Rephasing halted photon echoes using controlled optical deshelving

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Abstract. Quantum coherence control of two-pulse photon echoes has been demonstrated for a rephasing halt, resulting in storage-time extension using atom phase-controlled optical deshelving (optical locking) in a backward propagation scheme for the additional benefit of echo intensity enhancement. Compared with conventional forward two-pulse photon echoes, the backward two-pulse photon echo efficiency is enhanced by 15-fold even in a dilute sample, and the storage time is lengthened by spin dephasing time accelerated by spin inhomogeneous broadening. The mechanism of delayed photon echoes via optical locking is due to the temporal hold of the rephasing process by coherent population transfer to a robust spin state.

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Quantum optical interface between light and a collective ensemble of atoms plays an important role in all-optical (quantum) information processing. Due to the robustness of the spin state compared with its optical counterpart, coherent all-optical control of spin states is an important tool for photon interface onto a matter, where longer coherence time is highly beneficial [1–7]. Compared with a single atom or a spin [8], an atomic ensemble is advantageous for efficient light–matter interactions. Despite the multimode access with an ultrawide bandwidth, the optical version of Hahn echoes (two-pulse photon echoes (2PE)) has been practically limited due to both an extremely low retrieval efficiency and a short storage time confined by the optical phase decay process (Kurnit et al [9]; for Hahn echoes, see Hahn [9]). Because of the trade-off between data absorption and echo emission in a forward scheme, an extremely low retrieval efficiency of 2PE is inevitable [10]. Population inversion by a strong rephasing pulse can be beneficial to classical 2PE to enhance echo efficiency, whereas it causes quantum noise problems such as spontaneous emission decay and stimulated echo gain in quantum memories. Recently, various types of modified photon echoes were demonstrated for quantum memory applications to solve the rephasing-induced quantum noise problem [11–14]. To satisfy the multimode ultralong photon storage requirement in long-distance quantum communications using quantum repeaters, a resonant Raman echo [15] has been discussed for ultralong photon storage via controlled optical deshelving, the so-called optical locking, where the storage time can be lengthened in the order of minutes [16].

In this paper, a photon storage-time extension in a conventional 2PE using optical locking is demonstrated, whereas a related theoretical study has already been reported [17]. The aim of this work is to prove the coherence transfer of rephasing atoms into a spin state to lengthen the storage time. For this, we apply controlled deshelving or the optical locking method. Secondly, we demonstrate the backward echo scheme to overcome the inevitable echo absorption in a conventional (forward) 2PE scheme. Although it is known that a conventional 2PE cannot be accepted as a quantum memory protocol due to rephasing pulse-induced population inversion resulting in serious quantum noises [18], recently suggested methods of double rephasing make a direct use of 2PE in quantum memory applications by lifting off the fundamental population inversion problem [19, 20]. The physics of optical locking applied to 2PE is coherent transfer of rephasing atoms, where rephasing is the key process of photon echoes for time-reversed coherence evolution [17]. The mechanism of rephasing halt in the present paper has also been demonstrated in atomic frequency comb (AFC) echoes [21], where AFC belongs to the category of stimulated (three-pulse) photon echoes [22]. The function of the rephasing halt in the present scheme is obtained by transferring rephasing atoms on the excited state into an auxiliary ground spin state via optical locking [16–19]. Two decades ago, incoherent optical deshelving in 2PE was demonstrated for photon storage-time extension utilizing atom population transfer-based spin gratings [23].

In addition to the optical locking for extending photon storage time in 2PE, we have also demonstrated enhanced photon echoes in a backward propagation scheme suggested for near perfect echo efficiency [24]. Unlike the forward propagation scheme, the critical echo reabsorption is strongly alleviated in the backward scheme only because of echo retracing geometry along the data trajectory. Moreover, the backward scheme is beneficial for aberration corrections when dealing with quantum optical images [25, 26], enabling ultradense spatial multimode operation.
2. Experimental details

Before turning our attention to the experiments, we briefly address the optical locking by B1 and B2 (see figure 1). According to the photon echo theory, the $\pi$ pulse R rephases all atoms excited by D, and then population swapping between the excited state $|3\rangle$ and an auxiliary spin state $|2\rangle$ follows. In figure 1, the $\pi$ pulse B1 functions to temporally hold the coherence evolution of the rephasing atoms by transferring the excited atoms into the auxiliary spin state $|2\rangle$. By this population transfer, the optical coherence is simply transferred into the spin coherence. Although optical rephasing evolution is halted by the B1 pulse with a $\pi/2$ phase shift, the transferred coherence is dephased by spin inhomogeneous broadening. Thus, the delayed photon echo must be deteriorated by spin dephasing time accelerated by spin inhomogeneous width.

Figure 1(a) shows a partial energy-level diagram of a rare-earth Pr$^{3+}$ (0.05 at.%)-doped Y$_2$SiO$_5$ (Pr:YSO). The three laser beams are the modulated output of a ring-dye laser (Tecknoscan) pumped by a 532 nm laser (Coherent Verdi). Relative frequency adjustment for $\omega_1$, $\omega_2$ and $\omega_3$ is achieved by using acousto-optic modulators (Isomet) driven by radiofrequency (rf) synthesizers (PTS 250). Each laser pulse duration of the laser beam is controlled by an rf switch (Minicircuits) and a digital delay generator (SRS DG 535). Figures 1(b) and (c) show the pulse sequence and the propagation scheme of the laser beams. The light P is used for initial preparation of the ground state population redistribution, where the inhomogeneous width is determined by laser jitter, which is $\sim 300$ kHz. For this, both optical pulses at $\omega_1$ and $\omega_3$ are turned on for 10 ms, so that all atoms are incoherently pumped onto the state $|1\rangle$ ($\pm 3/2$, $^3H_4$): $\rho_{11} = 1$; $\rho_{22} = 0$; $\rho_{33} = 0$. We set the delay of R from D at 20 $\mu$s, where optical phase decay time is measured at 25 $\mu$s (see figure 2(d)). The repetition rate of the light pulse train is 20 Hz. The resultant absorption of the data pulse D is $\sim 70\%$, where the optical depth is $d \sim 1.0$.
Figure 2. Demonstration of the phase-locked photon echoes. (a) Phase matching condition. (b) Upper panel: Conventional two-pulse photon echo; lower panel: phase-locked photon echo using optical locking. D and R are saturated on detectors. (c) B1 position invariant phase-locked echoes. (d) Dephasing times of the conventional photon echo ($\Delta T_2$) and the phase-locked photon echo ($T$). In (b) and (c), the red (blue) line is from APD1 (APD2). In the blue line, B1 and B2 are scattered light to denote their positions.

from $I_{\text{out}} = I_{\text{in}} \exp(-d)$ by Beer’s law. All light beams are vertically polarized, and propagate along the crystal axis of the medium (Pr:YSO). A perfect phase conjugate scheme, however, is not satisfied due to the frequency difference between D/R ($\omega_2$) and B1/B2 ($\omega_3$), as shown in figure 1(c).

The controlled desheling pulses B1 and B2 are counterpropagating, and their frequency difference from D and R is 10.2 MHz, which is the energy splitting between two ground states $|1\rangle$ and $|2\rangle$ of Pr:YSO. The angle between D and B1 is 12.5 milliradians with an overlap of $\sim 90\%$ along the 1 mm sample. Within the allowed bandwidth given by modified optical inhomogeneous broadening, the pulse area of D and R is simply adjusted according to the pulse duration. An avalanche photodiode APD1 detects light in the line of D and R. With (without) B1 and B2 turned on, the echo direction becomes backward (forward) due to the phase matching condition, as shown in figures 1(c) and 2(a). The second avalanche photodiode APD2 detects the backward echo E. Both APD-captured signals are fed directly into a digital oscilloscope and recorded by averaging 30 samples. The spot diameter ($\exp(-2)$ in intensity) of B1, B2, D and

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R is 330, 200, 300 and 300 µm, respectively. The light power of R, D, B1 and B2 is 0.5, 0.5, 12 and 24 mW. The optimum power of light of B1 and B2 is predetermined by Rabi flopping measurement, where the B1 (B2) pulse area is set at π (3π). This nonidentical control pulse pair is to satisfy the phase recovery condition of 2π [17]. The Pr:YSO sample is in a liquid helium cryostat (Advanced Research System) kept at a temperature of ∼5 K, where one can use a closed cycle cryostat with the great benefit of energy saving.

3. Results and discussion

Figure 2 presents an experimental demonstration of the rephasing halt applied to 2PE in figure 1 using optical locking pulses, B1 and B2. According to the phase matching condition, the halted echo signal E satisfies the following:

\[ \vec{k}_E = \vec{k}_D - \vec{k}_{B1} + \vec{k}_{B2}, \]  

(1)

\[ \omega_E = \omega_D - \omega_{B1} + \omega_{B2}, \]  

(2)

where \( \vec{k}_i \) and \( \omega_i \) are the wave vector and angular frequency of pulse \( i \), respectively. Equations (1) and (2) denote a coherence transfer-based four-wave mixing resulting in a spin coherence \( \rho_{12} \) (excited by B1) readout signal, echo E (excited by B2). Unlike resonant Raman coherence excitation [15, 16], the spin coherence \( \rho_{12} \) is created via optical population transfer only by the deshelving pulse B1. Therefore, the four-wave mixing signal as an echo must depend on both optical (rephasing) and spin parameters (dephasing). Although E is not a phase conjugate of D, it actually works as a phase conjugate if the spatial shape of B1 and B2 is properly controlled for correlation and convolution theorem [27]. Conventional 2PE followed by D and R are detected by APD1, and the delayed (or halted) echo E is detected by APD2.

The upper panel of figure 2(b) represents the conventional two-pulse photon echo captured by APD1 (without B1 and B2) as a reference. The measured echo efficiency (echo intensity ratio to the intensity of D) is less than 1%. The lower panel of figure 2(b) represents the halted echo E, where \( T \) (B2 delay from B1) is 5 µs. The delay of B1 from R is 7 µs. The calculated delay of echo E from B2 in equation (3) matches the observed delay at 13 µs as shown in figure 2(b):

\[ T_E = T_{B2} + (T_R - T_D) - (T_{B1} - T_R), \]  

(3)

where \( T_x \) stands for the rising time of pulse \( x \) (for a more detailed analysis, see [17]). Because the auxiliary spin state \( |2\rangle \) in figure 1 is not isolated, but closely connected to state \( |1\rangle \) with a spin dephasing time \( \tau \), the delay \( T \) is limited by \( \tau \), which is given by the inverse of the spin inhomogeneous width.

In figure 2(b), the measured halted echo (blue) intensity is twice that of the conventional 2PE (red). Given the spin-dephasing-induced coherence loss of 67% during B1 and B2 (\( \exp(-5/9) \approx 0.33 \equiv \alpha \); discussed in figure 2(d)) and given the area ratio of B2 to D on the focal point (\( (2/3)^2 \approx 0.4 \equiv \beta \)), we conclude that the enhancement factor of 15 \( 2/(\alpha \beta) \) is achieved due to backward propagation of E, where the echo E traces back mostly along the same path as the data pulse. This is the first proof of [24], where the backward echo scheme was proposed to overcome the forward echo scheme-based lower retrieval efficiency discussed in [10]. Regarding storage-time extension, figure 2(b) demonstrates a direct proof of the rephasing halt in 2PE in the limit of fast spin dephasing time of 9 µs (explained in figure 2(d)).
Figure 2(c) proves equation (3), where the echo position $T_E$ is independent of the delay $B_1$ from $R$ for a fixed $T$. As discussed in [17], the experimental results in figure 2(c) prove that the delay $B_1$ does not change the echo intensity either, since the only function of $B_1$ and $B_2$, respectively, is to lock and unlock the rephasing process triggered by $R$. With the delay of $B_1$ from $R$ being longer than $(T_R - T_D)$, no halted echo signal is observed (not shown). Under the condition of a long spin dephasing time, the present scheme of using optical locking can be applied to tunable quantum optical delay [28].

In figure 2(d), the photon echo intensities are measured as a function of delay of $R$ (upper axis, $\Delta T_2$) without $B_1$ and $B_2$, and as a function of delay of $B_2$ (lower axis, $T$) for both fixed $R$ and $B_1$. We calculate the decay time $\tau$ for the $R$ delay at $\tau = 25 \mu s$ and that for the $B_2$ delay at $\tau = 9 \mu s$: $I_e(t) = I_e(0)\exp(2t/\tau)$, where $I_e$ is the intensity of echoes. The $B_2$ delay-dependent decay time $\tau$ is analogous to the inverse of the spin inhomogeneous width ($\Delta_{\text{spin}} = 1/\pi \tau = 35 \text{kHz}$) between the ground hyperfine states $|1\rangle$ and $|2\rangle$ [29]. This calculated inhomogeneous value is similar to that (30 kHz) measured by the rf-optical resonance method. The $R$ delay-dependent decay time is analogous to the inverse of the optical homogeneous decay rate, which increases rapidly as the temperature increases in Pr:YSO. To extend photon storage time using optical locking pulses, an external magnetic field is used to extend the spin dephasing time $\tau$ [30].

Figure 3(a) shows collective coherence transfer by the controlled desheling pulses. The conventional two-pulse photon echo intensity (filled squares; see also red echo in figure 2(b)) is gradually transferred into the rephasing halted (or delayed) echo (open circles; see blue echo in figure 2(b)) as $B_1$ pulse duration $\Delta T$ increases for a fixed $B_2$. At $\Delta T = 1.6 \mu s$, the 2PE is completely transferred into the delayed echo $E$, which denotes maximum population transfer from the state $|3\rangle$ to the state $|2\rangle$ with a $\pi$ pulse area of $B_1$. Figure 3(b) denotes that the $B_2$ pulse area satisfies $(4n - 1)\pi$, as discussed in [17]. For a fixed pulse area of $B_1$ ($1.6 \mu s$, $\pi$ pulse area), the first maximum (of the echo signal) appears at $0.9 \mu s$ of $B_2$ pulse duration, and the second and third maxima appear at $\sim 2.2$ and $\sim 3.6 \mu s$, respectively. These pulse durations correspond roughly to $3\pi$, $7\pi$ and $11\pi$, respectively, satisfying the phase recovery condition of...
the controlled deshelving pulse set B1 and B2: \( \Phi_{B2} = (4n-1)\pi \) for \( \Phi_{B1} = (4n-3)\pi \), where \( \Phi \) represents a pulse area and \( n \) is an integer [17]. The exact calculations are 0.9 \( \mu s \) for 3\( \pi \), 2.1 \( \mu s \) for 7\( \pi \) and 3.3 \( \mu s \) for 11\( \pi \). These echoes, however, turn out to be saturated due to both optical and spin damping, with a long pulse length of B2 as well as imperfect population transfer. Figure 3 thus demonstrates both population and phase-controlled coherence transfer in the present rephasing halted echoes by using independent control pulses.

4. Conclusion

In conclusion, coherence control of photon echoes using optical locking (atom phase-controlled optical deshelving) was experimentally demonstrated for photon storage-time extension as well as echo intensity enhancement in a rare-earth Pr\(^{3+}\)-doped Y\(_2\)SiO\(_5\). The observed photon echo efficiency in a backward propagation scheme was enhanced by a factor of 15 in a dilute sample. The observed photon storage-time extension was limited by spin inhomogeneous width. A very much longer storage time with near perfect retrieval efficiency can be obtained in an optically dense medium if an external dc magnetic field is applied. Quantum imaging with aberration corrections is another benefit of the present scheme based on the correlation and convolution theorem.

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