Electronic Supplementary Information for

## **Controlling Crystallization to Imprint Nanophotonic Structures into Halide Perovskites Using Soft Lithography**

Sarah Brittman, Sebastian Z. Oener, Ke Guo, Haralds Āboliņš, A. Femius Koenderink, and Erik C. Garnett\*

Center for Nanophotonics, AMOLF, Science Park 104, Amsterdam 1098 XG, the Netherlands

\*Corresponding author: garnett@amolf.nl

## Estimated refractive indices and mode indices of the waveguide

The waveguide structure consists of a film 140 nm thick underneath a corrugation that is 50 nm deep. First an effective refractive index for the layer was approximated. The value of the refractive index (*n*) of the perovskite at 543 nm was taken to be 2.25.<sup>1</sup> To approximate the effective refractive index, the layer was considered a stack of two layers:

(1) 50 nm layer with an index determined by its 50% fill fraction:

$$1.74 = \sqrt{0.5 * 2.25^2 + 0.5 * 1^2}$$

(2) 140 nm layer with an index of 2.25.

The low-index layer is then 26% of the total thickness, so the combined index is estimated as  $2.13 = \sqrt{0.74 * 2.25^2 + 0.26 * 1.74^2}$ .

Then the mode indices of the TE and TM waveguide modes of a 190-nm layer with this refractive index (n=2.13) on a fused silica substrate (n=1.46) were calculated from equations 4.4 and 4.17 in

Urbach, H. P. and Rikken, G. L. J. A. Spontaneous Emission for a Dielectric Slab. *Phys. Rev. A*, 1998, 57, 3913-3930.



**Fig. S1** Scheme of creating the waveguide underneath the gratings in the silicon master. After an array of gratings was etched in the silicon, specific lines of the array were protected by photoresist. All the unprotected silicon is etched away, leaving lines of gratings on top of an underlying waveguide. The unprotected gratings were damaged and not used. This scheme was applied for expediency - simply to reuse an existing photolithography mask on hand.



**Fig. S2** Absorptance, photoluminescence, and X-ray diffraction of the CH<sub>3</sub>NH<sub>3</sub>PbBr<sub>3</sub> film.



Fig. S3 Isolated crystals and wire-like structures that form because there is too much solvent in the film when it is imprinted.



**Fig. S4** Weak imprinting from too little solvent present for the array to fully recrystallize in contact with the stamp. Note the scattering of the array and lack of a highly uniform color from diffraction.



**Fig. S5** Isolated crystals form underneath the stamp when there is too much solvent from precursors in pure DMF (right). A continuous film forms when the solvent is removed before imprinting (left). The top and bottom bands are the patterned regions, while the center band is unpatterned. The patterns here are inverse nanoscale pyramids, not gratings.



Fig. S6 Increasing crystallization of the grating with a decrease in the time of the preannealing step. Longer pre-annealing removes the solvent that facilitates recrystallization.



Fig. S7 CH<sub>3</sub>NH<sub>3</sub>PbBr<sub>3</sub> grating imprinted using only DMSO



Fig. S8  $CH_3NH_3PbI_3$  grating from only DMSO. The 1 M precursor solution was made from a 1:1 ratio of  $PbI_2$  (Aldrich) and  $CH_3NH_3I$ , which was synthesized and recrystallized from ethanol. For the iodide perovskite, no pre-annealing step was necessary. Rather, the precursor was spincoated to form a grayish intermediate that was then imprinted.

ĺ –	Lines			Gaps		Periodicity
Si master	Perovskite	Ratio	Si master	Perovskite	Ratio	Ratio
128	134	1.05	110	113	1.03	1.05
126	140	1.11	121	108	0.89	1.04
130	134	1.03	116	124	1.06	1.04
129	140	1.08	126	132	1.04	1.10
137	142	1.04	129	134	1.04	1.08
148	169	1.15	129	121	0.94	1.09
159	164	1.03	129	137	1.06	1.09
164	191	1.16	134	126	0.94	1.10
175	199	1.14	132	124	0.94	1.08
164	194	1.18	153	140	0.91	1.09

**Table S1** Comparison of dimensions (in nm) of the lines and air gaps in the silicon master and the perovskite gratings, which were imprinted from the mixed DMF-DMSO precursor. The total periodicity of the structures is about 4-10% larger in the perovskite than in the silicon masters, likely from the tension in the PDMS stamp.