Strongly correlated photons on a chip

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Optical nonlinearities at the single-photon level are key ingredients for future photonic quantum technologies. Prime candidates for the realization of the strong photon–photon interactions necessary for implementing quantum information processing tasks, as well as for studying strongly correlated photons in an integrated photonic device setting, are quantum dots embedded in photonic-crystal nanocavities. Here, we report strong quantum correlations between photons on picosecond timescales. We observe (i) photon antibunching upon resonant excitation of the lowest-energy polariton state, proving that the first cavity photon blocks the subsequent injection events, and (ii) photon bunching when the laser field is in two-photon resonance with the polariton eigenstates of the second Jaynes–Cummings manifold, demonstrating that two photons at this colour are more likely to be injected into the cavity jointly than they would otherwise. Together, these results demonstrate unprecedented strong single-photon nonlinearities, paving the way for the realization of a quantum optical Josephson interferometer or a single-photon transistor.

Cavity quantum electrodynamics (cavity–QED) studies the quantum limit of the light–matter interaction when a single two-level quantum emitter is coupled to a single cavity mode. In the strong coupling regime of cavity–QED where the coherent interaction strength between the emitter and the cavity mode exceeds the dissipative rates, the energy level diagram is described by the anharmonic Jaynes–Cummings ladder (Fig. 1a). Transition energies from the ground state $|g\rangle$ to the first Jaynes–Cummings manifold $|\pm\rangle$ are distinct from transition energies higher up in the ladder. As a consequence, this system embodies the ultimate nonlinear optical device, enabling the observation of photon–photon interactions at the single-photon level. Various implementations of cavity–QED systems have been reported with atoms in highly-finesse cavities, with quantum dots (QDs) in different types of monolithic cavities and in the microwave domain. Recent experiments in the optical domain using a single atom coupled to Fabry–Perot or toroidal cavities have demonstrated the photon blockade effect by observing photon antibunching in correlation measurements. Photon bunching upon two-photon excitation of the second Jaynes–Cummings manifold has been observed for a single-atom cavity–QED system. In the solid state, early results in QD cavity–QED systems indicating optical nonlinearities have been reported.

Here, we show that a single QD deterministically coupled to a photonic-crystal defect cavity exhibits pronounced photon antibunching; this is indicated by a reduction in two-photon scattering events by more than 40% when resonantly exciting the fundamental system transitions (polaritons), demonstrating photon blockade. We also tuned either the laser or the cavity mode frequency to ensure resonant two-photon excitation of the higher polariton states. Because the ratio of the probability amplitudes for having two and one cavity photons is enhanced in this case compared to the Poissonian limit, we observe that the photon emission is strongly bunched by more than 50% in the maximal case. This observation is enabled by the high degree of anharmonicity of the strongly coupled QD–cavity system and allows for the first demonstration of spectrally resolved direct access to the second manifold of the Jaynes–Cummings ladder in an integrated solid-state photonic device. By positioning an InAs/GaAs QD at the electric-field maximum of a photonic-crystal defect cavity in L3 geometry, we achieved a coherent coupling constant $g$ corresponding to 141 μeV. Using photoluminescence spectroscopy together with correlation measurements, we first ensured that there was one, and only one, QD inside the photonic-crystal membrane. The cavity had a quality factor $Q$ of ~25,000, corresponding to a cavity linewidth $\kappa$ of 53 μeV. The ratio $g/\kappa \approx 2.7$ provides a measure of the degree of anharmonicity of the coupled QD–cavity system. To tune the cavity frequency, we used a thin-film deposition technique with nitrogen gas injection. A crossed-polarization technique ensured efficient suppression of the excitation-laser light, which was back-reflected from the sample surface. The set-up is sketched in Fig. 1b.

A large majority of QDs, as well as most other solid-state-based emitters such as nitrogen-vacancy centres, exhibit the phenomenon of blinking, which is induced by a spontaneous or induced change of the internal state of the QD, leading to an intermittent optical response. Experimentally, we found that the fractional amount of time our system spends in the neutral QD ground state strongly decreases with increasing resonant probe power, and is vanishingly small above 1 nW (see Supplementary Information). Incident laser photons simply scatter off the uncoupled cavity, resulting in a spectrum as shown in Fig. 1c (black trace). To overcome this problem, we used an off-resonant laser just below the edge of the wetting layer to re-pump the QD into its neutral ground state. By alternating re-pump and probe intervals (see Methods), we partly recovered the polariton spectrum shown in Fig. 1c (red trace).

We carried out correlation measurements using resonant pulses with a duration of $T_{\text{pulse}} \approx 72$ ps (full-width at half-maximum, FWHM), yielding a ratio between pulse length and polariton lifetime of $\kappa/2T_{\text{pulse}} \approx 2.9$. Figure 2 presents correlation histograms for different laser/cavity detunings. The respective detunings are indicated in the resonant scattering spectrum shown in Fig. 2a. We defined the correlation function for pulsed excitation at zero time delay $g_{\text{pulsec}}(0)$ as the area of the central peak divided by the average area of peaks at other times (see Supplementary Information). First, we chose the frequency of the pulsed laser to match the upper polariton transition and obtained significant
bunching feature on the two-photon transition, have their origin in the case of photon blockade, as well as the moderate ladder. We emphasize that the non-vanishing correlations at zero ground state to the second manifold of the Jaynes–Cummings bunching has its origin in a two-photon transition from the corresponding correlation histogram with $g^{(2)}$, we observed photon bunching. Figure 2e presents a correlation histogram with $g^{(2)}$ for various cavity and laser detunings from the exciton changed their sign. The kets $|n, +/−\rangle$ denote the upper/lower polariton states of the $n$th manifold, where $n$ is the number of energy quanta in the system. When exciting a state of the first manifold with a resonant laser (blue arrow), the second manifold cannot be accessed (photon blockade). Sketch of the experimental set-up with crossed-polarized laser suppression and a Hanbury Brown–Twiss set-up. APD, avalanche photodiode. c, On-resonance continuous-wave (c.w.) scattering spectrum for a probe power of 1 nW. The black trace was recorded without the additional re-pump laser. With the re-pump switched on, the resonant signal from the polaritons is restored (red trace).

antibunching with $g^{(2)}_{\text{pulsed}}(0) = 0.75 ± 0.06$ (Fig. 2c). For comparison, we turned off the re-pump laser and verified that photons scattered from the uncoupled cavity mode had Poissonian statistics (Fig. 2b). As a cross-check, we also confirmed that the scattered photons from a resonantly driven cavity mode far detuned from all QD transitions exhibited Poissonian statistics. These experiments together demonstrated that the applied re-pump laser could be used to switch on the single-photon nonlinearity and to control the statistics of the scattered photons. We emphasize that the degree of photon antibunching observed in the experiment is limited by the finite excitation pulse length. Accordingly, a higher degree of antibunching ($g^{(2)}_{\text{pulsed}}(0) = 0.55 ± 0.03$) is observed when the cavity is off-resonant from the exciton, as shown in Fig. 2d for the lower polariton in the case of blue-detuning of the cavity mode. Here, the polariton state lifetime is prolonged, making multiple photon absorption events within a single pulse less likely. Experimentally, we did not observe a difference between correlations recorded on the upper and lower polaritons if the respective laser and cavity detunings from the exciton changed their sign.

When tuning the laser photon energy to half the energy of the lower polariton eigenstate of the second Jaynes–Cummings manifold, we observed photon bunching. Figure 2e presents a corresponding correlation histogram with $g^{(2)}_{\text{pulsed}}(0) = 1.5 ± 0.1$. The bunching has its origin in a two-photon transition from the ground state to the second manifold of the Jaynes–Cummings ladder. We emphasize that the non-vanishing correlations at zero time delay in the case of photon blockade, as well as the moderate bunching feature on the two-photon transition, have their origin mostly in the particular implementation of the measurement using pulsed laser excitation and slow single-photon detectors. To confirm this and to explain the principal experimental features, we carried out numerical simulations of $g^{(2)}_{\text{pulsed}}(0)$ using a Monte Carlo wavefunction (MCWF) approach (see Supplementary Information). Here, we directly accounted for the pulsed laser excitation, the non-zero laser background due to imperfect extinction of the laser reflection, and the uncoupled cavity resonance due to the blinking of the dot. For the experimental settings corresponding to Fig. 2c–e, we estimated the temporal maximum of the average number of energy quanta in the Jaynes–Cummings system to be 0.12, 0.086 and 0.020, based on these MCWF simulations. Figure 3a displays the calculated autocorrelation function at zero time delay $g^{(2)}_{\text{pulsed}}(0)$ for varying cavity and laser detunings. The blue and red dashed lines denote the expected positions of the polariton frequencies and the two-photon resonances, respectively. The latter correspond to the optimal second-rung excitation conditions.

In addition to the bunching features originating from two-photon resonances, we would normally expect strong bunching
when the external laser that drives the cavity mode is resonant with the bare exciton, as can be seen in Fig. 3a (dotted vertical line). Unlike in previous experiments, we do not observe a bunching behaviour due to the observed QD blinking and a quantum interference based on an electromagnetically induced transparency effect (see Supplementary Information). Instead, our experiments demonstrate, for the first time, a coupled QD–cavity device realizing the Jaynes–Cummings model. However, photon antibunching can also be observed when the coupled emitter–cavity system is not in the strong coupling regime. In the simulations, we assumed a pure QD dephasing rate of $\hbar \gamma_{\text{deph}} = 13 \text{ meV}$ in addition to the cavity dissipation rate $\kappa$, consistent with the polaron linewidths observed in the experiment. This line broadening might stem from charge fluctuations in the QD environment, partially induced by the re-pump laser, and from phonon-induced dephasing.

In Fig. 3b,c we probed the upper and lower polariton branches for constant cavity detunings of $(−0.33 ± 0.04)g$ and $(1.04 ± 0.03)g$, respectively, by tuning the laser wavelength. The continuous change in photon correlations from strong antibunching to bunching maps out the (anharmonic) spectrum of the cavity–Jaynes–Cummings ladder. We also performed correlation measurements for varying cavity–exciton detunings at a constant laser–exciton detuning of $(0.94 ± 0.05)g$, demonstrating that the nature of strong photon correlations can be tuned by changing either the laser or cavity mode frequency. The agreement between the experimental values and theoretical expectations is very good in all cases, and we emphasize in this regard that the theoretical curves provide a prediction based on independently determined parameters. The dashed vertical lines in Fig. 3b–d correspond to the polaron energy (blue) and the energy of the two-photon transition to the second manifold (red), indicating the origin of both the antibunching and bunching features.

An obvious extension of the results presented here includes nonlinear optics experiments at the single-photon level, such as the realization of a single-photon transistor, where the laser excitation of a state of the second Jaynes–Cummings manifold is conditioned by a control laser exciting the fundamental manifold. Furthermore, our results elevate this system to the ultimate optical nonlinear building block for more complex structures investigating strong photon correlations in non-equilibrium settings, such as an optical Josephson interferometer or coupled arrays of nonlinear cavities. In combination with recent progress in the fabrication of site-controlled QDs within arrays of photonic-crystal cavities and the ability to tune QD transitions by up to 25 meV in p–i–n structures, the present work demonstrates the great potential of QD–cavity systems as candidates for quantum photon simulators.

**Methods**

**Sample fabrication and parameters.** The epitaxial structure of the sample consisted of a 12-nm-thick GaAs slab containing a layer of InGaAs QDs at the centre. Typical QD densities ranged from 0 to 3 $\mu$m$^{-2}$. The 130-nm-thick GaAs cavities were fabricated around the locations of individually selected dots. For the relative positioning of dots and cavities, atomic force microscopy (AFM) metrology was used as reported in a previous publication. The lattice constant of the photonic-crystal lattice was $a = 260$ nm, and the radius of the air holes $r = 60$ nm. Because of the precise positioning of the QD with an accuracy of $\sim 30$ nm at the electric field maximum of the cavity, the coherent coupling constant $g$ of the system was close to the optimum value for an L3 cavity structure and a self-assembled InGaAs QD.

**Optical characterization.** Optical characterization was performed in a liquid helium flow cryostat at $T = 5–7$ K. A $\times 50$ microscope objective (NA = 0.55) was used to illuminate the defect region of the photonic crystal with a spot size of $\sim 1$ $\mu$m. Emitted photons were collected with the same objective. After cooling, the cavity luminesced at a wavelength of $\sim 935$ nm, as determined by photoluminescence spectroscopy. To tune the cavity into resonance with the neutral excitation transition, we used a thin-film deposition technique with nitrogen gas injection. Even without active deposition of nitrogen, the cavity mode demonstrated an intrinsic red tuning at a rate of up to $0.008$ nm h$^{-1}$. For resonant scattering experiments, we excited the cavity mode with an actively power-stabilized narrow-bandwidth, mode-hop-free tunable diode laser. The sample was mounted so that there was a $45^\circ$ angle between the polarization axes of the cavity mode and the laser. By sending the collected light through a single-mode fibre with mounted fibre–polarization controllers and a subsequent analyser, we achieved precise control over the extinction of the reflected laser light. The photons scattered off the cavity were detected with an avalanche photodiode (APD) in single-photon counting mode.

**Off-resonant re-pump scheme.** When driving a polarization transition resonantly, a QD exhibits pronounced blinking; that is, either the QD charge changes or the electron or hole spin flips. To bring the QD back into the neutral ground state, we applied a pump–probe scheme at a repetition rate of 1 MHz. First, we probed the system for 500 ns. Next, with the APD readout disabled, we re-pumped the QD using an additional off-resonant laser pulse with a duration of 250 ns (using a c.w. Ti:sapphire laser at a wavelength of 857.2 nm, close to the wetting-layer resonance), followed by a waiting period of 250 ns before re-enabling the APD readout. Note that during the probe intervals, the re-pump laser was off, so we recorded autocorrelations of resonantly scattered photons only.

**Autocorrelation measurements.** For photon correlation measurements, we replaced the c.w. probe light by resonant laser pulses of a few picoseconds duration from a mode-locked Ti:sapphire laser, with a repetition rate of 76.3 MHz. To filter the broadband pulses we sent them through a 750 nm grating spectograph and coupled the diffracted light into a single-mode fibre. The transmitted pulse was approximately Gaussian, with a typical width of 0.018 nm. Using a streak camera we confirmed that the resulting pulses had near-Fourier-limited durations of $\sim 72$ ps. The dependence of photon correlations on the laser frequency were obtained by rotating the grating of the spectograph and consequently filtering out different parts...
of the pulse spectrum. Measurements depicting the dependence of photon correlations on the cavity resonance frequency on the other hand were carried out by exploiting an intrinsic cavity wavelength drift of ~0.006 nm h⁻¹ while keeping the laser frequency fixed. We performed autocorrelation measurements with a Hanbury–Brown and Twiss set-up, consisting of a 50:50 beamsplitter and an APD at each output. Photon arrival time differences of the two detectors were recorded and plotted in a histogram.

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Author contributions
A.R. and T.V. conducted the experiments, analysed the data and performed the simulations. M.W. made essential contributions to the experiment in its early stages. A.B., K.J.H. and A.R. and T.V. conducted the experiments, analysed the data and performed the simulations.

Additional information
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