Steep atomic dispersion induced by velocity-selective optical pumping

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Abstract: We demonstrate a method of preparation of broadband sign-reversible dispersion in alkali vapour based on velocity-selective optical pumping. The refractive index in Rb vapour has been measured using a heterodyne method. The magnitudes of the normal and anomalous dispersion, which are almost constant over a spectral region of approximately 40 MHz, can lead to a reduced ($V_g \approx c/230$) or negative ($V_g \approx -c/27$) group velocity of light.

OCIS codes: 300.6310 Spectroscopy, heterodyne; (270.5530) Nonlinear optics: Pulse propagation and temporal solitons

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1. Introduction

The possibility of large modification of the group velocity of a light pulse in dispersive media has been extensively studied both theoretically and experimentally and remarkable results have been achieved [1, 2, 3, 4]. The interest in this subject is mainly driven by a need for tunable optical delay lines, a key element making construction of an all-optical internet possible. This may enhance significantly the speed of signal processing in telecommunications [2, 5].

The peak of a pulse travels at the group velocity $V_g = c/[n + \nu(dn/d\nu)]$, where n is the refractive index and ν is the central frequency of the pulse. The group velocity can be positive or negative resulting in pulse delay (slow light) or pulse advance (fast light) [2]. In resonant atomic media, which are popular for proof-of-principle experiments, steep changes of the refractive index occur in the vicinity of optical transitions. Ground-state hyperfine or Zeeman coherence can dramatically enhance the dispersion of the sample [6, 7], but in a spectral region that is normally narrower than the natural width of the optical transition. This high selectivity imposes a lower limit on the pulse duration if it is essential for the shape of the pulse to be preserved. Wide-bandwidth delay lines based on linear-responding media may be considered as an alternative approach [8, 9]. In this case the dispersion is almost constant over a rather broad spectral region (of order 1 GHz), but the magnitude of the dispersion $dn/d\nu$ is rather modest.

In this Letter we demonstrate that steep normal and anomalous dispersion in an alkali vapour can be obtained using velocity-selective optical pumping. In the case of large inhomogeneous broadening this process may result in significant modification of the absorptive and, consequently, the dispersive properties of an atomic medium over a spectral region wider than the natural width, but narrower than Doppler width. This is an intermediate situation in terms of spectral selectivity and magnitude of dispersion compared with the linear and ground-state coherence mechanisms. This idea is similar to slow light generation in solid state media based on the hole burning method in inhomogeneously broadened absorption lines [10]. A modified experimental realization of the hole-burning method in a hot rubidium vapour was reported in [11], where large fractional pulse delays were achieved.

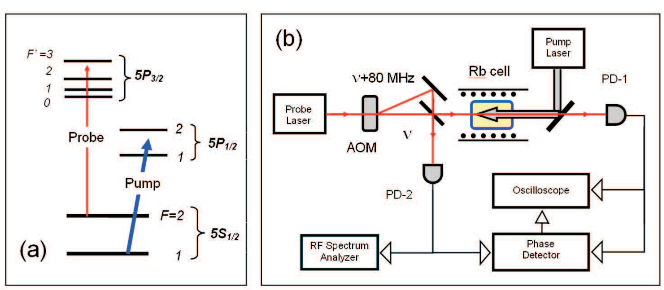

Fig. 1. (a) Energy levels diagram of the ^{87}Rb D_1 and D_2 lines. (b) Scheme of the experimental setup.

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2. Doppler-free resonances with frequency independent beams

Let us consider the origin of Doppler-free absorption and refractive index resonances in Rb vapour due to velocity-selective optical pumping, taking into account that the connection between absorption and dispersion is well established [2]. Using two independent lasers we can separate the frequencies of the pump and probe light and employ optical transitions which belong to different lines, such as the D_1 and D_2 lines. Doppler-free spectroscopy of alkali atoms with frequency independent pump and probe beams have some distinctive features compared to the commonly used saturation spectroscopy. For example, the number of Doppler-free resonances depends on the geometry of the experiment; also their spectral position and polarity depend on the frequency of the pump radiation. Typical peculiarities of Doppler-free spectroscopy of the Rb D_2 lines with co-propagating beams have recently been discussed [12]. Here we present results of dispersion measurements of Rb vapour for the D_2 line, when the pump radiation is tuned to the D_1 line (Fig. 1(a)).

Consider the case when fixed frequency pump radiation ν_{L1} is tuned to the transition $5S_{1/2}(F=1)-5P_{1/2}(F'=2)$ in ^{87}Rb, where F and F' are the total angular momenta of the ground and excited states, respectively. Due to the inhomogeneous broadening sub-MHz linewidth laser radiation interacts with a group of Rb atoms which have a certain velocity projection in the direction of the beam:

$$V_z = 2\pi(\nu_{L1}-\nu_{12})/k_1, \tag{1}$$

where $k_1 = 2\pi/\lambda_{D1}$ is the wave vector of the laser light tuned to the D_1 line. We denote the frequency of the transition $(F=n)-(F'=m)$ as ν_{nm}; thus ν_{12} is the resonant frequency of the $5S_{1/2}(F=1)-5P_{1/2}(F'=2)$ transition. Strong modification of the steady-state population in this group occurs if the pump beam is intense enough to produce efficient optical pumping:

$$I > I_S = [(1+\tau/T)/(1+2\tau/T)]\hbar\omega/\sigma_0 T, \tag{2}$$

where I_S is the optical-pumping saturation intensity [13], σ_0 is the absorption cross-section of the transition, T is the interaction time and τ is the upper level lifetime. Under these conditions the number of atoms in the resonant velocity group for the level F=1 is reduced, while for the F=2 level the population is enhanced. The redistribution of population among different ground-state sublevels probed by a laser with scanning frequency results in Doppler-free absorption resonances. The polarity of these resonances depends on the transitions used for probing. If the probe frequency ν_{L2} is swept across the Doppler profile for the $5S_{1/2}(F=2)-5P_{3/2}$ line, three peaks in the absorption profile can be observed, while for the $5S_{1/2}(F=1)-5P_{3/2}$ line three dips in the probe absorption occur. The resonant conditions for counter propagating probe and pump beams are:

$$\nu_{L2} = \nu_{2i} - k_2 V_z/2\pi; \;\; \nu_{L2} = \nu_{1i} - k_2 V_z/2\pi, \tag{3}$$

where $k_2 = 2\pi/\lambda_{D_2}$ is the wave vector for the D_2 line and ν_{2i} $(i=1,\,2,\,3)$ and ν_{1i} $(i=0,\,1,\,2)$ are the resonant frequencies of the optical transitions $5S_{1/2}(F=2)-5P_{3/2}$ and $5S_{1/2}(F=1)-5P_{3/2}$, respectively.

Such a method can be applied to all alkali atoms. In the case of ^{87}Rb or Cs atoms where the hyperfine splitting of the upper $5P_{1/2}$ or $6P_{1/2}$ state is larger than the Doppler width at room temperature we can ignore redistributions produced by adjacent hyperfine transitions within the D_1 line, such as $5S_{1/2}(F=1)-5P_{1/2}(F'=1)$ for ^{87}Rb.

3. Experimental setup

The dispersive properties of Rb vapour have been studied using the RF heterodyne technique [14]. The phase shift of the probe wave is proportional to the refractive index n: $\Delta\Phi = 2\pi(n-$

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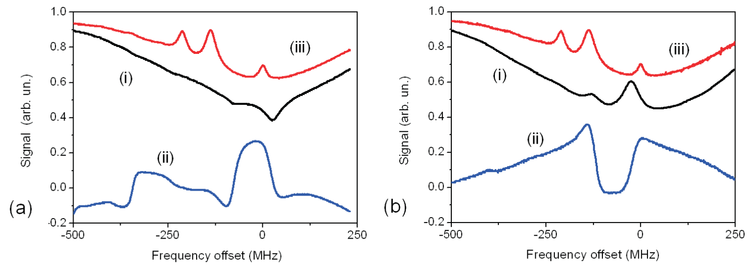

Fig. 2. Effect of velocity selective optical pumping on the transmission and dispersion of ^{87}Rb vapour probed on the $5S_{1/2}(F=2)-5P_{3/2}$ transitions. Figures (a) and (b) correspond to the optical frequency of the pump laser ν_{L1} tuned to the $5S_{1/2}(F=1)-5P_{1/2}(F'=2)$ and $5S_{1/2}(F=2)-5P_{1/2}(F'=2)$ transitions, respectively. Transmission of the bi-chromatic probe beam (i), phase detector output (ii) and probe transmission through the reference cell (iii) are presented as a function of probe frequency offset from the $5S_{1/2}(F=2)-5P_{3/2}(F'=3)$ transition.

$1)L/c$, where L is the length of the atomic sample. This allows us to measure the refractive index variation Δn and the dispersion $dn/d\nu$ of the medium.

The optical scheme of the experiment is shown in Fig. 1(b). Two extended cavity diode lasers tuned to the D_1 and D_2 absorption lines are used as sources of resonant light. The spectral width of the laser radiation is in the sub-MHz range. Doppler-free absorption resonances obtained in auxiliary cells, which are not shown in Fig. 1(b), are used as frequency references for the probe and pump lasers.

A 5 cm-long glass cell containing a natural mixture of Rb isotopes is heated up to 80 ^0C, at which temperature the Rb density is $N \approx 1.2 \times 10^{12} cm^{-3}$. The stray magnetic field produced by the heater is less than 0.5 Gauss. A counter propagating pump-probe scheme has been chosen to eliminate a parasitic contribution to the signal from the pump light. The diffracted output from the AOM, driven at 80 MHz, is used as an off-resonant reference beam for the heterodyne scheme, while the zero-order beam is used as the signal component. The two beams are superimposed on a beam splitter and sent to the signal PD-1 and reference PD-2 photodiodes. Typical transverse dimensions of the probe and pump beams are about 3-4 mm. The maximum intensity of the linearly polarized pump beam in the cell is $I_{L1} \approx 80\ mW/cm^2$, while that of the bi-chromatic probe is $I_{L2} \approx 1.5\ mW/cm^2$ with the intensity ratio between the frequency components 1 to 3.

We observe the spectral dependences of the absorption and refractive index simultaneously. The phase detector output and the signal, which is proportional to the probe intensity transmitted through the Rb cell, are recorded on a digital oscilloscope.

4. Heterodyne dispersion measurements and discussion

The disturbances of the velocity distribution of atoms in the ground state results in enhanced dispersion as shown in Fig. 2(a),(b). The absorption of the probe (curve (i)) is increased in the vicinity of the $5S_{1/2}(F=2)-5P_{3/2}(F'=3)$ transition because of velocity-selective pumping from the $F=1$ into the $F=2$ sublevels by the fixed frequency pump light tuned on the $5S_{1/2}(F=1)-5P_{1/2}(F'=2)$ transition (Fig. 2(a)). When the pump laser is tuned to the $5S_{1/2}(F=2)-5P_{1/2}(F'=2)$ transition, the absorption is reduced (curve (i)) due to velocity-selective depopulation (curve (i), Fig. 2(b)). These absorption resonances have a full width at half maximum (FWHM) linewidth of approximately 40 MHz, which is considerably larger

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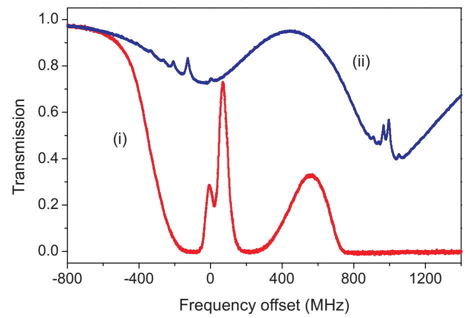

Fig. 3. (i) Curve (i) shows the transmission of the bi-chromatic probe radiation through the vapour cell at 75^0C containing a natural mixture of Rb isotopes in the vicinity of the transitions $5S_{1/2}(F=3)-5P_{3/2}$ for ^{85}Rb and $5S_{1/2}(F=2)-5P_{3/2}$ for ^{87}Rb. Curve (ii) represents the transmission through the reference cell.

than the natural linewidth ($\Gamma = 2\pi \times 6$ MHz) due to power broadening. Variations of the probe phase have a dispersive form. The slopes of the phase variations at the absorption peak and dip have opposite signs, indicating opposite atomic dispersion. One spectral region of steep atomic dispersion corresponds to two phase resonances with opposite polarity because of sequential scanning of the two-component probe across the region.

At rather low atomic density ($N \approx 2\times 10^{10} cm^{-3}$) the contrasts of the Doppler-free peaks and dips and, consequently, the absolute values of the normal and anomalous dispersion are comparable. The difference between the two cases becomes significant at higher atomic density when the absorption is high even without the pump light. Indeed, an initially opaque atomic medium can be made partly transparent by the velocity-selective depopulation (curve (i) in Fig. 3) and high contrast Doppler-free resonances result in large variations of refractive index. But there is an obvious limit for absorption enhancement. Above 75 ^0C, at which temperature the Rb density is $N \approx 8\times 10^{11} cm^{-3}$, the cell absorbs almost 100% of the resonant probe beam.

The phase shift is calibrated by replacing the signal from the reference photodiode with an output of the same amplitude from an auxiliary 80-MHz generator. The amplitudes are balanced using an RF spectrum analyser. Because of the small frequency offset between the auxiliary generator and the AOM driver the output of the phase detector exhibits oscillations. The peak-to-peak signal variation that corresponds to phase variations of π is employed for scaling the phase signal (Fig. 4(a)).

The phase variation in a dense atomic sample in the vicinity of the Doppler-free transparency window can exceed π, as shown in Fig. 4(b). The phase curve displays an oscillatory behaviour. We observe a phase shift of up to 3π, which corresponds to a refractive index variation $\Delta n \approx 2.4\times 10^{-5}$. Taking into account the width of the transparency window of 40 MHz, we find the dispersion $dn/d\nu \approx 0.6\times 10^{-12} Hz^{-1}$. This value of normal dispersion may result in light pulse propagation with a reduced group velocity $V_g \approx c/230$. It is worth mentioning that enhancement of the index of refraction and a phase shift of up to 7π was observed by Zibrov et al [15] using quantum coherence and interference effects.

We also find that the total refractive index variation Δn is approximately four times smaller if the pump laser is tuned to the $5S_{1/2}(F=2)-5P_{1/2}(F'=1)$ transition instead of the $5S_{1/2}(F=2)-5P_{1/2}(F'=2)$ transition.

In the case of anomalous dispersion, obtained when the pump laser is tuned to the $5S_{1/2}(F=1)-5P_{1/2}(F'=2)$ transition, the maximum observed variation of the refractive index is $\Delta n \approx 1.4\times 10^{-6}$. This value is lower compared to the case of reduced absorption because it has been obtained at lower atomic density ($N \approx 2.5\times 10^{10} cm^{-3}$). The absolute value of the anomalous

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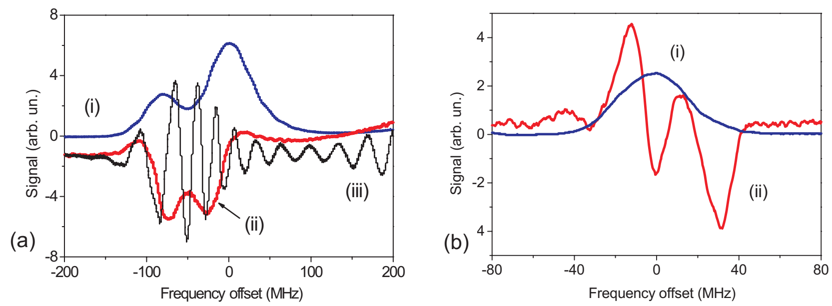

Fig. 4. Bi-chromatic probe transmission (i), phase detector output (ii) induced by atomic dispersion and phase calibration curve (iii) as the probe laser frequency is scanned across the ^{87}Rb $5S_{1/2}(F=2)-5P_{3/2}(F'=3)$ transition, with the pump frequency tuned to resonance with the $5S_{1/2}(F=2)-5P_{1/2}(F'=2)$ transition. Figs. (a) and (b) correspond to a Rb density of $N=7\times10^{11}cm^{-3}$ and $N=1.2\times10^{12}cm^{-3}$, respectively.

dispersion $dn/d\nu$ is also smaller, about $7\times10^{-14}Hz^{-1}$. However it is large enough to achieve a negative group velocity ($V_g \approx -c/27$).

The spectral region of steep dispersion, both normal and anomalous, can be easily tuned by changing the frequency of the pump laser within the Doppler profile. We have also investigated the intensity dependence of the dispersion. The contrasts of the Doppler-free absorption resonances and, consequently, refractive index variations depend on the intensity of the pump radiation. The transparency window for the probe becomes larger and wider at higher intensity of the pump beam as expected. For a given atomic density, the refractive index variation Δn grows until the pump intensity $I=(2-4)\ mW/cm^2$ and then saturates, while the dispersion $dn/d\nu$ reaches the maximum magnitude in this intensity range and after that decreases steadily with intensity due to power broadening.

5. Conclusion

The process of velocity-selective optical pumping has been used to produce steep normal and anomalous dispersion in Rb vapour. The spectral region of enhanced dispersion is wider than that provided by methods based on ground-state coherence. Furthermore, the approach based on optical pumping is less sensitive to stray magnetic fields and laser linewidth.

The refractive index has been measured using the RF heterodyne method. The obtained values of maximum dispersion, which are almost constant over a spectral region of approximately 40 MHz, may lead to a reduced ($V_g \approx c/230$), enhanced ($V_g > c$) or negative ($V_g \approx -c/27$) group velocity of light. It is interesting to note that a good agreement between the value of dispersion measured by the heterodyne method and the group velocity obtained by the pulse propagation method for a fast-light medium was recently reported [16].

Light-induced dispersive properties can be easily controlled by the frequency and intensity of the pump light. Even a reversal of sign of the dispersion may be achieved by tuning the pump laser from one transition to another. Media with controlled anomalous dispersion could allow the realization of a so-called white-light optical cavity, which combines high internal build-up and broadband response [17].

This method can be applied to all alkali atoms. Some peculiarities of Doppler-free spectroscopy with different frequency probe and pump lasers have been discussed. Atomic media with enhanced and laser controlled dispersion can also be considered as a convenient playground for modeling and constructing tuneable delay lines.

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