Experimental Demonstration of Enhanced Index of Refraction via Quantum Coherence in Rb

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We present a proof-of-principle experiment demonstrating a resonant enhancement of the index of refraction accompanied by vanishing absorption in a cell containing a coherently prepared Rb vapor. The results are in good agreement with detailed theoretical predictions. [S0031-9007(96)00142-1]

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In the present Letter we report the first demonstration of a resonant enhancement of the index of refraction without absorption [1]. Surprising and counterintuitive effects involving quantum coherence and interference have recently become laboratory realities. For example, phenomena such as electromagnetically induced transparency (EIT) [2] and lasing without population inversion (LWI) [3], predicted theoretically at the end of the 1980s have now been demonstrated experimentally [4–7]. In both of the above effects quantum coherence dramatically modifies the *absorptive* properties of the medium. The unusual behavior of the *dispersive* part of the susceptibility of coherently prepared medium is no less intriguing. For example, it was shown theoretically that it is possible to have a completely transparent medium with large dispersion (i.e., rapid variation of index of refraction with frequency) or with large index of refraction [1]. Both of these effects can be achieved using atomic phase coherence in a resonant medium, which is normally optically thick. Coherence effects, however, allow us to prepare an optical medium such that the medium has vanishing absorption, while the dispersive part of the susceptibility is enhanced.

The interest in these phenomena is due, on one hand, to the fundamental nature of coherence effects and, on the other hand, to possible applications. For example, the dispersive properties of coherently prepared media have been studied with an eye toward generation of pulses with very slow group velocity [8], effective control of nonlinearities [9], high precision magnetometry [10], and laser acceleration of particles [1].

Several recent experiments [11] have demonstrated the large dispersion of the index of refraction accompanying EIT. Index enhancement, however, allows not only for large dispersion, but also for a large refractive index itself, while maintaining a transparent medium.

The conceptual foundation of the present experiment can be understood by considering the simple Λ atomic configuration of Fig. 1. The coherent strong driving field with Rabi frequency (Ω) and weak coherent probe (Ω_p) allow us to prepare the atom in a coherent superposition of states b and c [12]. When the detunings of these two fields from their respective atomic resonances are equal, EIT is obtained. The incoherent pumping (represented by the rate r) alters this coherent superposition by pumping some of the population into other states. Depending upon the actual parameters of the system this may result in gain, loss, or complete transparency for the probe field.

The linear gain (absorption) coefficient (G) is proportional to the imaginary part of complex susceptibility (χ''). In the case when the driving field is in exact resonance with transition $a \rightarrow c$ it is given for this particular three-level configuration by [13]

$$G = \kappa \frac{\gamma_{bc} A(\gamma_{cb}\gamma_{ab} + |\Omega|^2 - \Delta^2) + \Delta^2 B(\gamma_{ab} + \gamma_{bc})}{(\gamma_{bc}\gamma_{ab} + |\Omega|^2 - \Delta^2)^2 + \Delta^2(\gamma_{ab} + \gamma_{bc})^2}, \tag{1}$$

where $\kappa = 3\lambda^2 N\gamma'_a L/4\pi$, and A and B are given by

$$A = [1 + (\gamma'_a + \gamma''_a)/2\gamma_{bc}]\rho_{aa} - \rho_{bb}, \tag{2}$$

$$B = \rho_{aa} - \rho_{bb}. \tag{3}$$

Here ρ_{ii} is the population of level i calculated to the zeroth order in the probe field, γ_{ij} is the relaxation rate of the density matrix element ρ_{ij}, N is the density of atoms, L is the length of the cell, λ is the wavelength,

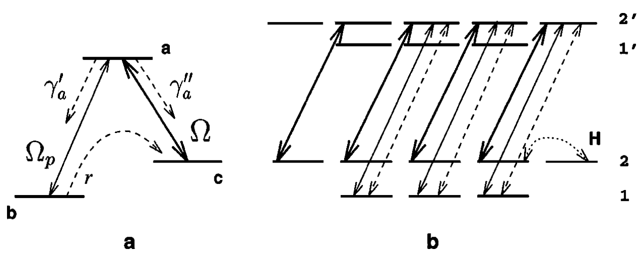

FIG. 1. (a) Simplified three-level model for index enhancement. (b) The actual level scheme of D_1 absorption line in Rb^{87} and the optical fields used in the present experiment. Energy separation between manifolds 1 and 2 is equivalent to 6.8 GHz.

Δ is the detuning of the probe laser, and atomic decays (γ_a', γ_a'') are indicated in Fig. 1(a). For the case $\Delta = 0$ we see that, in the presence of the coherent driving field, the gain (which is in that case proportional to A) can be positive even if most population remains in the ground state b, i.e., even if $B < 0$. That is, a weak probe field undergoes amplification without the need of population inversion [7,14]. If the probe field is detuned from atomic resonance, the gain coefficient decreases rapidly, and vanishes when

$$\Delta = \Delta_0 = \pm\sqrt{\frac{A\gamma_{bc}(\Omega^2 + \gamma_{ab}\gamma_{bc})}{A\gamma_{bc} - B(\gamma_{ab} + \gamma_{bc})}}. \tag{4}$$

Let us turn now to the real part of the complex susceptibility (χ'), i.e., to the resonant index of refraction. It results, in particular, in the phase shift (ϕ) for the probe light transmitted through the resonant vapor, which is proportional to χ':

$$\phi = -\frac{\kappa}{2}\Delta\frac{B(\gamma_{cb}\gamma_{ab} + |\Omega|^2 - \Delta^2) - A(\gamma_{ab} + \gamma_{bc})\gamma_{bc}}{(\gamma_{cb}\gamma_{ab} + |\Omega|^2 - \Delta^2)^2 + \Delta^2(\gamma_{ab} + \gamma_{bc})^2}. \tag{5}$$

The phase shift is equal to zero when the probe field is tuned precisely to the resonance with the $a \rightarrow b$ transition, but for nonzero probe field detuning it can become quite large. To be more specific, we consider the system with decay rates and pumping as indicated in Fig. 1(a) [13]. In this case and if we take $r \sim \gamma_a' \sim \gamma_a''$, $\Omega \gg \sqrt{\gamma_{ab} r}$, the gain coefficient vanishes at $\Delta_0 \sim \pm\sqrt{|\Omega_d|^2 + \gamma_{ab}\gamma_{bc}}$. At this point the phase shift is on the order of

$$\phi(\Delta_0) \sim (\kappa/12)/\Delta_0. \tag{6}$$

It follows, therefore, that a medium can become transparent at the point where the resonant index of refraction has a large value.

A Λ-type atomic configuration can be realized within the D_1 absorption line of Rb as shown in Fig. 1(b). Here right circularly polarized coherent driving and probe fields are tuned close to the $2 \rightarrow 2'$ and $1 \rightarrow 2'$ transitions, respectively [15]. The incoherent pumping out of the states 1 is accomplished by a broadband right circularly polarized field which couples the transition $1 \rightarrow 2'$. All of the fields are copropagating, which allows for the reduction of Doppler broadening.

We note here that optical properties of the real atomic systems are, in general, different from those of simplified 3- or 4-level models due to the presence of many hyperfine and Zeeman sublevels. In particular, optical pumping can play an important role. Indeed, in the case when both driving and pumping fields are present, atoms can be optically pumped into the state $F = 2$, $M_F = +2$ [Fig. 1(b)]. To avoid the trapping of the population in this state a weak magnetic field ($\sim 2 \times 10^{-4}$ T) is used, which mixes the populations via Larmor precession.

In order to account for realistic experimental conditions we developed a numerical model, wherein the field polarizations, the presence of hyperfine and Zeeman sublevels and Doppler broadening are taken into account. This is done by numerically solving the density matrix equations for the 16-level system of Fig. 1(b) and averaging over a Maxwell velocity distribution. The numerical analysis showed that for parameters typical of our experiment the effect of index enhancement depends upon the tuning of the drive laser within the Doppler absorption profile. This is the case when the intensity of a driving field is relatively low and the ground state relaxation rate is sufficiently high (determined in our case by the incoherent pump rate). Under these conditions we found the index enhancement is larger if the driving field is detuned to either side of the center of the Doppler profile by roughly a third of the Doppler width.

The results of the calculations are presented in Fig. 2. They clearly show that there is a region (indicated by I) where absorption is canceled and, at the same time, the real part of susceptibility (i.e., the phase shift) increases due to the presence of the drive and pump fields.

In the experiment we used a 4 cm long cell containing natural Rb. Drive and probe lasers were extended cavity diode lasers at 794 nm with linewidths of about 100 kHz. The powers of these beams in the region of the cell were on the order of 10 mW and 5 μW, respectively, and the corresponding spot sizes in the same region were 2 mm and 100 μm. The angle between two beams did not exceed 5×10^{-3} rad. The pump laser was a solitary laser diode (794 nm) with output power $\approx$5mW. Its linewidth was additionally broadened to about 150 MHz

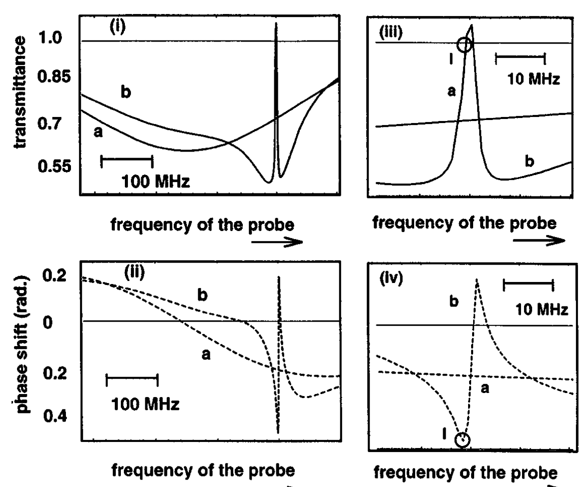

FIG. 2. Calculated absorption coefficient (i,iii) and phase shift (ii,iv) of the probe field in the Rb vapor. In all figures curve (a) corresponds to the case without driving and pumping fields, curve (b) corresponds to the case when these two fields are present. Parameters for numerical modeling are $\Omega = 16$ MHz, Doppler width is 500 MHz, incoherent pump corresponds to $r = 0.5$ MHz, and magnetic field $H_x = 2 \times 10^{-4}$ T. Natural linewidth of the Rb D_1 absorption line is 5.4 MHz.

by modulating the diode's input current with noise. The size of the pump beam was on the order of 3×5 mm.

The resonant index of refraction was determined via phase-shift measurements using a Mach-Zehnder interferometer (see Fig. 3). When the interferometer arms are of nearly equal length the difference current of photodetectors PD1 and PD2 is proportional to

$$S_{\mathrm{signal}} \sim I_0 \exp(G/2) \sin(\phi + \phi_0), \tag{7}$$

where I_0 is the intensity of the probe field and ϕ_0 is a constant phase shift determined by a balance of the interferometer arms. A piezoelectric transducer is used to vary ϕ_0 and thus obtain the scaling of the signal.

The first experiments were carried out with the low Rb density, such that maximum probe absorption was on the order of 40%. Under these conditions the signal given by Eq. (7) is the phase shift of the probe field in the Rb cell, provided that the arms of the interferometer are appropriately balanced. The absorption of the probe field was detected simultaneously. Without the drive and incoherent pump fields, the absorption and the phase shift show the ordinary features of Doppler-broadened two-level atomic resonance (curve *a* in Fig. 4). When the drive field is present and tuned within the Doppler profile, the absorption spectrum of the probe laser exhibits a narrow transmission peak at the frequency corresponding to that of a two-photon resonance condition. When the incoherent pump is added the narrow gain peak appears (curve *b* in Fig. 4). If the probe field is detuned slightly from the two-photon resonance (point *I* in Fig. 4) the medium is transparent; i.e., it has neither loss nor gain.

The phase shift, which is proportional to the real part of susceptibility, is shown in Figs. 4(ii) and 4(iv). It can be quite large in the transparency region. In particular, we note that at point *I* the change in the refractive index exceeds that found in the same system without the driving field. That is, the real part of the susceptibility is enhanced at the point where the absorption vanishes. We note the good agreement between the theoretical predictions of Fig. 2 and the experimental observations of Fig. 4.

Having observed and analyzed the absorption and dispersion in an optically thin medium we were also able to demonstrate the enhancement of refractive index in an optically dense medium. To accomplish this phase shift and absorption measurements were carried out for a variety of temperatures. Figure 5 shows the value of the phase shift as a function of atomic density. For low temperatures the phase shift in the coherently prepared transparent Rb vapor is somewhat larger than the maximum phase shift in the usual Rb resonant vapor. For temperatures $T > 80\,^{\circ}\mathrm{C}$ ($N > 3 \times 10^{11}\ \mathrm{cm}^{-3}$) the Rb absorbs 100% of the probe light near the resonance, i.e., in the region where the refractive index has its maximum. When the drive and incoherent pump are present the system is transparent and displays large phase shifts. The inset in Fig. 5 shows the transmission of the probe laser (curve *A*) and corresponding output S_{sig} of the phase shift measurement (curve *B*) at a Rb cell temperature $\sim 87\,^{\circ}\mathrm{C}$. In the presence of coherent drive and incoherent pumping, the narrow transparency peak appears (curve *a*) on an otherwise flat, completely absorbing background. The corresponding phase shift (curve *b*) measurements display the oscillations which correspond to large values of phase shift. Under the present experimental conditions we observed the phase shifts up to $\approx 7\pi$ at the point of complete transparency, which corresponds to the resonant change in the refractive index $\Delta n \sim 10^{-4}$. We note that such a phase shift is not observable in a usual absorber in the immediate vicinity of resonance because of the resonant absorption.

We remark here that quite large phase shifts accompanied by relatively good transparency can be obtained in a usual nondriven atomic system (e.g., within the Rb D_1 line without coherent preparation) at large detunings. This happens because the real part of the susceptibility χ' decreases slower than χ'' with detuning. However, for large detunings, χ' of a usual absorber increases with density much slower than the resonant χ'. In particular, for $N \sim 10^{12}\ \mathrm{cm}^{-3}$ we observed that the absorption of the

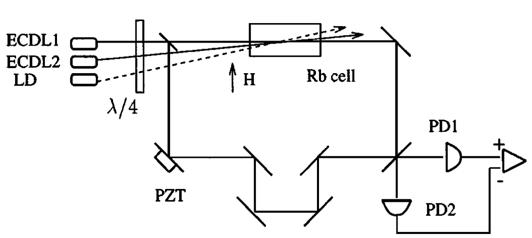

FIG. 3. Schematic of experimental setup. ECDL1 and ECDL2 are probe and driving lasers, respectively; LD is an incoherent pump laser.

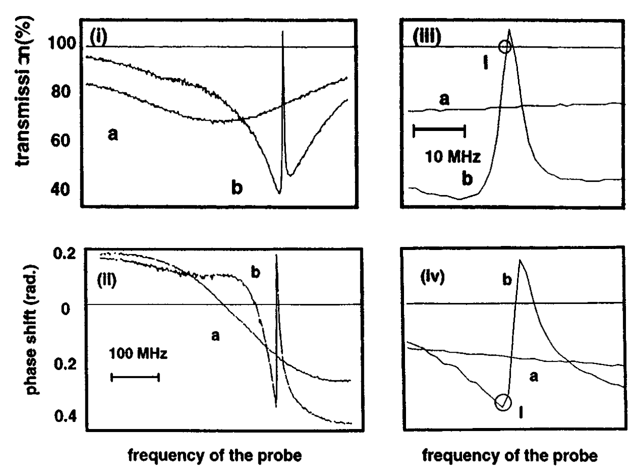

FIG. 4. Transmitted intensity (i,iii) of the probe light in the Rb cell and the difference current of photodetectors, converted into the phase shift (ii,iv). In all figures curve (*a*) was observed without driving and pumping fields, curve (*b*) was observed when those two fields were present.

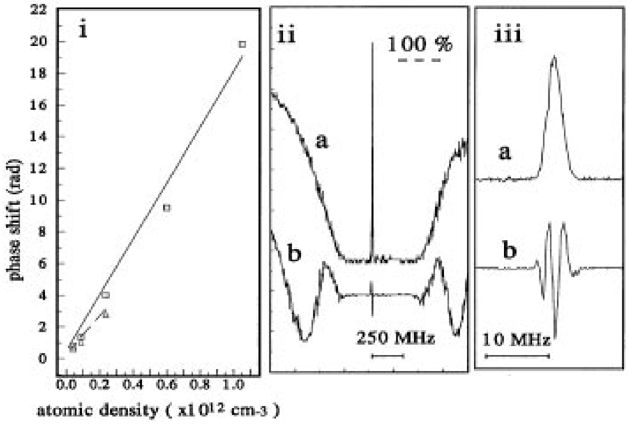

FIG. 5. (i) The phase shifts in Rb vapor as a function of a vapor density. □ is the phase shift in coherently prepared Rb at the point of complete transparency; △ is the maximum value of phase shift in the same system without coherent preparation. (ii) Transmission of the probe (curve *a*) and corresponding quadrature component of interferometer signal (curve *b*) in an optically dense Rb vapor with driving and incoherent pumping fields present. (iii) The enlarged central part of (ii).

probe field in the nondriven Rb vapor is essentially zero at detuning ~15 GHz. In this region the measured phase shift was approximately 1.5 rad.

In conclusion, we have presented an experimental demonstration of an enhancement of the refractive index in a coherently prepared atomic medium accompanied by vanishing absorption. The experimental results were predicted by and are in good agreement with a detailed theoretical analysis.

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[15] In such a configuration probe and driving fields form three effective Λ systems with the Rb D_1 manifold [Fig. 1(b)]. Note that the situation would be different if we used fields with linear polarizations: In that case fields form only two effective Λ systems (since the transition 2, $m_F = 0 \rightarrow 2' m_F = 0$ is forbidden) which implies a large absorption of the probe field on the transition 1, $m_F = 0 \rightarrow 2' m_F = 0$.