Femtosecond Laser Pulse Induced Third-Order Optical Nonlinearity in Quasi 2D Ruddlesden-Popper Perovskite Film Deciphered Using Z-Scan

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Experimental:

GIWAXS measurements have been performed at the Austrian SAXS beamline at the ELETTRA synchrotron in Trieste (Italy) at a photon energy of 8 keV.[1] The beam size was set to 0.6x0.2 mm^2 (HxV) while the sample to detector (Pilatus3 1M, Dectris) distance was determined to be 245 mm using a silver behenate as a reference pattern. Measurements have been performed in air at the incident angle of 0.22º and 1.3º with an exposure time of 10 s. The patterns have been corrected for the fluctuations of the primary intensity.[2] The simulations of the GIWAXS patterns have been performed with the GIXSGUI toolbox.[3] The film thickness evaluation has been performed using IGOR Pro (IGOR Pro 7.0.8.1, WaveMetrics). The average grainsize has been calculated as proposed in for (121), (141), (220), and (240) diffraction spots and averaged.[4] The $CsPbBr_3$ powder diffraction pattern has been simulated with Diamond 3.2i (Crystal Impact). The average grain size has been estimated using the Scherrer formula which accounts for the geometric and divergence smearing of the diffraction spot in the detector

plane.[5] The energy bandwidth smearing has been neglected since Si (111) optics is used.[1] The breadth of the most pronounced Bragg spots has been obtained by fitting the corresponding I(2θ) cuts with subsequent averaging of data. No asymmetry in nanocrystal size has been identified.
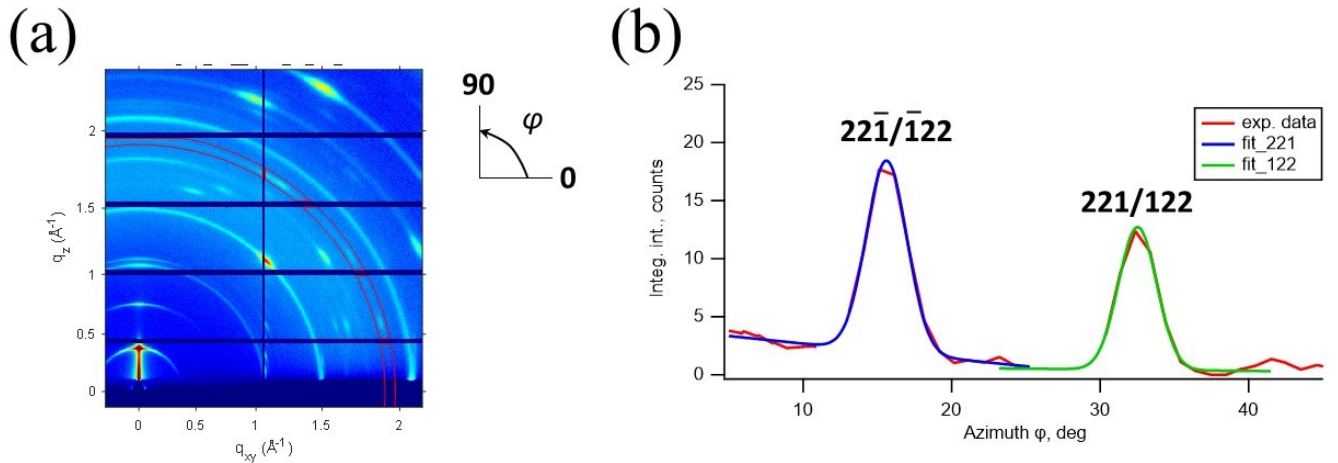

Fig. S1 (a) Raw 2D GIWAXS pattern of $(PEA)_2Cs_4Pb_5Br_{16}$ (RPP) film acquired at an incident angle of 1.3°. (b) The scattered intensity integrated as a function of azimuthal angle φ in the Δq interval between two red arcs depicted in panel (a). Note that prior to the integration Lorentz correction for 2D random orientation of the crystals in the surface has been performed. The contributions of two crystal orientations (see Figure 1d in the main text) have been estimated from the ratio of peak areas (the corresponding fit is shown in panel b). The ratio equals 1.43 or 58.8% to 41.2%.
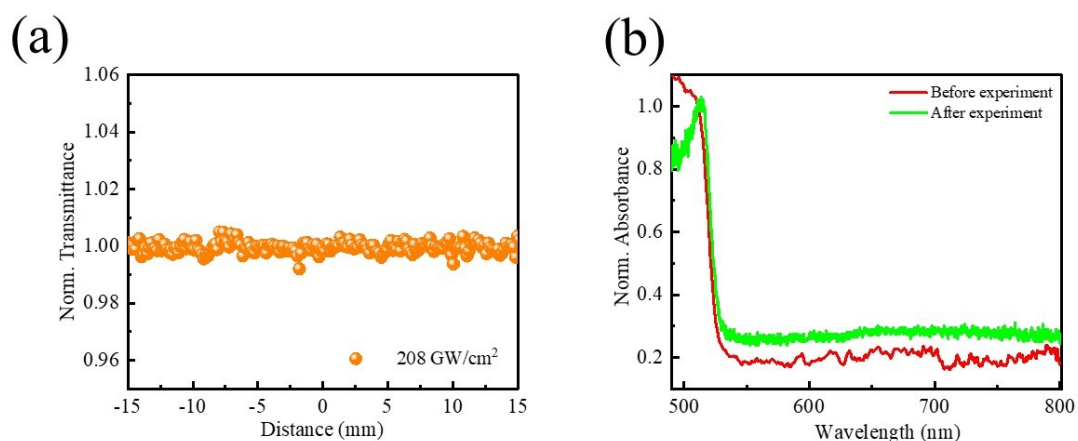
Fig. S2 (a) OA Z-scan curve of cover slip showing no NLO behaviour at 208 GW/cm². (b) Absorption spectrum of the perovskite film (measured at the irradiate area using a fibre optics spectrometer from Ocean Optics) after Z-scan experiments in comparison to absorption spectrum of the whole film before experiments (measured using Lamda 950 UV-Vis spectrophotometer).
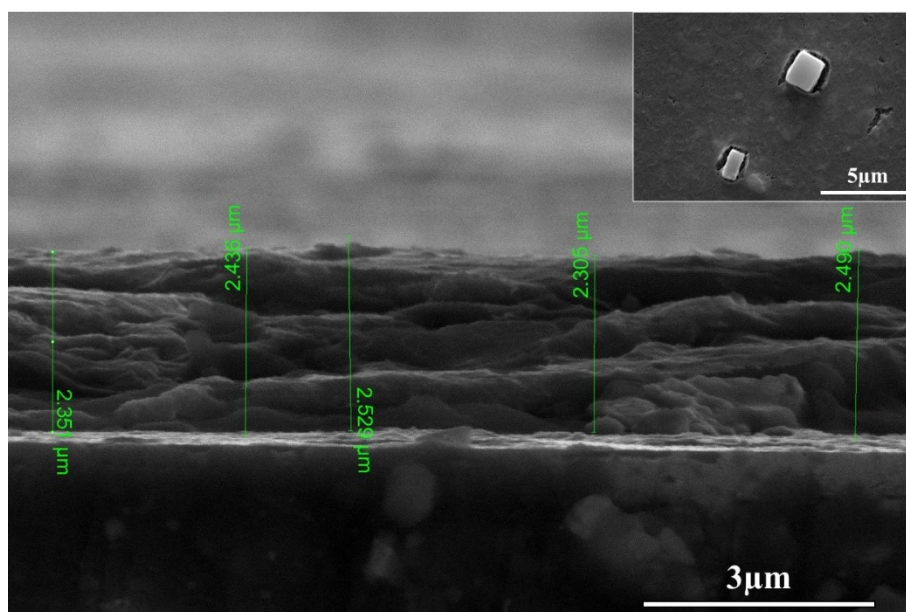

Fig. S3 Cross-sectional SEM image of perovskite film. Inset: SEM image of much thinner perovskite film.

References

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