Experimental detection of photons emitted during inhibited spontaneous emission

David Branning*[a], Alan L. Migdall[b], Paul G. Kwiat[c]
[a]Department of Physics, Trinity College, 300 Summit St., Hartford, CT 06106;
[b]Optical Technology Division, NIST, Gaithersburg, MD, 20899-8441;
[c]Department of Physics, University of Illinois,1110 W. Green St., Urbana, IL 61801

ABSTRACT

We present an experimental realization of a "sudden mirror replacement" thought experiment, in which a mirror that is inhibiting spontaneous emission is quickly replaced by a photodetector. The question is, can photons be counted immediately, or only after a retardation time that allows the emitter to couple to the changed modes of the cavity, and for light to propagate to the detector? Our results, obtained with a parametric downconverter, are consistent with the cavity QED prediction that photons can be counted immediately, and are in conflict with the retardation time prediction.

Keywords: inhibited spontaneous emission, cavity QED, parametric downconversion, quantum interference

1. INTRODUCTION

When an excited atom is placed near a mirror, it is allowed to radiate only into the set of electromagnetic modes that satisfy the boundary conditions imposed by the mirror. The result is enhancement or suppression of the spontaneous emission rate, depending on the structure of the allowed modes. In particular, if the atom is placed between two mirrors separated by a distance smaller than the shortest emission wavelength, it will not radiate into the cavity [1-3]. This phenomenon, known as inhibited spontaneous emission, seems paradoxical; for if the atom is prohibited from emitting a photon, then how can it "know" that the cavity is there? One explanation is that the vacuum fluctuations which produce spontaneous emission cannot exist in the cavity, because the modes themselves do not exist [1]. In the long cavity limit of this situation, the atom is sitting at a standing wave node, where the amplitude of the vacuum fluctuations is zero, and the emitted field consists of "virtual" photons [4]. A second, semiclassical explanation is that the atom initially emits a classical dipole field which is reflected and returns to the atom out of phase with itself, preventing any further emission or evolution of the atomic state [5].

However, as pointed out by Fearn, Cook, and Milonni, these two interpretations give conflicting answers to the following question: if a cavity mirror were suddenly replaced by a detector, how much time would elapse until a photon could be detected [6,7]? The first interpretation suggests that the atom must wait to "find out" about the absence of the mirror via a change in the mode structure of the cavity; only after the node is eliminated may the atom emit a photon. This photon would arrive at the detector after a minimum time $2d/c$, where d is the distance from the atom to the detector. In contrast, if counter-propagating fields exist at all times in the cavity, then the detector should be able to register a photon immediately.

We have conducted an experiment to distinguish between these two results, following a suggestion by Weinfurter *et al.* [4]. The data show that it is indeed possible to detect a photon immediately after the mirror is replaced. Because the associated timescales for atoms are too short (~1 fs) to be measured, we have used a spontaneous parametric downconverter (PDC) as an analog instead. Parametric downconversion occurs when a "pump" photon is annihilated within a nonlinear medium to create two lower-frequency photons, referred to as "signal" and "idler." It is possible to frustrate the spontaneous emission from this process by reflecting the pump, signal, and idler beams back into the medium [8-11] "analogous to the modification of single photon spontaneous emission of atoms in cavity QED." [12] However, unlike atomic emission, downconversion into the detected modes can be suppressed with cavities that are much longer than the wavelength of the light, so that the associated time scale can be made long enough (~10 ns) for the "sudden mirror replacement" to be carried out with an electro-optic switch (Pockels cell) [4].

*david.branning @trincoll.edu; phone 1 860 297-4048

The Nature of Light: What Are Photons?, edited by Chandrasekhar Roychoudhuri, Al F. Kracklauer, Katherine Creath, Proceedings of SPIE Vol. 6664, 66640E, (2007) · 0277-786X/07/$18 · doi: 10.1117/12.734807

2. EXPERIMENTAL ARRANGEMENT

In our experiment, an argon-ion laser produces 420 mW of cw light at 351.1 nm, which is directed by a dichroic mirror (D) to pump a type-I PDC (Fig. 1). The pump is vertically (V) polarized, so the PDC generates horizontally (H) polarized signal and idler photons at 702.2 nm. The pump and downconversion beams pass through a positive lens, a tilt glass, and a Pockels cell (PC) before impinging onto the cavity mirror (M). The light reflects back through the PDC, and D then reflects the pump beam away while transmitting the signal and idler through a colored Schott-glass filter (S) and a half-wave plate ($\lambda/2$). A polarizing beamsplitter (PBS) then directs the downconversion either to detector A or detector B, depending on its polarization.

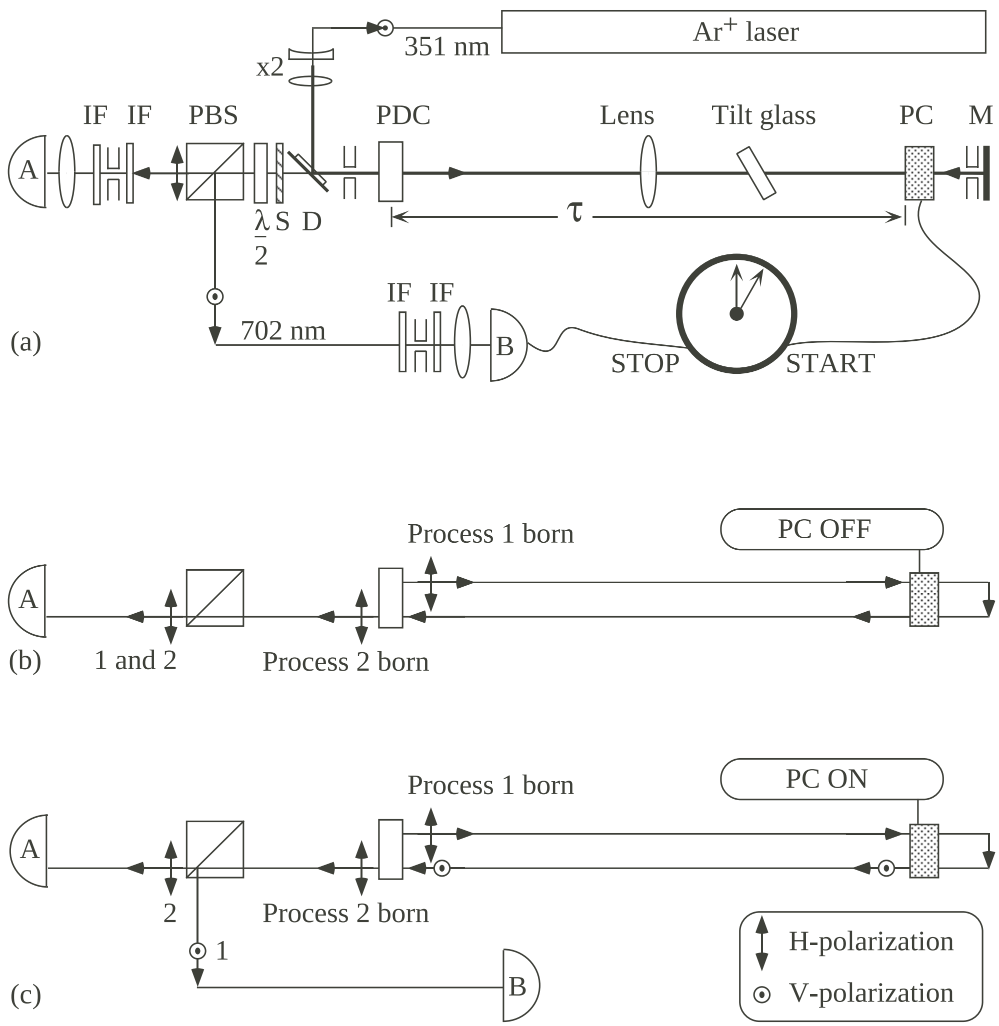

Fig. 1. (a) Schematic of the experiment. Residual laser fluorescence is removed by a pair of prisms (not shown), and the pump beam diameter is expanded to ~2 mm with a x2 telescope. The PDC is 6.25 mm of KDP, cut for collinear, frequency-degenerate downconversion. The lens (f = 500 mm) and tilt glass (12.7 mm thick) are SiO_2. The PC is 44.7 mm of KD*P. The optical path length from the PDC to M is 2048 ± 5 mm[1]. The optical delay from the PDC to the near edge of the PC is τ = 6.19 ± 0.02 ns. The detectors are photon-counting silicon avalanche photodiodes. In front of each detector are broadband (80 nm at A, 40 nm at B) and narrowband (5 nm) interference filters (IF) centered at 702.2 nm, followed by focusing lenses. (b) With the PC off, Process 1 and Process 2 photons go to the same detector. (c) With the PC on, Process 1 photons are rotated from H to V, and deflected to detector B..

[1] All uncertainties refer to combined standard uncertainties with coverage factor k=1.

While the PC is off, this system is simply a collinear version of the frustrated downconversion experiment of Herzog *et al.* [8]. There are two ways to produce photons in the detected 702-nm mode; either the photons are created on the first pass of the pump beam through the PDC and reflected back through (Process 1), or they are created on the second pass by the reflected pump beam (Process 2). Either way, the photons emerge with H polarization and are sent to detector A by the PBS (see Fig. 1b). Because there is no way to distinguish which downconversion process gives rise to the detected photons, their state is a superposition of amplitudes for each possibility[2]:

$$|\psi\rangle \propto \left(e^{i2\phi_{702}} + e^{i\phi_{351}}\right)|2\rangle_{702}. \quad (1)$$

The first amplitude is for "Process 1" photons, each of which accrues a phase ϕ_{702} on its trip through the cavity and back to the center of the PDC. The second amplitude is for "Process 2" photons, which are created after the pump has traveled through the cavity and accrued phase ϕ_{351}. The difference between these two phases is adjusted with the tilt glass. As the glass is tilted away from normal incidence, the optical path length through it is increased; but because of dispersion, the increase is not the same for the pump as it is for the signal and idler. Therefore, interference fringes are produced in the single-channel counting rate (see Fig. 2) according to [13]:

$$\begin{aligned} R &\propto \langle\psi|\hat{a}^{\dagger}_{702}\hat{a}_{702}|\psi\rangle \\ &\cong \alpha\left\{1+\cos\left(\phi_{351}-2\phi_{702}\right)\right\} \end{aligned} \quad (2)$$

where $\hat{a}^{\dagger}$ and $\hat{a}$ are the photon creation and annihilation operators, and α is the photon collection and detection efficiency, typically 0.1 or less.

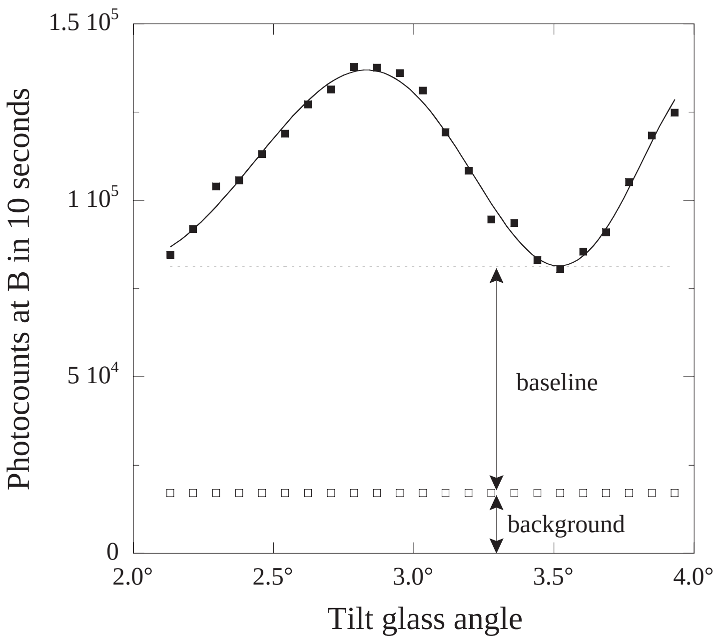

Fig. 2. Single-channel interference fringe obtained by scanning the tilt glass incidence angle within the cavity. The minimum corresponds to a counting rate of 8145 ± 19 s^{-1}. The measured background rate of 1712 ± 22 s^{-1} is plotted along the bottom. The difference is the non-interfering baseline rate, 6433 ± 29 s^{-1}. This fringe was recorded not at detector A, but at detector B (for later comparison with other counting rates at B), by changing the signal and idler polarizations from H to V with the $\lambda/2$ plate oriented at 45º.

[2] This state is an approximation that ignores the vacuum term, in which no photon pairs are produced, and also neglects the small (<< 0.001) relative probability for two or more photon pairs to be produced within the ~300 fs coherence time set by the interference filters.

In principle, when $2\phi_{702}$ differs from ϕ_{351} by π, the counting rate is zero and the emission of the downconverter is frustrated. Just as for an atom, this phenomenon can be described by "virtual" or real photons between the PDC and mirror; the former predicts a round-trip delay for the first photodetection after the mirror is replaced, while the latter predicts no such delay [4].

In practice, the interference visibility does not reach 100%, mainly because of imperfect overlap of the spatial modes for the two downconversion processes[3]. The fringe in Fig. 2 exhibits a visibility of 30% after subtraction of background (pump scatter, dark counts and stray light). To achieve even this visibility, the focal length and position of the lens were chosen to make the gaussian beam propagation matrix for the entire cavity, including all elements, equal to the identity matrix (times -1). However, this cannot be done perfectly for both the pump and downconversion wavelengths due to dispersion in the optical elements. The 1-mm collection irises at the detectors and the x2 beam-expanding telescope in the pump beam also improved the mode-matching by narrowing the range of propagation vectors produced and collected. Much of the mode mismatch can be directly attributed to the PC, which was observed to degrade the visibility in a shorter (one meter) cavity from 43% to 11%, probably by distorting the wavefronts.

The "sudden mirror replacement" experiment is conducted with the tilt glass fixed at the fringe minimum, where the emission is (partially) frustrated and the background-corrected counting rate at B drops to $6433 \pm 29\ s^{-1}$, well below the mean rate of $9206 \pm 25\ s^{-1}$. In this configuration, a 2.3-kV voltage is applied to the PC (within 7.5 ns), so that the PC acts like a $\lambda/4$ plate at 702 nm, oriented at 45º to the H axis. When the downconversion light passes twice through the activated PC (before and after being reflected off the cavity mirror), it is rotated from H to V polarization, and passes back through the cavity and PDC without interfering. The PBS then directs these V-polarized photons, *which can only have come from Process 1,* to detector B (see Fig. 1c)[4]. Because these photons have the wrong polarization for interference at the PDC, their trip back through the cavity amounts to a simple delay line before they impinge onto detector B. *From the point of view of the downconverter, it is as if the mirror has been removed entirely, because the mode that is reflected back is incapable of causing interference.*

A timer is started just before the PC is triggered, and is stopped by the detection of a photon at B. The elapsed time is recorded, the PC is reset to the "off" position, and the experiment is repeated.

---

[3] Differential losses for the pump and downconversion in the cavity cannot explain the low visibility. The collimating lens is SiO_2, with reflection losses at 351 nm and 702 nm limited to 0.25% and 0.5% per interface, respectively, by antireflection coatings. The measured tilt glass transmittance is 97% at 351 nm and 97.5% at 702 nm. The Pockels cell transmission is 70% at 351 nm and 92% at 702 nm. When combined, these values allow a maximum visibility of 95.4%.

[4] V-polarized photons cannot come from Process 2, because the downconverter is not phase-matched to emit photons with V polarization.

3. RESULTS AND INTERPRETATION

A histogram of the recorded arrival times is shown in Fig. 3. The mean number of counts in each of the first 24 bins, before the PC is switched, (region I) is 43.6 ± 1.5. Each bin represents 35.3 ms of total integration time, so this mean number corresponds to a counting rate of 1235 ± 42 s^{-1}. Because of electronic delays between the start and stop signals, the counts in this region are due entirely to background, and the rate is in good agreement with the independently-measured background rate of 1193 ± 20 s^{-1}. In the next 7.32 ns (region II), the counting rate rises to $(1 - 1/e)$ of its final value as the first photons from the PDC are detected. After another 22 ns, the rate settles to a final mean value of 6486 ± 61 s^{-1} (region III), including a background rate of 1238 ± 20 s^{-1}. The background rates are different in regions I and III because the PC scatters pump light differently in the "on" and "off" positions.

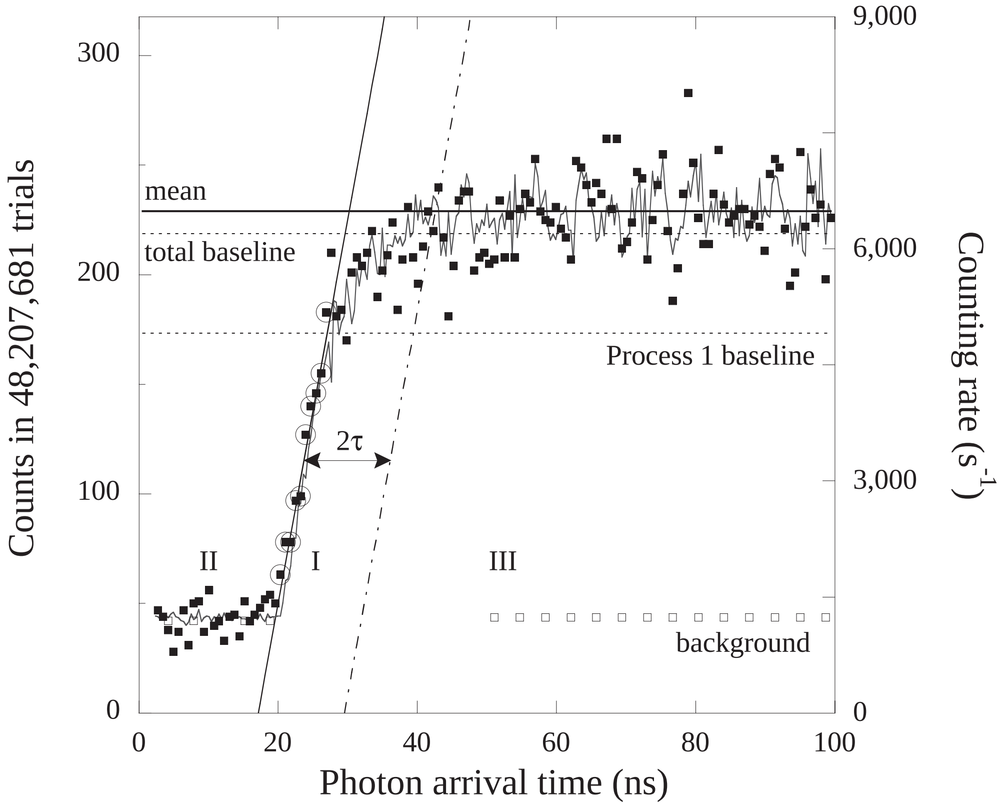

Fig. 3. Histogram of the arrival times of downconverted photons at detector B (solid squares), measured from just before the PC is activated. The width of each bin is 732 ps. The sloped solid line (region II) is a fit to the first 10 bins (circled) of the rise in the counting rate. The dot-dashed line is where the rise would occur if the PDC emission were delayed until after the mode structure of the cavity changed. The jagged line represents data taken under identical conditions with an attenuated HeNe laser, scaled to the PDC data. The thick horizontal line at 6486 s^{-1} is the mean counting rate of downconversion after the rise is completed. The dotted horizontal line is the baseline rate from non-interfering photons (see Fig. 2), corrected for losses and instabilities in the interferometer, and added to the background rate. The measured background rates of 1193 ± 20 s^{-1} before the PC is activated (region I) and 1238 ± 20 s^{-1} after the rise is completed (region III) are plotted along the bottom.

Did the photons arrive early, or after some round-trip delay? The jagged line superimposed on Fig. 3 shows data taken with a HeNe laser in the same cavity, under identical conditions. Because there are no interfering paths for the HeNe beam, there is no suppression of these photons, and no question that they must arrive at the "early" time -- there are always HeNe photons impinging on the PC, which will begin rotating their polarizations the moment it is activated. The downconverted photon detections in Fig. 3 closely follow the arrival times marked by this line, showing that they, too, must have been present in the cavity before the PC was switched. The optical path length from the PDC to the near edge of the PC is $d = 1865 \pm 5$ mm. The PDC therefore does not see the changed cavity mode structure until a time $\tau = d/c$ has elapsed from the beginning of the switch (see Fig. 1a). If the PDC were waiting to emit photons until information about the changed boundary condition reached it, these photons would also require another delay τ to propagate back to the PC. The total delay at detector B would then be $2\tau = 12.37 \pm 0.03$ ns after the "immediate" time defined by the HeNe arrival, and this delayed rise would correspond to the dot-dashed line in Fig. 3; but in fact the photons arrive earlier than this by 29 standard deviations[5].

Hence, in this experiment, there is a nonzero probability to detect a photon immediately after the mirror is "switched out" (modulo the final propagation time to the detector.) However, to understand whether this result is meaningful in the context of inhibited spontaneous emission, several practical issues must be addressed.

The first issue is that, because of the low fringe visibility, the majority of the detected photons do not have anything to do with inhibited spontaneous emission. Even at the minimum of the fringe in Fig. 2, the PDC is still emitting light, and we observe a non-interfering "baseline" rate of downconversion that is always taking place from both Process 1 and Process 2. It is then hardly surprising that some photons arrive at detector B at the same time as the HeNe photons. The only photons that can be unambiguously identified as having come from a frustrated downconversion process are those which remain after this baseline is subtracted. Fig. 3 shows that during the experiment, the mean counting rate in region III is $5252 \pm 74\ \mathrm{s}^{-1}$. This is actually *lower* than the baseline rate in Fig. 2, but a careful measurement of the counting rates at detectors A and B revealed that the PC in the "on" position only delivered $73.75 \pm 0.15\,\%$ of the available photons in the cavity to detector B[6]. Thus, all of the Process 1 photons that are rotated to V by the PC are depleted on their way to detector B. When the baseline rate is rescaled by this factor, it becomes $4744 \pm 31\ \mathrm{s}^{-1}$. The mean counting rate exceeds this by $508 \pm 80\ \mathrm{s}^{-1}$.

The second issue is stability. Although the PC switches from "off" to "on" in less than 8 ns, the repetition rate is limited by the high-voltage electronics to ~ 8 kHz. Thus, several hours of operation are required to collect the data in Fig. 3. Instabilities over this time period can affect the counting rates in two ways: the visibility may be reduced, and/or the interferometer may not stay in the "frustrated" position. Each of these instabilities serves to further increase the baseline counting rate due to non-interfering photons. To check for these effects, pauses were undertaken during the data collection to measure the counting rate and tilt-glass position at the fringe minimum. (After each check, the tilt glass was repositioned at the new minimum to acquire more data.) The result is that an excess counting rate of $218 \pm 100\ \mathrm{s}^{-1}$ is attributed to degradation in the visibility[7] while phase shifts (measured 4 times) contribute an additional $5 \pm 29\ \mathrm{s}^{-1}$ to the counting rate[8]. These contributions raise the baseline rate to $4966 \pm 105\ \mathrm{s}^{-1}$, which reduces the excess photons to a rate of $290 \pm 130\ \mathrm{s}^{-1}$, or 2.2 standard deviations above the baseline, as shown in Fig. 3.

Finally, it is crucial in all of these measurements that the background counting rate at detector B be minimized, and then measured correctly. The major source of counts at B which are not from downconversion is scattered light from the pump beam, which travels collinearly with the downconversion until the final pass through the dichroic mirror. Eliminating this scatter poses a challenge because the pump beam is 12 orders of magnitude brighter than the downconversion. The combination of the dichroic mirror, the PBS, the Schott glass, and the broadband and narrowband interference filters attenuate the pump scatter so that it contributes a background rate on the order of $1000\ \mathrm{s}^{-1}$ at B.

The total background rate at B, including the pump contribution, is measured by tilting the optic axis of the PDC by several degrees, so that the collinear phase-matching conditions are no longer well-satisfied for the detected downconversion wavelengths. This ensures that no more than $0.5\,\%$ of the usual downconversion is produced in the cavity, while the pump beam still travels through the cavity in virtually the same way as during the experiment. In this configuration, the measured counting rate $R^{\text{PDC off}}_{\text{cavity on}}$ at B is due entirely to background sources: dark counts, scattered

---

[5] The first 10 (circled) bins were fit to a straight line whose x-intercept has an uncertainty of 0.42 ns. This is the extent to which the arrival time of the earliest photons is known. The error on the delay, $2d/c$, is 0.03 ns.

[6] This loss is partially due to the dichroic mirror (D), which transmits $\sim 85\,\%$ of V-polarized light compared to H-polarized light because of its tilted orientation in the plane of incidence (see Figure 1). The remaining $\sim 10\,\%$ loss could be due to polarization or spatial mode effects within the Pockels cell.

[7] This is obtained from averaging a linear fit to the recorded fringe minima vs. time in each interval; the excess rate is then rescaled for cavity losses.

[8] This is computed by summing the counts above the fringe minimum that would be collected due to the shift in each interval. All shifts were less than 0.2λ. The ambient temperature did not fluctuate outside the values associated with these shifts.

pump light, and ambient light. However, because the crystal has been tilted away from normal incidence, this measure of the background does not include the backwards scattering of the pump from the faces of the PDC crystal itself. To correct for this, the backscatter alone is independently measured by removing the cavity mirror, so that any light traveling through the PDC cannot return to the detectors, and then taking the difference between the counting rates with the crystal at normal incidence and with it tilted. This difference, which represents only the backscatter from the PDC crystal, is then added to the initial background measurement. That is,

$$\underbrace{R_{\text{cavity on}}^{\text{PDC on}}}_{\text{total background}} = \underbrace{R_{\text{cavity on}}^{\text{PDC off}}}_{\substack{\text{no downconversion} \\ \text{no backscattered pump}}} + \underbrace{R_{\text{cavity off}}^{\text{PDC on}} - R_{\text{cavity off}}^{\text{PDC off}}}_{\text{backscattered pump}} \tag{3}$$

The background is estimated in this way for each condition of the Pockels cell (on or off) and the $\lambda/2$ plate (0º or 45º), and applied to the data appropriately. For example, with the Pockels cell on, and the $\lambda/2$ plate at 0º, $R_{\text{cavity on}}^{\text{PDC off}} = 1192 \pm 11\ \text{s}^{-1}$, $R_{\text{cavity off}}^{\text{PDC on}} = 1494 \pm 12\ \text{s}^{-1}$, and $R_{\text{cavity off}}^{\text{PDC off}} = 1448 \pm 12\ \text{s}^{-1}$, for a total background rate of $1238 \pm 20\ \text{s}^{-1}$ as shown in region III of Figure 3.

4. CONCLUSION

Even after taking into account the practical limitations of our experiment due to visibility, stability, and background rates, the data in Fig. 3 remain a strong indication of the immediate detection of photons from an inhibited spontaneous emitter. Although this experiment is unique in that sense, it does have features in common with some previous experiments. It is based on the frustrated downconversion first demonstrated by Herzog *et al.* [8], but with several major departures: the geometry is collinear, the cavity is seventeen times longer, and the interference is time-dependent. A more recent experiment by Wang *et al.* [14] investigated the time-dependent features of "induced coherence without induced emission," a similar quantum interference effect based on the overlap of downconversion modes; however, that effect does not suit our present purposes because emission from those downconverters is neither suppressed nor enhanced by the interference. In another related experiment, Kauranen and co-workers [15] measured the intracavity power in an upconverted beam, as the output beam was enhanced or suppressed by the cavity. The intracavity power was found to be independent of the enhancement or suppression, suggesting that a stimulated upconverter also does not "wait to know" about the boundary condition before emitting light. In our experiment we have taken the analogy with the "sudden replacement thought experiment" several steps further, by changing the boundary condition in a time-dependent way, and by using a spontaneous downconversion arrangement that is "a generalization of cavity QED experiments to a situation where the separation between the emitter and mirrors greatly exceeds the wavelength" [12]. (It has been shown elsewhere that the modification of atomic spontaneous emission rates is not qualitatively different for long or short cavities [16].)

In conclusion, we have experimentally demonstrated that there is a nonzero chance to detect a photon from inhibited spontaneous emission immediately after the inhibiting mirror is replaced with a detector. The photons arrive at the earliest possible time, 29 standard deviations before the retardation time $2d/c$ has elapsed.

ACKNOWLEDGEMENTS

We thank R. Erdmann, W.P. Grice, A.M. Steinberg, and I.A. Walmsley for helpful suggestions, and E.J. Heilweil for loan of the Pockels cell. This work was supported by the National Research Council.

REFERENCES

1. D. Kleppner, "Inhibited Spontaneous Emission," Phys. Rev. Lett. **47**, 233 (1981).
2. G. Gabrielse and H. Dehmelt, "Observation of Inhibited Spontaneous Emission," Phys Rev. Lett. **55**, 67 (1985).
3. R. Hulet, E. Hilfer, and D. Kleppner, "Inhibited Spontaneous Emission by a Rydberg Atom," Phys. Rev. Lett. **55**, 2137 (1985).

4. H. Weinfurter, T. Herzog, P. G. Kwiat, J. G. Rarity, A. Zeilinger, and M. Zukowski, "Frustrated Downconversion: Virtual or Real Photons?," Ann. New York Acad. Sci. **755**, 61 (1995).
5. J. Dowling, M. O. Scully, and F. De Martini, "Radiation pattern of a classical dipole in a cavity," Opt. Commun. **82**, 415 (1991).
6. H. Fearn, R. J. Cook, and P. W. Milonni, "Sudden Replacement of a Mirror by a Detector in Cavity QED: Are Photons Counted Immediately?" Phys. Rev. Lett. **74**, 1327 (1995).
7. J. Dowling, "Mirror, mirror, on the wall – is the photon there at all?" Phys. World **8**, 23 (1995).
8. T. J. Herzog, J. G. Rarity, H. Weinfurter, and A. Zeilinger, "Frustrated Two-Photon Creation via Interference," Phys. Rev. Lett. **72**, 629 (1994).
9. H. Paul and M. Pavicic," Nonclassical interaction-free detection of objects in a monolithic total-internal-reflection resonator," J. Opt. Soc. B **14**, 1275 (1997).
10. P. H. Souto Ribero and G. A. Barbosa, "Mirror effects and induced coherence in parametric down-conversion," Opt. Commun. **139**, 139 (1997).
11. A. Lamas-Linares, J. C. Howell and D. Bouwmeester, "Stimulated emission of polarization-entangled photons," Nature **412**, 887 (2001).
12. P. W. Milonni, H. Fearn, and A. Zeilinger, "Theory of two-photon down-conversion in the presence of mirrors," Phys. Rev. A **53**, 4556 (1996).
13. L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics* (Cambridge University Press, Cambridge, 1995).
14. L. J. Wang and J. –K. Rhee, "Propagation of transient quantum coherence," Phys. Rev. A **59**, 1654 (1999).
15. M. Kauranen, Y. Van Rompaey, J. J. Maki, and A. Persoons, "Nonvanishing Field between a Dipole Oscillator and a Reflecting Boundary during Suppression of Dipole Radiation," Phys. Rev. Lett. **80**, 952 (1998).
16. H. Gießen, J. D. Berger, G. Mohs, P. Meystre, and S. F. Yelin, "Cavity-modified spontaneous emission: From Rabi oscillations to exponential decay," Phys. Rev. A 53, 2816 (1996).