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Probing Exciton Properties and Dynamics in Lead Halide Perovskite Semiconductors by Optical Spectroscopy

The rapid development of perovskite-based devices have stimulated intensive research to unravel the behaviours of photo-excited electrons, holes and their interactions. However, the fundamental understanding of electronic band-structures and interparticle coupling is still at its early stage. Here, we investigate the exciton structures and the effects of quantum-confinement on optical responses of perovskite materials. Our optical study reveals that perovskites have complicated band-structures with multiple states near the bandedges, which are separated by only tens of meV. Under strong quantum-confinement regime, the spin-degeneracy of the exciton bands is lifted due to the enhanced electron-hole interaction that consequently produces exciton fine-structure. Particularly, in perovskite quantum-wells, we resolve the longitudinal and transverse excitons split by electron-hole exchange interaction, which, however, does not affect the spin-polarization of the indirect excitons. The interplay of different spin-interacting processes gives rise to the coexistence of anisotropic and isotropic excitons in two-dimensional perovskites. A unified understanding of fundamental properties on this emergent class of semiconductors will pave the way towards exciton manipulation for novel device applications.

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Supervisor: Prof Xiong Qihua