European XFEL

Workshop

Attosecond to Few-Femtosecond Ultrafast Science at Future FELs
(AsToFewFs@FutureFELs)

28 June – 30 June 2021
Venue: Online (Zoom)

Session VI: Charge transfer and charge migration

Francesca Calegari, DESY, Universität Hamburg

Speakers:

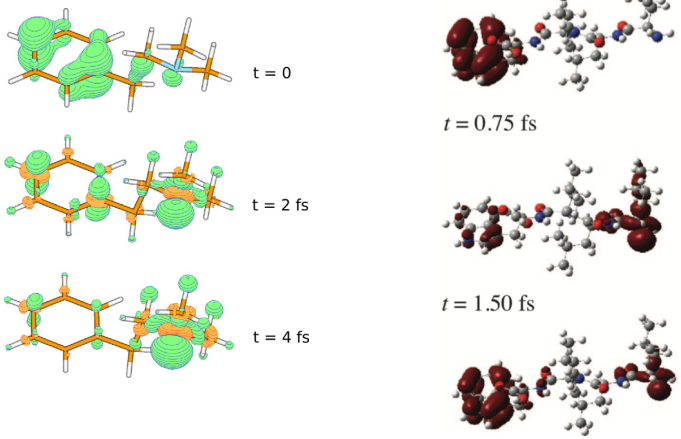
Jon Marangos
Imperial College London

Françoise Remacle
University of Liège

Valérie Blanchet
CELIA

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES DESY. UH CFEL SCIENCE

Charge migration & attochemistry



S. Lünnemann et al., Chemical Physics Letters 450, 232 (2008)

L. Cederbaum, J. Zobeley, Chem. Phys. Lett. 307, 205 (1999)

F. Remacle, R. Levine, PNAS 103, 6793 (2006)

A. Kuleff, L. Cederbaum, Chem. Phys. 338, 320 (2007)

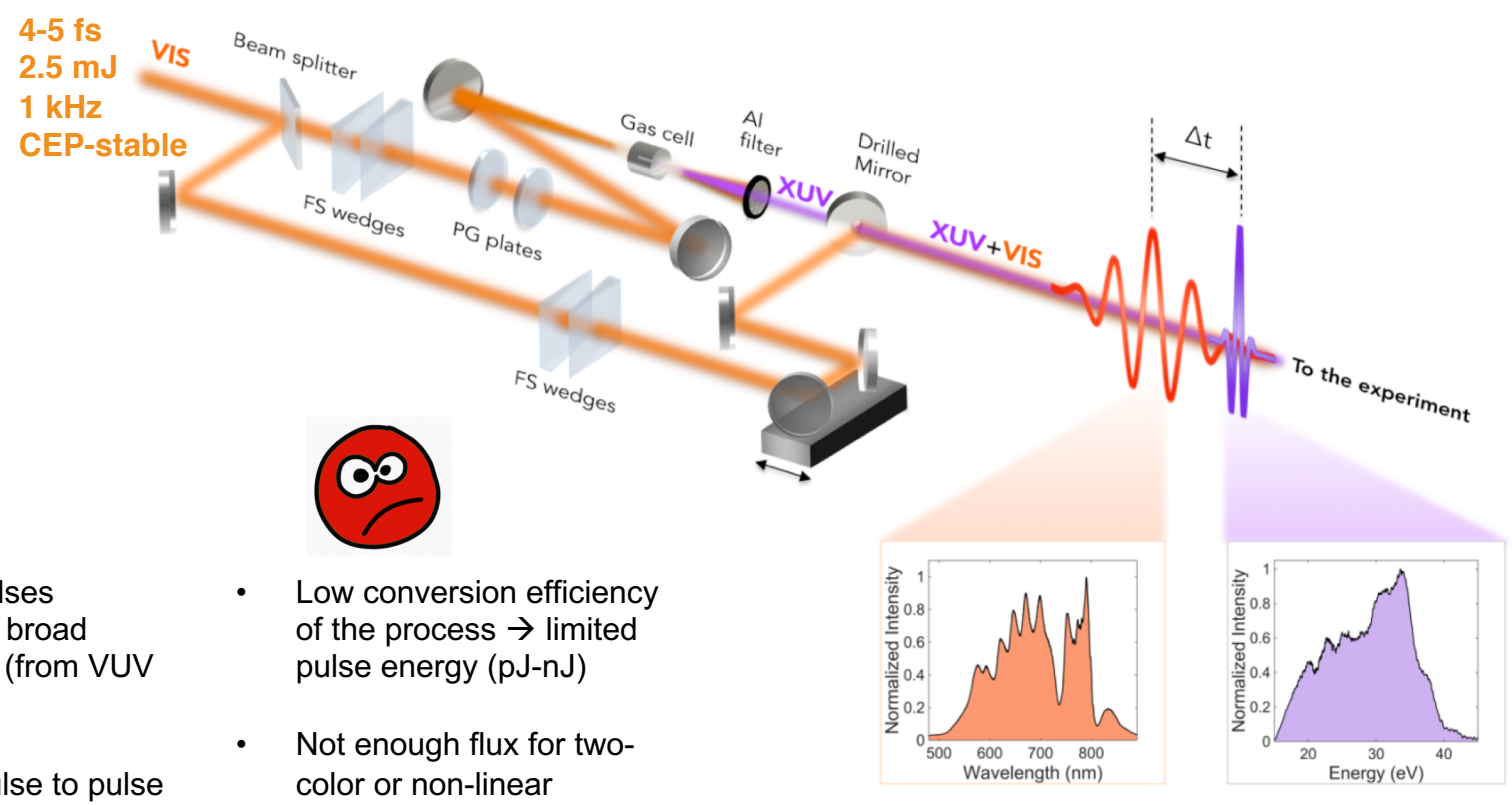
Charge migration: purely electronic (atto/few-femto)

Charge transfer: mediated by nuclei (several femto and more)

Is it possible to drive the charge on the attosecond/ few-femtosecond time scale?

Can we control the photochemistry by acting at this extreme time scale? Is attochemistry possible?

Table-top attosecond sources



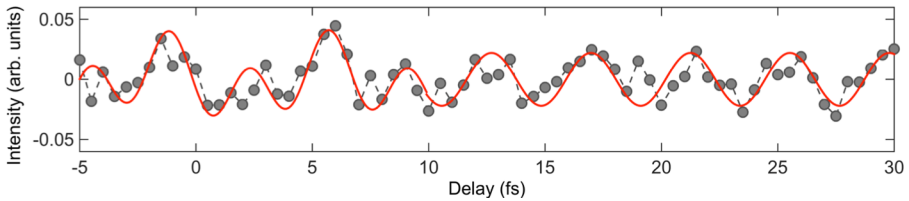
- Attosecond pulses generated in a broad spectral range (from VUV to soft-x)
- Exceptional pulse to pulse stability
- Attosecond synchronization with a second laser pulse

- Low conversion efficiency of the process → limited pulse energy (pJ-nJ)
- Not enough flux for two-color or non-linear experiments in the soft-x
- Limited photon energy (up to the end of the water window)
- Not enough photon flux for imaging

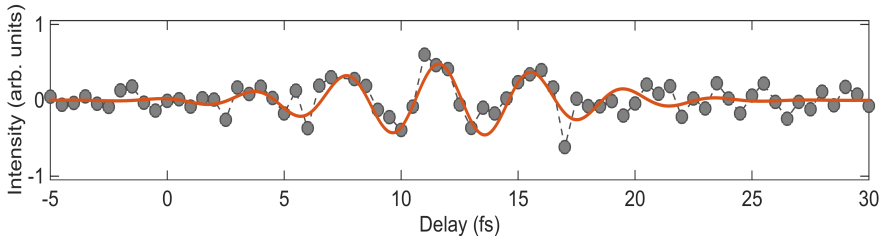
F. Calegari et al., *J. Phys. B: Atom. Mol. Opt. Phys.* ***49*** , 062001 (2016)

Charge migration in aromatic amino acids

Phenylalanine



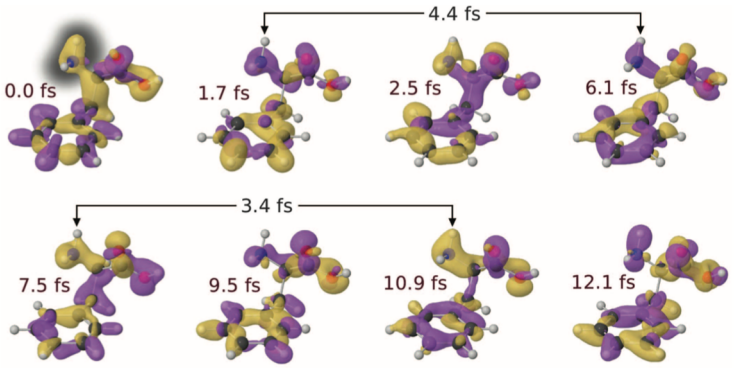
Tryptophan



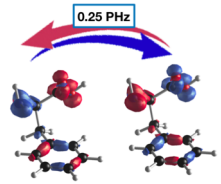
Charge migration resulting from the coherent superposition of 1-hole states

F. Calegari et al., Science 346, 336 (2014)
F. Calegari et al., IEEE JSTQE 21, 2419218 (2015)
M. Nisoli et al., Chem Rev 117 10760 (2017)
E. Perfetto, J. Phys. Chem. Lett. 11, 891–899 (2020)

Phenylalanine

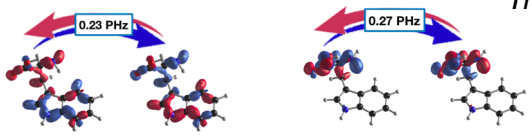


F. Martin, Universidad Autonoma de Madrid



Phenylalanine

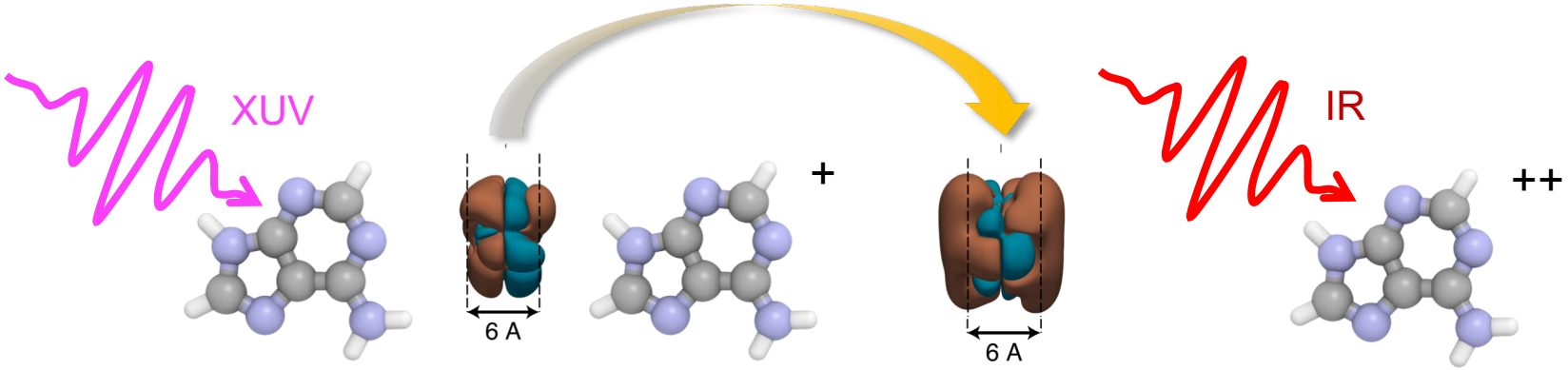
Tryptophan



G. Stefanucci, E. Perfetto, Università la Sapienza, Rome

Charge migration and stabilization in adenine

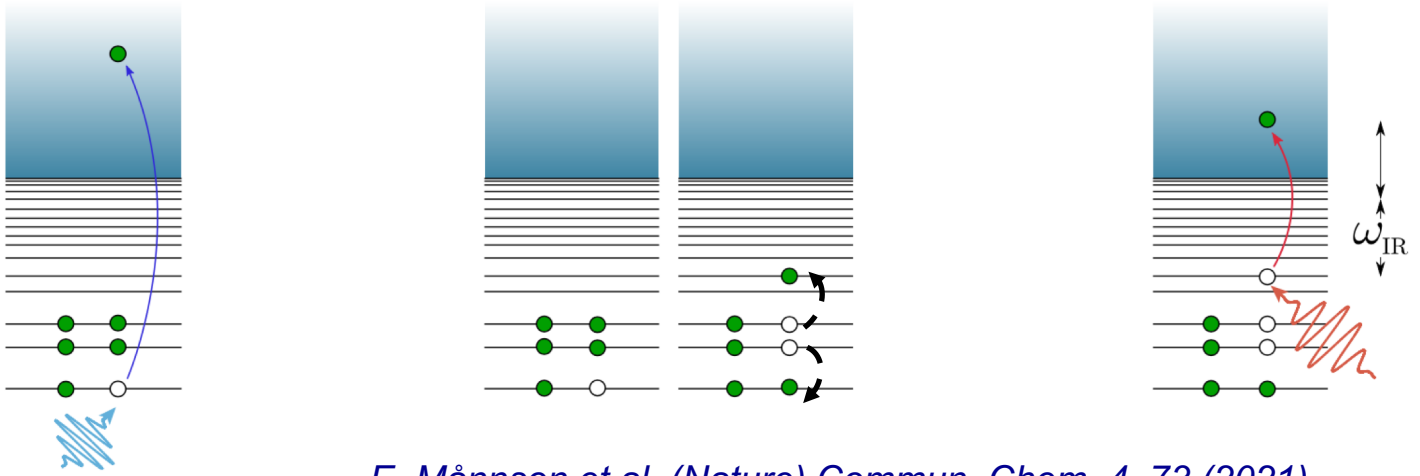
Correlation-driven
Charge migration/ inflation in 2.3 fs



① **XUV ionization**

② **Shake-up**

③ **IR ionization & stabilization**



E. Månnson et al, (Nature) Commun. Chem. 4, 73 (2021)

Open issues to be discussed in this session

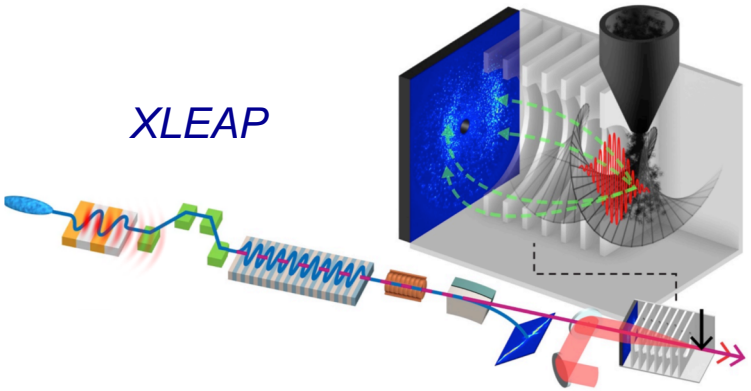
- **Better visualization of charge dynamics** (site specificity/sensitivity, x-ray absorption spectroscopy, x-ray photoelectron/auger spectroscopy, imaging…)
- **Demonstrate control of charge dynamics at attosecond time scales** (site selective ionization or controlled/localized valence excitation, non-linear schemes…)
- **Control of the molecular reactivity and/or other properties such as chiral response via electronic excitation** (control the charge location, control of dissociation and selective bond breaking)
- **Role of non-adiabatic couplings and bath**
- **and more...**

Can we use atto XFELs to address these issues?

New opportunities with atto XFELs

- Bright emission
- High photon energy
- High repetition rate (in CW up to MHz in future)
- Two-color experiments in the soft-x/x-ray
- Synchronization with a second laser pulse
- Possibility for single shot imaging

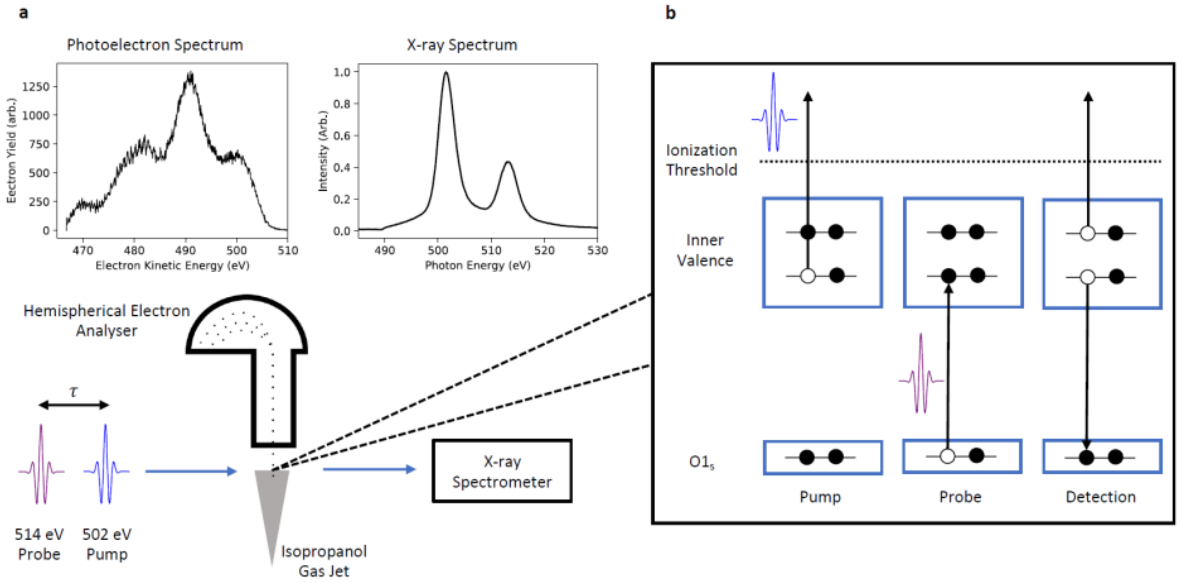
- SASE does not allow for pulse reproducibility
- Need for single shot detection
- Attosecond synchronization with an external laser?



*Duris, J. et al, Nat. Photonics **14,** 30–36 (2020)*

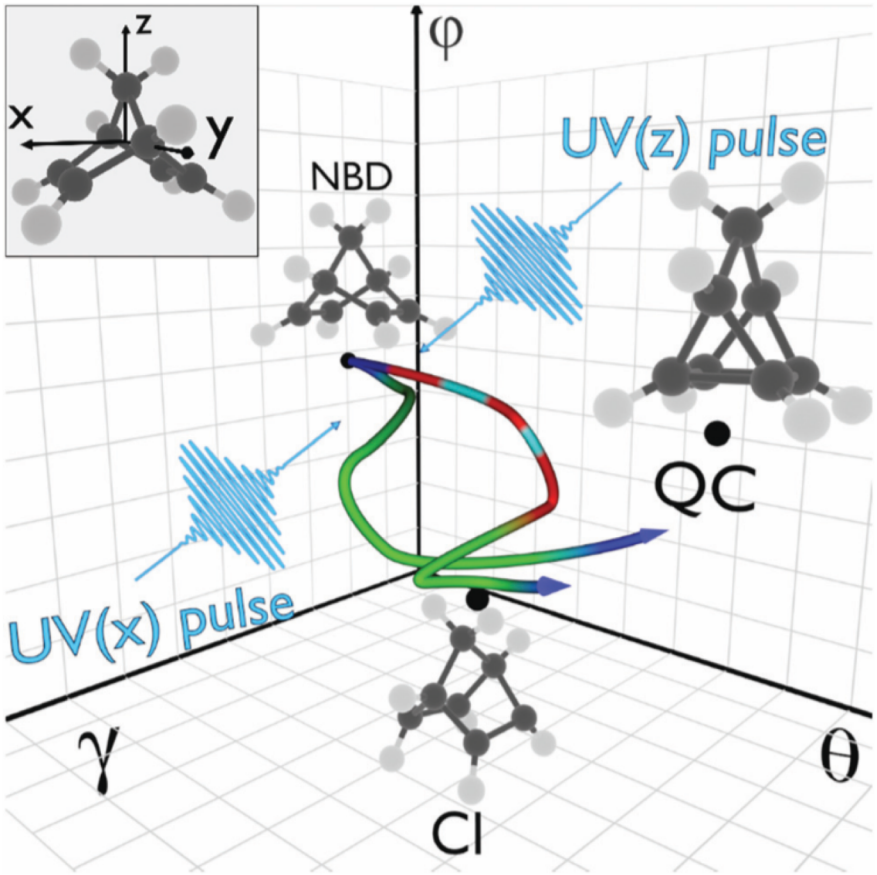
Two-color schemes to trigger and monitor charge migration

Jon Marangos
Imperial College London



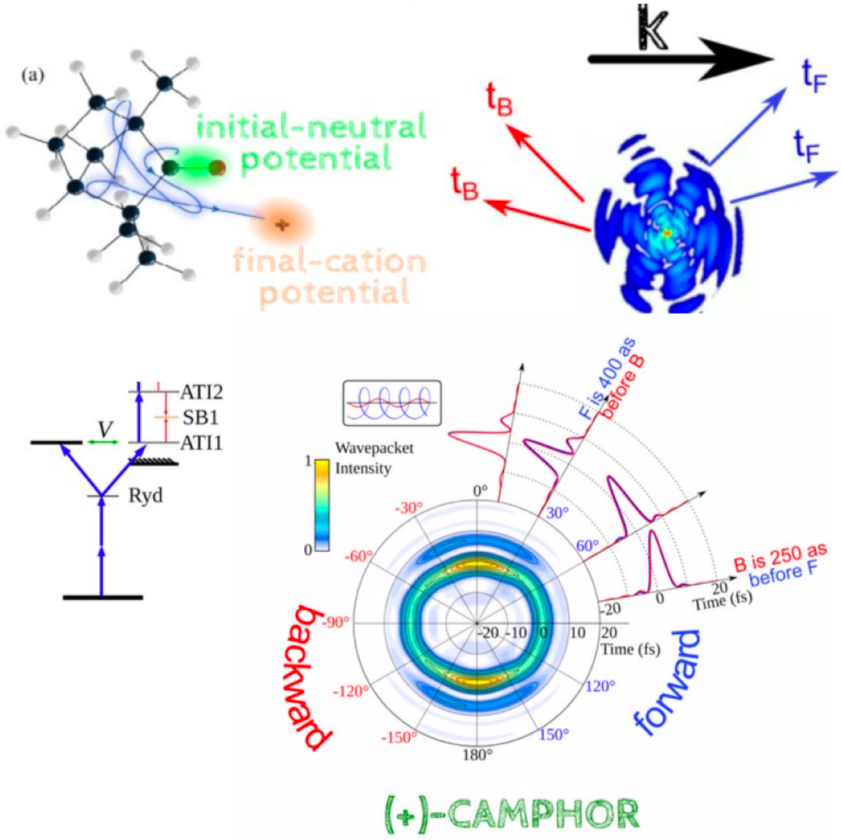
Theory of control of nuclear dynamics via electronic excitation

Françoise Remacle
University of Liège



Electronic coherences and ultrafast chiral response

Valérie Blanchet
CELIA



Session IV

19:20 – 19:35	Measurement of Ultrafast Electronic Dynamics with X-rays	J. Marangos (Imperial College London)
19:35 – 19:50	Exploiting Electronic Coherences for Steering Selectively Ultrafast Reactivity in Molecules	F. Remacle (Univ. of Liège)
19:50 – 20:05	Molecular Chirality on the Short Time-Scale	Valérie Blanchet (Univ. of Bordeaux)
20:05 – 20:40	Discussion	

- **Raise your hands or write in the chat for questions**
- **Only very urgent questions at the end of each talk**
- **Main discussion with questions after the talks**
- **Think to long term development and scientific impact (ten years or more, beyond what we can already do now)**