Nonlinear THz Control of the Highly Polarizable Lead Halide Perovskite Lattice

Maximilian Frenzel[1], Marie Cherasse[1,2], Feifan Wang[3], Bo Xiang[3], Joanna Urban[1], Leona Nest[1], Lucas Huber[3], Martin Wolf[1], X.-Y. Zhu[3], Sebastian F. Maehrlein[1]

[1]*Fritz Haber Institute of the Max Planck Society, Berlin, Germany*
[2]*LSI, CEA/DRF/IRAMIS, CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, Palaiseau, France*
[3]*Columbia University, New York City, USA*

Lead halide perovskites (LHPs) have recently emerged as promising semiconductor materials for optoelectronic devices such as solar cells or LEDs. They show a range of outstanding optoelectronic properties, including long charge carrier diffusion lengths and apparent defect tolerance, the microscopic origins of which remain under debate [1]. So far, to tailor material properties, the community focused mainly on changing the static design of perovskite compounds, by tuning their chemical composition or morphology. Meanwhile, the full potential for dynamic phonon-driven ultrafast material control, as was demonstrated for oxide perovskites, has not been exploited yet.

We employ intense, close to single-cycle, THz fields to excite coherent optical phonons in the LHP inorganic sublattice and probe the ensuing THz-induced Kerr effect. We observe strong transient birefringence in both inorganic CsPbBr3 and in hybrid MAPbBr3. In agreement with previous optical Kerr effect studies [2], our rigorous four-wave mixing simulations [3] prove that it is crucial to account for dispersion and optical anisotropy for the correct interpretation of the signals. At room temperature, the response is dominated by the instantaneous electronic polarizability. At lower temperatures, we observe a contribution from the lattice polarizability governed by a coherent phonon at ~1 THz, corresponding to octahedral twists of the lead halide cage. This mode is a suitable candidate for explaining dynamic screening, suggested as a charge carrier protection mechanism in LHPs [4]. Our findings demonstrate the potential to obtain direct coherent phonon control over the LHP lattice properties via nonlinear excitation of highly polarizable modes.

References

[1] P.P. Joshi, SF Maehrlein, XY Zhu, Adv. Mater. **31**, 1803054 (2019)

[2] S.F. Maehrlein et al. PNAS, **118**,7, e2022268118 (2021).

[3] L Huber, SF Maehrlein, et al., J. Chem. Phys., **154**, 094202 (2021)

[4] F. Wang, et al., J. Am. Chem. Soc. **143**, 5–16 (2021).