

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

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We have demonstrated electromagnetically induced transparency (EIT) in an inhomogeneously broadened spectral hole-burning system of  $\text{Pr}^{3+}$ -doped  $\text{Y}_2\text{SiO}_5$  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<sup>1</sup> (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,<sup>2</sup> image processing,<sup>3</sup> real-time holography,<sup>4</sup> and optical memory.<sup>5</sup>

Atom-field interactions in three-level  $\Lambda$ -type systems have been studied extensively in the past several years. Briefly, when two resonant laser beams interact with a  $\Lambda$ -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,<sup>6</sup> many potential applications have been explored. The most recent applications of EIT include nonlinear-optical processes for frequency conversion,<sup>7,8</sup> lasers without population inversion,<sup>9</sup> Raman-excited spin-echo data storage,<sup>10</sup> and high-gain, low-intensity optical phase conjugation.<sup>11</sup> 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.<sup>12</sup> Recent experiments showed enhanced second-harmonic gen-

eration<sup>7</sup> and third-harmonic generation<sup>8</sup> by use of EIT. In the area of lasers without population inversion there was an experimental demonstration in Rb vapor,<sup>13</sup> 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.<sup>14</sup> 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.<sup>11</sup> Li and Xiao also observed the enhancement of NDFWM as a result of the EIT effect in a  $\Lambda$ -type three-level system of Rb.<sup>15</sup>

Figure 1 is an energy-level diagram of  $\text{Pr}^{3+}$ -doped  $\text{Y}_2\text{SiO}_5$  (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  $\mu\text{s}$ , respectively, at 1.4 K, and the optical inhomogeneous width is  $\sim 4$  GHz.<sup>16</sup> The ground and the excited states each have three Kramers doublets.<sup>17</sup> 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.<sup>17</sup> For the 10.2-MHz transition, we measured an inhomogeneous linewidth of  $\sim 40$  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

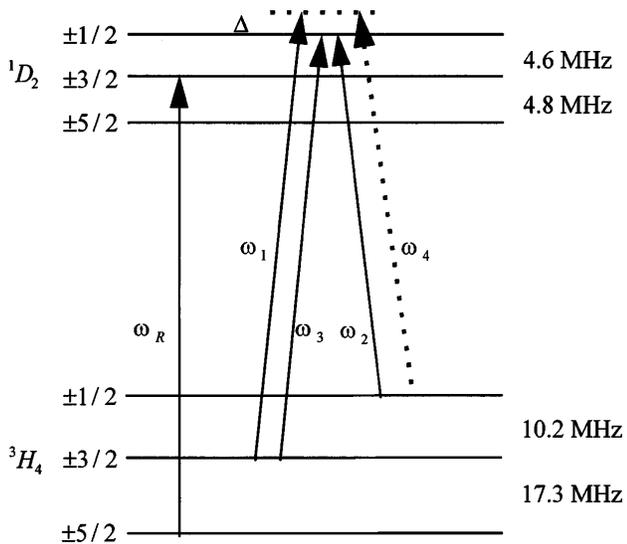


Fig. 1. Energy-level diagram of Pr:YSO for four-wave mixing.

optical density (when optical pump effects are neglected). Measured absorption coefficients<sup>17</sup> of Pr:YSO are as large as  $10 \text{ cm}^{-1}$ .

Laser fields  $\omega_2$  and  $\omega_3$  in Fig. 1 can be viewed as write beams, which create a ground-state coherence by EIT. Laser field  $\omega_1$  then acts as a read beam to generate a beam  $\omega_4$ , while laser field  $\omega_R$  acts as a repump beam. The read beam  $\omega_1$  is blue detuned from the write beam  $\omega_3$  by a frequency of  $\Delta = 100 \text{ kHz}$ , which is larger than the inhomogeneous width of any transition between Kramers doublets. The generated beam  $\omega_4$  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  $\omega_2$  and  $\omega_3$  were upshifted 54.8 and 65.0 MHz from the laser frequency by AO-2 and AO-3, respectively. Read beam  $\omega_1$  and repump beam  $\omega_R$  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  $\sim 150 \mu\text{m}$  in the crystal. Each applied laser intensity of  $\omega_1$ ,  $\omega_2$ ,  $\omega_3$ , and  $\omega_R$  was roughly 20, 40, 60, and  $60 \text{ W/cm}^2$ , respectively. The angle between the two write beams was  $\sim 70 \text{ mrad}$ . The hole-burning crystal Pr:YSO was inside the helium cryostat. We kept the temperature at 6 K. If the temperature was  $> 10 \text{ K}$ , 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  $\omega_2$  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  $\omega_1$ . The absorption width, seen as  $\omega_2$ , 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  $\omega_2$  with beams  $\omega_3$  and  $\omega_R$  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  $\sim 3.5 \text{ MHz}$ , 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  $\omega_R$  is blocked, as expected. The noise seen in the data is due to the

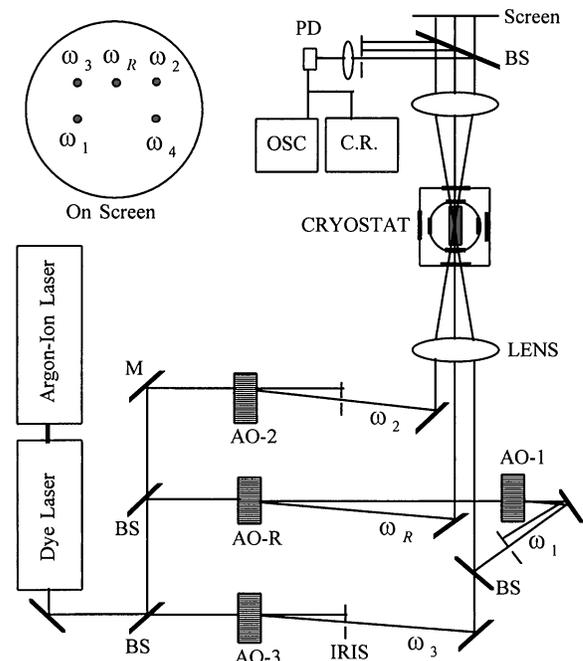


Fig. 2. Experimental setup for nondegenerate four-wave mixing in Pr:YSO: AO's, acousto-optic modulators; BS's, beam splitters; C.R., chart recorder; M, mirror; OSC, oscilloscope; PD, photodiode.

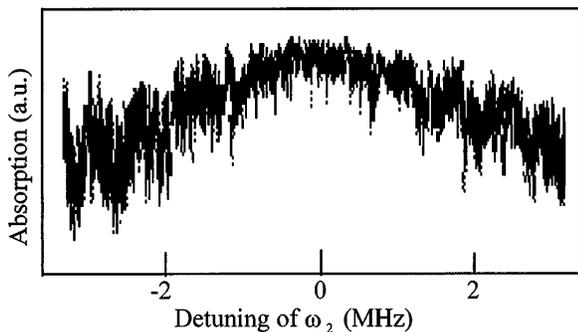


Fig. 3. Absorption spectrum of beam  $\omega_2$  with weak beams  $\omega_3$  and  $\omega_R$ .

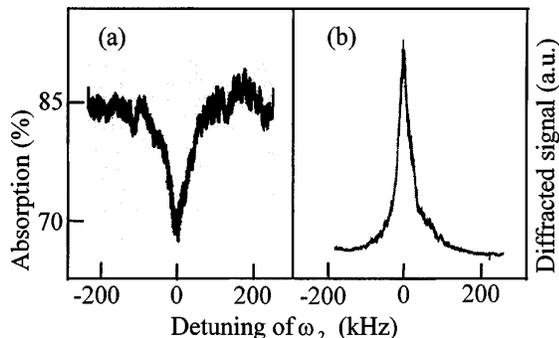


Fig. 4. (a) Absorption spectrum of beam  $\omega_2$  with strong beams  $\omega_3$  and  $\omega_R$ . (b) NDFWM generation in the inhomogeneously broadened spectral hole-burning crystal Pr:YSO at 6 K. The intensities of  $\omega_1$ ,  $\omega_2$ ,  $\omega_3$ , and  $\omega_R$  are 20, 40, 60, and 60 W/cm<sup>2</sup>, respectively.

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

To demonstrate EIT explicitly, we increased the laser intensities of  $\omega_3$  and  $\omega_R$  and again scanned the weak laser beam  $\omega_2$  across the two-photon resonance frequency, keeping  $\omega_3$  and  $\omega_R$  fixed. In Fig. 4(a), we demonstrate that the absorption of  $\omega_2$  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  $\sim 40$  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  $\omega_1$ , which was 100-kHz blue-detuned from write beam  $\omega_3$  (see Fig. 1). Even though this detuning is much smaller than the laser jitter,  $\omega_1$  and  $\omega_3$  are never degenerate, because the fields are generated with AO's from a single laser (see Fig. 2). The alignments of laser beams  $\omega_1$ ,  $\omega_2$ , and  $\omega_3$  satisfy the Bragg condition for the generation of  $\omega_4$  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  $\omega_4$  as a function of the detuning of  $\omega_2$  from two-photon resonances. Diffracted signal  $\omega_4$  is enhanced greatly as write beam  $\omega_2$  is tuned through two-photon resonance, and its FWHM is 40 kHz. The diffraction efficiency, i.e., the intensity ratio of  $\omega_4$

to  $\omega_1$ , is  $\sim 1\%$ . 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.

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