Scaling Law for Excitons in 2D Perovskite Quantum Wells

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Introduction

Ruddlesden–Popper halide perovskites (RPPs) are 2D solution-processed quantum wells where optoelectronic properties can be tuned by varying the perovskite layer thickness, and have recently emerged as efficient semiconductors with technologically relevant stability. However, fundamental questions concerning the nature of optical resonances (excitons or free carriers) and the exciton reduced mass, and their scaling with quantum well thickness, which are critical for designing efficient optoelectronic devices, remain unresolved. Here, using optical spectroscopy and 60-Tesla magneto-absorption supported by modeling, we unambiguously demonstrate that the optical resonances arise from tightly bound excitons with both exciton reduced masses and binding energies decreasing, respectively, from $0.221m_0$ to $0.186m_0$ and from 470 meV to 125 meV with increasing thickness from n = 1 to 5. Based on this study we propose a general scaling law to determine the binding energy of excitons in perovskite quantum wells of any layer thickness.

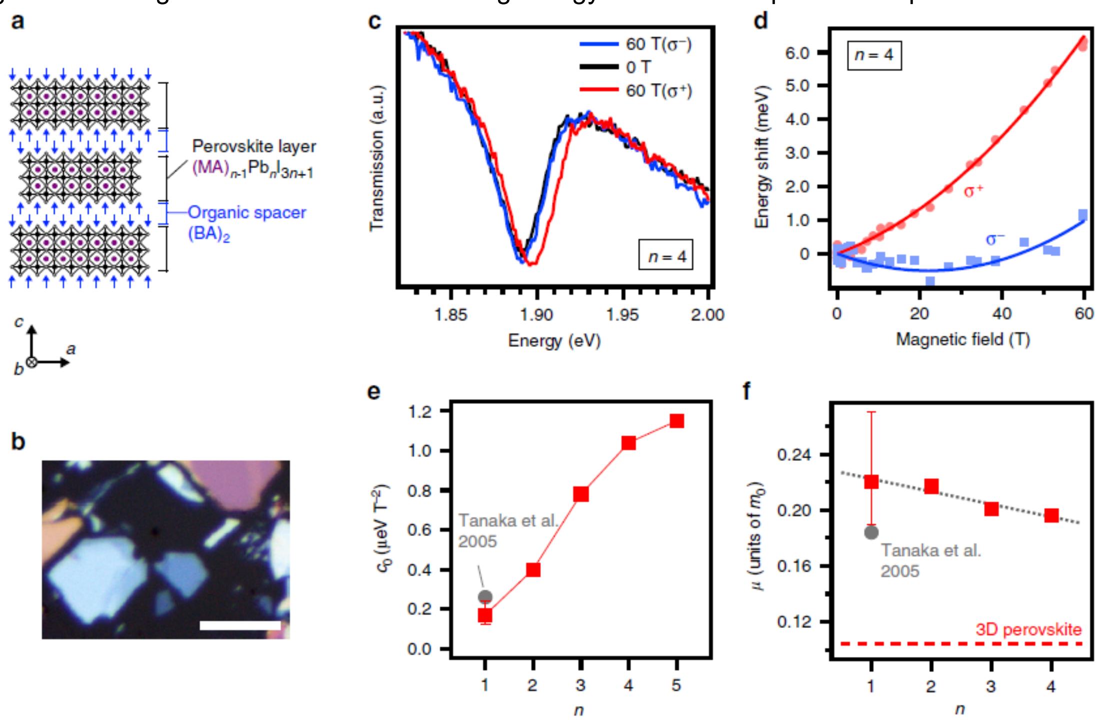

Figure: Exciton reduced mass from magneto-absorption spectroscopy. **a** Schematic of the RPP structure. **b** Image of mechanically exfoliated crystals. Scale bar is 10 μm. **c** Magnetic field dependence of the light transmission of an individual RPP with n = 4 crystal for right- (σ^-) and left-handed (σ^+) circular polarization. **d** Corresponding shift of the exciton energy as a function of the magnetic field. **e** Derived diamagnetic shift coefficient of the measured RPPs (red squares). **f** Exciton reduced mass derived from fitting the diamagnetic shifts. The red dashed line indicates average value of exciton reduced mass for the 3D perovskite $MAPbI_3$.

Results and Discussion

Here we present the study on using low-temperature (4 K) magneto-optical spectroscopy to accurately determine the exciton reduced mass for RPP crystals with perovskite layer thickness varying between 0.641 and 3.139 nm, corresponding to *n* between 1 and 5. The reduced mass is used to develop a generalized theoretical model for electron–hole interactions in RPPs and determine fundamental characteristics of the exciton states. In parallel, from low-temperature optical spectroscopy we experimentally determine the exciton binding energy in the RPPs with n from 1 to 5. From these results we produce a general scaling behavior for the binding energy of Wannier–Mott exciton states in RPPs, which allows for prediction of the exciton binding energy for any given thickness. This study closes a long-standing scientific gap and will lead to the rational design of next generation layered 2D perovskite-based optoelectronic devices.

References

[1] Blancon, J.-C. Nature Communications **9**, 2254 (2018).