

## Supporting Information for

### Evolution of Self-Trapped Exciton Emission Tuned by High Pressure in 2D All-Inorganic Cesium Lead Halides Nanosheets

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#### **Materials:**

Cesium acetate (CsOAc, 99.9%), octadecene (ODE, 95%), oleic acid (OA, 99.5%), n-octylamine (OLAm, 99.5%), lead acetate trihydrate (Pb(OAc)<sub>2</sub>·3H<sub>2</sub>O, 99.99%), ammonium chloride (NH<sub>4</sub>Cl, 99.99%), toluene (ACS grade, Fischer), ethylacetate (99.99%). All the materials were purchased from Macklin and all were used directly.

#### **Preparation of Cs precursor:**

CsOAc (1.25mmol) was mixed with OLAm (12mL) and OA(2mL) in a 50mL 3-neck round-bottom flask, the resulting solution was heated to 120°C under vacuum, until all power drug was absolutely dissolved. After that, the solution was kept 150°C under vacuum for use.

### **Synthesis of CsPbCl<sub>3</sub> perovskite NSs:**

Pb(OAc)<sub>2</sub>·3H<sub>2</sub>O (0.6 mmol), NH<sub>4</sub>Cl (1 mmol), OA(1.5mL), ODE(10mL) were added to a 100 mL round-bottom 3-neck flask, and refilled with N<sub>2</sub>, then heat the solution to 150°C for 1h. Finally, the solution was raised to 200°C, when the temperature was stable, Cs precursor (3.5 ml) were subsequently injected swiftly to the reaction solution and maintained for 1 minute, followed by an ice bath. Finally, the obtained perovskite QDs were repeatedly precipitated, centrifuged, and suspended in toluene.

### **High-pressure experiments:**

All of highpressure experiments was carried out by using a symmetric diamond anvil cell (DAC). A T301 steel gasket with a thickness of 50 μm and 0.15 mm diameter hole in the center part served as the sample chamber. The R1 fluorescence line of a ball was used to determine the actual pressure. The pressure-transmitting medium is silicone oil.

High-pressure crystal structure was measured by using XRD (Rigaku Synergy Custom FR-X diffractometer) with Mo K $\alpha$  radiation. The measured XRD patterns were fitted with the Rietveld method on GSAS software.

High-pressure Raman spectra were performed on a Renishaw inVia Raman Microscope with an Ar<sup>+</sup> 830 nm, 1800 g/mm grating.

Ultra-low fluorescence diamonds were used in high-pressure PL measurements. The PL and absorption spectra under different pressure were measured by a home-made optical measurement system. Excitation wavelength was 360 nm in PL measurements.

The fiber spectrometer of Ocean Optics QE65 Pro and a halogen light source were used to detect the absorption spectra.

In order to separate the excitation light (360 nm) and the fluorescence signal during the fluorescence measurements, a Longpass Dichroic Mirror with a cut-on wavelength of 380 nm was used as a Beamsplitter. The transmission band of the Beamsplitter is 380 nm-790 nm, therefore, the fluorescence signal with wavelength above 790 nm will be rapidly attenuated, resulting in the phenomenon of peak cut.

### Computational Details

Our calculations were performed by density functional theory (DFT). PBE was used as the exchange correlation functional. The Vienna ab initio simulation package (VASP) was employed. The cutoff for the plane-wave was set to be 500 eV and the convergence criteria of energy set as  $10^{-4}$  eV. The Brillouin zone use the Monkhorst-Pack  $6 \times 6 \times 6$  k-mesh. HSE06 hybrid functional was adopted to estimate the actual band gap. The lattice constants used in the calculations are all derived from experiments.

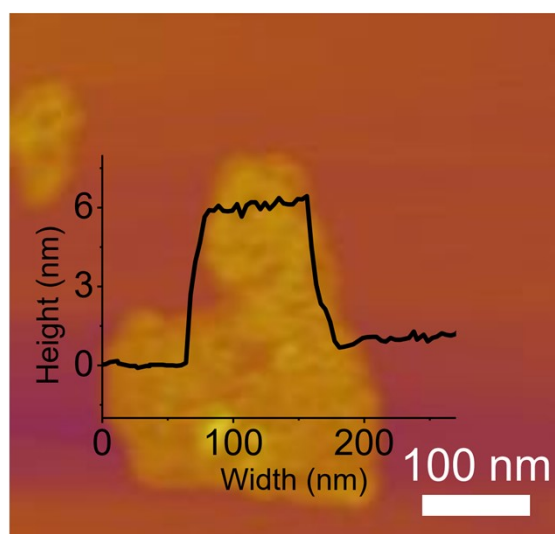


Figure S1. AFM image of CsPbCl<sub>3</sub> perovskite nanosheets.

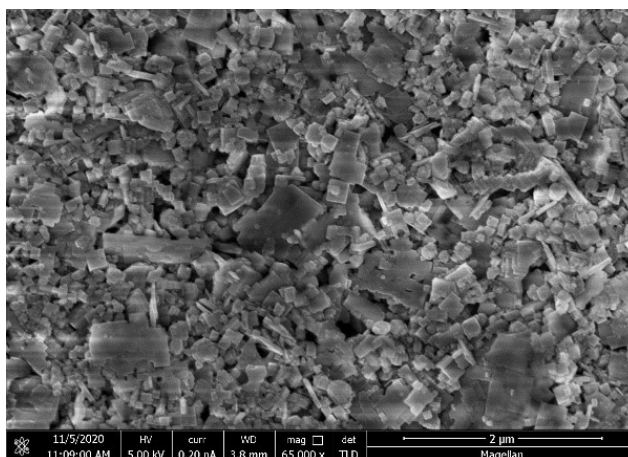


Figure S2. SEM image of the CsPbCl<sub>3</sub> perovskite nanosheets.

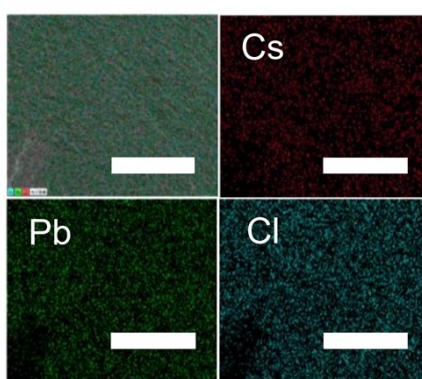


Figure S3. Elemental mapping images of CsPbCl<sub>3</sub> perovskite nanosheets. All scale bars are 2.5 μm.

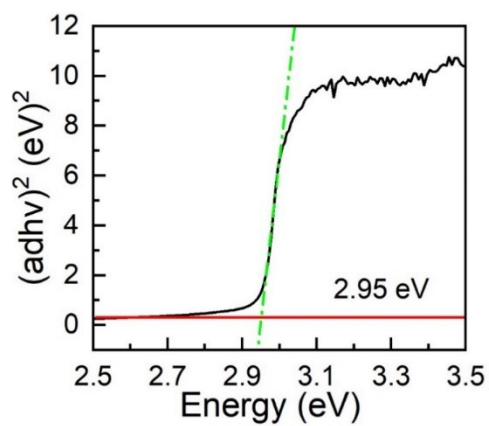


Figure S4. Band gap of CsPbCl<sub>3</sub> perovskite nanosheets. The bandgap of the sample is quantified from the Tauc plots of  $(\alpha hv)^2$  versus  $h\nu$ , where  $\alpha$  is the absorption coefficient,  $d$  is the sample thickness, and  $h\nu$  is the photon energy.

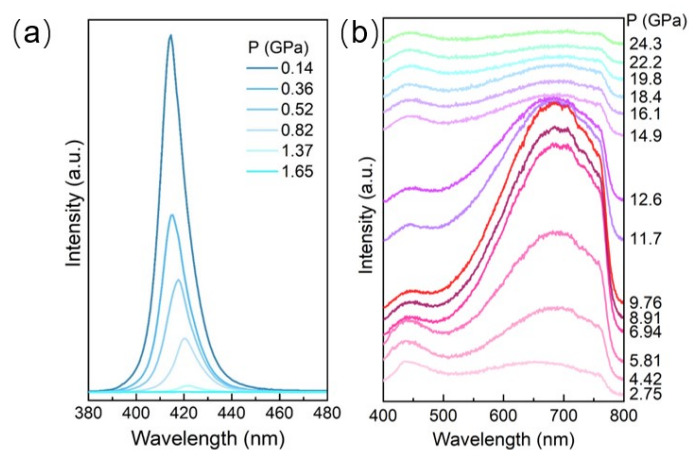


Figure S5. PL spectra from ambient pressure to 1.65 GPa and from 2.75 to 24.3 GPa, respectively.

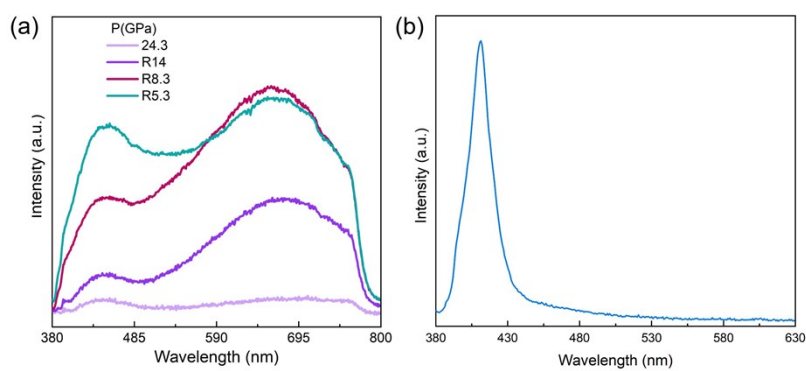


Figure S6. (a) PL spectra of CsPbCl<sub>3</sub> perovskite nanosheets collected after decompression.

(b) PL spectra of CsPbCl<sub>3</sub> perovskite nanosheets upon the release of pressure.

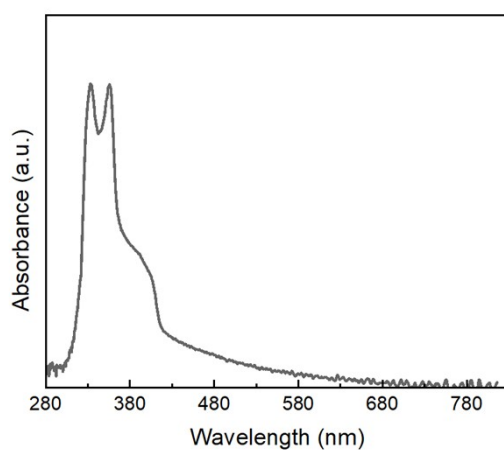


Figure S7. Absorption spectrum of CsPbCl<sub>3</sub> perovskite nanosheets upon the release of pressure.

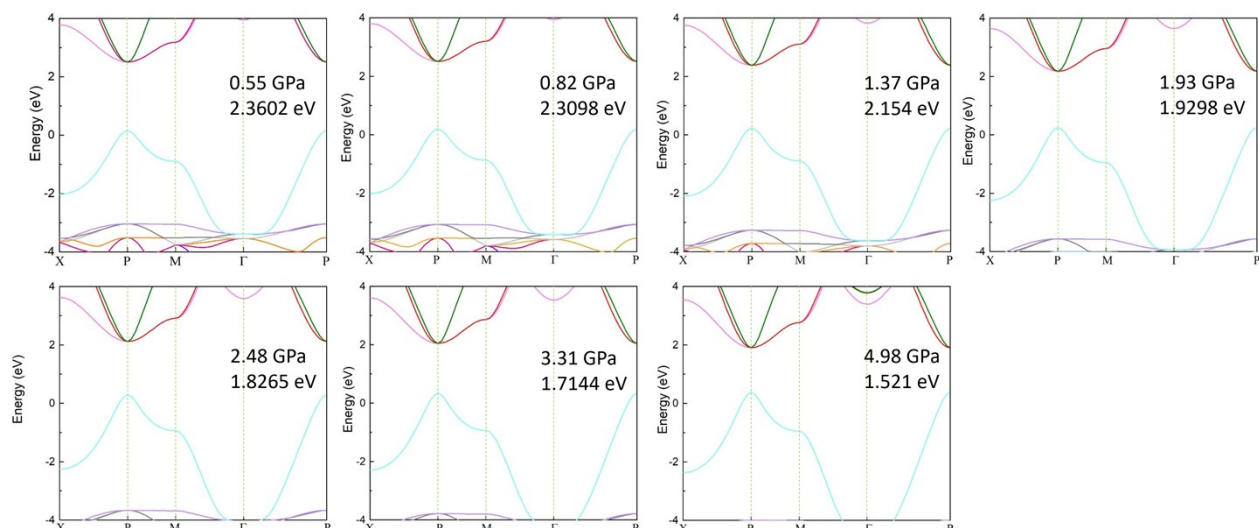


Figure S8. The calculated band structures of CsPbCl<sub>3</sub> perovskite nanosheets with representative pressures at room temperature.

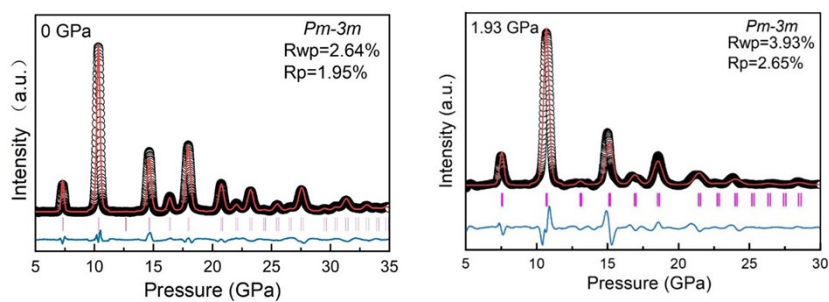


Figure S9. Rietveld analysis result of CsPbCl<sub>3</sub> perovskite nanosheets before and after structure change, respectively.

**Table S1.** Chromaticity coordinates (CIE) at selected pressures for CsPbCl<sub>3</sub> perovskite nanosheets.

Pressure	CIE x	CIE y	Pressure	CIE x	CIE y
0 GPa	0.1698	0.0067	0.14 GPa	0.1812	0.033
0.36 GPa	0.1685	0.0098	0.52 GPa	0.1689	0.0077
0.82 GPa	0.1684	0.0082	1.37 GPa	0.163	0.0304
2.2 GPa	0.1895	0.2052	3.3 GPa	0.241	0.2203
4.42 GPa	0.3793	0.3175	5.81 GPa	0.4125	0.3394
6.94 GPa	0.477	0.3711	8.91 GPa	0.4758	0.3722
9.76 GPa	0.4842	0.38	11.75 GPa	0.4675	0.3744
12.6 GPa	0.464	0.374	14.9 GPa	0.3869	0.3314

16.06 GPa	0.392	0.3277	18.38 GPa	0.3941	0.3291
19.84 GPa	0.3655	0.3091	22.2 GPa	0.3503	0.2972
24.27 GPa	0.3408	0.2954	Release	0.2019	0.1312

**Table S2.** Lattice constants and cell volume of CsPbCl<sub>3</sub> perovskite nanosheets at different pressures. Release represents the pressure completely released to the ambient conditions.

Pressure(GPa)	a (Å)	V(Å <sup>3</sup> )	Pressure(GPa)	a (Å)	V(Å <sup>3</sup> )
0	5.576	173.332	0.55	5.522	168.341
0.82	5.506	166.928	1.37	5.460	162.803
1.93	5.399	157.414	2.48	5.373	155.114
3.31	5.344	152.616	4.98	5.300	148.877
6.94	5.273	146.613	8.63	5.256	145.199
10.52	5.234	143.384	Release	5.580	173.741