Optical Precursor of a Single Photon

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We report the direct observation of optical precursors of heralded single photons with step- and square-modulated wave packets passing through cold atoms. Using electromagnetically induced transparency and the slow-light effect, we separate the single-photon precursor, which always travels at the speed of light in vacuum, from its delayed main wave packet. In the two-level superluminal medium, our result suggests that the causality holds for a single photon.

Wave-particle duality is at the heart of quantum mechanics. To pave the way towards the realization of practical quantum information processing [1], it is crucial to understand and demonstrate the duality at the single-photon level. The optical precursor, which refers to the propagation of the front of a step optical pulse, always travels at c, the speed of light in vacuum, in any dispersive medium. This wave property, first predicted by Sommerfeld and Brillouin in 1914 [2,3], is of great interest not only for fundamental reasons since it is related to Einstein's causality, but also for applications because of its connection to the maximum speed of information transmission [4–10]. Together with other experiments [11,12], now it is clear that the precursor is the fastest part in the propagation of an optical pulse even in a superluminal medium [7,8,10]. However, classical precursors are entirely based on macroscopic electromagnetic wave propagation, whereas envisioned applications, particularly quantum cryptography, involve interactions of single photons with atoms or molecules quantum mechanically. Does optical precursor exist at a single-photon level? Is the speed of a photon able to exceed c?

When a step-modulated optical pulse propagates through a dispersive medium, the optical precursor associated with the abrupt rising wave front travels at c with nearly no attenuation even in a highly absorptive medium. The existence of optical precursors seems surprising, but its nature can be understood physically. As a light field is switched on suddenly, its steep wave front experiences no barrier from the medium as the atoms have a finite response time. After a sufficiently long time when the atoms have established their induced electrodipoles, propagation of the main pulse gets modified and follows the group velocity. Therefore, in a slow-light medium a precursor can be separated from the delayed main field, as we first demonstrated in cold atoms with electromagnetically induced transparency (EIT) [8,13]. The nearly lossless nature of precursors may find application in optical communication through absorptive media. In a superluminal medium with negative group velocity, it was found that no light signal

components can move ahead of the precursor and thus the causality is verified [10].

A single photon is described by quantum mechanics. The propagation of a single photon through the slow and fast light medium has been studied in only a few and limited experiments [14,15] since it has been difficult to produce single photons with controllable waveforms. While some believe that the causality principle also applies to single photons [16,17], others argue that the group velocity may be the same as the velocity of individual photons [18]. Therefore, detection of single-photon precursors may shine light in the understanding of quantum information transmission. Recent progress in generating narrow-band nonclassical photon pairs [19–21] using EIT led to the observation of optical precursors in two-photon quantum correlation [22,23]. However, the observed biphoton precursors are produced during a nonlinear optical process and are not separable from the entire two-photon wave packet. In this work, we successfully separate these precursors from the main wave packet by passing one of the paired photons through an EIT slow-light system. Moreover, with heralded single photons whose waveform are shaped by an electro-optic modulator(EOM) [24], we can generate single-photon precursors. Our result suggests that the causality holds for a single photon.

The experimental configuration we use to generate single-photon precursors is illustrated in Fig. 1. We work with two two-dimensional (2D) ^{85}Rb magneto-optical traps (MOT) with a temperature of about 100 μK. In the first MOT (MOT1), with the presence of counterpropagating pump (ω_p, 780 nm) and coupling (ω_c, 795 nm) beams which are aligned at a 3° angle from the longitudinal axis, we produce Stokes (ω_s) and anti-Stokes (ω_{as}) paired photons [23]. When the Stokes photon is detected by the single-photon detector D_1, the heralded anti-Stokes photon is passed through an EOM (10 GHz, EOspace) which is driven by a fast waveform generator (Tektronix AFG3252) that is trigged by the detection of Stokes photons. In this way, we generate heralded single anti-Stokes photons with arbitrary waveforms within their temporal length [24].
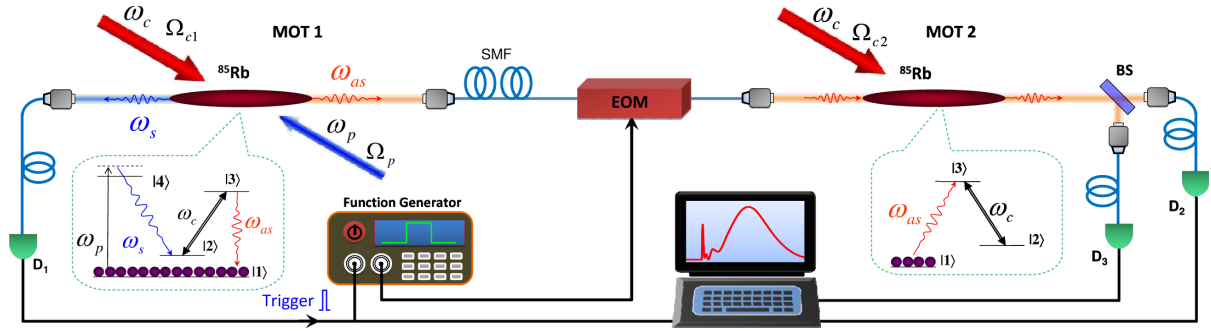
FIG. 1 (color online).   Schematics of the experimental setup. In the presence of a counterpropagating pump (ω_p) and coupling (ω_c) beams, narrow-band paired Stokes (ω_s) and anti-Stokes (ω_{as}) photons are produced from a cold ^{85}Rb atomic ensemble in MOT1 and coupled into opposing single-mode fibers (SMF). The anti-Stokes photons are passed through an EOM driven by a function generator triggered by the detection of Stokes photons at the single-photon detector D_1. We then send the heralded anti-Stokes photons with amplitude modulation to the second cold ^{85}Rb atomic ensemble at MOT2. A 50%-50% beam splitter (BS) and two detectors (D_2 and D_3) are used to confirm the single-photon quantum nature. The ^{85}Rb atomic energy levels are chosen as $|1\rangle = |5S_{1/2}, F = 2\rangle$, $|2\rangle = |5S_{1/2}, F = 3\rangle$, $|3\rangle = |5P_{1/2}, F = 3\rangle$ and $|4\rangle = |5P_{3/2}, F = 3\rangle$.

Then we send the modulated anti-Stokes photons through the cold atoms in the second MOT (MOT2) that is controlled by a coupling beam directed from the same coupling laser in MOT1. A beam splitter (BS) and two detectors (D_2 and D_3) are used to verify the quanutm nature of the transmitted anti-Stokes photons. In both MOTs, the atoms are prepared in the ground state $|1\rangle$, and the optical depths (OD) at the anti-Stokes transition can be varied from 0 to 55. The coupling laser is on resonance with $|2\rangle \leftrightarrow |3\rangle$ transition. The pump laser in MOT1 is blue detuned from $|1\rangle \leftrightarrow |4\rangle$ transition by 146 MHz. We run the experiment periodically with a MOT time of 4.5 ms and a single-photon generation window of 0.5 ms during each cycle.

Detection of the Stokes photon establishes the time origin of the anti-Stokes photon [24]. After passing though the EOM, the conditional single-photon wave packet can be described by

$$\psi_0(\tau) = \langle 0|m(\tau)\hat{a}_{as}(t_s + \tau)\hat{a}_s(t_s)|\Psi_{s,as}\rangle|_{t_s=0}$$
$$= \frac{1}{2\pi}\int \Phi_0(\omega)e^{-i\omega\tau}d\omega, \qquad (1)$$

where $|0\rangle$ and $|\Psi_{s,as}\rangle$ are the vacuum and two-photon state. $\hat{a}$ is the photon annihilation operator. $m(\tau)$ is the amplitude modulation function with $\tau = t_{as} - t_s$ as the relative time delay. $\Phi_0(\omega)$ describes the spectrum of the conditional single photon. After the anti-Stokes photon has interacted with the cold atoms in MOT2, we model the output wave packet as

$$\psi(\tau) = \frac{1}{2\pi}\int \Phi_0(\omega)e^{i[k_{as}(\omega)L-\omega\tau]}d\omega, \qquad (2)$$

where $k_{as}(\omega)$ is the complex wave number of the medium with a length L. The coincidence counts in a time period T with a bin width Δt can be calculated as $\eta|\psi(\tau)|^2T\Delta t$ where η is the two-photon detection efficiency. In our model, the single-photon wave packet propagates like a

"classical" wave. These heralded single photons with controllable waveforms allow us to directly verify their particle-wave duality in time domain.

We first characterize the photon source without modulation, i.e., $m(\tau) = 1$. In all measurements, the parameters in MOT1 are fixed. We set the pump and coupling Rabi frequencies as $\Omega_p = 0.5\gamma_{13}$ and $\Omega_{c1} = 3.0\gamma_{13}$, where $\gamma_{13} = 2\pi \times 3$ MHz is the electro dipole relaxation rate between $|1\rangle$ and $|3\rangle$. The OD of MOT1 is about 30. Figure 2(a) displays the two-photon correlation without passing the anti-Stokes photons through the EOM and MOT2. The spikeslike oscillatory structure at the leading edge results from the interference between the precursor and main waveform [22]. Figure 2(b) shows two-photon coincidence counts when the MOT2 is present with OD = 14 and $\Omega_{c2} = 3.5\gamma_{13}$. Because of EIT and slow-light effects, the main wave packet is delayed by about 100 ns. The precursor wave front always travels at c with nearly no attenuation and can be separated from the main wave packet, shown in Fig. 2(b) as the sharp peak at $\tau = 0$. Figure 2(c) shows that in the two-level system when the coupling laser is turned off ($\Omega_{c2} = 0$), only the unabsorbed precursor is left. In Fig. 2(d), we plot the relative time delays (compared to that through the vacuum) of the main wave packet and the precursors as functions of OD. It is clear that the main wave packet of the heralded anti-Stokes photon travels at the group velocity while the precursor shows no relative delay to the propagation through vacuum. These results confirm that the oscillatory structure previously reported in Refs. [22,23] indeed resulted from mixing of precursor with main waveform. The EIT effect of MOT2 allows us to separate the precursor from the main wave packet.

Next we drive the EOM with a step-on waveform. The modulated heralded anti-Stokes photon waveform is shown in Fig. 3(a) where the biphoton precursor induced oscillatory structure has been eliminated by properly adjusting the switch-on time. The two-photon correlation
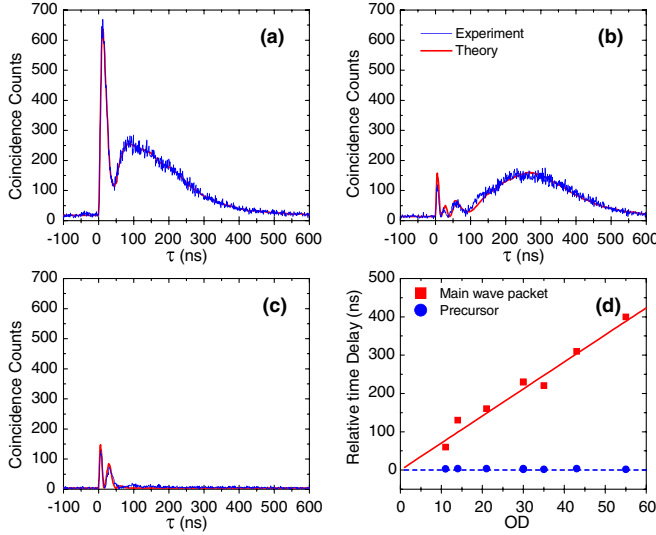
FIG. 2 (color online). Separation of optical precursors without modulation. Two-photon coincidences are measured after the anti-Stokes photons passing through (a) vacuum without atoms present, (b) the EIT system ($\Omega_{c2} = 3.5\gamma_{13}$, OD = 14) and (c) the two-level system ($\Omega_{c2} = 0$, OD = 14) in MOT2. The Stokes and anti-Stokes two-photon coincidence counts are collected for $T = 900$ s with $\Delta t = 1$ ns bin width. The theoretical curve in a is the best fitting using $\eta|\psi_0(\tau)|^2 T \Delta t$. Then the $\psi_0(\tau)$ is used to calculate the theoretical curves in b and c without any adjustable parameters following Eqs. (1) and (2). (d) The relative time delay (respected to the vacuum) of the precursor and main wave packet as functions of OD. The red solid line is the calculated EIT group delay.

after the anti-Stokes photon has passed through the EIT medium in MOT2 (OD = 18) is shown in Fig. 3(b). The precursor is clearly seen at the rising edge and separated from the delayed main waveform. To test single-photon causal propagation in a superluminal medium, we turn off the coupling laser in MOT2 to work in a two-level system. The negative group velocity of the two-level system at OD = 2.5 is measured by propagating a Gaussian pulse through the cold atoms. As shown in the inset of Fig. 3(d), we observe a peak advance of at least 40 ns and with about 10% transmission compared to the propagation through vacuum. However, as shown in Fig. 3(c), there is no observable advancement relative to the rising edge. This result indicates that the optical precursor is always the fastest part even in superluminal propagation and Einstein's causality holds.

We now study the heralded single-photon propagation with a square-modulated waveform that has a temporal length of 100 ns and a rise (fall) time of 3 ns, as shown in Fig. 4(a). After passing the anti-Stokes photon through the EIT system, we clearly observe the optical precursor at the rising edge, as shown in Fig. 4(b). The main wave packet arrives with 50 ns delay and interferes with the precursor at the falling edge. Figure 4(c) shows the result obtained in the two-level system. By varying the optical depth of MOT2, we measure the relative time delays of the
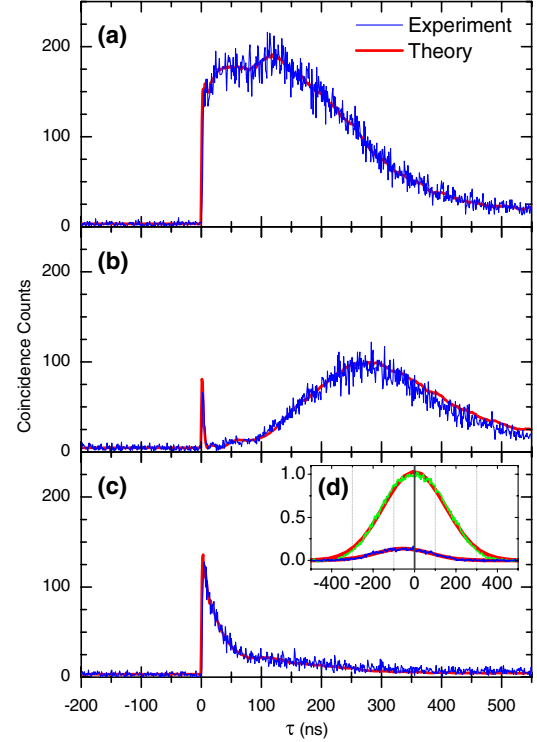

FIG. 3 (color online). Single-photon optical precursors from a step amplitude modulation. (a) The heralded anti-Stokes photon waveform with a step modulation. (b) and (c) are two-photon coincidences after the anti-Stokes photons passing through the EIT system ($\Omega_{c2} = 3.8\gamma_{13}$, OD = 18) and two-level system ($\Omega_{c2} = 0$, OD = 2.5) in MOT2, respectively. Inset (d) shows a Gaussian pulse propagation in the two-level system with a peak advancement of about 40 ns (the lower curve) compared to the reference pulse (the up curve).

main waveform and precursor compared to their propagation through vacuum without atoms. Our result in Fig. 4(d) suggests that the wave front of an optical precursor indeed propagates at c independent of OD and other material properties (in both EIT and two-level systems). The theory agrees well with experiment.

The nonclassical property of correlated paired photons can be quantified by the violation of Cauchy-Schwartz inequality [25]. The Glauber correlation function, measured as two-photon coincidence counts, is defined as $G_{i,j}^{(2)}(\tau) = \langle \hat{a}_i^\dagger(t)\hat{a}_j^\dagger(t+\tau)\hat{a}_j(t+\tau)\hat{a}_i(t)\rangle$. The normalized correlation function is obtained as $g_{i,j}^{(2)}(\tau) = G_{i,j}^{(2)}(\tau)/(R_i R_j)$ where $R_{i/j} = \langle \hat{a}_{i/j}^\dagger \hat{a}_{i/j}\rangle$ are the single-photon rates. For a classical light source, its normalized correlation functions are bounded by the Cauchy-Schwartz inequality $[g_{i,j}^{(2)}(\tau)]^2/[g_{i,i}^{(2)}(0)g_{j,j}^{(2)}(0)] \leq 1$ [25]. For the biphotons generated from MOT1, we measure their cross-correlation function from the Stokes and anti-Stokes coincidence counts [Fig. 2(a)] and obtain $g_{s,as}^{(2)}(\tau)$ that has a maximum value of 40. We measure the autocorrelations of the Stokes and anti-Stokes photons using a beam splitter and obtain $g_{s,s}^{(2)}(0) = g_{as,as}^{(2)}(0) = 2.0$. Then we obtain a
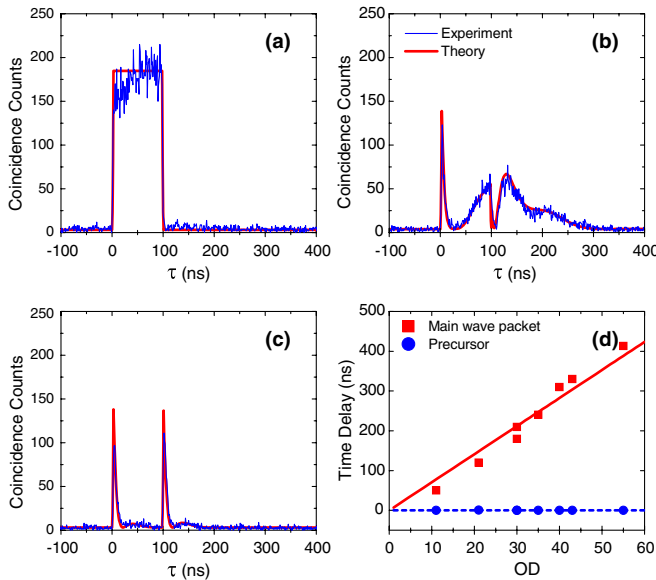
FIG. 4 (color online). Single-photon optical precursors from a square amplitude modulation. (a) The heralded anti-Stokes photon waveform with a square modulation. (b) and (c) are two-photon coincidences after the anti-Stokes photons passing through the EIT system ($\Omega_{c2} = 3.5\gamma_{13}$, OD = 10) and two-level system ($\Omega_{c2} = 0$, OD = 10) in MOT2, respectively. (d) The relative time delay (respected to the vacuum) of the precursor and main wave packet as functions of OD. The red solid line is the calculated EIT group delay.

violation of the inequality by a factor of 400. After the anti-Stokes photons are passed through MOT2, we obtain $g_{s,as}^{(2)}(\tau)$ with a maximum value of 10 and 35 at the precursor peak for the EIT [Fig. 2(b)] and two-level [Fig. 2(c)] system, respectively. Thus the inequality is still violated by a factor of 25 and 306. Therefore, the nonclassical property of the biphotons are preserved after the anti-Stokes photons have interacted with the atoms in MOT2. More importantly, the precursors observed in Fig. 2 are not from classical fields.

The single-photon quantum nature of the heralded anti-Stokes photons can be further verified by using a BS as shown in Fig. 1, because a single-photon incident at a BS must go to one or the other port. A measure of the quality of heralded single photons is given by the conditional correlation function $g_c^{(2)} = N_{123}N_1/(N_{12}N_{13})$ [26] where N_1 is the number of the Stokes counts at D_1, N_{12} and N_{13} are the number of twofold coincidence counts, and N_{123} is the threefold coincidence counts. A classical field must satisfy $g_c^{(2)} \geq 1$. With a coincidence window of 200 ns, we measure $g_c^{(2)} = 0.19 \pm 0.01$ for the case shown in Fig. 2(a). After passing the anti-Stokes photons through the EIT system in MOT2 [Fig. 2(b)], we obtain $g_c^{(2)} = 0.39 \pm 0.04$. In the modulation case we obtain $g_c^{(2)} = 0.16 \pm 0.11$ when the atoms are not present in MOT2 [Fig. 3(a)] and $g_c^{(2)} = 0.21 \pm 0.12$ for the EIT case [Fig. 3(b)]. In all these measurements, the precursors are within the coincidence window. Because a two-photon Fock state has $g_c^{(2)} = 0.5$, the fact that $g_c^{(2)} < 0.5$ suggests the near-single-photon character of the heralded anti-Stokes photons and the observed precursors.

Our observation of the optical precursor of heralded single photons provides a physical picture in understanding the transmission of quantum information. Our experimental results indicate that the optical precursor traveling at c is always the fastest part of the single-photon wave packet in both slow-light and superluminal media. It may bring a closure to the debate on the true speed of information carried by a single photon [18].

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[1] D. Bouwmeester, A. K. Ekert, and A. Zeilinger, *The Physics of Quantum Information* (Springer-Verlag, Berlin, 2000).
[2] A. Sommerfeld, Ann. Phys. (Leipzig) **349**, 177 (1914).
[3] L. Brillouin, Ann. Phys. (Leipzig) **349**, 203 (1914).
[4] K. E. Oughstun and G. C. Sherman, *Electromagnetic Pulse Propagation in Causal Dielectrics* (Springer-Verlag, Berlin, 1994).
[5] J. Aaviksoo, J. Kuhl, and K. Ploog, Phys. Rev. A **44**, R5353 (1991).
[6] S. -H. Choi and U. L. Österberg, Phys. Rev. Lett. **92**, 193903 (2004).
[7] H. Jeong, A. M. C. Dawes, and D. J. Gauthier, Phys. Rev. Lett. **96**, 143901 (2006).
[8] D. Wei et al., Phys. Rev. Lett. **103**, 093602 (2009).
[9] J. F. Chen et al., Phys. Rev. Lett. **104**, 223602 (2010).
[10] J. F. Chen et al., J. Opt. **12**, 104010 (2010).
[11] L. J. Wang, A. Kuzmich, and A. Dogariu, Nature (London) **406**, 277 (2000).
[12] M. D. Stenner, D. J. Gauthier, and M. A. Neifeld, Nature (London) **425**, 695 (2003).
[13] S. E. Harris, Phys. Today **50**, 36 (1997).
[14] M. D. Eisaman et al., Nature (London) **438**, 837 (2005).
[15] A. M. Steinberg, P. G. Kwiat, and R. Y. Chiao, Phys. Rev. Lett. **71**, 708 (1993).
[16] R. Y. Chiao, Phys. Rev. A **48**, R34 (1993).
[17] A. M. Steinberg and R. Y. Chiao, Phys. Rev. A **49**, 2071 (1994).
[18] J. Marangos, Nature (London) **406**, 243 (2000).
[19] C. H. van der Wal et al., Science **301**, 196 (2003).
[20] A. Kuzmich et al., Nature (London) **423**, 731 (2003).
[21] J. F. Chen et al., Phys. Rev. Lett. **104**, 183604 (2010).
[22] S. Du et al., Opt. Lett. **33**, 2149 (2008).
[23] S. Du et al., Phys. Rev. Lett. **100**, 183603 (2008).
[24] P. Kolchin et al., Phys. Rev. Lett. **101**, 103601 (2008).
[25] J. F. Clauser, Phys. Rev. D **9**, 853 (1974).
[26] P. Grangier, G. Roger, and A. Aspect, Europhys. Lett. **1**, 173 (1986).