Electronic Supplementary Material (ESI) for Nanoscale.
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Supplementary Materials

Evidence of Band Filling in PbS Colloidal Nanocrystal Square Superstructures

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A. QD Size Distribution and InterQD Distance

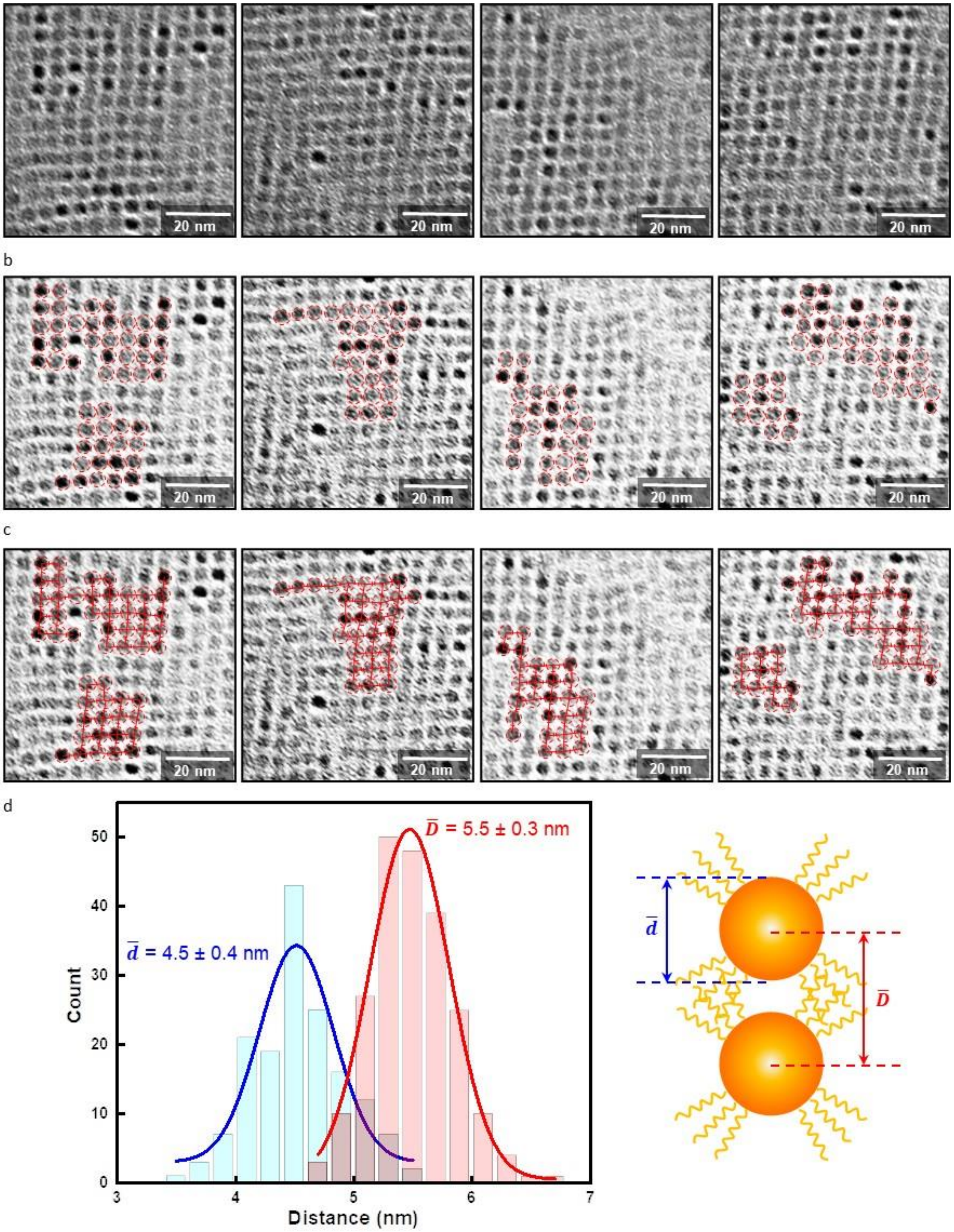

Figure S1. (a) Several high-resolution TEM images of PbS QD square superstructure formed by selective ligand stripping at [100] facets using DMSO. **(b)** The measurements of the QD diameter in the assemblies. **(c)** The measurement of the center-to-center interQD distance in the assemblies. **(d)** Distributions of the average diameter of the PbS QD and the center-to-center interQD distance. The difference of these two values with a broad distribution range suggests that **(e)** the remaining oleic acid in [111] facet may still hinder the epitaxial connection of the [100] facet.

B. Electric-Double-Layer Transistors and Spectroelectrochemistry of 1,2-ethanedithiol-capped PbS QD Assemblies

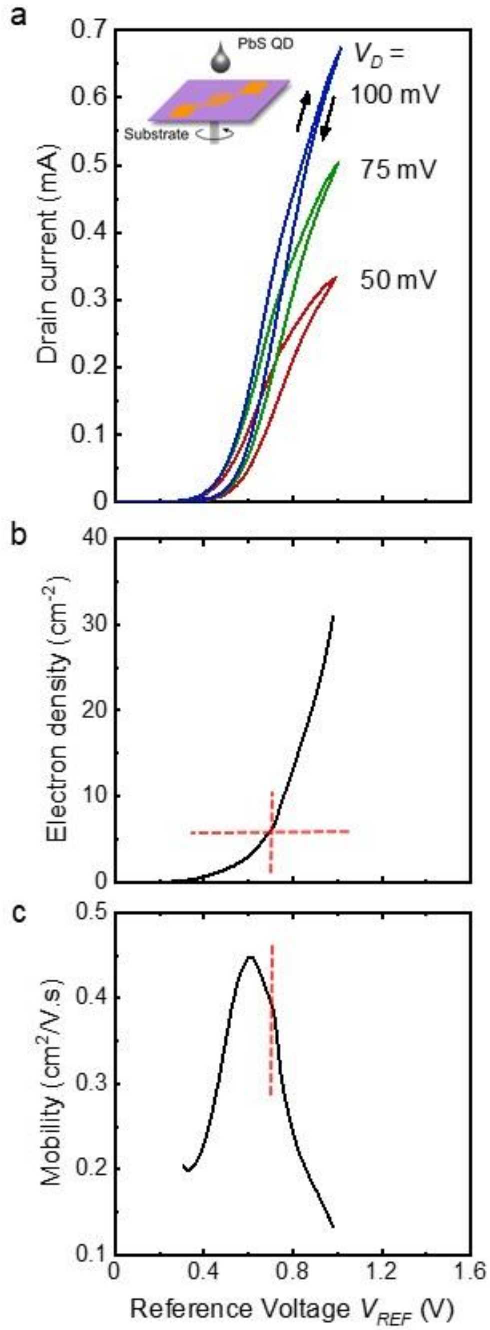

Figure S2. (a) Drain current (I_D) vs. gate reference voltage (V_{REF}) transfer characteristics of the EDLT using spin-coated EDT-capped PbS QD at various applied drain voltage (V_D) values. **(b)** Gate-dependent accumulated sheet electron density. The crosshair indicates the position of the half-filling of the 1*S* level. **(c)** Plots of electron mobility against the reference voltage at V_D= 100 mV.

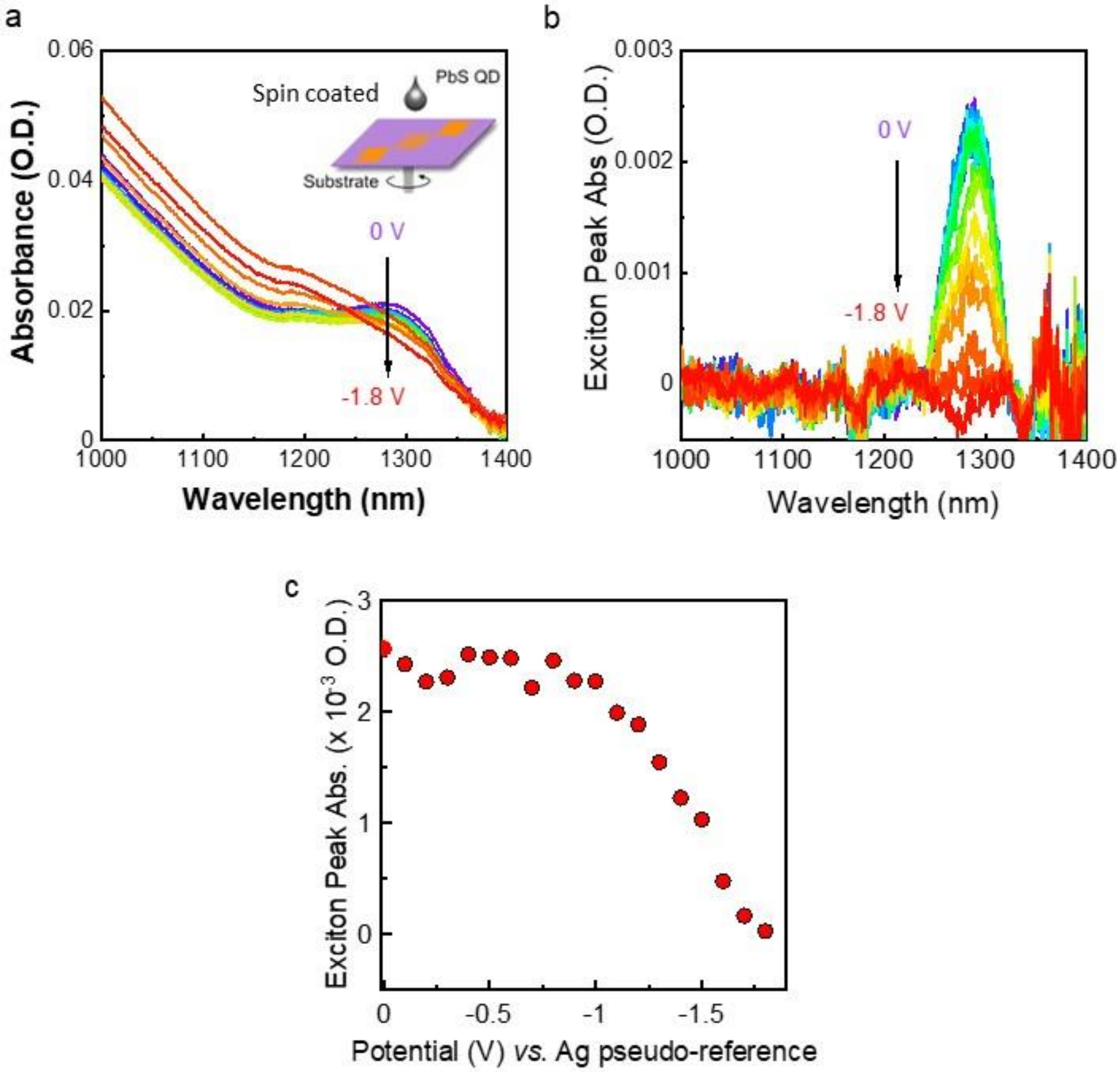

Figure S3. (a) Absorption spectra of the EDT-capped PbS QD film deposited by layer-by-layer spin-coating measured at different applied potential values. **(b)** The change of the QD excitonic peak absorbance by different applied potentials signifies the bleaching process. **(c)** The relative absorbance value of the excitonic peak (λ = 1290 nm) at different applied potential voltages showing that full bleach can only happen way above -1.7 V.

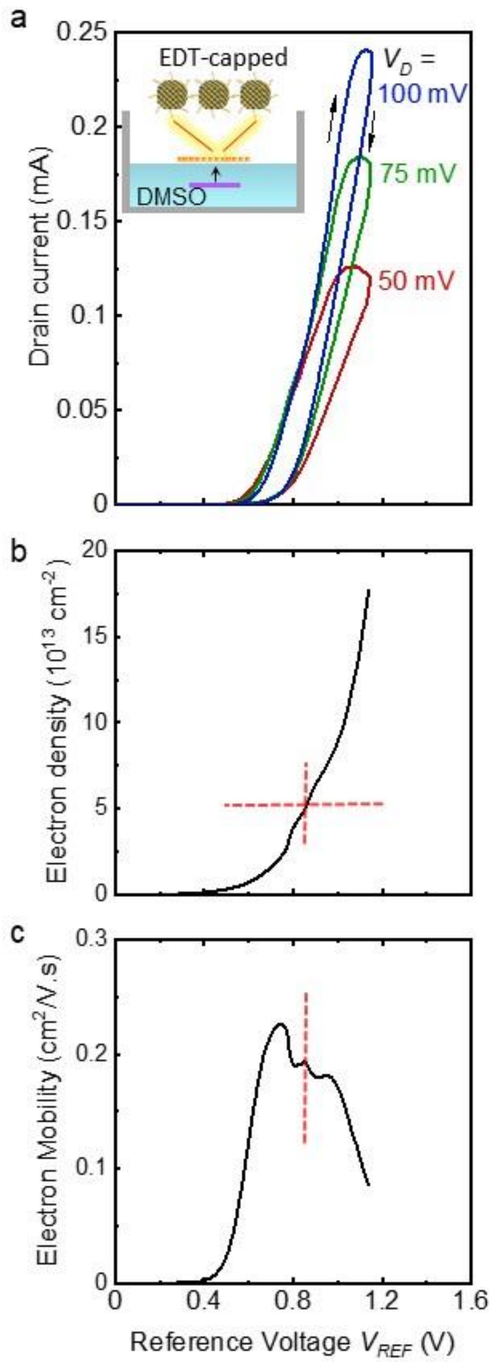

Figure S4. (a) Drain current (I_D) vs. gate reference voltage (V_{REF}) transfer characteristics, at various applied drain voltage (V_D) values, of the EDLT of EDT-capped PbS QD monolayer film prepared by liquid-air interface assembly. The crosshair indicates the position of the half-filling of the 1*S* level. **(b)** Gate-dependent accumulated sheet electron density **(c)** Plots of electron mobility against the reference voltage at V_D= 100 mV.

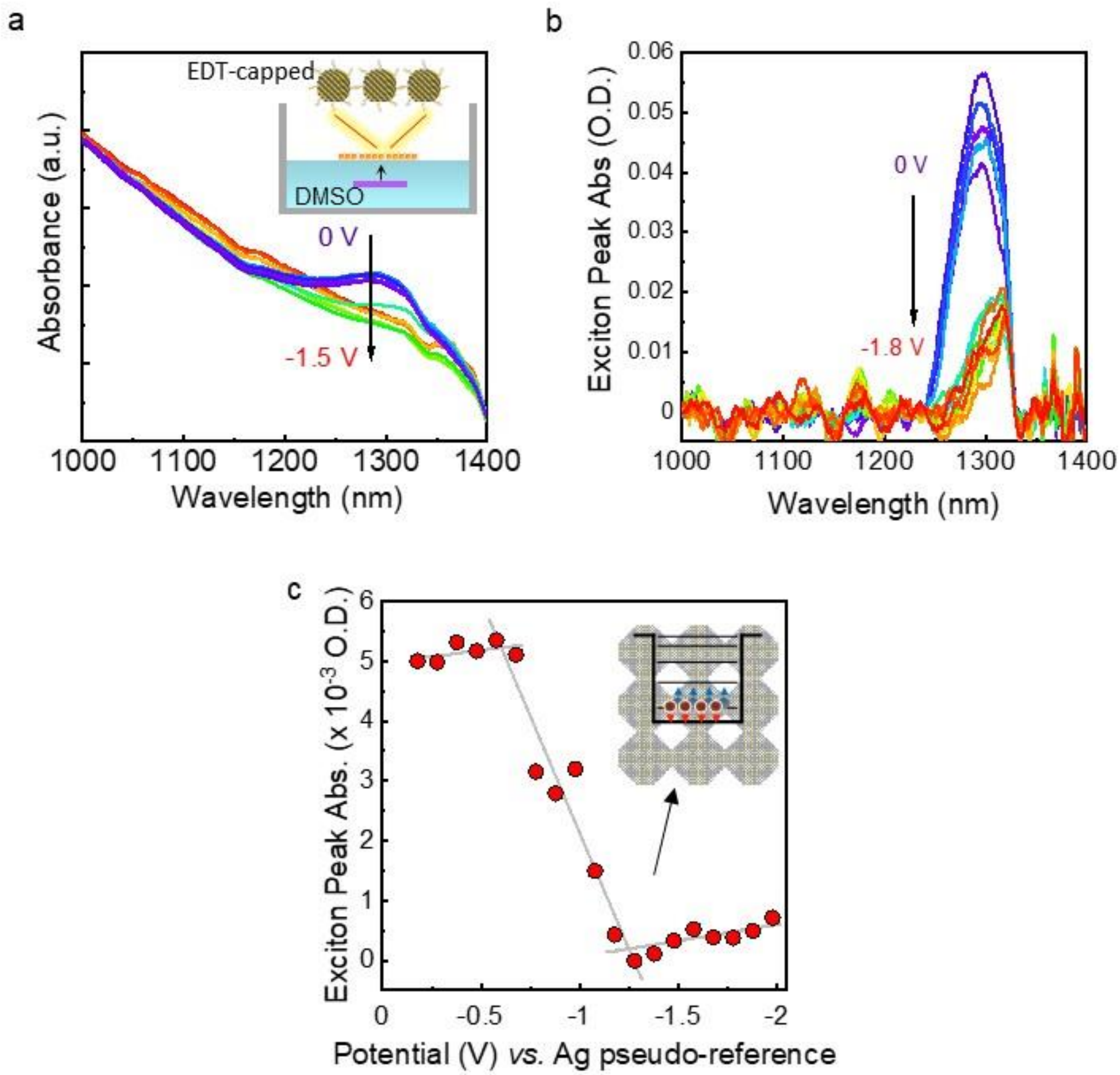

Figure S5. (a) Absorption spectra of the EDT-capped PbS QD film deposited by liquid/air interface assembly measured at different applied potential values. **(b)** The change of the QD excitonic peak absorbance by different applied potentials signifies the bleaching process. **(c)** The relative absorbance value of the excitonic peak (λ = 1290 nm) at different applied potential voltages showing that full bleach happens at -1.25 V.

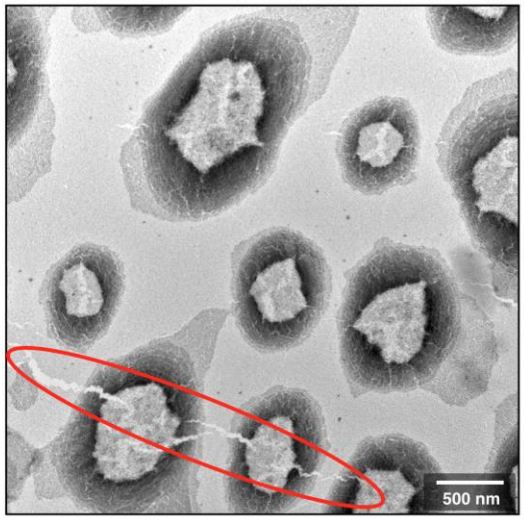

Fig. S5. Large window TEM image of the PbS square lattice. Cracks and multilayer formation of the QD assemblies are still found at this scale.