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Alloyed-core colloidal quantum dot DFB laser with encapsulated gain region

L. McLellan, B. Guilhabert, N. Laurand and M. D. Dawson
Institute of Photonics, University of Strathclyde, Glasgow G4 0NW, UK
Email: luke.mclellan@strath.ac.uk

Abstract: A CdS_xSe_{1-x}/ZnS colloidal quantum dot distributed-feedback laser operating in the nanosecond-regime with a threshold below 3 kW/cm^2 is reported. The laser emits vertically up to 40 nJ at 600 nm with an efficiency of 1.2%.

Introduction: Colloidal quantum dots (CQDs) are inorganic semiconductor nanocrystals that represent an attractive material for visible-wavelength laser technology. CQDs combine the quantum confinement effect desirable for wavelength versatility and efficient laser operation with solution-processing capabilities that enable great flexibility for device fabrication. A wide range of optically pumped CQD lasers has thus been demonstrated over recent years, including in mechanically-flexible formats [1-6]. While earlier CQD laser demonstrations made use of ultra-fast optical excitation (fs to few ps), the focus is now on closing the gap to real-world applications with compact systems [5]. This means improving the laser performance and establishing the technology for longer pulse operation, from the ns [4-6] to the continuous-wave regime, so that stable diode-pumping operation becomes possible. In this work, we utilise yellow-orange alloyed core/shell CdS_xSe_{1-x}/ZnS CQDs as the gain region of a bilayer vertically-emitting distributed-feedback (DFB) laser. Under 5ns-long pulse excitation at 355 nm, this laser emits up to 40 nJ at 600 nm, limited by the pump energy. Crucially, the threshold is below 3 kW/cm^2, to our knowledge the lowest threshold for a CQD laser operating in such temporal regime.

Device fabrication and experimental setup: The design of the DFB laser is shown in the schematic of Fig. 1. It consists of a 300nm CQD film on top of a surface-patterned glass substrate overcoated with a 180nm PVA layer. The pattern is a rectangular grating with a 380nm periodicity. The PVA acts as an encapsulant that symmetrises the refractive index profile of the overall laser structure. This design increases the laser mode overlap with the CQD region and therefore increases the modal gain. Alloyed-core/shell CdS_xSe_{1-x}/ZnS CQDs (Trilite™ from Cytodiagnostics) with an intrinsic photoluminescence (PL) centred at 575 nm were used. Auger recombination is known to be a limiting factor for laser performance in spherical CQDs with abrupt core/shell interfaces and the detrimental effect is exacerbated in CQDs of small size [7]. Alloying of the core enables the use of CQDs with relatively large size (here typically 6 nm) whatever the emission wavelength (450 nm to 665 nm) while alleviating the issue of the large and abrupt core/shell energy interface. The CQDs were initially in toluene at a concentration of 1 $mg.mL^{-1}$. The toluene was evaporated under vacuum before the CQDs in powder form were re-diluted in a mix of 1.6:1 PMMA in chloroform. This solution was then spun cast onto the glass substrate. After deposition of the PVA the sample was annealed at 30^0C for 120 hours. The amplified spontaneous emission (ASE) characteristics of the gain structure were measured independently of the laser cavity by depositing the same CQD and PVA layers onto a smooth glass substrate (CQD/PVA sample). Another sample on smooth glass but without the PVA layer was also characterized (CQD sample) to verify the effect of the latter. Samples were optically pumped with an Nd:YAG laser, emitting at 355 nm with 5ns pulses and a repetition rate of 10 Hz. The ASE samples were excited with a 1.5mm x 4.4mm stripe and the edge PL was collected for analysis. The CQD sample's edge PL was also measured under CW-excitation from a 10mW, 371nm laser diode with a 6.1mm x 1.7mm stripe. The DFB laser was excited with a 0.74mm x 4.4mm pump stripe and the vertical laser emission was collected for analysis.

Results and discussion: Fig. 1 (a) is an image of the CQD laser above threshold. Figure 2a shows the normalized edge emission of the ASE samples (CQD and CQD/PVA) pumped at a level above the onset of stimulated emission. The edge emission spectrum of the CQD/PVA sample under low intensity CW-excitation is also represented to act as the edge PL reference below the ASE threshold. Compared to the edge PL at 583 nm, the ASE peak for the CQD sample is red-shifted by 6 nm, which is attributed to the bi-excitonic binding energy (type I CQDs). We note that this binding energy is lower than for typical red-emitting CdSe/CdS CQDs [5]. The ASE peak for the CQD/PVA sample is further red-shifted by 9 nm. This further shift is caused by the higher modal confinement in the CQD region, exacerbating the effect of re-absorption of this quasi-three level laser material. The higher confinement though leads to a higher modal gain and the ASE threshold is lower for the CQD/PVA sample (382 µJ/cm^2 vs 815 µJ/cm^2, data not shown). Fig. 2b summarizes the performance of the equivalent CQD/PVA DFB laser. The laser emits principally in the TE$_0$ mode at 600 nm but the TM$_0$ mode can be seen at 597 nm at high pump level. The TE$_0$ total linewidth is 0.45 nm due to the presence of lateral modes. The threshold is only ~13.5 µJ/cm^2 (<3 kW/cm^2), on a par with some organic lasers operated in the same temporal regime. The laser energy for a pump at 3.7 mJ is 40 n J corresponding to an efficiency of 1.2%.

Conclusion: We have reported a CQD DFB laser with what is to our knowledge an ultra-low threshold for the ns temporal regime. This performance is attributed to the nature of the alloyed core CQDs and to the bilayer design of the laser cavity.
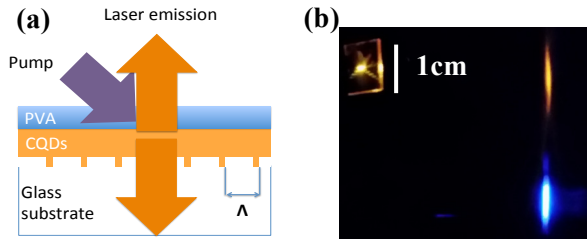

(a) **(b)**

Figure 1: (a) Schematic of the laser structure with grating substrate, a Λ = 380nm CQD layer and a 180nm PVA layer. The laser is pumped at an angle and the laser emits vertically. (b) Image of the CQD laser with its vertical emission (orange, top) and the diffracted pump beam (violet, bottom).
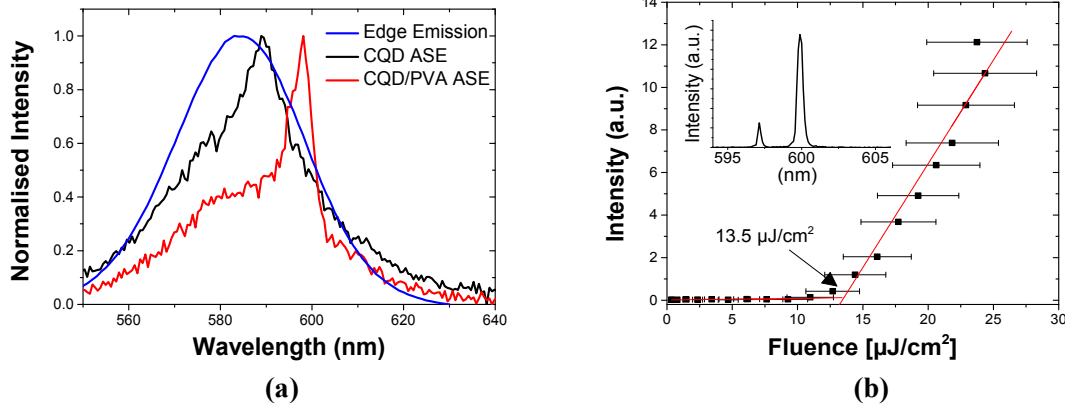

(a) **(b)**

Figure 2: (a) Normalised spectra of the edge emission of alloyed – core/shell CQDs in a film compared ASE shift with and without encapsulation (b) Transfer function of PVA/CQD/DFB laser with optical pump source resulting in a threshold fluence ~13.5 µJ/cm^2 and (inset) multimode laser spectrum with overall linewidth of 0.45nm.

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