Solution processed bulk colloidal nanocrystals as gain material in photonic crystal surface emitting lasers across the green-red spectrum

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ABSTRACT

Colloidal quantum dots (QDs) are heavily investigated for their applications in light emission such as light emitting diodes and, more challenging, lasers due to their appealing processing conditions, compared to e.g. epitaxy, lowering cost and enabling patterning, and tunable optical properties. Using quantum confined Cd-based QDs, several groups have shown light amplification and ensuing lasing action in the red part of the spectrum. Although impressive milestones were achieved, there is to date no single material that can provide the demanding combination of gain metrics to be truly competitive with existing epitaxial growth approaches.

In this work, we take a look at CdS/Se nanocrystals in the regime of vanishing quantum confinement, so-called 'bulk nanocrystals'. We show that these unique materials display disruptive optical gain metrics in the green optical region. Indeed, while showing similar gain thresholds compared to state-of-the-art QD materials, the gain window (440-600 nm, …), amplitude (up to 50.000/cm) and gain lifetime (up to 3 ns) vastly outpace other QD materials.

Using these novel gain materials, we demonstrate lasing in the highly demanded green spectral region (480 – 530 nm) and in the red (650 – 740 nm) both with pulsed and quasi-CW optical excitation. These lasers are made using a Photonic Crystal Surface Emitting Laser (PCSEL) type cavity. As a final step, we attempt to further optimize the lasing properties, be it either narrow linewidth lasers, or high power output, based on in-depth understanding of the hybrid QD-PCSEL laser system.

Keywords: Colloidal Nanocrystals, Optical Gain, Photonic Crystal, Surface Emitting Laser

1. INTRODUCTION

Solution-processible semiconductors have been heavily investigated for optical gain since the first demonstration in the year 2000[1]. Recent advancements have demonstrated optical gain in so-called 'bulk nanocrystals' (BNCs), defined by being larger than the Bohr diameter of the respective material. Because of this, there no longer is any strong confinement in the system. These BNCs, first demonstrated on CdS[2], show remarkable gain metrics that match or surpass the state-of-the-art, for gain magnitude (up to 50.000/cm) and gain lifetime (up to 3 ns). These metrics are achieved while maintaining similar gain thresholds. In this presentation, lasing using BNCs is demonstrated, with a cavity based on a Photonic Crystal Surface Emitting Laser (PCSEL) architecture. Focusing more on the lasers, we further explain the working mechanism behind these, by coupling theory and simulation to the experiment.

2. LASING WITH BULK NANOCRYSTALS

In Figure 1, data derived from measurements on PCSEL devices with CdS and CdSe BNCs spin-coated on a SiNx grating is presented (Figure 1a, sketch for a CdS-based device). All depicted measurements utilize 110 fs pulses, with CdS excited at 450 nm and CdSe at 515 nm. Figure 1b illustrates a $P_{in} - P_{out}$ plot, demonstrating a typical threshold fluence of 10 µJ/cm^2. These exceptionally low values are typically achieved near the material's band gap and increase with shorter periods (corresponding to shorter wavelengths). Spectra for various fluences of CdS and CdSe are displayed in Figure 1c and 1d, revealing lasing at 519 nm and around 670 nm, respectively. Figure 1e and 1f showcase how altering the period can change the resonance wavelength, indicating a broad gain bandwidth of 50 nm (480 – 530 nm) for CdS and 90 nm (650 – 740 nm) for CdSe. Looking at Figure 1d in particular, one can see a distinct multimode behaviour with three peaks. To understand this, we have to investigate the photonic crystal.

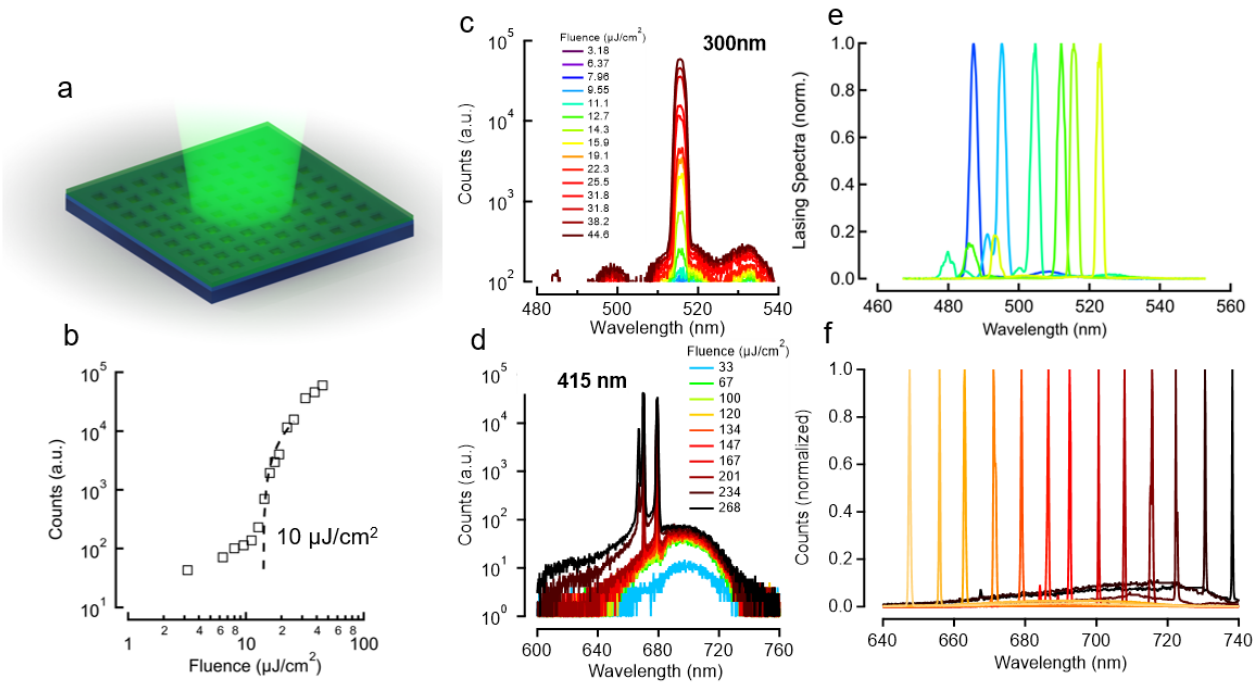

Figure 1. Lasing from BNCs. (a) Schematic of PCSEL device, overcoated with CdS BNCs. (b) Typical $P_{in} - P_{out}$ behaviour of PCSEL device, here with a period of 300 nm and lasing around 517 nm. (c) Spectra of previously mentioned device. (d) PCSEL overcoated with CdSe BNCs, showing clear multimode lasing with three distinct peaks. The period here is 415 nm. (e) Lasing of five different devices on the same chip, covered with CdS BNCs, showing a lasing bandwidth of about 50 nm. (f) Same as before but with CdSe BNCs, showing a lasing bandwidth of almost 100 nm.

3. PHOTONIC CRYSTAL SURFACE EMITTING LASERS

Lasing within these cavities is determined by second-order Bragg diffraction, expressed as $\lambda_{res} = n_{eff} \cdot \Lambda$. Here, λ_{res} represents the resonance wavelength, n_{eff} denotes the effective refractive index, and Λ is the grating period. Due to the second order grating, scattering can occur upward (first order scattering), or backward (second order scattering). Various modes exist in these types of systems, which each have a slightly different n_{eff}, due to the specific mode configuration. In such systems, various modes exist, each possessing a slightly different n_{eff} based on its specific mode configuration. To delve deeper, examining the photonic band structure is valuable, particularly around the Γ-point where second order diffraction occurs (Figure 2a). This analysis reveals eight distinct photonic bands, resulting in six modes that can lase at the Γ-point due to degeneracy of some of the modes. These six modes can be categorized into three Transverse-Electric

(TE) and three Transverse-Magnetic (TM) resonances, discerned by in-plane field distribution. TE modes exhibit only in-plane electrical field components and out-of-plane magnetic field components, while TM modes exhibit the reverse. These modes further classify based on in-plane symmetry, drawing from group theory terminology. A/B modes feature anti-symmetrical in-plane fields with respect to the x- and y-axis, while the E mode is symmetric (degenerate). The symmetry significantly influences mode confinement; anti-symmetric fields restrict upward scattering for A/B modes, confining light effectively within the plane. In an ideal infinite device, the Q-factor is infinite, but real-world constraints such as device finiteness and scattering losses reduce it. For E-modes, symmetry results in an equal probability of first and second order scattering, yielding a low Q-factor cavity. Electric and magnetic fields for A and E modes are depicted in Figure 2b and 2c. The B-mode closely resembles the A mode.

Far-field analysis in Figure 2d reveals that the anti-symmetric mode profile of A/B modes leads to emission at a small angle, creating a ring-shaped far field. The angular distribution depends on the device's period count. By employing a polarizer, the optical mode's nature (TE or TM) can be determined—TE modes exhibit azimuthal polarization, while TM modes have radial polarization. As can be seen from the bottom row of Figure 2d, the measured far field spectrum closely corresponds to the simulated far field, as do the lobes shown for horizontal and vertical polarisation, demonstrating that this particular mode has TE polarisation.

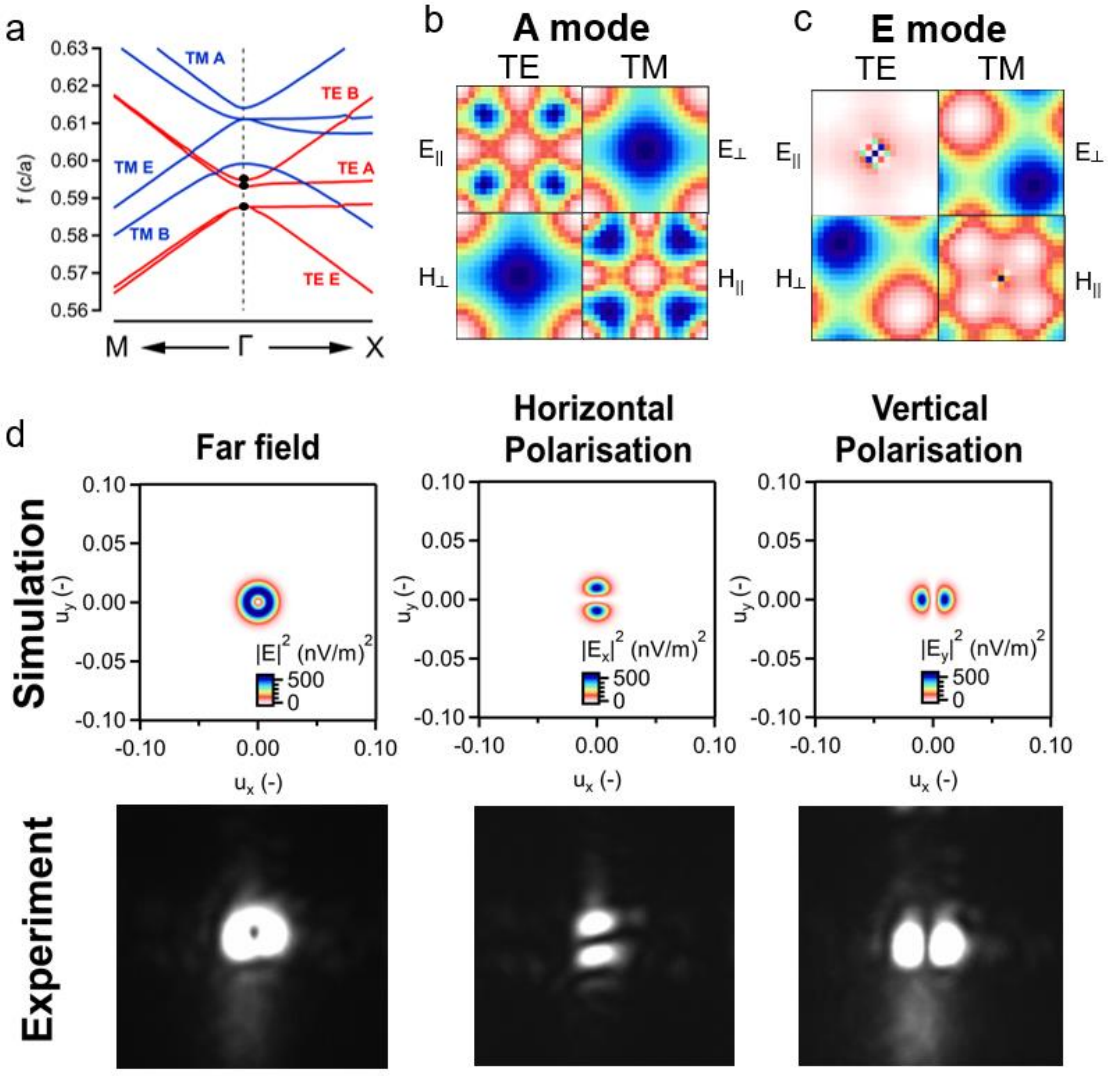

Figure 2. Modes in PCSEL lasers. (a) Photonic band diagram around the Γ-point. Red bands are TE bands, blue are TM. Names are denoted near the bands. (b) Electric and magnetic fields for the TE and TM version of mode A. (c) Same for

mode E. (d) Simulated and measured far field patterns for a mode, indicating it is the TE A mode through polarised measurements.

4. CONCLUSIONS

This work demonstrates lasing within a PCSEL architecture, and explains the lasing based on photonic crystal band structure. It furthermore demonstrates how to catalogue modes in simulation, and how they can be linked to the experiment. The low threshold and broad lasing bandwidth found indicate the usefulness of this hybrid material approach, and suggest the usefulness of using BNCs in these types of cavities in the future.

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