Analytic solution for retardation in two-atom systems

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The spontaneous emission of a pair of two identical and fixed two-level atoms, sharing initially a single excitation, is considered. It is shown that the solution which involves all the retardation times obtained by Milonni and Knight [Phys. Rev. A **10**, 1096 (1974)] is also valid for interatomic distances less than half of a transition wavelength. The effects of the interatomic distance on the time evolution of the atomic state populations are examined. [S1050-2947(99)07203-0]

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The energy transfer between a pair of atoms and the role of causality have been subjects of long-standing interest because of their fundamental importance [1–9]. The problem is usually formulated in terms of a pair of two-level atoms a fixed distance r apart, atom 1 in the upper state, atom 2 in the lower state, and no photon in the radiation field. From the causality principle, one would expect that at times less than $t=2r/c$, atom 1 decays as if isolated in free space. Meanwhile, atom 2 may have nonvanishing upper state population only after $t=r/c$. At time $t=2r/c$, atom 1 starts to be aware of atom 2, and so on. Formulas for the various probability amplitudes which show how all the retardation times nr/c enter have been obtained by Milonni and Knight [4,5] (see also [6]) under the condition that the atoms are larger than half of a transition wavelength apart. In this Brief Report, we will show that this condition is, in fact, unnecessary. In doing so, we extend the results of Milonni and Knight on higher-order retardation effects [4,5] and of Goldstein and Meystre on the dipole-dipole interaction [10] to interatomic distances less than half a wavelength. We analyze the effects of the interatomic distance r on the time behavior of the atomic state populations and demonstrate explicitly the transition from our model to the Dicke model of superradiance [11], which occurs when the two atoms get close enough to each other.

The process under discussion is governed by the Hamiltonian, in the electric dipole and rotating wave approximations,

$$H=\sum_{\mathbf{k}} \hbar\omega_k a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}+\sum_{m=1,2}\frac{1}{2}\hbar\omega_0\sigma_{mz} +\hbar\sum_{\mathbf{k}}\sum_{m=1,2}[g_m(\mathbf{k})\sigma_m a_{\mathbf{k}}^{\dagger}+g_m^*(\mathbf{k})\sigma_m^{\dagger}a_{\mathbf{k}}], \quad (1)$$

where $a_{\mathbf{k}}^{\dagger}$ and $a_{\mathbf{k}}$ are the photon creation and annihilation operators and σ_m, $\sigma_m^{\dagger}$, and σ_{mz} are Pauli operators for atom m. The coupling constants $g_m(\mathbf{k})$ read

$$g_m(\mathbf{k})=i\left(\frac{2\pi\omega_k}{\hbar V}\right)^{1/2}(\mu_m\cdot\hat{\boldsymbol{\epsilon}}_{\mathbf{k}})e^{i\mathbf{k}\cdot\mathbf{r}_m}, \quad (2)$$

with μ_m being the transition dipole moment for atom m located at $\mathbf{r}_m$ and $\hat{\boldsymbol{\epsilon}}_{\mathbf{k}}$ being a polarization unit vector. To facilitate the comparison, we follow closely the notation used in [4,5].

The system wave function at time t can be written as

$$|\psi(t)\rangle=b_1(t)|u,l\rangle|\{0\}\rangle+b_2(t)|l,u\rangle|\{0\}\rangle +\sum_{\mathbf{k}} b_{l\mathbf{k}}(t)|l,l\rangle|\{1_{\mathbf{k}}\}\rangle. \quad (3)$$

Using the Schrödinger equation and the initial condition $b_{l\mathbf{k}}(0)=0$, one can derive the following equations for the probability amplitudes of finding atom 1 (2) in the upper state:

$$\dot{b}_m(t)=\sum_{n=1,2}\int_0^t K_{mn}(t-t')b_n(t')dt', \quad m=1,2, \quad (4)$$

where

$$K_{mn}(t-t')=-\sum_{\mathbf{k}} g_m^*(\mathbf{k})g_n(\mathbf{k})e^{-i(\omega_k-\omega_0)(t-t')}. \quad (5)$$

The kernels $K_{mn}(t-t')$ can be evaluated (see the Appendix) and put in Eqs. (4) to yield

$$\dot{b}_m(t)=-\beta b_m(t)+f b_n\left(t-\frac{r}{c}\right)H\left(t-\frac{r}{c}\right), \quad m,n=1,2 \ (n\neq m), \quad (6)$$

where $\beta=2\mu^2\omega_0^3/(3\hbar c^3)$ is one-half the free-space decay rate, $H(x)$ is the Heaviside unit step function, and the coefficient f is defined as

$$f=-\frac{3}{2}\beta\left[\frac{p}{ik_0r}+\frac{q}{i(k_0r)^3}-\frac{q}{(k_0r)^2}\right]e^{ik_0r}, \quad p=0, \quad q=2 \quad \text{for} \quad \Delta m=0 \ \text{transitions}, \quad p=1, \quad q=-1 \quad \text{for} \quad \Delta m=\pm 1 \ \text{transitions}, \quad (7)$$

with $k_0=\omega_0/c$. It is convenient to introduce

$$C_{\pm}(t)=b_1(t)\pm b_2(t), \quad (8)$$

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which obey uncoupled delay differential equations

$$\dot{C}_{\pm}(t) = -\beta C_{\pm}(t) \pm f C_{\pm}\left(t - \frac{r}{c}\right) H\left(t - \frac{r}{c}\right). \tag{9}$$

The Laplace transform of these leads to

$$C_{\pm}(s) = \frac{1}{s + \beta \mp f \exp(-sr/c)}, \tag{10}$$

which is nothing but Eq. (17) given in [5]: $C_{\pm}(s) = 1/[s + A(s) \pm B(s)]$ with $A(s)$ and $B(s)$ given by Eqs. (27) and (40), respectively. To find the original functions $C_{\pm}(t)$ from Eq. (10), Milonni and Knight have assumed $k_0 r \gtrsim 3$, so that the integrand in the inverse Laplace transform can be expanded into a power series. The integral can then be performed using the contour integral techniques to give [5]

$$C_{\pm}(t) = \sum_{n=0}^{\infty} \frac{1}{n!} (\pm f)^n t_n^n e^{-\beta t_n} H(t_n), \tag{11}$$

where $t_n = t - nr/c$.

Here we show that Eqs. (9) can be solved exactly, without the additional assumption $k_0 r \gtrsim 3$ on the interatomic distance, as follows. First, for the sake of simplicity, we define

$$\tilde{C}_{\pm}(t) = e^{\beta t} C_{\pm}(t), \quad \tilde{f} = e^{\beta r/c} f, \tag{12}$$

so that Eqs. (9) become

$$\dot{\tilde{C}}_{\pm}(t) = \pm \tilde{f} \tilde{C}_{\pm}\left(t - \frac{r}{c}\right) H\left(t - \frac{r}{c}\right). \tag{13}$$

For times $t \leqslant r/c$, Eqs. (13) become ordinary differential equations, which can be integrated easily. Once the solution for $t \leqslant r/c$ is known, we can compute the solution for $r/c \leqslant t \leqslant 2r/c$, and so on. The results of several first steps are, with the initial conditions $\tilde{C}_{\pm}(0) = 1$,

$$\tilde{C}_{\pm}(t) = 1 \quad \text{for} \quad 0 \leqslant t \leqslant \frac{r}{c},$$

$$\tilde{C}_{\pm}(t) = 1 \pm \tilde{f} t_1 \quad \text{for} \quad \frac{r}{c} \leqslant t \leqslant \frac{2r}{c},$$

$$\tilde{C}_{\pm}(t) = 1 \pm \tilde{f} t_1 + \frac{1}{2!} (\pm \tilde{f})^2 t_2^2 \quad \text{for} \quad \frac{2r}{c} \leqslant t \leqslant \frac{3r}{c},$$

$$\tilde{C}_{\pm}(t) = 1 \pm \tilde{f} t_1 + \frac{1}{2!} (\pm \tilde{f})^2 t_2^2 + \frac{1}{3!} (\pm \tilde{f})^3 t_3^3$$

$$\text{for} \quad \frac{3r}{c} \leqslant t \leqslant \frac{4r}{c}, \quad \ldots . \tag{14}$$

From Eqs. (14), it can be deduced that the solution of Eqs. (13) is of the form

$$\tilde{C}_{\pm n}(t) = \sum_{k=0}^{n} \frac{1}{k!} (\pm \tilde{f})^k t_k^k \quad \text{for} \quad \frac{nr}{c} \leqslant t \leqslant \frac{(n+1)r}{c},$$

$$n = 0,1,2, \ldots . \tag{15}$$

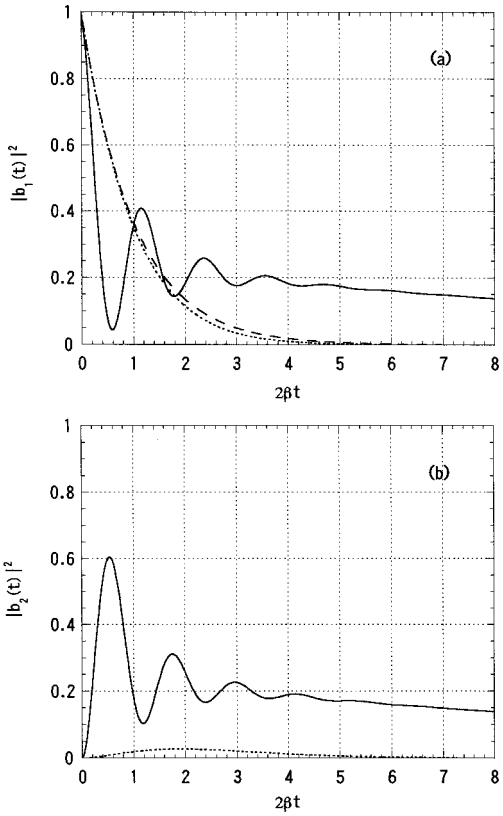

FIG. 1. Upper state populations of (a) atom 1 and (b) atom 2 as functions of the dimensionless time $2\beta t$ for $r = \lambda/2$ (dotted curve), $r = 0.2\lambda/2$ (solid curve), and free-space decay (dashed curve). Atom 1 is excited and atom 2 deexcited at $t = 0$. $\Delta m = \pm 1$ transitions are assumed and $\beta\lambda/c = 10^{-3}$.

To prove that Eqs. (15) are indeed solutions of Eqs. (13), we differentiate both sides of them to obtain

$$\dot{\tilde{C}}_{\pm n}(t) = \sum_{k=1}^{n} \frac{1}{(k-1)!} (\pm \tilde{f})^k t_k^{k-1} = \pm \tilde{f} \sum_{k=0}^{n-1} \frac{1}{k!} (\pm \tilde{f})^k t_{k+1}^k$$

$$= \pm \tilde{f} \tilde{C}_{\pm(n-1)}\left(t - \frac{r}{c}\right), \quad n = 1,2,3, \ldots . \tag{16}$$

It is not difficult to see that these together with the case of $n = 0$, which is trivial, are equivalent to Eqs. (13) and our proof is completed. The piecewise solutions (15), together with Eqs. (12), yield readily the result (11). Thus, we have shown that Eqs. (11) are exact solutions to Eqs. (9) for an arbitrary r and the condition $k_0 r \gtrsim 3$ required by Milonni and Knight [5] is only a specific feature of their method of derivation. Note, however, that the solutions (11) are still bound by the conditions imposed upon the delay differential equations (6) and (9) (see the Appendix).

In Fig. 1, we plot the probabilities of finding atom 1 [Fig. 1(a)] and atom 2 [Fig. 1(b)] in their upper states as functions of the dimensionless time $2\beta t$ for the interatomic distances $r = \lambda/2$ (dotted curve), $r = 0.2\lambda/2$ (solid curve), and for free-space decay (dashed curve). $\Delta m = \pm 1$ transitions are assumed; i.e., the two dipoles are parallel to each other and perpendicular to the interatomic axis z, and $\beta\lambda/c$ is taken to be 10^{-3}—a typical value for the optical range. From Fig. 1(a), it is visible that when the two atoms are half a wave-

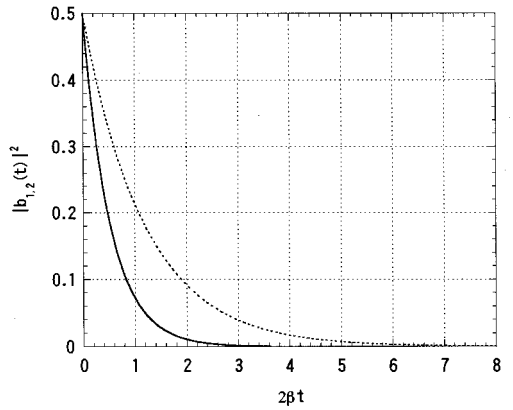

FIG. 2. Upper state populations of atoms 1 and 2 $[|b_1(t)|^2 = |b_2(t)|^2]$ as functions of the dimensionless time $2\beta t$ for the initial state $|+\rangle = (|u,l\rangle + |l,u\rangle)/\sqrt{2}$. Other parameters are the same as in Fig. 1.

length apart, atom 1 decays almost as it does in free space. When the distance between the two atoms decreases, the spontaneous emission is enhanced in agreement with the superradiance feature predicted by Dicke [11] and, as time goes on, the interference produced by multiple reemissions and reabsorptions of light results in an oscillatory energy exchange between atom 1 and atom 2. For longer times, $|b_1(t)|^2|$ and $|b_2(t)|^2$ become equal because of the symmetry inherent in our model. One can also see a considerable trapping of the energy within the atomic system for long times, an effect that can be explained by the fact that the initial atomic state $|u,l\rangle = (|+\rangle + |-\rangle)/\sqrt{2}$, where

$$|\pm\rangle = \frac{1}{\sqrt{2}}(|u,l\rangle \pm |l,u\rangle), \tag{17}$$

contains the antisymmetric state $|-\rangle$ which is a nonradiative one according to the Dicke model [11]. Note that the upper state population of atom 2 starts to increase from zero only after time intervals of r/c, which are too small to be seen in the scale of Fig. 1(b).

When the two atoms are close together, a more realistic initial state is the state $|\pm\rangle$ given in Eq. (17), which allows either atom 1 or atom 2 to be excited with equal probabilities at $t=0$. It is not difficult to solve the delay differential equations (9) under new initial conditions $C_+(0)=\sqrt{2}$, $C_-(0)=0$ by following the same lines outlined in Eqs. (12)–(16) and to find that $C_+(t)$ is equal to $\sqrt{2}$ times the right hand side of Eq. (11) and $C_-(t)=0$. In Fig. 2, we plot the upper state populations of atom 1 and atom 2 $[|b_1(t)|^2=|b_2(t)|^2]$ as functions of the dimensionless time $2\beta t$ with the same parameters as in Fig. 1, except that the initial state is now $|+\rangle$. It can be seen that the spontaneous emission is significantly enhanced for an interatomic distance less than half a wavelength. Numerical calculations (not shown here) also indicate a strong inhibition of spontaneous emission under the initial condition of antisymmetric atomic state $|-\rangle$.

In conclusion, with the aid of the ''method of steps,'' we have found that the solutions by Milonni and Knight to the delay differential equations which govern the radiative decay of a pair of two two-level atoms hold also when the atoms are less than half of a transition wavelength apart, and, as a consequence, we have extended the range of applicability of previous studies on phenomena such as high-order retardation effects [4,5] and dipole-dipole interactions [10]. We have investigated numerically the time behavior of the atomic state populations for different interatomic distances and different atomic initial states, and pointed out the new features appearing when the atoms are less than half of a transition wavelength apart.

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APPENDIX

To calculate the kernels $K_{mn}(t-t')$, Eq. (5), we replace the summation over $\mathbf{k}$ by an integral

$$\sum_{\mathbf{k}} \rightarrow \frac{V}{(2\pi)^3}\sum_{\lambda=1,2}\int d^3k.$$

Using Eqs. (2) and (5), after performing the integrations over the angle variables, we get

$$\begin{aligned} K_{mm}(t-t') &= -\frac{2\mu^2}{3\hbar\pi c^3}\int_0^\infty d\omega\omega^3 e^{-i(\omega-\omega_0)(t-t')} \\ &= -\frac{2\mu^2}{3\hbar\pi c^3}\int_{-\omega_0}^\infty dy(y+\omega_0)^3 e^{-iy(t-t')} \\ &= -2\beta\delta(t-t'), \end{aligned} \tag{A1}$$

where $m=1,2$. In going from the second equation to the third we have retained the dependence on y only in the exponential. This approximation can be explained as follows [12]. A y in $(y+\omega_0)^3$ in the second equation can be replaced by $\partial/\partial[-i(t-t')]$, which would give rise to a derivative of $\delta(t-t')$ with respect to time t in the third equation. This, when put in Eqs. (4), would result in a time derivative $(\partial/\partial t)b_m(t)$. If the time variation of $b_m(t)$ is assumed to be slow compared to the atomic oscillations at the optical frequency ω_0, the terms containing y can be neglected. We have also extended the frequency integral to minus infinity, which is equivalent to not making any rotating-wave approximation [4,5].

In a similar manner, we get

$$\begin{aligned} K_{mn}(t-t') = -\frac{3\beta}{2}\Bigg\{ & \frac{p}{ik_0r}[\,\delta(t-t'-r/c)e^{ik_0r} \\ & -\delta(t-t'+r/c)e^{-ik_0r}] + \frac{q}{i(k_0r)^3} \\ & \times[\,\delta(t-t'-r/c)e^{ik_0r} - \delta(t-t'+r/c)e^{-ik_0r}] \\ & -\frac{q}{(k_0r)^2}[\,\delta(t-t'-r/c)e^{ik_0r} \\ & +\delta(t-t'+r/c)e^{-ik_0r}]\Bigg\}, \end{aligned} \tag{A2}$$

where $m,n=1,2$ $(m\neq n)$ and p and q are defined as in Eqs. (7). Insertion of Eqs. (A1) and (A2) into Eqs. (4) leads to Eqs. (6).

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