Silica Coated Colloidal Semiconductor Quantum Dot Supracrystal Microlasers

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A novel approach for synthesizing silica-coated colloidal quantum dot supracrystal microsphere lasers is demonstrated. These lasers consist of red-emitting CdS_xSe_{1-x}/ZnS quantum dots that act as both the laser medium and the cavity, and have great potential for biosensing, bioimaging, and integrated photonics.

The bottom-up self-assembly of colloidal quantum dots into supraparticles (SPs) from an emulsion has been shown to be an attractive and simple method for creating microsize whispering gallery mode (WGM) lasers [1,2]. WGM lasers are excellent sensors, able to detect minute changes in their local refractive index. However, WGM SPs are not water soluble due to oleate molecules on their surface preventing their use in biosensing experiments. Herein, we present a method to coat the SPs with a silica shell, which not only allows for water solubility, but also acts as a platform for further biofunctionalisation.

The SPs were synthesized using an oil-in-water emulsion technique [1,2], yielding water insoluble SPs coated with oleate molecules. The silica shell was then grown onto the surface in a 2-step process [3]. Firstly, the oleate molecules were replaced with polyvinylpyrrolidone which allows for water solubility. Secondly, the SPs were mixed in a solution of ammonia and tetraethyl orthosilicate resulting in a thin (<5 nm) silica coating. The presence of a silica shell was confirmed through UV-Vis and energy dispersive X-ray spectrometry. The size of the SPs ranged between 500 nm – 15 µm as measured by optical microscopy and scanning electron microscopy (SEM) – an example showing two spheres is displayed in Fig 1(a). SPs were optically pumped with a 355nm, 5ns pulsed Nd:YAG laser at a 10Hz repetition rate and with a beam spot area 2.6 ± 1.5 x 10^{-5} cm^2. Oleate- and silica-coated SPs of ~10µm diameter exhibit lasing upon excitation with thresholds of 1.2 µJ and 6.1 µJ respectively. Fig. 1(b) shows the emission spectra of the silica coated SPs above (lasing at 605 nm) and below threshold for a single sphere. Despite a threshold increase, the silica coating broadens the applications for these microlasers.

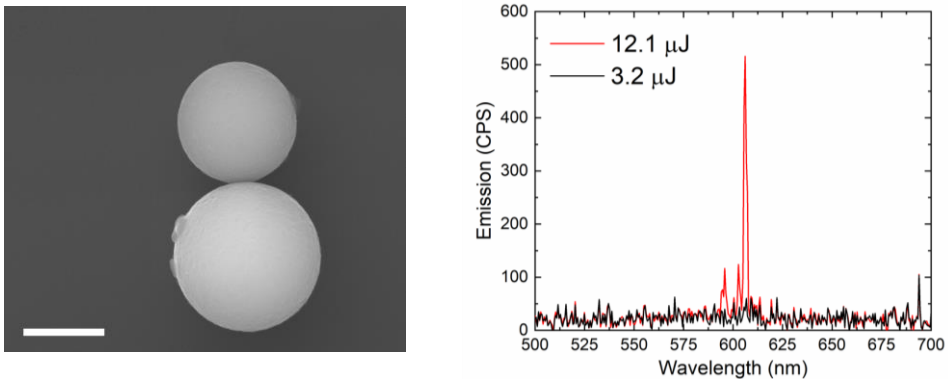

Fig. 1 (a) SEM image of silica coated SPs, scale bar is 1 µm; (b) Lasing spectrum of a single silica coated SP with a diameter of 10.4 µm, above and below threshold at 12.1 µJ and 3.2 µJ respectively.

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