Origins of Spectral Diffusion in the Micro-Photoluminescence of Single InGaN Quantum Dots

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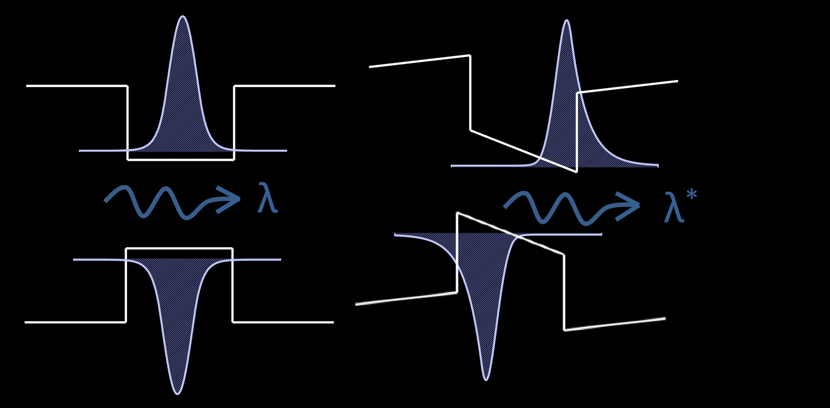
1. Introduction

A coupled optical microcavity-quantum dot system achieved with InGaN quantum dots (QDs) could provide an electrically driven, tunable single photon source, a significant boon for the development of linear optical quantum computation and quantum cryptography systems.

Micro-Photoluminescence measurements carried out on single InGaN quantum dots embedded in GaN show a significant temporal variation in the wavelength of the narrow (~300μeV – 3meV) μPL peaks characteristic of QD emission. This is ascribed to Stark shifts induced by randomly generated local electric fields. An understanding of the causes of this "spectral diffusion", and whether it can be overcome, is essential progress towards a coupled cavity-QD system, since temporal variation in emission wavelength of the QD would lead to a detuning from the cavity resonance.

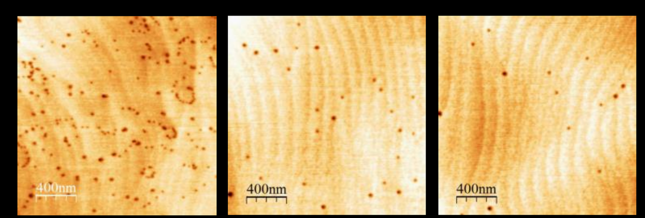


Quantum confined Stark effect. Left: No electric field. Right: Band-bending induced by internal piezoelectric field, this can also be influenced by external fields leading to spectral diffusion.

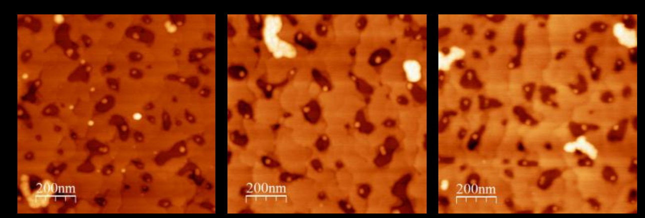
2. Sample Growth & Preparation

Three GaN epilayers (shown right) with threading dislocation densities of 3.5×10^8 cm^{-2} (LDD), 8×10^8 cm^{-2} (MDD) and 6×10^9 cm^{-2} (HDD) were grown by metal-organic vapour phase at Cambridge university.
QDs were formed on the different pseudo-substrates by the growth of a ca. 2.5nm InGaN epilayer which was then annealed in N_2 at the growth temperature, before being overgrown with a GaN cap.
The InGaN epilayer is damaged by this process but still acts as a quantum well (QW). This growth process, termed "modified droplet epitaxy", is still not fully understood.

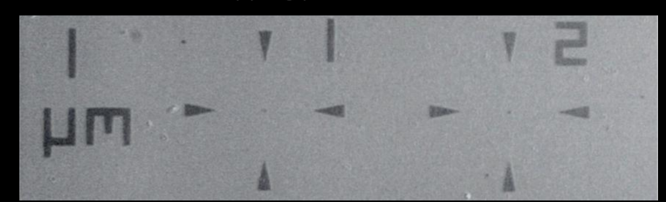
Each of the capped samples were masked with a pattern of 1μm diameter circular apertures using e-beam lithography and wet etching. This enables isolation of ~0.8μm^2 regions of each sample, meaning it's possible to isolate single QDs, and to return to a previously studied QD.



AFM images of HDD (left), MDD (centre) and LDD (right) GaN templates. The dots seen are the dislocation sites.



AFM images of HDD (left), MDD (centre) and LDD (right) InGaN epilayers. The light coloured dots are Indium rich droplets which are thought to form into QDs during the N_2 anneal and GaN capping processes.

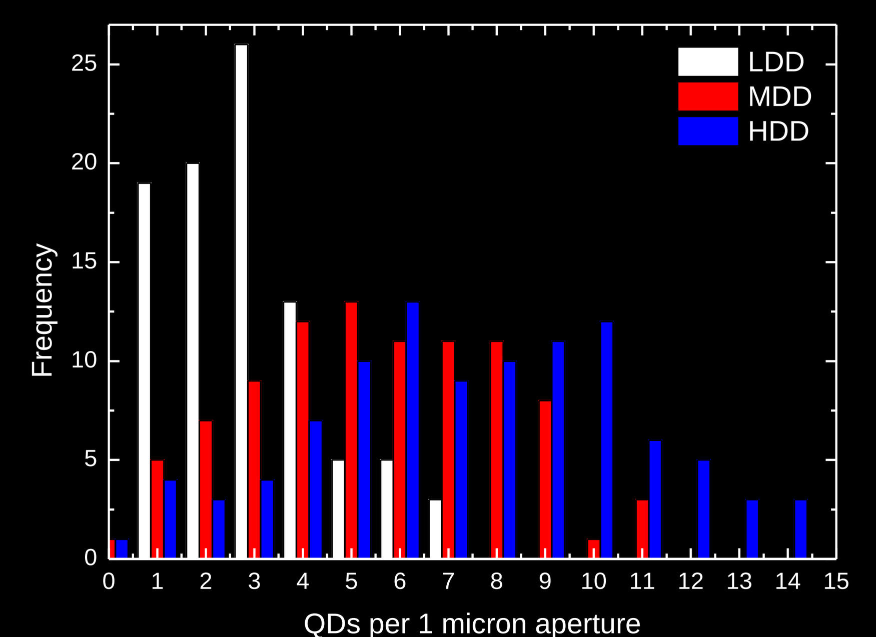


Optical microscope image of the Au mask.

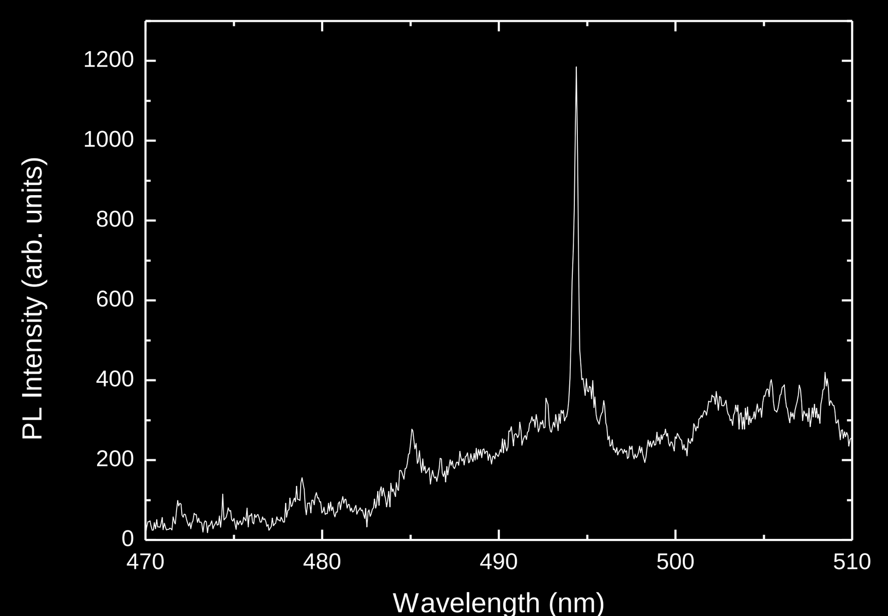
3. Micro-photoluminescence studies

Micro-photoluminescence measurements were performed using a two photon excitation technique, known to preferentially excite the 0D confined states of the QDs over that of the 2D confined QW, suppressing any background QW luminescence.

Low excitation power density (~1MWcm^{-2} 2-photon) studies were used to find the number of QD luminescence peaks per unit area, providing a metric for QD density. This metric was measured as 3.4×10^8 cm^{-2} (LDD), 6.4×10^8 cm^{-2} (MDD) and 9.5×10^8 cm^{-2} (HDD), in each case comparable to the dislocation density. This suggests that threading dislocations play a significant role in the formation of the QDs.

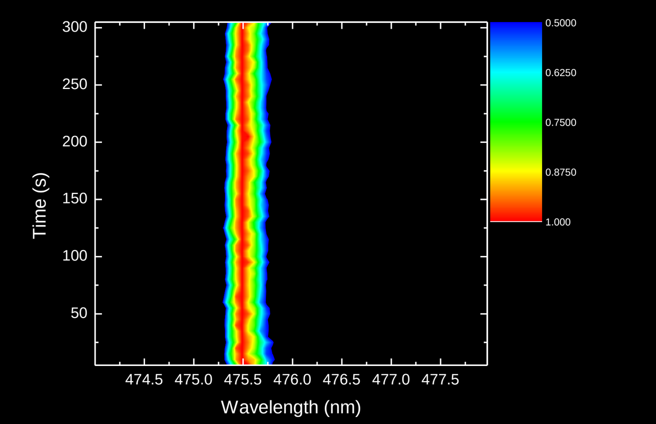


Histograms showing the number of QD emission lines present in a 1μm diameter circular aperture, for 100 of such apertures in each sample.

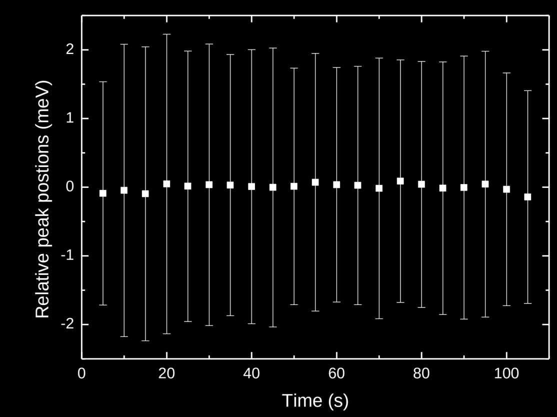


Typical μPL spectra from the HDD sample focused on one QD.

4. Temporal evolution of μPL

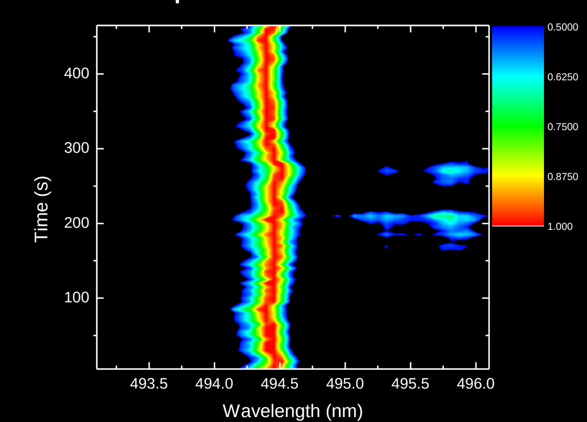


60 consecutive μPL spectra from a QD in the MDD with colour representing normalised PL intensity.

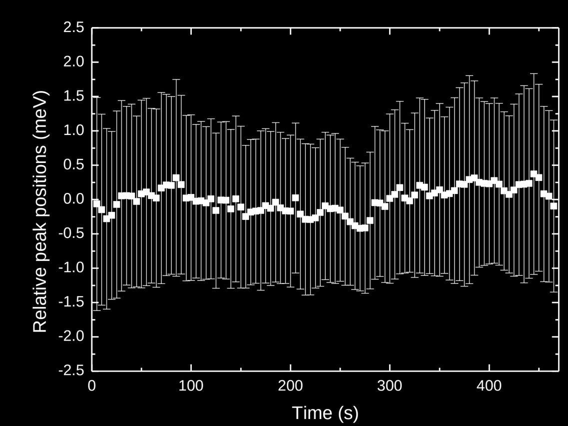


Emission energy shown as a function of time for QD in the MDD sample. The error bars represent the full width half maximum of the QD emission fitted to a Lorentzian peak.

Typical time series results from the MDD sample are shown above. The change in QD emission energy with time is small compared to the QD linewidth (1.6meV). The linewidth is larger than expected for pure radiative decay from a QD, this is attributed to spectral diffusion processes on the timescale of nanoseconds artificially broadening the measured emission peak.



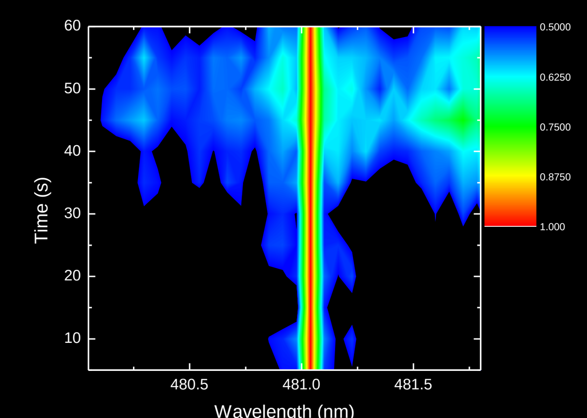
95 consecutive μPL spectra from a QD in the HDD with colour representing normalised PL intensity.



Emission energy shown as a function of time for QD in the HDD sample. The error bars represent the full width half maximum of the QD emission fitted to a Lorentzian peak.

Typical time series results from the HDD sample are shown above. Temporal variations in the emission are much larger (0.3-0.4meV) relative to the QD linewidth (1.2meV). This variations are visible as "hops" in the μPL peak energy clearly visible over timescales of seconds. Since this long timescale spectral diffusion is much more common in the HDD sample than the LDD and MDD, it is attributed to non-radiative carrier traps at threading dislocation sites.

Statistically, the LDD sample shows the least evidence of spectral diffusion, with an example of a particularly well isolated dot shown to the right – spectral diffusion processes are occurring on a scale not observable within the resolution limit (~300μeV) of the optical set-up.



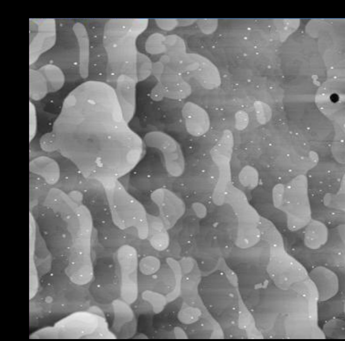
12 consecutive μPL spectra from a QD in the LDD with colour representing normalised PL intensity.

5. Summary

- QD formation is linked to threading dislocation sites
- Spectral diffusion processes occur in self-assembled InGaN QDs on two characteristic time scales: short (nanoseconds), and long (seconds)
- Long timescale spectral diffusion can be attributed to non-radiative carrier traps at threading dislocation sites.

6. Future Work

A new sample has been grown at Cambridge university (AFM right) using a thinner (1.2nm) InGaN epilayer and a prolonged N_2 anneal stage, in order to try to reduce the luminescence from the underlying QW layer. This should provide insight into the role of the QW, and whether carriers excited into the QW, or escaping from QD to QW are responsible for short timescale spectral diffusion.



AFM image of new sample with prolonged anneal time, the distribution of In droplets is more uniform.

Treatment of GaN pseudo-substrates with a SiH_4 flux has been found to increase dislocation pit width. Such a treated substrate will be used to grow InGaN QDs, which will then be masked in the normal way. Both μPL and AFM measurements in the same apertures will be used to investigate a correlation between the number of local dislocation pits and observed QD emission lines. If a correlation exists, using a dislocation pit as a marker for QD position would make spatial resonance between QD and optical microcavity relatively easy to achieve.