Supporting Information

Two-step MAPbl₃ deposition by Low-Vacuum Proximity-Space-Effusion for highefficiency inverted semitransparent perovskite solar cells

Emanuele Smecca,^a Vitantonio Valenzano,^b Salvatore Valastro,^{a,c} Ioannis Deretzis,^a Giovanni Mannino,^a Graziella Malandrino,^d Gianluca Accorsi,^e Silvia Colella,^f Aurora Rizzo,^e Antonino La Magna,^a Andrea Listorti,^{*e,g} Alessandra Alberti^{*a}

a CNR-IMM Zona industriale, Strada VIII 5, 95121, Catania, Italy

b Dipartimento di Matematica e Fisica "E. De Giorgi", Università del Salento, Campus Ecotekne, Via Arnesano, 73100 Lecce, Italy

c Dipartimento di Fisica e Astronomia, Università di Catania, Via S. Sofia 64, 95123 Catania, Italy

d Dipartimento di Scienze Chimiche. Università degli Studi di Catania. V.le A. Doria 95125 Catania. Italy

e CNR NANOTEC, Institute of Nanotechnology, c/o Campus Ecotekne, via Monteroni, 73100 Lecce, Italy

f CNRNANOTEC, Institute of Nanotechnology, c/o Department of Chemistry, University of Bari 'Aldo Moro', via Orabona 4, 70126 Bari, Italy

g Department of Chemistry, University of Bari "Aldo Moro", via Orabona 4, 70126, Bari, Italy *corresponding author

Email:alessandra.alberti@imm.cnr.it; andrea.listorti@uniba.it



Figure S1 XRD pattern collected for 30 min deposited Pbl2film as deposited and after annealed at 150°C for 1h. the XRD patterns collected before and after the annealing do not show any significant increase of the peak intensity confirming that the layer is already fully crystallized after deposition. The presence of some amorphous phase prior the annealing in fact, would increase the intensity of the peak due to the crystallization that would occur during the annealing at 150°C.



Figure S 2 (a) Polar figures acquired at fixed 2θ angle of 25.91° in samples deposited at different MAI time steps. Each circle in the polar frame corresponds to 10° in χ . (b) Representative spatial distribution of Lead (gray) and Iodine (green) in the PbI₂ lattice. Related octahedral configuration with respect to the [0001] growth direction for PbI₂ lattice.



Figure S 3 Pb 4f spectral region from high resolution XPS analyses performed on 30 minutes deposited PbI_2 (a) and $MAPbI_3$ (b). The deconvolution of the peaks does not show sign of metallic lead but rather a double peak centered at 138.6 eV ($Pb4f_{7/2}$) and 143.4 eV ($Pb4f_{5/2}$) typical of lead halides compounds. A slight shift at 138.2 ev ($Pb4f_{7/2}$) and 143.0 eV ($Pb4f_{5/2}$) can be seen in the MAPbI₃ sample due to the presence of MA⁺ cation inside the inorganic cage.

Table S1 Chemical analyses obtained from XPS analyses performed on 30 minutes deposited PbI_2 and $MAPbI_3$. It is worth to know the increasinfg of the C1s and N1s amount due to the presence of the MA⁺ cation inside the film. The Pb/I ratio is 0.51 for PbI_2 film and 0.35 for MAPbI₃ that are very close to the theoretical values.

	C 1s	<i>O1s</i>	N1s	Pb4f	I3d
Pbl ₂	32.5	9.6	-	19.3	38.4
MAPbl₃	43.3	29.8	4.8	5.8	16.1



Figure S 4 Energy Dispersive X-ray (EDX) spectroscopy performed on 30 minutes deposited PbI_2 on ITO/PTAA. In the inset we show the scan region.

Table S2 Chemical analyses obtained from EDX analyses performed on 30 minutes deposited PbI2 sample deposited on ITO/PTAA.. Due to the high sampling depth, elements from the glass, ITO and PTAA are recongnizable. It is worth to note the Pb/I ratio of 0.47 with a slight excess of lodine species exclude the presence of atomic lead in the film.

ELEMENT	WEIGHT	ATOMIC	
	%	%	
Ν	3.29	8.27	
0	20.05	44.77	
Na	2.95	4.52	
Mg	1.02	1.48	
SI	18.95	23.72	
Ca	3.34	2.93	
In	33.30	10.20	
Sn	3.86	1.14	
1	7.09	1.97	
Pb	5.56	0.94	



Figure S 5 The thickness of the perovskite, ~80 and ~150 nm, was determined by verifying that refractive index n obeys the Cauchy equation in the transparent region. In the graphs are reported the ellipsometric experimental data (Psi and Delta) collected at various angles and the Cauchy fit.



Figure S6 I-V Characteristics of the light soaked stabilized 150 nm MAPbI₃ thick champion device.

Here we reported the model for the calculation of the η_{ID} . In this model, J-V curve under illumination can be described by equation:

$$J = J_L - J_0 \left\{ e^{\frac{q(V + JR_s)}{(n_{ID})KT}} - 1 \right\} - \frac{V + JR_s}{R_{sh}}$$
eq S1

The previous equation can be written as follows:

$$\ln\left(J_{sc} - J - \frac{V}{R_{sh}}\right) = \frac{q}{(n_{ID})KT}(V + JR_s) + \ln J_0$$
eq S2

 J_0 and $n_{\text{\rm ID}}$ can be obtained by mathematical elaboration of experimental J-V curves under

illumination, calculating the intercept and the slope of fitting curve $ln \left(J_{sc} - J - \frac{V}{R_{sh}}\right)_{VS} \left(V + JR_{s}\right)$, respectively. In the calculation, R_{shunt} and R_s are taken in each device as done by Liao *et al.*⁷⁰