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Radiation from an N-Atom System. II. Spontaneous Emission from a Pair of Atoms

R. H. Lehmberg

U. S. Naval Air Development Center, Warminster, Pennsylvania

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Using the formalism developed earlier, we treat spontaneous emission from a pair of identical two-level atoms A_1, A_2, whose separation r_{21} can be comparable to the wavelength λ. We obtain expressions for time-dependent intensities and damping rates with the initial conditions (a) both atoms inverted, (b) prior excitation by a short $\frac{1}{2}\pi$ pulse, and (c) only A_1 inverted. The results in (a) are compared with those obtained for a model consisting of two initially excited harmonic oscillators O_1, O_2. The atoms exhibit superradiant behavior, whereas O_1, O_2 tend to trap radiation. In (a), the intensity pattern $\mathcal{R}(\theta, t)$ develops lobes in different directions at different times, so that the spatial distribution of photons at time $t\rightarrow\infty$ is the same as in the independent-atom case $r_{21}\gg\lambda$. For the oscillators, the lobes of $\mathcal{R}(\theta, t)$ do not change direction, but only become more pronounced as time increases. In (b) and (c), the lobes oscillate back and forth at frequency $2\Omega_{12}$ corresponding to the shifts $\pm\Omega_{12}$ of the triplet and singlet states due to the A_1-A_2 interaction. The intensity can therefore have a sinusoidal component. Field correlation functions calculated for (a) and (c) show that A_2 and A_2 radiate simultaneously around the frequencies $\epsilon\pm\Omega_{12}$, where ϵ is the single-atom resonant frequency. The spectrum is calculated for case (c), and shows the effects of coherent linewidth enhancement in addition to the frequency shifts.

I. INTRODUCTION

In the preceding paper[1] (henceforth referred to as I), a quantum-mechanical formalism was developed to study the radiation properties of N-atom systems. As an illustration, we now apply this formalism to the calculation of spontaneous emission in the case where $N=2$.

Other dynamical treatments of the two-atom problem have either been concerned mostly with interaction energy and coherent linewidth enhancement,[2-5] or with probability amplitudes for particular combinations of photon states.[6] The solutions presented here deal directly with physically measurable quantities, such as emission rates and field correlation functions. The treatment includes time-dependent radiation patterns,[6,7] photon trapping phenomena,[2,3,7] coherent linewidth enhancement, double frequency operation, and optical beating effects. Decay rates and radiation patterns will also be calculated for a model consisting of two harmonic oscillators in place of the atoms. The discrepancies between these two models (e.g., compare Figs. 2 and 3 with 4 and 5) tend to shed doubt on the validity of approximating inverted atoms by excited harmonic oscillators, even for times $t<\gamma^{-1}$.[8]

Although the two-atom system is admittedly an elementary model, it offers several advantages over the multiatom problem. Because of its simplicity, one can obtain detailed and nearly exact dynamical solutions with a variety of initial conditions. The initial conditions treated here include (a) both atoms inverted, (b) prior excitation by a short $\frac{1}{2}\pi$ pulse, and (c) only one atom inverted. Many of the results are analogous to phonomena that one would expect in multiatom systems. For example, the nonexponential decay law (Fig. 3) and simultaneous radiation at two frequencies [Eqs. (50) and (53)] are elementary examples of superradiant pulse formation[7-10] and interaction broadening,[11] respectively. On the other hand, the directional distribution of intensity (Fig. 2) and radiated energy [Eq. (20)] are in contrast to the ray-forming tendencies predicted by photon correlation arguments.[7] In an elementary model, these results (along with those found for the harmonic-oscillator system) can be easily interpreted physically, and thus provide insight into the behavior of more complicated systems. Finally, in a two-atom model, one can study a number of interesting effects that would tend to be obscured in a larger system. Examples of this include the optical beating [Eqs. (36) and (43)] and coherent linewidth enhancement [Eq. (55)] mentioned above.

II. RADIATION RATES

In this section, we solve for the average photon emission rates $W_{\hat{k}}(t)$ and $W(t)$ defined by Eqs. (I34), (I35), and (I31) where I refers to the preceding paper. According to (I37), the spontaneous rates are obtained from the factors

$$Q_{\alpha\beta}(t)\equiv\langle\sigma_\alpha^\dagger(t)\sigma_\beta(t)\rangle = \mathrm{Tr}\rho(0)\langle 0\,|\,\sigma_\alpha^\dagger(t)\,\sigma_\beta(t)\,|\,0\rangle\ , \qquad (1)$$

where $|\,0\rangle$ is the vacuum state and $\rho(0)$ is the initial atomic density operator. One could work directly with these factors, using expression (I29) to obtain equations such as

$$\dot{Q}_{11} = -\gamma Q_{11} - \Gamma_{12} Q_{12} - \Gamma_{12}^{*} Q_{21} \ ,$$

$$\dot{Q}_{12} = -\gamma Q_{12} - \Gamma_{12} Q_{11} - \Gamma_{12}^{*} Q_{22} + 2\gamma_{12}\langle \sigma_1^{\dagger}\sigma_1\sigma_2^{\dagger}\sigma_2\rangle ,$$

$$d\langle \sigma_1^{\dagger}\sigma_1\sigma_2^{\dagger}\sigma_2\rangle/dt = -2\gamma\langle \sigma_1^{\dagger}\sigma_1\sigma_2^{\dagger}\sigma_2\rangle \ , \ \text{etc.} \ ,$$

where $\Gamma_{12} \equiv \frac{1}{2}\gamma_{12} + i\Omega_{12}$ and γ_{12} $[\Omega_{12}]$ are defined by (I20) [(I25)]. It is more instructive, however, to deal with quantities that satisfy separable equations. To this end, we define the transition operators[12]

$$\mathcal{P}_{rm,\,r'm'}(t) \equiv e^{iHt} \,|\, r, m\rangle\langle r', m' \,|\, e^{-iHt} \ , \tag{2}$$

where $|\, r, m\rangle$ can be the singlet or triplet states[7]

$$|\,0, 0\rangle \equiv 2^{-1/2}(|+\rangle_1\,|-\rangle_2 - |-\rangle_1\,|+\rangle_2) \tag{3a}$$

or

$$|\,1, 0\rangle \equiv 2^{-1/2}(|+\rangle_1|-\rangle_2 + |-\rangle_1|+\rangle_2), \tag{3b}$$

$$|\,1, \pm 1\rangle \equiv |\pm\rangle_1\,|\pm\rangle_2 \ , \tag{3c}$$

respectively. These operators satisfy the identities

$$\sigma_1^{\dagger}\sigma_1 + \sigma_2^{\dagger}\sigma_2 = \mathcal{P}_{10,10} + \mathcal{P}_{00,00} + 2\mathcal{P}_{11,11} \ , \tag{4a}$$

$$\sigma_1^{\dagger}\sigma_2 = \tfrac{1}{2}(\mathcal{P}_{10,10} - \mathcal{P}_{00,00} + \mathcal{P}_{00,10} - \mathcal{P}_{10,00}) , \tag{4b}$$

and have the property

$$\langle P_{rm,r'm'}(t)\rangle = \rho_{r'm',rm}(t) \ , \tag{5}$$

where $\rho(t)$ is the reduced density operator containing only atomic variables. Thus, Eq. (I37) can be written as

$$\begin{aligned} W_{\hat{R}}(t) = (3\gamma/8\pi)[1-(\hat{R}\cdot\hat{p})^2]\{&\rho_{10,10}(t) + \rho_{00,00}(t) \\ &+ 2\rho_{11,11}(t) + [\rho_{10,10}(t) - \rho_{00,00}(t)]\cos(\kappa r_{21}\cos\theta) \\ &+ i[\rho_{00,10}(t) - \rho_{10,00}(t)]\sin(\kappa r_{21}\cos\theta)\} , \end{aligned} \tag{6}$$

where

$$\cos\theta \equiv \hat{R}\cdot\hat{r}_{21} \ , \qquad \vec{r}_{21} \equiv \vec{r}_2 - \vec{r}_1 \ , \tag{7}$$

and the R/c factor has been ignored, so that $t_R \to t$.

Substituting expression (2) into (I29), and using (5), we obtain the separable equations[13]

$$\dot{\rho}_{11,11} = -2\gamma\rho_{11,11} \ , \tag{8a}$$

$$\dot{\rho}_{10,10} = (\gamma+\gamma_{12})\rho_{11,11} - (\gamma+\gamma_{12})\rho_{10,10} \ , \tag{8b}$$

$$\dot{\rho}_{00,00} = (\gamma-\gamma_{12})\rho_{11,11} - (\gamma-\gamma_{12})\rho_{00,00} \ , \tag{8c}$$

$$\dot{\rho}_{10,00} = -(2i\Omega_{12}+\gamma)\rho_{10,00} \ . \tag{8d}$$

Equations (8a)–(8c) can be written directly from the transition rates shown in Fig. 1. One can also write (8d) from Fig. 1 by assuming that each level $|\,1, 0\rangle$ and $|\,0, 0\rangle$ contributes to the decay of $\rho_{10,00}$ at a rate equal to half the amount shown. The reason $\rho_{11,11}$ does not appear is that a nonzero value of $\rho_{10,00}$ requires the $|\,1, 0\rangle$ and $|\,0, 0\rangle$ amplitudes to be mutually coherent. Such coherence may be present initially (e.g., as a result of excitation by a $\frac{1}{2}\pi$ pulse), but cannot be produced or enhanced by spontaneous decay from above. Finally, we note that (8d) requires only that $|\,1, 0\rangle$ lie at energy $2\Omega_{12}$ above $|\,0, 0\rangle$.[14] The actual positions of the two levels will be examined in Sec. III.

The solutions of (8) are

$$\rho_{11,11}(t) = \rho_{11,11}(0)\,e^{-2\gamma t} \ , \tag{9a}$$

$$\begin{aligned}\rho_{10,10}(t) = e^{-\gamma t}[&\rho_{10,10}(0)\,e^{-\gamma_{12}t} \\ &+ \rho_{11,11}(0)\,(e^{-\gamma_{12}t} - e^{-\gamma t})(\gamma+\gamma_{12})/(\gamma-\gamma_{12})] ,\end{aligned} \tag{9b}$$

$$\begin{aligned}\rho_{00,00}(t) = e^{-\gamma t}[&\rho_{00,00}(0)e^{\gamma_{12}t} \\ &+ \rho_{11,11}(0)(e^{\gamma_{12}t} - e^{-\gamma t})(\gamma-\gamma_{12})/(\gamma+\gamma_{12})] ,\end{aligned} \tag{9c}$$

$$\rho_{10,00}(t) = \rho_{10,00}(0)e^{-(\gamma+2i\Omega_{12})t} = \rho_{00,10}^{*}(t) . \tag{9d}$$

Note that as $\vec{r}_1 \to \vec{r}_2$ (so that $\gamma_{12} \to \gamma$),

$$\rho_{10,10}(t) \to e^{-2\gamma t}[\rho_{10,10}(0) + 2\gamma t\rho_{11,11}(0)] , \tag{10a}$$

$$\rho_{00,00}(t) \to \rho_{00,00}(0) , \tag{10b}$$

in agreement with earlier results.[12]

If the atoms are replaced by harmonic oscillators O_1, O_2, then the average in (I37) must be replaced by $B_{\alpha\beta}(t) \equiv \langle b_{\alpha}^{\dagger}(t)\,b_{\beta}(t)\rangle$, where b_{α} is the lowering operator for the αth oscillator. From (I29), we obtain

$$\dot{B}_{11} = -\gamma B_{11} - \Gamma_{12}B_{12} - \Gamma_{12}^{*}B_{12}^{*} \ , \tag{11a}$$

$$\dot{B}_{22} = -\gamma B_{22} - \Gamma_{12}B_{12}^{*} - \Gamma_{12}^{*}B_{12} \ , \tag{11b}$$

$$\dot{B}_{12} = -\gamma B_{12} - \Gamma_{12}B_{11} - \Gamma_{12}^{*}B_{22} \ . \tag{11c}$$

The quantities

$$B_{\pm}' \equiv B_{11} + B_{22} \pm B_{12} \pm B_{12}^{*} \ , \tag{12a}$$

$$B_{\pm}'' \equiv B_{11} - B_{22} \pm B_{12} \mp B_{12}^{*} \ , \tag{12b}$$

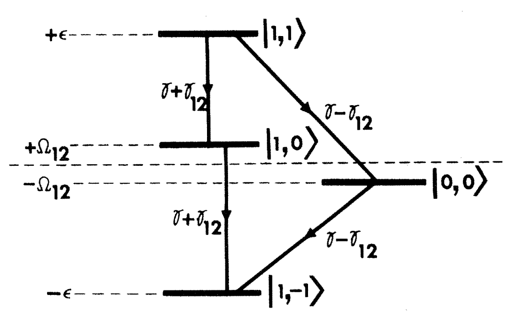

FIG. 1. Energy-level diagram for the $|\,r, m\rangle$ states, showing the frequency shifts $\pm\Omega_{12}(\hbar \equiv 1)$ and decay constants $\gamma \pm \gamma_{12}$.

satisfy separable equations with exponential solutions; hence, one can easily show that

$$B_{11}(t)+B_{22}(t)=\tfrac{1}{2}\left[B_{+}'(0)e^{-\gamma_{12}t}+B_{-}'(0)e^{\gamma_{12}t}\right]e^{-\gamma t}, \quad (13a)$$

$$B_{12}(t)=\tfrac{1}{4}\left[B_{+}'(0)e^{-\gamma_{12}t}-B_{-}'(0)e^{\gamma_{12}t} + B_{+}''(0)e^{-2i\Omega_{12}t}-B_{-}''(0)e^{2i\Omega_{12}t}\right]e^{-\gamma t}. \quad (13b)$$

π-Pulse Excitation

If the atoms are initially excited into their upper states (e.g., by a short π pulse), then

$$\rho_{11,11}(0)=1, \quad \rho_{10,10}(0)=\rho_{00,00}(0)=\rho_{10,00}(0)=0. \quad (14)$$

Equations (6) and (9) then yield the average photon emission rate

$$W_{\hat{R}}(t)=2W_{\hat{R}}^{(1)}(t)\,\mathcal{R}(\theta,t), \quad (15)$$

where

$$W_{\hat{R}}^{(1)}(t)\equiv(3\gamma/8\pi)[1-(\hat{R}\cdot\hat{p})^2]e^{-\gamma t} \quad (16)$$

is the radiation rate of a single isolated atom, and

$$\mathcal{R}(\theta,t)\equiv A(t)+B(t)\cos(\kappa r_{21}\cos\theta), \quad (17)$$

$$A(t)\equiv\tfrac{1}{2}(e^{-\gamma_{12}t}-e^{-\gamma t})(\gamma+\gamma_{12})(\gamma-\gamma_{12})^{-1} + \tfrac{1}{2}(e^{\gamma_{12}t}-e^{-\gamma t})(\gamma-\gamma_{12})(\gamma+\gamma_{12})^{-1}+e^{-\gamma t}, \quad (18a)$$

$$B(t)\equiv\tfrac{1}{2}(e^{-\gamma_{12}t}-e^{-\gamma t})(\gamma+\gamma_{12})(\gamma-\gamma_{12})^{-1} - \tfrac{1}{2}(e^{\gamma_{12}t}-e^{-\gamma t})(\gamma-\gamma_{12})(\gamma+\gamma_{12})^{-1}. \quad (18b)$$

If the atoms radiate independently (so that $\gamma_{12}=0$), then $\mathcal{R}(\theta,t)=1$.

The asymptotic values of $\mathcal{R}(\theta,t)$ are

$$\mathcal{R}(\theta,t)\simeq 1+\gamma_{12}t\cos(\kappa r_{21}\cos\theta), \quad \gamma t\ll 1, \quad (19a)$$

$$\mathcal{R}(\theta,t) \simeq\frac{1}{2}\,\frac{\gamma-|\gamma_{12}|}{\gamma+|\gamma_{12}|}\left[1-\frac{\gamma_{12}}{|\gamma_{12}|}\cos(\kappa r_{21}\cos\theta)\right]e^{|\gamma_{12}|t}, \quad \gamma_{12}t\gg 1. \quad (19b)$$

Apparently, $R(\theta,t)$ starts out spherically symmetric, develops a weak nonspherical radiation pattern, becomes spherically symmetric again, then begins to develop pronounced minima at the same angles at which the earlier pattern was maximum.

To explain such behavior in terms of $|r,m\rangle$ states, we consider as an example, the case $\kappa r_{21}=\frac{1}{2}\pi$ (Fig. 2). Since $\gamma_{12}>0$ in this case it is clear from Fig. 1 [or Eqs. (9) and (14)] that $\rho_{10,10}(t)>\rho_{00,00}(t)$ for the early times $\gamma t\leq 1$. However, level $|1,0\rangle$ also decays more rapidly than $|0,0\rangle$, so that $\rho_{10,10}(t)<\rho_{00,00}(t)$ when $\gamma t\gg 1$. Since A_1 and A_2 are a quarter wavelength apart, the radiation from symmetric state $|1,0\rangle$ has its maximum intensity in the plane $\theta=90°$, whereas the contributions from antisymmetric state $|0,0\rangle$ would tend to cancel in these directions.

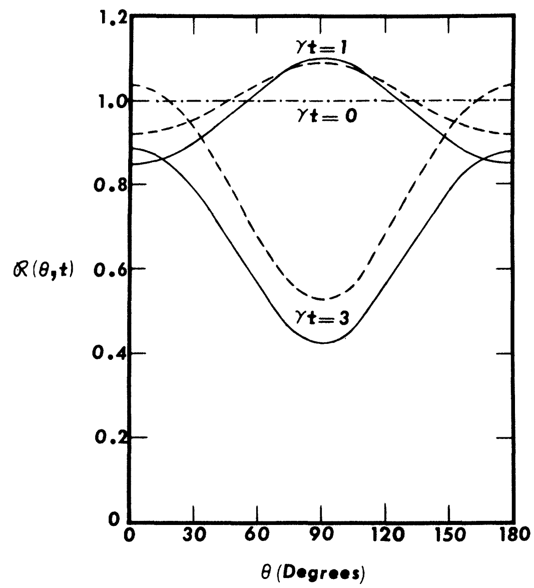

FIG. 2. Radiation pattern $\mathcal{R}(\theta,t)$ [defined by Eqs. (15) and (17)] at times $t=0$, γ^{-1}, and $3\gamma^{-1}$, for two atoms (initially inverted) with spacing $r_{21}=\frac{1}{4}\lambda$. Solid and dashed curves denote $\vec{p}\parallel\hat{r}_{21}$ and $\vec{p}\perp\hat{r}_{21}$, respectively, while the dot-dashed curve applies to either case. Here, γ is the decay constant of a single atom, θ is the angle between $\hat{r}_{21}$ and observation direction $\hat{R}$, and $\vec{p}$ is the dipole matrix element.

An interesting consequence of Eqs. (15)–(18) is that the average number of photons $n_{\hat{R}}$ emitted along $\hat{R}$ is independent of the A_1 - A_2 interaction; i.e.,

$$n_{\hat{R}}\equiv\int_0^\infty dt\,W_{\hat{R}}(t) =(3/4\pi)[1-(\hat{R}\cdot\hat{p})^2]=2\int_0^\infty dt\,W_{\hat{R}}^{(1)}(t). \quad (20)$$

These results, especially (20), seem to contradict the prediction of photon-correlation arguments[7] that the radiative coupling between A_1 and A_2 enhances emmission in certain directions. This point will be discussed in more detail in Sec. IV.

Integrating (15) over all $\Omega_{\hat{R}}$, we obtain the total emission rate

$$W(t)=2\gamma[A(t)+(\gamma_{12}/\gamma)B(t)]e^{-\gamma t} \quad (21a)$$

$$\xrightarrow[\gamma_{12}t\gg 1]{}\frac{\gamma(1-|\gamma_{12}|/\gamma)^2}{1+|\gamma_{12}|/\gamma}\,e^{-(\gamma-|\gamma_{12}|)t}. \quad (21b)$$

For $\vec{r}_2\to\vec{r}_1$ (so that $\gamma_{12}\to\gamma$), Eq. (21a) becomes

$$W(t)\to 2\gamma(1+2\gamma t)e^{-2\gamma t}, \quad (22)$$

in agreement with earlier results.[12,15] The curves of $W(t)/W(0)=W(t)/2\gamma$ (Fig. 3) show a clear tendency for the coupled atoms ($\kappa r_{21}\ll 1$ and $\kappa r_{21}=\frac{1}{2}\pi$)

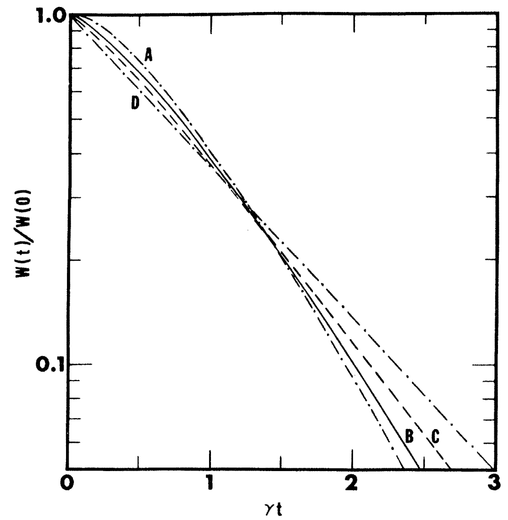

FIG. 3. Time dependence of the normalized damping rate $W(t)/W(0)$ for two atoms (initially inverted) with spacing: A: $r_{21} \ll \lambda(\vec{p} \parallel \vec{r}_{21}$ or $\perp \vec{r}_{21})$, B: $r_{21} = \frac{1}{4}\lambda(\vec{p} \parallel \vec{r}_{21})$, C: $r_{21} = \frac{1}{4}\lambda(\vec{p} \perp \vec{r}_{21})$, D: $r_{21} \ll \lambda(\vec{p} \parallel \vec{r}_{21}$ or $\perp \vec{r}_{21})$.

times $\gamma t < 1$, when they are both excited. The asymptotic forms of (24) and (25) for $\gamma_{12} t \gg 1$ are

$$\mathcal{R}(\theta, t) \simeq \tfrac{1}{2}[1 - (\gamma_{12}/|\gamma_{12}|)\cos(\kappa r_{21}\cos\theta)]\, e^{|\gamma_{12}|t}, \tag{26}$$

to decay more rapidly than they would if they were independent ($\kappa r_{21} \gg 1$). This is an elementary example of the superradiant pulse formation that one would expect in a larger system, and can be regarded, at least in part, as due to stimulated emission.

If A_1 and A_2 are replaced by harmonic oscillators initially in their lowest excited states, i.e.,

$$B_{11}(0) = B_{22}(0) = 1\ , \quad B_{12}(0) = 0\ , \tag{23}$$

then according to (13a), (13b) and (I37) (with $\sigma_\alpha \to b_\alpha$), we retain Eq. (15) with $R(\theta, t)$ now given by

$$\mathcal{R}(\theta, t) = \cosh(\gamma_{12} t) \times [1 - \tanh(\gamma_{12} t)\cos(\kappa r_{21}\cos\theta)]\ . \tag{24}$$

This pattern obviously has the same form for all $\gamma t > 0$, as shown, for example, in Fig. 4. (Note that the lobes need not be along $\pm \hat{r}_{21}$ in all cases; e.g., if $\hat{p} \perp \hat{r}_{21}$ and $\kappa r_{21} = \pi$, then $\gamma_{12} < 0$, and the lobes occur for $\hat{R} \perp \hat{r}_{21}$.)

The total emission rate is

$$W(t) = 2\gamma[1 - (\gamma_{12}/\gamma)\tanh(\gamma_{12} t)]\cosh(\gamma_{12} t)\, e^{-\gamma t}, \tag{25}$$

which is plotted in Fig. 5 for $\kappa r_{21} \ll 1$, $\kappa r_{21} = \frac{1}{2}\pi$, and $\kappa r_{21} \gg 1$. The first two curves show clearly that the oscillators tend to trap radiation, even at

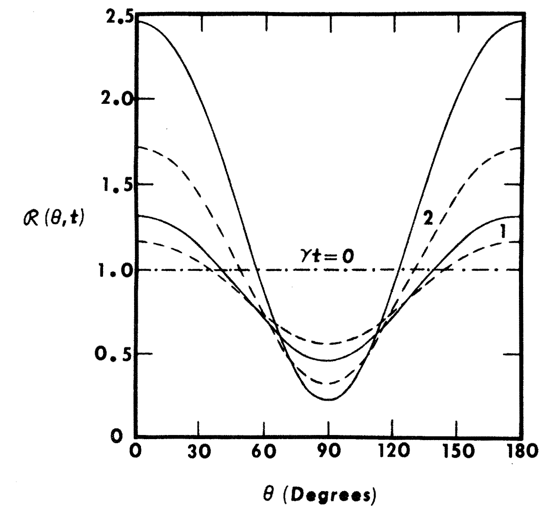

FIG. 4. Radiation pattern $\mathcal{R}(\theta, t)$ [defined by Eqs. (15), and (24)] at times $t = 0$, γ^{-1}, and $2\gamma^{-1}$, for two harmonic oscillators (initially in their first excited states) with spacing $r_{21} = \frac{1}{4}\lambda$. Solid and dashed curves denote $\vec{p} \parallel \vec{r}_{21}$ and $\vec{p} \perp \vec{r}_{21}$, respectively, while the dot-dashed curve applies to either case.

$$W(t) \simeq 2\gamma(1 - |\gamma_{12}|/\gamma)\, e^{-(\gamma - |\gamma_{12}|)t}\ , \tag{27}$$

which have the same (θ, t) dependence as the corresponding expressions (19b) and (21b) in the atomic case.

One can interpret many of these results in terms of a heuristic classical model. In particular, con-

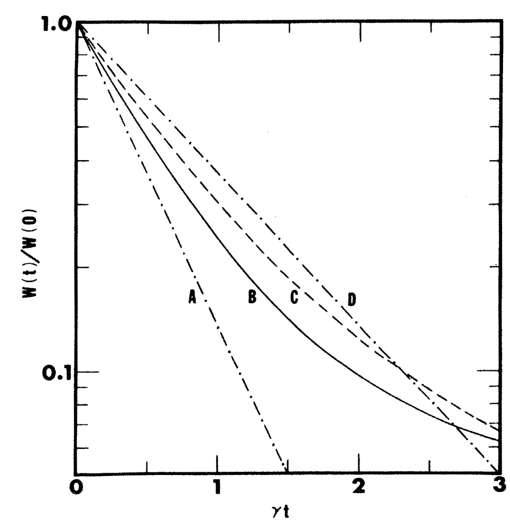

FIG. 5. Time dependence of the normalized damping rate $W(t)/W(0)$ for two harmonic oscillators (initially in their first excited states) with spacing: A: $r_{21} \ll (\vec{p} \parallel \vec{r}_{21}$ or $\perp \vec{r}_{21})$, B: $r_{21} = \frac{1}{4}\lambda(\vec{p} \parallel \vec{r}_{21})$, C: $r_{21} = \frac{1}{4}\lambda(\vec{p} \perp \vec{r}_{21})$, D: $r_{21} \ll \lambda(\vec{p} \parallel \vec{r}_{21}$ or $\perp \vec{r}_{21})$.

sider a pair of classical dipoles located at points $\vec{r}_1$, $\vec{r}_2$, with time dependence

$$\vec{p}_\alpha(t) \equiv \vec{p} \exp\{-i\epsilon t - i\psi_\alpha(t) - \tfrac{1}{2}\gamma t + \psi'(t)\}, \tag{28}$$

where $\psi'(t)$ is a real function, satisfying

$$|\psi'(t)| < \tfrac{1}{2}\gamma t, \quad \psi'(0) = 0.$$

The phase functions $\psi_\alpha(t)$ are random variables, assumed to be initially uncorrelated; i. e.,

$$\langle \cos \psi_{21}(0)\rangle = \langle \sin \psi_{21}(0)\rangle = 0,$$

where $\psi_{21} \equiv \psi_2 - \psi_1$, and angular brackets now denote a classical ensemble average. From symmetry considerations, it follows that $\langle \sin \psi_{21}(t)\rangle = 0$ for all times. The emission rates of the far field can therefore be written as

$$W_{\hat{R}}(t) = 2W_{\hat{R}}^{(1)}(t)\, e^{2\psi'(t)} \times [1 + \langle \cos \psi_{21}(t)\rangle \cos(K\gamma_{21}\cos\theta)], \tag{29}$$

$$W(t) = 2\gamma[1 + (\gamma_{12}/\gamma)\langle \cos\psi_{21}(t)\rangle]\, e^{-\gamma t + 2\psi'(t)}. \tag{30}$$

If $\vec{p}_1(t)$ and $\vec{p}_2(t)$ are harmonic oscillators, they exhibit positive susceptibilities[16]; thus, $\psi_1(t)$ and $\psi_2(t)$ will tend to correlate so as to trap the radiation. Hence, according to Eq. (30), we obtain

$$\gamma_{12}\langle \cos \psi_{21}(t)\rangle \leq 0$$

or

$$\langle \cos \psi_{21}(t)\rangle = -\,|\langle \cos \psi_{21}(t)\rangle|\,\gamma_{12}/|\gamma_{12}|, \tag{31}$$

and (29) has a radiation pattern similar to (24).

If $\vec{p}_1(t)$ and $\vec{p}_2(t)$ represent two-level atoms that are inverted at time $t=0$, then their initial susceptibilities will be negative, and the phases will, at first, tend to correlate so that

$$\langle \cos \psi_{21}(t)\rangle = +\,|\langle \cos \psi_{21}(t)\rangle|\,\gamma_{12}/|\gamma_{12}|. \tag{32}$$

For later times, when the upper levels are nearly depopulated, the susceptibilities become positive, and the phases realign so as to satisfy (31). Thus we see that (29) has the same type of radiation pattern as (19a) and (19b).

$\frac{1}{2}\pi$-Pulse Excitation

We now consider the case where atoms A_1, A_2 have been excited by a short $\frac{1}{2}\pi$ pulse of wave vector $\vec{k}$. The initial atomic state is then

$$\tfrac{1}{2}[\,|-\rangle_1 + i\eta \exp(i\vec{k}\cdot\vec{r}_1)|+\rangle][\,|-\rangle_2 + i\eta \exp(i\vec{k}\cdot\vec{r}_2)|+\rangle_2], \tag{33}$$

where $|\eta|^2 = 1$; therefore,

$$\rho_{11,11}(0) = \tfrac{1}{4}, \tag{34a}$$

$$\rho_{10,10}(0) = \tfrac{1}{4}(1 + \cos\vec{k}\cdot\vec{r}_{21}), \tag{34b}$$

$$\rho_{00,00}(0) = \tfrac{1}{4}(1 - \cos\vec{k}\cdot\vec{r}_{21}), \tag{34c}$$

$$\rho_{10,00}(0) = \tfrac{1}{4} i \sin\vec{k}\cdot\vec{r}_{21}. \tag{34d}$$

Equations (6) and (9) yield

$$W_{\hat{R}}(t) = W_{\hat{R}}^{(1)}(t)\, \mathcal{R}'(\theta, \vec{k}, t), \tag{35}$$

where

$$\mathcal{R}'(\theta,\vec{k},t) = \tfrac{1}{2}\{A(t) + \cosh(\gamma_{12}t) - \sinh(\gamma_{12}t)\cos\vec{k}\cdot\vec{r}_{21} + [B(t) - \sinh(\gamma_{12}t) + \cosh(\gamma_{12}t)\cos\vec{k}\cdot\vec{r}_{21}] \times \cos(\kappa r_{21}\cos\theta) + \cos(2\Omega_{12}t)\sin\vec{k}\cdot\vec{r}_{21}\sin(\kappa r_{21}\cos\theta)\}. \tag{36}$$

For $t=0$,

$$\mathcal{R}'(\theta,\vec{k},0) = 1 + \tfrac{1}{2}\cos(\vec{k} - \kappa\hat{R})\cdot\vec{r}_{21}, \tag{37}$$

in agreement with Dicke's result.[7] In the independent-atom limit where $\gamma_{12} = \Omega_{12} = 0$, expression (37) applies at all times.

For $\vec{k}\cdot\vec{r}_{21} \neq 0$, the maximum of $R'(\theta,\vec{k},t)$ occurs initially along $\hat{R} = +\hat{k}$, and oscillates back and forth between $+\hat{k}$ and $-\hat{k}$ at frequency $2\Omega_{12}$ (e.g., see Fig. 6). The intensity in a given direction (e.g.,

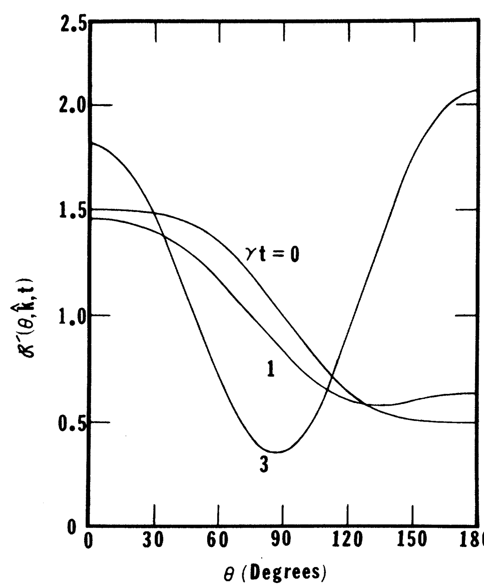

FIG. 6. Radiation pattern $\mathcal{R}'(\theta,\hat{k},t)$ [defined by Eqs. (35) and (36)] at times $t=0$, γ^{-1}, and $3\gamma^{-1}$, for two atoms initially excited by a short $\pi/2$ pulse. The atomic spacing is $\frac{1}{4}\lambda$. The pulse, at frequency $\omega = \epsilon$, is directed along $\vec{r}_{21}$, with polarization along $\vec{p}(\perp\vec{r}_{21})$.

$\hat{R}=\hat{k}$) therefore has a sinusoidal component. For $\gamma_{12}t \gg 1$,

$$\mathcal{R}'(\theta,\vec{k},t) \propto \tfrac{1}{2}\left[1-(\gamma_{12}/|\gamma_{12}|)\cos(\kappa r_{21}\cos\theta)\right]e^{|\gamma_{12}|t}, \quad (38)$$

which has the same θ, t dependence as the corresponding patterns (19b) and (26) in the π-pulse cases.

The oscillation arises because the definite phase relationship between levels $|1,0\rangle$ and $|0,0\rangle$ (established by the exciting pulse) changes periodically on account of their frequency difference $2\Omega_{12}$ (see Fig. 1). Similar effects have been discussed in connection with fine or hyperfine splitting in single atoms.[17] In the case of π-pulse excitation, no such phase correlation existed at the outset, nor can it be created by spontaneous decay from $|1,1\rangle$.

The total emission rate

$$W(\vec{k},t)=\tfrac{1}{2}\gamma\{A(t)+\cosh(\gamma_{12}t)-\sinh(\gamma_{12}t)\cos\vec{k}\cdot\vec{r}_{21} + (\gamma_{12}/\gamma)[B(t)-\sinh(\gamma_{12}t)+\cosh(\gamma_{12}t)\cos\vec{k}\cdot\vec{r}_{21}]\}e^{-\gamma t} \quad (39)$$

contains no periodic component. For the case where $\kappa r_{21} \ll 1$,

$$W(\vec{k},t)\to\gamma\left(\tfrac{3}{2}+\gamma t\right)e^{-2\gamma t},$$

in agreement with earlier results.[12]

Single-Atom Initial Excitation

As a final example, consider the case where only atom A_1 is excited initially; i.e.,

$$|\Psi(0)\rangle = |+\rangle_1|-\rangle_2|0\rangle, \quad (40)$$

and therefore

$$\rho_{10,10}(0)=\rho_{00,00}(0)=\rho_{10,00}(0)=\rho_{00,10}(0)=\tfrac{1}{2}, \quad (41a)$$

$$\rho_{11,11}(0)=\rho_{1-1,1-1}(0)=0 . \quad (41b)$$

The probability of finding only A_1 excited at time t is, according to Eqs. (9),

$${}_1\langle+|{}_2\langle-|\rho(t)|+\rangle_1|-\rangle_2=\tfrac{1}{2}e^{-\gamma t}[\cosh(\gamma_{12}t)+\cos(\Omega_{12}t)];$$

similarly, for A_2,

$${}_1\langle-|{}_2\langle+|\rho(t)|-\rangle_1|+\rangle_2 = \tfrac{1}{2}e^{-\gamma t}[\cosh(\gamma_{12}t)-\cos(\Omega_{12}t)] .$$

These results agree with those of Hutchinson and Hameka,[3] and provide a simple illustration of the radiation trapping discussed above. The photon evidently tends to be handed back and forth from one atom to the other, while the excitation decays as $e^{-(\gamma-\gamma_{12})t}$ for $\gamma_{12}t \gg 1$. Such effects also show up in the radiation rates

$$W_{\hat{R}}(t)=W_{\hat{R}}^{(1)}(t)\mathcal{R}(\theta,t), \quad (42)$$

where

$$\mathcal{R}(\theta,t)=\cosh\gamma_{12}t-\sinh(\gamma_{12}t)\cos(\kappa r_{21}\cos\theta) - \sin(\Omega_{12}t)\sin(\kappa r_{21}\cos\theta), \quad (43)$$

$$W(t)=\gamma[1-(\gamma_{12}/\gamma)\tanh(\gamma_{12}t)]\cosh(\gamma_{12}t)e^{-\gamma t}. \quad (44)$$

It is interesting to note that Eqs. (42)–(44) are also obtained for the analogous harmonic-oscillator model.

III. SPECTRAL PROPERTIES

In this section, we investigate the spectral properties of the radiation by evaluating expression (I40). If $t'=0$, then one can calculate this exactly from atomic operators

$$\langle\mathcal{P}_{rm,r'm-1}(t)\rangle_0=\langle 0|\mathcal{P}_{rm,r'm-1}(t)|0\rangle$$

by using the identities

$$\sigma_1^\dagger=2^{-1/2}(\mathcal{P}_{11,10}-\mathcal{P}_{11,00}+\mathcal{P}_{10,1-1}+\mathcal{P}_{00,1-1}), \quad (45a)$$

$$\sigma_2^\dagger=2^{-1/2}(\mathcal{P}_{11,10}+\mathcal{P}_{11,00}+\mathcal{P}_{10,1-1}-\mathcal{P}_{00,1-1}), \quad (45b)$$

and noting that

$$\langle\sigma_\alpha^\dagger(t)\sigma_\beta(0)\rangle = \mathrm{Tr}\rho(0)\langle\sigma_\alpha^\dagger(t)\rangle_0\sigma_\beta(0). \quad (46)$$

Expression (I40) can then be written as

$$f(t,0)=2^{-1/2}\,\mathrm{Tr}\rho(0)\{[\langle\mathcal{P}_{10,1-1}(t)\rangle_0+\langle\mathcal{P}_{11,10}(t)\rangle_0] \times[\sigma_1(0)+\sigma_2(0)](\gamma+\gamma_{12})+[\langle\mathcal{P}_{00,1-1}(t)\rangle_0 - \langle\mathcal{P}_{11,00}(t)\rangle_0][\sigma_1(0)-\sigma_2(0)](\gamma-\gamma_{12})\}. \quad (47)$$

From (I29), we obtain

$$\langle\dot{\mathcal{P}}_{11,10}\rangle_0=[i(\epsilon-\Omega_{12})-\tfrac{1}{2}(3\gamma+\gamma_{12})]\langle\mathcal{P}_{11,10}\rangle_0, \quad (48a)$$

$$\langle\dot{\mathcal{P}}_{11,00}\rangle_0=[i(\epsilon+\Omega_{12})-\tfrac{1}{2}(3\gamma-\gamma_{12})]\langle\mathcal{P}_{11,00}\rangle_0, \quad (48b)$$

$$\langle\dot{\mathcal{P}}_{10,1-1}\rangle_0=(\gamma+\gamma_{12})\langle\mathcal{P}_{11,10}\rangle_0 +[i(\epsilon+\Omega_{12})-\tfrac{1}{2}(\gamma+\gamma_{12})]\langle\mathcal{P}_{10,1-1}\rangle_0, \quad (48c)$$

$$\langle\dot{\mathcal{P}}_{00,1-1}\rangle_0=-(\gamma-\gamma_{12})\langle\mathcal{P}_{11,00}\rangle_0 +[i(\epsilon-\Omega_{12})-\tfrac{1}{2}(\gamma-\gamma_{12})]\langle\mathcal{P}_{00,1-1}\rangle_0, \quad (48d)$$

which confirm the location of levels $|1,0\rangle$ and $|0,0\rangle$ in Fig. 1, and yield the general solutions

$$\langle\mathcal{P}_{11,10}(t)\rangle_0=\mathcal{P}_{11,10}(0)\exp[i(\epsilon-\Omega_{12})-\tfrac{1}{2}(3\gamma+\gamma_{12})]t, \quad (49a)$$

$$\langle\mathcal{P}_{11,00}(t)\rangle_0=\mathcal{P}_{11,00}(0)\exp[i(\epsilon+\Omega_{12})-\tfrac{1}{2}(3\gamma-\gamma_{12})]t, \quad (49b)$$

$$\langle\mathcal{P}_{10,1-1}(t)\rangle_0=\exp[i(\epsilon+\Omega_{12})-\tfrac{1}{2}(\gamma+\gamma_{12})]t\{P_{10,1-1}(0) +(\gamma+\gamma_{12})(\gamma+2i\Omega_{12})^{-1}[1-e^{-(\gamma+2i\Omega_{12})t}]P_{11,10}(0)\}, \quad (49c)$$

$$\langle \mathcal{P}_{00,1-1}(t)\rangle_0 = \exp[i(\epsilon - \Omega_{12}) - \tfrac{1}{2}(\gamma - \gamma_{12})]\, t\{\mathcal{P}_{00,1-1}(0) - (\gamma - \gamma_{12})(\gamma - 2i\Omega_{12})^{-1}[1 - e^{-(\gamma - 2i\Omega_{12})t}]\,\mathcal{P}_{11,00}(0)\}. \tag{49d}$$

As an example, consider the case of π-pulse excitation described by Eqs. (14). Combining (47) and (49), one obtains

$$f(t,0) = C^{+}(t)\exp[i(\epsilon + \Omega_{12}) - \tfrac{1}{2}(\gamma + \gamma_{12})]t + C^{-}(t)\exp[i(\epsilon - \Omega_{12}) - \tfrac{1}{2}(\gamma - \gamma_{12})]t\ , \tag{50}$$

where

$$C^{\pm}(t) \equiv (\gamma \pm \gamma_{12})^2(\gamma \pm 2i\Omega_{12})^{-1} \pm (\gamma - 2i\Omega_{12}) \times (\gamma \mp \gamma_{12})(\gamma \mp 2i\Omega_{12})^{-1} e^{-(\gamma \mp \gamma_{12})t} \tag{51}$$

is nonperiodic. The system therefore radiates simultaneously around the frequencies $\epsilon \pm \Omega_{12}$, an effect analogous to interaction broadening in multiatom systems.[11] The periodic intensity in Sec. II can then be regarded as a beat note whose (ensemble) average phase is nonzero only if an initial correlation exists between $|1,0\rangle$ and $|0,0\rangle$. For γt and $\Omega_{12}t \ll 1$,

$$f(t,0) \simeq 2\gamma e^{i\epsilon t} \xrightarrow[t\to 0]{} W(0)\ .$$

For $\gamma_{12}t \gg 1$,

$$f(t,0) \propto \exp[i(\epsilon - \Omega_{12}) - \tfrac{1}{2}(\gamma - \gamma_{12})]t\ ,$$

which comes entirely from the $|0,0\rangle \to |1,-1\rangle$ transition.

So far, we have used no approximation except those described in I; however, in order to treat the general case where $t' \neq 0$ in (I40), one must assume the validity of the fluctuation-regression theorem. This theorem has been derived elsewhere,[18] and for the present purposes, can be written as[12]

$$\langle \sigma_\alpha^\dagger(t)\sigma_\beta(t')\rangle \simeq \langle e^{iHt'}\langle \sigma_\alpha^\dagger(t-t')\rangle_0 e^{-iHt'}\sigma_\beta(t')\rangle\ ,$$

where $t \geq t' \geq 0$. To implement it, one simply replaces all of the time arguments in Eqs. (47) and (49) according to the prescription

$$0 \to t'\ ,\ t \to t - t'\ . \tag{52}$$

Since the actual calculation can be tedious, it will be carried out only for the simple case where $|\Psi(0)\rangle = |+\rangle_1|-\rangle_2|0\rangle$ [Eqs. (41)]. The result is

$$f(t,t') = \tfrac{1}{2}(\gamma + \gamma_{12})\exp[i(\epsilon + \Omega_{12}) - \tfrac{1}{2}(\gamma + \gamma_{12})](t - t') \times \exp[-(\gamma + \gamma_{12})t'] + \tfrac{1}{2}(\gamma - \gamma_{12})\exp[i(\epsilon - \Omega_{12}) - \tfrac{1}{2}(\gamma - \gamma_{12})](t - t')\exp[-(\gamma - \gamma_{12})t']\ , \tag{53}$$

which reduces to (44) for $t' = t$.

Defining the spectral distribution of the radiation by

$$w(\omega) \equiv \mathrm{Re}\left\{\int_0^\infty dt \int_0^\infty dt' f(t,t')\, e^{-i\omega(t-t')}\right\}\ , \tag{54}$$

and noting that $f(t',t) = f^*(t,t')$, we obtain

$$w(\omega) = \frac{1}{2}\frac{\gamma + \gamma_{12}}{(\epsilon + \Omega_{12} - \omega)^2 + \frac{1}{4}(\gamma + \gamma_{12})^2} + \frac{1}{2}\frac{\gamma - \gamma_{12}}{(\epsilon - \Omega_{12} - \omega)^2 + \frac{1}{4}(\gamma - \gamma_{12})^2}\ . \tag{55}$$

Except for the shifts $\pm\Omega_{12}$ in the resonant frequencies, this result agrees with that of Lee and Lin[5] if the dipole matrix element $\vec{p}$ is averaged over all directions.

IV. DISCUSSION

The results shown in Fig. 2 and Eq. (20) seem puzzling at first, because they appear to contradict the predictions based upon photon correlation arguments.[7] These arguments can be stated as follows: If $I(\hat{k}_2)$ is the probability per unit time for finding a photon in direction $\hat{k}_2$ immediately after one is observed along $\hat{k}_1$, then[7]

$$I(\hat{k}_2) = I_0(\hat{k}_2)[1 + \cos\kappa(\hat{k}_2 - \hat{k}_1)\cdot\vec{r}_{21}]\ , \tag{56}$$

where $I_0(\hat{k}_2)$ is the probability rate for a single atom. The photons therefore tend to "bunch" around $\hat{k}_2 \simeq \hat{k}_1$, an effect that can be attributed to correlation between the atomic dipole moments. *If* this correlation persists throughout the decay process, then the radiation pattern $\mathcal{R}(\theta,t)$ observed at $\gamma t < 1$ simply becomes more pronounced at later times. The harmonic-oscillator model does behave in this fashion (see Fig. 4), but in the atomic model the initial correlation reverses itself, and $\mathcal{R}(\theta,t)$ changes accordingly. Photon bunching arguments are therefore unreliable (at least in the atomic case) for even a qualitative treatment of the dynamical behavior.

The solutions presented in this paper constitute only one application of the general formalism derived in I. Future work will concentrate on other examples, including (a) spontaneous emission from a multiatom system, and (b) fluorescence and scattering from two or more atoms driven by an external radiation field.

The multiatom case is complicated by the fact that the transition operators $\mathcal{P}_{rm,rm}$ do not satisfy separable equations for $N > 2$ unless $\kappa(r_{\alpha\beta})_{\max} \ll 1$. Thus, it will be necessary to impose additional restrictions, such as averaging over a smeared out distribution of atomic positions, and treating only the case where all atoms are initially inverted.

One would expect some of the phenomena discussed in this paper (e.g., coherent linewidth enhancement) to affect the response of a two-atom system to external radiation. For example, if it is driven by a weak field of wave vector $\vec{k}=\hat{k}\,\omega/c$, then only the $|1,-1\rangle \rightleftharpoons |1,0\rangle$ and $|1,-1\rangle \rightleftharpoons |0,0\rangle$ transitions will contribute appreciably to the scattering (Fig. 1). Since

$$|\langle 1,0|H_{\mathrm{em}}|1,\,-1\rangle|^2 \propto 1+\cos\vec{k}\cdot\vec{r}_{21}\,,$$

$$|\langle 0,0|H_{\mathrm{em}}|1,\,-1\rangle|^2 \propto 1-\cos\vec{k}\cdot\vec{r}_{21}\,,$$

we then expect the total scattering cross section to have the doubly resonant form

$$\sigma(\omega)\propto \frac{(1+\cos\vec{k}\cdot\vec{r}_{21})(\gamma+\gamma_{12})}{(\epsilon+\Omega_{12}-\omega)^2+\frac{1}{4}(\gamma+\gamma_{12})^2} + \frac{(1-\cos\vec{k}\cdot\vec{r}_{21})(\gamma-\gamma_{12})}{(\epsilon-\Omega_{12}-\omega)^2+\frac{1}{4}(\gamma-\gamma_{12})^2}\,. \tag{57}$$

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[1]R. H. Lehmberg, Phys. Rev., preceding paper, Phys. Rev. A 2, 883 (1970).

[2]M. J. Stephen, J. Chem. Phys. 40, 669 (1964).

[3]D. A. Hutchinson and H. F. Hameka, J. Chem. Phys. 41, 2006 (1964).

[4]V. L. Lyuboshitz, Zh. Eksperim. i Teor. Fiz. 52, 926 (1967) [Soviet Phys. JETP 25, 612 (1967)]; 53, 1630 (1967) [26, 937 (1968)].

[5]Y. C. Lee and D. L. Lin, Phys. Rev. 183, 147; 183, 150 (1969).

[6]V. Ernst and P. Stehle, Phys. Rev. 176, 1456 (1968).

[7]R. H. Dicke, Phys. Rev. 93, 99 (1954); in *Proceedings of the Third International Congress of Quantum Electronics*, edited by P. Grivet and N. Bloembergen (Columbia U. P., New York, 1964), p. 35.

[8]F. Schwabl and W. Thirring, Ergeb. Exakt. Naturw. 36, 219 (1964).

[9]E. R. Buley and F. W. Cummings, Phys. Rev. 134, A1454 (1964).

[10]J. H. Eberly and N. E. Rehler, Phys. Letters 29A, 142 (1969).

[11]A. Abragam, *The Principles of Nuclear Magnetism* (Oxford U. P., London, 1961), p. 103.

[12]R. H. Lehmberg, Phys. Rev. 181, 32 (1969).

[13]One can also obtain separable equations with other combinations of $Q_{\alpha\beta}$, such as $Q_{11}+Q_{22}\pm Q_{12}\;\pm Q_{21}$ and $Q_{11}-Q_{22}\pm Q_{12}\mp Q_{21}$; however, their physical interpretation is not as clear. For $N>2$, the $\rho_{r'm,rm}(t)$ matrices do not yield readily separable equations.

[14]Note that Ω_{12} need not be positive; e.g., see Fig. 1 in I.

[15]M. Dillard and H. R. Robl, Phys. Rev. 184, 312 (1969)

[16]Strictly speaking, susceptibility is a well-defined quantity only if the memory time of the atomic phase is short in comparison to the time required for large changes in the level populations or pulse amplitude. Its application here is entirely heuristic.

[17]J. Macek, Phys. Rev. Letters 23, 1 (1969); P. A. Franken, Phys. Rev. 121, 508 (1961).

[18]M. Lax, Phys. Rev. 129, 2342 (1963); M. Lax and W. H. Louisell, IEEE J. Quantum Electron. QE-3, 47 (1967).