

Ultrafast Dynamics of Strongly Correlated Materials

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We use femtosecond time- and angle-resolved photoemission to study photoinduced phase changes and electron-phonon coupling in highly correlated materials. The technique provides information about the interplay of the electronic band structure and the coupling of single particle states to collective excitations (e.g. coherent phonons), which are probed directly in the time domain.

Using trARPES we have studied the Mott insulator 1T-TaS₂ and demonstrate that the photoinduced insulator to metal transition is driven directly by electronic excitation as revealed by the instantaneous collapse of the electronic gap [1]. Photoexcitation by an intense laser pulse leads to an ultrafast (<50fs) insulator-to-metal transition towards a gapless phase. A coherently excited lattice mode results in a periodic shift of the spectra (CDW mode), which is lasting for 20 ps while the system has relaxed back to the insulating phase. These findings clearly demonstrate that the metal-insulator transition in TaS₂ follows a Mott-Hubbard scenario and does not support a Peierls-type mechanism. This is in clear contrast with the retarded (>100fs) response which we observe for the transient melting of the CDW phase in TbTe₃ [2]. Using trARPES we are able to identify the collective mode giving rise to the CDW transition and their highly anisotropic (k-dependent) coupling to the electronic system in real time.

Recently we have studied high-T_c superconductors (namely cuprates [3] and iron pnictides). The new class of FeAs based superconductors exhibits a complex interplay between electronic, lattice and magnetic degrees of freedom. In order to study this interplay, the electron dynamics following optical excitation of EuFe₂As₂, BaFe₂As₂ and BaCo_{0.15}Fe_{1.85}As₂ has been investigated by trARPES. We observe a momentum-dependent carrier dynamics (probed around the center of the Brillouin zone), whereby relaxation rates for holes are much (10x) faster compared to the electrons. This asymmetry in the dynamics of electrons and holes cannot be explained solely by scattering processes within one single band (i.e. the hole pocket at the Γ point) and is attributed to scattering processes between the center and boundary of the Brillouin zone (Γ and X points, respectively). In addition, pronounced periodic oscillations of the electronic structure in the vicinity of the Fermi level are observed. Analysis of these oscillations reveals three coherently excited modes with frequencies 5.6, 3.3 and 2.6 THz, respectively. Comparison with Raman scattering allows assigning the 5.6 THz mode to the A_{1g} mode, which changes the Fe-As distance and therefore modifies the Fe magnetic moment. The origin of the two other modes is less clear and might include magnetic excitations.

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