Ultrafast & XUV Science

Observation of charge density wave melt down in time resolved ARPES of single layer VSe_2

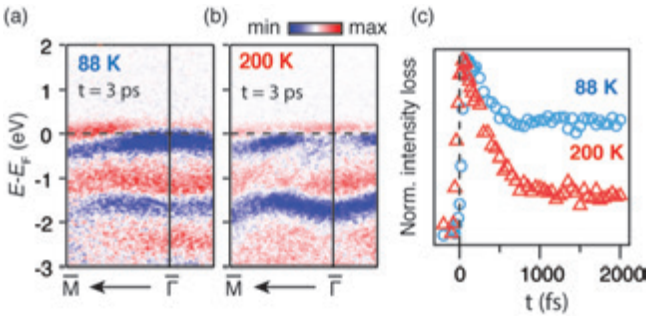
D. Biswas, A. Jones, K. Volckaert, C. Sanders, F. Andreatta, J. Miwa, P. Hofmann, N. Lanata, S. Ulstrup *(Department of Physics & Astronomy, Aarhus University, Denmark)*
P. Majchrzak, Y. Zhang, G. Karras, R.T. Chapman, A. Wyatt, E. Springate *(Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, UK)*

B.K. Choi, Y.J. Chang *(Department of Physics, University of Seoul, Republic of Korea)*
T.H. Lee, C.J. Kang *(Department of Physics & Astronomy, Rutgers University, Piscataway, New Jersey, USA)*
I. Marković, P.D.C. King *(SUPA, School of Physics & Astronomy, University of St Andrews, UK)*

Group V transition metal dichalcogenide (TMD) VSe_2 shows charge density wave (CDW) transition at 110 K with the CDW ordering (4 × 4 × 3), in its bulk form. Recent experiments have shown that the charge ordering gets enhanced in the monolayer (ML) VSe_2 with an increased transition temperature ~ 140 K. Moreover, in CDW phase almost the whole Fermi surface vanishes, forming CDW gap.

Multiple charge order structures are reported in literature – (4 × 4), (√3 × 2), (√3 × √7). It is very clear that the nature and the mechanism of CDW in bulk and ML VSe_2 are very different. Here we use time and angle resolved photoelectron spectroscopy (trARPES) to study the electron dynamics in both CDW and normal phase, and hence to shed light on the CDW mechanism in ML VSe_2. We have also simulated the ARPES intensity to extract different many-body and transient parameters.

Our measurements showed the melt down of the charge order on pumping, but there was no recovery even after 3 ps. This unusually long-time dependence points towards the possibility of a bottleneck in the hot carrier relaxation and the crucial role of the substrate in ML VSe_2.



(a), (b) The difference spectra at 3 ps time delay for 88 K and 200 K sample temperature respectively – different hot carrier dynamics

(c) Intensity variation close to $\bar{\Gamma}$ point showing the loss of back folded intensity for sample at 88 K after pumping – melting of charge order

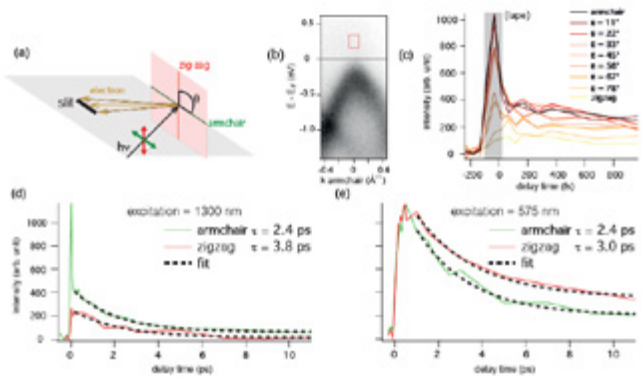
Contact: D. Biswas (deepnarayan7@phys.au.dk)

Assessing the anisotropic response in the out-of-equilibrium electron dynamics of black phosphorus

A. Crepaldi, S. Roth, G. Gatti, S. Polishuck, M. Puppin, M. Chergui, M. Grioni *(École Polytechnique Fédérale de Lausanne (EPFL), Switzerland)*
A.S. Wyatt, R.T. Chapman, G. Karras, Y. Zhang *(Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, UK)*

F. Grillo *(EaStCHEM and School of Chemistry, University of St. Andrews, UK)*

One of the most striking aspects of the physics of black phosphorus (BP) is the large anisotropy in its transport and optical properties. We have investigated the electron dynamics in the conduction band of BP, which is transiently populated by an ultrafast optical excitation. We resolve a change in the characteristic relaxation (τ) time when the light polarization is varied in the surface plane, from the armchair to the zig-zag direction. Our results suggest that the orbital polarization, which is responsible for the dichroism in the optical absorption, might also influence the electronic scattering mechanisms.



a) Experimental geometry. (b) Band dispersion of BP measured along the armchair direction at the G point of the 2nd Brillouin Zone. (c) Electron dynamics at the bottom of conduction band, in the region indicated in panel (b) by a red rectangle. The colour of the curves encodes the evolution of the pump polarization direction from armchair to zig-zag. (d) Comparison between the dynamics for an optical excitation centred at 1300 nm. The characteristic decay times are indicated. (e) The same comparison, but for an optical excitation centred at 575 nm.

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

A new three-dimensional probe of molecular structure and dynamics on the ultrafast timescale

J.W.L. Lee, H. Köckert, D. Heathcote, D. Popat, C. Vallance *(Department of Chemistry, University of Oxford)*

R. Chapman, P. Majchrzak, E. Springate *(Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, UK)*

Ultrafast laser pump-probe methods allow chemical reactions to be followed in real time, and have provided unprecedented insight into some of the most fundamental aspects of chemical reactivity. While evolution of the electronic structure of the system under study is immediately evident from changes in the observed spectral signatures, information on rearrangement of the nuclear framework is generally obtained indirectly. Disentangling the contributions to the signal from competing photochemical pathways can also be challenging in many cases.

Here, we introduce the new technique of three-dimensional covariance-map Coulomb explosion imaging, which has the potential to provide complete three-dimensional information on molecular structure and dynamics as they evolve in real time during a gas-phase chemical reaction. We describe our experimental approach and present first proof-of-concept data from recent measurements on CF_3I, performed at the Artemis ultrafast laser facility. Covariance analysis of the data allows the contributions from competing fragmentation pathways to be isolated and characterised. With further development, we envisage that the method will allow 'molecular movies' to be recorded for a wide variety of gas-phase chemical processes.

Reprinted from Lee, J.W.L., Köckert, H., Heathcote, D. et al. Three-dimensional covariance-map imaging of molecular structure and dynamics on the ultrafast timescale. Commun Chem 3, 72 (2020), under the terms of the Creative Commons CC BY license . doi: 10.1038/s42004-020-0320-3

Contact: C. Vallance (claire.vallance@chem.ox.ac.uk)

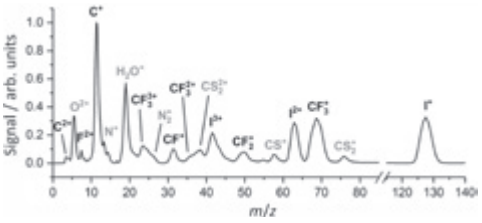


Figure 1: Time-of-flight spectrum for the charged fragments formed in the laser-induced Coulomb explosion of CF_3I

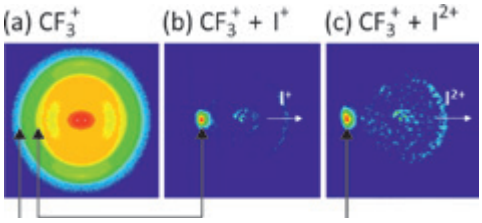


Figure 2: (a) 2D projection of the 3D velocity distribution recorded for CF_3^+ fragments; (b) covariance-map image of CF_3^+ formed with an I^+ partner ion; (c) covariance-map image of CF_3^+ formed with an I^{2+} partner ion

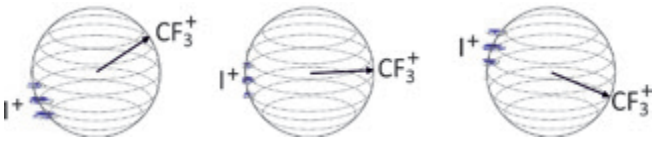


Figure 3: 3D covariance-map images for the $CF_3^+ + I^+$ fragmentation channel

Femtosecond UV and XUV Photoelectron Spectroscopy of Dissociating Methyl Iodide

B. Downes-Ward, E.M. Warne, J. Woodhouse, R.S. Minns *(Chemistry, University of Southampton, UK)*
A.S. Wyatt, P. Majchrzak, G. Karras, Y. Zhang, E. Springate, R.T. Chapman *(Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, UK)*

M.A. Parkes *(Department of Chemistry, University College London, UK)*

Pump probe photoelectron spectroscopy using UV and XUV probe photon energies has been used to monitor the ultrafast photodissociation of methyl iodide. The UV photoelectron imaging measurements provide a detailed view of the changing structure that is limited to very early times close to the initial geometry. By contrast the XUV photoelectron spectroscopy dynamics show the changes occurring over the full reaction. The two measurement provide complementary information on the dissociating molecule, allowing us to uncover complexities in the dynamics that have previously been missed.

Summary of the photoelectron spectroscopy data showing a photoelectron image (a) and time dependent photoelectron spectrum (b) obtained following excitation at 255 nm and ionisation with 400 nm light, and the photoelectron spectrum obtained following XUV ionisation (c) of the ground (blue) and excited state (red) population

Contact: R.S. Minns (r.s.minns@soton.ac.uk)