Ultrafast laser physics (ULP)

1 picosecond = 1 ps = 10^{-12} s
1 femtosecond = 1 fs = 10^{-15} s
1 attosecond = 1 as = 10^{-18} s
Measurement with μs time resolution

Flash photography:
Flash lights driven by electronics
⇒ triggered flash lights
⇒ ≈μs time resolution
  (already available 1935)
limited by flash duration
(„light pulse duration“)
The problem

• Straightforward: Measure slow event with fast event

• However, all detectors are time-integrating on these time scales

• Solution: Map dynamics/time axis to static observable!

E. Muybridge: Animal Locomotion (1887)
The solution

• The classical pump-probe approach:

![Diagram showing time mapping to space translation]

- Map time to translation in space: \( \tau = \frac{2\Delta x}{c} \propto \Delta x \)
- Therefore \( S(\Delta x) \leftrightarrow S(\tau) \)
- 1 nm resolution in \( \Delta x \) yields 7 as resolution in \( \tau \)
- Delay is equivalent to real time if duration of probe pulse is negligible and process is perfectly reproducible
- This idea can be generalized to other mappings of time to time-independent quantities
Pump-Probe Measurement

\[ z = c t \]

\[ t \approx 2 \times 3.3 \text{ fs} \]

\[ \Delta z = 1 \mu m \]

\[ 2 \cdot \Delta z = c \Delta t \]
Differential Transmission Spectroscopy

- Why a chopper?
- Why not the chopper in the probe pulse?
- Why do you use a lock-in amplifier?
Ultrafast measurements need some kind of nonlinearities in the measurement system (i.e. intensity dependent transmission)

\[
\text{signal} = \left[ T(\Delta t, I_{\text{pump}}) - T(I_{\text{pump}} = 0) \right] I_{\text{probe}}
\]
Different arrangements

- Noncollinear degenerate pump-probe measurements
- Collinear degenerate pump-probe measurements
Noncollinear: pump and probe beam **not** collinear
good for signal-to-noise because pump power is not on detector

Degenerate: pump and probe pulse have the same central wavelength
What is the reason for the PBS (polarizing beam splitters) in the set-up?
Potential problem? Detector can be saturated by strong pump beam.
Why is this set-up a degenerate four-wave mixing experiment?
Degenerate four-wave mixing

Parallel polarization creates a transient diffraction grating inside the sample. This grating exists as long as there is a coherent excitation (i.e. within the dephasing time)

**Review articles:**  
J. Shah, "Ultrafast Spectroscopy of Semiconductors," Springer-Verlag
Degenerate four-wave mixing

\[ E_1 \rightarrow \text{PUMP} \]
\[ E_2 \rightarrow \text{PROBE} \]
\[ \text{SAMPLE} \]

"Langsamer" Detektor

Polarisation => Beugungsgitter

\[ k_1 \]
\[ k_2 \]

\[ \Delta t \]

Polarisation

\[ 2k_2 - k_1 \]
Application: time resolved femtosecond luminescence measurement


Application of optical gating for “time-of-flight” imaging

Optical coherence tomography (OCT)

How does this work?

Science, 254, 1178, 1991
Optical coherence tomography (OCT)

Michelson Interferometer

Interference only within coherence length

Science, 254, 1178, 1991
Optical coherence tomography (OCT)

Normal versus Ultrahigh Resolution OCT

30 fs pulse duration $\rightarrow$ 10 $\mu$m axial resolution

10 fs pulse duration $\rightarrow$ 3 $\mu$m axial resolution

Prof. J. G. Fujimoto, MIT, USA
How do you do time resolved four-wave mixing?
Time resolved four-wave-mixing

PUMP

PROBE #1

Δt₁

Polarisation

LINSE

SAMPLE

k₁

k₂

2k₂ - k₁

NICHTLINEARER KRISTALL

PROBE #2

Δt₂

LINSE

LANGSAMER DETEKTOR
Photoconductive switch or Auston switch:
Photoconductive sampling gate
D. H. Auston, A. M. Johnson, P. R. Smith, J. C. Bean,
"Picosecond optoelectronic detection, sampling, and correlation measurements in amorphous semiconductors"
High-order harmonic generation in gases

Spectrum of harmonics

Log(Strength)

Plateau

Cutoff

Harmonic order

Step 1

Step 2

Step 3


tunnel ionization

acceleration in laser field

recombination

Classical electron-trajectories

→ Multiple trajectories with same recombination energy but different excursion time exist

e-trajectories

HHG

E-field

0

time

$t_i$

$t_f$

$\tau_1$

Short trajectory

$\tau_2$

Long trajectory
Laser-based HHG

Intense ultrafast Ti:sapphire CPA (≈800 nm, > 300μJ)
pulse repetition rate: ≈1 kHz (moving towards 10 kHz)
pulse energy center: up to 100 eV
pulse energy of attosecond pulses: < nJ
pulse duration: ≈100 as

Laser-based HHG: “a success story”

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

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

Challenges/Problems of laser based HHG:
Low pulse repetition rates and low pulse energy!
limits signal-to-noise (≈5 orders of magnitude reduction)
Streaking techniques instead of pump-probe

attosecond resolved measurements strongly signal-to-noise limited
use phase sensitive techniques instead:
  energy streaking: mapping time to energy
  angular streaking: mapping time to angular momentum

attosecond pulse synchronized with strong infrared field
strong infrared field can be used for streaking

energy streaking: mapping time to energy (linear polarized streaking field)

Attosecond streak camera

- Most versatile and most successful technique to date: 
  Hentschel et al., Nature 414, 509 (2001)

1. The attosecond pulse and an intense, short infrared pulse are overlapped in/on a medium being studied – they can be delayed with respect to each other
2. The attosecond pulse ionizes the medium
3. The vector potential of the infrared pulse shifts the resulting electron spectrum in energy as a function of the relative delay

Measured at ETH, 2012
Streaking techniques instead of pump-probe

Attosecond resolved measurements strongly signal-to-noise limited

Use phase sensitive techniques instead:

- Energy streaking: mapping time to energy
- Angular streaking: mapping time to angular momentum

Attosecond pulse synchronized with strong infrared field

Strong infrared field can be used for streaking

Angular streaking: mapping time to angular momentum (circular polarized)

How long does it take for an electron to traverse the tunneling barrier in tunnel-ionization of helium?

Optica 322, 1525 (2008)
and much more ….