Supporting Information

Thickness-Tunable Self-Assembled Colloidal Nanoplatelet Films Enable Ultra-Thin Optical Gain Media

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Synthesis of $CdSe/Cd_{0.25}Zn_{0.75}S$ core/shell NPLs. CdSe/CdZnS core/alloyed-shell NPLs have been synthesized with some modifications by using the hot-injection shell synthesis recipe reported in our recent works.[1,2] Cd-acetate (17.25 mg), Zn-acetate (41.25 mg), 1-octadecene (ODE, 7 mL), oleic acid (0.70 mL) and 4.5 ML CdSe core in hexane (1 mL, having absorbance of 1.5 at 350 nm along 1 cm-long optical path) have been loaded in quartz flask (50 mL) and stirred for 90 min at room temperature, for 30 min at 85 °C under vacuum environment. Then, oleylamine (0.7 mL) has been injected into the flask at 90 °C under argon gas and the temperature of the solution has been increased to 300 °C. The injection of ODE-octanethiol mixture that is prepared in glovebox using ODE (4 mL) and octanethiol (105 µL) have been started at 165 °C using 10 mL h^{-1} rate of syringe pump until the temperature reached 280 °C, and then the rate has been decreased to 4 mL h^{-1}. Before cooling to room temperature with cold water, the solution is kept for an hour at 300°C. Hexane (5 mL) is added at room temperature and the solution is cleaned four times with absolute ethanol. Finally, the precipitated NPLs is dissolved in hexane.

Surface treatment of the substrates prior to self-assembly deposition. For the cleaning before the silanization, the substrates have been kept in piranha solution for 15 min, followed by sonication in chemical detergent (Hellmanex, diluted 500 times with deionized water), deionized water, acetone and isopropanol, each for 5 min. The substrates were dried under nitrogen flow after sonication with isopropanol. For the silane treatment of the silicon and fused silica substrates, we adopted and modified the recipe of Beck *et al.*[3] Clean substrates were placed on a hot plate under nitrogen environment, which was then heated up to 200 °C. As the hot plate reached this targeted temperature, 20 µL of the silane solution was dropped onto a blank spot on the hot plate, which boils in a few seconds. The vapour is swiftly trapped by placing a glass beaker upside-down onto the hot plate. The hot plate is kept at 200 °C for 30 min. The substrates are immersed in hexane before being taken out into the atmosphere. The treated substrates are then sonicated for 1 min in acetone and then in isopropanol.

Optical gain measurements. Multilayered NPLs deposited on fused silica have been placed in front of a pulsed laser beam at normal incidence, whose intensity is adjusted with a neutral density filter on the optical path. A Ti:Sapphire laser amplifier (Spitfire Pro) generates laser pulses at 800 nm with a pulse width of ~110 fs and a repetition rate of 1 kHz. The wavelength of these pulses is converted to 400 nm with a frequency-doubling barium borate crystal. The residual 800 nm light is filtered out with a short pass filter. The 400 nm beam is focused along one dimension onto the sample with a cylindrical lens to obtain a stripe excitation having a width of ~150 μm. The emission of the sample is collected from the side with an optical fiber. The spectra are recorded with an optical spectrometer (Maya 2000, Ocean Optics).

Mode profile calculations. The modal analyses were carried out using a commercially available software package (Lumerical MODE solution) with two-dimensional layout. The refractive indices of CQW and fused silica used in the simulations were 2.10 and 1.45, respectively. The confinement factor was calculated based on the ratio of the fundamental transverse electric mode energy confined within the active NPL film to the total mode energy.

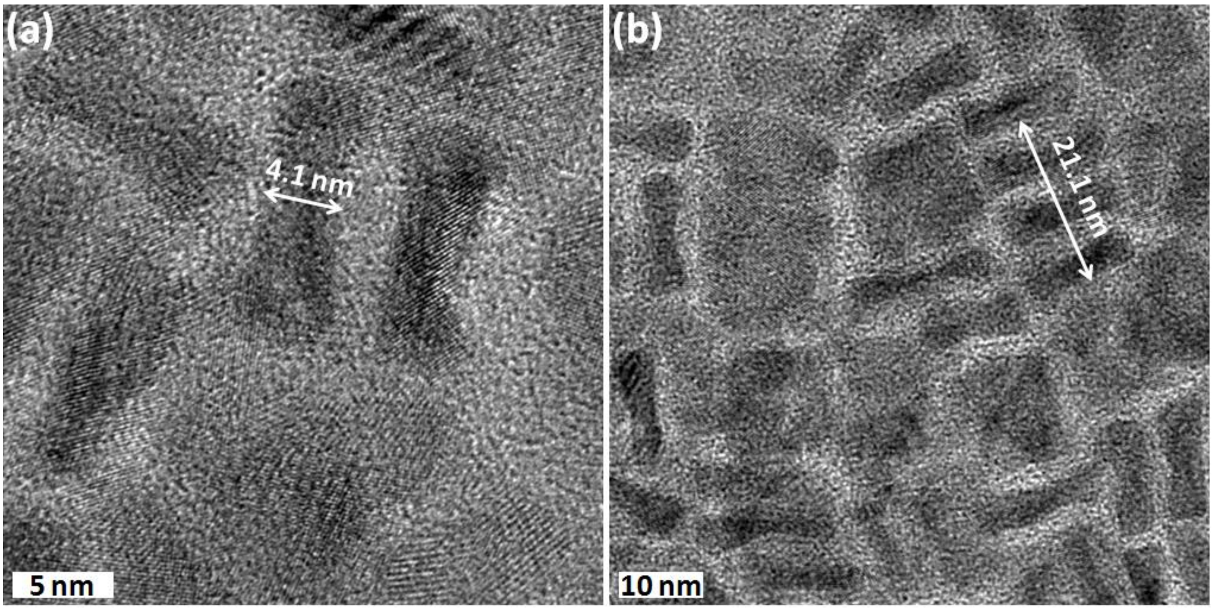

Figure S1: High-resolution transmission electron micrographs of CdSe/$Cd_{0.25}Zn_{0.75}S$ core/alloyed-shell NPLs that are vertically oriented. (a) The thickness of a single NPL is measured as ~4.1 nm. (b) Measurements on chains of vertically oriented NPLs reveal a center-to-center distance of ~7.0 nm, in accordance with the ellipsometric measurements (see Figure S4a).

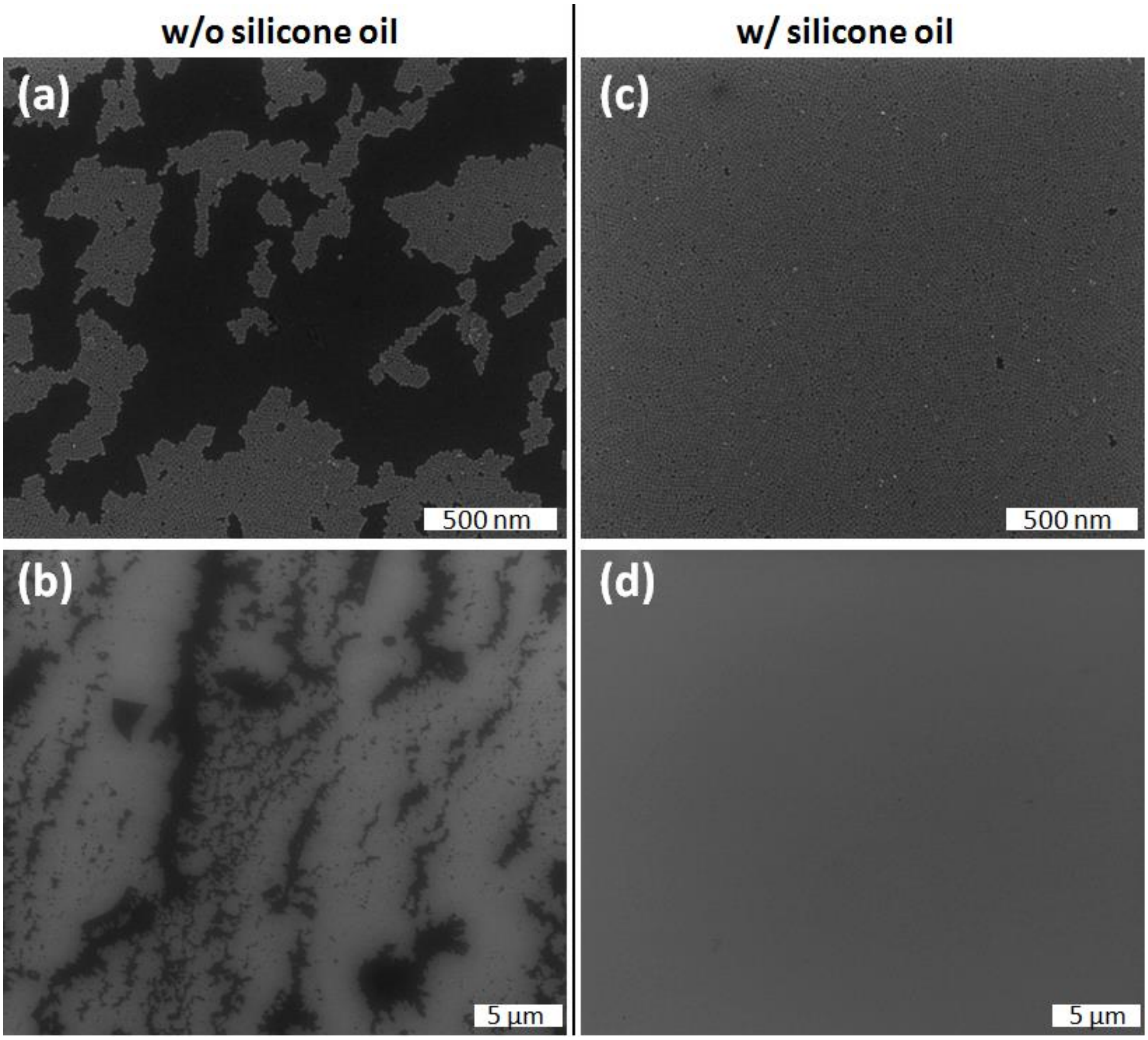

Figure S2: Scanning electron micrographs of self-assembled NPLs (a, b) without and (c, d) with silicone oil. In all panels, dark areas are void.

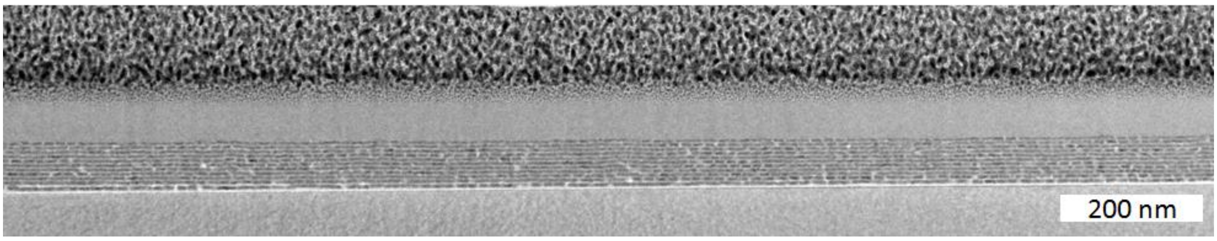

Figure S3: Cross-sectional transmission electron micrograph of 11-layered NPLs at lower magnification.

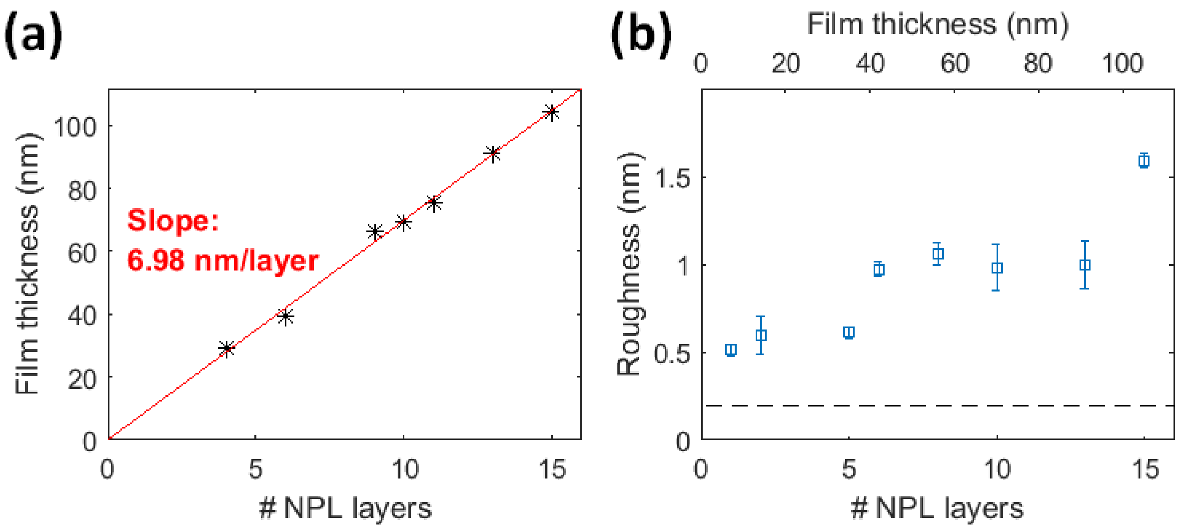

Figure S4: (a) Measurement of film thickness for the multilayered NPL constructs having different numbers of layers. The linear fit confirms 7.0 nm of thickness per NPL layer. (b) Surface roughness measurements of multilayer NPL films taken with atomic force microscopy over 5 different regions having an area of 2×2 μm^2. Dashed line shows the roughness of the bare fused silica substrate (~0.2 nm).

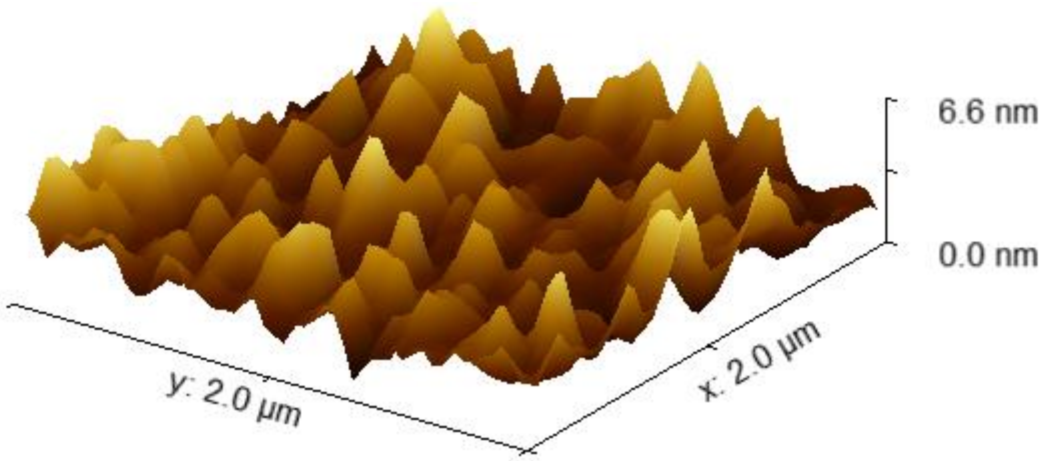

Figure S5: Topography of the surface of the 15-layered nanoplatelet film taken via atomic force microscopy.

Film homogeneity across large scale. To verify the uniformity of the film thickness across cm-large scales, we have taken ellipsometric measurements from different spots on a substrate with 1 × 2 cm^2 dimensions, which is deposited with 10 layers of our NPLs using our self-assembled deposition technique. The results of the ellipsometric fittings are given in Table S1.

Table S1: Ellipsometry measurements from different spots on a 10-layered sample

# Spot	Measured thickness (nm)	Refractive index
1	69.1	1.950
2	69.3	1.949
3	68.6	1.950
4	69.3	1.949
5	69.4	1.948

In addition, we have used confocal microscopy imaging to verify the homogeneity of the emission over larger scales. The confocal microscopy imaging taken from a 580 × 580 μm^2 region of the 14-layered NPL film is shown in Figure S6a. The PL spectra from four different areas in this region are plotted in Figure S6b.

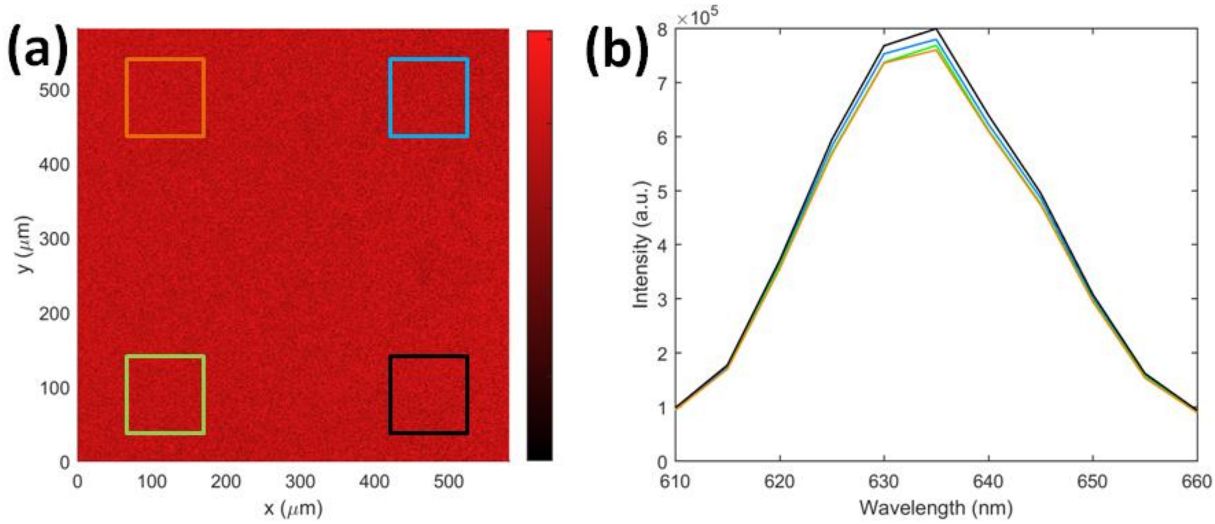

Figure S6: PL mapping of the 14-layered sample taken with a confocal microscope. (b) PL spectra collected at four different spots (marked with coloured squares in panel a) on the studied region.

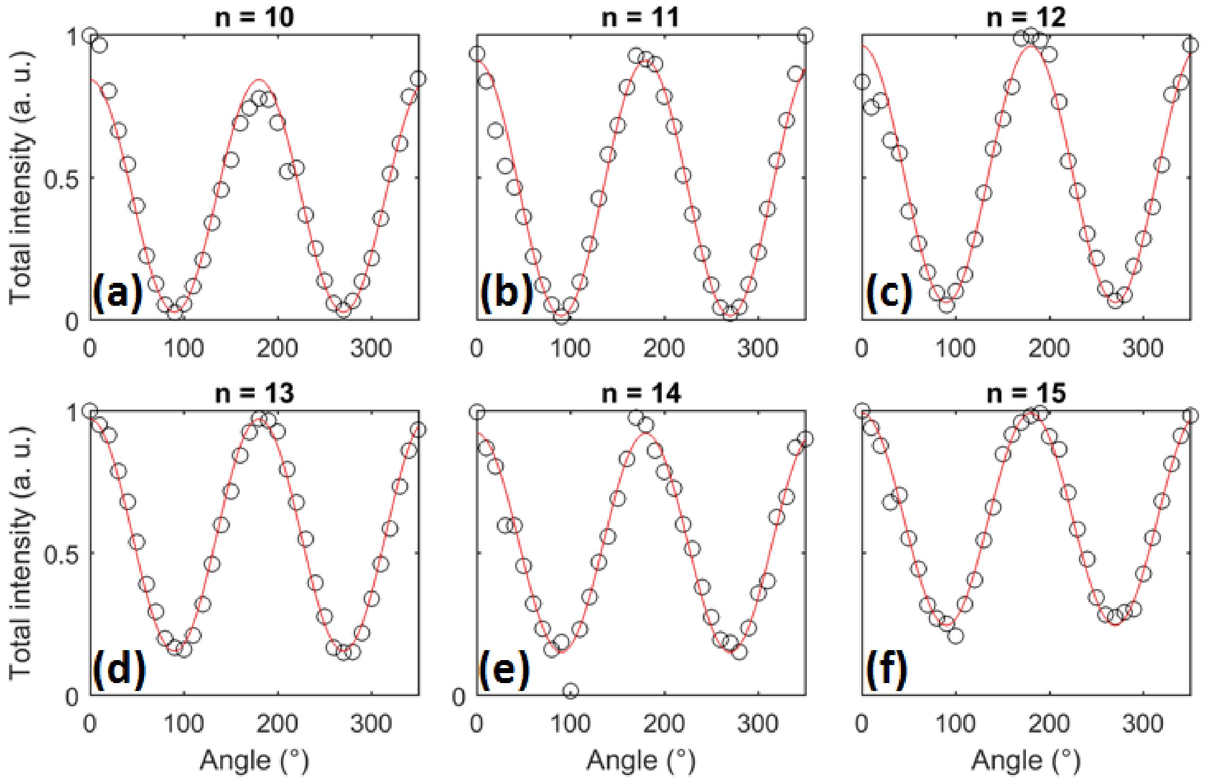

Figure S7: Polarization measurements of the ASE from 10- to 15-layered NPL films.

Table S2: Gaussian fitting parameters for the ASE spectra presented in Figure 2d. Peak and FWHM values for spontaneous emission (SE) and ASE features.

# NPL layers	SE peak (nm)	SE width (nm)	ASE peak (nm)	ASE width (nm)
6	647.6	31.8	650.2	7.2
7	648.8	32.6	647.4	7.4
8	650.3	29.1	653.7	5.9
9	647.8	30.3	652.7	7.6
10	651.3	35.0	657.1	6.2
11	649.7	35.5	660.8	5.9
12	648.0	34.0	661.9	5.7
13	647.7	32.3	663.0	5.5
14	647.7	33.0	664.8	5.5
15	645.9	31.1	664.1	7.2

References

(1) Altintas, Y.; Gungor, K.; Gao, Y.; Sak, M.; Quliyeva, U.; Bappi, G.; Mutlugun, E.; Sargent, E. H.; Demir, H. V. Giant Alloyed Hot Injection Shells Enable Ultralow Optical Gain Threshold in Colloidal Quantum Wells. *ACS Nano* **2019**, *13*, 10662–10670.

(2) Liu, B.; Altintas, Y.; Wang, L.; Shendre, S.; Sharma, M.; Sun, H.; Mutlugun, E.; Demir, H. V. Record High External Quantum Efficiency of 19.2% Achieved in Light-Emitting Diodes of Colloidal Quantum Wells Enabled by Hot-Injection Shell Growth. *Adv. Mater.* **2020**, *32*, 1905824.

(3) Beck, M.; Graczyk, M.; Maximov, I.; Sarwe, E. L.; Ling, T. G. I.; Keil, M.; Montelius, L. Improving Stamps for 10 Nm Level Wafer Scale Nanoimprint Lithography. *Microelectron. Eng.* **2002**, *61–62*, 441–448.