Magneto-spectroscopy of excitons in two-dimensional layered perovskites

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Over the past few years we have witnessed a booming interest in the field of two-dimensional (2D) layered metal halide perovskite. The revival of this class of materials is driven in part by their increased environmental stability with respect to the 3D perovskites as well as their outstanding performances in photovoltaic and light-emitting devices [1]. The reduction of dimensionality allows material engineering to go beyond the mere control of the thickness of the inorganic quantum wells. In such a hybrid organic-inorganic materials, the plethora of available organic molecules not only allows to tailor various material functionalities, but also gives rise to a considerable structural diversity. Due to the high contrast of dielectric constant of organic/inorganic sublattices, the 2D perovskites are characterized by very large exciton binding energies with low effective Bohr radius. As a result of large binding energies, the exchange interaction between the electron and hole spins is greatly enhanced, which in turn increases considerably the exciton fine structure. On top of that, the ionic nature and the softness of the perovskite lattice results in a significant coupling of electronic excitations to the lattice vibration (*i.e.*, electron-phonon interactions). Such coupling has a pronounced impact on the optoelectronic properties of 2D perovskites and is manifested in a complex absorption and emission spectra [2], which usually consist of multiple, equally spaced (in energy scale) resonances [3]. As the result, the mutual understanding of electronic and lattice excitations and their correlation – polarons – is crucial to adequately describe this material system.

In this work, we highlight our recent results on the determination of excitonic and polaronic properties of various 2D layered perovskites. In order to access such parameters, spectroscopy in high magnetic field, up to 160 T was employed. By these experiments, we not only determine important material parameters such as effective mass, exciton binding energy and exciton Bohr radius [4], but we also address the longstanding open question about the exciton fine structure in 2D perovskites. We elucidate the order of excitonic states and determine the bright–dark splitting energy [5]. Finally, by correlating the magneto-spectroscopy data with resonant Raman scattering we explain the complex multi-peak absorption response of 2D layered perovskites as being dominated by exciton-polarons.

[1] Gong *et al.*, *Nat. Mater.* **17**, 550 (2018).
[2] Urban *et al.*, *J. Phys. Chem. Lett.* **11**, 5830 (2020).
[3] Dyksik *et al.*, *ACS Energy Lett.* **5**, 3609 (2020).
[4] Dyksik *et al.*, *J. Phys. Chem. Lett.* **12**, 1638 (2021).
[5] Dyksik *et al.*, *Sci. Adv.* **7**, 1 (2021).