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Low-threshold optical gain and lasing of colloidal nanoplatelets

Yusuf Kelestemur,[1] Burak Guzelturk,[1] Murat Olutas,[1,2] Savas Delikanli,[1] and Hilmi Volkan Demir[1,3]
[1]Department of Electrical and Electronics Engineering, Department of Physics, UNAM - Institute of Materials Science and Nanotechnology, Bilkent University, Ankara 06800 Turkey; [2]Department of Physics, Abant Izzet Baysal University, Bolu 14280, Turkey
[3]School of Electrical and Electronic Engineering, School of Physical and Mathematical Sciences, Nanyang Technological University, Nanyang Avenue, Singapore 639798, Singapore
e-mail: volkan@stanfordalumni.org

Semiconductor nanocrystals, which are also known as colloidal quantum dots (CQDs), are highly attractive materials for high performance optoelectronic device applications such as lasers. With their size, shape and composition tunable electronic structure and optical properties, CQDs are highly desired for achieving full-color, temperature-insensitive, low-threshold and solution-processed lasers [1, 2]. However, due to their small size, they suffer from the nonradiative multiexciton Auger Recombination (AR), where energy of a bound electron-hole pair is transferred to a third particle of either an electron or a hole instead of radiative recombination. Therefore, CQDs having suppressed AR are strongly required for achieving high quality CQD-based lasers. To address this issue, CQDs having different size, shape and electronic structure have been synthesized and studied extensively [3-5]. Generally, suppression of AR and lower optical gain thresholds are achieved via reducing the wavefunction overlap of the electron and hole in a CQD. However, the separation of the electron and hole wavefunctions will dramatically decrease the oscillator strength and optical gain coefficient, which is highly critical for achieving high performance lasers. Therefore, colloidal materials with suppressed AR and high gain coefficients are highly welcomed. Here, we study optical gain performance of colloidal quantum wells [6] of CdSe-core and CdSe/CdS core/crown nanoplatelets (NPLs) that demonstrate remarkable optical properties with ultra-low threshold one- and two-photon optical pumping. As a result of their giant oscillator strength, superior optical gain and lasing performance are achieved from these colloidal NPLs with greatly enhanced gain coefficient [7].

In this study, we synthesize CdSe-core and CdSe/CdS core/crown NPLs with varying crown size by using the slightly modified recipe [8]. First, we synthesize 4 monolayer (ML) thick CdSe-core NPLs having peak spontaneous emission at 513 nm. Second, by using freshly synthesized 4 ML CdSe NPLs as seeds, CdS-crown layer is growth with the simultaneous injection of cadmium and sulfur precursors. By simply changing the amount of shell precursor, the size of CdS-crown layer can be tuned. The High-angle annular dark-field transmission electron microscopy (HAADF-TEM) images of CdSe-core and CdSe/CdS core/crown NPLs are depicted in the Figure 1. As it can be seen from the figure, CdSe/CdS NPLs having different CdS-crown size are achieved with uniform and homogenous coating by using different amount of shell precursors. According to long axis length, CdSe/CdS core/crown NPLs are denoted as 21, 25 and 32 nm, respectively. In addition, due to lateral growth of CdS-crown layer, the thickness of CdSe/CdS core/crown NPLs is remained the same as the thickness of the CdSe-core NPLs.

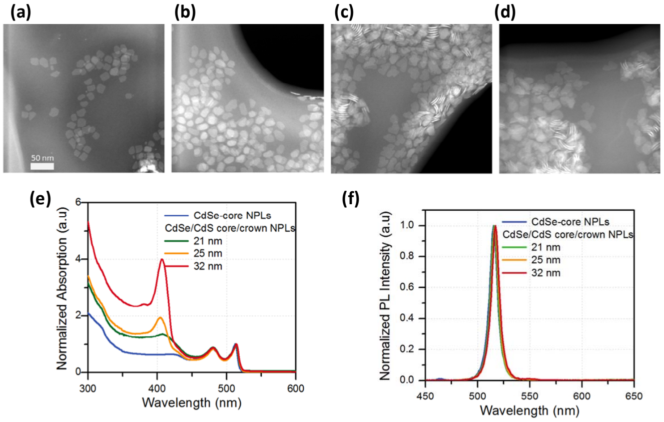

Figure 1. High-angle annular dark-field transmission electron microscopy (HAADF-TEM) images of the (a) CdSe-core NPLs, (b) 21, (c) 25 and (d) 32 nm CdSe/CdS core/crown NPLs with the scale bar of 50 nm. (e) Absorption and (f) photoluminescence spectra of CdSe-core and CdSe/CdS core/crown NPLs [7].

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The photoluminescence and absorption spectra of CdSe-core and CdSe/CdS core/crown NPLs having different CdS-crown size are plotted in the Figure 1e-f. As it can be seen from the absorption spectra, with the coating of CdS-crown layer, a new peak around 407 nm emerges and becomes dominant with further increased size of the CdS-crown layer. This peak is attributed the formation of 4 ML CdS and it makes great contribution to enhance the absorption cross-section of CdSe/CdS core/crown NPLs. However, the emission spectrum of CdSe/CdS core/crown NPLs is remained almost the same as compared to the only core NPLs. Although there is a strong absorption and exciton formation observed in the CdS-crown region, no emission is observed from the CdS-crown states, which confirms the efficient inter-exciton funneling from the CdS-crown to the CdSe-core [8]. It is also verified from the photoluminescence excitation (PLE) spectra.

After complete optical and structural characterization of the CdSe-core and CdSe/CdS core/crown NPLs, we study the amplified spontaneous emission with one- and two-photon optical pumping. For the optical gain study, highly close packed and dense films of NPLs are prepared with the drop casting of highly concentrated solution on the bare quartz substrates. The samples were pumped with 120 fs laser pulses with 1 kHz repetition rate at the optical wavelength of 800 nm. In the Figure 2, photoluminescence spectra of CdSe/CdS core/crown NPLs (21 nm) under intense two-photon optical pumping is given. When the excitation intensity exceeds the optical gain threshold of 5.81 mJ/cm^2, amplified spontaneous peak (ASE) emerges and dominates the whole emission spectra at higher excitation intensities with ~12 nm red-shifted ASE peak having a full-width-half-maximum (FWHM) of 5 nm. With the further increased CdS-crown region, the optical gain threshold is reduced to 4.48 mJ/cm^2. It can be attributed to the increased absorption cross-section and efficient inter-exciton funneling. In addition, we also investigate optical gain performance with one-photon optical pumping (1PA). While CdSe-core NPLs exhibits optical gain threshold of 214 μJ/cm^2, lower optical gain threshold (~41 μJ/cm^2) are achieved from CdSe/CdS core/crown NPLs. When compared to CQDs having emission in the same spectral range, CdSe/CdS core/crown NPLs exhibits the lowest optical gain threshold values for both 1PA and 2PA reported to date. In addition, the gain coefficient of NPLs is measured to be as high as 650 cm^{-1}. Finally, by using solution processed distributed Bragg reflectors (DBR), which are made from SiO_2 and TiO_2 nanoparticles, we also demonstrate proof of concept laser of the CdSe/CdS core/crown NPLs with 2PA lasing threshold of 2.49 mJ/cm^2.

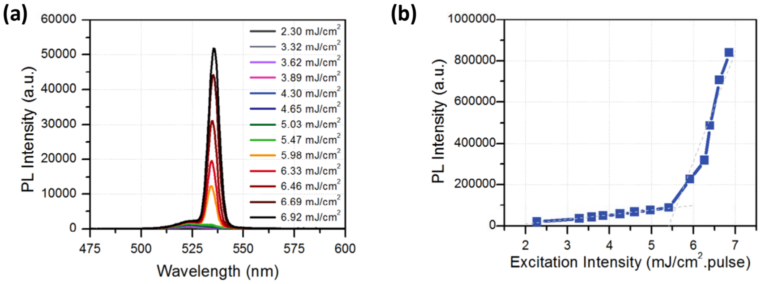

Figure 2. (a) Evolution of the PL spectra of CdSe/CdS core/crown NPLs (21 nm) under two photon optical pumping with varying excitation intensity. (b) Excitation pulse intensity dependence of the PL intensity at the wavelength of the ASE process. The linear fits shown as dashed lines are guides to the eye about the slopes of the curves [7].

In conclusion, we showed that CdSe/CdS core/crown NPLs are good candidates for lasing applications with both one- and two-photon optical pumping. As a result of the enhanced absorption cross-section and efficient inter exciton funneling, the lowest optical gain threshold of 41 μJ/cm^2 and 4.48 mJ/cm^2 are achieved from the CdSe/CdS core/crown NPLs when compared to the CQDs emitting in the green spectral emission region with 1PA and 2PA respectively. In addition, they exhibit the highest gain coefficient (~650 cm^{-1}) when compared to colloidal quantum dots and rods. Finally, 2PA lasing threshold of 2.49 mJ/cm^2 is achieved from these core/crown NPLs, which is record for the green emitting CQDs. These results show that the NPLs are promising materials for high performance lasing applications with suppressed Auger recombination and high gain coefficients.

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