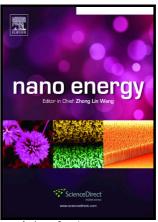
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www.elsevier.com/locate/nanoenergy

PII: S2211-2855(17)30595-5

DOI: http://dx.doi.org/10.1016/j.nanoen.2017.09.048

Reference: NANOEN2224

To appear in: Nano Energy

Received date: 14 July 2017 Revised date: 9 September 2017 Accepted date: 25 September 2017

Cite this article as: Can Huang, Yisheng Gao, Shuai Wang, Chen Zhang, Ningbo Yi, Shumin Xiao and Qinghai Song, Giant blueshifts of excitonic resonances in two-dimensional lead halide perovskite, *Nano Energy*, http://dx.doi.org/10.1016/j.nanoen.2017.09.048

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ACCEPTED MANUSCRIPT

Giant blueshifts of excitonic resonances in two-dimensional lead halide perovskite

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Abstract:

Two-dimensional (2D) methylammonium lead halide perovskites (MAPbX₃) have gained intensive research attention in past two years. Due to their quantum and dielectric confinements, 2D MAPbX₃ exhibited giant exciton binding energy, increased bandgap, and blueshifts of photoluminescence. Some of these novel characteristics have been successfully utilized to improve the performances of optoelectronic devices. However, up to now, the studies of quantum confinements are restricted within the linear region. Lots of important effects in quantum systems such as the exciton-exciton interaction have never been considered. Herein we synthesized the 2D MAPbBr₃ nano-sheets with layer number n = 7-10, 5, 3, 1 and characterized their nonlinear optical properties under high excitation density. Due to the strong excitonic resonance, we have achieved significantly increased third-harmonic generation (THG) from the 2D MAPbBr₃ nano-sheets. The deduced χ^3 of few-layer MAPbBr₃ nano-sheets was more than 50 times larger than the bulk MaPbBr₃ microblocks. While the THG was strongly dependent on the excitonic resonance, we found that their peak wavelengths were different from the ones of excitons in linear absorption

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