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Reversible nature of photo-induced phase segregation and origin of long carrier lifetime in triple cation mixed halide perovskite films

Subodh K. Gautam^{1,*}, Minjin Kim², Douglas R. Miquita^{1,3}, Jean-Eric Bouree², Bernard Geffroy^{2,4} and Olivier Plantevin¹

¹ Université Paris-Saclay, CNRS, Laboratoire de Physique des Solides, 91405, Orsay, France.

² LPICM, CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, route de Saclay, 91128 Palaiseau, France.

³ Centro de Microscopia - Universidade Federal de Minas Gerais, Belo Horizonte, MG, 31270-901- Brasil

⁴ Université Paris-Saclay, CEA, CNRS, NIMBE, LICSEN, 91191, Gif-sur-Yvette, France

Mixed-halide based hybrid perovskite semiconductors have attracted tremendous attention as a promising candidate for high efficient photovoltaic and light-emitting devices [1]. However, these advanced perovskite materials may undergo phase-segregation under light illumination due to halide ion migration and affecting their optoelectronic properties [2, 3]. In this contribution, we report such phase segregation effect in triple-cation mixed-halide perovskite film when subjected to photo-excitation and quantitatively analyze the processes that occur during phase segregation [4]. We highlight the relationship between photo-induced phase segregation and unusual increase in carrier lifetime in mixed halide perovskite under illumination ($> 1 \mu\text{s}$). Laser excitation induced halide ion migration lead to formation of smaller-bandgap iodide-rich and larger-bandgap bromide-rich domains which yield to red-shift in photoluminescence. The segregated iodide-rich domains efficiently trap the photo-excited-carriers where they are long lived before recombination, revealing their dominant role in the origin of the unusual long carrier lifetime. Interestingly, these photo-induced changes are fully reversible and thermally activated when laser-excitation is turned off measured in temperature range of 270K-330K. A significant difference in activation energies for halide ion migration is observed during photo-excitation and recovery process under dark. In addition, temperature-dependent PL studies (10 K - 300 K) have been performed for better understanding of the role of exciton-phonon coupling to interpret the phase segregation driving forces. These findings will help to understand the key issues in the perovskite materials for the development of efficient solar cells and optoelectronic devices.

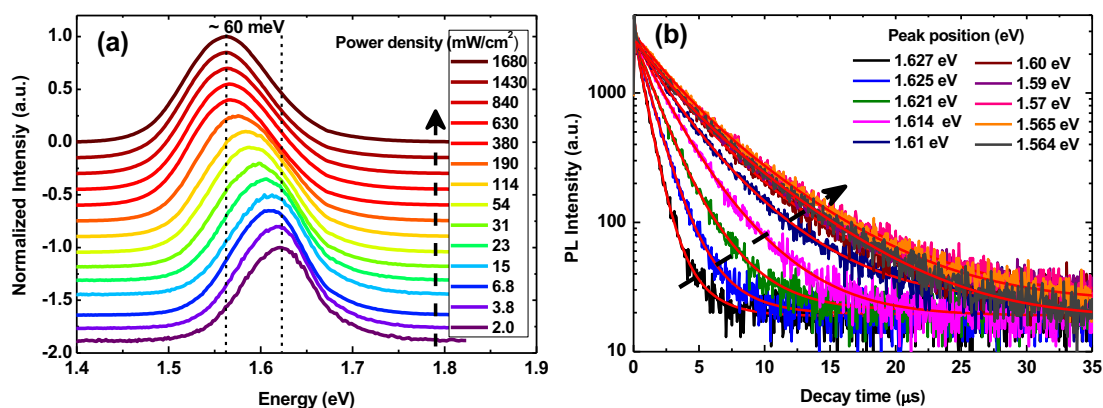


Figure: (a) Laser power-dependent photoluminescence (PL) spectra of triple-cation mixed-halide ($\text{MA}_{0.83}\text{FA}_{0.17}\text{Cs}_{0.05}\text{Pb}(\text{I}_{0.83}\text{Br}_{0.17})_3$) perovskite film showing red-shift in PL at room temperature and (b) PL-decay spectra measured on red-shifted PL positions.

References: [1] Fu *et al.* Nat. Rev. Mater. 4, 169–188 (2019); [2] Hoke *et al.* Chem. Sci. 6, 613–617 (2015); [3] Draguta *et al.* Nat. Commun. 8, 200 (2017); [4] Subodh K. Gautam *et al.* Adv. Funct. Mater. (2020) 2002622.