Radiation from an N-Atom System. I. General Formalism

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We consider the radiation from a system of N identical two-level atoms coupled to a continuum of quantized em modes, and possibly, to an external driving field near resonance. The atoms can be distributed over a region large in comparison to the resonant wavelength, but smaller than the spontaneous pulse length. Radiation rates and correlation functions are expressed in terms of expectation values of time-dependent atomic operators, which are shown to satisfy coupled first-order differential equations involving similar atomic operators and the *initial* radiation operators. The corresponding equations for the expectation values simplify considerably if no driving field is present. Similar results are derived for a model in which each atom is replaced by a harmonic oscillator.

I. INTRODUCTION

The development of ultrashort light-pulse techniques has led to renewed interest in a number of spontaneous radiation effects that are related to atomic coherence. Examples of such effects include superradiance and ray-forming properties,[1-5] photon trapping,[1,6,7] and coherent spectral line broadening.[6-9]

An appropriate model by which to study these phenomena consists of N identical two-level atoms coupled to a continuum of quantized em modes, and possibly to an external driving field near resonance. This model has recently been treated by a nearly exact operator formalism for the case in which the atoms are confined to a region small in comparison to the transition wavelength.[10,11] The present formalism removes this restriction, and also takes into account the frequency shifts due to em coupling between the atoms.[6,8] The only major limitation on the size of the system, expressed by condition (13), requires that the atoms be confined to a region small in comparison to the spontaneous pulse length. Although this precludes its application to macroscopic lasing materials, the formalism can still apply to multiatom systems extending over many wavelengths, and therefore capable of developing pronounced directional effects.

The methods and results presented here differ considerably from those of earlier treatments; in particular, the emphasis is upon time-dependent decay rates, intensity patterns, and correlation functions, rather than upon quantities such as the probability amplitude for finding a particular set of photon states. Although these admittedly contain less information than probability amplitudes, they are usually the quantities of most direct physical interest, and are easier to calculate.

In Sec. II, we derive a general equation of motion for an arbitrary atomic operator [Eq. (23)], and compare this with earlier results.[10,11] Expression (23) contains only *initial* radiation operators, ordered so that they can be eliminated if vacuum expectation values are taken; hence, if no photons are present initially, the equations for the expectation values simplify considerably [Eq. (29)]. Analogous results are presented for a model in which each atom is replaced by an equivalent harmonic oscillator.

In Sec. III, the average radiation intensity and correlation function are defined in terms of expectation values of appropriate atomic operators [Eqs. (37) and (40)]. They can therefore be evaluated if solutions to (23) or (29) can be found.

In Sec. IV, we discuss the size limitation mentioned above, and estimate the decay time for a many-atom system.

This formalism will be applied in the following paper to investigate the spontaneous emission properties of a two-atom and two-oscillator system.

II. EQUATIONS OF MOTION

Consider a collection of N identical nonoverlapping atoms, at positions $\vec{r}_1, \ldots, \vec{r}_N$, coupled to a quantized multimode em field. Each atom A_α is assumed to have only the two states $|\pm\rangle_\alpha$, separated by energy $\epsilon_\alpha = \epsilon = \epsilon_+ - \epsilon_-$. Using the dipole approximation, one can write for the Hamiltonian[12] (with $\hbar \equiv 1$)

$$H = \sum_{\alpha=1}^{N} \epsilon \sigma_\alpha^\dagger \sigma_\alpha + (1/8\pi)\int (|\vec{E}|^2 + |\vec{B}|^2) d^3r - \sum_{\alpha=1}^{N} \vec{E}(\vec{r}_\alpha)\cdot\vec{p}\,(\sigma_\alpha + \sigma_\alpha^\dagger), \quad (1)$$

where $\sigma_\alpha \equiv |-\rangle_{\alpha\alpha}\langle +|$ is the lowering operator for A_α, and $\vec{p} \equiv \langle +|e\vec{x}|-\rangle$ is the dipole matrix element[13] (assumed real). The assumption of no atomic overlap implies that $[\sigma_\alpha, \sigma_\beta^\dagger] = 0$ for $\alpha \neq \beta$.

Field operators $\vec{E}(\vec{r})$ and $\vec{B}(\vec{r})$ may be conveniently expanded in terms of transverse plane-wave modes; i.e.,

$$\begin{Bmatrix} \vec{E}(\vec{r}) \\ \vec{B}(\vec{r}) \end{Bmatrix} = \sum_q \left(\frac{2\pi\omega_q}{V}\right)^{1/2} \begin{Bmatrix} \vec{e}_q \\ \hat{k}\times\vec{e}_q \end{Bmatrix} e^{i\vec{k}_q\cdot\vec{r}} a_q + \text{H.c.}, \tag{2}$$

where V is the normalization volume, $\vec{e}_q$ the unit polarization vector ($\hat{k}_q\cdot\vec{e}_q = 0$), and a_q the annihilation operator for the qth mode ($[a_q, a_{q'}^\dagger] = \delta_{qq'}$). Substituting (2) into (1), and discarding the zero-point energy, we finally obtain

$$H = \sum_{\alpha=1}^{N} \epsilon\sigma_\alpha^\dagger\sigma_\alpha + \sum_q \omega_q a_q^\dagger a_q - \sum_{\alpha=1}^{N}\sum_q K_q(e^{i\vec{k}_q\cdot\vec{r}_\alpha} a_q + e^{-i\vec{k}_q\cdot\vec{r}_\alpha} a_q^\dagger) s_\alpha, \tag{3}$$

where

$$s_\alpha \equiv \sigma_\alpha + \sigma_\alpha^\dagger \quad , \tag{4}$$

$$K_q \equiv (2\pi\omega_q/V)^{1/2}\vec{e}_q\cdot\vec{p} \quad . \tag{5}$$

The radiation operators satisfy

$$\dot{a}_q = i[H, a_q] = -i\omega_q a_q + i\sum_\alpha K_q s_\alpha e^{-i\vec{k}_q\cdot\vec{r}_\alpha} \quad ; \tag{6}$$

hence,

$$a_q(t) = a_q(0)e^{-i\omega_q t} + i\sum_\alpha K_\alpha e^{-i\vec{k}_q\cdot\vec{r}_\alpha} \times \int_0^t dt' s_\alpha(t') e^{-i\omega_q(t-t')} \quad . \tag{7}$$

If $Q(t)$ is an arbitrary combination of atomic operators, then {with the definition $[A, B]_t \equiv [A(t), B(t)]$}

$$\dot{Q}(t) = i\epsilon\sum_\alpha [\sigma_\alpha^\dagger\sigma_\alpha, Q]_t - i\sum_\alpha \sum_q K_q \times \{e^{i\vec{k}_q\cdot\vec{r}_\alpha}[s_\alpha, Q]_t a_q(t) - e^{-i\vec{k}_q\cdot\vec{r}_\alpha} a_q^\dagger(t)[Q, s_\alpha]_t\} . \tag{8}$$

Consider the first term within the curly brackets. According to Eq. (7), its contribution to $\dot{Q}(t)$ can be written as

$$-i\sum_\alpha [s_\alpha, Q]_t \vec{E}_+^{(0)}(\vec{r}_\alpha, t)\cdot\vec{p} + \sum_{\alpha,\beta} K_q^2 e^{i\vec{k}_q\cdot\vec{r}_{\alpha\beta}} \times \int_0^t dt' e^{-i\omega_q(t-t')}[s_\alpha, Q]_t s_\beta(t') , \tag{9}$$

where

$$\vec{r}_{\alpha\beta} \equiv \vec{r}_\alpha - \vec{r}_\beta ,$$

$$\vec{E}_+^{(0)}(\vec{r}, t) \equiv \sum_q \left(\frac{2\pi\omega_q}{V}\right)^{1/2} \vec{e}_q a_q(0) e^{i(\vec{k}_q\cdot\vec{r} - \omega_q t)} . \tag{10}$$

To evaluate the second set of terms in (9), we allow $V \to \infty$ and $\vec{k}_q \to \vec{k}$ so that

$$\frac{1}{V}\sum_q \to \left(\frac{1}{2\pi c}\right)^3 \int_0^\infty \omega^2 d\omega \oint d\Omega_{\hat{k}} \sum_{\eta_{\hat{k}}=1}^{2} , \tag{11}$$

where $\hat{k} = \vec{k}/k$, and $\eta_{\hat{k}}$ specifies the orthogonal polarizations of the $\vec{k}$th mode. Substituting (5) and (11) into (9), then performing the $\eta_{\hat{k}}$ summation, we obtain

$$-i\sum_\alpha [s_\alpha, Q]_t \vec{E}_+^{(0)}(\vec{r}_\alpha, t)\cdot\vec{p} + (p^2/4\pi^2c^3) \times \sum_{\alpha,\beta} \oint d\Omega_{\hat{k}}[1 - (\hat{k}\cdot\hat{p})^2] \int_0^t dt' \int_0^\infty d\omega\, \omega^3 \times \exp[i\omega(t' - t + \hat{k}\cdot\vec{r}_{\alpha\beta}/c)][s_\alpha, Q]_t s_\beta(t') \quad . \tag{12}$$

Although the ω integral extends to ∞, the dipole approximation begins to break down as $\omega \to \omega_B \equiv c/a_B$, where a_B is the Bohr radius. In a more exact treatment, one would replace $\vec{p}$ by some function $\vec{p}(\omega)$ that decreases exponentially for $\omega > \omega_B$; i.e., the integral effectively cuts off around $\omega \simeq \omega_B$. For a given pair of atoms (α, β), the most important values of t' therefore lie within a region on the order of ω_B^{-1} around $t - \hat{k}\cdot\vec{r}_{\alpha\beta}/c$. Since $\omega_B \gg \epsilon$, one can certainly replace $\sigma_\beta(t')$ by

$$\sigma_\beta(t - \hat{k}\cdot\vec{r}_{\alpha\beta}/c)\exp[-i\epsilon(t' - t + \hat{k}\cdot\vec{r}_{\alpha\beta}/c)] + \text{H.c.}$$

in expression (12). We now impose the restriction that the time required for a light signal to cross the system be small in comparison to the time Δt required for appreciable (secular) changes in the atomic levels; i.e.,

$$(r_{\alpha\beta})_{\max} \ll c\Delta t \quad . \tag{13}$$

This assumption has been used explicitly in other quantum treatments of large systems,[1,4] and will be discussed in more detail in Sec. IV. If it applies, we have

$$s_\beta(t') \simeq \sigma_\beta(t)e^{-i\epsilon(t'-t)} + \sigma_\beta^\dagger(t)e^{i\epsilon(t'-t)} \quad . \tag{14}$$

All operators in (12) are now evaluated at times t; hence, the t' integral can be performed, yielding

$$-i\sum_\alpha [s_\alpha, Q]_t \vec{E}_+^{(0)}(\vec{r}_\alpha, t)\cdot\vec{p} + (p^2/4\pi^2c^3)\sum_{\alpha,\beta}[s_\alpha, Q]_t \times \oint d\Omega_{\hat{k}}[1 - (\hat{k}\cdot\hat{p})^2]\oint_0^\infty \omega^3 \exp(i\omega\hat{k}\cdot\vec{r}_{\alpha\beta}/c) \times \left\{\left[\frac{1 - \cos(\omega - \epsilon)t}{i(\omega - \epsilon)} + \frac{\sin(\omega - \epsilon)t}{\omega - \epsilon}\right]\sigma_\beta(t) + \left[\frac{1 - \cos(\omega + \epsilon)t}{i(\omega + \epsilon)} + \frac{\sin(\omega + \epsilon)t}{\omega + \epsilon}\right]\sigma_\beta^\dagger(t)\right\} \quad . \tag{15}$$

We are mainly interested in times

$$\epsilon t \gg 1, \quad ct \gg (r_{\alpha\beta})_{\max} \tag{16}$$

(however, t may still be $< \Delta t$), and under these conditions the expression in the square brackets can be replaced by

$$-iP(\omega \pm \epsilon)^{-1} + \pi\delta(\omega \pm \epsilon) \quad , \tag{17}$$

where P indicates a principal value.

Substituting this result into (15), integrating over Ω_k and ω, then discarding the high-frequency operators {such as $\sigma_\alpha(t)\sigma_\beta(t)$ and $[\sigma_\alpha, Q]_t \vec{E}_+^{(0)}(\vec{r}_\alpha, t) \cdot \vec{p}$}, we obtain

$$-\sum_\alpha [\sigma_\alpha^\dagger, Q]_t \vec{E}_+^{(0)}(\vec{r}_\alpha, t)\cdot\vec{p} + \sum_{\alpha,\beta} \{(-i\Omega_{\alpha\beta}^- + \tfrac{1}{2}\gamma_{\alpha\beta}) \times [\sigma_\alpha^\dagger, Q]_t \sigma_\beta(t) - i\Omega_{\alpha\beta}^+ [\sigma_\alpha, Q]_t \sigma_\beta^\dagger(t)\} \quad . \tag{18}$$

In terms of the normal decay rate of an isolated atom

$$\gamma = \tfrac{4}{3} p^2 \kappa^3, \quad \kappa \equiv \epsilon/c \tag{19}$$

the constants can be written

$$\gamma_{\alpha\beta} = \gamma_{\beta\alpha} = \gamma F_{\alpha\beta}(\kappa r_{\alpha\beta}), \quad \gamma_{\alpha\alpha} = \gamma \tag{20}$$

$$\Omega_{\alpha\beta}^\pm \sim \frac{\gamma}{\kappa^3} P \int_0^\infty \frac{dk}{2\pi} \frac{k^3 F_{\alpha\beta}(kr_{\alpha\beta})}{k \pm \kappa} \quad , \tag{21}$$

where

$$F_{\alpha\beta}(\xi) \equiv \tfrac{3}{2}\{[1 - (\hat{p}\cdot\hat{r}_{\alpha\beta})^2] \sin\xi/\xi + [1 - 3(\hat{p}\cdot\hat{r}_{\alpha\beta})^2](\cos\xi/\xi^2 - \sin\xi/\xi^3)\} \, . \tag{22}$$

The second term in the curly brackets of (8) can be written immediately by taking the Hermitean adjoint of the first one, then replacing $Q^\dagger$ by Q. Applying this procedure to (18), we finally obtain

$$\dot{Q} = i(\epsilon - \Omega)\sum_\alpha [\sigma_\alpha^\dagger \sigma_\alpha, Q] + (i\epsilon/c)\vec{p}\cdot\sum_\alpha \{[s_\alpha, Q]\vec{E}_+^{(0)}(\vec{r}_\alpha, t) - \vec{E}_+^{(0)\dagger}(\vec{r}_\alpha, t)[Q, s_\alpha]\} + \sum_{\alpha\neq\beta} i\Omega_{\alpha\beta}[\sigma_\alpha^\dagger \sigma_\beta, Q] + \sum_{\alpha,\beta} \gamma_{\alpha\beta}[\sigma_\alpha^\dagger Q \sigma_\beta - \tfrac{1}{2}(\sigma_\alpha^\dagger \sigma_\beta Q + Q\sigma_\alpha^\dagger \sigma_\beta)] \quad , \tag{23}$$

where all atomic operators are evaluated at time t, and the frequency shifts are defined as

$$\Omega \equiv \Omega_{\alpha\alpha}^- - \Omega_{\alpha\alpha}^+ \quad , \tag{24a}$$

$$\Omega_{\alpha\beta} \equiv -(\Omega_{\alpha\beta}^- + \Omega_{\alpha\beta}^+), \quad \alpha \neq \beta \quad . \tag{24b}$$

The form of (23) depends upon the ordering of radiation and atomic operators in Eq. (8). If they had been placed in antinormal order [e.g., $a_q(t)$ to the left of $\sigma^\dagger(t)$], then (23) would contain terms such as

$$\vec{E}_+^{(0)}(\vec{r}_\alpha, t)[\sigma_\alpha^\dagger, Q]_t \quad ,$$

$$\gamma_{\alpha\beta}[\sigma_\alpha Q \sigma_\beta^\dagger - \tfrac{1}{2}(\sigma_\alpha \sigma_\beta^\dagger Q + Q\sigma_\alpha \sigma_\beta^\dagger)],$$

instead of the normal-ordered combinations. Although this is formally correct, it is not very useful for obtaining equations for the expectation values with initial radiation states [e.g., expression (29)] because $\vec{E}_+^{(0)}(\vec{r}_\alpha, t)$ does not commute with the atomic operators at time t.

From Eqs. (21) and (22), it follows that Ω would diverge logarithmically were it not for the frequency cutoff around c/a_B. Since it always appears in the combination $\epsilon - \Omega$, it makes no contribution to the phenomena of interest here, and will henceforth be absorbed into ϵ.

To calculate $\Omega_{\alpha\beta}$, we use (21), (22a), and the identity $F_{\alpha\beta}(-\xi) = F_{\alpha\beta}(\xi)$ to write

$$\Omega_{\alpha\beta} = -\frac{\gamma}{(\kappa r_{\alpha\beta})^3} P \int_{-\infty}^\infty \frac{d\xi}{2\pi} \frac{\xi^3 F_{\alpha\beta}(\xi)}{\xi - \kappa r_{\alpha\beta}} \quad .$$

Evaluating this by contour methods, we obtain

$$\Omega_{\alpha\beta} = \Omega_{\beta\alpha} = \gamma G_{\alpha\beta}(\kappa r_{\alpha\beta}) \quad , \tag{25}$$

where

$$G_{\alpha\beta}(\xi) = \tfrac{3}{4}\{-[1 - (\hat{p}\cdot\hat{r}_{\alpha\beta})^2](\cos\xi)/\xi + [1 - 3(\hat{p}\cdot\hat{r}_{\alpha\beta})^2][(\sin\xi)/\xi^2 + (\cos\xi)/\xi^3]\} \, . \tag{26}$$

This is the interaction energy between the αth and βth atoms, and agrees with the result derived by Stephen.[6]

In Fig. 1, $G_{\alpha\beta}(\kappa r_{\alpha\beta}) = \Omega_{\alpha\beta}/\gamma$ has been plotted, along with $F_{\alpha\beta}(\kappa r_{\alpha\beta}) = \gamma_{\alpha\beta}/\gamma$, for the cases where $\hat{p}$ is parallel and perpendicular to $\hat{r}_{\alpha\beta}$. For $\kappa r_{\alpha\beta} > \pi$, we observe that $\gamma_{\alpha\beta}$ and $\Omega_{\alpha\beta}$ tend to be larger in the perpendicular case. This is understandable because the interaction at $\kappa r_{\alpha\beta} > \pi$ comes primarily from dipole radiation rather than from electrostatic or induction fields.[7] (The semiclassical interpretation of $\gamma_{\alpha\beta}$ and $\Omega_{\alpha\beta}$ is discussed by Lyuboshitz.[8])

If $\kappa r_{\alpha\beta} \ll 1$, then $\gamma_{\alpha\beta} \simeq \gamma$, while $\Omega_{\alpha\beta}$ becomes large and strongly dependent upon $r_{\alpha\beta}$. In the case where $\kappa(r_{\alpha\beta})_{\max} \ll 1$, Eq. (23) therefore reduces to

$$\dot{Q} = i\epsilon\sum_\alpha [\sigma_\alpha^\dagger \sigma_\alpha, Q] - i\vec{p}\cdot\{[R_+, Q]\vec{E}_+^{(0)}(t) - \vec{E}_+^{(0)\dagger}(t)[Q, R_-]\} + \sum_{\alpha\neq\beta} i\Omega_{\alpha\beta}[\sigma_\alpha^\dagger \sigma_\beta, Q] + \gamma[R_+ Q R_- - \tfrac{1}{2}(R_+ R_- Q + Q R_+ R_-)] \quad , \tag{27}$$

where

$$R_- \equiv \sum_\alpha \sigma_\alpha, \quad R_+ \equiv \sum_\alpha \sigma_\alpha^\dagger \, . \tag{28}$$

Except for the $\Omega_{\alpha\beta}$ terms, this yields the same operator equations of motion as those derived in earlier work[10,11]; however, in the following paper, it will be shown that $\Omega_{\alpha\beta}$ terms can, under some circumstances, have important effects on the radi-

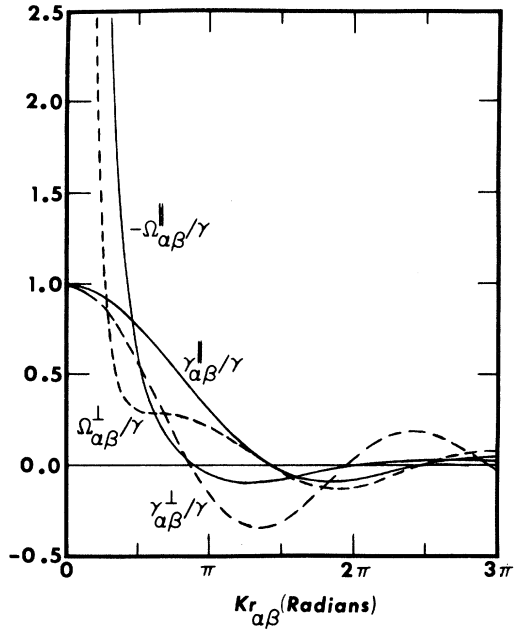

FIG. 1. Dependence of the damping constants γ_{12} and frequency shifts Ω_{12} on interatomic spacing r_{21} for dipole matrix elements $\vec{p}$ parallel and perpendicular to $\vec{r}_{21}$. Here, $\kappa = \epsilon/c = 2\pi/\lambda$, and γ is the damping constant of a single atom.

ation properties of a system.

Returning to the general expression (23), we note that for the case where the radiation is initially in the vacuum state $|0\rangle$, one can take vacuum expectation values to obtain the simplified form

$$\langle \dot{Q} \rangle_0 = i\epsilon \sum_\alpha \langle [\sigma_\alpha^\dagger \sigma_\alpha, Q] \rangle_0 + \sum_{\alpha \neq \beta} i\,\Omega_{\alpha\beta} \langle [\sigma_\alpha^\dagger \sigma_\beta, Q] \rangle_0 + \sum_{\alpha,\beta} \gamma_{\alpha\beta} \langle \sigma_\alpha^\dagger Q \sigma_\beta - \tfrac{1}{2}(\sigma_\alpha^\dagger \sigma_\beta Q + Q \sigma_\alpha^\dagger \sigma_\beta) \rangle_0 , \tag{29}$$

where $\langle\ \rangle_0 \equiv \langle 0|\ |0\rangle$. This expression will be the fundamental equation of motion in the theory of spontaneous emission from an N-atom system.

Consider now the case where each atom A_α is replaced by a harmonic oscillator O_α of frequency ϵ and energy states $|n\rangle_\alpha$. If $\vec{p}$ is now defined as $\langle n=1|\, e\vec{x}\,|n=0\rangle$, then $\langle n+1|\, e\vec{x}\,|n\rangle = (n+1)^{1/2}\vec{p}$. The radiation interaction is therefore identical to the last term in expression (3) with s_α replaced by $b_\alpha + b_\alpha^\dagger$, where b_α is the lowering operator ($b_\alpha |n\rangle_\alpha = n^{1/2}|n-1\rangle_\alpha$). If $Q_{os}(t)$ is an arbitrary combination of oscillator operators, then we again obtain Eq. (23) (by the same derivation), with $Q \to Q_{os}$ and $\sigma_\alpha \to b_\alpha$. The only difference is that Ω is now defined as $\Omega^-_{\alpha\alpha} + \Omega^+_{\alpha\alpha}$.

III. RADIATION PROPERTIES

We now derive general expressions for the radiation rates and spectral properties of the far field; in particular, let $\vec{R}$ be an observation point (referred to an arbitrary origin within the atomic system), and assume that $|\vec{R} - \vec{r}_\alpha| \equiv R_\alpha$ satisfies

$$\kappa R_\alpha \gg 1 \tag{30}$$

for all α.

The quantities of interest can be obtained from the correlation functions

$$f_{\hat{R}}(t,t') \equiv (R^2 c/2\pi\epsilon)\langle \vec{E}_+^\dagger(\vec{R},t) \circ \vec{E}_+(\vec{R},t')\rangle , \tag{31}$$

$$f(t,t') \equiv \oint d\Omega_{\hat{R}} f_{\hat{R}}(t,t') , \tag{32}$$

where the average is taken over initial states of the entire system, and $\vec{E}_+(\vec{R},t)$ is the positive frequency field operator

$$\vec{E}_+(\vec{R},t) = \sum_q (2\pi\omega_q/V)^{1/2} \vec{e}_q a_q(t) e^{i\vec{k}_q \cdot \vec{R}} . \tag{33}$$

For $t' = t$,

$$f_{\hat{R}}(t,t) = \dot{n}_{\hat{R}}(t) \equiv W_{\hat{R}}(t) \tag{34}$$

is the average photon emission rate into solid angle $d\Omega_{\hat{R}}$, and

$$f(t,t) = \dot{n}(t) \equiv W(t) \tag{35}$$

is the total radiation rate. We note that although $W_{\hat{R}}(t)$ does not correspond exactly to the classical expression (i.e., with real fields), it is the actual intensity that would be measured by an ordinary photodetector or similar device.[14]

Substituting Eq. (7) into (33), then applying conditions (13), (16), and (30), we obtain, by arguments similar to those used before (see the Appendix),

$$\vec{E}_+(\vec{R},t_R) = \vec{E}_+^{(0)}(\vec{R},t_R) + \frac{\epsilon^2}{c^2} \sum_\alpha \frac{\vec{p} - \hat{R}_\alpha \hat{R}_\alpha \cdot \vec{p}}{R_\alpha} \times \exp[i\kappa(R_\alpha - R)]\,\sigma_\alpha(t) , \tag{36}$$

where $t_R = t + R/c$, and $\vec{E}_+^{(0)}(\vec{R},t)$ is defined by (10).

The main interest here is in cases where the initial radiation is either confined to a narrow beam or absent entirely. If $\vec{R}$ lies outside any such beam, and if $R \gg (r_{\alpha\beta})_{\max}$ (so that $R_\alpha - R \simeq -\hat{R} \cdot \vec{r}_\alpha$), then (31) becomes

$$f_{\hat{R}}(t_R, t_R') = w_{\hat{R}}^{(1)} \sum_{\alpha,\beta} e^{i\kappa \hat{R} \circ \vec{r}_{\alpha\beta}} \langle \sigma_\alpha^\dagger(t)\sigma_\beta(t')\rangle , \tag{37a}$$

$$f_{\hat{R}}(t_R, t_R') = w_{\hat{R}}^{(1)} \langle R_{\vec{k}}^\dagger(t_R) R_{\vec{k}}(t_R')\rangle , \tag{37b}$$

where

$$\vec{k} \equiv \kappa \hat{R}, \tag{38}$$

$$R_{\vec{k}} \equiv \sum_\beta \sigma_\beta e^{-i\vec{k} \cdot \vec{r}_\beta} ,$$

and

$$w_{\hat{R}}^{(1)} \equiv \frac{3\gamma}{8\pi}[1 - (\hat{R} \circ \hat{p})^2] \tag{39}$$

is the initial radiation rate into $d\Omega_{\hat{R}}$ for a single excited atom. Expression (37b) generalizes a result first derived by Dicke[1] for the initial radiation rate.

Similarly, Eq. (32) becomes

$$f(t_R, t_R') = f^{(0)}(t_R, t_R') + \sum_{\alpha,\beta} \gamma_{\alpha\beta} \langle \sigma_\alpha^\dagger(t) \sigma_\beta(t') \rangle , \quad (40)$$

where

$$f^{(0)}(t_R, t_R') \equiv (R^2 c/2\pi\epsilon) \oint d\Omega_{\hat{R}} \times \langle \vec{E}_+^{(0)\dagger}(\vec{R}, t_R) \cdot \vec{E}_+^{(0)}(\vec{R}, t_R') \rangle , \quad (41)$$

and the interference between the incident and scattered radiation is negligible for R sufficiently large. In these expressions the factor R/c appears in both t_R and t_R'; thus, it is irrelevant for our purposes and will be ignored henceforth.

As a check on the methods used here, we note that $W_{\hat{R}}(t)$ can be calculated directly from the relation

$$W_{\hat{R}}(t) = \lim_{\Delta\Omega_{\hat{R}} \to 0} (1/\Delta\Omega_{\hat{R}}) \sum_{q(\Delta\Omega_{\hat{R}})} a_q^\dagger(t) a_q(t) + \text{H. c.}, \quad (42)$$

where the sum extends only over q states lying within $\Delta\Omega_{\hat{R}}$ around the $\hat{R}$ direction. Substituting Eqs. (6) and (7) into (42), using the relation

$$\lim_{\Delta\Omega_{\hat{R}} \to 0} \frac{1}{\Delta\Omega_{\hat{R}} V} \sum_{q(\Delta\Omega_{\hat{R}})} \xrightarrow[V \to \infty]{} \left(\frac{1}{2\pi c}\right)^3 \int_0^\infty \omega^2 d\omega \sum_{\eta}^{2} \hat{R} = 1 ,$$

and following the same procedure as the one used to derive (23), we recover (37) with $t' = t$.

For intensity calculations ($t' = t$), one can obtain differential equations for the factors $\langle \sigma_\alpha^\dagger(t) \sigma_\beta(t) \rangle$ by taking the expectation values of both sides of Eq. (23). This is considerably simpler than dealing with $\sigma_\beta(t)$ or $\sigma_\alpha^\dagger(t)\sigma_\beta(t)$ operators; e.g., in the case where no photons are present initially, one can deal with Eq. (29).

If $t' = 0$, then $\langle \sigma_\alpha^\dagger(t) \sigma_\beta(0) \rangle$ can be obtained from the reduced operators $\langle \text{rad} | \sigma_\alpha^\dagger(t) | \text{rad} \rangle$, where $| \text{rad} \rangle$ is the initial radiation state. Again, there is a considerable simplification, especially if no photons are present initially.

In the general case, $t > t' > 0$ or $t' > t > 0$, one is obliged either to solve for states such as $\sigma_\beta(t') | \text{rad} \rangle$, or to approximate $\langle \sigma_\alpha^\dagger(t) \sigma_\beta(t') \rangle$ by using the fluctuation-regression theorem.[15]

Finally, we note that expressions (36)–(41) apply equally well to the harmonic-oscillator model, the only difference being that all $\sigma_\alpha(t)$ are replaced by $b_\alpha(t)$ operators.

IV. DISCUSSION

The most important physical restriction in this paper is condition (13). For a system driven by a sufficiently strong resonant pulse, Δt is determined by the field amplitude and pulsewidth, and the problem may be adequately treated by semiclassical theory. In the absence of external radiation, we obtain $\Delta t \simeq \tau$, where τ is the duration of the spontaneous pulse; hence, if only a few atoms are present, we then have

$$\Delta t \simeq \gamma^{-1} . \quad (43)$$

One can obtain an approximate expression for Δt in the case where $N \gg 1$ from the results of Eberly and Rehler[5]:

$$\tau = 2/(\gamma \mu' N) , \quad (44)$$

where

$$\mu' \equiv \oint d\Omega_{\hat{R}} \frac{w_{\hat{R}}^{(1)}}{\gamma} \left| \frac{1}{N} \sum_\alpha \exp[i(\kappa \hat{R} - \vec{k}_1) \cdot \vec{r}_\alpha] \right|^2 , \quad (45)$$

$\vec{k}_1$ is the wave vector of a short pulse used to excite the atoms initially, and $w_{\hat{R}}^{(1)}$ is the initial radiation rate into $d\Omega_{\hat{R}}$ for a single atom. In the case of π-pulse excitation, $w_{\hat{R}}^{(1)}$ is given by Eq. (39).

To estimate μ' for a large system, we consider a model in which the atoms are distributed at random throughout a spherical region of diameter D; i.e.,

$$\frac{1}{N} \sum_\alpha \simeq \frac{6}{\pi D^3} \int_0^{D/2} r^2 dr \oint d\Omega_{\hat{R}} .$$

Substituting this into (45), taking $\vec{k}_1 = \kappa \hat{k}_1$, and approximating $w_{\hat{R}}^{(1)}$ by $\gamma/4\pi$, we obtain

$$\mu' \simeq 18 I/(\kappa D)^2 ,$$

where

$$I \equiv \int_0^{\kappa D} \frac{d\theta}{\theta^5} (\sin\theta - \theta \cos\theta)^2 \xrightarrow[\kappa D \to \infty]{} \frac{1}{4} .$$

Hence, we have

$$\Delta t \simeq \simeq \tau\, 10\pi\lambda/(n_\lambda D\gamma),$$

where

$$\lambda = 2\pi/\kappa \quad \text{and} \quad n_\lambda \equiv 6N\lambda^3/(\pi D^3)$$

is the average number of atoms per cubic wavelength. For a numerical example, we choose $\lambda = 10^{-4}$ cm, $D = 10^{-2}$ cm, and $n_\lambda = 10^2$; then we obtain $\gamma \simeq 10^7$ sec^{-1} for a dipole transition and $\Delta t \simeq 10^{-10}\pi$ sec. Since $(r_{\alpha\beta})_{\max} = D$, we have $(r_{\alpha\beta})_{\max}/c \simeq 3.3 \times 10^{-13}$ sec.

APPENDIX: DERIVATION OF EQ. (36)

Substituting expression (7) into (33) and using definition (10), we obtain

$$\vec{E}_+(\vec{R}, t_R) = \vec{E}_+^{(0)}(\vec{R}, t_R) + 2\pi i \sum_\alpha (1/V) \sum_q \omega_q \vec{e}_q \vec{e}_q \cdot \vec{p} \times \exp[i \vec{k}_q \cdot (\vec{R} - \vec{r}_\alpha)] \int_0^{t_R} dt' \exp[-i\omega_q(t_R - t')] s_\alpha(t') , \quad (A1)$$

where $t_R = t + R/c$. If relation (11) is applied, and the $\eta_{\hat{k}}$ summation carried out, the second term becomes

$$(i/4\pi^2c^3)\sum_\alpha \int_0^\infty \omega^3 d\omega \oint d\Omega_{\hat{k}} (\vec{p} - \hat{k}\hat{k}\cdot\vec{p}) \times e^{i\omega\hat{k}\circ\vec{R}_\alpha/c} \int_0^{t_R} dt' \exp[-i\omega(t_R - t')] s_\alpha(t'), \quad \text{(A2)}$$

where $\vec{R}_\alpha \equiv \vec{R} - \vec{r}_\alpha$.

The largest contributions to the field come from $\omega \simeq \epsilon$. Thus, if $\epsilon R_\alpha/c \gg 1$, then $\exp(i\omega\hat{k}\circ\vec{R}_\alpha/c)$ will oscillate rapidly over a range of $\hat{R}_\alpha \circ \hat{k}$ in which $\vec{p} - \hat{k}\hat{k}\circ\vec{p}$ remains essentially constant. The only important contributions come from those directions around $\hat{k} = \pm\hat{R}_\alpha$, where the phase $\pm\omega R_\alpha/c$ is stationary; hence, one can replace $\vec{p} - \hat{k}\hat{k}\cdot\vec{p}$ by $\vec{p} - \hat{R}_\alpha\hat{R}_\alpha\circ\vec{p}$, and remove it from the $\Omega_{\hat{k}}$ integral. The result is

$$\frac{1}{2\pi c^2}\sum_\alpha \frac{\vec{p} - \hat{R}_\alpha\hat{R}_\alpha\circ\vec{p}}{R_\alpha} \int_0^\infty \omega^2 d\omega \int_0^{t_R} dt' s_\alpha(t') \times e^{i\omega(t' - t - R/c)}\left[e^{i\omega R_\alpha/c} - e^{-i\omega R_\alpha/c}\right]. \quad \text{(A3)}$$

It is necessary to consider only the $e^{i\omega R_\alpha/c}$ term. Since $|R_\alpha - R| \le (r_{\alpha\beta})_{\max}$ if $\vec{R}$ lies within the atomic system, the same arguments that led to (14) also apply here. Thus, substituting (14) into (A3), then evaluating the t' integral, one is led to terms of the form

$$\frac{\exp[i(\omega \pm \epsilon)R_\alpha/c] - \exp\{-i(\omega \pm \epsilon)[t - (R_\alpha - R)/c]\}}{i(\omega \pm \epsilon)} \times e^{\mp i\epsilon(R_\alpha - R)/c}. \quad \text{(A4)}$$

We have already assumed that $\epsilon R_\alpha/c \gg 1$, and since $|R_\alpha - R| \le (r_{\alpha\beta})_{\max}$, conditions (16) lead to $\epsilon[t - (R - R_\alpha)/c] \gg 1$. Therefore, expression (A4) can be approximated by

$$2\pi\delta(\omega \pm \epsilon)\exp[i\epsilon(R_\alpha - R)/c],$$

and (A3) reduces to

$$\left(\frac{\epsilon}{c}\right)^2 \sum_\alpha \frac{\vec{p} - \hat{R}_\alpha\hat{R}_\alpha\cdot\vec{p}}{R_\alpha} \times \exp[i\kappa(R_\alpha - R)]\sigma_\alpha(t), \quad \text{(A5)}$$

where $\kappa \equiv \epsilon/c$. Adding the $\vec{E}_+^{(0)}(\vec{R}, t_R)$ to (A5), we obtain Eq. (36).

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