Origin of photo-absorption and photo-emission in two-dimensional Ruddlesden-Popper perovskites

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Two-dimensional Ruddlesden-Popper layered perovskites (RPLPs) have recently emerged as an alternative to the classic bulk organic-inorganic hybrid perovskites, mainly due to significantly improved photo- and chemical-stability in optoelectronic devices [1][2]. They also offer a richer chemical playground as compared to their 3D counterparts, which promises a wider range of functionalities for this layered material. Few recent encouraging developments in optoelectronic applications, notably in energy harvesting and light emitting [2][3], have already been demonstrated in these two-dimensional layered perovskites. However, further development and optimization of devices will require a deeper understanding of the intrinsic photo-physics and transport properties of the phase-pure RPLP materials. Here we investigate the physical properties of photo-generated charge carriers in phase-pure $(BA)_2(MA)_{n-1}Pb_nI_{3n+1}$ layered perovskite family [4], for which the *n*-number can be experimentally tuned thus the thickness of the perovskite layer. It is found that the photo-absorption and photo-emission properties of thin films with n>2 are dominated by bound excited states associated with edge-states of perovskite layers, which rule the characteristics of thin-film solar cells and light-emitting diodes. On the other hand, optical and electronic properties in RPLP crystals derive from both quantum and dielectric confinements of carriers in the two-dimensional perovskite layers, which are promising for future applications of color-tunable photon sources. Opportunities to control and switch between these states is also relevant for novel technology.

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