How to deal with femtosecond pulses
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I – Manipulation
Introduction: time-frequency travel

Keeping ultrashort pulses ultrashort

Self-phase modulation – the enemy within

Mirror mirror

Producing circularly polarized pulses – a perfect circle

Focus
The spectral width of a light source defines its **coherence time** \( \tau_c = 1/\Delta \nu \) (coherence = ability to produce interferences)

**Measurement:** linear (field) autocorrelation = Fourier transform spectroscopy
The spectral width of a light source defines its **coherence time** \( \tau_c = 1/\Delta \nu \) (coherence = ability to produce interferences)

Measurement: linear (field) autocorrelation = Fourier transform spectroscopy

An ultrashort light pulse necessarily has an ultrashort coherence time, and thus a **broad spectrum**

This is a necessary, but not sufficient, condition

The temporal profile is obtained by FT the **complex** spectrum
Definitions:

\[ E(t) = |E(t)| e^{i\varphi(t)} \]

\[ \mathcal{F} [E(\omega)] = \int E(\omega) e^{-i\omega t} d\omega = E(t) \]

\[ E(\omega) = |E(\omega)| e^{i\varphi(\omega)} \]

---

Real Gaussian function

\[ I(t) = I_0 \exp \left( -\frac{4 \ln(2) t^2}{\Delta t^2} \right) \]

\[ I(\omega) = |\mathcal{F} [E(t)]|^2 \propto I_0 \exp \left( -\frac{4 \ln(2) \omega^2}{\Delta \omega^2} \right) \]

\[ \Delta \omega \Delta t = 4 \ln(2) \approx 2.77 \]

\[ \Delta t \quad \text{Full Width at Half Maximum} \]

\[ \Delta \omega \quad \text{FWHM} \]
Definitions:

$$E(t) = |E(t)| e^{i\varphi(t)}$$

$$E(\omega) = |E(\omega)| e^{i\varphi(\omega)}$$

$$\mathcal{F} [E(\omega)] = \int E(\omega) e^{-i\omega t} d\omega = E(t)$$

Real Gaussian function

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$$\Delta \omega \Delta t = 4 \ln(2) \approx 2.77$$

Effect of a linear phase

$$\mathcal{F} [E(\omega)e^{i\omega t_0}] = \mathcal{F} [E(\omega)] \ast \mathcal{F} [e^{i\omega t_0}] = E(t)\delta_{t_0} = E(t - t_0)$$

$$\rightarrow$$ temporal shift

Group delay

$$\tau_g(\omega) = \frac{\partial \varphi(\omega)}{\partial \omega}$$

Linear spectral phase \(\omega \tau_0 \rightarrow\) group delay \(\tau_g = \tau_0\)
Fourier transform

Fourier transform

Spectrum FWHM = 40.0 nm

FWHM = 23.4 fs

Spectrum FWHM = 80.0 nm

FWHM = 11.8 fs

Electric field

Electric field
Fourier transform of a Gaussian

Constant spectral phase
Shortest pulse
= Fourier Limit
\[ \Delta \omega \Delta t = 4 \ln(2) \approx 2.77 \]

Quadratic spectral phase:
Gaussian pulse, but stretched
The group delay varies linearly with frequency
\[ \tau_g(\omega) = \frac{\partial \varphi(\omega)}{\partial \omega} \]
(and the instantaneous frequency varies linearly in time)
→ linear chirp

Cubic spectral phase:
Non gaussian pulse, asimetric, with pre or post pulses
(temporal interference
of the red and blue wings of the spectrum)

Random spectral phase:
No ultrashort pulse
Time-frequency representations

A Fourier-limited Gaussian narrowband pulse and a chirped broadband Gaussian pulse can have the same intensity profile.

The temporal/spectral phase may not be the most intuitive representation

→ Time frequency distributions: spectrogram

Goal: resolve temporally the evolution of the spectrum of a signal

→ Principle of the music score

Time-frequency representations are common in acoustics
Time-frequency representations

A sonogram using Sonic Visualiser
Track: Ultra Heat Treated, by Slugabed
Time-frequency representations

A sonogram using Sonic Visualiser
Track: Ultra Heat Treated, by Slugabed

Cut a temporal slice of the signal
Fourier transform it to obtain the spectrum
Time-frequency representations

A sonogram using Sonic Visualiser
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Cut a temporal slice of the signal
Fourier transform it to obtain the spectrum
A sonogram using Sonic Visualiser
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Move the center of the temporal window → Get a spectrum as a function of time
Time-frequency representations

A sonogram using Sonic Visualiser
Track: Ultra Heat Treated, by Slugabed

Crucial parameter: duration of the window

Long temporal window $\rightarrow$ Good spectral resolution but bad temporal resolution
Time-frequency representations

A sonogram using Sonic Visualiser
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Crucial parameter: duration of the window

Long temporal window → Good spectral resolution but bad temporal resolution
Short temporal window → Good temporal resolution but bad spectral resolution
Time-frequency representations

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Crucial parameter: duration of the window

Long temporal window → Good spectral resolution but bad temporal resolution
Short temporal window → Good temporal resolution but bad spectral resolution
Optimal window → Reveals the temporal evolution of the spectrum
Time-frequency representations

A sonogram using Sonic Visualiser
Track: Ultra Heat Treated, by Slugabed

Crucial parameter: duration of the window

Electronic Voice (tunable harmonics)

Electronic

Frequency (Hz)

Long temporal window → Good spectral resolution but bad temporal resolution
Short temporal window → Good temporal resolution but bad spectral resolution
Optimal window → Reveals the temporal evolution of the spectrum
Gabor Analysis: building a spectrogram

\[ S(\omega, \tau) = \left| \int E(t)G(t - \tau)e^{i\omega t} dt \right|^2 \]

Define Gaussian gate function \( G \)
Calculate the spectrum of the gated signal
Slide the gate

Example: Spectrogram of two delayed Fourier Limited pulses
Time-frequency representations of femtosecond pulses

Wigner distribution

\[ W(\omega, \tau) = \int E(t + \tau/2)E^*(t - \tau/2)e^{i\omega t} dt \]

Marginals:

\[ I(t) = \int W(\omega, \tau) d\omega \]

\[ I(\omega) = \int W(\omega, \tau) d\tau \]

Double pulse

Interference Between pulses
Time-frequency representations of femtosecond pulses
A newcomer’s guide to ultrashort pulse shaping and characterization

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Introduction: time-frequency travel

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Focus
A typical femtosecond experimental setup

Propagation in various media (air, glass...)
Reflections on mirrors
Polarization manipulation
Focusing by lenses, mirrors
Entrance windows to vacuum chambers

→ All of this can bring trouble, in particular stretch your pulses

This is an issue if:
- you need time resolution
- you need high intensity to drive extreme processes
- you need high intensity for multiphoton microscopy
- you simply want ultrashort pulses to remain ultrashort because it’s so satisfying (and you paid for that).
Dispersion → group velocity dispersion

The different frequency components travel at different speeds

Effect on fs pulse?

→ Get the dispersion formula \( n(\omega) \)

and calculate the accumulated spectral phase:

\[
\varphi(\omega) = n(\omega) l \omega / c
\]

over propagation distance \( l \)

Derivative of the spectral phase = group delay

Linear spectral phase: constant group delay

Quadratic spectral phase: group delay dispersion (GDD, \( \text{fs}^2 \))

\[
GDD = \frac{d^2 \varphi}{d\omega^2}
\]
Propagation in air

\[ E_{in}(t) \rightarrow E_{in}(\omega) \rightarrow E_{out}(\omega) = E_{in}(\omega) \exp(i\varphi(\omega)) \rightarrow E_{out}(t) \]
300 fs pulse at 1030 nm from Yb amplifier

After 100 m

Propagation in air

Time (s)

Angular frequency (rad s⁻¹)

z distance (m)

FWHM = 300.06 fs

After 1.00e+02 m of air

Initial pulse, fwhm=300.0 fs
Propagation in air

25 fs pulse at 800 nm from Ti:Sa amplifier

After 100 m
Propagation in air

5 fs pulse at 700 nm from postcompressed Ti:Sa amplifier

After 1m
$$n^2 - 1 = \frac{0.6961663\lambda^2}{\lambda^2 - 0.0684043^2} + \frac{0.4079426\lambda^2}{\lambda^2 - 0.1162414^2} + \frac{0.8974794\lambda^2}{\lambda^2 - 9.896161^2}$$
Propagation in fused silica

\[ E_{\text{in}}(t) \rightarrow E_{\text{in}}(\omega) \rightarrow E_{\text{out}}(\omega) = E_{\text{in}}(\omega) \exp(i\varphi(\omega)) \rightarrow E_{\text{out}}(t) \]
Propagation of 800nm pulse through 20mm of SiO2
Propagation in fused silica

- 300 fs  
  1030 nm  
  Up to 10 cm

- 25 fs  
  800 nm  
  Up to 5 mm

- 5 fs  
  700 nm  
  Up to 0.5 mm

Mostly linear spreading of the frequency components in time

→ Linear chirp, quadratic phase

Can be compensated by introducing an opposite quadratic phase in the laser
Dispersion compensation

\[ E_{\text{in}}(t) \rightarrow E_{\text{in}}(\omega) \rightarrow E_{\text{out}}(\omega) = E_{\text{in}}(\omega) \exp(i\varphi(\omega)) \rightarrow E_{\text{out}}(t) \]

\[ E_{\text{comp}}(\omega) = E_{\text{out}}(t) \exp(-i \alpha (\omega - \omega_0)^2) \rightarrow E_{\text{comp}}(t) \]

\[ \alpha = \text{quadratic spectral phase coefficient} \]
Dispersion compensation

Small residual deviation from Fourier Limit: higher order spectral phase, uncompensated in the compressor.
Clear signature of third order spectral phase
→ For very short pulses, we need better compensation
Multilayer dielectric coatings
Designed to optimize broadband reflectivity and introduce a well controlled Group Delay Dispersion
Chirped mirrors compensating the dispersion of fused silica can be bought
Chirped mirrors

Good average dispersion compensation

Oscillations in the GDD → Will create replicas in the temporal profile of the beam. This is bad.
Chirped mirrors

Good average dispersion compensation
Oscillations in the GDD → Will create replicas in the temporal profile of the beam. This is bad.
Avoided by matching two mirrors (different designs or different angles of incidence)

Chirped Mirror Set for Multiphoton Microscopy, 53.0 mm x 12.0 mm x 12.0 mm

- >99% Average Reflectance from 700 to 1000 nm
- Group Delay Dispersion (GDD) per Reflection: -175 fs² at 800 nm
- Coated Surface Dimensions: 50 mm x 8 mm
- 8° AOI
- Designed for P-polarized light
- Sold in Packs of 2

The DCM17S consists of a pair of reflective optics with >99% average reflectance over the 700 - 1000 nm wavelength range. These mirrors are designed to integrate with multiphoton microscopy setups, which typically include long path lengths through highly dispersive glass. The 8° AOI allows these mirrors to perform similarly for both s- and p-polarized light, and is ideal for a compact setup where multiple reflections are needed.

Mounting Option
As shown in the figure to the right, these mirrors can be mounted on the Kinematic Grating Mount Adapter, which is compatible with Ø1", front-loading, unthreaded mirror mounts, such as our Polaris UltraStable Kinematic Mirror Mount.

<table>
<thead>
<tr>
<th>+1</th>
<th>Quantité</th>
<th>Docs</th>
<th>Produit - Universel</th>
<th>Total HT</th>
<th>Disponibilité</th>
</tr>
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<tbody>
<tr>
<td>+1</td>
<td>DCMP17S</td>
<td></td>
<td>Dispersion-Compensating Mirror Set, 700 nm - 1000 nm, 8° AOI, Qty. 2</td>
<td>€ 2,424.68</td>
<td>5-8 Days</td>
</tr>
</tbody>
</table>
Good average dispersion compensation
Oscillations in the GDD → Will create replicas in the temporal profile of the beam. This is bad.
\textit{Avoided by matching two mirrors (different designs or different angles of incidence)}
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Focus
High intensity $\rightarrow$ Non-linear dispersion

Kerr effect: the refractive index is modulated by the laser intensity:

$$n(x,y,z,t) = n_0 + n_2 . I(x,y,z,t)$$

Already seen in Adeline’s talk: Kerr lens modelocking – spatial effect

Already seen in Clara’s talk: spectral/temporal effect
Self phase modulation

What is the effect of a Gaussian phase on a Gaussian pulse?

\[ E_{in}(t) \rightarrow E_{out}(t) = E_{in}(t) \exp(i \sigma l_{in}(t)) \rightarrow E_{out}(\omega) \]

\( \sigma = \) magnitude factor of the SPM
What is the effect of a Gaussian phase on a Gaussian pulse?

Parabolic approximation → quadratic temporal phase → spectral broadening

\[ \alpha = 9.8 \times 10^{-1} \]

**Pulse with Gaussian phase**

**FT limited**

**Modified spectrum by SPM**

**Initial spectrum**

**Wigner for \( \alpha = 9.8 \times 10^{-1} \)**

**Angular frequency (rad s\(^{-1}\))**

**Time (s)**
What is the effect of a Gaussian phase on a Gaussian pulse?

Parabolic approximation → quadratic temporal phase → spectral broadening

Strong SPM: fringes appear in the spectrum
When does SPM become a problem?

$$B = \frac{2\pi}{\lambda} \int n_2 I(z) \, dz$$

= Accumulated non-linear phase shift

Depends on the laser intensity
   Energy per pulse, pulse duration, beam diameter

Is accumulated along the whole beam path
   Air, glass windows, waveplates, lenses...

Can be avoided
   - by increasing the beam diameter
   - by shortening the optical path and avoiding transmissions
   - by stretching the pulses to propagate them, and compress them as late as possible
     (for instance in postcompression, do the final compression under vacuum to avoid SPM in windows)

SPM depends $x,y,z,t \rightarrow$ inhomogeneities in the beam
   The spectrum depends on position
   The spectral phase depends on position
   $\rightarrow$ The dispersion affects differently the various parts of the beam
   $\rightarrow$ The temporal profile becomes inhomogeneous
   $+$ spatial phase $\rightarrow$ focusing
When does SPM become a problem?

A few examples:
- Yb: fiber laser, waist=2mm, 300 fs, 500 μJ
  → $I_{\text{max}} = 0.44 \text{ GW/cm}^2 \rightarrow B=5$ in 7m of SiO2

- Ti:Sa laser, waist=20mm, 25 fs, 10 mJ
  → $I_{\text{max}} = 240 \text{ GW/cm}^2 \rightarrow B=5$ in 10 mm of SiO2

- Postcompressed pulse, waist = 10 mm, 5fs, 3mJ
  → $I_{\text{max}} = 1350 \text{ GW/cm}^2 \rightarrow B=5$ in 1.7 mm of SiO2
SPM induces spectral broadening → reduces the Fourier Limit duration that can be reached

Can we compensate the spectral phase?

Parabolic approximation of the phase → can be compensated by opposite second order phase

How can we introduce a negative chirp?

Mid Infrared: use negative GVD (eg in fused silica)

UV-Vis-IR: use chirped mirrors
Let's introduce a negative chirp to compensate the quadratic phase of our SPModulated pulse.

-175 fs² / bounce. How does it compare with the phase introduced by SPM?
Using SPM to compress fs pulses

Introduce a quadratic spectral phase to compensate for the positive chirp of the pulses

\[ E_{\text{in}}(t) \rightarrow E_{\text{out}}(t) = E_{\text{in}}(t) \exp(i \sigma l_{\text{in}}(t)) \rightarrow E_{\text{out}}(\omega) \]

\[ E_{\text{comp}}(\omega) = E_{\text{out}}(\omega) \exp(i \alpha (\omega - \omega_0)^2) \rightarrow E_{\text{comp}}(t) \]
Using SPM to compress fs pulses

Increasing quadratic phase compensation

Note:

Characterization of ultrashort electromagnetic pulses

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\begin{equation}
W_{\text{OUTPUT}}(t,\omega) = W_{\text{INPUT}}(t - \frac{\phi^{(2)}(\omega,\omega)}{2}).
\end{equation}

This corresponds to a shear of the chronocyclical Wigner function, as shown in Fig. 11(b), which encodes the spectrum of the input pulse onto the temporal intensity of the output pulse.
Postcompression in a bulk plate

2.3-cycle mid-infrared pulses from hybrid thin-plate post-compression at 7 W average power


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Why using two plates (Si and YAG)?

Efficient spectral broadening from a single bulk plate is limited by plasma formation due to ionization, which results in nonlinear losses and degradation of the beam profile at higher input intensities [18]. To overcome the limitations of single-plate compression of MIR pulses, different material thin plates of opposite group velocity dispersion (GVD) could be employed in alternating order, in a hybrid setup. Having the appropriate parameters for these plates, such as n2, GVD and thickness, may allow to compensate the spectral phase (up to the second order phase) on the subsequent plate, resulting in sufficient intensity to drive the nonlinear broadening efficiently.

Semiconductors: positive GVD at 3.2 μm, high n2

Semiconductors as silicon and germanium were the prime candidates as they have the highest nonlinear refractive index of 3.79·10⁻¹⁴ cm²/W and 3.68·10⁻¹³ cm²/W respectively [22].

Dielectrics: negative GVD at 3.2 μm, but low n2:

YAG has the highest nonlinearity of 7·10⁻¹⁶ cm²/W.

Fig. 1. Schematic view of the experimental arrangement. SM1 and SM2 are concave spherical mirrors.
Temporal characterization (see next course)

FROG

(a) FWHM = 22.8 fs

Efficient postcompression scheme
Inherent spatial inhomogeneity due to the intensity distribution in the plate? Can be mitigated by using multiple plates
Postcompression in bulk plates – more references

Compression of high-power optical pulses
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Greater than 50 times compression of 1030 nm Yb:KGW laser pulses to single-cycle duration
Chih-Hsuan Lu,1,2,5 Wei-Hsin Wu,1 Shiang-He Kuo,1 Jhan-Yu Guo,1 Ming-Chang Chen,1,3,4 Shiang-Da Yang,1, and A. H. Kung1,2,6
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Generation of intense supercontinuum in condensed media
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Efficient nonlinear compression of a mode-locked thin-disk oscillator to 27 fs at 98 W average power
Chih-Lun Tsai,1 Frank Meyer,2 Alan OMalley,2 Yicheng Wang,2 An-Yuan Liang,3 Chih-Hsuan Lu,1 Martin Hoffmann,2 Shiang-Da Yang,1 and Clara J. Saraceno2
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Generation of a single-cycle pulse using a two-stage compressor and its temporal characterization using a tunnelling ionization method
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Wavelength (nm)
Intensity (a.u.)

Fig. 5. Homogeneity measurement performed after multiple-pass stage supporting sub-30 fs. Dashed lines indicate 1/e² level of intensity.
**Postcompression in a hollow core fiber**

Generation of high power sub-15 fs pulses at 515 nm through nonlinear compression of an Yb-doped ultrafast fiber amplifier

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**Propagation in hollow-core fiber**: homogeneous effect of SPM  
Handwaving: Propagation mixes the different parts of the beam, leading to homogeneity

**Nonlinear medium?** Rare gas introduced at controlled pressure, ~ changing medium optical thickness

**Recompression:**  
Set of chirped mirrors. 18 bounces $\rightarrow -1000$ fs$^2$  
Additional silica plates $\rightarrow$ fine tune the dispersion, adding positive GDD.  
Optimal dispersion: -$850$fs$^2$
Postcompression in a hollow core fiber

Generation of high power sub-15 fs pulses at 515 nm through nonlinear compression of an Yb-doped ultrafast fiber amplifier

DOMINIQUE DESCAMPS,1* FLORENT GUICHARD,2 STÉPHANE PETIT,1 SANDRA BEAUVALET, ANTOINE COMBY,1 LOIC LAVENU,2 AND YOANN ZAOUTER,2

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Received XX Month XXXX; revised XX Month XXXX; accepted XX Month XXXX; posted XX Month XXXX (Doc. ID XXXXXX); published XX Month XXXX

Spectral measurements

Efficient postcompression scheme: 15fs, 20µJ, 500kHz (10W)

Some losses due to coupling to higher order modes in the capillary. Transmission = 78%

Temporal characterization (see next course)

20 µJ/pulse 10W average power
Postcompression in a hollow core fiber – more references

Generation of high energy 10 fs pulses by a new pulse compression technique

M. Nisoli, S. De Silvestri, and O. Svelto
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53 W average power few-cycle fiber laser system generating soft x rays up to the water window

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Fig. 1. Experimental setup of the two nonlinear compression stages that reduce the pulse duration from the initial 210 to 30 fs and 7.8 fs.

Memorandum

Vol. 6, No. 11 / November 2019 / Optica

Generation of three-cycle multi-millijoule laser pulses at 318 W average power

Tamás Nagy,1,a,1 Steffen Hädrich,1,2,3,5 Peter Simon,1,3,5 Andreas Blumenstein,1 Nico Walther,2 Robert Kläsi,6 Joachim Boldt,6 Henning Stark,4 Sven Breitenkopf,2 Peter Joujal,4 Imre Sérés,4 Zoltán Varallyay4,5 Tino Eidam,1 and Jens Limpert2,4,5,6

Fig. 1. Experimental layout: F-CPA, fiber chirped pulse amplifier; HCF, stretched hollow-core fiber; d-scan, dispersion scan device; PM, water-cooled power meter; 4D PSD, position sensitive detectors for near and far field; TFP, thin-film polarizer; J/2, half-wave plate; CCD, camera.
Propagation in cavity: homogeneous effect of SPM
Handwaving: inhomogeneities washed out by mode propagation in the cavity
34 roundtrips. Total distance 20 m

Nonlinear medium? 7 bars of argon. This gas pressure results in a nonlinear index
\[ n_2 = 6.5 \times 10^{-23} \text{ m}^2/\text{W} \] and a group velocity dispersion \( \beta_2 = 110 \text{ fs}^2/\text{m} \) [17].

Recompression:
Gires Tournoi Interferometer mirrors \( \rightarrow -250 \text{ fs}^2 \) per bounce + \(-100\text{fs}^2 \) per bounce
Optimal dispersion: \(-3100\text{fs}^2\)

What about spatial homogeneity of the beam?
Postcompression in a multipass cell

Overlap integral of the spectrum at location with the spectrum at center >90% wherever intensity >10%

Fig. 4. Spatio-spectral couplings at the output of the cavity in the vertical axis. (a) Experimental spectro-imaging measurement. (b) Simulated spectro-imaging measurement. (c) Spatio-spectral quality factor measured with a spectro-imaging setup: vertical axis (red), horizontal axis (blue).

Good spatial homogeneity of the spectrum
Nonlinear pulse compression in a multi-pass cell

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2Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Straße 1, 85748 Garching, Germany

**Postcompression in a multipass cell – more references**

Let us assume that the image contains text in Optics Letters and Optics Express journals. The text appears to be related to optical nonlinearities, pulse compression, and laser systems. The diagrams illustrate various optical setups, including nonlinear pulse compression in a multi-pass cell and spectral broadening in a multipass cell, with references to specific experimental setups and results.

**Nonlinear pulse compression in a multi-pass cell**

- Yb-fiber-based oscillator
  - 0.86 W, 10 MHz, 600fs
- Two-stage Yb-innoslab amplifier
  - 580 W, 850 fs, M² = 1.35 x 1.95
- Pointing stabilization & spatial filtering
  - 530 W, 850 fs, M² = 1.15 x 1.05
- Herriott-type MPC
  - 18 reflections per mirror
- Collimation
  - 2 spherical lenses
- Mirror compressor
  - 3 x (10^10) fs²
- Diagnostics
  - Power
  - Spectrum
  - Autocorrelation
  - Beam quality

**Postcompression of picosecond pulses into the few-cycle regime**

**Spectral broadening of 112 mJ, 1.3 ps pulses at 5 kHz in a LG10 multipass cell with compressibility to 37 fs**

**Let us consider the multiple references and diagrams provided.**

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**Notes:**
- The text and diagrams are related to advanced optical technologies and have been extracted from Optics Letters and Optics Express journals.
- The content includes theoretical and experimental aspects of nonlinear pulse compression and spectral broadening in multipass cells.
- Diagrams illustrate various optical configurations and components used in the experiments.
Recap on postcompression techniques

High-energy few-cycle pulses: post-compression techniques
Tamas Nagy, Peter Simon, and Laszlo Veisz

HC-PCF: Hollow Core Photonic Crystal Fiber
HCF: Hollow Core Fiber
SF-HCF: Stretched Hollow Core Fiber

Figure 10. Pulse energy versus pulse duration in compression experiments in the near-infrared range. The red-shaded area in represents the high-energy few-cycle regime.
Resolving the attosecond dynamics of the Kerr effect

Attosecond nonlinear polarization and light–matter energy transfer in solids

A. Sommer1, E. M. Bothschaft2, S. A. Sato3, C. Jakubet1, T. Latka1, O. Razskazovskaya1, H. Fattahi1, M. Jobst1, W. Schweinberger1, V. Shirvanyan1, V. S. Yakovlev1,2, R. Kienberger1, K. Yabana1, K. N. Karpowicz2, M. Schultze3, & F. Krausz1,2

Figure 2 | Sub-femtosecond-resolved optical Kerr effect in silica.

a. After passage through a 10-μm-thick fused silica sample, the electric field $E(t)$ of the few-cycle near-infrared pulse with a peak intensity of $1.3 \times 10^{14}$ W cm$^{-2}$, approximately 10% below the threshold for optical damage, is modulated as a result of the nonlinear light–matter interaction, as revealed by its comparison to a low-intensity ($I_{\text{peak}} = 7 \times 10^{12}$ W cm$^{-2}$) reference waveform $E_{\text{ref}}(t)$ (for $\beta = 0.27$). This comparison yields a transient positive phase shift induced by the strong field, as anticipated from the dynamic increase of the refractive index owing to the optical Kerr effect. The two insets show close-ups of the comparison near the centre and at the end of the pulse, revealing the full reversibility of the effect. $E(t)$ and $E_{\text{ref}}(t)$ are obtained from averaging a set of three recordings performed under identical conditions on individual samples. b. The phase shift $\Delta \phi_{\text{peak}}$ evaluated at the peak of the field envelope for different peak intensities $I_{\text{peak}}$ of $E(t)$ is found to exhibit a linear dependence on the field intensity. Each data point represents the mean value of three individual recordings under identical conditions; the error bars indicate the standard deviation.
Introduction: time-frequency travel

Keeping ultrashort pulses ultrashort

Self-phase modulation – the enemy within

Mirror mirror

Producing circularly polarized pulses – a perfect circle

Focus
Why bother about mirrors?

Ubiquitous in experimental setups

We’ve seen that chirped-mirrors could induce strong dispersion

What about “regular” mirrors?
Getting the dispersion of metallic mirrors

The complex refractive index gives the complex reflection coefficients through Fresnel equations.

\[ R_s = \left| \frac{n_1 \cos \theta_i - n_2 \cos \theta_t}{n_1 \cos \theta_i + n_2 \cos \theta_t} \right|^2 = \frac{n_1 \cos \theta_i - n_2 \sqrt{1 - \left( \frac{n_1}{n_2} \sin \theta_i \right)^2}}{n_1 \cos \theta_i + n_2 \sqrt{1 - \left( \frac{n_1}{n_2} \sin \theta_i \right)^2}}^2, \]

\[ R_p = \left| \frac{n_1 \cos \theta_t - n_2 \cos \theta_i}{n_1 \cos \theta_t + n_2 \cos \theta_i} \right|^2 = \frac{n_1 \sqrt{1 - \left( \frac{n_1}{n_2} \sin \theta_i \right)^2} - n_2 \cos \theta_i}{n_1 \sqrt{1 - \left( \frac{n_1}{n_2} \sin \theta_i \right)^2} + n_2 \cos \theta_i}^2. \]

The modulus square gives the reflectivity. The phase of \(\sqrt{R}\) gives the dephasing at reflection.
Silver mirror reflectivity and dephasing

The phase is not linear → GDD!
The spectral phase is defined along the frequency / angular frequency / photon energy coordinate. Not the wavelength.

$$\omega = \frac{2\pi c}{\lambda}$$ → nonlinear mapping between $$\omega$$ and $$\lambda$$
Getting the dispersion of metallic mirrors

Now you can say:
The phase is not linear $\rightarrow$ GDD!
The Protected Silver mirror has a different reflectivity than the bare Ag mirror (but you can’t really buy bare Ag mirrors)

Note the different reflectivity of S and P polarizations
Dip at 600 nm: scary. Probably means high dispersion. Dispersion not provided by company.
Ultrafast-enhanced silver mirrors

Reflectance of Ultrafast-Enhanced Silver, 45° AOI

Group Delay Dispersion of Ultrafast-Enhanced Silver, 45° AOI

Silver Mirror, fs Optimized, 0-45° AOI, 25.4 mm, 600-1000 nm

Silver Mirror, fs Optimized, 0-45° AOI, 25.4 mm, 470-1000 nm
Dielectric multilayer mirrors

68 bounces inside cavity

R=90% → Cell transmission T=0.9^{68}=0.07%
R=95% → T=3%
R=99% → T=50%
R=99.5% → T=71%
R=99.8% → T=87%
R=99.9% → T=93%

The bandwidth of dielectric mirrors can be a limitation in cavity postcompression.
More trouble: polarization state

The mirror reflectivity is defined for S and P polarizations

Reflectance of Ultrafast-Enhanced Silver, 45° AOI

In general, you work with S or P polarization

The beam is polarized at the exit of the laser
It remains in the horizontal plane
It doesn't go through birefringent optics
→ OK
More trouble: polarization state

The mirror reflectivity is defined for S and P polarizations

What if:

- You change beam height between two distant mirrors?
- You change beam height using a periscope?
- You go through a non-linear crystal for frequency conversion?

You may end up with a polarization state which is neither S or P
More trouble: polarization state

The mirror reflectivity is defined for S and P polarizations

Reflectance of Ultrafast-Enhanced Silver, 45° AOI

Let us decompose the input beam along S and P directions

Different reflectivity $\rightarrow$ Rotation of the polarization angle
Phase shift between the two components $\rightarrow$ Introduces some ellipticity in the laser field
Different GDD between the two components $\rightarrow$ Complex temporal evolution of the beam polarization

Can we calculate this?
We send a linearly polarized laser pulse to a set of 45° incidence silver mirror, in a horizontal plane. Mirror complex reflectivity calculated from Fresnel equations. We introduce an initial rotation of the polarization direction with respect to the vertical S polarization. We calculate the resulting pulse polarization, as a function of the number of bounces on mirrors.
We send a linearly polarized laser pulse to a set of 45° incidence silver mirror, in a horizontal plane. Mirror complex reflectivity calculated from Fresnel equations. We introduce an initial rotation of the polarization direction with respect to the vertical S polarization. We calculate the resulting pulse polarization, as a function of the number of bounces on mirrors.

Input
30° off
We send a linearly polarized laser pulse to a set of 45° incidence silver mirror, in a horizontal plane.

Mirror complex reflectivity calculated from Fresnel equations.

We introduce an initial rotation of the polarization direction with respect to the vertical S polarization.

We calculate the resulting pulse polarization, as a function of the number of bounces on mirrors.
We send a linearly polarized laser pulse to a set of 45° incidence silver mirror, in a horizontal plane. Mirror complex reflectivity calculated from Fresnel equations. We introduce an initial rotation of the polarization direction with respect to the vertical S polarization. We calculate the resulting pulse polarization, as a function of the number of bounces on mirrors.
Introduction: time-frequency travel

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Focus
Many experiments require circularly polarized light, or well defined elliptical light.
Circular Dichroisms, attoclock...

In principle, polarization manipulation is easy: use wave plates.

Decompose the field in two components in a birefringent crystal.
They accumulate different phases.
→ The resulting polarization is modified.

Ex: Half Wave Plate
Controlling the polarization state of femto pulses

The crystal thickness can be set to achieve a phase-shift of $\lambda/4$ between the two components
→ Converts linear light into circular

First constraint: we want zero order waveplates
Multiple order quarter waveplate: introduces a delay of $nT_0 + T_0/4$
$n=0 \rightarrow$ Zero-order quarter waveplate

20fs pulse, multiple order wp with $n=5$

20fs pulse, zero order waveplate

(can be useful for temporal shaping of polarization state, e.g. for polarization gating of attosecond pulse generation)
The crystal thickness can be set to achieve a phase-shift of $\lambda/4$ between the two components → Converts linear light into circular

First constraint: we want zero order waveplates
Multiple order quarter waveplate: introduces a delay of $nT_0 + T_0/4$
$n=0 \rightarrow$ Zero-order quarter waveplate

Technology:
- Very thin polymer film (true zero order)
- Quartz: stack two crystals, for instance one introducing $5T_0 + T_0/4$ and the other one $-5T_0$

Second constraint: we sometimes need broadband waveplates
Impossible with a single dispersive birefringent medium ($n_{\text{ordinary}}$ and $n_{\text{extraordinary}}$ vary with $\lambda$)
Use a combination of two media, with opposite dispersions: quartz and MgF$_2$
The bandwidth can be increased by increasing the complexity of the stacking
Which quarter wave plate?

Zero order

Achromatic

Super achromatic

*λ/4 Wavelength variation of the retardation R and the axis direction Ψ*
Controlling the polarization state of femto pulses

Transferring the attoclock technique to velocity map imaging

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Goal: produce an elliptical beam, and rotate the main axis of the ellipse during the acquisition (tomographic Velocity-Map Imaging)

The pulse characterization with a SPIDER resulted in a measured pulse length of 6.1 fs at a central wavelength of 735 nm. Before the pulse entered the vacuum chamber through the entrance window it passed a polarizer, a quarter quarter-wave plate (QWP) and a half-wave plate (HWP). The polarizer (Newport polarcor 05P109AR.16) ensured a clean linear polarization state of the beam before the pulse passes through the QWP. The desired ellipticity of 0.87 was induced by the quarter-wave plate (B.Halle Nachfl. GmbH RAC 5.4.10L)

Check: rotate polarizer → Record Malus’ law
Controlling the polarization state of femto pulses

Transferring the attoclock technique to velocity map imaging

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(C) 2013 OSA 23 September 2013 | Vol. 21, No. 19 | DOI:10.1364/OE.21.021981 | OPTICS EXPRESS 21981

Superachromatic waveplate
Controlling the polarization state of femto pulses

A superachromatic waveplate is necessary for this experiment.
Finding good waveplates can be difficult in DUV-VUV-XUV → Very challenging

Phase-shifts introduced by metallic reflections can be an alternative solution
Polarization by reflections

Calculation: reflection of a 5fs 400 nm pulse on Al mirrors: in the practical
Polarization by reflections

SU5: a calibrated variable-polarization synchrotron radiation beam line in the vacuum-ultraviolet range

Laurent Nahon and Christian Alcaraz

1024 APPLIED OPTICS / Vol. 43, No. 5 / 10 February 2004

Used as analysis QWP for synchrotron radiation

LETTER

https://doi.org/10.1364/AO.45.004024

Light-wave dynamic control of magnetism

Florian Siegrist1,2, Julia A. Geisser3,4, Marcus Ostinander5, Christian Denker5, Yi-Ping Chang6, Malte G. Schröder6, Alexander Guggenmos6,7, Yang Cui6, Jakob Wulofskeit8, Ulrike Martens9, J. K. Dewhurst10, Ulrike Klemm11, Markus Münzenberg12, Sangeeta Sharma13 & Martin Schultze14

Used to convert linear atto pulses to circular
Introduction: time-frequency travel

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Focus
Most straightforward tool: lenses

But:

Glass introduces GDD and SPM
The refractive index depends on wavelength → The focal length depends on wavelength
chromatic aberration
+ spherical aberrations...
While chromatic aberration plays only a small role in the pulse temporal phase, it does become evident, however, in the pulse’s temporal intensity and its distortions. For a lens free of aberrations, the pulse fronts are curved and perfectly symmetrical about the focus, and flat at the focus. Chromatic aberration shifts the position of the flat pulse front to a value of $z$ after the focus, resulting in pulse fronts that are not symmetric about the focus [3]. In Fig. 2, it is clear that the pulse fronts are, in fact, not symmetric about the focus, and the pulse front is flat at $z = 1.5$ mm in both the simulation and experimental data.

**Chromatic aberration and GDD, GDD dominates**
Directly measuring the spatio-temporal electric field of focusing ultrashort pulses

Pamela Bowlan, Pablo Gabilde, and Rick Trebino
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In Fig. 3, most of the color variation is again due to the GDD of the lens. Because the doublet is very thick (9.8 mm) and made of very dispersive glass, it introduces significant GDD, and this lengthens the pulse by about three times more than the aspheric lens does (using rms temporal width of the pulse averaged over x). Also, the pulse fronts are not symmetric about the focus, revealing the presence of chromatic aberration.

No chromatic aberration
GDD dominates

Fig. 3. $E(x,z,t)$ in the focal region of an achromatic doublet designed for visible light. Significant GDD is apparent due to the thickness of the lens. Because this lens was designed for the visible, and not 800 nm, the pulse fronts are not symmetric about the focus, revealing that some chromatic aberration is also present.
Spherical aberration and GDD dominates.

Fig. 4. \( E(x,z,t) \) in the focal region of a plano-convex lens. The spherical aberrations introduced by this lens result in ripples in the spatial profile that are particularly visible at \( z = -0.7 \text{mm} \).
Directly measuring the spatio-temporal electric field of focusing ultrashort pulses

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Pure effect of chromatic aberration → Pulse duration x 1.3 w/r Fourier Limit

30 nm bandwidth pulse
Focused by f=50mm
Plano-convex lens
With chirp compensation

Fig. 5. $E(x,z,t)$ in the focal region of a ZnSe lens with chirp compensation. In these plots, all of the color variation is due to chromatic aberration.
Angular dispersion can be caused by misaligned compressor or wedged components.

Fig. 5. $E(x,z,t)$ in the focal region of the beam that had angular dispersion. The data is displayed in the same way as in Fig. 4. The angular dispersion becomes purely spatial chirp at the focus because a lens is a Fourier transformer.
Focusing femtosecond pulses

30 nm bandwidth pulse with angular dispersion
Focused by f=25mm lens

Different colors sent to different angles → Spatial chirp at focus

Angular dispersion causes pulse front tilt: the femtosecond pulse arrival time depends on space

Generally detrimental, but can be useful – the attosecond lighthouse
The attosecond lighthouse

Applications of ultrafast wavefront rotation in highly nonlinear optics

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Attosecond lighthouse: ultrafast wavefront rotation, sending consecutive attosecond bursts to different directions
Focusing femtosecond pulses

Most straightforward tool: lenses

But:
Glass introduces GVD and SPM
The refractive index depends on wavelength → The focal length depends on wavelength
chromatic aberration

Spherical mirrors are broadly used

But:
Astigmatism is an issue when the incidence angle is too large

Off-axis parabola solve this issue

But:
They are more difficult to align
(large incidence angles also destroy circular polarization...)

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Focus
Many effects can degrade the pulse quality in an experiment
The most obvious issue is the increase of pulse duration
   We’ll see how to characterize this next course

Polarization issues and spatial inhomogeneities / space-time couplings can be tricky

In general one tries to avoid measuring stuff as long as things work
This can be risky and lead you to investigate artifacts believing they are interesting physics