Ultrafast multidimensional spectroscopy and diffraction of nanoscale materials

S. Dong, S. Beaulieu, M. Dendzik, T. Pincelli, P. Xian, W. Windsor, J. Maklar, D. Zahn, H. Seiler, T. Vasileiadis, Y. Qi, M. Puppin, C. Nicholson, Y. Deng, L. Waldecker, M. Wolf, L. Rettig, and R. Ernstorfer

Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin
ernstorfer@fhi-berlin.mpg.de

The dynamics of quasi-particles in non-equilibrium states of matter reveal the underlying microscopic coupling between electronic, spin and vibrational degrees of freedom. We aim for a quantum-state-resolved picture of coupling on the level of quasi-particle self-energies, which goes beyond established ensemble-average descriptions, and which requires ultrafast momentum-resolving techniques. The dynamics of electrons and excitons is measured with four-dimensional time- and angle-resolved photoelectron spectroscopy (trARPES), featuring a high-repetition rate XUV laser source [1] and momentum microscope detector, see Fig. 1. I will exemplify this experimental approach by discussing electron and exciton dynamics in the semiconducting transition metal dichalcogenide WSe_2 [2] and discuss its extension to nanoscale heterostructures. Upon strong excitation inducing phase transitions, trARPES reveals the full transient electronic structure driving the structural transition along the reaction coordinate [3]. The complementary view of ultrafast phonon dynamics is obtained through inelastic femtosecond electron diffraction [4].

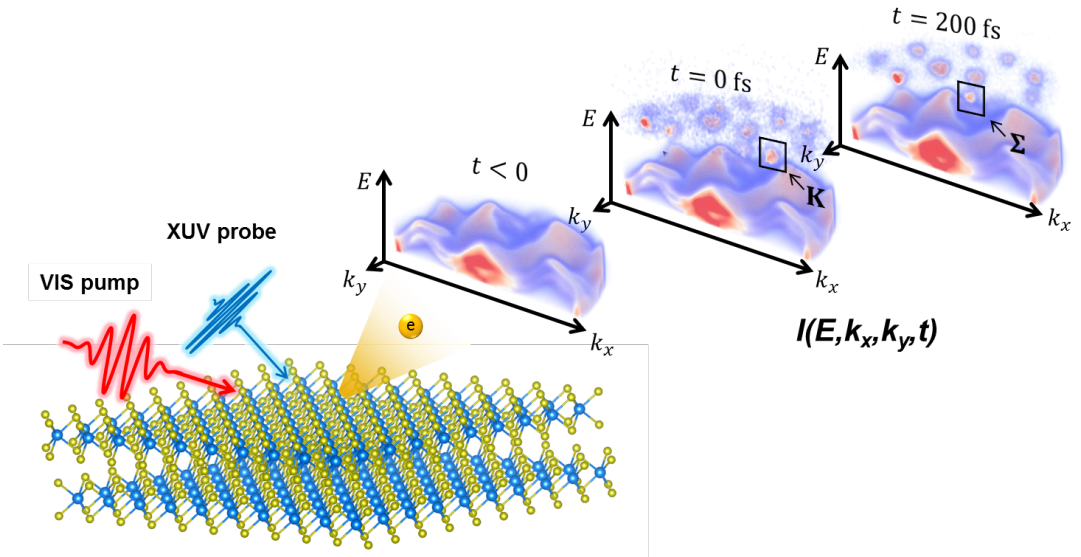

Fig. 1: Multidimensional photoemission spectroscopy. (a) Illustration of the trARPES scheme with a time-of-flight momentum microscope spectrometer, which resolves the photoelectrons in energy, both parallel momentum dimensions and (pump-probe) time. Three snapshots of a movie showing the generation of bright A excitons and their scattering to dark finite-momentum states is shown for WSe_2.

References:

[1] M. Puppin et al., Rev. Sci. Inst. 90, 23104 (2019).
[2] R. Bertoni et al., Phys Rev. Lett. 117, 277201 (2016).
[3] C.W. Nicholson et al., Science 362, 821 (2018); Phys. Rev. B 99, 155107 (2019).
[4] L. Waldecker et al., Phys. Rev. Lett. 119, 036803 (2017).