Counterintuitive temporal shape of single photons

Gurpreet Kaur Gulati,* Bharath Srivathsan,† Brenda Chng,‡ Alessandro Cerè,§ Dzmitry Matsukevich,∥ and Christian Kurtsiefer¶
(Dated: February 25, 2014)

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 antibunching 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. The rising envelope will be useful for efficient interaction between single photons and microscopic systems like single atoms and molecules. 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 atom-light interaction at the single quantum level is a prerequisite for many quantum communication and computation protocols [1, 4]. In free space the optimal coupling of photons to atoms requires the temporal profile of the incoming photons to match the time reversal of photons generated by spontaneous decay from the transition of interest [2–4]. Together with spectral and spatial mode matching [8–10], the efficient preparation of single photons with an exponentially rising temporal envelope remains an open challenge.

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 [11–13]. Single photons with an exponentially rising temporal envelope have been prepared from such a heralded single photon source by direct modulation of one of the photons of the pair [14]. 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 [15], and it has already been demonstrated that the resulting photon pairs are nearly Fourier-limited [16] with a coherence time long enough to be resolved with various optical detection techniques. We demonstrate how this source allows the direct preparation of single photons with a rising exponential temporal envelope.

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 $\approx 32$ 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 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 [17–19], 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 [20]. 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 $\Delta t_{12}$. 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 $g^{(2)}(0) = 0$ [21].

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) $D_{11}, D_{12}$ (quantum efficiency $\approx 40\%$, dark count rates 40 to 150 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 $\approx 600$ ps.

From our previous characterization of the source [16], we know that the correlation function between the signal and idler $g^{(2)}_{i1i2}(\Delta t_{si})$ has the shape of a decreasing exponential, with more than 98% of the coincidences occurring within a time window $T_c=30$ ns. We record a histogram $G^{(2)}_{i1i2}|s(\Delta t_{12})$ of idler detection events on $D_{11}$ and $D_{12}$ 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

$$g^{(2)}_{i1i2}|s(\Delta t_{12}) = G^{(2)}_{i1i2}|s(\Delta t_{12})/N_{i1i2}|s(\Delta t_{12}),$$

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 $\Delta t_{12}$ by integrating the time difference histograms between the signal and each arm of the HBT, $G^{(2)}_{s1}|s(\Delta t_{si})$ and $G^{(2)}_{s2}|s(\Delta t_{si})$ within $T_c$.

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 $\Delta t_{12} > 0$, we use

$$N^{(+)}_{i1i2}|s(\Delta t_{12}) = \int_0^{T_c} G^{(2)}_{s1}|s(\Delta t_{si}) G^{(2)}_{s2}|s(\Delta t_{si} + \Delta t_{12}) \, d\Delta t_{si},$$

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

$$N^{(-)}_{i1i2}|s(\Delta t_{12}) = \int_0^{T_c} G^{(2)}_{s1}|s(\Delta t_{si} + \Delta t_{12}) G^{(2)}_{s2}|s(\Delta t_{si}) \, d\Delta t_{si}.$$  

The resulting $g^{(2)}_{i1i2}|s(\Delta t_{12})$ is shown in Figure 2(a) as function of the delay $\Delta t_{12}$, sampled into 2 ns wide time bins. With a signal photon detection rate of 50000 s$^{-1}$ (at $\Delta_2 = 0$), we observe $g^{(2)}_{i1i2}|s(0) = 0.032 \pm 0.004$.

When switching the roles of the signal and idler arms, the resulting normalized correlation function shown in Figure 2(b) has a minimum $g^{(2)}_{i1i2}|s$ of 0.018$\pm$0.007 with an idler photon detection rate of 13000 s$^{-1}$.

We now proceed to determine the temporal envelope of the heralded single photon fields by measuring the field quadrature in the time domain [22, 23] 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 with 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 mW, the electronic noise is about 6-20 dB below the shot noise limit over a band of 10 kHz–210 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 $2.7 \times 10^5$ traces, normalized to the 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 $5 \times 10^5$ traces. Both results are shown in Figure 4. In both configurations, we set $\Delta_2 \approx 6$ MHz to maximize the heralding efficiency.

In Figure 4(a) the normalized field variance of the idler

![Graph](image)
detection time differences we observe in the cascade decrease or fall times are compatible with the distribution of solution [24, 25]. The time constants for the exponential of the idler photon according to the Weisskopf-Wigner exponentially, leading to the characteristic decaying envelope of the intermediate level, which subsequently decays exponentially back to the shot noise level, with a time constant of
\[ \tau_s = 7.4 \pm 0.2 \text{ ns} \]
obtained from a fit. Here, the supression of uncorrelated trigger (idler) photons by the etalon E (figure 3(b)) results in a higher heralding efficiency \( \eta_s \approx 19\% \), and therefore a higher signal to shot noise level.

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 with single atoms or molecules.

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 [27]). 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 [28], like the two-state vector formalism [29], 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 — efficiently be absorbed by a single atom in free-space in a time-reversed Weisskopf-Wigner situation. Such an experiment would therefore not only demonstrate strong
atom-photon interactions, but also provide a better understanding to what extent heralded photons are equivalent to single photons emerging from a setup with a well-defined initial condition.

We acknowledge the support of this work by the National Research Foundation & Ministry of Education in Singapore.

* Center for Quantum Technologies 3 Science Drive 2, Singapore, 117543
† Department of Physics, National University of Singapore, 2 Science Drive 3, Singapore, 117542; Center for Quantum Technologies, National University of Singapore, 3 Science Drive 2, Singapore, 117543
[1] J. I. Cirac, P. Zoller, H. J. Kimble, and H. Mabuchi, Phys. Rev. Lett. 78, 3221 (1997).
[2] L.-M. Duan, M. D. Lukin, J. I. Cirac, and P. Zoller, Nature 414, 413 (2001).
[3] T. Wilk, S. C. Webster, A. Kuhn, and G. Rempe, Science 317, 488 (2007).
[4] H. J. Kimble, Nature 453, 1023 (2008).
[5] Y. Wang, J. Minár, L. Sheridan, and V. Scarani, Phys. Rev. A 83, 063842 (2011).
[6] S. A. Aljunid, G. Maslennikov, Y. Wang, D. H. Lan, V. Scarani, and C. Kurtsiefer, Phys. Rev. Lett 111, 103601 (2013).
[7] M. Bader, S. Heugel, A. L. Chekhov, M. Sondermann, and G. Leuchs, New Journal of Physics 15, 123008 (2013).
[8] N. Schlosser, G. Reymond, I. Protsenko, and P. Grangier, Nature 411, 1024 (2001).
[9] M. Sondermann, R. Maiwald, H. Konermann, N. Lindlein, U. Peschel, and G. Leuchs, Appl. Phys. B 89, 489 (2007).
[10] M. K. Tey, Z. Chen, S. A. Aljunid, B. Chng, F. Huber, G. Maslennikov, and C. Kurtsiefer, Nat Phys 4, 924 (2008).
[11] P. Grangier, G. Roger, and A. Aspect, EPL 1, 173 (1986).
[12] C. K. Hong and L. Mandel, Phys. Rev. Lett. 56, 58 (1986).
[13] J. F. Clauser, Phys. Rev. D 9, 853 (1974).
[14] P. Kolchin, C. Belthangady, S. Du, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. 101, 103601 (2008).
[15] T. Chaneièère, D. N. Matsukevich, S. D. Jenkins, T. A. B. Kennedy, M. S. Chapman, and A. Kuzmich, Phys. Rev. Lett. 96, 093604 (2006).
[16] B. Srivathsan, G. K. Guliati, B. Chng, G. Maslennikov, D. Matsukevich, and C. Kurtsiefer, Phys. Rev. Lett. 111, 123602 (2013).
[17] C. Kurtsiefer, S. Mayer, P. Zarda, and H. Weinfurter, Phys. Rev. Lett. 85, 290 (2000).
[18] P. Michler, A. Kiraz, C. Becher, W. Schoenfeld, P. Petroff, L. Zhang, E. Hu, and A. Imamoglu, Science 290, 2282 (2000).
[19] A. Kuhn, M. Henrich, and G. Rempe, Phys. Rev. Lett. 89, 067901 (2002).
[20] L. Mandel and E. Wolf, Optical Coherence and Quantum Optics (Cambridge University Press, 1995), 1st ed., ISBN 0521417112.
[21] R. J. Glauber, Phys. Rev. 131, 2766 (1963).
[22] H. P. Yuen and V. W. S. Chan, Optics Letters 8, 177 (1983).
[23] A. I. Lvovsky, H. Hansen, T. Aichele, O. Benson, J. Mlynek, and S. Schiller, Phys. Rev. Lett. 87, 050402 (2001).
[24] V. Weisskopf and E. Wigner, Z. Physik 63, 54 (1930).
[25] J. D. Franson, Phys. Rev. Lett. 62, 2205 (1989).
[26] R. H. Dicke, Physical Review 93, 99 (1953).
[27] A. Einstein, B. Podolsky, and N. Rosen, Phys. Rev. 47, 777 (1935).
[28] S. Watanabe, Reviews of Modern Physics 27, 179 (1955).
[29] B. Reznik and Y. Aharonov, Phys. Rev. A 52, 2538 (1995).