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Spontaneous emission and scattering in a two-atom system: Conservation of probability and energy

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An explicit calculation of conservation of probability and energy in a two-atom system is presented. One of the atoms is excited initially and undergoes spontaneous emission. The field radiated by this atom can be scattered by the second atom. It is seen that the Weisskopf-Wigner approximation must be applied using a specific prescription to guarantee conservation of probability and energy. Moreover, for consistency, it is necessary to take into account the rescattering by the source atom of radiation scattered by the second atom.

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I. INTRODUCTION

Recently, there has been renewed interest in understanding problems involving emission in a dielectric from a microscopic point of view [1–3]. To understand more complex problems, such as those involving propagation in a dielectric and/or the recoil an atom undergoes in spontaneous emission, it is helpful to first consider the simpler problem of a single source atom and a single medium atom. In the case of *identical* atoms, this problem has received a great deal of attention [4,5]. There are also calculations in which cooperative emission involving two different atoms is considered [6]. In most of these treatments, however, there was little or no discussion of conservation of energy and probability. It might seem that this is a trivial consideration, but, as is often the case, appearances can be deceiving.

It should be recalled that probability conservation proves to be problematic even for the problem of an *isolated* atom undergoing spontaneous decay. This problem has never been solved exactly, although perturbative solutions based on quantum electrodynamics can be formulated. Weisskopf and Wigner [7] considered the problem of spontaneous decay and introduced two approximations that led to a consistent solution in which probability was conserved. The first approximation was to extend the frequency integration over vacuum field modes to minus infinity, based on the assumption that most contributions of field modes occur near the atomic frequency. The second approximation was to evaluate all vacuum field mode frequencies at the atomic transition frequency, *except* when those frequencies appeared in phase factors. There is no formal justification for this procedure; if one does not make such an assumption, but introduces some appropriate high-frequency cutoff, probability is conserved only approximately [8]. It is rather remarkable that the Weisskopf-Wigner approach leads to conservation of probability [9]. On the other hand, there is no guarantee that this procedure will work when analyzing more complex systems, such as the two-atom system considered in this work. One goal of this paper is, then, to establish explicitly the manner in which probability and energy are conserved in a two-atom system and to indicate the manner in which the Weisskopf-Wigner approximation must be used to achieve these results.

A secondary goal of this paper is to examine the various physical processes that give rise to interference effects between the field emitted by the source atom and the scatterer. In particular, the role played by retardation is brought into focus. To illustrate the conceptual problems that can arise, consider the scattering problem illustrated schematically in Fig. 1. A source atom A at the origin is excited at $t=0$ to a state having energy $\hbar\omega_0$ and undergoes spontaneous emission. The field radiated by the source atom can scatter from a second atom B located at position $\mathbf{R}_0$. Within the rotating-wave approximation, there are only three state amplitudes

$$b_{2,1;0},\ \ b_{1,2;0},\ \ b_{1,1;\mathbf{k}}$$

that enter the problem. The first subscript refers to atom A, the second to atom B, and the third to the radiation field. To first order in (Γ/Δ), where the detuning

$$\Delta = \omega_0 - \omega \tag{1}$$

and ω is the atom B transition frequency, the probability

$$\sum_{\mathbf{k}} |b_{1,1;\mathbf{k}}|^2 + |b_{2,1;0}|^2 = 1, \tag{2}$$

since the lowest contribution to $|b_{1,2;0}|^2$ is of order $(\Gamma/\Delta)^2$. The rate Γ is less than or of the order of the decay rates of atoms A or B.

As innocuous as it seems, Eq. (2) leads to an apparent paradox. The amplitude $b_{1,1;\mathbf{k}}$ has three contributions in lowest-order perturbation theory. The first arises from the direct emission of atom A with no scattering and is of order $(\Gamma/\Delta)^0$. The second, of order (Γ/Δ), involves scattering from atom B and depends on $b_{2,1;0}(t-R_0/c)$ since the radiation

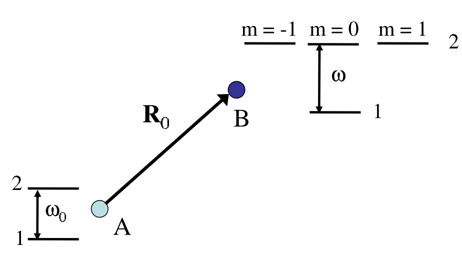

FIG. 1. (Color online) Source atom A is prepared in its excited state and undergoes spontaneous emission that can be scattered by atom B.

must travel from atom A to atom B before it is scattered. The third contribution, already discussed by Milonni and Knight and shown to be of the same order as the second contribution [4], can be linked to radiation that is emitted by atom A, scattered by atom B, and then rescattered from atom A. This contribution must depend on $b_{2,1;0}(t-2R_0/c)$ since the radiation must travel from atom A to atom B and back before it can be rescattered by atom A. It would appear that the interference terms in $|b_{1,1;\mathbf{k}}|^2$ involving the $(\Gamma/\Delta)^0$ and each of the (Γ/Δ) terms involve retardation at *different* times, namely, R_0/c and $2R_0/c$. But all contributions to the probability resulting from the presence of atom B are of order $(\Gamma/\Delta)^2$. As a consequence, all contributions to the probability of order (Γ/Δ) must vanish. How can these (Γ/Δ) terms involving retardation at two different times combine and cancel one another to ensure that overall probability is conserved? This apparent paradox is resolved in Sec. II.

Conservation of energy also proves to be problematic. Given the initial conditions, it is clear that the average energy in the system of atoms plus field is equal to $\hbar\omega_0$. Since this average energy must be conserved, we face a range of problems similar to those encountered in the probability calculation. An analysis of energy conservation allows one to see how the energy is partitioned between the atoms and the field.

The problems discussed in this paper are of fundamental importance when one considers the more complex problem of a medium of scatterers or a system in which the source atom is allowed to recoil on emission. As such, this calculation represents a type of "building-block" solution that may prove useful in treating such problems.

II. HAMILTONIAN

The source atom, located at $\mathbf{R}_0=0$, is modeled as having a $J=0$ ground and $J=1$ excited state separated in frequency by ω_0. Atom B is located at position $\mathbf{R}_0$ and has a $J=0$ ground state and $J=1$ excited state; the frequency separation of its ground and excited states is denoted by ω. It is assumed that $|\Delta|/\omega \ll 1$, enabling one to make a rotating-wave approximation in considering the field–atom-B interaction. On the other hand, it is assumed that $\Gamma/|\Delta| \ll 1$, allowing one to make an expansion of the probability amplitudes as a power series in this parameter. The source atom is excited by a z-polarized optical pulse into its $m_J=0$ excited state at $t=0$. As such, the source atom can be considered as a "two-level" atom with lower state $|1\rangle=|J=0\rangle$ and upper state $|2\rangle=|J=1,m_J=0\rangle$. The processes I consider are (i) radiation emitted by the source atom without scattering, (ii) radiation emitted by the source atom that is scattered by atom B, and (iii) radiation that is emitted by the source atom, scattered by atom B, and rescattered by the source atom.

In rotating-wave approximation, the Hamiltonian for the atom-field system is

$$H = H_0 + V, \tag{3}$$

where

$$H_0 = \frac{\hbar\omega_0}{2}\sigma_z^A + \sum_{m=-1}^{1}\frac{\hbar\omega}{2}\sigma_z^B(m) + \hbar\omega_k a_{\mathbf{k}_\lambda}^\dagger a_{\mathbf{k}_\lambda}, \tag{4}$$

$$V = \hbar g_{\mathbf{k}}(\sigma_+ a_{\mathbf{k}} - a_{\mathbf{k}}^\dagger\sigma_-) + \hbar\big(g'_{\mathbf{k}_\lambda}(m)\sigma_+^B(m)a_{\mathbf{k}_\lambda}e^{i\mathbf{k}\cdot\mathbf{R}_0} + g'_{\mathbf{k}_\lambda}(m)^* a_{\mathbf{k}_\lambda}^\dagger\sigma_-^B(m)e^{-i\mathbf{k}\cdot\mathbf{R}_0}\big), \tag{5}$$

$$g_{\mathbf{k}} = -i\left(\frac{\omega_k}{2\hbar\epsilon_0\mathcal{V}}\right)^{1/2}\mu(\boldsymbol{\epsilon}_{\mathbf{k}}^{(1)})_0,$$

$$g'_{\mathbf{k}_\lambda}(m) = -i\left(\frac{\omega_k}{2\hbar\epsilon_0\mathcal{V}}\right)^{1/2}\mu'(\boldsymbol{\epsilon}_{\mathbf{k}}^{(\lambda)})_m^*, \tag{6}$$

$\sigma_\pm$ are raising and lowering operators for the source atom, $\sigma_z=|2\rangle\langle 2|-|1\rangle\langle 1|$, $\sigma_\pm^B(m)$ are raising and lowering operators for atom B associated with the m sublevel of the $J=1$ excited state, $\sigma_z^B(m)=|m\rangle^B\langle m|-|g\rangle^B\langle g|$ is the population difference operator between excited state $(J=1,m)$ and ground state $(g,J=0)$ of atom B, μ is the z component of the dipole moment matrix element of the source atom between its ground and excited states (assumed real), μ' is $\sqrt{3}$ times the reduced matrix element of the dipole operator for atom B between its ground and excited states (assumed real), $a_{\mathbf{k}_\lambda}$ is an annihilation operator for a photon having propagation vector $\mathbf{k}$ and polarization $\boldsymbol{\epsilon}_{\mathbf{k}}^{(\lambda)}$, $\omega_k=kc$, $\mathcal{V}$ is the quantization volume, and $(\boldsymbol{\epsilon}_{\mathbf{k}}^{(\lambda)})_{\pm1}=\mp[(\boldsymbol{\epsilon}_{\mathbf{k}}^{(\lambda)})_x \pm i(\boldsymbol{\epsilon}_{\mathbf{k}}^{(\lambda)})_y]/\sqrt{2}$, $(\boldsymbol{\epsilon}_{\mathbf{k}}^{(\lambda)})_0=(\boldsymbol{\epsilon}_{\mathbf{k}}^{(\lambda)})_z$, where the unit polarization vectors are

$$\boldsymbol{\epsilon}_{\mathbf{k}}^{(1)} = \hat{\theta}_k = \cos\theta_k\cos\phi_k\hat{\mathbf{x}} + \cos\theta_k\sin\phi_k\hat{\mathbf{y}} - \sin\theta_k\hat{\mathbf{z}}; \tag{7a}$$

$$\boldsymbol{\epsilon}_{\mathbf{k}}^{(2)} = \hat{\phi}_k = -\sin\phi_k\hat{\mathbf{x}} + \cos\phi_k\hat{\mathbf{y}}. \tag{7b}$$

A summation convention is used in Eqs. (4) and (5), as well as all subsequent equations, in which any repeated symbol on the right-hand side of an equation is summed over, unless it also appears on the left-hand side of the equation. Equations (4) and (5) describe a system in which the source atom and atom B interact via the quantized radiation field.

It is convenient to expand the state vector as

$$|\psi(t,\mathbf{R}_0)\rangle = b_{2,0}(t,\mathbf{R}_0)e^{-i\omega_0 t}|2;0\rangle + \tilde{b}_{m,0}(t,\mathbf{R}_0)e^{-i\omega_0 t}|m;0\rangle + b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)e^{-i\omega_k t}|\mathbf{k}_\lambda\rangle, \tag{8}$$

where $b_{2,0}\equiv b_{2,1;0}$ is the amplitude (in an interaction representation) to find the source atom excited, atom B in its ground state, and no photons in the field; $\tilde{b}_{m,0}\equiv b_{1,m;0}$ is the amplitude (in an interaction representation) to find atom B in excited sublevel m, the source atom in its ground state, and no photons in the field; while $b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)\equiv b_{1,1;\mathbf{k}_\lambda}$ is the amplitude (in an interaction representation) to find both atoms in their ground states and a photon having wave vector $\mathbf{k}$ and polarization λ in the field. In the rotating-wave approximation, these are the only states that enter for the chosen initial conditions. Note that the third term is written as $\tilde{b}_{m,0}(t,\mathbf{R}_0)e^{-i\omega_0 t}$ rather than $\tilde{b}_{m,0}(t,\mathbf{R}_0)e^{-i\omega t}$ to reflect the fact that this state actually enters as a virtual state with energy $\hbar\omega_0$ rather than energy $\hbar\omega$. Probabilities are calculated to order (Γ/Δ) and energies to order $\hbar\Gamma^2/\Delta$. Higher-order corrections can be included, but they seriously complicate the solution.

III. CONSERVATION OF PROBABILITY

To prove explicitly that population is conserved, one must show that

$$P = |b_{2,0}(t,\mathbf{R}_0)|^2 + \sum_{\mathbf{k}_\lambda} |b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2 + |\tilde{b}_{m,0}(t,\mathbf{R}_0)|^2 \approx |b_{2,0}(t,\mathbf{R}_0)|^2 + \sum_{\mathbf{k}_\lambda} |b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2 = 1, \quad (9)$$

assuming $b_{2,0}(t=0)=1$. The $|\tilde{b}_{m,0}(t,\mathbf{R}_0)|^2$ term can be neglected in the probability calculation since it is of order $(\Gamma/\Delta)^2$. To proceed, I note first that $b_{2,0}(t)$ can be written as

$$b_{2,0}(t,\mathbf{R}_0) = b_{2,0}^{(0)}(t) + \beta(t,\mathbf{R}_0), \quad (10)$$

where $b_{2,0}^{(0)}(t)$ is the solution in the absence of atom B, and $\beta(t,\mathbf{R}_0)$ is a correction of order (Γ/Δ). Assuming the source atom to be excited "instantaneously" at $t=0$, it follows that [10]

$$b_{2,0}^{(0)}(t) = e^{-\gamma t}\Theta(t), \quad (11)$$

where $\gamma=(1/4\pi\epsilon_0)(2\mu^2\omega_0^3/3\hbar c^3)$ is one-half the source atom decay rate (in the absence of atom B) and $\Theta(t)$ is the Heaviside step function. Equation (11) is derived using the conventional Weisskopf-Wigner approximation in which integrals over ω_k are extended to $-\infty$ and all factors of ω_k are evaluated at ω_0, except when they appear in phase factors. To calculate P, one needs to find $b_{2,0}(t,\mathbf{R}_0)$ and $\Sigma_{\mathbf{k}_\lambda}|b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2$.

From Schrödinger's equation, it follows that the state amplitude $b_{\mathbf{k}_\lambda}(t)$ evolves as

$$\dot{b}_{\mathbf{k}_\lambda}(t,\mathbf{R}_0) = ig_{\mathbf{k}}e^{-i(\omega_0-\omega_k)t}b_{2,0}(t) - ig'_{\mathbf{k}_\lambda}(m)^* e^{-i(\omega_0-\omega_k)t}e^{-i\mathbf{k}\cdot\mathbf{R}_0}\tilde{b}_{m,0}(t) \quad (12)$$

or

$$\begin{aligned}\sum_{\mathbf{k}_\lambda} |b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2 &= \sum_{\mathbf{k}_\lambda} \left| \int_0^t dt' e^{-i(\omega_0-\omega_k)t'}[g_{\mathbf{k}}b_{2,0}(t',\mathbf{R}_0) - g'_{\mathbf{k}_\lambda}(m)^* e^{-i\mathbf{k}\cdot\mathbf{R}_0}\tilde{b}_{m,0}(t',\mathbf{R}_0)] \right|^2 \\ &= \sum_{\mathbf{k}_\lambda} \int_0^t dt' \int_0^t dt'' e^{i(\omega_k-\omega_0)(t'-t'')} \times [|g_{\mathbf{k}}|^2 b_{2,0}(t',\mathbf{R}_0)b_{2,0}^*(t'',\mathbf{R}_0) + |g'_{\mathbf{k}_\lambda}(m)|^2 \tilde{b}_{m,0}(t',\mathbf{R}_0)\tilde{b}_{m,0}^*(t'',\mathbf{R}_0)] \\ &\quad - \left[\int_0^t dt' \int_0^t dt'' e^{i(\omega_k-\omega_0)(t'-t'')} g_{\mathbf{k}} g'_{\mathbf{k}_\lambda}(m) \times e^{i\mathbf{k}\cdot\mathbf{R}_0} b_{2,0}(t',\mathbf{R}_0)\tilde{b}_{m,0}^*(t'',\mathbf{R}_0) + \text{c.c.} \right].\end{aligned} \quad (13)$$

The amplitudes $b_{2,0}(t,\mathbf{R}_0)$ and $\tilde{b}_{m,0}(t,\mathbf{R}_0)$ obey the equations of motion [2,3]

$$\dot{b}_{2,0} = -\gamma b_{2,0} - \gamma(\mu'/\mu)e^{ik_0R_0}F_{0;m}(\mathbf{R}_0,k_0)\tilde{b}_{m,0}(t-R_0/c), \quad (14a)$$

$$d\tilde{b}_{m,0}/dt = -[\gamma(\mu'/\mu)^2 - i\Delta]\tilde{b}_{m,0} - \gamma(\mu'/\mu)e^{ik_0R_0}F_{m;0}(\mathbf{R}_0,k_0)b_{2,0}(t-R_0/c), \quad (14b)$$

where

$$F_{0;0}(\mathbf{R}_0,k_0) = \sqrt{4\pi}\left(d_0(k_0R_0)Y_{0,0}(\theta_0,\phi_0) + \frac{1}{\sqrt{5}}d_2(k_0R_0)Y_{2,0}(\theta_0,\phi_0) \right), \quad (15a)$$

$$-F_{0;\mp 1}(\mathbf{R}_0,k_0) = F_{\pm 1;0}(\mathbf{R}_0,\omega_0) = -\sqrt{4\pi}\sqrt{3/20}d_2(k_0R_0)Y_{2,\mp 1}(\theta_0,\phi_0), \quad (15b)$$

$$d_0(x) = -\frac{i}{x}, \quad d_2(x) = -i\left(\frac{3}{x^3} - \frac{1}{x}\right) - \frac{3}{x^2}, \quad (16)$$

and $k_0=\omega_0/c$ [$d_\ell(x)=e^{-ix}h_\ell(x)$, where $h_\ell(x)$ is a spherical Hankel function of the first kind]. Equation (14b) can be integrated formally, assuming that $b_{2,0}$ varies slowly in a time of order $1/\Delta$ [10]. To order Γ/Δ, one obtains

$$\tilde{b}_{m,0}(t) \approx -i\frac{\gamma(\mu'/\mu)}{\Delta}e^{ik_0R_0}F_{m;0}(\mathbf{R}_0,k_0)b_{2,0}^{(0)}(t-R_0/c), \quad (17)$$

where

$$\dot{b}_{2,0} = -\gamma b_{2,0} - [\tilde{\Gamma}(\mathbf{R}_0) + i\tilde{S}(\mathbf{R}_0)]b_{2,0}^{(0)}(t-2R_0/c) \quad (18)$$

and

$$\tilde{\Gamma}(\mathbf{R}_0) + i\tilde{S}(\mathbf{R}_0) = -i\frac{\gamma^2(\mu'/\mu)^2}{\Delta}e^{2ik_0R_0}F_{0;m}(\mathbf{R}_0,k_0)F_{m;0}(\mathbf{R}_0,k_0) \quad (19)$$

with both $\tilde{\Gamma}(\mathbf{R}_0)$ and $\tilde{S}(\mathbf{R}_0)$ real.

Note that, for $k_0R_0 \gg 1$ (atom B in the radiation zone of atom A),

$$\tilde{\Gamma}(\mathbf{R}_0) = \Gamma(\mathbf{R}_0)\sin(2k_0R_0), \quad (20a)$$

$$\tilde{S}(\mathbf{R}_0) = -\Gamma(\mathbf{R}_0)\cos(2k_0R_0), \quad (20b)$$

where

$$\Gamma(\mathbf{R}_0) = -\frac{9}{4}\frac{\gamma^2(\mu'/\mu)^2}{\Delta(k_0R_0)^2}\sin^2\theta_0. \tag{21}$$

The general result (19) contains both near- and intermediate-field contributions; however, the adiabatic solution (17) is valid only if $[\gamma(\mu'/\mu)/\Delta]|F_{m;0}(\mathbf{R}_0,k_0)| \ll 1$ or

$$(k_0R_0)^3 \gg \frac{\gamma(\mu'/\mu)}{\Delta}. \tag{22}$$

For smaller separations, the two-atom system decays in a cooperative fashion. It is assumed that condition (22) holds.

Equation (17) can now be substituted into Eq. (13). The term proportional to $\tilde{b}_{m,0}(t',\mathbf{R}_0)\tilde{b}^*_{m,0}(t'',\mathbf{R}_0)$ is of order $(\Gamma/\Delta)^2$ and can be neglected. Evaluating the first term using the "normal" Weisskopf-Wigner approximation (setting $\omega_k = \omega_0$ except if it appears in a phase factor), one obtains

$$\sum_{\mathbf{k}_\lambda}|b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2 = 2\gamma\int_0^t dt'|b_{2,0}(t',\mathbf{R}_0)|^2 - \Bigg[\int_0^t dt'\int_0^t dt'' e^{i(\omega_k-\omega_0)(t'-t'')}g_{\mathbf{k}}g'_{\mathbf{k}_\lambda}(m)e^{i\mathbf{k}\cdot\mathbf{R}_0} \times b_{2,0}(t',\mathbf{R}_0)\tilde{b}^*_{m,0}(t'',\mathbf{R}_0) + \text{c.c.}\Bigg]. \tag{23}$$

By combining Eqs. (9), (17), and (18) {with $\gamma b_{2,0} = -\dot{b}_{2,0} - [\tilde{\Gamma}(\mathbf{R}_0) + i\tilde{S}(\mathbf{R}_0)]b^{(0)}_{2,0}(t-2R_0/c)$}, and (19), one can show, that for probability to be conserved, one must have

$$B_1 + B_2 = 0, \tag{24}$$

where

$$B_1 = -\tilde{\Gamma}(\mathbf{R}_0)(2\gamma)\int_0^t dt' b^{(0)}_{2,0}(t')b^{(0)}_{2,0}(t'-2R_0/c), \tag{25a}$$

$$B_2 = -i\frac{\gamma(\mu'/\mu)}{\Delta}F^*_{m;0}(\mathbf{R}_0,k_0)g_{\mathbf{k}}g'_{\mathbf{k}_\lambda}(m)e^{-ik_0R_0}e^{i\mathbf{k}\cdot\mathbf{R}_0}\int_0^t dt' \times b^{(0)}_{2,0}(t')\int_0^t dt'' e^{i(\omega_k-\omega_0)(t'-t'')}b^{(0)}_{2,0}(t''-R_0/c) + \text{c.c.}, \tag{25b}$$

and we used the fact that $b^{(0)}_{2,0}(t)$ is real. As mentioned in the Introduction, for probability to be conserved there must be cancellation of a term involving retardation at R_0/c with one at $2R_0/c$.

This apparent paradox is resolved when one explicitly carries out the integration in Eq. (25b). The interference of $b^{(0)}_{2,0}(t')$ and $b^{(0)}_{2,0}(t''-R_0/c)$ occurs only for $t''=t'-R_0/c$, leading to a term that varies as $b^{(0)}_{2,0}(t'-2R_0/c)$. To see this explicitly, one carries out the summation over $\mathbf{k}$ in Eq. (25b) by using the prescription $\Sigma_{\mathbf{k}} \rightarrow [\mathcal{V}/(2\pi)^3]\int d\mathbf{k} = [\mathcal{V}/(2\pi c)^3]\int_0^\infty \omega_k^2 d\omega_k \int d\Omega_k$, extending the ω_k integration to $-\infty$, setting $\omega_k = \omega_0$ except where it appears in phase factors, and using the relation

$$e^{i\mathbf{k}\cdot\mathbf{R}_0} = 4\pi\sum_{m=-\ell}^{\ell} i^\ell Y_{\ell m}(\theta_k,\phi_k)Y^*_{\ell m}(\theta_0,\phi_0)j_\ell(kR_0), \tag{26}$$

in which $Y_{\ell m}$ is a spherical harmonic and j_ℓ a spherical Bessel function. Only the $h^*_\ell(kR_0)$ part of $j_\ell(kR_0) = [h_\ell(kR_0) + h^*_\ell(kR_0)]/2$ contributes to B_2 since the first term in Eq. (25b) arising from the $h_\ell(kR_0)$ contribution is purely imaginary. After some algebra, one obtains

$$B_2 = \tilde{\Gamma}(\mathbf{R}_0)(2\gamma)\int_0^t dt' b^{(0)}_{2,0}(t')b^{(0)*}_{2,0}(t'-2R_0/c) = -B_1 \tag{27}$$

and probability is conserved, as expected.

One can understand the dependence $b^{(0)}_{2,0}(t')b^{(0)}_{2,0}(t'-2R_0/c)$ appearing in B_1 and B_2 in terms of interference in emission. The field intensity emitted from the source atom involves a product of excited-state amplitudes $b_{2,0}(t')b^*_{2,0}(t')$ evaluated at the *same* time. Since $b_{2,0}(t')$ has contributions from both $b^{(0)}_{2,0}(t')$ and $b^{(0)}_{2,0}(t'-2R_0/c)$, the interference between these components gives rise to the dependence in B_1. Thus B_1 corresponds to an interference between the field backscattered by atom B and *rescattered* by the source atom with the field emitted by the source atom at a later time.

The B_2 term is more complex. The B_2 term can be written as

$$B_2 = \sum_{\mathbf{k}_\lambda} I_{\mathbf{k}_\lambda}(t,\mathbf{R}_0), \tag{28}$$

where

$$I_{\mathbf{k}_\lambda}(t,\mathbf{R}_0) = \int_0^t dt'\int_0^t dt'' I_{\mathbf{k}_\lambda}(t,\mathbf{R}_0;t',t'')^{i(\omega_k-\omega_0)(t'-t'')} \tag{29}$$

and

$$I_{\mathbf{k}_\lambda}(t,\mathbf{R}_0;t',t'') = e^{i(\omega_k-\omega_0)(t'-t'')} \times [g_{\mathbf{k}}g'_{\mathbf{k}_\lambda}(m)e^{i\mathbf{k}\cdot\mathbf{R}_0}b_{2,0}(t')\tilde{b}^*_{m,0}(t'',\mathbf{R}_0) + \text{c.c.}]. \tag{30}$$

The quantity $I_{\mathbf{k}_\lambda}(t,\mathbf{R}_0;t',t'')$ is the contribution to $|b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2$ resulting from interference of the field emitted by atom A at time t' with that emitted by atom B at time t'' (recall that $\mathbf{R}_0 = \mathbf{R}_B - \mathbf{R}_A$). Actually, the quantity of interest here is

$$I_{\omega_k}(t,\mathbf{R}_0;t',t'') = \sum_\lambda \int d\Omega_k I_{\mathbf{k}_\lambda}(t,\mathbf{R}_0;t',t''), \tag{31}$$

which can be interpreted as a frequency distribution associated with this interference term. With the use of the addition theorem Eq. (26), and the fact that $j_\ell(x) = [e^{ix}d_\ell(x) + e^{-ix}d^*_\ell(x)]/2$, it follows that, to order (Γ/Δ),

$$I_{\omega_k}(t,\mathbf{R}_0;t',t'') = iC_m(\mathbf{R}_0)e^{ik_0R_0}b_{2,0}^{(0)}(t')\tilde{b}_{m,0}^{*}(t'',\mathbf{R}_0) \times e^{i(\omega_k-\omega_0)(t'-t''+R_0/c)} + \text{c.c.} + iD_m(\mathbf{R}_0)e^{-ik_0R_0} \times b_{2,0}^{(0)}(t')\tilde{b}_{m,0}^{*}(t'',\mathbf{R}_0)e^{i(\omega_k-\omega_0)(t'-t''-R_0/c)} + \text{c.c.}, \tag{32}$$

where $C_m(\mathbf{R}_0)$ is a purely *real* and $D_m(\mathbf{R}_0)$ a complex function of $\mathbf{R}_0$ resulting from the angular integration over Ω_k. In this form, $I_{\omega_k}(t,\mathbf{R}_0;t',t'')$ can be viewed as the interference of the outgoing wave emitted from atom A at time t' with the outgoing wave emitted from atom B at time t''.

In integrating over ω_k to get B_2, one finds that

$$B_2(t,\mathbf{R}_0) = \int_0^t dt' B_2(t,\mathbf{R}_0;t'), \tag{33}$$

where

$$B_2(t,\mathbf{R}_0;t') = 2\pi iC_m(\mathbf{R}_0)e^{ik_0R_0}b_{2,0}^{(0)}(t'-R_0/c)\tilde{b}_{m,0}^{*}(t',\mathbf{R}_0) + \text{c.c.} + 2\pi iD_m(\mathbf{R}_0)e^{-ik_0R_0}b_{2,0}^{(0)}(t' + R_0/c)\tilde{b}_{m,0}^{*}(t',\mathbf{R}_0) + \text{c.c.} \tag{34}$$

When the specific form (17) for $\tilde{b}_{m,0}^{*}$ is inserted into Eq. (34), the $C_m(\mathbf{R}_0)$ term varies as $|b_{2,0}^{(0)}(t'-R_0/c)|^2$; however, this term is purely imaginary and does not contribute to B_2. On the other hand, the $D_m(\mathbf{R}_0)$ term varies as $b_{2,0}^{(0)}(t'+R_0/c)b_{2,0}^{(0)}(t'-R_0/c)$; since this term is complex, it *does* contribute to $B_2(t,\mathbf{R}_0;t')$.

Using Eq. (17), one can write $B_2(t,\mathbf{R}_0;t')$ in the form

$$B_2(t,\mathbf{R}_0;t') = i\tilde{C}(\mathbf{R}_0)|b_{2,0}^{(0)}(t'-R_0/c)|^2 + \text{c.c.} + i\tilde{D}(\mathbf{R}_0)e^{-2ik_0R_0}b_{2,0}^{(0)}(t'+R_0/c)b_{2,0}^{(0)}(t'-R_0/c) + \text{c.c.}, \tag{35}$$

where $\tilde{C}(\mathbf{R}_0)$ is a purely *real* and $\tilde{D}(\mathbf{R}_0)$ a complex function of $\mathbf{R}_0$. It is tempting to interpret the $|b_{2,0}^{(0)}(t'-R_0/c)|^2$ term as a forward scattering term with no phase shift and the $e^{-2ik_0R_0}b_{2,0}^{(0)}(t'+R_0/c)b_{2,0}^{(0)}(t'-R_0/c)$ term as a backward scattering term with an $e^{-2ik_0R_0}$ phase shift. The backward scattering term corresponds to a wave backward scattered from atom B interfering with the wave emitted from atom A at a time R_0/c greater than the scattering time at atom B. In other words, this term corresponds to interference between amplitudes $\tilde{b}_{m,0}^{*}(t',\mathbf{R}_0)\sim b_{2,0}^{(0)}(t'-R_0/c)$ and $b_{2,0}^{(0)}(t'+R_0/c)$. Of course, for this term to contribute, one must have $2\gamma R_0/c \lesssim 1$. Then interpretation in terms of backward and forward scattering, while attractive from a mathematical point of view, is a bit misleading. It is apparent from Eqs. (31) and (30) that there are contributions to B_2 from scattering at all angles.

On the other hand, if $k_0R_0 \gg 1$, most contributions to the Ω_k integral in Eqs. (25a) and (25b) *do* come from the end points of integration, at $\theta=0,\pi$. In this limit and to order (Γ/Δ), there are four distinct processes that contribute to the probability of emission: (1) direct emission by the source atom A; (2) radiation emitted by the source atom interfering with radiation forward scattered from atom B—this term is purely imaginary and does not contribute to the probability; (3) radiation emitted by the source atom interfering with radiation backward scattered from atom B; and (4) radiation emitted by the source atom interfering with radiation backward scattered from atom B and rescattered by atom A. Contributions (3) and (4) cancel one another in the emission probability.

IV. CONSERVATION OF ENERGY

The expectation value of the energy must equal $\hbar\omega_0$ if we start the system with the source atom excited, atom B in its ground state, and no photons in the field. I wish to show that energy is conserved to order $\hbar\Gamma(\Gamma/\Delta)$. From Eqs. (3)–(5) and (8), it follows that conservation of energy can be expressed as

$$\hbar\omega_0|b_{2,0}(t,\mathbf{R}_0)|^2 + \hbar\omega\sum_m |\tilde{b}_{m,0}(t,\mathbf{R}_0)|^2 + \hbar\omega_k|b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2 + \langle V\rangle = \hbar\omega_0. \tag{36}$$

Note that, in contrast to the population calculation, I must keep the term $\hbar\omega|\tilde{b}_{m,0}(t,\mathbf{R}_0)|^2$ since it leads to a contribution to the energy of order $\hbar\Gamma(\Gamma/\Delta)$. By rewriting the equation as

$$\hbar\omega_0\left(|b_{2,0}(t,\mathbf{R}_0)|^2 + \sum_{\mathbf{k}_\lambda}|b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2\right) + \hbar\omega\sum_m |\tilde{b}_{m,0}(t,\mathbf{R}_0)|^2 + \hbar(\omega_k-\omega_0)|b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2 + \langle V\rangle = \hbar\omega_0$$

and using the fact that $(|b_{2,0}(t,\mathbf{R}_0)|^2+\Sigma_{\mathbf{k}_\lambda}|b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2)=1-\Sigma_m|\tilde{b}_{m,0}(t,\mathbf{R}_0)|^2$, one finds that the energy conservation condition can be restated as

$$-\hbar\Delta\sum_m |\tilde{b}_{m,0}(t,\mathbf{R}_0)|^2 + \hbar(\omega_k-\omega_0)|b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2 + \langle V\rangle = 0. \tag{37}$$

I now proceed to evaluate each of these terms separately.

From Eq. (17), one finds

$$-\hbar\Delta\sum_m |\tilde{b}_{m,0}(t,\mathbf{R}_0)|^2 = \hbar\Gamma'(\mathbf{R}_0)|b_{2,0}^{(0)}(t-R_0/c)|^2, \tag{38}$$

where

$$\Gamma'(\mathbf{R}_0) = -\frac{\gamma^2(\mu'/\mu)^2}{\Delta}\sum_m |F_{m;0}(\mathbf{R}_0,k_0)|^2. \tag{39}$$

For $k_0R_0 \gg 1$, $\Gamma'(\mathbf{R}_0)=\Gamma(\mathbf{R}_0)$ [Eq. (21)].

Next I calculate the interaction term. From Eqs. (5) and (8), it follows that

$$\langle V\rangle = \hbar[g_{\mathbf{k}}b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)b_{2,0}^{*}(t,\mathbf{R}_0) + g'_{\mathbf{k}_\lambda}(m)b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)\tilde{b}_{m,0}^{*}(t,\mathbf{R}_0)e^{i\mathbf{k}\cdot\mathbf{R}_0}]e^{-i(\omega_k-\omega_0)t} + \text{c.c.}$$

Substituting expression (17) and the integral of Eq. (12) into this equation, one finds

$$\langle V\rangle = i\hbar\sum_{\mathbf{k}_\lambda}|g_{\mathbf{k}}|^2 b_{2,0}^*(t,\mathbf{R}_0)\int_0^t dt' e^{-i(\omega_k-\omega_0)(t-t')}b_{2,0}(t',\mathbf{R}_0) + \text{c.c.} + i\hbar g_{\mathbf{k}} g'_{\mathbf{k}_\lambda}(m)\tilde{b}_{m,0}^*(t,\mathbf{R}_0)e^{i\mathbf{k}\cdot\mathbf{R}_0}\int_0^t dt' e^{-i(\omega_k-\omega_0)(t-t')} \times b_{2,0}(t',\mathbf{R}_0) + \text{c.c.} - i\hbar g_{\mathbf{k}} g'^*_{\mathbf{k}_\lambda}(m) b_{2,0}^*(t,\mathbf{R}_0) \times e^{-i\mathbf{k}\cdot\mathbf{R}_0}\int_0^t dt' e^{-i(\omega_k-\omega_0)(t-t')}\tilde{b}_{m,0}^*(t',\mathbf{R}_0) + \text{c.c.} \tag{40}$$

The first term is purely imaginary so that the first two terms do not contribute. The second and third terms involve sums very similar to those encountered in the probability calculations and, to order $\hbar\Gamma(\Gamma/\Delta)$, one finds

$$\langle V\rangle = -2\hbar\Gamma'(\mathbf{R}_0)|b_{2,0}^{(0)}(t-\mathbf{R}_0/c)|^2 - 2\hbar\tilde{S}(\mathbf{R}_0) \times b_{2,0}^{(0)}(t)b_{2,0}^{(0)}(t-2R_0/c). \tag{41}$$

I now turn my attention to the second term in Eq. (37), $\Sigma_{\mathbf{k}_\lambda}\hbar(\omega_k-\omega_0)|b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2$. If one used the "normal" prescription for applying the Weisskopf-Wigner approximation in which ω_k is replaced by ω_0 except in phase factors, this term would vanish and energy would not be conserved. Instead, in the integral over ω_k, I adopt an alternative procedure in which ω_k is evaluated at ω_0 except in phase factors *and* in the factor $(\omega_k-\omega_0)$. In some sense, the justification for this procedure is that it will lead to conservation of energy. Using the integral of Eq. (12), one finds that to order $\hbar\Gamma(\Gamma/\Delta)$,

$$\hbar\sum_{\mathbf{k}_\lambda}(\omega_k-\omega_0)|b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2 = K_1 + K_2, \tag{42}$$

where

$$K_1 = \frac{\hbar}{2}\sum_{\mathbf{k}_\lambda}|g_{\mathbf{k}}|^2(\omega_k-\omega_0)\int_0^t dt' b_{2,0}^*(t',\mathbf{R}_0) \times \int_0^t dt'' e^{-i(\omega_k-\omega_0)(t'-t'')}b_{2,0}(t'',\mathbf{R}_0) + \text{c.c.}, \tag{43a}$$

$$K_2 = -\hbar g_{\mathbf{k}} g'_{\mathbf{k}_\lambda}(m)e^{i\mathbf{k}\cdot\mathbf{R}_0}(\omega_k-\omega_0)\int_0^t dt' \tilde{b}_{m,0}^*(t',\mathbf{R}_0) \times \int_0^t dt'' e^{-i(\omega_k-\omega_0)(t'-t'')}b_{2,0}^{(0)}(t'') + \text{c.c.} \tag{43b}$$

Converting the summation over $\mathbf{k}$ to an integral using the modified form of the Weisskopf-Wigner approximation, I find that K_1 is given by [11]

$$\begin{aligned} K_1 &= i\frac{\hbar\gamma}{2\pi}\int_{-\infty}^{\infty} d\bar{\omega}_k\int_0^t dt' b_{2,0}^*(t')\frac{\partial}{\partial t'} \times \int_0^t dt'' e^{-i\bar{\omega}_k(t'-t'')}b_{2,0}(t'') + \text{c.c.} \\ &= i\hbar\gamma\int_0^t dt' b_{2,0}^*(t')\frac{\partial}{\partial t'}b_{2,0}(t') + \text{c.c.} \\ &= i\hbar\gamma\int_0^t dt' b_{2,0}^*(t')[-\gamma b_{2,0}(t') - \{\tilde{\Gamma}(\mathbf{R}_0) + i\tilde{S}(\mathbf{R}_0)\}b_{2,0}^{(0)}(t'-2R_0/c)] + \text{c.c.} \\ &= 2\hbar\tilde{S}(\mathbf{R}_0)\int_0^t dt' \gamma b_{2,0}^{(0)}(t')[b_{2,0}^{(0)}(t'-2R_0/c)] \\ &\approx 2\hbar\tilde{S}(\mathbf{R}_0)\int_0^t dt'[b_{2,0}^{(0)}(t'-2R_0/c)]\frac{\partial}{\partial t'}b_{2,0}^{(0)}(t'). \end{aligned} \tag{44}$$

For K_2, I use Eqs. (6) and (17) to obtain

$$\begin{aligned} K_2 &= -i\hbar g_{\mathbf{k}} g'_{\mathbf{k}_\lambda}(m)\frac{\gamma(\mu'/\mu)}{\Delta}F_{m;0}^*(\mathbf{R}_0,k_0)e^{-ikR_0}e^{i\mathbf{k}\cdot\mathbf{R}_0}(\omega_k-\omega_0) \times \int_0^t dt' b_{2,0}^{(0)}(t'-R_0/c)\int_0^t dt'' e^{-i(\omega_k-\omega_0)(t'-t'')}b_{2,0}^{(0)}(t'') + \text{c.c.} \\ &= -i\frac{\hbar\Gamma'(\mathbf{R}_0)}{2\pi}\int_{-\infty}^{\infty} d\bar{\omega}_k\bar{\omega}_k\int_0^t dt' b_{2,0}^{(0)}(t'-R_0/c) \times \int_0^t dt'' e^{-i\bar{\omega}_k(t'-t''-R_0/c)}b_{2,0}^{(0)}(t'') + \text{c.c.} \\ &\quad + \frac{\hbar\{\tilde{\Gamma}(\mathbf{R}_0)+i\tilde{S}(\mathbf{R}_0)\}}{2\pi}\int_{-\infty}^{\infty} d\bar{\omega}_k\bar{\omega}_k\int_0^t dt'' b_{2,0}^{(0)}(t'') \times \int_0^t dt' e^{i\bar{\omega}_k(t''-t'-R_0/c)}b_{2,0}^{(0)}(t'-R_0/c) + \text{c.c.} \\ &= \frac{\hbar\Gamma'(\mathbf{R}_0)}{2\pi}\int_{-\infty}^{\infty} d\bar{\omega}_k\int_0^t dt' b_{2,0}^{(0)}(t'-R_0/c)\frac{\partial}{\partial t'} \times \int_0^t dt'' e^{-i\bar{\omega}_k(t'-t''-R_0/c)}b_{2,0}^{(0)}(t'') + \text{c.c.} \\ &\quad - \frac{i\hbar\{\tilde{\Gamma}(\mathbf{R}_0)+i\tilde{S}(\mathbf{R}_0)\}}{2\pi}\int_{-\infty}^{\infty} d\bar{\omega}_k\int_0^t dt'' b_{2,0}^{(0)}(t'')\frac{\partial}{\partial t''} \times \int_0^t dt' e^{i\bar{\omega}_k(t''-t'-R_0/c)}b_{2,0}^{(0)}(t'-R_0/c) + \text{c.c.} \\ &= \hbar\Gamma'(\mathbf{R}_0)\int_0^t dt'\frac{\partial}{\partial t'}|b_{2,0}^{(0)}(t'-R_0/c)|^2 + 2\hbar\tilde{S}(\mathbf{R}_0) \times \int_0^t dt'' b_{2,0}^{(0)}(t'')\frac{\partial}{\partial t''}b_{2,0}^{(0)}(t''-2R_0/c). \end{aligned} \tag{45}$$

Combining Eqs. (44), (45), and (42), I obtain finally

$$\sum_{\mathbf{k}_\lambda}\hbar(\omega_k-\omega_0)|b_{\mathbf{k}_\lambda}(t,\mathbf{R}_0)|^2 = \hbar\Gamma'(\mathbf{R}_0)|b_{2,0}^{(0)}(t-R_0/c)|^2 + 2\hbar\tilde{S}(\mathbf{R}_0)b_{2,0}^{(0)}(t)b_{2,0}^{(0)}(t-2R_0/c). \tag{46}$$

It then follows from Eqs. (37), (38), (41), and (46) that energy is conserved.

The results can be summarized as follows: The average energy in atom B is

$$E_B = -\hbar\omega\frac{\Gamma'(\mathbf{R}_0)}{\Delta}|b_{2,0}^{(0)}(t-R_0/c)|^2, \tag{47}$$

the interaction energy is

$$\langle V\rangle = -2\hbar\Gamma'(\mathbf{R}_0)|b_{2,0}^{(0)}(t-\mathbf{R}_0/c)|^2 - 2\hbar\tilde{S}(\mathbf{R}_0)b_{2,0}^{(0)}(t)b_{2,0}^{(0)}(t-2R_0/c), \tag{48}$$

and the energy of the source atom plus the field is

$$E_A + E_F = \hbar\omega_0 + \hbar\omega\frac{\Gamma'(\mathbf{R}_0)}{\Delta}|b_{2,0}^{(0)}(t-R_0/c)|^2 + 2\hbar\Gamma'(\mathbf{R}_0)|b_{2,0}^{(0)}(t-R/c)|^2 + 2\hbar\tilde{S}(\mathbf{R}_0)b_{2,0}^{(0)}(t)b_{2,0}^{(0)}(t-2R_0/c). \tag{49}$$

If we look in the limit $\gamma(t-R_0/c)\lesssim 1$ and $\gamma R_0/c \gg 1$, then

$$E_F \sim \hbar\omega_0 + \hbar\omega\frac{\Gamma'(\mathbf{R}_0)}{\Delta}|b_{2,0}^{(0)}(t-R_0/c)|^2 + \hbar\Gamma'(\mathbf{R}_0)|b_{2,0}^{(0)}(t-R_0/c)|^2. \tag{50}$$

This limit corresponds to times for which the field is totally emitted from atom A and is being scattered by atom B, but has had insufficient time to rescatter from atom A. The second term in Eq. (50) corresponds to a reduction in field energy resulting from the excitation of atom B [since $\Gamma'(\mathbf{R}_0)$ is positive for negative Δ, this term is negative, regardless of the sign of Δ]. The third term in Eq. (50) is much smaller than the second and corresponds to an *increase* in field energy for $\Delta<0$. One interpretation of this term is that the field produces a light or ac Stark shift that lowers the energy of atom B; as a consequence the energy of the field increases. Based on Maxwell's equations, one usually concludes that it is the propagation vector rather than the frequency that changes when radiation propagates in a dielectric medium. In this problem, as well as in pulse propagation in a dielectric, there is nothing inconsistent with the fact that the average frequency in the field changes while it interacts with dielectric atoms, compared to the average frequency in vacuum.

Another interpretation of the third term in Eq. (50) can be given in terms of the frequency distribution (32), which, with the aid of Eq. (17), can be written as

$$I_{\omega_k}(t,\mathbf{R}_0) = C(\mathbf{R}_0)O_1(\omega_k-\omega_0) + \mathrm{Re}[D(\mathbf{R}_0)e^{-2ik_0R_0}] \times O_2(\omega_k-\omega_0) + \mathrm{Im}[D(\mathbf{R}_0)e^{-2ik_0R_0}]E(\omega_k-\omega_0), \tag{51}$$

where the real functions $O_i(x)$ $(i=1,2)$ and $E(x)$ are even and odd functions of x, respectively. In the limit $\gamma t \gg 1$ and $\gamma R_0/c \gg 1$, the second and third terms do not contribute since they are linked to a contribution varying as an integral of $b_{2,0}^{(0)}(t''-R_0/c)b_{2,0}^{(0)}(t''+R_0/c)$. On the other hand, the first term, while not contributing to the probability since it is an odd function of $(\omega_k-\omega_0)$, *does* contribute to the average value of $(\omega_k-\omega_0)$; in this way the average field energy increases as a result of the interference term.

V. SUMMARY

I have given an explicit calculation of conservation of probability and energy in a two-atom system. One of the atoms is excited initially and the spontaneous radiation emitted by this atom can be scattered by the second atom. It was seen that the Weisskopf-Wigner approximation must be applied in a specific manner if both probability and energy conservation is to be guaranteed. In a future planned work, I will extend this calculation to include the recoil the atoms undergo in the emission and scattering problem. This extension brings in yet another package of problems. The results must certainly be independent of the choice of the initial wave packets of the atoms, but the scattering and rescattering seem to depend in a sensitive way on this choice.

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[9] In considering the field emitted by an atomic dipole, one can justify extension of the frequency integral to minus infinity by including non-energy-conserving terms in the calculation. The resulting integrals are perfectly convergent and lead to expressions for the average field that take the same form as the classical results. Thus, no Weisskopf-Wigner approximation is

needed to obtain expressions for the field in terms of the dipole operator and its derivatives—see, for example, P. R. Berman, Phys. Rev. A **69**, 022101 (2004), and references therein.

[10] If the atom is excited "instantaneously," one can question the use of the adiabatic hypothesis. To avoid such problems, one can assume that the source atom is excited to its initial state by an optical pulse whose duration is much longer than $|\Delta|^{-1}$. The pulse duration is also assumed to be much shorter than γ^{-1}, enabling one to neglect any radiation *during* the excitation pulse. The only property of $b_{2,0}^{(0)}(t)$ used in this paper is that it is real and this property is in no way restrictive.

[11] In both Eqs. (44) and (45) one encounters terms of the form

$$\frac{\partial}{\partial t'}\int_0^t dt'' \delta(t'-t'')b_{2,0}(t'')$$

$$= \frac{\partial}{\partial t'}[b_{2,0}(t')\Theta(t-t')\Theta(t')]$$

$$= \Theta(t-t')\Theta(t')\frac{\partial}{\partial t'}b_{2,0}(t') - b_{2,0}(t)\delta(t-t')\Theta(t')$$

$$+ b_{2,0}(0)\Theta(t-t')\delta(t').$$

It can be shown, however, that if one simply sets

$$\frac{\partial}{\partial t'}\int_0^t dt'' \delta(t'-t'')b_{2,0}(t'') = \frac{\partial}{\partial t'}b_{2,0}(t')$$

in Eqs. (44) and (45) as I have done, the final results are the same that are obtained using the complete expression for the time derivatives of the integrals.