Transient coherence oscillation induced by a detuned Raman field in a rubidium Λ system

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The temporal evolution of coherent population trapping (CPT) was observed in rubidium atomic vapor when sudden changes were made to the detuning of a weak Raman field. The subsequent creation and destruction of CPT are caused by the temporal oscillations of optically induced Raman coherence, with their period depending on the Raman detuning. The oscillating signal was observed over a time of order of tens of milliseconds, and the dependence of the relaxation time on the cell temperature and laser power were investigated. The main features of the experimental observations were well explained by the time-dependent density-matrix equations.

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I. INTRODUCTION

The coherent interaction between an atomic system and light leads to a quantum coherence between coupled states of the atom. Of special interest is the case where a three-level atom composed of two ground-state hyperfine levels and one excited level interacts with two optical fields that constitute a Λ system. In this case, the ground-state coherence, referred to as Raman coherence, leads to novel optical phenomena such as electromagnetically induced transparency (EIT), lasing without inversion [1–5], dark states [6–8], coherent population trapping (CPT) maser [9], and more recently research on slow light [10].

A ''dark state'' is a superposition of internal atomic ground states with a definite amplitude and phase, such that the excitation amplitudes induced by the optical fields in the Λ system interfere destructively. Thus, after a few fluorescence cycles, most of the atomic population is optically pumped to the dark state. A perfect dark state, however, can only exist when the constituent states and optical fields sustain their relative phase. If the relative phase of the optical fields changes, the composition of the nonabsorbing superposition state changes, and accordingly, atoms are pumped to a new nonabsorbing state. The phase determines the dynamics and the steady state of the system.

The case for constant phase has been theoretically analyzed by Kosachiov *et al.* [11,12]. The creation and destruction of CPT as a function of time-dependent phase was observed in a double-Λ system [13–15]. Affolderbach *et al.* [16] reported on experiments with cesium vapor in a bichromatic standing wave where regions of EIT periodically interchange with regions of electromagnetically induced absorption, depending on the relative phase of the two local frequency components in the standing wave.

Transient excitation of a three-level system has been reported by many authors. Transient gain was first observed experimentally from a sodium atom with a Λ configuration by Fry *et al.* [17]. Zhu considered the conditions required for observing inversionless gain in the transient regime for V schemes [18] and Λ schemes [19]. Intentional phase modulation to produce such transient excitation was applied using several methods: phase modulation of the microwave oscillator which drives the electro-optic modulator [20], controlling the magnetic field [21,22], illuminating temporally separated laser fields [23], or using additional Raman laser fields [24].

Camparo *et al.* [25,26] studied transient phenomena of optical transmission (or absorption) in the rubidium ladder system interacting with a rf field and an optical field. They could observe the transient oscillation at the Rabi frequency of the rf field at the moment when the phase of the rf field was changed. Jyotsna *et al.* [27] explained theoretically that Rabi oscillations could be produced with only optical fields without the rf field in the atomic Λ system, but they disappeared as the field intensity was reduced. In contrast, Vanier *et al.* [28] could observe an oscillating behavior of the coherent microwave emission when weak optical fields, having some Raman detuning, were switched on in the cesium Λ system. In this case, the transient oscillation is produced by the periodic creation and destruction of CPT, depending on the Raman detuning.

In this paper we are concerned with the temporal evolution of the CPT signal in the rubidium Λ system when the Raman resonance condition is suddenly destroyed by changing the Raman detuning without changing the field intensity. This phenomenon is similar to the case where the Raman resonance condition is suddenly modified by the application of a magnetic field, producing a Zeeman shift of the ground-state sublevels [21,22]. However, in our case we focus on two aspects: first, where the period of oscillation depends only on the Raman detuning, and second, where the decay time of the oscillation depends on the cell temperature and laser power. In addition, an analytical solution is presented to compare with our experimental observations.

II. THEORY

Let us consider a closed Λ-type, three-level system with ground states $|1\rangle$ and $|2\rangle$, and excited state $|3\rangle$, as illustrated

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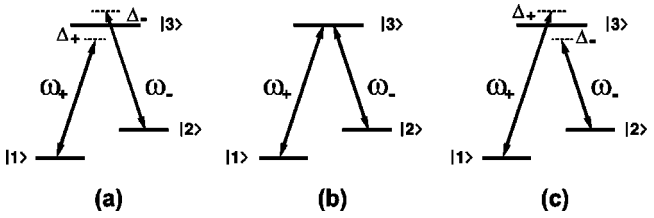

FIG. 1. Coherently coupled Λ-type three-level system with Raman detuning $\Delta_R<0$ (a), $\Delta_R=0$ (b), $\Delta_R>0$ (c).

in Fig. 1. $|1\rangle$ and $|2\rangle$ stand for the pair of levels within the hyperfine split $5S_{1/2}$ rubidium ground state, coupled by the laser fields to the common excited level $|3\rangle$ with the $5P_{1/2}$ manifold. The transition $|1\rangle\leftrightarrow|3\rangle$ of frequency ω_+ and the transition $|2\rangle\leftrightarrow|3\rangle$ of frequency ω_- are driven by two first-order sidebands created by an electro-optical modulator (EOM).

The atoms are considered to be at rest, and the atomic sample is assumed to be homogeneous. These assumptions are justified by the fact that the Raman resonance condition is unaffected by the Doppler effect. In addition, we do not account for the effect of the optical intensity distribution in the beam profile. The Zeeman structure of the ground level of Rb is well separated in the absorption spectrum by a suitable magnetic field. So we consider a Λ system consisting of two ground-state Zeeman sublevels $|5S_{1/2};F=1;m_F=0\rangle$ and $|5S_{1/2};F=2;m_F=0\rangle$. The semiclassical density-matrix equations of motion under the electric-dipole and the rotating-wave approximations can be written as

$$\dot{\rho}_{11}=\Gamma_{31}^{*}\rho_{33}+i\frac{\Omega_+}{2}(\rho_{31}-\rho_{13}), \tag{1a}$$

$$\dot{\rho}_{22}=\Gamma_{32}^{*}\rho_{33}+i\frac{\Omega_-}{2}(\rho_{32}-\rho_{23}), \tag{1b}$$

$$\dot{\rho}_{33}=-(\Gamma_{31}^{*}+\Gamma_{32}^{*})\rho_{33}+i\left[\frac{\Omega_+}{2}(\rho_{13}-\rho_{31})+\frac{\Omega_-}{2}(\rho_{23}-\rho_{32})\right], \tag{1c}$$

$$\dot{\rho}_{21}=[i\Delta_R(t)-\gamma]\rho_{21}+i\left[\frac{\Omega_-}{2}\rho_{31}-\frac{\Omega_+}{2}\rho_{23}\right], \tag{1d}$$

$$\dot{\rho}_{31}=[i\Delta_+(t)-\Gamma^{*}]\rho_{31}+i\left[\frac{\Omega_+}{2}(\rho_{11}-\rho_{33})+\frac{\Omega_-}{2}\rho_{21}\right], \tag{1e}$$

$$\dot{\rho}_{32}=[i\Delta_-(t)-\Gamma^{*}]\rho_{32}+i\left[\frac{\Omega_-}{2}(\rho_{22}-\rho_{33})+\frac{\Omega_+}{2}\rho_{12}\right]. \tag{1f}$$

Here $\Omega_\pm$ and $\Delta_\pm(t)$ are the Rabi and detuning frequencies, respectively, and $\Delta_R(t)=\Delta_+(t)-\Delta_-(t)$ is the Raman detuning. In our case the following conditions are satisfied: $\Omega_+=\Omega_-(=\Omega)$, $\Delta_+(t)+\Delta_-(t)=0$, and $|\Delta_+(t)|=|\Delta_-(t)|(=\Delta_R/2)$. Since our model is a three-level closed system, we assume a branching ratio of unity, i.e., $\Gamma_{31}^{*}=\Gamma_{32}^{*}=\Gamma$ where Γ_{31}^{*} and Γ_{32}^{*} are the spontaneous decay rates of the excited state $|3\rangle$ to the ground states $|1\rangle$ and $|2\rangle$, respectively, and $\Gamma^{*}\equiv(\Gamma_{31}^{*}+\Gamma_{32}^{*}+\gamma)/2\approx\Gamma$ where γ is the nonradiative decay rate between two ground states.

The gain-absorption coefficient for the two fields coupled to the transitions $|1\rangle\leftrightarrow|3\rangle$ and $|2\rangle\leftrightarrow|3\rangle$ is proportional to $\mathrm{Im}[\rho_{31}]$ and $\mathrm{Im}[\rho_{32}]$, respectively. Thus, we concentrate on these components of the density matrix to compare with our experimental observations. In our case, the transient behaviors of $\mathrm{Im}[\rho_{31}]$ and $\mathrm{Im}[\rho_{32}]$ are qualitatively the same. They all oscillate with the same frequency. The optical coherence ρ_{31} (or ρ_{32}) adiabatically follows the Raman coherence ρ_{21}, because the optical coherence builds up rapidly while the Raman coherence remains negligibly small [28]. We first solve for ρ_{31} (and ρ_{32}) in zero order; that is, $\rho_{33}=0$ and $\rho_{11}=\rho_{22}=1/2$ and near the Raman resonance ($\Delta_R\ll\Gamma$). Then, the optical coherence is simply given in terms of ρ_{21} by

$$\rho_{31}=-\frac{\Omega}{\Gamma^2}(\Delta_R-i\Gamma)\left(\frac{1}{2}+\rho_{21}\right). \tag{2}$$

Replacing this expression in the equation for $\dot{\rho}_{21}$, we obtain

$$\dot{\rho}_{21}=-\frac{(\Gamma+i\Delta_R)\Omega^2}{2\Gamma^2}-\left(\gamma+\frac{\Omega^2}{\Gamma}-i\Delta_R\right)\rho_{21} \tag{3}$$

with the additional condition

$$\frac{\Omega^2}{\Gamma^2}\ll 1. \tag{4}$$

Solving Eq. (3) with $\rho_{21}(t=0)=0$, and substituting its solution for the steady state into Eq. (2), we obtain

$$\mathrm{Im}[\rho_{31}^{s}]=\frac{\Gamma(\gamma^2+\Delta_R^2)\Omega+\gamma\Omega^3}{2[\Gamma^2(\gamma^2+\Delta_R^2)+2\gamma\Gamma\Omega^2+\Omega^4]}. \tag{5}$$

Consider the case where the Raman detuning is suddenly changed from zero detuning [Fig. 1(b)] to Δ_R [Fig. 1(c)] at $t=0$. The imaginary part of the optical coherence is obtained from Eqs. (3) and (2) as

$$\begin{aligned}\mathrm{Im}[\rho_{31}]=\mathrm{Im}[\rho_{31}^{s}]&+\frac{\Delta_R\Omega^3(\gamma\Gamma+\Gamma^2+\Omega^2)e^{-(\gamma+\Omega^2/\Gamma)t}}{2\Gamma^3(\gamma\Gamma+\Omega^2)[\Gamma^2(\gamma^2+\Delta_R^2)+2\gamma\Gamma\Omega^2+\Omega^4]}\\&\times[\Gamma(\gamma\Gamma-\Delta_R^2+\Omega^2)\sin(\Delta_R t)\\&-\Delta_R(\gamma\Gamma+\Gamma^2+\Omega^2)\cos(\Delta_R t)].\end{aligned} \tag{6}$$

It is interesting to note that the optical coherence decays at a rate $\gamma+\Omega^2/\Gamma$ from

$$\mathrm{Im}[\rho_{31}^{0}]=\frac{\gamma\Omega}{2(\gamma\Gamma+\Omega^2)} \tag{7}$$

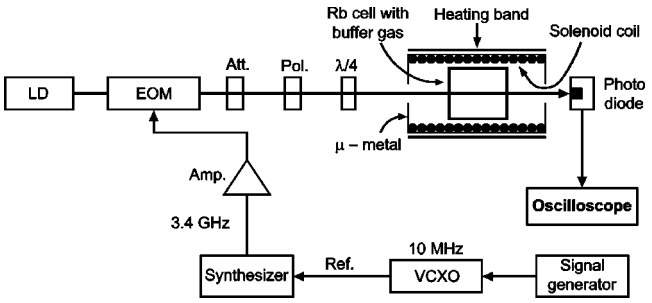

FIG. 2. Experimental setup.

to $\mathrm{Im}[\rho_{31}^{s}]$ and oscillates at a frequency Δ_R until equilibrium is reached. The main aspect of the optical coherence behavior can be described by a cosine-damped oscillation because the cosine term is dominant in the oscillation term of Eq. (6) for

$$2\Delta_R > \gamma + \frac{\Omega^2}{\Gamma}. \tag{8}$$

III. EXPERIMENTAL SETUP

The experimental setup is shown schematically in Fig. 2. The experiments were performed in a rubidium vapor cell with Ne as the buffer gas, at a temperature of 30–50 ○C. The cell wound with solenoid coils was placed in a μ-metal cylinder to provide shielding from the geomagnetic field perpendicular to the cylindrical axis.

We employed the $5^2S_{1/2}, F=1 \rightarrow 5^2P_{1/2}, F=2$ and $5^2S_{1/2}, F=2 \rightarrow 5^2P_{1/2}, F=2$ (i.e., D_1) transitions of ^{87}Rb atom. An external-cavity diode laser was used for the constant laser source. The laser linewidth was about 200 kHz [29]. An EOM was used to produce two first-order side modes with a frequency difference matching the splitting of the levels $|1\rangle$ and $|2\rangle$ (which is 6834 MHz). This EOM was driven by a tunable rf generator and a 10 MHz VCXO that was used as the reference frequency for the rf generator. The detuning of the two side modes was controlled by the input voltage of 10 MHz VCXO. In this way the two side modes form a Λ configuration with the atomic levels. The two side modes had equal intensities, and they were detuned at the same time by the same amount, but in opposite directions.

The two phase-correlated laser fields, which were circularly polarized and copropagating through the rubidium cell, coupled the ground states $|5S_{1/2}; F=1; m_F=0\rangle$ and $|5S_{1/2}; F=2; m_F=0\rangle$ to a common level of the $5P_{1/2}$ excited state. CPT signals due to transitions between other magnetic sublevels are separated by applying a 60-mG magnetic field. To make the condition for CPT, we drove two laser frequencies by carefully adjusting the rf to the input of the EOM. The phase-modulation index of the EOM was set to $m_\phi \approx 0.9$ and the carrier-to-sideband ratio was approximately 4:1. The intensity of the ω_+ and ω_- components was estimated to be a few tens of nW/cm^2. The input laser beams were collimated to a diameter of about 8 mm as they passed through the Rb vapor cell.

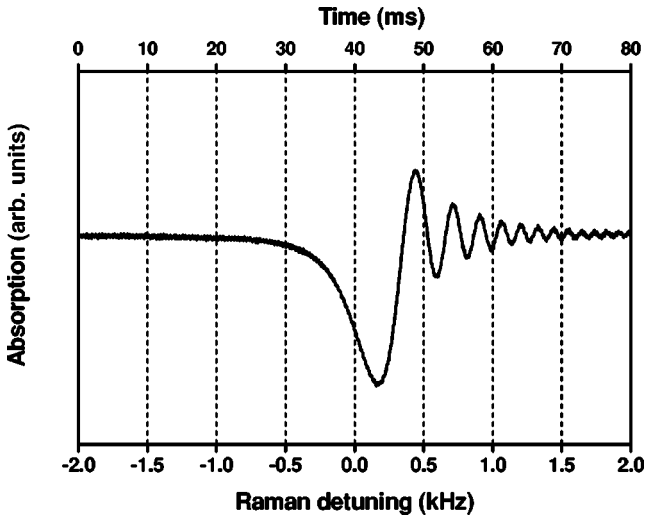

FIG. 3. The observed CPT signal has an oscillating tail. The total input laser intensity $I=0.2\ \mu$W/cm^2. The temperature of the cell is 30 °C.

IV. RESULTS AND DISCUSSION

When the laser frequency was continuously swept by triangular modulation from a red Raman detuning [Fig. 1(a)] to a blue Raman detuning [Fig. 1(c)] at a rate of about 50 kHz/s, the temporal evolution of the CPT signal was observed as shown in Fig. 3. As the Raman detuning gradually changed from the resonance, the subsequent periodic oscillation was observed. The period of this oscillation became shorter as the Raman detuning of the laser from the resonance increased. We obtained the same result (not shown here) from the simulation using Eqs. (1). From both results we found that the position of maximal CPT (minimal absorption) moved in the sweep direction as the sweep rate increased, but the decay time of the oscillation did not change. The similar transient phenomenon was studied in three-level ladder system [25] and two-level system [30]. It should be noted that, as described above, CPT mechanism plays an important role in transient phenomenon in this three-level Λ system.

Now we focus on the oscillation tail, the period of which decreases linearly as a function of the Raman detuning from the resonance. To characterize the transient behavior of the CPT resonance, stepwise Raman detuning was periodically made from zero [Fig. 1(b)] to Δ_R [Fig. 1(c)]. For this experiment, the VCXO was driven by a square-wave voltage. The voltage was changed at time $t=0$. Figure 4 shows three examples of the absorption signals for different values of Δ_R. The absorption amplitude of the plots has been normalized in order to show the difference in the oscillation periods. The absorption signal evolves toward a new steady state with a damped oscillation. The damping rate of the oscillation closely follows an exponential decay. Since the detuning is fixed at a constant value, the period of the oscillation does not depend on time, unlike that seen in Fig. 3 for continuous sweeping. The measured periods of oscillations are quite consistent with the inverse of the Raman detunings. These results agree with the theoretical prediction.

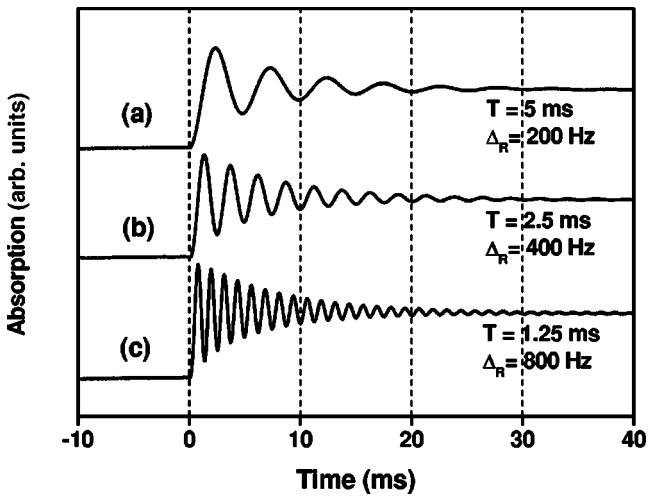

FIG. 4. Observed transient evolution of the CPT signal for different Raman detunings. (a) $\Delta_R = 200$ Hz, (b) $\Delta_R = 400$ Hz, (c) $\Delta_R = 800$ Hz.

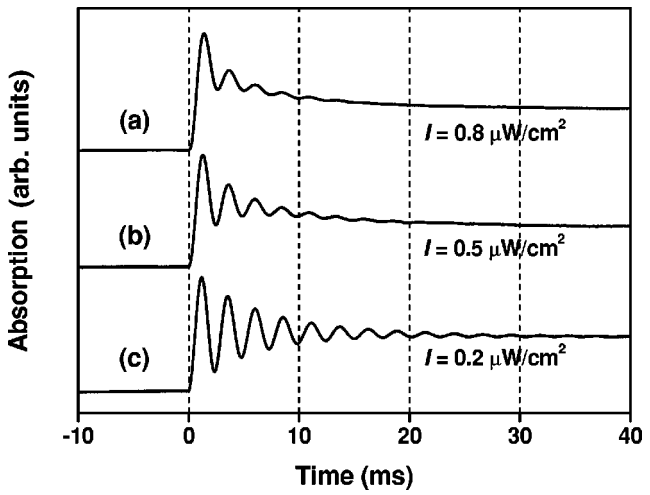

FIG. 6. Observed transient evolution of the CPT signal for different field intensities. (a) $I = 0.8\ \mu$W/cm^2, (b) $I = 0.5\ \mu$W/cm^2, (c) $I = 0.2\ \mu$W/cm^2.

We found that the decay of the atomic coherence (ρ_{31} or ρ_{32}), and of the corresponding oscillation, continues over 30 ms, and the decay rate is not changed by altering Δ_R. This long evolution is related to the long-lived Raman coherence. However, the decay of the dark state arising from atomic collisions [31], as well as from possible couplings to states outside the system, limits the evolution time. In previous work [20–23,32], most interest was focused on time scales comparable to the transition period ($\sim 10^{-8}$ s). However, in this paper, the entire decay time is of the order of tens of milliseconds. This long time is important, for example, in light storage experiments where the decay of spin coherence is related to the delay time or storage time [33]. For characterization of the transient behavior for this long time scale, we have conducted an experiment to show the dependence of the decay on the cell temperature and the laser power.

First, we changed the cell temperature. Experimental transient results are shown in Figs. 5. The measured amplitudes of the signals were normalized to compare their decay rates more precisely. Collisions with the buffer gas cause the Rb atoms to rapidly change their velocity, yet the ground-state coherence survives for typically $>10^7$ collisions [34]. The decoherence by spin exchange collisions between Rb atoms depends on the cell temperature [35,36]. In our simulation, we chose the values $\gamma = \Gamma/(3\times 10^5)$, $\Gamma/(6\times 10^5)$, and $\Gamma/(9\times 10^5)$ to match the calculated results to the measured results at temperatures of 50 °C, 40 °C, and 30 °C, respectively. In this way the experimental results were well reproduced by Eq. (6).

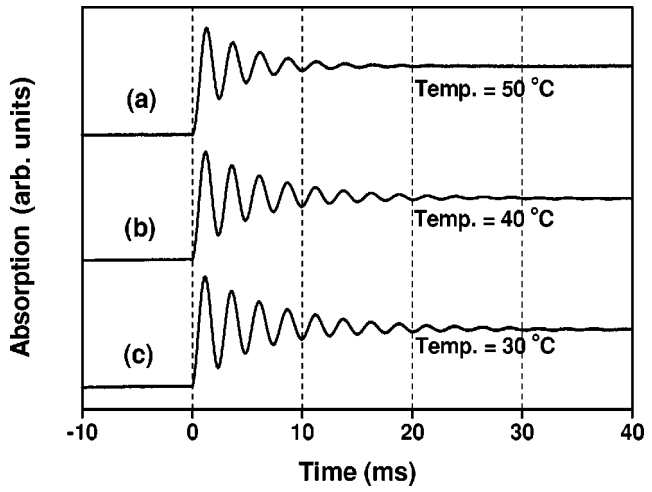

FIG. 5. Observed transient evolution of the CPT signal for different cell temperatures. $\Delta_R = 400$ Hz.

Next, we performed measurements for different laser intensities. The laser beam diameter was kept at 8 mm, while its intensity was varied by an attenuator. If the laser intensity increases, the position of the resonance shifts [37]. Therefore, we must adjust the laser frequency to exactly control the frequency of the Raman detuning. Figure 6 shows the experimental results obtained. At high intensity the temporal evolution deviates significantly from a single cosine-damped oscillation as shown in Fig. 6(a). It can be better described by the sum of a sine-damped oscillation plus an exponentially decaying term. However, the decay rate of this nonoscillating term does not appear in Eq. (6), because we considered only a closed Λ system and internal atomic dynamics. For more detailed explanation of the experimental results for high temperature and high intensity, one should consider inhomogeneous effects [38,39]. In any case, the simulation results show the main features of the experimental observation: that the decay rate is an increasing function of the laser intensity. The atomic coherence may be broken quickly by a detuned Raman field of high intensity: the atoms are optically pumped to a new noncoupled state generated by the detuned Raman field.

V. CONCLUSION

The transient evolution of atomic absorption of a Raman field was observed in a three-level Λ system. We have observed that the CPT signal has an oscillating tail. The oscil-

lating features were investigated by sudden change of the Raman detuning. The period of oscillation depends only on the Raman detuning. The decay rate of the oscillating CPT signal increases with the field intensity and cell temperature. The main features of the experimentally observed transients can be well reproduced by a simple theoretical model based on the analytical solution of density-matrix equations.

The observed effect may have potential applications for creating nonlinear optical media with periodically changing susceptibility, or time-resolved gratings. We suggest that the frequency modulation method can be used to enhance Raman coherence, which is of interest in the fields of atomic frequency standards based on CPT and quantum information storage.

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