Supplementary Information (SI) for Nanoscale.

Supporting Information

Study The Effect of Dimensions and Spacer Ligands on The Optical Properties of 2D Metal Halide Perovskites

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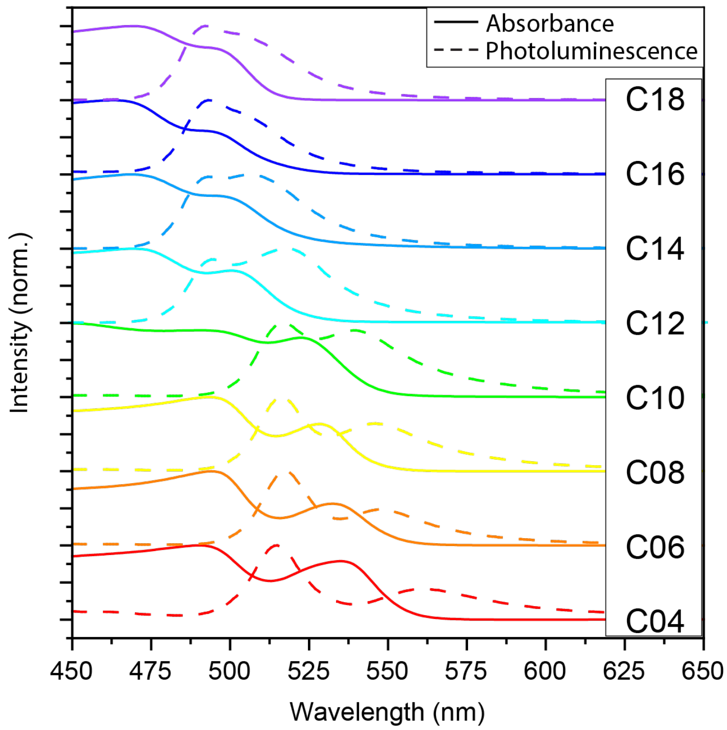
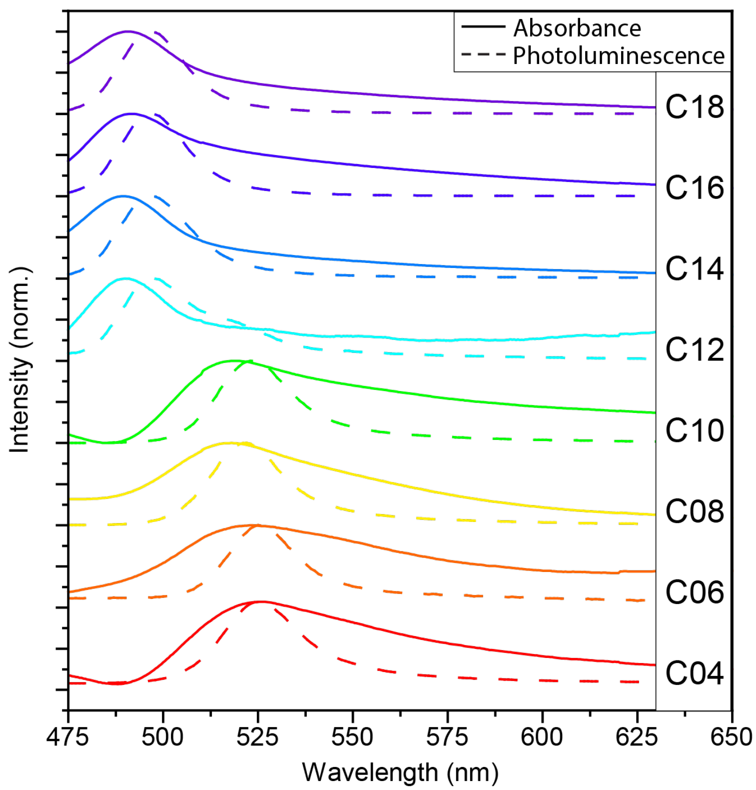
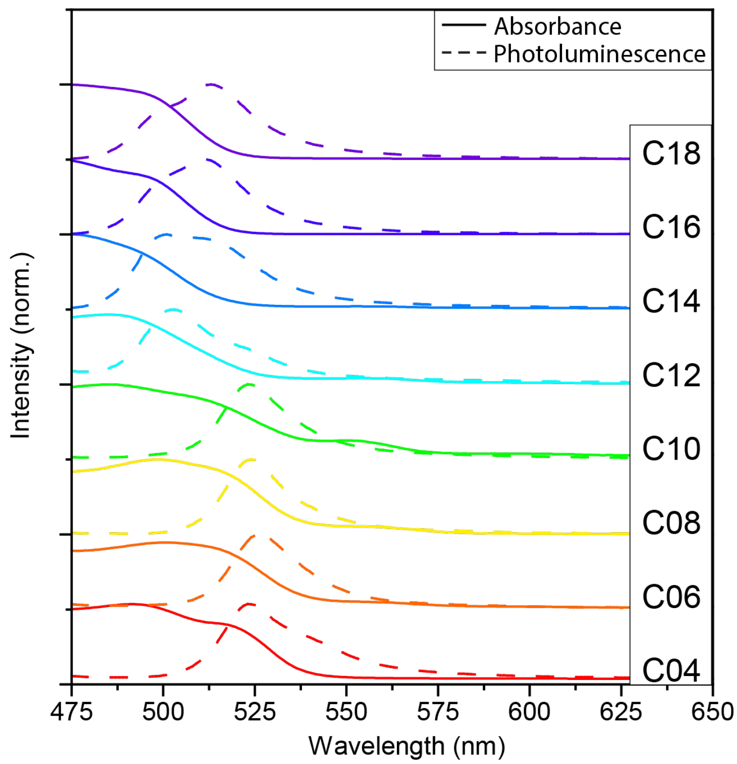
S1. Absorption and Photoluminescence of $(C_n)_2PbI_4$ poly-crystals – Absorption spectra (solid) and Photoluminescence spectra (PL, dashed) of 2D poly-crystals with C4-C18 spacers, showing double PL peaks. The lower energy PL peaks match well with the absorption onset while the higher energy PL peaks are correlated with the second feature in the absorption spectra. As the spacers becomes longer the separation between the peaks is narrowed.



S2. Absorption and Photoluminescence of $(C_n)_2PbI_4$ colloids – Absorption spectra (solid) and Photoluminescence spectra (PL, dashed) of 2D colloids with C4-C18 ligands. In the absorption spectra we observe scattering signal, monotonic increase of the intensity below the band gap caused by aggregation. The scattering is stronger for the short ligand samples indicating their lower colloidal stability, as expected. For all samples, the PL features a single peak, well correlated with the absorption peak.



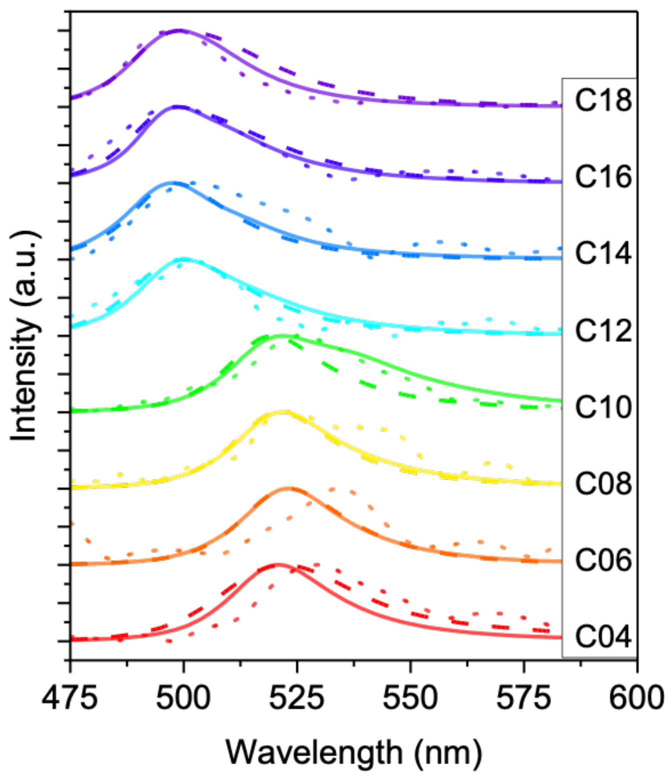
S3. Absorption and Photoluminescence of $(C_n)_2PbI_4$ thin films – Absorption spectra (solid) and Photoluminescence spectra (PL, dashed) of 2D thin films with C4-C18 spacers. For shorter spacers a single PL is observed matching with the absorption shoulder and highlighting the 'colloidal' nature of these films. For the longer spacers a seconda, lower energy, peak is observed highlighting the more 'bulk-like' nature of the films.



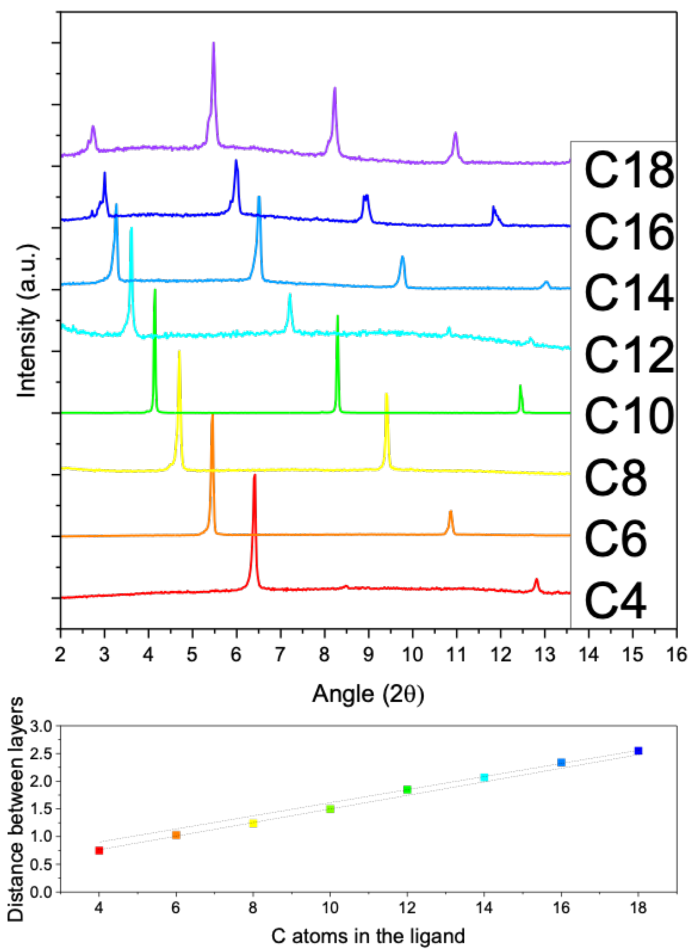
S4. Summary of structural and optical parameters of $(C_n)_2PbI_4$ samples – This table summarize some important properties of 2D metal halide perovskites based on the spacer and morphology. d is the interlayer distance extracted from pXRD measurements. The 'exciton' energy is extracted from the first peak in the absorbance spectra. PL energy is extracted from the peak energy of the PL spectra (or two peaks in case of poly-crystals). Average lifetime is extracted from the fitting of the TRPL measurements.

Ligand length	d (nm)	Exciton (nm)			PL (nm)				Average lifetime (ns)		
		Poly-Crystals	Thin Films	Colloids	Poly-Crystals		Thin Films	Colloids	Poly-Crystals	Thin Films	Colloids
					PL Peak 1	PL Peak 2					
4	0.75	535	518	523	515	561	524	526	0.445	0.422	0.627
6	1.03	533	518	519	517	548	526	526	0.381	0.457	0.954
8	1.24	528	518	517	517	546	524	524	0.464	0.965	0.610
10	1.50	526	517	519	517	540	523	525	0.494	0.459	0.433
12	1.85	502	490	491	493	518	503	496	0.562	0.224	0.669
14	2.07	500	495	491	497	508	500	500	0.498	0.473	0.350
16	3.24	495	496	491	494	505	500	497	0.704	0.588	0.892
18	2.55	496	495	491	493	504	499	497	0.428	0.411	0.489

S5. Photoluminescence spectra of $(C_n)_2PbI_4$ poly-crystals, colloids, and thin films extracted from the TRPL measurements – PL spectra of poly-crystals (solid), colloids (dashed), and thin films (dots) of 2D metal halide perovskites with C4-C18 spacers. These spectra were extracted from the TRPL measurements, the samples were excited by a strong pulsed laser in 90 degrees orientation. The PL spectra differ from the standard steady-state PL measurements featuring mainly a single peak. This is associated with the excitation orientation and power, highlighting mostly the 'surface' features of these samples.

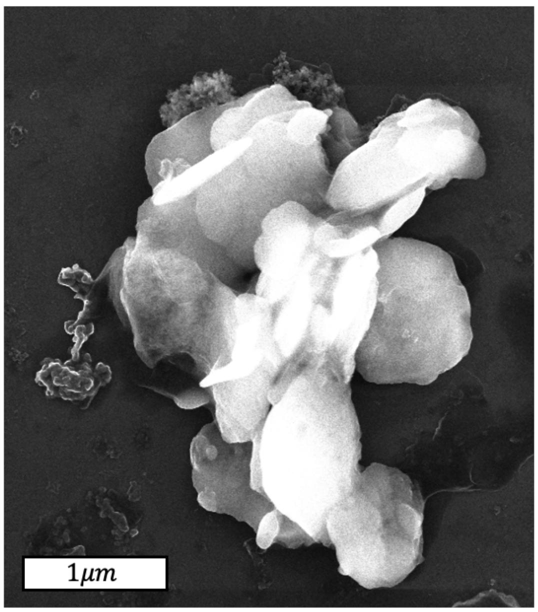


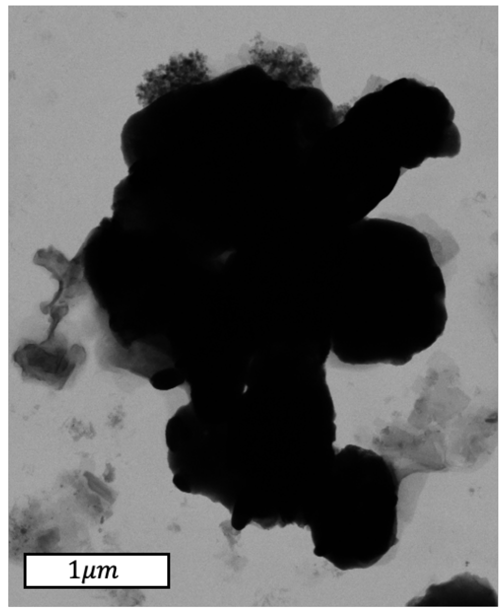
S6. Powder X-ray diffraction spectra of $(C_n)_2PbI_4$ poly-crystals – Room temperature pXRD spectra of 2D metal halide perovskite poly-crystals with C4-C18 spacers. A clear shift of the peaks towards lower 2-theta values (bigger unit cell) is observed as the spacers are elongated as expected. Plotting the angle of the first diffraction peak as function of the number of carbons in the chain shows two linear trends – one for short spacers C4-C10 and the second for longer spacers (C12-C18). The change between C10 to C12 is associated with the discussed phase transition from the orange (orthorhombic) phase to the yellow (monoclinic) phase.



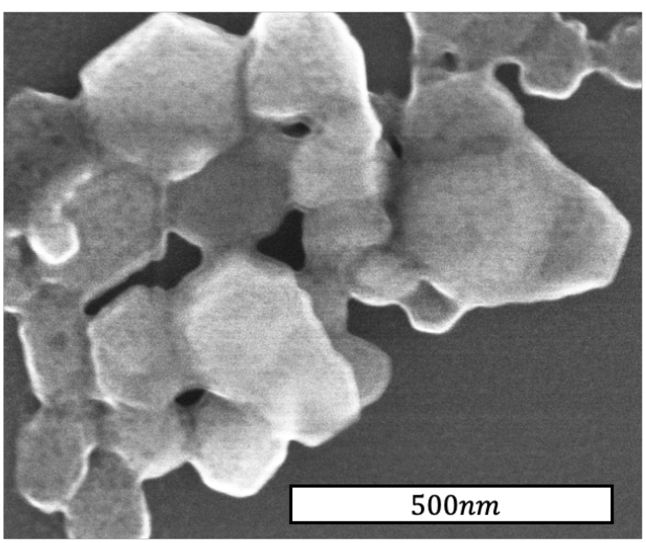
S7. Scanning electron microscopy images $(C_n)_2PbI_4$ colloids – Representative SEM images (Left – bright field, Right – Transmission) of 2D metal halide perovskites with C4, C10, and C18 ligands. The main difference we observed is in the thickness of the crystals. For the shorter ligands the transmittance image is black indicating the thicker nature of the crystals. While for the longer ligands the crystals appear transparent in the transmission images, indicating the thinner nature of these crystals.

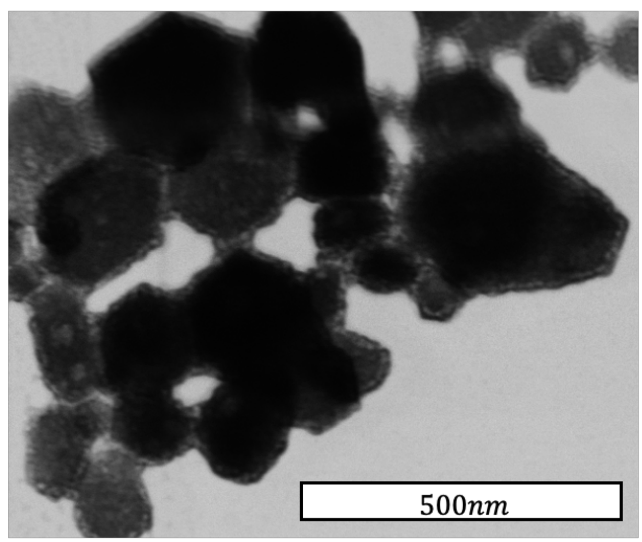
SEM images of $(C4)_2PbI_4$ colloids





SEM images of $(C10)_2PbI_4$ colloids





SEM images of $(C16)_2PbI_4$ colloids