

## SUPPORTING INFORMATION

# **A Photonic Crystal Laser from Solution Based Organo-Lead Iodide Perovskite Thin Films**

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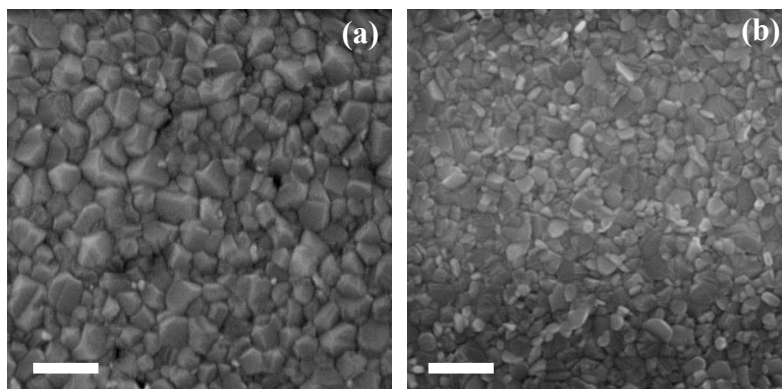
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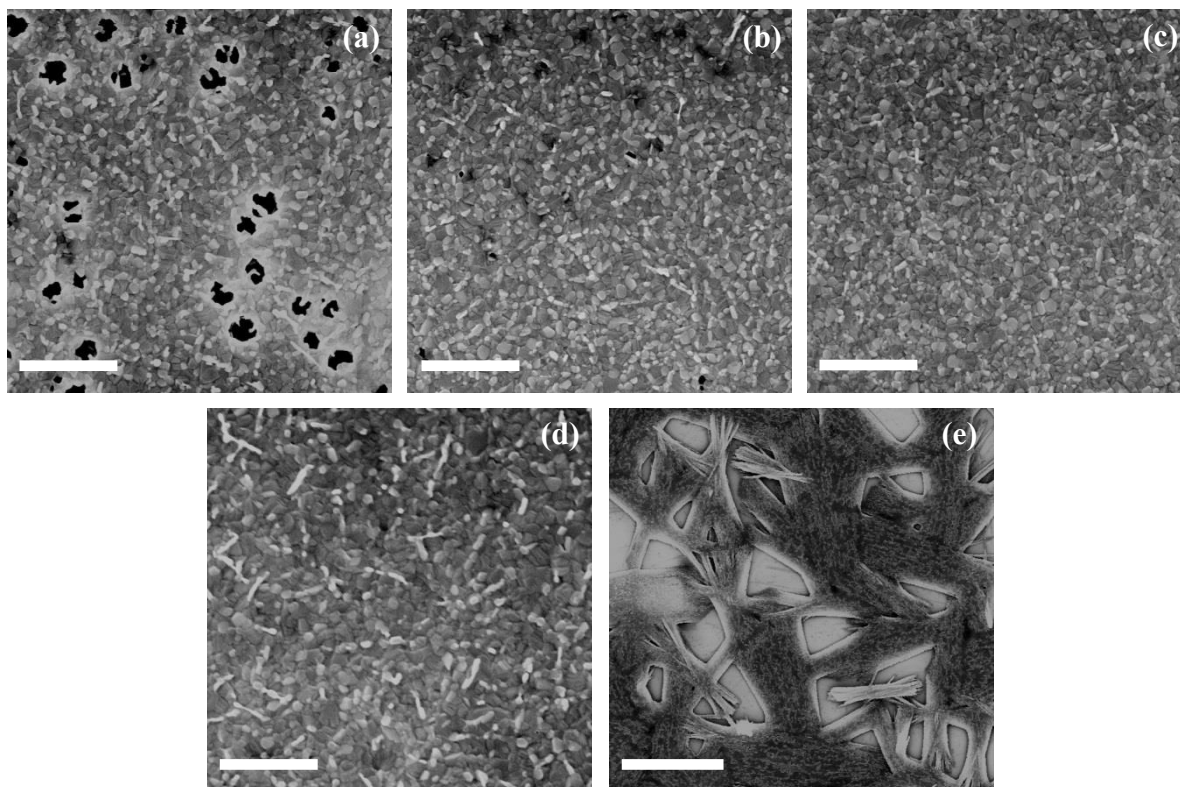
Compared with using DMF as the host solvent, using DMSO will achieve rapid crystallization of a more uniform distribution of perovskite nucleates followed by thermal annealing to finish the crystal growth. Figure S1 shows surface scanning electron microscope (SEM) images of films that were fabricated using DMF and DMSO as host solvent. In the former case, the film forms larger grain sizes with rather randomly crystallite orientation which makes it hard to conformally fill the spaces between the nano-pillars of the patterned PhC quartz substrate.



**Figure S1. Surface morphology differences between perovskite thin films:** **a**, using DMF and **b**, DMSO as the host solvent. In the case of DMF, toluene dripping takes place at 7 s delay time after the start of spin-casting while in the DMSO case, the corresponding delay is 37 s. Both films are thermally annealed at 100 °C for 4 mins. DMF method forms larger average grain size and more randomly distributed orientation, which leads to more pronounced surface roughness. The scale bars are 500 nm.

We also analyzed the effect of different dripping time on the surface morphology of perovskite films in the DMSO case as shown in Figure S2. If dripping takes place before the oversaturation of precursor solution (Figure S2a, b), toluene will play only a limited role of reducing the solubility of perovskite in the mixed solvent. This inhibits uniform crystallization and the film will develop holes (voids) during subsequent annealing process. By contrast, if dripping occurs after precursor solution gets oversaturated, crystallization will proceed under DMSO environment. The high boiling temperature (189 °C) of DMSO will promote slow crystallization of perovskite, resulting in texture structure and large void in the film (Figure S2d, e). We have

found that carefully timed toluene dripping approximately 37 s after beginning of spin-casting

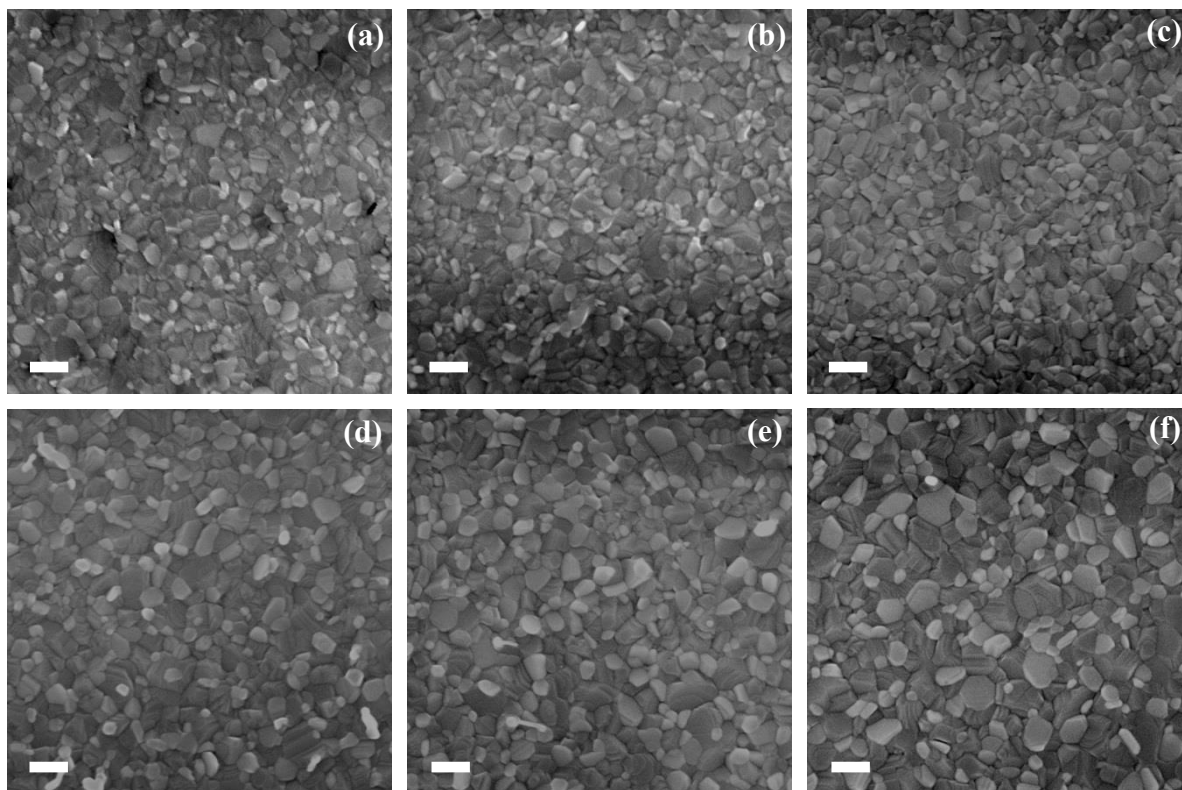


reveals the best result as demonstrated in Figure S2c.

**Figure S2. Effect of dripping time on film quality.** Surface SEM images of perovskite films with different toluene dripping time delays using DMSO as host solvent. **a**, 30 s, **b**, 35 s, **c**, 37 s, **d**, 40 s, **e**, no dripping. The scale bars for **a-d** are 1  $\mu\text{m}$  and scale bar in **e** is 5  $\mu\text{m}$ . If toluene is dripped when the precursor solution reaches over-saturation, the film will be pinhole and texture structure free. Here 37 s delay time for toluene dripping yields high-quality closely packed optically smooth thin film.

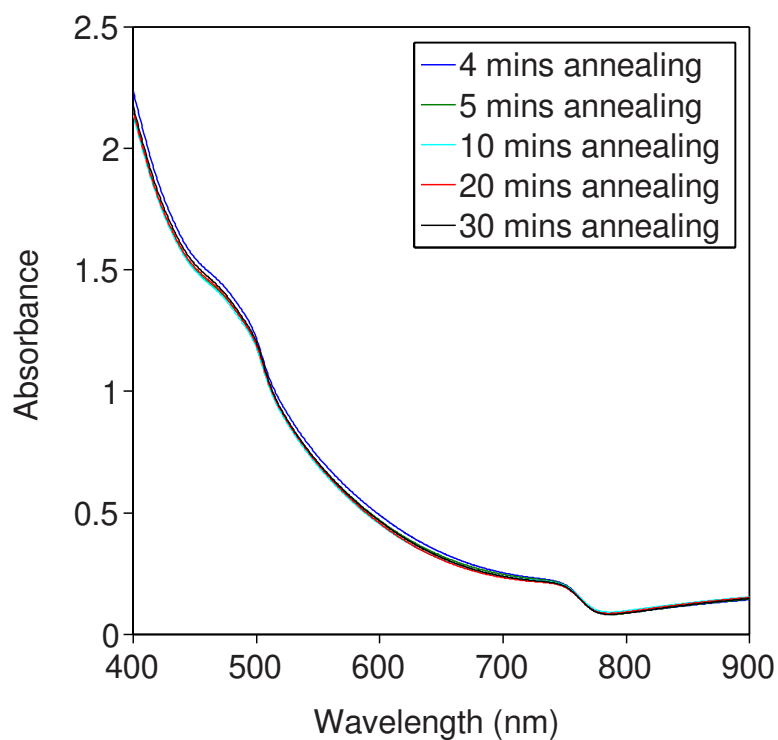
Figure S3 shows surface morphology images of perovskite films within a particular window of thermal annealing time. With 3 mins annealing, there are regions which are not fully converted (Figure S3a). After 4 minutes, the perovskites are seen to be fully crystallized and achieve 100% surface coverage in this polycrystalline structure (Figure S3b-f). Longer annealing will promote the crystal growth and form bigger grains; this, however, does not change the optical absorption

spectrum (Figure S4) indicating that the conversion process is fully completed in each case.

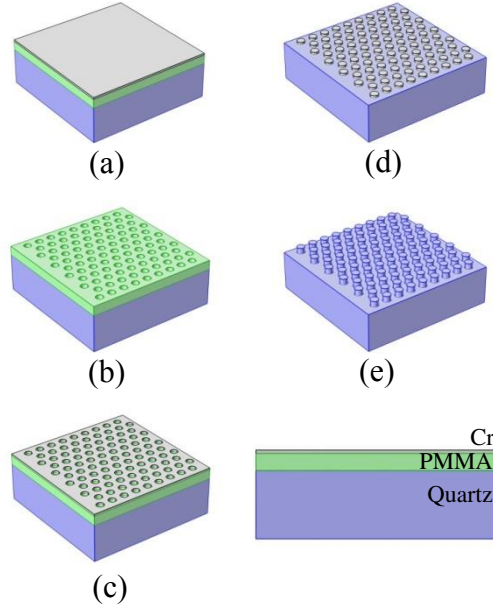


Therefore, considering the feature size of the photonic crystals we targeted from numerical simulations, an annealing time of 4 mins was chosen as optimal for perovskite films in this work.

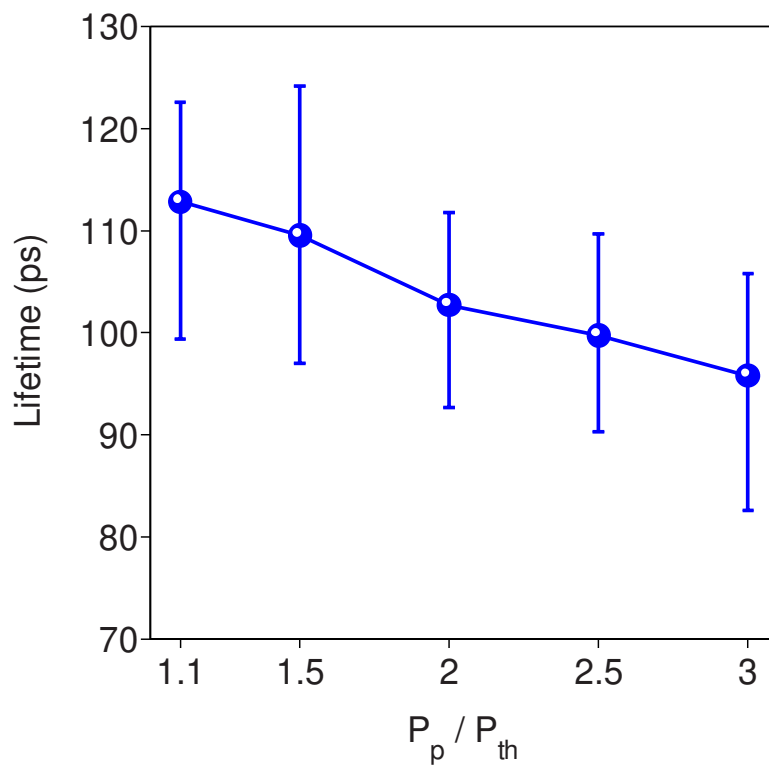
**Figure S3. Effect of thermal annealing time on perovskite grain size.** All samples used DMSO as the host solvent and 37 s delay for toluene dripping. Immediately after spin-casting, the samples were transferred onto a hot plate for annealing at 100 °C for **a**, 3 mins, **b**, 4 mins, **c**, 5 mins, **d**, 10 mins, **e**, 20 mins, **f**, 30 mins. A pronounced increase in average grain size can be seen with longer annealing time. For a 3 mins annealing time, pinhole-like defects on the surface of the film present, suggesting that annealing time  $\geq 4$  mins are necessary for good quality, homogeneous perovskite thin films. All scale bars are 200 nm.



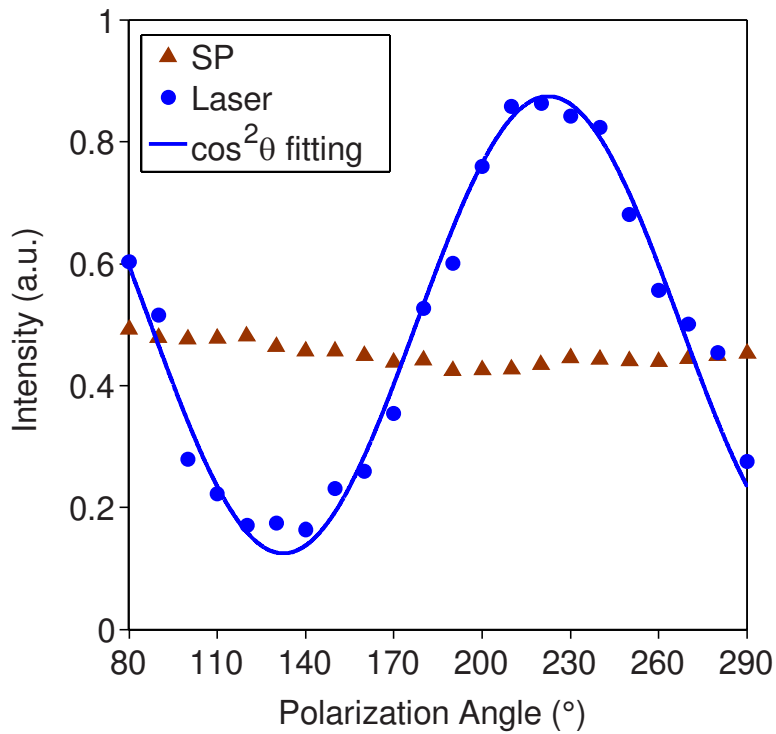
**Figure S4. Absorbance spectra for thin film samples with different annealing time.** Although longer annealing time promotes more grain growth, only small differences are seen in their optical absorption spectrum, indicating that the crystallization process is complete at  $t \approx 4$  mins; this was chosen to be optimum annealing time for the PePhC laser due to small grain size.



**Figure S5. Electron beam lithography (EBL) based fabrication processes flow for a 2D hexagonal lattice PhC.** **a**, 170 nm thick Poly(methyl methacrylate) (PMMA) was spin-cast (4000 rpm, 60 s) on 1'' × 1'' quartz and soft-baked at 180 °C for 90 s, then a 7.5 nm Cr film was evaporated onto the surface of PMMA layer followed by EBL (30 kV, 0.17 nA) process. **b**, The PMMA resist was developed in MIBK/IPA 1 : 3 developer after removing the Cr film by Cr etchant, obtaining the 2D PhC pattern. **c**, A 20 nm chromium layer was deposited on the patterned PMMA for plasma etch mask. **d**, PMMA lift-off process in piranha solution (volume ratio of 96% H<sub>2</sub>SO<sub>4</sub> : 30% H<sub>2</sub>O<sub>2</sub> = 2 : 1). **e**, Inductively coupled plasma reactive ion etching process was carried out to etch the sample to form the final 2D hexagonal PhC nano-pillar array. Cr etchant was used again to remove the Cr hard mask.

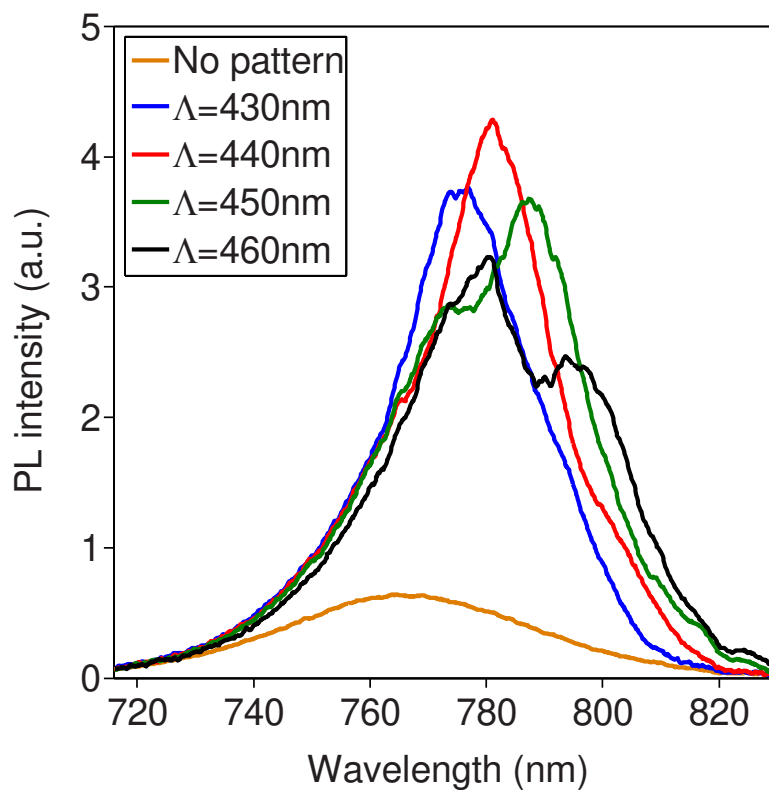


**Figure S6. PePhC stimulated emission decay time measured at different pump energy densities in the lasing regime.** With higher pump energy density, the stimulated emission rate is enhanced which reflected directly in the time-domain data shown in the figure. The error bars are due to synchronization source jitter effect of the electronic system ( $< 40$  ps). The time domain results are obtained by using sub-nanosecond pulsed pump source ( $\tau_{\text{pulse}} \sim 270$  ps) in the green at  $\lambda = 532$  nm, with 1 kHz repetition rate.



**Figure S7. Polarization measurement for PePhC laser output.** The PePhC laser beam is guided to pass through the Glan-Thompson polarizer and enter a spectrometer to collect signal. The setup is aligned so that the pump laser is polarized vertically at 90 degree in transverse plane, while 0 degree refers to polarization axis of Glan-Thompson polarizer aligned horizontally in transverse plane). The PePhC laser output is linearly polarized (blue dots in the figure) which can be well-fitted by  $y(\theta) = 0.16 + 0.7\cos^2(\theta - 222.5)$ . The degree of polarization (DOP) can thus be calculated as  $DOP = \frac{I_{max} - I_{min}}{I_{max} + I_{min}} \approx 70\%$ . The PL signal was also analyzed using the same setup, in contrast, the spontaneous emission fails to show any significant angular dependence when rotating the Glan-Thompson polarizer (brown triangles – the background variation is due to pump laser power fluctuations).





**Figure S8. PL spectra from PhC patterned perovskite samples with different pitch size.** The PL signal is collected by a 10× objective lens (NA = 0.25), and directed into the spectrometer. The enhancement by approximately factor of 6 ~ 7 in PL from the PhC patterned area versus non-patterned area of the films suggests the possible role of pump light intensity concentration by PhC cavity, thus increasing the absorption and PL emission. Different optical density of states spectrum originated from photonic band structure for different pitch size PhC patterns help to filter spontaneous emission spectrum, thus showing various PL lineshapes.