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Phonon-Driven Charge Dynamics in Solids – New Insight from Ultrafast Terahertz and X-Ray Experiments

The interplay of charge and lattice excitations has a decisive influence on the electronic and optical properties of polar and ionic crystals. Specific low-frequency phonons, the so-called soft modes, are strongly coupled to the electronic system and induce strong relocations of electronic charge in space and time. New methods of nonlinear terahertz spectroscopy and femtosecond x-ray diffraction allow for mapping such dynamics in a temporally and spatially resolved way. In this talk, recent results on phonon-driven charge dynamics in prototype molecular materials are presented. In polycrystalline aspirin (acetylsalicylic acid), rotations of methyl groups with a sub-picometer displacement induce charge relocations on the 100 pm length scale of chemical bonds [1,2]. In ferroelectric ammonium sulfate, the macroscopic electric polarization is periodically reversed by excitation of a soft-mode with a period of a few picoseconds [3]. A theoretical analysis of time dependent charge densities establishes a direct link between microscopic electron distributions and macroscopic electric properties [4].

[1] G. Folpini et al., Phys. Rev. Lett. 119, 097404 (2017)

[2] C. Hauf et al., Struct. Dyn. 6, 014503 (2019)

[3] C. Hauf et al., Struct. Dyn. 5**,** 024501 (2018)

[4] C. Hauf, M. Woerner, C. Hauf, T. Elsaesser, Phys. Rev. B 98, 054306 (2018)