PHYSICAL REVIEW A **69**, 061802(R) (2004)

Dynamic optical bistability in resonantly enhanced Raman generation

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(Received 9 April 2004; published 17 June 2004)

We report observations of novel dynamic behavior in resonantly enhanced stimulated Raman scattering in Rb vapor. In particular, we demonstrate a dynamic hysteresis of the Raman scattered optical field in response to changes of the drive laser field intensity and/or frequency. This effect may be described as a dynamic form of optical bistability resulting from the formation and decay of atomic coherence. We have applied this phenomenon to the realization of an all-optical switch.

DOI: 10.1103/PhysRevA.69.061802 PACS number(s): 42.50.Gy, 42.65.Dr, 42.65.Pc, 42.65.Sf

It is now well known that the optical properties of atomic media may be dramatically altered if the atoms are placed into an appropriate quantum superposition of states, enabling, e.g., electromagnetically induced transparency (EIT) and lasing without inversion [1]. Such coherently prepared media can also exhibit extremely large nonlinearity at very low light levels [2–6]. In this paper we demonstrate a novel form of nonlinearity—dynamic optical bistability—using resonantly enhanced Raman generation in warm Rb vapor [7]. The observed bistable behavior results from the formation of a long-lived coherence in the atomic ensemble due to the strong light-atom interaction provided by a double-Λ interaction scheme, which can be approximated as two strong pump fields $\Omega_{1,2}$ interacting with a three-level atom as shown in Fig. 1(a) [8]. Strong Raman gain is produced with this scheme, generating a pair of correlated Stokes and anti-Stokes fields $E_{1,2}$ [9–11]. We find dynamic optical bistability in the form of hysteresis in the response of either of the Raman fields to sufficiently fast variation in the corresponding pump field.

Optical bistability has been extensively studied for several decades (see [12,13] for reviews), and observed in many systems. Most typically, optical bistability occurs with a nonlinear medium placed in an optical cavity, such that there is more than one stable condition of output light intensity from the cavity for a given intensity of near-resonant input light, i.e., there is a hysteresis in the effective cavity transmission. Optical bistability with a cavity has been observed with various two- and three-level atomic systems serving as the nonlinear medium [14–19]. Optical bistability has also been demonstrated without a cavity using degenerate four-wave mixing in atomic vapor with two counterpropagating laser beams [20,21]. In these latter experiments, the bistability arises from a dependence of phase-matching conditions on the magnitude of the output field [22].

The unique features of the present results are (i) optical bistability arises due to the formation of long-lived atomic coherence, which in turn depends on the amplitudes and phases of all four optical fields ($\Omega_{1,2}$ and $E_{1,2}$); and (ii) bistability is observed in a co-propagating laser geometry without a cavity. The steady-state amplitudes of both the Stokes and anti-Stokes fields are self-adjusted by the light-atom interaction, so that a generalized four-photon dark state is established that is decoupled from all optical fields. The generalized dark state in the double-Λ system is analogous to the dark state polariton employed in "stored light" in the single-Λ system [23–25], with the dynamics controlled by three relevant time scales. First, there is the characteristic time of atomic response to change in the pump fields, i.e., the time for atomic coherence to be created and modified. This time is given by the inverse bandwidth of the four-photon Raman process [9,10], and may be quite short,

$$\tau_R \propto \frac{\Delta}{|\Omega_1||\Omega_2|}, \tag{1}$$

where Δ is the detuning of the off-resonant pump field Ω_1 (see Fig. 1(a)). In our experiment, $\tau_R < 1\ \mu$s for typical values of the pump fields $\Omega_{1,2}$ and Δ.

The second relevant time scale characterizes the equilibration of the four-photon dark state, and hence the response time of the amplitudes and phases of the generated Raman fields. This time is determined by the optical pumping rate of the far-detuned pump field Ω_1,

$$\tau_S \propto \frac{\Delta^2}{\gamma|\Omega_1|^2}, \tag{2}$$

where γ is the relaxation rate of the excited state. For comparable values of the pump field powers, one has

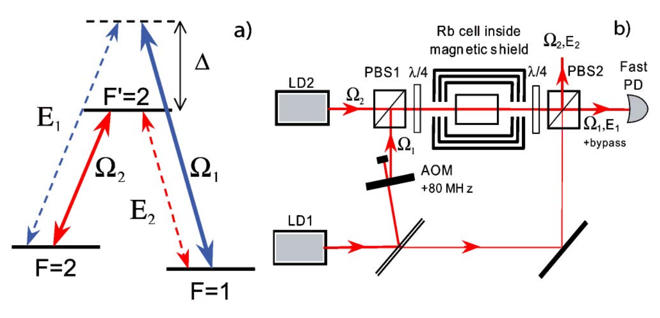

FIG. 1. (a) Three-level double-Λ scheme used in the experiment. The first diode laser LD1 (pump field Ω_1) is detuned by $\Delta \approx 4$ GHz to the blue side of the Rb transition $F=1 \rightarrow F'=2$. The second laser LD2 (pump field Ω_2) is resonant with $F=2 \rightarrow F'=2$. (b) Experimental setup.

$\tau_S \simeq (\Delta/\gamma)\tau_R \gg \tau_R$; for our system, $\tau_S \approx 30$ μs. This large difference in time scales enables dynamic optical bistability when either of the pump fields is modulated on a time scale between τ_S and τ_R, because the generated Raman fields depend on the magnitude and phase of the atomic coherence. Once the modulated pump field passes above the threshold for Raman generation, atomic coherence is created. However, the intensity of the Raman field reaches its steady-state value with some delay τ_S, effectively increasing the observed threshold pump power. Similarly, Raman generation continues at lower pump field power than the steady-state threshold when the pump field is reduced from its peak value. As the pump field is again increased, the hysteresis cycle is repeated.

The third relevant time scale is the atomic coherence lifetime, which for our system is limited by atomic diffusion in and out of the interaction region to be $T_2 \sim 2$ ms $\gg \tau_S$. Because atomic coherence provides a "reservoir" for Raman generation, dynamic optical bistability can be observed for very slow pump field modulation, down to a modulation period $\simeq T_2$.

A schematic of our experimental setup is shown in Fig. 1(b). We used two diode lasers LD1 and LD2 operating at 795 nm and resonant with the D_1 line ($5\,^2S_{1/2} \rightarrow 5\,^2P_{1/2}$) of ^{87}Rb. The first laser was detuned by Δ from the $F=1 \rightarrow F'=2$ transition, and the second laser was resonant with the $F=2 \rightarrow F'=2$ transition. To control the intensity of the off-resonant field Ω_1, a part of the beam from the first laser was depleted using an acousto-optic modulator (AOM) with a frequency shift of 80 MHz, and then combined with radiation from the second laser on a polarizing beam splitter (PBS1). The polarizations of the two fields were then transformed by a quarter-wave plate into orthogonal circular polarizations before entering the Rb cell. The maximum available optical power for the lasers LD1 and LD2 was approximately 3 mW and 4 mW, respectively, with the laser beams focused inside the cell to diameters of about 200 μm.

We employed a cylindrical glass cell filled with isotopically pure ^{87}Rb and 6 Torr of Ne buffer gas. We placed the cell inside a three-layer magnetic shield to reduce the influence of stray magnetic fields, and heated the cell to 106 °C, which corresponds to a Rb vapor density of 7×10^{12} cm^{-3}. Since counterpropagating laser beams can produce mirrorless oscillations in the Raman field [26], we took care to avoid retroreflection of either laser beam back into the atomic cell.

For the chosen pump field polarizations and resonant atomic levels, each of the generated Raman fields had circular polarization, with the same chirality as the corresponding pump field. Thus all fields had linear polarization after passing through the second quarter-wave plate placed after the Rb cell, with the polarization of fields Ω_1 and E_1 being orthogonal to Ω_2 and E_2. All these fields were then combined at the second polarizing beamsplitter (PBS2) together with an additional beam from the first laser which propagated outside of the atomic cell. The frequency of this "bypass" field was shifted down by 80 MHz with respect to the field Ω_1. We detected the beat-note signal between the bypass field and the Raman field E_1 using a fast photodetector and a spectrum analyzer in zero-span mode with a registration bandwidth of 3 MHz. Since the amplitude of the bypass field was constant, only changes in the amplitude of E_1 were detected.

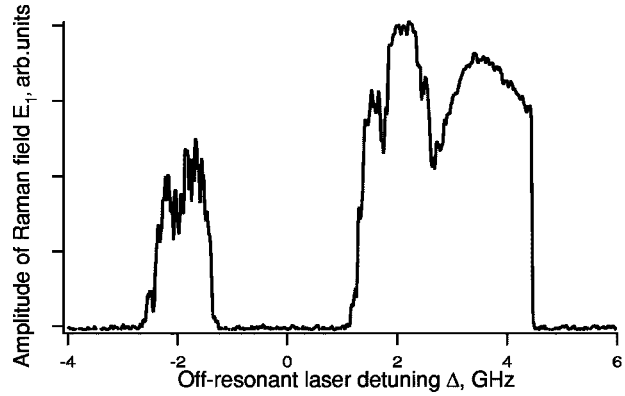

FIG. 2. Example spectrum of the generated Raman Stokes field E_1 as a function of the detuning Δ of the off-resonant pump field Ω_1.

Figure 2 shows a typical spectrum of the generated Stokes field E_1. We observed such Raman generation over a wide range of the first laser's detuning Δ. The absence of Raman generation around $\Delta = 0$ matches the resonant absorption for the pump field Ω_1. Raman generation also disappeared for large detunings once the frequency of the generated field E_1 approached the $F=1 \rightarrow F'$ transitions. Other smaller variations of the Stokes field amplitude were caused by effects such as switching between different modes of Raman generation. The frequencies of these modes differ by a few hundred kHz, and were found to depend on the intensity of the pump fields as well as details of the laser beams' spatial overlap, which likely affect the four-photon phase-matching conditions [7,27]. However, these smaller Raman variations did not change the qualitative behavior of the dynamic optical bistability.

To study the dynamics of Raman generation, we varied the intensity of the off-resonant laser field Ω_1 by modulating the voltage applied to the AOM at frequency f_{mod}, while the intensity and frequency of laser LD2 were kept constant. Figure 3(a) shows the observed dependence of the Stokes field amplitude E_1 on the corresponding pump field Ω_1, for the case of very slow variations of Ω_1 (i.e., $f_{\mathrm{mod}} T_2 \ll 1$). One can see that the amplitude of the Stokes field exhibits thresholdlike behavior, and then reaches a maximum (determined by the phase-matching conditions between all four optical fields). If the intensity of the pump field increases sufficiently, it drives the system out of the optimal conditions for Raman generation, and the amplitude of the generated field decreases. It is important to note that in the quasistatic case ($f_{\mathrm{mod}} < 10$ Hz), the Raman generation threshold is the same regardless of the "direction" of the pump field's change. Thus there is no hysteresis in the regime of low modulation frequency.

As shown in Figs. 3(b)–3(d), a hysteresis loop appears for the Stokes field amplitude as f_{mod} increases. We define the "threshold" values P_{on} and P_{off} as the pump field powers at which Raman generation starts (for increasing Ω_1) and ceases (for decreasing Ω_1). Their dependence on f_{mod} is pre-

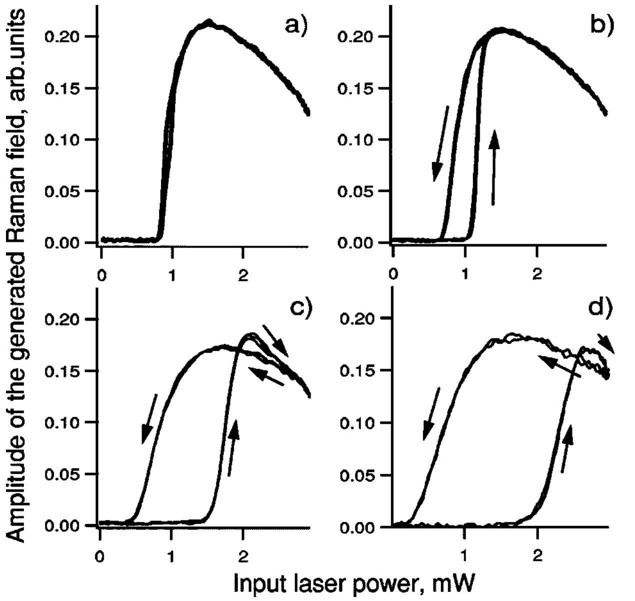

FIG. 3. Differing degrees of observed hysteresis in the generated Raman Stokes field E_1 as the corresponding Ω_1 pump field intensity is modulated at (a) 10 Hz (no hysteresis); (b) 150 Hz; (c) 1.2 kHz; (d) 4 kHz. Several (two to three) cycles are shown on each plot to demonstrate the reproducibility of the data, with arrows indicating the "direction" of change of the pump field. The off-resonant laser detuning Δ was approximately 4 GHz.

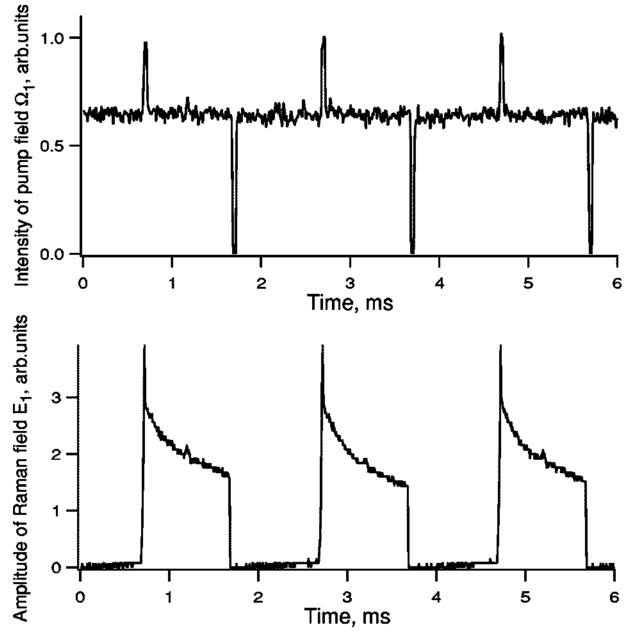

FIG. 5. Demonstration of an all-optical switch using dynamic optical bistability. The Stokes field E_1 could be switched on and off (lower graph) by pulsing the pump field Ω_1 (top graph). The duration of the Ω_1 pulses was 40 μs; the plotted intensity of these pulses is normalized to their maximum value ($\approx$2.5 mW) with a detuning $\Delta \approx 4$ GHz.

sented in Fig. 4. One can see that the difference between P_{on} and P_{off} increases with the modulation frequency, and these threshold powers are reasonably fit by $P_{\mathrm{on}}-P_{\mathrm{th}} \propto \sqrt{f_{\mathrm{mod}}}$ and $P_{\mathrm{off}}-P_{\mathrm{th}} \propto -\sqrt{f_{\mathrm{mod}}}$, where P_{th} is the threshold power for Raman generation in the cw regime. We can reproduce this scaling law by taking into account the delayed response of the generated Raman field to the pump field modulation, as characterized by the delay time τ_S [which depends on pump field power, see Eq. (2)]. This simple model yields

$$P_{\mathrm{on,off}} \simeq P_{\mathrm{th}}\left(1 \pm \tfrac{1}{2}\sqrt{f_{\mathrm{mod}}\tau_S^{\mathrm{th}}}\right), \qquad (3)$$

where τ_S^{th} is the four-photon dark-state equilibration time in the vicinity of the Raman generation threshold. This expression is in good qualitative agreement with the experimental results. However, a more complete description of the threshold behavior will require treatment of the diffusion of coherently prepared atoms in and out of the optical interaction region [28,29].

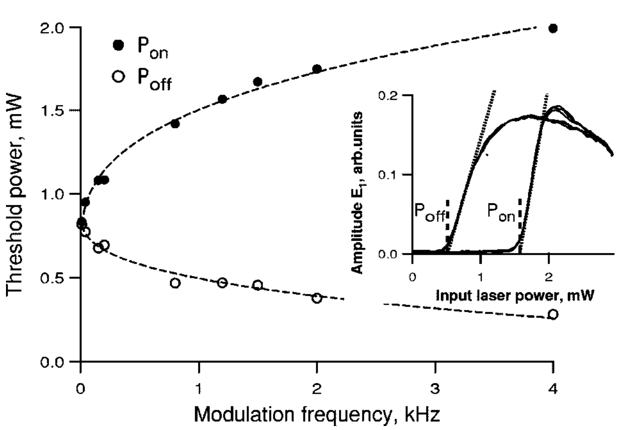

FIG. 4. Measured dependence on modulation frequency of the hysteresis threshold powers, P_{on} and P_{off}, of the off-resonant pump field. The dashed lines indicate independent fits of P_{on} and P_{off} to $\sqrt{f_{\mathrm{mod}}}$. Inset: Graphic definition of the manner in which P_{on} and P_{off} were determined from the hysteresis data.

At higher modulation frequencies ($f_{\mathrm{mod}} > 1$ kHz), the measured peaks of the Raman field E_1 output differ for increasing and decreasing pump field Ω_1, a result of changes in the four-photon phase-matching conditions (which determine optimal Raman generation) that are fast relative to τ_S.

Two additional notes about the observed dynamic optical bistability: (i) we found Raman hysteresis similar to that shown in Fig. 3 by fixing the intensity of both pump lasers, tuning Δ to be near a cutoff frequency for Raman generation (see Fig. 2), and modulating the frequency of either of the pump fields; and (ii) we observed similar bistable behavior for the anti-Stokes field E_2, although this field was generally much weaker than E_1 because of large residual absorption.

As an example application of dynamic optical bistability, we demonstrated an all-optical switch in which the Raman field E_1 was turned on and off by briefly pulsing the intensity of the Ω_1 pump field above or below a median level (see Fig. 5). During the turn-on pulse, the strength of the pump field

Ω_1 was large enough to establish significant atomic coherence, and thus to provide large Raman generation once Ω_1 returned to its median value. Similarly, the duration of the turn-off pulse was long enough for the resonant Ω_2 pump field to convert atomic coherence in the interaction region into anti-Stokes E_2 photons, in a process closely related to the release of stored light [10,11,23–25]. Thus the switching for both turn-on and turn-off can likely be made much faster with stronger pump fields.

In conclusion, we have studied the dynamics of resonantly enhanced Raman generation in a double-Λ configuration in Rb vapor. We observed a novel form of dynamic optical bistability based on long-lived atomic coherence, which did not involve an optical cavity or an induced Bragg grating in the medium. This bistability can be easily adjusted with changes to the pump laser fields, which may assist practical applications. Realization of dynamic optical bistability may also be possible in condensed-matter systems [30–32]. As an example application, we used dynamic optical bistability to demonstrate a simple, all-optical switch.

We thank M. Lukin for the use of his laboratory, and M. D. Eisaman, L. Childress, V. A. Sautenkov, and P. Zoller for useful and stimulating discussions. Financial support was provided by NSF Grant No. PHY-0113844, the Defense Advance Research Project Agency, the Office of Naval Research, NASA, and the Smithsonian Institution.

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