Electronic Supplementary Information (ESI)

Towards Scalable Synthesis of High-quality PbS Colloidal Quantum Dots for Photovoltaic Applications

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Content

1. The precipitates of PbS QDs synthesized at 19.8 M

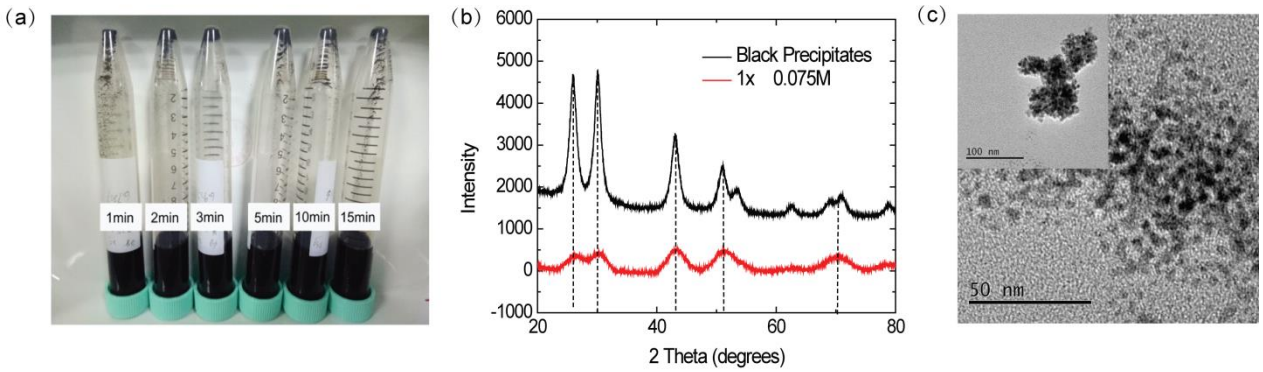
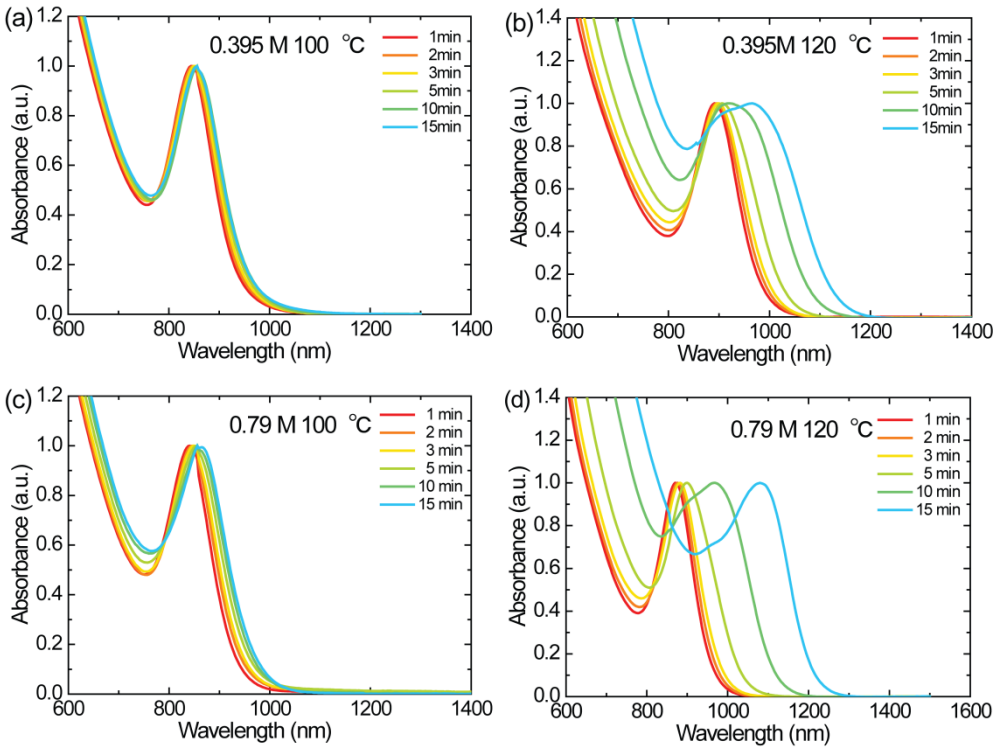


Figure S1. (a)The precipitates of PbS QDs synthesized at 19.8 M. The reaction temperature is 100°C. (b) The X-ray diffraction patterns of black precipitates and PbS QDs synthesized at conventional concentration. (c) The Transmission electron microscope images of black precipitates.

2. UV-vis absorption



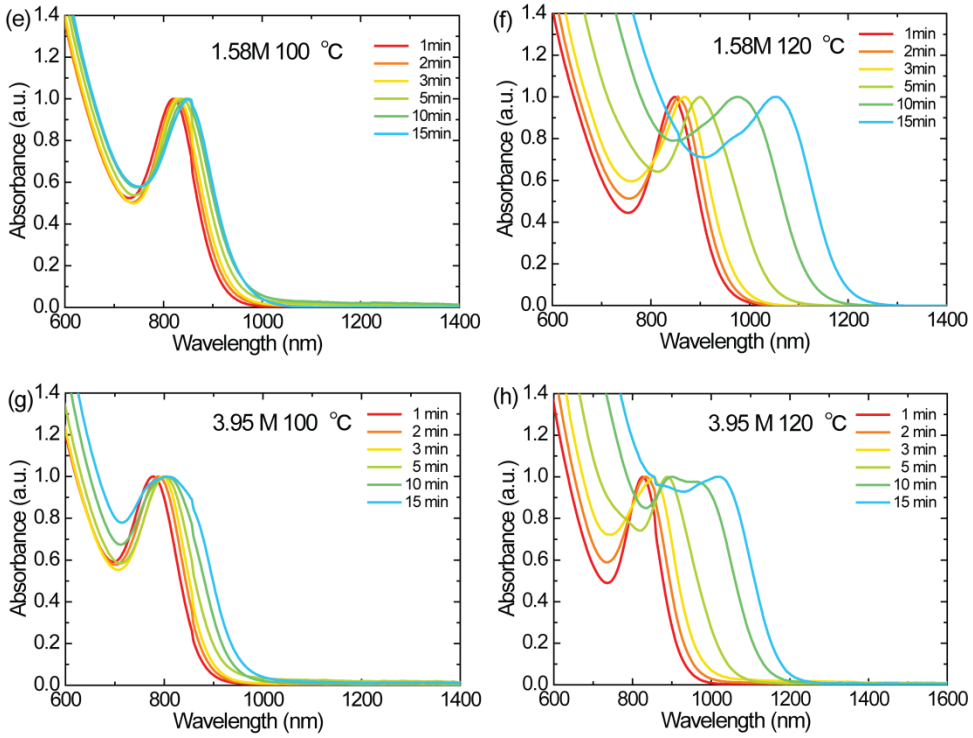


Figure S2. Ultraviolet-visible-near-infrared absorption spectra of PbS QDs synthesized using different precursor concentrations: (a)-(b): 0.395 M. (c)-(d): 0.79 M. (e)-(f): 1.58 M, (g)-(h): 3.95M.

3. The yield of PbS QDs

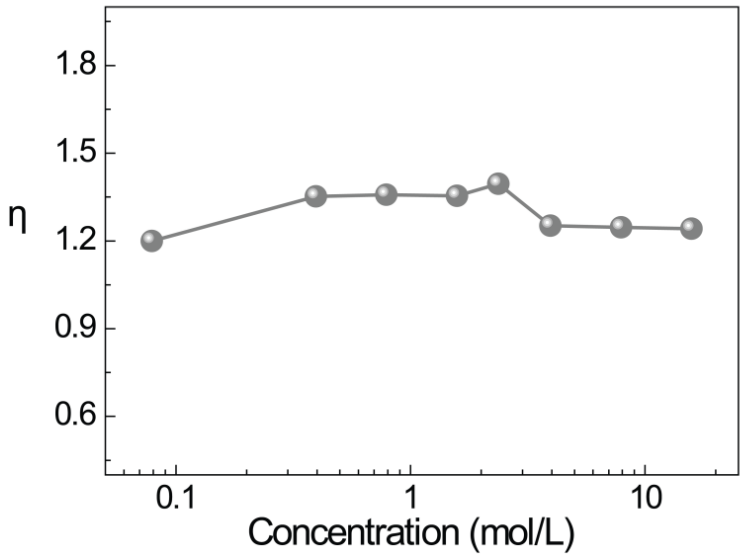


Figure S3. The yield of PbS QDs synthesized with different precursor concentrations.

4. The full-width at half-maximum (FWHM) of the Gaussian fitting

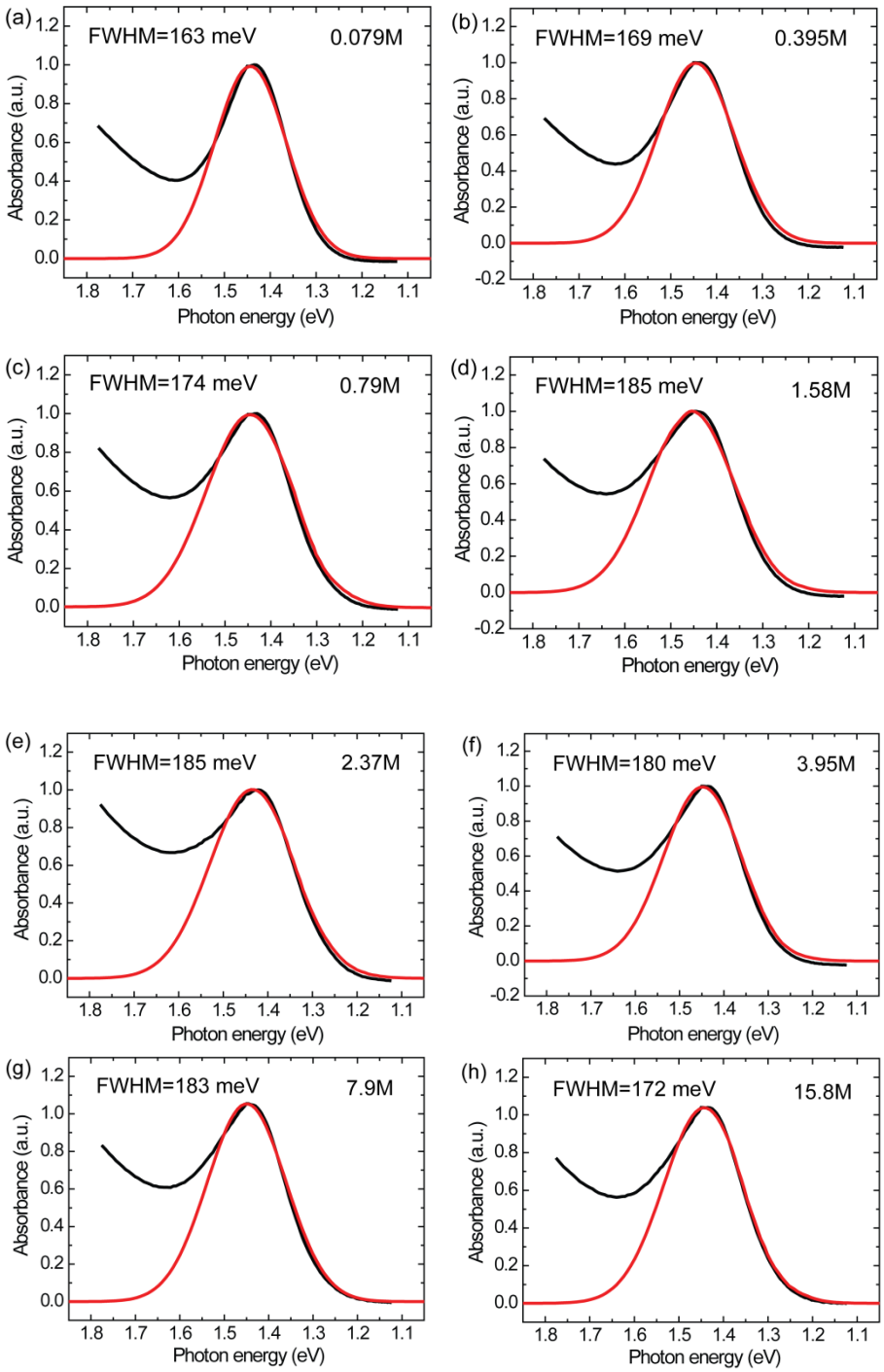


Figure S4. Normalized optical absorbance of PbS QDs synthesized under different precursor concentrations.

5. X-ray diffraction patterns

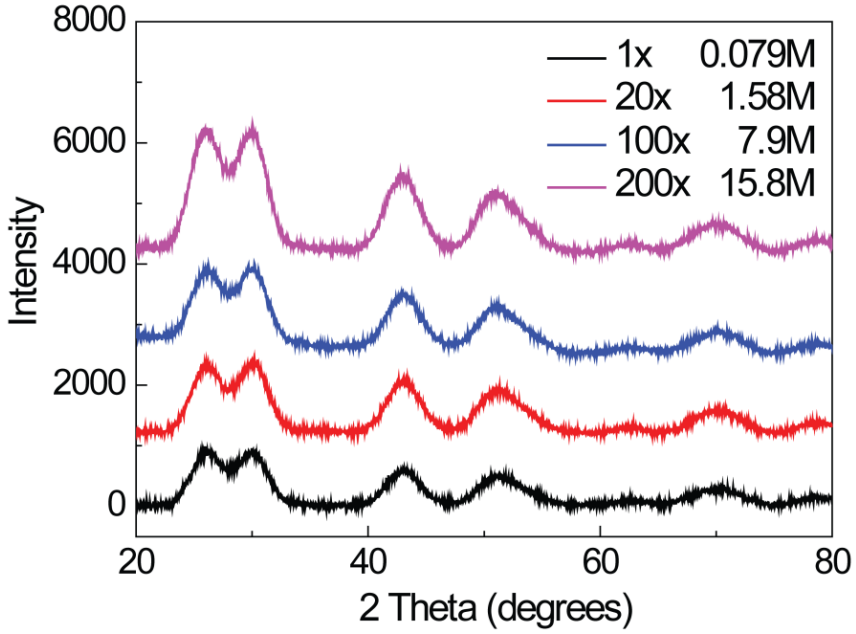


Figure S5. X-ray diffraction patterns of the oleic acid capped PbS QDs synthesized with different precursor concentrations.

6. X-ray photoelectron spectroscopy

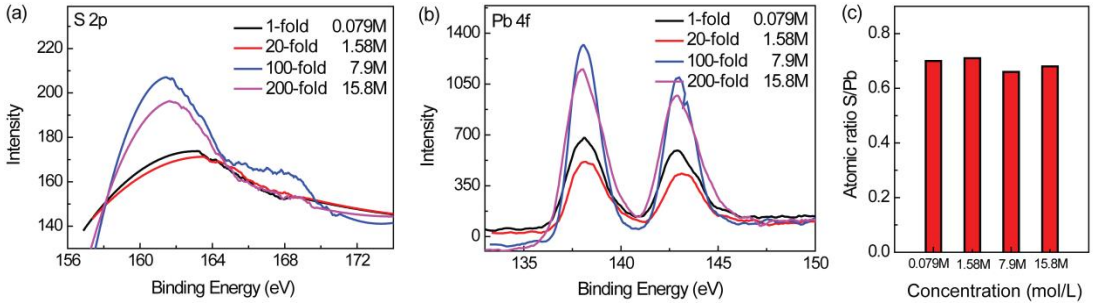


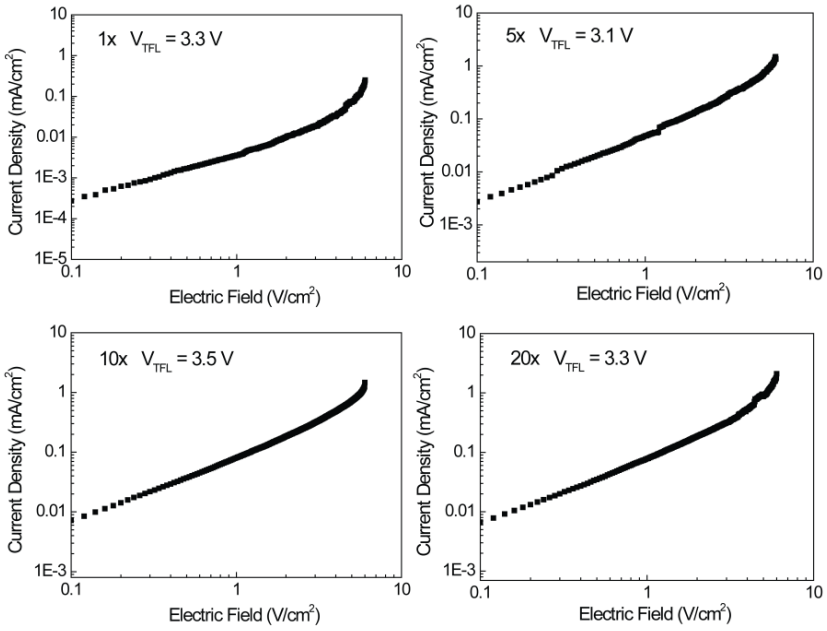
Figure S6. XPS analysis results of PbS QDs synthesized with different precursor concentrations.: (a) S 2p spectra of as-synthesized QDs. (b) Pb 4f spectra of as-synthesized QDs. (c) S/Pb atomic ratio of PbS QDs synthesized with different precursor concentrations.

7. Space charge limited current method (SCLC)

Electron only devices were fabricated using device structure Ag/PbS-TBAI/Ag for the measurement of trap density. *J-V* curves of devices exhibiting space charge limited current (SCLC) in the presence of traps show two characteristic regions. At low bias, thermally generated charges outnumber the injected charge carriers, and the device follows Ohm's law ($J \propto V$). At a certain voltage V_{TFL}, the injected charges exceed the thermally generated charges and injected charges fill the trap states leading to trap-filling SCLC behaviors. The onset voltage V_{TFL} is linearly proportional to the density of trap states N_{traps}:

$$\mathrm{V}_{TFL} = \frac{eN_{traps}d^2}{2\epsilon\epsilon_0}$$

In Figure S7, the VTFL for PbS-TBAI synthesized with different precursor concentrations are 3.3 V (0.075 M), 3.1 V (0.395 M), 3.5 V (0.79 M), 3.3 V (1.58 M), 3.6 V (2.37 M), 3.4 V (3.95 M), 3.7 V (7.9 M) and 3.6 V (15.8 M), respectively. Therefore, with a thickness of 200 nm, the trap density of PbS QD film are 1.7 × 10^{17} cm^{-3}, 1.6 × 10^{17} cm^{-3} , 1.8 × 10^{17} cm^{-3} , 1.7 × 10^{17} cm^{-3} , 1.86 × 10^{17} cm^{-3} , 1.75 × 10^{17} cm^{-3} , 1.9 × 10^{17} cm^{-3} , 1.86 × 10^{17} cm^{-3} , respectively.



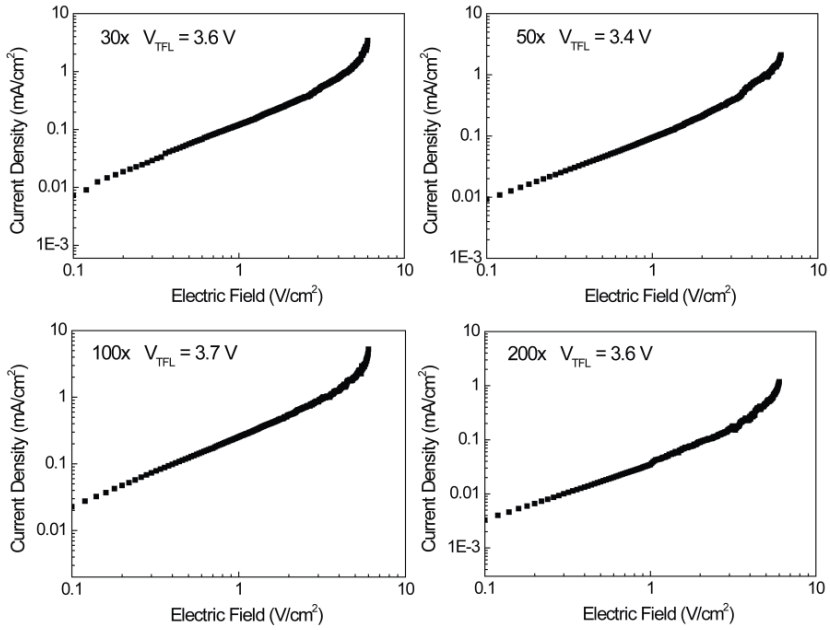


Figure S7. Space charge limited current using electron-only (Ag/PbS-TBAI/Ag).

8. Chemical quantities in PbS QDs synthesis.

Table S1: 1. Chemical quantities in PbS QDs synthesis

Lead precursor	Lead Concentration (M)	PbO Mass (g)	OA Mass (g)	ODE Mass (g)	$(TMS)_2S$ Volume (µl)
PbO	0.079 (1x)	0.223	3.5	10	105
	0.395 (5x)	1.115	3.5	10	525
	0.79 (10x)	1.115	3.5	5	525
	1.58 (20x)	1.115	3.5	2.5	525
	2.37 (30x)	1.115	3.5	1.7	525
	3.95 (50x)	1.115	3.5	1	525
	7.90 (100x)	1.115	3.5	0.5	525
	15.8 (200x)	1.115	3.5	0.25	525
	19.8 (250x)	1.115	3.5	0.2	525
$Pb(Ac)_2$	0.079 (1x)	0.38	3.5	10	105
	0.395 (5x)	1.9	3.5	10	525
	0.79 (10x)	1.9	3.5	5	525
	1.58 (20x)	1.9	3.5	2.5	525
	2.37 (30x)	1.9	3.5	1.7	525
	3.95 (50x)	1.9	3.5	1	525
	7.90 (100x)	1.9	3.5	0.5	525
	15.8 (200x)	1.9	3.5	0.25	525
	19.8 (250x)	1.9	3.5	0.2	525

9. Growth conditions of PbS QDs synthesized at different concentrations.

Table S2: Growth temperature and growth time to achieve the same size of PbS QDs with the absorption peak at 860 nm.

Lead Concentration (M)	Growth temperature (℃)	growth time (min)
0.079 (1x)	78	10
0.395 (5x)	93	5
0.79 (10x)	101	5
1.58 (20x)	113	3
2.37 (30x)	124	3
3.95 (50x)	140	1
7.90 (100x)	145	1
15.8 (200x)	140	1