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Reversing the Temporal Envelope of a Heralded Single Photon using a Cavity

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We demonstrate a way to prepare single photons with a temporal envelope that resembles the time reversal of photons from the spontaneous decay process. We use the photon pairs generated from a time-ordered cascade decay: the detection of the first photon of the cascade is used as a herald for the ground-state transition resonant second photon. We show how the interaction of the heralding photon with an asymmetric Fabry-Perot cavity reverses the temporal shape of its twin photon from a decaying to a rising exponential envelope. This single photon is expected to be ideal for interacting with two-level systems.

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The absorption of a single photon by a single atom or an ensemble of atoms is an interesting problem from a fundamental point of view and is also essential for many quantum information protocols [1–4]. One of the requirements for an efficient absorption is that the temporal shape of the incident photon is the time reversal of the photon from the spontaneous decay process [5,6]. Temporally shaped light pulses have been utilized in many recent experiments to achieve efficient interactions between light and matter [7,8]. In particular, the advantage of using a rising exponential shaped single photon for absorption in an atomic ensemble was demonstrated in Ref. [9], and shaped multiphoton pulses for exciting a single atom were demonstrated in Ref. [10]. This advantage also applies to interacting single photons with other systems such as quantum dots [11,12], single molecules [13], and superconducting circuits [14].

The efficient preparation of single photons with narrow bandwidth and a rising exponential envelope is not trivial. One solution is the direct modulation of a heralded photon generated by an atomic medium [15]. This technique results in unavoidable losses due to filtering. We have previously demonstrated a scheme to generate single photons with a rising exponential shape by heralding on photon pairs produced by cascade decay [16] without filtering. The drawback of this scheme is that the photon with the rising exponential envelope is not resonant with an atomic ground-state transition.

In this Letter, we combine the asymmetric cavity design used by Bader *et al.* [17] with the well-known temporal correlation properties of photon pairs [18] to invert the temporal envelope of the generated photon pairs: with the proper heralding sequence we obtain a rising exponential single photon resonant with a ground-state transition of ^{87}Rb. This concept is not limited to atoms, but can be equally applied to other physical system with a cascade level structure to obtain such photons [19–21]. A related idea has been used in the past for nonlocal dispersion cancellation [22].

The photons emerging from an atomic cascade decay have a well-defined time order. The first photon of the cascade (signal) is generated before the photon resonant with the ground state (idler). The resulting state can be described by a two-photon wave function [23] of the form

$$\psi(t_s, t_i) = A e^{-(t_i - t_s)/2\tau}\Theta(t_i - t_s), \tag{1}$$

where t_s, t_i are the detection times of the signal and idler photons and Θ is the Heaviside step function. In this notation, the probability of observing a pair is proportional to $|\psi(t_s, t_i)|^2$. The exponential envelope and the decay time τ is a consequence of the atomic evolution of the cascade decay. If the detection of a signal photon is used as a herald, the idler mode has a single photon state with an exponentially decaying temporal envelope starting at $t_i = t_s$. Similarly, if the detection of an idler photon acts as a herald, the signal photon has an exponentially rising temporal envelope.

An asymmetric cavity with the appropriate parameters transforms a light field with an exponentially rising envelope into one with a decaying envelope [17]. The main point of this work is that coupling the signal mode into a properly tuned asymmetric cavity before heralding results in a “time reversed” envelope for the idler photon.

The asymmetric cavity is formed by a partially reflective mirror M_1 and a highly reflective mirror M_2; see Fig. 1. The effect of the cavity on the signal mode is described as a frequency-dependent phase factor [24,25],

$$C(\delta') = \frac{\sqrt{R_1} - \sqrt{R_2}e^{i\delta'/\Delta\nu_f}}{1 - \sqrt{R_1 R_2}e^{i\delta'/\Delta\nu_f}}, \tag{2}$$

where $R_{1,2}$ are the reflectivities of M_1 and M_2, $\Delta\nu_f$ is the free spectral range of the cavity, and δ' is the detuning from

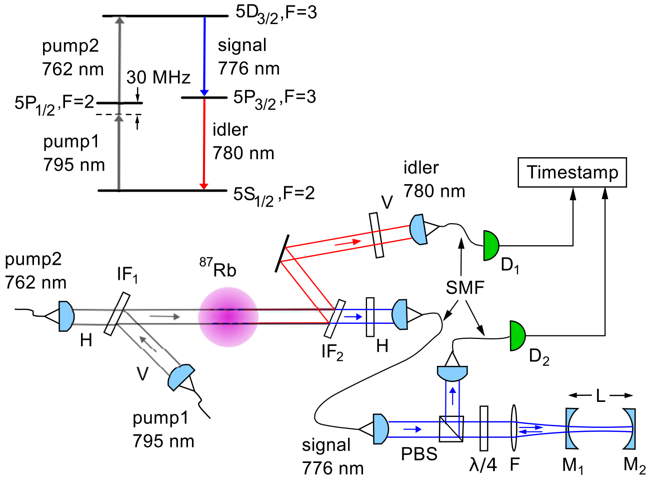

FIG. 1 (color online). Schematic of the four-wave mixing experiment in collinear geometry. IF$_1$, IF$_2$: interference filters, used to combine pump beams and to separate the photons pairs. SMF: Single-mode optical fibers. M_1, M_2: cavity mirrors. The incoming and outgoing modes of the cavity are separated by a polarizing beam splitter (PBS) and a quarter wave plate ($\lambda/4$). D_1, D_2: silicon avalanche photodiodes (APD). The inset shows the cascade level scheme for the generation of photon pairs in ^{87}Rb.

the cavity resonance. For $R_2 = 1$, the transformation of the incoming mode is lossless, i.e., $|C(\delta')| = 1$.

The cavity transforms the two-photon wave function in Eq. (1) into the two-photon wave function $\tilde{\psi}(t_s, t_i)$,

$$\tilde{\psi}(t_s, t_i) = \mathcal{F}_s^{-1}[C(\omega_s - \omega_s^0 - \delta)\mathcal{F}_s[\psi(t_s, t_i)]], \qquad (3)$$

where $\mathcal{F}_s$ denotes a Fourier transform from t_s to ω_s, and δ is the detuning of the cavity resonance from the signal photon center frequency $\omega_s^0/2\pi$.

If the ring-down time of the cavity matches the coherence time τ of the photon pair in Eq. (1), the resulting wave function is

$$\tilde{\psi}(t_s, t_i) = \frac{A}{\sqrt{1 + 4\delta^2\tau^2}}[2\delta\tau e^{-(t_i - t_s)/2\tau}\Theta(t_i - t_s) + e^{(t_i - t_s)/2\tau}\Theta(-t_i + t_s)] \qquad (4)$$

with an exponentially rising and an exponentially decaying component. Their relative weight can be controlled by the detuning δ, and for $\delta = 0$, a time-reversed version of Eq. (1) is obtained,

$$\tilde{\psi}(t_s, t_i) = A e^{(t_i - t_s)/2\tau}\Theta(-t_i + t_s). \qquad (5)$$

Heralding on the detection of the modified signal photon results in an idler photon state with a rising exponential envelope, ending at $t_i = t_s$. The cavity thus effects a reversal of the temporal envelope of the heralded idler photons from an exponential decay to a rise.

To experimentally investigate this method, we used the setup shown in Fig. 1. We generate time-ordered photon pairs by four-wave mixing in a cold ensemble of ^{87}Rb atoms in a cascade level scheme. Pump beams at 795 and 762 nm excite atoms from the $5S_{1/2}, F = 2$ ground level to the $5D_{3/2}, F = 3$ level via a two-photon transition. The 776 nm (signal) and 780 nm (idler) photon pairs emerge from a cascade decay back to the ground level and are coupled to single mode fibers. All four modes are collinear and propagate in the same direction. The coherence time τ of the photon pairs is determined by a time-resolved coincidence measurement between the detection of signal and idler photons to be 5.9 ns. Details about the photon pair source can be found in Refs. [16] and [26].

One of the modes of the photon pairs (signal in Fig. 1) is coupled to the fundamental transverse mode of an asymmetric cavity, formed by mirrors M_1, M_2 with radii of curvature of 100 and 200 mm, respectively. We characterize the cavity using a frequency-stabilized laser of wavelength 776 nm. The reflectivity of mirror M_1 is determined by direct measurement with a *p-i-n* photodiode to be $R_1 = 0.9410 \pm 0.0008$. Transmission through the mirror M_2 and absorption by the mirrors leads to losses in the cavity. The loss per round trip is determined from the transmission through and the reflection from the cavity and is included in the reflectivity of M_2, $R_2 = 0.998 \pm 0.001$. The mirrors are mounted on a 55 mm long fused silica spacer, corresponding to a free spectral range $\Delta\nu_f = 2.7$ GHz. Therefore, an incident photon of Fourier bandwidth $1/(2\pi\tau) = 27$ MHz interacts effectively with only one longitudinal mode of the cavity, ensuring that Eq. (2) is an adequate model. Temperature control of the spacer allows precise tuning and stabilization of the resonance frequency of the cavity. The light reflected off the cavity is separated from the incident mode by using a PBS and a quarter wave plate ($\lambda/4$).

We infer the temporal shape of the heralded photons from the time distribution of the coincidence rate $G_{si}^{(2)}$ between the APDs D_1 and D_2 (time resolution < 1 ns). In Fig. 2 we show $G_{si}^{(2)}$ for three different cavity-photon detunings. When the cavity resonance is tuned far away from the signal photon frequency ω_s^0, in this case about $\delta/2\pi = 120$ MHz, the temporal envelope remains nearly unchanged from the exponential decay obtained without cavity. Off-resonant coupling of the incident signal photon to the cavity leads to the residual coincidences at times $t_i - t_s < 0$. At $\delta/2\pi = 27$ MHz, the time distribution becomes a symmetric exponential, and on resonance, $\delta/2\pi = 0$, we obtain a rising exponential shape. For the three detunings the measurement agrees with the shape expected from Eq. (4): the exponential time constants remain unchanged and the new temporal shapes are determined by the phase shift across the cavity resonance via Eq. (2).

From the time distribution of the coincidence counts, it is evident that the situation is symmetrical to what we

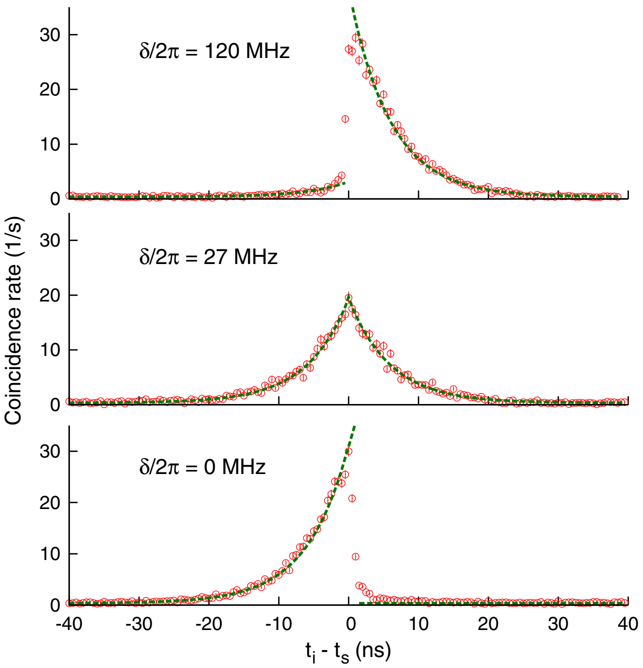

FIG. 2 (color online). Transformation of the temporal envelope of the heralded idler photon from exponential decay to rise when the cavity is in the signal mode. The y axis shows the coincidence rate $G_{si}^{(2)}$ between the detectors D_1 and D_2 as a function of the detection time difference. The dashed lines represent $|\tilde{\psi}(t_s, t_i)|^2$, obtained from the model described by Eq. (4) for the indicated cavity detunings δ, with amplitude A as the only free parameter used to fit the experimental points.

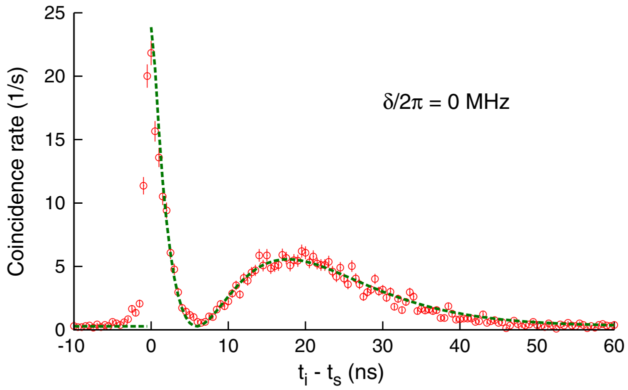

FIG. 3 (color online). Temporal envelope of the idler photon when the cavity is aligned and tuned to resonance with the idler mode. The dashed line represents $|\tilde{\psi}(t_s, t_i)|^2$ obtained from Eq. (3) by swapping i and s. Also in this case, the amplitude is the only free parameter used in the fit.

presented in Ref. [16]: by heralding on the signal photon, we now obtain an idler photon with a rising exponential temporal envelope. This result, though predicted by the theory, is particularly exciting: the idler photon is resonant with a ground-state transition and the obtained temporal envelope is similar to the time reversal of the one obtained by spontaneous decay. The only deviation from the predicted shape occurs for a short time interval after the detection of the herald ($t_i - t_s > 0$). We attribute this deviation to an imperfect matching between the signal and cavity spatial modes.

To confirm the predictive power of our model, we repeated the same experiment swapping the roles of the signal and idler modes. This corresponds to swapping the subscripts s and i in Eqs. (3) and (4). Figure 3 shows the time-resolved coincidence rate $G_{si}^{(2)}$ between the signal and modified idler photons with the cavity tuned to resonance with the idler central frequency. In this case, the cavity transforms the exponentially rising temporal envelope into a more complex shape. Our model accurately describes this complex shape, as can be seen from the dashed line in Fig. 3.

Using the same setup, we can infer the population of the cavity mode as a function of time and observe its dependence on the envelope of the incident photon. We estimate the mean photon number $\langle n(t)\rangle$ in the cavity using an algorithm similar to the one presented in Ref. [17]. We compare the time distributions of coincidence rates $G_{\rm fr}^{(2)}$ and $G_{\rm or}^{(2)}$ when the cavity is tuned far-off resonance ($\delta/2\pi = 200$ MHz) and on resonance ($\delta/2\pi = 0$ MHz) with the incident photons, normalized against the total far-off resonance coincidences,

$$\langle n(t)\rangle = \frac{e^{-\eta t\Delta\nu_f}\int_{-\infty}^{t}[G_{\rm fr}^{(2)}(t') - G_{\rm or}^{(2)}(t')]e^{\eta t'\Delta\nu_f}dt'}{\int_{-\infty}^{\infty}G_{\rm fr}^{(2)}(t')dt'}. \qquad (6)$$

We estimated η, which includes the cavity losses per round trip and transmission through M_2, to be 0.002 ± 0.001.

When the cavity is exposed to the idler mode, a heralded single photon with decaying exponential envelope interacts with the cavity: the mean photon number in the cavity

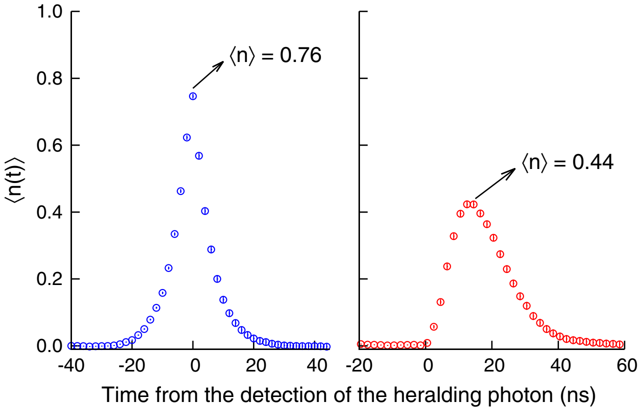

FIG. 4 (color online). Mean photonnumber in the cavity estimated using Eq. (6). On the left, the detection of an idler photon is used as a herald, and the cavity is in the signal mode. In this case, we observe the interaction of an exponentially rising waveform with the cavity. On the right, the roles of signal and idler are swapped, and the cavity interacts with an exponentially decaying incident photon.

reaches a maximum of 0.44 ± 0.01. On the other hand, when the cavity is aligned in the signal mode we have a heralded single photon with an increasing exponential envelope interacting with the cavity; in this case, $\langle n(t) \rangle$ reaches a maximum of 0.76 ± 0.01. As expected, the photon with the rising exponential waveform interacts more efficiently with the cavity. Following the analogy in Ref. [27], we expect this result to be extended to the probability of absorption of a single photon by a single atom. In the case of interaction with a single atom, it will be necessary to also match the bandwidth of the transition. We have already demonstrated how it is possible to control the bandwidth of the photon generated by the cascade process by adjusting the optical density of the atomic medium [26].

In summary, we have demonstrated a method to transform a heralded single photon with a decaying exponential temporal envelope to a rising exponential envelope using a cavity. Using this method, we obtain single photons that resemble the time-reversed versions of photons from spontaneous decay process resonant to the $D2$ line of ^{87}Rb atoms. Single photon states of this envelope and bandwidth would be useful for transferring information from photons back into atoms. As this time-reversal technique can be used with photon pairs from other sources with time-ordered emission, as found, e.g., in molecules and quantum dots, it completes the toolbox necessary to interconnect stationary qubits in a complex quantum information processing scenario.

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Note added in proof.—We became aware that during the review process a similar work independently performed by Liu *et al.* was published [28].

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