Superradiance in an inhomogeneously broadened atomic system*

R. Jodoin and L. Mandel
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627
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The problem of superradiant emission from a small atomic system is treated under conditions in which the effects of inhomogeneous broadening are important, as in a solid. The treatment is semiclassical, with the assumption that all atoms see the same electromagnetic field. A set of coupled integrodifferential equations is obtained characterizing the state of each atom of resonant frequency ω at time t. It is found that, in general, atoms of different frequencies evolve differently in time, whether or not they start from the same initial state. The effect of the inhomogeneous broadening is generally to delay the onset and to reduce the amplitude of the superradiant pulse. Some effects of inhomogeneous broadening are manifest even when the inhomogeneous lifetime is much longer than the usual superradiance time. When the atomic system starts in the ground state and is then exposed to a coherent excitation pulse, superradiant effects can appear even when the inhomogeneous lifetime is very short compared with the usual superradiance time.

I. INTRODUCTION

The theory of superradiance, or collective radiation from excited atoms, has been one of the most intensely studied subjects in recent years.[1] Following the results of the recent observations by Skribanowitz *et al.*,[2] there is now renewed interest in the possibility of observing superradiant effects in solids also. In principle, the formalisms that have been developed should apply to solids as well as to gases, except that inhomogeneous spectral broadening is often much more important in solids. It is, however, a little disappointing to find that very few of the published treatments of the superradiance problem incorporate the effect of inhomogeneous broadening. The treatments are therefore not applicable to solids in general, where the reciprocal T_2^* of the inhomogeneous linewidth may be of the same order as, or much shorter than, the superradiance time T_s. The only analyses that include some of the effects of inhomogeneous broadening appear to be those of Eberly[3] and Agarwal[4] (see also Ref. 21). Eberly made the assumption that all atoms behave identically, which applies in the limiting case $T_2^* \ll T_s$, while Agarwal made similar, though slightly less restrictive, assumptions regarding the expectation values of certain atomic operators. It is clear that a full solution of the superradiance problem, that includes the effects of inhomogeneous broadening, must be substantially more complicated than previous solutions, in that every atom belonging to a different frequency generally evolves differently in time.

In the following we present such a solution. However, while we include the effects associated with inhomogeneous broadening, we make substantial simplifications in the theory in other respects. The treatment is semiclassical, with the atoms described as two-level quantum systems and the field as a classical c-number field. In the limit of a large number of atoms, the results of this treatment generally coincide with those given by quantum electrodynamics. Secondly, we limit ourselves to a microscopic sample of linear dimensions much smaller than an optical wavelength, in order to avoid the complications associated with geometry and propagation delays. These effects have been discussed by Rehler and Eberly.[5] Thirdly, we simplify the analysis by assuming that all atoms interact very nearly with the same electromagnetic field, as in the treatment of Stroud *et al.*[6] The validity of this assumption has recently been criticized,[7] and there is no doubt that the atoms near the surface of the sample experience a different field from those near the center. Nevertheless, we believe that there is some merit in making this simplification. In the first place, there exist special geometries for which the equal-field assumption is strictly valid.[8] Secondly, for more realistic geometries, we show in the Appendix[9] that it is still a fair approximation under some conditions, that permits the complications associated with (nongeometric) inhomogeneous broadening to be emphasized. Strictly speaking, however, our treatment is valid only for the special geometries for which the equal-field assumption holds.

With the help of these assumptions we derive a set of coupled integrodifferential equations characterizing the state of each atom of resonance frequency ω at time t. The solutions show that different atoms evolve differently in time and that, moreover, there is no symmetry with respect to the midfrequency ω_0, even when all atoms are equally excited initially. The effect of the inho-

mogeneous broadening is generally to delay the onset and reduce the amplitude of the superradiant pulse, and such effects are manifest even when T_2^* is many times longer than the superradiance time T_s. We show that time-dependent frequency shifts, or chirps, appear in the superradiant pulse. They are generally associated with phase reversal when the field is close to zero.

We also consider the case in which all atoms start in the ground state and are then exposed to a coherent excitation pulse. In that case selective excitation takes place, and we find that, with a suitable excitation pulse, superradiant effects, as distinct from simple dephasing or free induction decay, appear even when the inhomogeneous lifetime T_2^* is much shorter than the usual superradiance time T_s.

II. EQUATIONS OF MOTION

We consider a set of N distinguishable two-level atoms interacting with a classical electromagnetic field, characterized by the electric vector $\vec{E}$, via an electric dipole interaction. If $\hat{\mu}_l(t)$ is the dipole moment of the lth atom, and $\vec{E}_l(t)$ is the electric field seen by this atom, then the total energy of interaction $\hat{H}_1$ of the whole system is taken to be

$$\hat{H}_1(t) = -\sum_l \hat{\mu}_l(t)\cdot\vec{E}_l(t) \ . \tag{1}$$

In this equation $\hat{\mu}_l(t)$ is a Hilbert-space operator[10] acting on the state vector $|\psi_l\rangle$ of the lth atom, but $\vec{E}_l(t)$ is a c number. We assume that the atom has no permanent dipole moment, only a transition dipole moment. We shall work in the interaction picture, where the operators associated with different atoms commute, so that the time-evolution operator for the whole system factorizes into a product of time-evolution operators for the individual atoms. As a result, an atomic product state at time $t=0$ remains a product state indefinitely. If we represent the state of the lth atom by the Bloch vector $\vec{r}_l$, then as is now well known, the Schrödinger equation of motion in the interaction picture for this atom can be expressed in the form[11]

$$\frac{d}{dt}\vec{r}_l = \vec{\Omega}_l \times \vec{r}_l \ , \tag{2}$$

where $\vec{\Omega}_l$ is a vector characterizing the interaction that is given by

$$\begin{aligned} \hbar\Omega_{l1} &= -2\vec{\mu}_l(t)\cdot\vec{E}_l(t)\cos\omega_l t \ , \\ \hbar\Omega_{l2} &= 2\vec{\mu}_l(t)\cdot\vec{E}_l(t)\sin\omega_l t \ , \\ \hbar\Omega_{l3} &= 0 \ . \end{aligned} \tag{3}$$

$\vec{\mu}_l$ is the matrix element (taken to be real) of the dipole-moment operator between the lower and upper atomic states of the lth atom, and $\hbar\omega_l$ is the energy difference between these states in the absence of any interaction.

The total field $\vec{E}_l(t)$ seen by the lth atom will have contributions from the atom's own radiation reaction field $\vec{E}_l^{(R)}(t)$, from the field due to the other atomic dipoles $\vec{E}_l^{(D)}(t)$, and from any external field $\vec{E}_l^{(E)}(t)$ that may be present. The first contribution has been given by Stroud and Jaynes,[12] and can be expressed in the form

$$\begin{aligned} \vec{E}_l^{(R)}(t) = &\frac{4\omega_l^2 K\vec{\mu}_l}{3\pi c^3}\left[x_l(t)\cos\omega_l t - y_l(t)\sin\omega_l t\right] \\ &+ \frac{2\omega_l^3\vec{\mu}_l}{3c^3}\left[x_l(t)\sin\omega_l t + y_l(t)\cos\omega_l t\right] \ , \end{aligned} \tag{4}$$

where x_l, y_l, z_l are the Cartesian components of the Bloch vector $\vec{r}_l$, and K is a cut-off frequency of order of the reciprocal of the transit time of light across the atom. $\vec{E}_l^{(D)}(t)$ is expressible in terms of the near fields of the other dipoles in the form[13]

$$\begin{aligned} \vec{E}_l^{(D)}(t) = &\sum_{k\neq l}\left(\frac{3(\vec{\mu}_l\cdot\vec{n}_{kl})\vec{n}_{kl}-\vec{\mu}_l}{d_{kl}^3} + \frac{(\vec{\mu}_l\cdot\vec{n}_{kl})\vec{n}_{kl}\omega_k^2 + \vec{\mu}_l\omega_k^2}{2c^2 d_{kl}}\right)\left[x_k(t)\cos\omega_k t - y_k(t)\sin\omega_k t\right] \\ &+ \sum_{k\neq l}\frac{2\vec{\mu}_l\omega_k^3}{3c^3}\left[x_k(t)\sin\omega_k t + y_k(t)\cos\omega_k t\right] \ , \end{aligned} \tag{5}$$

where $\vec{d}_{kl}$ is the position vector from the kth atom to the lth atom, and $\vec{n}_{kl}$ is the unit vector $\vec{d}_{kl}/d_{kl}$. We shall suppose that the external field $\vec{E}_l^{(E)}(t)$ is linearly polarized, as the field from a laser would be, and we represent it in the form

$$\vec{E}_l^{(E)}(t) = \vec{\epsilon}\,\mathcal{E}(t)\cos[\omega_0 t + \psi(t)] \ . \tag{6}$$

$\vec{\epsilon}$ is a unit polarization vector, ω_0 is some reference frequency that we take to be the most probable atomic resonance frequency (i.e., the peak frequency of the inhomogeneously broadened spectrum), and $\mathcal{E}(t)$ and $\psi(t)$ are amplitude and phase functions that vary only very slowly compared with the optical oscillations cos $\omega_0 t$. On writing

$$\vec{E}_l(t) = \vec{E}_l^{(R)}(t) + \vec{E}_l^{(D)}(t) + \vec{E}_l^{(E)}(t) \ , \tag{7}$$

and making use of Eqs. (4)–(6), we can obtain Ω_{l1}

and Ω_{l2} in Eqs. (3).

In carrying out the substitutions we make several approximations. We discard terms like $\cos(\omega_k + \omega_l)t$ oscillating at double the optical frequency, on the grounds that these average to zero over any measurable time interval. Secondly, we suppose that the first term in Eq. (4), representing the reaction- or self-field, will be negligible compared with the terms under the first summation in Eq. (5), representing the fields contributed by all other atoms, when the number of atoms is large. We then obtain

$$\hbar\Omega_{l1}(t) = \sum_{k\neq l} \left(\frac{3(\vec{\mu}_l \cdot \vec{n}_{kl})^2 - \vec{\mu}_l^{\,2}}{d_{kl}^{\,3}} + \frac{(\vec{\mu}_l \cdot \vec{n}_{kl})^2 + \vec{\mu}_l^{\,2}}{2c^2 d_{kl}} \omega_k^{\,2} \right) [-x_k(t)\cos(\omega_l - \omega_k)t - y_k(t)\sin(\omega_l - \omega_k)t]$$
$$+ \sum_k \frac{2\vec{\mu}_l^{\,2}\omega_k^{\,3}}{3c^3} [x_k(t)\sin(\omega_l - \omega_k)t - y_k(t)\cos(\omega_l - \omega_k)t] - \vec{\mu}_l \cdot \vec{\epsilon}\mathcal{E}(t)\cos[(\omega_l - \omega_0)t - \psi(t)] , \quad (8)$$

$$\hbar\Omega_{l2}(t) = \sum_{k\neq l} \left(\frac{3(\vec{\mu}_l \cdot \vec{n}_{kl})^2 - \vec{\mu}_l^{\,2}}{d_{kl}^{\,3}} + \frac{(\vec{\mu}_l \cdot \vec{n}_{kl})^2 + \vec{\mu}_l^{\,2}}{2c^2 d_{kl}} \omega_k^{\,2} \right) [x_k(t)\sin(\omega_l - \omega_k)t - y_k(t)\cos(\omega_l - \omega_k)t]$$
$$+ \sum_k \frac{2\vec{\mu}_l^{\,2}\omega_k^{\,3}}{3c^3} [x_k(t)\cos(\omega_l - \omega_k)t + y_k(t)\sin(\omega_l - \omega_k)t] + \vec{\mu}_l \cdot \vec{\epsilon}\mathcal{E}(t)\sin[(\omega_l - \omega_0)t - \psi(t)] , \quad (9)$$

$$\hbar\Omega_{l3}(t) = 0 . \quad (10)$$

Now the frequency spread, even for an appreciably inhomogeneously broadened spectrum, is generally very much less than the peak frequency ω_0. We may therefore, to a good approximation, replace the factors $\omega_k^{\,2}$ and $\omega_k^{\,3}$ appearing under the summations in Eqs. (8) and (9) by $\omega_0^{\,2}$ and $\omega_0^{\,3}$. We also make the reasonable assumption that the state of the kth atom is not appreciably correlated with the distance d_{kl} from the lth atom, nor with the scalar product $\vec{\mu}_l \cdot \vec{n}_{kl}$. We then replace the coefficients appearing under the summations in Eqs. (8) and (9) by their mean values over all atoms, and write

$$\gamma \equiv \frac{1}{\hbar} \left\{ \frac{3(\vec{\mu}_l \cdot \vec{n}_{kl})^2 - \vec{\mu}_l^{\,2}}{d_{kl}^{\,3}} + \frac{(\vec{\mu}_l \cdot \vec{n}_{kl})^2 + \vec{\mu}_l^{\,2}}{2c^2 d_{kl}} \omega_0^{\,2} \right\}_{\substack{\text{average} \\ \text{over atoms}}} , \quad (11)$$

$$\beta \equiv \left\{ \frac{2\mu_l^{\,2}\omega_0^{\,3}}{3c^3\hbar} \right\}_{\substack{\text{average} \\ \text{over atoms}}} , \quad (12)$$

$$\alpha \equiv \left\{ \vec{\mu}_l \cdot \vec{\epsilon}/\hbar \right\}_{\substack{\text{average} \\ \text{over atoms}}} , \quad (13)$$

where γ, β, $\alpha\mathcal{E}$ have dimensions of frequency. This amounts to the assumption that all atoms interact with nearly the same electromagnetic field. In general, some errors are introduced in this step, for the field varies somewhat throughout a typical sample. In the Appendix we examine the variation for a cylindrical sample of atoms of thickness c/ω_0 or $\lambda/2\pi$, when the dipole moments point along the axis. We show that the total variation covers a range of 2:1, although it is smaller for most of the atoms of the sample. This contributes an additional inhomogeneous linewidth of order $1/T_s$, which is certainly not negligible in general, although it is negligible if $T_2^* \ll T_s$. However, we shall be content to ignore these variations in the following treatment of the superradiance problem, in order to emphasize the effects of the nongeometric inhomogenous spectral broadening. Their inclusion would complicate the problem by another order of magnitude, for the desired solutions would become functions of three variables (time, detuning, and position). We may imagine, therefore, that the atoms are so arranged that they see very nearly the same field.[8]

We now pass to the continuum limit, in which the sums over atoms are replaced by integrals. For this purpose we introduce the inhomogeneous atomic spectral distribution function $g(\omega)$, which is peaked at ω_0, and is normalized so that

$$\int_0^\infty g(\omega)\, d\omega = 1 . \quad (14)$$

If there are N atoms altogether, the number having level spacing corresponding to a narrow frequency interval between ω and $\omega + d\omega$ is $Ng(\omega)\, d\omega$. The Bloch vector $\vec{r}$ and the interaction vector $\vec{\Omega}$ for each atom are now characterized by a frequency

ω and a time t, and instead of Eqs. (8)–(10) we may write

$$\Omega_1(\omega, t) = -N\gamma \int_0^\infty d\omega' [x(\omega', t)g(\omega')\cos(\omega-\omega')t + y(\omega', t)g(\omega')\sin(\omega-\omega')t]$$
$$+ N\beta \int_0^\infty d\omega' [x(\omega', t)g(\omega')\sin(\omega-\omega')t - y(\omega', t)g(\omega')\cos(\omega-\omega')t]$$
$$- \alpha \mathcal{E}(t)\cos[(\omega-\omega_0)t - \psi(t)] , \tag{15}$$

$$\Omega_2(\omega, t) = N\gamma \int_0^\infty d\omega' [x(\omega', t)g(\omega')\sin(\omega-\omega')t - y(\omega', t)g(\omega')\cos(\omega-\omega')t]$$
$$+ N\beta \int_0^\infty d\omega' [x(\omega', t)g(\omega')\cos(\omega-\omega')t + y(\omega', t)g(\omega')\sin(\omega-\omega')t]$$
$$+ \alpha \mathcal{E}(t)\sin[(\omega-\omega_0)t - \psi(t)], \tag{16}$$

$$\Omega_3(\omega, t) = 0 . \tag{17}$$

With the help of Eq. (2), these equations for $\vec{\Omega}(\omega, t)$ allow us to obtain the equations of motion for the components $x(\omega, t)$, $y(\omega, t)$, $z(\omega, t)$ of the Bloch vector. It is convenient to introduce the complex variable

$$\rho(\omega, t) \equiv x(\omega, t) + iy(\omega, t) , \tag{18}$$

which permits the equations of motion to be expressed in a somewhat more compact form. After some rearrangement of terms, we obtain the two equations

$$\dot{\rho}(\omega, t)e^{i\Delta\omega t} = Nz(\omega, t)(\beta+i\gamma)\int_0^\infty d\omega'\, \rho(\omega', t)g(\omega')e^{i\Delta\omega' t} + i\alpha z(\omega, t)\mathcal{E}(t)e^{i\psi(t)} , \tag{19}$$

$$\dot{z}(\omega, t) = \tfrac{1}{2}\rho^*(\omega, t)e^{-i\Delta\omega t}\left\{-N(\beta+i\gamma)\int_0^\infty d\omega'\, \rho(\omega', t)\, g(\omega')e^{i\Delta\omega' t} - i\alpha\mathcal{E}(t)e^{i\psi(t)}\right\} + \text{c.c.} , \tag{20}$$

where we have written $\Delta\omega \equiv \omega - \omega_0$. The second of these equations is actually derivable from the first and contains no additional information, as can be seen at once if we make use of the normalization condition for the Bloch vector

$$\frac{d}{dt}[z^2(\omega, t) + |\rho(\omega, t)|^2] = 0. \tag{21}$$

We now have a set of coupled integrodifferential equations for the components of the Bloch vector in the two variables ω and t. The solution allows us to determine how each atom evolves in time. In general, it is to be expected that different atoms having different resonant frequencies evolve differently.

III. ENERGY FLOW

The integral occurring in Eqs. (19) and (20) is rather closely related to the energy flow from the atomic system, in the absence of an external field. To see this, we recall that, for any atom, $\hbar\omega$ times the z component of the Bloch vector is the expectation value of the atomic energy, relative to the ground-state energy level. If U denotes the total mean energy of the atomic system in the absence of an external field, then

$$U(t) = N\int_0^\infty \hbar\omega g(\omega)z(\omega, t)\, d\omega$$
$$\approx N\hbar\omega_0 \int_0^\infty g(\omega)z(\omega, t)\, d\omega . \tag{22}$$

From Eq. (20) we then have for the rate of energy flow from the atomic system

$$-\frac{dU}{dt} = \tfrac{1}{2}N^2\hbar\,\omega_0(\beta+i\gamma)$$
$$\times \left|\int_0^\infty d\omega'\, \rho(\omega', t)g(\omega')e^{i\Delta\omega' t}\right|^2 + \text{c.c.}$$
$$= N^2\hbar\omega_0\beta|S(t)|^2 , \tag{23}$$

where

$$|S(t)| \equiv \left|\int_0^\infty d\omega'\, \rho(\omega', t)g(\omega')e^{i\Delta\omega' t}\right| . \tag{24}$$

Equation (24) does not define $S(t)$ unambiguously, but the ambiguity will be removed shortly. As usual for a superradiant system, we find that the energy flow rate is proportional to the square of the number of atoms. From energy-balance considerations of the whole system, it is clear that $|S(t)|^2$ is intimately related to the intensity of the total electromagnetic field radiated by the sample.

IV. EQUATIONS OF MOTION IN POLAR COORDINATES

Instead of working with the Cartesian components of the Bloch vector $\vec{r}$, we shall find it convenient to work with the two polar coordinates θ, ϕ, which form an irreducible set. On putting

$$x(\omega, t) = \sin\theta(\omega, t)\cos\phi(\omega, t)\ ,$$
$$y(\omega, t) = \sin\theta(\omega, t)\sin\phi(\omega, t)\ , \tag{25}$$
$$z(\omega, t) = \cos\theta(\omega, t)\ ,$$

in Eq. (19), we obtain the complex equation

$$[\cos\theta(\omega, t)\,\dot{\theta}(\omega, t) + i\sin\theta(\omega, t)\,\dot{\phi}(\omega, t)]\,e^{i[\Delta\omega t + \phi(\omega, t)]}$$
$$= \cos\theta(\omega, t)\left[N(\beta + i\gamma)\int_0^\infty \sin\theta(\omega', t)\ g(\omega')e^{i[\Delta\omega' t + \phi(\omega', t)]}d\omega' + i\alpha\mathcal{E}(t)e^{i\psi(t)}\right]\ , \tag{26}$$

which can be expressed in the form of two real coupled integrodifferential equations:

$$\dot{\theta}(\omega, t) = N\beta\,\mathrm{Re}[S(\omega, t)] - N\gamma\,\mathrm{Im}[S(\omega, t)] + \alpha\mathcal{E}(t)\sin[\Delta\omega t + \phi(\omega, t) - \psi(t)]\ , \tag{27}$$

$$\dot{\phi}(\omega, t) = \cot\theta(\omega, t)\{N\beta\,\mathrm{Im}[S(\omega, t)] + N\gamma\,\mathrm{Re}[S(\omega, t)] + \alpha\mathcal{E}(t)\cos[\Delta\omega t + \phi(\omega, t) - \psi(t)]\}\ , \tag{28}$$

with $S(\omega, t)$ given by

$$S(\omega, t) \equiv e^{-i[\Delta\omega t + \phi(\omega, t)]}\int_0^\infty d\omega'\sin\theta(\omega', t) \times g(\omega')e^{i[\Delta\omega' t + \phi(\omega', t)]}\ . \tag{29}$$

$S(\omega, t)$ will be seen to differ from $|S(t)|$ defined in Eq. (24) only by a phase that carries the ω dependence.

Inspection of Eq. (29) shows that, in the limit in which $g(\omega) \to \delta(\omega - \omega_0)$, and there is no inhomogeneous broadening, $S(\omega, t) \to \sin\theta(\omega_0, t)$, and Eqs. (27) and (28) reduce to the Eqs. (46) and (47) of Stroud *et al.*,[6] as expected. In this limit, when $\mathcal{E}(t) = 0$, we find the usual well-known solutions

$$\theta(\omega, t) = \theta(t) = 2\tan^{-1}e^{N\beta(t - t_0)}\ , \tag{30}$$

with

$$t_0 = (1/N\beta)\ln\cot[\tfrac{1}{2}\theta(0)]\ , \tag{31}$$

$$\phi(\omega, t) = \phi(t) = \phi(0) - \frac{\gamma}{\beta}\ln\left(\frac{\cosh N\beta(t - t_0)}{\cosh(N\beta t_0)}\right)\ , \tag{32}$$

and

$$\mathcal{R} = N^2\hbar\omega_0\beta\,|S(\omega, t)|^2 = N^2\hbar\omega_0\beta\,\mathrm{sech}^2 N\beta(t - t_0)\ , \tag{33}$$

where $\mathcal{R}$ is the rate of the energy flow. The width of the superradiant pulse, which is of order $1/N\beta$, is generally known as the supperradiance time T_s. The time of occurrence t_0 of the pulse peak depends on the initial state, but is generally a small multiple of T_s and of the same order of magnitude for atoms excited near the upper state.

V. EXTREME INHOMOGENEOUS BROADENING

Because of the two variables ω and t and the nonlinearities, the coupled equations (27)–(29) are not easy to solve when $g(\omega)$ is not a δ function. However, the other limiting case of extreme inhomogeneous broadening can be handled without difficulty. If $g(\omega)$ is a very broad spectral density of width $1/T_2{}^*$, with $T_2{}^* \ll 1/N\beta$, then the $S(\omega, t)$ integral in Eq. (29) will be very small for times appreciably in excess of $T_2{}^*$. Hence, except for such very short times, both $\dot{\theta}(\omega, t)$ and $\dot{\phi}(\omega, t)$ will be determined largely by the external electromagnetic field. If, in addition, there is no external field and $\mathcal{E}(t) = 0$, then $\int_0^t \dot{\theta}(\omega, t')\,dt'$ and $\int_0^t \dot{\phi}(\omega, t')\,dt'$ are negligibly small, and $\theta(\omega, t)$ and $\phi(\omega, t)$ retain their initial values $\theta(\omega, 0)$ and $\phi(\omega, 0)$ indefinitely. In that case it follows from Eq. (29) that

$$S(\omega, t) \approx e^{-i[\Delta\omega t + \phi(\omega, 0)]}\int_0^\infty \sin\theta(\omega', 0) \times g(\omega')e^{i\phi(\omega', 0)}e^{i\Delta\omega' t}\,d\omega'\ ,$$

so that

$$|S(\omega, t)|^2 \approx \left|\int_0^\infty \sin\theta(\omega', 0)g(\omega') \times e^{i\phi(\omega', 0)}e^{i\Delta\omega' t}\,d\omega'\right|^2\ , \tag{34}$$

and the energy flow from the sample is proportional to the square of the Fourier transform of $\sin\theta(\omega', 0)g(\omega')\exp[i\phi(\omega', 0)]$.

We notice that $|S(\omega, t)|^2$ is determined entirely by the initial orientations θ, ϕ of the atomic Bloch vectors, which gradually dephase in time. The radiated signal therefore has the character of an optical free-induction decay.

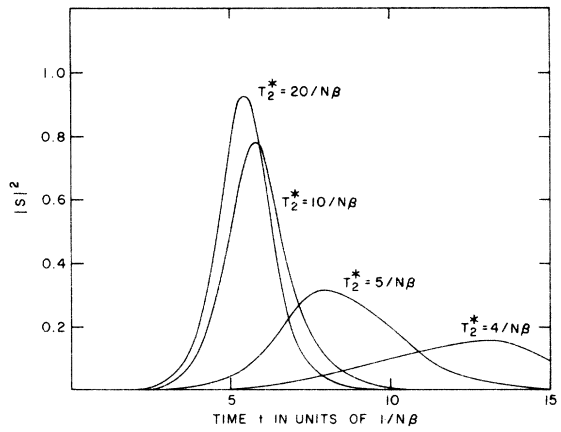

FIG. 1. Energy radiated by an initially excited atomic system as a function of time, for various values of the inhomogeneous lifetime T_2^*. The energy flow rate is given by $N^2\hbar\omega_0\beta|S(t)|^2$.

In the special case in which all atoms are excited equally initially, so that $\theta(\omega, 0) = \theta$ and $\phi(\omega, 0) = \phi$, the integral reduces to the Fourier transform of the spectral density $g(\omega)$, which is the amplitude correlation function $\gamma(t)$,

$$\gamma(t) \equiv \int_0^\infty g(\omega')e^{i\Delta\omega' t}\, d\omega' . \tag{35}$$

Hence, in this case

$$N^2\hbar\omega_0\beta|S(\omega, t)|^2 \approx N^2\hbar\omega_0\beta\sin^2\theta\,|\gamma(t)|^2 , \tag{36}$$

which is the result obtained previously[3] for the rate of energy flow. For example, for a Gaussian spectral density $g(\omega)$, this implies a Gaussian fall-off of the radiated intensity with time, and for a Lorentzian spectral density $g(\omega)$ it implies an exponential fall-off with time. But, irrespective of the form of $g(\omega)$, Eq. (36) implies that $|S(\omega, t)| \leq |S(\omega, 0)|$. The conclusion that $\theta(\omega, t)$ and $\phi(\omega, t)$ retain their initial values $\theta(\omega, 0)$ and $\phi(\omega, 0)$ indefinitely of course implies that extremely little of the stored atomic energy is radiated away by coherent processes. In practice, any excitation energy will tend to be dissipated by incoherent processes eventually, but, if the incoherent dissipation is slow, the excitation may persist for some time. It is this persistence of the stored energy in a strongly inhomogeneously broadened sample that makes it possible to observe the photon echo effect.[14]

VI. GENERAL SOLUTIONS FOR ZERO EXTERNAL FIELD

When the inhomogeneous linewidth $1/T_2^*$ is of the same order as $N\beta$, the solutions of Eqs. (27)–(29) become much more complicated, even when $\mathcal{E} = 0$, and we find that in general different atoms evolve differently in time. Because of the atomic dephasing that takes place during the growth of the superradiant pulse, fewer and fewer atoms radiate cooperatively or coherently as T_2^* decreases from infinity. As might be expected from inspection of Eqs. (30)–(33), the effect of reducing

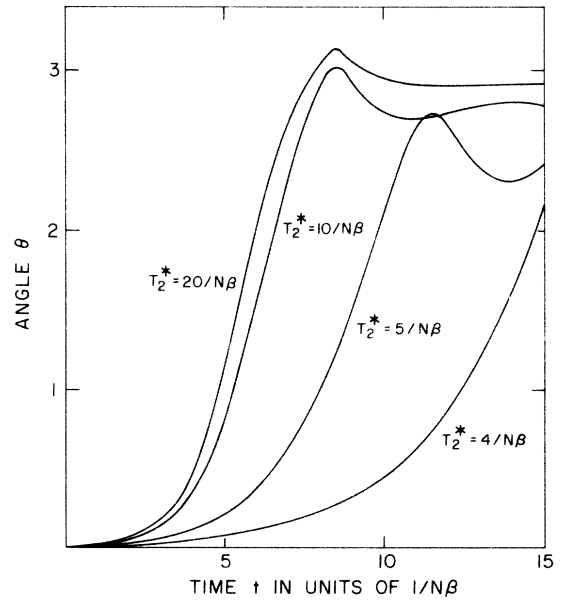

FIG. 2. Time variation of the polar angle $\theta(\omega_0, t)$ of the atomic Bloch vector, for atoms at the center of the inhomogeneously broadened line, for various values of the inhomogeneous lifetime T_2^*.

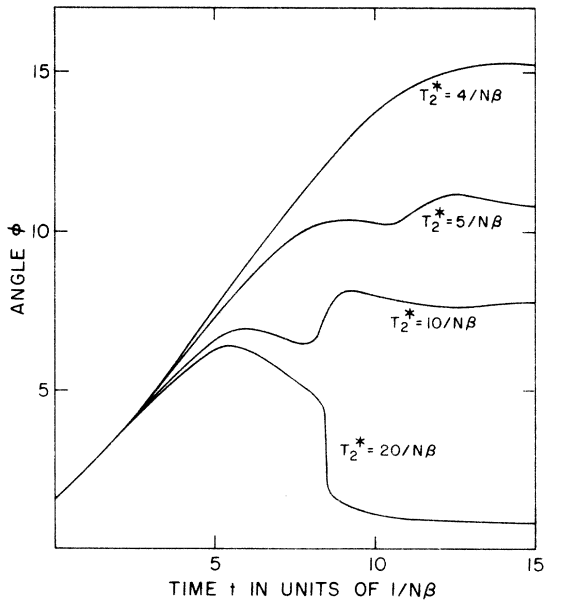

FIG. 3. Time variation of the azimuthal angle $\phi(\omega_0, t)$ of the atomic Bloch vector, for atoms at the center of the inhomogeneously broadened line, for various values of the inhomogeneous lifetime T_2^*.

the number of cooperating atoms is to broaden and to delay the superradiant pulse, and to reduce its amplitude.

We have not succeeded in finding analytic solutions of the coupled integrodifferential equations (27)–(29) in the general case. However, the equations readily lend themselves to step-by-step integration. For the purpose of numerical solution, we have taken $g(\omega)$ to be in the form of a Gaussian function:

$$g(\omega) = \frac{T_2^*}{2\pi^{3/2}} \exp\left(\frac{-(\omega-\omega_0)^2 T_2^*}{4\pi^2}\right), \qquad (37)$$

centered on ω_0 and having a standard deviation $1/\sqrt{2}\ T_2^*$ in frequency, or $\sqrt{2}\,\pi/T_2^*$ in angular frequency.[15] We have chosen γ and β given by Eqs. (11) and (12) to be equal, which is probably appropriate for a sample that is only modestly smaller than a wavelength, as the evaluations of the averages γ and β in the Appendix suggest. If the external field $\mathcal{E}=0$, and if we express all times in units of the superradiance time $T_s = 1/N\beta$, then the only adjustable parameter is the ratio of the inhomogeneous lifetime T_2^* to T_s.

To illustrate the solutions, we assume that all atoms start very nearly in the excited state,[16] with $\theta(\omega, 0) = 0.01$ and $\phi(\omega, 0) = \pi/2$ for all ω. Figure 1 illustrates the rate of energy flow from the system, which is proportional to $|S(\omega, t)|^2$, as a function of time t, for several different ratios of T_2^*/T_s. When $T_2^*/T_s = 20$, the solution is very close to that given by Eq. (33) for negligible inhomogeneous broadening. Smaller values of T_2^*/T_s lead to smaller, more spread out, and more delayed pulses, as might be expected from the fact that there is an increasing amount of dephasing among atoms, so that fewer atoms radiate cooperatively. When $T_2^*/T_s = 3$ the output pulse is already so small as to be invisible on the scale of this figure.

Figures 2 and 3 illustrate the solutions for the angles $\theta(\omega_0, t)$ and $\phi(\omega_0, t)$ of the Bloch vector at the center frequency ω_0 of the inhomogeneous line, for the same values of T_2^*/T_s. It will be seen that $\theta(\omega_0, t)$ does not quite reach the value π, corresponding to the ground state of the atom, in the steady state. This is so even when T_2^*/T_s is as large as 20, when it might be thought that the effects of inhomogeneous broadening would become negligible. The implication is that the atoms do not quite return to the ground state, but that some of the energy of excitation remains trapped within the system, to be dissipated eventually by incoherent processes.[17]

The azimuthal angles $\phi(\omega_0, t)$ shown in Fig. 3 at first increase with time, but, after some fairly complicated variations, they tend towards definite values depending on T_2^*/T_s in the steady state.

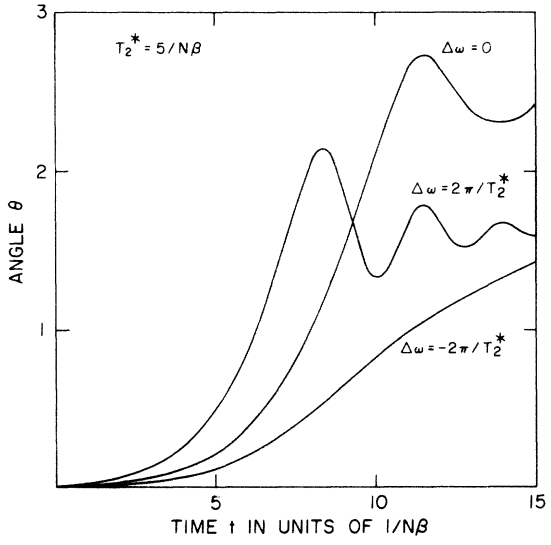

FIG. 4. Time variation of the polar angle $\theta(\omega,t)$ of the atomic Bloch vector, for atoms of three different frequencies, with inhomogeneous lifetime $T_2^* = 5T_s = 5/N\beta$.

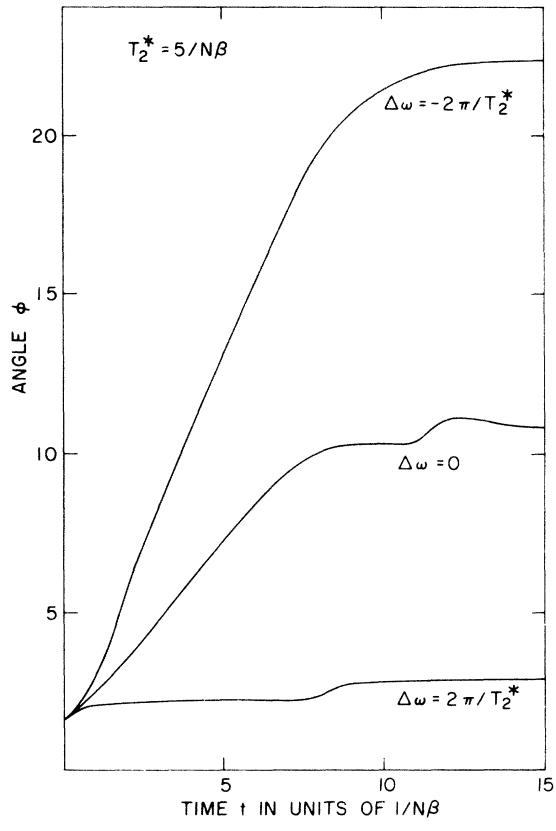

FIG. 5. Time variation of the azimuthal angle $\phi(\omega,t)$ of the atomic Bloch vector, for atoms of three different frequencies, with inhomogeneous lifetime $T_2^* = 5T_s = 5/N\beta$.

This behavior is quite different from that given by Eq. (32), in the absence of inhomogeneous broadening, when $\phi(\omega, t)$ varies linearly with time after a long time. The time derivative $\dot{\phi}(\omega, t)$ has a simple physical interpretation, whose significance is seen most easily in terms of the expectation value of the atomic dipole moment operator $\hat{\mu}(\omega, t)$. Recalling that $\langle\hat{\mu}(\omega, t)\rangle$ is proportional to the x component of the atomic Bloch vector, we find that

$$\langle\hat{\mu}(\omega, t)\rangle = \vec{\mu}(\omega)\sin\theta(\omega, t)\cos[\omega t + \phi(\omega, t)], \quad (38)$$

where $\vec{\mu}(\omega)$ is the matrix element of $\hat{\mu}$ between upper and lower states. This equation shows that $\dot{\phi}(\omega, t)$ represents a shift of the frequency of oscillation of the dipole from its "natural" frequency ω, and since the far field radiated by the atomic dipole is proportional to $\langle(d^2/dt^2)\hat{\mu}(\omega, t)\rangle$, the same shift will be manifest in the electromagnetic field. This frequency shift, or chirp, is itself time dependent, and can become very large at certain times. Figure 3 shows that, when $T_2^*/T_s = 20$, a large frequency shift occurs at time $8.5T_s$ for atoms of frequency ω_0. This frequency shift is, however, associated with phase reversal of the field when it is close to zero, rather than with any physical processes of interest. At this time the superradiant output pulse from the system is almost over.

Figures 4 and 5 showing the behavior of atoms at three different frequencies, for $T_2^*/T_s = 5$, illustrate another interesting feature of the decay process. It will be seen that the time development is quite distinct for atoms of different frequencies, and that, moreover, those atoms having frequencies equally far above and below the central frequency ω_0 do not, in general, behave similarly. This asymmetry is also reflected in the chirp or frequency shift characterized by $\dot{\phi}(\omega, t)$. The importance of the different behavior of different atoms is emphasized even more in Figs. 6 and 7. Here $T_2^*/T_s = 20$, and it might well be thought that effects associated with inhomogenous broadening would be negligible for such a long inhomogeneous lifetime. Indeed, the superradiant pulse emitted (see Fig. 1) has very nearly the form given by Eq. (33) for the strictly resonant atomic system. Nevertheless, even in this case, when we examine the behavior of atoms of different frequencies, we find very substantial differences in time development. In particular, while phase reversal effects appear at and below the midfrequency ω_0, these effects are much smaller for atoms resonant at higher frequencies.

VII. SOLUTIONS FOR A COHERENT EXCITATION PULSE

One obvious way of producing the initial excitation of the atomic system is to allow a coherent pulse of light to interact with it. Moreover, by appropriately choosing the frequency and duration of the excitation pulse, we may selectively excite a small group of atoms, lying within a narrow frequency range, out of the total system.

To illustrate this effect we have computed solutions of Eqs. (27)–(29) for an atomic system that is initially in the ground state, and is then exposed to a light pulse of constant amplitude and of duration $T = 20T_2^*$, centered on frequency ω_0,

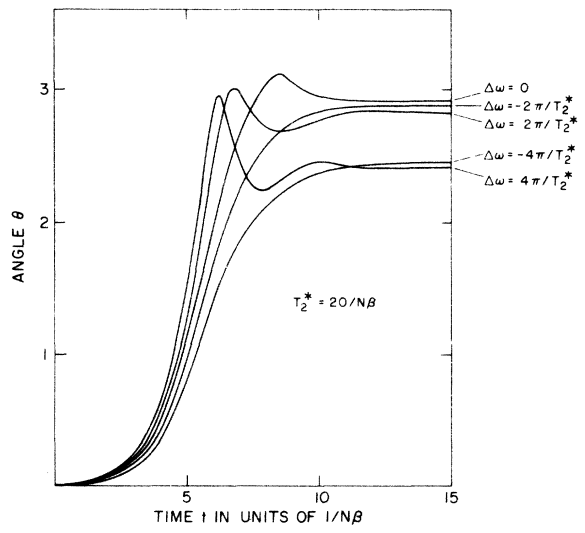

FIG. 6. Time variation of the polar angle $\theta(\omega,t)$ of the atomic Bloch vector, for atoms of five different frequencies, with inhomogeneous lifetime $T_2^* = 20T_s = 20/N\beta$.

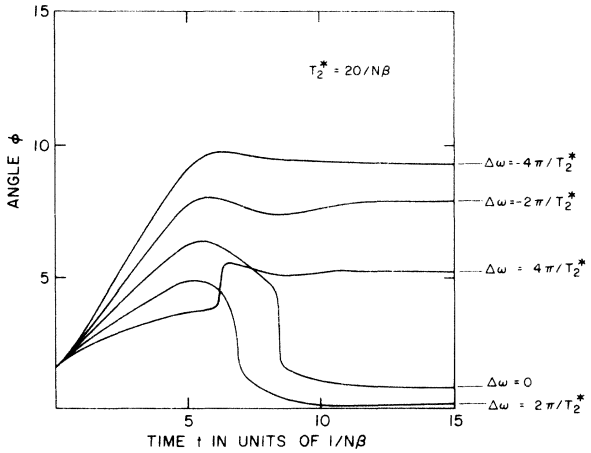

FIG. 7. Time variation of the azimuthal angle $\phi(\omega,t)$ of the atomic Bloch vector, for atoms of five different frequencies, with inhomogeneous lifetime $T_2^* = 20T_s = 20/N\beta$.

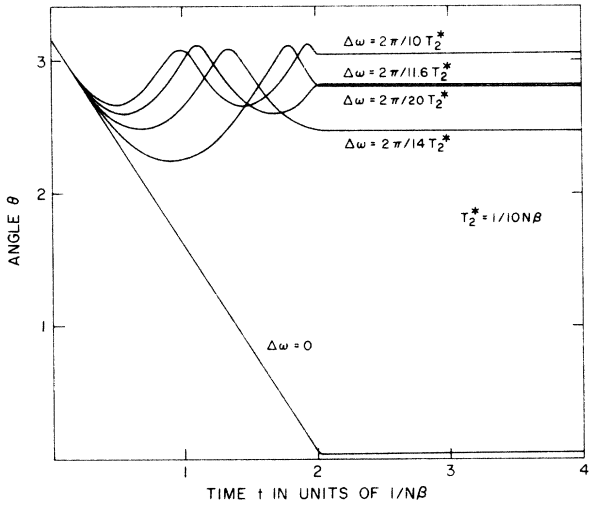

FIG. 8. Time variation of the polar angle of the atomic Bloch vector, for a system of atoms that is initially in the ground state and is then exposed to an exciting pulse of area π, of frequency ω_0, for a time $T=2/N\beta$, with $T_2{}^*=\frac{1}{10}T_s=1/10N\beta$. The response is shown at atomic frequency ω_0 and at four atomic frequencies slightly above ω_0. The response at frequencies below ω_0 is very similar.

with $T_2{}^*/T_s=\frac{1}{10}$ and $\psi(t)=0$. The amplitude of the external field $\mathcal{E}(t)$ was chosen so that $\alpha\int_0^\infty \mathcal{E}(t')dt'=\pi$, which, in the absence of inhomogeneous broadening, would produce almost total inversion. Ordinarily, we would not expect to find significant superradiant effects when the inhomogeneous lifetime $T_2{}^*$ is much shorter than the superradiance time $T_s=1/N\beta$, as is implied by Eq. (36). However, because of the long duration T of the excitation pulse, only those atoms having frequencies close to ω_0, within a range of order $1/T$, will be excited appreciably. This small group of atoms, corresponding to about $\frac{1}{20}$ of the total number, is able to radiate collectively within a much longer time than $T_2{}^*$, although the presence of the other atoms tends to quench the radiation.

Figure 8 shows the polar angle $\theta(\omega,t)$ of the Bloch vector for five different groups of atoms as a function of time. It will be seen that only the atoms of frequencies close to ω_0 are excited almost fully; the others become only partly excited at the end of the exciting pulse, after being taken through a few cycles of partial excitation. The figure illustrates the behavior of atoms having resonant frequencies at and above ω_0, but in this instance the time development is similar at frequencies below ω_0. The polar angles $\theta(\omega,t)$ change very little when $t>T(T=2/N\beta)$ and the excitation pulse is over, which indicates that relatively little of the energy of excitation emerges as radiation. Figure 9 shows the azimuthal angles $\phi(\omega,t)$ of the same groups of atoms as a function of time. Atoms appreciably off-resonance experience phase reversals when the field is close to zero, at times dependent on their resonant frequencies.

Figure 10 shows the value of $|S(t)|^2$ as a function of time. Here the interesting result is that, following the end of the excitation pulse, the system radiates a pulse of energy, whose duration is of the same order of magnitude as the duration of

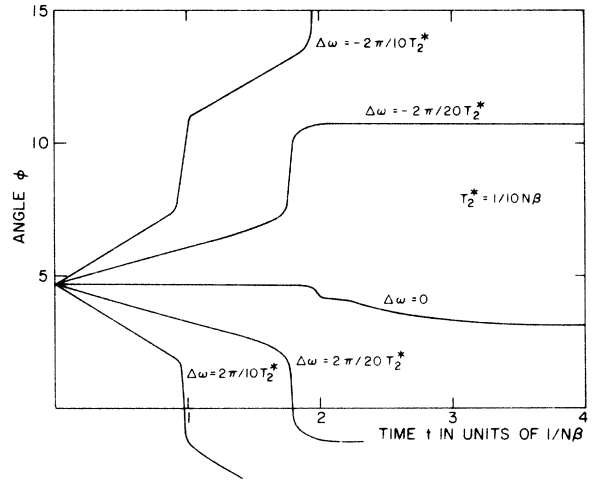

FIG. 9. Time variation of the azimuthal angle of the atomic Bloch vector, for a system of atoms that is initially in the ground state and is then exposed to an exciting pulse of area π, of frequency ω_0, for a time $T=2/N\beta$, with $T_2{}^*=\frac{1}{10}T_s=1/10N\beta$. The response is shown at five different atomic frequencies very close to ω_0.

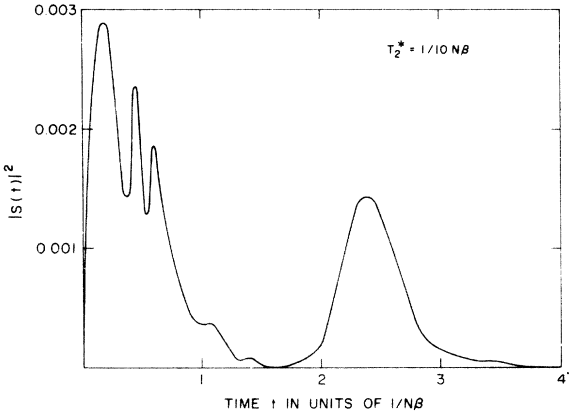

FIG. 10. Time variation of $|S(t)|^2$ for a system of atoms that is initially in the ground state and is then exposed to an exciting pulse of area π, of frequency ω_0, for a time $T=2/N\beta$, with $T_2{}^*=\frac{1}{10}T_s=1/10N\beta$. When the exciting pulse is over, the energy flow rate from the system is given by $N^2\hbar\omega_0\beta|S(t)|^2$. A small superradiant pulse is seen to follow the exciting pulse.

the exciting pulse. However the magnitude of the radiated pulse is very small; it is about 700 times smaller than the radiation pulse of which the system is capable in principle, if all atoms were to radiate cooperatively. But, remembering that only about 1/20 of the atoms were appreciably excited initially, we see that the pulse amplitude is still consistent with a superradiant effect proportional to the square of the number of atoms. The phenomenon is very similar to effects reported by Bloom in nuclear magnetic resonance experiments in inhomogeneously magnetic fields.[18]

In many respects the response of the atomic system resembles an optical free induction decay,[19] in which the atomic dipoles simply dephase progressively following the excitation pulse. Indeed the curves in Figs. 8 and 9 indicate that $\theta(\omega, t)$ and $\phi(\omega, t)$ change rather little after $t = 2/N\beta$, although a small rise in $\theta(\omega_0, t)$ can be discerned. We can carry out a rough test of this interpretation of Fig. 10, by comparing the shape of the output pulse with the form predicted by Eq. (34) for free induction decay. From Fig. 9 it appears that, immediately following the excitation pulse, $\phi(\omega)$ is very approximately a linear function of ω, with

$$\phi(\omega) - \phi(\omega_0) \sim -(60T_2^*/\pi)\Delta\omega. \tag{39}$$

It then follows from the Fourier transform structure of Eq. (34) that the output intensity $|S(\omega, t)|^2$ should reach its greatest value at a time $60T_2^*/\pi \sim 2N\beta$ following the excitation pulse, if the process were purely a free induction decay. In fact the pulse peak occurs after a time that is nearly an order of magnitude shorter, which suggests that superradiant effects, involving at least a partial decay of the atoms, are making a contribution. As the length of the coherent excitation pulse is increased, the number of atoms left sufficiently excited at the end of the pulse decreases, until superradiant effects become negligible compared with free induction decay. This appears to have been the situation in most of the observations reported so far.[19,20]

VIII. CONCLUSION

We have derived the integrodifferential equations yielding the behavior of different atoms in time, for an inhomogeneously broadened sample of matter interacting with radiation.[21] The results show that different atoms evolve differently in time, that there is no symmetry, in general, with respect to the midfrequency, even for a symmetric spectral distribution, and that the effects of inhomogeneous broadening are significant even when the inhomogeneous lifetime T_2^* is 20 times as great as the superradiance time $T_s = 1/N\beta$.

When $T_2^* \ll T_s$, the superradiant pulse from the sample is normally wiped out by the destructive interference of radiation from the different atoms. However, with the help of a coherent excitation pulse that selectively excites a small group of atoms of the sample, it is still possible to produce conditions under which a delayed superradiant-pulse appears. These results suggest that superradiant-effects, as distinct from free optical induction decay, should be observable in a solid, inhomogeneously broadened sample of matter.

APPENDIX: EVALUATION OF CERTAIN ATOMIC AVERAGES

In the following we shall examine the variation of the quantity

$$\gamma_l \equiv \frac{1}{\hbar}\left\langle \frac{3(\vec{\mu}_l\cdot\vec{n}_{kl})^2 - \vec{\mu}_l^{\,2}}{d_{kl}^{\,3}} + \frac{(\vec{\mu}_l\cdot\vec{n}_{kl})^2 + \vec{\mu}_l^{\,2}}{2c^2 d_{kl}}\,\omega_0^{\,2}\right\rangle_{\substack{\text{average over}\\ \text{atoms } k}}, \tag{A1}$$

where the average is to be taken over all atoms k, for various fixed positions of atom l, for a sample in the form of a disc. We take the dipole moment $\vec{\mu}_l$ to point along the axis of the disc, and choose the radius a and the height h to be of the same order of magnitude, with

$$a = c/\omega_0, \tag{A2}$$

which is the midwavelength multiplied by $1/2\pi$. This ensures that the sample is appreciably

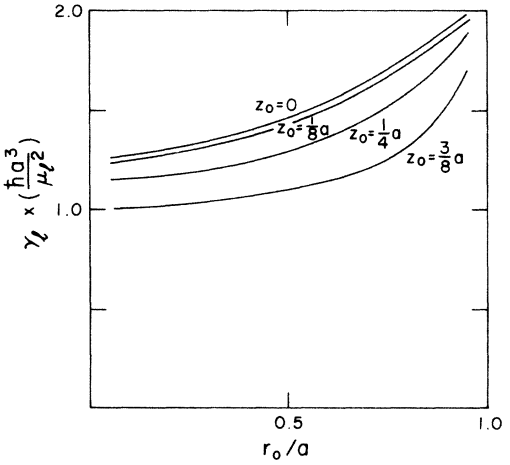

FIG. 11. Variation of γ_l over a cylindrical sample of radius a and height a for different positions of atom l. r_0 is the radial coordinate of atom l, and z_0 its axial coordinate measured from the center of the sample.

smaller than the wavelength of the midfrequency ω_0.

If the average in Eq. (A1) is to be finite, there must exist a minimum distance ϵ between the atoms k and atom l, and in general this will be determined by the atomic density ρ, such that

$$\rho \approx 1/\epsilon^3. \tag{A3}$$

In evaluating the average in Eq. (A1), we shall replace the sum over atoms by a continuous integral, and integrate over the volume of the sample up to a small region of linear dimensions ϵ surrounding atom l. We choose a system of cylindrical polar coordinates, with the z axis pointing along the sample axis and the origin at the center of the sample, and let (r_0, θ_0, z_0) be the coordinates of atom l. Then if $\epsilon \leq r_0 \leq a - \epsilon$ and $-\frac{1}{2}h + \epsilon \leq z_0 \leq \frac{1}{2}h - \epsilon$, we can express γ_l in the form:

$$\begin{aligned}\hbar\gamma_l = {} & \frac{\mu_l^2}{\pi a^2 h}\left(\int_0^{r_0-\epsilon} r\,dr + \int_{r_0+\epsilon}^{a} r\,dr\right)\int_0^{2\pi} d\theta \int_{-h/2}^{h/2} dz\, F(r, \theta, z; r_0, \theta_0, z_0) \\ & + \frac{\mu_l^2}{\pi a^2 h}(r_0 2\epsilon)\int_0^{2\pi} d\theta \left(\int_{-h/2}^{z_0-\epsilon} dz + \int_{z_0+\epsilon}^{h/2} dz\right) F(r_0, \theta, z; r_0, \theta_0, z_0) \\ & + \frac{\mu_l^2}{\pi a^2 h}(r_0 4\epsilon^2)\left(\int_0^{\theta_0-\epsilon/r_0} d\theta + \int_{\theta_0+\epsilon/r_0}^{2\pi} d\theta\right) F(r_0, \theta, z_0; r_0, \theta_0, z_0)\,,\end{aligned} \tag{A4}$$

where

$$\begin{aligned}F(r, \theta, z; r_0, \theta_0, z_0) = {} & 3(z - z_0)^2[r^2 + r_0^2 - 2rr_0\cos(\theta - \theta_0) + (z - z_0)^2]^{-5/2} - [r^2 + r_0^2 - 2rr_0\cos(\theta - \theta_0) + (z - z_0)^2]^{-3/2} \\ & + [(z - z_0)^2/2a^2][r^2 + r_0^2 - 2rr_0\cos(\theta - \theta_0) + (z_0 - z)^2]^{-3/2} \\ & + (1/2a^2)[r^2 + r_0^2 - 2rr_0\cos(\theta - \theta_0) + (z - z_0)^2]^{-1/2}\,.\end{aligned} \tag{A5}$$

The second and third terms in Eq. (A4) represent corrections to the first term, corresponding to volume elements excluded from the first term. If $r_0 = 0$, the r integral in the first term extends from ϵ to a, the factor $(r_0 2\epsilon)$ in the second term is replaced by $\frac{1}{2}\epsilon^2$, and the third term can be eliminated.

For certain positions (r_0, θ_0, z_0) of atom l, such as for points on the axis, the integrals can be evaulated and expressed in terms of elementary functions, but the expressions are quite long. We have evaluated the terms numerically for a number of different positions of atom l, for a sample for which $a = h = 100\epsilon$, and the results are summarized in Fig. 11. It will be seen that γ_l varies by a factor 2 over different positions, but that, for most of the atoms, the variation is within the range $\pm 20\%$.

For comparison, we find from Eq. (12) that $\hbar\beta \approx 0.67\mu_l^2/a^3$ under the same assumptions, so that γ and β are of the same order of magnitude.

If this variation of γ_l throughout the sample is interpreted as a frequency variation with a certain linewidth $1/T_2$, then since γ in Eqs. (27) and (28) is multiplied by the factor N, the effective linewidth $1/T_2$ resulting from the dispersion of γ is of order $N\mu^2/\hbar a^3 \sim N\beta = 1/T_s$. The geometric effects are therefore certainly not negligible. However, in the examples considered in Sec. VII, in which the system is selectively excited with a coherent excitation pulse and $T_2^* = T_s/10$, the geometric contribution to the linewidth is relatively small.

*Work supported by the National Science Foundation.

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[8]An example is any regular, plane polygon, with the atoms located at the vertices, and the polarization vector perpendicular to the plane.
[9]In the Appendix we examine the variation of the field experienced by atoms distributed throughout a cylindrical sample of thickness $c/\omega_0 = \lambda/2\pi$, when the dipole moments point along the axis. We show that the field variation may be characterized by an additional linewidth of order $1/T_s$. This is not negligible in general, but is relatively unimportant when $T_2^* \ll T_s$, as in some of the cases we examine.
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[15]Our definition of T_2^* may differ by a factor of order π from that used by other authors.
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[21]Since this work was completed, a paper was published by E. Ressayre and A. Tallet [Phys. Rev. Lett. 30, 1239 (1973)], in which the authors claim to have incorporated effects of inhomogeneous broadening in the analysis of the superradiance experiments of Skribanowitz *et al.* (Ref. 2). However, the solutions of the equations of motion for atoms of different frequencies were not presented.