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Transient electronic structure and coherent phonon dynamics during photoinduced insulator to metal transitions

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The electronic properties of complex materials are often governed by strong electron-phonon coupling and many-body correlation effects leading to phenomena like metal insulator transitions or superconductivity and the formation of broken symmetry ground states. This interplay between electronic and phonon degrees of freedom is of particular importance in thermally or optically driven transitions in charge-density wave (CDW) materials.

Time- and angle-resolved photoemission spectroscopy (trARPES) probes the evolution of single particle spectral function after optical excitation and makes the collective dynamics of a system (e.g. coherent phonons) directly visible through their influence on the quasiparticle band structure. Using this technique, we present a systematic study of TbTe3, a metal which exhibits a Fermi surface nesting driven CDW transition [1]. Time-resolved data taken at different positions in the Brillouin Zone and at different temperatures enable us to observe collective modes at 3.5 THz and 2.5 THz and their highly anisotropic (k-dependent) coupling to the electronic system in real time. The 2.5 THz mode, which occurs only in the CDW band near the gapped region, is identified as the amplitude mode and governs the retarded (>100fs) collapse of the CDW gap in TbTe3. This is in clear contrast to the previously observed quasi-instantaneous melting (« 50fs) observed for the Mott insulator TaS2. In addition, a systematic study of the pump fluence dependence in the gapped region of TbTe3 documents the crossover from a regime where mainly the amplitude mode gets excited to a regime where the CDW gap closes at least partially.

Employing a novel position-sensitive Time-of-Flight spectrometer (pTOF) [2], we are able to investigate the dynamics of both occupied and unoccupied electronic states over a con-tiguous (2D) area of the reciprocal space. We can thereby follow the evolution of the Fermi surface and can precisely determine the position of the CDW gap even in the unoccupied band structure. For example, we can identify, for the CDW compound HoTe3 the gap associated with the second CDW transition [1b] and investigate its dynamics in real time.

Using the well known photoinduced semiconductor-metal transition in VO2 we address the dynamics of transient changes in crystal symmetry, which accompany most photoinduced phase transitions. We show that an ultrafast change in symmetry can be probed by the coherent (phonon) response of the lattice. Below the threshold fluence, the four lowest Ag phonon modes of the monoclinic phase modulate the transient reflectivity of VO2. As the pump intensity is increased, a photoinduced transition is induced by excited electrons resulting in an abrupt change in the phonon dynamics which correspond to an ultrafast change of the lattice potential.

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[2] P.S. Kirchmann, et al., Appl. Phys. A 91, 211 (2008)

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