Emission from a resonantly excited quantum emitter is a fascinating research topic within the field of quantum optics and is a useful source for different types of quantum light fields. The resonance spectrum consists of a single spectral line that develops into a triplet above saturation of the quantum emitter1–3. This process is demonstrated in a number of experiments exhibiting resonance fluorescence from the Mollow triplet sidebands5. The bright and narrow sideband emission has been shown to be a valuable light source for quantum light spectroscopy and quantum information applications, for example.

The generation of non-classical light fields, such as single, entangled and cascaded or even heralded photons, forms a vital part of many schemes related to quantum information and computation. Atom optics has demonstrated the cascaded emission of single photons using resonance fluorescence from single atoms6 and from cold atoms in a cavity7. Semiconductor quantum dots have also been studied8,9,10, and cascaded photon sources11, but predominantly in non-resonant excitation schemes. Resonant excitation of these quantum emitters is an important precondition, because it promises to minimize most of the dephasing caused by non-resonant processes12. Coherent control of quantum dot excitons has been demonstrated in a number of experiments exhibiting Rabi splitting13, Rabi oscillations14 and resonant absorption15,16. Observations of oscillations in the first-order correlation17, characteristic emission spectra in the frequency domain, and oscillations of the second-order photon correlation function have all been measured directly in the resonance fluorescence from quantum dots. In particular, single-photon emission from the resonance line (below emitter saturation) as well as photon indistinguishability as high as 90% have been demonstrated18. Furthermore, the a.c. Stark shift of an exciton has been used to bring the initially split energy component and the ‘three photon’ line as the high-energy sideband4. These spectral components are a result of spontaneous emission down a ladder of paired states of so-called ‘dressed states’, which are a representation of the combined quantum dot state and laser mode while considering the coupling between them22. Each rung of the ladder (with N quanta) has two components, that is, (|g, N + 1⟩+|e, N⟩) and (|g, N + 1⟩−|e, N⟩), where (|g, N + 1⟩) and (|e, N⟩) represent the eigenstates of the uncoupled emitter laser states. The components of the dressed states are separated by the generalized Rabi frequency (Ω = √(Ωg + Δ)), where Ωg is the ‘bare’ Rabi frequency and Δ is the laser quantum dot detuning. The amplitudes of the dressed eigenstates are e = √(Ω + Δ)/(2Ω) and s = √(Ω − Δ)/(2Ω). The detuning Δ-dependence of the even energies of the dressed states is plotted in Fig. 1a.

For large positive detunings Δ ≫ 0, the steady-state solution5 of the dressed states reveals that state |1⟩ has a mainly |g, N + 1⟩ character and the system is largely prepared in state |1⟩ as c2 ≫ s2. Thus a T photon is emitted first, which puts the system into state |2⟩ of the next lower manifold E(N), from where an F photon emission can eventually occur (Fig. 1b). This time-ordered cascaded emission of a T photon ‘heralding’ the F transition results in a photon-bunching signature of the Mollow sidebands in photon cross-correlation experiments, as demonstrated below. In contrast, an autocorrelation measurement on an individual sideband should exhibit antibunching. This is because, after emission of a T photon for example, emission of an F sideband photon should occur before another T photon emission is possible (and vice versa; Fig. 1b,c). In contrast, photons from the central Rayleigh line should be totally uncorrelated in time (that is, Poissonian distributed), as the emission of an R photon is not accompanied by population modulation of the state of the emitter5, so the population remains in the same state, |1⟩ or |2⟩.

Figure 1d shows a Mollow triplet series obtained at a constant excitation power with systematic variation of laser detuning Δ. The central peak shifts along with the laser, and the satellite sidebands always remain spectrally symmetric with respect to the centre. As can be clearly observed in Fig. 1e, the total Rabi splitting Ω increases with increasing detuning Δ. The linewidth of the Mollow sidebands in the resonant spectrum (Δ = 0) is found to be Δν = 730 ± 20 MHz at the corresponding Rabi

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frequency of $\Omega_\nu/2\pi = 5.4$ GHz. This is somewhat larger than the Fourier-limited linewidth for a Mollow sideband given by $\Delta \nu / 2\pi = (3/2)\nu_0/(1/2\pi) = 300$ MHz due to excitation-induced dephasing effects.$^{20}$

A two-step signal filtering process, initially by means of a Michelson interferometer$^{23}$ (Fig. 2a–c) and then with a spectrometer, ensures a high degree of spectral purity of the photons in the Mollow triplet channels. It should be noted that an average photon count of 70,000 s$^{-1}$ was obtained on each Mollow sideband after spectral filtering, which corresponds to a collection of 5.9 million photons per second from the sample by the objective of our confocal microscope. From the measured lifetime $T_1$ we can estimate the emitted photon rate $R_{\text{SB}}$ of each Mollow sideband to be $R_{\text{SB}} = (c's'/(c^2 + s^2))(1/T_1)$ (ref. 4), which at resonance is $\sim 156$ MHz. We anticipate that higher collection efficiencies might be achieved by using improved sample structures such as high-Q microcavities and photonic nanowires.$^{24}$

Figure 2d shows a photon correlation measurement taken on the central Mollow peak (see also Fig. 2c). The autocorrelation demonstrates long-timescale bunching instead of pure Poissonian statistics. Such an effect is commonly detected for a quantum dot recombination signal subject to ‘blinking’ of the excitonic state between two or more neighbouring competing states.$^{25}$ The timescale of these processes can vary from 10 ns to a few hundred nanoseconds depending on the pump power.$^{25}$ The phenomenon of blinking under pure-resonant excitation can be assigned to the presence of a competing quantum dot exciton spin configuration known as the dark excitonic state, which is non-radiative in nature.$^{26}$ Our $g^{(2)}(\tau)$ correlation data are fitted by a bidirectional exponential fit, which reveals the explicit power-dependent blinking timescale and is observed in all the photon correlation measurements shown here.

The results of photon autocorrelation measurements carried out on a spectrally separated Mollow sideband (T) are presented in Fig. 2e. The entire signal is superimposed by the abovementioned bunching effect due to ‘blinking’. The signal around zero delay exhibits clean antibunching with a normalized value of $g^{(2)}(\tau) = 0.18$ at $\tau = 0$ (considering the time resolution of the detection set-up). From the experimental signature of antibunching we derived a time constant of $\tau_{\text{bl}} = 0.8 \pm 0.1$ ns, which is somewhat smaller than the theoretically expected value$^{2}$ (see Supplementary Information). We interpret the presence of a finite background in the correlation measurement to be a consequence of the signal contribution from the spectrally close central peak of the Mollow triplet as a non-vanishing background in the photoluminescence spectrum (Fig. 2e, inset).

Figure 3a presents excerpts of a series of correlation measurements taken on both sidebands simultaneously, without spectral separation between them. Consequently, there is no time ordering between the photon emission of the sidebands, as either of them can initiate a ‘start’ or ‘stop’ signal in the photon counting process. The data of Fig. 3a represent three distinct cases: the laser close to the exact quantum dot s-shell ($\Delta \approx 0$; centre), and blue-detuned ($\Delta > 0$; top) and red-detuned ($\Delta < 0$; bottom) from resonance. As expected, a short-timescale symmetric bunching feature is clearly observed for both detuned cases ($\Delta \neq 0$). The resonant case ($\Delta \approx 0$) does not show such bunching, as the probability of F or T being the first photon in the cascade is equal, and no cascaded emission is therefore expected. Once again, all three measurements

**Figure 1** | Laser detuning dependent Mollow triplet spectra. a. Laser quantum dot detuning dependence of dressed-state eigen energies (solid lines), as well as the quantum dot + laser uncoupled eigenstates (dashed lines)$^{20}$ b, c. Sideband photon emission sequence down the dressed-states ladder for strong blue-detuned ($\Delta > 0$), b and red-detuned ($\Delta \approx 0$), c lasers. d. Laser detuning dependent resonance fluorescence spectra recorded as high-resolution photoluminescence. All spectra are taken at the same excitation power, $P = 500 \mu$W. e. Positions of the spectral components of the fluorescence signal derived from the detuning series in d. Solid lines are fits according to $\nu = \Delta \pm \sqrt{(\Omega_\nu^2 + \Delta^2)}$ and $\nu = \Delta$.  

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Figure 2 | Photon correlations on Mollow spectral components. a, Michelson interferometer set-up used to spatially separate the Mollow sidebands from the central line (see Supplementary Information). b, Mollow spectrum obtained after filtering out the central peak, where only the sidebands remain observable in high-resolution photoluminescence. c, High-resolution spectrum of the central Rayleigh line filtered from the Mollow sidebands in b, d, Photon autocorrelation of the central Mollow triplet peak. The ‘bare’ Rabi frequency is 11.27 GHz while the laser is at exact resonance with the quantum dot exciton ($\Delta = 0$). The solid line is a bidirectional exponential fit to the data to extract the effect of ‘blinking’. Inset: resonance fluorescence emission spectrum of the full Mollow triplet obtained with the shaded region as the selected spectral region for photon correlation measurement. e, Autocorrelation on the T line of the Mollow spectrum. The experimental conditions were $\Omega_0/2\pi = 5.4$ GHz and $\Delta = 0$. The solid red line is a theoretical fit of the signal, convoluted with the instrumental detector response (480 ps), and the blue dashed line is the same, but deconvoluted. Inset: emission spectrum on the charge-coupled device after filtering out the central line. The shaded region shows the spectrally selected signal for the autocorrelation measurement.

Figure 3 | Laser detuning dependent correlation of the combined Mollow sidebands signal. a, Normalized photon correlation measurement on both Mollow triplet sidebands, without any time-ordering. The measurements are performed on resonance with the quantum dot s-shell (centre), blue-detuned (top) and red-detuned (bottom). Inset: corresponding emission spectrum with the highlighted area as the spectrally selected signal. b, Bunching parameter, derived from correlation measurements in a, as a function of laser–quantum dot detuning $\Delta$. Data points are plotted for two different excitation powers: red, 170 $\mu$W with $\Omega_0/2\pi = 3.4$ GHz; black, 500 $\mu$W with $\Omega_0/2\pi = 5.4$ GHz. Dashed lines are guides to the eye.
Figure 4 | Cross-correlation between spectrally selected Mollow sidebands. a. Normalized photon correlation between the T photon (‘start’) and F photon (‘stop’) lines, with the central Rayleigh line interferometrically suppressed. The bare Rabi frequency is $\Omega_0/2\pi = 11.5 \text{ GHz}$, and the laser is blue-detuned by $\Delta/2\pi = +9.3 \text{ GHz}$ from exact resonance. The solid line is a theoretical fit to the data with the exponential for negative (positive) delay indicating the rising (falling) edge of the correlation signal. For this case, the steady-state population of $|1\rangle$ is 95%. B. As in a, but for a case where $\Delta/2\pi = -7.6 \text{ GHz}$ and $\Delta/2\pi = -7.6 \text{ GHz}$. The rise and fall edges are time-reversed in this case, and the steady-state population of state $|2\rangle$ is 95%.

demonstrate the long-timescale ($\Gamma_\text{bl}^{-1} = 42 \text{ ns}$ here) bunching effect corresponding to ‘blinking’ of the state. Figure 3b displays the measured evolution of the bunching strength as a function of detuning $\Delta$ for two different fixed bare Rabi energies, $\Omega_0$. A symmetric increase in the bunching signature on either side of the detuning can be observed, as is made obvious by the dashed lines in the figure (provided as a guide to the eye). Even though the ‘cascaded’ nature of photon emission is observed, time ordering is still not observed due to the simultaneous detection of both sidebands.

Figure 4a,b presents cross-correlation measurements between the spectrally separated opposite Mollow triplet sidebands, in which the T photon always acts as a ‘start’ and the F photon as a ‘stop’ trigger signal. In each case, we observe a distinct bunching signature on a very short timescale of delays $|t| \approx 0$, on top of the aforementioned long-timescale blinking. The normalized cross-correlation bunching signal at $\tau \approx 0$ has been fitted with two single exponentials with characteristic time constants of $\tau_{\text{rise}}$ and $\tau_{\text{fall}}$, respectively. Figure 4a depicts the case of strongly blue-detuned excitation ($|\Delta/\Omega| \approx 83\%$), which for delays $\tau \approx 0$ reveals an abrupt rise with $\tau_{\text{rise}} = 0.48 \pm 0.10 \text{ ns}$, simply reflecting the detector response. In contrast, positive delays $\tau \geq 0$ show a significantly longer decay constant of $\tau_{\text{fall}} = 0.94 \pm 0.10 \text{ ns}$. This asymmetry in the bunched correlation signal clearly reflects time-ordering in the photon emission process of the sidebands, which means that the T and F photon emission forms a temporal radiative cascade in this case (Fig. 1b).

In contrast, Fig. 4b illustrates the case of strongly red-detuned excitation ($|\Delta/\Omega| \approx 79\%$). In this case, emission time ordering is reversed; that is, the T photon follows the F photon (Fig. 1c). Although the bunching again reveals $\tau_{\text{rise}} = 0.48 \pm 0.10 \text{ ns}$ (for $\tau > 0$), the falling edge of the correlation peak can be perfectly fitted by $\tau_{\text{fall}} = 0.89 \pm 0.10 \text{ ns}$. We emphasize that, for both blue and red excitation detuning, the derived values of $\tau_{\text{fall}}$ are close to the theoretically predicted modified emission rates $\Gamma = \Gamma_{\text{em}}(e^{-sT} + s)$, yielding lifetimes of $1.10 \pm 0.13 \text{ ns} (\Delta > 0)$ and $1.15 \pm 0.10 \text{ ns} (\Delta < 0)$, respectively (see Supplementary Information).

In conclusion, we have demonstrated that an individual Mollow sideband channel of the resonance fluorescence from a single quantum dot can act as an efficient single-photon source. By spectrally separating both Mollow sidebands, we have shown that this source can act as a solid state-based cascaded photon emitter with time-ordered photon pair emission process. Possible applications of such high-brightness photon sources include the realization of semiconductor–atom interfaces for the development of photon-based memories. Cascade photon sources may find applications in quantum communication protocols. These high-yield and narrowband sources might also be invaluable for spectroscopy of quantum emitters, where non-classical light is used for the excitation process. Using the Purcell effect by embedding the quantum dots in optical microcavities can result in spectrally ultra-sharp emission lines. Tuning the cavity into resonance with one of the Mollow triplet sidebands can result in population inversion of the system and, ultimately, single quantum emitter lasing. Quantum dots in a high-Q microcavity sample can also achieve an increased photon-cascade correlation or even heralded photon emission based on the scheme presented here, as hinted in ref. 21.

Methods
The sample consisted of a single layer of self-assembled In(Ga)As quantum dots grown by metal–organic vapour-phase epitaxy. The quantum dot layer was sandwiched between GaAs/AlAs distributed Bragg reflector (DBR) layers at the centre of a GaAs $\lambda$-cavity. The sample was kept in a helium-flow cryostat that was capable of stabilizing the temperature to $5 \pm 0.5 \text{ K}$. The measurements were performed on a special micro-photoluminescence setup with orthogonal geometry between the laser excitation in the growth plane, in combination with detection of luminescence perpendicular to the sample surface. The DBR structure of the sample acted as a waveguide for the excitation laser. In combination with the $90^\circ$ geometry, a pinhole assembly was used to minimize the contribution of scattered laser light. A polarizer setup containing a high-extinction Glan Thompson polarizer further suppressed the collection of laser stray light by means of polarization selection. An individual quantum dot was addressed by a narrowband ($\sim 500 \text{ kHz}$) continuous-wave Ti:sapphire laser by tuning it into the $s$-shell of a quantum dot. While scanning the narrowband laser over the quantum dot $s$-shell resonance, enhancement of the signal gave an indication of the onset of resonance fluorescence. The spectrometer/charge-coupled device combination in the set-up had a resolution of $8.4 \text{ GHz}$, which is not sufficient to resolve the components of the Mollow emission spectrum, particularly at lower excitation powers. To obtain a detailed spectrum of the full Mollow triplet, with well-resolved sidebands, we used a scanning Fabry–Perot interferometer providing a resolution of $\sim 250 \text{ MHz}$. To perform correlation measurements on different spectral components of the Mollow triplet, it is important to have an efficient filtering mechanism. For this purpose, a Michelson interferometer was used for pre-filtering. The delay of the interferometer was set in such a way that the photons from the Mollow sidebands interfered constructively at one output, and the central Mollow peak left the other output. After spatial separation of the sidebands and the central peak, further filtering was performed using two spectrometers with spectral resolutions of $8.4$ and $9.6 \text{ GHz}$, respectively. The same spectral component of the Mollow emission spectrum was selected to perform photon autocorrelation, and for cross-correlations different components were selected by each spectrometer.

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**Author contributions**

R.R. and M.J. designed the sample structure. A.U., S.W., S.M.U. and P.M. conceived the experiments. A.U., S.W. and S.M.U. performed the experiments and analysed the data. R.R. and M.J. designed the sample structure. A.U. acknowledges funding from the Carl-Zeiss-Stiftung.

**Additional information**

The authors declare no competing financial interests. Supplementary information accompanies this paper at www.nature.com/naturephotronics. Reprints and permission information is available online at http://www.nature.com/reprints. Correspondence and requests for materials should be addressed to A.U.