Effect of annealing on structure and luminescence of InP/AlGaInP quantum dots

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Lasers based on self-assembled quantum dots (QDs) have attracted widespread interest due to their unique characteristics that include low threshold current density and resistance to threading dislocations. While most work to date has focused on 1.3µm InAs/GaAs QDs, InP QDs have also aroused interest for low threshold lasers in the visible and near-infrared regime. For example, electrically injected room-temperature InP QD lasers showed a J_{th} of 190 A/cm^2 with emission at 740 nm [1]. Growth of phosphides by MBE typically requires a relatively low substrate temperature (< 500ºC) compared to MOVPE (~700ºC), which leads to degraded optical properties and makes post-growth annealing a crucial step to improve the material quality [2]. For InAs/GaAs QDs grown using MBE, thermal annealing leads to a significant blueshift of emission wavelength due to the interdiffusion of In between the QDs and the surrounding matrix [3]. In contrast, little is known about the effects of annealing on InP/AlGaInP QDs. Here we report the exceptional thermal stability of InP/AlGaInP QDs, showing ~50× improvement in room temperature photoluminescence intensity without signs of structural degradation or blueshifting for annealing temperatures T < 875ºC.

We found that annealing improves the optical properties of InP QDs, InGaP QWs and AlGaInP DHs, with QDs showing the greatest benefit and up to 50× enhancement in the integrated PL intensity. While high-T rapid thermal annealing (RTA) of InP QDs led to > 50× intensity increase, the emission spectra showed an abrupt blueshift of ~20 nm for T > 875ºC. Cross-sectional transmission electron microscopy (XTEM) of such QD samples showed a reduction in strain contrast, indicating strong interdiffusion between the matrix and the QDs. Given the lack of blueshift for T < 875^{0}C, we next investigated the effect of furnace annealing for longer times (~10-100 minutes) at lower T and found that similar PL intensity improvements of ~50× could be attained while avoiding any blueshift. XTEM and HAADF-STEM of furnace-annealed samples showed clear strain and composition contrast from individual QDs, similar to the as-grown structure. The remarkable improvement in emission intensity without significant structural changes at T < 875^{0}C proves thermal annealing to be a crucial step towards the development of visible InP QD LEDs and low-threshold lasers using MBE.

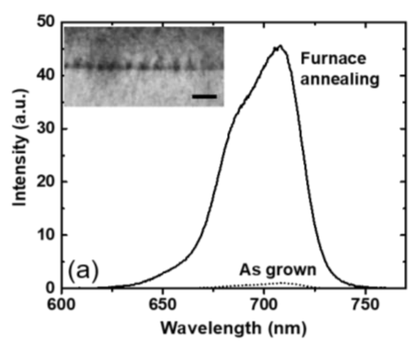

Figure 1: PL spectrum at 300K showing ~50× emission improvement after furnace annealing at 700ºC for 100 minutes (inset: XTEM image showing distinct QDs after annealing, scale bar: 50 nm)

[1] Lutti, Electron Lett., **5**, 247 (2005),
[2] Jalonen, Appl. Phys. Lett., **71**, 479 (1997),
[3] Xu, Appl. Phys. Lett., **72**, 3335 (1998)

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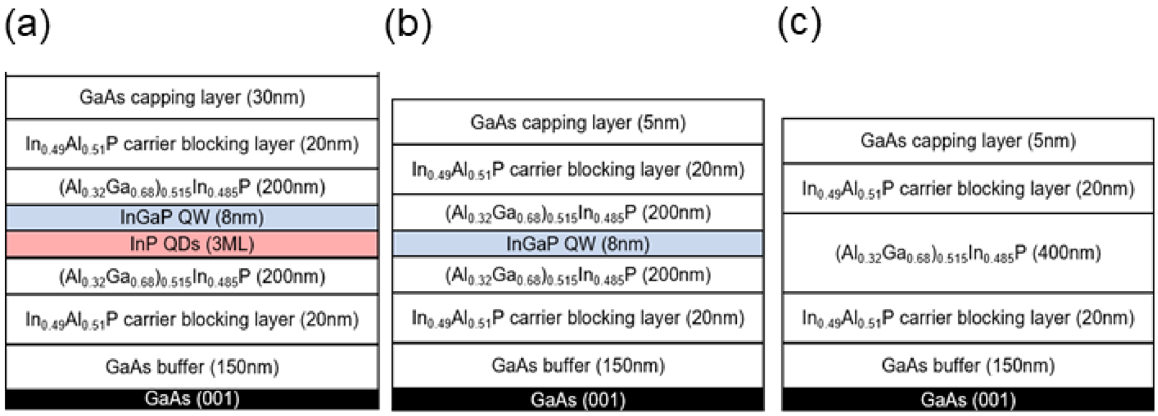

Figure 1: Schematic showing (a) InP QD, (b) InGaP QW and (c) AlGaInP DH structures for PL experiments and subsequent annealing studies.

Sample	I_{PL} as-grown (a.u.) / Peak λ (nm)	I_{PL} after annealing (a.u.) / Peak λ (nm)
AlGaInP DH/GaAs	1 / 589.3	1.9 / 588.3
InGaP QW/GaAs	5.2 / 648.5	72.1 / 641.5
InP QD/GaAs	10.5 / 707.8	522.2 / 705.1

Table I: Effect of optimized annealing conditions on integrated PL intensity of AlGaInP DH, InGaP QWs and InP QDs on GaAs substrate, with InP QDs showing ~50× improvement in the emission intensity. All P-based emitters showed improvement in integrated PL intensity upon thermal annealing. All intensities normalized to the as-grown AlGaInP DH on GaAs.

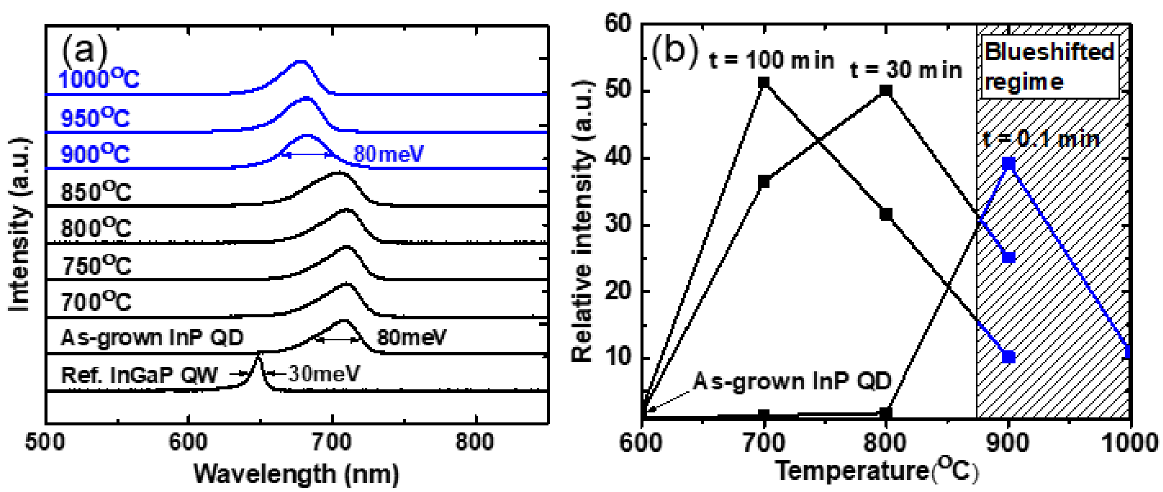

Figure 2: (a) Isochronous PL spectra at t = 0.1 minute demonstrating effect of RTA temperature on QD emission wavelength showing an abrupt blueshift for T ≥ 875°C. The wavelength at T ≥ 875°C still exceeds InGaP QW with a FWHM comparable to as-grown InP QDs. (b) Effect of annealing time on integrated PL intensity of InP/AlGaInP QDs relative to as-grown sample showing ~50× improvement in optical emission by annealing for long time at low temperatures without blueshift.

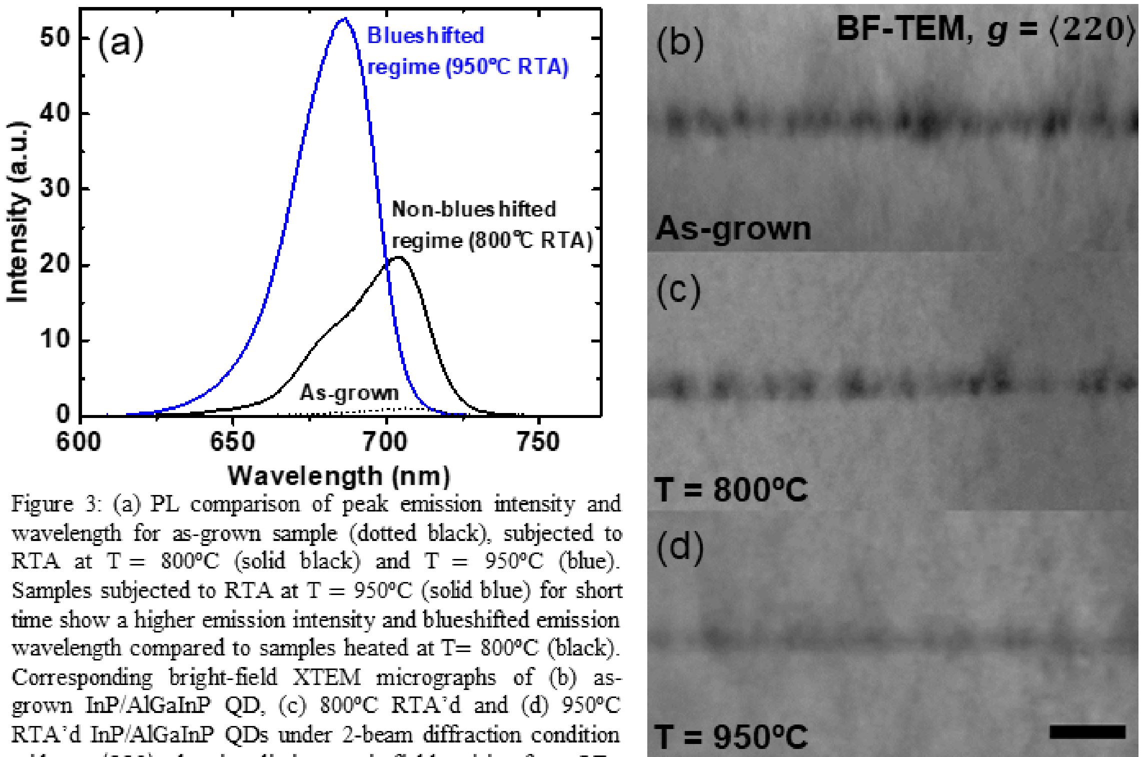

Figure 3: (a) PL comparison of peak emission intensity and wavelength for as-grown sample (dotted black), subjected to RTA at T = 800°C (solid black) and T = 950°C (blue). Samples subjected to RTA at T = 950°C (solid blue) for short time show a higher emission intensity and blueshifted emission wavelength compared to samples heated at T= 800°C (black). Corresponding bright-field XTEM micrographs of (b) as-grown InP/AlGaInP QD, (c) 800°C RTA'd and (d) 950°C RTA'd InP/AlGaInP QDs under 2-beam diffraction condition with $g = \langle 220 \rangle$, showing distinct strain fields arising from QDs in (b) and (c). The QD strain contrast reduces for (d) sample heated at 950°C indicating interdiffusion of InP QDs and surrounding layers; Scale bar = 50 nm.

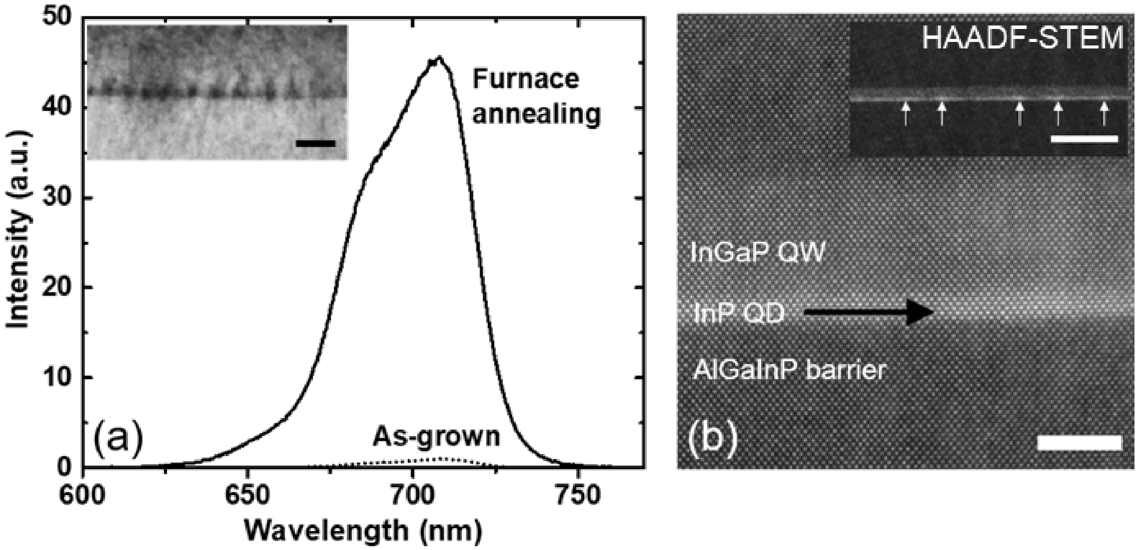

Figure 4: (a) PL comparison of as grown (dotted black) and 700°C/100 minute furnace annealed (solid black) InP/AlGaInP QDs showing ~50× intensity improvement without blueshift in emission wavelength (inset: bright - field XTEM image of 700°C/100 minute furnace annealed InP/AlGaInP QDs showing clear stain contrast from individual dots, $g = \langle 220 \rangle$, scale: 50 nm. (b) High resolution HAADF-STEM image of individual QD subjected to 700°C/100 minute furnace annealing showing composition contrast between InP, InGaP and AlGaInP layers, scale = 5 nm (inset: HAADF-STEM image of 700°C/100 minute furnace annealed sample showing presence of individual QDs marked by arrows, buried by InGaP QW and surrounded by AlGaInP barrier, scale = 50 nm).