Control of spontaneous emission from InP single quantum dots in GaInP photonic crystal nanocavities

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Abstract

We demonstrate semiconductor quantum dots coupled to photonic crystal cavity modes operating in the visible spectrum. We present the design, fabrication and characterisation of two dimensional photonic crystal cavities in GaInP and measure quality factors in excess of 7,500 at 680nm. We demonstrate full control over the spontaneous emission rate of InP quantum dots and by spectrally tuning the exciton emission energy into resonance with the fundamental cavity mode we observe a Purcell enhancement of ~8.

Two dimensional photonic crystal (PC) cavities provide a means to confine and manipulate light within a nanoscale volume. By integrating self assembled quantum dots (QDs) into the PC cavities, we can create a spatially well-defined system, ideal for studying cavity quantum electrodynamics[1] and a strong candidate for scalable quantum information processing applications.[2] Excellent progress has been made towards these goals using In(Ga)As quantum dots embedded in GaAs PC cavities and there have now been several demonstrations of strong coupling in this system.[3-6]

Another important application of photonic crystal cavities is the control of the spontaneous emission dynamics of single QDs. Single photon sources are of fundamental importance for quantum key distribution schemes[7] and linear quantum computation.[8] By coupling a quantum emitter to a cavity the Purcell effect can be utilised to improve the extraction efficiency and repetition rate, which has been demonstrated using In(Ga)As QDs in GaAs PC cavities.[9-11]

However, for certain applications the emission wavelength of these QDs (~900-1300nm) is not ideal, as it is far from the maximum efficiency of commercially available Si detectors. InP quantum dots embedded in GaInP provide an alternative and have the significant advantage of emitting in the red spectral range, at the maximum efficiency of Si detectors. This makes InP quantum dots an attractive proposition for implementation in optical quantum information processing and free space quantum communication.[12] A further advantage of InP QDs is that single photon emission has been demonstrated up to 80K, opening up the possibility of a liquid nitrogen cooled single photon source.[13]

Photonic crystals operating in the visible are also of interest for coupling to external light emitters, such as nitrogen vacancy centres in diamond nanocrystals[14] and fluorescent molecules.[15] Most work on PC cavities in this wavelength range has been carried out using SiN_4 membranes, where the Q factor is limited by the low refractive index (n=2), with reported Q factors of ~1500 for L3 cavities[15] and 3,400 for heterostructure cavities.[16] GaP[17] and GaInAlP[18] have also been used to fabricate L3 cavities with modes in the red, with maximum Q factors of 1,700 and 2,000 respectively.

In this work we show that L3 PC cavities can be fabricated in GaInP, with Q factors as high as 7,500 and we demonstrate semiconductor quantum dots coupled to photonic crystal cavity modes operating in the visible spectrum. Using time resolved photoluminescence measurements we observe spontaneous emission suppression of the QDs within the photonic bandgap and for a resonantly coupled QD we observe a Purcell enhancement of ~8.

The wafers used in this study are grown by metal organic vapour phase epitaxy (MOVPE) on 3˚ mis-orientated (100) GaAs wafers. In this work, two different wafers are investigated, which we will refer to as Sample A and B. Sample A consists of a 100nm GaInP waveguide with a layer of InP quantum dots at its centre, grown on top of a 670nm undoped $Al_{0.6}Ga_{0.4}As$ sacrificial layer. Sample B has a similar structure but incorporates a p-i-n junction into a 110nm thick waveguide region and was grown on an n-doped substrate. This sample was intended for devices incorporating electric

field tuning[5], although this will not be discussed here. For the QD growth, 3.3Å InP was deposited at a growth rate of 1.1Å/s at 650°C.

The photonic crystals were fabricated using electron beam lithography and a $SiCl_4$ reactive ion etch (RIE) and the suspended membrane was released by selectively wet etching the $Al_{0.6}Ga_{0.4}As$ layer using hydrofluoric acid. Fig.1(a) shows an SEM image of a completed L3 photonic crystal cavity. Finite difference time domain (FDTD) simulations of the modified L3 defect cavity[19] were performed to determine the parameters required for fundamental mode emission in the region 670-700nm and optimised for maximum Q-factor. Fig. 1(b) shows the calculated electric-field profile for the optimised cavity design, with lattice period a=187nm, hole radius r=0.3a and end hole shift s=0.18a. The simulated Q-factor for this structure was ~85,000.

The L3 cavities were characterised using micro-photoluminescence (μPL) in a helium flow cryostat at ~10K. Emission from the QD ensemble was excited using a HeNe laser emitting at 633nm, focused to a spot with diameter of ~1μm using a 100X objective lens (N.A =0.75). The emitted light was collected using the same objective, before being dispersed by a 0.55m spectrometer and detected with a charge coupled device (CCD) camera. For time resolved spectroscopy we used a 405nm pulsed diode laser, with a pulse width of ~50ps and an avalanche photo diode with a temporal resolution of ~40ps, resulting in a system response function with FWHM of ~100ps.

The photoluminescence spectrum of a typical L3 cavity on Sample A is shown in Fig. 1(c). Emission from the fundamental mode, labelled M0, can be seen at ~681nm. Higher order modes, M1 and M2, are also observed at ~657 and 650nm respectively.

Polarization dependent measurements reveal M0 and M1 to be linearly polarized along the y-direction of the cavity with M3 linearly polarized in the x-direction, verifying this assignment of the modes.[20] Fig. 1(d) shows a Lorentzian fit to the emission from M0 and reveals a Q-factor of ~7,500. Due to the limited spectral resolution of the setup this value represents a lower limit of the Q-factor but still represents a significant improvement for photonic crystals operating in the visible spectrum and is a promising indicator for achieving strong coupling in this system.

The QD density of sample A is high and prevents the observation of single QD lines in the μPL spectra. For this reason, we turn to Sample B to investigate the effect of the PC cavity on the emission properties of single QDs. Fig. 2(a) shows the μPL spectrum recorded at low pulsed excitation power from QDs in the bulk semiconductor, away from the patterned photonic crystals. Bright, narrow lines corresponding to individual QDs are observed and by selecting a well isolated line we can measure the decay dynamics of the QD emission. Similarly, for QDs embedded in photonic crystals we observe individual emission lines, as well as the cavity mode emission at ~695nm (Fig. 2(b)). We identify single emission lines from these spectra, labelled QD_{bulk} and QD_{PC1}, and perform power dependent and time resolved measurements. Both transitions saturate at a similar excitation power, but with a significantly higher intensity measured from QD_{PC1} than QD_{bulk}. This suggests the PC facilitates a ~3-fold enhancement of the extraction efficiency.[21] It is also interesting to note that the count rate is ~10 times higher than we observe from InAs QDs in GaAs PC cavities in our system, which we attribute to the improved efficiency of the Si CCD in this wavelength range and emphasises the advantage of this material system. In Fig. 2(c) we compare the decay transients of QD_{bulk} and QD_{PC1} and observe a

lifetime, τ_{bulk}=0.66ns, which is in good agreement with previous measurements of InP QDs.[13] In contrast to this for QD_{PC1} we see a significantly extended lifetime τ_{bulk}=13.1ns, which results from a reduction in the photonic density of states due to the 2D photonic bandgap.[22]

We now look at QDs not just in the photonic bandgap, but coupled to a cavity mode. Fig. 3(a) shows the μPL spectrum for a second cavity on sample B, where we observe several QD lines spectrally close to the fundamental mode. In this case, the mode has Q =3,300, which corresponds to a linewidth of ~0.21nm. Here, we concentrate on the QD detuned from the mode by $\Delta\lambda$=0.56nm, where $\Delta\lambda=\lambda_{mode}-\lambda_{QD}$, labelled QD_{PC2} in Fig. 3(a). By increasing the temperature we gradually decrease $\Delta\lambda$ until QD_{PC2} comes into resonance with the mode at 44.5K. Fig. 3(a) also shows the μPL spectrum for the $\Delta\lambda$=0 case. Fig. 3(b) shows that this tuning has a dramatic effect on the QD lifetime, which decreases from ~0.75ns when $\Delta\lambda$=0.56nm to ~90ps at $\Delta\lambda$=0. The decay transient is fitted with a bi-exponential decay, with the ~90ps decay attributed to the Purcell enhanced emission from the QD and a longer lived decay of ~9ns attributed to non-resonant feeding of the mode from other QDs within the PC.[23,24] The decrease in lifetime corresponds to a Purcell enhancement of ~8. To verify that this increase in the spontaneous emission rate is due to the Purcell effect, we measure the QD lifetime at various detuning, as shown in Fig. 3(c). The enhancement of the spontaneous emission rate is expected to follow the Lorentzian lineshape of the mode as the QD is detuned from resonance[25]. The solid line in Fig. 3(c) shows a Lorentzian fit to the lifetime measurements, with a FWHM of 0.25nm, which is in good agreement with the measured linewidth of the mode and confirms that the reduction in lifetime results from the Purcell effect.

In summary, we have demonstrated control over the spontaneous emission rate of InP QDs using L3 photonic crystal cavities in GaInP operating in the red spectral range. In measurements of off resonant QDs we observe a significant increase in the exciton lifetime. Furthermore, we observed a QD/cavity system in the weak coupling regime and measured a Purcell enhancement of ~8. Maximum Q-factors of ~7,500 were measured in the cavities, which opens up the possibility for reaching the strong coupling regime in this system where the emission wavelength is ideal for integration with Si detector technology.

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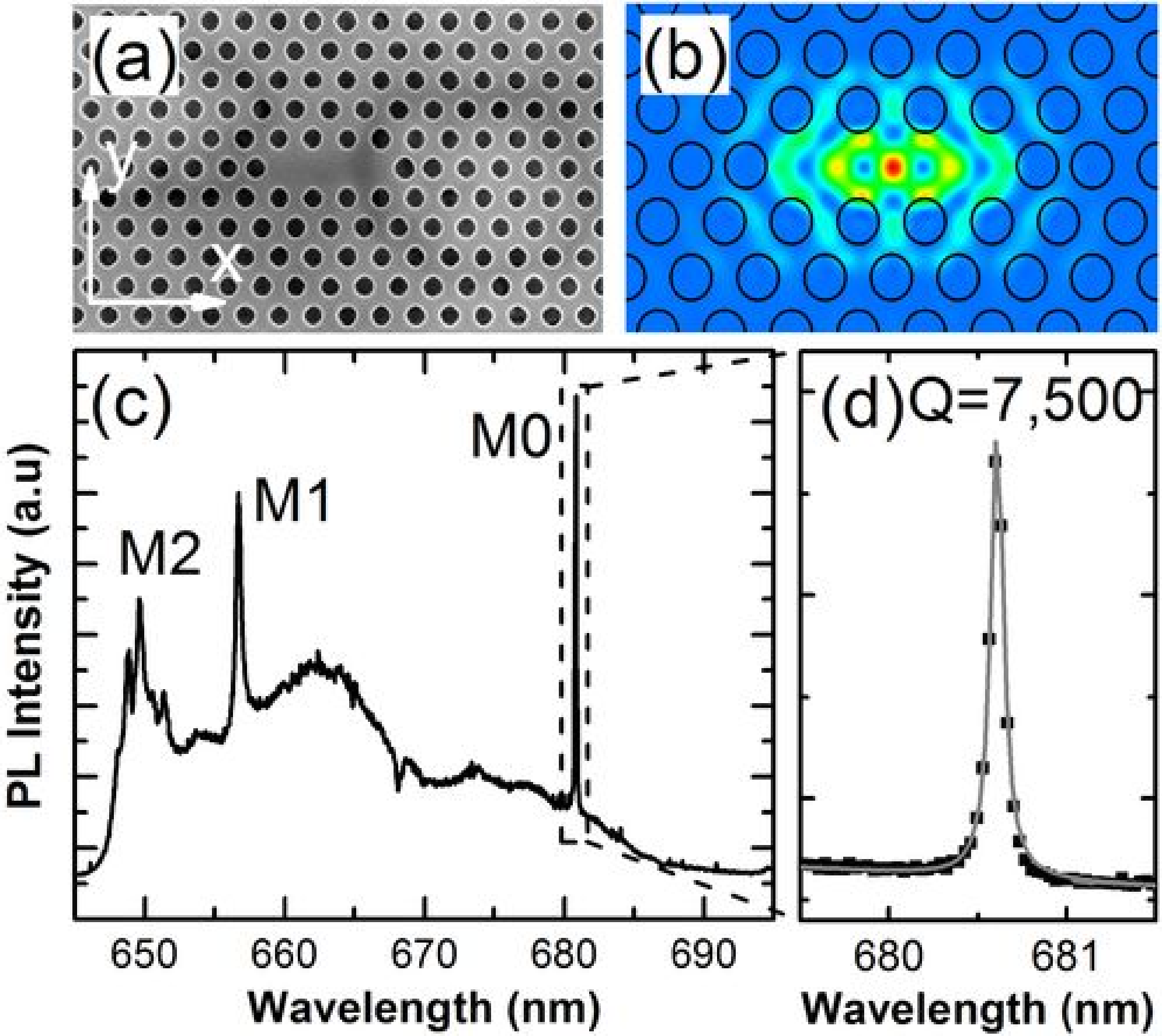
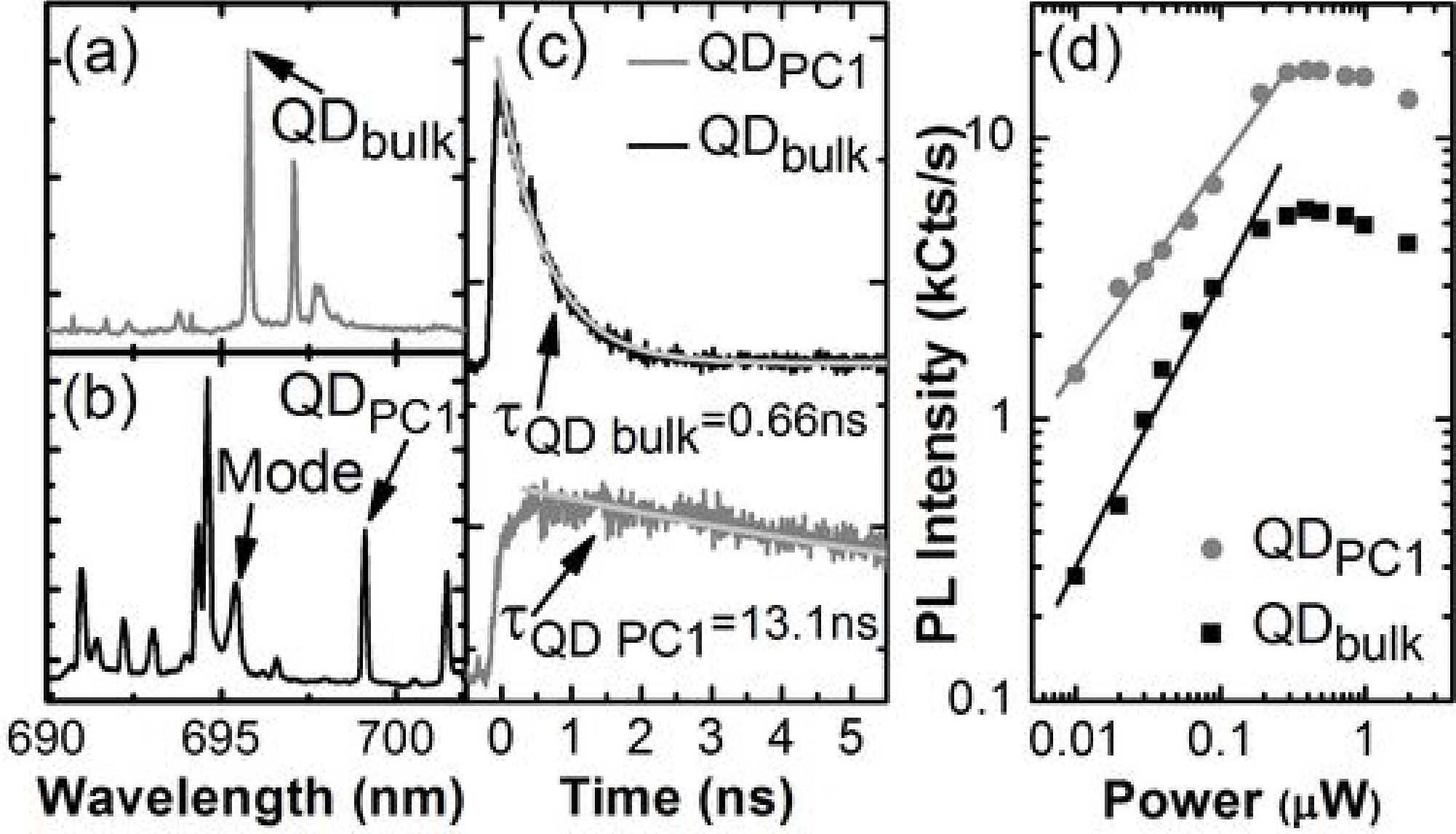
Figure Captions

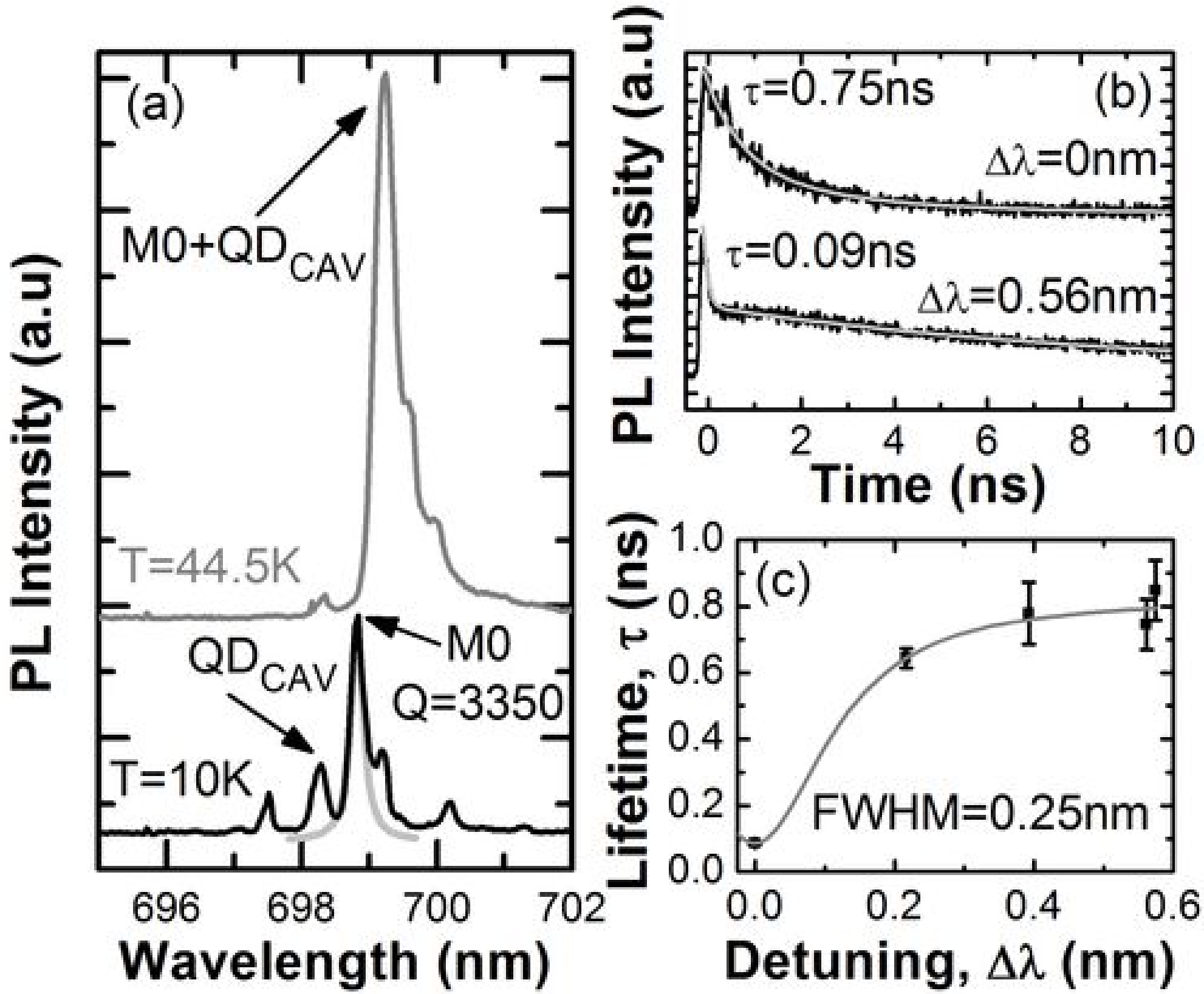
Fig. 1. (a) Scanning electron microscope image of an L3 photonic crystal cavity fabricated in a 100nm thick GaInP slab waveguide. The lattice constant a=187nm, hole radius r=0.29a and end hole shift s=0.18a. (b) Calculated electric field of the L3 fundamental mode. (c) Micro-photoluminescence spectra recorded from a typical GaInP L3 photonic crystal, showing emission from InP QD ensemble (650-685nm) and cavity modes. (d) Fundamental mode emission of cavity with quality factor of ~7,500.

Fig. 2. (a) Comparison of photoluminescence spectra recorded from QDs in the bulk GaInP (QD_{bulk}) and photonic crystal (QD_{PBG}) under pulsed excitation. (b) Decay transients recorded from QD_{bulk} and QD_{PC1}. Light grey lines show mono-exponential fits. (b) Pulsed power dependent PL Intensity of QD_{bulk} and QD_{PC1}, the solid lines show linear fits to the data.

Fig. 3. (a) Photoluminescence spectra of weakly coupled QD-cavity system recorded at 10K and 44.5K. Light grey line shows a Lorentzian fit to the cavity mode resonance at 10K, with FWHM=0.25nm. (b) Decay transients recorded from the QD at 10K ($\Delta\lambda$=0.56nm) and 44.5K ($\Delta\lambda$=0nm). (d) Measured QD lifetime as a function of QD/cavity detuning. Solid line shows Lorentzian fit, with FWHM=0.25nm.


(a)
y
x
(b)
(c)
M0
M1
M2
PL Intensity (a.u)
650
660
670
680
690
Wavelength (nm)
(d)Q=7,500
680
681
Wavelength (nm)


(a)
QDbulk
(b)
QDPC1
Mode
690
695
700
Wavelength (nm)
(c)
QDPC1
QDbulk
τQD bulk=0.66ns
τQD PC1=13.1ns
0
1
2
3
4
5
Time (ns)
(d)
PL Intensity (kCts/s)
10
1
0.1
QDPC1
QDbulk
0.01
0.1
1
Power (μW)


(a)
M0+QD$_{CAV}$
T=44.5K
QD$_{CAV}$
M0
Q=3350
T=10K
PL Intensity (a.u)
696
698
700
702
Wavelength (nm)
(b)
τ=0.75ns
Δλ=0nm
τ=0.09ns
Δλ=0.56nm
PL Intensity (a.u)
0
2
4
6
8
10
Time (ns)
(c)
FWHM=0.25nm
Lifetime, τ (ns)
1.0
0.8
0.6
0.4
0.2
0.0
0.0
0.2
0.4
0.6
Detuning, Δλ (nm)