Supplementary Information

Hot Carrier Cooling and Direct Observation of Electron-Phonon Coupling in Two-Dimensional Butylammonium Lead Iodide Perovskites

Vanga Ravali,[1] Ram Ratan,[2] E. Siva Subramaniam Iyer,[2] Tufan Ghosh[1,*]

[1]Department of Chemistry, School of Advance Sciences, VIT-AP University, Andhra Pradesh – 522237, India

[2] School of Chemical and Materials Sciences, Indian Institute of Technology Goa, Ponda, Goa 403401, India

[*]Email: tufan.ghosh@vitap.ac.in

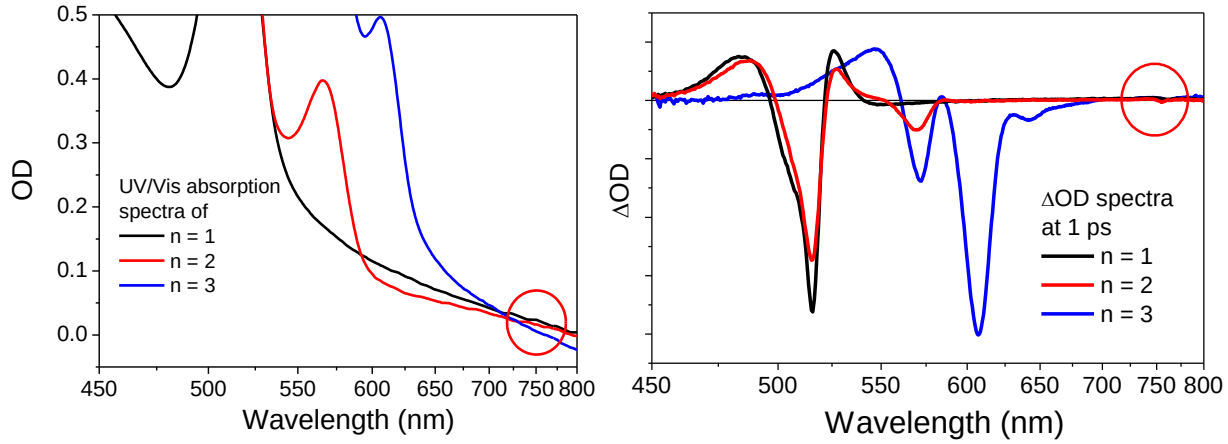

Figure S1: (a) UV-visible spectra and (b) TA spectra at 1 ps of 2D perovskites with n = 1, 2, and 3 layers. Red circles in each panels highlights the spectral changes around 750 nm.

Finite difference Spectra:

In this method, the ΔOD spectrum at a particular pump-probe delay is subtracted from that of a different pump-probe delay, as given in the following equation[1]:

$$\Delta\Delta\mathrm{OD}(\lambda, t, \Delta t) \equiv \Delta\mathrm{OD}\left(\lambda, t + \frac{\Delta t}{2}\right) - \Delta\mathrm{OD}\left(\lambda, t - \frac{\Delta t}{2}\right) \quad \ldots\ldots\ldots \text{Eq (1)}$$

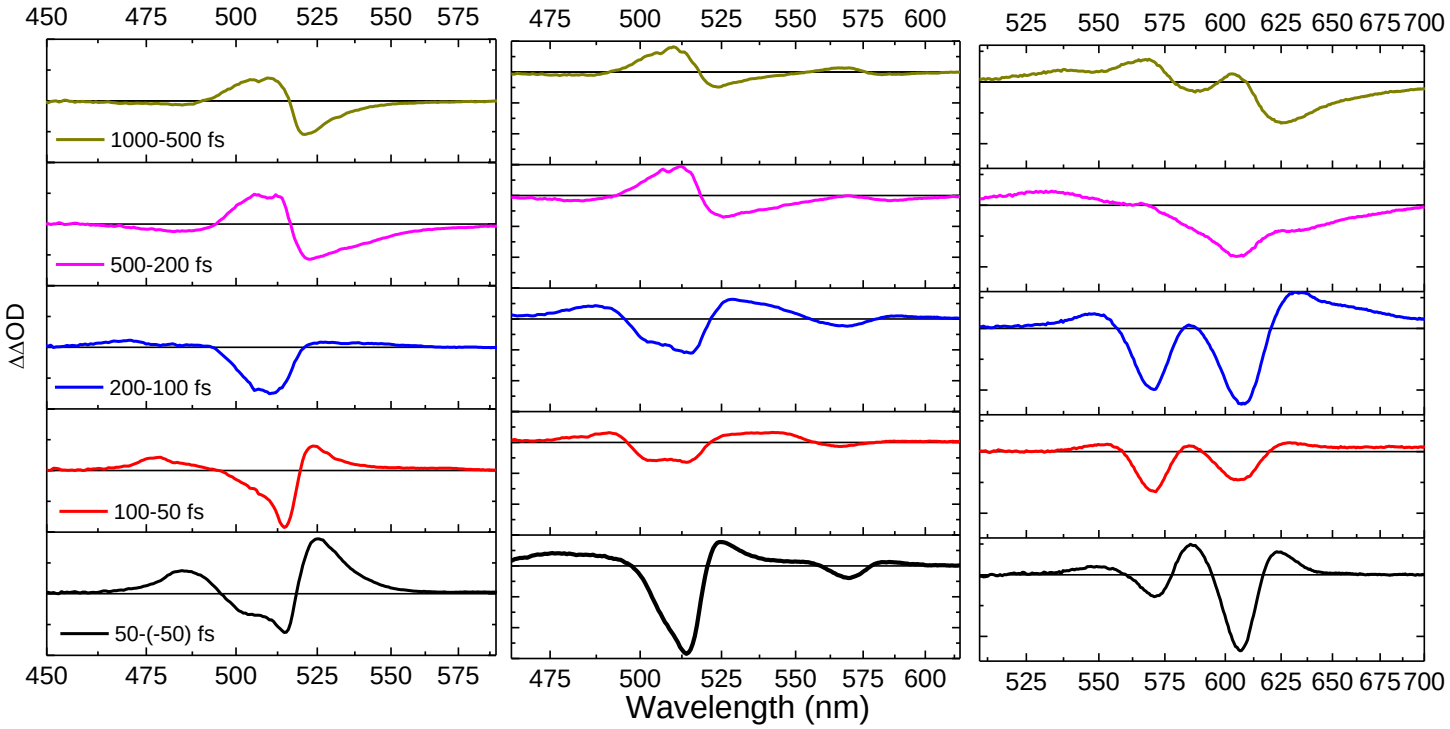

Figure S2: Finite difference spectra calculated from the TA data of $(BA)_2(MA)_{n-1}Pb_nI_{3n+1}$ perovskites with (a) n = 1, (b) n = 2, and (c) n = 3 layers.

Table S1: Transient decay time constants obtained global analysis of 2D layered perovskite thin films.

No. of layers, n	$n_0(cm^{-3})$	$\tau_1(ps)$	$\tau_2(ps)$
1	1.2×10^{18}	0.367±0.021	13.05±2.17
2	1×10^{18}	0.395±0.021	10.71±1.95
3	1.9×10^{18}	0.484±0.051	26.27±3.81

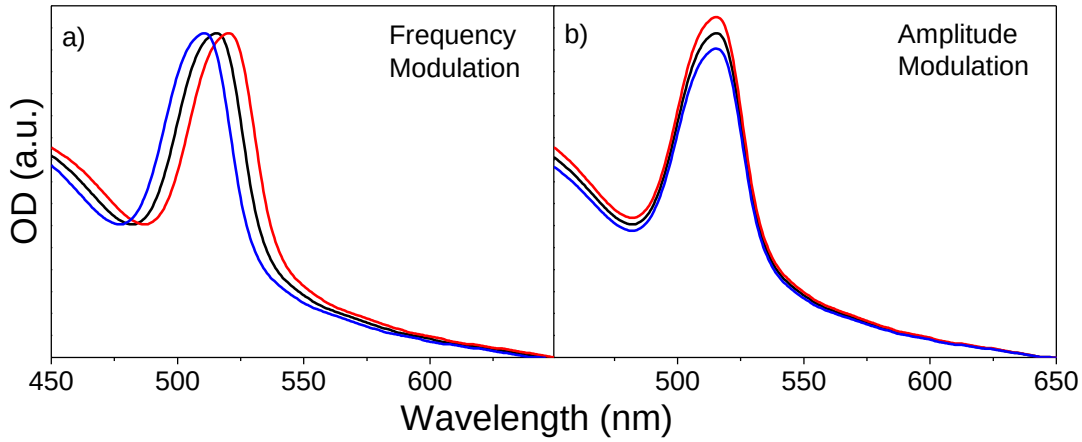

Figure S3: Schematic representation of (a) frequency modulation and (b) amplitude modulation of absorption spectra for n = 1 perovskite films.

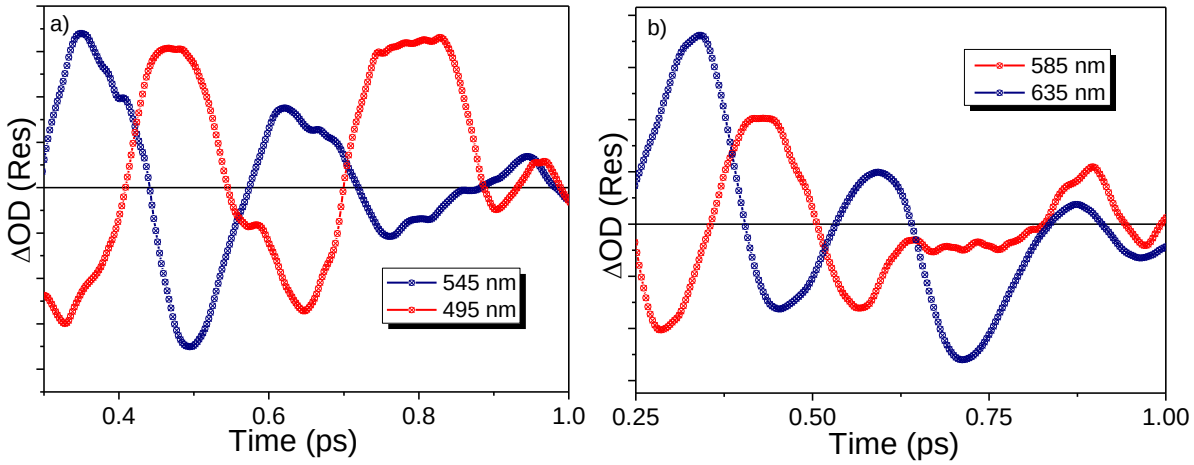

Figure S4: Residual modulations plotted at both sides of the exciton peak of $(BA)_2(MA)_{n-1}Pb_nI_{3n+1}$ perovskites films with (a) n = 1, and (c) n = 3 layers.

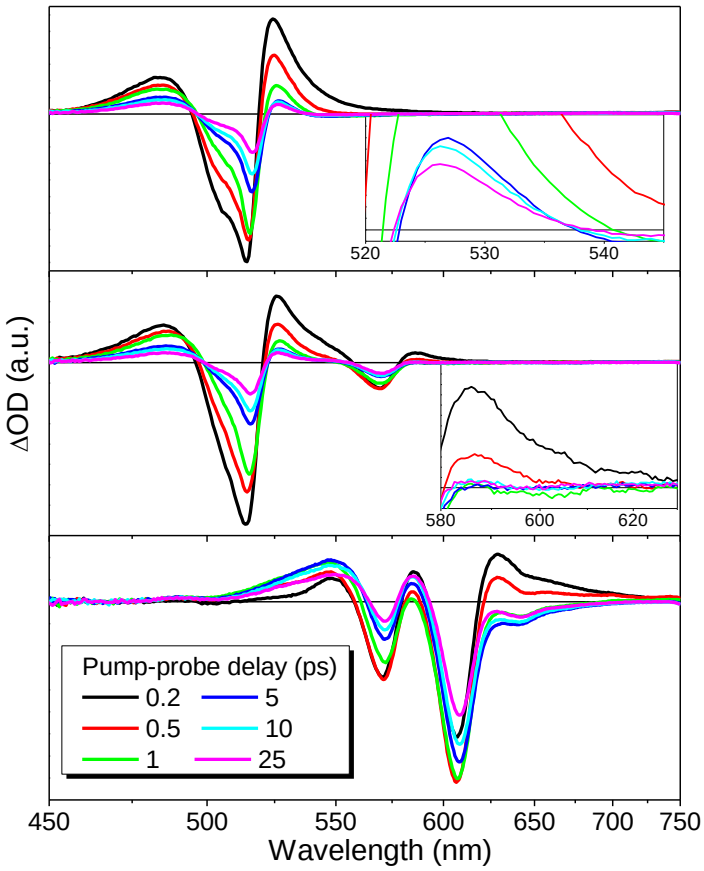

Figure S5: Transient differential absorption spectra of the 2D perovskite thin films up to pump-probe delays of 25 ps for (a) n = 1, (b) n = 2, and (c) n = 3 inorganic layers.

Characterization of the 2D perovskites by powder- and thin film XRD:

The 2D layered perovskite materials have been characterized by using the p-XRD technique. For n=1 (BA_2PbI_4) perovskite thin film and powder show reflections corresponding to diffraction planes of (002), (004), (006), and (008), which represents growth of crystals along the direction of (110) and reflecting (00l) direction. Here both in powder and thin films the crystal growth is in same directions. Whereas, when MA cation introduced, crystal tries to expand in all three directions (that is planar as well as vertically). For n=2 ($BA_2MAPb_2I_7$) powder and crystals grown along (h0l) plane, so we observed the reflections of (0k0) [that is (040), (060), (080), (0100), (0120)] planes along with (111), (202) and (222). The n=3 ($BA_2MA_2Pb_3I_{10}$) powder sample shows reflections

(0k0) [(040), (060), (080), (0160)] and (111) planes, when it comes to thin film the vertical crystal growth dominated the (0k0) reflections so only (111) and (222) planes are reflected. [2,3,4].

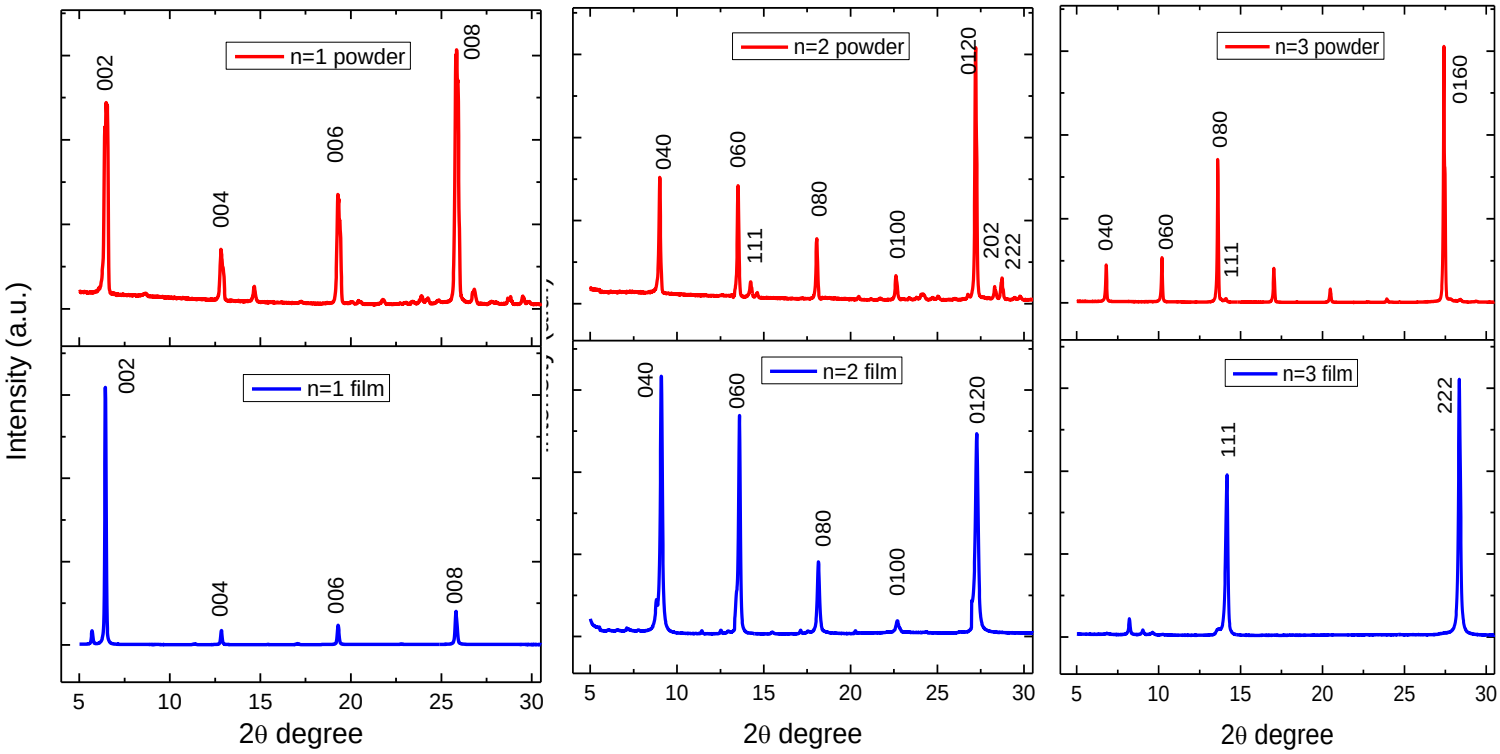

Figure S6: Powder- and thin film-XRD data of 2D perovskites with n = 1 (left panels), 2 (middle panels), and 3 (right panels).

REFERENCES:

[1] T. Ghosh, S. Aharon, L. Etgar and S. Ruhman, *J. Am. Chem. Soc.*, 2017, **139**, 18262–18270.

[2] D. H. Cao, C. C. Stoumpos, O. K. Farha, J. T. Hupp and M. G. Kanatzidis, *J. Am. Chem. Soc.*, 2015, **137**, 7843–7850.

[3] C. C. Stoumpos, D. H. Cao, D. J. Clark, J. Young, J. M. Rondinelli, J. I. Jang, J. T. Hupp and M. G. Kanatzidis, *Chem. Mater.*, 2016, **28**, 2852–2867.

[4] S. M. Gowdru, J.-C. Lin, S.-T. Wang, Y.-C. Chen, K.-C. Wu, C.-N. Jiang, Y.-D. Chen, S.-S. Li, Y. J. Chang and D.-Y. Wang, *Nanomaterials*, 2022, **12**, 1816.