

Complete Reconstruction of the Quantum State of a Single-Photon Wave Packet Absorbed by a Doppler-Broadened Transition

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An idea for how to reconstruct the quantum state of a nonstationary single-photon wave packet absorbed in a macroscopic medium with inhomogeneously broadened lines is presented. An analytical treatment of the problem is performed and the requirements on the proposed scheme for complete recovery of the recorded nonstationary quantum state with a probability close to unity is described. The physical nature of the present scheme is also discussed.

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Quantum optics opens new possibilities for construction of specific quantum states and for their use both in fundamental research and in numerous applications. Growing interest in quantum computation and the strong requirements that must be fulfilled to realize quantum calculations in practice have spurred an interest in many new issues related to the quantum dynamics of matter and light. A basic problem is the development of appropriate techniques for storage and recall of arbitrary quantum states of light. Recently a new technique for storage and recall of quantum states of light within a narrow spectral interval, based on electromagnetically induced transparency (EIT) [1], was proposed [2]. Mapping the quantum state of single-mode fields has also been attempted using trapped ions [3], whereas for the case of free space the common scheme of Raman-type interaction was also recently proposed for the mapping in [4]. It is interesting to consider techniques which would give a more general solution to quantum state storage and recall including a wider range of nonstationary fields. This is the problem we consider here. Our approach stems from the general ability of photon echoes techniques [5] to store and reproduce arbitrary, temporally shaped wave packets [6]. However, the efficiency in the photon echo process normally is low. For example, if the sample is transparent most of the input light would go through without absorption, while if, on the other hand, the optical density is high, the incoming wave packet will be completely absorbed by (mapped onto) the atomic ensemble, but most of the recalled wave packet will then also be absorbed as it propagates through the material. Here we propose an alternative photon echo storage procedure which solves both the above problems and which can be used for quantum state storage and total recall of nonstationary light fields. The proposed technique for restoring the field is based on the property that frequency shifts due to the Doppler effect are opposite for counterpropagating directions. We present the analytical solution for quantum state storage and recall of nonstationary single-photon wave packets in this medium.

We consider a gas localized between coordinates $z = 0$ and $z = L$. The gas consists of N three-level atoms all initially in the ground state, $|1\rangle$. There is an allowed electric dipole transition to the state $|3\rangle$ and there is a metastable state $|2\rangle$ situated just above state $|1\rangle$ in energy. We will neglect relaxation processes in the atomic system. A single-photon wave packet, resonant with the inhomogeneously broadened $|1\rangle$ - $|3\rangle$ transition, is incident along the z direction and absorbed in the medium. After a small time delay, τ , we apply a laser pulse in the z direction with pulse area π that transfers any excitation to state $|2\rangle$ using the electric dipole allowed transition $|2\rangle$ - $|3\rangle$. Information about the photon can now be saved for a longer time since the relaxation processes in level 2 are slow.

Time T after the first pulse we apply a second laser pulse also tuned to the $|2\rangle$ - $|3\rangle$ transition. This pulse also has pulse area π but propagates in negative z direction. This pulse will now revert the atomic excitation from state $|2\rangle$ to state $|3\rangle$. A rephasing of atomic coherence will now occur in the negative z direction a time interval τ after the second laser pulse (i.e., time interval $2\tau + T$ after the single-photon wave packet). Several different photon echo processes in three-level systems of similar type have been investigated and experimentally demonstrated [7]. However, the novel feature in our scheme is that here the rephasing process reproduces an exact time-reversed counterpropagating replica of the input single-photon wave packet; moreover, our scheme creates a rephasing process where the absorption process is replayed in a time-reversed mode, forcing the material to reemit what it previously absorbed. (Related considerations have been made previously in connection to schemes for quantum information transfer [8].) So the efficiency in the reemission process can be very high and the quantum state of the photon can be reproduced with a probability close to unity. Below we give the theory of the proposed process.

Before interaction of the photon with the medium the state vector $|\psi_f(t \rightarrow -\infty)\rangle$ of the combined system is the direct product of the field state and the atomic state:

$$|\psi_f(t \rightarrow -\infty)\rangle = \int dk f_k(-\infty) a_k^+ |0\rangle |A(t)\rangle. \quad (1)$$

Here $f_k(t)$ is the photon wave function with initial condition $\int dk |f_k(-\infty)|^2 = 1$ and a sharp maximum at $\vec{k} = \vec{e}_z \omega_{31}/c$. $|0\rangle$ is the vacuum state of light, the field operators satisfy the commutation relations $[a_{k'}, a_k^+] = \delta(k' - k)$. $|A(t)\rangle = \prod_{j=1}^N |\phi(t)\rangle_j$ is the atomic ground state, $|\phi(t)\rangle_j = \delta^{1/2}[r_j - r_j^0(t)] |1\rangle_j$, $|1\rangle_j$ is the ground state of atom j , the function $\delta^{1/2}[r_j - r_j^0(t)]$ describes the spatial movement of the j th atom with its coordinates at $t = 0$, $\vec{r}_j(t = 0) = \vec{r}_j^0(z_j^0, r_\perp^j(0))$, where the z coordinate $z_j^0(t) = z_j^0 + v_z^j t$, \vec{v}^j is the velocity of the j th atom. Below we treat the problem in one dimension with the function $f_k(-\infty) = f^{\text{in}}((\omega_k - \omega_{31})/c)$, where $k = \omega_k/c$, and we introduce the spectral width $\delta\omega_{\text{ph}}$ of the function with $\delta t_{\text{ph}} = \delta\omega_{\text{ph}}^{-1}$ as the temporal duration of the photon wave-packet state. The dynamics will be described by the Hamiltonian:

$$\begin{aligned} H &= H_a + H_f + V, \\ H_a &= \hbar\omega_{31} \sum_{j=1}^N P_{33}^j + \hbar\Delta \sum_{j=1}^N P_{22}^j, \\ H_f &= \int dk \hbar\omega_k a_k^+ a_k, \\ V &= \hbar g \int dk \sum_{j=1}^N \{a_k P_{31}^j \exp[ikz_j(t)] + \text{H.c.}\}. \end{aligned} \quad (2)$$

Here $P_{\mu\nu}^j = |\mu\rangle_{jj}\langle\nu|$ are the transition atomic operators between the states $|\nu\rangle_j$ and $|\mu\rangle_j$; g is the coupling constant of the photons with atoms on the transition $|1\rangle\text{-}|3\rangle$. We neglect the small frequency dependence [$g(k) \cong g(\omega_{31}/c) \equiv g$]. Initially we consider the mapping of the quantum state onto the atomic ensemble. The photon enters the medium at time $t = 0$. We have the following general form of the wave function:

$$\begin{aligned} |\psi(t)\rangle &= |\psi_m(t)\rangle + |\psi_f(t)\rangle, \\ |\psi_f(t)\rangle &= \int dk f_k(t) a_k^+ |0\rangle |A(t)\rangle, \\ |\psi_m(t)\rangle &= \sum_{j=1}^N b_j(t) P_{31}^j |0\rangle |A(t)\rangle, \end{aligned} \quad (3)$$

which is easily determined from the initial condition (1) and the interaction V , which conserves the total number of the excitations in the field plus atoms. Substituting Eq. (3) into the Schrödinger equation and taking into account that the wave vector k is parallel to the z direction we obtain

$$\begin{aligned} \frac{\partial}{\partial t} f_k &= -i\omega_k f_k - ig \sum_{j=1}^N b_j \\ &\quad \times \exp\{-ikz_j^0 - i(v_z^j/c)\omega_{31}t\}, \\ \frac{\partial}{\partial t} b_j &= -i\omega_{31} b_j - ig \int dk f_k \\ &\quad \times \exp\{-ikz_j^0 - i(v_z^j/c)\omega_{31}t\}. \end{aligned} \quad (4)$$

Equations (4) and (5) are valid assuming $\exp\{i(k - \omega_{31}/c)v_z^j t\} \cong 1$. This condition can be restated as $t < \omega_{31}/(\delta\omega_{\text{ph}}^2)$. Assuming $\delta\omega_{\text{ph}} \approx 10^9 \text{ sec}^{-1}$ and $\omega_{31} \approx 10^{15} \text{ sec}^{-1}$ we obtain $t < 1 \text{ ms}$. With the new variables $\beta_j = b_j(t) \exp\{-i(v_z^j/c)\omega_{31}t\}$, we obtain

$$\frac{\partial}{\partial t} f_k = -i\omega_k f_k - ig \sum_{j=1}^N \beta_j \exp\{-ikz_j^0\}, \quad (6)$$

$$\frac{\partial}{\partial t} \beta_j = -i\omega_{31}^{j(+)} \beta_j - ig \int dk f_k \exp\{-ikz_j^0\}, \quad (7)$$

where $\omega_{mn}^{j(+)} \equiv \omega_{mn}^j = \omega_{mn}(1 + v_z^j/c)$. Let $\delta\omega_{\text{ph}}$ be much narrower than the inhomogeneous broadening, Δ_n , of the atomic transition $|1\rangle\text{-}|3\rangle$ ($\delta\omega_{\text{ph}} \ll \Delta_n$). The result will then be independent of the type of line profile for the inhomogeneous broadening. For simplicity the Doppler broadening is therefore represented by a Lorentzian profile. Also for each value of z we sum over all atoms localized within the beam cross section [using notation $\beta_j(t) = \beta_j(\omega_{31}^j; t, z_j^0)$]:

$$\begin{aligned} \sum_{j=1}^N \beta_j(t) \delta(z - z_j^0) \dots &= n_0 \int_{-\infty}^{\infty} d(\omega_{31}^j - \omega_{31}) \\ &\quad \times \beta_j(\omega_{31}^j; t, z) \\ &\quad \times \frac{\Delta_n}{\pi[\Delta_n^2 + (\omega_{31}^j - \omega_{31})^2]} \dots \end{aligned} \quad (8)$$

Here $n_0 = N/L$ is the number of atoms per unit length. After multiplying Eq. (6) by e^{ikz} , integrating over k , and then using the notation $\int dk f_k(t) e^{ikz} = E(t, z) \times \exp[i\omega_{31}(z/c - t)]$, $\int dk e^{ik(z - z_j^0)} = 2\pi \delta(z - z_j^0)$ and Eqs. (7) and (8), we get

$$\left(\frac{\partial}{\partial t} + \frac{\partial}{\partial z}\right) E(t, z) = -\alpha E(t, z), \quad (9)$$

$$E(t, z) = E_0(t - z/c) \exp(-\alpha z), \quad (10)$$

$$E_0(t) = \int dk f^{\text{in}}((\omega_k - \omega_{31})/c) \exp\{-i(\omega_k - \omega_{31})t\}, \quad (11)$$

here $\alpha = 2\pi g^2 n_0 / (c\Delta_n)$, 2α corresponds to the absorption coefficient on the $|1\rangle \rightarrow |3\rangle$ transition. Using Eqs. (11) and (7) we find $\beta_j(t)$ for $t > \delta t_{\text{ph}}$:

$$\begin{aligned} \beta_j(t)|_{t > \delta t_{\text{ph}}} &\rightarrow \beta(\omega_{31}^j, z_j^0) e^{-i\omega_{31}^j t}, \\ \beta(\omega_{31}^j; z_j^0) &= \beta_0(\omega_{31}^j; z_j^0) e^{i\omega_{31}^j z_j^0 / c}, \\ \beta_0(\omega_{31}^j; z_j^0) &= -i2\pi g \exp(-\alpha z_j^0) \\ &\quad \times [f^{\text{in}}((\omega_{31}^{j(+)} - \omega_{31})/c)]/c. \end{aligned} \quad (12)$$

We note that $\beta(\omega_{31}^j, z_j^0)$ corresponds to the coefficients for the wave function $|\psi^i(t)\rangle$ in the interaction picture. The solution (12) reflects the fundamental principle that any atom absorbs only the resonant component of the field spectrum. Using (12) and (8) in the interaction picture we find that $\lim_{\substack{\alpha L \rightarrow \infty \\ \delta t_{\text{ph}} \gg 1}} \sum_{j=1}^N |\beta_j(t)|^2 \rightarrow 1$ and

$$|\psi^i(t)\rangle_{\substack{t \gg \delta t_{\text{ph}} \\ \alpha L \gg 1}} = \sum_{j=1}^N \beta_0(\omega_{31}^j; z_j^0) e^{i\omega_{31}^j z_j^0 / c} P_{31}^j |0\rangle |A(t)\rangle. \quad (13)$$

To store the single-photon quantum state for a longer time we apply the classical laser pulse $E_1(t, z) = E_{01}(t - \tau - z/c) \cos\{\omega_{32}(t - \tau - z/c) + \varphi_1\}$, after time delay $t_1 = \tau \ll T_2^{(3)}$ ($T_2^{(3)}$ is the phase relaxation time of state |3>). Since level |2) is unpopulated the laser pulse amplitude will be constant throughout the medium. Let the temporal duration of the laser pulse δt_1 be very short so that we can neglect the atomic dephasing ($\delta t_1 \delta \omega_{\text{ph}} \ll 1$). The pulse area is set to π ($\vartheta_1 = dE_{01} \hbar^{-1} \delta t_1 = \pi$). Here the atomic evolution is described by the unitary operator $U_1(\vartheta_1 = \pi, k_1, \varphi_1)$ (see, for example, Ref. [9]) which in the interaction picture can be written as $U_1(\tau + \delta t_1, \tau; \vartheta_1, k_1, \varphi_1) \equiv U_1(\vartheta_1, k_1, \varphi_1) = \prod_{j=1}^N U^j(\vartheta_1, \gamma_j^1)$, $U^j(\vartheta_1, \gamma_j^1) = P_{11}^j + \cos(\vartheta_1/2)(P_{22}^j + P_{33}^j) + i \sin(\vartheta_1/2)\{P_{23}^j e^{-i\gamma_j^1} + \text{H.c.}\}$,

where the phase $\gamma_j^1 = \omega_{32}^{j(+)}(\tau + z_j^0/c) + \varphi_1$. After this laser pulse we have the state

$$\begin{aligned} |\psi_m^i(\tau + \delta t_1)\rangle &= U_1(\vartheta_1 = \pi, k_1, \varphi_1) |\psi_m^i(\tau)\rangle \\ &= i \sum_{j=1}^N \beta_0(\omega_{31}^j; z_j^0) e^{-i(\omega_{32}^j \tau + \xi_j^1)} \\ &\quad \times P_{21}^j |0\rangle |A(r)\rangle. \end{aligned} \quad (14)$$

Here $\xi_j^1 = \varphi_1 - \Delta z_j^0/c$. The state $|\psi_m^i(\tau + \delta t_1)\rangle$ saves all information about the photon for a long time, $T_2^{(2)}$.

To reconstruct the single-photon state the gas is now excited by a second short laser pulse at time $t_2 = T + \tau$ ($T < T_2^{(2)}$, $\delta t_2 \delta \omega_{\text{ph}} \ll 1$) propagating in the negative z direction [$k_2 = -k_1$, so $\vec{k}_2 = -(\omega_{32}/c)\vec{e}_z$]. Similar to the first laser pulse, we have $E_2(t, z) = E_{02}(t - T - \tau + z/c) \cos\{\omega_{32}(t - T - \tau + z/c) + \varphi_2\}$. In this case the unitary evolution operator $U_2(\vartheta_2 = \pi, k_2, \varphi_2)$ will be $U_2(\vartheta_2, k_2, \varphi_2) = \prod_{j=1}^N U^j(\vartheta_2, \gamma_j^2)$, where $\gamma_j^2 = \omega_{32}^{j(-)}(T + \tau - z_j^0) + \varphi_2$, $\omega_{mn}^{j(-)} = \omega_{mn}(1 - \nu_z^j/c)$. Thus after the second laser pulse excitation we have the following wave function:

$$|\psi_m^i(T + \tau + \delta t_2)\rangle = U_2(\vartheta_2 = \pi, k_2, \varphi_2) |\psi_m^i(\tau + \delta t_1)\rangle = i^2 \sum_{j=1}^N \beta_0(\omega_{32}^j; z_j^0) \exp\{i\chi_j\} P_{31}^j |0\rangle |A(\tau + T)\rangle. \quad (15)$$

Here the phase $\chi_j = \omega_{32}^{j(-)}(T + \tau - z_j^0/c) - \omega_{32}^{j(+)}\tau + \Delta z_j^0/c + \varphi_2 - \varphi_1$. It is important to compare the state after the mapping, Eq. (13), with the state after the recall pulse, Eq. (15). We see that the changes in the state (15) are determined by the phase factor $e^{i\chi_j}$. After the transfer of the quantum state to level 3 we consider the interaction of the medium with the quantum electromagnetic field. We will investigate the radiation emitted in direction $\vec{k} = -k\vec{e}_z$. Using substitution $\eta_j = b_j \exp\{i(\nu_z^j/c)\omega_{31}^j t\}$ into the basic equations we obtain

$$\frac{\partial}{\partial t} f_k = -i\omega_k f_k - ig \sum_{j=1}^N \eta_j \exp\{ikz_j^0\}, \quad (16)$$

$$\frac{\partial}{\partial t} \eta_j = -i\omega_{31}^{j(-)} \eta_j - ig \int dk f_k \exp\{-ikz_j^0\}. \quad (17)$$

The parameters $b_j, \beta_j, \eta_j, f_k$ correspond to the Schrödinger representation so we have the initial conditions for Eqs. (16) and (17) at $t = T + \tau + \delta t_2$:

$$\begin{aligned} f_k(T + \tau + \delta t_2) &= 0, \\ \eta_j(T + \tau + \delta t_2) &= -\beta_0(\omega_{31}^j; z_j^0) e^{i\chi_j} \\ &\quad \times \exp\{-i\omega_{31}^{j(-)}(T + \tau + \delta t_2)\}. \end{aligned} \quad (18)$$

Below we again use the notation $\int dk f_k(t) e^{-ikz} = F(t, z) \exp[-i\omega_{31}(z/c + t)]$. The formal solution for $\eta_j(t)$ with the initial condition (18) is

$$\begin{aligned} \eta_j(t) &= -\beta_0(\omega_{31}^j; z_j^0) e^{-i\omega_{31}^{j(-)} t} - ig \int_{t_2 + \delta t_2}^t dt' \\ &\quad \times e^{-i\omega_{31}^{j(-)}(t-t')} F(t', z_j^0) \exp[-i\omega_{31}(z_j^0/c + t')]. \end{aligned} \quad (19)$$

Using (8), (18), (19) in (16) we obtain

$$\begin{aligned} \left(\frac{\partial}{\partial t} - \frac{\partial}{\partial z}\right) F(t, z) &= -\alpha F(t, z) + \exp[i\omega_{31}(z/c + t)] \\ &\quad \times P(t, z), \end{aligned} \quad (20)$$

where, using Eqs. (13) and (8) for $0 < z < L$,

$$\begin{aligned} P(t, z) &= \frac{2\pi ig}{c} \sum_{j=1}^N \beta_0(\omega_{31}^{j(+)}; z_j^0) \delta(z - z_j^0) \\ &\quad \times \exp\{-i(\omega_{31}^{j(-)} t - \chi_j)\} \Big|_{\exp\{2i(\Delta z_j^0/c)\} \cong 1} \\ &= 2i\alpha e^{-\alpha z} E_0(T + 2\tau - t - z/c) e^{i\gamma} \\ &\quad \times \exp\{-i\omega_{31}(z/c + t)\}. \end{aligned} \quad (21)$$

Here $\gamma = (\omega_{31} - \Delta)T + \varphi_{21}$. Since the absorption of the weak field occurs within a distance $z \approx \alpha^{-1}$, the process must be phase matched over distance α^{-1} , so we require that $\exp\{2i\Delta/(\alpha c)\} \cong 1$ and consequently $\Delta < c\alpha/2$. Thus we see that the value $P(t, z)$ is a source of radiation connected with the rephasing of atomic coherence and this radiation is the photon echo signal. Using (21) we have the following solution of (20):

$$F(t, z) = e^{-\alpha z} E_0(T + 2\tau - t - z/c) e^{i(\gamma + \pi/2)}. \quad (22)$$

We see that the output field will be radiated in the back direction from $z = 0$ at time $t = T + 2\tau$. Its amplitude exactly copies the field of the incident photon. But the temporal profile is reversed as would be the case also for a conventional photon echo process. Using Eqs. (22) and (19) we find that for $t > T + 2\tau + \delta t_{\text{ph}}$ the atomic parameters $\eta_j(t)|_{t \gg T + 2\tau + \delta t_{\text{ph}}} \rightarrow 0$. This means that all atoms have returned to the ground state. Using (22) and the inverse frequency domain Fourier transform, we finally

obtain the state of the output wave function which in the interaction picture has the form

$$\begin{aligned} \lim_{t \gg T+2\tau} |\psi^i(t)\rangle &\rightarrow |\psi_f^i\rangle = \int_{-\infty}^{\infty} dk f^{\text{out}}((\omega_k - \omega_{31})/c) \\ &\quad \times a_k^+ |0\rangle |A(t)\rangle, \quad (23) \\ f^{\text{out}}((\omega - \omega_{31})/c) &= \exp\{i\Phi\} f^{\text{in}}(-(\omega - \omega_{31})/c), \\ \Phi &= (\omega - \omega_{31})(T + 2\tau) \\ &\quad + (\omega_{31} - \Delta)T + \varphi_{21} + \pi/2. \quad (24) \end{aligned}$$

For comparison see the initial state in Eq. (1). Solution (24) gives the relation between the wave function of the recalled output photon, $f^{\text{out}}((\omega - \omega_{31})/c)$, and the wave function, $f^{\text{in}}((\omega - \omega_{31})/c)$, of the incident photon and it reflects the delay $(T + 2\tau)$ and unitary evolution of the photon in the medium. As seen from Eq. (24) we can vary the phase Φ of the photon using the phases of the laser pulses, φ_2 and φ_1 . We also note that in comparison to the input photon, the spectrum of the output photon is reversed around the center frequency of the transition. So if the spectrum of the first photon is symmetric: $f^{\text{in}}((\omega - \omega_{31})/c) = f^{\text{in}}(-(\omega - \omega_{31})/c)$, the recalled photon will reproduce the initial single-photon quantum state, where Φ will determine only the time delay and an overall additional phase shift. In general Eq. (24) opens interesting possibilities to manipulate single-photon states.

The possibility to truthfully map a nonstationary single-photon wave packet in a photon echo medium has been pointed out previously [9,10]. However, in the conventional two-level photon echo scheme [5] the reconstructed field is absorbed as it propagates in the sample, and although the state could be correctly mapped onto the atomic ensemble if the sample has a high optical density, it can be only partly extracted. In fact, the output signal intensity in a conventional photon echo experiment is typically only a few percent of the input field intensity (however, the experiment can be configured such that the sample coherently amplifies the output signal such that it becomes stronger than the input signal [11]). In comparison with quantum state mapping using slow light and EIT [2] we use the same energy level diagram configuration, but instead of causing the sample to be transparent through EIT we just time reverse the absorption process. At the same time we think that it would be important to study the spectral possibilities of both techniques more precisely, but this question requires further analysis. A potential problem with the photon echo approach might be the requirement of having pulse areas of exactly π for the pulses on the $|2\rangle$ - $|3\rangle$ transi-

tion. However, it is easy to find the probability P to retrieve a single photon wave packet for the pulse area ϑ of the laser pulses: $P(\vartheta) = \sin^4(\vartheta/2)$. If $\vartheta = \pi + \varepsilon$, where $|\varepsilon| \ll 1$, the probability P is very close to 1 and small variations of ϑ around π will then hardly affect the quality of the reconstructed quantum field. As our scheme is designed for use in macroscopic inhomogeneously broadened media, it is suitable for nonstationary fields and should readily be possible to extend to multiphoton fields. However, the in-depth analysis of this problem is an impetus for future research.

We conclude that our photon echo technique opens good possibilities to store, to recall, and also to change parameters of the photon wave function without destroying it. We mean that the techniques could be very interesting for studying intimate details of the interaction of nonstationary single-photon quantum fields with macroscopic systems of atoms and molecules [12].

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