Generation of terahertz impulse via an excited localized ensemble of identical particles with a permanent dipole moment

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Abstract. The paper states a simple and analytically solvable model for the generation of low-frequency emission via a localized excited ensemble of identical two-level quantum particles with a permanent dipole moment in conditions of collective spontaneous emission of the atomic ensemble.

1. Introduction
Electromagnetic processes in media with a permanent dipole moment (PDM), for example, in ensembles of quantum dots, are believed to be attractive both from the point of view of practical applications (generation of low-frequency terahertz emission) and from the theoretical point of view. Nevertheless, many theoretical papers are confined to numerical modeling for generation of low-frequency emission. Few exceptions are the papers which consider various questions of integrability of equations, see, for example, [1-5], or processes in optically thin media [6] and microcavities, for example [7]. These papers consider dynamics of quantum systems which consist of isolated PDM-based quantum particles, where the dipole-dipole interaction is neglected either due to sparseness of the system or any other reason. In particular, such assumptions can be made so as not to account possible phase transitions associated with the spontaneous occurrence of macroscopic polarization. Then dynamics of PDM-systems is reduced to the ordinary N-level optical models where certain quantum energy states are parity-free and are characterized as having PDM. Unlike N-level systems, an entire hierarchy of multi-quantum transitions is possible in PDM-systems for all possible resonances, with the time dynamics of a quantum system being characterized by low-frequency emission processes along with high-frequency (optical) components. The origin of low-frequency components can be easily comprehended on the basis of algebraic perturbation theory - according to such an approach, dynamics of the population of energy levels is characterized by slow time changes in comparison with the period of rapid oscillations of values determined by optical quantum transitions. Then, since particles have PDM, there arises an ordinary dipole generation of low-frequency emission.

It is to be noted that an adequate apparatus for the theory of optical resonance is the algebraic perturbation theory [8-10], which makes it possible both to obtain effective Hamiltonians of various optical processes and to describe all possible (time-frequency) hierarchies of polarizations of a quantum system. Under the given conditions the generation of terahertz emission is a side effect of superradiance of excited identical two-level PDM-based atoms in the ground and/or excited energy
states. The ensemble of two-level atoms is localized in a region with dimensions much smaller than the characteristic superradiance wavelength. Therefore, the dipole-dipole interaction does not manifest itself as the master equations for the populations of the energy levels under consideration. The suppression of superradiance, which becomes apparent in ensembles of a sufficient number of atoms, is also taken into account [11], which can be regarded as an additional control factor for the low-frequency emission.

2. Model definition
Suppose there are \( N \) fixed identical atoms in the subspace with dimensions much smaller than the characteristic wavelength, with some levels of PDM-based atoms. The Hamiltonian for such a system in the quantized electromagnetic field is given by a typical expression that includes the Hamiltonian of an isolated atoms \( \mathcal{H}^{t} \), the Hamiltonian of the quantized electromagnetic field \( \mathcal{H}^{F} \), and the operator of their interaction \( \mathcal{H}^{\text{int}} \):

\[
\mathcal{H}^{\text{int}} = \mathcal{H}^{t} \chi^{F} + \mathcal{H}^{\text{int}} + \mathcal{H}^{\text{int}}, \quad \mathcal{H}^{t} = \sum_{i,j} E_j | E_j > < i < E_j | ^{(i)}, \quad \mathcal{H}^{F} = \sum_{q} \hbar \omega_q b_q^\dagger b_q ,
\]

\[
\mathcal{H}^{\text{int}} = \sum_{q} \Gamma_q (b_q^\dagger + b_q) \sum_{i,j} d_{ij} | E_k > < i < E_j | ^{(i)}, \quad \sum_{j} | E_j > < i < E_j | ^{(i)}, \quad < E_j | ^{(i)} , \quad E_k > ^{(i)} = \delta_{jk} .
\]

Here \( | E_j > \) is the atomic quantum state of the energy \( E_j , d_{ij} =\langle E_k | d | E_j > , d = \sum_{ij} d_{ij} | E_k > < E_j | \)
is the dipole moment operator of an atom. The superscript of the atomic state vectors denotes the state space of the \( i \)-th atom and the summation over \( i \) is the summation over all atoms of the ensemble. Operators of creation \( b_q^\dagger \) and annihilation \( b_q \) of photons with a wave vector \( \vec{q} \) satisfy the typical commutation relation \( [b_q^\dagger, b_q] = \delta_{\vec{q}\vec{q}'} \), the dispersion relationship is given by the condition \( \omega_q = \omega c . \)

The coupling parameter of the electromagnetic field and atoms in three-dimensional space is \( \Gamma_q = (2\pi h \omega c / \ell^3)^{1/2} \), where \( \ell^3 \) is the quantization volume.

We will speak of two-level atoms, taking into account all the energy atomic states: a pair of levels is allocated by initial conditions - only the given atomic level \( | E_2 > \) is assumed to be populated at the initial instant of time, which is coupled by the frequency transition \( \omega_2 = (E_2 - E_1) / h \) with the low-lying ground level. For the sake of simplicity, let us suppose that there are no other single-quantum transitions from the excited level to the low-lying ones. In contrast to particles whose quantum states are characterized by parity, two-quantum transitions are possible in PDM-based media, along with single-quantum transitions from the excited level \( | E_2 > \) to the ground level \( | E_1 > , \) since there is an entire frequency spectrum in the vacuum electromagnetic field of ordinary three-dimensional space. However, two-quantum processes are second-order processes in the coupling constant of a quantum particle with an electromagnetic field and let us assume that there is no reason for a serious increase in the significance of such two-quantum processes in comparison with single-quantum processes in the system under consideration, as is the case for the same parity of states \( | E_1 > \) and \( | E_2 > , \) or in the case of processes in media with spectral singularities [12].

We will construct an effective Hamiltonian and obtain the master equation for the atoms. In case of open systems the major technique for constructing an effective Hamiltonian is the algebraic perturbation theory [8-10].

In the extended space of states of the atomic system and the quantized electromagnetic field, the initial Hamiltonian \( \mathcal{H}^{\text{int}} \) determines the Schrödinger equation for the state vector: \( i \hbar \frac{d}{dt} | \Psi^{AF} > >= \mathcal{H}^{\text{int}} | \Psi^{AF} > . \) According to the algebraic perturbation theory and methods of the effective Hamiltonian, assumptions about the nature of the initial state \( | \Psi^{AF} > \) at \( t = 0 \) are made after constructing the effective Hamiltonian of the problem.
3. The effective Hamiltonian, the master equation and low-frequency polarization of atoms

Following the general scheme [8-10], we transform the state vector $|Ψ^{A+F}>$

\[
|Ψ^{A+F} > = U |Ψ^{A+F} > , \quad U = e^{iS}, \quad S^+ = S .
\]

The transformed vector will satisfy the Schrödinger equation

\[
\frac{ih}{dτ} |Ψ^{A+F} > = \hat{H} |Ψ^{A+F} >
\]

with the transformed Hamiltonian

\[
\hat{H} = U\hat{H}^{bli}U^* - ihU \frac{d}{dτ} U^* .
\]

Expanding $\hat{H}$ and $S$ in series over the interaction with the vacuum field

\[
S = S^{(1)} + S^{(2)} + \ldots, \quad \hat{H} = \hat{H}^{(0)} + \hat{H}^{(1)} + \hat{H}^{(2)} + \ldots,
\]

we obtain (taking into account the Baker-Hausdorff formula [9])

\[
\hat{H}^{(0)} = \hat{H}^A + \hat{H}^F, \quad \hat{H}^{(1)} = \hat{H}^{bli} - i[S^{(1)}, \hat{H}^{(0)}] + \hbar \frac{d}{dτ} S^{(1)},
\]

\[
\hat{H}^{(2)} = -\frac{N}{2} [S^{(1)}, \hat{H}^{bli}] - \frac{1}{2} [S^{(1)}, \hat{H}^{(1)}] - i[S^{(2)}, \hat{H}^{(0)}] + \hbar \frac{d}{dτ} S^{(2)} .
\]

The major idea for finding the terms $S^{(i)}$ and the effective Hamiltonian $\hat{H}^{eff} = \hat{H}^{(0)} + \hat{H}^{(1)} + \hat{H}^{(2)}$ is the absence of rapidly varying terms in $\hat{H}^{(0)}$ in the interaction representation. The author's approach [8-10] differs from the other approaches to constructing the effective Hamiltonian of the theory of open quantum systems [13, 14] and leads to the notion that the algebraic perturbation theory has become self-consistent and efficient to describe both open quantum systems and quantum and nonlinear optical phenomena. Additionally, for open systems the differences are related to the Markov conditions that can be imposed on the initial non-transformed state vectors or the transformed state vectors. The differences arising here are vividly illustrated by the model problem considered in [15].

The conventional [8-11] calculations give rise to the terms in the second order of the effective Hamiltonian which are diagonal according to atomic variables and do not depend on photon variables (Lamb shifts). They can be easily included into $\hat{H}^{(0)}$. Then projection operators associated with the non-resonant energy levels can be omitted in the second-order term $\hat{H}^{(2)}$.

A term describing the dipole-dipole interaction of atoms tends to appear [11] in the second order of algebraic perturbation theory in case of an atomic interaction operator with a quantized electromagnetic field. This dipole-dipole interaction operator is neglected, since it does not exert a serious impact on the master equation for resonance level populations which is to be treated below. This leads to the following expressions (the tilde is also omitted):

\[
H^{(0)} = \hbar \omega_{21} R_{1}, \quad \omega_{21} = (E^{1}_2 - E^{1}_1)/\hbar, \quad H^{(1)} = \sum_\omega \Gamma_\omega^b \delta_{12} R_{1} + \sum_\omega \Gamma_\omega^b \delta_{23} R_{2},
\]

\[
H^{(2)} = \frac{N}{2} \sum_\omega \Gamma_\omega^b \sum_\omega \Gamma_\omega^b (\Pi(\omega) + \Pi(\omega')), \quad E^{1}_i = E_i + \sum_\omega \Gamma_\omega^2 \sum_j \frac{|d_{ij}|^2}{\hbar(\omega_{ij} - \omega)} \quad |E_i > > E_1, \quad E^{1}_{2} = E_2 + \sum_\omega \Gamma_\omega^2 \sum_j \frac{|d_{2j}|^2}{\hbar(\omega_{ij} - \omega)} \quad |E_2 > > E_2 |.
\]

During the calculations, unified parameters of the theory of optical resonance processes [8-10] are emerging regardless of the presence of classical and/or quantized fields, the presence or absence of PDM

\[
\Pi_\omega(\omega) = \sum_j \frac{|d_{ij}|^2}{\hbar} \left( \frac{1}{\omega_{ij} + \omega} + \frac{1}{\omega_{ij} - \omega} \right).
\]

The prime at the summation sign means that the summation eliminates the resonance terms $\omega_{jk} \mp \omega \approx 0$. In addition, the definition is used $\omega_{km} = (E^1_k - E^1_m)/\hbar$. 


The formula for the term of the effective Hamiltonian $H^{(2)}$ describing the Stark interaction suggests that the condition $\Pi_1(\omega) \approx \Pi_2(\omega) = \Pi(\omega)$ is satisfied for the resonance levels $|E_1>$ and $|E_2>$. The expression for $\Pi_2(\omega)$ contains no restrictions [8-11] for parity of the energy levels involved. Nevertheless, it is obvious that if only a pair of levels $|E_1>$ and $|E_2>$ were considered in the initial problem statement, then the presence of PDM would not have any effect on the system dynamics in the optically thin medium. The presence of nonresonant levels different from the considered ones leads to a change in the parameter $\Pi_2(\omega)$, triggers the dependence of this parameter on PDM and as result it becomes possible to discuss the PDM-based system dynamics for various values $\Pi_2(\omega)$.

The effective Hamiltonian $H^{eff} = \hat{H}^{(0)} + \hat{H}^{(1)} + \hat{H}^{(2)}$ is represented by atomic operators $R_j = \frac{1}{a} \sum_i (|E_j> <E_j| - |E_i> <E_i|)$, $R = \sum_i |E_i> <E_i|$, $R_2 = \sum_i |E_2> <E_2|.$

Satisfying commutation conditions $[R_j, R_k] = \pm R_i$, $[R_i, R_j] = 2R_3$.

The polarization of the medium $P$ is expressed in terms of the transformed density matrix, $ho = \mathbf{T}_F \mathbf{P}^{A+F} \mathbf{P}^{A+F}$ where the trace is taken over the states of the electromagnetic field. The general formula for polarization $P = N_a \mathbf{T}_\mathbf{F}D$ is determined by the effective dipole moment operator $D = e^{-i\sigma_d e^S} = d - i[S, d] - \frac{1}{2}[S, [S, d]] + \ldots \sum_{kj} D_{kj} |E_k> <E_j|.$

The low-frequency (LF) polarization component emerging in PDM-based media is of interest for our purposes. In this case, it can be written in the first non-vanishing order of the algebraic perturbation theory in its simple form

$$P^{LF} = N_a (d_{11} \rho_{11} + d_{22} \rho_{22}).$$

To calculate polarization $P^{LF}$ and determine parameters of the generated low-frequency emission it is necessary to obtain the master equation for the atomic density matrix. In case of absence of PDM, the formulated problem was solved in [11]. The results of algebraic perturbation theory make it possible to use the results of [11] under the conditions defined above and to immediately write down the master equation for the atomic density matrix according to the approximations described above.

We will consider the states of the atomic ensemble, symmetric in permutations of atoms. In that case they can be conveniently described by eigenvectors $|m>$ of the operator $R_j$: $R_j |m> = m |m>,$ $-r \leq m \leq r$. Here $r$ is the parameter characterizing the irreducible representation of the angular momentum algebra $su(2)$ for describing symmetric states of an ensemble of $2r = N_a$ identical atoms [11,16]. The state of an ensemble of fully excited quantum particles is given by a vector $|r>$, and the state of an ensemble, whose particles are in the ground state, is given by a vector $|-r>$. It is not difficult to write down master equations satisfying the effective Hamiltonian presented above in the Markov approximation [11] in the form:

$$\frac{dp_{mm}}{dt} = 2\gamma^2 (\eta_0 N_a)^{-2} [1 - \cos(\eta_0 N_a)] (g_{m+1} \rho_{m+1,m+1} - g_{m,m} \rho_{mm})$$

Here the dimensionless parameters are involved

$$\tau = \omega_0 f, \ \chi = \sqrt{2} \omega_0 d_2 \mu^{-1} c^{-3/2} h^{-1/2}, \ \eta_0 = 4d_2 \mu h c^{-3} \Pi(\omega_1).$$

Parameter $\mu \sim 1$ was involved to describe various models for a quantized electromagnetic field [11].

It can be seen that in neglecting the two-quantum decay of the excited level $|E_2>$ the main difference of the dynamics of the PDM-based system under study lies both in the magnitude $\eta_0$ and in inducing the low-frequency polarization (6), which takes the following form in terms of the introduced representation (henceforward everything is again expressed in dimensional values):
\[ P^{LF} = N_a \sum_{m=-r}^{r} \left( d_{22}(r+m) + d_{11}(r-m) \right) \rho_{mm} = p^{LF}(t) + \text{const}, \quad p^{LF}(t) = N_a (d_{22} - d_{11}) \sum_{m=-r}^{r} m \rho_{mm}. \] (8)

4. Impulse of low-frequency emission under superradiance

At the initial instant of time \( t = 0 \) all quantum particles of the ensemble are assumed to be excited, so that only the following one is nonzero \( \rho_{rr}(0) = 1 \) among the matrix elements of the density matrix. These initial conditions satisfy the Dicke ordinary superradiance [16], however, in contrast to the Dicke superradiance in Eqs. (7), the effect of collective-relaxation suppression by the Stark interaction, which is clearly manifested in localized atomic ensembles [11], is taken into account. In addition, equations (7) and (8) account for the presence of PDM-based atoms.

Let us consider a low-frequency component of the dipole emission of an atomic ensemble. For that purpose we will use the classical formula for the total intensity of dipole emission [17]. Using the dimensional values we have:

\[ I^{LF} = \frac{2}{\pi} e^{-3} \left( d^2 P^{LF}(t)/dr^2 \right)^2 = \frac{2}{\pi} e^{-3} N_a^2 (d_{22} - d_{11})^2 \left( \frac{d^2}{dr^2} \sum_{m=-r}^{r} m \rho_{mm} \right)^2. \]

The value \( \frac{d}{dt} \sum_{m=-r}^{r} m \rho_{mm} \) that determines the intensity of low-frequency emission is proportional to the rate of change in the average energy of the atomic system and, as a consequence, the intensity \( I^{SR}(t) \) of high-frequency superradiance:

\[ \frac{d}{dt} \sum_{m=-r}^{r} m \rho_{mm} = -(\hbar \omega_{21})^{-1} I^{SR}(t), \quad I^{SR}(t) = \hbar \omega_{21} \frac{d}{dt} n. \]

As a result, we obtain the relation between the required intensity of the low-frequency emission and the intensity of the superradiance of the atomic ensemble in the approximations under consideration:

\[ I^{LF} = \frac{2}{\pi} e^{-3} \left( d^2 P^{LF}(t)/dr^2 \right)^2 = \frac{2}{\pi} e^{-3} \hbar^2 \omega_{21}^{-2} N_a^2 (d_{22} - d_{11})^2 \left( d I^{SR}(t)/dr \right)^2. \]

To obtain the expression for the average number of \( n \) photons of the frequency \( \omega_{21} \) radiated by the time moment \( t \), we use the estimate of the mean emission time \( t_n \) of these \( n \) photons as

\[ t_n = \frac{(n_{\omega_{21}} N_a)^2}{2 \lambda^2 \omega_{21} [1 - \cos(n_{\omega_{21}} N_a)]} \sum_{n=1}^{n} g_{mm}. \]

Under the conditions of suppression of collective relaxation processes, the intensity of superradiance at \( N_a \gg 1 \) can be estimated as

\[ I^{SR}(t) \approx \tilde{\gamma} \hbar \omega_{21} \frac{1}{2} N_a^2 \text{sech}^2[\tilde{\gamma} N_a (t - t_D)/2], \quad \tilde{\gamma} = 2 \lambda^2 \beta \omega_{21} (n_{\omega_{21}} N_a)^2 [1 - \cos(n_{\omega_{21}} N_a)], \quad t_D = (\tilde{\gamma} N_a)^{-1} \ln \tilde{\gamma} N_a, \]

where \( \beta \) is the dimensionless geometry factor.

Eventually, the intensity of low-frequency emission takes the following form

\[ I^{LF} = \frac{2(d_{22} - d_{11})^2}{3e^c} \omega_{21} N_a \beta_0 \left( 1 - \cos(n_{\omega_{21}} N_a) \right)^4 \text{sh}^2[\tilde{\gamma} N_a (t - t_D)/2] \left( \frac{N_a}{n_{\omega_{21}}} \right)^4 \text{ch}^2[\tilde{\gamma} N_a (t - t_D)/2]. \] (9)

Compared with the intensity of ordinary superradiance, low-frequency generation depends heavily on the number of atoms in the ensemble \( I^{LF} \sim N_a^8 \) and appears to be of a two-humped form of a zero value corresponding to the maximum intensity of the superradiance pulse. Time interval \( T \) between the maxima of low-frequency emission can be estimated as \( T = 2(\tilde{\gamma} N_a)^{-1} \ln(2 + \sqrt{3}) \) and as the number of atoms increases, the distance between the peaks of low-frequency emission as well as their duration drop to zero value. In case of a significant number of atoms \( N_a \sim n_{\omega_{21}}^{-1} \) the emission intensity begins to decrease, and at \( N_a = 2\pi n_{\omega_{21}}^{-1} \) vanishes similar to the high-frequency superradiance [11].
Comparison of the derived formulae with the other cases of low-frequency generation emission under single-quantum resonance conditions [6,7] shows that the intensity of low-frequency emission depends on the difference of PDM values in the ground and excited states $d_{22} - d_{11}$.

5. Conclusion
The algebraic perturbation theory for optical processes provides a description of optical processes under resonant conditions, which causes the slow dependence of the populations of quantum levels on time, in comparison with the rapid time changes of optical properties within a quantum system. Then the quantum system always has a low-frequency component of polarization in PDM-based systems. Thus, the presented model analysis of the generation of low-frequency emission allows us to make a clear distinction between the low-frequency emission as a side action of optical effects in PDM-based media and the generation of low-frequency emission caused by the inherent property of the generated medium. And although the presence of PDM is often an integral property of the medium, the explanation of generation of terahertz emission in this case reduces frequently to the derivation of this very permanent dipole moment. Meanwhile, the very fact of generation of terahertz emission in optical processes in optically thin media with a permanent dipole moment does not require any new considerations other than the typical application of algebraic perturbation theory (the basis of the theory of optical resonance [10-12]) and the classical theory of dipole emission [17] (or certain approximations, for example, the approximation of unidirectional waves [18]).

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