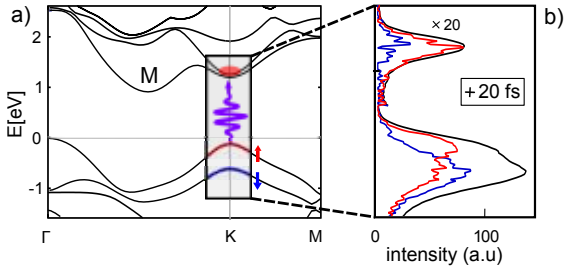
Ultrafast & XUV Science

Spin-resolved electronic dynamics in bulk WSe_2

R. Bertoni *(Institut de physique de Rennes, Université de Rennes 1, Rennes, France)*
C. Nicholson, L. Rettig, R. Ernstorfer *(Fritz-Haber-Institute der Max-Planck-Gesellschaft Faradayweg, Berlin, Germany)*
C. Monney *(University of Fribourg, Department of Physics, Fribourgh, Switzerland)*
M. Hoesch *(DESY Photon Science, Notkestrasse, Hamburg, Germany)*
C. Cacho *(Diamond Light Source, Harwell Campus, Didcot, UK)*
E. Springate, R.T. Chapman *(Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, UK)*

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.



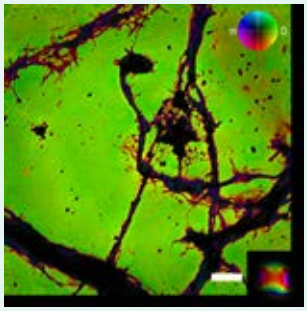
a) DFT calculations of the band structure of bulk WSe_2. At the K point, the two valence bands are purely spin polarized. The material is excited with 1.5 eV circular polarized light pulse populating the conduction band in the K valleys. The grey shaded area corresponds to the probed area with the spin-resolved detector.
b) Spin resolved photo-emitted signal 20 fs after photo-excitation. The red and blue curves correspond to the projected spin polarization of the photo-emitted electron. The generated photo-excited states in K valleys are almost purely spin-polarized.

Contact: R. Bertoni (roman.bertoni@univ-rennes1.fr)

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

P.D. Baksh, W.S. Brocklesby *(Optoelectronics Research Centre, University of Southampton, UK)*
M. Odstrčil *(Optoelectronics Research Centre, University of Southampton, UK; RWTH Aachen, Germany)*
M. Miszczak *(Optoelectronics Research Centre, University of Southampton; University of Padova, Italy)*
R. Chapman, E. Springate *(Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, UK)*
A. Wyatt *(Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, UK; Clarendon Laboratory, Department of Physics, University of Oxford, UK)*
J.G. Frey *(Department of Chemistry, University of Southampton, UK)*

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.



Transmission image at 29 nm of 7DIV neurons grown on SiN substrate. Scale bar is 10 μm.

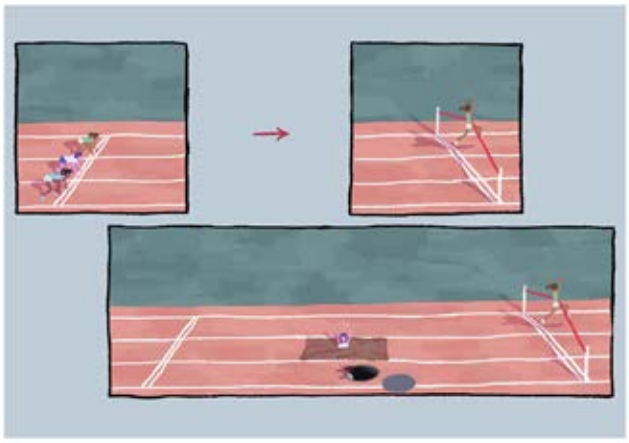
Contact: W.S. Brocklesby (W.S.Brocklesby@soton.ac.uk)

Observing the complete reaction pathway of CS_2 dissociation

A.D. Smith,, E.M. Warne, R.S. Minns *(Chemistry, University of Southampton, UK)*
D. Bellshaw, M. Tudorovskya, A. Kirrander *(EaStCHEM, School of Chemistry, University of Edinburgh, UK)*
A.J. Jones, P.E. Majchrzak, C. Cacho, E. Springate, R.T. Chapman *(Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, UK)*
D.A. Horke *(Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany; The Hamburg Centre for Ultrafast Imaging, Universität Hamburg, Hamburg, Germany)*

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.

Contact: R.T. Chapman (richard.chapman@stfc.ac.uk)
R.S. Minns (r.s.minns@soton.ac.uk)



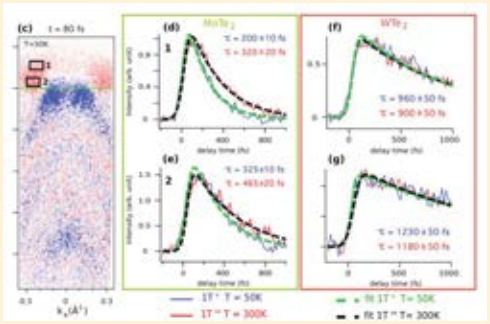
An artist's impression of the reported study's meaning and significance: if one only snapshots the beginning and the end of the reaction (here metaphorically depicted as a race), one is liable to miss the crucial events happening in-between, and misinterpret the entire situation. Courtesy of Helen Towrie, CLF

Out-of-equilibrium electronic dynamics of the transition metal dichalcogenides $MoTe_2$ and WTe_2

A. Crepaldi, G. Gatti, S. Roth, Ph. Bugnon, A. Magrez, H. Berger, M. Grioni *(Institute of Physics, Ecole Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland)*
G. Autès, O.V. Yazyev *(Institute of Physics, Ecole Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland ; National Centre for Computational Design and Discovery of Novel Materials MARVEL, Ecole Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland)*
A. Sterzi, G. Manzoni, F. Parmigiani *(Universitá degli Studi di Trieste - Via A. Valerio 2, Trieste 34127, Italy)*
M. Zacchigna *(C.N.R. - I.O.M., Strada Statale 14, km 163.5, Trieste 34149, Italy)*
C. Cacho, R.T. Chapman, E. Springate *(Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, UK)*
E.A. Seddon *(The Photon Science Institute, The University of Manchester, Manchester, UK; The Cockcroft Institute, Sci-Tech Daresbury, Warrington, UK)*

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, $MoTe_2$ and WTe_2. 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 $MoTe_2$ is found to act like a bottleneck. From the comparison with WTe_2, and the observation of a longer relaxation dynamics, we conclude that this material is a topological trivial semimetal.

Contact: A. Crepaldi (alberto.crepaldi@epfl.ch)



(c) Difference between the ARPES image 80 fs after and 200 fs before optical excitation. (d) - (e) Population dynamics of $MoTe_2$ integrated at 50 K (blue) and 300 K (red) in the area highlighted in panel (c) by the rectangles. (f) – (g) Comparison between the population dynamics measured at 50 K (blue) and 300 K (red) in WTe_2. The best fit and the corresponding characteristic time are shown, as well.

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

H.H. Fielding, M.A. Parkes, J.L. Woodhouse, Y. Zhang, G. Thornton *(Department of Chemistry, University College London, UK)*
K.L. Reid, L. Whalley *(Department of Chemistry, University of Nottingham, UK)*
R.S. Minns *(Chemistry, University of Southampton, UK)*

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 $2^1\pi\pi^*$ excited state. The preliminary results highlighted here indicate that it is practical to perform this experiment at Artemis.

Contact: H.H. Fielding (h.h.fielding@ucl.ac.uk)

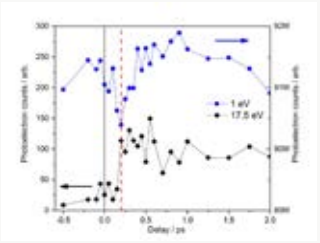


Figure 1 (left): Integrated photoelectron signal as a function of pump-probe delay. Top (blue) curve is in the region of the UV-pump + UV-pump signal, and the bottom (black) curve is in the region of the UV-pump + XUV-probe signal.

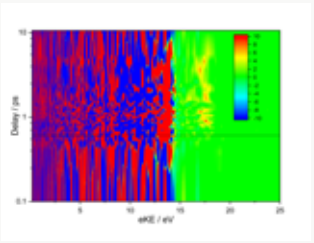


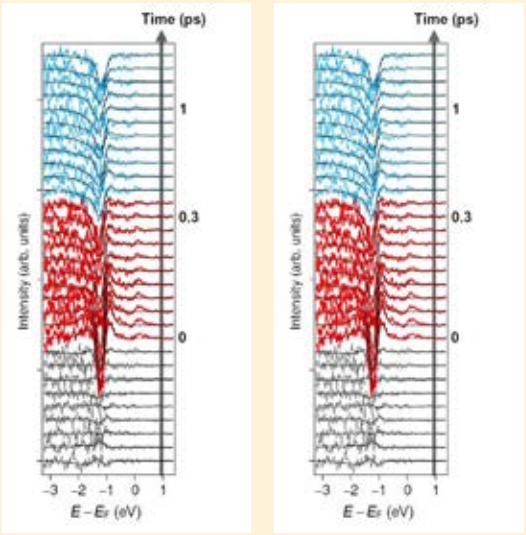
Figure 2 (right): Difference plot of the photoelectron spectra recorded using a 250 nm pump and 21.7 eV probe as a function of time. Note the increase in signal around 17.5 eV. This is the region where pump-probe photoionisation is observed.

Dynamics of correlation-frozen antinodal quasiparticles in superconducting cuprates

C. Giannetti, S. Peli *(Interdisciplinary Laboratories for Advanced Materials Physics, Università Cattolica del Sacro Cuore, I-25121 Brescia, Italy)*
F. Cilento *(Elettra-Sincrotrone Trieste S.C.p.A., Basovizza, Italy)*
G. Manzoni, A. Sterzi *(Elettra-Sincrotrone Trieste S.C.p.A., Basovizza, Italy; Dipartimento di Fisica, Università degli Studi di Trieste, Trieste, Italy)*
A. Ronchi *(Interdisciplinary Laboratories for Advanced Materials Physics, Università Cattolica del Sacro Cuore, Brescia, Italy; Department of Physics and Astronomy, Katholieke Universiteit Leuven, Celestijnenlaan, Leuven, Belgium)*
A. Crepaldi *(Institute of Physics, Ecole Polytechnique Fédérale de Lausanne, Lausanne, Switzerland)*
F. Boschini, M. Berciu, A. Damascelli *(Department of Physics and Astronomy, University of British Columbia, Vancouver, British Columbia, Canada; Quantum Matter Institute, University of British Columbia, Vancouver, British Columbia, Canada)*
C. Cacho, R. Chapman, E. Springate *(Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, UK)*
H. Eisaki *(Nanoelectronics Research Institute, National Institute of Advanced Industrial Science Technology, Tsukuba, Ibaraki, Japan)*
M. Greven *(School of Physics and Astronomy, University of Minnesota, Minneapolis, USA)*
A.F. Kemper *(Department of Physics, North Carolina State University, Raleigh, USA)*
M. Capone *(Scuola Internazionale Superiore di Studi Avanzati (SISSA) and Consiglio Nazionale delle Ricerche–IstitutoOfficina deiMateriali (CNR-IOM)Democritos National Simulation Center, Via Bonomea, Trieste, Italy)*
F. Parmigiani *(Elettra-Sincrotrone Trieste S.C.p.A., Basovizza, Italy; Dipartimento di Fisica, Università degli Studi di Trieste, Trieste, Italy; International Faculty, University of Cologne, Albertus-Magnus-Platz, Germany)*

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 bands, 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.

Contact: C. Giannetti (claudio.giannetti@unicatt.it)



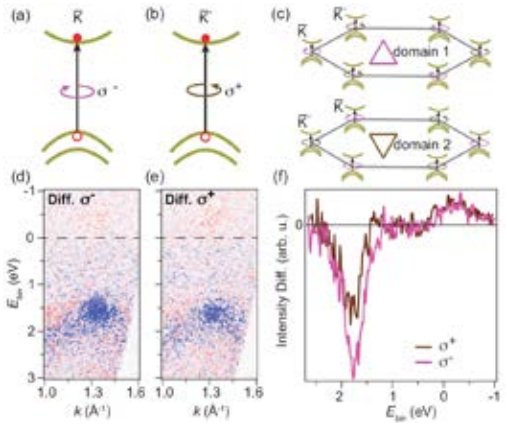
Differential EDC curves as a function of the time delay. The colours highlight three different characteristic temporal regions corresponding to negative delays (grey traces), short dynamics characterized by a transient broadening of the O-2p_π peak (red traces), and long dynamics characterized by a long-lived decrease of the O-2p_π peak spectral weight. The black lines are the differential fit to the data obtained by assuming both a Gaussian broadening and a spectral weight decrease of the O-2p_π.

Spin and valley control of free carriers in single-layer WS_2

S. Ulstrup *(Advanced Light Source, E.O. Lawrence Berkeley National Laboratory, Berkeley, California, USA)*
A. Grubišić Čabo, M. Dendzik, C.E. Sanders, M. Bianchi, J. A. Miwa, P. Hofmann *(Department of Physics & Astronomy, Interdisciplinary Nanoscience Center, Aarhus University, 8000 Aarhus C, Denmark)*
D. Biswas, J.M. Riley, P.D.C. King *(SUPA, School of Physics and Astronomy, University of St Andrews, St Andrews, UK)*
C. Cacho, D. Matselyukh, R.T. Chapman, E. Springgate *(Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, UK)*

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 (WS_2) 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.

Contact: J.A. Miwa (miwa@phys.au.dk)

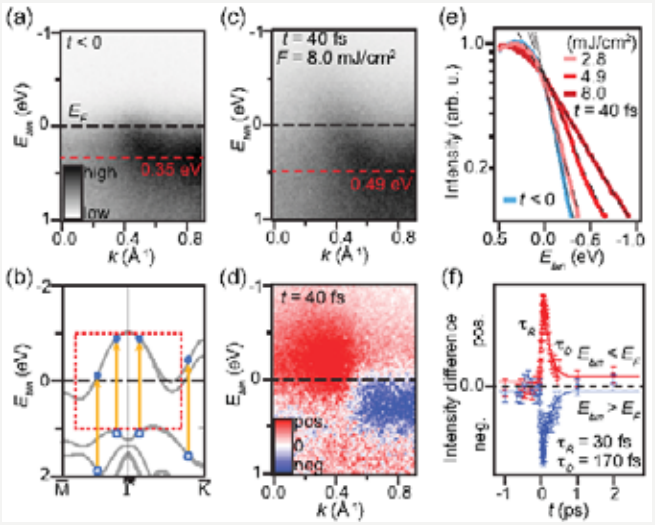


(a),(b) Schematics illustrating the selection rules for transitions from the upper VB to the CBM in the K and K' valleys. (c) BZ orientations and selection rules for the two mirror domains of the SL WS_2. (d),(e) Difference signal for optical pumping with (d) σ⁺- and (e) σ⁻-polarization. (f) EDCs of the difference integrated over the VBM and CBM regions for the data in (d) and (e).

Carrier dynamics in a two-dimensional metal

A. Grubišić Čabo, F. Andreatta, C. E. Sanders, M. Bianchi, J.A. Miwa, P. Hofmann, S. Ulstrup *(Department of Physics & Astronomy, Interdisciplinary Nanoscience Center, Aarhus University, 8000 Aarhus C, Denmark)*
H. Rostami, A. Balatsky *(Condensed Matter, Statistical and Biological Physics, NORDITA, Roslagstullsbacken, Stockholm, Sweden)*
D. Biswas, I. Marković, P.D.C. King *(SUPA, School of Physics and Astronomy, University of St Andrews, St Andrews, UK)*
C. Cacho, P. Majchrzak, R.T. Chapman, E. Springgate *(Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, UK)*

We provide the first direct femtosecond study of single-layer (SL) tantalum disulphide (TaS_2) — 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-TaS_2, the electrons reach an exceptionally hot 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.



TR-ARPES measurement of the 1H-TaS_2 dispersion around the Fermi level. (a) Measured spectrum before optical excitation, i.e. t<0. (b) Calculated dispersion from Ref. [4] with possible excitation processes. (c) TR-ARPES data acquired 40 fs after arrival of the pump pulse. (d) Difference spectrum obtained by subtracting panel (a) from (b). Red (blue) corresponds to excited electrons (holes). (e) Plot of the momentum-integrated intensity over the measured region in panel (a) and (c) for the stated time delays and pump fluence. Time-dependent intensity difference (markers) summed above and below the Fermi level.

Contact: J. A. Miwa (miwa@phys.au.dk)