Direct generation of an exponentially rising single photon field from parametric conversion in atoms

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We prepare heralded single photons from a photon pair source based on non-degenerate four-wave mixing in a cold atomic ensemble via a cascade decay scheme. Their statistics shows strong anti-bunching with $g^{(2)}(0) < 0.03$, indicating a near single photon character. In an optical homodyne experiment, we directly measure the temporal envelope of these photons and find, depending on the heralding scheme, an exponentially decaying or rising profile. Such a rising envelope is required for the efficient interaction between a single photon and a two level system. At the same time, their observation illustrates the breakdown of a realistic interpretation of the heralding process in terms of defining an initial condition of a physical system.

Strong interaction in free space between a single photon and a two-level quantum system is a prequisite for many quantum communication and computation protocols [1–4]. This interaction has been demonstrated to be optimal when the incident photon has an exponential rising temporal envelope [5–8].

A common way to obtain single photons is to generate time correlated photon pairs: the detection of one photon heralds the presence of the other [9–11]. Heralding has already been used to generate single photons with rising exponential temporal envelopes: the intensity of one photon of the pair is modulated in time using fast modulators [12, 13]. This modulation technique resulted in unavoidable losses due to the small overlap between the temporal shape of the generated photons (exponential decay) and the desired one (exponential rise).

In the work presented here, we use a photon pair source based on four-wave mixing in a cold atomic ensemble via a cascade decay level scheme. This process has been used in the past to generate narrow band photon pairs [14], and it has already been demonstrated that the resulting photon pairs are nearly Fourier-limited [15] with a coherence time long enough to be resolved with various optical detection techniques. We show how the use of a cascade decay scheme allows the direct preparation of single photons with a rising exponential temporal envelope by using the appropriate heralding scheme without resorting to any intensity modulation.

We initially demonstrate (see Fig. 1) the single photon character of our heralded photons. An ensemble of ^{87}Rb atoms is first cooled and confined in a magneto-optical trap (MOT) reaching an optical density of ≈ 32 on the $5S_{1/2}, F = 2 \rightarrow 5P_{3/2}, F = 3$ transition after about 12 ms. In the following 1 ms the MOT beams are switched off, and the atoms are excited to the $5D_{3/2}, F = 3$ level by two orthogonally linearly polarized pump beams (780 nm, 0.1 mW and 776 nm, 5 mW, beam waists 0.45 mm) intersecting at an angle of $0.5°$ in the cold atomic cloud. The 780 nm pump beam is red detuned by $\Delta_1 = 40$ MHz from
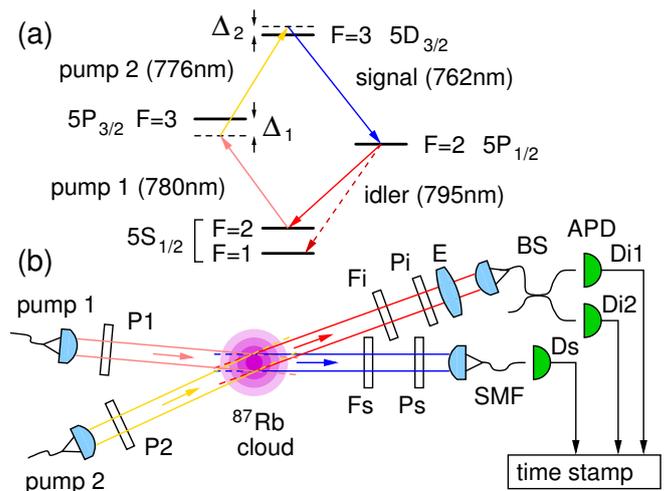
FIG. 1: (a) Cascade level scheme for four wave mixing in ^{87}Rb. (b) Setup of the heralded single photon source. Polarizers (P1,2, Pi, Ps) and interference filters (Fi, Fs) separate the signal and idler photons from residual pump light. An etalon (E) in the idler arm removes uncorrelated photons from a decay to the $5S_{1/2}, F = 1$ level. Signal photons detected by an avalanche photodiode (APD) trigger a Hanbury-Brown–Twiss measurement between idler photons distributed to two detectors, D_{i1} and D_{i2}, via a 50:50 fiber beam splitter (BS).

the intermediate level $5P_{3/2}, F = 3$ to reduce incoherent scattering. The combined detuning of both pumps from the two-photon transition ranges from $\Delta_2 = 0 - 6$ MHz to the blue. Photon pairs of wavelength 762 nm (signal) and 795 nm (idler) are generated by a cascade decay from the $5D_{3/2}, F = 3$ level via $5P_{1/2}, F = 2$ to $5S_{1/2}, F = 2$. We use interference filters and an etalon to filter any background light generated by other processes in the ensemble. Energy conservation and phase matching between pump and collection modes allow efficient coupling of photon pairs with a strong temporal correlation into single mode fibers.

Unlike single quantum emitters [16–18], the probability of generating more than one photon per heralding

event in a parametric process does not vanish due to the thermal nature of the emission process from the atomic ensemble [19]. We consider the second order correlation function $g^{(2)}(\Delta t_{12})$ for the probability of observing two photons in a given mode with a time difference Δt_{12}. Any classical light field exhibits $g^{(2)}(0) \geq 1$, while $g^{(2)}(\Delta t_{12}) < 1$ is referred to as photon antibunching, with an ideal single photon source reaching $g^{(2)}(0) = 0$ [20].

We determine this correlation function experimentally in a Hanbury-Brown–Twiss (HBT) geometry, where the idler light is distributed with a 50:50 fiber beam splitter onto two single photon counting silicon avalanche detectors (APD) D_{i1}, D_{i2} (quantum efficiency $\approx 40\%$, dark count rates 40 to $150\,\mathrm{s}^{-1}$), while signal photons are detected by D_s as heralds. The detection events are time-stamped with 125 ps resolution. The combined timing uncertainty of the photodetection process is $\approx 600\,\mathrm{ps}$.

From our previous characterization of the source [15], we know that the correlation function between the signal and idler $g_{si}^{(2)}(\Delta t_{si})$ has the shape of a decreasing exponential, with more than 98% of the coincidences occuring within a time window $T_c = 30\,\mathrm{ns}$. We record a histogram $G_{i1i2|s}^{(2)}(\Delta t_{12})$ of idler detection events on D_{i1} and D_{i2} with a time difference $\Delta t_{12} = t_2 - t_1$ if one of them occurs within a coincidence time window T_c after the detection of a heralding event in the signal mode. The normalized correlation function of heralded coincidences between the two idler modes is

$$g_{i1i2|s}^{(2)}(\Delta t_{12}) = G_{i1i2|s}^{(2)}(\Delta t_{12})/N_{i1i2|s}(\Delta t_{12}), \qquad (1)$$

where $N_{i1i2|s}(\Delta t_{12})$ is the estimated number of accidental coincidences. Due to the strong temporal correlation between signal and idler photons, the probability of accidental coincidences is not uniform. We thus estimate $N_{i1i2|s}(\Delta t_{12})$ for every Δt_{12} by integrating the time difference histograms between the signal and each arm of the HBT, $G_{si1}^{(2)}(\Delta t_{si})$ and $G_{si2}^{(2)}(\Delta t_{si})$ within T_c normalized to the total number of triggers N_s. Due to the time ordering of the cascade process, it is only meaningful to consider positive time delays after the detection of the heralding photon, thus splitting $N_{i1i2|s}$ into two cases. For $\Delta t_{12} \geq 0$, we use

$$N_{i1i2|s}^{(+)}(\Delta t_{12}) = \frac{1}{N_s} \int_0^{T_c} G_{si1}^{(2)}(\Delta t_{si}) \, G_{si2}^{(2)}(\Delta t_{si} + \Delta t_{12}) \; d\Delta t_{si} \qquad (2)$$

while for $\Delta t_{12} < 0$, we use

$$N_{i1i2|s}^{(-)}(\Delta t_{12}) = \frac{1}{N_s} \int_0^{T_c} G_{si1}^{(2)}(\Delta t_{si} + \Delta t_{12}) \, G_{si2}^{(2)}(\Delta t_{si}) \; d\Delta t_{si}. \qquad (3)$$

The resulting $g_{i1i2|s}^{(2)}(\Delta t_{12})$ is shown in Figure 2(a) as function of the delay Δt_{12}, sampled into 2 ns wide time
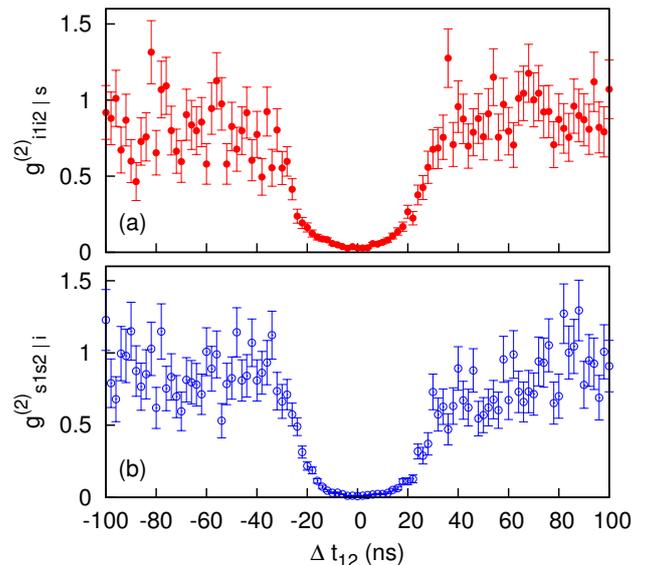
FIG. 2: (a) The correlation function $g_{i1i2|s}^{(2)}$ of idler photons separated by a time difference Δt_{12}, conditioned on detection of a heralding event in the signal mode, shows strong photon antibunching over a time scale of $\pm 20\,\mathrm{ns}$, indicating the single photon character of the heralded photons. The error bars indicate the propagated poissonian counting uncertainty from $G_{i1i2|s}^{(2)}$ and $N_{i1i2|s}$. (b) Same measurement, but with the signal and idler modes swapped.

bins. With a signal photon detection rate of $50000\,\mathrm{s}^{-1}$ (at $\Delta_2 = 0$), we observe $g_{i1i2|s}^{(2)}(0) = 0.032 \pm 0.004$.

When switching the roles of the signal and idler arms, the resulting normalized correlation function shown in Figure 2(b) has a minimum $g_{s1s2|i}^{(2)}$ of 0.018 ± 0.007 with an idler photon detection rate of $13000\,\mathrm{s}^{-1}$.

We now proceed to determine the temporal envelope of the heralded single photon fields by measuring the field quadrature in the time domain [21, 22] via a balanced homodyne detection. The experimental scheme is shown in Figure 3: the idler mode is mixed with a local oscillator (LO) which is frequency-stabilized to the idler transition $5S_{1/2}, F = 2 \rightarrow 5P_{1/2}, F = 2$. The balanced mixing is done with two polarizing beam splitters (PBS1,2) and a half-wave plate resulting in an interference visibility of $\approx 95\%$. The difference of photocurrents from pin silicon photodiodes (D+, D-; quantum efficiency $\approx 87\%$) is proportional to the optical field quadrature in the idler mode. With a LO power of $4.5\,\mathrm{mW}$, the electronic noise is about 6-20 dB below the shot noise limit over a band of $10\,\mathrm{kHz}$–$210\,\mathrm{MHz}$. We record the homodyne signal with a digital oscilloscope (analog bandwidth 1 GHz), with the click detection of the signal photon on the APD triggering the acquisition. We then calculate the variance of the optical field from 2.7×10^5 traces, normalized to the
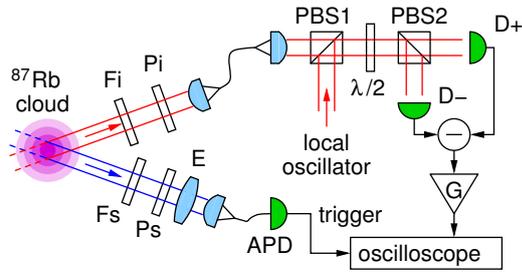
FIG. 3: Field measurement setup. One of the photons (idler in this figure) is combined with a coherent laser field as a local oscillator on a polarizing beam splitter (PBS1), and sent to a balanced pair of pin photodiodes D+, D- for a homodyne measurement. The photocurrent difference as a measure of the optical field strength is amplified and recorded with an oscilloscope on the arrival of a heralding event from the avalanche photodetector in the signal arm.

FIG. 4: Optical homodyne results. (a) Exponential decay of the field variance of heralded idler photons. (b) Exponential rise of the field variance of heralded signal photons

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shot noise, as a measure of the temporal envelope of the photon. We also switched the roles of the signal and idler modes for triggering and homodyne detection, this time using a local oscillator resonant with the transition $5P_{1/2}$, $F = 2 \rightarrow 5D_{3/2}$, $F = 3$ near 762 nm. The variance for this measurement is calculated from 5×10^5 traces. Both results are shown in Figure 4. In both configurations, we set $\Delta_2 \approx 6$ MHz to maximise the heralding efficiency.

In Figure 4(a) the normalized field variance of the idler photon suddenly rises about 4% above the shot noise level at the detection time of the trigger photon, and exponentially decays back to the shot noise level, with a time constant of $\tau_i = 7.2 \pm 0.2$ ns obtained from a fit. This can be easily understood by the timing sequence of a cascade decay, where the signal photon heralds the population of the intermediate level, which subsequently decays exponentially, leading to the characteristic decaying envelope of the idler photon according to the Weisskopf-Wigner solution [23, 24].

The probability of detecting an idler photon given the detection of a signal photon (heralding efficiency) was independently determined with two APDs to be $\eta_i \approx 13\%$ (uncorrected for APD efficiency and optical losses).

In Fig. 4(b) the normalized field variance of the signal photon exponentially increases a few 10 ns before the trigger event to a value of 1.06, and then quickly returns to the shot noise level, with a rise time constant $\tau_s = 7.4 \pm 0.2$ ns obtained from a fit. Here, the suppression of uncorrelated trigger (idler) photons by the etalon E (figure 1(b)) results in a higher heralding efficiency $\eta_s \approx 19\%$, and therefore a higher signal to shot noise level. The time constants for the exponential rise and decay profiles are shorter than the lifetime of the intermediate state. This is due to collective decay effects observed in dense atomic ensembles [25]. The measured value is compatible with our previous measurements of the distribution of detection time differences for this optical density [15].
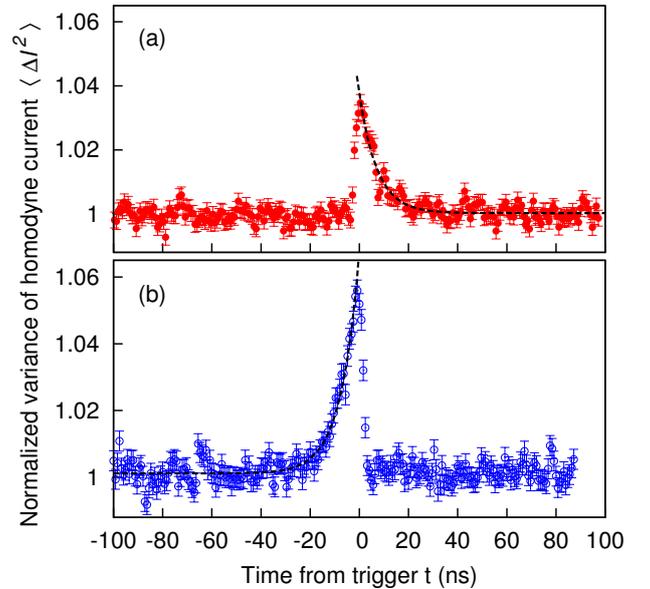
From the results of the HBT experiment, we know that the idler detection witnessed a single photon to a very good approximation. We therefore have to conclude that the heralded signal field is a single photon state with an exponentially rising temporal envelope as required for optimal absorption by a two-level system [5, 6].

In this case, however, the simple causal interpretation of the physical process in the Weisskopf-Wigner picture does not work: the trigger time is fixed by the herald that leaves the atoms in the ground state, but the signal field starts to rise to a maximum before that. So the heralding process does not set an initial condition of a physical system that then evolves forward in time, but marks the end of a (signal) field evolution that is compatible with the exponential rise that started *before* the heralding event. Formally this is not a problem, because the heralding event just sets a different boundary condition.

This experiment again highlights a problem with the definition of "real" physical quantities (in the spirit of an EPR definition [26]). The physical quantity here is the electrical field in the signal mode at any point in time. Nothing seems to set the initial condition leading to such an increase, with a dynamics governed by some laws of physics. Yet, when an idler event is registered in a photodetector, the recorded field is perfectly compatible with a single photon with an exponentially rising envelope. In this example, an interpretation that is more symmetric

between preparation and detection procedures [27], like the two-state vector formalism [28], may be adequate.

In summary, we have demonstrated a source of heralded single photons based on an ensemble of cold rubidium atoms. We observe antibunching with $g^{(2)}(0) < 0.03$, conditioned on the photon in the signal (idler) mode. Depending on which of the modes is chosen as a herald, we find either an exponentially decaying or rising temporal envelope of the heralded photon.

If heralded single photons are practically not distinguishable from "true" single photons, the latter should at least in principle be efficiently absorbed by a two-level system in free-space in a time-reversed Weisskopf-Wigner situation. Such an experiment also would provide a better understanding to what extent heralded photons are equivalent to single photons emerging from a setup with a well-defined initial condition.

This test would require a photon driving a ground state transition of a two level system. The heralded photons with the exponentially rising waveform generated by our scheme are resonant with an excited transition and therefore cannot be used directly. However, waveform reshaping techniques demonstrated in [8] can be employed to address this problem.

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