Degenerate four-wave mixing in atomic ytterbium

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We have performed degenerate four-wave mixing experiments with cryogenically cooled atomic ytterbium. We use buffer-gas cooling to prepare high optical density samples at a temperature of 5 K, cold enough to resolve the different isotopes and hyperfine transitions. We observe four-wave mixing with cross-polarized pump and probe beams. The four-wave mixing is strongly enhanced when the laser is closely detuned from the $^{1}S_0(F = 1/2) \rightarrow ^1P_1(F' = 1/2)$ transition of the $^{171}$Yb ($I = 1/2$) isotope. © 2012 Optical Society of America

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1. INTRODUCTION

Four-wave mixing (FWM) is a nonlinear interaction between light and matter that permits the transfer of energy between four modes of the electric field via their interaction with a nonlinear medium [1,2]. Degenerate four-wave mixing (DFWM), in which all four beams are of the same frequency, has been investigated both theoretically and experimentally with atomic gases. Many different effects have been explored, including amplified reflection and transmission, phase conjugation, and oscillation [3,4]; atomic motion (angular dependence) [5,6]; optical pumping [7]; pump intensities [8–11]; pump absorption influences [12–14]; field polarization properties [15,16]; ground state Zeeman coherence [17]; and reflectivity-bandwidth product [18]. Most of these results were obtained in two-level or three-level systems.

Recently, resonance-enhanced nondegenerate FWM in an atomic medium has shown great promise for applications in the field of quantum optics. Through the use of hot or room-temperature rubidium vapors, experiments have achieved the realization of a low-noise, phase-insensitive optical amplifier [19], strong relative intensity squeezing [20–24], optically tunable delay of Einstein–Podolsky–Rosen entangled beams of light [25], high degree of spatial entanglement [26], and the violation of the Cauchy–Schwartz inequality in the high-intensity regime [27].

In this paper, we present experimental results obtained from resonance-enhanced DFWM based on the $^1S_0 \rightarrow ^1P_1$ transition of the buffer-gas-cooled ytterbium atoms. By employing the $F = 1/2$ hyperfine levels of the $^{171}$Yb isotope, we create a four-level atomic system and demonstrate enhancement of DFWM relative to the “two-level” isotopes.

This experiment is motivated not only by an interest in investigating the performance of resonance-enhanced DFWM, but also by the goal of generating nonclassical narrowband light near the atomic resonance of ytterbium. Experiments using cold $^{171}$Yb have demonstrated squeezing of the nuclear spin states via quantum nondemolition experiments with classical light [28]; the performance of these experiments would be improved if squeezed light states could be used. Similarly, ytterbium has been shown to be a favorable medium for light storage [29]; it would be of interest to combine nonclassical optical states with a quantum memory for light. Finally, we note that—due to applications in optical clocks—nonlinear optics in alkaline-earth atoms (and alkaline-earthlike atoms, such as ytterbium) is currently of great interest [30].

2. EXPERIMENTS

A. Yb Atomic Structure

We use atomic Yb as our nonlinear medium for light near the strong $^1S_0 \rightarrow ^1P_1$ resonance at 399 nm [31]. The majority of naturally occurring isotopes have zero nuclear spin ($I = 0$), and hence serve as true two-level atoms. We are interested primarily in the $^{171}$Yb isotope, which has nuclear spin $I = 1/2$, and whose relevant energy level structure is shown in Fig. 1.

In zero magnetic field, both the ground and excited $F = 1/2$ states are doubly degenerate. Linearly polarized pump beams drive the $\pi$ transition, while an orthogonally polarized probe beam drives the $\sigma^+$ and $\sigma^–$ transitions. As a parametric process, a conjugate beam should be generated from the $\sigma^+$ or $\sigma^–$ transition to form a four-level loop.

B. Atomic Medium

To resolve the $^{171}$Yb transition from the other Yb isotopes, as well as the $F = 3/2$ hyperfine state of $^{171}$Yb, it is essential to work with a cold sample of atoms. We use laser ablation and buffer-gas cooling to create the cold atoms, a technique that has previously been demonstrated to be a favorable technique for nonlinear optics [32]. We create atomic Yb by laser ablation of a solid Yb target, and cool it to a translational temperature of 5 K via cryogenic buffer-gas cooling. The Yb sample is contained within a 10 cm copper cell, as described in [33].

C. Optics

A schematic of our optics setup is shown in Fig. 2. Two strong, linearly polarized counterpropagating pump beams of equal intensity are sent through the Yb atomic ensemble. A much weaker probe beam with an orthogonal polarization is overlapped with the pump beams at a small angle. Based on the phase-matching condition, the conjugate beam should be generated in the exact opposite direction of probe beam. We use a beam splitter (BS) to separate the outgoing conjugate beam.
from the incoming probe beam. The probe and conjugate beams are measured by two photodiodes.

We note that the beams are slightly elliptical. For convenience, we describe the beams by the geometric average of their Gaussian waists.

The degenerate pump and probe beams are generated from a single frequency-doubled titanium-sapphire laser. An additional low-intensity beam (not shown in Fig. 2) is sent through the cell to measure the optical density (OD) of the atomic sample; it is not overlapped with the pump or probe beams.

**D. Generation of Conjugate Beam**

The measured conjugate signal is shown in Fig. 3 as a function of the laser frequency. The conjugate signal is strongest at the blue and red sides of the $^{171}$Yb isotope, indicating a clear evidence of resonance-enhanced DFWM. Under these conditions—and under all conditions we experimentally observed—the $F = 1/2 \rightarrow F' = 1/2$ transition of $^{171}$Yb produces a significantly stronger conjugate beam than any other Yb $^1S_0 \rightarrow ^1P_1$ transition.

We note that the $F = 1/2$ ground and excited states are not truly degenerate two-level systems; an applied magnetic field will break the degeneracy (considerably more for the excited state than for the ground state, which possesses only a nuclear magnetic moment). However, we see no evidence of any effect due to this slight nondegeneracy: we applied external magnetic fields from 0.3 to 4.5 G, and no change in the conjugate beam was observed.

This enhancement appears very promising for DFWM. Unfortunately, the conjugate power produced scales poorly with the pump power. We define the gain as the ratio of the power of the measured conjugate beam on the photodetector to the probe power before it enters the cryostat. Figure 4 shows the gain as a function of the pump power, as measured at both the blue and red sides of the $^{171}$Yb resonance. At low powers, the gain increases with increasing pump power, as expected [14]. Unexpectedly, at higher powers, the gain plateaus and rolls off, an effect that can occur at pump powers as low as 10 mW.

Higher pump powers are expected to require higher atomic densities to achieve maximum gain [14], and our cryogenic cell cannot achieve the high densities possible with ovens. However, as seen in Fig. 5, our gain is not limited by the achievable OD in our cell; we can achieve optical densities on the $^1S_0(F = 1/2) \rightarrow ^1P_1(F' = 1/2)$ $^{171}$Yb transition exceeding 100.

We attribute our gain limitations to off-resonance absorption by the other Yb isotopes and the $^{171}$Yb $^1S_0(F = 1/2) \rightarrow ^1P_1(F' = 3/2)$ transition. We would expect that these deleterious effects will become greater at higher pump powers, as higher pump powers favor larger detunings from resonance.
to achieve maximum gain [14]. Unfortunately, our current apparatus does not have isotopically enriched $^{171}$Yb. However, we can modify the atomic transition linewidths via Doppler broadening and pressure broadening by varying the temperature of the cell and the density of the helium buffer gas [33]. Figure 6 shows the dependence of the conjugate signal on the cell temperature. As the temperature increases, a dramatic reduction in the conjugate signal is observed. Similarly, increasing the helium buffer-gas density also causes a reduction of the conjugate signal.

**E. Correlation of Probe and Conjugate Beams**

We also investigate the behavior of the conjugate signal as a function of time with the laser at a fixed frequency. We stabilize the laser frequency with a dichroic atomic vapor laser lock using Yb produced in a room-temperature sputtering cell. The time behavior of the probe and conjugate signals is shown in Fig. 7. We ablate the Yb target at time $t = 0$. Once created, the atoms diffuse through the helium buffer gas to the cell walls, where they are adsorbed. This time-varying density can be clearly seen in the time-varying absorption of the probe beam, as absorption losses dominate the small gain of our experiment. The conjugate beam is more complex, as it clearly exhibits both absorption and gain; the nonzero signal on the conjugate beam at $t < 0$ and at long times is from scattered pump light.

Using data obtained in this manner, we calculated the correlations between the probe and conjugate beams as well as the spectral noise density of the individual beams and their difference. We select the data for times at which the gain is high and Fourier transform the measured photocurrents to obtain the spectral noise power density. In the absence of atoms, the probe beam’s photocurrent has a spectral noise power density within 0.7 dB of the shot noise limit at frequencies above $10^4$ Hz. Unfortunately, at times of high gain we see a significant increase in the probe beam noise level, with a spectral noise power density roughly 4 dB above shot noise. This introduction of excess noise is common for atomic samples [20, 23]. Unfortunately, due to our low gain and low detector quantum efficiency, the conjugate beam is too weak to overcome this excess noise [34]. We subtract the probe and conjugate beam photocurrents and Fourier transform the result to obtain the intensity correlations between the conjugate and probe. While the probe and conjugate beams do exhibit correlations (the spectral noise power density of the difference of their photocurrents is less than the sum of their individual spectral noise power densities) their relative intensity noise remains above the standard quantum limit.

**3. CONCLUSION AND DISCUSSION**

We have performed DFWM experiments with buffer-gas-cooled atomic Yb. We have shown that, for the generation of a conjugate beam, different Yb isotopes work quite differently. We observe a conjugate beam only when the laser is closely detuned from the $^{1}S_0(F = 1/2) \rightarrow ^{1}P_1(F = 1/2)$ transition of the $^{171}$Yb ($I = 1/2$) isotope. We have achieved up to 20% gain; if corrected for optics losses, the gain would be 50%. We also observe correlations between probe and conjugate beams; however, because of the gain limitations of our system, we only observe classical correlations. Our measurements suggest that the gain could be improved in the future simply by operating at lower temperatures—buffer gas cells employing $^3$He have been able to attain temperatures below 1 K—or by using isotopically enriched $^{171}$Yb. However, these options would significantly increase the experimental cost.
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