Supporting Information

Impact of Background Oxygen Pressure on the Pulsed-Laser Deposition of ZnO Nanolayers and on Their Corresponding Performance as Electron Acceptors in PbS Quantum-Dot Solar Cells

Qian Dong,[†] Huiyan Liu,[‡] Yukihiro Hara,[§] Hannah E. Starr,[⊥] Jillian L. Dempsey,[⊥] and Rene Lopez[*, †, ‡, §]

[†]Department of Applied Physical Sciences, [§]Department of Physics and Astronomy, and [⊥]Department of Chemistry, University of North Carolina at Chapel Hill, Chapel Hill, North Carolina 27599, United States

[‡]School of Physical Science and Technology, Shanghai Tech University, 393 Middle Huaxia Road, Shanghai 201210, China

Corresponding author:

[*]E-mail: rln@physics.unc.edu.

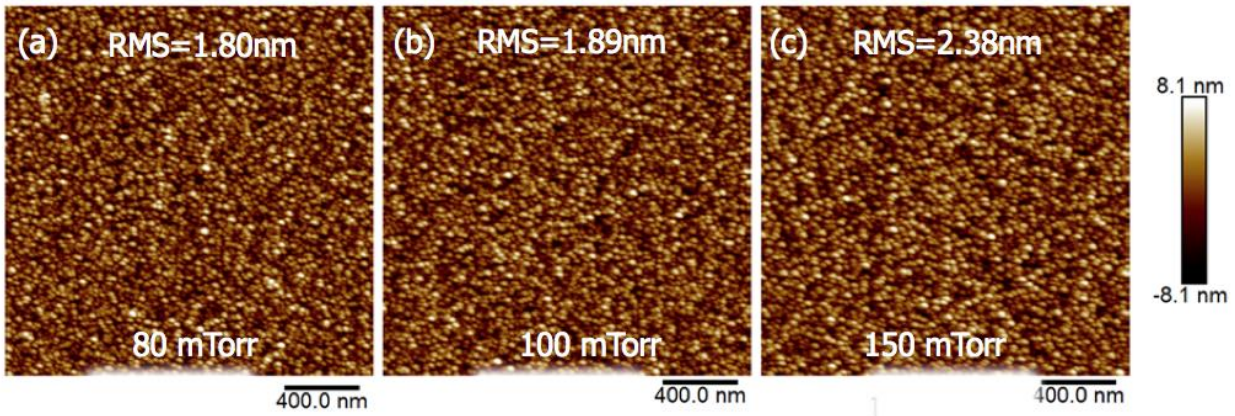

Figure S1. AFM images of films surface (scan size 2 μm × 2μm) for various deposited oxygen gas pressures, (a) 80 mTorr, (b) 100 mTorr, (c) 150 mTorr.

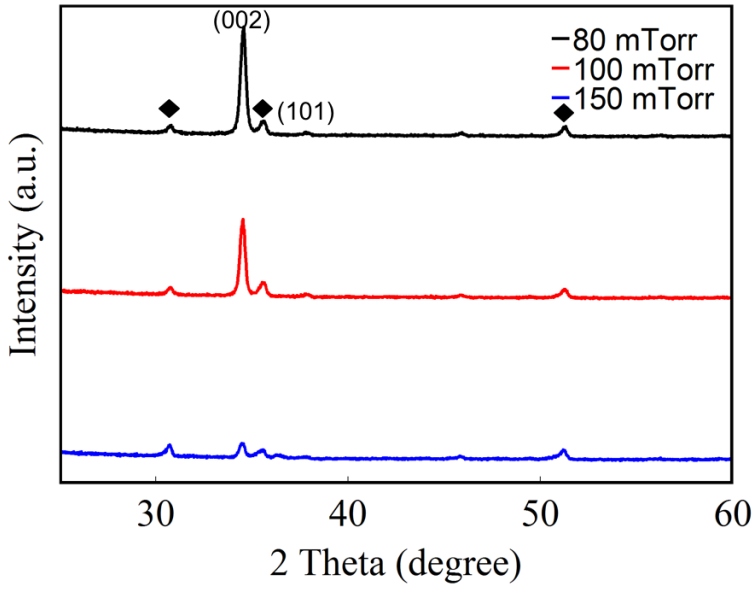

Figure S2. XRD patterns of ZnO films on ITO/glass substrate. ITO peaks are marked by black diamonds and ZnO peaks are identified by their Miller indices.

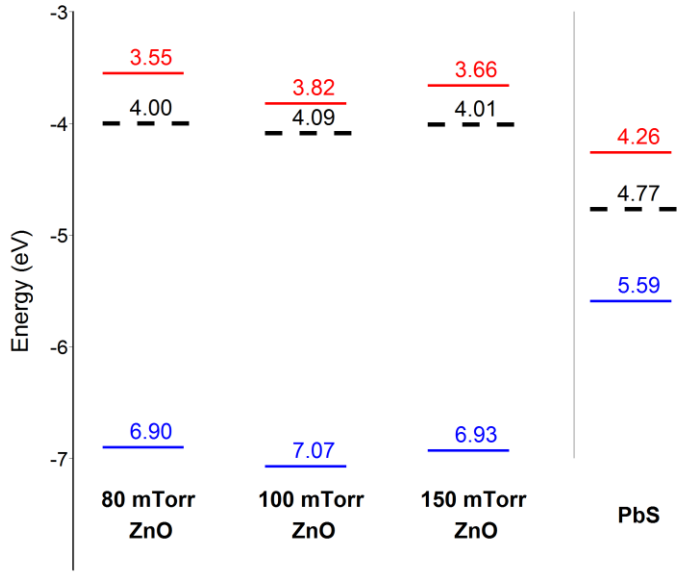

Figure S3. Energy levels with respect to vacuum for ZnO film deposited under different oxygen pressures, and PbS-TBAI film. The Fermi levels (E_F, dashed line) and valence band edges (E_V, blue lines) were determined by UPS. The conduction band edges (E_C, red lines) were calculated by adding the optical bandgap energy of 3.25 eV, 3.25 eV and 3.27 eV, respectively as determined from the first exciton absorption peak in the ZnO absorption spectrum to E_V.

Detailed PbS QD Synthesis.

PbS quantum dots were synthesized following a modified version of the procedure from Hines et al.[1] Briefly, lead (II) oxide (0.450 g, 2 mmol), oleic acid (1.13 g, 4 mmol), and octadecene (14.0 g) were combined in a 50-mL, three-neck round bottom flask and stirred under vacuum at 100 °C for one hour (flask 1). Simultaneously, hexamethyldisilathiane (TMS) (0.211 mL, 1 mmol) and octadecene (4 g) were combined in a 25 mL, two-neck pear flask and degassed (flask 2). The lead mixture (flask 1) was placed under positive nitrogen flow and heated to 100

°C. The TMS mixture (flask 2) was injected rapidly into the lead mixture (flask 1) and allowed to stir for 1.5 minutes before the QDs were quenched in acetone and left overnight. The QD mixture was dispersed in six 15 mL centrifuge tubes and centrifuged (8500 rpm) using acetone (three times, 10 min, 10 min, 8 min) and methanol (two times, 8 min and 6 min) as the antisolvents. QDs were resuspended in minimal pentane between centrifugation cycles. The yield was 550 mg, and the QDs were stored dry until use. PbS QDs absorbed at 896 nm with a diameter of 2.9 nm as determined by the sizing curve from Moreels, et al.[2, 3]

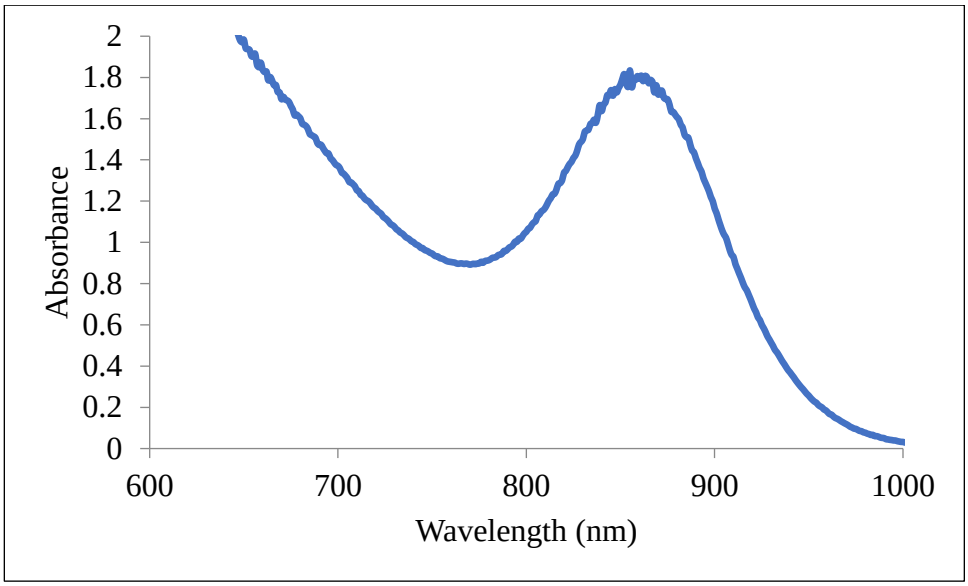

Figure S4. UV-Vis spectrum of 2.9 nm PbS CQDs.

References

(1) Hines, M. A.; Scholes, G. D. Colloidal PbS Nanocrystals with Size-Tunable Near-Infrared Emission: Observation of Post-Synthesis Self-Narrowing of the Particle Size Distribution. *Adv. Mater.* **2003**, *15*, 1844–1849.

(2) Moreels, I.; Lambert, K.; Smeets, D.; De Muynck, D.; Nollet, T.; Martins, J. C.; Vanhaecke, F.; Vantomme, A.; Delerue, C.; Allan, G.; Hens. Z. Size-Dependent Optical Properties of Colloidal PbS Quantum Dots. *ACS Nano* **2009**, *3*, 3023–3030.

(3) Kessler, M. L.; Starr, H. E.; Knauf, R. R.; Rountree, K. J.; Dempsey, J. L. Exchange Equilibria of Carboxylate-terminated Ligands at PbS Nanocrystal Surfaces. *Phys. Chem. Chem. Phys.*, **2018**, 20, 23649-23655.