Accounting for dipole-dipole interactions in the theory of superradiance

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(Submitted 10 January 1995)
Zh. Éksp. Teor. Fiz. **107**, 1845–1852 (June 1995)

On the basis of previously obtained equations for the partial atomic dipole densities, which are more general than the Bloch equations, we have carried out an analysis of the motion of dipoles in strongly optically excited gases. We show that dipole-dipole interactions can cause not only dephasing of the dipoles, but also coherent collective oscillations of the optical dipoles with characteristic frequency ω_{osc}. As a result, we have established that coherent alignment of dipoles, necessary for the generation of an observable pulse of superradiation, arises under the influence of strong dipole-dipole interactions, and not as a result of interactions of optical dipoles with the radiation reaction field, as was previously assumed in the theory of superradiance. © *1995 American Institute of Physics.*

1. INTRODUCTION

Superradiant pulses, emerging from strongly optically-excited matter, have been intensively studied both experimentally and theoretically.[1-3] However, so far, even the simplest concentration model of superradiance only takes account of the precession of the dipoles in the radiation reaction field[1-3]

$$E_r=\frac{2\omega^3 Nd}{3c^3}. \tag{1}$$

Here d is the matrix element of the dipole moment of the resonant transition, N is the number of dipoles in a small volume $V\ll\lambda^3$ of the material. Here we have not taken into account the precession of the dipoles in the local dipole fields $E_d\sim Nd/V$, even though

$$E_d/E_r\simeq\lambda^3/V. \tag{2}$$

This neglect of the dipole-dipole interactions in the theory of superradiance is in no way justified except by the difficulties of taking into account the contribution of this interaction to the collective coherent motions of the dipoles.

To describe the motions of the dipole densities $\mathscr{P}(t)$ observed in optics, as in other kinetic problems,[4] it is necessary first to obtain the kinetic equations. These equations can be derived from simple physical arguments, as was done by Bloch[5] in obtaining the equations for the magnetization. Another way to derive the kinetic equations is directly from the equations for the density matrix of the system of coupled dipole-dipole interactions of the quantum magnetic moments, as was proposed in Refs. 6 and 7.

The Bloch equations were later carried over with great success into optics[8] to describe the motion of the densities of quantum optical dipoles. At present, however, in optics[8] and the study of magnetic resonances it has been found[7] by analyzing a large quantity of experimental data on the decay of free polarization, echo effects, saturation, etc., that the Bloch equation provides a good description only of the dephasing of ensembles of dipoles precessing in external fields and local dipole fields, the latter various in magnitude and direction. Therefore these equations cannot describe the collective coherent motions of the dipoles in a superradiant field.

In this regard note that recently, by treating the elementary dynamic process in the form of precession of the dipoles in external and local dipole fields, kinetic equations were obtained[9,10] which are more general than the Bloch equations, for the partial densities $\sigma_\alpha(h,t)$, $\alpha=x,y,z$ of the quantum dipoles. Earlier, analogous equations for the magnetic dipole densities were approximately derived in Ref. 11 from the equations for the density matrix of the system of coupled dipole-dipole interactions of the magnetic moments. Here $\sigma_\alpha(h,t)$ is the density of dipoles found at time t in the longitudinal local dipole field h. Below for brevity we will call such densities layer polarizations. Recall that in a magnetic resonance and in optics we call those components of a vector longitudinal that are parallel to a strong constant magnetic field $\mathbf{H}_0$ or to the direction of propagation of electromagnetic waves. It should also be emphasized that in optics a longitudinal local field is not a true field, but an auxiliary field, convenient for describing optical phenomena.

With the help of the kinetic equations for the layer polarizations collective coherent oscillations of dipoles were first described in Ref. 9, where these oscillations have frequency

$$\omega_{\text{osc}}=\sqrt{a\omega_d\omega_a}\gg\omega_d, \tag{3}$$

where $\omega_d=d^2N/\hbar V$, ω_a is the resonant frequency of an atom, and $a\sim1$. Such oscillations, as was shown in Ref. 9, give rise to radiation peaks at frequencies $\omega_a\pm\omega_{\text{osc}}$ close to ω_a, observable in strongly optically-excited gases in the experimentally well-studied effect of spectral condensation of radiation.[12,13]

In order to use the kinetic equations for the layer polarizations to explain the motion of the dipoles under superradiant conditions, we added the contributions from the dipole precession in the radiation reaction field which are nonlinear in the dipole densities to the rates of variation of the layer polarizations.[9] As a result, solutions of the kinetic equations

1063-7761/95/061024-05$10.00

were obtained for the first time in Ref. 9 for small times $t<\tau$, T/N for $\omega_{\rm osc}t \gg 1$ in the form of a total complex dipole density [$g(h)$ is defined below in Eq. (8)]:

$$f_0(t)=\int dh g(h)[\sigma_x(h,t)+i\sigma_y(h,t)]=\sigma_{x0}(t)+i\sigma_{y0}(t). \tag{4}$$

(Here, T is the spontaneous transition time of the atom, τ is the characteristic time of variation of the local field h under the influence of the thermal motion of the atoms.) Thus, it was shown that dipole-dipole interactions are manifested in a superradiant field in the oscillations of the dipoles with frequency $\omega_{\rm osc}$ large in comparison with the characteristic frequency of the local field ω_d.

The goal of the present paper is to use solutions of the kinetic equation for the layer polarization to explain how the dipole-dipole interactions in strongly optically-excited materials lead to dephasing and to coherent collective oscillations of the dipoles with frequency $\omega_{\rm osc}$, and also, taking these interactions into account, to trace out how the process of collective spontaneous radiation proceeds at arbitrary times all the way to complete de-excitation of the material.

2. KINETIC EQUATIONS FOR SUPERRADIATION WITH ACCOUNT OF THE DIPOLE-DIPOLE INTERACTIONS

The kinetic equations for the layer polarization with account of the interactions of the dipoles with each other and with the radiation reaction field were obtained in Ref. 9 for the concentrated model of Dicke in the form (the fields h below are given in frequency units)

$$\frac{\partial F(h,t)}{\partial t}=ihF(h,t)+i[-hF(h,t)+hF_0+F_1]+\frac{F_0-F(h,t)}{\tau}+\frac{NF_0\sigma_z(h,t)}{2T}, \tag{5}$$

$$\frac{\partial \sigma_z(h,t)}{\partial t}=-\frac{N}{2T}[F_0^*F(h,t)+F_0F^*(h,t)], \tag{6}$$

where

$$F_{0,1}(t)=\int dh g(h) h^{0,1}F(h,t), \quad F(h,t)=\sigma_x(h,t)+i\sigma_y(h,t), \tag{7}$$

$$g(h)=\frac{1}{\pi}\frac{\gamma}{h^2+\sigma^2} \tag{8}$$

is the distribution function of the longitudinal local dipole fields,[7]

$$\gamma=3.8d^2N/\hbar V, \quad 1/\tau=\bar{v}(N/V)^{1/3}, \tag{9}$$

$\bar{v}$ is the thermal velocity of an atom, and its spontaneous transition rate is

$$\frac{1}{T}=\frac{2}{3}\frac{\omega_a^3d^2}{\hbar c^3}. \tag{10}$$

The contributions to the rate of variations of the layer polarizations which are proportional to N/T are associated with the radiation reaction field, and those proportional to $1/\tau$ are associated with random variations of the fields h due to the thermal motions of the atoms.

The first term on the right-hand side of Eq. (5), thanks to the choice of layer polarizations as the variables, exactly describes the contribution to the rate of variation of the layer polarizations from the precession of the dipoles in the field h. This contribution is characterized by the first of the two terms in the Hamiltonian of the secular part of the dipole-dipole interactions, which can be represented in the form[7,9]

$$H_d^0=-\sum_{i>k} b_{ik}\sigma_z^{(i)}\sigma_z^{(k)}+\sum_{i>k} b_{ik}\sigma^{(i)}\sigma^{(k)}\equiv H_{zz}+H_{is}. \tag{11}$$

Here $\sigma_\alpha^{(i)}$ are the operators of the components of the atomic pseudodipoles, identical with the Pauli matrices,

$$b_{ik}=d^2(3\cos^2\theta_{ik}-1)/2\hbar r_{ik}^2, \tag{12}$$

and r_{ik} and θ_{ik} are the magnitude and polar angle of the vector joining the ith and kth dipoles. H_{is} in Eq. (11) is the isotropic part of the Hamiltonian. This part simultaneously describes the precession of the dipoles in isotropic, local dipole fields and the exchange of polarizations between the pseudospins. Its contribution to the rate of variation of the layer polarizations is easily determined from five simple properties which it possesses. Since H_{is} corresponds to precession of the dipoles in the local fields, its contribution should be linear, first, in the field h, and second, in the layer polarizations $F(h,t)$. Since H_{is} contains H_{zz} within itself, its contribution should also contain a term proportional to ih-$F(h,t)$. Finally, since the contribution of H_{is} describes the exchange of polarizations between the layer h and all the remaining layers, it should vanish when the polarizations are equal and upon integration over all the layers. The contribution to the rate of variation of the layer polarizations corresponding to all these conditions can be represented in the form

$$\left.\frac{\partial F}{\partial t}\right|_{is}=i[-hF(h,t)+hF_0+F_1]. \tag{13}$$

It should be emphasized that it was specifically the added term H_d^0 to the well-known two-level Hamiltonian of noninteracting atoms[8] that made it possible in Ref. 9 to take multiparticle effects into account, which, in particular, are manifested under superradiant conditions in the coherent collective oscillations of the dipoles. At the same time, on the basis of the commonly used two-level model, which does not take H_d^0 into account, the Bloch equations were successfully used in Ref. 8 within the framework of resonance optics to describe just those phenomena in which the dipole-dipole interactions cause only a dephasing of the dipoles.

Let us move on now to analyze, with the help of kinetic equations (5) and (6), the contribution of the dipole-dipole interactions to the motion of the optical dipole densities in a gas strongly excited by a resonant field. We will take it to be enclosed in a volume with linear dimensions $1 \ll \lambda$, which will allow us to analyze the motions of the dipoles without accounting for wave effects in terms of the concentrated Dicke model.

3. SOLUTIONS OF THE KINETIC EQUATIONS FOR THE LAYER POLARIZATIONS

One can convince oneself by direct substitution in the kinetic equations (5) and (6) that these equations are satisfied by the solutions

$$F_{1,2}(h,t)=f_0(t)\left(1-\frac{ihk_{1,2}}{\langle h^2\rangle}\right)\exp(k_{1,2}t), \qquad (14)$$

where

$$\langle h^2\rangle=\int dhh^2g(h)=\mathrm{Tr}(b_{ik}\sigma_z^{(k)})^2/\mathrm{Tr}\ 1, \qquad (15)$$

and

$$\sigma_z^{1,2}(h,t)=\sigma_{z0}(t)\left(1-\frac{ihk_{1,2}}{\langle h^2\rangle}\right), \qquad (16)$$

where for $\langle h^2\rangle > 1/(2\tau)^2$ we have

$$k_{1,2}=-\frac{1}{2\tau}\pm i\sqrt{\langle h^2\rangle-\left(\frac{1}{2\tau}\right)^2}, \qquad (17)$$

and $f_0(t)$ and $\sigma_{z_0}(t)$ are defined by the system of equations

$$\frac{\partial f_0}{\partial t}=\frac{Nf_0\sigma_{z0}}{2T}, \qquad (18)$$

$$\frac{\partial\sigma_{z0}}{\partial t}=-\frac{2N}{T}f_0f_0^*\exp(k_1+k_2{}^*)t. \qquad (19)$$

It should be emphasized that relation (15) is approximately applicable for the case under consideration of strongly optically excited matter characterized by roughly identical populations of the upper and lower resonant levels.

Under the condition

$$T/N\tau \ll 1, \qquad (20)$$

which for rates of change of the atomic environment $1/\tau=n_0^{1/3}v$, where n_0 is the atomic density, is well satisfied for $n_0<10^{15}\ \mathrm{cm}^{-3}$, it is possible to neglect terms in Eq. (16) proportional to $1/\tau$, which leads to $k_{1,2}=\pm i\sqrt{\langle h^2\rangle}$.

Equations (18) and (19) then completely coincide with the kinetic equations derived in Ref. 13 for the concentrated Dicke model,[1] which do not take dipole-dipole interactions into account. These equations describe superradiance as the result of rotation under the influence of spontaneous transitions of a gigantic pseudodipole proportional to N

$$J=\sqrt{\sigma_{z0}^2+4|f_0|^2}. \qquad (21)$$

The appearance in solutions (14) of the decaying periodic factors which are proportional to $\exp(\pm i\omega_{\mathrm{osc}}-1/2\tau)t$ and are absent in Dicke's theory[1] implies that under the influence of the dipole-dipole interactions spatially homogeneous, coherent oscillations of the dipoles develop with large frequency (3). It is specifically these factors that distinguish the solutions of kinetic equations (5) and (6) from the solutions of Dicke's kinetic equations.[1]

To account for the appearance of a large frequency, we will make use of Eq. (5), discarding the terms proportional to $1/\tau$ and N/T, which are small in comparison with the frequency (3). Multiplying Eq. (5) by $g(h)$ and $hg(h)$ and integrating with respect to h, we find

$$\frac{\partial F_0}{\partial t}=if_1, \qquad (22)$$

$$\frac{\partial F_1}{\partial t}=i\langle h^2\rangle f_0. \qquad (23)$$

It is clear from these equations that an effective longitudinal field enters into the kinetics of strongly optically-excited dipoles not through the average dipole field ω_d, but through the root-mean-square field (3), which is much larger in gases. The solution of Eqs. (22) and (23) has the simple form

$$F_0=C\exp(kt), \quad F_1=-ikC\exp(kt), \quad k=\pm i\sqrt{\langle h^2\rangle}. \qquad (24)$$

It is clear from these solutions that under the influence of dipole-dipole interactions in strongly optically-excited matter, spatially homogeneous oscillations of the dipoles are established, leading to superradiation of a macroscopic sample.

In order to answer the question of the appearance of identical orientations of the dipoles over an entire macroscopic sample, it is important to turn our attention to the work of Rehler and Eberly.[14] Assuming simultaneous alignment of all the atomic dipoles of an extended sample and numerically calculating the intensity of the coherent radiation emitted by all the atoms of the sample, they succeeded in determining for some simple sample shapes the radiation reaction field acting on the atoms at the time t and in obtaining a kinetic equation for the optical dipole densities which provides a good description of the superradiant pulses observed in extended samples.[8,14]

In this equation the shape of the extended sample is taken into account by a constant μ, determined by numerical calculation.[8] The equation itself turns out to coincide with the Dicke equation[1] for $\mu=1$. It has been used successfully to describe superradiant pulses in extended samples having the same shape as in the concentrated model.

In connection with the importance in the superradiance phenomenon of the mechanism by which the dipoles become coherent, we will consider it in more detail in the following section.

4. MECHANISM FOR COLLECTIVE COHERENT OSCILLATIONS OF THE DIPOLES

We will now show that coherent oscillations of dipoles can also be considered as periodic echo chains. It is interesting to note that such oscillations were first detected[7] back in the 1950s in the context of nuclear magnetic resonance in the decays of the free polarization of crystalline materials.

The initial segment of these decays is well described by a Gaussian curve which characterizes the dephasing of the magnetic dipoles in solid materials with high density of magnetic dipoles.[7] Then, after a significant (roughly a factor of ten) decay of the magnetization over five periods, damped harmonic oscillations of the coupled dipole-dipole interactions of the magnetic dipoles were observed.[15]

These oscillations are echo chains. Such an interpretation is convincingly confirmed by the close coincidence of the observed decays[15] with those obtained by molecular dynamics calculations in Ref. 16, which take account of the precession of the dipoles in the local dipole fields.

Obviously, precession of the dipoles in the local fields can cause only phasing and dephasing of the dipoles. These processes are manifested in the calculated decays as periodic sequences of echos.

Because the precession of magnetic and optical dipoles is identical in magnetic or electric local dipole fields, it is obvious that coherent oscillations of dipoles in optics can also be considered as periodic echo sequences.

It is interesting to note that in the case $\omega_{\rm osc} \gg \bar{\delta}$, where $\bar{\delta}$ is the mean value of the Doppler shift, the linearly polarized oscillations of the dipoles suppress the Doppler shifts. In this case the influence of the Doppler shifts on the oscillations can be estimated from perturbation theory.

For this purpose it is enough to note that during the first and second half-periods of an oscillation with period $T_0 = 2\pi/\omega_{\rm osc}$ the Doppler shift $\bar{\delta}$ will create polarization increments $F_0 \bar{\delta} T_0$, identical in magnitude, but of opposite sign. As a result, the contribution of the Doppler shift to the dephasing of the dipoles decreases to $\bar{\delta}^2/\omega_{\rm osc}$.

In the case of circularly polarized dipole oscillations the Doppler shifts are not suppressed. Therefore, in strongly optically-excited gases the appearance of linearly polarized coherent oscillations is more probable.

5. CONCLUSION

In conclusion we may note that the kinetic equations for layer polarizations have been successfully applied in work on nuclear magnetic resonances[10,11] and electron paramagnetic resonances[10] to analyze a number of other problems in which the dipole kinetics could not be described by the Bloch equations.

The kinetic equations for the layer polarizations, for example, have been used successfully to describe nuclear magnetic resonance spectra in quantum crystals of ^{3}He in solutions with arbitrary radical concentration.[10] In crystals exchange interactions characterize the influence of tunnel motions of the atoms on the precession of the dipoles in the local dipole fields, but in solutions of radicals they cause polarization exchange between the electron spins.

We will use Eq. (13) to account for the contribution of these isotropic interactions to the kinetic equation for the layer polarizations, replacing h by $h_{\rm ex}$ (Ref. 10):

$$\left.\frac{\partial F}{\partial t}\right|_{\rm ex} \simeq i[-h_{\rm ex}F(h_{\rm ex},h,t)+h_{\rm ex}F_{0\,\rm ex}+F_{1\,\rm ex}], \qquad (25)$$

where

$$F_{0,1\,\rm ex} = \int dh g(h) \int dh_{\rm ex} g(h_{\rm ex}) h_{\rm ex}^{0,1} F(h_{\rm ex},h,t).$$

With the help of kinetic equations (5) and (6) for $N/T=0$ with the added contributions (25), it was possible in Ref. 10 to achieve a good description of the spectra observed in quantum crystals and radical solutions, with the exception of the high-frequency asymptotic behavior. In Ref. 10 it was shown that to describe this asymptotic behavior it is necessary to take account of memory effects in the spectral diffusion, i.e., in the term in the kinetic equations proportional to $1/\tau$.

Memory effects have no influence on the contributions of the dipole precession to the rate of variation of the layer polarizations because the precession rates are determined by the spin and the local field taken at the same instant of time.

It is important to emphasize here that in addition to the results considered above, the kinetic equations for the layer polarizations were used in Ref. 9 to obtain the Bloch equations for the optical dipole densities in gases and liquids, and in Ref. 17, to obtain the kinetic equations for the layer polarizations describing saturation effects in solids. For $\omega_1 \ll \omega_d$ (ω_1 is the amplitude of the variable magnetic field in frequency units), the latter reduce to the kinetic equations obtained in Ref. 18 for the magnetization and inverse temperature of the reservoir of the dipole-dipole interactions, which accurately describe the spectra observed[19] in nuclear magnetic resonances.

Summarizing, we can say that the kinetic equations for the layer polarizations obtained here have made it possible for the first time to describe on a single basis a wide range of resonance phenomena observed in optics, nuclear magnetic resonance, and electron paramagnetic resonance.

In conclusion it is interesting to pursue the following analogy. In a way similar to that in which simple physical considerations in quantum statistical physics[20] lead to the establishment of the form of the equilibrium density matrix

$$\rho_0 \propto \exp(\hat{H}/kT) \qquad (26)$$

($\hat{H}$ is the Hamiltonian of the material), which determines all the thermodynamic properties of the material, five simple physical considerations (see above) in kinetics lead to equations for the layer polarizations which well describe various motions of magnetic and optical dipole densities.

This work was carried out with the financial support of the Russian Fund for Fundamental Research (Project No. 93-02-15076).

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Translated by Paul F. Schippnick