Attosecond science with X-ray FELs

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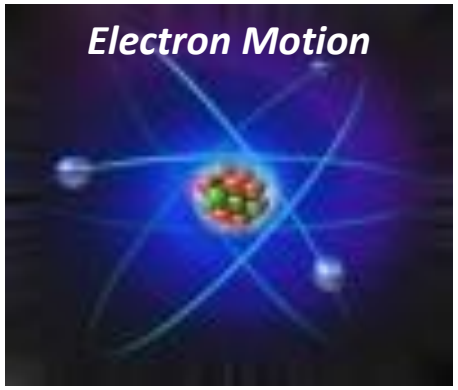
1. Motivation for attosecond science
2. Current capability
3. Future trends
4. Requirements

1. What is attoscience?

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.

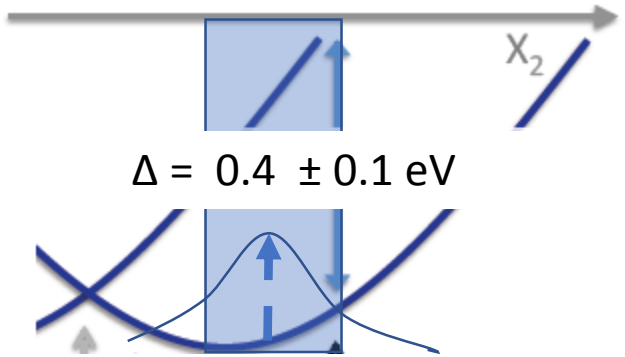
1. Attosecond science is not only about the electrons: Example of dephasing/damping of electronic coherence in charge migration

Rapid nuclear motion –can change evolution of amplitudes in a few fs

see Vacher et al *J Chem Phys*, 139, 044110 (2013)

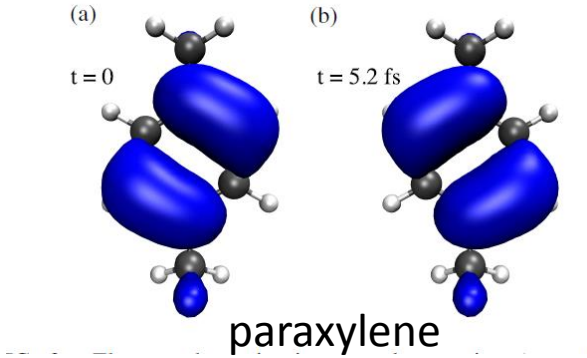
Initial quantum nuclear distribution also couples to a significant spread in electronic timescales and consequently dephasing

Vacher et al PRA 92, 040502(R) (2015)

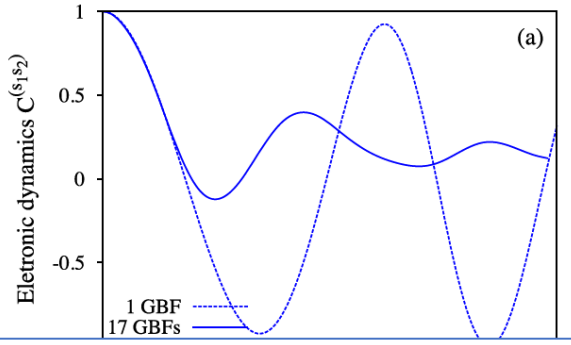


Superposition of cation states

Initial nuclear wavefunction

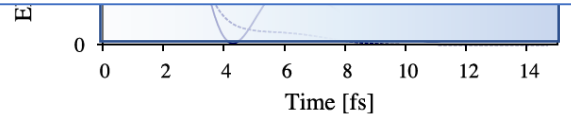


paraxylene

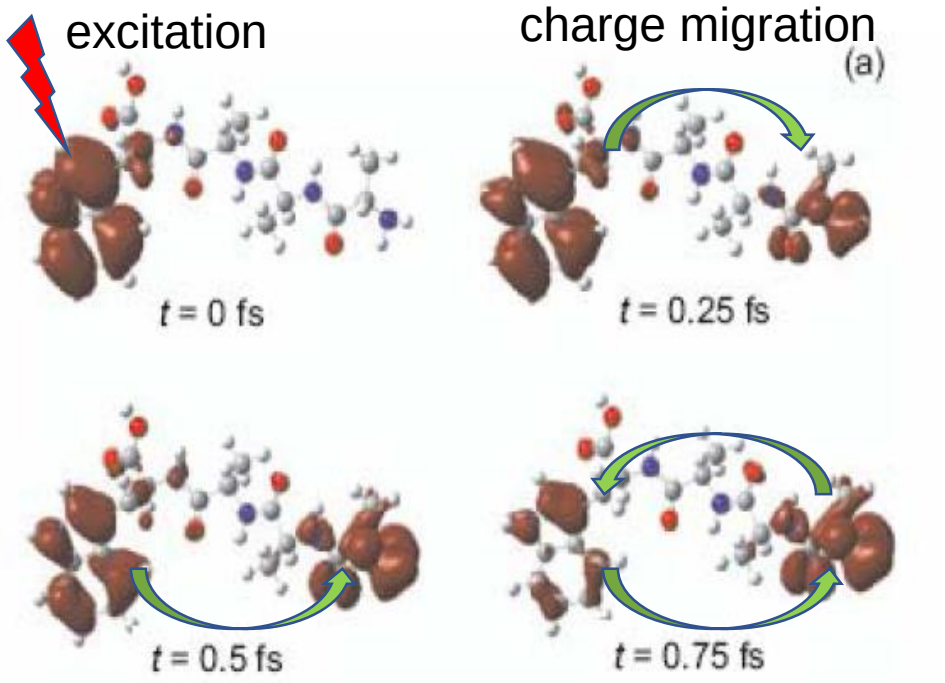


In general we must anticipate correlated electronic-nuclear state evolution with ultrafast vibronic coupling and non-adiabatic dynamics as well as strong traces of decoherence/dephasing at these early timescales

118, 083001
7)



1. Electronic motion in a molecule



Calculated electron dynamics in a dipeptide

Electron Orbit in Bohr Model

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

In a chemical reaction or physical change electronic, vibrational & rotational dynamics can occur in a highly correlated fashion on timescales from 0.01 – 1000 fs

1. Some important problems at ultrafast timescales

- Optimising artificial light harvesting systems (1 fs – 1 ns)
- Electronic events in photo-physics and chemistry
- Controlling chemistry and physics with laser fields (0.1 fs – 100 ps)

- Lightwave electronics (0.01-1 fs)

- Controlling materials (e.g. superconductivity) with light (1fs – 100ps)

- Understanding radiation damage in biomolecules (0.1fs – 1ps)

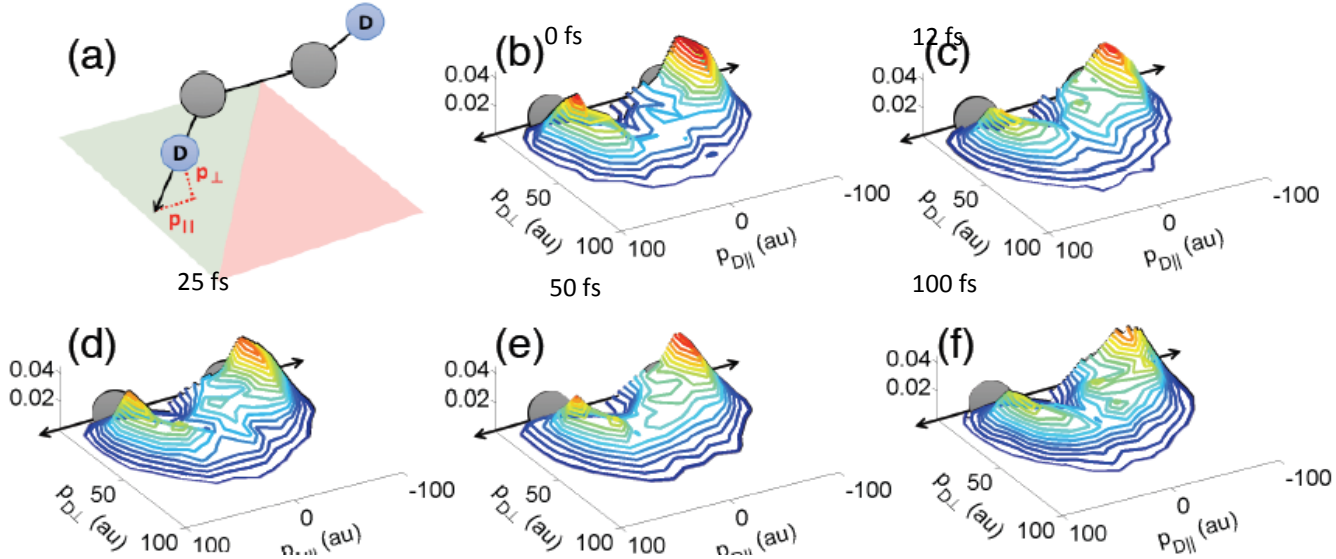
We must measure across a wide range of timescales from nanoseconds (1 ns = 10^{-9}s) picosecond (1 ps =10^{-12}s) $\rightarrow$ femtoseconds (1 fs =10^{-15}s) $\rightarrow$ attoseconds (1 as =10^{-18}s)

The fastest timescales are only now be accessed by ultrafast measurement technology

2. Approaching Few- to Sub-Femtosecond Time Resolution

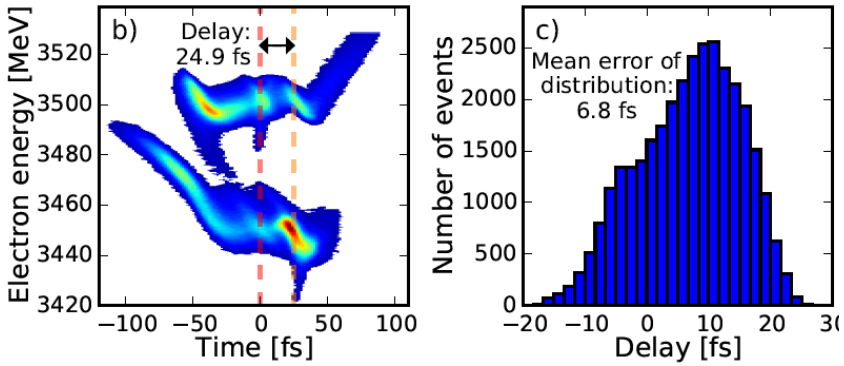
Imaging isomerization acetylene – vinyldiene using particle coincidences with 10 fs resolution at LCLS AMO end station using X-ray split and delay

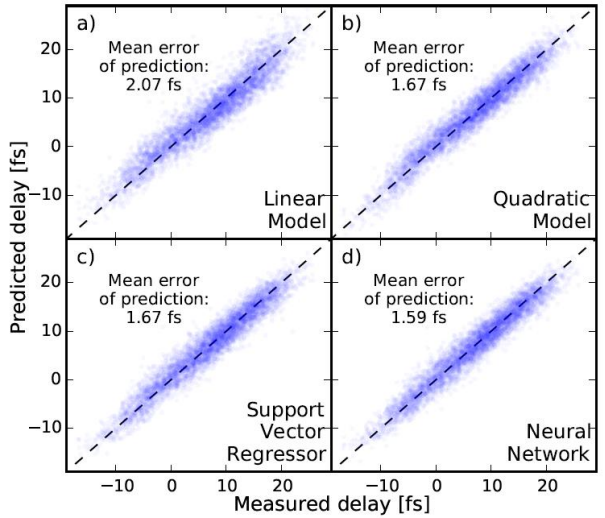
C.E.Liekhus-Schmaltz et al Nature Communications, 6, 8199 (2015)



Optimisation of time-resolved XFEL experiments using a machine learning approach to predict pulse parameters

A.Sanchez-Gonzalez et al, Nature Communications 10.1038/NCOMMS15461 (2017)





2. Probing valence hole dynamics

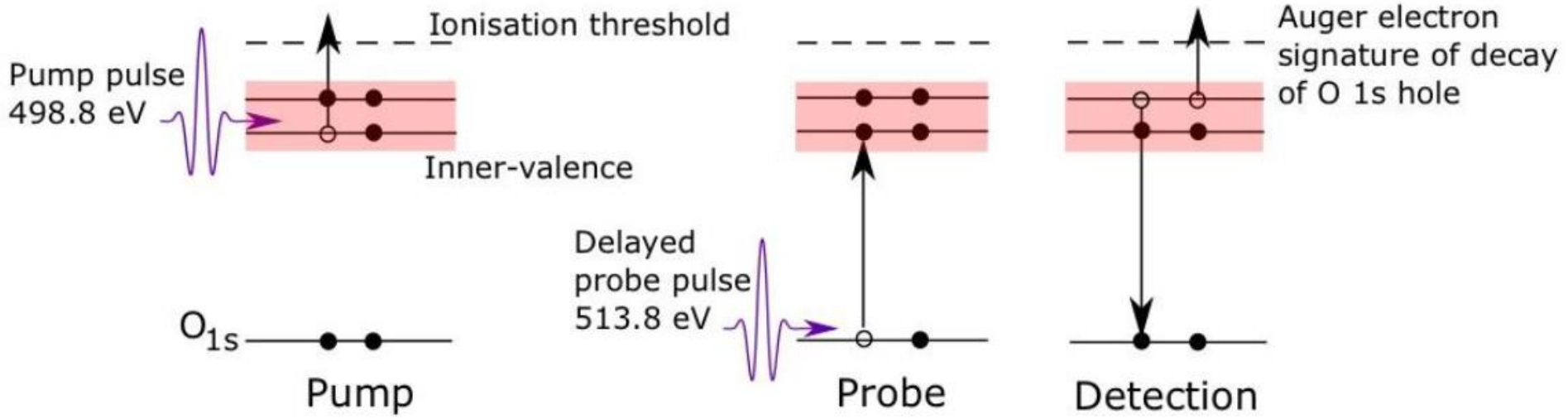


Figure 2: *Pump step opens a number of valence ionisation channels including creation of inner valence holes (IVH). The delayed probe pulse can strongly interact via a O 1s–IVH transition (a channel only open if inner valence ionisation has occurred). Following this there can be Auger decay back to the O 1s hole with the emission of Auger electrons of characteristic energy that can be detected.*

ARTICLES NATURE PHOTONICS DOI: 10.1038/NPHOTON.2016.201

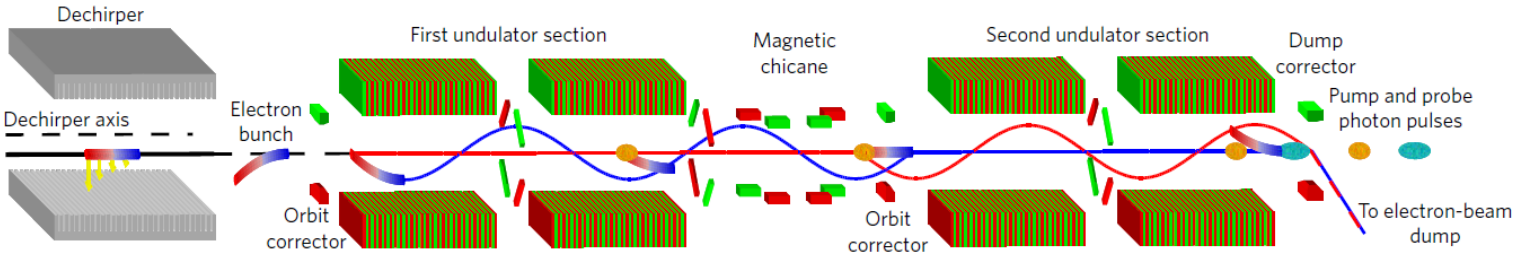
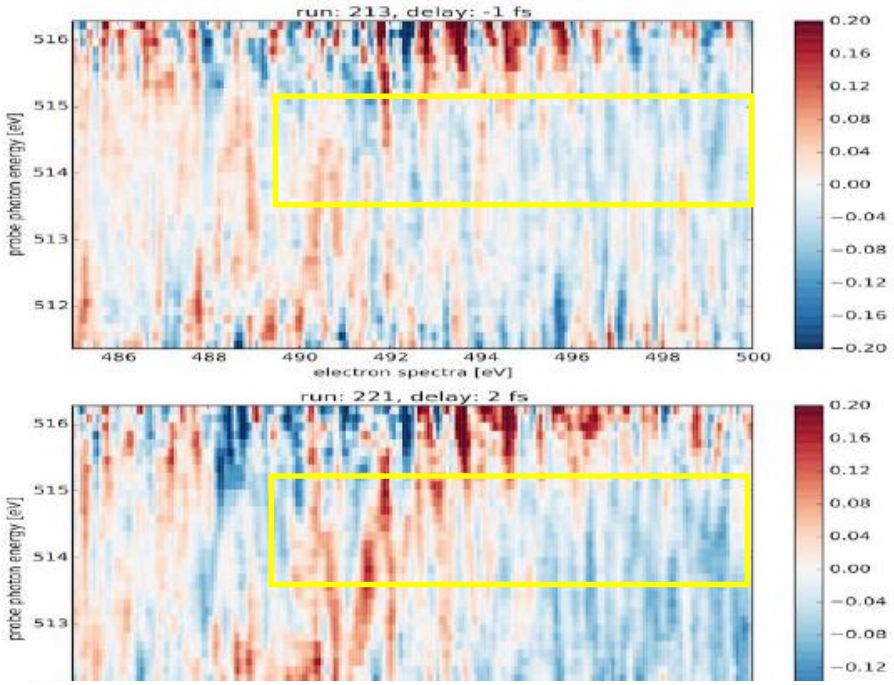
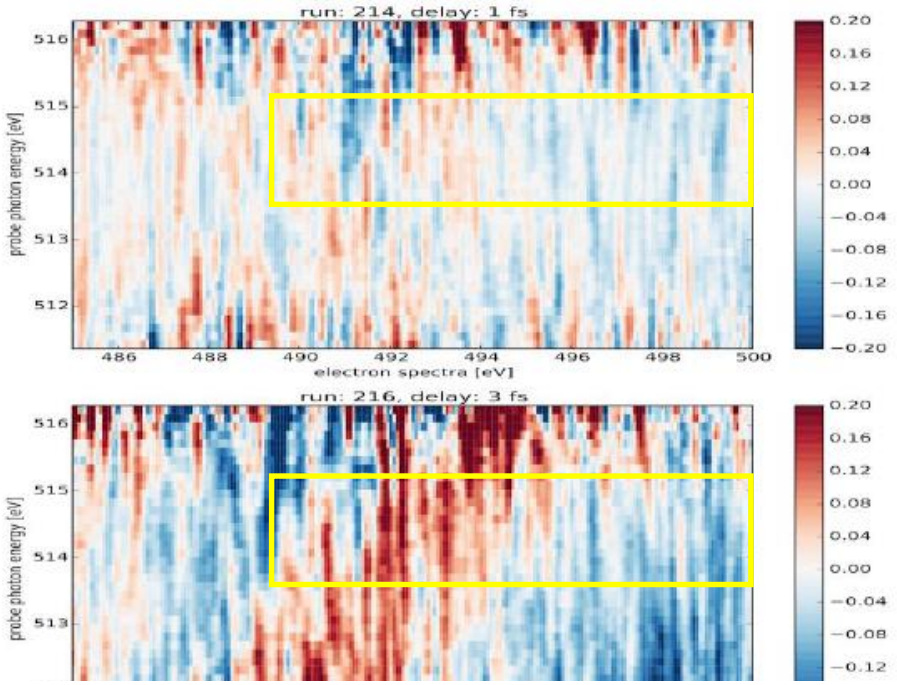


Figure 1 | Fresh-slice multipulse scheme. The electron bunch travels off-axis in the dechirper experiencing a strong transverse head–tail kick, represented by

"Fresh slice mode" was used to generate ~ 5 fs pump and probe pulses at the required photon energy with delay variable from -10 to +25 fs

2. Possible signature of valence hole dynamics





Photon energy v Electron spectrum at delays from (nominal)
t = -1 fs
to t = + 7 fs

Region of interest probe ~514 eV giving characteristic Auger spectrum

Analysis continues to

Currently a lot of fluctuation of pulse spectrum, intensity, temporal profile and delays between pulses, together with the only 120 Hz repetition rate, this makes getting statistically meaningful signals challenging –

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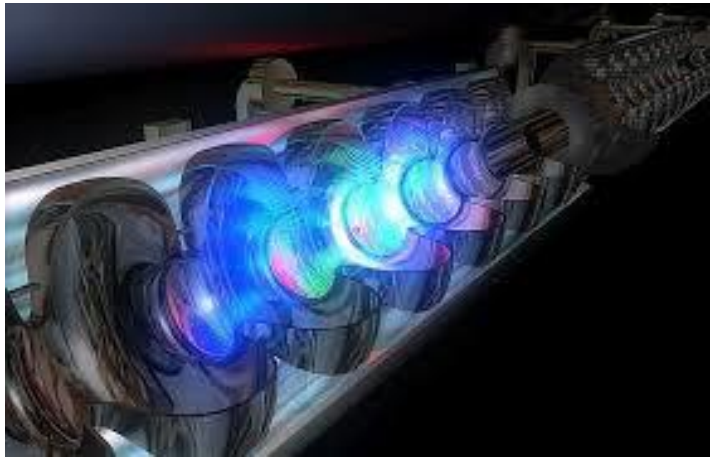
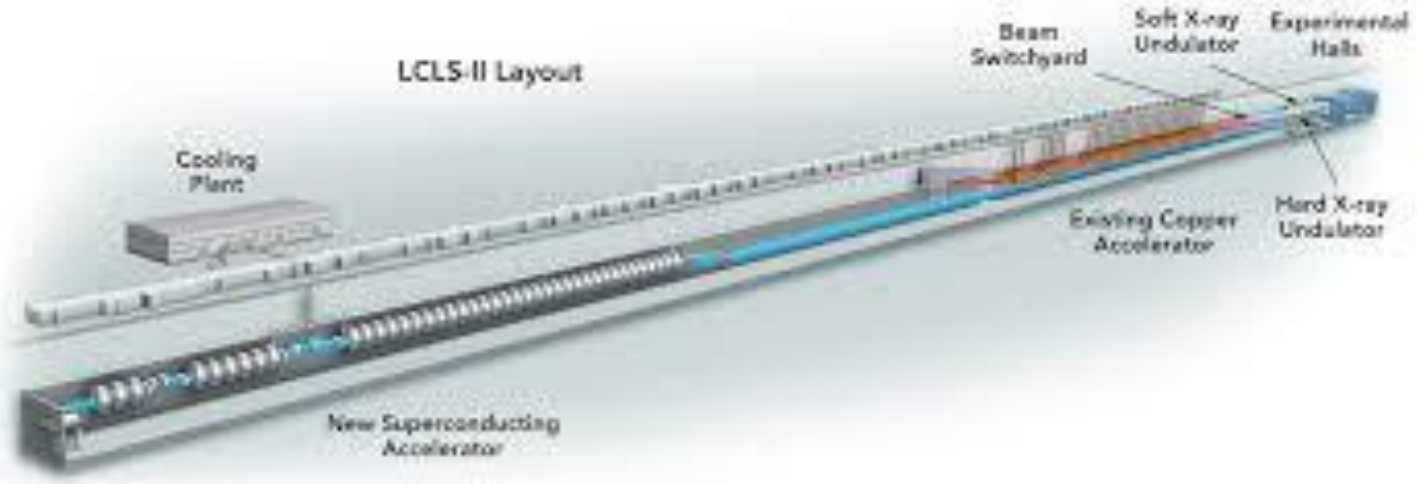
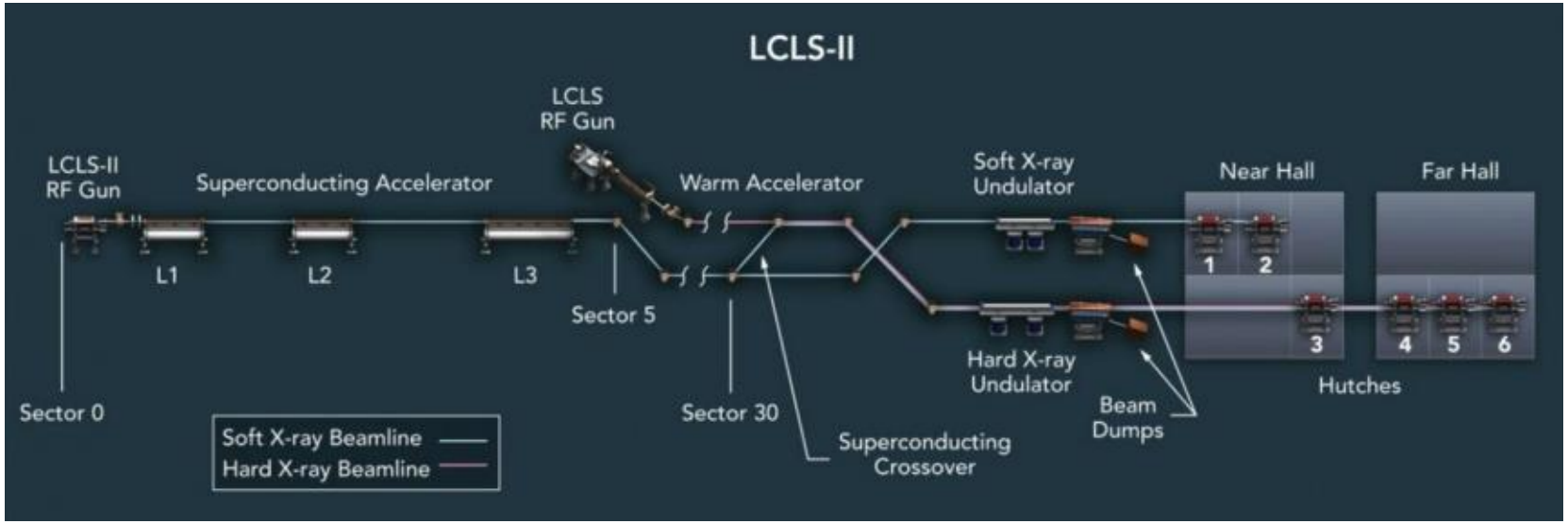
t against
AV

WE NEED A BETTER MACHINE !

3. Euro XFEL and LCLS II



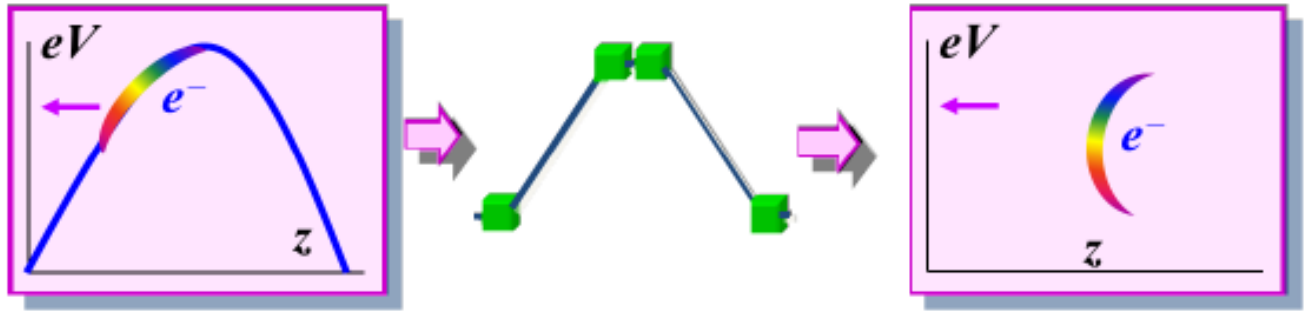




High repetition rate machines with up to 1 MHz rep-rate (10,000 higher) & potentially much less fluctuation

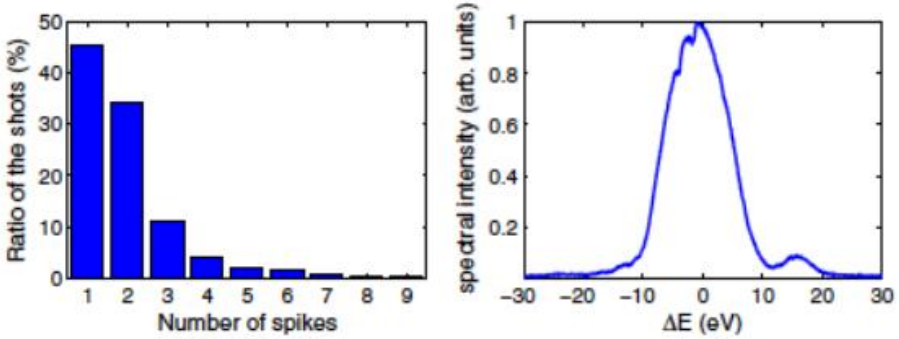
3 . Attosecond X-ray Pulses

HXR: Isolated 200 as pulse produced



Nonlinear compression produces
High density head with low density tail

Huang et al, PRL, 119, 154801 (2017)

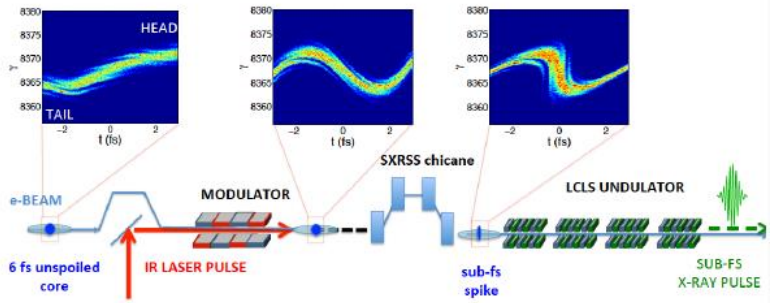


Huang et al, PRL 119, 154801 (2017)

Measurements:
179 ± 58 as @ 9 keV (14.4 eV BW)
228 ± 85 as @ 5.6 keV (11.3 eV BW)
10 uJ x-ray pulse from 20 pC bunch

Slotted foil results (400 as pulse): APL 111, 151101 (2017)

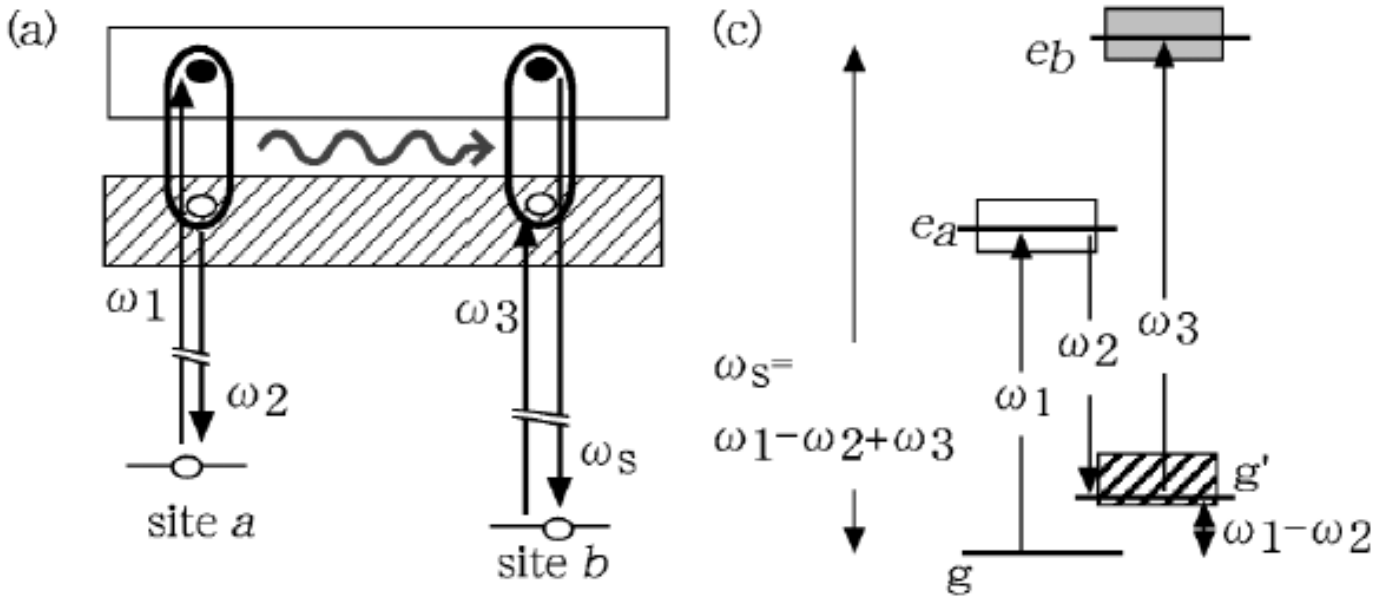
XLEAP - soft X-ray tests underway

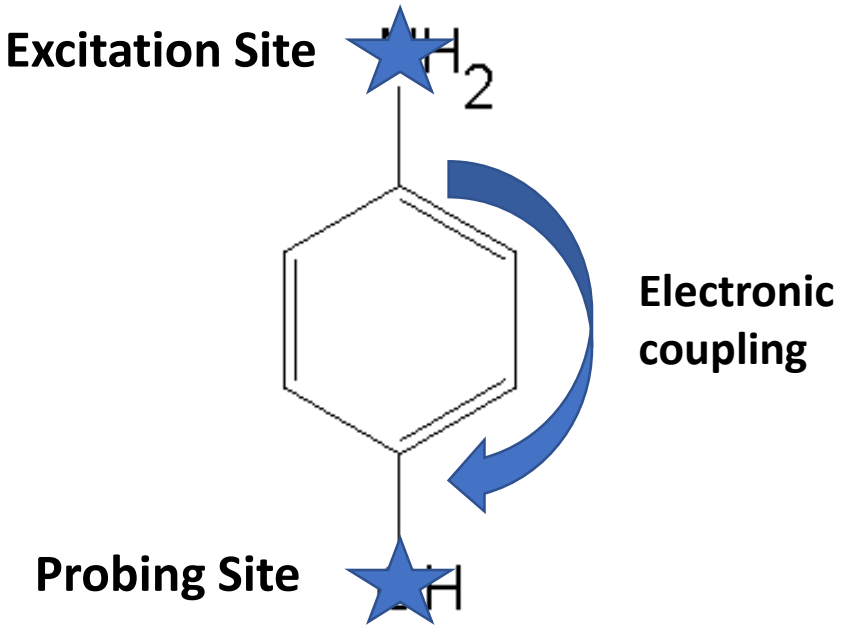


XLEAP Soft X-ray scheme now operating reliably.

1st sub-fs resolved circular field streaking measurements conducted in the last weeks

3. Attosecond pulses enable X-ray non-linear spectroscopy : Measuring electronic coupling between sites within a molecule





I.V. Schweigert and S. Mukamel, *PRL* **99**, 163001 (2007)

One of many potential methods that could lead to multi-dimensional time-resolved X-ray spectroscopy for tracking electronic dynamics within matter

4. X-ray requirements for attosecond science

- Sub- fs pulses (width < 0.5 fs)
- 2-colour (spanning multiple X-ray absorption edges) sub-fs pulses of variable delay (-100 to +100 fs) with delay accuracy to 0.5 fs
- SXR 50 eV – 1 keV, (eventually to 10 keV for accessibility to wide range of L and K edges)
- Accurate single-shot pulse diagnostics (pulse energy, photon spectrum, duration and delay)
- Synchronisation to external lasers (or post-sorting) to < 1 fs (for optical excitation and coherent control experiments)
- High peak and average flux (10 - 100 µJ pulses, >1 W average)
- High rep-rate (for statistics and to avoid severe sample damage or detector saturation limits) > 10 kHz
- Small X-ray beam focus (< 1 µm)
- Gas, liquid and solid sample environments operation in vacuuo