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Femtosecond induced third-order optical nonlinearity in quasi 2D Ruddlesden–Popper perovskite film deciphered using Z-scan†

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Ruddlesden–Popper perovskite is a class of layered halide perovskite which gained much attention over the past few years in the field of photovoltaics. Herein, we have presented a compendious sketch of third-order nonlinear optical (NLO) properties of (PEA)₂Cs₄Pb₅Br₁₆, a quasi-two-dimensional (2D) perovskite using femtosecond Z-scan technique at 800 nm. Our open aperture (OA) Z-scan measurements unveiled saturable absorption (SA) behaviour of the 2D perovskite with a nonlinear absorption coefficient of $-13.83 \text{ cm GW}^{-1}$, which is an order of magnitude higher than other 2D materials. Nonetheless, the self-defocusing effect has been demonstrated using a closed aperture (CA) Z-scan measurement. High nonlinear absorption coefficient and low saturation intensity (38.81 GW cm^{-2}) infer that (PEA)₂Cs₄Pb₅Br₁₆ perovskite film could be useful for ultrafast photonic applications.

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Introduction

Nonlinear optics has broad applications in laser technology which includes optical parametric amplification, electro-optic switching, frequency doubling and mixing. The study of nonlinear optical (NLO) properties becomes imperative from the technological point of view to design NLO devices and for the understanding of underlying mechanisms governing light-matter interaction. Ultrashort laser pulses can be generated by exploiting the NLO property, saturable absorption (SA) and hence saturable absorber is the key optical component in pulsed lasers. Semiconductor saturable absorber mirror (SESAM) is commercially used as a saturable absorber because of its high stability, but it has disadvantages of complex fabrication processes and limited bandwidth.¹ To develop ultrafast lasers, different NLO materials

have been investigated over recent years.² For example, carbon nanotubes (CNTs) were reported to possess excellent NLO responses. But, this NLO behaviour depends on the distribution of the diameter of CNT.^{3,4} On the other hand, graphene and graphene analogue two dimensional (2D) materials like transition metal dichalcogenides (TMDCs), transition metal monochalcogenides (TMMCs), black phosphorous and related materials have shown application potential in ultrafast photonics because of their striking NLO response.^{5–11}

Recently, perovskites materials have gained much importance in the field of photonics owing to their NLO properties.¹² Li *et al.* reported an SA response from the CsPbBr₃ quantum dots (QDs) with nonlinear absorption coefficient (α_{NL}) -0.35 cm GW^{-1} and saturation intensity (I_{sat}) 11 GW cm^{-2} .¹³ In another study, cubic crystals of CsPbBr₃ have been shown to exhibit two-photon absorption (TPA) responses with a TPA coefficient of $9.7 \times 10^{-2} \text{ cm GW}^{-1}$.¹⁴ Ruddlesden–Popper perovskite (RPP), are quasi-2D perovskites with the general formula $A^*(A)_{n-1}\text{Pb}_n\text{X}_{3n+1}$, where A^* is long-chain aromatic ammonium or alkyl cation, n is the number of octahedral layers in the perovskite-like stack and if n is large (infinity) the resultant structure will be an ordinary 3D perovskite. This class of layered halide perovskite material is on the rise with improved ambient stability.^{15,16} Cao *et al.* and Qiu *et al.* in separate reports demonstrated the role of solvent choice and spin coating conditions in enhancing the stability of 2D perovskite films.^{17,18} Recently, Liang *et al.* reported less than 10% efficiency degradation in 2D Ruddlesden–Popper perovskites

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