PERSEPHONe & SMART-X Winter School

January 8 - January 13, 2023

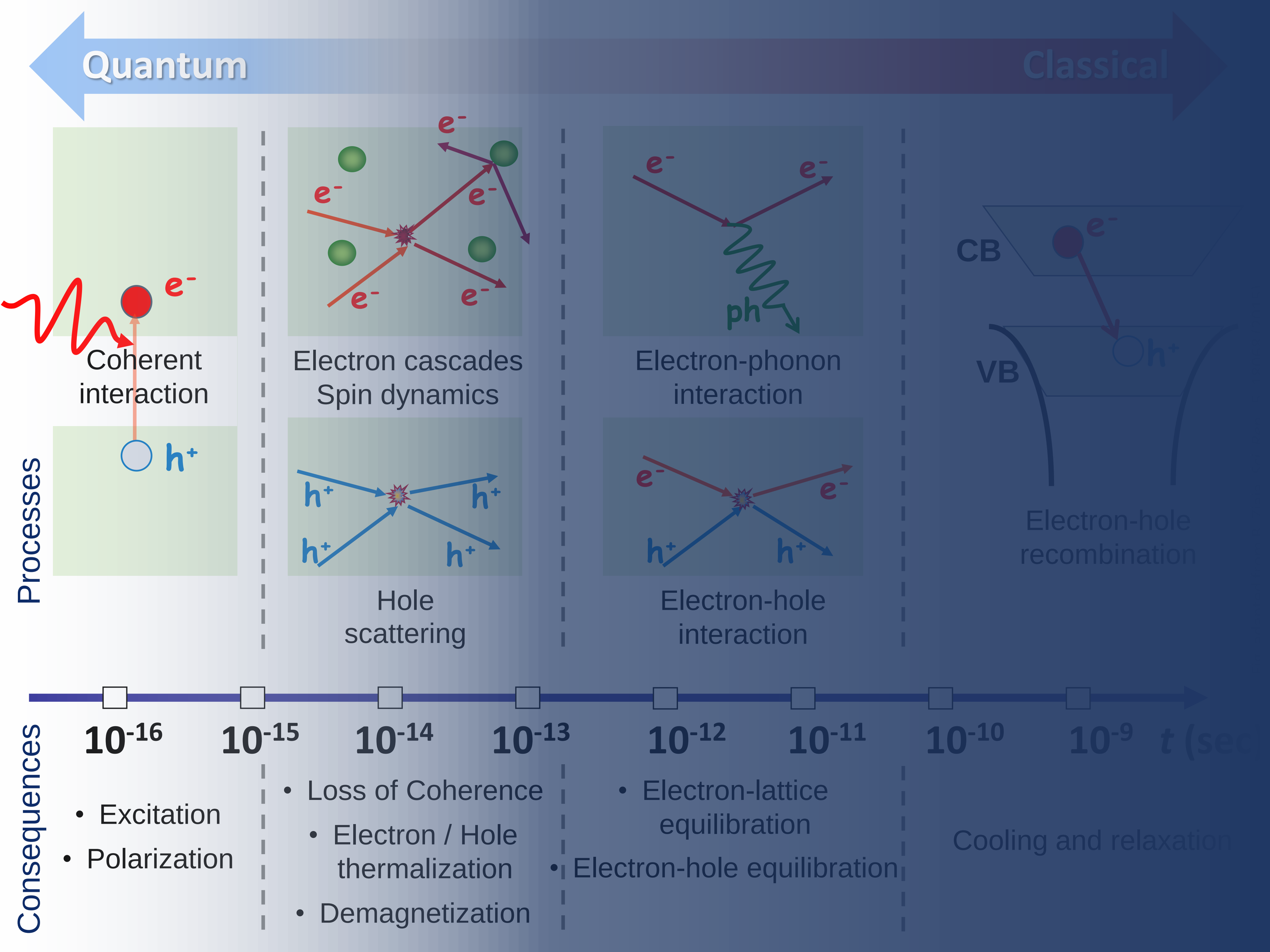
Bormio, Italy

Dyn properties

ht

Martin Schultze
TU Graz - Institute of Experimental Physics
www.tugraz.at/institute/iep

Tracking Charge and Spin in time

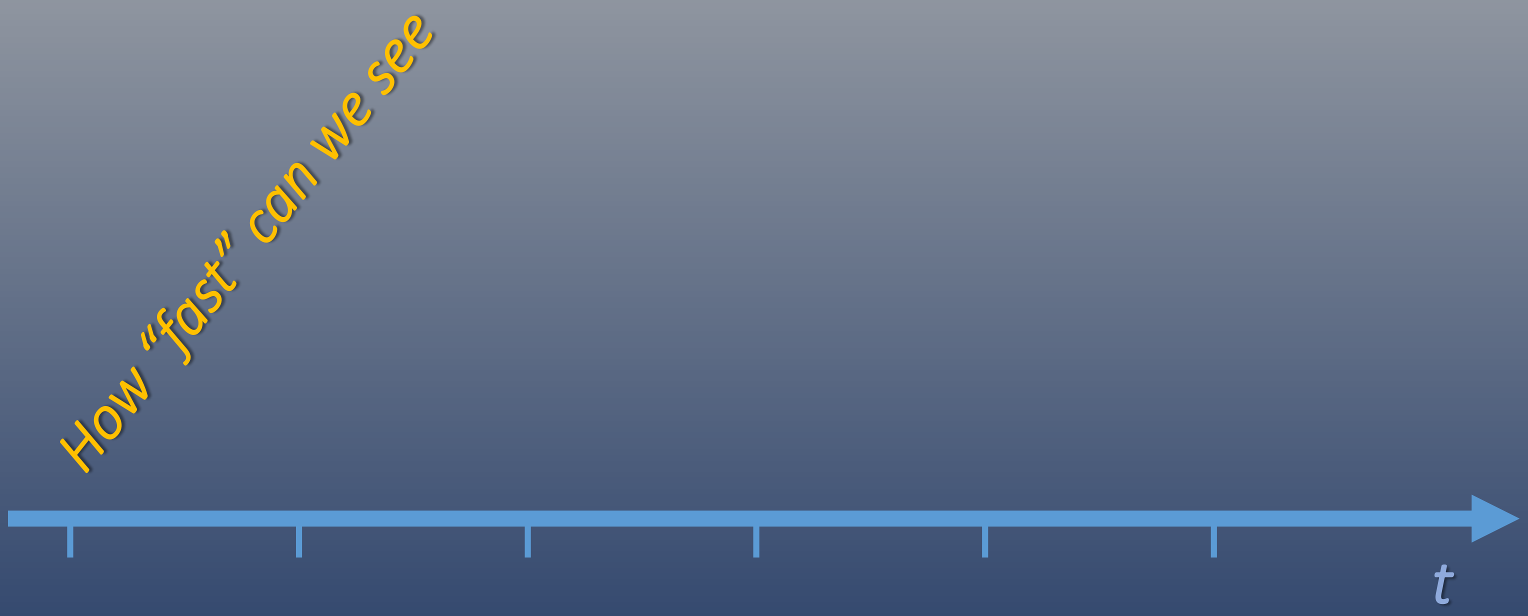

Quantum
Classical
Processes
e⁻
h⁺
Coherent interaction
Electron cascades
Spin dynamics
Hole scattering
Electron-phonon interaction
ph
Electron-hole interaction
CB
VB
Electron-hole recombination
10^{-16}
10^{-15}
10^{-14}
10^{-13}
10^{-12}
10^{-11}
10^{-10}
10^{-9}
t (sec)
Consequences
• Excitation
• Polarization
• Loss of Coherence
• Electron / Hole thermalization
• Demagnetization
• Electron-lattice equilibration
• Electron-hole equilibration
Cooling and relaxation

Outline

- How "fast" can we see
- Excitation across a band-gap takes how long?
- What if my material doesn't transmit XUV?
- Where is all the energy? And when?
- Can my laser make electronics faster?
- What? Really? Can it also make something more magnetic?

t

Outline



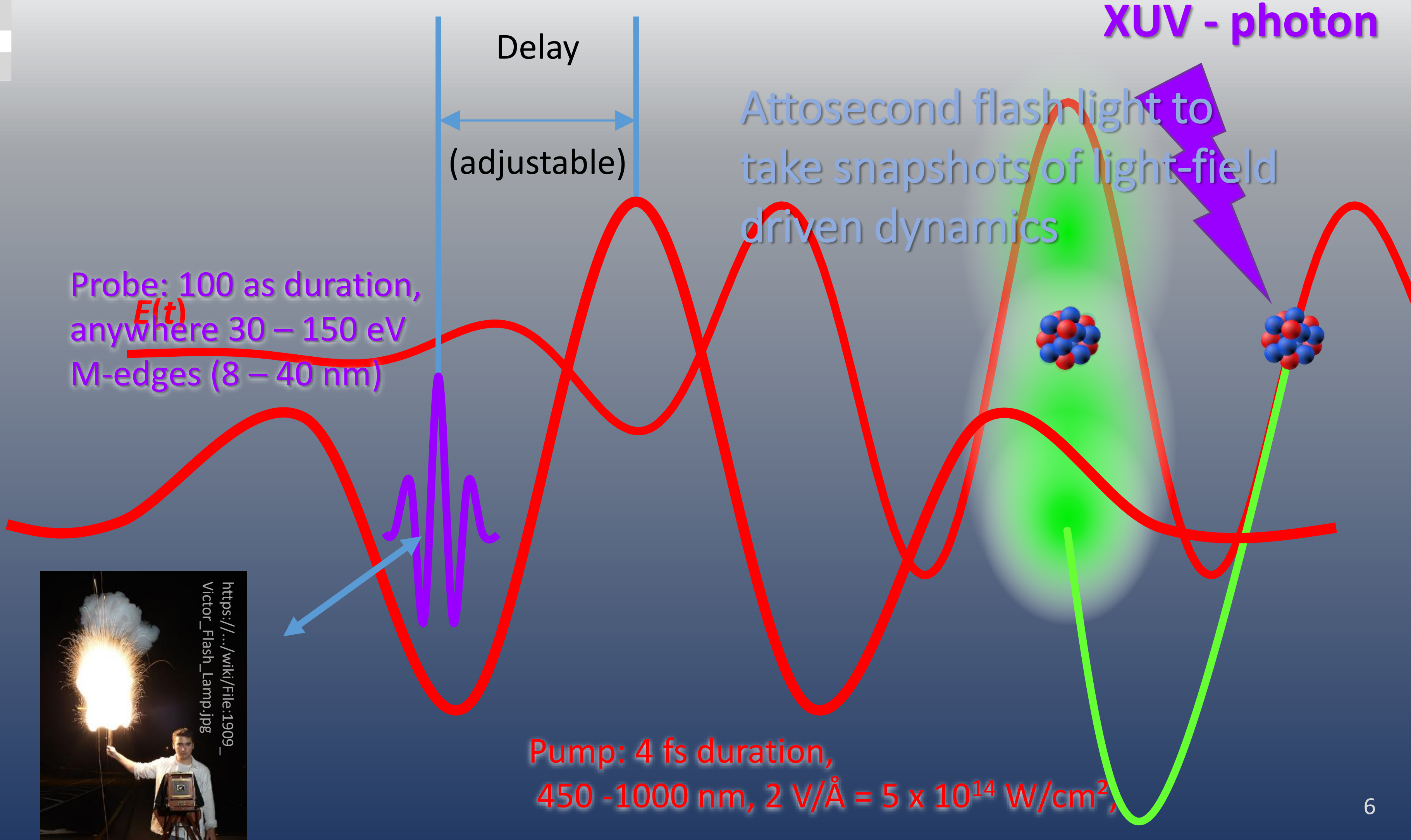
Stroboscopic effect

Flash lamp
~ 1/100 000 s

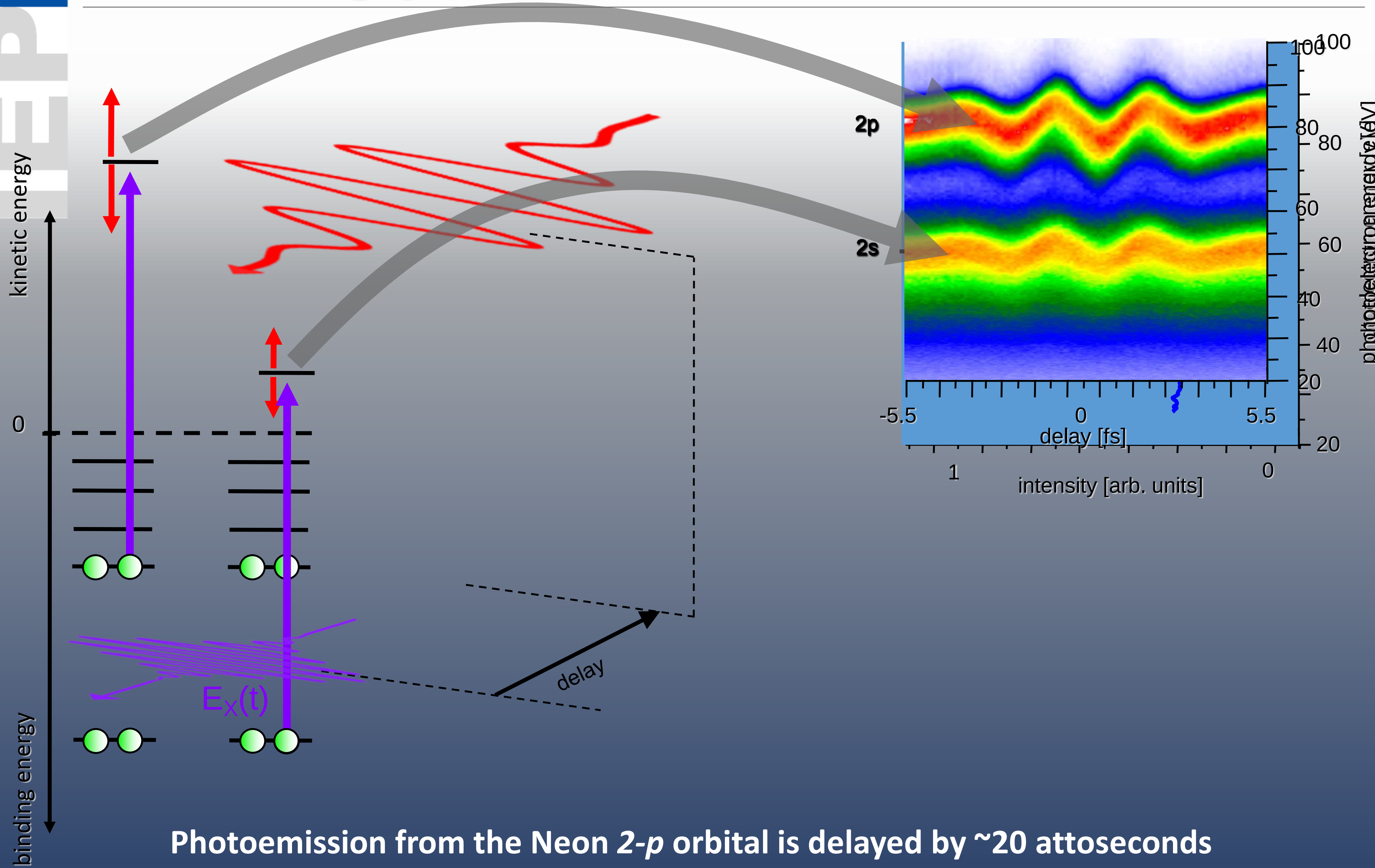
Freezing time via stroboscopic illumination

Attosecond stopwatch

Nonlinear optics via photo ionization:
The Three-Step-Model of High-Harmonic-Generation



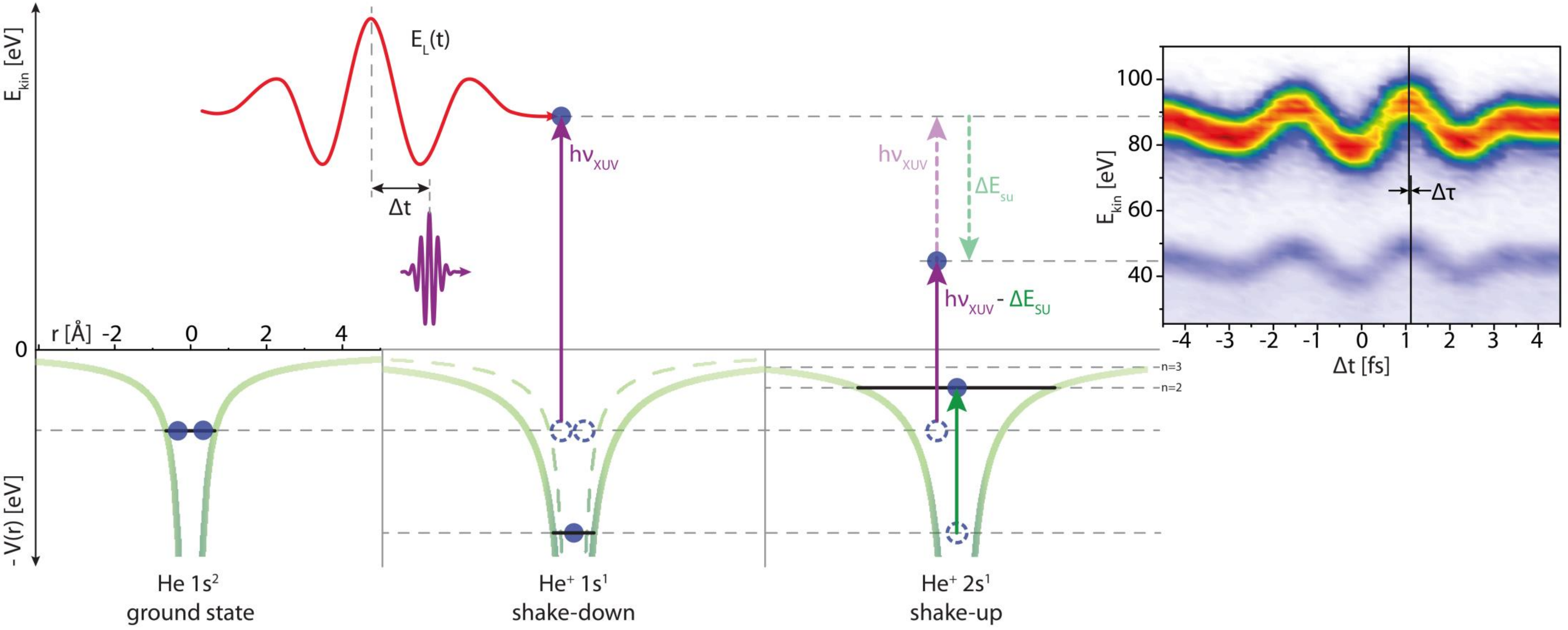
Clocking photoemission



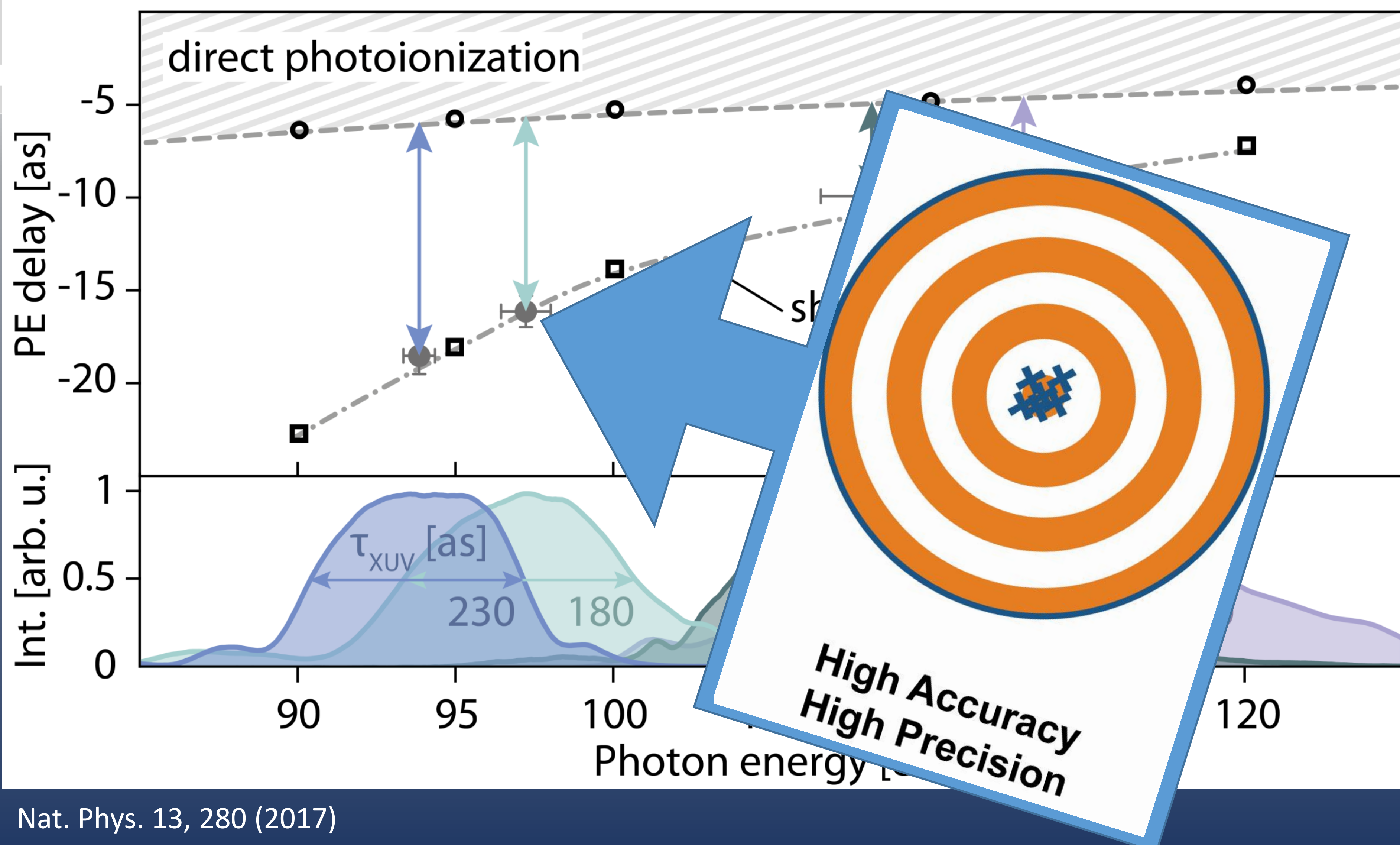
Photoemission from the Neon *2-p* orbital is delayed by ~20 attoseconds

Science 328 (5986) 2010

Clocking electronic correlations

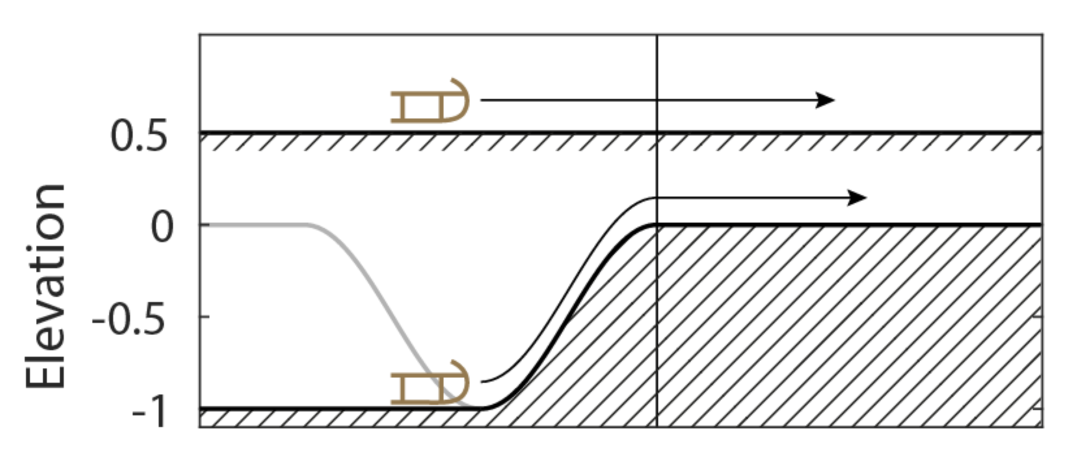


Clocking electronic correlations



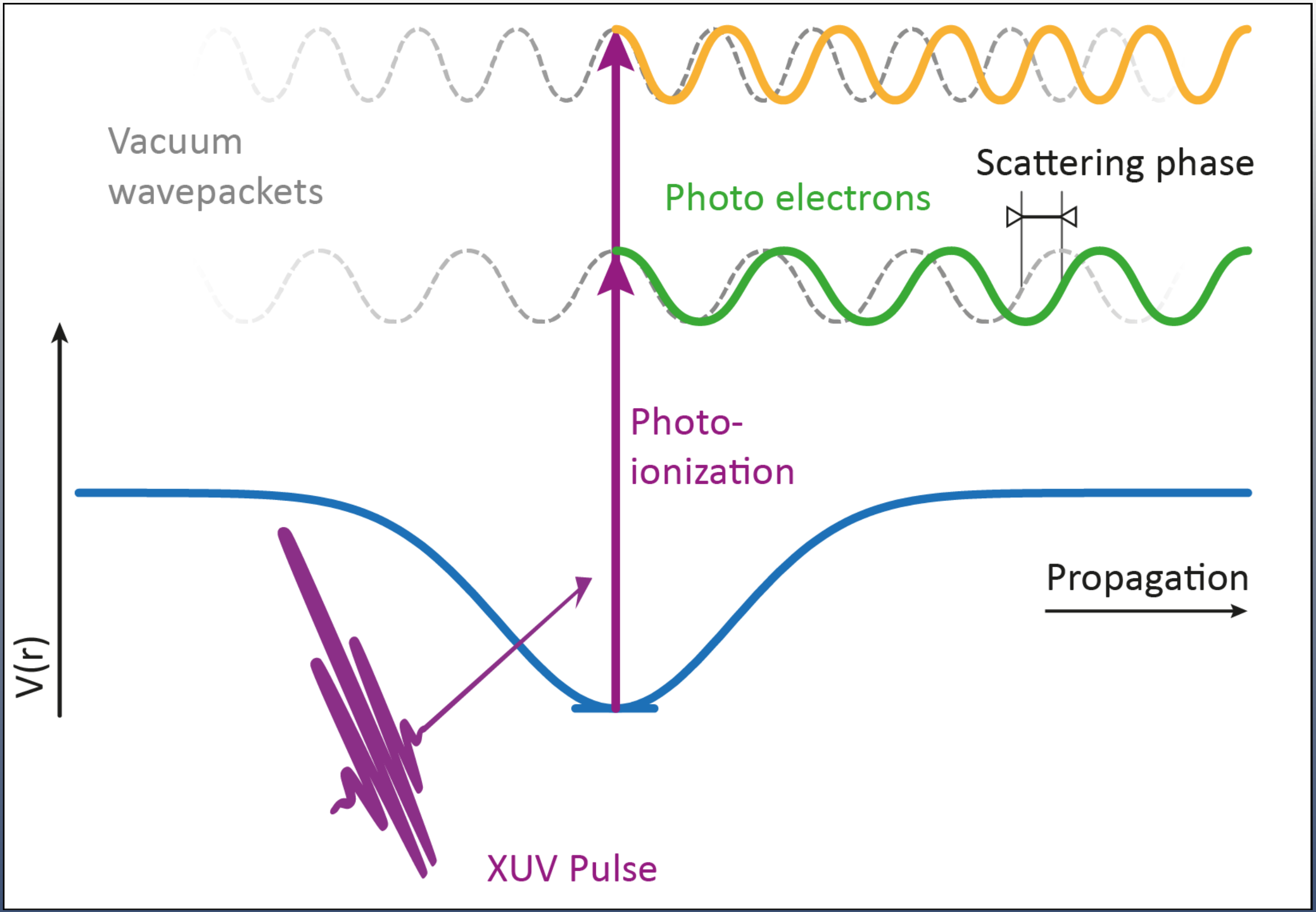
Clocking electronic correlations

…the phase of a wavepacket



Timing of Photoemission?

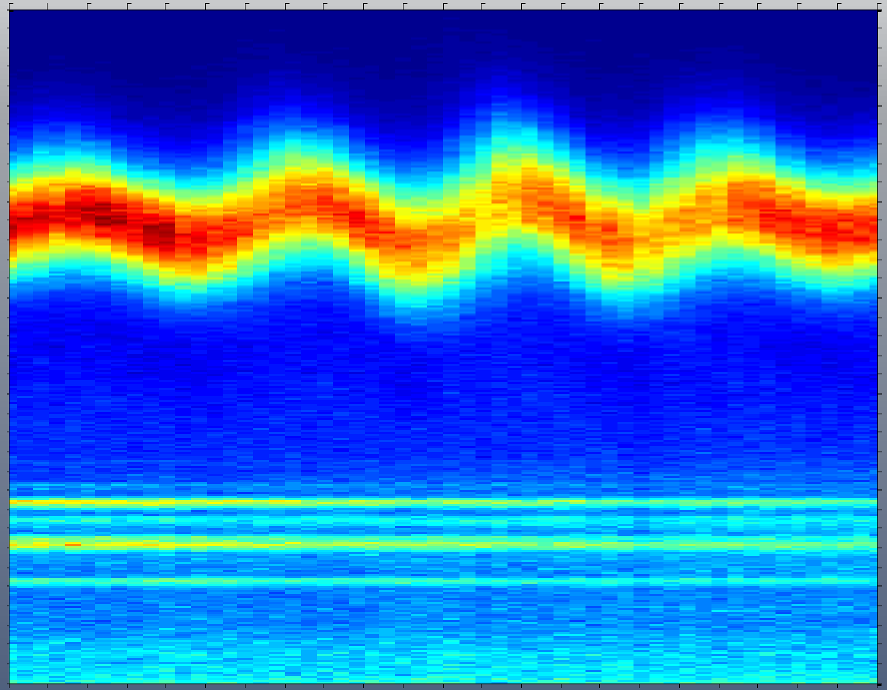
…the phase of a wavepacket



Accumulated scattering phase ➡ attosecond time shift in photoemission

Core-holes & time resolution

XUV pulse length ~ Wave-packet duration < $T_0/2$
→ streaking



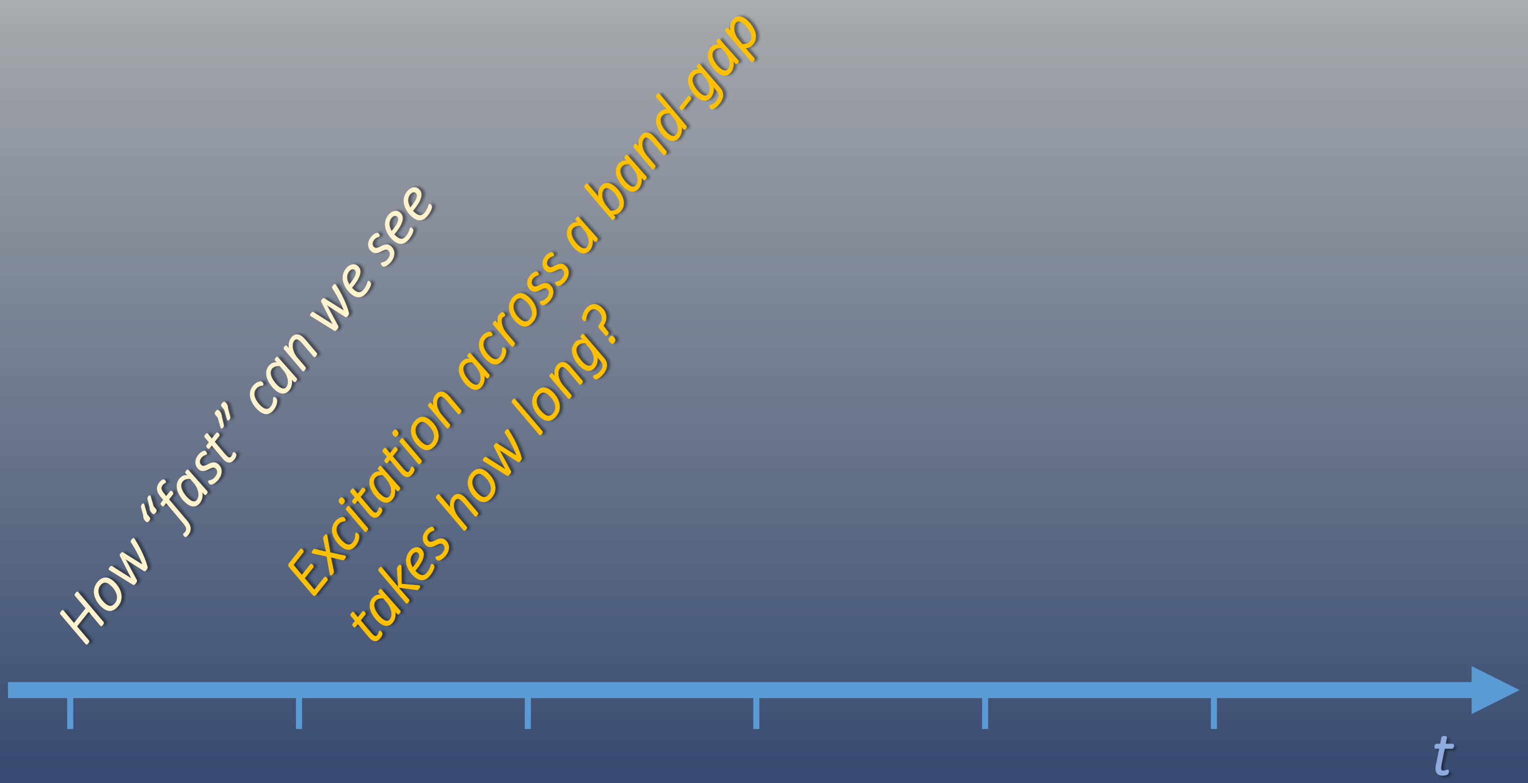
Xe 4d

Xe Auger decay

Wave-packet duration dominated by lifetime
(~ 10 fs Xenon Auger) >> $T_0/2$
→ sideband formation

Sometimes, core transitions live their own life

Outline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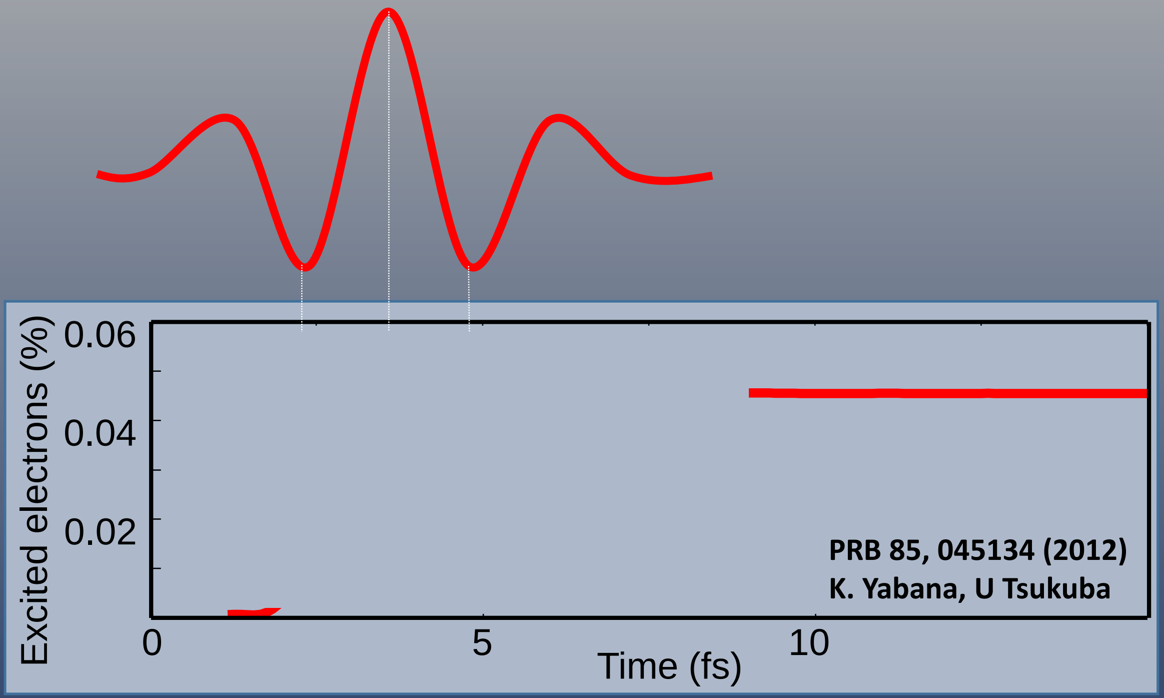
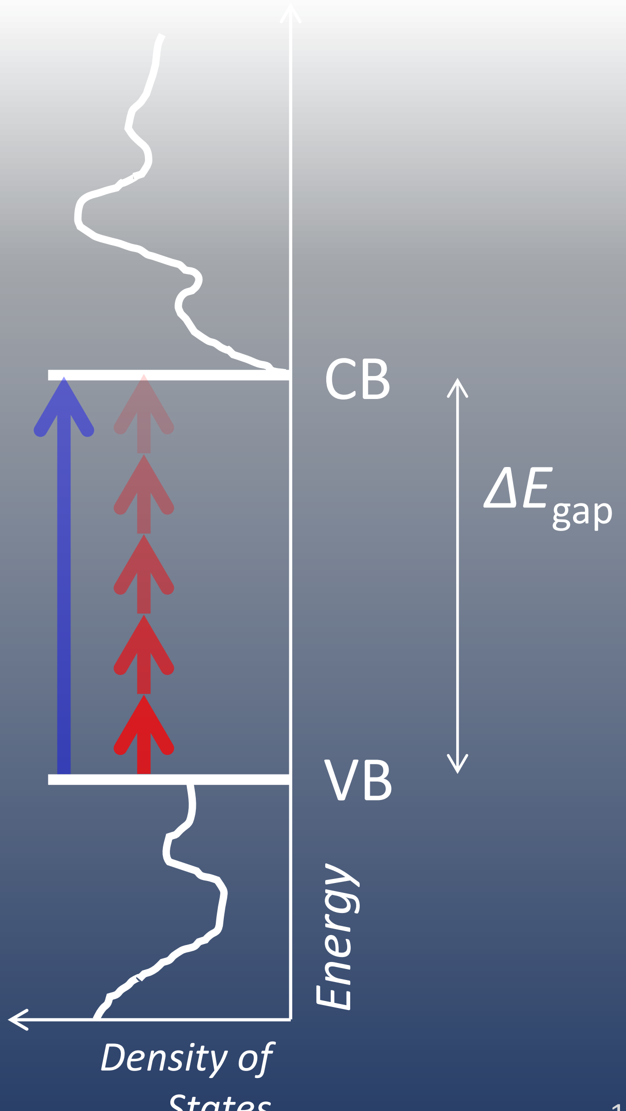
Excitation across a band-gap takes how long?
t

Charge dynamics in solids

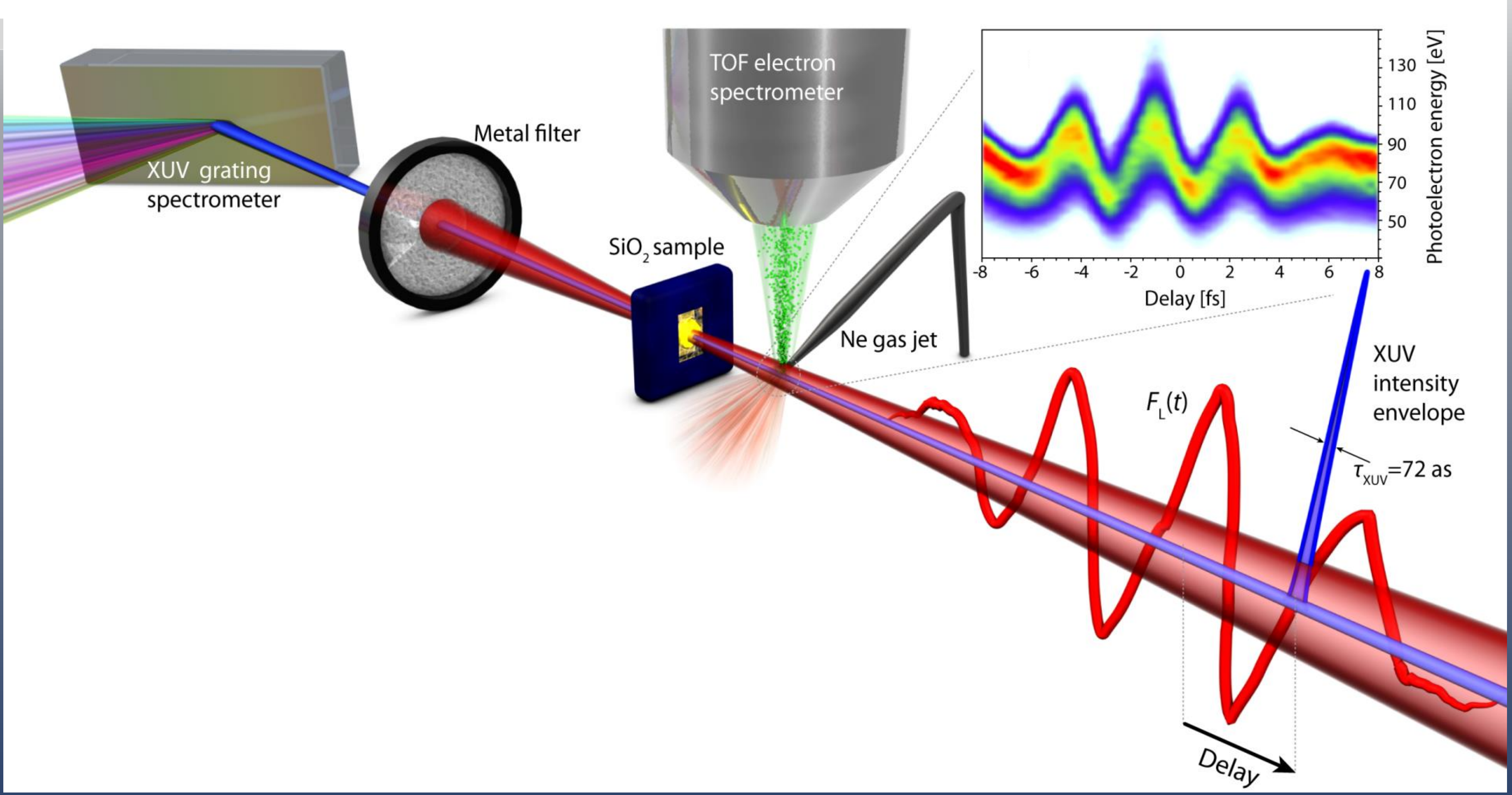
A material exposed to light can absorb energy by creating electron-hole pairs

Dispersion relation & Density of states

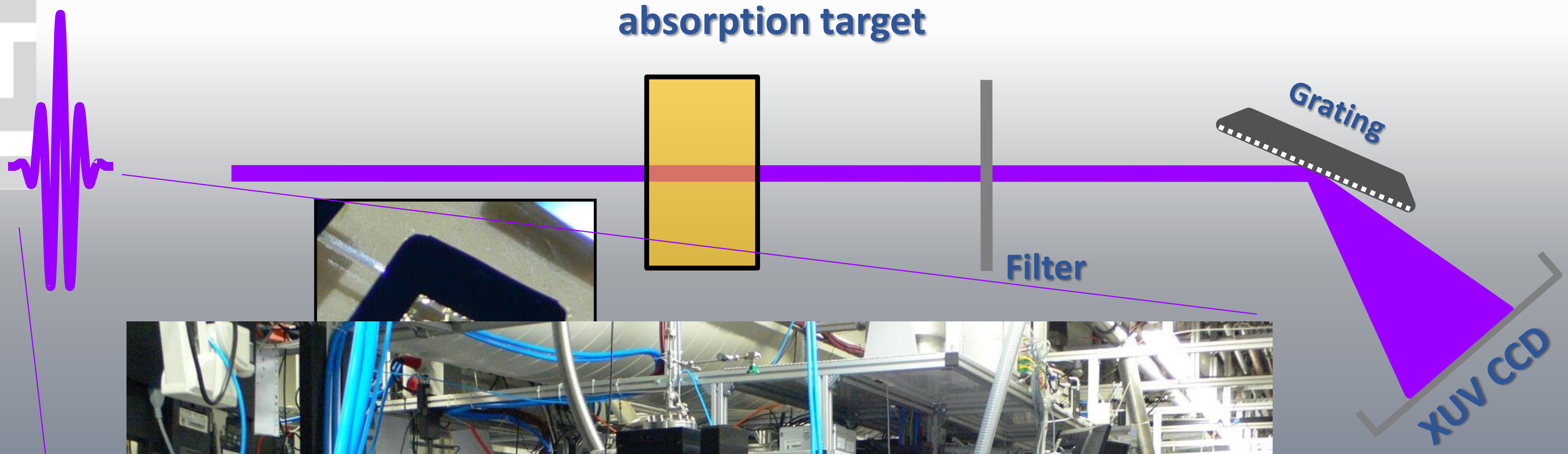


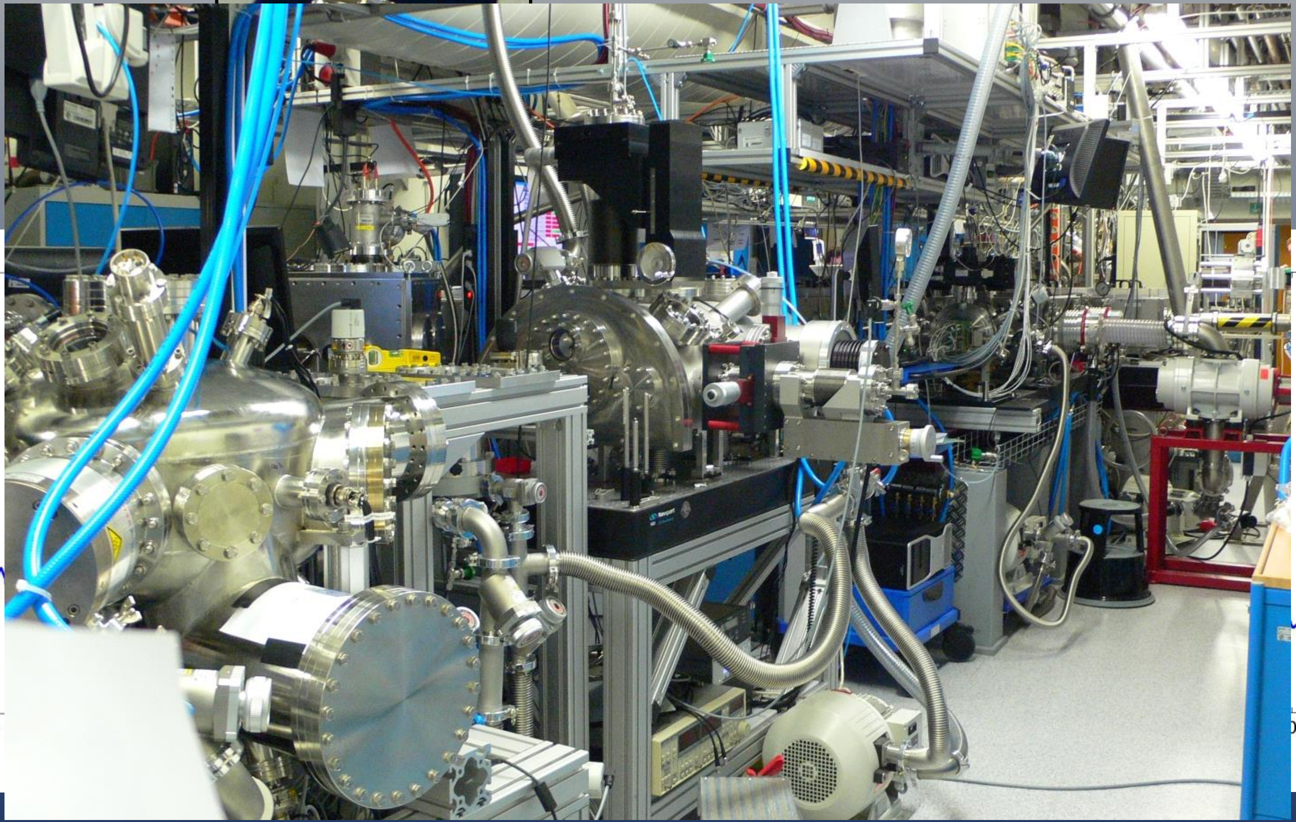


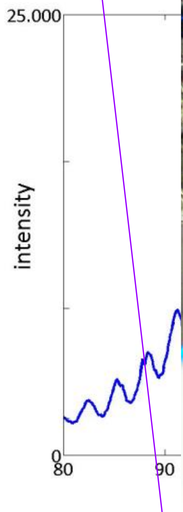
XUV - Transient absorption

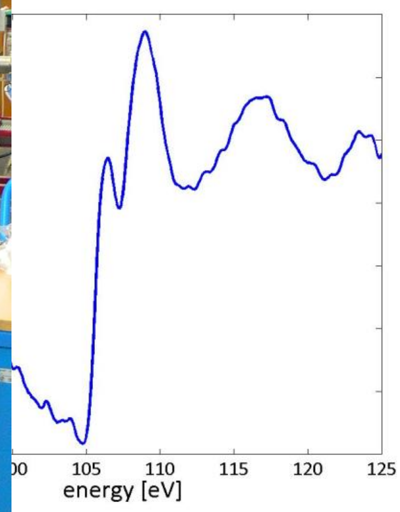


XUV - Transient absorption

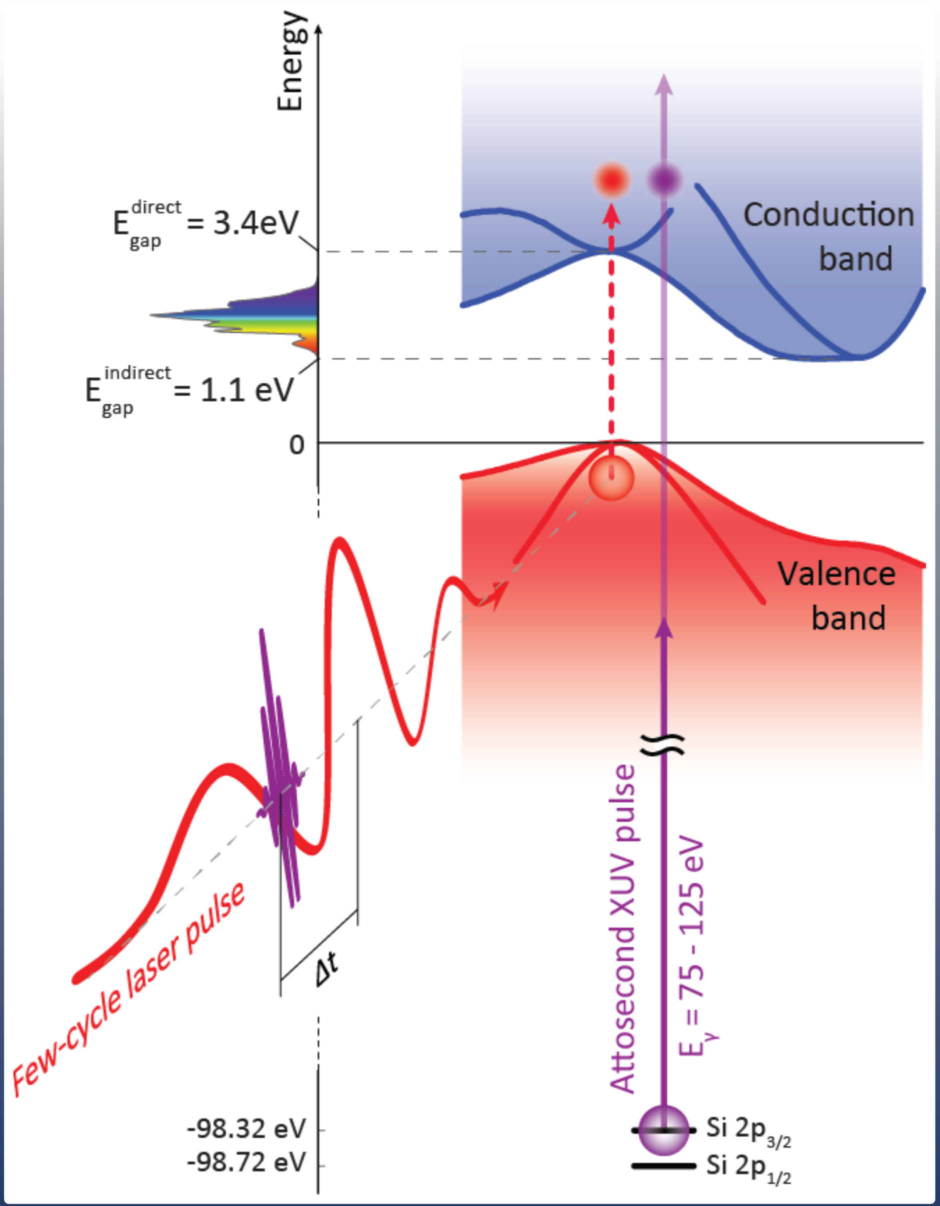




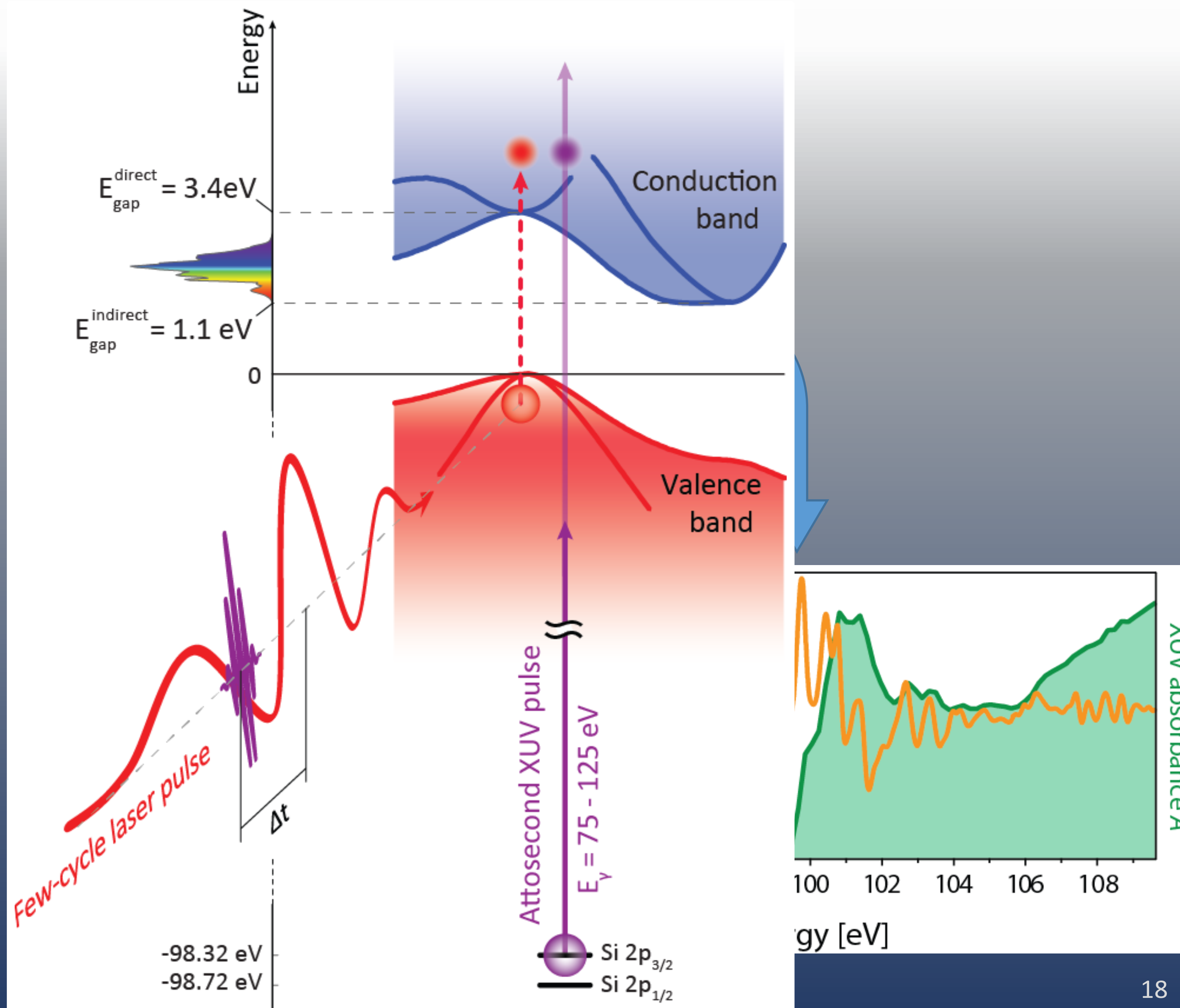




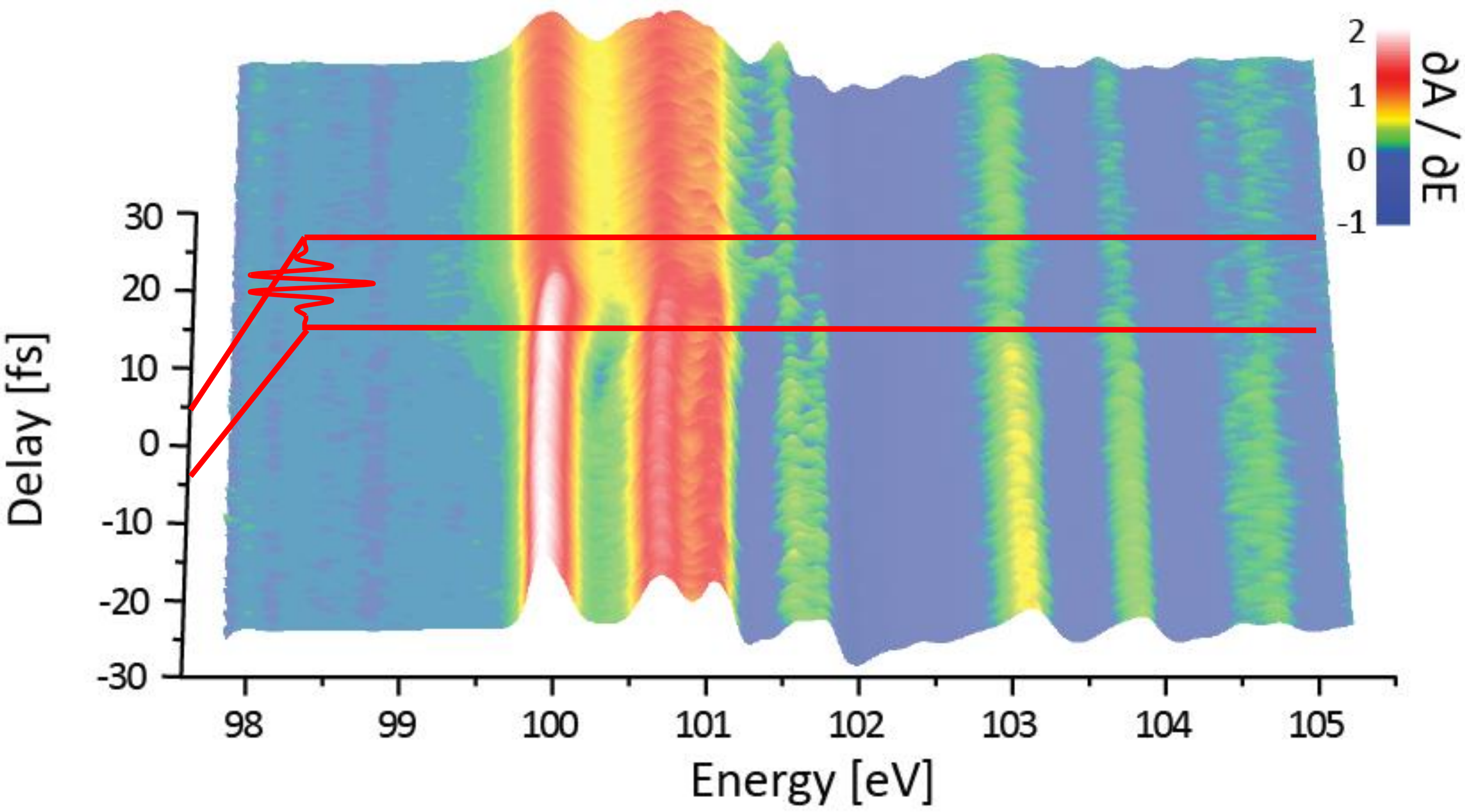
Band-gap dynamics in Silicon



Band-gap dynamics in Silicon

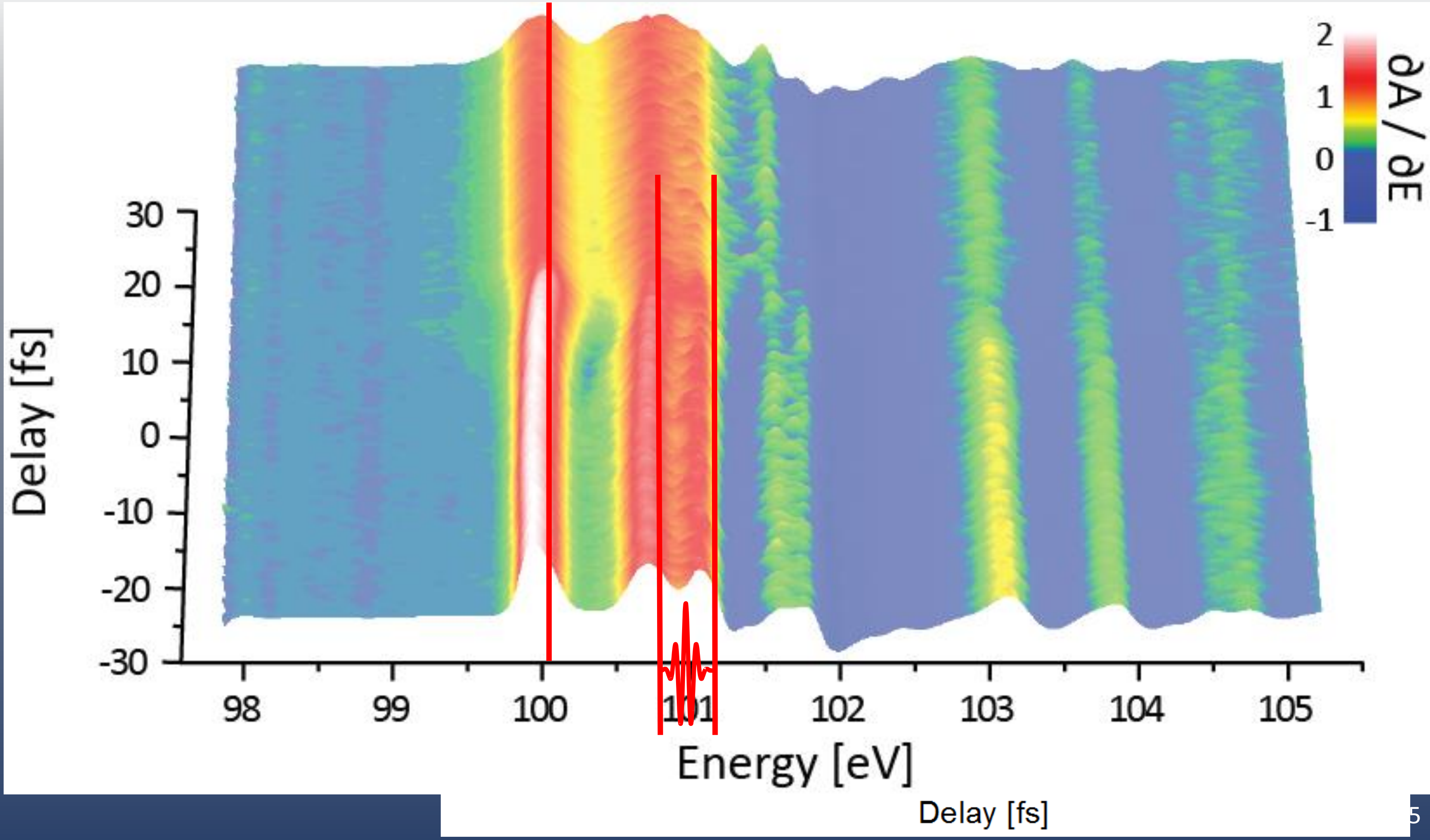


Band-gap dynamics in Silicon

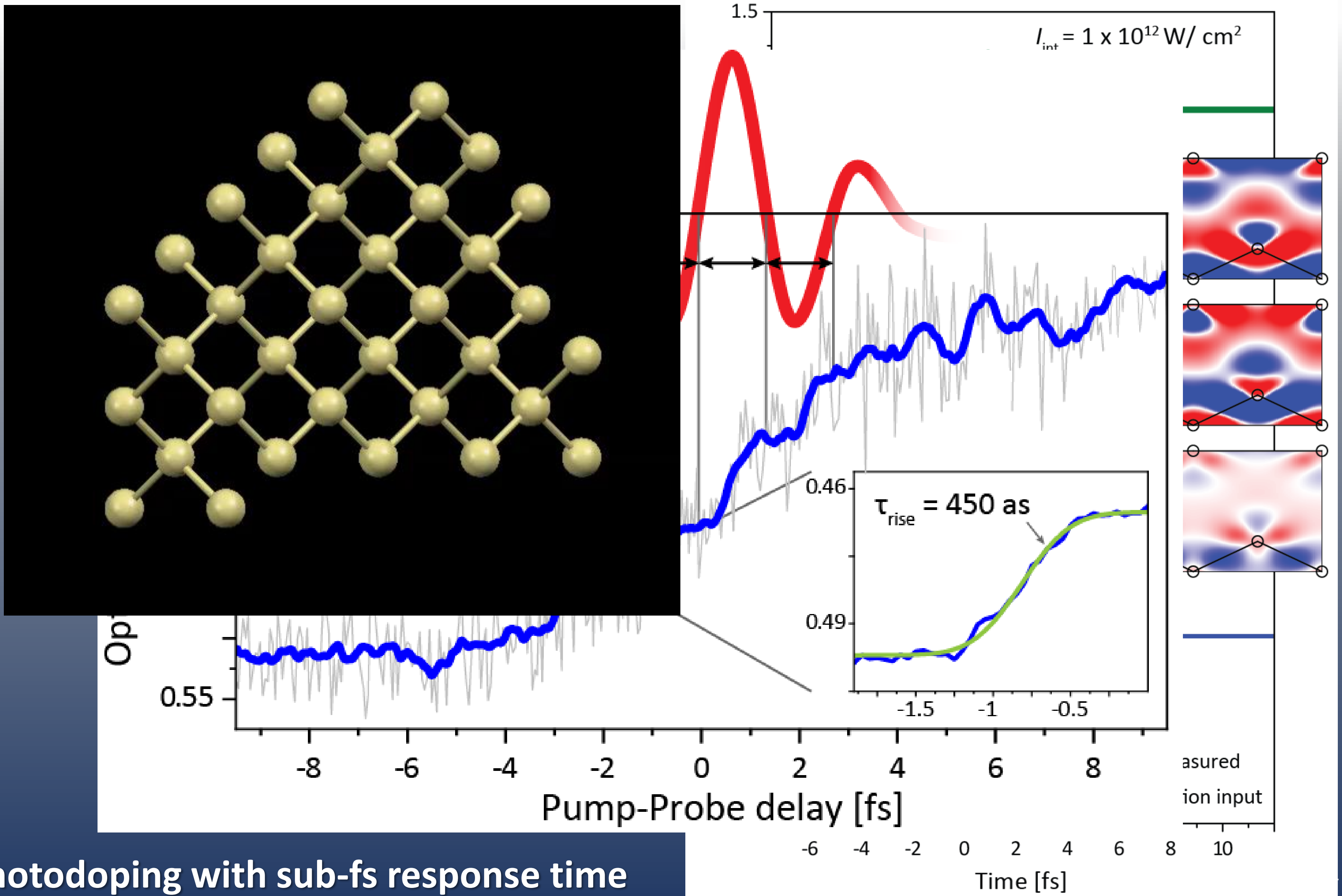


Science 346, 6215

Band-gap dynamics in Silicon

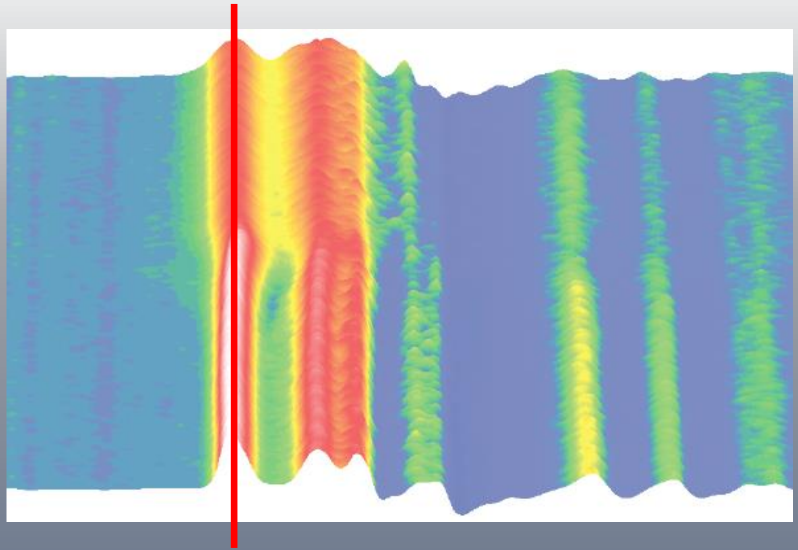


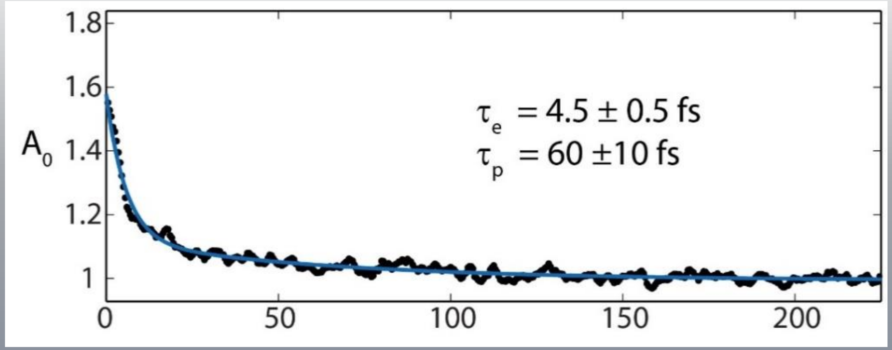
Interband tunneling

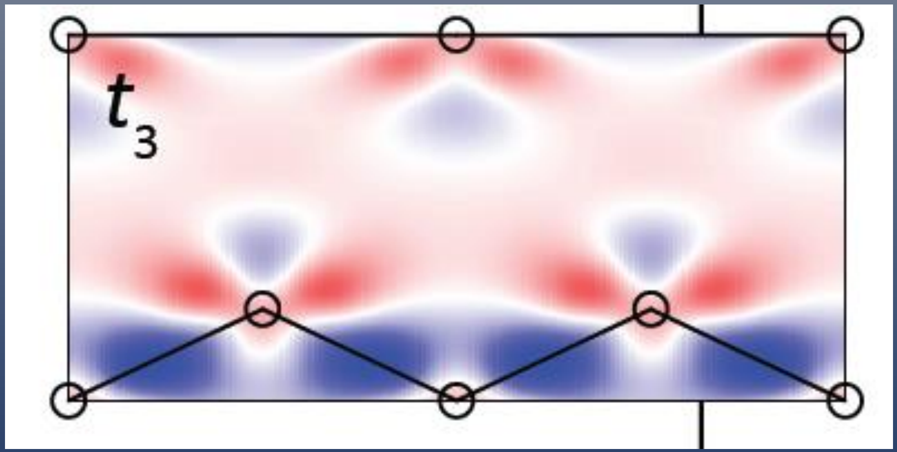


Photodoping with sub-fs response time

Ultrafast band-gap closing



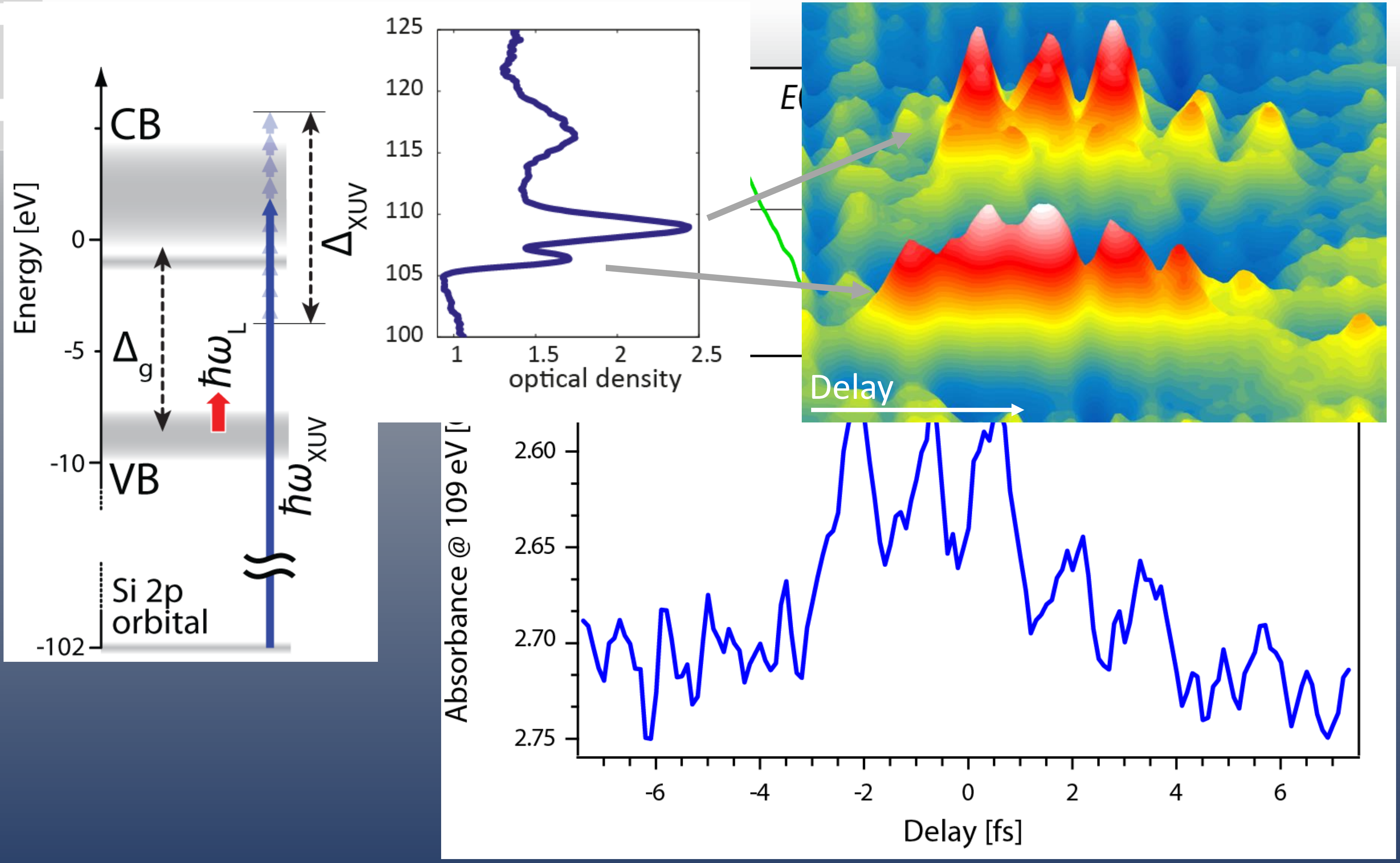




photodoping of silicon induces instantaneous (electronic) band gap narowing

the lattice follows with a time constant of the fastest optical phonon (64 fs)

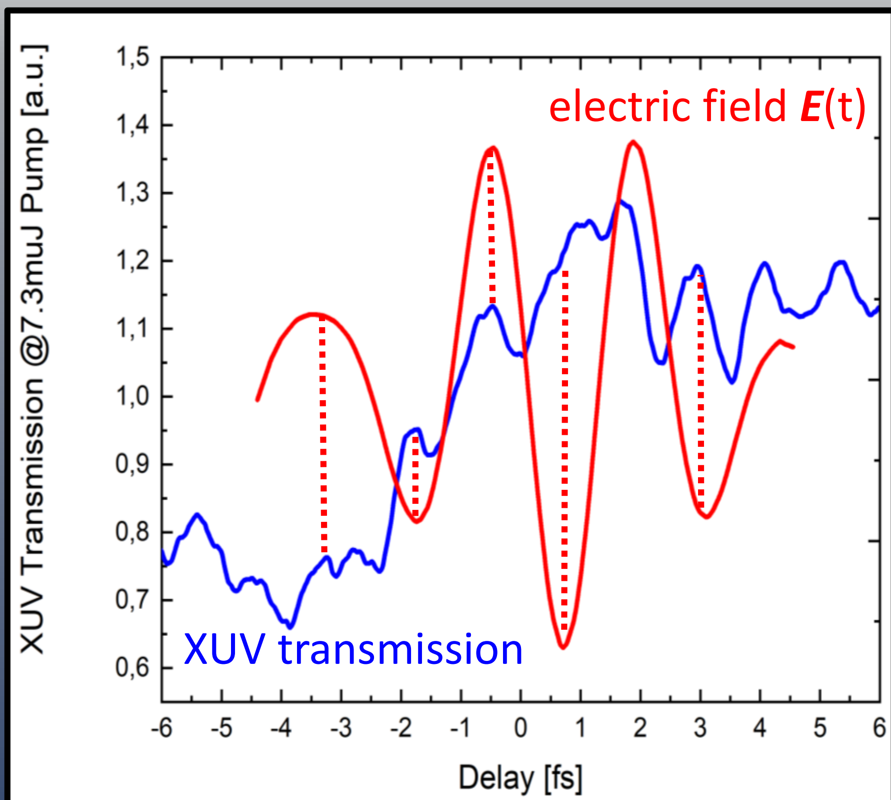
Strong field response of Quartz

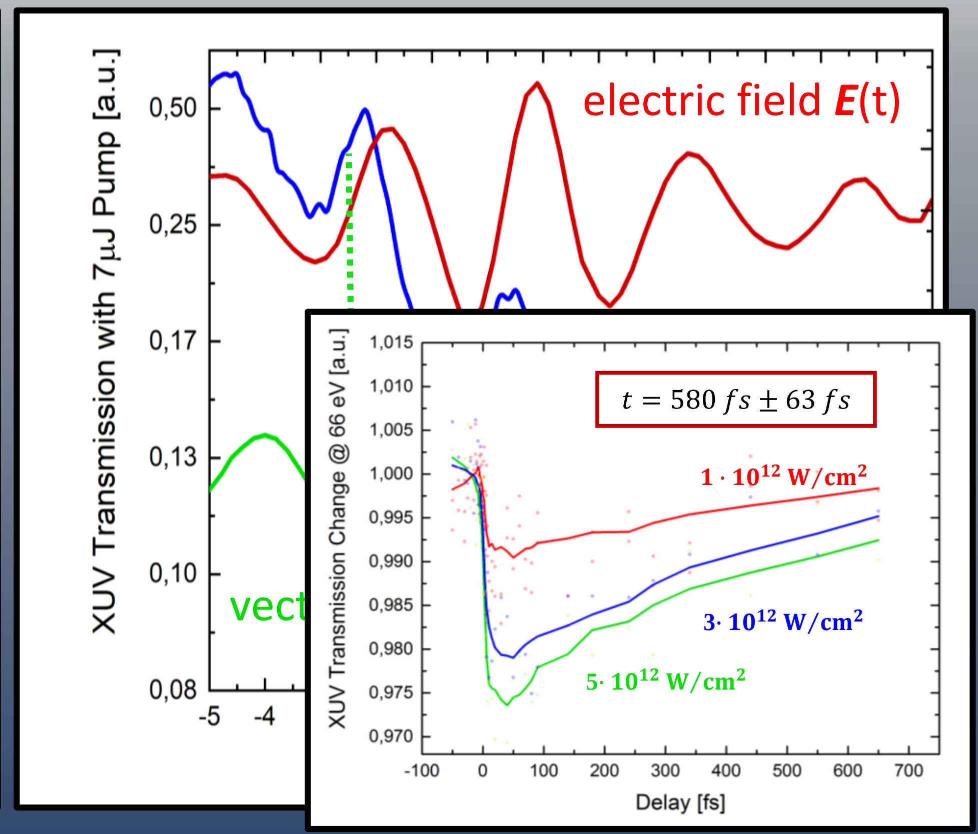


Semiconductor vs. metal

Silicon
semiconductor bandstructure

Nickel
metallic bandstructure





XUV Absorption can be sensitive to occupation dynamics or currents -> system dependent

Outline

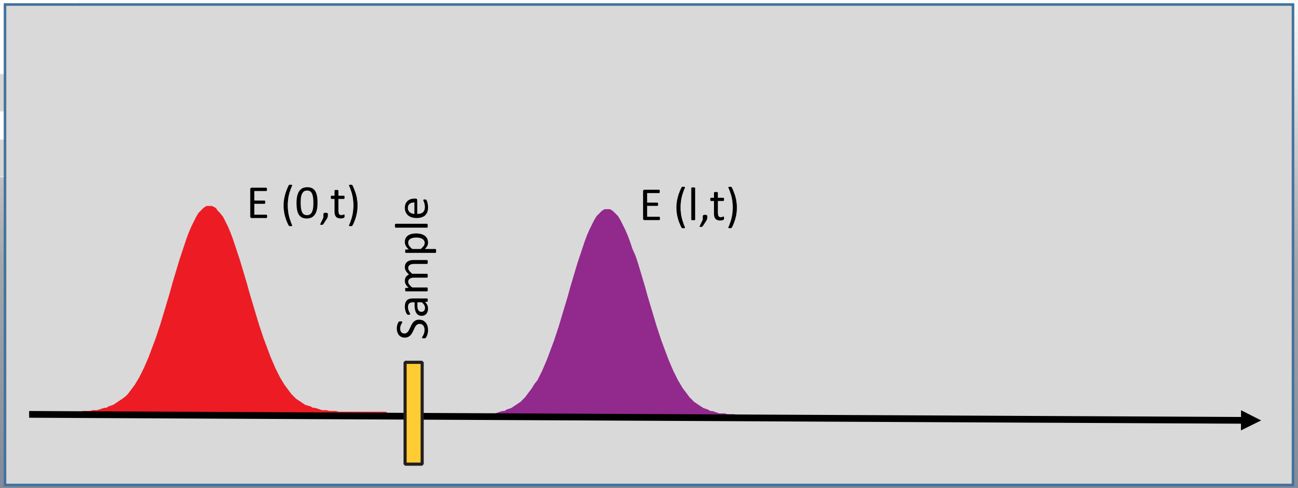
How “fast” can we see

Excitation across a band-gap takes how long?

What if my material doesn’t transmit XUV?

Where is all the energy? And when?

Tracking charge dynamics without XUV transition?

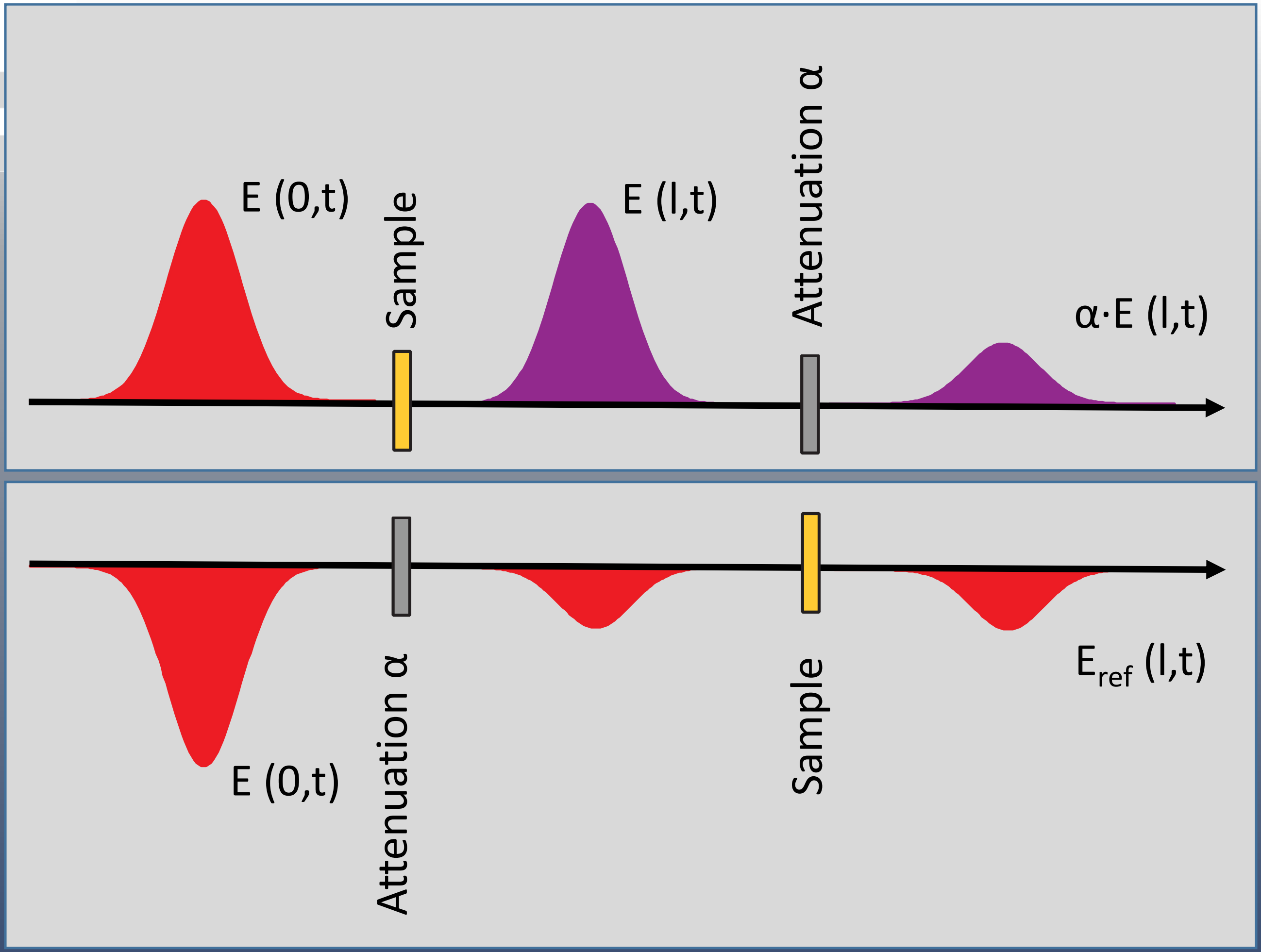


How did the eletcrons move in response to the electric field?

➡ **The dynamical polarization !**

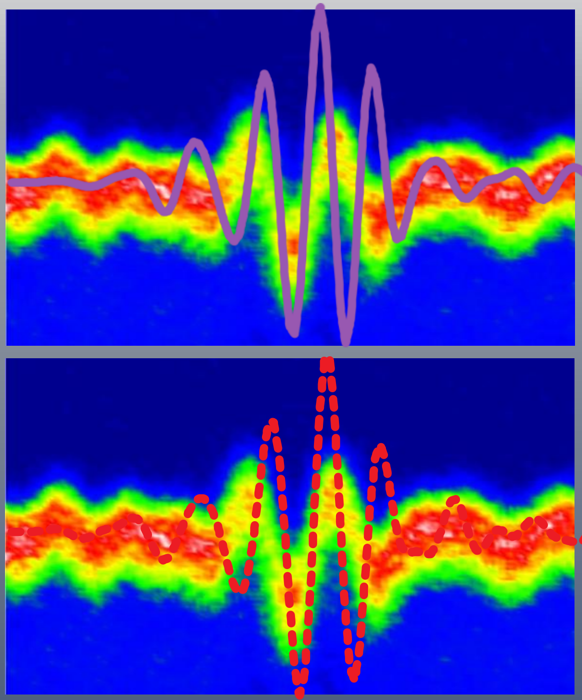
➡ **This information is radiated away. Can we detect it?**

Tracking charge dynamics without XUV transition?



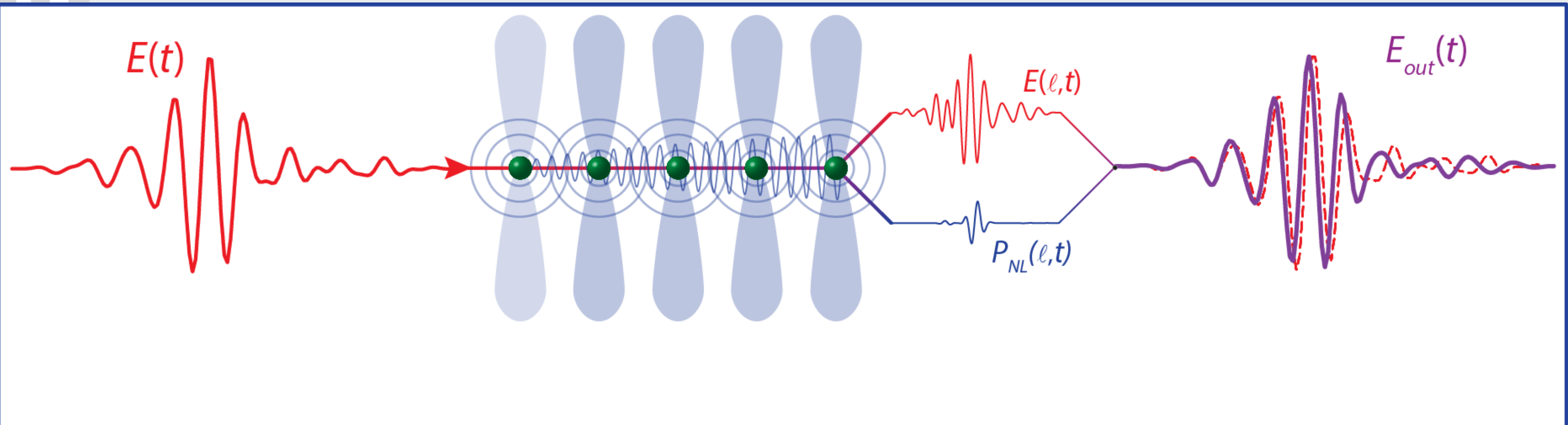
How to measure *E*(t)?

attosecond streak camera



$$P_{NL}(t) \propto E_{out}(t) - E(l,t)$$

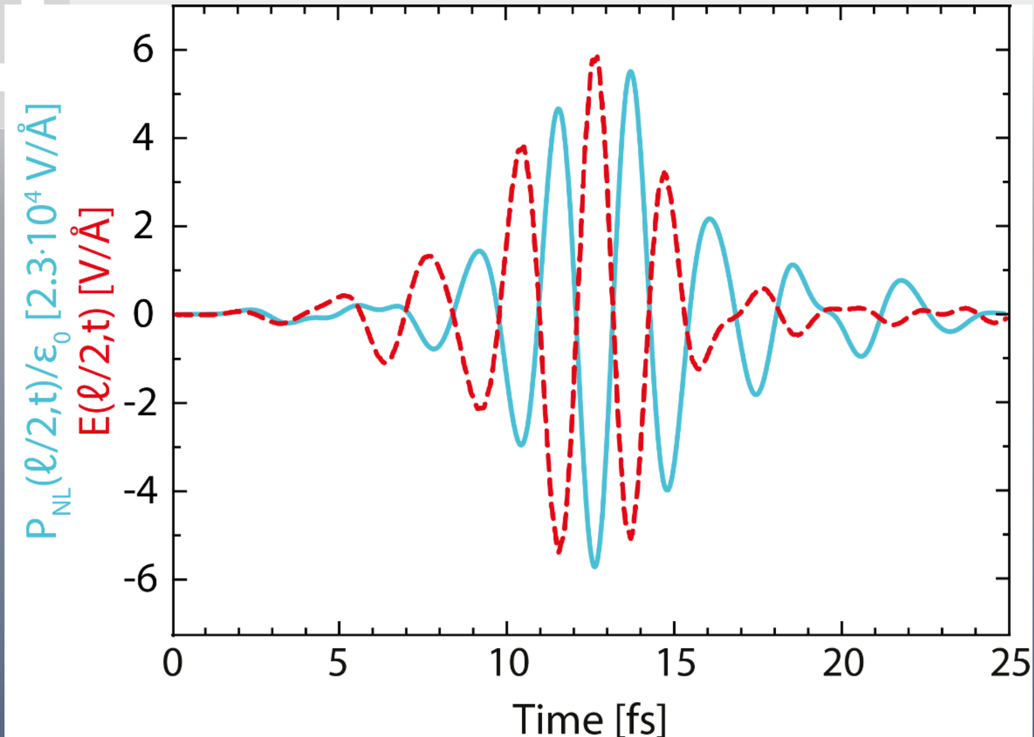
Polarization wave

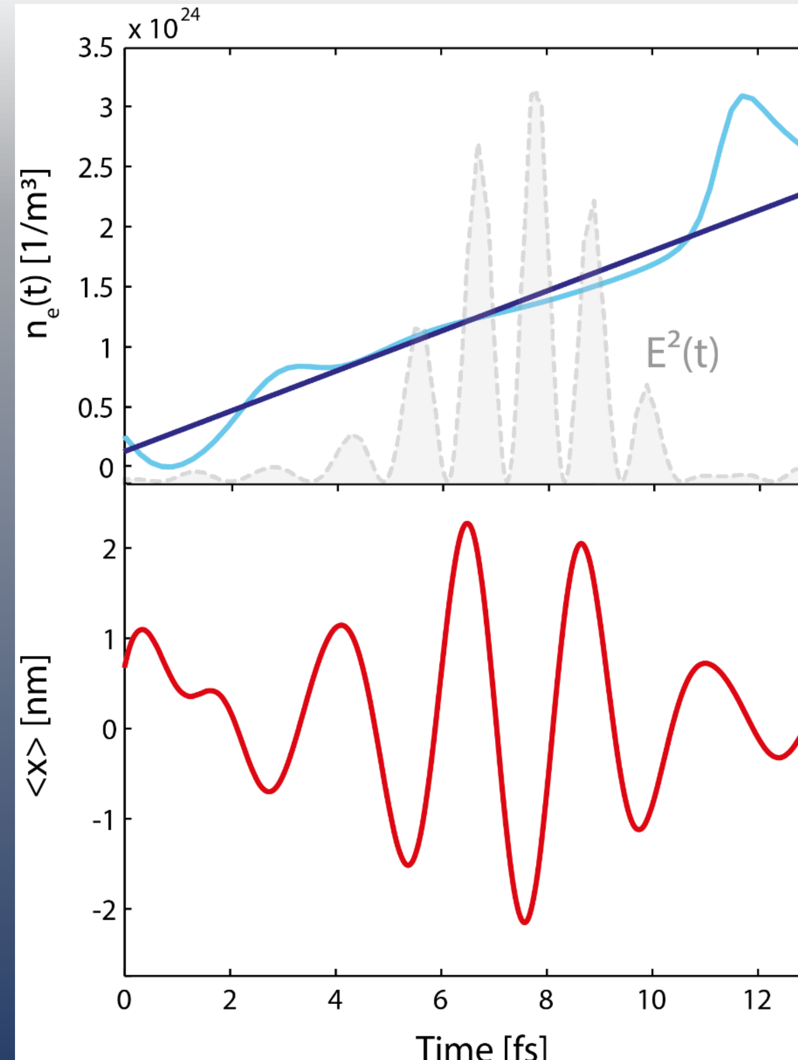


$$P(t) = P_L + P_{NL} = \varepsilon_0\left(X^{(1)}E(t) + X^{(2)}E^2(t) + X^{(3)}E^3(t) + \cdots\right)$$

$$P_{NL}(t) \propto E_{out}(t) - E(l,t)$$

Nonlinear Polarization: Plasma



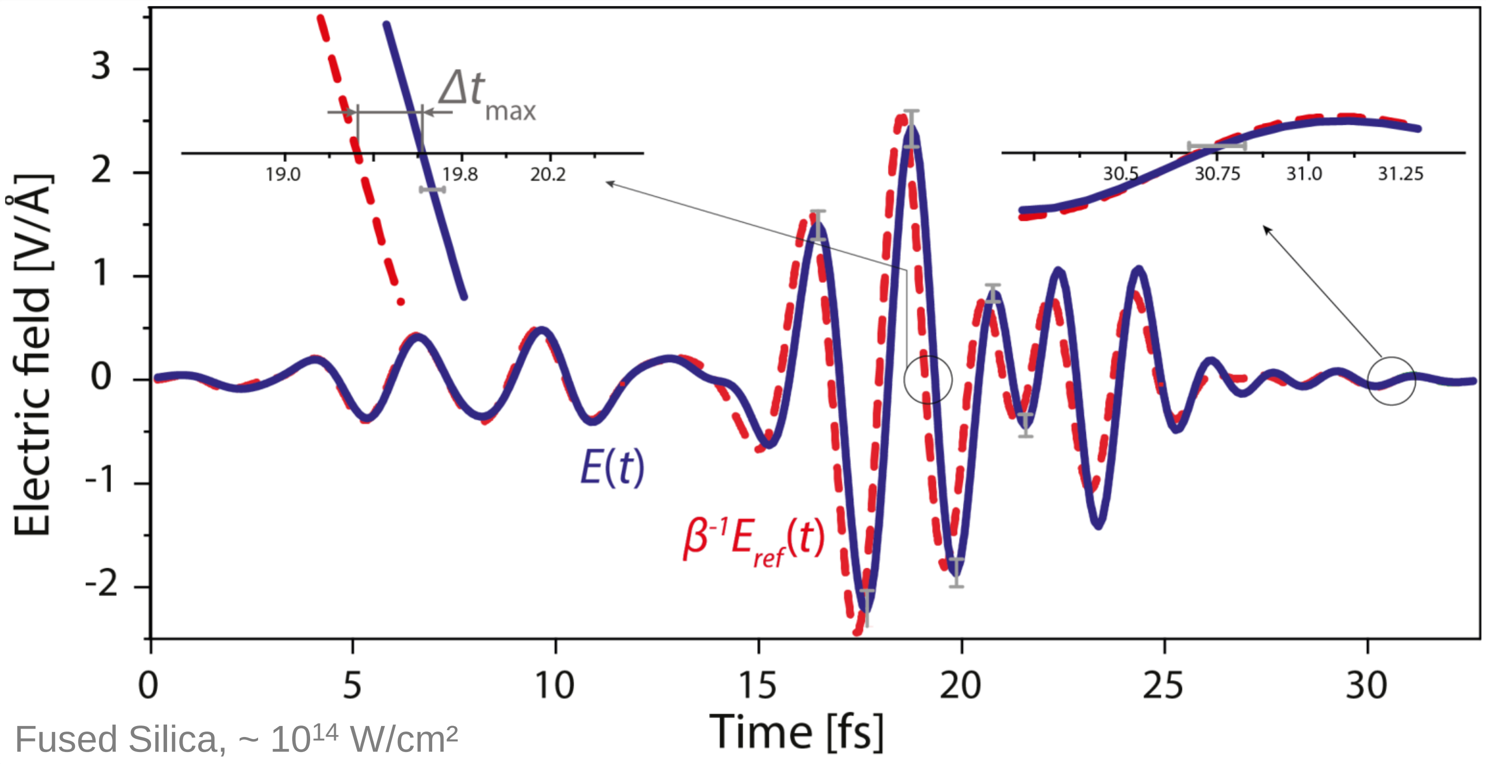


Neon, > 10^{14} W/cm²

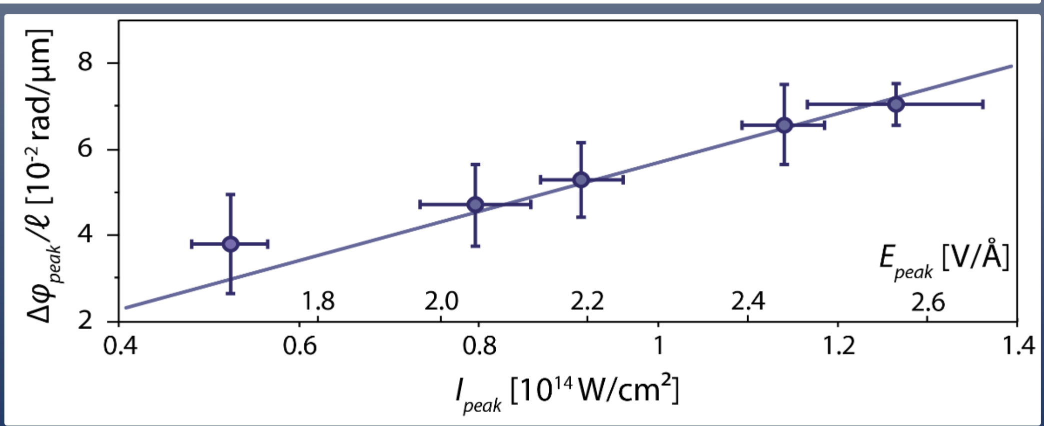
Electron trajectories in the ionization contiuuum

$$P_{NL}(t) = n_e(t) e \langle x(t) \rangle$$

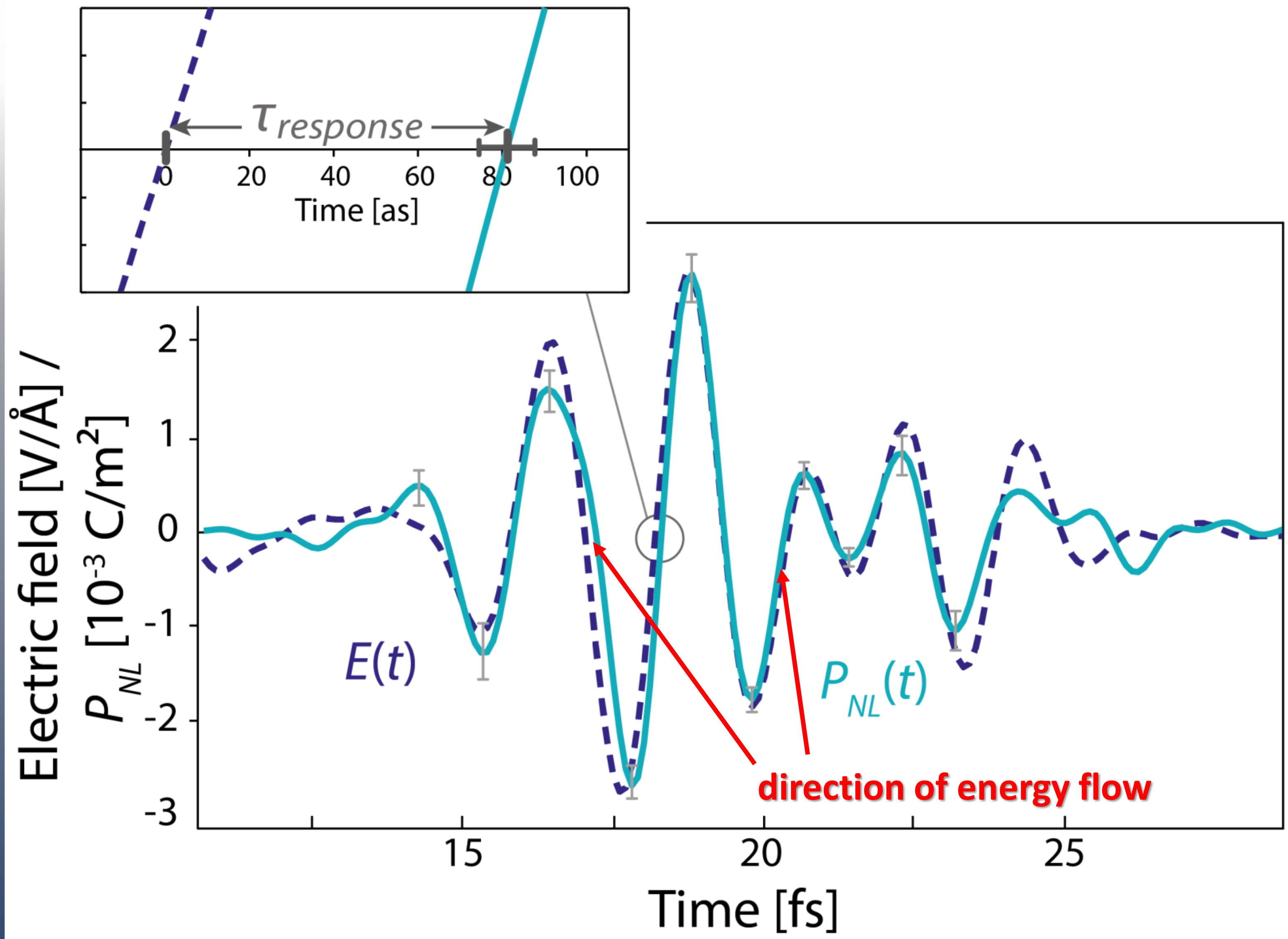
Kerr effect in time domain



$$n(I) = n_0 + n_2 I(t)$$



Nonlinear polarization wave



$$P_{NL}(t) \Leftrightarrow \langle x_{electrons}(t) \rangle$$

Energy transfer dynamics

A. Sommer et al. Nature 534, 86-90

Silica
dielectric bandstructure

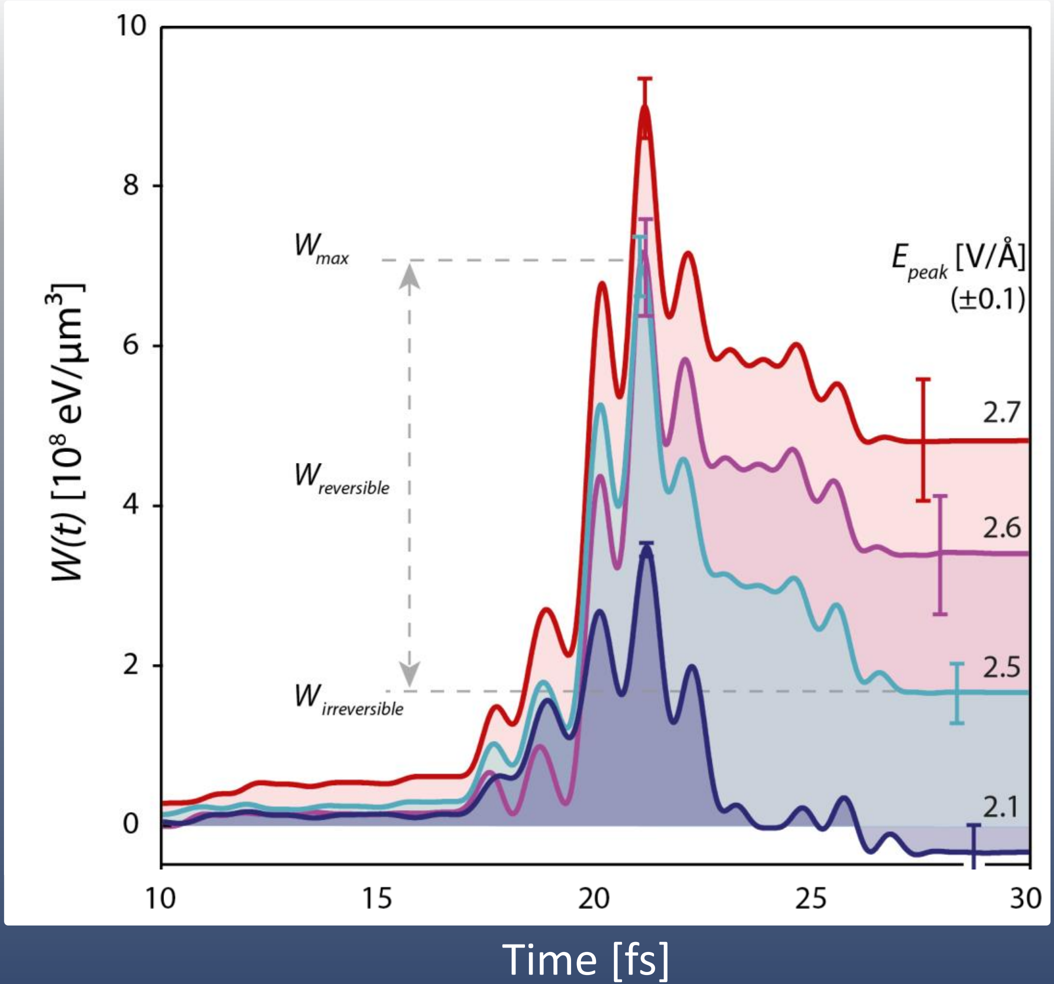
Work *W* done to the electronic system by the external field:

Electric field

$$W(t) = \int_{-\infty}^{t} E(t') * I(t')\, dt'$$

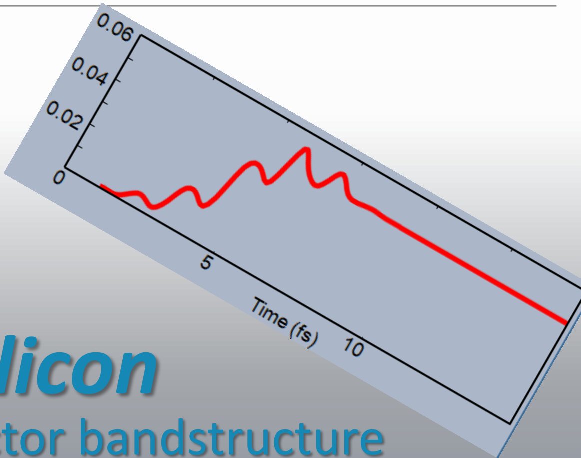
Current

We measure both!



Transient metallic behavior – without energy dissipation into the material

Energy transfer dynamics

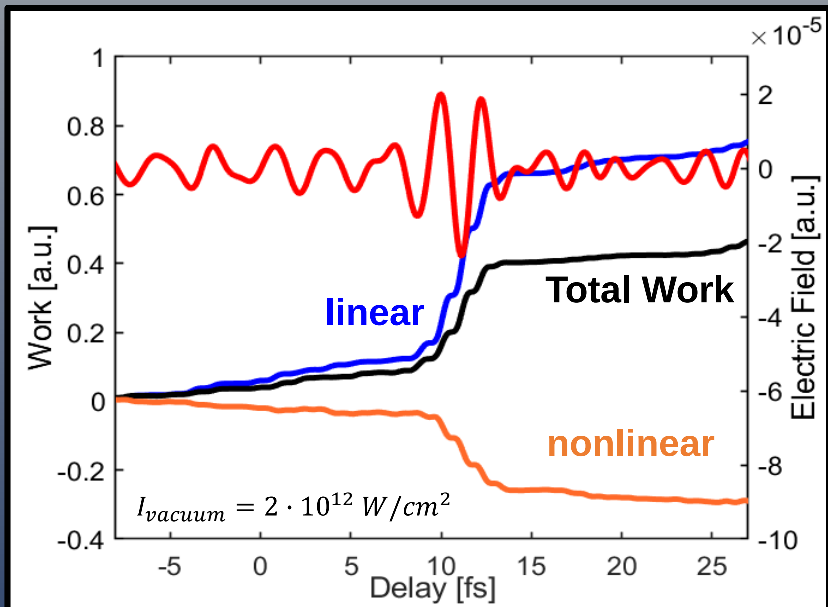


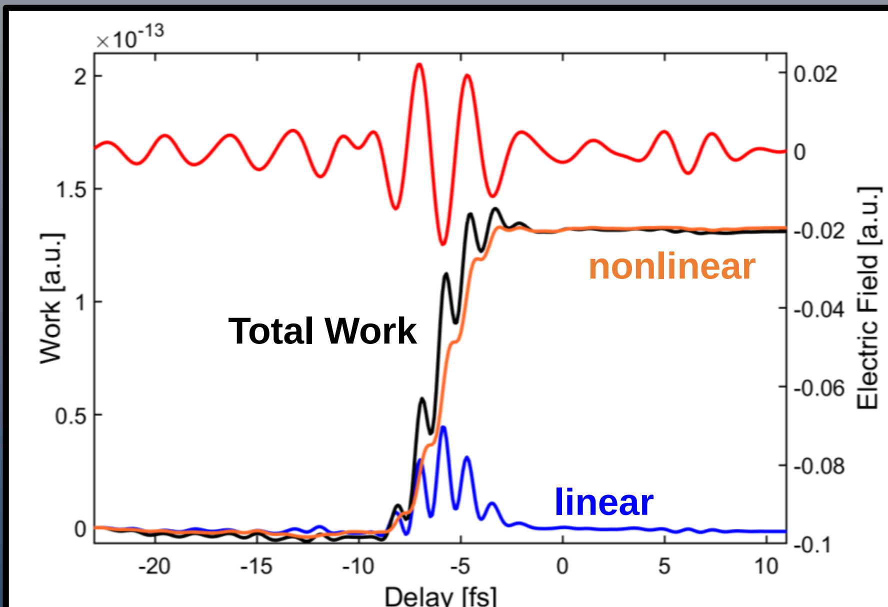
Nickel

metallic bandstructure

Silicon

semiconductor bandstructure





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t

Optoelectronics at max. speed

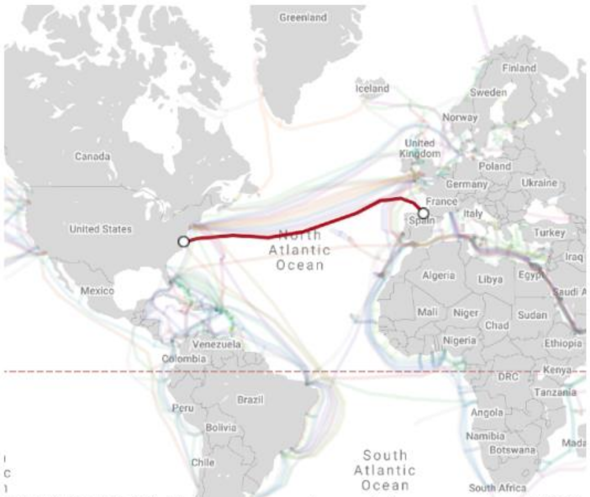
1 bit every 38 Femtoseconds

- Requires *phase* sensitivity: coherent/heterodyne detection

- Massive wavelength-division multiplexing vs. bandwidth limits

How fast could extreme opto-electronics be?

Light-Electronics Interconnect at PHz clock rates?



M2E.6.pdf OFC 2019 © OSA 2019

Real-time 16QAM Transatlantic Record Spectral Efficiency of 6.21 b/s/Hz Enabling 26.2 Tbps Capacity

Stephen Grubb[1], Pierre Mertz[2], Ales Kumpera[3], Lee Dardis[4], Jeffrey Rahn[4], James O'Connor[4], Matthew Mitchell[1]
[1]*Facebook, 1 Hacker Way, Menlo Park, CA 94025*
[2]*Infinera Maryland, 9005 Junction Dr., Savage, MD 20763*
[3]*Infinera Canada, 555 Legget Dr, Ottawa, ON K2K 2X3, Canada*
[4]*Infinera Corporation, 140 Caspian Ct., Sunnyvale, CA 94089*
E-mail address: pmertz@infinera.com

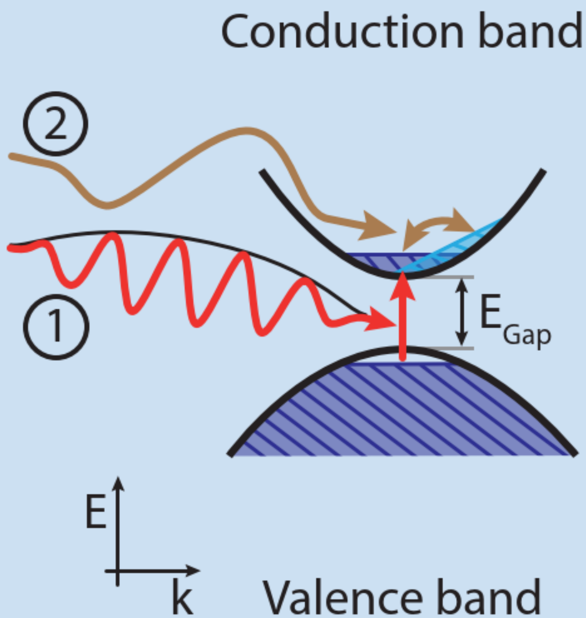
Abstract: Real-time, error-free 16QAM transmission at a record spectral efficiency of 6.21 b/s/Hz enables transatlantic (6,644 km) fiber capacity of 26.2 Tbps, using precision, multi-carrier common wavelocking; digitally synthesized subcarriers; near-Nyquist pulse shaping; and large-area, positive dispersion fiber. © 2019 The Author(s)
OCIS codes: (060.2330) Fiber optics communications; (060.1660) Coherent communications

Optical Antenna – ultrafast?

IEEE JOURNAL OF QUANTUM ELECTRONICS, VOL. 24, NO. 2, FEBRUARY 1988 255

Subpicosecond Photoconducting Dipole Antennas

PETER R. SMITH, DAVID H. AUSTON, MEMBER, IEEE, AND MARTIN C. NUSS

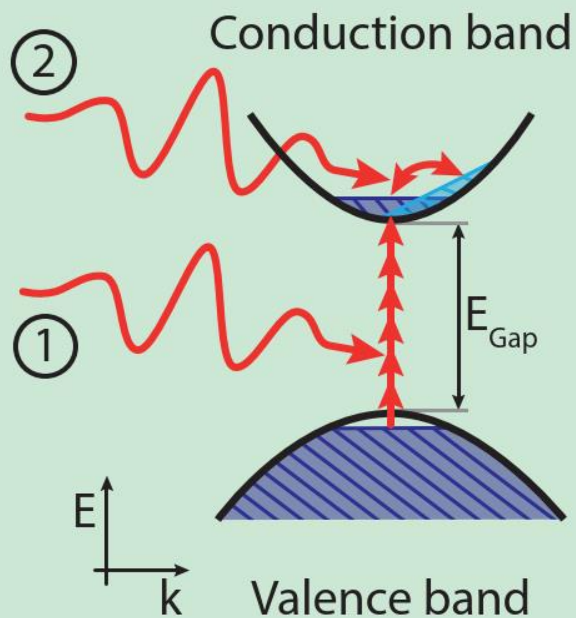


LETTER

doi:10.1038/nature11567

Optical-field-induced current in dielectrics

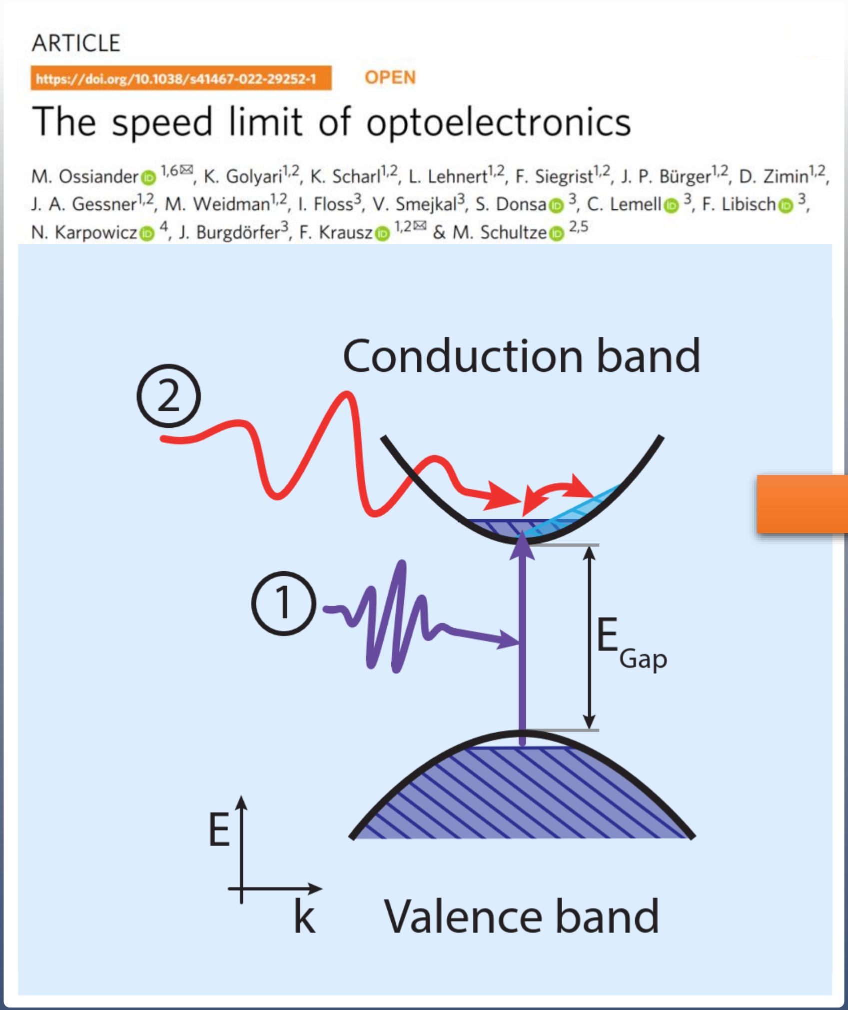
Agustin Schiffrin[1]†, Tim Paasch-Colberg[1], Nicholas Karpowicz[1], Vadym Apalkov[2], Daniel Gerster[3], Sascha Mühlbrandt[1,3], Michael Korbman[1], Joachim Reichert[3], Martin Schultze[1,4], Simon Holzner[1], Johannes V. Barth[3], Reinhard Kienberger[1,3], Ralph Ernstorfer[1,3,5], Vladislav S. Yakovlev[1,4], Mark I. Stockman[2] & Ferenc Krausz[1,4]

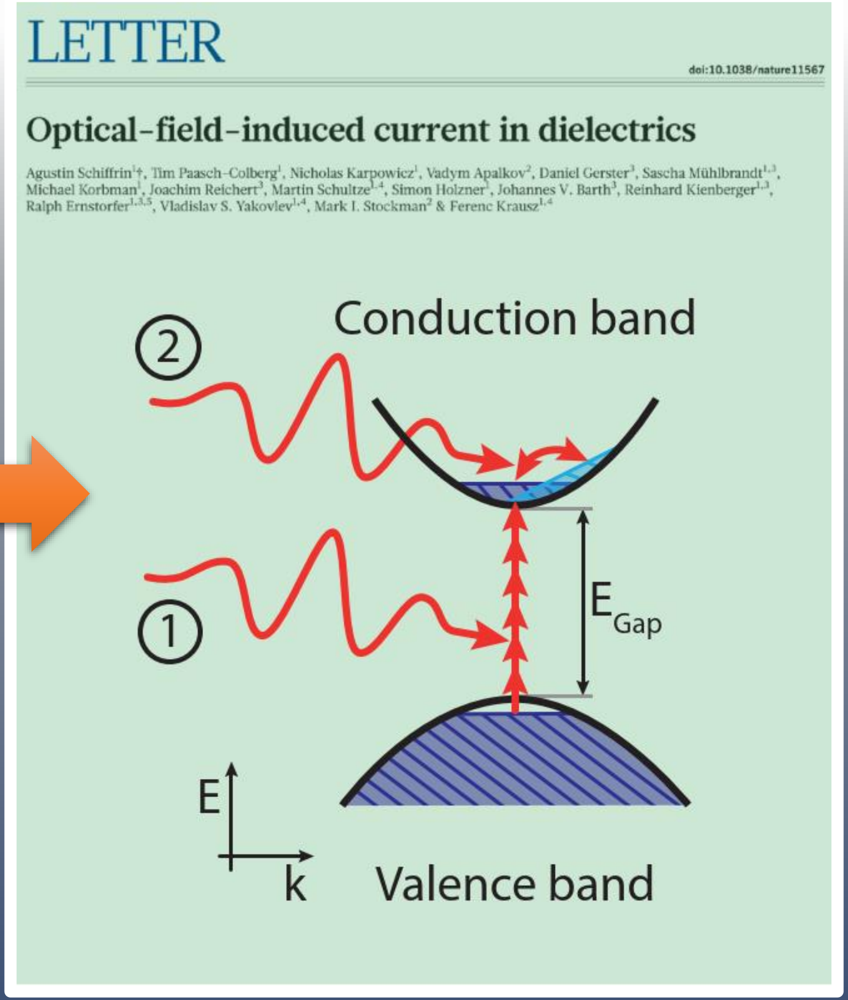


Linear Photodoping –
~DC carrier acceleration

Nonlinear excitation–
Strong field carrier acceleration

Ultrafast Optical Antenna = UV?



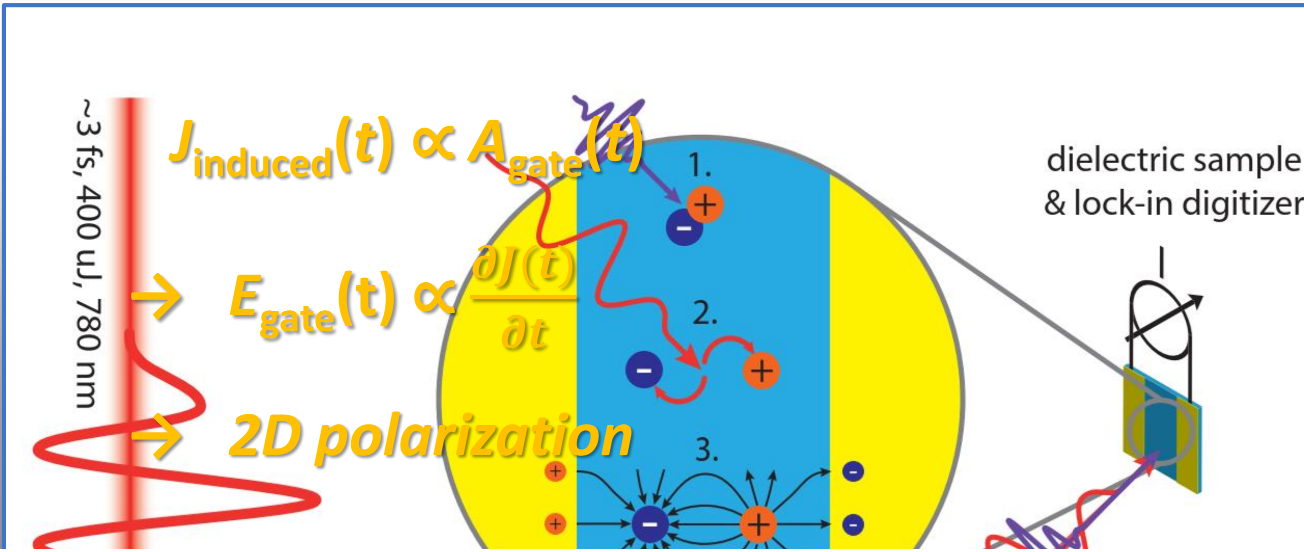


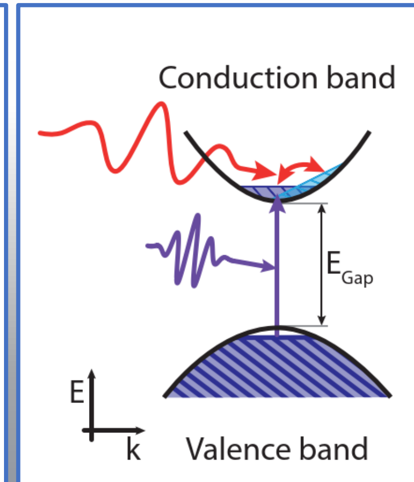
Attosecond Photodoping–
~DC gate field

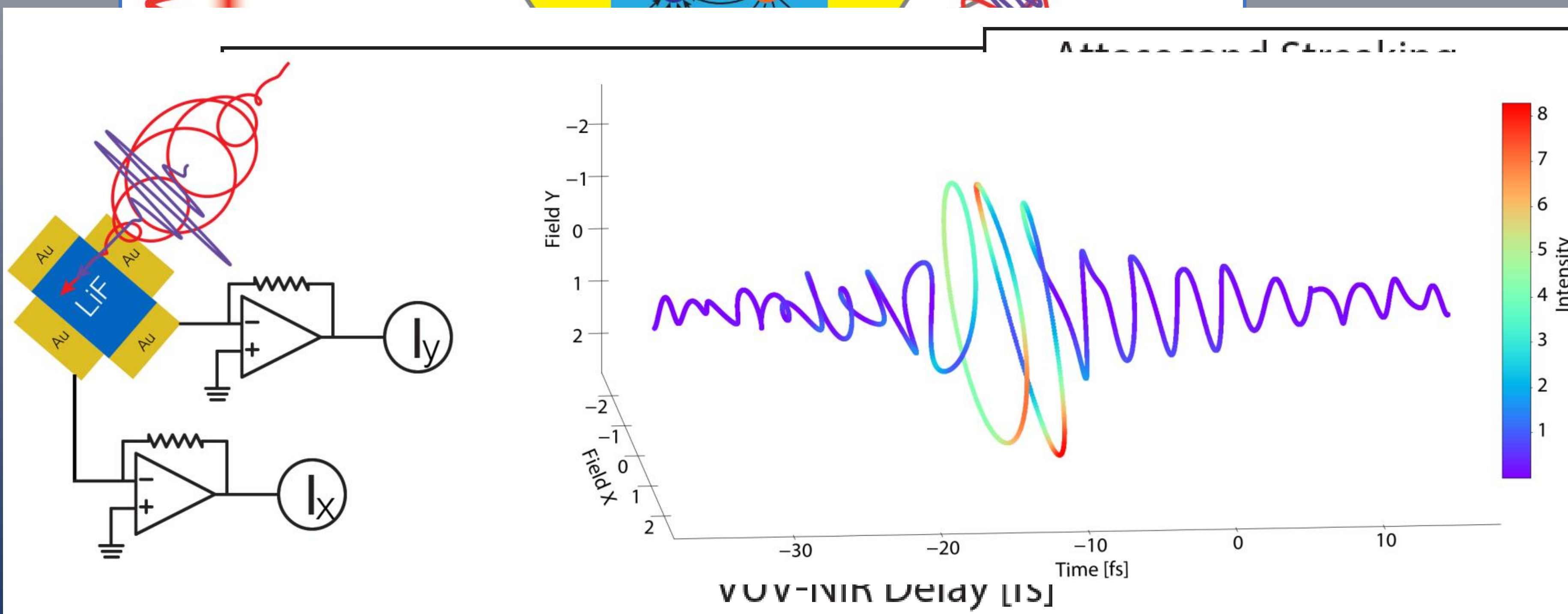
Linear Resonant injection–
Optical excitation

Nonlinear excitation–
Strong field carrier acceleration

Attosecond Optical Antenna





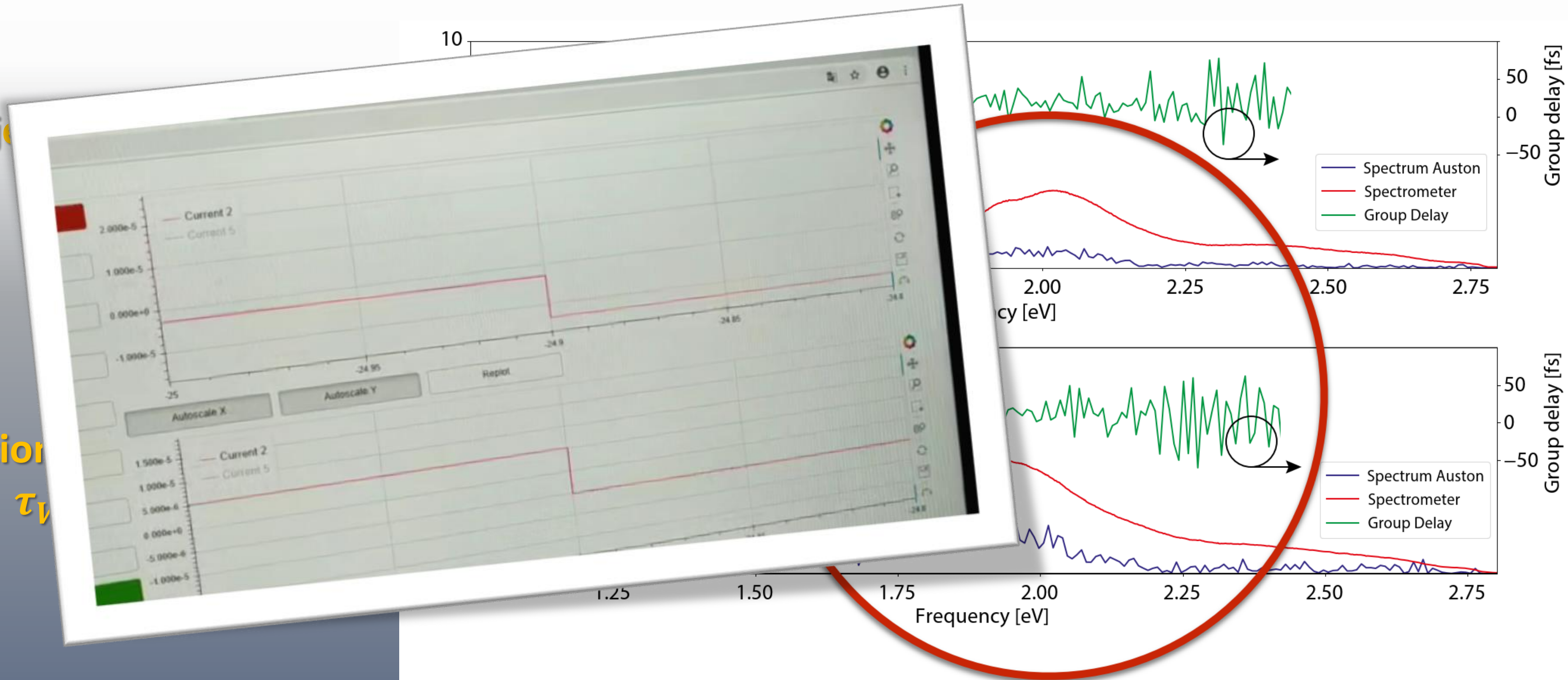


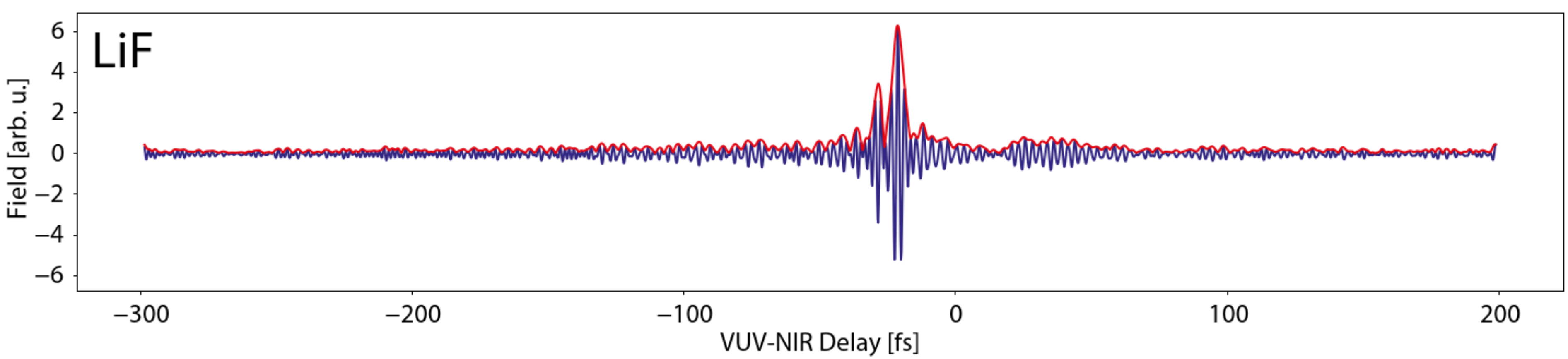
Solid-State Field Sampling

Inj

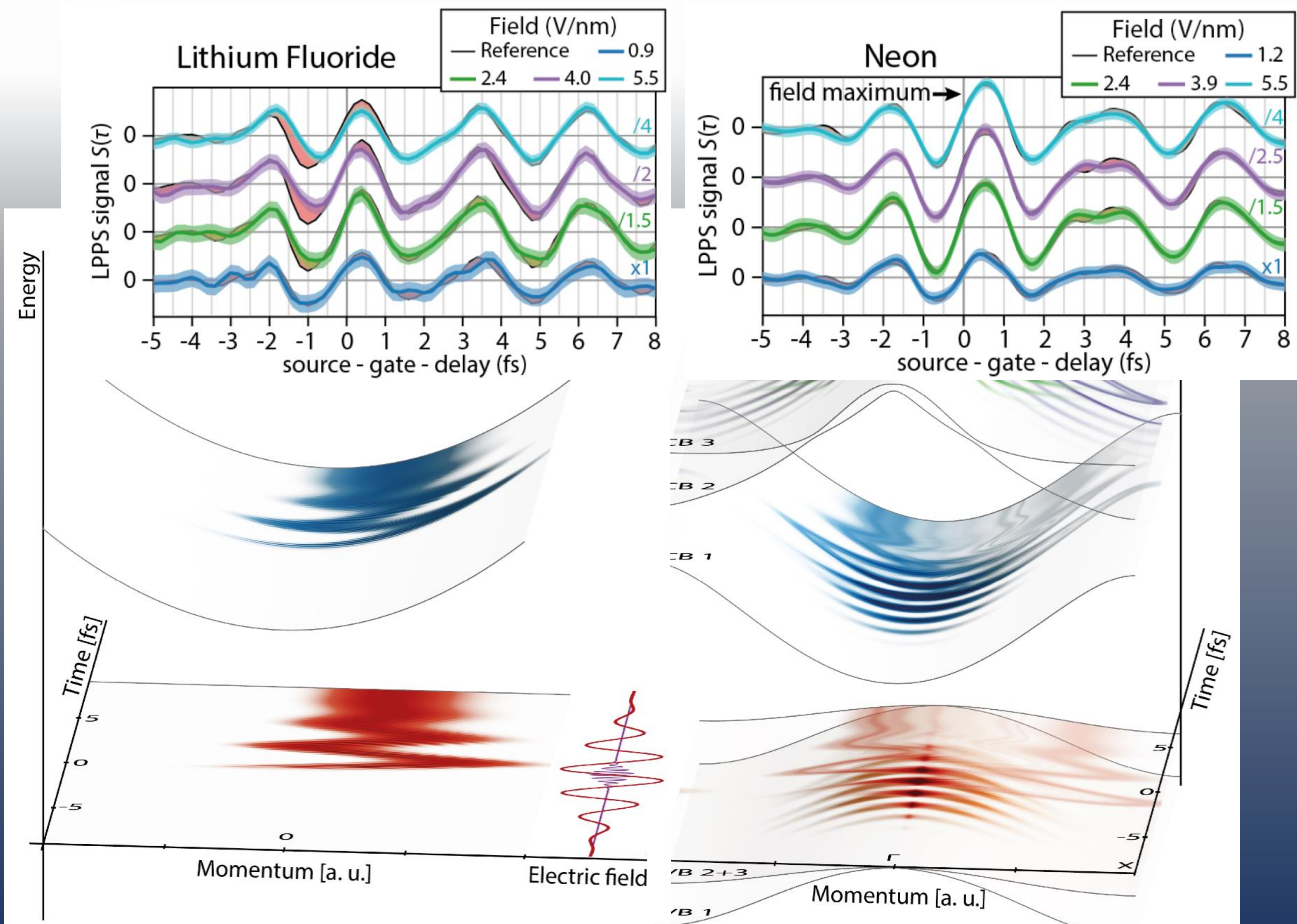
Injection

τ_V

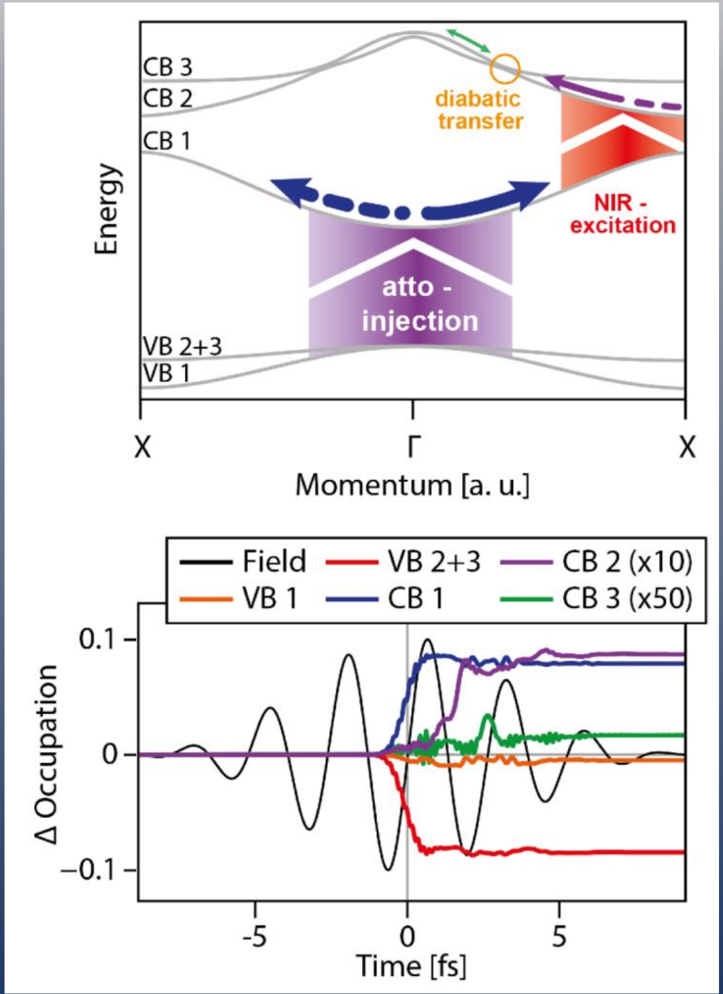


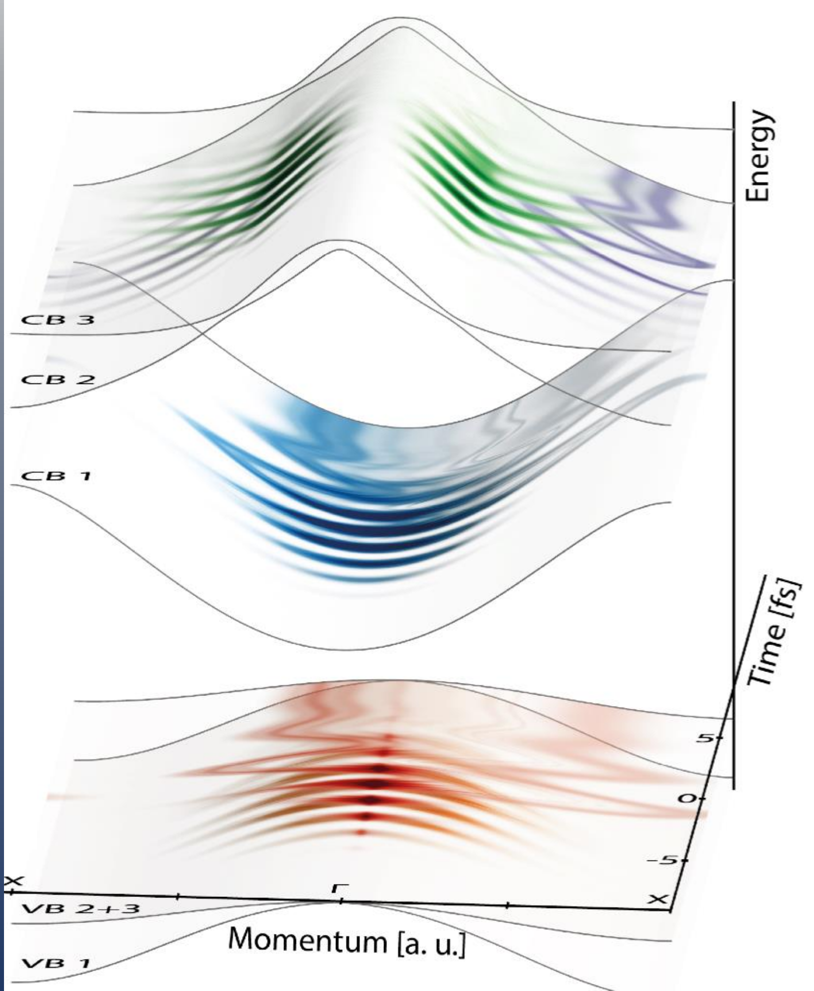


Momentum Transfer - Theory



Momentum Transfer - Theory





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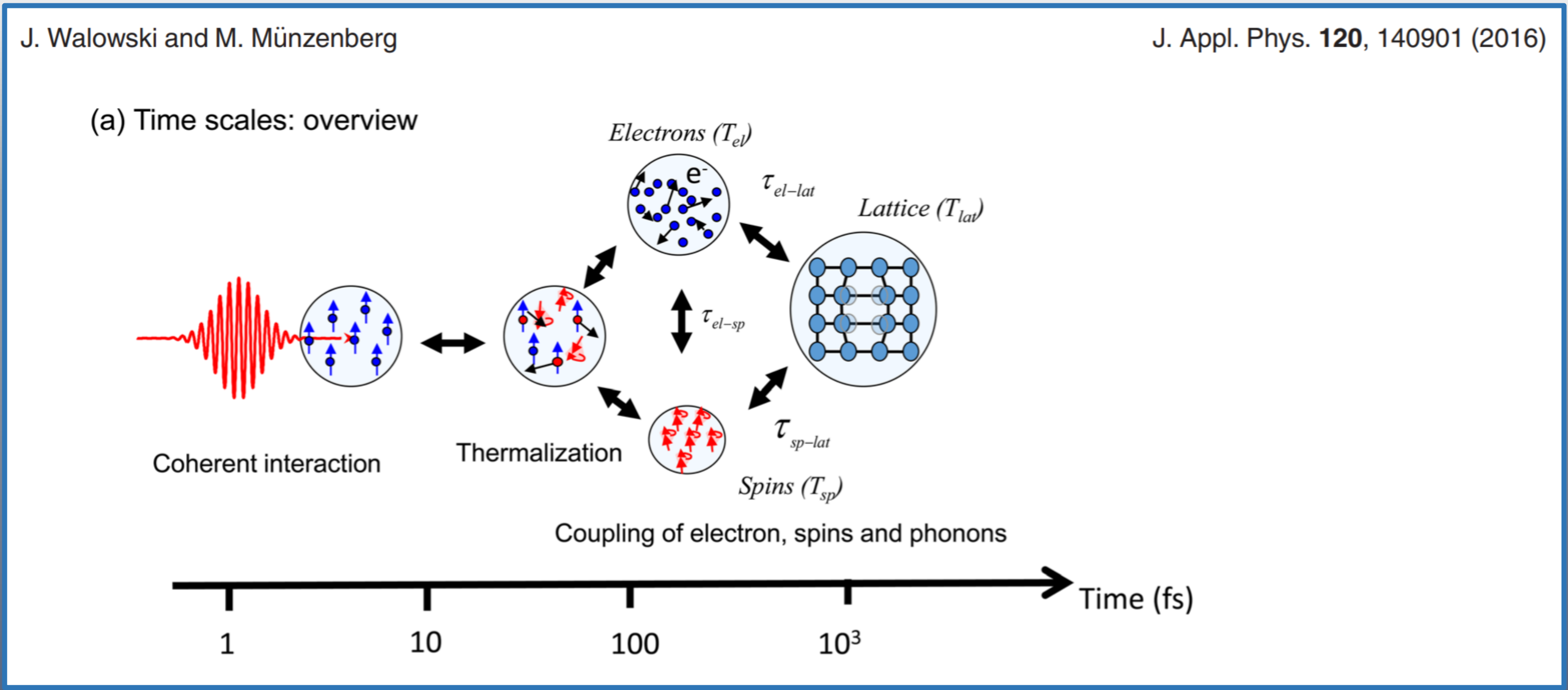
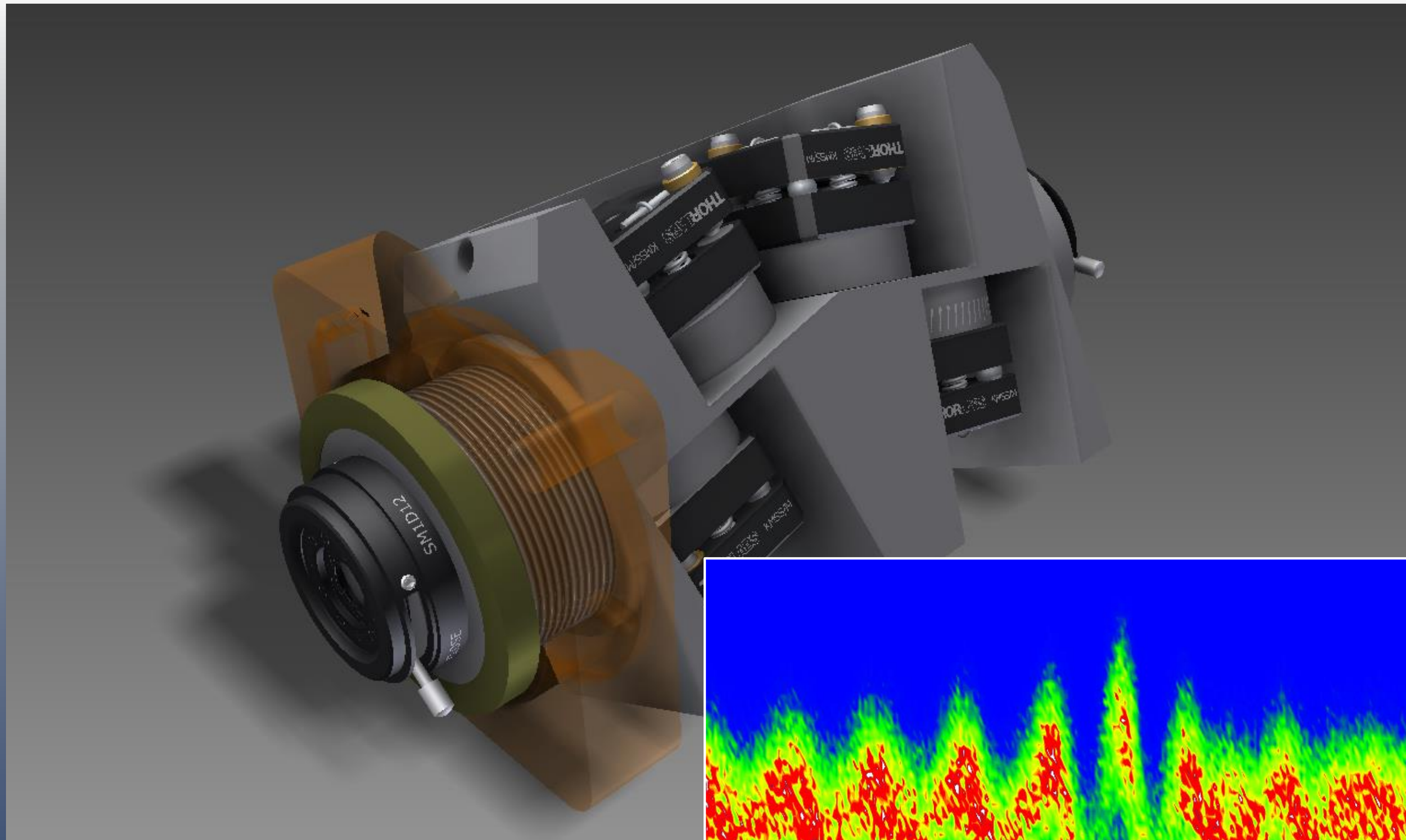
Spin dynamics & Ultrafast Magentism

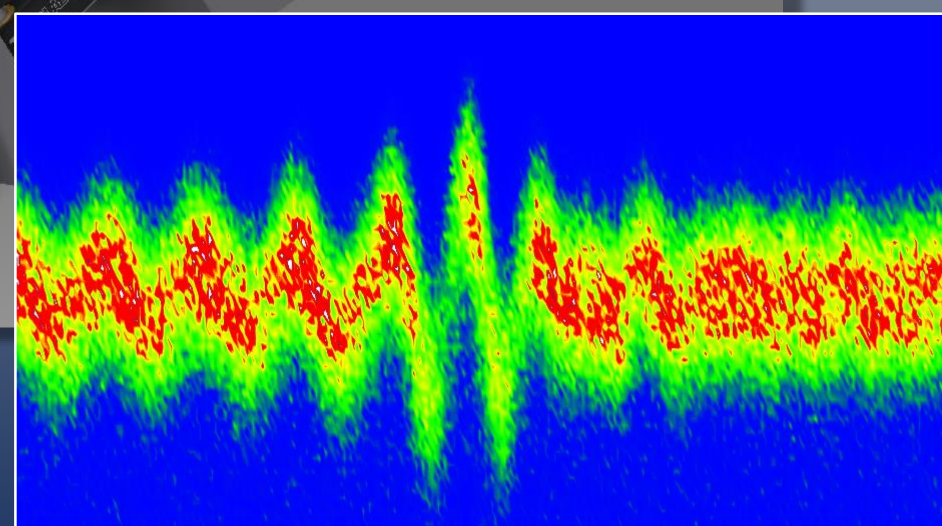


hook up spin system to optical carrier control ?

ultrafast

Circularly polarized XUV light

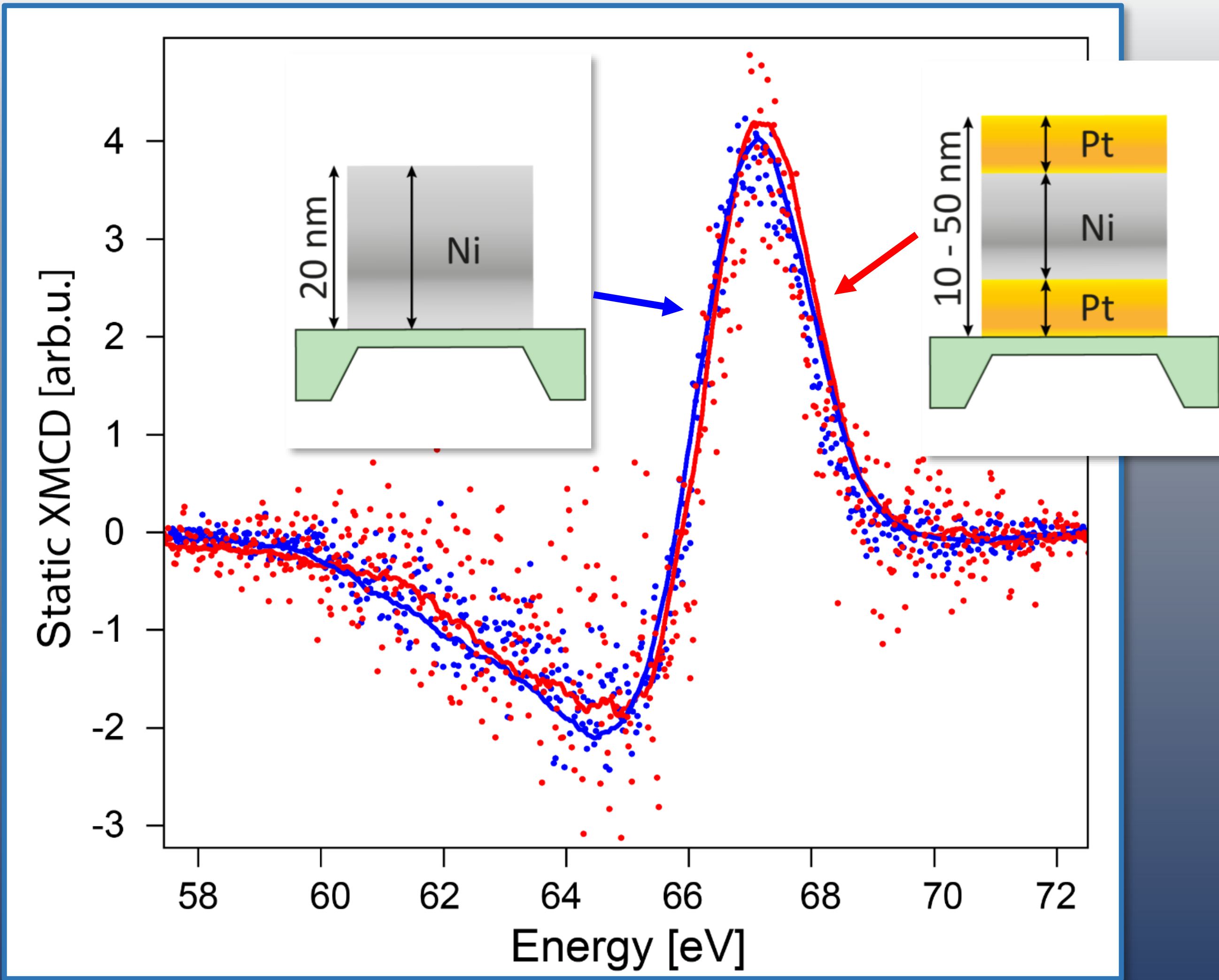


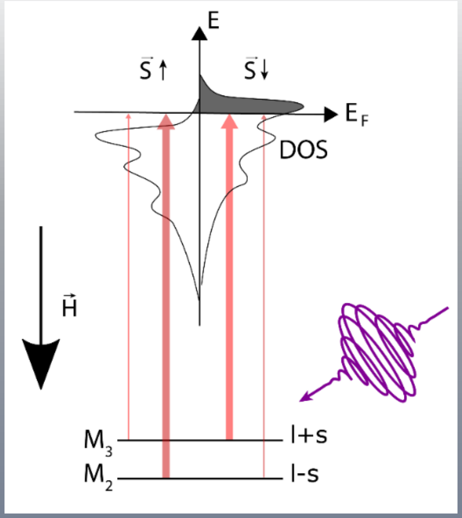


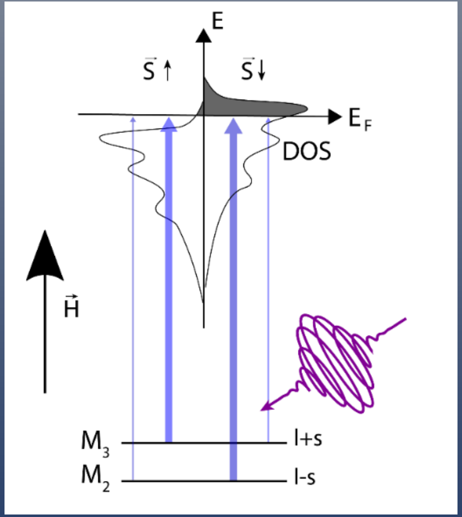
All-reflective
XUV waveplate

Attosecond XMCD?

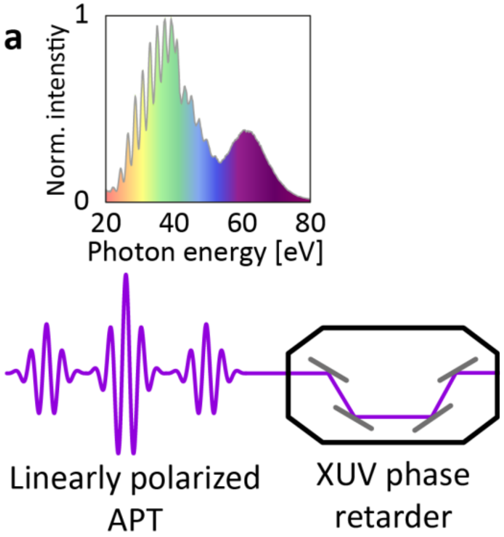
X-Ray **M**agnetic **C**ircular **D**ichroism

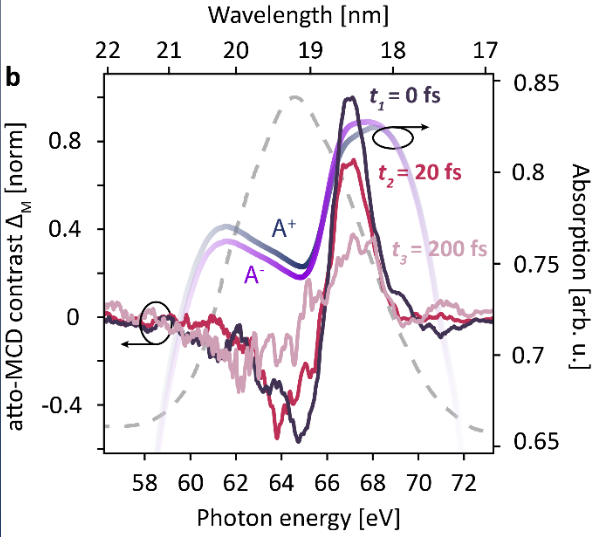






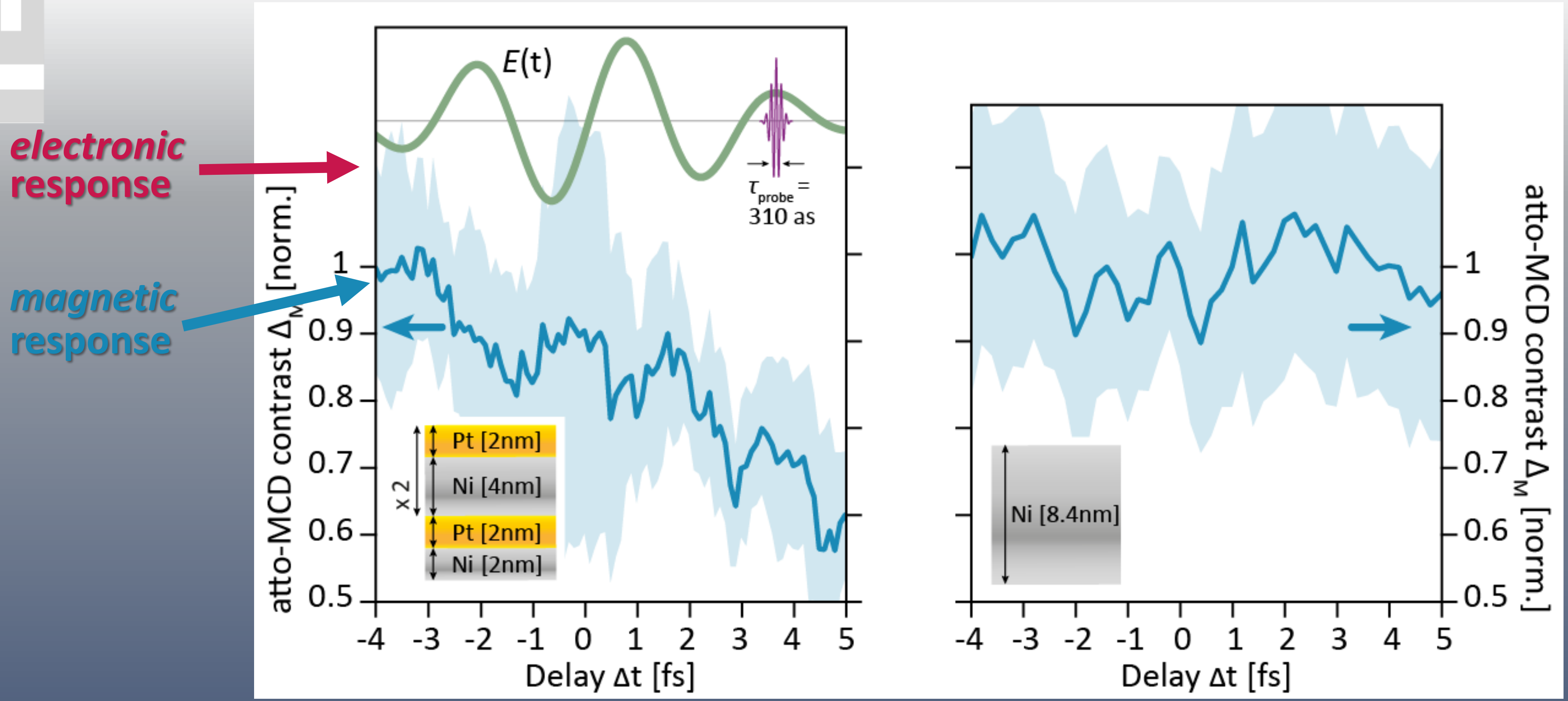
Spin-sensitive detection





Attosecond transient Absorption is capable of measuring Magnetization Dynamics!

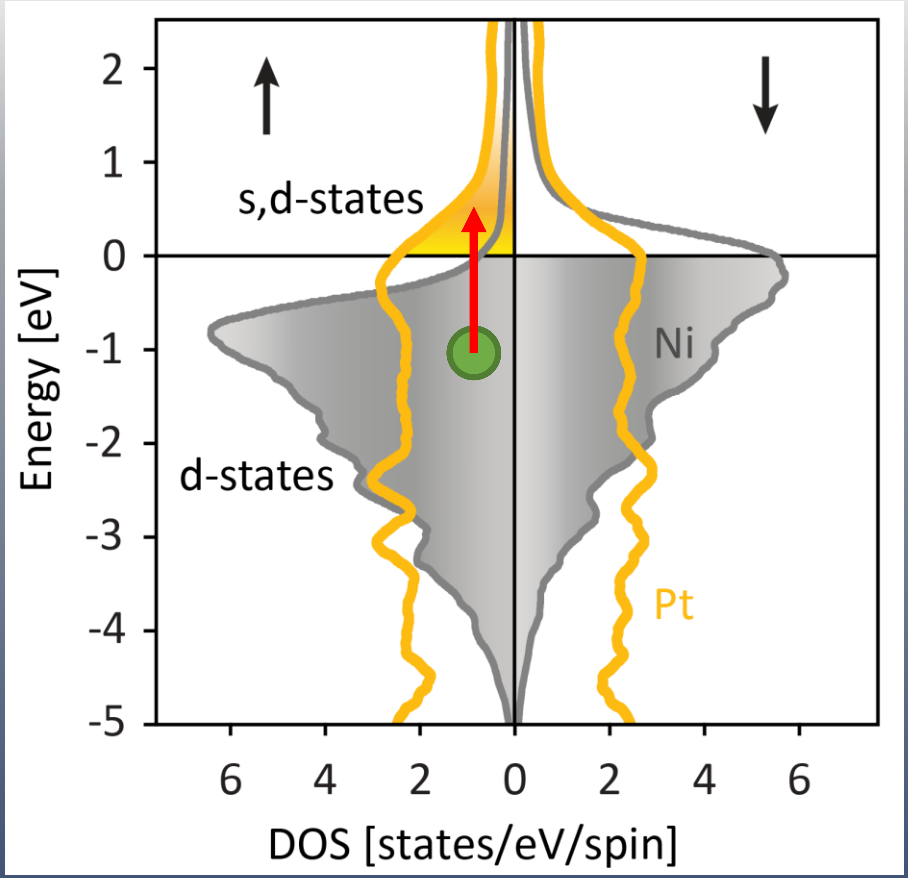
Coherent Magnetization Dynamics

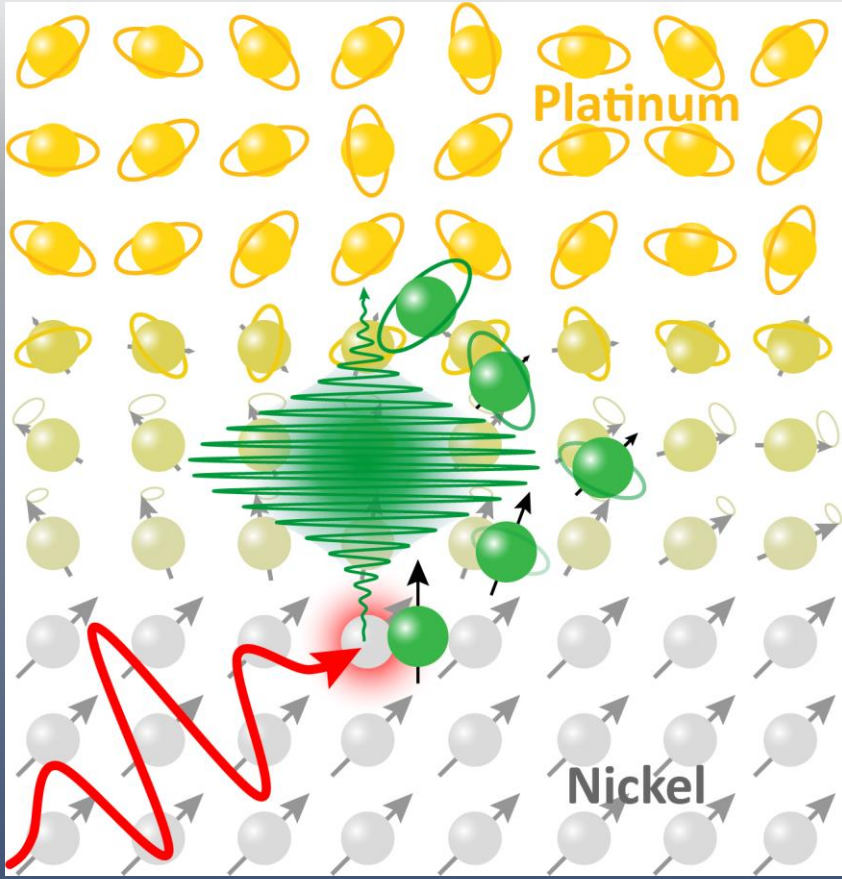


F. Siegrist *et al.*, Light-wave dynamic control of magnetism, Nature **571**, 240–244 (2019)

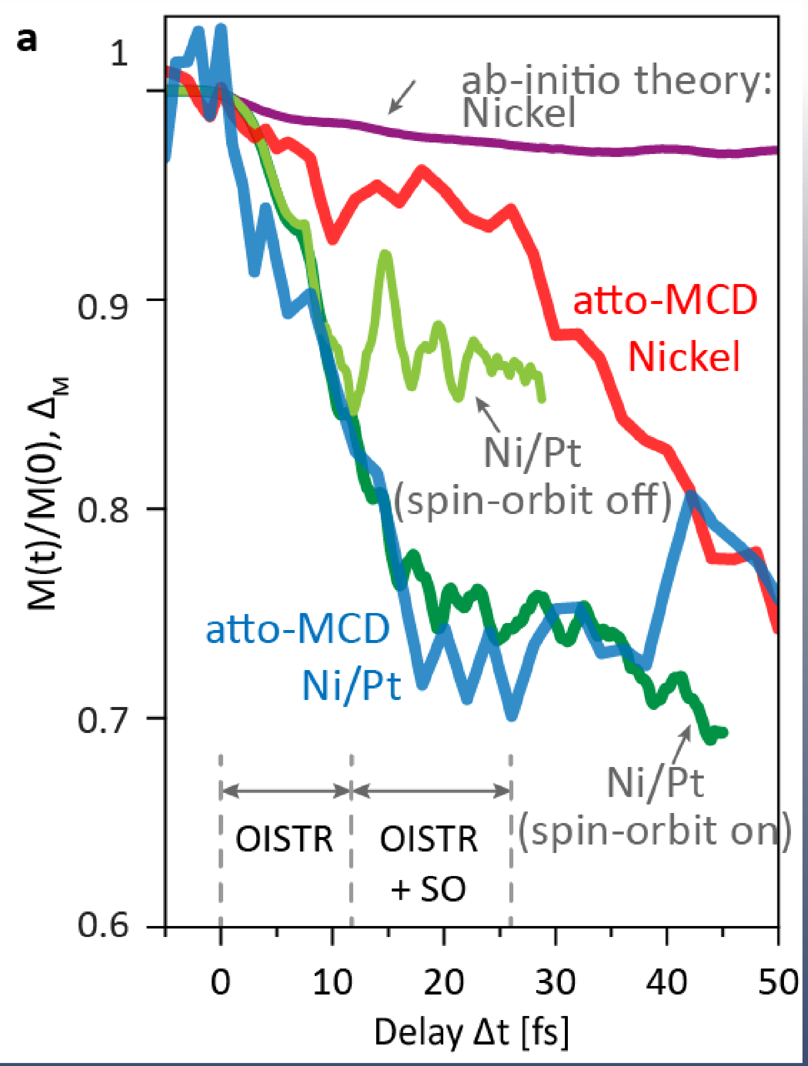
Optically Induced Spin Transfer

OISTR





Hierarchy of timescales



F. Siegrist *et al.*, Light-wave dynamic control of magnetism, Nature **571**, 240–244 (2019)

Short optical fields to control charge and spin in the condensed phase

Ultrafast laser fields can

manipulate electron and spin states

… possibly *faster* than the decoherence