two-photon properties have revealed unique information about electron densities in astrophysical plasmas and thermal x-ray sources, highly precise values of physical constants [13], structural properties of few-electron systems including subtle quantum electrodynamical (QED) effects [14] as well as about the basic concepts of quantum physics such as, e.g., nonlocality and nonseparability [15].

Beside the decay of metastable states of low-*Z* systems, much of today's interest is focused also on the two-photon transitions in high-*Z* ions and atoms which provide a sensitive tool for improving our understanding of the electron-photon interactions in the presence of extremely strong electromagnetic fields [16]. In such strong fields produced by heavy nuclei, relativistic and retardation effects become of paramount importance and may strongly affect the properties of two-photon emission. To explore these effects, therefore, theoretical investigations based on Dirac's equation have been carried out for the total and energy-differential decay rates [17–21] as well as for the angular and polarization correlations [22–24]. In general, relativistic predictions for the two-photon total and differential properties have been found in good agreement with experimental data obtained for the decay of inner-shell vacancies of heavy neutral atoms [25,26] and excited states of high-*Z* few-electron ions [27].

Although intensive experimental and theoretical efforts have been undertaken recently to understand relativistic effects on the two-photon transitions in heavy ions and atoms, a number of questions still remain open. One of the questions, which currently attracts much of interest, concerns the role of *negative energy* solutions of Dirac's equation in relativistic two-photon calculations. Usually, these calculations are performed within the framework of the second-order perturbation theory and, hence, require summation over the (virtual) intermediate ion states. Such a summation, running over the *complete* spectrum, should obviously include not only positive- (discrete and continuum) but also negative-eigenenergy Dirac states. One might expect, however, that since the energy release in two-photon bound-bound transitions is less than the energy required for the electron-positron pair production, the contribution from the negative part of Dirac's spectrum should be negligible even for the decay of heaviest elements. From practical viewpoint, this assumption justifies the restriction of the intermediate-state summation to the positive-energy solutions only. Exclusion of the negative continuum would lead, in turn, to a significant simplification of the second-order relativistic calculations especially for *many-electron* systems. Detailed theoretical investigations of the two-photon transitions in such systems are performed nowadays not only in heavy ion physics but also in chemical physics where the (two-photon) absorption rates for atoms and molecules are evaluated within the framework of relativistic four-component Hartree-Fock approximation [28–30].

Despite the (relatively) small energy of two-photon transitions, the influence of Dirac's negative continuum in second-order calculations should be further questioned because of possibility for production and subsequent annihilation of the *virtual* antiparticles. It has been argued, for example, that transitions involving positron states have to be taken into account for the proper description of Thomson scattering [31], interaction of ions with intense electromagnetic pulses [32,33] in the "undercritical" regime as well as magnetic transitions in two-electron ions [34–36]. Moreover, the first step toward the analysis of negative-energy contributions to the two-photon properties has been done by Labzowsky and co-workers [37] who focused on E1M1 and

E1E2 $2p_{1/2} \to 1s_{1/2}$ total decay probabilities. The relativistic calculations have indicated the importance of negative-energy contributions in hydrogenlike ions not only for high-Z but also for low-Z domain.

In this work, we apply the second-order perturbation theory based on relativistic Dirac's equation in order to reanalyze atomic two-photon decay. We pay special attention to the influence of negative continuum solutions on the evaluation of the transition amplitudes and, hence, on the total and energy-differential decay rates. For the sake of clarity, we restrict our analysis to the decay of hydrogenlike ions for which both the positive- and negative-energy parts of Dirac's spectrum can be still studied in a systematic way by making use of a finite basis set method [19]. Implementation of this method for computing relativistic second-order transition amplitudes is briefly discussed in Secs. II A and II B. Later, in Sec. II C, we consider an alternative, semiclassical, approach which allows *analytical* evaluation of the negative-energy contributions to the two-photon matrix elements and transition rates. These two—semiclassical and fully relativistic—approaches are used in Sec. III to calculate the energy differential and total decay rates for several multipole terms in the $2s_{1/2} \to 1s_{1/2}$ and $2p_{1/2} \to 1s_{1/2}$ two-photon decay of neutral hydrogen as well as hydrogenlike xenon Xe^{53+} and uranium U^{91+} ions. Based on the results of our calculations, we show that both the total transition probabilities and the photon energy distributions can be strongly affected by the negative-state contributions; this effect is most clearly observed for the nondipole transitions not only in high Z but also in (nonrelativistic) low Z domain. A brief summary of these findings and outlooks are given finally in Sec. IV.

II. THEORY

A. Differential and total decay rates

Not much has to be written about the basic formalism for studying the two-photon transitions in hydrogenlike ions. In the past, this formalism has been widely applied in order to investigate not only the total decay probabilities [17–19,37] but also the energy as well as angular distributions [23] and even the correlation in the polarization state of the photons [15,24]. Below, therefore, we restrict ourselves to a rather brief account of the basic expressions, just enough for discussing the role of negative-energy solutions of Dirac's equation in computing of the two-photon (total and differential) rates.

The properties of the two-photon atomic transitions are evaluated, usually, within the framework of the second-order perturbation theory. When based on Dirac's equation, this theory gives the following expression for the differential in energy decay rate,

$$\frac{dw}{d\omega_1} = \frac{\omega_1\omega_2}{(2\pi)^3 c^2} \left| \sum_\nu \left(\frac{\langle f|\boldsymbol{A}_2^*|\nu\rangle\langle\nu|\boldsymbol{A}_1^*|i\rangle}{E_\nu - E_i + \omega_1} + \frac{\langle f|\boldsymbol{A}_1^*|\nu\rangle\langle\nu|\boldsymbol{A}_2^*|i\rangle}{E_\nu - E_i + \omega_2} \right) \right|^2 d\Omega_1 d\Omega_2, \qquad (1)$$

where the transition operators $\boldsymbol{A}_j^*$ with $j=1,2$ describe the (relativistic) electron-photon interaction. For the emission of photons with wave vectors $\boldsymbol{k}_j$ and polarization vectors $\hat{\boldsymbol{e}}_j$ these operators read as

$$\boldsymbol{A}_j^* = \boldsymbol{\alpha}\cdot(\hat{\boldsymbol{e}}_j + G\hat{\boldsymbol{k}}_j)e^{-i\boldsymbol{k}_j\cdot\boldsymbol{r}} - Ge^{-i\boldsymbol{k}_j\cdot\boldsymbol{r}}, \qquad (2)$$

where $\boldsymbol{\alpha}$ is a vector of Dirac matrices and G is an arbitrary gauge parameter. In the calculations below, following Grant [38], we employ two different gauges that are known to lead to well-known nonrelativistic operators. First, we use the so-called Coulomb gauge, when $G=0$, which corresponds to the velocity form of electron-photon interaction operator in the nonrelativistic limit. As the second choice we adopt $G=\sqrt{(L+1)/L}$ in order to obtain Babushkin gauge which reduces, for the particular case of $L=1$, to the dipole length form of the transition operator.

In Eq. (1), $|i\rangle \equiv |n_i\kappa_i\mu_i\rangle$ and $|f\rangle \equiv |n_f\kappa_f\mu_f\rangle$ denote solutions of the Dirac's equation for the initial and final ionic states respectively, while $E_i \equiv E_{n_i\kappa_i}$ and $E_f \equiv E_{n_f\kappa_f}$ are the corresponding one-particle energies. Because of energy conservation, E_i and E_f are related to the energies $\omega_{1,2}=ck_{1,2}$ of the emitted photons by

$$E_i - E_f = \omega_1 + \omega_2. \qquad (3)$$

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

As usual in atomic physics, the second-order transition amplitudes in Eq. (1) and, hence, the two-photon transitions rates can be further simplified by applying the techniques of Racah's algebra if all the operators are presented in terms of spherical tensors and if the (standard) radial-angular representation of Dirac's wave functions is employed. For the interaction of the electron with electromagnetic field, the spherical tensor components are obtained from the *multipole* expansion of the operator $\boldsymbol{A}_j^*$ (see Refs. [18,19,42] for further details). By using such an expansion, we are able to rewrite Eq. (1) as a sum of partial multipole rates

$$\frac{dw}{d\omega_1} = \sum_{\Theta_1 L_1 \Theta_2 L_2} \frac{dW_{\Theta_1 L_1 \Theta_2 L_2}}{d\omega_1}, \qquad (4)$$

which describe the emission of two photons of electric ($\Theta_j=$E) and/or magnetic ($\Theta_j=$M) type carrying away the angular momenta L_1 and L_2. For the decay of unpolarized ionic state $|n_i\kappa_i\rangle$, in which the emission angles as well as polarization of both photons remain unobserved, these partial multipole rates are given by [18]

$$\frac{dW_{\Theta_1 L_1 \Theta_2 L_2}}{d\omega_1} = \frac{\omega_1\omega_2}{(2\pi)^3 c^2} \sum_{\lambda_{\Theta_1}\lambda_{\Theta_2}} \sum_{j_\nu} \Big[|S^{j_\nu}_{\lambda_{\Theta_1}\lambda_{\Theta_2}}(1,2)|^2 + |S^{j_\nu}_{\lambda_{\Theta_2}\lambda_{\Theta_1}}(2,1)|^2 + 2\sum_{j'_\nu} d(j_\nu,j'_\nu) S^{j_\nu}_{\lambda_{\Theta_2}\lambda_{\Theta_1}}(2,1) S^{j'_\nu}_{\lambda_{\Theta_1}\lambda_{\Theta_2}}(1,2) \Big], \qquad (5)$$

where j_ν is the total angular momentum of electron and summation over λ_{Θ_j} in Eq. (5) is restricted to $\lambda_{\Theta_j}=\pm 1$ for the electric (Θ_j=E) and $\lambda_{\Theta_j}=0$ for the magnetic (Θ_j=M) photon transitions. In Eq. (5), moreover, the angular coefficient $d(j_\nu,j'_\nu)$ is defined by the phase factor and $6j$ Wigner symbol,

$$d(j_\nu,j'_\nu) = \sqrt{(2j_\nu+1)(2j'_\nu+1)}(-1)^{2j'_\nu+L_1+L_2}\begin{Bmatrix} j_f & j'_\nu & L_1 \\ j_i & j_\nu & L_2 \end{Bmatrix}, \tag{6}$$

and the radial integral part is expressed in terms of the reduced matrix elements of the multipole (electric and magnetic) field operators,

$$S^{j_\nu}_{\lambda_{\Theta_1}\lambda_{\Theta_2}}(1,2) = \sum_{n_\nu} \frac{\langle n_f\kappa_f\|\hat{a}^{\lambda_{\Theta_1}*}_{L_1}\|n_\nu\kappa_\nu\rangle\langle n_\nu\kappa_\nu\|\hat{a}^{\lambda_{\Theta_2}*}_{L_2}\|n_i\kappa_i\rangle}{E_\nu - E_i + \omega_2}. \tag{7}$$

Here, summation over n_ν includes not only (infinite number of) bound ionic states but also integration over the positive as well as negative electron continua.

Until now, we have discussed the general expressions for the two-photon transition rates which are differential in energy ω_1 of one of the photons. By performing an integration over this energy one may easily obtain the total rate that is directly related to the lifetime of a particular excited state against the two-photon decay. It follows from Eq. (4) that such a total rate can be represented as a sum of its multipole components,

$$w_{\rm tot} = \sum_{\Theta_1 L_1 \Theta_2 L_2} W_{\Theta_1 L_1 \Theta_2 L_2} \equiv \sum_{\Theta_1 L_1 \Theta_2 L_2} \int_0^{\omega_t} \frac{dW_{\Theta_1 L_1 \Theta_2 L_2}}{d\omega_1} d\omega_1, \tag{8}$$

where $\omega_t = E_i - E_f$ is the transition energy.

As seen from Eqs. (4)–(8), any analysis of the differential as well as total two-photon decay rates can be traced back to the (reduced) matrix elements that describe the interaction of an electron with the (multipole) radiation field. Since the relativistic form of these matrix elements is applied very frequently in studying various atomic processes, we shall not discuss here their evaluation and just refer the reader for all details to Refs. [18,19,38]. Instead, in the next section we will focus on the summation over the intermediate states $|n_\nu\kappa_\nu\rangle$ which appears in the second-order transition amplitudes [see Eq. (7)].

B. Summation over the intermediate states

As mentioned already above, the summation over the intermediate states in Eq. (7) runs over the complete one-particle spectrum $|n_\nu\kappa_\nu\rangle$, including a summation over the discrete part of the spectrum as well as an integration over the positive and negative-energy continuum. A number of methods have been developed over the last decades in order to evaluate the second-order transition amplitudes consistently. Apart from the Green's function approach [23,39] which—in case of a purely Coulomb potential—allows for the analytical computation of Eq. (7), the *discrete-basis-set* summation is widely used nowadays in two-photon studies [19]. A great advantage of the latter method is that it allows to separate the contributions from the positive- and negative-energy solutions in the intermediate-state summation. Since the effects that arise from the negative-energy spectrum are in the focus of the present study, we apply for the calculations below the finite (discrete) basis solutions constructed from the B-spline sets.

Since the B-spline basis set approach has been discussed in detail elsewhere [19,40,41], here we just briefly recall its main features. In this way, we shall consider the ion (or atom) under consideration to be enclosed in a finite cavity with a radius R large enough to get a good approximation of the wave functions with some suitable set of boundary conditions, which allows for discretization of the continua. Wave functions that describe the quantum states $|\nu\rangle \equiv |n_\nu\kappa_\nu\rangle$ of such a "particle in box" system can be expanded in terms of basis set functions $\phi^i_\nu(r)$ with $i=1,\ldots,2N$ which, in turn, are found as solutions of the Dirac-Fock equation,

$$\begin{bmatrix} \dfrac{V(r)}{c} & \dfrac{d}{dr} - \dfrac{\kappa_\nu}{r} \\ -\left(\dfrac{d}{dr} + \dfrac{\kappa_\nu}{r}\right) & -2c + \dfrac{V(r)}{c} \end{bmatrix} \phi^i_\nu(r) = \frac{\epsilon^i_\nu}{c}\phi^i_\nu(r), \tag{9}$$

where $\epsilon^i_\nu = E^i_\nu - mc^2$ and $V(r)$ is a Coulomb potential of a uniformly charged finite-size nucleus. Due to computational reasons, each of $\phi^i_\nu(r)$ function is expressed as a linear combination of B splines as it was originally proposed in Ref. [40] by Johnson and coworkers.

For each quantum state $|\nu\rangle$ the set of basis functions $\phi^i_\nu(r)$ spans both positive and negative energy solutions. Solutions labeled by $i=1,\ldots,N$ describe the negative continuum with $\epsilon^i_\nu < -2mc^2$ while solutions labeled by $i=N+1,\ldots,2N$ correspond to the first few states of the bound-state spectrum as well as to positive continuum with $\epsilon^i_\nu > 0$. Thus, by selecting the proper subset of basis functions $\phi^i_\nu(r)$ we may explore the role of negative continuum in computing of the properties of two-photon emission from hydrogenlike ions.

C. Semirelativistic approximation

Based on the relativistic theory, the expressions obtained in the previous section allow to study the influence of the Dirac's negative continuum on the properties of two-photon emission from hydrogenlike ions with nuclear charge in the whole range $1 \le Z \le 92$. For the low-Z ions, moreover, it is also useful to estimate the negative-energy contributions within the semirelativistic approach as proposed in the work by Labzowsky and coworkers [37]. To perform such a semirelativistic analysis let us start from Eq. (1) in which we retain the sum only over the negative-energy continuum states. Since the total energy of these states is $E_\nu = -(T_\nu + mc^2)$, the corresponding energy denominator of the second-order transition amplitude can be written as $E_\nu - E_i + \omega_j \approx -2mc^2$ which leads to the following expression for the differential decay rate:

$$\frac{dw^{(-)}}{d\omega_1} = \frac{\omega_1\omega_2}{(2\pi)^3c^2}\frac{1}{4(mc^2)^2}\left|\sum_{\nu\in(-)}(\langle f|\boldsymbol{A}_2^*|\nu\rangle\langle\nu|\boldsymbol{A}_1^*|i\rangle + \langle f|\boldsymbol{A}_1^*|\nu\rangle\langle\nu|\boldsymbol{A}_2^*|i\rangle)\right|^2 d\Omega_1 d\Omega_2. \quad (10)$$

For further simplification of this expression, we shall make use of the multipole expansion of the electron-photon interaction operators (2). For the sake of simplicity, we restrict this semirelativistic analysis to the case of Coulomb gauge ($G=0$) in which operator $\boldsymbol{A}_j^*$ can be written as

$$\boldsymbol{A}_j^* = \boldsymbol{\alpha}\cdot\hat{\boldsymbol{e}}_j[1 - i\boldsymbol{k}\cdot\boldsymbol{r} + 1/2(-i\boldsymbol{k}\cdot\boldsymbol{r})^2 + \cdots], \quad (11)$$

if one expands the photon exponential $\exp(i\boldsymbol{k}\cdot\boldsymbol{r})$ into the Taylor series.

In contrast to the "standard" spherical tensor expansion [18,42], the series (11) usually does not allow one to make a clear distinction between the different multipole components of the electromagnetic field. For instance, while the first term in Eq. (11) describes—within the nonrelativistic limit—electric dipole (E1) transition, the term $(-i\boldsymbol{k}\cdot\boldsymbol{r})$ gives rise both, to magnetic dipole (M1) and electric quadrupole (E2) channels. Such an approximation, however, is well justified for our (semirelativistic) analysis which just aims to estimate the role of negative continuum states in the different (*groups of*) multipole two-photon transitions in light hydrogenlike ions. In particular, by adopting $\boldsymbol{A}_j^* = -\boldsymbol{\alpha}\cdot\hat{\boldsymbol{e}}_j(i\boldsymbol{k}\cdot\boldsymbol{r})$ for both operators in Eq. (10) we may find the contribution from the negative spectrum to the 2M1, 2E2, and E2M1 $2s_{1/2}\to 1s_{1/2}$ transition probabilities,

$$\frac{dw^{(-)}_{\mathrm{M1,E2}}}{d\omega_1} = \frac{\omega_1\omega_2}{(2\pi)^3c^2}\frac{1}{4(mc^2)^2}\left|\sum_{\nu\in(-)}[\langle f|\boldsymbol{\alpha}\cdot\hat{\boldsymbol{e}}_2(\boldsymbol{k}_2\cdot\boldsymbol{r})|\nu\rangle \times\langle\nu|\boldsymbol{\alpha}\cdot\hat{\boldsymbol{e}}_1(\boldsymbol{k}_1\cdot\boldsymbol{r})|i\rangle + \langle f|\boldsymbol{\alpha}\cdot\hat{\boldsymbol{e}}_1(\boldsymbol{k}_1\cdot\boldsymbol{r})|\nu\rangle \times\langle\nu|\boldsymbol{\alpha}\cdot\hat{\boldsymbol{e}}_2(\boldsymbol{k}_2\cdot\boldsymbol{r})|i\rangle]\right|^2 d\Omega_1 d\Omega_2. \quad (12)$$

Here, summation over the intermediate states $|\nu\rangle$ is restricted by the negative-energy solutions of the Dirac equation for the electron in the field of nucleus. In the nonrelativistic limits these states form a *complete* set of solutions of the Schrödinger equation for the particle in a repulsive Coulomb field [43]. By employing a closure relation for such a set we rewrite Eq. (12) in the form,

$$\frac{dw^{(-)}_{\mathrm{M1,E2}}}{d\omega_1} = \frac{\omega_1\omega_2}{(2\pi)^3c^2}\frac{1}{(mc^2)^2} \times|(\hat{\boldsymbol{e}}_1\hat{\boldsymbol{e}}_2)\langle f|(\boldsymbol{k}_1\cdot\boldsymbol{r})(\boldsymbol{k}_2\cdot\boldsymbol{r})|i\rangle|^2 d\Omega_1 d\Omega_2, \quad (13)$$

where $|i\rangle$ and $|f\rangle$ now denote the solutions of the Schrödinger equation for the initial and final ionic states, respectively. For the particular case of $2s_{1/2}\to 1s_{1/2}$ two-photon transition, i.e., when $|i\rangle=|2s\rangle$ and $|f\rangle=|1s\rangle$, this expression finally reads

$$\frac{dw^{(-)}_{\mathrm{M1,E2}}}{d\omega_1} = \frac{2^{22}}{3^{13}}\frac{\alpha^{10}}{5\pi Z^4}\omega_1^3\omega_2^3, \quad (14)$$

if one performs an integration over the photon emission angles as well as a summation over the polarization states (see Ref. [37] for further details).

Equation (14) provides the differential rate for the 2M1, 2E2, and E2M1 two-photon transitions as obtained within the nonrelativistic framework and by restricting the summation over the intermediate spectrum $|\nu\rangle$ to the negative energy states only. Being valid for low-Z ions, this expression may also help us to analyze the negative-energy contribution to the *total* decay rate,

$$w^{(-)}_{\mathrm{M1,E2}} = \int_0^{\omega_t}\frac{dw^{(-)}_{\mathrm{M1,E2}}}{d\omega_1}d\omega_1 = (\alpha Z)^{10}\frac{1}{14\pi 5^2 3^6} = 1.247\times 10^{-6}(\alpha Z)^{10}, \quad (15)$$

where the integration over the photon energy ω_1 is performed.

Apart from the 2M1, 2E2, and E2M1 $2s_{1/2}\to 1s_{1/2}$ two-photon transitions, Eqs. (10) and (11) may also be employed to study other decay channels. For example, the negative energy contributions to the differential as well as total rates for the E1M1 and E1E2 $2p_{1/2}\to 1s_{1/2}$ decay read as

$$\frac{dw^{(-)}_{\mathrm{E1,M1,E2}}}{d\omega_1} = \frac{2^{17}}{3^{12}}\frac{\alpha^8}{\pi Z^2}\omega_1\omega_2(\omega_1^2+\omega_2^2), \quad (16)$$

and

$$w^{(-)}_{\mathrm{E1,M1,E2}} = (\alpha Z)^8\frac{2}{5\pi 3^7} = 5.822\times 10^{-5}(\alpha Z)^8, \quad (17)$$

respectively [37]. Together with Eqs. (14) and (15), we shall later use these nonrelativistic predictions in order to check the validity of our numerical calculations in low-Z domain.

III. RESULTS AND DISCUSSION

Having discussed the theoretical background for the two-photon studies, we are prepared now to analyze the influence of the Dirac's negative continuum on the total as well as energy-differential decay rates. We shall start such an analysis for the $2s_{1/2}\to 1s_{1/2}$ transition, which is well established both in theory [18,19,23] and in experiment. For all hydrogenlike ions this transition is dominated by the 2E1 decay channel while all the higher multipoles contribute by less than 0.5% to the decay probability. The energy-differential decay rate given by Eq. (5) for the emission of two electric dipole photons is displayed in Fig. 1 for the decay of neutral hydrogen (H) as well as hydrogenlike xenon Xe^{53+} and uranium U^{91+} ions. For these ions, relativistic second-order calculations have been done within the Coulomb gauge and by performing intermediate-state summation over the complete Dirac spectrum (solid line) as well as over the positive- (dashed line) and negative-energy (dotted line) solutions only. As seen in this figure, the negative-energy contribution to the energy-differential decay rate is negligible for low-Z ions but becomes rather pronounced as the nuclear charge Z

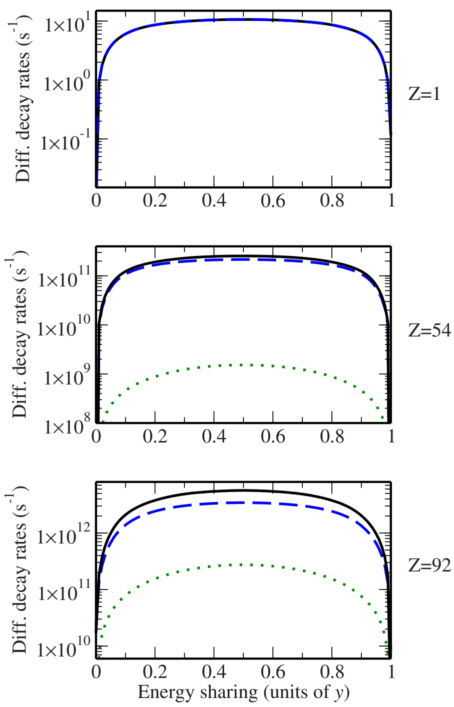

FIG. 1. (Color online) Energy-differential transition rates for the 2E1 $2s_{1/2}\to 1s_{1/2}$ two-photon decay of hydrogen and hydrogenlike ions. Relativistic calculations have been carried out by performing intermediate-state summation over complete Dirac's spectrum (solid line) as well as by restricting this summation to the positive- (dashed line) and negative-energy (dotted line) states only.

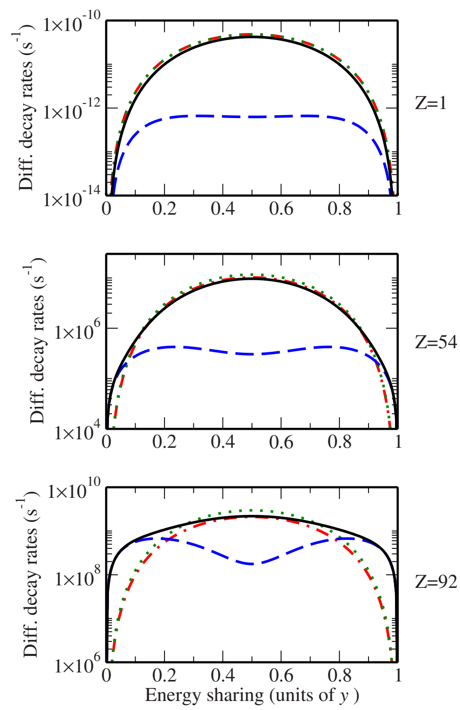

FIG. 2. (Color online) Energy-differential decay rates for the (sum of the) 2M1 and 2E2 $2s_{1/2}\to 1s_{1/2}$ multipole two-photon transitions in hydrogen and hydrogen-like ions. Relativistic calculations have been carried out by performing intermediate-state summation over complete Dirac's spectrum (solid line) as well as by restricting this summation to the positive- (dashed line) and negative-energy (dotted line) states only. Results of relativistic calculations are also compared with the semirelativistic prediction (dot-dashed line) as given by Eq. (14).

is increased. For the 2E1 decay of hydrogen-like uranium, for example, exclusion of the negative solutions from the intermediate-state summation in Eq. (7) leads to about 20% reduction of the decay rate when compared with the "exact" result.

While for the leading, 2E1 $2s_{1/2}\to 1s_{1/2}$ transition the negative continuum effects arise only for rather heavy ions, they might strongly affect properties of the higher multipole decay channels in low-Z domain. In Fig. 2, for example, we display the energy distributions of photons emitted in 2M1 and 2E2 transitions. As seen in the upper panel of the figure corresponding to the decay of neutral hydrogen, negative energy part of the Dirac's spectrum gives the dominant contribution to the (sum of the) differential rates for these decay channels. With the increasing nuclear charge Z, the role of positive energy solutions also becomes more pronounced. However, these solutions allow one to describe reasonably well the differential rates [Eq. (5)] only if one of the photons is much more energetic than the second one, i.e., when either $y<0.1$ or $y>0.9$. For a nearly equal energy sharing ($y \approx 0.5$), in contrast, accurate relativistic calculations of the 2M1 and 2E2 rates obviously require summation over both, the negative and the positive energy states.

Apart from the results of relativistic calculations, we also display in Fig. 2 the (sum of the) negative-energy contributions to the 2M1, 2E2, M1E2, and E2M1 $2s_{1/2}\to 1s_{1/2}$ transition probabilities as obtained within the semirelativistic approach discussed in Sec. II C. As expected, for low-Z ions both the relativistic (dotted line) and semirelativistic (dot-dashed line) results basically coincide and are well described by Eq. (14). As the nuclear charge Z is increased, however, semirelativistic treatment leads to a slight underestimation of the negative-energy contribution to the two-photon (differential) transition probabilities. For the $2s_{1/2}\to 1s_{1/2}$ decay of hydrogenlike uranium ion, for example, results obtained from Eq. (14) is about 30% smaller than the corresponding relativistic predictions.

Up to now, we have been considering the $2s_{1/2}\to 1s_{1/2}$ two-photon decay of the hydrogenlike ions. Apart from this—experimentally well studied—transition, recent theoretical interest has also been focused on the $2p_{1/2}\to 1s_{1/2}$ *two-photon* decay [37]. Although such a channel is rather weak when compared to the leading one-photon E1 transition, its detailed investigation is highly required for future experiments on the parity violation in simple atomic systems [44]. A number of calculations [37,45] have been performed, therefore, for the transition probabilities of the dominant E1M1 and E1E2 multipole components. In order to discuss the role of Dirac's negative continuum in these calculations, we display in Fig. 3 the energy-differential rate for the sum of the E1M1 and E1E2 $2p_{1/2}\to 1s_{1/2}$ two-photon transitions. Again, the calculations have been carried out within the Coulomb gauge for the electron-photon coupling and for three nuclear charges $Z=1$, 54, and 92. As seen in this figure, negative-energy summation in the second-order transition

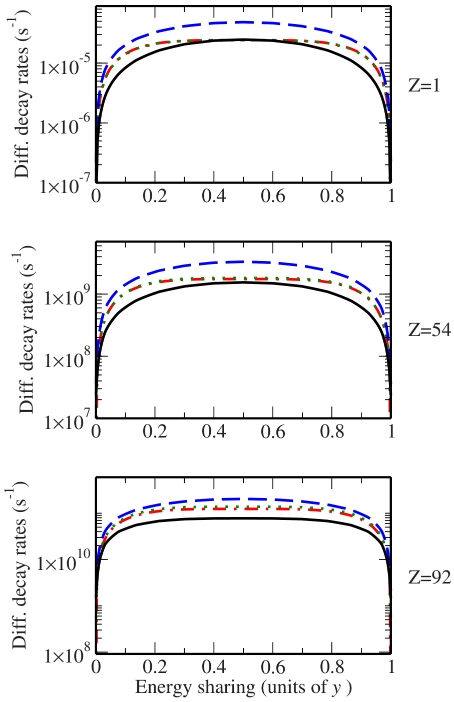

FIG. 3. (Color online) Energy-differential decay rates for the (sum of the) E1M1, M1E1, E1E2, and E2E1 $2p_{1/2}\rightarrow 1s_{1/2}$ multipole two-photon transitions in hydrogen and hydrogenlike ions. Relativistic calculations have been carried out by performing intermediate-state summation over complete Dirac's spectrum (solid line) as well as by restricting this summation to the positive- (dashed line) and negative-energy (dotted line) states only. Results of relativistic calculations are compared also with the semirelativistic prediction (dot-dashed line) as given by Eq. (16).

amplitude (7) is of great importance for accurate evaluation of $2p_{1/2}\rightarrow 1s_{1/2}$ transition probabilities both for low-Z and high-Z ions. That is, restriction of the intermediate-state summation to positive part of Dirac's spectrum results in an overestimation of the E1M1 and E1E2 energy-differential decay rates by factors of about 2 and 2.5 for the neutral hydrogen and hydrogenlike uranium, respectively.

Similar to the $2s_{1/2}\rightarrow 1s_{1/2}$ multipole transitions, we make use of semirelativistic formulas from Sec. II C to cross-check our relativistic computations for the negative-energy contribution to the E1M1 and E1E2 $2p_{1/2}\rightarrow 1s_{1/2}$ decay rates in low-Z domain. Again, while for neutral hydrogen both, semirelativistic (16) and relativistic approaches produce virtually identical results, they start to differ as the nuclear charge Z is increased.

So far we have discussed the energy-differential decay rates both for $2s_{1/2}\rightarrow 1s_{1/2}$ and $2p_{1/2}\rightarrow 1s_{1/2}$ two-photon transitions. Integration of these rates over the energy of one of the photons [see Eq. (8)] yields the *total* decay rates. In Table I we display the total decay rates for the various multipole channels of $2s_{1/2}\rightarrow 1s_{1/2}$ two-photon decay. In contrast to the photon energy distributions from above, here relativistic calculations have been performed in Coulomb (velocity) as well as Babushkin (length) gauges. In both gauges, negative-energy contribution to the (total) probability of the leading 2E1 transition is about eight orders of magnitude smaller than positive-energy term if decay of low-Z ions is considered but is significantly increased for higher nuclear charges. For the hydrogenlike uranium, for example, the total 2E1 decay rate is enhanced from 2.9041×10^{12} s^{-1} in the velocity gauge and 2.3939×10^{12} s^{-1} in the length gauge to the—gauge independent—"exact" value of 3.8256×10^{12} s^{-1} if, apart from the positive-energy states, the Dirac's states with negative energy are taken into account in the transition amplitude given in Eq. (7). These results clearly indicate the importance of the negative-state summation for the accurate evaluation of 2E1 $2s_{1/2}\rightarrow 1s_{1/2}$ total rates in both, velocity and length gauges. It worth mentioning, however, that while for velocity gauge our findings are in perfect agreement with results reported in Ref. [37] some *discrepancy* was found for

TABLE I. Total rates (in s^{-1}) for the several multipole combinations of $2s_{1/2}\rightarrow 1s_{1/2}$ two-photon decay. Relativistic calculations have been performed within the velocity and length gauges and by carrying out intermediate-state summation over the complete Dirac's spectrum (W_t) as well as over the positive- (W_+) and negative-energy (W_-) solutions only.

		Z=1		Z=54		Z=92	
		Length	Velocity	Length	Velocity	Length	Velocity
2E1	W_+	8.2291(+00)	8.2291(+00)	1.6311(+11)	1.6023(+11)	2.9041(+12)	2.3939(+12)
	W_-	2.4949(−08)	6.2372(−09)	3.8442(+09)	9.6290(+08)	6.8066(+11)	1.7044(+11)
	W_t	8.2291(+00)	8.2291(+00)	1.8592(+11)	1.8592(+11)	3.8256(+12)	3.8256(+12)
E1M2	W_+	2.5372(−10)	2.5372(−10)	4.7949(+07)	4.7940(+07)	8.2955(+09)	8.2714(+09)
	W_-	9.1743(−21)	4.5871(−21)	1.9521(+04)	9.7905(+03)	4.8084(+07)	2.4070(+07)
	W_t	2.5372(−10)	2.5372(−10)	4.9278(+07)	4.9278(+07)	9.1387(+09)	9.1387(+09)
2E2	W_+	3.7296(−11)	4.8617(−13)	9.1765(+06)	1.9624(+05)	2.4730(+09)	9.7383(+07)
	W_-	4.5092(−11)	8.2822(−12)	1.1000(+07)	2.0202(+06)	2.9087(+09)	5.3305(+08)
	W_t	4.9072(−12)	4.9072(−12)	9.8177(+05)	9.8177(+05)	1.7859(+08)	1.7859(+08)
2M1	W_+	5.9021(−20)		1.2691(+05)		3.3321(+08)	
	W_-	1.3804(−11)		3.2695(+06)		7.9720(+08)	
	W_t	1.3804(−11)		3.4027(+06)		1.1093(+09)	

TABLE II. Total rates (in s^{-1}) for the several multipole combinations of $2p_{1/2} \rightarrow 1s_{1/2}$ two-photon decay.

		$Z=1$		$Z=54$		$Z=92$	
		Length	Velocity	Length	Velocity	Length	Velocity
E1M1	W_+	4.1934(−05)	3.2256(−05)	3.0381(+09)	2.2422(+09)	2.1280(+11)	1.4126(+11)
	W_-	1.9355(−05)	9.6773(−06)	1.5701(+09)	7.7417(+08)	1.3745(+11)	6.5902(+10)
	W_t	9.6767(−06)	9.6767(−06)	6.3731(+08)	6.3731(+08)	3.8633(+10)	3.8633(+10)
E1E2	W_+	3.6716(−05)	1.2339(−06)	2.6077(+09)	8.6699(+07)	1.7827(+11)	6.3367(+09)
	W_-	4.5159(−05)	9.6769(−06)	3.1980(+09)	6.7698(+08)	2.1653(+11)	4.4598(+10)
	W_t	6.6117(−06)	6.6117(−06)	4.2942(+08)	4.2942(+08)	2.3584(+10)	2.3584(+10)

calculations performed in length gauge for which Labzowsky and co-workers have argued that the contribution from the Dirac's negative continuum is negligible even for heaviest ions. Based on our theoretical analysis, we argue that such a discrepancy is caused by the incorrect summation performed in Ref. [37] over the electric ($\lambda_\Theta=1$) and longitudinal ($\lambda_\Theta=-1$) components of the electron-photon interaction operator (2) and, hence, over the corresponding second-order amplitudes $S^{j_\nu}_{\lambda_{\Theta_1}\lambda_{\Theta_2}}$. Namely, Labzowsky and co-workers have added terms with $\lambda_\Theta=\pm 1$ *coherently*, constructing thus the (second-order) reduced matrix elements for electric type of transition: $S^{j_\nu}_{(e)}=S^{j_\nu}_{+1+1}+S^{j_\nu}_{+1-1}+S^{j_\nu}_{-1+1}+S^{j_\nu}_{-1-1}$ which—upon squaring—were employed later for the computation of the decay rates [cf. Eqs. (12) and (13) and (33)–(36) of Ref. [37]]. This interpretation is confirmed by our numerical calculations, which reproduce perfectly results from Ref. [37] when we sum matrix elements coherently. However, it has been proven by Drake and co-workers [17,18] that terms $S^{j_\nu}_{\lambda_{\Theta_1}\lambda_{\Theta_2}}$ with $\lambda_{\Theta_{1,2}}=\pm 1$ have to be added *incoherently* if neither the polarization states nor the emission angles of photons are observed. As seen from Eq. (5), such an incoherent summation was performed in this work in order to investigate the differential (in energy) as well as the total two-photon decay rates.

In Table I, besides the leading 2E1 decay channel, we present the results of relativistic calculations for the higher multipole contributions to the $2s_{1/2} \rightarrow 1s_{1/2}$ two-photon transition. The influence of Dirac's negative continuum is obviously different for various multipole combinations. While, for example, the negative-energy contribution to the intermediate-state summation in low-Z domain is negligible for the E1M2 decay it becomes of paramount importance for the 2E2 and 2M1 decay channels; an effect that has been already discussed for the case of the energy-differential decay rates (see upper panel of Fig. 2). Moreover, $2s_{1/2} \rightarrow 1s_{1/2}$ transition with emission of two magnetic dipole (2M1) photons in light ions seems to happen almost *exclusively* via the negative energy (virtual) intermediate states. The total decay rate for this transition together with the negative-energy contribution to the probability of the 2E2 channel (evaluated in Coulomb gauge) gives in atomic units,

$$w_{2M1} + w^{(-)}_{2E2} = 1.248 \times 10^{-6}(\alpha Z)^{10}, \qquad (18)$$

which is in perfect agreement with the semirelativistic formula (15).

One may observe in Table I that the total rates W_t for the leading 2E1 transition as well as for higher multipole decay channels in medium- and high-Z ions are not just sums of the corresponding rates W_+ and W_-. As seen from Eqs. (5)–(8), this comes from the fact that the two-photon transition probabilities contain also terms that arise due to *"interference"* between the positive- and negative-energy Dirac solutions. For the 2E1 and E1M2 transitions in the high-Z realm, the interference terms lead to an enhancement of the total decay rates by 10–30 % when compared with incoherent sum of W_+ and W_- contributions. In contrast, strong reduction of the total rates can be observed for the emission of two electric quadrupole photons (2E2); this effect is most pronounced in the length gauge where the (negative) interference term is as large as -5.21×10^9 s^{-1} for the decay of hydrogenlike uranium ion.

As mentioned above for the computation of the photon energy distributions in low-Z domain, negative-energy contribution to the intermediate-state summation is rather pronounced not only for the higher multipole terms of $2s_{1/2} \rightarrow 1s_{1/2}$ decay but also for the leading E1M1 and E1E2 (two-photon) channels of $2p_{1/2} \rightarrow 1s_{1/2}$ transition. Our relativistic calculations displayed in Table II indicate that one should also take into account the negative-continuum summation for an accurate evaluation of the *total* decay rates for these two decay channels. For the decay of light elements, sizable contribution from the negative-continuum intermediate states arises both in length and velocity gauges. Again, these results partially question the predictions by Labzowsky and coworkers [37] who claimed a minor role of negative energy terms for E1M1 and E1E2 calculations in length gauge. For the velocity gauge, in contrast, our relativistic calculations,

$$w^{(-)}_{E1M1} + w^{(-)}_{E1E2} = 5.822\times 10^{-5}(\alpha Z)^8, \qquad (19)$$

are in good agreement both, with the semirelativistic prediction [Eq. (17)] and data presented in Ref. [37].

Similar to the $2s_{1/2} \rightarrow 1s_{1/2}$ decay, the probabilities for the E1M1 and E1E2 transitions are also strongly affected by the mixing between the positive- and negative-energy states and, hence, cannot be represented as sums of W_+ and W_- terms. As seen in Table II, these (incoherent) sums sufficiently exceed the total rates W_t for all range of nuclear charge Z

indicating thus that such a mixing plays a destructive role for the two-photon decay of $2p_{1/2}$ hydrogenic state.

IV. SUMMARY AND OUTLOOK

In conclusion, the two-photon decay of hydrogenlike ions has been reinvestigated within the framework of second-order perturbation theory, based on Dirac's relativistic equation. Special attention has been paid to the summation over the intermediate ionic states which occurs in such a framework and runs over *complete* one-particle spectrum, including a summation over discrete (bound) states as well as the integration over the positive and negative continua. In particular, we discussed the role of the *negative* energy continuum in an accurate evaluation of the second-order transition amplitudes and, hence, the energy differential as well as total decay rates. Detailed calculations of these rates have been presented for the $2s_{1/2} \rightarrow 1s_{1/2}$ and $2p_{1/2} \rightarrow 1s_{1/2}$ two-photon transitions in neutral hydrogen as well as hydrogenlike xenon and uranium ions. As seen from the results obtained, both the total decay probabilities and the energy distributions of the simultaneously emitted photons can be strongly affected by the negative-state summation not only for heavy ions but also for low-Z ones. We demonstrate, however, that the role of Dirac's negative continuum becomes most pronounced for the higher (nondipole) terms in the expansion of the electron-photon interaction; similar effect has been recently reported for the theoretical description of hydrogenlike systems exposed to intense electromagnetic pulses [33].

In the present work, we have restricted our discussion of the negative energy contribution to the second-order calculations of the total and energy-differential decay rates. Even stronger effects due to the Dirac's negative continuum can be expected, however, for the angular and polarization correlations between emitted photons. Theoretical investigation of these correlations, which requires also detailed analysis of interference terms between the various (two-photon) multipole combinations is currently underway.

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