Local-field effects and configurational disorder in nonlinear optical systems with coherence

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We argue that in nonlinear optical systems with randomly distributed atoms one should go beyond the Clausius-Mossoti limit in order to take into account the effect of local-field fluctuations induced by *configurational* disorder in atom positions. This effect is analyzed by means of a random local-mean-field approach with the neglect of the correlations between dipole moments of different atoms. The formalism is applied to three-level Λ-type systems with quantum coherence possessing an absorptionless index of refraction and lasing without inversion. We show that the effect of configurational fluctuations results in the significant suppression of the atom susceptibility compared to the predictions based on the Clausious-Mossoti equation. [S1050-2947(98)08106-2]

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I. INTRODUCTION

It is well known that in a system of interacting atoms the local field acting on each atom is different from the macroscopic electric field described by Maxwell's equation [1–3]. In its simplest form the local field caused by the near dipole-dipole interactions between atoms is given by the Lorentz formula [1]

$$E_L(t)=E(t)+\frac{4\pi}{3}P(t), \tag{1}$$

with $E(t)$ being the time-dependent macroscopic electric field and $P(t)$ the macroscopic polarization of the system, which is determined self-consistently from

$$\tilde{P}(\omega)=n\chi(\omega)\tilde{E}_L(\omega), \tag{2}$$

where the variables $\tilde{P}(\omega)$, $\tilde{E}(\omega)$, and $\tilde{E}_L(\omega)$ are the Fourier components of their time dependent analogs, n is the atom volume density, and $\chi(\omega)$ is a frequency dependent single-atom nonlinear polarizability. Equations (1) and (2) result in the Clausius-Mossoti (CM) equation for the macroscopic susceptibility χ_{DD} related to the dielectric permittivity as $\epsilon=1+4\pi\chi_{DD}$

$$\chi_{DD}=\frac{n\chi}{1-\frac{4\pi}{3}n\chi}. \tag{3}$$

The local-field corrections in the form of Eq. (1), and hence Eq. (2), are valid for a static [1–3] as well as a time-dependent [4–6] field $E(t)$. In particular, in gases Eq. (1) corresponds to the account of hard-core interactions [4,5], which is equivalent to excluding the polarization-induced electric field inside the Lorentz sphere [2,6].

We will be interested here in nonlinear optical systems with quantum coherence, where the CM relation has been employed recently to treat a number of phenomena such as intrinsic optical bistability [7], linear and nonlinear spectral shifts [8,9], lasing without inversion and an absorptionless index of refraction [10,11], and electromagnetically induced transparency [12]. In particular, we will concentrate below on widely discussed [11–17] nonequilibrium three-level Λ systems (with their size less than the resonant wavelength) possessing quantum coherence between the lower two levels. It has been suggested [10,11] to use the atom concentration as a control parameter and shown through the employment of the CM relation that an enhancement of the two orders of magnitude would be achieved in the refractive index compared to the noninteracting atom limit. A possibility of such a remarkable increase of the system susceptibility arises from the unusual behavior of the real and imaginary parts of the susceptibility, so that in Λ systems with coherence a situation is possible with $\chi''\to 0$ just in the vicinity of the frequency ω_m where χ' reaches its maximum. It follows then from Eq. (3) that a dramatic increase of χ_{DD} is expected by properly adjusting the atom density such that $\chi'\approx 3$.

In this paper we go beyond the CM limit. Note first that Eqs. (1) and (2) and hence the CM relation (3) correspond to the account of dipole-dipole interactions between atoms in the mean-field approximation, an issue that has received intensive study (e.g., [18–21]). It is established that the CM relation properly reproduces the effect of the dipole interaction in the linear regime where the polarizability of each atom does not depend on the value of the local field. The situation is more complicated if the atom polarizability depends on the value of the local field and therefore is sensitive to the local-field fluctuations. As shown earlier [22,23] from the calculations of the distribution function of the local field (based on the Margenau statistical theory [24]), local-field fluctuations can be very large in random systems with dipole-dipole interactions due to effect of configurational disorder. For example, they eliminate the equilibrium phase transition, predicted by the mean-field theory, for an ensemble of randomly distributed atoms interacting with electromagnetic field (see the Appendix). It is apparent that the account of the local-field fluctuations should result in the modification of Eq. (3) for the frequency-dependent susceptibility of interacting atoms.

We will show in the following sections that the proposed formalism exploiting self-consistent calculations of the distribution function of the local field experienced by each atom from its neighbors allows one to give a quantitative predic-

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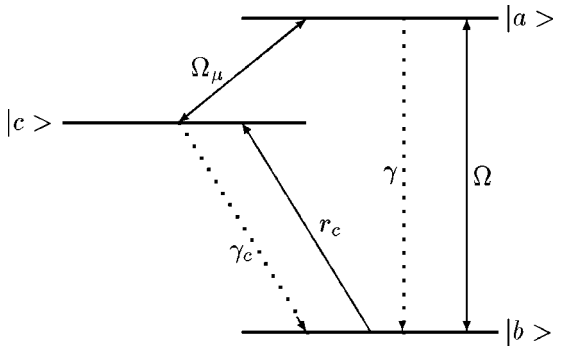

FIG. 1. Level scheme for the atomic system discussed.

tion for the behavior of the frequency-dependent susceptibility in systems of randomly distributed interacting atoms. The values of the susceptibility obtained are well below its values given by the CM equation. The proposed formalism is the most appropriate to treat an ensemble of atoms randomly frozen in crystals or amorphous hosts that (under the condition of vanishing χ_e'' at a particular frequency) undergoes a phase transition to nonequilibrium optical spin-glass state [25]. However, the estimates obtained for the susceptibility should be valid for gases as well (where a spin-glass state cannot be realized due to the effect of atom collisions).

II. MODEL

As a model possessing a quantum coherence we consider a three-level Λ system [17] shown in Fig. 1. In this system the coherence is maintained by a strong microwave field that couples the upper level of the probe field transition $|a\rangle \rightarrow |b\rangle$ to an auxiliary level $|c\rangle$. In Fig. 1 γ and γ_c are the decay rates and r_c is the pump rate. An indirect pump is necessary to maintain a nonvanishing population of the excited levels. Ω_μ is the Rabi frequency of the microwave field.

Kinetic equations for the density matrix elements [17] in the rotating reference frame with the decay and pump rates indicated in Fig. 1 reduce to

$$\dot{\rho}_{aa} = -\gamma\rho_{aa} - i\Omega(\rho_{ba} - \rho_{ab}) - i\Omega_\mu(\rho_{ca} - \rho_{ac}), \quad (4)$$

$$\dot{\rho}_{bb} = \gamma\rho_{aa} - r_c\rho_{bb} + \gamma_c\rho_{cc} + i\Omega(\rho_{ba} - \rho_{ab}), \quad (5)$$

$$\dot{\rho}_{cc} = -\gamma_c\rho_{cc} + r_c\rho_{bb} - \gamma_c\rho_{cc} + i\Omega_\mu(\rho_{ca} - \rho_{ac}), \quad (6)$$

$$\dot{\rho}_{ac} = -\gamma_{ac}\rho_{ac} + i\Omega_\mu(\rho_{aa} - \rho_{cc}) - i\Omega\rho_{bc}, \quad (7)$$

$$\dot{\rho}_{ab} = -(i\Delta + \gamma_{ab})\rho_{ab} + i\Omega(\rho_{aa} - \rho_{bb}) - i\Omega_\mu\rho_{cb}, \quad (8)$$

$$\dot{\rho}_{cb} = -(i\Delta + \gamma_{cb})\rho_{cb} + i\Omega\rho_{ca} - i\Omega_\mu\rho_{ab}. \quad (9)$$

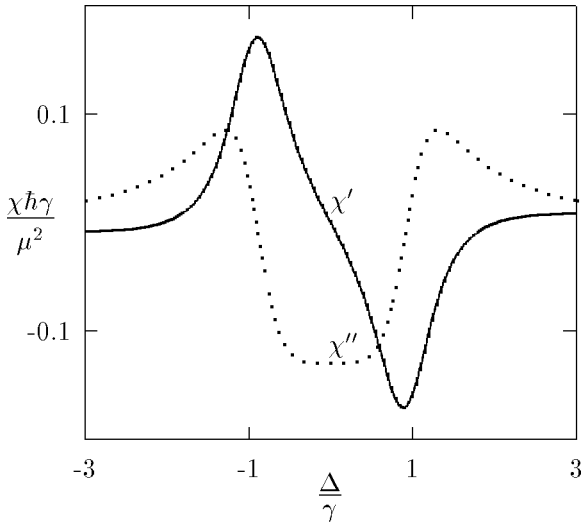

FIG. 2. Real and imaginary parts of the susceptibility for a three-level Λ system given by Eq. (10) for $\gamma_{ab} = \gamma_{ac} = 0.8\gamma$. $\gamma_{bc} = 0.08\gamma$.

Here $\Omega = \mu E_L/\hbar$ is the Rabi frequency associated with the field E_L, μ is the dipole moment matrix element between levels $|a\rangle$ and $|b\rangle$, $\Delta = \omega_{ab} - \omega$, and we assume that $E_L(t) = 2E_L\cos(\omega t)$

Using the conventional definition of the susceptibility $\chi = -\mu\rho_{ab}/E_L$ [17], and Eqs. (8), and (9), it is convenient to present χ in the form

$$\chi(\Delta) = \frac{\mu^2}{\hbar}\frac{i(\rho_{aa} - \rho_{bb})(i\Delta + \gamma_{bc}) + \Omega_\mu\rho_{ca}}{\Omega_\mu^2 + (i\Delta + \gamma_{ab})(i\Delta + \gamma_{bc})}. \quad (10)$$

The components ρ_{ca} of the density matrix in Eq. (10) represents the effect of the quantum coherence (induced by a microwave field) on the susceptibility of the system with respect to the local field E_L. For a weak E_L field (linear regime) one can neglect the dependence of ρ_{aa}, ρ_{bb}, and ρ_{ca} on Ω. The frequency dependence of the susceptibility in this linear regime, obtained by the numerical solution of Eqs. (4)–(7) and (10), is shown in Fig. 2, where the values of the parameters $\gamma_c = 0.1\gamma$ and $\omega_\mu = r_c = \gamma$ are chosen according to Ref. [17]. As already pointed out by Scully [15], a remarkable feature of the susceptibility in the system with coherence is the possibility of $\chi'' = 0$ at a frequency near the maximum of χ'. One can see from Fig. 2 that with the sets of parameters chosen $\chi'' = 0$ at $\Delta \approx -0.92\gamma$.

Note also that Eq. (10) can be easily generalized to the case of the linear response of the system to the additional weak field E_1 (with frequency ω_1 not necessarily equal to ω) in the presence of the strong field E_L controlling the density matrix components ρ_{aa}, ρ_{bb}, and ρ_{ca}. It is apparent that in this case the susceptibility $\chi(\Delta, \Delta_1, \Omega_1)$ describing the atom response to the weak field E_1 assumes the form

$$\chi(\Delta_1; \Delta, \Omega) = \frac{\mu^2}{\hbar}\frac{i[\rho_{aa}(\Delta, \Omega) - \rho_{bb}(\Delta, \Omega)](i\Delta_1 + \gamma_{bc}) + \Omega_\mu\rho_{ca}(\Delta, \Omega)}{\Omega_\mu^2 + (i\Delta_1 + \gamma_{ab})(i\Delta_1 + \gamma_{bc})}. \quad (11)$$

In the following sections Eqs. (10) and (11) will be used for the analysis of the effect of configurational fluctuations on the system macroscopic susceptibility.

III. LOCAL-MEAN-FIELD FORMALISM

The CM relation (3), based on Eq. (1) for the local field, does not take into account the effect of fluctuations of atom dipole moments. These fluctuations originate from the sign-changeable dipole-dipole interaction and random spatial atom positions. A physically transparent formalism to treat dipole moment configurational fluctuations, caused by the random atom positions, is a local-mean-field description. A starting point of the local-mean-field formalism is the assumption that each atom is characterized by the local dipole moment

$$m_i=\langle\hat{\mu}_i\rangle_{E_{Li}}, \tag{12}$$

where $\langle\hat{\mu}_i\rangle_{E_{Li}}$ denotes the quantum-statistical average of the ith atom dipole moment operator $\hat{\mu}_i$ in the local self-consistent field E_{Li}, given by

$$E_{Li}(t)=\sum_{j=1}^{N} J_{ij}m_j(t)+E_{ex}(t), \tag{13}$$

where $E_{ex}(t)$ is the applied external field. The function J_{ij} determines the dipole-dipole interaction between atoms i and j. In a retardationless approximation, which is valid if the size of the system is less than the resonance wavelength, we have

$$J_{ij}\equiv J(r_{ij})=\frac{3(n_{ij})_x^2-1}{r_{ij}^3}. \tag{14}$$

The effect of retardation on the form of the dipolar field has been discussed, e.g., in Refs. [26,4,5,27,7]. In Eqs. (13) and (14) we assumed, for simplicity, that all dipole moments are oriented along the axis x; $\mathbf{n}_{ij}=\mathbf{r}_{ij}/r_{ij}$, where $\mathbf{r}_{ij}=\mathbf{r}_i-\mathbf{r}_j$ is the radius-vector separating atoms i and j. E_{Li} in Eq. (13) is the x component of the local electric field.

The mean-field approximation (1) corresponds to the replacement of E_{Li} in Eq. (13) by

$$E_L=E_{ex}+\sum_j \overline{J_{ij}m_j}=E_{ex}+P\int d\mathbf{r}\; J(\mathbf{r}), \tag{15}$$

where the overbar denotes the configurational average

$$\overline{[\cdots]}=\frac{1}{V_0^N}\int dr_1\cdots\int dr_N[\cdots] \tag{16}$$

(V_0 is the sample volume). The volume integral in Eq. (15) can be replaced by two surface integrals over the outer surface of the sample giving rise to the depolarizing field E_{dep}, and the inner spherical Lorentz surface giving rise to the Lorentz local field $(4\pi/3)P$. Thus $E=E_{ex}+E_{dep}$ and therefore Eqs. (15) and (1) are identical.

It is convenient to rewrite Eq. (13) in the form

$$E_{iL}(t)=E(t)+\frac{4\pi}{3}P(t)+e_i(t), \tag{17}$$

$$e_i(t)=\sum_j \tilde{J}_{ij}m_j(t), \quad \tilde{J}_{ij}=J_{ij}-\frac{1}{V_0}\int d\mathbf{r}\; J(\mathbf{r}). \tag{18}$$

Equations (17) and (18) imply that $\Sigma_i m_i/V_0$ is a self-averaging variable equal to the macroscopic polarization P. The first moment $\overline{\tilde{J}_{ij}}$ of $\tilde{J}_{ij}$ given by Eq. (18) satisfies $\overline{\tilde{J}}_{ij}=0$ and the higher moments $\overline{\tilde{J}_{ij}^N}=\overline{J_{ij}^N}$, i.e., in practice, one can use $\tilde{J}_{ij}$ in the form of Eq. (14) and perform the integration in spherical reference frame providing $\overline{J_{ij}}=0$. Note that the local-mean-field approximation describing the system of interacting atoms in terms of local dipole moments, with the neglect of the quantum correlation effects, in some sense is similar to the well-known Hartree approximation.

Let us consider first the case of the absence of the external field and hence the absence of the macroscopic polarization P. The local polarization may still be nonzero due to the local-field fluctuations that have to be evaluated self-consistently. The key point in the consideration below is a special form of the atom susceptibility provided $\chi''=0$ at a particular frequency. In conventional systems with $\chi''\neq 0$ (e.g., two-level systems) the local polarization cannot exist in a steady state in the absence of the external field due to the effect of dissipation. In contrast, in systems with $\chi''=0$ at $\omega=\omega_m$ we have

$$\tilde{m}_i(\omega_m)=\chi'(\omega_m)\tilde{e}_i(\omega_m). \tag{19}$$

Here $\tilde{m}_i$ and $\tilde{e}_i$ are the Fourier components of the variables m_i and e_i, respectively. We will show that Eqs. (19) and (18) may have a nontrivial solution for the average local polarization characterized by a dimensionless parameter

$$M=\frac{1}{\mu}\overline{|\tilde{m}_i(\omega_m)|}. \tag{20}$$

Since in the absence of the external field macroscopic polarization $P=0$, the existence $M\neq 0$ implies the noncoherent steady state oscillations of the atom dipole moments at the frequency ω_m with random amplitudes. Thus the parameter M can be considered as the order parameter of the nonequilibrium optical spin-glass state [25].

In order to find the values of M note that the vanishing of χ'' results in the zero values of the imaginary part of the dipole moment amplitudes $\tilde{m}_i''(\omega_m)$ and hence, according to Eq. (13), the random local field $\tilde{e}_i$ in Eq. (19) is a real variable. We evaluated the distribution function of the local field

$$f(\tilde{e})=\overline{\delta(\tilde{e}-\tilde{e}_i)} \tag{21}$$

with the use of the self-consistent method [22,23], which is based on the Margenau [24] statistical theory. The function $f(e)$ possesses the Lorentzian form, which is characteristic of the dipole local-field distribution in random systems

$$f(\tilde{e})=\frac{1}{\pi}\frac{\delta}{\delta^2+\tilde{e}^2}, \quad \delta=5.1n\mu M. \tag{22}$$

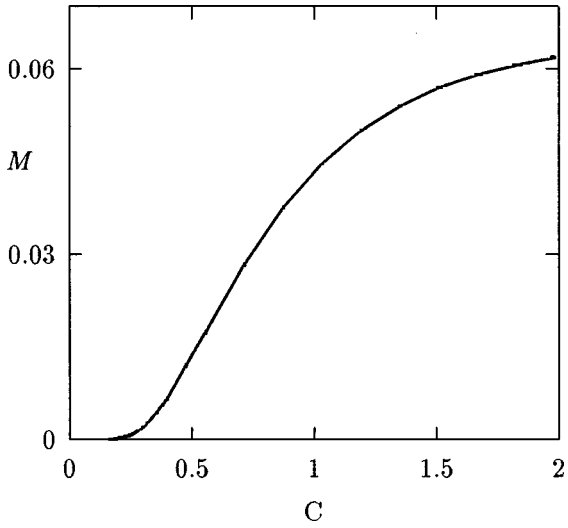

FIG. 3. Dependence of M on the parameter C ($\Delta=-0.92\gamma$).

One can see that the function $f(\tilde{e})$ depends parametrically on the parameter M, which can be obtained self-consistently from

$$M=\int d\tilde{e}|\chi'(\omega_m,\tilde{e})\tilde{e}|f(\tilde{e},M). \tag{23}$$

The value of M depends on the dimensionless parameter

$$C=\frac{\mu^2 n}{\hbar\gamma} \tag{24}$$

characterizing the strength of the dipole-dipole interaction. The values of $M(C)$ calculated from Eq. (23) are presented in Fig. 3.

Note that, as follows from Fig. 3, random local-mean-field theory predicts that the parameter M has finite (although very small) values even at very low dipole concentrations. This apparent defect of the theory originates from the neglect of correlation effects between closely separated atoms. As shown in our earlier work [28,25], closely separated atoms in random systems with a dipole-dipole interaction contribute only to localized states of the dipole-dipole interaction matrix and therefore do not affect the true cooperative phenomena. An estimate of the critical value of the parameter C_{cr} below which an optical spin-glass state does not exist (i.e., $M=0$) can be found from the criterion $5n_{cr}\chi'\approx 1$ [25] obtained with the use of computer simulations applied to the linearized local mean field equations (18) and (19). Taking into account the numerical value $\chi'\approx 0.17$ at $\Delta=-0.92\gamma$ (see Fig. 2), it gives

$$C_{cr}\approx 1.1. \tag{25}$$

It is interesting to note that the value of C_{cr} obtained is close to the inflection point of the curve $m(C)$.

Although the consideration above implies that optically active atoms are frozen in random positions in crystals or amorphous hosts, the obtained width δ of the local-field distribution function is a characteristic of the local-field fluctuations in gases too and should be taken into account in the calculations of the macroscopic susceptibility.

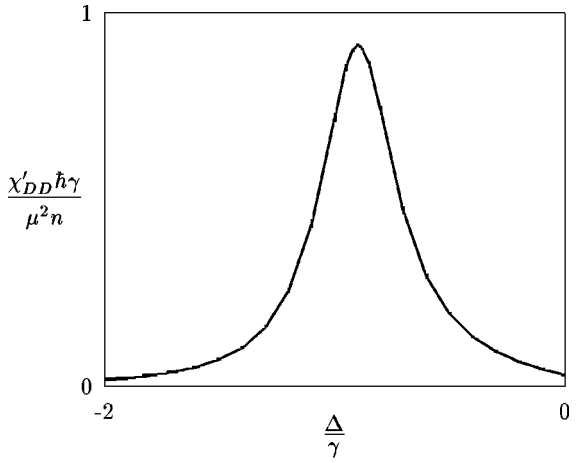

FIG. 4. Susceptibility χ'_{DD} calculated using Eq. (27) with $C=1.4$.

IV. MACROSCOPIC SUSCEPTIBILITY

The next step is to calculate the linear response of the system of interacting atoms in the external (macroscopic) field in the presence of local-field fluctuations. Since we assume that the external field is small [as well as the field-induced macroscopic polarization $P(t)$], one may neglect the change of the distribution function $f(\tilde{e})$ by the external field. In this case Eq. (17) contains two independent contributions: (a) a weak macroscopic field and a Lorentz field both oscillating at frequency ω of the external field and (b) a strong field $e_i(t)$ possessing a random amplitude and oscillating at frequency ω_m. A susceptibility describing the atom's response to the weak field in the presence of the strong oscillating field $e(t)$ is given by Eq. (11) with $\Delta=\Delta_m\equiv(\omega-\omega_{ab})$ and $\Omega=\Omega_{\tilde{e}}\equiv\mu\tilde{e}/\hbar\gamma$. In order to obtain the effective single-atom susceptibility κ describing the atom's response to the weak field one should average Eq. (11) over the random amplitudes $\tilde{e}$,

$$\kappa(\Delta_1)=\int d\tilde{e}\ \chi(\Delta_1;\Delta_m,\Omega_{\tilde{e}})f(\tilde{e}). \tag{26}$$

It follows then from Eq. (13) that in terms of the susceptibility κ the macroscopic polarization $\tilde{P}$ can be written as

$$\tilde{P}(\Delta_1)=n\kappa(\Delta_1)\left(\tilde{E}+\frac{4\pi}{3}\tilde{P}(\Delta_1)\right), \tag{27}$$

which results in the replacement of the susceptibility χ by κ in Eq. (3), i.e.,

$$\chi_{DD}(\Delta_1)=\frac{n\kappa(\Delta_1)}{1-\frac{4\pi}{3}n\kappa(\Delta_1)}. \tag{28}$$

In Fig. 4 we present the values of the real part of the macroscopic susceptibility χ'_{DD} obtained for $C=1.4$ and $\Delta=-0.92\gamma$. The other parameters were chosen as $\gamma_{ab}\approx\gamma_{ac}\approx 0.8$ and $\gamma_{bc}\approx 0.08$. For these values of the parameters the CM equation predicts more than two orders of magnitude enhancement of the χ'_{DD} compared to the susceptibility of

noninteracting atoms. At the same time, taking into account the local-field fluctuations, an increase in susceptibility of only five times is expected as follows from Fig. 4. Thus the effect of local-field fluctuations leads to a significant suppression of the susceptibility of interacting atoms in nonlinear optical systems with coherence.

V. CONCLUSION

We discussed the limitations of the applicability of the Clausius-Mossoti equation for the evaluation of the macroscopic susceptibility in nonlinear optical systems with configurational disorder in atom positions. It was shown that the corrections to the CM equation are especially important in nonlinear systems with quantum coherence with the vanishing imaginary part of the atom susceptibility for a particular frequency of the applied field. Under these conditions the CM equation predicts a dramatic increase of the real part of the macroscopic susceptibility of the ensemble of interacting atoms. We have shown that the latter effect is suppressed significantly due to the role of the configurational fluctuations of local fields, which should be taken into consideration.

APPENDIX: ABSENCE OF FERROELECTRIC PHASE TRANSITION FOR AN EQUILIBRIUM ENSEMBLE OF TWO-LEVEL ATOMS INTERACTING WITH AN ELECTROMAGNETIC FIELD

In this appendix we will apply the local-mean-field formalism for the analysis of the cooperative behavior of an ensemble of randomly positioned two-level atoms interacting with an electromagnetic field. It has been proposed [29–32] that such systems can undergo a second-order ferroelectric phase transition with the appearance of spontaneous atom polarization. Earlier papers [29–31] related to this problem have used a quantum formalism. In analogy to the Dicke model for superradiance, it was assumed that the main effect of atom-atom interactions comes from the coupling of each atom to the single mode of the electromagnetic field, corresponding to the wave vector $k=0$. However, the same conclusion can be reached [32] within the semiclassical formalism based on the CM equation (mean-field theory) and the concept of the Lorentz local field since the transition temperature T_c corresponds to the temperature of the divergence of χ_{DD}. A static susceptibility of a two-level atom is given by

$$\chi(T,E_L)=\frac{\mu^2}{\sqrt{(\hbar\omega_0)^2+(\mu E_L)^2}}\tanh\frac{\sqrt{(\hbar\omega_0)^2+(\mu E_L)^2}}{k_BT}, \tag{A1}$$

where $2\hbar\omega_0$ is the energy separation between the upper and lower levels. Combining Eqs. (3) and (A1), we obtain the equation for the phase transition temperature in mean-field approximation

$$\frac{4\pi}{3}n\chi(T_c,E_L=0)=1. \tag{A2}$$

In order to take into account the effect of configurational fluctuations of the local-field one should replace in Eq. (A2) (accordingly to the formalism discussed in the text) the susceptibility χ by the effective susceptibility

$$\kappa(T)=\int d\tilde{e}\ \chi(T,\tilde{e})f(\tilde{e}), \tag{A3}$$

where $f(\tilde{e})$ is given by Eq. (22). A solution of the self-consistent equation (23) with χ given by Eq. (A1) determines the values of the width δ of the function $f(\tilde{e})$. These values have then been used for the calculation of the effective susceptibility $\kappa(T)$ in order to establish the possibility of the appearance of ferroelectric ordering. We found that Eq. (A2), with χ replaced by κ, does not have solutions at any temperature and concentration, implying the absence of ferroelectric ordering in the system of randomly distributed two-level atoms being in thermal equilibrium with an electromagnetic field.

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