

Attosecond spectroscopy to track and control electron dynamics in matter

Francesca Calegari
CFEL-DESY, Universität Hamburg

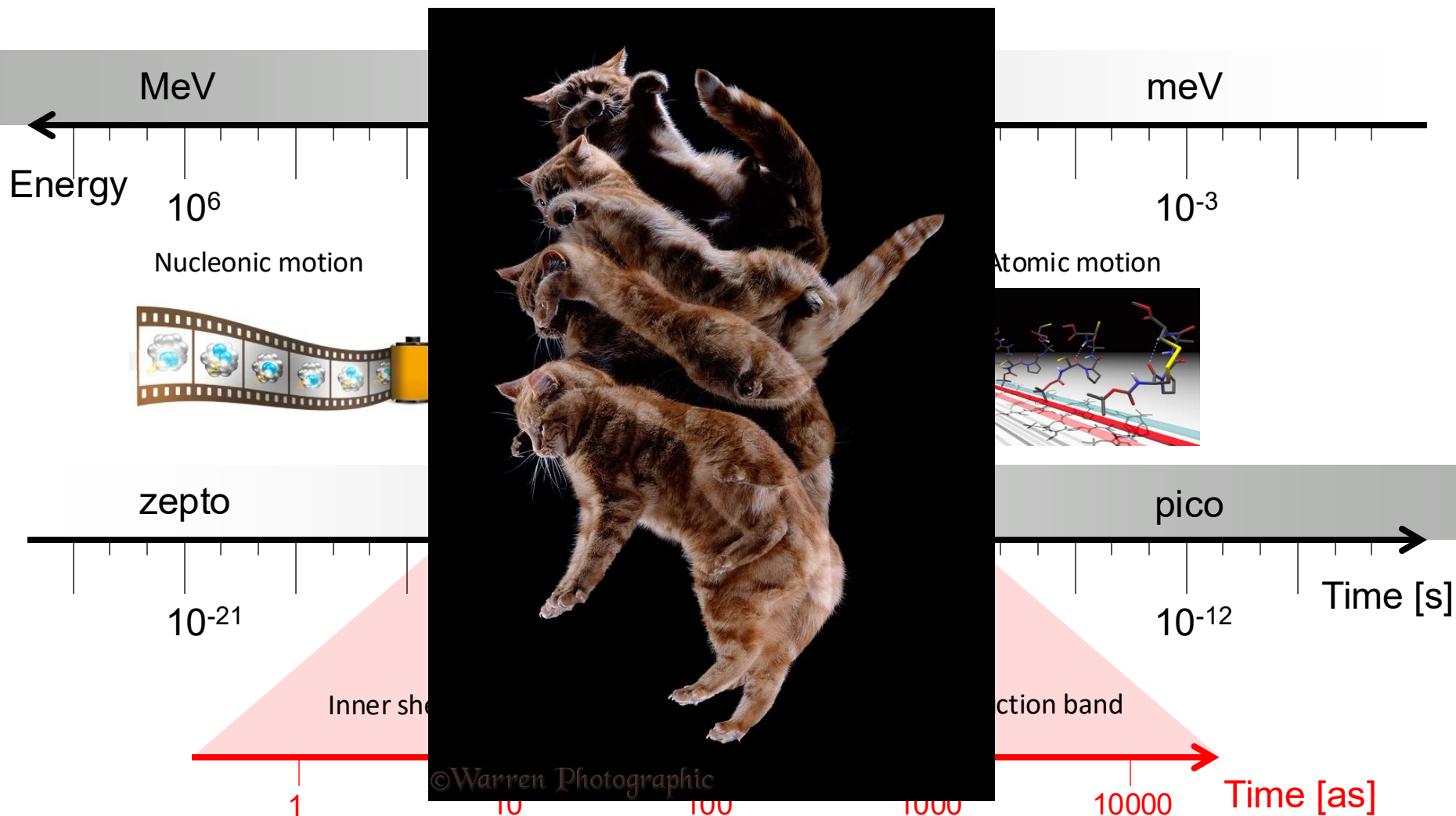
841. Heraeus Seminar Quantum
Technologies

CLUSTER OF EXCELLENCE
CUI: ADVANCED
IMAGING OF MATTER



Time scales in matter

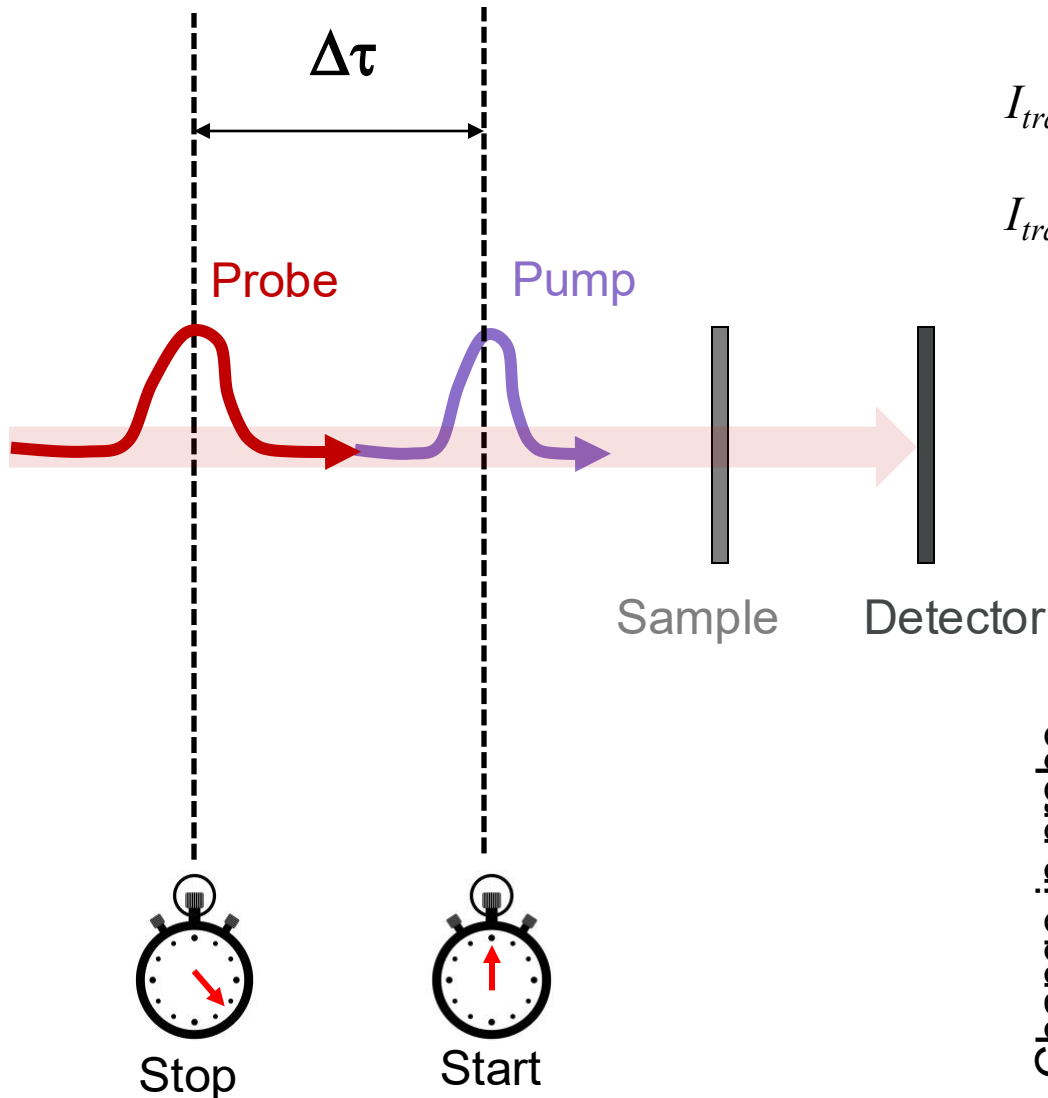
Matter is often driven by external stimuli out of equilibrium!



Outline

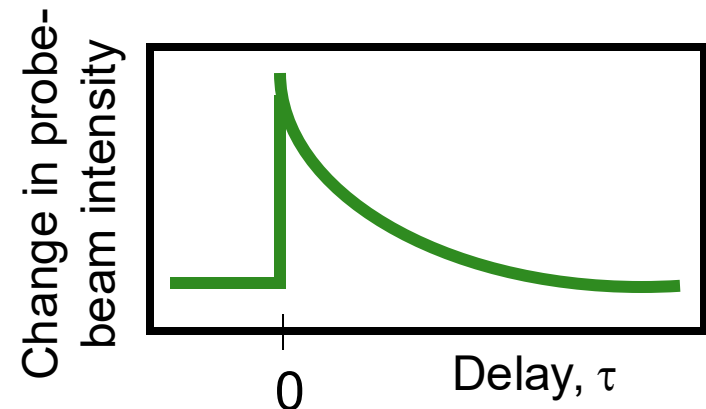
- **Attosecond pulse generation**
- **Charge migration: A route to attochemistry**
- **Photoemission Delays and Nanoplasmonics**

Time-resolved measurement: “pump & probe”

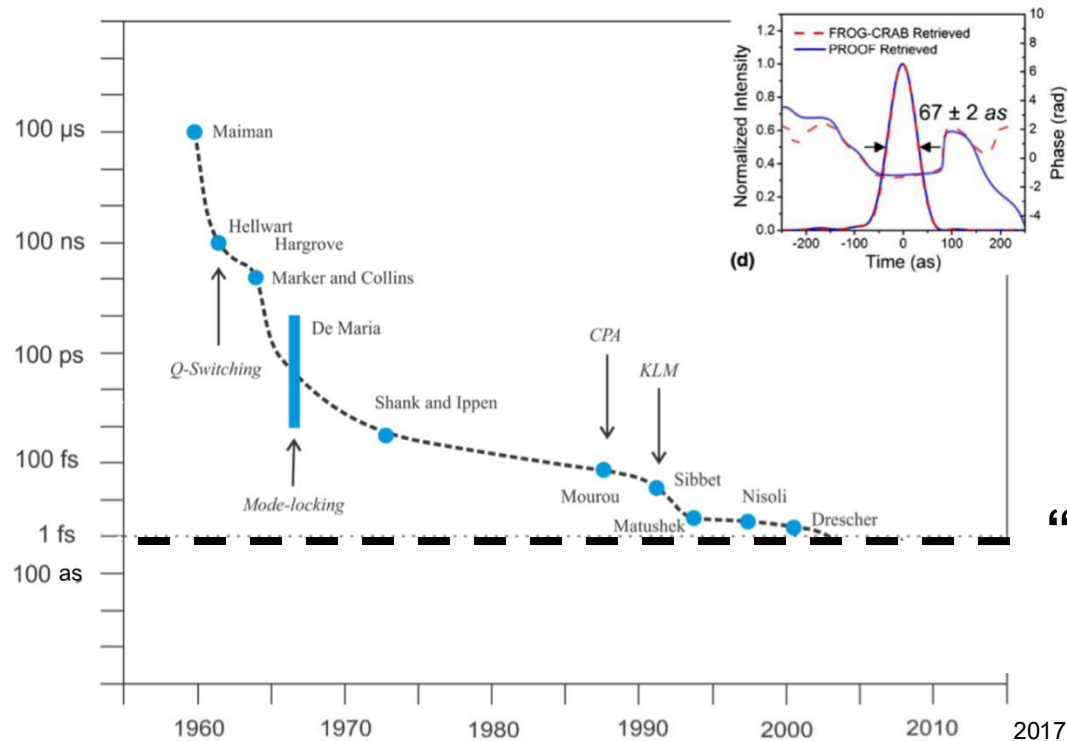


$$I_{\text{transmitted}} = I_{\text{incident}} \exp\{-\alpha_0 L\}$$

$$I_{\text{transmitted}}(\tau) = I_{\text{incident}} \exp\{-[\alpha_0 - \Delta\alpha(\tau)]L\}$$



Ultrashort laser pulses: a limit in pulse duration imposed by carrier wavelength



From Ursula Keller, Lecture notes “Ultrafast Lasers”, ETH Zürich

Femtosecond pulses
can be produced by
conventional lasers

“Femtosecond barrier”

The 0-fs pulse



Wayne Knox
University of Rochester

Observation of the 0-fs pulse

By W.H. Knox, R.S. Knox, J.E. Hoose, and R.N. Zare

The quest for the world's shortest laser pulse has led to a remarkable pace of development in ultrafast laser technology. Although pulses of only a few cycles duration have been made, clearly the observation of a 0-fs pulse would represent a key result in this field (see Fig. 1). In most experiments, a 100-fs pulse is amplified and passed through a nonlinear medium, generally a simple single-mode silica optical fiber. The resulting nonlinear propagation creates a large bandwidth increase, and, with careful control of subsequent optical phases, significant compression ratios have been obtained. A simple linear extrapolation predicted that continued progress should result in an important milestone (dashed line): the pulsewidth should have gone to zero in late 1986 and become negative in 1987. Incorporation of prisms have produced at best 6-fs (three-cycle) pulses.

We show that the reason for this

W.H. KNOX of AT&T Bell Laboratories, R.S. KNOX of the University of Rochester's Department of Physics and Astronomy, J.E. HOOSE of Milton Roy Inc., and R.N. ZARE of Stanford's Department of Chemistry carried out this work especially for the April issue of *Optics & Photonics News*.

pause in history has been the choice of nonlinear medium. Single-mode fused silica fibers are not capable of generating enough bandwidth to approach the 0-fs limit and beyond with realizable intensities. Our new nonlinear medium consists of a short length (1–10 mm) of zirconium encrusted high- T_c superconductor microcrystallite doped neodymium aluminate fiber maintained near the supercon-

ducting transition by a liquid nitrogen cooled cryostat with optical access ports. A strong pulsed magnetic field parallel to the fiber axis is synchronized with the laser, providing 40–80 T peak field strength at 8 kHz repetition rate. We study the pulse evolution as a function of propagation distance and record conventional autocorrelations.

Figure 2 shows our results. At 3

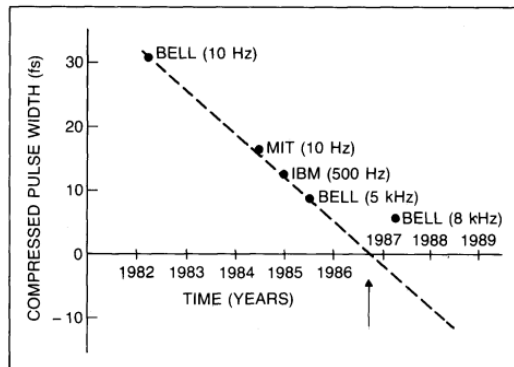


FIGURE 1. Evolution of pulse compression. The dashed line shows that in 1985 the 0-fs pulse was predicted to occur in late 1986. New nonlinear materials now allow us to attain this limit and beyond. Applications of large-negative-pulsewidth optical pulses should be abundant.

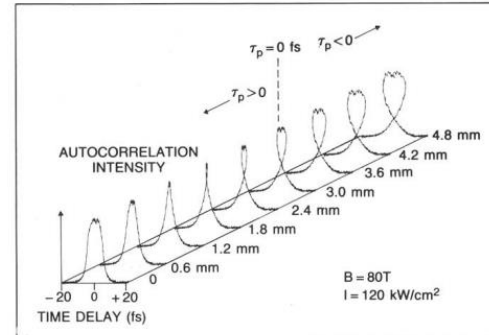


FIGURE 2. Autocorrelations of compressed pulses as a function of fiber length. We clearly observe the transition from normal pulse compression to 0-fs compression and beyond to the negative limit. We cannot test the uncertainty principle because the 0-fs pulse has no energy.

mm, note that the pulse FWHM actually goes to zero, and with longer propagation distance continues to go negative. This results, we believe, from the re-entrant behavior of the effective nonlinearity due to local-field effects in our microcrystallites. It is not clear how the nonlinear response exactly manages to modulate the dielectric function in such a way as to effect essentially a "wormhole" for passage of phase (not amplitude) in-

We tried to measure the power spectrum for the case closest to the 0-fs pulse. Uncertainty relations might appear to require an infinite bandwidth for a 0-fs pulse. Interestingly, the spectrum of the 0-fs pulse reveals an important feature of the 0-fs pulses that is not true of the 0-pi pulse. A 0-pi pulse has no area, but has energy. A 0-fs pulse has area, but no energy. This can easily be understood—the energy is just equal to the peak power

We are investigating possible violations of thermodynamics. Somebody's pulses must be getting longer.

THE OPTICS SHOPPE

Previously owned optics equipment

Have you hesitated about updating your lab because you're not sure what to do with the equipment you now have? Are you running out of storage space for equipment that's too good to discard but not quite what you need for your current projects? Have you ever wished your company would hold a garage sale?

Optics & Photonics News has a better idea.

"The Optics Shoppe" gives you the chance to let more than 16,000 colleagues know that you have optics equipment to sell or trade. Here are the details on how to take advantage of this feature:

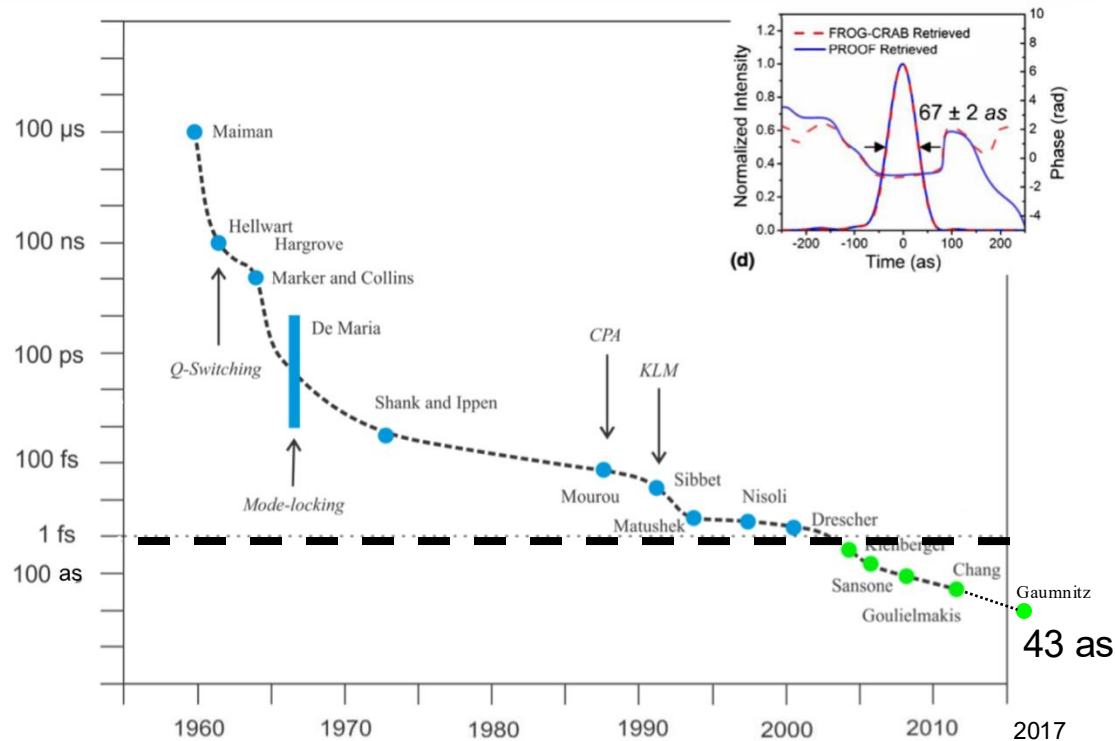
- Type an ad of no more than 25 words, including contact name and phone number.
- Enclose a \$10 check payable to The Optical Society of America and mail it to: The Optics Shoppe, *Optics & Photonics News*, 1816 Jefferson Place, N.W., Washington, D.C. 20036.
- Copy that reaches OSA by the 24th of the month will appear in the issue mailed about five weeks later (i.e., copy received by May 24 will appear in the July issue).

not advertised must be previously owned and in good condition.

ns? Call Larry Loash at (202) 462-1111. Then make a list of what you want to sell and what you want to buy. Put up those ads—that can find your key-words in all those extra in-

body's pulses must be getting longer.

Ultrashort laser pulses: a limit in pulse duration imposed by carrier wavelength



From Ursula Keller, Lecture notes "Ultrafast Lasers", ETH Zürich

Femtosecond pulses
can be produced by
conventional lasers

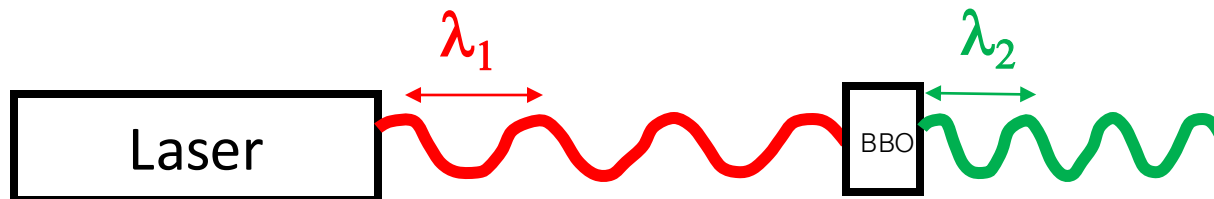
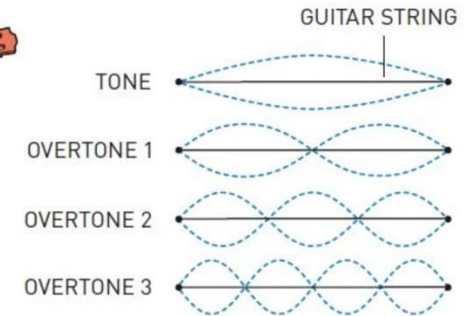
Attosecond pulses cannot
be produced by
conventional lasers: need
for shorter wavelengths

Nonlinear optics: perturbative regime

$$P = \epsilon_0 \left[\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots \right]$$

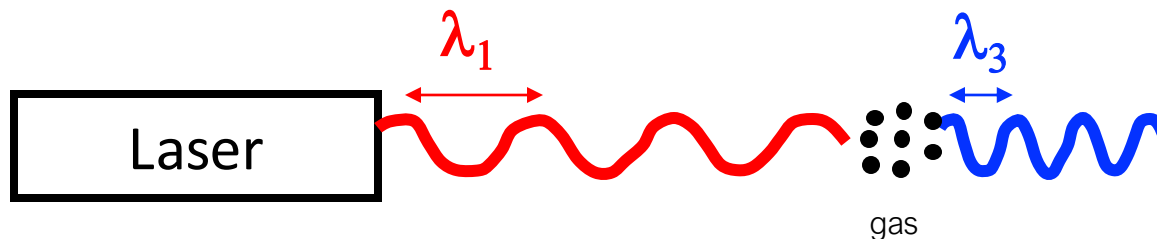


Overtone harmonics have several cycles for each cycle in the fundamental tone. Overtone harmonics work the same way in light waves.



$$\lambda_2 = \lambda_1 / 2$$

Second Harmonic Generation (SHG)

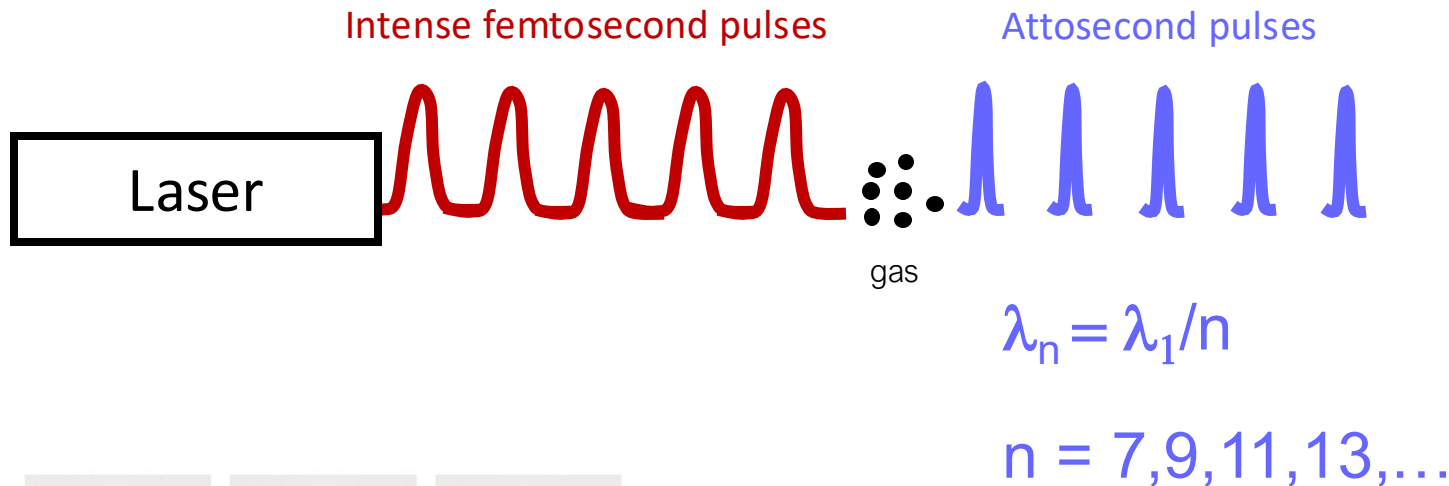


$$\lambda_3 = \lambda_1 / 3$$

Third Harmonic Generation (THG)

Extreme nonlinear optics: non-perturbative regime

$I > 10^{14} \text{ W/cm}^2 \rightarrow$ Highly non-perturbative interaction



III, Niklas Elmehed © Nobel Prize Outreach
Pierre Agostini
Prize share: 1/3



III, Niklas Elmehed © Nobel Prize Outreach
Ferenc Krausz
Prize share: 1/3



III, Niklas Elmehed © Nobel Prize Outreach
Anne L'Huillier
Prize share: 1/3



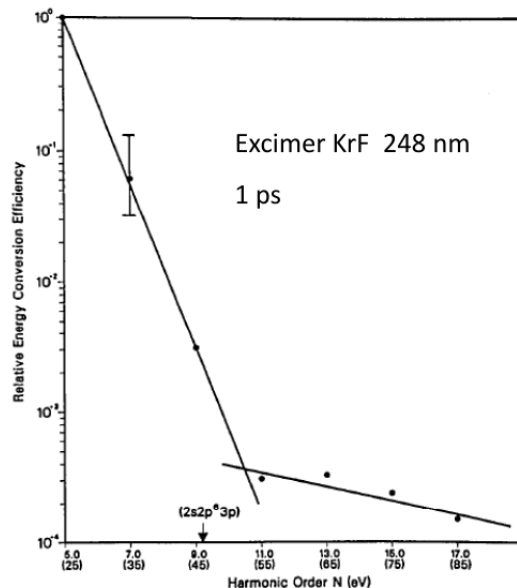
First observation of HHG

High-order Harmonic Generation - HHG

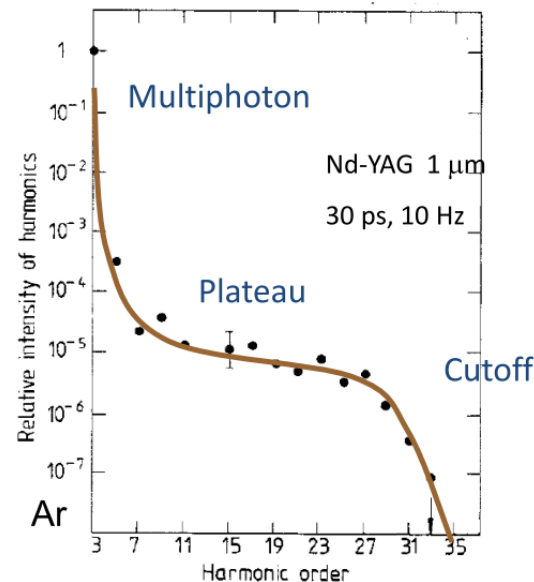
In the late 90's scientists observed frequency conversion of laser pulses to the XUV at orders incompatible with a perturbative multi-photon picture

Nonlinear optics was so far discussed only as a small perturbation of linear optics, giving rise to higher order terms

$$P = \varepsilon_0 [\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots]$$



McPherson et al, JOSA B (1987)



J Ferray et al, J. Phys. B (1988)

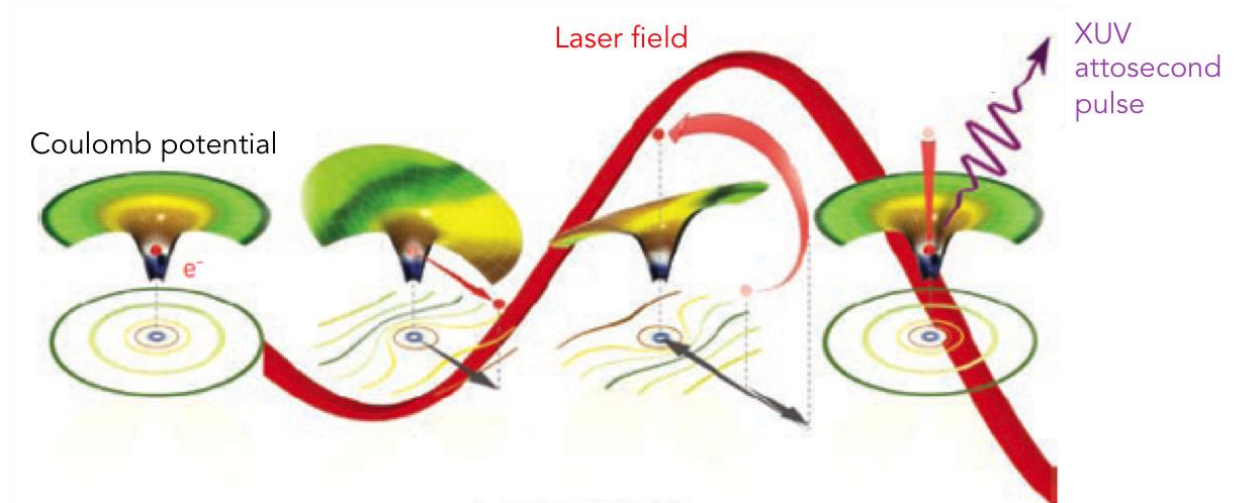
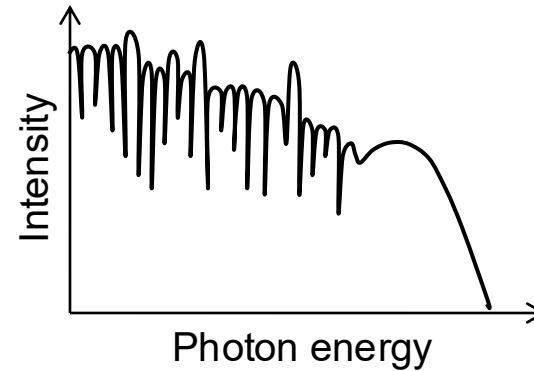


III. Niklas Elmehed © Nobel Prize Outreach
Anne L'Huillier
Prize share: 1/3

High-order Harmonic Generation

Three-step model

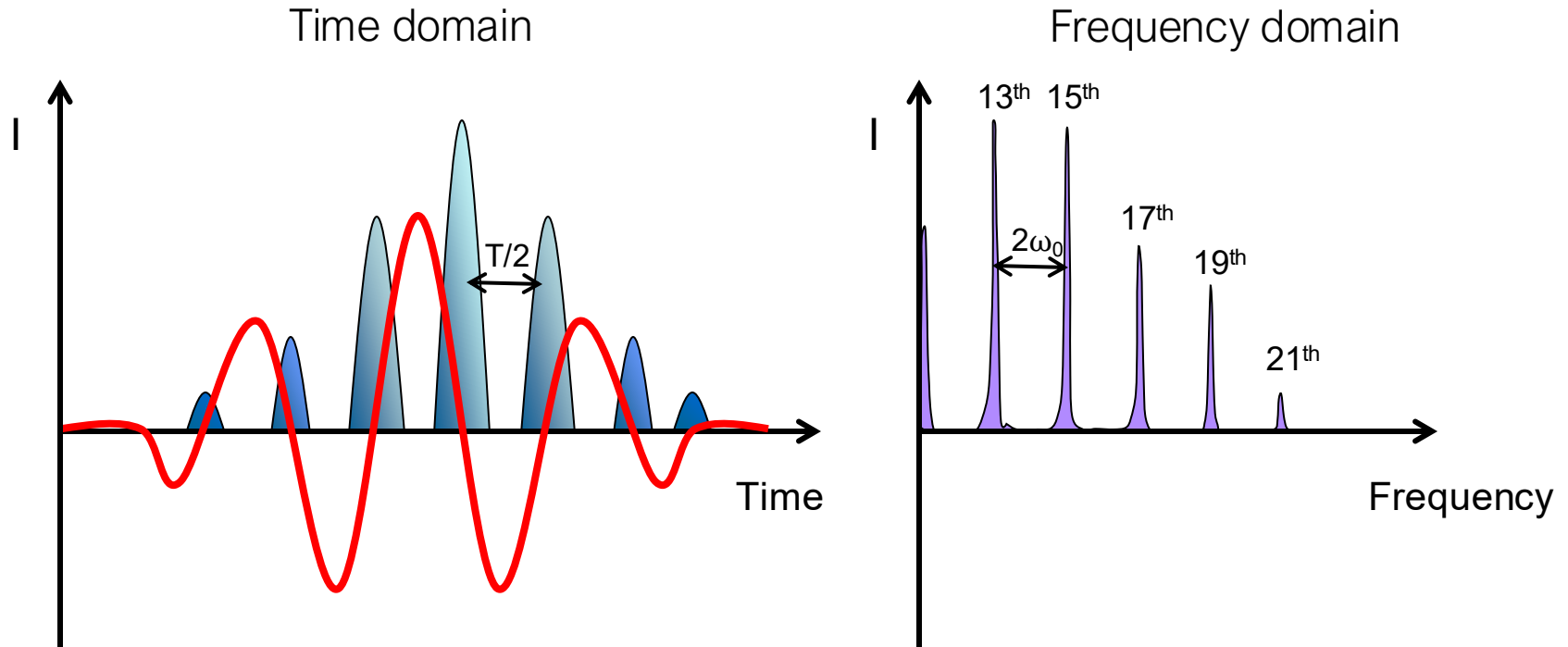
1. Tunnel ionization
2. Acceleration
3. Recollision



K.J. Schafer et al, Phys. Rev. Lett. 70, 1599 (1993)

P. B. Corkum, Phys. Rev. Lett. 71, 1994 (1993)

The attosecond pulse train

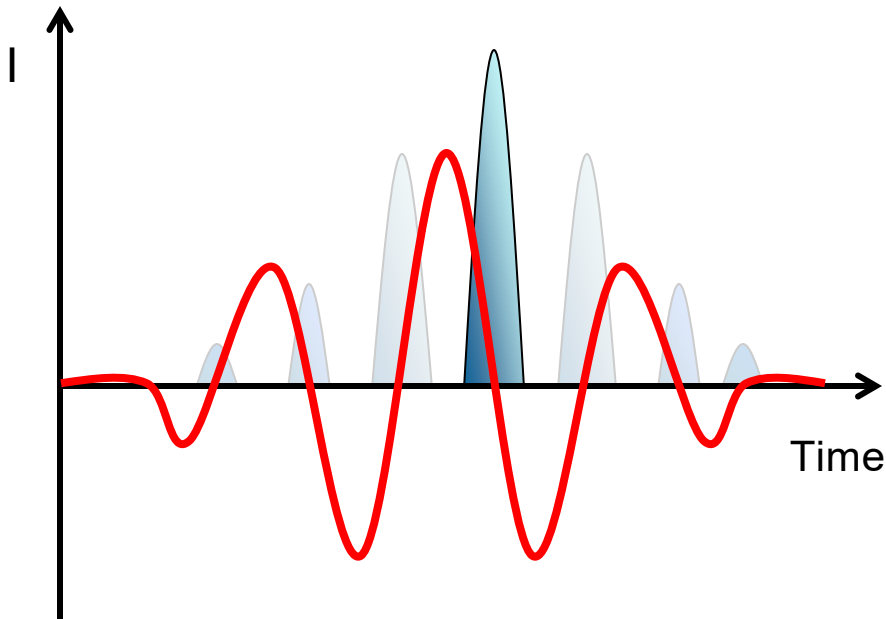


HHG every half cycle of the driving laser: attosecond pulse train

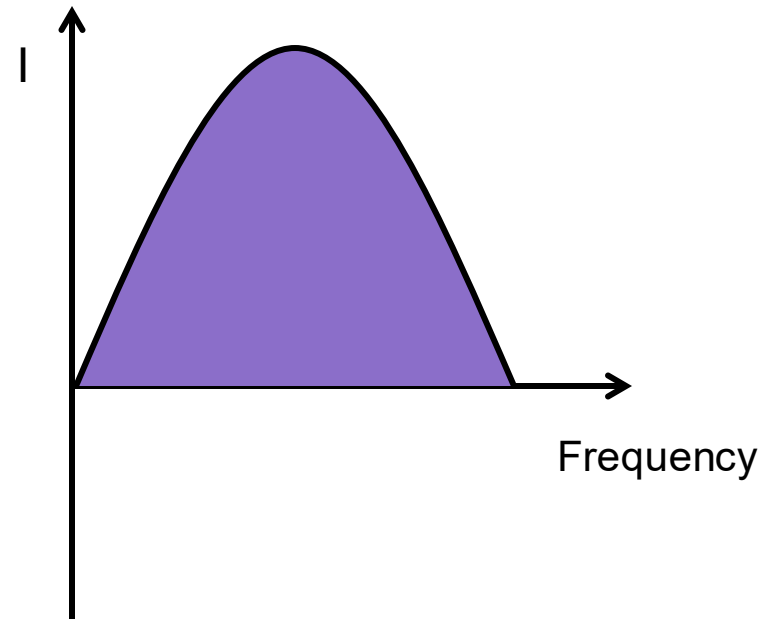
The interference between attosecond pulses separated $T/2$ gives rise to odd order harmonics of the fundamental frequency (spaced 2ω)

Isolated attosecond pulses

Time domain



Frequency domain

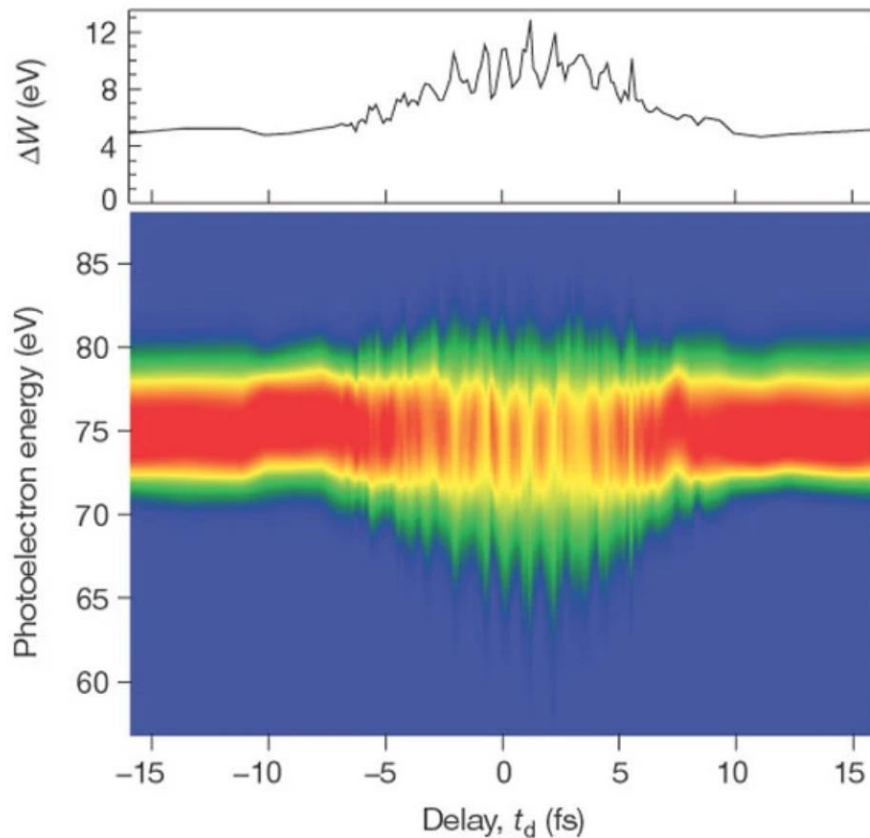


Spectral and temporal gating methods on HHG enable the generation of an isolated attosecond pulse

Shortest attosecond pulse 43 as!

T Gaumnitz et al, Optics Express 25 (2017)

First measurement of isolated attosecond pulses



III. Niklas Elmehed © Nobel Prize Outreach

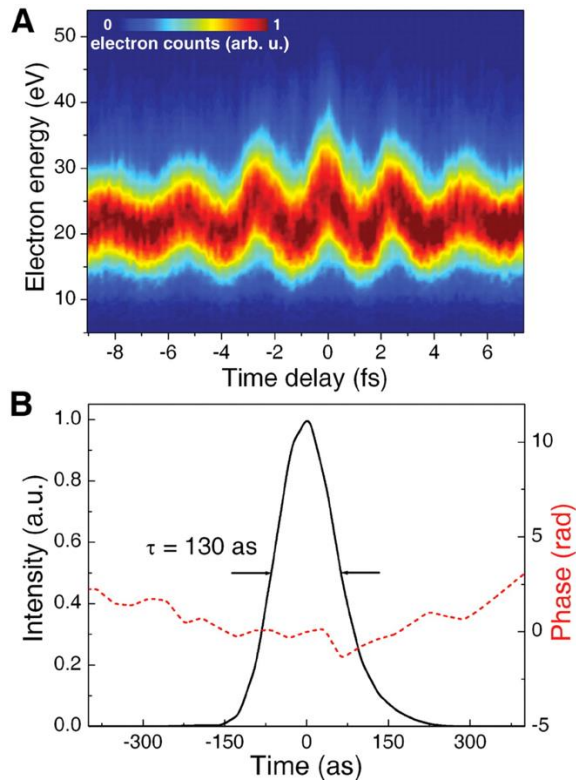
Ferenc Krausz

Prize share: 1/3

Hentschel, M. et al. Attosecond metrology. Nature 414, 509–513 (2001)

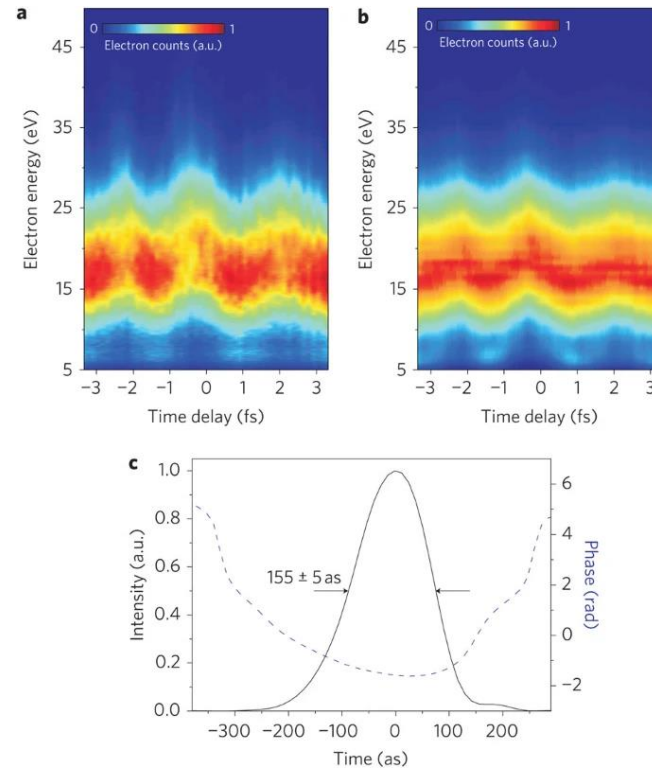
First temporal gating approaches

Polarization gating



G. Sansone et al., Science 314, 443 (2006)

Ionization gating



Ferrari, F., Calegari, F., et al. Nature Photon 4, 875–879 (2010)

Advancements in attosecond technology

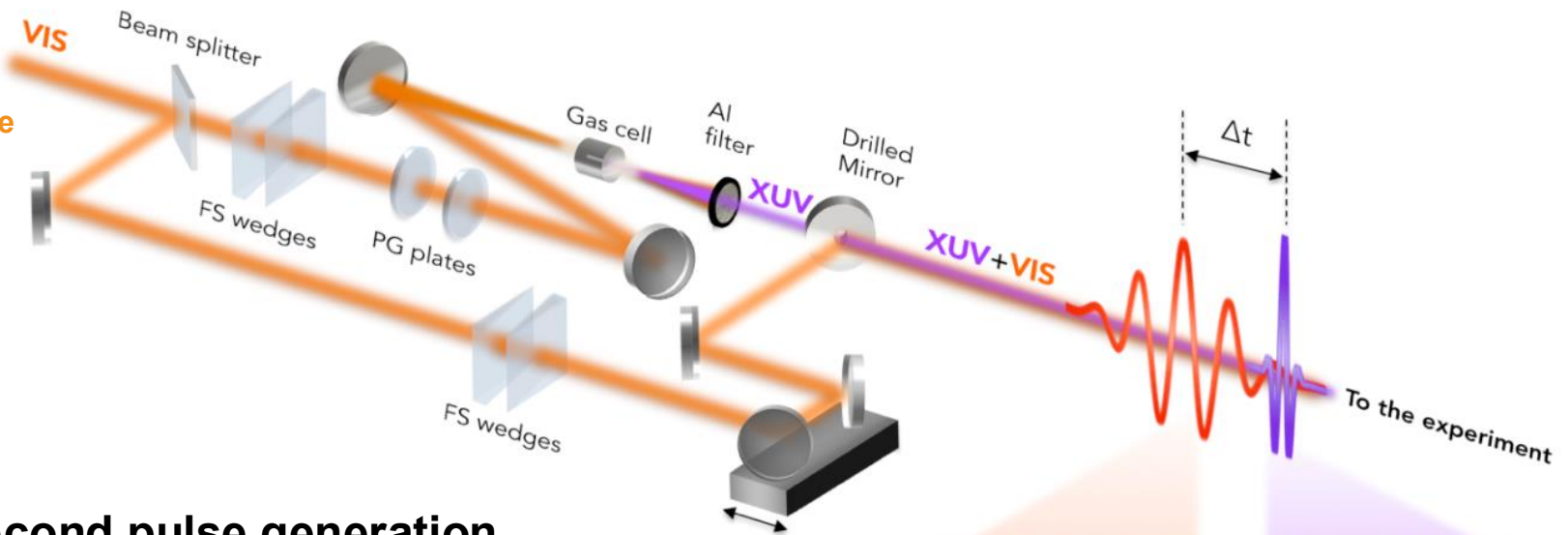
Gating method (gas)	Central energy/ FWHM (eV)	Pulse duration/FT limit (as)	Pulse energy	Peak power (MW)	Characterization	Year/refs.
PG (Ne)	100/22	130/45	70 pJ	0.5	FROG-CRAB	2006/CNR-IFN/Italy ¹³⁶
AG (Ne)	83/28	80/75	0.5 nJ	6	FROG-CRAB	2008/MPQ/Germany ¹³⁷
IG (Xe)	25/8	155/130	9 nJ	60	FROG-CRAB	2010/CNR-IFN/Italy ¹³⁸
DOG (Ne)	100/40	67/16	N/A	N/A	PROOF	2012/UCF/USA ¹³⁹
Lighthouse (Ne)	50/35	(N/A)/47	N/A	N/A	N/A	2013/NRC/Canada ¹⁴⁰
TC (Xe)	30/7	500/(N/A)	1.3 μ J	2600	Autocorrelation	2013/RIKEN/Japan ¹⁴¹
PG (Ne)	170/120	53/20	3 pJ	0.05	PROOF	2017/UCF/USA ³⁶
AG (Ne)	110/80	43/34	N/A	N/A	ML-VTGPA	2017/ETHZ/Switzerland ¹⁴²
AG (Ne)	250/100	<322/20	40 pJ	0.1	FROG-CRAB	2017/ICFO/Spain ^{26,143}
DOG (Ne)	150/100	305/(N/A)	0.3 nJ	1	Neural network	2019/UCF/USA ⁴⁰

Li, J., Lu, J., Chew, A. et al. Nat Commun 11, 2748 (2020)

- Attosecond sources deliver pulses in the mJ energy range
- Attosecond sources can cover the water window (200 – 500 eV)

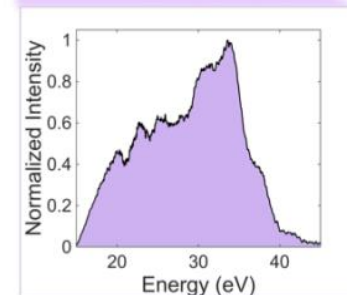
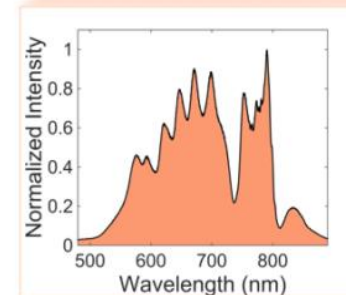
Attosecond pump-probe setup

4-5 fs
2.5 mJ
1 kHz
CEP-stable



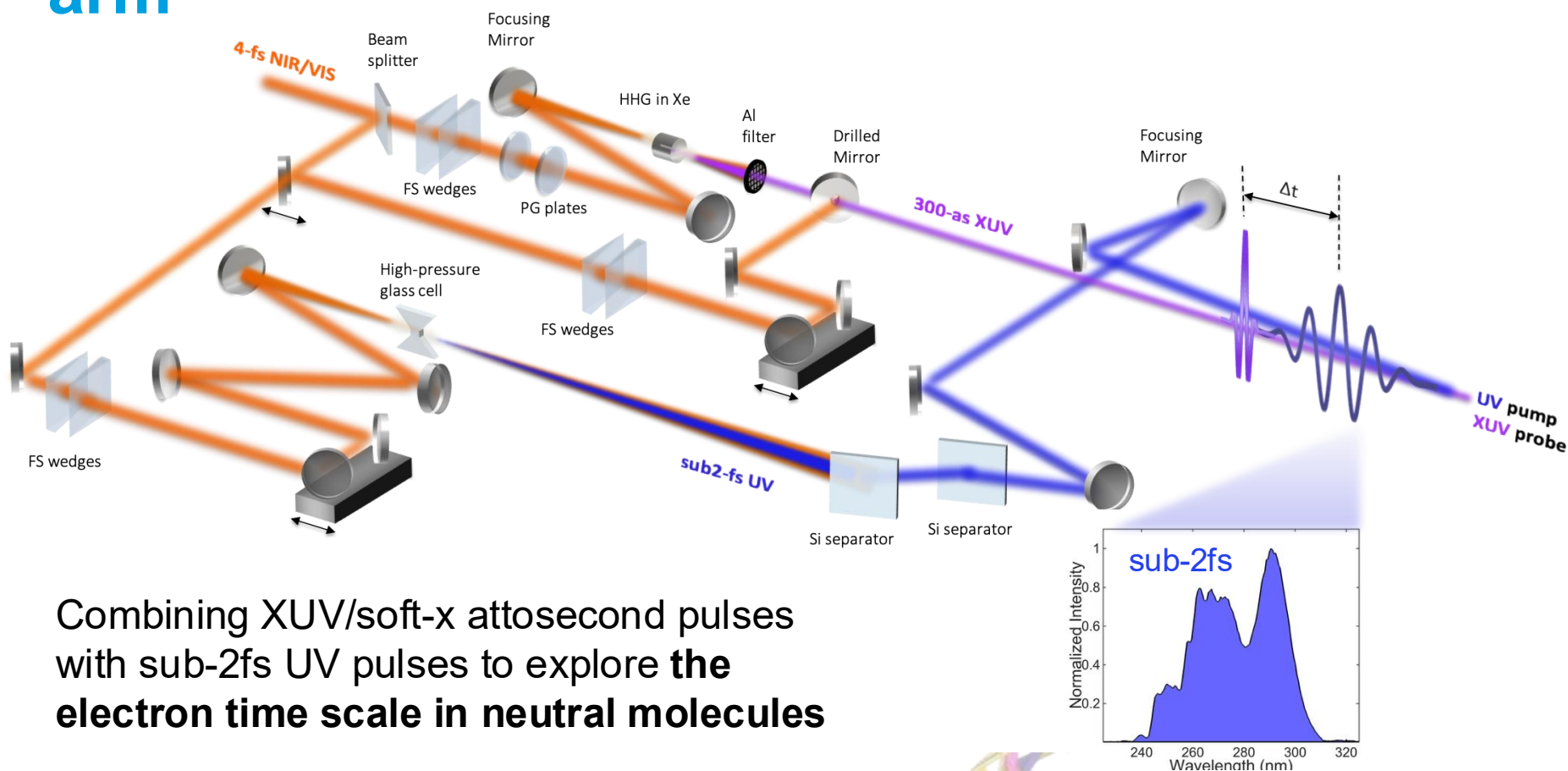
Attosecond pulse generation

- High-order Harmonic Generation (HHG)
- Polarization gating/ionization gating
- Energy range: 15 eV – 45 eV
- Temporal duration: 100 as



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

Attosecond pump-probe setup + UV arm



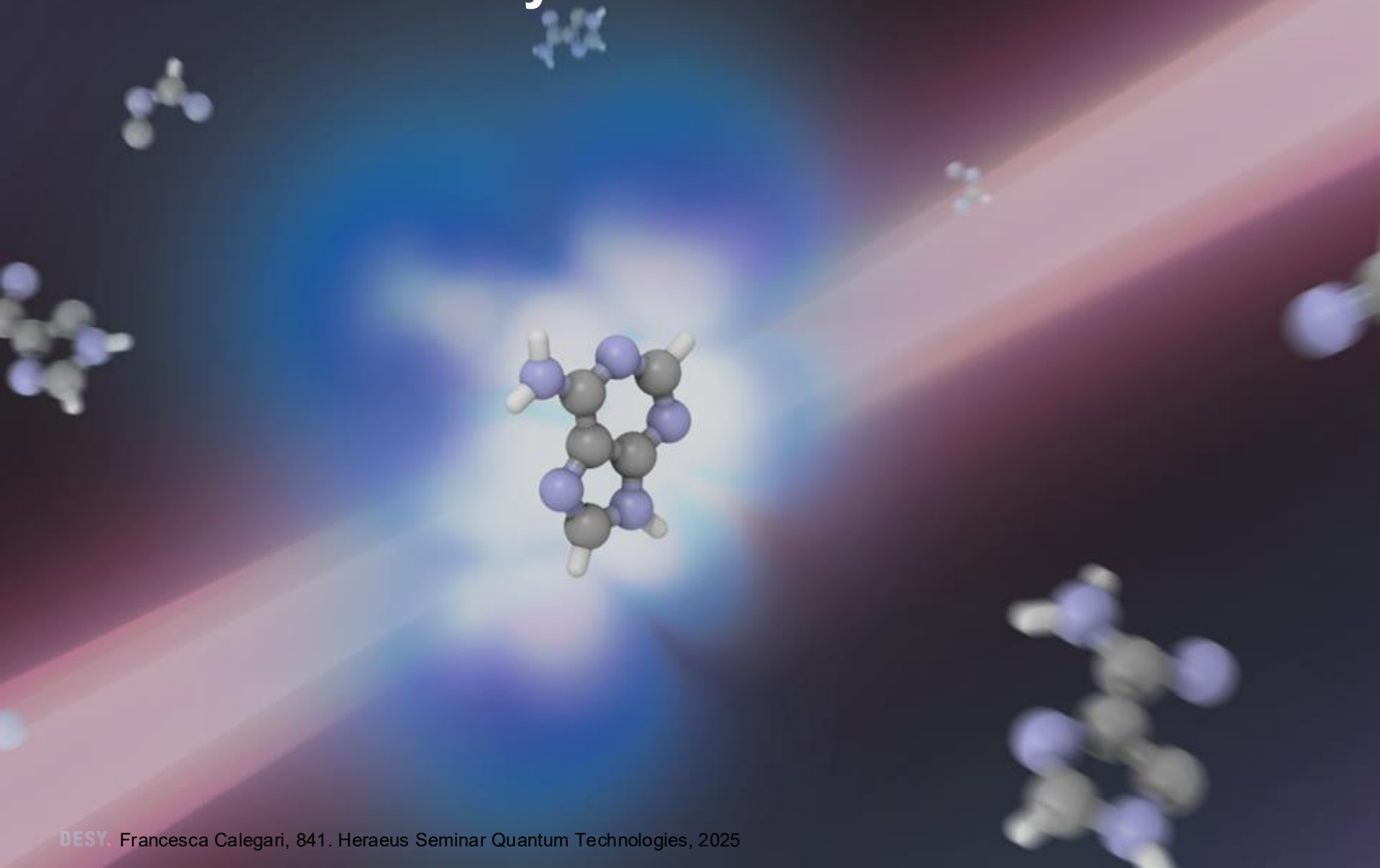
Combining XUV/soft-x attosecond pulses with sub-2fs UV pulses to explore **the electron time scale in neutral molecules**

V. Wanie et al., Rev. Sci. Instrum. 95, 083004 (2024)

M. Galli et al, Optics letters 44 (6), 1308-1311 (2019)

V. Wanie et al, J. Phys Photonics 6, 025005 (2023)

Charge migration and att chemistry



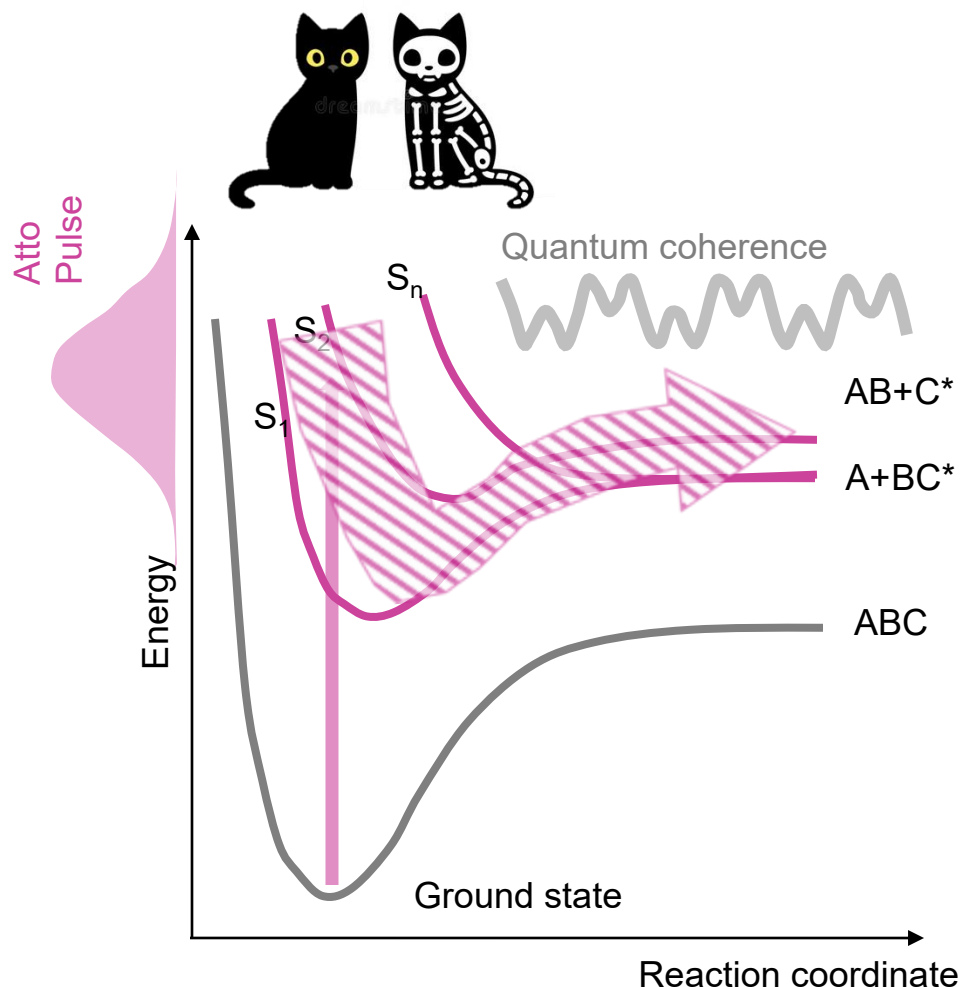
A portrait of a middle-aged man with dark hair and a mustache, wearing a dark suit, white shirt, and a red patterned tie. He is looking directly at the camera against a blue background.

The coherent excitation of a specific electronic state efficiently creates specific photoproducts.

$$|\Psi\rangle = c_1 |v_1\rangle + c_2 |v_2\rangle \dots + c_n |v_n\rangle$$

DESY. Francesca Calegari, 841. Heraeus Seminar Quantum Technologies, 2025

Attochemistry: new quantum landscape



Quantum forces on the atomic nuclei

Using **attosecond pulses**
broadband and sudden excitation
(fixed nuclei):

An electronic wavepacket (EWP) is created.

$$|\Psi\rangle = c_1 |S_1\rangle + c_2 |S_2\rangle \dots + c_n |S_n\rangle$$

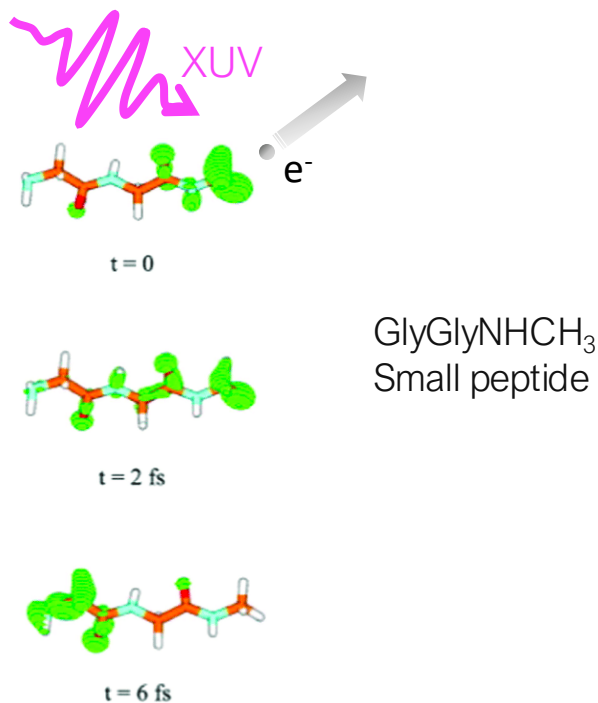
The quantum correlation between electron and nuclei creates new terms in the total force field → **shaping EWP**
allows to engineer the force.

Charge directed reactivity!

M. Cardosa-Gutierrez et al, J. Phys. B, 57 133501 (2024)

F. Calegari & F. Martin, Commun Chem 6, 184 (2023)

Charge migration



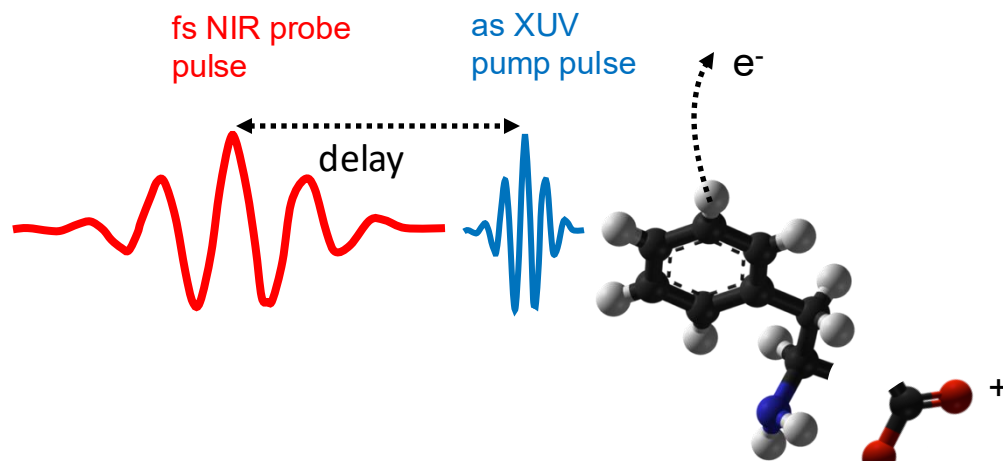
Sudden ionization/electronic excitation leads to EWP creation and a non-stationary charge density distribution: **charge migration**.

- How can we initiate charge migration?
- How can we observe charge migration?
- Can we exert control on chemical reactivity?

S. Lünnemann et al., Chem. Phys. Lett. 450, 232 (2008)
L. Cederbaum et al, Chem. Phys. Lett. 307, 205 (1999)
F. Remacle, R. Levine, PNAS 103, 6793 (2006)
A. Kuleff, L. Cederbaum, Chem. Phys. 338, 320 (2007)

Detecting charge migration

Attosecond time resolved photofragmentation



IR probe pulses:

Duration = 4 fs
Wavelength = 800 nm
Intensity = 10^{12} - 10^{13} W/cm²
CEP-stable
Interferometer actively stabilized
(RMS 10 as)

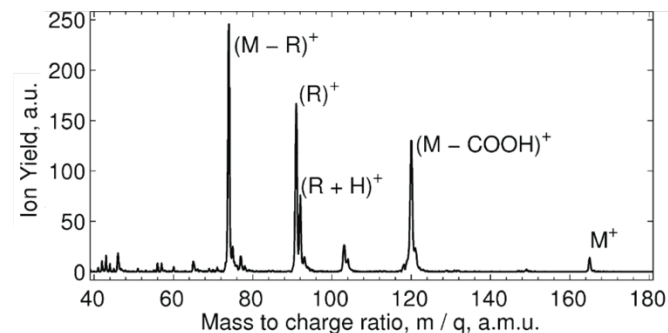
XUV pump pulses:

HHG in Krypton
Polarisation gating
Duration = 200 as
Energy range = 17-45 eV
Pulse energy = few nJ

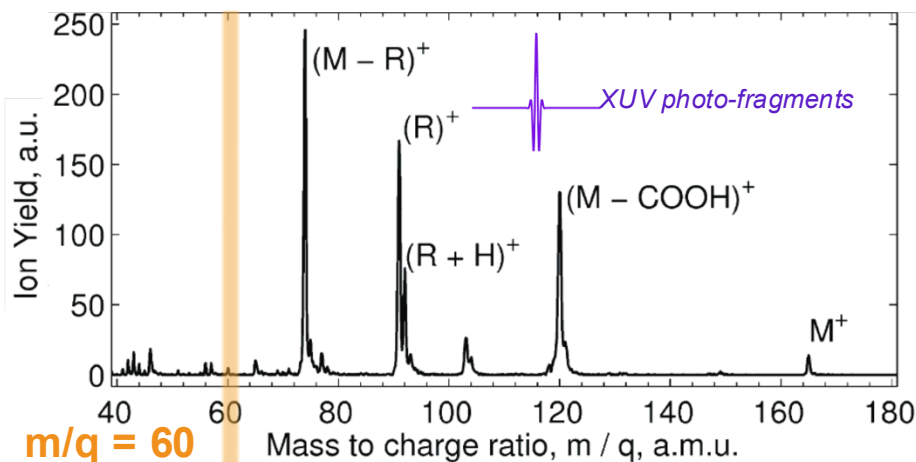


DETECTOR

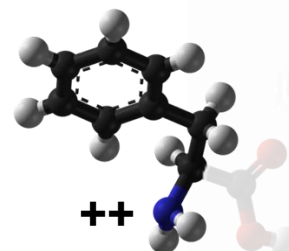
MASS SPECTRUM



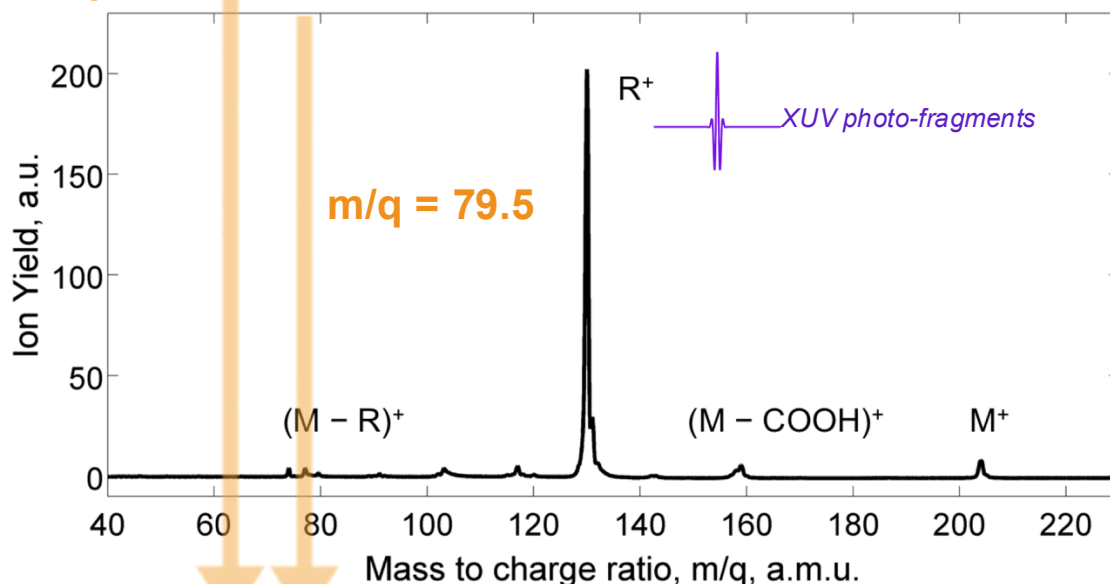
Charge migration in aromatic amino acids



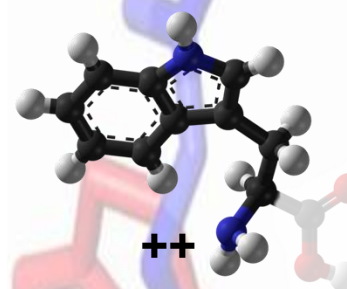
PHENYLALANINE



M^+
Parent ion
 $(M - COOH)^+$
Immonium ion
 R^+
Side chain



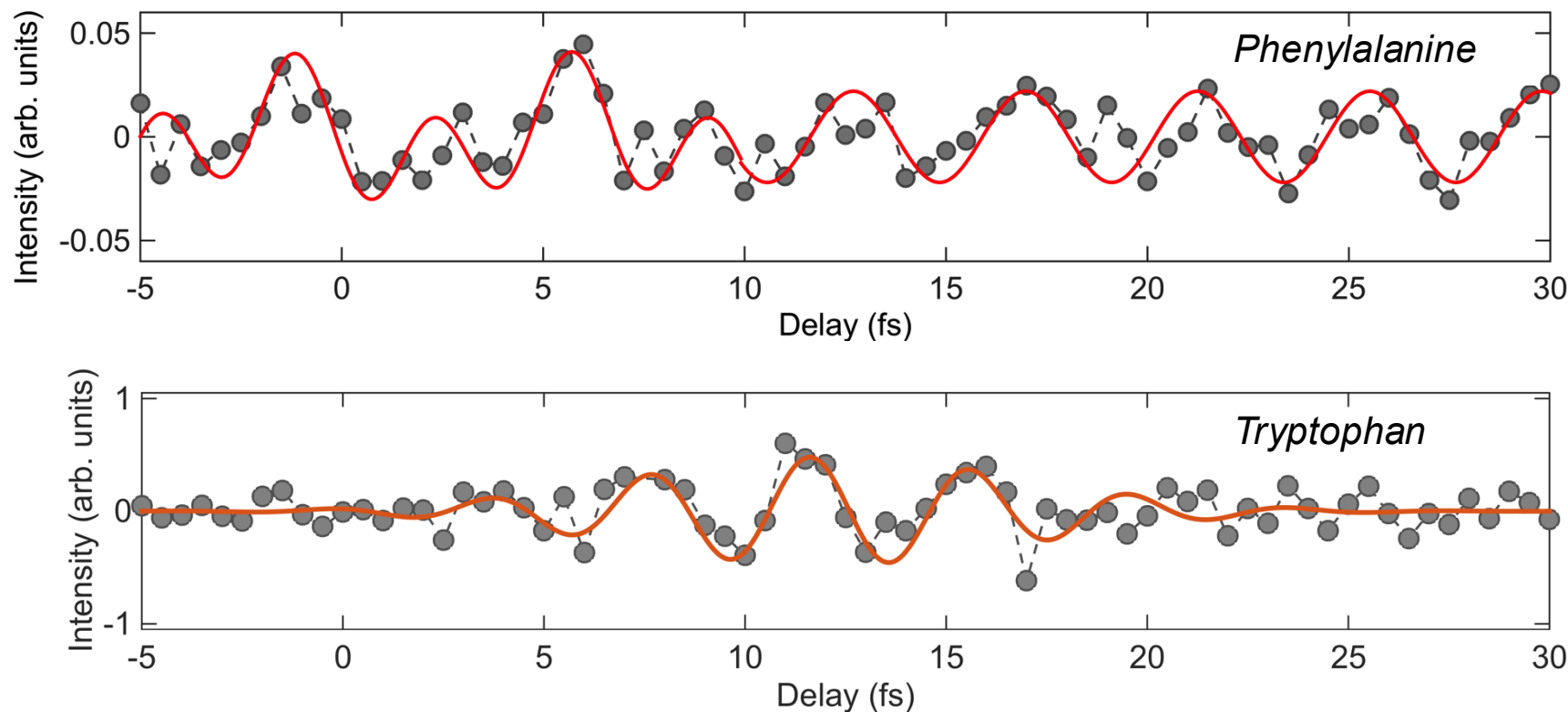
TRYPTOPHAN



$(M - COOH)^{++} =$ Doubly charged immonium ion

Charge migration in aromatic amino acids

Dication yield after subtraction of the 25-fs decay



F. Calegari et al., Science 346, 336 (2014)
M. Lara-Astiaso et al, The journal of physical chemistry letters 9 (16), 4570-4577, (2018)

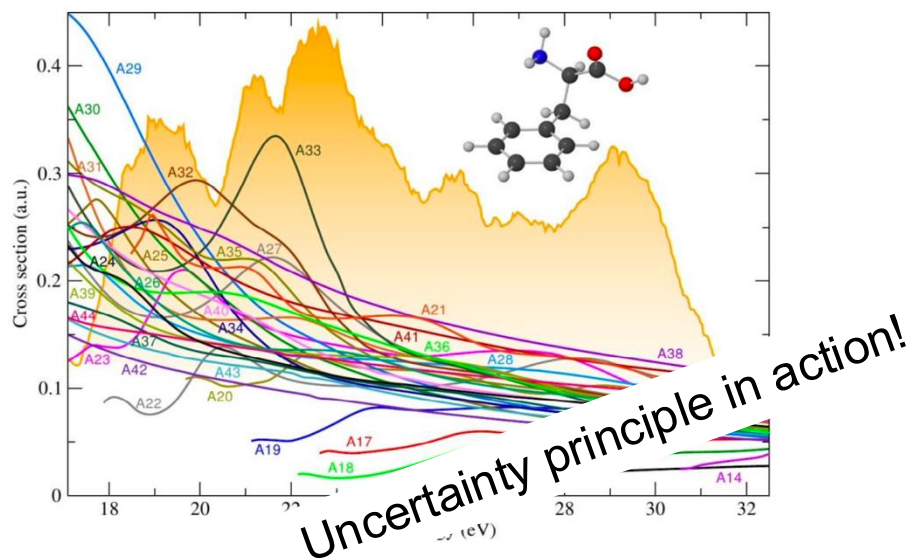
Sub 4.5 fs oscillations

Collaboration with M. Nisoli, Politecnico di Milano (Italy)

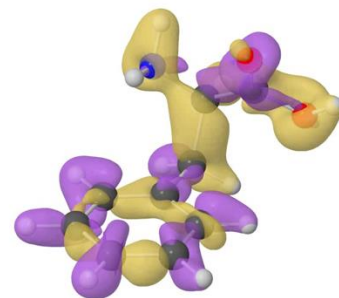
Charge migration in aromatic amino acids

An attosecond pulse is a broadband pulse!

→ Coherent superposition of a manifold of electronic states

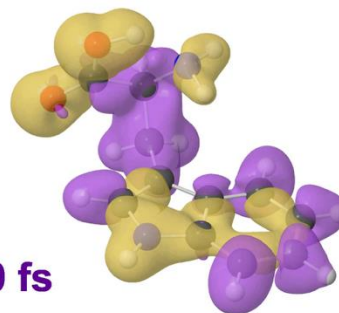


Phenylalanine



0.00 fs

Tryptophan



0.00 fs

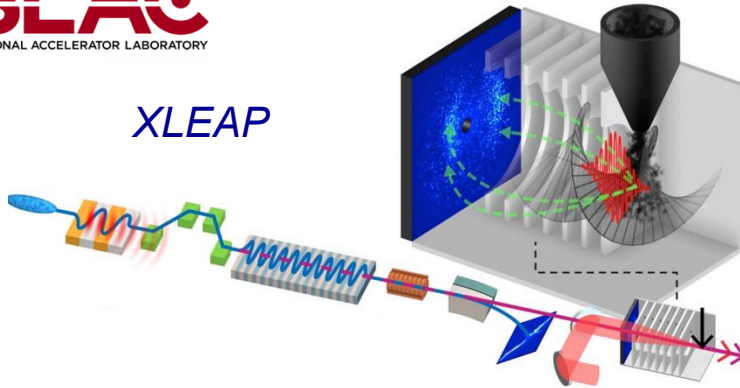
F. Calegari et al., Science 346, 336 (2014)

Collaboration with F. Martin, UAM (Madrid)

Attosecond pulses at XFELs



XLEAP



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

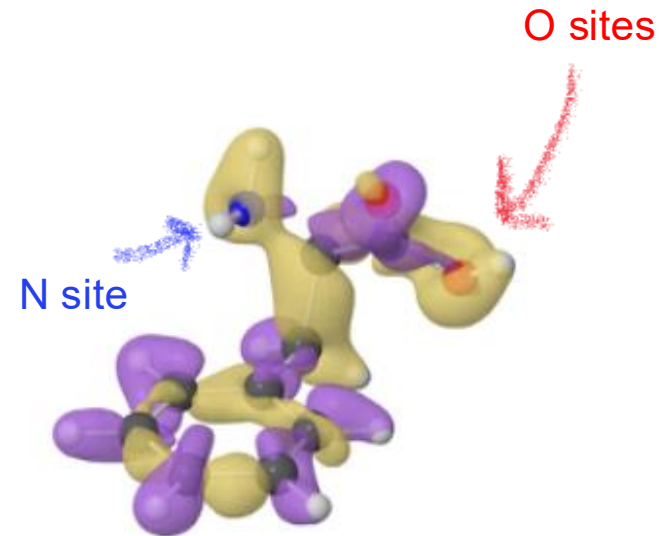
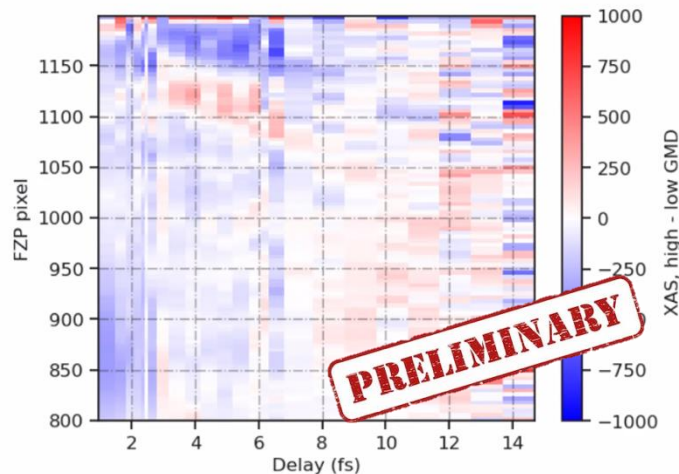


A. Marinelli



J. P. Cryan

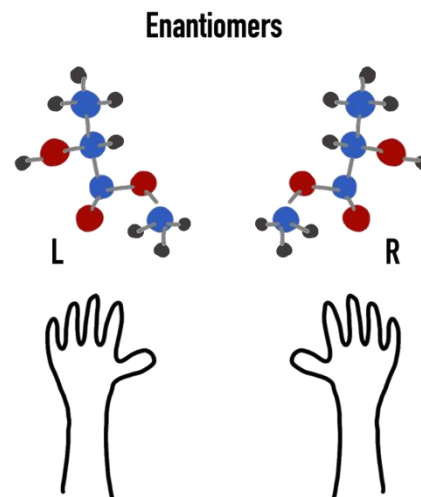
Look at the dynamics site selectively with XAS



Chirality

Concept

- Non-superimposable mirror images
- Handedness defines the interaction with another chiral object



Chiral recognition

Chiral molecules = enantiomers

- Homochirality of all living organisms (l - amino acids)
- Pharmacology → healing vs toxic effect

l-methamphetamine



Nasal
decongestant

d-methamphetamine

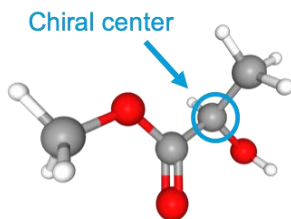
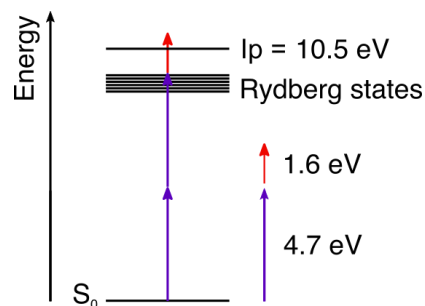


Recreational
drug

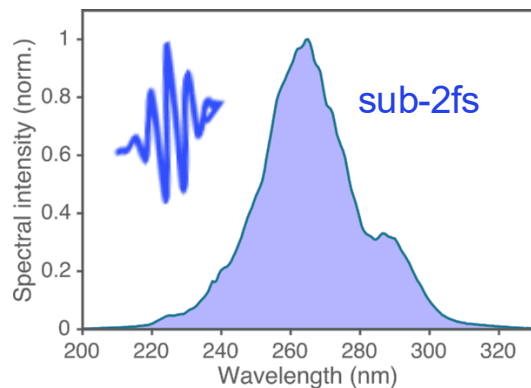
Charge migration in neutral chiral molecules

Time-resolved PECD in methyl lactate

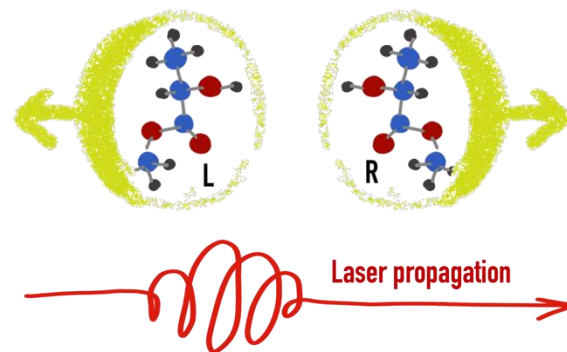
Methyl lactate



- The broadband UV pulse excites a coherent superposition of Rydberg states
- Photoelectron Circular Dichroism (PECD) can be used to detect the chiral response after electronic excitation



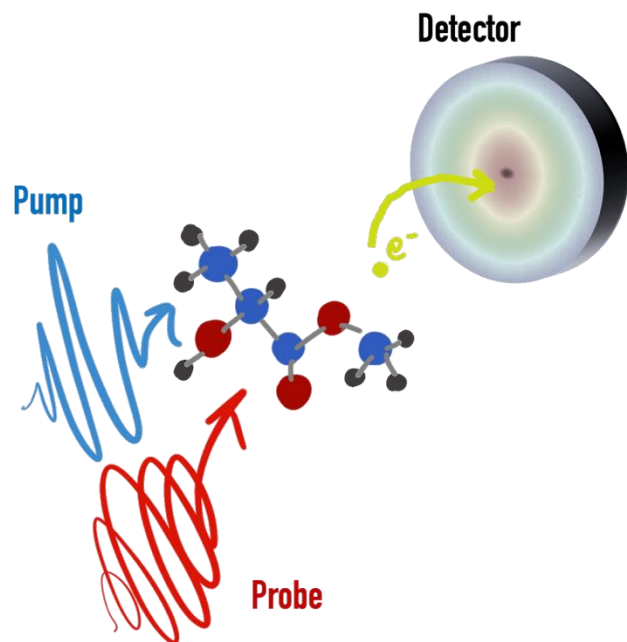
Photoelectron circular dychroism (PECD)



S. Beaulieu et al, Faraday Discuss., 194, 325 (2016)

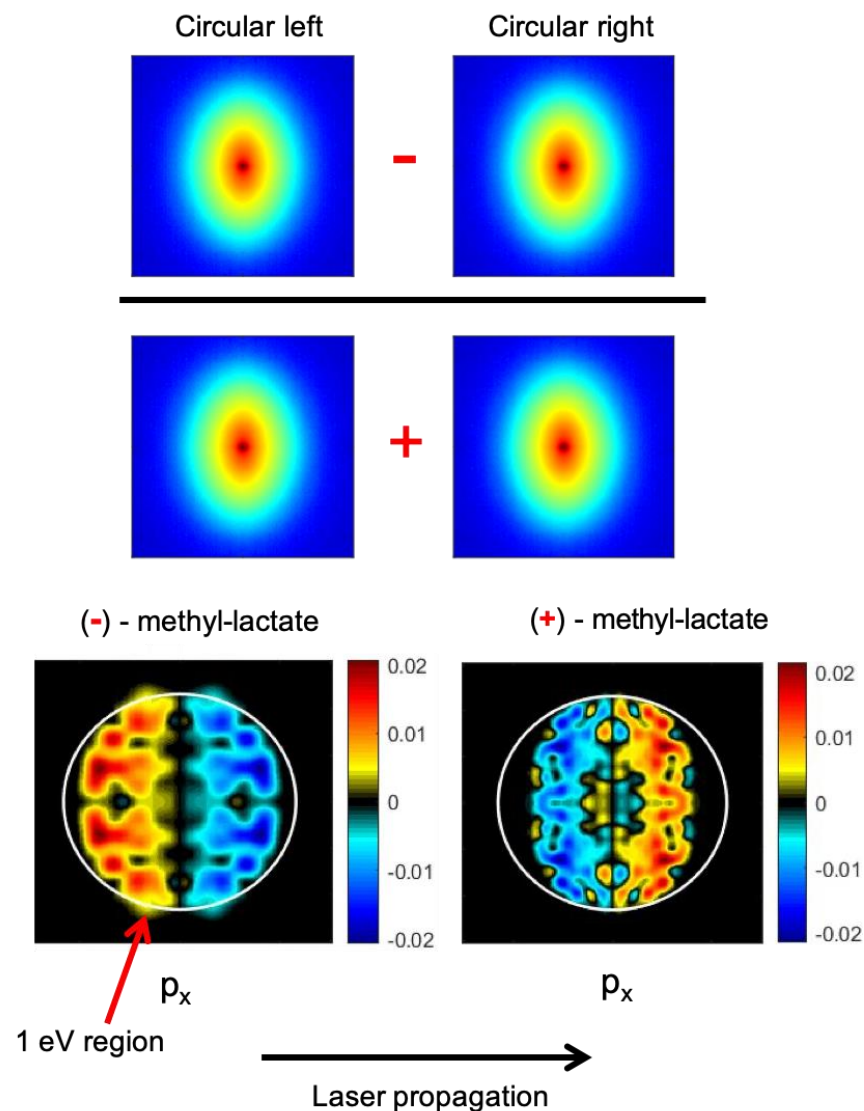
Collaboration with Y. Mairesse and V. Blanchet, CELIA, Bordeaux

TR-PECD setup



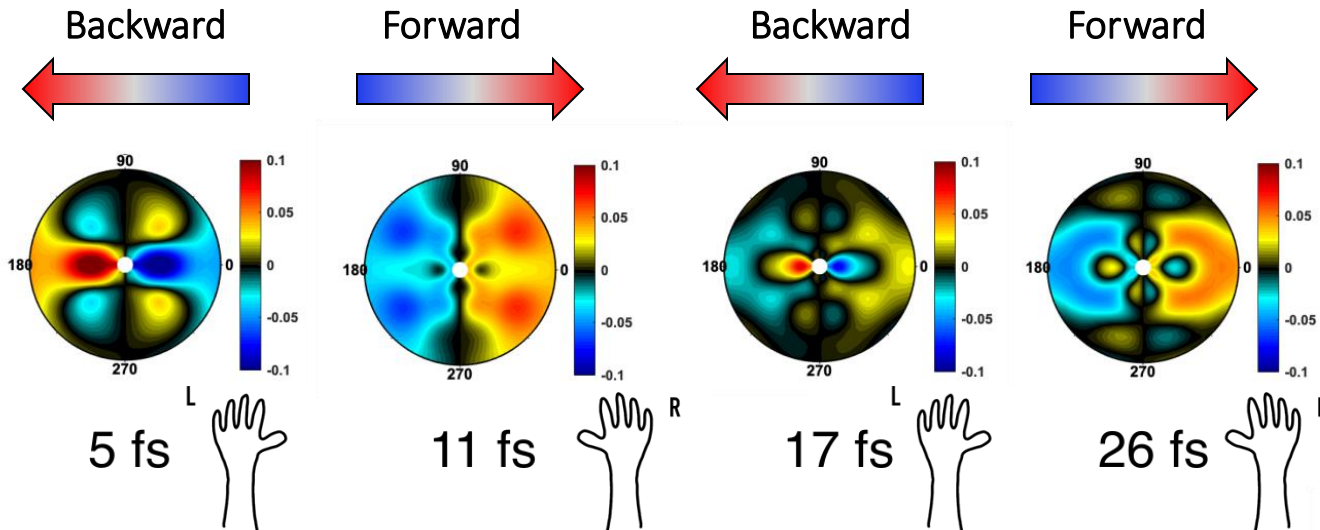
UV pump pulses:
 Duration = sub-2fs
 Energy range = 4-5 eV
 Pulse energy = 100 nJ

IR probe pulses:
 Duration = sub-5fs
 Circularly polarized

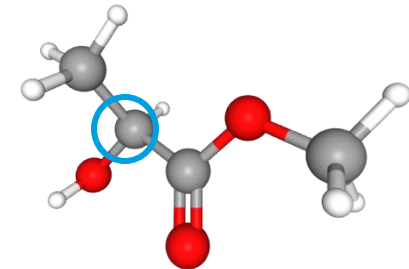


Time-dependent PECD

Ultrafast inversion of PECD



(S) - methyl-lactate

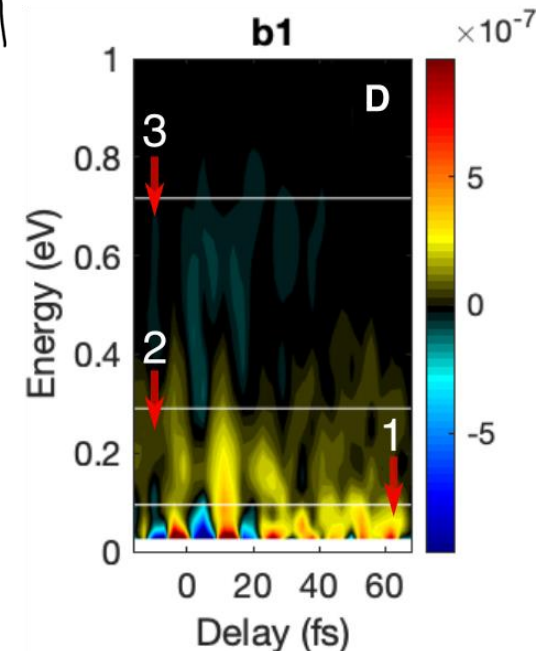


Energy-resolved fit of the angular distributions (p-Basex):

$$D(E, \theta) \approx \beta_0 \pm \beta_1 \cos(\theta) + \beta_2 P_2(\cos(\theta)) + \dots$$

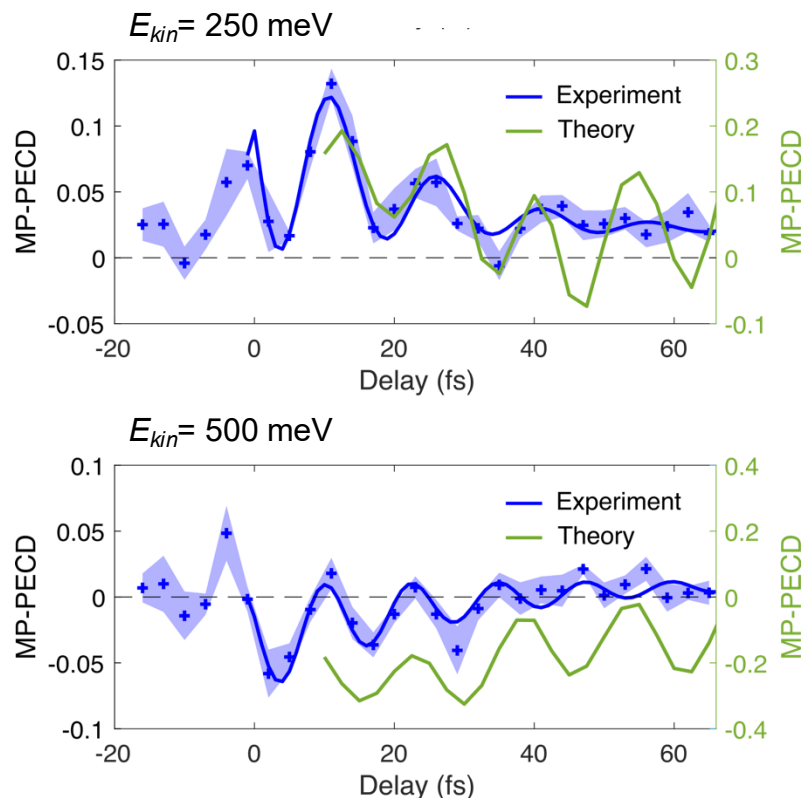
$$MP-PECD(E) = \frac{1}{\beta_0} \left(2\beta_1 - \frac{1}{2}\beta_3 + \frac{1}{4}\beta_5 - \frac{5}{32}\beta_7 + \dots \right)$$

V. Wanie et al, Nature 630, 109 (2024)

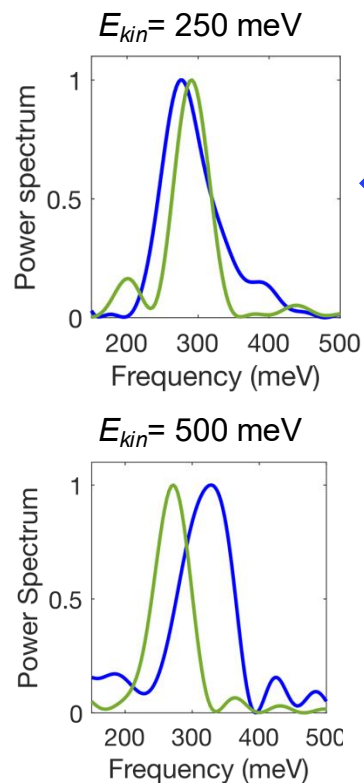


Comparison with theory (TDDFT)

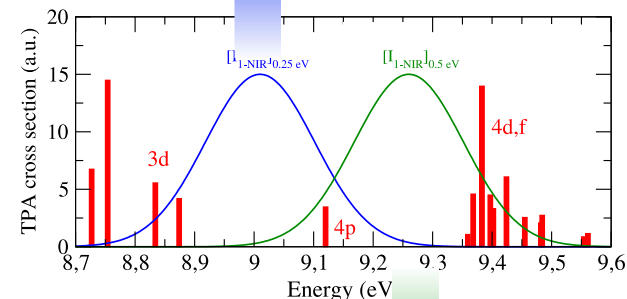
Time domain



Frequency domain



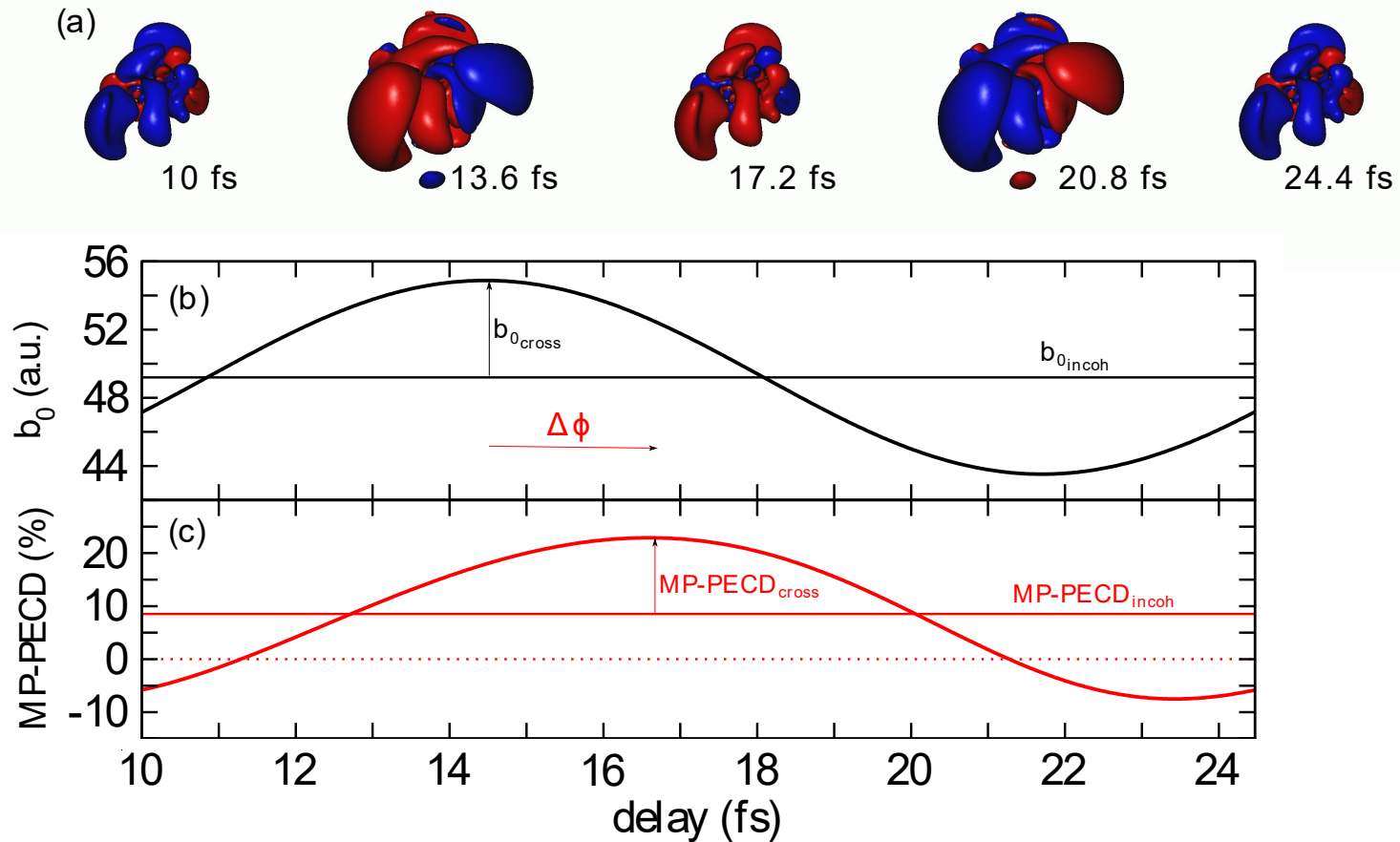
3d-4p coherence



4p-4d coherence

Coherent superposition of 3d-4p and 4p-4d,f states is responsible for the modulation of the PECD signal

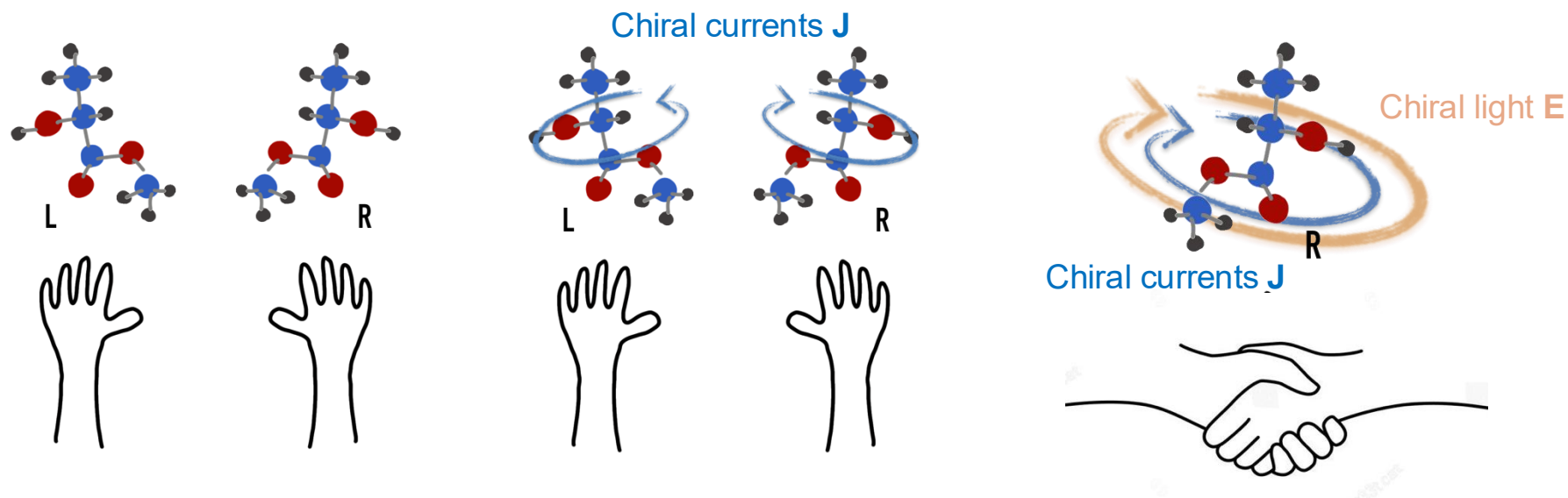
Charge migration



V. Wanie et al, Nature 630, 109 (2024)

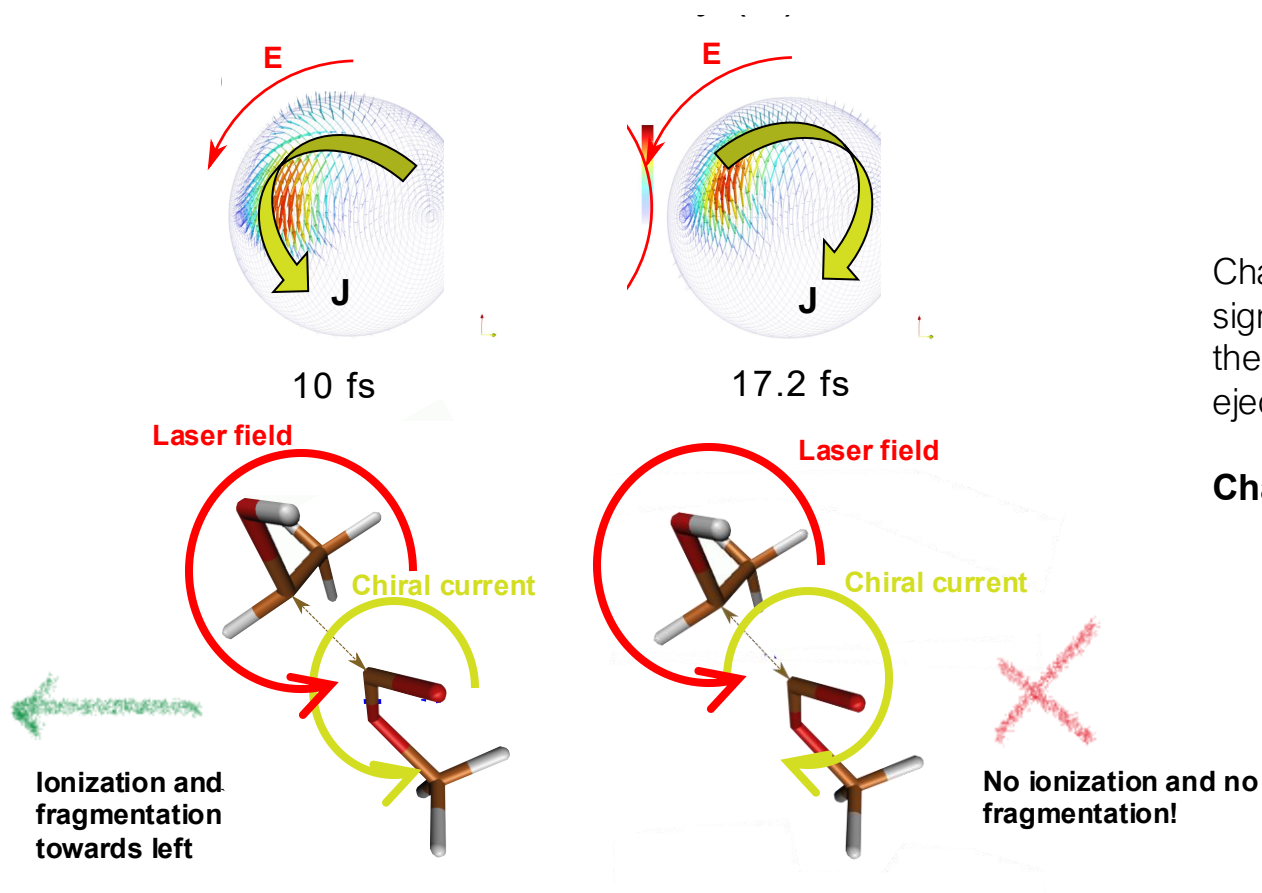
Chiral currents

Charge migration results in chiral currents switching sign with the periodicity of the electronic beatings.



Transient chiral currents enable **control of enantio-selective interactions**.

Molecular orientation effects

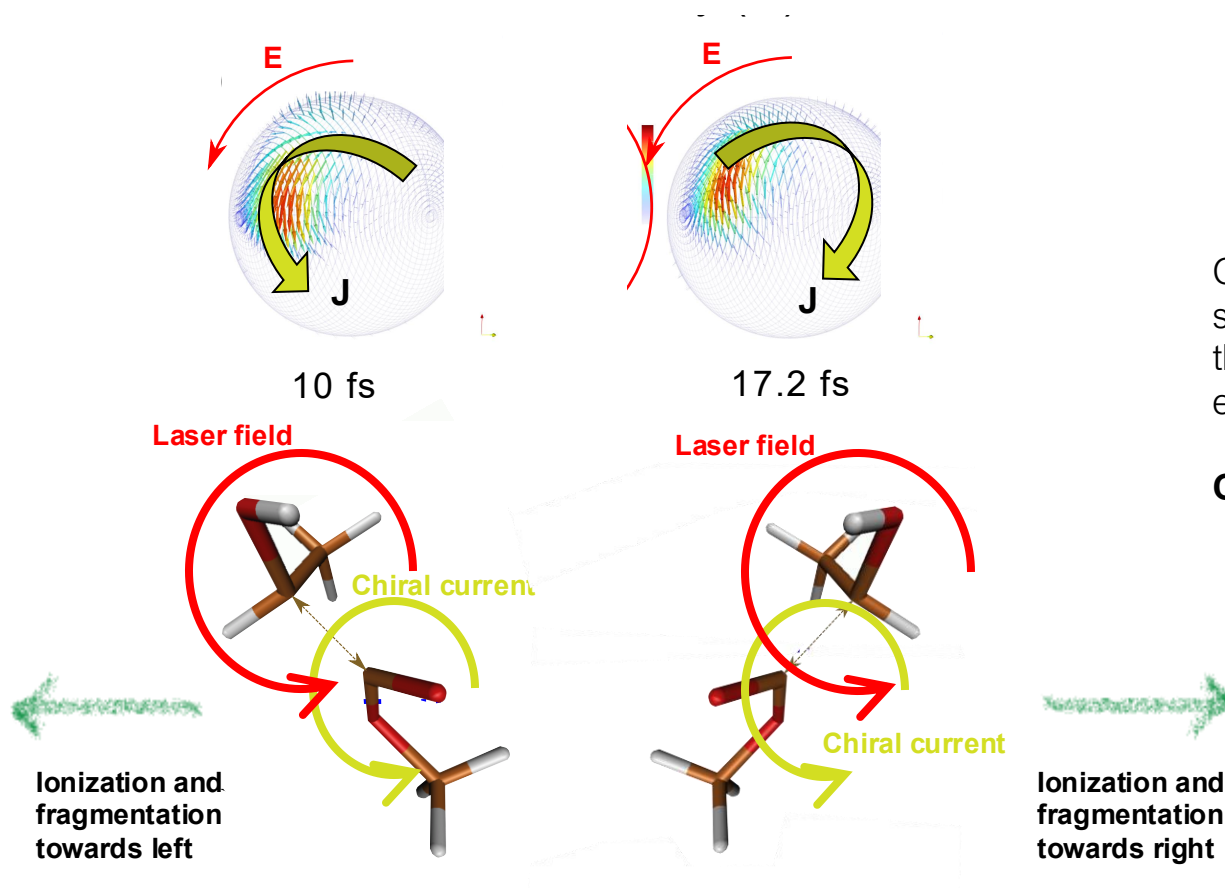


Charge migration dictates the sign of the chiral currents and the direction of the fragment ejection.

Charge-directed reactivity!

Collaboration with Olga Smirnova, MBI

Molecular orientation effects

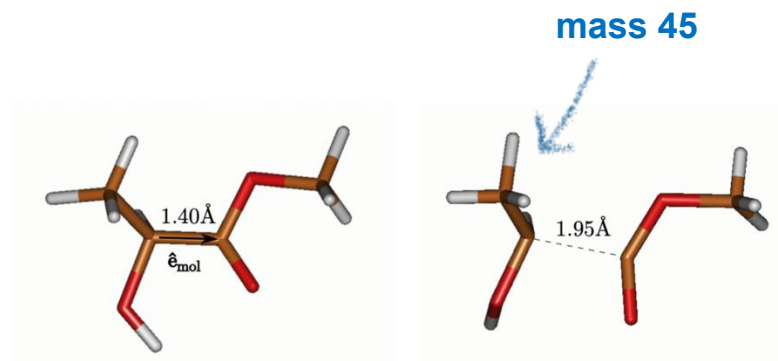
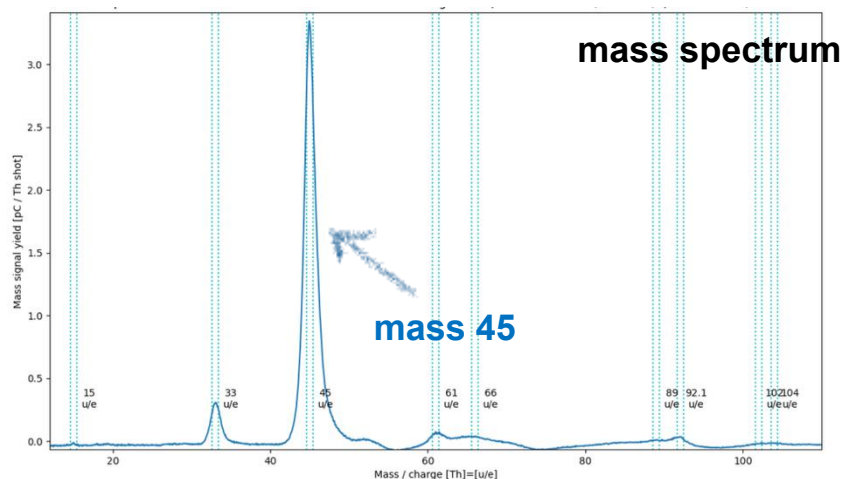


Charge migration dictates the sign of the chiral currents and the direction of the fragment ejection.

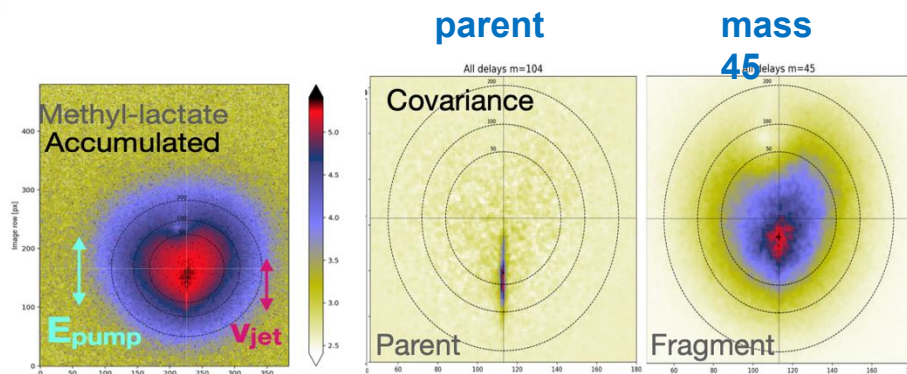
Charge-directed reactivity!

Collaboration with Olga Smirnova, MBI

Preliminary results in methyl lactate

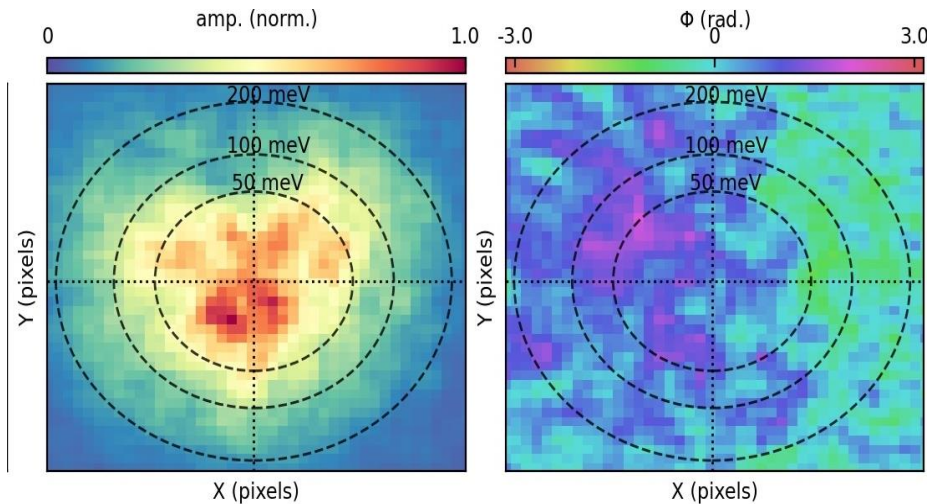


- Ion velocity map imaging (Ion VMI)
- Main contribution from fragment 45
- Breakage of C-C bond
- Covariance with ion ToF
- Measurement of RCP-LCP

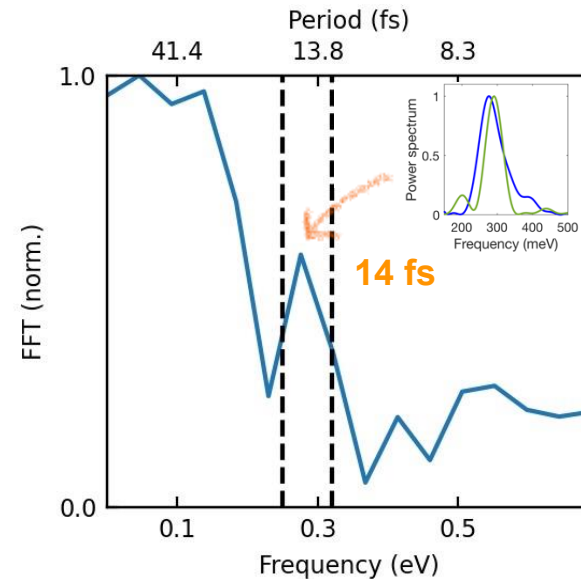


Time dependent asymmetry in the ions

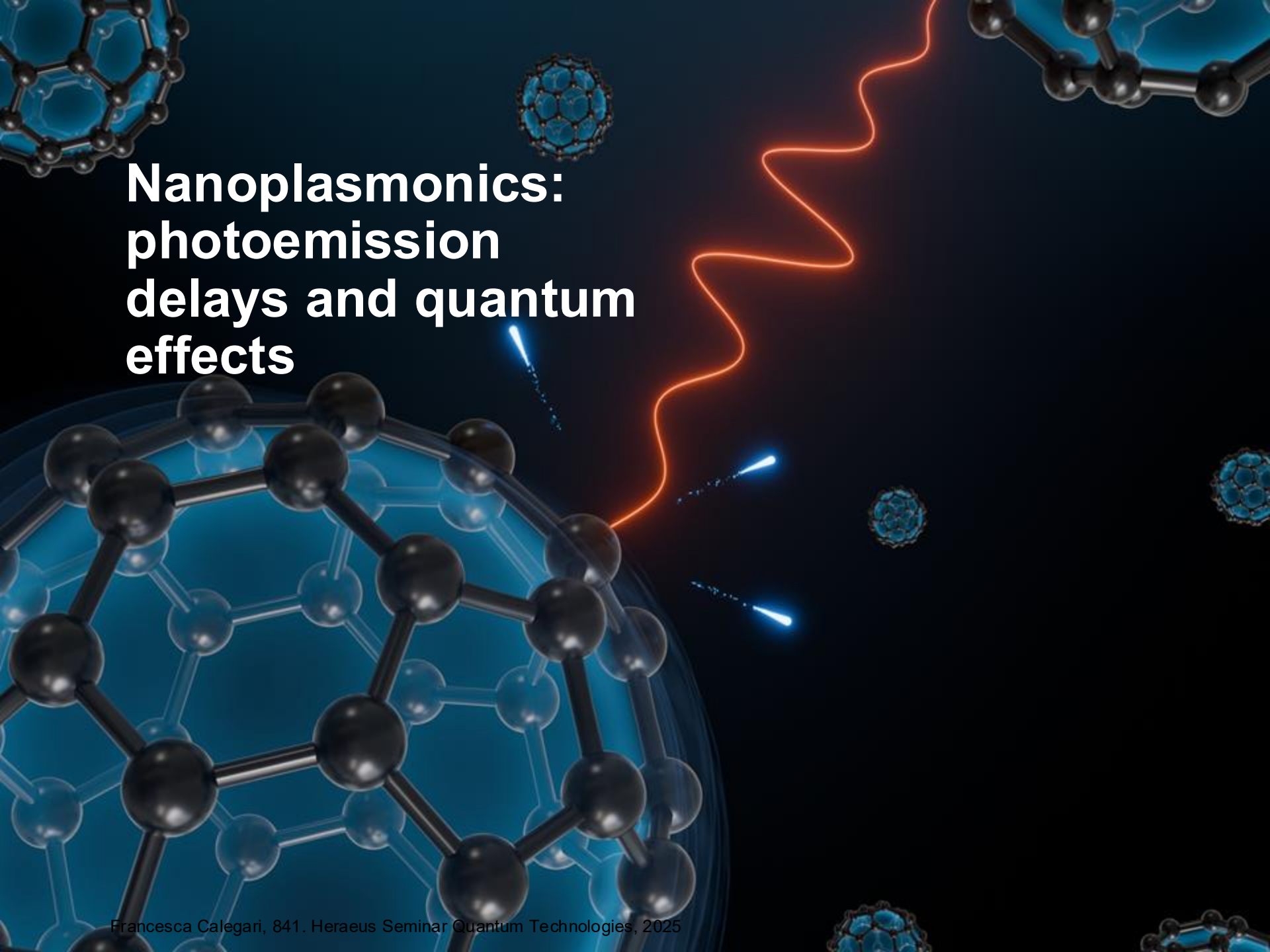
FFT analysis of the momentum distribution (RCP-LCP)



FFT amplitude at 200 meV



- Clear phase jump between the right and the left side of the FFT phase map
- FFT amplitude extracted at 200 meV shows the same periodicity of the PECD

The background of the slide is a dark blue space filled with various molecular and nanoscale structures. In the foreground, a large, detailed carbon nanotube or graphene-like structure is visible, composed of black spheres (atoms) connected by bonds. Other smaller, similar structures are scattered in the background. A prominent orange wavy line, representing a surface plasmon polariton, curves across the right side of the image. Several bright blue streaks, representing photoemitted electrons, are shown originating from the surface of the nanotube and moving away from it.

Nanoplasmonics: photoemission delays and quantum effects

Attosecond chronoscopy of photoemission

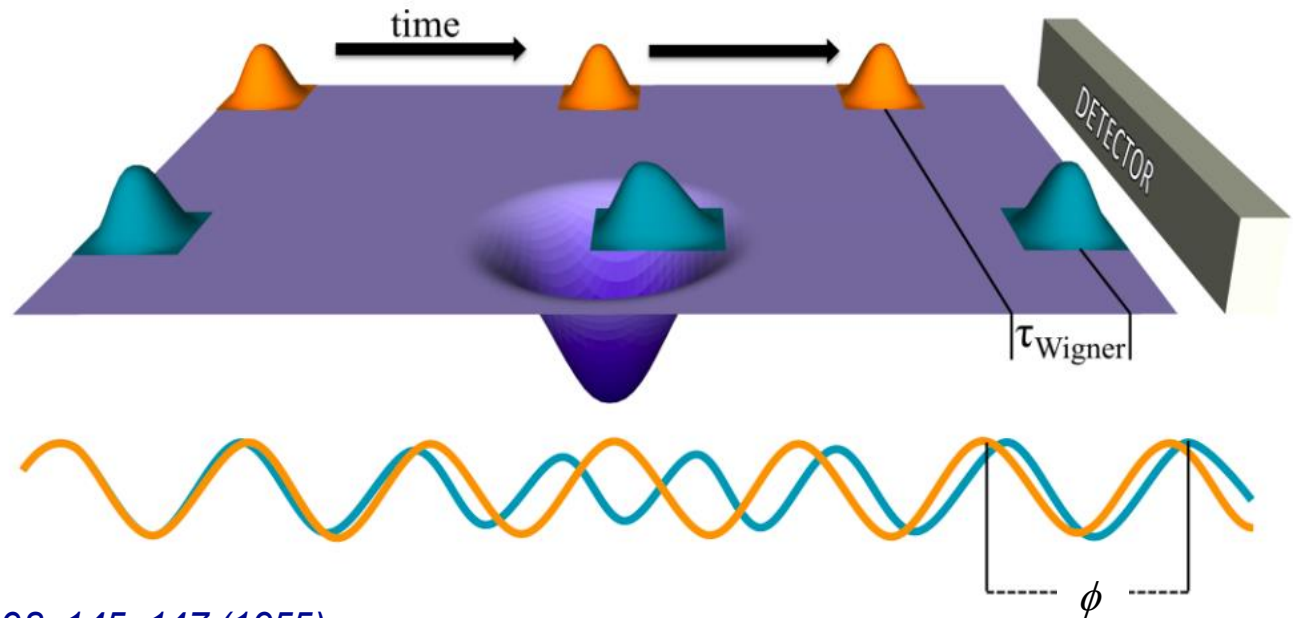
We could see ionization as half-collision

Scattering phase: associated with an **advance** or **retardation** of the outgoing electron wavepacket caused by its interaction with the **scattering potential**.

Eisenbud–Wigner–Smith (EWS) delay: energy derivative of the partial-wave scattering phase

$$\tau_W = \frac{\partial \phi(\epsilon, \omega)}{\partial \epsilon}$$

Group delay
accumulated by
the electron
wave-packet



E. P. Wigner, Phys. Rev. 98, 145–147 (1955)

L. Eisenbud, Ph. D. thesis, Princeton University (1948)

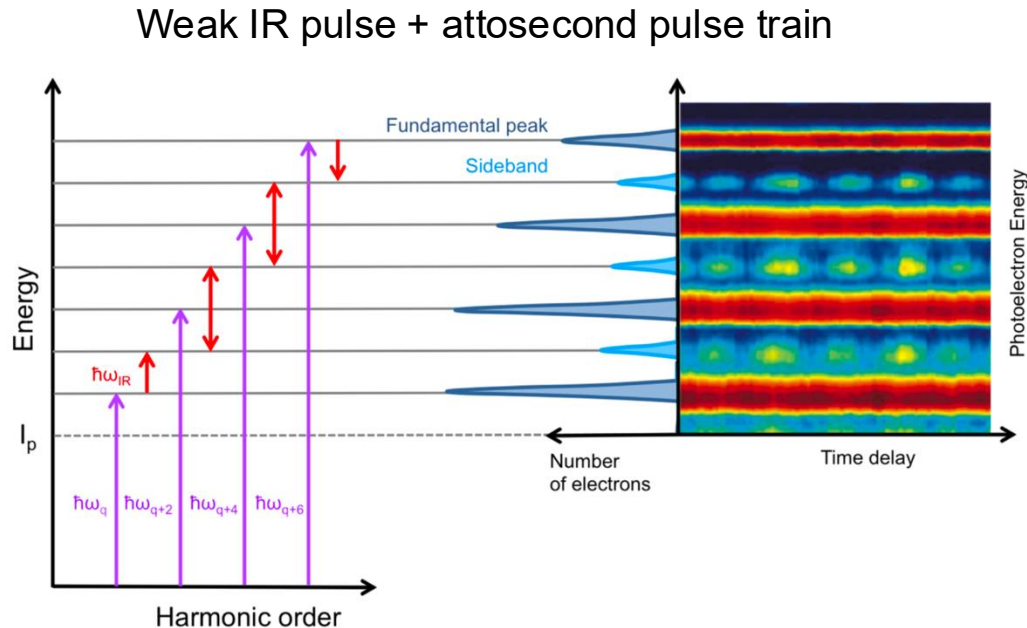
F.T. Smith, Phys. Rev. 118, 349 (1960)

R. Pazourek et al, Rev. Mod. Phys. 87, 765–802 (2015)

L. Argenti, Phys. Rev A 95, 043426 (2017)

The RABBITT technique

RABBITT: reconstruction of attosecond beating by interference of two-photon transitions



Ill. Niklas Elmehed © Nobel Prize Outreach
Pierre Agostini
Prize share: 1/3

*P. M. Paul et al, Science
292,1689-1692 (2001)*

Sidebands oscillations:

$$SB \propto A \cos (2\omega_{IR}\tau - \text{Attochirp} - \text{Target scattering phase})$$

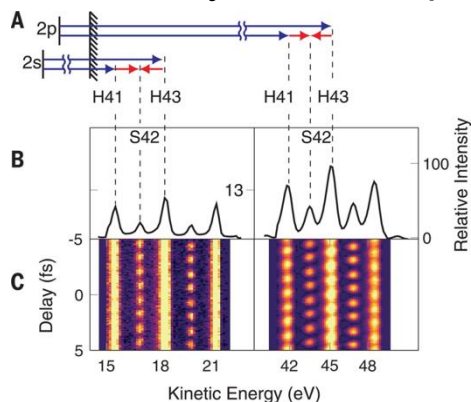
Attochirp
Target scattering phase

Extracted delay:

$$\tau_{\text{atomic}} = \hbar \frac{\partial \varphi_{\text{atomic}}}{\partial E} \cong \hbar \frac{\Delta \varphi_{\text{atomic}}}{\Delta E}$$

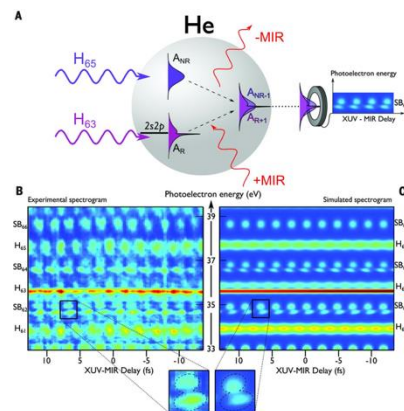
Photoemission delays extracted with RABBITT

Photoionization delay from Ne 2p and 2s



M. Isinger et al., Science 358,893-896 (2017)

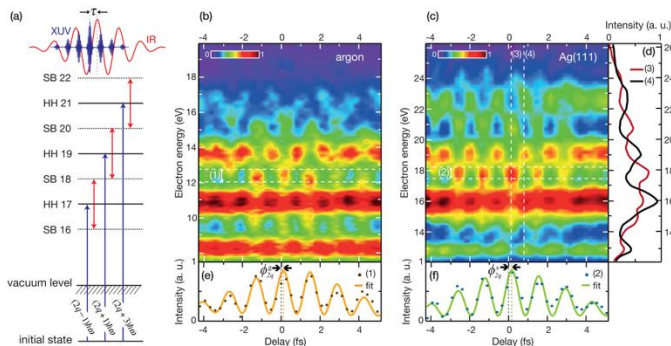
Autoionization in He



V. Gruson et al, Science 354,734-738 (2016)

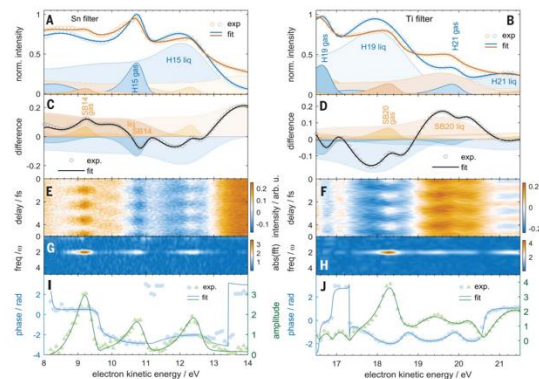
Delay extracted from Ag surface reveals transport mechanisms

R. Locher et al, Optica 2, 405–410 (2015)



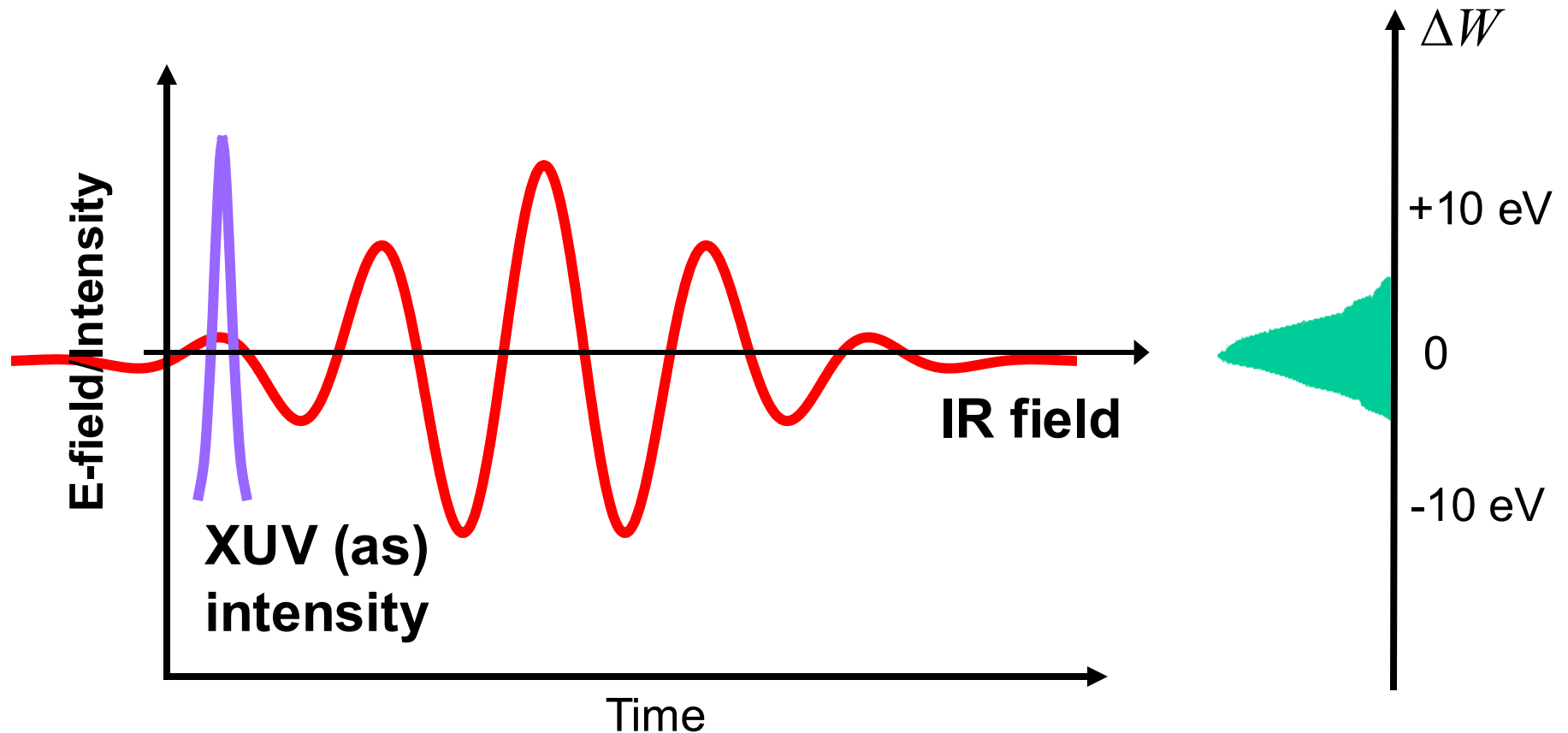
Photoemission delays extracted from liquid water

I. Jordan et al, Science, 369, 974-979 (2020)



The streaking technique

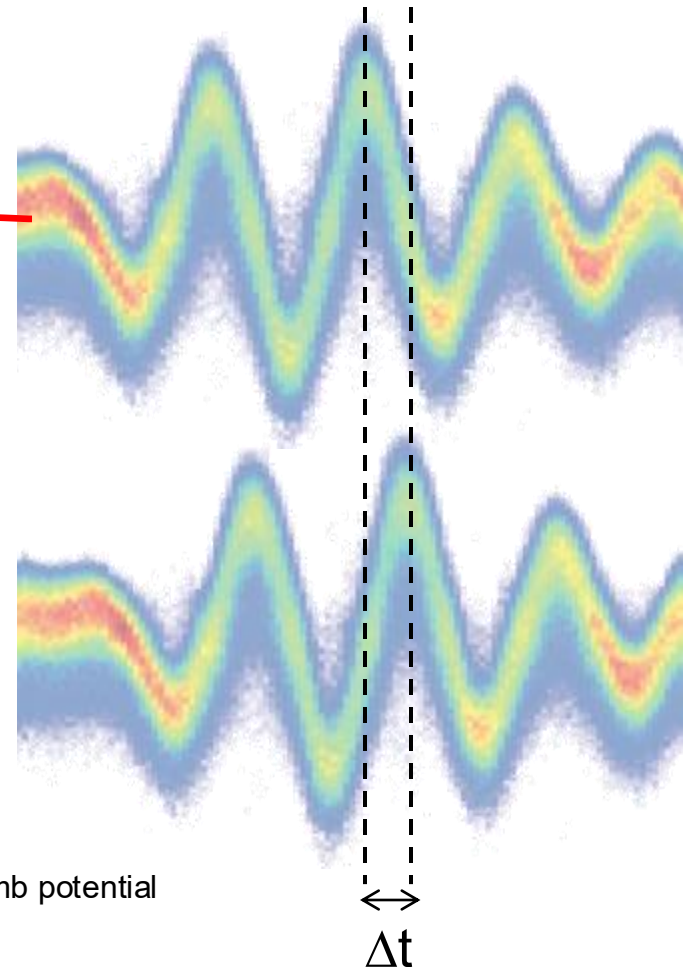
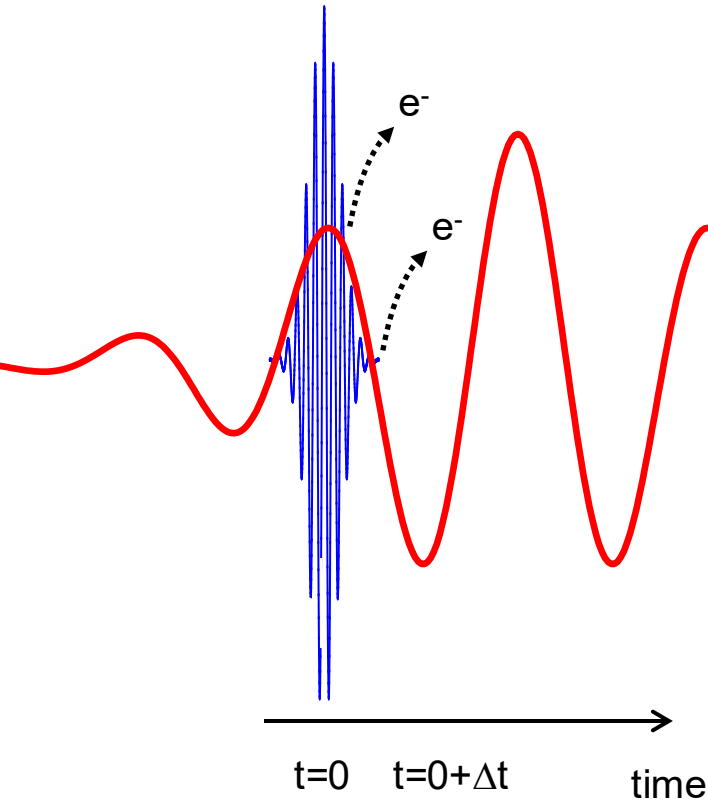
Attosecond streak camera



Hentschel, M. et al. Attosecond metrology. Nature 414, 509–513 (2001)

Extracting photoemission delays with attosecond streak camera

A delay in photoemission results in a delayed streaking trace



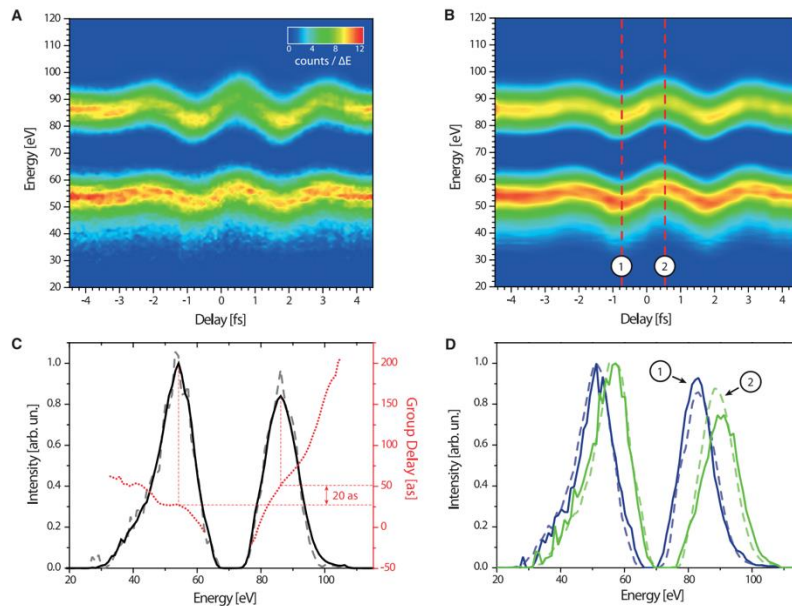
$$\Delta t = \tau_W + \tau_{CLC} + \tau_{attochirp}$$

τ_{CLC} : streaking field interacting with the asymptotic tail of the Coulomb potential

$\tau_{attochirp}$: residual chirp of the attosecond pulse

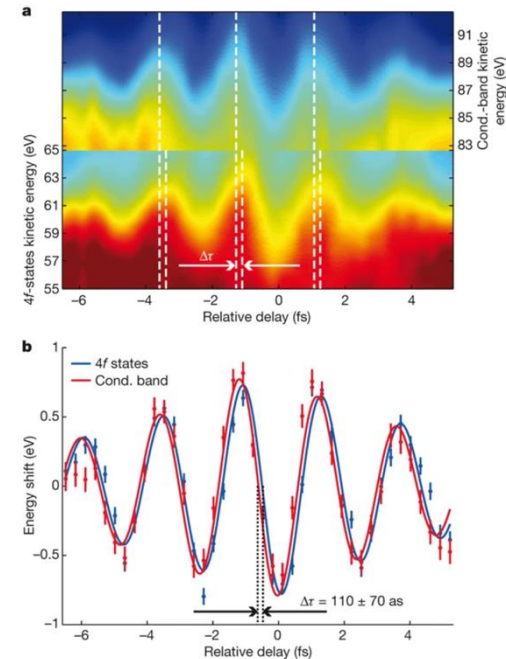
Photoemission delays with attosecond streak camera

Photoionization delay from Ne 2p and 2s



M. Schultze et al, Science 328, 1658-1662 (2010)

Photoemission delays from 4f states and conduction band of tungsten

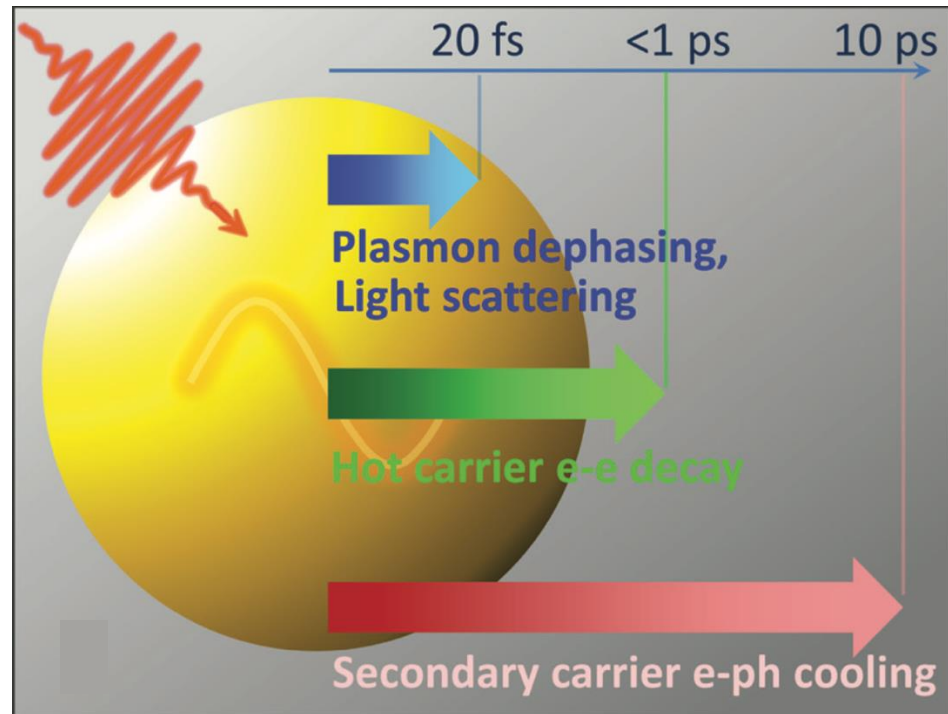


A. Cavalieri et al, Nature 449, 1029-1032 (2007)

Timescale of the surface plasmon evolution

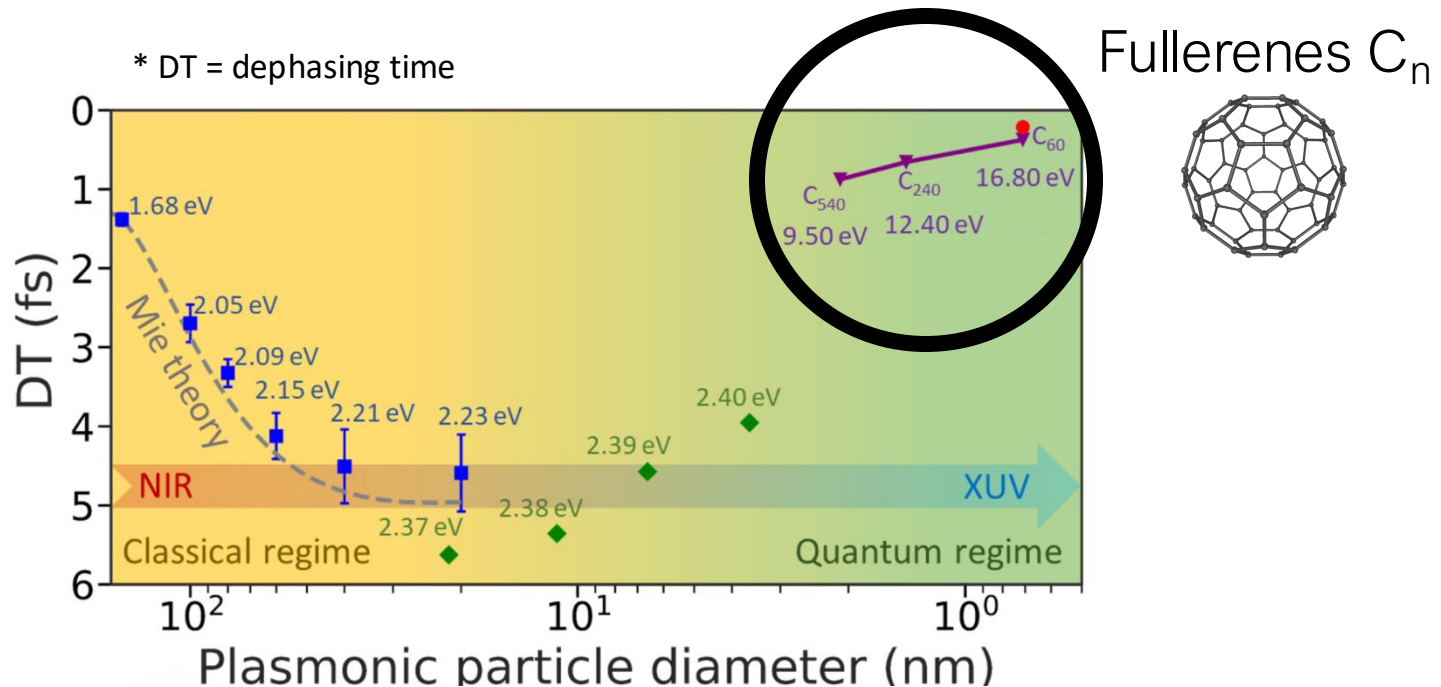
- Plasmon resonances near metal surfaces concentrates the incident radiation far beyond the limits of the geometrical optics
- Classic interpretation: collective electron dynamics
- Fast dephasing routes
- Important applications: **solar energy harvesting, sensing, photocatalysis.**

N. Kholmicheva et al, Nanophotonics, vol. 8, no. 4, 2019, pp. 613-628.



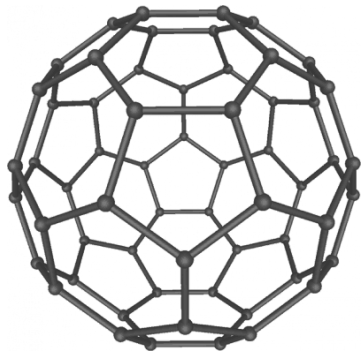
Plasmonics in the quantum limit

Sub-nm regime: quantum limit, ultrafast dephasing, role of electron correlations?

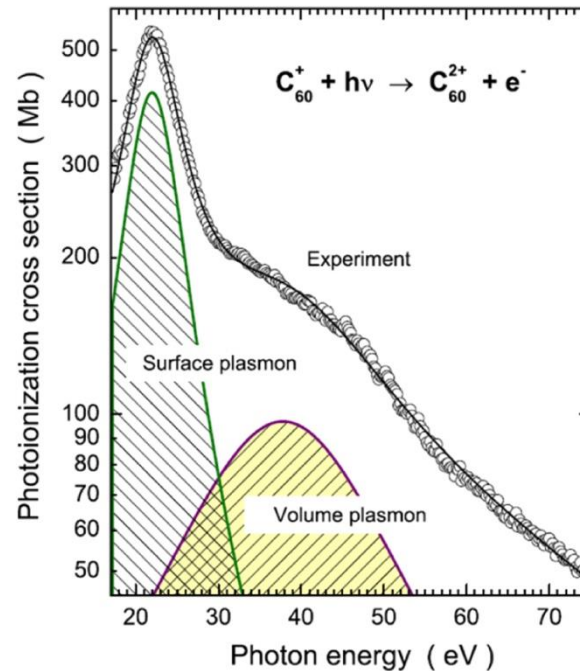


Sonnichsen, C., et al., New J Phys 4, 93 (2002)
Piella, J., et al., Chem Mater 28, 1066-1075 (2016)
Scholl, J. A., et al., Nature 483, 421-427 (2012)

The fullerene C₆₀



sub-nm diameter
240 valence electrons



T. Barillot et al, Phys. Rev. A 91, 033413 (2015)

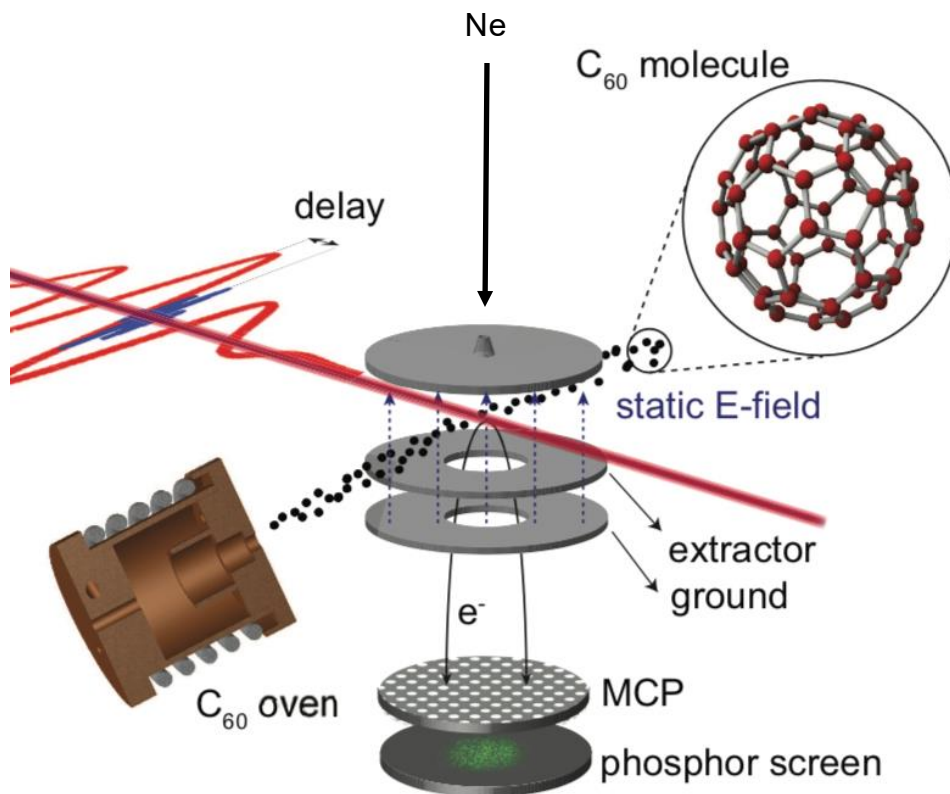
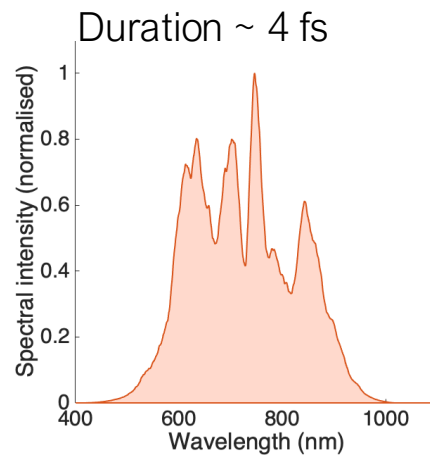
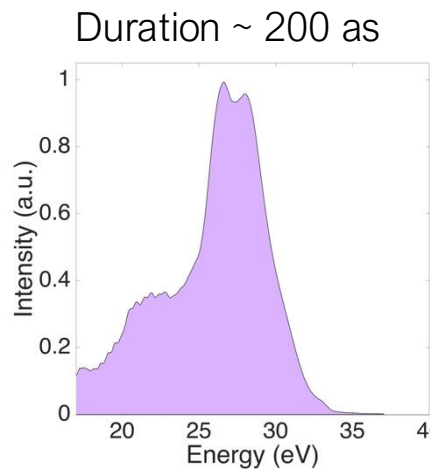
Hertel I, Phys. Rev. Lett., 68 784–7 (1992)

F. Lépine, J. Phys. B: Atomic, Mol. and Opt.Phys., 48, 12 (2015)

Ultrabroad (ultrafast) **giant plasmonic resonance** in the continuum (~20 eV, Ip ~7.6 eV).

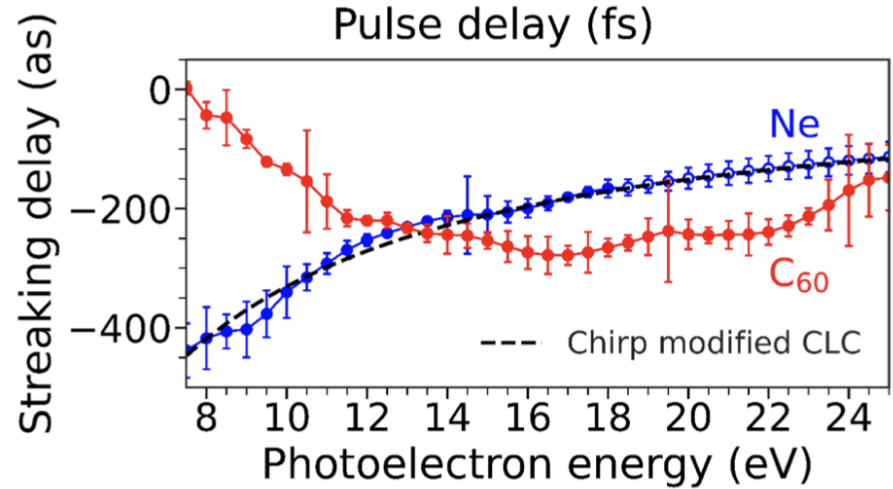
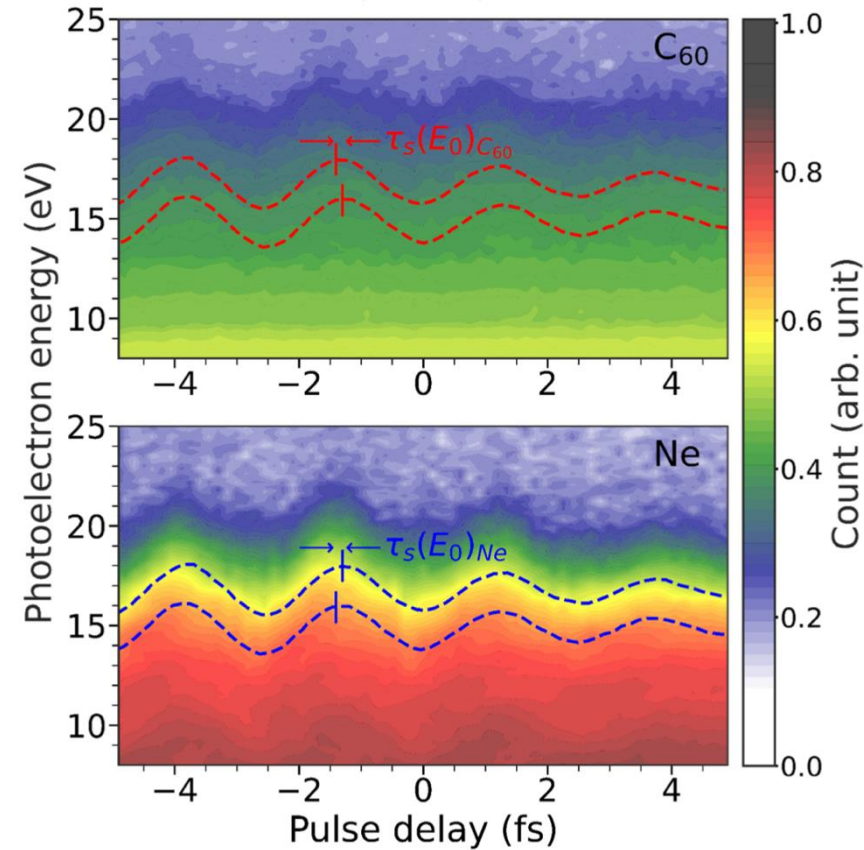
Attosecond-resolution spectroscopy is required.

The experiment: attosecond streaking setup



In collaboration with M. Kling, MPQ and Stanford and M. Nisoli, Politecnico di Milano

Extraction of photoemission delays from C₆₀



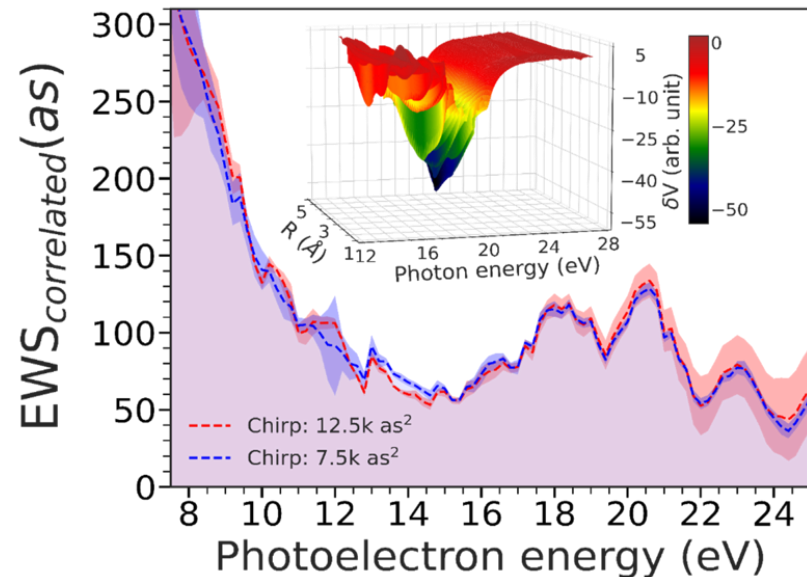
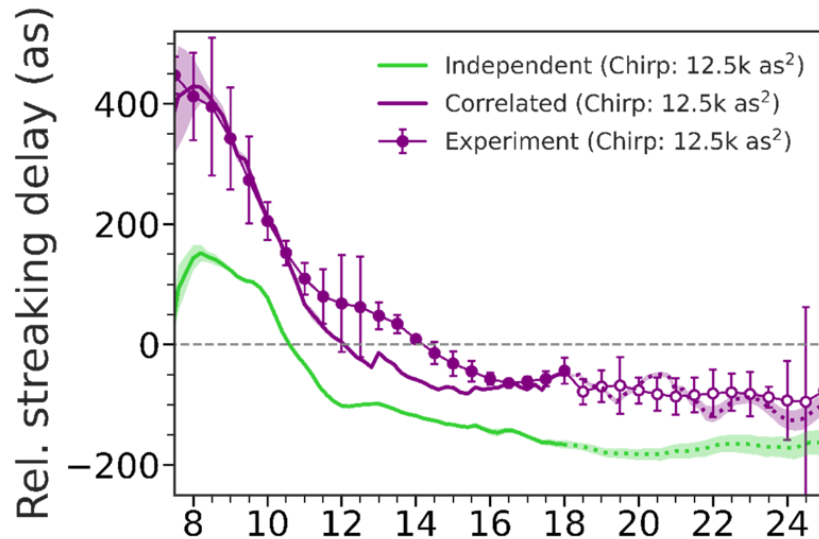
Streaking in neon is used to create a reference clock, characterize the attochirp and cancel out t_{CLC} :

$$\Delta t = (\tau_W + \cancel{\tau_{CLC}} + \cancel{\tau_{attochirp}} + \tau_{dipole})_{C60} - (\tau_W + \cancel{\tau_{CLC}} + \cancel{\tau_{attochirp}})_{Ne}$$

Role of quantum correlations

Theoretical modeling:

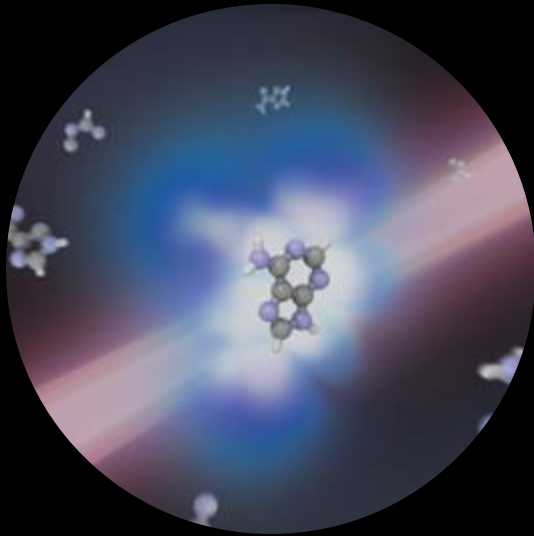
- Linear-response-TDDFT for the XUV-induced photoionization
- Classical trajectory Montecarlo simulations for the streaking where near-field effects are taken into account



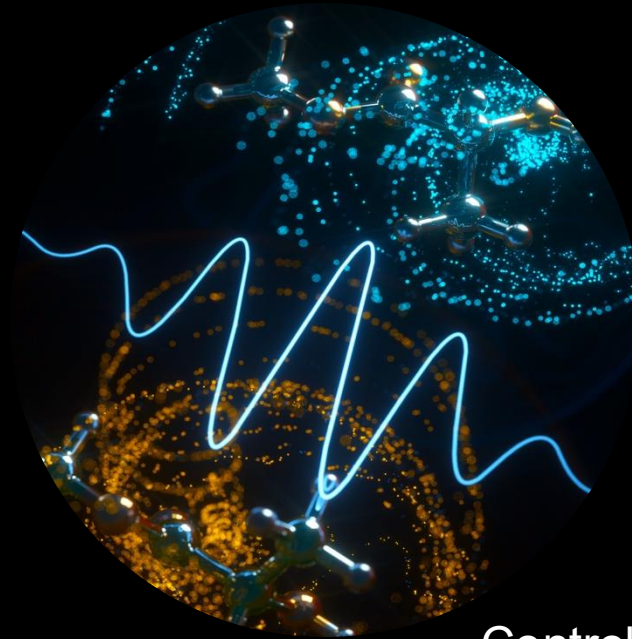
Photoionization is the fastest dephasing channel and it is **dominated by electron correlations**.

S. Biswas et al, Sci. Adv. 11, eads0494 (2025)

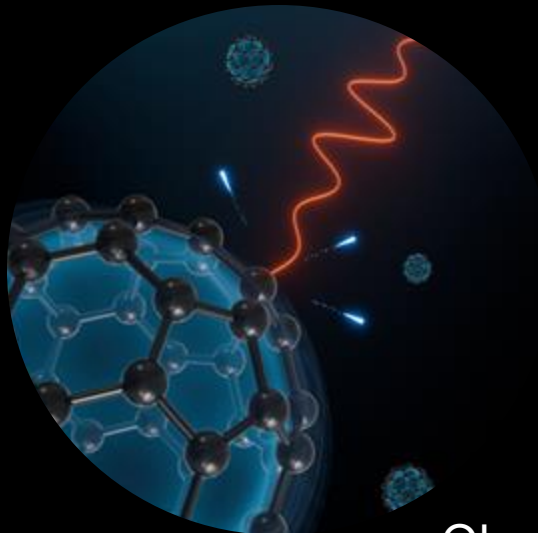
In collaboration with H. Himadri Chakraborty,
M. Madjet, M. Magrakvelidze, Missouri
University
U. De Giovannini, A. Rubio, MPSD, Hamburg



Observe electron
migration in
molecules



Control chiral optical
response



Observe quantum dynamics
in nanosystems

Attosecond Science Group at DESY- CFEL



- Attochemistry
- Ultrafast chirality
- Ultrafast nanoplasmonics
- Photoemission delays from complex targets
- Ultrafast dynamics in quantum materials

<https://atto.cfel.de>



Thank you!

DESY-CFEL/ UHH

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Lorenzo Pratolli
Ana Ferreira de Oliveira e Silva
Erik Månsson
Sergey Ryabchuk
Ammar Bin Wahid
Sabine Rockenstein
Oliviero Cannelli
Vincent Wanie
Terry Mullins
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CELIA

Valerie Blanchet
Etienne Bloch
Yann Mairesse
Bernard Pons

MBI

Olga Smirnova

Imperial College

David Ayuso
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Université P. Sabatier

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INRS

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MPSD

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University of Mary Washington

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