

## Supporting Information

# Giant Alloyed Hot Injection Shells Enable Ultra-Low Optical Gain Threshold in Colloidal Quantum Wells

*Yemliha Altintas,<sup>a,b,†</sup> Kivanc Gungor,<sup>a,†</sup> Yuan Gao,<sup>c,†</sup> Mustafa Sak,<sup>a</sup> Ulviyya Quliyeva,<sup>a</sup> Golam Bappi,<sup>c</sup> Evren Mutlugun\*,<sup>a,b</sup> Edward H. Sargent\*,<sup>c</sup> and Hilmi Volkan Demir\*,<sup>a,d</sup>*

<sup>a</sup>Department of Electrical and Electronics Engineering and Department of Physics UNAM – Institute of Materials Science and Nanotechnology, Bilkent University Ankara 06800, Turkey

<sup>b</sup>Department of Materials Science and Nanotechnology and Department of Electrical-Electronics Engineering, Abdullah Gül University, Kayseri, TR-38080, Turkey

<sup>c</sup>Department of Electrical and Computer Engineering, University of Toronto, 10 King's College Road, Toronto, ON M5S 3G4, Canada

<sup>d</sup>Luminous! Center of Excellence for Semiconductor Lighting and Displays, School of Electrical and Electronic Engineering, School of Physical and Mathematical Sciences, School of Materials Science and Nanotechnology, Nanyang Technological University, Singapore 639798, Singapore

\*E-mail: [hvdemir@ntu.edu.sg](mailto:hvdemir@ntu.edu.sg), [volkan@bilkent.edu.tr](mailto:volkan@bilkent.edu.tr) \*E-mail: [ted.sargent@utoronto.ca](mailto:ted.sargent@utoronto.ca)

\* E-mail: [evren.mutlugun@agu.edu.tr](mailto:evren.mutlugun@agu.edu.tr)

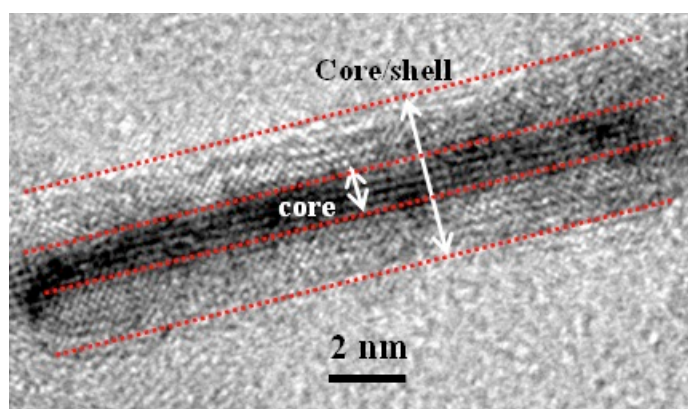
**Chemicals.** All chemicals were used without any additional purification as received from Sigma Aldrich as follows: zinc acetate (99.99%), cadmium acetate (anhydrous, 99.995%), 1-octadecene (ODE, 90%), oleic acid (OA, 90%), 1-octanethiol ( $\geq 98.5\%$ ), oleylamine (OLA, 70%), sodium myristate ( $\geq 99.0\%$ ), cadmium nitrate tetrahydrate ( $\geq 99.0\%$ ), selenium (99.99%), n-hexane ( $\geq 97.0\%$ ), methanol ( $\geq 99.7\%$ ), absolute ethanol, toluene ( $\geq 99.5\%$ ).

**Preparation of the Cd-myristate:** Cd-myristate was prepared according to the previous literature.<sup>1</sup> Sodium myristate and cadmium nitrate tetrahydrate were separately dissolved in methanol by using a magnetic stirrer at room temperature until the solution became clear. Then, both solutions were mixed and stirred until becoming bulky. Then, the solution was filtered and washed two times and left overnight to dry under vacuum.

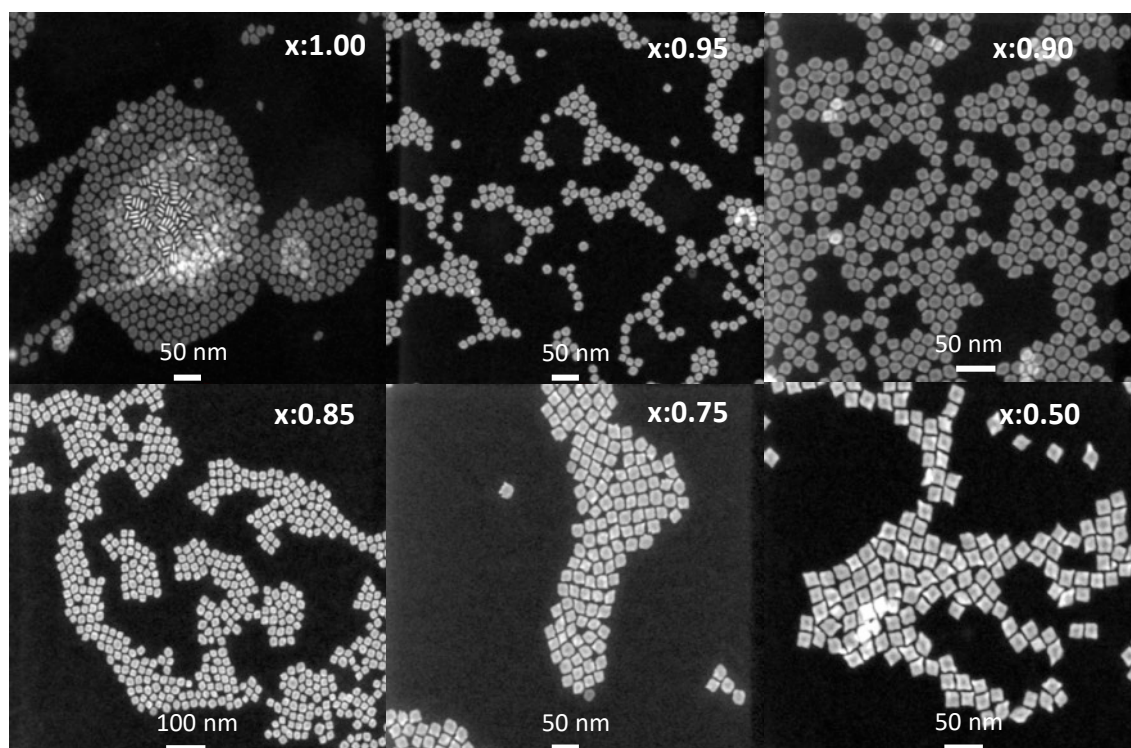
**4 ML CdSe core NPL synthesis.** CdSe core NPLs were synthesized by using a well-known synthesis protocol with slight modifications.<sup>1</sup> Cd myristate (340 mg), selenium (24 mg) and ODE (30 mL) were mixed in 100 mL of four-necked flask and heated to 95 °C under vacuum for one hour. The temperature of the solution was increased to 240 °C under argon gas flow. At 195 °C, 120 mg of cadmium acetate dihydrate was rapidly added into the solution and the solution was kept at this temperature for 9 min. Before cooling to the room temperature, 1 mL of OA was added into the reaction flask and then the solution was cooled down quickly with cold water. As synthesized NCs were cleaned two times with ethanol and 4ML pure CdSe core NPLs was dispersed in hexane after cleaning.

## Characterization

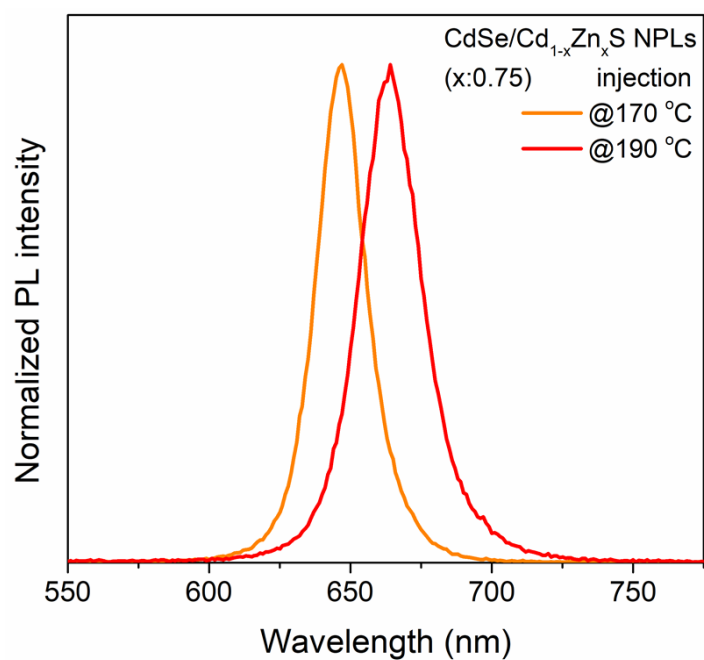
Photoluminescence and absorption characterizations were performed by using Cary Eclipse fluorescence spectrophotometer and UV-Visible spectrophotometer of Varian Cary 100 Bio, respectively. Panalytical Xpert Pro MPD and Thermo Scientific K-Alpha X-Ray spectrometer were used for structural characterization of the synthesized core/shell NPLs to obtain X-ray diffraction pattern and X-ray photoelectron spectroscopy (XPS), respectively. Transmission electron microscopy (TEM) images and energy-dispersive X-ray spectroscopy (EDS) data were collected by using Tecnai G2 F30 at 300 kV. QY of the samples was determined by using absolute QY measurement system. The methodology of QY measurements can be found in Mello *et al.*<sup>2</sup> The system is composed of a spectrometer (Ocean Optics Maya 2000), an integrating sphere (Hamamatsu) and a monochromator integrated Xenon lamp. The excitation wavelength was set to 400 nm for these measurements.



**Figure S1.** Core/shell structure of the synthesized core/g-HIS NPLs shown from cross-sectional view.



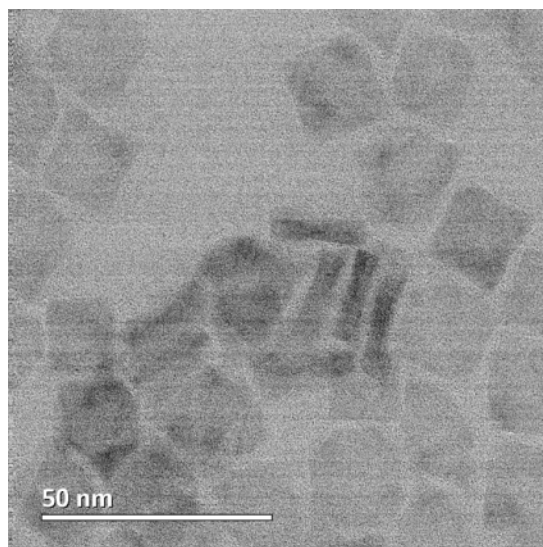
**Figure S2.** HAADF-STEM images of CdSe/Cd<sub>1-x</sub>Zn<sub>x</sub>S core/ alloyed g-HIS NPLs using the tuned values of  $x$  composition.



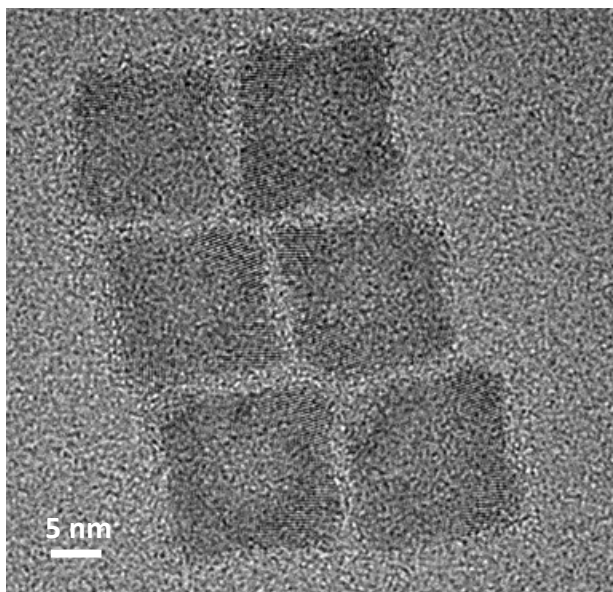
**Figure S3.** Effect of the injection temperature of the S-precursor on the PL of synthesized core/alloyed g-HIS NPLs.

**Table S1.** Optical properties of 10 CdSe/Cd<sub>0.25</sub>Zn<sub>0.75</sub>S core/alloyed g-HIS NPL samples consecutively synthesized. QY of these samples was measured by using absolute QY measurement system at an excitation wavelength of 400 nm.

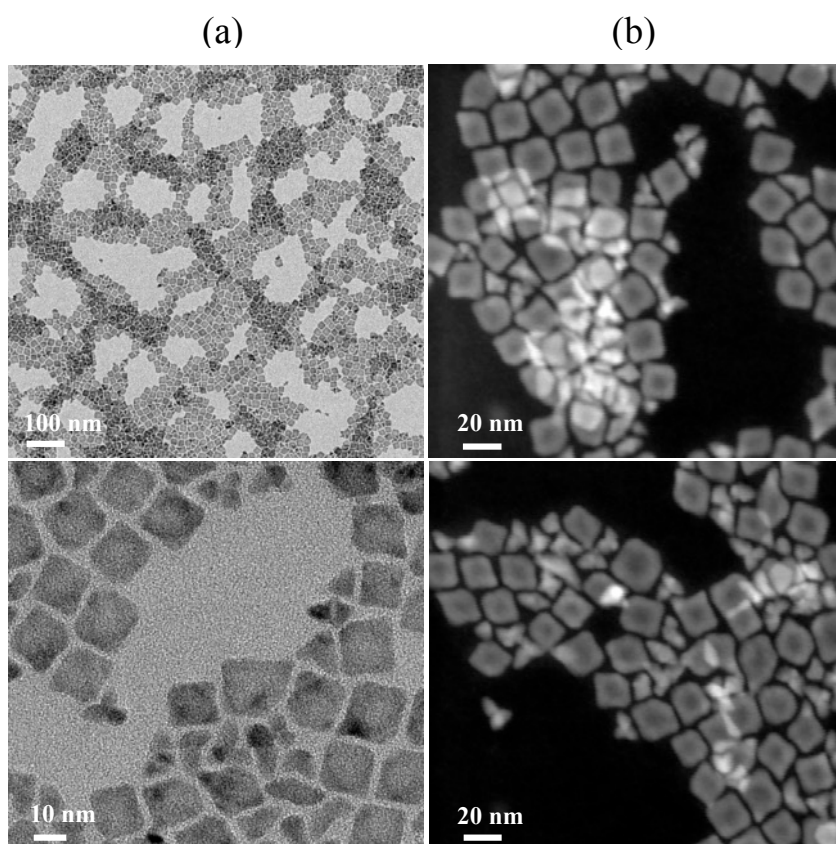
# of the synthesis	Peak Emission Wavelength (nm)	FWHM (nm)	QY (%)
1	650	21	98
2	648	22	100
3	648	22	94
4	650	24	96
5	651	22	92
6	650	22	100
7	648	23	94
8	651	23	96
9	648	20	95
10	650	24	93



**Figure S4.** Cross-sectional TEM images of the synthesized CdSe/Cd<sub>0.25</sub>Zn<sub>0.75</sub>S core/alloyed g-HIS NPLs.

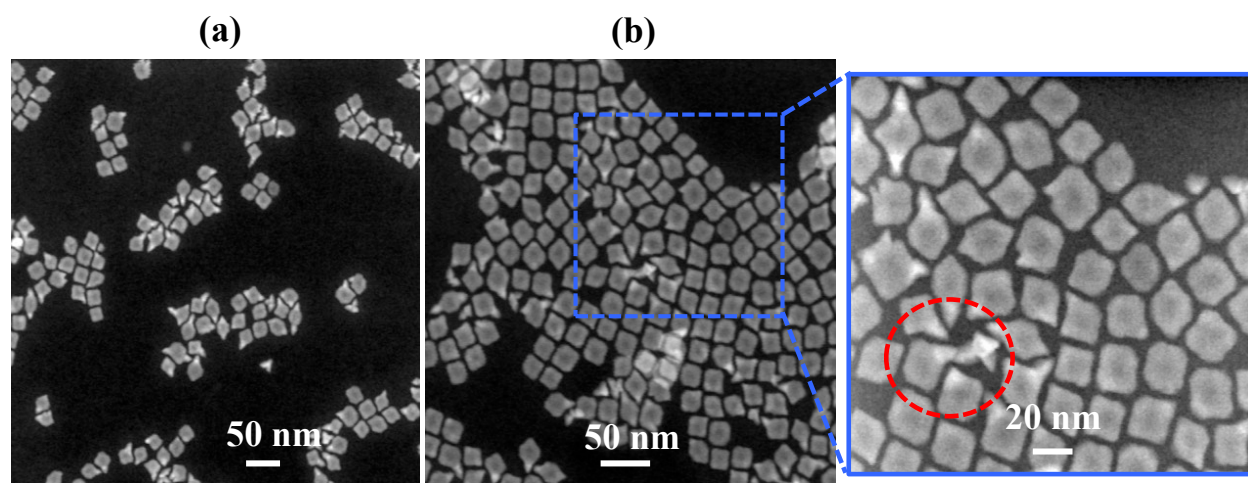


**Figure S5.** HR-TEM image of the CdSe/Cd<sub>1-x</sub>Zn<sub>x</sub>S core/alloyed g-HIS NPLs sample.

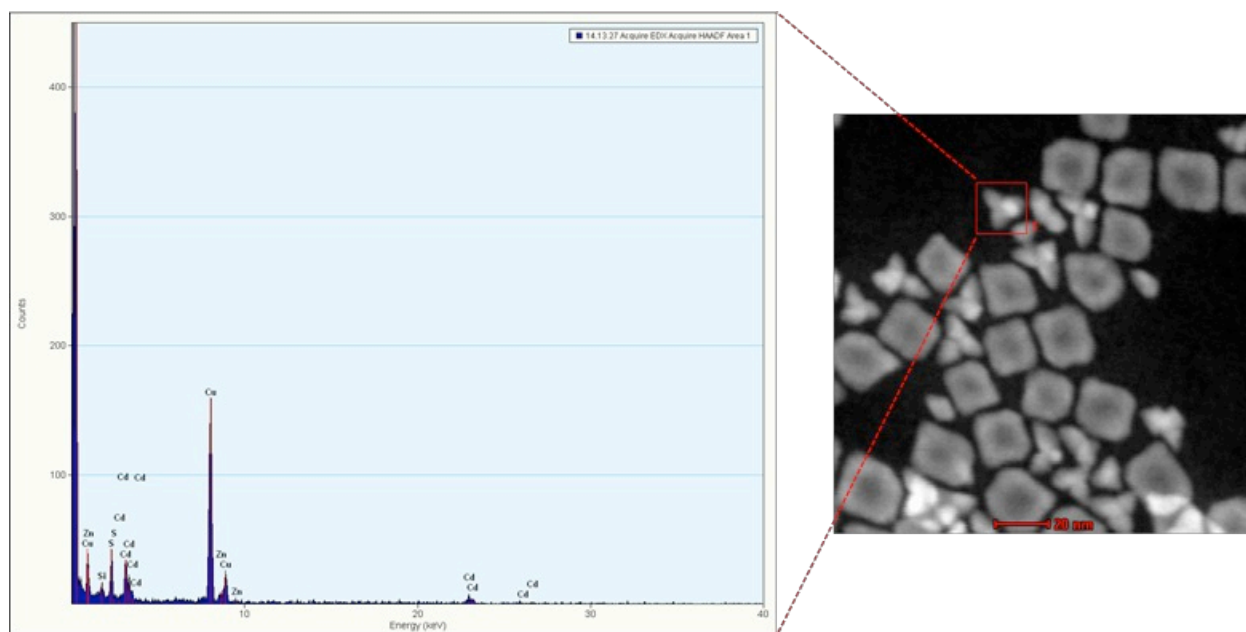




**Figure S6.** a) HR-TEM and b) HAADF-STEM images of the synthesized CdSe/Cd<sub>0.75</sub>Zn<sub>0.25</sub>S core/alloyed g-HIS NPLs.

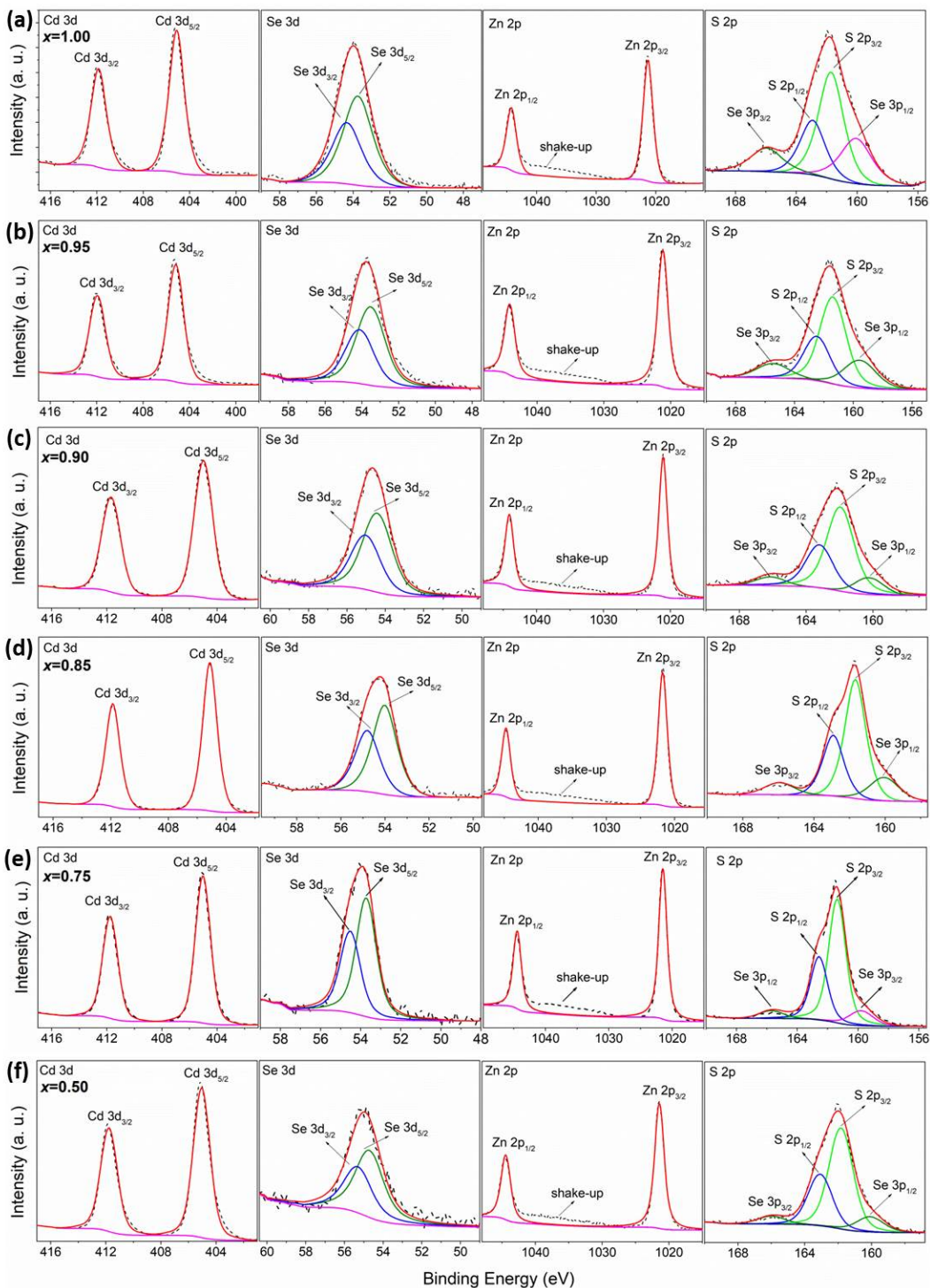


**Figure S7.** a) and b) HAADF-STEM images of the CdSe/Cd<sub>0.75</sub>Zn<sub>0.25</sub>S core/alloyed g-HIS NPLs. These images show outward growing profile of the NPLs from their edges and also a second particle formation has been observed in the TEM images just before breaking off from the edges of NPL as presented in the circled area.



**Figure S8.** When the concentration of the Cd was set to 0.75 (*i.e.*,  $x=0.25$  in  $\text{CdSe/Cd}_{1-x}\text{Zn}_x\text{S}$  core/alloyed g-HIS NPLs), extensions of NPLs from edges broke off and appeared as separate nanocrystals as shown in Figures S7 and S8. From the STEM images and energy-dispersive X-ray (EDX) analyses of the samples given here, broken parts of the NPLs were identified as CdZnS nanocrystals.

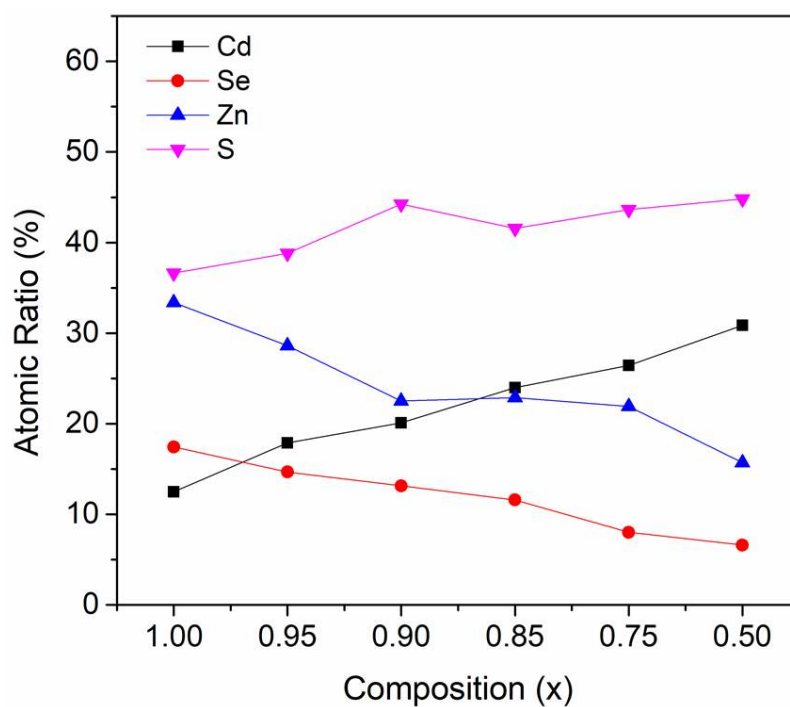




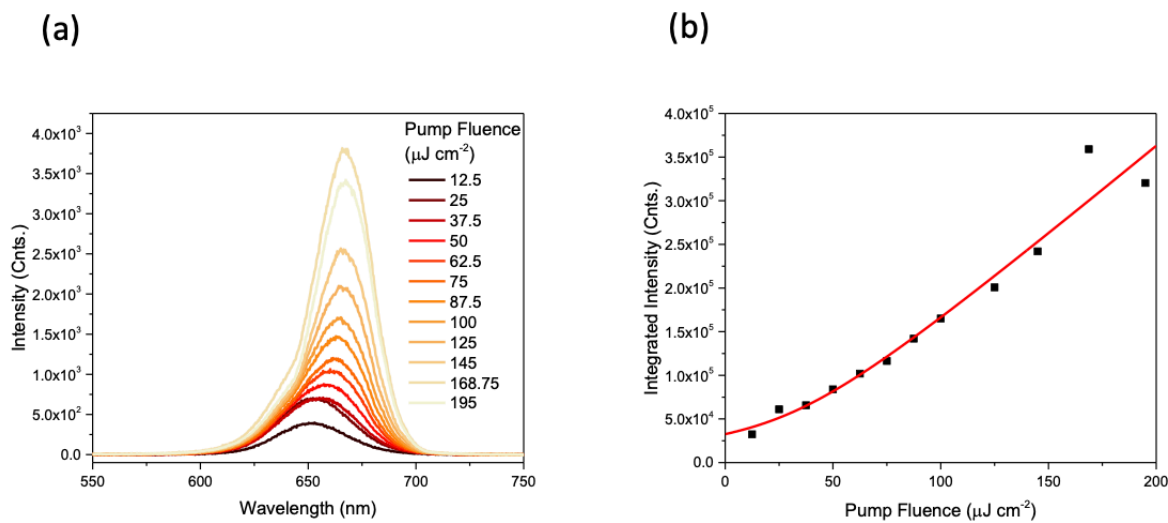
**Figure S9.** XPS spectra of the CdSe/Cd<sub>1-x</sub>Zn<sub>x</sub>S core/ alloyed g-HIS NPLs samples a)  $x=1.00$ , b)  $x=0.95$ , c)  $x=0.90$ , d)  $x=0.85$ , e)  $x=0.75$ , and f)  $x=0.50$ . As-synthesized NPLs were cleaned twice and spin-coated on silicon substrates. The elements of the core/shell NPLs structure and their elemental binding energies were measured by XPS, and all curves were fit to precisely calculate the atomic percentages of each element in the structure.

**Table S2.** Measured atomic ratios of Zn and Cd elements in the structure of the core/ alloyed g-HIS NPLs from XPS and EDX analyses.

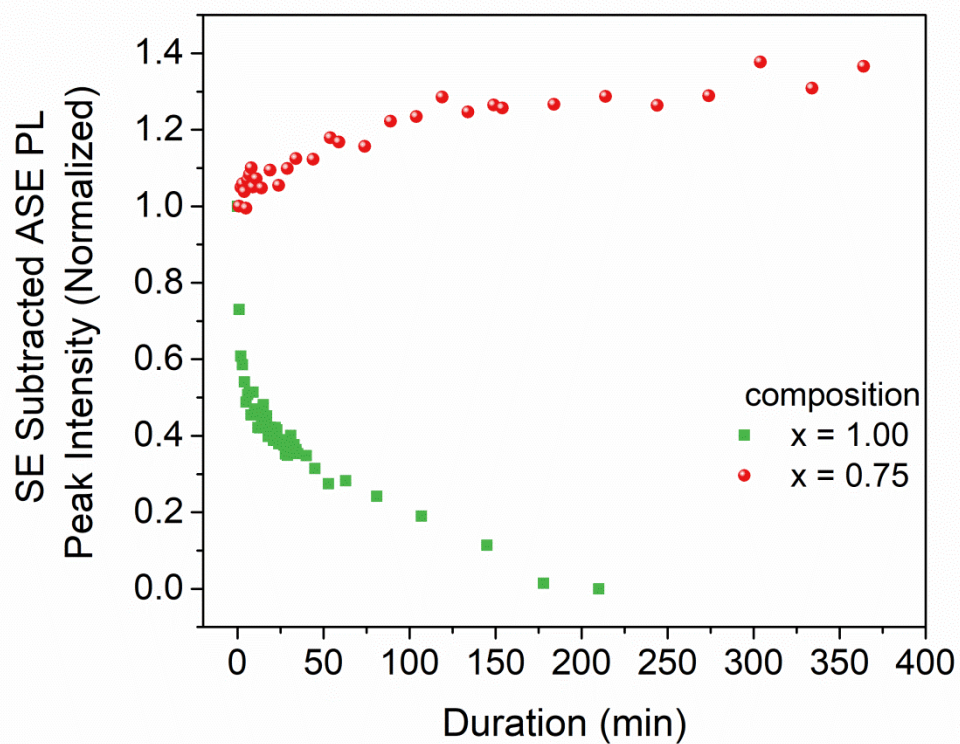
	<b>Zn/Cd atomic ratio</b>	
<i>x</i> composition	From XPS analyses	From EDX analyses
1.00	2.66	2.67
0.95	1.60	1.66
0.90	1.12	1.00
0.85	0.95	0.99
0.75	0.83	0.63
0.50	0.50	0.42



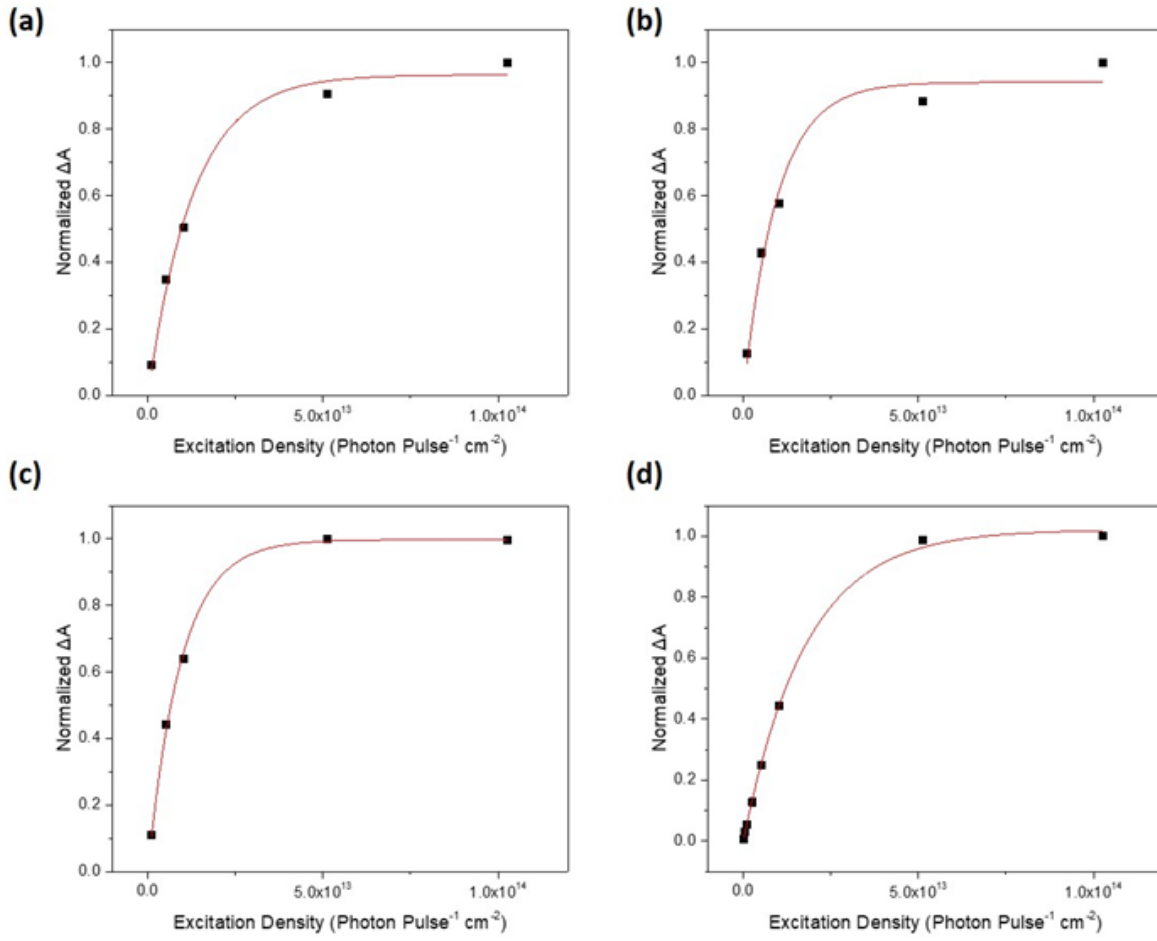
**Figure S10.** Determined atomic ratios of the elements (given in %) with respect to different shell compositions of the CdSe/Cd<sub>1-x</sub>Zn<sub>x</sub>S core/alloyed g-HIS NPLs samples obtained from XPS measurements.



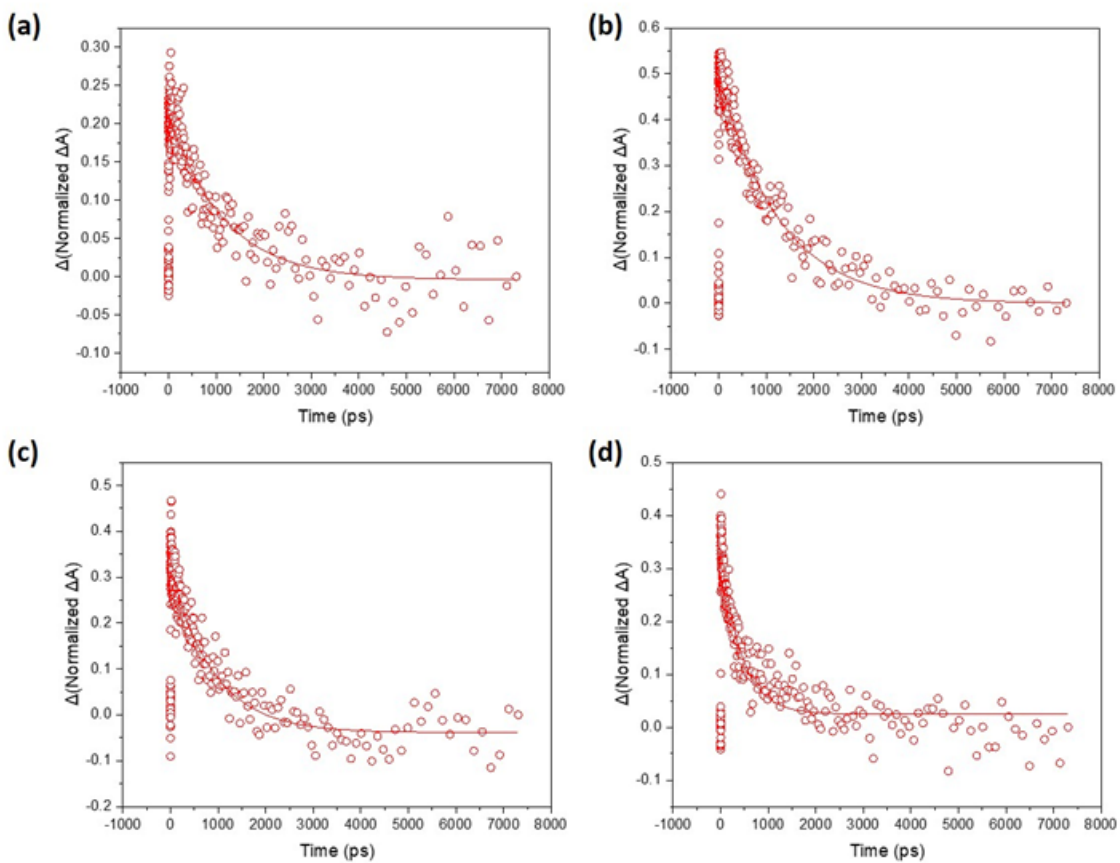
**Figure S11.** (a) The variation of the emission spectra as a function of the pump fluence and (b) the transfer function under 355nm Nd YAG laser pumping with 1 ns pulse width and 100 Hz repetition rate.



**Figure S12.** Amplified spontaneous emission (ASE) peak intensity subtracted from spontaneous emission (SE) under constant optical pumping shows highly stable ASE for  $x=0.75$  shell composition under 1PA.

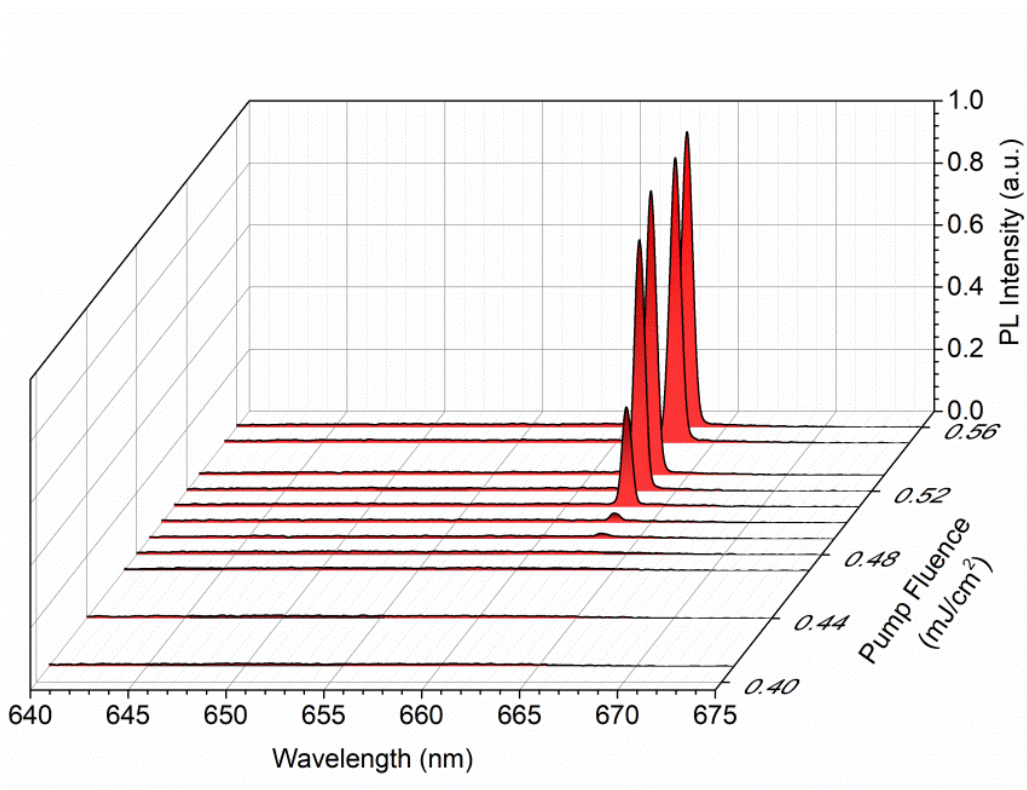


**Figure S13.** Determination of the average number of electron and hole pairs. Normalized intensity of the bleaching bands of (a)  $x=0.50$ , (b)  $x=0.75$ , (c)  $x=0.90$ , and (d)  $x=1.00$  CdSe/Cd<sub>1-x</sub>Zn<sub>x</sub>S core/alloyed g-HIS NPLs as a function of the pump fluence measured at delay time of 7.5 ns when multicarrier recombination is completed. The data points are fitted with  $\Delta A \propto [1 - \exp(-J\sigma)]$ , where  $J$  is the pump fluence and  $\sigma$  is the absorption cross-section of NPLs.



**Figure S14.** Determination of the Auger lifetime of (a)  $x=0.50$ , (b)  $x=0.75$ , (c)  $x=0.90$ , and (d)  $x=1.00$  CdSe/Cd<sub>1-x</sub>Zn<sub>x</sub>S core/alloyed g-HIS NPLs.





**Figure S15.** Evolution of the lasing peak with increasing pump fluence values shown for the same measurement in Figure 4d.

## References

- (1) Tessier, M. D.; Spinicelli, P.; Dupont, D.; Patriarche, G.; Ithurria, S.; Dubertret, B. Efficient Exciton Concentrators Built from Colloidal Core/Crown CdSe/CdS Semiconductor Nanoplatelets. *Nano Lett.* **2014**, *14*, 207–213.
- (2) De Mello, J.; Wittmann, H.; Friend, R. An Improved Experimental Determination of External Photoluminescence Quantum Efficiency. *Adv. Mater.* **1997**, *9*, 230–232.