Fundamental couplings in strongly correlated materials investigated by complementary time-domain techniques

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Correlated materials are characterized by the variety of couplings between the elementary degrees of freedom, leading to novel ground states with broken symmetries and often intriguing properties. Yet the quantitative determination of those couplings, and their relevance for the formation of broken symmetry ground states and phase transitions remains a major challenge. In particular, in thermal equilibrium the various interactions are present simultaneously in a system, making it difficult to separate them due to their similar energy scale. Studies of those interactions in the time domain in a non-equilibrium system created after ultrafast optical excitation promise a way to separate such contributions by their intrinsically different dynamics.

In particular using quantitative and complementary femtosecond time-resolved spectroscopies, which directly address the dynamics of specific degrees of freedom on their individual time scales, allows access to the couplings between those degrees of freedom and their relevance for a phase transition from their temporal evolution. Specifically, combining femtosecond time- and angle-resolved photoemission spectroscopy (trARPES) and time-resolved (resonant) x-ray diffraction (trXRD) techniques allows us to follow the ultrafast dynamics of electronic, structural and magnetic degrees of freedom and their orderings individually, yielding direct access to the coupling of the electronic, phononic and spin systems.

A showcase example is the Fe-pnictide parent compound $BaFe_2As_2$, where trXRD allows us to observe fluence dependent intensity oscillations of two specific Bragg reflections with a period of ~200 fs due to the coherent excitation of the A_{1g} phonon mode. Comparison of the quantitative coherent modifications of the Fe-As tetrahedra with time-resolved photoemission data yields the electron-phonon deformation potential for this particular mode. Our results demonstrate the importance of this structural degree of freedom for the electron-phonon coupling in the Fe pnictides and indicate a transient increase of the Fe magnetic moments on an ultrafast timescale.

Furthermore, I will discuss the case of the optically driven charge-density wave (CDW) transition in $TbTe_3$, where coherent and incoherent dynamics of the electronic and structural CDW order parameters investigated by trARPES and trXRD, respectively, demonstrate a strong coupling of the CDW condensate to particular phonon modes. In addition, the transient order parameter dynamics directly encodes the transient energy potential landscape.

Finally, the rare-earth intermetallics $RERh_2Si_2$ (RE=Gd, Sm) shows an intriguing interplay of bulk antiferromagnetic (AFM) order and a ferromagnetic (FM) surface state. Here, the dynamics of FM exchange splitting investigated by trARPES is complemented by femtosecond time-resolved resonant soft-xray diffraction at the RE M5-edges which directly measures the long-rage AFM order parameter.