Two-photon decays reexamined: cascade contributions and gauge invariance

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Ulrich D Jentschura

Max-Planck-Institut für Kernphysik, Postfach 10 39 80, 69029 Heidelberg, Germany
and
Institut für Theoretische Physik, Philosophenweg 16, 69120 Heidelberg, Germany

Abstract

The purpose of this paper is to calculate the two-photon decay rate corresponding to the two-photon transitions nS $\to$ 1S and nD $\to$ 1S in hydrogen-like ions with a low nuclear charge number Z (for principal quantum numbers $n = 2, \ldots, 8$). Numerical results are obtained within a nonrelativistic framework, and the results are found to scale approximately as $(Z\alpha)^6/n^3$, where α is the fine-structure constant. We also attempt to clarify a number of subtle issues regarding the treatment of the coherent, quasi-simultaneous emission of the two photons as opposed to one-photon cascades. In particular, the gauge invariance of the decay rate is shown explicitly.

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1. Introduction

The subject of the current paper is the two-photon decay rate of excited atomic states, interpreted as the imaginary part of the two-loop self-energy. We follow our previous investigation reported in [1] and augment the analysis by treating the decay rate in both length and velocity gauges. Special emphasis is placed on the role of singularities, infinitesimally displaced from the integration contours for the photon energy integrations, which are generated by bound-state poles of lower energy than the reference state (in the sense of the two-loop self-energy). The reference state is equivalent to the initial state of the two-photon decay process. A good quantitative understanding of the two-photon decay processes from highly excited hydrogenic bound states is important for astrophysics, as emphasized in a recent paper by Chluba and Sunyaev [2]. As the physics of the process is in principle well known and has been discussed in a previous fast track communication [1], we see no obstacle to going *in medias res* with the analysis.

Our purpose here, in addition to providing numerical data concerning the D $\to$ S transitions, is to clarify the role of cascades of one-photon decays through so-called resonant intermediate states, which are addressed using concepts developed in field theory [3, 4].

Natural units with $\hbar = c = \epsilon_0 = 1$, i.e. $e^2 = 4\pi\alpha$, are used throughout this paper, which is organized as follows. In section 2, the gauge invariance of the two-photon decay rate, as derived from the two-loop self-energy, is reanalyzed. In section 3, numerical results for $n\mathrm{D} \to 1\mathrm{S}$ transitions are presented; these were not treated in the previous paper [1]. A discussion of our results, including a comparison to previous investigations of two-photon decay from highly excited states (see [5–7]) is given in section 4. Cascade contributions are analyzed in section 5. Conclusions are drawn in section 6.

2. Gauge invariance

We start by considering the two-photon self-energy for a reference state $|\phi_i\rangle$ in a hydrogen-like ion, as derived from nonrelativistic quantum electrodynamics (NRQED). In the velocity gauge, the interaction Hamiltonian of the quantized electromagnetic field with the electron is given by

$$H_I = -\frac{e}{2m}(\vec{p}\cdot\vec{A} + \vec{A}\cdot\vec{p}) + \frac{e^2\vec{A}^2}{2m}, \tag{1}$$

where $\vec{A}$ is the vector potential of the quantized electromagnetic field.

The well-known expression (see, e.g., [1, 8]) for the two-loop self-energy reads (ω_1 and ω_2 denote the energies of the two virtual quanta)

$$\Delta E_i^{(2)} = \lim_{\epsilon\to 0}\left(\frac{2\alpha}{3\pi m^2}\right)^2 \int_0^{\Lambda_1} \mathrm{d}\omega_1\,\omega_1 \int_0^{\Lambda_2} \mathrm{d}\omega_2\,\omega_2\, f_\epsilon(\omega_1,\omega_2) = \mathrm{Re}\,\Delta E_i^{(2)} - \mathrm{i}\frac{\delta\Gamma_i^{(1)}}{2} - \mathrm{i}\frac{\Gamma_i^{(2)}}{2}. \tag{2}$$

Here, $\mathrm{Re}\,\Delta E_i^{(2)}$ is the real part of the energy shift, which gives rise, in particular, to the so-called two-loop Bethe logarithms [9]. Our treatment relies on the identification of the imaginary part of the energy shift in terms of the decay rate of the reference state, as suggested by Barbieri and Sucher in [10]. In equation (2), $\delta\Gamma_i^{(1)}$ is a correction to the one-photon decay rate, whereas $\Gamma_i^{(2)}$ is the two-photon decay rate. The former is obtained by terms where the integration over ω_1 or ω_2 meets a bound-state pole and generates an imaginary part, in the sense of equation (4) of [1], but the other photon energy is integrated with a principal-value prescription. The latter term, $\Gamma_i^{(2)}$, is obtained by selecting exclusively the imaginary part generated by the singularities at $\omega_1 + \omega_2 = E_i - E_v$, where E_v is a virtual state contained in one of the propagators. All expressions on the right-hand side of equation (2) are manifestly of order $\alpha^2(Z\alpha)^6 m$, i.e. $(Z\alpha)^6 R_\infty$, where R_∞ is the Rydberg constant.

The function f_ϵ reads as follows (with all infinitesimal imaginary parts duly taken into account):

$$\begin{aligned} f_\epsilon(\omega_1,\omega_2) &= \langle\phi_i|p^j\frac{1}{E-H-\omega_1+\mathrm{i}\epsilon}p^k\frac{1}{E-H-\omega_1-\omega_2+\mathrm{i}\epsilon}p^j\frac{1}{E-H-\omega_2+\mathrm{i}\epsilon}p^k|\phi_i\rangle \\ &+ \frac{1}{2}\langle\phi_i|p^j\frac{1}{E-H-\omega_1+\mathrm{i}\epsilon}p^k\frac{1}{E-H-\omega_1-\omega_2+\mathrm{i}\epsilon}p^k\frac{1}{E-H-\omega_1+\mathrm{i}\epsilon}p^j|\phi_i\rangle \\ &+ \frac{1}{2}\langle\phi_i|p^j\frac{1}{E-H-\omega_2+\mathrm{i}\epsilon}p^k\frac{1}{E-H-\omega_1-\omega_2+\mathrm{i}\epsilon}p^k\frac{1}{E-H-\omega_2+\mathrm{i}\epsilon}p^j|\phi_i\rangle + \cdots, \end{aligned} \tag{3}$$

where the terms denoted by the ellipsis are given in equation (3) of [1], being irrelevant for the current investigation, because the two-photon decay rate is generated exclusively by the poles where the sum $\omega_1 + \omega_2$ of both photon energies is on resonance. In a basis-set representation,

the expression for the two-photon decay rate $\Gamma^{(2)}$ is thus found from the first three terms in equation (3) as [1]

$$
\Gamma^{(2)} = \frac{4\alpha^2}{9\pi m^2} \, \mathrm{Re} \int_0^{E_i - E_f} \mathrm{d}\omega \, \omega (E_i - E_f - \omega)
$$

$$
\times \left(\sum_v \left\{ \frac{\langle \phi_f | p^k | \phi_v \rangle \langle \phi_v | p^j | \phi_i \rangle}{E_i - E_v - \omega + \mathrm{i}\epsilon} + \frac{\langle \phi_f | p^k | \phi_v \rangle \langle \phi_v | p^j | \phi_i \rangle}{E_f - E_v + \omega + \mathrm{i}\epsilon} \right\} \right)
$$

$$
\times \left(\sum_w \left\{ \frac{\langle \phi_i | p^k | \phi_w \rangle \langle \phi_w | p^j | \phi_f \rangle}{E_i - E_w - \omega + \mathrm{i}\epsilon} + \frac{\langle \phi_i | p^k | \phi_w \rangle \langle \phi_w | p^j | \phi_f \rangle}{E_f - E_w + \omega + \mathrm{i}\epsilon} \right\} \right), \tag{4}
$$

where we use the summation convention for the Cartesian coordinates labeled by the indices $j \in \{1, 2, 3\}$ and $k \in \{1, 2, 3\}$. The sum over v contains all virtual states, i.e. over the entire bound and continuous spectrum. We here imply a sum over the magnetic projections of the intermediate states, and the final state of the decay process, but an averaging over magnetic projections of the initial state (since the decay rate does not depend on the magnetic projection of the initial state, one may alternatively choose any allowed value for the initial state magnetic projection).

We now assume all initial and final, and virtual states to be given in terms of hydrogen wavefunctions in the standard representation (see, e.g., [11]), so that

$$
\langle \phi_f | p^j | \phi_v \rangle \langle \phi_v | p^j | \phi_i \rangle = \langle \phi_i | p^j | \phi_v \rangle \langle \phi_v | p^j | \phi_f \rangle, \tag{5}
$$

where the sum over j is assumed. We then do the angular algebra [12]. For $n\mathrm{S} \rightarrow 1\mathrm{S}$ decays, one obtains a result [1] which reproduces the well-known expression obtained by Göppert-Mayer in [13] for the particular case of $|\phi_i\rangle = |2\mathrm{S}\rangle$,

$$
\Gamma_{nS}^{(2)} = \frac{4\alpha^2}{27\pi m^2} \lim_{\epsilon \to 0} \mathrm{Re} \int_0^{E_{nS} - E_{1S}} \mathrm{d}\omega \, \omega (E_{nS} - E_{1S} - \omega)
$$

$$
\times \left(\sum_v \left\{ \frac{\langle 1\mathrm{S} \| \vec{p} \| v\mathrm{P} \rangle \langle v\mathrm{P} \| \vec{p} \| n\mathrm{S} \rangle}{E_{nS} - E_{vP} - \omega + \mathrm{i}\epsilon} + \frac{\langle 1\mathrm{S} \| \vec{p} \| v\mathrm{P} \rangle \langle v\mathrm{P} \| \vec{p} \| n\mathrm{S} \rangle}{E_{1S} - E_{vP} + \omega + \mathrm{i}\epsilon} \right\} \right)^2, \tag{6}
$$

where we use the definition of the reduced matrix elements according to [12]. The virtual P states are also relevant for the $n\mathrm{D} \rightarrow 1\mathrm{S}$ decays, but the well-known prefactor is different [7], and the result is

$$
\Gamma_{nD}^{(2)} = \frac{4\alpha^2}{135\pi m^2} \lim_{\epsilon \to 0} \mathrm{Re} \int_0^{E_{nD} - E_{1S}} \mathrm{d}\omega \, \omega (E_{nD} - E_{1S} - \omega)
$$

$$
\times \left(\sum_v \left\{ \frac{\langle 1\mathrm{S} \| \vec{p} \| v\mathrm{P} \rangle \langle v\mathrm{P} \| \vec{p} \| n\mathrm{D} \rangle}{E_{nD} - E_{vP} - \omega + \mathrm{i}\epsilon} + \frac{\langle 1\mathrm{S} \| \vec{p} \| v\mathrm{P} \rangle \langle v\mathrm{P} \| \vec{p} \| n\mathrm{D} \rangle}{E_{1S} - E_{vP} + \omega + \mathrm{i}\epsilon} \right\} \right)^2, \tag{7}
$$

where for completeness we note that the reduced matrix element for $P \rightarrow D$ transitions differs from the 'radial' component of the matrix element by a factor $\sqrt{2}$.

In the length gauge, the atom–field interaction is given by

$$
H_I = -e \vec{E} \cdot \vec{r}, \tag{8}
$$

where $\vec{E}$ is the quantized electric-field operator. The length-gauge two-photon self-energy is obtained by straightforward fourth-order perturbation theory as

$$
\Delta E_i^{(2)} = \lim_{\epsilon \to 0} \left(\frac{2\alpha}{3\pi m^2} \right)^2 \int_0^{\Lambda_1} \mathrm{d}\omega_1 \, \omega_1^3 \int_0^{\Lambda_2} \mathrm{d}\omega_2 \, \omega_2^3 g_\epsilon(\omega_1, \omega_2) = \mathrm{Re} \, \Delta E_i^{(2)} - \mathrm{i} \frac{\delta \Gamma_i^{(1)}}{2} - \mathrm{i} \frac{\Gamma_i^{(2)}}{2}. \tag{9}
$$

We observe the factor $\omega_1^3 \omega_2^3$, which is characteristic of the length-gauge formulation. The absence of the seagull term as opposed to the velocity gauge leads to a somewhat simplified expression,

$$
\begin{aligned}
g_\epsilon(\omega_1, \omega_2) =\ & \langle \phi_i | x^j \frac{1}{E - H - \omega_1 + i\epsilon} x^k \frac{1}{E - H - \omega_1 - \omega_2 + i\epsilon} x^j \frac{1}{E - H - \omega_2 + i\epsilon} x^k | \phi_i \rangle \\
& + \frac{1}{2} \langle \phi_i | x^j \frac{1}{E - H - \omega_1 + i\epsilon} x^k \frac{1}{E - H - \omega_1 - \omega_2 + i\epsilon} x^k \frac{1}{E - H - \omega_1 + i\epsilon} x^j | \phi_i \rangle \\
& + \frac{1}{2} \langle \phi_i | x^j \frac{1}{E - H - \omega_2 + i\epsilon} x^k \frac{1}{E - H - \omega_1 - \omega_2 + i\epsilon} x^k \frac{1}{E - H - \omega_2 + i\epsilon} x^j | \phi_i \rangle \\
& + \langle \phi_i | x^j \frac{1}{E - H - \omega_1 + i\epsilon} x^j \left(\frac{1}{E - H} \right)' x^k \frac{1}{E - H - \omega_2 + i\epsilon} x^k | \phi_i \rangle \\
& - \frac{1}{2} \langle \phi_i | x^j \frac{1}{E - H - \omega_1 + i\epsilon} x^j | \phi_i \rangle \langle \phi_i | x^k \left(\frac{1}{E - H - \omega_2 + i\epsilon} \right)^2 x^k | \phi_i \rangle \\
& - \frac{1}{2} \langle \phi_i | x^j \frac{1}{E - H - \omega_2 + i\epsilon} x^j | \phi_i \rangle \langle \phi_i | x^k \left(\frac{1}{E - H - \omega_1 + i\epsilon} \right)^2 x^k | \phi_i \rangle. \quad (10)
\end{aligned}
$$

In contrast to equation (3), the momentum operators are replaced by position operators. In analogy to equation (3), only the first three terms are relevant for the two-photon decay rate. Using a basis-set representation, the expression for the two-photon decay rate derived in the length gauge thus reads

$$
\begin{aligned}
\Gamma^{(2)} =\ & \frac{4\alpha^2}{9\pi m^2} \, \mathrm{Re} \int_0^{E_i - E_f} \mathrm{d}\omega \, \omega^3 (E_i - E_f - \omega)^3 \\
& \times \left(\sum_v \left\{ \frac{\langle \phi_f | x^k | \phi_v \rangle \langle \phi_v | x^j | \phi_i \rangle}{E_i - E_v - \omega + i\epsilon} + \frac{\langle \phi_f | x^k | \phi_v \rangle \langle \phi_v | x^j | \phi_i \rangle}{E_f - E_v + \omega + i\epsilon} \right\} \right) \\
& \times \left(\sum_w \left\{ \frac{\langle \phi_i | x^k | \phi_w \rangle \langle \phi_w | x^j | \phi_f \rangle}{E_i - E_w - \omega + i\epsilon} + \frac{\langle \phi_i | x^k | \phi_w \rangle \langle \phi_w | x^j | \phi_f \rangle}{E_f - E_w + \omega + i\epsilon} \right\} \right), \quad (11)
\end{aligned}
$$

where the sum over v contains all virtual states. Using the identity

$$
\begin{aligned}
\sum_v & \left\{ \frac{\langle \phi_f | p^k | \phi_v \rangle \langle \phi_v | p^j | \phi_i \rangle}{E_i - E_v - \omega + i\epsilon} + \frac{\langle \phi_f | p^k | \phi_v \rangle \langle \phi_v | p^j | \phi_i \rangle}{E_f - E_v + \omega + i\epsilon} \right\} \\
& = \omega (E_i - E_f - \omega) \sum_v \left\{ \frac{\langle \phi_f | x^k | \phi_v \rangle \langle \phi_v | x^j | \phi_i \rangle}{E_i - E_v - \omega + i\epsilon} + \frac{\langle \phi_f | x^k | \phi_v \rangle \langle \phi_v | x^j | \phi_i \rangle}{E_f - E_v + \omega + i\epsilon} \right\}, \quad (12)
\end{aligned}
$$

it is easy to show the equivalence of the two expressions for the two-photon decay rate given in equations (2) and (9). Note that this equivalence can be shown easily using the commutator relation $p^i = i[H, x^j]$, but it holds only if the sum over v extends over the complete spectrum.

Assuming hydrogen wavefunctions in the standard representation, we have that in analogy to equation (5),

$$
\langle \phi_f | x^j | \phi_v \rangle \langle \phi_v | x^j | \phi_i \rangle = \langle \phi_i | x^j | \phi_v \rangle \langle \phi_v | x^j | \phi_f \rangle. \quad (13)
$$

After angular algebra, one obtains for the decay $nS \to 1S$

$$
\begin{aligned}
\Gamma_{nS}^{(2)} =\ & \frac{4\alpha^2}{27\pi m^2} \lim_{\epsilon \to 0} \mathrm{Re} \int_0^{E_{nS} - E_{1S}} \mathrm{d}\omega \, \omega^3 (E_{nS} - E_{1S} - \omega)^3 \\
& \times \left(\sum_v \left\{ \frac{\langle 1S \| \vec{x} \| v\mathrm{P} \rangle \langle v\mathrm{P} \| \vec{x} \| nS \rangle}{E_{nS} - E_{v\mathrm{P}} - \omega + i\epsilon} + \frac{\langle 1S \| \vec{x} \| v\mathrm{P} \rangle \langle v\mathrm{P} \| \vec{x} \| nS \rangle}{E_{1S} - E_{v\mathrm{P}} + \omega + i\epsilon} \right\} \right)^2, \quad (14)
\end{aligned}
$$

Table 1. Numerical results for the decay rates nS $\to$ 1S and nD $\to$ 1S for hydrogen. The rates scale with Z^6 for hydrogen-like ions with nuclear charge number Z. Units are in inverse seconds. To obtain the decay rate in hertz, one needs to divide by a factor of 2π. We here supplement the results given in [1] by some values for higher excited S states and also indicated results for nD $\to$ 1S, which were not treated in [1].

	$\lvert\phi_f\rangle = \lvert 1\mathrm{S}\rangle$		$\lvert\phi_f\rangle = \lvert 1\mathrm{S}\rangle$
$\lvert\phi_i\rangle = \lvert 2\mathrm{S}\rangle$	8.229 352		
$\lvert\phi_i\rangle = \lvert 3\mathrm{S}\rangle$	2.082 853	$\lvert\phi_i\rangle = \lvert 3\mathrm{D}\rangle$	1.042 896
$\lvert\phi_i\rangle = \lvert 4\mathrm{S}\rangle$	0.698 897	$\lvert\phi_i\rangle = \lvert 4\mathrm{D}\rangle$	0.598 798
$\lvert\phi_i\rangle = \lvert 5\mathrm{S}\rangle$	0.287 110	$\lvert\phi_i\rangle = \lvert 5\mathrm{D}\rangle$	0.340 883
$\lvert\phi_i\rangle = \lvert 6\mathrm{S}\rangle$	0.135 935	$\lvert\phi_i\rangle = \lvert 6\mathrm{D}\rangle$	0.206 523
$\lvert\phi_i\rangle = \lvert 7\mathrm{S}\rangle$	0.071 402	$\lvert\phi_i\rangle = \lvert 7\mathrm{D}\rangle$	0.132 928
$\lvert\phi_i\rangle = \lvert 8\mathrm{S}\rangle$	0.040 587	$\lvert\phi_i\rangle = \lvert 8\mathrm{D}\rangle$	0.090 016

whereas for nD $\to$ 1S decays

$$\Gamma_{n\mathrm{D}}^{(2)} = \frac{4\alpha^2}{135\pi m^2} \lim_{\epsilon \to 0} \mathrm{Re} \int_0^{E_i - E_f} \mathrm{d}\omega\, \omega^3 (E_{n\mathrm{D}} - E_{1\mathrm{S}} - \omega)^3$$

$$\times \left(\sum_\nu \left\{ \frac{\langle 1\mathrm{S}\|\vec{x}\|\nu\mathrm{P}\rangle\langle\nu\mathrm{P}\|\vec{x}\|n\mathrm{D}\rangle}{E_{n\mathrm{D}} - E_{\nu\mathrm{P}} - \omega + \mathrm{i}\epsilon} + \frac{\langle 1\mathrm{S}\|\vec{x}\|\nu\mathrm{P}\rangle\langle\nu\mathrm{P}\|\vec{x}\|n\mathrm{D}\rangle}{E_{1\mathrm{S}} - E_{\nu\mathrm{P}} + \omega + \mathrm{i}\epsilon} \right\} \right)^2, \tag{15}$$

again in complete analogy to equations (6) and (7), respectively.

3. Numerical results

We here focus on the nS $\to$ 1S and nD $\to$ 1S decays, as indicated in equations (14) and (15), respectively. Decays to the ground state have the highest rate for both one-photon [14] as well as two-photon processes and are therefore of special interest. Due to the infinitesimal imaginary parts explicitly indicated in equations (14) and (15), we can extend the sum over intermediate, virtual states over the entire hydrogenic spectrum, including those P states which have a lower energy than the reference state. We recall here that the double poles at intermediate resonances are naturally treated using the formula [1]

$$\lim_{\epsilon \to 0} \mathrm{Re} \int_0^1 \mathrm{d}\omega \left(\frac{1}{a - \omega + \mathrm{i}\epsilon} \right)^2 = \frac{1}{a(a-1)}. \tag{16}$$

Simple poles are treated using the well-known Dirac prescription, and the principal-value integration then yields the real part of the integrals. Numerical results can be obtained by expressing the matrix elements with the propagators in terms of hypergeometric functions, following [15, 16]. Final values are indicated in table 1.

The one-loop as well as the two-loop self-energy shifts of hydrogenic states are well known to follow scaling laws of the form of inverse powers of the principal quantum number n, as analyzed in [17]. The two-photon decay rate is the imaginary part of this energy shift and is thus expected to follow an analogous trend with the principal quantum numbers. Analyzing the data in table 1, we find that the nD $\to$ 1S state results appear to follow the asymptotic behavior (expressed in inverse seconds)

$$\Gamma_{n\mathrm{D}}^{(2)} = \frac{49(2)}{n^3} Z^6 \mathrm{s}^{-1}, \qquad n \to \infty, \tag{17}$$

whereas for $nS \rightarrow 1S$ decay, a fractional power apparently leads to a more satisfactory representation of the data,

$$\Gamma_{nS}^{(2)} = \frac{330(20)}{n^{4.3}} Z^6 s^{-1}, \qquad n \rightarrow \infty. \tag{18}$$

The results indicated in table 1 are consistent with a decrease of the two-photon decay rate with increasing n.

4. Discussion and comparison

When comparing to the existing literature, it is useful, first of all, to note the calculations [2, 5, 6], which are apparently based on second-order perturbation theory for the two-photon transition amplitude. As a consequence, they present singularities when the energy of one of the photons reaches a level situated between the initial and final states, and no procedure is given in the cited references if one does not go beyond second order. When evaluating differential transition rates [5, 18, 19], the absence of the infinitesimal imaginary part does not matter, and the numerical results in the velocity gauge [5, 19] and in the length gauge [18] fully agree. The problem arises when one tries to evaluate the total decay rate, as the existing singularities are not integrable. Although in [7] fourth-order perturbation theory was used, a consistent answer does not appear to be found.

It appears that, in general, two approaches have been used so far in the literature in order to deal with the problematic double poles for the photon energy integrations: (i) the explicit removal of particular states from the sum over virtual states and (ii) the inclusion of a width for the intermediate, virtual states.

Let us begin the discussion with the removal of states. Indeed, Chluba and Sunyaev [2], Florescu *et al* [6] as well as Cresser *et al* [7] have used different formulae than those used here, in order to evaluate the two-photon decay rates. In particular, they use instead of equation (14) the following formula for $nS \rightarrow 1S$ decays:

$$\gamma_{nS}^{(2)} = \frac{4\alpha^2}{27\pi m^2} \int_0^{E_{nS}-E_{1S}} d\omega\, \omega^3 (E_{nS} - E_{1S} - \omega)^3$$
$$\times \left| \sum_{\nu \geqslant N} \left\{ \frac{\langle 1S\|\vec{x}\|\nu P\rangle\langle \nu P\|\vec{x}\|nS\rangle}{E_{nS} - E_{\nu P} - \omega} + \frac{\langle 1S\|\vec{x}\|\nu P\rangle\langle \nu P\|\vec{x}\|nS\rangle}{E_{1S} - E_{\nu P} + \omega} \right\} \right|^2, \tag{19}$$

where $N = n$ (Chluba and Sunyaev [2]) or $N = n + 1$ (Florescu *et al* [5, 6] and Cresser *et al* [7]), and the notation $\nu \geqslant N$ of course means that one should sum over the discrete spectrum for all virtual states with principal quantum numbers as indicated, and of course integrate over the entire continuum spectrum in addition. For $nD \rightarrow 1S$ decays, the cited authors use

$$\gamma_{nD}^{(2)} = \frac{4\alpha^2}{135\pi m^2} \int_0^{E_{nD}-E_{1S}} d\omega\, \omega^3 (E_{nD} - E_{1S} - \omega)^3$$
$$\times \left| \sum_{\nu \geqslant N} \left\{ \frac{\langle 1S\|\vec{x}\|\nu P\rangle\langle \nu P\|\vec{x}\|nD\rangle}{E_{nD} - E_{\nu P} - \omega} + \frac{\langle 1S\|\vec{x}\|\nu P\rangle\langle \nu P\|\vec{x}\|nD\rangle}{E_{1S} - E_{\nu P} + \omega} \right\} \right|^2, \tag{20}$$

with the same proposed values for N. In this case, because the problematic virtual states of lower energy than the initial state $|\phi_i\rangle$ have been explicitly removed from the sum over virtual states, there are no more singularities infinitesimally displaced from the integration contours present, and there is therefore no need for any infinitesimal imaginary part $i\epsilon$ in the propagator

denominators. Furthermore, $|\cdot|^2$ is equivalent to $(\cdot)^2$ provided our assumption formulated in equation (5) holds. The corresponding velocity-gauge expressions

$$
\eta_{nS}^{(2)} = \frac{4\alpha^2}{27\pi m^2} \int_0^{E_{nS}-E_{1S}} d\omega\, \omega(E_{nS} - E_{1S} - \omega)
$$

$$
\times \left| \sum_{\nu \geqslant N} \left\{ \frac{\langle 1S\|\vec{p}\|\nu P\rangle\langle \nu P\|\vec{p}\|nS\rangle}{E_{nS} - E_{\nu P} - \omega} + \frac{\langle 1S\|\vec{p}\|\nu P\rangle\langle \nu P\|\vec{p}\|nS\rangle}{E_{1S} - E_{\nu P} + \omega} \right\} \right|^2 \tag{21}
$$

and

$$
\eta_{nD}^{(2)} = \frac{4\alpha^2}{135\pi m^2} \int_0^{E_{nD}-E_{1S}} d\omega\, \omega(E_{nD} - E_{1S} - \omega)
$$

$$
\times \left| \sum_{\nu \geqslant N} \left\{ \frac{\langle 1S\|\vec{p}\|\nu P\rangle\langle \nu P\|\vec{p}\|nD\rangle}{E_{nD} - E_{\nu P} - \omega} + \frac{\langle 1S\|\vec{p}\|\nu P\rangle\langle \nu P\|\vec{p}\|nD\rangle}{E_{1S} - E_{\nu P} + \omega} \right\} \right|^2 \tag{22}
$$

are not equivalent to the length-gauge expressions in equations (19) and (20), because relation (12) breaks down if the sum over ν does not extend over the entire hydrogen spectrum. The explicit removal of the 'problematic' virtual states from the propagators avoids the necessity of indicating the infinitesimal imaginary terms in the propagator denominators, but the removal operation leads to different expressions in the length and velocity gauges and is thus not gauge invariant.

To illustrate this finding by a numerical example, we observe that we can reproduce the value of $\gamma_{3D}^{(2)} = 0.131\,813$ s^{-1} for the decay 3D $\to$ 1S with $N = n + 1$ using the length-gauge expression (20), in agreement with equation (20) of [7]. However, the velocity-gauge expression (22) gives a different result, namely, $\eta_{3D}^{(2)} = 0.439\,368$ s^{-1}. These two results have to be contrasted with the gauge-invariant result of $\Gamma_{3D}^{(2)} = 1.042\,896$ s^{-1}, indicated in table 1. For the decay 3S $\to$ 1S, the values are $\gamma_{3S}^{(2)} = 8.225\,796$ s^{-1} in agreement with equation (19) of [7], and the velocity-gauge result with 2P and 3P virtual states removed is $\eta_{3S}^{(2)} = 6.192\,881$ s^{-1}, whereas the gauge-invariant result with the full hydrogenic spectrum of virtual states reads $\Gamma_{3S}^{(2)} = 2.082\,853$ s^{-1} (see table 1). It is interesting to observe that $\Gamma_{3D}^{(2)} > \gamma_{3D}^{(2)}$, but $\Gamma_{3S}^{(2)} < \gamma_{3S}^{(2)}$.

Let us now turn our attention to the inclusion of a decay width for the intermediate states. Indeed, the authors of [2, 5–7] arrive at expressions (21) and (22) after analyzing the expression (for illustrative purposes we restrict ourselves here to the nS $\to$ 1S decay)

$$
\frac{4\alpha^2}{27\pi m^2} \int_0^{E_{nS}-E_{1S}} d\omega\, \omega^3(E_{nS} - E_{1S} - \omega)^3
$$

$$
\times \left| \sum_{\nu} \left\{ \frac{\langle 1S\|\vec{x}\|\nu P\rangle\langle \nu P\|\vec{x}\|nS\rangle}{E_{nS} - E_{\nu P} - \omega + \frac{i}{2}\Gamma_\nu^{(1)}} + \frac{\langle 1S\|\vec{x}\|\nu P\rangle\langle \nu P\|\vec{x}\|nS\rangle}{E_{1S} - E_{\nu P} + \omega + \frac{i}{2}\Gamma_\nu^{(1)}} \right\} \right|^2. \tag{23}
$$

Let us consider the 3S state as an example. The only 'problematic' virtual state is the 2P state ($\nu = 2$), and using the formula

$$
\int_0^1 d\omega \left| \frac{1}{a - \omega + i\Gamma} \right|^2 = \frac{\pi}{\Gamma} + \frac{1}{a(a-1)} + \mathcal{O}(\Gamma^2), \tag{24}
$$

it is possible to show that the term with $\nu = 2$ in expression (23) gives rise to a contribution which is equivalent to the one-photon decay rate 3S $\to$ 2P, and this decay rate is just the total one-photon decay rate of the 3S state, and it is equal to the imaginary part of the one-loop

self-energy of the 3S state (in the dipole approximation). The authors of [2, 5–7] thus conclude that this term should be interpreted as the one-photon decay rate of the 3S state, which has got nothing to do with the two-photon decay process, and this observation appears to be the basis for their removal of the 2P state from the sum over virtual states (see also the analysis in footnote 4 of [7]).

Despite the appealing aspects of the removal operation, it is unfortunately not gauge invariant, as shown above, and we would like to point out two more aspects that merit a discussion. First and foremost, the discussion in footnote 4 of [7] shows that expression (23) gives rise to a one-photon decay rate, effectively mixing the two-loop self-energy with the one-photon self-energy (according to the interpretation of the decay rate as an imaginary part of an energy shift). If one would take expression (23) literally, then one should be careful to avoid a double counting of the one-photon decay rate, which is already contained in the one-loop self-energy and should not be obtained once more from the imaginary part of the two-loop self-energy. Cascade contributions are discussed in more detail in section 5.

The second aspect is observed when the analysis in footnote 4 of [7] is generalized to the 4S → 1S decay. In that case, two cascades are possible, namely, 4S → 3P → 1S and 4S → 2P → 1S. As an easy generalization of the analysis in footnote 4 of [7] shows, the full one-photon decay rate of the 4S state is obtained from expression (23) only if the virtual 2P and 3P are endowed with their *partial* decay rates to the 1S ground state, i.e. the 3P decay rate should be inserted into the propagator denominators as the *partial* decay rate 3P → 1S, excluding the decay process 3P → 2S. If one generalizes these considerations further, namely, to a general decay nS → 1S, then this would imply that one should use different decay rates $\Gamma_v^{(1)}$ in equation (23) to regularize the divergence in $1/\Gamma_v^{(1)}$ in equation (24), adjusting them according to the decay process under study. That prescription would be highly counterintuitive as the virtual states should somehow 'know' about properties of the initial and final states of the decay process. The ensuing questions have already been noticed by Chluba and Sunyaev [2].

Let us conclude this section with a remark on asymptotics (17) and (18), which permit an extrapolation of our results to Rydberg states with high principal quantum numbers. Some investigations, including [2], lead to results for the two-photon decay rates of higher excited state which exhibit a linear increase with n instead of a decrease with at least n^{-3}, as indicated in equations (17) and (18). It is well known that the one-photon rates decrease approximately with n^{-3} (see [14]). If the two-photon rates would indeed increase linearly, then there would be a relative factor n^4 with which two-photon rates would grow in comparison to one-photon rates as the principal quantum number of a state increases. If we take into account the relative scaling factor of $Z^2\alpha^3/\pi$ by which two-photon rates are suppressed with respect to one-photon rates, then we would have to conclude that the two-photon rates overtake the one-photon rates for states with a comparatively low principal quantum number of $n \approx 50/\sqrt{Z}$ in a hydrogen-like ion with nuclear charge number Z. For our results as indicated in table 1, the two-photon rates are suppressed with respect to one-photon rates by a relative factor $Z^2\alpha^3/\pi$ for all hydrogenic states, because the scaling with n is obtained to be approximately the same for the one-photon as well as the two-photon rates, and the natural hierarchy of the likelihood of one- and two-photon events is preserved for all states.

5. Extraction of the cascade contribution

As in section 4, let us focus on a particular example whose generalization is obvious, namely (this time) the 3S → 1S decays, for which the cascade 3S → 2P → 1S needs to be addressed.

Let us go back once more to equation (14),

$$\Gamma_{3S}^{(2)} = \frac{4\alpha^2}{27\pi m^2} \lim_{\epsilon \to 0} \text{Re} \int_0^{E_{3S}-E_{1S}} d\omega\, \omega^3 (E_{3S} - E_{1S} - \omega)^3$$

$$\times \left(\sum_\nu \left\{ \frac{\langle 1S\|\vec{x}\|\nu P\rangle\langle\nu P\|\vec{x}\|3S\rangle}{(E_{3S} - E_{\nu P} - \omega + i\epsilon)} + \frac{\langle 1S\|\vec{x}\|\nu P\rangle\langle\nu P\|\vec{x}\|3S\rangle}{(E_{1S} - E_{\nu P} + \omega + i\epsilon)} \right\} \right)^2 \quad (25)$$

and adopt the cumbersome, but absolutely unique notation P.V. for the principal value part of the distribution. If we use the formula

$$\frac{1}{x + i\epsilon} = (\text{P.V.})\frac{1}{x} - i\pi\delta(x) \quad (26)$$

for all propagator denominators in equation (25) and extract only the contribution due to the delta functions, then the only contributing virtual state is the 2P state. Because there is a product of two terms both of which become singular, we cannot avoid to obtain the square of the delta function,

$$\delta^2(\omega - E_{3S} - E_{2P}) = \delta(0)\delta(\omega - E_{3S} - E_{2P}) = \frac{T}{2\pi}\delta(\omega - E_{3S} - E_{2P}), \quad (27)$$

and a further term proportional to $\delta(\omega - E_{2P} + E_{1S})T/(2\pi)$. Here, T is the (long) observation time proportional to $\delta(0)$ in energy space (see, e.g., [3]). The sum of the terms proportional to $\delta(0)$ reads

$$C_{3S}^{(2)} = -T\Gamma_{3S\to2P}^{(1)}\Gamma_{2P\to1S}^{(1)}, \qquad \int dT\, C_{3S}^{(2)} = -\frac{T^2}{2}\Gamma_{3S\to2P}^{(1)}\Gamma_{2P\to1S}^{(1)}, \quad (28)$$

where we introduce an obvious notation for the partial one-photon rates $\Gamma_{3S\to2P}^{(1)}$ and $\Gamma_{2P\to1S}^{(1)}$. Note, in particular, that the resulting expression for $C_{3S}^{(2)}$ is gauge invariant.

Our result (28) has just the right form to describe the cascade decay, except for the 'wrong' sign. For the term to contribute to the decay of the 3S state, it should be positive, but it turns out to be negative. Let us defer a discussion of this issue and instead consider the extraction of the cascade contribution from the expression

$$\tilde{\Gamma}_{3S}^{(2)} = \frac{4\alpha^2}{27\pi m^2} \lim_{\epsilon \to 0} \text{Re} \int_0^{E_{3S}-E_{1S}} d\omega\, \omega^3 (E_{3S} - E_{1S} - \omega)^3$$

$$\times \prod_\pm \sum_\nu \left\{ \frac{\langle 1S\|\vec{x}\|\nu P\rangle\langle\nu P\|\vec{x}\|3S\rangle}{E_{3S} - E_{\nu P} - \omega \pm i\epsilon} + \frac{\langle 1S\|\vec{x}\|\nu P\rangle\langle\nu P\|\vec{x}\|3S\rangle}{E_{1S} - E_{\nu P} + \omega \pm i\epsilon} \right\}, \quad (29)$$

where $\prod_\pm$ means the product of two terms, with either sign. The product over the two terms with $\pm i\epsilon$ is of course equivalent to the square of the modulus of the two terms in the integrand, in analogy to equation (23). If we now use (26), then we obtain

$$\tilde{C}_{3S}^{(2)} = T\Gamma_{3S\to2P}^{(1)}\Gamma_{2P\to1S}^{(1)}, \qquad \int dT\, \tilde{C}_{3S}^{(2)} = \frac{T^2}{2}\Gamma_{3S\to2P}^{(1)}\Gamma_{2P\to1S}^{(1)}. \quad (30)$$

This result has the 'right' sign, and has, for large T, the right temporal dependence for a cascade process.

In order to resolve the paradox, one first should note that both signs found in equations (28) and (30) actually have a valid interpretation. The two-loop self-energy contains both radiative corrections to the one-photon decay as well as the full two-photon decay amplitude. The

radiative corrections to the one-photon decay are obtained by 'cutting' appropriate internal lines in the diagrams, and indeed, we can rederive the first three radiative corrections to one-photon as given in equation (27) of [20] by considering resonant intermediate states in the 'outer' electron propagators in terms of equation (3). (The remaining terms used in equation (27) of [20] follow from standard third-order perturbation theory.) The magnitude of the radiative corrections to one-photon decay is decreased by the possibility of cascade processes, due to the virtual-to-real conversion of the photons appearing in the integrals for the radiative corrections to the one-photon decay at resonance, and this decrease is consistent with the sign of the right-hand side of equation (28). On the other hand, the two-photon decay amplitude should be increased by the cascade processes, and this increase is consistent with the sign of the right-hand side of equation (30).

In the sum of the radiative corrections to the one- and two-photon decays, the incoherent cascade contributions cancel, and this is in analogy to the discussion in [3] for a different, but physically related process, namely, the coherent/incoherent pair production via a virtual/real photon intermediate state by an electron in crossed, static electromagnetic fields.

Immediately, new questions arise. Our considerations suggest that our formulation in equation (3) provides infinitesimal imaginary parts that are appropriate for the evaluation of radiative corrections to the one-photon decay, but provides the 'wrong' sign of the cascades for two-photon decays. This could lead to new doubt regarding whether we can extract a valid expression for the two-photon decay rate from our equation (3) in the first place. The question is: Can we extract, by some mathematically justifiable procedure, from equation (29), an expression for the two-photon decay rate which either confirms or invalidates our result for the two-photon decay rate, under a suitable gauge-invariant subtraction of the cascade contribution from the integrand in (29)?

First, since the cascade contributions correspond to the delta function in equation (26), it is clear that the two-photon decay rate corresponds to the product of two principal-value distributions of the form,

$$\left(\text{P.V.} \frac{1}{\omega - \omega_0} \right) \left(\text{P.V.} \frac{1}{\omega - \omega_0} \right) = \left(\text{P.V.} \frac{1}{\omega - \omega_0} \right)^2, \tag{31}$$

which is integrated over ω. As similar problems have occurred in field theory (see equation (6.23) on p 168 of [4]), we are provided with a guiding principle for the calculation. Namely, we consider an arbitrary function f, integrated over a finite interval $(0, \omega_{\max})$ with $f(0) = f(\omega_{\max}) = 0$:

$$\int_0^{\omega_{\max}} \mathrm{d}\omega \, f(\omega) \left(\text{P.V.} \frac{1}{\omega - \omega_0} \right)^2 = \lim_{\eta \to 0} \int_0^{\omega_{\max}} \mathrm{d}\omega \, f(\omega) \left(\text{P.V.} \frac{1}{\omega - \omega_0 + \eta} \right) \left(\text{P.V.} \frac{1}{\omega - \omega_0} \right)$$

$$= \lim_{\eta \to 0} \frac{1}{\eta} \int_0^{\omega_{\max}} \mathrm{d}\omega \, f(\omega) \left(\text{P.V.} \frac{1}{\omega - \omega_0} - \text{P.V.} \frac{1}{\omega - \omega_0 + \eta} \right)$$

$$= \lim_{\eta \to 0} \frac{1}{\eta} \int_0^{\omega_{\max}} \mathrm{d}\omega [f(\omega) - f(\omega - \eta)] \left(\text{P.V.} \frac{1}{\omega - \omega_0} \right)$$

$$= \int_0^{\omega_{\max}} \mathrm{d}\omega \left[\frac{\partial}{\partial \omega} (f(\omega) - f(\omega_0)) \right] \left(\text{P.V.} \frac{1}{\omega - \omega_0} \right)$$

$$= -\frac{\omega_{\max} f(\omega_0)}{\omega_0(\omega_{\max} - \omega_0)} + \text{P.V.} \int_0^{\omega_{\max}} \mathrm{d}\omega \, \frac{f(\omega) - f(\omega_0)}{(\omega - \omega_0)^2}. \tag{32}$$

This subtraction, applied to equation (29), gives rise to

$$
\Gamma_{3S}^{(2)} = \frac{4\alpha^2}{27\pi m^2} \text{P.V.} \int_0^{E_{3S}-E_{1S}} d\omega
$$

$$
\times \left(\omega^3 (E_{3S} - E_{1S} - \omega)^3 \left| \sum_\nu \left\{ \frac{\langle 1S\|\vec{x}\|\nu P\rangle \langle \nu P\|\vec{x}\|3S\rangle}{E_{3S} - E_{\nu P} - \omega} + \frac{\langle 1S\|\vec{x}\|\nu P\rangle \langle \nu P\|\vec{x}\|3S\rangle}{E_{1S} - E_{\nu P} + \omega} \right\} \right|^2 \right.
$$

$$
- (E_{3S} - E_{2P})^3 (E_{2P} - E_{1S})^3
$$

$$
\left. \times \left(\frac{\langle 1S\|\vec{x}\|2P\rangle^2 \langle 2P\|\vec{x}\|3S\rangle^2}{(E_{3S} - E_{2P} - \omega)^2} + \frac{\langle 1S\|\vec{x}\|2P\rangle^2 \langle 2P\|\vec{x}\|3S\rangle^2}{(E_{2P} - E_{1S} - \omega)^2} \right) \right) + \frac{4\alpha^2}{27\pi m^2} \mathcal{F}. \tag{33}
$$

Here, we have subtract the cascade-generating terms according to equation (32), thus leading to an integral which is finite under a principal-value prescription, because the double poles have explicitly been subtracted. Because prescription (32) takes the numerators to exact resonance, the subtraction terms in (33) are gauge invariant and indeed proportional to $\Gamma_{3S\to2P}^{(1)}\Gamma_{2P\to1S}^{(1)}$. The additional term $\mathcal{F}$ is due to the boundary term found in (32),

$$
\mathcal{F} = -2(E_{3S} - E_{1S})(E_{3S} - E_{2P})^2 (E_{2P} - E_{1S})^2 \langle 1S\|\vec{x}\|2P\rangle^2 \langle 2P\|\vec{x}\|3S\rangle^2. \tag{34}
$$

Finally, returning to our original $i\epsilon$ prescription, we have according to equation (16),

$$
\lim_{\epsilon\to0} \text{Re} \int_0^{\omega_{\max}} d\omega \frac{f(\omega_0)}{(\omega - \omega_0 + i\epsilon)^2} = -\frac{\omega_{\max} f(\omega_0)}{\omega_0(\omega_{\max} - \omega_0)} \tag{35}
$$

and in view of (32) and (35), we obtain the (perhaps somewhat surprising) equality

$$
\lim_{\epsilon\to0} \text{Re} \int_0^{\omega_{\max}} d\omega \frac{f(\omega)}{(\omega - \omega_0 + i\epsilon)^2} = \int_0^{\omega_{\max}} d\omega\, f(\omega) \left(\text{P.V.} \frac{1}{\omega - \omega_0} \right)^2, \tag{36}
$$

which is subject to the interpretation of the squared principal-value contribution according to equation (32). We can finally state that result (33) agrees with formula (25), so that, under the provisions of the regularization implied by equation (32), it is irrelevant if we start from an expression where the integrand for the two-photon decay is formulated as a modulus squared or with two infinitesimal imaginary parts pointing in the same direction.

6. Conclusions

We have analyzed two-photon decay processes involving $n\text{S} \to 1\text{S}$ and $n\text{D} \to 1\text{S}$ channels in hydrogen-like ions. Our general formulae (4) and (11) are gauge-invariant and are obtained with otherwise unspecified, arbitrary infinitesimal imaginary parts $i\epsilon$, provided the limit $\epsilon \to 0$ is taken after the integrations over the photon energies have been performed (non-uniform convergence). Numerical results are presented in table 1. These are nonrelativistic results which scale as Z^6 with the nuclear charge number Z. For a relativistic generalization, see [21].

From a more philosophical point of view, we can say that the two-photon decay process turns out to be an extremely subtle physical phenomenon, which demands a lot of mathematical sophistication in its analysis. Without a careful handling of the distributions, including ill-defined squares of delta functions, it is impossible to obtain consistent answers. The

current work attempts to provide a proposal for a consistent framework in which the resonant intermediate states and the generated double poles can be addressed, while preserving the interpretation of the integrand of the two-photon decay rate as a differential decay rate with respect to the photon energy.

Three remarks conclude this work. (i) Following [3], we should point out that there is no guarantee that the coherent two-photon decay rate as evaluated here always needs to be positive (except for the 2S state, where no resonant intermediate states are present). Indeed, as equation (33) shows, the result is obtained as a subtracted integral, and the integrand is not necessarily positive. For all transitions considered here, the rate is positive (see table 1), but it is known that radiative corrections to decay rates can be negative, and the coherent two-particle contribution to the decay rate beyond the cascade constitutes a correction to the decay rate which need not be positive. This statement is paradoxical, but we can point out that this statement has already been confirmed after equation (20) of [3] in an absolutely analogous situation. (ii) The observation time T as implied in equation (30) has to be sufficiently large (larger than the typical formation time of radiation in the system, according to [3], or otherwise the decay process will proceed in a different way). In our case, the natural formation time of radiation is given by a time inversely related to the decay width of the initial state of the process, which is naturally identified as the one-photon decay rate of the highly excited states. (iii) It may seem that the agreement of the integration around the infinitesimally displaced poles as described in section 2 and the regularized principal-value prescription as described in section 5 is purely accidental. However, one should remember that similar integrals appear in Lamb-shift related self-energy calculations (e.g., [22]), and therefore, the predictions for the Lamb shift of excited states would have had to be reinvestigated if we had not found agreement of the two computational schemes discussed here. Fortunately, the internal consistency of mathematics protects us from having to reinvestigate accurate theoretical predictions based on quantum electrodynamics.

Acknowledgments

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