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Unraveling exciton dynamics in 2D quantum materials on ultrashort time- and lengthscales using time-resolved momentum microscopy

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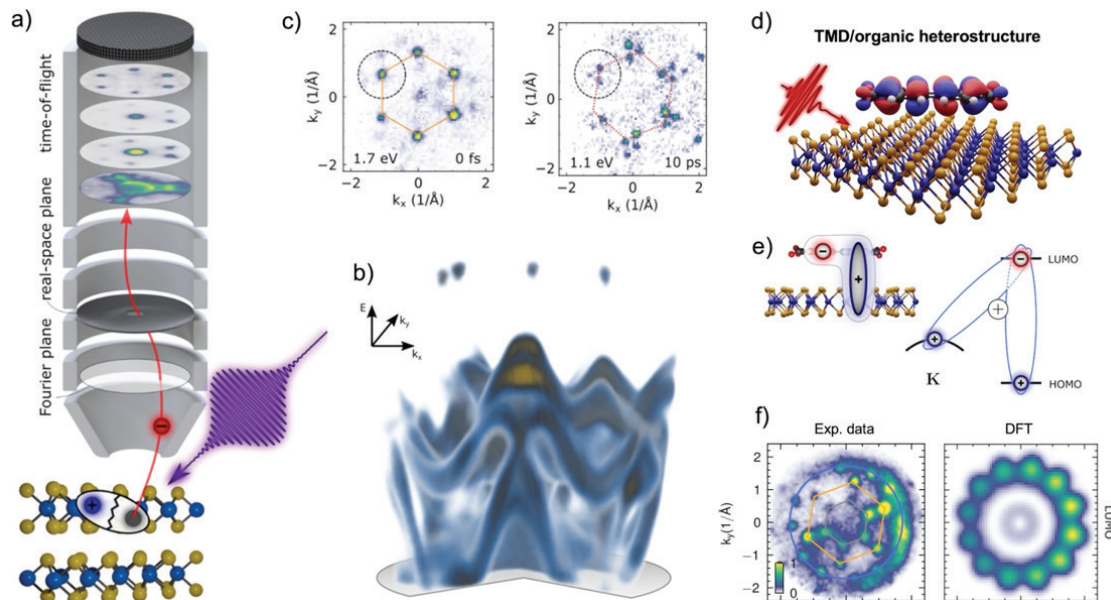


Fig. 1. a) Scheme of the ultrafast momentum microscopy experiment [1]. b) 3D electronic structure of WSe2/MoS2 with excitonic signals above the valence band maxima [3,4]. c) Selected momentum cuts showing the momentum fingerprint of the optically excited A-exciton in WSe2 (left) and an interlayer exciton in WSe2/MoS2 [3]. d) Transition metal dichalcogenide – organic semiconductor heterostructure [4]. e) Scheme of the hybrid exciton (left) and the involved electronic single-particle states (top of valence band of TMD and HOMO/LUMO of molecular layer). f) Exciton fingerprints from experiment (left) showing bright and dark excitons in the TMD and the signature of the molecular orbital being indicative for the hybrid exciton. Right: DFT calculation of LUMO state of PTCCA for comparison [6].

Two-dimensional semiconducting quantum materials and organic semiconductors have emerged as promising materials for next-generation optoelectronics and energy harvesting. At their heart lies the process of exciton formation: when light is absorbed, Coulomb-bound electron-hole pairs, termed excitons, carry and convert energy on nanometer spatial and femtosecond temporal scales. Understanding the very first moments of light-matter interaction, along with subsequent exciton relaxation and energy conversion, is vital for designing devices with new functionality and high efficiency.

To study light-matter interaction and exciton dynamics in such 2D quantum material heterostructures, we have developed a state-of-the-art, photoemission-based experiment [1] that enables us to observe exciton dynamics in space and time, spanning nanometers and femtoseconds. This approach uncovers the characteristic fingerprints of exciton generation, tracks their dynamics of energy transfer and thermalization, and lets us map the elusive dark exciton landscape in 2D semiconductors.

In my talk, I will show how we probe ultrafast light-matter interactions [2] and resolve the formation dynamics of dark interlayer excitons across a variety of 2D quantum materials [3-7]. I will discuss the identification of distinctive photoemission signatures linked to these processes, show how the dark exciton landscape can be mapped, and introduce “photoemission exciton tomography” [6], a new technique that enables tracing ultrafast charge transfer from transition metal dichalcogenides (TMDs) to organic layers in real time [7].