Local-field effects in a dense collection of two-level atoms embedded in a dielectric medium: Intrinsic optical bistability enhancement and local cooperative effects

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We investigate the effect of a linear host medium on the dynamics of a dense collection of resonant atoms. We find that near dipole-dipole interaction is enhanced by the presence of the host dielectric, resulting in a lower threshold density and a greater hysteresis area for intrinsic optical bistability. Cooperative decay terms appear that represent the interaction of near dipoles mediated by the absorptive component of the dielectric function of the host medium. These terms are part of the local response and correspond to local cooperative decay effects.

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Recent experiments [1,2] by Hehlen, Güdel, Shu, Rai, Rai, and Rand (HGSR[3]) have provided dramatic verification of theoretical predictions [3] of intrinsic optical bistability (IOB) due to near dipole-dipole (NDD) interactions. These experiments were performed with Yb^{3+} ions in a $Cs_3Y_2Br_9$ crystal at densities sufficient to cause strong local-field effects, but radiationless exchange interactions, as well. The exchange interaction contribution, which corresponds to pair upconversion in the excited state, evidently produces enhancement of the IOB by increased hysteresis area. Thus, we could say that the HGSR[3] experiments exhibit exchange interaction assisted IOB.

While the HGSR[3] experiments were performed with resonant systems embedded in a crystal host, previous theoretical treatments of NDD interactions have assumed a dense collection of resonant systems *in vacuo* [1,3–10]. In this paper, we investigate the effect of the linear polarizability of a host material, such as a crystal or amorphous material, on the dynamics of a dense embedded collection of two-level atoms. Allowing for dispersion and absorption, we assume that the dielectric function of the host can be represented by a complex constant, where the value of the constant depends on the frequency of the driving field. In the context of the Kramers-Kronig relations, this can be viewed as an approximation in which the dielectric function is essentially constant over the range of frequencies in the local field. In the Drude model, this condition can be satisfied if the range of frequencies in the local field is sufficiently small compared to the detuning of the driving frequency from the resonance frequency of the background dielectric medium. In the steady-state limit, the dielectric function must be constant regardless of the model of the background polarizability. While the generalized Bloch-Drude model will be discussed in a future publication, the assumption in the current work that the dielectric function of the host material has a constant value clarifies the presentation, allows identification of the underlying physical processes in dispersive dielectrics, and makes the results independent of a specific model of the background polarizability.

After deriving the equations of motion, we find that the inversion-dependent atomic resonance frequency renormalization arising from the NDD interaction is enhanced by the presence of the host material. This enhancement leads, in the steady-state limit, to a lower threshold density and a greater hysteresis area for intrinsic optical bistability. Further, cooperative decay terms appear which are quite novel in that they represent the interaction of near dipoles mediated by the imaginary component of the dielectric function of the host material. Significantly, these terms, which are not dependent on propagation or sample size in the macroscopic equations of motion, contribute to the local response and correspond to local cooperative decay effects.

The polarization of the medium, which is composed of an isotropic homogeneous distribution of resonant systems in a linearly polarizable host, is calculated using a phenomenological approach due to Lorentz [11] and Bloembergen [12]. The total polarization,

$$\mathcal{P}=\mathcal{P}^{\rm bg}+\mathcal{P}^{\rm res}=\alpha N_\alpha \mathcal{E}_L+\mathcal{P}^{\rm res},$$

is the sum of a background polarization, linear in the local field, that is due to the host material and a nonlinear polarization that is due to the resonant systems. Fields are represented throughout by envelope functions which can be defined implicitly by $P=\frac{1}{2}(\mathcal{P}e^{-i\omega t}+\text{c.c.})$, $E_L=\frac{1}{2}(\mathcal{E}_L e^{-i\omega t}+\text{c.c.})$, etc. In addition, α and N_α are the linear polarizability and the number density of linear systems, respectively, of the isotropic homogeneous host material. (Inhomogeneity of the host material due to the resonant systems is neglected for simplicity.) Then, using the Lorentz local-field condition to eliminate the microscopic local field $\mathcal{E}_L=\mathcal{E}+(4\pi/3)\mathcal{P}$ in favor of the macroscopic Maxwell field $\mathcal{E}$ and polarization $\mathcal{P}$ and using the Clausius-Mossotti-Lorentz-Lorenz relation $(4\pi/3)\alpha N_\alpha=(\varepsilon-1)/(\varepsilon+2)$ to eliminate the microscopic polarizability in favor of the macroscopic dielectric function $\varepsilon=n^2$, where n is the linear index of refraction of the host material, one obtains

$$\mathcal{P}=\frac{\varepsilon-1}{4\pi}\mathcal{E}+\frac{\varepsilon+2}{3}\mathcal{P}^{\rm res}. \tag{1}$$

The dynamics of the two-level systems are described by the generalized Bloch equations in the rotating-wave approximation [4]:

$$\frac{\partial R_{21}}{\partial t}=i\Delta R_{21}-\frac{i\mu}{2\hbar}\left(\mathcal{E}+\frac{4\pi}{3}\mathcal{P}\right)W-\gamma_{\perp}R_{21}, \tag{2a}$$

$$\frac{\partial W}{\partial t}=-\frac{i\mu}{\hbar}\left[\left(\mathcal{E}^*+\frac{4\pi}{3}\mathcal{P}^*\right)R_{21}-\left(\mathcal{E}+\frac{4\pi}{3}\mathcal{P}\right)R_{21}^*\right]-\gamma_{\parallel}(W-W_{eq}). \tag{2b}$$

The macroscopic, spatially averaged, atomic variables in the rotating frame of reference are $R_{21}=\langle\rho_{21}e^{i\omega t}\rangle_{sp}$, $R_{12}=\langle\rho_{12}e^{-i\omega t}\rangle_{sp}$, and $W=R_{22}-R_{11}=\langle\rho_{22}\rangle_{sp}-\langle\rho_{11}\rangle_{sp}$. Here, $\langle\cdots\rangle_{sp}$ corresponds to a spatial average over a volume of the order of a resonance wavelength cubed and the ρ_{ij} are the density matrix elements for a two-level system with a lower state $|1\rangle$ and an upper state $|2\rangle$. In addition, μ is the transition dipole moment, $\Delta=\omega-\omega_0$ is the detuning from resonance, $\gamma_{\perp}$ is the dipole dephasing rate, $\gamma_{\parallel}$ is the population relaxation rate, and W_{eq} is the population difference at equilibrium.

Although impurities in solids are typically inhomogeneously broadened, we begin with the more familiar case of homogeneous broadening. For homogeneously broadened two-level atoms, the contribution to the polarization envelope is $\mathcal{P}^{res}=2N\mu R_{21}$. Then, using Eq. (1) to eliminate the polarization, the generalized macroscopic Bloch equations become

$$\frac{\partial R_{21}}{\partial t}=i(\Delta-\ell\epsilon W)R_{21}-\frac{i\mu}{2\hbar}\ell\mathcal{E}W-\gamma_{\perp}R_{21}, \tag{3a}$$

$$\frac{\partial W}{\partial t}=-\frac{i\mu}{\hbar}(\ell^*\mathcal{E}^*R_{21}-\ell\mathcal{E}R_{21}^*)-2i(\ell^*-\ell)\epsilon|R_{21}|^2-\gamma_{\parallel}(W-W_{eq}), \tag{3b}$$

where $\epsilon=4\pi N\mu^2/3\hbar$ is the NDD interaction parameter, N is the number density of two-level systems, and $\ell=(\varepsilon+2)/3$ is the local-field enhancement factor arising from the elimination of the total polarization using Eq. (1).

For dilute concentrations of resonant atoms ($\epsilon\equiv 0$) embedded in a linear dielectric, it is convenient to eliminate the explicit dependence on ℓ from the equations of motion by renormalizing the dipole moment to $\ell\mu$ [13]. However, renormalization of the dipole moment fails to eliminate the explicit dependence on ℓ in dense media because the NDD interaction term is proportional to $\ell\mu^2$. Although ϵ could be renormalized as well, it is not an independent parameter and inconsistencies can arise, for example, when the generalized Bloch equations are coupled to the Maxwell wave equation. Therefore, we retain the explicit presentation of the enhancement factors and, since the dielectric function is, in general, complex, recast Eqs. (3) in terms of the real and imaginary parts of the enhancement factor $\ell=\ell'+i\ell''$ to obtain

$$\frac{\partial R_{21}}{\partial t}=i(\Delta-\ell'\epsilon W)R_{21}-\frac{i\mu}{2\hbar}\ell\mathcal{E}W+\ell''\epsilon WR_{21}-\gamma_{\perp}R_{21}, \tag{4a}$$

$$\frac{\partial W}{\partial t}=-\frac{i\mu}{\hbar}(\ell^*\mathcal{E}^*R_{21}-\ell\mathcal{E}R_{21}^*)-4\ell''\epsilon|R_{21}|^2-\gamma_{\parallel}(W-W_{eq}). \tag{4b}$$

Equations (4) display several local-field enhancement effects. (i) As in the case of a dilute concentration of two-level atoms in a host dielectric, there is an apparent enhancement of the magnitude and an overall phase shift of the field that drives the atoms. (ii) The inversion-dependent detuning (nonlinear Lorentz frequency shift) that is due to the NDD interaction is intensified by the real part of the local-field enhancement factor. (iii) The damping rates $\gamma_{\perp}$ and $\gamma_{\parallel}$ are taken as phenomenological material parameters. (iv) Cooperative decay terms appear due to the interaction of near dipoles mediated by the imaginary component of the dielectric function of the host material.

Although ℓ appears as a coefficient of $\mathcal{E}$ in the generalized Bloch equations, the field that drives the atoms is not necessarily enhanced by the presence of the dielectric. The basis of the argument is the fact that a field, incident on a dispersionless dielectric, is reduced by the factor ℓ at the vacuum/dielectric interface as a consequence of local-field effects described by the Ewald-Oseen extinction theorem [14]. Then, in the limiting case in which the density and dipole moment of the embedded atoms are sufficiently small that nonlinear propagation effects are negligible, the enhancement and reduction are offsetting and the ''enhanced'' field $\ell\mathcal{E}$ in the dielectric ($\ell>1$) is the same as the field incident from the vacuum, if reflections at the boundary are neglected. In the general case of a dielectric containing two-level atoms, the extinction theorem and reflections become nonlinear [15,16] and, consequently, quantitative calculations must include propagation with appropriate initial and boundary conditions. The point to be made is that, based on the limiting case, the inversion-dependent detuning is enhanced relative to the Rabi frequency $\Omega=\mu\ell\mathcal{E}/\hbar$, as well as to the detuning and dephasing, even though ℓ appears as a coefficient of $\mathcal{E}$ in the generalized Bloch equations.

Because NDD effects are only important at sufficiently high densities and large oscillator strengths, it is significant that the local-field enhancement factor increases the effect of the inversion-dependent detuning in dense media. For example, Friedberg, Hartmann, and Manassah have derived a threshold condition for intrinsic optical bistability in a dense vapor, namely $\epsilon>4\gamma_{\perp}$ [5]. This density threshold condition cannot be controlled in a dense vapor of two-level systems because the dephasing rate increases in direct proportion to the density due to collisional broadening [5]. For two-level systems in a dispersionless dielectric, the threshold condition becomes $\ell\epsilon>4\gamma_{\perp}$ and the threshold condition can be satisfied by a smaller value of ϵ, because (i) the inversion-dependent detuning is enhanced relative to the dephasing rate and (ii) in condensed matter, particularly at cryogenic temperatures, the homogeneous linewidth is no longer restricted to the formula for a collisionally broadened vapor. Significantly, it is in a similar case that intrinsic optical bistability, analyzable by a two-level model, was observed experimentally [1].

A linear dielectric which is dispersive must also be absorptive in accordance with the Kramers-Kronig relations.

This is manifested as a complex dielectric function, causing the local-field enhancement factor to be complex, as well. The imaginary part of the enhancement factor appears as a coefficient, along with the NDD parameter, of bilinear products of the macroscopic atomic variables in two terms of Eqs. (4). These terms are part of the local response and correspond to local cooperative decay effects representing the interaction of near dipoles mediated by the imaginary component of the dielectric function of the host medium. Although the local cooperative terms in the macroscopic Bloch equations, Eqs. (4), have the same dependence on the atomic variables as for cooperative effects in extended systems, which, for example, in thin films containing dilute concentrations of resonant atoms arise from the elimination of the propagating field in the mean-field approximation [8,9], these terms are intrinsic and are not dependent on propagation or on the sample size as in the case for superfluorescence or superradiance. In order to study propagation effects, the equations of motion for the local response, including the local cooperative decay terms, must be coupled to the wave equation. Any cooperative decay effects that are related to nonlinear propagation are, therefore, in addition to the local cooperative decay effects described here. Because the slowly varying envelope approximation is not generally valid in dense media [3] or in dispersive dielectrics, nonlinear propagation effects are beyond the scope of the present paper, but will be treated in a future publication.

The imaginary part of the local-field enhancement factor is given by $\ell''=2\eta\kappa/3$ in terms of the real and imaginary components of the index of refraction $n=\eta+i\kappa$. The imaginary part of the index of refraction, the extinction coefficient κ, is typically associated with the absorption of a field that is propagating through a linear dielectric. The field is reduced by $1/e$ upon propagating one skin depth, $1/4\pi\kappa$ vacuum wavelengths. In dense media, the effects of NDD interactions can be manifested in films that are significantly thinner than a vacuum wavelength [3]. Consequently, the detrimental effects, absorption and heating, associated with an extinction coefficient can be mitigated for dense media. Then, for a thin film of a strongly dispersive dielectric containing a dense collection of two-level atoms, one can expect local cooperative decay effects to play a significant role in the dynamics.

Next, we solve the generalized Bloch equations in the steady-state limit in order to provide a specific example of the effects of the complex local-field enhancement factor on the dynamics. From Eq. (4a), we obtain the steady-state result for R_{21}, namely

$$R_{21}=\frac{\mu\ell\mathcal{E}W/2\hbar}{\Delta-\ell\epsilon W+i\gamma_\perp}=\frac{\mu\ell\mathcal{E}W/2\hbar}{\Delta-\ell'\epsilon W+i(\gamma_\perp+\ell'')}. \quad (5)$$

Then, in the weak-field limit, $W=-1$, the susceptibility is

$$\chi=\frac{\mathcal{P}}{\mathcal{E}}=\frac{\varepsilon-1}{4\pi}+\frac{3}{4\pi}\frac{\ell^2\epsilon}{\omega_0-\ell'\epsilon-\omega-i(\gamma_\perp+\ell'')}$$

and we find that the Lorentz redshift is increased by the real part of the local-field enhancement factor, while the imaginary part contributes to the linewidth. Combining Eqs. (4b) and (5) yields a cubic equation for the inversion

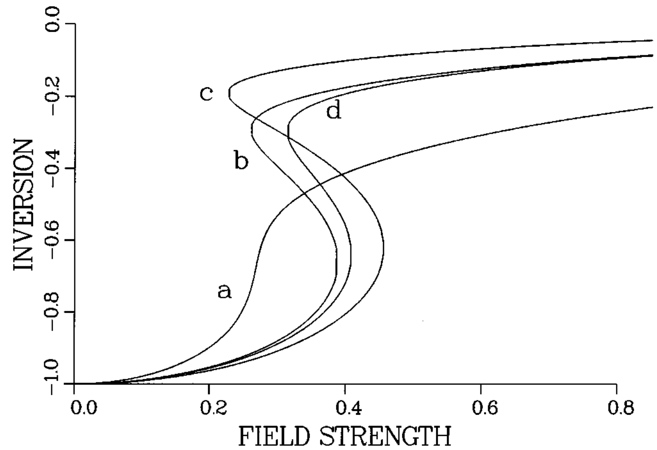

FIG. 1. Inversion as a function of normalized field strength $\mu\mathcal{E}/\hbar\gamma_\perp$ for (a) $\ell=1$, (b) $\ell=2$, (c) $\ell=3$, and (d) $\ell=2+0.2i$. Also, $\Delta=-2\gamma_\perp$, $\gamma_\parallel=0.1\gamma_\perp$, $\epsilon=4\gamma_\perp$, and $W_{\rm eq}=-1$.

$$|\ell|^2\epsilon^2W^3+(-|\ell|^2\epsilon^2W_{\rm eq}-2\ell'\epsilon\Delta-2\epsilon\ell''\gamma_\perp)W^2 + \left(\Delta^2+\gamma_\perp^2+|\ell|^2|\mathcal{E}|^2\frac{\mu^2\gamma_\perp}{\hbar^2\gamma_\parallel}+2\ell'\epsilon\Delta W_{\rm eq} + 2\ell''\gamma_\perp\epsilon W_{\rm eq}\right)W-W_{\rm eq}(\Delta^2-\gamma_\perp^2)=0, \quad (6)$$

in the steady-state limit. Figure 1 shows the roots of Eq. (6) as a function of the field for the cases of (i) no enhancement, $\ell=1$, (ii) real enhancement, $\ell=2$ and $\ell=3$, and (iii) complex enhancement, $\ell=2+0.2i$. The enhanced inversion-dependent detuning, curve *b*, is sufficiently strong to satisfy the density threshold condition for intrinsic optical bistability while the unenhanced NDD interaction, curve *a*, is not. If the inversion-dependent detuning is further enhanced, curve *c*, the bistability region widens, increasing the dynamical range $\Delta\mathcal{E}/\mathcal{E}$. Curve *d* shows that the local cooperative decay terms affect the bistability curve by shifting and compressing the bistability region slightly, as do cooperative effects in extended systems [9]. In addition, large cooperative decay terms can result in an increased hysteresis area, although the dynamical range is reduced [9].

For inhomogeneous broadening, the generalized Bloch equations become

$$\frac{\partial\langle R_{21}\rangle}{\partial t}=\langle i\Delta R_{21}\rangle-i\ell\epsilon\langle W\rangle\langle R_{21}\rangle-\frac{i\mu}{2\hbar}\ell\mathcal{E}\langle W\rangle-\gamma_\perp\langle R_{21}\rangle,$$

$$\frac{\partial\langle W\rangle}{\partial t}=-\frac{i\mu}{\hbar}(\ell^*\mathcal{E}^*\langle R_{21}\rangle-\ell\mathcal{E}\langle R_{21}^*\rangle) -2i(\ell^*-\ell)\epsilon\langle|R_{21}|^2\rangle-\gamma_\parallel(\langle W\rangle-W_{\rm eq})$$

in terms of averages, denoted by angle brackets, over the resonance frequencies of the atoms. The local-field enhancement factors enter into the equations of motion in the same way for both the homogeneously and inhomogeneously broadened cases, however the analysis is considerably more complicated for inhomogeneous broadening due to the presence of the frequency-averaged term $\langle i\Delta R_{21}\rangle$. In some circumstances, perturbative techniques can be used to reduce, by the introduction of new dynamical variables, the integro-differential Bloch equations for an inhomogeneously broadened system to a hierarchy of coupled differential equations [17].

In summary, we have described the derivation of self-consistent semiclassical equations of motion for a dense collection of resonant systems embedded in a linear dielectric. We found that the NDD interaction of dense collections of two-level systems is enhanced by the presence of the host material, lowering the threshold density and increasing the hysteresis area for intrinsic optical bistability. We showed that renormalization of the dipole moment, which is sufficient to account for local-field effects for a collection of tenuous atoms in a linear dielectric host, does not eliminate the local-field enhancement factor when applied to a dense collection of atoms. In addition, the imaginary component of the dielectric function of the host material was found to induce intrinsic quantum coherences. These coherences are manifested as local cooperative terms which have the same dependence on the atomic variables as nonlocal superradiation terms arising from the elimination of the propagating field in the mean-field approximation. Unlike superradiation terms, the local cooperative terms are independent of propagation or sample size, and must be included with the local response when the generalized Bloch equations are coupled to the Maxwell wave equation. The results presented here for two-level atoms have obvious extensions to dense collections of multilevel systems [10] embedded in a linear dielectric medium, for example, enhancement factors and local cooperative terms are obtained for the three-level system case. Finally, these results are indicative of the novel quantum coherence effects that can be expected in other dense multicomponent materials, such as two species of two-level atoms [18]. These effects, as well as effects on spontaneous emission [19], will be treated in future publications.

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