Enhanced Cross-Phase Modulation Based on a Double Electromagnetically Induced Transparency in a Four-Level Tripod Atomic System

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We report experimental observations on the simultaneous electromagnetically induced transparency (EIT) effects for probe and trigger fields (double EIT) as well as the enhanced cross-phase modulation (XPM) between the two fields in a four-level tripod EIT system of the D1 line of $^{87}$Rb atoms. The XPM coefficients (larger than $2 \times 10^{-5}$ cm$^2$/W) and the accompanying transmissions (higher than 60%) are measured at a slight detuning of the probe field from the exact EIT-resonance condition. The system and enhanced cross-Kerr nonlinearities presented here can be applied to quantum information processes.

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Cross-Kerr nonlinearity, or the so-called cross-phase modulation (XPM), has received more attention since it can find applications in demonstrating quantum phase gates (QPG) [1–3], generating quantum entanglement of single photons [4], constructing a near deterministic controlled-NOT gate [5], and performing a nondestructive Bell-state detection [6]. However, the absence of sufficient XPM in conventional media becomes an obstacle to its applications in quantum information processing (QIP). Electromagnetically induced transparency (EIT) technology is a promising avenue for solving the problem [2–4]. The large cross-Kerr nonlinearity in a four-level N-type EIT system has been theoretically proposed [7] and experimentally demonstrated in cold atoms [8]. However, in the N-type EIT system, the large cross-Kerr nonlinearity between probe and trigger (signal) pulses may not occur due to their group velocity mismatching [9–11]. Recently, several schemes have been proposed to obtain a large XPM based on the simultaneous EIT for two weak fields in various multilevel systems [2,3,10,11]; that is because in this case the group velocity matching will be possible, thus the long interacting time for producing a large XPM can be obtained. Until now, to the best of our knowledge, the experimental demonstration of a large XPM effect between two weak fields based on double EIT has not been presented. The experimental search for the effect is highly anticipated. Under this motivation we carried out this experimental study.

In this Letter, we report the first experimental observations on double EIT and the enhanced cross-phase modulation between the two fields in a four-level tripod EIT system of the D1 line of $^{87}$Rb atoms. The relevant atomic levels are shown in Fig. 1 of Ref. [12]. The probe field $E_p$ of frequency $\omega_p$ is left circularly polarized ($\sigma^-$), with a Rabi frequency $\Omega_p = \mu E_p/\hbar$, coupling between the transitions from levels $|a_i+1\rangle$ to $|e_i\rangle$ ($i = 1, 2$). The coupling field $E_C$ of frequency $\omega_C$ is also $\sigma^-$ polarized, with a Rabi frequency $\Omega_C = \mu E_C/\hbar$, to drive the levels $|b_{i+1}\rangle$ to $|e_i\rangle$ transitions ($i = 1, 2, 3$). The trigger field $E_T$ of frequency $\omega_T$ is right circularly polarized ($\sigma^+$), with a Rabi frequency $\Omega_T = \mu E_T/\hbar$, coupling to the levels $|b_i\rangle$ to $|e_i\rangle$ transitions ($i = 1, 2, 3$), $\mu$ is the dipole moment for $^{87}$Rb D1 transitions. In this case, the system is coherently prepared into two four-level tripod-type systems [12]. One is formed by the levels $|a_2\rangle - |b_3\rangle - |b_1\rangle - |e_1\rangle$ and the other by the levels $|a_3\rangle - |b_2\rangle - |b_2\rangle - |e_2\rangle$. The $\sigma^-$ probe transition and $\sigma^+$ trigger transition share a common excited state $|e_1\rangle$ ($|e_2\rangle$), which induces a coupling between the probe and trigger fields in such a coherently prepared tripod system, so that the cross-Kerr phase modulation between the two fields will be enhanced. The total probe (trigger) susceptibility $\chi_P$ ($\chi_T$) should include the contributions of both systems; that is, $\chi_P = \chi_{P1} + \chi_{P2}$ ($\chi_T = \chi_{T1} + \chi_{T2}$). In the present measurement of the XPM between the probe and trigger beams, the condition of $|\Omega_{p,T}|^2 \ll |\Omega_C|^2$ is satisfied. Solving the density-matrix equations of Eq. (3) in Ref. [12] for these conditions, under the steady-state condition [2,3], the susceptibilities $\chi_{pi}$ ($i = 1, 2$) and $\chi_{Ti}$ ($i = 1, 2$) are obtained:

$$\chi_{P1} = \frac{N|\mu_{ai,1,ei}|^2}{\hbar\epsilon_0} \left\{ \frac{\rho_{ai,1,ai+1} - \rho_{ei,ei}}{\Delta_T + |\Omega_{Ti}|^2/4\Delta_{PT} + |\Omega_C|^2/4\Delta_{PC}} - \frac{|\Omega_{Ti}|^2(\rho_{bi,bi} - \rho_{ei,ei})/4}{\Delta_{PT}(-\Delta_T^* + |\Omega_C|^2/4\Delta_{TC})(-\Delta_P + |\Omega_C|^2/4\Delta_{PC})} \right\}$$

$$\chi_{T1} = \frac{N|\mu_{bi,bi}|^2}{\hbar\epsilon_0} \left\{ \frac{\rho_{bi,bi} - \rho_{ei,ei}}{\Delta_T - |\Omega_{Pi}|^2/4\Delta_{PT} + |\Omega_C|^2/4\Delta_{TC}} - \frac{|\Omega_{Pi}|^2(\rho_{ai,ai+1} - \rho_{ei,ei})/4}{\Delta_{PT}(-\Delta_P^* + |\Omega_C|^2/4\Delta_{PC})(-\Delta_T + |\Omega_C|^2/4\Delta_{TC})} \right\},$$

where $\Delta_P = \Delta_P + i\gamma_0$, $\Delta_T = \Delta_T + i\gamma_0$, $\Delta_{PT} = \Delta_P - \Delta_T + i\gamma_1$, $\Delta_{PC} = \Delta_P - \Delta_C + i\gamma_2$, $\Delta_{TC} = \Delta_T - \Delta_C + i\gamma_3$, and $\Delta_P$, $\Delta_C$, and $\Delta_T$ are the probe, coupling, and trigger frequency detunings [12], respectively. $\gamma_i$ ($i = 0, 1, 2, 3$) describes decay of populations and coherences. $\rho_{ai,ai}$, $\rho_{bi,bi}$, and $\rho_{ei,ei}$ are the populations of states $|ai\rangle$, $|bi\rangle$, and $|ei\rangle$, respectively.
\[ \Omega_{P1} = \Omega_p C_{ei,ai+1}, \quad \Omega_{Cl} = \Omega_C C_{el,bi+2}, \quad \text{and} \quad \Omega_{T1} = \Omega_T C_{ei,bi} \]

are the probe, coupling, and trigger Rabi frequencies for the transitions from Zeeman levels \([|a_i\rangle + 1] \rightarrow |e_i\rangle\), \([|b_{j+2}\rangle \rightarrow |e_i\rangle\), and \([|b_j\rangle \rightarrow |e_i\rangle\) respectively. \(C_{ei,bi}(ak)\) is a coupling coefficient related to the dipole moment \(\mu_{ei,bi}(ak)\) for the transition from level \([|b_j\rangle \langle a_k|) \rightarrow |e_i\rangle\) with a expression \(\mu_{ei,bi}(ak) = C_{ei,bi}(ak) \mu\). For comparing the cross-Kerr non-linearity produced from the four-level tripod systems with and without EIT, we respectively calculated the XPM coefficients in the two different systems from Eqs. (1). For the EIT scheme, the calculated XPM coefficients are \(n_{P,E}^{(2)} = 4.2 \times 10^{-6} \text{cm}^2/\text{W}\) and \(n_{T,E}^{(2)} = 7.3 \times 10^{-6} \text{cm}^2/\text{W}\) for \(\Omega_p = 70 \text{ MHz}\) and \(\Omega_P = \Omega_T = 6 \text{ MHz}\), respectively, and with a slight probe detuning \(\Delta_P = -0.5 \text{ MHz}\) from EIT-resonance (\(\Delta_C = \Delta_T = \Delta_P = 0\)). The accompanying probe and trigger absorptions are 42% and 57%, respectively. However, in the conventional scheme, if the probe (or trigger) absorption equals ~42% (~57%), the system has to operate at the large detunings of \(\Delta_p = -200 \text{ MHz}\) and \(\Delta_T = 200 \text{ MHz}\) to avoid the very strong resonance absorption. In this case, the calculated XPM coefficients are \(n_{P,E}^{(2)} = 3.78 \times 10^{-9} \text{ cm}^2/\text{W}\) and \(n_{T,E}^{(2)} = 0.94 \times 10^{-9} \text{ cm}^2/\text{W}\), respectively. The calculation shows that the EIT enhancement factors of \(n_{P,E}^{(2)}/n_{P,C}^{(2)} = 1000\) and \(n_{T,E}^{(2)}/n_{T,C}^{(2)} = 5000\) are achieved in the presented scheme.

Figure 1 depicts the experimental setup. LD1 (for probe beam) and LD2 (for coupling and trigger beams) are the frequency-stabilized diode lasers (linewidths ~1 MHz) with grating coating. The LD2 laser beam is split into two parts by a beam splitter (BS1). One of them serves as the trigger beam, and the other one serves as the coupling beam. The trigger beam passes through an acousto-optical modulator system for scanning its frequency around \(\omega_{be}\) (see Ref. [12] for the details). The EIT dispersion curves of the probe and trigger beams are measured with a common Mach-Zehnder interferometer. The Mach-Zehnder interferometer consists of two beam displacing polarizers, BD1 and BD2, which play the role of the two beam splitters used in Fig. 2 of Ref. [13]. The linearly polarized input probe beam (with a chosen polarization angle) is separated into two orthogonally polarized beams, s- and p-polarized beams, by BD1. They are used for the probe and the probe reference beams, and they copropagate along line \(L_2\) and line \(L_1\), respectively. Similarly, the linearly polarized input trigger beam is also separated into orthogonal s- and p-polarized output beams by BD1. The p-polarized trigger beam propagates along line \(L_2\) and overlaps with the s-polarized probe beam. The s-polarized trigger reference beam propagates along line \(L_3\), which is parallel with line \(L_2\). The s-polarized coupling beam propagates through a Rb cell with a small angle (~1°) relative to line \(L_2\) and overlaps with the probe and trigger beams inside the Rb cell. The s-polarized probe and coupling beams become \(\sigma^-\) polarized, while the p-polarized trigger beam becomes \(\sigma^+\) polarized after they respectively pass through a \(\lambda/4\) wave plate. The length of the atomic cell without buffer gas is \(l = 50 \text{ mm}\) with magnetic shielding, and its temperature is stabilized to about 63.5 °C. A weak magnetic field (~150 mG) in the z direction of the atomic cell (parallel to line \(L_2\)) is applied by means of Helmholtz coils to provide a quantization axis. The \(1/e\) intensity diameters of the probe, trigger, and coupling beams are about 1, 1, and 3 mm at the center of the cell, respectively. The probe and trigger beams are recombined to their original linear polarizations after respectively passing through another \(\lambda/4\) wave plate. The intensities of the probe, trigger, probe reference, and trigger reference beams are detected by detectors D3, D2, D1, and D4, respectively. The transmitted probe (s-polarized) and probe reference (p-polarized) beams from BS2 overlap each other at the exit of BD2 to be combined into beam \(P\) again. Similarly, the trigger (p-polarized) and trigger reference (s-polarized) beams are combined into another beam (T) at the exit of BD2. Then, the \(P (T)\) beam passes through a MgO:LiNbO\(_3\) crystal Li1 (Li2) and a \(\lambda/2\) wave plate. Similar to Ref. [14], the detectors D7 and D8 as well as D5 and D6 form two homodyne detectors, H1 and H2, respectively. The differential signals \(\Delta I_{H1} \approx 2 |E_{RP}||E_p|e^{-ae/2}k_p n_p l\) and \(\Delta I_{H2} \approx 2 |E_{RT}||E_T|e^{-ae/2}k_t n_t l\) from the H1 and H2 will give the probe and trigger dispersions \(kn_p l\) and \(kn_t l\), where \(E_{RP} (\gg E_P)\) and \(E_{RT} (\gg E_T)\) are the probe and trigger reference fields, respectively.

We observed the EIT windows for probe and trigger fields when coupling detuning \(\Delta_C = 0\). The powers of the coupling, probe, and trigger beams are set to \(P_C = 14 \text{ mW}\) (\(\Omega_C \approx 70 \text{ MHz}\)), \(P_p = 8 \mu\text{W}\) (\(\Omega_P \approx 3 \text{ MHz}\)), and \(P_T = 10 \mu\text{W}\) (\(\Omega_T = 3 \text{ MHz}\)), respectively. When the coupling beam was on, we scanned the probe frequency across \(\omega_{ae}\) to measure the probe absorption spectrum at \(\Delta_T = 0\). An EIT window [Fig. 2(a)] for the probe beam, with a linewidth of ~2 MHz, appears at the resonance \(\Delta_C = \Delta_P = 0\), which derives from the three-level A system \(|b_j\rangle - |e_i\rangle - |a_k\rangle, (|b_j\rangle - |e_i\rangle - |a_k\rangle)\). At \(\Delta_P = 0\), scanning the trigger frequency around \(\omega_{be}\) with the acousto-optical modulator system [12], the trigger EIT signal [Fig. 2(b)] at the resonance of \(\Delta_C = \Delta_T = 0\) with
a linewidth of $\sim 2$ MHz was observed, which derives from another three-level $\Lambda$ system $|b_1\rangle - |e_1\rangle - |b_1\rangle$ ($|b_2\rangle - |e_2\rangle - |b_2\rangle$).

We also observed the cross-phase modulation between the probe and trigger fields under the conditions of different powers for the two beams. The measured results are shown in Fig. 3. During the measurements of Fig. 3, the coupling beam with a power of $P_C = 14$ mW was always on and the probe frequency was scanned across $\omega_{\text{det}}$. First, under the conditions $P_p = 8 \mu$W and $P_T = 300 \mu$W ($\Omega_T = 18$ MHz), i.e., $\Omega_p < \Omega_T$ ($\ll \Omega_T$), we measured the modulation of the probe field by the trigger field. The curves $I$ of Fig. 3(a) and $I'$ of Fig. 3(b), respectively, present the probe EIT absorption $\alpha_p(\omega_p)l$ and dispersion $k\Delta_p(\omega_p)l$ as the function of $\Delta_T$ when the trigger beam is blocked, which derive from the three-level $\Lambda$-type systems. When the trigger beam is applied, the four-level tripod systems are formed and the EIT window [curve II of Fig. 3(a)] and dispersion [curve $I''$ of Fig. 3(b)] from the tripod systems become much larger than the corresponding results from the $\Lambda$-type systems, respectively. The top of the probe EIT window with the trigger beam is obviously beyond that without trigger beam, while in Ref. [12] this phenomenon did not occur. The reason is that the probe beam propagates through the Rb cell with a small angle relative to the trigger beam in Ref. [12], while, in the presented system, the two beams almost totally overlap. Thus, the interaction between two beams in the presented system is larger than that in Ref. [12]. From Fig. 3(b), we can see that the two EIT dispersion peaks of the probe beam occur at $\Delta_p = -0.7$ MHz and $\Delta_p = 0.6$ MHz, respectively. At the left (right) dispersion peak, a XPM phase shift $\Psi_{\text{XPM}} \sim -2.5^\circ$ ($5^\circ$) was achieved, with a transmission of 70%. Next, we measured the modulation of the weaker trigger beam with a power of 10 $\mu$W by the stronger probe beam with a power of 300 $\mu$W. Traces $i$ of Fig. 3(c) and $i'$ of Fig. 3(d) present the trigger EIT absorption and dispersion $k\Delta_T(\omega_T)l_{|\Delta_T=0\rangle}$ signals when the probe beam is blocked, which correspond to the absorption and dispersion at resonance $\Delta_C = \Delta_T = 0$, respectively. Curves $ii$ of Fig. 3(c) and $ii'$ of Fig. 3(d) are the measured trigger EIT absorption and dispersion $k\Delta_T(\omega_T)l_{|\Delta_T\neq0\rangle}$, respectively, when the probe beam is applied. The top of EIT in curve $ii$ of Fig. 3(c) (four-level tripod systems) is far beyond the trace $i$ of Fig. 3(c) (three-level systems) when both the probe and trigger fields are tuned to the dark states ($\Delta_C = \Delta_T = \Delta_p$). Simultaneously, a sharp EIT dispersion curve $ii'$ of Fig. 3(d) appears at $\Delta_C = \Delta_T = \Delta_p = 0$, which is greatly different with that on the trace $i'$ of Fig. 3(d). The trigger XPM phase shift can be calculated through $\Delta(T) = \Delta(T_0) - n_T l_{|T=0\rangle}$ according to the data given from curves $i'$ and $ii'$ in Fig. 3(d). At $\Delta_p = \pm 0.5$ MHz, a maximum of the trigger XPM phase shift $\Delta(T)$ of $\sim \pm 5^\circ$ is achieved.

Successively, we observed the XPM between the weak probe and trigger fields, both of which have the same power $P_p = P_T = 14 \mu$W ($\Omega_p = \Omega_T = 4$ MHz). Curves $I$ of Fig. 3(e) and $I'$ of Fig. 3(f) are the measured probe EIT absorption and dispersion without the trigger beam on, the curve $i$ of Fig. 3(g) and $i'$ of Fig. 3(h) are the measured trigger absorption and dispersion without the probe beam on. When the two beams were on, we simultaneously measured the EIT absorption curves II (probe) in Fig. 3(e) and $ii$ (trigger) in Fig. 3(g), as well as the dispersion curves II' (probe) in Fig. 3(f) and $ii'$ (trigger) in Fig. 3(h) in a scanning of $\Delta_p$, respectively. The results show that the modulations of the probe EIT absorption and dispersion by the trigger beam are small, but the modulations of the trigger EIT absorption and dispersion by the probe beam are quite obvious. At $\Delta_p = \pm 0.5$ MHz, the
measured XPM coefficients in Ref. [2], in which the solute values of XPM coefficients are suppressed down to 5 kHz, a conditional phase shift (ΔNp + ΦN) based on the XPM between the two pulses with one photon will reach 1.2 rad for Ωc = 30 MHz, which may allow us to perform the QPG operation.

In summary, we have experimentally demonstrated the enhanced cross-Kerr nonlinearity based on double EIT. The double EIT windows are important for matching the group velocities of the probe and trigger beams, which produces an enhanced XPM between the two optical pulses [2].

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![Graph](Image)

**FIG. 4** (color online). Curves (A) and (B) are the measured probe and trigger XPM coefficients at Δρ = ΔC ≈ 0.5 MHz as a function of ΔC, respectively, for Pp ≈ Pt ≈ 14 μW. Curves (A’) and (B’) are corresponding theoretical results with experimental parameters γ0 = 3.5 MHz, γ1 = 0.5 MHz, γ2 = 1.5 MHz, γ3 = 1.0 MHz, Ωc = 70 MHz, Ωp = ΩT = 4 MHz, and N = 3.72 × 10^13/cm^2. Curves (C) and (D) in the inset are the accompanying transmissions of probe and trigger beams at Δρ = ΔC ≈ 0.5 MHz.

maximal trigger nonlinear phase shift ~ ± 1.4° is achieved with an absorption of ~74%. However, such a large absorption is not desired in the practical application of QIP.

For exploring the optimal condition to obtain a large XPM and an accompanying small absorption, we measured the XPM phase shifts and the transmissions at the different ΔC under the condition of the weak probe and trigger fields (Pp = Pt = 14 μW). Curves A and B of Fig. 4 plot the measured XPM coefficients n_p^2 and n_T^2 as the function of ΔC. Curves C and D in the inset of Fig. 4 plot the simultaneously measured transmissions. Increasing ΔC, the absolute values of XPM coefficients n_p^2 and n_T^2 go down, but the accompanying transmissions of the probe and trigger beams become larger, which allows the choice of an appropriate coupling frequency detuning to achieve substantial XPM phase shift with a smaller absorption. Figure 4 shows that n_p^2 is much larger than n_T^2. This asymmetry of n_T^2 and n_p^2 is not in agreement with the prediction in Ref. [2], in which the n_T^2 ↔ n_p^2 exchange is symmetric. A possible reason is that the large differences between the probe Rabi frequencies (Ω1 = Ω2 = 1.15 MHz) and the trigger Rabi frequencies (Ω1 = 2.83 MHz, Ω2 = 2 MHz) could result in the difference of the populations of two ground state (|a1⟩ and |b1⟩ (|a2⟩ and |b2⟩). In this case, the calculated populations with Eq. (3) of Ref. [12] are ρ_a1,a2 = 0.38, ρ_a3,a3 = 0.3, ρ_b1,b1 ≈ 0.12, and ρ_b2,b2 ≈ 0.2. At the same time, we theoretically calculated the fitting curves n_p^2 and n_T^2 at Δρ = ΔC = 0.5 MHz with Eq. (1), which are shown in curves A' and B'. The theoretical calculations are in reasonable agreement with the experimental results.

The enhanced XPM in the presented tripod system is based on simultaneous EIT for probe and trigger fields, and thus both the probe and trigger absorptions are small. For example, the trigger XPM coefficient can reach 2 × 10^{-5} cm^2/W with a transmission of ~60% (see Fig. 4). Such a property is much better than that obtained with the N-type system, in which the probe EIT window will move into an absorption peak when a significant XPM phase shift is acquired [8]. If a probe beam with the intensity of ~0.2 mW/cm^2 (Ωp = 1 MHz) is applied, which corresponds to the case that a probe pulse consisting of one photon is tightly focused to a spot size of a half wavelength at 795 nm, for a duration of 1 μs, the trigger XPM phase shift induced by the probe field will reach ~0.001 rad. Such a XPM phase shift may satisfy the requirement [5] of √m0^2 ≈ 5 (the mean photon number n per pulse is on the order of 4 × 10^12); thus, the presented system may have practical applications in QIP based on a photon number quantum nondemolition detector [5,6]. Although the conditional phase shift which is defined as Φ_p^N + Φ_T^N [1] in QPG still cannot reach −π in our experimental conditions, if the laser linewidths of probe and trigger beams are suppressed down to 5 kHz, a conditional phase shift (Φ_p^N + Φ_T^N) based on the XPM between the two pulses with one photon will reach 1.2 rad for Ωc = 30 MHz, which may allow us to perform the QPG operation.

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