Purcell enhancement of a deterministically coupled quantum dot in an SU-8 laser patterned photonic crystal heterostructure

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Enhancement of single photon source emission through cavity quantum electrodynamics is key to the realization of applicable emitters in many quantum optics technologies. In this work we present a flexible and convenient cavity fabrication process that writes a SU-8 microstrip onto a photonic crystal waveguide deterministically, in which InGaAs/GaAs quantum dots are present as emitters. The strip cavity is laser patterned at the location of a quantum dot with a chosen emission wavelength. Micro-photoluminescence studies are undertaken that demonstrate an enhanced emission intensity by a factor of 2.1 with weak coupling to a single quantum dot and time-resolved photoluminescence further shows a Purcell enhancement factor of 2.16. The fabrication process is thus verified as a reliable recipe to introduce deterministic cavity coupling to a chosen quantum dot.

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Single photon sources (SPS) are highly sought after for applications in the fields of quantum information processing and quantum communication[1–3], such as for the well-known BB84 quantum key distribution protocol[4]. A common approach to creating a SPS based on a single quantum dot (QD) is to embed it into an optical cavity. By achieving spectral resonance between a QD emission line and a cavity mode, a coupling can be achieved in which photons are emitted into the cavity mode. This coupled cavity-emitter system enables modification of the spontaneous emission dynamics of the QD, known as the Purcell effect[5,6]. Coupling of a photonic crystal (PhC) cavity mode to a single QD can offer the potential to realize a Purcell-enhanced SPS with highly directional free-space emission[7,8]. Such SPSs can also be coupled into optical fibers[9] or waveguides[10–12]. Early work on QD- cavity coupling had many dots coupling to a single cavity, because of the high density of QDs in the samples[13]. Progress in growth techniques now allows reliable fabrication of low density QD samples[14], which are suited to achieving coupling between a single QD and a cavity in order to achieve a Purcell- enhanced SPS[15,16]. More extensive research on coupling in low density samples can be found in references[6,17–19] . It remains a challenge to produce a cavity mode which couples to an individual dot, as the positional accuracy has to be within the cavity mode volume and simultaneously the mode wavelength needs to match that of the chosen dot. In this work, we demonstrate a convenient method of achieving deterministic coupling between a single QD and a PhC cavity mode. Using microphotoluminescence (μPL) techniques, a target single QD in a PhC waveguide is identified, and a laser-patterning procedure incorporating SU-8 negative photoresist is used to create a PhC cavity mode to couple to the target QD. In the resulting device, a PhC cavity mode with a good Q/V_0 ratio is defined by a cross-linked SU-8 structure on top of the PhC waveguide[20,21]. The technique allows this cavity mode to be positioned to achieve spatial overlap with the target QD; close spectral proximity between the emitter and cavity is achieved by selecting the target QD based on known properties of the cavity mode.

The photonic crystal sample was grown by molecular beam epitaxy and consisted of a GaAs wafer with a 1 μm thick sacrificial layer of $Al_{0.7}Ga_{0.3}As$ capped with 100 nm of GaAs. A 0.7 nm layer of InAs was then deposited followed by 6 nm of InGaAs, resulting in the formation of QDs ~ 20 nm wide, at a density of $\sim 5\times10^8\,cm^{-2}$ and a capping layer of 94 nm of GaAs[22]. This structure is later processed via e-beam lithography and reactive ion etching to produce a series of 2D PhC waveguide as shown in figure 1[21].

The deposition of a SU-8 film on the PhC waveguide red shifts the waveguide band edge as shown in Sfig1. This shift is sufficient to create a mode gap cavity[23,24] when patterning a SU-8 strip across the PhC waveguide (Sfig2). This confinement of photons in the mode gap cavity is akin to the electronic quantum confinement in a quantum well. Previous theoretical and experimental results confirm that a cavity mode is created successfully by writing either a SU-8 disk or a SU-8 strip (running perpendicular to the waveguide) on top of a PhC waveguide[20,21].

Figure 1a demonstrates how SU-8 photoresist can be pro-

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cessed to fabricate the mode gap cavity on a registered dot. A mixture of SU-8 and cyclopentanone (ratio of 1:7) is first spin coated onto the sample surface, followed by baking at 95°C for 5 min to give a green colored film (figure 1b)[20]. Then the coated sample is cooled down to 4 K in a cryostat. A weak 532 nm continuous wave (CW) laser (which does not expose the SU-8 photoresist) is used to excite the sample through an optical fiber to look for dots of the desired emission wavelength within the missing line of holes by using a piezo-controlled scanning objective. Once a dot has been identified and registered (to an accuracy of ± 40 nm)[25], a 405 nm continuous wave laser, which is coupled into the same fiber, is then focused onto the deposited SU-8 layer with a power of 210 μW to expose a line passing over the selected QD perpendicular to the missing line of holes with a writing speed of 0.04 μms^{-1}. Thus, the registration and exposure occur in one experimental session, and many such mode gap cavities can be written on different PhC waveguides in succession, or on the same PhC waveguide to create a photonic molecule[20]. After laser patterning, the sample is removed from the cryostat and is heated at 90°C again for 5 min and the surrounding unexposed SU-8 residue is then washed out using PGMEA. The resulting SU-8 strip acts as an optical cavity. The overall mode volume is determined by both the waveguide and SU-8 strip[20].

A schematic and an image of a typical fabricated cavity are shown in figures 1c and 1d. A comparison of parameters for strip writing processes at 4 K and 298 K to give a similar strip size of $1-2\,\mu$m width and 0.1 μm thickness is shown in table I to illustrate the change in exposure power and dose necessary at cryogenic temperatures[20]. Previous mode gap cavities defined by patterning strips on the surface of PhCs were created at room temperature[26,27], here we demonstrate PhC cavity creation at cryogenic temperatures, allowing deterministic coupling with a quantum dot. The SU-8 strip has a width of $1-2\,\mu$m and a predicted thickness of 0.1 μm, which is patterned on a PhC waveguide with a 340 nm lattice spacing, 185 nm hole diameter and 200 nm slab thickness. Ths produces a cavity mode resonance wavelength of 1283 nm at 4 K. This wavelength is within the spectral region of possible InGaAs/GaAs QD emission and determines the QD emission wavelength that is chosen for a registered dot to enable cavity coupling.

	4 K	298 K
Exposure laser power (μW)	210	17.5
Writing speed (μs^{-1})	0.04	0.25 – 0.50

TABLE I. A comparison of parameters for strip writing processes at 4 K and 298 K onto a film of thickness 100 nm to give a strip of 1–2 μm width.

At cryogenic temperatures, the emission of a single QD itself becomes visible at a relatively low excitation power (<0.25 μW) as thermally induced broadening through phonon coupling is less prominent. However, collective emission from an ensemble of QDs can still undergo inhomogeneous broadening due to uncontrollable minor variations in their geometries and sizes, which is inevitable in the sample growth

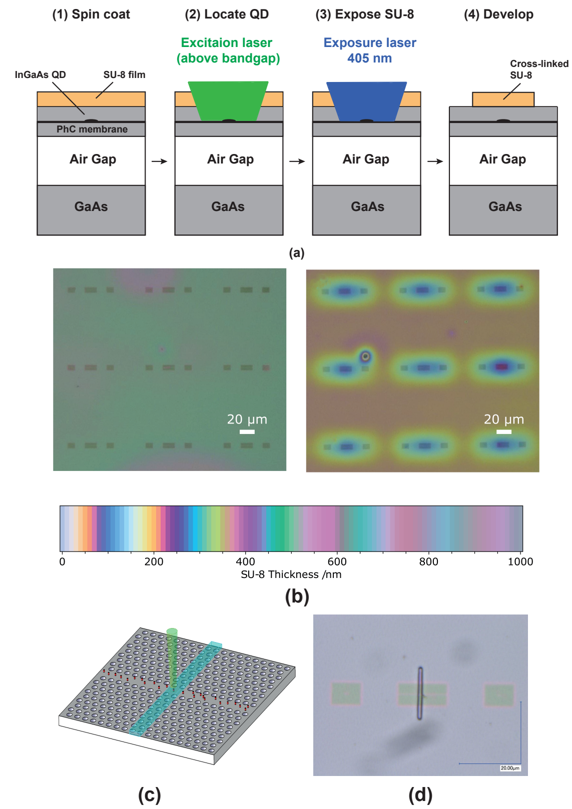

FIG. 1. (a) Cavity fabrication via laser exposure of SU-8 at 4 K. (b) Microscope image of spin-coated SU-8 thin film with color scale for thickness reference, left: coated uniform film of ~ 500 nm thickness; right: Thinning of the SU-8 film on top of the photonic crystal for a thin spin-coated layer (about 300 nm thick) (c) 3D schematic of the SU-8 strip cavity. The green transparent cone represents excitation laser 532 nm. Cyan strip marks the cavity and red dots represent QDs. (d) An image of our fabricated cavity.

process as shown in figure 2a[28]. It is hence necessary to pick out a single emitter by filtering out a QD both spatially and spectrally. μPL studies of devices doped with high and low densities of dots before depositing SU-8 (figure 2) show that in the case of a sample a with low density of QDs the individual emission lines can be distinguished clearly enough from each other for a 1 μm^2 excitation area and a single emitter can then be spectrally addressed via cavity coupling. In figure 2b a single QD emission at the predicted cavity mode wavelength of an SU-8 strip is indicated at 1283.5 nm and the cavity is then drawn at the same location to evaluate the coupling effect. The challenge in finding a QD at the exact same wavelength as the cavity mode is reduced by having many dots lying within the missing line of holes along the waveguide. Thus, performing a 1D μPL scan to choose a particular QD becomes feasible. This procedure together with the ease of writing a strip cavity anywhere along the waveguide make the production of a coupled system much more likely

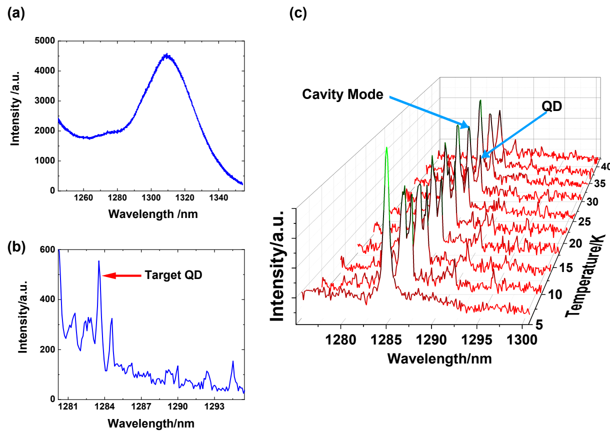

FIG. 2. (a) PL of an ensemble of high density InGaAs/GaAs quantum dots at 4 K. (b) PL of target QD near 1283.5 nm well separated from the emission from other QDs before cavity deposition. (c) temperature run after creating SU-8 cavity at the location from (b) showing coupled emission at 6 K; the QD starts shifting away from the cavity mode as the temperature increases.

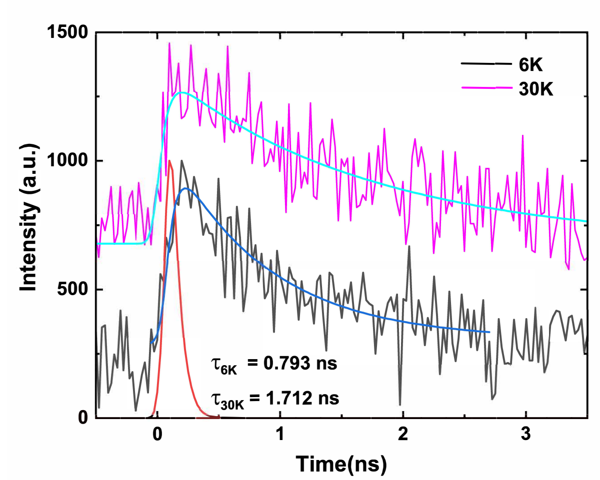

FIG. 3. Time-resolved photoluminescence comparison from cavity coupled emission at 6 K towards uncoupled emission at 30 K. A Purcell Enhancement factor of 2.16 is calculated.

and allows for reproducible coupling. The μPL emission as a function of temperature was measured after creating an SU-8 cavity on top of the location of a single QD with matching emission wavelength, to enable temperature tuning of the coupling. Figure 2c shows enhanced emission from the coupling effect at 6 K when the QD wavelength coincides with the cavity mode, with a maximum enhancement in intensity of 2.1 ± 0.1 at resonance. As the QD line redshifts with temperature and weakens the coupling effect, both lines exhibit a drop in intensity until the QD becomes almost invisible next to the background ensemble emission at 30 K. This crossover of the two emissions clearly demonstrates a coupled system where the emitter is at resonance with the optical cavity. Ideally, this crossover should be engineered to occur at a low temperature to preserve the best emission characteristics of the QD. The coupling at 6 K therefore fulfils this enhancement condition. Coupling of another system ,but at 10K, is shown in the supplementary information (Sfig3). The FWHM of the emission is 0.64 nm and results in a quality factor of over 2000, which is large enough to enable the system to enter the weak coupling regime with Purcell enhancement. Limitation factors such as non-perfect cavity geometry, thick cavity strip, defects in the substrate and the morphology of the QD itself can prevent the quality factor from getting even higher, however, this quality factor is a more realistic number for practical weak coupling performance[29].

In order to verify Purcell enhancement, a comparison of the decay lifetime is performed at 6 K and 30 K (figure 3) when the QD emission is on ($\tau = 0.793 \pm 0.102$ ns) and off ($\tau = 1.712 \pm 0.146$ ns) resonance with the SU-8 cavity mode. These lifetimes were obtained after deconvoluting the instrument response function of the detector (130 ps FWHM). A micrometer-controlled output slit together with a multimode fiber of 25 μm core is used to spectrally filter the emission line and transmit it to an InGaAs APD to measure the photons. A clear reduction in the lifetime is observed at 6 K consistent with weak coupling. The maximum Purcell factor $F_{\mathrm{P}} = \frac{3}{4\pi^2}\left(\frac{\lambda}{n}\right)^3\left(\frac{Q}{V}\right)$ derived from Fermi's golden rule, using our quality factor of 2000 and mode volume of $1.3\,(\lambda/n)^3$ is estimated to be 117 while the measured enhancement at resonance is 2.16 ± 0.14 compared to the uncoupled emission. Such a difference from the theoretically predicted value can be explained by the non-perfect spatial matching between the QD and the cavity and to the fact that the excitation is non-resonant[6]. Indeed, the position of the cavity mode antinodes created by the presence of the strip is dictated by the lattice geometry, not by the strip as shown in Sfig2. The position of the strip dictates the intensity of these nodes, this can be generalised to all multilayer PhC slabs[24]. Thus if a QD is located between the antinodes no coupling would occur even though the cavity is created at the strip location where the QD has been registered. However additional markers on the PhC sample could help determine if the chosen QD is at an ideal location within the PhC waveguide for cavity coupling (see mode pattern in Sfig2). Other reasons for the discrepancy could be due to polarisation mismatch, crystalline imperfections as well as a background contribution from other uncoupled QDs[13].

We have tested a fabrication method for writing a SU-8 cavity on a PhC waveguide and used a μPL setup to obtain deterministic coupling between a QD emitter and the cavity mode. The SU-8 strip can be developed via a laser patterning procedure at cryogenic temperatures at the QD position. The QD is chosen so that its emission wavelength coincides with the predicted cavity mode resonance and therefore increase the likelihood of coupling. The writing process further provides convenience and flexibility in creating a cavity after finding the emitter. μPL temperature dependence clearly demonstrates coupling of a single QD emission and a cavity mode and an increased intensity as they overlap spectrally.

Our procedure enables us to simultaneously identify a suitable QD and pattern a PhC cavity at the same position and opens the possibility for scaling the system to coupled cavities with QDs in one or both cavities, allowing electric field control of emission via a photon blockade mechanism. There is also the possibility of forming a series of coupled cavities, resulting in electromagnetically induced transparency.

I. SUPPLEMENTARY MATERIAL

The supplementary material shows a comparison of one band of a PhC waveguide with and without a layer of $0.1\,\mu$m thick cross-linked SU-8 on the top surface, a schematic of the strip-defined mode gap cavity with the electric field envelope and a further example of deterministic coupling from a different cavity.

II. AUTHOR CONTRIBUTIONS

H. Shao and G. Ying contributed equally to this work, other authors contributed to the analysis and the text and planned the experiments. We acknowledge from the Engineering and Physical Sciences Research Council (EPSRC) (grant EP/K014978/1) and from Hitachi Cambridge Research Laboratory.

III. DATA AVAILABILITY

The data that support the findings of this study are openly available at https://ora.ox.ac.uk/objects/uuid:839ad408-eaa6-4552-a9b5-7f713874363b.

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