Effect of optical pumping and Raman scattering on the degenerate four-wave mixing in coherently pumped rubidium atoms

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We present an experimental study of cw degenerate four-wave mixing (DFWM) in Doppler-broadened rubidium atoms coupled by an additional coherent field. The coupling field and the DFWM fields were arranged in a Λ-type configuration from the rubidium D_2 transitions. We found that the presence of the coherent coupling field modifies the DFWM emission: at lower intensities of the coupling field, the DFWM emission is enhanced; at higher intensities of the coupling field, the DFWM is reduced. The observed effect of the coupling laser on the DFWM emission may be explained by the direct competition between the resonance-enhanced two-photon Raman scattering and the optical-pumping-enhanced DFWM emission. [S1050-2947(96)02112-9]

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I. INTRODUCTION

Degenerate four-wave mixing (DFWM) has been extensively studied during recent years, partly because the possibility of obtaining phase conjugation using DFWM [1]. Many experiments utilized an alkali vapor as the nonlinear medium [2–6]. Detailed theoretical treatments including inhomogeneous Doppler shifts and/or the effect of the atomic collisions in a vapor cell for two- and three-level atomic systems have been developed [7–12]. The hyperfine structures of alkali atoms, together with the inhomogeneous Doppler broadening, limit the density of atoms contributing to the optical nonlinearity to be a small fraction of the total density of atoms. Also optical pumping by the DFWM lasers to a different hyperfine ground state can further reduce the population of the interacting atoms so the DFWM signal may disappear. One way to avoid the unwanted optical pumping effect is to use a cycling transition in which the excited-state atoms can only decay back to the same hyperfine level of the ground state from which they were excited [2]. Recently, Knize *et al.* reported the enhanced DFWM emission in cesium atoms by adding a hyperfine pumping laser that recycles the atoms from the noninteracting hyperfine level of the ground state back to the interacting hyperfine level of the ground state, thus allowing the DFWM to be observed at noncycling transitions [13]; Schiffer and co-workers used bichromatic DFWM laser fields sharing the same upper-excited state and observed the DFWM signal from the pressure-induced optical pumping [14]. In view of the recent interest in the study of nonlinear optical phenomena and the spectroscopic measurements modified by an independent coupling laser (control laser), here we report an experimental study of DFWM emission in an essentially collision-free, Doppler-broadened Rb atomic system with the addition of a coupling laser driving the reverse hyperfine transition. The coupling laser and the DFWM lasers form a Λ-type atomic configuration. It is interesting to note that a variety of physical phenomena may occur in a coherently coupled Λ-type atomic system, such as coherent population trapping [15], electromagnetically-induced transparency [16–19], and enhanced third-harmonic generation [20]. Recently, it has been shown theoretically that atomic coherence induced by a coupling laser may enhance four-wave-mixing emission in a three-level system [21]. For our experimental study of the DFWM emission in a Λ-type system, we found that the mutual coupling of the additional laser and the DFWM modifies the DFWM emission. As the intensity of the additional coupling laser increases from null value, the DFWM emission was first increased, and then maximized at intermediate intensities of the coupling laser. Further intensity increases of the coupling laser resulted in the reduced DFWM emission. The increases of the DFWM emission at weak to moderate intensities of the coupling laser may be explained by the reversed optical pumping such as that observed by Knize *et al.* [13]. At high intensities of the coupling laser, the reverse optical pumping is saturated, and a resonance-enhanced two-photon Raman-scattering process (two-photon-induced atomic coherence) becomes dominant in the Λ-type system. The Raman-scattering process competes with the DFWM process, which leads to the reduction of the DFWM emission.

II. EXPERIMENTAL RESULTS

The experiment was carried out on the D_2 transition of ^{87}Rb atoms. The energy level structure of the ^{87}Rb D_2 transition lines and DFWM scheme is depicted in Fig. 1. The DFWM laser fields (the forward pump, the backward pump, and the probe) couple the Rb $5S_{1/2}(F=1)\leftrightarrow 5P_{3/2}$ transition. An additional coupling laser drives the Rb $5S_{1/2}(F=2)\leftrightarrow 5P_{3/2}$ transition and induces atomic coherence between the two hyperfine ground states. The phase-conjugated DFWM signal is generated in the backward direction relative to the forward probe. The coherent coupling laser and the DFWM lasers form a quasi-three-level Λ-type system, and under appropriate conditions, lead to the atomic population trapping in the two hyperfine ground states [15].

The experimental apparatus is shown schematically in Fig. 2. A 75-mm-long rubidium vapor cell made of Pyrex glass was heated to about 40 °C at which we measured that the linear absorption coefficient at the line center of the ^{87}Rb

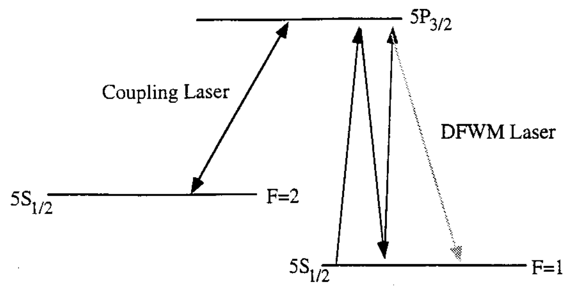

FIG. 1. Energy levels of rubidium D_2 lines and the DFWM pump and probe scheme. A population-trapping Λ-type configuration is formed by the coupling laser and the DFWM laser.

$5S_{1/2}(F=1)\leftrightarrow 5P_{3/2}$ transition was about 0.2 cm^{-1}. An external-cavity diode laser was used to produce a probe, a forward and a backward pump beam in a standard backward phase-conjugate DFWM configuration. The DFWM probe and the pump lasers were linearly polarized parallel to each other. Before incident on a 30-70 beam splitter, the diode laser beam was collimated and passed through a Faraday isolator to reduce the optical feedback from the backward pump beam. A frequency-stabilized cw Ti:sapphire laser was used as the additional coupling laser. The output beam from the Ti:sapphire laser was linearly polarized, collimated, and made to overlap with the DFWM laser beams in the Rb vapor cell. The angle between the forward pump and the forward probe beams was about 10 mrad. The generated backward DFWM signal was picked off a 50-50 beam splitter placed in the path of the forward probe beam and was detected by a photodiode. A spatial filter was used to minimize scattered light entering the detector. The beam diameter of the Ti:sapphire laser was about 2 mm and the beam diameter of the diode laser was about 1 mm. Typically, the power of the probe beam was about 0.5 mW and that of the forward pump beam was about 1.5 mW at the entrance of the Rb vapor cell. The power of the Ti:sapphire laser was varied in the experiment between 0 to 400 mW, corresponding to the estimated Rabi frequencies of the coupling laser between 0 to 400 MHz. The natural linewidth of the Rb D_2 transition is about 6 MHz and the Doppler width is about 500 MHz. During the experiment, the Ti:sapphire laser was tuned to the center of the ^{87}Rb $5S_{1/2}(F=2)\leftrightarrow 5P_{3/2}$ transition, and the DFWM signal was recorded while the frequency of the DFWM diode laser was scanned across the Rb $5S_{1/2}(F=1)\leftrightarrow 5P_{3/2}$ transition.

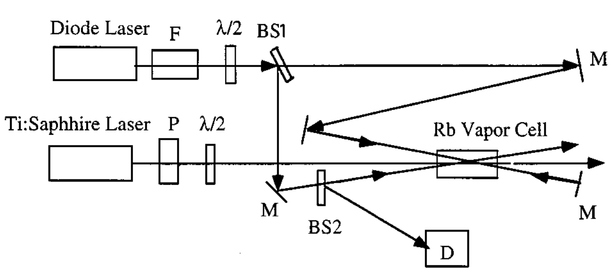

FIG. 2. Experimental arrangement. M, mirrors; D, photodetector; $\lambda/2$, half-wave plate; P, polarizer; F, Faraday isolator; BS1, 30-70 beam splitter; BS2, 50-50 beam splitter.

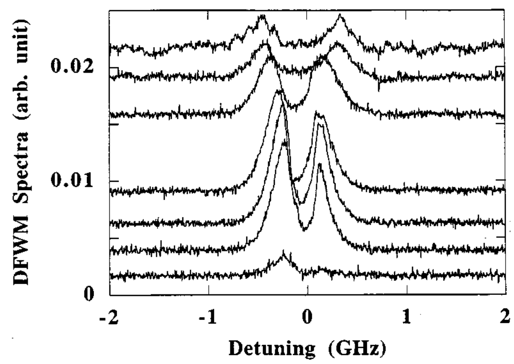

FIG. 3. The measured DFWM signal versus the DFWM laser detuning Δ for several values of the coupling laser powers. The DFWM probe field and the pump fields (both forward and backward) were linearly polarized in the same direction. The coupling laser is linearly polarized. The curves from the bottom to the top correspond to the coupling laser power 0, 2, 10, 30, 100, 200, and 400 mW, respectively. For clarity, different curves have been displaced vertically.

Figure 3 shows the measured DFWM spectra versus the frequency detuning of the DFWM diode laser from the ^{87}Rb $5S_{1/2}(F=1)\leftrightarrow 5P_{3/2}$ transition for several values of the coupling laser powers. The probe field and the pump fields (both forward and backward) were linearly polarized in the same direction, and the coupling laser was linearly polarized. We found that the DFWM emission depends sensitively on the relative polarization states of the DFWM lasers, but is essentially independent of the polarization state of the coupling laser. The recorded DFWM spectra from the bottom curve to the top curve correspond to the coupling laser power of 0, 2, 10, 30, 100, 200, and 400 mW, respectively. Without the coupling laser, the DFWM emission is very weak and shows a double-peaked spectrum. The observed double-peaked DFWM spectrum is consistent with the previously published work, which has been interpreted in terms of the ac Stark shift of the saturating DFWM fields [2–5,22]. Because the probe beam intensity was well above the Rb saturation intensity and the DFWM signal beam was completely overlapped with the probe beam in its backward direction, the linear absorption of the DFWM signal beam due to the nonperfect overlap of the pump and probe beams in the vapor cell should not be appreciable. Therefore the peak splitting observed in the DFWM spectrum should not be due largely to the linear absorption. This was verified by increasing the pump beam diameter to about 2 mm and observing the similar double-peaked DFWM spectrum. The weak DFWM emission is apparently caused by two factors: first, the optical pumping in the noncycling transitions Rb $5S_{1/2}(F=1)\leftrightarrow 5P_{3/2}(F=1$ and $2)$ that transfers the Rb atoms to $5S_{1/2}(F=2)$ hyperfine level of the ground state and terminates the DFWM laser-atom interaction; second, the optical pumping for the cycling transition Rb $5S_{1/2}(F=1)\leftrightarrow 5P_{3/2}(F=0)$ in which the DFWM laser fields induce only the Rb $5S_{1/2}(F=1,\ m_F=0)\leftrightarrow 5P_{3/2}(F=0,\ m_F=0)$ transition and transfer the atomic population to the Rb $5S_{1/2}(F=1,\ m_F=\pm 1)$ magnetic sublevels. These two factors reduce the Rb atomic population interacting with the DFWM lasers, thus diminishing the DFWM emission. When

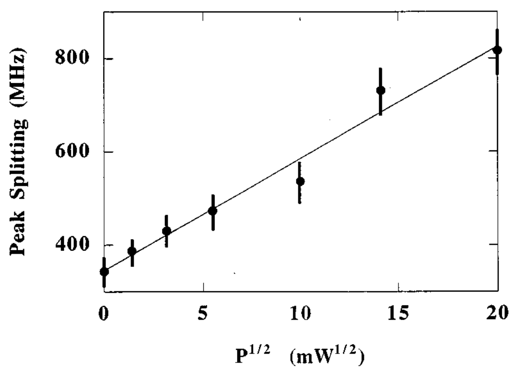

FIG. 4. The peak splitting of the DFWM emission spectra versus $\sqrt{P}$, where P is the coupling laser power. The linear dependence shows that the splitting is proportional to the Rabi frequency of the coupling laser.

the coupling laser is turned on, the optical pumping is reversed and the DFWM emission is substantially increased as shown by the curve with the coupling laser power 2 mW. As the power of the coupling laser increases, the DFWM signal increases and the separation of the two emission peaks becomes wider. The intensity of the DFWM signal is maximized at a moderate intensity of the coupling laser (~30 mW). Further increases of the coupling laser intensity result in decreases of the DFWM signal as shown by the top three curves with the coupling laser power of 100, 200, and 400 mW, respectively. Overall, the peak phase conjugate reflectivity measured from the data shown in Fig. 3 ranges from 0.2% (the zero coupling laser power) to 2% (30 mW coupling laser power). The increased peak splitting in the DFWM emission spectra at higher coupling laser intensities can be attributed to the ac Stark shift of the coupling laser. In Fig. 4, we plot the measured peak separation of the DFWM emission versus the squared root of the coupling laser power, proportional to the coupling laser Rabi frequency $\Omega=\mu E/\hbar\propto\sqrt{P}$. Here μ is the atomic dipole moment between the $5S_{1/2}(F=2)$ and $5P_{3/2}$ states, and E is the coupling field amplitude. The linear dependence of the peak splitting versus Ω is apparent. The double-peaked, DFWM emission spectrum at zero coupling laser intensity is due to the ac Stark shift induced by the moderately strong DFWM pump field [2–5]. When the coupling laser is on, an additional ac Stark splitting between the $5S_{1/2}(F=2)$ and $5P_{3/2}$ states is introduced, which results in further separation of the DFWM emission peaks.

III. DISCUSSIONS

In the effective Λ-type atomic system coupled by two laser fields studied here, two physical mechanisms influence the DFWM process. First, the reversed optical hyperfine pumping is present, which transfers the atoms back to the hyperfine level of the ground state coupled by the DFWM lasers, and thus increases the DFWM signal. The optical pumping rate is proportional to [23]

$$\gamma_{\text{pump}}\propto\frac{\Omega^2\tau_3^2}{1+4(\omega_{31}-\omega_c)^2\tau_3^2+2\Omega^2\tau_3^2}. \tag{1}$$

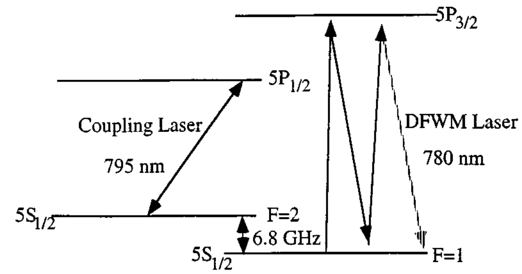

FIG. 5. The reverse optical pumping and the DFWM pump and probe scheme. The coupling laser acts as an optical pumping laser driving the Rb D_1 transition at 795 nm while the DFWM lasers are coupled to the Rb $D2$ transitions. Due to the large energy separation between the $5P_{1/2}$ and $5P_{3/2}$ states, the two-photon Raman scattering is far off resonance and its contribution is negligible.

Here τ_3 is the population lifetime of the upper $5P_{3/2}$ states, ω_{31} is the frequency separation between the $5S_{1/2}(F=2)$ and $5P_{3/2}$ states, and ω_c is the coupling laser frequency. Second, there is a resonance-enhanced two-photon Raman scattering process, $5S_{1/2}(F=1)\leftrightarrow5P_{3/2}\leftrightarrow5S_{1/2}(F=1)$, which induces the atomic population trapping between the two hyperfine ground states and directly competes with the DFWM process. The two-photon Raman scattering process involves the annihilation (creation) of one photon from the coupling laser and creation (annihilation) of one photon from one of the DFWM lasers. The Raman pump rate is proportional to [24]

$$\gamma_{\text{Raman}}\propto\frac{\Omega^2\Omega_d^2}{(\omega_{31}-\omega_c)^2\{(\omega_{32}-\omega_d+\omega_{31}-\omega_c)^2+\gamma^2\}}. \tag{2}$$

Here Ω_d and ω_d are the Rabi frequency and the frequency of the DFWM fields, respectively. ω_{32} is the transition frequency between the $5S_{1/2}(F=1)$ and $5P_{3/2}$ states and γ is the two-photon Raman linewidth. Note that for the coupling laser and the forward pump laser (or probe laser), the two-photon Raman scattering is essentially Doppler-free, and therefore, can be very effective. The Raman process competes with the DFWM process. As shown by Eqs. (1) and (2), at lower intensities of the coupling laser, the reverse optical pumping is dominant, and the DFWM signal increases due to the increase of the number of the atoms interacting with the DFWM lasers. At higher intensities of the coupling laser, the reversed optical pumping becomes saturated, and the two-photon Raman scattering becomes more important. The competition between the two-photon Raman process and the DFWM process leads to the reduction of the DFWM signal at higher intensities of the coupling laser.

To verify the competition between the two-photon Raman scattering and the optical pumping-enhanced DFWM emission, we carried out the experimental measurement of the DFWM emission in a laser coupling configuration shown in Fig. 5 in which the coupling laser is tuned to the ^{87}Rb D_1 transition while the DFWM laser is kept tuned to the ^{87}Rb D_2 transition. This configuration effectively removes the two-photon Raman scattering from the DFWM process by creating a large frequency detuning for the Raman transitions. The reverse optical pumping is achieved first by the coupling laser excitation of the Rb atoms to the $5P_{1/2}$ states and then followed by the spontaneous decay of the Rb atoms from the

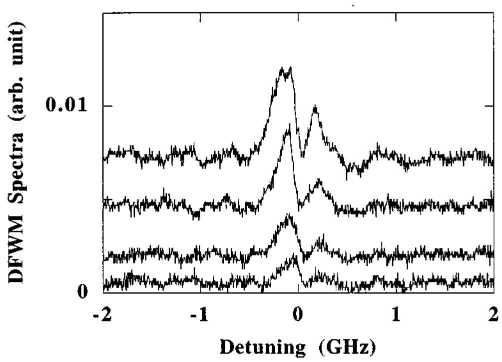

FIG. 6. The measured DFWM emission spectra for several coupling laser powers versus the DFWM laser detuning D. The curves from the bottom to the top correspond to the coupling laser power 0, 100, 200, and 400 mW. The coupling laser recycles the Rb atoms back to the Rb $5S_{1/2}(F=1)$ state and results in the enhanced DFWM emission. This reverse optical pumping increases with the increasing coupling laser power and saturates at about 200 mW. For clarity, different curves have been displaced vertically.

$5P_{1/2}$ state to the $5S_{1/2}(F=1)$ state. Once the Rb atoms are in the $5S_{1/2}(F=1)$ state, their interactions with the DFWM pump and probe fields lead to the emission of the phase-conjugated DFWM field. Since the coupling laser and the DFWM lasers were connected to the Rb D_1 and D_2 transitions, respectively, there is a large frequency detuning for the two-photon Raman scattering process. This effectively eliminates the contribution from the two-photon Raman scattering. The measured DFWM emission spectra for several values of the coupling laser powers are plotted in Fig. 6. As expected, the DFWM emission increases with the coupling laser power, no suppression of the DFWM emission is observed at higher coupling laser powers, and the coupling laser causes no further increase of the peak splitting in the double-peaked DFWM spectra. At the maximum coupling power of 400 mW, the DFWM emission is increased by about five times. For the DFWM emission in the Λ-type configuration shown in Fig. 3, at a moderate coupling laser power of about 30 mW, the maximum DFWM emission was about 10 times greater than that without the coupling laser. This may indicate that when the coupling laser is connected to the same $5P_{3/2}$ state as the DFWM lasers are, the atomic coherence induced by a weak coupling laser may have a positive effect on the DFWM emission and/or the reverse optical pumping is more efficient than that where the coupling laser is connected to the $5P_{1/2}$ state, which is isolated from the DFWM lasers. Due to the complicated and different hyperfine structures between the $5P_{3/2}$ and $5P_{1/2}$ states, uncoupled magnetic sublevels are present, and unbalanced population accumulation and trapping among different magnetic sublevels differ for the two optical pumping schemes. Thus, it is expected that the DFWM emissions will be different for the two situations. Since the two-photon Raman scattering is negligible for the experimental arrangement shown in Fig. 5, the measured DFWM emission in Fig. 6 is free of the competition from the two-photon Raman scattering, is a monotonically increasing function of the coupling power, and maximized when the reverse optical pumping is saturated at the high coupling laser power.

IV. CONCLUSION

We have measured the DFWM emission spectra in a collision-free, Doppler-broadened Rb atomic system coupled by an additional coherent field. Our measurement shows that the DFWM signal increases with an additional coupling laser at lower intensities and is maximized at a moderate intensity of the coupling laser. A further increase of the coupling laser intensity results in decreases of the DFWM emission. The experimental measurements demonstrate that the observed effects of the coupling laser on the DFWM emission can be qualitatively understood in terms of a reversed optical pumping process that transfers the atoms back to the interacting hyperfine ground state and a two-photon Raman scattering process that directly competes with the DFWM process. At higher intensities of the coupling laser, the two-photon Raman-scattering process is dominant, and therefore, the DFWM emission decreases. This conclusion is supported by the additional experimental measurements in a laser coupling configuration essentially free of the Raman-scattering contribution. However, a quantitative understanding of the experimental results requires a detailed theoretical analysis. Because of the complicated hyperfine structures in the Rb D_2 and D_1 transitions, the DFWM emission should depend sensitively on the relative polarizations of the DFWM laser fields, and the influence of the coupling laser on the DFWM emission may vary. Further experimental study along these lines and a detailed theoretical analysis should be worthwhile.

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