Quantum Entanglement of Fock States with Perfectly Efficient Ultraslow Single-Probe Photon Four-Wave Mixing

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We propose a method to achieve quantum entanglement of two Fock states with perfectly efficient, ultraslow propagation enhanced four-wave mixing. A cold atomic medium is illuminated with a twomode cw control laser to produce coherent mixtures of excited states. An ultraslowly propagating, single-photon quantum probe field completes the four-wave mixing with 100% photon flux conversion efficiency, creating a depth dependent entanglement of two Fock states. We show that at a suitable propagation distance, a maximum entangled state is created with a single-photon wave-packet state that has 50% probability of being in each of two product-type Fock states.

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Quantum entanglement $[1-3]$ is one of the most striking features of quantum mechanics. The entanglement of the quantum states of separate particles, such as the entanglement of photon pairs [4–6], plays a crucial role in quantum information science [7–10] and has been intensively studied. The entanglement of multiple Fock states with a single (or a few) ultraslow photon via fourwave mixing (FWM), however, has not been reported [11]. Here, we discuss a scheme to achieve quantum entanglement of two Fock states with perfectly efficient, ultraslow propagation enhanced FWM. An atomic medium is illuminated with a two-mode continuous-wave (cw) control laser to coherently mix two excited states. An ultraslowly propagating single-photon quantum probe field completes the FWM with 100% photon flux conversion efficiency, creating a depth dependent entanglement of two Fock states [12]. At a suitable propagation distance, a maximum entangled state is created with a single-photon wave-packet state that has 50% probability of being in each of two product-type Fock states. We further demonstrate the ''storage and retrieval'' of such an entangled state, thereby opening the possibility of applications of the technique to quantum information science.

We consider a lifetime broadened three-state system interacting with a single, ultraslowly propagating probe photon (ω_{p1} , assumed to be in a wave-packet state) that is detuned far from the one-photon resonance (Fig. 1). A strong cw control laser (ω_{c1}) is tuned so that the exact two-photon resonance between the ground state and the lower excited state is established. We show that when a second control laser at ω_{c2} is present, a 100% efficient FWM process occurs with a new, ultraslow photon being created at the frequency $\omega_{p2} = \omega_{p1} - \omega_{c1} + \omega_{c2}$. Therefore, deep in the medium the state of the quantized electromagnetic (EM) field oscillates, as it propagates, between the product states $|1_{\omega_{p1}}\rangle|0_{\omega_{p2}}\rangle$ and $|0_{\omega_{p1}}\rangle|1_{\omega_{p2}}\rangle$. This Fock state entanglement occurs as a result of conserving the number of probe photons during the FWM process.

We use the Heisenberg representation to calculate the atomic dynamics and the probe photon field, but we treat the strong two-mode control fields and their interactions with the medium classically. Taking the plane wave and the slowly varying-phase-and-amplitude approximations we arrive at the following equation of motion for the complex amplitude of the quantized probe field:

$$
\left(\frac{\partial \hat{E}_{pj}^+}{\partial z}\right)_t + \frac{1}{c} \left(\frac{\partial \hat{E}_{pj}^+}{\partial t}\right)_z = \frac{i\kappa_{12}\hbar}{D_{12}} \hat{S}_{pj} \qquad (j = 1, 2), \quad (1)
$$

where \hat{S}_{pj} are the polarization operators, κ_{12} = $2\pi N\omega_{12}|\tilde{D}_{12}|^2/\hbar c$, and *N* and D_{12} are the density of

FIG. 1. A three-level purely lifetime broadened atomic system couples with a quantized probe field $\hat{E}_{p1}(\omega_{p1})$ and a strong calssically treated cw two-mode control field $(\Omega_{c1}, \omega_{c1})$; Ω_{c2}, ω_{c2}). The probe field frequency ω_{p1} and the control field frequency ω_{c1} are such that the exact two-photon resonance between $|1\rangle$ and $|3\rangle$ is achieved. The complex detunings are defined as $d_{pj} = \delta_{pj} + i\gamma_2/2$ (*j* = 1, 2), where δ_{pj} and γ_2 are the detunings from level $|2\rangle$ and the decay rate of level $|2\rangle$, respectively.

atoms and the dipole moment for the transition $|1\rangle \rightarrow |2\rangle$, respectively. We assume that $|d_{pj}\tau| \gg 1$, $|d_{pj}| \gg |\Omega_{cj}|$, and $|(|\Omega_{c1}|^2/d_{p1}) + (|\Omega_{c2}|^2/d_{p2})| \gg 1/\tau$, so that near adiabatic behavior will be achieved. Since only one probe photon propagates through the medium, the ground-state diagonal element of the density matrix remains close to unity. With these assumptions an adiabatic following approximation for \hat{S}_{pj} is valid, resulting in

$$
\hat{S}_{pj} = -\frac{D_{12}}{\hbar d_{pj}} \hat{E}_{pj}^{+} - \frac{\Omega_{cj}^{*}}{d_{pj}} e^{-i(\omega_3 - \omega_1)z/c} \hat{R}_{3}.
$$
 (2)

In Eq. (2) \hat{R}_3 is the operator for $\hat{\rho}_{13}$ (after appropriate phase transformations), which obeys

$$
\frac{\partial \hat{R}_3}{\partial t} = -i D_{12} \left(\frac{\Omega_{c1}}{\hbar d_{p1}} \hat{E}_{p1}^+ + \frac{\Omega_{c2}}{\hbar d_{p2}} \hat{E}_{p2}^+ \right) e^{i(\omega_2 - \omega_1)z/c} \n- i \left(\frac{|\Omega_{c1}|^2}{d_{p1}} + \frac{|\Omega_{c2}|^2}{d_{p2}} \right) \hat{R}_3.
$$
\n(3)

Taking the Fourier transform of Eqs. (1) – (3) , we obtain, with $D = d_{p1} |\Omega_{c2}|^2 + d_{p2} |\Omega_{c1}|^2$,

$$
\left(\frac{\partial \hat{Z}_{pj}^+}{\partial z}\right)_{\omega} - i\frac{\omega}{c} \hat{Z}_{pj}^+ = i\kappa_{12} \left[\frac{(\omega d_{pl} - |\Omega_{cl}|^2) \hat{Z}_{pj}^+ + \Omega_{cj}^* \Omega_{cl} \hat{Z}_{pl}^+}{D - \omega d_{pj} d_{pl}} \right] \qquad (j, l = 1, 2; j \neq l), \tag{4}
$$

where $\hat{Z}_{pj}^+(z, \omega)$ is the Fourier transform of $\hat{E}_{pj}^+(z, t)$ with ω being the transform variable. Equation (4) can be solved analytically for arbitrary initial conditions. Here, we consider only the case where initially there is no photon in the ω_{p2} mode. In order to be able to carry out the inverse transform analytically, thereby gaining important physical insight, we neglect the ω dependent terms in the coefficients of the solutions of Eq. (4) and keep only terms that are linear in ω in the exponents [13]. This step is justified because the assumptions made previously lead to a well behaved adiabatic following solution. We thus have

$$
\hat{E}_{pj}^{+} = \sqrt{\frac{2\pi\omega_{pj}\hbar}{A_{\rm eff}c\tau}} \frac{\Omega_{cj}}{|\Omega|^{2}} [\Omega_{c1}^{*}M_{j}^{(1)}(z, t)\hat{\alpha}_{p1} + \Omega_{c2}^{*}M_{j}^{(2)}(z, t)\hat{\alpha}_{p2}] \qquad (j = 1, 2),
$$
\n(5a)

$$
M_j^{(m)}(z,t) = P_{p1}\left(t - \frac{z}{V_{g1}}\right) + (-1)^j \frac{|\Omega_{c1}|^2 \delta_{m2} - |\Omega_{c2}|^2 \delta_{m1}}{|\Omega_{cj}|^2} e^{ib_0 z} P_{p1}\left(t - \frac{z}{V_{g2}}\right),\tag{5b}
$$

where $|\Omega|^2 = |\Omega_{c1}|^2 + |\Omega_{c2}|^2$, A_{eff} is the effective beam cross section, $b_0 = -\kappa_{12} |\Omega|^2 / D$, $P_{p1}(t)$ is the pulse shape function for the initial photon wave packet, $\delta_{m1(m2)}$ is the Kronecker δ function, $\hat{\alpha}_{pj}$ are the annihilation operators for photons of frequency ω_{pi} , and

$$
\frac{1}{V_{g1}} = \frac{1}{c} + \frac{\kappa_{12}}{|\Omega|^2},
$$
\n
$$
\frac{1}{V_{g2}} = \frac{1}{c} + \frac{\kappa_{12}}{|\Omega|^2} \Big(\frac{|\Omega_{c1}|^2 |\Omega_{c2}|^2 (d_{p1} - d_{p2})^2}{D^2} \Big).
$$
\n(6)

Equations (5) and (6) indicate that in general the wave packet will break into two parts that travel with different group velocities, but both retain a pulse shape identical to that of the input probe field. We consider here only cases where initially there are photons in the ω_{p1} mode but no photon in the ω_{p2} mode. In this case all of the following results are independent of $M_j^{(2)}$. Specifically, let us consider the case where $|\Omega_{c1}| = |\Omega_{c2}|$, $\delta_{p1} = 0$, $\delta_{p2} \gg \gamma_2$, $\gamma_2 \tau \gg 1$, and $|\Omega_{c1}|^2 / \gamma_{p2} \gg 1/\tau$. This is the case that may have potential applications in quantum information science. A close inspection of the above treatment shows that in this case $1/V_{g2} = 1/c + \kappa_{12}(1$ $i2\gamma_2/\delta_{p2}$ ²/ $|\Omega_{c1}|^2$. Thus, both fields now propagate with the same group velocity. The state vector of the EM field when it exits from the medium can now be expressed as [14]

$$
|\Psi(z, t)\rangle = a_{10}(z, t)|1_{\omega_{p1}}\rangle|0_{\omega_{p2}}\rangle
$$

+ $a_{01}(z, t)|0_{\omega_{p1}}\rangle|1_{\omega_{p2}}\rangle,$ (7a)
 $(1 + e^{-2Bz_0} - e^{-Bz_0})$

$$
|a_{10(01)}(z, t)|^2 = \left(\frac{1 + e^{-2Bz_0}}{4} \pm \frac{e^{-Bz_0}}{2} \cos(Az_0)\right) \times P_{p1}^2 \left(t_1 = t - \frac{z_0}{V_{g1}} - \frac{z - z_0}{c}\right), \quad (7b)
$$

where $A = \text{Re}(b_0) = -2\kappa$ $B = Im(b_0) =$ $2\kappa_{12}\gamma_2/(\delta_{p2})^2$ [15], and z_0 is the length of the medium. The absorption terms in Eq. (7b) are due to spontaneous emission out of level $|2\rangle$ and can be made small even in the case where the wave packet becomes completely trapped in the medium. Equation (7b) shows that when the absorption is small, by choosing $Az_0 = (2n + 1)\pi/2$ the absorption is small, by choosing $Az_0 = (2n + 1)\pi/2$
we get $|a_{10}| = |a_{01}| = P_{p1}(t_1)/\sqrt{2}$. The state vector is then given by

$$
|\Psi(z,t)\rangle = \frac{P_{p1}(t_1)}{\sqrt{2}} (|1_{\omega_{p1}}\rangle |0_{\omega_{p2}}\rangle \pm i|0_{\omega_{p1}}\rangle |1_{\omega_{p2}}\rangle). \tag{8}
$$

This is a maximally entangled linear combination of product-type Fock states. If we choose $Az_0 = n\pi$, the wave packet is given by $|\Psi(z, t)\rangle = P_{p1}(t_1)|0_{\omega_{p1}}\rangle|1_{\omega_{p2}}\rangle.$ Thus, with a single photon at ω_{p1} as input, the exiting wave packet contains only one photon at ω_{p2} . Such a

100% efficient FWM process can serve as a perfectly efficient single-photon frequency switch.

It is interesting to examine the entanglement resulting from having exactly two photons at ω_{p1} and none at ω_{p2} in the initial wave packet. In this case, if the absorption is small, the state vector of the EM field after exiting the medium will be $|\Psi(z, t)\rangle = a_{20}(z, t)|2_{\omega_{p1}}\rangle|0_{\omega_{p2}}\rangle +$ $a_{11}(z, t) \left| \frac{1}{\omega_{p1}} \right\rangle \left| \frac{1}{\omega_{p2}} \right\rangle + a_{02}(z, t) \left| \frac{0}{\omega_{p1}} \right\rangle \left| \frac{2}{\omega_{p2}} \right\rangle$. It can be shown that if $Az_0 = n\pi$, the wave packet exiting the medium will contain two photons at ω_{p2} and none at ω_{p1} , yielding a 100% photon flux conversion efficiency. On the other hand, if $Az_0 = (2n + 1)\pi/2$, we obtain On the other hand, if $Az_0 = (2n + 1)\pi/2$, we obtain $|a_{20}| = |a_{02}| = P_{p1}(t_1)/2$, $|a_{11}| = P_{p1}(t_1)/2$, again an entangled linear combination of product-type Fock states.

We now demonstrate through rigorous numerical calculations, using a set of experimentally achievable parameters, the validity of the analytical treatment described [16]. We take $\tau = 10 \mu s$, $\Omega_{c1} \tau = \Omega_{c2} \tau =$ 2500, $\delta_{p1} = 0$, $\delta_{p2}\tau = 6.28 \times 10^5$, $\gamma_2\tau = 628$, $\gamma_3\tau =$

0.005, $z_0 = 5$ cm, and $\kappa_{12}\tau z_0 = 2.356 \times 10^7$. With these parameters, the spatial width of the wave packet as a function of position is $\Delta z/z_0 \approx 0.265$. Thus, when the peak of the wave packet reaches $z_0/2$, almost all of the area under the packet is contained inside the medium. In Fig. 2 we show a surface plot of $I_{p1}(z, t)/I_{p1}(0, 0)$ as a function of time and position. It oscillates between 0 and 1 with a period of $2\pi/|A_{z_0}| = 0.0837$. At the exit of the medium, the delayed wave packet is shown in Fig. 3.

One of the main motivations of the present study is the possibility of ''storing and retrieving'' a single-photon wave packet with its entanglement properties preserved. For this purpose, we calculate

$$
N\hat{\rho}_{33}(z, t) = \frac{\kappa_{12}}{2|\Omega_{c1}|^2} \frac{[1 + \cos(Az)]}{A_{\text{eff}}\tau} P_{p1}^2 \left(t - \frac{z}{V_{g1}}\right)
$$

$$
\approx \frac{1}{A_{\text{eff}}V_{g1}\tau} P_{p1}^2 \left(t - \frac{z}{V_{g1}}\right).
$$

When the peak of the packet reaches $z_0/2$, we have

$$
I = N \int dS dz \langle \hat{\rho}_{33} \rangle = \int_0^{z_0} \frac{dz}{V_{g1} \tau} \left| P_{p1} \left(\frac{z_0/2 - z}{V_{g1}} \right) \right|^2 \approx 1 \qquad \left[\int_{-\infty}^{\infty} P_{p1}^2 \left(t - \frac{z}{c} \right) \frac{dz}{c \tau} = 1 \right].
$$

This indicates that if the two-mode control laser is cut off, the probe photon wave packet is absorbed and an atom is in state $|3\rangle$; i.e., the population is trapped. Indeed, after

1.0 0.8 T 40.6

the control lasers are cut off the ''wave of atomic coherence" no longer propagates and $|\rho_{13}|$ remains the same value. Thus, the entangled photon state is coherently mapped to the atomic spin state. If the control lasers are restored to the same values, the wave packet that was originally propagating at the time of cutoff is restored with its original amplitude and shape so that a

FIG. 2. Surface plot of $I_{p1}(z, t)/I_{p1}(0, 0)$ versus t/τ and z/z_0 for $|\Omega_{12}\tau| = |\Omega_{c2}\tau| = 2500$, $\delta_{p1}\tau = 0$, $\delta_{p2}\tau = 6.28 \times 10^5$, $\gamma_2 \tau = 628$, $\gamma_3 \tau = 0.005$, and $\kappa_{12} \tau z_0 = 2.356 \times 10^7$. Note the oscillations between 0 and 1. At the points where the ω_{p1} component is unity, the ω_{p2} component will be near zero. Also, note that the difference in t_r/τ between when the peak entered the medium and when it exits is about 1.885. This means that when the peak of the pulse reaches $z_0/2$, more than 99% of the pulse lies in the medium. Such a probe pulse trapping is required for ''storing'' and ''retrieving'' such pulses.

FIG. 3. Plot of the time dependence of $I_{p1}(z_0, t)/I_{p1}(0, 0)$ at the exit of the medium. The parameters used are the same as in Fig. 2. Note that the amplitude is about 0.9 and that its full width at $1/e$ is very close to 1, indicating that the absorption is small and the adiabatic approximation works very well in this case. The solid line is obtained with full numerical calculation with no adiabatic approximation. The dashed line is from the analytical solution Eq. (5).

photon is regenerated with its state vector exactly the same as that of Eq. (8). Therefore, the entanglement properties of the wave packet are fully recovered. This replication of the probe photon state with its entanglement properties preserved is crucially important for quantum computation where the preservation of the entangled properties during the storage and recovery is of primary importance.

We have presented the first study on the production of a frequency entangled state using a single ultraslow probe photon. Such an entanglement of Fock states is of fundamental importance in quantum information science. The theory and method proposed here are drastically different from the usual down-conversion method where polarization entangled photon pairs can be produced only by using high energy, short-pulsed lasers with low efficiency. The two-mode ultraslow, single probe photon FWM model described here may lead to further understanding of the fundamental physics of ultraslow wave entanglement, efficient generation and ultraslow propagation of entangled photon pairs [17], ''storage'' and subsequent regeneration of entangled photons, and perhaps new technology for novel applications in quantum communication and quantum computing.

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- [13] Inclusion of ω^2 terms in the exponent will lead to additional, distance dependent attenuation and pulse spreading. With our assumptions these higher-order contributions are negligible even when the concentration and thickness are such that ''storage'' and ''retrieval'' can be achieved.
- [14] For convenience, we use the Schrödinger picture to express the state vector.
- [15] A small factor due to $Im(V_{g2})$ has been neglected.
- [16] In the numerical calculation, we solve both the atomic equations of motion (not shown here) and Eq. (1) simultaneously without invoking the strict adiabatic following. The parameters used are very similar to those of a typical alkali-metal vapor.
- [17] A significantly attenuated dye laser can provide the single-photon source needed. The familiar downconversion source may also be used as a single-photon source. In this case, one of the photons can be used as the even/electronic trigger with the other photon as the input photon. It can also serve as a source of the entangled photon pair. It would then be interesting to investigate the ultraslow propagation and four-wave mixing associated with such photon pairs in a separate vapor cell using the scheme described here. See also T. Nakanishi *et al.*, Phys. Rev. A **67**, 043809 (2003).C. Becher *et al.*, Physica (Amsterdam) **13E**, 412 (2002).