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Semiclassical theory of cooperative radiation of a polyatomic system

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The kinetics of a pulse of cooperative radiation is considered with account taken of the homogeneous and inhomogeneous broadening of the spectrum. It is shown that in the case of sufficiently large homogeneous broadening, the Dicke superradiance is transformed into superluminescence, which can be described by the balance equations. The existence of an optimal gain length corresponding to the maximum intensity of the cooperative radiation is proved.

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I. INTRODUCTION

The cooperative character of the spontaneous emission of a polyatomic system is due to exchange of real and virtual transverse photons between atoms. Dicke,[1] followed by others,[2,3] have shown that for a system of N atoms in a small volume, with linear dimensions shorter than the wavelength, the radiation intensity is proportional to N. The proportionality of the intensity to the square of the number of particles shows that phase alignment of the atomic dipole takes place in the emission process, although the macroscopic moment may be equal to zero in the initial state.

The present paper is devoted to an investigation of the cooperative spontaneous emission of extended polyatomic system with account taken of the homogeneous and inhomogeneous broadening of the luminescence spectrum. Examples of such systems are pulsed gas lasers without mirrors.[4–6] Introduction of the broadenings is essential not only for the determination of the details of the structure of a cooperative-emission pulse, but also to find the conditions for the manifestation of the cooperative effect. The quantum theory of superradiance of extensive systems without allowance for the broadening of the spectrum was developed in[7–9]. The use of a more elaborate model makes a semiclassical approximation (a quantum description of the atomic system and a classical description of the electromagnetic field) more advantageous. In particular, it becomes possible to trace the connection between the theory that describes the Dicke superradiance and the balance equations that are used in the theory of pulsed lasers.[10] The transition to the latter is realized in the case of sufficiently large homogeneous broadening of the spectrum, which leads to loss of the phase memory of the atomic system. Pulsed cooperative emission, during the course of which the phase memory of the atomic system is preserved, will be called superradiance. On the other hand, if the relaxation of the off-diagonal elements of the density matrix of the atoms is effective enough, so that the conditions for the validity of the balance equations are satisfied, then the corresponding radiation regime will be called superluminescence.

2. THE SEMICLASSICAL APPROXIMATION

In the semiclassical approximation, a system of two-level atoms interacting with an electromagnetic field can be described by the system of equations for the single-atom density matrix $\|\rho_{ab}\|$ and the equation for the intensity of the electric field E due to the polarization $\mathbf{P}$ of the medium[11]:

$$
\begin{gathered}
i\hbar\dot{\rho}_{aa}=V_{ab}\rho_{ba}-\rho_{ab}V_{ba},\\
i\hbar\dot{\rho}_{ba}=\hbar\omega\rho_{ba}+V_{ba}\rho_{aa}-\rho_{bb}V_{ba}-i\hbar\frac{\rho_{ba}}{T_2},\\
\left(\Delta-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}-\varkappa'\frac{\partial}{\partial t}\right)\mathbf{E}=\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{P}.
\end{gathered}
\tag{1}
$$

Here $V_{ba}=V_{ab}^*$ is the matrix element of the interaction; in the dipole element we have $V_{ba}=-\boldsymbol{\mu}\cdot\mathbf{E}$, where $\boldsymbol{\mu}$ is the dipole-moment matrix element corresponding to the transition from the excited state b to the ground state a. The density matrix is locally averaged over the positions of the atoms and is a function of the time, of the spatial coordinates, and of the natural frequency ω of the

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atoms, for which we shall introduce a renormalized distribution function

$$g(\Delta)=T_2^*\pi^{-1/2}\exp(-(T_2^*\Delta)^2), \quad \Delta=\omega-\omega_0,$$

that describes the inhomogeneous-broadening contour. The relaxation term in the second equation is responsible for the homogeneous broadening of the spectrum, and T_2 is the transverse relaxation time. The polarization **P** can be expressed in terms of the off-diagonal elements of the density matrix:

$$\mathbf{P}(\mathbf{r},t)=\frac{N}{V}\mu\int g(\Delta)(\rho_{ba}+\rho_{ab})d\Delta, \tag{2}$$

where N is the total number of the atoms and V is the volume of the system. The last term in the left-hand side of the third equation in (1) describes the losses due to the emergence of the radiation from the volume in question; the meaning of the parameter $\varkappa'$ will be made clear later on.

The purpose of our study is to investigate the form of emission pulse, assuming that population inversion exists at the initial instant of time. The parameters that determine the kinetics of the pulse, besides the single-atom characteristics, are the length of the system, the density of the radiating atoms, and the homogeneous and inhomogeneous widths of the spectrum.

In the solution of this problem it is natural to make the following assumptions.

1. We assume that the radiation has a preferred direction and take into account dependence of the density-matrix and of the field on only the one coordinate z along this direction.

2. We assume the polarization of all the vector quantities to be fixed and the same, and will omit henceforth the vector symbols.

3. We shall seek the electric field and the off-diagonal elements of the density matrix in the form of plane waves with slowly varying complex amplitudes:

$$\begin{aligned} E&=\mathscr{E}(z,t)\exp[-i\omega_0 t+ikz]+\text{c.c.},\\ \rho_{ba}&=R^-(z,t)\exp[-i\omega t+ikz],\\ \rho_{ab}&=R^+(z,t)\exp[i\omega t-ikz]. \end{aligned} \tag{3}$$

4. We choose as the initial condition a small homogeneous polarization (with an amplitude independent of z). In this case the amplitudes of the solutions will likewise not depend on z.[12] We note that variation of the initial conditions that imitate the initial radiation or the polarization of the medium have little effect on the shape of the output pulse.[4]

Under the foregoing assumptions, we can transform Eqs. (1) into equations for the amplitudes:

$$\begin{aligned} \left(\frac{d}{dt}+\varkappa\right)A(t)&=g_0N\int R^-(t,\Delta)e^{-i\Delta t}g(\Delta)d\Delta,\\ \frac{d}{dt}R^-(t,\Delta)&=2g_0A(t)Z(t,\Delta)e^{i\Delta t}-\frac{R^-}{T_2},\\ \frac{d}{dt}Z(t,\Delta)&=-g_0(A(t)R^+(t,\Delta)e^{i\Delta t}+A^*(t)R^-(t,\Delta)e^{-i\Delta t}). \end{aligned} \tag{4}$$

Here A is the dimensionless amplitude of the electric field, and the square of its modulus is equal to the number of photons in the volume:

$$A=-i\left(\frac{V}{2\pi\hbar\omega_0}\right)^{1/2}\mathscr{E};\quad \varkappa=\frac{\varkappa'}{2}c^2;\quad Z=\frac{\rho_{bb}-\rho_{aa}}{2},\quad g_0^2=\frac{2\pi\mu^2\omega_0}{\hbar V}.$$

With the aid of the system (4) we can obtain an equation for the balance of the excitations in the volume

$$\frac{d}{dt}\left(A^*A+N\int Z(t,\Delta)g(\Delta)d\Delta\right)=-2\varkappa A^*A. \tag{5}$$

If we assume that the change in the number of excitations is connected only with the emission leaving the volume, then we can put $2\varkappa=c/L$, where L is the length of the volume in the direction of the radiation.

3. SUPERRADIANCE OF AN EXTENDED SYSTEM

Before we proceed to the analysis of the solution of the system (4), with account taken of the homogeneous and inhomogeneous broadening of the spectrum, we recall the principal results pertaining to the idealized case $T_2^{-1}=T_2^{*-1}=0$, which was investigated by Bonifacio *et al.*[12,13] and which lends itself to a graphic interpretation based on the analogy with the problem of the mathematical pendulum. It is convenient to introduce for this purpose, instead of the density matrix, a Bloch vector with components X, Y, Z; $X\pm iY=R^\pm$. By virtue of (4), under the condition $T_2^{-1}=T_2^{*-1}=0$, the Bloch vector conserves its length, $X^2+Y^2+Z^2=\text{const}$. If the initial condition corresponds to total inversion, $Z(0)=1/2$, $X(0)=Y(0)=0$, the length of the Bloch vector is 1/2. The initial state corresponds to a Bloch-vector polar angle $\theta=0$, while the final state corresponds to $\theta=\pi$. It follows from the system (4) that the pendulum equation is satisfied for θ:

$$\ddot\theta+\varkappa\dot\theta-\Omega^2\sin\theta=0, \tag{6}$$

$$\Omega^2=g_0^2N=2\pi\mu^2\omega_0N/\hbar V. \tag{7}$$

The intensity of the radiation (the number of photons emitted from the volume per unit time) is expressed in terms of the kinetic energy of the pendulum:

$$I(t)=2\varkappa A^*A=\dot\theta^2\varkappa/2g_0^2=\dot\theta^2\varkappa N/2\Omega^2. \tag{8}$$

In the case of large damping $\varkappa\gg\Omega$ (we recall that $\varkappa=c/2L$), i.e., at sufficiently small lengths of the system, the regime is aperiodic. The maximum of the radiation intensity (of the kinetic energy of the pendulum) is reached at $\theta\approx\pi/2$. The form of the pulse is given by

$$I(t)=\frac{N}{2\tau_R}\operatorname{sech}^2\frac{t-t_0}{2\tau_R}, \tag{9}$$

$$\tau_R=\varkappa/g_0^2N=\varkappa/\Omega^2. \tag{10}$$

The intensity is proportional in this case to N^2 and is bell-shaped with a width on the order of $2\tau_R$. The parameter t_0 determines the position of the maximum (i.e., the pulse delay time) and can be obtained from the initial conditions. In the case of total inversion, the pendulum is in a position of unstable equilibrium at the initial instant of time, and then $t_0=\infty$. On the other hand if we assume

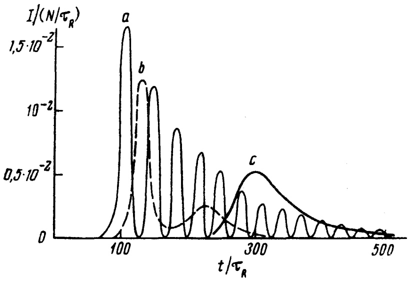

FIG. 1. Pulses of cooperative radiation at different values of the homogeneous width of the spectrum, $\theta(0)=2\cdot 10^{-4}$: a) $\varkappa=0.01\tau_R^{-1}$, $T_2^{*-1}=0$, $\tau_R/T_2=0$; b) $\varkappa=0.01\tau_R^{-1}$, $T_2^{*-1}=0$, $\tau_R/T_2=0.04$; c) $\varkappa=0.01\tau_R^{-1}$, $T_2^{*-1}=0$, $\tau_R/T_2=0.2$.

$$Z(0)={}^1/_2-1/N,\quad X(0)=(1/N)^{1/2},\quad Y(0)=0.$$

then

$$t_0=2\tau_R\ln(2/\theta(0))=\tau_R\ln N.$$

The fast-damping condition $\varkappa\gg\Omega$ is equivalent, by virtue of (10), to the inequality $\tau_R\gg\Omega^{-1}$. When the inverse inequality is satisfied, the Bloch vector executes weakly damped oscillations of frequency Ω, i.e., periodic energy exchange will take place between the atoms and the field. In this case the radiation pulse will consist of a series of decreasing maxima separated by time intervals $\sim\pi/\Omega$. The position of the first maximum is determined by the delay time (1/4 of the period)

$$t_0=\Omega^{-1}\ln(8/\theta(0))=\Omega^{-1}\ln(4N^{-1/2}).$$

The maximum intensity (the intensity of the first maximum) in the weak damping regime can be expressed in terms of the maximum kinetic energy of the pendulum

$$\max I(t)=\max\frac{\theta^2}{2}\frac{\varkappa}{g_0^2}=2\Omega^2\frac{\varkappa}{g_0^2}=2N\varkappa \tag{11}$$

and is consequently proportional to the first power of the number of particles in the system. The duration of the pulse is determined in this case by the time $\sim\varkappa^{-1}$ required for the light to pass through the system.

4. COOPERATIVE RADIATION WITH ALLOWANCE FOR THE HOMOGENEOUS AND INHOMOGENEOUS BROADENING

Homogeneous broadening of the spectrum upsets the phase of the state of the radiating atoms. If the transverse-relaxation time is short enough, so that the inequality

$$\dot R^{\pm}\ll R^{\pm}/T_2 \tag{12}$$

is satisfied and $T_2^*>T_2$ (i.e., the homogeneous broadening exceeds the inhomogeneous broadening), then Eqs. (4) can be reduced to a system of balance equations

$$\dot n=-2\varkappa n+4T_2g_0^2N'n,\quad \dot N'=-4T_2g_0^2nN', \tag{13}$$

where $n=A^*A$ is the number of photons in the volume, and

$$N'=N\int Z(t,\Delta)g(\Delta)\,d\Delta$$

is half the population difference.

It is known that these equations describe the process of amplification of spontaneous luminescence (superluminescence), which stops when the populations become equal, i.e., at $N'=0$, whereas in the superradiance regime we have $N'\to(-1/2)N$.

If the condition $\varkappa>\tau_R^{-1}$ is satisfied (i.e., for short lengths), the characteristic emission time is τ_R, and therefore the condition (12) can be written in the form $\tau_R>T_2$. But the ratio T_2/τ_R is equal to the product αL of the gain by the length, so that satisfaction of the condition (12) is equivalent to the absence of gain.

For a sufficiently extended system ($\varkappa>\tau_R^{-1}$), the characteristic time of variation of $R^{\pm}$ is the half-period of the oscillations $\pi/\Omega=\pi(\tau_R/\varkappa)^{1/2}$ and the damping time $\varkappa^{-1}$, therefore condition (12) does not contradict the presence of gain. A numerical solution of the system (4) shows that homogeneous broadening leads to a decrease and to a broadening of the intensity maxim, to a smoothing of the oscillations, and to an increase of the delay time.

Figure 1 shows cooperative-radiation pulses for different values of the homogeneous width.

A numerical solution of the system (4) has explained also the influence of the inhomogeneous broadening of the spectrum on the radiation kinetics. Inhomogeneous broadening leads to dephasing of the Bloch vectors of the individual atoms in the (X, Y) plane. The appearance of an oscillatory pulse structure shows that an inverse process, analogous to photon echo, takes place also in the radiation process. Just as in the case of homogeneous broadening, an increase of the inhomogeneous width is accompanied by a decrease of the gain (there is no cooperative radiation at $\tau_R/T_2^*\geqslant 2$) and by an increase of the delay time. Comparison with the results of Bonifacio and Lugiato[12] shows that the approximate allowance used by them for the inhomogeneous broadening is apparently valid only at asymptotically small values of τ_R/T_2^*. Figure 2 shows radiation pulses for several values of T_2^*.

Figure 3 shows the dependence of the intensity of the cooperative radiation (the maximum of the first peak) on the number of atoms N. With increasing N the influence of the homogeneous and inhomogeneous broadening

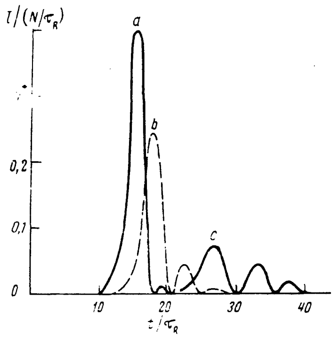

FIG. 2. Pulses of cooperative radiation at different values of the inhomogeneous width of the spectrum, $\theta(0)=2\cdot 10^{-4}$: a) $\varkappa=\tau_R^{-1}$, $T_2^{*-1}=0$, $\tau_R/T_2=0$; b) $\varkappa=\tau_R^{-1}$, $T_2^{*-1}=0.5\tau_R^{-1}$, $\tau_R/T_2=0$; c) $\varkappa=\tau_R^{-1}$, $T_2^{*-1}=\tau_R^{-1}$, $\tau_R/T_2=0$.

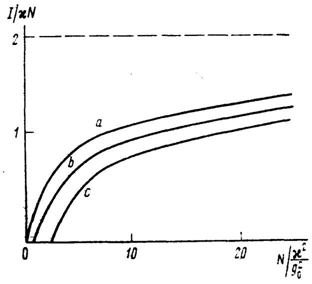

FIG. 3. Dependence of the maximum of the intensity of the cooperative radiation on the number of atoms: a) $T_2^{*-1} = 0, \varkappa^{-1}/T_2 = 0$; b) $T_2^{*-1} = 0$. $\varkappa^{-1}/T_2 = 0.5$; c) $T_2^{*-1} = 0$, $\varkappa^{-1}/T_2 = 1$

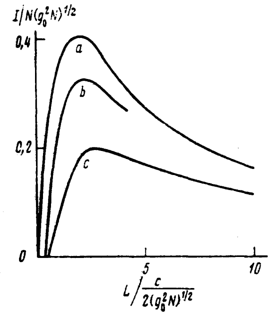

FIG. 4. Dependence of the maximum intensity of the cooperative radiation on the length of the system: a) $T_2^{-1} = 0$, $T_2^{*-1} = 0$; b) $T_2^{-1} = 0$. $T_2^{*-1} = 0.5\,\Omega$; c) $T_2^{-1} = 0.5\,\Omega$, $T_2^{*-1} = 0$.

becomes weaker (if it assumed that T_2 and T_2^* are independent of N), and a superradiance regime with oscillations sets in. The intensity reaches in this case a maximum value equal, as already noted, to $2N\varkappa$. We note that the quadratic dependence of the intensity on N goes over into a linear one in this case.

The most interesting is the dependence of the cooperative radiation on the length of the system. At short lengths, such that $T_2/\tau_R < 1$, there is no gain. With increasing length, the gain increases. If $T_2/\tau_R > 1$ and $\tau_R > \varkappa^{-1}$, then superradiance takes place. With further increase of the length, τ_R becomes smaller than $\varkappa^{-1}$, and at $(\varkappa^{-1}\tau_R)^{1/2} = \Omega^{-1} \gtrsim T_2$ the superradiance goes over into superluminescence. Even though the gain αL increases with increasing length, the maximum intensity per atom decreases, since the pulse time, which is determined by the value of $\varkappa^{-1}$, decreases. At a certain optimal length the intensity maximum per atom becomes largest (see Fig. 4). In the absence of homogeneous or inhomogeneous broadening, this optimal length is determined by the realtion $\tau_R \varkappa = 0.25$.

We can expect in this connection that under pulsed excitation of the system (such as a platelet or a rod) the radiation will not propagate in the direction of the maximum gain if the optimal relation is satisfied for another direction. A similar phenomenon was observed by Galanin *et al.* [14] for superluminescence of thin anthracene plates, although the system parameters cited by them do not allow us to conclude that the optimal condition was satisfied.

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