Using Colloidal Quantum Shells as a Novel Gain Medium for Integrated Visible Lasers

I. Tanghe,[1,2,3] A. Waters,[4] J. Huang,[4] K. Molkens,[1,2,3] T. Vandekerckhove,[1] D. Van Thourhout,[1] M. Zamkov,[4,5] and P. Geiregat[2,3].

[1] Photonics Research Group, Ghent University, Belgium
[2] Physics and Chemistry of Nanostructures, Ghent University, Belgium
[3] NoLIMITS Center for Non-Linear Microscopy and Spectroscopy, Ghent University, Belgium
[4] Department of Physics, Bowling Green State University, Bowling Green, Ohio, USA
[5] The Center of Photochemical Sciences, Bowling Green State University, Bowling Green, Ohio, USA

Colloidal Quantum Shells (QSs) are a class of novel solution-processible nanocrystals. In this work, their potential for optical gain is investigated and lasing with optical excitation is demonstrated.

Introduction

Colloidal Quantum Shells (QSs) based on CdSe/CdS, are investigated as a promising new solution-processible gain material[1,2]. This material consists of multiple shells, where contrary to typical QDs, the material possessing the smallest band gap (CdSe) is located not within the core, but rather in the initial shell, which has a thickness of 1.9 nm. The core and the outer shell consist of a material with a larger band gap (CdS). The core is 8.5 nm, and the secondary CdS shell with ZnS passivation is 21-22 nm, found through a TEM analysis (Figure 1b). The resulting material emits around 660 nm, with a Quantum Yield close to 100%. It is very similar to the material used in the work of Harankahage et al.[2]

Transient Absorption Spectroscopy

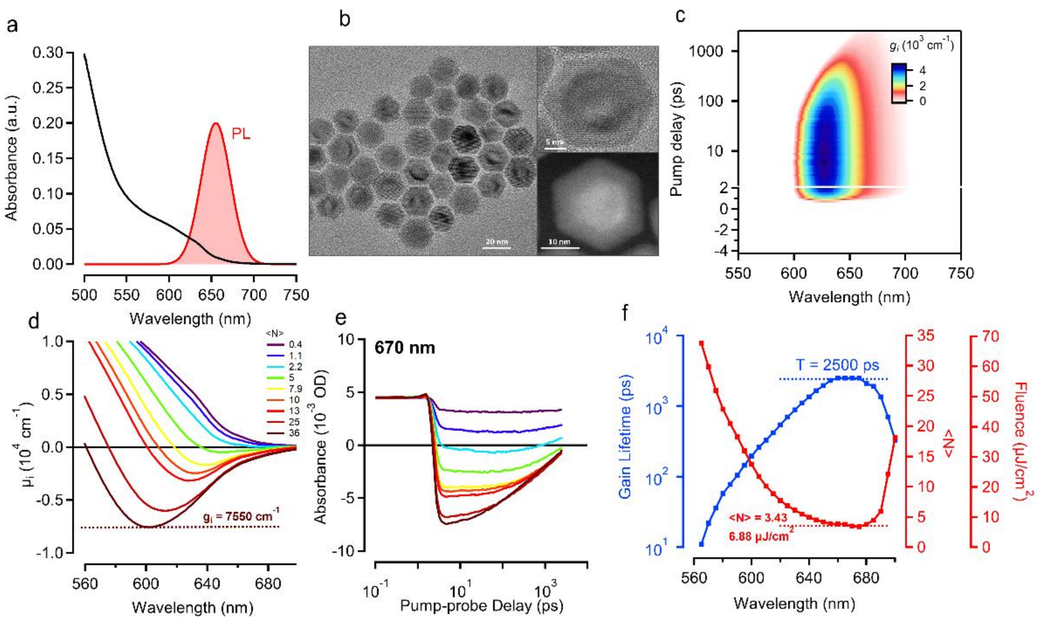

Figure 1: Linear characterization and Transient Absorption Spectroscopy of Colloidal Quantum Shells.

We studied the optical gain by means of femtosecond Transient Absorption (TA), where the pump consists of 110 fs pulses at a wavelength of 515 nm. After the initial pump pulse, a second short but broadband probe pulse is injected into the sample. From this measurement, gain maps can be constructed as exemplified in Figure 1c. By varying the excitation power, multiple of these maps can be measured to find the gain threshold.

Taking slices for various excitation powers yield both a spectrum (Figure 1d) and kinetics of the gain (Figure 1e). This can give us the gain magnitude and gain lifetime, yielding values of up to 7500 and 2.5 ns, respectively. Finally, in Figure 1f, the found gain lifetime (blue), and the threshold (red) are plotted together, showing the inversely proportional relationship between these two parameters.

Demonstrating Lasing with Photonic Crystal based Lasers

The resonating wavelength within a cavity follows Bragg's Law, where a second order grating was used to achieve surface emission. In this study, gratings were designed with a square lattice, featuring periods spanning from 320 to 460 nm at 5 nm intervals. This range was determined based on an estimate of the effective index.

Laterally, the devices are 600x600 periods. A schematic of a device is shown in Figure 2a. They are designed with a duty cycle of 60%. SEM images in Figure 2b,c show a device with a period of 395 nm.

A 515 nm fs laser is used to optically excite the cavities. An example of a measurement is illustrated in Figure 2b, for a period of 385 nm which shows lasing at 637.5 nm. The inset graph depicts peak counts as a function of fluency, revealing distinct threshold behavior, with a measured threshold of 35.8 µJ/cm^2. Devices exhibited lasing across periods ranging from 335 nm to 405 nm, corresponding to resonant wavelengths spanning 580 to 660 nm, as shown in Figure 2e. Figure 2f illustrates device thresholds in red, revealing the lowest threshold within the range of 640-660 nm, measured at 15.9 µJ/cm^2.

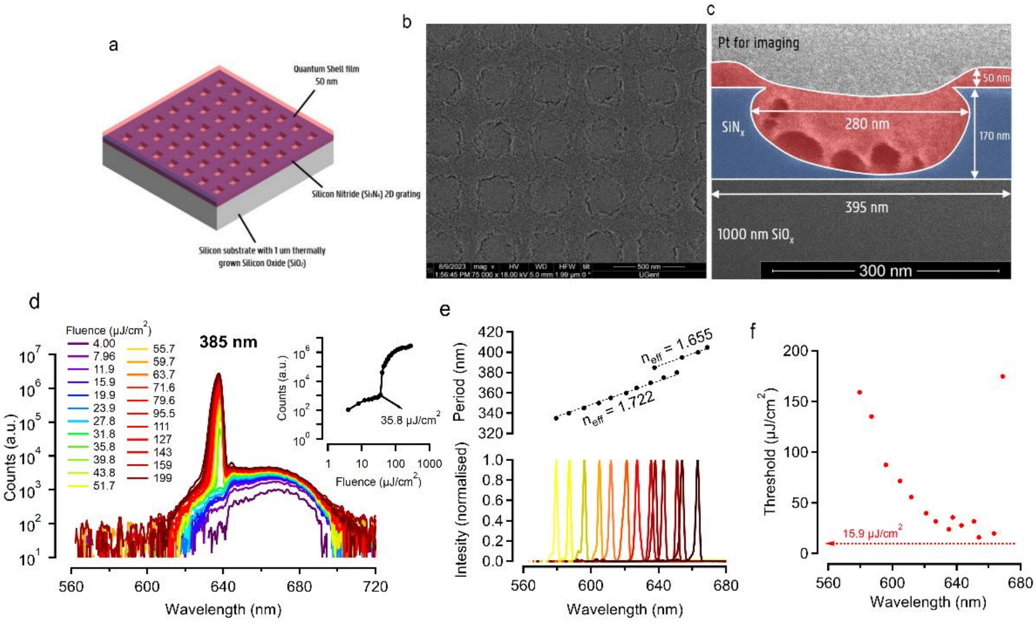

Figure 2: Lasing with Colloidal Quantum Shells through Photonic Crystal Surface Emitting Lasers

Conclusions

In this study the strong gain properties of colloidal QSs are determined, and low threshold femtosecond lasing is demonstrated.

References

[1] Cassidy, J.; …, Zamkov, M. Quantum Shells Boost the Optical Gain of Lasing Media. ACS Nano 2022, 16, 3017–3026.

[2] Harankahage, D.; …, Zamkov, M. Quantum Shell in a Shell: Engineering Colloidal Nanocrystals for a High-Intensity Excitation Regime. Journal of the American Chemical Society 2023.