## Supporting Information

## Structural Engineering of Single-Crystal-like Perovskite Nanocrystals for Ultrasensitive Photodetector Applications

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**Figure S1**. XPS spectra of the CsPbBr<sub>3</sub> QD, NW, and NR for (a) Cs 3d, (b) Pb 4f, (c) Br 3d, (d) C 1s, (e) O 1s, and (f) N 1s. (g) Total XPS spectra of the CsPbBr<sub>3</sub> QD, NW, and NR.



**Figure S2.** HRTEM images of the perovskite NWs and NRs. The red lines indicate crystal diffraction planes.



**Figure S3.** Temporal photocurrent response of the CsPbBr<sub>3</sub> NWs and NRs photodetectors under 8-mW illumination. Compared to the NWs device, the NRs device showed a higher photocurrent and a constant response.



**Figure S4.** Dark I-V curve of the CsPbBr<sub>3</sub> QDs-based photodetector depending on the scan direction.



Figure S5. Logarithmic plots of the photocurrent versus illumination power.



Figure S6. Responsivity of the fabricated QDs, NWs, and NRs-based photodetector devices.



**Figure S7.** XRD of the pristine CsPbBr<sub>3</sub> (a) QDs, (b) NWs, and (c) NRs. XRD of the aged CsPbBr<sub>3</sub> (d) QDs, (e) NWs, and (f) NRs after 9 months.



**Figure S8**. FESEM images of the fresh photodetectors based on the CsPbBr<sub>3</sub> perovskite (a) QDs, (b) NWs, and (c) NRs. (d-f) FESEM images of the aged CsPbBr<sub>3</sub> photodetectors based on (d) QDs, (e) NWs, and (f) NRs after 9 months.

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**Figure S9.** (a) X-ray diffraction, (b) SEM image, and (c) OM image of the CsPbBr<sub>3</sub> bulk thin film. (d) X-ray diffraction, (e) SEM image, and (f) OM image of the CsPbBr<sub>3</sub> single crystals.



**Figure S10.** (a) Dark current density and (b) photocurrent density of the CsPbBr<sub>3</sub> QDs, NWs, and NRs-based photodetector devices.