Electronic Supplementary Material (ESI) for Energy & Environmental Science.

Supporting information

Frustrated Lewis Pair-Mediated Recrystallization of $CH_3NH_3PbI_3$ for Improved Optoelectronic Quality and High Voltage Planar Perovskite Solar Cells

Sagar Motilal Jain[1,†], Zhen Qiu[2], Leif Häggman[1], Mohammad Mirmohades[1], Malin B. Johansson[1], Tomas Edvinsson[2*] and Gerrit Boschloo[1*]

[1]*Uppsala University, Department of Chemistry-Ångström Laboratory, Physical Chemistry, Box 523, SE 751 20 Uppsala, Sweden*
[2]*Department of Engineering Sciences, Solid State Physics, Uppsala University, Box 534, SE 751 21 Uppsala, Sweden*

[†]*SPECIFIC, College of Engineering Engineering, Swansea University Bay Campus, Fabian Way, SA1 8EN Swansea, United Kingdom*

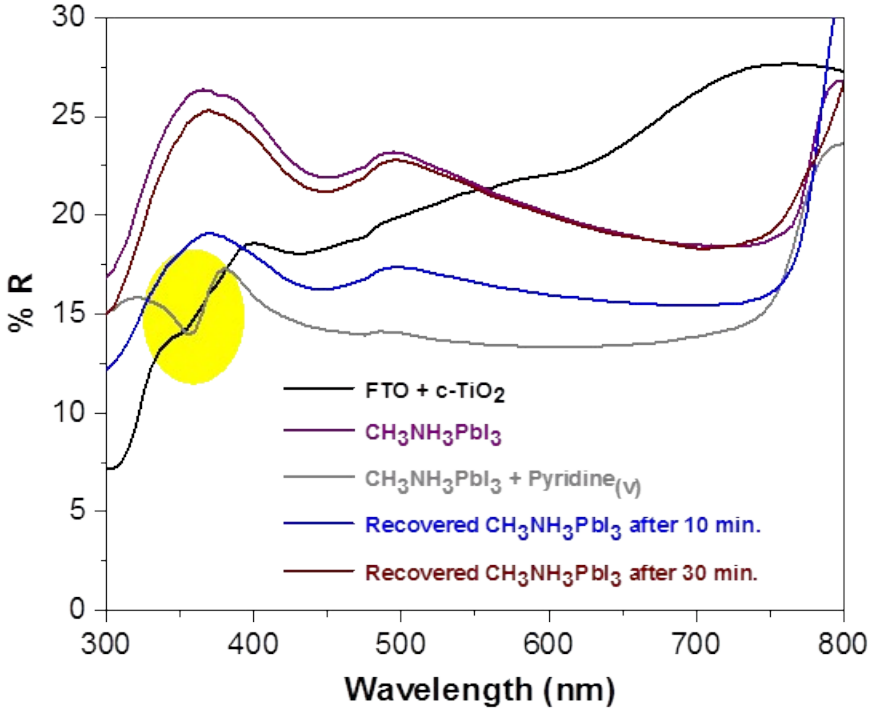

Figure S1: UV-Vis reflectance spectra or the recovery process of $CH_3NH_3PbI_3$ (film prepared on FTO + c-TiO_2) on exposure to pyridine vapor. The yellow marking indicates absorption due to the $PbI_2(pyridine)_2$ complex.

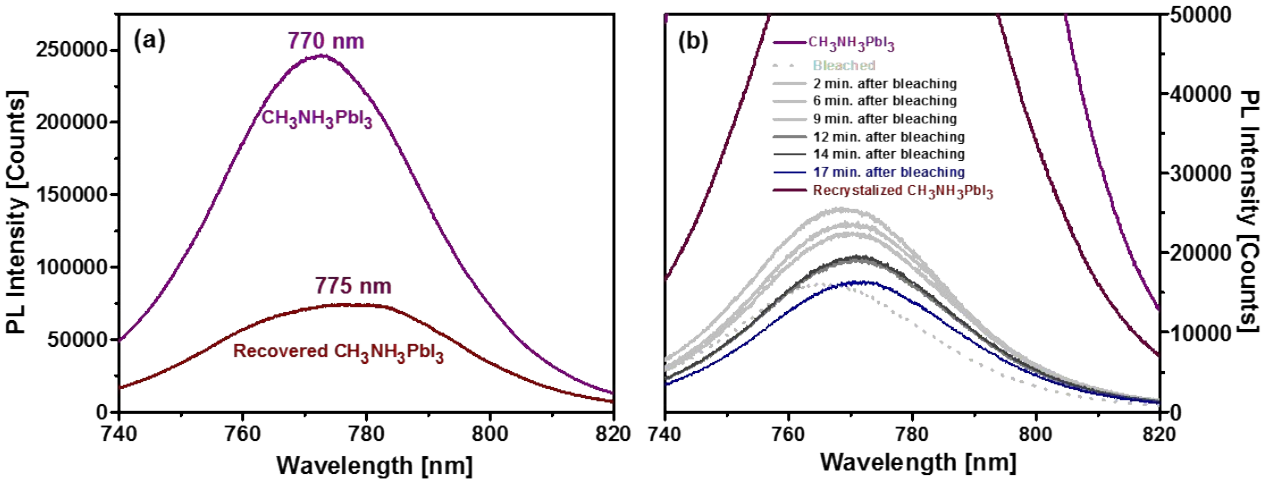

Figure S2: Steady state photoluminescence spectroscopy of pyridine-induced bleaching and recovery of $CH_3NH_3PbI_3$. (a) Photoluminescence spectra of $CH_3NH_3PbI_3$ before and after exposure to pyridine vapors, (b) PL-Spectroscopy of reversible bleaching process followed in time under air ambient conditions.

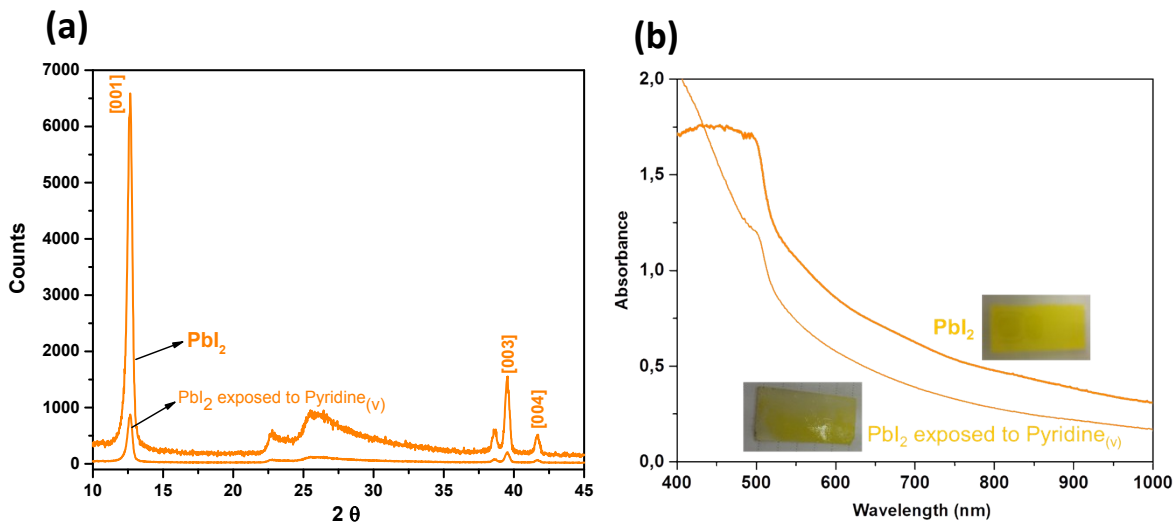

Figure S3: (a) XRD pattern and (b) UV-visible absorbance spectrum of PbI_2 and PbI_2 on exposure to pyridine vapor. Pyridine vapor exposure leads to loss of PbI_2 crystallinity and reduced absorption, the film becomes semi-transparent yellow colored (*inset* photographs).

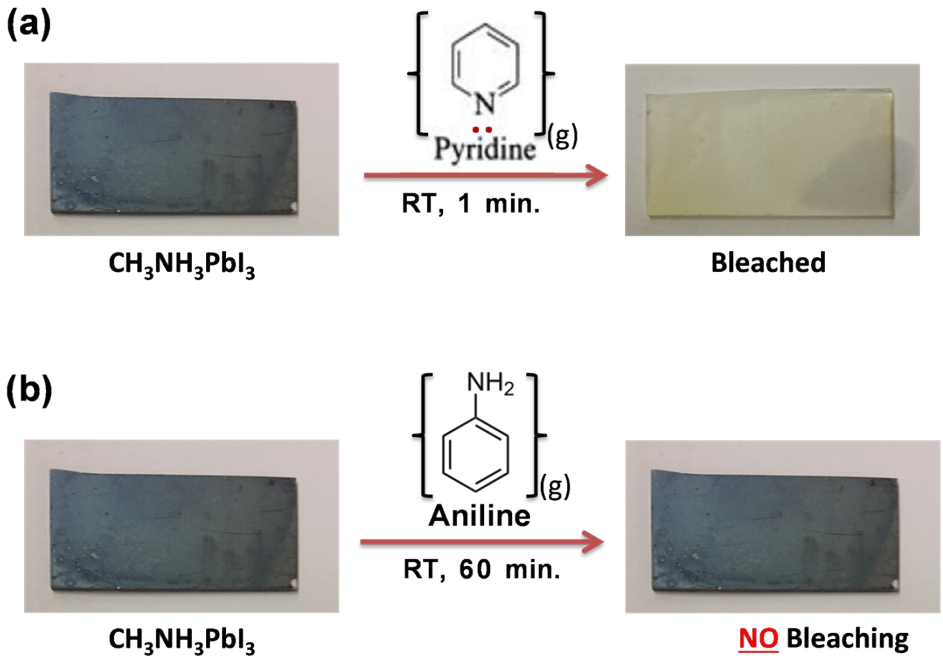

Figure S4: Experimental evidence for electron donation as reason for $CH_3NH_3PbI_3$ bleaching reaction. Pyridine is a relatively strong Lewis base, leading to availability of the free electron pair in the bleaching reaction with $CH_3NH_3PbI_3$, see photographs in (a). The aromatic group in aniline is electron withdrawing, strongly decreasing the Lewis basicity of the electron pair on the nitrogen, resulting in absence of reaction with perovskite (b).

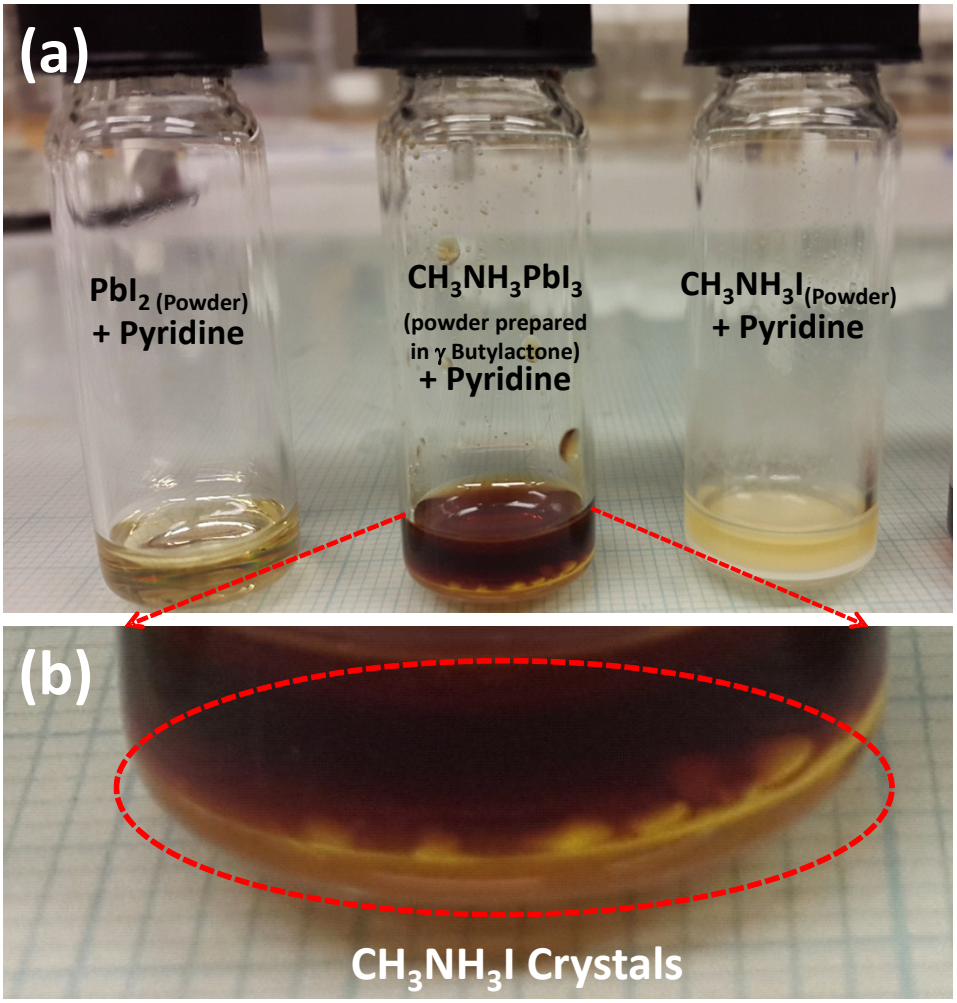

Figure S5: (a) PbI_2 powder is soluble in pyridine, $CH_3NH_3PbI_3$ as-prepared (powder scraped from substrate) dissolves partially in pyridine –white color crystals of CH_3NH_3I (result conclusion from further XRD analysis) are seen on the bottom. CH_3NH_3I powder dispersed in pyridine - the white powder is insoluble and precipitates on the bottom. (b) Magnification of $CH_3NH_3PbI_3$ in pyridine.

Table S2: Photovoltaic performance statistics of 48 - $CH_3NH_3PbI_3$ planar thin film solar cells made via pyridine bleaching treatment. (FS – Forward scan; BS – Backward scan). All devices measured at a constant scan rate and intensity of 30 mV s^{-1} and 1000 wm^{-2} respectively from scan direction of the open circuit to the short-circuit and reverse. (Note that we have selected the best statistics of 48 devices out of 74 solar cells prepared)

Number of Solar cells	Scan direction	*J*sc ($mAcm^{-2}$)	Voc (V)	FF	PCE (%)
Pyridine treated $CH_3NH_3PbI_3$					
4	FS BS	20.0 19.8	1.00 0.98	0.65 0.62	13.0 12.3
6	FS BS	21.8 21.8	1.12 1.10	0.72 0.69	17.5 16.6
10	FS BS	22.0 22.0	1.15 1.15	0.73 0.71	**18.5** **17.9**
11	FS BS	22.0 22.0	0.99 0.96	0.72 0.69	15.7 15.0
17	FS BS	20.2 20.0	1.01 0.99	0.70 0.68	15.1 14.6

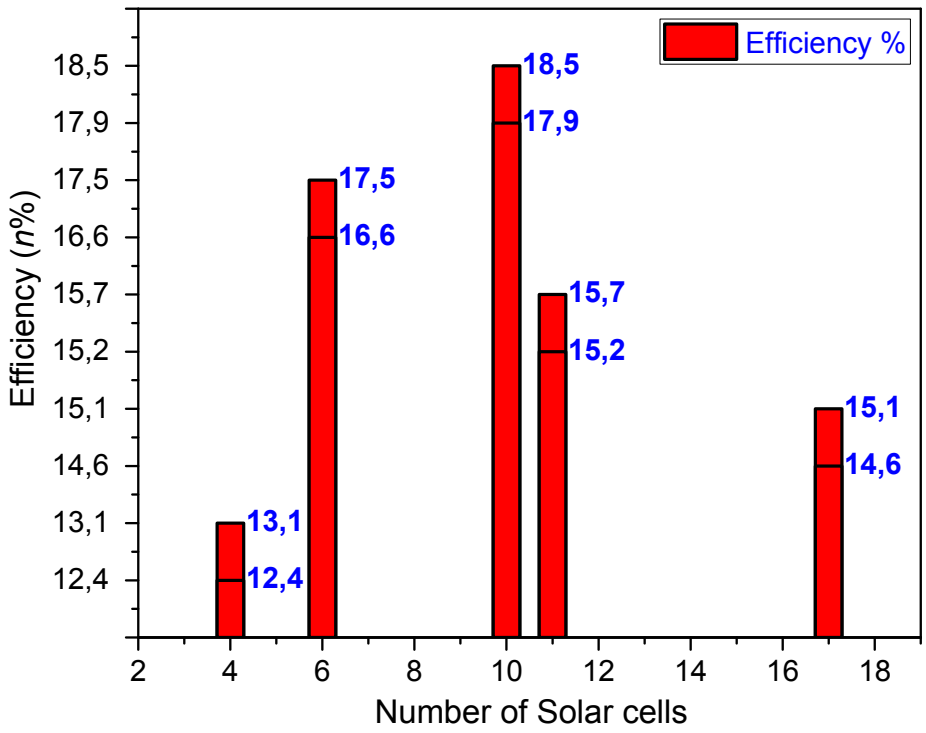

Figure S6: Photovoltaic performance statistics of the 48 planar solar cell devices on different scan direction. All devices measured at constant scan rate and intensity of 30 mV S^{-1} and 1000 W m^{-2} respectively. (On the bar, the top numbers represent solar cell performance on forward scan while the bottom numbers represent solar cell performance on backward scan)

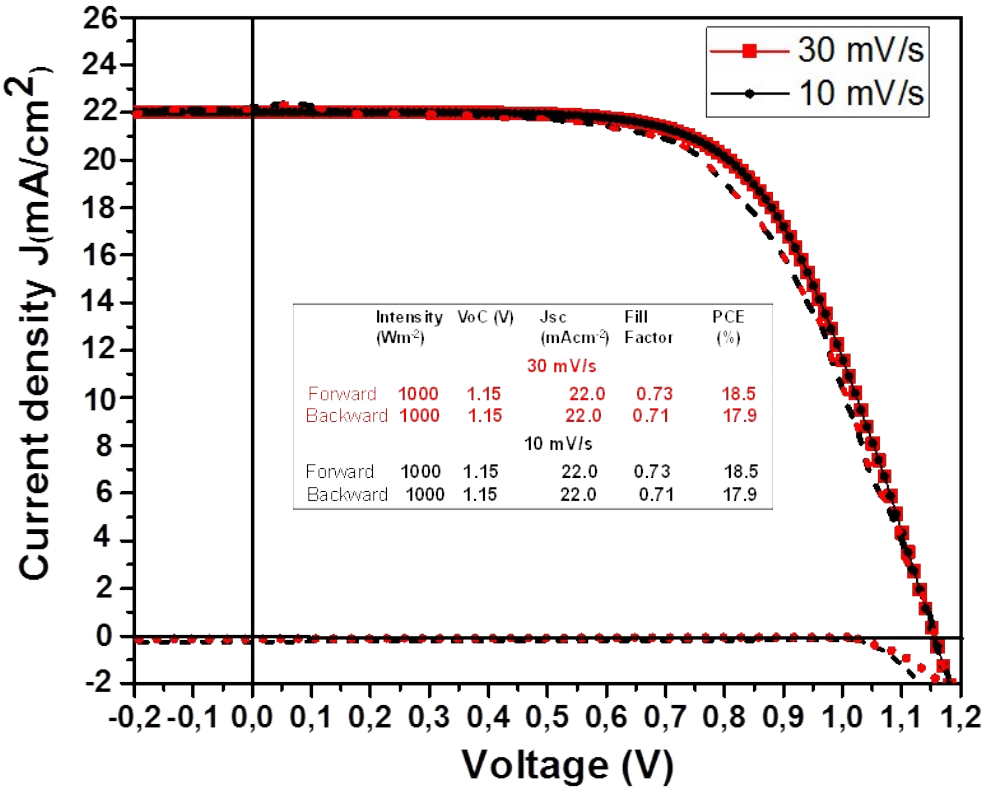

Figure S7: Device performance measured at variable scan rates of 30 mV/s and 10 mV/s and solar light intensity of 1000 W m^{-2}.