



Light induced binding energy modification of excitons in hybrid perovskites

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Introduction

Organic-inorganic (hybrid) perovskites feature unique structural characteristics that are related to the hybrid nature of their crystal lattice. This includes a soft and dynamical lattice structure, which can be altered by external stimuli such as light and fields, as well as organic-inorganic atomically thin interfaces. Recently it was demonstrated that the crystal quality of lead halide perovskites (e.g. MAPbI₃ and (FA)_{0.7}MA_{0.25}Cs_{0.05}PbI₃) as well as the solar cell performances based on these materials can be improved from continuous sunlight illumination (soaking) for tens of minutes which initiate a lattice expansion of about 1% in the perovskite lattice [1]. Here we investigate the impact of light irradiation and lattice expansion on the exciton properties of the related material (FA)_{0.7}MA_{0.25}Cs_{0.05}PbI₅. We also investigate the impact of the organic-inorganic size on the exciton properties [2] of phase pure members of the Ruddlesden-Popper 2D perovskites by changing the organic molecule length.

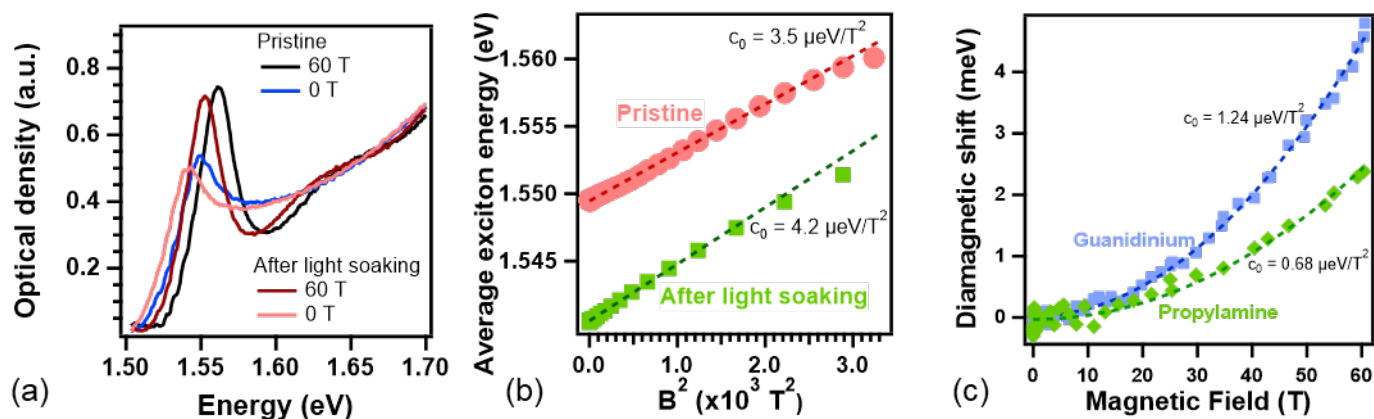
Experimental Results and Discussion

Using a technique that was developed for the spectroscopy of monolayer semiconductors [3], we affixed thin crystals of (FA)_{0.7}MA_{0.25}Cs_{0.05}PbI₅ on the tip of a single mode optical fiber and performed polarized low-temperature magneto-absorption spectroscopy to 60 tesla (Fig. 1a & b).

We find that the diamagnetic shift (c_0) of the fundamental exciton reversibly *increases* (from $\sim 3.5 \mu\text{eV}/\text{T}^2$ to $\sim 4.2 \mu\text{eV}/\text{T}^2$) after the sample is bathed in white light at room temperature. This appears concurrently with a red shift of the exciton feature from 800 nm to 804 nm at $T=4\text{K}$. We note that this effect only happens at room temperature and does not occur when the sample is held at cryogenic temperatures. We specifically note that this effect is reversible again after another thermal cycle without illumination of the sample without noticeable degradation of the optical spectrum. We further note that – unlike 2D Ruddlesden Popper perovskites – that we observe essentially no spin splitting of the fundamental exciton feature. We also note that we can control the exciton properties (diamagnetic shift change indicating of a change of the exciton binding energy and/or exciton reduced mass) by changing the organic cation in Ruddlesden-Popper perovskites (Fig. 1c).

Conclusions

These results pave the way to controlling exciton states in hybrid perovskite semiconductors either via chemical synthesis or by external stimulation.



Acknowledgements

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References

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