ARTICLES

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# Efficient $MAPbI_3$ solar cells made *via* drop-coating at room temperature

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**Abstract:** Here we demonstrate a room-temperature drop-coating method for $MAPbI_3$ films. By using low-boiling-point solvent, high-quality $MAPbI_3$ films were made by simply casting a drop of solution onto the substrate at room temperature. This approach took advantage of the synergistic effect of good wettability and volatility of the solvent, enabling high nuclei density and compact film at room temperature. The crystal growth in different solvents was *in-situ* observed by using optical microscope, which helped us to understand the mechanism for the formation of different film morphology. Perovskite solar cells gave a PCE of 18.21%.

**Key words:** perovskite solar cells; drop-coating; room temperature; low-boiling-point solvent; crystal growth

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## SUPPORTING INFORMATION

### Experimental section

### Materials

All materials were purchased and used without further purification unless specified. Dimethyl formamide (DMF) (99.8%), dimethyl sulfoxide (DMSO) (99.9%), and chlorobenzene (99.8%) were purchased from Alfa Aesar. Methylammonium iodide (MAI) (≥99.5%), MACl (≥99.5%), $PbI_2$ (≥99.99%) were purchased from Xi'an Polymer Light Technology Corp. Acetonitrile (ACN, anhydrous, 99.9%), Isopropanol (IPA) (99.5%), and Bathocuproine (BCP, 99%) were purchased from J&K. The methylamine (33% wt.% in absolute ethanol) was purchased from Energy chemical.

### Solution preparation

For DMF/DMSO-based solution, 0.6 M MAI and $PbI_2$ (1 : 1) were dissolved in a mixed solvent of DMF and DMSO (4 : 1, v/v) and stirred for 60 min at room temperature. MACl was added to the solution with a concentration of 5 mg $mL^{-1}$. Then the solution was stirred for 30 min at room temperature.

For MA(EtOH)/ACN-based solution, the preparation route is illustrated in Fig. S1. Firstly, 1.2 M MAI and 1.26 M $PbI_2$ (5% excess $PbI_2$ yields the best device performance) were dissolved in the MA(EtOH) solvent. The solution was stirred until MAI and $PbI_2$ were completely dissolved (~1 h), resulting in a clear, light yellow solution. After that, the solution was diluted to 0.4 M (for MAI) with ACN and 5 mg $mL^{-1}$ MACl was added. Note that high-concentration (> 1 M) solution should be prepared first before diluting with ACN, because low-concentration $MAPbI_3$ in MA(EtOH) yields a milky solution and poor film morphology.

### Device fabrication

Patterned ITO glass with a sheet resistance of 15 Ω $sq^{-1}$ was cleaned in detergent, deionized water, acetone, and isopropanol sequentially by ultrasonics and then treated with UV-ozone for 10 min. The PEDOT:PSS solution was prepared by mixing 1 mL PEDOT:PSS (Clevios P VP AI 4083) with 4 mL $H_2O$ and stirred for 1 h. PEDOT:PSS layer was deposited on ITO glass by spin coating the diluted PEDOT:PSS solution (3000 rpm for 30 s) and drying at 150 °C for 10 min. For MA(EtOH)/ACN based precursor, after cooling, 3 μL perovskite solution was dropped onto the center of the 1.5 × 1.5 $cm^2$ substrate. The solution can spread on the substrate spontaneously and dry naturally. After the film turns black (about 10 s), the substrate was transferred to a 110 °C hot plate for 1 min. For the conventional DMF/DMSO-based solution, the deposition route was the same except for the change of precursor volume to 0.5 μL. After cooling down, $PC_{61}BM$ in chlorobenzene (20 mg $mL^{-1}$), and BCP in IPA (0.5 mg $mL^{-1}$) were spin coated onto the perovskite films sequentially at 1000 rpm for 30 s and 4000 rpm for 30 s, respectively. Finally, 80 nm Ag was deposited onto the BCP layer through a shadow mask ($10^{-4}$ Pa).

### Materials characterization

Optical microscopy images were taken by using a LW200-3JT microscope. Absorption spectra for the films were recorded on a Shimadzu UV-1800 spectrophotometer. The SEM images were taken with a Zeiss Merlin field emission SEM (FE-SEM) operated at an accelerating voltage of 5 kV. XRD was performed on a RIGAKU D/MAX-TTRIII (CBO) with Cu Kα radiation.

### Device characterization

The effective area for the cells is 4 $mm^2$. *J–V* curves were measured by using a computerized Keithley 2400 Source-

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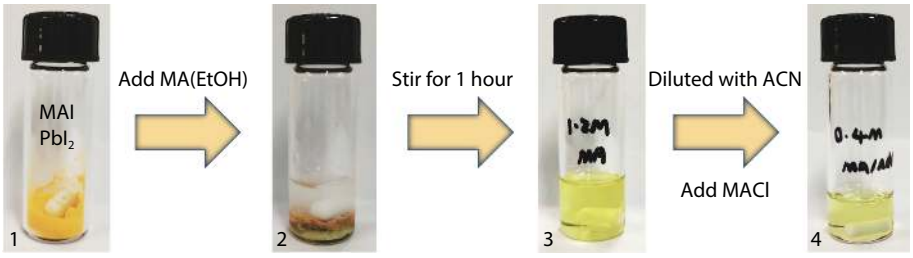


Fig. S1. (Color online) The solution preparation process and photographs of the samples.

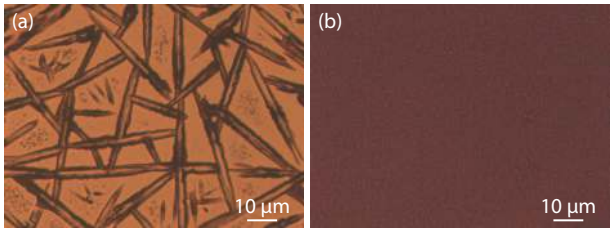


Fig. S2. (Color online) Optical microscope images for $MAPbI_3$ films made with (a) DMF/DMSO solution and (b) MA(EtOH)/ACN solution.

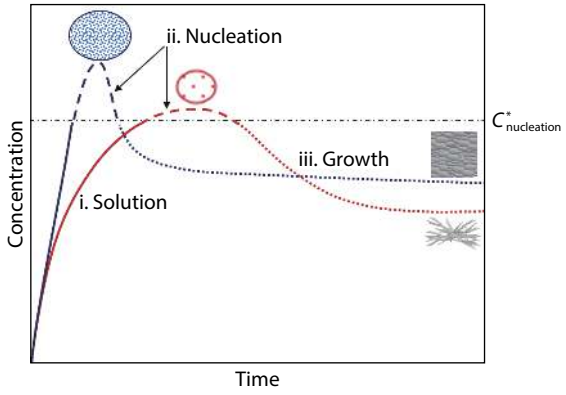


Fig. S3. (Color online) Lamer model to explain the nucleation and crystal growth in DMF/DMSO solution (red line) and MA(EtOH)/ACN solution (blue line). Solid line for solution stage; long dash line for nucleation stage; dot line for crystal growth stage.

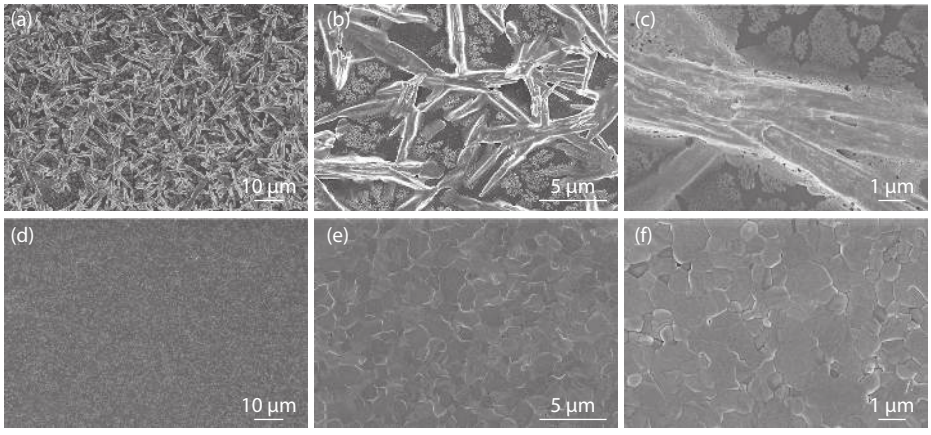


Fig. S4. (Color online) Top-view SEM images for perovskite films made from (a–c) DMF/DMSO solution, and (d–f) MA(EtOH)/ACN solution.

Meter and a Xenon-lamp-based solar simulator (Enli Tech, AM 1.5G, 100 mW $cm^{-2}$). The illumination intensity was determined by using a monocrystalline silicon solar cell (Oriel 91150, 2 × 2 $cm^2$) calibrated by NIM. The external quantum efficiency (EQE) spectra were measured by using a QE-R3011 measurement system (Enli Tech).