PHz electronics – attosecond field-induced dynamics in condensed matter

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Zoom Telecon on Applications of Dielectric Accelerators
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Questions to be addressed

1. What are the characteristics of an on-chip electron source that make it attractive for this application area?
2. Are there new capabilities that would be uniquely enabled (i.e. which could not be done with a conventional accelerator system)?

   Potential applications: Petahertz electronics

   Attosecond transient absorption

   needs state-of-the art theory to unravel underlying electron dynamics

   Electron and/or X-ray diffraction (or imaging) could measure charge density dynamics more directly
3. What are the minimum required accelerator performance parameters for useful application of the technology?

   Theory is available – need to get better prediction for diffraction signal

Outline

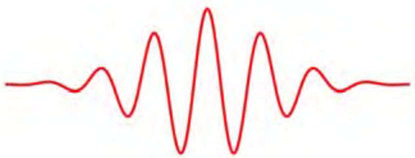
- Moving towards condensed matter:
  - Attosecond transient absorption spectroscopy (ATAS)
  - Example: dynamical Franz-Keldysh effect in diamond and GaAs
  - Theory helps to unravel underlying physics
  - Example: attosecond screening dynamics by electron-localization in Ti and Zr
  - Current issues and alternative complementary measurements?

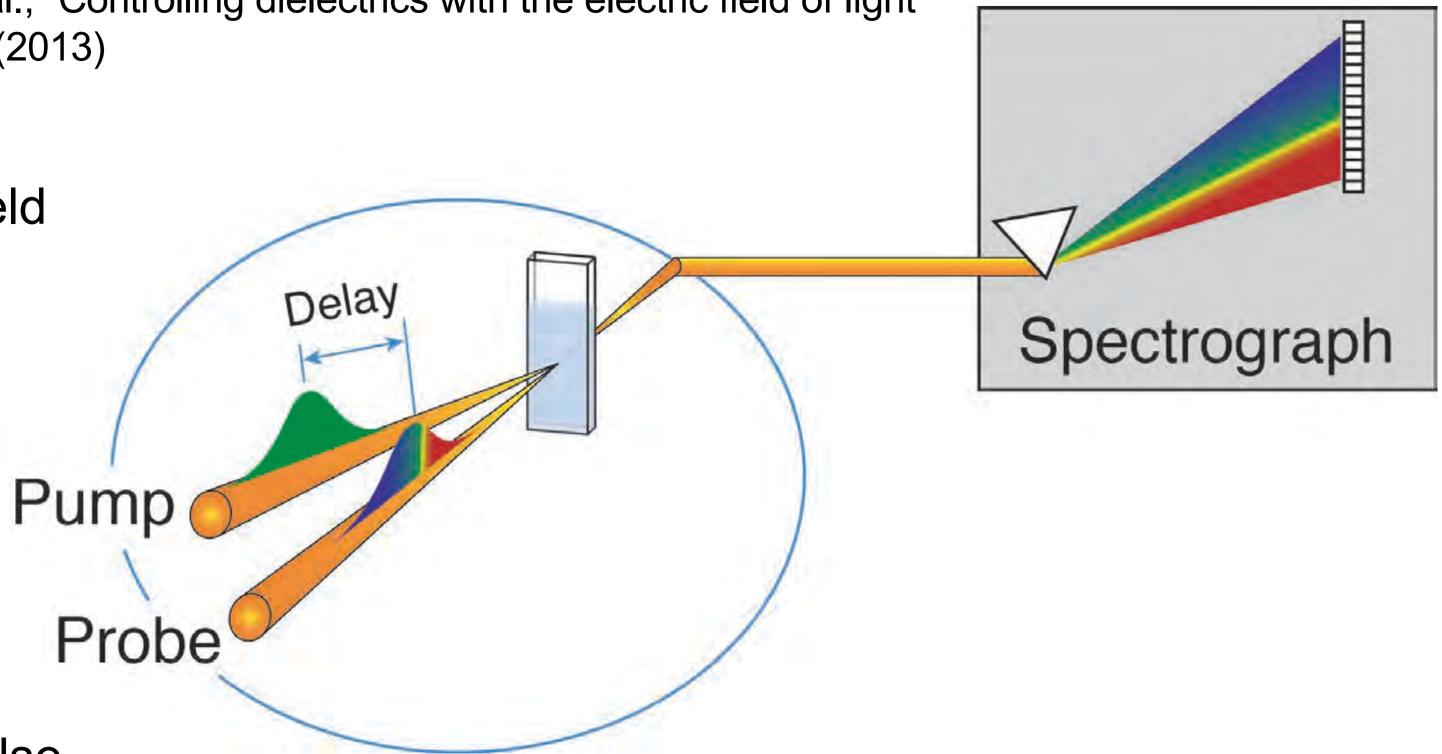
Attosecond transient absorption spectroscopy (ATAS)

- Pioneering ATAS applied to atoms and molecules in gas phase:
  - E. Goulielmakis *et al*, "Real-time observation of valence electron motion", *Nature* **466**, 739 (2010).
  - M. Holler et al.," Attosecond Electron Wave-Packet Interference Observed by Transient Absorption" *Phys. Rev. Lett.* **106**, 123601 (2011).
  - M. Chini et al., "Sub-cycle Oscillations in Virtual States Brought to Light", *Sci. Rep.* **3,** 1105 (2013).
  - M. Lucchini et al., "Role of electron wavepacket interference in the optical response of helium atoms", *New J. Phys.* **15**, 103010 (2013).
- Pioneering ATAS in condensed matter:
  - M. Schultze et al., "Controlling dielectrics with the electric field of light" *Nature* **493**, 75 (2013)

Pump:
Infrared (IR) laser field

$\vec{E}(t)$

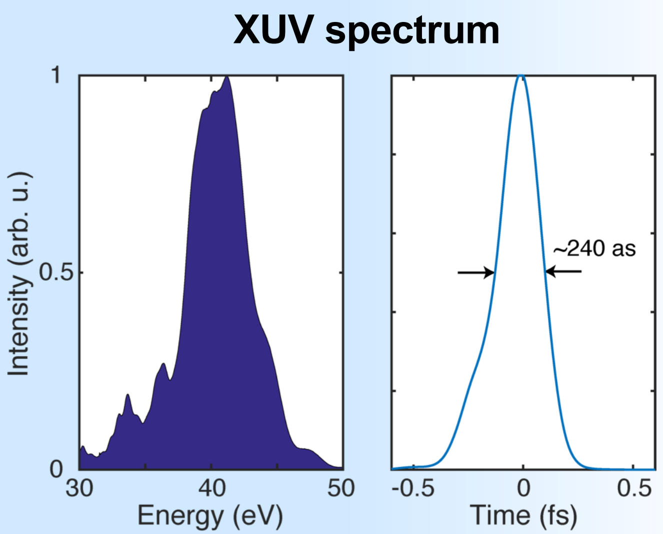


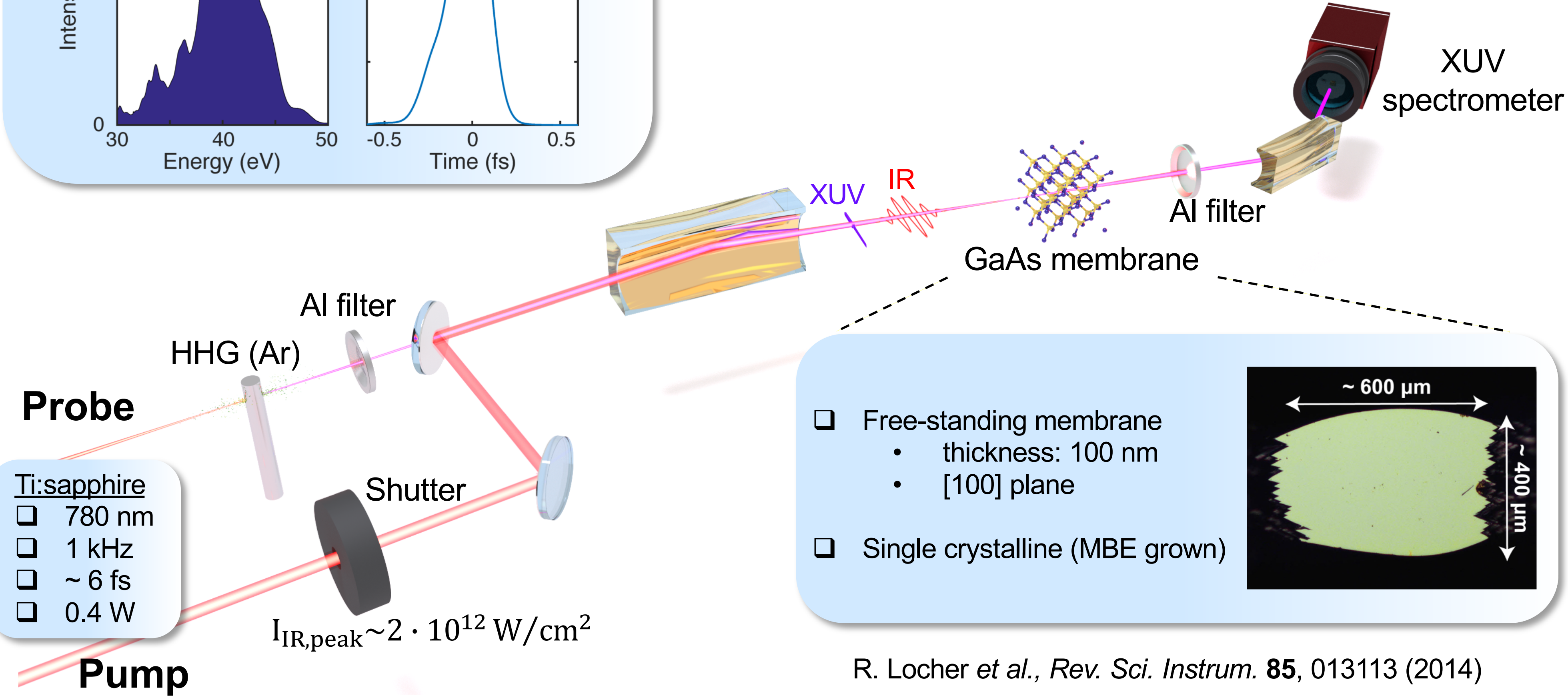


Probe:
Attosecond XUV pulse

Experimental ATAS setup

Probe: Attosecond XUV pulse





Ti:sapphire

- 780 nm
- 1 kHz
- ~ 6 fs
- 0.4 W

- Free-standing membrane
  - thickness: 100 nm
  - [100] plane
- Single crystalline (MBE grown)

R. Locher *et al.*, *Rev. Sci. Instrum.* **85**, 013113 (2014)

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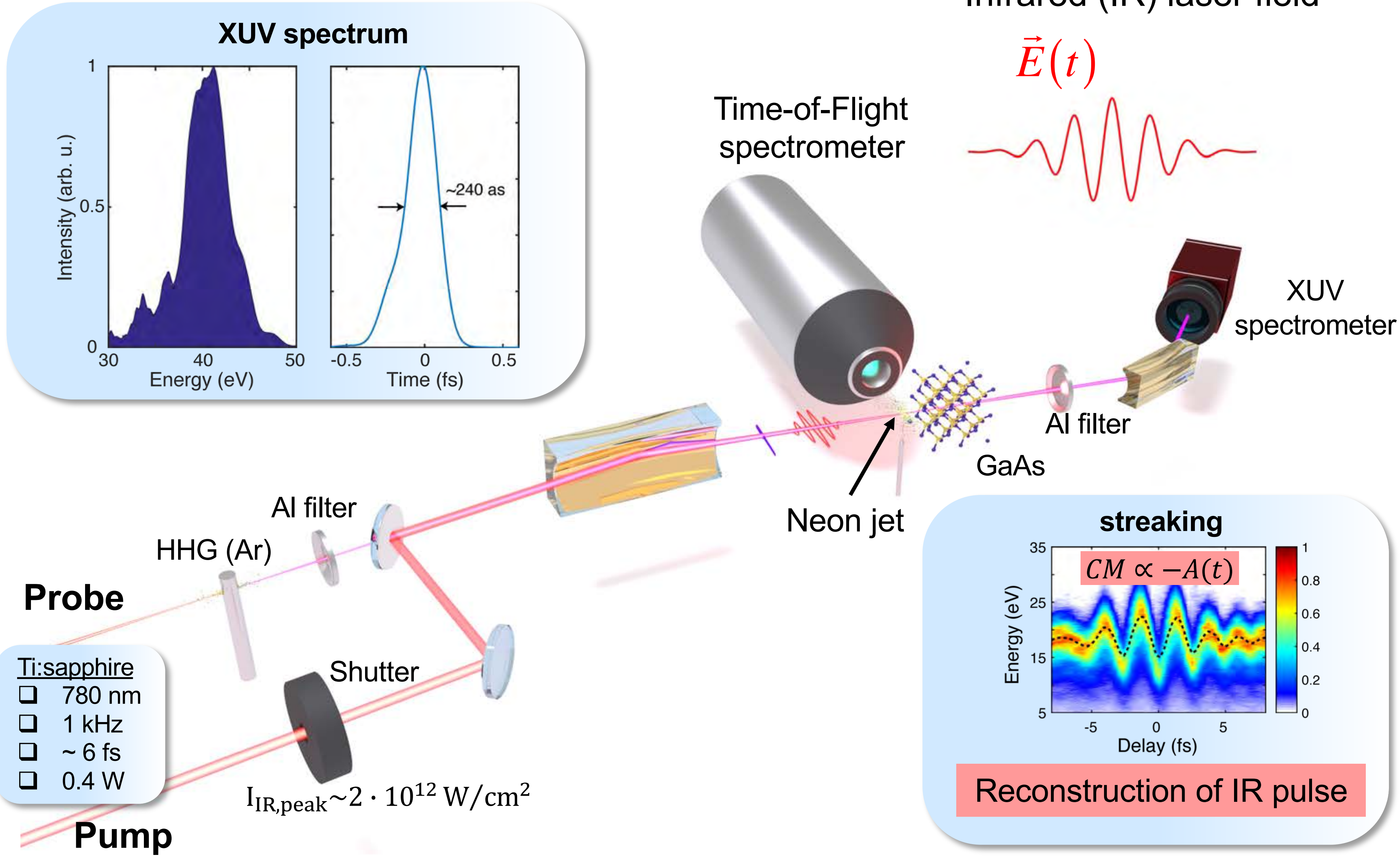
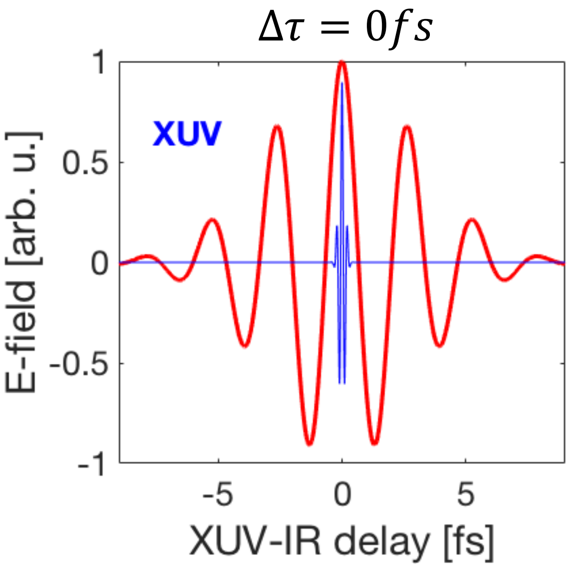
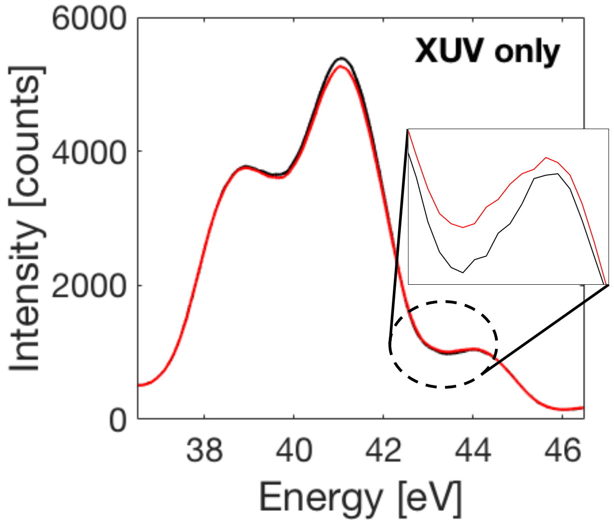
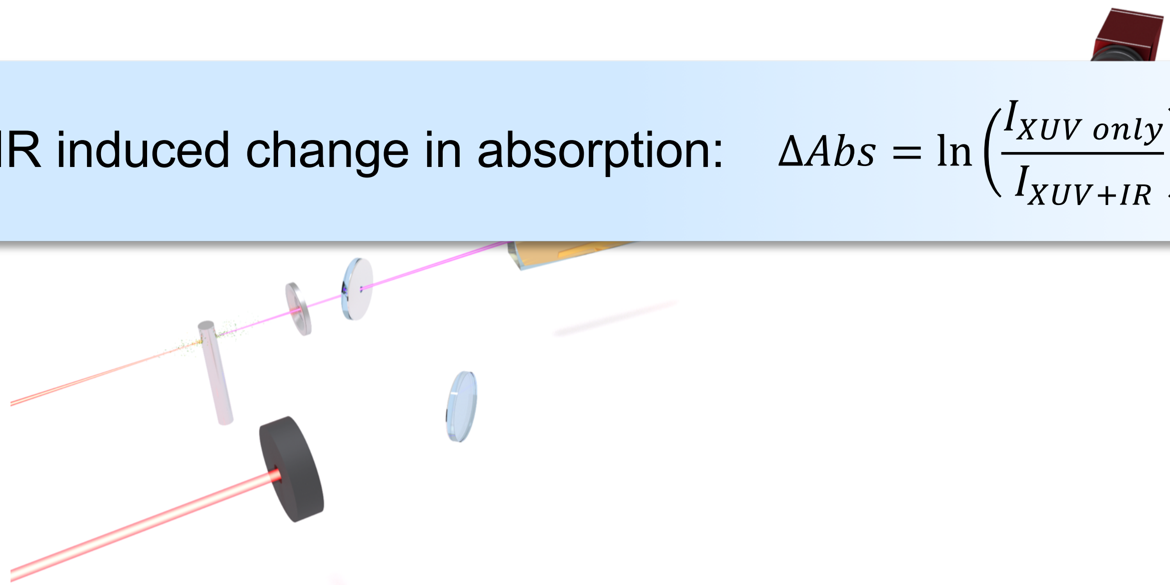


Transient absorption spectroscopy

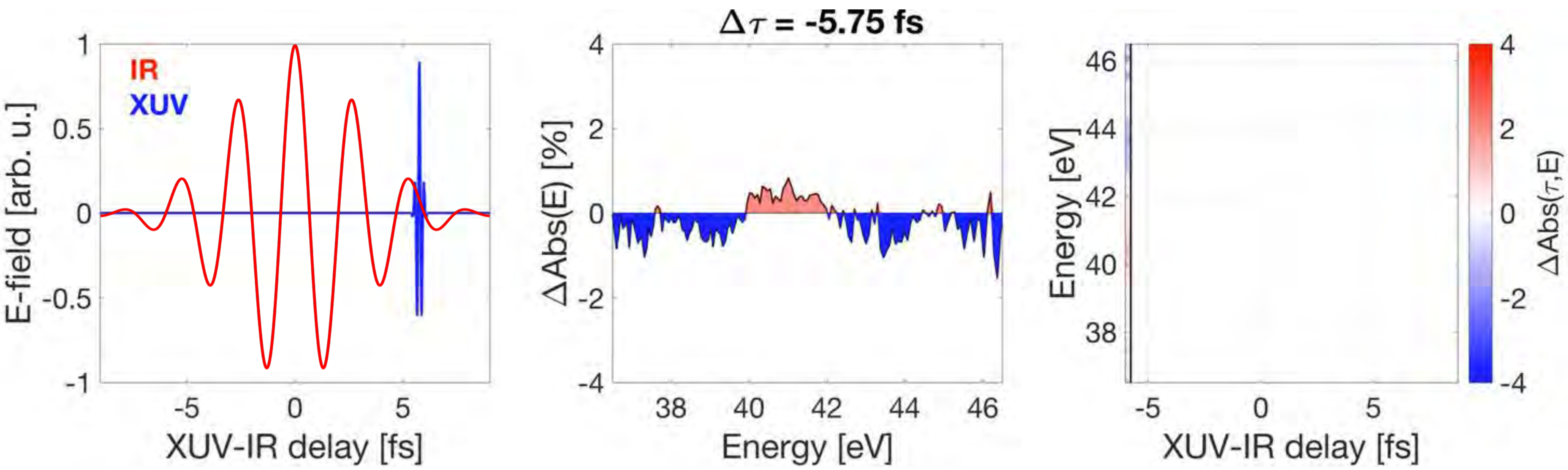




IR induced change in absorption: $\Delta Abs = \ln\left(\frac{I_{XUV\ only}}{I_{XUV+IR}}\right)$



Transient absorption spectroscopy



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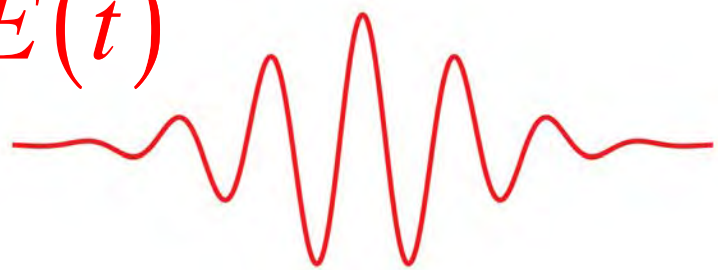
Dynamical Franz-Keldysh Effect (DFKE)

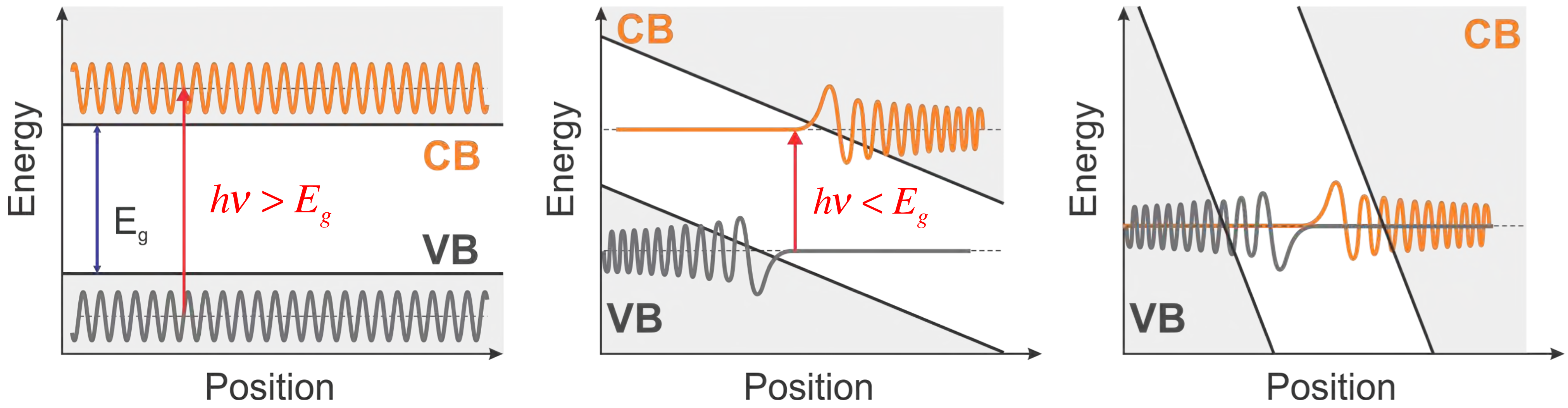
Example: Diamond
non-resonant excitation

short laser pulse $h\nu << E_g$

$\vec{E}(t)$



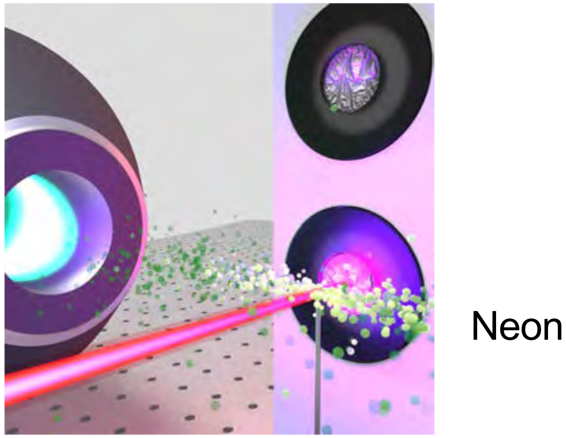
Photoabsorption | Mixed behavior (DFKE) | Tunneling

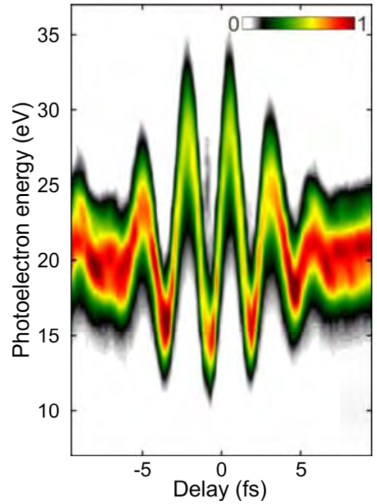


Excitation process?
intra-band or **inter**-band ("dressed states")

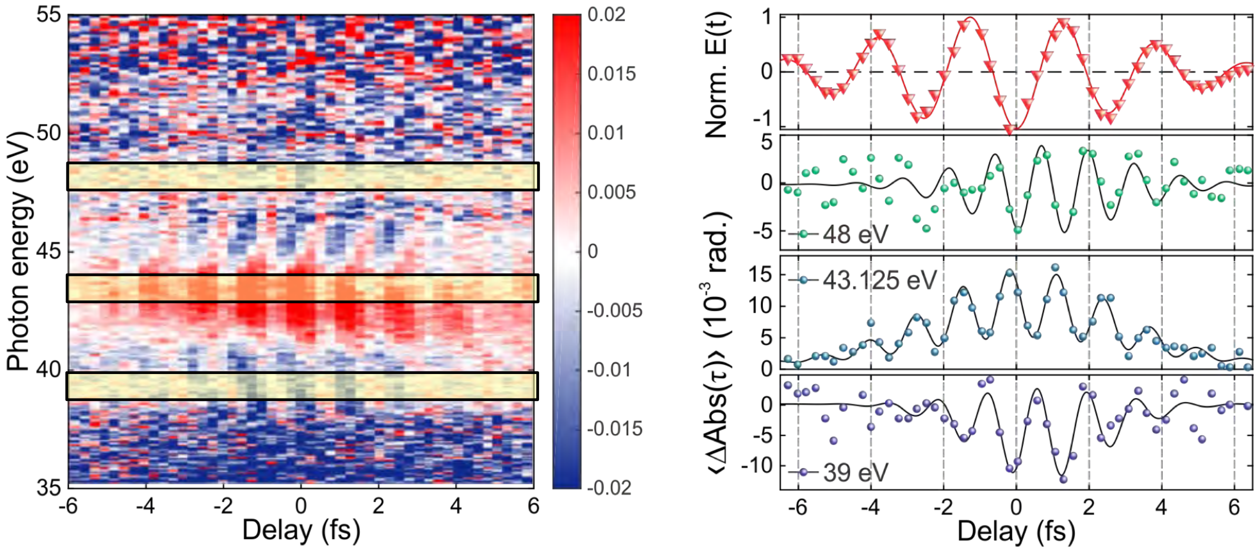
Phase relation with the IR field

- Double target for simultaneous photoelectron and photon detection



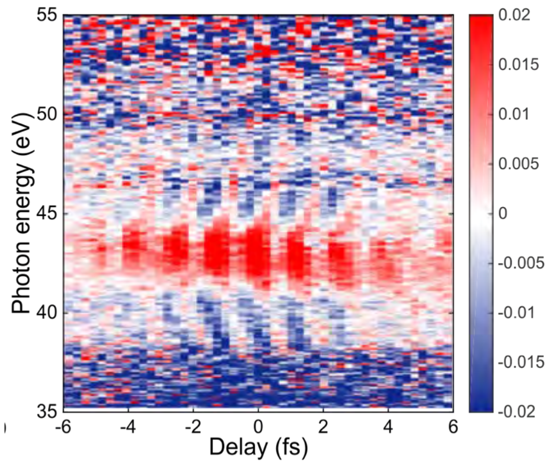


- We can extract the relative phase between the transient features and the IR field amplitude

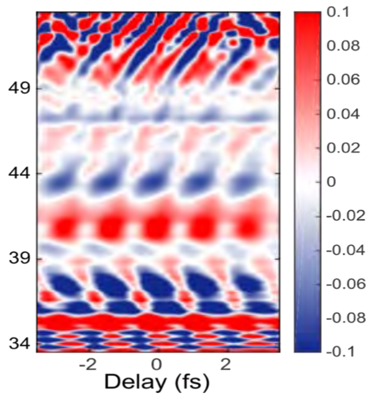


- We can extract the delay between the ATAS signal and $E^2_{IR}(t)$. The main features are not exactly in phase with the IR field.

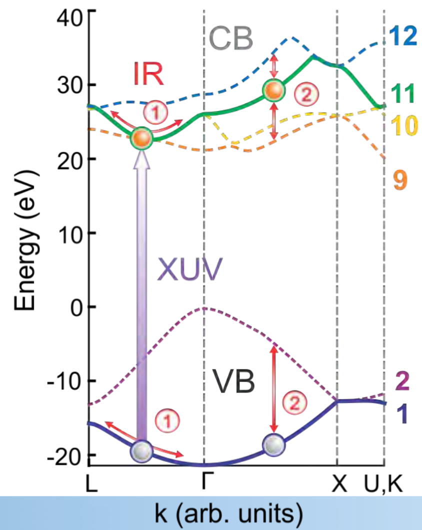
Conclusions from diamond (nonresonant)



1) First observation of close-to-PHz oscillations in diamond transient absorption pumped with field-controlled few-cycle IR pulses.



2) *Ab initio* numerical calculations fully reproduce the experimental results with the non-trivial energy-dependent phase:
TDDFT to solve the time-dependent Kohn-Sham equation

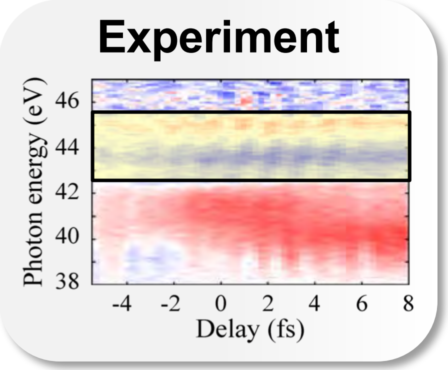


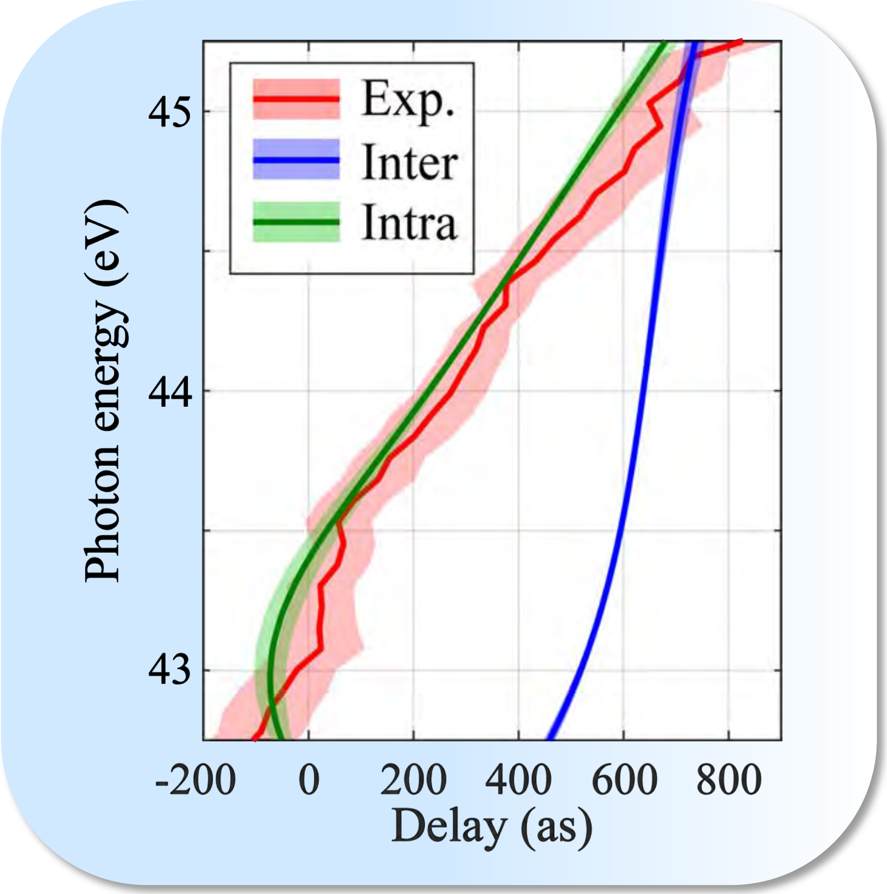
3) Orbital decomposition reveals that only two states are relevant. Comparison with a 2-band parabolic model allows to explain in the results in the framework of DFKE.

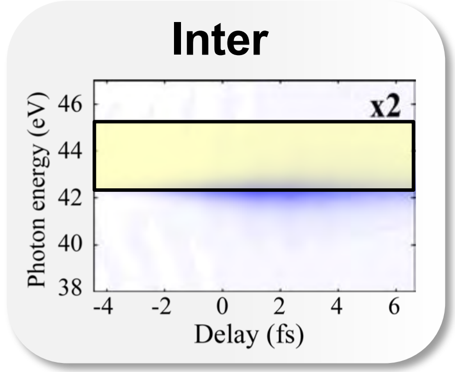
M. Lucchini, S. A. Sato, A. Ludwig, J. Herrmann, M. Volkov, L. Kasmi, Y. Shinohara, K. Yabana, L. Gallmann, U. Keller, *Science* **353**, 916, 2016

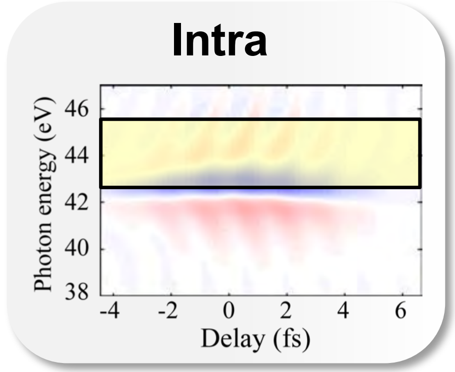
Conclusion from GaAs (resonant)

Comparison of the oscillation phase between experiment and simulation.







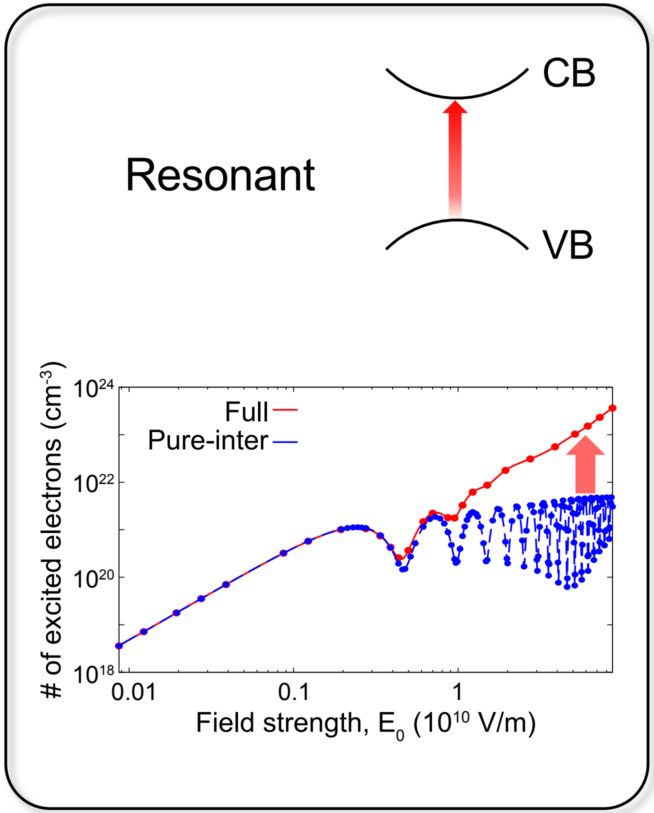


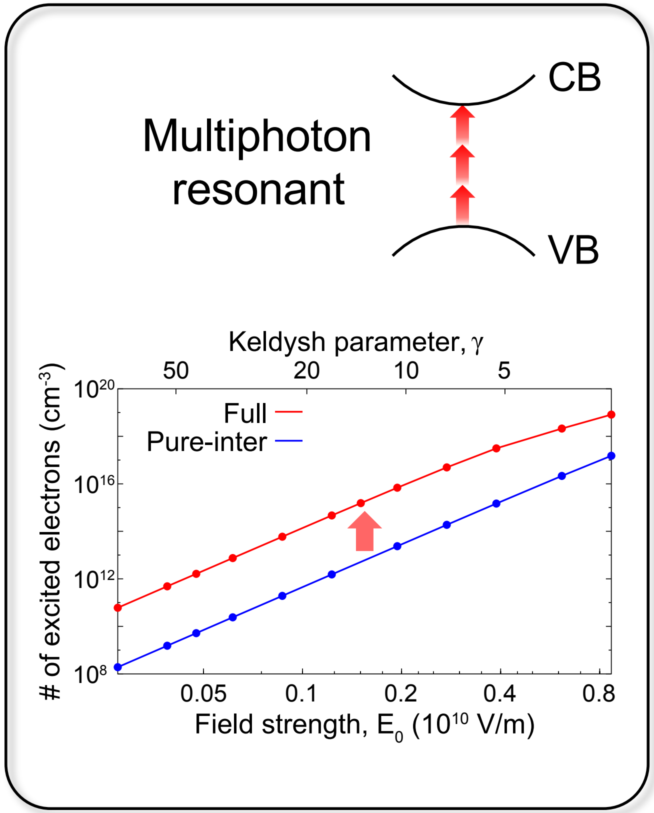
Oscillation delay of experiment perfectly reproduced by intra-band motion!

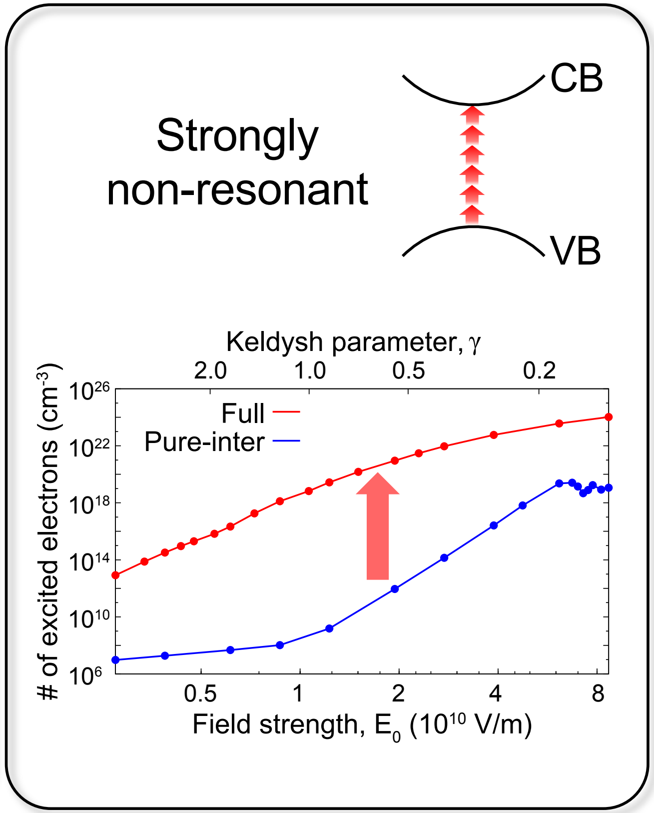
→ **Ultrafast optical response is dominated by intra-band motion!**

F. Schlaepfer, M. Lucchini, S. A. Sato, M. Volkov, L. Kasmi, N. Hartmann, A. Rubio, L. Gallmann, U. Keller
Nature Physics 14, 560 (2018)

Injection process for different limits







Intra-band motion largely enhances the carrier injection in a **large range of excitation parameters.**

→ **Universal** process

S. A. Sato, M. Lucchini, M. Volkov, F. Schlaepfer, L. Gallmann, U. Keller, and A. Rubio, *PRB* **98**, 035202 (2018)

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  - Attosecond transient absorption spectroscopy (ATAS)
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TDDFT with the exchange and correlation functional

TD Kohn-Sham equations

$$i\frac{\partial}{\partial t}\varphi_i(\mathbf{r},t) = \left[\frac{1}{2}\left(-i\nabla - \frac{\mathbf{A}(t)}{c}\right)^2 + v_{\mathrm{KS}}[n](\mathbf{r},t)\right]\varphi_i(\mathbf{r},t)$$

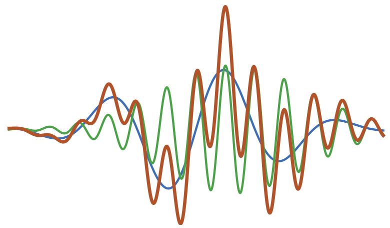
$$v_{\mathrm{KS}}[n](\mathbf{r},t) = v_{\mathrm{ion}}(\mathbf{r},t) + v_{\mathrm{H}}[n](\mathbf{r},t) + v_{\mathrm{xc}}[n](\mathbf{r},t)$$

$$n(\mathbf{r},t) = 2\sum_{i=1}^{N/2}|\varphi_i(\mathbf{r},t)|^2$$

Our approach

Propagation of the KS equations in real-space and real-time

The external field is an arbitrary function of time



It can describe any linear combination of laser pulses

Key features

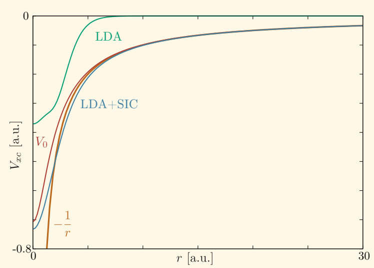
- Exact reformulation of the TD Schrödinger equation
- Very good compromise between computational efficiency and accuracy.
- All observables as functional of the TD density

exchange and correlation functional

it accounts for many-body effects

Correct asymptotic decay is crucial for ionization potential (molecules) and excitons (solids)

Available options:
Self-interaction correction (SIC)
LB94
EXX + hybrids



Courtesy of Dr. Umberto De Giovannini in the group of Prof. Angel Rubio, MPI Hamburg

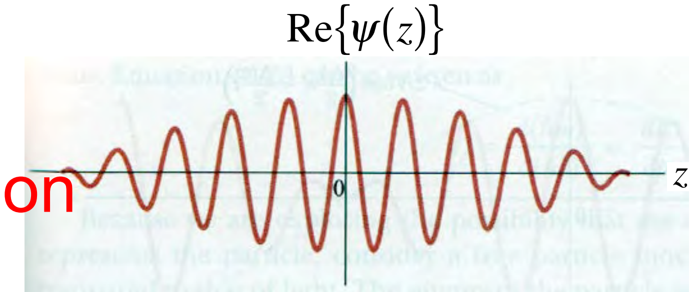
Quantum Mechanics – approximations required

- **Time Dependent Schrödinger Equation (TDSE)**
  wave function $\psi(\boldsymbol{r},t)$

$$\hat{H}\;\psi(\boldsymbol{r},t) = i\hbar\frac{\partial}{\partial t}\psi(\boldsymbol{r},t)$$

- **Born Oppenheimer Approximation**
  electrons much faster than atoms

$$\Psi_{total} = \Psi_{electronic}\cdot\Psi_{nuclear}$$



- **Single Active Electron (SAE) approximation and wave packet dynamics**
  very successful in solid-state physics, electron transport in semiconductor nanostructures, HHG, molecular dynamics ....

- **Time-Dependent Density Functional Theory (TDDFT)**
  challenge: loose phase information
  \+ successful for weakly correlated systems
  \- not successful for strongly correlated systems (high-T_c superconductivity)

$$\mathrm{TDSE} \rightarrow \mathrm{TDDFT}$$

$$\psi(\boldsymbol{r},t) \rightarrow |\psi(\boldsymbol{r},t)|^2$$

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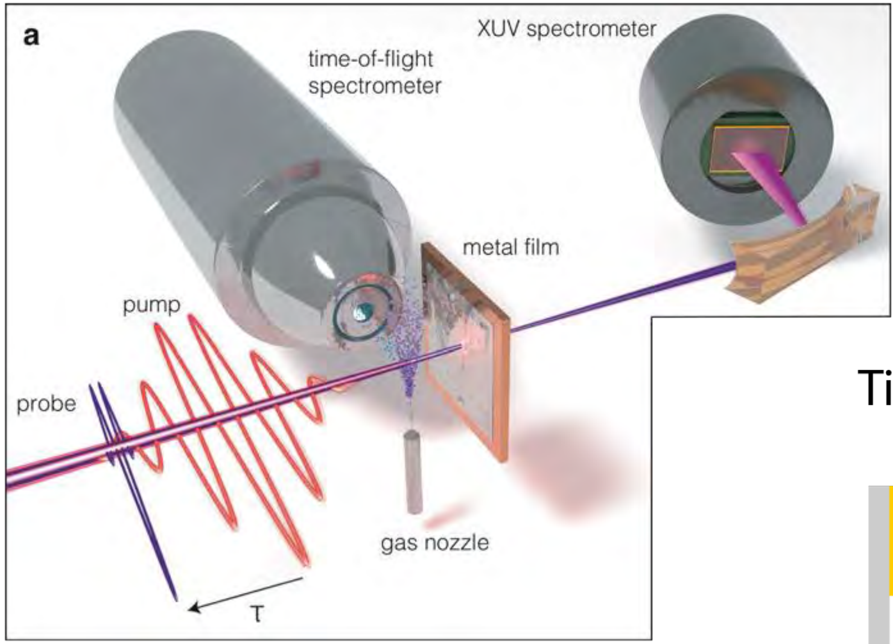
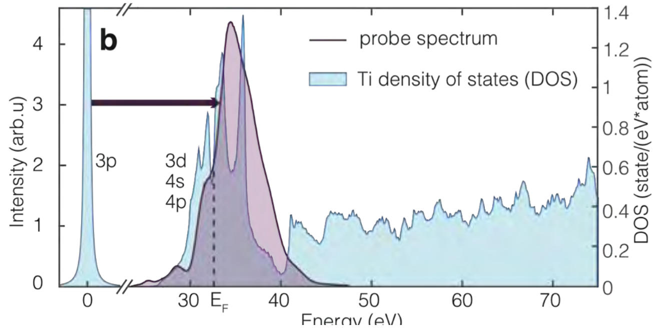
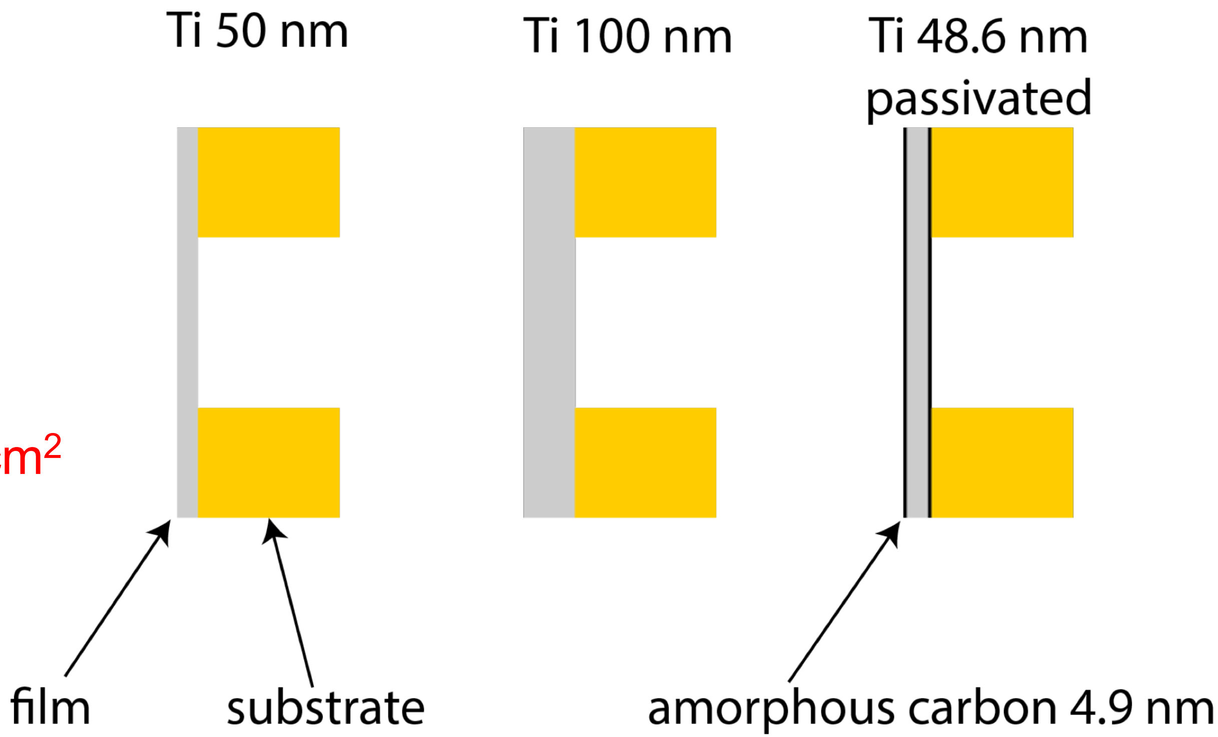
ATAS on transition metal Ti (and Zr)







Probe XUV, ≈35 eV, 260 as
dominated by 3p-3d transition
Pump IR, 1.55 eV, ≈7x10^{11} W/cm^2

Ti, Titanium
[Ar] 3d^2 4s^2

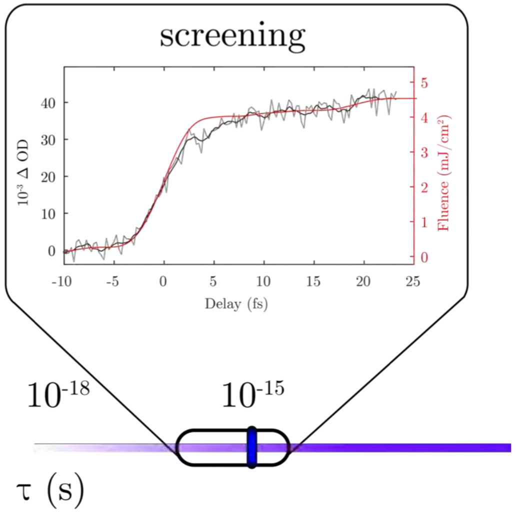
Zr, Zirconium
[Kr] 4d^2 5s^2

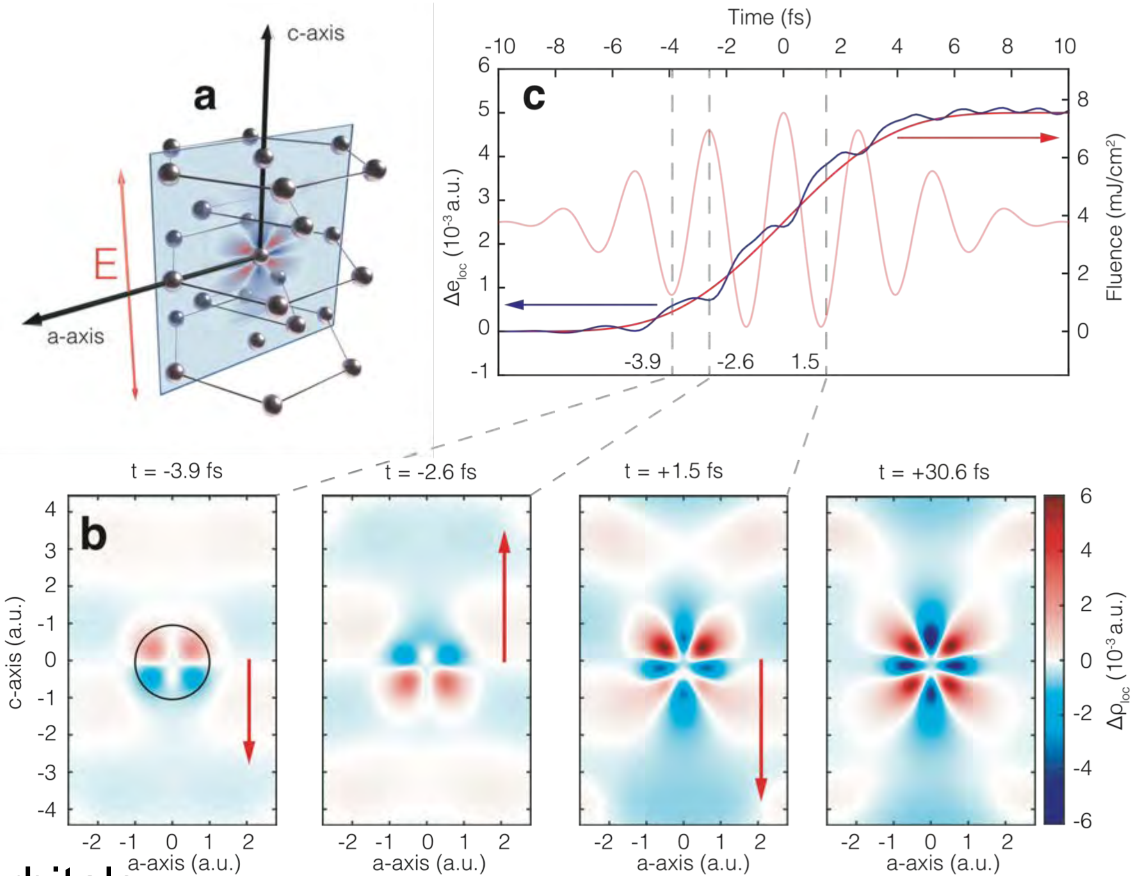
M. Volkov, S. A. Sato, F. Schlaepfer, L. Kasmi, N. Hartmann, M. Lucchini, L. Gallmann, A. Rubio, U. Keller, "Attosecond screening dynamics mediated by electron-localization", *Nature Physics* **15**, 1145 (2019)

Attosecond screening dynamics

Many-body electron dynamics in transition metals before thermalization sets in

Ultrafast electronic localization on d-orbitals





Electron density difference induced by near-IR electric field

At the end of the near-IR pump pulse creates an inflow of electronic density towards the Ti atom and localizes it on the d-orbitals.

M. Volkov, et al., *Nature Physics* **15**, 1145 (2019)

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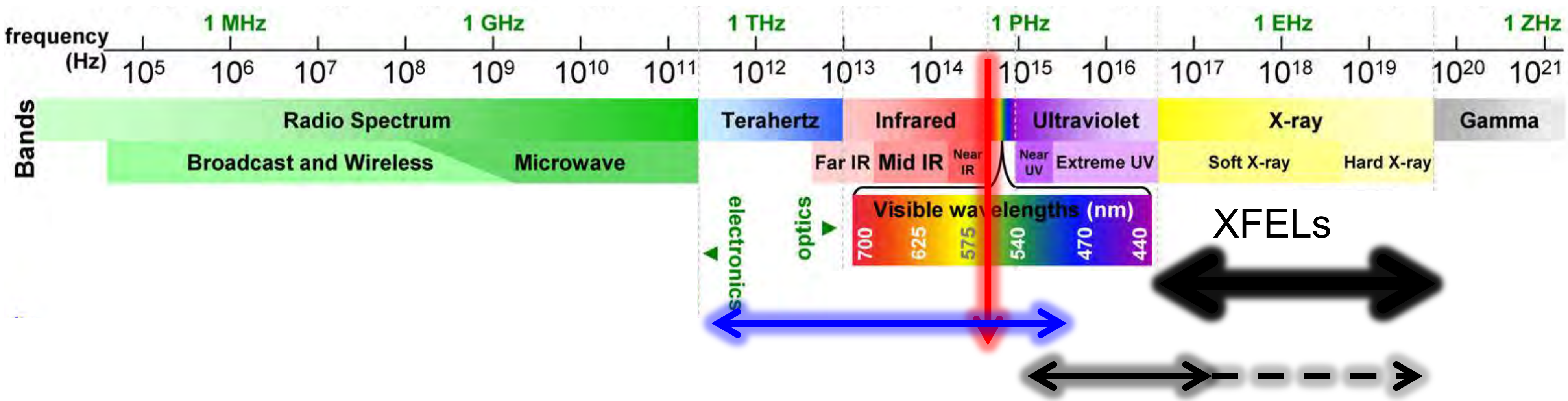
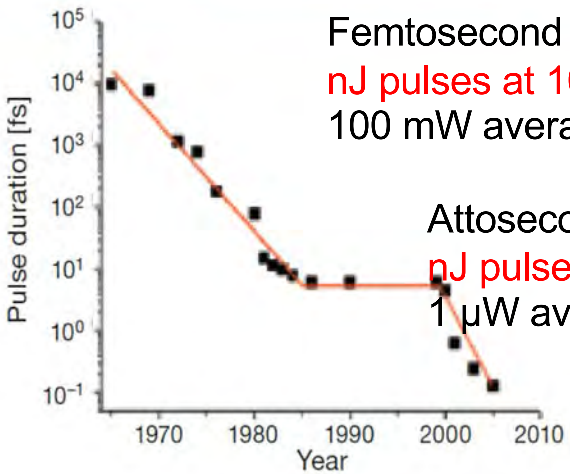
Future Lasers ...



High Harmonic Generation (HHG)

nonlinear optics beyond perturbation theory

Transient absorption spectroscopy



Femtosecond domain:
nJ pulses at 100 MHz
100 mW average power

Attosecond domain:
nJ pulses at 1 kHz
1 μW average power

Future developments:

- Higher repetition rate attosecond HHG sources with higher photon flux
- Attosecond pump and probe
- Higher photon flux in soft and hard X-ray
- Attosecond XFELs
- Attosecond electrons

Challenge for attosecond science with HHG:
Low pulse repetition rates
limits signal-to-noise (≈5 orders of magnitude reduction)

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Prof. Dr. Angel Rubio

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