Supplementary Materials

Evidence of Band Filling in PbS Colloidal Nanocrystal Square Superstructures

Liming Liu†, Ricky Dwi Septianto‡^, Satria Zulkarnaen Bisri‡^*, Yasuhiro Ishida‡, Takuzo Aida,†‡, and Yoshihiro Iwasa‡§

† Department of Chemistry and Biotechnology, School of Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan.
‡ RIKEN Center for Emergent Matter Science, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan.
^Department of Materials Science and Engineering, Tokyo Institute of Technology, 2-12-1 Ookayama, Meguro, Tokyo 152-8550, Japan
§ Quantum-Phase Electronics Center and Department of Applied Physics, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan.

Corresponding Author Email: satria.bisri@riken.jp
A. QD Size Distribution and InterQD Distance

![TEM images of PbS QD square superstructure](image1)

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 1S 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 ($\lambda = 1290$ 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 1S 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 ($\lambda = 1290$ 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.