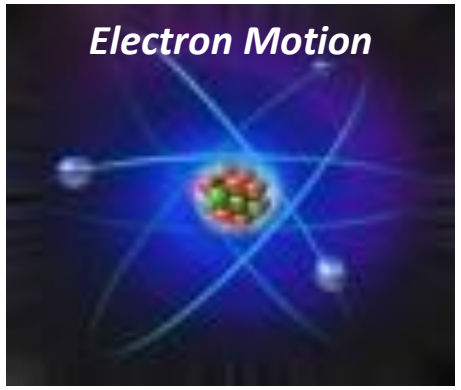
Attosecond Science

Jon Marangos, Director Extreme Light Consortium,
Imperial College London

Electron Orbit in Bohr Model

$T_{orbit} \approx 150$ *as* for H ground state



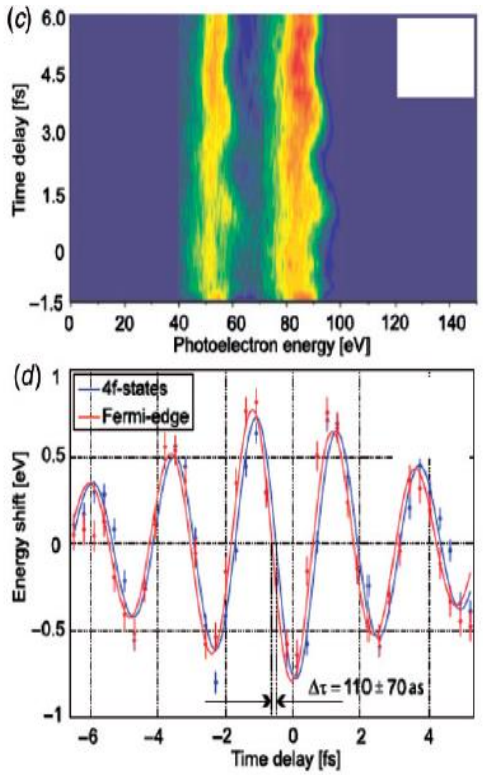
In most matter electrons are in close proximity to one another and so both classical and quantum correlation will play a vital role in the electron dynamics

Attosecond Science = study and ultimately control of attosecond time-scale electron dynamics in matter.

These dynamics determine how physical and chemical changes occur at a fundamental level.

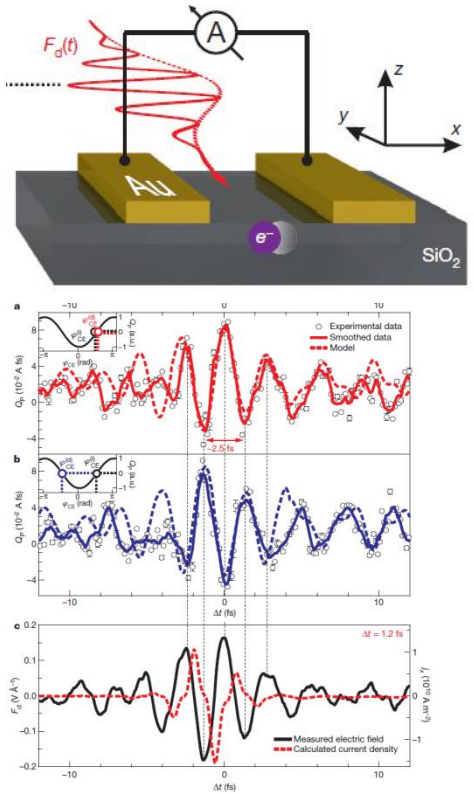
Attosecond Domain Electron Dynamics of Chemical and Physical Systems

Controlling photoemission from a surface



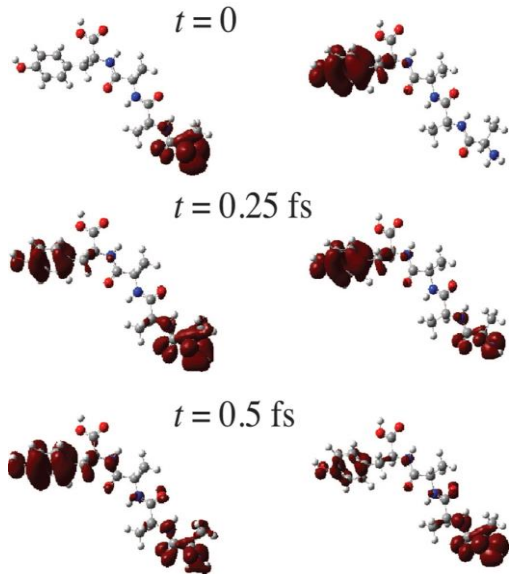
A. Cavalieri et al. Nature 2007, 449, 1029

Light wave electronics – controlling photocurrents with attosecond precision



A.Schiffirin et al Nature, (2012)
Schultze et al Nature (2012)

Measurement of attosecond charge migration in molecules – tracking from the electronic to nuclear motion



e.g. calculated charge migration in the peptide Trp-Ala-Ala-Ala

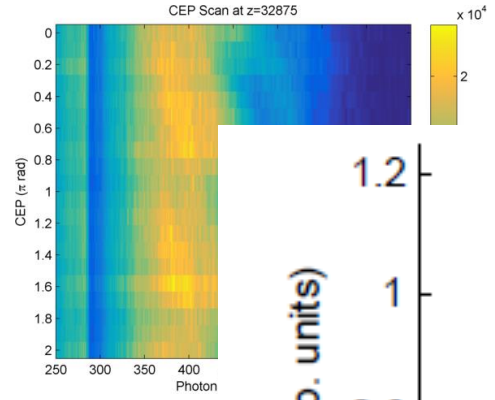
Remacle & Levine, PNAS 103 6793 (2005) & Cederbaum et al *Chem. Phys. Lett.* **307** 205 (1999)
Calegari et al, Science (2014)
Vacher et al, PRL (2017)

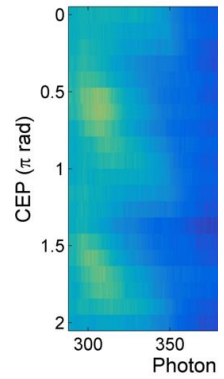
Some important problems at ultrafast timescales

- Understanding light harvesting systems (photosynthesis) (1fs – 1ps)
- Controlling chemistry with laser fields (0.1 – 1000fs)
- Quantum control of physical processes (0.1-1000fs)
- Light-wave electronics (0.01-1 fs)
- Fundamental understanding of photo-catalysis, catalysis and enzymes (1fs – 100ps)
- Controlling superconductivity with light (1fs – 100ps)
- Understanding radiation damage in DNA (0.1fs – 1ps)

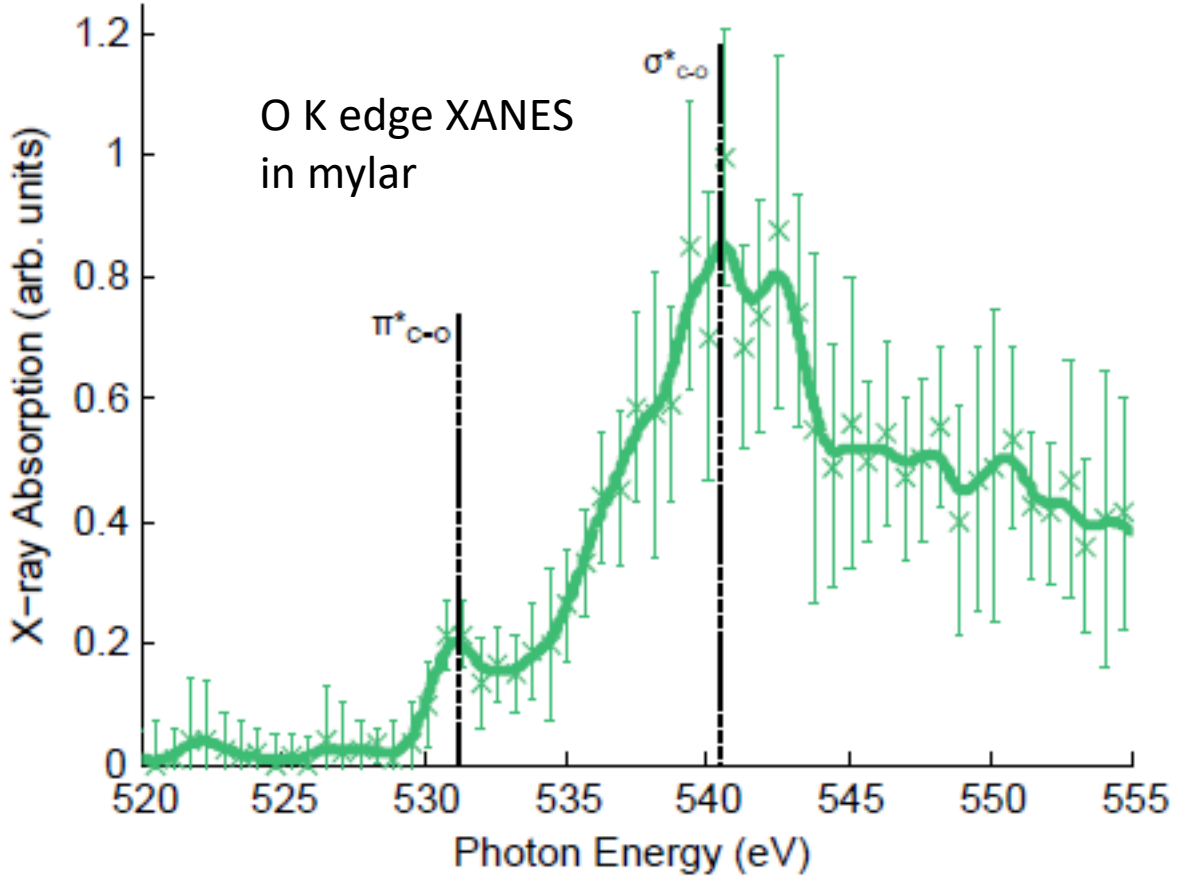
We must measure across a wide range of timescales from nanosecond (1 ns = 10^{-9}s) & picosecond (1 ps =10^{-12}s) $\rightarrow$ femtoseconds (1 fs =10^{-15}s) $\rightarrow$ attoseconds (1 as =10^{-18}s) to get a full picture of the dynamics of these complex systems.

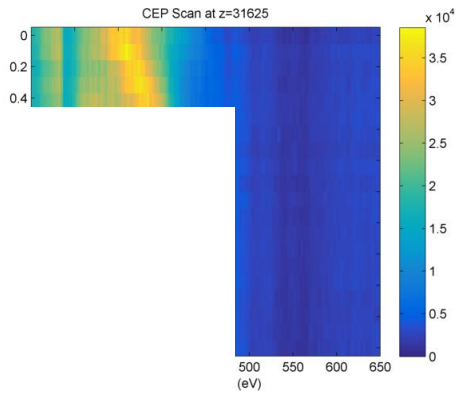
Tunable attosecond pulses across the water window

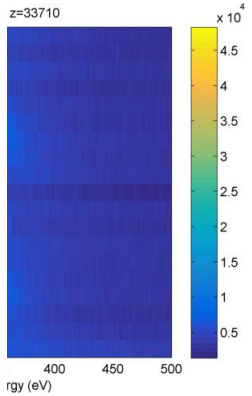




By changing gas species, tuning gas pressure, and





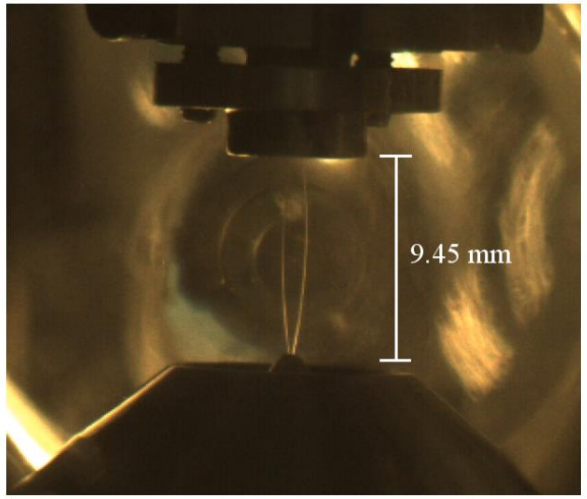


A.S.Johnson et al Science Advances, 4, 3761 (2018)

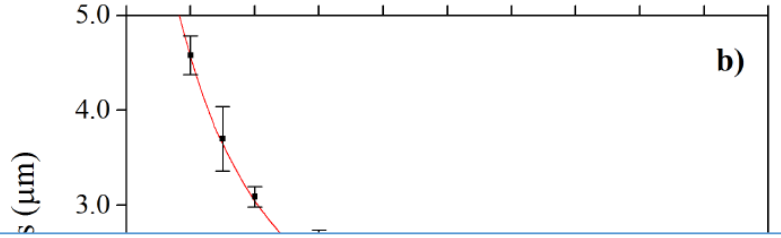
Ref.	Ga				lux
This work	N				4) × 10^9 photons/s
This work	H				.4) × 10^8 photons/s
[4]	N				.1) × 10^7 photons/s
[4]	He	-	-	-	0.9 ± 0.2 pJ (1 kHz)
[5]	Ne	-	-	-	> 1 × 10^8 photons/s
[6]	He	-	-	-	6 × 10^7 photons/s

Measured flux is 10x greater than previously achieved in the water window with few-cycle pulses.

Liquid phase time resolved SXR absorption spectroscopy

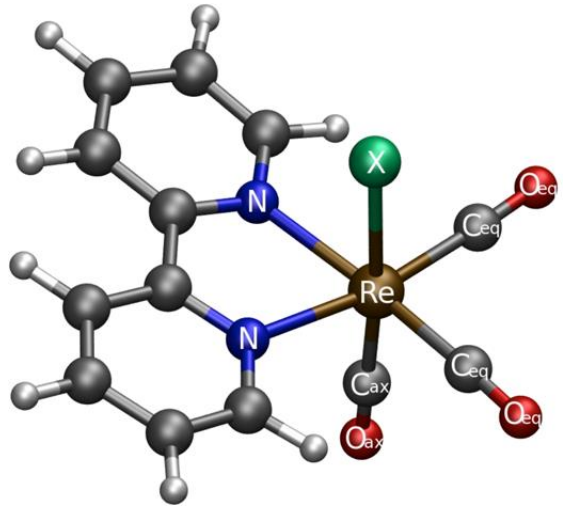


Sheets stable (isopropanol in air or vacuum) of thickness ~ 1.0 µm using shaped nozzles we have developed



Time-resolved X-ray spectroscopy:

Objective is to track electronic and structural dynamics in photochemical reactions (e.g. electronic transfer following excitation of metal-ligand complexes)



White light interferometry demonstrates thickness of 1.2 micron in a stable and uniform flow

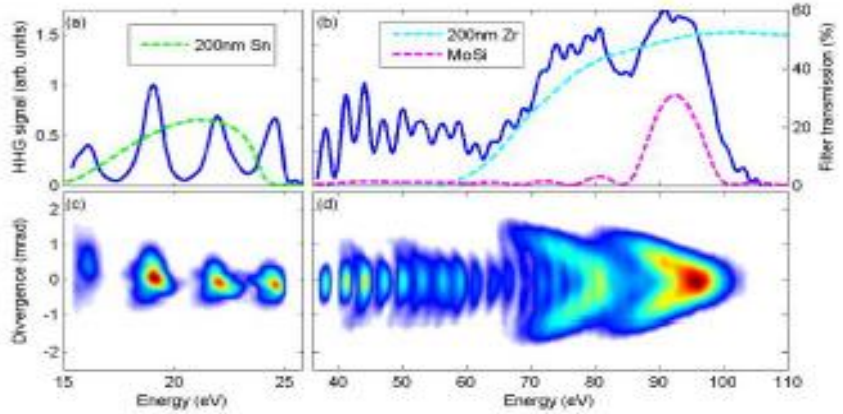
G.Galinas et al

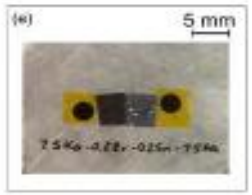
published Reviews of Scientific Instruments August 30th 2017

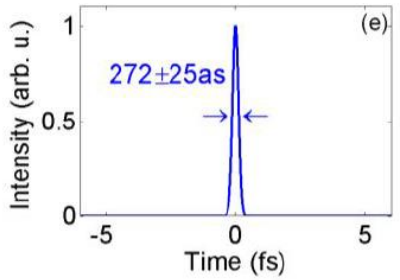
RSI, 88, 083117 (2017)

Studies at low concentrations or low pump fluence of relevance to biochemistry & light harvesting technology will require high rep-rate, high power lasers such as those at ELI-ALP

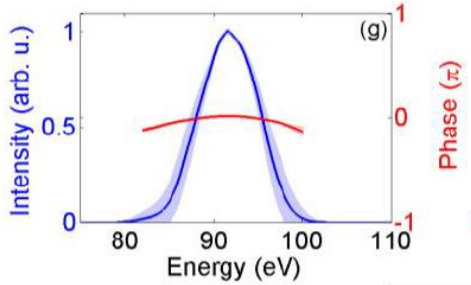
Simultaneous generation of single sub-femtosecond HHG XUV and VUV pulses retrieved by streaking measurements

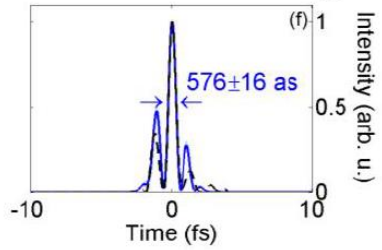




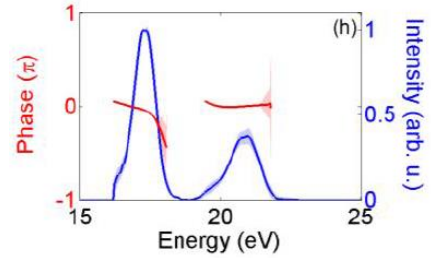


90 eV
"XUV"



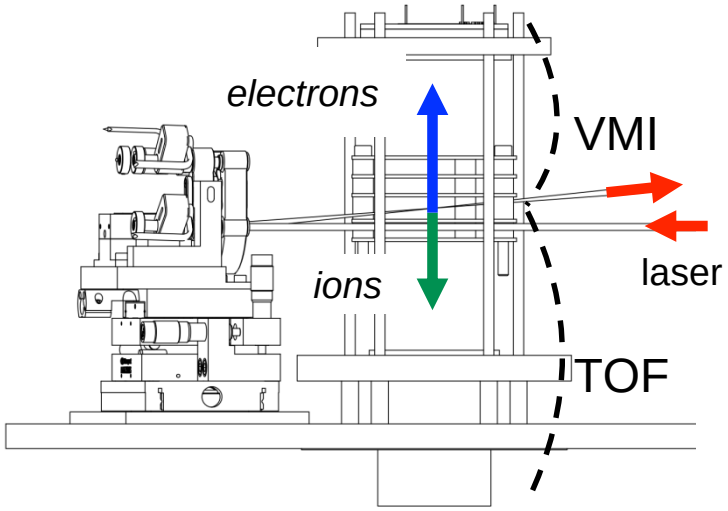


20 eV
"VUV"

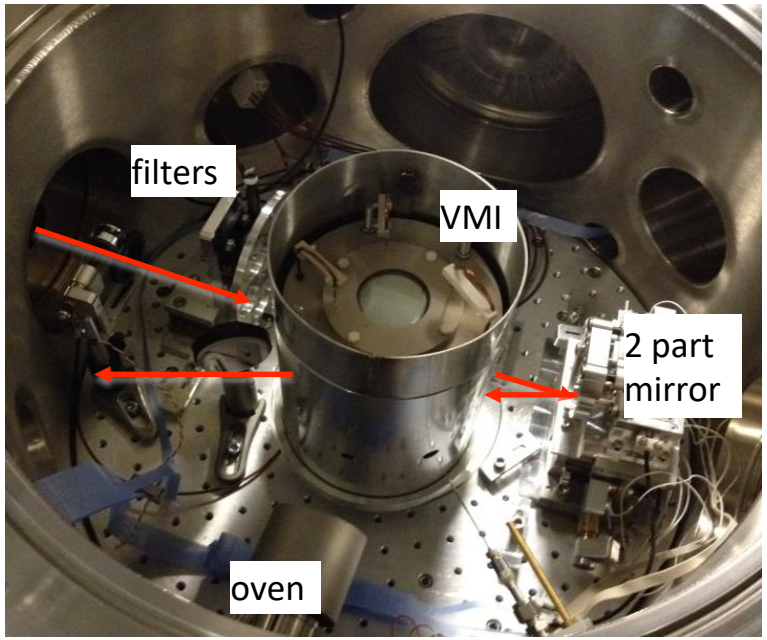


D. Fabris et al Nature Photonics , 9, 383 (2015)

Toward pump-probe measurements of organic molecule photoinduced dynamics

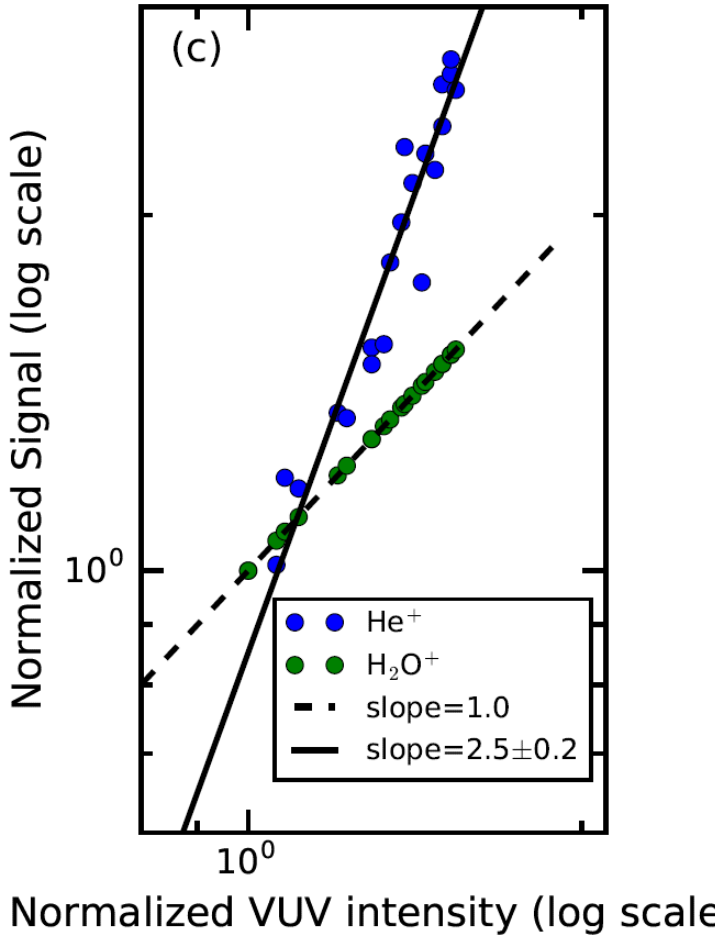


Simultaneous measurement of electrons (electron VMI) and ions (ion TOF) at 1kHz rate.



Sub-fs VUV at 21 eV only 0.1 nJ pulses but still intense enough to measure two-photon processes – a necessary condition for attosecond pump-attosecond probe

T.Barillot et al, CPL (2017) in press



With high power, high rep-rate lasers at ELI-ALP true attosecond pump -attosecond probe experiments are enabled

Attosecond Time Domain Science at ELI-ALP

- Important and wide ranging scientific problems associated with measuring and controlling ultrafast electronic and electronic-nuclear coupled dynamics
- UK research labs can tackle some of these challenges – but lack a high rep-rate, high flux source of attosecond pulses
- ELI-ALP offers a suite of highly appropriate lasers and attosecond beamlines that will be “the best in the world” capability for the next decade or more