Lecture Series Buenos Aires

18-3-2024 until 22-3-2024

Lecture M8 – Attosecond solid state physics

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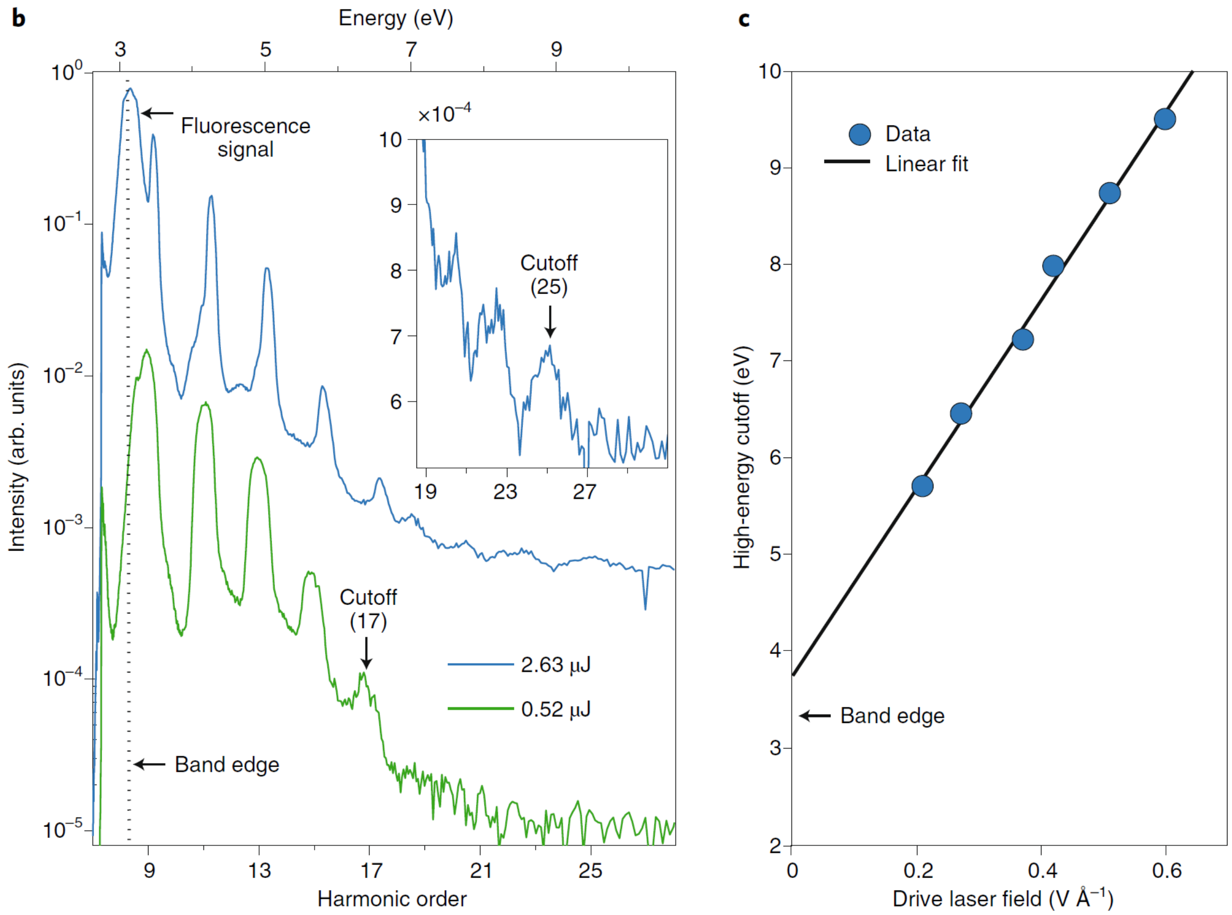
Contents

- High-harmonic generation in solid samples
- Attosecond and femtosecond time-resolved pump-probe spectroscopy in solid samples
  - Strong field-driven electron dynamics
  - Perturbative electron dynamics

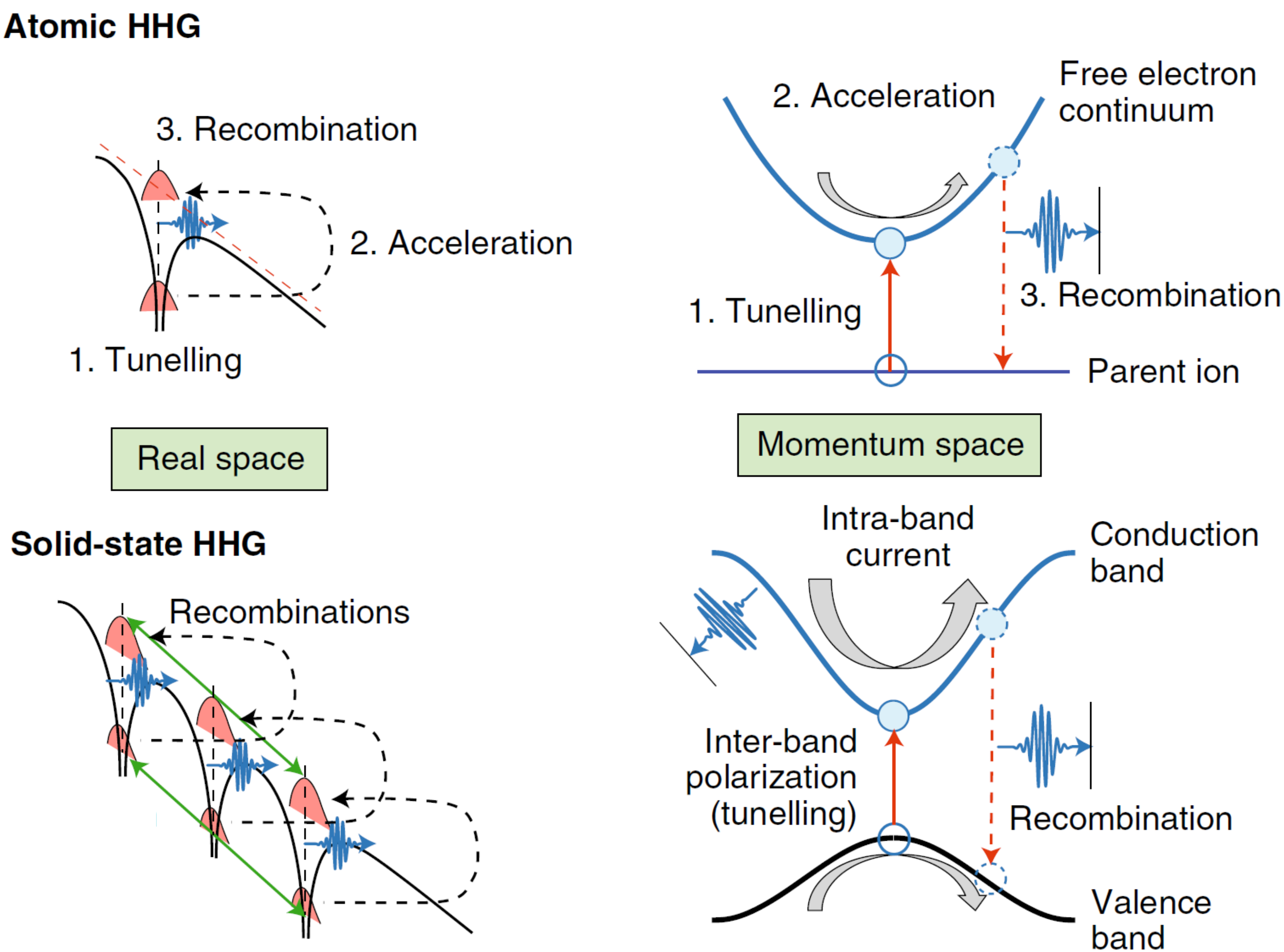
Discovery of HHG in solids



High-order harmonic generation in ZnO crystals: (left) measured spectra for two pulse energies; (right) high-energy cutoff as a function of the peak field, along with a linear fit. The intercept along the energy axis is near the bandgap (3.2 eV)

Ghimire, Nat. Phys. 7, 138 (2011)

Mechanism: intraband vs interband



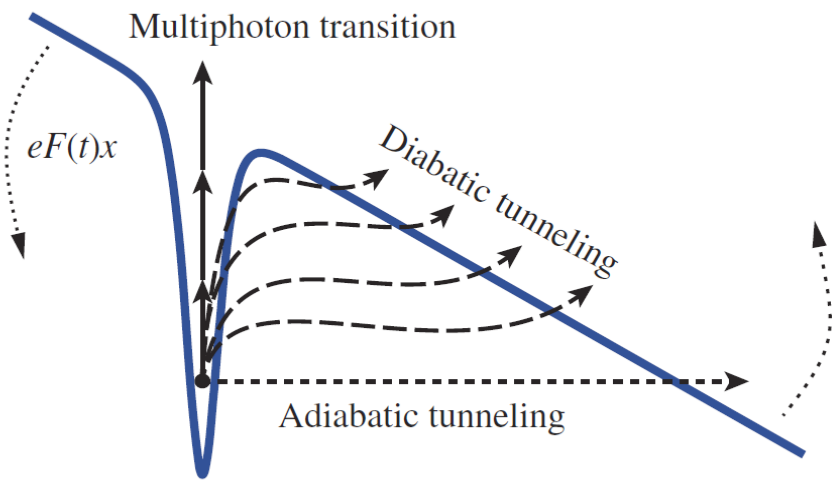
Ghimire, Nat. Phys. 7, 138 (2011)

Attosecond and femtosecond time-resolved pump-probe spectroscopy in solid samples

Strong field-driven electron dynamics

Strong field interactions: atoms vs. solids

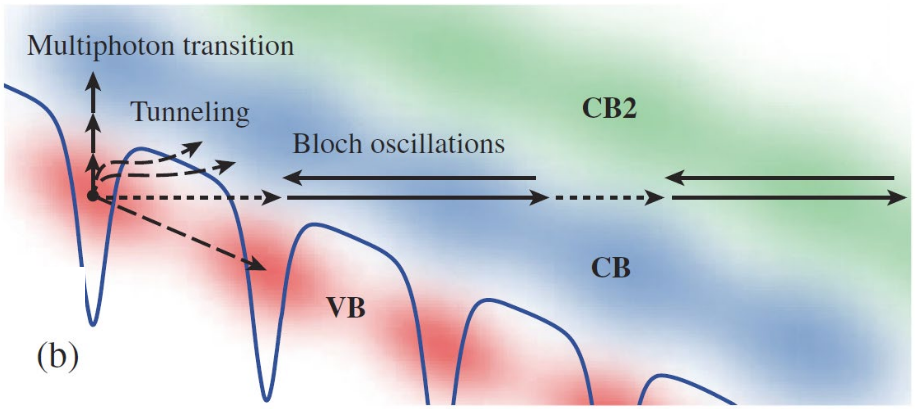
Atom in strong field



- When γ<<1, the ionization proceeds by adiabatic tunneling – horizontal process
- When γ>>1, the ionization proceeds by multiphoton absorption – vertical process
- When γ≈1, non-adiabatic tunneling

M. Y. Ivanov et al. J. Modern Optics 52, 165 (2005)

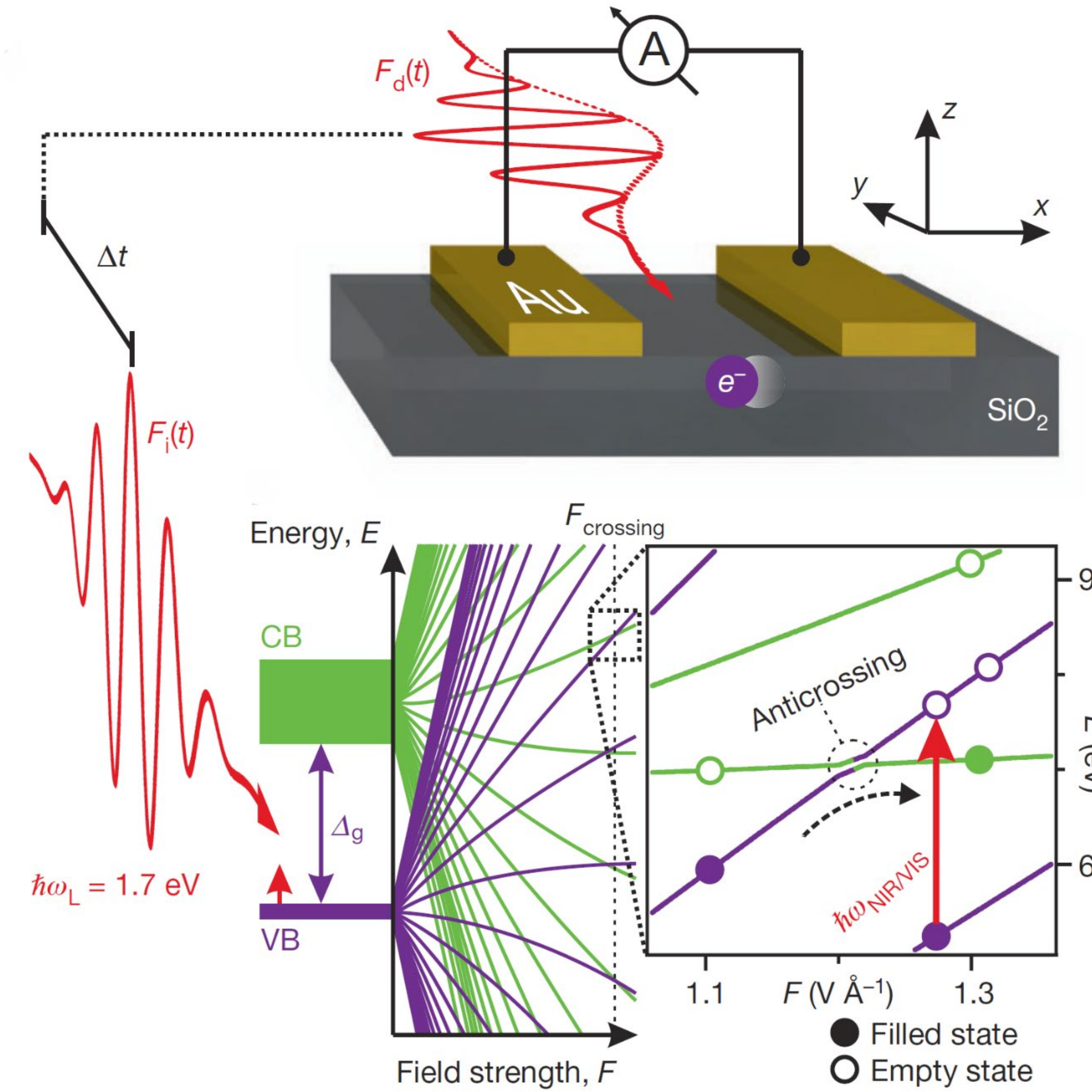
Periodic system in strong field



- When γ<<1, the ionization *intraband* motion leads to adiabatic tunneling and transfer from the valence (VB) band to the conduction (CB) band
- When γ>>1, (multi)-photon *interband* transitions from the valence (VB) band to the conduction (CB) band

S. Y. Kruchinin, Rev. Mod. Phys. 90, 021002 (2018)

First: an attosecond experiment without attosecond pulses



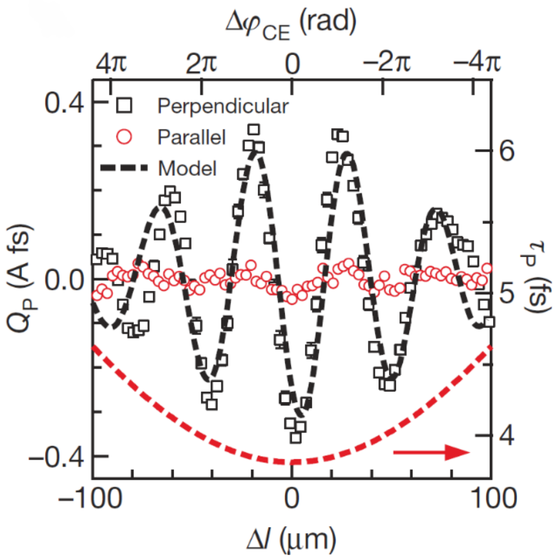
Excitation of a SiO_2 sample (dielectric, negligible conductivity) with an intense, few-cycle near-IR laser

Measurement, using two electrodes, of a photo-induced current

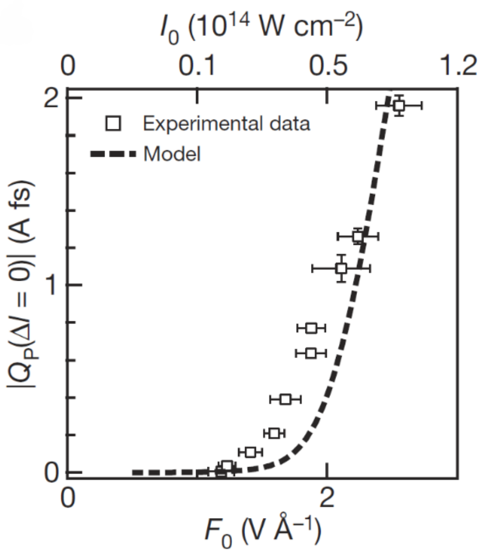
N.B. Bandgap 9 eV, photon energy 1.7 eV → falls within adiabatic (tunneling) picture

A. Schiffrin et al., Nature 493, 70 (2013)

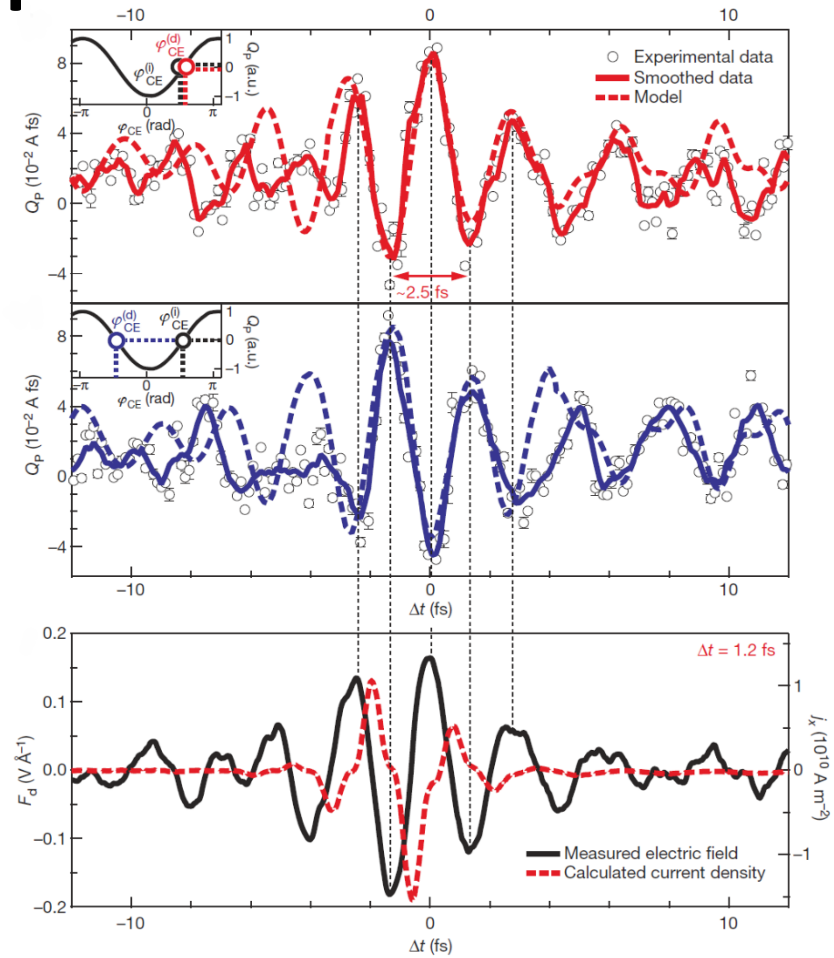
First: an attosecond experiment without attosecond pulses



Observation of a current when the laser polarization is perpendicular to the electrodes, with the direction controlled by the CEP



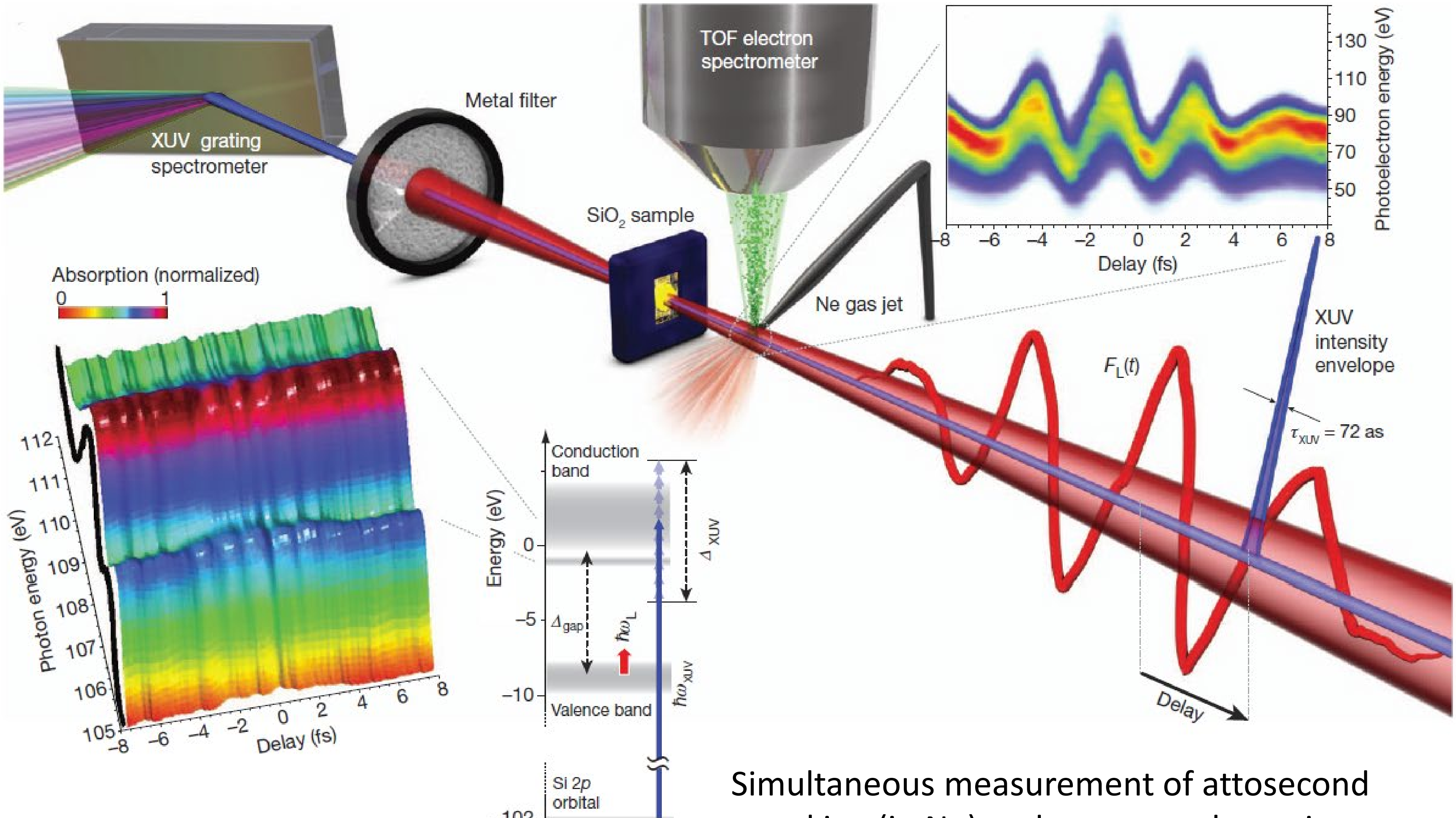
Laser electric field dependence of the current, revealing the high non-linearity involved in the process



Two-pulse experiment with a strong pulse (2 V/cm) polarized parallel to the electrodes creating the conduction band population and a weak pulse (0.2 V/cm) driving the current + comparison of the field of the latter pulse to attosecond streaking

A. Schiffrin et al., Nature 493, 70 (2013)

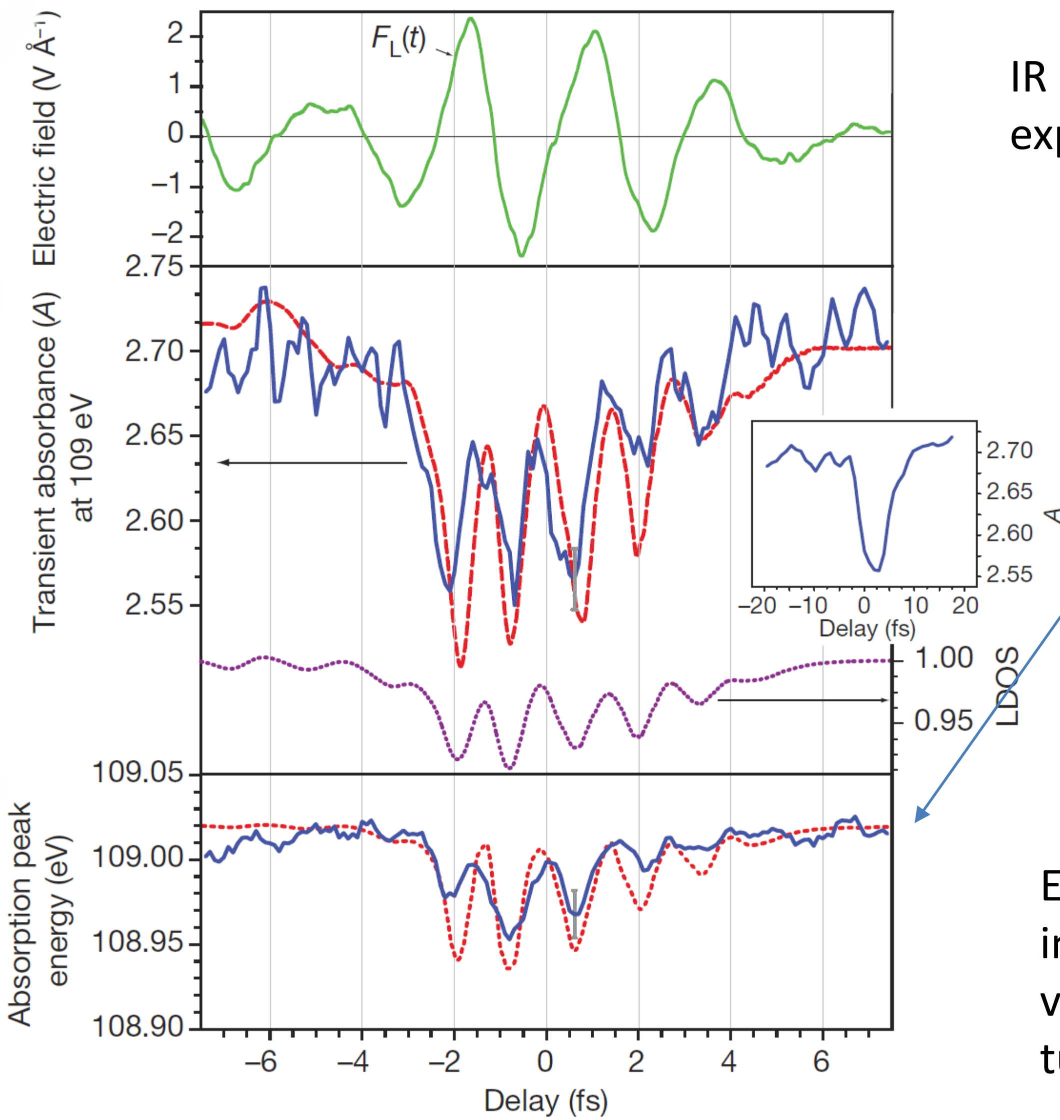
Next: using attosecond transient absorption to probe the injection process



Simultaneous measurement of attosecond streaking (in Ne) and attosecond transient absorption (in SiO_2)

M. Schultze et al., Nature 493, 75 (2013)

Next: using attosecond transient absorption to probe the injection process



IR driver pulse (determined by streaking experiment

Experimental transient absorption into conduction band (blue) and comparison to theory (red)

Transient lineshifts in the experiment (blue) in theory (red)

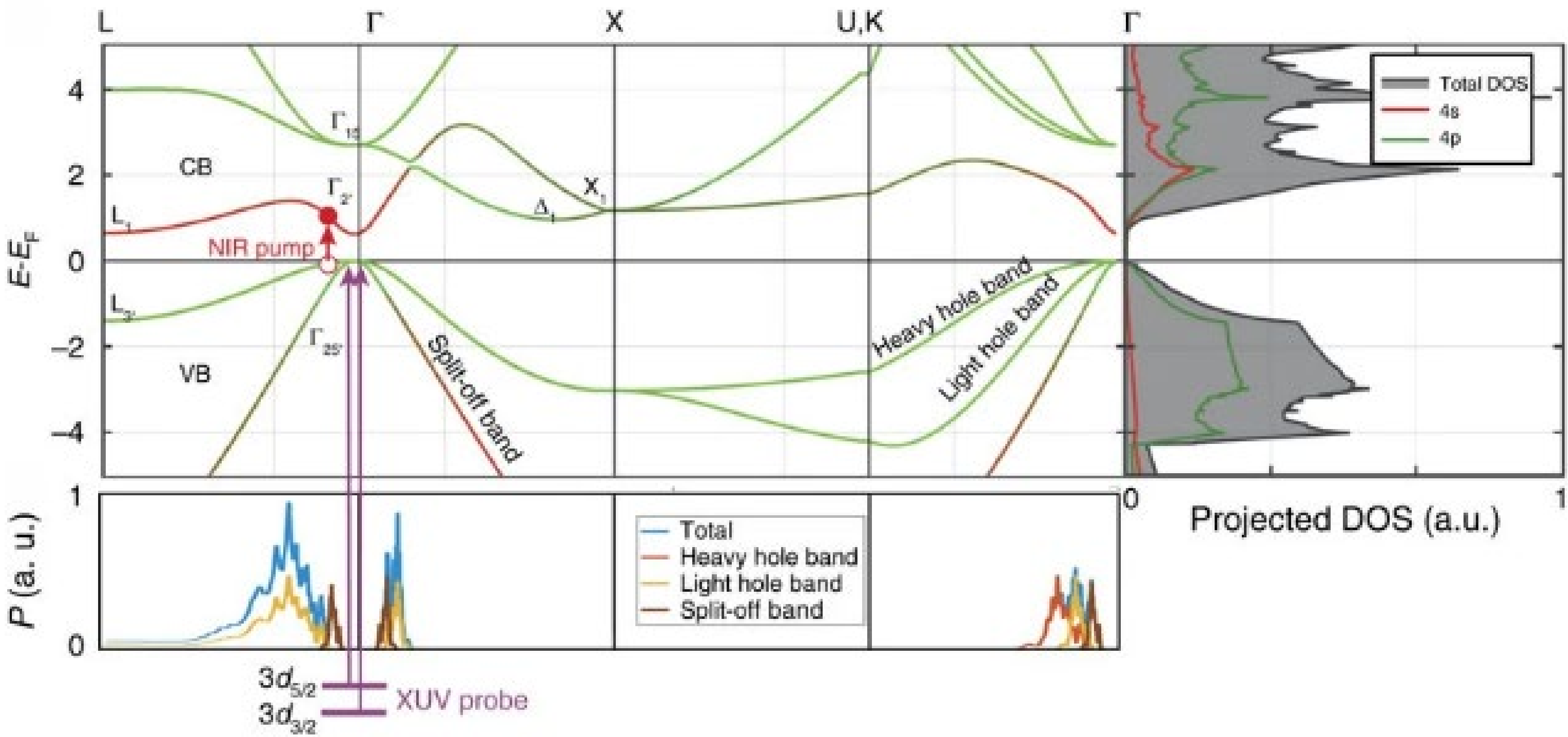
Evidence of a reversible, near-instantaneus response – production of virtual carriers in the CB by field-driven tunneling

M. Schultze et al., Nature 493, 75 (2013)

Attosecond and femtosecond time-resolved pump-probe spectroscopy in solid samples

Perturbative electron dynamics

Transient soft X-ray spectroscopy in Germanium



IR/VIS excitation from the valence (VB) to the conduction (CB) band

What are the relaxation times and the relaxation mechanisms of the CB electrons and VB holes that are produced?

Approach: excited $3d_{3/2}$ and $3d_{5/2}$ core electrons into available states (i.e. 4p character)

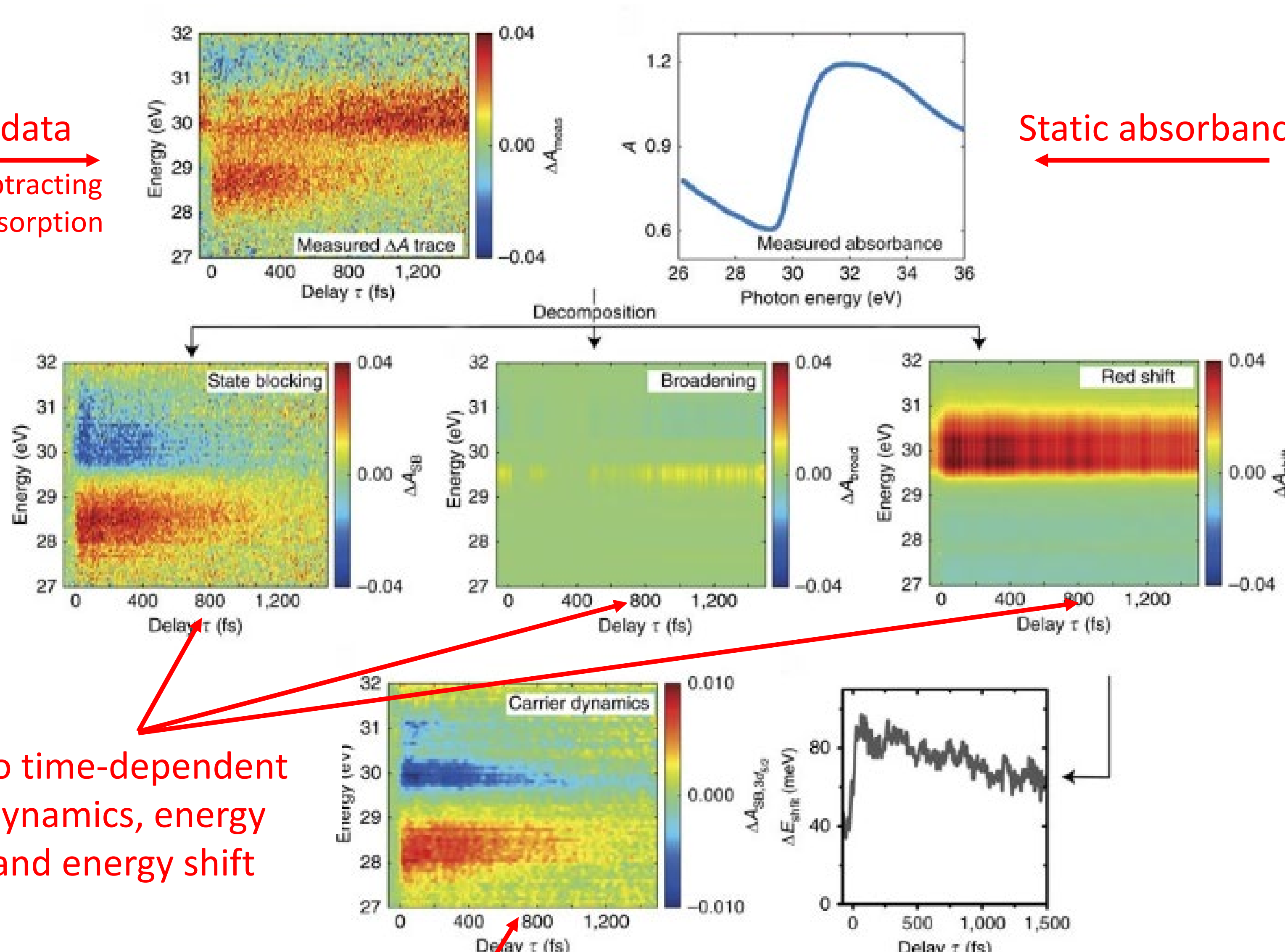
M. Zürch et al., Nat. Comm. 8, 15734 (2017)

Transient soft X-ray spectroscopy in Germanium

Raw data

after subtracting static absorption

Static absorbance

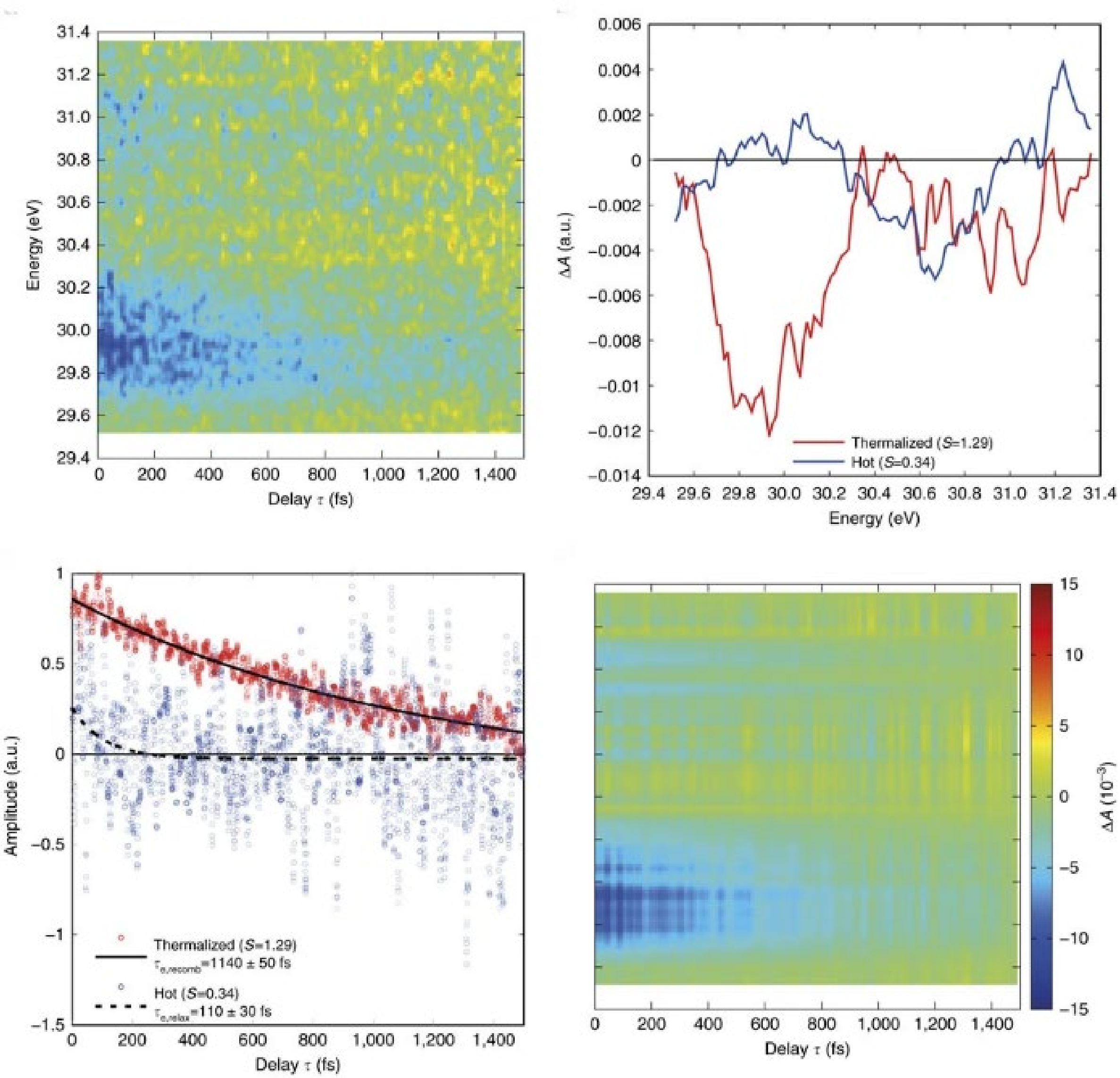


Separate into time-dependent Population dynamics, energy broadening and energy shift

Deconvolute $3d_{3/2}$ and $3d_{5/2}$

M. Zürch et al., Nat. Comm. 8, 15734 (2017)

Transient soft X-ray spectroscopy in Germanium

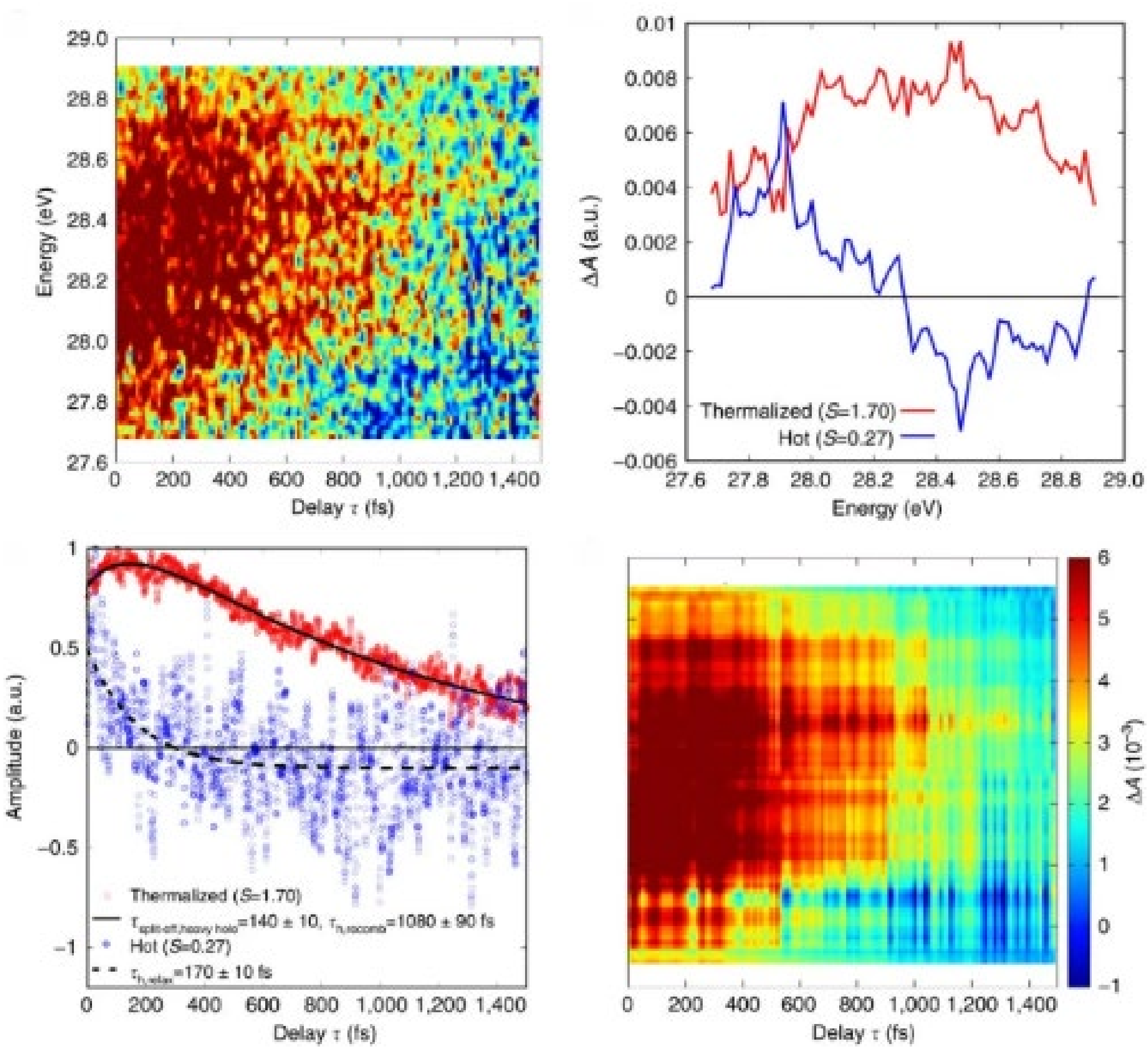


Electron dynamics (higher photon energies)

- Separate the measured response into two parts using singular value decomposition (SVD)
- Slower component (~1 ps): electron-hole recombination
- Faster component (~140 fs): electron relaxation
- Dependence of observed relaxation time on XUV photon energy: relaxations from valleys in CB

M. Zürch et al., Nat. Comm. 8, 15734 (2017)

Transient soft X-ray spectroscopy in Germanium

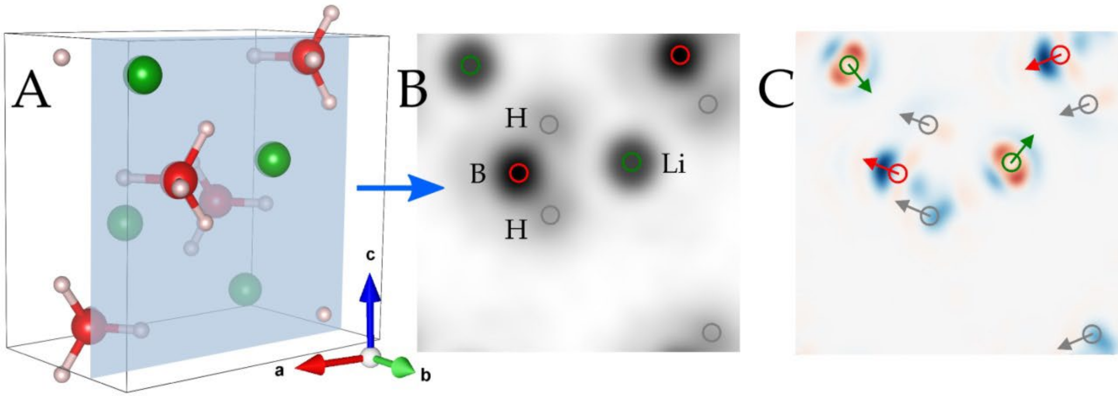


Hole dynamics
(lower photon energies)

- Separate the measured response into two parts using singular value decomposition (SVD)
- Slower component (~1 ps): electron-hole recombination
- Faster component (~140 fs): scattering between different hole states
- Additional fast component (~170 fs): hole relaxation

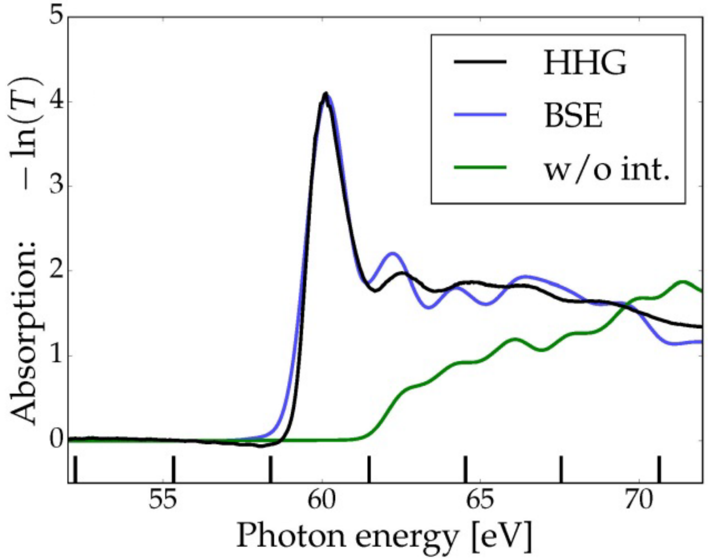
M. Zürch et al., Nat. Comm. 8, 15734 (2017)

Correlated electronic/nuclear motion in $LiBH_4$

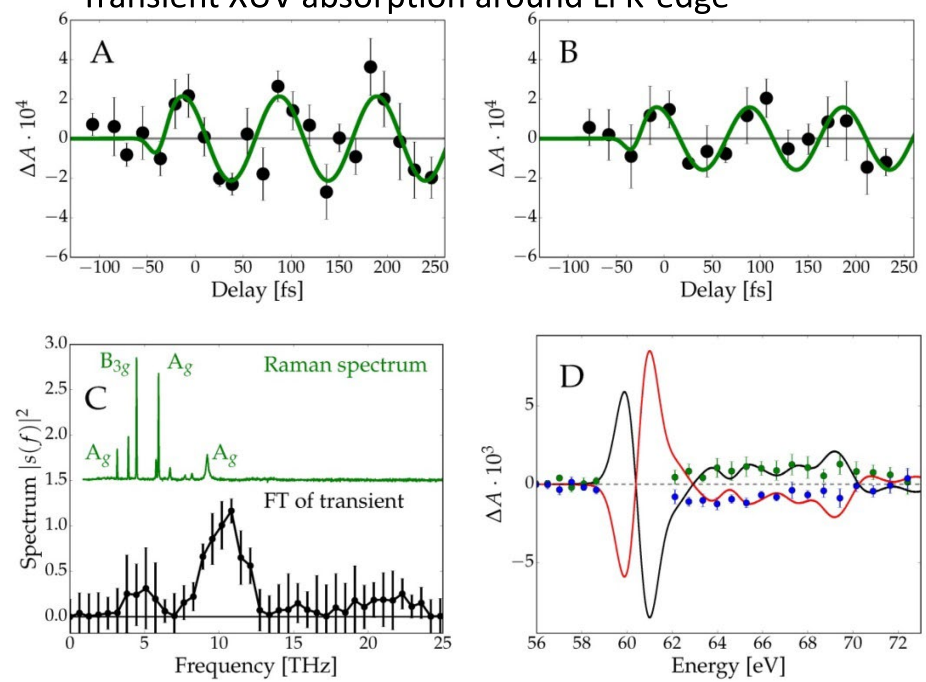


Time-resolved X-ray diffraction
T. Elsaesser et al., Phys. Rev. Lett 109, 147402 (2012)

Steady-state XUV absorption around Li K-edge



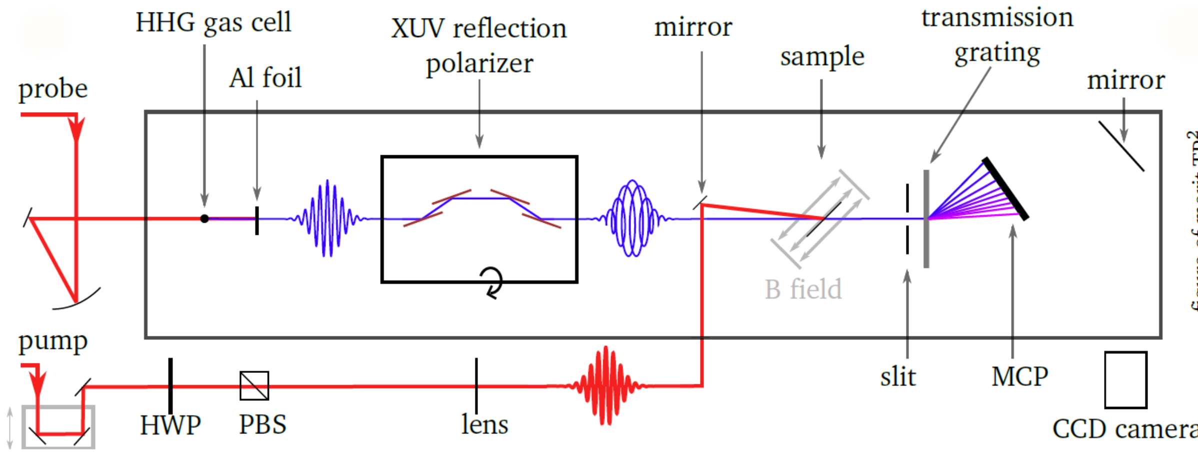
Transient XUV absorption around Li K-edge

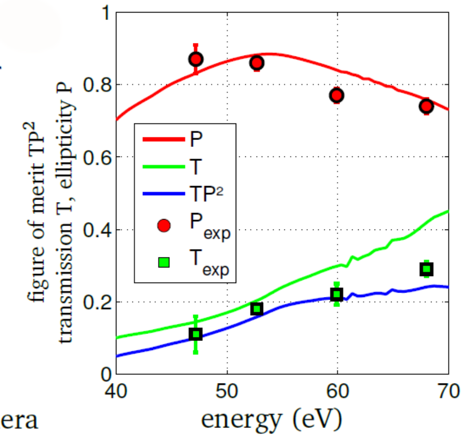


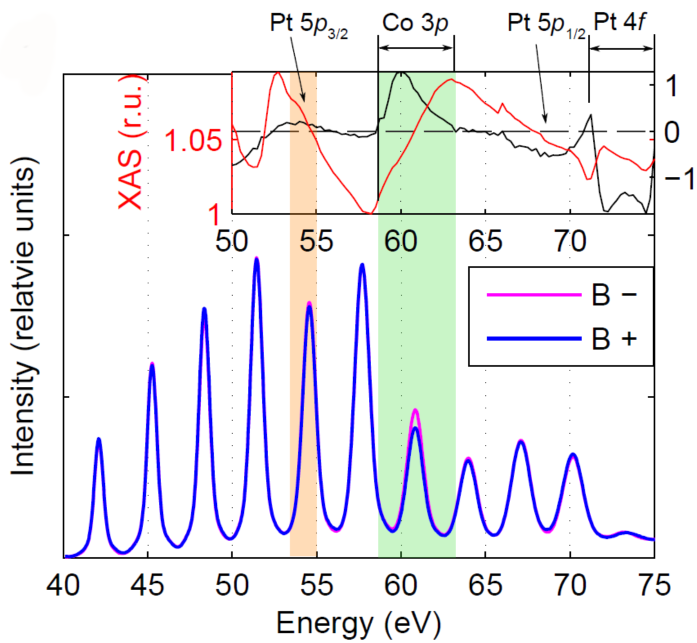
Time-resolved X-ray diffraction and XUV absorption reveal correlated electronic and nuclear dynamics!!

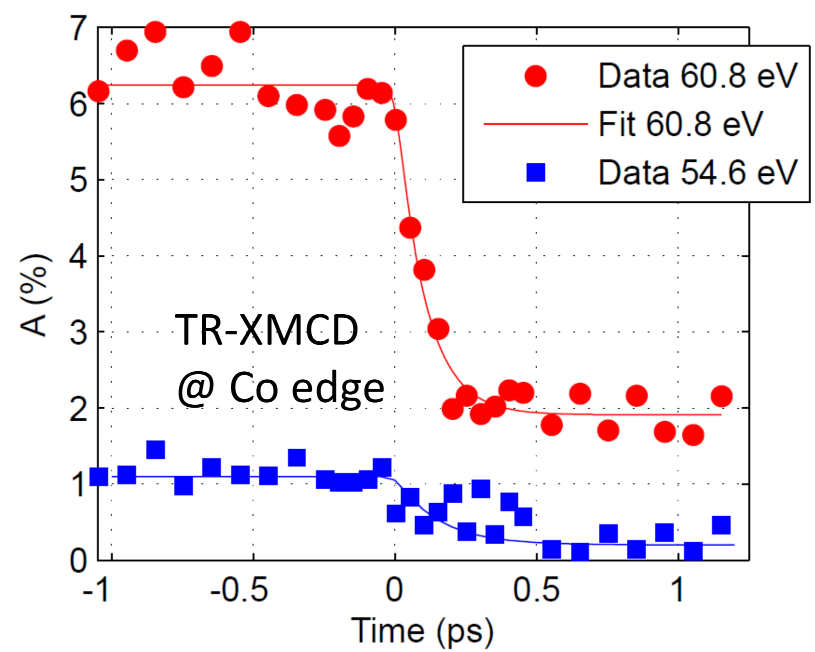
J. Weisshaupt et al., Phys. Rev. B 95, 081101(R) (2017)

Time-resolved XMCD using HHG



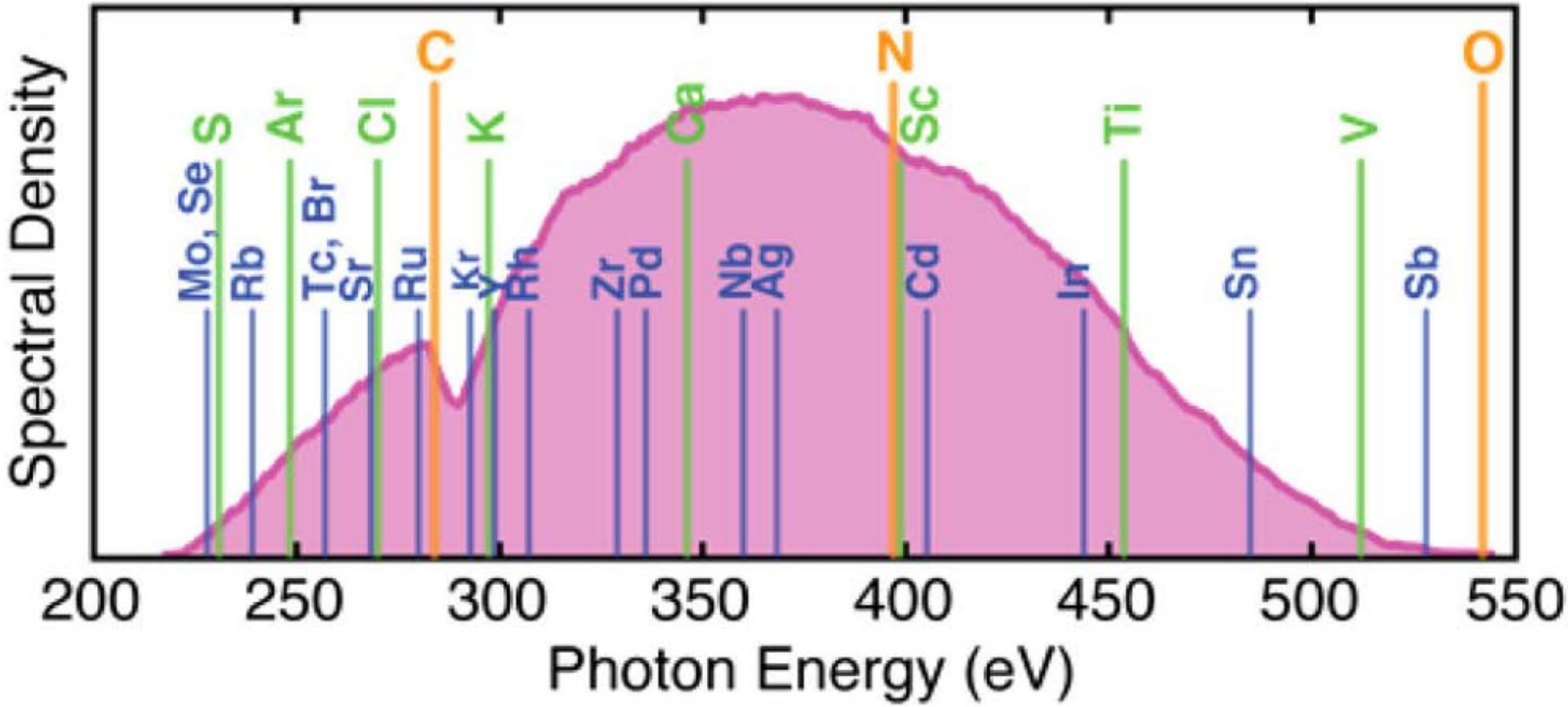






Willems et al., Phys. Rev. B 92, 220405(R) (2015)

ATAS in the water window: the case of TiS_2

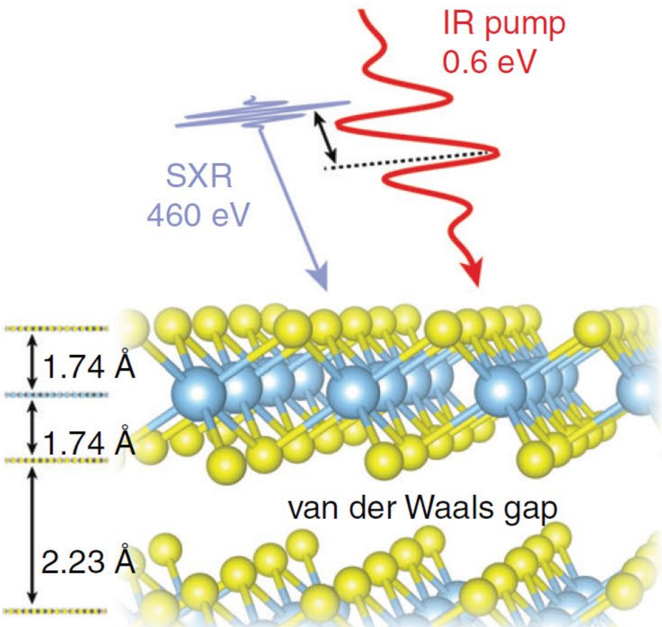


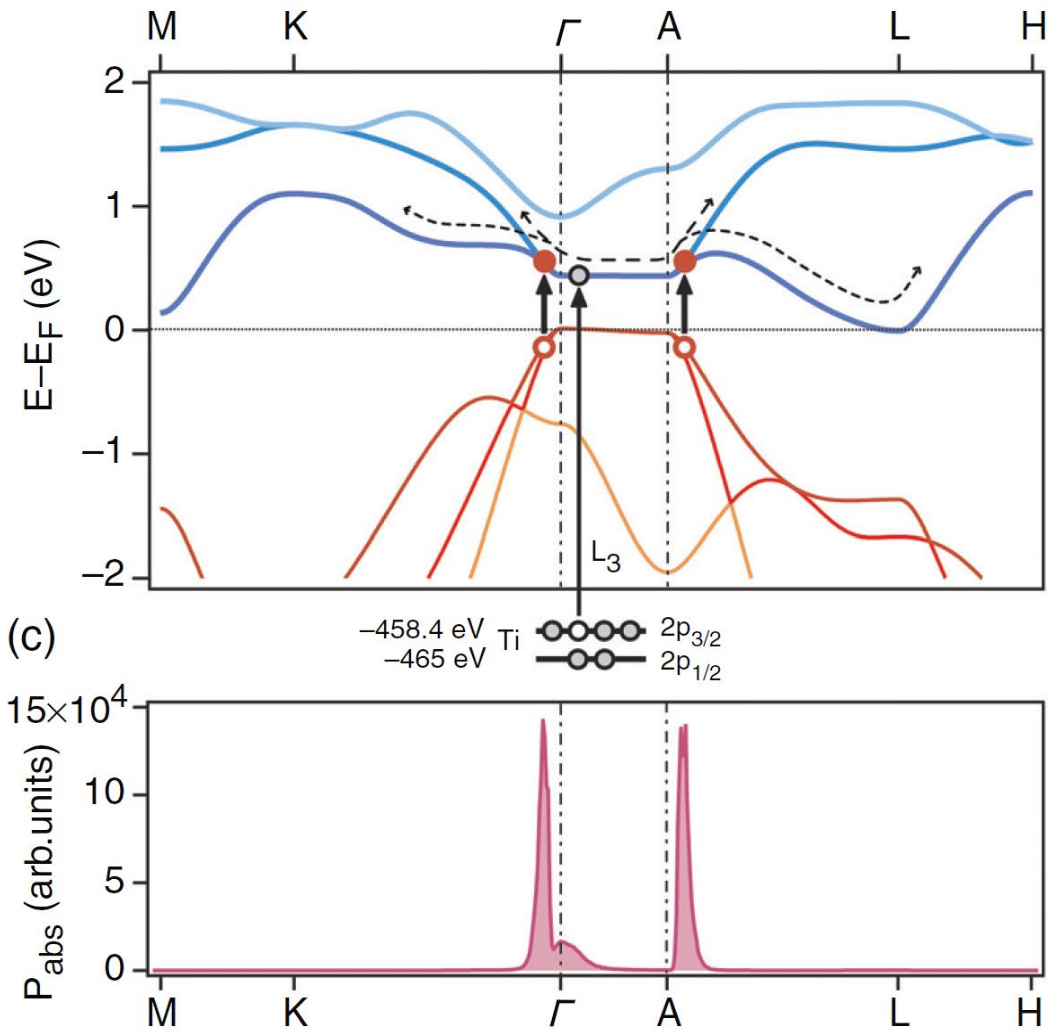
K-edges L-edges M-edges

When absorption edges from deeper-lying core levels are exploited, the theoretical description and – thus – interpretation of XAS experiments becomes easier → drive towards implementation of ATAS in the water window

B. Buades et al., Optica 5, 502 (2018)

ATAS in the water window: the case of TiS_2

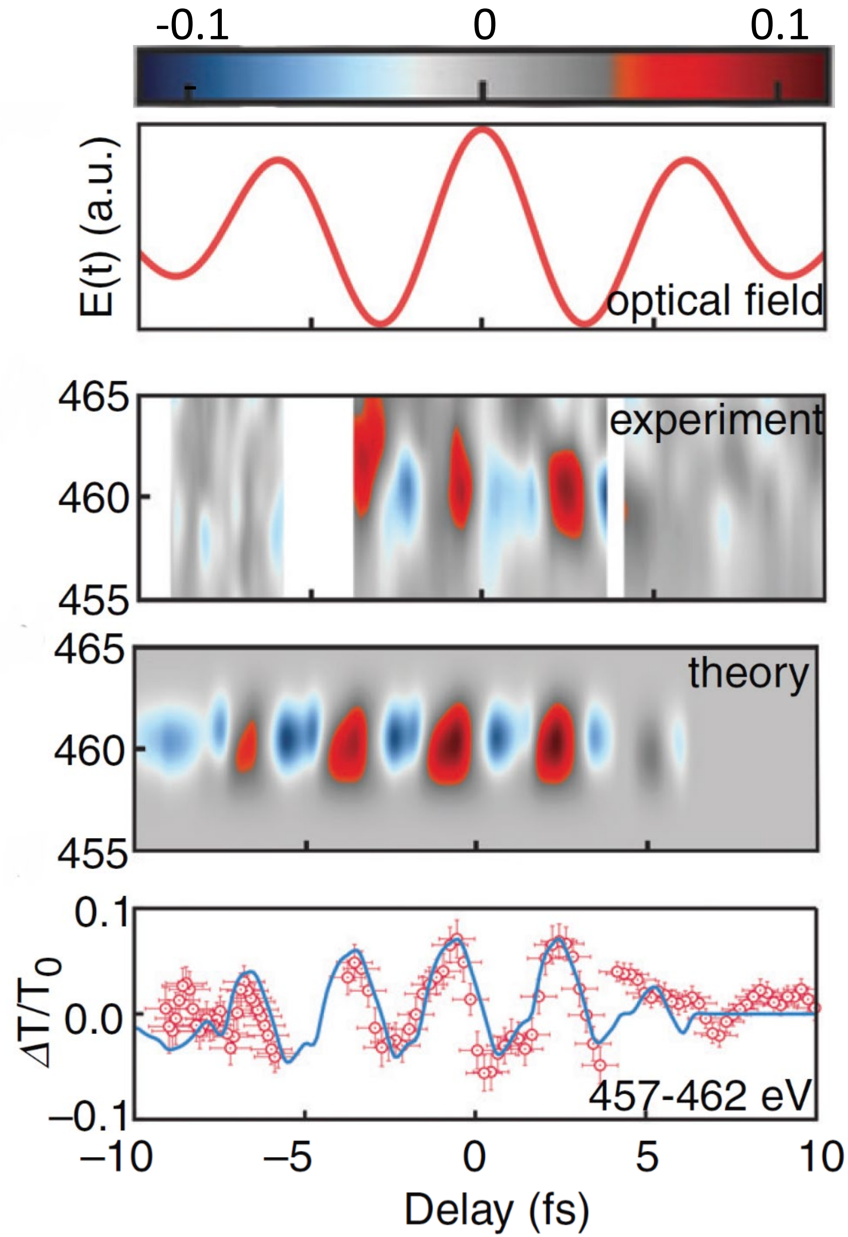




- Excitation from valence (3s/3p character) to conduction (3d character) band
- Probing of conduction band electrons using 2p→3d Ti L-edge (~460 eV)

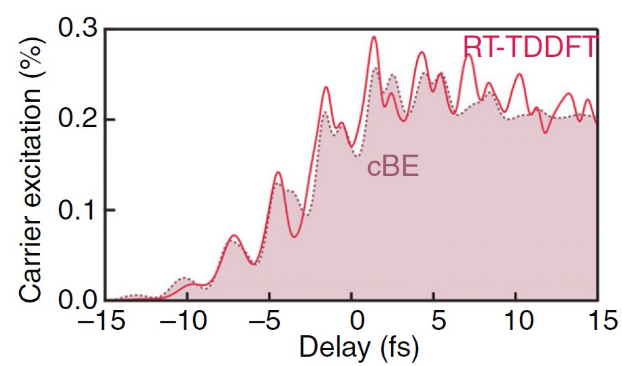
B. Buades et al., Appl Phys. Rev. 8, 011408 (2021)

ATAS in the water window: the case of TiS_2



B. Buades et al., Appl Phys. Rev. 8, 011408 (2021)

- Plot the relative change in the absorption (compared to the static absorption, without pump laser)
- The experiment shows that the L-edge absorption increases and decreases as a function of IR-XUV delay, with two oscillations occurring per driver laser period
- These observations are well reproduced by theoretical calculations (both TDDFT and core-state resolved Bloch Equation (cBE) model

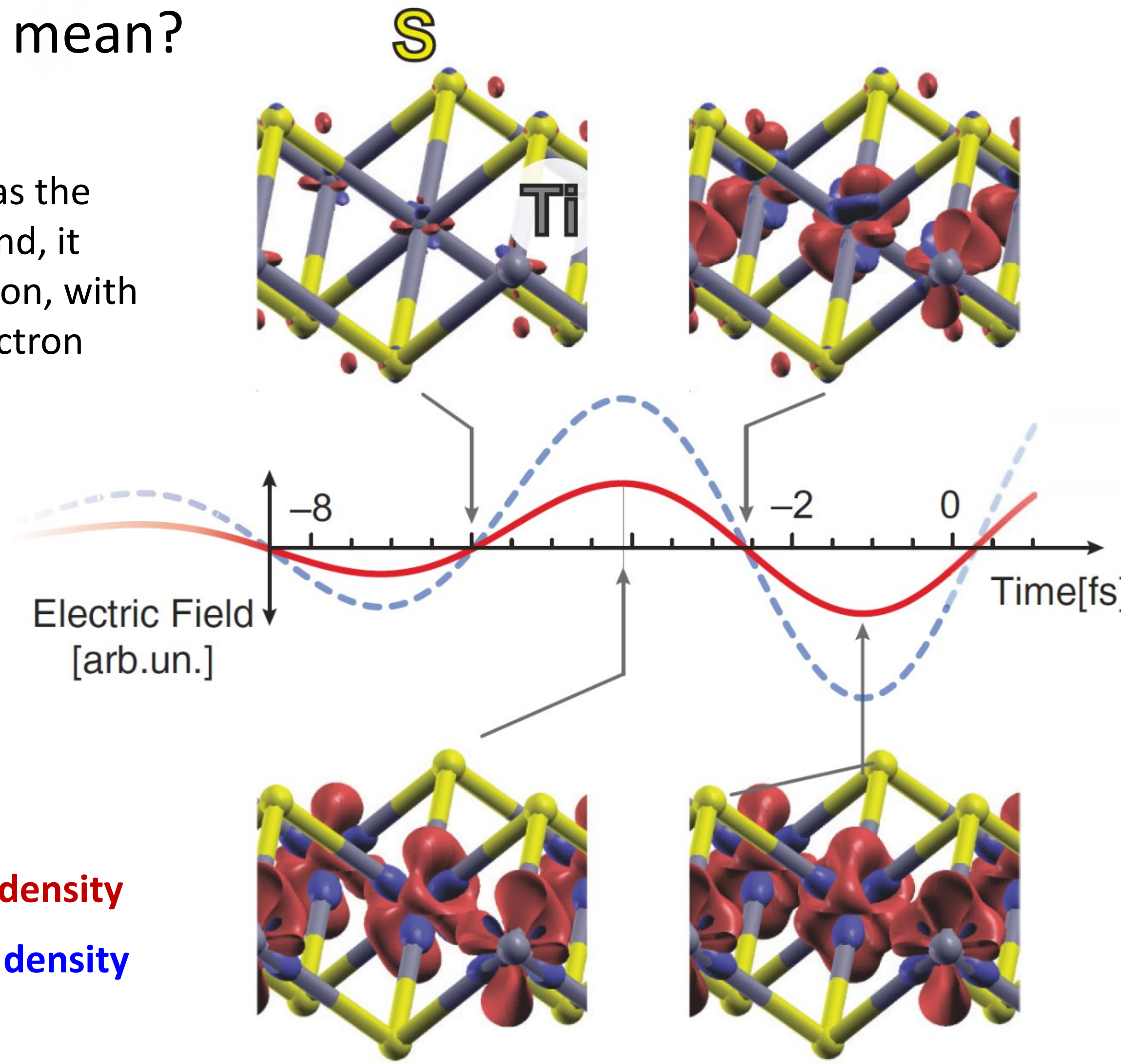


- Build-up of charge in the conduction band in TDDFT and cBE calculations

ATAS in the water window: the case of TiS_2

What do the oscillations mean?

- The authors conclude that as soon as the charge arrives in the conduction band, it starts to perform an intraband motion, with – particular – an increase of the electron density on the Ti atom



Increase of electron density

Reduction of the electron density

B. Buades et al., Appl Phys. Rev. 8, 011408 (2021)

Conclusion

The plan for this week

<table>
<tr><th></th><th>Monday</th><th>Tuesday</th><th>Wednesday</th><th>Thursday</th><th>Friday</th></tr>
<tr><td>8.00 – 9.00</td><td rowspan="2">Time-resolved spectroscopy (120)</td><td rowspan="2">Oscillator including carrier envelope phase (CEP) stability (120)</td><td rowspan="2">Generation and characterization of attosecond pulses (120)</td><td>Optical parametric chirped pulse amplification (OPCPA) (60/120)</td><td rowspan="2">Attosecond atomic physics (120/240)</td></tr>
<tr><td>9.00 – 10.00</td><td rowspan="2">Post-compression (120)</td></tr>
<tr><td>10.00 – 11.00</td><td>Mathematical description of laser pulses (60)</td><td>Pulse Characterization (60)</td><td rowspan="2">Chirped pulse amplification (CPA) (cont.) (60/120)</td><td rowspan="2">Attosecond molecular physics (90)</td></tr>
<tr><td>11.00 – 12.00</td><td rowspan="2">Non-perturbative physics (120)</td><td rowspan="2">High-harmonic generation (120)</td><td rowspan="2">Attosecond atomic physics (120/240)</td></tr>
<tr><td>12.00 – 13.00</td><td>Optical parametric chirped pulse amplification (OPCPA) (60/120)</td><td>Attosecond condensed phase physics (90)</td></tr>
<tr><td>13.00 – 14.00</td><td>Open discussion</td><td>Open discussion</td><td>Open discussion</td><td>Open discussion</td><td>Open discussion</td></tr>
</table>

Marc Vrakking

Federico Furch

Key ingredients

- We will discuss how materials can be studied by means of pump-probe experiments
- We will discuss the interaction of materials with intense laser light, and see how this can give rise to the formation of extreme ultraviolet (XUV) attosecond laser pulses
- We will discuss how pump-probe experiments can be configured using attosecond laser pulses
- We will learn about key experimental techniques, i.e. how intense lasers are built, sample preparation, detection techniques, etc.
- To understand the lectures, we will understand a basic knowledge of optics and quantum mechanics – if something is not clear, please ask!

Selected take home messages that you will understand on Friday

- *The proliferation of attosecond science became possible because of major developments in ultrafast laser technology, and attosecond science drives numerous new developments*
- *High-harmonic generation and the formation of attosecond pulses are best understood in a field picture of the laser-matter interaction*
- *Pump-probe experiments are interference experiments, where time-dependent signals arise through interference between multiple quantum paths that connect the initial state of a system to the final state detected in the experiment*
- *The techniques that were introduced in 2001 for the characterization of attosecond pulse trains and isolated attosecond pulses continue to be workhorses in experiments where attosecond pulses are used to probe electron dynamics*
- *The attosecond pulse structure of High-harmonic radiation is not its only important feature, HHG moreover permits core-level specific time-resolved spectroscopy with enormous impact in atomic, molecular and condensed phase physics*
- *The future of attosecond science looks extremely bright!*


Thank you for your attention!
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