

Controlled Single-Photon Emission from a Single Trapped Two-Level Atom

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By illuminating an individual rubidium atom stored in a tight optical tweezer with short resonant light pulses, we created an efficient triggered source of single photons with a well-defined polarization. The measured intensity correlation of the emitted light pulses exhibits almost perfect antibunching. Such a source of high-rate, fully controlled single-photon pulses has many potential applications for quantum information processing.

Implementing a deterministic or conditional two-qubit quantum gate is a key step toward quantum computation. Deterministic gates generally require a strong interaction between the particles that are used to carry the physical qubits (1). Recently, controlled-not gates have been realized with the use of trapped ions and incorporated in elaborate quantum algorithms (2–4). So far, individually addressed two-qubit gates have not been demonstrated with neutral atoms. Promising results have been obtained on entangling neutral atoms with the use of cold controlled collisions in an optical lattice (5), but the single-qubit operations are difficult to perform in such a system.

Another approach is to bypass the requirement for a direct interaction between the qubits and use instead an interference effect and a measurement-induced state projection to create the desired operation (6, 7). An interesting recent development of this idea is the use of photon detection events to create entangled states of two atoms (8–10). This provides “conditional” quantum gates, where the success of the logical operation is heralded by appropriate detection events. These schemes can be extended to realize a full controlled-not gate, or a Bell-state measurement, or more generally to implement conditional unitary operations (8, 11). They could be implemented by using, for instance, trapped ions (12) or atoms in microscopic dipole traps. These proposals require the controlled emission of indistinguishable single photons by at least two identical emitters.

Various single-photon sources have been implemented using solid-state systems as well as atoms or ions. Solid-state systems such as single molecules, nitrogen-vacancy centers in diamond, and quantum dots allow high single-photon rates (13). However, realizing truly identical sources is a major problem for such systems because of inhomogeneities in both

the environment of the emitters and the emitters themselves. Another approach is provided by sources based on neutral atoms (14, 15) or ions (16) strongly coupled to a mode of a high-finesse optical cavity. Such sources are spectrally narrow, and the photons are emitted into a well-defined spatial mode, thus opening the way to coherent coupling of the quantum state of single atoms and single photons. However, the rate at which the system can emit photons is limited by the cavity and is often low in practice. Moreover, the need to achieve the strong coupling regime of cavity quantum electrodynamics remains a demanding experimental requirement.

We present a triggered single-photon source based on a single rubidium atom trapped at the focal point of a lens with a high numerical aperture of 0.7. We also show that we have full control of the optical transition by observing Rabi oscillations. Under these conditions, our system is equivalent to the textbook model formed by a two-level atom driven by monochromatic light pulses. Previous work has shown that by using holographic techniques one can create arrays of dipole traps, each containing a single atom, which can be addressed individually (17). The work presented here can therefore be directly scaled to two or more identical emitters.

We trapped the single rubidium-87 atom at the focus of the lens using a far-detuned optical dipole trap (810 nm), loaded from an optical molasses. The same lens was used to collect the fluorescence emitted by the atom (Fig. 1). The experimental apparatus is described in more detail by Schlosser *et al.* (18, 19). A crucial feature of our experiment is the existence of a “collisional blockade” mechanism (19) which allows only one atom at a time to be stored in the trap; if a second atom enters the trap, both are immediately ejected. In this regime, the atom statistics are sub-Poissonian and the trap contains either one or zero (and never two) atoms, with an average atom number of 0.5.

The trapped atom was excited with 4-ns pulses of laser light, resonant with the $S_{1/2}$, $F = 2 \rightarrow P_{3/2}$, $F' = 3$ transition at 780.2 nm. The laser pulses were generated by frequency

doubling pulses at 1560 nm, generated by using an electrooptic modulator to chop the output of a continuous-wave diode laser. A fiber amplifier was used to boost the peak power of the pulses before the doubling crystal. The repetition rate of the source was 5 MHz.

Fluorescence photons were produced by spontaneous emission from the upper state, which has a lifetime of 26 ns. The pulsed laser beam was right-circularly polarized (σ^+ -polarized) with respect to the quantization axis defined by a magnetic field applied during the excitation. The trapped atom was optically pumped into the $F = 2$, $m_F = +2$ ground state by the first few laser pulses. It then cycled on the $F = 2$, $m_F = +2 \rightarrow F' = 3$, $m_{F'} = +3$ transition, which forms a closed two-level system emitting σ^+ -polarized photons. Impurities in the polarization of the pulsed laser beam with respect to the quantization axis, together with the large bandwidth of the exciting pulse (250 MHz), resulted in off-resonant excitation to the $F' = 2$ upper state, leading to possible de-excitation to the $F = 1$ ground state. To counteract this, we added a repumping laser resonant with the $F = 1 \rightarrow F' = 2$ transition. We checked that our two-level description was still valid in the presence of the repumper by analyzing the polarization of the emitted single photons (supporting online material text).

The overall detection and collection efficiency for the light emitted from the atom was measured to be $0.60 \pm 0.04\%$. This was obtained by measuring the fluorescence rate of the atom for the same atomic transition driven by a continuous-wave probe beam and confirmed by a direct measurement of the transmission of our detection system (SOM text).

For a two-level atom and exactly resonant square light pulses of fixed duration T , the probability that an atom in the ground state

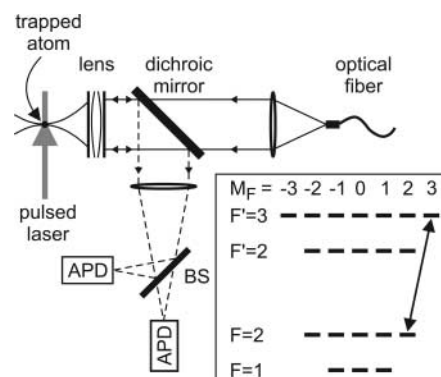


Fig. 1. Schematic of the experiment. The same lens is used to focus the dipole trap and collect the fluorescence light. The fluorescence is separated by a dichroic mirror and imaged onto two photon-counting avalanche photodiodes (APD), placed after a beam-splitter (BS). The insert shows the relevant hyperfine levels and Zeeman sub-levels of rubidium-87. The cycling transition is shown by the arrow. Also shown is the nearby $F = 2$ level responsible for the depumping.

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will be transferred to the excited state is $\sin^2(\Omega T/2)$, where the Rabi frequency Ω is proportional to the square root of the power. Therefore, the excited state population and hence the fluorescence rate oscillates as the intensity is increased. To observe these Rabi oscillations, we illuminated the trapped atom with the laser pulses during 1 ms. We kept the length of each laser pulse fixed at 4 ns, with a repetition rate of 5 MHz, and measured the total fluorescence rate as a function of the laser power. The Rabi oscillations are clearly visible on our results (Fig. 2). From the height of the first peak and the calibrated detection efficiency measured previously, we derived a maximum excitation efficiency per pulse of $95 \pm 5\%$.

The reduction in the contrast of the oscillations at high laser power is mostly due to fluctuations of the pulsed laser peak power. This is shown by the theoretical curve in Fig. 2, based on a simple two-level model. This model shows that the 10% relative intensity fluctuations that we measured on the laser beam are enough to smear out the oscillations as observed.

The behavior of the atom in the time domain can be studied by using time-resolved photon-counting techniques to record the arrival times of the photons relative to the excitation pulses, thus constructing a time spectrum. By adjusting the laser pulse intensity, we observe an adjustable number of Rabi oscillations during the duration of the pulse,

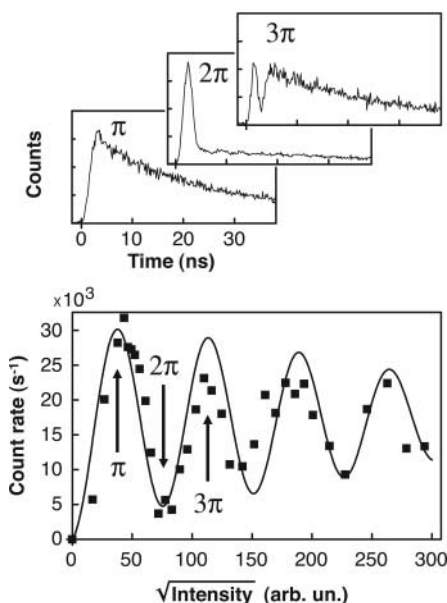


Fig. 2. Total count rate (squares) as a function of the average power of the pulsed beam, for a fixed pulse length of 4 ns and a repetition rate of 5 MHz. The solid line is a theoretical curve based on a simple two-level model that includes spontaneous emission and intensity fluctuations. The inserts show the time spectra for the laser intensities corresponding to π , 2π , and 3π pulses. arb. un., arbitrary units.

followed by the free decay of the atom once the laser has been turned off. The effects of pulses close to π , 2π , and 3π are displayed as inserts in Fig. 2 and show the quality of the coherent control achieved on a single atom.

To use this system as a single photon source, the laser power was set to realize a π pulse. To maximize the number of single photons emitted before the atom was heated out of the trap, we used the following sequence. First, we detected the presence of an atom in the dipole trap in real time using its fluorescence from the molasses light. Then, the magnetic field was switched on and we triggered an experimental sequence that alternated 115- μ s periods of pulsed excitation with 885- μ s periods of cooling by the molasses light (Fig. 3). The repumping laser remained on throughout, and the trap lifetime during the sequence was measured to be 34 ms. After 100 excitation/cooling cycles, the magnetic field was switched off and the molasses was turned back on until a new atom was recaptured and the process began again. On average, three atoms were captured per second under these conditions. The average count rate during the excitation was 9600 s^{-1} , with a peak rate of $29,000 \text{ s}^{-1}$ (corresponding to twice the first peak in Fig. 3).

To characterize the statistics of the emitted light, we measured the second-order temporal correlation function, using a Hanbury Brown and Twiss type set-up. To do this, we used the beam splitter in the imaging system (Fig. 1), which sent the fluorescence light to two photon-counting avalanche photodiodes that were connected to a high-resolution time-to-digital conversion counting card in a start-stop configuration (resolution of about 1 ns). The card was gated so that only photons scattered during the 115 μ s periods of pulsed excitation were counted, and the number of

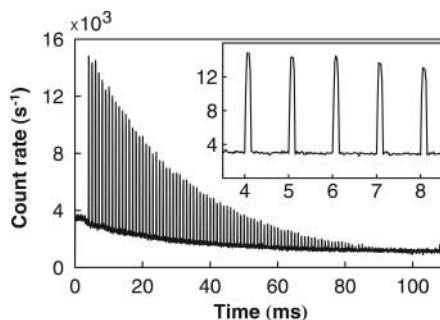


Fig. 3. Fluorescence signal measured by one of the two photodiodes during the experimental sequence, averaged over 22,958 cycles. Peaks are observed corresponding to the 115- μ s periods of pulsed excitation, separated by periods of lower fluorescence induced by the molasses light during the 885 μ s of cooling. The exponential decay of the signal is due to the lifetime of the atom in the trap, which is 34 ms under these conditions. (Insert) A close-up of the signal clearly shows the alternating excitation and cooling periods.

coincidence events was measured as a function of delay. The histogram obtained after 4 hours of continuous operation is displayed in Fig. 4 and shows a series of spikes separated by the period of the excitation pulses (200 ns). The $1/e$ half width of the peaks is $27 \pm 3 \text{ ns}$, in agreement with the lifetime of the upper state. No background correction was done on the displayed data. The small flat background is attributed to coincidences between a fluorescence photon, and an event coming either from stray laser light (about 175 counts/s), or dark counts of the avalanche photodiodes (about 150 counts/s). When we correct for these events, the integrated residual area around zero delay is $3.4 \pm 1.2\%$ of the area of the other peaks.

We calculated (20) that under our experimental conditions, the probability to emit exactly one photon per pulse is 0.981, whereas the probability to emit two photons is 0.019. These two-photon events would show up in the correlation curve as coincidences close to zero delay (still with no coincidences at exactly zero delay). From our calculation, the value for the ratio of the area around zero delay to the area of the other peaks is 3.7%, in excellent agreement with the experimental results.

Finally, we discuss the coherence properties of the emitted photons, necessary for entanglement protocols based on the interference between two emitted photons, either from the same atom or from different atoms. As our collection optics are diffraction limited, the outgoing photons should be in a single spatial mode of the electromagnetic field. As far as temporal coherence is concerned, the main limiting factor appears to be the motion of the atom in the trap, which can be controlled by optimized cooling sequences. We then anticipate that our source should be Fourier limited by the lifetime of the excited state. We are now working to characterize the coherence of our single-photon source and to use it to observe multiple atom interference effects.

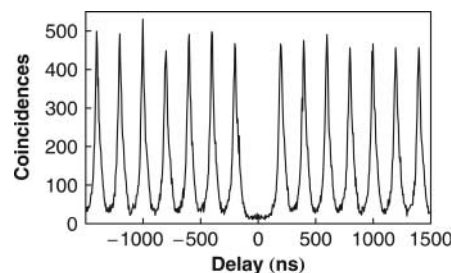


Fig. 4. Histogram of the time delays in the start-stop experiment. The histogram has been binned 4 times, leading to a 4.7-ns time resolution. No correction for background has been made. The absence of a peak at zero delay shows that the source is emitting single photons. During the 4-hour experimental run, 43,895 sequences were completed, which corresponds to a total of 505 s of excitation. A total of 4.83×10^6 photons were detected by the two photodiodes.

References and Notes

1. P. Zoller, J. I. Cirac, L. Duan, J. J. García-Ripoll, in *Les Houches 2003: Quantum Entanglement and Information Processing*, D. Estève, J.-M. Raimond, J. Dalibard, Eds. (Elsevier, Amsterdam, 2004), pp. 187–222.
2. M. Riebe *et al.*, *Nature* **429**, 734 (2004).
3. M. D. Barrett *et al.*, *Nature* **429**, 737 (2004).
4. J. Chiaverini *et al.*, *Nature* **432**, 602 (2004).
5. O. Mandel *et al.*, *Nature* **425**, 937 (2003).
6. E. Knill, R. Laflamme, G. J. Milburn, *Nature* **409**, 46 (2001).
7. J. P. Dowling, J. D. Franson, H. Lee, G. J. Milburn, *Quantum Inf. Process.* **3**, 205 (2004).
8. I. Protzenko, G. Reymond, N. Schlosser, P. Grangier, *Phys. Rev. A* **66**, 062306 (2002).
9. C. Simon, W. T. M. Irvine, *Phys. Rev. Lett.* **91**, 110405 (2003).
10. L.-M. Duan, H. J. Kimble, *Phys. Rev. Lett.* **90**, 253601 (2003).
11. Y. L. Lim, A. Beige, L. C. Kwek, in press; preprint available at <http://arXiv.org/abs/quant-ph/0408043>.
12. B. B. Blinov, D. L. Moehring, L.-M. Duan, C. Monroe, *Nature* **428**, 153 (2004).
13. Single Photons on Demand, P. Grangier, B. Sanders, J. Vukovic, Eds. *New J. Phys.* **6**, 85–100, 129–163 (2004).
14. A. Kuhn, M. Hennrich, G. Rempe, *Phys. Rev. Lett.* **89**, 067901 (2002).
15. J. McKeever *et al.*, *Science* **303**, 1992 (2004).
16. M. Keller, B. Lange, K. Hayasaka, W. Lange, H. Walther, *Nature* **431**, 1075 (2004).
17. S. Bergamini *et al.*, *J. Opt. Soc. Am. B* **21**, 1889 (2004).
18. N. Schlosser, G. Reymond, I. Protzenko, P. Grangier, *Nature* **411**, 1024 (2001).
19. N. Schlosser, G. Reymond, P. Grangier, *Phys. Rev. Lett.* **89**, 023005 (2002).
20. We performed a full calculation of the second-order correlation function using the Heisenberg-Langevin

equations, which gives the areas of the peaks. We also calculated the photon emission probabilities using density matrix and Monte Carlo methods, which lead to the same result.

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Structural Relaxation of Polymer Glasses at Surfaces, Interfaces, and In Between

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We analyzed the glassy-state structural relaxation of polymers near surfaces and interfaces by monitoring fluorescence in multilayer films. Relative to that of bulk, the rate of structural relaxation of poly(methyl methacrylate) is reduced by a factor of 2 at a free surface and by a factor of 15 at a silica substrate interface; the latter exhibits a nearly complete arresting of relaxation. The distribution in relaxation rates extends more than 100 nanometers into the film interior, a distance greater than that over which surfaces and interfaces affect the glass transition temperature.

When cooled below the glass transition temperature (T_g), an amorphous material has higher specific volume, enthalpy, and entropy than in its equilibrium state at the same temperature. The resulting material is called a glass, which relaxes toward thermodynamic equilibrium in a process called structural relaxation (1–3). With polymers, glassy-state structural relaxation, often referred to as physical aging (4–6), can result in a time dependence of end-use properties of critical technological importance (4–7) such as increased modulus, increased brittleness, and reduced permeability. This glassy-state structural relaxation can be distinguished from the cooperative motion associated with T_g ; the latter is associated with the α -relaxation [i.e., relaxation of cooperatively rearranging regions of some tens to hundreds of repeat units (1, 8, 9)], whereas the former has been commonly associated with the β -relaxation (6) (i.e., subsegmental relaxation such as the reorientation of an ester side group).

The glassy state and its associated phenomena, T_g and structural relaxation, are

central challenges in condensed-matter physics (10–14) and are important in fields as varied as optical materials and the preservation of food and insect life (11, 12). Numerous studies have established that confinement of amorphous materials between surfaces and interfaces at a length scale on the order of 100 nm can change T_g relative to its bulk value (9, 15–29), whereas a limited number of studies have provided examples in which T_g is independent of confinement (30, 31). Although many advanced technologies and materials (e.g., thin-film and asymmetric membranes and polymer nanocomposites) demand the long-time use of confined glasses (4, 28, 29), few studies have characterized the effect of nanoconfinement on structural relaxation in the glassy state.

Relative to bulk systems, enthalpy relaxation studies have indicated accelerated structural relaxation of a low molecular weight glass, *ortho*-terphenyl, confined to nanopores 11 to 12 nm in diameter (32). Physical aging in ultrathin polystyrene films was shown to be dependent on thickness and absent when the aging temperature was above the T_g of the confined film but below that of bulk polymer (33, 34). In contrast, the presence of structural relaxation was observed above the bulk T_g for an ultrathin poly(methyl methacrylate)

(PMMA) film supported on a silica substrate (34). The latter result was obtained because confinement leads to an increase in T_g due to attractive polymer-substrate interactions [hydrogen bond formation (25) between hydroxyl groups on the substrate surface and the polymer repeat unit]. Other investigations found little impact of confinement on structural relaxation of supported poly(isobutyl methacrylate) films (18) or superposed studies of structural relaxation with solvent evaporation in the case of supported PMMA films (35).

There is now substantial evidence that the origin of the T_g -nanoconfinement effect is related to surfaces and interfaces modifying relevant T_g dynamics in the interior regions of glass formers (20). Here, we report how surfaces and interfaces modify structural relaxation and to what extent these effects extend into the film interior. We use a fluorescence method in which dye-labeled polymer is inserted into known positions in a film in order to study the glassy-state relaxation dynamics. The dyes exhibit fluorescence intensities that increase substantially with minute increases in local density near the dye (36) and thereby provide an amplified sensitivity to structural relaxation. The method provides for highly sensitive, continuous monitoring of aging as well as the ability to measure aging rate distributions near surfaces, near interfaces, and in between. The distributions have been obtained with the use of 4-tricyanovinyl-*N*-(2-hydroxyethyl)-*N*-ethyl-aniline (TC1)-labeled PMMA in single layers of multilayer PMMA films (37). We chose TC1 dye for the present study because its molecular structure allows for conformational mobility, which yields a high sensitivity of the dye fluorescence to the small densification that accompanies physical aging (34, 36).

Figure 1 shows the increase in fluorescence intensity during relaxation at 305 K of a 400-nm-thick TC1-labeled PMMA film [$T_g(\text{bulk}) = 393$ K]. In agreement with specific volume and enthalpy relaxation measurements during physical aging of bulk polymer (7, 38), the fluorescence intensity is roughly linear with logarithmic aging time. The intensity increases

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Polarization analysis of the emitted single photons

As explained in the main text, polarization imperfections lead to a depumping process to the $F = 1$ ground state. A repumping laser is used to counteract this process and minimize deviations from the two-level behavior.

To check the validity of our two-level description, we have investigated the effect of impurities in the polarization of the pulsed laser beam with respect to the quantization axis. We measure that on average the atom is pumped into the $F = 1$ ground state by spontaneous emission after 120 excitations. In the presence of the repumping light, a rate equation model of the repumping process (including the repumping laser as well as the pulsed excitation) shows that the atom spends more than 90% of its time on the cycling $F = 2, m_F = +2 \rightarrow F' = 3, m'_F = +3$ transition, as desired.

In addition, we have measured the polarization of the emitted light, using a polarizer to select fluorescence light either polarized perpendicularly ($\perp$) or parallel ($\parallel$) to the quantization axis. For a narrow collection angle, $\perp$ would correspond to the circularly polarized photons emitted on the cycling transition, and $\parallel$ to π polarized photons. Here, the measured contrast $(R_{\perp} - R_{\parallel})/(R_{\perp} + R_{\parallel}) = 72(\pm 2)\%$, where $R_{\perp}$ and $R_{\parallel}$ are the count rates perpendicular and parallel to the quantization axis respectively. The largest part of this depolarization is actually due to the very large numerical aperture (N.A. = 0.7) of the collection lens, which decreases to 77% the maximum contrast obtainable for purely σ -polarized fluorescence. From the measured contrast of 72%, we calculate that 3% of the collected photons are π -polarized and we attribute this to photons induced by the depumping-repumping processes. This number is compatible with the results of the rate equation model discussed in the previous paragraph.

Collection and detection efficiency

The overall collection and detection efficiency of $(0.60 \pm 0.04)\%$ is obtained by measuring the fluorescence rate of the atom as a function of the power of a continuous-wave probe beam. Since the saturated photon emission rate for a closed two-level system is $\Gamma/2$, where Γ is the inverse of the natural lifetime, the collection and detection efficiency can be obtained directly from the measured count rate.

This value is compatible with that obtained from a direct evaluation of the transmission of our detection system. The transmission of our lens is measured to be 87% and its collection solid angle is $0.15 \times 4\pi$ sr. Because the emission pattern for σ^+ -polarized photons is not isotropic, the effective solid angle of collection must be corrected by a factor of 85%. The transmission of the optical elements in the imaging system is 58%. Finally the light passes through a pinhole before illuminating the avalanche photodiode. The largest uncertainty is in the combination of the pinhole transmission and photodiode quantum efficiency, which is estimated to be around 10%. Multiplying all factors gives an overall collection and detection efficiency compatible with the 0.6% quoted above.