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Phase-resolved two-dimensional terahertz spectroscopy - a probe of highly nonlinear light-matter interactions

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Terahertz (THz) spectroscopy gives insight into low-frequency excitations and charge dynamics in condensed matter. So far, most experiments in a frequency range from 0.5 to 30 THz have focused on the linear THz response to determine linear absorption and disperion spectra, and/or electric conductivities. The generation of ultrashort THz transients with peak electric fields up to megavolts/cm has allowed for addressing nonlinear light-matter interactions and inducing excitations far from equilibrium. The novel method of two-dimensional THz (2D-THz) spectroscopy allows for mapping ultrafast dynamics and couplings of elementary excitations up to arbitrary nonlinear order in the electric field, both under resonant and nonresonant excitation conditions. In particular, different contributions to the overall nonlinear response are separated by dissecting it as a function of excitation and detection frequencies and for different waiting times after excitation. This talk gives an introduction in 2D-THz spectroscopy, including its recent extension to 3-pulse sequences and interaction schemes. To illustrate the potential of the method, recent results on two-phonon coherences and high-order interband excitations in the semiconductor InSb will be presented. Nonlinear THz excitation of two-phonon coherences exploits a resonance enhancement by the large electronic interband dipole of InSb and is, thus, far more efficient than linear excitation via resonant two-phonon absorption. As a second application, the nonlinear softmode response in a crystal consisting of aspirin molecules will be discussed. At moderate THz driving fields, the pronounced correlation of rotational modes of CH_3 groups with collective oscillations of π-electrons drives the system into the regime of nonperturbative light-matter interaction. Nonlinear absorption around 1.1 THz leads to a blue-shifted coherent emission at 1.5 THz, revealing a dynamic breakup of the strong electron-phonon correlations.