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Nearly degenerate four-wave mixing with bichromatic laser fields in a Rb atomic system

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We present an experimental study of cw nearly degenerate four-wave-mixing emission in a Doppler broadened Rb atomic system. With the fixed detuning of an intense pump laser for the Rb D_2 line, the nearly degenerate four-wave-mixing emission exhibits three peaks as a probe laser is scanned across the same Rb D_2 line. The observed three-peaked spectrum can be interpreted as the resonances in the energy structure of the dressed atomic states.

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I. INTRODUCTION

Degenerate four-wave mixing (DFWM) has been extensively studied during recent years, partly because of the possibility of obtaining phase conjugation using DFWM [1]. Many experiments utilized an alkali-metal vapor as the nonlinear medium and were carried out in strong pump and weak probe configurations [2–6]. Detailed theoretical treatments including inhomogeneous Doppler shifts and/or the effects of the atomic collisions in a vapor cell for two- and three-level atomic systems have been developed [7–12]. Interestingly, when the Rabi frequency of the pump field is greater than the atomic linewidth but smaller than the Doppler width, it was observed that DFWM in a two-level system exhibits two peaks that can be attributed to the ac Stark shift [2–9,13]. The two-peaked DFWM or nearly DFWM spectrum can be observed under various strong pump and weak probe configurations and its origin can be explained in terms of a dressed-state picture [8]. Here we report an experimental study of nearly DFWM in an essentially collision-free, Doppler-broadened Rb atomic system. Unlike previous studies, our experiments were carried out with at least two strong laser fields. With the frequency of a strong pump laser fixed and the frequency of a probe laser tuned across the ^{85}Rb $5S_{1/2}(F=2)\leftrightarrow 5P_{3/2}(F=1)$ transition, we observed a three-peaked spectrum in the nearly DFWM signal.

II. EXPERIMENTAL RESULTS

The ^{85}Rb D_2 transition lines and nearly DFWM scheme is depicted in Fig. 1. The pump laser fields (both forward and backward) couple the Rb $5S_{1/2}(F=2)\leftrightarrow 5P_{3/2}$ transition and their frequency is detuned from the atomic resonance by Δ_1. A probe laser field (forward) couples the same Rb $5S_{1/2}(F=2)\leftrightarrow 5P_{3/2}$ transition and the phase-conjugated signal at the probe frequency ω_p is detected in the backward direction relative to the forward probe. It should be noted that optical pumping by the lasers to a different hyperfine level of the ground state $[5S_{1/2}(F=3)]$ reduces the population of the interacting atoms and terminates the interaction of the atoms with the laser fields. Thus the four-wave-mixing signal disappears for the noncycling transitions [2,14], only the cycling transition $5S_{1/2}(F=2)\leftrightarrow 5P_{3/2}(F=1)$ contributes to the DFWM or nearly DFWM, and the Rb atoms can be treated as an effective two-level system.

The experimental apparatus is shown schematically in Fig. 2. A 75-mm-long rubidium vapor cell made of Pyrex glass was heated to 45 °C. A frequency stabilized cw Ti:sapphire laser was used to provide a strong pump field. The output beam from the Ti:sapphire laser was collimated and passed through a Faraday isolator to reduce the optical feedback from the backward pump beam. After traversing through the vapor cell, the forward pump beam was attenuated by a neutral density filter (attenuation 5 times) and then reflected back to form the backward pump beam and overlapped with the forward pump beam. An external-cavity diode laser was used as the forward probe and was aligned with the pump beams in the vapor cell. The angle between the forward pump and the forward probe beams was about 10 mrad. The pump and the probe lasers were linearly polarized. The generated backward nearly 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 diameters of the Ti:sapphire laser and the diode laser were about 1 mm. Typically, the total powers incident at the Rb cell were about 20 mW for the Ti:sapphire laser and 2 mW for the diode laser. The corresponding Rabi frequencies were about 150 MHz for the forward pump field and 45 MHz for the forward probe field (the natural linewidth of the Rb transition is about 6 MHz). So the experiment was carried out in a regime where the Rabi frequencies of the pump and probe lasers were greater than the natural

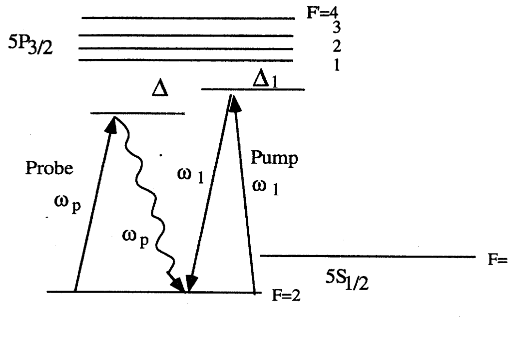

FIG. 1. Energy levels of rubidium D_2 lines and the nearly DFWM pump and probe scheme.

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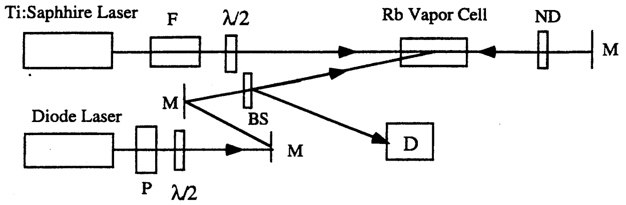

FIG. 2. Experimental arrangement. M, mirrors; D, photodetector; $\lambda/2$, half wave plate; P, polarizer; F, Faraday isolator; ND, neutral density filter; BS, 50%-50% beam splitter.

linewidth but smaller than the Doppler width (about 500 MHz at the cell temperature of 45 °C). During the experiment, the Ti:sapphire laser was tuned near the Rb $5S_{1/2}(F=2)\leftrightarrow 5P_{3/2}(F=1)$ transition and the nearly DFWM signal was recorded while the diode laser frequency was swept across the same Rb $5S_{1/2}(F=2)\leftrightarrow 5P_{3/2}(F=1)$ transition.

Figure 3(a) shows the measured nearly DFWM signal when the total power of the pump laser was 0.2 mW and the pump laser was tuned on resonance with the Rb transition (the probe power was about 0.3 mW). As expected, a single, essentially Doppler-free DFWM peak approximately located at the line center was recorded. With the total pump power of 20 mW and the probe power 2 mW, the measured nearly DFWM signal exhibits three peaks as shown in Fig. 3(b).

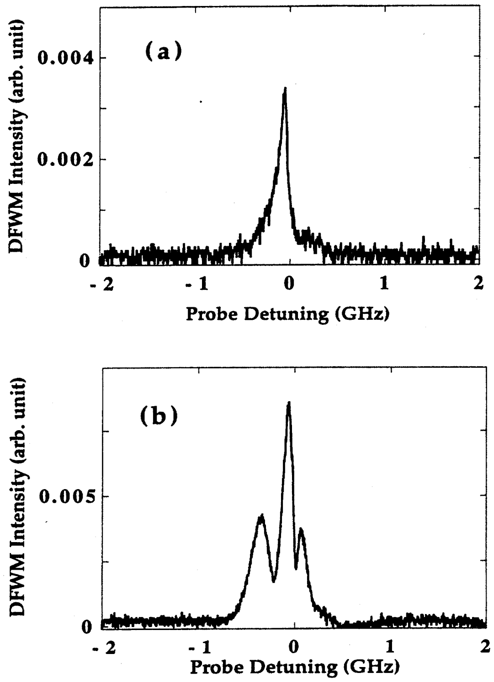

FIG. 3. (a) Nearly DFWM signal versus the probe detuning Δ with a weak pump laser (the total pump power 0.2 mW and the pump detuning $\Delta_1\approx 0$). The probe laser was about 0.1 mW. (b) Nearly DFWM signal versus the probe detuning Δ with a strong pump laser (the total pump power 20 mW and the pump detuning $\Delta_1\approx 0$). The probe power was about 0.5 mW.

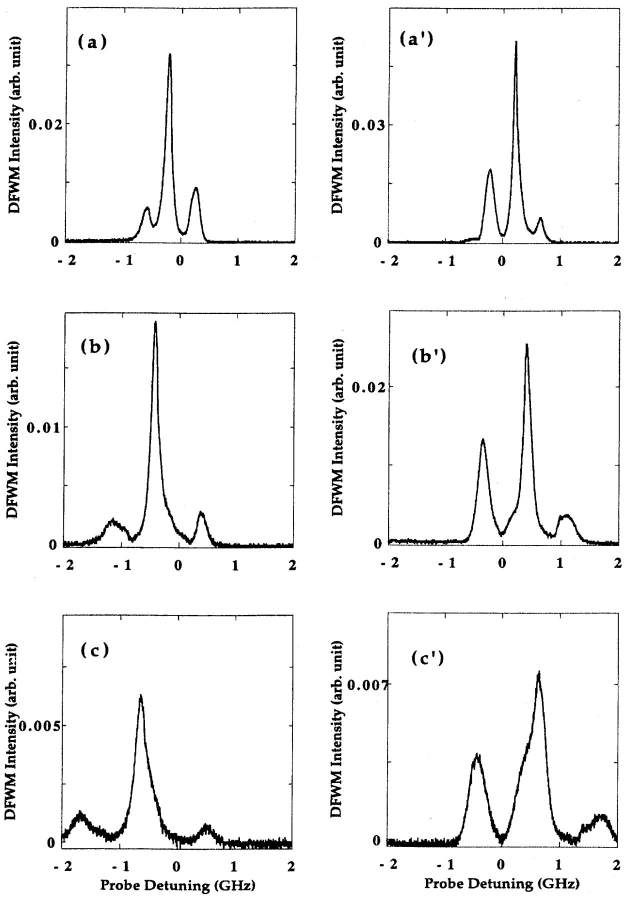

FIG. 4. Spectra of the nearly DFWM emission versus the probe detuning Δ with a strong pump laser (the total power 20 mW) tuned off the atomic transition. (a) $\Delta_1=-200$ MHz, (a′) $\Delta_1=200$ MHz, (b) $\Delta_1=-400$ MHz; (b′) $\Delta_1=400$ MHz, (c) $\Delta_1=-600$ MHz, and (c′) $\Delta_1=600$ MHz.

Figure 4 shows the measured nearly DFWM spectra versus the probe detuning Δ for a series of different pump laser detunings Δ_1. Shown in Fig. 4(a) is the nearly DFWM spectrum recorded with $\Delta_1=-200$ MHz and Fig. 4(a′) is the nearly DFWM spectrum recorded with $\Delta_1=200$ MHz. The spectrum in Fig. 4(b) [4(b′)] was recorded with $\Delta_1=-400$ MHz (400 MHz) and the nearly DFWM spectrum in Fig. 4(c) [4(c′)] was recorded with $\Delta_1=-600$ MHz (600 MHz). It is seen that as the magnitude of the pump detuning $|\Delta_1|$ increases, the peak separation increases. When the detuning Δ_1 is smaller than the Doppler width ($\sim$500 MHz under the experimental condition), the nearly DFWM signal exhibits sub-Doppler spectral peaks. When the detuning Δ_1 is greater than the Doppler width, the spectral features become broadened.

III. DISCUSSIONS

Qualitatively, the observed three-peaked nearly DFWM emission can be understood in terms of the dressed-state resonances. With the measured optical power for each coupling beam, we estimate that the Rabi frequency Ω for the

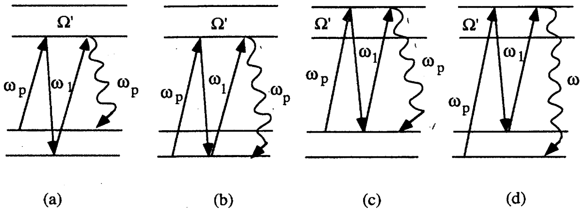

FIG. 5. Nearly DFWM resonances in terms of the dressed-state transitions: (a) the dressed-state resonance at the probe detuning $\Delta=\omega_p-\omega_0=\Delta_1-\Omega'$, (b) and (c) the dressed-state resonances at the probe detuning $\Delta=\Delta_1$, and (d) the dressed-state resonance at the probe detuning $\Delta=\omega_p-\omega_0=\Delta_1+\Omega'$.

forward pump beam is about 150 MHz, the Rabi frequency for the backward pump beam is about 30 MHz, and the probe Rabi frequency is about 45 MHz. The effective two-level Rb system is dominantly driven by the forward pump beam. The strong forward pump beam couples the Rb $5S_{1/2}(F=2)$ (state $|1\rangle$) and $5P_{3/2}(F=1)$ (state $|2\rangle$) states and creates a pair of dressed states $|+\rangle$ and $|-\rangle$ [15]. The energy eigenvalues of the two dressed states are

$$E_\pm=\frac{\Delta_1}{2}\pm\frac{1}{2}[\Delta_1^2+\Omega^2]^{1/2}, \tag{1}$$

where $\Omega=2\mu E/\hbar$ is the Rabi frequency of the forward pump field E (μ is the atomic dipole moment). The corresponding dressed states are

$$|\pm\rangle=a_\pm|1\rangle+b_\pm|2\rangle, \tag{2}$$

where the coefficients are given by

$$a_\pm=\frac{E_\pm}{(E_\pm^2+(\Omega/2)^2)^{1/2}}, \quad b_\pm=\frac{\Omega/2}{(E_\pm^2+(\Omega/2)^2)^{1/2}}.$$

The dressed states form a doublet ladder and there are three transitions with different energies among the infinite dressed-state ladder [15]. Measured from the atomic transition frequency ω_0, the three dressed-state transitions have frequencies Δ_1, $\Delta_1+\Omega'$, and $\Delta_1-\Omega'$; here $\Omega'=[\Delta_1^2+\Omega^2]^{1/2}$ is the generalized Rabi frequency. The three peaks are usually referred to as Mollow triplet [16] and have been extensively studied in the resonance fluorescence of a strongly coupled two-level atomic system [17–19]. The physical picture of nearly DFWM in the dressed states is depicted in Fig. 5. Note that in the experiment, the pump laser frequency is fixed and the probe laser frequency was swept across the atomic transition. The generated nearly DFWM emission in the dressed states is shown by the wavy lines in Fig. 5 and different combinations of the dressed-state transitions lead to three frequency components given above. For example, Fig. 5(a) represents the nearly DFWM emission at the frequency $\omega_p-\omega_0=\Delta_1-\Omega'$, Figs. 5(b) and 5(c) show the nearly DFWM emission at $\omega_p-\omega_0=\Delta_1$, i.e., $\omega_p=\omega_1$, and Fig. 5(d) shows the nearly DFWM emission at $\omega_p-\omega_0=\Delta_1+\Omega'$. Plotted in Fig. 6 are the measured positions of the nearly DFWM peaks versus the pump laser detuning Δ_1. The experimental data are plotted by the dashed lines [dots, the lower (red) nearly DFWM peak; diamonds, the central nearly DFWM peak; and triangles, the upper (blue) nearly DFWM peak] and the solid lines are the calculated positions of the three dressed-state transitions at Δ_1, $\Delta_1+\Omega'$, and $\Delta_1-\Omega'$. Qualitatively, the measurements follow the values of the dressed-state transitions. Only the positions of the central peak agree with the dressed-state transitions very well. It is not surprising that no quantitative agreement can be achieved, considering the facts that no Doppler average has been taken into account and the perturbation of the backward field and the probe field on the dressed-state levels is neglected.

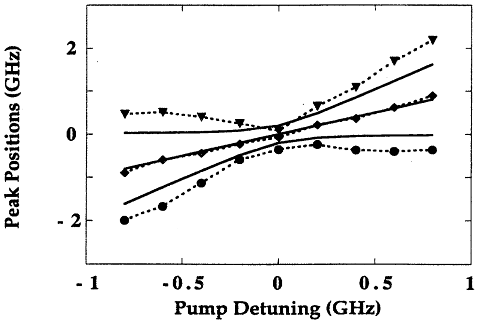

FIG. 6. Peak positions of the three nearly DFWM resonances versus the pump laser detuning Δ. Dashed lines are the measured nearly DFWM resonances, the dots represent the blue sideband, the diamonds represent the central peak, and the triangles represent the red sideband. The solid lines are the positions of the three dressed-state resonances depicted in Fig. 5.

We studied the dependence of the nearly DFWM signal on the probe and pump powers. Shown in Fig. 7 are the measured nearly DFWM spectra versus the probe detuning for several values of the probe power. The spectral peak positions essentially are independent of the probe power. We also observed the three-peaked spectra with a strong forward pump field, a saturating probe field, and a weak backward pump field. When both the probe field and the backward pump field were weak, we did not observe the three-peaked spectrum. Shown in Fig. 8 are the measured nearly DFWM

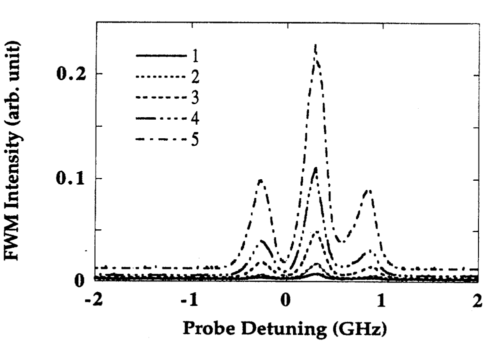

FIG. 7. Measured nearly DFWM spectra versus the probe laser detuning for several values of the probe laser powers. The forward pump laser power was about 15 mW and the reflected backward pump beam was about 3 mW ($\Delta_1\approx200$ MHz). The probe powers were 2, 0.6, 0.2, 0.06, and 0.02 mW for curves 5, 4, 3, 2, and 1, respectively.

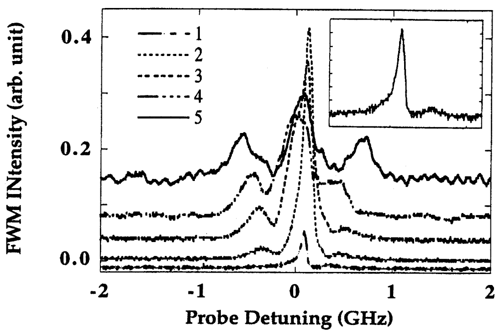

FIG. 8. Measured nearly DFWM spectra versus the probe laser detuning for several values of the pump laser powers. The pump detuning $\Delta_1 \approx 0$. The probe laser power was about 2 mW. The pump powers were 0.2, 1, 3, 15, 75, and 300 mW for curves 1, 2, 3, 4, and 5. The inset shows curve 1 (0.2 mW) in an expanded scale.

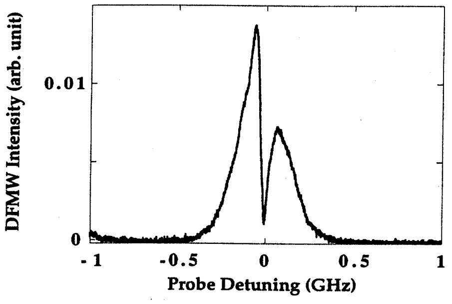

FIG. 9. Observed DFWM spectrum versus the frequency detuning with a monochromatic laser providing the forward pump, the backward pump, and the probe fields. The two-peaked DFWM emission is consistent with the previous studies.

spectra versus the probe detuning for several values of the forward pump intensity (the attenuated backward pump intensity is proportional to the forward pump intensity as shown in Fig. 2). When the pump power was reduced to about 0.2 mW, a two-peaked spectrum was observed, as shown by the inset in Fig. 8. As the pump power increases, the spectrum develops three peaks. The peak separation and the peak linewidth increase with the increasing pump intensity, but the intensity of the nearly DFWM decreases at higher pump intensities. Under our experimental conditions, the observation of the three-peaked spectrum requires at least two strong saturating fields. Furthermore, we have found that the observation of the three-peaked nearly DFWM spectrum requires two-color pump and probe fields. With a monochromatic laser providing the forward pump, the backward pump, and the probe beams, we observed only a double-peaked DFWM spectrum as shown in Fig. 9. This is consistent with the previous studies [2–9]. Grynberg and co-workers studied the DFWM with bichromatic pump and probe fields under various experimental configurations in a neon discharge before [8] and observed double-peaked DFWM or nearly DFWM spectra. Their theoretical model is based on a two-level system having identical lifetimes for both the upper state and lower state and interacting with a strong pump field, a weak pump field, and a weak probe field. Their results indicate that the nearly DFWM may have additional frequency components, but the amplitude is too small to be observed under the experiments conditions with only a single strong field.

IV. CONCLUSION

We have measured the nearly DFWM emission with two frequency fields in a collision-free, Doppler-broadened Rb atomic system. A three-peaked nearly DFWM spectrum was observed that can be attributed to the effect of the formation of the dressed atomic states by the strong forward pump laser. The experiment was carried out in a regime where the Rabi frequencies of all three beams, the forward pump, the backward pump, and the forward probe, are greater than the atomic natural linewidth, but smaller than the Doppler width. When the pump detuning Δ_1 is smaller than the Doppler width, the sub-Doppler nearly DFWM emission features were observed. When Δ_1 is greater than the Doppler width, the nearly DFWM emission becomes broadened. The observed three-peaked nearly DFWM emission can be qualitatively understood in terms of the dress-state resonances. However, a quantitative understanding of the experimental results requires a detailed theoretical analysis.

ACKNOWLEDGMENT

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