

# Highly indistinguishable on-demand resonance fluorescence photons from a deterministic quantum dot micropillar device with 74 % extraction efficiency

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**Abstract:** The implementation and engineering of bright and coherent solid state quantum light sources is key for the realization of both on chip and remote quantum networks. Despite tremendous efforts for more than 15 years, the combination of these two key prerequisites in a single, potentially scalable device is a major challenge. Here, we report on the observation of bright single photon emission generated via pulsed, resonance fluorescence conditions from a single quantum dot (QD) deterministically centered in a micropillar cavity device via cryogenic optical lithography. The brightness of the QD fluorescence is greatly enhanced on resonance with the fundamental mode of the pillar, leading to an overall device efficiency of  $\eta = (74 \pm 4) \%$  for a single photon emission as pure as  $g^{(2)}(0) = 0.0092 \pm 0.0004$ . The combination of large Purcell enhancement and resonant pumping conditions allows us to observe a two-photon wave packet overlap up to  $v = (88 \pm 3) \%$ .

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## 1. Introduction

Building compact and bright solid state sources of on-demand and Fourier-limited single photons is a major challenge in engineering quantum emitters and solid state microcavities [1]. Such sources are of crucial importance for establishing optical quantum networks on- and off chip [3, 4], to enable quantum teleportation [5, 6] and to implement building blocks for quantum repeater networks [7]. Among the most promising candidate for solid state single-photon emitters are epitaxially grown semiconductor quantum dots (QDs). Due to their near-unity quantum efficiency and spontaneous emission lifetimes in the lower ns range they allow for operation frequencies in the GHz range. As InAs or InP QDs are embedded in a high refractive index medium and thus suffer from total internal reflection, improving the extraction efficiency of the single photon flux out of the crystal is crucial and can be established via various designs [8]. Thus far, microcavities [9, 10], lensing [11, 12, 13] and waveguiding approaches [14, 15, 16, 17] have been most successfully applied to this long standing problem, and single photon sources with extraction efficiencies in excess of 70 % [14] have been reported. The indistinguishability of the emitted photons, however, which is an equally important characteristics of a quantum light source for the above mentioned applications, is a parameter which is harder to maximize. Indistinguishable photons share all spectral characteristics, including polarization and color. Furthermore, the characteristic destructive quantum interference between photons leaving separate output ports on a beamsplitter can only be established if the single photons impinge the splitter simultaneously, and if their wave packet overlap equals unity. Equal timing requires an excitation technique which minimizes time jittering of the emission event, while maximizing the wave packet overlap requires close-to Fourier limited photons. In principal, the latter can be achieved more easy using the Purcell effect to enhance the emission rate of the emitter [18]. While single photon streams can be generated from a single quantum dot under non-resonant or quasi-resonant excitation conditions, it has been recognized that resonance fluorescence excitation is the most suitable configuration to simultaneously minimize time jittering and maximize coherence [19, 20].

In recent works, we discussed the successful implementation of resonant excitation to trigger streams of single [21] and highly indistinguishable [22] photons. These devices were, however, based on a random alignment of QD and cavity. Thus the effort to find a device which combines the criteria of brightness and coherence is painstaking and not very practicable for scalable applications. To circumvent this limitation, in our current work we choose a deterministic technique to put a coherent, pre-selected QD in the maximum of the optical field in a micropillar cavity. The resulting device, operated deeply in the Purcell regime, yields an overall device efficiency of  $\eta = (74 \pm 4) \%$  of resonance fluorescence single photons and an extraction efficiency of  $\eta_{Lin} = (37 \pm 2) \%$  of linearly polarized single photons. The coherence of the resonance fluorescence photons is measured via Hong-Ou-Mandel type interference experiments resulting in a maximal two-photon wave packet overlap of up to  $v = (88 \pm 3) \%$ .

## 2. Experimental setup and Hong-Ou-Mandel interference

The experimental setup for the resonant excitation of such a QD cavity system consist of a linearly polarized Ti:Sapphire laser (repetition rate 82 MHz, pulse length  $\tau \approx 1.3$  ps) which is

coupled into the beam path via a 92/8 Pellicle beamsplitter. The micropillar sample is mounted on the coldfinger of a  $\text{He}$  flow cryostat and the emitted light from the sample is collected with a microscope objective ( $\text{NA} = 0.42$ ) and coupled into a single mode fiber. A second linear polarizer in front of the fiber coupler is orientated perpendicular to the laser polarization and selects the detected polarization axis of the QD signal. After spectrally filtering the emission with a  $1500 \frac{\text{lines}}{\text{mm}}$  grating, we either detect the signal on a charged-coupled device (CCD) or we couple it into a fiber. The single photon statistics are measured via a fiber-coupled Hanbury Brown and Twiss setup. For the two-photon interference experiments, we divide each laser pulse into two equal pulses with 2 ns delay. The interference experiments are carried out with an unbalanced free beam Mach-Zehnder-interferometer, where the two arms exactly compensate the separation between two consecutive emitted photons. We measure the second order auto-correlation of this interferometer via two single photon sensitive Silicon based avalanche photo diodes at the exit ports of the second 50/50 beamsplitter.

### 3. Sample growth and insitu lithography

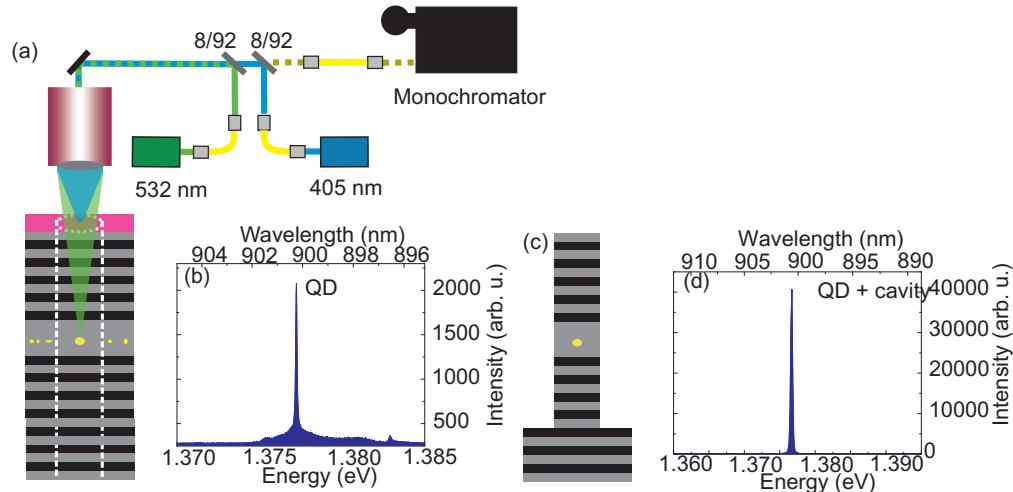


Fig. 1. Schematic drawing of the deterministic device fabrication: (a) We excite the planar structure with a green laser to pump the QDs without exposing the photo resist. (b) We select a single, bright QD line and optimize the signal. (c) After exposing the photoresist with a violet laser, we etch the micropillar into the planar structure. (d) The result is a bright, background free, Purcell-enhanced single QD line.

Figure 1 depicts a sketch of the fabrication scheme which we applied to fabricate our aligned QD-micropillar device, in a similar approach as discussed by Dousse et al.[23]. The process starts from a planar cavity structure grown via means of molecular beam epitaxy (MBE). It consists of 25.5 (15)  $\lambda/4$ -thick AlAs/GaAs mirror pairs which form the lower (upper) distributed Bragg reflector (DBR). The DBR mirrors sandwich a  $\lambda$ -thick GaAs cavity with a single layer of low density In(Ga)As QDs. To shift the emission wavelength of the QDs to the 900 nm range, we performed an in-situ partial capping and annealing step while MBE growth. A silicon  $\delta$ -doping was introduced 10 nm below the QD layer to stochastically charge the single QDs with an excess electron. The sample is spin-coated with a conventional photo resist for optical lithography (sensitive in the ultra violet to blue spectral range) and mounted on the cold finger of a  $\text{He}$  flow cryostat. We excite the sample with a green, SM-fiber coupled, continuous wave laser ( $\lambda = 532 \text{ nm}$ ) to pump the QDs without exposing the resist. After selecting a bright,

resolution limited single QD line (see Fig. 1(b)), we maximize the signal to ensure that the QD is located in the center of the laser spot. An additional SM-fiber coupled, violet laser ( $\lambda = 405$  nm), which beam path is identically to the one of the green laser is used to expose the photo resist. Via varying the exposure time, we can control the diameter of the exposed area with sub-micron precision and thus can match the optical resonance to the emission frequency of the QD. Subsequent to this cryogenic optical lithography step, we deposit a BaF/Cr hard mask and after lift-off, the mask pattern is transferred into the sample via electron-cyclotron-resonance reactive-ion-etching. Next, the sample is planarized by a transparent polymer to stabilize the pillars and protect them from sidewall oxidation, and the hard mask is fully removed in an ultra sonic bath. With this technique, we observe a yield of approximately 20 % to match the emission wavelength between QD and fundamental mode below 15- 20 K. The deviation from an ideal fabrication process is mainly due to slight shifts of the QD emission over several cooling and heating cycles and within the etching process altering the strain environment of the QD.

#### 4. Experimental results

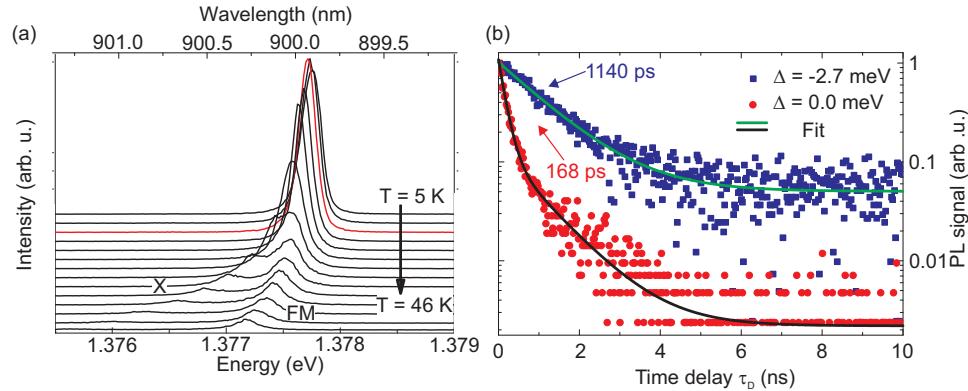


Fig. 2. (a) Temperature dependent spectra of an insitu defined micropillar with a diameter of  $d \approx 3 \mu\text{m}$  under above bandgap excitation. A strong enhancement on spectral resonance due to the Purcell effect is observed. (b) Time resolved measurements on and off resonance reveal a Purcell factor of  $F_P = 5.8 \pm 0.2$ .

In the following, we discuss the emission characteristics of a micropillar with a diameter of  $d = 3 \mu\text{m}$  ( $Q=5930$ ,  $\gamma_C = 232 \mu\text{eV}$ ). Figure 2(a) shows a series of spectra acquired under non-resonant excitation and for varying temperature. We observe a clear signature of weak light matter coupling with a strong enhancement of the QD emission for spectral resonance between QD and fundamental optical mode. To determine the Purcell enhancement of the system, we carried out time-resolved  $\mu$ -photoluminescence measurements ( $t_{\text{Res}} \approx 40$  ps). The time-depended emission of the saturated QD is plotted in Fig. 2(b). Via fitting the decay curve for spectral resonance between QD and cavity with a biexponential decay we can extract the lifetime of the QD exciton as  $T_{\text{on}} = (168 \pm 5)$  ps. While the origin of the biexponential decay is not fully clarified, we suggest that an off resonant contribution of the same emitter (via another charge state) is a reasonable assumption [24]. The lifetime for a spectral detuning of  $\Delta = E_X - E_C = -2.7 \text{ meV} \approx 12 * \gamma_C$  is measured to be  $T_{\text{off}} = (1140 \pm 19)$  ps. This leads to a Purcell factor of  $F_P = \frac{T_{\text{off}}}{T_{\text{on}}} - 1 = 5.8 \pm 0.2$ , under the common assumption that suppression of spontaneous emission off resonance is insignificant in our case.

Full inversion of the two level system can be established most efficiently under strictly resonant pumping: Figure 3(a) shows the integrated QD emission taken from the CCD of our setup

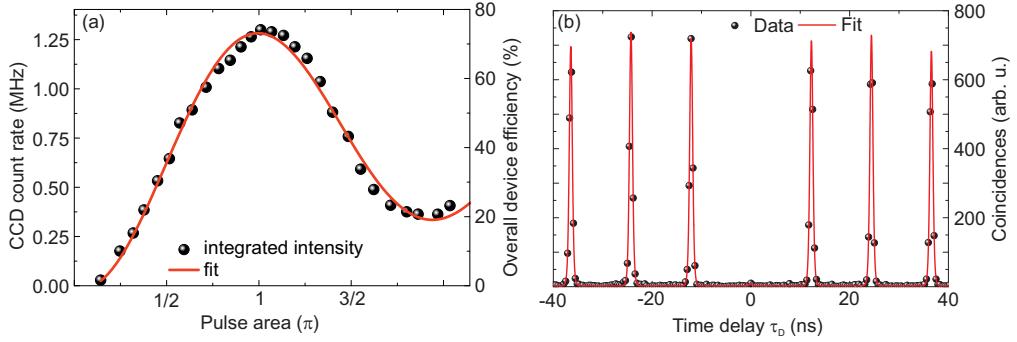


Fig. 3. (a) Measured source efficiency and count rate on the monochromator CCD versus the pulse area of the driving laser field for spectral resonance between QD and cavity mode. We extract an overall device efficiency of  $\eta = (74 \pm 4) \%$ . (b) 2<sup>nd</sup> order auto-correlation histogram for pulsed resonant excitation with a  $\pi$ -pulse. We extract a  $g^{(2)}$ -value as low as  $g^{(2)}(0) = 0.0092 \pm 0.0004$ .

to avoid laser induced errors from a lowered signal to laser ratio for high pumping strength. We observe an oscillating behaviour which is a signature for the pulsed resonant driving of the system [25]. For  $\pi$ -pulse excitation, we observe count rates on the CCD up to 1.3 million counts per second. In order to extract the overall device efficiency of our QD micropillar device from this measurement, we carefully calibrated our setup revealing a setup efficiency for the detected linear polarization of  $\eta_{Setup} = (2.1 \pm 0.1) \%$ . Therefore, our device yields an overall efficiency of  $\eta = (74 \pm 4) \%$ . This value is comparable to the reported state of the art values for single QD-based light sources. However, we note that it is significantly larger than any other value reported thus far for a deterministically inverted quantum dot via resonant pumping. For possible applications, the figure of merit of such a device is the number of linear polarized photons emitted by the SPS. For our device, this number is half of the overall device efficiency which leads to an efficiency of linear polarized photons of  $\eta_{Lin} = (37 \pm 2) \%$ . In order to compare our results to the maximal achievable extraction efficiency of a micropillar with the same specifications, we simulated our structure using an eigenmode expansion technique [26]. We assumed an oxidation of 75 nm of the AlAs layer, as well as a 130 nm thick SiO<sub>2</sub>-layer on the sidewalls of the micropillar induced by the reactive ion etching process on a silicon sample holder [27]. This leads to a maximum extraction efficiency of 75 % for a 3  $\mu\text{m}$  diameter pillar, which compares very well with our measured efficiency. Without the AlAs oxidation and the SiO<sub>2</sub>-surface, the calculated efficiency is slightly decreased to approx. 72 % resulting from a slight increase of the cavity mode volume. The high brightness, and comparably large setup efficiency allows us to record quasi background free single photon correlation charts via a fiber coupled Hanbury Brown and Twiss setup with very good signal to noise ratios on the minute scale. Figure 3(b) shows the recorded coincidence histogram for  $\pi$ -pulse excitation. The vanishing peak around  $\tau \approx 0$  ns is a clear signature of the non classical light emission from the QD. We fit each pulse with a two sided exponential decay convolved with a Gaussian distribution, where the width is the time resolution ( $t_{Res} \approx 520$  ps) of our setup. This allows us to extract a value of  $g^{(2)}(0)$  via dividing the area of the central peak by the average area of the surrounding peaks, which amounts to  $g^{(2)}(0) = 0.0092 \pm 0.0004$ . These results unambiguously proof pure and bright single photon emission from a deterministically prepared exciton state.

In order to test the coherence of consecutive emitted single photons from our device, we split the excitation pulses in a train of two pulses with 2 ns separation. The emitted photons

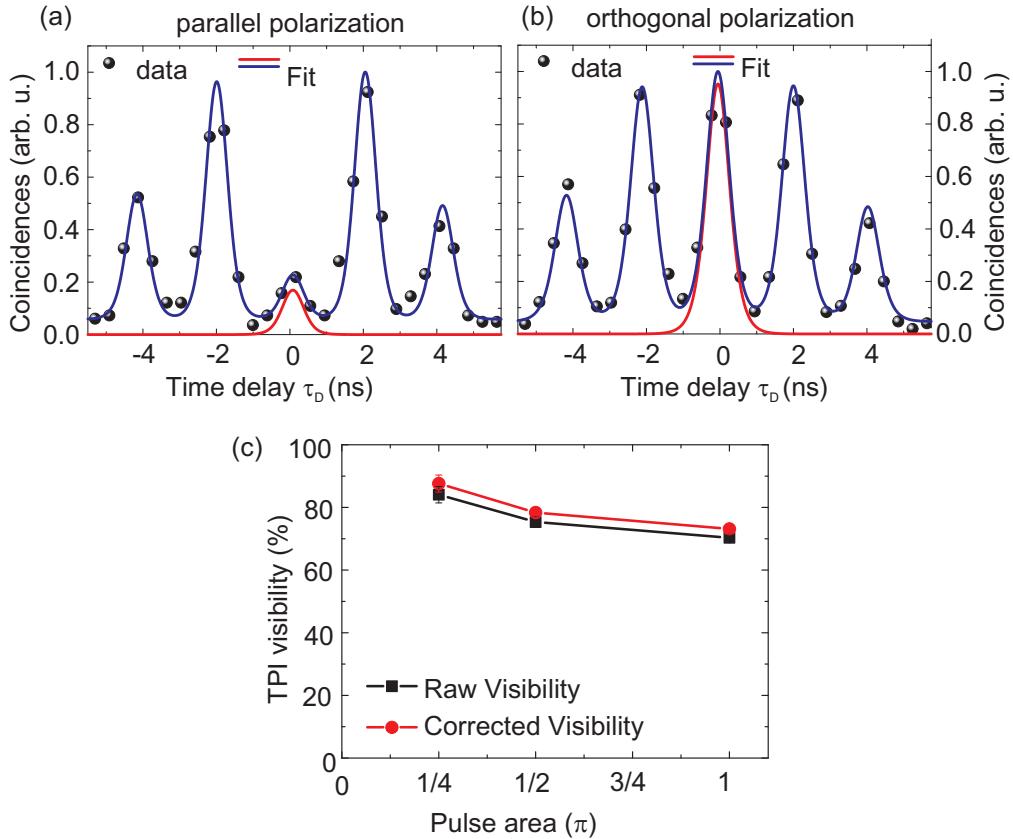


Fig. 4. Measured two-photon interference histograms for (a) parallel polarization of both photons and (b) orthogonal polarization. We fit each histogram with a sum of five two sided exponential functions each convolved with a Gaussian distribution and extract a visibility of  $v = (88 \pm 3) \%$ . (c) For higher pumping strength we observe a modest decrease of the two-photon interference visibility.

are then coupled into an unbalanced Mach-Zehnder-interferometer. One arm of the interferometer is precisely adjusted to compensate the delay between the two photons. If the early photon takes the long arm of the interferometer and the late photon the short path, both meet each other at the second 50/50 beam splitter where they can interfere if they are indistinguishable. Via an additional  $\lambda/2$ -wave plate, we can rotate the polarization in one arm of the interferometer by  $90^\circ$  to make the two photons distinguishable. Figure 4(a) and (b) show the measured coincidence histograms for parallel and orthogonal polarization of the photons for driving the system with a  $\pi/4$ -pulse. The suppression of the central peak in Fig. 4(a) is a clear proof, that our device is capable as a source for single and highly coherent photons. In order to extract the degree of indistinguishability, we fit each histogram by a sum of five two sided exponential functions, each convoluted with a Gaussian distribution. For the fitting procedure, we keep the time constant of the exponential decay fixed to the measured lifetime of the QD transition (see also Fig. 2(b)). Via the expression  $v = 1 - \frac{A_{\text{parallel}}}{A_{\text{orthogonal}}}$  we can extract the two-photon interference visibility  $v$  from our fit results which reveals a raw visibility of  $v_{\text{raw}} = (84 \pm 3) \%$ . When taking into account the imperfections of the 50/50 beamsplitter ( $R/T \approx 1.1$ ), the contrast of the Mach-Zehnder-interferometer ( $(1 - \varepsilon) = 0.98$ ) and the slight deviation from a non zero  $g^{(2)}(0)$ -value

( $g^{(2)} = 0.0092 \pm 0.0004$ ), we can use the expressions given by Santori et al. [28] to correct the two-photon interference visibility which results in a corrected value of  $v_{corr.} = (88 \pm 3) \%$ . This number compares favourably to previously reported values from deterministically fabricated QD based quantum light sources [12, 29]. When we increase the pumping strength, we observe a slight decrease of the two-photon wave packet overlap to  $v = (73 \pm 1) \%$  for  $\pi$ -pulse excitation (see Fig. 4(c)), most likely as a consequence of power induced dephasing of the system via coupling of the QD to longitudinal acoustic phonons.

## 5. Conclusions

In conclusion, we have discussed the implementation of a QD based single photon source based on a micropillar aligned to a pre-selected QD via cryogenic optical lithography. Ideal lateral alignment leads to a close to ideal coupling of the QD to the fundamental mode of the micropillar, characterized by a large Purcell factor of  $F_P = 5.8 \pm 0.2$ . As a result, we observe bright emission of single photons with an overall efficiency of  $\eta = (74 \pm 4) \%$  ( $(37 \pm 2) \%$  for linearly polarized single photons) and single photons statistics close to perfection with  $g^{(2)} = 0.0092 \pm 0.0004$  under pulsed resonance fluorescence conditions. In a Hong-Ou-Mandel interference experiment, we tested the indistinguishability of the emitted photons revealing a two-photon wave packet overlap as high as  $v = (88 \pm 3) \%$  for exciting the QD with a  $\pi/4$ -pulse. We believe that our work represents a significant step towards advanced solid state quantum optics experience experiments based on single, indistinguishable photons. Furthermore, the possibility to combine very efficient mode coupling and photon interference in aligned, microstructured devices is very encouraging for the implementation of integrated quantum dot based quantum circuits. As a next step one could also implement a reduction of the coupling to leaky modes to achieve an overall device efficiency close to 100 %[30]. The efficiency of linear polarized photons could be increased via fabricating elliptically shaped pillars to enhance the emission rate of one polarisation[31].

*Note:* During the submission of this work, we became aware of a similar work [32].

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