Resonant enhancement of refractive index in a cascade scheme

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(*Received 15 March 2001; revision received 15 May 2001*)

Abstract. The refractive index of coherently driven rubidium vapour is experimentally investigated in a three-level cascade configuration using a selective reflection technique. The maximum measured resonant change in the refractive index is $\Delta n \simeq 0.1$. The selective reflection is accompanied by a four-wave-mixing process that can reach ~90% efficiency.

1. Introduction

In transparent solids the index of refraction can be as high as 3 for visible light. An important question is whether or not it is possible to enhance the index of refraction in resonant atomic media and what maximum value can be achieved in principle. In a two-level medium, the index of refraction can be very high near resonance compared with the nonresonant case. However, this high index is accompanied by large resonant absorption, which makes it less useful for many experiments and applications.

In coherently driven atomic media the situation might be different because of the strong modification of susceptibility that occurs when atoms are prepared in a coherent superposition of states. In particular, one of the most striking phenomena associated with this quantum coherence is the ability to produce a large resonant index of refraction with vanishing absorption [1–10]. Preparation of matter in such a state may provide a new type of optical material of interest both in its own right, and in applications to laser particle acceleration, optical microscopy, atomic tests of fundamental interactions, and precision magnetometry.

Proof-of-principle experiments demonstrating resonant enhancement of the index of refraction without absorption in Λ-type level configuration were reported in [8]. The experiment was performed in a warm vapour of ^{87}Rb ($T \sim 90^\circ$C). The resonant index of refraction was determined via phase-shift measurements using a Mach–Zehnder interferometer. Phase shifts up to 7π at the point of complete transparency were observed, which corresponds to resonant change of the refractive index $\Delta n \simeq 10^{-4}$. Such a phase shift is not normally observable in the vicinity of resonance because of the absorption.

Journal of Modern Optics ISSN 0950–0340 print/ISSN 1362–3044 online
http://www.tandf.co.uk/journals
DOI: 10.1080/09500340110088588

Currently, the greatest enhancement of the index of refraction reported for an atomic vapour is still much less than unity, but research is continuing and many theoretical schemes have been proposed. Examples are four-level systems in which double-dark resonance is achieved [9, 10], an asymmetric double quantum well [11], and index enhancement due to local field effects [12, 13].

In an atomic vapour at low density, the index of refraction scales linearly with the density. However, simply increasing the density of atomic vapour and applying more powerful drive radiation to suppress absorption, which appears to be an obvious way to increase the index, is not a simple task. The increase of the density is often accompanied by an increase of collective incoherent processes, such as radiation trapping [14] and collisional broadening [15–23], that can be detrimental to obtaining a high index of refraction.

In this paper we study selective reflection [16, 20–28] of laser radiation from coherently driven rubidium vapour at an atomic vapour density $\sim 10^{15}\,\mathrm{cm}^{-3}$. It is shown that some coherence is preserved in a thin boundary layer even in this dense atomic gas, and that it leads to resonant enhancement of the refractive index. The optical selective reflection due to coherence grows to as high as 8%, meaning that the refractive index of the coherent atomic media is increased by $\Delta n \sim 0.1$. The experiment is performed for a cascade-level configuration which is different from the experiment [8] performed for a Λ-level configuration.

The atomic coherence also leads to four-wave-mixing (FWM) processes. The drive and probe fields combine to create a coherence grating [29, 30] that can then effectively scatter the probe field. FWM reflection with an efficiency up to 90% was observed. This is especially interesting because the atomic vapour has a high optical density and the linear absorption length for the probe field is approximately equal to the probe wavelength. Applying the drive radiation allows the probe light to penetrate deeper into the medium and to increase the interaction length necessary for an efficient FWM process.

2. Experimental set-up

In the experiment two extended-cavity diode lasers are used. The probe laser is tuned to the ^{87}Rb D_2 line ($\lambda = 780\,\mathrm{nm}$) in the vicinity of the cycling transition $5S_{1/2},\ F=2 \rightarrow 5P_{3/2},\ F'=3$. The drive laser is tuned to the $5P_{3/2},\ F'=3 \rightarrow 5D_{5/2},\ F''=2,3,4$ ($\lambda' = 776$ nm) transitions. Both lasers are linearly polarized. Polarizations of the probe and drive are set orthogonal to each other.

The intensity of the probe laser is $1.3\,\mathrm{mW\,cm^{-2}}$, which corresponds to a Rabi frequency $\Omega_p \simeq 2.4\,\mathrm{MHz}$ (natural population decay rate for the D_2 line is $\gamma \simeq 6\,\mathrm{MHz}$). The drive laser is amplified by a tapered amplifier up to $3\,\mathrm{W\,cm^{-2}}$, which corresponds to a Rabi frequency $\Omega_d \simeq 28\,\mathrm{MHz}$ (natural decay rate for $5P_{3/2} \rightarrow 5D_{5/2}$ transition is $\gamma' \simeq 0.7\,\mathrm{MHz}$). Both laser beams have diameters of $\sim 0.04\,\mathrm{cm}$ and divergence of $\sim 10^{-3}$ rad.

The laser beams are superimposed on the inner window surface of a glass cell containing a natural mixture of Rb (figure 1). The length of the cell is $L = 2.5\,\mathrm{cm}$ and its diameter is $D = 2.5\,\mathrm{cm}$. The density of the Rb vapour is determined by its vapour pressure at the temperature of the cell. In our experiment the cell temperature is $\sim 150^{\circ}\mathrm{C}$, which corresponds to a concentration of Rb atoms of $N \sim 10^{15}\mathrm{cm}^{-3}$ and a Doppler width of $\Delta_D \simeq 600\,\mathrm{MHz}$. The homogeneous broadening of the probe transition due to atomic collisions is $\gamma_{coll} \simeq 100\,\mathrm{MHz}$.

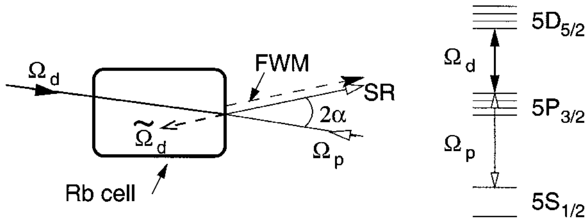

Figure 1. Experimental set-up and scheme of atomic levels.

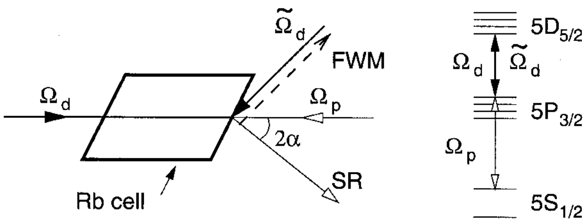

Figure 2. Experimental set-up to study four-wave-mixing process on the interface between glass and rubidium vapour.

Probe and drive lasers are counter-propagating to reduce the Doppler width of the two-photon transition. Under this condition the residual width is ~ 3 MHz which is comparable with the homogeneous width of the drive transition. It is interesting to mention here, that the homogeneous width of the drive transition is much less than the width of the probe transition. Because Rb vapour is completely transparent for the drive field, the drive field is sent through the cell. In the presence of the driving field the absorption length of the probe field is about 3×10^{-4} cm.

In addition to selective reflection we have observed four-wave mixing (FWM) in the atomic vapour. Weak FWM (several per cent reflection of probe field power) can be observed in the scheme shown at figure 1. This process appears because of reflection of some of the driving field from the surface of the cell window. Power of the reflected drive radiation $\tilde{\Omega}_d$ is about 4% of the power of incident drive Ω_d, but it is enough to create a coherence grating that generates FWM and changes the selective reflection spectrum (FWM and selective reflection propagates in the same direction for the geometry shown in figure 1).

To study this FWM more precisely we split the drive into two equal parts and send them into an atomic cell as shown in figure 2. Windows of this cell have antireflection coatings and are tilted at the Brewster's angle. The polarization of the forward drive field Ω_d is chosen such that this field does not experience reflection at the cell windows. This measurement allows one to subtract the signal due to the FWM process from the selective reflection signal and, thus, measure the index of refraction of the medium.

3. Basic results and discussion

Typical selective reflection spectra are shown in figures 3 and 4. For resonant tuning of the drive field the width of the spectral features is determined by the

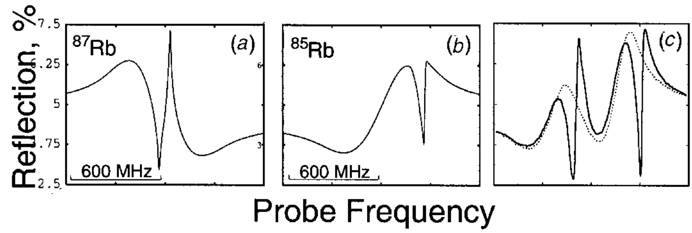

Figure 3. Calculated (a, b) and experimental (c) selective reflection spectrum when the probe frequency is swept around D_2 line of ^{85}Rb $(F = 3 \rightarrow F')$ and ^{87}Rb $(F = 2 \rightarrow F')$. The drive frequency is fixed and tuned to the $5P_{3/2}$, $F' = 3 \rightarrow 5D_{5/2}$, $F'' = 4$ transition of ^{87}Rb. Dotted experimental curve (c) shows the reflection spectrum without the driving field. Incident angle is equal to $\alpha = 0.1$ rad.

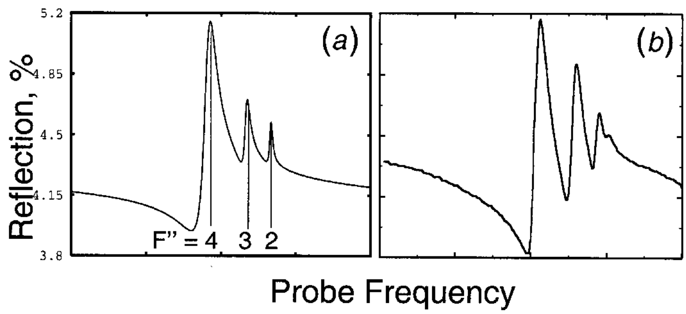

Figure 4. Calculated (a) and experimental (b) selective reflection spectrum when the probe frequency is swept in the area red shifted (~ 800 MHz) from the D_2 line of ^{87}Rb $(F = 2 \rightarrow F')$. The drive frequency is fixed and tuned to the $5P_{3/2}$, $F' = 3 \rightarrow 5D_{5/2}$ transition of ^{87}Rb. Observed resonances correspond to the fine structure of the $5D_{5/2}$ state. Incident angle is equal to $\alpha = 0.1$ rad.

radiation broadening. Because the cell contains a natural mixture of rubidium, we see D_2 lines for both ^{85}Rb and ^{87}Rb isotopes.

For the far detuned drive field the selective reflection features are very narrow. In this case we are able to observe the fine structure of the $5D_{5/2}$ state, namely transitions to $F'' = 4, 3, 2$. This is possible because radiation broadening decreases as the detuning increase. The minimum width of the spectral lines is determined by residual Doppler as well as the homogeneous width of the two-photon transition.

It is useful to draw attention to the fourth resonance observed in the experimental spectrum (figure 4 (b)). This resonance corresponds to the $5D_{5/2}$, $F'' = 1$ level and appears because the Doppler broadening is broader than the hyperfine structure of the $5P_{3/2}$ state. In other words, two-photon transitions $5S_{1/2}$, $F = 2 \rightarrow 5P_{3/2}$, $F = 3 \rightarrow 5D_{5/2}$, $F'' = 2, 3, 4$ are allowed while the transition $5S_{1/2}$, $F = 2 \rightarrow 5P_{3/2}$, $F = 3 \rightarrow 5D_{5/2}$, $F'' = 1$ is forbidden. The fourth line appears as the result of $5S_{1/2}$, $F = 2 \rightarrow 5P_{3/2}$, $F = 2 \rightarrow 5D_{5/2}$, $F'' = 1$ transition. This 'extra' resonance is much weaker because both drive and probe fields are detuned from resonance with the corresponding one-photon atomic transitions. Only fast atoms that belong to the tails of the Doppler distribution take part in the interaction. Our numerical model includes $5S_{1/2}$, $F = 2$, $5P_{3/2}$, $F = 3$, and

$5D_{5/2}$, $F'' = 2, 3, 4$ levels only and, as the result, the fourth spectral feature is absent in the theoretical plot (figure 4 (*a*)).

To describe selective reflection theoretically the Rb atoms are modelled by a five-level system including $5S_{1/2}$, $F = 2$, $5P_{3/2}$, $F = 3$, and $5D_{5/2}$, $F'' = 2, 3, 4$ levels. The Bloch equations for the atomic populations and polarizations are solved in steady state. Taking into account collisional as well as Doppler broadening the index of refraction (surface admittance) of the medium, $\Delta n = \Delta n' + i\Delta n''$ is found, and finally the Fresnel refraction R is calculated as

$$R = \left| \frac{n_0 - 1 - \Delta n}{n_0 + 1 + \Delta n} \right|^2, \tag{1}$$

where $n_0 = 1.5$ is the glass refractive index. Equation (1) is valid for small incidence angles α.

A simple analysis of the reflected probe radiation allows study of the index of refraction of the Rb vapour. Because $|\Delta n| \gg 1$, the reflection can be estimated as $R \approx 0.04 - \Delta n'$.

Direct application of equation (1) is inappropriate because the selective reflection is altered by FWM processes resulted from the drive reflection on the cell window. For the experiment shown in figure 1 the efficiency of FWM can be 1- 3%. This significantly changes the inferred value of refractive index if FWM is not taken into account. To study FWM we use a cell with Brewster windows (figure 2). This allows us to avoid reflection of the drive field Ω_d from the inside surface of the cell window and, therefore, to measure separately selective reflection and FWM with good precision. Accurate calculations show that in this experiment $\Delta n' \simeq 0.1$.

The FWM process can be strong (see figure 5). For a red detuned drive field the efficiency can reach $\sim 90\%$. The coherence grating for the FWM is created by Ω_d and Ω_p while $\tilde{\Omega}_d$ is scattered by the grating. The detailed theory of this effect will be presented elsewhere.

Increasing the temperature of the atomic vapour leads to a decrease of both the two-photon resonance feature of selective reflection and also the FWM signal, whereas the single photon reflection continues to increase. The maximum observed selective reflection due to one-photon processes was about 10%, which corresponds to an index of refraction $\Delta n' \approx 0.1$. The two-photon coherent pro-

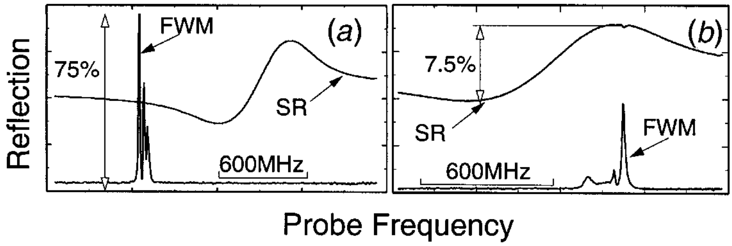

Figure 5. Selective reflection and four-wave-mixing spectra for the configuration shown in figure 2. (*a*) The drive field is red shifted (~ 950 MHz) and (*b*) the drive field is blue shifted (~ 300 MHz) from the $5P_{3/2}$, $F' = 3 \rightarrow 5D_{5/2}$ transition of ^{87}Rb. Forward Ω_d and backward $\tilde{\Omega}_d$ drive fields have the same intensity and polarization. Incident angle is set to Brewster's angle. Vertical scales for both pictures are the same.

cesses vanish for high atomic densities due to collisional broadening and radiation trapping, which destroy coherence effects.

4. Conclusion

It has been shown that coherence processes are present in very dense, up to $N = 3 \times 10^{15}\,\mathrm{cm}^{-3}$, atomic medium. This coherence leads to significant change in the refractive index of the atomic vapour and to efficient four-wave mixing. However, the problem of realizing a medium with a high index of refraction and small absorption remains unsolved. The absorptive part of the refractive index measured in these experiments is reduced, but still of the same order of magnitude as the refractive part. Therefore, we cannot say that we have a high index of refraction without absorption. Nonetheless, the experiment gives some new insight on the question of the maximum possible refraction obtainable with a resonant atomic vapour.

Acknowledgments

The authors gratefully acknowledge useful discussions with V. A. Sautenkov and Y. V. Rostovtsev, and support from the Office of Naval Research, the National Science Foundation, and the Welch Foundation.

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