

Observation of intensity squeezing in resonance fluorescence from a solid-state device

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Abstract

Intensity squeezing¹—i.e., photon number fluctuations below the shot noise limit—is a fundamental aspect of quantum optics and has wide applications in quantum metrology²⁻⁴. It was predicted in 1979 that the intensity squeezing could be observed in resonance fluorescence from a two-level quantum system⁵. Yet, its experimental observation in solid states was hindered by inefficiencies in generating, collecting and detecting resonance fluorescence. Here, we report the intensity squeezing in a single-mode fibre-coupled resonance fluorescence single-photon source based on a quantum dot-micropillar system. We detect pulsed single-photon streams with 22.6% system efficiency, which show subshot-noise intensity fluctuation with an intensity squeezing of 0.59 dB. We estimate a corrected squeezing of 3.29 dB at the first lens. The observed intensity squeezing provides the last piece of the fundamental picture of resonance fluorescence; which can be used as a new standard for optical radiation and in scalable quantum metrology with indistinguishable single photons.

A fundamental tool in optical quantum information science is the single-photon source⁶⁻⁷, which ideally should emit one, and only one, indistinguishable light quanta on demand. The absence of two- or more-photon components gives rise to antibunching—a highly nonclassical behaviour—which is relevant for quantum cryptography applications⁸. The indistinguishability between independent single photons, which can be measured by two-photon Hong-Ou-Mandel quantum interference⁹, lies at the heart of quantum teleportation¹⁰, quantum networks¹¹, and linear optical quantum computing^{12,13}.

In principle, an ideal single-photon device is also useful for quantum metrology. For example, there is a growing interest in redefining the international standard base unit of luminous intensity, candela, by counting the number of fundamental light quanta, photons¹⁴. Compared to conventional light sources, such as lasers or light-emitting diodes, an ideal single-photon source has the unique quantum advantage that its intensity uncertainty can be greatly suppressed, thus offer a way to beat the fundamental shot noise limit. Such a feature would find applications in various fields, such as imaging or measurement in the ultralow power regime of light-sensitive biological samples^{15,16}.

While the high single-photon purity, indistinguishability, and extraction efficiency has been demonstrated¹⁷⁻¹⁹, the simultaneous observation of intensity squeezing in a single-photon source remains challenging. This is due to the inevitable photon loss from the generation, transmission, collection, and detection in experiments; such that, in each time slot, there was much more often vacuum rather than one photon detected. While the low overall efficiency only affected the data accumulation time in the measurements of the single-photon purity and indistinguishability, the intensity squeezing, which heavily relies on the overall efficiency; can be diminished by the low efficiency and washed out in the presence of fluctuation of the experimental parameters. Therefore, there were rarely observations of intensity squeezing in single-photon sources. The first observation, with a very small squeezing of $\sim 0.00183 \pm 0.00038$ dB, dated back to 1983 from a single atom¹. Recently, a single

molecule embedded inside a metallodielectric antenna was designed for a strong enhancement of photon collection efficiency and showed a squeezing of 2.2 dB²⁰. However, these single photons were not in a single mode, and due to the incoherent excitation, they were distinguishable. A related work reported intensity squeezing with confined polaritons in semiconductor micropillars²¹.

Recently, semiconductor quantum dots embedded in microcavities emerged as a scalable solid-state platform for quantum information technologies²²⁻²³. Pulsed and resonant excitation of the single quantum dots was used to efficiently generate single photons with near-unity purity and indistinguishability²⁴. Polarized microcavities were deterministically coupled to the emitter and efficiently funnelled the single photons into a single spatial mode²⁵. In all physical systems, the long sought-after goal of observing intensity squeezing in pulsed resonance fluorescence remained a challenge. Here, we report the first single-mode fibre-coupled semiconductor source of indistinguishable single photons with subshot-noise intensity fluctuation.

The reduction of the intensity fluctuation in a single-photon source compared to the shot noise can be expressed as²⁰: $N_{SPS} / N_{SN} = \sqrt{1 - \rho T}$, where ρ is the internal efficiency of the quantum emitter (including its quantum efficiency and excited-state preparation efficiency), and T denotes the total external efficiency (including extraction efficiency and the overall transmission efficiency in the experiment setup). Due to the quantum uncertainty principle, when the uncertainty of the intensity is smaller than the shot noise, the uncertainty of its conjugate variable—phase—is accordingly increased.

In our experiment, we use a self-assembled semiconductor InAs/GaAs quantum dot coupled in a 2- μ m diameter micropillar cavity. The sample is cooled at 4 K where the emitter is resonant with the cavity, so that both the efficiency and indistinguishability are improved due to the Purcell effect. A confocal microscope is utilized to excite the emitter and collect resonance fluorescence single photons. A cross-polarization set-up suppresses the laser leakage with an extinction ratio of $\sim 10^7:1$.

First, we study the resonance fluorescence single photons under continuous-wave

(CW) laser excitation. Due to the high photon flux and the limited recovery time (~ 5 ns) of the superconducting nanowire single-photon detectors used, we attenuate the single-photon stream by 1000 times. The corrected single-photon count rate as a function of laser power is shown in Fig. 1(a), from which we extract a saturation power of $P_{sat} = 4.9$ nW and a saturated photon flux of $I_{\infty} = (1.87 \pm 0.03) \times 10^9 / s$. This is so far the brightest single-photon source reported in any physical system.

A CW laser can prepare the emitter in its excited state with a maximal probability of 50% in the limit of high laser power²⁶. The theoretically predicted single-photon flux is $1/2T_1$, where T_1 is the excited state lifetime. Fig. 1(b) shows a time-resolved resonance fluorescence measurement which gives $T_1 = 58.60 \pm 0.02$ ps. Thus the

overall single-photon efficiency is $\frac{I_{\infty}}{1/2T_1} = 0.219 \pm 0.004$. Compared to the average

lifetime of many quantum dots in the slab, $T_{slab} = 1.08$ ns, we estimate the Purcell factor to be ~ 18.4 .

Next, we resonantly pump the quantum dot by a pulsed laser with a bandwidth of ~ 50 GHz to match the cavity mode. The detected single-photon counts show a Rabi oscillation as a function of excitation power, as plotted in Fig. 1(c). At π pulse, with a repetition rate of 76 MHz, we finally detect 17.2 million single photons per second on a superconducting nanowire single-photon detector with an efficiency of $\sim 86\%$. The corresponding overall single-photon efficiency is 22.6%, in a good agreement with the value (21.9%) extracted from CW excitation.

The single photon nature of generated resonance fluorescence is demonstrated by a Hanbury Brown and Twiss measurement that shows $g_2(0) = 0.025(1)$ at a π pulse.

Fig. 1(d) is the measured high-resolution spectrum of the resonance fluorescence single photons, showing a full width at half maximum of 2.74 ± 0.02 GHz. By fast Fourier transformation, the fitted coherent time is $T_2 = 108.8 \pm 0.9$ ps. This allows us to estimate that $T_2 / 2T_1 = 0.928 \pm 0.008$, indicate that the pulsed single photons are

92.8% Fourier transform limit. This is in a good agreement with the measured photon indistinguishability via Hong-Ou-Mandel interferences that show a corrected visibility of 0.935(1) for two single photons separated by $\sim 10 \mu\text{s}$.

We now examine the intensity squeezing of resonance fluorescence. The single photons are directly detected by a superconducting nanowire single-photon detector, and the arrival time is recorded by a time-to-digit converter. Figure 2(a) is the real-time monitoring of resonance fluorescence single photon counts at π pulse with a time bin of $1.0 \mu\text{s}$. The average count per time bin is $17.5/\mu\text{s}$. The corresponding histogram is plotted in Fig. 2b. The directly observed standard deviation of photon counts is

$N_{SPS} = 3.65$, which is significantly below the shot noise limit of

$N_{SN} = \sqrt{17.48} = 4.18$. Therefore, the directly measured intensity squeezing is

$N_{SPS} / N_{SN} = 87.32\%$ (0.59 dB) at the π pulse. This value is very close to, but slightly smaller than the theoretically predicted value of 87.75%. This is due to the imperfect recovery time of the detector (see supplementary information). As a comparison experiment, we replace the single-photon source by a CW laser attenuated to an average count rate of ~ 17.2 million per second and send it into the same detector. The observed standard deviation is 99.98% of the shot noise limit. This indicates that the observed subshot-noise intensity fluctuation indeed comes from the single photons.

In Fig. 2(b), the occurrences at different single-photon counts are plotted (deep blue) and fitted by a binominal function (see Supplementary Information), which is reduced by 12.68% compared to the shot noise limit (green). Considering the optical transmission loss, cross-polarization filtering loss, coupling loss into the single-mode fiber, and the detection efficiency at the detector, the intensity squeezing out of the first lens should be 3.29 dB (light blue). Fig. 2(c) shows the amount of intensity squeezing as a function of the excitation laser amplitude. As expected, by tuning the laser power gradually to π pulse, the excited-state population—proportional to the single-photon generation efficiency—grows, which increases the amount of intensity squeezing.

It is clear that the intensity squeezing critically relies on the overall efficiency. We analyse the photon loss budget in detail. First, we estimate the external efficiency, which accounts for all photon loss from the first lens to the single-photon detector: collection efficiency by the first objective, polarization filtering, single-mode fibre coupling, and detection. To estimate the single-mode fibre coupling efficiency, we performed a separate experiment using multimode fibres (with diameters of 100 μm , 200 μm , which gives the same counts, indicating the efficiency is saturated and equal to unity). The ratio of single-photon count using a single-mode fibre is measured to be 74% of that using multimode fibres, which determines the single-mode coupling efficiency. The overall transmission efficiency of the optical path (including the first lens, optical window, polarized beam splitter, and two wave plates) is measured to be 83%. The first lens collection efficiency is estimated to be 78%. The superconducting nanowire single-photon detection efficiency is 86%. The micropillar cavity shows a small mode splitting due to a slight ellipticity of its cross section. In this case, the intensity of one eigenmode will be higher than that of another one³². By measuring the single photon counts in two modes respectively, the determined polarization filtering efficiency is 55%.

The internal efficiency of the quantum dot-micropillar system includes the quantum efficiency of the emitter (QE), excited-state preparation efficiency (ESPE), and the photon extraction efficiency (PEE) out of the micropillar. The QE and ESPE can be affected by power-induced²⁷ and phonon-induced damping²⁸ in quantum dots. The PEE accounts for the losses due to the side leakage and the part not coupled to the fundamental mode of the microcavity, which can be estimated by²⁹

$$\eta_c = \frac{F_p}{F_p + 1} \frac{Q}{Q_0},$$

where F_p is the Purcell factor, Q is the quality factor, and Q_0 is the quality factor of planar microcavity. In this experiment, $F_p=18.4$, $Q=6800$, $Q_0=7600$, so the photon extraction efficiency by the microcavity is $\sim 85\%$. This result is consistent with the numerically simulated number of 87% using finite difference time domain. The

product of QE and ESPE is estimated to be 92%.

In summary, we have directly observed the first intensity squeezing in pulsed resonance fluorescence, in a single-mode fibre-coupled highly-indistinguishable solid-state single-photon source. The uncorrected squeezing at the output of the single-mode fibre is 0.59 dB, and the corrected intensity squeezing at the first lens is 3.29 dB. From a fundamental perspective, our work fills the a long sought-after missing element in textbook quantum optical phenomena²⁶ using quantum dots. This is addition to the previously reported antibunching³⁰, two-photon interference³¹, weak³² and strong^{33,34} coupling, Rabi oscillation³⁵, Autler-Townes splitting³⁶, coherent population trapping³⁷, Mollow triplet^{38,39}, and quadrature squeezing⁴⁰. For practical applications, our work combines for the first time high levels of single-photon efficiency, purity and indistinguishability together with intensity squeezing in a semiconductor chip. The intensity squeezing can be further improved by driving the quantum dot with dichromatic laser pulses⁴¹ and/or coupling the quantum dot to polarized microcavities¹⁹. We note that such an intensity squeezed single-photon source with subshot noise fluctuations has natural application to benchmarking single-photon detector efficiency⁴², redefining the standard base unit of luminous intensity at ultralow power level⁴³, and optical spectroscopy of light-sensitive biological samples^{15,16}.

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Figure captions:

Figure 1: (a) Single-photon counts with CW laser resonant excitation. The saturation count is 1.87 billion per second. The data is fitted by the Eqn. (2). (b) Time-resolved measurement of the resonance fluorescence by a superconducting nanowire single photon detector with a time resolution of ~ 20 ps. The fitted lifetime of quantum dot is $T_1 = 58.60 \pm 0.02$ ps. Comparing to the quantum dot lifetime in the slab (~ 1.08 ns), the Purcell factor is 18.4. (c) Rabi oscillation under pulsed resonant excitation. At π pulse, 17.2 million pure single photons are detected per second, by a superconducting nanowire single-photon detector with an efficiency of 0.86. (d) High-resolution spectrum of resonance fluorescence measured by a Fabry-Perot cavity with a frequency resolution of 220 MHz and a free spectral range of 37.4 GHz. The data is fitted by a Voigt function (red line), and the linewidth is 2.74 ± 0.02 GHz.

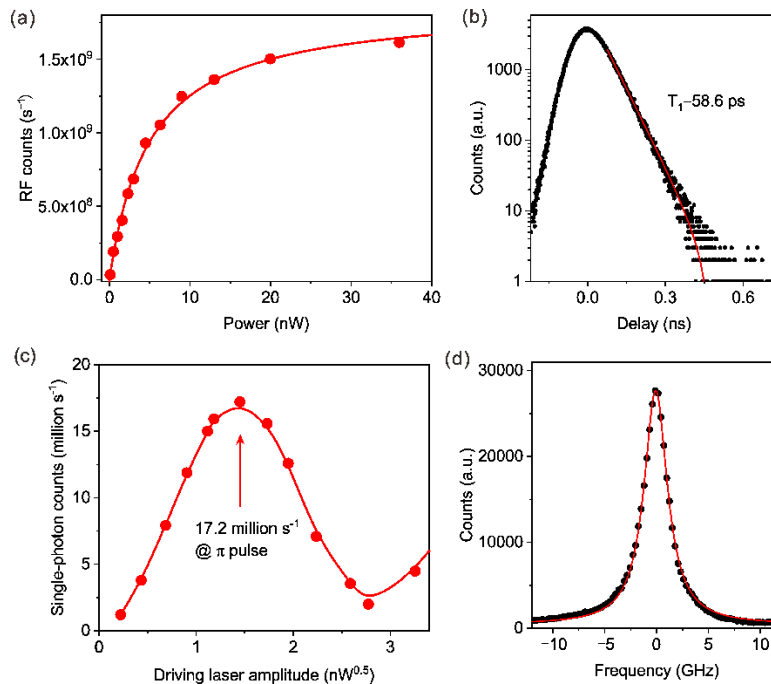


Figure 2: (a) Real-time monitoring of resonance fluorescence single photon counts at π

pulse with a time bin of $1.0 \mu\text{s}$. The average count per time bin is $17.5/\mu\text{s}$. (b) The corresponding histogram where the frequency of observed counts (i.e., the intensity fluctuation, shown as deep blue dot) is fitted by a binomial function. Comparing to the shot-noise-limited source with the same intensity (green line), the quantum dot single-photon source shows a 12.68% reduction of histogram linewidth, that is, intensity fluctuation noise. Blue line displays the corrected intensity squeezing at the first lens (see main text). (c) Intensity squeezing parameter as a function of the excitation laser amplitude. At π pulse, the intensity squeezing reaches a minimal value of 87.32%. Black dotted line is the normalized shot-noise limit.

