

# **Effect of Perovskite Thickness on Electroluminescence and Solar Cell Conversion Efficiency**

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## **Experimental Methods:**

### **Device fabrication:**

**Cleaning:** A conducting glass coated with FTO (sheet resistance 14Ω/sq.) was etched with Zn powder and HCl (37%) to prevent direct contact between cathode and anode. The FTO glass was washed with deacon soap, deionized water, and ethanol for 15 min and then subjected to ultraviolet-ozone treatment for 15 min.

**Electron Transport Layer:** A thin planar layer of SnO<sub>2</sub> as electron transport layer was deposited using a 0.05 M solution of SnCl<sub>2</sub>·2H<sub>2</sub>O (Sigma Aldrich) in ethanol at 1500 rpm for 10s followed by fast pinning at 5000 rpm for 10 s and transferring to a 80 °C hot plate followed by annealing at 180 °C for 1 h. After cooling at room temperature, the SnO<sub>2</sub> substrates were transferred into a nitrogen glove box.

**Perovskite coating:** Perovskite solution was prepared using 1.1 M concentration of each PbI<sub>2</sub> (TCI, L0279), PbBr<sub>2</sub> (TCI, L0346), CsI (Sigma Aldrich) and FAI (GreatCell Solar ) in Dimethylformamide (DMF, Sigma Aldrich) and Dimethyl sulfoxide (DMSO, Sigma Aldrich) to maintain the stoichiometry of Cs<sub>0.20</sub>FA<sub>0.80</sub>Pb(I<sub>0.85</sub>Br<sub>0.15</sub>)<sub>3</sub>, DMF: DMSO ratio was 4:1. Perovskite solution was spin coated on SnO<sub>2</sub> substrates at 2000 rpm/10 s followed by 6000 rpm/30 s. Chlorobenzene as antisolvent was dripped in last few seconds. Substrates were preheated at 80 °C followed by 120 °C for 30 min.

**Hole Transport Layer:** 50 mg of spiro-MeOTAD (99%, Sigma Aldrich) was dissolved in chlorobenzene (700μl), and 4-tertbutylpyridine (20μl). 12.4 μL Li salt solution (26 mg lithium bis-(trifluoromethylsulfonyl) imide in 50 μl acetonitrile) and 28 μL Co salt solution (8.5 mg tris (2-(1H-pyrazol-1-yl)-4-tert-butylpyridine)-cobalt(III) tris(bis(trifluoromethylsulfonyl) imide in 28 μl acetonitrile) was then added and the whole solution was stirred for 15 min at 70 °C. This solution was spin coated on the perovskite layer at 4000 rpm for 30 s.

**Top Electrode:** Finally, 80 nm layer of Au was thermally evaporated at a rate of 0.3 Å s<sup>-1</sup>.

### **Characterizations:**

**XRD:** Thin film XRD data was collected by a Bruker D8 Advance diffractometer using CuKα radiation at a step size of 0.02° 2θ and counting time of 1 s per step.

**SEM:** SEM images were acquired on ZEISS Field Emission Scanning Electron Microscope.

**UV/Vis spectroscopy:** Optical absorption measurement was performed on UV-3600 (Shimadzu) spectrophotometer.

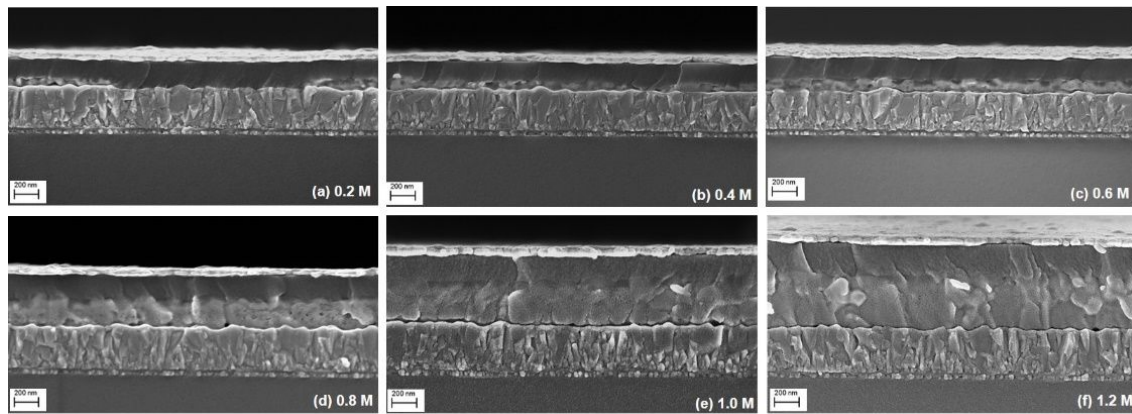
**PL measurements:** were done on Fluorolog, FL-1057, Horiba Instruments equipped with 450 W Xe Lamp. TRPL measurements were performed on same set up using single photon counting

controller with nano LED of 455 nm as excitation source, pulse duration 1.3 ns and repetition rate 1 MHz. The sample was FTO-SnO<sub>2</sub>-Perovskite.

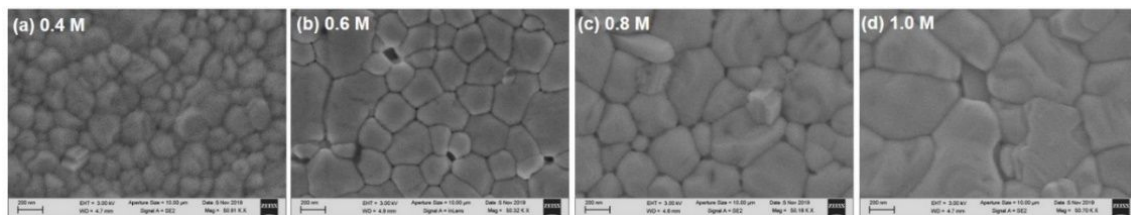
**PV measurements:** The J–V curves were measured by using a SanEI Electric, XEC-301S solar simulator under standard simulated AM1.5G illumination using a 0.09 cm<sup>2</sup> mask. Calibration was done using a standard reference silicon cell (Newport).

**EL measurements:** The characteristic current density–voltage–radiance was recorded with a Keithley 2612B source meter and an OceanOptics QE Pro spectrometer connected to an integrating sphere and operated using Ciemo LabVIEW software.

**EQE of solar cell:** The incident photon-to-current conversion efficiency (IPCE) was measured by using a PVE300 (Bentham), with xenon/quartz halogen light source in DC mode.



**Figure S1. (a-f) SEM cross section of different thickness devices.**



**Figure S2. Top view SEM showing increment in grain size from 0.4 M to 1.0 M**

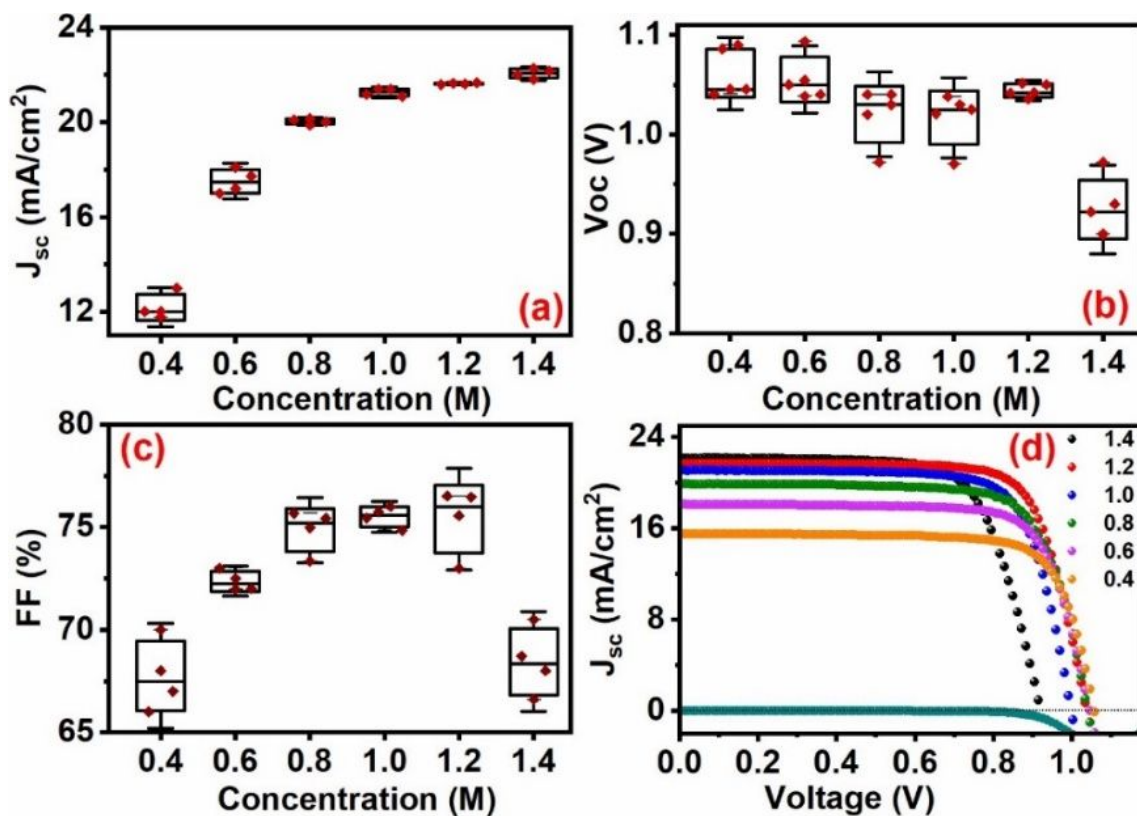


Figure S3. Statistical distribution of (a) current density ( $J_{sc}$ ), (b) open circuit voltage ( $V_{oc}$ ), (c) Fill factor (FF) and (d) J-V curve for different thicknesses.

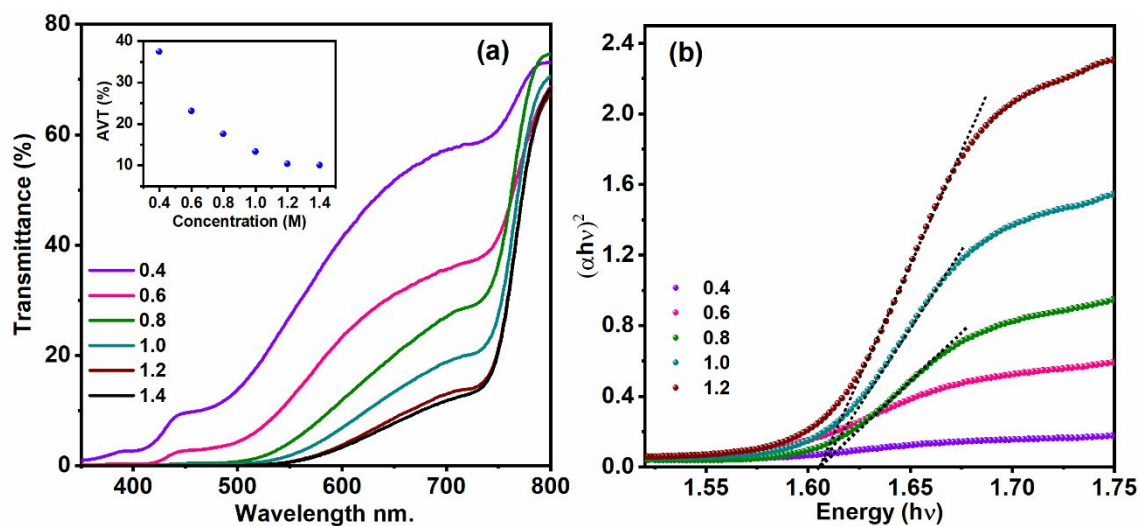


Figure S4. (a) Transmittance spectra showing variation in AVT (b) Tauc plot for different concentration

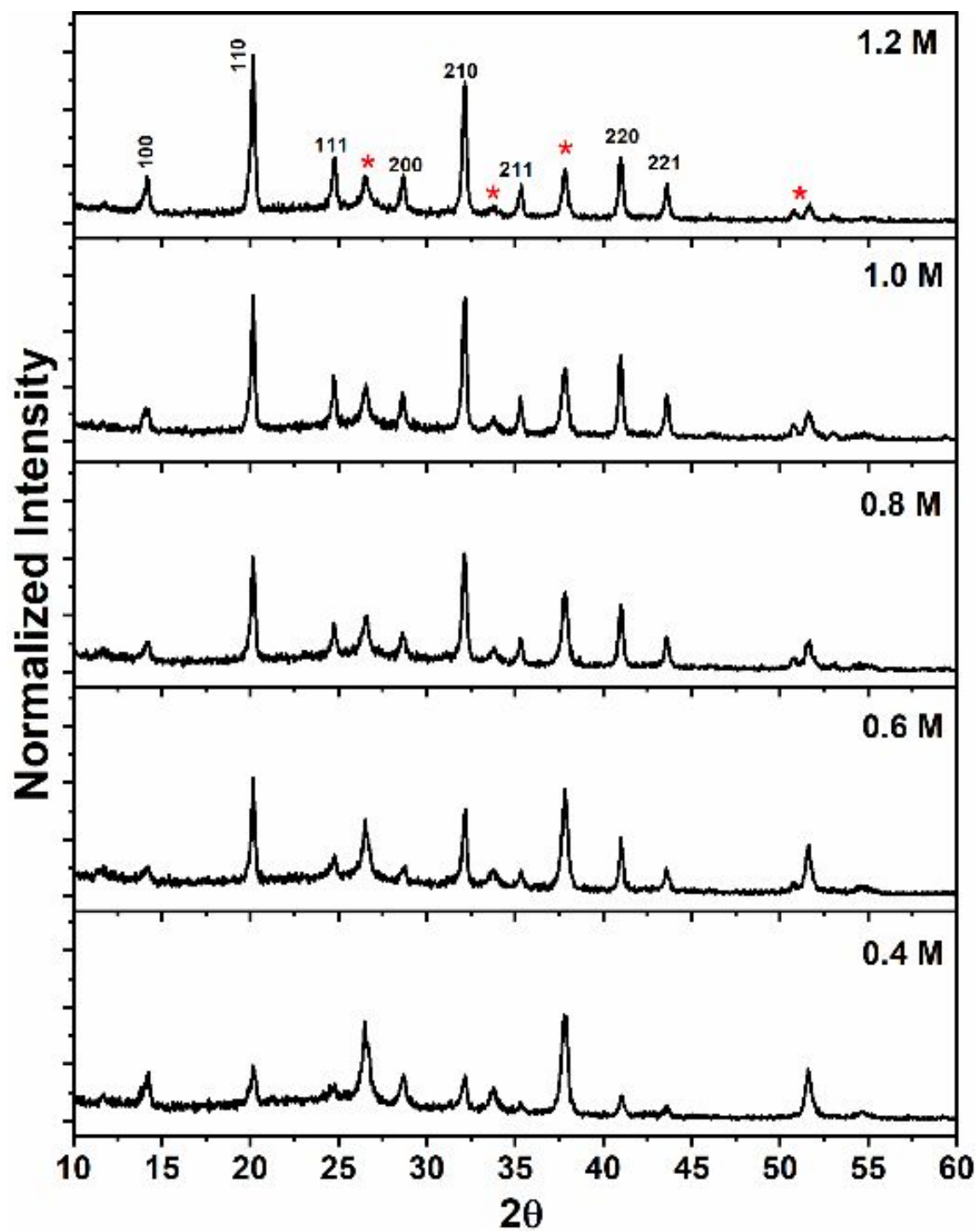


Figure S5. XRD peak showing the evolution and enhancement of Perovskite peak with increasing thickness. Red dots show peaks due to FTO substrate.

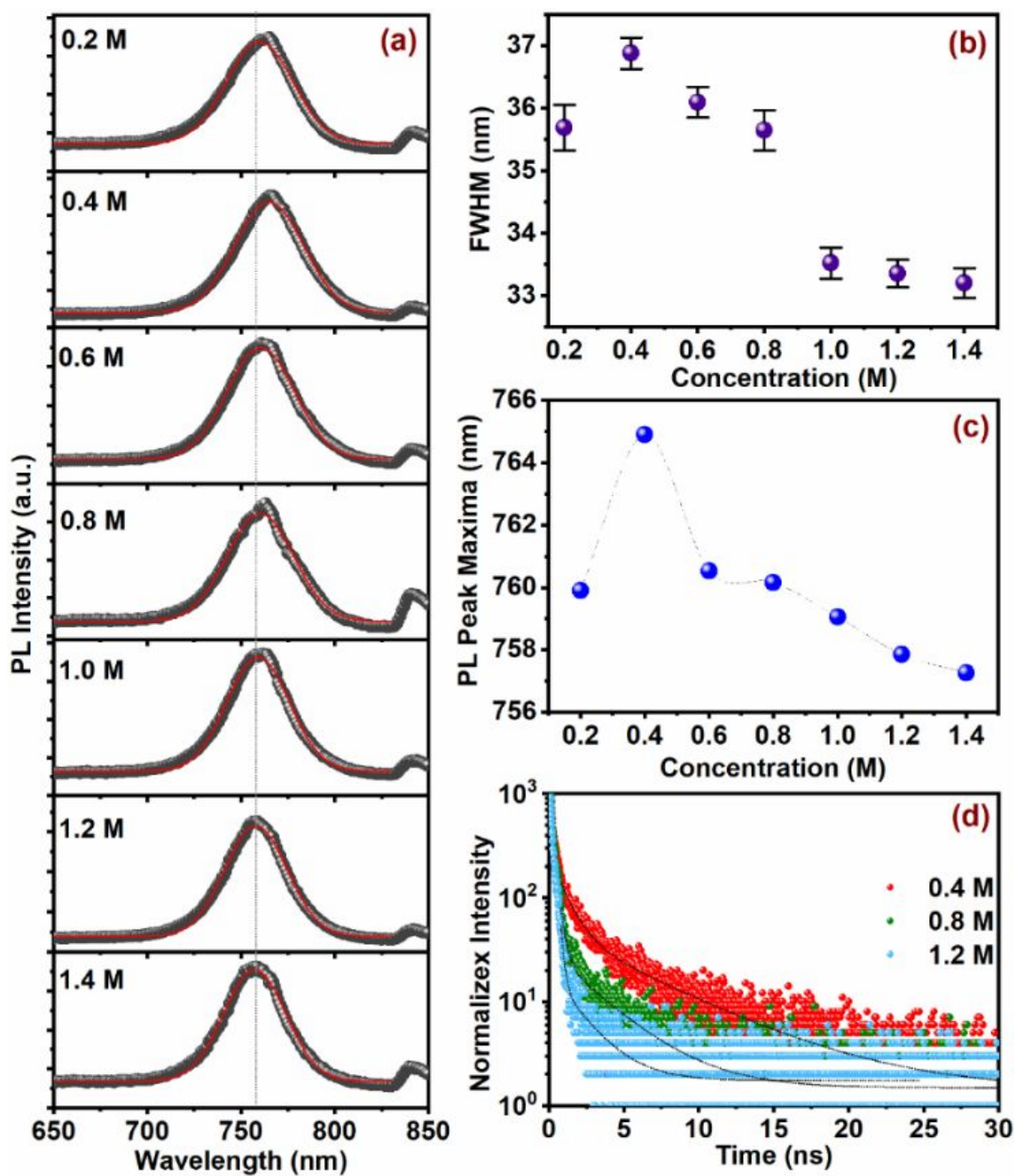
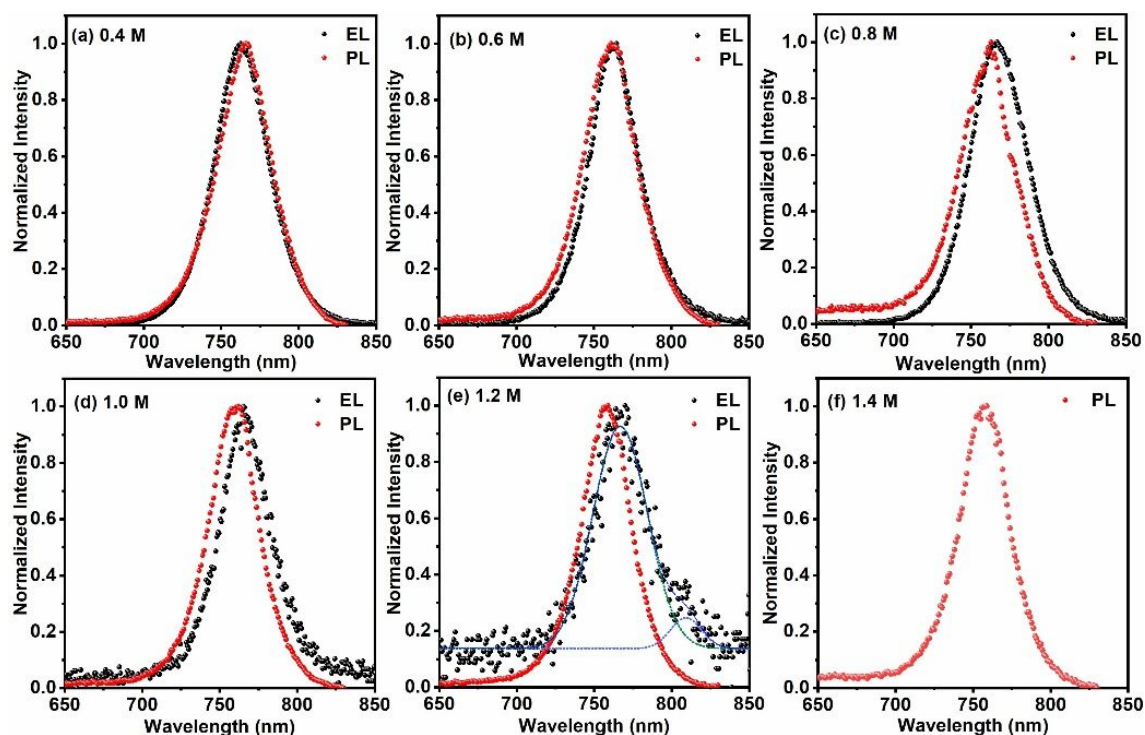
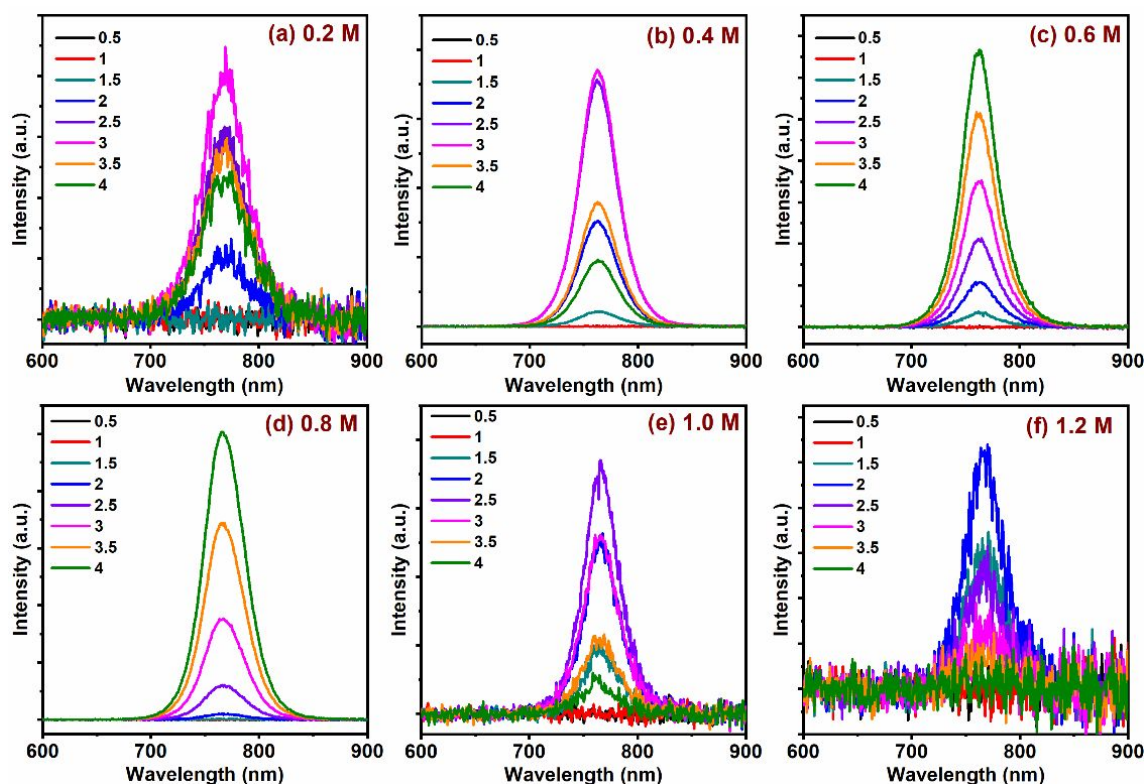


Figure S6. (a) Photoluminescence spectra with different concentration. Red line shows gaussian fitting. (b,c) variation of FWHM and peak maxima with concentration, respectively. (d) TRPL results for 0.4, 0.8 and 1.2 M. Black line shows exponential fitting.





**Figure S7.** Overlay spectra of EL and PL to show the shift in peak maxima for different thickness. No EL is observed for 1.4 M.



**Figure S8.** EL spectra at different applied bias (0.5-4V) for different thickness sample.