Multicolor laser oscillation in a single self-assembled colloidal quantum dot microsphere

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***Abstract* — Self-assembled microsphere lasers oscillating simultaneously at more than one wavelength in the visible are reported. The lasers consist of micron-scale supraparticles made of CdS_xSe_{1-x}/ ZnS quantum dots that emit between 585-605 nm and 625-655 nm.**

Keywords* — *Quantum dots, lasers, nanocrystals, semiconductors, microresonators.

I. Introduction

The advantageous characteristics of colloidal quantum dots (CQDs), including solution processability, near-unity emission quantum yield and tunable emission wavelength, make them attractive for photonic applications. Specifically in terms of laser operation, they have potential in integrated photonics, imaging, sensing and diagnostics [1]. To achieve lasing with CQDs, different types of cavity designs can be used, such as: Fabry-Perot, spherical and micro-ring resonators, vertical cavities implemented with distributed Bragg reflectors, and DFB gratings [1]. While single color laser oscillation has been demonstrated with CQDs, to our knowledge no reports regarding multicolor laser oscillation have been made to date. Such multicolor lasers would pave the way for a broad range of applications including miniature tunable lasers, high density color displays and sensing devices [2]. In this work, advantage of the whispering gallery modes present in spherical supraparticles (SPs) - self-assembled microspheres of CQDs - is taken to demonstrate multicolor lasing in individual SPs.

II. Materials and Methodology

A. *Synthesis of the Supra Particles*

An oil-in-water emulsion technique [3] was used to synthesize spherical SPs made of CdS_xSe_{1-x}/ ZnS quantum dots. Three different batches were produced: red SPs made of CQDs emitting at ~630 nm; yellow SPs made of CQDs emitting at ~575 nm and SPs with a 50:50 ratio of red and yellow CQDs. Resulting SPs in water were then drop-cast onto a glass substrate and optically-pumped individually for the characterization process. The size dispersion of the synthesised SPs ranged approximately between 1 and 10 µm in diameter (Fig. 1).

B. *Optical Characterization*

SPs were selected and optically pumped one at a time with a 1.6 ns pulsed frequency-doubled Nd:YAG laser at 532 nm, at a repetition rate of 10 kHz, with a beam spot area of approximately 1.9×10^{-5} cm^2. The beam was attenuated with a variable wheel neutral density attenuator and focused on the sample with an objective lens (4×/ 0.13, Nikon). A spectrometer (AvaSpec-2048-4-DT, Avantes) was used to acquire the spectrum data. The µPhotoluminescence (µPL) setup described above follows the same design as in previous works [4]. The size of the SPs was measured by both optical microscopy and scanning electron microscopy (SEM).

III. Results and Discussion

Two batches of SPs emitting in a single color (red, yellow) were fabricated and characterized, before demonstrating emission at two colors (red and yellow) simultaneously. Fig. 2. and Fig. 3. show the typical emission intensity responses of a red SP laser (630 nm) and a yellow SP laser (575 nm) as a function of the pump fluence, as well as their characteristic photoluminescence below and above threshold. For both single color lasers, the threshold fluence was between 0.5 and 0.7 mJ.cm^{-2}. To test if multicolor laser oscillation could be obtained with a single SP, the third batch of SPs was prepared with 50 % of red CQDs and 50 % of yellow CQDs (Fig. 4). Depending on the pump fluence, three laser regimes were observed. In the first regime, the first modes to go above the lasing threshold were in the red band and this happened approximately at the same fluence as their single wavelength counterparts (0.5 to 0.7 mJ.cm^{-2}). For the second regime, at around

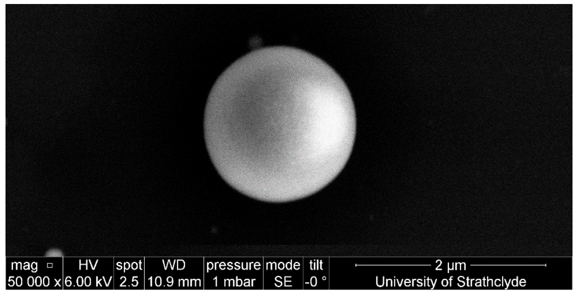

Fig. 1. SEM image of a red SP approximately 1.5 µm in diameter (representative example).

2.0 mJ.cm^{-2}, modes in the yellow band started to oscillate while the modes in the red band started to lose energy. Stable dual color lasing was obtained at approximately 2.4 mJ.cm^{-2} (Fig. 4). The third regime, at higher fluences, had the modes in the yellow band dominating the emission spectrum whilst modes in the red returned to their below threshold state.

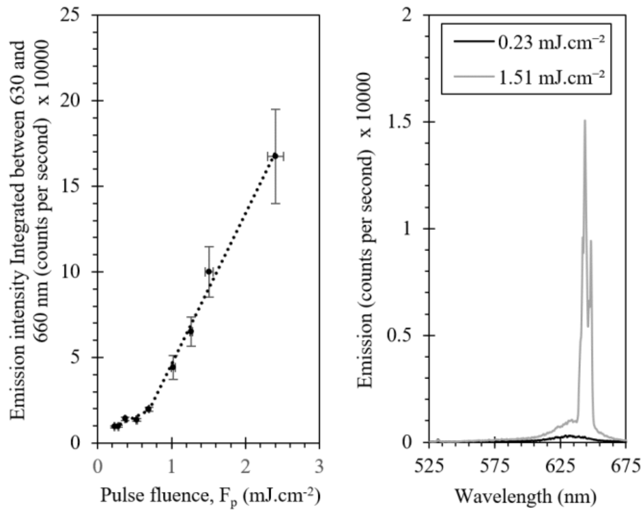

Fig. 2. Laser transfer function of a red SP (diameter: 7.6 µm) and its typical spectrum below and above threshold.

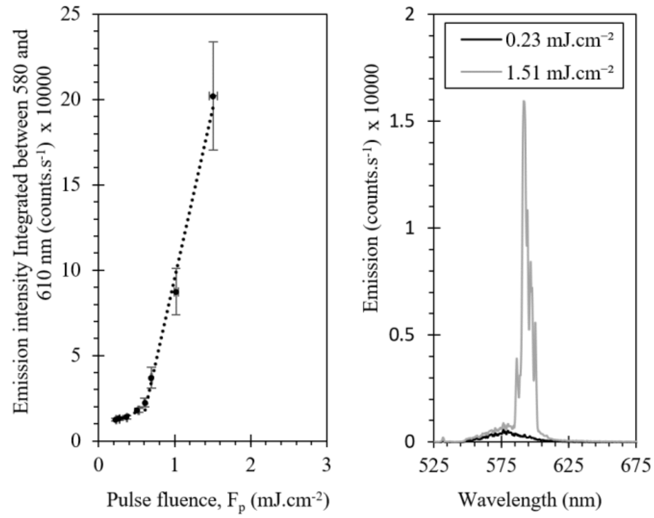

Fig. 3. Laser transfer function of a yellow SP (diameter: 6.8 µm) and its typical spectrum below and above threshold.

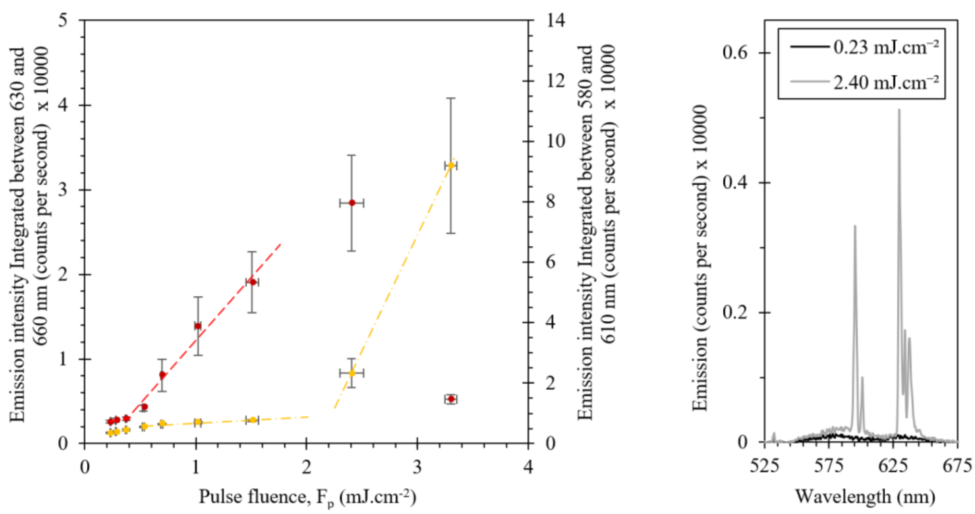

Fig. 4. Laser transfer function of a multicolor SP (diameter: 6.4 µm) and its typical spectrum below and above threshold.

IV. Conclusions

Multicolor lasing in individual self-assembled CQD SPs has been demonstrated. Three lasing regimes were successfully demonstrated in a single SP, as a function of the pump fluence (namely single color red, single color yellow and simultaneous dual color red and yellow). Future work will focus on studying these properties further and demonstrating possible applications.

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