Supplementary Information

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Title: "Exciton-Phonon interaction in quasi-two dimensional layered (PEA)₂(CsPbBr₃)_{n-1}PbBr₄ perovskite"

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PXRD data and analyzation:



Fig.S.1: Low angle PXRD of (a) quasi-2D mixed phase (PEA)₂(CsPbBr₃)_{n-1}PbBr₄, (b) pure phase 2D (PEA)₂PbBr₄ perovskite showing formation of 2D PbBr₄ layer.

Three primary PXRD peaks were observed in quasi-2D mixed phase $(PEA)_2(CsPbBr_3)_{n-1}PbBr_4$ and pure phase 2D $(PEA)_2PbBr_4$ perovskite. These three peaks came from (002), (004) and (006) diffraction of PbBr_4 octahedrons [1], which were direct proof of 2D structure formation in our samples.

TRPL at room temperature:



Fig.S.2: (a) TRPL of quasi-2D (PEA)₂(CsPbBr₃)_{n-1}PbBr₄ perovskite at 407 nm (n=1 energy level); (b) carriers' transfer and recombination diagram derived by TRPL

The room temperature time resolved PL (TRPL) spectrums were taken at the peak wavelengths corresponding to the n=1-4 and bulk (PEA)₂ (CsPbBr₃)_{n-1} PbBr₄ perovskite. Each TRPL could be perfectly fitted by double exponential decay models with one slow channel (10ns-30ns) and another fast channel (1ns-3ns), as shown in Fig.S.2(a). The fast channel was ascribed with the carrier recombination process. The slow channel could be ascribed with the energy funnel process. Because the recombination process was prompt in our quasi-2D material, five distinct PL peaks can be detected by the integrated PL measurements. Therefore, we concluded that the recombination process dominated in our quasi-2D samples, while energy funnel was not the dominating factor.

• n=1 1.0 Intensity (a.u.) 0.95 Intensity (a.u.) Intensity (a.u.) 0.8 0. 0.90 0.7 0.6 0.5 0.85 0.80 0.75 0.70 0.65 0.4 0.3 (b) • (c) (a 0.12 0.2 0.60 100 150 200 250 Temperature (K) 00 150 200 25 Temperature (K) 150 200 250 Temperature (K) 250 300 300 300 100 n=4 1.1 n=3 . 0 1 1. 1.0 0.9 0.8 0.7 0.6 0.5 0.4 0.3 0.9 0.8 0.7 0.6 0.5 0.4 0.3 0.2 0.1 Intensity (a.u.) Intensity (a.u.) Intensity (a.u.) 0.8 0.7 0.6 0.5 0.4 0.3 0.2 0.1 0.0 F 66.2 me (f) (e) (d) 0. 100 150 200 Temperature (K) 100 150 200 250 Temperature (K) 300 300 250 100 150 200 250 Temperature (K) 300 50 50

PL intensity quenching fitting:

Fig.S.3: Normalized PL intensities of (PEA)₂(CsPbBr₃)_{n-1}PbBr₄ with various n layer numbers.

The intensity quenching of (PEA)₂(CsPbBr₃)_{n-1}PbBr₄ was fitted by equation[1]:

$$I(T) = \frac{I(0)}{D \cdot exp\left(-\frac{E_b}{kT}\right) + 1}$$
(1)

where D represents the non-radiative density, E_b was the exciton binding energies.

TDPL of pure phase 2D (PEA)₂PbBr₄:



Fig.S.4: (a) photoluminescence (PL) spectrum of pure phase 2D (PEA)₂PbBr₄ perovskite at 50K;
(b) monotonous blueshift of PL peak with temperature; (c) intensity quenching with temperature;
(d) width broadening by temperature by exciton-phonon coupling.

References:

- [1]. Liang, Y.; Shang, Q.; Wei, Q.; Zhao, L.; Liu, Z.; Shi, J.; Zhong, Y.; Chen, J.; Gao, Y.; Li, M.; et al. Adv. Mater. 2019, 1903030.
- [2]. Long, H.; Peng, X.; Lin, K.; Xie, L.; Lu, J.; Zhang, B.; Ying, L.; Wei, Z. Applied Physics Express 2019, 12 (5), 052003.