

Generation of Paired Photons with Controllable Waveforms

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We describe experiments and theory showing the generation of counterpropagating paired photons with coherence times of about 50 ns and waveforms that are controllable at a rudimentary level. Using cw lasers, electromagnetically induced transparency and cold ^{87}Rb atoms we generate paired photons into opposing single-mode optical fibers at a rate of $\sim 12\,000$ pairs per second.

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For many years, spontaneous parametric down-conversion in nonlinear crystals has become the near-standard method for generating correlated and entangled photon pairs [1]. These paired photons are routinely used in areas such as quantum measurement, imaging, and information transfer [2]. The existing sources of paired photons have limitations for certain applications: (a) their linewidth is too broad, and their spectral brightness is too low to allow excitation of atomic species; (b) because their coherence time is in the subpicosecond range, photon waveforms are not resolvable by existing photodetectors; and (c) their short coherence length, $\sim 100\ \mu\text{m}$, is prohibitively small for long distance quantum communication.

In this Letter, following the field-opening papers of the Lukin [3] and Kimble [4] groups, we describe a source of paired photons that decisively overcomes the aforementioned limitations. This source makes use of slow light with a variable group velocity thereby allowing the control of the width, and to some extent, of the shape of the quantum wave packet. We describe experimental results showing the generation of paired photons into opposing single-mode fibers at a rate of 12 000 pairs/sec with a controllable wave packet width in the range of 50 ns.

To motivate what follows, we first consider an ideal nondegenerate parametric down-converter where pump photons are spontaneously converted to signal and idler photons and where the signal propagates with a group velocity V_g that is much slower than that of the idler. Figure 1 shows the analytical solution for the normalized intensity correlation function $g^{(2)}$ of the signal with the idler. Because paired signal and idler photons have an equal probability of being generated anywhere in the crystal of length L , the correlation function is rectangular with a temporal width that is equal to the group delay time L/V_g . With R defined as the paired generation rate (photons/sec), the height of the correlation function is $V_g/(RL)$ and is equal to the ratio of the average time between emitted photons to the length of the photon wave packet of a single pair of photons. If the group velocity is varied, the width and height of $g^{(2)}$ vary accordingly. The two-photon waveform is determined by $g^{(2)}$. For example, if the paired photons are used in a sum-frequency

experiment and are focused to an area A , the power per area of both photons is determined by the length of the generating crystal and is $\hbar\omega V_g/(LA)$.

The seminal paper by Duan *et al.* [5] described the entanglement of distant atomic ensembles and thereby motivated the search for methods for generating narrow-band correlated photons. van der Wal *et al.* have demonstrated both atomic memory and correlation between classical fields [3]. Kuzmich *et al.* have used the detected Stokes photon to time the generation of an anti-Stokes photon, thereby generating nonclassical photon pairs [4]. Braje *et al.* have reported electromagnetically induced transparency (EIT)-based classical field backward-wave generation [6]. Further connections to earlier work include the demonstration of interference of quantum wave packets [7], the effect of dispersion on these wave packets [8], the use of EIT for nonlinear optical processes [9], the observation of slow light [10], the experiments by Scully and co-workers on a backward-wave EIT process, and the theoretical studies of photon correlation [11]. Recently, Chou *et al.* and Eisaman *et al.* have demonstrated single photon generation [12] and have shaped few-photon light pulses [13]. There also exist techniques for combining resonators with crystal-based parametric oscillators to produce photons of narrow bandwidth [14].

Figure 2 shows a schematic of the process considered here. In the presence of two counterpropagating cw pump

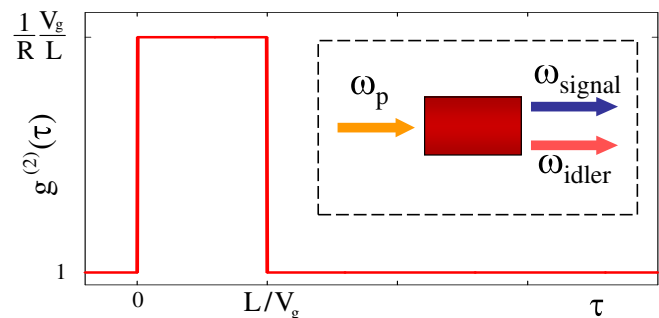


FIG. 1 (color online). Intensity correlation function for a spontaneous down-converter with a relative group delay L/V_g and paired emission rate R .

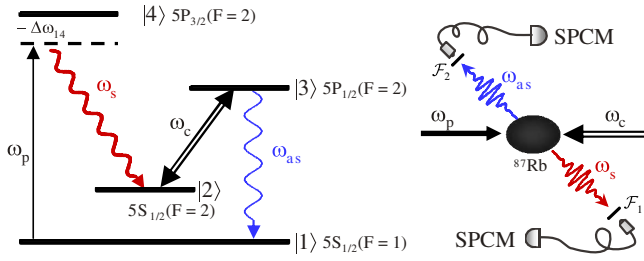


FIG. 2 (color online). In the presence of strong pump and coupling lasers, phase-matched, counterpropagating Stokes and anti-Stokes photons are generated into opposing single-mode fibers and detected by single photon-counting modules (SPCM). Filters \mathcal{F}_1 and \mathcal{F}_2 are narrow-band interference filters at 780 and 795 nm, respectively.

and coupling lasers with angular frequencies ω_p and ω_c , paired spontaneous Stokes and anti-Stokes photons are generated and propagate in opposite directions. These frequencies satisfy $\omega_s + \omega_{as} = \omega_p + \omega_c$. The wavelengths of the Stokes and anti-Stokes photons are 780 and 795 nm, respectively. The anti-Stokes photon has a center frequency equal to that of the highly absorbing $|1\rangle \rightarrow |3\rangle$ transition. The coupling laser creates a transparency window and causes the anti-Stokes wave to travel at a slow and variable group velocity V_g . This group velocity determines the width of the paired photon wave packet. The dominant process of Fig. 2 is the phase-matched, backward-wave parametric interaction between the Stokes and anti-Stokes waves. The complexity of this interaction is increased by the presence of Raman gain and scattering at the Stokes frequency and by EIT and slow light at the anti-Stokes frequency.

Working in the Heisenberg picture, the slowly varying envelope equations for the frequency domain operators $a_s^\dagger(-\omega)$ and $a_{as}(\omega)$ are

$$\begin{aligned} \frac{\partial a_s^\dagger}{\partial z} + g_R a_s^\dagger + \kappa_s a_{as} &= F_s, \\ -\frac{\partial a_{as}}{\partial z} + \Gamma a_{as} + \kappa_{as} a_s^\dagger &= F_{as}, \end{aligned} \quad (1a)$$

where $F_s = \xi_s f_{12} + \eta_s f_{13}$ and $F_{as} = \xi_{as} f_{12} + \eta_{as} f_{13}$ are the commutator preserving Langevin operators and $\langle f_{ij}(\omega, z) f_{ij}^\dagger(\omega', z') \rangle = (1/2\pi) \delta(\omega - \omega') \delta(z - z')$. In Eq. (1a), $\Gamma(\omega)$ is the EIT profile, $g_R(\omega)$ is the (complex) Raman gain, and $\kappa_s(\omega)$ and $\kappa_{as}(\omega)$ are the parametric coupling constants. We calculate these coefficients using the assumption that the atomic population remains in the ground state. With ω as the detuning of the anti-Stokes frequency from the $|1\rangle \rightarrow |3\rangle$ transition, $\Delta\omega_{14}$ as the detuning of the pump laser from $|1\rangle \rightarrow |4\rangle$ transition, dephasing rates γ_{12} and γ_{13} , atom density N , an absorption cross section σ for all allowed transitions, and the definitions $\tilde{\omega}_{12} = \omega + i\gamma_{12}$, $\tilde{\omega}_{13} = \omega + i\gamma_{13}$, and $D(\omega) = |\Omega_c|^2 -$

$4\tilde{\omega}_{12}\tilde{\omega}_{13}$, the quantities in Eq. (1a) are

$$\begin{aligned} \Gamma &= -\frac{2iN\sigma\gamma_{13}\tilde{\omega}_{12}}{D(\omega)}, & g_R &= -\left(\frac{\Omega_p^2\tilde{\omega}_{13}}{4\Delta\omega_{14}^2\tilde{\omega}_{12}}\right)\Gamma, \\ \kappa_s &= -\kappa_{as} = \left(\frac{\Omega_p}{2\Delta\omega_{14}}\right)\frac{iN\sigma\gamma_{13}\Omega_c}{D(\omega)}, \\ \xi_s &= \left(\frac{\Omega_p}{\Delta\omega_{14}}\right)\frac{2\tilde{\omega}_{13}\sqrt{N\sigma\gamma_{13}\gamma_{12}}}{D(\omega)}, & (1b) \\ \eta_s &= -\left(\frac{\Omega_c}{2\tilde{\omega}_{13}}\right)\sqrt{\frac{\gamma_{13}}{\gamma_{12}}}\xi_s, & \xi_{as} &= -\frac{2\Omega_c\sqrt{N\sigma\gamma_{13}\gamma_{12}}}{D(\omega)}, \\ \eta_{as} &= -\sqrt{\frac{\gamma_{13}}{\gamma_{12}}}\left(\frac{2\tilde{\omega}_{12}}{\Omega_c}\right)\xi_{as}. \end{aligned}$$

We first estimate the expected rate of paired counts by setting the Langevin terms in Eq. (1a) equal to zero, neglecting the Raman term g_R , and assuming that the optical depth $N\sigma L$ is sufficiently large such that the anti-Stokes counts are generated near the line center of the EIT profile. When this is the case, we may approximate the EIT profile as $\Gamma(\omega) = -i\omega/V_g$. The solution of Eq. (1a) is then written as a linear combination of the boundary conditions $a_s^\dagger(z=0)$ and $a_{as}^\dagger(z=L)$. The quantities of interest, both here and later, are obtained as expected values of product (boundary) vacuum states. The single transverse mode spectral generation rate $S(\omega)$ and the spectrally integrated generation rate $S = \frac{1}{2\pi} \int S(\omega) d\omega$ are

$$S(\omega) = |\kappa|^2 L^2 \text{sinc}^2\left(\frac{\omega L}{2V_g}\right), \quad S = V_g |\kappa|^2 L. \quad (2)$$

For the parameters of this work Eq. (2) predicts $c/V_g = 3500$ and a paired count rate of 2.6×10^5 pairs/sec.

Analogous to parametric down-conversion in crystals, the spectral generation rate $S(\omega)$ scales as $(NL)^2$, while the total rate of paired counts scales linearly as NL . The Langevin noise terms in Eq. (1a) result in additional unpaired Stokes counts. Because of the strong approximation that all atoms are initially in the ground state, no unpaired anti-Stokes counts are predicted.

Paired photon generation is achieved in a gas of cold ^{87}Rb atoms which are cooled and trapped in a dark magneto-optical trap in the $5S_{1/2}(F=1)$ level [6]. The optical depth $N\sigma L$ is varied from 1 to 11 by changing the detuning of the trapping laser from the $5P_{3/2}(F'=3)$ level. The photon-counting experiment is performed in the presence of the trapping, magnetic quadrupole field in a $500 \mu\text{s}$ window $10 \mu\text{s}$ after shutting off the trapping beams. By turning on the trapping and repumping lasers before the cold atoms have expanded out of the trapping region, the atom cloud is quickly recycled. This procedure allows a duty cycle (experimental time to trapping time) of up to 30%. Electromagnetically induced transparency is obtained at the anti-Stokes wavelength by using pump and coupling lasers with opposite circular polarizations.

The pump laser is frequency stabilized $7.5\gamma_{13}$ below the $5S_{1/2}(F=1) \rightarrow 5P_{3/2}(F'=2)$ transition of ^{87}Rb . The 680 nW laser is focused to a $1/e^2$ diameter of $280 \mu\text{m}$ with a Rabi frequency of $\Omega_p = 0.8\gamma_{13}$. The coupling laser, locked on the $5S_{1/2}(F=2) \rightarrow 5P_{1/2}(F'=2)$ transition, has a $1/e^2$ diameter of $950 \mu\text{m}$ in the atom cloud with a Rabi frequency variable from 4 to $30\gamma_{13}$.

As shown in Fig. 2, anti-Stokes and Stokes photons are counterpropagating through two single-mode fibers separated by 0.8 m. These fibers are aligned to each other through the atom cloud with a measured fiber-coupling efficiency of 80%. We choose the angle between the Stokes/anti-Stokes axis and the axis defined by the pump and coupling lasers to be $\sim 2^\circ$, which provides spatial separation between the weak anti-Stokes and Stokes photons from the two strong beams. The phase matching of these lasers is discussed in detail in our earlier work [6]. Narrow-band, interference filters \mathcal{F}_1 and \mathcal{F}_2 are used to reduce the uncorrelated noise. The single-mode fibers are connected to photon-counting modules (Perkin-Elmer SPCM-AQR-13). A time-to-digital converter (FAST Comtec TDC 7886S) measures the time between each “start” Stokes photon and “stop” anti-Stokes photon. Correlation statistics are binned into 128-bin histograms with a bin width of 1 ns.

We turn now to the main work of this Letter: the discussion and the measurement of the shape of the (non-normalized) Glauber two-photon correlation function:

$$G^{(2)}(\tau) = \langle a_s^\dagger(t) a_{as}^\dagger(t+\tau) a_{as}(t+\tau) a_s(t) \rangle. \quad (3)$$

The shape of $G^{(2)}(\tau)$ is expected to depend on the relation of two characteristic times that are the essence of this

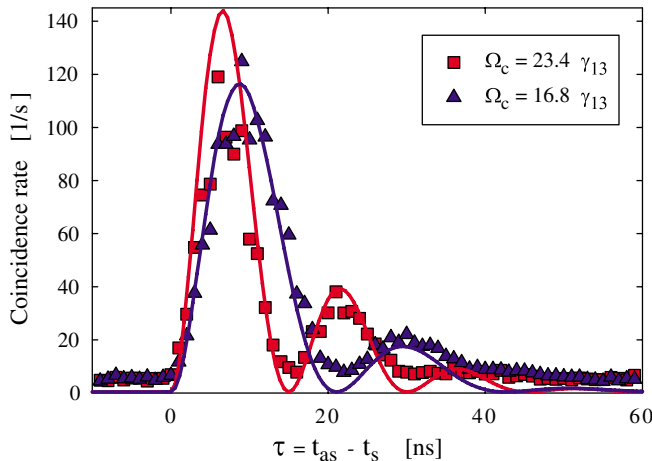


FIG. 3 (color online). Correlation function in the oscillatory regime. Coincidence rate $R_c(\tau)$ as a function of the time difference between the detection of anti-Stokes and Stokes photons is shown at $N\sigma L = 11$. Data were collected over 200 sec with a duty cycle of 10% and a bin size of 1 ns. Theoretical curves (solid lines) have been scaled vertically by a common factor.

problem. The first of these times is a Rabi-like time $t_r = (\Omega_c^2 - \gamma_{13}^2)^{-1/2}$, and the second is the group delay time of the medium $t_g = L/V_g$.

When $t_g < t_r$ and $\Omega_c > \gamma_{13}$, then $G^{(2)}(\tau)$ is oscillatory and damped [15]. The experimental results in this regime are shown in Fig. 3. One observes behavior like that of a single atom, where at $t = 0$ a Stokes photon causes the excitation of state $|2\rangle$, and the probability amplitude oscillates between states $|2\rangle$ and $|3\rangle$ as the atom decays. In Fig. 3 the optical depth is fixed at $N\sigma L = 11$ and Ω_c is varied. The squares and triangles are the coincidence rates $R_c(\tau)$ acquired over 200 sec and corrected for the 10% duty cycle. Statistical uncertainty of the peak coincidence rates is less than 3%. The theoretical curves (solid lines) are obtained by numerically solving Eq. (1a) with all terms retained and then evaluating $G^{(2)}(\tau)$ as given in Eq. (3). The parameters for the theoretical curves are $N\sigma L = 11$, $\Omega_p = 0.8\gamma_{13}$, and $\Delta\omega_{14} = 7.5\gamma_{13}$. All of the theoretical curves are vertically scaled by a factor of 0.009; this factor is the product of a detection efficiency and a theory-experiment discrepancy of 0.10.

When the group delay is sufficiently large such that $t_g > t_r$, $G^{(2)}(\tau)$ approximates the correlation function of the ideal parametric emitter of Fig. 1, and the width of the wave packet is approximately t_g . Figure 4 shows experiment and theory in this limit where the correlation width is set by the variable group delay. As in Fig. 3, the optical depth is fixed at $N\sigma L = 11$, and the group delay is varied by changing Ω_c . In the first experimental curve (squares), corresponding to $\Omega_c = 8.4\gamma_{13}$, the width of the correlation function is ~ 20 ns. By increasing the group delay, the peak height of the coincidence curve decreases; this trend can be seen in the subsequent data curves. The theoretical curves

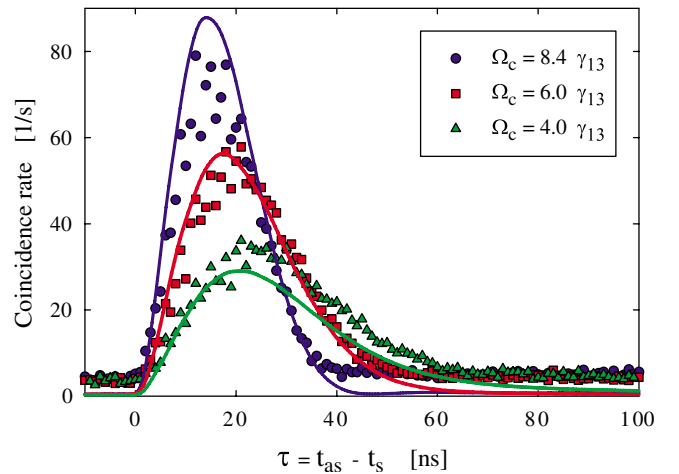


FIG. 4 (color online). Correlation function in the group delay regime. Coincidence rate $R_c(\tau)$ as a function of the time difference between the detection of anti-Stokes and Stokes photons is shown at $N\sigma L = 11$. Data were collected over 200 sec, and the bin size is 1 ns. Theoretical curves are vertically scaled.

are vertically scaled by a factor of 0.013; here the theory-experiment discrepancy is 0.14.

In any EIT process, the maximum group delay that may be observed is limited by the linewidth of the $|1\rangle \rightarrow |2\rangle$ transition [16]. Here, we estimate this linewidth from a least-squares fit of the observed EIT profiles at small Ω_c . This yields an effective dephasing rate of $\gamma_{12} = 0.6\gamma_{13}$. The maximum delay and thus the maximum paired photon pulse width is, therefore, $1/(2\gamma_{12}) = 45$ ns. This maximum delay is in reasonable agreement with the $\Omega_c = 4.0\gamma_{13}$ curve of Fig. 4.

We next compare the experimental results and the predictions of Eq. (1a). The agreement between theory and experiment describing the shapes of the non-normalized $G^{(2)}(\tau)$ is good over our parameter range. The generation rate for our paired counts is estimated from correlation curves by subtracting the uncorrelated flat background and measuring the area. Taking into account detection efficiencies, we typically generate paired photons at a rate of $\sim 12\,000$ s $^{-1}$, which is low as compared to theory by a factor of ~ 8 . The measured conversion efficiency between a weak input Stokes beam and the generated anti-Stokes beam is also low by this same factor. We believe that this discrepancy is the result of our use of focused (nonplanar) pump and coupling laser beams.

The theoretical ratio of anti-Stokes counts to Stokes counts depends on the optical depth and for very large $N\sigma L$ approaches unity. For the parameters of this work, we calculate an anti-Stokes to Stokes count ratio of 0.58; however, we find experimentally that the total anti-Stokes rate is approximately equal to the observed Stokes rate. It appears that our anti-Stokes counts are of two types: paired counts that comprise about 6.7% of the total anti-Stokes counts and occur with a delay of less than 50 ns and nonpaired anti-Stokes counts that occur over much longer times. We suspect, but have no evidence, that the unpaired anti-Stokes counts result from the dephased $|1\rangle \rightarrow |2\rangle$ transition atoms that must ultimately return to the ground level if the system is to remain in steady state.

When the bin width ΔT is small as compared to the temporal correlation time, $G^{(2)}$ is related to the measured coincidence rates of Figs. 3 and 4 by $G^{(2)}(\tau) \approx R_c(\tau)/(\epsilon^2\Delta T)$. In this experiment $\Delta T = 1$ ns, and the efficiency factor $\epsilon = 0.3$. This factor accounts for the photon counter efficiency of 50%, the filter transmission of 75%, and the fiber coupling of 80%. With R_s and R_{as} defined as the similarly corrected Stokes and anti-Stokes rates, the normalized intensity correlation function is then $g^{(2)}(\tau) = G^{(2)}(\tau)/(R_s R_{as})$. We thereby estimate $G^{(2)}(\tau) = 6.6 \times 10^{11}$ s $^{-2}$ and for $R_s = R_{as} = 180\,000$ s $^{-1}$, $g^{(2)}(\tau) = 20$. For a classical light source, the cross correlation is bounded by the product of autocorrelations as described by the Cauchy-Schwartz inequality [17]: $[g_{s,as}^{(2)}(\tau)]^2 / (g_{s,s}^{(2)} g_{as,as}^{(2)}) \leq 1$. The self-correlations $g_{s,s}^{(2)}$

$g_{as,as}^{(2)}$ were each measured using a fiber beam splitter (not shown in Fig. 2). Both self-correlations were equal to 1.0 ± 0.1 . The Cauchy-Schwartz inequality is therefore violated by a factor of ~ 400 .

This work has described experiments and theory showing the generation of oppositely propagating paired photons with coherence times of ~ 50 ns and waveforms that are electronically controllable at a rudimentary level. It is expected that these nondegenerate paired photons will exhibit time-frequency entanglement. Operation with appropriately polarized pump and coupling laser beams will result in polarization entanglement of the paired photons. By detuning the pumping laser, the frequencies of the Stokes and anti-Stokes photons may be made the same. Because they are narrow band, paired photons that are generated by this technique should be useful for transferring correlated and entangled momenta to cold atoms.

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