LETTER TO THE EDITOR

Rapid refractive index enhancements via laser-mediated collectivity

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Abstract

The collective interaction via the environmental vacuum is investigated for a mixture of two different multi-atom ensembles in a moderately intense laser field. Due to the numerous inter-atomic couplings, the laser-dressed system may react sensitively and rapidly with respect to changes in the atomic and laser parameters. We show for weak probe fields that in the absence of absorption both the index of refraction and the group velocity may be modified strongly and rapidly due to the collectivity.

The presence of strong laser fields is known to substantially modify the absorptive and dispersive properties of atomic samples [1–12]. The numerous effects put forward already for single atoms include the splitting of spectral lines [1, 2], electromagnetically induced transparency [3], lasing without population inversion [4], enhanced indices of refraction [5, 6], stopping of light [7] and ultra-narrow spectral lines [8]. For larger ensembles and not too low densities, collective phenomena were further pointed out to drastically affect laser-driven media [9, 10, 12, 14, 15]. In particular, collectivity due to nonlinearities in a plasma may render the medium transparent [9], while laser-mediated local effects arising from dipole–dipole interactions between atoms may alter the appearance of band gaps in optically dense materials [10] and induce piezophotonic switching [11]. Efficient schemes to control the collective quantum dynamics, in general, were further demonstrated [12] via employing interferences and fast switching schemes among collective atomic dressed states. There is particular interest, however, in the rapid control of dispersive properties of collective systems, e.g. for quantum gates or high precision measurements [13].

In this letter, we show that the mutual interactions of atoms via the quantum fluctuations of the surrounding electromagnetic field (EMF) [16] are suitable to generate transparent media...
with large indices of refraction of order 10 or high dispersion of arbitrary sign. In particular, we point out that the effects may be considerably larger than for the same amount of independent atoms and may be set up on a time scale of $10^{-8}$ s. Furthermore, the group velocity of a weak probe field propagating through the strongly driven two-level medium may be substantially slowed down or strongly accelerated.

For this purpose, we consider an atomic system consisting of two ensembles of two-level atoms, numbered $N_a$ and $N_b$, with densities of order $10^{13} - 10^{14}$ cm$^{-3}$, and with somewhat different transition frequencies, and interacting with a single moderately strong laser field (see figure 1). The corresponding Rabi frequencies are $|2\Omega_a, 2\Omega_b|$ and spontaneous decay of all closely spaced atoms occurs via interaction with a common electromagnetic field reservoir with rates $|2\gamma_a, 2\gamma_b|$ excited states $|2\lambda, |2\mu\rangle$, respectively. In order to treat the atoms uniformly we suppose that $\{L/c, \Omega_{a,b}^{-1} \ll \tau_s\}$, where $L, c$ and $\tau_s$ are the largest dimension of the sample, the light velocity and the collective decay time, respectively. When scanning the composed atomic sample with, say for instance, a dye or diode laser [17], and depending on the resonance condition for each kind of atom and thus the employed laser frequency, generally one or the other atomic species may dominate the final steady-state collective behaviour.

In the conventional mean-field and rotating-wave approximation, the interaction of the laser-driven atomic sample with the surrounding EMF bath is described in a frame rotating with the laser frequency $\omega_L$, via the Hamiltonian:

$$H = H_i + H_0 + H_s,$$

where $H_i = \sum_i \hbar (\omega_i - \omega_L) a_i^\dagger a_i$, $H_0 = \sum_i \hbar \left[ \Delta_i S_i^{(i)} + \Omega_i \left( S_i^{(i)} + S_i^{(i)} \right) \right]$ and $H_s = i \sum_i (\tilde{g}_i \cdot \vec{d}_i) (a_i S_i^{(i)} - a_i^\dagger S_i^{(i)})$. Here the first and the second terms in equation (1) represent the free EMF ($H_i$) and the free atomic plus laser-atom interaction Hamiltonian ($H_0$), respectively. The last term of equation (1) describes the interaction ($H_s$) of the atoms with the environmental vacuum modes ($a_i$). The collective atomic operators $S_i^{(i)} = \sum_{n=1}^N \{2\lambda_{i1}, 2\mu_{i1}\}$, $S_{i}^{(i)} = S_{i}^{(i)}$, $S_{i}^{(i)} = S_{i}^{(i)}$ satisfy the standard commutation relations for quasiparticle operators, i.e., $[S_{i}^{(i)}, S_{j}^{(j)}] = \pm \delta_{ij} S_{i}^{(j)}$, $[S_{i}^{(i)}, S_{j}^{(j)}] = \pm \delta_{ij} S_{i}^{(j)}$, $[(i, j) \in \{a, b\}$]. $\Delta_i = \omega_i - \omega_L$ denote the detuning of the atomic transition frequencies $\omega_i$ to the laser frequency, and $\{\Delta_i, \Omega_i \ll \omega_L\}$, $\tilde{d}_i$ corresponds to the transition dipole matrix elements of the atoms. $\tilde{g}_k = \sqrt{\frac{2\pi}{\hbar}} \hbar \omega_k \vec{e}_k$, where $\vec{e}_k$ is the photon polarization vector while $V$ is the EMF quantization volume.
The master equation corresponding to the Hamiltonian (1) in the Born–Markov approximation then reads
\[
\dot{\rho}(t) + \frac{1}{\hbar}[H_0, \rho] = - \sum_{i,j \in \{a,b\}} \left\{ \sqrt{r_i \gamma_i} [\mathcal{S}^{(i)}_+, \mathcal{S}^{(j)}_- \rho] + \sqrt{r_j \gamma_j} [\mathcal{S}^{(i)}_- \mathcal{S}^{(j)}_+ \rho] \right\} + \text{h.c.} \quad (2)
\]
The diagonal \((i = j)\) contribution of the first term on the right-hand side describes the collective damping due to the spontaneous emission of atoms, while that proportional to \(\sqrt{r_i \gamma_i} \) involves the mutual exchange of photons among the different types of atoms in the sample and is very sensitive relative to the splitting frequency \(\Delta \omega = \omega_a - \omega_b\) with \(\Delta_a = \Delta_b = \Delta \omega\). The dipole–dipole interactions \(\Theta_{\text{dd}}\) among the emitters are omitted here, an approximation valid as soon as \(\Omega_{a,b}/N_{a,b} \gg \Theta_{\text{dd}}\). The last term of the master equation (2) involving the collision rates \(r_i\) accounts for collisional damping of atoms which alter the phase of the atomic state but not its population [15, 18].

In the intense-field limit \((\Omega_i \gg \gamma_i N_i)\), the master equation (2) transforms into the dressed-state picture \((|\Psi_i^{(j)}\rangle\text{ for } i \in \{a, b\}, j \in \{1, 2\})\) via
\[
|1_i\rangle = |\Psi_1^{(i)}\rangle \cos \theta_i + |\Psi_2^{(i)}\rangle \sin \theta_i, \quad |2_i\rangle = -|\Psi_1^{(i)}\rangle \sin \theta_i + |\Psi_2^{(i)}\rangle \cos \theta_i,
\]
with \(\cot 2\theta_i = \Delta_i/2\Omega_i, \{i \in \{a, b\}\}\). In the secular approximation, i.e. upon omission of the terms (in the master equation) oscillating with Rabi frequency \(\hat{\Omega}_{ab} = \sqrt{\Omega_{ab}^2 + (\Delta_{ab}/2)^2}\) and larger, and without the cross-damping contribution \((\gamma_{ab} = \sqrt{\gamma_a \gamma_b} = r_{ab} = \sqrt{r_a r_b} = 0)\), the resulting dressed-state master equation results in the exact steady-state solution of the form
\[
\rho_s = Z^{-1} \prod_{i \in \{a, b\}} e^{-\xi_i R_i^{(i)}}. \quad (4)
\]
Here \(2\xi_i = \ln(|\gamma_i \cos^2 \theta_i + r_i \sin^2(2\theta_i)/4|)/|\gamma_i \sin^4 \theta_i + r_i \sin^2(2\theta_i)/4|\), \(R_i^{(i)} = |\Psi_1^{(i)}||\Psi_2^{(i)}| - |\Psi_2^{(i)}||\Psi_1^{(i)}|, \{i \in \{a, b\}\}\), while \(Z\) is chosen such that \(\text{Tr} \rho_s = 1\).

The influence of the cross-damping terms with respect to the final collective steady-state dynamics can be estimated approximately for larger samples, while for single-atom systems \((N_a = N_b = 1)\) this can be carried out exactly by solving the respective equations of motion for the atomic variables. In the dressed master equation, the sideband contribution proportional to \(\gamma_{ab}\), \(r_{ab}\) oscillates at the relative frequency \(2(\Omega_a - \Omega_b)/\Delta\) which can be nonzero in the general, the atoms are subject to different detunings and laser intensities. If \(\Omega_a - \Omega_b \sim \Omega_{a,b}\) such contributions can be omitted in the secular approximation (as well as for smaller samples), and the solution in equation (4) is then applicable. When \(\Omega_a \approx \Omega_b\) and the strong laser field is detuned far off the frequency range \(0 \ll \Delta \ll \Delta \omega\), i.e. \(\Omega_a - \Omega_b \approx 0\), then equation (4) can be employed with \(\xi_a \approx \xi_b\). Assuming \(\Delta_a = |\Delta_b| =: \Delta \omega/2\), and \(\Omega_a = \Omega_b \gg \Delta \omega/2\), the steady-state solution can further be obtained from equation (4) in the limit \(\xi_{a,b} \rightarrow 0\). In what follows, however, we neglect the cross-damping contributions and thus restrict ourselves to the case \(N\gamma_{a,b} \ll \Delta \omega < \Omega_{a,b}\). Therefore, collective interactions via the environmental vacuum occur between atoms possessing identical transition frequencies, i.e. not different ones coupling with distinct parts of the vacuum.

On employing the atomic coherent states for two-level atoms [19] and the solution in equation (4), we derive the expectation values for any collective atomic correlators of interest. In particular, the steady-state expectation values for the collective dressed-state inversion operators \(\langle R_i^{(i)} \rangle_s\) can be obtained for \(i \in \{a, b\}\) and with \(Z = Z_a Z_b\):
\[
\langle R_i^{(i)} \rangle_s = -\frac{\partial}{\partial \xi_i} \ln Z_i, \quad Z_i = e^{\xi_i N_i} \frac{1 - e^{-2\xi_i (N_i + 1)}}{1 - e^{-2\xi_i}}.
\]
In what follows we concentrate on the case with almost equal parameters for the two types of atoms, i.e. \( \Omega_a \approx \Omega_b \equiv \Omega, \gamma_b \approx \gamma_a(1 - \Delta\omega/\omega_0)^3 \approx \gamma_a \equiv \gamma \), \( r_a \approx r_b \equiv r \) and \( N_a \approx N_b \equiv N \).

Figure 2 depicts the dependence of the dressed-state inversion operators as a function of \( \Delta_\omega/(2\Omega) \). As the atomic transition frequencies for the atoms of types a and b differ from each other, the steady-state collective populations behave differently as well. Note that, due to collectivity, the collective dressed state populations may be transferred abruptly and rapidly from one dressed state to another as the laser detuning \( \Delta_\omega \) changes its sign. A few-atom system is less sensitive relative to the laser detuning in the sense of fast switching. Thus, at this particular point, \( \Delta\omega/(2\Omega) = \pm \varepsilon \) with \( \varepsilon \ll 1 \), one may switch the absorption properties of a weak probe field from positive to negative gain (or vice versa) while the dispersive features are strongly enhanced. For a pencil-shaped sample with length \( L \sim 5\lambda \), transverse area \( S \approx 2\lambda^2 \), \( \lambda \sim 10^{-4} \) cm, \( \gamma \sim 10^7 \) Hz and \( N \sim 10^5 \) we estimate a switching time \( \tau_s \sim 2L/(\lambda\gamma N) \) between the dressed-state populations, \( (R)_{s}/N = \pm 1 \), of about \( 10^{-9} \) s [12, 14, 20]. The secular approximation can be applied here if \( \Omega \sim 10^{10} \) Hz, as \( \Omega \gg \tau_{s}^{-1} \).

We proceed by calculating the refractive properties of a very weak field probing the strongly driven atomic sample. The linear susceptibility \( \chi(\omega) \) of the probe field, at frequency \( \nu \), can be represented in terms of the Fourier transform of the average value of the two-time commutator of the atomic operator as

\[
\chi(\nu) = \frac{i}{\hbar} \sum_{i=\{a,b\}} \frac{\tilde{N}_i d_i^2}{\gamma_i \hbar} \int_0^\infty dr \, e^{i\nu t} \langle [S^{(i)}(t), \tilde{S}^{(i)}(0)] \rangle_{s}.
\]  

(5)

Note that the steady state (subindex \( s \)) of the atomic correlators in equation (5) should be calculated with the help of equations (3), (4).

Introducing equation (3) in equation (5), and making use of both the secular approximation and the quantum regression theorem [18], together with equation (4), the dispersion and absorption features can be described via

\[
\chi'(\Delta_\rho) = \sum_{i=\{a,b\}} \frac{\tilde{N}_i d_i^2 (R^{(i)}_{z})}{\gamma_i \hbar} \frac{\tilde{N}_i}{N_i} \left[ \frac{\cos^4 \theta_i - 2\tilde{\theta}_i}{\tilde{\gamma}_i^2 + (\tilde{\Delta}_i - 2\tilde{\Omega}_i)^2} - \sin^4 \theta_i - \frac{\tilde{\Delta}_i(1) + 2\tilde{\Omega}_i}{\tilde{\gamma}_i^2 + (\tilde{\Delta}_i + 2\tilde{\Omega}_i)^2} \right],
\]

\[
\chi''(\Delta_\rho) = \sum_{i=\{a,b\}} \frac{\tilde{N}_i d_i^2 (R^{(i)}_{z})}{\gamma_i \hbar} \frac{\tilde{N}_i}{N_i} \left[ \frac{\sin^4 \theta_i - \tilde{\gamma}_i}{\tilde{\gamma}_i^2 + (\tilde{\Delta}_i + 2\tilde{\Omega}_i)^2} - \cos^4 \theta_i - \frac{\tilde{\Delta}_i(1)}{\tilde{\gamma}_i^2 + (\tilde{\Delta}_i - 2\tilde{\Omega}_i)^2} \right].
\]

(6)
Here $\tilde{\Omega}_i = \tilde{\Omega}_i / (\gamma_i N_i)$, while $\tilde{N}_i$ is the atomic density. $\tilde{\Delta}_d^{(ij)} = \Delta_d / (\gamma_i N_i) = (v - \omega_a + \Delta_d) / (\gamma_i N_i)$ corresponds to the detuning of the weak probe field frequency with respect to the driving field, while $\tilde{\gamma}_i = (\gamma_c^{(ij)} - \gamma_i^{(ij)}) / (\gamma_i N_i)$ describes the non-diagonal collective damping with $\gamma_c^{(ij)} = \gamma_i [\sin^2(2\theta_i) + \cos^2(2\theta_i) + \sin^2(2\theta_i)] + r_i [\cos^2(2\theta_i) + \sin^2(2\theta_i)] / 2$ and $\gamma_i^{(ij)} = \gamma_i \cos(2\theta_i) |R^{(ij)}_a|$, respectively. Thus, all decay rates of the atomic variables scale linearly with the atomic number in the sample. Therefore, the absorption–refraction properties of a weak field that probes the strongly driven atomic ensembles will modify accordingly, i.e. rapidly. It should further be noted that in equations (6) we have employed the so-called decoupling scheme for symmetrical atomic correlators as valid for $N \gg 1$ [14]. In the absence of collective effects ($\gamma_c^{(ij)} \equiv 0$), equations (6) reduce, as it should, to the correct result for $N_i$ and $N_0$ independent atoms.

On inspecting equations (6) (involving $|R^{(ij)}_a|$), and figure 2(a) one can easily recognize that the susceptibility $\{x', x''\}$ is substantially enhanced via collective effects. Figure 3 depicts the steady-state dependence of the linear susceptibility with respect to the strong laser detunings while keeping fixed probe-field frequencies. Strong gain, strong positive or negative dispersion with zero absorption are then feasible. The interpretation of these results is straightforward via a dressed-state analysis [1, 2]. When $v - \omega_a = -2\Omega$ (see figures 3(a), (b)), the probe field at exact resonance with the dressed-state transition $|\Psi_1^{(a)}| \leftrightarrow |\Psi_2^{(a)}|$. If $\Delta_d / (2\Omega) < 0$ most population is placed in the dressed states $|\Psi_1^{(a)}|$ (see figure 2(a)) and, thus, the probe field is absorbed. Here $\Delta_d / (2\Omega) = 0$ means that $|R^{(a)}_a| \neq 0$ while $|R^{(b)}_a| \neq 0$ and, respectively, the susceptibility is small though nonzero. Note that for a single-type atomic ensemble one can achieve complete transparency at this point. Increasing further $\Delta_d$, i.e. $\Delta_d / (2\Omega) > 0$, the dressed-state population transfers to $|\Psi_1^{(a)}|$ and the probe field is amplified. Thus, the second ensemble contributes here to a strong shift of the susceptibility resulting in zero absorption with large dispersive features. In particular, the index of refraction yield with close to vanishing absorption $n(v) \approx \sqrt{1 + x''(v)}$ [18] which for the sample parameters

![Figure 3](image)

**Figure 3.** The steady-state dependence of the linear susceptibility $\chi$ (in units of $\tilde{N} a^2 / \gamma \hbar$) as well as that of the derivative $(d/dv)\chi'$ (in units of $\tilde{N} a^2 / \gamma \hbar$) as a function of $\Delta_d / (2\Omega)$. The solid and dashed lines correspond to the real and imaginary parts of $\chi$, respectively, while the dotted curve stands for $(d/dv)\chi''$. Here $\tilde{\Omega}_i = \tilde{\Omega}_i / (\gamma_i N_i)$, while $\tilde{N}_i$ is the atomic density. $\tilde{\Delta}_d^{(ij)} = \Delta_d / (\gamma_i N_i) = (v - \omega_a + \Delta_d) / (\gamma_i N_i)$ corresponds to the detuning of the weak probe field frequency with respect to the driving field, while $\tilde{\gamma}_i = (\gamma_c^{(ij)} - \gamma_i^{(ij)}) / (\gamma_i N_i)$ describes the non-diagonal collective damping with $\gamma_c^{(ij)} = \gamma_i [\sin^2(2\theta_i) + \cos^2(2\theta_i) + \sin^2(2\theta_i)] + r_i [\cos^2(2\theta_i) + \sin^2(2\theta_i)] / 2$ and $\gamma_i^{(ij)} = \gamma_i \cos(2\theta_i) |R^{(ij)}_a|$, respectively. Thus, all decay rates of the atomic variables scale linearly with the atomic number in the sample. Therefore, the absorption–refraction properties of a weak field that probes the strongly driven atomic ensembles will modify accordingly, i.e. rapidly. It should further be noted that in equations (6) we have employed the so-called decoupling scheme for symmetrical atomic correlators as valid for $N \gg 1$ [14]. In the absence of collective effects ($\gamma_c^{(ij)} \equiv 0$), equations (6) reduce, as it should, to the correct result for $N_i$ and $N_0$ independent atoms.

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given above takes values larger than $n > 8$ (see figure 3(a) near $\Delta_{\omega}(2\Omega) \approx 10^{-3}$). However, without collective effects, i.e. a noninteracting ensemble of $N_a = N_b = 1000$ atoms, the index of refraction $n$ can be further enhanced by increasing $\lambda^3 N$, but then the atoms would be so close to each other that short-range dipole–dipole interactions need to be taken into account. Moreover, $\chi'$ may be smaller than $-1$ at frequencies where the absorption vanishes meaning that the weak-probe field can no longer propagate through the atomic medium (see figure 3). The switching time of the susceptibilities for the same sample parameters is of the order of $10^{-8}$ s because now $-1 < \langle \dot{R}^{(a)}_{10} \rangle / N < 1$.

Furthermore, once the probe field frequency is adjusted near resonance with the bare-state transition frequencies of atom a or b, one can observe steep dispersive features at $\Delta_{\omega} \in [0, \Delta\omega]$ (see figure 2(b)). However, the magnitude of the susceptibility, in this case, is the same as for a few-atom sample, except the dispersive slopes. These results can be employed to induce a rapid phase shift for a weak field travelling through the strongly driven atomic sample. Note that at the points, where the $\Delta_i$ change the sign, the two-level emitters are in a strong collective phase [15] and, thus, abrupt changes of $\chi'$ are due to strong collectivity. Also, the collisional damping does not affect considerably the collective steady-state behaviour, and its influence can be balanced by increasing the number of atoms. This means that the collisional damping influences small atomic samples while larger atomic systems are less sensitive with regard to kind of phase damping.

We demonstrate further that collections of two-level atoms are suitable for rapidly switching between strongly accelerating or slowing down of a weak probe pulse traversing through driven two-level media. The light group velocity can be estimated from the following expressions: $1/v_g = n_g/c = d(k(v))/dv$, where $k = n(v)v/c$. For $\bar{N}d^2/\gamma \hbar \sim 0.1$, i.e. $\bar{N} \sim 10^{12}$ cm$^{-2}$, $v/\gamma \sim 10^8$, $n_g$ may reach values of the order of $10^7$ of either sign (see figures 3(b), (d) where $\chi'' = 0$). The refractive index may take values below unity at such moderate atomic densities. Thus, by properly choosing the external parameters one can arrive at rather low subluminal or large superluminal group velocities.

In summary we have demonstrated that collective interactions among two-level radiators are suitable to generate highly refractive media with close to GHz switching times to strongly deviating properties. Further, the group velocity of a weak electromagnetic field pulse probing the laser-driven atomic sample may be abruptly altered depending sensitively on the external atomic and laser parameters.

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