Supporting Information

Photo-Rechargeable Organo-Halide Perovskite Batteries

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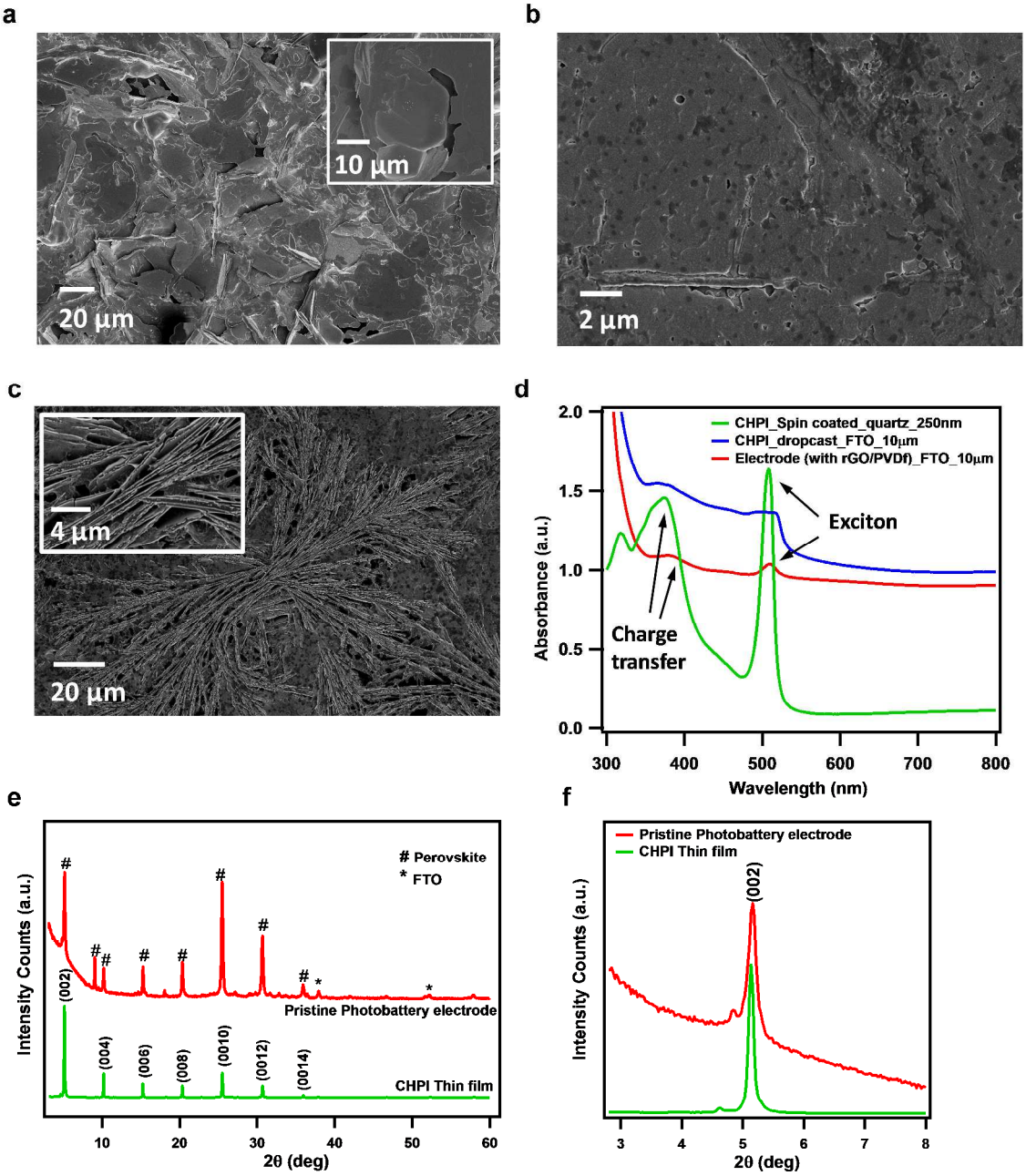

Figure S1: SEM images of ordinary (a) Drop casted and (b) Hot-casted films of pristine CHPI solution prepared on FTO substrates. The hot-casting process is adopted from a recently published paper[1]. Inset in (a) shows the hexagonal shaped bulk perovskite crystals[2,3]. (c) SEM image (top view) shows the vertical assembly of crystalline platelets of 2D perovskite on an FTO substrate. (d) Optical absorption spectra of perovskite photo-battery electrode (*B2*) prepared on pre-cleaned FTO substrates (red line). The absorbance of drop-casted and spin-coated films of pristine CHPI is added for comparison. (e) X-Ray Diffraction measurements of a pristine perovskite photo-battery electrode (*B2*) and pristine perovskite thin film (CHPI). (f) Close-up view of characteristic *(002)* peak for the

photo-battery electrode, which does not show any significant shift in the diffraction angle position when compared with the pristine perovskite thin film (CHPI).

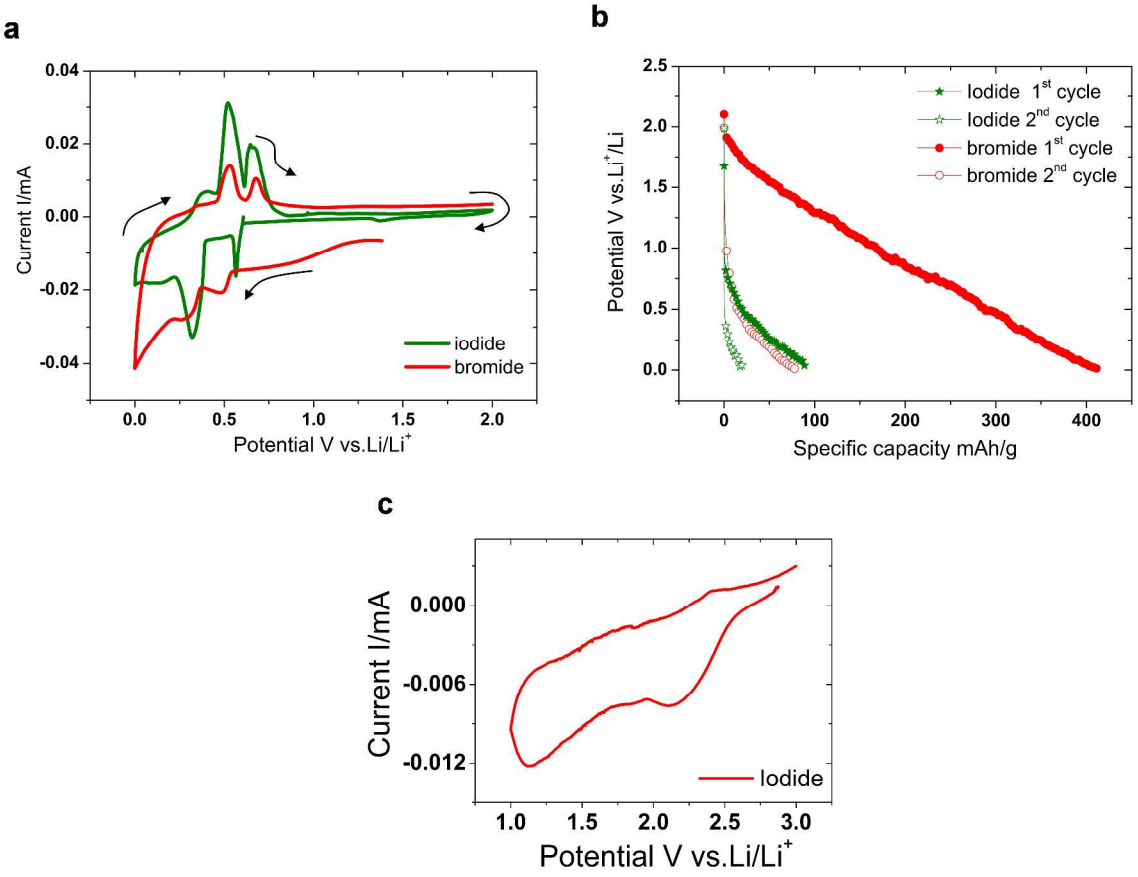

Figure S2: (a) Cyclic-voltammetry measurement of CHPI and CHPB 2D perovskites at 0.05 mV/s, measured in standard coin cell configuration under dark conditions. (b) First and second charge-discharge voltage profile of CHPI and CHPB 2D perovskites at ~30 mA/g and ~40 mA/g respectively from corresponding standard coin cells, the measurements were done in dark conditions. (c) Cyclic-voltammetry of CHPI photo-battery at 0.1 mV/s, measured in dark conditions.

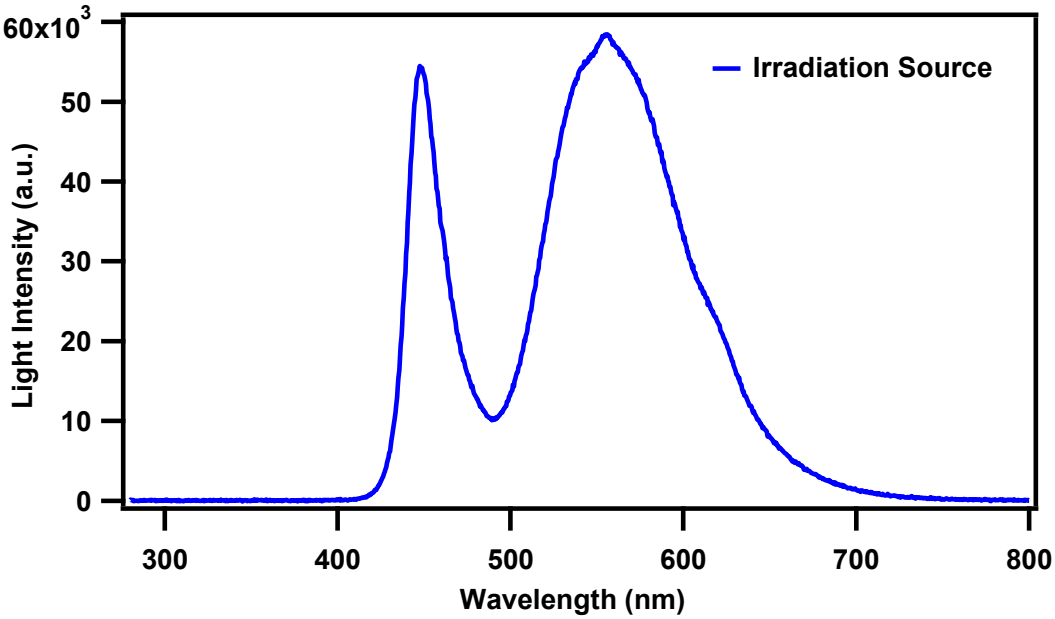

Figure S3: Spectrum of the LED irradiation source used for photo-charging.

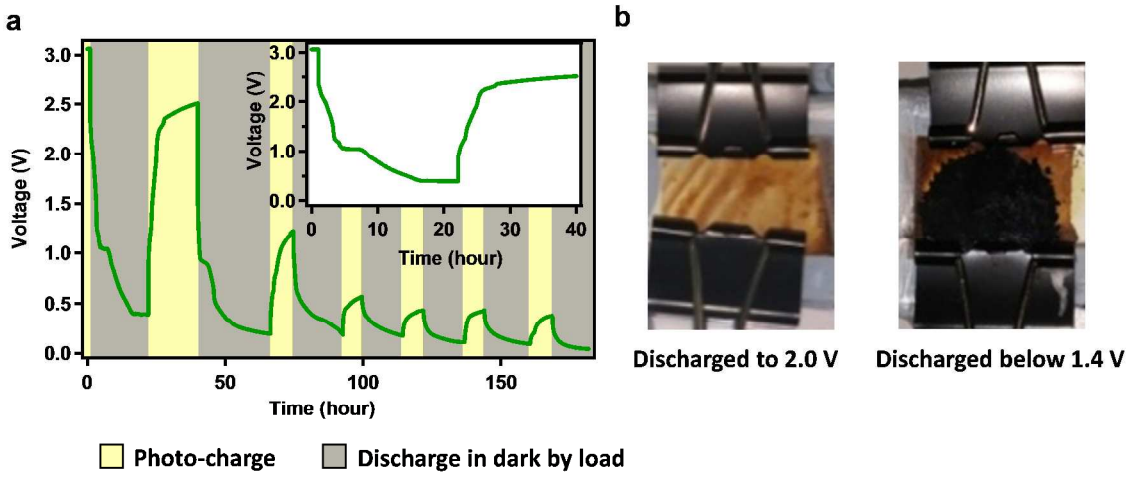

Figure S4: (a) Photocharge-discharge voltage profiles of a perovskite photobattery electrode with PCBM (*B1*) as additive. (b) Close-up photographs of electrodes when discharged to 2.0V and below 1.4V.

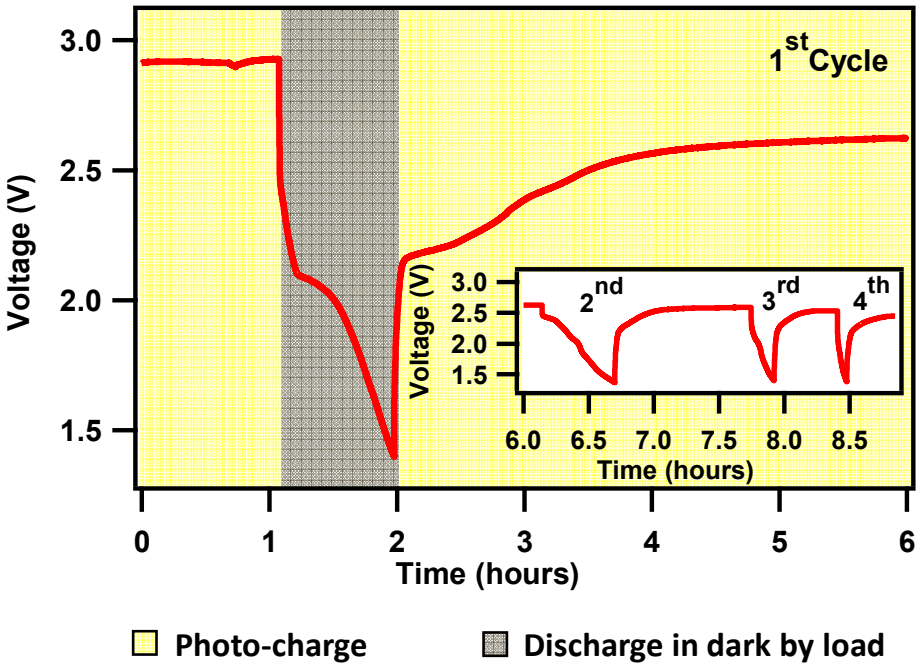

Figure S5: Photocharge-discharge voltage profiles of a perovskite photobattery electrode (*B2*) fabricated on a graphene based transparent electrode instead of FTO. The device shows a similar performance as ones with FTO as transparent substrate.

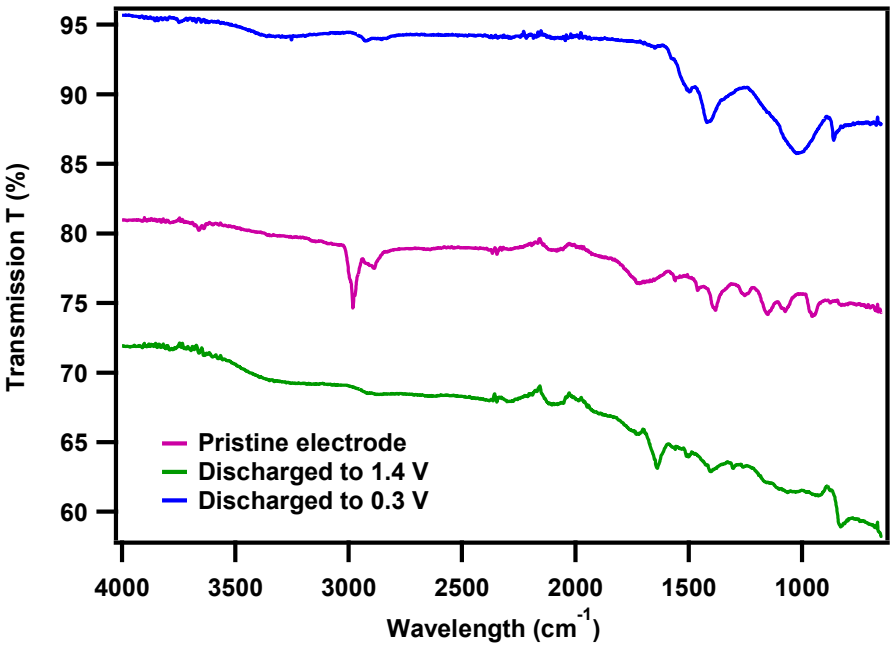

Figure S6: Fourier transform infrared (FTIR) spectroscopy using attenuated total reflection (ATR-FTIR) of a photo-battery electrodes discharged to 1.4 V and 0.3 V. FTIR spectra of pristine electrode is added for comparison. The electrode material is collected from the cell and analysed by ATR-FTIR. Occurrence of relatively strong peaks at 1016 cm^{-1} and 1423 cm^{-1} in case of photo-battery discharged to 0.3 V could be due to the electrochemical reduction of electrolyte[4].

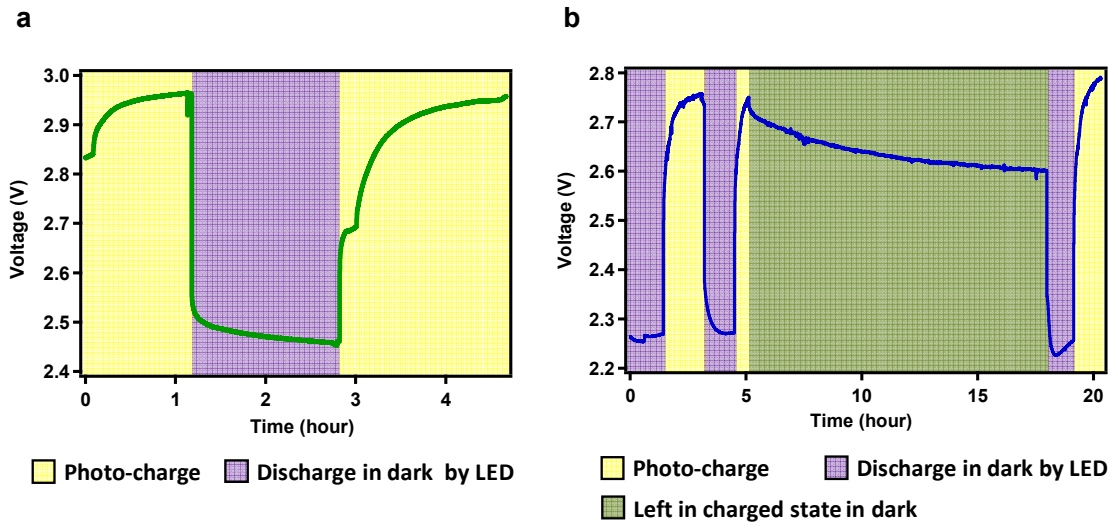

Figure S7: (a) Photocharge-discharge voltage curves of a perovskite photobattery electrodes (*B2*) when discharged by a white light LED in dark conditions. The LED has a turn-on voltage of about 2.2 V. This demonstrates that perovskite photobatteries are capable of powering an LED for more than an hour when fully photocharged. (**b**) Photocharge-discharge voltage curves of a perovskite photobattery electrodes (*B1*) showing the capacity retention of the photo-battery in charged state.

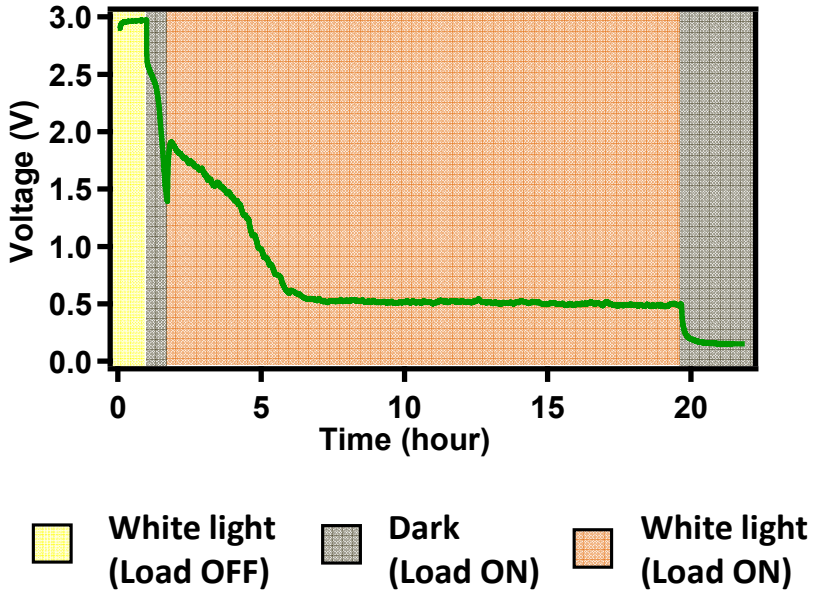

Figure S8: *In-situ* discharge potential from a device used for 5 cycles (shown in Fig 2C). Here, a fully photo-charged device was first discharged in dark conditions to 1.4 V (grey region), then the battery was illuminated, inducing photo-charging while discharging (red region). Since the device was already cycled previously, the performance was not as good as a new device (Fig 3A), but still, a stable potential value of ~ 0.5 V (η~ 0.011 %) is achieved for more than 13 hours. When the light was finally turned OFF (grey region), the voltage dropped to ~ 0.15 V (grey region).

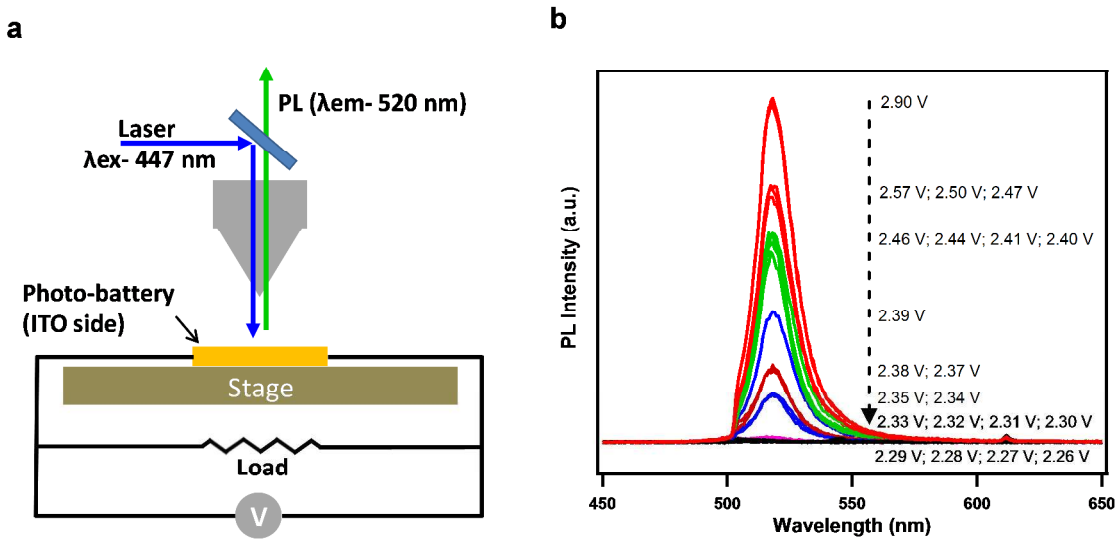

Figure S9: (a) Shows the confocal PL set-up designed to measure the in-situ PL of perovskite films (*B2*) while discharging them through a resistive load (21.5 kΩ). (b) Exciton PL spectra collected against the corresponding values of the open circuit voltage (OCV) of the photo-battery.

References:

(1) Nie, W.; Tsai, H.; Asadpour, R.; Blancon, J.-C.; Neukirch, A. J.; Gupta, G.; Crochet, J. J.; Chhowalla, M.; Tretiak, S.; Alam, M. A.; Wang, H. L.; Mohite, A. D. *Science* **2015**, *347*, 522-525.

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(4) Zhuang, G. V.; Yang, H.; Blizanac, B.; Jr. Ross, P. N. *Electrochem. Solid-State Lett.* **2005**, *8*, A441-A445.