A short review of how femtosecond laser pulses are generated and what they are good for…

Ch 6 Ultrafast Laser Physics/Chemistry Lab

Geoff Blake & Matthew Kelley

Most slides adapted from: http://www.chem.ucsb.edu/~ocf/lecture_ford.ppt
LASER = Light Amplification by the Stimulated Emission of Radiation

With a population inversion in a gain medium, the LASER converts energy input into a coherent output radiation field. The medium determines the ultimate range of wavelengths that can be emitted, the cavity determines the precise wavelengths that are seen. With high finesse cavities line widths of <1 Hz can be achieved even at optical wavelengths! For these (CW) applications, one and only one cavity mode is selected.
What’s so Special About Femtosecond Lasers?

- **Short optical pulse.**
  - Most energy dissipation and transfer processes occur on time scales longer than 100 fs.
  - Femtosecond laser pulses enable one to excite the species studied “instantly” ($t_{exc} \ll t_{rel}$)
  - Dynamics of the excited state can be monitored with high temporal resolution ($\sim 0.5 \tau_{pulse} \approx 12-50$ fs for most commercial lasers)
  - **Visualization of ultrafast dynamics (fluorescence, excited state absorption)**
What’s so Special About Femtosecond Lasers?

- **High peak power of the light**
  - $I \sim J/\tau_{\text{pulse}}$, $I$ – Power, $J$ – pulse energy.
  - 1 mJ pulse with 10 ns duration - 0.1 MW
  - 1 mJ pulse with 100 fs duration - 10 GW
- **Non-linear spectroscopy and materials processing**
  - (e.g., multi-photon absorption, optical harmonics generation, materials ablation, and, as we’ll see, imaging/microscopy)

How to Prepare a Femtosecond Pulse I

Femtosecond laser pulses are usually *Fourier transform-limited pulses*

\[ \Delta \omega \cdot \Delta t \approx 2\pi \quad \Rightarrow \quad \Delta \omega \approx \frac{2\pi}{\Delta t} \]

Large spectral bandwidth for short pulses

\[ \Delta \lambda \approx \frac{\lambda^2}{c \Delta t} \quad \Rightarrow \quad \Delta \lambda \approx 21 \text{ nm for 100 fs pulses with } \lambda_0 = 800 \text{ nm} \]

The need for large bandwidth limits the choice of the laser active medium (broad-band materials only, e.g., Ti:Sapphire, laser dyes) & laser cavity design (no bandwidth limiting elements, such as narrowband mirrors)
How to Prepare a Femtosecond Pulse II

Laser mode – combination of frequency ($\omega$) and direction ($\mathbf{k}$) of the electromagnetic wave allowed by the laser cavity geometry.

The spectrum of laser modes is not continuous $\lambda_n = 2L/n$

$I(t) = \sum_{n=0}^{N} A_n \sin(\omega_n t + \varphi_n)$ Laser pulse as a sum of modes

Relative phase of the modes has to be constant (locked) in order to obtain a stable output pulse

Lock

No lock

Time
How to Prepare a Femtosecond Pulse III

Laser mode – combination of frequency ($\omega$) and direction ($\mathbf{k}$) of the electromagnetic wave allowed by the laser cavity geometry.

$$I(t) = \sum_{n=0}^{N} A_n \sin(\omega_n t + \varphi_n)$$

Relative phase of the modes has to be constant (locked) in order to obtain a stable train of output pulses (at the cavity repetition rate=1/cavity frequency spacing).
Passive Mode-Locking

Saturating absorber technique – Slow, hard to produce <ps pulse widths.

![Diagram showing Passive Mode-Locking setup with mirrors, active medium, saturating absorber, and absorption (cavity losses) graph.]

- Initial noise “seed”
- Steady-state operation
Passive Mode-Locking II

Kerr-lens mode-locking

• Kerr’s effect – intensity-dependent index of refraction: \( n = n_0 + n_2 I \)
• The e/m field inside the laser cavity has Gaussian distribution of intensity which creates similar distribution of the refractive index.
• High-intensity beam is self-focused by the photoinduced lens.

• High-intensity modes have smaller cross-section and are less lossy. Thus, Kerr-lens is similar to a saturating absorber!
• Some lasing materials (e.g. Ti:Sapphire) can act as Kerr-media
• Kerr’s effect is much faster than saturating absorber allowing one generate very short pulses (~5 fs).
Group Velocity Dispersion (GVD)

Optical pulse in a transparent medium stretches because of GVD

- $v = c / n$ – speed of light in a medium
- $n$ – depends on wavelength, $dn/d\lambda < 0$ – normal dispersion

Because of GVD, red components (longer wavelengths) of the pulse propagate faster than blue components (shorter wavelengths) leading to pulse stretching (aka “chirp”).

- Uncompensated GVD makes fs laser operation impossible
- GVD can be compensated by material with abnormal dispersion

Electric field of chirped (non-transform limited) pulse
GVD Compensation

GVD can be compensated if optical pathlength is different for “blue” and “red” components of the pulse.

If OR + RR’ > OB, GVD < 0

“Red” component of the pulse propagates in glass more than the “blue” one and has longer optical path (n x L).
Typical fs Oscillator

**Typical Ti:Sapphire fs Oscillator Layout**

- Tuning range 690-1050 nm
- Pulse duration > 5 fs (typically 50 -100 fs)
- Pulse energy < 10 nJ
- Repetition rate 40 – 1000 MHz (determined by the cavity length)
- Pump source:
  - Ar-ion laser (488+514 nm)
  - DPSS CW YAG laser (532 nm)
- Typical applications:
  - time-resolved emission studies,
  - multi-photon absorption spectroscopy and imaging

CIT Ti:Sapphire fs Oscillator(s)

Ti$^{3+}$ (d$^1$) $t_{2g} - e_g$ Transition
$\tau(e_g) \sim 3$ microseconds
Rep rate $\sim 100$ MHz – 1 GHz
$<$Power$> \sim 100$ – 1000 mW
Pulse length $\sim 10$ – 100 fs
Frequency Conversion of fs Pulses

With fs pulses non-linear optical processes are very efficient due to high intensity of input light: $I_{out} = A I_{in}^m$

**Parametric down-conversion**

- Pump: 800 nm, 1mJ, 100 fs
- Signal: 1100 - 1600 nm, 0.12 mJ
- Idler: 1600 – 3000 nm, 0.08 mJ

**Optical harmonic generation**

**Second harmonic**

$1/\lambda_{SH} = 2/\lambda_F$

$k_{SH} = 2 k_F$

- Pump: 800 nm, 1mJ, 100 fs
- SHG: 400 nm, 0.2 mJ

Harmonic generation can be used to upconvert signal or idler into the visible range of spectrum
Femtosecond Continuum

White-light continuum generation

- Self-focusing and self-phase-modulation broadens the spectrum
- Extremely broad-band, ultrafast pulses (Vis and IR ranges)
- Strongly chirped

Two-Photon Absorption

\[ \Delta I = -\gamma I_1 I_2 \Delta x \]
\[ \gamma = \beta c \]

\[ \Delta I = -\gamma I^2 \Delta x \]

Degenerate case

\[ \beta - \text{TPA cross-section, } c - \text{concentration of material} \]

**1PA**

\[ \frac{dI}{dx} = -\sigma c I \]

Beer’s Law

\[ I(x) = I_0 \exp(-\sigma cx) \]

\[ T = \exp(-\sigma cx) \]

**TPA**

\[ \frac{dI}{dx} = -\beta c I^2 = -(\beta c I) I \]

\[ I(x) = \frac{I_0}{1 + \beta c I_0 x} \]

\[ T = \frac{1}{1 + \beta c x I_0} \]
TPA Cross-Section Units

\[ [\beta cI_0 x] = 1 \quad \rightarrow \quad [\beta] = \left[ \frac{1}{cI_0 x} \right] = \frac{cm^3 \cdot s \cdot cm^2}{phot} \cdot \frac{1}{cm} = \frac{cm^4 \cdot s}{phot} \]

10^-50 cm^4 \cdot s / phot = 1 GM

Typical TPA absorption cross-section is 1 - 10 GM

Do We Really Need a fs Pulse?

\[ \frac{\Delta I}{I} \geq 10^{-5} \quad \text{Accuracy limit of the most of intensity measurements} \]

If beam diameter is 10 μm, required lasers power/pulse energy is:

- CW laser power 12000W
- YAG:Nd laser (10 ns pulse, 25 Hz rep. rate) 120 μJ pulse energy (3 mW)
- Ti:Sapphire laser (100 fs pulse, 100 MHz rep. rate), 1.2 nJ pulse energy (120 mW)
TPA PL excitation

**Pros:**
- Very sensitive
- Easy to setup
- Works without amplifier

**Cons:**
- Works only for PL emitting materials
- Not absolute (requires reference material)
TPA PLE II

\[ I_{PL} = A \cdot \frac{\beta cxI^2}{1 + \beta cxI} \cdot \eta_{PL} \], if \( \beta cxI \ll 1 \), then \( I_{PL} \approx A \beta cxI^2 \eta_{PL} \)

\( \beta \) – TPA cross-section, \( c \) – concentration, \( x \) – length of interaction, \( I \) – laser light intensity, \( A \) – geometrical factor (usually unknown)

TPA PL technique requires a reference measurement

\[ \beta = \beta_{ref} \frac{c_{ref} \eta_{PL}^{ref}}{c \eta_{PL}} \cdot \frac{I^2}{I_{ref}^2} B \quad B = \frac{n^2}{n_{ref}^2} \] for collimated beams

Good reference materials: laser dyes (Fluorescein, Rhodamin, Coumarin)

TPA Imaging

Single photon imaging

$\sim \lambda$

Two photon imaging (works even under the surface!)

$\sim \lambda/2$

Water spectrum
TPA Imaging

Single photon imaging

Two photon imaging (works even under the surface!)
Pump-Probe Experiments I

Idea of the experiment

Before excitation

After

$\Delta \alpha$

$\Delta t$
Pump-Probe Experiments II

• PPE enable one to trace the relaxation dynamics with sub-100 fs resolution
• Types of the data generated by PPE: time-resolved absorption spectra and absorption transients at a certain wavelength.
• Numerous combinations of pump and probe beams are possible (UV pump + visible probe, UV-pump+continuum probe, etc.)
• High pump intensities are required in order to produce noticeable change in the optical absorption of the sample (GW/cm$^2$ – TW/cm$^2$) (Ti:Sapphire amplifiers are generally required)
• Interpretation of the data is sometimes complicated
Pump-Probe Experiments III

- Detects $10^{-5}$ transmission change
- PPE spectra can be chirp-corrected during the experiment
- Use of continuum as a probe enables one to cover the entire visible and NIR ranges

Solvent Reorganization Dynamics

Coumarin 153

Solvent must *adjust* to the large charge separation in the excited state! This happens much faster than fluorescence.