

## Deterministic Single Photons via Conditional Quantum Evolution

D. N. Matsukevich, T. Chanelière, S. D. Jenkins, S.-Y. Lan, T. A. B. Kennedy, and A. Kuzmich

*School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332-0430, USA*

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A source of deterministic single photons is proposed and demonstrated by the application of a measurement-based feedback protocol to a heralded single-photon source consisting of an ensemble of cold rubidium atoms. Our source is stationary and produces a photoelectric detection record with sub-Poissonian statistics.

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Quantum state transfer between photonic- and matter-based quantum systems is a key element of quantum information science, particularly of quantum communication networks. Its importance is rooted in the ability of atomic systems to provide excellent long-term quantum information storage, whereas the long-distance transmission of quantum information is nowadays accomplished using light. Inspired by the work of Duan *et al.* [1], emission of nonclassical radiation has been observed in first-generation atomic ensemble experiments [2].

In 2004 the first realization of coherent quantum state transfer from a matter qubit onto a photonic qubit was achieved [3]. This breakthrough laid the groundwork for several further advances towards the realization of a long-distance, distributed network of atomic qubits, linear optical elements, and single-photon detectors [4–8]. A seminal proposal for universal quantum computation with a similar set of physical resources has also been made [9].

An important additional tool for quantum information science is a deterministic source of single photons. Previous implementations of such a source used *single emitters*, such as quantum dots [10,11], color centers [12,13], neutral atoms [14,15], ions [16], and molecules [17]. The measured efficiency  $\eta_D$  to detect a single photon per trial with these sources is typically less than 1%, with the highest reported measured value of about 2.4% [14], to our knowledge.

We propose a deterministic single-photon source based on an *ensemble of atomic emitters*, measurement, and conditional quantum evolution. We report the implementation of this scheme using a cold rubidium vapor, with a measured efficiency  $\eta_D \approx 1\%–2\%$ . In common with the cavity QED system, our source is suitable for reversible quantum state transfer between atoms and light, a prerequisite for a quantum network. However, unlike cavity QED implementations [14], it is unaffected by intrinsically probabilistic single atom loading. Therefore, it is stationary and produces a photoelectric detection record with truly sub-Poissonian statistics.

The key idea of our protocol is that a single photon can be generated at a predetermined time if we know that the medium contains an atomic excitation. The presence of the latter is heralded by the measurement of a scattered photon

in a *write* process. Since this is intrinsically probabilistic, it is necessary to perform independent, sequential *write* trials before the excitation is heralded. After this point one simply waits and reads out the excitation at the predetermined time. The performance of repeated trials and heralding measurements represents a conditional feedback process and the duration of the protocol is limited by the coherence time of the atomic excitation. Our system has therefore two crucial elements: (a) a high-quality probabilistic source of heralded photons and (b) long atomic coherence times. We note that related schemes using parametric down-conversion have been discussed [18].

Heralded single-photon sources are characterized by mean photon number  $\langle \hat{n} \rangle \ll 1$ , as the unconditioned state consists mostly of vacuum [19,20]. More importantly, in the absence of the heralding information the reduced density operator of the atomic excitation is thermal [21]. In contrast, its evolution conditioned on the recorded measurement history of the signal field in our protocol ideally results in a single atomic excitation. However, without exception all prior experiments with atomic ensembles did not have sufficiently long coherence times to implement such a feedback protocol [2–7,22–24]. In earlier work quantum feedback protocols have demonstrated control of nonclassical states of light [25] and motion of a single atom [26] in cavity QED.

We first outline the procedure for heralded single-photon generation. A schematic of our experiment is shown in Fig. 1. An atomic cloud of optical thickness  $\approx 7$  is provided by a magneto-optical trap (MOT) of  $^{85}\text{Rb}$ . The ground levels  $\{|a\rangle; |b\rangle\}$  correspond to the  $5S_{1/2}$ ,  $F_{a,b} = \{3, 2\}$  hyperfine levels, while the excited level  $|c\rangle$  represents the  $\{5P_{1/2}, F_c = 3\}$  level of the  $D_1$  line at 795 nm. The experimental sequence starts with all of the atoms prepared in level  $|a\rangle$ . An amplitude modulator generates a linearly polarized 70 ns long *write* pulse tuned to the  $|a\rangle \rightarrow |c\rangle$  transition, and focused into the MOT with a Gaussian waist of about  $430 \mu\text{m}$ . We describe the *write* process using a simple model based on nondegenerate parametric amplification. The light induces spontaneous Raman scattering via the  $|c\rangle \rightarrow |b\rangle$  transition. The annihilation of a *write* photon creates a pair of excitations: namely, a signal photon and a quasibosonic collective atomic excitation

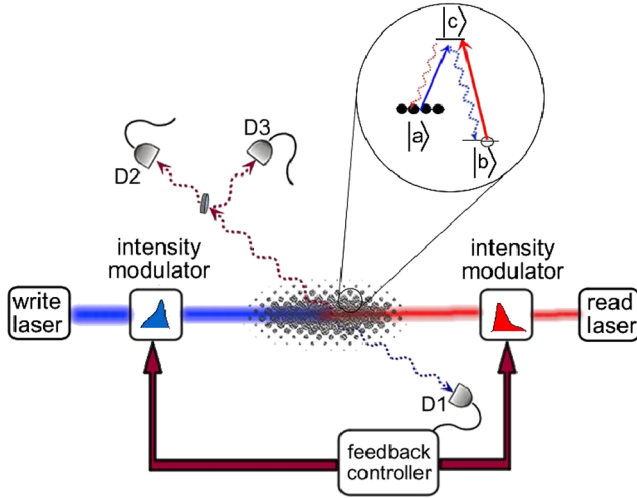


FIG. 1 (color online). Schematic of experimental setup, with the inset showing the atomic level scheme (see text).

[1]. The scattered light with polarization orthogonal to the *write* pulse is collected by a single-mode fiber and directed onto a single-photon detector D1, with overall propagation and detection efficiency  $\eta_s$ . Starting with the correlated state of signal field and atomic excitation, we project out the vacuum from the state produced by the write pulse using the projection operator  $:\hat{1} - e^{-\hat{d}^\dagger \hat{d}}:$ , where  $\hat{d} = \sqrt{\eta_s} \hat{a}_s + \sqrt{1 - \eta_s} \hat{\xi}_s$ ,  $\hat{a}_s$  is the detected signal mode, and  $\hat{\xi}_s$  is a bosonic operator accounting for degrees of freedom other than those detected. Tracing over the signal and all other undetected modes, we find that the density matrix for the atomic excitation  $A$  conditioned on having at least one photoelectric detection event is given by [27]

$$\rho_{A|1} = \frac{1}{p_1} \sum_{n=1}^{\infty} \frac{\tanh^{2n} \chi}{\cosh^2 \chi} (1 - (1 - \eta_s)^n) |n\rangle \langle n|, \quad (1)$$

where  $p_1 \ll 1$  is the probability of a signal photoelectric detection event per *write* pulse, and the interaction parameter  $\chi$  is given in terms of  $p_1$  and  $\eta_s$  by

$$\sinh^2 \chi = p_1 / [\eta_s (1 - p_1)], \quad (2)$$

where  $|n\rangle \equiv \hat{A}^{\dagger n} |0\rangle / \sqrt{n!}$ , and  $|0\rangle$  is the atomic vacuum. Note that in Eq. (1) there is zero probability to find  $|0\rangle$ .

After a storage time  $\tau$ , a *read* pulse of length 80 ns containing around  $3 \times 10^7$  photons, and with polarization orthogonal to that of the *write* pulse, tuned to the  $|b\rangle \rightarrow |c\rangle$  transition, illuminates the atomic ensemble (Fig. 1). Ideally, the *read* pulse converts atomic spin excitations into the idler field emitted on the  $|c\rangle \rightarrow |a\rangle$  transition. The elastically scattered light from the write beam is filtered out, while the idler field polarization orthogonal to that of the *read* beam is directed into a 50:50 single-mode fiber beam splitter. Both *write-read* and signal-idler pairs of fields are counterpropagating [23]. The waist of the signal-idler mode in the MOT is about  $180 \mu\text{m}$ . The two

outputs of the fiber beam splitter are connected to detectors D2 and D3. Electronic pulses from the detectors are gated with 120 ns (D1) and 100 ns (D2 and D3) windows centered on times determined by the *write* and *read* light pulses, respectively. Subsequently, the electronic pulses from D1, D2, and D3 are fed into a time-interval analyzer which records photoelectric detection events with a 2 ns time resolution.

The transfer of atomic excitation to the detected idler field at either  $Dk$  ( $k = 2, 3$ ) is given by a linear optics relation  $\hat{a}_k = \sqrt{\eta_i(\tau)/2} \hat{A} + \sqrt{1 - \eta_i(\tau)/2} \hat{\xi}_k(\tau)$ , where  $\hat{a}_k$  depends parametrically on  $\tau$  and corresponds to a mode with an associated temporal envelope  $\phi(t)$ , normalized so that  $\int_0^\infty dt |\phi(t)|^2 = 1$ , and  $\hat{\xi}_k(\tau)$  is a bosonic operator which accounts for coupling to degrees of freedom other than those detected. The efficiency  $\eta_i(\tau)/2$  is the probability that a single atomic excitation stored for  $\tau$  results in a photoelectric event at  $Dk$ , and includes the effects of idler retrieval and propagation losses, symmetric beam splitter (factor of  $1/2$ ) and nonunit detector efficiency. We start from the elementary probability density  $Q_{k|1}(t_c)$  for a count at time  $t_c$  and no other counts in the interval  $[0, t_c)$ ,  $Q_{k|1}(t_c) = |\phi(t_c)|^2 \langle : \hat{a}_k^\dagger \hat{a}_k \exp(-\int_0^{t_c} dt |\phi(t)|^2 \hat{a}_k^\dagger \hat{a}_k) : \rangle$  [28]. Using Eq. (1), we then calculate probability  $p_{k|1} \equiv \int_0^\infty dt Q_{k|1}(t)$  that detector  $Dk$  registers at least one photoelectric detection event. We similarly calculate the probability  $p_{23|1}$  of at least one photoelectric event occurring at both detectors. These probabilities are given by

$$p_{2|1}(\tau) = p_{3|1}(\tau) = \Pi(\eta_i(\tau)/2; p_1, \eta_s), \quad (3)$$

$$p_{23|1}(\tau) = p_{2|1}(\tau) + p_{3|1}(\tau) - \Pi(\eta_i(\tau); p_1, \eta_s), \quad (4)$$

where we show the explicit dependence on  $\tau$ . Here  $1 - \Pi(\eta; p_1, \eta_s)$  is given by

$$\frac{1}{p_1} \left( \frac{1}{1 + \eta \sinh^2 \chi} - \frac{1}{1 + (\eta_s + \eta(1 - \eta_s)) \sinh^2 \chi} \right).$$

Our conditional quantum evolution protocol transforms a heralded single-photon source into a deterministic one. The critical requirements for this transformation are higher efficiency and longer memory time of the heralded source than those previously reported [4,5]. In Fig. 2 we show the results of our characterization of an improved source of heralded single photons. Figure 2(a) shows the measured intensity cross-correlation function  $g_{si} \equiv [p_{2|1} + p_{3|1}] / [p_2 + p_3]$  as a function of  $p_1$ . Large values of  $g_{si}$  under conditions of weak excitation—i.e., small  $p_1$ —indicate strong pairwise correlations between signal and idler photons. The efficiency of the signal photon generation and detection is given by  $\eta_s \rightarrow g_{si} p_1$ , in the limit  $\sinh^2 \chi \ll 1$ . We have measured  $\eta_s \approx 0.08$ , which includes the effects of passive propagation and detection losses  $\epsilon_s$ . It is important to distinguish the *measured* efficiency from the *intrinsic* efficiency which is sometimes employed. The intrinsic efficiency of having a signal photon in a single

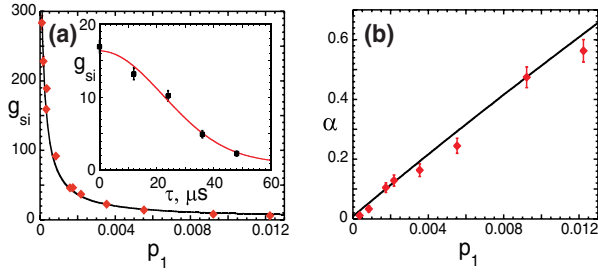


FIG. 2 (color online).  $g_{si}$  (a) and  $\alpha$  (b) vs  $p_1$ , for  $\tau = 80$  ns. The solid lines are based on Eqs. (3) and (4), with a nearly negligible background [5]. The inset shows  $g_{si}$  vs storage time  $\tau$ . The full curve is a fit of the form  $1 + B \exp(-\tau^2/\tau_c^2)$  with  $B = 16$  and  $\tau_c = 31.5 \mu\text{s}$  as adjustable parameters.

spatial mode at the input of the single-mode optical fiber  $\eta_s^0 \equiv (\eta_s/\epsilon_s) \approx 0.24$ . We measure  $\epsilon_s \equiv \epsilon_s^f \epsilon_s^t \epsilon_s^d \approx 0.3$  independently using coherent laser light, where the fiber coupling efficiency  $\epsilon_s^f \approx 0.7$ , optical elements transmission  $\epsilon_s^t \approx 0.85$ , and the detection efficiency  $\epsilon_s^d \approx 0.55$ . The measured efficiency of the idler photon detection is  $\eta_i \rightarrow g_{si}(p_2 + p_3) \approx 0.075$ . Here  $p_2$  and  $p_3$  are defined by expressions analogous to Eq. (2). Similarly, the intrinsic efficiency for the idler field  $\eta_i^0 \equiv (\eta_i/\epsilon_i) \approx 0.34$ , where we measure  $\epsilon_i \equiv \epsilon_i^f \epsilon_i^t \epsilon_i^d \approx 0.22$ , with  $\epsilon_i^f \approx 0.75$ ,  $\epsilon_i^t \approx 0.59$ , and  $\epsilon_i^d \approx 0.55$ . The reported values of  $\eta_s \approx 0.08$  and  $\eta_i \approx 0.075$  represent slight improvements on the previous highest measured efficiencies in atomic ensemble experiments of 0.04–0.07 [5,7].

The quality of the heralded single photons produced by our source is assessed using the procedure of Grangier *et al.*, which involves a beam splitter followed by two single-photon counters, as shown in Fig. 1 [20]. An ideal single-photon input to the beam splitter results in photoelectric detection at either D2 or D3, but not both. An imperfect single-photon input will result in strong anticorrelation of the coincidence counts. Quantitatively, this is determined by the anticorrelation parameter  $\alpha$  given by the ratio of various photoelectric detection probabilities measured by the set of detectors D1, D2, and D3:  $\alpha = p_{23|1}/(p_{2|1}p_{3|1})$ . Classical fields must satisfy a criterion  $\alpha \geq 1$  based on the Cauchy-Schwarz inequality [20]. For an ideally prepared single-photon state  $\alpha \rightarrow 0$ . Panel (b) in Fig. 2 shows the measured values of  $\alpha$  as a function of  $p_1$ , with  $\min\{\alpha\} = 0.012 \pm 0.007$  representing a tenfold improvement on the lowest previously reported value in atomic ensembles [5].

In order to evaluate the atomic memory coherence time  $\tau_c$ , we measure  $g_{si}$  as a function of the storage time  $\tau$ , inset of Fig. 2(a). To maximize  $\tau_c$ , the quadrupole coils of the MOT are switched off, with the ambient magnetic field compensated by three pairs of Helmholtz coils [4]. The measured value of  $\tau_c \approx 31.5 \mu\text{s}$ , a threefold improvement over the previously reported value, is limited by dephasing of different Zeeman components in the residual magnetic field [5,6].

The long coherence time enables us to implement a conditional quantum evolution protocol. In order to generate a single photon at a predetermined time  $t_p$ , we initiate the first of a series of trials at a time  $t_p - \Delta t$ , where  $\Delta t$  is on the order of the atomic coherence time  $\tau_c$ . Each trial begins with a *write* pulse. If D1 registers a signal photoelectric event, the protocol is halted. The atomic memory is now armed with an excitation and is left undisturbed until the time  $t_p$  when a *read* pulse converts it into the idler field. If D1 does not register an event, the atomic memory is reset to its initial state with a cleaning pulse, and the trial is repeated. The duration of a single trial  $t_0 = 300$  ns. If D1 does not register a heralding photoelectric event after  $N$  trials, the protocol is halted  $1.5 \mu\text{s}$  prior to  $t_p$ , and any background counts in the idler channel are detected and included in the measurement record.

Armed with Eqs. (3) and (4), we can calculate the unconditioned detection and coincidence probabilities for the complete protocol. The probability that the atomic excitation is produced on the  $j$ th trial is  $p_1(1 - p_1)^{j-1}$ . This excitation is stored for a time  $(N - j)t_0$  before it is retrieved and detected;  $N = \Delta t/t_0$  is the maximum number of trials that can be performed in the protocol (we ignore the  $1.5 \mu\text{s}$  halting period before the readout).

One can express the probability of a photoelectric event at  $D_k$  ( $k = 2, 3$ ),  $P_k$ , and the coincidence probabilities  $P_{23}$

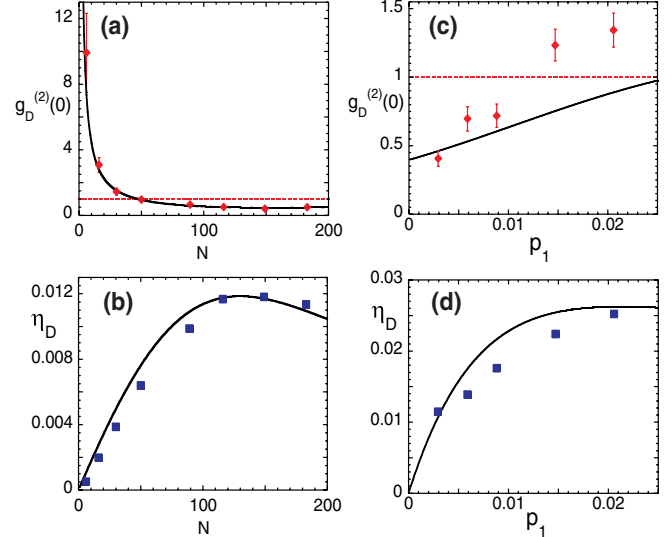


FIG. 3 (color online).  $g_D^{(2)}(0)$  as a function of  $N$  (a) and  $p_1$  (c);  $\eta_D$  as a function of  $N$  (b) and  $p_1$  (d). In (a) and (b)  $p_1 = 0.003$  (about  $6 \times 10^5$  photons per *write* pulse were used), whereas in (c) and (d)  $N = 150$ . The full curves are based on Eq. (5) with the values of efficiencies and coherence times given in the text, with, however,  $\eta_D$  multiplied by an empirical factor of  $2/3$ . We believe this reduced efficiency is due to imperfect switching of the *read* light in the feedback-based protocol (we note that there are no other adjustable parameters in the theory). Deviations from the theory in (c) and (d), could be explained by drifts in the residual magnetic field and *read* light leakage.

in terms of the conditional probabilities of Eqs. (3) and (4),

$$P_\mu = p_1 \sum_{j=1}^N (1 - p_1)^{j-1} p_{\mu|1}(\Delta t - jt_0), \quad (5)$$

$\mu = 2, 3, 23$ . In the limit of infinite atomic coherence time and  $N \rightarrow \infty$ ,  $P_\mu \rightarrow p_{\mu|1}$ . Hence, if the memory time is sufficiently long for an adequate number of trials, the protocol ideally results in deterministic preparation of a single atomic excitation, which can be converted into a single photon at a desired time. Consistent with Fig. 2(a) inset, we assume a combined retrieval-detection efficiency that decays as a Gaussian function of storage time,  $\eta_i(\tau) = \eta_i(0)e^{-(\tau/\tau_c)^2}$ , where  $\tau_c$  is the atomic spin-wave coherence time.

In Fig. 3 we present the measured degree of 2nd order coherence for zero time delay  $g_D^{(2)}(0) \equiv P_{23}/(P_2P_3)$  [29] and the measured efficiency  $\eta_D \equiv P_2 + P_3$  as a function of  $N$  [Figs. 3(a) and 3(b)], and as a function of  $p_1$  [Figs. 3(c) and 3(d)]. The solid curves are based on Eq. (5). The dashed lines in Figs. 3(a) and 3(c) show the expected value of  $g_D^{(2)}(0) = 1$  for a weak coherent state (as we have confirmed in separate measurements). The particular value of  $\Delta t$  is chosen to optimize  $g_D^{(2)}(0)$  and  $\eta_D$ . The minimum value of  $g_D^{(2)}(0) = 0.41 \pm 0.04$  indicates substantial suppression of two-photon events and under the same conditions  $\eta_D = 0.012$  [30]. As shown in Fig. 3(a), when  $N$  is small, the protocol does not result in deterministic single photons. Instead, the cleaning pulse-induced vacuum component of the idler field leads to additional classical noise. Large  $N$ , and hence long coherence times, are crucial to reduce this noise below the coherent state level and to approach a single-photon source. Note that in the limit of infinite atomic memory and  $N \rightarrow \infty$ ,  $g_D^{(2)}(0) \rightarrow \min\{\alpha\} \approx 0.012 \pm 0.007$  and  $\eta_D \rightarrow \eta_i \approx 0.075$ , substantially exceeding the performance of any demonstrated deterministic single-photon source.

Moreover,  $\eta_D$  can be further increased with a larger optical thickness and by optimizing the spatial modes of the signal and idler fields [31]. The spatial signal-idler correlations from an atomic ensemble (and, therefore  $\eta_i^0$ ) can also be improved by use of an optical cavity. However, in the absence of special precautions the use of a cavity will itself introduce additional losses associated, e.g., with the mirror coatings or the cavity locking optics [14,16,24]. The measured efficiency  $\eta_D$  would involve a trade-off between improved spatial correlations due to the cavity and the concomitant losses that it introduces.

In conclusion, we have proposed and demonstrated a stationary source of deterministic single photons based on an ensemble of cold rubidium atoms.

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- [1] L.-M. Duan *et al.*, *Nature (London)* **414**, 413 (2001).
- [2] A. Kuzmich *et al.*, *Nature (London)* **423**, 731 (2003); C. H. van der Wal *et al.*, *Science* **301**, 196 (2003); C. W. Chou *et al.*, *Phys. Rev. Lett.* **92**, 213601 (2004).
- [3] D. N. Matsukevich and A. Kuzmich, *Science* **306**, 663 (2004); similar results were reported a year later in C. W. Chou *et al.*, *Nature (London)* **438**, 828 (2005).
- [4] D. N. Matsukevich *et al.*, *Phys. Rev. Lett.* **95**, 040405 (2005).
- [5] T. Chanelière *et al.*, *Nature (London)* **438**, 833 (2005) and accompanying Supplementary Information.
- [6] D. N. Matsukevich *et al.*, *Phys. Rev. Lett.* **96**, 033601 (2006); S. D. Jenkins *et al.*, *Phys. Rev. A* **73**, 021803(R) (2006).
- [7] D. N. Matsukevich *et al.*, *Phys. Rev. Lett.* **96**, 030405 (2006).
- [8] T. Chanelière *et al.*, *Phys. Rev. Lett.* **96**, 093604 (2006).
- [9] E. Knill, R. Laflamme, and G. J. Milburn, *Nature (London)* **409**, 46 (2001).
- [10] P. Michler *et al.*, *Science* **290**, 2282 (2000).
- [11] C. Santori *et al.*, *Phys. Rev. Lett.* **86**, 1502 (2001); M. Pelton *et al.*, *Phys. Rev. Lett.* **89**, 233602 (2002).
- [12] R. Brouri *et al.*, *Opt. Lett.* **25**, 1294 (2000).
- [13] C. Kurtsiefer *et al.*, *Phys. Rev. Lett.* **85**, 290 (2000).
- [14] A. Kuhn, M. Hennrich, and G. Rempe, *Phys. Rev. Lett.* **89**, 067901 (2002); J. McKeever *et al.*, *Science* **303**, 1992 (2004).
- [15] B. Darquie *et al.*, *Science* **309**, 454 (2005).
- [16] M. Keller *et al.*, *Nature (London)* **431**, 1075 (2004).
- [17] B. Lounis and W. E. Moerner, *Nature (London)* **407**, 491 (2000).
- [18] T. B. Pittman, B. C. Jacobs, and J. D. Franson, *Phys. Rev. A* **66**, 042303 (2002); E. Jeffrey, N. A. Peters, and P. G. Kwiat, *New J. Phys.* **6**, 100 (2004).
- [19] C. K. Hong and L. Mandel, *Phys. Rev. Lett.* **56**, 58 (1986).
- [20] P. Grangier, G. Roger, and A. Aspect, *Europhys. Lett.* **1**, 173 (1986).
- [21] D. F. Walls and G. J. Milburn, *Quantum Optics* (Springer-Verlag, Berlin, 1994).
- [22] M. D. Eisaman *et al.*, *Phys. Rev. Lett.* **93**, 233602 (2004); *Nature (London)* **438**, 837 (2005).
- [23] V. Balic *et al.*, *Phys. Rev. Lett.* **94**, 183601 (2005).
- [24] A. T. Black *et al.*, *Phys. Rev. Lett.* **95**, 133601 (2005).
- [25] W. P. Smith *et al.*, *Phys. Rev. Lett.* **89**, 133601 (2002).
- [26] T. Fischer *et al.*, *Phys. Rev. Lett.* **88**, 163002 (2002).
- [27] This result can also be derived using arguments based on elementary photon counting probabilities [28].
- [28] C. W. Gardiner and P. Zoller, *Quantum Noise* (Springer-Verlag, Berlin, 2000), Chapter 8.
- [29] R. Loudon, *The Quantum Theory of Light* (Oxford University Press, New York, 2000).
- [30] The corresponding value of the measured Mandel parameter  $Q_D \equiv -\eta_D[1 - g_D^{(2)}(0)] \approx -0.007 \pm 10\%$  and is largely determined by  $\eta_D$  [29].
- [31] In separate sets of measurements, we have observed  $\eta_s \approx 0.2$ , for the intrinsic signal efficiency  $\eta_s^0 \approx 0.6$ .