Refractive Index Enhancement in a Far-Off Resonant Atomic System

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(Received 5 May 2005; published 22 November 2005)

We demonstrate a scheme where a laser beam which is very far detuned from an atomic resonance experiences a large index of refraction with vanishing absorption. The essential idea is to excite two Raman resonances with appropriately chosen strong control lasers.

DOI: 10.1103/PhysRevLett.95.223601 PACS numbers: 42.50.Gy, 42.65.An, 42.65.Dr, 78.20.Ci

It is well known that a laser beam tuned close to an atomic resonance can experience a large index of refraction. However, such a large index is usually accompanied by large absorption. This is because, at frequencies near an optical resonance, the real and imaginary parts of the linear susceptibility are of the same order. Over the last decade, Scully and colleagues have predicted [1–3] and demonstrated [4] that, by using quantum interference, it is possible to obtain a large index of refraction with vanishing absorption. The essential idea is to establish a Raman coherence such that there is complete destructive interference in the imaginary part of the linear susceptibility. This interference is obtained very close to an atomic resonance with substantial excited state fraction [1–4]. In this Letter, we extend this idea to a far-off resonant atomic or molecular system. We show that the refractive index of a weak probe beam which is very far detuned from an optical resonance can be enhanced by many orders of magnitude while maintaining vanishing absorption.

Noting Fig. 1, we consider a model atomic or molecular system with a ground Raman state $|g\rangle$, two excited Raman states $|1\rangle$, and $|2\rangle$, and an excited upper state $|e\rangle$. The probe beam, E_p, is weak and is largely detuned from any one-photon resonance. Together with the probe beam, two strong control fields, E_{c1} and E_{c2}, two-photon couple the ground state $|g\rangle$ to excited Raman states $|1\rangle$ and $|2\rangle$, respectively. In the absence of the control fields, the probe beam experiences the usual largely detuned linear susceptibility. As will be demonstrated later, the presence of the control fields strongly modify the susceptibility of the probe beam. In particular, one can obtain a great enhancement in the real part of the susceptibility while maintaining perfect destructive interference in the imaginary part.

Before proceeding further, we would like to cite pertinent earlier work: over the recent years, there has been substantial work utilizing unusual dispersive and absorptive properties of systems exhibiting quantum interference. Of particular importance is lasers without inversion and electromagnetically induced transparency (EIT) [5]. Harris *et. al.* have shown how to reduce the refractive index of a probe beam to unity in a far-off resonant system in an EIT-like manner [6]. Several papers have discussed the possibility of refractive index control for a comb of Raman sidebands [7,8]. Walker and colleagues have demonstrated refractive index enhancement and reduction with maximally coherent molecules [9]. The simultaneous excitation of two Raman resonances and its utility in producing single cycle pulses has been recently suggested [10].

We proceed with the analysis of the schematic of Fig. 1. We follow the formalism of Harris and colleagues [6–8]. The two-photon detunings from the Raman resonances 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 quantities Γ_e, γ_1, and γ_2 denote the (amplitude) decay rates of states $|e\rangle$, $|1\rangle$, and $|2\rangle$, respectively. To avoid the need for a density matrix formalism, we take all of the decay rates to be decay outside the system. Since we are considering a far-off resonant system, we can adiabatically eliminate the derivative of the probability amplitude of the upper state $|e\rangle$ when compared with the detunings from this state. With these assumptions, the equations for the probability amplitudes of the three Raman states are [6–8]:

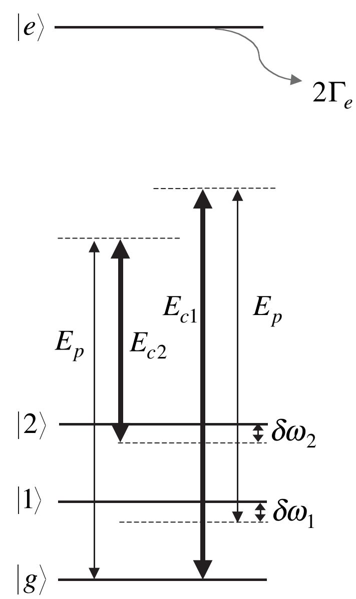

FIG. 1. The schematic of the proposed scheme. A weak far-off resonant probe beam E_p, and two strong control lasers, E_{c1} and E_{c2}, two-photon couple the ground state $|g\rangle$ to excited Raman states $|1\rangle$ and $|2\rangle$.

$$\frac{\partial c_g}{\partial t}+\frac{\Im(A)}{2}c_g=j\frac{B_1}{2}c_1+j\frac{B_2}{2}c_2,$$
$$\frac{\partial c_1}{\partial t}+j\left[\delta\omega_1-\frac{\Re(D_1-A)}{2}\right]c_1+\left[\gamma_1+\frac{\Im(D_1)}{2}\right]c_1=j\frac{B_1^*}{2}c_g,$$
$$\frac{\partial c_2}{\partial t}+j\left[\delta\omega_2-\frac{\Re(D_2-A)}{2}\right]c_2+\left[\gamma_2+\frac{\Im(D_2)}{2}\right]c_2=j\frac{B_2^*}{2}c_g, \tag{1}$$

where

$$\begin{aligned} A &= a_p|E_p|^2 + a_{c1}|E_{c1}|^2 + a_{c2}|E_{c2}|^2,\\ B_1 &= b_1 E_{c1}E_p^*,\\ B_2 &= b_2 E_p E_{c2}^*,\\ D_1 &= d_{1,p}|E_p|^2 + d_{1,c1}|E_{c1}|^2 + d_{1,c2}|E_{c2}|^2,\\ D_2 &= d_{2,p}|E_p|^2 + d_{2,c1}|E_{c1}|^2 + d_{2,c2}|E_{c2}|^2. \end{aligned} \tag{2}$$

In Eq. (1), the symbols $\Re$ and $\Im$ stand for the real and imaginary parts of the relevant complex quantities, respectively. In Eq. (2), the constants a, b, and d determine the stark shifts and the coupling, and they are (including rotating and nonrotating terms):

$$\begin{aligned} b_1 &= \frac{1}{2\hbar^2}\left[\frac{\mu_{ge}\mu_{1e}^*}{(\omega_e-\omega_g)-\omega_{c1}-j\Gamma_e} + \frac{\mu_{ge}\mu_{1e}^*}{(\omega_e-\omega_g)+\omega_{c1}-j\Gamma_e}\right],\\ b_2 &= \frac{1}{2\hbar^2}\left[\frac{\mu_{ge}\mu_{2e}^*}{(\omega_e-\omega_g)-\omega_p-j\Gamma_e} + \frac{\mu_{ge}\mu_{2e}^*}{(\omega_e-\omega_g)+\omega_p-j\Gamma_e}\right],\\ a_p &= \frac{1}{2\hbar^2}\left[\frac{|\mu_{ge}|^2}{(\omega_e-\omega_g)-\omega_p-j\Gamma_e} + \frac{|\mu_{ge}|^2}{(\omega_e-\omega_g)+\omega_p-j\Gamma_e}\right]. \end{aligned} \tag{3}$$

Here, μ_{ij} are the dipole matrix elements between respective transitions. The constants $a_{c1}, a_{c2}, d_{1,p}, d_{1,c1}, d_{1,c2}, d_{2,p}, d_{2,c1}, d_{2,c2}$ have the same form as a_p of Eq. (3) with the matrix elements and the angular frequencies replaced accordingly. When the decay rate of the upper state, Γ_e, is ignored, Eqs. (1)–(3) reduce to the formalism of Harris and colleagues [6–8]. Throughout this Letter, for simplicity, we will consider a single excited state $|e\rangle$. The formalism easily extends to an arbitrary number of upper states by summing through all these states while evaluating the constants of Eq. (3).

With the evolution of atomic system described by Eq. (1), we can evaluate the generated dipole moment at the probe frequency [6–8]:

$$\begin{aligned} P_p = 2\hbar N(&a_p|c_g|^2E_p + d_{1,p}|c_1|^2E_p + d_{2,p}|c_2|^2E_p\\ &+ b_1c_g^*c_1E_{c1} + b_2^*c_gc_2^*E_{c2}). \end{aligned} \tag{4}$$

We now proceed with an analysis of Eqs. (1) and (4). We proceed perturbatively, and assume that E_p is sufficiently weak and the detuning from the excited state $|e\rangle$ is sufficiently large such that most of the population remains in the ground state $|g\rangle$, and take $c_g \approx 1$. From Eq. (1), the steady state solution for the probability amplitudes of the excited Raman states are:

$$\begin{aligned} c_1 &\approx \frac{B_1^*}{2\{[\delta\omega_1 - \Re(D_1-A)/2] - j[\gamma_1 + \Im(D_1)/2]\}},\\ c_2 &\approx \frac{B_2^*}{2\{[\delta\omega_2 - \Re(D_2-A)/2] - j[\gamma_2 + \Im(D_2)/2]\}}. \end{aligned} \tag{5}$$

This steady state solution neglects the depletion of the ground state population and is therefore valid for times short compared to the inverse of $\Im(A)$. In Eq. (5), the quantities $\Re(D_1 - A)/2$ and $\Re(D_2 - A)/2$ are due to the stark shifts of the Raman states. We redefine the quantities $\delta\omega_1$ and $\delta\omega_2$ to include these stark shifts. From Eq. (4), the dipole moment at the probe frequency then is:

$$\begin{aligned} P_p = 2\hbar N\Bigg(&a_p + \frac{|b_1|^2}{2\{\delta\omega_1 - j[\gamma_1+\Im(D_1)/2]\}}|E_{c1}|^2\\ &+ \frac{|b_2|^2}{2\{\delta\omega_2 + j[\gamma_2+\Im(D_2)/2]\}}|E_{c2}|^2\Bigg)E_p. \end{aligned} \tag{6}$$

From Eq. (6) and using $P_p = \epsilon_0\chi E_p$, we can find the susceptibility of the medium at the probe wave:

$$\begin{aligned} \chi &\equiv \chi' + j\chi''\\ &= \frac{2\hbar N}{\epsilon_0}\Bigg(a_p + \frac{|b_1|^2}{2\{\delta\omega_1 - j[\gamma_1+\Im(D_1)/2]\}}|E_{c1}|^2\\ &\quad + \frac{|b_2|^2}{2\{\delta\omega_2 + j[\gamma_2+\Im(D_2)/2]\}}|E_{c2}|^2\Bigg), \end{aligned} \tag{7}$$

where χ' and χ'' stand for the real and imaginary parts of the susceptibility, respectively. As expected, the susceptibility of Eq. (7) is a sum of three terms. The first term in the right-hand side of Eq. (7) is a background susceptibility that would be present even in the absence of the strong control fields. The second term is due to two-photon resonance with the upper Raman state $|1\rangle$, and the third term is due to two-photon resonance with state $|2\rangle$. The two resonances occur when the probe laser wavelength is chosen such that $\omega_p = \omega_g + \omega_{c1} - \omega_1$ ($\delta\omega_1 = 0$) or $\omega_p = \omega_2 + \omega_{c2} - \omega_g$ ($\delta\omega_2 = 0$), respectively. We define the quantity $\Delta \equiv (\omega_1 + \omega_2 - 2\omega_g) - (\omega_{c1} - \omega_{c2})$ that determines the separation of these two resonances in frequency space. This quantity can also be rewritten in terms of the two-photon detunings: $\Delta = \delta\omega_1 + \delta\omega_2$.

The second term in the right-hand side of Eq. (7) causes gain on the probe beam whereas the third term causes absorption. These two terms can interfere to produce vanishing imaginary part of the susceptibility accompanied by a large index of refraction. For the case of equal matrix elements and equal quantities for the two Raman excitations ($b_1 = b_2$, $\gamma_1 = \gamma_2$, $I_{c1} = I_{c2}$), there is perfect destructive interference in the imaginary part of the susceptibility when $\delta\omega_1 = \delta\omega_2 = \Delta/2$. For this case, the real part of the susceptibility is (neglecting the background contribution):

$$\chi' = \frac{2\hbar N}{\epsilon_0} \frac{|b_1|^2 \delta\omega_1}{\{\delta\omega_1^2 + [\gamma_1 + \Im(D_1)/2]^2\}} |E_{c1}|^2$$
$$= \frac{2\hbar N}{\epsilon_0} \frac{|b_1|^2 (\Delta/2)}{\{(\Delta/2)^2 + [\gamma_1 + \Im(b_1)|E_{c1}|^2]^2\}} |E_{c1}|^2. \quad (8)$$

We proceed with a numerical example. We consider a sample alkili vapor cell where the three Raman states are different hyperfine states of the ground electronic state. We take the wavelength of the probe beam to be $\lambda_p = 800$ nm and take the excited state to be at a wavelength of $\lambda_e = 790$ nm. The probe beam is therefore very far detuned from the excited state. We take the matrix elements μ_{ge}, μ_{1e}, and μ_{2e} to be 1 a.u. and assume $N = 10^{13}/\mathrm{cm}^3$. We take the excited state decay rate to be $\Gamma_e = 2\pi \times 3$ MHz. For these parameters, the constants of Eq. (2) are equal and they are $a_p = b_1 = b_2 = 1.08 \times 10^{-4}$ in mks units. We take the ground state relaxation rates to be $\gamma_1 = \gamma_2 = 1$ KHz [11].

In Fig. 2, we plot the real and imaginary parts of the susceptibility of Eq. (7) as a function of $\delta\omega_1$ for $\Delta = 10, 5$, and 1 KHz, respectively. Here we take the power densities of the control lasers to be $I_{c1} = I_{c2} = 10^3$ W/cm^2. As also noted above, for $\delta\omega_1 = 0$, two-photon resonance with state $|1\rangle$ occurs. For $\delta\omega_1 = \Delta$ ($\delta\omega_2 = 0$), two-photon resonance with state $|2\rangle$ occurs. For $\delta\omega_1 = \Delta/2$ ($\delta\omega_2 = \Delta/2$), there is destructive interference in the imaginary part of the susceptibility. Conversely, the real part of the susceptibility obtains a large value of $\approx 10^{-6}$ due to constructive interference. This may be compared with the value of the susceptibility in the absence of the control fields which is 2.59×10^{-8}.

In Fig. 3, we use parameters identical to that of Fig. 2, and plot the real part of the susceptibility, χ', as a function of the separation of two resonances, Δ, at the point of quantum interference, $\delta\omega_1 = \delta\omega_2 = \Delta/2$. The real part of the susceptibility gets its largest value for $\Delta = 2\gamma_1 = 2$ KHz. For $\Delta \gg \gamma_1$, the system becomes two isolated Raman resonances and χ' drops. In the opposite limit of $\Delta \ll \gamma_1$, the effects of two Raman resonances cancel each other, resulting again in a small value of χ'. To obtain a large χ', it is therefore critical that Δ is on the order of the linewidth of the Raman transitions, γ_1.

We now ask the question: how much can the real part of the susceptibility be increased while maintaining vanishing absorption? In Eq. (8), the largest value of χ' is obtained when the control laser intensities are increased such that we have $\Im(D_1)/2 = \Im(b_1)|E_{c1}|^2 = \sqrt{\gamma_1^2 + (\Delta/2)^2}$. Using $\Im(b_1) \approx \frac{\Gamma_e}{(\omega_e - \omega_g) - \omega_p}|b_1|$ from Eq. (3), this maximum value of the susceptibility is:

$$\chi' = \xi \frac{N}{\hbar\epsilon_0} \frac{|\mu_{ge}|^2}{2\Gamma_e}. \quad (9)$$

Here ξ is a numerical factor that depends on the choice of Δ. For $\Delta = 2\gamma_1$, $\xi = 0.41$. Remarkably, Eq. (9) is the value of maximum susceptibility (within a factor of ξ) for a usual two level medium which would be obtained by tuning close to the atomic resonance. However, the

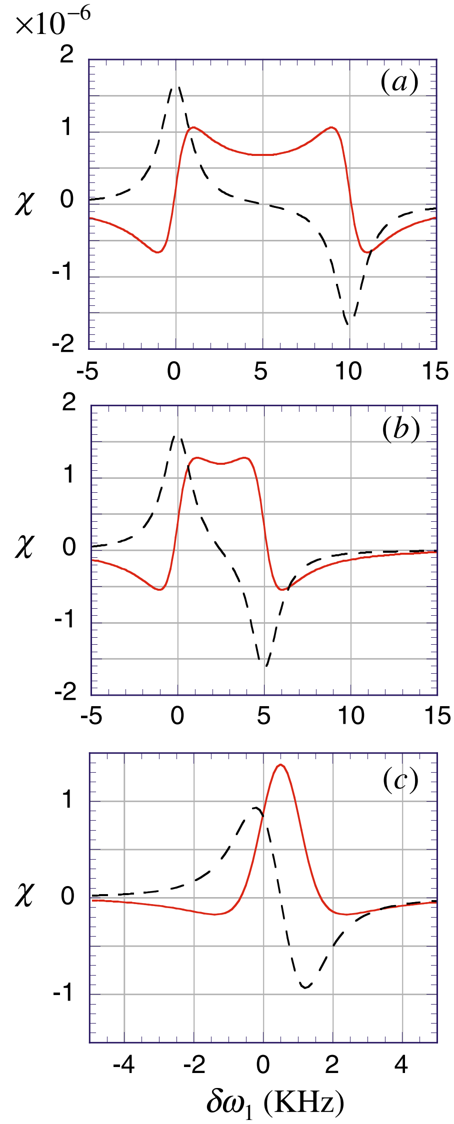

FIG. 2 (color online). The real part, χ' (solid line), and the imaginary part, χ'' (dashed line), of the susceptibility for (a) $\Delta = 10$ KHz, (b) $\Delta = 5$ KHz, and (c) $\Delta = 1$ KHz. The power densities of the control lasers are $I_{c1} = I_{c2} = 10^3$ W/cm^2. For $\delta\omega_1 = \Delta/2$, there is perfect destructive interference in the imaginary part of the susceptibility.

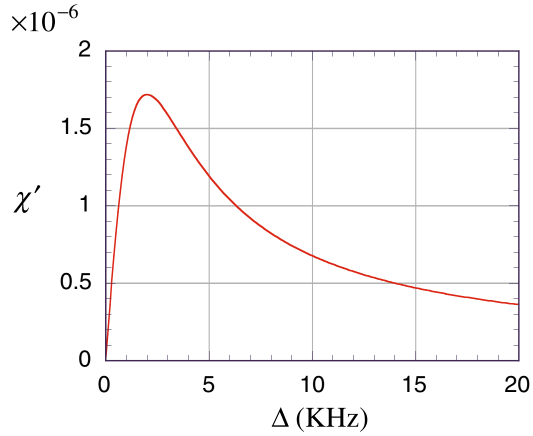

FIG. 3 (color online). The real part of the susceptibility, χ', as a function of the separation of two resonances Δ at the point of quantum interference $\delta\omega_1 = \delta\omega_2 = \Delta/2$. The parameters are identical to that of Fig. 2.

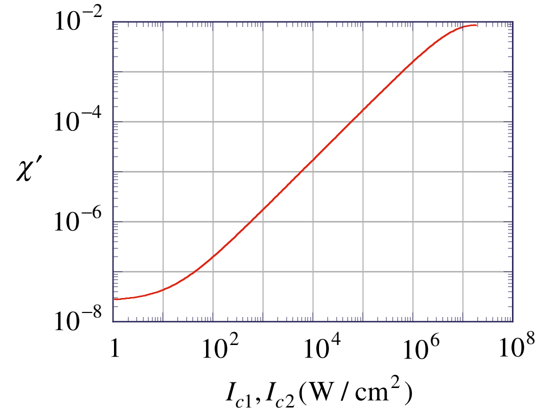

FIG. 4 (color online). The real part of the susceptibility as a function of the intensity of the control fields. For $I_{c1} = I_{c2} \approx 10^7\ \mathrm{W/cm^2}$, the susceptibility obtains its largest possible value. The imaginary part of the susceptibility vanishes for all these intensities.

difference here is that this value is obtained with vanishing imaginary part of the susceptibility.

In Fig. 4, we use parameters identical to that of Fig. 2 with $\Delta = 2$ KHz and plot the real part of the susceptibility as a function the intensity of the control fields $I_{c1} = I_{c2}$ at $\delta\omega_1 = \delta\omega_2 = \Delta/2 = 1$ KHz. At low intensities, the susceptibility has its background value of 2.59×10^{-8}. As the control laser intensities are increased, the susceptibility approaches its maximum value of $\approx 10^{-2}$. The imaginary part of the susceptibility vanishes for all these intensities.

We next discuss the relevant time scales of the problem. The susceptibility of Eq. (7) is valid for laser pulses long compared to $1/\gamma_1$. The results of Eqs. (5)–(9) are perturbative and therefore are valid for times short compared to $1/\Im(A)$, which is the depletion time of the ground state $|g\rangle$. The depletion of the ground state is due to the excited state spontaneous decay to the states outside the system. At the high intensity limit of Eq. (9), this depletion time is $1/\Im(A) \approx 1/\gamma_1$. Therefore, at the high intensity limit of Eq. (9) (for intensities $I_{c1} = I_{c2} > 10^6\ \mathrm{W/cm^2}$ in Fig. 4), the perturbative approximation breaks down and the ground state transients have to be taken into account. We have performed numerical simulations of Eq. (1) at high intensities and obtained maximum susceptibilities of $\chi' \approx 10^{-3}$ which is in reasonable agreement with the results of Fig. 4. We expect that if the atoms that are lost through excited state spontaneous decay are recycled back to the ground state (through optical pumping, for example), then maximum susceptibilities of Fig. 4, $\chi' \approx 10^{-2}$, will be attainable.

In summary, we have suggested a scheme to increase the refractive index of a far-off resonant probe beam while maintaining vanishing absorption. As also pointed out in Ref. [1], one exciting application of such schemes is to increase the spatial resolution of an optical microscope. For alkili vapor cells with densities $10^{17}/\mathrm{cm}^3$, the real part of the susceptibility may get values as large as $\chi' \approx 100$ and hence the refractive index $n \approx 10$. These schemes may provide an alternative to negative index materials in constructing lenses that beat the diffraction limit [12,13].

I would like to thank Miroslav Shverdin from Stanford University for many helpful discussions. This work was supported by the U. S. Army Research Office under Contract No. DAAD19-02-1-0083 and NSF Grant No. PHY-0205236.

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