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To cite this article: Byoung S Ham 2012 New J. Phys. 14 013003

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Atom phase-locked coherence conversion using optical locking for ultralong photon storage beyond the spin $T_2$ constraint

Byoung S Ham
Center for Photon Information Processing, School of Electrical Engineering, Inha University, 253 Yonghyun-dong, Nam-gu, Incheon 402-751, Korea
E-mail: bham@inha.ac.kr

Received 27 August 2011
Published 5 January 2012
Online at http://www.njp.org/
doi:10.1088/1367-2630/14/1/013003

Abstract. Using on-demand coherence conversion via optical locking, a dynamic coherent control of the collective atom phase has been demonstrated for longer photon storage beyond the critical constraint of spin phase decay time, where the storage time can be extended up to hours in a rare-earth-doped solid. Coherent transient phenomena such as photon echoes have been investigated for frozen phase decay via coherent population transfer using a simple deshelving optical pulse pair. Unlike the rephasing halt applied to two-pulse photon echoes, where optical decoherence is accelerated by spin inhomogeneous broadening, a completely atom phase-locked coherence conversion has been observed in three-pulse photon echoes, resulting in spin dephasing-free coherence control. Here, the mechanism of the atom phase-locked coherence conversion via optical locking has been investigated in a solid medium whose optical transition is imperfect, where partial coherence is lost via optical depth-dependent imperfect population transfer. The relationship between coherence loss and optical depth is analyzed, where nearly perfect photon echo efficiency can be obtained for ultralong photon storage in an optically thick medium.
Coherence conversion between optical and spin states has been an important research topic in nonlinear quantum optics, where recently the quantum optical interface has drawn much attention [1–10]. Compared with robust spin states, optical states are fragile in decoherence due to intrinsically fast decay processes. Thus, coherence conversion from a fragile optical state to a robust spin state has been studied for extended coherence time of collective atoms [7–10].

In particular in ensemble solid media, electromagnetically induced transparency (EIT) [1] has drawn much attention in nonlinear quantum optics for ultraslow light-based quantum optical memories [3], efficient four-wave mixing [11], photon switching [12] and photon logic gates [13]. Compared with atomic media whose EIT condition can be easily reached even far below optical saturation point, most solid media encounter difficulties in satisfying EIT due to a wide inhomogeneous broadening greater than GHz and weak oscillator strength. However, solids have benefits in the application, such as wide bandwidth, robustness and absence of atomic diffusion.

Simultaneous quantum optical interface in a resonant Raman scheme between optical and spin states used for EIT and EIT-based slow light is limited by faster spin dephasing accelerated by spin inhomogeneous broadening [3, 14]. As already shown in resonant Raman echoes, however, reversibility of spin ensemble in a spectral domain overcomes the limited coherence time and extends it up to spin phase decay time of the order of a 100 µs [15]. A recent proposal for extended photon storage using resonant Raman echoes is a breakthrough in quantum coherence control of collective atoms (or ions), where the photon storage time can be extended up to hours [16]. To retrieve input photons, however, an optical readout process of the Raman echoes is necessary, requiring preparation of readout pulses synchronized with data pulses [17].

Recently, quantum coherence control in inhomogeneously broadened optical media was studied theoretically [18] and experimentally [14] for a direct photon coherence control using reversible mapping processes in two-pulse photon echoes. However, the extended photon storage time is still limited by spin dephasing accelerated by spin inhomogeneous width. Here, we present a solution to overcome the critical limitation of quantum coherence control in conventional two-pulse photon echoes or quantum mapping processes for a spin inhomogeneously broadened optical system. For this, a three-pulse photon echo scheme is used, where an intrinsic property of atom phase locking is the key element to be utilized [19].

Like two-pulse photon echoes, three-pulse photon echoes rely on reversibility of inhomogeneously broadened atoms. The difference between them lies in the storage method:
three-pulse photon echoes use population grating, whereas two-pulse photon echoes use phase grating. In both cases the phase and amplitude of input photons are stored and retrieved. Thus, the quantum coherence control applied to two-pulse photon echoes can also be applied to the three-pulse photon echoes via the same population transfer method. As demonstrated for quantum storage in atomic frequency comb echoes using quantum coherence control [20] whose storage mechanism belongs to the three-pulse photon echoes [21], the quantum coherence control applied to the present three-pulse photon echoes has potential for application to quantum memory, too, if the rephasing-induced population inversion problem is solved. Very recently, population inversion-free photon echo protocols have been proposed using a double rephasing technique for quantum memory [22–24].

2. Theory

Figure 1(a) shows partial energy levels of Pr$^{3+}$ ion-doped Y$_2$SiO$_5$ (Pr:YSO) for the present phase-locked coherence conversion scheme using controlled optical population transfer, the so-called optical locking [16]. Figure 1(b) shows the pulse sequence of figure 1(a), where the temporal position of the optical locking pulse pair B1 and B2 at frequency $\omega_2$ can be adjusted. Via the population transfer process each transferred atom from state $|3\rangle$ to state $|2\rangle$ or vice versa gains a $\pi/2$ phase, where the control pulses B1 and B2 at frequency $\omega_2$ must satisfy the following conditions to avoid an unwanted phase shift [18]:

$$\Phi_{B1} + \Phi_{B2} = 4n\pi, \quad (1)$$

$$\Phi_{B1} = (2n - 1)\pi, \quad (2)$$

where $\Phi_{BX}$ is the pulse area of the desheling pulse BX and $n$ is an integer.
For the experiments, the three laser beams in figure 1 are the modulated output of a ring-dye laser (Tecknoscan) pumped by a 532 nm pump laser (Coherent Verdi). Relative frequency adjustment for \( \omega_0, \omega_1 \) and \( \omega_2 \) is achieved by using acousto-optic modulators (Isomet) driven by radio-frequency (rf) synthesizers (PTS 250). The data pulse \( D \) is made by an arbitrary waveform generator (AWG610) for a hyperbolic secant pulse: \( \Omega(t) = \Omega_0 \sec h [\beta (t - t_0)] \), where \( \Omega \) is the maximum Rabi frequency of the pulse and \( \beta \) is real to determine the pulse duration. Other pulses are square pulses. The duration of each pulse of the laser beam is controlled by an rf switch (Minicircuits) and a digital delay generator (SRS DG 535). The repetition rate of the light pulse train is 10 Hz. The lights P, D, W, B1, B2 and Re are generated from the same laser beam. The light pulse \( P (\omega_0; \omega_2) \) is used for initial preparation of ground-state population redistribution in a persistent spectral hole-burning medium of Pr:YSO: \( \rho_{11} = 1; \rho_{22} = 0; \rho_{33} = 0 \). The pulses W (write) and Re (read) are for the conventional three-pulse photon echoes at the frequency \( \omega_1 \) with \( \pi/2 \) pulse area each. The angle between D and B1 is 12.5 mrad with an overlap of \( \sim 90\% \) (\( \sim 80\% \)) along the 1 mm (3 mm) sample. Within the allowed bandwidth (\( \sim 300 \) kHz) given by modified optical inhomogeneous broadening by P, each laser pulse area is simply adjusted by its duration. The avalanche photodiode (Hamamatsu APD)-captured signals are directly fed into a digital oscilloscope and recorded by averaging ten samples. The lights B1 and B2 are predetermined for the pulse area of \( \pi \) and \( 3\pi \) to satisfy equations (1) and (2), respectively, unless otherwise indicated. The Pr:YSO sample in a liquid helium cryostat (Advanced Research System) is kept at temperatures of 5–6 K. All light pulses are vertically polarized and propagate along the crystal axis of the medium (Pr:YSO).

To briefly address the rephasing phenomenon in an inhomogeneously broadened medium and phase-locked coherence transfer in three-pulse photon echoes, time-dependent density matrix equations are directly solved without approximations in figure 2. In an inhomogeneously broadened optical system neglecting all decay parameters, two-pulse photon echoes are solved in figures 2(a) and (b): the pulse area of D at \( t = 5 \mu s \) is set at \( \pi/2 \) for maximum coherence excitation. The pulse \( R \) is for rephasing with a pulse area of \( \pi \). By the rephasing pulse \( R \) at \( t = 20.0 \mu s \), the overall system coherence \( \langle \rho_{13} \rangle \) completely decayed out quickly right after \( D \) shows a full recovery as a photon echo at \( t = 35 \mu s \). This rephasing is the key mechanism of photon storage in the context of the time reversal process with a \( \pi \) phase shift.

We now virtually divide the rephasing pulse \( R \) into two pulses to understand the atom phase-locking mechanism via phase-population coherence swapping between off-diagonal elements \( \rho_{13} \) (coherence between states \( |1\rangle \) and \( |3\rangle \)) and diagonal elements \( \rho_{ii} \) (population in state \( |i\rangle \)), where \( \rho_{33} = 1 - \rho_{11} \). By the atom detuning \( \Delta \)-dependent phase evolution \( \exp(i\Delta t) \), D-excited coherence \( \rho_{13} \) results in a phase grating (see the top panels of figures 2(c) and (d)). Here \( \text{Re}\rho_{13} \) has a \( \pi/2 \) phase shift against \( \text{Im}\rho_{13} \) (not shown). When the rephasing pulse \( R \) comes in at \( t = 20.0 \mu s \), the phase grating \( \rho_{13} \) starts to transfer into the population grating \( \rho_{11} \) (see the middle panel of figure 2(c)). Note here that no population grating exists before \( R \). At the end of the first half of \( R \) at \( t = 20.1 \mu s \) for a \( \pi/2 \) pulse area, the coherence conversion from phase grating \( \rho_{13} \) to population grating \( \rho_{11} \) is completed (see the middle panel of figure 2(d)). At the end of \( R \) at \( t = 20.2 \mu s \) for a \( \pi \) pulse area, the conversion process by the first half of \( R \) is completely reversed, where the initial phase grating is recovered but at the cost of a \( \pi \) phase shift (see the bottom panels of figures 2(c) and (d)). This phase reversal of the atom ensemble makes a coherence burst as an emissive echo at \( t = 35 \mu s \). Here, the three-pulse photon echo system is achieved by simply dividing the \( R \) pulse into identical two pulses (see figure 2(e)), where both the phase and amplitude of \( D \) are stored.
Figure 2. Numerical simulations for coherence conversion. (a) Absorption ($\text{Im} \rho_{13}$) as functions of time and detuning for two-pulse photon echoes. (b) Spectral sum of (a). Inset: pulse sequence. The pulse area of $D$ and $R$ is $\pi/2$ and $\pi$, respectively. Optical inhomogeneous broadening $\Delta = 340 \text{ kHz (FWHM)}$. All decay rates are zero. (c, d) Coherence conversion between $\text{Im} \rho_{13}$ and $\rho_{11}$ by a rephasing pulse $R$ in (b). $R$ is on for $20.0 < t \leq 20.2$. (e, f) Optical deshelving-based coherence conversion in three-pulse photon echoes. (e) Top panel: pulse sequence; bottom panel: extended photon storage. (f) Red: $\text{Re} \rho_{13}$; blue: $\text{Im} \rho_{12}$; green: $\rho_{33}$; dotted: $\rho_{22}$. The dotted curve is overlapping with the blue one. $W$, $B1$ and $B2$ end at $t = 20.1$, $20.2$ and $120.3 \mu s$, respectively. The pulse area of $D$ is $\pi/4$. Each pulse area of $W$ and $\text{Re}$ is $\pi/2$. The pulse area of $B1$ and $B2$ is $\pi$ and $3\pi$, respectively. The pulse length of $D$, $W$, $B1$, $B2$ and $\text{Re}$ is $0.1$, $0.1$, $0.1$, $0.3$ and $0.1 \mu s$. Optical inhomogeneous broadening, $\Delta = 680 \text{ kHz (FWHM)}$. $\Gamma_1 = 0$; $\Gamma_3 = \Gamma_2 = \gamma_3 = \gamma_1 = \gamma_2 = 5 \text{ kHz}$.

in the population grating of both $\rho_{11}$ and $\rho_{33}$. The spectral grating as a population modulation in the optical inhomogeneous broadening is achieved even with nonclassical light of the data pulse $D$.
Figure 2(e) represents the present phase-locked coherence conversion using optical locking in three-pulse photon echoes. Here, the rephasing pulse \( R \) in figure 2(b) is divided into two halves, W and Re, and the control pulse pair B1 (\( \pi \)) and B2 (3\( \pi \)) satisfying equations (1) and (2) are inserted between them. For the coherence transfer via optical locking, an auxiliary ground state \( |2\rangle \) is added. As shown in figure 2(e) (bottom panel), the photon storage time is extended by \( T \), where the storage-time extension \( T \) (B2 delay from B1) is normally longer than the optical population decay time \( T_{\text{opt}} \) \( (T_{\text{opt}} \approx 165 \mu s) \). This photon storage time extension is due to atom-phase locking as discussed in figures 2(c) and (d). Because the three-pulse photon echo is locked to the optical phase decay, the transferred atom coherence into the spin state \( |2\rangle \) is also locked to the spin phase decay process (will be discussed in figure 3).

As shown in figure 2(f), quantum coherence transfer between the optical state (\( \rho_{13} \)) and the spin state (\( \rho_{12} \)) is performed with direct population transfer by the control pulse pair, B1 and B2, where both phase decay processes are locked to the transferred atoms. Here, \( \rho_{22} \) (dotted curve) by B1 is exactly the same as \( \rho_{33} \) (green curve) by W, where \( \rho_{33} = 1 - \rho_{11} \) (see figure 2(d)). Thus, quantum coherence control of collective atoms in three-pulse photon echoes is satisfied for spin dephasing-free photon storage.

### 3. Experiments and discussions

In figure 3, the atom phase-locked coherence conversion applied to three-pulse photon echoes is demonstrated for both forward and backward propagation schemes for different optical depths. The use of two different optical depths is to analyze imperfect coherent population transfer via optical locking, resulting in coherence loss [25]. Since B1 and B2 work for only coherence transfer of the phase-locked atoms in state \( |3\rangle \) to state \( |2\rangle \) (see figure 2(e)), the phase-matching condition is the same as that for the conventional three-pulse photon echoes:

\[
\begin{align*}
\vec{k}_E &= -\vec{k}_D + \vec{k}_W + \vec{k}_{\text{Re}}, \\
\omega_E &= -\omega_D + \omega_W + \omega_{\text{Re}},
\end{align*}
\]

where \( \vec{k}_i \) and \( \omega_i \) are the wave vector and angular frequency of pulse \( i \), respectively. Unlike the optical locking applied to the rephasing process of two-pulse photon echoes [14, 18], the coherently transferred atoms by the optical locking pulses are frozen from both optical and spin dephasing, as discussed in figure 2. Thus, spin dephasing-independent photon storage is obtained.

Figure 3(a) presents a forward propagation scheme, and figure 3(b) shows the results as a function of delay time \( T \) (B2 delay from B1) in a lower optical depth \( d \) (\( d = \alpha l = 1.0; \ l = 1 \text{ mm} \)). The experiments are performed at temperatures of 5–6 K. Due to echo reabsorption in a forward scheme, the measured retrieval efficiency (the echo intensity ratio to the data D intensity at \( T = 0 \)) is extremely weak (0.2%), as expected. The observed coherence decay time \( \tau \) of the optically locked echoes is \( \tau = 165 \mu s \), which is much longer than the spin dephasing time of \( \sim 10 \mu s \) [14] in the coherence control of two-pulse photon echoes. Unexpectedly, however, the \( \tau \) is the same as the optical population decay time \( T_{\text{opt}} \) \( (T_{\text{opt}} \sim 165 \mu s) \) [26]. This is discussed below. The data points in figures 3(b) and (d) are for the peak intensity measurement of each echo signal averaged (see figure 3(f)).

The optical population decay-dependent decoherence phenomenon observed in figure 3(b) is due to remnant (or leaked) population in state \( |3\rangle \), where imperfect coherence control by the B1 pulse results in the remnant population [25]. Although the remnant atoms are decreased as
Figure 3. Observations of ultralong optically locked echoes. (a) Schematic diagram of forward propagation. (b) Observed echoes with B1 and B2 as a function of delay $T$ (in figure 1). The delays of $R$ from B2 and $W$ from $D$ are fixed at 2 $\mu$s. Red lines are for $\{\exp[-(t - \Delta T)/\tau] + n\}^2/(n + 1)^2$, where $\Delta T$ is the minimum delay and $n \geq 0$. The pulse duration of $D$, $W$ and $R$ in figure 1 is set at 1 $\mu$s for the $\pi/2$ pulse area and 4.6 mW in power. The pulse duration of control pulse B1 (B2) is set at 1.2 $\mu$s (3.6 $\mu$s) for the $\pi$ (3$\pi$) pulse area at 14.5 mW in power. The optimum power of the lights B1 and B2 is predetermined by Rabi flopping measurement. An avalanche photodiode APD1 detects light in the line of $D$, $W$ and Re, including conventional photon echoes. The spot diameter (exp$(-2)$ in intensity) of B1, B2, $D$, $W$ and Re is $\sim$ 300 $\mu$m. The Pr:YSO sample in a liquid helium cryostat (Advanced Research System) is kept at a temperature of $\sim$ 5 K. The resultant absorption of a data pulse $D$ is $\sim$ 70%, where optical depth $d$ ($d' = \alpha l$, where $l$ = 1 mm) is 1.0. All light pulses are vertically polarized,
the optical depth increases, the remnant atom-induced three-pulse photon echoes as a defect cannot be distinguished from the optically locked echoes, as discussed in [25]. Thus, the observed decay pattern of the optically locked echo signals in figure 3(b) must show two different decay curves; one by the three-pulse photon echoes in a short time scale and the other by the optically locked echoes in a long time scale. For $t > T_1^{\text{opt}}$, the $T_1^{\text{spin}}$-dependent echo should decay out, while $T_1^{\text{spin}}$-dependent echo dominates. As a result, the echo intensity level ($I_e$) must be determined by the amount of leaked atoms caused by imperfect coherence control by the B1 pulse (in an optically dilute medium):

$$I_e = \left\{ \exp\left[\frac{-(t - 30)}{\tau}\right] + n \right\}^2 / (n + 1)^2,$$  \hspace{1cm} (5)

where $n$ indicates a saturation (or defect) level determined by the optical depth $d = 1.0: n = 1.2$; $\tau = 165$. Equation (5) is obtained intuitively to include the coherence leakage due to the remnant atoms.

Figure 3(c) illustrates a backward propagation (phase conjugate) scheme, and figure 3(d) shows the experimental results of figure 3(c) as a function of delay $T$ in an optically dense medium ($d = 2.4$) using a longer sample ($l = 3$ mm). The measured decay time of the observed echoes is the same as that in figure 3(b). However, the observed signal loss for the defect level $n$ in equation (5) decreases from $\sim 70\%$ in figure 3(b) to $\sim 50\%$ due to reduced remnant atoms in the excited state $|3\rangle$ in a higher optical depth medium, as discussed in figure 3(b). The dash-dot curve indicates the best fit curve of the echo decay, which represents the spin population decay time $T_1^{\text{spin}} = 2$ s at $\sim 6$ K (see figure 9 of [27]): exp$[-(t - 5)/2000] - 0.5$, where $t$ is in ms. Clearly, the existence of remnant atoms due to the imperfect population transfer by B1 causes the systemic coherence leakage. Here, the coherence drop to 50% in figure 3(d) indicates a no-cloning regime without post-selection, where quantum error correction protocols can be applied [28]. Compared with figure 3(b), the higher echo saturation level in figure 3(d) provides the proof of the remnant population-caused coherence leakage [25]. Thus, in an optically dense medium, where nearly perfect population transfer occurs, the decay curve should follow the spin population decay time without the $T_1^{\text{opt}}$-dependent defect in echo intensity (discussed in figure 5). Note that the dash-dot curve in figure 3(d) is temperature dependent, where its longest time is $\sim 100$ s at $\sim 2$ K [27]. In Eu$^{3+}$-doped Y$_2$SiO$_5$, spin $T_1$ is as long as a few hours [29].
Figure 4. Measurement of the optical depth and echo efficiency in the conventional stimulated photon echoes in a backward scheme (see figure 3(c)). (a) Raw data of D and E. ‘Ref. data’ is for D before entering the medium. ‘Data passed’ is for D after passing through the medium. The pulses W and R are placed between D and E (not shown). (b) Integration of each pulse in (a). The measured echo efficiency is ∼ 20% of Ref. data by considering the use of a 50/50 beam splitter. The measured optical depth $d$ is $d = 2.4$.

Owing to the backward propagation scheme, the observed echo retrieval efficiency (at delay $T = 0$) is increased from 0.2% in figure 3(b) to 20%, which is 100 times higher than that in figure 3(b) (see also figure 4). Considering the rough overlap scheme in figure 3(d), nearly 100% retrieval efficiency can be obtained in an optically dense medium if the beams are nearly perfectly overlapped inside the medium. Here, a three-pulse photon echo decay curve should follow $T_{\text{opt}}^{\text{opt}}$ if the delay $T_W$ from $T_D$ is too short compared with the optical phase decay time $T_{\text{opt}}^{\text{opt}}$ ($T_{\text{opt}}^{\text{opt}} \sim 110 \mu s$ in Pr:YSO (see [26])), otherwise, shortens [30]. In figure 3, the delay of write pulse W from the data pulse D is just 2 $\mu s$. Therefore, the observed echoes in figure 3 must be independent of the phase parameters as shown. Here, the temperature of 6 K in the present experiments is of great benefit from a practical point of view because it can be easily obtained by an electrically pumped closed cycle cryostat.

Figure 3(f) shows overlapped echo signals observed in figure 3(d), and the method is shown in figure 3(e). For each data point in figure 3(d), three sets of echoes from figure 3(f) are collected, measured and averaged. As shown in Figure 3(f), the echo intensity becomes decreased but saturated at a defect level as the delay time $T$ increases. The defect level of echo intensity depends on the optical density of the medium as discussed above.

Figure 4(a) shows normalized intensity of each pulse for an optically locked photon echo observed in figure 3(d) for a backward propagation scheme. Figure 4(b) represents the integration of each pulse in figure 4(a). As shown in figure 4(b), the observed backward echo efficiency is ∼ 20% considering the use of a 50/50 beam splitter.

4. Analysis

To support the analysis of imperfect population transfer-induced (or remnant population-induced) decoherence discussed in figure 3, numerical simulations of the optically locked echoes with improper deshelving pulses, B1 and B2, are shown in figure 5. To satisfy the
Figure 5. Numerical simulations of the remnant population-caused coherence leakage. (a–d) Delay $T$ of B2 from B1 is set at 0.9 (red), 10 (blue), 20 (magenta), 40 (cyan), 70 (green) and 110 $\mu$s (black). The positions of light pulses $D$, $W$ and B1 are $T_D = 5 \mu$s, $T_W = 10 \mu$s and $T_{B1} = 10.1 \mu$s, respectively. The pulse duration of $D$, W, B1 and Re is 0.1 $\mu$s. The pulse duration of B3 is 0.3 $\mu$s. The pulses Re and B2 move together, where their separation is 0.1 $\mu$s. The pulse area of $D$ is $\pi/4$. All resultant echoes are superposed. The pulse area $\Phi_{B2}$ of B2 is fixed at $\Phi_{B2} = 0.8\pi$, while $\Phi_{B1}$ varies to test the coherence loss due to imperfect population transfer (or remnant atom-induced coherence loss) by B1 pulse excitation. The $n$ is for equation (6). The optical inhomogeneous width (FWHM) is 680 kHz. $\Gamma_{31} = \Gamma_{32} = \gamma_{31} = 5 $kHz.

remnant atom-caused coherence loss, the following equation is used for the decay curve from equation (5): $y(t) = \{\exp[-(t - \Delta T)/\tau] + n\}^2/(n + 1)^2$, where $\Delta T$ is the shortest $T$ for the first echo data, $\tau$ is $T_1^{\text{opt}}$, and $n$ $(n \geq 0)$ is determined by the remnant atom-caused coherence leakage. In comparison, the pulse area of B1 varies for a fixed B2 ($\Phi_{B2} = 2.4\pi$) not to satisfy equations (1) and (2). For simplification, spin decay rates are set to zero, resulting in no decay if a perfect population transfer is achieved by B1 and B2. The dotted curves refer to equation (5). As shown in figure 5, echo retrieval efficiency increases as coherence leakage (due to remnant atoms) decreases. For all cases in figure 5, the echo decay rate is the same as the optical population decay rate $\Gamma_3$ ($\Gamma_3 = \Gamma_{31} + \Gamma_{32}$) supporting that the observed sudden coherence drop in figures 3(b) and (d) is due to the imperfect (or leaked) population transfer in a dilute sample.
With the same optical depth, the coherence recovery in figure 5(b) (88% in amplitude) and figure 5(d) (46% in amplitude) corresponds to figure 3(d) (50% in intensity; 70% in amplitude) and figure 3(b) (30% in intensity; 55% in amplitude), respectively. Therefore, to satisfy zero coherence leakage of the present atom phase-locked coherence transfer via optical locking, the use of an optically dense medium satisfying perfect transitions (like in nitrogen-vacancy centers of diamond) is essential. Moreover, the use of an optically dense medium hardly deteriorates echo retrieval efficiency in the backward propagation scheme if a perfect overlap is achieved [7, 8, 14].

5. Conclusion

In conclusion, on-demand quantum coherence control of three-pulse photon echoes via optical locking was presented in an optically leaked system for photon storage time extension, where the photon storage time was much longer than the spin dephasing time measured by the same method applied to two-pulse photon echoes. The storage time extension is due to the inherent phase-locking property of three-pulse photon echoes. The observed echo decay features two independent parameters (one for optical population decay as a defect and the other for spin population decay), where imperfect coherence transfer by optical locking pulses induces coherence leakage, resulting in conventional three-pulse echoes which cannot be separated from the optically locked echoes. The defect echo decay is analyzed by the theory of imperfect population transfer, and the results matched quite well with experimental measurements. In a backward propagation scheme, the observed echo efficiency was increased a 100-fold in intensity due to retracing the data pulse trajectory. Thus, the critical limitation of spin dephasing observed in quantum coherence control in two-pulse photon echoes is solved and can be applied to ultralong photon storage even in the case of nonclassical light if the double rephasing technique is combined.

Acknowledgments

This work was supported by the Creative Research Initiative program (grant no. 2011-0000433) of the Korean Ministry of Education, Science, and Technology through the National Research Foundation. J Hahn contributed to the experiment.

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