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Time-resolved photoelectron spectroscopy

Martin Weinelt, Fachbereich Physik, Freie Universität Berlin, Berlin, Germany

Abstract

In this lecture I will discuss aspects of time-, angle-, energy-, and spin-resolved photoemission spectroscopy. We use this technique to study the dynamics of electronic excitations at solid state surfaces and transients of the electronic band structure. Ultrashort optical and vacuum-ultraviolet (VUV) pulses from laser sources and/or a higher-order harmonic generation beamline serve as the sources for time-resolved experiments. Photoelectrons are detected after a hemispherical or a time-of-flight electron analyzer optimized for low kinetic energies.

Laser excitation allows us to populate unoccupied electronic states between the Fermi and the vacuum level and probe their dynamics. Comparing lifetime and linewidth or the decay of quantum oscillations gives access to inelastic and quasi-elastic electron scattering. Image-potential states are a well-defined model system of unoccupied surface states for which decay and dephasing processes can be studied in great detail. We will discuss a few examples illustrating scattering at surface defects and steps. For ferromagnetic substrates the image-potential states become exchange split which allows us to study spin-dependent electron dynamics. We will highlight magnon emission and electron-electron exchange scattering for the 3d-metals.

To investigate the occupied band structure of solids, laser pulses with photon energies exceeding the work function are necessary. 6-eV pulses can be produced generating the fourth harmonics of the Ti:sapphire fundamental in non-linear crystals. Therefore still moderate laser intensities are required allowing for high repetition rates and thus sufficient signal-to-noise ratios. In pump-probe experiments we can study the dynamics of occupied and unoccupied states close to the Fermi level and probe the electronic band structure in a small range of the Brillouin zone. For 6-eV photoemission 2d-systems are particular suitable due to a sizeable coupling of 2d initial states to photocurrent-carrying final states. We will give two examples studying the thermalization of hot electrons and the laser-induced melting of a charge-density wave.

To study particular regions of the bulk Brillouin zone VUV pulses with tunable photon energy are required. Here higher-order harmonic generation offers an emerging radiation source for mapping the transient band structure after laser excitation. We focus on the lanthanide Gadolinium and study the breakdown of the magnetization after femtosecond laser excitation. To compare 5d and 4f spin dynamics we record in parallel the transient band structure and the dichroism in 4f photoemission. We observe distinctly different spin dynamics, which suggest the breakdown of the intra-atomic exchange upon femtosecond laser excitation.