Self-assembled semiconductor microlaser based on colloidal nanoplatelets

Pedro Urbano Alves*, Manoj Sharma†‡, Emek Goksu Durmusoglu‡, Merve Ïzmir‡, Martin D. Dawson*, Hilmi Volkan Demir‡ and Nicolas Laurand*
*Institute of Photonics, Department of Physics, SUPA, University of Strathclyde, Glasgow, UK
†Exciton Science, Chemistry Building, University of Monash, Melbourne, Australia
‡Luminous!, School of Electrical and Electronic Engineering, Nanyang Technological University, Singapore
pedro.alves@strath.ac.uk

***Abstract* — A semiconductor microsphere laser based on colloidal nanoplatelets is demonstrated comprising a micron-sized supraparticle obtained by self-assembly of core/shell CdSe/ CdS nanoplatelets with peak luminescence at 660nm. It shows multimode laser emission between 665 and 695nm with threshold at 200 nJ (28 ± 17 mJ.cm^{-2}).**

Keywords — Nanoplatelets, lasers, nanocrystals, semiconductors, microresonators.

I. Introduction

Colloidal nanoplatelets (NPLs), or colloidal quantum wells, are 2D semiconductor nanocrystals (NCs) that share many attributes with other colloidal NCs, including the flexibility of solution processing, composition and size control of their optical and electronic properties, and high luminescence quantum efficiency. In addition, they benefit from narrower emission linewidths, higher oscillator strengths, larger exciton binding energies, higher absorption cross-sections and lower Auger recombination rate when compared to colloidal quantum dots (QDs) [1]. For these reasons, CdSe-based NPLs are particularly attractive for visible colloidal lasers [2]. They are also promising for Förster resonance energy transfer between them and other NCs such as QDs [3]. Up to now and to the authors' knowledge, demonstrations of NPL lasers have been obtained by combining NPLs with a laser cavity fabricated separately through top-down methods [4]. In this work, the bottom-up self-assembly of NPLs into supraparticles (SPs) – in this case microspheres – is reported. This follows a similar method used for QDs, where SPs act both as the laser gain medium and the optical cavity [5]. While this is 'a priori' challenging due to the different geometries between QDs and NPLs, CdSe/ CdS NPL-based SP microspheres capable of laser emission are synthesized and demonstrated for the first time.

II. Materials and Methodology

A. Synthesis of the Supraparticles

An oil-in-water emulsion technique [5] at cold temperatures (close to 0ºC) was used to synthesize spherical SPs with NPLs as building blocks. The NPLs were synthesized as in [6] and were approximately 14 × 15 nm in width × length. They consisted of a CdSe core 4 monolayers thick and a CdS shell approximately 8 monolayer thick synthesized by the hot-injection shell growth method. The emission peak of individual NPLs was centered at 660 nm. Resulting SPs in solution were drop-cast onto a glass substrate and optically-pumped individually after complete solvent evaporation.

B. Optical Characterization

A single SP was selected and optically pumped on a μ-photoluminescence (μ-PL) setup with a 5 ns pulsed laser (λ = 355 nm) at a repetition rate of 10 Hz and with a beam spot radius of approximately 18 ± 6 μm. The beam intensity was controlled by a variable wheel neutral density attenuator and focused on the sample with an objective lens (10×/ 0.25). A spectrometer (AvaSpec-2048-4-DT with 0.7 nm spectral resolution between 220-1100 nm, Avantes) was used to acquire the spectral data [5]. A microscope was used to measure the size of the SPs (Fig. 1). The size was verified by the pseudo-Free Spectral Range (FSR) of their cavities, which can be expressed as the wavelength spacing between two consecutive modes of the same polarization, $\Delta\nu_{n,l}^{\Delta l} \approx \frac{c}{2\pi.N.a}$, with the radius a of a sphere of refractive index N [7]. The 12 μm of diameter measured on the microscopic image (Fig. 1) matches the size extracted from the FSR in Fig. 2 ($\frac{\Delta\nu}{c} \approx 6.5$ nm) for $N \approx 1.8$.

III. Results and Discussion

The microscope images and emission spectra of the measured NPL SP below threshold (normalized PL) and at 1.8 μJ, i.e. above lasing threshold, are shown in Fig.1 and 2, respectively. The spacing between grey lines in the background (6.5 nm) corresponds to the pseudo FSR for this SP as it matches the spacing between whispering gallery mode adjacent maxima (normalized PL). The lasing peaks at approximately 690 and 696 nm also match that same distance, suggesting that they correspond to consecutive modes.

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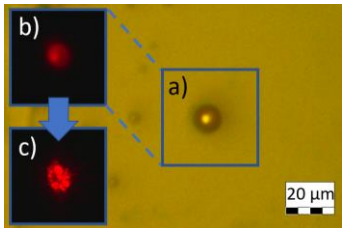

Figure 1. Microscope image of the SP under bright light (a) and pumped below (b) and above (c) laser threshold (diameter: 12 µm).

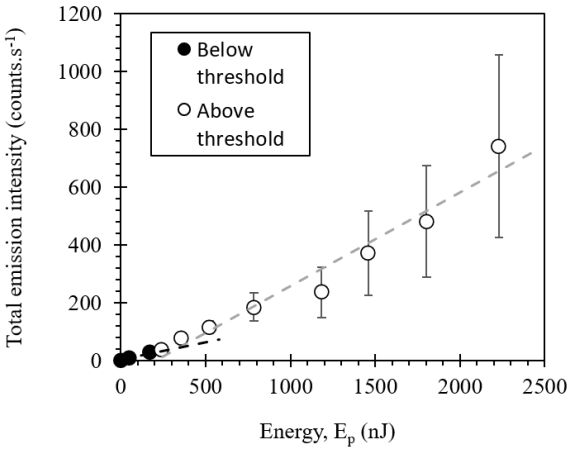

Figure 3. The 3 main laser emission peak intensities (Fig. 2) and the laser transfer function of the SP as their sum, with data points and trends below (black) and above (white) threshold.

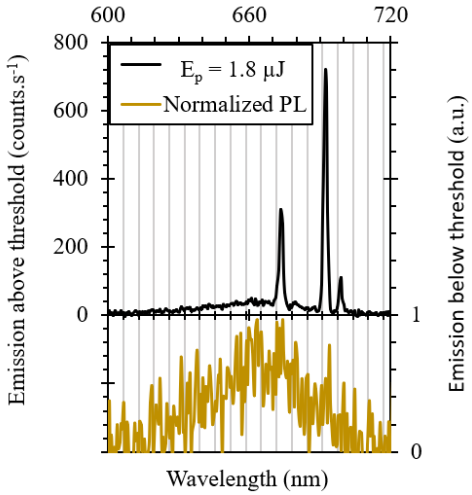

Figure 2. Normalized PL of the characterized SP (bottom) and an emission spectrum above threshold, for E_p = 1.8 µJ (top).

The total emission intensity of the measured NPL SP (Fig. 3) is given by the sum of the emission of the 3 main laser peaks (673, 693 and 699 nm) at different pump energies. The lasing threshold for this SP is at 200 nJ (28 ± 17 mJ.cm^{-2}) and the SP was able to operate at the full range of fluences while showing little to no degradation. Laser emission starts almost simultaneously at 673 and 693 nm. Above 1 µJ, the mode at 699 nm appears and it becomes a competition between these 3 main lasing peaks.

IV. Conclusion

Synthesis of individual self-assembled NPL SPs for lasing applications has been successfully demonstrated. Future work will focus on further exploring the properties of NPL SPs.

Acknowledgments

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