



Missouri University of Science and Technology
Scholars' Mine

Physics Faculty Research & Creative Works

Physics

01 Jan 2010

Separation of Transitions with Two Quantum Jumps from Cascades

Ulrich D. Jentschura

Missouri University of Science and Technology, ulj@mst.edu

Follow this and additional works at: https://scholarsmine.mst.edu/phys_facwork

 Part of the [Physics Commons](#)

Recommended Citation

U. D. Jentschura, "Separation of Transitions with Two Quantum Jumps from Cascades," *Physical Review A - Atomic, Molecular, and Optical Physics*, vol. 81, no. 1, pp. 012112-1-012112-8, American Physical Society (APS), Jan 2010.

The definitive version is available at <https://doi.org/10.1103/PhysRevA.81.012112>

This Article - Journal is brought to you for free and open access by Scholars' Mine. It has been accepted for inclusion in Physics Faculty Research & Creative Works by an authorized administrator of Scholars' Mine. This work is protected by U. S. Copyright Law. Unauthorized use including reproduction for redistribution requires the permission of the copyright holder. For more information, please contact scholarsmine@mst.edu.

Separation of transitions with two quantum jumps from cascades

Ulrich D. Jentschura

Department of Physics, Missouri University of Science and Technology, Rolla, Missouri 65409-0640, USA

(Received 22 July 2009; published 20 January 2010)

We consider the general scenario of an excited level $|i\rangle$ of a quantum system that can decay via two channels: (i) via a single-quantum jump to an intermediate, resonant level $|\bar{m}\rangle$, followed by a second single-quantum jump to a final level $|f\rangle$, and (ii) via a two-quantum transition to a final level $|f\rangle$. Cascade processes $|i\rangle \rightarrow |\bar{m}\rangle \rightarrow |f\rangle$ and two-quantum transitions $|i\rangle \rightarrow |m\rangle \rightarrow |f\rangle$ compete (in the latter case, $|m\rangle$ can be both a nonresonant as well as a resonant level). General expressions are derived within second-order time-dependent perturbation theory, and the cascade contribution is identified. When the one-quantum decay rates of the virtual states are included into the complex resonance energies that enter the propagator denominator, it is found that the second-order decay rate contains the one-quantum decay rate of the initial state as a lower-order term. For atomic transitions, this implies that the differential-in-energy two-photon transition rate with complex resonance energies in the propagator denominators can be used to good accuracy even in the vicinity of resonance poles.

DOI: 10.1103/PhysRevA.81.012112

PACS number(s): 12.20.Ds, 31.30.jc

I. INTRODUCTION

In this article, we consider quite a general problem which is illustrated on the basis of the radiative decay of excited atomic levels. Let us suppose that an initial state $|i\rangle$ of a quantum system can decay into a final state $|f\rangle$ via an intermediate, virtual state $|m\rangle$, under the influence of an interaction potential V , with relevant matrix elements V_{fm} and V_{mi} . If all available levels $|m\rangle$ are nonresonant, then the decay rate can be computed using time-ordered second-order perturbation theory [1]. One famous example is the decay of the $2S$ state of hydrogen, whose decay to the ground state is dipole-forbidden. Nevertheless, the main contribution to the $2S$ state decays to the ground state is caused by the very *electric dipole* coupling of the bound electron to the quantized electromagnetic field: the transition proceeds via virtual nP levels ($n \geq 2$), which are all nonresonant in the nonrelativistic approximation. One has to formulate the problem in second-order as opposed to first-order time-dependent perturbation theory.

Cascade decay accompanying the process $|i\rangle \rightarrow |m\rangle \rightarrow |f\rangle$ can proceed when some of the available virtual levels $|\bar{m}\rangle \in \{|m\rangle\}$ are resonant. In that case, the atom may first undergo a transition $|i\rangle \rightarrow |\bar{m}\rangle$, then $|\bar{m}\rangle \rightarrow |f\rangle$ (cascade decay). An example is the decay $3S \rightarrow nP \rightarrow 1S$ in atomic hydrogen, where the atom may first radiate a photon at the resonant frequency of the $3S \rightarrow 2P$ transition, and then radiate a second photon at the resonant frequency of the $2P \rightarrow 1S$ transition. However, the transition $3S \rightarrow nP \rightarrow 1S$ may also proceed via a nonresonant nP level, in which case it is a true two-photon (two-quantum) transition. Indeed, the second-order transition amplitude for the sum of the processes $3S \rightarrow nP \rightarrow 1S$ (n being summed over) contains both the transition amplitude due to nonresonant virtual states as well as the transition amplitude due to resonant intermediate states. Strictly speaking, the situation is even a little more complicated: the electric-dipole coupling of the atom with the radiation field couples a state with the atom in the $3S$ state to a combined atom + field state with the atom in the $|nP\rangle$ state and one photon in the radiation field. We can denote this state as $|m\rangle = |nP, 1_{\vec{k}\lambda}\rangle$ for the particular transition

mentioned. Here, \vec{k} is the photon wave vector, and λ is its polarization. Unless simultaneously $n = 2$ and the photon fulfills the resonance condition $E_{3S} - E_{2P} = \hbar ck$, where $k \equiv |\vec{k}|$ is the wave number of the photon, the intermediate state $|m\rangle$ is nonresonant. An intermediate level $|m\rangle = |2P, 1_{\vec{k}\lambda}\rangle$ with $E_{3S} - E_{2P} \neq \hbar ck'$ constitutes a nonresonant level even if the atomic part of the intermediate state—the $2P$ level—can become resonant. To give another example, an intermediate state $|m\rangle = |4P, 1_{\vec{k}\lambda}\rangle$ with arbitrary \vec{k} is always nonresonant because there is no photon frequency available which could turn this level into a resonant state. The question then is how to separate the decay through resonant intermediate states from the decay via nonresonant intermediate states. Certainly, it is impossible to do this by excluding the $|2P\rangle$ level from the sum over the intermediate atomic levels, because this level can be both resonant (if the photon frequency in the intermediate state is resonant with respect to the $3S \rightarrow 2P$ transition) or nonresonant (if the photon frequency in the intermediate state is nonresonant with respect to the $3S \rightarrow 2P$ transition). The exclusion of the $2P$ state had been proposed in Ref. [2] but has since been scrutinized [3,4].

Related questions are investigated here in more general terms: How can we formulate the problem, within time-dependent second-order perturbation theory, so that the resonant intermediate levels in the process $|i\rangle \rightarrow |m\rangle \rightarrow |f\rangle$ are separated from the nonresonant levels, and so that the cascade contribution due to resonant intermediate levels $|\bar{m}\rangle$ is clearly identified within the time-dependent formalism? In order to answer this question, we first recall that under rather general assumptions about the process, the intermediate states $|m\rangle$ represent a *continuum* of states. This is the case even in transitions of *discrete* atomic levels because the intermediate states $|m\rangle$ in this case are product states of the atom in a discrete state and one or more excited modes of the electromagnetic field. While the bound states of the atom are discrete, the photon modes represent a continuum of energies. In particular, the photon wave vector \vec{k} represents a continuous variable. A resonant process involves a transition to a lower atomic level with a simultaneous emission of a photon of the resonant frequency; in that case, the resonant state $|\bar{m}\rangle$ is an eigenstate

of the unperturbed Hamiltonian of atom + radiation field with exactly the same energy as the initial state (the sum of the energies of the lower atomic state and of the energy of the radiated photon is equal to the energy of the initial atomic state).

When the decay $|i\rangle \rightarrow |m\rangle \rightarrow |f\rangle$ can proceed via a resonant state $|\bar{m}\rangle$ which can be reached from $|i\rangle$ via a single quantum jump, we have to take into account both possibilities: (a) the sequential transition (cascade) and (b) the two-quantum transition via the nonresonant levels. One possibility to identify the cascade within time-dependent perturbation theory is given by the functional form of its time dependence: for a cascade decay $|i\rangle \rightarrow |\bar{m}\rangle \rightarrow |f\rangle$, the probability of finding the system in the final state $|f\rangle$ is proportional to the square of the elapsed time t : The system first has to make a quantum jump $|i\rangle \rightarrow |\bar{m}\rangle$, leading to a linear increase (with time) of the population of the resonant intermediate level $|\bar{m}\rangle$. The second quantum jump $|\bar{m}\rangle \rightarrow |f\rangle$ then leads to a quadratic increase of the probability of finding the system in state $|f\rangle$ with time. By contrast, the true nonsequential two-quantum transition $|i\rangle \rightarrow |m\rangle \rightarrow |f\rangle$ via nonresonant intermediate states leads to a linear increase (with time) of the probability of finding the system in state $|f\rangle$ with time. Here, we identify, in a general formalism, those contributions of the two-quanta transition which contribute to the linear behavior (in time), and separate them from the (quadratic in time) cascade effect.

We follow Ref. [1] in our conventions and proceed as follows: First, the basics of a single-quantum transition are recalled (Sec. II). We then proceed to the discussion of a two-quantum transition without cascades (Sec. III), before including the cascades/resonant levels in Sec. IV. Conclusions are reserved for Sec. V. The interaction is switched off adiabatically in the distant past and in the distant future, but the rate is calculated near $t = 0$. We work in natural units ($\hbar = c = \epsilon_0 = 1$).

II. SINGLE-QUANTUM TRANSITION

A. General formulation

Following Chap. 5 of Ref. [1], we first consider a single-quantum transition $|i\rangle$ to $|f\rangle$, with $c_f(t)$ being the time-dependent expansion coefficient of the final-state Hilbert vector with respect to the state $|f\rangle$. The interaction is adiabatically damped on in the infinite past $t \rightarrow -\infty$ and suppressed by an exponential factor $\exp(\eta t)$, with $\eta > 0$ being an infinitesimal parameter. We then start the time evolution with $c_f(0) = 0$ and $c_i(0) = 1$ (initially, the system is in the state $|i\rangle$). For the complex probability amplitude $c_f(t)$ of finding the system in state $|f\rangle$ at time t , one finds [see Eq. (5.8.2) of Ref. [1]],

$$c_f(t) = -i \int_{-\infty}^t V_{fi} e^{\eta t'} e^{i\omega_{fi} t'} dt' = -\frac{e^{\eta t + i\omega_{fi} t}}{\omega_{fi} - i\eta} V_{fi}, \quad (1)$$

where V_{fi} is the matrix element of the interaction Hamiltonian V in the Schrödinger picture, i.e., $V_{fi} = \langle f|V|i\rangle$. Note that there is a somewhat subtle difference between the interaction Hamiltonian V in the Schrödinger picture, and the interaction Hamiltonian $\exp(iH_0 t)V\exp(-iH_0 t)$ in the interaction picture, because in the latter case, matrix elements of V acquire

a time dependence. This time dependence is explicitly written out in the term $V_{fi} e^{i\omega_{fi} t'}$ in Eq. (1).

In the case of an electric-dipole transition in an atom, V is the coupling of the bound electron to the quantized radiation field. The expression $\omega_{fi} = E_f - E_i$ is the energy difference of the initial and final state of the transition with respect to the unperturbed Hamiltonian H_0 of the system. In the case of an electric-dipole transition in an atom, H_0 is the sum of the unperturbed Hamiltonian of the atom and of the electromagnetic Hamiltonian counting the modes of the radiation field. From Eq. (1), we find $|c_f(t)|^2 = e^{2\eta t} |V_{fi}|^2 / (\omega_{fi}^2 + \eta^2)$. Differentiating this expression with respect to time, we obtain

$$\frac{d}{dt} |c_f(t)|^2 = \frac{e^{2\eta t} 2\eta |V_{fi}|^2}{\omega_{fi}^2 + \eta^2}. \quad (2)$$

With the identification [see Eq. (5.8.5) of Ref. [1]]

$$\frac{\eta}{\omega_{fi}^2 + \eta^2} \rightarrow \pi \delta(\omega_{fi}), \quad \eta \rightarrow 0^+, \quad (3)$$

we obtain in the limit $\eta \rightarrow 0^+$

$$\Gamma_{fi}^{(1)} = \left. \frac{d}{dt} |c_f(t)|^2 \right|_{t=0} = 2\pi |V_{fi}|^2 \delta(\omega_{fi}), \quad (4)$$

where by definition, $\Gamma_{fi}^{(1)}$ is the decay rate associated with the transition $|i\rangle \rightarrow |f\rangle$ via a single quantum jump (we reemphasize that the time derivative is taken at $t = 0$). This result is known as Fermi's golden rule [see Eqs. (5.6.35) and (5.8.6) of [1]].

One might wonder why the Dirac δ persists in the final result, although Fermi's Golden Rule is known to be directly applicable to experimentally relevant calculations, and an expression containing a Dirac δ might otherwise be assumed not to be applicable to an experiment. Just after Eq. (5.6.35) of Ref. [1], which is equivalent to Eq. (4) in this work, it is stated that the final state must be integrated over an (infinitesimal) interval about the final-state energy. This statement is useful, but it may need a more complex explanation for a full elucidation. Indeed, the solution to this question involves two observations: (a) that Eq. (4) needs to be summed over the state variables of the radiated quanta (in the case of an atomic transition, photons) in order to make experimentally relevant predictions, and (b) that the Dirac δ disappears when all possible energies and all possible polarizations of the emitted quanta are taken into account in the final state. In order to illustrate this aspect, we now discuss the application of Eq. (4) to an electric dipole transition in an atom.

B. Specialization to an atomic transition

In the case of an electric-dipole transition of an atom, the final state $|f\rangle$ is a product state of the atom in state $|f_A\rangle$ and one radiated photon $|1_{\vec{k}\lambda}\rangle$ in the radiation field. In the following, we will write a general product state $|f\rangle$ of the system composed of the atom + radiation field as

$$|f\rangle = |f_A, \vec{f}\rangle, \quad (5)$$

where $|f_A\rangle$ is atomic part of the product state, and $|\vec{f}\rangle$ is the photon part of the product state. The unperturbed Hamiltonian

of the system is

$$H_0 = \sum_{f_A} E_{f_A} |f_A\rangle \langle f_A| + \sum_{\vec{k}\lambda} k a_{\vec{k}\lambda}^+ a_{\vec{k}\lambda}, \quad (6)$$

where the $a_{\vec{k}\lambda}$ and $a_{\vec{k}\lambda}^+$ are photon annihilation and creation operators (here, we work in a representation with a finite normalization volume \mathcal{V} , i.e., $[a_{\vec{k}\lambda}, a_{\vec{k}\lambda}^+] = \delta_{\vec{k}\vec{k}'} \delta_{\lambda\lambda'}$). Eigenstates of the Hamiltonian (6) are product states of the atom in eigenstate $|f_A\rangle$ and a Fock state of the electromagnetic field such as $|1_{\vec{k}\lambda}\rangle$.

In the Schrödinger picture, the dipole interaction of an electron at point \vec{x} with the quantized electromagnetic field is given by

$$V = -e\vec{x} \cdot \vec{E}, \quad (7)$$

$$\vec{E} = \sum_{\vec{k}\lambda} \sqrt{\frac{k}{2\mathcal{V}}} (\hat{\epsilon}_{\vec{k}\lambda} a_{\vec{k}\lambda} + \hat{\epsilon}_{\vec{k}\lambda}^+ a_{\vec{k}\lambda}^+),$$

with the unit polarization vectors $\hat{\epsilon}_{\vec{k}\lambda}$ and the electric field operator \vec{E} . In atomic physics, one distinguishes between the $(\vec{p} \cdot \vec{A})$ and $(\vec{x} \cdot \vec{E})$ forms of the interaction with the electromagnetic field. The former is called the velocity gauge because of the appearance of the electron momentum in the interaction Hamiltonian. The latter is commonly referred to as the length gauge, because the electron coordinate \vec{x} in the interaction Hamiltonian has physical dimension of length. In some situations, the length gauge is preferable because the interaction is formulated in terms of physically observable electric field strength \vec{E} instead of the gauge-dependent vector potential \vec{A} (see Refs. [5–7]). All results presented here are given in the length gauge.

While the initial state of the atom $|i_A\rangle$ and the final state of the atom $|f_A\rangle$ are well-defined for an atomic decay rate, we have to sum over the degrees of freedom of the radiated photons in order to obtain the decay rate for the one-photon transition $|i_A\rangle \rightarrow |f_A\rangle$. The Dirac δ function in Eq. (4) ensures the fulfillment of the resonance condition. For a one-photon final state, we can replace

$$\sum_{\vec{f}} \rightarrow \sum_{\vec{k}\lambda} \quad (8)$$

for the sum over the photon degrees of freedom of the final state. Indeed, the atomic one-photon (1γ) decay rate for the transition $|i_A\rangle \rightarrow |f_A\rangle$, which we denote as $\Gamma_{f_A i_A}^{(1\gamma)}$, is obtained as the sum

$$\Gamma_{f_A i_A}^{(1\gamma)} = \sum_{\vec{f}} \Gamma_{f_i}^{(1)} = \sum_{\vec{k}\lambda} \Gamma_{f_i}^{(1)} = \sum_{\vec{k}\lambda} \Gamma_{|f_A, 1_{\vec{k}\lambda}\rangle, |i_A, 0\rangle}^{(1)}$$

$$= 2\pi \sum_{\vec{k}\lambda} \delta(E_{f_A} - E_{i_A} - k) |\langle f_A, 1_{\vec{k}\lambda} | V | i_A, 0 \rangle|^2, \quad (9)$$

where we recall that the sum over \vec{k} and λ transforms into an integral in the continuum limit,

$$\sum_{\vec{k}\lambda} \rightarrow \mathcal{V} \sum_{\lambda} \int \frac{d^3k}{(2\pi)^3}. \quad (10)$$

This integral cancels the Dirac δ . We reemphasize that the Dirac- δ function is eliminated after a summation over specific

degrees of freedom of the final state, namely, the degrees of freedom of the electromagnetic field.

One might wonder why the single-quantum transitions apparently conserve energy according to the above formalism [persistence of the $\delta(\omega_{fi})$ in Eq. (4)], while spontaneous decay of an atomic state always tends to lower the energy of the bound electron. The answer is that the final state of the process, which is a bound electron in a lower state plus a single resonant photon, has the same energy as the initial state (electron in the excited state and no photon in the radiation field). This is manifest in the expression $\delta(\omega_{fi}) = \delta(E_{f_A} - E_{i_A} - k)$ in Eq. (9).

In view of Eq. (7), the transition matrix element $\langle f_A, 1_{\vec{k}\lambda} | V | i_A, 0 \rangle$ in Eq. (9) can be written as

$$\langle f_A, 1_{\vec{k}\lambda} | (-e\vec{x} \cdot \vec{E}) | i_A, 0 \rangle = -e \sqrt{\frac{k}{2\mathcal{V}}} \hat{\epsilon}_{\vec{k}\lambda} \cdot \langle f_A | \vec{x} | i_A \rangle, \quad (11)$$

where we denote the atomic component of the bra and ket vectors by a subscript A . The sum over the photon modes in Eq. (7) collapses because there is exactly one definite photon mode occupied in the state $|f_A, 1_{\vec{k}\lambda}\rangle$. Summing over the available photon modes in the exit channel, we obtain ($k \equiv |\vec{k}|$)

$$\Gamma_{f_A i_A}^{(1\gamma)} = \sum_{\vec{k}\lambda} 2\pi |V_{fi}|^2 \delta(\omega_{fi})$$

$$= \sum_{\vec{k}\lambda} 2\pi e^2 \frac{k}{2\mathcal{V}} |\hat{\epsilon}_{\vec{k}\lambda} \cdot \langle f_A | \vec{x} | i_A \rangle|^2 \delta(E_{f_A} - E_{i_A} - \omega_{\vec{k}})$$

$$= \sum_{\lambda} \int \frac{d^3k}{(2\pi)^3} 4\pi^2 \alpha k |\hat{\epsilon}_{\vec{k}\lambda} \cdot \langle f_A | \vec{x} | i_A \rangle|^2$$

$$\times \delta(E_{f_A} - E_{i_A} - \omega_{\vec{k}})$$

$$= \int \frac{d\Omega_k}{4\pi} 2\alpha (E_{f_A} - E_{i_A})^3 \delta^{T,ij} \langle f_A | x^j | i_A \rangle \langle i_A | x^k | f_A \rangle$$

$$= \frac{4}{3} \alpha (E_f - E_i)^3 |\langle f_A | \vec{x} | i_A \rangle|^2, \quad (12)$$

which is the familiar result for a one-photon electric-dipole decay rate. The transverse delta function is $\delta^{T,ij} = \delta^{ij} - k^i k^j / k^2$. We denote the Cartesian components of a vector by superscripts. Note, in particular, that the sum over the photon modes in Eq. (7) is not enough in order to calculate the familiar expression for the one-photon decay rate; an additional summation over final states is necessary.

III. TWO-QUANTUM TRANSITION WITHOUT CASCADES

A. General formulation

In second-order time-dependent perturbation theory, the amplitude $c_f(t)$ to find the system in state $|f\rangle$ at time t due to the transition $|i\rangle \rightarrow |m\rangle \rightarrow |f\rangle$ is given by

$$c_f(t) = (-i)^2 \int_{-\infty}^t dt' e^{i\eta t' + i\omega_{fm} t'} V_{fm} \int_{-\infty}^{t'} dt'' e^{i\eta t'' + i\omega_{mi} t''} V_{mi}, \quad (13)$$

which leads to [see Eq. (5.6.37) of Ref. [1]],

$$|c_f(t)|^2 = \frac{e^{4\eta t}}{(4\eta^2 + \omega_{fi}^2)} \left| \sum_m \frac{V_{fm} V_{mi}}{\omega_{mi} - i\eta} \right|^2. \quad (14)$$

This is a generalization of Eq. (1) to second order. When no cascades are allowed, we can differentiate with respect to time and assume that $\omega_{mi} \neq 0$ is always nonvanishing. In order to fix ideas by comparison to a concrete example, we recall that in the case of the $2S \rightarrow 1S$ two-photon transition in atomic hydrogen, the intermediate state $|m\rangle = |nP, 1_{\vec{k}\lambda}\rangle$ has a higher energy than the initial state $|i\rangle = |2S, 0\rangle$ where the atom is in the $2S$ state and the electromagnetic field is in the vacuum state $|0\rangle$. No cascades are relevant in this case, and Eq. (14) is immediately applicable.

We can thus differentiate Eq. (13) with respect to time and obtain

$$\begin{aligned} \Gamma_{fi}^{(2)} &= \left(\frac{d}{dt} |c_f|^2 \right) \Big|_{t=0} \\ &= \frac{4\eta}{(2\eta)^2 + \omega_{fi}^2} \left| \sum_m \frac{V_{fm} V_{mi}}{\omega_{mi} - i\eta} \right|^2 \\ &= 2\pi \delta(\omega_{fi}) \left| \sum_m \frac{V_{fm} V_{mi}}{\omega_{mi}} \right|^2, \quad \eta \rightarrow 0^+. \end{aligned} \quad (15)$$

In analogy to the single-quantum transition described by Eq. (4), the Dirac δ disappears when the final states are summed over the experimentally relevant degrees of the radiated quanta. We now verify that Eq. (15) exactly reproduces the known expressions [8,9] for two-photon decay rates in atoms.

B. Specialization to an atomic transition

For a two-photon transition in an atom, we can write the initial state as $|i\rangle = |i_A, 0\rangle$, where $|i_A\rangle$ is the atomic final state, and $|0\rangle$ is the vacuum state of the electromagnetic field. The intermediate state is $|m\rangle = |m_A, 1_{\vec{k}\lambda}\rangle$, where the atom is in state $|m_A\rangle$, and the electromagnetic field is in the one-photon Fock state $|1_{\vec{k}\lambda}\rangle$. The final state is $|f\rangle = |f_A, 1_{\vec{k}_1\lambda_1}, 1_{\vec{k}_2\lambda_2}\rangle$, where the $|\vec{k}_i|$ and λ_i are the wave vectors and polarizations of the two radiated photons ($i = 1, 2$).

In specializing Eq. (15) to a two-photon transition in atoms, we have to take into account a subtlety, which we outline in greater detail because it becomes relevant for all discussions in the following. Namely, the atomic decay rate is obtained after summing the rate $\Gamma_{fi}^{(2)}$ over the degrees of freedom of all possible radiated photons. Now, if we sum the final states over all $\vec{k}_1\lambda_1$ and all $\vec{k}_2\lambda_2$, we count the photons twice, because the Fock state $|1_{\vec{k}_2\lambda_2}, 1_{\vec{k}_1\lambda_1}\rangle$ obtained under the simultaneous exchange $\vec{k}_1 \leftrightarrow \vec{k}_2$ and $\lambda_1 \leftrightarrow \lambda_2$ is identical to the original state $|1_{\vec{k}_1\lambda_1}, 1_{\vec{k}_2\lambda_2}\rangle$. Hence,

$$\Gamma_{f_A i_A}^{(2\gamma)} = \frac{1}{2} \sum_{\vec{k}_1\lambda_1} \sum_{\vec{k}_2\lambda_2} \Gamma_{fi}^{(2)}. \quad (16)$$

The factor 1/2 is discussed after Eq. (5.108) on p. 169 of the quantum field theory textbook [10] and in the text preceding Eq. (3.316) of the textbook [11].

With reference to Eq. (15), we now turn our attention to the two-quantum decay rate (without cascades). Here, two quantum paths are possible which must be added coherently. These correspond to a different time ordering for the emissions of the photons with photon wave vector \vec{k}_i and polarization λ_i

($i = 1, 2$). Summing over the final-state photon polarizations, the result then is

$$\begin{aligned} \Gamma_{f_A i_A}^{(2\gamma)} &= \frac{1}{2} \sum_{\vec{k}_1\lambda_1} \sum_{\vec{k}_2\lambda_2} 2\pi \delta(\omega_{fi}) \left| \sum_m \frac{V_{fm} V_{mi}}{\omega_{mi}} \right|^2, \\ &= \sum_{\vec{k}_1\lambda_1} \sum_{\vec{k}_2\lambda_2} \pi e^4 \frac{k_1}{2\mathcal{V}} \frac{k_2}{2\mathcal{V}} \delta(E_f - E_i - k_1 - k_2) \\ &\quad \times \left| \sum_{m_A} \left(\frac{(\hat{\epsilon}_{\vec{k}_1\lambda_1} \cdot \langle f_A | \vec{x} | m_A \rangle)(\hat{\epsilon}_{\vec{k}_2\lambda_2} \cdot \langle m_A | \vec{x} | i_A \rangle)}{E_{m_A} - E_{i_A} + k_2} \right. \right. \\ &\quad \left. \left. + \frac{(\hat{\epsilon}_{\vec{k}_2\lambda_2} \cdot \langle f_A | \vec{x} | m_A \rangle)(\hat{\epsilon}_{\vec{k}_1\lambda_1} \cdot \langle m_A | \vec{x} | i_A \rangle)}{E_{m_A} - E_{i_A} + k_1} \right) \right|^2. \end{aligned} \quad (17)$$

Separating angular and radial variables for the photon energies, we finally obtain the following known result [8] in the continuum limit [see Eq. (10)]:

$$\begin{aligned} \Gamma_{f_A i_A}^{(2\gamma)} &= \frac{4\alpha^2}{27\pi} \int_0^{E_{f_A} - E_{i_A}} dk k^3 (E_{f_A} - E_{i_A} - k)^3 \\ &\quad \times \left| \sum_{m_A} \left(\frac{\langle f_A | x^j | m_A \rangle \langle m_A | x^j | i_A \rangle}{E_{m_A} - E_{i_A} + k} \right. \right. \\ &\quad \left. \left. + \frac{\langle f_A | x^j | m_A \rangle \langle m_A | x^j | i_A \rangle}{E_{m_A} - E_{f_A} - k} \right) \right|^2. \end{aligned} \quad (18)$$

The integration over k extends over the allowed frequency range for a two-photon transition [12]. The subtlety with respect to the counting of photon modes illustrates that Eq. (15) cannot be applied to atomic transitions without a proper interpretation of all physical quantities involved.

IV. TWO-QUANTUM TRANSITION WITH CASCADES

A. General formulation

We return once more to Eq. (14) which gives the result for the two-photon decay rate [see also Eq. (5.6.37) of Ref. [1]],

$$|c_f(t)|^2 = \frac{e^{4\eta t}}{(4\eta^2 + \omega_{fi}^2)} \left| \sum_m \frac{V_{fm} V_{mi}}{\omega_{mi} - i\eta} \right|^2. \quad (19)$$

In the text directly following Eq. (5.6.37) of Ref. [1], it is stated that the best way to deal with the situation of a resonant intermediate state with $\omega_{mi} \approx 0$ is to use an adiabatic turn-on of the perturbation that leads to the transition. We have already incorporated this adiabatic turn-on into Eq. (13). It is also stated in Eq. (5.6.38) of Ref. [1] that the turn-on amounts to the replacement

$$\omega_{mi} \rightarrow \omega_{mi} - i\eta \quad (20)$$

in the denominator of the expression on the right-hand side of Eq. (19). Again, we have already incorporated the infinitesimal imaginary part in Eq. (19). Here, we extend the discussion beyond that in Ref. [1] and analyze the resonant and nonresonant levels separately.

We now assume that some of the intermediate states of the system are close in energy to the initial state of the process, i.e., that there exist states $|\bar{m}\rangle$ with $E_{\bar{m}} = E_i$. We

recall that $E_{\bar{m}}$ here represents the total energy if the system. In the case of an atomic transition, this would be the sum of the energy of the intermediate atomic level and of energy of the photons radiated. In order to analyze this process, we restrict, in Eq. (19), the sum over intermediate states to the resonant states \bar{m} . Then,

$$|\bar{c}_f(t)|^2 = \frac{e^{4\eta t}}{(2\eta)^2 + \omega_{fi}^2} \left| \sum_{\bar{m}} \frac{V_{f\bar{m}} V_{\bar{m}i}}{\omega_{\bar{m}i} - i\eta} \right|^2, \quad (21)$$

where $\omega_{\bar{m}i}$ tends to zero. The cascade contribution associated with the resonant levels $|\bar{m}\rangle$ needs to be differentiated twice with respect to the time. We obtain

$$\begin{aligned} \left(\frac{d^2}{dt^2} |\bar{c}_f|^2 \right) \Big|_{t=0} &= \frac{16\eta^2}{(2\eta)^2 + \omega_{fi}^2} \left| \sum_{\bar{m}} \frac{V_{f\bar{m}} V_{\bar{m}i}}{\omega_{\bar{m}i} - i\eta} \right|^2 \\ &\stackrel{\eta \rightarrow 0^+}{=} 4\pi^2 \sum_{\bar{m}} |V_{f\bar{m}} V_{\bar{m}i}|^2 \delta(\omega_{f\bar{m}}) \delta(\omega_{fi}) \\ &= 4\pi^2 \sum_{\bar{m}} |V_{f\bar{m}} V_{\bar{m}i}|^2 \delta(\omega_{f\bar{m}}) \delta(\omega_{\bar{m}i}) \\ &= \sum_{\bar{m}} \Gamma_{f\bar{m}}^{(1)} \Gamma_{\bar{m}i}^{(1)} \equiv C_{fi}. \end{aligned} \quad (22)$$

In the last step, we define the expression C_{fi} as the relevant cascade term which we evaluate for atomic transitions in Sec. IV B below. We also assume that interference terms among the different resonant levels $|\bar{m}\rangle$ vanish.

Equation (22) is just the expected result: the level $|i\rangle$ feeds the resonant intermediate levels $|\bar{m}\rangle$ with a time dependence $\Gamma_{\bar{m}i}^{(1)} t$, and the resonant intermediate levels, in turn, feed the final state population as

$$|\bar{c}_f(t)|^2 = \sum_{\bar{m}} \int_0^t dt' \Gamma_{f\bar{m}}^{(1)} \Gamma_{\bar{m}i}^{(1)} t' = \frac{1}{2} \sum_{\bar{m}} \Gamma_{f\bar{m}}^{(1)} \Gamma_{\bar{m}i}^{(1)} t^2. \quad (23)$$

The necessity of the sum over \bar{m} is also clear, because all intermediate resonant levels have to be included.

Now that we have treated the resonant levels separately, we have to subtract them from the remaining expression. We thereby obtain a modified probability $|c_f(t)|^2$ of finding the system in state $|f\rangle$,

$$\begin{aligned} |c_f(t)|^2 &= |c_f(t)|^2 - |\bar{c}_f(t)|^2 \\ &= \frac{e^{4\eta t}}{(4\eta^2 + \omega_{fi}^2)} \left| \sum_m \frac{V_{fm} V_{mi}}{\omega_{mi} - i\eta} \right|^2 \\ &\quad - \frac{e^{4\eta t}}{(4\eta^2 + \omega_{fi}^2)} \left| \sum_{\bar{m}} \frac{V_{f\bar{m}} V_{\bar{m}i}}{\omega_{\bar{m}i} - i\eta} \right|^2. \end{aligned} \quad (24)$$

One might think that the subtraction term (second term on the right-hand side of the above equation) would imply, e.g., the subtraction of the intermediate $2P$ state in the two-photon decay of the $3S$ state of hydrogen. However, that is not the case. The intermediate states are quantum states of the coupled system of atom + radiation field. As already outlined in Sec. I, the product state composed of the $2P$ level and a resonant photon would qualify as a resonant state $|\bar{m}\rangle$, but a $2P$ state with a slightly off-resonant photon would not constitute a

resonant intermediate state. Therefore, the $2P$ state may not be taken out of the sum over the atomic-state components of the virtual states. The time derivative of the subtracted expression $|c_f|^2$ is

$$\begin{aligned} \left(\frac{d}{dt} |c_f|^2 \right) \Big|_{t=0} &= \frac{4\eta}{(4\eta^2 + \omega_{fi}^2)} \left| \sum_m \frac{V_{fm} V_{mi}}{\omega_{mi} - i\eta} \right|^2 \\ &\quad - \frac{4\eta}{(4\eta^2 + \omega_{fi}^2)} \left| \sum_{\bar{m}} \frac{V_{f\bar{m}} V_{\bar{m}i}}{\omega_{\bar{m}i} - i\eta} \right|^2. \end{aligned} \quad (25)$$

The second term on the right-hand side of Eq. (25) is divergent in the limit $\eta \rightarrow 0^+$ and $\omega_{\bar{m}i} \rightarrow 0$. We cannot proceed without giving a physical interpretation to the adiabatic parameter η . First, in the subtraction term

$$S = -\frac{4\eta}{(2\eta)^2 + \omega_{fi}^2} \left| \sum_{\bar{m}} \frac{V_{f\bar{m}} V_{\bar{m}i}}{\omega_{\bar{m}i} - i\eta} \right|^2, \quad (26)$$

we carry out the limit $\eta \rightarrow 0^+$ in the prefactor; this leads to a Dirac δ . Then, for the sum over \bar{m} , we match the adiabatic parameter with the imaginary part of the interaction Hamiltonian. The in and out states $|i\rangle$ and $|f\rangle$ are assumed to be asymptotic, stable states in the infinite past and future within the context of adiabatic perturbation theory. Adiabatically, we therefore switch on only the virtual intermediate states. We should thus replace

$$\eta \rightarrow \frac{1}{2} \Gamma_{fm}^{(1)} \quad (27)$$

for every term in the sum. We then obtain

$$\begin{aligned} S &= -2\pi \delta(\omega_{fi}) \left| \sum_{\bar{m}} \frac{V_{f\bar{m}} V_{\bar{m}i}}{\omega_{\bar{m}i} - i \frac{1}{2} \Gamma_{f\bar{m}}^{(1)}} \right|^2 \\ &= -2\pi \delta(\omega_{fi}) \sum_{\bar{m}} \frac{|V_{f\bar{m}} V_{\bar{m}i}|^2}{\omega_{\bar{m}i}^2 + \left(\frac{1}{2} \Gamma_{f\bar{m}}^{(1)} \right)^2} \\ &= -2\pi \delta(\omega_{fi}) \sum_{\bar{m}} \frac{2 \frac{1}{2} \Gamma_{f\bar{m}}^{(1)} |V_{f\bar{m}} V_{\bar{m}i}|^2}{\Gamma_{f\bar{m}}^{(1)} \omega_{\bar{m}i}^2 + \left(\frac{1}{2} \Gamma_{f\bar{m}}^{(1)} \right)^2} \\ &= -4\pi^2 \delta(\omega_{fi}) \sum_{\bar{m}} \frac{1}{\Gamma_{f\bar{m}}^{(1)}} |V_{f\bar{m}} V_{\bar{m}i}|^2 \delta(\omega_{\bar{m}i}) \\ &= -4\pi^2 \delta(\omega_{fi}) \sum_{\bar{m}} \frac{|V_{f\bar{m}}|^2 |V_{\bar{m}i}|^2}{\Gamma_{f\bar{m}}^{(1)}} \delta(\omega_{f\bar{m}}) \\ &= -2\pi \delta(\omega_{\bar{m}i}) \sum_{\bar{m}} |V_{\bar{m}i}|^2 = -\sum_{\bar{m}} \Gamma_{\bar{m}i}^{(1)}. \end{aligned} \quad (28)$$

In going from the fourth to the fifth line of the above equation, we have neglected interference terms. This deserves some comments, which we give by way of example. Let us consider a situation with an initial $4S$ state without any photons, and resonant $2P$ and $3P$ virtual states (each endowed with a single resonant photon), and a $1S$ final state (with two resonant photons). A conceivable $2P$ - $3P$ interference term would necessitate the final states $|f\rangle$ to be equivalent in regards to both their atomic components as well as electromagnetic-field components. However, because the emitted resonant photons

for $4S \rightarrow 3P \rightarrow 1S$ have different energy as compared to $4S \rightarrow 2P \rightarrow 1S$, the interference term vanishes.

The derivation (28) clarifies that the subtraction term is nothing but the sum of the one-quantum decay rates of the initial state to all accessible resonant intermediate states. The result coincides with the lower-order subtraction term found in Ref. [12] for the two-photon decay rate, but the above derivation is much more general. It means that under this regularization, the two-quantum correction to the decay rate is obtained as

$$\begin{aligned} \overline{\Gamma_{fi}^{(2)}} &= \left(\frac{d}{dt} |c_f|^2 \right) \Big|_{t=0} \\ &= 2\pi\delta(\omega_{fi}) \left| \sum_m \frac{V_{fm} V_{mi}}{\omega_{mi} - \frac{1}{2}i\Gamma_{fm}^{(1)}} \right|^2 - \sum_m \overline{\Gamma_{mi}^{(1)}}, \end{aligned} \quad (29)$$

where we introduce the overlining in order to differentiate $\overline{\Gamma_{fi}^{(2)}}$ from $\Gamma_{fi}^{(2)}$. The result (29) is well defined and gauge invariant [12]. We also note that the total (one-quantum plus two-quantum) decay rate of level $|i\rangle$ thus is

$$\Gamma_i = \sum_m \overline{\Gamma_{mi}^{(1)}} + \overline{\Gamma_{fi}^{(2)}} = 2\pi\delta(\omega_{fi}) \left| \sum_m \frac{V_{fm} V_{mi}}{\omega_{mi} - \frac{1}{2}i\Gamma_{fm}^{(1)}} \right|^2. \quad (30)$$

This result states, in general terms, that the expression for the two-quantum decay rate, in the presence of allowed cascade transitions and with propagator denominators regularized by the total one-quantum decay rate, has to be interpreted as a one+two quantum decay rate.

B. Specialization to an atomic transition

In view of the result given in Eq. (18), it is immediately clear how to apply Eq. (30) to two-photon transitions in atoms. Namely, when Eq. (30) is evaluated for two-photon transitions, the correct result is obtained when the virtual-state energies in formula (18) are regularized by their total one-photon decay widths [see Eq. (1) of Ref. [12] for a concrete example and extensive further discussion in Refs. [12–15]]. For an atomic two-photon transition, $\overline{\Gamma_{fi}^{(2)}}$ as written in Eq. (29) coincides with the imaginary part of the two-loop self-energy due to cut diagrams with two-photon emission [12,16], with the photons fulfilling the two-photon resonance condition $k_1 + k_2 = E_{f_A} - E_{i_A}$.

It has been shown in Refs. [12,16] that $\overline{\Gamma_{fi}^{(2)}}$ is of the order of $\alpha^2(Z\alpha)^6$ in units of the electron rest mass energy, and is thus of the same order as the result obtained for two-photon transitions without cascades [8]. It is therefore appropriate to refer to $\overline{\Gamma_{fi}^{(2)}}$ as a two-photon *correction* to the decay rate of an initial state which otherwise decays via one-photon decay. For completeness, we note that the two terms on the right-hand side of Eq. (29) are both of order $\alpha^2(Z\alpha)^4$, but their difference is of order $\alpha^2(Z\alpha)^6$ and thus smaller by two orders of $Z\alpha$.

The only calculation remaining concerns the verification of the fact that Eq. (22) reproduces the product of one-photon decay rates for the cascade process $|i_A\rangle \rightarrow |m_A\rangle \rightarrow |f_A\rangle$. We use Eq. (16) in order to sum over the two-photon final states and

Eq. (10) in order to proceed to the continuum limit. Summing the cascade term (22) over the degrees of freedom of the emitted photons, we obtain

$$\begin{aligned} C_{f_A i_A} &= \left(\frac{1}{2} \sum_{\vec{k}_1 \lambda_1} \sum_{\vec{k}_2 \lambda_2} \right) \sum_{\overline{m}} C_{fi} \\ &= \frac{1}{2} \sum_{\vec{k}_1 \lambda_1} \sum_{\vec{k}_2 \lambda_2} \sum_{\overline{m}} \Gamma_{f\overline{m}}^{(1)} \Gamma_{\overline{m}i}^{(1)} \\ &= \frac{1}{2} \sum_{\overline{m}_A} \sum_{\vec{k}_1 \lambda_1} \sum_{\vec{k}_2 \lambda_2} \sum_{\vec{k}\lambda} 2\pi\delta(E_{f_A} + k_1 + k_2 - E_{\overline{m}_A} - k) \\ &\quad \times 2\pi\delta(E_{\overline{m}_A} - E_{i_A} - k) \\ &\quad \times |\langle \overline{m}_A, 1_{\vec{k}_1 \lambda_1}, 1_{\vec{k}_2 \lambda_2} | (-e\vec{x} \cdot \vec{E}) | i_A, 1_{\vec{k}\lambda} \rangle|^2 \\ &\quad \times |\langle \overline{m}_A, 1_{\vec{k}\lambda} | (-e\vec{x} \cdot \vec{E}) | i_A, 0 \rangle|^2. \end{aligned} \quad (31)$$

The summation over $\vec{k}\lambda$ is over both polarizations λ and over an energy interval for $k = |\vec{k}|$ which contains the resonance frequency of the intermediate atomic resonant state $|\overline{m}_A\rangle$. After performing the sum over $\vec{k}\lambda$ and going to the continuum limit with the help of Eq. (10), we obtain

$$\begin{aligned} C_{f_A i_A} &= e^4 \sum_{\overline{m}_A} \sum_{\vec{k}_1 \lambda_1} \sum_{\vec{k}_2 \lambda_2} (2\pi)^2 \delta[k_1 - (E_{f_A} - E_{\overline{m}_A})] \\ &\quad \times \delta[k_2 - (E_{\overline{m}_A} - E_{i_A})] \frac{k_1}{2V} |\langle f_A | \hat{\epsilon}_{\vec{k}_1 \lambda_1} \cdot \vec{x} | \overline{m}_A \rangle|^2 \\ &\quad \times \frac{k_2}{2V} |\langle \overline{m}_A | \hat{\epsilon}_{\vec{k}_2 \lambda_2} \cdot \vec{x} | i_A \rangle|^2 \\ &= \sum_{\overline{m}_A} \left[\frac{4\alpha}{3} (E_{f_A} - E_{\overline{m}_A})^3 |\langle f_A | \vec{x} | \overline{m}_A \rangle|^2 \right. \\ &\quad \left. \times \frac{4\alpha}{3} (E_{\overline{m}_A} - E_{i_A})^3 |\langle \overline{m}_A | \vec{x} | i_A \rangle|^2 \right] \\ &= \sum_{\overline{m}_A} \Gamma_{f_A \overline{m}_A}^{(1\gamma)} \Gamma_{\overline{m}_A i_A}^{(1\gamma)}. \end{aligned} \quad (32)$$

This result confirms that the cascade terms can indeed be written as the product of atomic one-photon decay rates.

V. CONCLUSIONS

In this paper, we have reviewed the formulation of a transition with a single quantum jump within time-dependent perturbation theory (see Sec. II A). The result, which is Fermi's golden rule [see Eq. (4)], is evaluated for an atomic dipole transition in Sec. II B. We find that the familiar result for the one-photon decay rate [see Eq. (12)] is obtained after a summation/integration over the degrees of freedom of the emitted photon in the continuum limit, as given in Eq. (10). The general formulation of a transition with two quantum jumps is carried out in Sec. III A, within second-order time-dependent perturbation theory. The result for the two-quantum decay rate $\Gamma_{fi}^{(2)}$ as given in Eq. (15) is valid if there are no resonant intermediate states through which a cascade decay could possibly proceed. The specialization to a transition with two quantum jumps is carried out in Sec. III B, where it is shown that a summation over the two-photon final states of the process [see Eq. (16)] yields the familiar result (18) for a two-photon

transition rate in a hydrogenlike ion (such as the $2S$ - $1S$ decay). Cascade contributions are analyzed in Sec. IV A.

By isolating the resonant states within the intermediate states of the process, we obtain the cascade contribution (23) after differentiating the probability of finding the system in the final state *twice* with respect to the elapsed time. Subtracting the contribution of the resonant states within the *first* time derivative of said probability, we obtain an expression for the two-quantum correction to the decay rate of a system which can simultaneously decay via cascades and two-quantum transitions. If we use the identification (27) for the regularization parameter η that parametrizes the width of the intermediate states, then the effect of subtracting the resonant states from the two-quantum decay rate is equivalent to the subtraction of the total one-photon decay rate of the initial state [see Eq. (29)]. The difference of the formal two-quantum decay rate (with the propagator denominators of the resonant states regularized by their widths) and the one-photon decay rate therefore constitutes the two-quantum correction $\overline{\Gamma}_{fi}^{(2)}$ to the decay rate, as noted in Eq. (29).

The formal two-quantum decay rate (with the propagator denominators of the resonant states regularized by their widths) therefore constitutes more than its name might suggest: namely, according to Eq. (30), it is the sum of the one-quantum decay rate and of the two-quantum correction and therefore constitutes, in some sense, a one + two quantum decay rate. This finding provides a formal and general justification for the observation made in Ref. [12]: namely, that the formal two-photon decay rate of, say, a $4S$ state in a hydrogenlike ion contains the one-photon decay rate of $4S$ as a lower-order term which needs to be subtracted in order to obtain the pure two-photon correction. It is instructive to also remember that competing one- and two-photon decays are not restricted to atomic hydrogen, but also occur in other atomic systems of fundamental importance like rubidium [17].

Finally, in Sec. IV B, we analyze the cascade contribution for atomic transitions and show that under the appropriate normalization of the two-photon final state [see Eqs. (16) and (31)], the familiar result is obtained [see Eq. (32)]. The results reported here have an interesting consequence, because they imply that the one + two-photon decay rate (30) gives the correct differential-in-energy rate coefficients for recombination codes [13,14], where the spectrum of emitted photons from both two-photon and cascade transitions is needed over the entire range of resonant and nonresonant frequencies. In particular, the corresponding expression (30) can be used even in the vicinity of resonant bound-state poles, provided these are regularized by their respective decay widths.

One particular limitation of the treatment discussed here is immediately obvious. Adiabatic perturbation theory does not make any statement about the time points t' and t'' where the two photons are emitted. Thus, the total regularized two-photon decay rate as described by Eq. (30) contains both correlated emission (with a small time difference $|t' - t''|$) and also sequential emission (with a large time difference $|t' - t''|$). Since it is only the photon spectrum, not the correlation [18], that matters for cosmological recombination, we can use the regularized two-photon decay rate (30), differential in energy, with good accuracy for recombination codes [13,14] which need the differential-in-energy spectrum of the emitted photons as input. In particular, this means

that it is not necessary to distinguish specific contributions to the complete rate (30); this rate contains both cascade photons and correlated two-photon processes in a natural way. Therefore, it is understandable that the authors of Ref. [19] could not give a unique value to their parameter $\Delta\omega$; this parameter was introduced in order to distinguish between cascade photons and two-photon decays. If one would like to make a more refined distinction between cascade photons and correlated two-photon processes, then one has to go beyond adiabatic perturbation theory and analyze the dependence of the process on the emission times t' and t'' , including loss of correlation as a function of $|t' - t''|$, which may be process- as well as environment-dependent (e.g., there may be a dependence on the average mean free path of the atoms in their environment).

Our article illustrates both the usefulness but also the limitations of adiabatic perturbation theory. Namely, if we apply the regularization (27) consistently, to both the first term as well as the second term on the right-hand side of Eq. (25), then the subtraction term S defined in Eq. (26) attains a finite value and can be evaluated in closed form [see Eq. (28)]. Furthermore, as shown in Ref. [12], if the regularization (27) is applied to a two-photon transition in atoms, then there are significant cancellations between the two terms on the right-hand side of Eq. (29), which are both of order $\alpha^2(Z\alpha)^4$, but their difference is of order $\alpha^2(Z\alpha)^6$, where Z is the nuclear charge number, and α is the fine-structure constant. Two-photon decay rates are of order $\alpha^2(Z\alpha)^6$. As evident from Eq. (11) of Ref. [12] and from Eq. (28) in the current work, the cancellation of the lower-order terms depends on the particular choice of the regularization. Within adiabatic perturbation theory, the regularization (27) thus appears to be the only one which leads to a consistent removal of the infinities that plague the two-quantum decay rate in the presence of allowed cascade transitions. Therefore, our article offers a connection of the adiabatic parameter η used in time-dependent perturbation theory to a physical concept, namely, the lifetime of virtual intermediate states.

An interesting connection to the theory of energy shifts of atomic levels can be drawn. Low [20–22] observed that the calculation of energy shifts of excited states of hydrogenlike ions becomes problematic at order $\alpha^2(Z\alpha)^6$, due to interference effects of the resonance line shapes of atomic levels of different principal quantum number. It has been argued that at order $\alpha^2(Z\alpha)^6$, two-loop energy shifts of excited states cannot be uniquely associated any more with a particular atomic level, due to the predictive limits of adiabatic perturbation theory [20–22]. The decay rate at order $\alpha^2(Z\alpha)^6$ constitutes the imaginary part of the energy shift of that same order. It is thus not surprising that its calculation requires considerable effort within the formalism of adiabatic perturbation theory.

ACKNOWLEDGMENTS

The author acknowledges helpful conversations with Professor A. I. Milstein. Helpful remarks of an anonymous referee are also gratefully acknowledged. This research was supported by the National Science Foundation (PHY-8555454) and by a precision measurement grant from the National Institute of Standards and Technology.

- [1] J. J. Sakurai, *Modern Quantum Mechanics* (Addison-Wesley, Reading, MA, 1994).
- [2] J. D. Cresser, A. Z. Tang, G. J. Salamo, and F. T. Chan, *Phys. Rev. A* **33**, 1677 (1986).
- [3] U. D. Jentschura, *J. Phys. A* **41**, 155307 (2008).
- [4] U. D. Jentschura and A. Surzhykov, *Phys. Rev. A* **77**, 042507 (2008).
- [5] D. H. Kobe, *Phys. Rev. Lett.* **40**, 538 (1978).
- [6] D. H. Kobe, *J. Phys. B* **16**, 1159 (1983).
- [7] R. R. Schlicher, W. Becker, J. Bergou, and M. O. Scully, in *Quantum Electrodynamics and Quantum Optics*, edited by A.-O. Barut (Plenum, New York, 1984), pp. 405–441.
- [8] M. Göppert-Mayer, *Ann. Phys. (Leipzig)* **401**, 273 (1931).
- [9] U. D. Jentschura, *Phys. Rev. A* **69**, 052118 (2004).
- [10] M. E. Peskin and D. V. Schroeder, *An Introduction to Quantum Field Theory* (Perseus, Reading, MA, 1995).
- [11] W. Greiner and J. Reinhardt, *Quantum Electrodynamics* (Springer, New York, 1992).
- [12] U. D. Jentschura, *Phys. Rev. A* **79**, 022510 (2009).
- [13] J. Chluba and R. A. Sunyaev, *Astron. Astrophys.* **480**, 629 (2008).
- [14] E. R. Switzer and C. M. Hirata, *Phys. Rev. D* **77**, 083006 (2008).
- [15] P. Amaro, J. P. Santos, F. Parente, A. Surzhykov, and P. Indelicato, *Phys. Rev. A* **79**, 062504 (2009).
- [16] U. D. Jentschura, *J. Phys. A* **40**, F223 (2007).
- [17] D. Touahri, O. Acef, A. Clairon, J. J. Zondy, R. Felder, L. Hilico, B. de Beauvoir, F. Biraben, and F. Nez, *Opt. Commun.* **133**, 471 (1997).
- [18] T. Radtke, A. Surzhykov, and S. Fritzsche, *Phys. Rev. A* **77**, 022507 (2008).
- [19] L. Labzowsky, D. Solovyev, and G. Plunien, e-print arXiv:0904.2346 [physics.atom-ph].
- [20] F. Low, *Phys. Rev.* **88**, 53 (1952).
- [21] U. D. Jentschura, J. Evers, C. H. Keitel, and K. Pachucki, *New J. Phys.* **4**, 49 (2002).
- [22] L. N. Labzowsky, D. A. Solovyev, G. Plunien, and G. Soff, *Phys. Rev. Lett.* **87**, 143003 (2001).