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Retardation in the resonant interaction of two identical atoms*

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The problem considered is that of two identical two-level atoms a fixed distance r apart, one of which is excited at $t=0$. A simple improvement on the usual on-shell approximation (which in the single-atom case is part of the Weisskopf-Wigner approximation) yields new solutions for the various probability amplitudes in the form of infinite series involving all the retardation times nr/c. The truncated solutions involving only the single retardation time r/c are compared with previously published results both when all photon modes are allowed and when only photons propagating along the interatomic axis are allowed. When the retardation times are neglected, the series are summed to give the well-known results of Stephen and others.

I. INTRODUCTION

One of the classic problems in the quantum theory of radiation is to show that the mutual influence of two systems interacting through the electromagnetic radiation field propagates with the velocity of light. The problem is usually formulated in terms of two atoms a fixed distance r apart, atom 1 in the first excited state, atom 2 in the ground state, and the electromagnetic field in the vacuum state. Using the solution of Weisskopf and Wigner[1] for the spontaneous emission from atom 1, several authors[2] showed that atom 2 has vanishing probability of being excited until time r/c. The problem was reexamined following a suggestion by Ferretti and Peierls[3] that quantum electrodynamics may not provide a correct solution. Hamilton[4] and Heitler and Ma[5] answered this objection and again showed that atom 2 has nonvanishing probability of being excited only after time $t=r/c$. The problem is now textbook material.[6]

In a different context, the two-atom system with one excitation is again of fundamental importance. Lord Rayleigh[7] first suggested that resonance broadening due to identical atoms is of greater magnitude than the broadening due to the presence of foreign atoms. Dicke[8] was the first to emphasize the cooperative nature of the spontaneous emission from a system of identical sources. As an example, he considered the two-atom case with one atom excited, the atoms being located within a transition wavelength of each other. He pointed out that there are two equally likely possibilities: the system will either decay at twice the single-atom rate, or it will be nonradiative. Stephen[9] and others[10] further studied this simplest example of cooperative decay and expressed the cooperative linewidth and line shift as a function of interatomic separation. The problem continues to be of interest, especially in the light of increasing efforts to study "superradiance" experimentally.[11]

It appears to us that the second aspect of the two-atom problem is now better understood than the first. The causal aspect of the problem deserves more careful study, especially in view of its fundamental importance. The solutions presented by several authors[2] for the various probability amplitudes are discontinuous in time, exhibiting a step-function behavior at $t=r/c$; the same departure from the continuity demanded by the Schrödinger equation is reproduced in a popular textbook.[6] Furthermore, even the continuous solution given by Hamilton[4] is only valid for times less than $2r/c$; it does not demonstrate the possibility of atom 1 absorbing its emitted photon after it was absorbed and then emitted by atom 2. It would be desirable to have a solution that is not only continuous, but shows how all the retardation times nr/c enter. When all the retardation times are neglected, it should reduce to the results of Stephen[9] for cooperative decay.

Such a solution has been briefly reported recently.[12] The results of that work are derived here in detail and extended. The solution unifies the two traditionally separate aspects of the two-atom problem discussed above. In Sec. II we introduce our notation and briefly review the results of Stephen. In Sec. III we show how the approach may be modified to show the dependence upon the retardation times nr/c. The solution reduces to Hamilton's when the higher order processes depending upon $nr/c, n\geq 2$, are omitted. In Sec. IV we consider the problem treated by Arecchi and Courtens,[13] in which only photons propagating along the interatomic axis are allowed. Some concluding remarks are made in Sec. V.

II. RESONANT INTERACTION IN FREE SPACE

Consider two identical one-electron atoms separated by a distance r in the free-space vacuum. Suppose that at $t=0$, atom 1 is in the first excited state and atom 2 is in the ground state (we shall not be concerned with how such a system was initially excited). The ground state is taken to be a nondegenerate s state from which no real spontaneous transitions are possible, while the first excited state is a triply degenerate p state. Since we are not interested in single-atom radiative frequency shifts here, we may use the two-level model of the atom as a reasonable approximation.[14] While this assumption ignores spatial degeneracy, no real difficulty is introduced, since the three transition possibilities $\Delta m=0, \pm 1$ may be treated separately. Thus, the "excited state" at this point need not carry a specific value of m.

The Hamiltonian for this system is

$$H=H_A+H_F+H_{\mathrm{INT}}, \tag{1}$$

where H_A and H_F are, respectively, the atomic and field Hamiltonians, and H_{INT} is the interaction Hamiltonian[15] (see Appendix A)

$$H_{\mathrm{INT}}=-\vec{\mu}_1\cdot\vec{\mathrm{E}}(\vec{\mathrm{x}}_1)-\vec{\mu}_2\cdot\vec{\mathrm{E}}(\vec{\mathrm{x}}_2). \tag{2}$$

Here, $\vec{\mu}_l$ is the transition dipole moment operator $\vec{\mu}_l\equiv e\vec{\mathrm{x}}_l$ for atom l, and $\vec{\mathrm{E}}(\vec{\mathrm{x}}_l)$ is the electric field operator evaluated at the position $\vec{\mathrm{x}}_l$ of atom l[16];

$$\vec{\mathrm{E}}(\vec{\mathrm{x}})=i\sum_{\vec{\mathrm{k}}_\lambda}\left(\frac{2\pi\hbar\omega_k}{V}\right)^{1/2}[a(\vec{\mathrm{k}}_\lambda)e^{i\vec{\mathrm{k}}\cdot\vec{\mathrm{x}}}\hat{\epsilon}(\vec{\mathrm{k}}_\lambda)-\mathrm{H.c.}], \tag{3}$$

where V is the quantization volume; $\hat{\epsilon}(\vec{\mathrm{k}}_\lambda)$ is a polarization unit vector, $\vec{\mathrm{k}}\cdot\hat{\epsilon}(\vec{\mathrm{k}}_\lambda)=0$, $\lambda=1,2$; and $a(\vec{\mathrm{k}}_\lambda)$ and $a^\dagger(\vec{\mathrm{k}}_\lambda)$ are, respectively, the photon annihilation and creation operators for mode $(\vec{\mathrm{k}}_\lambda)$.

The Hamiltonian (1) may be written, using the two-level approximation, as

$$H=H_A+H_F-i\hbar\sum_{l=1,2}\sum_{\vec{\mathrm{k}}_\lambda}[g_l(\vec{\mathrm{k}}_\lambda)\sigma_l^-+g_l^*(\vec{\mathrm{k}}_\lambda)\sigma_l^+]\times[a(\vec{\mathrm{k}}_\lambda)-a^\dagger(\vec{\mathrm{k}}_\lambda)], \tag{4}$$

where

$$g_l(\vec{\mathrm{k}}_\lambda)\equiv\hbar^{-1}(2\pi\hbar\omega_k/V)^{1/2}[\vec{\mu}_l\cdot\hat{\epsilon}(\vec{\mathrm{k}}_\lambda)]e^{i\vec{\mathrm{k}}\cdot\vec{\mathrm{x}}_l}. \tag{5}$$

We have taken a real polarization basis, but the transition moments

$$\vec{\mu}_l\equiv{}_l\langle -|e\vec{\mathrm{x}}_l|+\rangle_l=\mu\hat{\mu}_l \tag{6}$$

may be complex. As usual, $|-\rangle_l$ and $|+\rangle_l$ are, respectively, the ground- and excited-state vectors of atom l. The operators $\sigma_l^\pm$ are defined by

$$\sigma_l^-=|-\rangle_l\,{}_l\langle +|,\quad \sigma_l^+=|+\rangle_l\,{}_l\langle -|. \tag{7}$$

We may write the state vector for the system in the Schrödinger picture as

$$|\psi(t)\rangle=\sum_m b_m(t)|\phi_m\rangle, \tag{8}$$

where the $|\phi_m\rangle$ are eigenstates of the unperturbed system

$$(H_A+H_F)|\phi_m\rangle=E_m|\phi_m\rangle.$$

Then the Schrödinger equation becomes (upon using orthonormality)

$$i\hbar\dot{b}_n(t)=E_n b_n(t)+\sum_m b_m(t)\langle\phi_n|H_{\mathrm{INT}}|\phi_m\rangle. \tag{9}$$

With the given initial conditions, an approximate complete set of states is given by[9]

$$|\phi_1\rangle=|+\rangle_1|-\rangle_2|\{0\}\rangle,\quad E_1=0, \tag{10a}$$

$$|\phi_2\rangle=|-\rangle_1|+\rangle_2|\{0\}\rangle,\quad E_2=0, \tag{10b}$$

$$|\phi(\vec{\mathrm{k}}_{\lambda1})\rangle=|-\rangle_1|-\rangle_2|1_{\vec{\mathrm{k}}_\lambda}\rangle,\quad E(\vec{\mathrm{k}}_{\lambda1})=\hbar(\omega_k-\omega_0), \tag{10c}$$

$$|\phi(\vec{\mathrm{k}}_{\lambda2})\rangle=|+\rangle_1|+\rangle_2|1_{\vec{\mathrm{k}}_\lambda}\rangle,\quad E(\vec{\mathrm{k}}_{\lambda2})=\hbar(\omega_k+\omega_0). \tag{10d}$$

The states $|\phi_1\rangle$ and $|\phi_2\rangle$ have one atom excited and the radiation field in the vacuum state, while $|\phi(\vec{\mathrm{k}}_{\lambda1})\rangle$ has both atoms in the ground state and one photon in mode $(\vec{\mathrm{k}}_\lambda)$. The state $|\phi(\vec{\mathrm{k}}_{\lambda2})\rangle$ has both atoms excited and one photon in mode $(\vec{\mathrm{k}}_\lambda)$; $|\phi(\vec{\mathrm{k}}_{\lambda2})\rangle$ would be omitted in the rotating-wave approximation (RWA), or resonant approximation[17] of replacing Eq. (4) with

$$H_{\mathrm{RWA}}=H_A+H_F-i\hbar\sum_{l=1,2}\sum_{\vec{\mathrm{k}}_\lambda}[g_l^*(\vec{\mathrm{k}}_\lambda)\sigma_l^+a(\vec{\mathrm{k}}_\lambda)-g_l(\vec{\mathrm{k}}_\lambda)\sigma_l^-a^\dagger(\vec{\mathrm{k}}_\lambda)]. \tag{11}$$

This approximation admits only processes corresponding to the emission of a photon and the simultaneous lowering of an atomic state, and vice versa. The rotating-wave approximation is discussed further in Appendix A.

With the states (10), the Schrödinger equation takes the form

$$\dot{b}_1(t)=-\sum_{\vec{\mathrm{k}}_\lambda}g_1^*(\vec{\mathrm{k}}_\lambda)b_{\vec{\mathrm{k}}_{\lambda1}}(t)-\sum_{\vec{\mathrm{k}}_\lambda}g_2(\vec{\mathrm{k}}_\lambda)b_{\vec{\mathrm{k}}_{\lambda2}}(t), \tag{12a}$$

$$\dot{b}_2(t) = -\sum_{\vec{k}_\lambda} g_2^*(\vec{k}_\lambda) b_{\vec{k}_{\lambda 1}}(t) - \sum_{\vec{k}_\lambda} g_1(\vec{k}_\lambda) b_{\vec{k}_{\lambda 2}}(t), \tag{12b}$$

$$\dot{b}_{\vec{k}_{\lambda 1}}(t) = -i(\omega_k - \omega_0) b_{\vec{k}_{\lambda 1}}(t) + g_1(\vec{k}_\lambda) b_1(t) + g_2(\vec{k}_\lambda) b_2(t), \tag{12c}$$

$$\dot{b}_{\vec{k}_{\lambda 2}}(t) = -i(\omega_k + \omega_0) b_{\vec{k}_{\lambda 2}}(t) + g_2^*(\vec{k}_\lambda) b_1(t) + g_1^*(\vec{k}_\lambda) b_2(t), \tag{12d}$$

with initial conditions $b_1(0) = 1$, $b_2(0) = b_{\vec{k}_{\lambda 1}}(0) = b_{\vec{k}_{\lambda 2}}(0) = 0$. To solve this set of coupled equations, we Laplace transform the set (12) using these initial conditions to obtain

$$\hat{b}_1(s) = [s + A(s)] / \{[s + A(s)]^2 - B^2(s)\}, \tag{13}$$

$$\hat{b}_2(s) = -B(s) / \{[s + A(s)]^2 - B^2(s)\}, \tag{14}$$

with

$$\hat{b}(s) \equiv \int_0^\infty dt\, e^{-st} b(t),$$

and where[18]

$$A(s) = \sum_{\vec{k}_\lambda} |g_1(\vec{k}_\lambda)|^2 \left(\frac{1}{s + i(\omega_k - \omega_0)} + \frac{1}{s + i(\omega_k + \omega_0)} \right), \tag{15}$$

$$B(s) = \sum_{\vec{k}_\lambda} g_1^*(\vec{k}_\lambda) g_2(\vec{k}_\lambda) \left(\frac{1}{s + i(\omega_k - \omega_0)} + \frac{1}{s + i(\omega_k + \omega_0)} \right). \tag{16}$$

The function $B(s)$ given by Eq. (16) will lead to the r-dependent complex cooperative self-energy describing the coupling between the atoms. The function $A(s)$ given by Eq. (15) will lead to the r-independent single-atom self-energies; it describes the self-energy of the excited state of atom 1 *and* the self-energy of the ground state of atom 2. Despite superficial resemblances, it does *not* lead to the shift in the transition frequency of atom 1; the antiresonant term involving $\omega_k + \omega_0$ represents the level shift of the ground state of atom 2, not the shift in the ground-state energy level of atom 1. Thus, $A(s)$ together with $B(s)$ yields the energy-level-shift parameter of the initial state $|\phi_1\rangle$. To obtain the frequency-shift parameter involved in the decay, we must subtract from this the level shift of the state with two ground-state atoms and no photons. The r-dependent shift produced by the coupling of two ground-state atoms is the van der Waals energy, and is negligible compared with the first-order dispersion effects we are concerned with. Hence, we may equate the level shift derived from $B(s)$ with the frequency shift of the transition of the coupled system.

The r-independent self-energy of the two-ground-state atoms involves two equal terms in $\omega_k + \omega_0$, and represents the effects of wholly virtual transitions from the ground state of *each* atom. One of these terms will cancel the ground-state nonresonant term in the shift of $|\phi_1\rangle$, the other will enter with the correct sign to give the shift in the transition frequency of atom 1. Symbolically,

$$[(\omega_k - \omega_0)^{-1} + (\omega_k + \omega_0)^{-1}] - [(\omega_k + \omega_0)^{-1} + (\omega_k + \omega_0)^{-1}] = (\omega_k - \omega_0)^{-1} - (\omega_k + \omega_0)^{-1}$$

and

$$\Delta = \sum_{\vec{k}_\lambda} |g_1(\vec{k}_\lambda)|^2 \left(\frac{1}{\omega_k - \omega_0} - \frac{1}{\omega_k + \omega_0} \right)$$

is the single-atom frequency shift. Thus, as $r \to \infty$ $[B(s) \to 0]$ in the all-mode case, the shift in the central resonance frequency from a system in initial state $|\phi_1\rangle$ is independent of the presence of the remote ground-state atom, as it should be. As long as we are not interested in single-atom frequency shifts (for which the two-level approximation is a poor representation anyway), the level shift which results from $B(s)$ can be regarded as a frequency shift, since all other contributions are either irrelevant or negligible.

It will prove useful to define

$$C_\pm(s) = 1/[s + A(s) \pm B(s)], \tag{17}$$

so that

$$b_1(t) = \tfrac{1}{2}[C_+(t) + C_-(t)] \tag{18}$$

and

$$b_2(t) = \tfrac{1}{2}[C_+(t) - C_-(t)], \tag{19}$$

where

$$C_\pm(t) = \frac{1}{2\pi i} \int_{-i\infty+\epsilon}^{i\infty+\epsilon} ds\, \frac{e^{st}}{s + A(s) \pm B(s)}. \tag{20}$$

The path of integration is parallel to the imaginary axis, and ϵ is a small positive number. Using $\epsilon \ll 1$, we may write Eq. (20) as

$$C_\pm(t) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{e^{izt}\, dz}{z - iA(iz + \epsilon) \mp iB(iz + \epsilon)}. \tag{21}$$

If we now consider all modes of the radiation field available for spontaneous emission (we shall later restrict the number of modes in Sec. IV) and go to the free-space continuum limit

$$\sum_{\vec{k}_\lambda} \to \frac{V}{(2\pi)^3}\int d^3k \sum_{\lambda=1,2},$$

we obtain from Eqs. (15) and (16) (using the relative dipole approximation[18]),

$$A(iz+\epsilon) = -\frac{2i\mu^2}{3\pi\hbar c^3}\left(\int_0^\infty \frac{\omega^3 d\omega}{\omega-\omega_0+z-i\epsilon} + \int_0^\infty \frac{\omega^3 d\omega}{\omega+\omega_0+z-i\epsilon}\right) \tag{22}$$

and

$$B(iz+\epsilon) = -\frac{i\mu^2}{\pi\hbar c^3}\left(\int_0^\infty \frac{\omega^3 f(\omega r/c)\, d\omega}{\omega-\omega_0+z-i\epsilon} + \int_0^\infty \frac{\omega^3 f(\omega r/c)\, d\omega}{\omega+\omega_0+z-i\epsilon}\right). \tag{23}$$

The function $f(\omega r/c)$ contains the interatomic separation, and is given by

$$f(x) = p\,\sin x/x + q(\sin x/x^3 - \cos x/x^2), \tag{24}$$

where

$$\begin{aligned} &p=0,\quad q=2 \quad \text{for } \Delta m = 0 \text{ transitions},\\ &p=1,\quad q=-1 \quad \text{for } \Delta m = \pm 1 \text{ transitions}. \end{aligned} \tag{25}$$

The quantization axis has been taken to be the interatomic axis. The details of the derivation of Eqs. (24) and (25) are given in Appendix B.

The above has served to present our notation and method of approach. At this point, the usual analysis (when concerned with the cooperative aspect of the two-atom system) proceeds by making a pole, or "on-shell," approximation in the spirit of the Weisskopf-Wigner approach to single-atom spontaneous emission. The pole approximation[19] for single-atom emission consists of neglecting the z dependence of $A(iz+\epsilon)$, which may be generalized in our case to also neglecting the z dependence of $B(iz+\epsilon)$. Our new solution is a result of going outside the pole, or "on-shell," approximation, and keeping the leading z-dependent effects.

For purposes of comparison, we shall derive the on-shell results in this section. Using this approximation in Eq. (22), and making use of the prescription

$$\frac{1}{x-x_0-i\epsilon} = i\pi\delta(x-x_0) + P\left(\frac{1}{x-x_0}\right), \tag{26}$$

we obtain

$$A(iz+\epsilon)|_{z=0} = \frac{2\mu^2\omega_0^3}{3\hbar c^3} = \beta, \tag{27}$$

where β is one-half the Einstein A coefficient. In obtaining Eq. (27), we have neglected all single-atom radiative level shifts[20]; these are irrelevant for our purposes.

If we use the same approximation in the interatomic term $B(iz+\epsilon)$, we have

$$\begin{aligned} B(iz+\epsilon)|_{z=0} &= -\frac{i\mu^2}{\pi\hbar c^3}\left(\int_0^\infty \frac{\omega^3 f(\omega r/c)\, d\omega}{\omega-\omega_0-i\epsilon} + \int_0^\infty \frac{\omega^3 f(\omega r/c)\, d\omega}{\omega+\omega_0-i\epsilon}\right)\\ &= -\frac{i\mu^2}{\pi\hbar c^3}\int_{-\infty}^\infty \frac{\omega^3 f(\omega r/c)\, d\omega}{\omega-\omega_0-i\epsilon}. \end{aligned}$$

This integral may be evaluated to give

$$B(iz+\epsilon)|_{z=0} = -\tfrac{3}{2}\beta e^{ik_0 r}\left(\frac{ip}{k_0 r} + \frac{iq}{k_0^3 r^3} + \frac{q}{k_0^2 r^2}\right), \tag{28}$$

where $k_0 = \omega_0/c$.

Thus, in the pole approximation, we obtain from Eq. (21) [using the results (27) and (28)],

$$C_\pm(t) = \exp\{-\beta[1 \pm g(r)]\,t\}, \tag{29}$$

where

$$g(r) \equiv \frac{3}{2}\left(p\frac{\sin k_0 r}{k_0 r} + q\frac{\sin k_0 r}{k_0^3 r^3} - q\frac{\cos k_0 r}{k_0^2 r^2}\right) - \frac{3i}{2}\left(p\frac{\cos k_0 r}{k_0 r} + q\frac{\cos k_0 r}{k_0^3 r^3} + q\frac{\sin k_0 r}{k_0^2 r^2}\right). \tag{30}$$

Then from Eqs. (18) and (19), the state probability amplitudes $b_1(t)$ and $b_2(t)$ are

$$b_1(t) = \tfrac{1}{2}(\exp\{-\beta[1+g(r)]\,t\} + \exp\{-\beta[1-g(r)]\,t\}), \tag{31}$$

$$b_2(t) = \tfrac{1}{2}(\exp\{-\beta[1+g(r)]\,t\} - \exp\{-\beta[1-g(r)]\,t\}). \tag{32}$$

The probability amplitudes $b_1(t)$ and $b_2(t)$ are those appropriate for our initial conditions, when we know for certain at $t=0$ which atom is excited. When the atoms are sufficiently close together, this initial condition is unrealistic. An alternative initial state, which allows either atom 1 or atom 2 to be excited with equal probability at $t=0$, is

$$|\phi_\pm\rangle = \frac{1}{\sqrt{2}}(|\phi_1\rangle \pm |\phi_2\rangle), \tag{33}$$

where $|\phi_{1,2}\rangle$ are given by Eqs. (10a) and (10b). Using the correlated states $|\phi_\pm\rangle$, it is easy to show that $C_\pm(t)$ is the amplitude for remaining in the initial state. Then the state amplitudes for $|\phi_\pm\rangle$ decay at a rate

$$\beta_\pm = \beta[1 \pm \mathrm{Re}\,g(r)], \tag{34}$$

which in the limit $k_0 r \ll 1$ becomes

$$\beta_\pm = \beta(1 \pm \tfrac{3}{2}p \pm \tfrac{1}{2}q) = \beta(1 \pm 1) \tag{35}$$

for either $\Delta m = 0$ or for $\Delta m = \pm 1$ transitions. In the symmetric state $|\phi_+\rangle$ the rate of spontaneous emission is doubled, whereas the antisymmetric state $|\phi_-\rangle$ is nonradiative. This is exactly the result of Dicke[8] for the special case of two atoms; the generalization (34) for arbitrary $k_0 r$ is well known from the work of Stephen[9] and others.[10] The imaginary part of $g(r)$ represents a radiative level shift and is similarly well known.[9,10,21]

III. RETARDATION IN RESONANT INTERACTION

While the solutions in Sec. II are well known, they possess some disagreeable features. The main one is the absence of retardation times nr/c; because of the approximations employed in solving the equations of motion, we have neglected the causal propagation time. Each atom is allowed to influence the other instantaneously. The limitations of this approximation have been stressed by Arecchi and Courtens.[13] We now consider an improvement over the pole approximation used in Sec. II, and show how the retardation times appear naturally from our approach. In this paper, we are not concerned with the features of purely single-atom spontaneous emission,[22] so we will continue to make the pole approximation on the single-atom self-energy term $A(iz+\epsilon)$. We may use the prescription (26) to decompose the interatomic term $B(iz+\epsilon)$ to

$$\begin{aligned} B(iz+\epsilon) &= -\frac{i\mu^2}{\pi\hbar c^3}\left(\int_0^\infty \frac{\omega^3 f(\omega r/c)\,d\omega}{\omega-\omega_0+z-i\epsilon} + \int_{-\infty}^0 \frac{\omega^3 f(\omega r/c)\,d\omega}{\omega-\omega_0-z+i\epsilon}\right) \\ &= -\frac{i\mu^2}{\pi\hbar c^3}\int_0^\infty d\omega\,\omega^3 f(\omega r/c)\,[i\pi\delta(\omega-\omega_0+z)+P(1/\omega-\omega_0+z)] \\ &\quad -\frac{i\mu^2}{\pi\hbar c^3}\int_{-\infty}^0 d\omega\,\omega^3 f(\omega r/c)[-i\pi\delta(\omega-\omega_0-z)+P(1/\omega-\omega_0-z)]. \end{aligned} \tag{36}$$

Rather than neglect the z dependence of $B(iz+\epsilon)$ entirely,[23] we shall assume $|z| \ll \omega_0$ in Eq. (36). Then the third term on the right-hand side of Eq. (36) may be neglected. Furthermore, we shall replace the term $P(1/\omega-\omega_0-z)$ by $P(1/\omega-\omega_0+z)$ (see Appendix C). Then

$$\begin{aligned} B(iz+\epsilon) &\simeq \frac{\mu^2}{\hbar c^3}\int_{-\infty}^\infty d\omega\,\omega^3 f\left(\frac{\omega r}{c}\right)\delta(\omega-\omega_0+z) \\ &\quad -\frac{i\mu^2}{\pi\hbar c^3}P\int_{-\infty}^\infty \frac{\omega^3 f(\omega r/c)}{\omega-\omega_0+z}\,d\omega. \end{aligned} \tag{37}$$

Since the integrals now run over a common range, and the arguments of the principal part and delta function are identical, Eq. (37) can be written as

$$B(iz+\epsilon) \simeq -\frac{i\mu^2}{\pi\hbar c^3}\int_{-\infty}^\infty \frac{\omega^3 f(\omega r/c)\,d\omega}{\omega-\omega_0+z-i\epsilon}.$$

Within the full on-shell or pole approximation ($z \to 0$), the step from Eq. (36) to Eq. (37) is exact. The "semi-pole" approximation involved in replacing Eq. (36) by Eq. (37) is discussed in more detail in Appendix C.

Evaluating $B(iz+\epsilon)$ from Eq. (37) gives

$$B(iz+\epsilon) = -(\mu^2/\hbar c^3)(\omega_0-z)^3 f_1[(\omega_0-z)r/c], \tag{38}$$

where

$$f_1(x) = e^{ix}(ip/x + iq/x^3 + q/x^2). \tag{39}$$

Since the principal effect of retaining z will be in the exponent, we shall neglect z everywhere except in the exponential in $f_1(x)$,

$$\begin{aligned} B(iz+\epsilon) &\simeq -(\tfrac{3}{2}\beta)e^{i(\omega_0-z)r/c} \\ &\quad \times (ip/k_0 r + iq/k_0^3 r^3 + q/k_0^2 r^2). \end{aligned} \tag{40}$$

To facilitate comparison with earlier work, we assume $k_0 r \gg 1$ so that from Eq. (21)

$$\begin{aligned} C_\pm(t) &= \frac{1}{2\pi i}\int_{-\infty}^\infty dz\, e^{izt}[z - i\beta \pm (\tfrac{3}{2}i\beta)e^{i(\omega_0-z)r/c} \\ &\quad \times (ip/k_0 r + iq/k_0^3 r^3 + q/k_0^2 r^2)]^{-1}. \end{aligned} \tag{41}$$

Since $|B(iz+\epsilon)/(z-i\beta)| < 1$ for all z, the integrand may be expanded into a power series

$$\begin{aligned} C_\pm(t) &= \frac{1}{2\pi i}\int_{-\infty}^\infty dz\, e^{izt} \\ &\quad \times \sum_{n=0}^\infty \left[\pm(\tfrac{3}{2}\beta)\left(\frac{p}{k_0 r} + \frac{q}{k_0^3 r^3} - \frac{iq}{k_0^2 r^2}\right)\right]^n \\ &\quad \times \frac{e^{ink_0 r}\, e^{-inzr/c}}{(z-i\beta)^{n+1}}. \end{aligned}$$

Actually, our method applies if only $k_0 r \gtrsim 3$ for either $\Delta m = 0$ or $\Delta m = \pm 1$ transitions. This means that the atoms must be more than half of a transition wavelength apart. For separations less than this (where retardation does not play an important role), we can extend our method, but in this case it is necessary to include single-atom level shifts. Provided this restriction on r is satisfied

$$C_\pm(t)=\frac{1}{2\pi i}\sum_{n=0}^{\infty}\left[\pm\tfrac{3}{2}\beta\left(\frac{p}{k_0r}+\frac{q}{k_0^3r^3}-\frac{iq}{k_0^2r^2}\right)\right]^n e^{ink_0r}\int_{-\infty}^{\infty}\frac{\exp[iz(t-nr/c)]\,dz}{(z-i\beta)^{n+1}}$$

$$=\sum_{n=0}^{\infty}\frac{1}{n!}\left[\pm\tfrac{3}{2}i\left(\frac{p}{k_0r}+\frac{q}{k_0^3r^3}-\frac{iq}{k_0^2r^2}\right)e^{ik_0r}\right]^n\left[\beta\left(t-\frac{nr}{c}\right)\right]^n U\left(t-\frac{nr}{c}\right)e^{-\beta(t-nr/c)}, \tag{42}$$

where U is the unit step function. From Eqs. (18) and (19) we obtain the probability amplitudes

$$b_1(t)=\sum_{n=0,2,4\ldots}^{\infty}\frac{1}{n!}\left[\tfrac{3}{2}i\left(\frac{p}{k_0r}+\frac{q}{k_0^3r^3}-\frac{iq}{k_0^2r^2}\right)e^{ik_0r}\right]^n\left[\beta\left(t-\frac{nr}{c}\right)\right]^n U\left(t-\frac{nr}{c}\right)e^{-\beta(t-nr/c)}, \tag{43}$$

$$b_2(t)=\sum_{n=1,3,5\ldots}^{\infty}\frac{1}{n!}\left[\tfrac{3}{2}i\left(\frac{p}{k_0r}+\frac{q}{k_0^3r^3}-\frac{iq}{k_0^2r^2}\right)e^{ik_0r}\right]^n\left[\beta\left(t-\frac{nr}{c}\right)\right]^n U\left(t-\frac{nr}{c}\right)e^{-\beta(t-nr/c)}. \tag{44}$$

Equations (43) and (44) represent our new improved solutions, and exhibit several interesting features not contained in earlier treatments of this problem. Since the results (31) and (32) were obtained by neglecting retardation times, each atom then feels the influence of the other instantaneously, and the system in this approximation *immediately* decays in a cooperative fashion. However, if $k_0r\gg 1$, retardation will play a significant role; the way each atom begins to feel the influence of the other is of interest and importance.

If we neglect the terms which are nonvanishing only after times $t=2r/c$, and retain only the leading terms in $(k_0r)^{-1}$, we have for $\Delta m=\pm 1$ transitions

$$b_1(t)=e^{-\beta t}, \tag{45}$$

$$b_2(t)=(3i/2k_0r)\,e^{ik_0r}\,\beta(t-r/c)\times U(t-r/c)\,e^{-\beta(t-r/c)}. \tag{46}$$

For the probabilities, we have from Eqs. (45) and (46),

$$|b_1(t)|^2=e^{-2\beta t}, \tag{47}$$

$$|b_2(t)|^2=(3/2k_0r)^2U(t-r/c)\times[\beta(t-r/c)]^2e^{-2\beta(t-r/c)}, \tag{48}$$

which are essentially the results of Hamilton,[4] explicitly keeping track of numerical constants.

Equation (43) predicts that at short times the excited atom 1 decays exponentially as if isolated in free space. But at time $t=2r/c$, it begins to be aware of atom 2; clearly this represents the possibility of the excitation being first emitted by atom 1, being absorbed and then re-emitted by atom 2, and finally reabsorbed by atom 1. The other terms have an analogous interpretation. Similarly, Eq. (44) predicts that the "signal" from atom 1 does not arrive at atom 2 until $t=r/c$. This specific feature has, of course, been derived previously,[2] but in that case, the solution was discontinuous. Our solution, in common with that of Hamilton,[4] is continuous, but in contrast to Hamilton's solution, it exhibits all the multiple retardation times associated with successive exchanges of the photon.

To see how the effects of reabsorption (and the subsequent higher-order retardation times) modify these simple lowest-order results, it is interesting to retain terms including the second retardation time $2r/c$, so that the reexcitation of atom 1 can be described. We may again restrict ourselves to $k_0r\gg 1$ and retain only the leading term p/k_0r in Eqs. (43) and (44) for the probability amplitudes

$$b_1(t)=\sum_{n=0,2,4\ldots}^{\infty}\frac{1}{n!}\left(\frac{3ip}{2k_0r}\,e^{ik_0r}\right)^n\times\left[\beta\left(t-\frac{nr}{c}\right)\right]^n U\left(t-\frac{nr}{c}\right)e^{-\beta(t-nr/c)}, \tag{49}$$

$$b_2(t)=\sum_{n=1,3,5\ldots}^{\infty}\frac{1}{n!}\left(\frac{3ip}{2k_0r}\,e^{ik_0r}\right)^n\times\left[\beta\left(t-\frac{nr}{c}\right)\right]^n U\left(t-\frac{nr}{c}\right)e^{-\beta(t-nr/c)}, \tag{50}$$

and for the probabilities

$$|b_1(t)|^2=e^{-2\beta t}-(3p/2k_0r)^2[\beta(t-2r/c)]^2\times U(t-2r/c)\,e^{-2\beta(t-r/c)}\cos(2k_0r)+\cdots, \tag{51}$$

$$|b_2(t)|^2=(3p/2k_0r)^2[\beta(t-r/c)]^2\times U(t-r/c)\,e^{-2\beta(t-r/c)}+\cdots. \tag{52}$$

Consider Eq. (49). The first term on the right-hand side corresponds to the spontaneous emission from atom 1. The second term corresponds to the possibility of a photon being emitted from atom 1, absorbed and emitted by atom 2, and then reabsorbed by atom 1. The second term in Eq. (51) for the probability is an *interference* between these two terms. Hamilton[4] obtains essentially Eq. (52) for $|b_2(t)|^2$; it is identical to Eq. (48) obtained including only a single retardation time (for $\Delta m = \pm 1$ transitions) for the obvious reason that the higher-order terms modify $b_2(t)$ only after time $t = 3r/c$, i.e., when the excitation has had time to get back to atom 2. Hamilton has also noted that the r^{-2} dependence of $|b_2(t)|^2$ contained in Eq. (52) agrees with the classical notion that atom 2 absorbs proportionately to the intensity of the light incident upon it.

On the basis of such classical notions, we might have expected the inverse square dependence of the intensity of the light from atom 2, times the inverse square dependence of the probability of atom 2 to be excited, to lead to an r^{-4} dependence for the probability of reabsorption of the first atom's emitted photon after it has been absorbed and then emitted by atom 2. As Eq. (51) shows, this is not the case. The first two terms in Eq. (49) interfere with each other and lead to an r^{-2} dependence for reabsorption.

The oscillatory factor $\cos(2k_0 r)$ in the second term of $|b_1(t)|^2$ has an obvious classical interpretation. Suppose the interatomic distance is large enough for the first two terms on the right-hand side of Eq. (51) to be a good approximation of $|b_1(t)|^2$. If the atoms are an odd integral number of quarter-wavelengths apart, $|b_1(t)|^2$ is a maximum, whereas if the atoms are separated by an integral number of half-wavelengths, $|b_1(t)|^2$ is a minimum. This is the same type of behavior exhibited by a classical oscillating dipole in the presence of an initially unexcited dipole of the same resonant frequency. The field from the first dipole drives the second dipole, whose field then acts upon the first dipole. If the dipoles are separated by an odd integral number of quarter-wavelengths, the field seen initially by the first dipole from the second dipole is π out of phase with its oscillation and tends to decrease its emitted power. This is analogous in the two-atom problem to a *decrease* in the probability of photon emission from atom 1, i.e., a *maximum* $|b_1(t)|^2$.

If we neglect all retardation times, we should recover the standard results of Sec. II. For times much longer than all but the largest retardation times

$$C_\pm(t) \approx \sum_{n=0}^{\infty} \frac{1}{n!}\left[\pm\tfrac{3}{2} i\left(\frac{p}{k_0 r} + \frac{q}{k_0^3 r^3} - \frac{iq}{k_0^2 r^2}\right) e^{ik_0 r}\right]^n (\beta t)^n e^{-\beta t}$$
$$= e^{-\beta[1 \pm g(r)]t} . \qquad (53)$$

Thus, we have recovered the results of Stephen[9] discussed in Sec. II. The result of Stephen is seen in this light as the consequence of infinitely many photon exchanges between the atoms concerned, and is valid provided that we may neglect retardation times.

IV. RESTRICTION TO PHOTONS PROPAGATING ALONG INTERATOMIC AXIS

In Sec. III, the photon could be emitted into any of the free-space modes. However, for the purpose of discussing super-radiance, it is sometimes convenient to restrict the number of available modes. We may anticipate that for a long pencil-shaped system of atoms, super-radiant emission will be overwhelmingly into the axial modes of the system; indeed the approximation of allowing super-radiant emission only into this "end-fire" mode forms the basis of a thorough discussion[24] of super-radiance. Arecchi and Courtens[13] have discussed the effects of retardation for such a system, and in particular for a two-atom system. In this section, we show how multiple retardation times play a role in such a system, and how the result of Arecchi and Courtens may be regarded as a truncated version of the full solution in a way analogous to Hamilton's solution for the free-space problem.

Up until now, we have assumed an initial condition of atom 1 excited and atom 2 in its ground state. If we allow the states $|+\rangle_1 |-\rangle_2 |\{0\}\rangle = |\phi_1\rangle$ and $|-\rangle_1 |+\rangle_2 |\{0\}\rangle = |\phi_2\rangle$ to have *arbitrary* (consistent with orthonormality) amplitudes $b_1(0)$ and $b_2(0)$ at $t = 0$, then the amplitudes at the time t are

$$b_1(t) = \tfrac{1}{2}[b_1(0) + b_2(0)]\, C_+(t) + \tfrac{1}{2}[b_1(0) - b_2(0)]\, C_-(t), \qquad (54)$$

$$b_2(t) = \tfrac{1}{2}[b_1(0) + b_2(0)]\, C_+(t) - \tfrac{1}{2}[b_1(0) - b_2(0)]\, C_-(t), \qquad (55)$$

where $C_\pm(t)$ are given by Eq. (20). Arecchi and Courtens consider the initial state

$$|\theta_1\rangle = (1/\sqrt{2})(e^{i\vec{k}_0\cdot\vec{r}/2}|\phi_1\rangle + e^{-i\vec{k}_0\cdot\vec{r}/2}|\phi_2\rangle), \qquad (56a)$$

and require the amplitude $a_1(t)$ and $a_2(t)$ for the system to be in the states $|\theta_1\rangle$ and

$$|\theta_2\rangle = (1/\sqrt{2})(e^{i\vec{k}_0\cdot\vec{r}/2}|\phi_1\rangle - e^{-i\vec{k}_0\cdot\vec{r}/2}|\phi_2\rangle), \qquad (56b)$$

respectively, at time t. Here $k_0 = |\vec{k}_0| = \omega_0/c$. As the interatomic distance $r \to 0$, or if $k_0 = 0$, $|\theta_1\rangle$ and $|\theta_2\rangle$ correspond to the symmetric and antisymmetric states discussed previously. The state $|\theta_1\rangle$ corresponds to an initial plane-wave excitation by an incident field of wave vector $\vec{k}_0$ parallel to $\vec{r}$.

The amplitudes $a_1(t)$ and $a_2(t)$ can be related to $b_1(t)$ and $b_2(t)$ by writing

$$b_1(t)|\phi_1\rangle + b_2(t)|\phi_2\rangle = a_1(t)|\theta_1\rangle + a_2(t)|\theta_2\rangle,$$

which gives

$$a_1(t) = b_2(0)b_1(t) + b_1(0)b_2(t) \tag{57}$$

and

$$a_2(t) = b_2(0)b_1(t) - b_1(0)b_2(t) \tag{58}$$

for

$$\begin{aligned} b_1(0) &= (1/\sqrt{2}\,)(e^{i\vec{k}_0\cdot\vec{r}/2}), \\ b_2(0) &= (1/\sqrt{2}\,)(e^{-i\vec{k}_0\cdot\vec{r}/2}). \end{aligned} \tag{59}$$

As in Sec. III, we find it convenient to work with the functions $c_\pm(t)$. Using Eqs. (54), (55), and (57)–(59), we have

$$a_1(t) = \tfrac{1}{2}(1 + \cos k_0 r)\,c_+(t) + \tfrac{1}{2}(1 - \cos k_0 r)c_-(t), \tag{60}$$

$$a_2(t) = -\tfrac{1}{2}i\,\sin k_0 r[c_+(t) - c_-(t)]. \tag{61}$$

For $k_0 r \to 0$

$$a_1(t) = c_+(t), \quad a_2(t) = 0.$$

We choose the situation appropriate to an idealized description of super-radiance in a pencil-shaped cavity by considering only radiation modes with wave vectors along the interatomic axis; this corresponds to the directional emission along the cavity axis. Following Arecchi and Courtens,[13] we consider only a single polarization, and replace Eq. (5) with

$$\begin{aligned} g_l(k) &= (\mu/\hbar)(2\pi\hbar\omega_k/V)^{1/2}\,e^{ik\hat{r}\cdot\vec{x}_l} \\ &= C(k)\,e^{ik\hat{r}\cdot\vec{x}_l}, \end{aligned} \tag{62}$$

where again

$$\vec{x}_2 - \vec{x}_1 = r\hat{r}.$$

$c_\pm(t)$ is given by Eq. (21), where instead of the free-space all-mode results (15) and (16), we now have

$$\begin{aligned} A(s) = \sum_k C^2(k)\{&[s + i(\omega_k - \omega_0)]^{-1} \\ &+ [s + i(\omega_k + \omega_0)]^{-1}\}, \end{aligned} \tag{63}$$

$$\begin{aligned} B(s) = \sum_k C^2(k)\cos kr\{&[s + i(\omega_k - \omega_0)]^{-1} \\ &+ [s + i(\omega_k + \omega_0)]^{-1}\}. \end{aligned} \tag{64}$$

For reflecting boundary conditions $k = n\pi/L$, following Arecchi and Courtens,[13]

$$\sum_k \to \frac{L}{\pi}\int dk,$$

so that

$$\begin{aligned} A(iz + \epsilon) = \frac{-iL}{\pi c}\int_0^\infty d\omega\, C^2\!\left(\frac{\omega}{c}\right) \\ \times\left(\frac{1}{\omega - \omega_0 + z - i\epsilon} + \frac{1}{\omega + \omega_0 + z - i\epsilon}\right), \end{aligned} \tag{65}$$

$$\begin{aligned} B(iz + \epsilon) = \frac{-iL}{\pi c}\int_0^\infty d\omega\, C^2\!\left(\frac{\omega}{c}\right)\cos\!\left(\frac{\omega r}{c}\right) \\ \times\left(\frac{1}{\omega - \omega_0 + z - i\epsilon} + \frac{1}{\omega + \omega_0 + z - i\epsilon}\right). \end{aligned} \tag{66}$$

If we again make the pole approximation on $A(iz + \epsilon)$, and neglect single-atom frequency shifts, we have

$$A(iz + \epsilon)|_{z=0} = (L/c)C^2(k_0) = \tfrac{1}{2}\gamma, \tag{67}$$

where γ is the single-atom decay rate replacing the Einstein A coefficient.

Using the same approximations as we used in Sec. III to evaluate $B(iz + \epsilon)$, we obtain

$$B(iz + \epsilon) \cong (\tfrac{1}{2}\gamma)\exp[i(\omega_0 - z)r/c], \tag{68}$$

and

$$\begin{aligned} c_\pm(t) = \sum_{n=0}^{\infty}\frac{(\mp 1)^n}{n!}\,e^{ink_0 r}[\tfrac{1}{2}\gamma(t - nr/c)]^n \\ \times U(t - nr/c)\exp[-\tfrac{1}{2}\gamma(t - nr/c)]. \end{aligned} \tag{69}$$

If we neglect all terms which are nonvanishing only for $t > 2r/c$, we have from Eqs. (60), (61), and (69)

$$\begin{aligned} a_1(t) = e^{-\gamma t/2} - \cos k_0 r\, e^{ik_0 r}[(\tfrac{1}{2}\gamma)(t - r/c)] \\ \times U(t - r/c)\exp[-\tfrac{1}{2}\gamma(t - r/c)], \end{aligned} \tag{70}$$

and

$$\begin{aligned} a_2(t) = i\,\sin k_0 r\, e^{ik_0 r}[(\tfrac{1}{2}\gamma)(t - r/c)] \\ \times U(t - r/c)\exp[-\tfrac{1}{2}\gamma(t - r/c)]. \end{aligned} \tag{71}$$

Equations (70) and (71) should be compared with the corresponding results of Arecchi and Courtens:

$$\begin{aligned} a_1(t) = e^{-\gamma t/2} - (\tfrac{1}{2}\gamma)(t - r/c) \\ \times U(t - r/c)\exp[-\tfrac{1}{2}\gamma(t - r/c)], \end{aligned} \tag{72}$$

$$a_2(t) = -(\tfrac{1}{2}\gamma)(t - r/c)\,U(t - r/c)\exp[-\tfrac{1}{2}\gamma(t - r/c)]. \tag{73}$$

For $k_0 r \to 0$, $a_1(t)$ and $a_2(t)$ are, respectively, the amplitudes for the symmetric and antisymmetric states $|\phi_+\rangle$ and $|\phi_-\rangle$. Equation (61) [or (71)] indicates that, in this limit, the antisymmetric state cannot be reached. This is certainly as it should be, since in the limit $k_0 r \to 0$, the Hamiltonian becomes invariant under particle permutation, and can only connect states of the same symmetry. $|\phi_-\rangle$ has symmetry opposite to that of $|\phi_+\rangle$ under particle permutation, and so its amplitude must remain zero for all times. In Dicke's terminology,[8] the cooperation number, initially equal to 1, must be a constant of the motion for $k_0r\to 0$; the state $|\phi_-\rangle$ has cooperation number 0, and therefore cannot be reached.

The Arecchi-Courtens[13] result (73) indicates that the antisymmetric state has some nonvanishing amplitude for $t>0$ in the limit $r\to 0$; this disagreeable feature is not found in Eq. (71).

When all the retardation times are neglected, the complete set of photon "exchanges" may be summed to yield

$$\begin{aligned} a_1(t)={}&\tfrac{1}{2}(1+\cos k_0r)\exp[-\tfrac{1}{2}\gamma(1+\cos k_0r)t] \\ &\times\exp[-\tfrac{1}{2}i\gamma t\sin k_0r]+\tfrac{1}{2}(1-\cos k_0r) \\ &\times\exp[-\tfrac{1}{2}\gamma(1-\cos k_0r)t] \\ &\times\exp[\tfrac{1}{2}i\gamma t\sin k_0r]\,, \end{aligned} \tag{74}$$

and

$$\begin{aligned} a_2(t)=-\tfrac{1}{2}i\sin k_0r\{&\exp[-\tfrac{1}{2}\gamma(1+\cos k_0r)t] \\ &\times\exp(-\tfrac{1}{2}i\gamma t\sin k_0r) \\ &-\exp[-\tfrac{1}{2}\gamma(1-\cos k_0r)t] \\ &\times\exp(\tfrac{1}{2}i\gamma t\sin k_0r)\}. \end{aligned} \tag{75}$$

The fully retarded results indicate that there is no cooperative decay until time r/c. On the basis of this result, Arecchi and Courtens[13] have introduced a "cooperation length" $c\tau_c$, where τ_c is the super-radiant lifetime. Atoms separated by more than a cooperation length will decay approximately independently. This argument is, of course, only meaningful for the case of highly directional emission; if all propagation directions are allowed, the Stephen interaction cuts off at distances much shorter than those for which the causal retardation times play an important role. From Eq. (30), it is seen that the cutoff length in this case is approximately a transition wavelength. In a similar context, this result was discussed by Lord Rayleigh.[25]

It should be emphasized, of course, that the restriction to photons propagating along the interatomic axis is unrealistic. We have discussed this idealized model only to draw comparison with the work of Arecchi and Courtens.[13] A more realistic treatment of cooperative decay, including emission into all available free-space modes, should *predict* the directional characteristics of the emission. Such an analysis could proceed in a manner similar to that presented in Sec. III.

It should also be borne in mind that $a_1(t)$ and $a_2(t)$ refer to the correlated states $|\theta_1\rangle$ and $|\theta_2\rangle$ and not to the product states associated with $b_1(t)$ and $b_2(t)$. The product states $|\phi_1\rangle$ and $|\phi_2\rangle$ are useful for discussing the retardation aspect of the problem where the emphasis is on excitation exchange. Any realistic excitation mechanism will excite some superposition of correlated states which is not necessarily a product state. For example, for atoms located within a wavelength, a plane-wave electric field can only excite a completely symmetric state.

V. DISCUSSION

We have presented a solution to the retardation problem which (i) is continuous; (ii) reduces to previously reported continuous solutions when higher-order retardation effects are omitted; and (iii) reduces to the well-known results for the resonant interaction for times much greater than any of the retardation times (or neglecting all retardation times). In addition, we show that both the causal and cooperative aspects of the two-atom problem are manifested within a single unified formalism. The full solution exhibits the effects of multiple photon exchanges between the two atoms; since very many exchanges occur during a spontaneous emission lifetime for small interatomic spacing ($r\sim\lambda$), cooperation appears quite naturally as a smoothed approximation to the full solution. The calculation of Sec. III can be extended to the case $k_0r<1$.

The results we have obtained enable us to comment upon the usefulness of the various basis states employed. If we take the solutions neglecting retardation times from Sec. III, in the limit $k_0r\ll 1$, we have

$$g(r)\simeq 1-\tfrac{3}{2}iq(k_0r)^{-3}, \tag{76}$$

and

$$\begin{aligned} |b_1(t)|^2&=\tfrac{1}{4}(1+e^{-4\beta t}+2e^{-2\beta t}\cos 2\Delta t) \\ &\simeq\cos^2\Delta t\quad\text{for }\beta t\ll 1, \end{aligned} \tag{77}$$

$$\begin{aligned} |b_2(t)|^2&=\tfrac{1}{4}(1+e^{-4\beta t}-2e^{-2\beta t}\cos 2\Delta t) \\ &\simeq\sin^2\Delta t\quad\text{for }\beta t\ll 1, \end{aligned} \tag{78}$$

where

$$\hbar\Delta=\mu^2q/r^3. \tag{79}$$

This indicates that for $\beta t\ll 1$, the excitation oscillates back and forth between atoms 1 and 2 with frequency Δ (the dipole-dipole coupling).[26] This should not be confused with Förster's resonance energy transfer rate.[27]

For large Δ (small separation), it is clear that the initial condition we originally specified (a simple product state) is not very meaningful. After a very short time the excitation is equally likely to be on atom 1 or on atom 2, with the state vector more precisely defined by $|\phi_\pm\rangle$, the symmetric and antisymmetric states. These states provide a more natural basis for close separations; they are exactly analogous to the two normal modes of the classical system of two coupled harmonic oscillators. In fact, when radiation damping is neglected, the states $|\phi_\pm\rangle$ are stationary states of the system. For this reason, they are sometimes referred to as "quasi-stationary" states. The imaginary part of $g(r)$ directly describes the energy shifts of these states and not the product states $|\phi_1\rangle$ and $|\phi_2\rangle$.

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APPENDIX A: COMMENTS ON THE FORM OF THE INTERACTION HAMILTONIAN

Throughout this paper, we have used the electric dipole interaction Hamiltonian $H_{\text{INT}} = -\vec{\mu}\cdot\vec{\text{E}}(\vec{\text{r}})$, and taken into account energy nonconserving virtual processes by removing the rotating-wave approximation (RWA). The RWA has been used quite often in discussions of cooperative decay: in the two-atom problem by Hamilton,[4] Hutchinson and Hameka,[10] Arecchi and Courtens,[13] and others.

In the RWA, the expressions (13) and (14) for $\hat{b}_1(s)$ and $\hat{b}_2(s)$ remain formally the same, but $A(s)$ and $B(s)$ from Eqs. (15) and (16) are replaced by

$$A(s) = \sum_{\vec{\text{k}}\lambda} \frac{|g_1(\vec{\text{k}}_\lambda)|^2}{s + i(\omega_k - \omega_0)}, \tag{A1}$$

$$B(s) = \sum_{\vec{\text{k}}\lambda} \frac{g_1^*(\vec{\text{k}}_\lambda) g_2(\vec{\text{k}}_\lambda)}{s + i(\omega_k - \omega_0)}. \tag{A2}$$

Then Eqs. (22) and (23) become

$$A = -\frac{2i\mu^2}{3\pi\hbar c^3} \int_0^\infty \frac{\omega^3 d\omega}{\omega - \omega_0 - i\epsilon}, \tag{A3}$$

and

$$B = -\frac{i\mu^2}{\pi\hbar c^3} \int_0^\infty \frac{\omega^3 f(\omega r/c)\, d\omega}{\omega - \omega_0 - i\epsilon} \tag{A4}$$

in the pole approximation.

Using Eq. (26),

$$A = \beta - \frac{2i\mu^2}{3\pi\hbar c^3} P \int_0^\infty \frac{\omega^3 d\omega}{\omega - \omega_0}. \tag{A5}$$

The divergent (unrenormalized) energy shift term represents the level shift of the upper state of our excited two-level atom. Without the RWA we have, using the "essential states,"[10]

$$A = \beta - \frac{2i\mu^2}{3\pi\hbar c^3} \left(P \int_0^\infty \frac{\omega^3 d\omega}{\omega - \omega_0} + \int_0^\infty \frac{\omega^3 d\omega}{\omega + \omega_0} \right). \tag{A6}$$

The additional term represents (as explained in the text) the level shift of the lower state of the ground-state two-level atom. The total energy shift is the energy shift of the initial state $|\phi_1\rangle = |+\rangle_1 |-\rangle_2 |\{0\}\rangle$. The RWA therefore gives the correct decay rate but not the proper level shift. Ground states are not shifted in the RWA. This is not a severe limitation, since the two-level atom model is inadequate to deal with real radiative level shifts.

We do encounter a serious limitation of the RWA when we consider the r-dependent term B in Eq. (A4)

$$B = \frac{\mu^2 \omega_0^3}{\hbar c^3} f(\omega_0 r/c) - \frac{i\mu^2}{\pi\hbar c^3} P \int_0^\infty \frac{\omega^3 f(\omega r/c)\, d\omega}{\omega - \omega_0}. \tag{A7}$$

If we do not make the RWA, the level shift is

$$-\frac{i\mu^2}{\pi\hbar c^3} P \int_{-\infty}^\infty \frac{\omega^3 f(\omega r/c)\, d\omega}{\omega - \omega_0},$$

which gives our previous results. Therefore, treatments based upon the RWA cannot give the correct (retarded first-order dispersion) dipole-dipole level shift. However, it is clearly permissible to use the RWA to calculate A and B if we adopt the *Ansatz* of extending the frequency integrals to minus infinity.

Another problem encountered in spontaneous emission processes is the choice of the interaction Hamiltonian.[15] We have chosen to use the dipole form $-\vec{\mu}\cdot\vec{\text{E}}(\vec{\text{r}})$, although this presents difficulties in interpreting the level shift in a renormalized fashion. This form is well suited for describing the retarded dipole-dipole interaction. Other authors have chosen to use the minimal substitution Hamiltonian $-(e/mc)\vec{\text{p}}\cdot\vec{\text{A}}(\vec{\text{r}})$ ($\vec{\text{A}}$ is the quantized vector potential), and to neglect the $(e^2/2mc^2)\vec{\text{A}}^2$ quadratic term. Without the RWA, but using the pole approximation, we may repeat the analysis of Sec. III using the $\vec{\text{p}}\cdot\vec{\text{A}}$ form to obtain

$$B'(iz + \epsilon)|_{z=0} = -\tfrac{3}{2}\beta f_1(\omega_0 r/c) + (3iq/2k_0^3 r^3)\beta, \tag{A8}$$

where $f_1(\omega_0 r/c)$ is given by Eq. (39). The first term is identical to that obtained using the form $-\vec{\mu}\cdot\vec{E}(\vec{r})$, but we now have another term, arising from a pole at $\omega=0$, which is unretarded. This must now be cancelled by including in the Hamiltonian the static dipole-dipole interaction,[9] although some authors not interested in the form of the dipole-dipole level shift have not done this. For our two-level atom, the static dipole-dipole interaction is given for our purposes by

$$V=-\frac{3q\beta\hbar}{2k_0^3r^3}(\sigma_1^+\sigma_2^-+\sigma_2^+\sigma_1^-). \tag{A9}$$

Including this term, we obtain a term exactly cancelling the second term of Eq. (A8),

$$B'(iz+\epsilon)|_{z=0}=-\tfrac{3}{2}\beta f_1(\omega_0 r/c),$$

in exact agreement with our earlier result, obtained using the $-\vec{\mu}\cdot\vec{E}$ form.

APPENDIX B: DERIVATION OF EQS. (23)-(25)

Our assumption of parallel transition moments $\vec{\mu}_1$ and $\vec{\mu}_2$ is actually required by conservation of angular momentum. For example, if atom 1 makes a $\Delta m=-1$ transition (axis of quantization equals the interatomic axis), atom 2, after absorbing atom 1's emitted photon, will also emit via a $\Delta m=-1$ transition.

The function $f(kr)$ may be written

$$\begin{aligned} f(kr)&=\frac{1}{4\pi}\oint d\Omega_{\hat{k}}\sum_{\lambda=1,2}|\hat{\mu}\cdot\hat{\epsilon}(\vec{k}_\lambda)|^2e^{i\vec{k}\cdot\vec{r}}\\ &=\frac{1}{4\pi}\oint d\Omega_{\hat{k}}(1-|\hat{k}\cdot\hat{\mu}|^2)e^{i\vec{k}\cdot\vec{r}}, \end{aligned} \tag{B1}$$

taking the unit vectors $\hat{\mu}_1$ and $\hat{\mu}_2$ to be equal, and making the relative dipole approximation.

Equation (B1) may be written

$$\begin{aligned} f(kr)=\frac{1}{(4\pi)^{1/2}}\sum_{l=0}^{\infty}i^l(2l+1)^{1/2}j_l(kr)\int_0^{2\pi}d\phi\\ \times\int_0^{\pi}d\theta\sin\theta(1-|\hat{k}\cdot\hat{\mu}|^2)\,Y_{l0}(\theta,0), \end{aligned} \tag{B2}$$

where the z axis joins the two atoms.

For $\Delta m=0$ transitions, $\hat{\mu}=\hat{z}$ and $1-|\hat{k}\cdot\hat{\mu}|^2=\sin^2\theta$. Then

$$\begin{aligned} f(kr)&=\frac{2\pi}{(4\pi)^{1/2}}\sum_{l=0}^{\infty}i^l(2l+1)^{1/2}j_l(kr)\int_0^{\pi}d\theta\sin^3\theta\,Y_{l0}(\theta,0)=\frac{1}{2}\sum_{l=0}^{\infty}i^l(2l+1)j_l(kr)\int_{-1}^{1}dx(1-x^2)P_l^{(0)}(x)\\ &=\frac{2}{3}\sum_{l=0}^{\infty}i^lj_l(kr)(\delta_{0l}-\delta_{2l})=2\left(\frac{\sin kr}{(kr)^3}-\frac{\cos kr}{(kr)^2}\right). \end{aligned} \tag{B3}$$

For $\Delta m=\pm1$ transitions, we have $\hat{\mu}=\mp2^{-1/2}(\hat{x}\pm i\hat{y})$ and $(1-|\hat{k}\cdot\hat{\mu}|^2)=\frac{1}{2}(1+\cos^2\theta)$. In this case, it is easily shown that

$$f(kr)=\frac{\sin kr}{kr}-\frac{\sin kr}{(kr)^3}+\frac{\cos kr}{(kr)^2}. \tag{B4}$$

Thus, for $k_0r\gtrsim1$, cooperative effects are generally more important for $\Delta m=\pm1$ transitions where a transverse photon may be emitted along the interatomic axis, than for $\Delta m=0$ transitions. This discrimination of $\Delta m=\pm1$ from $\Delta m=0$ transitions in the form of $f(kr)$ has also been used by Hutchinson and Hameka.[10]

APPENDIX C: DETAILS OF SEMIPOLE APPROXIMATION

For $|z|\ll\omega_0$ we have already remarked that we may drop the third term on the right-hand side of Eq. (36). Then

$$\begin{aligned} B(iz+\epsilon)&\to\frac{\mu^2}{\hbar c^3}\int_0^{\infty}d\omega\,\omega^3f\left(\frac{\omega r}{c}\right)\delta(\omega-\omega_0+z)-\frac{i\mu^2}{\pi\hbar c^3}\left(P\int_0^{\infty}\frac{\omega^3f(\omega r/c)}{\omega-\omega_0+z}d\omega+P\int_{-\infty}^{0}\frac{\omega^3f(\omega r/c)}{\omega-\omega_0-z}d\omega\right)\\ &=\frac{\mu^2}{\hbar c^3}\int_0^{\infty}d\omega\,\omega^3f\left(\frac{\omega r}{c}\right)\delta(\omega-\omega_0+z)-\frac{i\mu^2}{\pi\hbar c^3}\left(P\int_0^{\infty}\frac{\omega^3f(\omega r/c)}{\omega-\omega_0+z}d\omega+\int_0^{\infty}\frac{\omega^3f(\omega r/c)}{\omega+\omega_0+z}d\omega\right), \end{aligned} \tag{C1}$$

where we need not be concerned with a principal-part integration in the last integral for $|z|\ll\omega_0$.

We require the integrals

$$P\int_0^{\infty}\frac{\omega^2\sin(\omega r/c)d\omega}{\omega\mp\omega_0+z},\qquad P\int_0^{\infty}\frac{\omega\cos(\omega r/c)d\omega}{\omega\mp\omega_0+z},\qquad P\int_0^{\infty}\frac{\sin(\omega r/c)d\omega}{\omega\mp\omega_0+z}. \tag{C2}$$

Since the first two integrals are obtained from the third by differentiation with respect to r, we need only consider the third integral

$$X_{\mp}(z)=P\int_0^{\infty}\frac{\sin(\omega r/c)\,d\omega}{\omega\mp\omega_0+z}\,, \tag{C3}$$

which can be evaluated to give

$$\begin{aligned}X_-(z)=&-Ci(k_0r-zr/c)\sin(k_0r-zr/c)\\&+si(k_0r-zr/c)\cos(k_0r-zr/c)\\&+\pi\cos(k_0r-zr/c),\end{aligned} \tag{C4}$$

and

$$\begin{aligned}X_+(z)=&\,Ci(k_0r+zr/c)\sin(k_0r+zr/c)\\&-si(k_0r+zr/c)\cos(k_0r+zr/c).\end{aligned} \tag{C5}$$

From Eq. (C1), it is seen that $X_{\mp}(z)$ are required in the combination

$$\begin{aligned}X_-(z)+X_+(z)=&\,Ci(k_0r+zr/c)\sin(k_0r+zr/c)\\&-Ci(k_0r-zr/c)\sin(k_0r-zr/c)\\&+si(k_0r-zr/c)\cos(k_0r-zr/c)\\&-si(k_0r+zr/c)\cos(k_0r+zr/c)\\&+\pi\cos(k_0r-zr/c)\\\approx&\,\pi\cos(k_0r-zr/c)\end{aligned} \tag{C6}$$

for $|z|\ll\omega_0$. In fact, in the limit $k_0r\gg1$ we consider in Sec. III, the asymptotic series

$$\begin{aligned}Ci(x)=&\sin x\left(\frac{1}{x}-\frac{2!}{x^3}+\frac{4!}{x^5}-\cdots\right)\\&-\cos x\left(\frac{1}{x^2}-\frac{3!}{x^4}+\frac{5!}{x^6}-\cdots\right)\,,\end{aligned} \tag{C7}$$

$$si(x)=-\cos x\left(\frac{1}{x}-\frac{2!}{x^3}+\cdots\right)-\sin x\left(\frac{1}{x^2}-\frac{3!}{x^4}+\cdots\right)\,, \tag{C8}$$

indicates the validity of replacing $X_-(z)+X_+(z)$ by $\pi\cos(k_0r-zr/c)$, the semipole approximation,

$$X_-(z)+X_+(z)\rightarrow\pi\cos(k_0r-zr/c) \tag{C9}$$

or

$$\begin{aligned}&P\int_0^{\infty}\frac{\sin(\omega r/c)\,d\omega}{\omega-\omega_0+z}+\int_0^{\infty}\frac{\sin(\omega r/c)\,d\omega}{\omega+\omega_0+z}\\&\qquad\rightarrow P\int_{-\infty}^{\infty}\frac{\sin(\omega r/c)\,d\omega}{\omega-\omega_0+z}\,.\end{aligned} \tag{C10}$$

The replacement indicated by Eq. (C10) is, of course, exact in the full pole approximation ($z=0$).

The pole approximation can perhaps be best appreciated by considering the case of single-atom spontaneous emission. Suppose first that there is only a single field mode interacting with the atom. The amplitude for remaining in the initially excited state with no photons in the field is, in the RWA, the inverse Laplace transform of

$$\hat{b}(s)=[s+A(s)]^{-1}, \tag{C11}$$

where

$$A(s)=g^2/(s+i\delta), \tag{C12}$$

and g is the coupling constant between the atom and the single field mode, and $\delta=\omega-\omega_0$, where ω is the circular frequency of the field mode. We obtain from Eqs. (C11) and (C12) the probability[28]

$$|b(t)|^2=\cos^2\tfrac{1}{2}\Omega t+(\delta^2/\Omega^2)\sin^2\tfrac{1}{2}\Omega t\,, \tag{C13}$$

where $\Omega=(\delta^2+4g^2)^{1/2}$. The pole approximation is meaningless in this case. It would imply $|b(t)|^2=1$.

Consider now the case of spontaneous emission into the free-space vacuum. Now all field modes are allowed, and in

$$A(s)=\sum_{\vec{k}\lambda}\frac{|g(\vec{k}_\lambda)|^2}{s+i(\omega_k-\omega_0)} \tag{C14}$$

we effectively encounter an infinite number of poles (in the free-space limit where the field frequencies form a continuous variable, these poles become a branch cut). The essence of the pole approximation is the assumption that, in the free-space limit, rather than having the excitation energy oscillate back and forth between the atom and field [as indicated by Eq. (C13) for a single mode], the number of field modes is so large that this excitation energy never returns to the atom. Our "semipole" approximation does not alter this irreversibility; it merely represents an improvement over the full pole approximation, to show how retardation is accounted for in this case.

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