# Strongly bound excitons in Ruddlesden-Popper 2D perovskites

Blancon, J.C., Mohite, A.D. (Los Alamos National Lab); Stier, A.V., Crooker, S.A. (NHMFL-Los Alamos)

## Introduction

Since the discovery of phase-pure higher members of the Ruddlesden-Popper 2D perovskites [1] (2D perovskite layer of thickness of more than one unit cell, or n value >1), there has been an intense debate regarding the nature of their optical resonances (excitons versus free carriers) and the value of exciton binding energies. This originates from the lack of knowledge of the fundamental quantities defining electron-hole interactions such as the structure of phase-pure Ruddlesden-Popper perovskites, the exciton reduced mass, effective dielectric constant, spatial extend of electron and hole wavefunctions. As a result of these uncertainties, it has been impossible to develop a universal scaling behaviour for the exciton binding energy in 2D perovskites.

## Experimental

Using a variety of zero magnetic field optical spectroscopy techniques (PL, PLE) and 60-Tesla short pulse magneto-absorption spectroscopy supported by modelling, we determine the effective mass and exciton binding energy in 2D perovskites with increasing thickness from n=1 to 5 (Fig. 1a). The reduced exciton mass was derived for the sequence of samples through rigorous theoretical simulation that utilizes the experimentally measured diamagnetic shift as a constraint for the exciton size (Fig. 1b,c).

## Results and Discussion

High magnetic field spectroscopic measurements form the basis to address the long-standing challenges regarding the exciton properties in 2D perovskites by answering two of the central scientific questions. We unequivocally elucidate that the dominant optical resonances in the layered 2D perovskites arise from strongly bound excitons (>125 meV, Fig. 1d) due to ultra-strong dielectric confinement and present a universal scaling law for the exciton binding energy as a function of 2D perovskite layer thickness (or n value). Key results include [2]: (i) the first direct and accurate determination of the exciton reduced mass in 2D perovskites using magneto-optical spectroscopy (up to 60 T), (ii) the direct measurement of the exciton binding energy, which ranges from 470 meV to 125 meV as the perovskite layer thickness increases from 0.64 nm (n=1) and 3.14 nm (n=5), and (iii) the development of a universal theoretical model, which accurately describes the Wannier-Mott exciton states in 2D perovskites and provided insight into the confinement mechanisms (dielectric vs. quantum confinement).

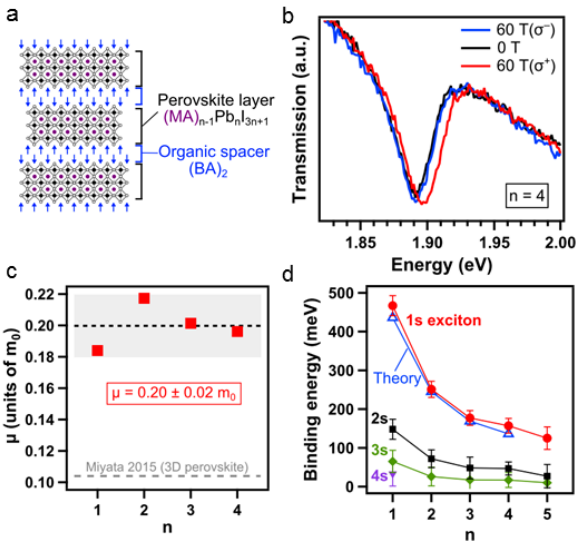


**Fig.1** (a) Ruddlesden-Popper perovskite structure. (b) Example of high-field magneto-absorption results. (c) Effective mass derived from the magneto-absorption measurements. (d) Corresponding *ns*-exciton binding energy for different perovskite layer thickness.

## Conclusions

(i) We emphasize that these results are extremely important for the design of any optoelectronic or photonic devices based on excitonic material system (quantum-wells or low-dimensional materials) [2,3], where typically, the light-generated electron-hole pairs, form strongly bound excitonic states dominating the photo-physics of materials (absorption and photoemission). To the best of our knowledge, this is the first direct evidence of strongly bound excitons in 2D perovskites with thickness up to 12 nm.

(ii) More generally, this study has been a unique opportunity to explore the physical properties of natural quantum-well semiconducting crystals situated between true monolayer 2D materials and bulk 3D materials. We are able to access quasi-dimensional physics in synthetic inorganic semiconductor quantum-well structures.

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## References

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[2] Blancon, J.-C., *et al.*, arXiv:1710.07653 (2017).
[3] Blancon, J.-C., *et al*., Science **355,** 1288–1292 (2017).