Phase-Transition-Induced Carrier Mass Enhancement in 2D Ruddlesden−Popper Perovskites

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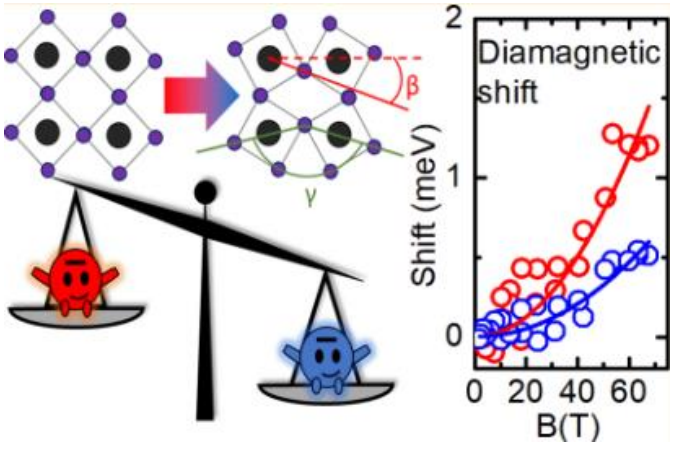
Hybrid organometal-halide perovskites have recently conquered the field of low-temperature processed optoelectronic materials, excelling their counterparts in performance [1]. A lot of effort in perovskite research is directed towards improving the stability of these materials, where 2D perovskites have been demonstrated as a more robust alternative to bulk materials [2]. There is a variety of possible ways to tune the optical properties of 2D perovskites, though the mutual dependence between tuning parameters hinders our fundamental understanding of their properties. In this work, we attempt to address this issue for Ruddlesden−Popper $(C_nH_{2n+1}NH_3)_2PbI_4$ perovskites with varying length of organic cations (n = 4, 6, 8, 10, 12) using optical spectroscopy in high magnetic fields up to 67 T.

Our experimental results, clearly demonstrate that the exciton reduced mass increases by around 30% in the low-temperature phase, which is reflected by a 2−3-fold decrease of the diamagnetic coefficient. This observation is supported by our DFT calculations, which find the dependence of the reduced mass value on octahedral tilting, increasing during the transition to the structure of lower symmetry. Our studies show that the effective mass, that determines carrier mobility in semiconductors therefore is an essential parameter for optoelectronic device operation, can be tuned by the variation of organic spacers and/or moderate cooling achievable using Peltier coolers. Moreover, we show that the complex absorption features visible in absorption/transmission spectra track each other in a magnetic field, providing strong evidence for the phonon-related nature of the observed side bands.



[1] S. D. Stranks, H. J. Snaith, Nat. Nanotech. **10**, 391 (2015).
[2] Y. Cheng *et al.*, Adv. Mat. **30**, 1703487 (2018).