Synchronised attosecond XUV and VUV pulses for pump-probe experiments


Blackett Laboratory, Imperial College London, Prince Consort Road, London SW7 2AZ, UK

Jost Henkel, Manfred Lein
Institut für Theoretische Physik and Centre for Quantum Engineering and Space-Time Research (QUEST), Leibniz Universität Hannover Appelstraße 2, 30167 Hannover, Germany
Charge migration studied by attosecond pump - attosecond probe spectroscopy

1. **Attosecond ionization** in the inner valence region (XUV pump)
2. followed by **VUV probe** inducing single-photon laser-enabled Auger decay SP-LEAD (weak field, background free channel)

Observation of Ultrafast Charge Migration in an Amino Acid (phenylalanine)
Louise Belshaw, Francesca Calegari, Martin J. Duffy, Andrea Trabattoni, Luca Poletto, Mauro Nisoli, and Jason B. Greenwood

Extreme-ultraviolet pump-probe studies or one-femtosecond-scale electron dynamics
P. Tzallas, E. Skantzakis, L.A.A. Nikolopoulos, G. D. Tsakiris and D. Charalambidis

Attosecond pump – attosecond probe with a choice of photon energies

Glycine

1. **XUV+XUV**
2. **VUV+IR**
3. **Strong field probe**

High energy laser systems

Observation of Ultrafast Charge Migration in an Amino Acid (phenylalanine)
Louise Belshaw, Francesca Calegari, Martin J. Duffy, Andrea Trabattoni, Luca Poletto, Mauro Nisoli, and Jason B. Greenwood

Extreme-ultraviolet pump-probe studies or one-femtosecond-scale electron dynamics
P. Tzallas, E. Skantzakis, L.A.A. Nikolopoulos, G. D. Tsakiris and D. Charalambidis

Attosecond pump – attosecond probe with a choice of photon energies

Glycine

1. **XUV+XUV**
2. **VUV+IR**
3. **Strong field probe**

High energy laser systems

Observation of Ultrafast Charge Migration in an Amino Acid (phenylalanine)
Louise Belshaw, Francesca Calegari, Martin J. Duffy, Andrea Trabattoni, Luca Poletto, Mauro Nisoli, and Jason B. Greenwood

Extreme-ultraviolet pump-probe studies or one-femtosecond-scale electron dynamics
P. Tzallas, E. Skantzakis, L.A.A. Nikolopoulos, G. D. Tsakiris and D. Charalambidis

Attosecond pump – attosecond probe with a choice of photon energies

Glycine

1. **XUV+XUV**
2. **VUV+IR**
3. **Strong field probe**

High energy laser systems

Observation of Ultrafast Charge Migration in an Amino Acid (phenylalanine)
Louise Belshaw, Francesca Calegari, Martin J. Duffy, Andrea Trabattoni, Luca Poletto, Mauro Nisoli, and Jason B. Greenwood

Extreme-ultraviolet pump-probe studies or one-femtosecond-scale electron dynamics
P. Tzallas, E. Skantzakis, L.A.A. Nikolopoulos, G. D. Tsakiris and D. Charalambidis

Attosecond pump – attosecond probe with a choice of photon energies

Glycine

1. **XUV+XUV**
2. **VUV+IR**
3. **Strong field probe**

High energy laser systems

Observation of Ultrafast Charge Migration in an Amino Acid (phenylalanine)
Louise Belshaw, Francesca Calegari, Martin J. Duffy, Andrea Trabattoni, Luca Poletto, Mauro Nisoli, and Jason B. Greenwood

Extreme-ultraviolet pump-probe studies or one-femtosecond-scale electron dynamics
P. Tzallas, E. Skantzakis, L.A.A. Nikolopoulos, G. D. Tsakiris and D. Charalambidis

Attosecond pump – attosecond probe with a choice of photon energies

Glycine

1. **XUV+XUV**
2. **VUV+IR**
3. **Strong field probe**

High energy laser systems

Observation of Ultrafast Charge Migration in an Amino Acid (phenylalanine)
Louise Belshaw, Francesca Calegari, Martin J. Duffy, Andrea Trabattoni, Luca Poletto, Mauro Nisoli, and Jason B. Greenwood

Extreme-ultraviolet pump-probe studies or one-femtosecond-scale electron dynamics
P. Tzallas, E. Skantzakis, L.A.A. Nikolopoulos, G. D. Tsakiris and D. Charalambidis

Attosecond pump – attosecond probe with a choice of photon energies

Glycine

1. **XUV+XUV**
2. **VUV+IR**
3. **Strong field probe**

High energy laser systems

Observation of Ultrafast Charge Migration in an Amino Acid (phenylalanine)
Louise Belshaw, Francesca Calegari, Martin J. Duffy, Andrea Trabattoni, Luca Poletto, Mauro Nisoli, and Jason B. Greenwood

Extreme-ultraviolet pump-probe studies or one-femtosecond-scale electron dynamics
P. Tzallas, E. Skantzakis, L.A.A. Nikolopoulos, G. D. Tsakiris and D. Charalambidis
- **Isolated Attosecond pulses in the XUV at 100eV**

- **Isolated Attosecond (~1/sub-fs) pulses in the VUV (15-20eV)**

- Isolated Attosecond pulses in the harder XUV
  - Water window: 280-530eV between Carbon-K and Oxygen-K
1.5-cycle laser source

- Up to 0.6 mJ in 3.5 fs
- Further improvements in progress:
- Larger core fibre $\Rightarrow$ $\sim1.5$ mJ anticipated

- W. Okell et al. *OL* 38, 3918 (2013)
Collinear VUV & XUV attosecond beamline

- pump-probe studies require
- **stable delay** between the two pulses  
  *(with attosecond precision)*

=> **Collinear geometry**
- Common path interferometer
- no delay jitter
- no active locking required

Collinear generation of ultrashort UV and XUV pulses
Spatio-Spectral characterisation of VUV & XUV

- Gas jet z-scans, flat field spectrometer

- XUV cutoff phasematched at specific (downstream) z-position
- VUV positioning is less stringent
- first VUV jet, then XUV jet

VUV 15 to 25 eV
Spatio-Spectral characterisation of VUV & XUV

- 'VUV' 15 to 25 eV
- 'XUV' 50 to 140 eV range

- XUV cutoff not disturbed by VUV
- Absorption of VUV generated in jet1 in XUV jet2
VUV spectral bandpass filter options

transmission of 200nm of In Sn Ge Al Si Zr Mo

- In
- Sn
- Ge
- Al
- Si
- Zr
- Mo

Transmission (%)

Energy (eV)

10 20 30 40 50 60 70 80 90 100 110 120

- 14eV
- 20eV
- 25eV
VUV pulse duration prediction from simulations

- **TDSE calculations** from Jost Henkel, Manfred Lein group
  - Model **Kr atom** for HHG, s, and **px ground state**, softcore potential

![Graphs showing energy vs. time for different elements](image)

- Indium: $939\text{as}$
- Tin: $501\text{as}$
Attosecond streaking

- Measure VUV pulse durations
- Confirm that XUV pulse is preserved with VUV jet in operation
Attosecond Streaking

\[
a(\vec{v}, \tau) = -i \int_{-\infty}^{+\infty} e^{i\phi_{IR}(t)} \vec{d}_p \vec{E}(t - \tau) e^{i(W + I_p)} dt
\]

\[
\phi_{IR}(t) = -\int_{t}^{+\infty} \vec{v} \cdot \vec{A}(t') + \frac{1}{2} \vec{A}^2(t') dt'
\]

Electric field of streaking pulse!
Streaking with varying XUV pulse duration
Confirm that XUV pulse is preserved with VUV jet in operation.
XUV streaking

- VUV OFF
- VUV ON

Energy (eV)

Intensity (arb. u.)

Time (fs)

Energy (eV)

Intensity (arb. u.)

Phase (°)

271±25as
257±21as

XUV-NIR
VUV-NIR
XUV-VUV

2-part focusing mirror
delay
time-of-flight electron spectrometer
effusive gas target
VUV streaking

- Measure VUV pulse duration
- Confirm it survives the XUV jet
VUV streaking 20eV
VUV streaking 15eV
Absolute photon flux measurement

- Sodium salicylate (Aspirin): flat QE response from 300 to 30nm
- Fluorescence in blue -> detection with PMT
- Cross-calibration with 266nm from laser / NIST traceable powermeter head

1) calibration

2) measurement

([Graphs and diagrams showing photon flux measurement process])
Absolute photon flux measurement results

- Sodium salicylate (Aspirin): flat QE response from 300 to 30nm
- Fluorescence in blue -> detection with PMT
- Cross-calibration with 266nm from laser / NIST traceable powermeter head

$10^8 \text{ ph/pulse} = 0.5 \text{ nJ}$
(at generation)
$10^6 \text{ photons/pulse on target}$
New light source capabilities for attosecond pump-probe experiments

- **XUV** pulse is preserved **95 eV, 266 as**
- **VUV** pulse at **14 eV, 1.7 fs** (0.94 fs TDSE)
- **VUV** pulse at **20 eV, 590 as** (501 as TDSE)
- **3.5 fs IR** pulse

- **VUV+XUV** pulses
  - produced collinear & simultaneously
  - Common path interferometer
  - No delay jitter between IR/XUV/VUV
  - ready for pump-probe experiments!

- Improvements:
  - HCF upgrade, 1.5mJ expected
  - 2-colour gating to improve VUV pulse contrast
Acknowledgements:
Davide Fabris
Daniel Walke
Paloma Matia Hernando
William Okell
Thomas Barillot
Peter Knight
Jon Marangos
John Tisch

Jost Henkel
Manfred Lein