Supporting Information

Low-Temperature, Solution-Based Sulfurization and Necking of PbS CQD Films

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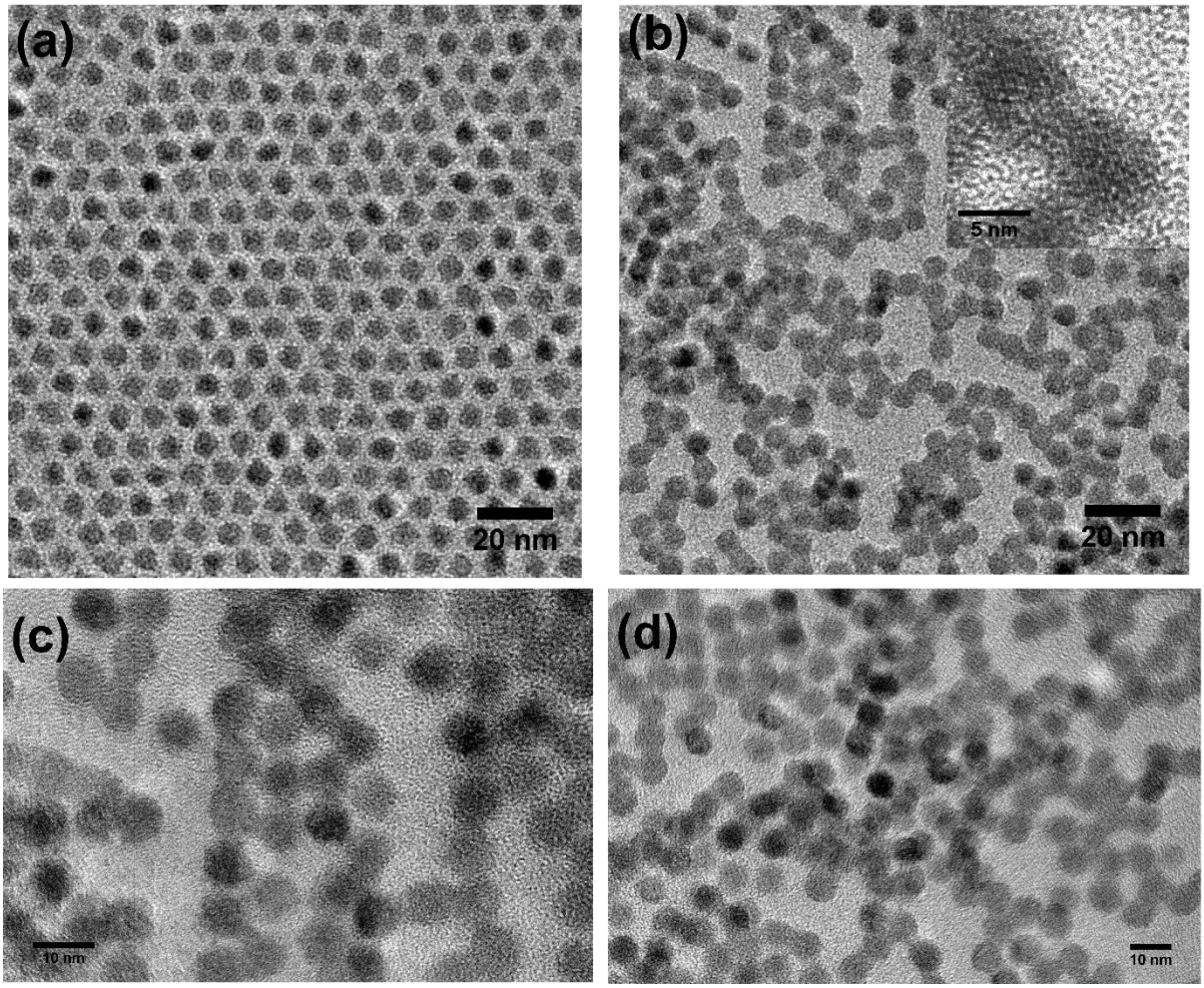
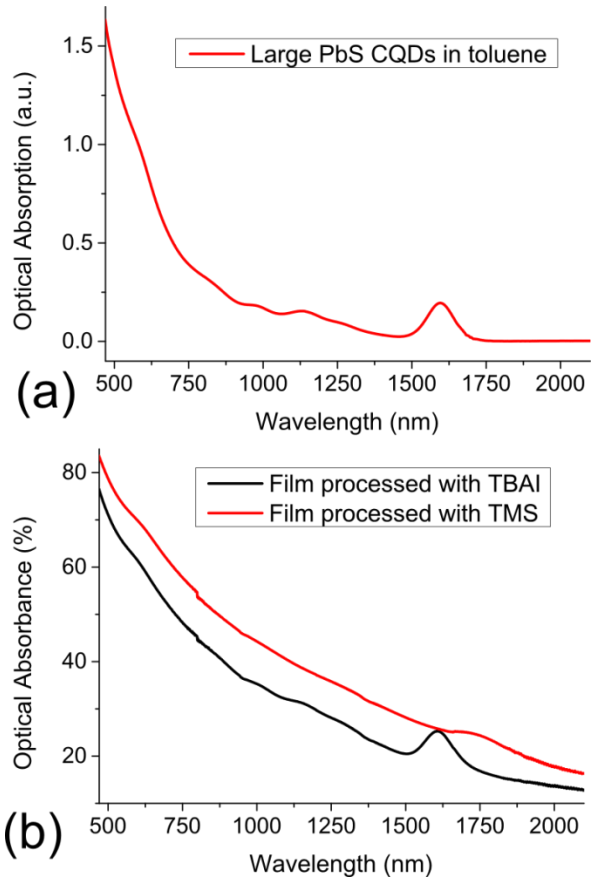
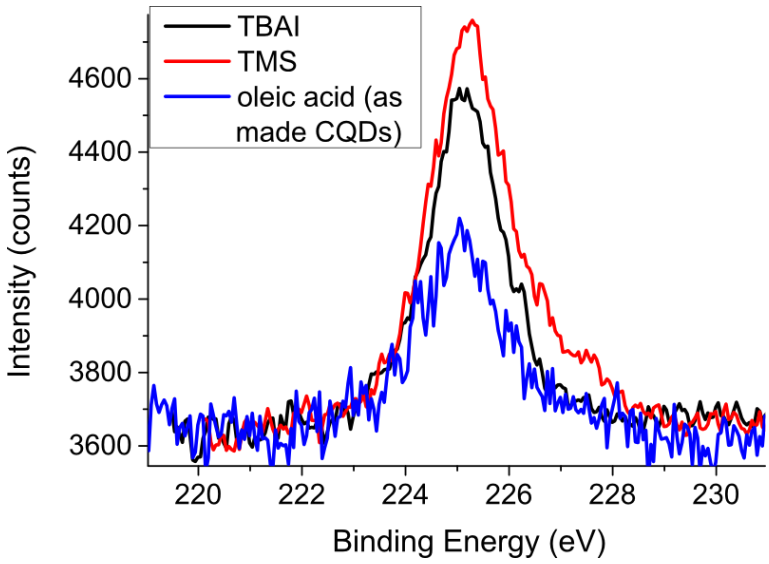


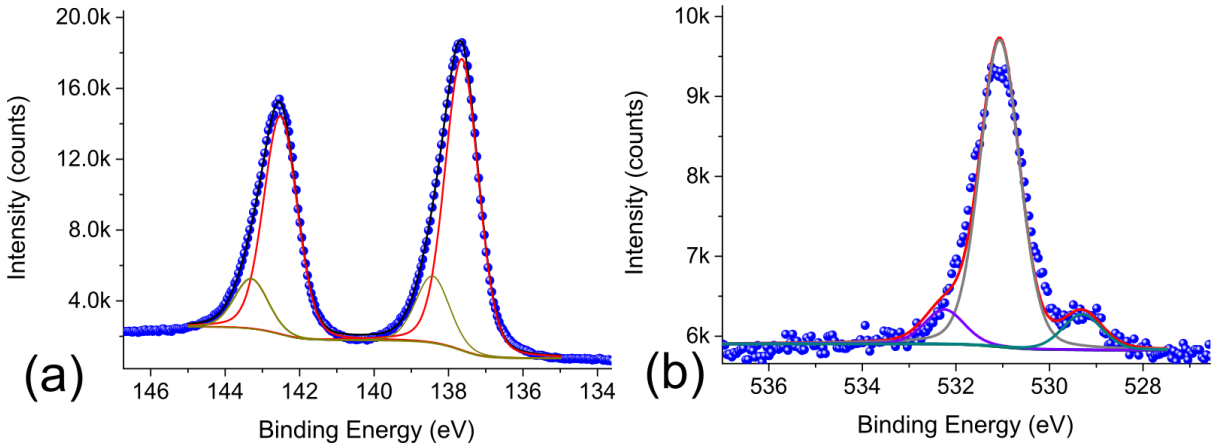
Supporting Figure S1. (a) Figure 1. (a) TEM micrograph of PbS CQDs (exciton peak at 1600 nm) on an amorphous carbon film. (b-d) TEM micrograph of the same PbS CQDs forming aggregates after being treated on film with TMS. The inset of (b) shows an HREM micrograph of two necked CQDs that exhibit a set of aligned crystal plane fringes with respect to each other [scale bars are: 20 nm for (a) and (b), 10 nm for (c) and (d), 5 nm for inset of (b)].



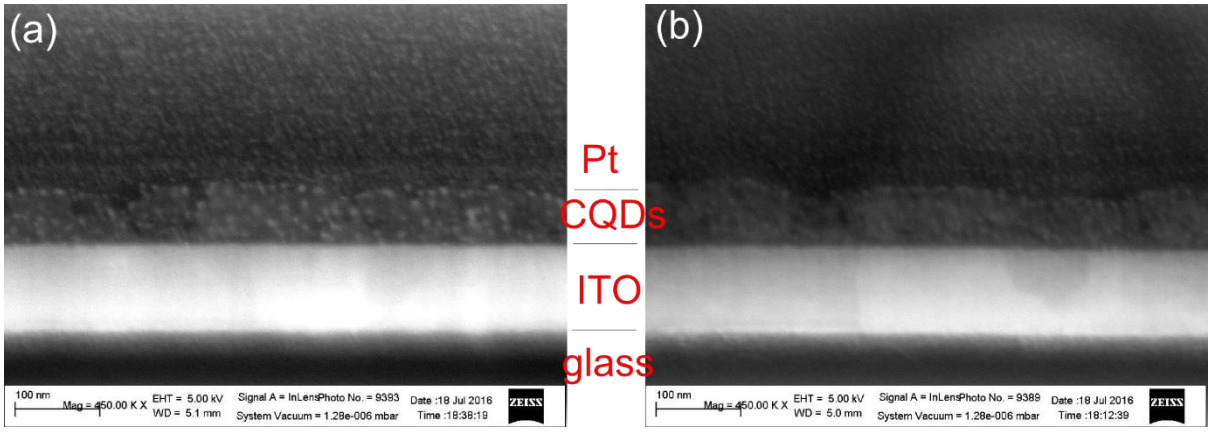
Supporting Figure S2. (a) Optical absoprtion of large (average diameter 5.7nm) PbS CQDs dispersed in toluene. (b) Optical absorbance of films of large PbS CQDs processed with TBAI and TMS.



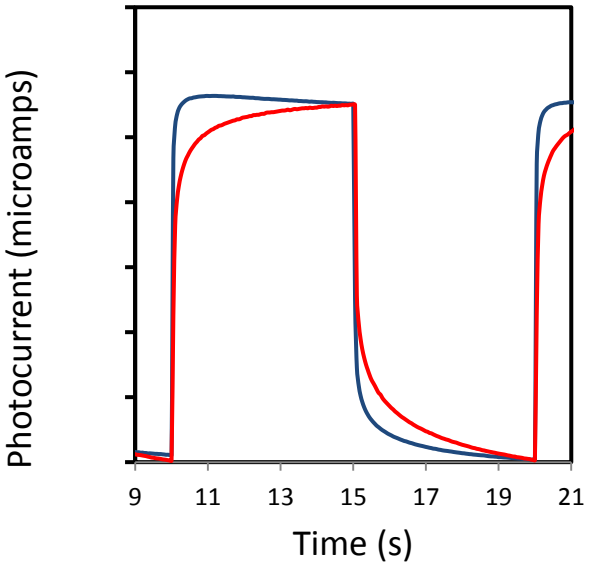
Supporting Figure S3. S2s spectra of the the TBAI, TMS (1%) and as-made (oleic acid, no further ligand was applied) films (backgrounds have been offset for clarity of presentation). Apart from the main peak at 225.2 eV attributed to PbS, a second peak/shoulder appears at 227.2 eV for the TMS-treated film.



Supporting Figure S4. XPS spectra of oleic acid capped CQD (untreated) film. (a) Pb4f spectrum with a $4f_{5/2}$ component at 137.6 eV assigned to PbS, and a second $4f_{5/2}$ (and rather strong) component at 138.5 assigned to Pb-oleate. (b) O1s spectrum with components at 529.3 eV, 531.05 eV, 532.3 eV assigned respectively to PbO, Pb-oleate, and -COO-. The presence of PbO may originate from residual PbO precursor from the CQD synthesis. We note the absence a component at 533.8 eV that would be assigned to -OH, that may indicate that all present oleic acid moieties have been deprotonated.



Supporting Figure S5. SEM images of cross sections (prepared with FIB) of CQD thin films fabricated in a layers-by-layer (LBL) with (a) TBAI and, (b) with TMS. Both samples were made via 4 applied CQD layers. The CQD films are on glass/ITO substrates and -for measurement purposes- have been covered with a thick layer of Pt. Scale bars are 100 nm.



Supporting Figure S6. Normalized data of Figure 4d of main paper further illustrating that the photoresponse of TBAI-treated (blue trace) CQD films is faster compared to the TMS-treated films (red trace)