Two-photon transitions with cascades: Two-photon transition rates and two-photon level widths

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An ambiguity of the separation of cascades from "pure" two-photon decay is confirmed with accurate numerical calculations in a gauge-invariant way. A direct evaluation of the two-photon decay width of excited states in H-like ions via the imaginary part of two-loop self-energy is presented. We demonstrate that there is a fundamental difference between the level width and the transition probability in the presence of cascades. The two-photon widths are shown to be different from the two-photon decay rates for the transitions including cascades.

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I. INTRODUCTION

The theory of the multiphoton transitions in atoms on the basis of quantum mechanics (QM) started with the work by Göppert-Mayer [1]. The first evaluation of the two-photon decay rate $2s \to 1s + 2\gamma(E1)$ in H was performed by Breit and Teller [2] (see correction to this work in Ref. [3]). The accurate nonrelativistic evaluation of the $2s \to 1s + 2\gamma(E1)$ transition rate in H was performed by the authors of [4]. The first fully relativistic calculation of this transition was given in Ref. [5] and later in Refs. [6–8]. Quantum electrodynamical (QED) corrections to the two-photon decay of the 2s level in H were studied in Ref. [9]. Fully relativistic calculations of different multiphoton transitions can be found in Refs. [10–14].

During the last decade the multiphoton transitions became of high interest for astrophysics. This interest was triggered by the accurate measurements of the temperature and polarization distribution of the cosmic microwave background (CMB) [15,16]. The CMB was formed in the epoch of the cosmological H recombination and an accurate theory of this formation is therefore required. The modern theory of the cosmological recombination starts from works by Zel'dovich, Kurt, and Sunyaev [17] and Peebles [18]. According to the authors of [17,18], the $2s \to 1s + 2\gamma(E1)$ transition was found to be the main channel for the radiation escape from the matter and formation of CMB. Hence the recent properties of the CMB are defined by the two-photon processes during the cosmological recombination epoch.

Apart from the $2s \to 1s + 2\gamma(E1)$ transition as it was noted recently in Ref. [19,20] the two-photon decays from the excited states with the principal quantum numbers $n > 2$ also can contribute to the radiation escape at the 1% level of accuracy. This idea was further developed in Ref. [21–24]. There is a difference between the decay of ns ($n > 2$) and nd states and the decay of the 2s state. This difference is due to the presence of cascade transitions as the dominant decay channels in case of ns ($n > 2$) and nd levels. For the 2s level the cascades are absent. The problem of the separation of "pure" two-photon emission from the cascade photons arises in connection with the radiation escape probability.

The laboratory measurements of the two-photon decay rates were reported for $2s \to 1s + 2\gamma$ transitions in H [24], in H-like He [25], and in the H-like highly charged ions (HCI) [26,27].

The cascade separation problem appeared to be nontrivial. For the first time this question was raised in Ref. [28] for the two-photon transitions in HCI. The same problem was considered later in Ref. [29]. In Refs. [30,31] a general QED approach was developed which allowed for a rigorous description of the multiphoton cascade transitions. This approach was based on Low's theory [32] of the spectral line profile in QED. In Refs. [30,31] it was demonstrated that the separation of the cascade contribution from the "pure" two-photon decay rate can not be achieved in an unambiguous way. While in Ref. [30] this problem was studied for HCI, in Refs. [33,34] it was discussed in connection with the cosmological recombination. As an example the two-photon decay of the 3s level in H was considered: $3s \to 1s + 2\gamma(E1)$. The ambiguity of the separation of this cascade was demonstrated numerically: The result was dependent on the method of separation. In this paper we reconsider the separation problem for the same example. On a basis of more accurate numerical calculations we are able to show that while the contribution of "pure" two-photon and interference terms vary essentially with the method of cascade separation, the total two-photon transition rate remains invariant. The later circumstance was not obvious from the results in Ref. [33] (because of the poorer accuracy of the numerical calculations). Moreover, in this paper we prove the gauge invariance of our results by a direct numerical check.

In the first part of our paper we will apply exclusively nonrelativistic theory which is sufficient for the description of the processes in H. This means that we neglect relativistic and QED corrections to the transition probabilities though we will use the QED description for the derivation of the expressions for the decay probabilities. We will neglect also the multipole radiation, i.e., we restrict our studies with $E1$ photons, considering only the $E1E1$ two-photon, $E1E1E1$ three-photon, and so on, transitions. These transitions are dominant for one-, two-, three-photon, and so on processes respectively, though particular transition rates for the one-photon multipole electric or magnetic transitions can exceed the transition rates for the multiphoton $E1$ transitions.

In the second part of our paper we investigate the problem of the evaluation of the two-photon level width via the imaginary part of two-loop electron self-energy. This evaluation was performed in a series of works [35–37] and it was claimed that the results obtained present an unambiguous way to evaluate the "pure" two-photon contributions to the two-photon transitions with cascades. The derivations were made nonrelativistically. The numerical values differ essentially from the ones in Ref. [33]. In the present paper we rederive the imaginary part of the two-loop contribution in a fully relativistic way and confirm exactly the results [35–37]. However, we argue that these results have no direct connection to the two-photon decay rates for the transitions with cascades as it was claimed in Refs. [35–37]. The numbers obtained in Refs. [35–37] and confirmed in our present paper are the "pure" two-photon contributions to the level widths. They represent small corrections to the total level widths. Moreover, these contributions can be even negative (see Sec. V), which confirms our treatment of these contributions.

The QED perturbation theory for the self-energy radiative corrections to the electron energy levels in atoms treats consequently the one-loop (second-order in the coupling constant) and two-loop (fourth-order in the coupling constant) contributions. The imaginary parts of these corrections correspond, respectively, to the one-photon width, two-photon width, three-photon width and so on

$$\Gamma_n = \Gamma_n^{1\gamma} + \Gamma_n^{2\gamma} + \Gamma_n^{3\gamma} + \cdots . \quad (1)$$

Here the index n denotes an excited atomic level. It is well known that the one-photon width $\Gamma_n^{1\gamma}$ is equal to the sum of the one-photon transitions to lower levels

$$\Gamma_n^{1\gamma} = \sum_{n'<n} \Gamma_{nn'}^{1\gamma} = \sum_{n'<n} W_{nn'}^{1\gamma}. \quad (2)$$

Here we distinguish the partial widths $\Gamma_{nn'}^{1\gamma}$ corresponding to the transitions $n \to n'$ and the transition rates $W_{nn'}^{1\gamma}$ for these transitions. In the case of 1γ transitions

$$\Gamma_{nn'}^{1\gamma} = W_{nn'}^{1\gamma}. \quad (3)$$

However, the situation with 2γ transitions is different. Similar to Eq. (2) we can express the width $\Gamma_n^{2\gamma}$ via partial widths

$$\Gamma_n^{2\gamma} = \sum_{n'<n} \Gamma_{nn'}^{2\gamma}, \quad (4)$$

but now the partial widths $\Gamma_{nn'}^{2\gamma}$, in general, are not equal to the corresponding transition rates $W_{nn'}^{2\gamma}$. The equality

$$\Gamma_{nn'}^{2\gamma} = W_{nn'}^{2\gamma} \quad (5)$$

holds only in the absence of cascades. Examples with $E1$ photons only, are $2s \to 1s + 2\gamma$, $3s \to 2s + 2\gamma$, $3p \to 2p + 2\gamma$, and so on. For the transitions with cascades

$$\Gamma_{nn'}^{2\gamma} \neq W_{nn'}^{2\gamma}. \quad (6)$$

The widths in Eq. (1) satisfy the inequalities

$$\Gamma_n^{1\gamma} \gg \Gamma_n^{2\gamma} \gg \Gamma_n^{3\gamma} \gg \cdots , \quad (7)$$

so that the perturbation expansion for the imaginary part of the energy converges in the same (asymptotic) sense as the real part. For transitions with cascades the order of magnitude for the transition rates involving any number of photons is parametrically the same as $W_{nn'}^{1\gamma}$:

$$W_{2p1s}^{1\gamma} \sim W_{3s1s}^{2\gamma} \sim W_{3p1s}^{3\gamma}. \quad (8)$$

In Eq. (8) only transitions with $E1$ photons are included. In Refs. [35–37] the widths $\Gamma_{nn'}^{2\gamma}$ were evaluated for different nn'. The evaluation itself is quite correct but the corresponding values cannot be assigned to the "pure" two-photon transition rates due to the inequality (6). We will extend the calculations performed in Refs. [35–37] to the more wide set of transitions.

To confirm our conclusions we will also employ the adiabatic S-matrix approach for the evaluation of the two-photon level widths [33]. This will justify the derivations made by the direct evaluation of the imaginary part of the two-loop self-energy graphs.

II. TRANSITIONS RATES FOR THE TWO-PHOTON TRANSITIONS WITH CASCADES

In this section we describe the two-photon transitions to the ground state taking the $ns \to 1s + 2\gamma$ transitions as an example. We will be interested in transitions with cascades, so we will consider $n > 2$ since the $2s \to 1s + 2\gamma$ transition does not contain cascades. The full QED description of any process in an atom should start with the ground state, i.e., the excitation of the decaying state should be always included. In this way the theory of multiphoton processes in atoms was developed by the authors of [30,31]. For the resonant processes, e.g., for the resonant photon scattering the absorption part of the process can be well separated from the emission part, so that the description of the decay process independent of the excitation becomes possible. Still the way of the excitation influences to some extent the decay. Having in mind the cosmological recombination processes in H we consider the resonance two-photon scattering on the ground $1s$ state with

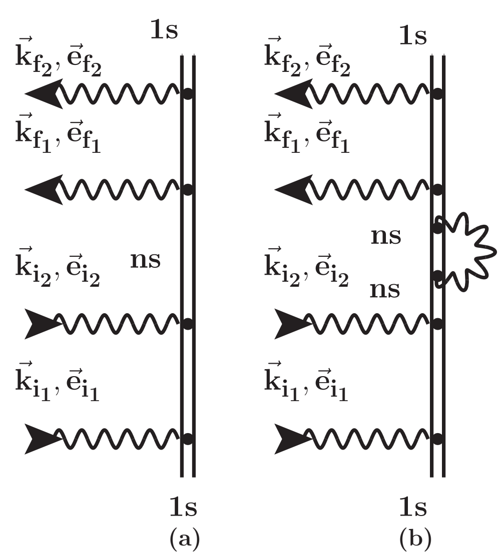

FIG. 1. Feynman graph describing the two-photon resonance scattering on the ground state of a H atom with excitation of the ns state and the resonance condition $\omega_{i_1} + \omega_{i_2} = E_{ns} - E_{1s}$. The double solid lines denote an electron in the field of the nucleus and the wavy lines denote the absorbed, emitted, and virtual photons. The notations $\vec{k}_{i_1}\vec{e}_{i_1}$, $\vec{k}_{i_2}\vec{e}_{i_2}$ correspond to the momentum and polarization of the absorbed (initial) photons, $\vec{k}_{f_1}\vec{e}_{f_1}$, $\vec{k}_{f_2}\vec{e}_{f_2}$ correspond to the emitted (final) photons. (a) The generic process of the resonant scattering is depicted. (b) The electron self-energy insertion in the central electron propagator is made.

resonances corresponding to the ns states. This process is natural in the case of cosmological recombination since the two photons released in the process of the decay of one atom can be absorbed by another atom. The Feynman graph corresponding to the resonant photon scattering is depicted in Fig. 1(a).

The S-matrix element, corresponding to Fig. 1(a) looks like

$$S_{1s}^{(4)sc} = (-ie)^4 \int d^4x_1 d^4x_2 d^4x_3 d^4x_4 \overline{\psi}_{1s}(x_1)\gamma_{\mu_1} A_{\mu_1}^{*(\vec{k}_{f_2}\vec{e}_{f_2})}(x_1) S(x_1,x_2)\gamma_{\mu_2} A_{\mu_2}^{*(\vec{k}_{f_1}\vec{e}_{f_1})}(x_2) S(x_2,x_3)\gamma_{\mu_3} A_{\mu_3}^{(\vec{k}_{i_2}\vec{e}_{i_2})}(x_3) \times S(x_3,x_4)\gamma_{\mu_4} A_{\mu_3}^{(\vec{k}_{i_1}\vec{e}_{i_1})}(x_4)\psi_{1s}(x_4), \tag{9}$$

where

$$\psi_n(x) = \psi_n(\vec{r})e^{-iE_n t}, \tag{10}$$

the spatial part $\psi_n(\vec{r})$ is the solution of the Dirac equation for the atomic electron, E_n is the Dirac energy, $\overline{\psi}_n = \psi_n^+\gamma_0$ is the Dirac conjugated wave function, $\gamma_\mu \equiv (\gamma_0,\vec{\gamma})$ are the Dirac matrices, and $x \equiv (\vec{r},t)$ are the space-time coordinates. In this paper the Euclidean metric with an imaginary fourth component is employed. The photon wave function (photon field) is described by

$$A_\mu^{(\vec{k},\vec{e})}(x) = \sqrt{\frac{2\pi}{\omega}} e_\mu e^{ik_\mu x_\mu} = A_\mu^{(\vec{k},\vec{e})}(\vec{r})e^{-i\omega t}, \tag{11}$$

where $k \equiv (\vec{k},i\omega)$ is the photon momentum four-vector, $\vec{k}$ is the photon wave vector, $\omega = |\vec{k}|$ is the photon frequency, e_μ are the components of the photon polarization four-vector, $\vec{e}$ is the three-dimensional polarization vector for real photons, $A_\mu^{(\vec{k},\vec{e})}$ corresponds to the absorbed photon, and $A_\mu^{*(\vec{k},\vec{e})}$ corresponds to the emitted photon. The electron propagator for bound electrons we present in the form of the eigenmode decomposition with respect to one-electron eigenstates [38]

$$S(x_1,x_2) = \frac{1}{2\pi i}\int_{-\infty}^{\infty} d\omega e^{i\omega(t_1-t_2)} \sum_n \frac{\psi_n(\vec{r}_1)\overline{\psi}_n(\vec{r}_2)}{E_n(1-i0)+\omega}. \tag{12}$$

The insertion of the expressions (10) to (12) into Eq. (9) and performing the integrations over time and frequency variables yields

$$S_{1s}^{(4)sc} = -2\pi i e^4 \delta(\omega_{f_1}+\omega_{f_2}-\omega_{i_1}-\omega_{i_2}) \sum_{n_1n_2n_3} \frac{\left(\gamma_{\mu_1}A_{\mu_1}^{*(\vec{k}_{f_2}\vec{e}_{f_2})}\right)_{1sn_1}\left(\gamma_{\mu_2}A_{\mu_2}^{*(\vec{k}_{f_1}\vec{e}_{f_1})}\right)_{n_1n_2}}{[E_{n_1}(1-i0)-\omega_{f_2}-E_{1s}][E_{n_2}(1-i0)-\omega_{f_2}-\omega_{f_1}-E_{1s}]} \times \frac{\left(\gamma_{\mu_3}A_{\mu_3}^{(\vec{k}_{i_2}\vec{e}_{i_2})}\right)_{n_2n_3}\left(\gamma_{\mu_4}A_{\mu_4}^{(\vec{k}_{i_1}\vec{e}_{i_1})}\right)_{n_31s}}{E_{n_3}(1-i0)+\omega_{i_2}-\omega_{f_2}-\omega_{f_1}-E_{1s}}. \tag{13}$$

The amplitude U of the elastic photon scattering is related to the S matrix via [38]

$$S = -2\pi i\delta(\omega_{f_1}+\omega_{f_2}-\omega_{i_1}-\omega_{i_2})U. \tag{14}$$

Accordingly, for the scattering amplitude we have

$$U_{1s}^{(4)sc} = e^4 \sum_{n_1n_2n_3} \frac{\left(\gamma_{\mu_1}A_{\mu_1}^{*(\vec{k}_{f_2}\vec{e}_{f_2})}\right)_{1sn_1}\left(\gamma_{\mu_2}A_{\mu_2}^{*(\vec{k}_{f_1}\vec{e}_{f_1})}\right)_{n_1n_2}}{[E_{n_1}(1-i0)-\omega_{f_2}-E_{1s}][E_{n_2}(1-i0)-\omega_{f_2}-\omega_{f_1}-E_{1s}]} \frac{\left(\gamma_{\mu_3}A_{\mu_3}^{(\vec{k}_{i_2}\vec{e}_{i_2})}\right)_{n_2n_3}\left(\gamma_{\mu_4}A_{\mu_4}^{(\vec{k}_{i_1}\vec{e}_{i_1})}\right)_{n_31s}}{E_{n_3}(1-i0)+\omega_{i_2}-\omega_{f_2}-\omega_{f_1}-E_{1s}}. \tag{15}$$

For the resonant scattering process the photon frequencies satisfy the relation

$$\omega_{f_1}+\omega_{f_2} = \omega_{i_1}+\omega_{i_2} = E_{ns}-E_{1s}, \tag{16}$$

so that we have to retain only one term in the sum over n_2: $n_2 = ns$. Then, replacing the last denominator in Eq. (15) by

$$E_{n_3}+\omega_{i_2}-\omega_{f_1}-\omega_{f_2}-E_{1s} = E_{n_3}-\omega_{i_1}-E_{1s}, \tag{17}$$

we obtain

$$U_{1s(ns)}^{(4)sc} = e^4 \frac{1}{E_{ns}-\omega_{f_2}-\omega_{f_1}-E_{1s}} \sum_{n_1} \frac{\left(\gamma_{\mu_1}A_{\mu_1}^{*(\vec{k}_{f_2}\vec{e}_{f_2})}\right)_{1sn_1}\left(\gamma_{\mu_2}A_{\mu_2}^{*(\vec{k}_{f_1}\vec{e}_{f_1})}\right)_{n_1ns}}{E_{n_1}(1-i0)-\omega_{f_2}-E_{1s}} \sum_{n_3} \frac{\left(\gamma_{\mu_3}A_{\mu_3}^{(\vec{k}_{i_2}\vec{e}_{i_2})}\right)_{nsn_3}\left(\gamma_{\mu_4}A_{\mu_4}^{(\vec{k}_{i_1}\vec{e}_{i_1})}\right)_{n_31s}}{E_{n_3}(1-i0)-\omega_{i_1}-E_{1s}}. \tag{18}$$

Equation (18) reveals that in the resonance approximation a scattering amplitude factorizes into emission and absorption parts. The first energy denominator in Eq. (18) should be attached to the emission or absorption part depending on what we want to describe: the emission or absorption process. The two-photon emission amplitude can be presented as

$$U_{ns-1s}^{(2\gamma)em} = e^2 \frac{1}{E_{ns}-\omega_{f_2}-\omega_{f_1}-E_{1s}} \sum_{n_1} \frac{\left(\gamma_{\mu_1}A_{\mu_1}^{*(\vec{k}_{f_2}\vec{e}_{f_2})}\right)_{1sn_1}\left(\gamma_{\mu_2}A_{\mu_2}^{*(\vec{k}_{f_1}\vec{e}_{f_1})}\right)_{n_1ns}}{E_{n_1}(1-i0)-\omega_{f_2}-E_{1s}}. \tag{19}$$

The energy conservation law which follows from Eq. (14) reads

$$\omega_{f_1} + \omega_{f_2} = \omega_{i_1} + \omega_{i_2}. \tag{20}$$

The resonance condition can be written in the form

$$\left|\omega_{i_1} + \omega_{i_2} - E_{ns} + E_{1s}\right| = \left|\omega_{f_1} + \omega_{f_2} - E_{ns} + E_{1s}\right| \lesssim \Gamma_{ns}, \tag{21}$$

where Γ_{ns} is the total width of the ns level. In cases when we can neglect Γ_{ns} in Eq. (21) this equation takes the form of the energy conservation law

$$\omega_{f_1} + \omega_{f_2} = E_{ns} - E_{1s}, \tag{22}$$

in particular, we can employ condition (22) in the second energy denominator in Eq. (19), but not in the first denominator.

Now we have to take into account the form of the resonance corresponding to the first energy denominator in Eq. (19). For this purpose we will apply the procedure first introduced in QED by Low [32] (see also [31]). This procedure consists of an infinite number of the electron one-loop self-energy insertions in the central electron propagator in Fig. 1(a) in the resonance approximation. The first term of this sequence is depicted in Fig. 1(b). Returning back to the scattering amplitude Eq. (15) we proceed along the same way as earlier in the resonance approximation. Continuing Low's sequence and summing up the arising geometric progression yields

$$U^{(2\gamma)em}_{ns-1s} = e^2 \frac{1}{\omega_{f_2} + \omega_{f_1} + E_{1s} - E_{ns} - \left[\widehat{\Sigma}\left(\omega_{f_1} + \omega_{f_2} + E_{1s}\right)\right]_{ns,ns}} \times \sum_{n_1} \frac{\left(\gamma_{\mu_1} A^{*(\vec{k}_{f_2}\vec{e}_{f_2})}_{\mu_1}\right)_{n_1 ns} \left(\gamma_{\mu_2} A^{*(\vec{k}_{f_1}\vec{e}_{f_1})}_{\mu_2}\right)_{n_1 ns}}{\omega_{f_1} + E_{1s} - E_{n_1}}, \tag{23}$$

where the matrix element of the one-loop electron self-energy operator is defined as [31]

$$[\widehat{\Sigma}(\xi)]_{AB} = \frac{e^2}{2\pi i} \sum_n \int d\Omega \frac{\left[\gamma_{\mu_1}\gamma_{\mu_2} I_{\mu_1\mu_2}(|\Omega|, r_{12})\right]_{AnnB}}{\xi - \Omega - E_n(1 - i0)}, \tag{24}$$

$$I_{\mu_1\mu_2} = \frac{\delta_{\mu_1\mu_2}}{r_{12}} e^{i|\Omega| r_{12}}. \tag{25}$$

Here $x \equiv (\vec{r}, t)$, $r_{12} = |\vec{r}_1 - \vec{r}_2|$.

At the point of the resonance we expand the operator

$$\widehat{\Sigma}\left(\omega_{f_1} + \omega_{f_2} + E_{1s}\right) = \widehat{\Sigma}(E_{ns}) + \cdots \tag{26}$$

and use the equality

$$[\widehat{\Sigma}(E_{ns})]_{ns,ns} = L^{SE}_{ns} - \frac{i}{2}\Gamma^{1\gamma}_{ns}, \tag{27}$$

where $L^{\rm SE}_{ns}$ is the lowest-order (one-loop) electron self-energy part of the Lamb shift for the level ns and $\Gamma^{1\gamma}_{ns}$ is the one-photon level width. An explicit relativistic expression for $\Gamma^{1\gamma}_A$ for an arbitrary level A can be found in Ref. [39]. The contribution of $L^{\rm SE}_{ns}$ does not play any significant role in our further derivations and will be omitted. To the expression (23) we have to add also another term corresponding to the graphs in Fig. 1 with interchanged positions of the $\vec{k}_{f_1}\vec{e}_{f_1}$ and $\vec{k}_{f_2}\vec{e}_{f_2}$ photon lines.

Taking Eq. (23) by the square modulus, integrating over the momenta of the emitted photons, and summing over the photon polarizations we obtain the absolute probability for the two-photon emission $db^{2\gamma}_{ns-1s}(\omega)$, where ω is the frequency of one of the emitted photons; the frequency of the second photon is determined via Eq. (22). The quantity $b^{2\gamma}_{ns-1s}$ presents the differential branching ratio

$$db^{2\gamma}_{ns-1s}(\omega) = \frac{dW^{2\gamma}_{ns-1s}(\omega)}{\Gamma_{ns}}, \tag{28}$$

where $dW^{2\gamma}_{ns-1s}(\omega)$ is the differential transition rate. The total transition rate then results as

$$W^{2\gamma}_{ns-1s} = \frac{1}{2}\int_0^{\omega_0} dW^{2\gamma}_{ns-1s}(\omega), \tag{29}$$

$\omega_0 = E_{ns} - E_{1s}$. The factor of 1/2 in Eq. (29) reflects the integration over $d\omega_{f_1} d\omega_{f_2}$ together with the additional condition Eq. (22).

III. TWO-PHOTON 3s-1s TRANSITION

The further investigation of the two-photon transitions with cascades should be performed separately for different n values. In this section we will continue this investigation for the $3s \to 1s + 2\gamma$ transition. The Feynman graphs which describe this process are depicted in Fig. 2. The two-photon emission

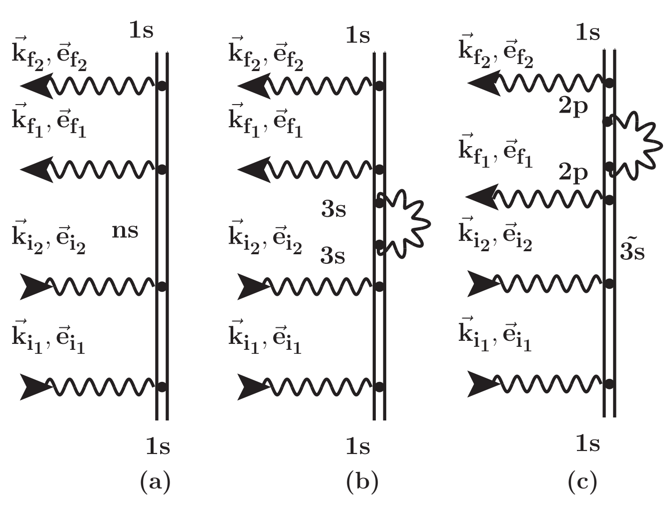

FIG. 2. Feynman graph describing the two-photon resonance scattering on the ground state of a H atom with excitation of the $3s$ state. Resonance condition is $\omega_{i_1} + \omega_{i_2} = E_{3s} - E_{1s}$ and the decay resonances are $\omega_f^{\rm res.1} = E_{3s} - E_{2p}$, $\omega_f^{\rm res.2} = E_{3s} - E_{2p}$. (a) The basic process of the resonance scattering is depicted. (b) The electron self-energy insertion in the central propagator is made. (a), (b) repeat the graphs in Figs. 1(a) and 1(b) for the ns resonance for the particular case of $n = 3$. (c) The electron self-energy insertion in the upper electron propagator is shown. Notation $\tilde{3}s$ means that the Low procedure is already performed for the central electron propagator. The other notations are the same as in Fig. 1.

amplitude for this process looks like

$$U^{2\gamma}_{3s-1s} = e^2 \frac{1}{\omega_{f_2} + \omega_{f_1} + E_{1s} - E_{3s} + \frac{i}{2}\Gamma_{3s}} \times \sum_{n_1} \left\{ \frac{\left(\gamma_{\mu_1} A^{*(\vec{k}_{f_2}\vec{e}_{f_2})}_{\mu_1}\right)_{1sn_1} \left(\gamma_{\mu_2} A^{*(\vec{k}_{f_1}\vec{e}_{f_1})}_{\mu_2}\right)_{n_1 3s}}{\omega_{f_2} + E_{1s} - E_{n_1}} + \frac{\left(\gamma_{\mu_1} A^{*(\vec{k}_{f_1}\vec{e}_{f_1})}_{\mu_1}\right)_{1sn_1} \left(\gamma_{\mu_2} A^{*(\vec{k}_{f_2}\vec{e}_{f_2})}_{\mu_2}\right)_{n_1 3s}}{\omega_{f_1} + E_{1s} - E_{n_1}} \right\}. \tag{30}$$

For the $3s - 1s$ two-photon transition only one cascade is possible: $3s - 2p - 1s$. Accordingly, the two new resonance conditions arise:

$$\omega^{\text{res.1}} = E_{3s} - E_{2p}, \tag{31}$$

$$\omega^{\text{res.2}} = E_{2p} - E_{1s}. \tag{32}$$

Let us consider first the cascade contribution to Eq. (30). For this purpose we have to set $n_1 = 2p$, which gives

$$U^{2\gamma,\text{ cascade}}_{3s-2p-1s} = e^2 \frac{1}{\omega_{f_2} + \omega_{f_1} + E_{1s} - E_{3s} + \frac{i}{2}\Gamma_{3s}} \times \left\{ \frac{\left(\gamma_{\mu_1} A^{*(\vec{k}_{f_2}\vec{e}_{f_2})}_{\mu_1}\right)_{1s2p} \left(\gamma_{\mu_2} A^{*(\vec{k}_{f_1}\vec{e}_{f_1})}_{\mu_2}\right)_{2p3s}}{\omega_{f_2} + E_{1s} - E_{2p}} + \frac{\left(\gamma_{\mu_1} A^{*(\vec{k}_{f_1}\vec{e}_{f_1})}_{\mu_1}\right)_{1s2p} \left(\gamma_{\mu_2} A^{*(\vec{k}_{f_2}\vec{e}_{f_2})}_{\mu_2}\right)_{2p3s}}{\omega_{f_1} + E_{1s} - E_{2p}} \right\}. \tag{33}$$

The first term in the curly brackets in Eq. (33) corresponds to the resonance (31). Applying the Low procedure (the insertion of the infinite chain of the electron self-energy corrections in the resonance approximation and summation of the arising geometric progression) to the upper electron propagator in Fig. 2 [see Fig. 2(c)] we find

$$U^{2\gamma,\text{ cascade}}_{3s-2p-1s} = e^2 \frac{1}{\omega_{f_2} + \omega_{f_1} + E_{1s} - E_{3s} + \frac{i}{2}\Gamma_{3s}} \times \left\{ \frac{\left(\gamma_{\mu_1} A^{*(\vec{k}_{f_2}\vec{e}_{f_2})}_{\mu_1}\right)_{1s2p} \left(\gamma_{\mu_2} A^{*(\vec{k}_{f_1}\vec{e}_{f_1})}_{\mu_2}\right)_{2p3s}}{\omega_{f_2} + E_{1s} - E_{2p} + \frac{i}{2}\Gamma_{2p}} + \frac{\left(\gamma_{\mu_1} A^{*(\vec{k}_{f_1}\vec{e}_{f_1})}_{\mu_1}\right)_{1s2p} \left(\gamma_{\mu_2} A^{*(\vec{k}_{f_2}\vec{e}_{f_2})}_{\mu_2}\right)_{2p3s}}{\omega_{f_1} + E_{1s} - E_{2p} + \frac{i}{2}\Gamma_{2p}} \right\}. \tag{34}$$

Now we take $U^{2\gamma,\text{ cascade}}_{3s-2p-1s}$ by the square modulus, integrate over emitted photon directions, and sum over the polarizations. The one-photon transition rates arise as a result:

$$W^{1\gamma}_{AB} = 2\pi\omega^2 e^2 \sum_{\vec{e}} \int \frac{d\vec{\nu}}{(2\pi)^3} \left|\left(\gamma_\mu A^{*(\vec{k}\vec{e})}_\mu\right)_{AB}\right|^2, \tag{35}$$

where $\vec{\nu} \equiv \vec{k}/\omega$, A, B are the atomic states, $\omega = E_A - E_B$, E_A, E_B are the energy of the states A, B, respectively.

Consider first the square modulus of the first term in the curly brackets in Eq. (34) together with the factor outside the brackets. This term corresponds to the contribution of the resonance 1 in Eq. (31), i.e., the upper link of the cascade $3s - 2p - 1s$. Therefore we have to integrate first over frequency ω_{f_2} of the second emitted photon. In principle, the integration over both photon frequencies should be done with Eq. (22) taken into account. However, we perform the integration over ω_{f_2} in the complex plane. Since only the pole terms contribute we can extend the interval of integration to $(-\infty,+\infty)$ and disregard Eq. (22). Then using the Cauchy theorem after some algebraic transformations we obtain the following cascade (resonance 1) contribution to the differential branching ratio:

$$db^{2\gamma(\text{res.1})}_{3s-2p-1s}(\omega) = \frac{1}{2\pi}\frac{\Gamma_{3s}+\Gamma_{2p}}{\Gamma_{3s}\Gamma_{2p}} \frac{W^{1\gamma}_{3s-2p}(\omega^{\text{res.1}})W^{1\gamma}_{2p-1s}(\omega^{\text{res.2}})d\omega}{(\omega-\omega^{\text{res.1}})^2 + \frac{1}{4}(\Gamma_{3s}+\Gamma_{2p})^2} \tag{36}$$

(here we have changed notation for the frequency from ω_{f_1} to ω). The differential branching ratio $db^{2\gamma}$ is connected with the differential transition rate $dW^{2\gamma}$ via Eq. (28). Combining now the formulas (36) and (28) we arrive at the expression for the cascade (upper link) contribution to the transit rate

$$dW^{2\gamma(\text{res.1})}_{3s-2p-1s}(\omega) = \frac{1}{2\pi}\frac{\Gamma_{3s}+\Gamma_{2p}}{\Gamma_{2p}} \frac{W^{1\gamma}_{3s-2p}(\omega^{\text{res.1}})W^{1\gamma}_{2p-1s}(\omega^{\text{res.2}})d\omega}{(\omega-\omega^{\text{res.1}})^2 + \frac{1}{4}(\Gamma_{3s}+\Gamma_{2p})^2}. \tag{37}$$

The definition (28) concerns not only the cascade contribution to $dW^{2\gamma}_{ns-1s}$ but all other contributions, i.e., noncascade, or "pure" two-photon contribution and the interference between cascade and noncascade contributions. The interference between different resonances (links of one cascade) or between different cascades are usually negligible since they correspond to different frequency regions. Hence we can write

$$db^{2\gamma}_{ns-1s} = db^{2\gamma,\text{ cascade}}_{ns-1s} + db^{2\gamma,\text{ pure}}_{ns-1s} + db^{2\gamma,\text{ interference}}_{ns-1s} = \frac{dW^{2\gamma}_{ns-1s}}{\Gamma_{ns}} = \frac{1}{\Gamma_{ns}}\left(dW^{2\gamma,\text{ cascade}}_{ns-1s} + dW^{2\gamma,\text{ pure}}_{ns-1s} + dW^{2\gamma,\text{ interference}}_{ns-1s}\right). \tag{38}$$

Integration Eq. (28) over the remaining frequency ω [see Eq. (29)] will give the total branching ratio

$$b^{2\gamma}_{ns-1s} = \frac{W^{2\gamma}_{ns-1s}}{\Gamma_{ns}}. \tag{39}$$

The cascade contribution consists of two terms

$$dW^{2\gamma,\text{ cascade}}_{3s-1s} = dW^{2\gamma,\text{ res.1}}_{3s-2p-1s} + dW^{2\gamma,\text{ res.2}}_{3s-2p-1s}. \tag{40}$$

The second term in the curly brackets in Eq. (34) corresponds to the resonance 2 Eq. (32), i.e., to the lower link of the cascade. Taking it by the square modulus, after integration over the directions of emitted photons, and the summation over the photon polarizations, we have to integrate it over the frequency of the first emitted photon, i.e., again over ω_{f_2} [the photons are

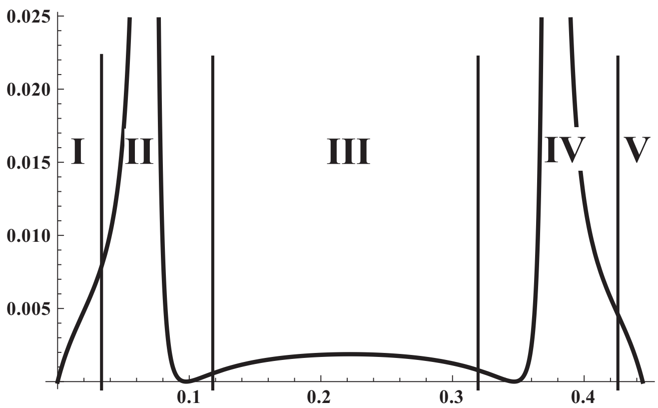

FIG. 3. The frequency distribution $dW^{2\gamma}_{3s-1s}(\omega)/d\omega$ for the two-photon $3s \to 1s + 2\gamma$ transition, divided by α^6 (α is the fine structure constant). The boundaries for the frequency subintervals I–V are indicated as vertical lines.

interchanged compared to the first term in Eq. (34)]. Replacing notation ω_{f_1} to ω we obtain

$$dW^{2\gamma(\text{res.2})}_{3s-2p-1s}(\omega) = \frac{1}{2\pi}\frac{W^{1\gamma}_{3s-2p}(\omega^{\text{res.1}})W^{1\gamma}_{2p-1s}(\omega^{\text{res.2}})d\omega}{(\omega-\omega^{\text{res.2}})^2+\frac{1}{4}\Gamma^2_{2p}}. \tag{41}$$

IV. AMBIGUITY OF THE SEPARATION OF CASCADES AND PURE TWO-PHOTON EMISSION

In this section we will demonstrate the ambiguity of the separation of contributions $dW^{2\gamma,\text{ cascade}}_{ns-1s}$, $dW^{2\gamma,\text{ pure}}_{ns-1s}$ and $dW^{2\gamma,\text{ interference}}_{ns-1s}$ in Eq. (38), taking the transition $3s \to 1s + 2\gamma$ as an example. This ambiguity was demonstrated numerically by the authors of [33], however, the accuracy of the calculations was not sufficient to show that the total value for the transition rate $W^{2\gamma}_{3s-1s}$ is always the same, independent of the way of defining the separate terms in Eq. (38). Now, with more accurate calculations we can show this. Moreover we can prove also the gauge invariance of the total transition rate $W^{2\gamma}_{3s-1s}$ with cascade terms, regularized according to the Low procedure, i.e., with Eqs. (37) and (41).

The integration over the entire frequency interval $[0,\omega_0]$ in Eq. (29) we split into several intervals, namely five in the case of the $3s \to 1s + 2\gamma$ transition (see Fig. 3). The first interval I extends from $\omega = 0$ up to the lower boundary of the interval II. The second one encloses the resonance frequency value Eq. (31). Within the interval II the resonant term $n_1 = 2p$ in Eq. (30) after taking the square modulus of expression (30) should be replaced by the expression (37). The third interval III extends from the upper boundary of the interval II up to the lower boundary of the interval IV, the later one enclosing another resonance frequency defined by Eq. (32). Within the interval IV again the resonant term $n_1 = 2p$ in Eq. (30) after taking the square modulus should be replaced by Eq. (41). Finally, the fifth interval V ranges from the upper boundary of the interval IV up to the maximum frequency $\omega_0 = E_{3s} - E_{1s}$. The frequency distribution $dW^{2\gamma}_{3s-1s}$ is symmetric with respect to the point $\omega = \frac{1}{2}\omega_0$ with an accuracy of about 1%. The deviation from the symmetry is due to the difference between $\Gamma_{3s} + \Gamma_{2p}$ and Γ_{2p} in the denominators of Eqs. (37) and (41).

Going over to the fully nonrelativistic approximation and performing the angular integration in the matrix elements in Eq. (30) we obtain the following contributions to the total transition rate $W^{2\gamma}_{3s-1s}$:

$$W^{2\gamma}_{3s-1s} = W^{2\gamma,\text{ cascade}}_{3s-1s} + W^{2\gamma,\text{ pure}}_{3s-1s} + W^{2\gamma,\text{ interference}}_{3s-1s}, \tag{42}$$

$$W^{(\text{cascade})}_{3s;1s} = \frac{4}{27\pi}\frac{\Gamma_{3s}+\Gamma_{2p}}{\Gamma_{2p}}\int_{(\text{II})}\omega^3(\omega_0-\omega)^3\left|\frac{(r)_{3s2p}(r)_{2p1s}}{E_{2p}-E_{3s}+\omega-\frac{i}{2}(\Gamma_{3s}+\Gamma_{2p})}\right|^2 d\omega + \frac{4}{27\pi}\int_{(\text{IV})}\omega^3(\omega_0-\omega)^3\left|\frac{(r)_{3s2p}(r)_{2p1s}}{E_{2p}-E_{1s}-\omega-\frac{i}{2}\Gamma_{2p}}\right|^2 d\omega, \tag{43}$$

$$W^{(\text{pure}2\gamma)}_{3s;1s} = \frac{4}{27\pi}\int_{(\text{II})}\omega^3(\omega_0-\omega)^3\left|S^{(2p)}_{1s;3s}(\omega)+S_{1s;3s}(\omega_0-\omega)\right|^2 d\omega + \frac{4}{27\pi}\int_{(\text{IV})}\omega^3(\omega_0-\omega)^3\left|S_{1s;3s}(\omega) + S^{(2p)}_{1s;3s}(\omega_0-\omega)\right|^2 d\omega + \frac{4}{27\pi}\int_{(\text{I+III+V})}\omega^3(\omega_0-\omega)^3|S_{1s;3s}(\omega)+S_{1s;3s}(\omega_0-\omega)|^2 d\omega, \tag{44}$$

$$dW^{(\text{interference})}_{3s;1s} = \frac{\Gamma_{3s}+\Gamma_{2p}}{\Gamma_{2p}}\int_{(\text{II})}\frac{4\omega^3(\omega_0-\omega)^3}{27\pi}\text{Re}\left[\frac{(r)_{3s2p}(r)_{2p1s}}{E_{2p}-E_{3s}+\omega-\frac{i}{2}(\Gamma_{3s}+\Gamma_{2p})}\right] \tag{45}$$

$$\left[S^{(2p)}_{1s;3s}(\omega)+S_{1s;3s}(\omega_0-\omega)\right]d\omega + \int_{(\text{IV})}\frac{4\omega^3(\omega_0-\omega)^3}{27\pi}\text{Re}\left[\frac{(r)_{3s2p}(r)_{2p1s}}{E_{2p}-E_{1s}-\omega-\frac{i}{2}\Gamma_{2p}}\right]\left[S_{1s;3s}(\omega)+S^{(2p)}_{1s;3s}(\omega_0-\omega)\right]d\omega, \tag{46}$$

$$S_{1s;3s} = \sum_{n'p}\frac{(r)_{3sn'p}(r)_{n'p1s}}{E_{n'p}-E_{3s}+\omega}, \tag{47}$$

$$(r)_{nln'l'} = \int_0^\infty r^3 R_{nl}(r)R_{n'l'}(r)dr. \tag{48}$$

TABLE I. Transition rates (in s^{-1}) for the different decay channels for the decay probability of the $3s$ level with different frequency interval size (l) in length gauge. The last column corresponds to the limiting case where intervals II and IV touch one another with the use of three parameters $l_1 = 4.53 \times 10^6$, $l_2 = 4.58 \times 10^6$, and $l_3 = 10^7$.

l	10^4	10^5	2.5×10^5	5×10^5	10^6	l_1, l_2, l_3
$W_{\mathrm{I}}^{(\mathrm{pure})}$	53.05375194	7.05462259	3.57430739	2.18976651	1.27735735	0
$W_{\mathrm{II}}^{(\mathrm{pure})}$	0.00624713	0.06246845	0.15613353	0.31199777	0.62181876	3.98083823
$W_{\mathrm{III}}^{(\mathrm{pure})}$	95.53585438	7.87779112	2.79274511	1.45164839	1.04561648	0
$W_{\mathrm{IV}}^{(\mathrm{pure})}$	0.00618480	0.06184525	0.15457664	0.30889200	0.61567230	3.95741133
$W_{\mathrm{V}}^{(\mathrm{pure})}$	53.56051962	7.11011114	3.59987543	2.20557518	1.28862529	0
$W^{(\mathrm{pure})}$	202.16255787	22.16683855	10.27763810	6.46787984	4.84909017	7.93824956
$W^{(\mathrm{inter})}$	−0.00910554	−0.09106015	−0.22761672	−0.45498984	−0.90802360	−6.976045531
$W^{(\mathrm{casc})}$	6.31675910×10^6	6.3169401×10^6	6.31695216×10^6	6.31695622×10^6	6.31695830×10^6	6.31696010×10^6
$W^{(\mathrm{total})}$	6.31696125×10^6	6.31696215×10^6	6.31696221×10^6	6.31696223×10^6	6.31696224×10^6	6.31696196×10^6

The notation $S^{(2p)}_{1s;3s}$ means that the state $2p$ is excluded from sum over the $n'p$ in Eq. (47).

In Eqs. (43) to (48) R_{nl} are the radial Schrödinger wave functions for the H atoms and E_{nl} are the corresponding energies. All the probabilities are given in the "length" gauge, in atomic units.

The resonance contributions are concentrated exclusively in the intervals II and IV for the resonances 1 and 2 [see Eqs. (31) and (32)], respectively. Within the other intervals the cascade contributions vanish. Unlike the cascades, the "pure" two-photon contributions are present in all the intervals. The interference contribution is zero within the I, III, and V intervals, i.e., where the cascade contributions are absent.

The results of our calculations are presented in Table I. It is convenient to define the size of the second interval II as $\Delta\omega_{\mathrm{II}} = 2(\Gamma_{3s} + \Gamma_{2p})l$ where l is some number and the size of the fourth interval (IV) as $\Delta\omega_{\mathrm{IV}} = 2\Gamma_{2p}l$, the numbers l ranging from $l = 10^4$ up to $l = 1.5 \times 10^6$. The upper boundary of interval II equals $\omega^{\mathrm{res.1}} + l(\Gamma_{3s} + \Gamma_{2p}) = \frac{5}{72} + l(\Gamma_{3s} + \Gamma_{2p})$ in a.u. while the lower boundary of the interval IV equals $\omega^{\mathrm{res.2}} - l\Gamma_{2p} = \frac{3}{8} - l\Gamma_{2p}$. The different lines in Table I present the contributions of the different intervals to the "pure" two-photon decay rate $W^{2\gamma,\mathrm{pure}}_{3s-1s}$ for different choices of the size of intervals (i.e., the l values). The three last lines present the contributions of the $W^{2\gamma,\mathrm{interference}}_{3s-1s}$, $W^{2\gamma,\mathrm{cascade}}_{3s-1s}$ and the total value $W^{2\gamma}_{3s-1s}$. From Table I we can draw the following conclusion: the "pure" two-photon transition rate depends strongly on the choice of the of the intervals $\Delta\omega_{\mathrm{II}}$, $\Delta\omega_{\mathrm{IV}}$ and cannot be separated out of the total value of $W^{2\gamma}_{2s-1s}$. Only the sum of all contributions $W^{2\gamma}_{3s-1s} = W^{2\gamma,\mathrm{cascade}}_{3s-1s} + W^{2\gamma,\mathrm{pure}}_{3s-1s} + W^{2\gamma,\mathrm{interference}}_{3s-1s}$ can be defined unambiguously, independent of the intervals' choice, i.e., remain invariant. This invariance is clearly seen from comparison of the two last lines in Table I: the cascade contributions changes in the sixth digit with the choice of different l values (at this sixth digit the contributions of the "pure" and "interference" terms becomes significant) while the total value $W^{2\gamma}_{3s-1s}$ remains invariant with the sixth digits. This is an exact proof that was missing in Ref. [33] due to the poorer numerical accuracy. Moreover, in Table II we present the results of the same calculations performed in the "velocity" gauge. Although all the numbers differ quite considerably, the final results for $W^{2\gamma}_{3s-1s}$ again remain invariant with respect

TABLE II. Transition rates (in s^{-1}) for the different decay channels for the decay probability of the $3s$ level with different frequency interval size (l) in velocity gauge. The last column corresponds to the limiting case where intervals II and IV touch one another with the use of three parameters $l_1 = 4.53 \times 10^6$, $l_2 = 4.58 \times 10^6$, and $l_3 = 10^7$.

l	10^4	10^5	2.5×10^5	5×10^5	10^6	l_1, l_2, l_3
$W_{\mathrm{I}}^{(\mathrm{pure})}$	53.05371101	7.05459545	3.57428602	2.18974982	1.27734585	0
$W_{\mathrm{II}}^{(\mathrm{pure})}$	0.00373447	0.03734343	0.09334150	0.18656065	0.37212616	2.45141850
$W_{\mathrm{III}}^{(\mathrm{pure})}$	95.53586269	7.87777276	2.79271674	1.45161319	1.04557595	0
$W_{\mathrm{IV}}^{(\mathrm{pure})}$	0.00369721	0.03697087	0.09241062	0.18470252	0.36843982	2.43568051
$W_{\mathrm{V}}^{(\mathrm{pure})}$	53.56047863	7.11008395	3.59985400	2.20555842	1.28861370	0
$W^{(\mathrm{pure})}$	202.15748401	22.11676645	10.15260889	6.21818460	4.35210148	4.88709901
$W^{(\mathrm{inter})}$	−0.00251068	−0.02510978	−0.06278658	−0.125659184	−0.25201266	−2.71427111
$W^{(\mathrm{casc})}$	6.31675909×10^6	6.31694006×10^6	6.31695212×10^6	6.31695614×10^6	6.31695814×10^6	6.31695986×10^6
$W^{(\mathrm{total})}$	6.31696125×10^6	6.31696215×10^6	6.31696221×10^6	6.31696223×10^6	6.31696224×10^6	6.31696203×10^6

to the choice of the intervals' size and coincide with the value in the "length" gauge with six digits. Our conclusion is that there is no way to define separately the contributions of the "pure" two-photon transition rate for the transitions with cascades.

The accuracy of the numbers in Tables I and II is higher than the contributions of the relativistic and QED corrections to the transition rates, not included in our calculations. Though these corrections will change the value of the $W^{2\gamma}_{3s-1s}$ transition rate (in the fourth digit by the relativistic corrections, in the sixth digit by the QED ones) they will not change our argument for the "inseparability" of the "pure" two-photon contribution from the total $3s \to 1s + 2\gamma$ decay rate.

V. TWO-PHOTON WIDTHS FROM THE IMAGINARY PART OF THE ENERGY LEVEL SHIFT

There is a familiar statement that the total width Γ of the excited atomic level equals the sum of the transition rates to all the lower levels. Since all the transitions end up finally at the ground state we could write

$$\Gamma^{\rm total}_A = \sum_i W^i_{A\to 0}, \tag{49}$$

where index A denotes the arbitrary excited state of an atom, 0 denotes the ground state, and the summation over i corresponds to all possible decay channels, including cascades. From the other side, the width of the level can be defined via imaginary part of the level shift ΔE_A which is due to the radiative corrections and interelectron corrections

$$\Gamma_A = -2{\rm Im}\Delta E_A. \tag{50}$$

As it was argued in the Introduction, these two definitions do not coincide with the cascade contributions taken into account. The cascades do not contribute to Eq. (50). The proof of this statement is the aim of the present section.

The contributions of the cascades to the $\Gamma^{\rm total}_A$ become singular due to the energy denominators which tend to zero as was demonstrated in Secs. II and III. The regularizations of these singularities require the insertion of the energy widths in the energy denominators and these widths are defined exactly by Eq. (50).

In the case of the one-photon transitions there are no cascades and Eq. (2) for $\Gamma^{1\gamma}_n$ takes place. For the one-electron atom the closed fully relativistic expression for $\Gamma^{1\gamma}_n$ can be derived (see [31,39])

$$\Gamma^{1\gamma}_n = \sum_{n'<n}\Gamma^{1\gamma}_{nn'} = -\frac{\alpha}{2}\sum_{n'<n}\left(1-\frac{E_{n'}}{|E_{n'}|}\right) \times\left(\frac{1-\vec{\alpha}_1\vec{\alpha}_2}{r_{12}}\sin(|E_{n'}-E_n|r_{12})\right)_{n'nnn'}, \tag{51}$$

where $\vec{\alpha}_i$ $(i = 1, 2)$ are the Dirac matrices acting on the Dirac atomic wave functions depending on the variables $\vec{r}_1$ and $\vec{r}_2$, respectively. The summation in Eq. (51) is extended over all positive energy levels lower than n: $E_{n'} < E_n$. In Eq. (51) the integration over the emitted photon directions and the summation over the photon polarization are automatically included.

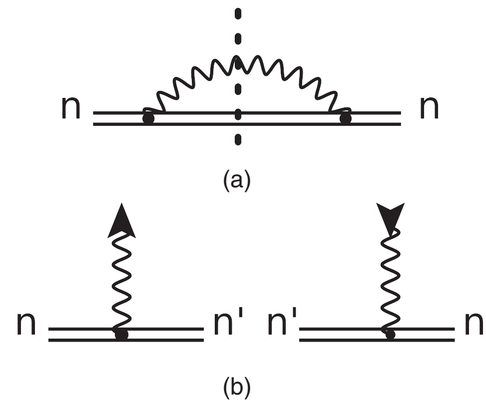

FIG. 4. The Feynman one-loop graph corresponding to the one-photon width $\Gamma^{1\gamma}_n$ [see Eq. (51)]. The double solid line denotes the electron in the field of the nucleus (Furry picture), the wavy lines denote (a) the virtual or (b) real photons. The dashed vertical line in (a) indicates the cut of the graph. This cut corresponds to the emission of the photon and the right graph corresponds to the absorbed photon.

The expression Eq. (51) arises as an imaginary part of the lowest-order radiative electron self-energy correction. The Feynman graph corresponding to this correction is depicted in Fig. 4. There is a simple rule which helps to understand to which transition corresponds the imaginary part of one or another Feynman graph for radiative corrections. This rule consists of cutting the graphs, namely the electron self-energy loops as it is demonstrated in Fig. 4(a) for the lowest-order electron self-energy correction. One should not cut the vacuum loops since the vacuum polarization corrections do not contribute to the imaginary part of the energy correction. The graph in Fig. 4(b) illustrates that the imaginary part of the graph in Fig. 4(a) corresponds to the sum of the one-photon transitions to the lower levels. A justification of this rule can be seen from the derivations below in this section.

The second-order electron self-energy corrections are depicted in Figs. 5–7. The Feynman graph Fig. 5(a) is reducible, i.e., this graph can be divided into two parts by cutting only the internal electron line. The graphs in Figs. 6(a) and 7(a) are irreducible in this sense. As it was proved in Refs. [31,39], corrections to the energy ΔE_A corresponding to any irreducible graphs can be obtained via relations

$$\langle A'|\widehat{S}|A\rangle = 2\pi i\delta(E_{A'}-E_A)\langle A'|\widehat{U}|A\rangle, \tag{52}$$

$$\Delta E_A = \langle A|\widehat{U}|A\rangle, \tag{53}$$

where $\langle A'|\widehat{U}|A\rangle$ is the matrix element of the amplitude. Figure 5(b) demonstrates that the graph in Fig. 5(a) contributes only to the electron self-energy (SE) correction to the one-photon amplitude and does not contribute to the two-photon width $\Gamma^{2\gamma}_n$. Radiative corrections to the one-photon transition

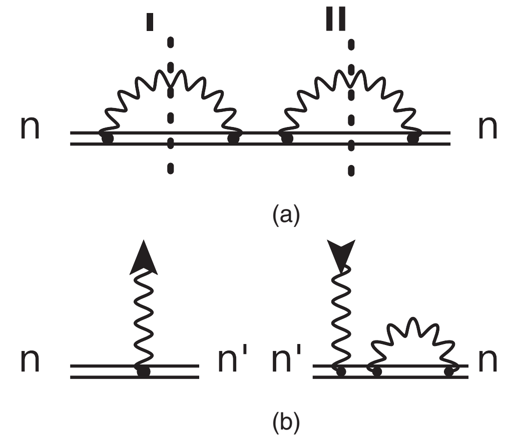

FIG. 5. The two-loop graph (loop after loop). Notations are the same as in Fig. 4. The two cuts I and II in (a) correspond to the products of the amplitudes corresponding for the Feynman graphs in (b) as depicted for cut I. A similar picture results for cut II. These pictures demonstrate that the graph in panel (a) contributes only to the electron self-energy (SE) correction to the one-photon amplitudes and does not contribute to the two-photon width $\Gamma_n^{2\gamma}$.

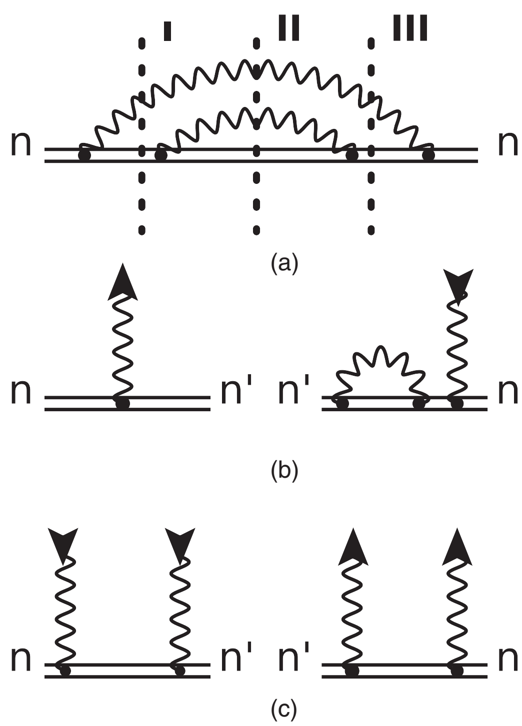

FIG. 6. The two-loop Feynman graph (loop inside loop). Notations are the same as in Figs. 4 and 5. The three cuts I, II, II in (a) correspond to the products of amplitudes as depicted for the cut I in (b) and for the cut II in (c). A picture for the cut III is similar to that in (b). These pictures demonstrate that only the cut II corresponds to the contribution to the two-photon width $\Gamma_n^{2\gamma}$.

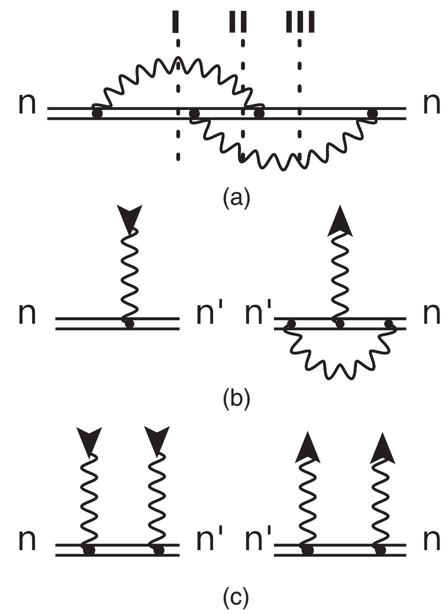

FIG. 7. The two-loop graph (crossed loops). Notations are the same as in Fig. 4. The three cuts in (a) correspond to the products of the Feynman graphs as depicted for the cut I in (b) and for the cut II in (c). A picture for the cut III is similar to that in (b). These pictures demonstrate that the cuts I and II contribute to the vertex corrections to the one-photon amplitudes and only the cut II contributes to the two-photon width $\Gamma_n^{2\gamma}$.

rates via imaginary parts of the radiative corrections to the energy levels were evaluated in Refs. [40–42]. Here we will not be interested in these corrections and will evaluate exclusively the two-photon decay widths. Therefore we will concentrate on the graphs in Figs. 6 and 7.

The fourth-order S-matrix elements which correspond to Fig. 6(a) "loop inside loop" (lil) and Fig. 7(a) "crossed loops" (cl) graphs are

$$\langle A|\widehat{S}^{(4)\mathrm{lil}}|A\rangle = e^4\int d^4x_1d^4x_2d^4x_3d^4x_4\big(\overline{\psi}_A(x_1)\gamma_{\mu_1}S(x_1x_2)\gamma_{\mu_2} \times S(x_2x_3)\gamma_{\mu_3}S(x_3x_4)\gamma_{\mu_4}\psi_A(x_4)\big) \times D_{\mu_1\mu_4}(x_1x_4)D_{\mu_2\mu_3}(x_2x_3), \tag{54}$$

$$\langle A|\widehat{S}^{(4)\mathrm{cl}}|A\rangle = e^4\int d^4x_1d^4x_2d^4x_3d^4x_4\big(\overline{\psi}_A(x_1)\gamma_{\mu_1}S(x_1x_2)\gamma_{\mu_2} \times S(x_2x_3)\gamma_{\mu_3}\,S(x_3x_4)\gamma_{\mu_4}\psi_A(x_4)\big) \times D_{\mu_1\mu_3}(x_1x_3)D_{\mu_2\mu_4}(x_2x_4). \tag{55}$$

Here the notations are the same as in Sec. II and $D_{\mu\nu}(x_1x_2)$ denotes the photon propagator in the Feynman gauge

$$D_{\mu\nu}(x_1x_2) = \frac{\delta_{\mu\nu}}{2\pi i r_{12}}\int_{-\infty}^{\infty} e^{i\omega(t_1-t_2)+i|\omega|r_{12}}d\omega. \tag{56}$$

Inserting expressions (10), (12), and (56) in Eqs. (54) and (55) for the Dirac wave functions, electron propagators, and photon propagators, respectively, integrating over time and frequency variables, and using Eqs. (52) and (53) we find

$$U_A^{(4)\mathrm{lil}} = e^4 \sum_{nmk} \left(\frac{\delta_{\mu_1\mu_4}}{r_{14}} \frac{\delta_{\mu_2\mu_3}}{r_{23}} I^{\mathrm{lil}}_{nmkA}(r_{14}r_{23}) \right)_{Anmk}, \tag{57}$$

$$I^{\mathrm{lil}}_{nmkA}(r_{14}r_{23}) = \left(\frac{1}{2\pi i}\right)^2 \int_{-\infty}^{+\infty} d\omega_1 \int_{-\infty}^{+\infty} d\omega_3 \frac{e^{i|\omega_1|r_{14}} e^{i|\omega_3|r_{23}}}{[E_A - \omega_1 - E_n(1-i0)][E_A - \omega_1 - \omega_3 - E_k(1-i0)][E_A - \omega_1 - E_m(1-i0)]}, \tag{58}$$

$$U_A^{(4)\mathrm{cl}} = e^4 \sum_{nmk} \left(\frac{\delta_{\mu_1\mu_3}}{r_{13}} \frac{\delta_{\mu_2\mu_4}}{r_{24}} I^{cl}_{nmkA}(r_{13}r_{24}) \right)_{Anmk}, \tag{59}$$

$$I^{\mathrm{cl}}_{nmkA}(r_{13}r_{24}) = \left(\frac{1}{2\pi i}\right)^2 \int_{-\infty}^{+\infty} d\omega_1 \int_{-\infty}^{+\infty} d\omega_3 \frac{e^{i|\omega_1|r_{13}} e^{i|\omega_3|r_{24}}}{[E_A - \omega_1 - E_n(1-i0)][E_A - \omega_1 - \omega_3 - E_k(1-i0)][E_A - \omega_3 - E_m(1-i0)]}. \tag{60}$$

According to the analysis given in the beginning of this section the two-photon width is connected exclusively with the poles generated by the central energy denominators in Eqs. (58) and (60). For further evaluation we employ the following representation of the integrals, containing $e^{i|\omega|r}$:

$$\mathrm{Re} \int_{-\infty}^{\infty} \frac{e^{i|\omega|r}}{E_A - E_s(1-i0) - \omega} = -\frac{\pi}{2}\left(1 + \frac{E_s}{|E_s|}\right)\left(1 + \frac{\beta_{As}}{|\beta_{As}|}\right)\sin(\beta_{As} r), \tag{61}$$

where $\beta_{As} = E_A - E_s$. Performing the integration over ω_3 with the help of Eq. (61) we find that the two-photon contributions to Eqs. (58) and (60) reduce to

$$I^{\mathrm{lil}.2\gamma}_{nmkA}(r_{14},r_{23}) = \frac{e^4}{2\pi}\left(1 + \frac{E_k}{|E_k|}\right) \int_0^{\beta_{Ak}} d\omega_1 \frac{\sin(\omega_1 r_{23}) e^{i(\beta_{Ak} - \omega_1) r_{14}}}{[E_A + \omega_1 - E_n(1-i0)][E_A - \omega_1 - E_m(1-i0)]}, \tag{62}$$

$$I^{cl.2\gamma}_{nmkA}(r_{13},r_{24}) = \frac{e^4}{2\pi}\left(1 + \frac{E_k}{|E_k|}\right) \int_0^{\beta_{Ak}} d\omega_1 \frac{\sin(\omega_1 r_{23}) e^{i(\beta_{Ak} - \omega_1) r_{14}}}{[E_A + \omega_1 - E_n(1-i0)][E_A - \omega_1 - E_m(1-i0)]}, \tag{63}$$

where $E_A > E_{n(m)} > E_k$.

Collecting all the contributions together, using the definition Eq. (50) and introducing explicitly the Feynman parameters $i0 \to i\varepsilon$ we get

$$\Gamma_A^{2\gamma} = \frac{e^4}{2\pi} \lim_{\varepsilon \to 0} \mathrm{Re} \left(\sum_{\substack{nkm \\ E_A > E_{n(m)} \\ > E_k}} \int_0^{\beta_{Ak}} \left(1 + \frac{E_k}{|E_k|}\right) \left\{ \frac{\left(\frac{(1-\vec{\alpha}_2\vec{\alpha}_4)\sin(\omega r_{24})}{r_{24}}\right)_{nmkA} \left(\frac{(1-\vec{\alpha}_1\vec{\alpha}_3)\sin[(\beta_{Ak}-\omega)r_{13}]}{r_{13}}\right)_{Akmn}}{[(E_k - E_n)(1-i\varepsilon) + \omega][E_A - \omega - E_m(1-i\varepsilon)]} + \frac{\left(\frac{(1-\vec{\alpha}_2\vec{\alpha}_3)\sin(\omega r_{23})}{r_{23}}\right)_{AmnA} \left(\frac{(1-\vec{\alpha}_1\vec{\alpha}_4)\sin[(\beta_{Ak}-\omega)r_{14}]}{r_{14}}\right)_{nkkm}}{[E_A - \omega - E_n(1-i\varepsilon)][E_A - \omega - E_m(1-i\varepsilon)]} \right\} d\omega \right). \tag{64}$$

The matrix elements in Eq. (64) $[F(12)]_{abcd}$ should be understood as $[F(12)]_{a(1)b(2)c(1)d(2)}$ where 1, 2 denote the variables and $\vec{\alpha}_i$ are the Dirac matrices acting on the corresponding wave functions $\psi(i)$. The first term in curly brackets in Eq. (64) corresponds to the contribution of the loop inside loop Feynman graph and the second term corresponds to the crossed loops Feynman graph.

To express Eq. (64) as the sum of the partial widths for the two-photon transitions $A \to k + 2\gamma$ we may use an equality [39]

$$\beta_{Ak} \sum_{\vec{e}} \int d\vec{\nu} |[(\vec{e}\vec{\alpha}) e^{-i\beta_{nA}(\vec{\nu}\vec{r})}]_{kA}|^2 = -\pi \left(\frac{1 - \vec{\alpha}_1\vec{\alpha}_2}{r_{12}} \sin(\beta_{Ak} r_{12}) \right)_{AkkA}. \tag{65}$$

Then

$$\Gamma_A^{2\gamma} = \sum_{E_A > E_{n(m)} > E_k} \Gamma_{Ak}^{2\gamma}, \tag{66}$$

$$\Gamma_{Ak}^{2\gamma} = e^4 \lim_{\varepsilon\to 0} \mathrm{Re} \int_0^{\beta_{Ak}} \left(\frac{\omega(\beta_{Ak}-\omega)}{2^4\pi^3} \int d\vec{\nu} d\vec{\nu}' \sum_{\vec{e}\,\vec{e}\,'} \sum_n \left\{ \frac{[(\vec{e}\,^*\vec{\alpha})e^{-i\vec{k}\vec{r}}]_{kn}[(\vec{e}\,'^*\vec{\alpha})e^{-i\vec{k}'\vec{r}}]_{nA}}{E_A-\omega-E_n(1-i\varepsilon)} + \frac{[(\vec{e}\,'^*\vec{\alpha})e^{-i\vec{k}'\vec{r}}]_{kn}[(\vec{e}\,^*\vec{\alpha})e^{-i\vec{k}\vec{r}}]_{nA}}{(E_k-E_n)(1-i\varepsilon)+\omega} \right\} \right.$$
$$\left. \times \sum_m \left\{ \frac{[(\vec{e}\,^*\vec{\alpha})e^{-i\vec{k}\vec{r}}]^*_{km}[(\vec{e}\,'^*\vec{\alpha})e^{-i\vec{k}'\vec{r}}]^*_{mA}}{E_A-\omega-E_m(1-i\varepsilon)} + \frac{[(\vec{e}\,'^*\vec{\alpha})e^{-i\vec{k}'\vec{r}}]^*_{km}[(\vec{e}\,^*\vec{\alpha})e^{-i\vec{k}\vec{r}}]^*_{mA}}{(E_k-E_m)(1-i\varepsilon)+\omega} \right\} \right) d\omega. \tag{67}$$

Note that in Eq. (67) there is no square modulus. Therefore we can employ the method suggested by the authors of [35–37] for avoiding the double pole singularities in the integration over ω in Eq. (64) when $n(m) = A, k$. According to the prescription given in Refs. [35–37] these singular terms should be regularized with the use of equality

$$\lim_{\varepsilon\to 0} \mathrm{Re} \int_0^1 d\omega \left(\frac{1}{a-\omega+i\varepsilon} \right)^2 = \frac{1}{a(a-1)}. \tag{68}$$

The regularization Eq. (68) holds with two special assumptions.

(1) We have to keep the Feynman parameters $i\varepsilon$ in both energy denominators in Eq. (64) and below as equal to each other.

(2) In Eq. (68) we have first to integrate over ω and only then evaluate the limit $\varepsilon \to 0$.

Both assumptions are not evident, but in the next section we will present a justification for them.

In the nonrelativistic limit in the "length" form Eq. (64) after integration over the photon emission directions and summation over the photon polarizations reduces to

$$\Gamma_{Ak}^{2\gamma} = e^4 \lim_{\varepsilon\to 0} \mathrm{Re} \int_0^{\beta_{Ak}} \frac{\omega^3(\beta_{Ak}-\omega)^3}{2^4\pi^3} \sum_n \times \left\{ \frac{(\vec{r})_{kn}(\vec{r})_{nA}}{E_A-\omega-E_n(1-i\varepsilon)} + \frac{(\vec{r})_{kn}(\vec{r})_{nA}}{(E_k-E_n)(1-i\varepsilon)+\omega} \right\} \times \sum_m \left\{ \frac{(\vec{r})^*_{km}(\vec{r})^*_{mA}}{E_A-\omega-E_m(1-i\varepsilon)} + \frac{(\vec{r})^*_{km}(\vec{r})^*_{mA}}{(E_k-E_m)(1-i\varepsilon)+\omega} \right\} d\omega, \tag{69}$$

where $(\ldots)_{ab}$ denote the matrix elements with the Schrödinger wave functions ψ_a^*, ψ_b. The double pole contribution in Eq. (69) should be understood according to Eq. (68). Following [35–37] we performed also the numerical calculations of the widths $\Gamma_{Ak}^{2\gamma}$ with indices A,k running through the set of nl states: for A $n(m) = 2,\ldots,5, l = 0,1,2,3$, for k $n(m) = 1,\ldots,4, l = 0,1,2$. The results are summarized in Table III. We should stress once more, that the widths $\Gamma_{Ak}^{2\gamma}$ cannot be considered as the "pure" two-photon transition rates, but present the two-photon contributions to the total level width. This statement is supported by the circumstance that some quantities $\Gamma_{Ak}^{2\gamma}$ appear to be negative (see Table III). This prevents the treatment of $\Gamma_{Ak}^{2\gamma}$ as transition rates. Our calculations in Table III were performed in three different ways: (1) direct summation over the entire H spectrum in Eq. (69) using the "length" form of the matrix elements; (2) the same with the "velocity" form of the matrix elements; and (3) the use of the Coulomb Green's function. All the results coincide with three to six digits.

VI. APPLICATION OF THE ADIABATIC *S*-MATRIX THEORY

In the Sec. V Eqs. (52) and (53) for the energy level shift were applied for the evaluation of the two-photon width $\Gamma_A^{2\gamma}$ of the energy level A as an imaginary part of the total energy shift ΔE_A. As was already mentioned in Sec. V, the applicability of Eqs. (52) and (53) was proven for the corrections described by the irreducible Feynman graphs in Refs. [31,39]. This proof was based on the adiabatic *S*-matrix theory, i.e., on the Gell-Mann and Low formula [43] modified by Sucher [44], and on an assumption that the energy denominators in the *S*-matrix elements corresponding to the irreducible Feynman graphs never turn to zero. The imaginary parts of the irreducible Feynman graphs in Figs. 6 and 7, wherefrom we extract the two-photon width contribution, contain singular denominators

TABLE III. The widths $\Gamma_{Ak}^{2\gamma}$ for the hydrogen atom (in s^{-1}). With the (∗) are denoted the values calculated earlier in Refs. [35–37].

state		n'			
nl	$n'l$	1	2	3	4
2s	n's	8.229355 8.229352 ∗	–	–	–
3s	n's	2.083086 2.082853 ∗	0.064531	–	–
3d	n 's	1.042708 1.042896 ∗	0.000776	–	–
4s	n's	0.699717 0.698897 ∗	0.016843	0.002925	–
4s	n'd	–	–	9.69×10^{-6}	–
4p	n'p	–	0.015623	0.002503	–
4d	n's	0.598406 0.598798 ∗	−0.007319	0.000030	–
4d	n'd	–	–	0.001685	–
4f	n'p	–	0.031754	0.000044	
5s	n's	0.288117 0.287110 ∗	0.081741	0.000704	0.000298
5s	n'd	–	–	−0.000028	1.82×10^{-6}

as was shown in Sec. V. Though this singularity can be avoided with the help of the formula (68), we would like to support the derivations made in Sec. V, by another approach which does not refer to Eqs. (52) and (53) and is based directly on the Gell-Mann-Low-Sucher adiabatic S matrix.

The adiabatic S-matrix approach was applied by the authors of [33] for the description of the one- and two-photon transitions with cascades. A general approach and the one-photon part of the studies in Ref. [33] are fully adequate, but the two-photon part including the cascade contribution requires revision. The Gell-Mann-Low-Sucher formula for the energy shift ΔE_A for an electron in the one-electron atom or ion looks like

$$\Delta E_A = \lim_{\eta\to 0}\frac{1}{2}i\eta\frac{e\frac{\partial}{\partial e}\langle A|\widehat{S}_\eta|A\rangle}{\langle A|\widehat{S}_\eta|A\rangle}, \tag{70}$$

where e is the electron charge (absolute value). The adiabatic S matrix S_η differs from the ordinary S matrix by the presence of the adiabatic exponential factor $e^{-\eta|t|}$ in the every vertex. The level shift is considered with respect to the zero-order Dirac energy of the level E_A^0. The shift may be caused by the radiative corrections or by the interelectron interaction corrections in a few-electron atom. An imaginary part of the energy shift due to the radiative corrections corresponds to the radiative widths of the levels, an imaginary part of the second-order interelectron interaction corrections corresponds to the Auge width [39]. The use of Eq. (70) helps to avoid the assumption (1) made in the previous section.

The first evaluation of the first- and second-order energy shift due to the Coulomb interelectron interaction was given in Ref. [45], the first evaluation of the imaginary part of ΔE_A (radiative correction to the one-photon decay of the excited state in one-electron atom) was made in Ref. [40].

For a one-electron atom (ion) in the state $|A\rangle$ interacting with the photon vacuum a complex energy correction ΔE_A contains only the diagonal S-matrix elements of even order. The expansion of Eq. (67) up to the fourth order in e reads [45]

$$\Delta E_A = \lim_{\eta\to 0} i\eta\big[\langle A|\widehat{S}^{(2)}_\eta|A\rangle + 2\langle A|\widehat{S}^{(4)}_\eta|A\rangle - \langle A|\widehat{S}^{(2)}_\eta|A\rangle^2\big]. \tag{71}$$

Separating the real and imaginary parts of the matrix element $\langle A|\widehat{S}^{(i)}_\eta|A\rangle$ and using Eqs. (50) and (71) results [33]

$$\Gamma_A = -\lim_{\eta\to 0}\eta\big[\mathrm{Re}\langle A|\widehat{S}^{(2)}_\eta|A\rangle + 2\mathrm{Re}\langle A|\widehat{S}^{(4)}_\eta|A\rangle + \big|\langle A|\widehat{S}^{(2)}_\eta|A\rangle\big|^2 - 2\big(\mathrm{Re}\langle A|\widehat{S}^{(2)}_\eta|A\rangle\big)^2\big]. \tag{72}$$

Equation (72) is valid up to the e^4 terms inclusively.

For further analysis in Ref. [33] using the optical theorem was suggested. This theorem is a consequence of the unitarity of the S matrix and in the general way can be formulated via the introduction of the T matrix

$$\widehat{S} = 1 + i\widehat{T}. \tag{73}$$

Then the optical theorem reads

$$i(\widehat{T} - \widehat{T}^+) = -\widehat{T}^+\widehat{T} = -\widehat{T}\widehat{T}^+. \tag{74}$$

In terms of the matrix elements Eq. (74) looks like

$$2\mathrm{Im}\langle I|\widehat{T}|I\rangle = \sum_F |\langle F|\widehat{T}|I\rangle|^2, \tag{75}$$

where the wave function $|I\rangle$ corresponds to the initial excited state of an atomic electron and the photon vacuum and the summation in Eq. (75) runs over the states $|F\rangle$, corresponding to the final (not necessarily ground) state of atomic electron plus a number (one or two in our case) of photons. Using the optical theorem in the form Eq. (75) and the relation between the matrix elements

$$\mathrm{Re}\langle I|\widehat{S}^{(i)}|I\rangle = -\mathrm{Im}\langle I|\widehat{T}^{(i)}|I\rangle, \tag{76}$$

where $i = 1,2,\ldots$, the following equality can be derived [33]

$$-2\mathrm{Re}\langle I|\widehat{S}^{(2i)}|I\rangle = \sum_F |\langle F|\widehat{S}^{(i)}|I\rangle|^2 + \sum_F\sum_{j<i} 2\mathrm{Re}\langle I|\times\widehat{S}^{(2j)}|F\rangle\langle F|\widehat{S}^{(2i-j)}|I\rangle. \tag{77}$$

For the fourth-order matrix elements of our interest Eq. (77) results

$$-2\mathrm{Re}\langle I|\widehat{S}^{(4)}|I\rangle = \sum_F |\langle F|\widehat{S}^{(2)}|I\rangle|^2 + \sum_F\sum_{j<i} 2\mathrm{Re}\langle I|\widehat{S}^{(1)}|F\rangle\langle F|\widehat{S}^{(3)}|I\rangle. \tag{78}$$

The last term in Eq. (78) represents evidently the radiative correction to the one-photon width evaluated in Ref. [40] and will not be discussed here anymore.

As it was argued in Ref. [33], it is also possible to apply the optical theorem to the adiabatic S matrix. Then from Eq. (72) it follows for $I = A,0_\gamma$ (excited state, no photons) an expression for the two-photon width

$$\Gamma_A^{2\gamma} = \lim_{\eta\to 0}\eta\left\{2\sum_{F\neq A,0_\gamma}\big|\langle F|\widehat{S}^{(2)}_\eta|A,0_\gamma\rangle\big|^2 + 4\big(\mathrm{Re}\langle A,0_\gamma|\widehat{S}^{(2)}_\eta|A,0_\gamma\rangle\big)^2\right\}. \tag{79}$$

It should be noted that the state $F = A,2_\gamma$ formally is present in the sum over F in Eq. (79). So we can rewrite Eq. (79) finally in the form

$$\Gamma_A^{2\gamma} = \lim_{\eta\to 0}\eta\left\{2\sum_{\substack{F\neq A,0_\gamma\\ F\neq A,2_\gamma}}\big|\langle F|\widehat{S}^{(2)}_\eta|A,0_\gamma\rangle\big|^2 + 2\big|\langle A,2_\gamma|\widehat{S}^{(2)}_\eta|A,0_\gamma\rangle\big|^2 + 4\big(\mathrm{Re}\langle A,0_\gamma|\widehat{S}^{(2)}_\eta|A,0_\gamma\rangle\big)^2\right\}. \tag{80}$$

The middle term in Eq. (80) corresponds to the apparently nonphysical transition $A \to A + 2\gamma$ as canceled by the last term as it was shown in Ref. [33]. The summation over F in Eq. (79) assumes also an integration over the wave vectors of both the emitted photons and summation over the photon polarizations.

Note that our analysis of Eq. (80) differs from that in Ref. [33]. Below we will demonstrate that this analysis leads to the same conclusions as in Sec. V.

The substitution of the corresponding matrix elements in Eq. (80) and integration over photon frequency leads to

$$\Gamma_A^{2\gamma} = \sum_F \Gamma_{AF}^{2\gamma} = \lim_{\eta\to 0}\frac{e^4}{2^4\pi^3}\sum_F \left\{\sum_{e\,e'}\int d\nu d\nu' \int d\omega\, \omega(\omega_0-\omega) \times \sum_n \left|\frac{[(\vec{e}'^{\,*}\vec{\alpha})e^{-i\vec{k}'\vec{r}}]_{Fn}[(\vec{e}^{\,*}\vec{\alpha})e^{-i\vec{k}\vec{r}}]_{nA}}{E_n - E_A + \omega + i\eta} + \frac{[(\vec{e}^{\,*}\vec{\alpha})e^{-i\vec{k}\vec{r}}]_{Fn}[(\vec{e}'^{\,*}\vec{\alpha})e^{-i\vec{k}'\vec{r}}]_{nA}}{E_n - E_F - \omega + i\eta}\right|^2\right\}. \tag{81}$$

To demonstrate that Eqs. (81) and (66) coincide with each other in the presence of cascades (in the absence of cascades it is obvious since both amplitudes are pure real) it is convenient to consider transition $3s \to 1s + 2\gamma$ ($A = 3s$, $F = 1s$) as an example:

$$\Gamma_{3s,1s}^{2\gamma} = \lim_{\eta\to 0}\frac{e^4}{2^4\pi^3}\sum_{e\,e'}\int d\nu d\nu' \int d\omega\, \omega(\omega_0-\omega)\sum_n \left|\frac{[(\vec{e}'^{\,*}\vec{\alpha})e^{-i\vec{k}'\vec{r}}]_{1sn}[(\vec{e}^{\,*}\vec{\alpha})e^{-i\vec{k}\vec{r}}]_{n3s}}{E_n - E_{3s} + \omega + i\eta} + \frac{[(\vec{e}^{\,*}\vec{\alpha})e^{-i\vec{k}\vec{r}}]_{1sn}[(\vec{e}'^{\,*}\vec{\alpha})e^{-i\vec{k}'\vec{r}}]_{n3s}}{E_n - E_{1s} - \omega + i\eta}\right|^2. \tag{82}$$

The resonant terms when $n = 2p$ in Eq. (82) are

$$\Gamma_{3s-2p-1s}^{2\gamma,\text{res}.1} = \lim_{\eta\to 0}\frac{e^4}{2^4\pi^3}\sum_{e\,e'}\int d\nu d\nu' \int d\omega\, \omega(\omega_0-\omega)|[(\vec{e}'^{\,*}\vec{\alpha})e^{-i\vec{k}'\vec{r}}]_{1s2p}[(\vec{e}^{\,*}\vec{\alpha})e^{-i\vec{k}\vec{r}}]_{2p3s}|^2\left|\frac{1}{E_{2p} - E_{3s} + \omega + i\eta}\right|^2 \tag{83}$$

and

$$\Gamma_{3s-2p-1s}^{2\gamma,\text{res}.2} = \lim_{\eta\to 0}\frac{e^4}{2^4\pi^3}\sum_{e\,e'}\int d\nu d\nu' \int d\omega\, \omega(\omega_0-\omega)|[(\vec{e}'^{\,*}\vec{\alpha})e^{-i\vec{k}'\vec{r}}]_{1s2p}[(\vec{e}^{\,*}\vec{\alpha})e^{-i\vec{k}\vec{r}}]_{2p3s}|^2\left|\frac{1}{E_{2p} - E_{1s} - \omega + i\eta}\right|^2. \tag{84}$$

It is important to note that parameter η in both Eqs. (83) and (84) is the same. The limit $\eta \to 0$ in the adiabatic theory should be evaluated after all the integrations are performed. This justifies the assumptions made in the previous section.

The equality

$$\lim_{\varepsilon\to 0}\int_0^1 d\omega\left|\frac{1}{a-\omega+i\varepsilon}\right|^2 = \frac{\pi}{\varepsilon} + \frac{1}{a(a-1)} + O(\varepsilon^2) \tag{85}$$

employed in Ref. [35] being applied to Eq. (84) yields the positive contribution $+\frac{\pi}{\eta}$ while for Eq. (83) it gives negative contribution $-\frac{\pi}{\eta}$. So these singular terms cancel each other in Eq. (82) after using Eq. (85). Having in mind Eq. (68), we can write the following chain of equalities for two resonant terms in Eq. (82):

$$\lim_{\eta\to 0}\int_0^1 d\omega\left|\frac{1}{a-\omega+i\eta}\right|^2 + \lim_{\eta\to 0}\int_0^1 d\omega\left|\frac{1}{a'-\omega-i\eta}\right|^2 = \frac{\pi}{\eta} + \frac{1}{a(a-1)} - \frac{\pi}{\eta} + \frac{1}{a'(a'-1)} + O(\eta^2)$$
$$= \lim_{\eta\to 0}\text{Re}\int_0^1 d\omega\left(\frac{1}{a-\omega+i\eta}\right)^2 + \lim_{\eta\to 0}\text{Re}\int_0^1 d\omega\left(\frac{1}{a'-\omega+i\eta}\right)^2. \tag{86}$$

Substituting Eq. (87) in Eq. (82) finally we have

$$\Gamma_{3s,1s}^{2\gamma} = e^4 \lim_{\eta\to 0}\text{Re}\int_0^{\omega_0}\left(\frac{\omega(\omega_0-\omega)}{2^4\pi^3}\int d\vec{\nu}d\vec{\nu}\,'\sum_{\vec{e}\,\vec{e}\,'}\sum_n\left\{\frac{[(\vec{e}\,'^{\,*}\vec{\alpha})e^{-i\vec{k}\,'\vec{r}}]_{1sn}[(\vec{e}^{\,*}\vec{\alpha})e^{-i\vec{k}\vec{r}}]_{n3s}}{E_n - E_{3s} + \omega + i\eta} + \frac{[(\vec{e}^{\,*}\vec{\alpha})e^{-i\vec{k}\vec{r}}]_{1sn}[(\vec{e}\,'^{\,*}\vec{\alpha})e^{-i\vec{k}\,'\vec{r}}]_{n3s}}{E_n - E_{1s} - \omega + i\eta}\right\} \times \sum_m\left\{\frac{[(\vec{e}^{\,*}\vec{\alpha})e^{-i\vec{k}\vec{r}}]_{1sm}^*[(\vec{e}\,'^{\,*}\vec{\alpha})e^{-i\vec{k}\,'\vec{r}}]_{m3s}^*}{E_m - E_{3s} + \omega + i\eta} + \frac{[(\vec{e}\,'^{\,*}\vec{\alpha})e^{-i\vec{k}\,'\vec{r}}]_{1sm}^*[(\vec{e}^{\,*}\vec{\alpha})e^{-i\vec{k}\vec{r}}]_{m3s}^*}{E_m - E_{1s} - \omega + i\eta}\right\}\right)d\omega. \tag{87}$$

The final equation for $\Gamma_{3s,1s}^{2\gamma}$ fully coincides with Eq. (67) for $A = 3s$, $k = 1s$.

The approach described above can be applied also for the evaluation of transition rate $W_{3s,1s}^{2\gamma}$, however, in this case the singular terms in Eq. (87) will not cancel each other. Actually each electron propagator in the two-photon decay rate amplitude has a different infinitesimal parameter ε and the limits with respect to these parameters should be taken independently.

Thus the singularity remains in the cascade terms in the two-photon transition rate and should be regularized as it was described in Sec. III.

VII. CONCLUSION

To summarize the results of our present studies we should mention first that we believe that we have presented a convincing demonstration of the inseparability of the contributions of cascades and "pure" two-photon transitions in the total two-photon transition rates. We have shown with a high numerical accuracy of calculations that, while the "pure" two-photon and interference contributions can vary essentially with the choice of the method of calculation, the total two-photon transition rates remains strongly invariant. For the regularization of the cascade (singular) terms in the expressions for the two-photon decay rates we employed the QED procedure suggested by Low [32]. This procedure in an unambiguous way introduces the level width Γ_A in the "dangerous" energy denominators. These widths are the imaginary parts of the corresponding radiative level shifts. The dominant contributions to these widths come from the one-photon width $\Gamma_A^{1\gamma}$, which can be presented as a sum of the one-photon transition rates to the lower levels [Eq. (2)]. However, in principle Γ_A should contain also a two-photon part $\Gamma_A^{2\gamma}$. If $\Gamma_A^{2\gamma}$ would also coincide with the sum of the total two-photon decay rates (including cascades), the perturbation theory for the imaginary part of the radiative energy level shift would not exist: cascade contributions are always of the same order (parametrically) as the one-photon widths. Our analysis shows that it does not happen. An explicit evaluation of the two-photon contributions to the imaginary part of the radiative level shift shows that the cascades do not contribute to this imaginary part. The "dangerous" denominators do not require regularization and reduce to the finite terms, so that the two-photon width $\Gamma_A^{2\gamma}$ does not violate the perturbation theory [see Eq. (7)]. The most important (and new) statement made in the present work is expressed by an inequality (6): the two-photon transition rate in the presence of cascades does not coincide with the two-photon width evaluated as an imaginary part of the radiative level shift. The two-photon transition rate $W^{2\gamma}$ contains singularity corresponding to the cascades and requires regularization. The two-photon width $\Gamma_A^{2\gamma}$ is finite and presents a small correction to the one-photon width $\Gamma_A^{1\gamma}$. For the one-electron ions with only $E1$ photons taken into account $\Gamma_A^{1\gamma} \sim m\alpha(\alpha Z)^4$ r.u., where m is the electron mass, α is the fine structure constant, and Z is the nuclear charge. For the two-photon width the corresponding estimate is $\Gamma_A^{2\gamma} \sim m\alpha^2(\alpha Z)^6$, so that the inequality (7) always holds.

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