High performance single photon sources from photolithographically defined pillar microcavities

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Abstract: We demonstrate that single photons can be generated from single InAs/GaAs quantum dots in photolithographically defined pillar microcavities. Pillars with a 1.9 μm diameter cavity show a four fold enhancement in the radiative decay rate due to the Purcell effect and a photon collection efficiency into a lens of up to 10%. Measurements of the second order correlation function reveal a greater than fifty fold reduction in the multi-photon emission rate compared to a laser of the same intensity.

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OCIS codes: (270.5290) Photon statistics; (300.6250) Spectroscopy, condensed matter.

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1. Introduction

There has recently been great interest in a semiconductor source of single photons based on the luminescence of individual quantum dots [1, 2, 3]. Such a source allows for the integration of n and p-doped contacts which could facilitate electrical injection of carriers into a robust, compact device [4]. Of central importance in applications of these sources is the efficiency with which photons are collected from the device. For dots embedded in a high refractive index semiconductor this number is small. However, it is well known that pillar microcavity structures alter the distribution of photonic modes into which the quantum dot emits, greatly increasing the collection efficiency by [i] channeling emission into a single optical mode via the Purcell effect and [ii] collimating emission from this mode into a traveling wave that can be efficiently collected by a lens. In addition, the Purcell effect acts to reduce the radiative lifetime, reducing the jitter in the emission time, which can lead to the creation of time-bandwidth limited photons [2].

Previous work on pillar microcavities focused on sub-micron diameter pillars that were defined using sapphire dust as an etch mask [2] or electron beam lithography [5, 6]. However, a study by Barnes *et al* [7] on one set of pillars indicates that the highest collection efficiencies occur for diameters around 2 µm, although the precise diameter depends on how the quality factor (Q) varies with pillar diameter. Pillars on this scale have the added advantage that they can be defined using standard optical lithography, which allows mass production of devices with reproducible control of their shape, dimension and position. Here we present results from an array of 1.9 µm diameter pillar microcavities fabricated using standard photolithography techniques. We demonstrate that these pillars have figures of merit comparable to, and in some cases exceeding, those of smaller pillars.

2. Growth, processing and characterization of samples

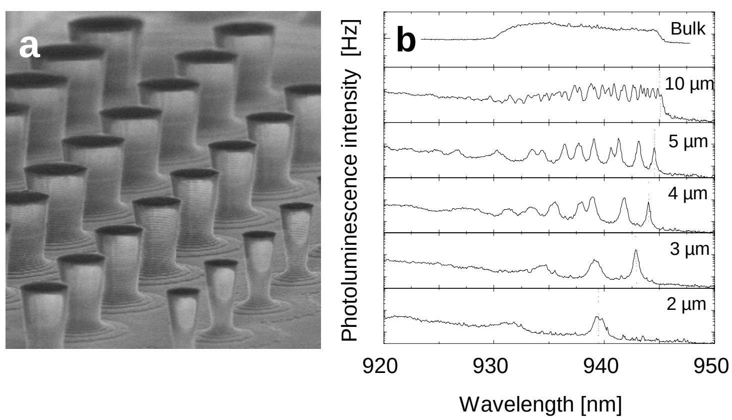

Fig. 1 (a). SEM of pillar microcavities with diameters (left to right) of 5, 4, 3 and 2 µm. (b) micro-PL spectra, recorded at high excitation densities (0.3 KW cm^{-2}) and averaged over several "empty" pillars. A dotted line indicates the wavelength of the HE_{11} mode.

The sample was grown by Molecular Beam Epitaxy and contains a single layer of low density self–assembled InAs/GaAs quantum dots at the centre of a microcavity of GaAs (λ = 945 nm). The GaAs/AlAs distributed Bragg reflector above (below) the cavity had 17 (20) mirror repeats. Standard photolithography and a $SiCl_4$ reactive ion etch was then used to pattern the sample into arrays of circular pillars of various diameters and height 4.6 µm. An SEM image of the sample (Fig. 1(a)) shows the side-walls of the pillars are undercut by 5.5^o.

All measurements were carried out in a continuous flow Helium cryostat at 5 K. Micro-photoluminescence (µ-PL) was excited with a tuneable picosecond-pulsed laser focused, at

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normal incidence, to a 2 μm spot on the sample surface by a microscope objective (NA = 0.5). Luminescence was collected with the same objective, dispersed using a grating spectrometer and passed to either a nitrogen cooled CCD, or a pair of avalanche photodiodes for recording time resolved or autocorrelation traces in a Hanbury-Brown and Twiss experiment. We also performed single photon interference experiments using a Michelson interferometer in front of the spectrometer in order to determine the average 1/e coherence length of photons emitted by the device [8]. In this experiment the variable delay arm of the interferometer is stepped in large (10-50 μm) steps to determine the shape of the wavepacket envelope.

3. Photoluminescence spectra and photon collection efficiency

With a high excitation density, pumping above the GaAs bandgap, it was possible to observe the cavity modes of the pillars. This is due to some spectrally continuous emission, which may be due to the InAs wetting layer or sub-bandgap emission from the surrounding GaAs layers [9], being filtered through the cavity modes. Fig. 1(b) shows a series of emission spectra for pillars of different sizes, averaged over several nominally identical pillars each of which showed no strong excitonic emission lines. These spectra were remarkably reproducible between pillars, showing an increasing broadening and blue-shift of the HE_{11} mode (dashed line, Fig. 1(b)) as the pillar diameter was reduced.

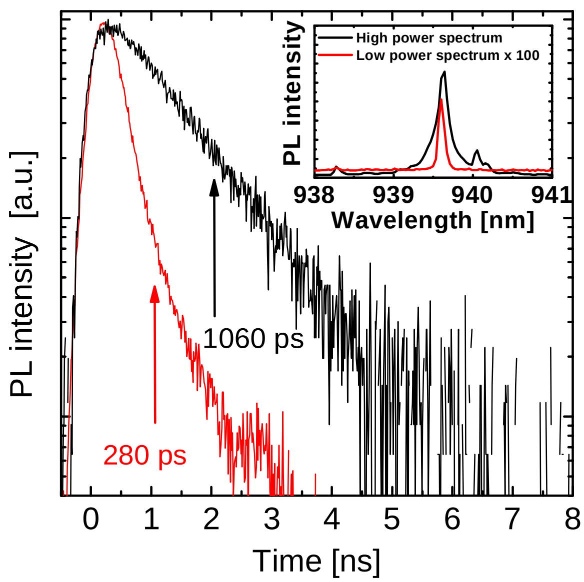

Fig. 2. Time-resolved decays of an exciton (*X*) line on resonance (red line) and away from resonance (black line) with the cavity mode at 5 K with and excitation density of 0.3 W cm^{-2}. Insert: micro-PL spectra from the same pillar with and *X* state resonant with the cavity mode under low (red, 0.3 W cm^{-2}) and high excitation levels (black, 0.9 KW cm^{-2}).

A more detailed study was made on one array of circular pillars with a diameter of 1.9 μm at the cavity center which had quality factors, $Q=\lambda/\delta\lambda$, for the HE_{11} mode of 2100. No polarisation splitting of the optical modes was observed. At low excitation densities (0.3 W cm^{-2}) it was possible to identify pillars where there was a single strong emission line near resonance with the HE_{11} mode. These lines were identified as excitons (X) due to their linear power dependence. At higher excitation powers biexcitons (X_2) with near-quadratic power dependence were visible. At still higher excitation powers both X and X_2 lines saturated and

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the emission tended towards a spectrum that was common to all nominally identical pillars in the array.

One pillar was identified with an X line on resonance with the cavity mode at 939.65 nm. The insert to Fig. 2 shows, in black, a spectrum from this pillar at a high excitation density, well above the point where the X state is saturated. Also shown (in red) is a spectrum recorded at 3.5 orders of magnitude lower excitation level where the dominant line is from recombination of the X state. Time-resolved measurements, with low excitation density, on pillars with the exciton on resonance with the HE_{11} cavity mode revealed decay times as low as 280 ps (fitted to the first part of the decay curve, Fig. 2). A longer decay time is also apparent in this plot which is due to the background, spectrally continuous emission, from other states in the cavity. Deconvolving this decay trace with the system response (recorded using a train of modelocked laser pulses) suggests the exciton radiative decay time was nearer 240 ps. In comparison, exciton lines that were spectrally well separated from the cavity mode had radiative lifetimes of 0.95-1.10 ns. Similar lifetimes were observed for the exciton states of QDs that were not in a microcavity. This corresponds to measured reduction in the radiative lifetime of ~ 4. Using a mode volume, V, calculated using an eigenmode solver package [10] we estimate a maximum Purcell factor, $F_p=3Q\lambda^3/4\pi^2Vn^3$, of 10. The measured value may be lower than the predicted maximum Purcell factor due to a number of factors, including possible misalignment of the spatial position and polarisation directions of the excitonic states and cavity modes and/or the presence of "leaky" optical modes [6, 11, 12, 13, 14].

In order to determine the collection efficiency for this device we have calibrated the transmission coefficients of all optics in our system and the detection efficiency of the CCD. Measurement of the saturation intensity of the exciton line of a single InAs QD under a 2 µm diameter metallic aperture (not in cavity) gave the efficiency which photons were collected into our microscope objective to be ~ 0.5 %. This agrees well with a simple calculation that assumes the QD emits isotropically. In a direct comparison, the pillar microcavity sample gave a saturated X intensity ~ 20 times higher corresponding to a collection efficiency into our microscope objective of 9.6 ± 1.9 %.

4. Correlation and interference experiments

We have also carried out single photon interference experiments to determine the 1/e coherence times of these photons, and Hanbury-Brown and Twiss (HBT) experiments to determine the degree of single photon emission. Figure 3 shows correlation measurements for a pillar with the X line on resonance with the cavity mode. The area of the peak at time zero, $g^{(2)}(0)$, gives the probability of measuring coincidences on the two single-photon detectors. With excitation at 750 nm we observe a central peak area of 0.25 (Fig. 3(b)). Independent spectral measurements of the intensity of the background emission indicate this is consistent with perfect single photon emission from the X state and poissonian emission from the background. Under similar conditions single photon interference measurements on the X emission lines of these sources reveals coherence times, τ_{coh}, of between 80 and 100 ps.

Finally, excitation resonant with an excited state in the QD at 910.9 nm was used to avoid population of the wetting layer or surrounding GaAs. Again, this was carried out using on-axis excitation. A significantly higher excitation density was required so as to obtain similar count rates because the upper Bragg mirror is highly reflective at these wavelengths. Figure 4 is a typical single photon interferogram of the emission from the X state under these conditions. Here we have measured coherence times as long as 200 ps which is significantly longer than that obtained with non-resonant excitation. When considering possible use of this source in quantum computation schemes the important figure of merit is $2\tau_{rad}/\tau_{coh}$ which provides a measure of the degree of indistinguishability of the emitted photons. This parameter is 1 when the photons are fourier transform limited and totally indistinguishable. For this pillar microcavity the ratio $2\tau_{rad}/\tau_{coh}$ is 2.4. This compares well to previously published values [2] where consecutively emitted photons were shown to have a high degree of indistinguishablity in a 2-photon interference experiment. We anticipate that with further optimisation of this source we could approach the time-bandwidth limit. Figure 3(a) shows a correlation measurement recorded with the excitation tuned to an excited state of the QD at 910.9 nm. We observe $g^{(2)}(0)$ is less than 2 %, which represents a fifty-fold reduction in the probability of generating more than 1 photon in a pulse relative to a classical, poissonian source of the same intensity.

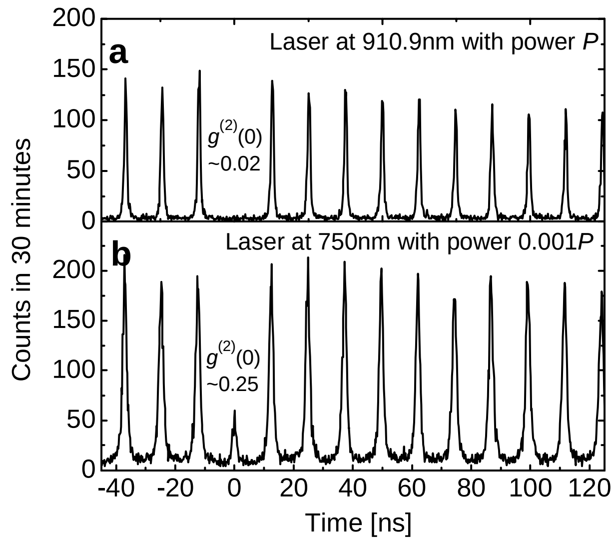

Fig. 3. Pulsed correlation measurements from a pillar with a single exciton emission line near the cavity resonance at 939.2 nm, for (a) quasi-resonant excitation at a wavelength of 910.9 nm, coincident with an excited state in the QD and (b) above bandgap excitation at 750 nm. *P* corresponds to a power density of 16 $KWcm^{-2}$.

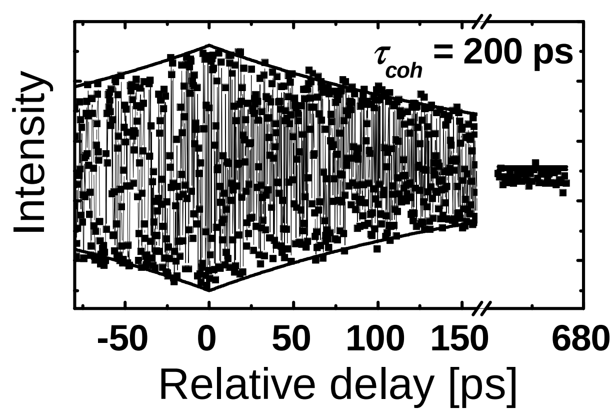

Fig. 4. Single photon interferogram from the *X* state, with excitation resonant with an excited state in the QD.

5. Conclusions

To conclude, we have used photolithoghrapic techniques to reproducibly fabricate pillar microcavities. For the exciton states of single quantum dots in pillars with dimensions significantly larger than those previously reported we have directly measured Purcell factors up to 4, coherence times of 200 ps, collection efficiencies of 10 % and probabilities of multi-photon emission below 2 %.

Acknowledgments

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