Fritz-Haber-Institut der Max-Planck-Gesellschaft

Physikalische Chemie — Direktor: Prof. Dr. Martin Wolf

MAX-PLANCK-GESELLSCHAFT

Department Seminar:

Wednesday, July 10, 2019, at **11:00** a.m.;
— all are invited to meet at around 10:40 for a chat with coffee & cookies —

Dr. Sebastian F. Maehrlein Department of Chemistry,
Columbia University, New York City.

Charge Carrier-induced Lattice Anharmonicities in Lead Halide Perovskites

PC Seminar Room G2.06, Building G, Faradayweg 4 M. Wolf

Abstract:

Lead halide perovskites (LHPs) have emerged as new darlings of materials science with broad potential for high performance solar cells and light emitting devices. The dynamically disordered and soft perovskite lattice is found to be a key component for their extraordinary optoelectronic properties, leading to a multitude of exciting phenomena such as (dynamic) Rashba effect, ferroelectricity, and large polarons[1–4]. The unifying goal of these studies is finding the fundamental mechanism of charge carrier protection in LHPs, but so far, specific electronic state coupled lattice potentials have eluded experimental probes.

Here, we perform coherent phonon spectroscopy (CPS) of the all-inorganic $CsPbBr_3$ perovskite and extend it to electronic state specific two-dimensional CPS. This novel method allows to establish the phonon response with precise excitation energy resolution in the highly dilute charge injection regime. We find a rich structure of coherent lattice response over a wide spectral range including strong phonon anharmonicities leading to coherent softening and phonon-phonon coupling. Additionally, we witness an extreme pump energy-dependent shift in phonon frequency compared to the equilibrium normal mode analysis through ab-initio calculations. These results unveil the energy-dependent mapping of strongly modified non-equilibrium lattice potentials, likely induced by ferroelectric charge localization. Such lattice response may form the basis for the design principle of defect tolerant semiconductors from efficient dynamic screening..

1. Miyata, K. et al. Large polarons in lead halide perovskites. *Sci Adv* **3**, e1701217 (2017).
2. Miyata, K. & Zhu, X.-Y. Ferroelectric large polarons. *Nature Materials* **17**, 379–381 (2018).
3. Evans, T. J. S., Miyata, K.; Joshi, P. P., Maehrlein, S.F. Liu, F. Zhu, X.-Y. Competition Between Hot-Electron Cooling and Large Polaron Screening in CsPbBr3 Perovskite Single Crystals. *Journal of Physical Chemistry C* (2018). doi:10.1021/acs.jpcc.8b00476.
4. Joshi, P. P., Maehrlein, S. F. & Zhu, X. Dynamic Screening and Slow Cooling of Hot Carriers in Lead Halide Perovskites. *Adv. Mater.*, 1803054 (2019).