# Transient Electronic Structure of Solids and Surfaces studied with Time- and Angle-Resolved Photoemission

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**Abstract:** The dynamics of highly correlated materials are studied by femtosecond time- and angle-resolved photoemission spectroscopy. In the new FeAs based high-Tc superconductors electron-phonon coupling plays a decisive role leading to strongly momentum dependent carrier relaxation.

One of the key questions in solid state physics is the origin of the electric conductivity of a material. While simple band structure theory can explain basic electronic and optical properties of most insulators, semi-conductors and metals, other more complex materials exhibit properties based on many body interactions. The electronic properties of such highly correlated materials are often governed by strong electron-phonon coupling and correlation effects leading to phenomena like formation of charge density waves (CDW), metal insulator transitions or superconductivity. This interplay between electronic and phonon degrees of freedom is of particular importance for the understanding of highly correlated materials. As the dynamics of the underlying elementary processes (like the electron-electron correlations or electron-phonon interaction) occur typically on different and characteristic timescales, it can be expected that time-resolved spectroscopy can provide complementary and new information on these elementary processes and coupling between various degrees freedom.

Here we use femtosecond time- and angle-resolved photoemission (trARPES) to investigate the electron dynamics of various photoexcited correlated materials. In these experiments the samples are cleaved in Ultra-High-Vacuum (UHV) to obtain an atomically well defined surface and are excited by 1.5 eV femtosecond pump pulse at cryogenic temperatures. The temporal evolution of the electronic band structure after photoexcitation is probed by time-delayed 6 eV femtosecond pulses giving rise to a photoemission spectrum originating from both occupied and transiently populated (i.e. normally unoccupied) electronic states above the Fermi level. Angular resolution of the photoelectrons provides access to the electron momentum parallel to the surface. The overall time-resolution of the setup is limited to ~90 fs by the duration of the 6 eV laser pulse. The trARPES technique can simultaneously provide information about the single particle spectral function in the frequency domain and collective excitations (e.g. coherent phonons) in the time domain, allowing to directly relate these excitations with the electronic band structure.

Using trARPES we have studied the Mott insulator 1T-$TaS_2$ 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 has relaxed back the insulating phase. These findings clearly demonstrate that the metal-insulator transition in $TaS_2$ follows a Mott-Hubbard scenario and do 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 $TbTe_3$ [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 highTc 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 $EuFe_2As_2$, $BaFe_2As_2$ and $BaCo_{0.15}Fe_{1.85}As_2$ 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 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). We thus propose that scattering processes between the center and boundary of the Brillouin zone (Γ and X points, respectively) might be relevant, which would require momentum transfer and may lead to emission of zone boundary phonons.

In addition, pronounced periodic oscillations of the electronic structure in the vicinity of the Fermi level are observed (see Fig.1). Analysis of these oscillations reveals three coherently excited phonon modes with frequencies 5.6, 3.3 and 2.6 THz, respectively. Comparison with Raman scattering results allows assigning the mode at 5.6 THz to the $A_{1g}$ mode, which changes the Fe-As distance and therefore modifies the Fe magnetic moment. The two other modes may originate from the emission of zone boundary phonons as discussed above.

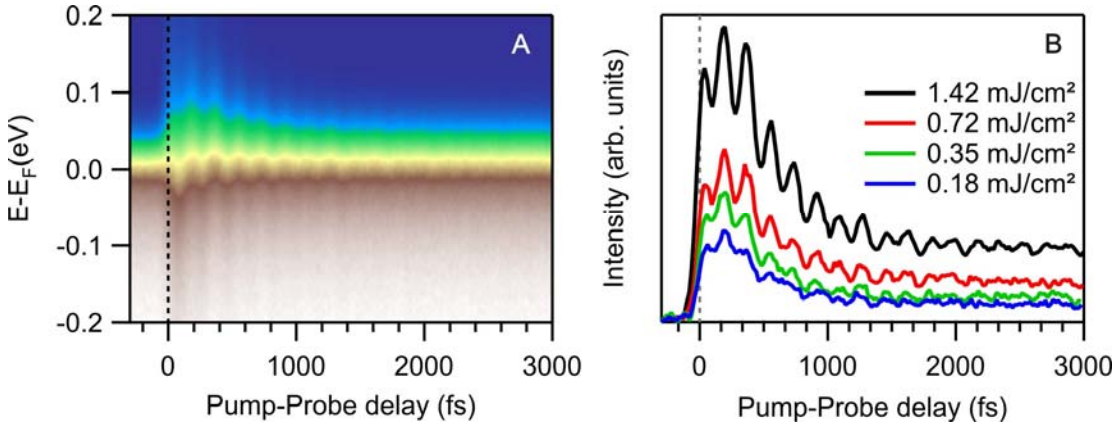


Fig.1 (A) Time evolution of the electronic structure of $BaCo_{0.15}Fe_{1.85}As_2$ recorded at $k_F$ of the hole pocket (at the Γ point). The spectral function shows pronounced periodic oscillation in the vicinity of the Fermi level. (B) Integrated photoemission intensity above the Fermi level as a function of pump probe delay for various pump fluencies, which exhibits beating between several modes.

Note that in trARPES all coherent phonon modes are observed via their coupling to the electronic structure. Therefore only those modes, which are coherently excited by a femtosecond laser pulse and which exhibit a sufficiently strong electron-phonon coupling, can be probed. This, however, provides a unique opportunity to study the dynamics of electron-phonon coupling with electron momentiúm resolution.

In summary, we have used femtosecond time- and angle-resolved photoemission to study photoincued phase changes and electron-phonon coupling in highly correlated materials. The technique provide 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.

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