Collective spontaneous emission (Dicke superradiance)

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The present state of theoretical and experimental research on collective spontaneous emission (superradiance) in a system of radiators (atoms, molecules, or nuclei) is reviewed. The distinction between superradiance and the amplification of spontaneous emission is discussed. There is also a discussion of conditions under which the effect can be observed and of various theoretical methods for describing superradiance: the quantum single-mode and multimode models and the semiclassical approach. Theoretical papers on superradiance in systems with dimensions smaller than the wavelength of the radiation and also in extended systems are reviewed. It is shown that superradiance may occur in weakly amplifying media. A situation in which the superradiance is oscillatory is described. The possible use of superradiance to generate coherent emission in the x-ray and γ ranges is discussed. The superradiance accompanying Raman scattering of light in atomic and molecular media is studied. The theoretical results are compared with experimental observations of superradiance in the optical range. In an appendix, the nature of the phase transition in a system of radiators interacting through an electromagnetic field is discussed.

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CONTENTS

I. INTRODUCTION

In his pioneering 1954 paper,[1] Dicke showed that a system of N two-level atoms with a population inversion could spontaneously revert to the ground state in a time inversely proportional to the number of atoms, $\tau_e \sim 1/N$. This effect occurs because a correlation is induced between the transition moments of spatially separated radiators as they interact with each other through the radiation field. As a result, the atoms in a volume of macroscopic size emit coherently. Since the total energy radiated by N atoms is $N\hbar\omega_0$, where ω_0 is the transition frequency, the emission intensity is $I \sim N\hbar\omega_0/\tau_e \sim N^2$. This emission has been labeled "collective spontaneous emission" and "Dicke superradiance."[1)] For ordinary spontaneous emission, in contrast, in which case the atoms decay independently of each other, with a spontaneous-decay time T_1 which is independent of the number of radiators, the emission intensity is proportional to the number of radiators, $I \sim N\hbar\omega_0/T_1$

[1)]The effect is also referred to as "superfluorescence" in the non-Soviet literature.

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$\sim N$. (See Agarwal's book[82] for an up-to-date review of the theory of spontaneous emission.)

An effective self-induction of correlations between dipole moments can occur only if the characteristic time of this process (τ_c) is shorter than the relaxation time of the atomic dipole moment, T_2, and also shorter than T_1 (usually, $T_2 < T_1$). From the standpoint of the dynamics of the atomic subsystem, therefore, superradiance is a transient process, which occurs over times shorter than T_1 and T_2. We emphasize that this onset of correlations between radiators is an event which occurs spontaneously in the course of the emission process. This circumstance represents a fundamental distinction between superradiance and other transient coherent processes, such as the decay of free optical induction,[78] self-induced transparency,[78] and the photon echo,[79] in which cases the individual radiators are in phase, and the emission intensity is also proportional to N^2, but the phase coherence has been imposed by a coherent external pump.

The properties of superradiance and the conditions under which it can be observed are qualitatively different from those for ordinary spontaneous emission and also for stimulated emission. The distinctive features of superradiance can be seen on the example of a typical experiment carried out to observe it.[24] We assume that there are N two-level atoms in a macroscopic cylinder, of length L and case area A, which is long and open at both ends ($L \gg \sqrt{A}$, $V=AL, n=N/V$). All the atoms are initially put in the upper state by a short pump pulse ($\tau_{pump} < \tau_c$), in such a manner that the initial state of the system is noncoherent (in other words, there are no correlations between the dipole moments of the working transitions of the different atoms) (Fig. 1). Then the system of atoms with a population inversion begins to decay freely; the nature of the decay depends on the relations among the characteristic times T_1, T_2, τ_c, and also $\tau = L/c$, which is the transit time of a photon through the medium (c is the speed of light). We assume that the number density of atoms is so low that the following inequality holds:

$$T_1 = \left(\frac{4}{3}\frac{d^2\omega_0^3}{\hbar c^3}\right)^{-1} < \tau_c \sim \left(\frac{2\pi n d^2 \omega_0}{\hbar \tau^{-1}}\right)^{-1}$$

(d is the dipole matrix element of the transition). Then each atom decays independently of the others, and the energy stored in the atomic system is radiated in a characteristic time T_1 in an isotropic fashion (Fig. 2b).

We now assume

$$\tau \ll \tau_c \ll T_2, \; T_1. \tag{1.1}$$

The right-hand inequality means that the collective processes occur more rapidly than the relaxation in the individual atoms. The left-hand inequality means that the photons leave the volume under consideration in a time shorter than the characteristic time for the induction of interatomic correlations, so that stimulated processes can be ignored during the superradiance. Conditions (1.1) determine the type of superradiance. If all these conditions hold, a system of N atoms will emit a superradiance pulse with a peak intensity several orders of magnitude higher than the intensity of spontaneous emission (about ten orders of magnitude higher in the experiments of Skribanowitz *et al.*).[24] Most of the energy is radiated into small solid angles along the greatest dimension of the volume (Fig. 2c). This directionality results from interference of the different radiators and is determined by the geometric configuration of the medium. Under the condition $\tau \sim \tau_c$, some of the radiated energy reenters the atomic subsystem, and the emission takes the form of a train of pulses of decreasing height ("oscillatory superradiance"; Fig. 2c). An important characteristic of the superradiance pulse is the delay time t_0, determined from the time at which the crest of the pulse is observed (Fig. 2c); this time is roughly an order of magnitude longer than the length of the pulse itself ($t_0 \sim \tau_c \ln N$). The reason for the delay is that the decay begins with isotropic spontaneous emission, and only gradually, as the result of the interaction of atoms through the radiation field, do correlations grow among the atomic dipole moments. It is at the time $t = t_0$ that these correlations reach their maximum (at t_0, the populations of the upper and lower working levels are equal). The directionality of the superradiance, along the greatest dimension of the volume, is reminiscent of a corresponding property of the amplified spontaneous emission in mirror-free systems. In the time-dependent case, this situation is determined by the condition $T_2 < \tau_c' = (2\pi n d^2 \omega_0 / \hbar T_2^{-1})^{-1} \ll \tau$, in contrast with superradiance. The inequality at the left here means that the polarization is rapidly adjusted by the field; the inequality at the right means that the photons remain in the medium for a time τ sufficient for an avalanche increase in the stimulated

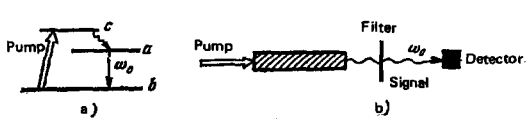

FIG. 1. a) Working-level diagram; b) typical experimental arrangement for observing superradiance. a, b) Working levels; c) intermediate level.

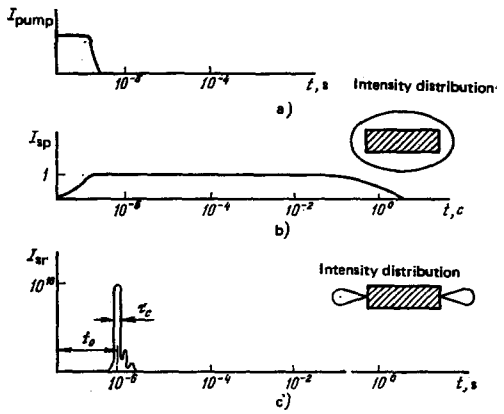

FIG. 2. Comparison of superradiance and noncoherent spontaneous emission.[24] The time scale is logarithmic. a) Pump pulse which creates a population inversion for the working transition, $a \to b$; b) emission intensity in the case of noncoherent spontaneous decay ($T_1 \sim 1s$): a slow exponential decay with an isotropic directional distribution of the intensity; c) the observed highly directional superradiance signal (in gaseous HF; Ref. 24). The peak intensity I_{sr} is roughly 10^{10} times I_{sp}.

emission (which occurs over a time $\sim \tau_c'$). We might note that in the literature the amplification of spontaneous emission is also frequently referred to as "superradiance" (or "superluminescence"). Superradiance, a collective spontaneous process, differs from this other process in a fundamental way: Stimulated atomic transitions play no role in superradiance. To see the distinction between superradiance and stimulated emission, we consider the example of a single-pass mirror-free laser, with a geometric configuration and a working-level arrangement the same as for the superradiance which we have been discussing (Fig. 1a). Now, however, the pump acts continuously. At a large gain, $\mu L = 2\pi n d^2 \omega_0 L/\hbar T_2^{-1} c \gg 1$, the system of two-level atoms will be in a saturation state (the populations of the upper and lower working levels will be equal, at $N/2$). In this state, a maximum fraction of the energy stored in the atomic medium is converted into radiation. Since the rate at which atoms are pumped to the upper working level is proportional to $N/2$, the output intensity of this laser (in the steady state) is also proportional to N. Consequently, if we change (for example) the pressure of the working gas, then the intensity of the amplified spontaneous emission will vary in proportion to the pressure, while the intensity of superradiance will vary in proportion to the square of the pressure. This circumstance is exploited experimentally to identify superradiance.

Superradiance is of both general physical interest and applied interest. From the physical standpoint, superradiance is an example of cooperative behavior of a system of N particles ($N \gg 1$) which are interacting with an electromagnetic field. The formation of the superradiant correlated state in a many-body system of this type, the role played by the geometry of the medium in shaping the spatial coherence of the superradiance, and the relationship between superradiance effects and nonequilibrium phase transitions—all these questions are of general interest.

From the standpoint of applications, superradiance is interesting as one method for producing coherent emission in a system without mirrors. The difficulties in devising effective mirrors for the x-ray and γ ranges have prevented the use of ordinary stimulated-emission processes for generating short-wave emission. Consequently, superradiance may prove successful as a mechanism for generating coherent emission in these ranges (this possibility is supported by the theory of Refs. 48–52).

Superradiance can also be exploited to find spectroscopic information,[57,58] to generate ultrashort pulses, etc.

For a long time after the publication of Dicke's paper, new theoretical work on the subject appeared only sporadically. This period of relative inactivity gave way to a new stage of intense theoretical and experimental work on superradiance just recently, after the first experimental observation of superradiance in the optical range.[24] A large number of experiments have now been carried out to observe superradiance, and their results have shown that the existing theories fall short of satisfactory agreement with experiments in several important ways. As a result, further effort has gone into deriving a theory for the superradiance of extended systems (with dimensions much greater than the radiation wavelength). Among the topics which have been treated theoretically are multimode superradiance, superradiance in weakly amplifying media, and oscillatory superradiance. The theory has been developed further through a generalization to the case of two-photon processes (Raman-scattering superradiance in molecular and atomic media).[62,64,22] Sczaniecki and Buchert[80] report that it is also possible in principle to observe superradiance in many-photon processes.

It should be pointed out that several approaches have been taken toward a theoretical description of superradiance, with significant differences in initial assumptions. This situation is a consequence of the complexity of the problem in general formulation (the nonlinearity and distributed nature of the atomic system, the need to take into account the geometric configuration of the medium, etc.). Approaches which focus on only one aspect of the phenomenon may not describe other important aspects. In this review we will accordingly discuss or briefly summarize all the basic approaches to a description of superradiance, and we will compare the theoretical results with experiment.

2. SUPERRADIANCE OF SYSTEMS WITH DIMENSIONS SMALLER THAN THE WAVELENGTH

The system of two-level atoms interacting with radiation which we discussed above is described by the Hamiltonian

$$H = \frac{\hbar\omega_0}{2} R_3 + \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}} a_{\mathbf{k}}^+ a_{\mathbf{k}} + \sum_{\mathbf{k}} (g_{\mathbf{k}} R_{\mathbf{k}}^+ a_{\mathbf{k}} + g_{\mathbf{k}}^* a_{\mathbf{k}}^+ R_{\mathbf{k}}^-) = H_0 + \sum_{\mathbf{k}} H_{\mathbf{k}}, \quad (2.1)$$

where $a_{\mathbf{k},\lambda}^+$ ($a_{\mathbf{k},\lambda}$) is the creation (annihilation) operator for a field quantum in the mode $(\mathbf{k}, \lambda)$, with wave vector $\mathbf{k}$, frequency $\omega_{\mathbf{k}}$, and polarization $\mathbf{e}_\lambda$. The coupling constant $g_{\mathbf{k},\lambda}^+$ is

$$g_{\mathbf{k},\lambda} = \sqrt{\frac{2\pi\hbar}{V\omega_{\mathbf{k}}}} \langle + | \mathbf{j}^+ \mathbf{e}_\lambda | - \rangle, \quad (2.1a)$$

where $\mathbf{j}^+$ is the operator representing the transition current density (in the dipole approximation, $\mathbf{j}^+ = i\omega_0 \mathbf{d}$), and $|+\rangle$ and $|-\rangle$ are the wave functions of the excited and ground states of the atom respectively. We will be omitting the polarization index λ, adopting the notation $\mathbf{k} \equiv (\mathbf{k}, \lambda)$.

The collective atomic operators $R_{\mathbf{k}}^\pm$ and R_3 can be expressed in terms of the Pauli spinors $\sigma_\pm = (\sigma_1 \pm i\sigma_2)/2$ of the individual atoms:

$$R_{\mathbf{k}}^\pm = \sum_{i=1}^N \sigma_\pm^{(i)} \exp(\pm i\mathbf{k}\mathbf{r}_i), \qquad R_3 = \sum_{i=1}^N \sigma_3^{(i)}. \quad (2.2)$$

These operators satisfy the commutation relations

$$[R_{\mathbf{k}}^+, R_{\mathbf{k}'}^-] = R_{3,\mathbf{k}-\mathbf{k}'}, \qquad [R_{\mathbf{k}}^\pm, R_3] = \mp 2R_{\mathbf{k}}^\pm, \quad (2.3)$$

where

$$R_{3\mathbf{k}} = \sum_{i=1}^N \sigma_3^{(i)} \exp(i\mathbf{k}\mathbf{r}_i).$$

The intensity of the emission into a unit solid angle around the direction $\mathbf{k}$ is expressed in terms of the cor-

relators of the collective Heisenberg operators,[1]

$$\frac{d}{dt}\langle a_k^+ a_k\rangle = I_k(t) = I_{sp}\langle R_k^+(t)\, R_k^-(t)\rangle, \tag{2.4}$$

where $I_{sp}=1/4\pi T_1$ is the intensity of the isotropic spontaneous decay per unit solid angle, and the angle brackets denote the quantum-statistics average. Here and below, the intensities are expressed in photons per second.

Before we take up the dynamics of the correlators $\langle R_k^+ R_k^-\rangle$ in the general case of extended systems, we will illustrate the basic ideas by examining the simple case in which the volume of the system satisfies $V \ll \lambda^3$, where λ is the radiation wavelength. Then for each atom we have $\mathbf{k}\cdot\mathbf{r}_i \ll 1$ in (2.1), and this circumstance can be taken into account formally by letting $\mathbf{k}\to 0$ in (2.2)–(2.3) and by using the replacement $R_k^\pm \to R_0^\pm$ in (2.4).

We introduce the collective operator

$$R^2 = \frac{1}{2}(R_0^+ R_0^- + R_0^- R_0^+) + \frac{1}{4}R_3^2, \tag{2.5}$$

and we describe the atomic system by means of the eigenfunctions of the operator R^2 (the Dicke states[1]):

$$R^2 \mid r, m\rangle = r(r+1)\mid r, m\rangle,\quad R_3 \mid r, m\rangle = 2m \mid r, m\rangle,$$

where $m=(N_a - N_b)/2$ is half the difference between the populations ($N_a + N_b = N$), $0 \le r \le N/2$, $|m| \le r$. For radiative transitions, the cooperation number r does not change, since R^2 commutes with the Hamiltonian in (2.1) (where $\mathbf{k}=0$).

According to (2.4), the emission intensity at the time $t=0$ depends on the initial state of the atomic system. Let us assume that at $t=0$ the atoms have a population inversion: $m=r=N/2$. The intensity of the radiation from this state is

$$I_k = I_{sp}\langle r, m \mid R_0^+ R_0^- \mid r, m\rangle = I_{sp}(r+m)(r-m+1) = I_{sp}N;$$

i.e., each atom begins to decay independently of the others, and the intensities of the various radiators are summed.

We now assume that the system is in the state $r=N/2$, $m=0$ (the populations of the upper and lower levels are equal). The radiation intensity in this state is

$$I_k = \left\langle \frac{N}{2}, 0 \mid R_0^+ R_0^- \mid \frac{N}{2}, 0\right\rangle = I_{sp}\frac{N}{2}\left(\frac{N}{2}+1\right) \approx I_{sp}\frac{N^2}{4}.$$

The decay of a system of N atoms from a Dicke state with $r=N/2$ and $m=0$ thus occurs in a cooperative manner, and the emission intensity is at a maximum and is proportional to N^2 (a superradiative state).

The superradiative state can be produced by irradiating a system of atoms in the ground state ($r=N/2$, $m=-N/2$) with an intense pulse of resonant radiation, which causes the populations to become equal at times $t \ll T_2$.

Here, however, we are interested in a different process: the decay of a system of N atoms which initially has a complete population inversion ($r=m=N/2$ at $t=0$), accompanied by a spontaneous transition to a superradiative state.

To describe this process, we make use of the circumstance that the operator in (2.5) commutes with the Hamiltonian in (2.1) (where $R_k^\pm \to R_0^\pm$), so that its expectation value is conserved at times $t < T_2$:

$$\langle R^2\rangle_t = \frac{1}{2}\langle R_0^+ R_0^- + R_0^- R_0^+\rangle_t + \frac{1}{4}\langle R_3^2\rangle = \langle R^2\rangle_{t=0} \equiv r^2, \tag{2.6}$$

where

$$r^2 = \frac{N}{2}\left(\frac{N}{2}+1\right).$$

Equation (2.6) is known as the "conservation law for the length of the Bloch vector." Now using the commutation rules in (2.3) and the expression for the intensity in (2.4), and ignoring the mean square fluctuation $\langle \delta R_3^2\rangle$ (at $t=0$, with a complete noncoherent population inversion, $\langle \delta R_3^2\rangle = 0$), we obtain from (2.6)

$$T_1 4\pi I_k + \frac{\langle R_3\rangle^2}{4} - \frac{\langle R_3\rangle}{2} = \frac{N}{2}\left(\frac{N}{2}+1\right). \tag{2.7}$$

One of the variables, $I_k(t)$ or $\langle R_3(t)\rangle$, can be eliminated by making use of the fact that energy is conserved during the emission. Using (2.4) (where $R_k^\pm \to R_0^\pm$), we can write this conservation law as

$$\frac{d}{dt}\left\langle \frac{R_3}{2} + \sum_k a_k^+ a_k\right\rangle = \frac{d\langle R_3\rangle}{2dt} + 4\pi I_k = 0. \tag{2.8}$$

Then from (2.8) and (2.7) we obtain an equation which describes the dynamics of the difference between the populations of the atomic levels,

$$\frac{d\langle R_3\rangle}{dt} = -\frac{2}{T_1}\left(\frac{N^2}{4} - \frac{\langle R_3\rangle^2}{4} + \frac{N+\langle R_3\rangle}{2}\right) \tag{2.9}$$

with the initial condition $\langle R_3\rangle_{t=0} = N$. It solution (within terms of order $1/N$) is

$$\langle R_3\rangle_t = -N \operatorname{th}\frac{t-t_0}{2\tau_c}, \tag{2.10}$$

where

$$\tau_c = \frac{T_1}{N}, \tag{2.11}$$

$$t_0 = \frac{T_1}{N}\ln N = \tau_c \ln N. \tag{2.12}$$

The populations of the upper and lower levels become equal at $t=t_0$.

For the intensity of the radiation into a unit solid angle, we find from (2.7) and (2.10)

$$I_k(t) = \frac{I_{max}}{4\pi}\operatorname{sech}^2\frac{t-t_0}{2\tau_c}, \tag{2.13}$$

where

$$I_{max} = \frac{N^2}{4T_1}.$$

The system of N atoms emits in an arbitrary direction $\mathbf{k}$ a pulse whose intensity reaches a maximum $I_{max}/4\pi \sim N^2$ (superradiance) at the time t_0 (the delay time). The length of the pulse is τ_c. Since $N \gg 1$, we have $\ln N \gg 1$ in (2.12) (in a real situation, we could have $\ln N \approx 20$), so that the condition $t_0 \gg \tau_c$ holds. It is thus t_0 which determines the characteristic time interval for emission of the system. Equation (2.12) for t_0 was first derived by Fain.[2] We recall that in deriving (2.13) we used the conservation of the length of the Bloch vector, (2.6) ($t < T_2$), so that one of the conditions for superradiance is the inequality $t_0 < T_2$.

We have briefly mentioned the wave function of a system of N atoms which corresponds to a superradiative state. In Refs. 3 and 4, where a group-theory approach to superradiance was developed on the basis of a permutation group, it was shown that the superradiative state corresponds to a wave function of a system of N identical atoms which is completely symmetric with re-

spect to interchange of particles. A wave function which is completely antisymmetric with respect to the interchange of particles corresponds to a state with a zero decay rate; in other words, radiation would be trapped in such a state.

Shelepin[5] was the first to take up the problem of superradiance for a many-level system. The number of papers on this subject is now quite large, and the present state of the question is covered thoroughly in Ref. 6.

It should be noted that superradiance has not yet been observed in systems significantly shorter than the radiation wavelength. One possible reason is that dipole–dipole interactions broaden the line to the extent that the condition $t_0 < T_2$ may not hold for such systems. In a superradiative state, the dipole–dipole level width is $(1/T_2)_{\text{pump}} \sim Nd^2/\hbar r_{\text{av}}^3$, where r_{av} is the average linear dimension of the system; from (2.12), on the other hand, we find $1/t_0 = -N/T_1 \ln N \approx [(2\pi^3)N/\ln N]d^2/\hbar\lambda^3 \ll (1/T_2)_{\text{pump}}$ if $r_{\text{av}} \ll \lambda$. The effect of the shape of the sample the dipole relaxation rate in systems with $V \ll \lambda^3$ has been discussed by Friedberg *et al.*[7,8]

That superradiance can be observed in samples with dimensions comparable to the radiation wavelength has been demonstrated experimentally by Gross *et al.*,[9] who observed the effect in the Rydberg levels of the sodium atom (in a transition from an nS level with a principal quantum number $n = 25$). The emission had a wavelength of $\lambda = 1.5$ mm, and the active region had dimensions $L = 5$ mm and $d = 1$ mm.

3. SUPERRADIANCE OF EXTENDED SYSTEMS

a) General considerations

Developments in quantum electronics spurred much new interest in superradiance, as the appearance of sources of high-power coherent light pulses, with pulse lengths shorter than the relaxation time for atomic transitions, made it possible to study in the optical range several effects which had previously been observable only in the microwave range, among them the photon echo, optical nutation, and optical adiabatic inversion. Common to all these effects is a macroscopic induced transition moment which arises because the radiators come to act in phase. As mentioned above, it is the onset of correlations between the transition momoments of the radiators in volumes of macroscopic size during the decay which is responsible for superradiance. In order to evaluate the possibility of observing superradiance in the optical range, it became necessary to generalize the theory to the case of large radiating volumes. This problem has been taken up in many papers. Afanas'ev and Kagan,[10] for example, discussed the possibility of observing the effect in the γ range, and they estimated the maximum emission intensity. The time evolution of superradiance in extended systems was described in Refs. 11–16, where various approaches led to similar equations for the dynamics of superradiative systems.

For extended systems, the Hamiltonian of a system of N two-level atoms which are interacting with an electromagnetic field is given by Eq. (2.1).

By analogy with the case of systems with dimensions smaller than the wavelength, we introduce the operator

$$R_{\mathbf{k}}^2 = \frac{1}{2}(R_{\mathbf{k}}^+ R_{\mathbf{k}}^- + R_{\mathbf{k}}^- R_{\mathbf{k}}^+) + \frac{1}{4} R_3^2. \tag{3.1}$$

As we have seen, the possibility of superradiance stems from the appearance of correlations $\langle R_{\mathbf{k}}^+ R_{\mathbf{k}}^- \rangle = \sum_{i \neq j} \times e^{i\mathbf{k}(\mathbf{r}_i - \mathbf{r}_j)} \langle \sigma_+^{(i)} \sigma_-^{(j)} \rangle$; i.e., the correlators of the individual radiators, $\langle \sigma_+^{(i)} \sigma^{(j)} \rangle$, must be nonzero in a volume of macroscopic size. The geometric configuration of the volume occupied by the atoms is important in determining the correlators. To see this, we assume that the volume is isotropic in shape, so that all the field modes $\mathbf{k}$ are equivalent; i.e., the $\langle R_{\mathbf{k}}^+ R_{\mathbf{k}}^- \rangle$ are independent of the direction of $\mathbf{k}$. Since the medium is homogeneous, we can write $\langle \sigma_+^{(i)} \sigma_-^{(j)} \rangle = f(\mathbf{r}_{ij})$, $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$, and thus $\langle R_{\mathbf{k}}^+ R_{\mathbf{k}}^- \rangle \sim \int d\mathbf{r}_{ij} f(\mathbf{r}_{ij}) \exp(i\mathbf{k}\mathbf{r}_{ij})$. Then it follows that

$$\langle \sigma_+^{(i)} \sigma_-^{(j)} \rangle \sim \int d\mathbf{k} e^{-i\mathbf{k}\mathbf{r}_{ij}} \langle R_{\mathbf{k}}^+ R_{\mathbf{k}}^- \rangle \sim \frac{\sin(kr_{ij})}{kr_{ij}};$$

i.e., this correlator falls off in inverse proportion to the distance between radiators. If, on the other hand, we discriminate among radiation modes, for example, by using a long, cylindrical sample, so that modes with wave vectors directed along the axis of the cylinder play a governing role in the emission [see Eq. (3.14) below], the correlations between radiators will span regions of macroscopic size.

In the extreme case of a single mode $\mathbf{k}$, the individual radiators will be correlated in a volume whose longitudinal dimension along the $\mathbf{k}$ direction, l_{eff}, is determined by the uncertainty in the wave vector, $\Delta k = 2\pi/l_{\text{eff}}$. The difference Δk may stem from several factors. First, it may result from an inhomogeneous or homogeneous width of the emission line, $l_{\text{eff}_1} = cT_2$. Second, since the photon is emitted within a region L (L is the length of the sample), it has a momentum uncertainty $\Delta k = 2\pi/L$, so that the corresponding value of l_{eff_2} is equal to L. Furthermore, if a radiation pulse of length τ_e results from the decay of the system, the corresponding value is $l_{\text{eff}_3} = c\tau_e$ (Ref. 16). The maximum size of the coherence region— the region within which the radiators can be put in phase—is thus determined by the length of the system. This maximum value is reached in systems with $L/c < \tau_e, T_2$.

Ressayre and Tallet[17] have shown that the situation in a cylindrical sample with Fresnel numbers $F = A/\lambda L \sim 1$ corresponds approximately to the single-mode case. The single-mode model has been used widely to describe the superradiance in extended systems because of its simplicity.[14,18,19] Let us review the basic results obtained from this model.

b) Single-mode model for superradiance

In the single-mode approximation [in which there is only one definite mode $\mathbf{k}$ in (2.1)], a theory can be constructed for superradiance in complete analogy with the case of a system with dimensions smaller than the wavelength. The commutator for the operator $R_{\mathbf{k}}^2$ in (3.1) with the Hamiltonian in (2.1) is

$$[R_{\mathbf{k}}^2, H] = -2i \sum_{\mathbf{k}'} \sum_{i \neq j} [g_{\mathbf{k}} \sigma_+^{(i)} \sigma_3^{(j)} a_{\mathbf{k}'} \exp\left(i\frac{\mathbf{k}}{2}\mathbf{r}_{ij} + i\frac{\mathbf{k}'}{2}\rho_{ij}\right) - g_{\mathbf{k}}^* a_{\mathbf{k}}^+ \sigma_3^{(j)} \sigma_-^{(i)} \exp\left(-i\frac{\mathbf{k}}{2}\mathbf{r}_{ij} - i\frac{\mathbf{k}'}{2}\rho_{ij}\right)] \sin\frac{\mathbf{k}-\mathbf{k}'}{2}\mathbf{r}_{ij},$$

where

$$\mathbf{r}_{ij}=\mathbf{r}_i-\mathbf{r}_j,\quad \boldsymbol{\rho}_{ij}=\mathbf{r}_i+\mathbf{r}_j.$$

It follows that in the single-mode approximation ($g_{\mathbf{k}}=g_{\mathbf{k}}\delta_{\mathbf{k}\mathbf{k}'}$) the operator $R_{\mathbf{k}}^2$ commutes with $H_{\mathbf{k}}$ and is conserved (over times $t<T_2$).

For $R_{\mathbf{k}}^2$ we thus find an analog of the conservation law for the length of the Bloch vector, again in the form of (2.6), where we must use the substitution $R_0^2\to R_{\mathbf{k}}^2$. Then in (2.8) we must allow for the fact that in this case the radiation is now emitted into a small solid angle $\Delta\Omega_{\mathbf{k}}\simeq\lambda^2/A$ determined by diffraction instead of into a solid angle of 4π. This small solid angle is around the axis of the cylinder (which is parallel to $\mathbf{k}$):

$$\frac{\mathrm{d}}{dt}\frac{\langle R_3\rangle}{2}+\Delta\Omega_{\mathbf{k}}I_{\mathbf{k}}=0. \tag{3.2}$$

This means that Eq. (2.9) remains valid for an extended system in the single-mode case if we use the substitution

$$\frac{1}{T_1}\to\frac{1}{T_1}\frac{\Delta\Omega_{\mathbf{k}}}{4\pi}. \tag{3.2'}$$

Repeating the arguments which were made for the case of a system with dimensions smaller than a wavelength, we find that the intensity of the radiation into the solid angle $\Delta\Omega_{\mathbf{k}}$ is

$$I(t)=I_{\mathbf{k}}\Delta\Omega_{\mathbf{k}}=\frac{N}{4\tau_c}\operatorname{sech}^2\frac{t-t_0}{2\tau_c}, \tag{3.3}$$

where

$$\tau_c=\frac{4\pi A}{\lambda^2}\frac{T_1}{N}, \tag{3.4}$$

$$t_0=\tau_c\ln N. \tag{3.5}$$

In the case of an extended system, the pulse length τ_c and the delay time t_0 are thus greater by a factor $4\pi A/\lambda^2\gg1$ than in the case of a system with dimensions smaller than a wavelength [cf. (2.11) and (2.12)]. A more rigorous derivation[14b] of Eq. (3.3) shows that this equation holds only if $l_c=c\tau_c>L$, in which case the change in the envelope of the superradiance pulse along the medium can be ignored.

In the semiclassical approximation, the expression for $R_{\mathbf{k}}^2$ in (3.1) can be converted to the form $R_{\mathbf{k}}^2=R_x^2+R_y^2+R_z^2$ by using the change of variables $R_{\mathbf{k}}^+=R_x+iR_y$, $R_{\mathbf{k}}^-=R_x-iR_y$, $\frac{1}{2}R_3=R_z$. The conservation of $R_{\mathbf{k}}^2$ over times $t<T_2$ allows us to describe the superradiative decay formally as the motion of a vector of constant length with components R_x, R_y, R_z (a Bloch vector) along the surface of a sphere of radius r. Introducing the new variable θ (the azimuthal angle) by means of $R_z=-r\cos\theta$; using (2.9), where we ignore the term $(N+R_3)/2$; and using (3.2'), we obtain

$$\frac{\mathrm{d}\theta}{dt}+\frac{1}{\tau_c}r\sin\theta=0. \tag{3.6}$$

This equation, which is frequently used, describes superradiative decay except near an initial state of complete inversion $\theta_{t=0}=\theta_0=\pi$. The description of superradiance in the Bloch representation will be reviewed more thoroughly in Subsection 3e.

c) Quantum multimode theory of superradiance

The single-mode model of superradiance is simple, graphic, and a close analogy of a system with dimensions smaller than a wavelength. On the other hand, it does not tell us about many important characteristics of superradiance (in particular, the very condition for the applicability of the single-mode approximation). Some important questions which cannot be studied in the single-mode theory are how the non-conservation of the length of the Bloch vector over short time intervals affects the superradiance delay, the angular distribution of the superradiance, the role played by the geometric configuration of the medium in forming the superradiance, and the relationship between superradiance and nonequilibrium phase transitions. We must turn to the multimode theory for a complete description of superradiance.

A multimode theory can be formulated either in the atomic–field representation, in which case we would use the dynamic equations for both the atomic and field subsystems, which are interacting with each other; or in the purely atomic representation, in which case the field variable would be eliminated through a solution of the space–time Maxwell equation.[15,17,22]

In this subsection we will take the first of these approaches, which is the one commonly used in the quantum theory of radiation. The second approach, which is more convenient, particularly for determining the relationship between superradiance and nonequilibrium phase transitions, will be discussed in Section 6 for the case of superradiance in two-photon processes (Raman scattering).

1. *Conditions for the occurrence of the effect.* The Hamiltonian in (2.1) with the relaxation terms, the commutation relations in (2.3), and the commutation rules for the operators $a_{\mathbf{k}}^+$ and $a_{\mathbf{k}}$ lead to the following system of equations in the mode representation for an atomic–field system which is interacting with its surroundings[18,20]:

$$\left.\begin{aligned}
&\frac{dn_{\mathbf{k}}}{dt}+\frac{n_{\mathbf{k}}}{\tau_{\mathbf{k}}}=F_{\mathbf{k}},\\
&\frac{dF_{\mathbf{k}}}{dt}+\frac{1}{2}\left(\frac{1}{\tau_{\mathbf{k}}}+\frac{1}{T_2}\right)F_{\mathbf{k}}=P_{\mathbf{k}}+\frac{1}{T_{\mathbf{k}}^2}(S_0+S_{\mathbf{k}})+i(\omega_{\mathbf{k}}-\omega_0)G_{\mathbf{k}},\\
&\frac{dG_{\mathbf{k}}}{dt}+\frac{1}{2}\left(\frac{1}{\tau_{\mathbf{k}}}+\frac{1}{T_2}\right)G_{\mathbf{k}}=i(\omega_{\mathbf{k}}-\omega_0)F_{\mathbf{k}},\\
&\frac{dS_{\mathbf{k}}}{dt}+\frac{S_{\mathbf{k}}}{T_2}=F_{\mathbf{k}}R_3,\\
&\frac{dR_3}{dt}+\frac{N+R_3}{T_1^*}=-2\sum_{\mathbf{k}}F_{\mathbf{k}},
\end{aligned}\right\} \tag{3.7}$$

where $n_{\mathbf{k}}=\langle a_{\mathbf{k}}^+a_{\mathbf{k}}\rangle$ is the number of photons in mode $\mathbf{k}$, $\tau_{\mathbf{k}}=L_{\mathbf{k}}/c$, $L_{\mathbf{k}}$ is the length of the sample along the direction of the vector $\mathbf{k}$ (the term $n_{\mathbf{k}}/\tau_{\mathbf{k}}$ effectively allows for the radiation which leaves the volume),

$$\left.\begin{aligned}
&\frac{1}{T_{\mathbf{k}}^2}=\frac{2|g_{\mathbf{k}}|^2}{\hbar^2},\qquad R_3=\langle R_3\rangle,\\
&F_{\mathbf{k}}=\frac{2i}{\hbar}(g_{\mathbf{k}}\langle a_{\mathbf{k}}^+R_{\mathbf{k}}^-\rangle-g_{\mathbf{k}}^*\langle a_{\mathbf{k}}R_{\mathbf{k}}^+\rangle),\\
&P_{\mathbf{k}}=\frac{2}{\hbar^2}\sum_{\mathbf{k}'}[g_{\mathbf{k}}g_{\mathbf{k}'}^*\langle a_{\mathbf{k}}^+a_{\mathbf{k}'}\rangle R_{3,\,\mathbf{k}-\mathbf{k}'}+g_{\mathbf{k}'}g_{\mathbf{k}}^*\langle a_{\mathbf{k}'}^+a_{\mathbf{k}}\rangle R_{3,\,\mathbf{k}'-\mathbf{k}}],\\
&S_0=\sum_{i=1}^{N}\langle\sigma_+^{(i)}\sigma_-^{(i)}\rangle,\qquad S_{\mathbf{k}}=\sum_{i\neq j}\langle\sigma_+^{(i)}\sigma_-^{(j)}\rangle\exp(i\mathbf{k}\mathbf{r}_{ij}),
\end{aligned}\right\} \tag{3.8}$$

and $1/T_1^*$ is the rate of radiationless relaxation of the population difference. An equation for $G_{\mathbf{k}}$, can be found from (3.8) by changing the sign of $g_{\mathbf{k}}^*$.

If the atomic system is homogeneously broadened,

then $\omega_k \approx \omega_0$ and $G_k = 0$, and we will adopt these assumptions everywhere below except in Subsection 3e. The system of equations in (3.7) simplifies substantially if

$$R_{3,\,k-k'} = R_3 \delta_{k,\,k'}. \tag{3.9}$$

This condition holds quite accurately if the field amplitude varies only slightly over the length of the sample, i.e., if the pulse length τ_c is much longer than the transit time τ. In this case we have $\langle \sigma_3^{(i)} \rangle \approx \langle \sigma^{(j)} \rangle$ for any values of i and j, so that $\langle R_{3,\,k-k'} \rangle = N \langle \sigma_3 \rangle \delta_{kk'}$, since $\sum_i \exp[i(\mathbf{k}-\mathbf{k}')\mathbf{r}_i] = \delta_{kk'}$. In systems satisfying the condition $\tau \ll \tau_c$, the second equation in system (3.7) thus becomes

$$\frac{dF_k}{dt} + \frac{1}{2}\left(\frac{1}{\tau_k} + \frac{1}{T_2}\right) F_k = \frac{1}{T_k^2}(n_k R_3 + S_0 + S_k). \tag{3.10}$$

Let us examine the right side of Eq. (3.10) in more detail. The variable F_k is the rate at which the energy stored in the atomic system is transferred to the k-th field mode (or in the opposite direction, depending on the sign of F_k). This energy exchange occurs through induced emission ($n_k R_3$), the spontaneous-decay processes described by the term $S_0 = \sum_{i=1}^{N} \langle \sigma_+^{(i)} \sigma_-^{(j)} \rangle = (N + R_3)/2$ (which is equal to the number of excited atoms), and collective spontaneous emission, described by the term S_k. The term S_k results from a correlation which arises among the various radiators as light is emitted from the system. It is this correlation which can lead, under certain conditions, to a cooperative decay of the system.

The first two equations in system (3.7), (3.10) show that the time evolution $n_k(t)$ is determined by the right side of Eq. (3.10), i.e., by the resultant effect of induced, spontaneous, and collective processes. Let us compare the effects of induced and collective processes on the emission intensity. From the first and fourth equations in system (3.7), (3.10) we have

$$n_k R_3 + S_k = \int_0^t F_k(t') \left\{ R_3(t) \exp\left[-\frac{t-t'}{\tau}\right] + R_3(t') \exp\left[-\frac{t-t'}{T_2}\right] \right\} dt'. \tag{3.11}$$

In systems with $\tau = L/c \ll T_2$, the emission intensity is thus determined primarily by collective processes (S_k) (since in this case the first term in braces is much smaller than the second at times $t > \tau$), while in systems with $\tau \gg T_2$ the induced processes are predominant.

We have thus found two conditions which must be satisfied if we wish to observe superradiance: First, the length of the sample must satisfy the condition $L \ll cT_2$ (the dimensions of the coherence region are maximized when the condition $L \ll l_c$ also holds). Second, as was shown above, there must be a discrimination among radiation modes.

To see how we can arrange this discrimination, we consider the kinetics of the appearance of superradiative states in a completely excited and initially uncorrelated system of two-level atoms. In this case, system (3.7), (3.10) must be supplemented with the initial conditions

$$n(0) = 0, \quad F(0) = 0, \quad S(0) = 0, \quad R_3(0) = N. \tag{3.12}$$

The set of eigenvalues of system (3.7)–(3.10), which is linear in the initial stage, while the condition $N - R_3 \ll R_3$ holds, is given by $[n_k(t) \sim e^{\lambda_k t}]$

$$\lambda_{1,\,k} = -\frac{1}{2}\left(\frac{1}{\tau_k} + \frac{1}{T_2}\right), \quad \lambda_{2,\,3k} = \lambda_{1,\,k} \pm \sqrt{\frac{1}{4}\left(\frac{1}{\tau_k} - \frac{1}{T_2}\right)^2 + \frac{2N}{T_k^2}}. \tag{3.13}$$

Exponentially growing solutions thus arise only if

$$\mu_0 L_k \equiv \frac{2N\tau_k T_2}{T_k^2} = \frac{\lambda^2}{4\pi}\frac{N}{V}\frac{T_2}{T_1} L_k > 1. \tag{3.14}$$

This mode discrimination can thus be arranged by using a needle-shaped sample. In this case, axial modes, for which $L_k \approx L$, where L is the length of the sample, will play a governing role.

Let us assume that the number of axial (working) modes is $\varkappa_0$. Summing separately over the working and the nonworking modes in (3.7), we find a system of equations in which the working and nonworking modes are coupled only by the equation for the population difference:

$$\frac{dR_3}{dt} + \frac{N + R_3}{T_1^3} = -2(F^{(1)} + F^{(2)}), \tag{3.15}$$

where $F^{(1)}$ and $F^{(2)}$ are the sums of F_k over the working and nonworking modes, respectively. In the first part of the equation for $F^{(2)}$, the term with S_0 is dominant. In systems with $\tau \ll T_2$, the induced term is much smaller than the collective term, according to (3.11). Let us compare the terms $\sum_{k'} S_0$ and $\sum_{k'} S_{k'}$ in order of magnitude, where k' are the wave vectors of the nonworking modes. The first of these sums is equal to the product of N_2 and the number of nonworking modes [in long samples, the number of nonworking modes is essentially equal to the total number of modes; see (3.44)]:

$$\sum_{k'} S_0 \sim \frac{Vk^2}{2\pi L_{\text{eff}}} N,$$

where L_{eff} is the effective length of the sample for the nonworking modes. For the second term we find

$$\sum_{k'} S_{k'} \ll \sum_{ij} \frac{\sin kr_{ij}}{kr_{ij}} \sim \left(\frac{N}{V}\right)^2 \cdot \frac{4\pi V}{k^3}.$$

In systems with $N/V \sim 10^{10}$ cm^{-3} (this condition usually holds in experiments carried out to observe superradiance; see Table I in Section 5), with $\lambda \lesssim 10^{-2}$ cm, we have

$$\sum_{k'} S_{k'} \ll \sum_{k'} S_0.$$

Then it is not difficult to see that the expression for $F^{(2)}$ is

$$F^{(2)} = \frac{S_0}{T_1'}, \tag{3.16}$$

where $1/T_1'$ is the rate of the isotropic spontaneous decay into a solid angle of 4π (after the effect of the working modes has been subtracted), and system (3.15) can be rewritten as

$$\left.\begin{aligned} \frac{dn}{dt} + \frac{n}{\tau} &= F, \\ \frac{dF}{dt} + \frac{1}{2}\left(\frac{1}{\tau} + \frac{1}{T_2}\right) F &= \frac{1}{T_0^2}(nR_3 + \varkappa_0 S_0 + S), \\ \frac{dS}{dt} + \frac{S}{T_2} &= FR_3, \\ \frac{dR_3}{dt} + \frac{N + R_3}{T_{1\,\text{eff}}} &= -2F, \end{aligned}\right\} \tag{3.17}$$

where $1/T_{1\,\mathrm{eff}} = (1/T_1^*) + (1/T_1')$ is the effective rate of the noncoherent decay; all the variables refer to the working modes; $1/T_0^2 = \sum_{\mathbf{k}_\varkappa}(1/T_{\mathbf{k}}^2)$; and $\mathbf{k}_{\varkappa_0}$ are the wave vectors of the working modes.

To determine the number of working modes, we sum the values of $S_{\mathbf{k}}$, which appears in (3.7) and which determines the directionality of the emission, according to (2.4):

$$\sum_{\mathbf{k}} S_{\mathbf{k}} = \sum_{\mathbf{k}} \sum_{i \neq j} \langle \sigma_+^{(i)} \sigma_-^{(j)} \rangle\, e^{i\mathbf{k}(\mathbf{r}_i - \mathbf{r}_j)} = \sum_{i \neq j} C_{ij} \langle \sigma_+^{(i)} \sigma_-^{(j)} \rangle, \tag{3.18}$$

where $C_{ij} = \sin k_0 r_{ij}/k_0 r_{ij}$ is the interaction matrix, and $k_0 = \omega_0/c$.

To determine the number of modes which effectively contribute to (3.18), we adopt the procedure of Ref. 17, introducing the eigenfunctions $\psi_\lambda(\mathbf{r}_i)$ and the eigenvalues λ of the matrix C_{ij}:

$$\sum_{j=1}^{N} C_{ij} \psi_\lambda(\mathbf{r}_j) = \lambda \psi_\lambda(\mathbf{r}_i). \tag{3.19}$$

The eigenfunctions $\psi_\lambda(\mathbf{r}_i)$ satisfy completeness and orthonormality conditions:

$$\sum_{i=1}^{N} \psi_\lambda(\mathbf{r}_i)\, \psi_{\lambda'}(\mathbf{r}_j) = \delta_{\lambda\lambda'}, \quad \sum_{\lambda} \psi_\lambda(\mathbf{r}_i)\, \psi_\lambda(\mathbf{r}_j) = \delta_{ij}. \tag{3.20}$$

From (3.19) and (3.20) we obtain

$$C_{ij} = \sum_{\lambda} \lambda \psi_\lambda(\mathbf{r}_i)\, \psi_\lambda(\mathbf{r}_j). \tag{3.21}$$

Using this expression in (3.18), we obtain

$$\sum_{\mathbf{k}} S_{\mathbf{k}} = \sum_{\lambda} \lambda \langle R^+(\lambda)\, R^-(\lambda) \rangle,$$

where

$$R^{\pm}(\lambda) = \sum_{i=1}^{N} \sigma_{\pm}^{(i)} \psi_\lambda(\mathbf{r}_i).$$

The eigenfunctions ψ_λ and the corresponding eigenvalues were determined by Ressayre and Tallet[17] for a cylindrical volume in the limits of small and large Fresnel numbers $\mathcal{F} + A/L\lambda$. All the largest values of λ_0 are $\varkappa$-fold degenerate and are given by

$$\begin{aligned} &\lambda_0 = \frac{N\pi}{k_0 L}, \quad \varkappa = \frac{1}{\mathcal{F}} \gg 1 \quad \text{for} \quad \mathcal{F} \ll 1 \\ &\lambda_0 = \frac{N\pi}{k_0^2 A}, \quad \varkappa = 2\mathcal{F} \gg 1 \quad \text{for} \quad \mathcal{F} \gg 1. \end{aligned} \tag{3.22}$$

The other eigenvalues satisfy $\lambda \ll \lambda_0$. The eigenfunctions are

$$\psi_\lambda(\mathbf{r}_i) \approx \frac{1}{\sqrt{N}} (\cos \mathbf{k}_0 \mathbf{r}_i + \sin \mathbf{k}_0 \mathbf{r}_i), \tag{3.23}$$

where $\mathbf{k}_0 = \omega_0/c\hat{\mathbf{k}}_0$, and $\hat{\mathbf{k}}_0$ is the unit vector along the axis of the cylinder.

It follows from system (3.7) that the quantity $(2\tau/T_0^2)\lambda$ determines the rate of superradiative decay in the given mode λ, which is determined by the shape of the sample. The number of working modes in (3.17) is thus determined by (3.22); i.e., $\varkappa_0 = \varkappa$.

Physically, the introduction of the functions $\psi_\lambda(\mathbf{r}_i)$ corresponds to a transformation from the two-dimensional modes of the quantization volume to the eigenmodes of the volume of the sample.

2. *Analysis of the solution of the system of equations for multimode superradiative decay.* The solution of system (3.17) depends on the relations among the relaxation times τ, T_2, and $T_{1\,\mathrm{eff}}$ and the collective-decay time τ_c. In the low-density gases in which superradiance is observed, we have $T_2, T_1' \ll T_1^*$ (T_1^* is of the order of the characteristic time for collisions between atoms), so that $1/T_{1\,\mathrm{eff}} \approx 1/T_1'$. If we ignore transverse and longitudinal relaxation and set $T_2 = T_{1\,\mathrm{eff}} = \infty$ in the last two equations in (3.7), we obtain the conservation law

$$\frac{1}{2} \sum_{\mathbf{k}_\varkappa} (R_{\mathbf{k}}^+ R_{\mathbf{k}}^- + R_{\mathbf{k}}^- R_{\mathbf{k}}^+) + \frac{1}{4} R_3^2 = \frac{N}{2} \left(\frac{N}{2} + \varkappa \right), \tag{3.24}$$

which is a generalization to the case of multimode superradiance of the law stating the conservation of the Bloch vector in (3.1). This conservation law holds because of (3.9). Using (3.24), we find the following expression for the integrated radiation intensity in the working modes:

$$I = \frac{n}{\tau} = \frac{N}{4\tau_c} \operatorname{sech}^2 \frac{t - t_0}{2\tau_c}, \tag{3.25}$$

where now we have

$$t_0 = \tau_c \ln \frac{N}{\varkappa}, \tag{3.26}$$

$$\tau_c = \frac{2\tau (N + \varkappa)}{T_0^2}. \tag{3.27}$$

Although $T_2 \leq T_1'$, the relaxation term with the relaxation time T_1' is frequently more important than the transverse relaxation in systems with $\tau_c \ll T_2$. The reason is that the superradiative decay begins from noncoherent spontaneous decay, so that the processes governing T_1' can have a significant effect on the delay of the superradiance pulse. From system (3.17) for the case of pure superradiance ($\tau_c \gg \tau$), we easily obtain the following expression[22]:

$$\frac{d^2 R_3}{dt^2} - \frac{1}{2N\tau_c} \frac{dR_3^2}{dt} + \frac{1}{T_1} \frac{dR_3}{dt} = \frac{1}{T_1' \tau_c} \frac{(N + R_3) R_3}{N}, \tag{3.28}$$

where $1/T_1' = (1/T_1') + (1/T_{1\varkappa})$ is the total probability for the spontaneous decay, and $1/T_{1\varkappa}$ is the probability for spontaneous decay into the working modes.

Taking the relaxation with the time T_1 into account leads to a breakdown of the conservation of the length of the Bloch vector described by (3.24). In this sense, Eq. (3.28) replaces (3.24).

The importance of processes which do not conserve $R_{\mathbf{k}}^2$ was first pointed out by Lee,[21] who derived the following expression for the moment of the transition from a state of isotropic spontaneous decay (with $R_{\mathbf{k}}$ not conserved) to a state of correlated emission, in which the most important processes conserve $R_{\mathbf{k}}^2$:

$$t_1 = \tau_c \ln \frac{1 - (\lambda_0/N)}{\lambda_0/N}.$$

The effect of the nonconservation of the length of the Bloch vector on the superradiance pulses, which has been studied by Emel'yanov and Seminogov,[22] will be discussed in more detail in Section 6.

3. *Superradiance in weakly amplifying media.* We will now see how transverse relaxation affects superradiance. The solution of system (3.17) with the initial conditions (3.12) can be broken up into three stages. In the first stage, correlated states are formed from

states which are completely uncorrelated initially. In this stage, the condition $N - R_3 \ll N$ holds, and system (3.17) becomes linear. The set of eigenvalues of the problem for this stage is given by (3.13), where all the τ_k satisfy $\tau_k \simeq L/c$. System (3.17) also becomes linear in the third stage, where $N - |R_3| \ll N$. In this stage, the correlated states decay. Depending on the parameters of the system, the decay is either exponential or oscillatory. Such parameters of the system as the delay time t_0 and the oscillation frequency can thus be found by analyzing the set of eigenvalues of the linearized system. Using (3.13) for example, we easily find the following expression for the delay of the superradiance pulse:

$$t_0 = 2\tau \ln\left(\frac{N}{\varkappa}\right)\left[\sqrt{\left(1-\frac{\tau}{T_2}\right)^2+4\frac{\tau}{\tau_c}}-\left(1+\frac{\tau}{T_2}\right)\right]^{-1}. \quad (3.29)$$

In the intermediate (second stage, the superradiance pulse is formed. This stage is described by definitely nonlinear equations, but again in this case we can find ways to simplify the analytic solution of the problem. For example, for systems with $\tau \ll \tau_c$ we have $d^2R/dt^2 \ll 1/\tau\, dR/dt$. In this case, $T(t)$ is determined by

$$R(t) = \frac{N\tau_c}{T_2} - N\sqrt{\left(1-\frac{\tau_c}{T_2}\right)^2+\frac{4\varkappa}{N}}\,\mathrm{th}\left[\frac{1}{2}\left(\frac{1}{\tau_c}-\frac{1}{T_2}\right)(t-t_0)\right], \quad (3.30)$$

and the radiation intensity is determined by[23]

$$I = \frac{N}{4\tau_c}\left(1-\frac{\tau_c}{T_2}\right)^2 \mathrm{sech}^2\left[\frac{1}{2}\left(\frac{1}{\tau_c}-\frac{1}{T_2}\right)(t-t_0)\right], \quad (3.31)$$

where

$$t_0 = \left(\frac{1}{\tau_c}-\frac{1}{T_2}\right)^{-1}\ln\left[\frac{N}{\varkappa}\left(1-\frac{\tau_c}{T_2}\right)\right]. \quad (3.32)$$

Equation (3.31) shows that the condition $T_2 \gg t_0$, i.e.,

$$\mu_0 L > \ln N,$$

which is usually stipulated for superradiance, is actually not a necessary condition for the observation of superradiance. From (3.31) we find the following dependence of the maximum radiation intensity on the amplification of the medium:

$$I_{\max} = \frac{N}{4\tau_c}\left(1-\frac{1}{\mu_0 L}\right)^2. \quad (3.33)$$

Then the condition for superradiative decay is

$$\mu_0 L > 1. \quad (3.34)$$

From (3.30)–(3.32) we see that, while all N radiators come into phase in a strongly amplifying medium, only a certain effective number $N_{\text{eff}} = N[1 - (1/\mu_0 L)]$ do so in a weakly amplifying medium, and it is only this number which is involved in the cooperative decay. The population difference immediately after the superradiance pulse is thus

$$R_0 = -N\left(1-\frac{2}{\mu_0 L}\right). \quad (3.35)$$

Let us compare the equations for the delay time in (3.29) and (3.32). The first was found by equating the sum of $\varkappa$ exponential functions with the arguments (3.13) to their maximum value, N. The second was derived through an approximate solution of the nonlinear system in (3.17). A numerical solution of (3.17) shows that in the case $\tau \ll \tau_c$ this approximate solution is essentially exact. The analysis above thus shows that the most general equation for the delay time is Eq. (3.29) with N replaced by $N_{\text{eff}} = N[1 - (1/\mu_0 L)]$ in the logarithm. It is not difficult to see that the coefficient of the logarithm in (3.32) follows from the corresponding coefficient in (3.29) in the case $\tau \ll \tau_c T_2$.

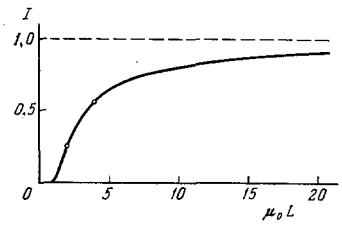

FIG. 3. Maximum radiation intensity as a function of the amplification of the medium. The intensity is normalized to the maximum intensity of strongly amplifying media. The circles are the results of a numerical solution of system (3.17) (see Fig. 4).

Figure 3 shows the dependence of the maximum intensity, normalized to the maximum intensity in strongly amplifying media, $I_0 = N/4\tau_c$, on the amplification of the medium. We see that at $\mu_0 L \approx 3$ the radiation intensity is lower than that calculated from the equations for a strongly amplifying medium by a factor of only two. Clearly, the behavior in Fig. 3 is not correct, since the value of I_0 itself depends on $\mu_0 L$. Figure 4 shows how the shape of the superradiance pulse is affected by the amplification of the medium; these curves were found through a numerical solution (3.17) for systems with $\tau/\tau_c = 2 \cdot 10^{-3}$.

4. *Oscillatory superradiance.* The successful experiments by Skribanowitz *et al.*,[24] which revealed the basic features of the effect—the highly directional radiation and the quadratic dependence on the intensity on the density of radiating particles—generated a wave of interest in the problem. An interesting feature of the decay intensity was its oscillatory structure. Some typical oscilloscope traces with superradiance pulses from Ref. 24 are reproduced in Fig. 5. Many papers have been devoted to a derivation of kinetic equations describing the oscillatory superradiance and to determining the conditions under which the oscillation occurs.[18–20,24,26–32] The equations of oscillatory superradiance were analyzed numerically in Refs. 29–32. An analytic expression has been derived for the oscillation frequency, and the threshold for the oscillatory case has been determined.[18–20]

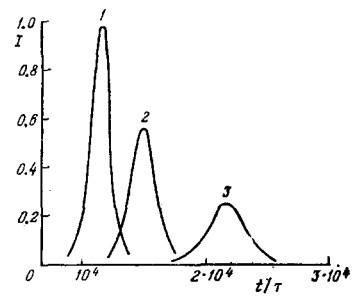

FIG. 4. Dependence of the shape of the superradiance pulse on the amplification of the medium.[31] $\tau/\tau_c = 2.10^{-3}$. 1) $\mu_0 L = 200$; 2) 4; 3) 2.

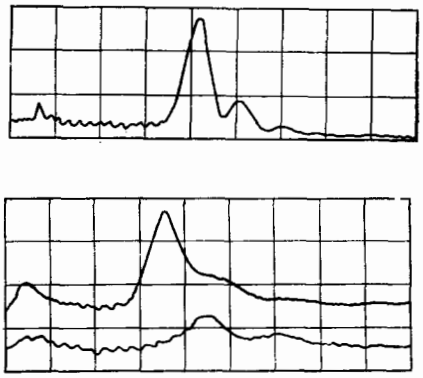

FIG. 5. Oscilloscope traces of superradiance pulses (from Ref. 24; gaseous HF, $\lambda = 84\ \mu$m).

It is simple to see the reason for the oscillation: In the case $\tau_c < \tau$, cooperative emission occurs only within regions with dimensions of the order of[16] $l_c = c\tau_c$. As the radiation propagates from the central regions of the sample through adjacent "coherence regions," energy may be transferred from the field back to the medium. In other words, the field may induce transitions of atoms, which have decayed, back to the excited state (in complete analogy with the events which occur during propagation of radiation in a resonantly absorbing medium).[33] The subsequent decay of these twice-excited atoms leads to the additional peaks.

The set of eigenvalues of the problem in the third stage is[18,20]

$$\lambda_1 = -\frac{1}{2}\left(\frac{1}{\tau}+\frac{1}{T_2}\right), \quad \lambda_{2,3} = \lambda_1 \pm \sqrt{\frac{1}{4}\left(\frac{1}{\tau}-\frac{1}{T_2}\right)^2 + \frac{2R_0}{T_0^2}}. \tag{3.36}$$

We see from (3.36) that in the case $R_0 < 0$ there may be an oscillation in this stage. The condition for the occurrence of the oscillation is

$$\mu_0 L > \frac{1}{4}\left(6 + \frac{\tau}{T_2} + \frac{T_2}{\tau}\right). \tag{3.37}$$

and the oscillation frequency is

$$\Omega = \sqrt{\frac{2|R_0|}{T_0^2} - \frac{1}{4}\left(\frac{1}{\tau}-\frac{1}{T_2}\right)^2}. \tag{3.38}$$

5. *Superradiance of inhomogeneously broadened systems.* We assumed above that the atoms were subject only to homogeneous broadening, and we correspondingly assumed that the photon energy $\hbar\omega_{\mathbf{k}}$ was equal to the atomic excitation energy, $\hbar\omega_{\mathbf{k}} = \hbar\omega_0$. Consequently, taking a multimode situation in the transverse direction into account, i.e., in considering the dependence of $n_{\mathbf{k}}$ on the direction of $\mathbf{k}$, we are not taking into account a multimode situation in the longitudinal direction; i.e., we are not considering a dependence of $n_{\mathbf{k}}$ on the magnitude of the wave vector, $|\mathbf{k}| = \omega_{\mathbf{k}}/c$. We introduce the average frequency of the atomic transition, $\bar{\omega}_0 = \sum_{i=1}^{N}\omega_{0i}/N$; the photons which arise in the system now have a frequency $\omega_{\mathbf{k}} = \bar{\omega}_0 + \Delta\omega_{\mathbf{k}}$. From system (3.7), instead of (3.17), we now have the system of equations

$$\left.\begin{aligned}
&\frac{dn}{dt} + \frac{n}{\tau} = F,\\
&\frac{d^2F}{dt^2} + \left(\frac{1}{\tau}+\frac{1}{T_2}\right)\frac{dF}{dt} + \left[\frac{1}{4}\left(\frac{1}{\tau}+\frac{1}{T_2}\right)^2 + \overline{\Delta\omega^2}\right]F\\
&\quad = \frac{1}{T_0^2}\left[\frac{d}{dt}(nR + \varkappa S_0 + S) + \left(\frac{1}{\tau}+\frac{1}{T_2}\right)(nR + \varkappa S_0 + S)\right],\\
&\frac{dS}{dt} + \frac{S}{T_2} = FR_3, \qquad \frac{dR_3}{dt} + \frac{N+R_3}{T_{1\,\mathrm{eff}}} = -2F,
\end{aligned}\right\} \tag{3.39}$$

where

$$\overline{\Delta\omega^2} = \frac{\sum_{\mathbf{k}} \Delta\omega_{\mathbf{k}}^2 F_{\mathbf{k}}}{\sum_{\mathbf{k}} F_{\mathbf{k}}}.$$

In the initial stage, system (3.39) has the eigenvalues

$$\left.\begin{aligned}
\lambda_{1,2} &= -\frac{1}{2}\left(\frac{1}{\tau}+\frac{1}{T_2}\right) \pm \sqrt{\nu_1+\nu_2},\\
\lambda_{3,4} &= -\frac{1}{2}\left(\frac{1}{\tau}+\frac{1}{T_2}\right) \pm i\sqrt{\nu_2-\nu_1},
\end{aligned}\right\} \tag{3.40}$$

where

$$\nu_1 = \frac{1}{2}\left[\frac{1}{4}\left(\frac{1}{\tau}-\frac{1}{T_2}\right)^2 + \frac{2N}{T_0^2} - \overline{\Delta\omega^2}\right],$$

$$\nu_2 = \sqrt{\nu_1^2 + \frac{\overline{\Delta\omega^2}}{4}\left(\frac{1}{\tau}-\frac{1}{T_2}\right)^2}.$$

A growing solution thus exists if

$$\mu_0 L > 1 + 4\tau^2\overline{\Delta\omega^2}\left(1+\frac{\tau}{T_2}\right)^{-2}. \tag{3.41}$$

For systems with $\tau \ll T_2 \ll T_{1\,\mathrm{eff}}$, we easily find the following expression from (3.39):

$$S(t) = \frac{N^2}{4}\left(1-\frac{1}{\mu L}\right)^2 - \frac{1}{4}\left(R(t) - \frac{N}{\mu L}\right)^2, \tag{3.42}$$

where

$$\mu = \mu_0\left[1 + 4\tau^2\overline{\Delta\omega^2}\left(1+\frac{\tau}{T_2}\right)^{-2}\right]^{-1}.$$

Using (3.42), we can show in a straightforward manner that all the equations of the two preceding sections remain the same, except that μ_0 is replaced by μ. For example, the maximum radiation intensity is now

$$I = \frac{N}{4\tau_c}\left(1-\frac{1}{\mu L}\right)^2. \tag{3.43}$$

An important characteristic of inhomogeneously broadened systems is that the radiation intensity is always an oscillatory function, since the two eigenvalues in (3.40) are always complex. The reason for the oscillation, however, is twofold. One of the mechanisms leading to the oscillation was explained in the preceding section. The second reason lies in the longitudinal multimode nature of the radiation. While the first mechanism leads to an oscillation after the radiation intensity reaches its maximum, the second leads to an oscillation which occurs immediately after the end of the pump pulse. This oscillatory structure is in fact visible in Fig. 5.

Changes similar to those described above also occur in the third stage of the evolution of the superradiance pulse.

Let us summarize the basic changes caused in the dynamics of superradiance decay by the processes responsible for inhomogeneous broadening. In systems with $\overline{\Delta\omega^2} \ll 1/\tau^2$, the main features of the effect remain the same; there is simply an insignificant change in the parameters of the pulses. The only distinctive feature of the decay of such systems is the appearance of an additional small-scale oscillation, whose frequency in this case is higher than Ω, as shown in Ref. 20. Inhomogeneous-broadening processes are important in systems with $\overline{\Delta\omega^2} \sim 1/\tau^2$. In this case, even if the conditions for oscillatory superradiance, (3.7), are not satisfied in the given system, the emission pulse will break up into a train of pulses of comparable height

(see Figs. 9 and 11 below). In this case there are significant changes in the parameters of the superradiance pulses, and the threshold condition for observation of the effect becomes $\mu L > 1$.

6. *Competition between collective and spontaneous processes.* We turn now to the decay of slightly anisotropic systems. In studying the decay of extended systems, we summed over the working and nonworking modes separately in (3.7). For nearly isotropic systems, all modes are equivalent, and Eq. (3.7) must now be summed over all modes. The quantity $\varkappa$ in (3.17) is now determined by the total number of modes, and $T_{1\,\mathrm{eff}}$ is equal to T_1^*. It is a simple matter to calculate $\varkappa$:

$$\varkappa = 8\pi \frac{V\omega^2}{(2\pi c)^3}\Delta\omega = \frac{V\omega^2}{2\pi c^3 \tau}. \tag{3.44}$$

On the other hand, the probability for spontaneous decay is

$$\frac{1}{T_1} = \frac{2\pi}{\hbar}|g|^2\rho(\hbar\omega) = \frac{2\tau\varkappa}{T_2^2}, \tag{3.45}$$

where $\rho(\hbar\omega)$ is the density of final states. Accordingly,

$$\frac{\varkappa}{\tau_c} = \frac{N}{T_1}.$$

In a completely isotropic system, with $\varkappa = N$, there is absolutely no correlated emission. If $\varkappa \lesssim N$, on the other hand, a slight correlation may be observed. The following equation can be derived[20] in a straightforward fashion from (3.17) with the help of (3.44) and (3.45):

$$R(t) = N\alpha - N\beta \operatorname{th}\frac{\beta(t-t_0)}{2\tau_c}, \tag{3.46}$$

where

$$\alpha = \frac{\tau_c}{T_2} + \frac{\tau_c}{T_1}, \quad \beta = \sqrt{\left[1-\left(\frac{\tau_c}{T_2}-\frac{\tau_c}{T_1}\right)\right]^2 + \frac{4\tau_c^2}{T_2 T_1}}, \quad t_0 = \frac{\tau_c}{\beta}\ln\frac{\beta+(1-\alpha)}{\beta-(1-\alpha)}. \tag{3.47}$$

The maximum radiation intensity is

$$I = \frac{N}{2T_1}\left(1+\frac{\tau_c}{T_2}+\frac{\tau_c}{T_1}\right) + \frac{N}{4\tau_c}\left[\left(1-\frac{\tau_c}{T_2}\right)^2 - \left(\frac{\tau_c}{T_1}\right)^2\right]. \tag{3.48}$$

where the first term describes the intensity of the noncoherent spontaneous decay, while the second describes the intensity of the coherent decay. The coherent component of the radiation is thus lost when $T_1 = [(1/\tau_c) - (1/T_2)]^{-1}$. In this case, the radiation intensity is described by $I = N/T_1$; i.e., there is a purely spontaneous decay in such systems. We note that the time t_0 given by expression (3.47) vanishes in this case.

We note that the decay of systems with $\tau_c \sim T_2 \sim T_1$ can no longer be referred to as superradiance, since the intensities of the coherent and noncoherent components of the radiation in such systems become comparable, according to (3.48), so that I ceases to vary in proportion to n^2.

d) Semiclassical theory of superraidance. Spatial variation of the field envelope

The semiclassical system of equations for the slowly varying field amplitude $\mathbf{E}(\mathbf{r},t) = \mathbf{A}(x,t)e^{i(\omega t - kx)} + \text{c.c.}$, the polarization $\mathbf{P}$, and the density of the population difference $\langle R_3\rangle$ is widely used in research on laser kinetics. If Doppler broadening is ignored, this system can be written in the form

$$\left.\begin{aligned} &\frac{\partial \mathbf{A}}{\partial t} + c\frac{\partial \mathbf{A}}{\partial x} = i2\pi k\mathbf{P}, \\ &\left(\frac{\partial}{\partial t} + \frac{1}{T_2}\right)\mathbf{P} = \frac{d^2\langle R_3\rangle}{\hbar}\mathbf{A} + \xi_n, \\ &\frac{\partial}{\partial t}\langle R_3\rangle + \frac{1}{T_1}\langle R_3\rangle = -\frac{1}{\hbar}\operatorname{Re}(\mathbf{A}^*\mathbf{P}), \end{aligned}\right\} \tag{3.49}$$

where $P \sim R^*$ and ξ_n is the fluctuation source. Since we are using the plane-wave approximation in (3.49), this system of equations is applicable only for large Fresnel numbers, $\mathcal{F} \gg 1$.

Equations (3.49) were applied to the superradiance problem in Refs. 24, 25, and 34. For these equations, the fluctuation source ξ_n is important in the initial stage of the superradiance. Skribanowitz *et al.*[24] determined ξ_n on the basis of other considerations, as a source of polarization noise. Polder *et al.*[34] derived semiclassical equations by a quantum-mechanical approach, finding an expression for the source in the equation for $\mathbf{P}$ resulting from zero-point field fluctuations. The expression differs from that used by Skribanowitz *et al.*[24]

The approaches discussed above have made use of a splitting of ternary correlations of the type

$$\langle a_k^+ a_k R_3\rangle = \langle a_k^+ a_k\rangle\langle R_3\rangle, \quad \langle a_k^+ R_k^- R_3\rangle = \langle a_k^+ R_k^-\rangle\langle R_3\rangle \tag{3.50}$$

or

$$\langle \sigma_+^{(i)}\sigma_-^{(j)}\sigma_3^{(l)}\rangle = \langle \sigma_+^{(i)}\sigma_-^{(j)}\rangle\langle \sigma_3^{(l)}\rangle, \quad \langle \sigma_3^{(i)}\sigma_3^{(j)}\rangle = \langle \sigma_3^{(i)}\rangle\langle \sigma_3^{(j)}\rangle, \tag{3.51}$$

while the semiclassical equations instead use a splitting of binary correlations,

$$\langle a^+R^-\rangle = \langle a^+\rangle\langle R^-\rangle, \quad \langle a^+a\rangle = \langle a^+\rangle\langle a\rangle, \tag{3.52}$$

and this difference leads to a slightly less accurate description of the initial stage of the avalanche. Trifonov *et al.*[35] have reported a comparative analysis of the semiclassical and quantum approaches. One of the most important achievements of the semiclassical approach has been the incorporation of the change in the field envelope along the length of the sample. This effect leads to results different from those found by other approaches. For example, instead of Eq. (3.6), the semiclassical method leads to[24]

$$\frac{\partial^2\theta}{\partial x\,\partial t} + \frac{1}{L\tau_c}\sin\theta = 0, \tag{3.53}$$

where

$$R_3(x,\ t) = -R_0\cos\theta, \quad \langle R^+\rangle = R_0\sin\theta.$$

An asymptotic solution of (3.53) which satisfies the boundary condition $\theta(x=0,t)=\theta_0$ is[36]

$$\theta = \pi - \frac{C}{\sqrt{\eta}}\cos\left(\eta - \frac{\pi}{4} + \frac{C^2}{16}\ln\eta + \varphi_0\right) + O(\eta^{-3/2}),$$

where $\eta = 2\sqrt{xt/L\tau_c}$, and C and φ_0 are constants.

Equation (3.53) yields the following value for the delay of the superradiance pulse:

$$t_0 = \frac{\tau_0}{2}\ln\frac{\pi}{2\theta_0}, \tag{3.54}$$

where $\tau_0 = (\tau_c/2)\ln(\pi/2\theta_0)$ is the time over which the intensity falls by a factor of e from its maximum. The value of θ_0 depends on which source of fluctuations is used in the calculations.[24,34]

Vrehen and Schuurmans[37] have recently reported a direct experimental determination of θ_0. In those experiments, a cell holding cesium vapor was irradiated with a superradiance pulse from a second, identical cell immediately after the pump pulse. The intensity of the superradiance pulse was adjusted with an attenuator. If the area of the incident pulse, θ_p is greater than θ_0, such a pulse causes a decrease in the delay of the decay pulse of the test cell. If, on the other hand, $\theta_p < \theta_0$, this pulse should not affect the delay of the pulse from the second cell. The results found in those experiments agree quite well with the theory of Polder *et al.*[34]

e) Role played by fluctuations in superradiative decay (Bloch representation)

As mentioned in the preceding section, closed kinetic equations for superradiance can be derived only by using approximations like those in (3.50) and (3.51), i.e., only by ignoring the fluctuations in the number of photons. When these fluctuations are taken into account, they lead to a system of coupled equations which are too complicated to analyze analytically or even numerically.[27] Gronchi and Lugiato[40] have shown that the solution of the problem of limiting the system of coupled equations can be physically interpreted with the greatest clarity in the so-called Bloch representation, which was used to describe superradiance in Refs. 38–40. The Bloch or coherent atomic representation was also introduced independently in Refs. 41 and 42. This representation is introduced on the basis of the conservation of the vector R_2 with the components R_x, R_y, R_z, the square of whose length is given by (2.6). While the state of the atomic system in the Dicke representation is determined by specifying the cooperation number r and the population half-difference m, the state of the system in the Bloch representation is determined by specifying the azimuthal angle θ and the polar angle φ of the vector R in pseudospin space. The transformation from the wave functions in the Dicke representation to the wave functions in the coherent atomic representation is made by[38]

$$|\theta, \varphi\rangle = \sum_{m=-r}^{r} |r, m\rangle\langle r, m|\theta, \varphi\rangle = \sum_{m=-r}^{r} |r, m\rangle \binom{2r}{m+r}^{1/2} \left(\sin\frac{\theta}{2}\right)^{r+m}\left(\cos\frac{\theta}{2}\right)^{r-m} e^{-i(r+m)\varphi}. \tag{3.55}$$

Arecchi *et al.*[42] have demonstrated the analogy between the coherent atomic representation and the coherent representation of the electromagnetic field introduced by Glauber.[43] In particular, the state of a system of atoms with specified values of θ and φ has a minimum uncertainty; i.e., the product of the mean square deviations of canonically conjugate quantities is at a minimum in these states. This formal analogy has served as a basis for a further use of this representation. For example, by expressing the eigenvalues of the operator representing the density of the atomic subsystem in terms of the vectors of the coherent states in (3.55), we can calculate the normally ordered products of the operators R^+ and R^- by means of the following correspondence rules:

$$(R^+)^l (R^-)^l \to (r\sin\theta)^{2l}, \quad R_3^n \to (-r\cos\theta)^n.$$

We denote by $\rho_A(t)$ the density matrix of the atomic subsystem. In the diagonal representation,[42] we have

$$\rho_A(t) = \int d\Omega P(\theta, \varphi, t)\,|\theta, \varphi\rangle\langle\theta, \varphi|, \tag{3.56}$$

where $d\Omega = \sin\theta\, d\theta\, d\varphi$, and the weight function $P(\theta, \varphi, t)$ has a meaning analogous to a probability density for the distribution of the values of θ and φ over the (θ, φ) Bloch sphere. Narducci *et al.*[38] derived an equation for the function

$$Q(\theta, t) = \sin\theta \int_0^{2\pi} d\varphi P(\theta, \varphi, t).$$

The result is

$$\frac{\partial}{\partial t} Q(\theta, t) = \frac{2g^2\tau}{\hbar^2}\left\{\frac{\partial}{\partial\theta}\left[\left(r\sin\theta + \frac{\sin\theta}{2(1+\cos\theta)}\right) Q(\theta, t)\right] + \frac{\partial^2}{\partial\theta^2}\left[\frac{1-\cos\theta}{2} Q(\theta, t)\right]\right\}. \tag{3.57}$$

The Fokker–Planck equation (3.57) describes the behavior of the function $Q(\theta, t)$ on the Bloch sphere. The term with the first derivative on the right side of (3.57) describes the motion of the maximum of the function $Q(\theta, t)$; the diffusion term with the second derivative describes the distribution of the function $Q(\theta, t)$ on the Bloch sphere. Far from the completely inverted state, $\theta = \pi$, the first term in brackets with the first derivative is dominant. This term gives us the ordinary classical description of superradiance. If we retain only this term in (3.57), we find the following equation for the evolution of the angle θ:

$$\frac{d\theta}{dt} = -\frac{2g^2\tau}{\hbar^2} r\sin\theta, \tag{3.58}$$

which is the same as Eq. (3.56). The second term in the expression with the first derivative spontaneous sources into account. Analysis of Eq. (3.57) has shown[38] that the average product $\langle R^{+l} R_3^n R^{-l}\rangle$ can be factorized in the form of the product $\langle R^+R^-\rangle^l\langle R_3\rangle^n$ everywhere except in a region near the completely inverted state, with $\theta = \pi$. Glauber and Haake[39] have shown that the classical description becomes progressively poorer as the initial state approaches the state with $\theta = \pi$, and in the limit in which the initial state is a state of complete inversion the maximum intensity and the delay calculated from (3.57) may differ from the classical results by 20%. Gronchi and Lugiato[40] showed that there is yet another factor, neglected in (3.57), which can be important: A term $2g^2\tau^2\gamma^2\sin\theta/\hbar$ must be taken into account in the term with the second derivative on the right side of (3.57). In other words, we must make the replacement

$$\frac{\partial^2}{\partial\theta^2}\left[\frac{1-\cos\theta}{2} Q(\theta, t)\right] \to \frac{\partial^2}{\partial\theta^2}\left[\frac{1-\cos\theta}{2} + \frac{2g^2\tau^2\gamma^2}{\hbar}\sin^2\theta\right]$$

in (3.57). This new term describes fluctuations of a collective nature, while the term $(1-\cos\theta)/2$ results from the spontaneous decay of individual atoms. In fact, the quantity $N(1-\cos\theta)/2$ is none other than the number of atoms in the excited state. Another difference between these two terms is that while the term proportional to $1-\cos\theta$ is important only near $\theta = \pi$ the second term is important throughout the formation of the superradiance pulse.

4. SUPERRADIANCE AT SHORT WAVELENGTHS

a) Superradiance in the γ range

The difficulties in developing resonators for electromagnetic waves in the x-ray and γ ranges make the single-pass γ-ray laser, in which the directionality of the radiation is achieved through the use of needle-shaped active crystals, the most promising possibility. As a result, the very first papers in which γ-ray lasers were proposed discussed the possibility of producing superradiative states of a system of excited nuclei. Terhune and Baldwin[44] and Zaretskiĭ and Lomonosov[45] have suggested that this could be arranged by choosing a lattice satisfying the condition

$$\mathbf{k} = 2\pi\mathbf{b}, \tag{4.1}$$

where k is the wave vector of the γ ray, and b is the reciprocal lattice vector. It is not true, of course, that just any arbitrary system of excited nuclei satisfying condition (4.1) would be put in a superradiative state as a result of the decay. Further developments in the theory for superradiance yielded the conditions for superradiative decay. It should be noted that the periodic nature of the crystal lattice should nevertheless play a key role in a possible observation of superradiative decay of nuclei. The reason is that it is the anomalous transmission of γ rays along Bragg directions which leads to the mode discrimination [see (3.14)] required for the appearance of a macroscopic polarization.[46] Consequently, condition (4.1), which corresponds to the particular case of Bragg diffraction, is not a relation which must be imposed between the radiation wavelength and the period of the crystal lattice if we are to observe the effect.

The possibility of achieving superradiance in the γ range has recently been discussed in Refs. 19, 23, and 46–49, where this question was examined in the light of recent developments in the theory of superradiance. One of the most important conclusions reached in Refs. 19, 47, and 48 was that a system of excited nuclei would be put in a superradiative state if the condition $\mu L > \ln N$ were met. The attainment of a high amplification coefficient at short wavelengths is extremely problematical, however, so that the possibility of arranging superradiance in weakly amplifying media, with $1 < \mu L < \ln N$, which was demonstrated by Andreev,[23] has opened up more realistic opportunities. Andreev *et al.*[49] have shown that relatively narrow lines for Mössbauer transitions, combined with the large cross sections for photoabsorption of γ rays in the Mössbauer energy range, has the consequence that collective spontaneous emission should play a leading role in the generation of coherent Mössbauer γ radiation.

To illustrate the consequences of the conditions for the superradiative decay of a system of excited nuclei, we will estimate the critical number density of nuclei required. The characteristic lifetimes of low-lying Mössbauer levels range from 10^{-9} s to several hours. For isomers with a lifetime $T_1 < 10^{-5}$ s the line width is equal to the natural width, while for isomers with $T_1 > 10^{-5}$ s we have $\Gamma = 1/T_2 \sim 10^5$ Hz. Strong photoabsorption of γ rays in the Mössbauer energy range has the consequence that the range of a γ ray in a solid is $l_{abs} \lesssim 0.1$ cm. Since the coherence length is smaller than or equal to the range, we would not want to use a crystal with dimensions larger than l_{abs}. The Borman effect may have the consequence that l_{abs} increases to $l_{abs} \sim 1$ cm. The transit time $\tau \sim l_{abs}/c$ in the Mössbauer region is thus always shorter than T_2. For the isotope Ag_{47}^{107}, for example, we have $T_1 = 44.3$ s, $T_2 = 10^{-5}$ s, $l_{abs} = 0.05$ cm, $\hbar\omega = 93.1$ keV, and $(N/V)_{cr} = 10^{20}\ \Gamma T_1$ cm^{-3}. Accordingly, when a γ line without narrowing is used, the critical number density of nuclei reaches a value beyond the densities of solids.

The possibilities for achieving superradiance in the γ range are more restricted than in the optical range, since now the distance between radiators is comparable to the wavelength. While it is possible in the optical range to arrange mode discrimination (i.e., to supress the secondary maxima which result from the spatial distribution of the radiators), by using needle-shaped samples, it is essentially impossible to satisfy the condition of a unit value for the Fresnel number in the γ range. This condition on the Fresnel number determines the optimum shape of the sample. Accordingly, superradiance can be observed in the γ range only if the nuclei are in a regular arrangement, i.e., in a crystal lattice, since in this case there are directions along which interference maxima are observed: Bragg directions. The interference maxima do not themselves, however, lead to an anisotropy of the threshold conditions; only the Borman effect,[46,49] i.e., the sharp increase in the range of γ rays along the Bragg directions, will lead to the anisotropy of the directional pattern of the radiation which is required for the occurrence of superradiance.

b) Superradiance in the x-ray range

When we examine the possibility of generating coherent x radiation, we run into the same problem as in the γ range: the absence of reflecting mirrors. Another problem, this time unique to the x-ray range, is the absence of long-lived isomer levels. This latter circumstance imposes some very stringent requirements on the pump intensity and forces a search for new types of pump sources: pumping by a travelling wave, transverse pumping by an electron beam, pumping by a scanning ion beam, etc.[50] In these cases, the atoms are in an excited state at precisely that time at which they are reached by the radiation emitted by previously excited atoms. Clearly, the properties of such systems will be quite different from those of ordinary laser systems. A second interesting feature of the ion-beam method is that the Doppler line broadening is eliminated in this case, so that the transverse relaxation time is determined by the decay time for the population difference. It can thus be hoped that effects related to the phase memory of the system will influence the kinetics of the decay. This possibility has been analyzed by Hopf *et al.*[51]

Miller[81] has recently analyzed the possibility of obtaining coherent x radiation by making use of the Doppler shift and the $2P - 1S$ transition of one-electron

atoms accelerated in a high-energy accelerator. Miller showed that superradiative processes may lead to coherent emission in this case also.

MacGillivray and Feld[52] used the theory of Refs. 24 and 25 to analyze the possibility of superradiance in the x-ray range. Here again, as in the preceding section, we might note that the more stringent conditions $\mu L > \ln N$, which McGillivray and Feld identified as necessary for observation of the effect,[52] are relaxed by the possibility of arranging superradiance in a weakly amplifying medium, which was demonstrated by Andreev.[23]

The possibility of arranging conditions for superradiative decay in the x-ray range can be demonstrated by the example of the $L\alpha$ line of sodium at $\lambda = 372$ Å. The mechanism for producing a population inversion for this transition was discussed by Duguay and Rentzepis.[53] Here $P = 0.02$ torr, $T_1 = 4 \cdot 10^{-10}$ s, $T_2^* \simeq 0.17 \cdot 10^{-10}$ s, and $l_{abs} = 9.3 \cdot 10^4$ cm, so that in samples with a length less than $L < T^* c = 0.5$ cm one of the necessary conditions for superradiative decay is satisfied.

The conditions $\mu L > 1$ and $\tau_{pump} < \tau_e$, where τ_{pump} is the length of the pump pulse, determine the pump power required. Under the conditions of Ref. 53, with 0.3% of a pump with $\hbar\omega = 50$ eV absorbed in the medium, the power of the pump pulse must be $I = 4$ GW/cm^3.

5. EXPERIMENTAL WORK ON SUPERRADIANCE

Superradiance has been observed successfully in the IR range in several experiments. The characteristics of the superradiance pulses observed in these experiments are listed in Table I.

a) Superradiance of rotational transitions of molecules

The first experiment carried out to observe superradiance in the IR region was that reported by Skribanowitz *et al.*,[24] in which they observed the effect in HF vapor. The key parts of their experimental apparatus were (1) a HF laser which emitted pump pulses with a wavelength $\lambda = 2.5$ μm and a length of 50–100 ns and (2) a cell holding HF vapor at room temperature, with dimensions from 30 to 100 cm and an inside diameter between 12 and 28 mm. The HF vapor pressure was of the order of 1–2 mtorr. Since the first vibrational excited level of the HF molecule lies 20 $k_B T$ above the ground level at room temperature, all the rotational sublevels of this vibrational level are essentially unpopulated. Consequently, an optical pump pulse which

TABLE I. Summary of the characteristics of superradiative media and of the temporal characteristics of the superradiance pulses.

Gas	λ, μm	L, cm	τ, ns	T_2, ns	$\frac{N}{V}$, cm^{-3}	t_0 ns	Ref.
Tl	1.3	15	0.5	1	$2 \cdot 10^{15}$	12	59
Na	2.21	14	0.47	1.1	$6 \cdot 10^9$—$2 \cdot 10^{10}$	2—5	60
Ca	2.9	5	0.17	5	$2 \cdot 10^{10}$	15	35
Na	3.4	14	0.47	1.7	$6 \cdot 10^9$—$2 \cdot 10^{10}$	2—7	60
HF	84	100	3.3	220	10^{12}	400	24
CH_3F	496	600	20	($T_1 = 60$)	$3 \cdot 10^{12}$	100	54

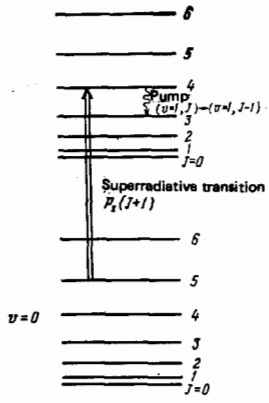

FIG. 6. Energy-level diagram of the HF molecule.

sends the molecule from its ground state ($v = 0$) to one of the rotational sublevels of the first vibrational state ($v = 1$) produces an essentially complete population inversion between two coupled rotational sublevels in the $v = 1$ state. Figure 6 shows the energy diagram of the HF levels.

Similarly, superradiance pulses could be produced by making use of other rotational transitions of the $v = 1$ level. The corresponding transition wavelengths at which superradiance was observed in Ref. 24 are summarized in Table II.

Below 5 mtorr, the superradiance pulses are delayed 500–2000 ns with respect to the pump pulse. As the pressure is reduced, the delay and length of the superradiance pulse increase, while the intensity falls off. At these pressures, the radiation has an oscillatory structure; i.e., there is a train of pulses of decreasing height (Fig. 5a). At pressures above 10 mtorr, a single superradiance pulse is observed (Fig. 5b).

If we ignore the relaxation of the molecules with two rotational levels between which the superradiance transitions occur, we can adopt a three-level model for the superradiance in this case. A similar arrangement for producing superradiance was used by Rosenberger *et al.*[54] They observed superradiance in purely rotational transitions of the CH_3F molecule. For the optical pumping they used a pulse from a CO_2TEA laser with $\lambda = 9.55$ μm, exciting the $\nu_3^9 Q(12.2)$-mode of CH_3F. The length of the cell holding the CH_3F was varied from 2.3 m to 9.7 m, and the gas pressure was varied from 0.08 to 0.8 torr. When the cell was less than 6.3 m long, the superradiance pulses emitted toward the two ends of the

TABLE II. Summary of the rotational transitions of HF at which superradiance was observed in Ref. 24.

Pump				Superradiative transition	
$R_1(J)$	λ, μm	$P_1(J)$	λ, μm	$J_U \rightarrow J_L$	λ, μm
$R_1(0)$	2.50	$P_1(2)$	2.58	1—0	252.7
$R_1(1)$	2.48	$P_1(3)$	2.61	2—1	126.4
$R_1(2)$	2.45	$P_1(4)$	2.64	3—2	84.4
$R_1(3)$	2.43	$P_1(5)$	2.67	4—3	63.4
$R_1(4)$	2.41	$P_1(6)$	2.71	5—4	50.8

$R_1(J)$ denotes the transition $(v=1, J+1) \rightarrow (v=0, J)$,
$P_1(J)$ denotes the transition $(v=1, J-1) \rightarrow (v=0, J)$.

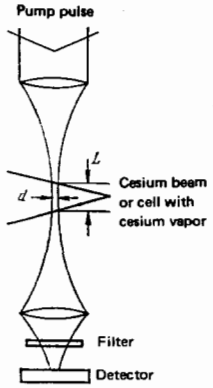

FIG. 7. Experimental arrangement of Ref. 56.

cell had the same shape. In this case, therefore, the pumping was homogeneous. As the length of the cell was increased, the shapes of the pulses leaving the two ends of the cell became different. This change corresponded to a transition to a travelling pump. The dependence of the properties of the superradiance pulses on the pressure and length of the sample, for short lengths, were the same as in the experiments of Ref. 24.

b) Superradiance of optical transitions of atoms

Level degeneracy can strongly affect superradiance, as has been demonstrated particularly clearly in the experiments of Refs. 55 and 56, where the effect was observed during electronic transitions in a vapor of cesium atoms. The experimental arrangement used in Refs. 55 and 56 is shown in Fig. 7. In some of the experiments, the cesium vapor was held in a cell from 1 to 10 cm long at 30–100° C; in other experiments, it was injected through a slit with dimensions of 0.5 × 3 mm from an oven containing several grams of cesium at 200–300° C. Figure 8 shows the level diagram of the cesium atom. In Ref. 55, the effect was observed in two situations. In a first experiment, a pulse from a N_2 laser with a spectral width of 400–500 mHz excited, from one of the sublevels of the hyperfine structure of

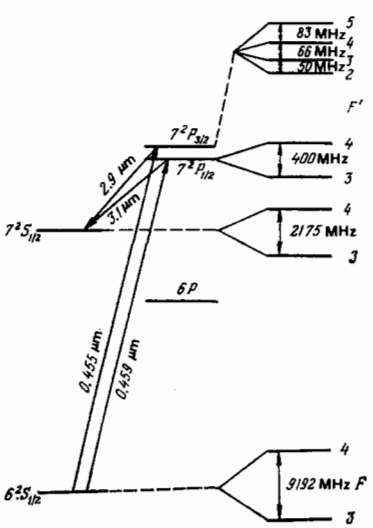

FIG. 8. Energy-level diagram of the cesium atom.

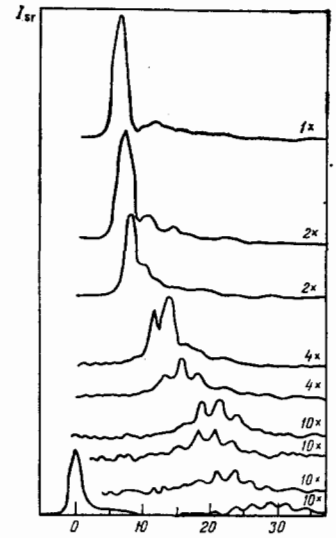

FIG. 9. Quantum beats in the superradiance intensity caused by hyperfine structure of the $7P_{1/2}$ level of the cesium atom.[55] The abscissa scale shows the time in nanoseconds.

the $6S_{1/2}$ state, a hyperfine sublevel of the $7P$ state, which decayed through superradiative decay to one of the hyperfine sublevels of the $7S$ state. Since the detector did not resolve the beats which resulted from the hyperfine structure of the $7S$ state (of the order of 2 GHz), the observed beats were due entirely to the hyperfine structure of the $7P$ level. Figure 9 shows oscilloscope traces of the superradiance pulses for the case of the excitation of the $7P_{1/2}$ level. The beats are at 400 MHz, which corresponds to the hyperfine structure of the $7P_{1/2}$ level. In the second experiment, a transverse magnetic field of 2.8 kOe was applied to the sample, and excitation was brought about by a pulse with linear σ polarization.

Figure 10 shows the Cs level diagram in a magnetic field. The $M = -5/2$ sublevel of the $7P$ state could be excited independently of the $M = -3/2$ sublevel, and in this case a superradiance pulse was observed without beats. If, on the other hand, the intensity of the exciting pulse was such that the nutation frequency exceeded the splitting of the ground level, 1.3-GHz beats appeared (Fig. 11). It should be noted that these beats cannot be observed in spontaneous decay, since the sublevels of the $7P$ state were excited from different sublevels of the ground level and decayed to different sublevels of the final state.

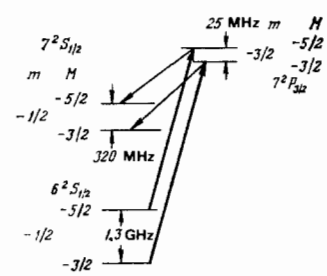

FIG. 10. Energy-level diagram of cesium in a magnetic field of 1.8 kOe.

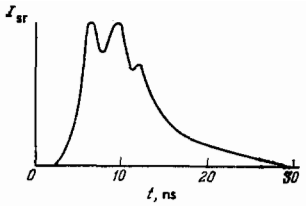

FIG. 11. Quantum beats in the superradiance intensity in a magnetic field of 2.8 kOe (Ref. 55).

Gibbs *et al.*[56] observed single superradiance pulses. Their observation conditions corresponded to the second experiment of Ref. 55, but they varied the vapor density and the characteristics of the cells over broad ranges. The results of their experiments are shown in Fig. 12.

The experiments by Gross *et al.*[57] were an extremely interesting continuation of the experimental research on superradiance with cesium. Here beats were observed from the interaction of two groups of atoms with different veloicites within the Doppler width (500 MHz). In contrast with Refs. 55 and 56, the sublevels of the $7P_{1/2}$ state were excited in Ref. 57 by a pulse from a N_2 laser with a bandwidth of 100 MHz, i.e., less than the distance between the hyperfine levels of the $7P_{1/2}$ state. Consequently, the atoms excited to the $7P_{1/2}$ state were those whose velocities v_f ($v_{f'}$) satisfied

$$\nu_{gF}=\nu_L-k_Lv_F,\quad \nu_{gF'}=\nu_L-k_Lv_{F'},$$

where ν_L and $\mathbf{k}_L$ are the average frequency and average wave vector of the exciting pulse, and $\nu_{gF}(\nu_{gF'})$ is the difference between the frequencies of the upper (lower) sublevel of the $7P_{1/2}$ state and one of the sublevels of the $6S_{1/2}$ state. Accordingly, this method leads to the excitation of two groups of atoms, which have different velocities and accordingly emit pulses with different frequencies, $\nu_{fF}+k_{sr}v_F$ and $\nu_{fF'}+k_{sr}v_{F'}$, respectively, where $\nu_{fF}(\nu_{fF'})$ is the difference between the frequencies of the upper (lower) sublevel of the $7P_{1/2}$ state and one of the sublevels of the $7S_{1/2}$ state, and k_{sr} is the average wave vector of the superradiance photons. We thus expect the superradiance pulses to be modulated at a frequency

$$\nu=\nu_{FF'}\left(1-\frac{k_{SR}}{k_L}\right),$$

where

$$\nu_{FF'}=\nu_{gF}-\nu_{gF'}=\nu_{fF}-\nu_{fF'}.$$

For emission along the direction of the exciting field

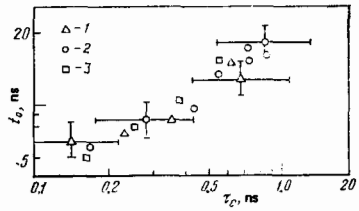

FIG. 12. Dependence of the delay time t_0 on the coherence time τ_c (ref. 56). 1) Beam, $L=2$ cm, $R=273$ μm, $T_2^*=32$ ns; 2) beam, $L=3.6$ cm, $R=366$ μm, $T_2^*=18$ ns; 3) cell, $L=5$ cm, $R=432$ μm, $T_2^*=5$ ns.

TABLE III. Comparison of the theoretical and experimental delay times.

Experimental[56]				Theoretical, t_0, ns		
$\frac{N}{V}10^{-10}$, (cm^{-3})	$\mu_0 L$	τ_c, ns	t_0, ns	Equation (3.5)	Equation (3.32)	Equation (3.29)
6.7	31.3	0.16	6±1	3.2	3.3	5.5
4.3	20	0.25	8±1	4.9	5.0	7.6
2.0	13.5	0.37	10±2	7.1	7.6	10.3
1.9	8.9	0.56	15±3	10.5	11.7	14.3

Note:
Equation (3.5): $t_0=\tau_c\ln N$,
Equation (3.32): $t_0=\left(\frac{1}{\tau_c}-\frac{1}{T_2}\right)^{-1}\ln\frac{N}{\varkappa}\left(1-\frac{\tau_c}{T_2}\right)$,
Equation (3.29): $t\tau=2_0\left[\sqrt{\left(1-\frac{\tau}{T_2}\right)^2+4\frac{\tau}{\tau_c}}-\left(1+\frac{\tau}{T_2}\right)\right]^{-1}\ln\frac{N}{\varkappa}$.

($k_{sr}/k_L>0$), the frequency will be red-shifted; if k_{sr} and k_L have opposite signs, the frequency will be blue-shifted. Such a shift was observed experimentally in Ref. 57. An important feature of the experiments of Ref. 57 was that the beats arose, not as a result of the formation of a coherent superposition of states of one atom, but because of a coherent superposition of states of physically distinct atoms. A similar procedure can be used to measure isotopic shifts.

Superradiance was observed in rubidium vapor by Grubellier *et al.*[58] Transverse pumping of atomic beams kept the Doppler width small. A study of the polarization dependence of the superradiance pulses revealed several new features of the superradiative decay—not found in the case of the isotropic spontaneous decay of a system of atoms. These new features may find applications in spectroscopy.

Flusberg *et al.*[59] observed superradiance in thallium vapor. They observed superradiance at a record short wavelength, $\lambda=1.7$ μm.

Gross *et al.*[60] observed superradiance in sodium vapor vapor. This was the first observation of superradiance in the case of cascading transitions of atoms.

To conclude this section, we will briefly compare the theoretical and experimental results. As we mentioned earlier, "pure" single-pulse superradiance was observed in the experiments of Ref. 56, where special measures were in addition taken to arrange conditions conforming with the single-mode model.

It is thus natural to compare the theoretical results specifically with the results of those experiments (Table III). The experimental data in Table III correspond to the series of measurements indicated by the squares in Fig. 12. We see, first, that the effect was also observed in weakly amplifying media in Ref. 56, since the condition $\ln N\approx 19$ held in the experiments of Ref. 56. According to the data in Table III, the results calculated from Eq. (3.29) agree best with the results of this series of experiments. This conclusion is not surprising, since Eq. (3.32) follows from (3.29) in the case $\tau\ll\tau_c$, as we mentioned eariler. In turn, Eq. (3.5) follows from (3.32) in the case $\mu L\gg\ln N$, while for the characteristics listed in Table I we have $\mu L\lesssim\ln N$, as we mentioned above, and $\tau\sim\tau_c$.

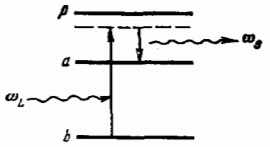

FIG. 13. Scheme of intramolecular transitions in superradiative Raman scattering. a, b) Working levels; p) intermediate level. $\omega_L = \omega_{ab} + \omega_s$; $\hbar\omega_L$ is the pump photon; and $\hbar\omega_s$ is the photon of the Stokes component of supperradiative Raman scattering.

6. SUPERRADIANCE IN RAMAN SCATTERING (MULTIMODE THEORY)

The questions which we have been discussing in the preceding sections refer to the case of single-photon superradiance, in which the frequency of the emitted light is equal to the transition frequency, $\omega = \omega_0$. There is of course also interest in theoretical and experimental study of the corresponding effect in two-photon (or, in general, multiphoton) processes. A first step in this direction was taken in a study of the superradiative Raman scattering of light in molecular and atomic systems. This scattering is an example of a two-photon process in which the pump photon ω_L generates a photon corresponding to Stokes scattering, ω_s (Fig. 13).

As in the case of single-photon superradiance, there are two ways to produce superradiative states in the case of superradiative Raman scattering. The first way is to excite the medium beforehand with a coherent field. The macroscopic polarization induced by this field leads to a transient Raman scattering whose intensity is proportional to the square of the number of scattering particles.[61,62] In this case the interatomic interactions caused by the field radiated by the atoms themselves are unimportant. This effect has been observed experimentally.[63] The second process, in which we are also interested, is the occurrence of superradiative Raman scattering in an originally noncoherent system of atoms by virtue of the spontaneous induction of interatomic correlations in the course of the scattering.[64,22]

The approach taken below[22] toward a description of multimode superradiative Raman scattering automatically also yields the results for the case of single-photon superradiance. (In this sense, this approach is an alternative to that used in Subsection 3c). The basic advantages of this approach are the clarity of the physical interpretation of the formation of the superradiative state, the description of the angular distribution of the superradiance, and the identification of the relationship between superradiance and nonequilibrium phase transitions.

Basic equations of the theory of superradiative Raman scattering and analysis of their solution

We assume that a plane electromagnetic pump wave acts on a system of N multilevel atoms (or molecules) in a volume of arbitrary geometric shape:

$$\mathbf{E}_L(\mathbf{r}_j, t) = \mathbf{E}_{Lj}^{-}(t) + \mathbf{E}_{Lj}^{+}(t) = \mathbf{e}_L\{E_L \exp[-i(\omega_L t - \mathbf{k}_L\mathbf{r}_j)] + \text{c.c.}\}, \quad (6.1)$$

where $E_L = 0$ for $t < 0$ and $E_L = \text{const}$ for $t > 0$.

All the atoms are initially in the ground state (b), and the average polarization of the medium is zero. Raman scattering in the medium gives rise to a Stokes field at the frequency $\omega_s = \omega_L - \omega_{ab}$, where $\omega_{ab} > 0$ is the frequency of the transition between the pair of levels in which we are interested:

$$\mathbf{E}_S(\mathbf{r}_j, t) = \mathbf{E}_{Sj}^{-}(t) + \mathbf{E}_{Sj}^{+}(t) = \mathbf{E}_{Sj}^{-}e^{-i\omega_S t} + \mathbf{E}_{Sj}^{+}e^{i\omega_S t}. \quad (6.2)$$

To describe the superradiative Raman scattering we introduce operators representing the fields, $\mathbf{E}(\mathbf{r}_j, t)$, instead of the creation and annihilation operators for the photons, $a_\mathbf{k}$ and $a_\mathbf{k}^+$, which we used previously. The relationships between these operators are described by Eq. (6.6) below. This description is the conventional one for stimulated Raman scattering, and it makes it easier to compare the theory of superradiative Raman scattering with the theory of transient stimulated Raman scattering.[65]

Equations for the atomic variables which describe the population difference and the dipole moment of the selected pair of levels, a and b, are written in the form[22]

$$\frac{d\sigma_3^{(j)}}{dt} = -\frac{i r_{ab}}{\hbar}(E_{Lj}^{-}(t)\,\mathbf{E}_{Sj}^{+}(t)\,\sigma_+^{(j)} + \sigma_+^{(j)}E_{Lj}^{-}(t)\,\mathbf{E}_{Sj}^{+}(t)) + \text{H.c.}, \quad (6.3)$$

$$\frac{d\sigma_+^{(j)}}{dt} - \left(i\omega_{ab} - \frac{1}{T_2}\right)\sigma_+^{(j)} = -\frac{i r_{ba}}{2\hbar}[\sigma_3^{(j)}\mathbf{E}_{Sj}^{-}(t)\,E_{Lj}^{+}(t) + E_{Sj}^{-}(t)\,E_{Lj}^{+}(t)\,\sigma_3^{(j)}], \quad (6.4)$$

where

$$r_{bv} = \frac{1}{\hbar}\sum_p\left[\frac{(\mathbf{d}_{bp}\mathbf{e}_L)\,\mathbf{d}_{pa}}{\omega_{bp}+\omega_L} + \frac{\mathbf{d}_{bp}(\mathbf{d}_{pa}\mathbf{e}_L)}{\omega_{bp}-\omega_S}\right].$$

Solving Maxwell's equation, we can express the Stokes scattered field in terms of the sum of the contributions of the individual atomic dipole moments at the Stokes frequency, $\mathbf{P}_{Si} = \mathbf{d}_S\sigma_+^i e^{-i(\omega_L t - \mathbf{K}_L\mathbf{r}_i)} + \text{H.c.}$ (with $d_S = r_{ab}E_L$), to the vacuum field $\mathbf{E}_{S0}$:

$$\mathbf{E}_{Sj}^{+} = \frac{\omega_S^2}{c^2}\sum_i \frac{1}{r_{ij}}\rho_-^{(i)}(t')\,[[\mathbf{d}_S^*\mathbf{n}_{ij}]\,\mathbf{n}_{ij}]\exp\left(-i\frac{\omega_S}{c}r_{ij}\right), \quad (6.5)$$

where

$$\rho_-^{(i)}(t') = \sigma_-^{(i)}\exp[i(\omega_{ab}t' - \mathbf{k}_L\mathbf{r}_i)], \quad t' = t - \frac{r_{ij}}{c}, \quad \mathbf{n}_{ij} = \frac{\mathbf{r}_{ij}}{r_{ij}},$$
$$\mathbf{E}_{S0}(\mathbf{r}_j, t) = i\sum_\mathbf{k}\sqrt{\frac{2\pi\hbar kc}{V}}\,\mathbf{e}_\mathbf{k}a_\mathbf{k}\exp[-i(kct - \mathbf{k}\mathbf{r}_j)] + \text{H.c.} \quad (6.6)$$

and $a_\mathbf{k}^+$ and $a_\mathbf{k}$ are the creation and annihilation operators for the field quanta, which satisfy the conditions

$$\langle a_\mathbf{k}a_{\mathbf{k}'}^+\rangle = \delta_{\mathbf{k}\mathbf{k}'}, \quad \langle a_\mathbf{k}^+a_{\mathbf{k}'}\rangle = 0. \quad (6.7)$$

Here and below, the angle brackets denote the average of the operators over the vacuum field and over the ensemble.

By virtue of (6.5), a study of the scattered field $\mathbf{E}_{Sj}^{+}$ reduces to an analysis of the dynamics of the polarization of the atomic system, which is governed in turn by the dynamics of the population difference σ_3 and by the fields E_L and $\mathbf{E}_{S0}$. At this point it is convenient to introduce

$$S_{ij} = \langle\rho_-^{(i)}\rho_+^{(j)} + \rho_+^{(j)}\rho_-^{(i)}\rangle$$

and the total population difference $R_3 = \sum_j\langle\sigma_3^{(j)}\rangle$. Substituting (6.5) into (6.3) and (6.4), using expansion (6.6) for $\mathbf{E}_{S0}$, and averaging the resulting equations over the vacuum state and the ensemble,[22] we find the following

system of equations:

$$\frac{dR_3}{dt}=\frac{1}{T_{1S}}\sum_{i\neq j}C_{ij}S_{ij}+\frac{1}{T_{1S}}(N-R_3), \tag{6.8}$$

$$\left(\frac{d}{dt}+\frac{1}{T_2}\right)S_{ij}=\frac{1}{2T_{1S}}C_{ij}\{(\langle\sigma_3^{(i)}\rangle-1)\langle\sigma_3^{(j)}\rangle+(\langle\sigma_3^{(j)}\rangle-1)\langle\sigma_3^{(i)}\rangle\}$$
$$-\frac{1}{2T_{1S}}\sum_{h\neq i\neq j}C_{hi}S_{hj}\langle\sigma_3^{(i)}\rangle-\frac{1}{2T_{1S}}\sum_{h\neq i\neq j}C_{hj}S_{ih}\langle\sigma_3^{(j)}\rangle. \tag{6.9}$$

Here $C_{ij}=\sin(\omega_S r_{ij}/c)(\omega_S/c)r_{ij}$ is the matrix of the effective interaction between atoms which results from the radiation at the Stokes frequency. In deriving (6.8) and (6.9) we ignored the retardation in the slow part of the density matrix, setting $t'=t-r_{ij}/c\approx t$ in (6.5), as we are justified in doing under the condition $L/c\ll\tau_c$:

$$\frac{1}{T_{1S}}=2\omega_S^3|r|^2|E_L|^2(5\hbar c^3)^{-1},\quad |r|=\frac{1}{\hbar}\left|\sum_p\left(\frac{d_{bp}d_{pa}}{\omega_{bp}+\omega_L}+\frac{d_{bp}d_{pa}}{\omega_{bp}-\omega_S}\right)\right|. \tag{6.9'}$$

Equation (6.8) describes the dynamics of the total population difference. At $t=0$ we have $S_{ij}=0$, and $R_3(0)=-N$. The beginning of the time evolution of R_3 is determined by the spontaneous Raman scattering, which corresponds to the last term in (6.8). The first term on the right side of (6.9) describes the induction of interatomic correlations by virtue of the spontaneous emission of a Stokes photon by one atom and a reaction to it by another atom. If this term were not here, we would have $S_{ij}=0$ at all times, and there would be no superradiance, as can be seen from (6.9) and the given initial conditions ($S_{ij}=0$ at $t=0$). Since the spatial dimension of the superradiative Raman scattering pulse satisfies $c\tau_c>L$ by assumption, we can assume that all the population differences are the same in (6.9): $\langle\sigma_3^{(j)}\rangle=R_3/N$.

The incorporation of the dynamics of the populations is a fundamental distinction between superradiative Raman scattering and transient stimulated Raman scattering, which is usually described by ignoring population changes.[65]

We can use Eq. (6.9) to relate the theory of superradiance to the theory of phase transitions. If we set $(d/dt)S_{ij}=0$ in the equation for S_{ij}, this equation assumes the form of the Ornstein–Zernike equation for a binary correlation function in the theory of equilibrium phase transitions.[66] From this standpoint, superradiance could be interpreted as a nonequilibrium phase transition described by a time-dependent equation. This analogy has not been pursued elsewhere, and we would like to do so here.

To solve Eqs. (6.3) and (6.9), we use the eigenfunctions ψ_λ and the eigenvalues of the matrix λ in Eqs. (3.19) and (3.20).

Transforming to the collective quantity $S(\lambda,t)=\sum_{i\neq j}S_{ij}(t)\psi_\lambda(r_i)\psi_\lambda(r_j)$ in (6.8) and (6.9), and using the properties (3.20), we obtain the system of equations

$$\frac{dR_3}{dt}=\frac{1}{T_{1S}}[\lambda_0\varkappa S(\lambda_0,t)+(N-R_3)], \tag{6.10}$$

$$\frac{dS(\lambda_0,t)}{dt}=-\frac{1}{T_{1S}}\frac{R_3(N-R_3)}{N^2}\lambda_0-\frac{1}{T_{1S}}\frac{R_3}{N}\lambda_0 S(\lambda_0,t). \tag{6.11}$$

In deriving (6.10) and (6.11) we used the relation $\sum_\lambda\lambda S(\lambda,t)\approx\varkappa\lambda_0 S(\lambda_0,t)$ and we set $1/T_2=0$.

Eliminating $S(\lambda_0,t)$, we obtain an equation for $R_3(t)$:

$$\frac{d^2R_3}{dt^2}+\frac{\lambda_0}{2T_{1S}N}\frac{dR_3^2}{dt}-\frac{1}{T_{1S}}\frac{d(N-R_3)}{dt}=\frac{1}{T_{1S}^2}\frac{R_3(R_3-N)}{N}\lambda_0\left(1-\frac{\lambda_0\varkappa}{N}\right) \tag{6.12}$$

with the initial conditions $R_3(0)=-N$, $dR_3^{(0)}/dt=2N/T_{1S}$. After the replacement $R_3\to-R_3$, $T_{1S}\to T_1$, Eq. (6.12) describes the superradiative dynamics of the population difference in a system of two-level atoms which is initially inverted [cf. (3.28)]. This equation contains two important generalizations compared to Eq. (2.9), which was derived for the case of a system with dimensions smaller than a wavelength. Equation (6.12) corresponds to (2.9) if we set its right side equal to zero. Then its exact solution is

$$R_3(t)=N\left[\left(1+\frac{1}{\xi}\right)\operatorname{th}\frac{t-t_0^*}{2\tau_c}-\frac{1}{\xi}\right], \tag{6.13}$$

$$\tau_c=\frac{T_{1S}}{\lambda_0},\quad t_0^*=\tau_c\ln\lambda_0,\quad \xi\equiv\lambda_0, \tag{6.14}$$

which corresponds to an increase in the duration of the pulse of the distributed system, τ_c, by a factor of N/λ_0 in comparison with the case of the system with dimensions smaller than a wavelength [cf. (2.11)].

Ignoring the spontaneous terms, we obtain from (6.10) and (6.11) an analog of the law expressing the conservation of the length of the Bloch vector:

$$R_3^2+2N\varkappa S(\lambda_0,t)\approx N^2. \tag{6.15}$$

Using (6.13), we then find

$$S(\lambda_0,t)=\frac{N}{2\varkappa}\left\{1-\left[\left(1+\frac{1}{\xi}\right)\operatorname{th}\frac{t-t_0^*}{2\tau_c}-\frac{1}{\xi}\right]^2\right\}\approx\frac{N}{2\varkappa}\operatorname{sech}^2\frac{t-t_0^*}{2\tau_c}. \tag{6.16}$$

At the time $t=t_0^*$, we have $R_3(t_0)=0$, $S(\lambda_0,t_0)=\max$, and the system is in a superradiative state. The complete equation (6.12) cannot be solved analytically, but it follows from the general form of this equation that the delay t_0 must in fact be greater than predicted by Eq. (6.14), since the right side of (6.12) vanishes when the spontaneous "seed" in (6.11) (the first term on the right) is increased by a factor of $N/\lambda_0\varkappa\gg1$. It is easy to see that with such an increase in the spontaneous term the initial growth rate S which follows from (6.11) becomes the same as the corresponding expression which we obtain from the conservation law (6.15) with the help of (6.10). Thus, the increase in the delay t_0 results from a violation of the conservation law (6.15) at early times.

We can approximate t_0 in the following manner. Assuming $R_3(t)\approx-N$ for $0<t\leq t_0$, we obtain from (6.11) and (6.16)

$$S(\lambda_0,t_0)=2\left(\exp\frac{\lambda_0 t_0}{T_{1S}}\right)\approx\frac{N}{2\varkappa}.$$

Hence we have

$$t_0=\frac{T_{1S}}{\lambda_0}\ln\frac{N}{\varkappa}. \tag{6.17}$$

It follows from (6.17), (3.22), and (6.9') that the delay of the superradiative Raman scattering pulse t_0 (and also the pulse length, τ_c) is in the case $\mathcal{F}\gg1$ inversely proportional to the product of the number density of scattering particles, $n=N/V$, the pump intensity $|E_L|^2$, and the length of the medium, L.

Using (6.12) and (6.13), we can write t_0 as

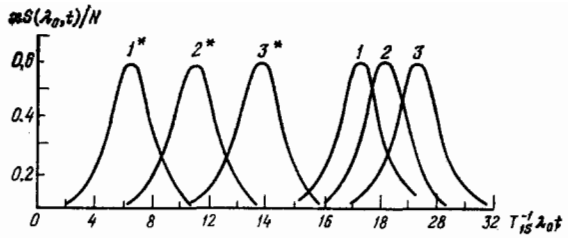

FIG. 14. Shape of the superradiative Raman scattering pulse; comparison of the delay times t_0 and t_0^*. 1–3) Numerical solution of (6.10) and (6.11); 1*–3*) calculation from Eq. (6.16). $L=10$ cm, $A=1$ cm^2, $N=10^{14}$. 1, 1*) $\lambda_s=2\pi\omega_s/c=10^{-5}$ cm; 2, 2*) $\lambda_s=10^{-4}$ cm; 3, 3*) $\lambda_s=10^{-3}$ cm.

$$t_0=\frac{T_{1S}}{\lambda_0}\left(\ln\lambda_0+\ln\frac{\omega_S L}{c}\right), \quad \mathcal{F}\gg 1,$$
$$t_0=\frac{T_{1S}}{\lambda_0}\left(\ln\lambda_0+\ln\frac{\omega_S^2 R^2}{\pi c^2}\right), \quad \mathcal{F}\ll 1,$$

i.e., $t_0 > t_0^*$.

If $\varkappa\simeq 1$, we find, by using $\lambda_0=N\pi/k_s^2 A$ in (6.17), that the result in (6.17) is the same as Eq. (3.5), which holds for the single-mode model. Thus, the condition for the applicability of the single-mode approximation in describing superradiative Raman scattering is the condition $\varkappa\simeq 1$, i.e., $\mathcal{F}\simeq 1$. A numerical solution of the system (6.10), (6.11) confirms the estimate (6.17). Figures 14 and 15 show the results found for $S(\lambda_0,t)$ and $R_3(t)$ through a numerical solution of (6.10), (6.11); shown for comparison are the results on $S(\lambda_0,t)$ and R_3 found from Eqs. (6.13) and (6.16).

It follows that the results derived above hold under the condition $t_0 \ll T_2$. If, on the other hand, $t_0 \gtrsim T_2$, then we must add a term $-T_2^{-1}S(\lambda_0,t)$ to the right side of (6.11). Then by analogy with the derivation of (6.17), we obtain from (6.11) and (6.16)

$$t_0=\tau_c\left(1-\frac{\tau_c}{T_2}\right)^{-1}\ln\frac{N}{\varkappa}\left(1-\frac{\tau_0}{T_2}\right).$$

Superradiative Raman scattering is thus also possible in the case $t_0 \gtrsim T_2$, but with $\tau_c < T_2$. This situation corresponds to resonant superradiance in weakly amplifying media (see Subsection 3c, part 3).

We turn now to the shape of the superradiative Raman-scattering pulse and the angular distribution of the radiation. Using (6.5), we can express the energy scattered per unit time into a unit solid angle along the direction of the unit vector $\hat{k}$ in terms of S_{ij} and R_3;

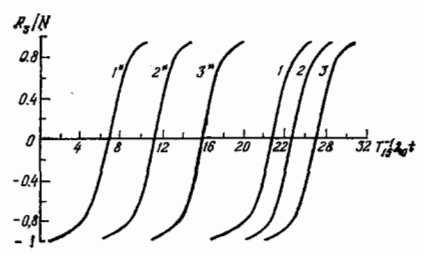

FIG. 15. Dynamics of the population difference in superradiative Raman scattering. The parameters and notation are the same as in Fig. 14.

then we can transform to the collective quantiy $S(\lambda,t)$. We find[22]

$$I_{S,\hat{k}}(t) \quad \frac{\hbar\omega_S}{4\pi T_{1S}}\left[\frac{N-R_3(t)}{2}+\frac{1}{2}\sum_{\lambda}S(\lambda,t)\sum_{i\neq j}\psi_\lambda(\mathbf{r}_i)\psi_\lambda(\mathbf{r}_j)e^{i\mathbf{k}\mathbf{r}_{ij}}\right].$$

In the limits $\mathcal{F}\ll 1$ and $\mathcal{F}\gg 1$ we find with the help of (3.23) explicit expressions for $I_{S,\hat{k}}$ explicit expressions for $I_{S,\hat{k}}$

$$I_{S,\hat{k}}=\frac{\hbar\omega_S}{4\pi T_{1S}}\left[\frac{N-R_3(t)}{2}+\frac{N}{2}\varkappa S(\lambda_0,t)\Gamma(\hat{k})\right], \tag{6.18}$$

where $R_3(t)$ and $S(\lambda_0,t)$ are given by Eqs. (6.13) and (6.16) (with $t_0^*\to t_0$), and the angular-directionality factor of the superradiative Raman scattering is

$$\Gamma(\hat{k})=\frac{1}{2N^2}\sum_{i\neq j}\left[\exp\left(i(\mathbf{k}_0+\mathbf{k})\mathbf{r}_{ij}\right)+\exp\left(i(\mathbf{k}_0-\mathbf{k})\mathbf{r}_{ij}\right)\right]$$
$$=2\sum_{(\pm)}\left[\frac{\sin\frac{1}{2}H(1\mp\cos\varphi)}{\frac{1}{2}H(1\mp\cos\varphi)}\right]^2\left(\frac{J_1(h\sin\varphi)}{h\sin\varphi}\right)^2. \tag{6.19}$$

Here $H=\omega_S L/c$, $h=\omega_S R/c$, $\cos\varphi=\mathbf{k}\mathbf{k}_0/k^2$ (we recall that $\mathbf{k}_0$ is directed along the cylinder axis, $|\mathbf{k}|=|\mathbf{k}_0|$), and J_1 is the Bessel function of order one.

The first term in (6.18) describes isotropic spontaneous Raman scattering which occurs at $t=0$; the second term describes the collective emission which takes the form of a pulse whose maximum is reached at the time t_0. It can be seen from (6.18) and (6.19) that the emission during superradiative Raman scattering is into small solid angles in opposite directions along the cylinder axis. We see from (6.16) and (6.18) that at the time t_0 the scattered power is at a maximum and we have $I_{S,\hat{k}}(t_0)\sim N^2$. When we use the replacements $\omega_S\to\omega_0$, $T_{1S}\to T_1$, $R_3\to -R_3$, we see that Eqs. (6.18) and (6.19) describe the pulse shape and the angular distribution of the superradiance in a system of two-level atoms which are initially inverted in a noncoherent manner. Using Eqs. (6.13) and (6.16) for R_3 and $S(\lambda_0,t)$, we can write, for this case

$$I_{\hat{k}}=\frac{\hbar\omega_0 N}{4\pi T_1}(1+\xi)\frac{1}{e^{t/\tau_c}+\xi}\left[1+N\frac{e^{t/\tau_c}-1}{e^{t/\tau_c}+\xi}\Gamma(\hat{k})\right]. \tag{6.20}$$

If we set $\xi=\lambda_0$, then we find that (6.20) assumes the form of the result found by Eberly and Rehler,[13] which was derived for a medium excited beforehand by a short pulse of coherent light with wave vector $\mathbf{k}_0$. There is a difference: The expression for $\Gamma(k)$ in (6.19) contains two terms which describe the symmetric formation of superradiance in opposite directions along the cylinder axis, and $\mathbf{k}_0$ is now the wave vector of the eigenfunction of the interaction matrix in (3.23). If we take into account, that we should in fact actually use the replacements $t_0^*\to t_0$ and $\xi\to N/\varkappa$ in (6.13) and (6.16), we find that Eq. (6.20) remains valid if we set $\xi=N/\varkappa$ in it. This approach corresponds to an increase in the delay of the superradiance pulse to the value t_0 in (6.17).

Equation (6.20) clearly shows the time evolution of the intensity and angular distribution of the superradiance (and, correspondingly, the superradiative Raman scattering). At $t=0$ we have

$$I_{\hat{k}}(0)=\frac{\hbar\omega_0}{4\pi T_1}N,$$

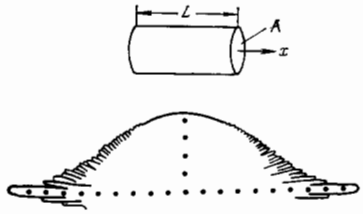

FIG. 16. Spatial distribution of the superradiance from a cylindrical medium [according to Eq. (6.20)]. The cylinder axis coincides with the x axis. The distribution is found by imposing a symmetry with respect to the two directions along the x axis on the diagram in Ref. 13 (the result of a numerical calculation). The Fresnel number is $\mathcal{F}=A/\lambda L$, where λ is the wavelength, $V=(10^5\lambda^3/8\pi^2)$ is the volume of the cylinder, and the density of active atoms is $61.2/\lambda^3$. The scale is radially logarithmic; each successive point along the axes represents an increase in the intensity by an order of magnitude. We see that in the superradiative state the emission intensity along the axis of the cylinder is roughly six orders of magnitude higher than that in the perpendicular direction. The diagram also shows the intensity distributions for superradiative Raman scattering.

which corresponds to isotropic spontaneous scattering. At the time $t=t_0=\tau_c \ln N/\varkappa$ we have

$$I_{\hat{k}}(t_0)=\frac{\hbar\omega_0}{4\pi T_1}\frac{1+\xi}{2\xi}\left[1+N\frac{\xi-1}{2\xi}\Gamma(\hat{k})\right]\approx\frac{\hbar\omega_0}{8\pi T_1}N^2\Gamma(\hat{k}). \qquad (6.21)$$

(We used $\xi=N/\varkappa\gg 1$ in the derivation of the last equality.) Thus, at the time t_0, the system radiates in opposite directions along the cylinder axis, for which we have $\Gamma(\hat{k})\simeq 1$, at a rate N^2 higher than the rate of the spontaneous emission from an individual atom (see Fig. 16).

This analysis shows that in the approximation of a given pump field the problem of describing superradiative Raman scattering turns out to be completely analogous to the problem of describing single-photon superradiance. Correspondingly, in this approximation, the condition for observation of Raman-scattering superradiance turns out to be analogous to the condition for superradiance: $L/c\ll\tau_c\ll T_2(\tau_c\sim 1/n)$. Then for a given pump intensity I_L there is a lower limit on the density of the medium, $n>n_{\min}(I_L)$.

For clarity, we can express the conditions for observation of superradiative Raman scattering in the approximation of a given pump field in terms of the amplification coefficient for steady-state stimulated Raman scattering, $G=2\pi n d_{\mathrm{R}}^2\omega_S/\hbar T_2^{-1}c$. Using the expression for τ_c in (6.14), with λ_0 given by (3.22), we find

$$\frac{L}{c}\ll\frac{T_2}{GL}\ll T_2. \qquad (6.22)$$

In time-dependent stimulated Raman scattering, which occurs (like superradiative Raman scattering) over times shorter than T_2, the growth of the amplitude of the Stokes wave over the length of the medium, L, is proportional to $\exp\sqrt{GL^2/T_2c}$. By virtue of the inequality at the left in (6.22), we have $\sqrt{GL^2/T_2c}\ll 1$ in superradiative Raman scattering, and we can ignore stimulated amplification.

The pump field can be assumed given if the input energy over the radiation time $2\tau_c$ is greater than the energy drawn from the pump field, i.e., if

$$\frac{|E_L|^2 c 2\tau_c A}{2\pi}>n\hbar\omega_L AL,$$

Also using (6.14), (3.22), and the expression for T_{1S} in (6.9′), we obtain

$$n<n_{\max}=\frac{c/L}{\pi\sqrt{2\omega_S\omega_L|r|}}. \qquad (6.23)$$

Accordingly, depletion of the pump leads to an additional upper limit on the number density of scattering particles. For $L\sim 1$ cm, $\omega_S\sim\omega_L\sim 10^{15}$ s^{-1}, and $|r|\sim 10^{-24}$ esu, we find $n_{\max}\sim 10^{19}$ cm^{-3}. The calculation for superradiative Raman scattering in the single-mode model in Ref. 7, with allowance for pump depletion, confirms (6.23).

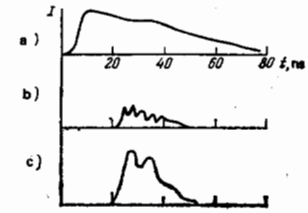

FIG. 17. Oscilloscope traces of various pulses. a) Pump; b) anti-Stokes component of superradiative Raman scattering; c) Stokes component of superradiative Raman scattering.[68]

Superradiative Raman scattering was apparently first observed by Pivtsov *et al.*[68] In their experiments, they studied the $Q_{01}(1)$ Raman line in H_2, for which $T_2\sim 7.5\cdot 10^{-9}$ s·atm and $T_1\gg T_2$. The pump wavelength was $\lambda_L=694.3$ nm; $\lambda_S=976$ nm; the length of the cell was $L=30$ cm; and $n\sim 7.2\cdot 10^{19}$ cm^{-3}. The pump energy was $W=0.2$–0.5 J; the pulse length was 50 ns, the rise time was 2–3 ns (Fig. 17); and the light was focused to a spot 0.2 mm in diameter. The most important result of those experiments was the observation of a substantial delay of the Stokes and anti-Stokes Raman-scattering pulses with respect to the peak of the pump pulse. With $P=2.7$ atm and $W=0.35$ J, this delay was 17 ± 5 ns, falling off with increasing pump power and with increasing pressure, roughly in proportion to the reciprocal of the product $n|E_L|^2$. The pulses had an oscillatory structure (Fig. 17).

7. CONCLUSION

As mentioned above, the basic characteristics of superradiance are a high directionality, a quadratic dependence of the maximum intensity on the particle density, and an inverse proportionality between the pulse length and the particle density. These characteristics determine the possible applications of the effect as a source of intense coherent pulses whose intensity and length can be varied over broad ranges in a rather simple manner. Furthermore, while superradiance sources would become alternatives to existing coherent laser sources in the infrared, visible, and ultraviolet ranges, they would be the only possible type of coherent sources in the vacuum-UV, x-ray, and γ ranges, as mentioned above. The two-photon superradiance may prove pertinent to the development of a

tunable laser which makes use of two-photon emission in an inverted medium.

Research on superradiance is continuing, and the number of papers on this subject is continuously growing.

A complete theory of superradiance will require further work on such questions as how to incorporate the field propagation in the quantum model for an arbitrary geometric configuration of the medium, for both single photon and multiphoton processes: the spatial and temporal coherence of superradiance; and the relationship between superradiance and nonequilibrium phase transitions. So far, most of the experimental work on superradiance has been restricted to single-photon transitions in the optical part of the spectrum. It would be interesting to see a demonstration of superradiance corresponding to multiphoton transitions, an experimental study of the spatial and temporal coherence of the superradiance, and a demonstration of superradiance at shorter wavelengths.

In this review we have been interested in electromagnetic superradiance. Methods using effects similar to superradiance (for example, the photon echo[78,79]) are widely used in spectroscopy.

Corresponding methods have been developed extensively in acoustics.[75,76] They have been used to develop new types of acoustic sources, to find a more detailed explanation of the relaxation of impurity centers, and to improve substantially our ability to study metals and impurity centers, by supplementing rf-spectroscopy methods.

The method of exciton superradiance[77] promises to become an effective tool for studying molecular crystals and semiconductors.

This progress raises the hope that optical superradiance will also find widespread applications in spectroscopy, where it may prove effective as a method for amplifying the response of systems to transitions with small dipole moments.

APPENDIX. PHASE TRANSITION IN AN EQUILIBRIUM SYSTEM OF TWO-LEVEL ATOMS INTERACTING THROUGH AN ELECTROMAGNETIC FIELD

The superradiance which we have discussed here is a process of relaxation to equilibrium in an originally very non-equilibrium system of two-level atoms which are interacting with a radiation field. It is also of interest to examine equilibrium properties of this system.

Hepp and Lieb[69] (see also Ref. 70) have found that in the equilibrium system with the Hamiltonian in (2.1), taken in the single-mode approximation, a phase transition may occur with the appearance, at a temperature above a certain critical level, T_{cr}, of an average photon-mode occupation number $\langle a_k^+ a_k \rangle \sim N$, where N is the number of atoms (Bose condensation of photons). At $T > T_{cr}$, $\langle a_k^+ a_k \rangle$ is determined by the Planck formula and is independent of N. A necessary condition for the phase transition is $8\pi n d^2/\hbar\omega_0 > 1$, where d is the dipole matrix element, and n is the density of atoms.

It should be noted that the fundamental question of the frequency at which the Bose condensation occurs was not discussed in the original papers,[69,70] and in some subsequent papers it was asserted that there is a condensation of photons with a frequency ω_k or with the frequency of the atomic transition, ω_0 (phase transition to a "superradiative state"). The reader is referred to Refs. 71 and 72 for a critical analysis of the possibility of an equilibrium phase transition to a superradiative state and for a more extensive bibliography.

The frequency of the Bose condensate was studied in Refs. 73 and 74. Elesin and Kopaev[74] used a model with the Hamiltonian in the Dicke approximation.[1] Emel'yanov and Klimontovich[73] studied the situation for extended systems on the basis of the total Hamiltonian, and they found the relationship between the Dicke approximation and the concept of an effective field. It was shown in these papers that there is a "soft" mode, with a frequency which vanishes at $T = T_{cr}$, in the spectrum of collective excitations of this system. This result means that the frequency of the Bose condensate is zero, and the phase transition is accompanied by the spontaneous appearance of a macroscopic polarization at zero frequency or a constant electric field $E = \lim_{k\to 0} \sqrt{(2\pi\omega_k \hbar/V)\langle a_k^+ a_k \rangle}$. In other words, a ferrolectric phase transition occurs.

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