Electronic Supplementary Material (ESI) for Nanoscale.

Mechanical Switching of Orientation-related Photoluminescence in Deep-blue 2D Layered Perovskite Ensembles

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Table S1. Different loading amount of stock and benzylamine solutions added to 1 ml of PDMS in toluene. The final MHLP concentrations were calculated based on the total amount in volume of perovskite precursors added to 1 ml of polymer solution.

Stock	Volume used from the stock solution, µl	Benzylamine, µl	MHLP in Polymer, vol %
$PbBr_2$ (0.41 mmol, 150 mg) HBr (0.82 mmol, 95 µl) Acetone (640 µl)	20	2.84 µl	2.3
	30	4.26 µl	3.4
	50	7.1 µl	5.7

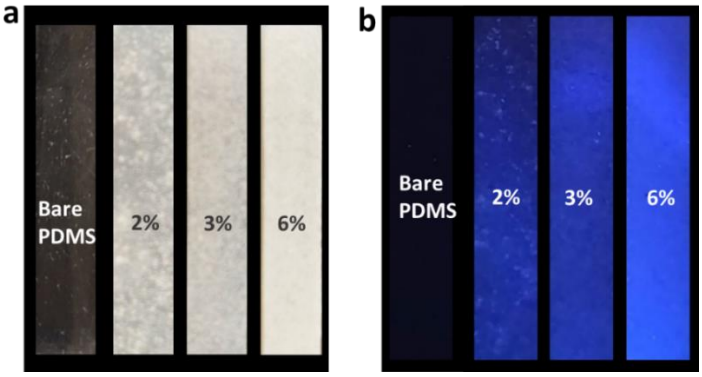


Figure S1. Representative photographs of a set of MHLP-PDMS films prepared with different loading of MHLP precursors, from 2 to 6 % in vol, inside the PDMS taken under normal (a) and ultra-violet (UV) light (b). All the films show a bluish-white emission under UV light except bare PDMS (0% MHLP precursors), which does not show any emission under the same experimental conditions.

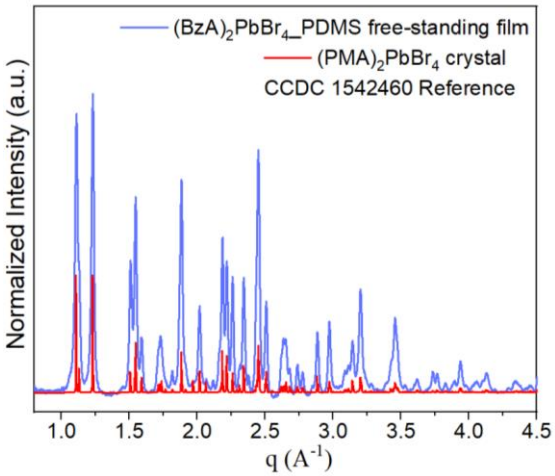


Figure S2. XRD patterns collected from the perovskite polymer films and compared with the reference for $(PMA)_2PbBr_4$ crystals from Ref. [34] in the main text. Note that the authors of this work reported the benzylammonium (BzA) with its IUPAC name, that is, phenylmethanamine (PMA).

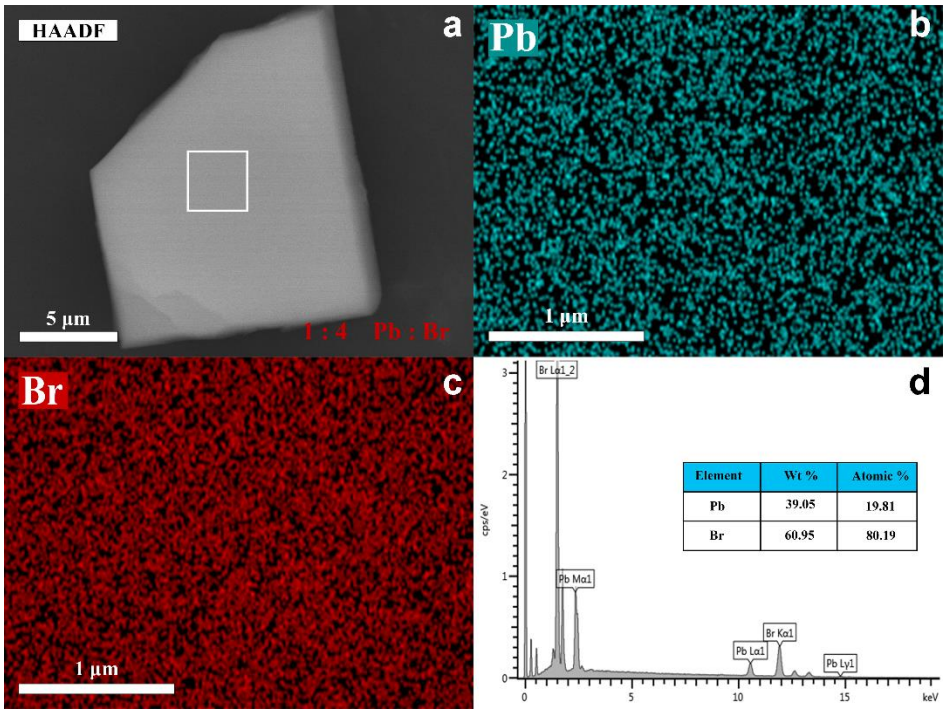
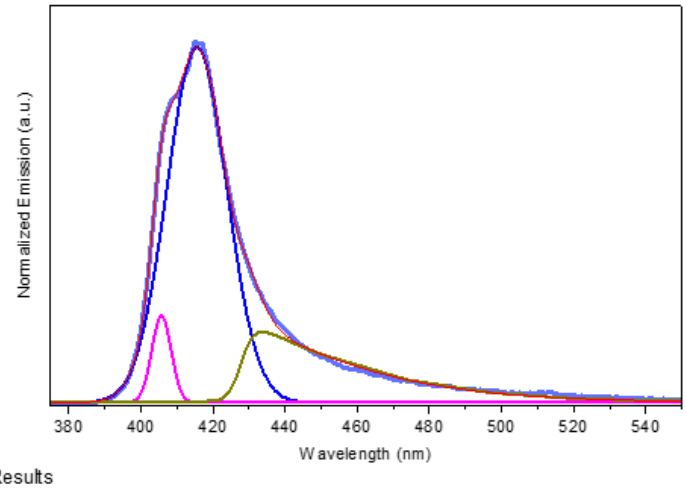


Figure S3. (a) SEM image of a piece of MHLP-PDMS film. (b-c) Quantitative EDS/SEM maps performed on the white framed region in (a) for Pb and Br, respectively. (d) Collected EDS spectra from where the Br:Pb atomic ratio of 4 ± 0.05 is calculated by considering the components of Lα = 10.5 keV for Pbr and Kα = 11.9 keV for Br.



Fitting Results

Peak Index	Peak Type	Area Intg	FWHM	Max Height	Center Grvty	Area IntgP
1	Gaussian	20.31508	19.55285	0.97606	415.5148	68.1693
2	Gaussian	1.5972	6.32565	0.2372	405.70191	5.35958
3	Asym2Sig	7.88864	33.33942	0.19329	457.3864	26.47112

Figure S4. Results of the fitting PL profile of the MHLP-PDMS film, with an adjusted R-squared of 0.9991. The PL of the film and the resulting fitting curve are shown in blue and red, respectively. The two characteristic emission peaks are centered at 406 nm (P_1, in magenta) and 416 nm (P_2, in blue), with corresponding full width at high maxima (FWHM) of 6 nm and 20 nm.

Table S2. Optical characteristics of the investigated samples. A slight blue-shift in the emission of the composite films with respect to the bare platelets is due to the different sample conditions and related changes on the density of platelets under the illuminated spot. The PL fitting profiles were obtained following the peak types described on Figure S4. *PL measured in situ by using the customized set up showed in Figure S5 and reported in Figure S9.

	PL peaks		**FWHM, nm**		**Absorption peaks**	**CIE**	
	P_1, nm (eV)	**P_2, nm (eV)**	**P_1, nm**	**P_2, nm**		**x**	y
$(BzA)_2PbBr_4$ platelet deposit	408 (304)	418 (2.97)	6.5	18	395 (with a shoulder at 408)	0.1733	0.0644-
$(BzA)_2PbBr_4$ PDMS free-standing film, 0%	406 (3.05)	416 (2.98)	7.0	17.0	395 (with a shoulder at 403)	0.1623	0.0260
$(BzA)_2PbBr_4$ PDMS free-standing* film, 70%.	408.5 (303)	416.5 (2.968)	6.0	12.0	395	0.1677	0.0130

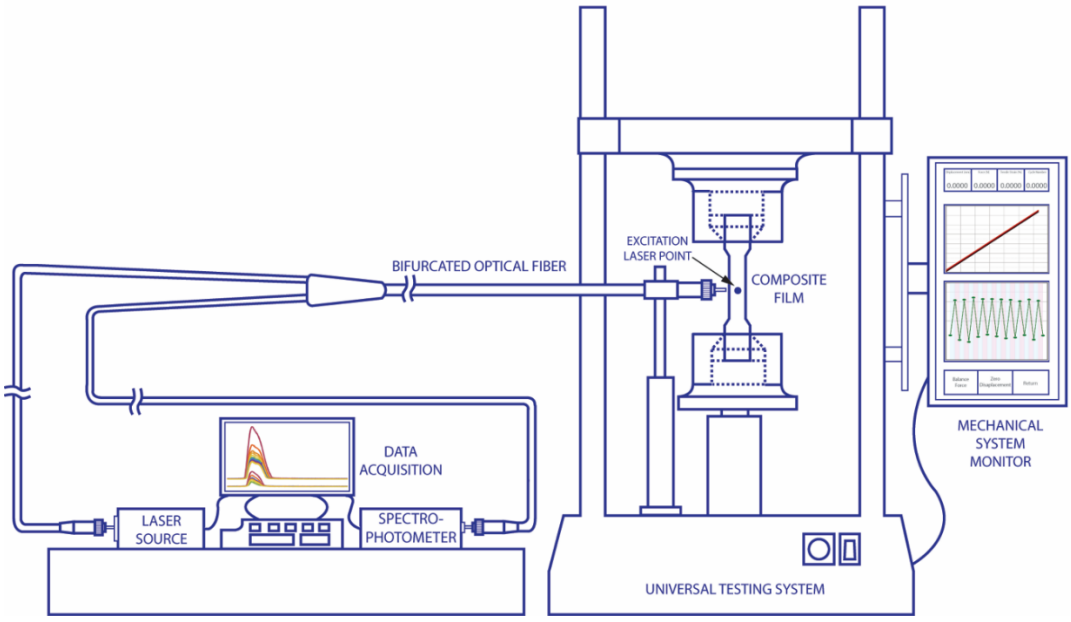


Figure S5. Customized set-up used for investigating in-situ the photoluminescence of the MHLP-PDMS free-standing films under mechanical cyclic stretching.

Additional $(BzA)_2PbBr_4$-PDMS samples investigated under stretch/release cycles

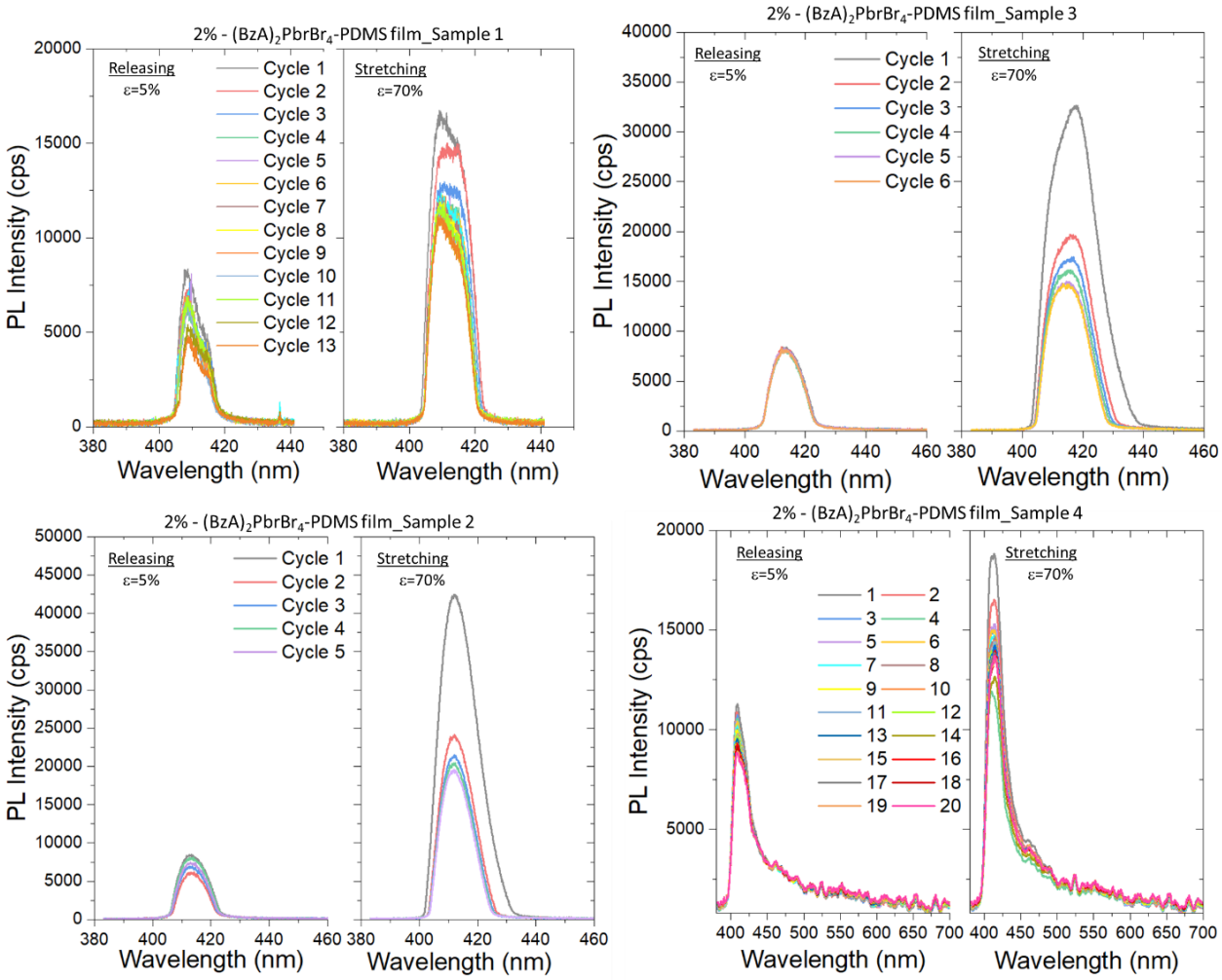


Figure S6. Emission spectra acquired from four different $(BzA)_2PbBr_4$-PDMS free-standing films prepared with 2% of perovskite precursors under continuous stretch/release mechanical cycles. All of them show around 1.5 (sample 4) to 2.5 fold increase in PL intensity after stretching.

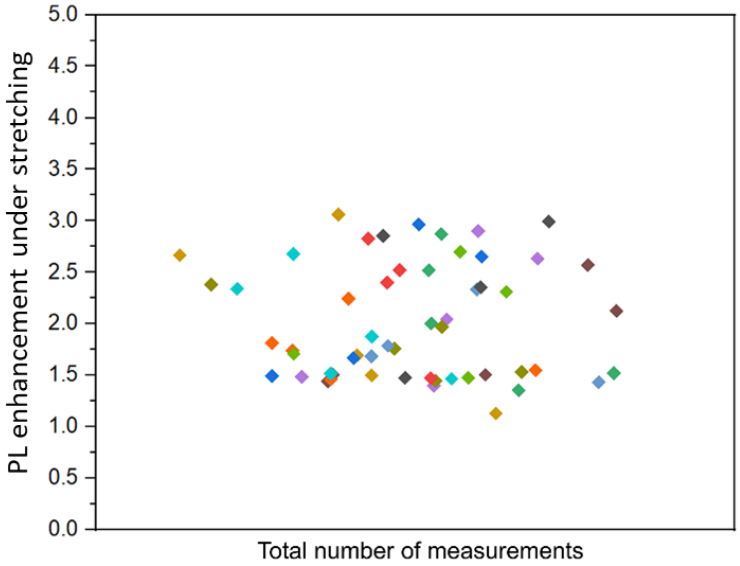


Figure S7. Increment of PL intensity under stretching as a function of the number of measurements performed, that is the total number of loading (70% strain) and unloading (5% strain) cycles recorded (55 measurements highlighted by different colours in the plot). The increment was calculated as the ratio between the maximum peak intensity observed at 70% and 5% for each cycle. This results in an average PL intensity increment of 2.0 ± 0.5, which considers potential mechanical induced degradation over cycle; the average value from the first cycle recorded from all the samples resulted in 2.5 ± 0.5.

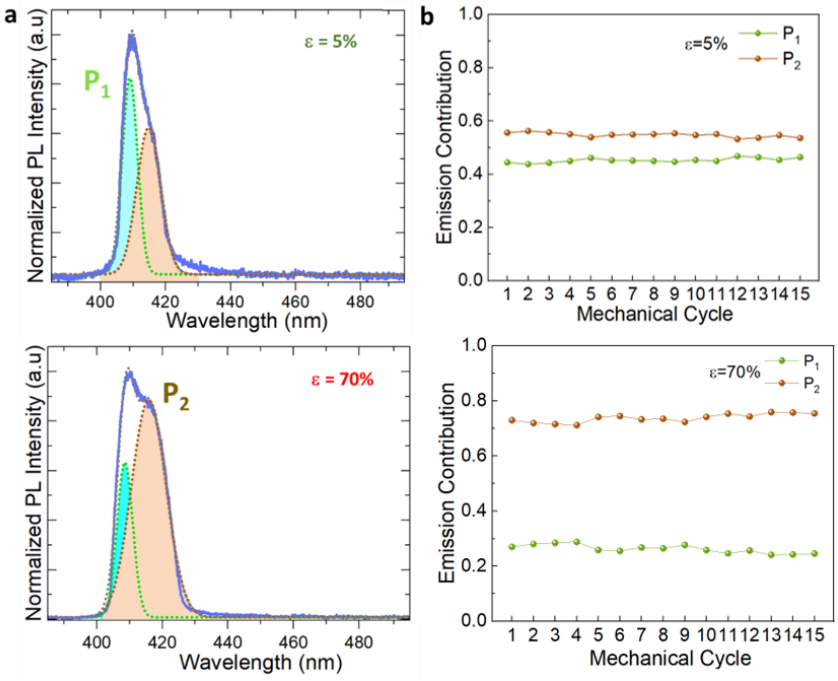


Figure S8. (a) PL spectra collected from one stretching/releasing cycle at 0% and 70% showing the fitting curves for P_1 and P_2. (b) Contributions on the area under the emission profile from P_1 and P_2 at 0% and 70% strain as a function of the number of cycles.

Table S3. Maximum emission intensity from the $(BzA)_2PbBr_4$_PDMS free-standing films recorded in situ at 5% and 70% strain for the first 5 mechanical cycles, and the PL intensity increment calculated from their ratio. The table includes the average (Avg) values per cycle, as well as per sample (S), with the corresponding standard deviations (SD).

	Maximum PL Intensity at 0%, cps					**Maximum PL Intensity at 70%, cps**					**PL Intensity Increment**					**Avg per cycle**	
Cycle	**S1**	**S2**	**S3**	**S4**	**S5**	**S1**	**S2**	**S3**	**S4**	**S5**	**S1**	**S2**	**S3**	**S4**	**S5**	**Avg**	**SD**
1	8348	8501	8374	11285	6268	16735	25452	25627	18822	16928	2.00	3.00	3.06	1.70	2.70	2.48	0.55
2	7336	7187	8416	10872	5897	15007	18136	19700	16523	16837	2.04	2.52	2.34	1.52	2.85	2.26	0.45
3	7640	6955	8201	10737	5956	12950	18459	17432	15027	16838	1.7	2.65	2.12	1.40	2.82	2.143	0.54
4	6431	8107	8182	10555	5642	12065	20436	16133	11929	16728	1.90	2.52	1.97	1.13	2.96	2.09	0.62
5	8109	7443	8248	10426	5821	12214	19604	14972	15304	16720	1.50	2.63	1.81	1.47	2.87	2.06	0.58
										Avg	1.82	2.7	2.26	1.44	2.84		
										SD	0.20	0.17	0.43	0.18	0.08		

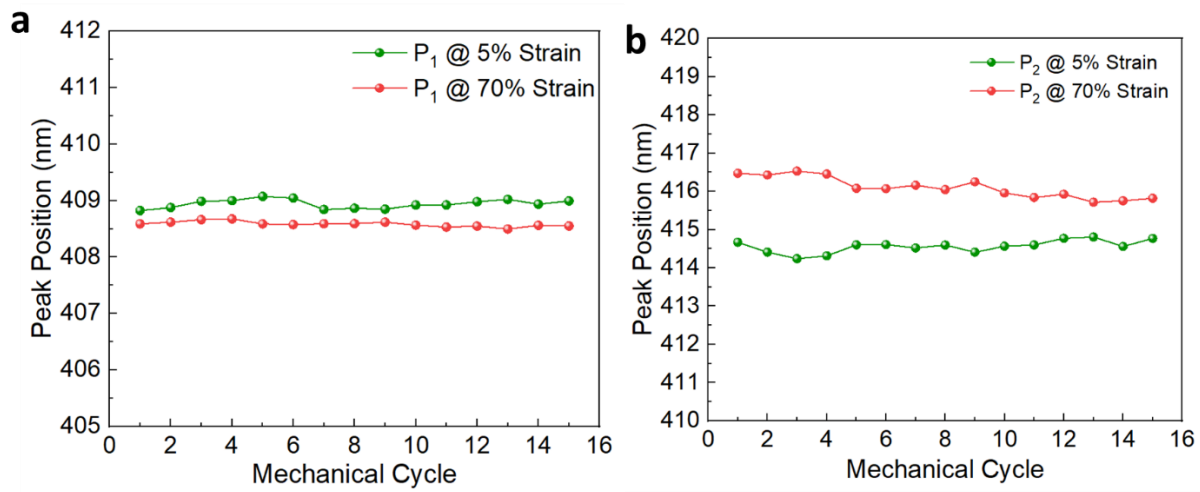


Figure S9. Position of the emission peaks P_1 and P_2 as a function of the number of stretch/release cycle applied to the $(BzA)_2PbBr_4$-PDMS free-standing films prepared with 2% in vol. of MHLP. A red-shift of less than 2 nm is observed on both peak while stretching the films up to 70% strain.

Table S4. Detailed analysis of the PL time decay profiles presented in Figure 3a-b of the main text for P_1 emission peak. τ_1, τ_2, τ_3, and τ_{avg} are the three-exponential time decay fitting and their average value, respectively. All the τ values are reported in ns.

Sample	A1%	τ_1	A2%	τ_2	A3%	τ_3	τ_{Avg}
$(BzA)_2PbBr_4$	67.30	1.08	31.30	4.22	1.40	20.00	5.12
$(BzA)_2PbBr_4$_PDMS free standing film	64.80	1.03	33.80	3.05	1.40	11.50	3.04
Film, ε=40%	63.10	1.21	36.50	2.80	0.40	14.61	2.45
Film, ε=70%	70.10	1.11	29.60	2.70	0.30	14.63	2.19

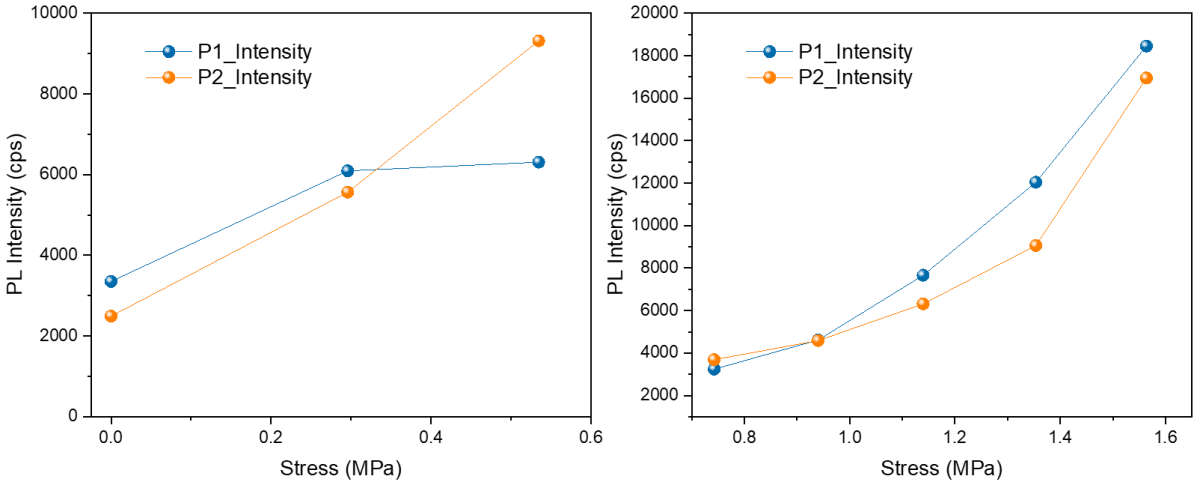


Figure S10. PL intensity of P_1 and P_2 emission peaks as a function of the applied stress/strain collected from the films under stretching up to 70% elongation, corresponding to Figure 3c-d of the main document. The data were collected from two set of experiments by using different acquisition time to avoid signal saturation due to the increment of emission intensity with the elongation.

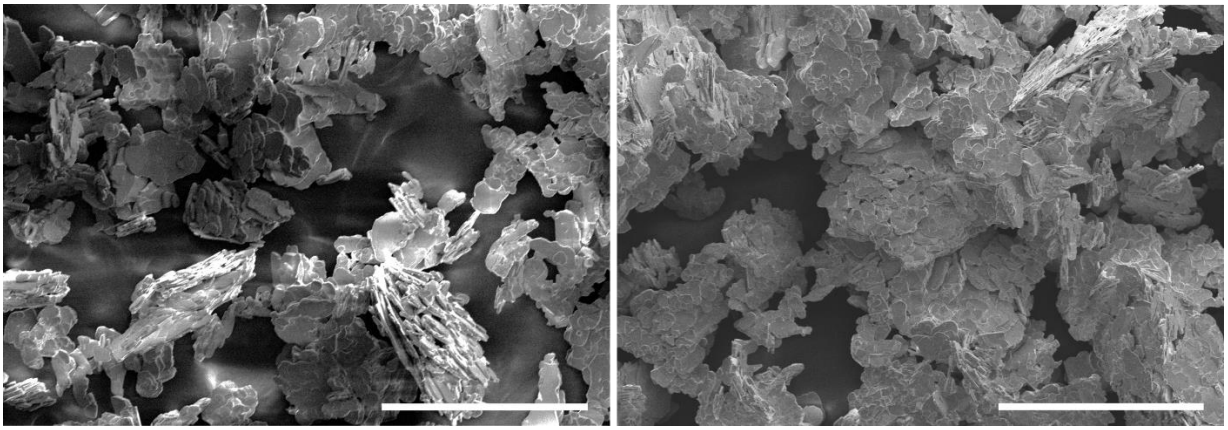


Figure S11. SEM images collected from the $(BzA)_2PbBr_4$ platelets prepared following the protocol used for their in situ grow in the polymer solution and using only pure toluene without PDMS. The images show the formation of randomly oriented stacks of platelets. Scale bars: 100 μm.

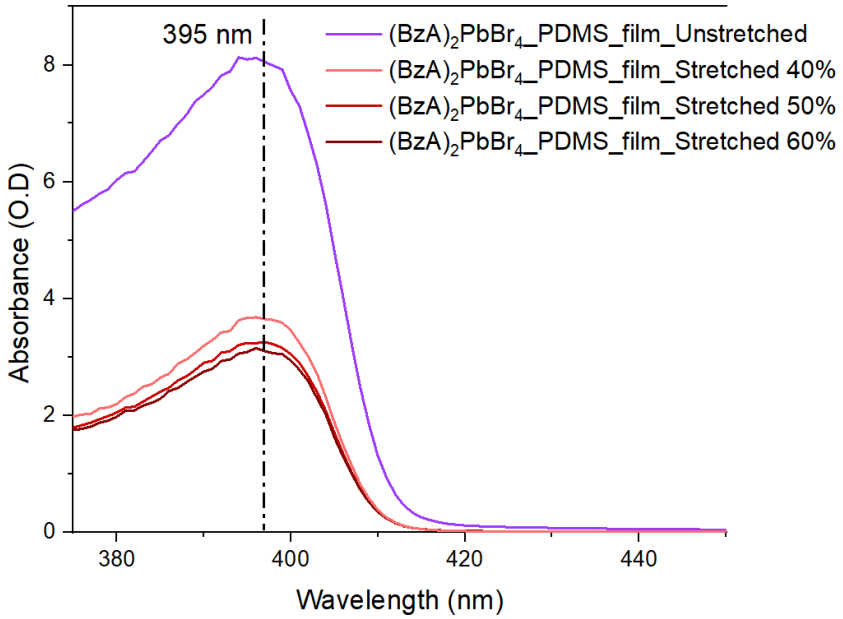


Figure S12. Absorbance spectrum collected in reflectance mode from the composite films under 0% strain and a maximum stretching of ca. 50%.

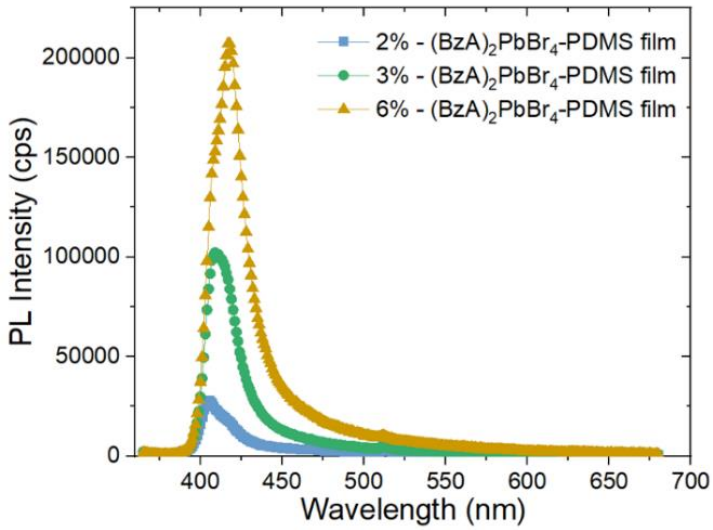


Figure S13. Representative PL spectra of a set of MHLP-PDMS films prepared with different loading of MHLP precursors, from 2 to 6 % in vol.

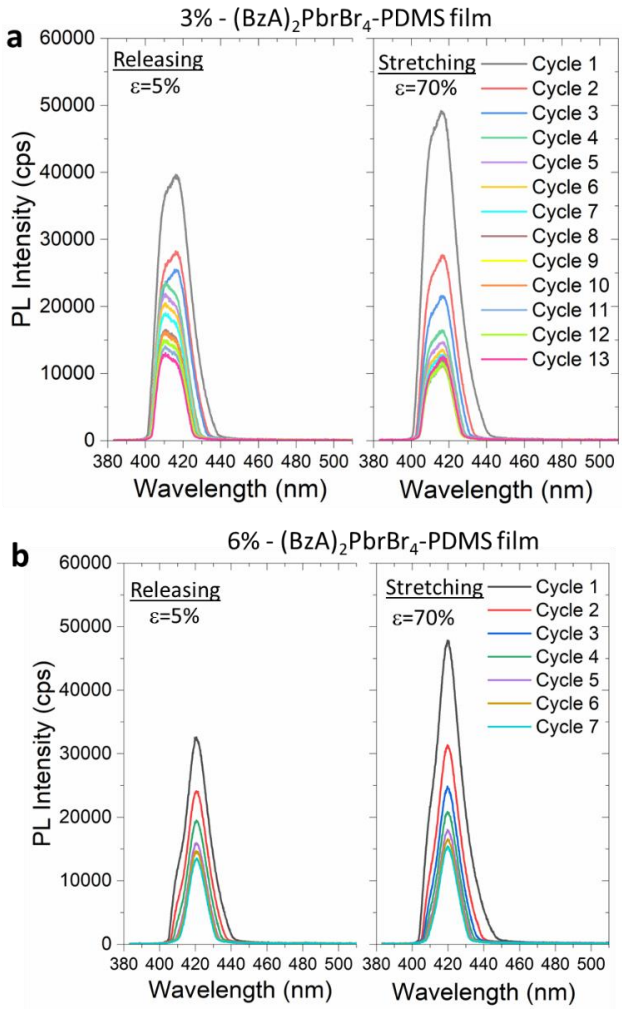


Figure S14. Emission spectra acquired from $(BzA)_2PbBr_4$-PDMS free-standing film prepared with 3% (a) and 6% (b) of perovskite precursors under continuous stretch/release mechanical cycles.

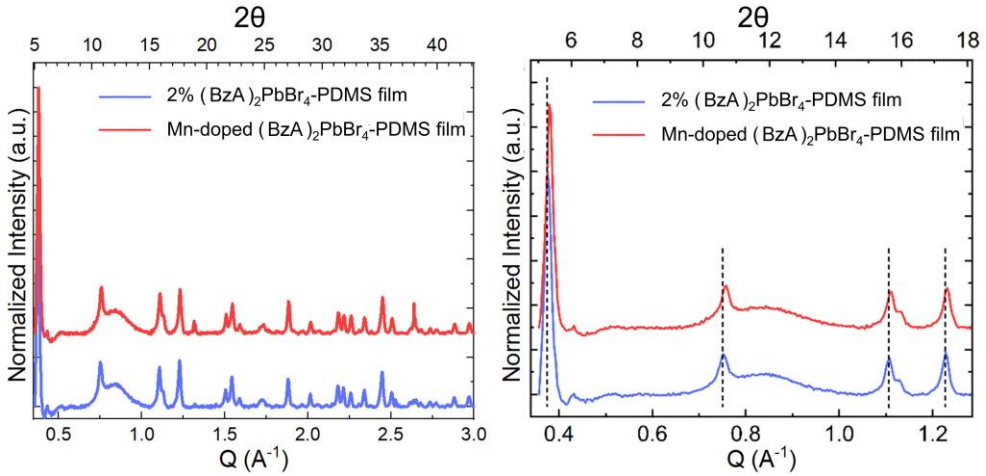


Figure S15. XRD pattern acquired from Mn-doped $(BzA)_2PbBr_4$-PDMS free-standing film (left) showing a slight shift towards the higher angle as compared to the undoped films (right) due to the successful incorporation of smaller Mn atoms in the Pb-Br network.

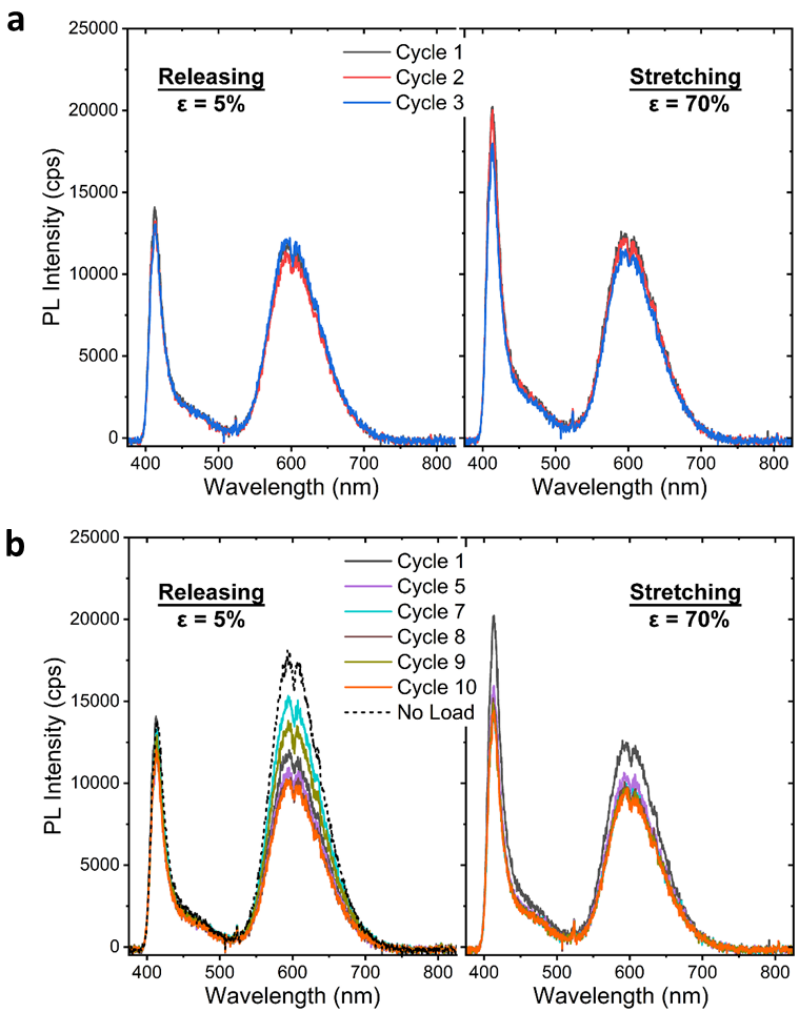


Figure S16. PL spectra collected in situ from the Mn-doped $(BzA)_2PbBr_4$-PDMS free-standing films under continuous stretch (up to 70%)/release (up to 5%) mechanical cycles. The first three cycles are shown in (a). It shows an increment in the intensity of the blue emission peak while the red emission peak remains at similar intensity throughout these cycles. The following cycles are shown in (b) and we observe a relative increment on the intensity of the red emission peak for ε = 5% that is maintained after full removal of the load (black dotted line labelled 'No Load' in (b)).