Enhanced nondegenerate four-wave mixing owing to electromagnetically induced transparency in a spectral hole-burning crystal

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Received March 20, 1997

We have demonstrated electromagnetically induced transparency (EIT) in an inhomogeneously broadened spectral hole-burning system of Pr^{3+}-doped Y_2SiO_5 at 6 K. We have also shown enhancement of four-wave mixing under conditions of reduced absorption. This demonstration opens the possibilities of pursuing EIT applications such as high-resolution optical image processing and optical data storage in solids. © 1997 Optical Society of America

In this Letter we demonstrate suppressed absorption and enhanced nondegenerate four-wave mixing (NDFWM) based on electromagnetically induced transparency[1] (EIT) in a spectral hole-burning crystal at 6 K with a cw laser. EIT has attracted much recent interest because it permits the use of optically dense, resonant media for nonlinear-optical applications. However, previous observations of EIT were restricted to vapors and atomic beams. In these materials the diffusion of atoms is a major problem and limits application, especially when one is working with images. Spectral hole-burning solid materials have many similarities to vapors and beams in that the optical transitions are between energy levels of isolated atoms but have the advantages of permanent optical pumping (storage capability) and the absence of diffusion. These advantages open the door to numerous additional applications of EIT to spectroscopy,[2] image processing,[3] real-time holography,[4] and optical memory.[5]

Atom–field interactions in three-level Λ-type systems have been studied extensively in the past several years. Briefly, when two resonant laser beams interact with a Λ-type three-level system, the atoms can be trapped in a coherent superposition state, which is decoupled from the excited state so that the laser beam is no longer absorbed by the medium. This is called coherent population trapping or EIT in the context of optically dense media. Since the first observation of coherent population trapping in a sodium beam,[6] many potential applications have been explored. The most recent applications of EIT include nonlinear-optical processes for frequency conversion,[7,8] lasers without population inversion,[9] Raman-excited spin-echo data storage,[10] and high-gain, low-intensity optical phase conjugation.[11] In the area of frequency conversion, Harris *et al.* proposed the creation of nonlinear media with resonantly enhanced nonlinear susceptibilities and at the same time reduced absorption at the resonant transition frequency owing to EIT.[12] Recent experiments showed enhanced second-harmonic generation[7] and third-harmonic generation[8] by use of EIT. In the area of lasers without population inversion there was an experimental demonstration in Rb vapor,[13] and there is current interest in finding suitable solid material. For Raman-excited spin echoes, a sodium atomic beam experiment was performed to verify the basic physics. This process is being considered for high-temperature persistent spectral hole-burning memories, because spin coherences tend to survive longer than optical coherences at high temperatures.[14] In the area of optical phase conjugation by four-wave mixing Hemmer *et al.* recently observed high-gain phase conjugation in sodium vapor by using coherent population trapping.[11] Li and Xiao also observed the enhancement of NDFWM as a result of the EIT effect in a Λ-type three-level system of Rb.[15]

Figure 1 is an energy-level diagram of Pr^{3+}-doped Y_2SiO_5 (Pr:YSO) for NDFWM. Our system consists of 0.05-at. % Pr:YSO in which Pr ions act as an inhomogeneously broadened six-level atomic system. Pr:YSO is known as a good material for optical spectral hole-burning data storage because of its large ratio of inhomogeneous to homogeneous widths and long optical pumping lifetime. For this study, the relevant optical transition is $^3H_4 \rightarrow {}^1D_2$, which has a frequency of 605.7 nm at site 1. Optical population relaxation time T_1 and transverse relaxation time T_2 are 164 and 111 μs, respectively, at 1.4 K, and the optical inhomogeneous width is ~4 GHz.[16] The ground and the excited states each have three Kramers doublets.[17] The transition frequencies of the ground-state Kramers doublets are 10.2 and 17.3 MHz. T_2 between Kramers doublets has not been directly measured, but T_1 is as long as several minutes. The inhomogeneous widths of the transitions between ground-state Kramers doublets was measured by optically detected nuclear magnetic resonance to be less than 80 kHz.[17] For the 10.2-MHz transition, we measured an inhomogeneous linewidth of ~40 kHz. In general, EIT implies the use of high-optical-density material. Pr:YSO is a good material for such studies because it can have a high

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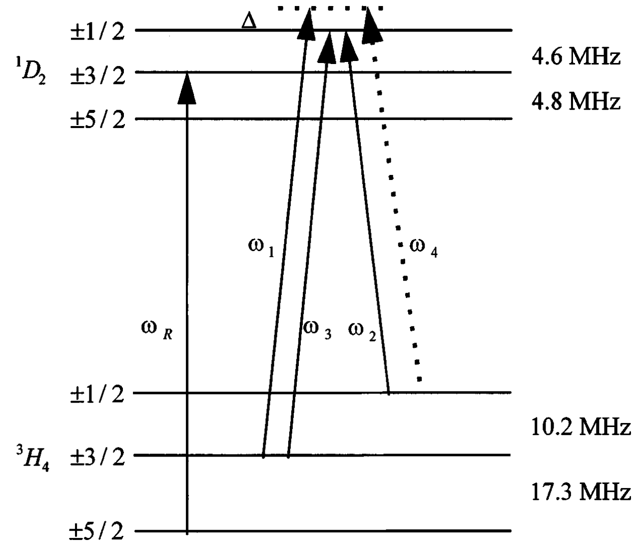

Fig. 1. Energy-level diagram of Pr:YSO for four-wave mixing.

optical density (when optical pump effects are neglected). Measured absorption coefficients[17] of Pr:YSO are as large as 10 cm^{-1}.

Laser fields ω_2 and ω_3 in Fig. 1 can be viewed as write beams, which create a ground-state coherence by EIT. Laser field ω_1 then acts as a read beam to generate a beam ω_4, while laser field ω_R acts as a repump beam. The read beam ω_1 is blue detuned from the write beam ω_3 by a frequency of $\Delta = 100\,\mathrm{kHz}$, which is larger than the inhomogeneous width of any transition between Kramers doublets. The generated beam ω_4 must satisfy the phase-matching condition $\mathbf{k}_4 = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3$. Here it should be noted that Fig. 1 applies to only a small subset of Pr ions. Because of the large inhomogeneous broadening, each laser field can pump other transitions in the manifold for a subset of Pr ions that have the appropriate transition frequency. However, owing to persistent optical pumping (spectral hole burning), only Pr ions that have simultaneous coupling of the optical fields to all three ground states contribute to the signal. One can easily verify this spectral selectivity by scanning the repump beam frequency over a range spanning all three excited states. When the repump beam is resonant with one of the excited states, the probe beam is absorbed; otherwise the crystal is transparent.

Figure 2 shows the schematic experimental setup for observing EIT and NDFWM in Pr:YSO. We used a frequency-stabilized ring dye laser (Coherent Model 699) pumped by a Spectra-Physics argon-ion laser. The laser is cw, and its estimated linewidth is 3 MHz. We used acousto-optic modulators driven by frequency synthesizers to make four different coherent laser beams as shown. To match the setup in Fig. 1, the write beams ω_2 and ω_3 were upshifted 54.8 and 65.0 MHz from the laser frequency by AO-2 and AO-3, respectively. Read beam ω_1 and repump beam ω_R were upshifted 65.1 and 77.7 MHz by AO-1 and AO-R, respectively. To avoid any possible contribution of coherent interaction between the repump and the two-photon transitions, we chose the repump transition as shown in Fig. 1. All four laser beams were focused into the sample by a 40-cm focal-length lens. The beam diameters ($1/e$ in intensity) were ~150 μm in the crystal. Each applied laser intensity of ω_1, ω_2, ω_3, and ω_R was roughly 20, 40, 60, and 60 $\mathrm{W/cm^2}$, respectively. The angle between the two write beams was ~70 mrad. The hole-burning crystal Pr:YSO was inside the helium cryostat. We kept the temperature at 6 K. If the temperature was >10 K, no spectral hole burning was observed. The crystal was 3 mm in diameter and 10 mm long. Its optical b axis was unknown, and the absorption was very polarization sensitive. Under the conditions described, we measured a maximum absorption for the laser beam ω_2 of 85%, which was limited by the strength of the repump beam.

To measure the effective optical linewidth of the repumped atom source, we reduced the laser intensities by a factor of 10 and blocked read beam ω_1. The absorption width, seen as ω_2, that is scanned depends on both the homogeneous width of the optical transition and the applied laser field linewidth, which can be broadened by laser jitter. In our case the laser linewidth dominated because it was much broader than the optical homogeneous width. Figure 3 shows the absorption spectrum of beam ω_2 with beams ω_3 and ω_R tuned to near the center of the 4-GHz inhomogeneously broadened absorption profile of Pr:YSO. No two-photon transition is seen because of insufficient laser intensity. The FWHM of the absorption curve is ~3.5 MHz, which is similar to the estimated laser linewidth based on laser-jitter measurements made with a Fabry–Perot spectrum analyzer. The absorption curve disappears when repump field ω_R is blocked, as expected. The noise seen in the data is due to the

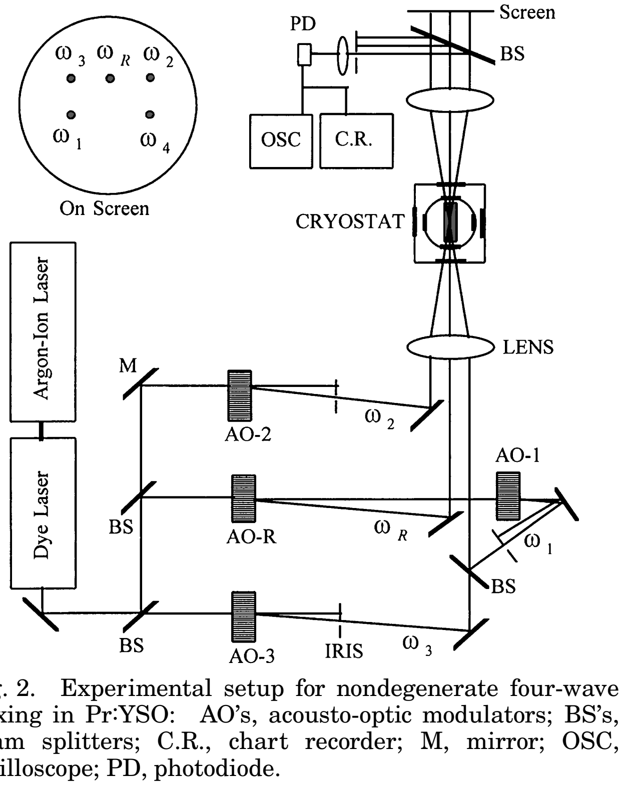

Fig. 2. Experimental setup for nondegenerate four-wave mixing in Pr:YSO: AO's, acousto-optic modulators; BS's, beam splitters; C.R., chart recorder; M, mirror; OSC, oscilloscope; PD, photodiode.

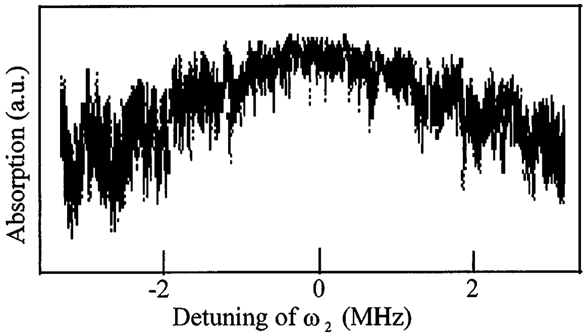

Fig. 3. Absorption spectrum of beam ω_2 with weak beams ω_3 and ω_R.

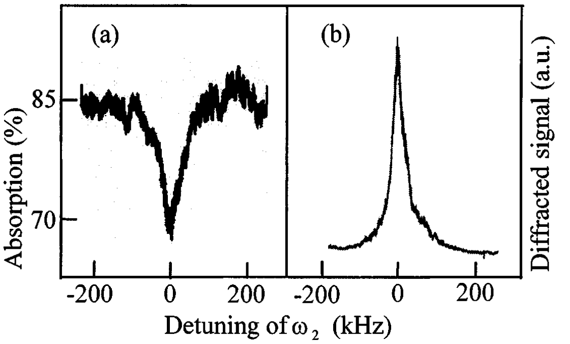

Fig. 4. (a) Absorption spectrum of beam ω_2 with strong beams ω_3 and ω_R. (b) NDFWM generation in the inhomogeneously broadened spectral hole-burning crystal Pr:YSO at 6 K. The intensities of ω_1, ω_2, ω_3, and ω_R are 20, 40, 60, and 60 W/cm^2, respectively.

combined effects of laser jitter and the spectral hole-burning process.

To demonstrate EIT explicitly, we increased the laser intensities of ω_3 and ω_R and again scanned the weak laser beam ω_2 across the two-photon resonance frequency, keeping ω_3 and ω_R fixed. In Fig. 4(a), we demonstrate that the absorption of ω_2 at two-photon resonance frequency is reduced from 85% to 70%. The FWHM of the reduced absorption is 60 kHz, which is much narrower than the laser linewidth and comparable with the spin-state inhomogeneous linewidth of $^3H_4(\pm 3/2) \leftrightarrow {}^3H_4(\pm 1/2)$, which is ~40 kHz. This sub-laser-jitter linewidth is the signature of the two-photon transition responsible for EIT. Thus the reduced absorption is due to the EIT effect.

For the four-wave mixing experiment, we added read beam ω_1, which was 100-kHz blue-detuned from write beam ω_3 (see Fig. 1). Even though this detuning is much smaller than the laser jitter, ω_1 and ω_3 are never degenerate, because the fields are generated with AO's from a single laser (see Fig. 2). The alignments of laser beams ω_1, ω_2, and ω_3 satisfy the Bragg condition for the generation of ω_4 at the position indicated inside the circle in Fig. 2, which shows the spatial positions of the four laser beams on a screen. Figure 4(b) shows the intensity of diffracted signal ω_4 as a function of the detuning of ω_2 from two-photon resonances. Diffracted signal ω_4 is enhanced greatly as write beam ω_2 is tuned through two-photon resonance, and its FWHM is 40 kHz. The diffraction efficiency, i.e., the intensity ratio of ω_4 to ω_1, is ~1%. Again, sub-laser-jitter linewidth is the evidence of EIT. Therefore the enhancement of nonlinear generation in NDFWM is due to the ground-state coherence produced by EIT.

In summary, we have observed electromagnetically induced transparency in an inhomogeneously broadened spectral hole-burning system of Pr:YSO at 6 K. We have shown simultaneous reduction of absorption and enhancement of four-wave mixing. Pr:YSO is an attractive alternative to the use of vapors because it opens the possibility of pursuing EIT applications, such as high-resolution nonlinear-optical image processing and optical data storage, in solid media.

We acknowledge discussions with S. Ezekiel of the Massachusetts Institute of Technology. This study was supported by Rome Laboratory (grant F30602-96-2-0100) and by the U.S. Air Force Office of Scientific Research (grant F49620-96-1-0395).

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