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Negative refraction with low absorption using Raman transitions with magnetoelectric coupling

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We suggest a scheme for obtaining negative refraction that does not require the simultaneous presence of an electric-dipole and a magnetic-dipole transition near the same transition frequency. The key idea of the scheme is to obtain a strong electric response by using far-off-resonant Raman transitions. We propose to use a pair of electric-dipole Raman transitions and utilize magneto-electric cross coupling to achieve a negative index of refraction without requiring negative permeability. The interference of the two Raman transitions allows tunable negative refraction with low absorption.

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More than four decades ago, Veselago predicted that materials with a simultaneous negative permittivity and permeability would acquire a negative index of refraction, $n < 0$ [1]. These materials exhibit many seemingly strange properties such as the electromagnetic vectors forming a left-handed triad (hence the term left-handed materials) and the Poynting vector being antiparallel to the k vector. The key practical application for these materials was discovered in the year 2000 when Pendry demonstrated that a slab with a negative index of refraction can image objects with, in principle, unlimited resolution [2]. Since Pendry's suggestion, the interest in these materials has been continuously growing and there have been a large number of theoretical developments and experimental advances [3–8]. Initial experiments have demonstrated such materials in the microwave region of the spectrum by using metal wires and split-ring resonators [5–8]. Recently, utilizing advances in nanolithography techniques, several groups have reported a negative index of refraction at optical frequencies in metal and dielectric nanostructures [9–12]. A key difficulty of these experiments, which is particularly pronounced in the optical domain, is the large absorption that accompanies negative refraction. The performance of the left-handed materials is typically characterized by the figure of merit $F = -\text{Re}(n)/|\text{Im}(n)|$. For all recent experiments in the optical region, the figure of merit is of order unity, $F \approx 1$, which is a key limitation for many potential applications. It therefore remains a big challenge to obtain negative refraction with low absorption in the optical region of the spectrum.

The chief difficulty for obtaining negative refraction in the optical region of the spectrum is the weakness of the magnetic response. It is difficult to obtain a negative permeability with low absorption since typical transition magnetic-dipole moments are much smaller than the electric-dipole moments. To alleviate this problem, recently, a chiral route to negative refraction has been suggested [13,14]. Here, the key idea is to use a magnetoelectric cross coupling where the medium's electric polarization is coupled to the magnetic field of the wave and the medium's magnetization is coupled to the electric field. Under such conditions, a negative index of refraction can be achieved without requiring a negative permeability. Building on this idea, Fleischhauer and colleagues have recently suggested a promising scheme that achieves negative refraction with low absorption using quantum interference [15,16]. Their scheme utilizes the dark state of the electromagnetically induced transparency (EIT) to reduce absorption while enhancing the chiral response. The work of Fleischhauer and colleagues improves on the pioneering efforts of Oktel and Mustecaplioglu [17] and Thommen and Mandel [18].

All of these current suggestions require a strong magnetic-dipole transition and a strong electric-dipole transition near the same transition frequency. This requirement puts a stringent constraint on the energy level structure of systems where negative refraction can be achieved. In this article, we suggest an alternative route that overcomes this constraint. To overcome the need for an electric-dipole transition at a specific frequency, we use two-photon Raman transitions that can be very far detuned from the electronic state. We obtain a strong electric-dipole response with low absorption by using a pair of Raman transitions: one amplifying and one absorptive in nature. The interference of these two transitions result in a strong enhancement of the permittivity while minimizing absorption. We then coherently couple to a magnetic-dipole transition to obtain a chiral response and achieve a negative index of refraction through magnetoelectric cross coupling.

Before proceeding with a detailed description of our suggestion, we summarize the chiral approach to negative refraction. Consider a probe beam with electric-field and magnetic-field components E_p and B_p, respectively. In a material with magnetoelectric cross coupling, the medium polarization P_p and the magnetization M_p are given by [15,16]

$$P_p = \epsilon_0 \chi_E E_p + \frac{\xi_{EB}}{c\mu_0} B_p,$$
$$M_p = \frac{\xi_{BE}}{c\mu_0} E_p + \frac{\chi_M}{\mu_0} B_p, \qquad (1)$$

where χ_E and χ_M are the electric and magnetic susceptibilities, and ξ_{EB} and ξ_{BE} are the complex magnetoelectric coupling (chirality) coefficients, respectively. The index of refraction of the medium for a plane wave of a particular circular polarization can be found by using Eqs. (1) and Maxwell's equations:

$$n = \sqrt{\epsilon\mu - \frac{(\xi_{EB} + \xi_{BE})^2}{4}} + \frac{i}{2}(\xi_{EB} - \xi_{BE}). \qquad (2)$$

Here, $\epsilon = 1 + \chi_E$ and $\mu = 1 + \chi_M$ are the relative permittivity and permeability of the medium. As shown in Eq. (2), the chirality coefficients result in additional contributions to the index of refraction. The key idea behind the chiral approach is that, in the optical region, one typically has the scaling

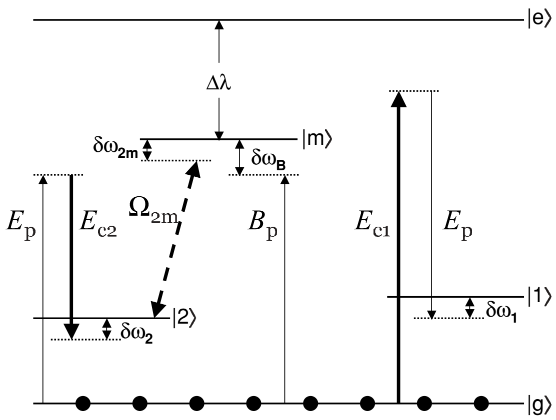

FIG. 1. The energy level diagram of our suggestion. E_p and B_p are the electric-field and magnetic-field components for the probe laser beam. $|g\rangle \rightarrow |m\rangle$ is a magnetic-dipole transition induced by the probe magnetic-field B_p. Two strong control laser beams, E_{c1} and E_{c2}, induce two electric-dipole Raman transitions for the probe beam. The Raman transitions can be very far detuned from the excited state $|e\rangle$. Therefore, the system does not require $|g\rangle \rightarrow |m\rangle$ and $|g\rangle \rightarrow |e\rangle$ transitions to be near the same frequency. Ω_{2m} induces magnetoelectric cross coupling (chirality).

$\chi_M \sim \alpha^2 \chi_E$ and $(\xi_{EB},\xi_{BE}) \sim \alpha \chi_E$, where $\alpha = 1/137$ is the fine structure constant. Since the values of the chirality coefficients are smaller only by a factor of α instead of α^2, negative refraction can be achieved without the need for negative permeability and at much smaller atomic densities compared to those of nonchiral schemes. Negative refraction with chirality requires appropriate phase control of the chirality coefficients which can be achieved through coherent magnetoelectric coupling. One typically chooses the phase such that the chirality coefficients are imaginary, $\xi_{EB} = -\xi_{BE} = i\xi$, and Eq. (2) reads $n = \sqrt{\epsilon\mu} - \xi$. Achieving $n < 0$ then requires $\xi > \sqrt{\epsilon\mu}$.

We proceed with a detailed description of our suggestion. Noting Fig. 1, we consider a five-level system interacting with four laser beams. We wish to achieve a negative index of refraction for the probe laser beam with field components E_p and B_p, respectively. We take the atomic system to have a strong magnetic transition with the dipole-moment μ_{gm} near the frequency of the probe laser beam. As mentioned previously, the system does not have a strong electric-dipole transition near the probe laser frequency. The electric-dipole response is obtained by using two-photon Raman transitions through the excited state $|e\rangle$. At the heart of the scheme is the recently predicted and experimentally demonstrated "refractive index enhancement with vanishing absorption" technique [19,20]. Starting with the ground state $|g\rangle$, we induce two Raman transitions using the probe laser and two intense control lasers with electric field amplitudes E_{c1} and E_{c2}, respectively. Since the order at which the probe laser beam is involved in each Raman transition is different, this scheme achieves two resonances: one amplifying and one absorptive in nature. The strength and the position of these two resonances can be controlled by varying the intensities and the frequencies of the control laser beams. It is the interference of these two resonances that results in the control of the index of refraction while maintaining small absorption. The magnetoelectric cross coupling is achieved through coherent coupling of states $|2\rangle$ and $|m\rangle$ with a separate laser beam of Rabi frequency Ω_{2m}. States $|g\rangle$, $|1\rangle$, $|2\rangle$, and $|m\rangle$ have the same parity, which is opposite to the parity of state $|e\rangle$. Since states $|2\rangle$ and $|m\rangle$ have the same parity, the coherent coupling Ω_{2m} cannot be electric-dipole, but instead can be achieved through the magnetic-field of a strong laser or through a separate two-photon transition (not shown). The two-photon detunings from the two Raman transitions are defined as $\delta\omega_1 = (\omega_1 - \omega_g) - (\omega_{c1} - \omega_p)$ and $\delta\omega_2 = (\omega_2 - \omega_g) - (\omega_p - \omega_{c2})$. The quantity $\delta\omega_B = (\omega_m - \omega_g) - \omega_p$ is the detuning of the probe laser beam from the $|g\rangle \rightarrow |m\rangle$ magnetic transition. $\delta\omega_{2m}$ is the detuning of the magnetoelectric coupling laser beam from the $|2\rangle \rightarrow |m\rangle$ transition. To obtain a closed loop, we require $\delta\omega_B = \delta\omega_2 + \delta\omega_{2m}$.

We focus our attention on the case where the single photon detunings of the laser beams from the excited state $|e\rangle$ are much larger than the coupling rates. This allows adiabatic elimination of the probability amplitude of state $|e\rangle$ and therefore reduces the problem to an effective four-level system. We solve the Schrödinger's equation with the decay rates added phenomenologically at steady state and in the perturbative limit, where most of the population stays in the ground state, $\rho_{gg} \approx 1$. We have verified the validity of this steady-state and perturbative solution by direct numerical integration. From the solution of the Schrödinger's equation, we calculate the relevant coherences and the medium's polarization and magnetization. The details of these calculations will be reported elsewhere. The end result for the susceptibilities and the chirality coefficients, within the rotating wave approximation, are

$$\chi_E = \frac{N}{\epsilon_0}\left[\frac{|d_{ge}|^2}{\hbar(\Delta_p - i\Gamma_e/2)} + \frac{|d_{ge}|^2|d_{1e}|^2}{4\hbar^3\Delta_1^2(\delta\omega_1 + i\gamma_1)}|E_{c1}|^2 + \frac{|d_{ge}|^2|d_{2e}|^2}{4\hbar^3\Delta_2^2\left(\delta\omega_2 - \frac{|\Omega_{2m}|^2}{4(\delta\omega_B - i\gamma_m)} - i\gamma_2\right)}|E_{c2}|^2\right],$$

$$\chi_M = N\mu_0\left[\frac{|\mu_{gm}|^2}{\hbar(\delta\omega_B - i\gamma_m)} + \frac{|\mu_{gm}|^2}{4\hbar(\delta\omega_B - i\gamma_m)^2\left(\delta\omega_2 - \frac{|\Omega_{2m}|^2}{4(\delta\omega_B - i\gamma_m)} - i\gamma_2\right)}|\Omega_{2m}|^2\right],$$

$$\xi_{EB} = N\mu_0 c\frac{d_{ge}d_{2e}^*\mu_{gm}^*}{4\hbar^2\Delta_2(\delta\omega_B - i\gamma_m)\left(\delta\omega_2 - \frac{|\Omega_{2m}|^2}{4(\delta\omega_B - i\gamma_m)} - i\gamma_2\right)}\Omega_{2m}E_{c2}, \tag{3}$$

$$\xi_{BE} = N\mu_0 c\frac{d_{ge}^*d_{2e}\mu_{gm}}{4\hbar^2\Delta_2(\delta\omega_B - i\gamma_m)\left(\delta\omega_2 - \frac{|\Omega_{2m}|^2}{4(\delta\omega_B - i\gamma_m)} - i\gamma_2\right)}\Omega_{2m}^*E_{c2}^*,$$

where N is the atomic density and Γ_e is the decay rate of the excited state $|e\rangle$. The quantities d_{ge}, d_{1e}, and d_{2e} are the electric-dipole matrix elements between relevant states, and μ_{gm} is the magnetic-dipole matrix element for the $|g\rangle \rightarrow |m\rangle$ transition. Δ_p, Δ_1, and Δ_2 are the large single photon detunings of the relevant transitions from the excited state $|e\rangle$. The quantities $\delta\omega_1$,$\delta\omega_2$, and $\delta\omega_B$ are the detunings as defined in Fig. 1, but modified to take into account the Stark shifts due to the intense control beams E_{c1} and E_{c2}. γ_1, γ_2, and γ_m are the total linewidths of the respective states, which include radiative decay, possible broadening mechanisms such as collisions, and the power broadening due to control laser beams.

We proceed with a numerical example for a model atomic system. We consider a probe beam at a wavelength of $\lambda_p = 600$ nm. We take the radiative decay rate of state $|e\rangle$ to be $\Gamma_e = 2\pi \times 10$ MHz and calculate the dipole matrix elements, d_{ge}, d_{1e}, and d_{2e}, using the Wigner-Weisskopf result and assuming equal branching ratios, $d_{ij} = \sqrt{\pi\epsilon_0\Gamma_e\hbar c^3/\omega_p^3}$. We apply a similar procedure and assume a radiative decay rate of $\Gamma_e/137^2$ for the magnetic level $|m\rangle$ and calculate the corresponding magnetic-dipole matrix element, μ_{gm}. To simulate a realistic system, we assume an additional broadening mechanism (for example, collisions) with a rate of $\gamma_c = 2\pi \times 1$ MHz and add this broadening to the linewidths of states $|1\rangle$, $|2\rangle$, and $|m\rangle$. We take the wavelengths of electric-dipole ($|g\rangle \rightarrow |e\rangle$) and magnetic-dipole ($|g\rangle \rightarrow |m\rangle$) transitions to be different by $\Delta\lambda = 0.1$ nm. As we will discuss subsequently, this difference can be larger at the expense of an increase in the required control laser intensities. For simplicity, we assume the large single photon detunings to be the same, $\Delta_p = \Delta_1 = \Delta_2$. We take the magnetoelectric coupling laser beam to be resonant with the $|2\rangle \rightarrow |m\rangle$ transition and therefore take $\delta\omega_B = \delta\omega_2$. We also assume that the control laser frequencies are appropriately adjusted such that the two Raman resonance frequencies coincide as the probe laser frequency is scanned, $\delta\omega_1 = -\delta\omega_2$.

Figure 2 shows the susceptibilities and the chirality coefficients, χ_E, χ_M, ξ_{EB}, and ξ_{BE}, without the local-field corrections as the frequency of the probe laser beam is varied for an atomic density of $N = 5 \times 10^{16}/\text{cm}^3$. Here we take the intensities of the two control laser beams to be $I_{c1} = 0.251\ \text{MW/cm}^2$ and $I_{c2} = 0.5\ \text{MW/cm}^2$ and assume $\Omega_{2m} = i2\pi \times 2.12$ MHz. As shown in Fig. 2, the magnetoelectric coupling causes an EIT-like level splitting for χ_E. The imaginary part of χ_E becomes small near $\delta\omega_B = 0$ due to the interference of the two Raman resonances. One of the key differences of our approach compared to the scheme of Fleischhauer [15,16] is that, since the electric-dipole response is due to Raman transitions, its strength is controlled by the intensity of the control laser beams. As a result, we do not have the usual scaling $\chi_M \sim \alpha^2\chi_E$ and $(\xi_{EB},\xi_{BE}) \sim \alpha\chi_E$. Therefore, the magnitude of χ_E can be made more comparable to the chirality coefficients.

For materials with a refractive index substantially different from unity, the microscopic local fields can be substantially different than the averaged macroscopic fields. To calculate the refractive index, we include both the electric and magnetic Clausius-Mossotti-type local-field effects [16,21,22]. Figure 3

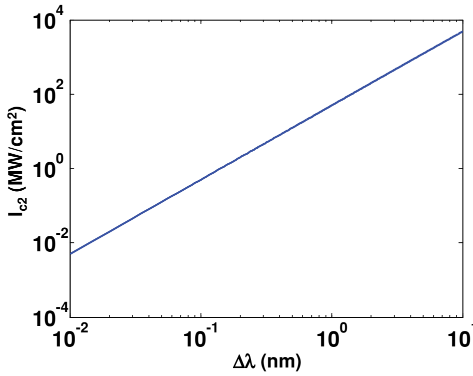

FIG. 5. (Color online) The required control laser intensity, I_{c2}, as the wavelength separation between the transitions, $\Delta\lambda$, is varied to obtain results comparable to those of Figs. 2–4.

shows the real and imaginary parts of the refractive index for parameters identical to those in Fig. 2. The index of refraction reaches a value of $n \approx -1$ with a figure of merit of $F > 10$. To show the critical dependence on atomic density, Fig. 4 shows the refractive index for $N = 2 \times 10^{16}/\text{cm}^3$ and $N = 3 \times 10^{17}/\text{cm}^3$ with parameters otherwise identical to those of Fig. 3. For $N = 3 \times 10^{17}$, we obtain an index of refraction of $n = -4$ with low absorption. For $n = -1$ the figure of merit is $F \approx 200$. We note that in Figs. 3 and 4, near $\delta\omega_B = 0$, the absorption is small and the figure of merit becomes very large. However, at these probe laser frequencies, the index of refraction is also small.

As mentioned previously, for Figs. 2–4, the transition wavelengths for the electric-dipole ($|g\rangle \rightarrow |e\rangle$) and magnetic-dipole ($|g\rangle \rightarrow |m\rangle$) transitions are assumed to be different by $\Delta\lambda = 0.1$ nm. This wavelength separation can be larger at the expense of an increase in the required control laser intensities. Figure 5 demonstrates this result. Here we plot the required control laser intensity, I_{c2}, as the wavelength separation between the transitions, $\Delta\lambda$, is varied to obtain results comparable to those of Figs. 2–4. The two transition wavelengths may be different by as much as $\Delta\lambda = 10$ nm and the scheme will still work with experimentally accessible laser systems. This gives considerable flexibility on the energy level structure of our scheme which may enable an experimental implementation.

In conclusion, we have suggested an alternative approach for achieving negative refraction that does not require the simultaneous presence of an electric-dipole and a magnetic-dipole transition near the same frequency. We are currently investigating a suitable system where our approach may be experimentally implemented. As mentioned in Ref. [15], rare-earth atoms such as dysprosium vapor show considerable promise. Detailed assessment of our approach in such systems will be among our future investigations. We also expect our technique to be applicable in other systems including molecules and solid-state structures [23].

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