Enhancement of four-wave mixing and line narrowing by use of quantum coherence in an optically dense double-Λ solid

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We have demonstrated enhanced nondegenerate four-wave mixing by use of a resonant probe in a double-Λ system consisting of an optically dense spectral hole–burning solid. The observed probe diffraction efficiency is ~16% in amplitude at 6 K, which is higher than for an off-resonant probe in a Λ-type scheme. We have also observed two-photon coherence line narrowing, which has potential application to high-resolution spectroscopy. © 1999 Optical Society of America

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Since the first proposal of enhanced nondegenerate four-wave mixing,[1] there have been several observations of the enhancement of nonlinear optical processes by use of two-photon coherence in gas media[2–7] and solids.[8] In particular, enhancement of four-wave mixing generation in Λ-type systems is found to be large under the conditions in which coherent population trapping[9] plays an essential role.[5] Without two-photon coherence, four-wave mixing efficiency is lower on resonance, because the linear susceptibility $\mathrm{Im}[\chi^{(1)}]$ competes with the nonlinear susceptibility $\mathrm{Re}[\chi^{(3)}]$ and suppresses the nonlinear optical processes. However, by use of coherent population trapping or electromagnetically induced transparency[8,10,11] the absorption can be suppressed even at the exact resonance. Recently, nondegenerate four-wave mixing was studied by use of double-Λ systems in atomic[5,6] and molecular[7] vapors. Lu *et al.* demonstrated higher four-wave mixing efficiency in a double-Λ system than in a single-Λ system using Rb vapor.[6]

In this Letter we report enhanced nondegenerate four-wave mixing by use of a double-Λ system in an optically thick spectral hole–burning solid, Pr^{3+}-doped $\mathrm{Y_2SiO_5}$ (Pr:YSO). We have observed that the probe diffraction efficiency is 2.4% in intensity (15.6% in amplitude) at 6 K, which is higher than the efficiency (<1%) observed in a single-Λ system.[8,12] The observed diffraction efficiency obtained with the double-Λ scheme in Pr:YSO is also higher than that observed in atomic[5,6] and molecular[7] vapors. Owing to the very low matrix element (oscillator strength, $\sim 10^{-7}$) in Pr:YSO, much higher laser intensity is expected for similar diffraction efficiency observed in atomic vapors. Therefore, the observed high diffraction efficiency in a solid medium is important for potential applications such as optical memories,[12,13] high-resolution coherence spectroscopy,[14,15] lasers without population inversion,[16] and aberration correction.[17] We also report line narrowing of the four-wave mixing signal to below the inhomogeneous width of the sublevel transition. This line narrowing is due to the compression of the two-photon transparency window in an optically dense medium.[15] The observed line narrowing of the four-wave mixing signal has potential application to high-resolution spectroscopy.

Figure 1 shows an energy-level diagram of Pr:YSO. Our system consists of 0.05-at.% Pr-doped YSO in which Pr^{3+} is substituted for Y^{3+}. The relevant optical transition is $^3H_4 \rightarrow {}^1D_2$, which has a resonant frequency of 606 nm. The inhomogeneous width of the optical transition is ~4 GHz at 1.4 K.[18] Optical population decay time T_1 and transverse decay time T_2 are 164 and 111 μs, respectively, at 1.4 K.[18] The ground ($^3H_4$) and the excited ($^1D_2$) states each have three degenerate hyperfine states. The split among the ground-hyperfine states is 10.2 MHz (±1/2 ↔ ±3/2), 17.3 MHz (±3/2 ↔ ±5/2), and 27.5 MHz (±1/2 ↔ ±5/2).[18] The splitting among the excited-hyperfine states is 4.6 MHz (±1/2 ↔ ±3/2), 4.8 MHz (±3/2 ↔ ±5/2), and 9.4 MHz

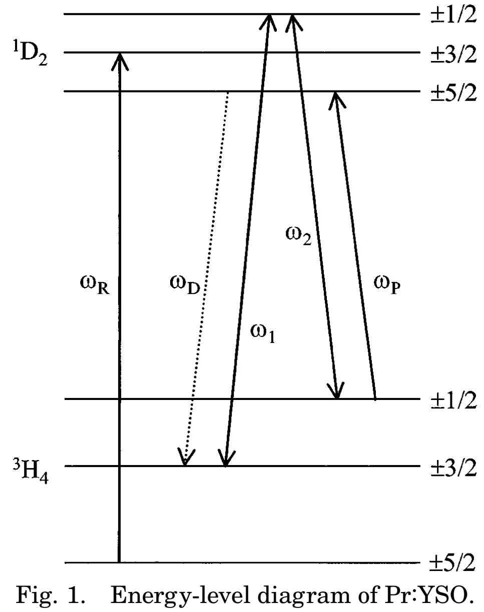

Fig. 1. Energy-level diagram of Pr:YSO.

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($\pm 1/2 \leftrightarrow \pm 5/2$).[18] Ground-state population decay time T_1 is ~100 s,[19] and spin transverse decay time T_2 for the 10.2-MHz transition is 500 ms at 6 K.[12] The spin inhomogeneous width for the 10.2-MHz transition is 30 kHz at 1.6 K.[19]

Laser fields ω_1 and ω_2 in Fig. 1 act as pump beams that create two-photon ground-state coherence through coherent population trapping. Laser field ω_P acts as a probe (read) beam, which scatters off of the two-photon coherence phase gratings created by the pump beams and generates the four-wave mixing signal, ω_D, that satisfies the phase-matching condition $\mathbf{k}_D = \mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k}_P$. Repump field ω_R is used to provide spectral selectivity in the otherwise inhomogeneously broadened system (~4-GHz inhomogeneous width). The amount of spectral selectivity provided by the repump depends on the laser jitter.

Figure 2 shows a schematic of the experimental setup for the observation of nondegenerate four-wave mixing in Pr:YSO. We use a cw frequency-stabilized ring dye laser pumped by an Ar-ion laser. The dye laser frequency jitter is ~2 MHz. We used acousto-optic modulators driven by frequency synthesizers (PTS 160) to make four different coherent laser beams, as shown in the figure. For the resonant Raman transition, pump beams ω_1 and ω_2 are downshifted by 60.0 and 70.2 MHz from the laser frequency, respectively. The probe and the repump fields are downshifted from the dye laser output by 79.6 and 47.3 MHz, respectively. All laser beams are linearly polarized and focused into the sample by a 30-cm focal-length lens, so that the focused beam diameter (e^{-1} in intensity) is ~100 μm. Pump lasers ω_1 and ω_2 have 12.5 and 9 mW of power, respectively. The power of the probe laser ω_P, is 18 mW, and the power of the repump laser, ω_R, is 11 mW. To produce laser pulses we use rf switches driven by pulse generators. The pulse width of the pump and repump beams is fixed at 1 ms. The probe pulse width is 3 μs and is delayed 2 μs after the end of the pump and repump pulses. A boxcar averager averages 30 samples of the four-wave mixing signal, ω_D. The pulse repetition rate is 50 Hz. The angle between the pump and probe fields is ~70 mrad. The spectral hole–burning crystal of Pr:YSO is inside a cryostat, and its temperature is fixed at 6 K. The size of the crystal is 3.5 mm × 4 mm × 3 mm. Its optical B axis is along the 3-mm length, and laser propagation direction is almost parallel to the optical axis.

Figure 3 shows the efficiency of ω_D as a function of the detuning of pump beam ω_2. The measured width (FWHM) is 97.0 kHz, which is two-photon power broadened. The maximum magnitude of ω_D corresponds to a diffraction efficiency of 2.4% in intensity. The actual diffraction efficiency, however, should be higher, because the beams do not copropagate and the sample is optically dense. In the limit in which length l is longer than the beam overlap length, the four-wave mixing signal intensity (I_D) is attenuated by a factor of $\exp(\alpha l)$:

$$I_D(l) \propto [\mathrm{Re}(\rho_{12})]^2 I_P(0) \exp(-\alpha l), \qquad (1)$$

where α is an absorption coefficient, ρ_{12} is the pump-pulse-excited coherence, and $I_P(0)$ is the probe intensity at $z = 0$. In relation (1), the fact that four-wave mixing signal is proportional to the product of the pump intensities (until saturated) was demonstrated in atomic and molecular vapors,[6,7] because the Raman coherence ρ_{12} is proportional to the product of pump Rabi frequencies (for weak pump beams). Although ρ_{12} should be position dependent owing to linear absorption when it is not copropagating, the pump energy used in this experiment is enough for saturation. Therefore, the Raman pump-pulse-excited spin coherence ρ_{12} is nearly position invariant and dependent on only the optical density αl. The factor $\exp(-\alpha l)$ in relation (1) is based on an unsaturated diffracted signal, which is definitely true in this experiment. This fact gives us useful information that using a thin sample is better.

For comparison, we also used an off-resonant probe beam and observed maximum diffraction efficiency of less than 1% in intensity. This was observed when the probe beam frequency was detuned ~1 MHz from the pump beam, ω_2.

To measure the minimum width of the two-photon coherence we reduced the pump powers by factors of 100 and 10 for ω_1 and ω_2, respectively. Figure 4(a) shows the resulting diffracted signal ω_D versus the detuning of the pump beam, ω_2. We lengthened the

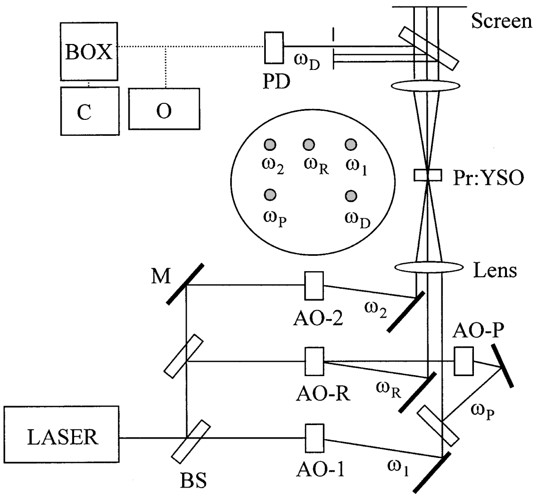

Fig. 2. Schematic of the experimental setup: AO, acousto-optic modulator; BOX, boxcar integrator; BS, beam splitter; C, chart recorder; M, mirror; PD, photodiode; O, oscilloscope. Inset, laser beams on screen.

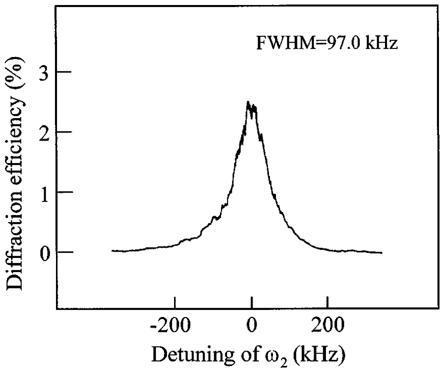

Fig. 3. Four-wave mixing signal efficiency at 6 K.

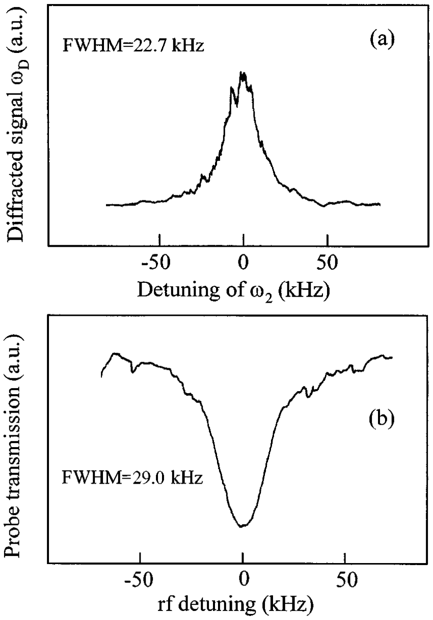

Fig. 4. (a) Line narrowing of the four-wave mixing signal and (b) spin inhomogeneous linewidth of 10.2-MHz transition by rf–optical double-resonance technique.

pump pulses to 5 ms to increase the total pulse areas to compensate lower pump intensities. We also reduced the probe power by a factor of 2. The observed width (FWHM) of the four-wave mixing signal ω_D in Fig. 4(a) is 22.7 kHz, which is narrower by a factor of 1.3 than the inhomogeneous width (29 kHz) of the 10.2-MHz transition. This line narrowing is attributed to compression of the two-photon transparency window in an optically dense medium. Such line narrowing is explained by the effects of nonlinear dispersion on parametric processes in a dense phase-coherent medium.[15] In Ref. 15 the line narrowing was shown to be approximately proportional to the square root of the optical density, which results in a factor of 1.2 when the effective length l' is used in our Pr:YSO. This factor is close to the observed narrowing.

To compare the linewidth in Fig. 4(a) with the spin inhomogeneous width, we use a conventional method, the rf–optical double-resonance technique.[20] We simultaneously applied 1-ms rf pulses (10.2 MHz) and power-attenuated optical probe pulses at a repetition rate of 30 to the sample and detected the probe transmission. To avoid rf-power broadening we reduced the rf power until the linewidth reached minimum. Figure 4(b) shows the probe transmission versus the rf detuning. The measured linewidth (FWHM) for the 10.2-MHz transition is 29 kHz. This width is broader than the measured width shown in Fig. 4(a).

In conclusion, we have observed enhanced nondegenerate four-wave mixing generation by use of a resonantly probed double-Λ scheme in an optically dense spectral hole–burning solid. The measured diffraction efficiency is 2.4% in intensity. This efficient four-wave mixing has potential application to nonlinear optical processes such as optical memories, high-resolution coherence spectroscopy, lasers without population inversion, and aberration correction. We also observed line narrowing of the four-wave mixing signal. The observed line narrowing is due to the high optical density of the medium and is useful for high-resolution spectroscopy.

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