

Excitons and Phonons in 2D perovskites

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High environmental stability and surprisingly high efficiency of solar cells based on 2D perovskites have renewed interest in these materials. These natural quantum wells consist of planes of metal-halide octahedra, separated by organic spacers. Remarkably the organic spacers play crucial role in optoelectronic properties of these compounds. The characteristic for ionic crystal coupling of excitonic species to lattice vibration became particularly important in case of soft perovskite lattice. The nontrivial mutual dependencies between lattice dynamics, organic spacers and electronic excitation manifest in a complex absorption and emission spectrum which detailed origin is subject of ongoing controversy. First, I will discuss electronic properties of 2D perovskites with different thicknesses of the octahedral layers and two types of organic spacer. I will demonstrate that the energy spacing of excitonic features depends on organic spacer but very weakly depends on octahedral layer thickness. This indicates the vibronic progression scenario which is confirmed by high magnetic fields studies up to 67T. Next, I will show that in 2D perovskites, the distortion imposed by the organic spacers governs the effective mass of the carriers. As a result, and unlike in any other semiconductor, the effective mass in 2D perovskites can be easily tailored. In the end, I will discuss exciton fine structure. The bright-dark splitting is also of paramount importance for light emitters which rely on the radiative recombination of excitons, since the excitons usually relax to the lowest lying dark state, which is detrimental for the device efficiency. I will discuss our optical spectroscopy measurements with an applied in-plane magnetic field to mix the bright and dark excitonic states of (PEA)₂PbI₄, providing the first direct measurement of the bright-dark splitting. The induced brightening of the dark state allows us to directly observe an enhancement of the absorption at the low-energy side of the spectrum related to the dark state. The evolution of the PL signal in the magnetic field, suggests that at low temperatures the exciton population is not fully thermalized due to the existence of a phonon bottleneck, which occurs due to the specific nature of the exciton-phonon coupling in soft perovskite materials.