Snapshots of cooperative atomic motions in the optical becalming of charge density waves.

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Femtosecond (fs) time-resolved techniques are becoming established as new tools for studying correlated electron phenomena in solids. By tracking energy relaxation pathways between various degrees of freedom (electron, spin, lattice etc.) on fs timescale, the interaction strengths amongst these subsystems can be investigated or manipulated. Charge-Density-Wave (CDW) systems, with their inherently multi-component order parameter, present a prime example of such cooperative phenomena in solids. The CDW state is characterized by the periodic modulation of the electron density and the periodic lattice distortion (PLD). In equilibrium the order parameter is given by a macroscopically occupied "frozen-in" phonon mode (static displacement of atoms away from that of the host lattice). Following perturbation with a fs optical pulse, however, the situation can be realized where on the fs timescale the electronic and lattice parts of the order parameter are uncoupled. While numerous fs time-resolved experiments have been performed on this class of materials, all were essentially probing the dynamics of the electronic subsystem and only inferred the dynamics of the PLD.

Here we report on the first study of the fs structural dynamics in a CDW system 1T-TaS2, where dynamics of the PLD was studied directly utilizing fs electron diffraction [1]. The results reveal an ultrafast non-thermal suppression of the CDW order with a timescale of 170 fs. The high degree of correlated motions is further highlighted by the rapid electron-phonon energy transfer (350 fs) and the exceptionally fast recovery of the CDW (4 ps). These observed timescales and the degree of cooperativity are remarkable and illustrate the importance of obtaining atomic level perspectives of the dynamics directing the physics of strongly correlated systems.

[1] M. Eichberger et al., submitted