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Ultrafast Science and Development at the Artemis Facility

Edmond Turcu
Central Laser facility
STFC, Rutherford Appleton Laboratory, UK

Micro to Nano-Photonics II, Romopto 2009
Sibiu, Romania , 3rd of September

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Artemis facility is open to UK and EU research groups

Contents

I. Artemis facility development

II. Ultrafast time-resolved experiments in Artemis: atomic and molecular physics and biophysics

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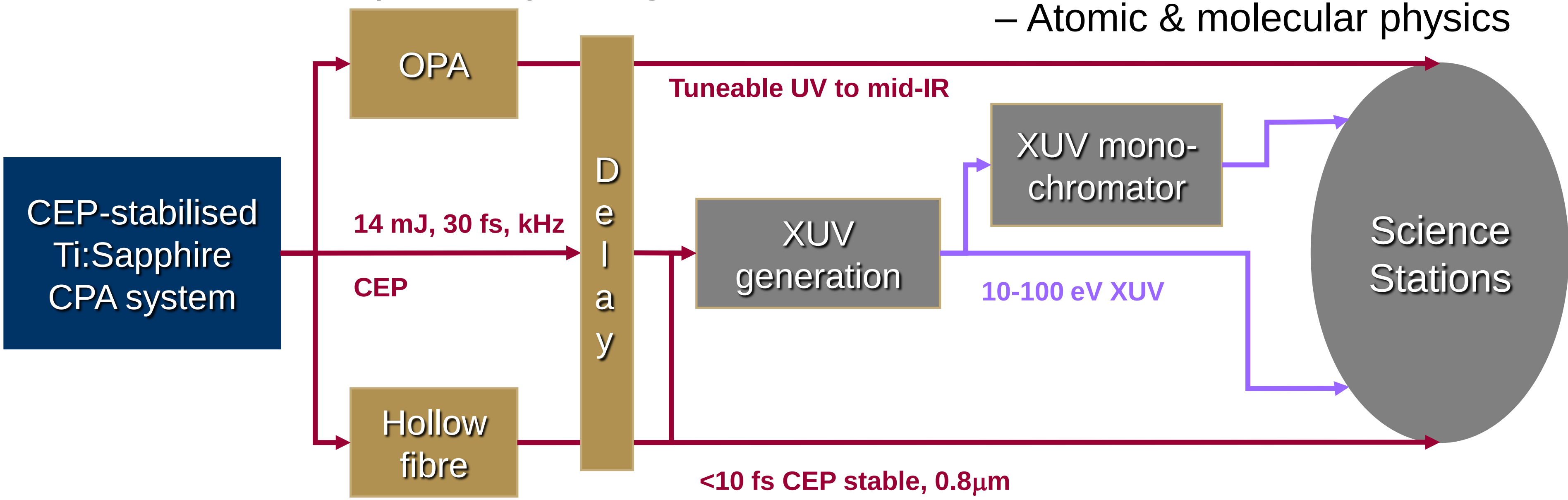
Artemis Facility

- **Three femtosecond laser beams**
  - 14 mJ, 30 fs, 1-3 kHz CEP-stabilised
  - <10fs 800nm pulses
  - 30 fs, tuneable UV to mid-IR
- **Two femtosecond XUV beamlines**
  - Broadband 10-100 eV XUV
  - Monochromatic short pulse (10-50 fs)
  - Future attosecond capability
- All beams synchronised and independently configurable
- **Two End-Stations**
  - Materials science
  - Atomic & molecular physics



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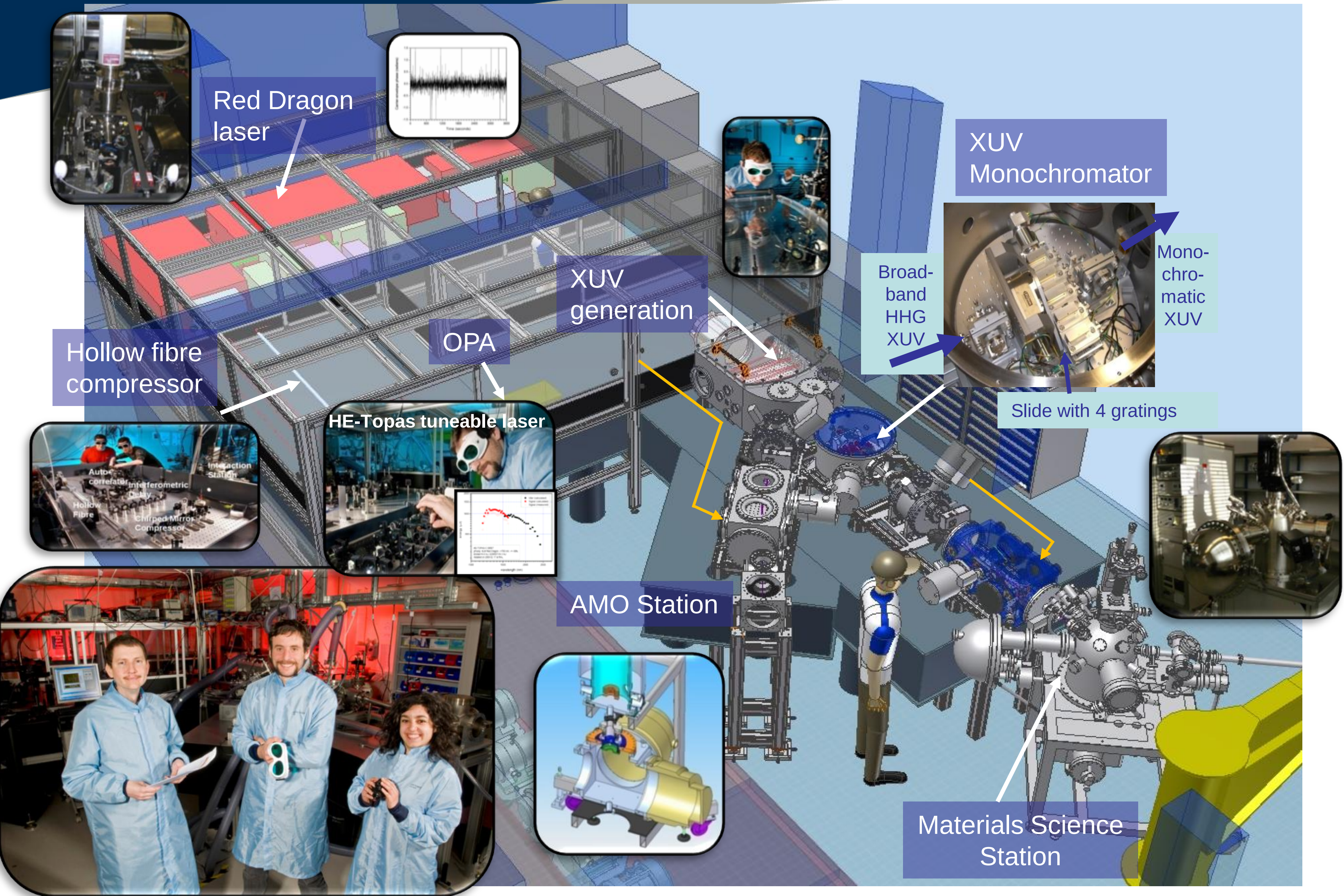
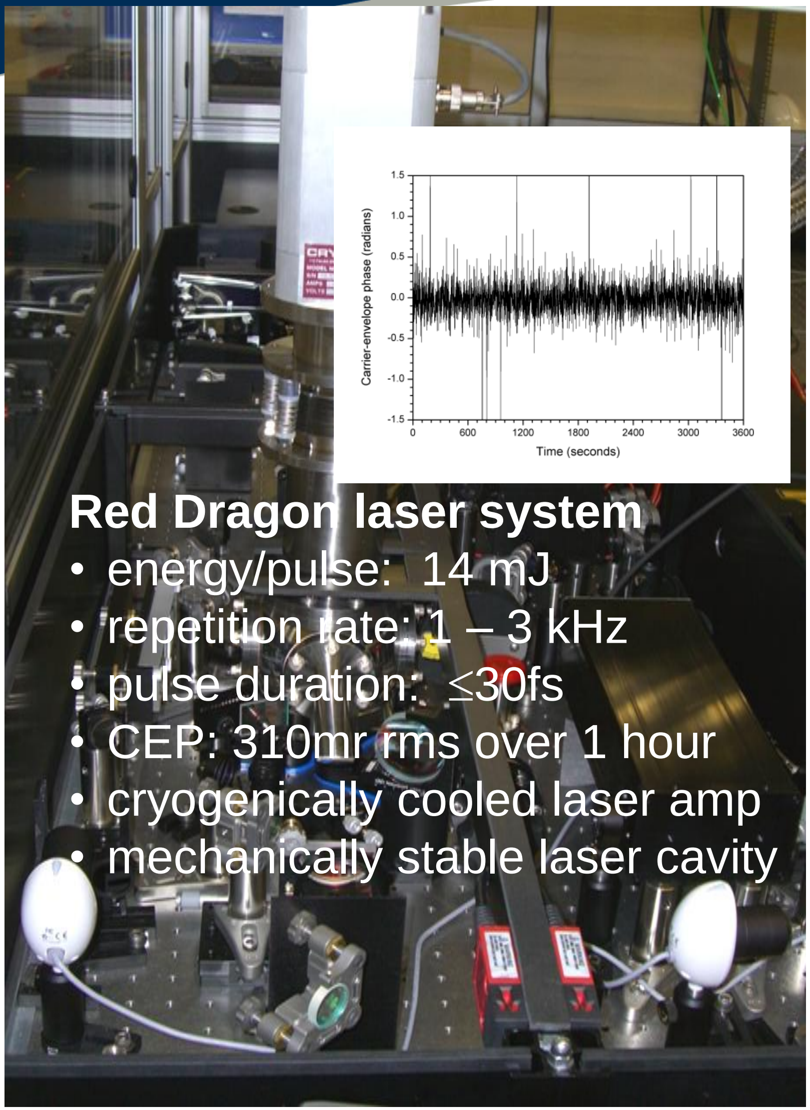
Facility Layout



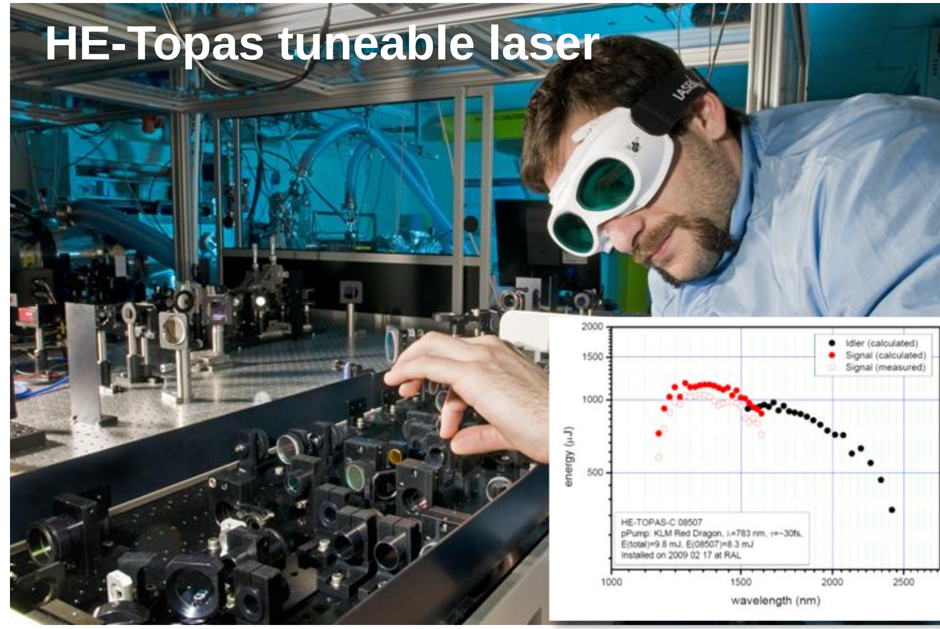
Artemis :Laser Beams



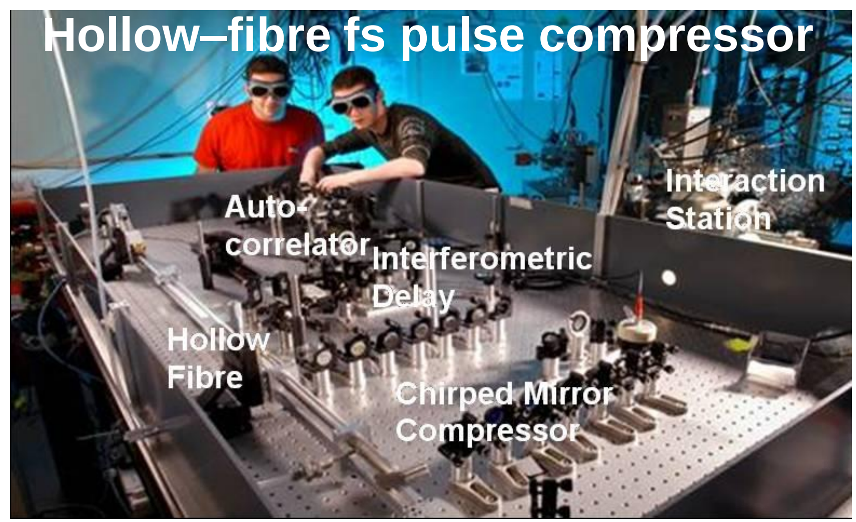
Red Dragon laser system

- energy/pulse: 14 mJ
- repetition rate: 1 – 3 kHz
- pulse duration: ≤30fs
- CEP: 310mr rms over 1 hour
- cryogenically cooled laser amp
- mechanically stable laser cavity

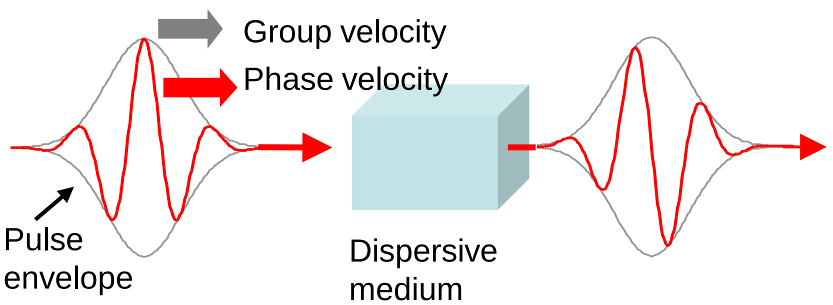
HE-Topas tuneable laser



Hollow–fibre fs pulse compressor

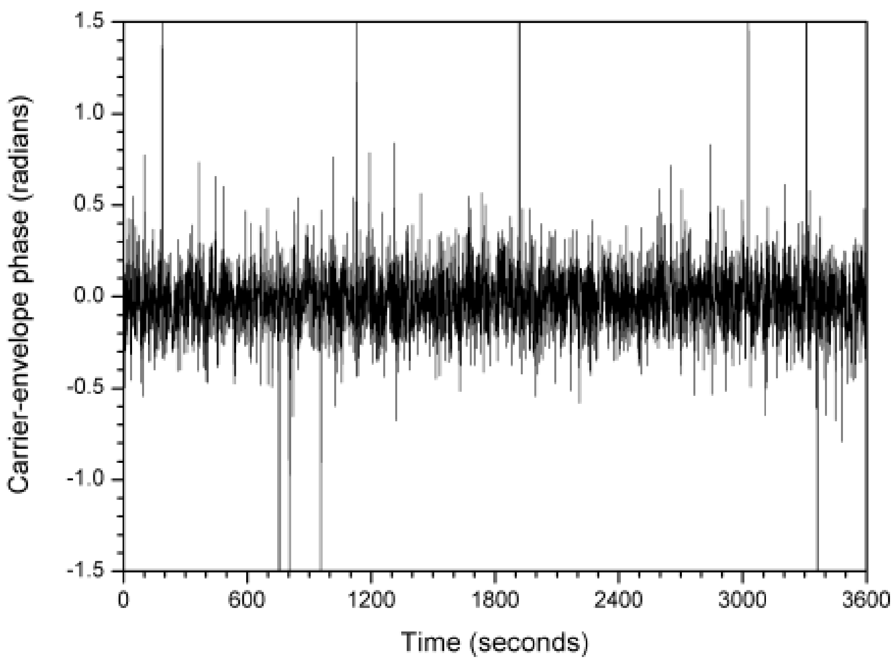


CEP Stabilisation

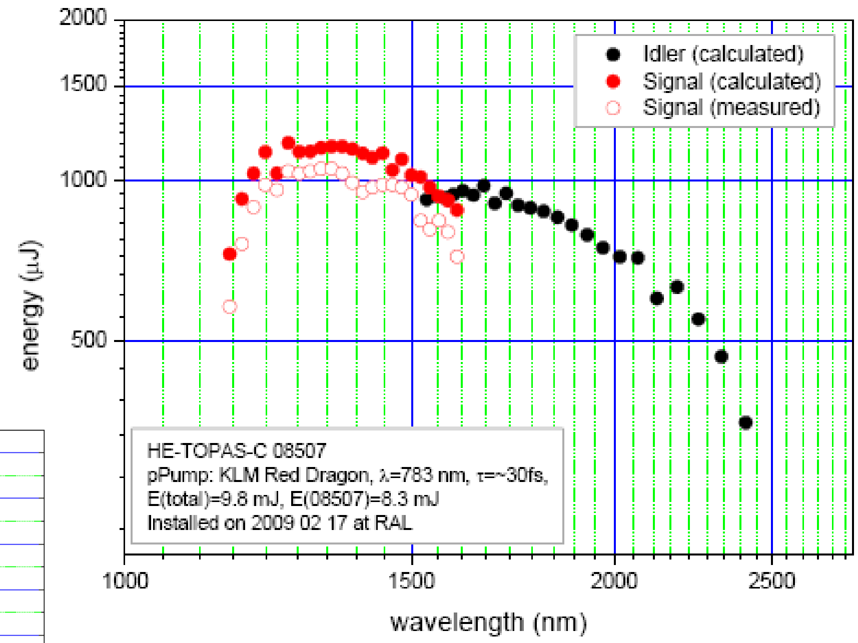


- 14 mJ Red Dragon laser from KML is first multi-stage amplifier system on which CEP has been demonstrated
- 310 mrad rms CEP for over one hour demonstrated in Artemis

- Carrier-Envelope-Phase stability becomes important for few-cycle pulses (<10 fs)
- Requires stabilising path through entire system
- More difficult with grating-compressor based systems as pointing variation into compressor can give large CEP slip (10^4 rad/rad)

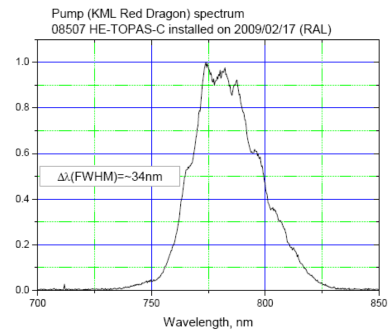


HE-TOPAS Tuneable Laser

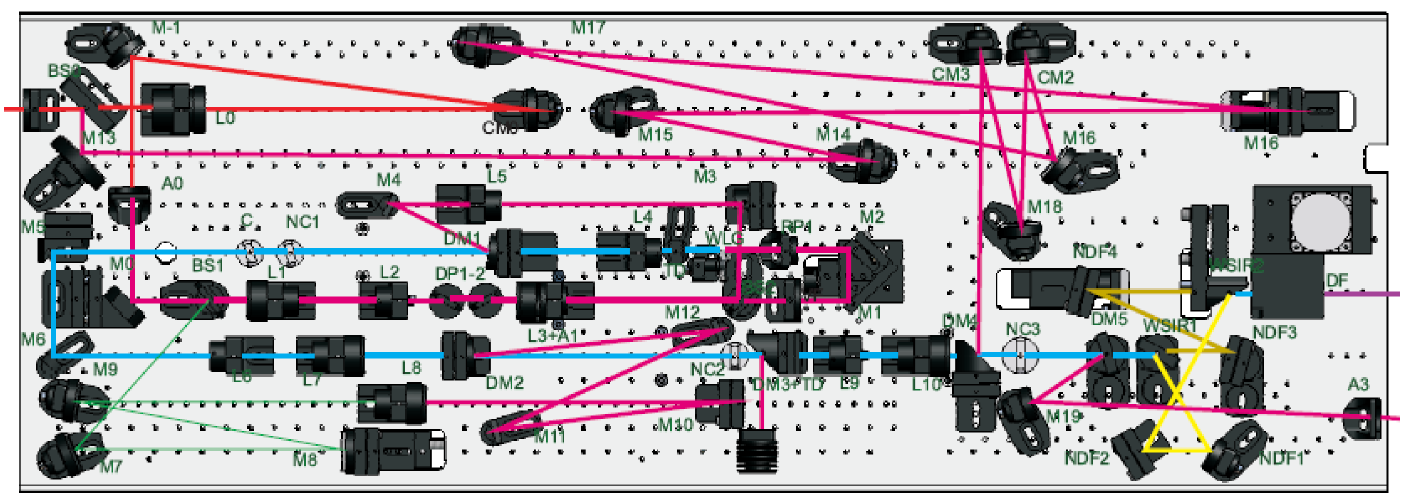


HE-TOPAS OPA from 'Light Conversion'

- Energy/pulse in: 8.3mJ
- Energy/pulse (signal+idler): 2.5mJ@1300nm
- Tuneable:1.2µm -20µm
- Will extend to: 0.2µm - 20µm
- Pulse duration: ≥35fs
- Repetition rate: 1 kHz



Input:
~8.3mJ
~30fs
780nm



Signal

Idler

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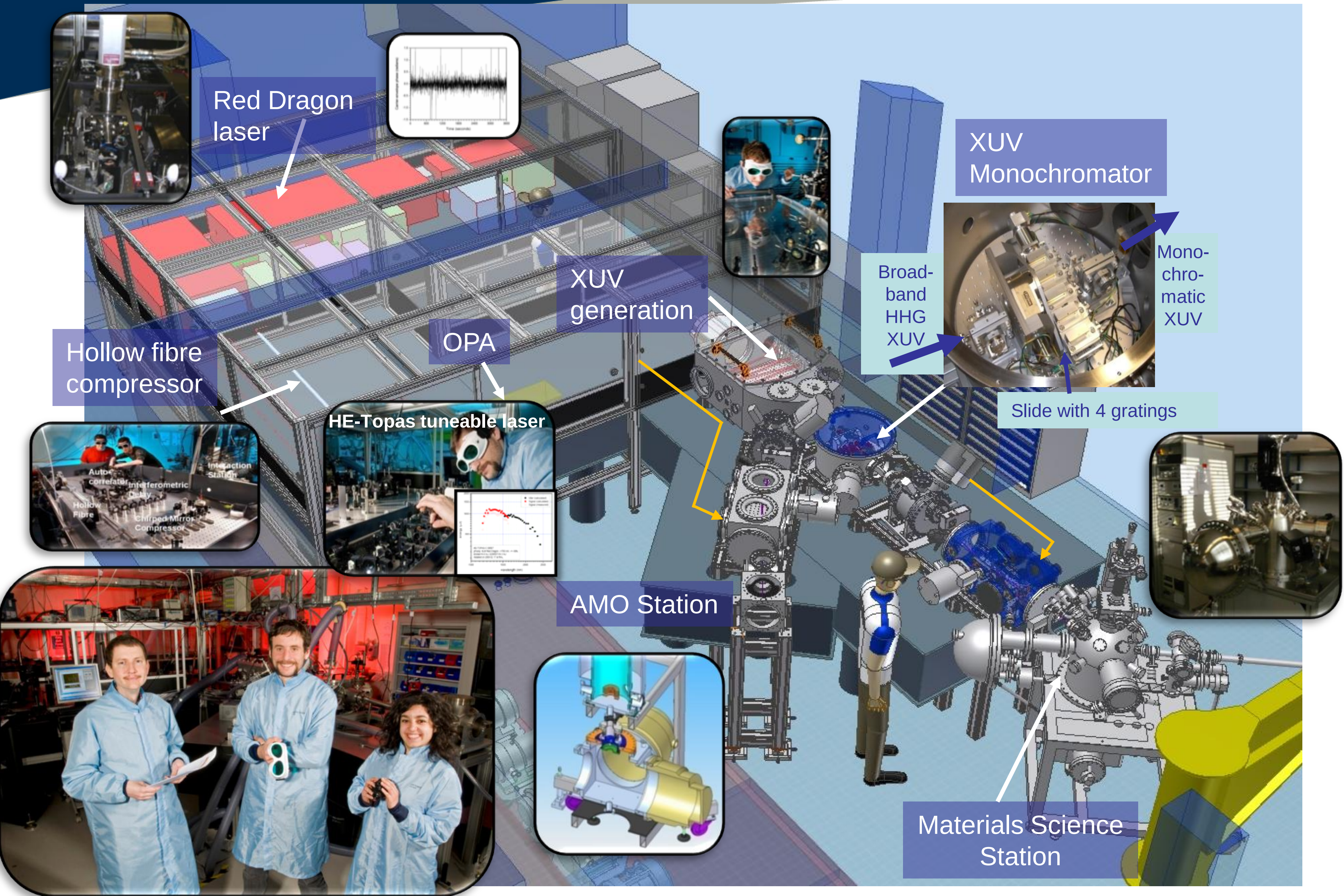
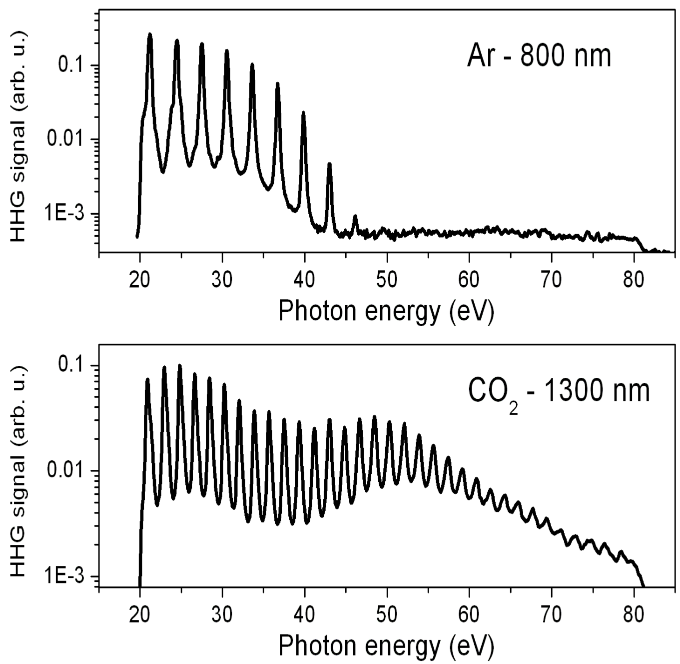
XUV Beamlines



XUV Beamlines

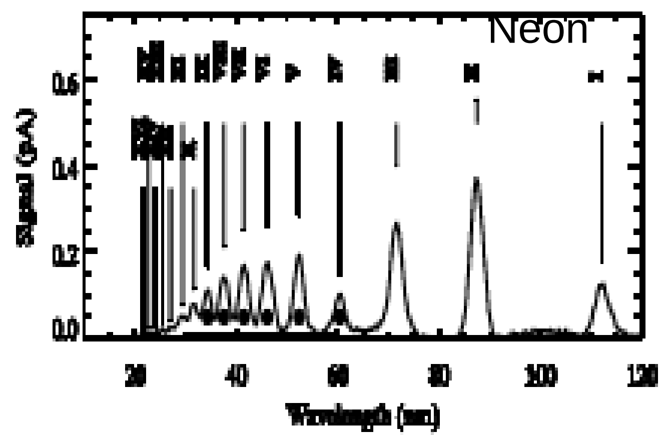
XUV broadband beamline for Atomic and Molecular Physics End-Station

- HHG from 800nm or 1300nm
- Flat-field spectrometer –diagnostics



XUV monochromatic beamline for Materials science End-Station

- >10^9 photon/sec @ 40eV
- Tunable in the 10 eV -100 eV
- XUV bandwidth ~ 20 -100 = $\lambda / \Delta\lambda$
- XUV pulse duration ~10 – 40fs



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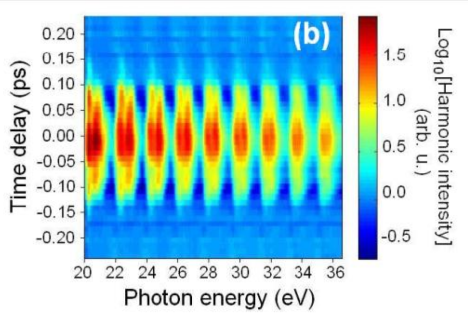
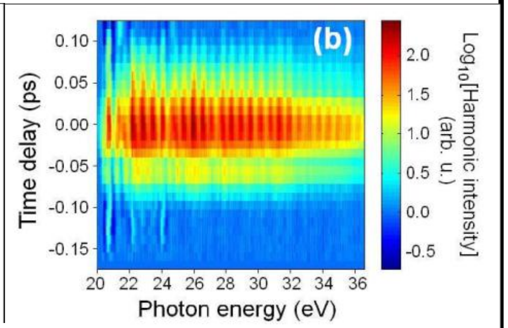
Enhanced HHG with 2-Colour Laser

IC London, UC London, U Napoli



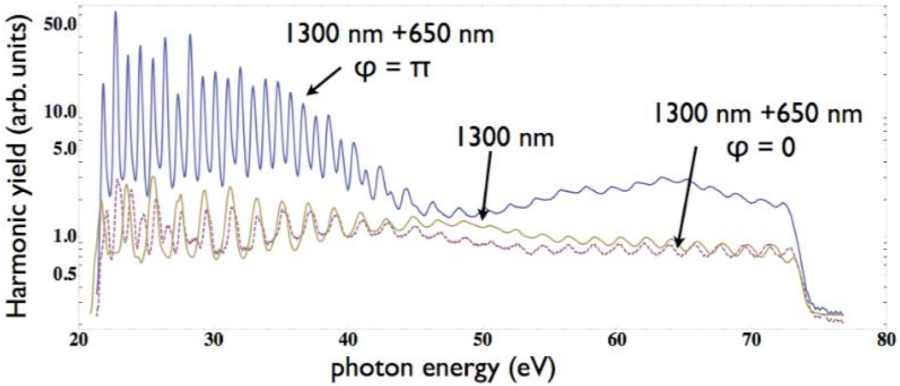


HHG spectra of argon function of delay between λ_1=1300 nm and λ_2=780 nm pulses, parallel polarisation (normalised 1300 nm HHG).

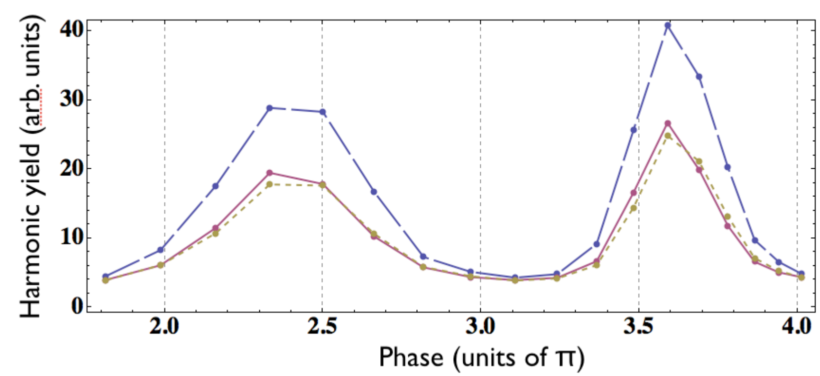
(a) I_1/I_2=1.5 10^{14} W/cm^2/0.2 10^{14} W/cm^2;

(b) I_1=I_2= 0.5 10^{14} W/cm^2;

- **>100x HHG enhancement**
- **Non-integer order HHG**



HHG spectra of argon λ_1=1300 nm and λ_2=650 nm pulses (ω + 2 ω), orthogonal polarisation. **10x HHG enhancement – compensates lower 1300nm efficiency.**



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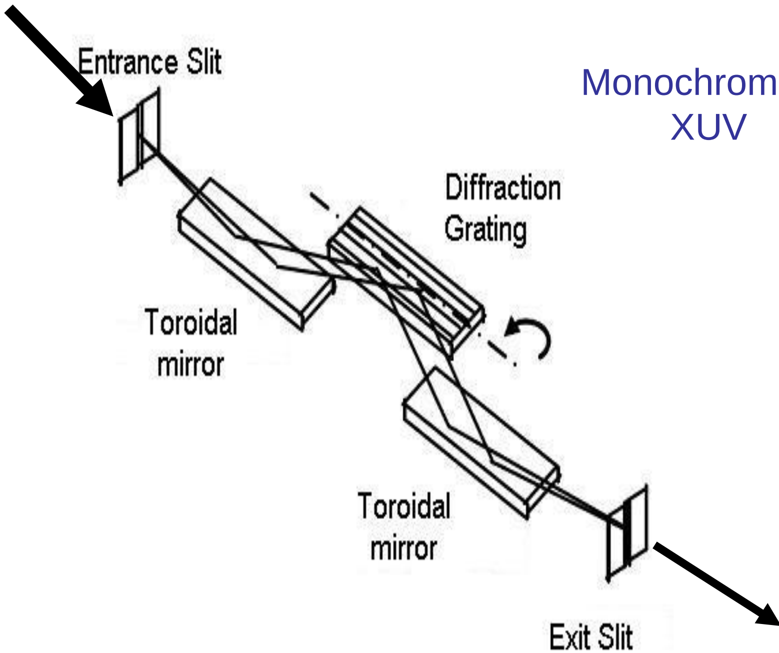
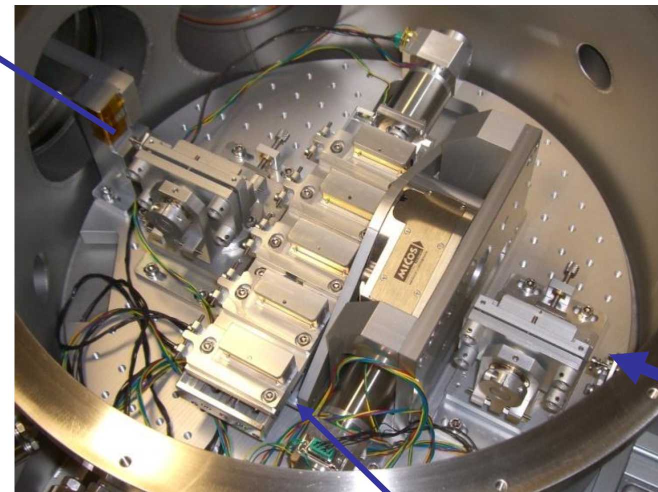
10fs XUV Monochromator

Monochromator will select a single harmonic within a broad band XUV spectrum while preserving pulse-length.





Monochromatic XUV

Broadband HHG spectrum

Slide with 4 gratings for short/long wavelength regions and high/low resolution

- Tunable: 10 eV -100 eV photon energy, 120nm – 12nm wavelength
- Low resolution gratings ($\lambda/\Delta\Lambda$~20): pulse duration ~10fs
- High resolution gratings ($\lambda/\Delta\lambda$~80): pulse duration ~40fs
- Peak efficiency ≈30%

XUV Beamline: fsec Resolution

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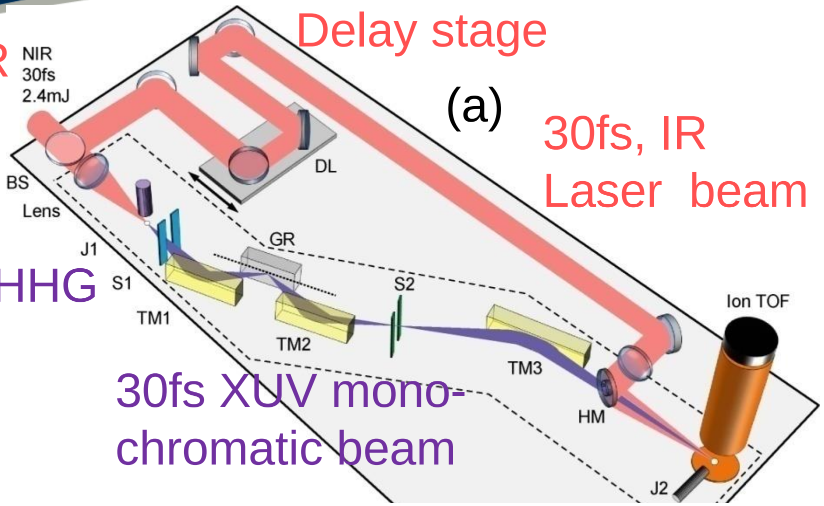
30fs, IR Laser

Splitter

Gas Jet: HHG

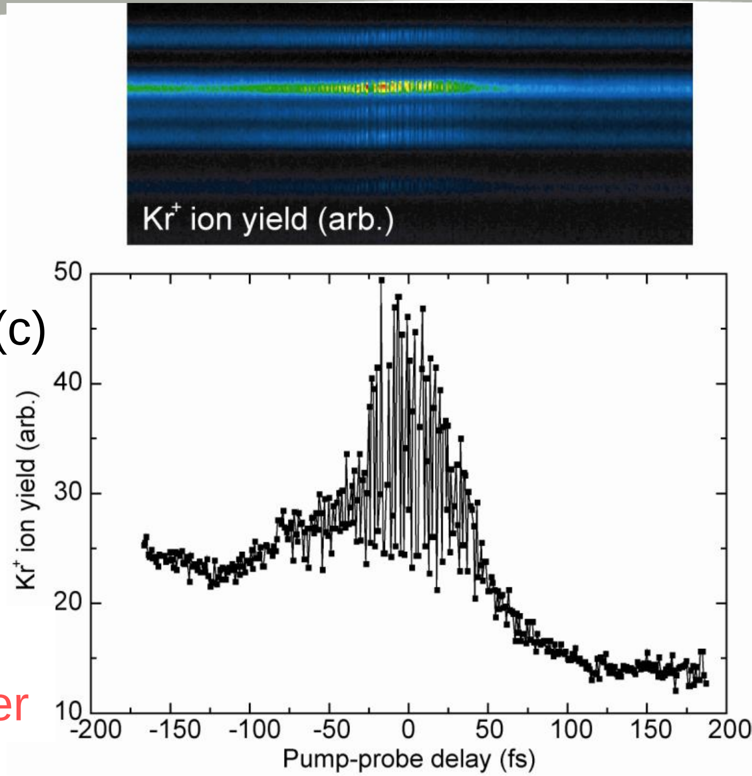
Delay stage

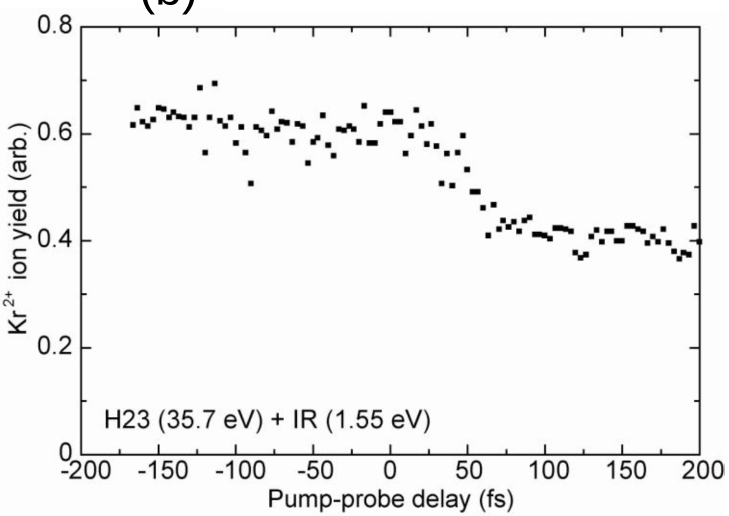
30fs, IR Laser beam



30fs XUV mono-chromatic beam

30fs XUV+IR focused on Kr atoms in ToF spectrometer





(a) <0.6fs Resolution Interferometer XUV + IR : XUV-Pump + IR-Probe for ultrafast science. Stability >12hours.

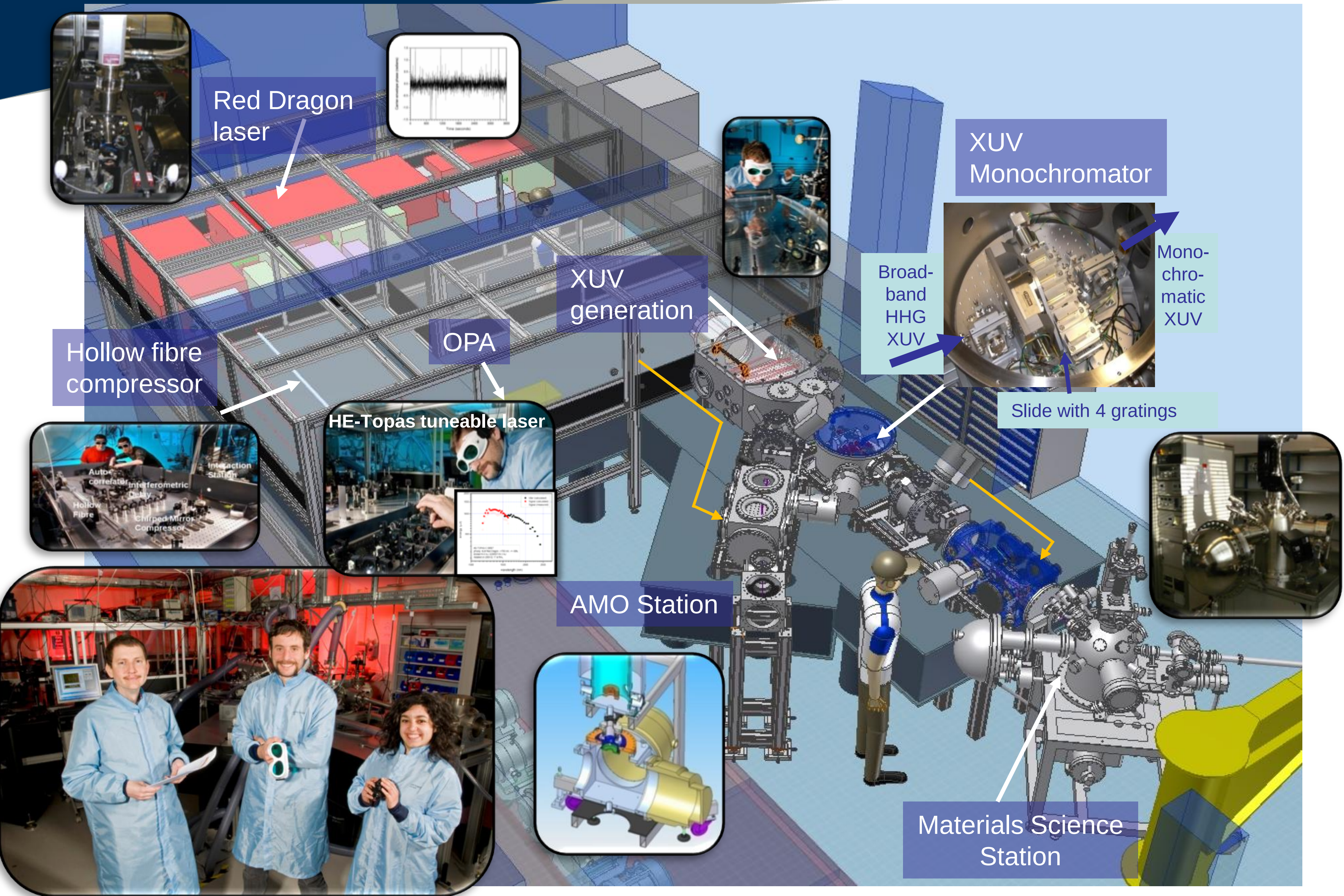
Kr^{2+} ion yield function of temporal delay XUV+IR

(b) XUV pulse duration measured ~ 30fs:

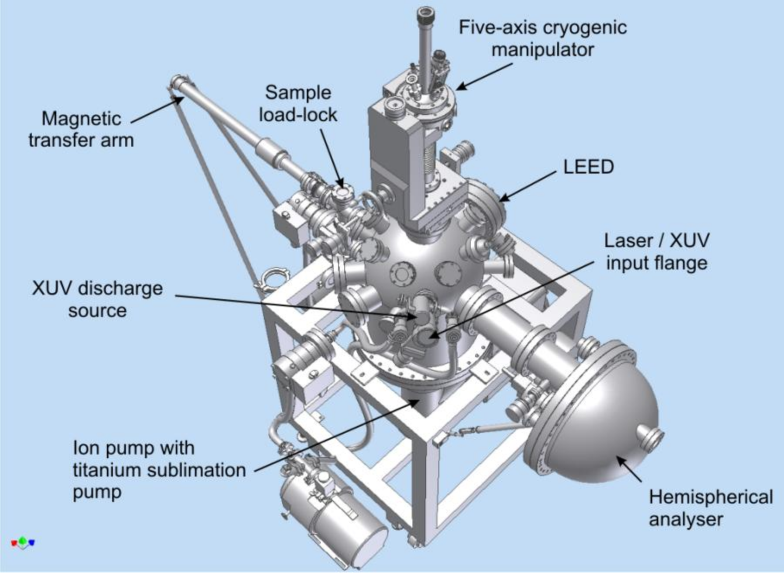
XUV Harmonic 23 (hν=35.7eV) + IR (hν=1.55eV).

(c) Interferometer resolution is < 1/4 laser periods (0.6fs) XUV broadband + (IR + IR)

TR Science End-Stations



Materials Science Station

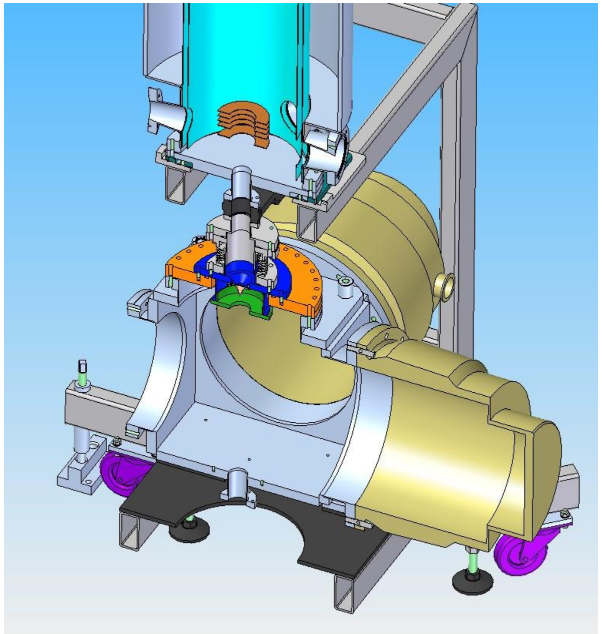


- UHV end-station from SPEC
- < $2\text{x}10^{-10}$ mbar
- Manipulator: 14K liquid-He cooled 5-axis
- Hemispherical electron spectrometer with
- 2-dimensional detector for
- energy- and angle-resolved photo-emission experiments
- Mu-metal chamber
- Sample transfer apparatus
- LEED

Investigation of angle- and time-resolved photo- electron emission for:

- Coherent control and Fermi surface dynamics in complex oxides;
- Non-adiabatic melting of charge order and Mott-gap dynamics;
- Ultrafast core-level photo-emission

AMO End-Station



Atomic and Molecular Physics End-Station

Two coupled chambers:

- molecular beam source in the lower chamber
- velocity-map imaging (VMI) detector for
- ions and electrons in upper section.

Time-Resolved Atomic and Molecular Physics:

- control of electron recollisions,
- time-resolved photoelectron imaging of excited state molecular processes,
- Coulomb explosion imaging of molecular wavepackets.

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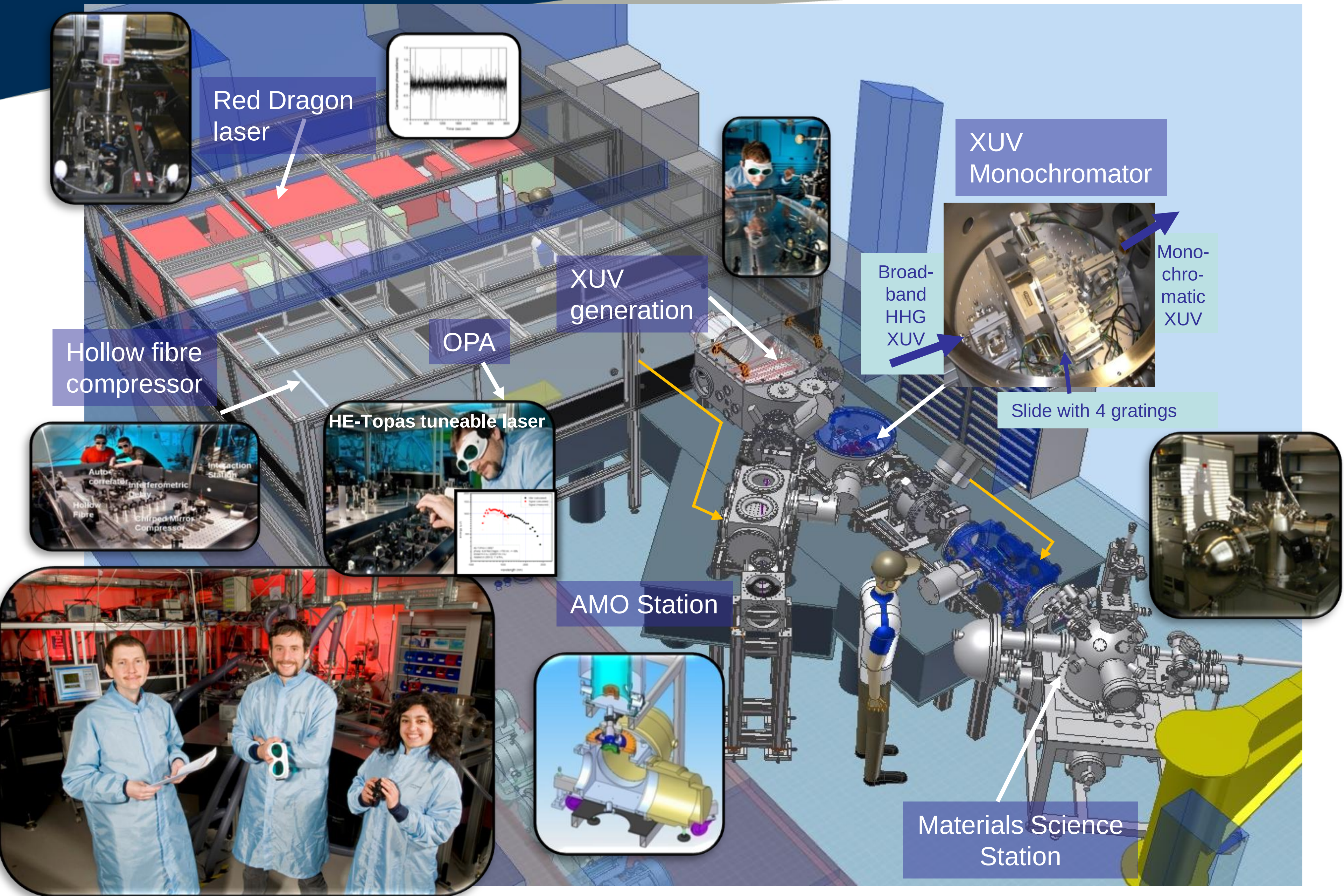
Ultrafast Science



Cold molecule dissociation in ion-traps

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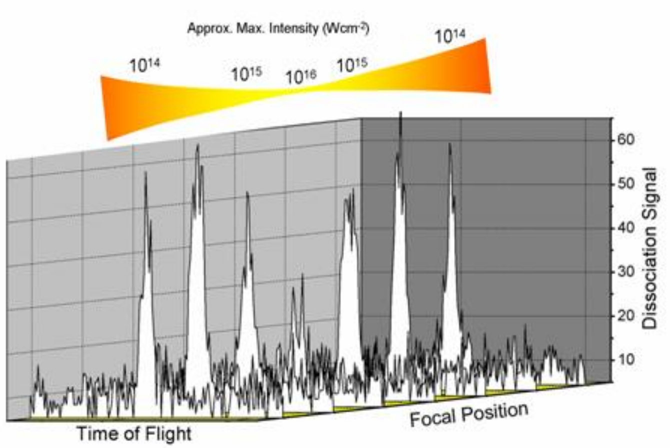
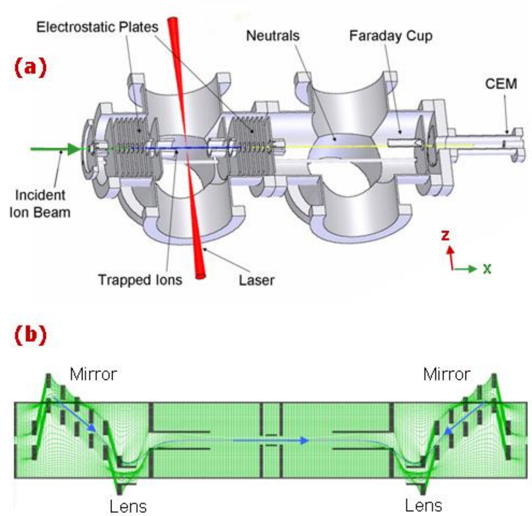
QU Belfast, UCL, Swansea U



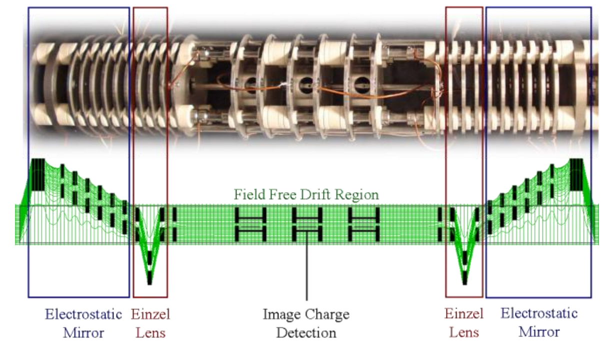


Z-scan of D_3^+ photodissociation in intense fs laser field 10^{16} W/cm^2.

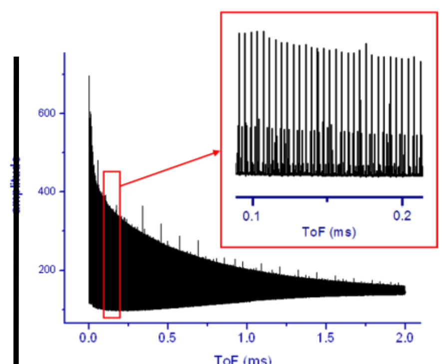
- **first strong field dissociation of D_3^+**
- **two photon process**
- **very stable molecule in strong field**
- **only highly excited ro-vibrational levels of ground state can dissociate - trapping time <10ms**
- **future: TR pump-and-probe 10fs**

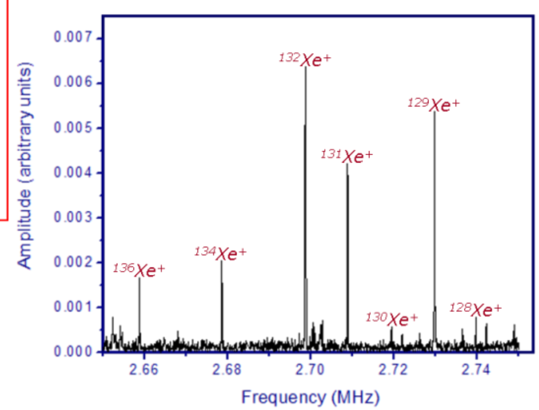
JD Alexander J Phys B 2 141004(2009)
JD Alexander J Phys B42 154027 (2009)

KEIRA: Kilovolt Electrostatic Ion Reflection Analyser



High Resolution Mass Spectrometer

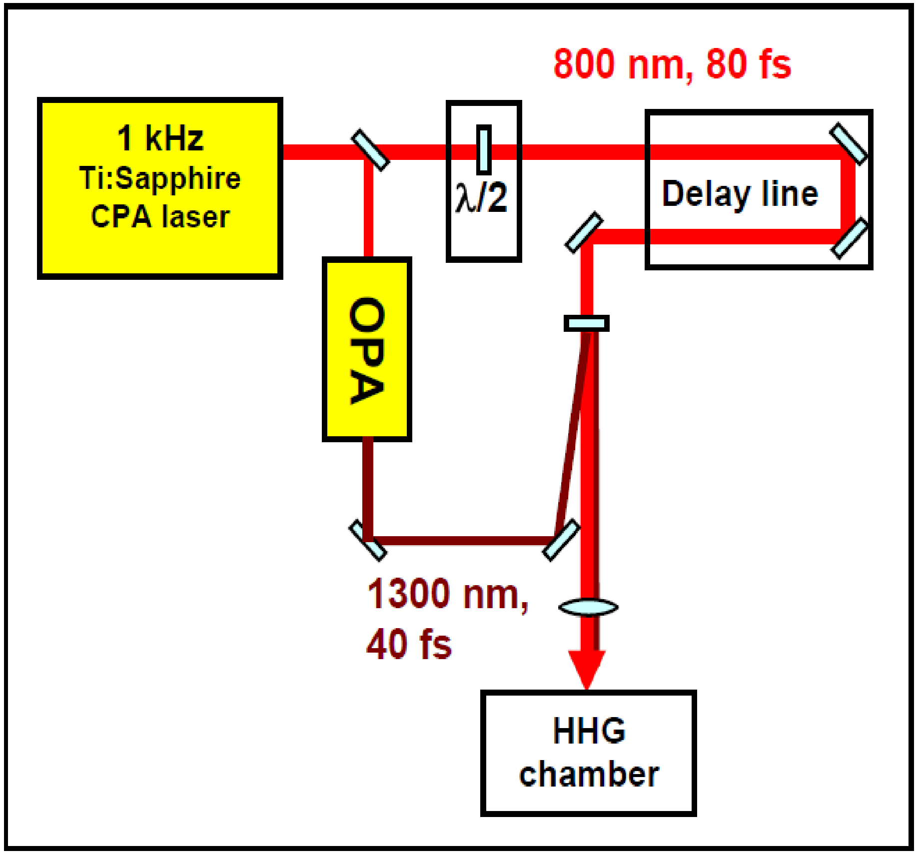


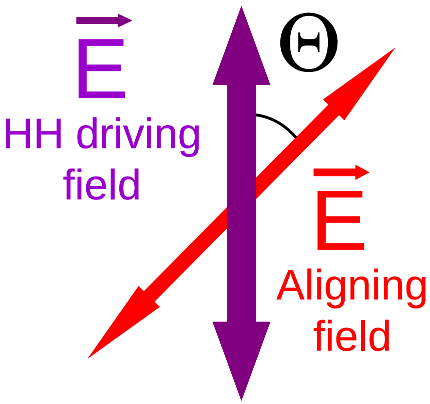


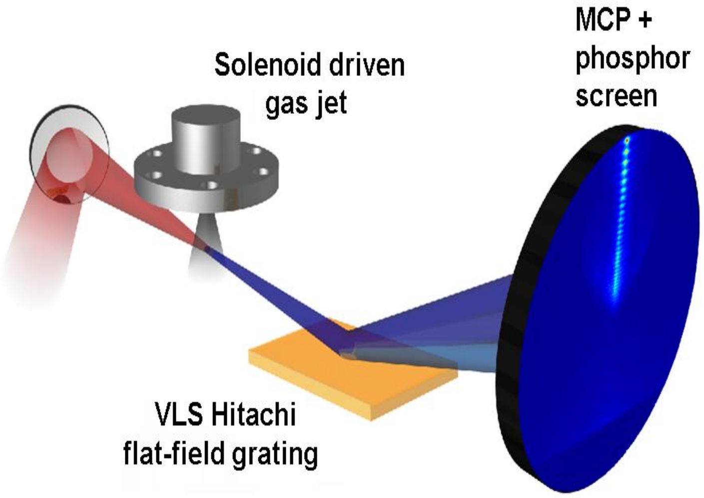
The 21st harmonic (in frequency space) of trapped xenon isotopes, shows a **mass resolution of ~ 10^4.**

IC London, UC London,
U Napoli

Probing Molecular Structure and Dynamics With Mid-IR HHG in Aligned Molecules







Artemis laser at CLF was used as it has a high power TOPAS synchronised to 800nm used for molecular alignment

Benefit of 1300nm vs. 800nm laser: cut-off extension of HH spectrum

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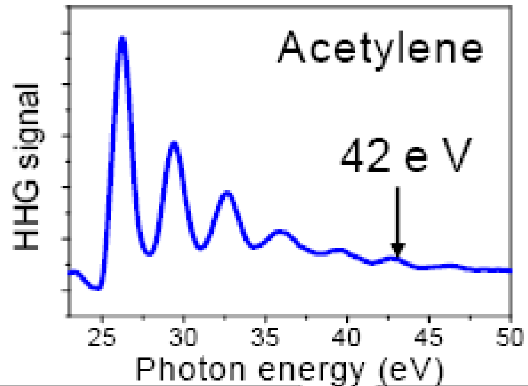
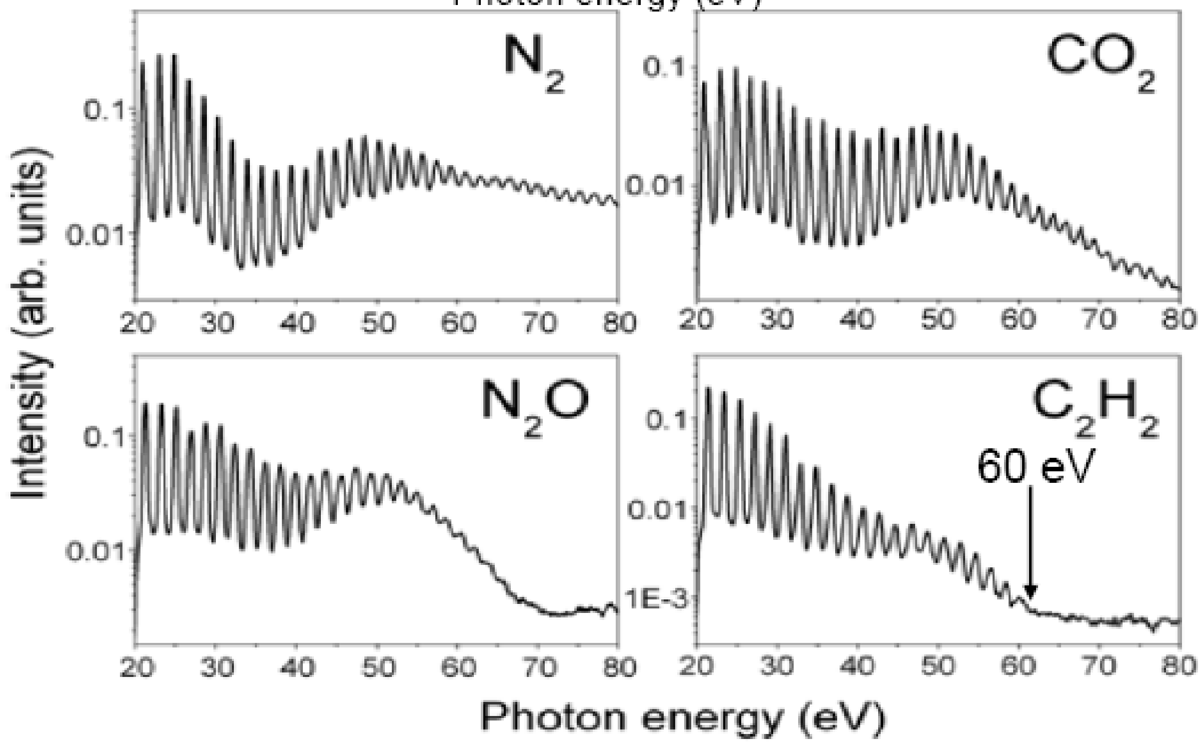


10fs 800nm – cut-off fixed by ionisation saturation intensity, especially if Ip lower.



40 fs 1300nm
Un-normalized
spectra

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Evidence for Structural Interference

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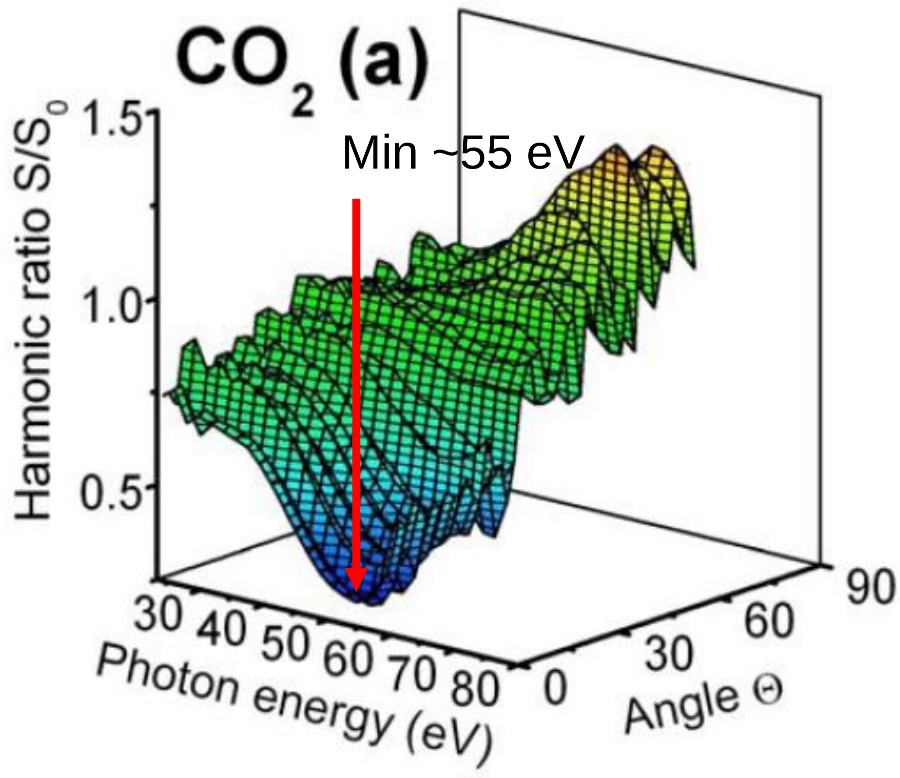
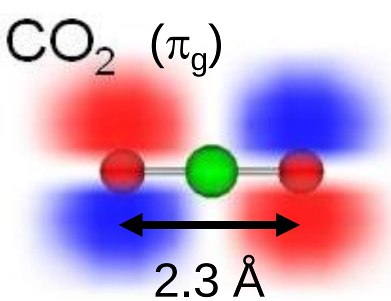


High-Harmonic XUV spectrum modulation in CO_2 molecules shows:

- strong minimum at same position for 1300nm as at 800nm laser driver
- minimum at similar XUV energy as in N_2O which has same spacing
- Evidence that the dominant contribution is structural interference.



Destructive if
λ_{deB}= 2.3 $\cos\theta$ Å
(~ 51 eV if θ~ 30^0)
~60 eV if θ~ 40^0

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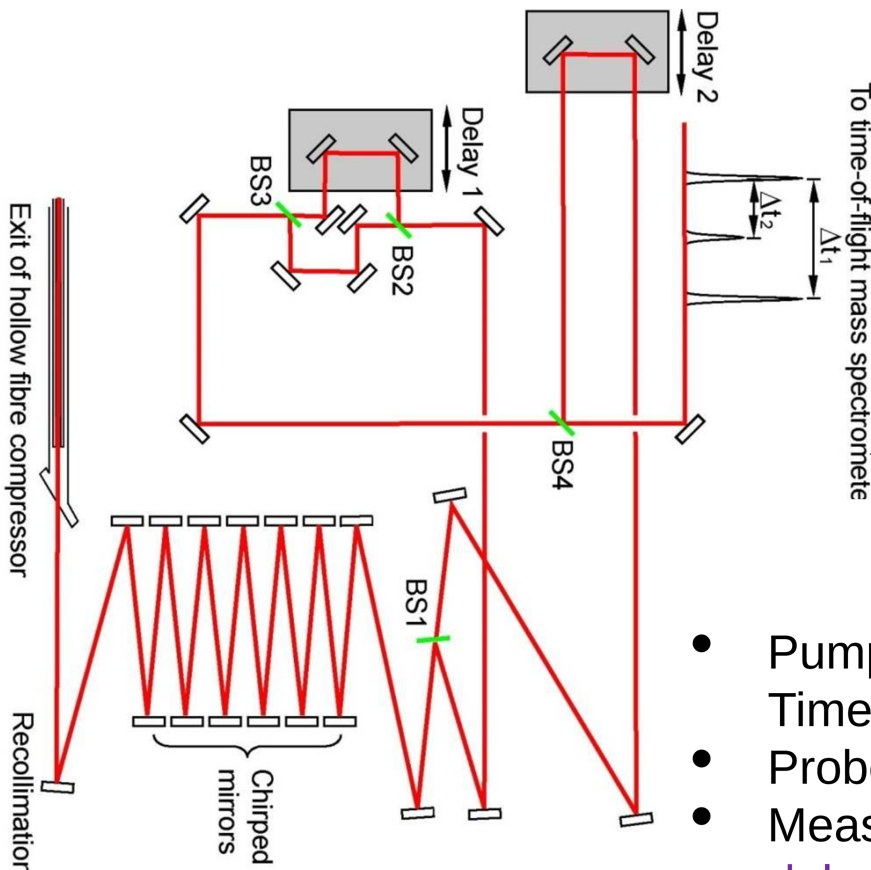
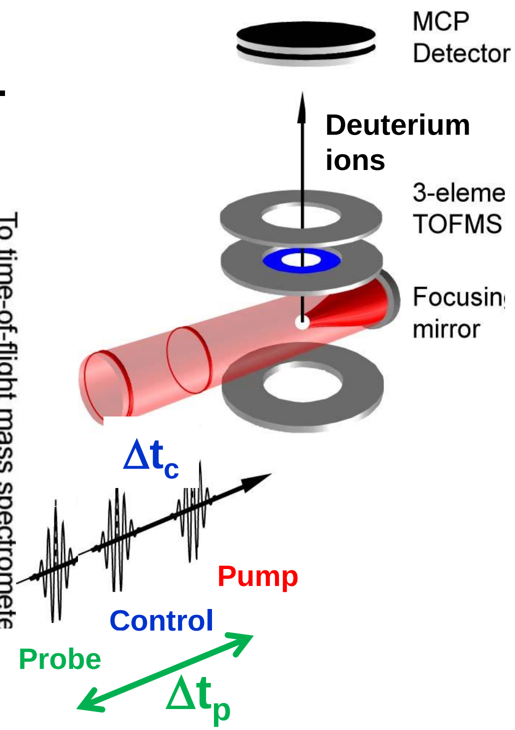
UCL, QU Belfast

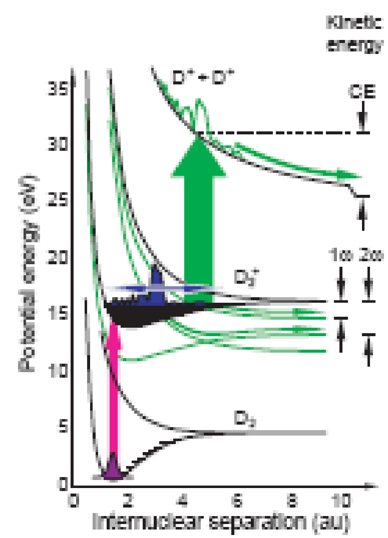
Quantum phase- and population-control in a vibrational wavepacket

Three-pulse Mach-Zehnder interferometer with **two independent delays: pump-probe and pump-control.**

- 0.3 mJ, 10 fs from hollow fibre compressor (λ_0 = 800 nm).
- Stable to < 1/6th optical cycle.
- Resolution ~ 300 attoseconds.







- Pump-control-probe pulse train focused into Time-of-Flight Ion Spectrometer
- Probe photo-dissociated ($D_2^+ \rightarrow D^+ + D$).
- Measure D^+ yield function of pump-probe delay for different pump-control delay.

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Controlling vibrational population and phase

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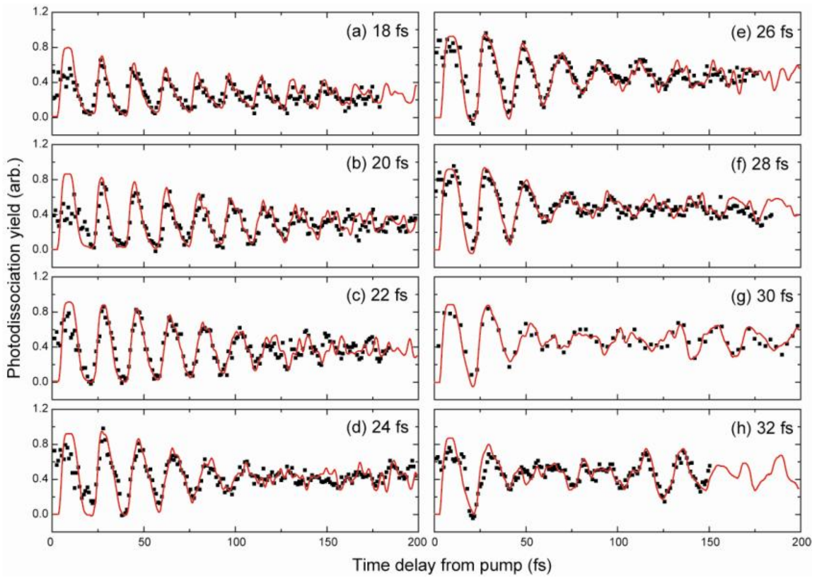
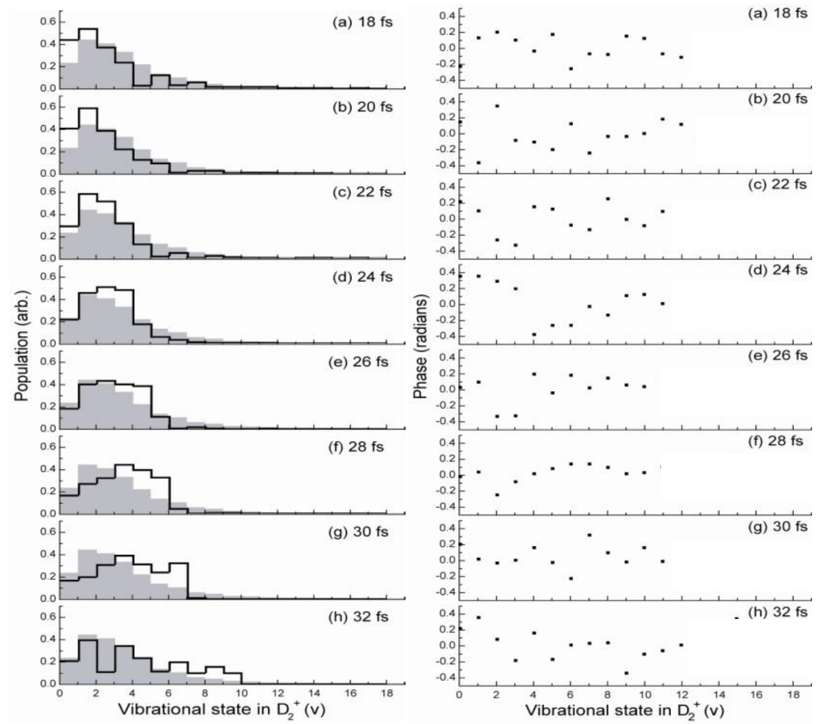
UCL, QU Belfast





Time-delay between 10fs pump-probe pulses for variable control pulse delays.

Good agreement between: experimental PD yield and the Quasi-Classical-Model of wavepacket manipulation.

- Initial and final vibrational populations: **control pulse drives the population** significantly. Control 18fs to 32 fs: distribution shifts from “cooler” to “hotter”...
- Phase variation has less structure.

Better control expected with two or more control pulses.

Future : Transfer of energy → applications in **chemical reactivity**.
Phase and population → single molecule **quantum computation**.

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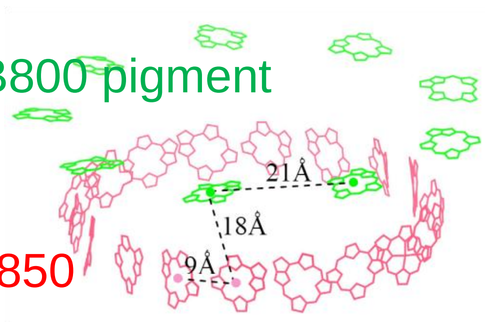
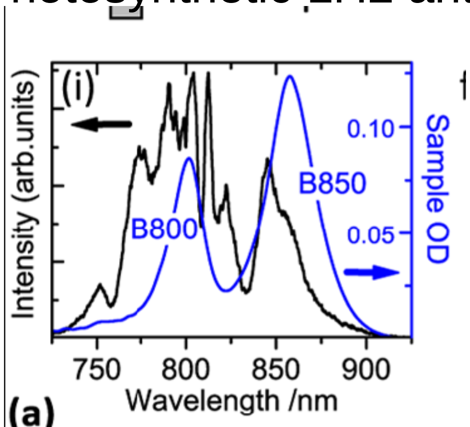
UC Dublin, IC, Glasgow U

Photosynthetic protein: energy transfers and coherences

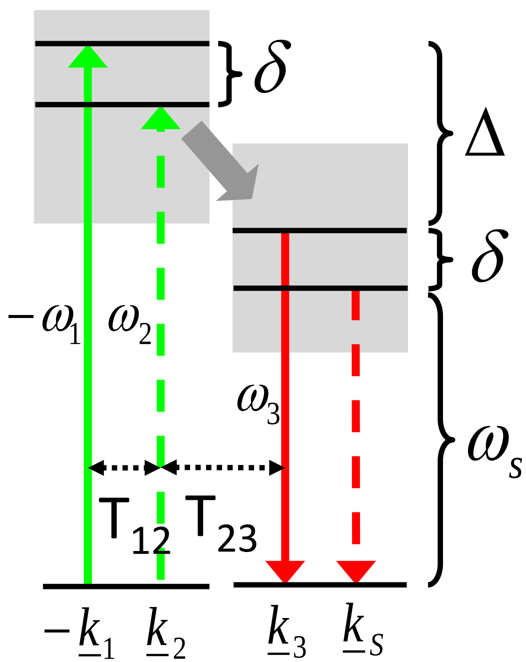
Angular Resolved Coherent (ARC) imaging : New 4-wave mixing method for imaging an arbitrary number of molecular quantum couplings with a single 10fs laser pulse and 2-D CCD camera



Photosynthetic LH2 antenna



10fs laser spectrum overlaps both B800/B850 absorption spectra



ARC RESULTS:
Coherence beat frequency
δ-decay time = 160fs

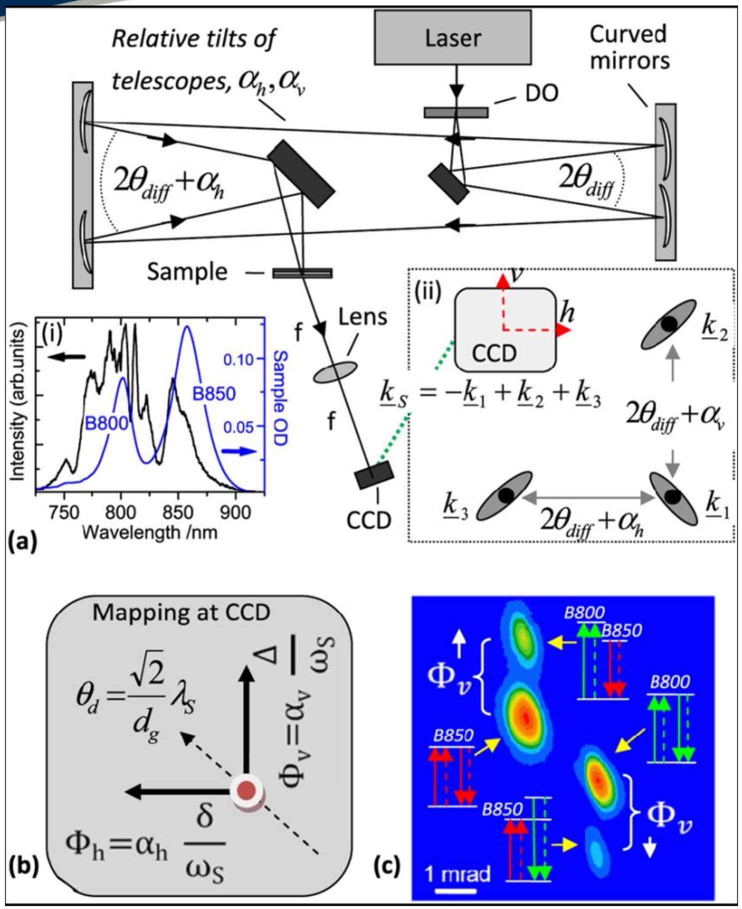
Can determine quantum transition energies directly from position of feature on map

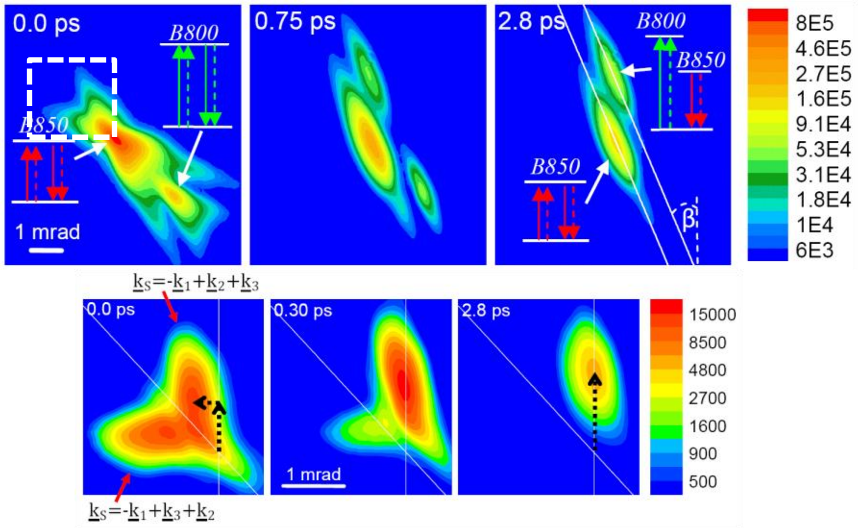
Time-ordered light-matter interaction sequence for B850 pigment of the light harvesting complex II of purple bacteria observed in this experiment. T_{12} and T_{23} represent times between light-field interactions

Ian P. Mercer , Phys. Rev. Lett. 102, 57402, 2009

Angular Resolved Coherent Imaging: Full Map with Single Laser Pulse

UC Dublin, IC, Glasgow U





Measured time-dependence of energy transfer. ARC-TG maps of LH2 for pulse time-delays T_{23}=0, 0.75ps, 2.8ps.
Top: full map.
Bottom: white insert detail of map (a) with filtering at 880nm, 10nm BW

ARC experimental apparatus.
(a) laser-beams with Diffractive Optic (DO)
(b) Illustration of ARC signal mapping at the CCD detector
(c) Measured ARC map of LH2 at T_{23}=1.3ps

Conclusions

Artemis science facility development:

- *Ultrafast synchronized beams:*
  - ***Laser**: Red Dragon, HE-TOPAS, Few cycle hollow fibre*
  - ***XUV**: Monochromatic-tuneable and Broad-band*
- *Ultrafast time-resolved science end-stations:*
  - *Materials science,*
  - *Atomic and molecular physics and chemistry.*

Ultrafast time-resolved science:

- *Molecular structure and dynamics*
- *Quantum control*
- *Energy transfer in photosynthesis*
- *Mass spectrometry with cold molecules*

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Contributors

STFC Central Laser Facility: Edmond Turcu, Emma Springate, Chris Froud, John Collier, STFC Daresbury Laboratory: Mark Roper

Imperial College London, University College London , Universita di Napoli Collaboration: Jon Marangos, John Tisch, Ricardo Torres La Porte, Thomas Siegel, Yasin C. El-Taha,, Leonardo Brugnera, Jonathan G. Underwood, Immacolata Procino, C. Altucci, R. Velotta,

University College Dublin, Imperial College London, University of Glasgow Collaboration: Ian Mercer, Yasin El-Taha, Nathaniel Kajumba, Jon Marangos, John Tisch, Mads Gabrielsen, Richard J. Cogdell

Queens University Belfast, University College London, Swansea University Collaboration: Jason Greenwood, Chris Calvert, Orla Kelly, Raymond King, Ian Williams, Roy Newell, William Bryan, Jamie Nemeth

University of Padova Collaboration (XUV monochromator): Luca Poletto, Paolo Villoresi, Fabio Frassetto, Stefano Bonora

Diamond Light Source Ltd, MPG Structural Dynamics Hamburg, University of Oxford Collaboration (Materials Science): Andrea Cavalleri, Sarnjeet Dhesi,

- **Artemis facility call for proposals: September 2009**
- **New Artemis permanent position expected shortly: Experimental Scientist: Post-Doc/Senior Post-Doc Contact Emma.Springate@stfc.ac.uk and check website.**

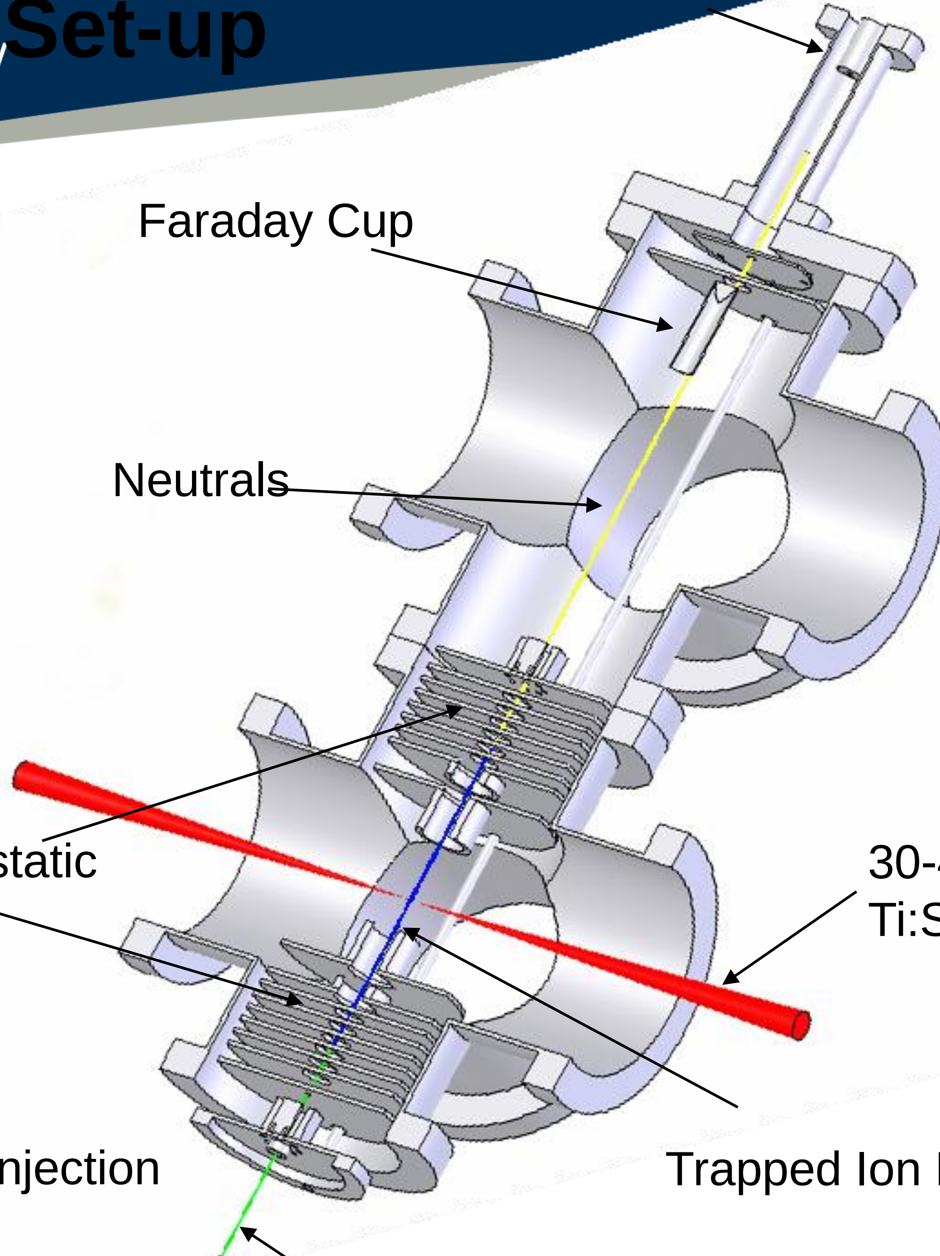
www.clf.stfc.ac.uk

Thank You!

New Facilities

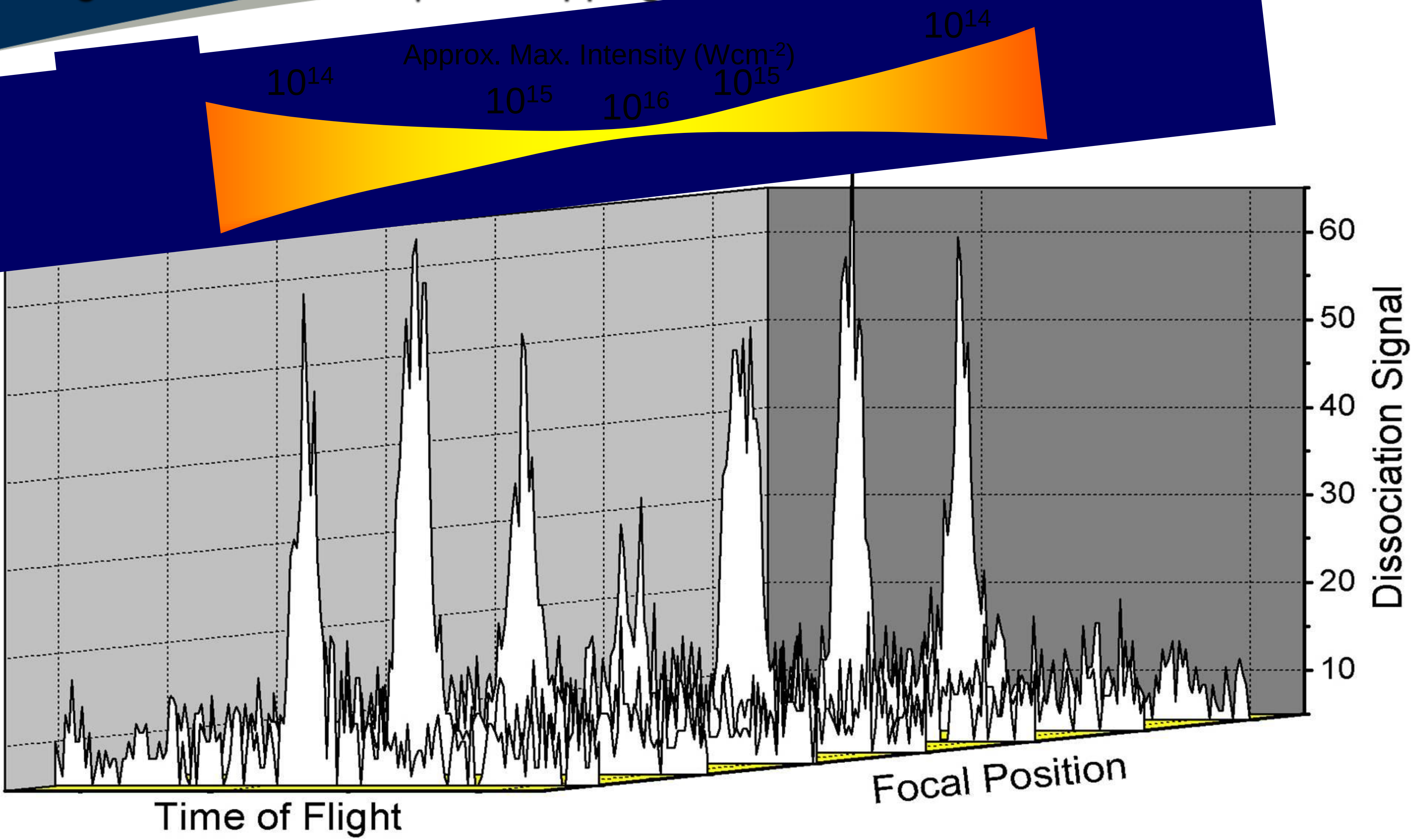
- Vulcan Petawatt: 400 J in 400fs
  - Upgrade to 10PW: 400J in 40fs
- Gemini Petawatt: 2x15J in 30fs, 3pulses/min
- Artemis: fsec XUV+laser beams, kHz
- Ultra: fsec laser beams, 10kHz
- Future facilities/proposals
  - HiPER fast ignition fusion test facility (EU).
  - NLS, 4th generation light source (UK)
  - Dipole, diode pumped laser (EU)
  - ELI, extreme light (EU)

Experimental Set-up



D_3^+ Dissociation z-scan

- Signal from first 500 μs of trapping

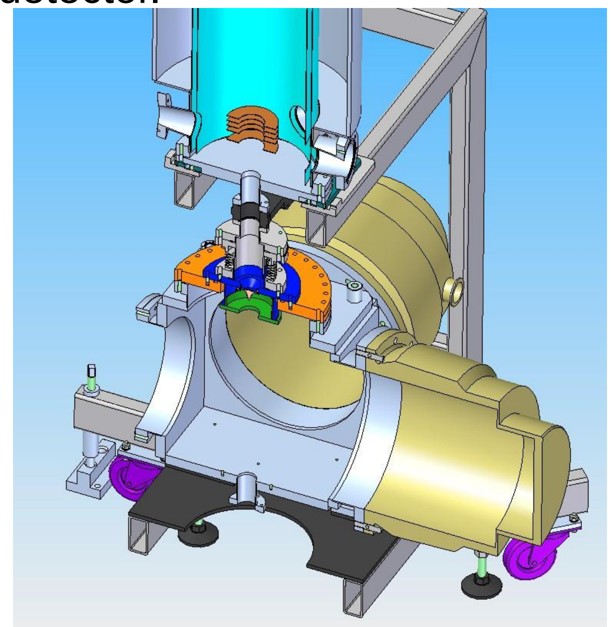


- Low order process (2 or 3 photon)

Artemis: End Stations

Science & Technology Facilities Council
Central Laser Facility

Atomic and Molecular Physics End-Station:
cross-section showing the molecular beam chamber located underneath the velocity-map imaging detector.



Two coupled chambers, with molecular beam source in the lower chamber and velocity-map imaging (VMI) detector for ions and electrons in upper section.

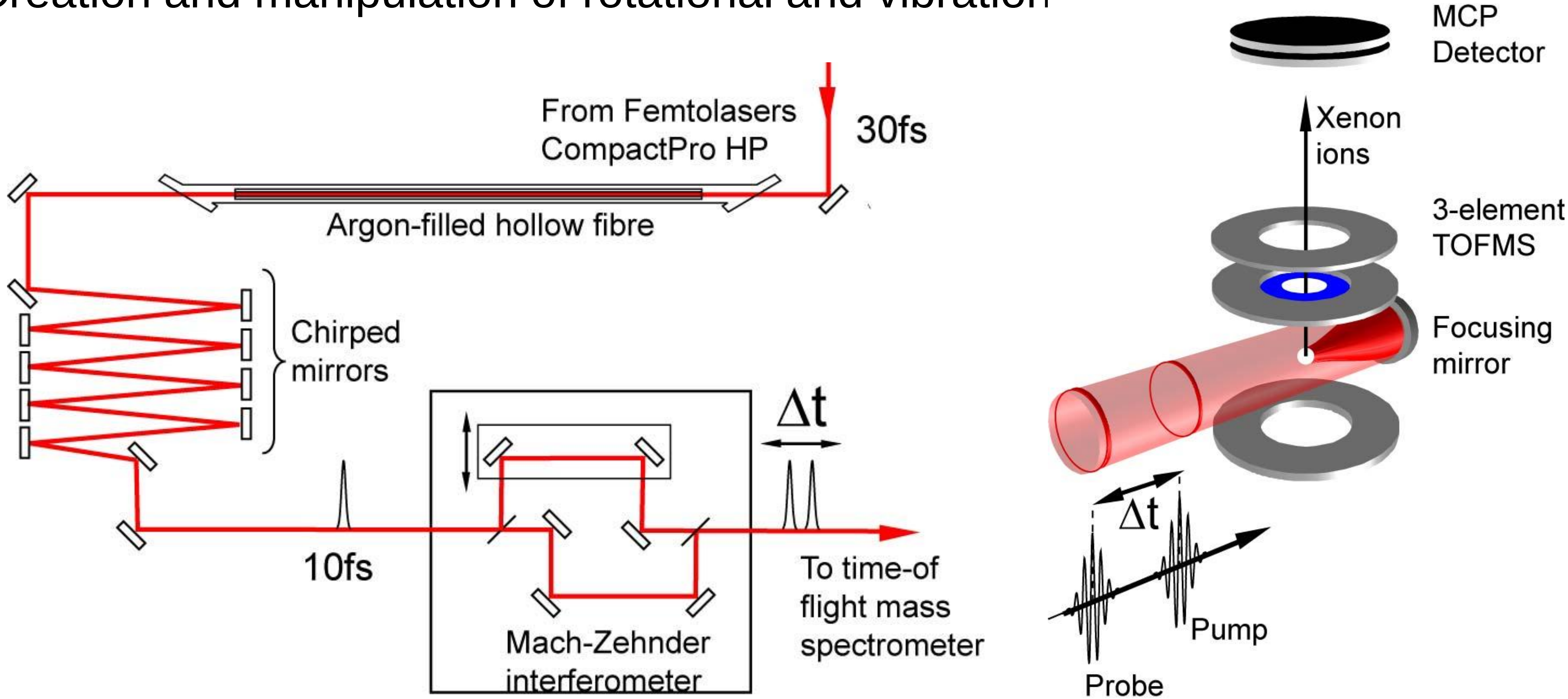
Science: control of electron recollisions,
time-resolved photoelectron imaging of excited state molecular processes,
Coulomb explosion imaging of molecular wavepackets.

Materials Science End-Station
showing the hemispherical electron analyser.

UHV (< 2×10^{-10} mbar) chamber,
a liquid helium-cooled five-axis manipulator and a hemispherical analyser equipped with 2-dimensional detector for energy- and angle-resolved photoemission experiments

Measuring ultrafast molecular processes

- Co-linear interferometer for 10fs pump and probe pulses
- Delay resolution ~0.15 fs, range 0 - 150 ps
- Creation and manipulation of rotational and vibrational wavepackets



Creating vibrational wavepackets in HD

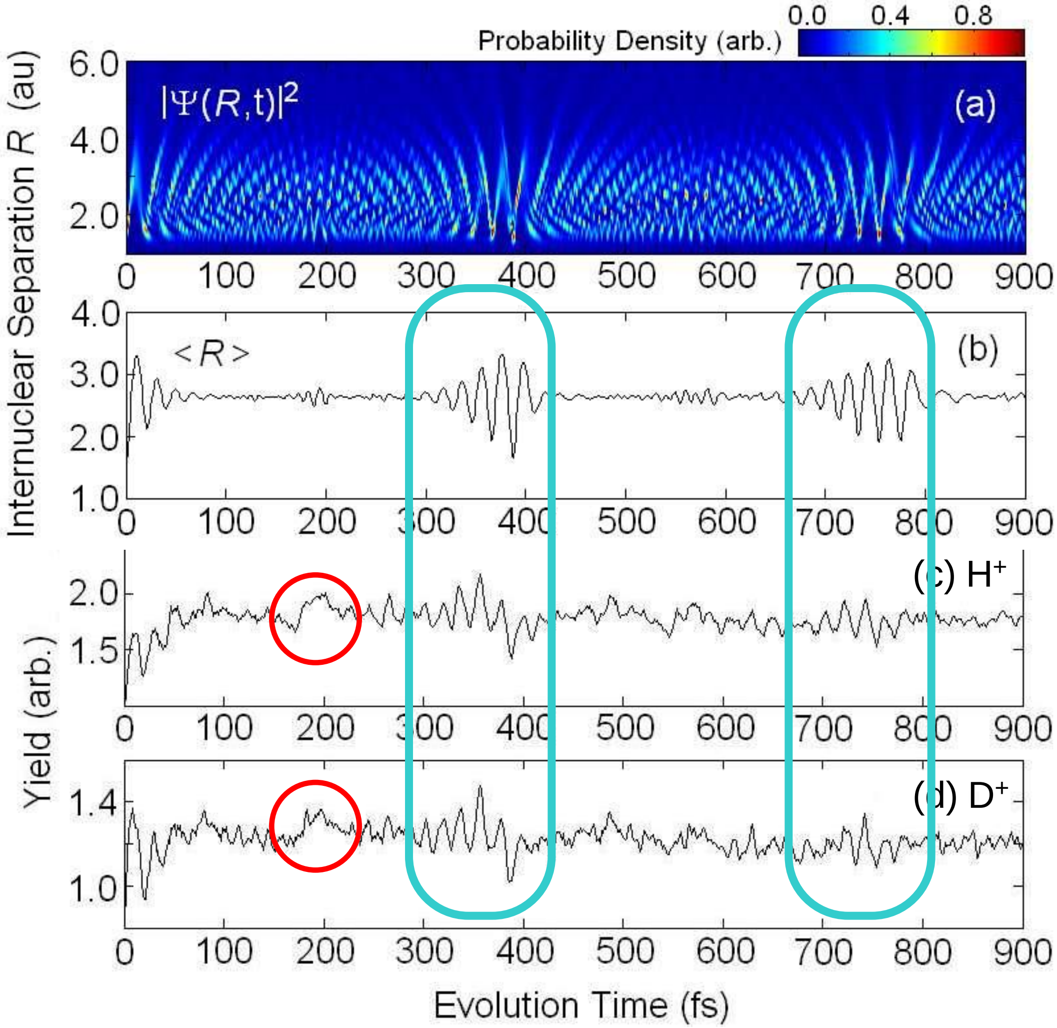
J McKenna et al, J. Mod. Opt. vol. 54 p1127 (2007)

Simultaneous observation of rotational and vibrational dynamics:

(a) Motion of a pure vibrational wavepacket in HD^+ (theory)
(b) Expectation of wavepacket position (theory)

Observe vibrational wavepacket
(c) Measured H^+ integrated yield.
(d) Measured D^+ integrated yield.

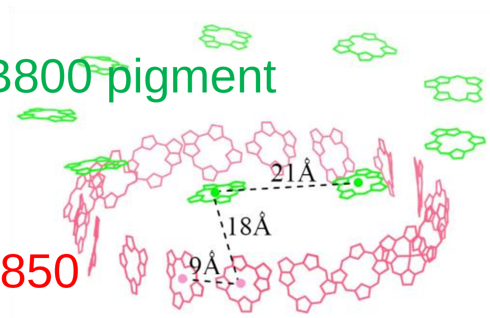
Additional features due to rotational wavepackets in HD (red).



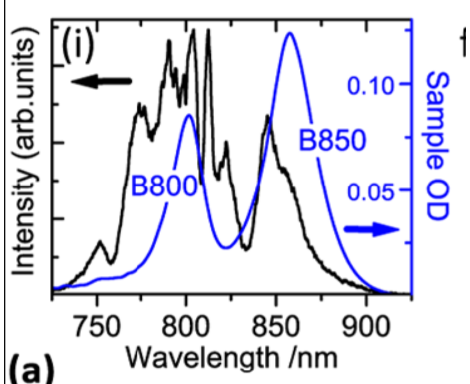
Photosynthetic protein: energy transfers and coherences

UC Dublin, IC, Glasgow U

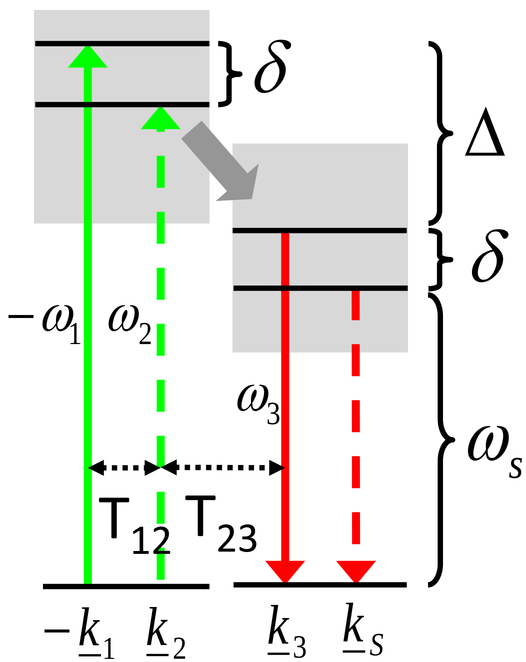
Angular Resolved Coherent (ARC) imaging : New 4-wave mixing method for imaging an arbitrary number of molecular quantum couplings with a single 10fs laser pulse and 2-D CCD camera



Photosynthetic LH2 antenna



10fs laser spectrum over-laps both B800/B850 absorption spectra



ARC RESULTS:

T_{12}=0, T_{23}=0,
ω_1=11740cm^{-1}; ω_2=11620cm^{-1} ,
ω_3=11480cm^{-1} , ω_s=11360cm^{-1}
δ-decay time = 160fs

T_{12}=0, T_{23}= 2.8ps,
ω_1= ω_2=11760cm^{-1} ,
ω_3= ω_s=11360cm^{-1} (population state)

Time-ordered light-matter interaction sequence for B850 pigment of the light harvesting complex II of purple bacteria observed in this experiment. T12 and T23 represent times between light-field interactions

Ian P. Mercer , Phys. Rev. Lett. 102, 57402, 2009