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Article

Ultrafast and Nonlinear Optical Properties of Two-Dimensional Mo-Doped Dual Phase Inorganic Lead Halide Perovskite

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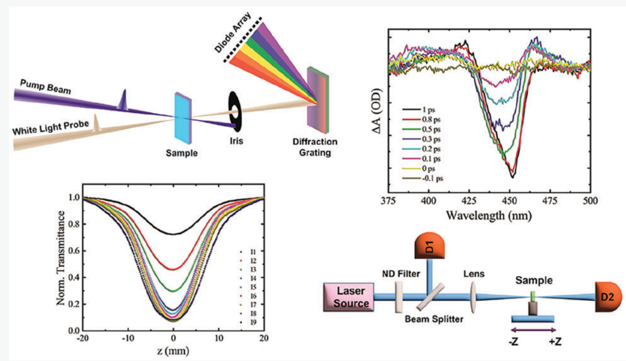
ABSTRACT: Two-dimensional (2D) inorganic halide nanomaterials are finding tremendous attention owing to ease of synthesis, solution processability, superior electronic, optical, and mechanical properties resulting in versatile applications. All inorganic dual phase perovskite composites can have superior optoelectronic properties than a single phase one. In addition to dual phase, doping with various ions may open up new avenues for emerging applications of such materials. In this work, we report on the synthesis of colloidal 2D Mo-doped all inorganic dual phase mix halide perovskite ($CsPbBr_3/CsPb_2Br_5$). The charge carrier dynamics in these nanostructures has been studied with femtosecond transient absorption technique. Our studies reveal that at low pump fluences cold carrier trapping predominantly affects relaxation dynamics at early time. However, Auger recombination of excitons dominates the early time carrier relaxation at high fluences. Additionally, femtosecond *Z*-scan studies have been performed to elucidate the effect of photoinduced charge carriers on two photon absorption (TPA) properties of Mo-doped 2D perovskite. Our results could be useful for designing new low-dimensional perovskites for future linear and nonlinear optoelectronic applications.



1. INTRODUCTION

Nanomaterials find various applications in diverse field of science and technology owing to their superior, sometime outstandingly new properties over their bulk counterparts.[1] These properties emerge due to quantum confinement of the charges and large surface to volume ratio. Beside size-dependent properties, these materials exhibit dimensionality dependent features which have been harnessed in various applications.[2−7] With the continuous development and realization of extensive applications of numerous nanomaterials, a new class of nanomaterials was invented by Schmidt et al.[8] and Protesescu et al.[9] from perovskites.[10] This family of materials possess crystal structure of ABX_3 type where A and B are cations of different sizes and X is an anion. Perovskite nanomaterials exhibit wide variety of excellent properties, e.g., ease of synthesis and upscaling, high defect tolerance, tunable optical properties, high extinction coefficient, long carrier diffusion length, and small exciton binding energy.[11] Among the various perovskite nanomaterials, all inorganic lead halide perovskite (LHP) nanomaterials have attracted tremendous attention after successful demonstration of application possibilities of organic−inorganic halide perovskite nanomaterials in diverse fields.[12,13] All inorganic LHPs, in comparison to organic−inorganic perovskites are found to be more stable.[14−16] High photoluminescence quantum yield (PLQY) (>95%) and narrow PL line width[17,18] with higher stability against moisture and photodegradation make them better candidate for optoelectronic devices such as solar cells, photodetectors, memory devices, light-emitting diodes (LEDs), and lasers.[19] To exploit additional benefits of nanomaterials, dimension can be reduced by retaining original stochiometric ratios of the constituents. The reduction of dimension results in the decrease of dielectric screening between the charges and overlap of electron and hole wave functions increases by allowing them to interact strongly. Consequently, enhanced Coulomb binding energy assists the formation of strongly bound excitons with increased oscillator strength (for transition) which leads to enhancement in absorption coefficient.[20−22] 2D nanomaterials offer extra benefits over their 3D counterparts due to increased specific surface area which is desirable for efficient charge collection in optoelectronic devices.[23−27] Though $CsPbBr_3$ possesses excellent optoelectronic properties further improvements have been achieved by synthesizing dual phase $CsPbBr_3$−$CsPb_2Br_5$ perovskite. These dual phases of tetragonal $CsPb_2Br_5$ and orthorhombic $CsPbBr_3$ can be achieved in single chemical

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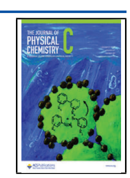

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