Quantum memory for nonstationary light fields based on controlled reversible inhomogeneous broadening

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We propose a method for efficient storage and recall of arbitrary nonstationary light fields, such as, for instance, single photon time-bin qubits or intense fields, in optically dense atomic ensembles. Our approach to quantum memory is based on controlled, reversible, inhomogeneous broadening and relies on a hidden time-reversal symmetry of the optical Bloch equations describing the propagation of the light field. We briefly discuss experimental realizations of our proposal.

Photons are good carriers to transmit quantum information, since they travel fast and only interact weakly with the environment [1]. However, their advantage is also their weakness: Photons can only be used in a probabilistic way for quantum-information processing where interaction between different carriers is necessary [2], and are hard to store. Therefore, many applications in quantum communication and information processing call for reversible and efficient mapping of nonclassical photon states onto electronic excitations in atoms or solids. Such quantum memories enable one to build sources of single photons on demand based on heralded single photon sources [3], serve as a buffer to store quantum information, and form a basic ingredient for both linear optic quantum computation [2] and quantum repeaters [4]. In particular, the possibility to store and recall single-photon time-bin qubits at a wavelength of around 1.5 μm would have a large impact on quantum communication, since they have been shown to be well suited for transmission in optical fibers over large distances [1,5].

Many different approaches towards quantum memory have been proposed, based on single absorbers or emitters in a cavity [6], as well as in optically dense atomic ensembles [7–9]. On the experimental side, storage and retrieval of classical light has been achieved using electromagnetically induced transparency (EIT) [10] and photon echoes [11]. All-optical storage of single photons and recall on pseudodemand has been achieved [12]. Finally, quantum coherent states of light have recently been stored onto an atomic ensemble with a higher-than-classical fidelity [13].

In this paper we present a method to efficiently store and recall nonstationary light fields based on controlled reversible inhomogeneous broadening (CRIB) in optically dense atomic ensembles. We demonstrate that by reversing the inhomogeneous broadening after absorption of an unknown, arbitrary incoming light state, one can force the atom-light system to evolve "backwards" in time. Our method shares several similarities with the ones proposed in Refs. [8,9,11], in that the pulse retrieval is obtained by a rephasing mechanism. However, we obtain this rephasing by controlling the artificial broadening of an unbroadened absorption line using external fields. This eliminates the difficulty associated with reversing natural, randomly distributed level shifts which

may not always be possible. Furthermore, the physical phenomenon we use for the storage and retrieval of the quantum state of light follows from a "hidden" symmetry of the optical Bloch equations describing the propagation and absorption of the light field in the atomic ensemble, and thus one does not have to solve those equations in order to see this effect. Our scheme enables, in principle, unit efficiency storage and recall of arbitrary light states, such as, for instance, single-photon time-bin qubits, or intense fields. Our protocol is general in the sense that it relates to any interaction that enables controlled, reversible, inhomogeneous broadening and applies to a variety of materials, e.g., rare-earth-metal-ion-doped solids, atomic vapor, nitrogen-vacancy (NV) centers, or quantum dots. Moreover, as compared to other schemes [10,13], ours has the advantage that the retrieved pulse is obtained when the control laser beams are off, and thus they do not mix with the signal photons.

In the following, we will describe mathematically the physical effect on which our scheme is based. We consider a series of atoms, confined in some region of space, interacting with a pulse of light propagating along the $+z$ direction, of central frequency ω_0, and a given polarization. Atom n at position z_n is initially prepared in a certain ground atomic level $|g\rangle_n$ and is driven by the laser pulse to some excited state $|e\rangle_n$ with corresponding transition frequency ω_n. The Hamiltonian describing this situation can be written as ($\hbar = 1$),

$$H = \sum_n \omega_n \sigma_n^z + \sum_k c|k|a_k^\dagger a_k - d\sum_n [\sigma_n^+ E^+(z_n) + \text{H.c.}]. \quad (1)$$

Here, the σ's are Pauli operators describing the atomic two-level transition, a_k is the annihilation operator of a field mode of momentum k, and $E^+(z) = i\Sigma_k g_k e^{ikz} a_k$ is the positive frequency part of the electric field operator. For the sake of simplicity, we have assumed that the dipole matrix element d corresponding to the two-level transitions is the same for all atoms. In Eq. (1) we have used a one-dimensional (1D) description; i.e., only included the field modes along the propagation direction. The rest of the modes are the ones responsible for spontaneous emission, which will be neglected here since we assume the total time of the whole considered pro-

cess to be much shorter than the spontaneous emission time for each atom. We further assume that in the incoming electromagnetic field only the modes with $\omega_k \in [\omega_0 - \delta, \omega_0 + \delta]$, with $\delta \ll \omega_0$ are occupied, a fact which has allowed us to perform the rotating wave approximation in H. Accordingly, we define the slowly varying electric field operators corresponding to the forward and backward modes as

$$\epsilon_f^+(z,t) := i \sum_{k>0} g_k a_k(t) e^{ikz} e^{i(\omega_0 t - k_0 z)}, \quad (2)$$

$$\epsilon_b^+(z,t) := i \sum_{k<0} g_k a_k(t) e^{ikz} e^{i(\omega_0 t + k_0 z)}, \quad (3)$$

and $\epsilon_{f,b}^-(z,t) := \epsilon_{f,b}^+(z,t)^\dagger$, where $k_0 = \omega_0/c$.

Let us first consider the propagation equation of the light pulse, which is moving from left to right, through the atomic medium. We define new atomic operators as

$$s_{n,f}^-(t) := \sigma_n^-(t) e^{i(\omega_0 t - k_0 z_n)}, \quad (4a)$$

$$s_{n,f}^+(t) := [s_{n,f}^-(t)]^\dagger, \quad s_{n,f}^z(t) := \sigma_n^z(t), \quad (4b)$$

and obtain the following Maxwell-Bloch equations [14]:

$$i\partial_t s_{n,f}^-(t) = -\Delta_n s_{n,f}^-(t) + d s_{n,f}^z(t) \epsilon_f^+(z_n,t),$$

$$i\partial_t s_{n,f}^z(t) = -d s_{n,f}^+(t) \epsilon_f^+(z_n,t) + \text{H.c.},$$

$$\left[\partial_z + \frac{1}{c}\partial_t\right]\epsilon_f^+(z,t) = q \sum_{k>0} g_k^2 \sum_n s_{n,f}^-(t) e^{i(k-k_0)(z-z_n)}, \quad (5)$$

where $\Delta_n := \omega_0 - \omega_n$ is the detuning and $q = id/c$. In the last equation, the sum over k gives rise to a $\delta(z-z_n)$, which indicates that the atoms are the sources (or drains) of the electric field. Equations (5) describe, in the Heisenberg picture, how the photons in the incoming pulse are absorbed as they travel through the medium. In particular, if the medium is optically thick, after a sufficiently long time t_0 the photons will be absorbed. In the Heisenberg picture, this is manifested by the fact that all expectation values of normally order field operators will vanish. In the Schrödinger picture, the state of the atom+light system factorizes

$$|\Psi(t_0)\rangle \simeq |\phi(t_0)\rangle_{\text{atoms}} \otimes |\text{vac}\rangle_{\text{field}}. \quad (6)$$

Thus the quantum state of light is stored in the atomic state. The main issue is now to find a way to recover the state, i.e., to map it back to the field state. We propose to use the photon-echo effect, i.e., to instantaneously change some of the atomic properties such that the field is restored. In order to show how this works, we just have to look at some symmetry properties of those equations, and thus we do not need to solve the complicated Maxwell-Bloch equations nor to make further approximations. The main idea is to carry out those instantaneous changes so that the atomic and field operators evolve "backwards in time," i.e., at the end all the atoms end up in their ground states and the field is restored but now propagating from right to left.

At time t_0, the state of the atoms and field is $|\Psi(t_0)\rangle$. In the spirit of the proposals in Refs. [8,9], we suddenly change

the atomic frequency of each atom from $\omega_n = \omega_0 + \Delta_n$ to $\omega_n = \omega_0 - \Delta_n$ (i.e., we change the sign of the detuning). Simultaneously, a phase shift is applied to each atom that changes $|g\rangle_n \rightarrow e^{2ik_0 z_n}|g\rangle_n$ [15]. Since we are going to use the Heisenberg picture for subsequent times, this is equivalent to keeping the initial state as $|\Psi(t_0)\rangle$ and replacing

$$\sigma_-(t_0) \rightarrow \sigma_-(t_0) e^{2ik_0 z_n}. \quad (7)$$

Defining the new Heisenberg operators,

$$s_{n,b}^-(t) := \sigma_n^-(t) e^{i(\omega_0 t + k_0 z_n)}, \quad (8a)$$

$$s_{n,b}^+(t) := [s_{n,b}^-(t)]^\dagger, \quad s_{n,b}^z(t) := \sigma_n^z(t), \quad (8b)$$

one can easily obtain the new Maxwell-Bloch equations. Taking into account that

$$\sum_{k>0} g_k^2 e^{i(k-k_0)(z-z_n)} \simeq \sum_{k<0} g_k^2 e^{i(k+k_0)(z-z_n)}, \quad (9)$$

since, as mentioned before, this expression is basically proportional to $\delta(z-z_n)$ (i.e., real), we have that the operators $s_{n,b}^-(t_0+\tau)$, $s_{n,b}^z(t_0+\tau)$, and $-\epsilon_b^+(z,t_0+\tau)$ fulfill the same (first order) differential equations as $s_{n,f}^-(t_0-\tau)$, $s_{n,f}^z(t_0-\tau)$ and $\epsilon_f^+(z,t_0-\tau)$, respectively [16]. Using (4) and (8) and taking into account (7) one can verify that $s_{n,b}^{\pm,z}(t_0) = s_{n,f}^{\pm,z}(t_0)$. Thus, $s_{n,b}^{\pm,z}(t_0+\tau) = s_{n,f}^{\pm,z}(t_0-\tau)$, i.e., the atomic operators evolve backwards in time. It is a bit more involved to prove that the same applies for the field operators.

Obviously it is not possible to show that the operators describing the backward modes evolve time reversed to the operators describing the forward modes. However, we only need to show that in the Heisenberg picture, the expectation value of any observable, measured at time $t_0+\tau$ is the same, independent of whether we use the operators $s_{n,f}^{\pm,z}$, ϵ_f^+ at time $t_0-\tau$ or the operators $s_{n,b}^{\pm,z}$, $-\epsilon_b^+$ at time $t_0+\tau$. This implies the desired result, namely that the evolution of the system at time $t_0+\tau$ will be closely connected to that at $t_0-\tau$. In particular, at time $2t_0$ we will recover the initial pulse propagating in the backward direction and with a global π phase shift.

In order to show that, we consider a solution for any operator describing the forward modes in terms of the operators at time $\tau=0$; for example,

$$\epsilon_f^+(z,t_0-\tau) = \tilde{f}(\tau, s_{n,f}^{\pm,z}(t_0), \epsilon_f^\pm(z,t_0)). \quad (10)$$

We will then have

$$\epsilon_b^+(z,t_0+\tau) = -\tilde{f}(\tau, s_{n,b}^{\pm,z}(t_0), -\epsilon_b^\pm(z,t_0)). \quad (11)$$

As explained above we have to show that

$$\langle\Psi(t_0)|f(\tau, s_{n,b}^{\pm,z}(t_0), \epsilon_f^\pm(z,t_0))|\Psi(t_0)\rangle$$
$$= \langle\Psi(t_0)|f(\tau, s_{n,f}^{\pm,z}(t_0), -\epsilon_b^\pm(z,t_0))|\Psi(t_0)\rangle \quad (12)$$

for all (analytic) functions f, and therefore that the expectation values of any observable at time τ will give the same result. Equation (12) shows that any observable measured at time $t_0+\tau$ can be described either with $f(\tau, s_{n,f}^{\pm,z}(t_0), \epsilon_f(z,t_0))$ or $f(\tau, s_{n,b}^{\pm,z}(t_0), -\epsilon_b(z,t_0))$. In order to prove Eq. (12) we use

that $s_{n,b}^{\pm,z}(t_0)=s_{n,f}^{\pm,z}(t_0)$. On the other hand, if we expand f in the left-hand side of Eq. (12) in normally ordered powers of $\epsilon_f^{\pm}(z,t_0)$ all the terms except the zeroth power will vanish as they are evaluated in the vacuum $|\text{vac}\rangle$ [cf. (6)]. These zeroth order terms appear when we commute $\epsilon_f^-(z,t_0)$ with $\epsilon_f^+(z',t_0)$. Analogously, in the right-hand side of Eq. (12) only the zeroth order terms will survive, and they will appear when we commute $\epsilon_b^-(z,t_0)$ with $\epsilon_b^+(z',t_0)$. However, using their definitions one can readily see that

$$\left[\epsilon_f^-(z,t_0),\epsilon_f^+(z',t_0)\right] \simeq \left[\epsilon_b^-(z,t_0),\epsilon_b^+(z',t_0)\right] \qquad (13)$$

as it follows from (9). Note also that this explanation becomes much simpler in the semiclassical case, where the electric field operators are substituted by c numbers; in that case, $\epsilon_f^+(z,t_0-\varepsilon)=-\epsilon_b^+(z,t_0+\varepsilon)\to 0$ as $\varepsilon\to 0$, since at time t_0 the field has been completely absorbed, and thus $\epsilon_b^+(z,t_0+\tau)=-\epsilon_f^+(z,t_0-\tau)$.

Thus, according to this analysis, one should proceed as follows: First, one should prepare a single atomic absorption line and then broaden it inhomogenously up to a value of δ. This has to be carried out in a controlled way such that later on one can change the sign of the detunings of all the atoms. Second, the light pulse of duration δt_{light} is sent into the optically thick atomic ensemble and is completely absorbed [17]. In order to retrieve the pulse the position dependent phase shift is imprinted in all atoms, and the sign of the detunings are suddenly changed during a time δt. The whole process should happen within a time, much shorter than the atomic spontaneous emission time or any other decoherence time. Thus, the conditions for this scheme to work are $\delta t \ll \delta t_{light}$, $t_0 \ll T_{decoh}$. In order to extend the storage time beyond T_{decoh}, one might proceed as follows. After the absorption the field introducing the inhomogeneous broadening is switched off. Then, one can coherently transfer the excited state population to another internal atomic ground state with extended coherence time. When readout is desired, a counterpropagating beam is used to inverse this process and the field introducing the inhomogeneous broadening is reestablished with opposite sign. It has been shown in Ref. [18] that this process automatically leads to the position-dependent phase shift.

In the following, we will briefly discuss several experimental realizations of our proposal. As mentioned before, CRIB requires an atomic medium with a long decoherence time, i.e., a small homogeneous linewidth. For instance, alkaline metals with typical linewidths in the MHz range enable storage during a few hundred nanoseconds, and linewidths in rare-earth-ion-doped (RE) crystals at cryogenic temperatures may be below 100 Hz, i.e., storage times of several milliseconds have been reported [19,20].

First, an isolated absorption line on a nonabsorbing background has to be prepared. To this aim, the natural inhomogeneous broadening has to be suppressed. Regarding free atoms, the Doppler broadening can be decreased by cooling. In the case of rare-earth-metal-ion-doped crystals, inhomogeneous broadening is caused by the fact that the ions are located in slightly different surroundings in the host. Through optical pumping, it is possible to transfer ions absorbing at undesired frequencies to other long-lived (hyperfine) levels, and thus to empty a spectral region of absorbing ions and form the desired narrow absorptive feature [21].

Next, the absorption line has to be broadened in a controlled and reversible way. After the absorption of the light field, the broadening can be suddenly reduced to zero and the phase shift of $2kz$ applied. When recall is required the broadening is reestablished in the inverse way, leading to the desired rephasing. Finally, let us briefly comment on possibilities of implementing the controlled reversible broadening. One can take advantage of the interaction between atoms and a magnetic or an electric field, i.e., the Zeeman or Stark shift. Note that the maximum broadening has to remain smaller than the hyperfine or fine structure splitting, respectively. Regarding the Zeeman effect, typical level shifts are of the order of the Bohr magneton divided by Planck's constant, i.e., ≈ 13 MHz/mT. When using atoms (ions) featuring a permanent dipole moment, one might also take advantage of a dc electric field. For instance, in the case of rare-earth-metal-ion-doped crystals, shifts of the order of 100 kHz/V cm^{-1} have been observed [22]. Application of a magnetic, or electric (dc) field gradient leads thus to a desired position-dependent detuning, Δ, that can be reversed by inverting the field [23]. More details regarding CRIB based on dc Stark shifts in rare-earth-metal-ion-doped crystals can be found in Ref. [18]. To complete these examples, the energy levels of neutral atoms (lacking a permanent dipole moment) can be shifted in a controlled way employing ac electric fields, e.g., strongly detuned light fields (ac Stark or light shift). In this case, the shift depends linearly on the light intensity and is inversely proportional to the detuning of the light field with respect to the unperturbed absorption line. Typical line shifts, for instance for the cesium D1 line, are of the order of 200 MHz/10^9 W m^{-2} at a laser detuning of 10 nm. The shifts can be reversed by changing the detuning of the laser to its inverse value.

In this paper, we proposed a protocol for quantum state storage for nonstationary light fields in an inhomogeneously broadened atomic ensemble. Our proposal is not limited to single photon fields, but also enables storage and recall of intense pulses. Instead of solving the underlying equations of motion, we showed that if one applies suitable phase and frequency shifts once the light field has been absorbed, the atom-light system will evolve backwards in time. In order to implement these shifts, the inhomogeneous broadening must be controllable and reversible. We briefly discussed some experimental realizations of CRIB. Several experimental groups, including ourselves, are working on the realization of the proposed scheme [18,24]. To conclude, let us mention that it is possible to acquire information about successful loading of the memory by employing teleportation-based state transfer, along the lines exploited in quantum relays [1,25]. This is an important necessity to implement a quantum repeater, and also enables to deterministically entangle distant atomic ensembles for quantum networks by starting with entangled photon pairs. Note also that CRIB enables storage of a whole sequence of light states and recall in inverse order. In addition, using several broadened, well-separated absorption lines, frequency multiplexing can be employed. This enables recall of photonic qubits in arbitrary

order by triggering only rephasing of atoms within a desired range of resonance frequencies. Finally, it is likely that tailored rephasing can also be used to implement general "interatomic" unitary transformations [26] or measurements, e.g., pulse compression [27].

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[14] In these equations we have omitted the backward field $\epsilon_b(z,t)$; i.e., we have ignored backscattering, which can be shown to be negligible.

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[16] The forward terms $-\epsilon_f^+(z,t_0+t)$ can be neglected using the same arguments that allowed us to neglect the backward scattering terms (cf. [14]) in Eq. (5).

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