Two-photon free-induction decay with electromagnetically induced transparency

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Received December 21, 2009; revised March 11, 2010; accepted May 11, 2010;
posted May 14, 2010 (Doc. ID 121824); published May 28, 2010

We report the observation of coherent two-photon free-induction decay (FID) in a three-level Λ system excited by a low-intensity square-modulated laser pulse. Using electromagnetically induced transparency in an ^{85}Rb cold atomic cloud, for what we believe to be the first time, we observe FID signals with coherence time exceeding the relevant atomic excited-state natural lifetime. Because of the on-resonance enhancement, the two-photon FID signal can be observed at the falling edge without using heterodyne means. © 2010 Optical Society of America

OCIS codes: 190.4180, 020.1670.

Transient optical phenomena associated with coherent interaction of radiation field with matter have been a fascinating aspect of the field of atomic, molecular, and optical physics. These fundamental studies have led to the development of many powerful analytical tools, such as nuclear magnetic resonance (NMR) spectroscopy and the atomic clock. Following the invention of the laser, there was much interest in optical coherent transitions from a two-level system, which is formally equivalent to the well-studied NMR [1]. This led to the realization of optical analogs of spin resonance effects such as nutation and free-induction decay (FID) [2,3]. Soon afterwards, two-photon nutation and FID in three-level systems were studied and demonstrated [4–8]. Recently, FIDs have also been reported in cold atoms [9,10] and linked to the phenomenon of optical precursors [11].

However, all two-photon FID experiments in the past focused on three-level ladder schemes. As a result, the FID temporal duration is limited by the atomic lifetimes of the excited states. Meanwhile, to avoid single-photon absorption, the applied laser fields are often far detuned from the intermediate states. Therefore, the two-photon FID signals are always very weak and can only be indirectly detected by heterodyne means through interference between radiation and the driving field.

In this Letter, we report the observation of coherent two-photon FIDs in a three-level Λ system excited by a weak square-modulated laser pulse. Using electromagnetically induced transparency (EIT) [12] in a ^{85}Rb cold atomic cloud, the two-photon coherence between the two ground states is very long. Moreover, the elimination of single-photon on-resonance linear absorption allows us to tune both two driving laser fields on atomic resonances to enhance the two-photon interaction. As a result, we are able to demonstrate, for the first time (to our knowledge), direct observation of two-photon FIDs at the falling edge of the square pulse without having to use heterodyne means. We obtain FID signals with a temporal length more than four times longer than the atomic lifetime of the excited state.

We work with ^{85}Rb cold atoms in a two-dimensional (2D) magneto-optical trap (MOT) at a low optical depth of about 2. Schematics of the three-level Λ EIT system is shown in Figs. 1(a1) and (b1). A coupling laser (ω_c) is applied to the transition $|2\rangle \to |3\rangle$, and a weak probe laser (ω_p) to the transition $|1\rangle \to |3\rangle$. Δ denotes the coupling laser detuning. The two-photon detuning is defined as $\delta = \omega_p - \omega_c - \Delta\omega_{21}$, where $\Delta\omega_{21}$ is the energy difference between $|1\rangle$ and $|2\rangle$. When the coupling laser is on resonance [$\Delta = 0$, Fig. 1(a1)], the EIT transparency occurs at the probe single-photon resonance, as shown in the probe transmission spectrum in Fig. 1(a2). In the off-resonance EIT case with $\Delta = 2\pi \times 10$ MHz in Figs. 1 (b1) and (b2), the induced transparency lies also at $\delta = 0$. In both cases when the two-photon detuning is zero, the two-photon coherence time can be very long because of the two long-lived ground states.

The experimental apparatus has been described in our recent work [13]. The 2D MOT has a length of $L = 1.5$ cm and a temperature of about 100 μK. We periodically run the system with an MOT trapping time of 4.5 ms followed by a measurement window of 0.5 ms. At the end of the trapping time, all the atoms are optically pumped to the ground level $|1\rangle$. During the measurement window when

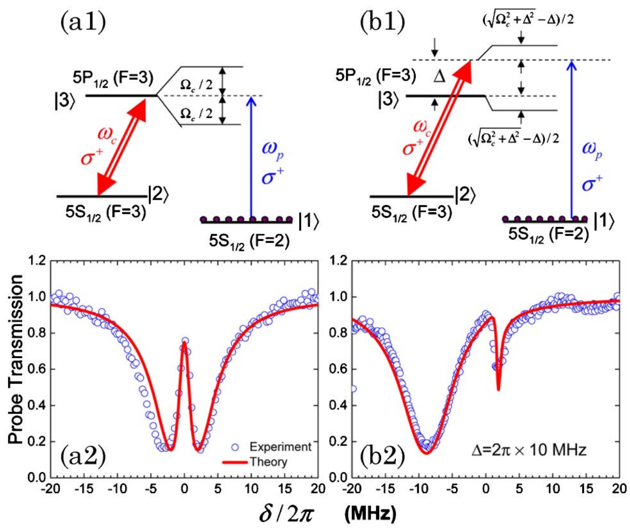

Fig. 1. (Color online) Schematics of ^{85}Rb energy levels for (a1) on-resonance and (b1) off-resonance EIT systems. (a2) and (b2) are, respectively, the measured on-resonance and off-resonance EIT transmission as functions of the two-photon detuning δ, taken at an optical depth $\alpha_0 L = 2$ and coupling Rabi frequency $\Omega_c = 1.4\gamma_{13}$.

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the probe and coupling lasers are on, all the MOT laser beams are switched off. The MOT magnetic field has a transverse gradient of 10 Gauss/cm and remains on all the time. The coupling laser is aligned collinearly with a small angle of 2° respect to the probe beam. The weak square-modulated probe pulse propagates through the cold atoms and is detected by a photon-multiplier tube (Hamamatsu, H6780-20, 0.78 ns rise time). The data are recorded by a 1 GHz real-time digital oscilloscope (Tektronix, TDS684B) and averaged over 30 traces.

We focus on two-photon FIDs excited by a weak step-modulated probe pulse. The probe optical field is described classically as $\frac{1}{2}E_p(z,t)e^{i[k_p z-\omega_p t]}+c.c.$ with $k_p=\omega_p/c$. With the probe laser that is sufficiently weak, the atomic population remains mostly at the ground state $|1\rangle$. This ground state approximation allows us to avoid the complexity of the density matrix approach. Instead, we will study the atomic response using $|\Psi(t)\rangle = a_1(t)|1\rangle + a_2(t)|2\rangle + a_3(t)|3\rangle$. In the counterrotating reference frame, with $a_1 \simeq 1$ and two-photon resonance condition $\delta=0$, we have the Schrodinger equations for the probability amplitudes a_2 and a_3:

$$\dot{a}_2 = -\gamma_{12}a_2 + i\frac{\Omega_c^*}{2}a_3,$$
$$\dot{a}_3 = i\frac{\Omega_p}{2} + i\frac{\Omega_c}{2}a_2 + i(\Delta + i\gamma_{13})a_3, \tag{1}$$

where $\Omega_p = E_p\mu_{31}/\hbar$ and $\Omega_c = E_c\mu_{32}/\hbar$ are the probe and coupling Rabi frequencies, respectively. μ_{ij} is the electric dipole matrix element between $|i\rangle$ and $|j\rangle$. $\gamma_{13} = 2\pi \times 3$ MHz is the electric dipole relaxation rate between $|1\rangle$ and $|3\rangle$. The two-photon dephasing rate between the two ground states $|1\rangle$ and $|2\rangle$ is measured to be $\gamma_{12} = 0.07\gamma_{13}$. The probe wave propagation is governed by the slowly varying envelop equation:

$$\frac{\partial E_p(z,t)}{\partial z} + \frac{1}{c}\frac{\partial E_p(z,t)}{\partial t} = \frac{i}{2}\frac{\omega_p}{c\varepsilon_0}P(z,t), \tag{2}$$

where $P(z,t) = 2N\mu_{13}a_3$ is the induced electric dipole at the probe transition with atomic density N.

We first consider the transient response of the on-resonance ($\Delta=0$) EIT system from an input step-off on-resonance pulse $E_{\text{in}}(t) = E_0\Theta(-t)$, where Θ is the Heaviside function. At time $t=0$ before the probe field is switched off, the atoms are prepared in a dark state: $|\Psi(0)\rangle \simeq |1\rangle - \frac{\Omega_p}{\Omega_c}|2\rangle$ [14]. With this initial condition, one can solve the master Eq. (1) by setting $\Omega_p(t>0)=0$ and obtain

$$a_3(t>0) = \frac{-i\Omega_p}{\Omega_e}\sin\left(\frac{\Omega_e t}{2}\right)e^{-(\gamma_{13}+\gamma_{12})t/2}, \tag{3}$$

where the effective coupling Rabi frequency is defined as $\Omega_e = \sqrt{|\Omega_c|^2 - (\gamma_{13}-\gamma_{12})^2}$. At low optical depth when the propagation effect is negligible, Eq. (2) gives an approximate solution for the FID radiation field:

$$E_{\text{fid}}(t) \simeq \frac{i\omega_p L}{2c\varepsilon_0}P(t) = \frac{\alpha_0 L\gamma_{13}}{\Omega_e}E_0\Theta(t)\sin\left(\frac{\Omega_e t}{2}\right)e^{-(\gamma_{13}+\gamma_{12})t/2}, \tag{4}$$

where $\alpha_0 = Nk_p|\mu_{13}|^2/(\varepsilon_0\hbar\gamma_{13})$ is the on-resonance absorption coefficient. Equation (4) shows some interesting features about the two-photon FID. Similar to that in a two-level system, the FID field is proportional to the optical depth $\alpha_0 L$ due to the collective coherent effect. However, different from the FID in the two-level system with an initial excited macroscopic electric dipole, the initial electric dipole of the EIT system is zero, because the atoms are prepared in the dark state. After the probe laser is switched off, the stored two-photon coherence is released by the coupling laser. There are three characteristic times that determine the transient pattern. The first is the Rabi time $\tau_r = 2\pi/\Omega_e$ of the oscillation period. The second is the excited-state dipole relaxation time $\tau_{13} = 1/\gamma_{13} = 53$ ns. The third is the ground state dephasing time $\tau_{12} = 1/\gamma_{12} = 758$ ns. Because τ_{12} is very long

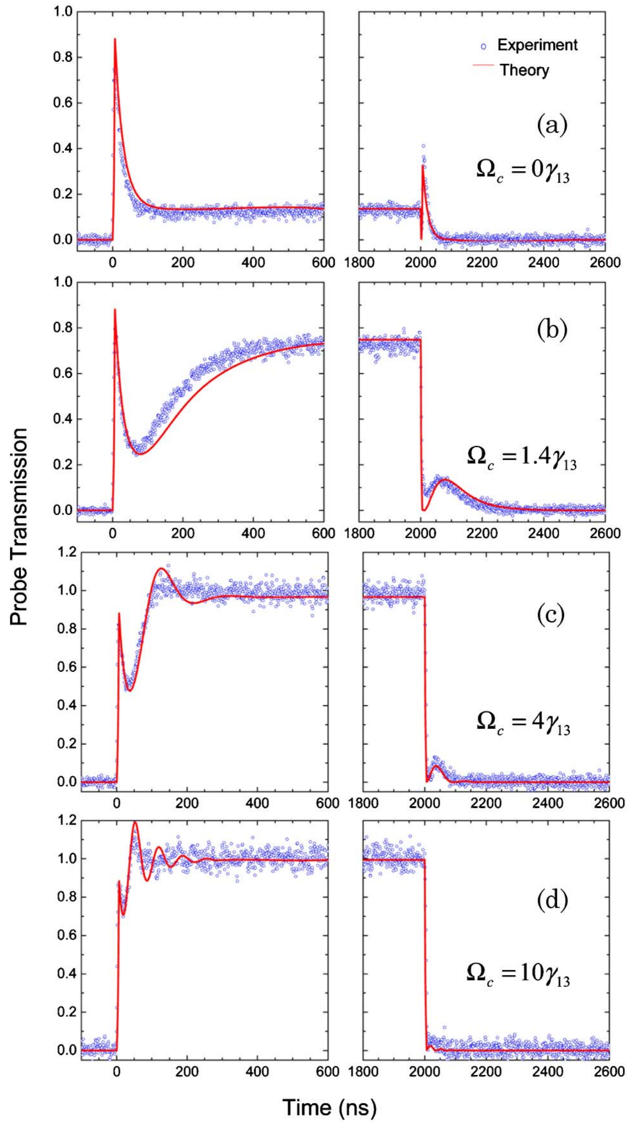

Fig. 2. (Color online) Two-photon FIDs with on-resonance ($\Delta = 0$) EIT excited by a weak square probe pulse driven by different coupling laser powers at an optical depth $\alpha_0 L = 2$.

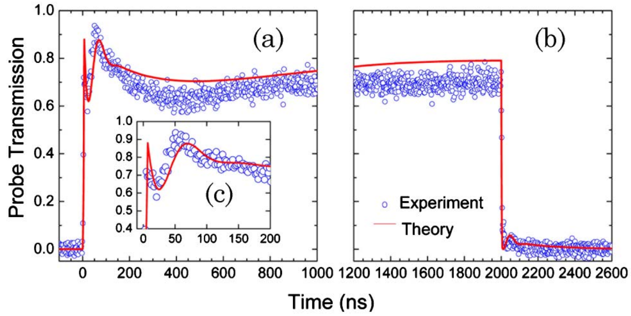

Fig. 3. (Color online) Two-photon FIDs with off-resonance ($\Delta = 2\pi \times 10$ MHz) EIT excited by a weak square probe pulse at $\alpha_0 L = 2$ and $\Omega_c = 1.4\gamma_{13}$.

compared to the other two times, the FID duration is determined by Max$\{\tau_r, \tau_{13}\}$. The physics can be understood in the following two-photon process picture. As the coupling laser is weak, it takes a very long time to release the atoms from the dark state and the time scale is mainly determined by the coupling Rabi frequency. On the other hand, as the coupling laser becomes strong, the atoms are cycled between $|2\rangle$ and $|3\rangle$, and the leakage of photons to the transition $|3\rangle \to |1\rangle$ is determined by τ_{13}. Therefore, the two-photon FID signal duration can be controlled by the coupling laser. The ultimate coherence time is limited by τ_{12}.

The on-resonance ($\Delta = 0$) EIT experimental data (dots) of probe square pulse transmission with different coupling powers are shown in Fig. 2. The weak input square pulse has a length of 2 μs and a rise (fall) time of 7 ns. The solid red curves are obtained by numerically solving the atom-field coupled Eqs. (1) and (2). When the coupling laser is off, Fig. 2(a) shows the FID in the two-level system where the exponential decay is determined by γ_{13}. When we apply the coupling laser, the FID signals are no longer a single exponential decay curve. At $\Omega_c = 1.4\gamma_{13}$, the long transient duration of about 200 ns agrees with the Rabi time τ_r and is four times longer than τ_{13}. As we further increase the coupling power, the transient signal becomes weaker and shorter. These behaviors can be explained in the dressed-state picture, as shown in Fig. 1(a1), where the excited state is split into two with an energy separation of $\hbar\Omega_c$. The radiations from these two dressed states interfere and result in beating. We believe that the transient signals after the falling edge are the first direct observation of two-photon FID without using heterodyne means.

We then study two-photon FID in the off-resonance EIT system. Figure 3 shows the results with $\Delta = 2\pi \times 10$ MHz and $\Omega_c = 1.4\gamma_{13}$. Compared to that of the on-resonance case in Fig. 2(b), the transient signal is much weaker and is barely detectable at the falling edge. The oscillation time of about 100 ns at the rising edge is well described by the effective Rabi time $2\pi/\sqrt{|\Omega_c|^2 + \Delta^2} \simeq 2\pi/\Delta$. This can be explained again in the dressed-state picture, depicted in Fig. 1(b1).

While the observed FID signals are consistent with the theory, we note there is slight mismatching at a long time scale in Figs. 2(c), 2(d), and 3. As shown in the theoretical curves in Figs. 2(c) and 2(d), the damped oscillations last for 300 ns following the rising edge. However, these oscillations are hardly observed in the experimental data after $t > 100$ ns. For the off-resonance EIT case in Fig. 3, the mismatching also occurs as $t > 200$ ns. Besides the detection noise, the degradation of the measured two-photon coherence time may result from the multi-Zeeman-state effect, inhomogeneous broadening caused by the remaining magnetic field, and laser frequency drift during the average time, which cannot be easily counted by the simple three-state EIT model.

The work was supported by a grant from the Research Grants Council of the Hong Kong Special Administrative Region, China (project HKUST600809). Some pieces of equipment used in the experiment were provided by the Joyce M. Kuok Lasers and Photonics Laboratory and the William Mong Clusters Laboratory.

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