

## Supplementary Information

### Light-Sensitive Monolayer-Thick Nanocrystal Skins of Face-Down Self-Oriented Colloidal Quantum Wells

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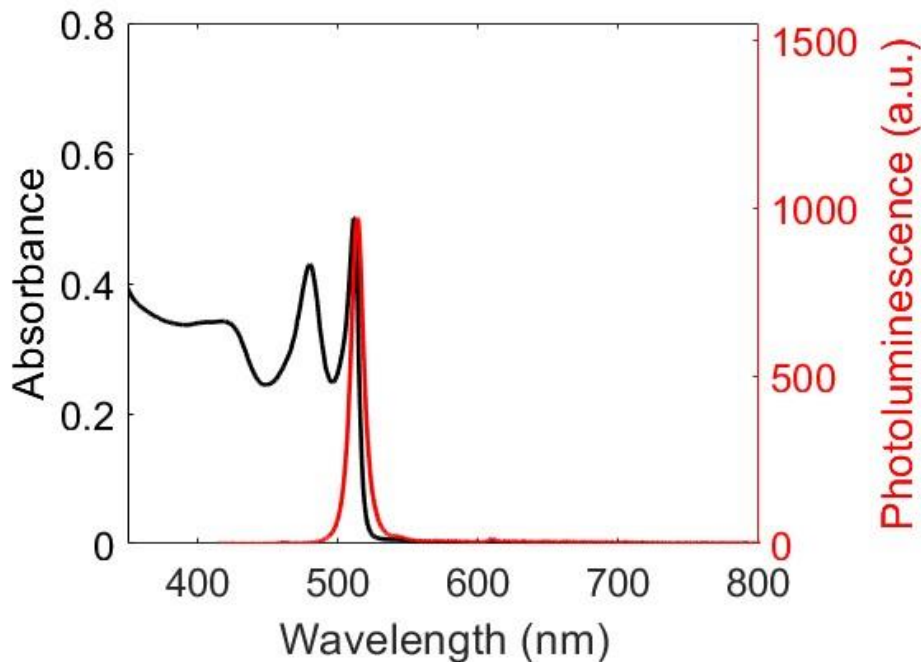
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## **Synthesis of CdSe colloidal quantum wells**

CdSe colloidal quantum wells (CQWs) with the thickness of 4.5 monolayers (ML) were synthesized as given by a previously reported recipe in the literature.<sup>1,2</sup> As the initial step, the reaction mixture in a three-neck flask, which consists of 340 mg of cadmium myristate, 24 mg of selenium, and 15 mL of octadecene (ODE), was vacuumed at 95°C for an hour and then heated to 240°C under Ar atmosphere. At roughly 195°C, 120 mg of cadmium acetate dihydrate was quickly introduced to the reaction mixture when the color of the solution turned dark orange. For 8 min, the solution was stirred at 240°C, and with the addition of 1 mL of oleic acid, the reaction was cooled to room temperature in a cold-water bath. Further purification processes were used to isolate the 4.5 ML CdSe CQWs from other reaction products. The first step involved centrifuging the resultant mixture at 6,000 rpm for 6 min, separating the supernatant into another centrifuge tube. Second, ethanol was added to the supernatant solution until it turned turbid, and the precipitate was dissolved in hexane following the centrifugation at 6,000 rpm for 6 min.

## Optical absorption and photoluminescence spectroscopy



**Figure S1.** Absorption and photoluminescence spectra of the as-synthesized 4.5 ML CdSe colloidal quantum wells in solution.

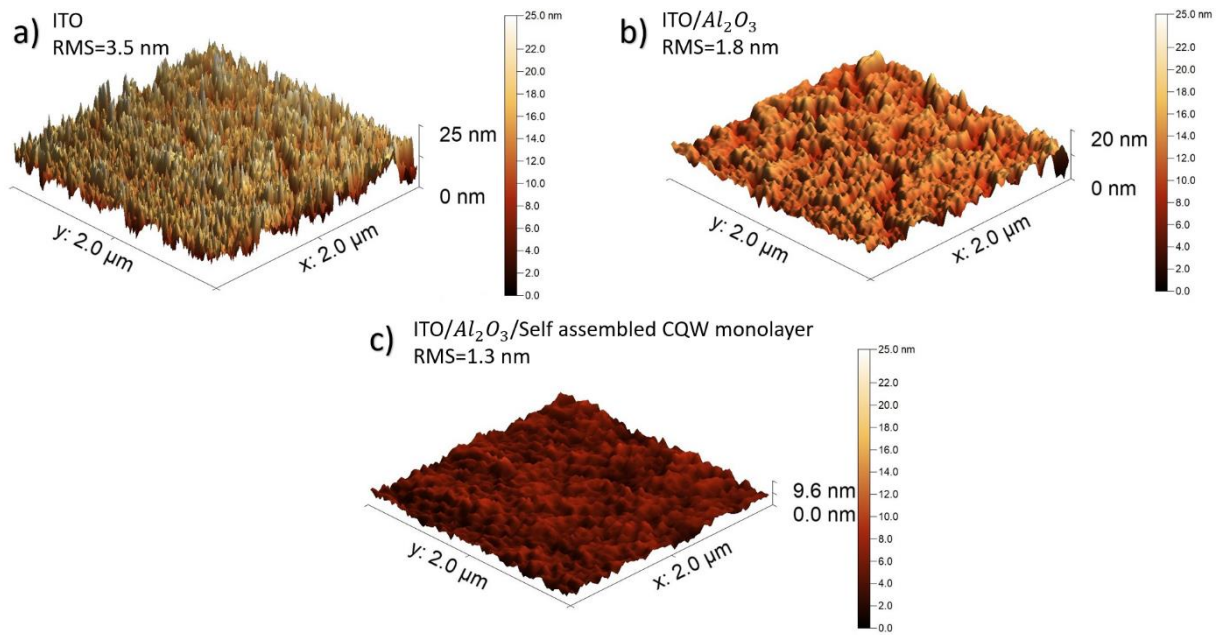
### Fabrication of the 4.5 ML CdSe CQW-based LS-NS devices

To fabricate the LS-NS devices, we used commercially available 100 nm thick ITO-coated glass substrates from Osilla with dimensions of 7.5 cm x 2.5 cm. We cut each of the substrate into three even pieces at a time, as follows: We started cleaning the substrate pieces for 10 min in Hellmanex solution of deionized water. Then, we washed them three times in deionized water for 5 min each. This is important to eliminate any residual contamination of the Hellmanex detergent. Afterward, we sonicated the pieces in acetone and isopropanol for 10 min each time. This completes the first cleaning procedure, which removed most of the major residual contamination on the substrates. Next we took the substrates to the cleanroom for delicate surface cleaning as the second procedure of cleaning before the thermal atomic layer deposition (ALD). In this second step, we also cleaned each piece separately. It is vital to use Teflon tweezers for this washing process. We cleaned the ITO surface of the substrate by first washing with a DI water fountain for around one minute and then washed the surface with

acetone and isopropanol alcohol. Finally, we covered one of the edges for each substrate piece with Kapton tape (0.5 cm) to access the ITO contact when the tape was removed.

Subsequently, we used ALD to coat a 50 nm thick  $Al_2O_3$  film at 300°C. After letting the samples cool to room temperature, we took them out and washed them with DI water, acetone, and isopropanol again. The reason behind this additional cleaning procedure was to eliminate any possible tiny flakes that could possibly come from the ALD chamber during deposition. If such flakes are generated at an early phase of the deposition, they prohibit the growth of a uniform dielectric film, generating bare sites to possibly cause a short circuit on the device. After this washing step, we continued ALD to deposit the next oxide layer of 50 nm in thickness, and the oxide deposition process was completed with a total thickness of 100 nm of the deposited  $Al_2O_3$  film. At this point, we moved on to the self-assembly procedure explained in the main text. Finally, we coated the aluminum contact on the top with thermal evaporation. One critical step to point out before the thermal evaporation is that we isolated each side of every substrate piece by 0.5 cm with Kapton tape. This was necessary to isolate the edges. Since edges are rough regions, it is possible to come across defects that might result in short circuits at the end. Finally, we ended up with an active area of 2.25 cm<sup>2</sup>, since the dimensions of the active area in which we collected photogenerated charges is 1.5 cm × 1.5 cm.

## Atomic force microscopy of ITO, ITO/Alumina and ITO/ $Al_2O_3$ /CQWs



**Figure S2.** Atomic force microscopy of the device sample surface at different stages throughout the fabrication process to show the reduced RMS surface roughness: **a)** Commercial ITO substrate (RMS roughness: 3.5 nm) **b)** alumina-coated ITO substrate (RMS roughness: 1.8 nm) and **c)** ITO/ $Al_2O_3$ /CQW structure (RMS roughness: 1.3 nm).

### Experimental device characterization

To measure voltage, we used Keysight B1500A semiconductor parameter analyzer probe station, where samples were placed on top of a glass stand and contacts were made by utilizing its probes at both ITO and aluminum sides. A 405-nm emitting LED was connected to an external power source for the excitation. Also, we used a custom-made monochromator in the same setup to further characterize the spectral sensitivity by changing the LED excitation source with this monochromator when needed. To assess the optical excitation power, we used an optical powermeter for both cases before placing the LS-NS device into the setup. The probes connected to the device itself were also connected to a shunt resistor in parallel so that the device discharge could also take place at a controlled rate.

## References

1. Bertrand, G. H. V., Polovitsyn, A., Christodoulou, S., Khan, A. H. & Moreels, I. Shape control of zincblende CdSe nanoplatelets. *Chemical Communications* **52**, 11975–11978 (2016).
2. Olutas, M., Guzelturk, B., Kelestemur, Y., Yeltik, A., Delikanli, S., & Demir, H. V. Lateral Size-Dependent spontaneous and Stimulated emission properties in colloidal CDSE nanoplatelets. *ACS Nano*, 9(5), 5041–5050 (2015).