

# Deterministic Generation of Single Photons from One Atom Trapped in a Cavity: Supporting Online Material - Materials and Methods

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## EXPERIMENTAL DETAILS

Our experimental setup is depicted by the simple drawing in Fig. 1A of the manuscript, with many of the technical aspects described in more detail in Refs. [S1, S2]. After releasing a cloud of atoms from a magneto-optical trap (MOT) above the cavity, transverse cooling beams illuminate the cavity region, at which point an atom can be loaded into the intracavity far-off resonance trap (FORT), which is matched to a standing-wave, TEM<sub>00</sub> mode along the cavity axis. The trap depth is  $U_0/k_B = 2.3$  mK (47 MHz), and because its wavelength is  $\lambda_F = 935.6$  nm, the potential for the atomic center-of-mass motion is only weakly dependent on the atom's internal state [S2]. The cavity length is actively stabilized with an auxiliary laser at wavelength  $\lambda_C = 835.8$  nm that does not interfere with the trapping or the cQED interactions. Relevant cavity parameters are length  $l_0 = 42.2$   $\mu\text{m}$ , waist  $w_0 = 23.6$   $\mu\text{m}$ , and finesse  $\mathcal{F} = 4.2 \times 10^5$  at 852 nm.

For our system, the Rabi frequency  $2g_0$  for a single quantum of excitation is given by  $g_0/2\pi = 16$  MHz, where  $g_0$  is based upon the reduced dipole moment for the  $6S_{1/2}, F = 4 \leftrightarrow 6P_{3/2}, F' = 3'$  transition in atomic Cs (Fig 1B). The amplitude decay rates ( $\kappa, \gamma$ ) due to cavity losses and atomic spontaneous emission are  $\kappa/2\pi = 4.2$  MHz, and  $\gamma/2\pi = 2.6$  MHz. Since  $g_0 \gg (\kappa, \gamma)$ , strong coupling is achieved, resulting in critical photon and atom numbers  $n_0 \equiv \gamma^2/(2g_0^2) \simeq 0.013$ ,  $N_0 \equiv 2\kappa\gamma/g_0^2 \simeq 0.084$ .

With an atom loaded into the intracavity FORT, our protocol for the generation of single-photon pulses consists in illuminating the atom with a sequence of laser pulses according to the timing diagram shown in Fig. 1(c) of the manuscript. Within each trial, the first pulse  $\Omega_3(t)$  contains light tuned 10 MHz blue of  $F = 3 \rightarrow F' = 3'$ , which initiates the adiabatic transfer  $F = 3 \rightarrow 4$  between the ground hyperfine levels, with the emission of a photon into the cavity mode. This transformation is principally accomplished via “dark” eigenstates of the atom-cavity system, with no contribution from the excited level  $F' = 3'$ , and hence with a concomitant reduction of fluorescent loss [S3, S4, S5]. The second pulse  $\Omega_4(t)$  is tuned 17 MHz blue of  $F = 4 \rightarrow F' = 4'$  and recycles the atom back to the  $F = 3$  ground state through spontaneous decay  $F' = 4' \rightarrow F = 3$ . Each  $\Omega_{3,4}$  field consists of two orthogonal pairs of counter-propagating

beams in a  $\sigma^+ - \sigma^-$  configuration. The detuning between the  $3' \rightarrow 4$  transition at  $\omega_{43}$  and the cavity resonance  $\omega_C$  is  $\Delta_{CA} \equiv \omega_C - \omega_{43} = 2\pi \times 9$  MHz [S6].

We now provide some additional details on the optical path from the cavity to the detectors. After emerging from the vacuum chamber window, the path includes a polarizing beam splitter (PBS), several dichroic mirrors and two interference filters. The light is next coupled into a single-mode fiber, and then split using a 50/50 fiber coupler. The two output fibers of the coupler are connected to fiber-coupled avalanche photodiodes (APD), labelled  $D_A$  and  $D_B$ .

## LOSSES AND EFFICIENCIES

Photons generated in the cavity are subject to various types of loss along their path to the detectors. These are summarized in Table S1.

Independent diagnostic measurements reveal the ratio of scattering/absorption losses to transmission of our cavity mirrors [S7]. From this we infer the cavity escape efficiency  $\alpha_e$ . Our cavity is also symmetric (nominally identical mirrors) meaning that 1/2 of the generated photons leave through the mirror  $M_1$  and are not detected ( $\alpha_{2s}$ ). Once the light exits the vacuum chamber, the unpolarized stream of output photons is first reflected from the PBS, resulting in a 50% loss ( $\alpha_{PBS}$ ). The remaining optics, including the fiber coupling efficiency, on the way to the APD's gives the quantity  $\alpha_P$ . The quantum efficiency  $\alpha_{APD}$  of the APD's is also independently measured.

The uncertainty in  $\alpha_e$  comes from the difficulties related to mode matching and scattering in the reflection dip measurement we used [S7]. The efficiencies  $\alpha_P$  and  $\alpha_{APD}$  are obtained using measurements of the fiber transmission, which has fluctuations in our system of around  $\pm 10\%$ . The values and uncertainties enumerated in Table S1 combine to give a total efficiency of escape, propagation and detection

$$\alpha = \alpha_e \times \alpha_{2s} \times \alpha_{PBS} \times \alpha_P \times \alpha_{APD} = 0.024 \pm 0.004 \quad (1)$$

From these efficiencies, their associated uncertainties and our measurements of photon statistics of the emitted light, we infer that each generation attempt succeeds with probability  $\phi_G = 1.15 \pm 0.18$ . The fact that we suc-

ceed with efficiency consistent with unity (within error) derives from the strong coupling of atom and cavity field.

## DATA ANALYSIS

### Determination of the Presence of a Trapped Atom

For each attempt to load an atom into the FORT, we obtain an output stream of photoelectric events. The first step in the analysis of such an output stream is to determine whether an atom was indeed loaded into the trap. The procedure is as follows: we assume an atom is present at time  $t$  if more than  $n_p$  photons total were recorded during the  $W_d$  detection windows immediately prior to  $t$ . The result is not sensitive to the exact values of  $n_p$  and  $W_d$ ; typical values are  $n_p = 1-5$ ,  $W_d = 500$  (i.e., 5 ms).

### Calculation of $C(\tau)$

Counts recorded outside the gating intervals  $[t_0^j - \frac{1}{2}\delta t, t_0^j + \frac{3}{2}\delta t]$  are removed from the record due to an excess of stray light between trials  $\{j\}$ . This occurs because of our use of an optically pumped Cs cell in the output path for the purposes of filtering residual scattered  $\Omega_3$  light. These records are then converted into a pair of lists  $(a_1^k, \dots, a_N^k)$  and  $(b_1^k, \dots, b_N^k)$  for detectors  $D_A$  and  $D_B$ , respectively. The  $n^{\text{th}}$  entry in each list is 1 if a photoelectric event was recorded in the time interval  $[n\delta, (n+1)\delta]$ , and zero otherwise, where  $\delta$  is the time resolution of our data acquisition system. The correlation function is obtained by convolving the two lists against each other, and then convolving the result against a smoothing function  $f(t) = (2\pi\sigma^2)^{-1/2}e^{-t^2/2\sigma^2}$ , with  $\sigma = 20$  ns:

$$C(\tau) = \sum_m f(\tau - m\delta) \sum_k \sum_n a_n^k b_{n+m}^k. \quad (2)$$

## TWO-EVENT PROCESSES

We have investigated several optical processes that might yield more than one photon per trial, including the atom's recycling by stray light from the  $\Omega_4$  beam or by diverse off-resonant excitation mechanisms, including from the FORT itself. We conclude that none of these are responsible for the observed disparity between  $R$  and  $R_B$ . Instead, as demonstrated by Fig. 4 in our manuscript, the excess coincidences likely arise from infrequent events in which two atoms are trapped within the cavity, each atom contributing one photon during the detection window.

## Model for Inferring the Two-Atom Probability

In a straightforward fashion, we derive that the photodetection probabilities  $\tilde{P}_1, \tilde{P}_2$  for one and two-photon detection in the absence of detector dark counts are:

$$\begin{aligned} \tilde{P}_1 &= \eta_I \alpha \phi_G + 2\eta_{II} \alpha \phi_G (1 - \alpha \phi_G) + P_S \\ \tilde{P}_2 &= \eta_I \alpha \phi_G P_S + \eta_{II} \alpha \phi_G (\alpha \phi_G + 2P_S (1 - \alpha \phi_G)) + P_S^2. \end{aligned} \quad (3)$$

The principal assumptions in the derivation of these equations are (i) that there are at most two atoms in the cavity and (ii) that two atoms act independently inside the cavity both in their decay from the trap (assumption supported by separate measurements) and in their generation of ideal single photons.

In Eq. (3),  $\phi_G$  is the probability that one atom within the cavity will generate one photon during a given trial  $j$ , and  $P_S = P_B - P_D$  is the probability of a background event other than from detector dark counts in the detection window  $[t_0^j, t_0^j + \delta t]$  (i.e., scattered light). From the measured photoelectric counting statistics, the known probability  $P_S = 4.9 \times 10^{-5}$ , and the overall efficiency  $\alpha$ , we can solve this pair of equations for the two unknowns  $\phi_G$  and  $\eta_{II}$ . For the first bin of Fig. 4 of the manuscript (with the values  $\tilde{P}_1 = 0.0285, \tilde{P}_2 = 2.68 \times 10^{-5}$  derived from  $P_1, P_2$  by correcting for dark counts), we find  $\alpha \phi_G = 0.0276$  and  $\eta_{II} = 0.033$ .

The full curve in Fig. 4 of our manuscript is obtained by employing these values for  $\alpha \phi_G$  and  $\eta_{II}$  as initial conditions to deduce the time dependence of the photon statistics over the duration of the trapping interval. The time dependence of  $\eta_I, \eta_{II}$  is modelled by simple rate equations with initial conditions  $(\eta_I^0, \eta_{II}^0) = (0.967, 0.033)$ . We also use the experimentally determined decay rate  $\Gamma_1 = 1/0.14$  s, and the assumption  $\Gamma_2 = 2\Gamma_1$ .

Description	Symbol	Value	Error
Cavity escape	$\alpha_e$	0.6	0.1
Two-sided cavity	$\alpha_{2s}$	0.50	N/A
Polarizer	$\alpha_{PBS}$	0.50	N/A
Propagation	$\alpha_P$	0.32	0.03
Detection	$\alpha_{APD}$	0.49	0.05
Total	$\alpha$	0.024	0.004

TABLE S1: List of efficiencies associated with photon propagation and detection.

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