Tunable Negative Refraction without Absorption via Electromagnetically Induced Chirality

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We show that negative refraction with minimal absorption can be obtained by means of quantum interference effects similar to electromagnetically induced transparency (EIT). Coupling a magnetic dipole transition coherently with an electric dipole transition leads to electromagnetically induced chirality, which can provide negative refraction without requiring negative permeability and also suppress absorption. This technique allows negative refraction in the optical regime at densities where the magnetic susceptibility is still small and with refraction/absorption ratios that are orders of magnitude larger than those achievable previously. Furthermore, the refractive index can be fine-tuned, which is essential for practical realization of subdiffraction-limit imaging. As with EIT, electromagnetically induced chirality should be applicable to a wide range of systems.

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Negative refraction of electromagnetic radiation [1] is currently a very active area of research, driven by goals such as the development of a "perfect lens" in which imaging resolution is not limited by the wavelength [2]. Despite remarkable recent progress in demonstrating negative refraction using technologies such as metamaterials [3–6] and photonic crystals [7–9], a key challenge remains the realization of negative refraction *without absorption*, which is particularly important in the optical regime [10–14]. Here we propose a promising new approach: the use of quantum interference effects similar to electromagnetically induced transparency (EIT) [15] to suppress absorption and induce chirality [16]. As with EIT, electromagnetically induced chirality should be applicable to a wide range of systems: atoms, molecules, quantum dots, excitons, etc.

Early proposals for negative refraction required media with both negative permittivity and permeability ($\varepsilon, \mu < 0$) in the frequency range of interest. In the optical regime, however, it is difficult to realize negative permeability with low loss, since typical transition magnetic dipole moments (μ_a) are smaller than transition electric dipole moments (d_a) by a factor of the order of the fine structure constant $\alpha \simeq \frac{1}{137}$. As a consequence, magnetic susceptibilities χ_m are much smaller than electric susceptibilities χ_e: $|\chi_m| \sim (\mu_a/d_a)^2|\chi_e| \sim \alpha^2|\chi_e| \simeq (1/137)^2|\chi_e|$, where $\varepsilon = 1 + \chi_e$ and $\mu = 1 + \chi_m$. Recently, an elegant suggestion was made [17,18] to alleviate this problem by using a chiral medium, i.e., a medium in which the electric polarization $\mathbf{P}$ is coupled to the magnetic field $\mathbf{H}$ of an electromagnetic wave and the magnetization $\mathbf{M}$ is coupled to the electric field $\mathbf{E}$:

$$\mathbf{P} = \varepsilon_0 \chi_e \mathbf{E} + \frac{\xi_{EH}}{c}\mathbf{H}, \qquad \mathbf{M} = \frac{\xi_{HE}}{c\mu_0}\mathbf{E} + \chi_m \mathbf{H}. \quad (1)$$

Here ξ_{EH} and ξ_{HE} are the complex chirality coefficients. They lead to additional contributions to the refractive index for one circular polarization:

$$n = \sqrt{\varepsilon\mu - \frac{(\xi_{EH} + \xi_{HE})^2}{4}} + \frac{i}{2}(\xi_{EH} - \xi_{HE}). \quad (2)$$

As Pendry noted [17], such a chiral medium allows $n < 0$ without requiring negative μ if there is a positive imaginary part of $(\xi_{EH} - \xi_{HE})$ of sufficiently large magnitude. For example, choosing the phases of the complex chirality coefficients such that $\xi_{EH} = -\xi_{HE} = i\xi$, with $\xi, \varepsilon, \mu > 0$, the index of refraction becomes $n = \sqrt{\varepsilon\mu} - \xi$, and $n < 0$ when $\xi > \sqrt{\varepsilon\mu}$. Because chirality coefficients can scale as $\xi_{EH}, \xi_{HE} \sim \frac{\mu_a}{d_a}\chi_e \sim \alpha\chi_e$, they suffer only one factor of α suppression, as compared to the challenge of realizing negative permeability with $\chi_m \sim \alpha^2\chi_e$. Nevertheless, the use of a chiral medium to achieve negative refraction in the optical regime still faces the demanding requirement of minimizing loss while realizing $|\xi_{EH}|, |\xi_{HE}| \sim 1$. In the following we describe how quantum interference effects similar to EIT allow this requirement to be met and also enable fine-tuning of the refractive index by means of external fields.

To introduce concepts underlying electromagnetically induced chiral negative refraction, we begin with the simplistic three-level system shown in Fig. 1(a), which we will later modify to a more realistic scheme [see Fig. 1(b)]. As seen in Fig. 1(a), the electric ($\mathbf{E}$) and magnetic ($\mathbf{B}$) components of the probe field couple state $|1\rangle$ to state $|3\rangle$ by an electric dipole ($E1$) transition, and to state $|2\rangle$ by a magnetic dipole ($M1$) transition, respectively. (We neglect the electric quadrupole coupling on the $|1\rangle - |2\rangle$ transition since it contributes insignificantly to induced chirality [19].) There is also a strong resonant coherent field coupling states $|2\rangle$ and $|3\rangle$ with Rabi frequency Ω_c. The parities in this system are $|1\rangle$ even, $|2\rangle$ even, $|3\rangle$ odd or

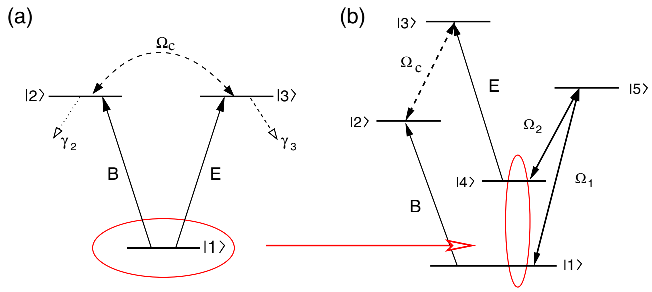

FIG. 1 (color online). (a) Simplistic three-level system for electromagnetically induced chiral negative refraction. (b) Modified level scheme for realistic media. The dark state created by the two-photon resonant Raman coupling of levels $|1\rangle$ and $|4\rangle$ takes over the role of level $|1\rangle$ in (a). In both level schemes, **E**, **B** are the electric, magnetic components of the probe field, which can experience negative refraction.

vice versa. An analogous parity distribution will apply to the more realistic system of Fig. 1(b). Since the $|2\rangle - |1\rangle$ transition is not $E1$, level $|2\rangle$ can be considered metastable with a decay rate $\gamma_2 \sim (\mu_a/d_a)^2\gamma_3 \sim (1/137)^2\gamma_3$.

The scheme of Fig. 1(a) has similarities to resonant nonlinear optics based on EIT [20,21]. For such schemes it is well known that there is destructive interference for the absorption and constructive interference for the dispersive cross coupling between the $|1\rangle - |3\rangle$ and $|1\rangle - |2\rangle$ transitions in the limit $\gamma_2 \ll \gamma_3$ and if the transition $|1\rangle - |2\rangle$ is two-photon resonant with the probe field E and the coupling field Ω_c [15]. A similar setup has been discussed recently by Oktel and Müstecaplioglu [22] to generate negative refraction by means of a large negative μ. However, they did not examine chirality and its effect on the index of refraction, which, as we show below, is the most important contribution.

To enable electromagnetically induced chiral negative refraction in realistic media, we modify the simplistic scheme of Fig. 1(a) to satisfy three criteria: (i) Ω_c must be an ac field, so that its phase can be adjusted to induce chirality; (ii) there must be high-contrast EIT for the probe field; and (iii) the energy level structure must be appropriate for media of interest (atoms, molecules, excitons, etc.). In Fig. 1(b) we show one example of a modified level structure that meets the above criteria. This scheme employs strong coherent Raman coupling by two coherent fields with complex Rabi frequencies Ω_1 and Ω_2 and carrier frequencies ω_1 and ω_2, which creates a dark superposition of states $|1\rangle$ and $|4\rangle$. This dark state takes over the role of the ground state $|1\rangle$ in the three-level scheme of Fig. 1(a), such that the electric component of the probe field has a transition from the dark state to state $|3\rangle$ and the magnetic component a transition to level $|2\rangle$. Therefore, the modified scheme remains effectively three-level (consisting of $|2\rangle$, $|3\rangle$, and the dark state), but with sufficient additional degrees of freedom under experimental control to allow the above three criteria to be satisfied.

The coherent preparation of dark states by external laser fields is a well-established technique realized in many systems. Existence of a dark state requires only two-photon resonance and is largely insensitive to the intensities of the laser fields. In the scheme of Fig. 1(b), coupling of the probe field to a dark state greatly enhances the freedom of choice of energy levels and operating conditions, and thus makes the scheme applicable to realistic systems. The coupling between $|2\rangle$ and $|3\rangle$ is now an ac coupling with carrier frequency ω_c and Rabi frequency Ω_c, which can be chosen to be complex. By varying the phase of Ω_c the phase of ξ_{EH} and ξ_{HE} can be controlled. To ensure degeneracy between the electric and magnetic carrier frequencies, $\omega_1 - \omega_2 = \omega_c$ must hold. We note that the possibility of cross coupling the electric and magnetic field components of a probe field without a common level coupled to both components was recently suggested by Thommen and Mandel [23] for metastable neon with a far-infrared probe field. However, their scheme exploits neither dark states nor chirality and thus the achieved ratio of negative refraction to absorption is small [24]. Finally, since the EIT transition in the level scheme of Fig. 1(b) is predominantly between levels $|4\rangle$ and $|2\rangle$, which can have comparable energies, the two-photon resonance can remain narrow even in the presence of one-photon-transition broadening and thus high-contrast EIT is possible.

For an ensemble of radiators described by the level scheme of Fig. 1(b), we calculate the real and imaginary parts of the refractive index in linear response. States $|2\rangle$ and $|4\rangle$ are assumed to be metastable. To model realistic situations we take into account broadening γ_P of the narrow magnetic transition $|1\rangle - |2\rangle$, which could be due to relaxation other than radiative decay or due to inhomogeneous (e.g., Doppler) broadening. We also require that the $|2\rangle - |4\rangle$ transition, used for high-contrast EIT, has a very narrow linewidth. We solve for the steady-state values of the density matrix elements ρ_{34} and ρ_{21} in a linear approximation in which the probe field components **E** and **B** are assumed to be small variables:

$$\rho_{34} = \alpha_{EE}\mathbf{E} + \alpha_{EH}\mathbf{B}, \qquad \rho_{21} = \alpha_{HE}\mathbf{E} + \alpha_{HH}\mathbf{B}. \quad (3)$$

Here α_{EE} and α_{HH} are the polarizabilities and α_{EH} and α_{HE} are the chirality parameters. From this we determine the ensemble polarization $\mathbf{P} = Nd_{34}\rho_{34}$ and magnetization $\mathbf{M} = N\mu_{21}\rho_{21}$, where N is the density of radiators and d_{34} and μ_{21} are the probe field transition electric and magnetic dipole moments. Figure 2 shows typical calculated spectra for the electric and magnetic susceptibilities as well as the chirality parameters without local-field corrections. The induced reduction of $\mathrm{Im}[\chi_e]$ and thus of the absorption on resonance is apparent. Likewise, ξ_{EH} is resonantly enhanced while ξ_{HE} and χ_m remain small.

Since we are interested in values for the refractive index substantially different from unity, we include both electric and magnetic local-field effects [25,26] by replacing the macroscopic field values **E** and **B** in Eq. (3) with the local (i.e., microscopic) fields $\mathbf{E}_m = \mathbf{E} + \mathbf{P}/3\varepsilon_0$ and $\mathbf{B}_m/\mu_0 = \mathbf{H}_m = \mathbf{H} + \mathbf{M}/3$. We then use the polarizabilities and

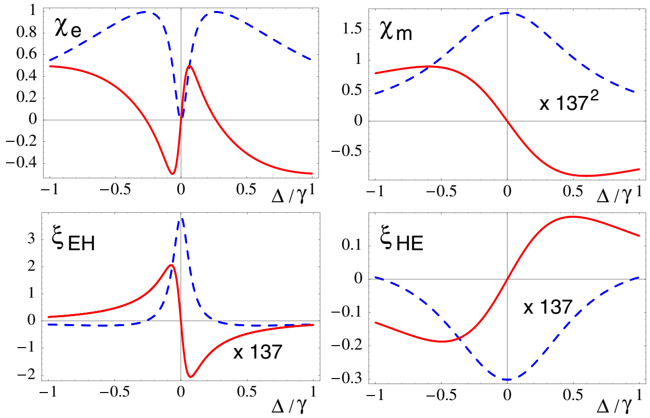

FIG. 2 (color online). Real (solid lines) and imaginary (dashed lines) parts of the electric (χ_e) and magnetic (χ_m) susceptibilities as well as the chirality parameters (ξ_{HE}, ξ_{EH}) without local-field corrections in arbitrary but the same units, as a function of the probe field detuning Δ relative to the radiative decay rate γ_3 from level $|3\rangle$.

chirality parameters to calculate χ_e, χ_m, ε, μ, ξ_{EH}, and ξ_{HE} with local-field corrections, and determine the refractive index from Eq. (2). As noted above, we find ξ_{EH}, $\xi_{HE} \sim \alpha\chi_e$, which makes negative refraction with minimal absorption feasible at moderate densities. (A detailed treatment of this calculation will be presented in an upcoming, longer paper.)

As an example, Fig. 3(a) shows the calculated real and imaginary parts of the refractive index as a function of probe field detuning for a density of $N = 5 \times 10^{16}$ cm^{-3} and using the realistic parameter values $\gamma_2 = 10^3$ s^{-1}, $\Omega_c = 10^4 e^{i\pi/2}\gamma_2$, $\Omega_1 = \Omega_2 = 10^6$ s^{-1}, $\gamma_3 = \gamma_5 = (137)^2\gamma_2$, $\gamma_4 \simeq 0$, and $\gamma_P = 10^4\gamma_2$. The transition dipole matrix elements are related to the radiative decay rates via $d_{34}(\mu_{21}/c) = \sqrt{3\pi\varepsilon_0\gamma_3(\gamma_2)\hbar c^3/\omega^3}$ for a typical optical wavelength of 600 nm. We find substantial negative refraction and minimal absorption for this density, which is about a factor of 10^2 smaller than the density needed without taking chirality into account [22]. It should also be noted that the magnetic permeability differs by less than 10% from unity for the density considered here. Therefore, the negative refraction shown in Fig. 3(a) is clearly a consequence of chirality.

For larger N the optical response of the medium increases. For example, Fig. 3(b) shows the refractive index for $N = 5 \times 10^{17}$ cm^{-3}. As expected the refraction reaches increasingly larger negative values. Remarkably, we find that the absorption reaches a maximum and then decreases with increasing density. This effect is illustrated in Fig. 4, where we show the real and imaginary parts of the refractive index as well as the real part of μ (including local-field effects) as functions of the density. This peculiar behavior is a result of the combined electric and magnetic local-field corrections [26]. As a consequence the refraction/absorption ratio, shown in the inset of Fig. 4, continues

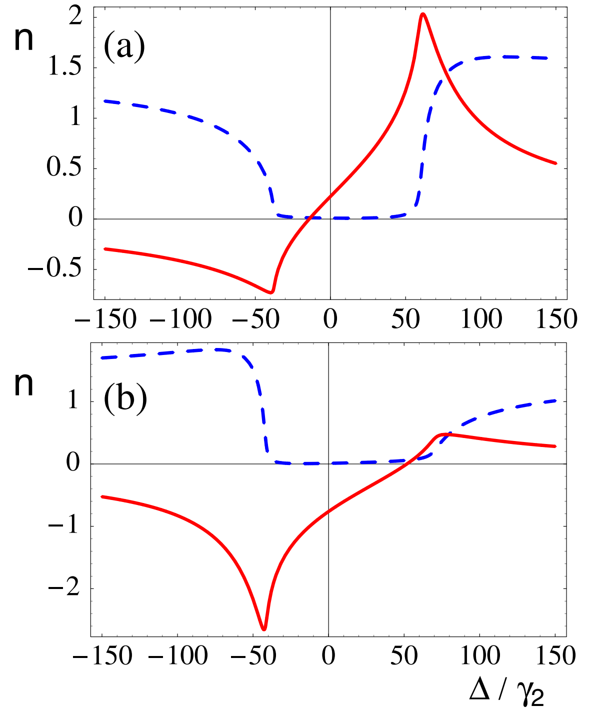

FIG. 3 (color online). Real (solid lines) and imaginary (dashed lines) parts of the refractive index as a function of the probe field detuning Δ relative to the radiative decay rate γ_2 from level $|2\rangle$, for a density of (a) $N = 5 \times 10^{16}$ cm^{-3} and (b) $N = 5 \times 10^{17}$ cm^{-3}.

to increase with density and reaches rather large values on the order of 10^2. These results should be contrasted to previous theoretical proposals and experiments on negative refraction in the optical regime, for which $-\mathrm{Re}[n]/\mathrm{Im}[n]$ is typically on the order of unity [10–12,14].

It has been pointed out by Smith *et al.* and Merlin [27] that the realization of subdiffraction-limit imaging with a lens of thickness d and resolution Δx requires an extreme

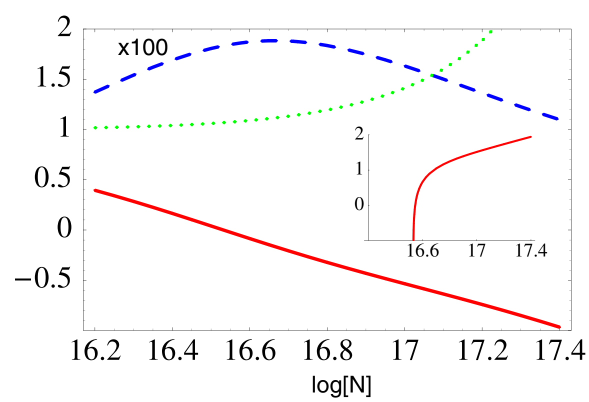

FIG. 4 (color online). Real (solid line) and imaginary (dashed line, $\times 100$) parts of the refractive index, as well as the real part of the permeability (dotted line), as a function of the logarithm of the density N, at a frequency slightly below resonance ($\Delta = -25\gamma_2$). Inset: $\log|\mathrm{Re}(n)/\mathrm{Im}(n)|$ as a function of $\log[N]$.

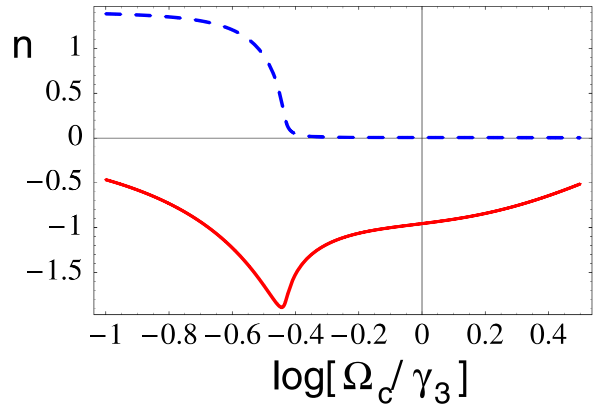

FIG. 5 (color online). Real (solid line) and imaginary (dashed line) parts of the refractive index as a function of the coupling field Rabi frequency Ω_c relative to the radiative decay rate γ_3, for $N = 3.5 \times 10^{17}$ cm^{-3} and $\Delta = -25\gamma_2$.

fine-tuning of the index of refraction to the value $n = -1$ with accuracy $\Delta n = 1 - \exp\{-\frac{\Delta x}{2\pi d}\}$. The quantum interference scheme presented here provides a handle for this. For example, as shown in Fig. 5, the real part of the refractive index can be fine-tuned by relatively coarse adjustments of the strength of the coupling field Ω_c.

As with EIT and its applications, electromagnetically induced chirality should be realizable in a wide range of atomic, molecular, and condensed matter systems: already, a basic form has been observed in Rb vapor [16]. In preparation for further experimental investigations, we are currently performing a detailed assessment of systems such as metastable neon, rare-earth atoms, and donor-bound electrons and bound excitons in semiconductors, all of which can have level structures and interactions analogous to those of Fig. 1(b). A promising example is dysprosium vapor, which has been studied spectroscopically at densities $>10^{16}$ cm^{-3} [28] and for which there are many appropriate combinations of energy levels: e.g., a probe field at 710 nm with the $E1$ transition between $4f^{10}5d6s$, $J = 8$ and $4f^{9}5d^{2}6s$, $J = 7$ and the $M1$ transition between $4f^{10}6s6p$, $J = 7$ and $4f^{10}6s6p$, $J = 6$. Collaborative experimental investigation of electromagnetically induced chirality and negative refraction in rare-earth atoms are planned using ablation or buffer-gas-cooling technology [29]. Results from these investigations will be reported in the future.

In conclusion, we showed that quantum interference effects similar to EIT can lead to a large induced chirality under realistic conditions, and thus enable tunable negative refraction with minimal absorption. Such electromagnetically induced chiral negative refraction should be applicable to a wide array of systems in the optical regime, including atoms, molecules, quantum dots, and excitons.

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