Spektroskopie und Photonik mit Femtosekunden-Lasern

1. Femtosecond (fs) laser pulses:
   general properties, generation,
   light matter interaction (linear), dispersion

2. Nonlinear optics:
   nonlinear light matter interaction,
   new frequencies, THz spectroscopy with a single shot,
   white light from lasers

3. Ultrafast spectroscopy:
   ultrafast Ohm-meters, Ampere-meters,
   magnetometers etc.

4. Ultrafast nano-optics:
   plasmons, photonic crystals,
   manipulation of light on-the-fly
1. Femtosecond (fs) laser pulses
   - general properties
   - applications
2. Fs pulse generation
   - laser
   - mode locking
3. Pulse propagation in vacuum
4. Excursus: light matter coupling
   - Lorentz model
5. Pulse propagation in matter
   - dispersion
   - dispersion compensation
6. Pulse shaping
What is a femtosecond laser pulse?

- electromagnetic pulse with duration of ~1 to 1000fs
- here: 100-fs pulse, centered at 800 nm (400THz, 2.5fs per cycle)
- peak field can be ~ 10 V/Å
- pulses come periodically with frequency $f_{\text{rep}}$

Typically ~1 W average power for Ti:sapphire

$\Rightarrow$ pulse energy = 10 nJ at $f_{\text{rep}} = 100$ MHz, pulse energy = 1 mJ at $f_{\text{rep}} = 1$ kHz etc. (⇒ laser is on for 1s/d, 1s/300y)
What is a femtosecond laser pulse?

Pulses can have a quite complex temporal structure, e.g. chirp.

\[ E(t) = Re \left[ A(t) \exp(i \omega_c t) \right] \]

carrier oscillation

complex-valued envelope
How short is a femtosecond?

1 fs = $10^{-15}$ s

1 s:
light travels from earth to moon

100 fs:
light propagates only 30 μm
(thickness of a human hair)
How short is a femtosecond?

8 fs:
one vibration of H-H bond of H₂

10 fs:
~ time between subsequent collisions of an electron in a metal

500 fs:
bacteriorhodopsin turns from cis into trans conformation
Femtosecond pulses - what for?

Femtosecond laser pulses have unique properties:

1. short duration
2. high peak intensities
3. stable repetition rate
Ultrafast time-domain spectroscopy

Pulse (“pump”) triggers ultrafast processes, e.g.
• molecular or lattice vibration
• spin precession
• electron-hole pairs

Pulse (“probe”) monitors ultrafast processes, e.g.
• pump-induced dynamics
• thermally induced random dynamics (noise spectroscopy)
Ultrafast photonics

Nonlinear optics: light interacts with light

Ultrafast optical switches
• for controlling light with light

Generation of new wavelengths
• longest wavelengths (terahertz pulses, $\lambda \sim 1$mm):
  probe low-energy resonances
• shortest wavelengths (soft x ray, $\lambda \sim 1$nm):
  for x-ray diffraction and core-level spectroscopy

Generation of electron pulses
• for ultrafast electron diffraction
Scanning microscopy

Two-photon microscopy
• two photons absorbed, one emitted
• increases resolution by $\sqrt{2}$

Stimulated-emission depletion (STED) microscopy
• exploits nonlinear saturation of stimulated emission
• breaks diffraction limit ($\sim \lambda/2$) by factor of $\approx 20$
Scanning microscopy

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Micromachining

Reduction of pulse duration

\[ \tau = 20 \text{ ps} \quad \tau = 1 \text{ ps} \quad \tau = 150 \text{ fs} \]

Absorbed laser pulse heats electrons at first

\[ \Rightarrow \text{shorter laser pulses} \]
- give higher (electron) temperatures for same energy
- minimize the heat-affected zones
- yields higher reproducibility
- give higher process efficiency
Frequency comb

pulse train ("comb") in time domain
⇒ comb in frequency domain


application: measurement of optical frequencies with high accuracy
How to generate fs pulses?

Laser: self-sustained oscillator for light

We need:
1. light-amplifying medium
2. feedback

Lasing condition: gain ≥ loss

losses due to outcoupling, scattering, absorption
Gain by laser-pumped Ti:sapphire

Ti$^{3+}$ level scheme: levels split by coupling to phonons

$|1\rangle$ $|2\rangle$ $|3\rangle$ (long-lived) $|4\rangle$ (short-lived)

pump (514...532nm)
broad emission (650...1080nm)
Gain by laser-pumped Ti:sapphire

Ti$^{3+}$ level scheme:
levels split by coupling to phonons

|0\rangle
|2\rangle (long-lived)
|3\rangle (long-lived)
|4\rangle (short-lived)

pump (514...532nm)
broad emission (650...1080nm)

Light amplification by stimulated emission of radiation (LASER)

Consider: Light oscillation by stimulated emission of radiation
Fabry-Perot resonator for feedback

only wavelengths \( \lambda_j = \frac{2L}{j} \) survive

\( \Rightarrow \) frequency comb of modes

Idea: superposition of modes should give a wavepacket or pulse
Example for mode superposition

more modes...
Example for mode superposition

even more modes...
Example for mode superposition

\[ E(t, m) = \frac{1}{2m+1} \sum_{n=-m}^{n=m} \cos(2\pi (\nu + n\nu_f) t) \]

energy-time uncertainty: \( \Delta t \Delta f \sim 1 \)

\( \nu = 25, \Delta \nu_f = 0.05, m = 1 \)

\( \nu = 25, \Delta \nu_f = 0.05, m = 3 \)

\( \nu = 25, \Delta \nu_f = 0.05, m = 10 \)

\( \nu = 25, \Delta \nu_f = 0.05, m = 20 \)

Ti:sapphire laser oscillator: \( m \sim 10^6 \)
The laser

- Light wave
- Gain medium
- Gain curve
- Mode comb
- Loss

Mirror

Mirror
Switch gain on:
only one mode depletes gain medium

No pulse!

- reason: homogeneous gain profile
- How can we excite and phase-couple the other modes?
Active resonator modulation

modulator generates side bands at modulation frequency $\omega_{\text{mod}} = c/2L$

side bands are amplified, new sidebands etc.
Active resonator modulation

The modulator generates side bands at modulation frequency $\omega_{\text{mod}} = c/2L$

Side bands are amplified, new sidebands etc.

Mode locking $\Rightarrow$ pulse
Active resonator modulation

modulator generates side bands at modulation frequency $\omega_{\text{mod}} = c/2L$

side bands are amplified, new sidebands etc.

Mode locking $\Rightarrow$ pulse
Passive (self-) mode locking

assume we have pulse: oscillates in resonator with frequency \( c/2L \)
\[\Rightarrow \text{can be used for self-modulation of resonator}\]

Several approaches:

1. Saturable absorber

   becomes transparent at high intensities

2. Transient Kerr effect

   leads to self-focusing at high intensities
Fs laser in the lab

pump laser: Nd:YVO$_4$, 532 nm, 6.75 W

Ti:sapphire laser oscillator
• 10 fs pulse duration
• 10 nJ pulse energy
• 80 MHz repetition rate

~ $10^6$ coupled modes
Fs laser in the lab

intensity autocorrelation

![Autocorrelation signal](image)

\[ t_p = 10 \pm 1 \text{ fs} \]

delay time (fs)

intensity spectrum

![Intensity spectrum](image)

FWHM:
\[ \Delta \lambda = 103 \text{ nm} \]
\[ \Delta \nu = 51.3 \text{ THz} \]
\[ \Delta E = 1710 \text{ cm}^{-1} \]
Frequency comb

Pulse train (“comb”) in time domain ⇒ comb in frequency domain


Application: measurement of optical frequencies with high accuracy
<table>
<thead>
<tr>
<th>Year</th>
<th>Time</th>
<th>Prefix</th>
<th>Description</th>
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<td>$10^{-6}$ sec</td>
<td>micro</td>
<td>Laser, extern moduliert</td>
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<td>≈ 1965</td>
<td>$10^{-9}$ sec</td>
<td>nano</td>
<td>Güteschaltung aktiv</td>
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<td>$10^{-15}$ sec</td>
<td>femto</td>
<td>Dispersions-kompensation</td>
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<td>≈ 2000</td>
<td>$10^{-18}$ sec</td>
<td>atto</td>
<td>extrem nicht-lineare Optik</td>
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Pulse propagation

• Eq of motion for light in vacuum: Maxwell eqs
  \[ \Rightarrow \left( \nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) E = 0 \]

• plane wave \( E = E(z,t)e \) \( \Rightarrow \left( \frac{\partial^2}{\partial z^2} - c^{-2} \frac{\partial^2}{\partial t^2} \right) E = 0 \)

  general solution: \( E = u_- (z+ct) + u_+ (z-ct) \)

  pulse propagates with \( \pm c \), no dispersion

Reality:
1. beam is not plane wave \( \Rightarrow \) e.g. Gouy phase shift in focus
2. Beam propagates in media

We only consider 2.
Light-matter interaction

Light: Maxwell eqs, $j \rightarrow (E,B)$

Matter: Newton eqs, $j \leftarrow (E,B)$

Lorentz force $\propto E + (v/c) \times B$

equations of motion coupled by current $j$
Model: Lorentz oscillator

Assumptions:
• particle (electron, ion), charge \( q \), in potential minimum at \( x=0 \)
• harmonic approximation:
  \[ \nabla V = \omega_0^2 x \]  \( \Rightarrow \) linear theory
• electric-dipole approximation:
  \[ E(x,t) = E(0,t), \; B=0 \]
• friction force \(-2\gamma \partial_x x\)

\[ \Rightarrow \text{Eq of motion} \quad \hat{L}x = qE(t) \quad \text{with} \quad \hat{L} = \partial_t^2 + 2\gamma \partial_t + \omega_0^2 \]

\[ \Rightarrow \text{Fourier transformation} \quad x(\omega) = L^{-1}(\omega) qE(\omega) \quad L^{-1}(\omega) = \frac{1}{\omega_0^2 - \omega^2 - 2i\gamma\omega} \]

Lorentz function
Electric susceptibility

\[ P(\omega) = \frac{\text{dipole moment}}{\text{volume}} = Nq\chi = \frac{Nq^2}{L(\omega)} E(\omega) \]

- \( \chi \) quantifies how easily matter can be polarized (vacuum: \( \chi = 0 \), glass: 0.1 @800nm)
- \( \omega \Im \chi(\omega) \) quantifies locally absorbed light power
- time domain: \( \chi(t) \) is polarization response to \( \delta(t) \)
- \[ j(t) = \partial P(t)/\partial t \Rightarrow j(\omega) = -i\omega\chi(\omega)E(\omega) \Rightarrow \sigma(\omega) = -i\omega\chi(\omega) \]
Example: $\omega_0 = 25, \gamma = 1$
Fig. 15. Fit of a single Lorentz oscillator to the dielectric function of a pure CO ice at 10 K from E97 a), Baratta & Palumbo (1998) b) and Elisa et al. (1997) c). The best fitting parameters are: a) \( \gamma = 1.5 \text{ cm}^{-1}, \ \omega_p = 195 \text{ cm}^{-1} \) and \( \epsilon_0 = 1.67 \). b) \( \gamma = 1.5 \text{ cm}^{-1}, \ \omega_p = 160 \text{ cm}^{-1} \) and \( \epsilon_0 = 1.57 \). c) \( \gamma = 1.75 \text{ cm}^{-1}, \ \omega_p = 175 \text{ cm}^{-1} \) and \( \epsilon_0 = 1.67 \). All have \( \omega_0 = 2138.5 \text{ cm}^{-1} \). Additionally a second component has been fitted in panel c) with parameters: \( \gamma = 1.2 \text{ cm}^{-1}, \ \omega_p = 30 \text{ cm}^{-1} \) and \( \omega_0 = 2142.3 \).
Maxwell eqs in matter

Maxwell eqs \[\Rightarrow \quad \left( \nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) E = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} P \]

go to \(\omega\) space: \(P(\omega) = \chi(\omega) E(\omega)\)

\[\Rightarrow \quad [\nabla^2 + k^2(x, \omega)] E(x, \omega) = 0\]

Helmholtz eq, describes linear optics in isotropic space

\[k(x, \omega) = \frac{n(x, \omega) \omega}{c} \quad n = \sqrt{\varepsilon} \quad \varepsilon = 1 + 4\pi\chi\]

dispersion relation refractive index dielectric function

medium homogeneous, only one dimension \(z\), wave from \(z = -\infty\):

\[E(z, \omega) = E(0, \omega) \exp \left[ ik(\omega) z \right]\]
Pulse propagation in homogenous medium

\[ E(z,t) \propto \int d\omega \ E(0,\omega) \exp[ik(\omega)z - i\omega t] \]

Superposition of plane harmonic waves

\[ k(\omega) = \frac{n(\omega)\omega}{c} \]

Dispersion relation determines pulse evolution

• Assume Gaussian input: \[ E(0,\omega) \propto \exp\left[-\frac{(\omega - \omega_c)^2}{\Delta\omega^2}\right] \]

• Taylor expansion:

\[ k(\omega) \approx k_c + k'_c \cdot (\omega - \omega_c) + \frac{1}{2} k''_c \cdot (\omega - \omega_c)^2 \]
Pulse propagation in homogenous medium

\[ E(z,t) \propto \exp[\text{i}k_c z - \text{i} \omega_c t] \exp \left[ -\frac{(k'_c z - t)^2}{4\Delta \omega^{-2} - 2\text{i} k''_c z} \right] \sqrt{\frac{2\pi}{2\Delta \omega^{-2} - \text{i} k''_c z}} \]

carrier wave

Gaussian envelope:
- group velocity \( 1/ k'_c \)
- disperses if \( k''_c \neq 0 \)
- chirp

conserves energy
Chromatic dispersion

output pulse duration $\approx \left[ \frac{1}{v(\omega_2)} - \frac{1}{v(\omega_1)} \right] z \approx k'_cz \cdot (\omega_2 - \omega_1)$

- Example: 10-fs-pulse as input
  $\Rightarrow$ output pulse duration = 50THz 2000fs$^2$ = 100fs

- application: chirped-pulse amplification
Dispersion compensation

Goal: longer path for red than blue

prism or grating pairs

angular dispersion

“chirped” mirrors

local Bragg reflections

5 μm
More flexible: LCD pulse shaper

Fourier plane with amplitude and phase mask

Can create nearly arbitrary waveforms

(C) T. Brixner, U Würzburg