PHYSICAL REVIEW A, VOLUME 61, 033805

Coherent population trapping and four-wave mixing via dark states in a Doppler-broadened open Rb system

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(Received 25 August 1999; published 11 February 2000)

We report an experimental study of coherent population trapping and four-wave mixing via multilevel dark states prepared in the ground hyperfine states of ^{87}Rb atoms. The dark states are created by Λ-type coupling chains driven by a linearly polarized pump laser tuned to the D_1 $F=2\leftrightarrow F'=1$ transition. The Doppler shift in the coupling chains is compensated by the left- and right-circularly polarized components of the pump laser. The multilevel dark states are explored by a probe laser and their existence is manifested by a strong dependence of the probe excitation and four-wave-mixing emission on the probe polarization. Our experiment shows that coherent population trapping in multilevel dark states of a degenerate atomic system should be useful in the coherence control of optical nonlinearity.

PACS number(s): 42.50.Gy, 42.50.Hz, 32.80.Bx, 42.65.−k

Coherent population trapping (CPT) was originally observed in Na, which was modeled as a closed three-level Λ system in which a dark state composed of a coherent superposition of the two ground states is generated by two radiation fields [1,2]. CPT plays an important role in a variety of physical phenomena, such as cooling of atoms below the photon recoil limit [3], trapping of atoms in optical lattices [4–5], electromagnetically induced transparency [6], lasing without population inversion [7], control of nonlinear atomic susceptibilities [8], and enhancement of nonlinear optical processes [9–12]. Studies of these phenomena have been based on closed three or four-level atomic systems in which total atomic population is conserved.

In practical atomic systems, atomic transition often involves magnetic sublevel structures that can lead to interesting consequences. Coherent radiation-atom interaction in multilevel degenerate systems has received increasing attention in recent years [13]. It has been found that CPT is a more general phenomenon that exists in a two-state atomic system with multiple degenerate ground and excited sublevels [14–16]. When the number of degenerate ground-state sublevels is greater than or equal to the number of degenerate excited-state sublevels, dark states that consist of a coherent mixture of the ground sublevels are generated by an elliptically polarized pump field and CPT is therefore realized in a degenerate multilevel system [16,17]. In open multilevel atomic systems, optical pumping transfers atomic population to a noninteracting ground state, which is detrimental to the multilevel CPT. Theoretical studies in homogeneous atomic systems show that the multilevel CPT competes effectively with the optical pumping and can be preserved in an open atomic system [16]. Specifically, Ling *et al.* showed theoretically that CPT exists in the open multilevel Rb D_1 $F=2\leftrightarrow F=1$ transitions [18]. Recently, Milner and Prior demonstrated experimentally the multilevel CPT on the D_1 $F=2\leftrightarrow F'=2$ transitions of sodium atoms in an atomic beam apparatus [19].

In a Doppler-broadened system, atoms with different thermal velocities experience different frequency detunings from the pump laser. The two circular components of a linearly polarized pump laser that couple the $\Delta m=1$ and $\Delta m=-1$ sublevel transitions separately have exactly the same Doppler shift, which leads to a complete cancellation of the Doppler shifts for Λ-type transitions induced by the two circular components of the same pump laser (see Fig. 1). The Λ-type coupling chain of the degenerate hyperfine ground sublevels is then Doppler free and resonant for all atoms. This establishes the dark states and results in coherent population trapping in a Doppler broadened, multilevel system [20]. CPT should play an important role in nonlinear optical phenomena in multilevel atomic systems. Below, we report an experimental study of CPT in a Doppler-broadened open Rb atomic system and show that CPT competes effectively with optical pumping and can be used to induce four-wave-mixing (FWM) emission in the open system.

Consider the ^{87}Rb D_1 $F=2\leftrightarrow F'=1$ transition with relevant energy levels, links of the transition among magnetic sublevels, and the coupling constants depicted in Fig. 1. Atoms in the excited state spontaneously decay to the hyperfine ground state $F=1$, which makes the degenerate multilevel system open. Ling *et al.* showed theoretically that CPT can be preserved in this open system and electromagnetically

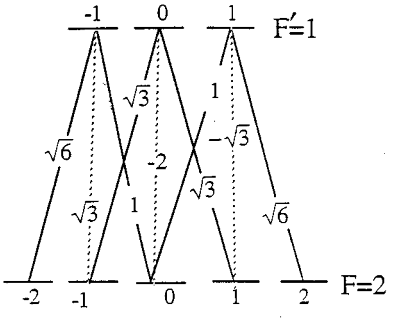

FIG. 1. Energy levels and relative dipole moments for the ^{87}Rb D_1 $F=2\leftrightarrow F'=1$ transition. A linearly polarized pump laser is resolved into the left- and right-circular polarization components that induce the Λ-type coupling chains (shown by solid lines) and create dark states.

induced transparency may be observed by a probe laser coupled to the transition $F=1\leftrightarrow F=2$ [18]. General treatments of CPT in a degenerate multilevel system coupled by an elliptically polarized field can be found in Refs. [17] and [21]. A physical picture of the CPT can be derived if one resolves an elliptically polarized field into the left and right circular components that induce $\Delta m=+1$ and $\Delta m=-1$ transitions respectively among the degenerate sublevels. The character of the dark states is determined by the ellipticity, eccentricity, and the phase difference between the two circular components of the light field. CPT occurs in the transition $F\leftrightarrow F'$ only when $F'\leqslant F$ and is manifested by Λ-type transition chains connecting the degenerate hyperfine ground sublevels [16–17]. For the $F=2\leftrightarrow F'=1$ transition coupled by a pump field that is linearly polarized in the x direction (assuming the wave vector k is in the z direction) and is written as a superposition of the left ($\hat{\varepsilon}_+$) and the right ($\hat{\varepsilon}_-$) circularly polarized components: $\vec{E}=\hat{\varepsilon}_x E\exp(-i\omega t)+\text{c.c.}=(1/\sqrt{2})(\hat{\varepsilon}_+ +\hat{\varepsilon}_-)E\exp(-i\omega t)+\text{c.c.}$, two Λ-type coupling chains exist and are shown by the solid lines in Fig. 1. The two corresponding dark states are given by [18] $|\phi_1\rangle=\sqrt{1/8}\{|-2\rangle-\sqrt{6}|0\rangle+|2\rangle\}$ and $|\phi_2\rangle=\sqrt{1/2}\{|-1\rangle-|1\rangle\}$, respectively. Note that the dark states are a coherent mixture of three or two sublevels among the five magnetic sublevels in the ground $F=2$ hyperfine state. Since characteristics of the CPT depends on the polarization of the pump field, it can be explored by a probe laser tuned to the same atomic transition as the pump laser and used to induce a nearly degenerate FWM process via the darks states in an open multilevel system. By controlling the polarization state of the probe laser, the probe absorption and the induced FWM emission can be manipulated.

The transition probability induced by a probe field E_p between the dark states ϕ_i ($i=1$ and 2) and the excited hyperfine state F' (including all magnetic m' sublevels) is given by

$$P(\phi,F')=\sum_{i,m'}|\langle\phi_i|\vec{D}\cdot\vec{E}_p|F',m'\rangle|^2. \tag{1}$$

When the probe laser is polarized parallel to the linearly polarized pump field, i.e., $\vec{E}_p=\hat{\varepsilon}_x E_p\exp(-i\omega t)+\text{c.c.}=(1/\sqrt{2})(\hat{\varepsilon}_+ +\hat{\varepsilon}_-)E_p\exp(-i\omega t)+\text{c.c.}$, we derive $P(\phi,F')=0$. The dark states generated by the pump laser are decoupled from the probe laser due to destructive interference among the multiple excitation paths of the transition $|\phi_i\rangle\rightarrow|F',m'\rangle$. When the probe laser is polarized perpendicular to the pump laser, i.e., $\vec{E}_p=\hat{\varepsilon}_y E_p\exp(-i\omega t)+\text{c.c.}=(-i/\sqrt{2})(\hat{\varepsilon}_+ -\hat{\varepsilon}_-)\{E_p\exp(-i\omega t)+\text{c.c.}\}$, constructive interference among the excitation paths occurs because the two circularly polarized components in $\hat{\varepsilon}_+$ and $\hat{\varepsilon}_-$ have a π phase difference between the x polarized probe field and the y polarized probe field. The probability of the transition $F=2\leftrightarrow F'=1$ is enhanced by the constructive interference. The interference contrast for the two orthogonal polarization directions is $[P_\perp(\phi,F')-P_\parallel(\phi,F')]/[P_\perp(\phi,F')+P_\parallel(\phi,F')]=100\%$. Here $P_\parallel(\phi,F')$ $[P_\perp(\phi,F')]$ is the transition probability for the x [y] polarized probe field.

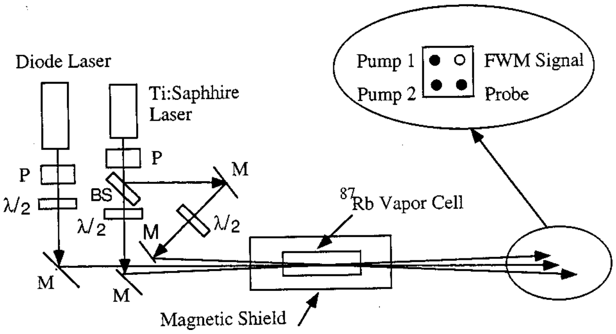

FIG. 2. Experimental arrangement for observation of the multilevel CPT and dark-state FWM in ^{87}Rb atoms. P's: polarizers; $\lambda/2$'s: half-wave plates; *M*'s: mirrors; *D*: photodiode detector. BS: 50-50 beam splitter. The inset panel shows the phase-matching beam configuration for the nearly degenerate FWM.

The experimental arrangement is shown schematically in Fig. 2. The experiment was performed in a room temperature, ^{87}Rb isotope vapor cell (the isotope abundance $\geqslant 95\%$) without any buffer gas. The Rb atomic density was estimated to be $\sim 2\times 10^{10}\,\text{cm}^{-3}$. The vapor cell was 7.5 cm long and was magnetic shielded by a 15-cm long, μ-metal tube. The measured residual magnetic field was $\leqslant 0.03$ G. The hyperfine splitting of the excited $5P_{1/2}$, $F'=2$ and $F'=1$ states is 816 MHz, which is greater than the Doppler width of ~ 540 MHz. The pump fields were provided by a cw Ti: Sapphire laser (Coherent 899-21) with a beam diameter ~ 2 mm and power ~ 50 mW. The Ti: Sapphire laser was split into two pump beams of equal power (~ 25 mW for each beam and the estimated Rabi frequency ~ 50 MHz) and same linear polarization. An extended-cavity diode laser with a beam diameter ~ 1 mm was used as the probe laser. The linewidth of the Ti:Sapphire laser was ~ 5 MHz and the linewidth of the diode laser was ~ 1 MHz. The two pump beams and the probe beam were overlapped in the Rb vapor cell with a mutual angle $\sim 0.5°$ and were aligned into a forward FWM configuration in which the phase matching condition expressed in terms of the wave vector $\vec{k}_i$ ($i=1$, 2, p, and FWM stand for the two pump fields, the probe field, and the generated FWM field, respectively), $\vec{k}_1+\vec{k}_p=\vec{k}_2+\vec{k}_{\text{FWM}}$, is satisfied as shown in Fig. 2. For the probe absorption measurement, the diode laser power was ~ 0.2 μW and the transmitted probe beam was detected by a photodiode. For the FWM measurement, the diode laser power was ~ 0.4 mW, and the FWM emission was picked up by another photodiode placed 2 m away from the Rb cell and spatially aligned to the FWM emission. The photodiode signals were sent to a digital oscilloscope and the digitized data were stored in a PC. During the experiment, the Ti: Sapphire laser was tuned to the center of the D_1 $F=2\leftrightarrow F'=1$ transition at ~ 795 nm while the diode laser frequency was scanned across the D_1 $F=2\leftrightarrow F'$ transitions.

Figure 3(a) shows the probe absorption spectrum without the pump laser as a reference. When the linearly polarized pump laser was turned on, the probe absorption spectra are shown in Fig. 3(b). When the probe laser polarization was parallel to that of the pump laser (dashed line), the absorp-

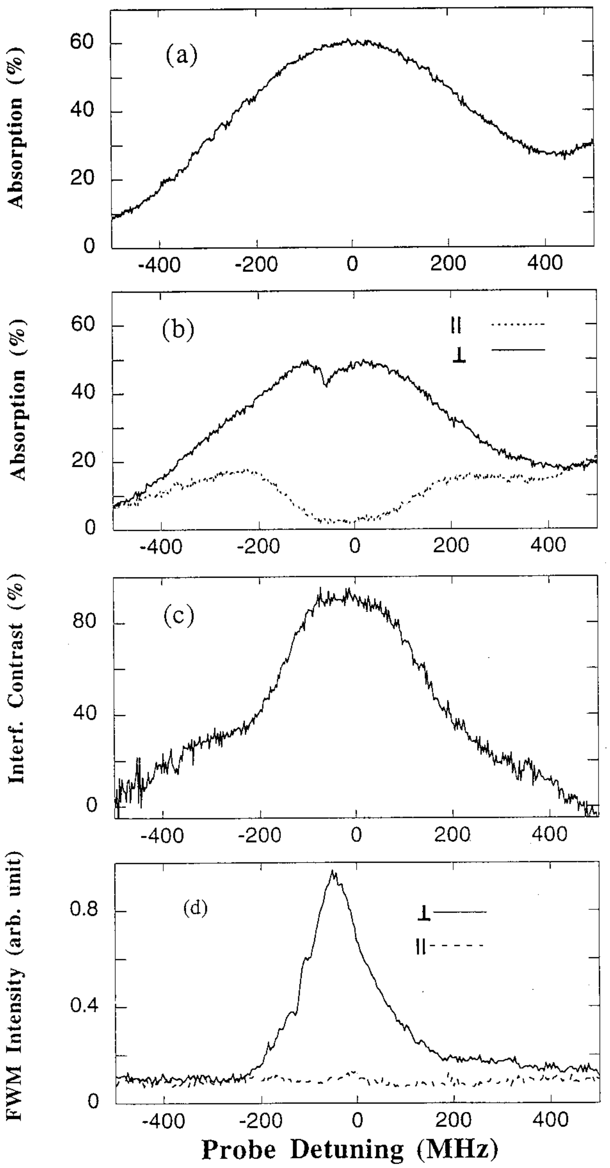

FIG. 3. Measured probe absorption and FWM spectra versus the probe laser detuning from the D_1 $F=2\leftrightarrow F'=1$ transition. (a) The probe absorption spectrum without the pump laser near the ^{87}Rb D_1 $F=2\leftrightarrow F'=1$ transition. (b), (c), and (d) show the probe absorption spectra, the interference contrast, and the FWM spectra with a linearly polarized pump laser tuned to the center of the $F=2\leftrightarrow F'=1$ transition. (b) Dashed (solid) line shows the probe linear absorption spectrum when the probe field was linearly polarized parallel (perpendicular) to the pump fields. (c) The interference contrast for the two orthogonal polarization directions of the probe field. (d) The FWM spectra when the probe field was linearly polarized perpendicular (solid line) or parallel (dashed line) to the pump fields.

tion at the $F=2\leftrightarrow F'=1$ transition is suppressed by the destructive interference. The broad minimum at the $F=2\leftrightarrow F'=1$ transition reflects the thermal distribution of the trapped Rb atoms and shows that CPT is effective for most of the Rb atoms in the thermal velocity distribution. This is expected from the Doppler-free Λ-type coupling of the two circular components of the linearly polarized laser. The weak probe laser simply maps out the trapped Rb atoms in the dark states and plays no role in the creation of the dark states. This is different from the narrow-peaked CPT observed in the nondegenerate three-level Λ-type system coupled by two lasers with different frequencies. As the coupling laser intensity decreases, the absorption near the line center increases and the broad minimum becomes narrower. At higher coupling laser intensities, the off-resonant excitation of the Rb atoms to the $F'=2$ state becomes appreciable, which leads to the transfer of some Rb atoms to the ground $F=1$ hyperfine state and results in fewer Rb atoms trapped in the ground $F=2$ dark states. When the probe laser was polarized perpendicular to the pump laser (solid line), the absorption is enhanced at the $F=2\leftrightarrow F'=1$ transition by the constructive interference. The interference contrast ($\perp$ probe absorption $-\parallel$ probe absorption)/($\perp$ probe absorption + $\parallel$ probe absorption), is plotted in Fig. 3(c). Near the line center of the $F=2\leftrightarrow F'=1$ transition, it is ~92%, in good agreement with the analysis above. Note that the maximum absorption at the constructive interference is ~50%, indicating that a large number of atoms are trapped in the dark states in spite of the competing optical pumping that transfers the atoms to the noninteracting, ground $F=1$ hyperfine state. This is consistent with the theoretical study of Ref. [18] based on the homogeneous Rb system. Quantitative comparison with the theory requires a detailed analysis including the Doppler effect and the off-resonant coupling with the $F=2\leftrightarrow F'=2$ transition, which is quite complicated and will be left for future consideration. When the pump laser was tuned to the center of the $F=2\leftrightarrow F'=2$ transition, we also observed similar polarization dependence of the probe absorption manifested by the multilevel CPT [20].

The FWM spectrum is shown in Fig. 3(d). For the probe laser polarized perpendicular to the pump laser, the intense, nearly degenerate FWM emission was observed near the $F=2\leftrightarrow F'=1$ transition (solid line). While for the probe laser polarized parallel to the pump laser, the FWM was suppressed (dashed line). The FWM is produced by a coherence grating created by the probe laser and one of the pump laser beams [22], which is optimized when CPT is generated. The observed polarization dependence of the nearly degenerate FWM demonstrates that CPT in multilevel atomic systems can be used to control nonlinear optical phenomena. We note that Grove *et al.* observed the efficient backward FWM on the Rb D_1 $F=2\leftrightarrow F=1$ transition in a B field of ~1.5 G that lifts the degeneracy of the magnetic sublevels [11]. Our experiment was done in a near-zero B environment in which the degenerate multilevel CPT is clearly demonstrated in Fig. 3(b) and is the determining factor as shown by the dependence of the FWM on the probe polarization.

For comparison, we also measured the probe absorption spectrum when the pump laser was circularly polarized and tuned to the same $F=2\leftrightarrow F'=1$ transition. As expected, no polarization dependence of the probe absorption was observed. Also no FWM emission was observed since a circularly polarized pump laser optically pumps the atoms to the $m_F=-1$ and -2 (or $+1$ and $+2$) magnetic sublevels and does not create a coherence grating.

As an independent check, we measured polarization dependence of the probe absorption across the ^{87}Rb $F=1\leftrightarrow F'$ transitions when the linearly polarized pump laser was tuned to the center of the $F=1\leftrightarrow F'=2$ transition. No CPT occurs for the transition of $F'>F$, and the optical pumping depletes atomic population from the ground $F=1$ hyperfine state. The measurements showed that the probe absorption is near zero and does not depend on the probe polarization. Since the atoms are depleted by the optical pumping from the ground $F=1$ state to the noninteracting ground $F=2$ state, no FWM emission can be observed.

In conclusion, we have reported an observation of CPT in an inhomogeneously broadened, multilevel open atomic system and have used the CPT to generate the nearly degenerate FWM emission. Although the competing optical pumping transfers atoms to the noninteracting ground hyperfine state, a large number of the atoms survives the optical pumping and are trapped in the dark states. CPT is demonstrated by the polarization dependence of the weak probe absorption and can be used to realize polarization control of the degenerate FWM emission. CPT may be important in diverse physical phenomena in which a multi-level atomic system (either open or close) interacts with a moderate pump laser. In particular, CPT in multilevel atomic systems should be useful in a number of applications such as polarization control of multiple laser excitation and ionization of atoms and molecules, enhancement of nonlinear optical susceptibility, and manipulation of neutral atoms in atom optics [23].

This work was supported by the National Science Foundation (Grant No. PHY-9729133) and the U.S. Army Research Office (Grant No. DAAH04-95-1-0534).

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