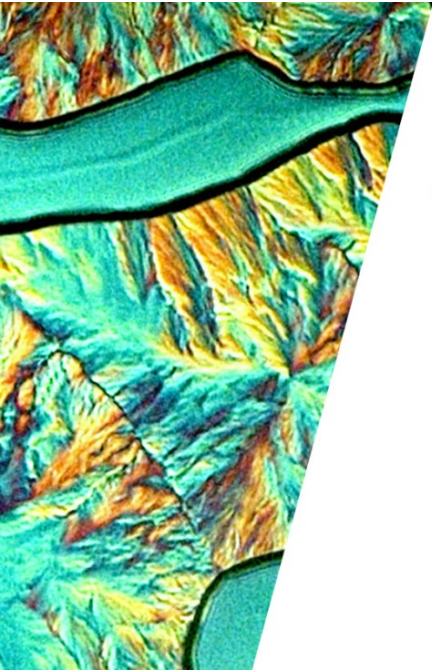


Attosecond Spectroscopy to Track Ultrafast Electron Dynamics

Presented by:



The OSA Nonlinear Optics Technical Group Welcomes You!



ATTOSECOND SPECTROSCOPY TO TRACK ULTRAFAST ELECTRON DYNAMICS WEBINAR

13 June 2018 • 10:30 EDT

OSA Nonlinear
Optics
Technical Group

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Technical Group Leadership 2018



Chair

Ajanta Barh

DTU Fotonik, Denmark



Event officer

Ryan T. Glasser

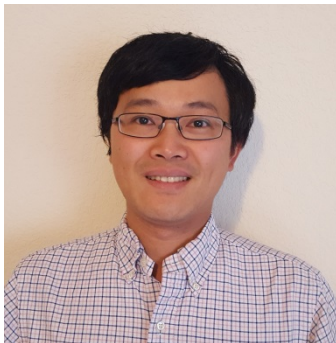
Tulane University, USA



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Technical Group at a glance

- **Focus**

- “Physics of nonlinear optical materials, processes, devices, & applications”
- **3800** members (**largest** in OIS, 3rd largest in OSA)

- **Mission**

- To benefit YOU
- webinars, e-Presence, publications, technical events, business events, outreach
- Interested in presenting your research? Have ideas for TG events? Contact us at

- **Email:** TGNonlinearOptics@osa.org

- Find us here

- www.osa.org

- **Facebook:** www.facebook.com/osanonlinearoptics

- **LinkedIn:** www.linkedin.com/groups/8302249

Today's webinar



Attosecond spectroscopy to track ultrafast electron dynamics

Speaker's short Bio:

Graduation in Physics at ETH Zürich, CH

Ph.D. degree from Ludwig-Maximilians-Universität of Munich, DE

Postdoc at the University of California in Berkeley, USA.

Dr. Martin Schultze

Research Group leader of Attosecond experiments

Max-Planck-Institut für Quantenoptik, Garching, Germany

martin.schultze@mpq.mpg.de



ATTOWORLD
Laboratory for Attosecond Physics



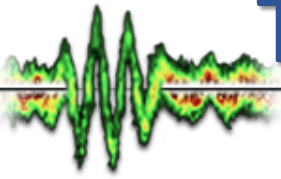
OSA Technical Group Webinar

13.06.2018

Martin Schultze
LMU München & MPI f. Quantenoptik
www.attoworld.de

Attosecond Spectroscopy to Track Ultrafast Electron Dynamics

Time-domain Measurements ?



to observe ultrafast processes

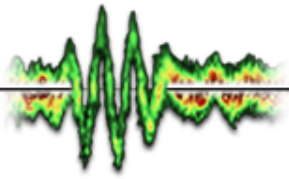
$\tau_{\text{shutter}}: 1300 \mu\text{s}$



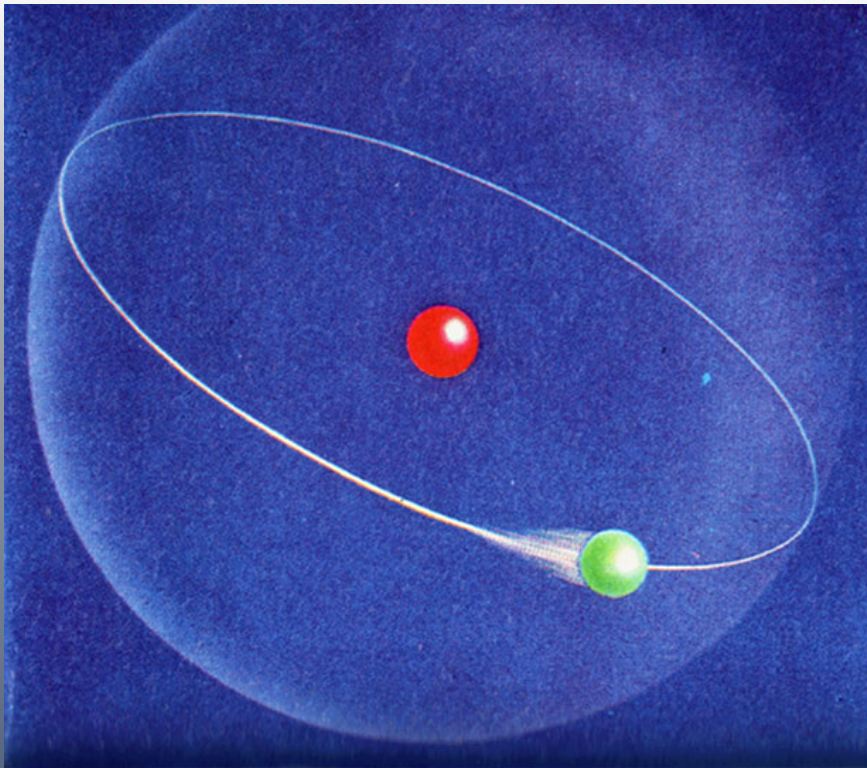
$\tau_{\text{shutter}}: 400 \mu\text{s}$



How fast do we need to get



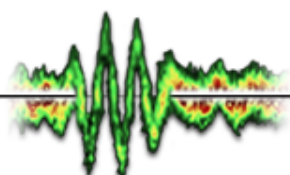
to track quantum mechanical effects in time domain?



For Hydrogen one revolution
according to Bohr's model takes
150 Attoseconds
= 0.000 000 000 000 000 15 s

Light travels about 0.3 nm in this time interval

⇒ Approx. the size of an atom



What do we want to know ?

RESPONSE TIMES:

How quickly can we switch between two states ?

CORRELATIONS:

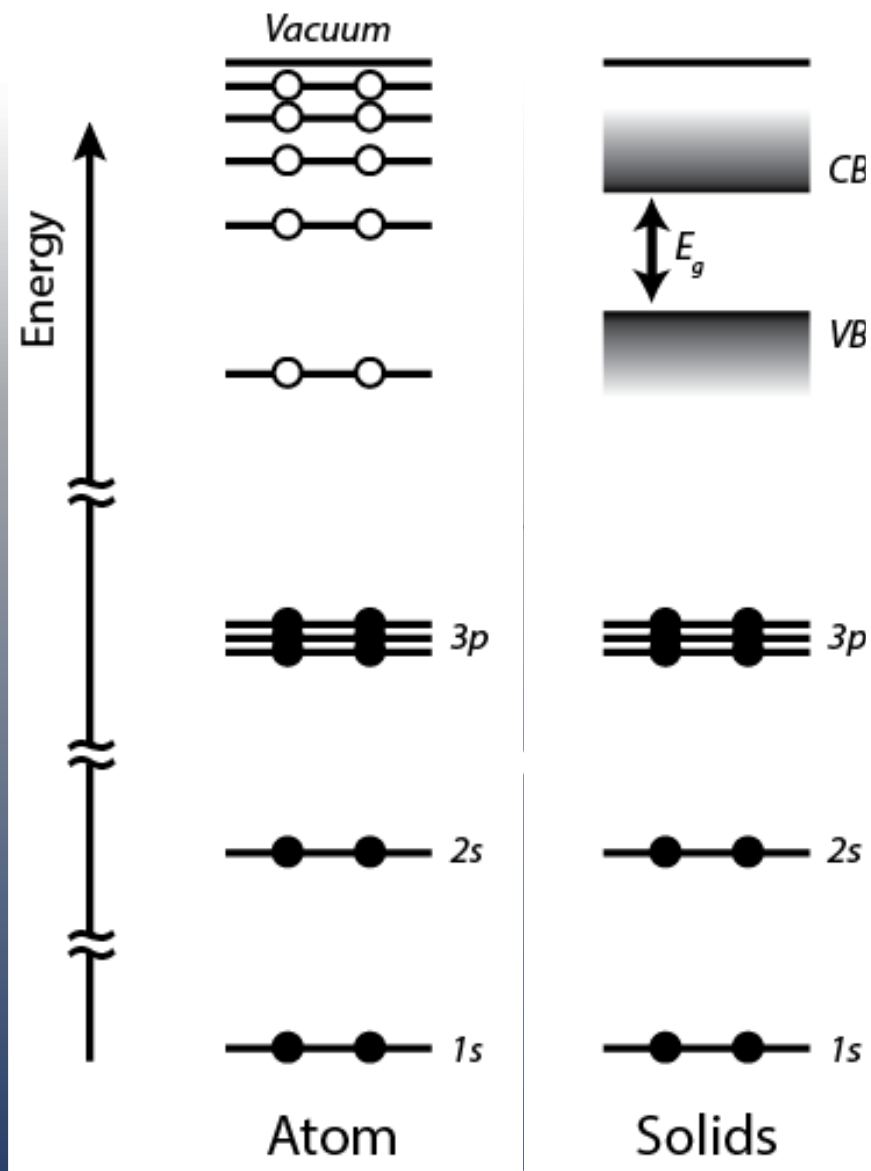
Effects of multi-electron interactions

DISSIPATION:

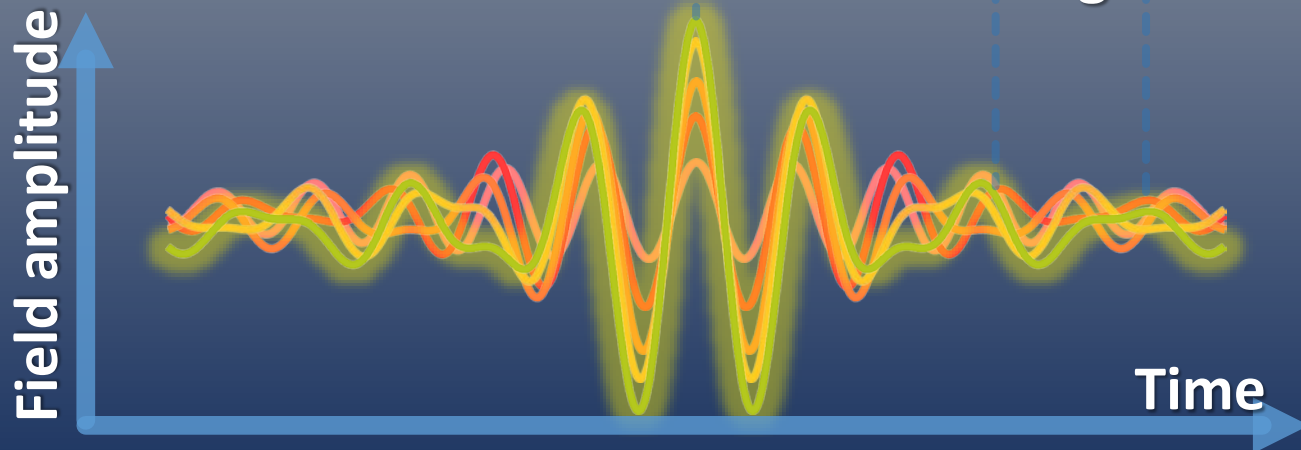
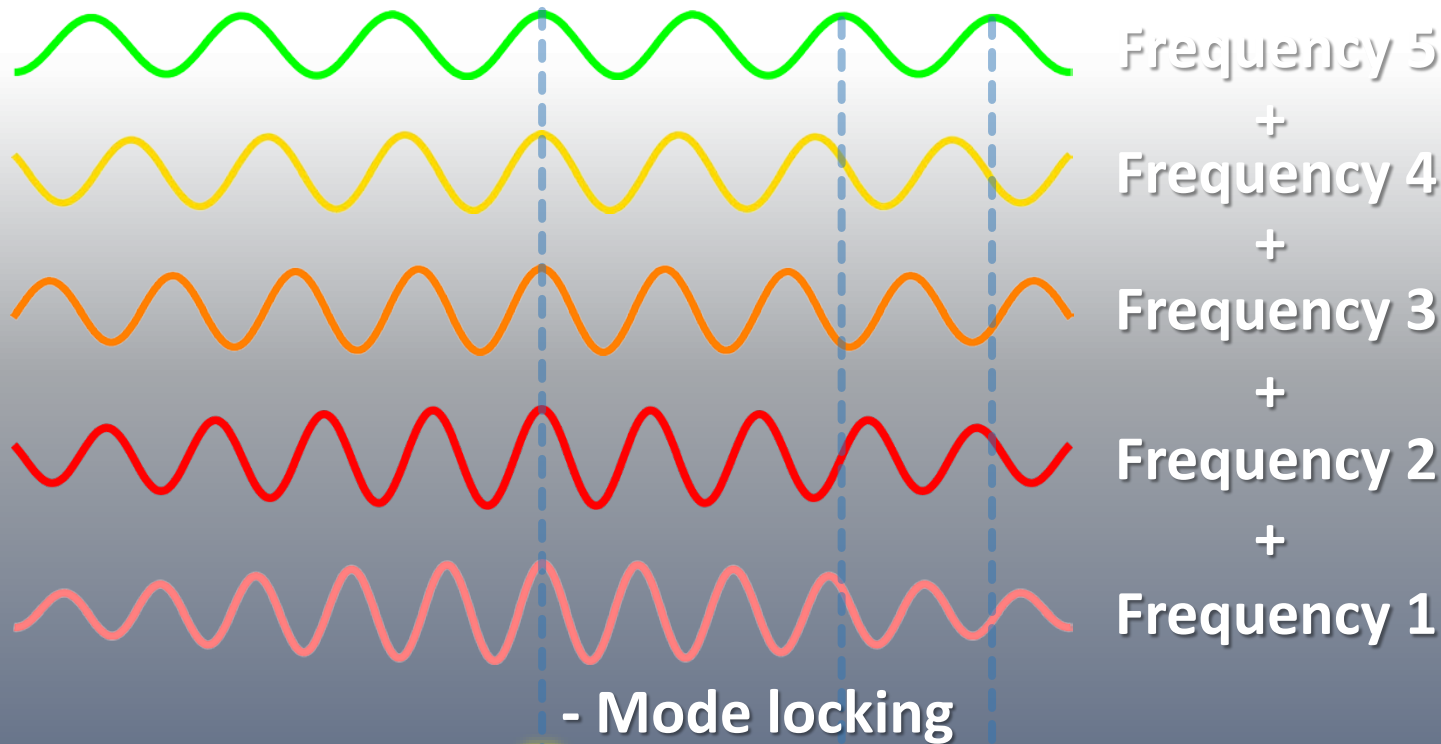
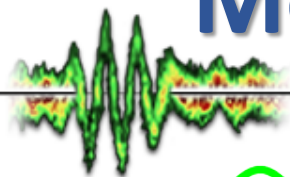
Where does the energy end up, and when?

REVERSIBILITY:

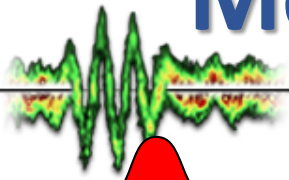
How quickly can a system return back into the ground state?



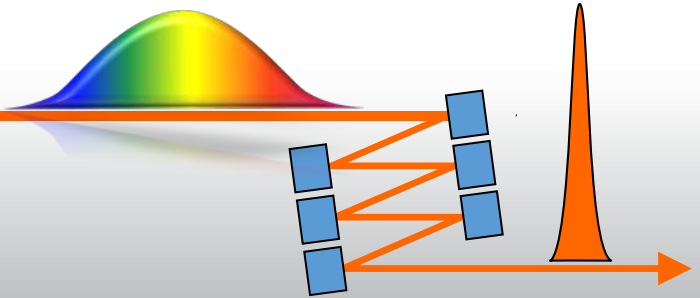
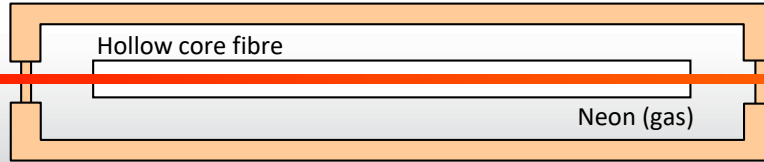
Mode locked lasers as strobe light



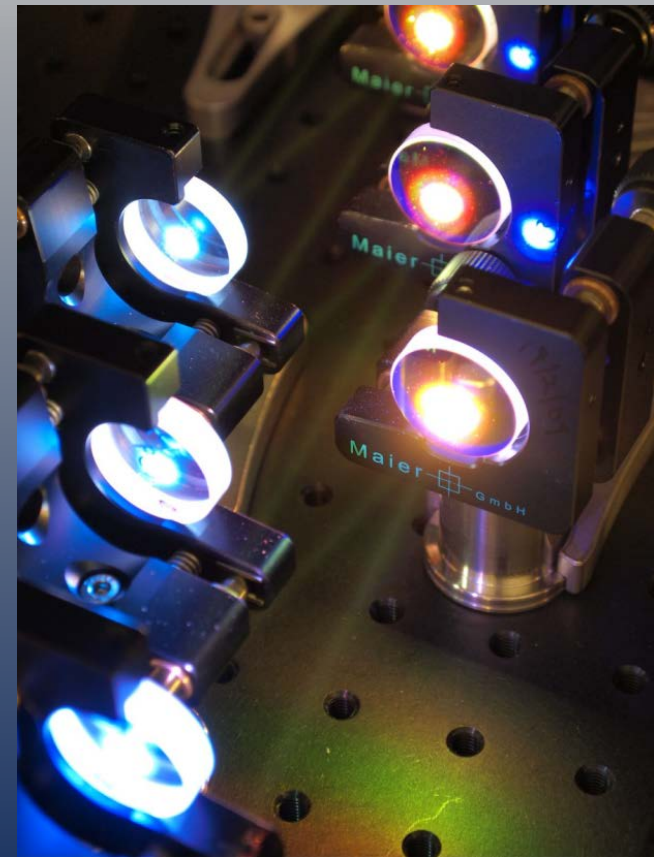
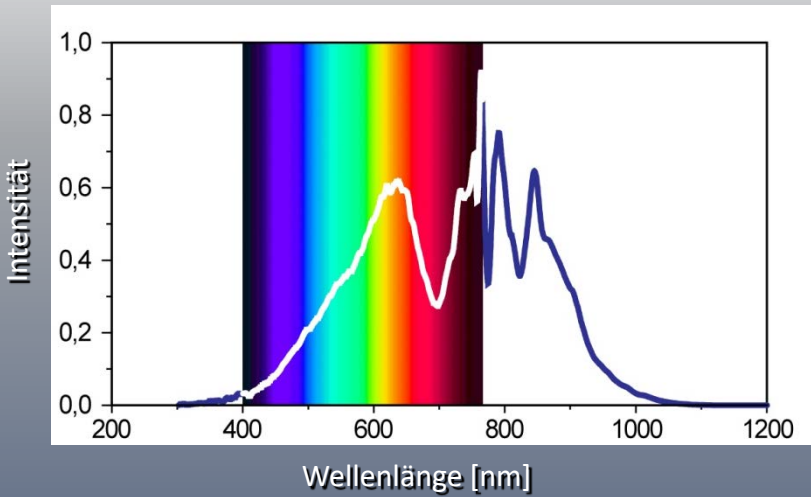
Mode locked lasers as strobe light



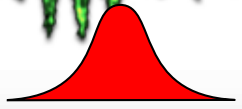
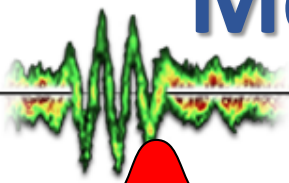
mJ / 25 fs



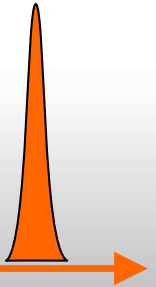
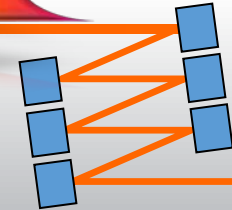
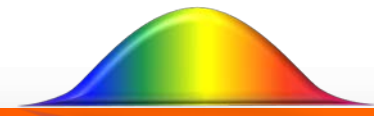
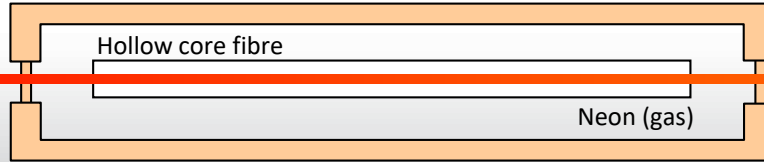
$\frac{1}{2}$ mJ / 4 fs



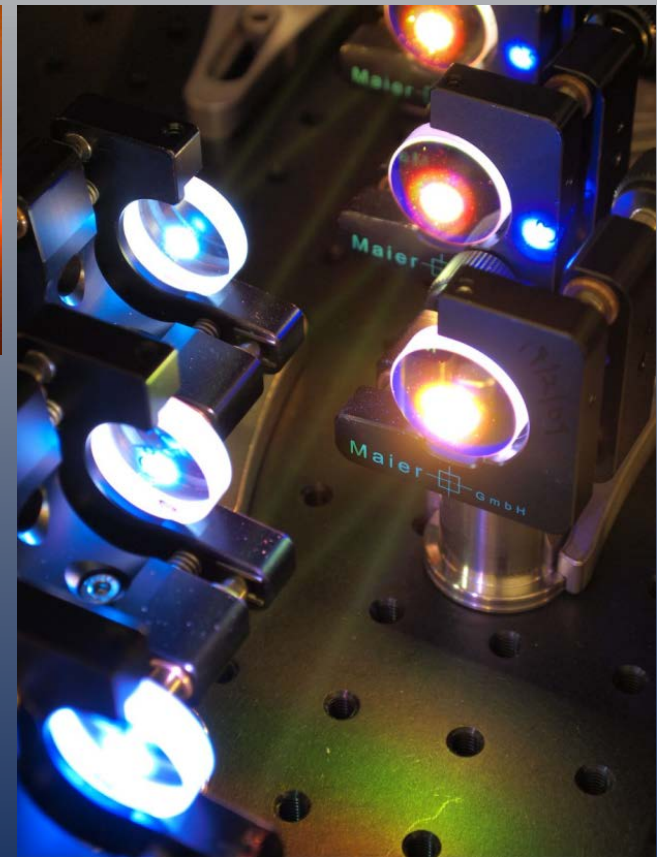
Mode locked lasers as strobe light



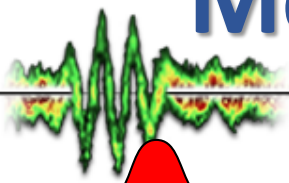
mJ / 25 fs



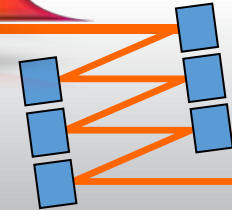
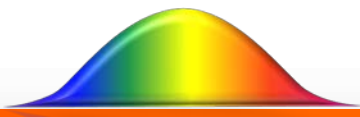
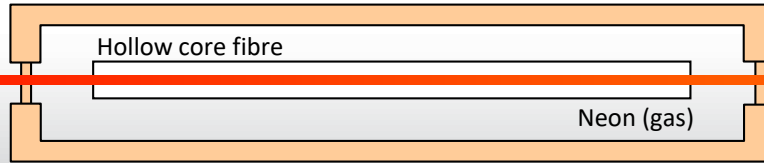
1/2 mJ / 4 fs



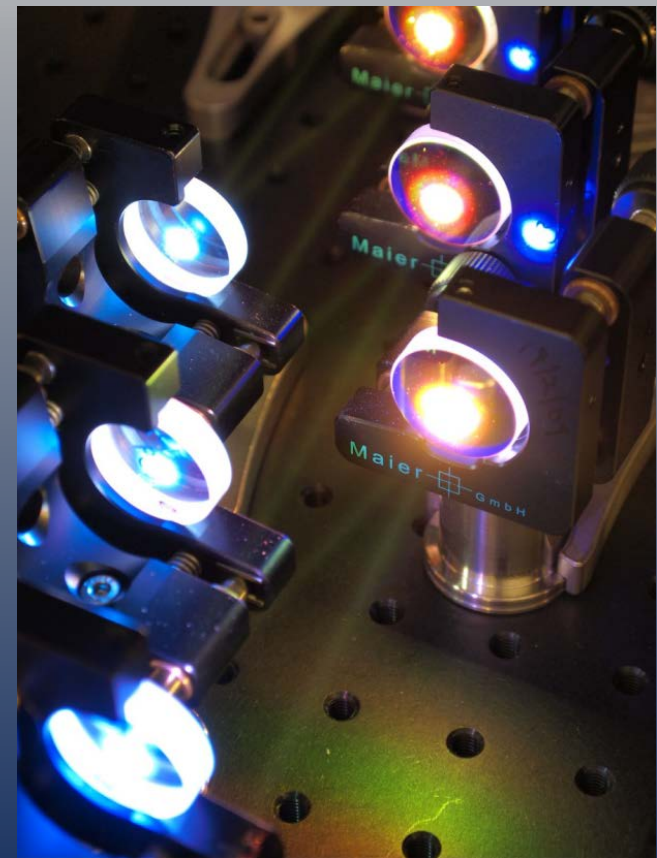
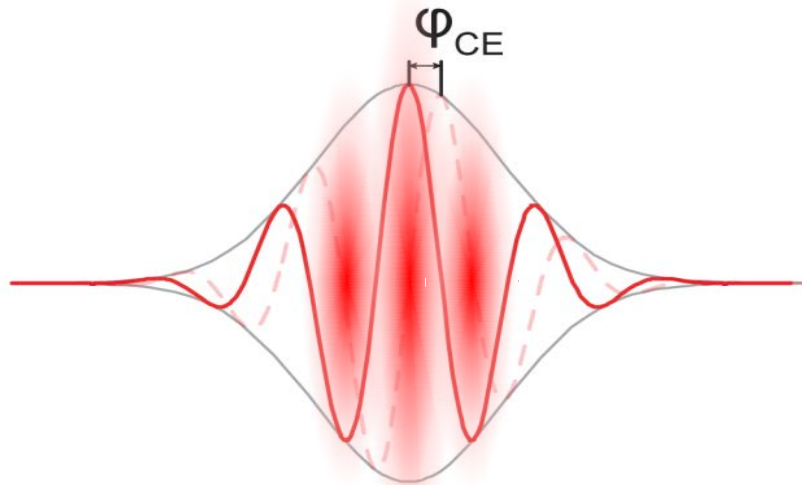
Mode locked lasers as strobe light



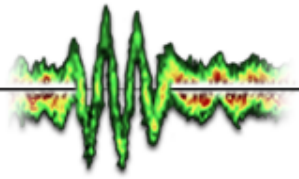
mJ / 25 fs



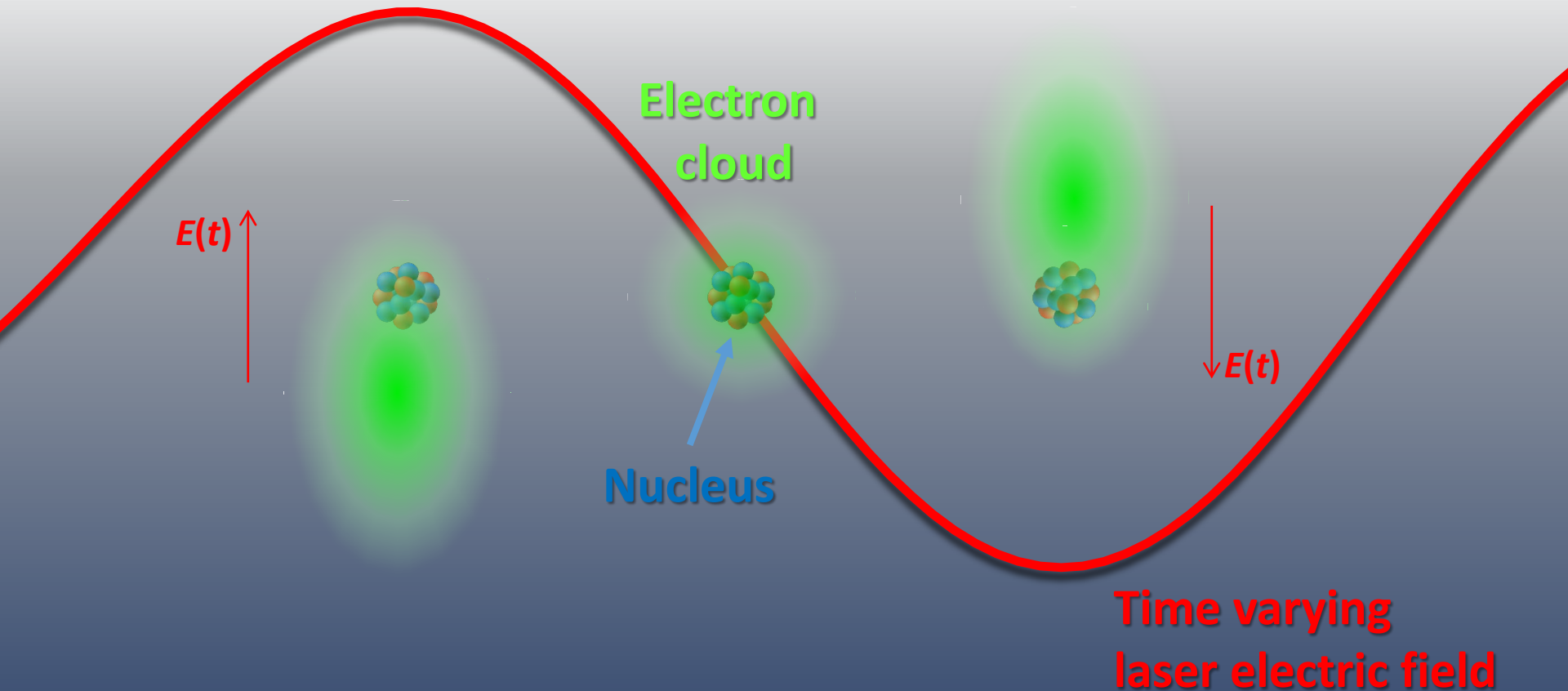
1/2 mJ / 4 fs



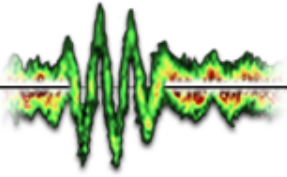
Attosecond pulses



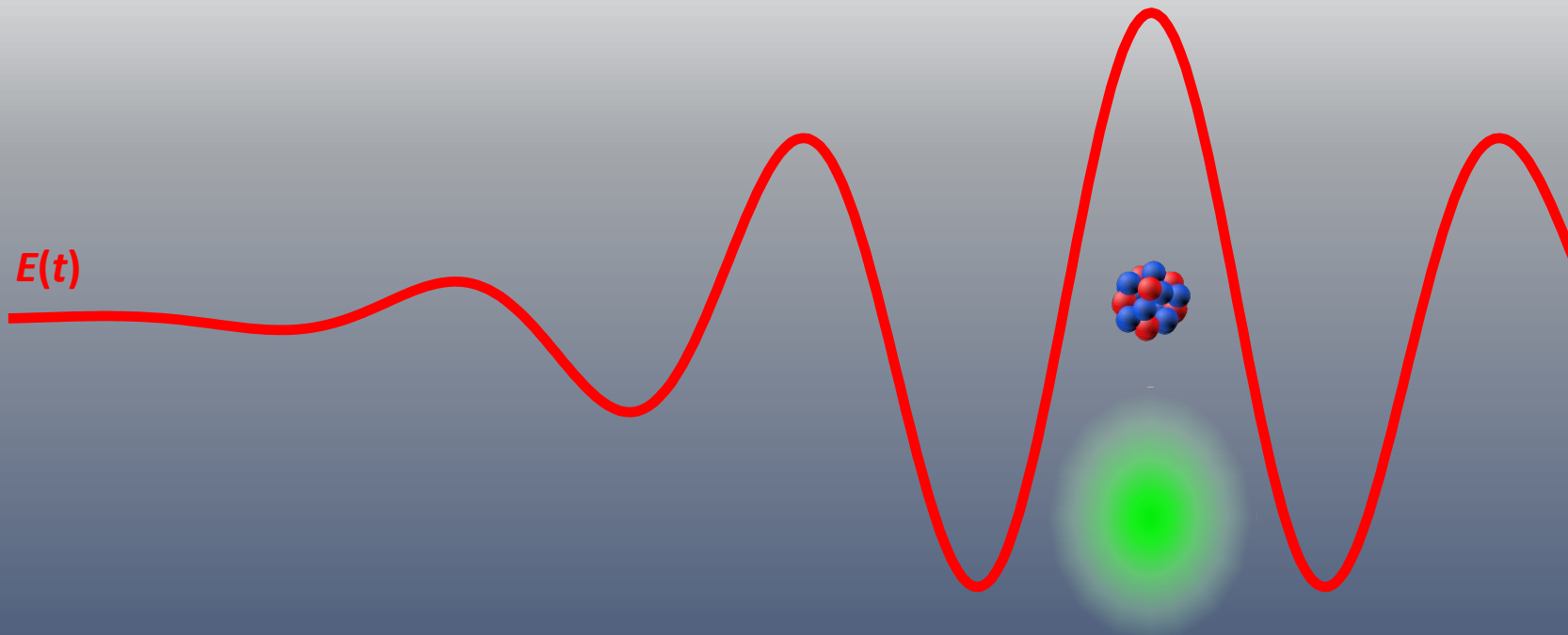
Nonlinear optics via photo ionization



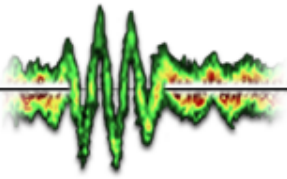
Attosecond pulses



Nonlinear optics via photo ionization

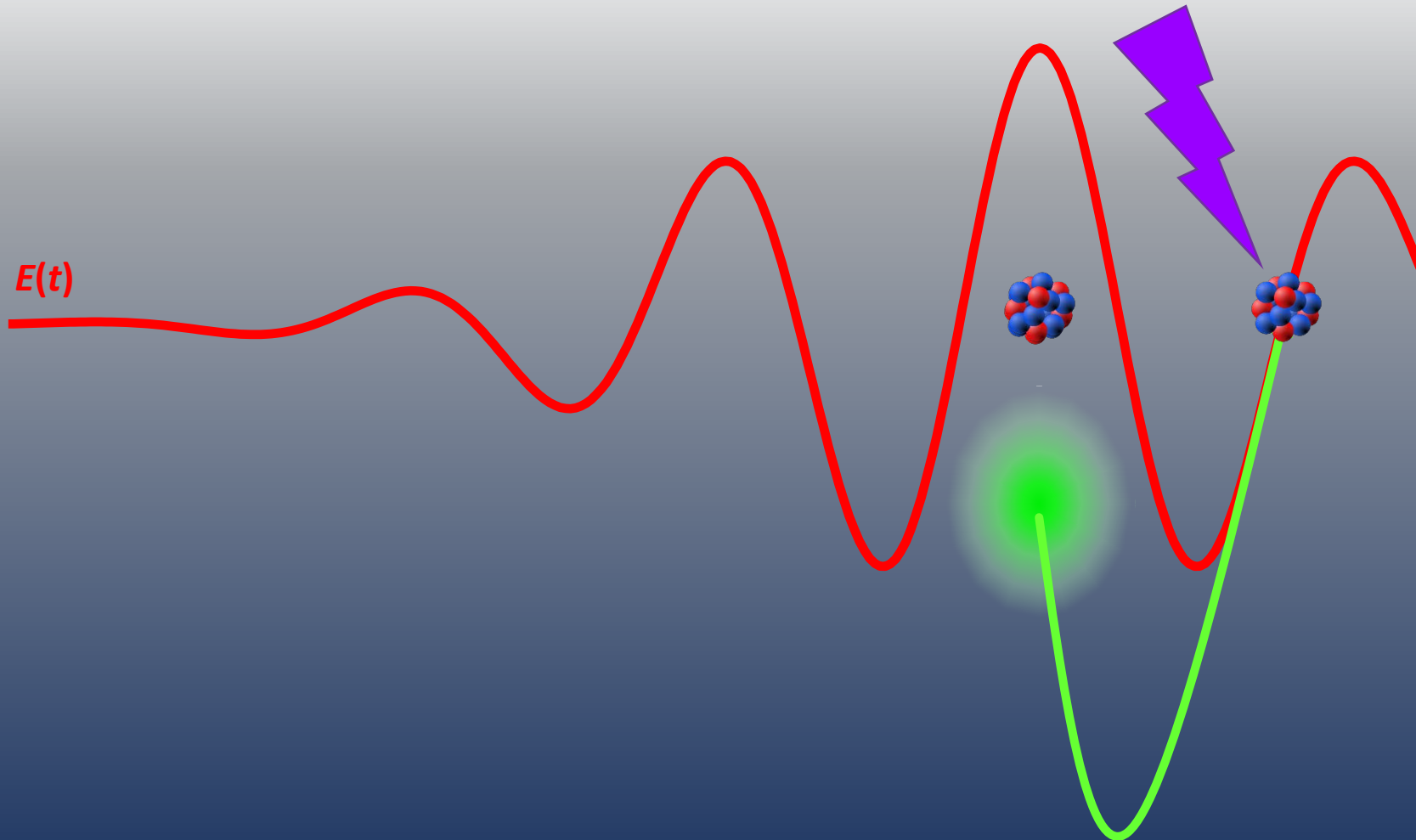


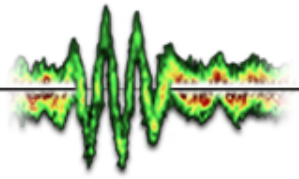
Attosecond pulses



Nonlinear optics via photo ionization

XUV
photon

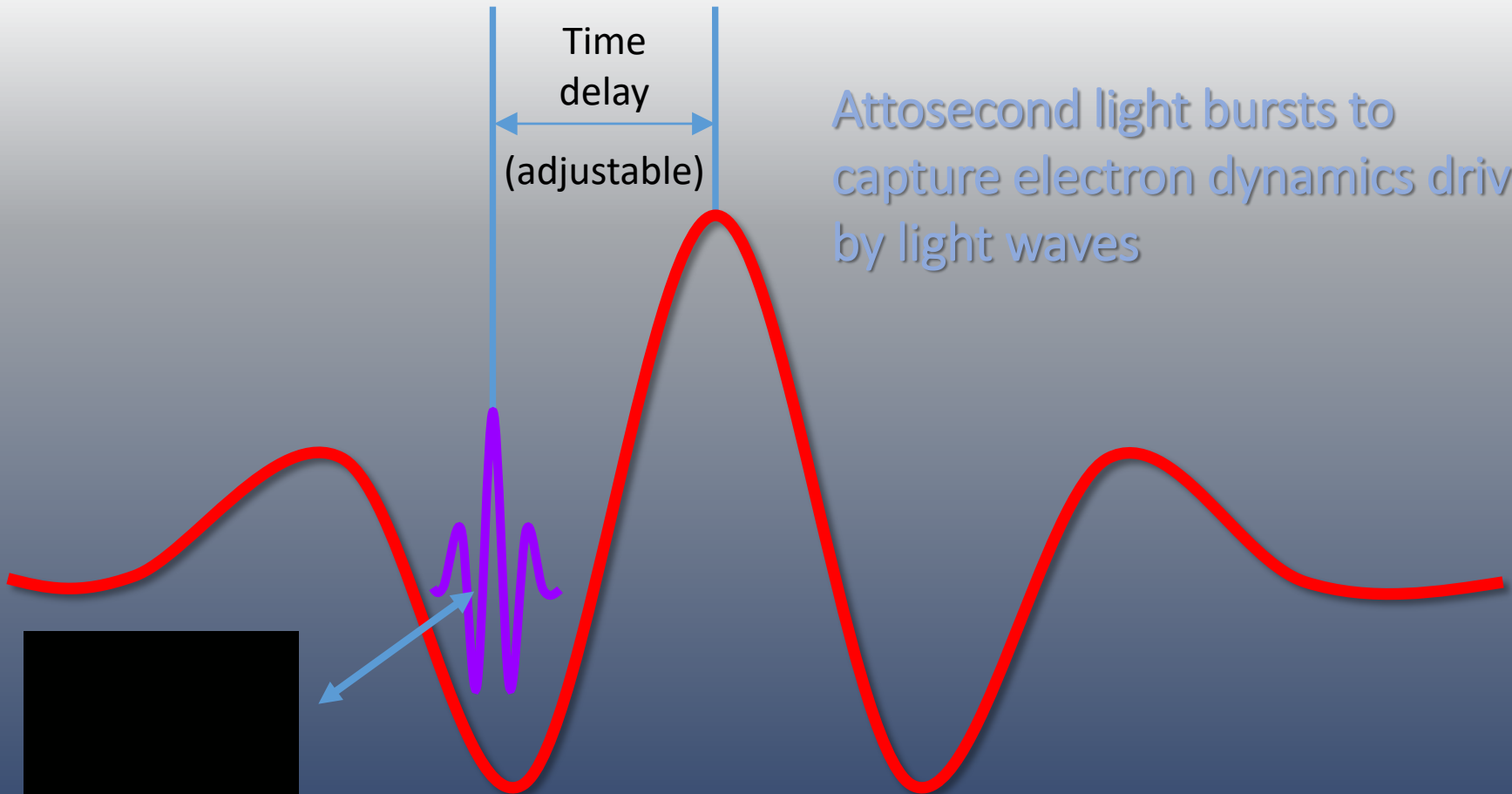




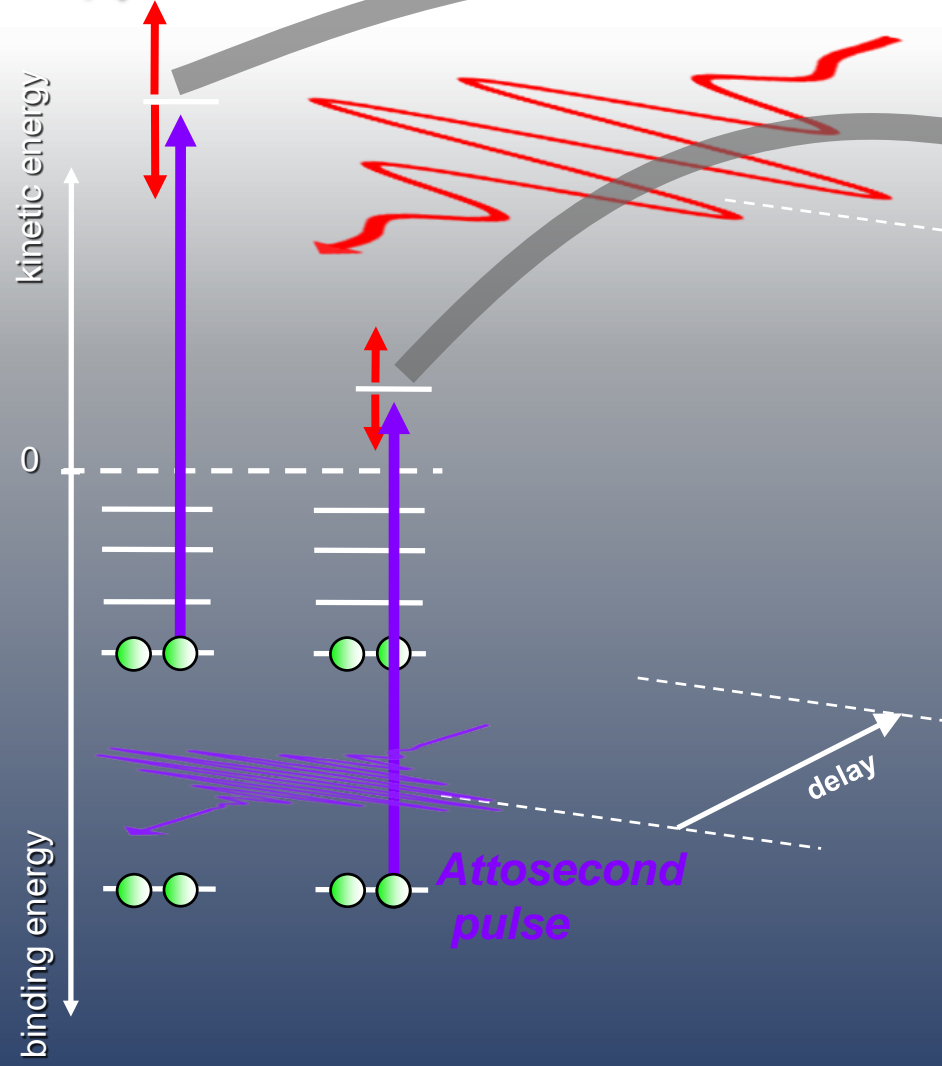
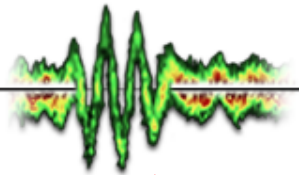
Attosecond pulses

Nonlinear optics via photo ionization

Attosecond light bursts to capture electron dynamics driven by light waves

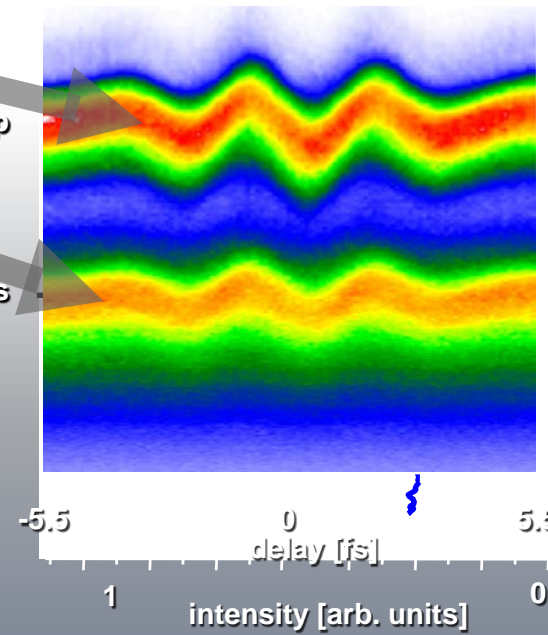


Attosecond streak camera



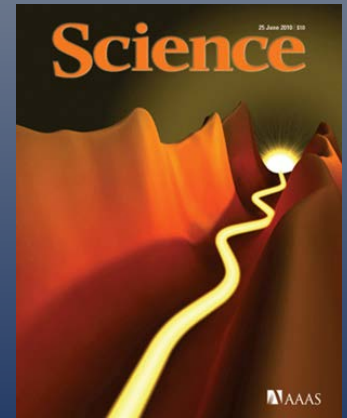
2p

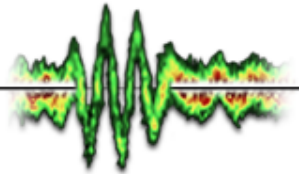
2s



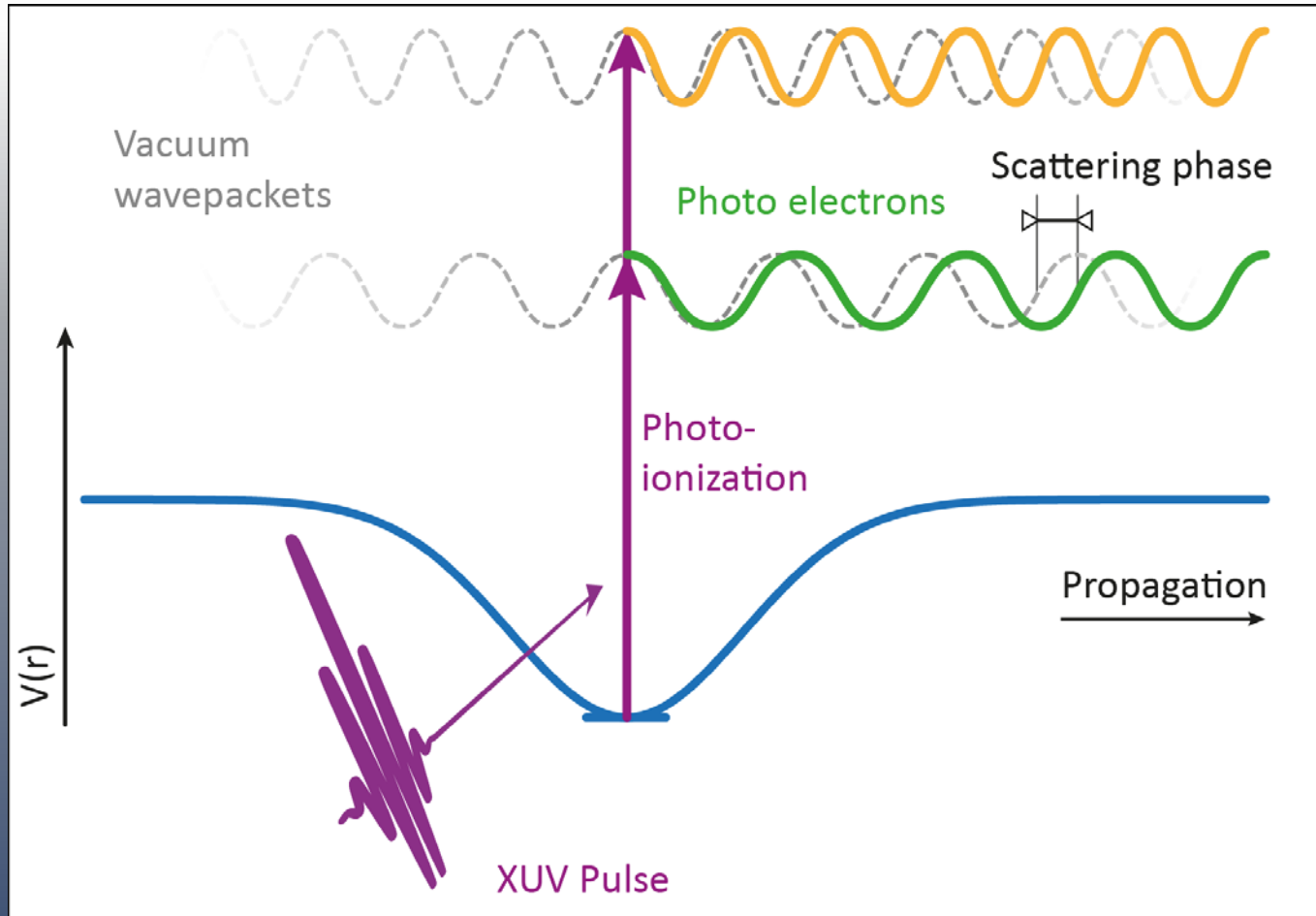
intensity [arb. units]

Observed Delay:
~ 20 Attoseconds





Delay in Photoemission?

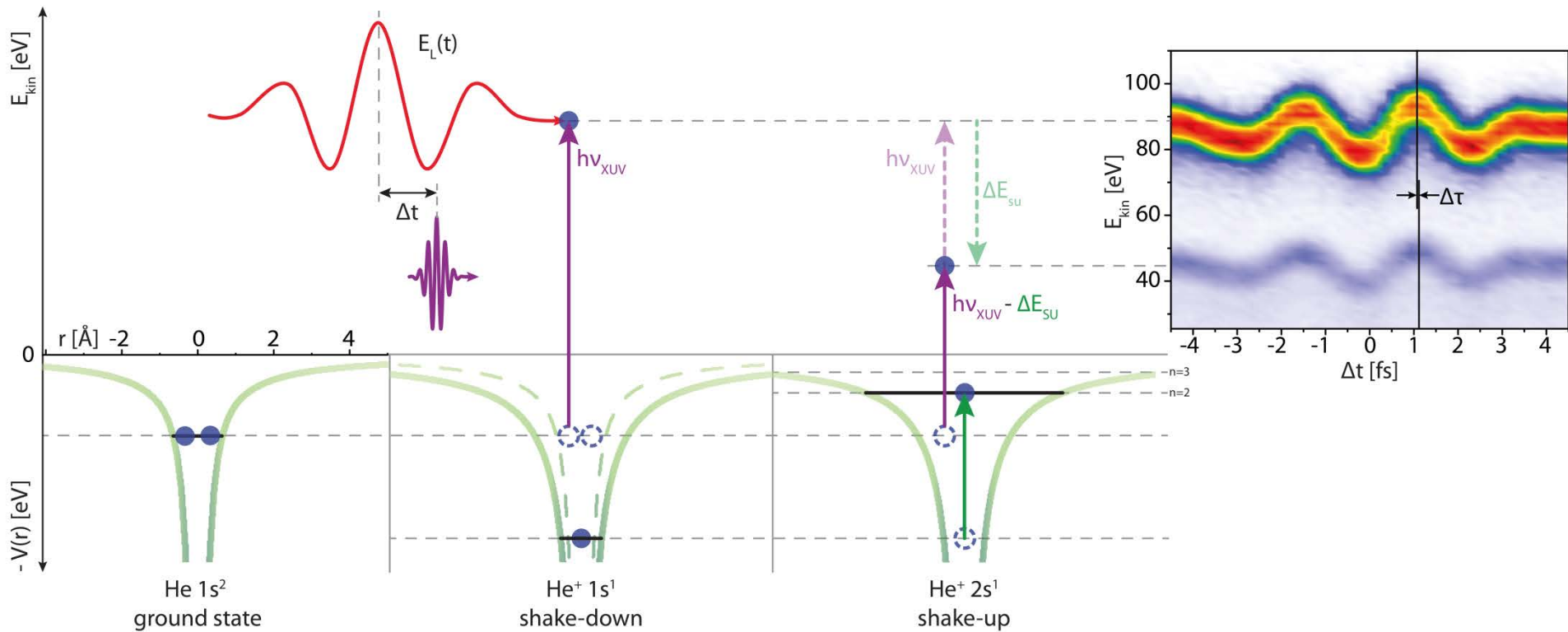
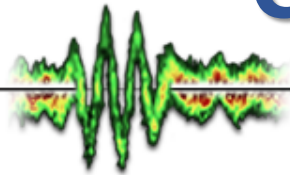


$$\Delta\tau_{2s-2p}$$

experiment: 21 as
theory: 9 - 16 as

Residual sub-10 as discrepancy
-> **electronic correlations ?**

Clocking electronic correlations



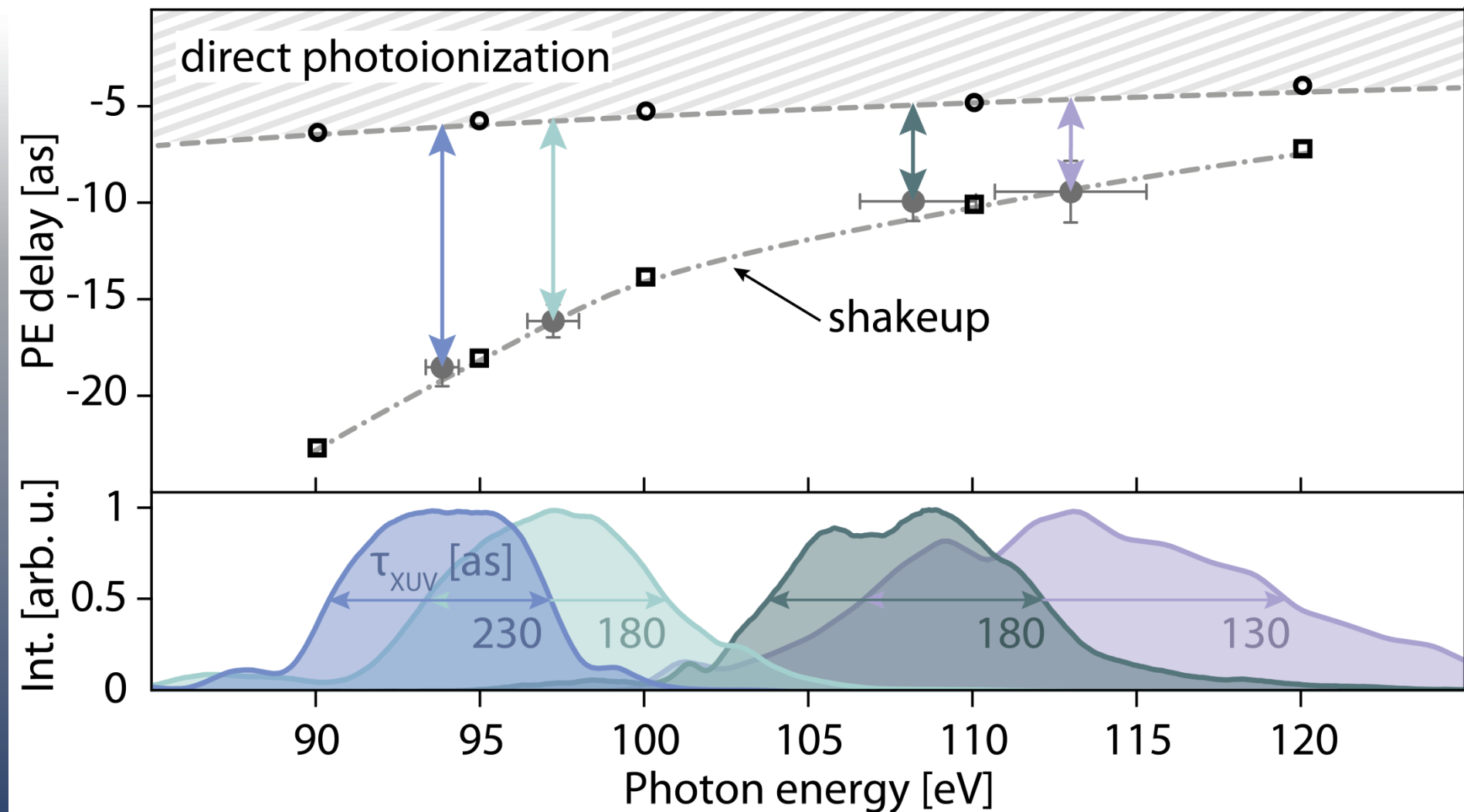
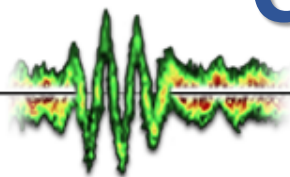
Helium
ground state

Photoelectric effect

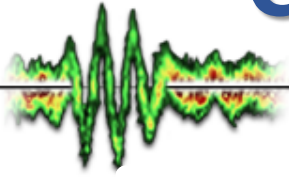
Photoelectric effect
& electronic excitation

Observe the influence of
electronic correlations ?

Clocking electronic correlations

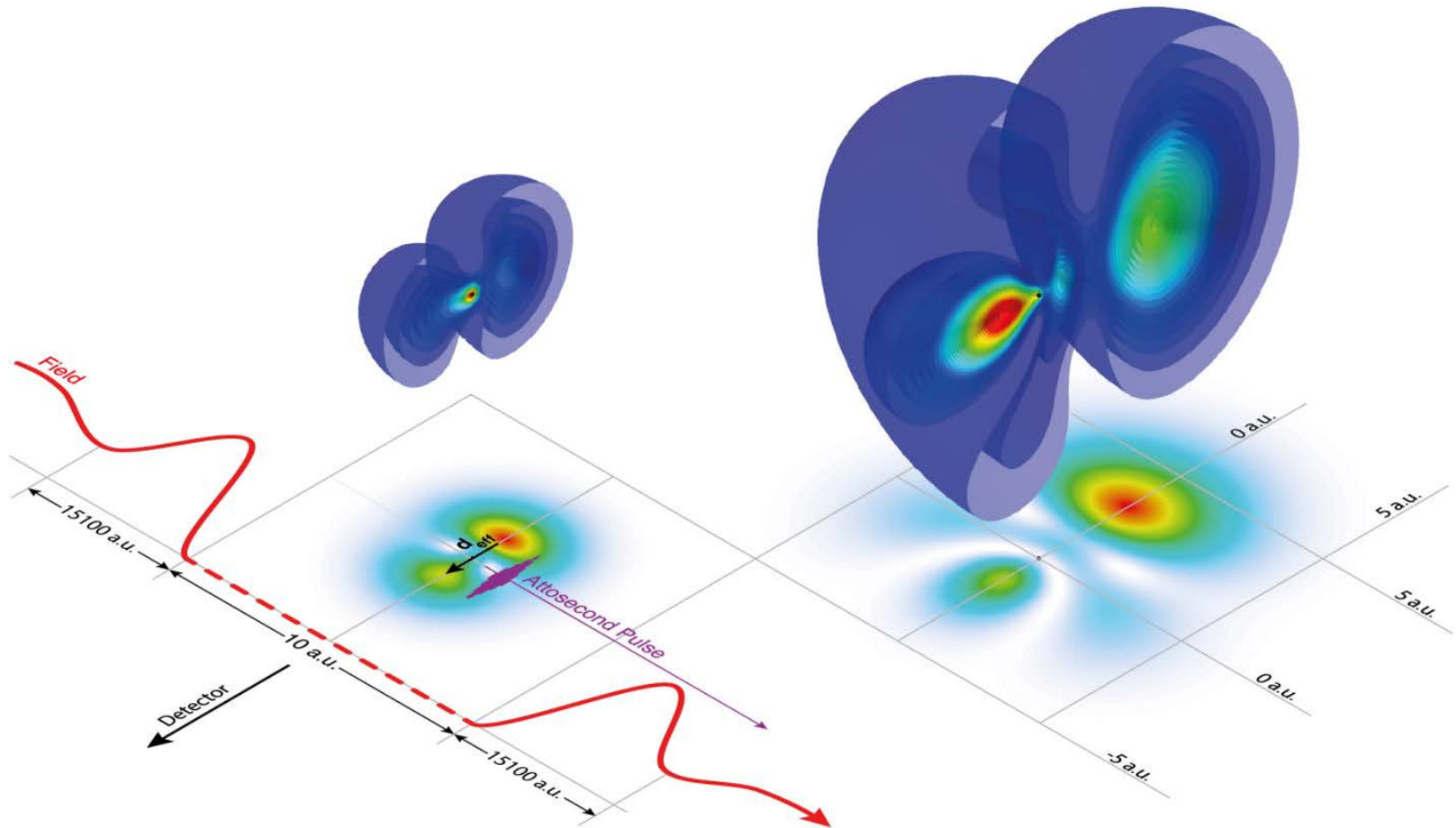


Clocking electronic correlations

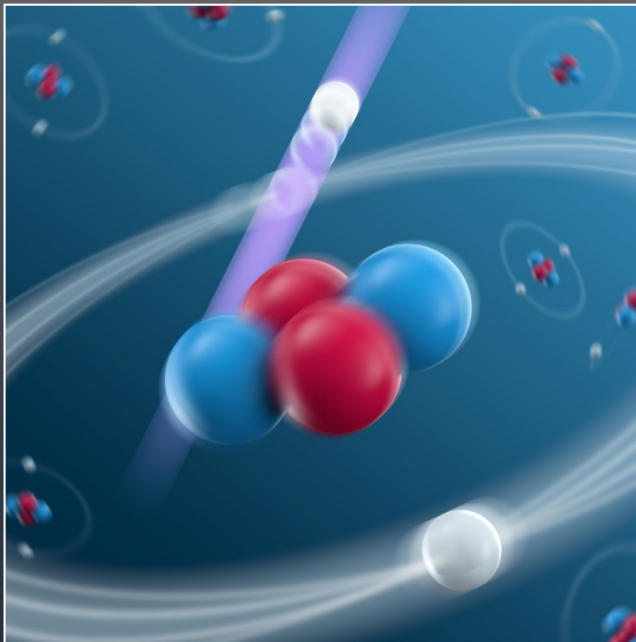
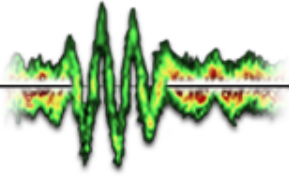


$n=2$

$n=3$



Electron dynamics in atoms



The photo-electric effect¹

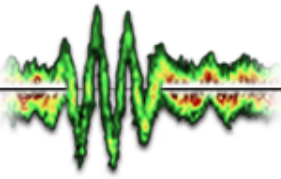
in Helium exposed to strong electric fields oscillating at optical frequencies

takes 5.9 attoseconds

this time-shift depends on the excitation photon energy and photo-electron and – ion form a quantum mechanically entangled pair

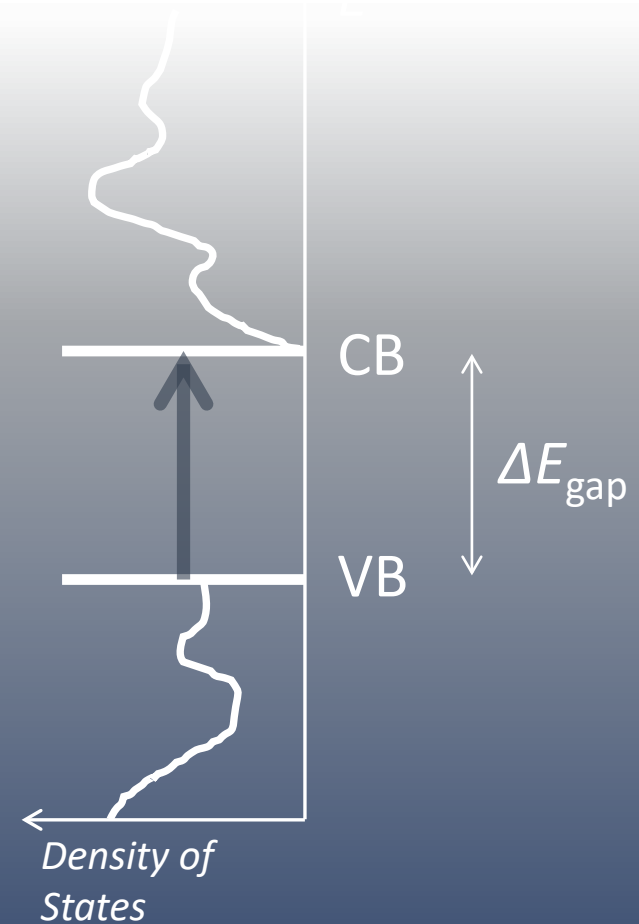
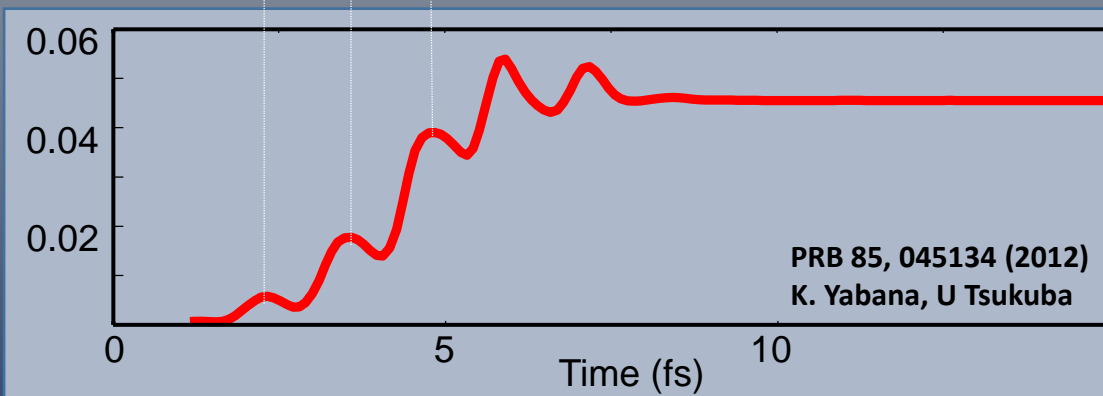
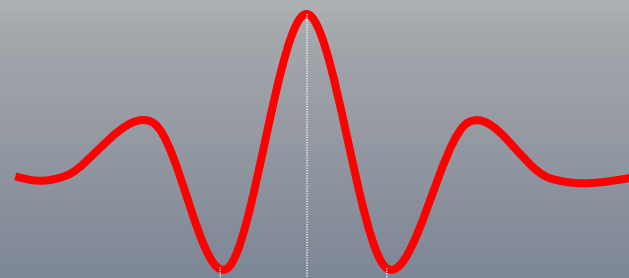
¹ A. Einstein, Über einen die Erzeugung und Verwandlung des Lichtes betreffenden heuristischen Gesichtspunkt. Ann. Phys. 322, 132–148 (1905).

Charge dynamics in solids



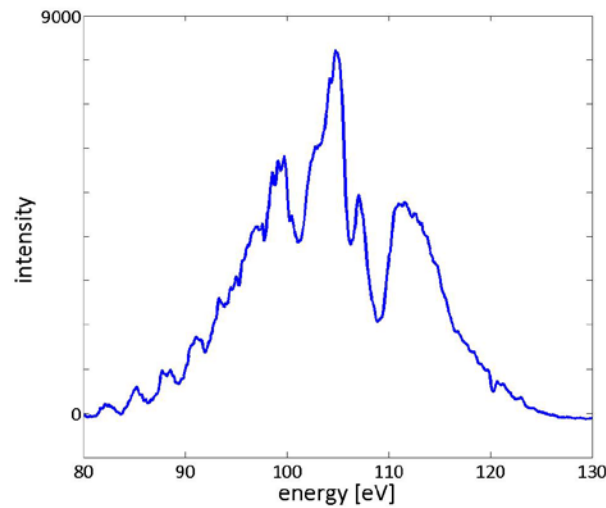
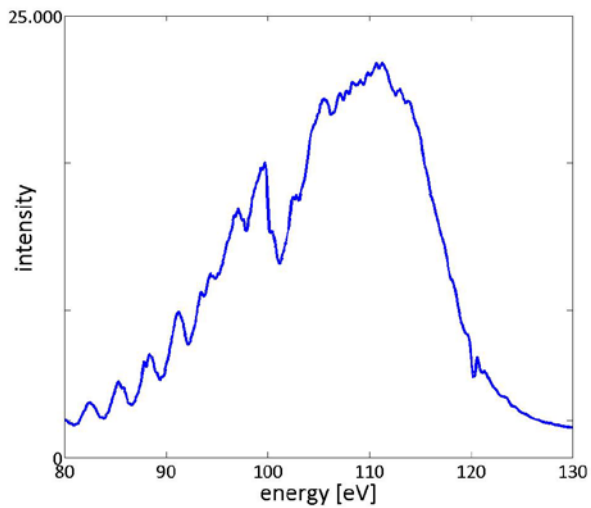
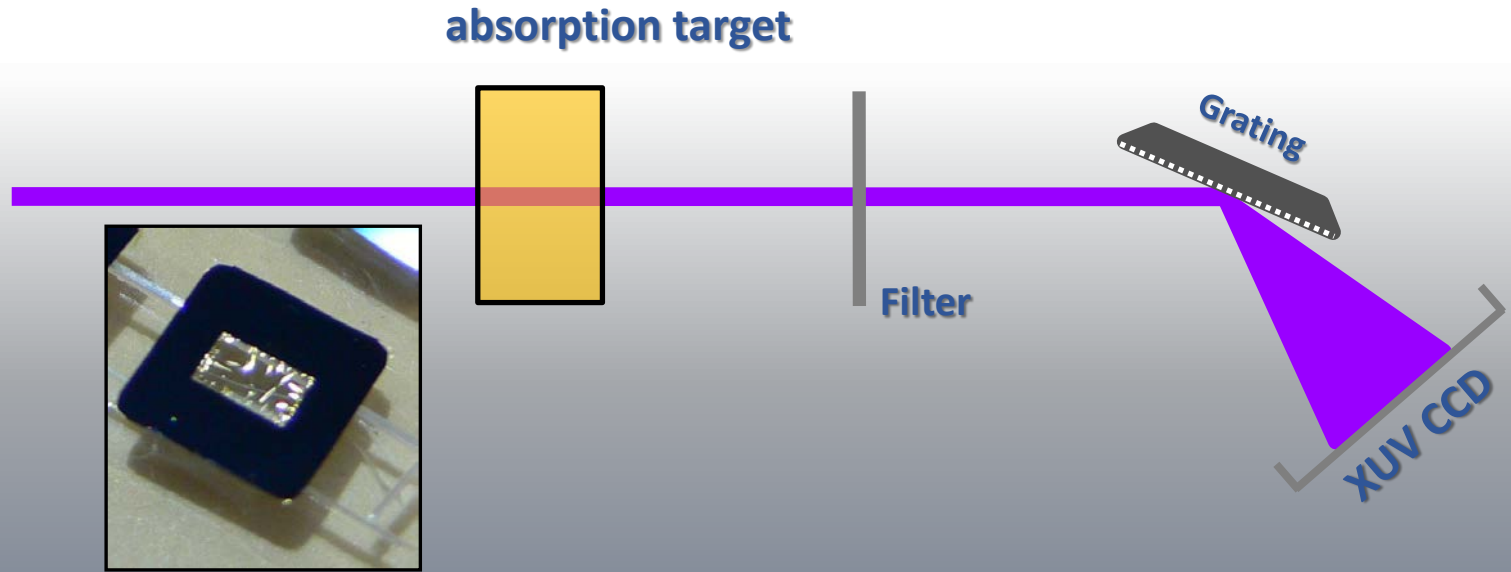
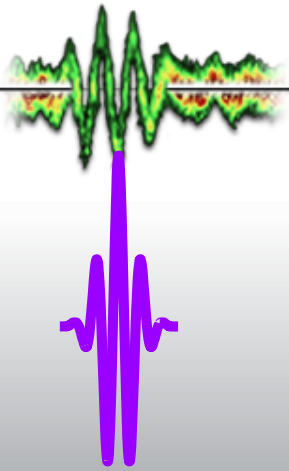
Semiconductor exposed to light can absorb energy by creating electron-hole pairs

Dispersion relation & Density of states

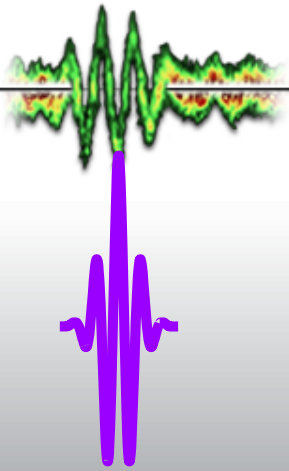


attosecond XUV tools to quantify charge transfer dynamics?

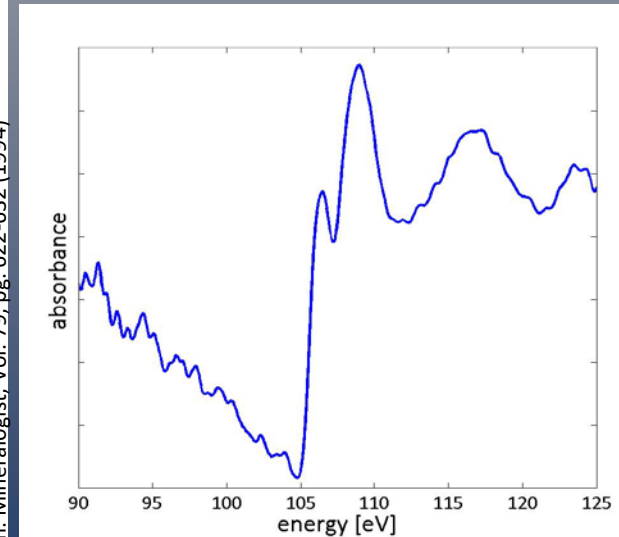
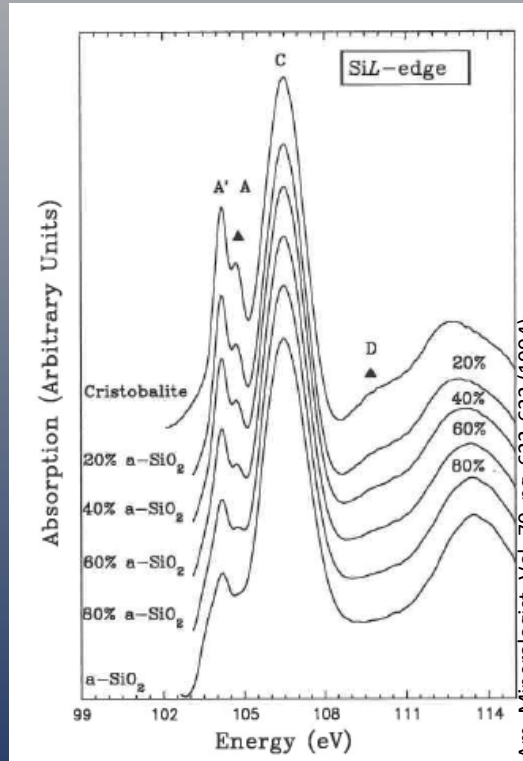
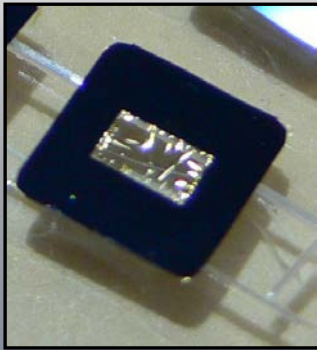
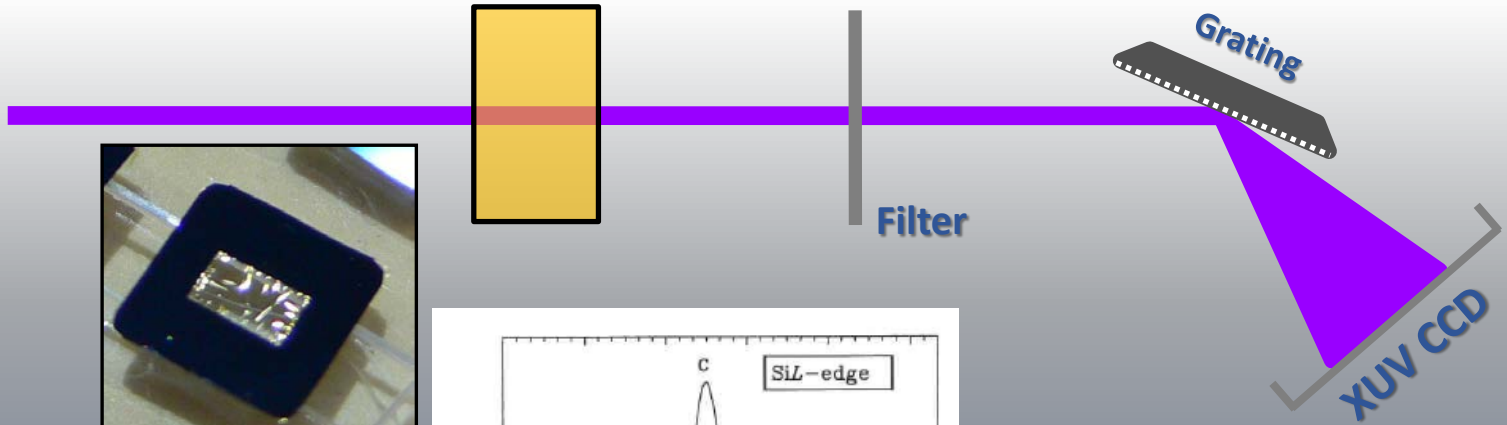
XUV - Transient absorption



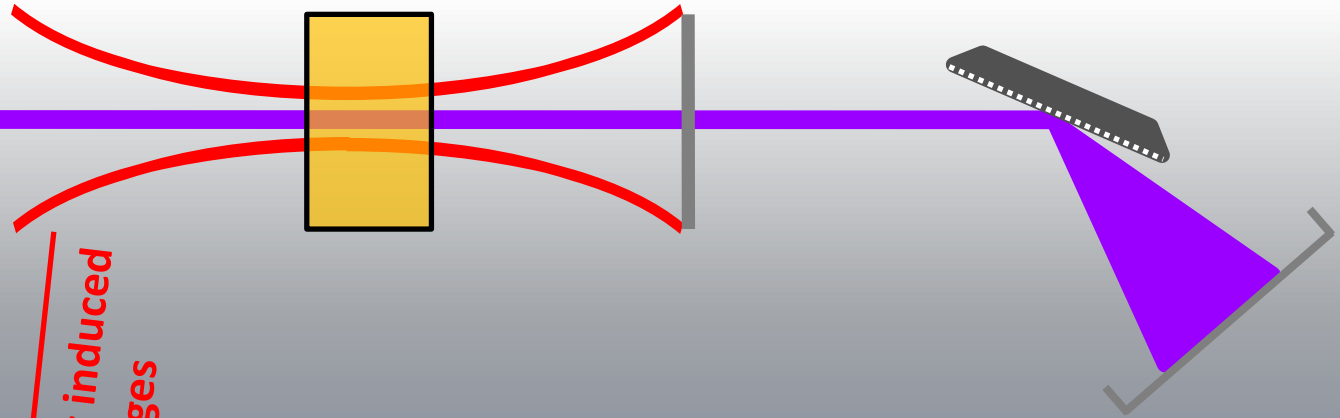
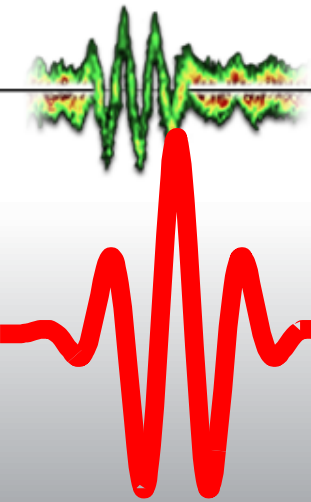
XUV - Transient absorption



absorption target

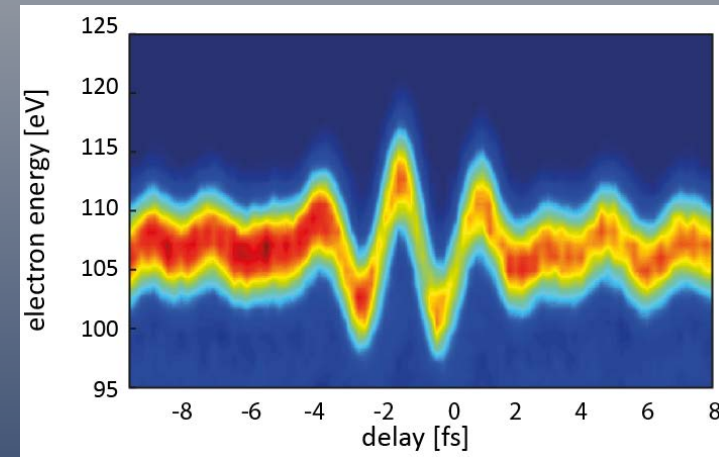
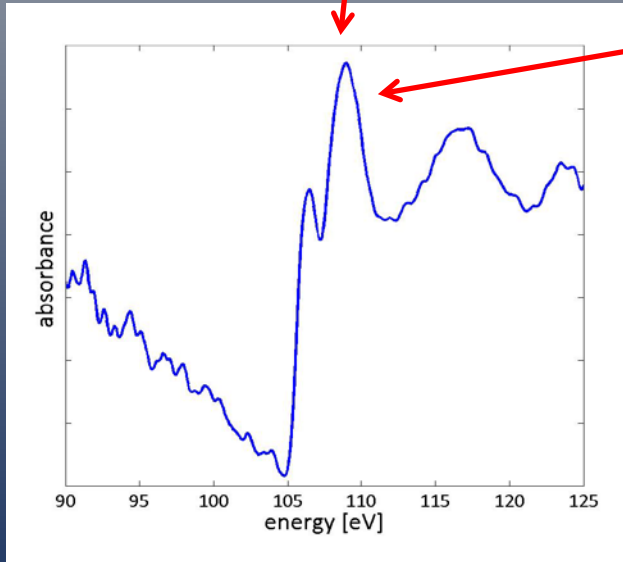


XUV - Transient absorption



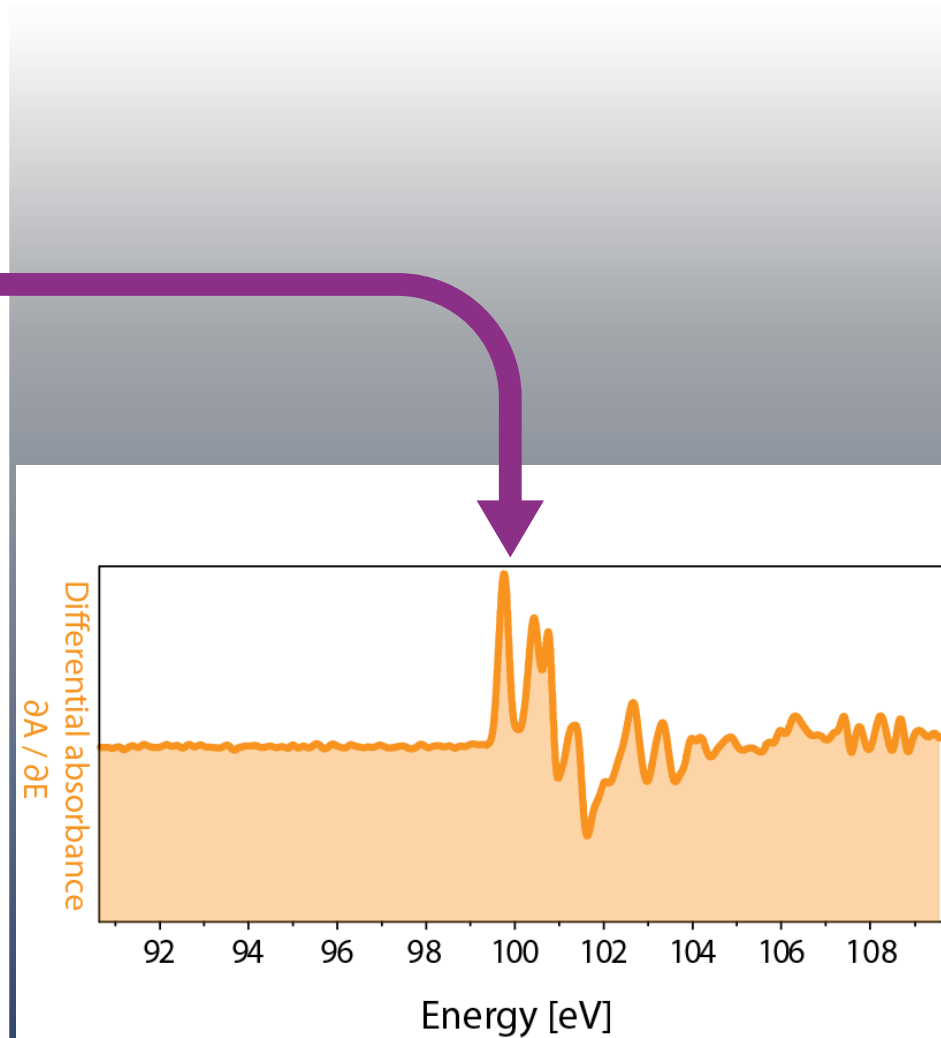
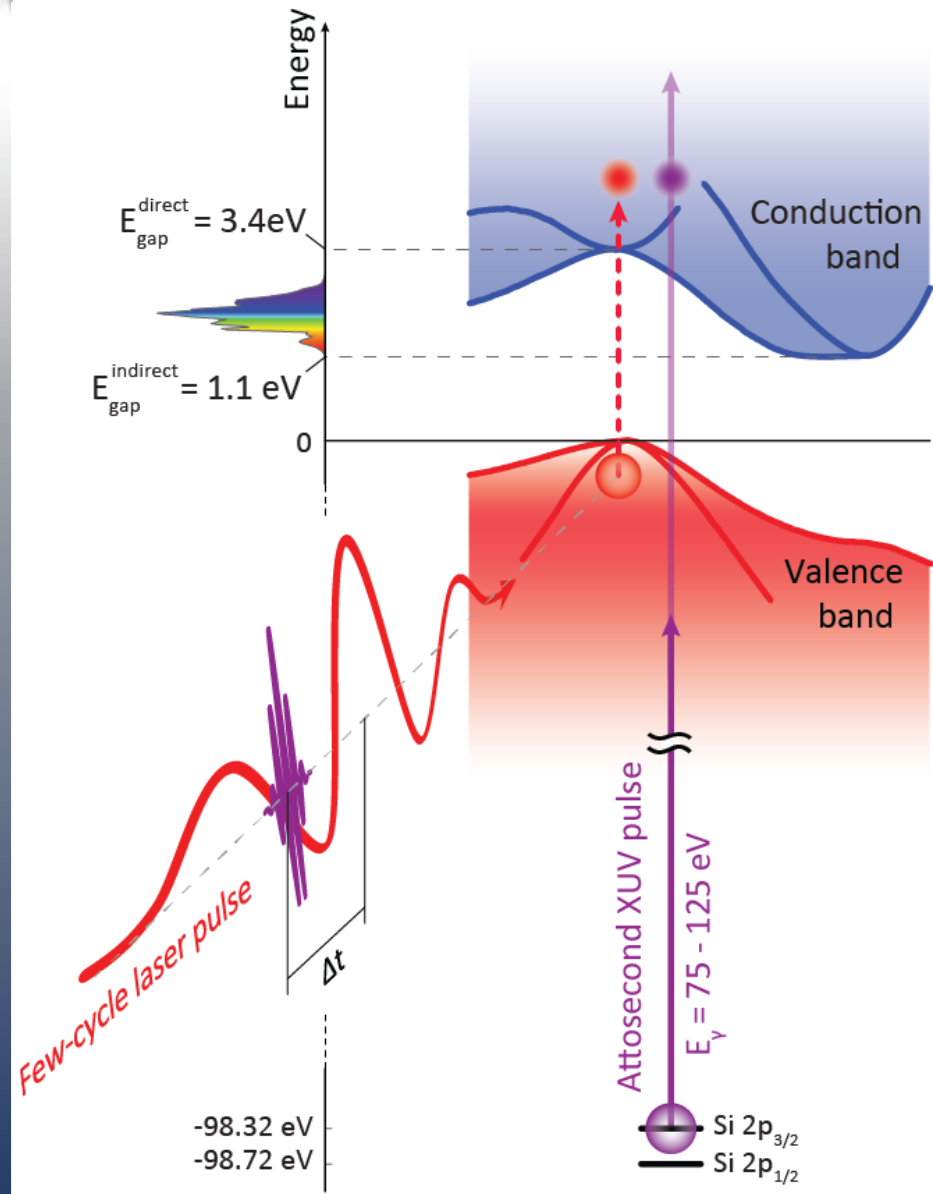
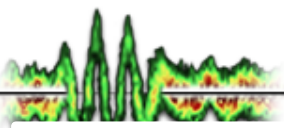
Light induced changes

Electric field sampling

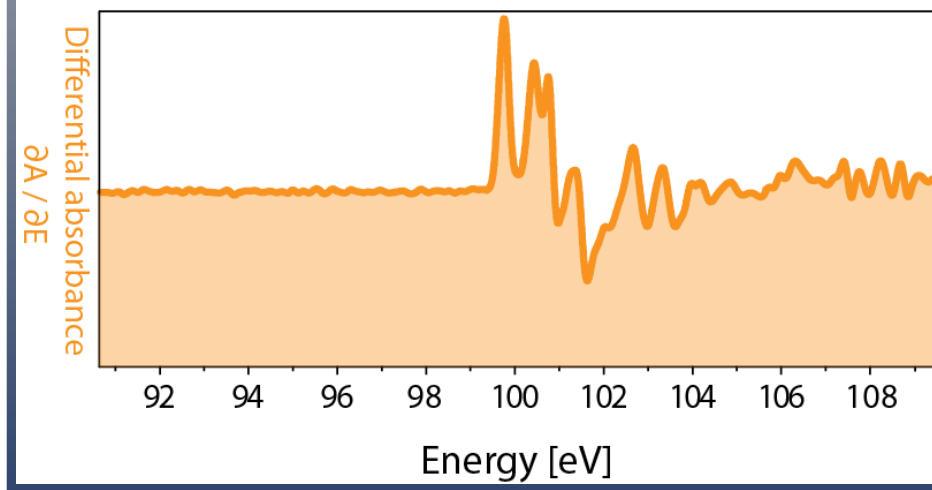
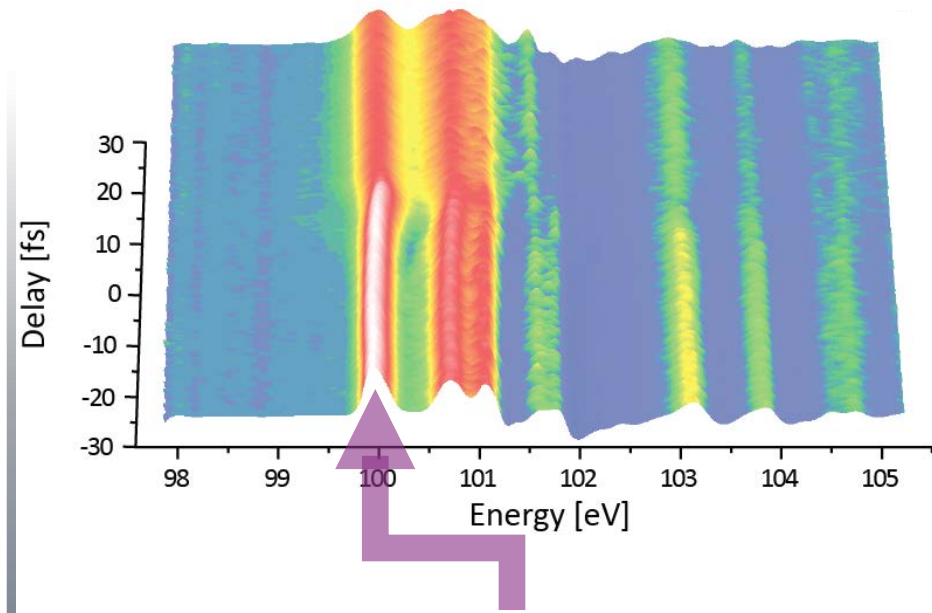
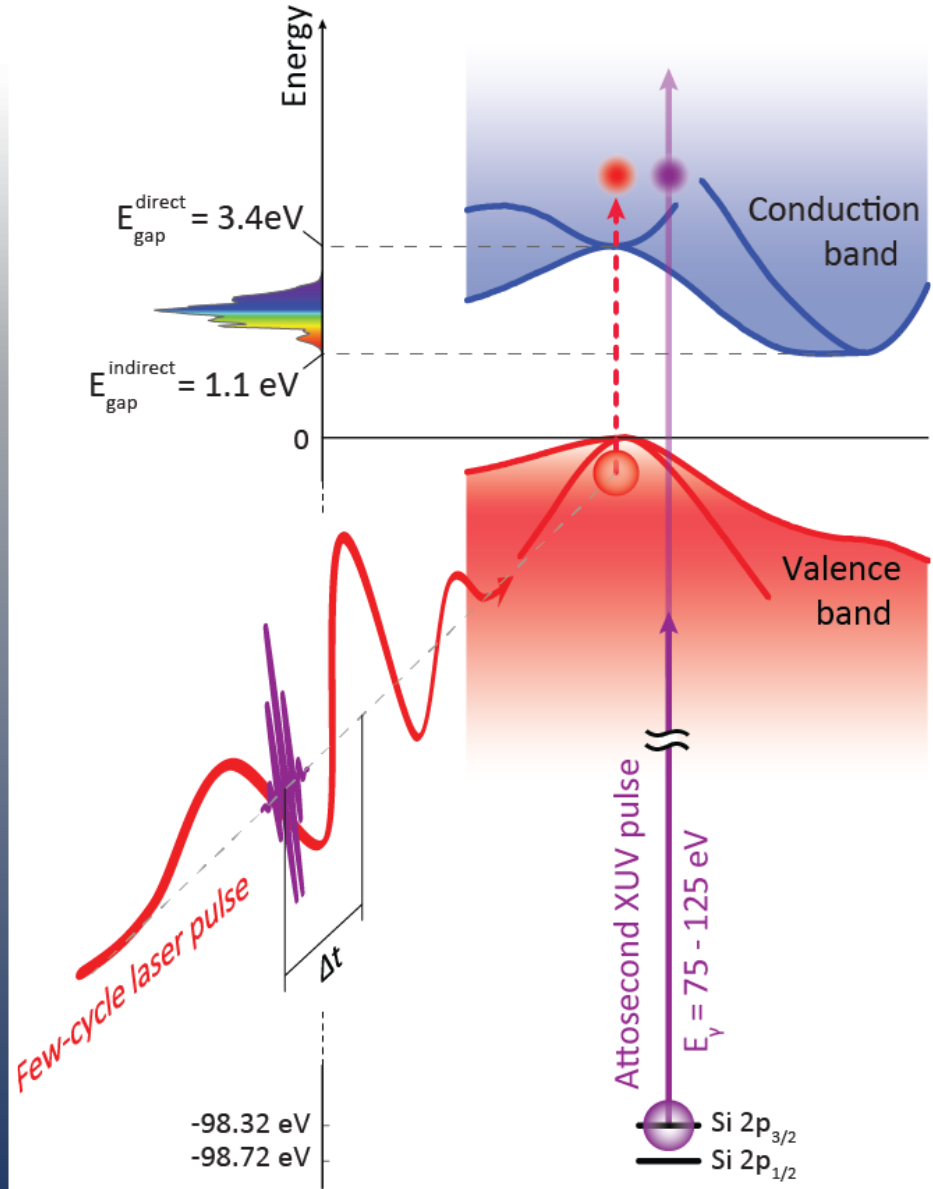
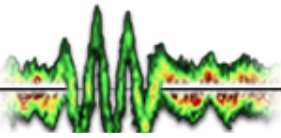


→ Time resolve optical excitations

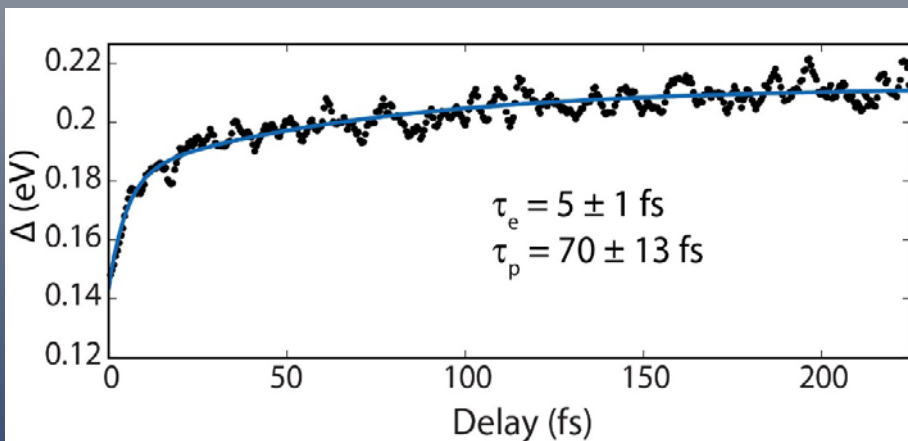
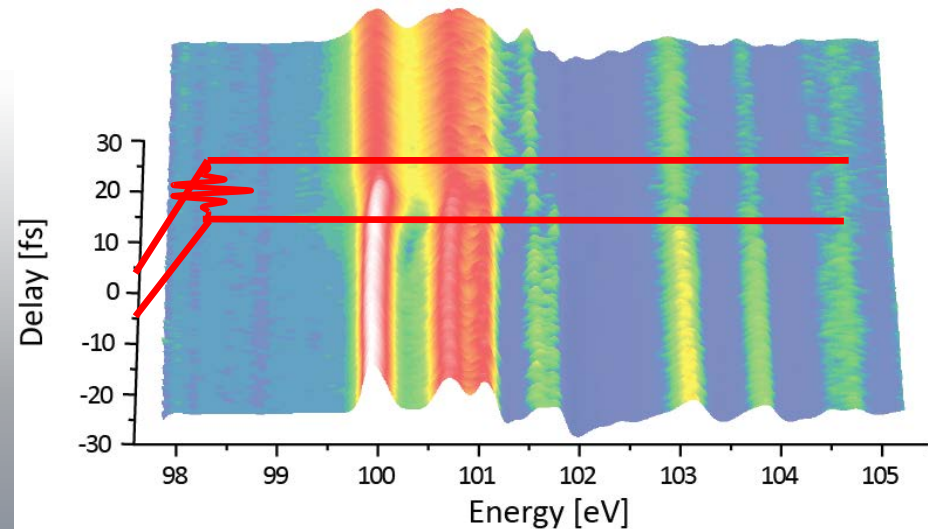
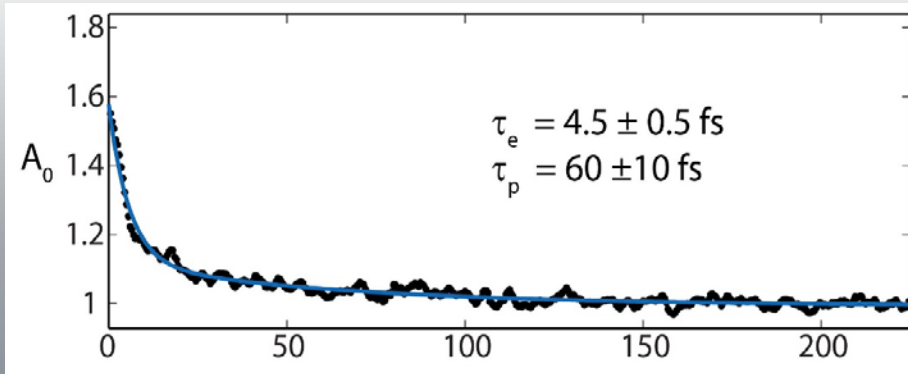
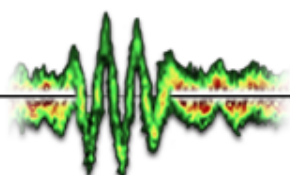
Band-gap dynamics: Silicon



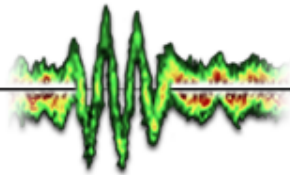
Band-gap dynamics: Silicon



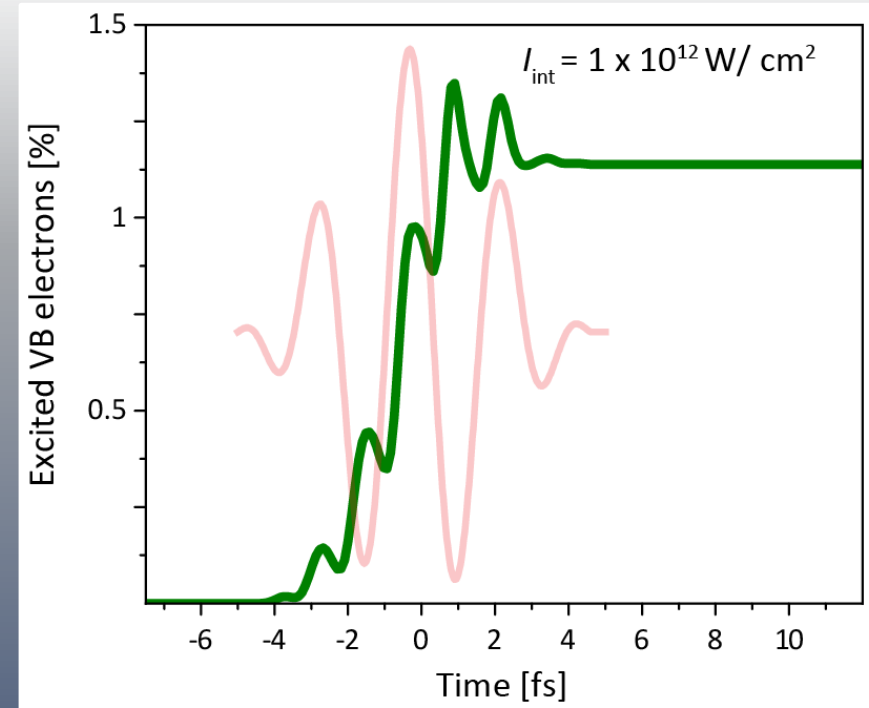
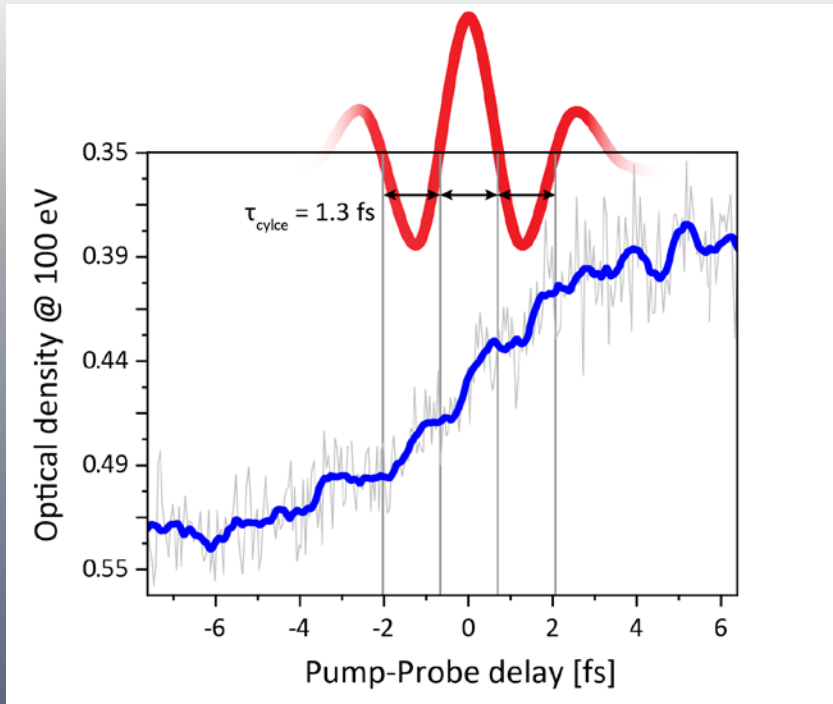
Ultrafast band-gap collapse

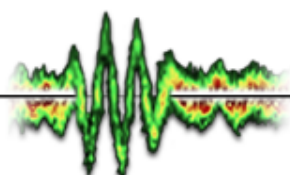


photodoping of silicon induces
instantaneous (electronic) band gap narrowing
&
the lattice follows with a time constant of the
fastest optical phonon (64 fs)

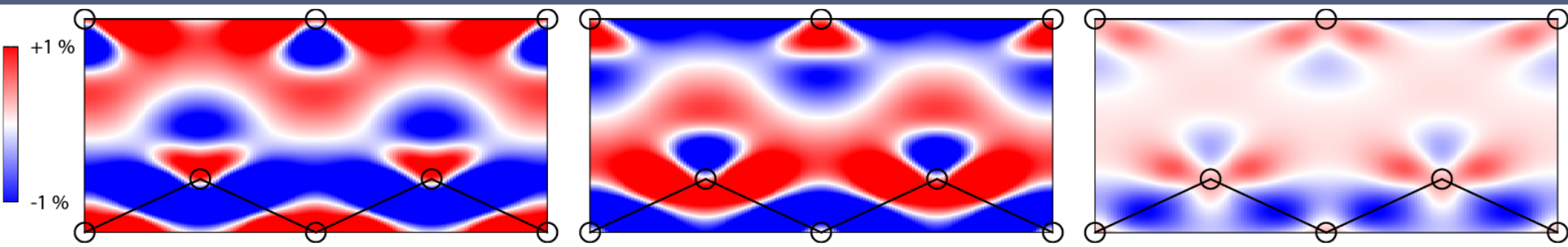
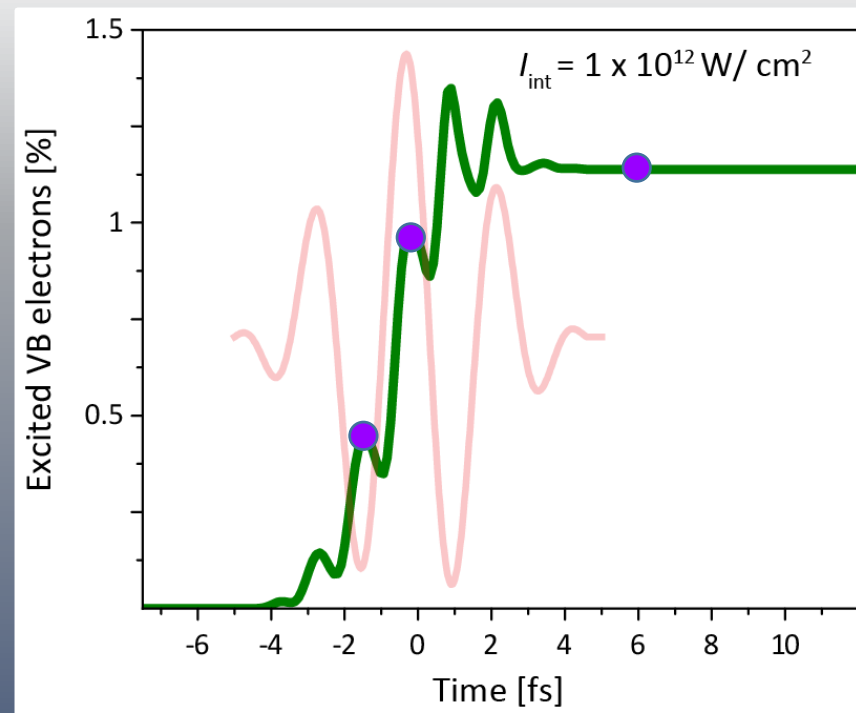
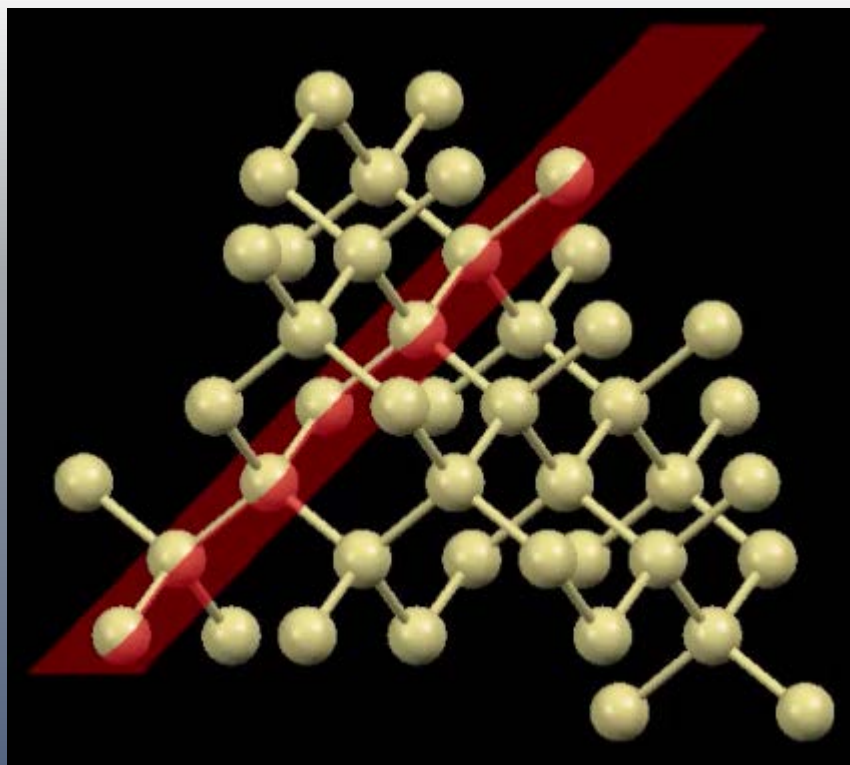


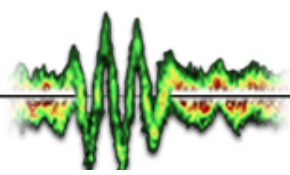
Tunneling into the CB



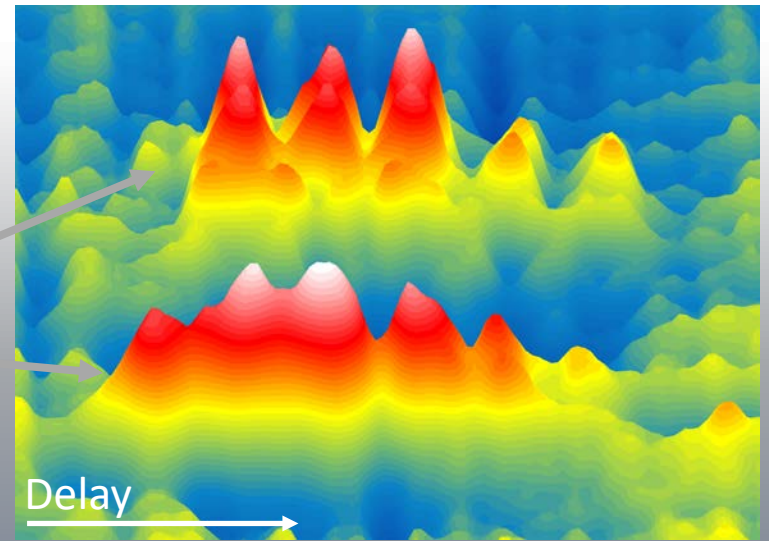
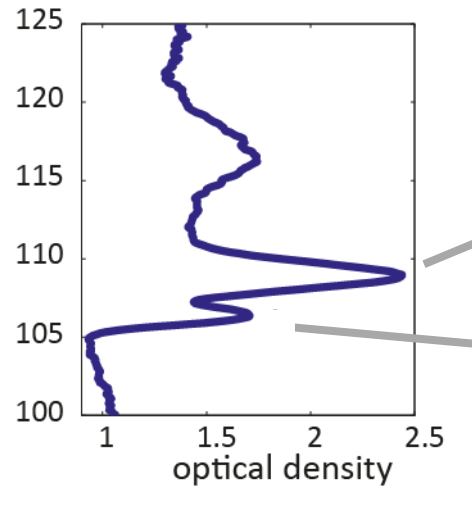
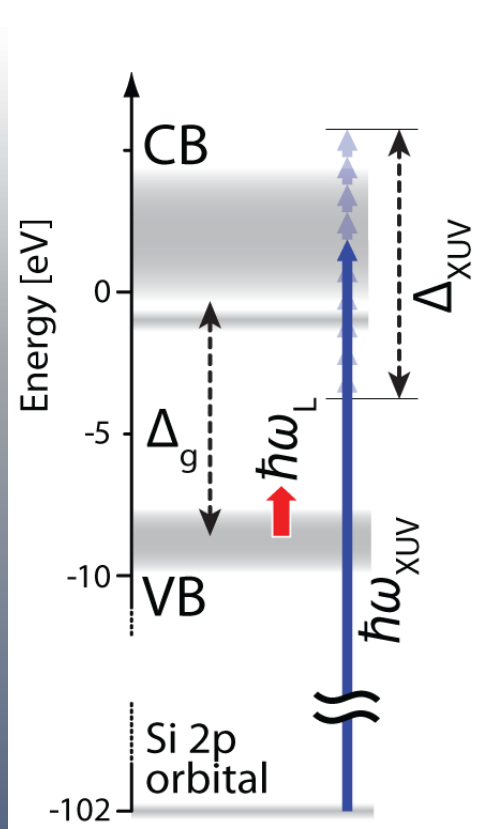


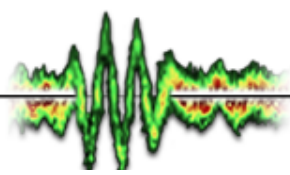
Tunneling into the CB



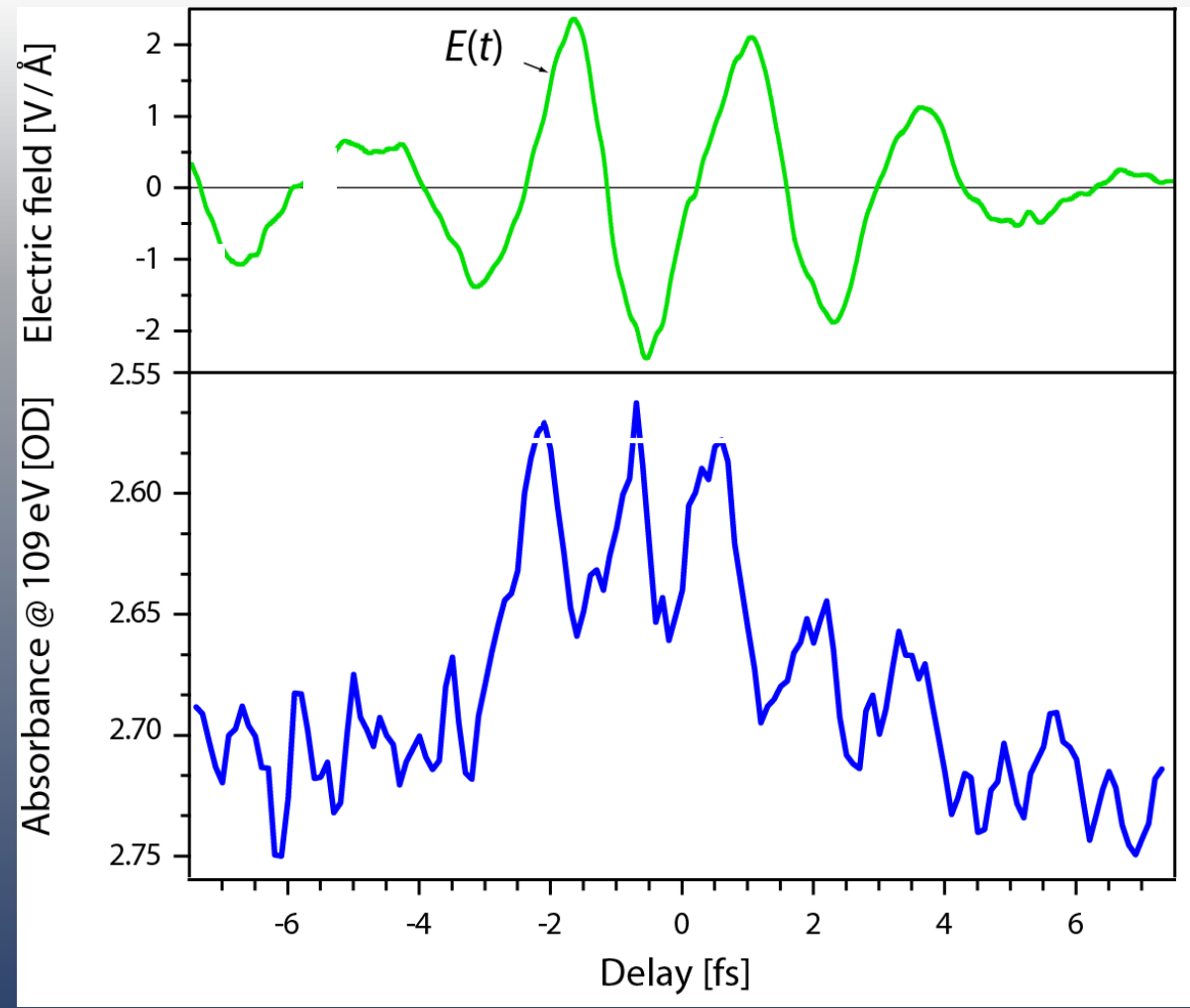
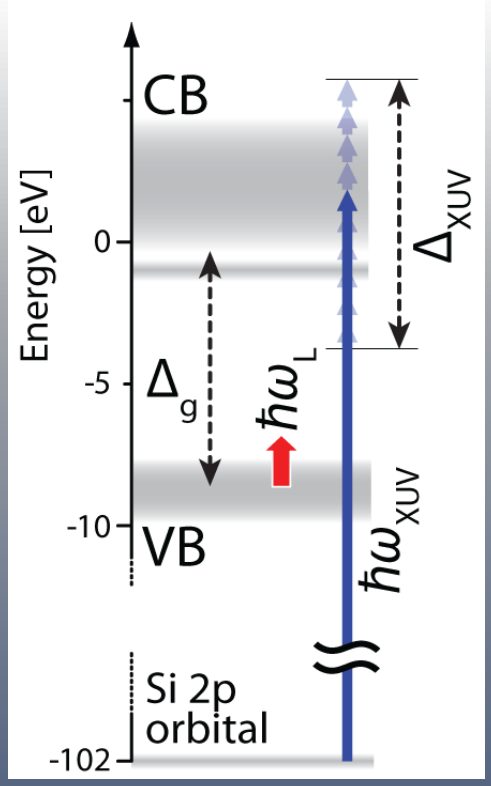


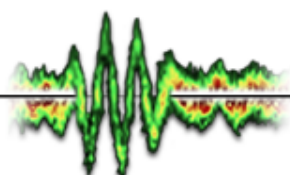
Dynamics in Dielectrics



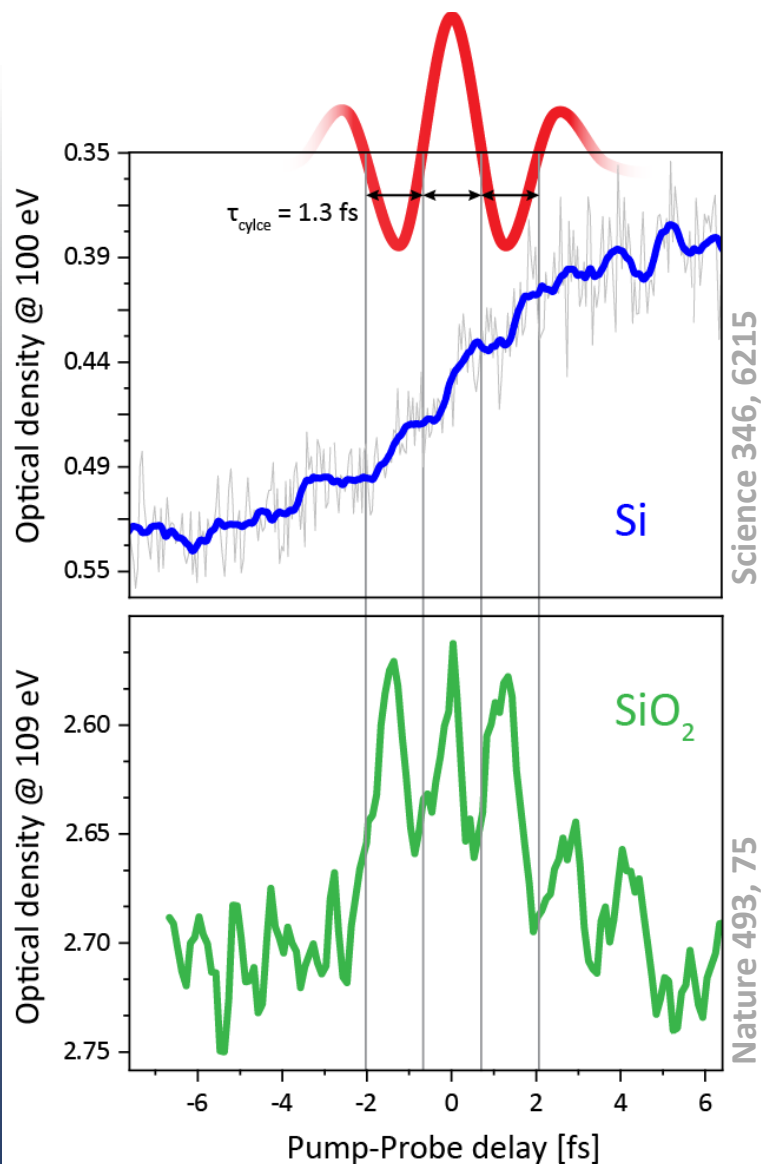


Dynamics in Dielectrics





Ultrafast Nonlinearities

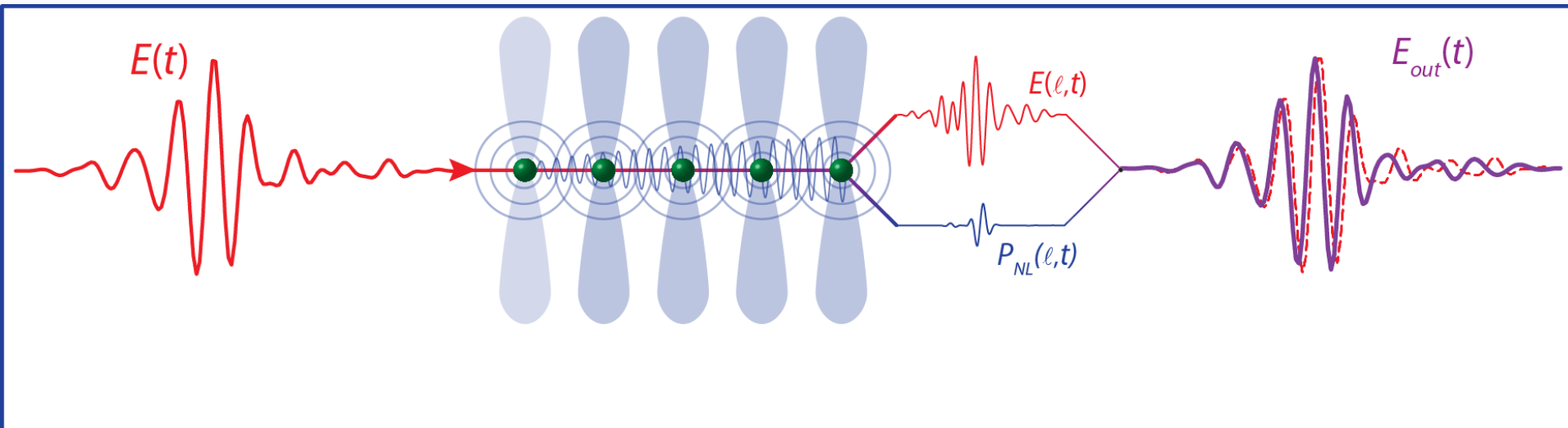
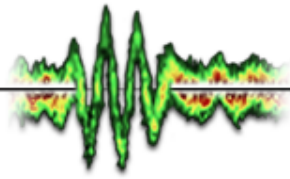


Photodoping of
CB states

Transient
Photoconductivity

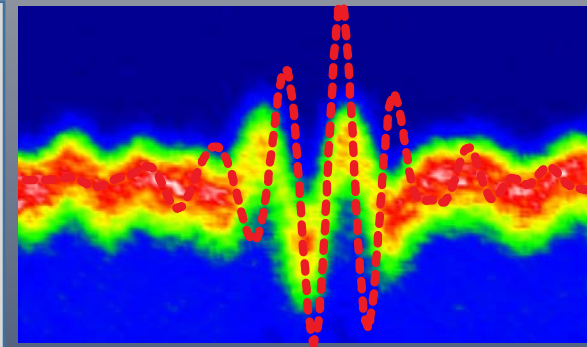
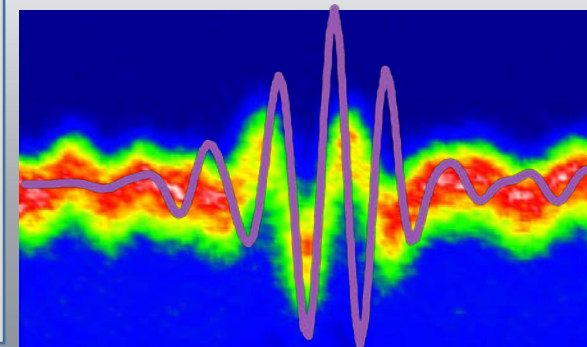
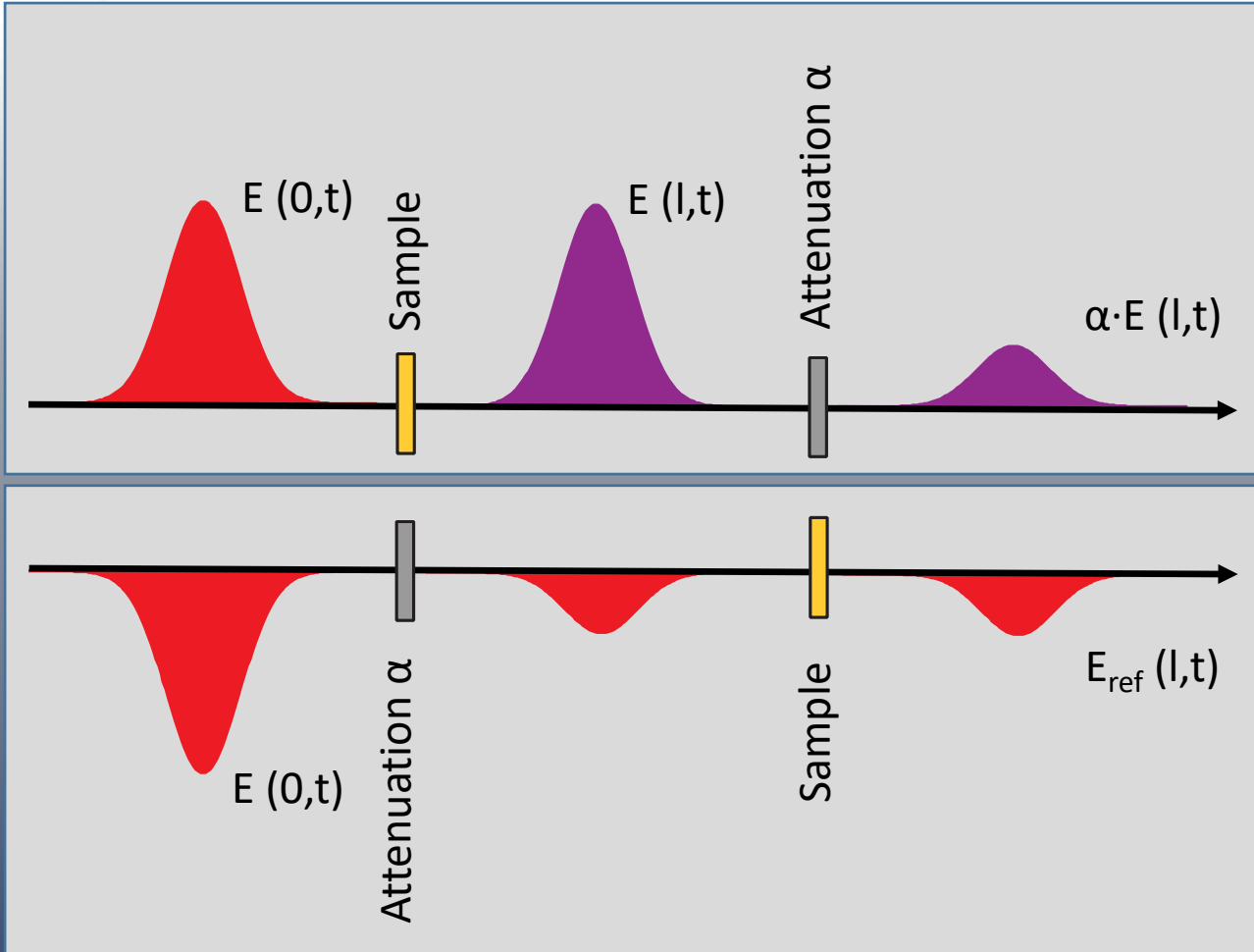
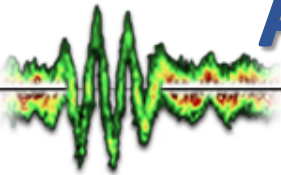
**Nonlinearities with
sub-fs response time!**

Nonlinear polarization response

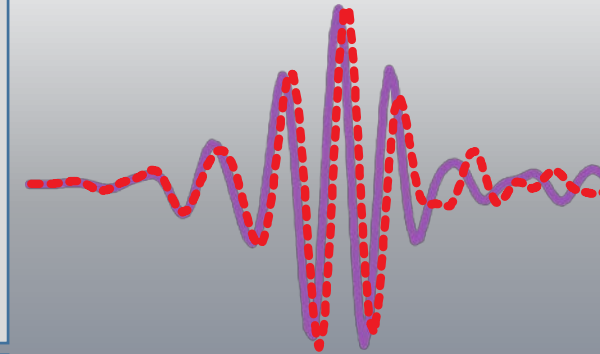
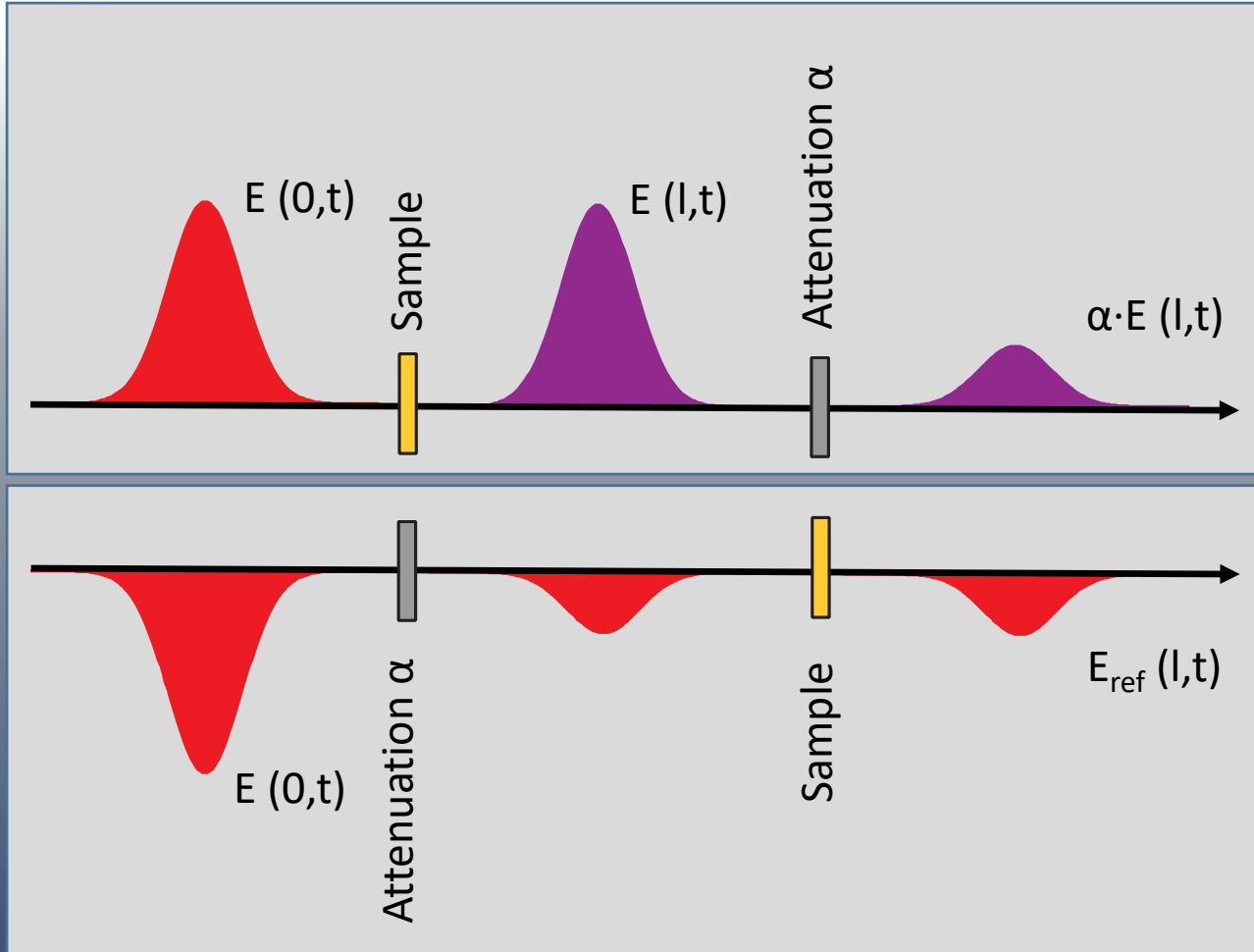
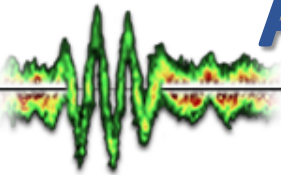


$$P_{NL}(t) \propto E_{out}(t) - E(l, t)$$

Attosecond polarization sampling

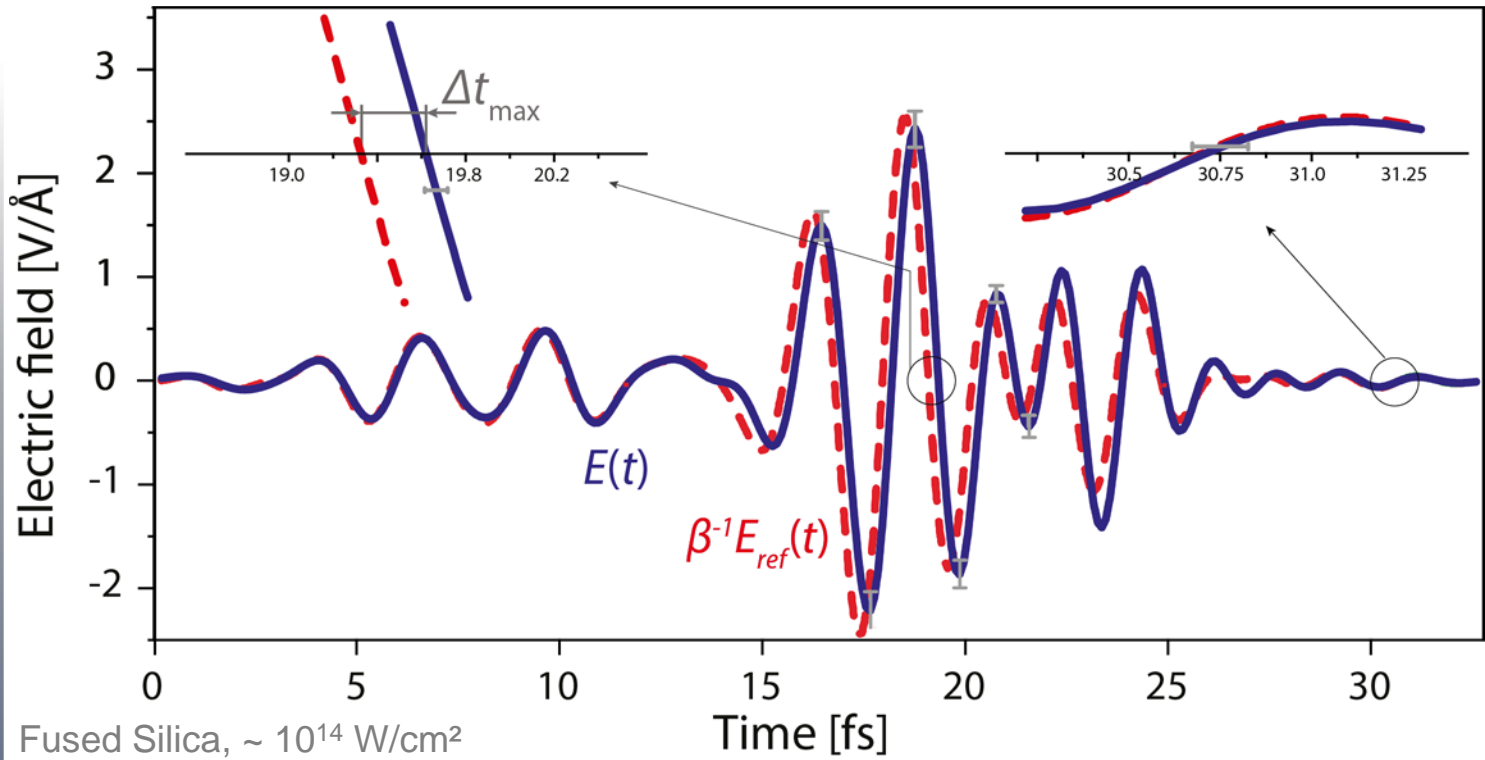
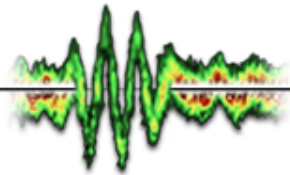


Attosecond polarization sampling

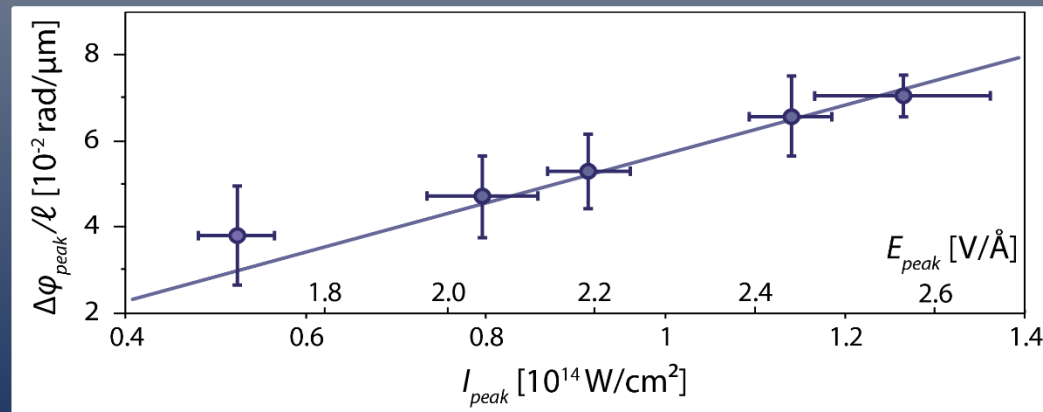


$$P_{NL}(t) \propto E_{out}(t) - E(l,t)$$

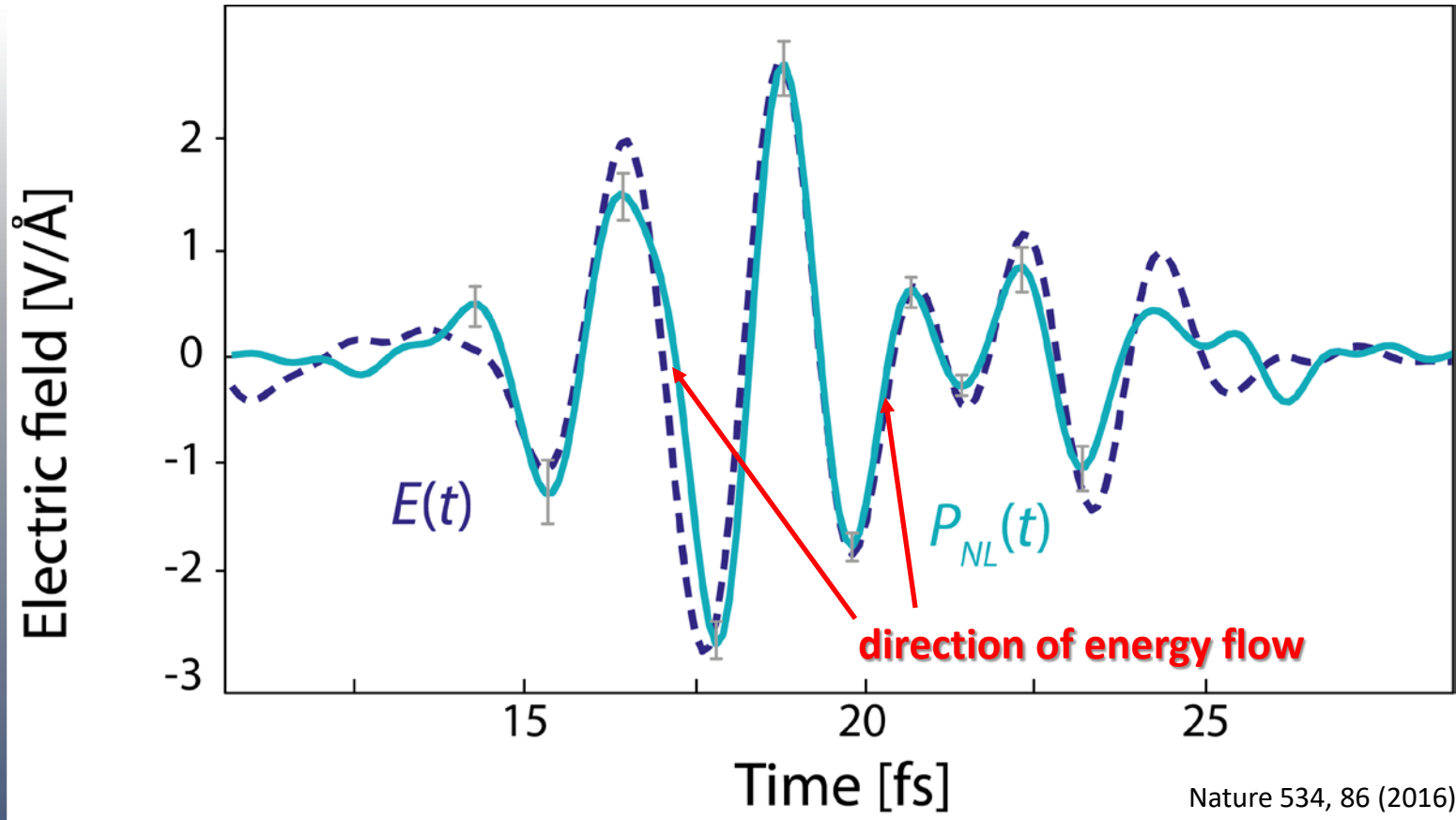
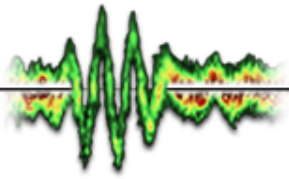
Nonlinear refractive index



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



Nonlinear polarization density

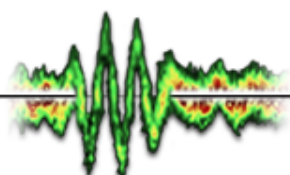


$$P_{NL} \sim \langle x_{electrons}(t) \rangle$$

Nonlinear polarization density



Electron displacement

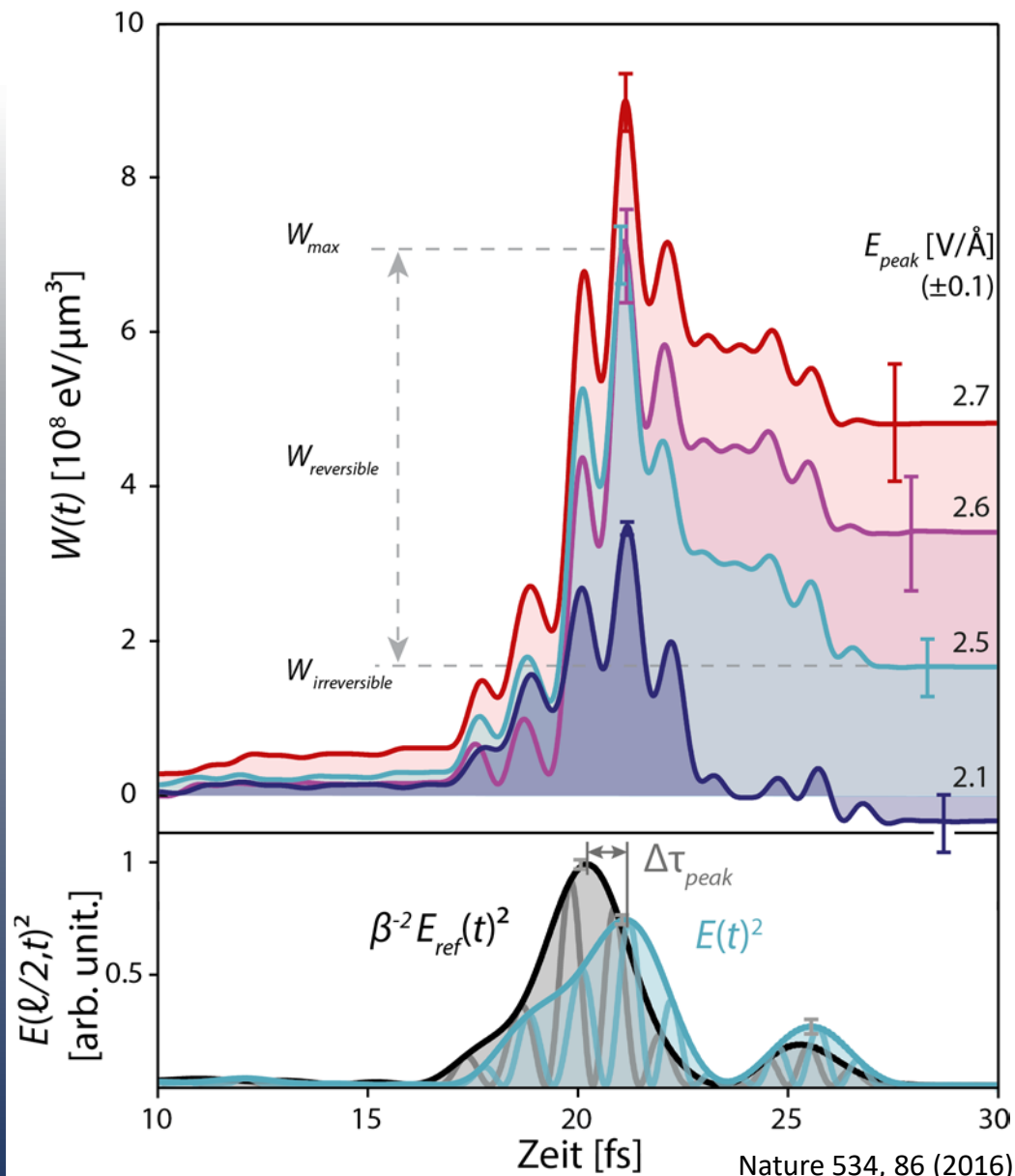


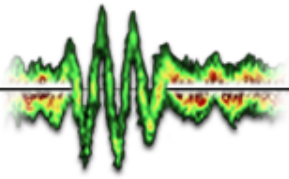
Energy transfer dynamics

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

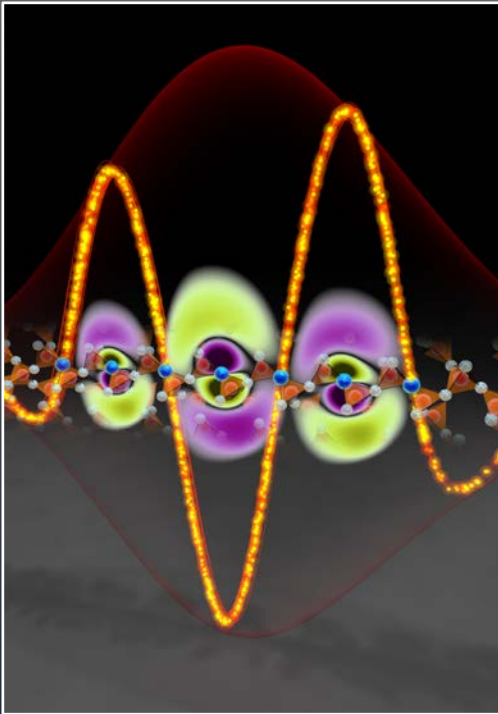
$$W(t) = \int_{-\infty}^t \overset{\text{measured}}{E(t')} * I(t') dt'$$

$$\overset{\text{measured too!}}{\frac{d}{dt} P_{NL}(t)}$$





Electron dynamics in solids



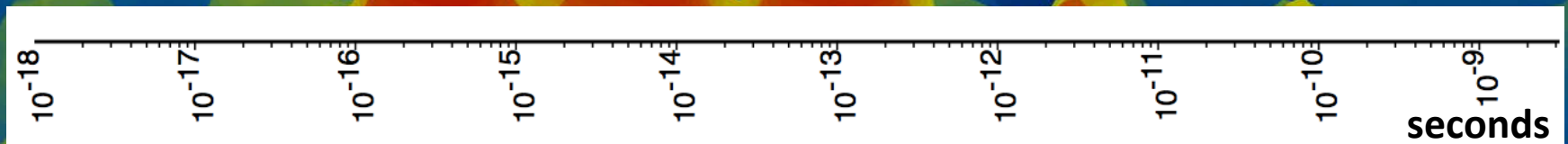
Ultrafast spectroscopy in solids

Ultrafast laser pulse can reversibly modify the electronic properties of band-gap materials

Electronics at optical clock rates

These developments hold promise for opto-electronic metrology and signal manipulation at a clock rate 10000 times faster than current micro electronics

Attosecond solid state spectroscopy



e-h creation

e-e scattering

e-phonon scattering

coherent electronics

optical phonon decay

recombination

- Quantitative observation of optical excitations
- Decouples electron and lattice response
- Explores transient electronic states

dynamics into & inside the bands, energy transfer dynamics, scattering times, electron-phonon coupling, carrier relaxation

Attosecond electron dynamics in band-gap materials

Thanks to:

Ferenc Krausz & Team at MPQ Garching / LMU Munich

Annkatrin Sommer, Florian Siegrist, Nicholas Karpowicz

Steve Leone, Dan Neumark & Team at UC Berkeley

Krupa Ramasesha

**K. Yabana, U Tsukuba, U Tsukuba & D. Prendergast, LBNL Berkeley & M. Stockman,
GSU Atlanta & J. Burgdörfer, TU Vienna**

S.A. Sato, D. Pemmaraju, V. Apalkov, R. Pazourek, S. Nagele

