Control of Photon Storage Time in Photon Echoes using a Deshelving Process

Byoung S. Ham
School of Electrical Engineering, Inha University
S. Korea

1. Introduction

Since the first protocol of quantum algorithm was put forth in 1994 (Shor, 1994), quantum information processing has been intensively studied (Nielsen & Chuang, 2000). The quantum approach has benefits over the classical optical information processing in areas such as prime number factorization (Shor, 1994), data searching (Grover, 1996), and high-resolution lithography (Boto et al., 2000; Yablonovitch, 1999). Compared to conventional cryptography based on public key cryptosystem (RSA cryptosystem) using conventional computers, prime number factorization using quantum computers has demonstrated a potential for a formidable attack on existing cryptographic systems. Like conventional memory which serves in the information processing unit, such as a processing unit together with logic gates, quantum memory is also essential to quantum information and communications networks. For quantum communications via a classical optical channel, the longest communication distance a quantum light can be transmitted is determined by the sensitivity of optical detectors and a lossy classical channel such as an optical fibre. Based on current technologies, the longest distance a single photon can propagate through an optical fibre is about 100 km (Zbinden et al., 1998). This distance should limit applications of quantum information especially for long-distance quantum communications. To solve the limited photon transmission, a quantum repeater has been introduced for virtually unlimited transmission distance (Duan et al., 2001; Jiang et al., 2007; Simon et al., 2007; Waks et al., 2002). Quantum memory is an essential element for the entangled photon swapping in the quantum repeaters. Because quantum repeaters swap entangled photons shared by neighboring remote quantum nodes in a quantum network, and the quantum information must be kept coherently through the quantum network, the minimum storage time of quantum memory is determined by the longest transmission distance of the lossy optical channel. For tranceatlantic quantum communications, roughly a one-second or more storage time is required. So far, such a long photon storage has not been demonstrated, where conventional quantum memory protocols limit the storage time to spin phase decay time at most ($\leq 10^{-3}$ second).

Unlike classical memories, quantum memory must satisfy a coherent process. Since the first observation of coherent retrieval of a stored optical pulse in a Bose Einstein condensate using slow light (Liu et al., 2001), interest in quantum memories has increased in the last decade (Alexander et al., 2006; Afzelius et al., 2010; Chaneliere et al., 2005. Choi et al., 2008;

Ham, 1998; Ham 2009a; Ham, 2010a; Hedges et al., 2010; Hetet et al., 2008; Hosseini et al., 2009; Julsgaard et al., 2004; Kocharovskaya et al., 2001; Kraus et al., 2006; Liu et al., 2001; Moiseev & Kroll, 2001; Moiseev et al., 2003; Neumann et al., 2009; Nilsson & Kroll, 2005; Sangouard et al., 2007; Turukhin et al., 2002; Van der Wal, et al., 2003). Because temporal multimode storage capability is required for the quantum repeaters, a photon echo-type protocol has emerged as a best candidate. Unlike a single atom-based quantum memory, echo-type quantum memory has the advantage of using an ensemble of atoms, where a quantum light is efficiently absorbed by many atoms. This ensemble system also provides near perfect storage capability as well as inherent temporal multimode capability. Following the first observation of echo-type optical memory in a spin system (Hahn, 1950), photon echoes were intensively studied in the 1980s and 1990s for spatiotemporal ultrahigh-speed all-optical information processing. Unlike all-optical memories, retrieval efficiency in quantum memories must satisfy at least a two thirds level of fidelity. In this chapter photon echo type quantum memory protocols are reviewed and compared. The chapter is composed of the following sections. In section 2, photon echoes are reviewed as a background of modified echo-type quantum memories. Section 3 presents the advantages and disadvantages of several modified photon echoes for quantum memory protocols. In Section 4, an optical locking technique is introduced for an ultralong photon storage method that can be applied to long-distance quantum communications. Section 5 discusses a phase matching condition for optical locking applied to different photon echo protocols, solving a main drawback in conventional photon echoes. Section 6 presents conclusions.

2. Review of photon echoes

Like spin echoes (Hahn, 1950), photon echoes (Kurnit, et al., 1964) use optical inhomogeneity of an atomic ensemble. Figure 1 shows numerical simulations of a two-pulse photon echo in a two-level atomic system. The first pulse D in Fig. 1(b) interacting with a two-level optical system excites atoms onto the excited state |2>. For a visualization purpose of maximum coherence, the first pulse D is set at a $\pi/2$ pulse area, where the pulse area Φ is defined by: $\Phi = \int \Omega dt,$ and Ω is the Rabi frequency. By the interaction of the first pulse D, atomic coherence is created between states |1> and |3>. A phase relaxation-dependent decoherence is inevitable in any optical system. Because the atoms are inhomogeneously broadened, randomly detuned atoms from the absorption linecentre cause a fast dephasing of sum coherence for the atomic system. Later but before each individual atom diphases completely, the second pulse R, whose pulse area is π, interacts with all atoms whose sum coherence is washed out, and inverts the system to rephase. The rephasing by the second pulse R results in a time reversal process, where initial coherence should be retrieved after the same elapse as taken with R. Here, the photon echo as a coherent burst has nothing to do with a population transfer process but relates only to coherent phase retrieval of all individual atoms. The retrieval efficiency degrades as a function of time due to the optical phase decay process as well as to optical population decay of the excited atoms. In general, the optical phase decay time in rare-earth doped solids is ~0.1 ms, which is too short to quantum repeaters (Macfarlane & Shelby, 1987). Another problem of the two-pulse photon echoes is the echo reabsorption by the noninteracted (or nonabsorbed) atoms along the propagation direction, common in an optical medium governed by Beer's law, where the number of atoms excited by the light

pulse D is exponentially reduced as a function of propagation distance inside the medium (Sangouard, N. et al., (2007). Because the retrieval efficiency is defined by the ratio of emitted photon echo intensity to the data intensity, an optically dense medium is needed for near 100% data photon absorption. This optically thick medium is, however, disadvantageous to the echo generation due to echo reabsorption. As a result of reabsorption, the observed photon echo efficiency or retrieval efficiency in most rare-earth doped solids is less than 1 %. Hence the original photon echo protocols cannot be adapted for a quantum memory protocol unless the reabsorption problem is solved.
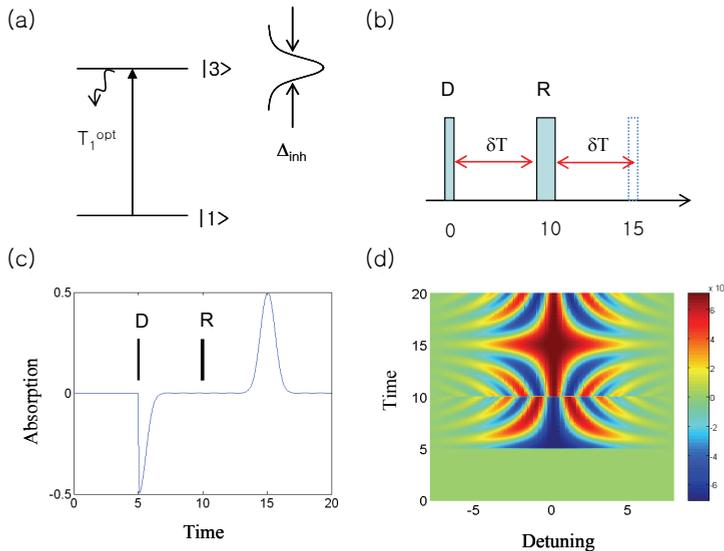

Fig. 1. Two-pulse photon echoes. (a) Energy level diagram interacting with light pulses, (b) pulse sequence for (a), (c) and (d) numerical simulations for (b). The pulse area of D and R is $\pi/2$ and π, respectively. All decay rates are assumed zero for visualization purposes. Optical inhomogeneous width Δ_{inh} is 680 kHz, where Rabi frequency of each pulse is 1 MHz

Compared with the two-pulse photon echoes (Kurnit, et al., 1964), a stimulated photon echo protocol was introduced to lengthen the storage time (Mossberg, 1982). In the stimulated photon echoes, the rephasing pulse R in the two-pulse photon scheme in Fig. 1(b) is divided into two $\pi/2$ pulses – that is W and R [see Fig. 2(a)]. By the first $\pi/2$ pulse, W, the atoms in both ground and excited states become spectrally modulated resulting in a spectral grating or frequency comb as shown in Fig. 2. Because the spectral modulation results from atom population modulation in the frequency domain caused by two consecutive optical pulses, D and W, the lifetime of the spectral grating is determined only by atom population decay time. Since the ground state population decay time is much longer than the optical counterpart, an optical deshelving technique to evacuate the excited atoms to a third state has been developed to increase the lifetime of the spectral grating (Mitsunaga & Uesugi, 1990). Thus, in the stimulated photon echoes, the storage mechanism is free from the optical phase decay process, which is the main storage mechanism to the two-pulse photon echoes. The third pulse R functions to rephase the coherence half-way stopped by W, resulting in a

stimulated echo (not shown). Here, the stimulated echo is a four-wave mixing process in the time domain, where R scatters off the spectral grating made by D and W, thus generating a time-delayed echo signal as shown in Fig. 3(a). The time delay of the echo from R is exactly the same as that between D and W due to the temporal four-wave mixing process. Thus, the storage time in the stimulated photon echoes can be eventually lengthened up to the spin population decay time, which is several orders of magnitude longer than the optical phase decay time (Macfarlane & Shelby, 1987). However, due to the excited state population loss during the storage process, the retrieval efficiency of the stimulated photon echoes must be less than 50%, which cannot satisfy the minimum fidelity of quantum memories.
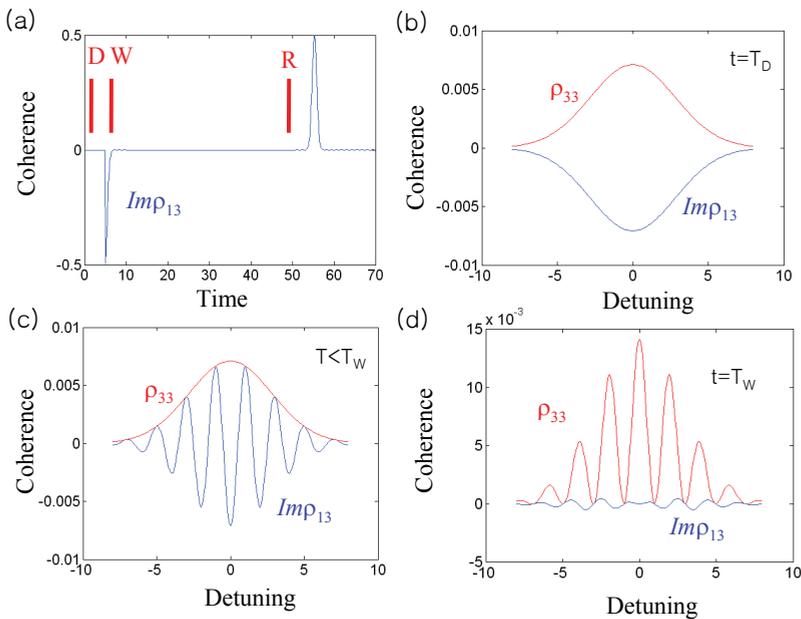

Fig. 2. Numerical simulations of stimulated photon echo. (a) ~ (d) sum of coherence $Im\rho_{13}$ and excited state population ρ_{33}, where ρ_{ij} is a density matrix element defined by $\rho_{ij}=|i><j|$
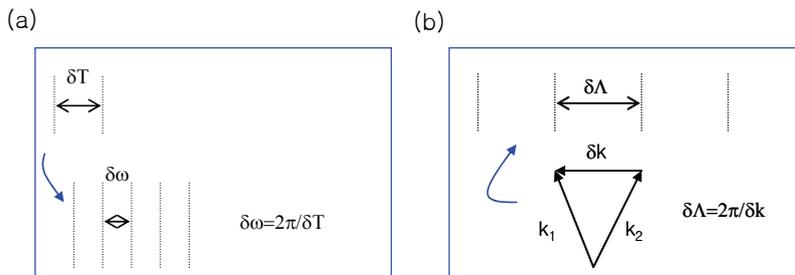

Fig. 3. Schematic diagram of (a) a spectral grating by D and W in Fig. 1(b) and (b) a spatial grating by two angled light beams k_1 and k_2

In both two-pulse and stimulated photon echoes, spontaneous emission noise due to population excitation should be a critical problem in quantum memory applications using single photons. The spontaneous emission noise problem, however, can be practically removed or alleviated if squeezed light or multiphoton entangled light (Marino et al., 2009) is used. Even in single photon-based quantum memory protocols, the spontaneous emission decay-caused quantum noise can be practically removed if an ultrashort pulse is used in a pencil-like geometry, where the pulse duration is still confined by optical inhomogenous width of the optical medium. Although Swiss and Calgary groups jointly criticised that photon echoes cannot be used for quantum memories due to the spontaneous emission noise, it fails with practical conditions in a rare-earth doped solids (Sangouard, N. et al., 2010).

In a rare-earth Pr^{3+} (0.05 at. %) doped Y_2SiO_5, which has been used for most modified photon echo based quantum memories (Afzelius et al., 2010, Ham, 2010d), total atom number per unit volume (cm^3) is 4.7×10^{18} (Maksimov et al., 1969). Either in the two-pulse photon echoes or in the stimulated photon echoes, at least one half the ground atoms are excited and spontaneously resulting in quantum noise. Thus, it seems obvious to say that even one out of 10^{18} atoms could affect the single photon-based echo signal to destroy the quantum fidelity. However, in a pencil-like propagation geometry, whose light cross section is 1 mm in diameter, the interaction volume decreases to 10^{-6} cm^3. For a 100 ps data pulse to cover a 4 GHz inhomogeneous width of the medium, the temporal ratio of the echo to the spontaneous decay time is 10^{-9}. Owing to the symmetry of echo to the data pulse in a virtual sphere made by a 10 cm focal length lens, the area ratio for the echo signal to the noise on the sphere is 10^{-5}. Thus, the effective number of spontaneously emitted photons affected to the echo signal is ~ 0.01. This number is nearly negligible to alter the photon echo fidelity.

3. Modified photon echoes for quantum memory applications

3.1 To solve the echo reabsorption problem in two-pulse photon echoes

Due to Beer's law, a trade-off exists between echo intensity and data absorption in an optically thick medium. If the echo propagation direction can be reversed to trace exactly along the data path, then no echo signals from the excited atoms interact with any nonexcited atoms due to the backward propagation scheme (Moiseev & Kroll, 2001). This idea has been experimentally demonstrated in 2009, where the echo enhancement factor even in an optically dilute medium is 15 times (Ham, 2009b). Another modified protocol to avoid echo reabsorption in the two-pulse photon echoes has been demonstrated by both a Lund group (Nilsson & Kroll, 2005) and Australian groups (Alexander et al., 2006; Hetet et al., 2008) using an electrical Stark effect. Instead of using π rephasing optical pulse, a pair of electrical stripe lines with opposing current flow spectrally controls the Stark effect, resulting in the same effect as the optical π rephsing pulse. Because the Rabi frequency of the electrical pulse is limited in most rare-earth doped solids, this electrical Stark method, however, limits the inhomogeneous width of atoms. Here, atom spectral width or inhomogeneous broadening determines the maximum amount of data, where the inverse of the spectral width determines the minimum pulse duration of the data D. Although echo efficiency can be maximized using this technique, the photon storage time is still limited by the optical phase decay time T_2^{opt} (in the order of 100 μs), which cannot satisfy the storage

time requirement for quantum repeaters (in the order of seconds) used for long-distance quantum communications.

3.2 To solve short storage time in two-pulse photon echoes

In a two-level system, the data pulse D excites optical coherence as mentioned in Fig. 1. Due to decoherence by optical phase decay time T_2^{opt}, however, individually excited coherence decreases as time elapses. Compared with optical coherence, spin coherence is much more robust, roughly ten times longer than the optical counterpart (Ham et al., 1997). Thus, if the optical coherence can be transferred into spin ensembles, longer storage time can be obtained (Moiseev & Kroll, 2001; Moiseev et al., 2003). In 1998, spin coherence excitation using temporally separated Raman optical pulses was investigated, where optical coherence between the optical pulses forming a Raman pulse plays a major role (Ham et al., 1998). The optical coherence in a time delayed Raman pulse is determined by inhomogeneous broadening of excited atoms. Contrary to general four-wave mixing processes, however, rephasing-based coherence transfer such as the stimulated photon echo is free from the optical coherence between control pulses. This will be discussed in more detail in Section 4.

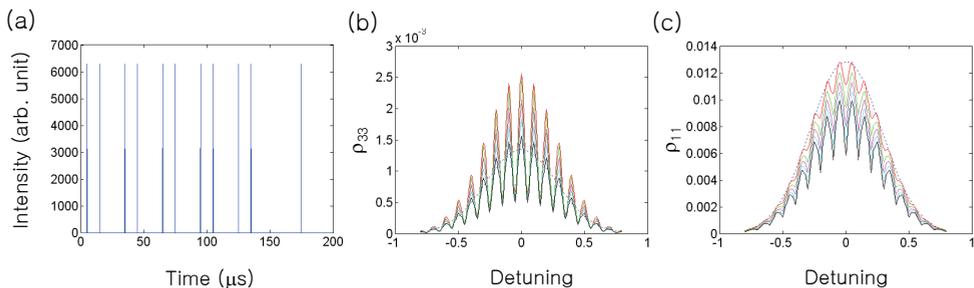## 3.3 To solve the spontaneous emission noise problem

Fig. 4. Numerical simulations of AFC using five sets of two consecutive pulses. (a) pulse sequence, (b) excited state population, and (c) ground state population. Dotted: after the first pulse; Red: after the second pulse; Green: after the fourth pulse; Magenta: after the sixth pulse; Cyan: after the eight pulse; Black: after the tenth pulse in (a)

The spontaneous emission noise originates in the excited atoms due to optical population decay. Especially for quantum memories, the data pulse D must be weak, where only a small number of atoms are excited. By the rephasing pulse R, however, population inversion results in potential spontaneous emission noise. To solve this problem, an atomic frequency comb (AFC) method was introduced by a Swiss group (de Riedmatten et al., 2008). In AFC, the excited atoms are freely removed by a spontaneous emission decay process during atom preparation by a long optical train composed of two consecutive weak pulse pairs, as shown in Fig. 4(a). By the way, in the stimulated photon echoes, two consecutive optical pulses D and W in Fig. 2(a) create a spectral grating on both ground and excited states. If a $\pi/2$ optical pulse set is used, then ideally the spectral grating forms a 50% duty cycle with an equal distribution of atoms [see Fig. 2(d)]. In AFC, many weak-pulse sets accumulate to form one spectral grating on top of another to sharpen it, so that the increased finesse can be obtained as shown in Fig. 4. At the same time the excited state atoms freely decay down to a third

state. Eventually no excited state population remains. Thus, a spontaneous emission-free optical system can be achieved. Regarding the spectral grating, the physics of AFC for the retrieval process is exactly the same as for the stimulated photon echoes as discussed (Ham, 2010b). In AFC, however, a trade-off exists between high finesse and optical depth regarding enhanced retrieval efficiency. Even though the spectral grating can last up to spin population decay time, the storage time in AFC is determined by optical phase decay time $T_2{}^{opt}$ (de Riedmatten et al., 2008), which is too short to quantum repeaters.

3.4 To solve the excited state population loss in stimulated photon echoes

This subsection somewhat overlaps with the modified two-pulse photon echoes in Section 3.2. In stimulated photon echoes, where longer storage time can be achieved, the excited atoms contain the same magnitude of coherence as the ground state. To avoid coherence loss due to optical population decay during the storage process, the excited atoms must be intentionally transferred into a third state for an on-demand halt, such as an auxiliary spin state. The coherence transfer technique suggested by the Lund group has been modified to transfer the spectrally modulated atoms from the excited state to the auxiliary spin state (Afzelius et al., 2010; Ham, 2009b). Because spectral grating is free from the optical phase decay process, storage time can be lengthened if population decay-caused coherence loss is halted. Unlike rephased atoms in the two-pulse photon echoes, the phase decay-dependent coherence loss can be frozen in the stimulated photon echoes using spectral grating, explaining why the coherence in AFC echoes (Afzelius et al., 2010) and phase locked echoes (Ham, 2009b) are degraded by spin dephasing. This will be discussed in more detail in Section 4.

4. Optical locking

As discussed in previous sections, a coherence transfer method is used to modify photon echoes to lengthen storage time. Here, an auxiliary deshelving pulse set (B1 and B2) is used to transfer atom population between the excited state ($|3>$) and an auxiliary spin state ($|2>$) as shown in Figs. 5(a) and (b). However, the atom population transfer between the optical and auxiliary spin states creates a $\pi/2$ phase gain, which applies evenly to all individual atoms assuming all light pulses are in phase. By a round-trip population transfer (by B1 and B2), the total phase gain accumulated becomes π. With the rephasing process serving to give a π phase shift to all individual atoms, this population transfer-based π phase gain completely washes out the rephasing performed by R leading to no echo generation. To avoid the odd phase gain obtained in the coherence transfer process, a phase recovery condition was investigated for an optical locking technique by an Inha group, S. Korea (Ham, 2009b). To create a multiple 2π phase shift during the coherence transfer process, the second deshelving pulse area must be 3π in order to make another round-trip population transfer for an additional π phase shift. As a general rule, the phase recovery condition of the optical deshelving pulses satisfies the followings (Ham, 2010a):

$$\Phi_{B1} = (4n-3)\pi, \tag{1}$$

$$\Phi_{B2} = (4n-1)\pi, \tag{2}$$

where n is an integer. Thus, the usage of an identical pulse set (Afzelius et al., 2010; Moiseev & Kroll, 2001) brings a contradiction, and violates the rephrasing process for echo

generation. The observation of delayed AFC echoes under this contradiction, however, can be explained as a result of coherence leakage due to imperfect population transfer in an optically dilute sample (Ham, 2010c; Ham, 2010d). The detected delayed echo signals in this case may be from the conventional photon echoes or at least mixed echoes. The remnant atom-generated echo signals from the excited state due to the imperfect population transfer cannot be separated from the delayed echoes (Ham, 2010c).
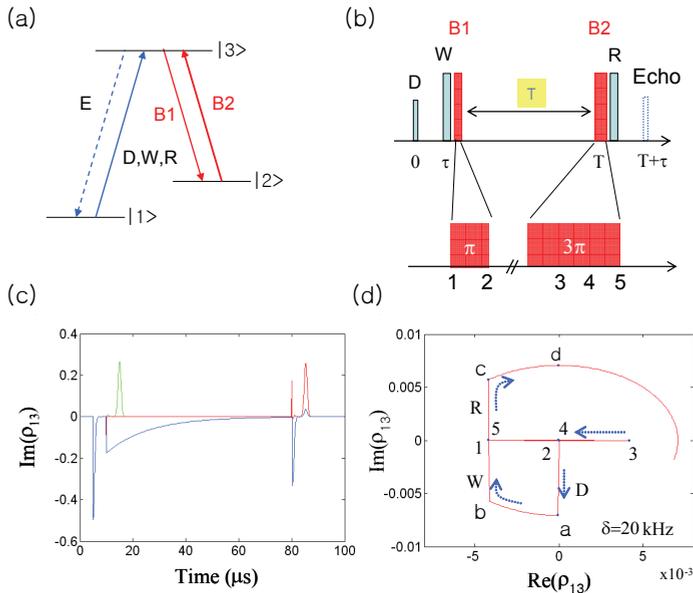
Fig. 5. Optical locking applied to stimulated photon echo. (a) An energy level diagram interacting with optical locking pulses, where D, W, and R represent DATA, WRITE, and READ pulses, respectively. (b) Optical pulse sequence for (a). (c) Numerical simulations. Red: for (b); Blue: without B1 and B2; Green: for two-pulse photon echo as a reference. (d) Bloch vector model without population decay loss. The numbers represent those in (b) indicating the phase recovery condition

In the experimental proofs of optical locking applied to both two-pulse photon echoes and stimulated photon echoes, the storage time extension of the photon echoes yields completely different results. First, in the two-pulse photon echoes, the storage time extension is governed by the overall spin dephasing rate, determined predominantly by spin inhomogeneous broadening, which is one tenth of the optical phase decay time (Afzelius et al., 2010; Ham, 2009b). As shown in the AFC and the phase locked echoes, this storage time extension is too short to solve the optical phase decay time constraint in conventional quantum memory protocols. With the stimulated photon echoes, however, the storage time extension is greatly increased due to the inherent property of optical phase locking resulting from the spectral grating based on population redistribution. The observed storage time extension, applying optical locking to the stimulated photon echoes in a rare-earth Pr^{3+} doped Y_2SiO_5 is five orders of magnitude longer than with the two-pulse photon echoes or AFC echoes (Ham, 2010d).

5. Phase matching in optical locking

As discussed in Section 3.1, a backward propagation technique has been introduced to solve the echo reabsorption problem. In two-pulse photon echoes using a rephasing halt, the phase matching condition for echo signal has nothing to do with the rephasing pulse vector, but relates with the data D and the optical locking pulses B1 and B2 (Ham, 2009b). Conversely, in the stimulated photon echoes in Section 3.4, the phase matching condition for echo signals includes the Data, Write, and Read pulses only, where optical locking pulses do not contribute at all to the phase matching condition (Ham, 2010d). From the results of these two cases, the important conclusion regarding the phase matching using optical locking is that the storage mechanism in each system is completely different. Thus, optical locking can result in an immense storage time extension in the stimulated photon echoes. For comparison, Fig. 6 represents a schematic of using optical locking pulses to these different cases. Only Fig. 6(b) can be applied to any meaningful storage time extension potential for quantum repeaters because of long photon storage time.
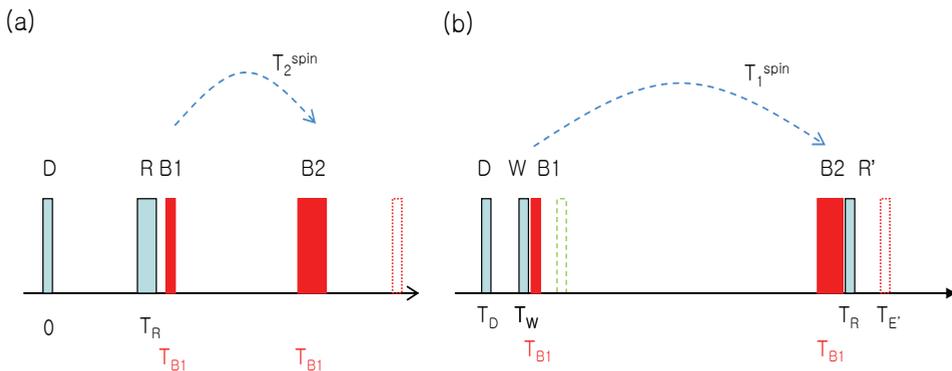

Fig. 6. Schematic diagram for (a) phase locked echo applied to rephased atoms in two-pulse photon echoes, and (b) optically locked echo applied to spectral grating in stimulated photon echoes. R and R' represent for rephasing (2π) and READ (π) pulses, respectively

The physics of photon storage time extension in Fig. 6(b) is atom phase locking. This is accomplished by the spectral grating discussed in Fig. 2. This means that the phase grating excited by D is fully transferred into population grating by W, so that phase dependent decoherence is completely locked. This optical population information is coherently transferred into an auxiliary spin state |2> by B1 as optical-spin coherence conversion process in Fig. 5. In this stage, the spin dephasing becomes also an independent parameter to the coherence. Then, the last-long spin coherence is returned into state |3> by B2, and the optical population information is fully recovered into the optical phase grating by R'. In the experiment, a Korean group demonstrated one second storage time of photon echoes with 50% retrievcal efficiency (Ham, 2010d). Multi-photon entangled light or squeezed light could be the best candidate to this method. However, a single photon data pulse scheme can also be applied because of extremely low noise by the spontaneous emission decay process for a wide bandwidth, pencil-like propagation geometry as discussed above.

6. Conclusion

For potential applications of long-distance quantum communications using quantum memories, modified photon echo protocols have been reviewed. Although AFC echoes and gradient echoes have successfully solved the intrinsically low retrieval efficiency and spontaneous emission noise problems in the original photon echoes, the ultrashort photon storage time limits the usage to quantum memory applications. Instead, optical locking applied to the stimulated photon echoes has been demonstrated to prove ultralong photon storage time limited by spin population decay time, which is much longer than the minimum required storage time for quantum repeaters. Unlike critical objection by Swiss and Calgary group, the intrinsic atom population-caused spontaneous emission noise problem in the conventional photon echoes, however, can not be a serious problem due to low noise to the echo signal in a wide-bandwidth scheme. The key idea of storage time extension is locking phase decay process to the storage mechanism as well as optical-spin coherence transfer.

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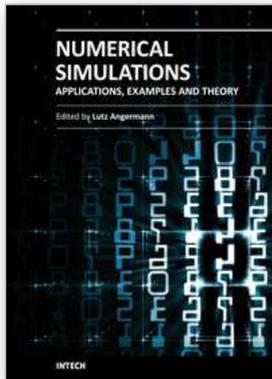
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