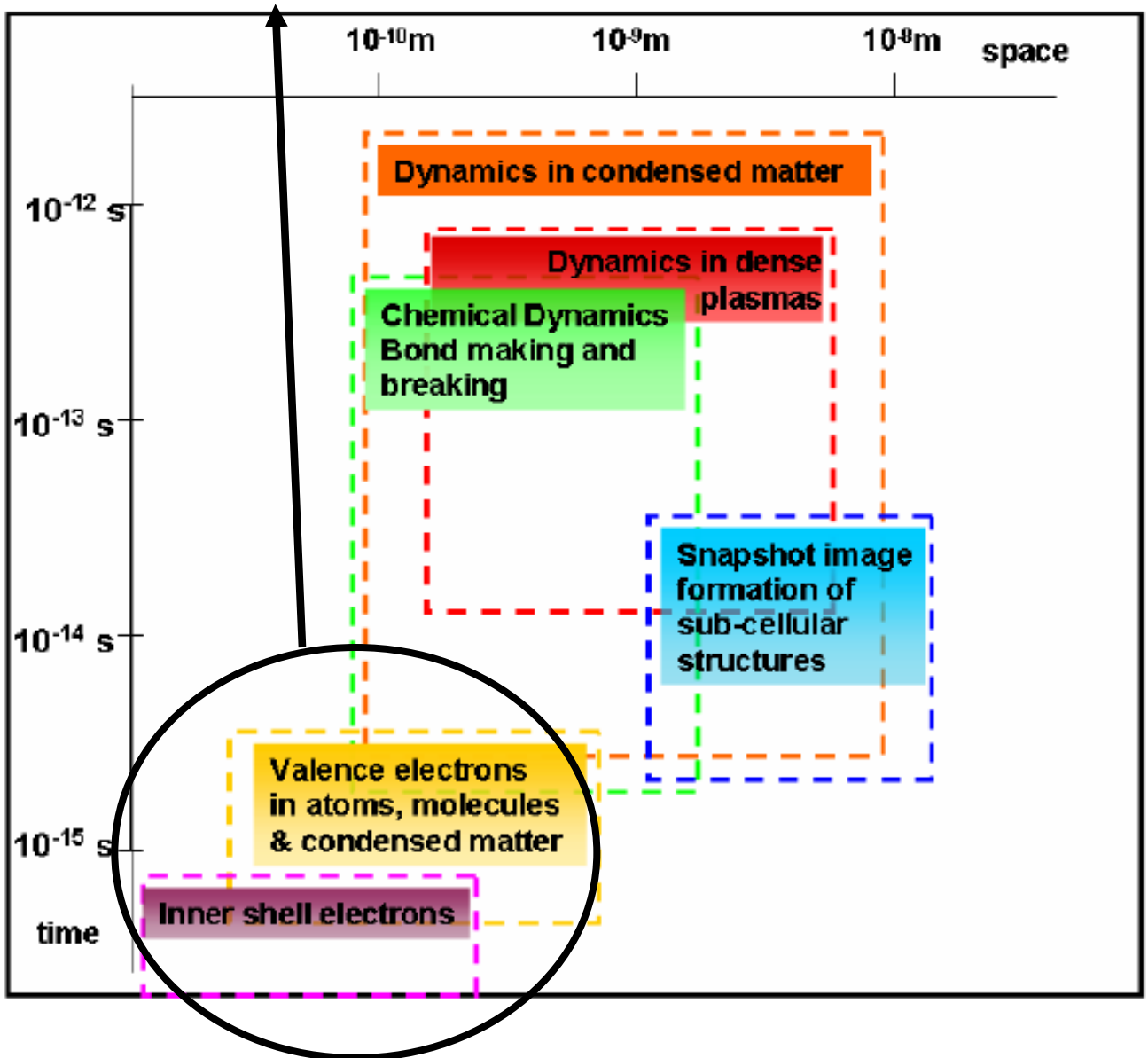
Ultrafast Electron Dynamics And Attosecond Science



Two topics of importance will be briefly covered here:

- Attosecond timescale dynamics in multi-electron systems.

  How will FELs have an impact?

- Ultra-fast electron dynamics driven by intense soft-X-ray fields.

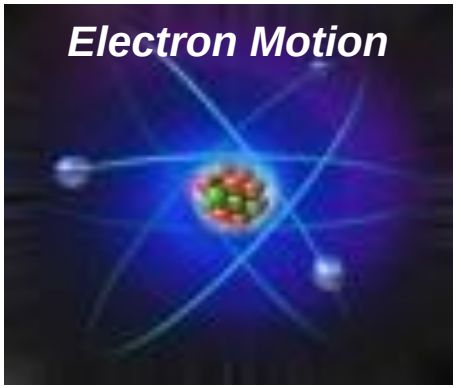
  How can we capture what is going on and build on this for new classes of measurement?

Why do we need to do attoscience?

Electron Orbits in Bohr Model

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

1 as = 10^{-18} sec



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.

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

Why Study Multi-Electron Correlated Dynamics

Fast changes (chemical reactions, phase changes etc) require real time information to give a complete understanding at the electronic level

Formation of band structure in real time, understanding this and the formation of other electronic properties will provide profound new insights in material science

Quantum control – a frontier of 21st century technology - concerns optical manipulation of electrons. For complex matter this will require an understanding and control over multi-electron correlations

Ultrafast Science relies on ultrashort light pulses

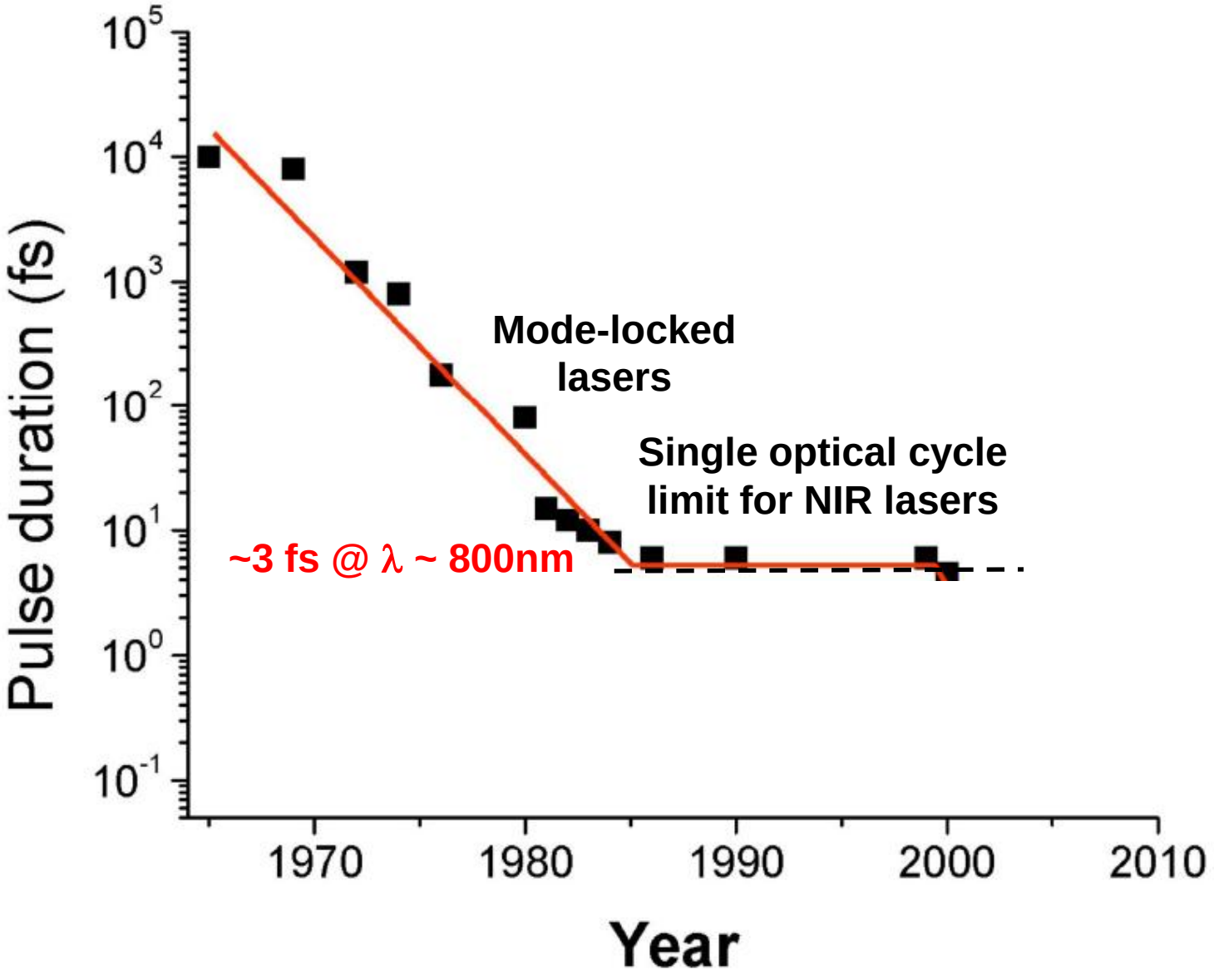
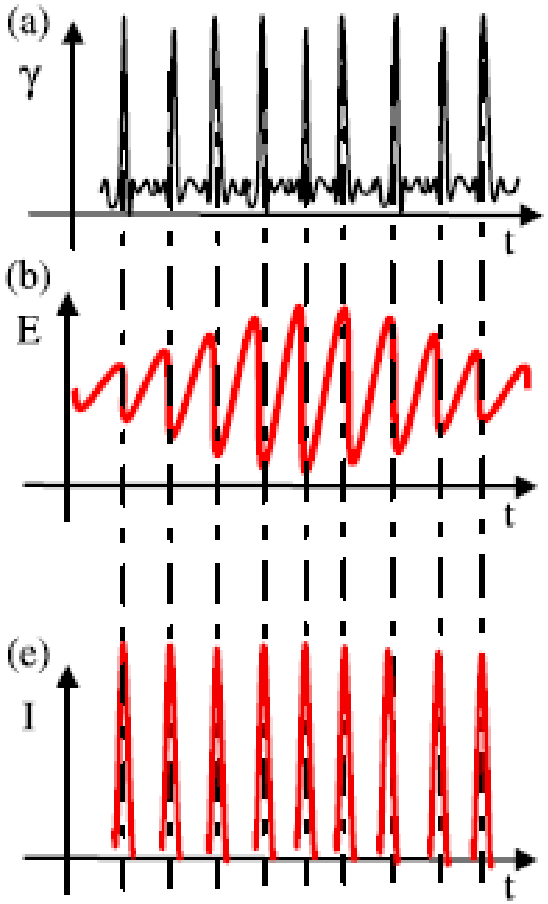


Fig. adapted from Corkum & Krausz Nature Physics **3,** 381 (2007)

Routes to brighter attosecond probes

- **RELATIVISTIC HHG**

  Dromey et al. Nature Physics **2** 456 (2006)



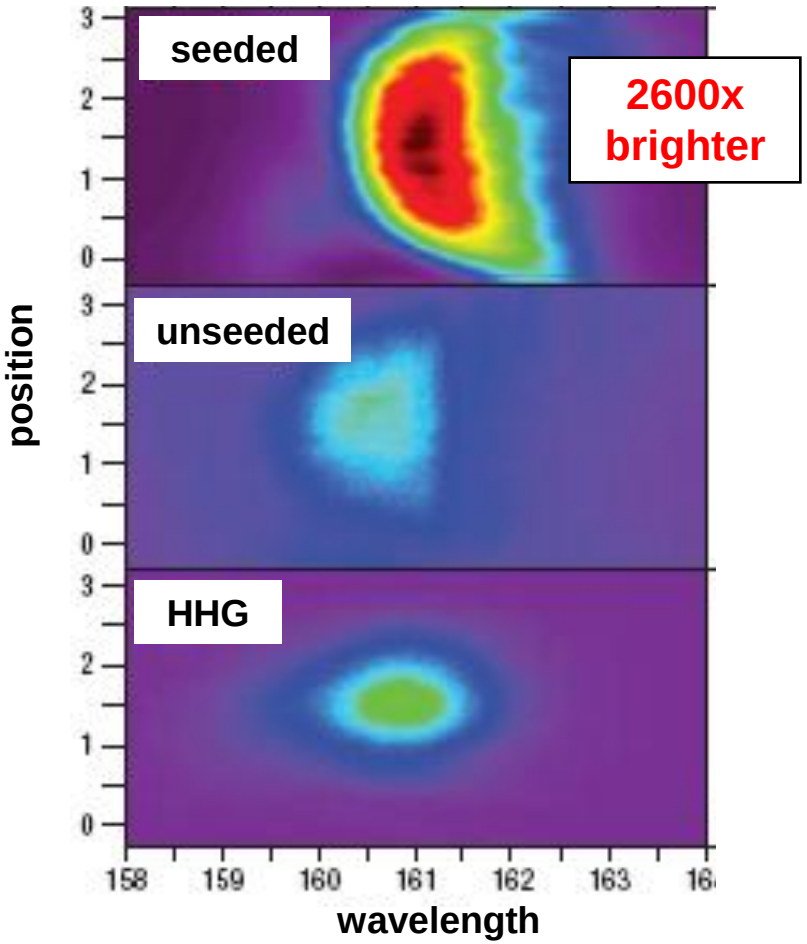
Zepf et al. Plasma Phys. & Controlled Fusion **49** B149 (2007)

- **XFELs**
  - **Schemes for attosec pulses**

    Zholents & Fawley PRL **92** 224801 (2004)
  - **HHG seeding**

    Lambert et al Nature Physics **4** 296 (2008)

    (SPRING8)



Unique Capabilities of NLS For Attosecond Science

- Access to inner shell states from 10eV-5keV (HHG 10-100eV)
- >10,000 times brighter than existing sources even at <100eV (where HHG is available), so hugely extends the range of measurements
- Non-linear excitations will become accessible by photons up to 1keV
- Challenge is to get FEL pulses making measurements in attosecond domain:

  (a) attosecond pulses from FEL, (b) femtosecond seeded FEL pulses phase locked to auxillary laser

Attosecond Temporal Resolution of Correlated Multi-Electron Processes with Femtosecond X-ray Pulse

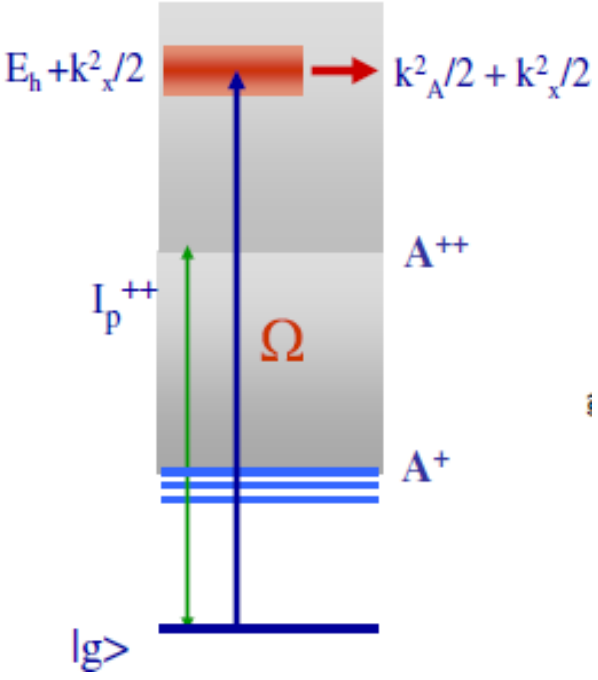
Correlated electron release in presence of IR dressing field, interference between PE sidebands allows retrieval of spectral phase of entangled electrons.

Auxillary Laser Field Allows Full Retrieval of 2-electron Amplitude and Phase

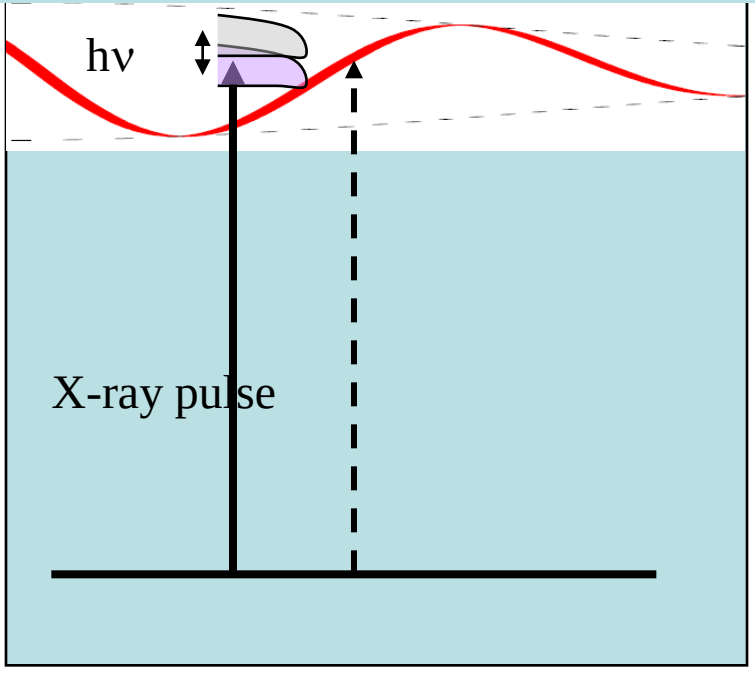
PRL 94, 213001 (2005) PHYSICAL REVIEW LETTERS

Use of Electron Correlation to Make Attosecond Measurements without Attosecond Pu

Olga Smirnova,[1] Vladislav S. Yakovlev,[2] and Misha Ivanov[3]



X-ray absorbtion leading to emission of two correlated electrons of momenta k_x (direct) and k_A (Auger)



10fs X-ray pulse + tightly phase synchronised 5μm field can give <100as temporal resolution

Revealing Electron Dynamics into Attosecond Domain

Attosecond electron dynamics are amenable to study through the interaction with bright short wavelength fields.

-Probing of hole dynamics in atoms, molecules and condensed matter in real time

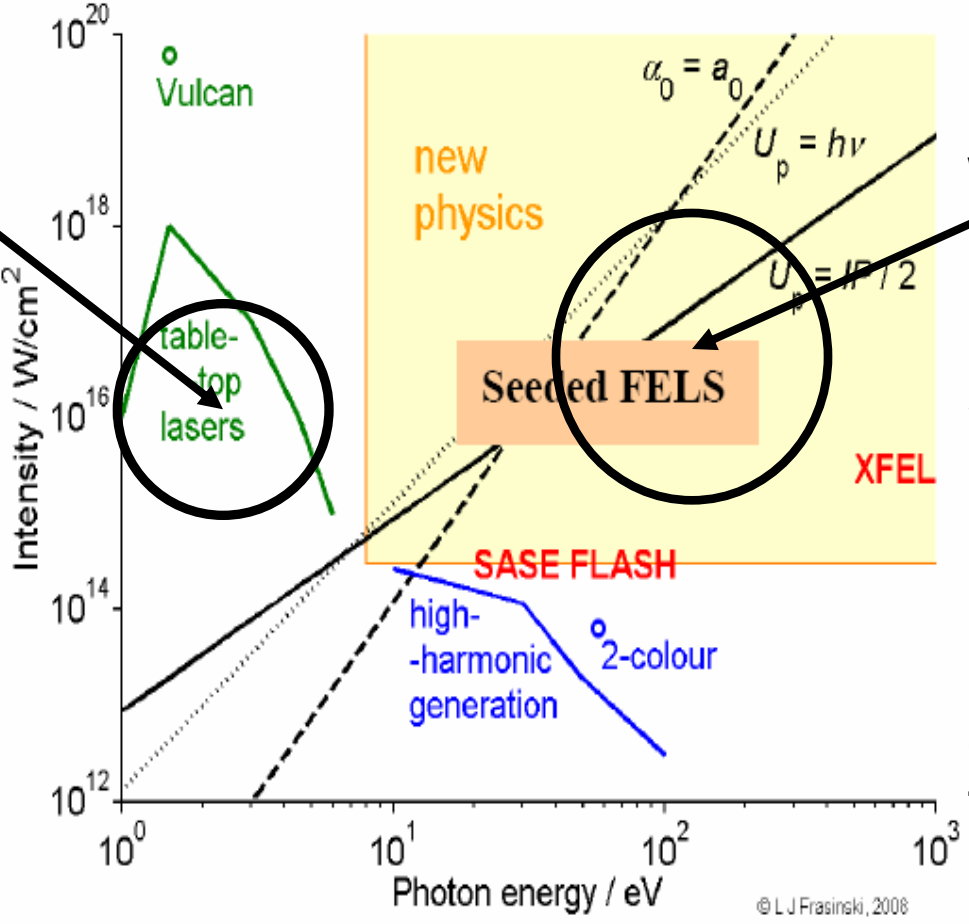
- Time-space resolved studies of nanoscale electron dynamics, e.g. in nanoplasmonic structures

-Real time probing of coherently driven processes for optimised quantum control of matter

ULTRA-FAST DYNAMICS DRIVEN BY INTENSE X-RAYS

A new regime in short pulse high intensity X-ray interaction physics

Strong visible/IR fields “low frequency”:
single electron interaction poorly coupled to inner valence
and core states



Intense soft X-rays, “High frequency” can penetrate and interact with inner valence and core electrons leading to collective effects and strong signatures of electron correlation.

New Regime
-Core electron removal with high probability
-Multiple core hole formation
-2-photon/ 2-electron processes

Strong field physics enters a new regime for high frequency fields. There is special interest in the soft X-ray range (10eV- 1keV) where the fields couple efficiently to multi-electron – multi-photon processes in matter.

Non-linear and High Field X-ray Processes

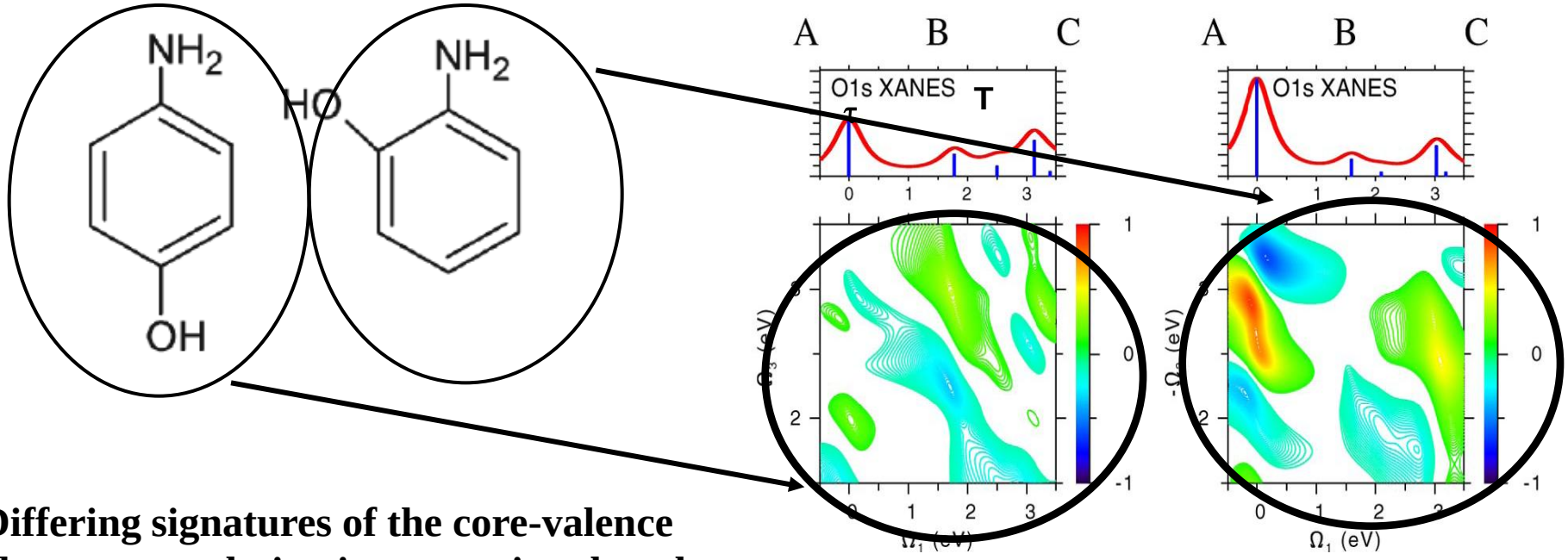
- **Matter in High X-ray Fields: A New Physical Regime for Fragmentation that we Must Understand if New Concepts in Nanoscale Imaging to be Fully Realised**

- **Multi-Photon Processes and Potential Applications for Temporal Measurement of Electronic Reorganization following Core Hole Formation**

- **Non-Linear X-ray Spectroscopy**

New attosecond techniques (e.g. NLO) will be enabled that lead to future science

Schweigert, I.V.and Mukamel, S., *Coherent ultrafast core-hole correlation spectroscopy: X-ray analogues of multi-dimensional NMR,* Phys.Rev.Lett. 2007, 99(6), 163001.



Differing signatures of the core-valence electron correlation in two aminophenol isomers

Essential information about the structurally dependent coupling between the O and N holes will be contained in the cross-peaks
(so far all theory)

Quantitative Non-linear X-ray Science will be Enabled by NLS due to Seeding

- NLS will be a well characterized, well controlled, X-ray source as required for quantitative studies.
- Full spatial and temporal characterization of intensity (full retrieval – going beyond autocorrelation) will be available.
- X-rays will be synchronised to additional laser fields (including HHG XUV fields).
- Sub-fs pulse duration may be available from pulse slicing.

New Science of Ultra-Fast Electron Dynamics will be Enabled by NLS

New Technology for Control and Measurement of Complex Systems is Likely to Follow

Acknowledgements

John Tisch	Imperial	Jonathan Underwood	UCL
Matt Zepff	QUB	Misha Ivanov	Imperial
Will Bryan	Swansea	Olga Sminova	MBI, Berlin
Leszek Frasinski	Imperial	Fred Currell	QUB
Ian Walmsley	Oxford	Roger Falcone	Berkeley, USA
Brian McNeil	Strathclyde		
Riccardo Bartolini	DLS		
Justin Wark	Oxford		