Refractive Index Enhancement with Vanishing Absorption in an Atomic Vapor

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We report a proof-of-principle experiment where the refractive index of an atomic vapor is enhanced while maintaining vanishing absorption of the beam. The key idea is to drive alkali atoms in a vapor with appropriate control lasers and induce a gain resonance and an absorption resonance for a probe beam in a two-photon Raman configuration. The strength and the position of these two resonances can be manipulated by changing the parameters of the control lasers. By using the interference between these two resonances, we obtain an enhanced refractive index without an increase in the absorption.

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Since the birth of quantum and nonlinear optics, one of the key challenges has been if one can achieve a very large refractive index for a laser beam [1]. A key application of a large refractive index is to optical imaging science. It is well known that the wavelength of light inside a refractive medium is $\lambda = \lambda_0/n$, where λ_0 is the wavelength in free space and n is the refractive index. A large refractive index, therefore, corresponds to a reduced wavelength inside the medium and enhanced imaging resolution. Another important application of a large refractive index is to optical lithography where the smallest feature size of a lithographic mask is determined by the wavelength of light. A simple and an efficient way to achieve a large refractive index is likely to have significant practical implications since lithographic resolution currently determines the size and the processing power of every semiconductor integrated circuit.

A laser beam which is tuned close to an atomic resonance can experience a large refractive index. As an example, the refractive index for a gas at a pressure of 1 torr can reach values as high as 10. However, such a large index is usually accompanied by large absorption and the effect is not useful. This is because, near an optical resonance, the real and imaginary parts of the optical susceptibility are of the same order. It was first pointed out by Scully that, by using interference in a three state atomic system, it is possible to obtain a large refractive index with negligible absorption [2,3]. The pioneering work of Scully was extended to different configurations by Fleischhauer and colleagues [4–6]. Although these ideas were experimentally demonstrated in a Rb vapor cell by Zibrov *et al.* [7], it has not yet been possible to achieve a refractive index in a vapor that is large enough to be of practical importance. In this Letter, we report a proof-of-principle experiment that demonstrates a new approach to this long-standing problem [8,9]. By utilizing the interference of two Raman resonances, we show that the refractive index of a laser beam that is very far detuned from an electronic resonance can be enhanced while maintaining vanishing absorption.

Before proceeding with a detailed description of our experiment, we note that in recent years, a number of counter-intuitive effects in driven atomic systems have been predicted and demonstrated [10–22]. Of particular importance is the demonstration of slow light [12–14], stopped light [15–17], fast and backward light [18,19], and enhanced nonlinearities using electromagnetically induced transparency [20–22]. Broadly, most of these effects can be thought as engineering the variation (the slope) of the refractive index as a function of frequency. In contrast, our technique allows us to engineer the actual value of the refractive index while maintaining vanishing absorption to the beam. In this sense, our approach complements the existing techniques in modifying the optical response of an atomic medium.

In our experiment, we follow the suggestion of Yavuz *et al.* [8] which has recently been extended to high alkali densities by Kocharovskaya and colleagues [9]. The essential features of this idea are presented in Fig. 1. It is well-known that, the interference of the dipole moments of an absorptive resonance and an amplifying resonance can lead to an enhanced refractive index with vanishing absorption [4]. As shown in Fig. 1(a), the most straightforward way to realize such an interference would be to have two different two-level atomic species. In practice, such a multiple two-level atom scheme has not yet been realized since it is difficult to find two different atomic species with very close and easily tunable resonance frequencies. The key idea that we experimentally demonstrate in this work is that such a multiple two-level scheme can be realized by using Raman resonances in far-off resonant atomic systems. As shown in Fig. 1(b), with an atom starting in the ground state $|g\rangle$, a Raman transition involves absorption of one photon and emission of another photon of different frequency such that the two-photon resonance condition is satisfied. By changing the order at which the probe laser, E_p, is involved in the process, such a Raman resonance can be made absorptive or amplifying. This approach circumvents the difficulties of the scheme of Fig. 1(a). The two Raman transition frequencies can be arbitrarily different since we have the freedom to choose the frequencies of the control lasers, E_{c1} and E_{c2}. Figure 1(c) shows the real and the imaginary parts of the susceptibility for the case of

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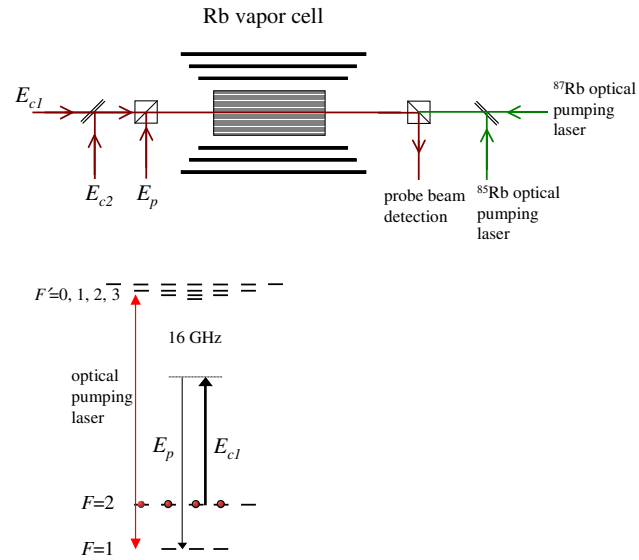

equal strength of the two resonances. The real part of the susceptibility, χ', and therefore the refractive index, peaks at the point of vanishing imaginary part, χ''.

We demonstrate the scheme of Fig. 1(b) in two isotopes of atomic Rubidium, ^{87}Rb and ^{85}Rb. Figure 2 shows the simplified experimental setup and the relevant energy level diagrams. We work with a triple layer magnetically shielded and temperature controlled natural abundance Rb (28% ^{87}Rb, 72% ^{85}Rb) vapor cell. The vapor cell is $L = 7.5$ cm long and contains 10 torr of Neon (Ne) as a buffer gas. The temperature of the vapor cell is kept at $T = 90$ degrees Celsius which gives a total atomic density of $N \approx 2.4 \times 10^{12}/\text{cm}^3$. We use $F = 2 \rightarrow F = 1$ and $F = 2 \rightarrow F = 3$ hyperfine transitions in ^{87}Rb and ^{85}Rb respectively (in ground electronic state $5S_{1/2}$). For Raman transition between the hyperfine states, we utilize far-off resonant excitation through the excited electronic state $5P_{3/2}$ ($D2$ line) near a wavelength of $\lambda = 780.2$ nm. We drive the two Raman transitions with a weak probe beam, E_p, and two strong control lasers, E_{c1} and E_{c2}. The frequency differences between respective beams are tuned close to hyperfine transition frequencies, $\omega_p - \omega_{c1} \approx 6.834$ and $\omega_p - \omega_{c2} \approx 3.035$ GHz. The frequency of the probe laser beam, ω_p, is detuned ≈ 16 GHz from the $D2$ line in ^{87}Rb.

We start the experiment by optically pumping both of the atomic species to the $F = 2$ hyperfine state manifold. This is achieved by two optical pumping lasers locked to $F = 1 \rightarrow F' = 2$ transition in ^{87}Rb and $F = 3 \rightarrow F' = 3$ transition in ^{85}Rb, respectively. Each optical pumping beam has a power of about 0.5 W and is obtained by seeding a semiconductor tapered amplifier with an external cavity diode laser. The optical pumping beams have a collimated beam radius of ≈ 3 mm and counter-propagate the three experimental beams as shown in Fig. 2(a).

The three experimental beams, E_p, E_{c1}, and E_{c2}, are derived from a single external cavity master diode laser. The output of this laser is appropriately shifted by three high frequency acousto-optic modulators (AOM) in parallel to produce the desired frequency spacing between respective beams. After the AOMs, the control beams are amplified by tapered amplifiers to achieve the required power levels. The frequency of each of the three laser beams can be tuned by changing the modulation frequency of the AOMs. This setup gives us complete control over the position of the two Raman resonances when we scan the frequency of the probe laser beam. Further details regarding our laser system can be found in our previous publica-

pinhole. The pinhole transmission is appropriately normalized to take into account the gain or absorption of the beam. In essence, the intensity measurement of Fig. 4(a) probes the imaginary part of the susceptibility (χ'') whereas the pinhole transmission measurement of Fig. 4(b) probes the real part of the susceptibility (χ'). For this data, we adjust the control laser frequencies such that the two resonances are separated by 0.2 MHz which is roughly twice the width of each resonance. The pinhole transmission, and therefore the refractive index, is maximized at the point of vanishing gain or loss on the beam. The solid line in Fig. 4(a) is a fit to the data that assumes each resonance to be Lorentzian. The solid line in Fig. 4(b) is the calculated refractive index based on the fit to the data of Fig. 4(a). We see good qualitative agreement between the refractive index calculation and the pinhole transmission data.

Currently, our experiment suffers from an undesired cross coupling of the two optical pumping processes. The ^{87}Rb optical pumping laser tries to pump the ^{85}Rb atoms to the wrong state and vice versa. Because of this cross coupling, our optical pumping efficiency is low and the observed refractive index in our experiment ($\Delta n \approx 10^{-6}$) is about an order of magnitude lower than the ideal limit for our experimental parameters. This problem can be solved by going to a different laser system and pumping both species to their lower energy hyperfine ground states ($F = 1$ in ^{87}Rb and $F = 2$ in ^{85}Rb) which we plan to implement in the near future.

One key advantage of our scheme is that by increasing the intensities of the control lasers one can obtain the maximum possible refractive index of the medium while maintaining vanishing absorption [8,9]. The maximum refractive index of an alkali vapor at a density of about 10^{15} atoms/cm^3 is ≈ 2. The collisional broadening of the excited state prevents the refractive index to be increased further in an alkali vapor [9]. Demonstrating a refractive index of 2 in an alkali vapor with vanishing absorption may have significant practical implications. Since the wavelength of light is reduced inside the vapor, an optical microscope with 2 times better resolution may be constructed. Similarly, a better lithographic resolution may be obtained inside the medium. Furthermore, our scheme is general and may be applied to molecular species such as molecular N_2. In molecules, Raman transitions between vibrational and rotational states may be utilized. More favorable pressure broadening coefficients of molecular gases may allow refractive index values approaching 10 to be obtained.

We also note the relation of our work to the growing field of negative index (left-handed) metamaterials [25]. Recently, there has been a number of theoretical suggestions that achieve negative refractive index with low loss in the optical region of the spectrum in atomic systems [26–28]. We note that, by changing the order of gain and absorption resonances [such as in Fig. 3(c) and Fig. 3(d)], our scheme may also be used to obtain a reduced refractive index ($n < 1$). At large densities, this effect may be used to obtain a negative value of the dielectric permittivity, $\epsilon = 1 + \chi' < 0$, with vanishing absorption. While this is not sufficient to construct a negative index medium (since one also needs negative permeability, $\mu < 0$), it may be possible to combine our technique with a strong magnetic resonance in another species and obtain a negative refractive index. A detailed analysis of this effect will be among our future investigations.

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