Excited-state mapping with XUV-based trARPES

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Time-resolved photoemission combined with scanning of the emission angle can extend established band-structure mapping to excited states which are only occupied out-of-equilibrium. The full potential of time- and angle-resolved photoemission spectroscopy (trARPES) is reached by performing the experiment at high repetition rates of hundreds of kHz, limiting space charge effects and data acquisition time. Extreme ultraviolet (XUV) photon energies grant access to the whole Brillouin zone: by performing high-harmonic generation at 0.5 MHz with a novel laser light source [1], we demonstrate trARPES with a photon energy of 21 eV, ≈100 meV resolution and sub-50 fs time resolution. A 3.1 eV pump pulse populates the conduction band of the layered transition metal dichalcogenide WSe_2 followed by relaxation toward the band minimum within 1 picosecond. About 100 fs after excitation, most of the normally unoccupied conduction band states are populated due to scattering of the photo-excited carriers, allowing excited state band mapping throughout the whole Brillouin zone. This demonstrates how trARPES can measure the bandgap of a material in a single experiment with full time and momentum resolution. The system was further excited in resonance with an excitonic-feature of the optical spectrum. In this case, the observed signal was localized at the K point CB minimum (direct bandgap), overlapping with Floquet-like replica bands within the bandgap. By temperature-tuning the material's bandgap the true population could be distinguished from the replica bands in the time domain. An extremely fast population decay (~ 20 fs) from the K point to the Σ sigma point (CB minimum) was observed in this case, showing that an efficient scattering mechanism transfer the excited state population to optically-dark states.

References:

[1] M. Puppin, Y. Deng, O. Prochnow, J. Ahrens, T. Binhammer, U. Morgner, M. Krenz, M. Wolf, and R. Ernstorfer, Opt. Express, **23**, 2, 1491 (2015).