Near Dipole-Dipole Effects in Lasing without Inversion: An Enhancement of Gain and Absorptionless Index of Refraction

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We include the effect of density-dependent near dipole-dipole interactions in order to generalize the theory of a simple three-level system that exhibits lasing without inversion and an enhanced index of refraction at zero absorption. For certain values of atomic density, our generalized theory predicts a novel enhancement of inversionless gain and absorptionless index by more than 2 orders of magnitude.

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It is well known that, for static fields, the local microscopic electric field $\mathbf{E}_L$ that couples to the atomic dipole moment $\mathbf{p}$ is related to the macroscopic field $\mathbf{E}$ and volume polarization $\mathbf{P}$, in a dense dielectric medium, by the Lorentz-Lorenz (LL) relation [1], namely,

$$\mathbf{E}_L = \mathbf{E} + \mathbf{P}/3\epsilon_0 . \tag{1}$$

For dense media, this relation leads to a Clausius-Mossotti relationship for the susceptibility $\chi = \alpha N/(1-\alpha N/3\epsilon_0 N)$ that implies a *nonlinear* relationship between the microscopic molecular polarizability α and the macroscopic susceptibility χ, for a material volume density N. The LL relation (1) also holds for dynamic fields in a linear medium [2], and in fact—as shown conclusively by the present authors—holds for arbitrary time-dependent fields in a nonlinear medium [3]. What this implies is that all of quantum optics that uses an external electric field rather than the local field is internally inconsistent and will yield only approximate results for an optically dense medium. Such an observation had led to the discovery of a large number of new effects such as intrinsic optical bistability [4], self-phase modulation in self-induced transparency [5], linear and nonlinear spectral shifts [6], propagational effects in nonlinear media [7], novel inversion and ultrafast optical switching effects [8], and statistical effects in superfluorescence and amplified spontaneous emission [9]. The current theory of lasing without inversion is not immune, either, to the inconsistency of not using the local field. We show now an example of how the theory must be generated for optically dense media. In the present work we will consider the effect of the near dipole-dipole (NDD) interaction, Eq. (1), on a simple three-level Λ system known to exhibit lasing without inversion, and a high index of refraction at zero absorption.

It is known that the principles of atomic coherence and quantum interference can be used to construct a three-or-more-level atomic system that has nonabsorbing resonances [10] and lasing without inversion [11]. In a collection of such systems, a small but nonzero percentage of atoms in the excited state yields an absorption curve that vanishes at points where the real part of the susceptibility is nonzero—suggesting the possibility of a high refractivity in a nonabsorbing medium [12]. In all this work, however, there has not been an account taken for the effects of the NDD interaction, Eq. (1), that would be of great importance when the medium is sufficiently dense [3]. We rectify this situation now by considering the effect of the NDD contributions to lasing without inversion and enhanced index in a simple three-level system considered by Scully and co-workers [13].

The three-level scheme is depicted in the upper left inset of Fig. 1. The idea is to have an upper level a that is relatively far above the two lower levels, b and b'. In practice, lasing without inversion at optical frequencies would occur between the a and b,b' levels, while coherences are induced, say, by microwaves, at the longer wavelength b to b' transition. The $a \to b,b'$ transition is assumed to be pumped by a plane wave light beam of electric field strength E_0 and circular frequency ω. We assume that the atoms are prepared in a coherent superposition of the two lower levels b and b', yielding an initial density matrix for each atom as

$$\hat{\rho}_{\text{initial}} = \begin{pmatrix} \rho^0_{aa} & 0 & 0 \\ 0 & \rho^0_{b'b'} & \rho^0_{b'b} \\ 0 & \rho^0_{bb'} & \rho^0_{bb} \end{pmatrix} . \tag{2}$$

We assume further that the atoms are injected into the electric field interaction region at a rate r.

Let us assume that $\hbar\omega_{\alpha\beta}$ is the energy difference between levels α and β where $\alpha,\beta \in \{a,b,b'\}$. We then may define three frequency shifts of interest as $\Delta_{\alpha\beta} \equiv \omega_{\alpha\beta} - \omega$, with $\beta \in \{b,b'\}$, and $\Delta \equiv (\Delta_{ab} + \Delta_{ab'})/2$ with the condition $\Delta_{ab} - \Delta_{ab'} = \omega_{b'b}$. Let us assume that γ_a and $\gamma_b = \gamma_{b'} \equiv \gamma$ are the depopulation rates of levels a and b,b', respectively. We shall also assume, as do Scully and co-workers [13], that $\omega_{b'b} = \gamma$, and that $\rho^0_{bb} = \rho^0_{b'b'} = |\rho^0_{b'b}| = \rho^0$, where the phase φ of $\rho^0_{b'b}$ is fixed by the relation $\mu_{ab'}\mu_{ab} \times \rho^0_{b'b} e^{-i\pi/4} \equiv \mu^2 |\rho^0_{b'b}| e^{i\varphi}$. Here, $\mu_{ab'}$ and μ_{ab} are the complex dipole matrix elements with $|\mu_{ab'}| = |\mu_{ab}| \equiv \mu$.

With these definitions and assumptions in mind, we define the unitless parameters $\delta \equiv 2\Delta/\gamma$, $\delta_\pm \equiv \delta \pm 1$, $R \equiv \rho^0_{aa}/\rho^0$, $\Gamma \equiv \gamma_a/\gamma$, and $\Gamma_+ \equiv \Gamma + 1$. With all this notation, the Heisenberg equation of motion for the density matrix, $\dot{\hat{\rho}} = i\hbar[\hat{\rho}, H]$, in component form becomes

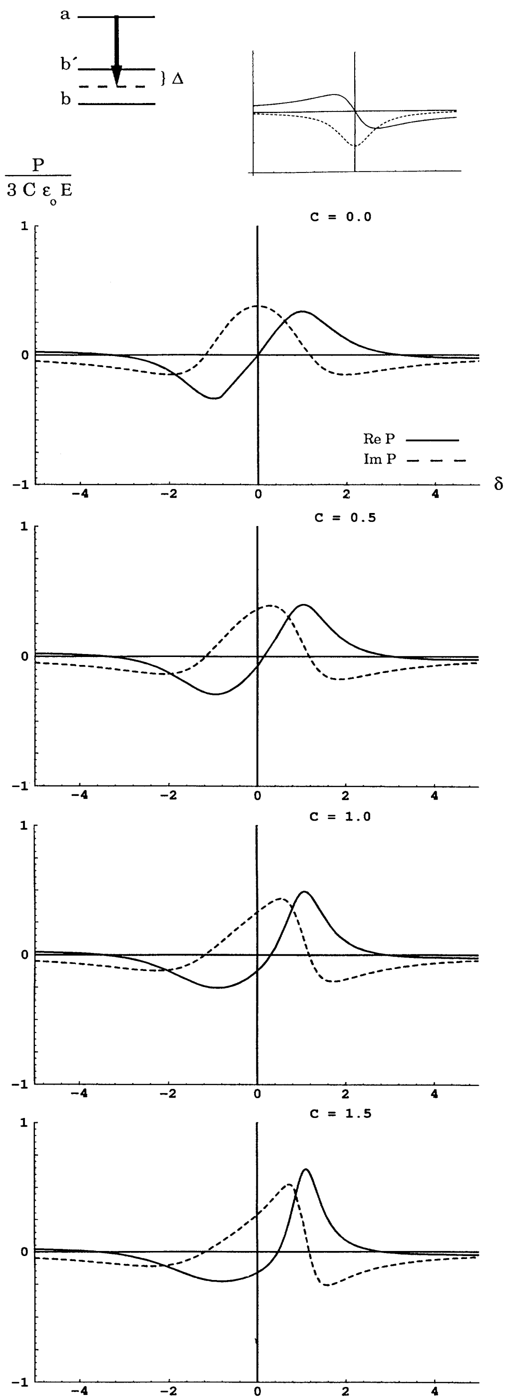

FIG. 1. The effect of small increases in the NDD parameter C on the dispersion $\mathrm{Re}\{P\}$ and absorption $\mathrm{Im}\{P\}$ curves. Noting that $\mathrm{Im}\{P\} > 0$ implies gain, we see for $C=0.0$ the usual curves indicating inversionless gain and two points of absorptionless dispersion. As we increase C there is at first only a central frequency shift to the right as well as a modest distortion of the curves. Top left inset: The three-level system in use. Top right inset: Dispersion and absorption curves for an equivalent two-level system showing no inversionless gain or absorptionless dispersion.

$$\dot\rho_{ab} = -\frac{\gamma}{2}(\Gamma_+ + i\delta_+)\rho_{ab} - \frac{iE_L}{\hbar}[\mu_{ab}(\rho_{bb}-\rho_{aa}) - \mu_{ab'}\rho_{b'b}]\,, \tag{3a}$$

$$\dot\rho_{ab'} = -\frac{\gamma}{2}(\Gamma_+ + i\delta_-)\rho_{ab'} - \frac{iE_L}{\hbar}[\mu_{ab'}(\rho_{b'b'}-\rho_{aa}) - \mu_{ab}\rho_{bb'}]\,, \tag{3b}$$

$$\dot\rho_{aa} = r\rho^0_{aa} - \gamma_a\rho_{aa} + \frac{i}{\hbar}[(\mu^*_{ab}\rho_{ab} + \mu^*_{ab'}\rho_{ab'})E_L^* - \text{c.c.}]\,, \tag{3c}$$

$$\dot\rho_{bb} = r\rho^0_{bb} - \gamma\rho_{bb} - \frac{i}{\hbar}(\mu_{ab}\rho_{ab}E_L^* - \text{c.c.})\,, \tag{3d}$$

$$\dot\rho_{b'b'} = r\rho^0_{b'b'} - \gamma\rho_{b'b'} - \frac{i}{\hbar}(\mu_{ab'}\rho_{ab'}E_L^* - \text{c.c.})\,, \tag{3e}$$

$$\dot\rho_{b'b} = r\rho^0_{b'b} - \sqrt{2}\gamma e^{i\pi/4}\rho_{b'b} - \frac{i}{\hbar}(\mu^*_{ab'}\rho_{ab}E_L^* - \mu_{ab}\rho^*_{ab'}E_L)\,. \tag{3f}$$

To first order in E_L, we may neglect products of the form $\rho_{\alpha\beta}E_L$ for the off-diagonal terms, $\alpha\neq\beta$. Then, in steady state, we find

$$\rho_{ab} = \frac{2i\mu E_L}{\hbar\gamma}\left(\frac{r\rho^0}{\gamma}\right)\frac{(R/\Gamma-1)-e^{i\varphi}/\sqrt{2}}{\Gamma_+ + i\delta_+} \equiv iAE_L G(\delta)\,, \tag{4a}$$

$$\rho_{ab'} = \frac{2i\mu E_L}{\hbar\gamma}\frac{r\rho^0}{\gamma}\frac{(R/\Gamma-1)-e^{-i\varphi}/\sqrt{2}}{\Gamma_+ + i\delta_-} \equiv iAE_L G'(\delta)\,, \tag{4b}$$

and

$$\rho_{aa} = \frac{r\rho^0_{aa}}{\gamma_a} = \frac{r\rho^0}{\gamma}\left[\left(\frac{\rho^0_{aa}}{\rho^0}\right)\Big/\left(\frac{\gamma_a}{\gamma}\right)\right] \equiv \frac{r\rho^0}{\gamma}[R/\Gamma]\,,$$

$\rho_{bb} = r\rho^0/\gamma$, $\rho_{b'b'} = r\rho^0/\gamma$, $\rho_{b'b} = (r\rho^0/\gamma)e^{i\varphi}/\sqrt{2}$, where we have introduced a constant $A\equiv(2\mu/\hbar\gamma)r\rho^0/\gamma$ that has units of inverse E, and also two unitless, complex functions $G(\delta)$ and $G'(\delta)$, defined by

$$\begin{Bmatrix} G(\delta) \\ G'(\delta) \end{Bmatrix} \equiv \frac{R/\Gamma - 1 - e^{\pm i\varphi}}{\Gamma_+ + i\delta_\pm}\,,$$

where the top and bottom terms in braces correspond to plus and minus signs, respectively.

We are now ready to include the effect of the NDD relation, Eq. (1). Let us express the macroscopic polarization P in terms of the macroscopic variables ρ_{ab} and $\rho_{ab'}$,

$$\begin{aligned} P &= \mu(\rho_{ab}+\rho_{ab'}) = i\mu AE_L[G(\delta)+G'(\delta)] \\ &= i\mu A\{E + P/3\epsilon_0\}[G(\delta)+G'(\delta)]\,. \end{aligned} \tag{5}$$

When Eq. (5) is now solved for the polarization, the novel presence of P on the right-hand side of this equation leads to a nonlinear equation of the Clausius-Mossotti sort, namely,

$$P(\delta) = 3C\epsilon_0 E_0 \frac{iG_+(\delta)}{1 - iCG_+(\delta)}\,, \tag{6}$$

where the unitless NDD constant C is defined by

$$C = 2\frac{\mu^2/3\epsilon_0}{\hbar\gamma}\left(\frac{r\rho^0}{\gamma}\right) = \frac{2\mu^2\mathcal{N}/3\epsilon_0}{\hbar\gamma} = \frac{2\varepsilon}{\gamma}\,,$$

and $G_+(\delta) \equiv G(\delta) + G'(\delta)$. Here, $\mathcal{N} = r\rho^0/\gamma$ is an effective volume density, and $\varepsilon \equiv \mu^2\mathcal{N}/3\hbar\epsilon_0$, with units of frequency, is the standard notation for the NDD parameter that is a measure of the strength of the NDD interaction, as used in previous works [3]. We assume that γ is the radiative decay rate out of levels b and b' and is hence independent of the density. The dispersion and absorption functions are proportional to $\mathrm{Re}\{P\}$ and $\mathrm{Im}\{P\}$, respectively,

$$\mathrm{Re}\{P\} = 3C\epsilon_0 E_0 \frac{\mathrm{Re}\{iG_+(\delta)\} - C|G_+(\delta)|^2}{|1 - iCG_+(\delta)|^2}\,, \tag{7a}$$

$$\mathrm{Im}\{P\} = 3C\epsilon_0 E_0 \frac{\mathrm{Im}\{iG_+(\delta)\}}{|1 - iCG_+(\delta)|^2}\,, \tag{7b}$$

where we note that only the real part contains the *difference* of two terms in the numerator, with the first term proportional to C and the second to C^2.

To reproduce exactly previous results [13], we take the population ratio as $R \equiv \rho_{aa}^0/\rho^0 = 0.02$, the decay ratio as $\Gamma \equiv \gamma_a/\gamma = 0.05$, the phase $\varphi = 5\pi/4$, and of course, the NDD parameter as $C = 0.0$ in Eqs. (7). The result is shown in Fig. 1 and exhibits the usual gain and absorptionless points in the dispersion for this system that is clearly noninverted, since $R = 0.02$. (We plot P for an equivalent two-level system in a top right inset in Fig. 1 to show that there is no inversionless gain or absorptionless dispersion there [14].) Now, as shown in Fig. 1, we slowly crank up the NDD constant, $C = 0.0$, 0.5, 1.0, and 1.5, and we see that the effect is to shift the central frequency to the right and to distort both the absorption and dispersion relations, while leaving the inversionless gain and absorptionless index more or less intact. This behavior maintains as we sweep C over larger values, except for a sharp transition near $C = 3.0$. As depicted in Fig. 2, we now focus in on this region, taking the NDD constant over a small range, $C = 3.05$, 3.10, 3.15, and 3.20. Notice first that we have enlarged the vertical scale so we can see that there is a remarkable increase of about a factor of 75 in both inversionless gain and absorptionless index in the area of $C = 3.10$. This is an enhancement of *2 orders of magnitude* over the original $C = 0.0$ case without the NDD effect. Moreover, near this value we can see that a threshold in C has been crossed that changes the sign of the dispersion $\mathrm{Re}\{P\}$ but not the absorption $\mathrm{Im}\{P\}$. The reason for this can easily be seen by considering Eq. (7). The numerator of $\mathrm{Re}\{P\}$, Eq. (7a), is the difference of two terms that describe a family of functions of δ

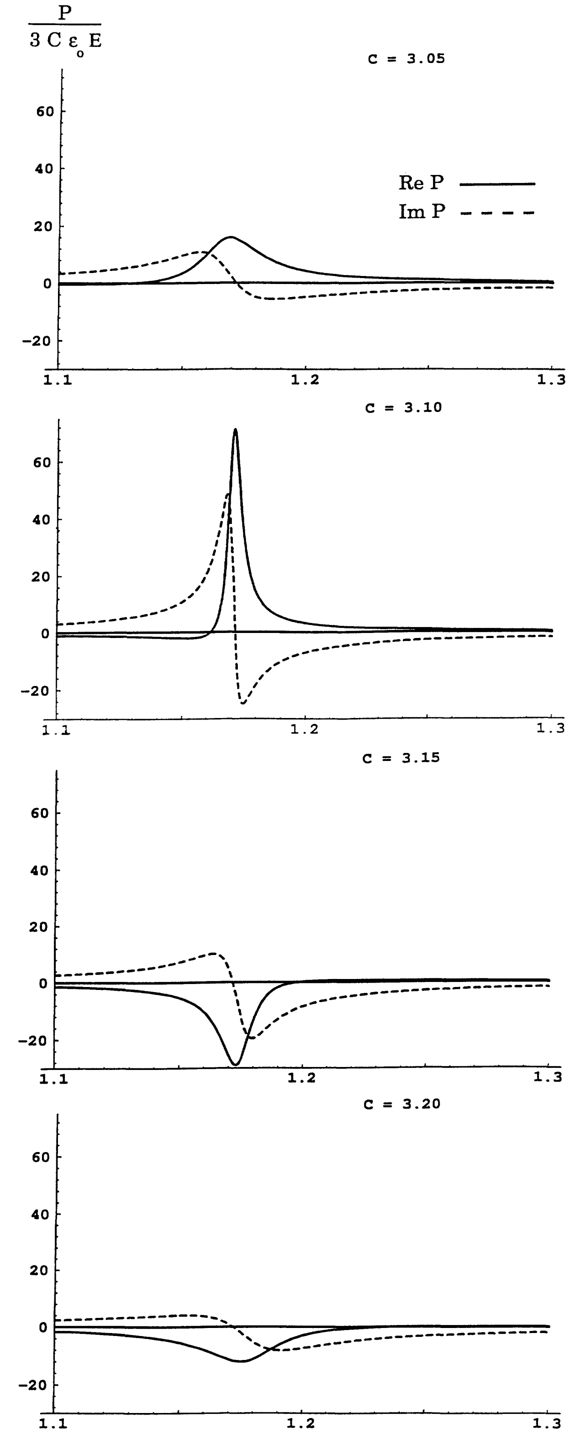

FIG. 2. As we sweep through a small range of values of the NDD parameter C near the threshold near 3.0 we see a huge enhancement of inversionless gain and absorptionless dispersion by 2 orders of magnitude over the $C = 0.0$ case in Fig. 1. We also notice an interchange of odd and even symmetry of $\mathrm{Re}\{P\}$ and $\mathrm{Im}\{P\}$ as compared to $C \cong 0$, Fig. 1, and an overall sign change in $\mathrm{Re}\{P\}$ near $C \approx 3.10$ with no corresponding sign change in $\mathrm{Im}\{P\}$.

parametrized by the constant C. If C becomes large enough, the amplitude of the function changes sign, flipping the curve. This does not occur for the absorption $\mathrm{Im}\{P\}$, Eq. (7b), because there is only a single C-independent term in the numerator, and hence no flip mechanism. Another interesting feature to notice in Fig. 2 is that the absorption curve—originally symmetric in Fig. 1—now is almost antisymmetric, while the reverse has occurred for the dispersion curve. In some sense, the NDD correction interchanges the functional form of the two curves.

The above discussion may be cast into a form using the injection rate r rather than the unitless parameter C. For an order of magnitude estimate, let us choose atomic parameters: $\mu \sim 10^{-29}$ cm and $\gamma \sim 10^{10}\ \mathrm{s}^{-1}$. For local field effects to play a role, we need many atoms per cubic wavelength λ injected with the b,b' coherence. Taking $\lambda = 300$ nm and "many" as a hundred, then $\rho^0 \sim 10^{21}$ m^{-3}. Inserting these values into the expression for C, we obtain $r = C \times 10^{12}\ \mathrm{s}^{-1}$, where we recall that C is unitless. Hence, one needs on the order of 10^{12} atoms entering the interaction volume per unit time.

In conclusion, we have included the near dipole-dipole interaction contributions for dense media in the density matrix equations of a simple three-level system that exhibits inversionless lasing and absorptionless index of refraction. For small values of the unitless NDD parameter C we see a frequency shift and a small distortion of the absorption and dispersion curves. However, a threshold effect in the parameter C allows us to choose a value that enhances inversionless gain and absorptionless dispersion by at least 2 orders of magnitude, as long as the increase is not so large that the assumption of a linearization in E_L is violated.

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