Superradiance by ferroelectrics in cavity resonators

V I Yukalov

Bogolubov Laboratory of Theoretical Physics, Joint Institute for Nuclear Research, Dubna 141980, Russia
Instituto de Física de Sã o Carlos, Universidade de Sã o Paulo, CP 369, Sã o Carlos 13560-970, Sã o Paulo, Brazil

E-mail: yukalov@theor.jinr.ru

Received 23 May 2019
Accepted for publication 19 August 2019
Published 24 October 2019

Abstract
A theory is presented showing that, under appropriate conditions, a ferroelectric in a cavity resonator can emit superradiant pulses. Initially, the ferroelectric has to be prepared in a nonequilibrium state from which it relaxes emitting a coherent pulse in the infrared region. Polarization dipolar waves play the role of the triggering mechanism initiating the beginning of the process.

Keywords: superradiance, cavity resonators, Purcell effect, scale separation

1. Introduction

It is known that an ensemble of atoms or molecules can emit superradiant electromagnetic pulses due to atomic interactions through the common radiation field [1–11]. This type of coherent emission, where the process starts with spontaneous atomic radiation, was first described by Dicke [12] and is often called Dicke superradiance [13]. The same mathematical description as for atomic superradiance is valid for exciton superradiance [14–33], including superradiance from quantum dots and wells [34–40] and for polariton superradiance [41–46].

There also exists superradiance of non-Dicke type, when radiating dipoles cannot be collectivized through the common radiation field, but can become correlated by a resonator feedback field, which is called the Purcell effect [47]. This type of superradiance, due to the Purcell effect, is typical of superradiance produced by spin and quasi-spin assemblies composed, e.g. of polarized nuclei, magnetic nanomolecules, magnetic nanoclusters, dipolar atoms and molecules, spinor atoms and molecules, and ferromagnets. Numerous citations can be found in review articles [48, 49] and recent papers [50–53].

In the present paper, we show that ferroelectrics can also produce superradiance. Similarly to spin systems, ferroelectric superradiance is possible only when the ferroelectric sample is connected to a resonator, so that the ferroelectric superradiance is also of non-Dicke type. A kind of luminescence can be emitted by polarized samples not coupled to a resonator [54], but this radiation is not coherent. The possibility of coherent radiation by ferroelectrics was mentioned in [55], but the full theory was not given. Here the theory of ferroelectric superradiance is developed with all details that are necessary for proving the feasibility of this phenomenon.

Throughout the paper, the system of units is used, where the Planck and Boltzmann constants are set to one.

2. Ferroelectric model

Let us consider a ferroelectric of the order–disorder type, whose Hamiltonian is [56]

\[ \hat{H} = -\Omega \sum_j S_j^x - \frac{1}{2} \sum_{i \neq j} J_{ij} S_i^x S_j^x - \sum_j E_{\text{ext}} \cdot \hat{P}_j. \]  (1)

Here \( \Omega \) is the tunneling frequency; \( J_{ij} \) is the interaction potential between two lattice sites enumerated by the indices \( i,j = 1,2,\ldots,N \), where the self-action is excluded through the condition \( J_{jj} = 0 \); the site polarization operators \( \hat{P}_j = d_0 S_j \)

are expressed through the electric dipole \( d_0 \) and the spin one half (quasi-spin) operators with the commutation relations

\[ [S_i^x, S_j^x] = i\delta_{ij} S_j^y, \quad [S_i^y, S_j^y] = i\delta_{ij} S_j^x, \quad [S_i^z, S_j^z] = i\delta_{ij} S_j^y. \]
The total electric field
\[ \mathbf{E}_{tot} = E_0 \mathbf{e}_x + E_0 \mathbf{e}_z \]  
consists of a resonator feedback field \( E_0 \) and an external field \( E_0 \).

The Heisenberg equations of motion for the spin operators yield
\[ \frac{dS^i_j}{dt} = \left( \sum_j J_{ij} S^j_i + d_0 E_0 \right) S^i_j , \]
\[ \frac{dS^y_j}{dt} = -\left( \sum_j J_{ij} S^j_i + d_0 E_0 \right) S^y_j + (\Omega + d_0 E) S^y_j , \]
\[ \frac{dS^z_j}{dt} = - (\Omega + d_0 E) S^z_j . \]  
(4)

Our aim is to find the temporal behavior of the averages
\[ \langle S^i \rangle = \frac{2}{N} \sum_j \langle S^i_j \rangle . \]  
(5)

For the average interaction potential, we shall use the notation
\[ J \equiv \frac{1}{N} \sum_{i \neq j} J_{ij} . \]  
(6)

To take into account spin relaxation, we employ the method of local fields [57, 58], where particle interactions are considered as acting in the local field formed by other particles so that there appears the attenuation of spin motion, which forces the spin variables at each moment of time to relax to their local equilibrium values. The latter are defined as having the form of the equilibrium averages
\[ \zeta_0 \equiv \frac{2}{N} \sum_j \langle S^i_j \rangle_{eq} , \]  
(7)

but expressed through the variables (5) taken at the given moment of time.

The analysis of the evolution equations can be done by invoking a scale separation approach [48–53, 55]. The pair spin correlators are decoupled by means of the stochastic mean-field approximation [48–50] giving
\[ \langle S^i_j S^j_i \rangle = \langle S^i_j \rangle \langle S^j_i \rangle + \langle S^i_j \rangle \delta S^i_j + \langle S^j_i \rangle \delta S^j_i , \]  
(8)

where \( i \neq j \) and \( \delta S^i_j \) are treated as stochastic variables with zero mean,
\[ \langle \delta S^i_j \rangle = 0 . \]  
(9)

This approximation makes it possible to take into account spin correlations caused by spin waves, which is principally important at the initial stage of spin relaxation.

Averaging equation (4) with decoupling (8), we meet the stochastic variable
\[ \xi_0^i \equiv \sum_j J_{ij} \delta S^i_j . \]  
(10)

Under the averaging over the sample, we use the mean-field-type approximation
\[ \frac{1}{N} \sum_j \xi_0^i \langle \xi_j^i \rangle = \xi_0 \frac{1}{N} \sum_j \langle \xi_j^i \rangle , \]
\[ \frac{1}{N} \sum_j \xi_0^\alpha \langle \xi_j^\alpha \rangle = \xi_\alpha \frac{1}{N} \sum_j \langle \xi_j^\alpha \rangle , \]  
(11)
in which \( \xi_0 \) and \( \xi_\alpha \) are stochastic variables. Thus we come to the equations
\[ \frac{ds_i}{dt} = \left( \frac{J}{2} s_x + d_0 E_0 + \xi_0 \right) s_y + \xi_\alpha s_x - \gamma_2 (s_x - \xi_\alpha) , \]
\[ \frac{ds_y}{dt} = - \left( \frac{J}{2} s_x + d_0 E_0 + \xi_0 \right) s_y + (\Omega + d_0 E) s_x - \gamma_2 (s_x - \xi_\alpha) , \]
\[ \frac{ds_z}{dt} = - (\Omega + d_0 E) s_y - \gamma_1 (s_x - \xi_\alpha) . \]  
(12)

It is also useful to consider the ladder operator
\[ S^\pm_j \equiv S^j_x \pm i S^j_y , \]
whose average yields the variable
\[ u \equiv \frac{2}{N} \sum_j \langle S^\pm_j \rangle = s_x - i s_y . \]  
(13)

Then, denoting the quantities
\[ \xi \equiv \xi_\alpha - i \xi_0 , \quad \zeta \equiv \xi_\alpha - i \xi_\alpha , \]  
(14)

we obtain the equation
\[ \frac{du}{dt} = i \left( \frac{J}{2} s_x + d_0 E_0 + \xi_0 \right) u - i (\Omega + d_0 E - \xi) s_y - \gamma_2 (u - \zeta) . \]  
(15)

The stochastic variables are assumed to satisfy the correlation conditions
\[ \langle \langle \xi_0 (t) \rangle \rangle = \langle \langle \xi (t) \rangle \rangle = 0 , \]
\[ \langle \langle \xi_0 (t) \xi_0 (t') \rangle \rangle = 0 , \]
\[ \langle \langle \xi_0 (t) \xi_0 (t') \rangle \rangle = \langle \langle \xi (t) \xi (t') \rangle \rangle = 2 \gamma_3 \delta (t - t') , \]  
(16)

where stochastic averaging is implied.

3. Local equilibrium state

In order to define the local equilibrium (quasi-equilibrium) values (7), it is necessary to consider a Hamiltonian without the resonator feedback field,
\[ \hat{H}_{eq} = - \Omega \sum_j S^z_j - \frac{1}{2} \sum_{i \neq j} J_{ij} S^i_j S^j_i - d_0 E_0 \sum_j S^y_j . \]  
(17)

Resorting to the mean-field approximation gives...
\[ H_{eq} = -\Omega \sum_j S_j^t - J_{\text{eff}} \sum_j S_j^d, \]  
(18)  
where
\[ J_{\text{eff}} \equiv J\langle S_j^d \rangle + d_0 E_0. \]  
(19)  
Calculating statistical averages of spin components at temperature \( T \), we employ the notation
\[ \Omega_{\text{eff}} \equiv \sqrt{\Omega^2 + J_{\text{eff}}^2}. \]  
(20)  
Thus we obtain
\[ \langle S_j^t \rangle_{eq} = \frac{\Omega}{2\Omega_{\text{eff}}} \tanh \left( \frac{\Omega_{\text{eff}}}{2T} \right), \quad \langle S_j^d \rangle_{eq} = 0, \]  
(21)  
The external electric field is directed down, so that
\[ \omega_0 \equiv -d_0 E_0 > 0. \]  
(22)  
Replacing in equation (21) the average spins by their time-dependent values at zero temperature yields
\[ \zeta_\alpha = \frac{\Omega}{\Omega_{\text{eff}}}, \quad \zeta_\nu = \frac{2\Omega}{4\Omega^2 + (J_\nu - 2\omega_0)^2}^{1/2}, \quad \zeta_\nu = 0, \]  
(23)  
Since \( j_\nu = j_\nu(t) \) is a function of time, the local equilibrium quantities also depend on time,
\[ \zeta_\alpha = \zeta_\alpha(t), \quad \zeta = \zeta(t) = \zeta_\nu(t). \]  
(24)  

4. Resonator field

To derive an equation for the resonator feedback field, it is possible to follow the general methods for treating cavity resonators [59, 60]. Here we keep in mind a cylindrical resonator cavity of radius \( R \), length \( L \), and volume \( V_{\text{res}} = \pi R^2 L \). The axis of the resonator is along the \( x \)-axis. The Gaussian system of units will be used.

Electromagnetic fields inside the resonator obey the Maxwell equations
\[ \nabla \cdot \mathbf{D} = 4\pi \rho, \quad \nabla \cdot \mathbf{B} = 0 \]
\[ \nabla \times \mathbf{E} = -\frac{1}{c^2} \frac{\partial \mathbf{B}}{\partial t}, \quad \nabla \times \mathbf{H} = \frac{4\pi}{c} \mathbf{J} + \frac{1}{c} \frac{\partial \mathbf{D}}{\partial t}, \]  
(24)  
in which
\[ \mathbf{D} = \mathbf{E} + 4\pi \mathbf{P}, \quad \mathbf{B} = \mathbf{H} + 4\pi \mathbf{M}. \]  
(25)  
We consider the case of a ferroelectric inserted into the resonator, where there are no free charges and magnetic inclusions, so that the material equations are
\[ \rho = 0, \quad \mathbf{M} = 0, \quad \mathbf{J} = \sigma \mathbf{E}. \]  
(26)  
From the Maxwell equations, we find
\[ \nabla^2 \mathbf{E} = \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} - \frac{4\pi \sigma}{c^2} \frac{\partial \mathbf{E}}{\partial t} = \frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}}{\partial t^2} - 4\pi \nabla \left( \nabla \cdot \mathbf{P} \right). \]  
(27)  
The polarization vector can be represented as
\[ \mathbf{P} = \frac{1}{V} \sum_j \langle \mathbf{P}_j \rangle = \frac{d_0}{V} \sum_j \langle S_j \rangle. \]  
(28)  
We look for the solution of equation (27) in the form
\[ \mathbf{E}(\mathbf{r}, t) = \mathbf{e}(\mathbf{r}) \mathbf{E}(t), \]  
(29)  
in which \( \mathbf{e}(\mathbf{r}) \) describes the normal resonator modes satisfying the Helmholtz equation
\[ \left( \nabla^2 + \frac{\omega^2}{c^2} \right) \mathbf{e}(\mathbf{r}) = 0 \]  
(30)  
and being normalized to the resonator volume,
\[ \int | \mathbf{e}(\mathbf{r}) |^2 d\mathbf{r} = V_{\text{res}}. \]  
(31)  
Here \( \omega \) is the resonator natural frequency. Since we have chosen the resonator axis along the \( x \)-axis, we take the Laplacian in the form
\[ \nabla^2 = \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} + \frac{1}{r^2} \frac{\partial^2}{\partial \phi^2} + \frac{\partial^2}{\partial z^2}. \]  
(32)  
The solutions to the Helmholtz equation are termed TM_{\nu m} modes. We are interested in the solution whose electric field would be along the resonator axis, such that
\[ e_\nu(\mathbf{r}) \neq 0, \quad e_\lambda(\mathbf{r}) = 0, \quad e_\nu(\mathbf{r}) = 0, \]  
(33)  
except the boundary of the resonator, where
\[ e_\nu(\mathbf{r}) |_{r=R} = 0. \]  
(34)  
The corresponding solution is given by the TM_{010} fundamental mode
\[ e_\nu(\mathbf{r}) = C_0 J_0 \left( \frac{\omega}{c} r \right), \]  
(35)  
where \( J_0 \) is the Bessel function of the first kind. The boundary condition (33), defining the first zero of the Bessel function,
\[ J_0 \left( \frac{\omega}{c} R \right) = 0, \]  
(36)  
prescribes the resonator natural frequency
\[ \omega = 2.4048 \frac{c}{R}. \]  
(37)  
The coefficient \( C_0 \) has to be found from the normalization condition (31). To this end, we use the integral
\[ \int_0^R j_\nu'(kr) rdr = \frac{R^2}{2} \left| j_\nu'(kr) \right|^2 + \frac{1}{2} \left( R^2 - \frac{\nu^2}{k^2} \right) J_\nu^2(kR), \]  
(38)  
in which
\[ J_\nu'(kr) = \frac{\nu}{kR} J_\nu(kR) - J_{\nu+1}(kR). \]  
(39)
Keeping in mind the boundary condition (33), we have
\[ J'_0(kR) = -J_1(kR) \quad \left( k = \omega / c \right). \]
Then the normalization condition (31) results in the integral
\[ \int | e_i(\mathbf{r})|^2 \, d\mathbf{r} = C_0^2 \frac{2\pi}{k} J_0(kr) r \, dr = C_0^2 V_{\text{res}} J_1(kr) = V_{\text{res}}. \]
where \( k = \omega / c \), which gives
\[ C_0 = J_1^{-1} \left( \frac{\omega}{c} R \right). \]
Taking into account the natural frequency (36) and the value
\[ J_1(2.04048) = 0.519153, \]
we get
\[ C_0 = 1.926214. \quad (37) \]
Substituting form (29) into equation (27), multiplying it by
\[ e(\mathbf{r}) = C_0 j_0 \left( \frac{\omega}{c} r \right) e_i, \quad (38) \]
and integrating over space yields
\[ \frac{\partial^2 E}{\partial t^2} + 4\pi \sigma \frac{\partial E}{\partial t} + \omega^2 E = -4\pi \frac{\partial^2 P_s}{\partial t^2} \int e(\mathbf{r}) \, d\mathbf{r}, \quad (39) \]
where the filling factor is
\[ \eta_f \equiv \frac{1}{V_{\text{res}}} \int e_i(\mathbf{r}) \, d\mathbf{r}. \quad (40) \]
Introducing the resonator attenuation
\[ \gamma \equiv 2\pi \sigma \quad (41) \]
results in the equation
\[ \frac{\partial^2 E}{\partial t^2} + 2\gamma \frac{\partial E}{\partial t} + \omega^2 E = -4\pi \eta_f \frac{\partial^2 P_s}{\partial t^2}, \quad (42) \]
in which
\[ P_s = \frac{d_0}{V} \sum_{j} \langle S^y_j \rangle. \quad (43) \]
Because of the integral
\[ \int_0^R J_0(kr) r \, dr = \frac{R}{k} J_1(kR), \]
we get
\[ \int e_i(\mathbf{r}) \, d\mathbf{r} = 0.83167 V_{\text{res}}, \]
which gives
\[ \eta_f = 0.83167. \quad (44) \]
Polarization (43) can be written as
\[ P_s = \frac{1}{2} \rho d_0 s_x \left( \rho \equiv \frac{N}{V} \right), \quad (45) \]
Under the initial conditions
\[ E(0) = 0, \quad \frac{\partial E}{\partial t} \bigg|_{t=0} = 0, \quad \frac{\partial P_s}{\partial t} \bigg|_{t=0} = 0, \quad (46) \]
the resonator equation (42) can be rewritten as
\[ \frac{dE}{dt} + 2\gamma E + \omega^2 \int E(f) \, df = -4\pi \eta_f \frac{dP_s}{dt}. \quad (47) \]
This equation has the same form as the Kirchhoff equation for a resonant electric circuit with a magnetic sample inside it. Therefore the consideration of the dipole dynamics in a ferroelectric can be done similarly to the study of spin dynamics in magnets [48–53].

5. Evolution equations
The quantity
\[ \omega_e \equiv \omega_0 - \frac{J}{2} s_z \quad (48) \]
plays the role of an effective rotation frequency. Using the equality
\[ s_i = -\frac{i}{2} (u^* - u), \]
equation (12) can be represented as
\[ \frac{ds_x}{dt} = - (\omega_e - \xi_0)s_x + \xi_s s_z - \gamma_2 s_x - \zeta_s, \quad \frac{ds_y}{dt} = (\omega_e - \xi_0)s_x + (\Omega + d_0 E - \xi_s) s_z - \gamma_2 s_y, \quad \frac{ds_z}{dt} = \frac{i}{2} (\Omega + d_0 E)(u^* - u) - \gamma_1 (s_x - \zeta_s). \quad (49) \]
Equation (15) takes the form
\[ \frac{d\rho}{dt} = -i(\omega_e - \xi_0 - t\gamma_2)u - i(\Omega + d_0 E - \xi_s)s_z + \gamma_2 \zeta. \quad (50) \]
In addition, we shall need to consider the temporal behavior of the coherence intensity
\[ w \equiv \frac{1}{N(N-1)} \sum_{ij} \langle S^+_i S^-_j \rangle = |u|^2, \quad (51) \]
whose evolution equation reads as
\[ \frac{dw}{dt} = -2\gamma_2 w - i(\Omega + d_0 E - \xi_s) s_z u^* + i(\Omega + d_0 E - \xi^*) s_z u + \gamma_2 \zeta (u^* + u). \quad (52) \]
We assume that the detuning from the resonance is small,
\[ \frac{\Delta}{\omega} \ll 1 \quad (\Delta \equiv \omega - \omega_0). \quad (53) \]
It is possible to take a sufficiently large external electric field, such that
\[ \left| \frac{J}{\omega_0} \right| \ll 1. \quad (54) \]
All attenuations are supposed to be much smaller than the frequency \( \omega_0 \).
As follows from equation (42), the effective coupling rate between the ferroelectric sample and resonator is

\[
\frac{\gamma_c}{\omega} \ll 1, \quad \frac{\eta}{\omega_0} \ll 1, \quad \frac{\gamma}{\omega_0} \ll 1 \,.
\]  

(55)

which is much smaller than the resonator natural frequency,

\[
\left| \frac{\gamma_c}{\omega} \right| \ll 1 \,.
\]  

(57)

Solving the resonator equation (42) by perturbation theory in powers of the coupling rate in first order yields

\[
d_0 E = i (u X - X^* u^*) \,.
\]  

(58)

with the coupling function

\[
X = \gamma \omega u \left[ 1 - \exp \left\{ -i(\omega - \omega_s) t - \gamma t \right\} \right. \\
\left. + \frac{1 - \exp \left\{ -i(\omega + \omega_s) t - \gamma t \right\}}{\gamma - i(\omega + \omega_s)} \right] \,.
\]  

(59)

If the effective detuning

\[
\Delta_s \equiv \omega - \omega_s = \Delta + \frac{J}{2} s_t
\]  

(60)

is also small, such that

\[
\left| \frac{\Delta_s}{\omega} \right| \ll 1 \,.
\]  

(61)

then in the coupling function (59) it is possible to keep only the resonant part, obtaining

\[
X = \gamma \omega_s \left[ 1 - \exp \left\{ -i \Delta_s t - \gamma t \right\} \right] \frac{1}{\gamma + i \Delta_s} \,.
\]  

(62)

Substituting expression (58) into the evolution equation (50) gives

\[
\frac{du}{dt} = -i \omega_{eff} u + i \xi u + i \xi s_t - i \Omega s_t + \gamma_2 \zeta - X^* u^* s_t \,.
\]  

(63)

where

\[
\omega_{eff} \equiv -i(\gamma_2 - X t) \,.
\]  

(64)

Equation (52) leads to

\[
\frac{dw}{dt} = -2 \gamma_2 (1 - \alpha s_t) w + i (u^* \zeta - \xi^* u) s_t \,.
\]  

+\Omega (u - u^*) \label{eq:65}

\[
+ \Omega (u - u^*) + \gamma_2 \zeta (u - u^*) - [X^*(u^*)^2 + X u^2] s_t \,.
\]

with the notation

\[
\alpha \equiv \frac{1}{2 \gamma_2} (X^* + X) = \frac{\text{Re} X}{\gamma_2} \,.
\]  

(66)

The equation for the polarization becomes

\[
\frac{ds_t}{dt} = -\alpha \gamma_2 w - \gamma_1 (s_t - \zeta) + \frac{i}{2} \Omega (u^* - u) \,.
\]  

(67)

Taking account of the existing small parameters shows that the functional variable \(u\) can be treated as fast, while the variables \(w\) and \(s_t\) are slow. Then averaging techniques are applicable [55, 61]. Solving equation (63) for the fast variable, the slow variables are kept as integrals of motion. This results in the solution

\[
u = u_0 \exp \left\{ -i \omega_{eff} t + i \int_0^t \xi_0 (t') \, dt' \right\} + i \int_0^t [\xi (t') s_t - \Omega s_t - i \gamma_2 \zeta] \exp \left\{ -i \omega_{eff} (t - t') + i \int_r^t \xi_0 (t'') \, dt'' \right\} \, dt' \,.
\]  

(68)

The found fast variable is to be substituted into the equations for the slow variables and their right-hand sides are to be averaged over time and over the stochastic variables. In that way, we obtain the equations for the guiding centers

\[
\frac{dw}{dt} = -2 \gamma_2 (1 - \alpha s_t) w + 2 \gamma_3 s_t^2 \,.
\]  

(69)

These equations are complimented by the initial conditions

\[
w(0) = w_0 \,, \quad s_t(0) = s_0 \,.
\]  

(70)

Assuming that \(\Delta_s\) is much smaller than \(\gamma\) makes it possible to simplify the coupling function (66), getting

\[
\alpha = \frac{g^2}{\gamma^2 + \Delta_s^2} \left( 1 - \frac{J}{2 \gamma_0} s_t \right) (1 - e^{-\gamma t}) \,.
\]  

(71)

where

\[
g \equiv \frac{\gamma \omega_0}{\gamma_2} \,.
\]  

(72)

is a dimensionless coupling parameter. Under this condition, it is convenient to write the coupling function in the form

\[
\alpha = g (1 - A s_t) (1 - e^{-\gamma t}) \,.
\]  

(73)

in which

\[
A \equiv \frac{J}{2 \gamma_0} \,.
\]  

(74)

### 6. Development of coherence

First of all, it is necessary to emphasize that the presence of the resonator is crucial for organizing collective motion of dipoles. Really, the absence of the resonator implies that the coupling function \(\alpha = 0\). Then from equation (69), it is evident that the polarization \(s_t\) slowly relaxes with the relaxation rate \(\gamma_1\), while the coherence intensity \(w\) slowly relaxes with the relaxation rate \(\gamma_2\); provided coherence was imposed through the initial condition. In addition, no coherence appears if \(w_0 = 0\).

The existence of the dynamic attenuation \(\gamma_3\) due to dipolar waves is also of principal importance. If \(\gamma_3 = 0\) and no initial coherence is imposed, so that \(w_0 = 0\), then coherence can
never arise. The presence of $\gamma_3$ initiates the motion of dipoles at the initial stage and leads to the arising coherence.

Let us consider the initial stage of the process, when the coupling function is still small,

$$\alpha \simeq 0 \quad (\gamma t \ll 1).$$

Then equation (69) is

$$\frac{dw}{dt} = -2\gamma_2 w + 2\gamma_3 s_z^2 \quad \frac{ds_z}{dt} = -\gamma_1 (s_z - \zeta).$$

Keeping in mind that $\gamma_1 \ll \gamma$, we see that the polarization practically does not change,

$$s_z \simeq s_0 \quad (\gamma t \ll \gamma t \ll 1),$$

and the coherence intensity is

$$w \simeq \left( w_0 - \frac{\gamma_1}{\gamma_2} s_0^2 \right) e^{-2\gamma_1 t} + \frac{\gamma_3}{\gamma_2} s_0^2.$$

(78)

When either $\gamma_3 = 0$ or $s_0 = 0$, and $w_0 \neq 0$, the function $w$ slowly relaxes with the relaxation rate $\gamma_2$. If $w_0 = 0$ and $s_0 \neq 0$, then

$$w \simeq \frac{\gamma_3}{\gamma_2} s_0^2 \left( 1 - e^{-2\gamma_1 t} \right) \quad (w_0 = 0),$$

(79)

which is rather small, since usually $\gamma_3 \ll \gamma_2$.

There is no essential coherence in the process, when $\alpha s_z \ll 1$. But the coupling function $\alpha$ grows with time, inducing coherence. The incoherent regime lasts until $\alpha s_z$, becomes of the order of unity, after which coherence starts quickly growing. The time of the beginning of the coherent stage can be defined by the equality

$$\alpha s_z = 1 \quad (t = t_{coh}).$$

(80)

which yields

$$t_{coh} = \frac{1}{\gamma} \ln \left[ \frac{g s_0 (1 - A s_0)}{g s_0 (1 - A s_0) - 1} \right].$$

(81)

The strength of the coupling with the resonator depends on the magnitude of the coupling parameter $g$ and on the initial polarization $s_0$. Under strong coupling, the coherence time becomes

$$t_{coh} \simeq \frac{1}{\gamma g s_0 (1 - A s_0) \gamma \gamma_3} \quad (g s_0 \gg 1).$$

(82)

This also shows that coherence can develop in the sample only if the initial polarization corresponds to a nonequilibrium state, when $s_0$ is positive, hence directed against the applied electric field $E_0$. Notice that

$$\gamma t_{coh} \ll 1 \quad (g s_0 \gg 1).$$

(83)

Therefore, if $\gamma_2 \ll \gamma$, then

$$\gamma_2 t_{coh} \ll \gamma t_{coh} \ll 1 \quad (g s_0 \gg 1).$$

(84)

Then the coherence intensity (78) reads as

$$w \simeq w_0 + 2\gamma_3 s_0^2 t \quad (\gamma_2 t \ll 1).$$

(85)

At the coherence time, the variables $w$ and $s$ reach the values

$$w(t_{coh}) \equiv w_{coh}, \quad s(t_{coh}) \equiv s_{coh}.$$

(86)

for which we have

$$w_{coh} = w_0 + 2\gamma_3 s_0^2 t_{coh}, \quad s_{coh} = s_0.$$

(87)

Again, let us stress the necessity for the existence of the resonator that induces coherence in the presence of an initial nonequilibrium polarization and the dynamic attenuation due to dipolar waves.

7. Coherent stage

Essential coherence of dipole motion develops in the sample at large $\gamma t$. Then, assuming that $|A| \ll 1$, one has

$$\alpha \simeq g \quad (|A| \ll 1, \gamma t \gg 1).$$

(88)

Also, taking into account the standard case where

$$\gamma_1 \ll \gamma_2, \quad \gamma_3 \ll \gamma_2,$$

(89)

we get the equations

$$\frac{dw}{dt} = 2\gamma_2 (g s_z - 1) w, \quad \frac{ds_z}{dt} = -g\gamma_2 w.$$

(90)

These equations enjoy exact solutions giving the coherence intensity

$$w = \left( \frac{\gamma_p}{g\gamma_2} \right)^2 \text{sech}^2 \left( \frac{t - t_0}{\tau_p} \right)$$

and polarization

$$s_z = \frac{1}{g} - \frac{\gamma_p}{g\gamma_2} \text{tanh} \left( \frac{t - t_0}{\tau_p} \right).$$

(91)

(92)

Here the quantities $t_0$ and $\gamma_p \equiv 1/\tau_p$ are defined by conditions (86) resulting in the equalities for the delay time

$$t_0 = t_{coh} + \frac{\tau_p}{2} \ln \left| \frac{\gamma_p + \gamma_2}{\gamma_p - \gamma_2} \right|$$

(93)

and pulse time

$$\tau_p \equiv \frac{1}{\gamma_p}, \quad \gamma_p = \frac{\gamma_2}{2} \left[ 1 + \sqrt{1 + 4 \left( \frac{g s_0}{w_{coh}} \right)^2} \right],$$

(94)

where

$$\gamma_2 = (g s_0 - 1) \gamma_2.$$

(95)

Under strong coupling and weak initially imposed coherence, equation (94) simplifies to

$$\gamma_p \simeq g\gamma_2 \sqrt{\frac{s_0^2 + w_{coh}}{g s_0 \gg 1, \frac{w_{coh}}{s_0} \ll 1}}.$$

(96)

At the time $t_0$, the coherence intensity reaches its maximum, where

$$w(t_0) = s_0^2 + w_{coh}, \quad s_z(t_0) = \frac{1}{g}.$$

(97)
After the delay time \( t_0 \), the coherence intensity quickly diminishes,

\[
w \approx 4w(t_0) \exp \left( -\frac{2}{\tau_p} t \right) \quad (t \gg t_0)
\]  

while the polarization reverses and tends to the expression

\[
s_z \approx -s_0 + \frac{2}{g} + 2s_0 \exp \left( -\frac{2}{\tau_p} t \right) \quad (t \approx t_0).
\]  

It is useful to remark that the limit \( -s_0 + 2/g \) is not an equilibrium limit, although the sample stays close to that state for quite a long time, slowly relaxing to an equilibrium value during the time \( T_1 = 1/\gamma_1 \). In that sense, this effect can be termed pre-equilibration.

8. Radiation intensity

The intensity of radiation by dipoles in the direction of \( n \equiv \hat{r}/|\hat{r}| \) at time \( t \) consists of two parts,

\[
I(n,t) = I_{\text{inc}}(n,t) + I_{\text{coh}}(n,t),
\]

incoherent radiation intensity

\[
I_{\text{inc}}(n,t) = 2\omega_0\gamma_0 \sum_j \varphi(n) \langle S_j^+(t)S_j^-(t) \rangle,
\]

and coherent radiation intensity

\[
I_{\text{coh}}(n,t) = 2\omega_0\gamma_0 \sum_{i\neq j} \varphi_i(n) \langle S_i^+(t)S_i^-(t) \rangle,
\]

in which

\[
\varphi(n) = \frac{3}{16\pi} (1 + \cos^2 \vartheta), \quad \cos \vartheta = (n \cdot e_z),
\]

\[
\varphi_i(n) = \varphi(n) \exp \{i\delta_0 n \cdot (r_i - r_j)\},
\]

\[
\gamma_0 \equiv \frac{2}{|d_0|} \frac{2}{\hbar} \left( k_0 \equiv \frac{\omega_0}{c} \right).
\]

The derivation of these expressions can be found in [50]. In the present case, we have

\[
I_{\text{inc}}(n,t) = N\omega_0\gamma_0\varphi(n) \left[ 1 + s_z(t) \right],
\]

\[
I_{\text{coh}}(n,t) = \frac{1}{2} N^2\omega_0\gamma_0\varphi(n) F(k_0 n) w(t),
\]

where

\[
F(k) = \left[ \frac{1}{N} \sum_{j=1}^{N} e^{i k \cdot r_j} \right]^2.
\]

A sample inside a cavity resonator can radiate only in the direction of the cavity axis, that is, in the direction \( n = e_z \), when \( \vartheta = \pi/2 \). In this case,

\[
\varphi(e_z) = \frac{3}{16\pi}, \quad F(k_0 e_z) = \frac{4}{k_0^2 L^2} \sin^2 \left( \frac{k_0 L}{2} \right).
\]

Then the incoherent radiation intensity is

\[
I_{\text{inc}}(e_z,t) = \frac{3}{16\pi} N\omega_0\gamma_0 \left[ 1 + s_z(t) \right],
\]

while the coherent radiation intensity reads as

\[
I_{\text{coh}}(e_z,t) = \frac{3}{32\pi} N^2\omega_0\gamma_0 F(k_0 e_z) w(t).
\]  

To make estimates for the radiation intensity, let us consider the system parameters typical of ferroelectrics [56]. The dipole interaction is \( J \sim 10^2 \) K \( \approx 10^{13} \) Hz. Therefore, in order to make \( J \ll \omega_0 \), we should take an external electric field such that \( \omega_0 \) is least \( 10^{14} \) Hz. This corresponds to the mid-infrared range of the electromagnetic spectrum, with the wavelength \( \lambda \sim 10^{-3} \) cm.

Keeping in mind ferroelectrics of the order–disorder type, with proton or deuteron bonds, we have the electric dipoles \( d_0 \sim e_0 f_0 \), in which \( e_0 = 1.602 \times 10^{-19} \) C is the proton charge and the length \( f_0 \sim 10^{-8} \) cm is the distance between the minima of the effective double-well potential. Since one Coulomb is \( 1 \text{C} = 2.997 \times 10^9 \text{g}^{-1/2} \text{cm}^{-1/2} \text{s}^{-1} \), then

\[
ed_0 = 4.803 \times 10^{-19} \text{g}^{-1/2} \text{cm}^{-1/2} \text{s}^{-1}.
\]

Using the relation

\[1 \text{D} \sim 10^{-18} \text{erg}/\text{G} = 0.333 \times 10^{-22} \text{C} \text{m},\]

with \( 1 \text{G} = 1 \text{erg}/\text{cm}^3 \), we find

\[
d_0 \sim 10^{-18} \text{erg}/\text{G} \sim 1 \text{D}.
\]

This gives the peak radiation intensities (104) and (105) at the moment of time \( t_0 \) of the order

\[I_{\text{inc}}(e_z,t_0) \sim N \times 10^{-20} \text{W}, \quad I_{\text{coh}}(e_z,t_0) \sim N^2 F(k_0 e_z) \times 10^{-21} \text{W}.
\]

The number of atoms that are certain to radiate coherently is \( N_{\text{coh}} \sim \rho \lambda^3 \). With the average density \( \rho \sim 10^{23} \text{cm}^{-3} \) and the wavelength \( \lambda \sim 10^{-3} \), we get \( N_{\text{coh}} \sim 10^{14} \). If the wavelength is larger than the sample length, then

\[F(k_0 e_z) \equiv 1 \quad (\lambda > L).
\]

Therefore, we get

\[I_{\text{inc}}(e_z,t_0) \sim 10^{-6} \text{W}, \quad I_{\text{coh}}(e_z,t_0) \sim 10^{8} \text{W}.
\]

If the sample has a cylindrical shape, then the radiation beam, spreading along the cylinder axis, can split into filaments, each radiating coherently. This phenomenon of filamentation is well known experimentally (see, e.g. [62–64] and review articles [48, 65]) and has been explained theoretically [40, 66–69]. Each filament, radiating coherently, and having the volume \( V_{\text{coh}} = \pi r_f^2 L \), contains the number of particles

\[N_{\text{coh}} = \rho V_{\text{coh}} = \pi \rho r_f^2 L.
\]

The filament radius is found [40, 66–69] to be \( r_f = 0.3 \sqrt{\lambda L} \), hence

\[N_{\text{coh}} \approx 0.283 \rho \lambda L^2.
\]

For a sample of length \( L = 0.1 \) cm, the number of coherently radiating atoms in a filament can reach \( N \sim 10^{17} \). But when \( \lambda \ll L \), it is necessary to take into account that for real
samples, the scaling of the coherent radiation intensity with the number of atoms $N$ is not exactly $N^2$, but is usually lower, being caused by the geometric factor $F(k)$ and different experimental imperfections [70]. For an estimate, we can take

$$F(k_0 \epsilon_s) \sim \frac{\lambda^2}{\pi L^2} \quad (\lambda \ll L).$$

Then, with the sample length $L = 0.1$ cm, we get

$$I_{\text{neu}}(\epsilon_s, t_0) \sim 10^{-3} \text{ W}, \quad I_{\text{coh}}(\epsilon_s, t_0) \sim 10^{10} \text{ W}.$$

9. Conclusion

A ferroelectric is inserted into a resonator cavity. The motion of ferroelectric dipoles induces in the cavity an electric field acting back on these dipoles. The ferroelectric is prepared in a nonequilibrium initial state, in an electric field directed opposite to the ferroelectric dipoles. The motion of dipoles is triggered by the dipolar waves. The correlation between the moving dipoles is due to the resonator feedback field. The coherent motion of these electric dipoles produces coherent radiation, called superradiance.

The physics of the ferroelectric superradiance is principally different from atomic superradiance. In the latter, the process is initiated by spontaneous atomic radiation, while in ferroelectric superradiance, the process is triggered by dipolar waves. In atomic superradiance, collective radiation is induced by effective atom interactions through the common electromagnetic radiation field, while in ferroelectric superradiance, the collectivization of dipole motion is induced by the resonator feedback field. No ferroelectric superradiance can develop without the resonator. The enhancement of radiation by a resonator is termed the Purcell effect [47]. But here, the resonator not only enhances the coherent radiation, but induces coherence as such. Although mathematically there are direct analogies between the spontaneous radiation of atoms and dipole waves and between the common radiation field of atoms and the resonator feedback field [65, 71], the physics of atomic superradiance and ferroelectric superradiance are rather different.

Ferroelectric superradiance occurs in the infrared region. In that region, there exist many high-quality infrared resonators (see, e.g. [72–77]). Therefore it looks quite straightforward to realize ferroelectric superradiance in experiments.

Acknowledgments

I dedicate this paper to my dear friend V V Samartsev on the occasion of his 80th birthday. I have had the great pleasure of discussing with him a variety of problems related to the topic of the present paper.

Also, I am grateful for the discussions and help from E P Yukalova.

References

[1] Allen L and Eberly J 1975 Optical Resonance and Two-Level Atoms (New York: Wiley)

[2] Samartsev V V and Sheibut Y E 1975 Electromagnetic Superradiance (New York: Academic)

[3] Gross M and Haroche S 1982 Phys. Rep. 93 301

[4] Naboikin Y V, Samartsev V V, Zhinov P V and Silaeva N B 1986 Coherent Spectroscopy of Molecular Crystals (Kiev: Naukova Dumka)

[5] Andrianov S N, Naboikin Y V, Samartsev V V, Silaeva N B and Sheibut Y E 1986 Phys.—Usp. 29 1060

[6] Zinoviev P V, Samartsev V V and Silaeva N B 1990 Interaction of Electromagnetic Field with Condensed Matter eds N N Bogolubov et al (Singapore: World Scientific) p 244

[7] Zinoviev P V, Samartsev V V and Silaeva N B 1991 Laser Phys. 11

[8] Andreev A V, Emelyanov V I and Illinskii Y A 1993 Cooperative Effects in Optics (Bristol: Institute of Physics)

[9] Benedikt M G, Ermolov A M, Malyshev A V, Sokolov I V and Trifonov E D 1996 Superradiance—Multiatomic Coherent Emission (Bristol: Institute of Physics)

[10] Zinoviev P V, Zinov V V, Kalochev A A and Samartsev V V 2001 Laser Phys. 11 1307

[11] Kalachev A A and Samartsev V V 2003 Coherent Phenomena in Optics (Kazan: Kazan University)

[12] Dicke R H 1954 Phys. Rev. 93 99

[13] Yukalov V I 1998 Mathematical Physics ed L D Faddeev (Moscow: Grand Russian Encyclopedia) p 169

[14] Kopvillem U K, Samartsev V V and Sheibut Y E 1975 Phys. Status Solidi B 70 799

[15] Bashkirov E K, Shumovsky A S and Yukalov V I 1985 Dokl. Phys. 30 367

[16] Aavikosov Y, Lippmaa Y and Reinit T 1987 Opt. Spectros. 62 419

[17] Andrianov S N and Samartsev V V 1987 Laser Phys. 6 179

[18] Andrianov S N, Samartsev V V and Sheibut Y E 1987 Theor. Math. Phys. 72 884

[19] Hanamura E 1988 Phys. Rev. B 38 1228

[20] Sazonov C V 1988 Solid State Phys. 30 3226

[21] Itoh T, Iekhara T and Iabuchi Y 1990 J. Lumin. 45 29

[22] Boer S D and Wiersma D A 1990 Chem. Phys. Lett. 165 45

[23] Fiddler H, Knoester J and Wiersma D A 1990 Chem. Phys. Lett. 171 529

[24] Devead B, Clerot F, Roy N, Satzike K, Sermage B and Knoester J 1992 J. Lumin. 53 1001

[25] Bjork G, Pau S, Jacobson J M and Yamamoto Y 1994 Phys. Rev. B 50 17336

[26] Tokihiro T, Manabe Y and Hanamura E 1995 Phys. Rev. B 51 7655

[27] Bjork G, Pau S, Jacobson J M, Cao H and Yamamoto N 1995 Phys. Rev. B 52 17310

[28] Wang H Z, Zheng X G, Zhao Z L, Gao Z L and Yu Z X 1995 Phys. Rev. Lett. 74 4079

[29] Agranovich M, Basko D M and Dubovsky O A 1997 J. Chem. Phys. 106 3896

[30] Chen Y N and Chuu D S 2000 Phys. Rev. B 61 10815

[31] Chen Y N, Chuu D S, Brandes T and Kramer B 2001 Phys. Rev. B 64 125307

[32] Jiu G R, Zhang P, Liu Y X and Sun C P 2003 Phys. Rev. B 68 134301

[33] Singh M, Yukalov V I and Lau W W 1998 Nanostructures: Physics and Technology ed Z Alferov and L Esaki (Saint Petersburg: Ioffe Institute) p 327

[34] Chen Y N, Chuu D S and Brandes T 2003 Phys. Rev. Lett. 90 166802

[35] Temnov V V and Woggon U 2005 Phys. Rev. Lett. 95 243602

[36] Parascandolo G and Savona V 2005 Phys. Rev. B 71 045335

[37] Scheibner M, Schmidt T, Worschec L, Fischer A, Bacher G, Passow T and Hommel D 2007 Nat. Phys. 3 106

[38] Sitek A and Machnikowski P 2009 Phys. Rev. B 80 115319

[39] Yukalov V I and Yukalova E P 2010 Phys. Rev. B 81 075308
[41] John S and Quang T 1995 Phys. Rev. Lett. 74 3419
[42] Malyshev V and Jarque E C 1995 Opt. Soc. Am. B 12 1868
[43] Yukalov V I 1998 Laser Phys. 8 1182
[44] Yukalov V I 1999 Opt. Spectrosc. 87 550
[45] Yukalov V I 2000 Quantum Electron. 30 911
[46] Yukalov V I 2001 Eur. Phys. J. D 13 83
[47] Purcell E M 1946 Phys. Rev. 69 681
[48] Yukalov V I and Yukalova E P 2000 Phys. Part. Nucl. 31 561
[49] Yukalov V I and Yukalova E P 2004 Phys. Part. Nucl. 35 348
[50] Yukalov V I and Yukalova E P 2015 Laser Phys. 25 085801
[51] Yukalov V I 2018 Laser Phys. 28 053001
[52] Yukalov V I and Yukalova E P 2018 Phys. Rev. A 98 144438
[53] Yukalov V I and Yukalova E P 2018 Eur. Phys. J. D 72 190
[54] Patel J S and Hanson D M 1981 Nature 293 445
[55] Yukalov V I 1993 Laser Phys. 3 870
[56] Blinc R and Zeks B 1974 Soft Modes in Ferroelectrics and Antiferroelectrics (Amsterdam: North Holland)
[57] Wangsness R K 1955 Phys. Rev. 98 927
[58] Yukalov V I and Yukalova E P 2008 Phys. Rev. A 78 063610
[59] Mandel L and Wolf E 1995 Optical Coherence and Quantum Optics (Cambridge: Cambridge University Press)
[60] Checchin M and Martinello M 2016 (arXiv:1610.02083)
[61] Yukalov V I 1995 Laser Phys. 5 970
[62] Encinas-Sanz F, Leyva I and Guerra J M 2000 Phys. Rev. Lett. 84 883
[63] Encinas-Sanz F, Leyva I and Guerra J M 2000 Phys. Rev. A 62 043821
[64] Leyva I and Guerra J M 2002 Phys. Rev. A 66 023820
[65] Yukalov V I 2014 Laser Phys. 24 094015
[66] Yukalov V I 1988 J. Mod. Opt. 35 35
[67] Yukalov V I 1990 J. Mod. Opt. 37 1361
[68] Yukalov V I 1991 Laser Phys. 1 81
[69] Yukalov V I 2000 Phys. Lett. A 278 30
[70] Angerer A, Streitsov K, Astner T, Putz S, Sumiya H, Onoda S, Munro W J, Nemoto K, Schmiedmayer J and Majer J 2018 Nat. Phys. 14 1168
[71] Yukalov V I 2005 Laser Phys. Lett. 2 356
[72] Garin M, Fenollosa R, Alcubilla R, Shi L, Marsal L F and Meseguer F 2014 Nat. Commun. 5 3440
[73] Lecaplain C, Javerzac-Galy C, Gorodetsky M L and Kippenberg T J 2016 Nat. Commun. 7 13383
[74] Osman A, Nedeljkovic M, Soler Penades J, Wu Y, Qu Z, Khokhar A Z, Debnath K and Mashanovich G Z 2018 Opt. Lett. 43 5997
[75] Radosavljevic S, Beneitez N T, Katumba A, Muneeb M, Vanslembrouck M, Kuyken B and Roelkens G 2018 Opt. Mater. Express 8 824
[76] Yao K and Liu Y 2018 ACS Photon. 5 844
[77] Xiao T H, Zhao Z, Zhou W, Chang C Y, Set S Y, Takenaka M, Tsang H K, Cheng Z and Goda K 2018 Opt. Lett. 43 2885