Science & Technology Facilities Council
Central Laser Facility

Ultrafast Materials Science with the Artemis XUV beamline

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

Workshop: 'High Power Lasers' at Conference:
'Diaspora in Cercetarea Stiintifica din Romania',
Academia Romana, Bucuresti, 22nd September 2010

edmond.turcu@stfc.ac.uk; www.clf.stfc.ac.uk

Artemis facility is open to UK and EU research groups

Contents

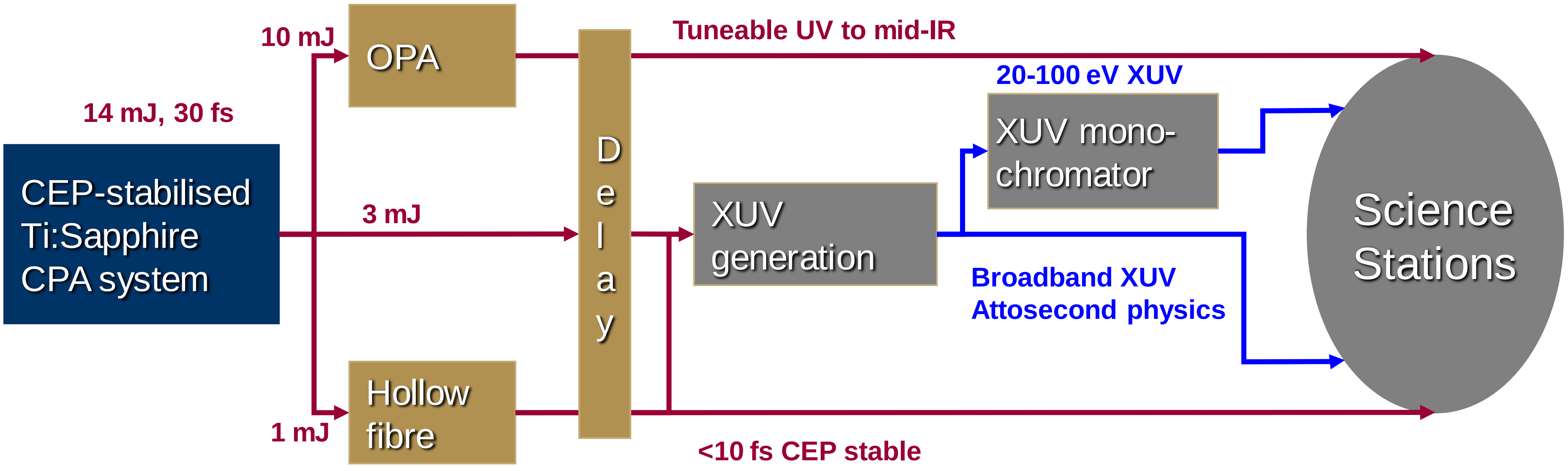
I. Artemis facility

II. Ultrafast time-resolved materials science experiments

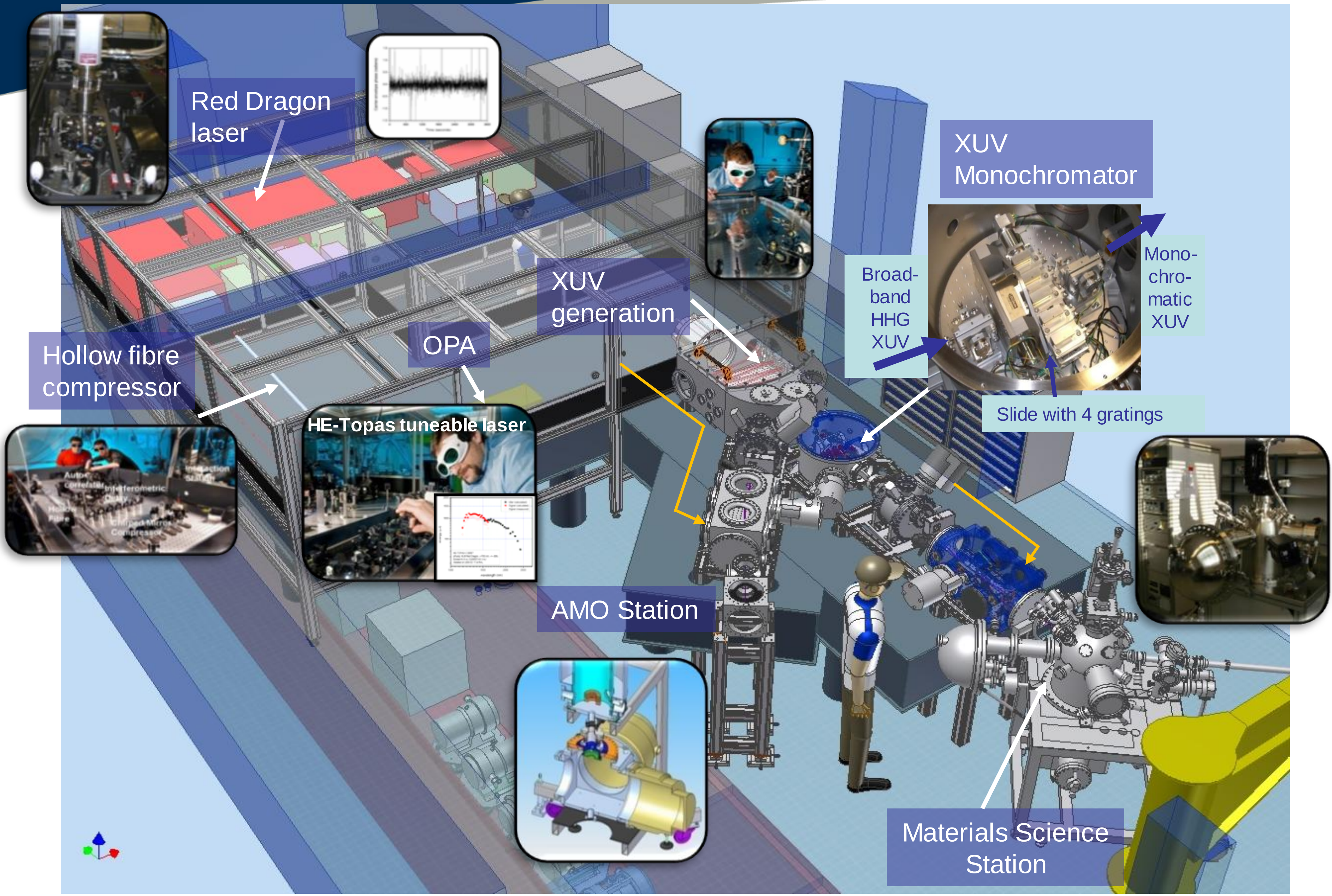
Artemis Facility

- **Combining ultrafast laser beams**
  - 14 mJ, 30 fs, 1 kHz CEP-stabilised drive laser (Red Dragon from KML)
  - 30 fs, tuneable UV to mid-IR
  - <10fs 800nm pulses
  - 30 fs IR
  - All three beams synchronised and independently configurable

- **Two XUV beamlines**
  - Short pulse (10-50 fs) monochromatised XUV
  - Broadband, attosecond harmonic beam
- **Materials science end-station**
- **Atomic & molecular physics end-station**
- **Visiting end-station**



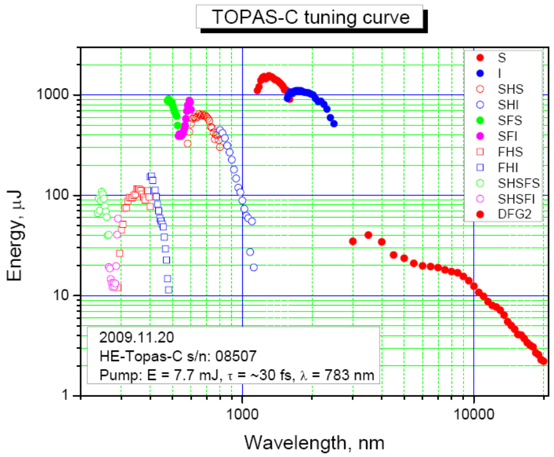
Facility Layout



Beam Parameters

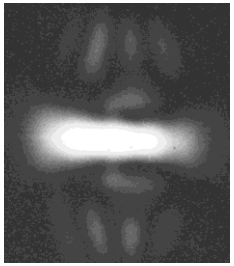
High-Energy TOPAS Tuneable Laser

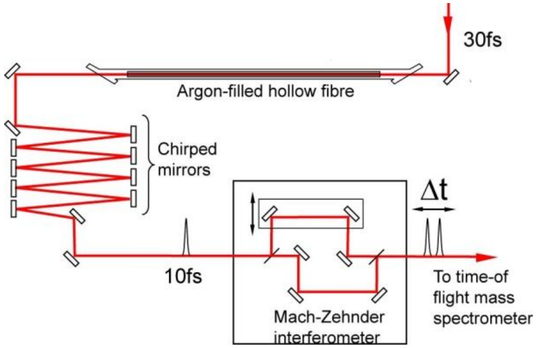
- **Energy/pulse in: 8 mJ**
- **Energy/pulse (signal+idler): 2.5mJ@1300nm**
- **Tuneable:1.2µm -20µm**
- **Pulse duration: ≥35fs**
- **Repetition rate: 1 kHz**
- **HE-TOPAS from 'Light Conversion'**



Hollow fibre compression

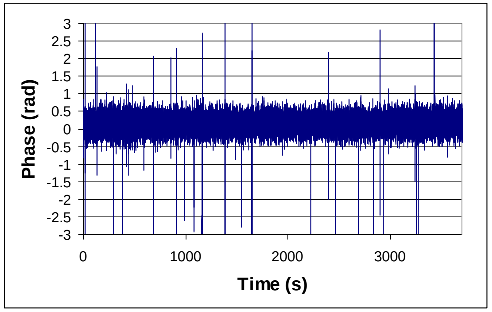
- **7 fs pulse duration measured with FROG for ultra short pulses.**
- **Energy/pulse: 600 µ J**

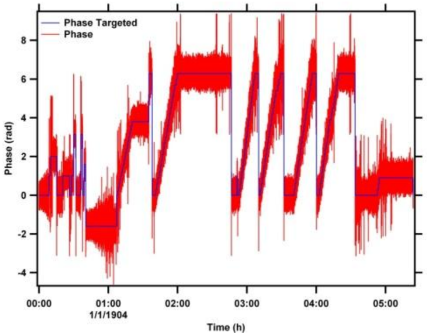




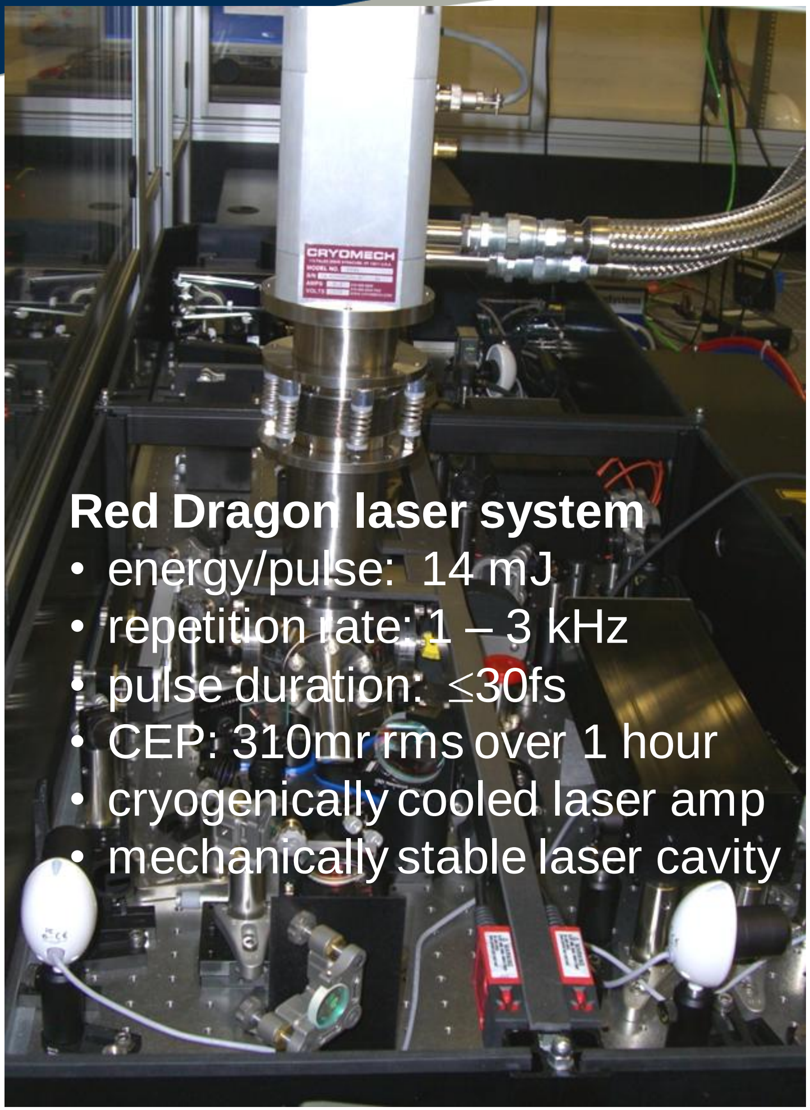
Carrier Envelope Phase stabilised

The 14 mJ multi-stage amplifier system is Carrier Envelope Phase stabilised and we archived in our lab a 340 mrad rms noise with control over eight hours.





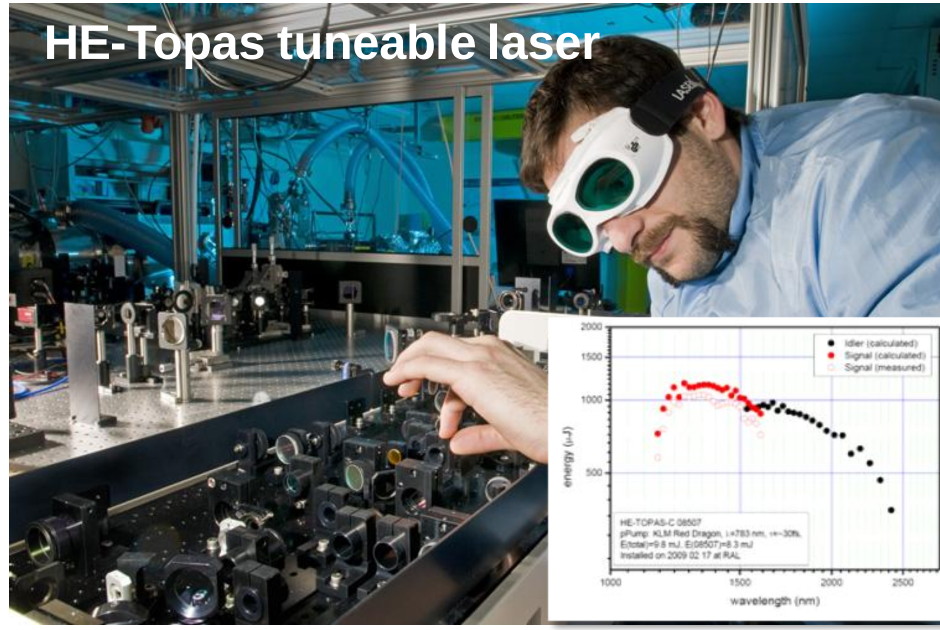
Artemis :Laser Beams



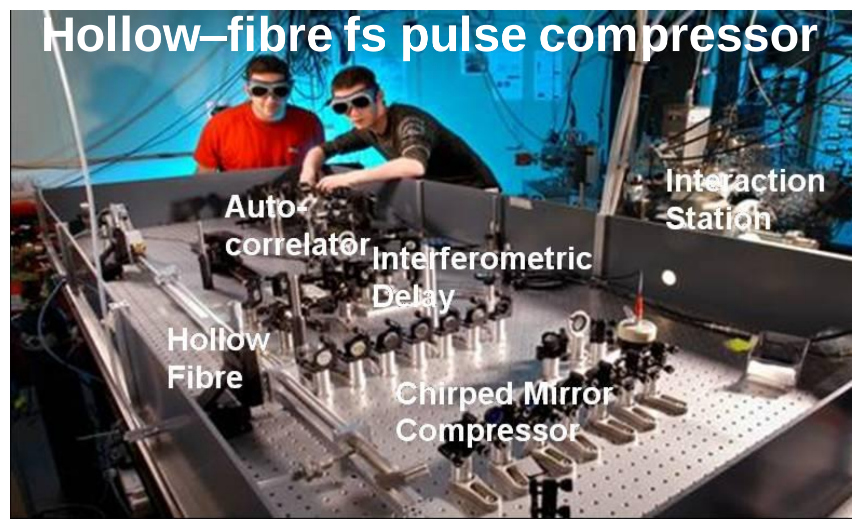
Red Dragon laser system

- 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

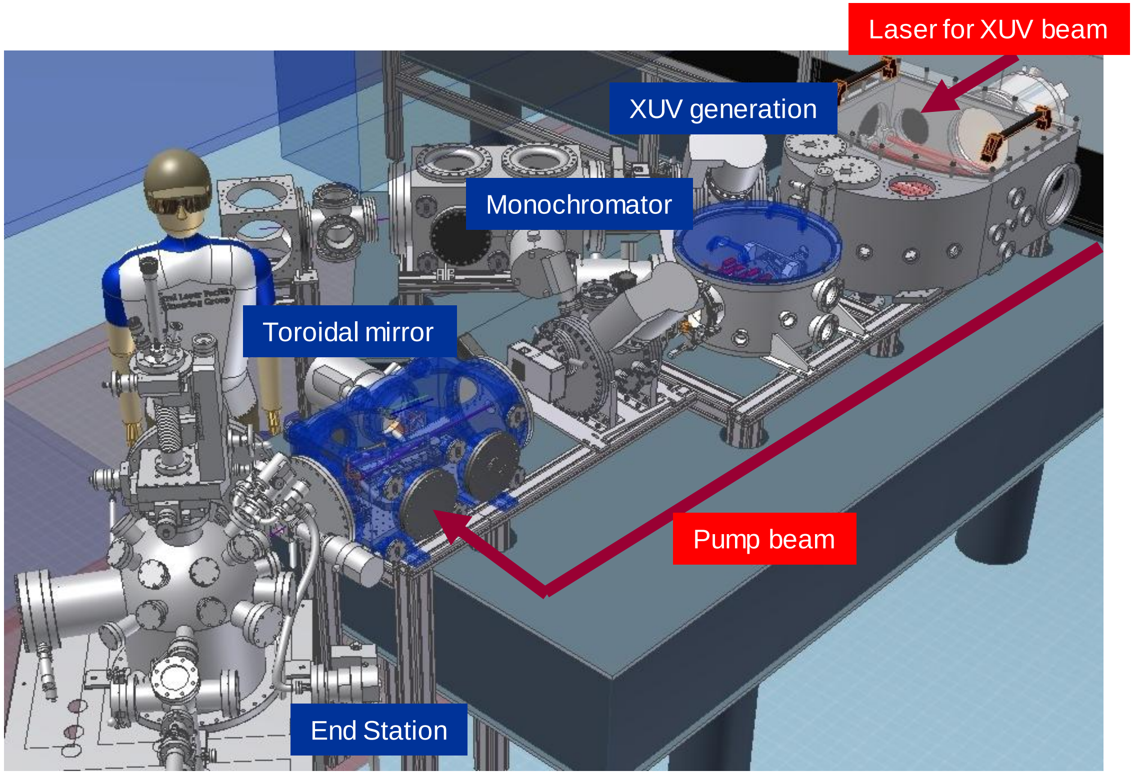
HE-Topas tuneable laser

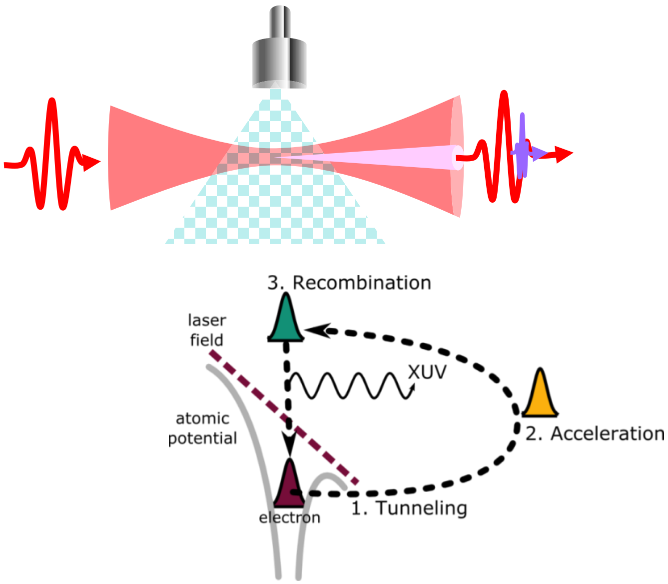


Hollow–fibre fs pulse compressor



XUV monochromatic beamline





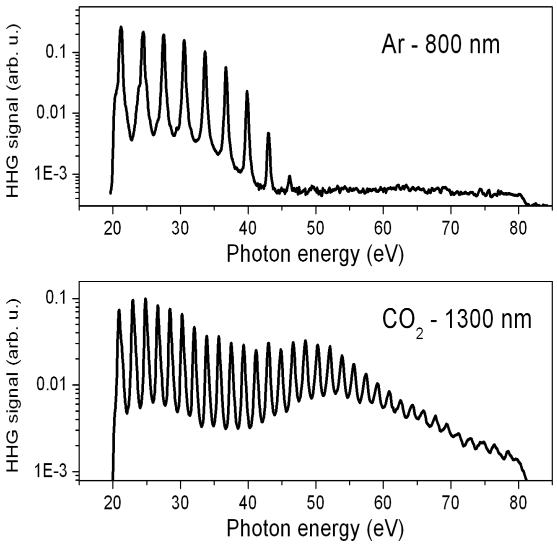
HHG from HE-Topas laser driver: 1300nm, 40fs, 1mJ, 1kHz

Temporal resolution from Pump-Probe with controlled fs time-delay.

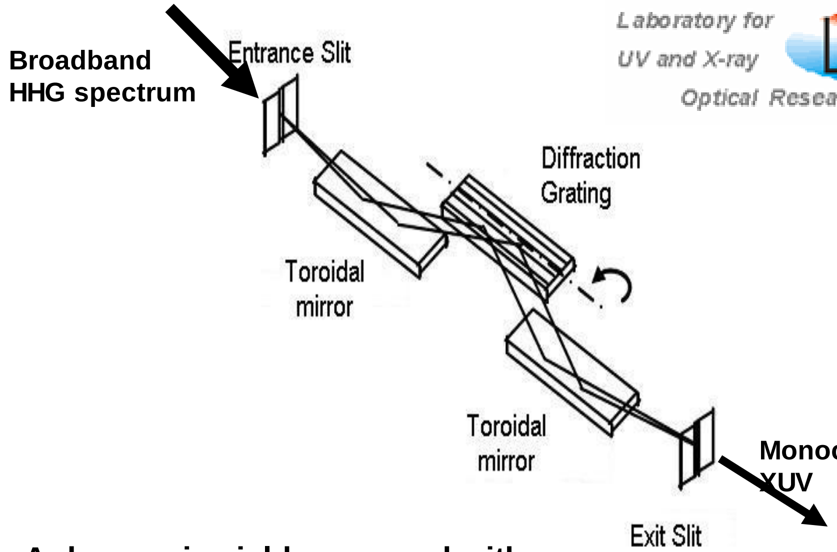
Pump: 800nm or tuneable laser: λ = 0.2-20μm

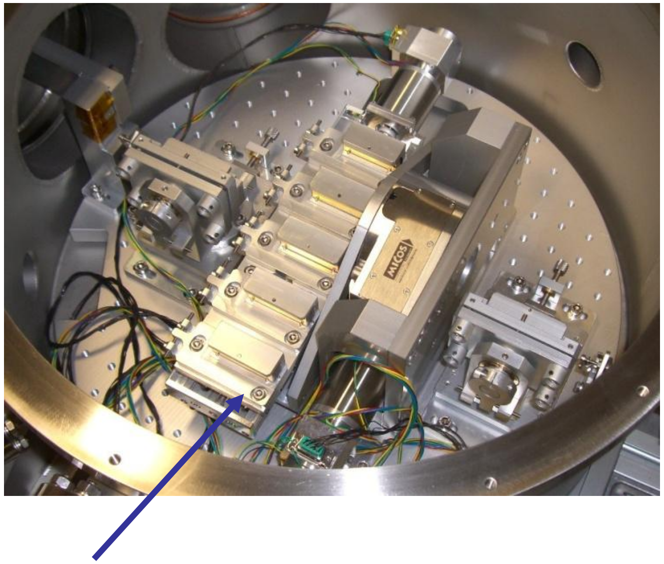
Probe: XUV: hν = 10 – 100eV monochromatic

Time-zero from laser interferometry.



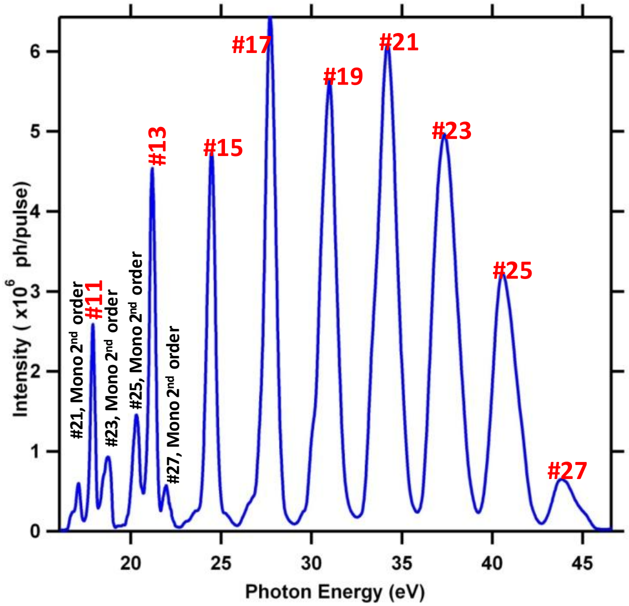
Pulse-length preserving XUV monochromator





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

Ar harmonic yield measured with grating 3 (λ/Δ λ=100)

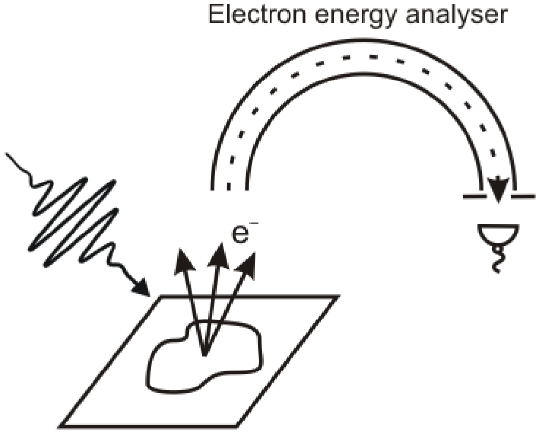


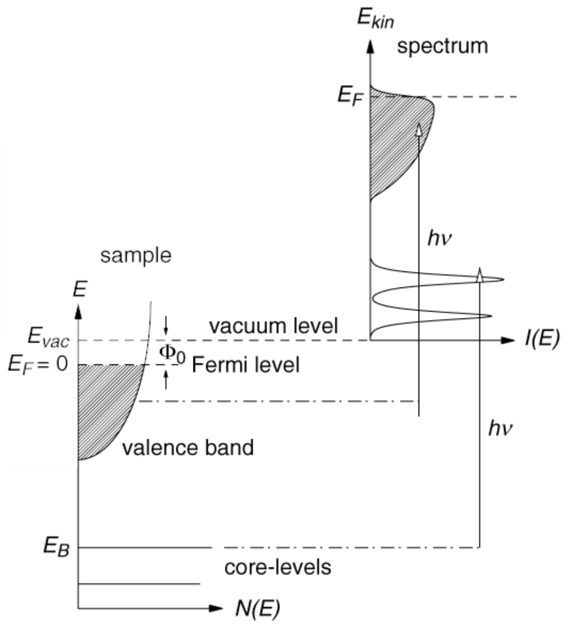
Grating calculated parameters

Grating #	Photon energy range	Pulse duration (40% for reduced flux)	Pulse bandwidth λ/Δ λ
1 (LR)	20-40 eV	~10 fs	~27
2 (LR)	35-100 eV	~10 fs	~23
3 (HR)	20-40 eV	~56 fs	~100
4 (HR)	35-100 eV	~40fs	~70

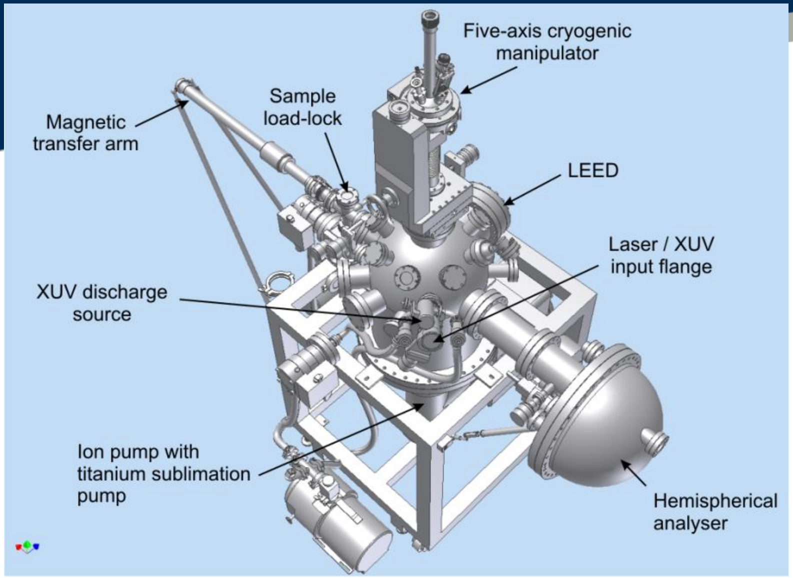
ARPES Analyzer

- Incident photons eject electrons with kinetic energy E=hv− EB−φ
- Allows direct access to electronic (surface) states of material
- **Angle resolved PES (ARPES) can map out the entire Fermi surface and band structure**
- Analyser static image; ARPES angular momentum dispersion mapping. XUV photons probe the whole Brillouin Zone:
- **k = ± 2.0 A-1 for hv=20.9eV compared to**
- **k = ± 0.36 A-1 for hv=6eV**





Materials Science Station



– UHV chamber with mu-metal shielding
– 5 axis manipulator
– He cooling (14K)
– In situ cleaving
– 2D electron analyser (PHOBOIS 100) for energy and angular resolution
– LEED , He lamp
– Load lock, sample transfer

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

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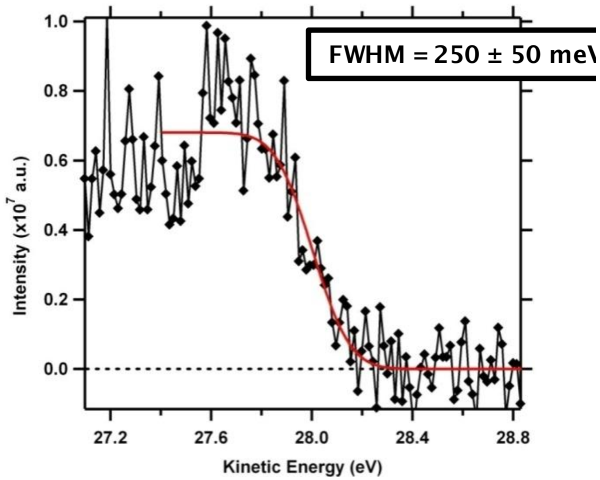
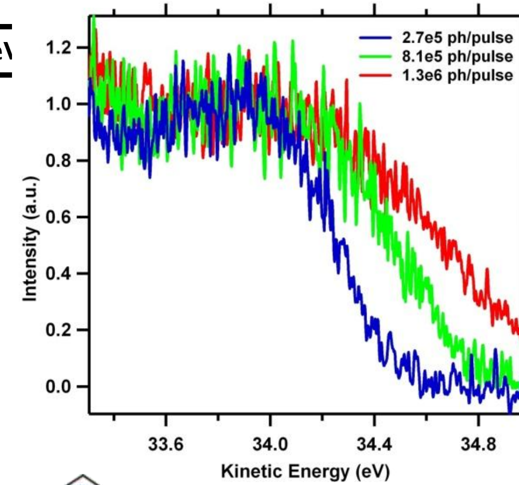
Time and Energy resolution of the XUV beamline

Overall Energy resolution

- PHOIBOS 100 with cooled CCD camera. The best analyser resolution measured on Ag with the He lamp is 25 meV.
- Measured TaS_2 Fermi edge with the 17th harmonic at 28 eV. The *250 meV energy resolution* is limited by the grating resolving power.
- We have carried out systematic measurement to establish the space charge effects limit.

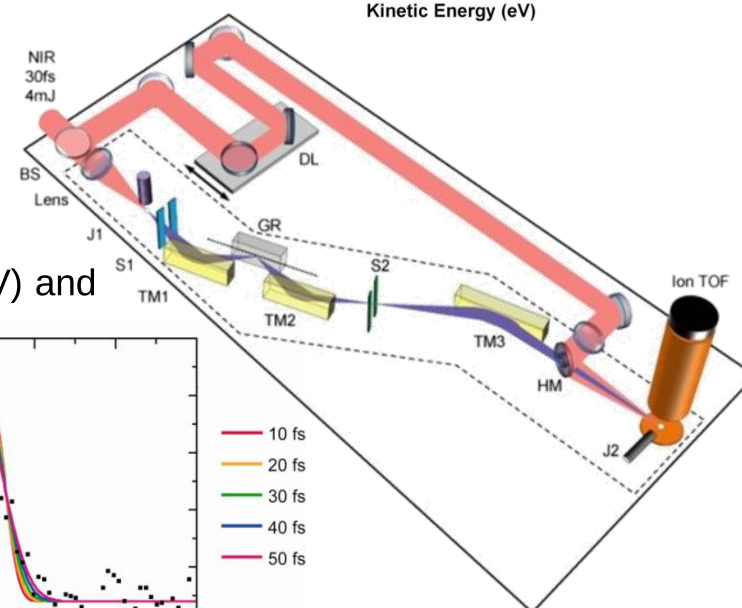


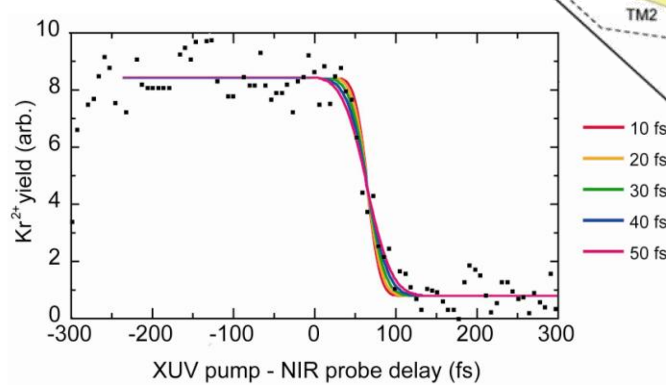


Time resolution

- Kr^{2+} ion yield measured in ion time-of-flight spectrometer
- Change in Kr^{2+} ion yield measured as delay between IR (1.55eV) and 23rd harmonic (35.7 eV) .
- The IR laser pulse duration is 28 fs and
- from the step slope we find an *XUV pulse duration of 24 fs*.
- Resolution Interferometer XUV + IR is < ¼ laser periods or <0.6fs.

Stability is >12hours.





Mott Gap Insulator Dynamics by

TR-ARPES

(Time-Resolved Angular-Resolved-Photoemission-Spectroscopy)

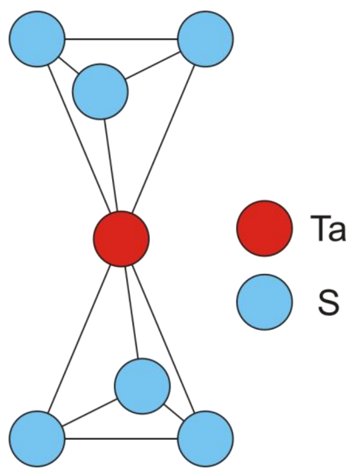
Tantalum disulphide (TaS2)

MPSD
Oxford U

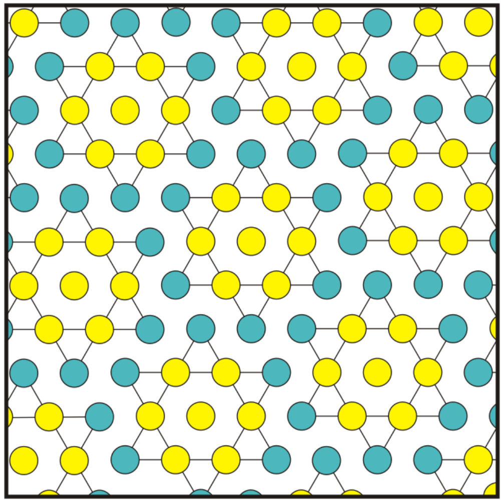
Below 180K

- **TaS_2 is quasi-two-dimensional. Structural unit in Ta plane: 13 atom clusters –'stars' with central atom and 2 rings of 6 Ta atoms. Associated with formation of CDW = charge transfer from from outer ring to inner parts.**
- **Transition Metal to Mott Insulator (100 meV gap). The 13 Ta atom have d^1 orbitals. Cluster has 6 orbitals totally occupied and 7^{th} half filled. the electronic band splits into 2 occupied manifolds (each with 3 orbitals) and a conduction band (uppermost cluster orbital).**
- **At low temperature the lattice contraction CDW amplitude increases reducing overlap between orbitals. At 180 K the energy U necessary of double occupation of one UCO is smaller than W – bandwidth. The metalic state is unstable an electrons localize into the Mott insulating (MI) phase.**

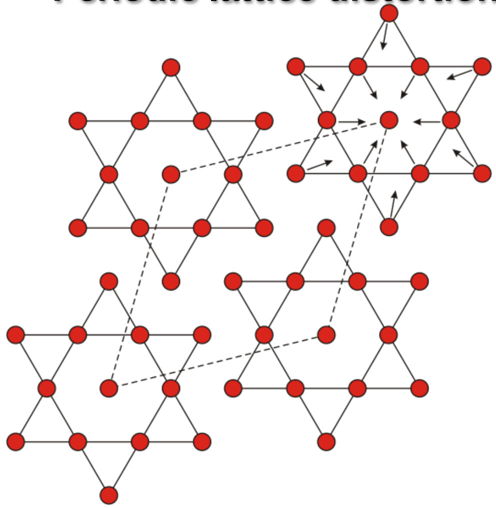
Layered structure

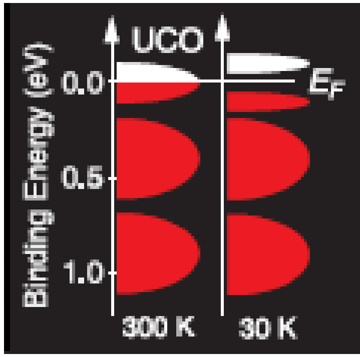


Charge Density Wave



Periodic lattice distortion





→ **The MI can be destroyed by photoinduced excitation and we need a spectroscopic technique to be able to observe the dynamics of the electronic structure. Our aim is to perform a detail study of the electronic dynamics in the momentum space.**

Time-resolved ARPES (our first results)

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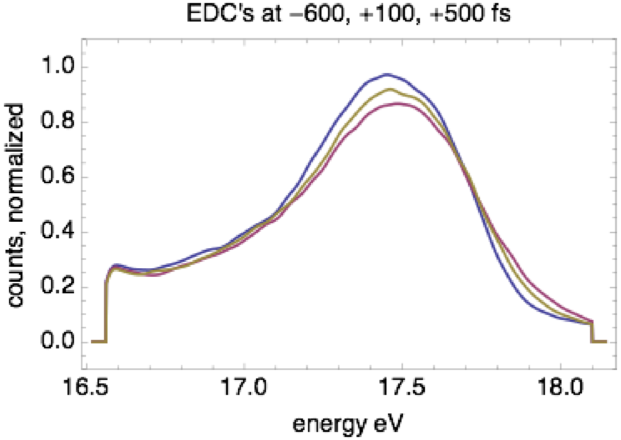
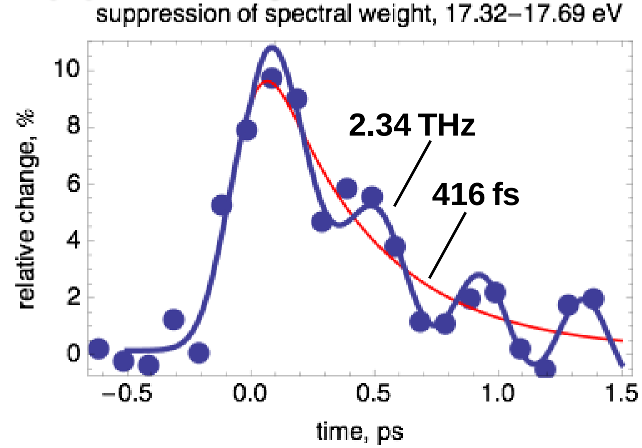
Oxford U

Energy Distribution Curve at different time delays

Pump = 800 nm
Probe = 17.9 eV
T = 14K
Angle = ±18˚
k = ±0.57 Å



Reduction of the Hubbard peak versus pump-probe delay time.

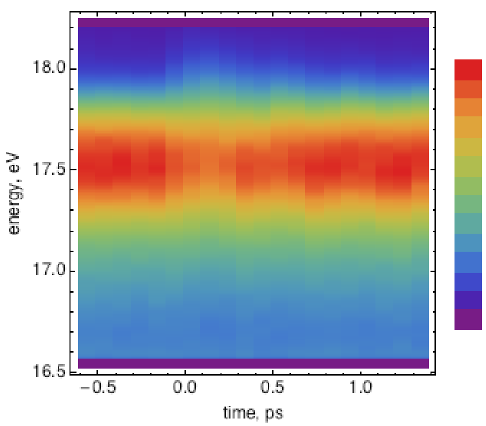


We observe a clear suppression of the spectral weight at the Hubbard peak (17.5 eV) and a collapse of MI gap (17.9 eV). The density of state near the Fermi level is modified by the destruction of the Mott order.

- **At time zero an ultrafast collapse (200fs) of the Mott Insulator gap is observed corresponding to a photoinduced MI-metal phase transition.**
- **The MI phase is recovered on the same time scale as the relaxation of the hot electrons (416 fs lifetime). It is well establish that in TaS2 the MI collapse is driven by the high temperature electrons.**

- **During the pump excitation there is a transfer of charge from the center of the Ta star to the outer rings inducing atomic displacements. The 2.3 THz oscillation observed is attributed to a breathing mode of the star associated to a coherent excitation of the CDW amplitude mode by phonon excitation.**

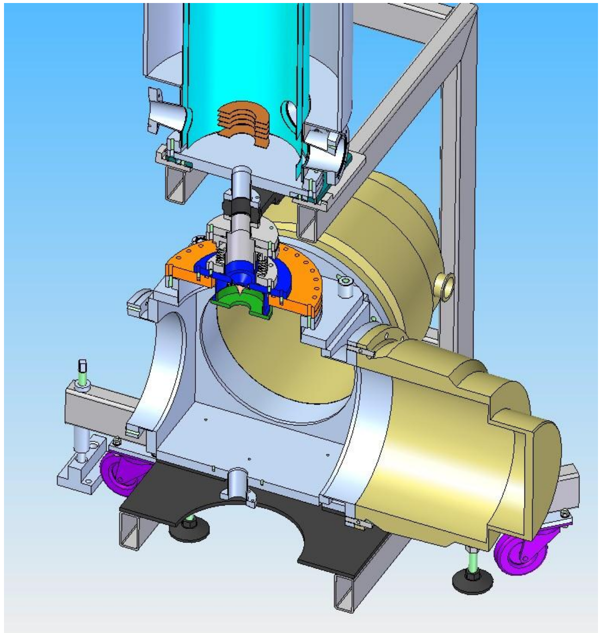
Energy-time intensity map photoelectron intensity



These results agree well with the measurements performed by Perfetti *et al.* at 6 eV photon energy.

L. Perfetti, *et al.*, PRL **97**, 067402 (2006)

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.

Conclusions

Artemis science facility :

- *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:

- *Mot-Gap insulator dynamics with TR-ARPES*

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Contributors

STFC Central Laser Facility: Emma Springate, Edmond Turcu, Cephise Cacho, John Collier, Steve Hook, Toby Strange, Brian Landowski

STFC Daresbury Laboratory: Mark Roper

Swansea University: Will Bryan, Jamie Nemeth

Oxford University: Andrea Cavalleri, Jesse Peterson, Nicky Dean.

MPG Structural Dynamics Hamburg: Andrea Cavalleri, Jesse Peterson, Stefan Keiser, Adrian Cavalieri, Alberto Alberto Simoncig

Diamond Light Source: Sarnjeet Dhesi

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

Central Laser Facility

- Vulcan Petawatt: 400 J in 400fs
  - Upgrade to 10PW: 400J in 40fs
- Gemini Petawatt: 2x15J in 30fs, 3pulses/min
- Artemis: fsec XUV+laser beams, 1-3kHz
- Ultra: fsec laser beams, 10kHz
- Future facilities/proposals
  - HiPER fast ignition fusion test facility (EU).
  - Dipole, diode pumped laser (EU)
  - ELI, extreme light (EU)

Va multumesc pentru atentie!

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Artemis facility is open to UK and EU research groups

XUV Beamline: fsec Resolution

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Swansea University
Prifysgol Abertawe

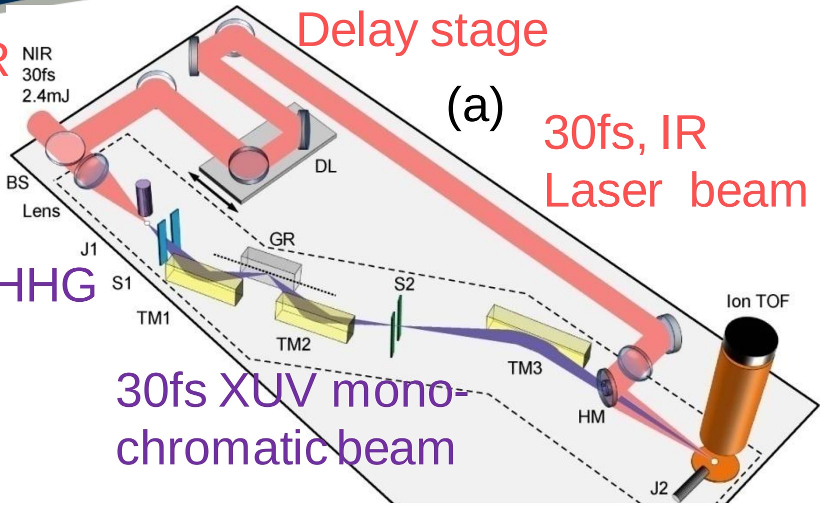
Laboratory for UV and X-ray Optical Research

30fs, IR Laser

Delay stage

Splitter

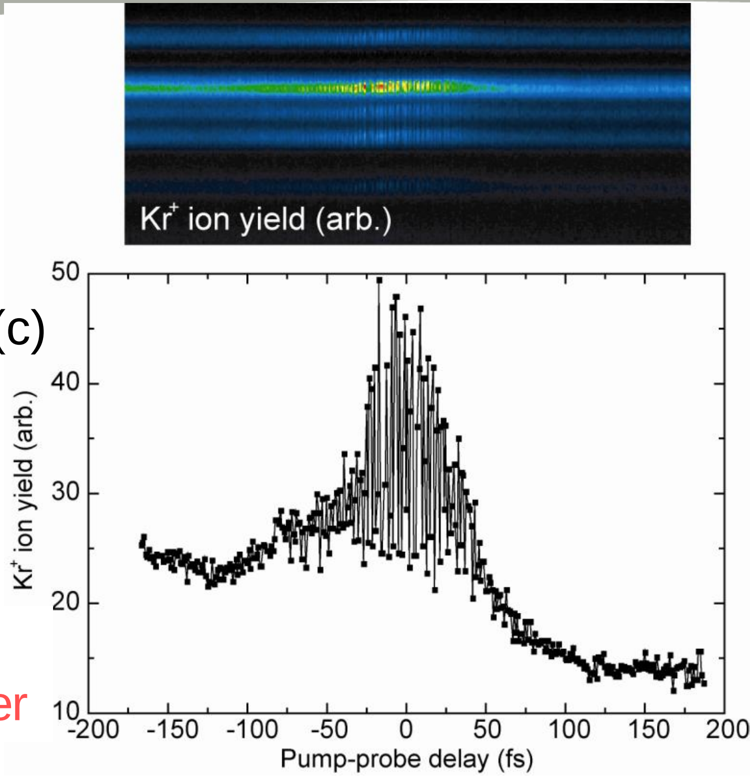
Gas Jet: HHG

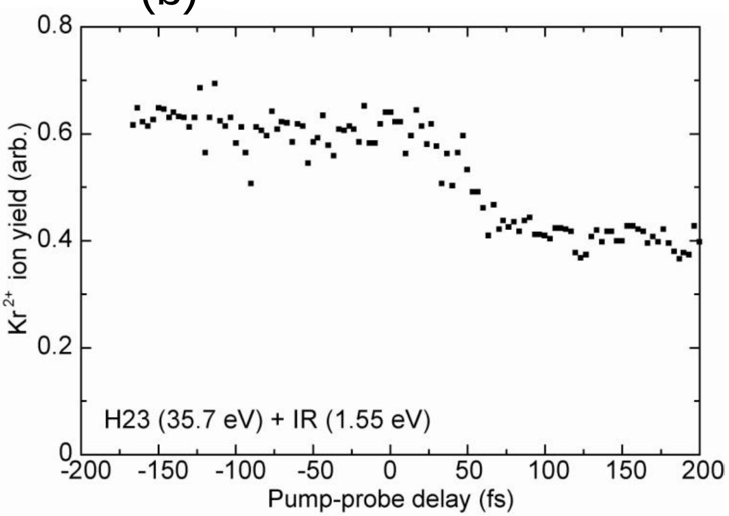


30fs, IR Laser beam

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.

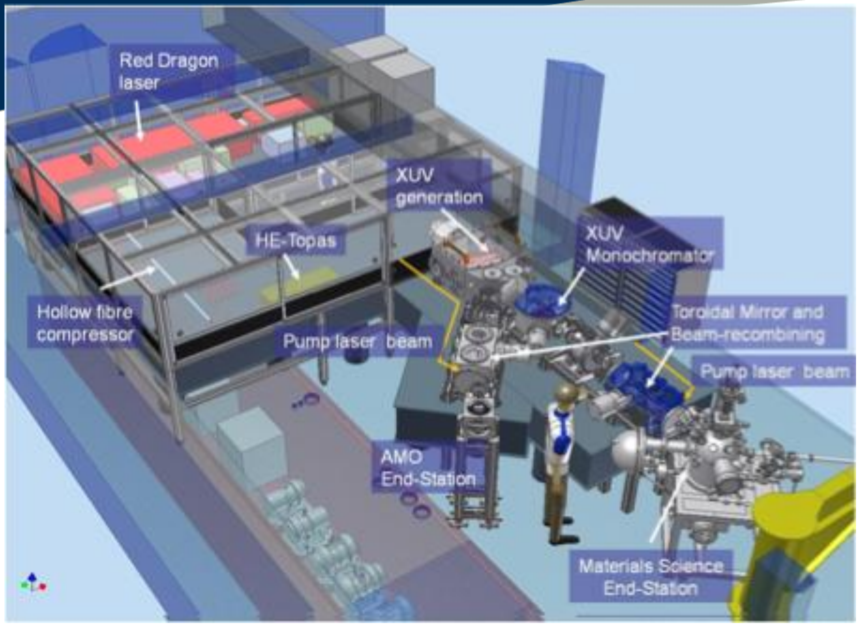
Kr^{2+} ion yield function of temporal delay XUV+IR

(b) XUV pulse duration measured ~ 30fs:
XUV Harmonic 23 (hν=35.7eV) + IR (hν=1.55eV).

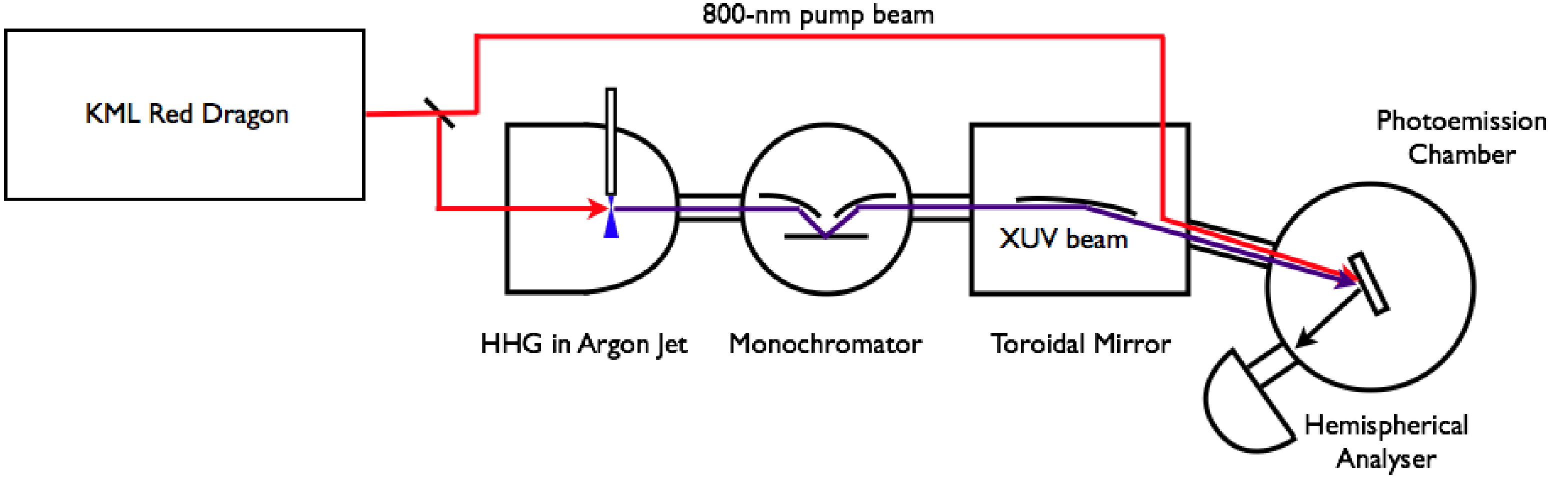
(c) Interferometer resolution is < 1/4 laser periods (0.6fs) XUV broadband + (IR + IR)

TR-ARPES Beamline

MPSD
Oxford U

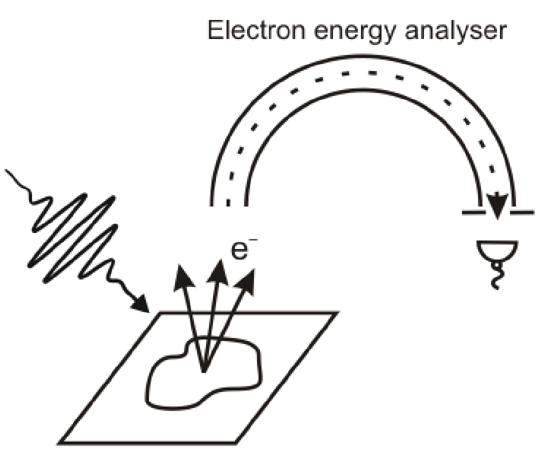


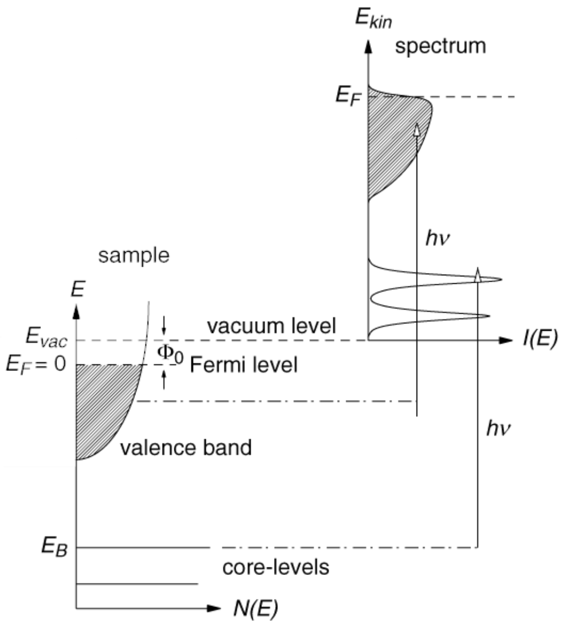
- Rutherford Appleton Laboratory, UK
- Laser-based TR-ARPES with HHG-XUV source
- Initial resolution: 30 fs, 200 meV
- Eventual: 10 fs XUV
- Pump excitation from vis. to MIR
- CEP-stable



ARPES Analyzer

- Incident photons eject electrons with kinetic energy E=hv– EB–φ
- Allows direct access to electronic (surface) states of material
- Angle resolved PES (ARPES) can map out the entire Fermi surface and band structure

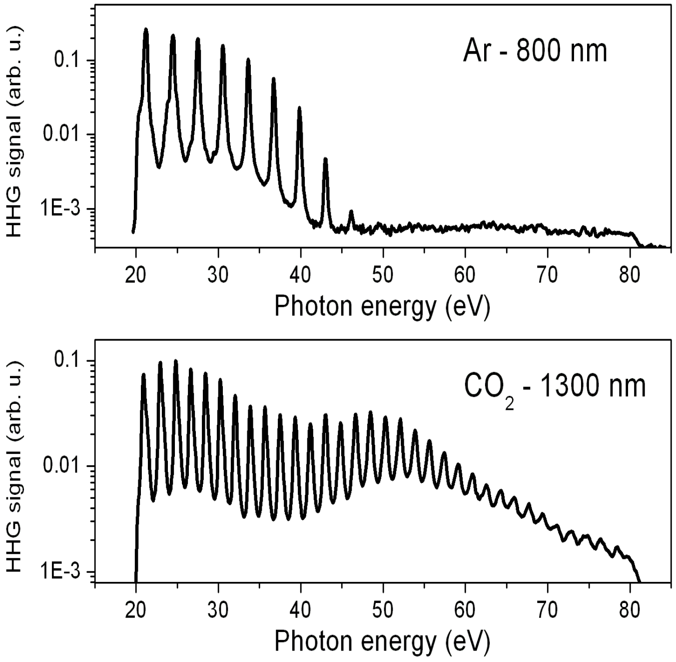




XUV broadband beamline for Atomic and Molecular Physics

XUV-HHG from 800nm or 1300nm

XUV Flat-field spectrometer



Enhanced HHG with 2-Colour Laser

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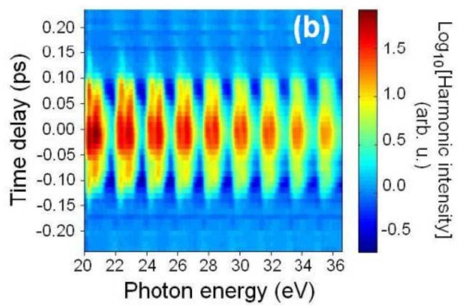
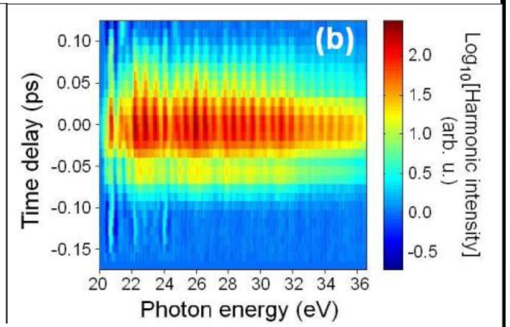
IC London, UC London, U Napoli



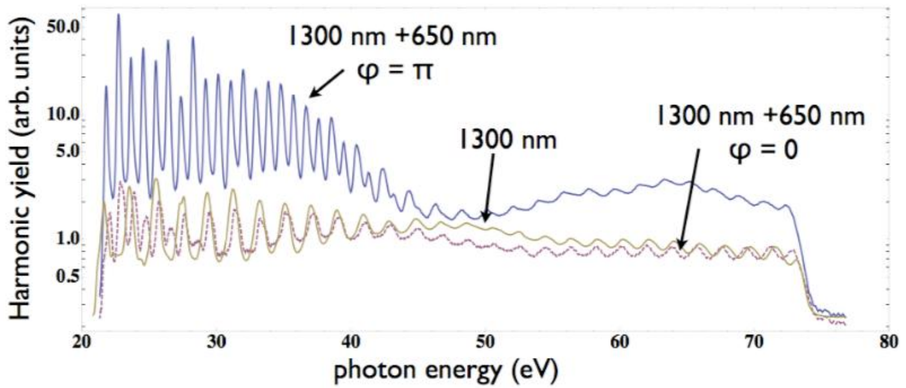


HHG spectra of argon function of delay between λ_1=1300 nm and λ_2=780 nm pulses, parallel polarisation (normalised 1300 nm HHG).

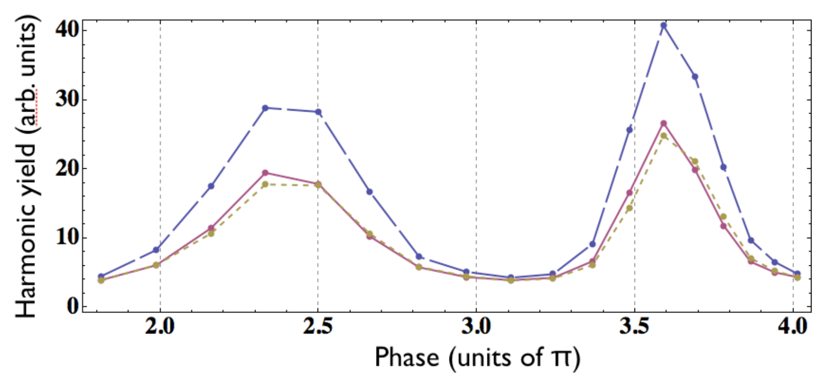
(a) $I_1/I_2=1.5\times10^{14}$ W/cm^2/0.2×10^{14} W/cm^2;

(b) $I_1=I_2=0.5\times10^{14}$ 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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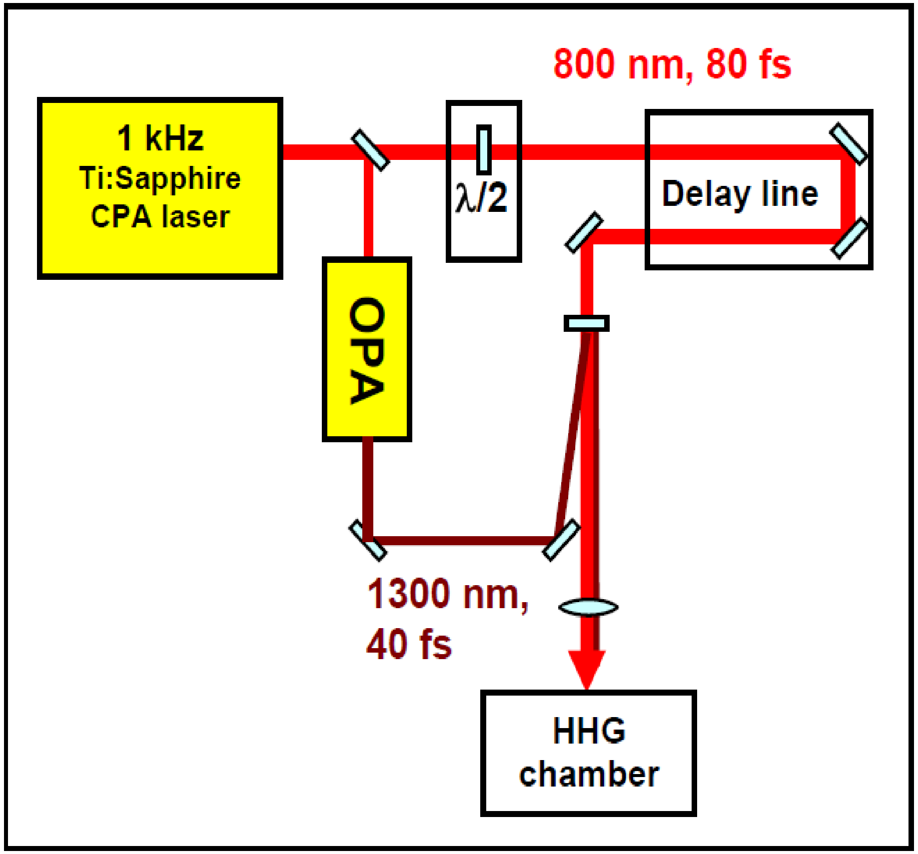
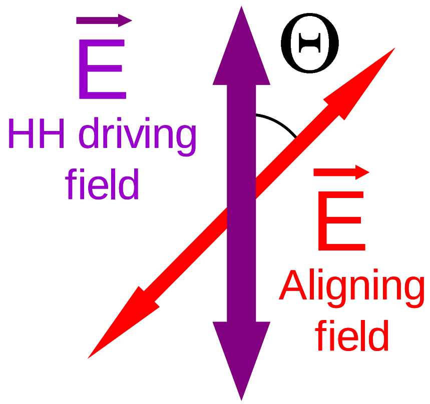
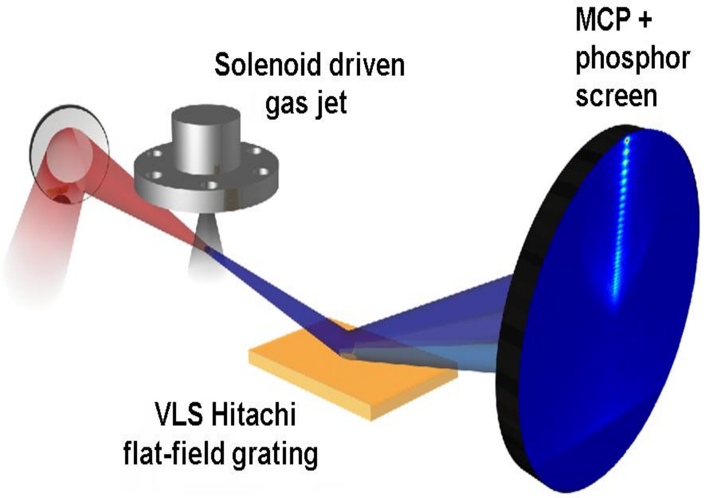
U Napoli

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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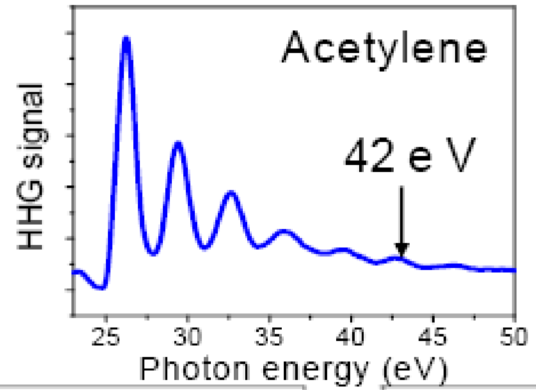
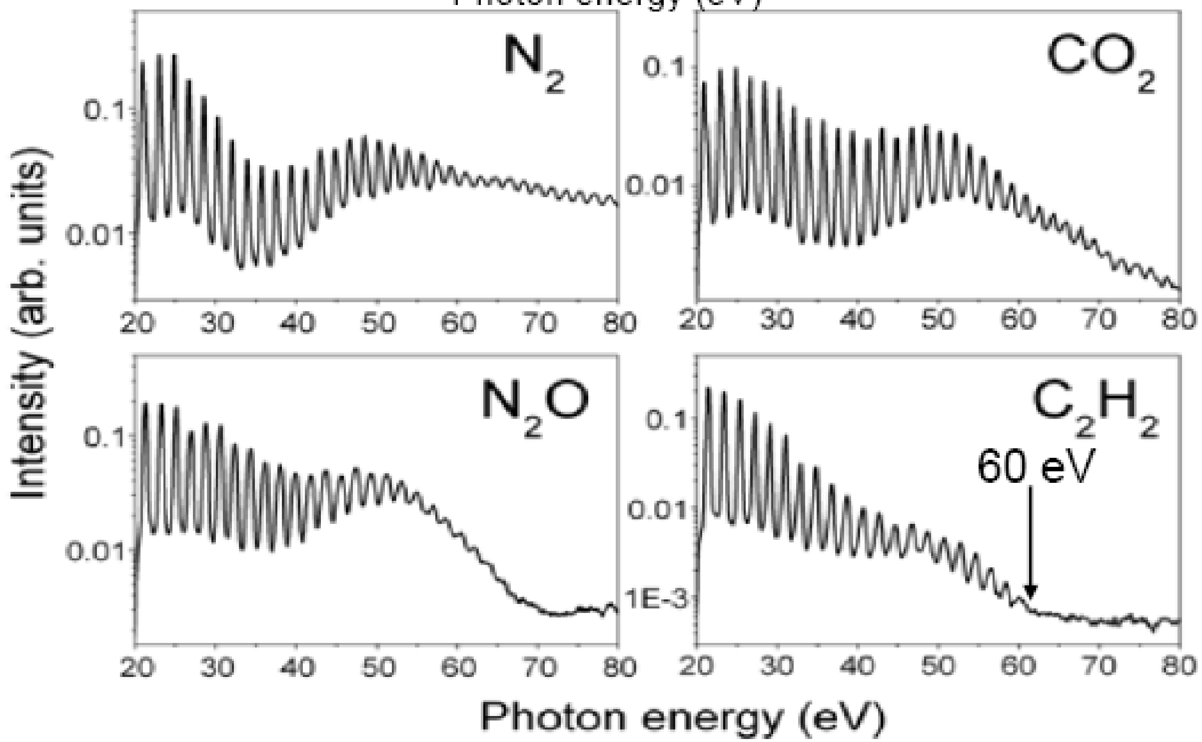
Central Laser Facility

IC London, UC London, U Napoli



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



40 fs 1300nm Un-normalized spectra

Evidence for Structural Interference

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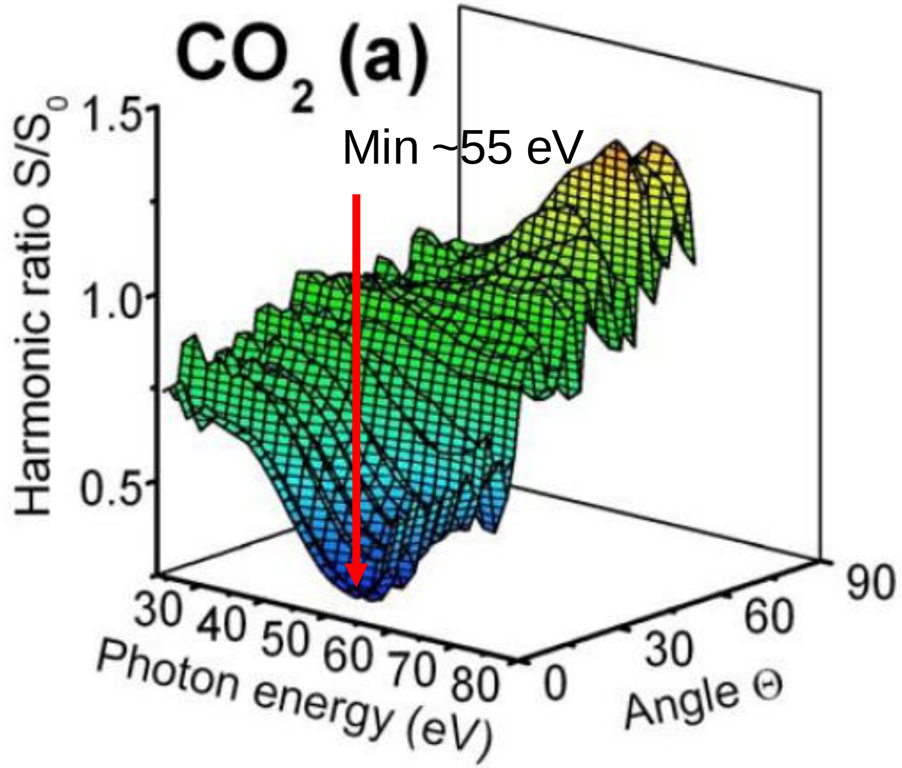
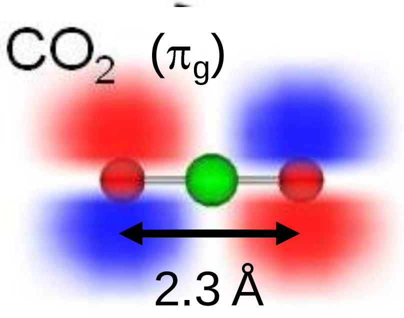


High-Harmonic XUV spectrum modulation in CO_2 molecules shows:

- 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 cosθ Å
(~ 51 eV if θ~ 30^0)
~60 eV if θ~ 40^0

Conclusions

Artemis science facility :

- *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:

- *Mot-Gap insulator dynamics with TR-ARPES in*
- *Molecular structure and dynamics*

Science & Technology Facilities Council
Central Laser Facility

Contributors

STFC Central Laser Facility: Emma Springate, Edmond Turcu, Cephise Cacho, John Collier, Steve Hook, Brian Landowski

STFC Daresbury Laboratory: Mark Roper

Swansea University: Will Bryan, Jamie Nemeth

Oxford University: Andrea Cavalleri, Jesse Peterson, Nicky Dean.

MPG Structural Dynamics Hamburg: Andrea Cavalleri, Jesse Peterson, Stefan Keiser, Adrian Cavalieri, Alberto Alberto Simoncig

Diamond Light Source: Sarnjeet Dhesi

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

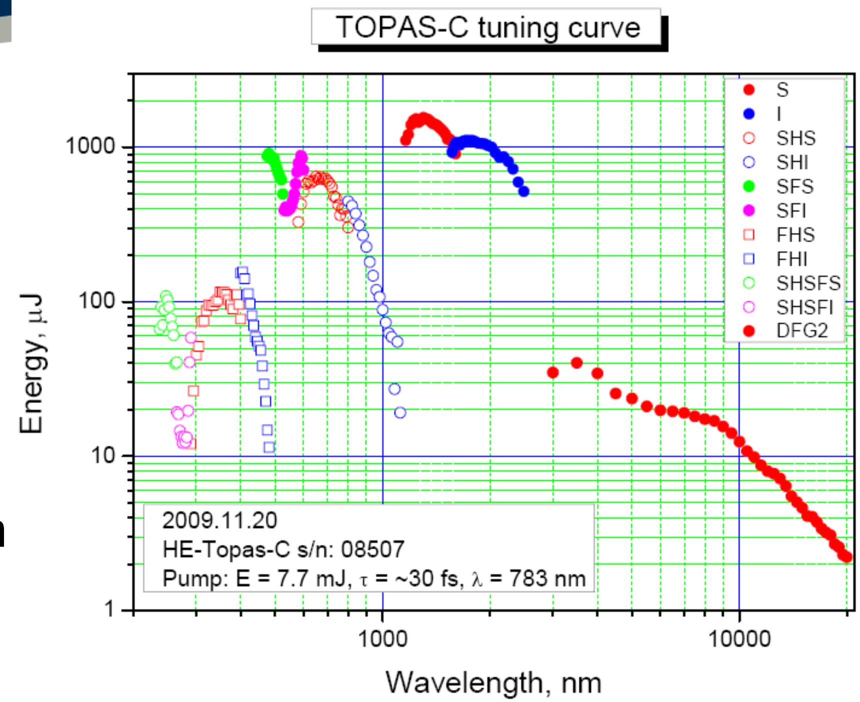
Imperial College London, University College London , Universita di Napoli Collaboration: Jon Marangos, John Tisch, Ricardo Torres La Porte, Thomas Siegel, Yasin C. El-Taha,, Leonardo Brugnera, Jonathan G. Underwood, Immacolata Procino, C. Altucci, R. Velotta,

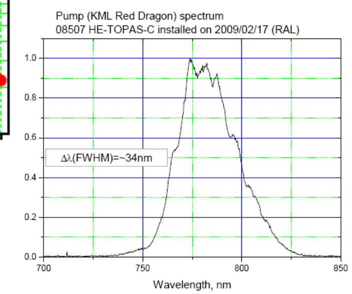
Queens University Belfast: Jason Greenwood, Chris Calvert, Raymond King, Orla Kelly.

HE-TOPAS Tuneable Laser

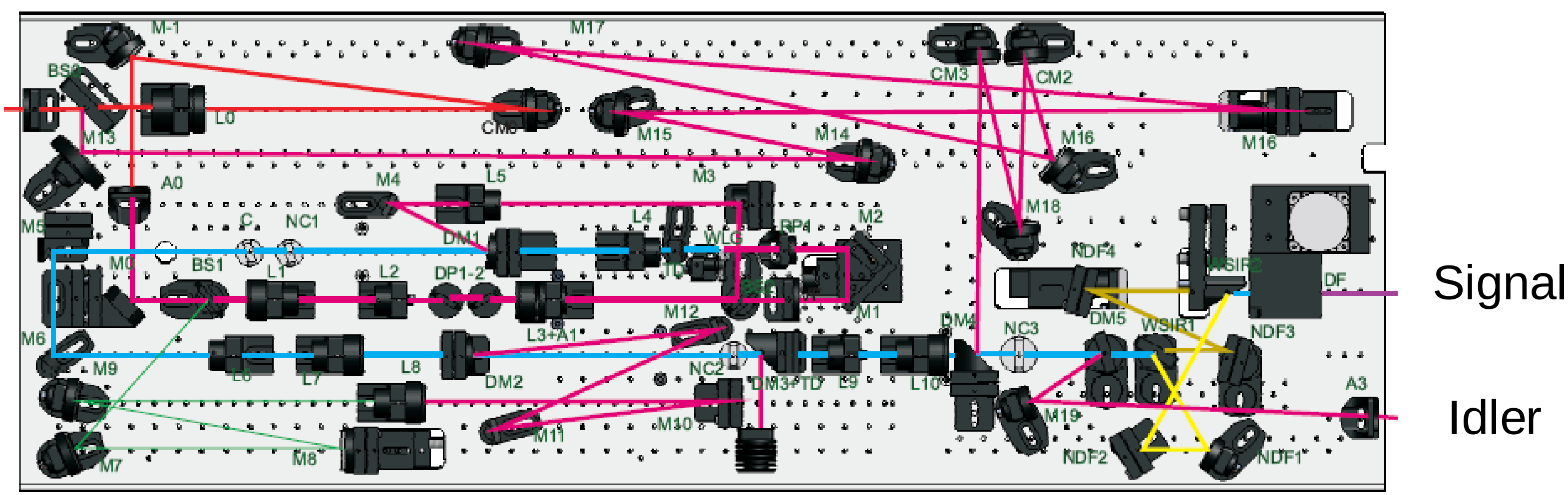
HE-TOPAS OPA from 'Light Conversion'

- Energy/pulse in: 8 mJ
- Energy/pulse (signal+idler): 2.5mJ@1300nm
- Tuneable:0.2µm -20µm
- Will extend to: 0.2µm - 20µm
- Pulse duration: ≥35fs
- Repetition rate: 1 kHz

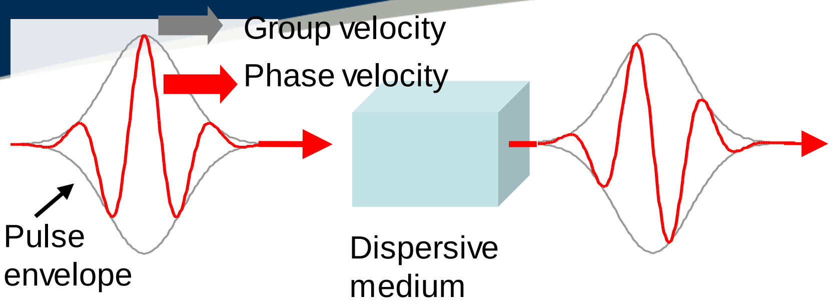




Input:
~8mJ
~30fs
780nm



CEP Stabilisation



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

- 14 mJ Red Dragon laser from KML is first multi-stage amplifier CEP stable
- 310 mrad rms CEP for over one many hours demonstrated in Artemis.
- CEP control demonstrated over five hours