Building on the foundations of attosecond physics

We seek to develop:

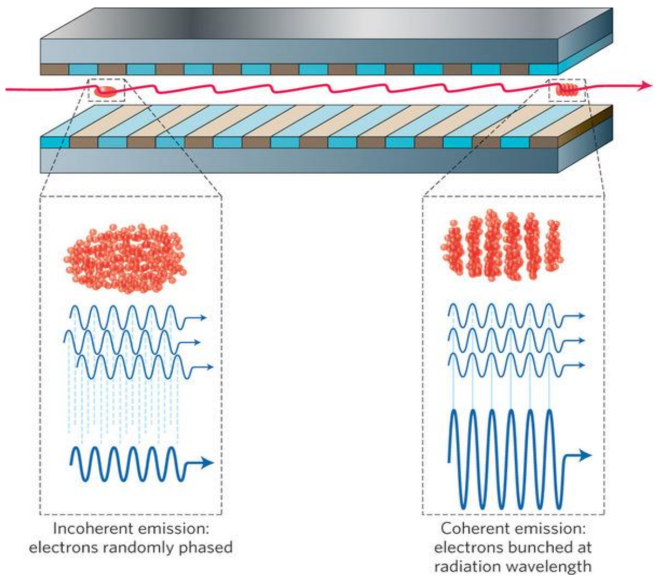
- **High brightness attosecond x-rays from XFELs**
- **Non-linear and linear attosecond spectroscopy**
- **Applications to photo-physics, photo-chemistry and energy materials to capture the electronic dynamics underlaying critical processes**

XFELs offer a new route to measuring ultrafast electron dynamics

Input

Low emittance,
narrow energy spread,
relativisitic electron
bunch
4 – 15 GeV

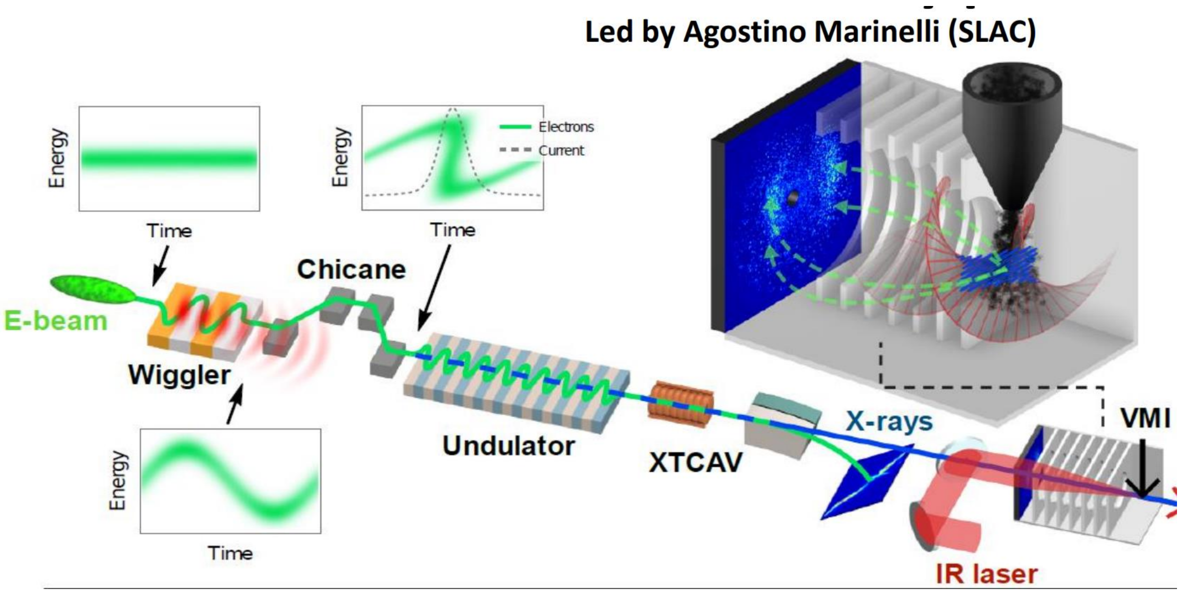
Undulator

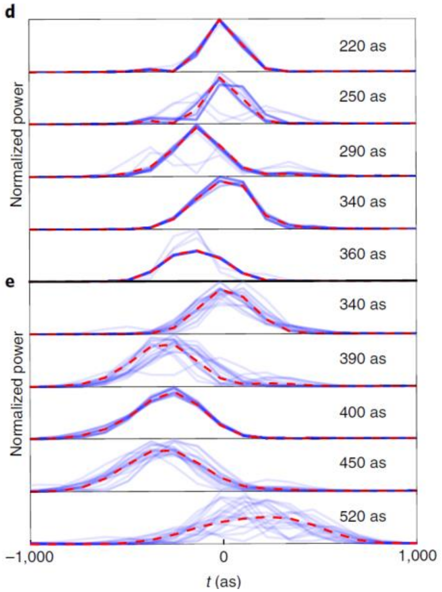


Output

High brightness,
short pulse of coherent
soft to hard X-rays
But multiple random transform limited
pulses

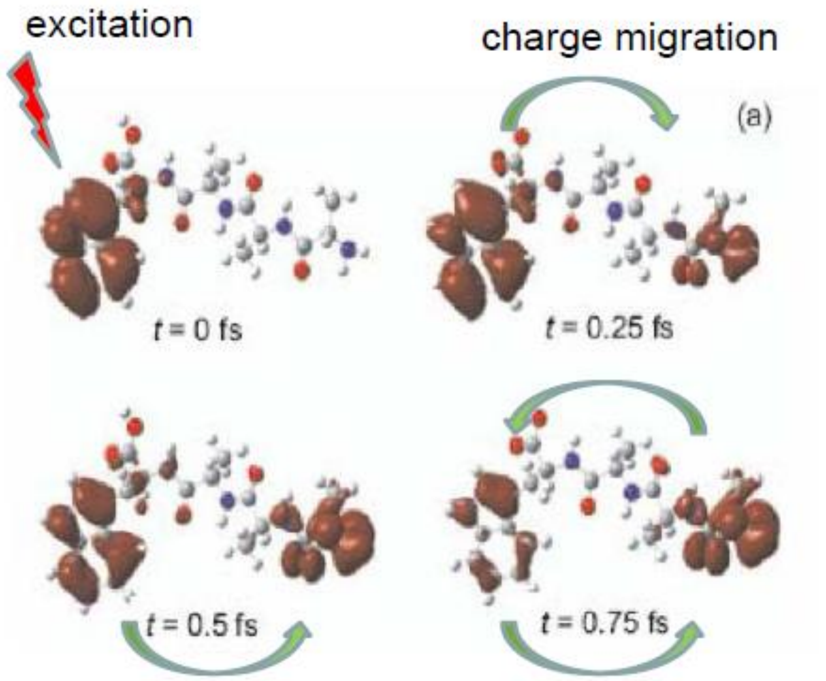
Controlling electron bunch properties to create a high current spike to force lasing in a single transform limited attosecond pulse – achieved at LCLS Stanford 2020





J.Duris et al Nature Photonics 14, 30 (2020)

An example of attosecond dynamics: charge migration



Charge (hole) migration in the peptide Trp-Ala-Ala-Ala (Remacle & Levine Z.Phys.Chem. 221, 647 (2007))

F. Calegari et al, Science 346, 336 (2014)
P. Kraus et al, Science, 350, 790 (2015)

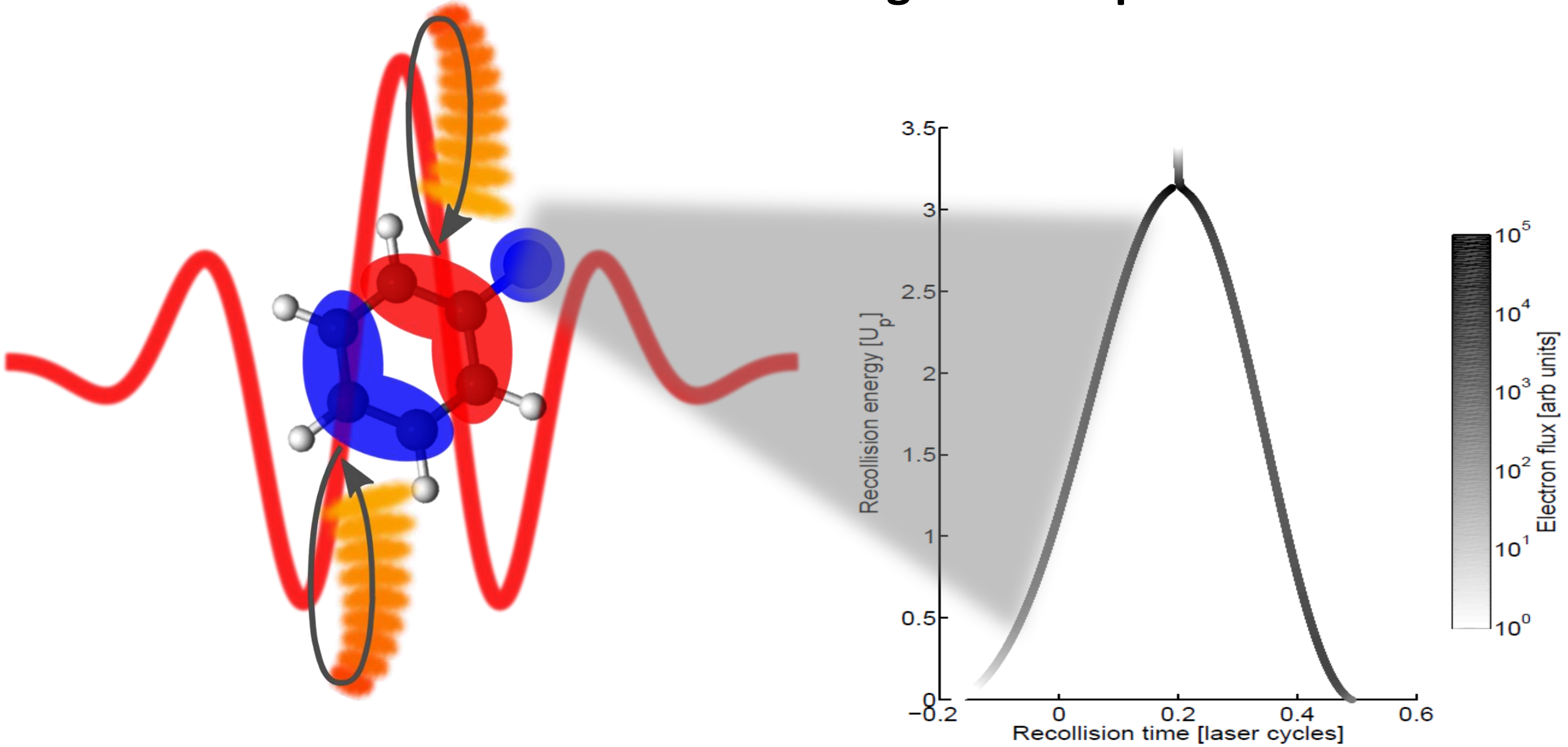
Charge Migration

Sudden electron removal can form a localised hole state that is a coherent superposition of the electronic eigenstates of the molecular ion and so undergoes rapid evolution. This results in large amplitude charge oscillation across the molecule on an attosecond timescale (first identified by Cederbaum et al).

How long does electronic coherence survive?
How does electronic coherence evolve into longer lived vibronic coherence?
What are the consequences for "chemical change"?
Can we control it?

Measurement Strategy: HHG spectroscopy of small molecules

HHG provides an attosecond probe of the response of a molecule to a strong field that can access the first few-fs after ionisation through the chirped harmonic emission

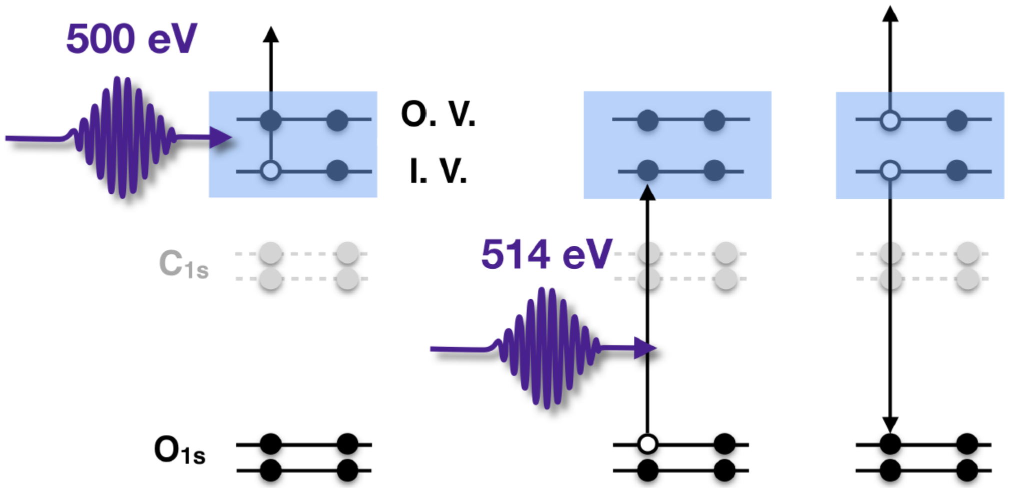


Nuclear Autocorrelation Function
S. Baker *et al.*, **Science 312**, 424 (2006)
J.Marangos **J.Phys.B** (2016)
J.Leeuwenburgh et al, **PRL** 111, 123002 (2013)
D.R.Austin *et al,* **Scientific Reports 11,** 2485, (2021)

Electronic Dynamics
O. Smirnova *et al.*, **Nature** 460 (2009)
P.M.Kraus et al **Science 350 ,** 790 (2015)

Autocorrelation+Electronic Dynamics
Ferte et al **PRL** 133, 203201 (2024)

Measurement strategy: X-ray pump - X-ray probe implemented at LCLS

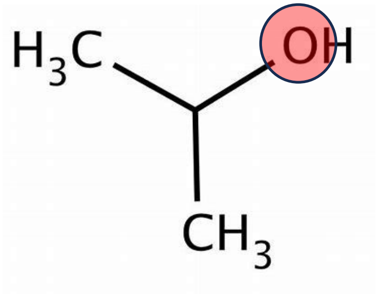


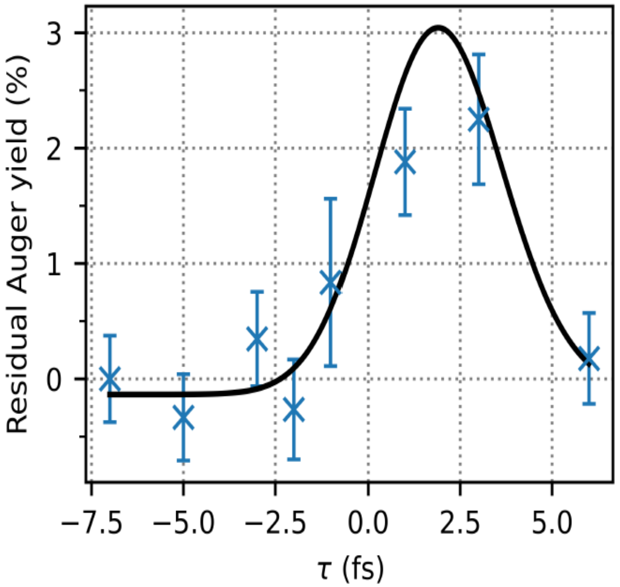
X-ray absorption spectroscopy (XAS) – direct probe of hole state dynamics:

- Localised to a single atomic site.

- Weak field/no perturbation of dynamics

B.Cooper et al Faraday Discussion 171, 93 (2014)





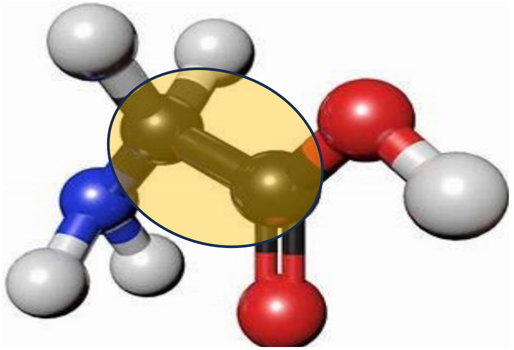
Measurement at LCLS of a highly transient hole (few-fs lifetime) in isopropanol localised near the O atom

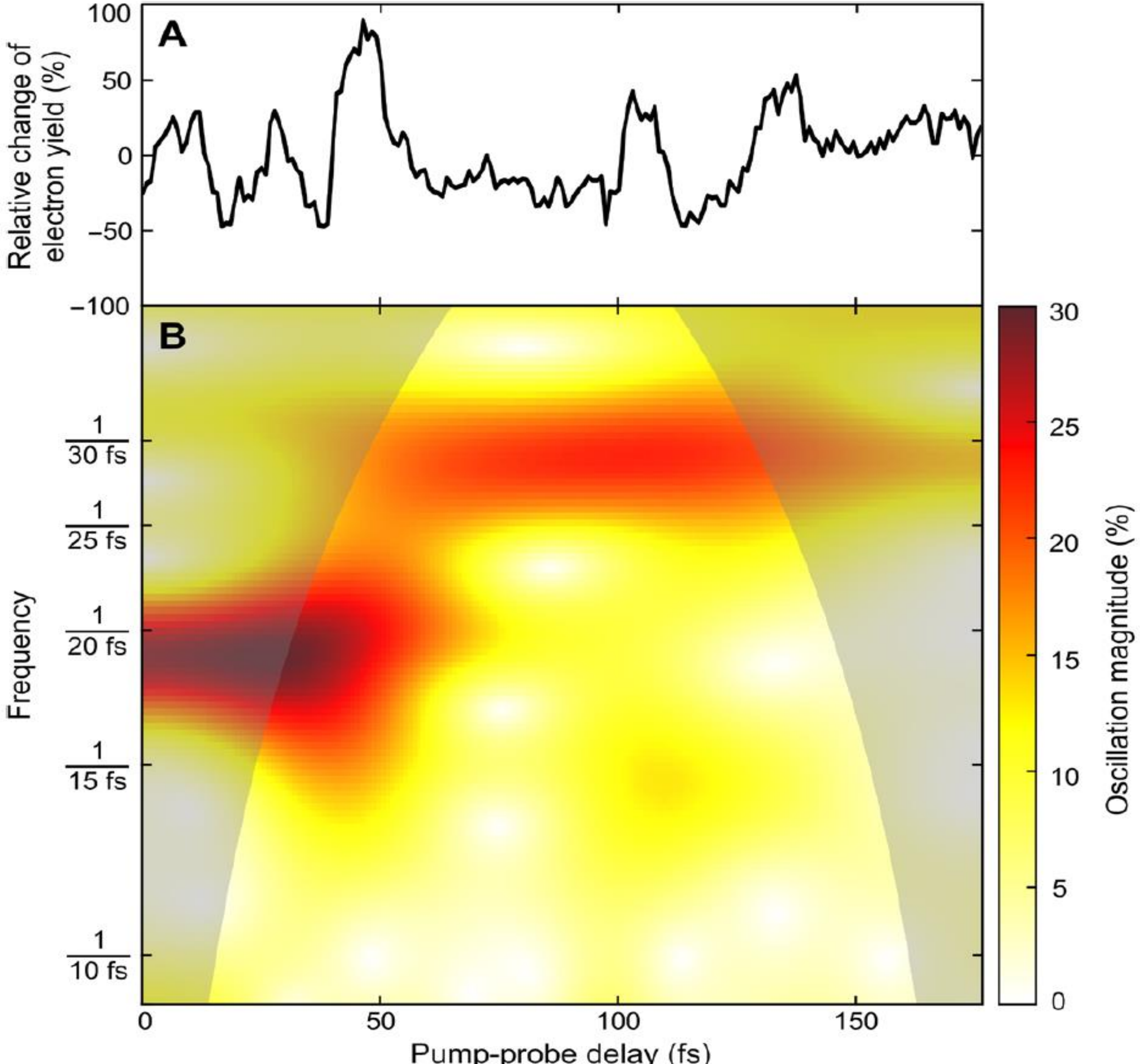
T.Barillot et al, "Correlation Driven Transient Hole Dynamics Resolved in Space and Time in the Isopropanol Molecule", PRX 11, 031048 (2021)

Transient x-ray absorption with 3 fs near transform limited pulses in glycine show initial electronic coherence coupling to vibronic coherence

Tim Laarmann (DESY), Marco Ruberti (Imperial) et al measurements @ FLASH

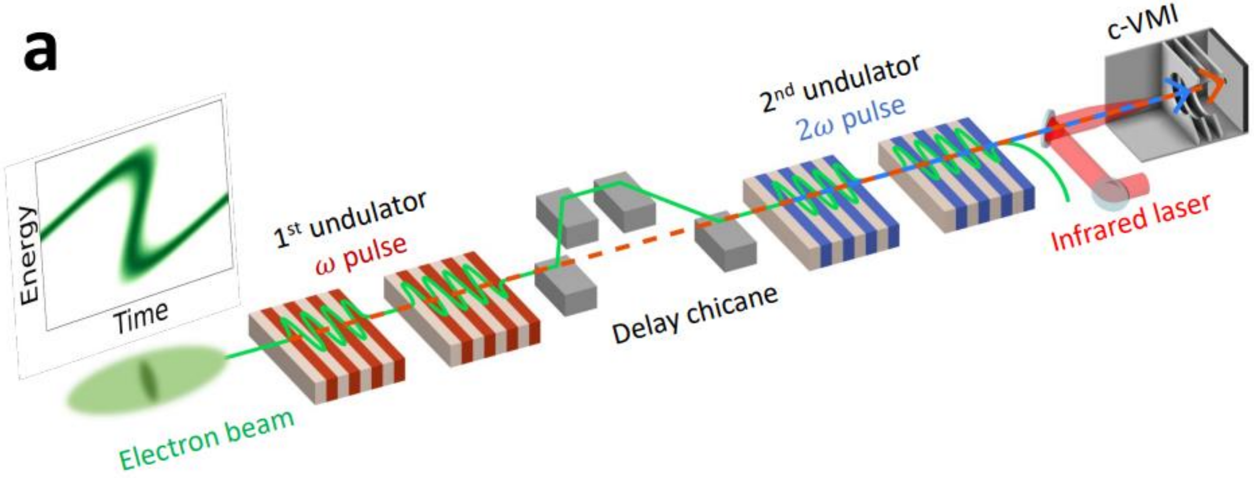
Probing the hole near C sites following x-ray ionisation of glycine we observe both oscillatory charge migration and evidence of vibronic coupling. A near transform limited 3 fs pulse enabled the temporal and energy resolution needed for this measurement.





Schwickert et al, **Science Advances, 8,** eabn6848 (2022) DOI: 10.1126/sciadv.abn6848

Attosecond x-ray pump-probe measurements at LCLS in aminophenol



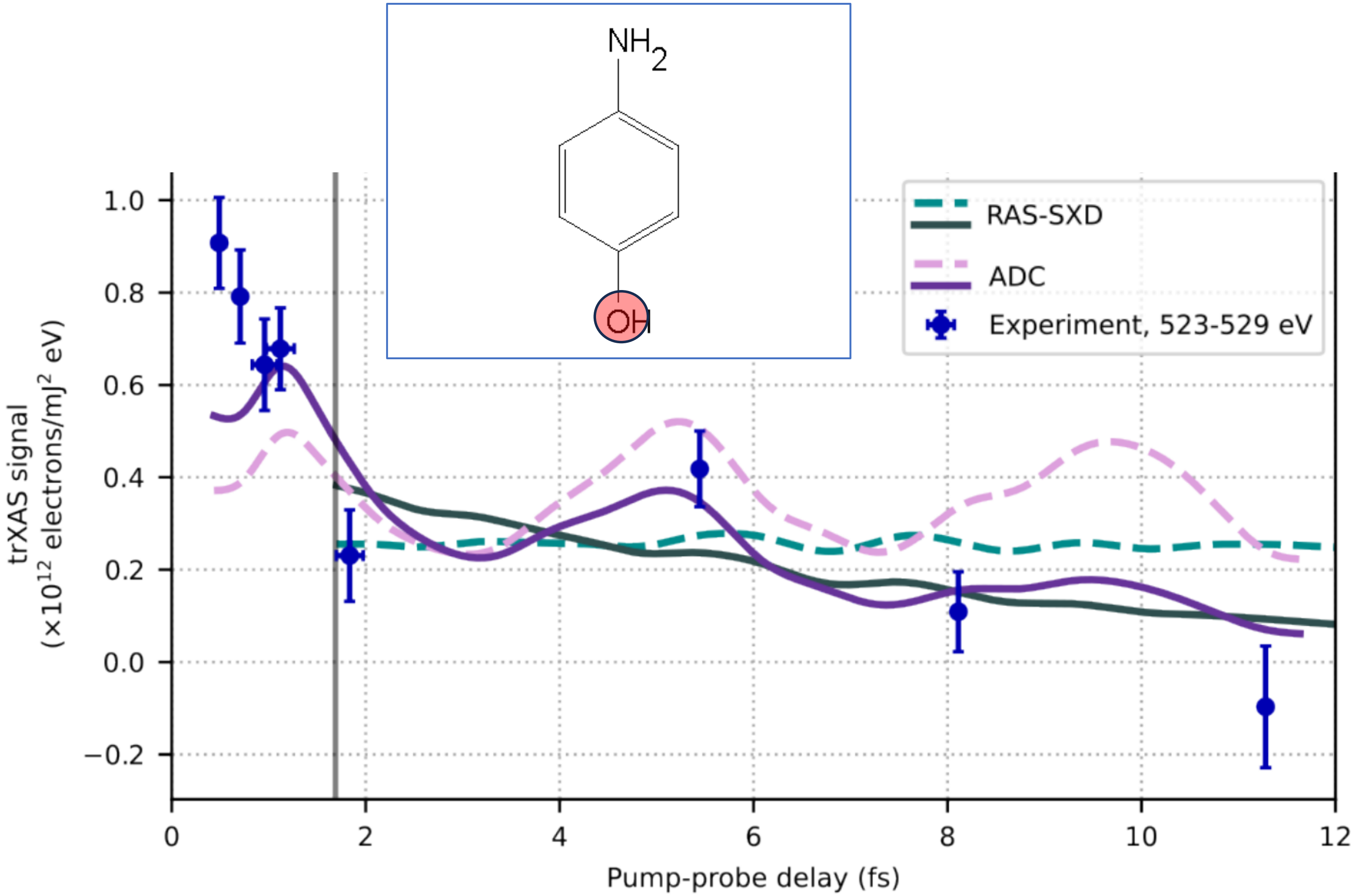
J.Duris et al Nature Photonics 14, 30 (2020)

Z.Ghuo et al Nature Photonics *18* 691 (2024)

Ionisation physics of water ***Science*** eadn6059 (2024)

Measurements @ LCLS with Imperial collaborators "Attosecond Campaign"
ArXiv: 2411.01700v1 (Nov 2024)

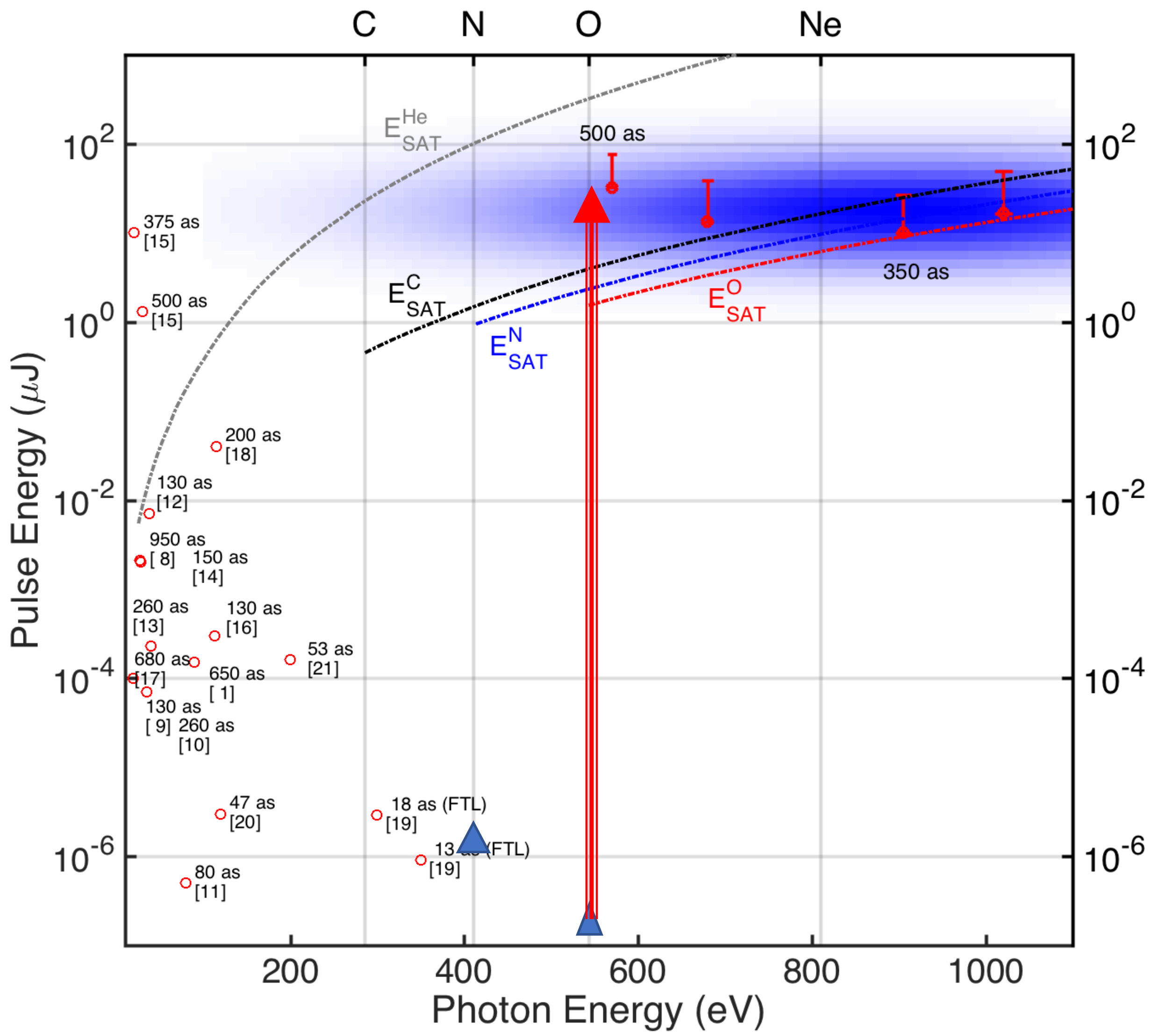
New tools for fundamental understanding of electron-hole dynamics in many-electron systems with applications to the photon-electron coupling in molecules, metals, semiconductors, dielectrics and amorphous systems



Imperial College London

SLAC

XFELs give high brightness attosecond X-ray pulses

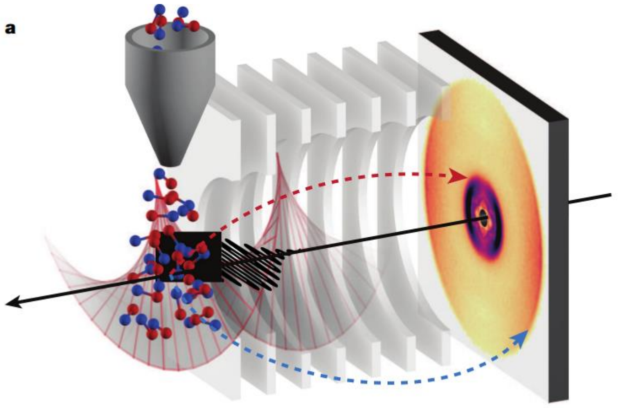


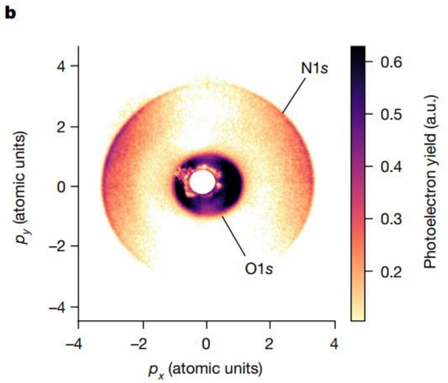
- **Near-transform limited with 5-10 eV coherent bandwidth**
- **8 - 9 orders of magnitude brighter than HHG sources in X-ray range.**
- **Unique source for non-linear (2 or more photon) x-ray interactions and ultrafast measurement:**
- **x-ray pump-probe measurements**
- **x-ray photo/Auger electron streaking**
- **x-ray impulsive electronic Raman**

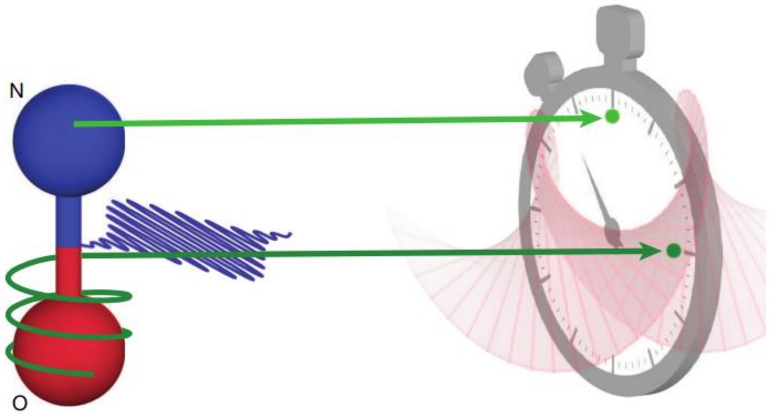
J.Duris et al Nature Photonics 14, 30 (2020)

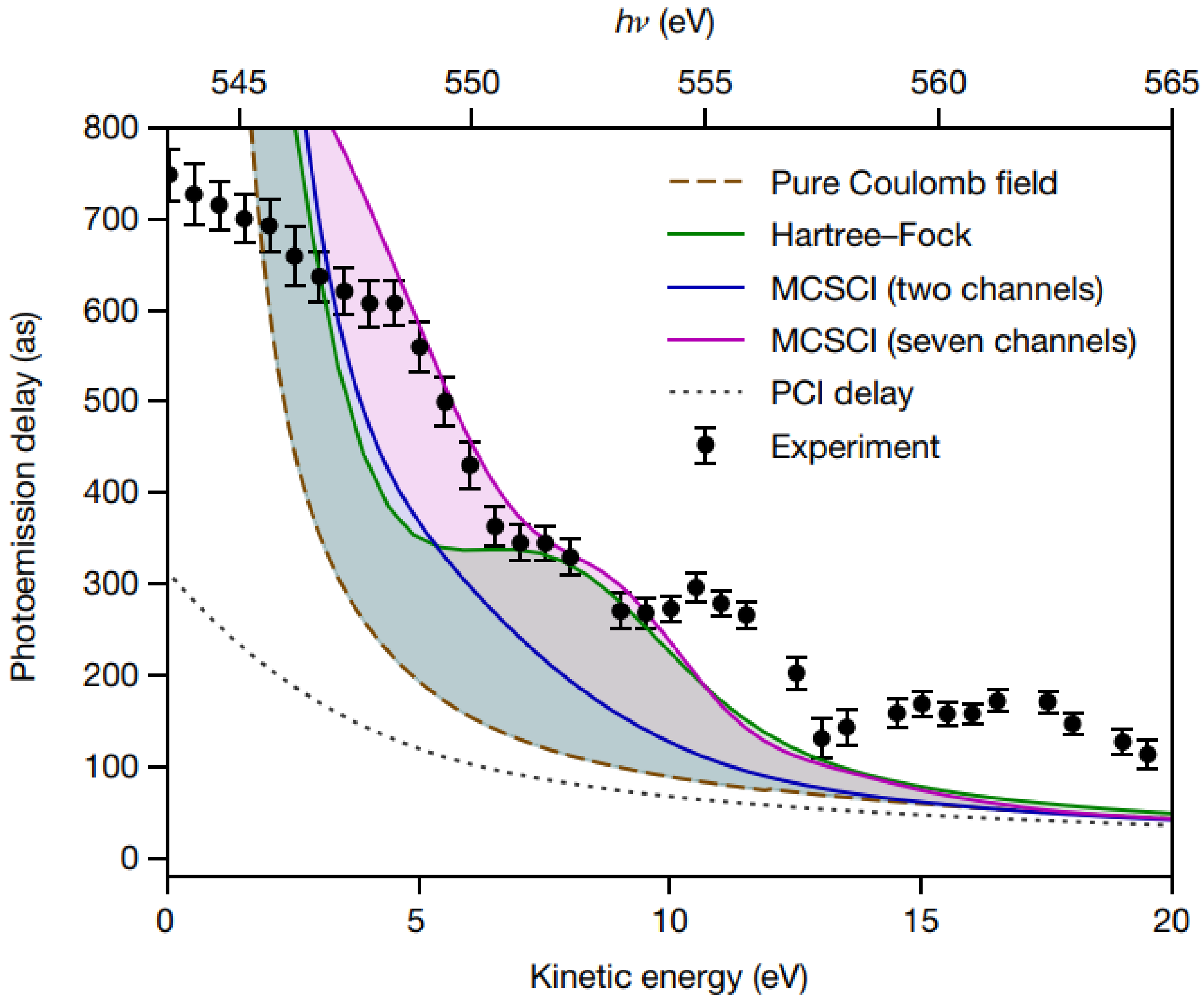
Mapping potential landscape of a molecule through photoelectron time-delays

Led by James Cryan (SLAC) with Imperial and UCL (Agapi and team)





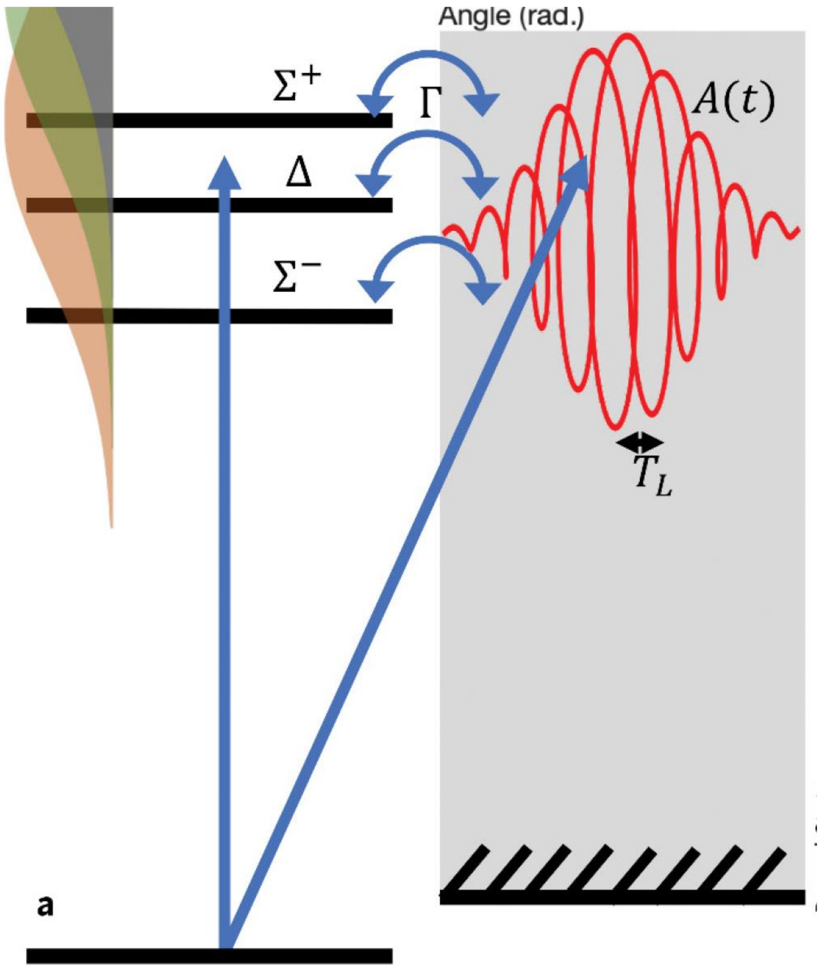


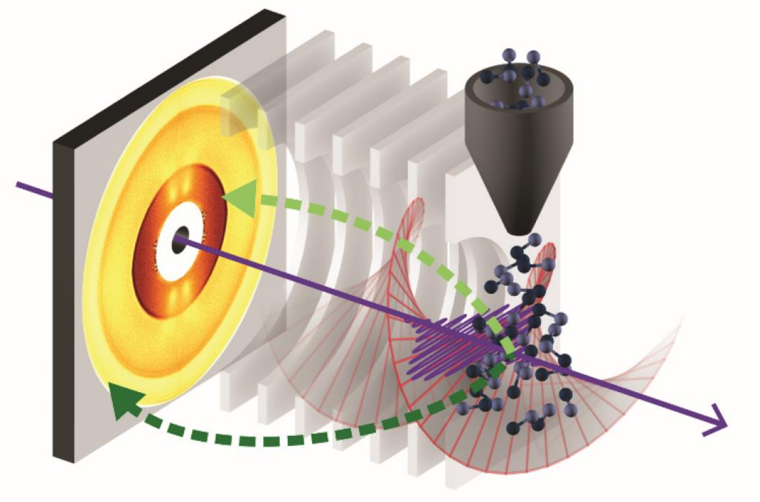


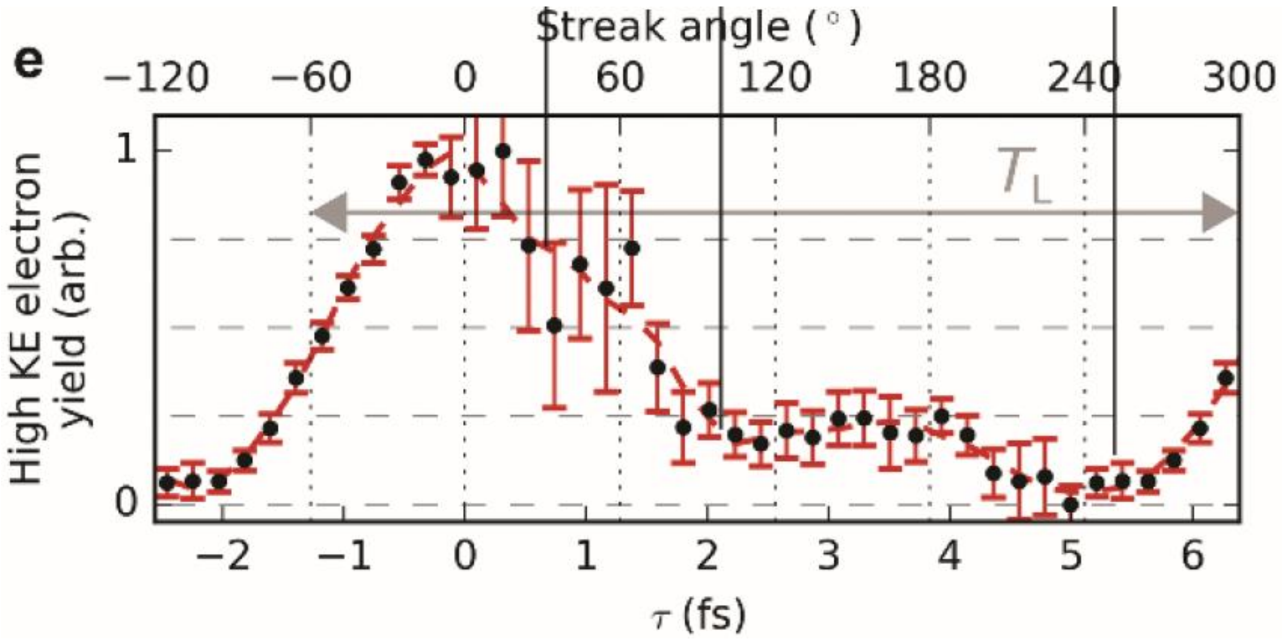
T.Driver et al ***Nature* 632**, *762 (2024)*

Core excited electronic wavepackets in NO – quantum interference in Auger-Meitner decay registered in time-domain

Led by James Cryan (SLAC) with Jon Marangos Team XLC



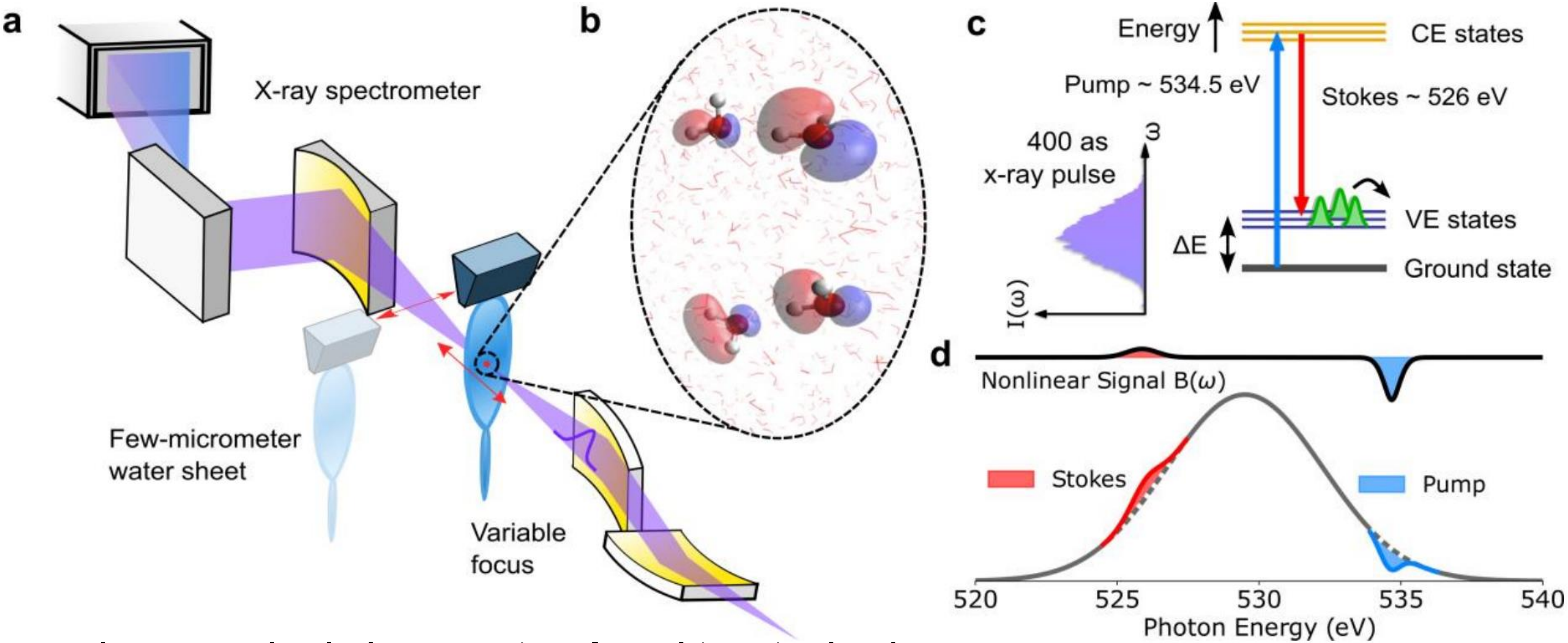




S.Li et al ***Science* 375**, *285 (2022)*

Non-linear X-ray optics in liquid water: Impulsive stimulated electronic Raman with high intensity attosecond X-ray pulses

Imperial College + Stanford, LCLS + Autonoma Madrid



Gas phase NO molecule demonstration of Impulsive Stimulated X-ray Raman Scattering (ISXRS): J.T.O'Neal et al, PRL, 125, 073203 (2020)

Non-linear X-ray optics in liquid water: Impulsive electronic Raman with high intensity attosecond X-ray pulses

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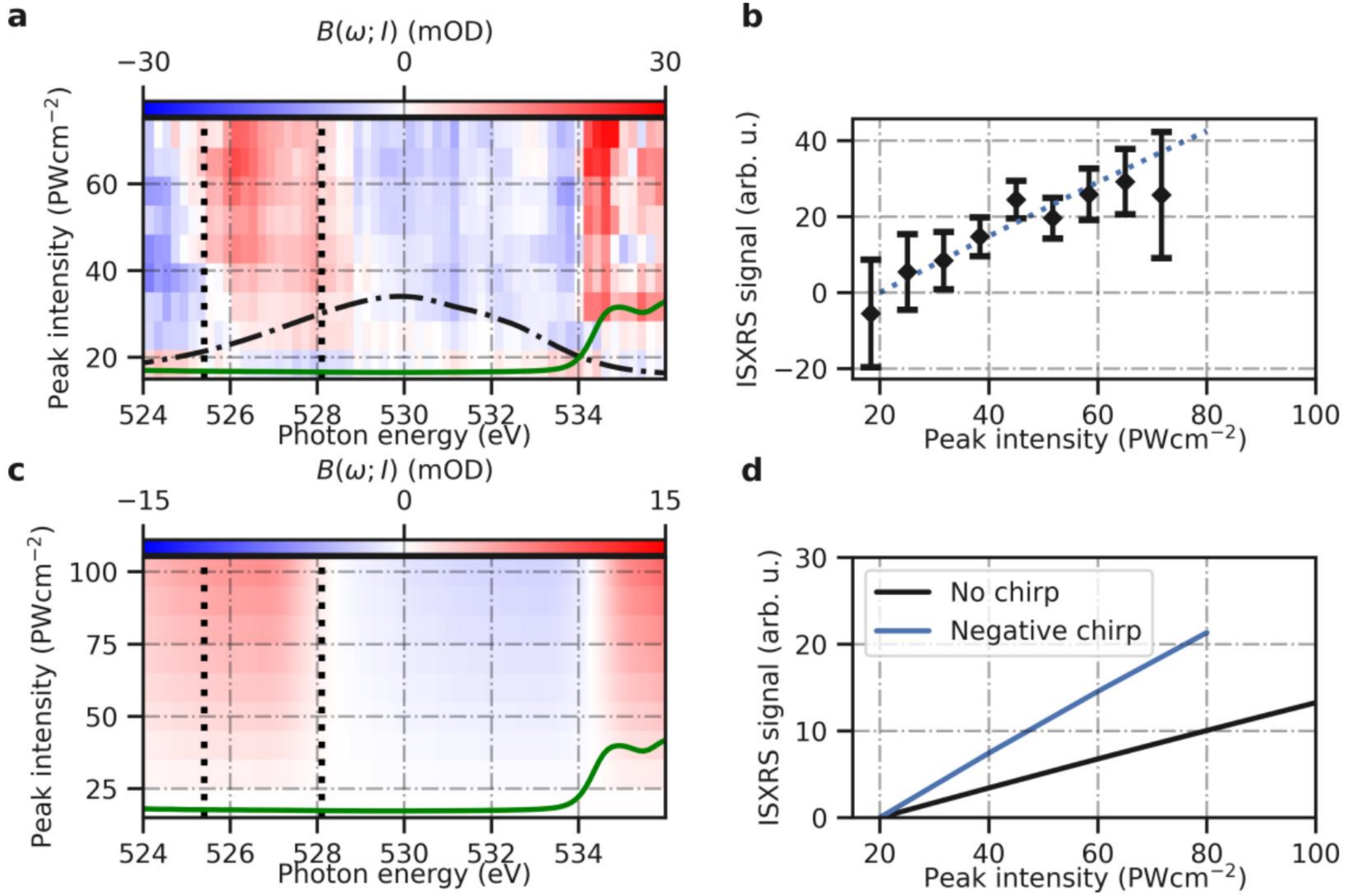
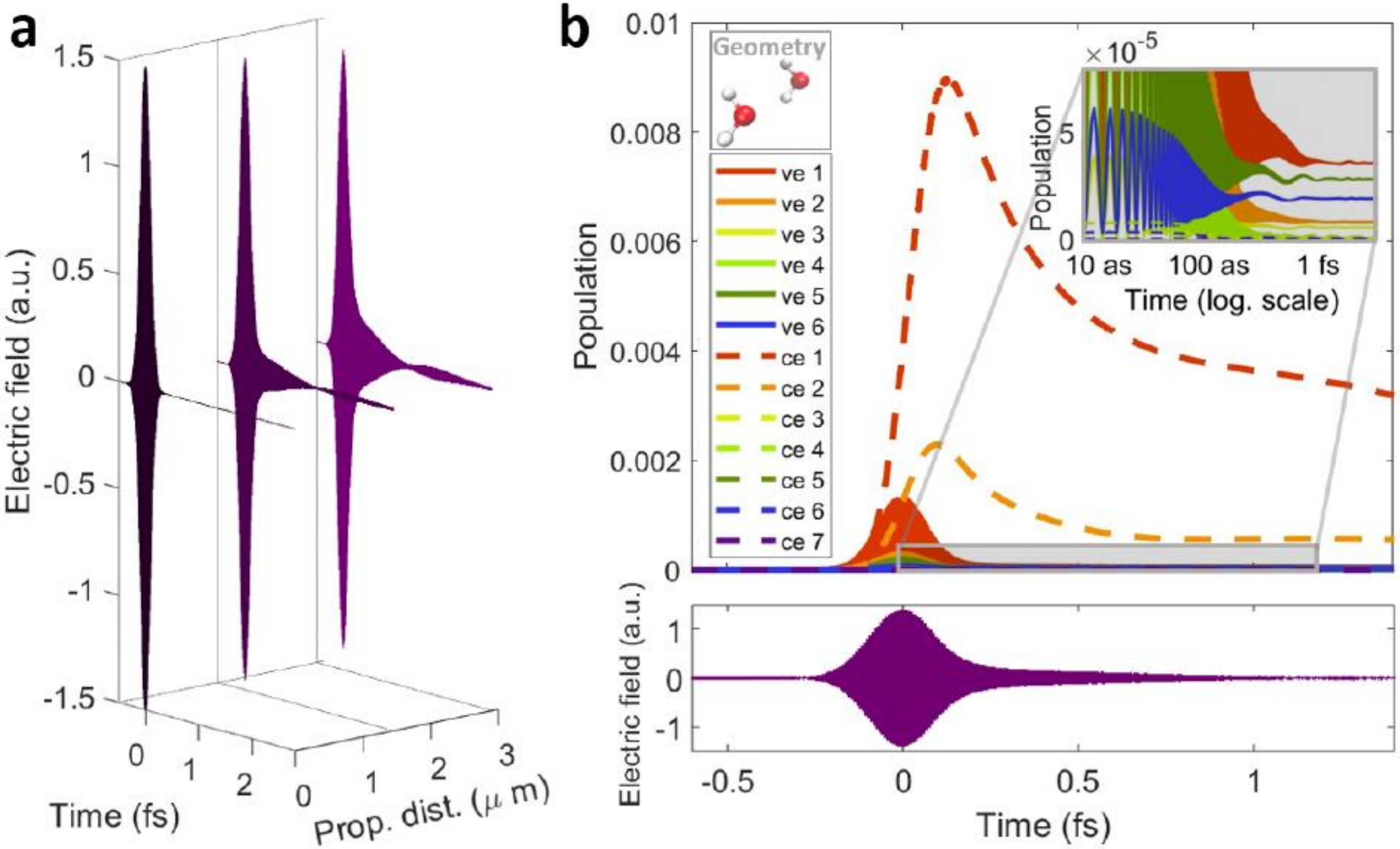


Impulsive electronic Raman creates excited electronic wavepackets localised in space and time in neutral matter

Imperial College + Stanford, LCLS + Autonoma Madrid



Science Advances, 10, eadp0841 (2024)

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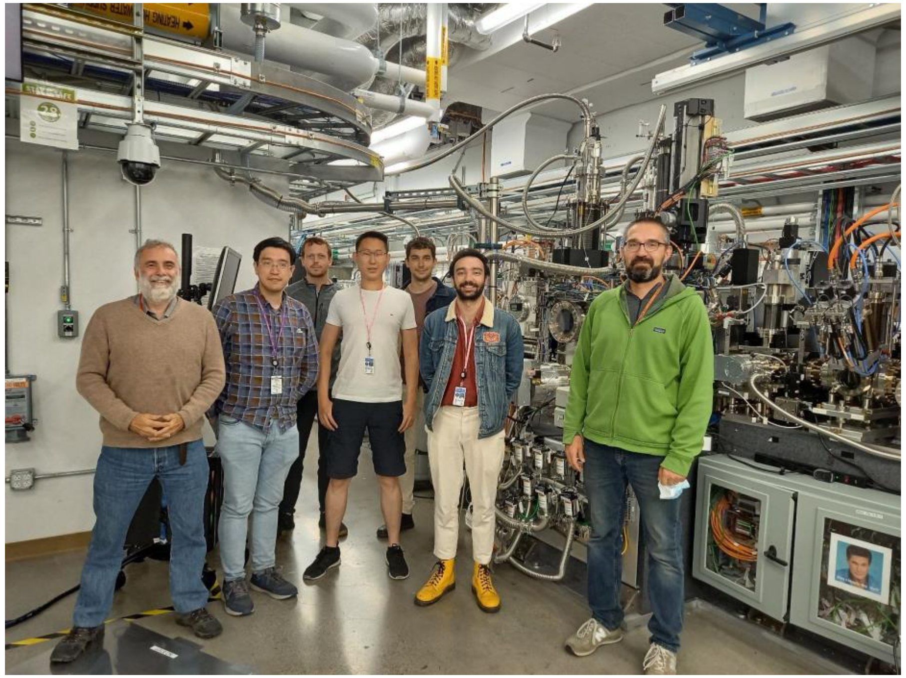
LR25 and LX52 collaborations

Tim Laarmann and DESY "Glycine" collaboration

LCLS Attosecond Campaign



UNIVERSIDAD AUTONOMA DE MADRID

Ana Gutierrez
Antonio Picón
Juanjo Perez
GustavoFernández
Fernando Martin
Gilbert Grell
Alicia Palacios

Imperial College London

Felix Egun
Jacob Lee
Kasia Kowalczyk
Laura Rego
James Turner
Keven Zhao
David Ayuso
Oliver Alexander
Mary Matthews

Taran Driver
Douglas Garratt
Kirk Larsen
Jordan O'Neal
Emily Thierstein
James Cryan
Ryan Coffee
Phillip Bucksbaum

SLAC NATIONAL ACCELERATOR LABORATORY

Georgi Dakovski
Daniel DePonte
Ming-Fu Lin
Agostino Marinelli
Stefan Moeller

Ru-Pan Wang
Nils Huse

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Sandra Beauvarlet
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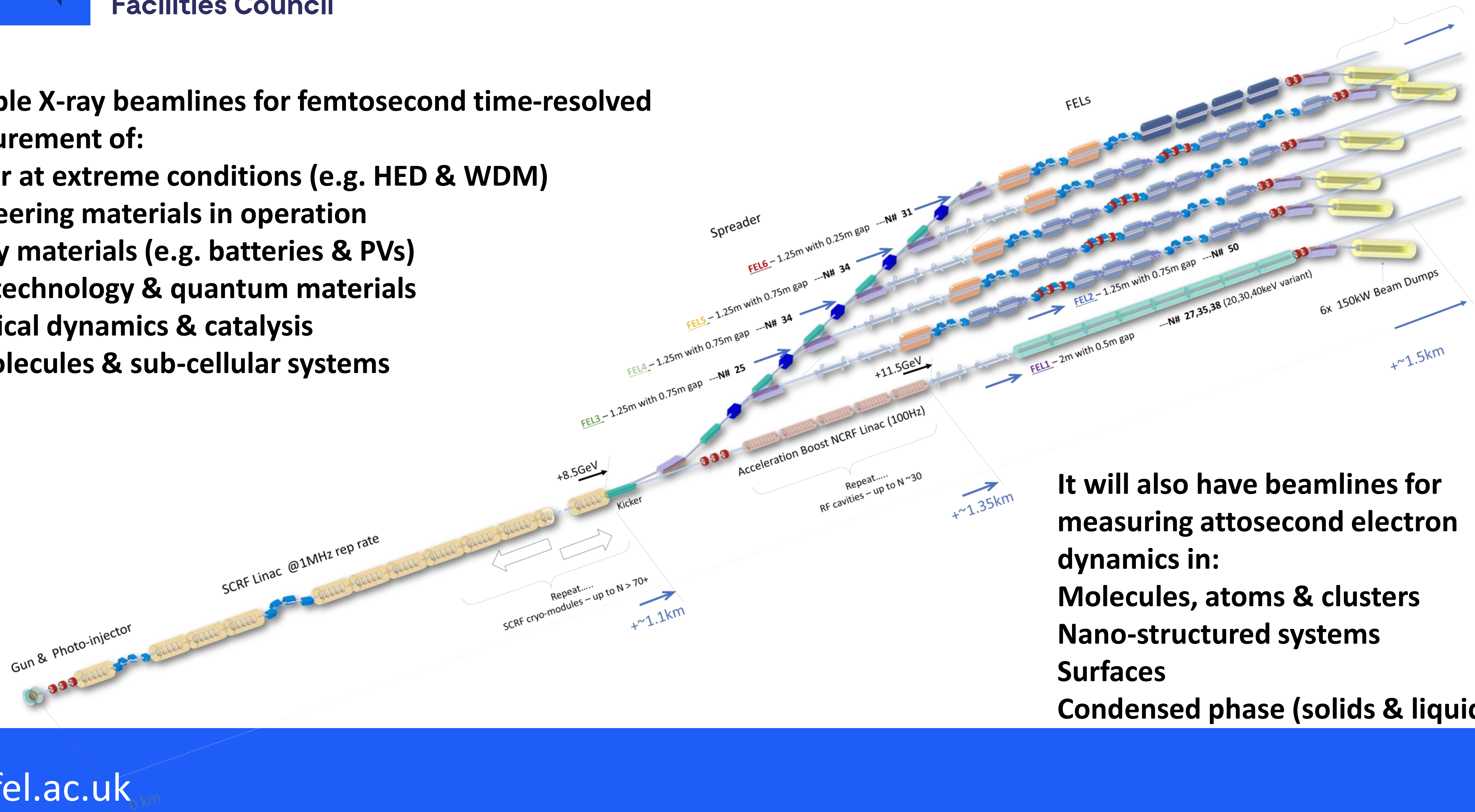
Science and Technology Facilities Council

UK XFEL - Addressing Future Research Needs

Multiple X-ray beamlines for femtosecond time-resolved measurement of:
Matter at extreme conditions (e.g. HED & WDM)
Engineering materials in operation
Energy materials (e.g. batteries & PVs)
Nanotechnology & quantum materials
Chemical dynamics & catalysis
Biomolecules & sub-cellular systems



It will also have beamlines for measuring attosecond electron dynamics in:
Molecules, atoms & clusters
Nano-structured systems
Surfaces
Condensed phase (solids & liquids)