Theory of superradiance in an extended, optically thick medium*

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This paper presents a semiclassical treatment of the evolution of an initially inverted system into a superradiant state in an extended, optically thick medium. In this process spontaneous emission and background thermal radiation initiate the collective radiative decay and produce a superradiant output pulse of intensity proportional to the square of the number of radiators. The treatment is based on the coupled Maxwell-Schrödinger equations, modified to include a fluctuating polarization source properly constructed to account for the effects of spontaneous emission. Computer results show that for a high-gain system only two parameters significantly influence the evolution process: T_R, the characteristic radiation damping time of the collective system; and θ_0, a function of the conditions which initiate the superradiant process. In this limit one obtains a normalized emission curve and simple analytical expressions for the time delay, pulse width, and peak intensity of the output radiation. These results are in good agreement with experiments. A comparison of our model with previous treatments of superradiance is given.

I. INTRODUCTION

Superradiance, the idea that the spontaneous emission rate of an assembly of atoms (or molecules) can be much greater than that of the same number of isolated atoms, has been the subject of much theoretical discussion since it was originally proposed by Dicke[1] in 1954. In the process of superradiant emission the atoms are coupled together by their common radiation field, and so decay cooperatively. The intensity emitted by N atoms is therefore proportional to N^2 instead of N. Thus, superradiance is a fundamental effect.

Historically, observations of cooperative emission effects go back at least as far as Hahn's spin-echo experiment of 1950.[2] In the ensuing years experimental observations of free-induction decay, echos, and other cooperative emission effects have been made in both optical[3] and longer-wavelength[4] regimes. Such phenomena can be termed "limited superradiance," in that only a small fraction of the energy stored in the sample is emitted cooperatively, so that the decay of the sample is essentially unaffected by the cooperative radiation.[5]

In 1973 the first observation of "strong superradiance" was made in optically pumped HF gas.[6] In this experiment virtually all of the energy stored in the sample was emitted cooperatively, and so the decay of the sample was dramatically accelerated. Although the emitted radiation had all the characteristics of superradiance described by Dicke, the detailed behavior (e.g., ringing, time delay of output) differed substantially from that predicted by the theoretical elaborations of Dicke's work then available.[7-13] Guided by the experimental observations, a simple theoretical model was developed[6,14] which accurately described the features of the emitted radiation. The present paper is an elaboration of that model. Recent experimental observations and their analysis are presented in another paper.[15,16]

Dicke considered two regimes, distinguished by whether the sample is small ("point sample") or large ("extended medium") compared to the wavelength of the emitted radiation. The description of superradiant emission in an extended sample, such as occurs in the HF experiments, requires a more complex analysis, since propagation effects must be fully taken into account. Although most theoretical treatments of superradiance have used quantized fields,[7-12,17-20] propagation effects are more easily included in the semiclassical approach (classical fields, quantized molecules) used by Dicke[1,21] and a few others.[6,13,14,22-25] The model presented here makes use of the semiclassical approach which, as discussed below, adequately describes a superradiant system. In fact, there is nothing inherently quantum mechanical about superradiance, as is illustrated by Dicke's description[1,21] of how a collection of classical dipoles appropriately prepared can exhibit superradiant behavior. Later, our semiclassical results will be compared with some results obtained in quantized field treatments.

The theory developed in this paper is relevant to several potentially useful applications of superradiance. For example, it may be possible to produce ultrashort superradiant pulses of large peak intensity.[26] The results of this work should also be applicable to x-ray lasers[27] where, owing to the lack of suitable reflective surfaces, feedback is absent and one must depend on single-pass gain. Superradiant emission should also be observable in spin-lattice systems.[28,29]

In an extremely long (or dense) sample, features of the superradiant output may be modified. The maximum spatial extent of a system which can superradiate as a whole, and the minimum duration of its output pulses, are limited by effects such as finite transit time, diffraction, and the nature and duration of the excitation process. These effects are discussed in another paper[26]; they are insignificant in the case of optically pumped HF gas at mTorr pressures in samples shorter than ~10 m (about 10 times the sample length of the experiments).

The remainder of this paper contains the following sections: II, Experimental Observation of Superradiance. III, Physical Principles. IV, Theoretical Model with Polarization Source, with subsections: A, Introduction; B, Coupled Maxwell-Schrödinger equations; C, Polarization source; D, Blackbody radiation and "equivalent input field." V, Numerical Results. VI, Simplified Theory. VII, Connections with Other Work.

Lengthy mathematical discussions have been placed in the appendixes.

II. EXPERIMENTAL OBSERVATION OF SUPERRADIANCE

In the experiments[6,14,15] a long sample cell of low-pressure HF gas is pumped by a short intense pulse from an HF laser operating on a single R or P branch transition between the $v=0$ and 1 vibrational levels (Fig. 1). This produces a nearly complete population inversion between two adjacent rotational levels in the first excited vibrational state, corresponding to a transition in the 50–250-μm range. The infrared radiation from this coupled transition (which is not at the same frequency as the pump transition) is studied as a function of time. There are no mirrors, and care is taken to minimize feedback. A detailed description of the experiments and their comparison with theory is given in Ref. 15.

An example of the observed output intensity is shown in Fig. 2(c). After a considerable delay (~1–2 μsec) with respect to the ~100-nsec pump pulse [Fig. 2(a)], radiation is emitted in a series of intense, short (~100 nsec) bursts of diminishing

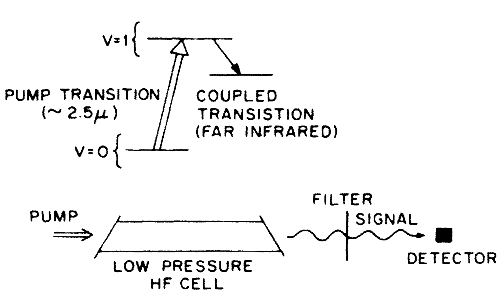

FIG. 1. HF level scheme and schematic of experimental setup.

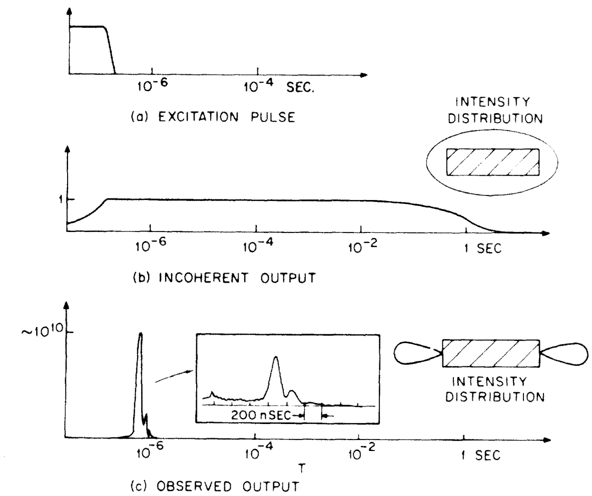

FIG. 2. Comparison of observed output and incoherent spontaneous emission. Time is plotted on a logarithmic scale. (a) Population-inverting laser pulse. (b) Output expected from incoherent spontaneous emission, exhibiting exponential decay and an isotropic radiation pattern. (c) Observed output, exhibiting ringing, a highly directional radiation pattern, and a peak intensity of ~ 10^{10} times that of (b). The inset shows the time evolution of the same pulse with a linear time scale.

size ("ringing"); the process is completed within a few μsec. The radiation pattern is highly directional; almost all of the radiation is emitted into a very small angle along the axis of the pump beam.

If the radiation emitted by this system were incoherent spontaneous emission, then it would have a long exponential decay (the radiative lifetime of these transitions is in the 1–10-sec range), and the radiation pattern would be isotropic [Fig. 2(b)]. Furthermore, the observed peak intensity is ten orders of magnitude greater than that expected for incoherent radiation. It is therefore clear that the process observed is not incoherent spontaneous emission. It is also clear that the output signal is not "amplified spontaneous emission"[30-32] in the usual sense, since the pulse evolution time is much longer than the time over which population inversion occurs.

The observed radiation is also distinct from that of an "ordinary" high-gain laser, even one which can oscillate without mirrors, such as a saturated-molecular-nitrogen laser (3300 Å).[33,34] In the nitrogen system the peak output intensity is directly proportional to the total population difference between the levels of the laser transition. Therefore, when the length or pressure is increased, the peak intensity increases proportionally. In contrast, the peak intensity of the observed output pulses in HF is proportional to the square of the pressure.[14,15] This proportionality is in agreement with the theoretical conclusions illustrated in Fig. 4 below.

The observed output radiation is therefore distinct from both incoherent emission and normal laser radiation. The N^2 dependence and the directionality of the observed radiation agree with the predictions of Dicke[1,21] for the behavior of a superradiant system (Sec. III). The present analysis of a superradiant system is based on the semiclassical formalism[35] which, as shown below, predicts the same N^2 dependence and directionality as Dicke's analysis. The semiclassical formalism makes possible a detailed description of the time evolution of the system and allows one to establish the specific conditions under which superradiance can occur in an extended sample. Furthermore, in the special case of "limited superradiance" described below, the results of the present analysis reduce to those obtained in previous theoretical treatments of superradiance.[11,12,17] The interpretation of the present experiment as the observation of superradiance is based on the connections of the present theory with previous treatments of Dicke and others, and the agreement of this theory with our experiments.

III. PHYSICAL PRINCIPLES

In his original treatment, Dicke[1] considered the radiative decay of an assembly of molecules for both a point sample (where the wavelength of emitted radiation λ is much greater than the sample length L) and an extended medium ($L \gg \lambda$). In a point sample,[36] by treating the entire collection of N molecules as a single quantum-mechanical system, he found that an initially totally inverted system will evolve into a "superradiant" state whose intensity is $N^2/4$ times the intensity radiated by a single molecule. This is larger by a factor of $N/4$ than the intensity radiated by N incoherent molecules.

For an extended medium, Dicke[1] showed that an array of quantum-mechanical dipoles phased along an axis could also produce superradiant emission.[36] Most of the radiation from the array is emitted into a small solid angle along the axis. For systems in which the Fresnel number $2A/\lambda L \gg 1$ ("disk"), the fraction of solid angle within which the radiation adds coherently is[11]

$$f = \Delta\Omega/4\pi = \lambda^2/4\pi A \,, \tag{1}$$

where A is the cross-sectional area of the sample. In our HF system, where the Fresnel number is of order unity, the correct formula[11] gives a value only slightly different from that of Eq. (1), and $f \sim 10^{-4}$. The output radiation intensity of the array of dipoles is larger than the radiated intensity of N incoherent dipoles by a factor $Nf \sim 10^8$.

The power I radiated by this array of N dipoles can be written in the form[15]

$$I = N(\tfrac{3}{4}\hbar\omega/T_R) \,, \tag{2}$$

where T_R is the characteristic radiation damping time of the collective system,[37]

$$T_R = T_{sp}(8\pi/n\lambda^2 L) \,, \tag{3}$$

$n = N/AL$, and T_{sp} is the lifetime of an isolated molecular dipole. Therefore, the emitted power is proportional to $N^2 f$.

In an extended molecular sample which is evolving to a superradiant state, a macroscopic polarization is established over a region of space. This polarization is equivalent to a phased array of dipoles which can be represented quantum mechanically as coherent mixtures of the stationary states of the molecules.[21] In the case of a ($J_{\text{upper}} = J+1$) $\to$ ($J_{\text{lower}} = J$) rotational transition of a diatomic molecule such as HF, T_{sp} is given by

$$\hbar\omega_0/T_{sp} = \tfrac{4}{3}\,|\mu|^2\omega_0^4/c^3 \,, \tag{4}$$

where[38]

$$|\mu|^2 = \mu_0^2 \frac{J+1}{2J+3} \,, \tag{5}$$

and μ_0 is the dipole moment of the molecule. Combining Eqs. (3) and (4), we have

$$T_R^{-1} = \frac{2\pi}{\lambda}\,\frac{2\pi}{\hbar}\,\mu_z^2 n_T L \,, \tag{6}$$

where $n(M)$ is the population inversion density between the states $|J_{\text{upper}}, M\rangle$ and $|J_{\text{lower}}, M\rangle$, n_T is the sum of $n(M)$ over all M levels in J_{upper}, and

$$\mu_z^2 \equiv \langle |\mu_z(M)|^2 \rangle_{M_{\text{lower}}} = \sum_{M=-J}^{J} \frac{|\mu_z(M)|^2 n(M)}{n_T} \,, \tag{7}$$

where

$$\mu_z(M) \equiv \mu_z[(J+1, M) \to (J, M)] \,. \tag{8}$$

In our HF case,

$$\mu_z^2 = \frac{\mu_0^2}{3}\,\frac{J+1}{2J+3} \tag{9}$$

when the excitation pulse is a P branch transition, and

$$\mu_z^2 = \frac{\mu_0^2}{3}\,\frac{J+1}{2J+1} \tag{10}$$

for an R branch excitation pulse.

Equation (2) shows that when the sample radiates as a collective system, the lifetime decreases from T_{sp} to $\sim T_R$. T_R can therefore be interpreted as a characteristic radiation damping time of the collective system. For the HF system, $T_{sp} \sim 1$ sec and $n \sim 10^{11}-10^{12}$ cm^{-3}, so $T_R \sim 10^{-8}$ sec. In order for the system to decay by the collective mode rather than as independent radiators it is necessary that $T_R \ll T_{sp}$. For a "disk" this requires that $n\lambda^2 L \gg 1$; i.e., there must be a large number of molecules in a "diffraction volume" $\lambda^2 L = (\lambda^2/A)AL$. This requirement also insures that there are many molecules in a cylinder of cross section λ^2 and length L, so that a coherent wave front can be properly reconstructed.

Consider next the process by which a macroscopic polarization builds up in an initially inverted system (the two adjacent rotational levels of interest in the HF $v=1$ level). In the Bloch formalism[39] for a point sample, a totally inverted system corresponds to a vector pointing straight up, analogous to a rigid pendulum balanced exactly on end. Similarly, our initially inverted extended medium corresponds to a spatial distribution of Bloch vectors all pointing straight up. The individual Bloch vectors of the extended medium, which are coupled together via the common radiation field, may evolve differently owing to propagation effects.

Just as the pendulum is unstable to small fluctuations, so the excited molecular system is unstable to a small perturbing field, initiated by spontaneous emission from one of the excited molecules or by background thermal radiation. This weak propagating electric field induces a small macroscopic polarization in the medium, which acts as a source to create additional electric field in the medium which, in turn, produces more polarization. This regenerative process gives rise to a growing electric field and an increasing polarization throughout the medium [Fig. 3(a)]. Therefore, a superradiant state slowly evolves over a sizable portion of the sample cell.[41] This state corresponds to the Bloch vectors pointing sideways over a sizable region of space, at which time radiation is emitted at a greatly enhanced rate. This process leads to a rapid deexcitation of that region of the medium, after which essentially all of the population is in the lower level; i.e., the Bloch vectors all point downward. Deexcited regions can then be reexcited by radiation from other regions, which gives rise to the "ringing" observed in the output radiation.

Figure 3 plots the polarization envelope $\mathcal{P}$ and the population inversion density n, respectively, throughout the sample at several instants of time. These values have been calculated from a theoretical model (developed in the following sections) based on the intuitive picture presented above. Notice that $\mathcal{P}$ and n vary slowly in space and time throughout the medium, giving rise to several regions of locally uniform polarization. These

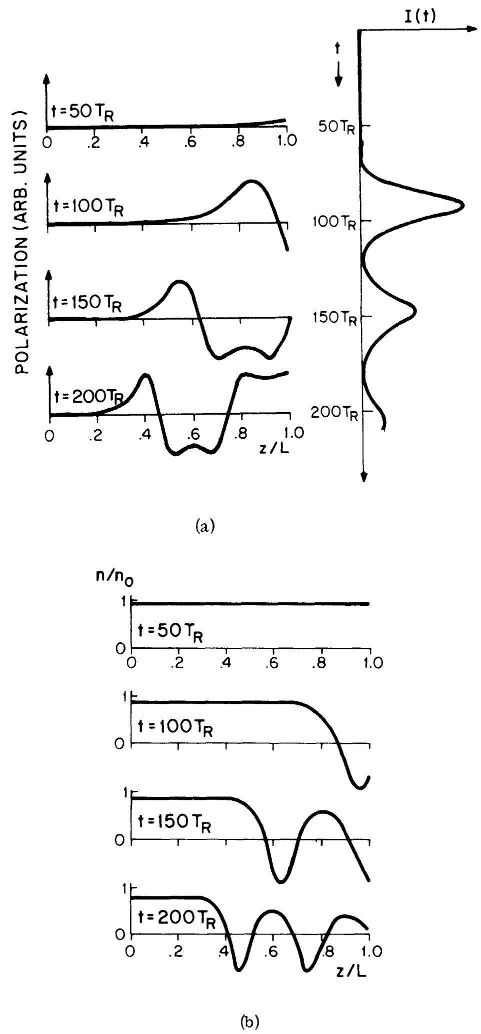

FIG. 3. Sketch of (a) the polarization envelope $\mathcal{P}$ and (b) the population inversion density n in the medium as functions of x at $t=50T_R$, $100T_R$, $150T_R$, and $200T_R$. The corresponding output intensity pattern is shown at the right of (a). The double peaks in $\mathcal{P}(x)$ occur whenever the ringing is sufficiently large (Ref. 40).

spatial variations in $\mathcal{P}$ and n are due to propagation effects in a high-gain medium, a crucial point which was not appreciated in some earlier work. The ringing in the output radiation (Fig. 3) is a direct consequence of these spatial variations (see Sec. VI).

The time evolution of the radiation emitted by an initially inverted system depends on many factors, including broadening, diffraction loss, and level degeneracy. However, as shown in Sec. V, in a high-gain system the major features of the output radiation pulse are determined by T_R and the logarithm of θ_0 (described below), and a normalized curve can be drawn which gives the output intensity $I(T)$ multiplied by T_R^2 as a function of time in units of T_R (T is the retarded time at the end of the sample). This curve (Fig. 4) depicts a burst of radiation with ringing preceded by a long delay. The scaling of the curve is such that when T_R is halved, the system radiates twice as fast and the peak intensity is quadrupled. This curve exhibits the N^2 intensity dependence which is characteristic of superradiant emission, since the peak intensity is proportional to T_R^{-2} and T_R is proportional to N^{-1}. Experimental data exhibiting this scaling are given in Ref. 15.

The shape of this curve depends on θ_0, a parameter which is a measure of the conditions which initiate the superradiant pulse [Eq. (20) below]. Whenever $\theta_0 \ll 1$, the delay time T_D from the time of inversion to the peak of the first lobe of emitted radiation is much greater than T_R. This long delay time may be understood by considering a pendulum which is initially balanced exactly on end. The time required for the pendulum to fall following a small perturbation can be much longer than the oscillation period. In an analogous manner, the time necessary for the completely inverted system to develop a macroscopic polarization can be much longer than the collective radiation time T_R. This point will be discussed in Sec. VI.

In the experiments, superradiant pulses are observed simultaneously in both the forward and backward directions with respect to the population-inverting laser pulse. This comes about because in the HF system the transit time through the sample cell is short compared to the time needed for the system to superradiate ($\sim T_D$), so that the electromagnetic waves can grow in both directions simultaneously without interacting appreciably during most of the pulse evolution time. (Long transit times and different excitation configurations are discussed in Sec. VI and Ref. 26.) This is so because in order for the waves to interact, either (i) they must become large in the same region of the medium at the same time, so that the coherent interactions which couple the forward and backward waves become important, or (ii) the population depletion by one wave must affect the subsequent growth of the other wave. The former effect is negligible because the buildup of polarization of a given wave is confined to one end of the medium until well after the main pulse has already been emitted [$\sim 150\,T_R$, Fig. 3(a)]. The latter effect is negligible because each wave depletes the population inversion primarily at one end of the medium [Fig. 3(b)]. An iterative computer solution using the theoretical model shows that the growth of each wave is almost completely unaffected by the population depletion caused by the other wave. We therefore conclude that for systems with small transit time the two waves will radiate essentially independently without significant correlation, and henceforth, we will only deal with the forward traveling wave. A discussion of the interaction of forward and backward waves in systems with long transit times will be found in Ref. 26.

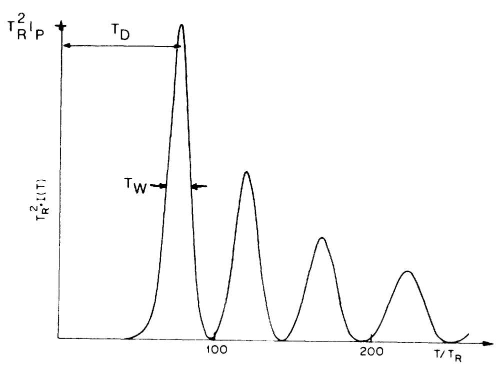

FIG. 4. Normalized output curve. This curve is the output response to a small rectangular input pulse of area θ_0 in a nondegenerate system where $T_2 = T_2^* = \infty$ and $\kappa L = 0$. The time scales as T_R and the intensity scales as T_R^{-2}. Note that the shape of the normalized output curve depends on θ_0. I_p, T_D, and T_W can all be expressed in terms of T_R and θ_0 (see Sec. VI).

IV. THEORETICAL MODEL WITH POLARIZATION SOURCE

A. Introduction

In numerous treatment of superradiance the radiation field is quantized and the molecular system is described in terms of collective Dicke states.[7-12,17-20] As pointed out by Arecchi, Courtens, Gilmore, and Thomas,[35] these states can be used to construct a new set of states, Bloch states, which also describe superradiant ensembles. These new states can be treated by means of the semiclassical formalism (coupled Maxwell-Schrödinger equations), in which the molecular system is quantized but the electromagnetic field is treated classically.

One shortcoming of the Maxwell-Schrödinger equations as they are usually written[42] is that there is no mechanism for spontaneous emission, so that an initially inverted system (such as in the HF experiment) cannot evolve.[43] This problem can be overcome by adding a phenomenological fluctuating polarization source term to simulate the effect of spontaneous emission. This term can be constructed to be consistent with requirements of thermal equilibrium as well as energy and number conservation.

Let us consider in more detail the initial stage of the evolution of an inverted system to a superradiant state. At first the system undergoes spontaneous emission in various directions at random times. Eventually a photon is emitted along the axis of the sample cell, leading to the excitation of one of the modes of the inverted medium. In the case of large Fresnel number (disk) this mode subtends a solid angle $\Delta\Omega = \lambda^2/A$ [Eq. (1)], determined by diffraction. The average number of photons emitted into this mode in a time t is equal to $(N/T_{sp})(\lambda^2/4\pi A)t = 2t/T_R$, so that the first photon is emitted into the diffraction mode in a time of order T_R.[43] Therefore, at times greater than T_R there will be many photons per mode, so that the radiation field can be treated classically.

Note that up to time $\sim T_R$, when the medium is starting to evolve, the number of photons per mode is small and one can question the validity of the semiclassical model with polarization source term included. However, in the present problem, in which an initially unstable system is subjected to small perturbations, this model is a good approximation. To justify this conclusion, we note the following:

(i) Computer results (Sec. V) obtained from this model show that the superradiant output of the initially inverted system is insensitive to the specific form of the source fluctuations, depending only on their sum over a time $\sim T_R$. This is easy to understand in terms of the pendulum analogy presented previously: For a system in an unstable initial state, the response to a rapid series of small destabilizing forces depends only on the sum of these forces over a period of time (here, $\sim T_R$) and not on the form of the individual forces.

(ii) The numerical results also show that the contribution of the polarization source term to the evolution of the system becomes negligible for times larger than $\sim T_R$. This can be understood by using Eq. (47) below to show that the time necessary for the output resulting from a small input pulse to become much larger than the input pulse itself is of order T_R. Therefore, for times greater than T_R, the input pulse and, accordingly, the polarization source which gives rise to it, no longer significantly influences the output behavior. (It will be shown in Sec. IV D that the polarization source of this model can be accurately approximated by an equivalent input pulse.)

Since the output radiation becomes independent of the polarization source after a time $\sim T_R$ following the inversion of the medium, phase fluctuations associated with spontaneous emission at subsequent times will have little effect on the output radiation. In other words, the first photon emitted into the diffraction mode initiates the evolution of the system, and phase fluctuations associated with subsequent spontaneous-emission events are unimportant. In effect, the first photon determines the phase of the output radiation.

(iii) The delay time between the inversion of the medium and the peak of the superradiant output pulse, which is of the order of $100\,T_R$ in the HF experiments [see Eq. (49)], is not sensitive to the fluctuations of the spontaneous emission which initiates the superradiant process. This is so because the fluctuation time before the first photon is emitted into the diffraction mode is of order T_R. This time is only $\sim 1\%$ of the delay time in HF, and can be ignored in calculating the delay time.

This explains why the semiclassical formalism with a fluctuating polarization source included gives a good description of the evolution of a superradiant state in an initially inverted medium.

B. Coupled Maxwell-Schrödinger equations

Adopting the semiclassical formalism, we consider a gas of two-level molecules interacting with an electromagnetic wave. As in the discussion following Eq. (1), we confine our attention to a system of large Fresnel number (disk-shaped system) where the electromagnetic field can be approximated by a plane wave. In this limit the two-level system is described by the coupled Maxwell-Schrödinger equations given by many authors.[22-24,44,45] We shall adopt notation similar to that of Icsevgi and Lamb,[24] extended to include level degeneracy, as is necessary in treating rotational transitions of a molecular system, and the polarization source described previously. The coupled equations in the slowly-varying-envelope approximation, written in complex form, are

$$\frac{\partial \mathcal{E}}{\partial x} = -\kappa\mathcal{E} + 2\pi k \sum_{v,M} \mathcal{P}\,, \tag{11a}$$

$$\frac{\partial \mathcal{P}}{\partial T} = -(\gamma - ikv)\mathcal{P} + (\mu_z^2)_M \mathcal{E} n/\hbar + \Lambda_p\,, \tag{11b}$$

$$\frac{\partial n}{\partial T} = \Lambda - \gamma n - (1/\hbar)\,\mathrm{Re}\,(\mathcal{E}\mathcal{P}^*)\,. \tag{11c}$$

Here $\mathcal{E}(x,T)$ and $\mathcal{P}(x,T,v,M)$ are the slowly varying envelopes of the electric field $\vec{E}(\vec{r},t)$ and the polarization density per velocity interval dv, $\vec{P}(\vec{r},t,v,M)$, respectively. They are defined by

$$\vec{E}(\vec{r},t) = \hat{z}E(x,t) = \hat{z}\,\mathrm{Re}\,[\mathcal{E}(x,T)\,e^{i(\omega_0 t - kx)}]\,, \tag{12}$$

$$\vec{P}(\vec{r},t,v,M) = \hat{z}P(x,t,v,M) = \hat{z}\,\mathrm{Re}\,[i\mathcal{P}(x,T,v,M)\,e^{i(\omega_0 t - kx)}]\,, \tag{13}$$

at position x, time t, and retarded time $T = t - x/c$. The carrier frequency is assumed to be the molecular center frequency ω_0 with no loss of generality. Note that $\mathcal{E}$ and $\mathcal{P}$ are complex, and that $\mathcal{E}(x,T) = |\mathcal{E}(x,T)|\,e^{i\xi}$ where $\xi(x,T)$ is a slowly varying real phase. In these equations, $n(x,T,v,M)$

$=n_2-n_1$, where n_2 and n_1 are the number densities per velocity interval dv of molecules in the upper and lower states (J,M) of the superradiant transition, respectively; κ is a loss term which accounts for diffraction (see Sec. V), which can be a function of x but not of $\mathcal{E}$; and γ is the decay rate of n_1, n_2, and $\mathcal{P}$, assumed equal for simplicity (i.e., $T_1=T_2=\gamma^{-1}$).[46] $\Lambda=\lambda(z,T,M)W(v)$ is a source term describing the rate of production of n, due to optical pumping in our case, where $W(v)$ is the distribution of molecular velocities, normalized to unity [Eq. (15)]. $\Lambda_p(x,T,v,M)$ is the polarization source term describing the rate of production of $\mathcal{P}$ due to spontaneous emission. The exact form is discussed in Sec. IV C. The symbol $\sum_{v,M}$ denotes an integral over the velocity distribution and a sum over degenerate M states of the rotational levels:

$$\sum_{v,M} f(v,M)=\int_{-\infty}^{\infty} dv \sum_{M=-J}^{J} f(v,M). \tag{14}$$

In the experiments the optical pumping process produces excited molecules in the upper level of an initially unpopulated two-level system (Fig. 1) with a Lorentzian velocity distribution,[47]

$$W(v)=u_1/\pi(v^2+u_1^2), \tag{15}$$

where

$$u_1 \approx \mu_p \mathcal{E}_p/\hbar k_p. \tag{16}$$

Here k_p is the wave number and μ_p the matrix element of the pump transition, and $\mathcal{E}_p$ is the amplitude of the intense pump field. In HF,[14,15] $u_1 \sim 10^4$ cm/sec. The total number density of excited molecules in all M_J states is then (for R branch pumping)

$$n_T=\sum_{v,M} n(v,M), \tag{17}$$

$$n_{T0}=n_T(t=0)=\frac{1}{2}\left(\frac{\mu_p\mathcal{E}_p/\hbar}{k_p u/\sqrt{\pi}}\right)n_G, \tag{18}$$

where n_G is the total number of molecules in the rotational level (of the $v=0$ vibrational state) which is being pumped.[48]

Although the growth of the electric field in the medium does not follow simple exponential gain, the time integral of the electric field obeys an exponential law, the area theorem.[44] This states that for a nondegenerate Doppler-broadened collisionless system subjected to an incident field $\mathcal{E}(x=0,t)$ of constant phase, the pulse area $\theta(x)$ obeys the equation

$$\tan\tfrac{1}{2}\theta(x)=(\tan\tfrac{1}{2}\theta_0)\,e^{\alpha_0 x}, \tag{19}$$

where

$$\theta(x)\equiv\frac{\mu_z}{\hbar}\int_{-\infty}^{\infty}\mathcal{E}(x,t)\,dt, \tag{20}$$

$\theta_0=\theta(x=0)$, and α_0 is the small signal field gain at the molecular center frequency ω_0. For a system whose linewidth contains both homogeneous and inhomogeneous contributions,

$$\alpha_0 L=2\pi k(\mu_z^2/\hbar)n_{T0}LT_2', \tag{21}$$

where n_{T0} is the initial total inversion density. In the Doppler-broadened limit, which applies in the HF system, the characteristic broadening time $T_2'\approx T_2^*=1/ku_1$. [Note that in our case, ku_1 is an effective Doppler width, given by Eq. (16) above.] Combining Eqs. (6) and (21) gives the relationship[49]

$$\alpha_0 L=T_2'/T_R, \tag{22}$$

so that $\alpha_0 L$ is independent of the pump field intensity I.

Equation (19) shows that in a high-gain medium the area of a pulse of small initial area begins to grow exponentially,

$$\theta(x)=\theta_0 e^{\alpha_0 x}, \tag{23}$$

but then evolves towards π. In this process the field envelope may develop positive and negative lobes (ringing) whose contributions to the area substantially cancel one another. Accordingly, in a high-gain system the pulse energy continues to grow even though the area remains constant.

The presence of level degeneracy does not invalidate this conclusion, although level degeneracy can inhibit pulse propagation in an absorber. The considerations of Rhodes, Szöke, and Javan[50] applied to an amplifier show that level degeneracy does not prevent pulse formation, and that pulses of increasing energy and area near π can evolve.

High gain ($\alpha_0 L \gg 1$) is necessary for superradiance to occur: This condition is equivalent [Eq. (22)] to $T_R \ll T_2'$, which is necessary so that collective radiation can occur rapidly with respect to incoherent decay. The requirement $\alpha_0 L \gg 1$ also ensures that $\alpha_0 L \gtrsim |\ln\theta_0|$, so that a π pulse will evolve [Eq. (19)]. The high-gain condition will be discussed further in Sec. VII.

C. Polarization source

As discussed earlier, the superradiant evolution process is initiated by spontaneous emission from the excited molecules and by background thermal radiation. In our model spontaneous emission is simulated by a randomly phased polarization source term Λ_p which is distributed throughout the medium. As is shown below, although Λ_p is essential for initiating the growth of $\mathcal{E}$, as the $\mathcal{E}$ field evolves in a high-gain system the influence of Λ_p soon becomes unimportant. Therefore, to calculate the amplitude of Λ_p for a high-gain system, we consider the contribution of Λ_p to the growth of $\mathcal{E}$ only during the first stage of pulse evolution, when $\mathcal{E}$ is small and the field produced by Λ_p is sizable compared to the field already present.

The amplitude of Λ_p will be calculated in the following manner: First, starting from the coupled Maxwell-Schrödinger equations, specialized to the case of weak field and steady state, we obtain an equation for the intensity $I(x)$ as a function of the amplitude of Λ_p. This equation is then compared to an intensity equation derived from thermal equilibrium considerations which also holds for linear steady-state systems. Since these two equations are of the same form, we can evaluate the amplitude of Λ_p in terms of the molecular parameters of the system. The resulting expression for Λ_p, which is not a function of dynamical variables, can be used in the coupled Maxwell-Schrödinger equations to describe a system not at thermal equilibrium.

We consider the medium to be divided into small regions of volume $A\Delta x$ ($\Delta x \ll L$), and time into intervals $\Delta t < T_R$. Since the spontaneous-emission intensity is proportional to $n_2(x,t)$ and otherwise depends only on molecular constants (except for a linewidth-narrowing effect in high-gain systems, described below), Λ_p is proportional to $(n_2)^{1/2}$. We choose the form of Λ_p to be a series of pulses in space and time with constant amplitude[51] (except for the n_2 dependence) and random phases.[52] It is convenient to evaluate Λ_p at points separated by Δx in space and ΔT in retarded time. Then Λ_p will be of the form

$$\Lambda_p(x,T,v,M)=B_1[n_2(x,T,v,M)]^{1/2}\sum_{j=1}^{j'}\delta(x-x_j)\sum_{l=-\infty}^{\infty}\delta(T-T_l)\sum_{k=1}^{k'}\delta(v-v_k)\,e^{i\phi_{jlkM}}, \tag{24}$$

where B_1 is a (real) constant to be determined, $x_j=(j-\frac{1}{2})\Delta x$, $\Delta x=L/j$, $T_l=l\Delta T$, v_k are k' discrete velocities,[53] and ϕ_{jlkM} are independent random phases.

We consider the case where n and $\mathcal{E}$ are independent, $\mathcal{E}$ is very slowly varying in time ($\partial\mathcal{E}/\partial T \ll \gamma\mathcal{E}$),[54] and n is near a constant value n_0 and is very slowly varying in space and time ($\partial n/\partial T \ll \gamma n$, $\partial n/\partial x \ll \gamma n/c$).[55] Such a system is linear and nearly steady state ($n \sim n_0$). As shown in Appendix A, the coupled Maxwell-Schrödinger equations [Eqs. (11a) and (11b)] for such a system may be integrated to give an intensity gain equation

$$I(x)=I(x=0)\,e^{2\alpha_0 x}+K_0(e^{2\alpha_0 x}-1), \tag{25}$$

where α_0 is the gain of the system at the molecular center frequency ω_0, $I(x)=cA\,|\mathcal{E}(x)|^2/8\pi$ is the power at position x, and

$$K_0=\frac{\pi}{4}\frac{\omega_0^2}{c}AB_s^2\frac{T_2 n_2}{\Delta T}(1-e^{-2\alpha_0\Delta x})^{-1}. \tag{26}$$

Equation (25) holds for any linear steady-state system, independent of the sign and magnitude of the gain.

We require that Eq. (25) be consistent with the Einstein intensity equation,[56,57] which states that throughout the active region of any linear steady-state medium, the total energy per mode $I_\omega(x)$ must satisfy the equation

$$\frac{dI_\omega(x)}{dx}=2\alpha(\omega)\left(I_\omega(x)+\frac{\hbar\omega n_2}{n_2-n_1}\right), \tag{27}$$

where $\alpha(\omega)$ is the gain at frequency ω. [Note that Eq. (27) is consistent with the requirements of thermal equilibrium, since when $I_\omega(x)=\hbar\omega/(e^{\hbar\omega/kT}-1)$ and $n_2=n_1e^{-\hbar\omega/kT}$, Eq. (27) simplifies to $dI_\omega(x)/dx=0$.] To compare Eq. (27) with Eq. (25), Eq. (27) must be rewritten in terms of the power per unit frequency interval $I(\omega,x)$ in the plane-wave direction. As shown in Appendix B, for a system with Fresnel number not signficantly less than unity, Eq. (27) becomes

$$\frac{\partial I(\omega,x)}{\partial x}=2\alpha(\omega)\left(I(\omega,x)+\frac{\hbar\omega}{4\pi}\frac{n_2}{n_2-n_1}\right). \tag{28}$$

The integration of Eq. (28) in a high-gain ($\alpha_0 L \gg 1$) linear steady-state system with finite bandwidth is performed in Appendix C. In such a system the linewidth of $I(\omega,x)$ decreases as $\alpha_0 x$ increases ("gain narrowing"). For a high-gain system interacting with a broadband field of input bandwidth $\Delta\omega \gg (T_2')^{-1}(\pi/2\alpha_0 L)^{1/2}$ centered at frequency ω_0 (where $1/T_2'$ is the bandwidth of the gain profile), the total output power $I(x=L)$ can be obtained by integrating Eq. (28) as in Appendix C:

$$\begin{aligned}I(x=L)&=\int I(\omega,x=L)\,d\omega\\&\approx\left(I(\omega_0,x=0)+\frac{\hbar\omega}{4\pi}\frac{n_2}{n_2-n_1}\right)(T_2')^{-1}\\&\quad\times\left(\frac{\pi}{2\alpha_0 L}\right)^{1/2}e^{2\alpha_0 L}\,.\end{aligned} \tag{29}$$

We can compare this result with the high-gain limit ($e^{2\alpha_0 x} \gg 1$) of Eq. (25) to obtain[58]

$$I(x=0)=I(\omega_0, x=0)\,(T_2')^{-1}(\pi/2\alpha_0 L)^{1/2} \tag{30}$$

and

$$K_0=\frac{n_2}{n_2-n_1}(T_2')^{-1}\left(\frac{\pi}{2\alpha_0 L}\right)^{1/2}\frac{\hbar\omega_0}{4\pi}. \tag{31}$$

Equation (30) tells us what the input power $I(x=0)$ in the intensity gain equation [Eq. (25)] must be for a high-gain system in order to be consistent with the Einstein equation. Equation (30) can be rewritten in the form

$$I(x=0)=I(\omega_0, x=0)\,(\Delta\omega)_{\text{eff}}, \tag{32}$$

where $(\Delta\omega)_{\text{eff}} \equiv 1/(T_2')_{\text{eff}}=(T_2')^{-1}(\pi/2\alpha_0 L)^{1/2}$ is the mathematically derived effective bandwidth of the input radiation which interacts with the gain profile of the high-gain system. This gain-narrowing effect can be explained physically by noting that in the early stages of growth, a high-gain system preferentially amplifies the central portion of the frequency profile.

Inserting Eq. (31) into (26), we obtain B_1, and if Δx is chosen such that $\alpha_0\Delta x \lesssim 1$ (as in the case of our computer program), Eq. (24) becomes[55]

$$\Lambda_p(x,T,v,M) = \left(\frac{4\Delta T}{T_2'}\frac{\Delta x}{L}\frac{n_2 AL}{(2\pi\alpha_0 L)^{1/2}}\right)^{1/2}\left(\frac{\mu_z}{A}\right) \times\sum_j\sum_l\sum_k \delta(x-x_j)\,\delta(T-T_l)\,\delta(v-v_k)\,e^{i\phi_{jlkM}}. \tag{33}$$

Note that Λ_p is proportional to μ_z and depends on the total number of excited molecules $n_2 A\Delta x\Delta T$ associated with one point in the space-time grid; the square-root dependence occurs because the radiation from independent spontaneous-emission events adds incoherently.

D. Blackbody radiation and "equivalent input field"

In the computer model background thermal radiation is simulated by an input field of randomly fluctuating phase, the intensity of which depends on the bandwidth and the solid angle of the input radiation which interacts with the system. The result of Appendix B can be used to convert the blackbody power per mode $I_\omega(x=0)=\hbar\omega/(e^{\hbar\omega/kT}-1)$ to the plane-wave blackbody power per unit frequency interval $I(\omega,x=0)=(\hbar\omega/4\pi)/(e^{\hbar\omega/kT}-1)$. As described above, the effective bandwidth of radiation which interacts with a high-gain system is $\Delta\omega_{\text{eff}}=(\pi/2\alpha_0 L)^{1/2}/T_2'$ (see Appendix C), so that the total input power which must be used in order to correctly apply Eq. (25) to this case is

$$\frac{cA}{8\pi}|\mathcal{E}(x=0,T)|^2=\frac{\hbar\omega_0}{4\pi}\frac{\Delta\omega_{\text{eff}}}{e^{\hbar\omega_0/kT}-1} \tag{34}$$

or, equivalently,

$$\frac{|\mathcal{E}|^2}{8\pi}=\frac{\hbar\omega_0}{e^{\hbar\omega_0/kT}-1}\frac{2\pi(\omega_0/2\pi)^2}{c^3} \times\frac{\lambda^2}{4\pi A}\frac{ku_1}{2\pi(2\alpha_0 L/\pi)^{1/2}}. \tag{35}$$

The factors on the right-hand side of Eq. (35) are, respectively, the background thermal radiation energy per mode, the number of modes of one polarization per unit value per frequency interval $\Delta\nu$, the solid angle factor f, and the gain-dependent effective bandwidth (in units of $\Delta\nu$). Using this value of $|\mathcal{E}(x=0,T)|$ as an input boundary condition, and using Eq. (33) for Λ_p, Eqs. (11) can be numerically integrated to obtain the output radiation intensity. Examples of these computer results are given in Figs. 3 and 5.

Alternatively, spontaneous emission in a high-gain system can be described by replacing the distributed polarization source by an "equivalent"

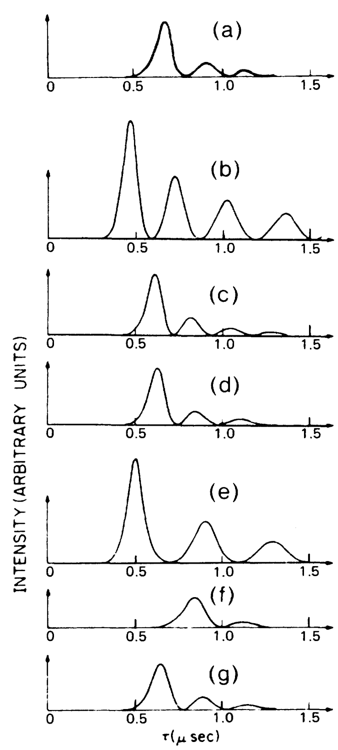

FIG. 5. Computer results showing the influence of parameters on output intensity. The same intensity scale is used throughout. (a) A theoretical fit with parameters T_R=6.1 nsec, T_2^*=330 nsec, T_2=5.4 μsec, κL=2.5, J_{lower}=2. All parameters have the same values as in this curve except when stated otherwise. (b) No level degeneracy, $T_2=T_2^*=\infty$, $\kappa L=0$. (c) $T_2=\infty$. (d) $T_2^*=\infty$. (e) $\kappa L=0$. (f) $\kappa L=5$. (g) No level degeneracy.

input electric field. The input field (of constant amplitude) which would produce the same output intensity as the distributed source (see Appendix C) for a high-gain system, which combines the contributions of blackbody radiation [Eq. (34)] and spontaneous emission, is[59]

$$\frac{cA}{8\pi}|\mathcal{E}_{\rm eff}(x=0,T)|^2 = \frac{\hbar\omega_0}{4\pi}\frac{ku_1}{(2\alpha_0 L/\pi)^{1/2}}\left(\frac{n_2}{n_2-n_1}+(e^{\hbar\omega_0/kT}-1)^{-1}\right) = \frac{\hbar\omega_0}{2T_R\sqrt{\pi}}\frac{1}{(2\alpha_0 L)^{3/2}}\left(\frac{n_2}{n_2-n_1}+(e^{\hbar\omega_0/kT}-1)^{-1}\right). \tag{36}$$

The validity of the "equivalent-input" approximation can be understood by noting that in a high-gain system the spontaneous emission which occurs near the input face is most important in initiating the superradiant evolution process. Although all of the output radiation curves in this paper (and in Ref. 15) were fitted using the distributed-polarization-source model, essentially identical computer curves are obtained using the equivalent-input-field approximation. This finding strongly supports the validity of the equivalent-field approach and confirms the interpretation and analaysis of Ref. 6.

Since the distributed source can be replaced by an effective input field and since phase fluctuations in the input field do not significantly affect the computer curves (as explained in Sec. IV A and verified by the computer results of Sec. V below), the results of the area theorem [Eq. (19)] may be used for a qualitative understanding of the behavior of the system.[60] This makes it possible to derive an effective input area θ_0, which can be used in the area theorem and for obtaining simple analytical expressions for the delay time, pulse width, and peak intensity (Sec. VI). Owing to the exponential nature of the growth of the area [Eq. (19)], the parameter which enters into these expressions is a logarithmic function of θ_0. Because $|\ln\theta_0|\gg 1$, the expressions are insensitive to the exact numerical coefficient of θ_0.

As stated earlier, the influence of the effective input field on the evolution of the superradiant state is only significant for a short time after the system is pumped into excitation, and since the output field more than doubles in a time T_R after excitation [Eq. (47)], the input field can be ignored after a time T_R. Therefore, to a good approximation,

$$\theta_0 \approx \mu\mathcal{E}_{\rm eff}\, T_R/\hbar \tag{37}$$

can be chosen to be the area of the effective input pulse. Combining Eqs. (3), (4), and (36) with this expression yields[59]

$$\theta_0^2 \approx \frac{N_2^{-1}}{\sqrt{2\pi}}(\alpha_0 L)^{-3/2}\left(\frac{n_2}{n_2-n_1}+\frac{1}{e^{\hbar\omega_0/kT}-1}\right). \tag{38}$$

For our system, $\theta_0 \sim 10^{-8}$, and in order for the output to evolve to a pulse of area $\sim\pi$, $\alpha_0 L$ [Eq. (19)] must be $\gtrsim 20$. This is readily achieved in the experiments.

V. NUMERICAL RESULTS

The coupled Maxwell-Schrödinger equations [Eqs. (11)] have been integrated by computer, with level degeneracy taken into account in pump and superradiant transitions. Computer-generated curves obtained using input conditions appropriate for the HF case[14,15] are in good agreement with experimental results. For example, decreasing the sample cell pressure or pump intensity reduces the output intensity and increases the delay time and the width of the output pulse. A detailed comparison of theory and experiment can be found in Ref. 15.

The following general features are evident from the numerical results, and confirm the qualitative statements of Secs. III and IV:

(i) The effects of relaxation are unimportant as long as the homogeneous relaxation time T_2 exceeds the pulse delay T_D. When T_2 becomes comparable to T_D the pulses are reduced in size and the ringing is cut down. At the mTorr pressures of the experiments T_2, determined by collisions, is always much longer than T_D.

(ii) The influence of the polarization source Λ_p is limited to the first few T_R's. In addition, the output is unchanged when the polarization source [Eq. (33)] is replaced by an input field whose amplitude is given by Eq. (36). This verifies the effective input-field approximation presented in Sec. IV D and the value of $\mathcal{E}_{\rm eff}$ given by Eq. (36).

(iii) The effect of a weak input field (to simulate blackbody radiation) is limited to the first few T_R's. The output is insensitive to the exact shape and phase of the input field. Delta function, step function, Gaussian pulses, and pulse trains of varying phase all give output pulses of about the same shape and size, as long as their input areas are equal. Also, the presence of random jumps in the phase of the input field has no significant effect on the output pulses.

(iv) The area of the output pulse is determined by the gain $\alpha_0 L$ and the size of the input pulse θ_0, in accordance with the area theorem [Eq. (19)]. Since in our case $\theta_0 \sim 10^{-8}$, $\alpha_0 L \gtrsim 20$ is needed for appreciable pulse buildup. Such gains are available in HF at mTorr pressure.

(v) The time scale of the pulse evolution process (i.e., delays, pulse shapes) is almost completely determined by T_R and θ_0, as long as $\alpha_0 L \gg 1$.

(vi) The output pulses are insensitive to the specific time dependence of the population excitation. This is true for excitation pulses of duration approaching the superradiant pulse delay. (Effects of longer excitation pulses are discussed in Ref. 26.)

(vii) Two forms of the loss coefficient κ [Eq. (11a)] might be important under different experimental conditions: the case of linear (constant) κ, as was used in Ref. 24 and our previous work[6,14]; and the case of diffraction loss, where the plane-wave $\mathcal{E}$ field can be modeled as a Gaussian beam, in which case $\kappa = x/(x^2+L_0^2)$, with $L_0 = A/\lambda$.[61] For the same value of the total loss (κL in the linear-loss case; $\int \kappa\, dx = \frac{1}{2}\ln[1+(\lambda L/A)^2]$ in the Gaussian-beam case), the two models give virtually identical output pulses.

(viii) Replacing each of the different matrix elements $(\mu_z)_M$ in a degenerate system by the average value μ_z defined by Eq. (7) does not significantly affect the behavior of the output pulse; i.e., the effects of level degeneracy are negligible.

(ix) The output intensity at a given superradiant transition depends on the pump transition branch used (P or R), in agreement with the experiments. This dependence is due to the different matrix elements and widely different populations of the ground-state levels selected.

In summary, as long as $\alpha_0 L \gg 1$, for a given T_R homogeneous and inhomogeneous broadening, level degeneracy, and moderate variations in the value of κL change the results in only minor ways. The weak influence of these effects on the output pulse can be seen in Fig. 5, which shows the changes in the pulse shape and delay caused by varying these parameters. The simple results obtainable by setting $1/T_2 = 1/T_2^* = \kappa = 0$ and neglecting level degeneracy may be used for a qualitative understanding of the behavior.[60] This simplified model gives the normalized emission curve of Fig. 4. (Other effects which occur in very long samples, but which are not relevant to the HF experiments, are discussed in Ref. 26.)

VI. SIMPLIFIED THEORY

As indicated by the computer analysis of Sec. V, most of the parameters of the system have very little effect on the superradiant output pulses (Fig. 5). If we set $\gamma = ku = \Lambda = \Lambda_p = \kappa = 0$ in Eqs. (11) and ignore the effects of level degeneracy, a set of simplified equations is obtained:

$$\frac{\partial \mathcal{E}}{\partial x} = -2\pi k S, \tag{39}$$

$$\frac{\partial S}{\partial T} = -\frac{\mu_z^2 n \mathcal{E}}{\hbar}, \tag{40}$$

$$\frac{\partial n}{\partial T} = \frac{\mathcal{E} S}{\hbar}. \tag{41}$$

Here, $S = -\mathcal{P}$ and n, $\mathcal{E}$, and S are all real. Note that in this simplified model, in which $\Lambda_p = 0$, the effect of spontaneous emission (which initiates the superradiant evolution process in an initially inverted system) can be included by means of the effective-input-field approximation. In the discussions of this section, an effective input pulse [as given by Eq. (38)] will be used as an initial condition.

Equations (40) and (41) have the solution

$$n = n_0 \cos\psi, \tag{42}$$

$$S = -\mu_z n_0 \sin\psi, \tag{43}$$

$$\frac{\partial \psi}{\partial T} = \frac{\mu_z \mathcal{E}}{\hbar}, \tag{44}$$

where

$$\psi(x,T) = \int_{-\infty}^{T} (\mu_z/\hbar)\, \mathcal{E}(x,T')\, dT' \tag{45}$$

is the "partial area" of the pulse, so that $\psi(x, T = \infty) = \theta(x)$. From Eqs. (42) and (43), it can be seen that ψ may also be interpreted in the geometrical representation[62] as the "tipping angle" of a Bloch vector[39] of length n_0 which evolves in the z-y [$n - (S/\mu_z)$] plane. Note [Sec. III] that an extended medium must be represented as a distribution of Bloch vectors whose spatial variations are determined by Eq. (39). The initially inverted system, corresponding to the Bloch vectors all standing on end ($n = n_0, S = 0$), gradually evolves into a superradiant state ($n = 0, S/\mu_z = -n_0$); the evolution of the individual Bloch vector at any point in space is described by Eqs. (42) and (43).

Combining Eqs. (39) and (44) gives an equation for $\psi(x,T)$[22,63]:

$$\frac{\partial^2 \psi}{\partial x\, \partial(T/T_R)} = \frac{\sin\psi}{L}. \tag{46}$$

This equation was studied for the case of an absorber by Burnham and Chiao[63] and others,[13,45] who showed that for a δ-function input field, the time dependence of $\psi(x,T)$ and therefore [Eq. (44)] of $T_R \mathcal{E}(x,T)$ was a function of T/T_R only. Our case is that of an amplifier, and the time dependence of the δ-function response is again a function of T/T_R only, from which follows the scaling property of the normalized emission curve of Fig. 4.[63a] Furthermore, since the effective input field is only important during the first $\sim T_R$, any input pulse of small effective area θ_0 gives the same response as a δ-function input pulse of the same area. [A more formal statement of the scaling properties of Fig. 4 is that the transformation $T \to T/T_R$, $\mathcal{E} \to T_R\mathcal{E}$ in Eqs. (39)–(41) leaves these equations unchanged and does not affect the area of the input pulse (the boundary condition).] This simple scaling property holds for "end-excited" systems in which the system is excited by a pulse propagating through the medium at velocity c, so that all molecules are excited at the same *retarded* time. It also holds for "side-excited" systems (in which all molecules are excited simultaneously) unless the transit time $L/c \gtrsim 2(T_R/4)[\ln(\theta_0/2\pi)]^2$.[26] In the latter case, spontaneous emission in different parts of the medium causes the sample to break into a number of independently superradiating segments as described by Arecchi and Courtens[13] (see Ref. 26 for further discussion).

The computer results also exhibit these scaling properties, even when all of the refinements of the theory are included (Fig. 5). The main requirement for the validity of the normalized emission curve is that T_R be much shorter than all other times associated with the system ($T_R \ll T_2'$), so that superradiant emission can occur before dephasing or relaxation processes set in. It immediately follows from Eq. (22) that this condition is equivalent to $\alpha_0 L \gg 1$, so that any sufficiently-high-gain system will undergo superradiant emission when suitably excited.

As a consequence of the long delay times which result from small effective input pulses, ψ remains small for almost all of the pulse evolution time. Accordingly, in the expression for $\mathcal{E}(x,T)$ the small-angle approximation ($\sin\psi = \psi$) can be used. (In the final stages of pulse evolution, ψ approaches π and this approximation breaks down.) Also, since the exact form of the effective input field does not matter (Sec. V), a step function can be chosen. A number of useful results can be derived from this model.

Consider first the delay T_D from the time at which the sample is inverted to the time of the first peak of the emitted radiation.[64] In the small-angle limit a solution of Eq. (46) can be found using the transformation $w = 2(xT)^{1/2}$, which gives Bessel's equation in w. For the initial condition of a small step-function input electric field with envelope $\mathcal{E}(x=0, T>0) = \mathcal{E}_0$, which corresponds to $\mathcal{E}(w=0) = \mathcal{E}_0$, the output field at $x = L$ is of the form

$$\mathcal{E}(L,T) = \mathcal{E}_0 I_0(u)\,, \tag{47}$$

where $I_n(x)$ is the modified Bessel function of order n, and $u = 2(T/T_R)^{1/2}$. Then $\Phi(T) = \psi(x=L,T)$, the partial area at $x = L$, is given by

$$\Phi(T) = \int_0^T (\mu_z/\hbar)\,\mathcal{E}(L,T')\,dT' = \theta_0 u I_1(u)\,, \tag{48}$$

where $\theta_0 = \mu_z \mathcal{E}_0 T_R/\hbar$. A property of solutions of Eq. (46) is that for $\theta_0 \ll 1$, $\Phi(T_D) \sim \pi$.[65] Setting $\Phi(I_D = \pi)$ in Eq. (48) and solving for T_D in the limit $T_D \gg T_R$, one obtains[66]

$$T_D \approx (T_R/4)[\ln(\theta_0/2\pi)]^2\,. \tag{49}$$

Note that Eq. (49) was derived under the condition of small θ_0. However, since Φ only deviates from Eq. (48) during the last few T_R before $T = T_D$, only a small error in calculating T_D is made by using Eq. (48) to approximate $\Phi(T_D)$.

In the experimental system, $\theta_0 \sim 10^{-8}$, so $|\ln(\theta_0/2\pi)| \sim 20$ and T_D is insensitive to changes in θ_0. Therefore $T_D \sim 100\,T_R$ is a convenient estimate of the time required for the superradiant state to evolve. Since T_D is proportional to T_R, T_D should be inversely proportional to the excitation density, and therefore, the pressure, in the sample cell. This inverse proportionality and the estimate of T_D, Eq. (49), agree with experimental results.[15]

An estimate of the width T_W of the first lobe of the radiation pulse can be obtained from the $1/e$ width of $\Phi(T)$ at $T = T_D$, i.e., by solving the equation $\Phi(T_D - T_W/2) = \Phi(T_D)/e$ for T_W:

$$T_W \sim T_R\,|\ln(\theta_0/2\pi)|\,, \tag{50}$$

since $T_W \ll T_D$. Equation (50) predicts a pulse width $\sim 20\,T_R$ for our system, which is somewhat smaller than the observed pulse widths of $\sim(20\text{–}40)\,T_R$.[15] This is expected, since the growth of $\Phi(T)$ near T_D is actually slower than that given by Eq. (48).

It was explained above that because the sample is optically thick (i.e., $\alpha_0 L \gg 1$) the polarization envelope $\mathcal{P}(z,T)$ varies over the medium and undergoes changes in sign [Fig. 3(a)]. Since $\mathcal{E} = 2\pi k \int \mathcal{P}\,dx$, regions of $\mathcal{P}$ of opposite sign tend to cancel and the output radiation at a particular time can be considered to originate from a single spatial region, of width L'_{eff}. Accordingly, L'_{eff} and T_W, the duration of one lobe of the superradiant output, are related; T_W is the "effective" T_R of a sample of length L'_{eff}, $T_W = T_R(L = L'_{\text{eff}}) = 8\pi T_{sp}/n\lambda^2 L'_{\text{eff}}$, from which $L'_{\text{eff}} \approx L/|\ln(\theta_0/2\pi)|$. Comparison with computer plots of $\mathcal{P}(x,T)$ [Fig. 3(a)] shows that L'_{eff} is approximately the half-width at half-maximum of a spatial region. The total extent of a region of polarization (between points of $\mathcal{P} = 0$) is several times larger. We define

$$L_{\text{eff}} = 4L'_{\text{eff}} \approx \frac{4L}{|\ln(\theta_0/2\pi)|} = \frac{4T_R L}{T_W}\,. \tag{51}$$

Although both L_{eff} and L'_{eff} have the correct functional dependence, the choice of L_{eff} leads to much better agreement between Eq. (54), below, and computer results.

Equation (51) indicates that a fraction $\sim 4/|\ln(\theta_0/2\pi)|$ of the medium contributes to each lobe of ringing in the superradiant bursts. This suggests that the normalized emission curve should contain $\sim|\ln(\theta_0/2\pi)|/4$ lobes. This is confirmed by computer results, which give this number of lobes (up to the $1/e$ point) for various values of θ_0. Systems with finite T_2' will have fewer lobes.

The total energy of the first lobe E_p is

$$E_p \approx \hbar\omega_0 n A L_{\text{eff}} = \frac{4\hbar\omega_0 n A L}{|\ln(\theta_0/2\pi)|}, \tag{52}$$

and the total peak radiation intensity I_p is approximately the total energy of the first lobe E_p divided by the width of the first lobe T_W,

$$I_p \approx \frac{E_p}{T_W} = \frac{\hbar\omega_0 n A L_{\text{eff}}}{T_W} \tag{53}$$

$$= \frac{4N\hbar\omega_0/T_R}{|\ln(\theta_0/2\pi)|^2} = \frac{4}{|\ln(\theta_0/2\pi)|^2}\,\frac{\hbar\omega_0 A n^2\lambda^2 L^2}{8\pi T_{sp}}, \tag{54}$$

and is proportional to L^2 (Fig. 4). As anticipated in the qualitative discussion of Sec. III, the peak radiation rate is enhanced from $N\hbar\omega_0/T_{sp}$ by a factor proportional to T_{sp}/T_R. Note that in terms of the peak value of the electric field, $\mathcal{E}_p$, Eq. (54) gives

$$(\mu\mathcal{E}_p/\hbar)(T_W/4) = 1, \tag{55}$$

which shows that the ringing of the superradiant output can be viewed as a form of Rabi nutation.[67]

The formulas for I_p [Eq. (54)], T_D [Eq. (49)], and T_W [Eq. (50)] agree well with experimental data,[15] computer predictions, and the normalized emission curve (Fig. 4). For extremely long samples (in the HF case, $\gtrsim 10$ m), the considerations of Ref. 26 must be taken into account.

VII. CONNECTIONS WITH OTHER WORK

Some previous treatments[11,12,17] of superradiance are based on equations of motion written in terms of the variables $I(t)$ and $n(t)$, the time-dependent intensity and inversion density. To compare the present work with these treatments, consider the approximation in which Eqs. (11) can be rewritten in these two variables, rather than the five real quantities $\mathcal{E}$, $\mathcal{P}$, and n.

An equation for $\partial I/\partial x$ can be obtained by multiplying Eq. (11a) by $(cA\mathcal{E}^*/8\pi)$:

$$\frac{\partial I}{\partial x} = -2\kappa I + (\omega_0 A/2)\,\mathrm{Re}[\mathcal{E}^*\langle\mathcal{P}\rangle_v], \tag{56}$$

which is a statement of energy balance. An expression for $\partial n/\partial t$ can be obtained by integrating Eq. (11c) over velocity:

$$\frac{\partial}{\partial t}\langle n\rangle_v + \frac{1}{T_2}\langle(n - T_2\Lambda)\rangle_v = -\frac{1}{\hbar}\mathrm{Re}[\mathcal{E}\langle\mathcal{P}^*\rangle_v] \tag{57}$$

$$= -\frac{2}{\hbar\omega_0 A}\left(\frac{\partial I}{\partial x} + 2\kappa I\right), \tag{58}$$

using Eq. (56). An expression for the total population difference $N = \int nA\,dx$ is then readily obtained[68]:

$$\frac{d}{dt}\langle N\rangle_v = -\frac{1}{T_2}\langle(N - T_2\Lambda AL)\rangle_v - \frac{2}{\hbar\omega_0}\left(I(L) - I(0) + 2\int_0^L \kappa I\,dx\right), \tag{59}$$

which has an obvious interpretation in terms of number conservation.

Equations (56) and (59) are exact. To simplify Eq. (56), we set $\kappa = 0$, neglect Doppler broadening and level degeneracy, and assume that $\mathcal{E}$ and $\mathcal{P}$ have a constant phase.[69,70] Differentiating Eq. (56) with respect to T then gives

$$\frac{2}{\omega_0 A}\frac{\partial^2 I}{\partial x\partial T} = \frac{\partial\mathcal{E}}{\partial T}\mathcal{P}^* + \mathcal{E}\frac{\partial\mathcal{P}^*}{\partial T} \tag{60}$$

$$= \frac{1}{2I}\frac{\partial I}{\partial T}\mathcal{E}\mathcal{P}^* - \frac{\mathcal{E}\mathcal{P}^*}{T_2} + \frac{\mu_g^2}{\hbar}n|\mathcal{E}|^2 - \mathcal{E}\Lambda_p, \tag{61}$$

using Eq. (11b). Then, using Eq. (56),

$$\frac{\partial^2 I}{\partial x\partial T} = \frac{1}{2I}\frac{\partial I}{\partial T}\frac{\partial I}{\partial x} - \frac{1}{T_2}\left(\frac{\partial I}{\partial x} - \frac{2nIT_2}{n_0 L T_R} - 2\alpha_0 K_0\right). \tag{62}$$

The last term in Eq. (62) is obtained by replacing the source term $\mathcal{E}\Lambda_p$ of Eq. (61) by $4\alpha_0 K_0/\omega_0 A T_2$, where the form of K_0 depends on the gain of the system [Eq. (31) or Ref. 61]. This substitution is consistent with the averaging procedure used in Sec. IV in deriving the polarization source. Note that in the equilibrium limit, where $\partial I/\partial t = 0$, Eq. (62) reduces to the usual expression for intensity gain [Eq. (A13)],

$$\frac{dI}{dx} = 2\left(\frac{nT_2}{n_0 L T_R}\right)I + 2\alpha_0 K_0, \tag{63}$$

the term in parentheses being α_0.

Equation (62) is exact, subject to the stated assumptions. A system which satisfies these conditions can be described by two real equations, Eqs. (58) [or (59)] and (62), in two unknowns (n and I), rather than by Eqs. (11). Note that the influence of spontaneous emission is included in these equations. In general, they must be solved subject to appropriate initial conditions on I and

n. For simplicity, in the following discussions we will assume that the input field is negligible.

Equations (59) and (62) are a useful starting point to connect the present analysis with some previous treatments[11,12,17] which developed equations appropriate to a regime which we would like to call "limited superradiance." This regime is characterized by the conditions

$$T_R \ll T_{sp}, \tag{64a}$$

but

$$\alpha_0 L \ll 1, \tag{64b}$$

i.e.,

$$T_2' \ll T_R \ll T_{sp}. \tag{64c}$$

In this limit, spatial variations in $\mathcal{P}$ and n are negligible throughout the sample,[71] and Eq. (62) can be integrated to obtain

$$\frac{dI}{dt} = -\frac{2I}{T_2} + \frac{4IN}{3T_R N_0} + \frac{\hbar\omega_0}{\pi T_2 T_R}, \tag{65a}$$

where, henceforth, $I = I(L,t)$. Similarly, Eq. (59) becomes

$$\frac{dN}{dt} = -\frac{1}{T_2}(N - T_2 \Lambda A L) - \frac{2I}{\hbar\omega_0}. \tag{65b}$$

As will be seen below, solutions of these equations are very different from those obtained in the high-gain case and from the experimental results (different delay times, no ringing, etc.).

Equations (65) are almost identical to the superradiant rate equations of Bonifacio, Schwendimann, and Haake,[12] which in our notation become

$$\frac{dI}{dt} = -\frac{2I}{T_2} + \frac{4IN}{T_R N_0} - \frac{4I}{T_R N_0}, \tag{66a}$$

$$\frac{dN}{dt} = -\frac{1}{T_1}(N - T_1 \Lambda A L) - \frac{2I}{\hbar\omega_0}. \tag{66b}$$

(They did not assume $T_1 = T_2$.)

Equations (4.10) and (4.11) of Rehler and Eberly[11] can be differentiated to obtain equations similar to ours (they assume $T_1^{-1} = 0$):

$$\frac{dI}{dt} = -\frac{2I}{T_2} + \frac{4IN}{T_R N_0}, \tag{67a}$$

$$\frac{dN}{dt} = -\frac{2I}{\hbar\omega_0}. \tag{67b}$$

Similar equations obtained by Ressayre and Tallet[17] include finite Doppler broadening:

$$\frac{dI}{dt} = -\left(\frac{1}{T_2} + \frac{1}{T_2^*}\right) I - \frac{IN}{T_R N_0} \exp\left(-\frac{t}{T_2^*}\right), \tag{68a}$$

$$\frac{dN}{dt} = -\frac{N - T_2 \Lambda A L}{T_2} - \frac{2I}{\hbar\omega}. \tag{68b}$$

In the limit of $T_2^* \to \infty$, these equations are of the same form as the others.[72]

These three sets of equations [(66), (67), and (68)] essentially agree with Eqs. (65), which are derived in the thin-sample limit and do not apply to $\alpha_0 L \gg 1$.

In Refs. 11, 12, and 17, these equations are solved in the limit where $2I/T_2$ is negligible compared to $4IN/3T_R N_0$. The solutions for I are then hyperbolic-secant-squared functions of time. However, the ratio of the first term to the second term in Eq. (65a) is

$$\frac{2I/T_2}{4IN/3T_R N_0} = \frac{3}{2\alpha_0 L}\frac{N_0}{N}. \tag{69}$$

In the thin-sample limit, in which Eqs. (65) are valid, $\alpha_0 L = T_2/T_R \ll 1$, and since $|N| \le N_0$ always, the magnitude of this ratio will always be much greater than 1. As a result, the $2I/T_2$ term may not be neglected, but the term $4IN/3T_R N_0$ is negligible and may be ignored. Therefore, the hyperbolic-secant-squared intensity solutions of Refs. 11, 12, and 17 are incorrect.

The approximate solution of Eqs. (65) may be obtained by neglecting the second term of Eq. (65a):

$$I(L,t) = I(L,t=0)\,e^{-2t/T_2} + (\hbar\omega_0/4\pi T_R)(1 - e^{-2t/T_2}), \tag{70a}$$

$$N = \frac{2I(L,t=0)}{\hbar\omega_0}(e^{-2t/T_2} - e^{-t/T_2}) + N(t=0)\,e^{-t/T_2} + T_2 \Lambda A L(1 - e^{-t/T_2}), \tag{70b}$$

where $N(t=0)$ is the initial population inversion and $I(L,t=0)$ is the initial output intensity,[73] produced by the population existing at $t=0$ in a medium having initial conditions $n(t=0) = n_0 \cos\phi$ and $\mathcal{P}(t=0) = \mu_z n_0 \sin\phi$ (corresponding to an initial tipping angle ϕ of the Bloch vector):

$$I(L,t=0) = (cA/8\pi)(2\pi k L \mu_z n_0 \sin\phi)^2 = (N\hbar\omega_0/4T_R)\sin^2\phi. \tag{71}$$

The second term in Eq. (70a), which is due to the source term of Eq. (65a), is only important when $I(L,t=0)$ is negligible ($\lesssim \hbar\omega_0/4\pi T_R$). In this case incoherent processes initiate the evolution of the system, and Eq. (70a) becomes

$$I = \frac{\hbar\omega_0}{4\pi T_R}(1 - e^{-2t/T_2}) = \frac{N_0\hbar\omega_0}{T_{sp}}\frac{\lambda^2}{32\pi^2 A}(1 - e^{-2t/T_2}). \tag{72}$$

Here $I(t)$ is proportional to n_0 and decays exponentially in the usual fashion.

The first term in Eq. (70a) is the free-induction decay of $I(L,t=0)$. For $I(L,t=0) \gtrsim \hbar\omega_0/4\pi T_R$; the second term in Eq. (70a) is negligible, and $I(t)$ is proportional to n_0^2 (coherent emission). This solution is a decaying exponential with decay time $T_2/2=\alpha_0 L T_R/2 \ll T_R$, rather than the hyperbolic-secant-squared solution of width $\sim T_R$ of Refs. 11 and 12. Therefore, collective radiation occurs for a time much shorter than T_R, after which the excited-state population will have decayed. Integrating $I(t)=(N\hbar\omega_0/4T_R)e^{-2t/T_2}\sin^2\phi$ over time, the total energy radiated coherently is at most $(N\hbar\omega_0/8)(T_2/T_R)$, so that only a small fraction $T_2/8T_R=\alpha_0 L/8 \ll 1$ of the energy is radiated coherently. The decay of the system is determined by ordinary incoherent processes.

Experimental techniques such as free-induction decay and photon echos,[3] which study the collective radiative decay of a system in the limited superradiance regime, are useful for measuring incoherent relaxation processes. In fact, these experiments must be performed in the thin sample ($\alpha_0 L \ll 1$) so that coherent emission does not affect the decay rate. In the regime of limited superradiance, the coherent emission acts as a probe of the state of the system, without significantly affecting its state.

Although limited superradiance can be considered superradiance in the sense that $T_R \ll T_{sp}$, so that collective effects are important [as is seen in the n_0^2 dependence of Eq. (71)], only a very small fraction of the energy is radiated coherently since $T_2 \ll T_R$. This is in sharp contrast to the regime of strong superradiance studied in the HF experiments, which is characterized by the conditions

$$T_R \ll T_{sp} \tag{73a}$$

and

$$\alpha_0 L \gg 1\,, \tag{73b}$$

i.e.,

$$T_2' \gg T_R\,. \tag{73c}$$

The latter condition implies that the sample can superradiate before decay processes set in. Accordingly, in the strong superradiant regime essentially all of the energy stored in the sample can be emitted coherently. [The condition of Eq. (73b) for strong superradiance can be made more precise in some cases.[74]]

The long delays preceding the observed output pulses deserve further discussion, in light of the fact that the conditions for strong superradiance [Eqs. (7)] only require $T_2^* \gg T_R$, not $T_2^* > T_D$. (However, $T_2 > T_D$ is necessary.) For example, typically $T_D \sim 100T_R$, whereas $T_2^* \sim 50T_R$, so that $T_D > 2T_2^*$. It is remarkable that fully developed pulses with ringing can evolve over such long times, despite the presence of dephasing processes. This behavior is unique to high-gain amplifiers, where the high gain can overcome dephasing during the early stages of pulse evolution. The system eventually dephases, but the effective dephasing time is increased to $\alpha_0 L T_2^*$, as can be established by considering the response of a high-gain inhomogeneously broadened amplifying medium to a short input pulse of small area θ_0. (A small step-function input pulse gives similar results.) Analytical expressions have been given by Crisp.[75] For a Lorentzian line shape the output $\mathcal{E}$ field is of the form

$$\mathcal{E}(x=L,T) \approx \frac{\mathcal{E}_0}{2\sqrt{\pi}}\left(\frac{T}{T_R}\right)^{-3/4} \times \exp\left[2\left(\frac{T}{T_R}\right)^{1/2} - \frac{T}{T_R\alpha_0 L}\right], \tag{74}$$

for an input pulse of small area $\theta_0=\mu\mathcal{E}_0 T_R/\hbar$. Initially the square-root term in the exponential dominates and $\mathcal{E}$ increases. Its maximum value is reached at $T\approx T_2^*\alpha_0 L$, after which exponential decay due to dephasing sets in. Therefore, the effective dephasing time is

$$(T_2^*)_{\rm eff} \approx T_2^*\alpha_0 L\,. \tag{75}$$

When this expression is compared with Eq. (49) for the delay time, one finds that

$$\frac{T_D}{(T_2^*)_{\rm eff}} \approx \left(\frac{\ln(\theta_0/2\pi)}{2\alpha_0 L}\right)^2, \tag{76}$$

which is always smaller than unity for a high-gain system (where $\alpha_0 L \gg 1$). Therefore, an inhomogeneously broadened system of sufficiently high gain will always superradiate before it can dephase.[76]

In a recent series of papers, Bonifacio and Banfi[18] and Bonifacio and Lugiato[19,20] have developed equations in terms of $\mathcal{E}$, $\mathcal{P}$, and n (rather than I and N) for the cooperative radiation from an initially inverted two-level system, which they call superfluorescence. In our notation, the semiclassical equations which they obtain[77] can be written in the form

$$\left[\frac{1}{c}\left(\frac{\partial}{\partial t}\right)_x + \left(\frac{\partial}{\partial x}\right)_t + \frac{1}{2L}\right]\mathcal{E}(x,t) = \left[\left(\frac{\partial}{\partial x}\right)_T + \frac{1}{2L}\right]\mathcal{E}(x,T) = \frac{2\pi\omega_0}{c}\mathcal{P}(x,T)\exp\left(-\frac{t}{2T_2^*}\right), \tag{77a}$$

$$\frac{\partial}{\partial T}\mathcal{P}(x,T) = \frac{\mu_z^2}{\hbar} n\mathcal{E}\exp\left(-\frac{t}{2T_2^*}\right), \tag{77b}$$

$$\frac{\partial}{\partial T} n(x,T) = -\frac{1}{\hbar}\mathrm{Re}(\mathcal{E}^*\mathcal{P})\exp\left(-\frac{t}{2T_2^*}\right), \tag{77c}$$

where $T=t-x/c$. Although these equations are similar to Eqs. (11) above, they contain two major differences: (i) The term $(1/2L)$ in Eq. (77a) has been included to account for "the irreversible escape (propagation) of the total Maxwell field from the active volume to the outside."[19] However, wave propagation is built into Maxwell's equations (the $\partial/\partial x$ terms) so that the $1/2L$ term appears to be incorrect and was added unnecessarily. (ii) To account for inhomogeneous broadening, in Eqs. (77) the factor $\exp(-t/2T_2^*)$ is included. This factor arises from the assumption[19] that there is no correlation between frequency and position during the evolution of the superradiant pulse, an assumption which we believe is incorrect. Henceforth, we restrict ourselves to the case $T_2^*=\infty$, in which case Eq. (77) agrees with Eq. (59).

In Ref. 19, Eqs. (77) are solved by neglecting the $(\partial/\partial x)_t$ term. This incorrect approximation is justified[19] by the argument that in the case of "uniform excitation" (side pumping), Eqs. (77) are spatially invariant for all time. However, since the inverted medium is finite in length, this is not the case—the initial condition is spatially constant only over a finite region of space. Therefore, after a time of order L/c spatial variations will have developed throughout the sample. Computer solutions also substantiate this point.

In summary, we doubt that any optically thick ($\alpha_0 L \gg 1$) superradiant system can accurately be described by any set of differential equations which ignores spatial variations. We also doubt that the pendulum equation of Ref. 19 [Eq. (11.12)] applies to strong superradiance.[77a]

Another recent paper by Bonifacio, Hopf, Meystre, and Scully[78] deals with the evolution of steady-state pulses. This work is an extension of and in the same spirit as discussions of this subject in Refs. 22 and 24. This work with steady-state pulses is applicable to a system in which the loss coefficient κ is sufficiently high so as to suppress ringing [the decrease in ringing for large κ can be seen in the $\kappa L=5$ curve in Fig. 5].

VIII. CONCLUSION

This paper has developed a semiclassical description of the evolution of an initially inverted system into a superradiant state, and shown that the inclusion of a properly constructed fluctuating polarization source in the coupled Maxwell-Schrödinger equations gives rise to output pulses which quantitatively agree with experimental results and have all the features expected of a superradiant system. The success of this model makes clear the simple nature of the superradiant process and places it in perspective with other coherent phenomena.

As shown in this paper, in the optical region spontaneous emission initiates the pulse evolution process and greatly influences the time delay and other experimentally observable features of the output radiation. The quantitative agreement of the semiclassical treatments with experiment is largely due to the fact that in a high-gain system the effects of spontaneous emission can be combined into a single parameter θ_0 which enters the equations logarithmically, so that the exact details of the spontaneous-emission process become unimportant. Nevertheless, the need still exists for a quantized-field treatment to substantiate the semiclassical results and to explore quantum-mechanical features of superradiance such as fluctuations. Since it is now generally recognized that a semiclassical description is adequate once the pulse evolution process is under way, the quantized-field-theory treatment can be restricted to the small-signal regime, where important simplifications occur. Several recent papers[79] have made useful advances along these lines. It is our hope that the present work will stimulate further research in this direction.

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APPENDIX A: DERIVATION OF INTENSITY GAIN EQUATION FROM COUPLED MAXWELL-SCHRÖDINGER EQUATIONS FOR A SYSTEM NEAR THERMAL EQUILIBRIUM

We consider a linear steady-state system. Schrödinger's equation [Eq. (11b)] can be rewritten in the form

$$\left(\frac{\partial}{\partial T}\right)[\mathcal{P} e^{T(\gamma-i\Gamma)}] = \left(\frac{\mu_z^2}{\hbar}\right) n\mathcal{E}\, e^{T(\gamma-i\Gamma)} + \Lambda_p e^{T(\gamma-i\Gamma)}, \tag{A1}$$

where $\Gamma=kv$ and Λ_p, which includes level degeneracy and velocity dependence, is given by Eq. (24),

$$\Lambda_p(x,T,v,M)=B_1[n_2(x,T,v,M)]^{1/2}\times\sum_{j=1}^{j'}\delta(x-x_j)\sum_{l=-\infty}^{\infty}\delta(T-T_l)\times\sum_{k=1}^{k'}\delta(v-v_k)e^{i\phi_{jlkM}}, \tag{A2}$$

where $x_j=(j-\frac{1}{2})\Delta x$, $\Delta x=L/j'$, $T_l=l\Delta T$, and the v_k are k' discrete velocities. Then integrating Eq. (A1) over T from $-\infty$ to T gives

$$\mathcal{P}(T)\,e^{T(\gamma-i\Gamma)}=\frac{\mu_z^2}{\hbar}\int_{-\infty}^{T}n\mathcal{E}\,e^{T'(\gamma-i\Gamma)}dT' +B_1\sum_j\sum_l\sum_k e^{i\phi_{jlkM}}\delta(x-x_j)\,\delta(v-v_k)\times\int_{-\infty}^{T}\delta(T'-T_l)\,(n_2)^{1/2}\times e^{(T'/T_2-i\Gamma T')}dT'. \tag{A3}$$

In the case where n and $\mathcal{E}$ are very slowly varying in time and n is approximately constant, we can replace the populations $n(x,T,v,M)$ by their equilibrium values n_0, and Eq. (A3) becomes

$$\mathcal{P}(T)=\frac{\mu_z^2}{\hbar}\,\frac{n_0\mathcal{E}}{1/T_2-i\Gamma} +B_1(n_{20})^{1/2}\sum_j\sum_l\sum_k e^{i\phi_{jlkM}}\,u(T-T_l)\,\delta(x-x_j)\times\delta(v-v_k)\,e^{-(T-T_l)/T_2}, \tag{A4}$$

where $u(t)$ is the unit step function, and n_{20} is the equilibrium value of n_2.

Substituting this result into Maxwell's equation [Eq. (11a)] in the case of no linear loss (which is the case of interest), we obtain

$$\frac{\partial\mathcal{E}}{\partial x}-\left[\frac{2\pi k\mu_z^2}{\hbar}\left\langle\frac{n_0}{1/T_2-i\Gamma}\right\rangle_{v,M}\right]\mathcal{E} =2\pi k\sum_j\sum_l e^{-(T-T_l)/T_2}\,u(T-T_l)\,\delta(x-x_j)\times\left\langle B_1[n_{20}(x,T,v,M)]^{1/2}\sum_k\delta(v-v_k)\,e^{i\phi_{jlkM}}\right\rangle_{v,M} \tag{A5}$$

$$=2\pi k\sum_j\sum_l\sum_k\sum_{M=-J}^{J}e^{-(T-T_l)/T_2}\,u(T-T_l)\,\delta(x-x_j)\times B_1[n_{20}(x,T,v_k,M)]^{1/2}e^{i\phi_{jlkM}}. \tag{A6}$$

Since the imaginary part of the coefficient of $\mathcal{E}$ in Eq. (A5) vanishes when integrated over velocity, and the real part is the gain at line center α_0, Eq. (A6) becomes

$$e^{\alpha_0x}\frac{\partial}{\partial x}[\mathcal{E}e^{-\alpha_0x}] =\sum_j\sum_l\sum_k\sum_M e^{-(T-T_l)/T_2}\,u(T-T_l)\times\delta(x-x_j)\,B_2e^{i\phi_{jlkM}}, \tag{A7}$$

where $B_2(v_k,M)=2\pi k\,B_1[n_{20}(v_k,M)]^{1/2}$. Integrating along a path of constant T from $x=(j-1)\Delta x$ to $x=j\Delta x$ gives

$$\mathcal{E}(j\Delta x)=\mathcal{E}((j-1)\Delta x)\,e^{\alpha_0\Delta x} +\sum_k\sum_l\sum_M e^{-(T-T_l)/T_2}\,u(T-T_l)\times B_2e^{\alpha_0\Delta x/2}e^{i\phi_{jlkM}} \tag{A8}$$

for $j=1,2,3,\ldots,j'$. Simultaneous solution gives

$$\mathcal{E}(L,T)=\mathcal{E}(0,T)\,e^{\alpha_0L}+\sum_k\sum_l\sum_M B_2\sum_{j=1}^{j'}e^{\alpha_0(j-1/2)\Delta x}u(T-T_l)\,e^{-(T-T_l)/T_2}e^{i\phi_{jlkM}} \tag{A9}$$

and

$$I(L,T)=|\mathcal{E}(L,T)|^2\frac{cA}{8\pi}=|\mathcal{E}(0,T)|^2e^{2\alpha_0L}\frac{cA}{8\pi}+\frac{cA}{8\pi}\sum_j\sum_l\sum_k\sum_M|B_2|^2e^{(2j-1)\alpha_0\Delta x}u(T-T_l)\,e^{-2(T-T_l)/T_2} +(\text{terms containing random phases}). \tag{A10}$$

For $\alpha_0\Delta x\lesssim1$ and $\Delta T/T_2\lesssim1$, no single terms dominate the sum of random-phase terms, and the sum of these terms (which individually average to zero) will be negligible compared to the other two terms in Eq. (A10). Then Eq. (A10) can be written as the intensity gain equation of the text [Eq. (25)],

$$I(x,T)=I(0,T)\,e^{2\alpha_0x}+K_0(e^{2\alpha_0x}-1), \tag{A11}$$

where

$$K_0 = \frac{\pi}{2}\,\frac{\omega_0^2}{c}\,\frac{AT_2}{2\Delta T}(1-e^{-2\alpha_0\Delta x})^{-1}B_1^2 \times \sum_k \sum_M n_{20}(v_k, M)\,. \tag{A12}$$

[Note that Eq. (A11) is the solution of the differential equation

$$\frac{dI}{dx} = 2\alpha_0 I + 2\alpha_0 K_0\,, \tag{A13}$$

which is therefore an equivalent intensity gain equation.] Examination of Eq. (A12) shows that if the effects of level degeneracy and velocity dependence were eliminated by choosing one velocity and by assuming $M=0$ for all molecules, we would obtain the same result,

$$K_0 = \frac{\pi}{2}\,\frac{\omega_0^2}{c}\,\frac{AT_2}{2\Delta T}(1-e^{-2\alpha_0\Delta x})^{-1}B_1^2 n_{20}(x,T)\,, \tag{A14}$$

that would be obtained by summing $n_{20}(v_k, M)$ over k and M. [Here $n_{20}(x,T) = \sum_k \sum_M n_{20}(v_k, M)$.]

APPENDIX B: CONVERSION OF INTENSITY PER MODE TO PLANE-WAVE INTENSITY PER UNIT BANDWIDTH

The Einstein intensity equation[56] [Eq. (27)]

$$\frac{dI_\omega(x)}{dx} = 2\alpha(\omega)\left(I_\omega(x) + \frac{\hbar\omega n_2}{n_2-n_1}\right) \tag{B1}$$

gives the total energy $I_\omega(x)$ per mode in the active region of any linear-gain medium. To convert $I_\omega(x)$ into an intensity $I(\omega,x)$ per unit bandwidth in the plane-wave direction, we note that the total power $I(x)$ in the plane-wave direction is

$$I(x) = \int_0^\infty \left(\frac{2\pi\nu^2 d\nu}{c^3}\,cA\right)\left(\frac{\lambda^2}{4\pi A}\right) I_\omega(x)\,, \tag{B2}$$

where the first factor is the number of modes of *one* polarization[81] in the active region in a frequency interval $d\nu = d\omega/2\pi$ and the second factor is that fraction f of the sphere over which plane waves contribute coherently. In the plane-wave limit, valid for a system with large Fresnel number (a "disk"), $f = \Delta\Omega/4\pi = \lambda^2/4\pi A$ [Eq. (1)]. If we define $I(\omega,x)$ by $I(x) \equiv \int I(\omega,x)\,d\omega$, then $I(\omega,x) = (1/4\pi) I_\omega(x)$, and Eq. (B1) becomes [Eq. (28)]

$$\frac{\partial I(\omega,x)}{\partial x} = 2\alpha(\omega)\left(I(\omega,x) + \frac{\hbar\omega}{4\pi}\,\frac{n_2}{n_2-n_1}\right). \tag{B3}$$

APPENDIX C: BANDWIDTH NARROWING IN LINEAR HIGH-GAIN SYSTEM (REFS. 31 AND 39)

The intensity gain equation for the plane-wave intensity $I(\omega,x)$ per unit bandwidth is [Eq. (28) or (B3)]

$$\frac{\partial I(\omega,x)}{\partial x} = 2\alpha(\omega)\left(I(\omega,x) + \frac{\hbar\omega}{4\pi}\,\frac{n_2}{n_2-n_1}\right). \tag{C1}$$

Integrating Eq. (C1) over x gives

$$I(\omega,x) = (e^{2\alpha(\omega)x} - 1) I_x(\omega) + I(\omega, x=0)\,, \tag{C2}$$

where

$$I_x(\omega) = I(\omega, x=0) + \frac{\hbar\omega}{4\pi}\,\frac{n_2}{n_2-n_1}. \tag{C3}$$

The total power at $x=L$ can then be obtained by integrating over ω:

$$I(x=L) = \int_0^\infty I(\omega, x=0)\,d\omega + \int_0^\infty d\omega\, I_x(\omega)\,(e^{2\alpha(\omega)L} - 1)\,. \tag{C4}$$

For a high-gain system [$\alpha(\omega=\omega_0)L \gtrsim 20$], the exponential factor in Eq. (C4) is very sharply peaked near $\omega=\omega_0$, so that $I_x(\omega)$ can be replaced by $I_x(\omega_0)$ and removed from the integral. Also for a high-gain system, the first integral can be neglected, being much smaller than the second integral. Equation (C4) becomes

$$I(L) \approx I_x(\omega_0) \int_0^\infty d\omega\,(e^{2\alpha(\omega)L} - 1)\,. \tag{C5}$$

To find $I(L)$, an integral of the form

$$I_1 = \int_0^\infty d\omega\,[e^{2\alpha_0 LG(\omega)} - 1] \approx \int_{-\infty}^\infty d\omega (e^{2\alpha_0 LG(\omega)} - 1)\,, \tag{C6}$$

where $\alpha_0 = \alpha(\omega=0)$ and $G(\omega) = \alpha(\omega)/\alpha_0$, must be solved. Expanding the exponential in a power series and interchanging integration and summation gives

$$I_1 = \sum_{J=1}^\infty \frac{(2\alpha_0 L)^J}{J!} \int_{-\infty}^\infty [G(\omega)]^J d\omega\,. \tag{C7}$$

For $G(\omega)$ that occur in slowly varying physical systems, such as Lorentzians and Gaussians, $[G(\omega)]^J d\omega \approx [G(\omega)]^{(J-1)} d\omega$ for $J \gg 1$, so that the ratio of term J to term $J-1$ of Eq. (C7) is approximately $2\alpha_0 L/J$. Therefore, for $\alpha_0 L \gg 1$ (high-gain system), the terms which contribute most to the sum will be those for which $J \approx 2\alpha_0 L$. We can therefore approximate the factor $\int [G(\omega)]^J d\omega$ appearing in Eq. (C7) by $\int [G(\omega)]^{2\alpha_0 L} d\omega$ to obtain

$$I_1 \approx \sum_{J=1}^\infty \frac{(2\alpha_0 L)^J}{J!} \int_{-\infty}^\infty [G(\omega)]^{2\alpha_0 L} d\omega = e^{2\alpha_0 L} \int_{-\infty}^\infty [G(\omega)]^{2\alpha_0 L} d\omega\,. \tag{C8}$$

For our system, the gain profile is Lorentzian [see discussion above Eq. (15)], with $G(\omega)=(ku_1)^2/[(\omega-\omega_0)^2+(ku_1)^2]$.[82] Then,

$$I_1=(\pi/2\alpha_0 L)^{1/2}ku_1 e^{2\alpha_0 L}\,, \tag{C9}$$

and we obtain the result of Eq. (29),

$$I(L)=\left(I(\omega_0,x=0)+\frac{\hbar\omega_0}{4\pi}\,\frac{n_2}{n_2-n_1}\right)e^{2\alpha_0 L}(\Delta\omega)^{-1}_{\rm eff}\,, \tag{C10}$$

where $(\Delta\omega)^{-1}_{\rm eff}=T_2'(\pi/2\alpha_0 L)^{1/2}$, as in the text.

Examination of the form of $I_x(\omega)$ [Eq. (C3)] or of $I(L)$ [Eq. (C10)] shows that in a high-gain system, a distributed source can be replaced by an "equivalent" intensity source at the input face which will give the same output intensity. In other words, a high-gain system described by the intensity gain equation

$$\frac{dI(x)}{dx}=2\alpha_0 I(x) \tag{C11}$$

and an input intensity

$$I(x=0)=\frac{\hbar\omega_0}{4\pi}\left(\frac{n_2}{n_2-n_1}+\frac{1}{e^{\hbar\omega_0/kT}-1}\right)(\Delta\omega)_{\rm eff} \tag{C12}$$

would give the same output intensity $I(L)$ which we derived [Eq. (C10)] using our distributed-source model. In a high-gain system, therefore, the distributed source of Eq. (C1) can be replaced by an equivalent input field.

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[41]Accordingly, in the preceding discussion the length of the dipole array could be considered to be the length L_{eff} ($<L$) over which the polarization envelope is approximately constant. See Eq. (51) below.

[42]See, for example, Ref. 24.

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[47]This formula is a consequence of the optical pumping process and is not basic to the superradiant pulse evolution process.

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[52]A similar expression for Λ_p without spatial dependence was used by J. A. Fleck, Jr. [Phys. Rev. B 1, 84 (1970)]. In the present case, spatial dependence of Λ_p must be included, since in a high-gain system spatial variations are essential to superradiant behavior.

[53]The discrete velocities should be chosen to optimize the convergence of Eqs. (11). See Ref. 24.

[54]"Very slowly varying" should not be confused with "slowly varying" ($\partial/\partial T \ll \omega_0$, $\partial/\partial z \ll \omega_0/c$), which has already been assumed in the derivation of Eqs. (11a) and (11b). Note that $\mathcal{E}$ is not necessarily very slowly varying in space, even near thermal equilibrium; i.e., we do not require $\partial\mathcal{E}/\partial z \ll \gamma\mathcal{E}/c$.

[55]These conditions hold, for example, near thermal equilibrium. Note that the presence of delta functions in Λ_p is not inconsistent with very slowly varying n and $\mathcal{E}$; although Λ_p has delta functions, the source terms for n and $\mathcal{E}$ do not.

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[59]There are some errors in the numerical factors in the corresponding equations in Ref. 14.

[60]The derivation (Ref. 44) of the area theorem assumes a zero-phase external field incident upon a nondegenerate medium with no distributed sources.

[61]This follows from the expression for the cross section of a divergent Gaussian beam. See, for example, M. Sargent III *et al.*, *Laser Physics* (Addison-Wesley, Reading, Mass., 1974), p. 369.

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[63a] *Note added in proof.* The normalized emission curve is obtained from Eq. (46) using the substitution $w=2(xT)^{1/2}$, as in Ref. 63. One obtains $d^2\psi/dw^2 + (1/w)\,d\psi/dw = \sin\psi/LT_R$, which for the initial condition $\psi(w=0) << 1$ gives the normalized emission curve. This initial condition is equivalent to a δ-function input $\mathcal{E}$ field, which is approximately equivalent (Sec. V) to any other input pulse of equal small effective area.

[64]If the population excitation time is appreciable, T_D should be measured from the central part of the excitation pulse. See Ref. 26.

[65]This can be seen from the figures of D. C. Burnham and R. Y. Chiao (Ref. 63).

[66]In the derivation of Eq. (49), the argument of the logarithm depends on T_D and is of order θ_0/π; the chosen numerical factor provides a very accurate fit to the numerical results of Sec. V.

[67]From these approximate expressions a picture emerges in which the normalized output emission curve is similar to a damped sine-squared curve, $I(T) = I_p e^{-(T-T_D)/\tau}\cos^2[\mu\mathcal{E}_p(T-T_D)/4\hbar]$, of frequency $(\mu\mathcal{E}_p/4\hbar)^{-1}$ $[=T_R|\ln(\theta_0/2\pi)|=T_W]$ and decay constant τ

$= (\pi/16)T_R[\ln(\theta_0/2\pi)]^2$. This equation gives a qualitative fit to the normalized output curve, and as such illustrates the interpretation of superradiance as a damped oscillatory process, where damping is due to the release of coherent radiation from the sample. Behavior of this type can be derived from a differential equation obtained by considering the fraction of the energy radiated in one lobe of radiation and the time between the first two output peaks. Combining this differential equation with Eq. (55) gives $I(T)$ as above except for a factor of 4 in τ, a difference attributable to the fact that the true decay of energy slows temporarily each time $\mathcal{E}$ passes through zero.

[68]To simplify intermediate calculations we have assumed negligible transit time so that $t = T$ in Eqs. (59)–(62).

[69]These approximations were justified in Sec. IV and are confirmed by the numerical results (Sec. V) which, in particular, show that the phase of $\mathcal{P}$ is reasonably constant throughout the medium and in time.

[70]The constant-phase assumption implies that the carrier frequency is close to resonance ($\nu \sim \omega_0$). This approximation is valid because the spontaneous-emission profile is symmetric about its peak at ω_0.

[71]In integrating Eq. (62) care must be taken to keep all terms of order $\alpha_0 L$ in order to obtain the correct numerical coefficient of the second term in Eq. (65a). However, as seen below, that numerical factor is unimportant because that term is negligible ($\alpha_0 L \ll 1$).

[72]The solutions of Eqs. (68) obtained in Ref. 17 do not agree at all with the experimental results of Ref. 6, notwithstanding the analysis of Ref. 17. In the fit therein to the experimental results, the time axis has been shifted arbitrarily and the experimentally observed ringing is disregarded. Although Ref. 17 claims that large delays are due to inhomogeneous broadening, their calculations do not give the observed delays. In fact, our results show that for constant T_R, the delay *decreases* as the inhomogeneously broadened width $1/T_2^*$ increases [Eq. (75)].

[73]Equations (66) do not allow $I(L, t=0) = 0$, because Ref. 12 requires that $I(L, t=0)$ must be at least as large as a value corresponding to spontaneous emission.

[74]As can be seen from the discussion of $(T_2^*)_{\rm eff}$ [Eq. (75)], $\alpha_0 L \gtrsim |\ln\theta_0|$ is sufficient if T_2^* is the dominant broadening time, but $\alpha_0 L \gg |\ln\theta_0|$ is necessary if T_2 dominates. This follows from the fact that T_D must be shorter than both the incoherent decay time and the effective dephasing time of the system so that coherent decay will dominate.

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[77a] *Note added in proof.* The pendulum equation of Ref. 19, $d^2\psi/dt^2 + [(c/2L) + (1/2T_2^*)]\, d\psi/dt = (c/2LT_R)e^{-t/T_2^*} \times \sin\psi$, is a function of time only. Note that the pendulum equation obtained from the present treatment (Ref. 63a) is a function of both space and time.

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[81]The intensities of the two polarizations grow independently, but the one which starts first will dominate.

[82]For a Gaussian $G(\omega) = e^{-(\Delta\omega/ku)^2}$, $I_1 = (\pi/2\alpha_0 L)^{1/2} \times ku e^{2\alpha_0 L} = [\pi/(2\alpha_0 L)^{1/2}] e^{2\alpha_0 L}/T_2^*$.