

## Supporting Information

### Observation of Optical Gain from Aqueous Quantum Well Heterostructures in Water

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**Chemicals:** Sodium myristate, cadmium acetate dihydrate ( $\text{Cd}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$ ), cadmium nitrate tetrahydrate ( $\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ ), zinc nitrate hexahydrate ( $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ), oleylamine (OLA), N-methylformamide (NMF), ammonium sulfide solution (40-48 wt. % in water), 1-octadecene (ODE), oleic acid (OA), selenium, hexane, acetonitrile, toluene, chloroform, ethylenediamine (EDA) and 3-mercaptopropionic acid (MPA) were purchased from Sigma-Aldrich.

**Synthesis of cadmium myristate:** Cadmium myristate was synthesized according to a previously published protocol with slight modifications.<sup>[1]</sup> In a typical synthesis, 6.26 g of sodium myristate and 2.46 g of cadmium nitrate tetrahydrate were separately dissolved in 500 and 80 mL of methanol, respectively. After complete dissolution, they were mixed and stirred vigorously for 3 h. Finally, the white cadmium myristate powder was precipitated by centrifugation. To remove any undesired impurities and/or side products, the precipitate was washed a couple of times with methanol. The final product was dried overnight under vacuum at room temperature (RT) and stored under ambient conditions.

**Synthesis of CdSe core of CQWs:** 4 monolayer (ML) CdSe CQWs (i.e. consisting of four Se atomic layers and five Cd atomic layers) were synthesized using a former procedure reported

in the literature. <sup>[1]</sup> In this protocol, 68 mg of Se powder and 800 mg of cadmium myristate were added to 60 mL of ODE in a 100 mL three-neck flask and degassed at 95 °C for 1 h. Then, the temperature was set to 240 °C under the argon flow. At 195 °C, 200 mg of cadmium acetate dihydrate was added to the reaction. This solution was kept at 240 °C for 8 min. Finally, growth was terminated by addition 4 mL of oleic acid and quenched with a cold-water bath. Monodisperse 4 ML CdSe CQWs were obtained through size selective precipitation of the reaction mixture. The final product was stored in hexane.

Synthesis of crown growth precursor: A previously reported procedure was followed with slight modifications. <sup>[1]</sup> Typically, 480 mg of cadmium acetate dihydrate, 340  $\mu$ L of OA, and 2 mL of ODE were loaded in a beaker. The solution was sonicated for 30 min at room temperature under ambient atmosphere. By continuous stirring at 160 °C and sonication at 100 °C alternately, a white homogeneous solution was obtained. After cooling the sample to the room temperature, 3 mL of 0.1 M sulfur in ODE was added to the mixture. The final solution was kept under constant stirring.

Synthesis of CdSe/CdS core/crown CQWs: A typical core-seeded synthesis method reported previously was used with slight modifications. <sup>[1]</sup> 4 ML CdSe core CQWs and 15 mL of ODE were introduced into a three-neck flask. The solution was degassed at 90 °C for 1h. Then, a certain amount of the growth solution was injected into the mixture at 90°C and degassed for 1h. After that, the solution was heated to 240 °C under argon flow and held at this temperature for 5 min to ensure the proper growth of crown. The reaction was quenched with a cold-water bath. The synthesized core/crown CQWs were precipitated by addition of 5 mL hexane and 3 mL ethanol and centrifugation. Finally, the precipitate was dispersed and stored in hexane.

Deposition of Cd<sub>1-x</sub>Zn<sub>x</sub>S shell layers on CdSe/CdS core/crown CQWs: Cd<sub>1-x</sub>Zn<sub>x</sub>S shell layers were grown on CdSe/CdS core/crown CQW to obtain CdSe/CdS@Cd<sub>1-x</sub>Zn<sub>x</sub>S core/crown@Shell CQW by following a previously reported c-ALD recipe with some

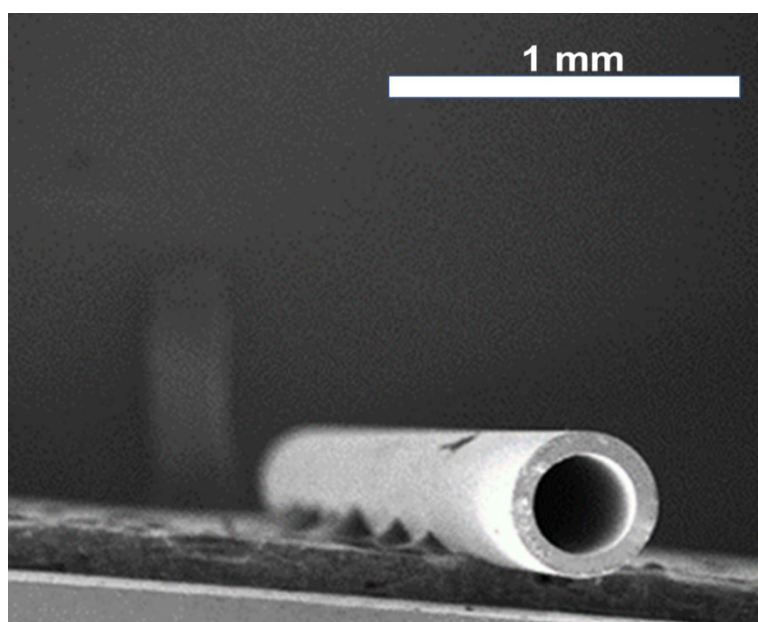
modifications.<sup>[2]</sup> 4 mL of NMF was added to the dispersion of CdSe/CdS core/crown CQWs in 1 mL hexane. Then, 40  $\mu$ L of 40-48% aqueous solution of ammonium sulfide as sulfur shell growth precursor was added and stirred for 2 min. Growth of the sulfur layer was terminated by the addition of 1 mL acetonitrile and excess toluene (till the mixture became blurry) and the mixture was centrifuged. The precipitate was dispersed in NMF and the same cleaning procedure was repeated to remove unreacted precursor. Finally, the CQWs were dispersed in 4 mL of NMF. Cation precursor was prepared by mixing solutions of 0.4 M cadmium nitrate tetrahydrate and 0.4 M zinc nitrate hexahydrate in NMF by a desired volume fraction to obtain a gradient shell.

For the cation deposition, 1 mL of cation precursor mixture was added to CQW dispersion in NMF and the solution was stirred for 45 min. Growth of the cation shell layer was terminated by the addition of 1 mL acetonitrile and excess toluene (till the mixture became blurry) and the mixture was centrifuged. The precipitate was dispersed in NMF and the same cleaning procedure was repeated to remove unreacted precursor.

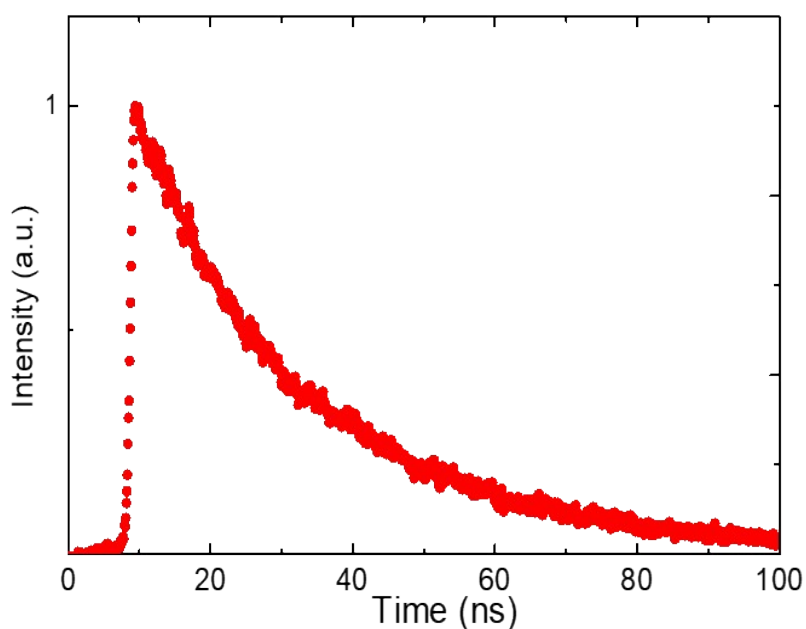
To increase number of shells, the aforementioned steps above were repeated. The fractions of the zinc to cadmium in cation precursors used in this work were 50:50, 95:5, 95:5 and 99:1 for 1<sup>st</sup>, 2<sup>nd</sup>, 3<sup>rd</sup> and 4<sup>th</sup> shell layers, respectively. Lastly, CQWs were precipitated, and 5 mL of hexane and 100 mL of OLA were added and the mixture was stirred overnight. To get rid of excess ligands, the dispersion of CQWs was precipitated by addition of a minimum amount of ethanol and stored in hexane.

Ligand exchange: We followed a previously reported method to obtain CQWs that are dispersible in water.<sup>[2]</sup> Core/crown@shell CQWs in hexane were precipitated with addition of ethanol and centrifugation. The precipitate was redispersed in 3 mL chloroform. 1 mL of EDA was added to this mixture and stirred for 1 h under ambient conditions. Then, 3 mL of 0.15 M MPA in DI water was added. As the CQWs were transferred from chloroform to water phase,

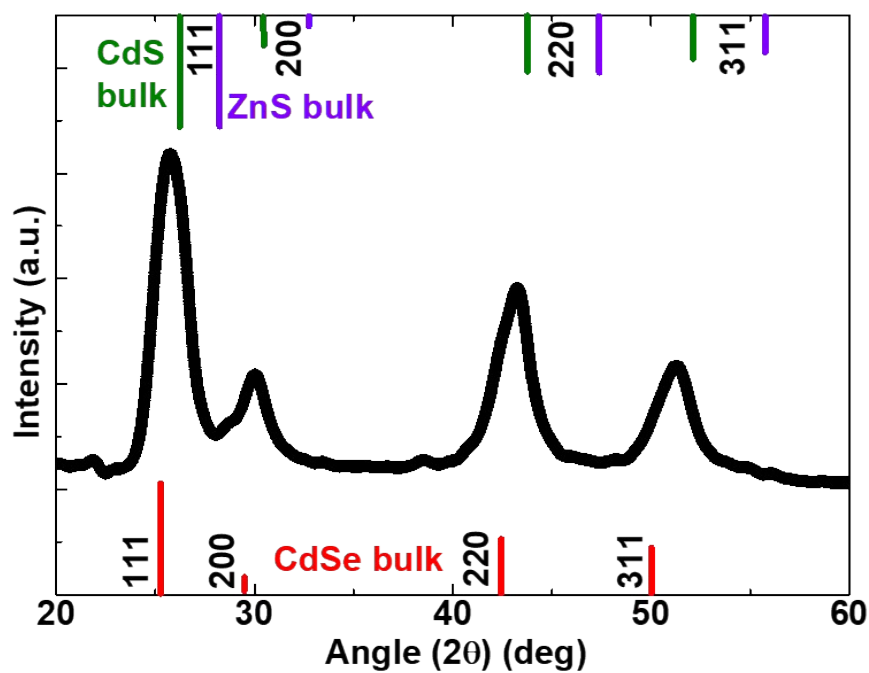
chloroform was removed with the help of a pipette and adding 10 mg of zinc nitrate hexahydrate to the CQWs in water.



**Figure S1.** A scanning electron microscope (SEM) image of the capillary tube used for the in-solution ASE measurements.



**Figure S2.** Time-resolved PL measurements of our CdSe/CdS@Cd<sub>x</sub>Zn<sub>1-x</sub>S core/crown@gradient-alloyed shell CQWs. The intensity weighted lifetime of the PL emission is 33 ns.



**Figure S3.** XRD spectrum of our CdSe/CdS@Cd<sub>x</sub>Zn<sub>1-x</sub>S core/crown@gradient-alloyed shell CQWs.

## References

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