### Ultrafast & XUV Science

#### Spin-resolved electronic dynamics in bulk WSe$_2$

The ability to generate and control spin-polarized carriers is at the heart of spintronic science. One way of doing so is to use materials where several quantum degrees of freedom are strongly entangled. Natural candidates are transition metal dichalcogenides (TMDCs) where electrons, spins and valley indexes are coupled and can be simultaneously addressed via proper light excitation. However, the direct monitoring of the spin polarization of such short-lived states remains an experimental challenge.

Using time- and spin-resolved photoemission spectroscopy at the CLF Artemis facility, we were able to probe in the time domain, the spin-polarization of photo-excited carriers in bulk WSe$_2$. By using circular optical excitation, we observe the generation of an almost purely spin-polarized electron gas in the conduction band (K valley) of the material. It demonstrates that such excitation gives direct access to the spin and the valley degree of freedom in this class of material. Knowing the way to effectively generate such spin-polarized excited states, and their following evolution in the time domain, is mandatory in order to harvest these peculiar properties for spintronic devices.

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#### XUV Ptychographic imaging of mouse hippocampal neurons with 50nm resolution using the Artemis HHG source

The availability of coherent sources of XUV and soft x-ray radiation from sources based on high harmonic generation (HHG) means that imaging in these spectral regions can be performed using lens-less techniques such as ptychography. This allows for much increased resolution, as no imaging optics are required, and for measurement of the full complex transmission of the sample. In this paper, we describe coherent imaging of mouse hippocampal neurons grown on silicon nitride substrates at a wavelength of 29 nm with diffraction-limited resolution of ~100 nm. Transmission imaging was performed with an illumination-forming pinhole close to the sample, and also with an illumination-forming aperture at a large distance (~1 m) from the sample demagnified by EUV reflective optics. This allows much greater flexibility in designing measurement geometries without the need for a pinhole in close proximity to the sample.

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Observing the complete reaction pathway of CS₂ dissociation

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Much of our current understanding of chemical dynamics relies on carefully applied assumptions, most famously the Born-Oppenheimer approximation, which allows us to disentangle the motion of electrons and nuclei from each other. Photochemistry, however, occurs on timescales too fast for those assumptions to stand. As a result, even relatively small and structurally simple molecules, when treated with light, undergo a complex cascade of competing processes and pass through a large number of electronic states of near-identical energy. We have shown that it is possible to use a high harmonic XUV probe to monitor the evolution of a photodissociation reaction initiated by an ultrafast UV pulse, following all of the intermediates involved. This work is backed up by kinetic modelling showing agreement with the observed spectra.

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Out-of-equilibrium electronic dynamics of the transition metal dichalcogenides MoTe₂ and WTe₂

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Weyl semimetals are at the frontier of research activity on novel topological phases of matter. We have investigated the out-of-equilibrium electronic properties of two transition metal dichalcogenides, MoTe₂ and WTe₂. The former exhibits a topological phase transition as a function of temperature, while the topology of the latter is still under debate. Our data show that the electron dynamics are strongly sensitive to details of the material unoccupied band structure. In particular, the opening of bandgap in the topological trivial phase of MoTe₂ is found to act like a bottleneck. From the comparison with WTe₂, and the observation of a longer relaxation dynamics, we conclude that this material is a topological trivial semimetal.

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UV-pump–VUV-probe photoelectron spectroscopy experiments at ARTEMIS: unravelling the ultrafast relaxation dynamics of aniline

Using an XUV photon as a universal probe capable of detecting all of the possible product states, we tracked the dynamics of aniline following excitation into its $2\,\pi\pi^*$ excited state. The preliminary results highlighted here indicate that it is practical to perform this experiment at Artemis.

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Figure 1 (left): Integrated photoelectron signal as a function of pump-probe delay. Top (blue) curve is in the region of the UV-pump + UV-pump signal, and the bottom (black) curve is in the region of the UV-pump + XUV-probe signal.

Figure 2 (right): Difference plot of the photoelectron spectra recorded using a 250 nm pump and 21.7 eV probe as a function of time. Note the increase in signal around 17.5 eV. This is the region where pump-probe photoionisation is observed.

Dynamics of correlation-frozen antinodal quasiparticles in superconducting cuprates

Many puzzling properties of high–critical temperature ($T_c$) superconducting (HTSC) copper oxides have deep roots in the nature of the antinodal quasiparticles, the elementary excitations with wave vector parallel to the Cu–O bonds. These electronic states are most affected by the onset of antiferromagnetic correlations and charge instabilities, and they host the maximum of the anisotropic superconducting gap and pseudogap. We use time-resolved extreme-ultraviolet photoemission with appropriate photon energy (18 eV) and time resolution (50 fs) to reveal the ultrafast dynamics of the antinodal states in a prototypical HTSC cuprate. After photoinducing a nonthermal charge redistribution within the Cu and O orbitals, we reveal a dramatic momentum-space differentiation of the transient electron dynamics. Whereas the nodal quasiparticle distribution is heated up as in a conventional metal, new quasiparticle states transiently emerge at the antinodes, similarly to what is expected for a photoexcited Mott insulator, where the frozen charges can be released by an impulsive excitation. This transient antinodal metallicity is mapped into the dynamics of the O–2p$_x$ peak, thus directly demonstrating the intertwining between the low- and high-energy scales that is typical of correlated materials. Our results suggest that the correlation-driven freezing of the electrons moving along the Cu–O bonds, analogous to the Mott localization mechanism, constitutes the starting point for any model of high-$T_c$ superconductivity and other exotic phases of HTSC cuprates.

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Spin and valley control of free carriers in single-layer WS₂

Time- and angle-resolved photoemission spectroscopy (TR-ARPES) is used to measure optically excited free carriers in the electronic band structure of single-layer (SL) tungsten disulphide (WS₂) grown on the (111) face of silver (Ag). This SL transition metal dichalcogenide (TMDC) is characterized by a strong spin splitting on the order of 420 meV at the valence band maximum. Such a strong spin-orbit coupling is desirable for a TR-ARPES experiment that aims to directly detect free carriers selectively excited in the spin-split states in a given valley. Our experiments reveal excited electron and hole populations that are at their maximum for a resonant excitation between the upper valence band spin state and the conduction band. In addition, a noticeable valley polarization of the free carriers results when the material is pumped with circularly polarized light.

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Carrier dynamics in a two-dimensional metal

We provide the first direct femtosecond study of single-layer (SL) tantalum disulphide (TaS₂) — a two-dimensional (2D) metal — using time- and angle-resolved photoemission spectroscopy (TR-ARPES) at the Artemis facility. We simulate the measured spectral function procedure in order to extract the electronic dispersion and temperature. We find that a decay time of 170 fs for the excited electrons. 40 fs after optical excitation of the SL-TaS₂, the electrons reach an exceptionally high temperature of 3080 K. The elevated electronic temperature is accompanied by a surprising renormalization of the electronic structure bandwidth, driven by interactions of the hot electron gas. Upon exploring the excitation and its temporal evolution for different fluences and sample temperatures, we find an ultrafast single-exponential decay of hot electrons.

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