Purification of a single-photon nonlinearity

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Single photon nonlinearities based on a semiconductor quantum dot in an optical microcavity are a promising candidate for integrated optical quantum information processing nodes. In practice, however, the finite quantum dot lifetime and cavity-quantum dot coupling lead to reduced fidelity. Here we show that, with a nearly polarization degenerate microcavity in the weak coupling regime, polarization pre- and postselection can be used to restore high fidelity. The two orthogonally polarized transmission amplitudes interfere at the output polarizer; for special polarization angles, which depend only on the device cooperativity, this enables cancellation of light that did not interact with the quantum dot. With this, we can transform incident coherent light into a stream of strongly correlated photons with a second-order correlation value up to 40, larger than previous experimental results, even in the strong-coupling regime. This purification technique might also be useful to improve the fidelity of quantum dot based logic gates.
Single-photon nonlinearities enabled by quantum two-level systems are essential for future quantum information technologies, as they are the building block of quantum photonics logic gates, deterministic entanglers of independent photons and for coupling distant nodes to form a quantum network. Near-unity fidelity interaction of photons with a two-level system such as an atom or quantum dot (QD) is enabled by embedding it into an optical cavity. The strong coupling regime, however, requires a small optical mode volume, which in turn makes it extremely hard to achieve polarization degeneracy of the fundamental cavity mode. This is due to unavoidable deviations from the ideal shape and intrinsic birefringence on the GaAs platform, precluding implementation of deterministic polarization-based quantum gates.

Here we show, using a nearly polarization-degenerate cavity in the weak coupling CQED regime, that we can transform incident coherent light into a stream of strongly correlated photons with \( g^2(0) = 25.7 \pm 0.9 \), corresponding to \( \gtrsim 40 \) in the absence of detector jitter. The polarization-degenerate cavity enables us to choose the incident polarization \( \theta_{\text{in}} = 45^\circ \) such that both fine-structure split QD transitions along \( \theta_{\text{QD}} = 0^\circ \) and \( \theta_{\text{QD}} = 90^\circ \) are excited, and we can use a postselection polarizer behind the cavity (\( \theta_{\text{out}} \)) to induce quantum interference of the two transmitted orthogonal polarization components (Fig. 1a). This leads to the appearance of two special postselection polarization angles \( \theta_{\text{out}}^\pm \) (depending on sample parameters), which can be used to restore perfect QD contrast (red curves in Fig. 1b). This compensates fully for reduced QD cavity coupling due to finite QD lifetime and QD cavity coupling strength, leading to complete suppression of transmission of the single-photon component in the low excitation limit. The transmission of higher-photon number states remains largely intact, allowing us to observe in Fig. 1c the strongest photon correlations to date in a solid-state system, reaching the range of strongly coupled atomic systems.

Figure 1 | The purification technique. (a) Cartoon of the experiment: polarization pre- and postselection in a resonant transmission CQED experiment enables tuning of the photon statistics from antibunched to bunched. (b) Theoretical resonant transmission spectra for coherent light with mean photon number \( \sim 2 \) (refs 15–18) have been obtained experimentally with QDs, which hardly exceeds even the classical case of thermal light following Bose statistics of \( g^2(0) = 2 \). In atomic systems with much longer coherence times, values up to \( \sim 50 \) have been obtained (ref. 6) and it is known that strict two-photon light sources exhibit diverging \( g^2(0) \) if the two-photon flux is reduced. Most related QD experiments to date have been operating in the strong-coupling regime of CQED, which is considered to be essential due to its photon-number-dependent energy structure (refs 6, 17, 18). In the weak-coupling regime, the energy structure is not resolved and it is an open question whether photon-number-dependent JC effects can still be observed. The strong coupling regime, however, requires a small optical mode volume, which in turn makes it extremely hard to achieve polarization degeneracy of the fundamental cavity mode. This is due to unavoidable deviations from the ideal shape and intrinsic birefringence on the GaAs platform, precluding implementation of deterministic polarization-based quantum gates.

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In terms of the second-order photon correlation function \( g^2(0) \), values up to \( \sim 2 \) (refs 15–18) have been obtained experimentally with QDs, which hardly exceeds even the classical case of thermal light following Bose statistics of \( g^2(0) = 2 \). In atomic systems with much longer coherence times, values up to \( \sim 50 \) have been obtained (ref. 6) and it is known that strict two-photon light sources exhibit diverging \( g^2(0) \) if the two-photon flux is reduced. Most related QD experiments to date have been operating in the strong-coupling regime of CQED, which is considered to be essential due to its photon-number-dependent energy structure (refs 6, 17, 18). In the weak-coupling regime, the energy structure is not resolved and it is an open question whether photon-number-dependent JC effects can still be observed. The strong coupling regime, however, requires a small optical mode volume, which in turn makes it extremely hard to achieve polarization degeneracy of the fundamental cavity mode. This is due to unavoidable deviations from the ideal shape and intrinsic birefringence on the GaAs platform, precluding implementation of deterministic polarization-based quantum gates.

Here we show, using a nearly polarization-degenerate cavity in the weak coupling CQED regime, that we can transform incident coherent light into a stream of strongly correlated photons with \( g^2(0) = 25.7 \pm 0.9 \), corresponding to \( \gtrsim 40 \) in the absence of detector jitter. The polarization-degenerate cavity enables us to choose the incident polarization \( \theta_{\text{in}} = 45^\circ \) such that both fine-structure split QD transitions along \( \theta_{\text{QD}} = 0^\circ \) and \( \theta_{\text{QD}} = 90^\circ \) are excited, and we can use a postselection polarizer behind the cavity (\( \theta_{\text{out}} \)) to induce quantum interference of the two transmitted orthogonal polarization components (Fig. 1a). This leads to the appearance of two special postselection polarization angles \( \theta_{\text{out}}^\pm \) (depending on sample parameters), which can be used to restore perfect QD contrast (red curves in Fig. 1b). This compensates fully for reduced QD cavity coupling due to finite QD lifetime and QD cavity coupling strength, leading to complete suppression of transmission of the single-photon component in the low excitation limit. The transmission of higher-photon number states remains largely intact, allowing us to observe in Fig. 1c the strongest photon correlations to date in a solid-state system, reaching the range of strongly coupled atomic systems (refs 6, 17, 18). In the following, a detailed
experimental and theoretical investigation of this effect, which can be seen as a purification of a single-photon nonlinearity, will be presented.

Results

Device structure. Our device consists of self-assembled InAs/GaAs QDs embedded in a micropillar Fabry–Perot cavity grown by molecular beam epitaxy25 (see Supplementary Fig. 1 and Supplementary Note 1). The QD layer is embedded in a P–I–N junction, separated by a 35 nm-thick tunnel barrier from the electron reservoir, to enable tuning of the QD resonance frequency by the quantum-confined Stark effect. For transverse mode confinement and to achieve polarization degenerate cavity modes, we first ion-etch micropillars of large diameter (35 µm) and slightly elliptical shape, then we use wet-chemical oxidation of an AlAs layer26 to prepare an intra-cavity lens for transverse-mode confinement27, avoiding loss by surface scattering at the side walls. Finally, we fine-tune the cavity modes by laser induced surface defects28,29 to obtain a polarization mode splitting much smaller than the cavity linewidth.

Device parameters and theoretical model. The system we study here is tuned to contain a single neutral QD within the cavity linewidth. The excitonic fine-structure splitting leads to 4.8 GHz linewidth. The excitonic fine-structure splitting leads to 4.8 GHz linewidth and its second-order photon correlation function \( g^{(2)}(0) \). The fundamental cavity modes show a residual polarization splitting of 4 GHz \( (f^X = 0 \ \text{GHz}, f^Y = -4 \ \text{GHz}) \) and the cavity axes are rotated by 5° with respect to the QD axes. To determine further system parameters, we model our QD cavity system by a two-polarization JC Hamiltonian coupled to the incident coherent field and take care of cavity and QD dissipation by the quantum master equation formalism30,31. We compare experiment and theory for six different input–output polarizer settings to faithfully determine the model parameters, these measurements were performed for an input power of 100 pW to avoid saturation effects32. We obtain \( g^{(2)}(0) \) measurements are compared in Fig. 1c), and the corresponding \( g^{(2)}(\tau) \) measurements are compared in Fig. 1c), both results agree very well to the convolution of the theoretically expected \( g^{(2)}(\tau) \) with the detector responses (Fig. 1c; see also Supplementary Fig. 3 and Supplementary Note 3).

Discussion

We have shown by experiment and theory that the reduced fidelity of a QD nonlinearity, caused by imperfect QD-cavity coupling, can be strongly enhanced by pre- and post-selection of specific polarization states. This enables transformation of a weak coherent input beam into highly bunched light with \( g^{(2)}(0) \approx 40 \), a value that has not been reached before, not even in the strong coupling regime. How is it possible to reach such high photon correlations, how does the polarization-based purification technique work?

We consider incident light with a frequency in the vicinity of one of the QD resonances, say \( \omega_{\text{QD}} \), and let us decompose the

Figure 2 | Coherent probing of the QD cavity system. Experimental (a) and theoretical (b) false colour plot of the columnwise normalized optical transmission as a function of the laser detuning \( \Delta \lambda_{\text{Laser}} \) and the polarization \( \theta_{\text{out}} \) (\( \theta_{\text{in}} = 45^\circ \)). The fine-split QD transition frequencies are at \( f_{0}\text{QD} = -2.4 \ \text{GHz} \) and \( f_{0}\text{QD} = 2.4 \ \text{GHz} \). The red circles indicate the special polarization conditions; the white square indicates the area explored in Fig. 3.
electromagnetic field transmitted through the cavity in two orthogonally polarized components: the signal Field $E_S$ polarized along the QD resonance polarization $\theta_{\text{QD}}^0 = 0^\circ$ and the local oscillator $E_{\text{LO}}$, which has interacted with an empty cavity, because it is polarized orthogonally to $\theta_{\text{QD}}^0$. Now, we consider three cases: (i) efficient interaction of the QD with incident light (cooperativity $C > 1$), (ii) intermediate interaction ($C \approx 1$) and (iii) weak interaction ($C \rightarrow 0$). The special polarization angles for various cooperativities are shown in Fig. 4.

In case (i), the QD leads to a nearly complete removal of the single-photon component from the incident coherent light polarized along the QD polarization: these photons are in principle perfectly reflected from the cavity and we simply have to detect along the same axis ($\theta_{\text{out}}^0 = \theta_{\text{QD}}^0 = 0^\circ$, see Fig. 4) to observe strong photon correlations. A significant proportion of higher photon number states are transmitted. As the second-order correlation function can be expressed in terms of the photon number distribution as $g^2(0) \propto 2P_2/P_1^2$, which is $N > 2$ photon number states, which for $P_2 < < P_1$ and $P_{N>2} < < P_2$, this leads to diverging photon correlations such as $g^2(0) \propto 1/\alpha^2$ if the single-photon component is attenuated as $P_1 \rightarrow \alpha P_1$.

Now in case (ii), for realistic systems, the finite lifetime of the QD transition and/or limited QD-cavity coupling $\gamma$ leads to a reduced cooperativity: even in the low-excitation limit, not every single-photon state is filtered out. Therefore, the signal field $E_S$ contains a fraction of coherent light reducing the photon bunching along the QD polarization $\theta_{\text{QD}}^0$, compare Fig. 3. This effect has been called ‘self-homodyning’ in literature. With the purification technique, we now rotate the postselection polarizer to interfere a portion of the local oscillator field $E_{\text{LO}}$ with the signal field, leading to the superimposed field $E_{\text{SLO}} = e^{i\phi_{\text{LO}}}E_S + e^{i\phi_{\text{LO}}}E_{\text{LO}}$. The polarizer angle controls the relative intensity of the two components and we can control the transmission phases $\phi_{\text{LO}}$ and $\phi_{\text{LO}}$ by adjusting the laser frequency, because the phases vary strongly in the vicinity of the QD and cavity resonances. We simply have to choose the local oscillator intensity that it matches the intensity of the portion of $E_S$ and adjust the phases for destructive interference. The result is that we detect in transmission mainly the single-photon filtered portion of $E_S$, which leads to very high photon correlations in the transmitted light despite limited cooperativity.

Finally, in case (iii) for $C \rightarrow 0$, only a vanishing fraction of the photons have interacted with the QD. We have to tune the postselection polarizer to $-45^\circ$ to destructively interfere nearly equal amounts of $E_S$ and $E_{\text{LO}}$ to observe enhanced photon correlations. This case is similar to that recently investigated in ref. 36, where (weak) photon bunching is observed for a relative phase of $\pi (\phi_{\text{LO}} - \phi_{\text{LO}} = \pi)$. We have a high-finesse ($F \approx 800$) cavity and significant cooperativity, which enables us to observe much stronger photon correlations (Supplementary Fig. 4 and Supplementary Note 4).

The special postselection angle $\theta_{\text{out}}^0$ and laser frequency have to be optimized numerically in principle, because pure dephasing cannot be taken care of in a semiclassical model. Despite this, we found that the special polarization angle shows approximately a very simple dependency on the cooperativity: $\theta_{\text{out}}^* \approx -45^\circ \exp(-C)$, see Fig. 4, which agrees well to our intuitive explanation here.

As a last point, we analyse the strong photon bunching in terms of the photon number distribution $P_n$. We use our theoretical model to determine $P_n$ as direct experimental determination thereof is strongly complicated by its sensitivity to loss. However, also the simulation of narrow-band photon number Fock input states is challenging in the quantum master model. Therefore, we use coherent input light and analyse the intra-cavity light in terms of its polarized photon number distribution, taking care of quantum interference at the postselection polarizer acting on the intra-cavity field. This is an approximation, because imperfect transmission through the cavity reshapes $P_n$. We found that the photon statistics $P_n$ can be calculated best by projection on the required Fock states using polarization-rotated Fock space ladder operators $b_{n/2} = a_{n/2}\cos \theta_{\text{out}} + a_{n/2}\sin \theta_{\text{out}}$ and tracing out the undesirable polarization component afterwards. With the numerically calculated steady-state density matrix operator $\rho$ of

![Figure 3 | Photon bunching and purification.](image)

**Figure 3** | Photon bunching and purification. Experimental (a) and theoretical (b) data of the second-order correlation function as a function of the QD frequency and output polarization ($\theta_{\text{out}}$), taken in the area marked with a white rectangle in Fig. 2. The vertical dashed lines indicate the special polarization angle and the black horizontal line indicate the QD resonance frequency.

![Figure 4 | Special polarization and cooperativity.](image)

**Figure 4** | Special polarization and cooperativity. Black: numerically determined special polarization angle, where photon bunching in transmission is maximized, as a function of the cooperativity $C$, which in turn is modified by varying only the QD lifetime $\gamma$. The green curve is given by the phenomenological expression $\sim 45^\circ \exp(-C)$: in the limit of high $C$, the QD alone can efficiently filter out single-photon states, leading to photon bunching. However, for low cooperativity $C$, it is advantageous to mix the quantum-dot scattered light with a ‘local oscillator’ provided by orthogonal polarization. The red data point corresponds to the sample presented here; the blue data corresponds to another device with lower cooperativity.
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dip (P2) is not exactly at cross-polarization, which suggests the
following intuitive explanation: apparently, in the photon number
basis, the different Fock states pick up a different phase
during transmission through the QD cavity system. In the
weak-coupling regime, but often also in the strong coupling
regime, the individual JC dressed states cannot be resolved
spectrally, because g ≪ k. However, the CQED system is still
photon-number sensitive, which implies lifetime-dependent JC
effects in the weak coupling regime: the decay rate of the CQED
system increases with the number of photons in the cavity20,38.
As consequence, higher photon-number states have a
modified interaction cross-section and experience a reduced
phase shift. The dip in P2 in Fig. 5 is already very close to
the cross-polarization angle θout = −45°, whereas the dips for
higher photon number states Pn>2 are indistinguishable from
θout = −45°.

In conclusion, we found that the nonlinear response of a lossy
cavity QD system can be strongly enhanced by postselection of
a particular polarization state. This leads to interference between
Fock states that experienced different modifications by the QD
nonlinearity and results in strong photon correlations of the
transmitted light. As the underlying effect, interference of the two
polarizations modes leads to high-fidelity cancellation of the

single-photon transmission for the special polarization
postselection condition. By correlating the results with a
theoretical model, we found indications of photon-number
sensitive JC physics in the weak coupling regime of CQED.

Data availability. All relevant data are available on request.

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Figure 5 | Photon number distributions. Calculated photon number
distribution after the polarizer, with (through curves) and without
(dashed curves) coupling to the QD in the cavity, the laser frequency is set
to one of the QD resonances. With QD, we clearly see the photon-number-
dependent shift of the transmission dip. Only the photon number
distribution of the detected polarization component is shown; therefore, the
total number of photons in case with QD can exceed the case without QD
due to polarization conversion by the dot. For clarity, pure dephasing has
been neglected here, making the special polarization angle different from
the other simulations and experimental results.

Figure 5 shows the four lowest photon number probabilities as
a function of the polarizer angle θout for the case with and
without QD. In the empty-cavity case we see, as expected,
lowest transmission under the cross-polarization condition
(θout = −45°). For the case with the QD, we observe a photon number-
dependent shift of the transmission dip. At the special
polarization angle θout, we see that the one-photon component
reaches a minimum, while the higher-photon number states do
not, which explains the enhanced photon bunching enabled by
the purification technique.

It is important to note that also the two-photon transmission
dip (P2) is not exactly at cross-polarization, which suggests the
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**Acknowledgements**

We thank G. Nienhuis for fruitful discussions. We acknowledge funding from FOM-NWO (08QIP6-2), from NWO/OCW as part of the Frontiers of Nanoscience program, and from the National Science Foundation (NSF) (0901886, 0960331).

**Author contributions**

J.A.F., J.N., E.C.L., A.G., J.E.B., D.B. and W.L. designed and fabricated the devices. M.P.B., H.S., M.P.v.E., D.B. and W.L. conceived and conducted the optical experiments. All authors contributed to the manuscript.

**Additional information**

Supplementary Information accompanies this paper at http://www.nature.com/naturecommunications

Competing financial interests: The authors declare no competing financial interests.

How to cite this article: Snijders, H. *et al.* Purification of a single-photon nonlinearity. *Nat. Commun.* **7**:12578 doi: 10.1038/ncomms12578 (2016).

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