Supplementary Materials for

Room Temperature Epitaxy of $CH_3NH_3PbI_3$ Halide Perovskite by Pulsed Laser Deposition

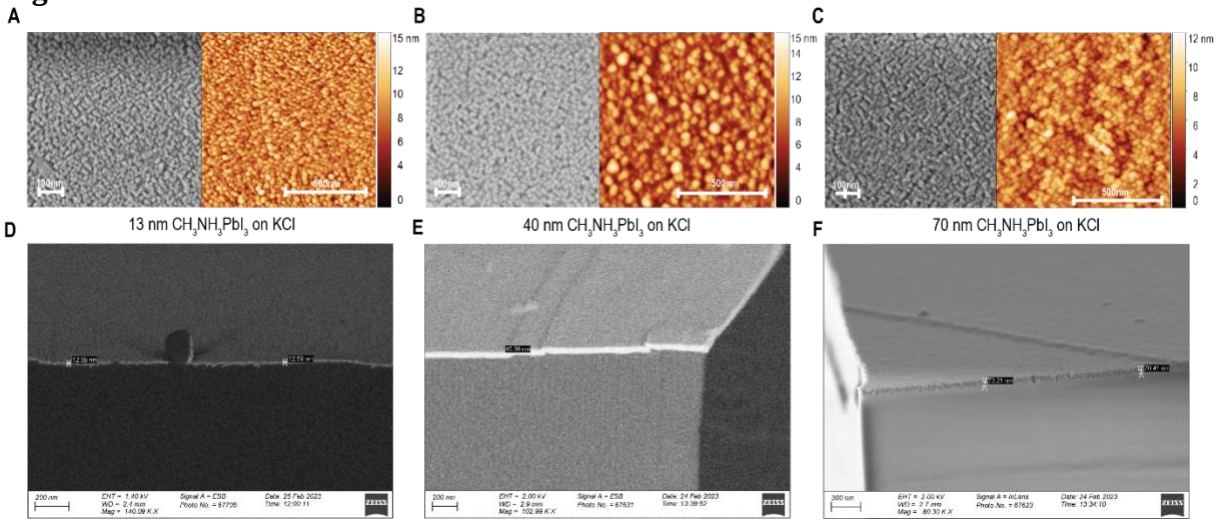
Junia S. Solomon,[1]* Tatiana Soto-Montero,[1] Yorick A. Birkhölzer,[1] Daniel M. Cunha,[1] Wiria Soltanpoor,[1] Martin Ledinský,[2] Nikolai Orlov,[3] Erik C. Garnett,[3] Nicolás Forero-Correa,[4] Sebastian E. Reyes-Lillo,[5] Gertjan Koster,[1] Guus Rijnders,[1] Linn Leppert,[1] Monica Morales-Masis [1*]†

[1]MESA+ Institute for Nanotechnology, University of Twente, 7500 AE Enschede, The Netherlands
[2]Institute of Physics, Academy of Sciences of the Czech Republic, Cukrovarnická 10, Prague 162 00, Czech Republic
[3]AMOLF Institute, Amsterdam, The Netherlands
[4]Doctorado en Fisicoquímica Molecular, Facultad de Ciencias Exactas, Universidad Andres Bello, Santiago 837-0136, Chile
[5]Departamento de Ciencias Físicas, Universidad Andres Bello, Santiago 837-0136, Chile

* Email: j.s.s.solomonsathiaraj@utwente.nl; m.moralesmasis@utwente.nl

This PDF file includes:

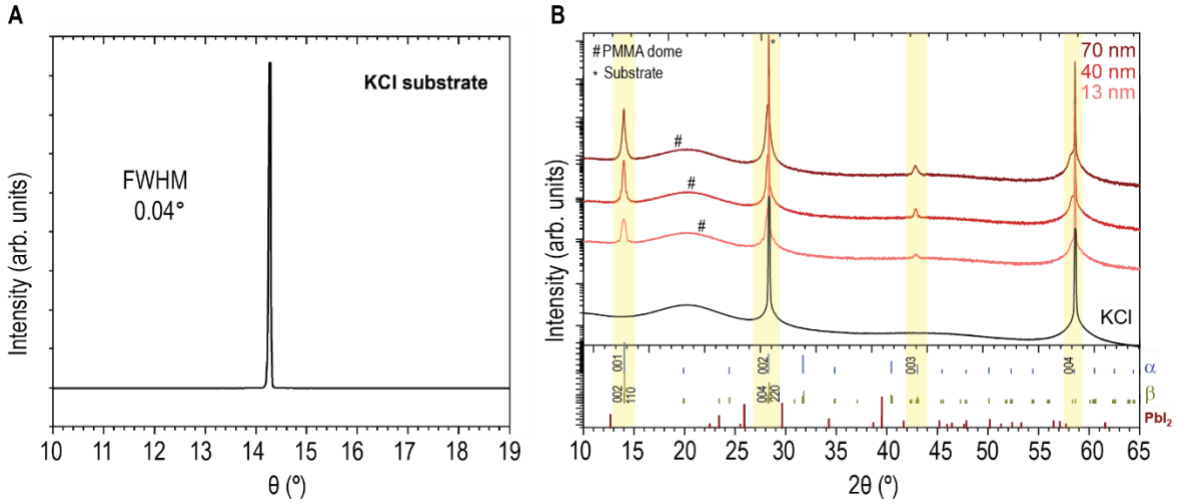
Fig. S1.



Morphology and thickness of the epitaxial $CH_3NH_3PbI_3$ thin films grown at different laser pulses.

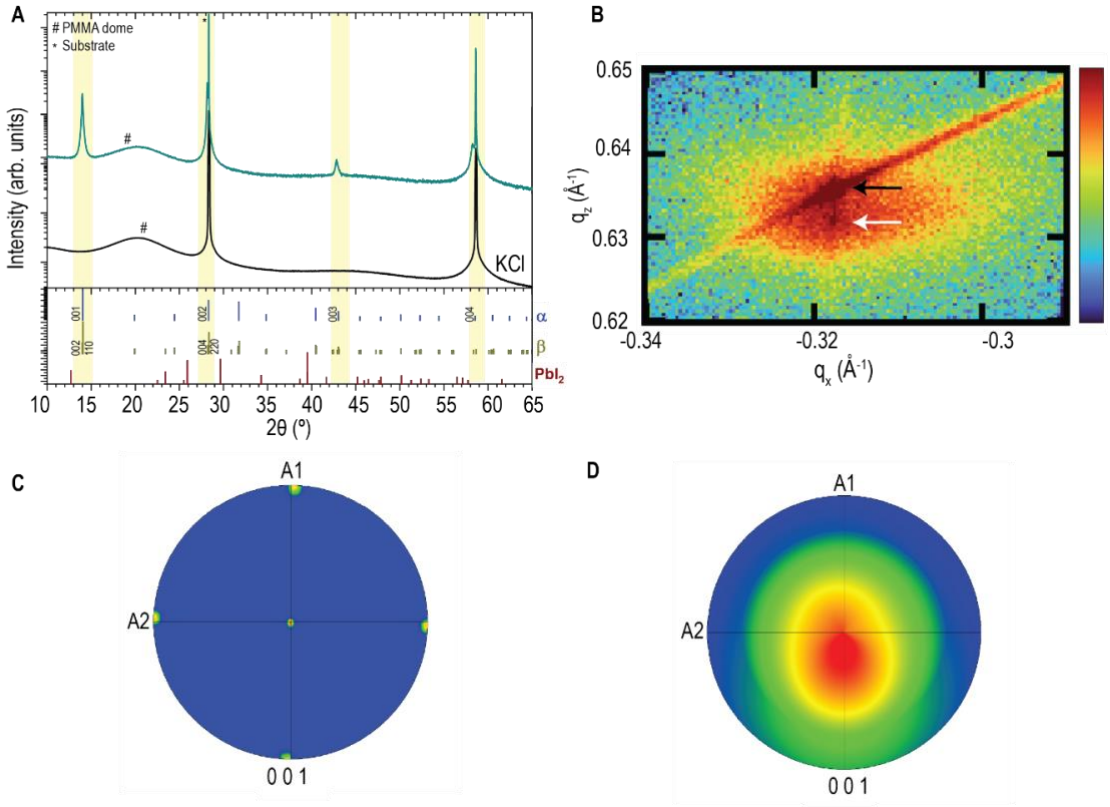
(A-C) shows top-view from SEM and AFM of 13 nm, 40 nm, 70 nm thick epitaxial $CH_3NH_3PbI_3$ thin films, respectively. AFM image in (A) has a distortion in the image due to drift while performing the scan. (D-F) shows the cross-section SEM images of $CH_3NH_3PbI_3$ thin films grown at 1500, 3000 and 6000 pulses.

Fig. S2.



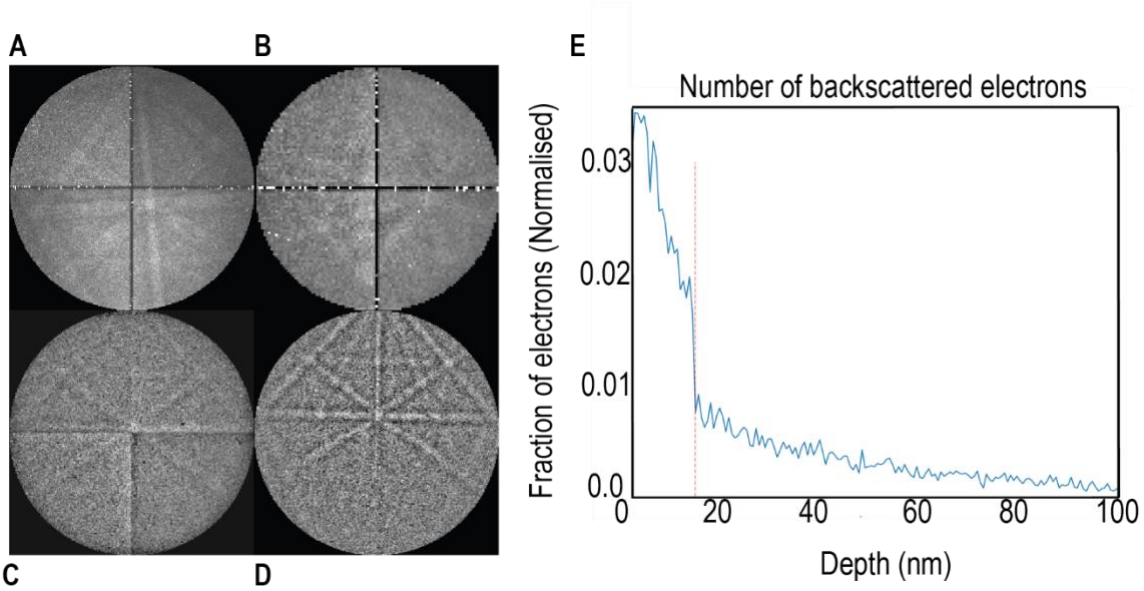
(A) Rocking Curve of KCl substrate and (B) XRD pattern of $CH_3NH_3PbI_3$ thin film of three different thickness with a longer scan range.

Fig. S3.



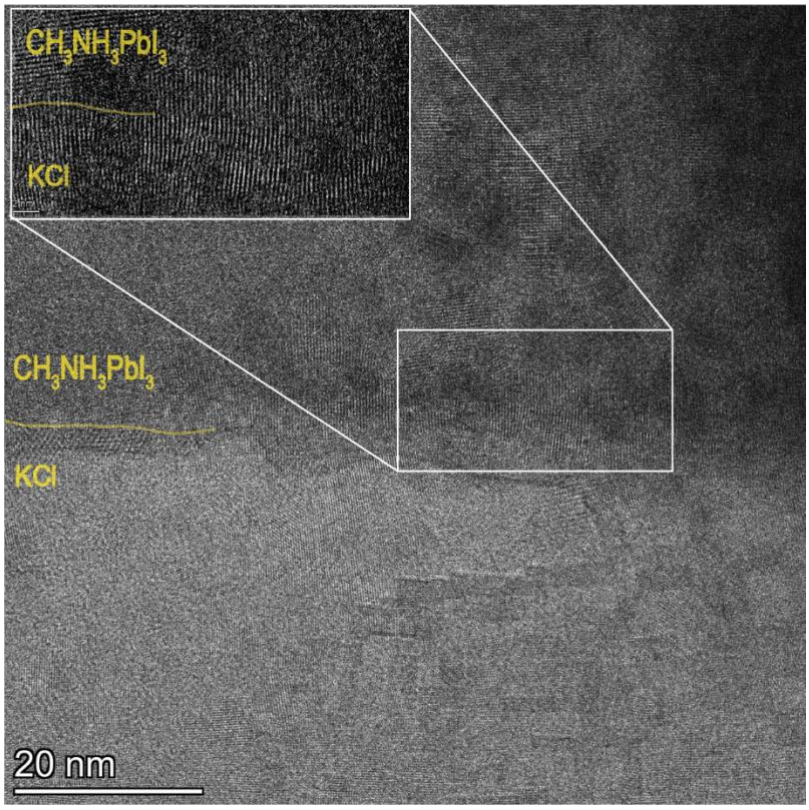
(A) XRD pattern and (B) RSM of $CH_3NH_3PbI_3$ thin film grown higher deposition rate of 5 nm/min. Inverse pole figure of the 100 nm $CH_3NH_3PbI_3$ film on KCl. (C) Maximum polar angle is 90°, (D) Maximum polar angle is 4° (the chosen threshold for grain boundaries)

Fig. S4.



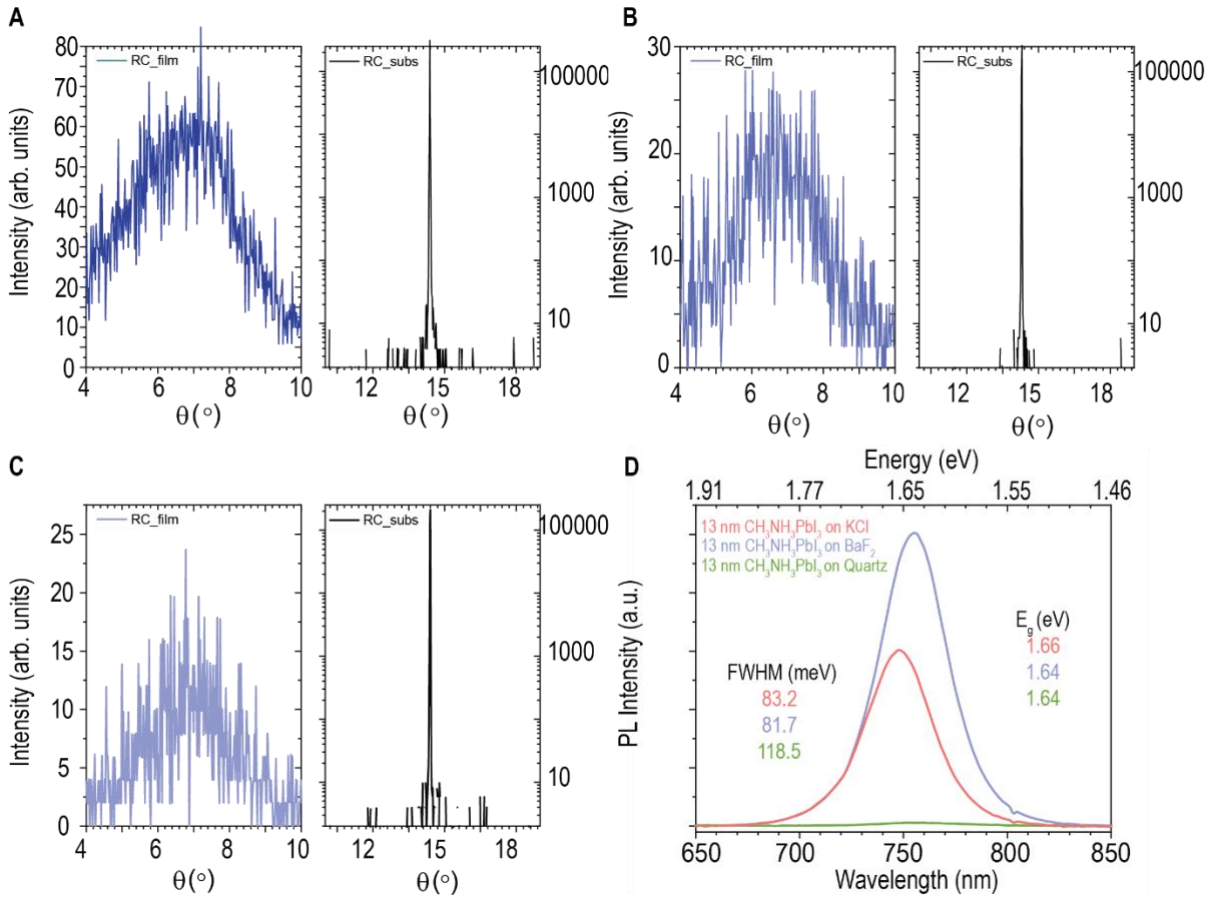
(A-D) EBSD Kikuchi patterns of KCl substrate, 13 nm , 70 nm and 100 nm $CH_3NH_3PbI_3$ film on KCl, respectively. (E) Depth penetration histogram for BSE (5 kV, KCl substrate and 13 nm $CH_3NH_3PbI_3$ layer, red dashed line corresponds to the thickness of the $CH_3NH_3PbI_3$ film).

Fig. S5.



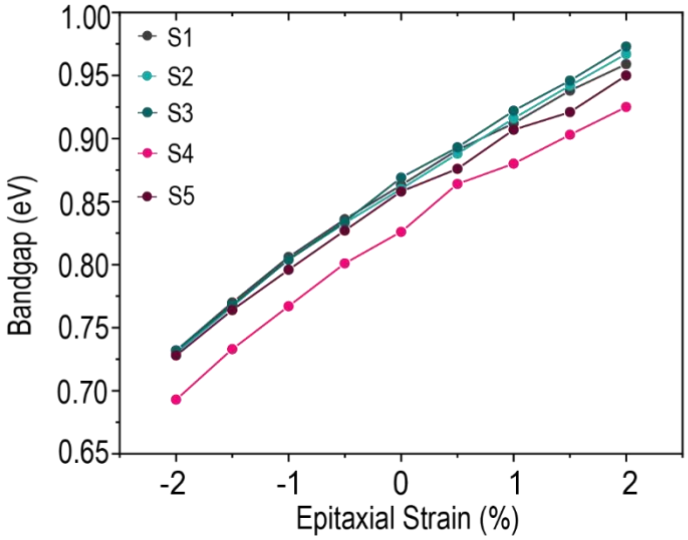
TEM measurements of the interface between $CH_3NH_3PbI_3$ thin film and KCl substrate
(less damaged regions show the epitaxial relationship between the substrate and film.)

Fig. S6.



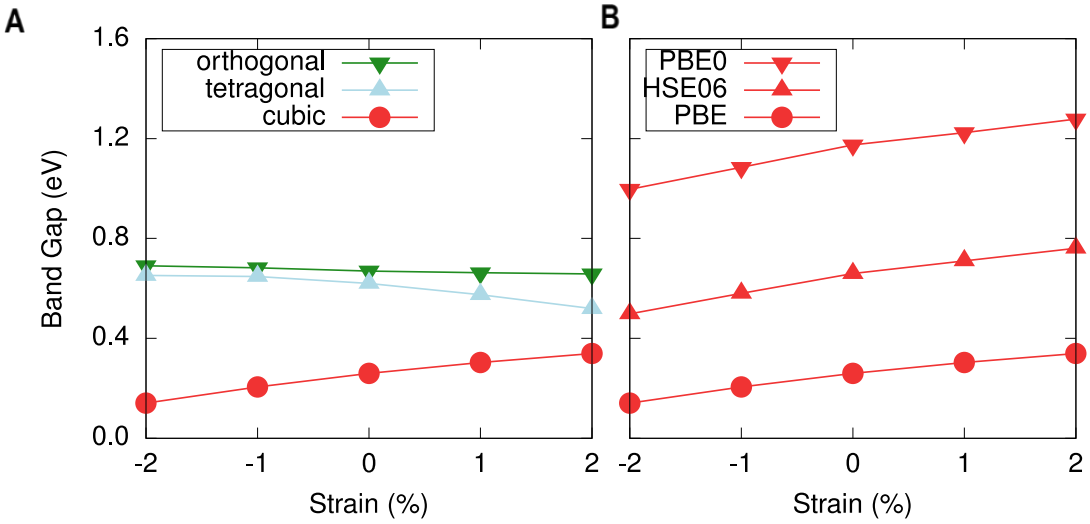
Rocking curve measurements of $CH_3NH_3PbI_3$ on BaF_2 for different thicknesses (A-C) and (D) PL measurements of 13 nm $CH_3NH_3PbI_3$ on KCl, BaF_2 and Quartz.

Fig. S7.



DFT band gaps of HSE06, averaged over five low-energy MA configurations.

Fig. S8



(A) Band gaps of strained orthogonal, tetragonal, and cubic $CsPbI_3$ calculated with the PBE functional including spin-orbit coupling (SOC) self-consistently. (B) Band gaps of strained cubic $CsPbI_3$ calculated with PBE+SOC, HSE06+SOC, and PBE0+SOC.