Control of dispersion effects for resonant ultrashort pulses

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- Dispersion distorts the pulse. The sample is excited by a different field.
- A lot of physical and chemical processes depend on pulse temporal shape and phase

Shaping devices are limited:
- Wavelength
- Passive: No Amplification, Cannot create new frequencies

Resonant atomic dispersion and light-shifts may be an alternative
At atomic resonance:

- Gain
- Modification of pulse shape
1. Propagation of ultrashort pulses
2. Direct compensation with a pulse shaper
3. Case of an ultrashort pulse train.
4. Propagation in an atomic system driven by a strong pulse
5. Towards « active » pulse shaping
1. Propagation of ultrashort pulses

a) **Non resonant medium**
   - **Transparent**
   - **Dispersion**:

\[
\phi(\omega) = n(\omega)\omega L/c \\
\phi(\omega) = \phi_0 + \phi'_0(\omega - \omega_L) + \phi''_0(\omega - \omega_L)^2/2
\]

\[
\tau_C = 2\phi''_0/\tau_0 \\
\omega_i = \omega_0 + t/2\phi''_0
\]

\[
\sqrt{|\phi''_0|} \quad \quad \quad \quad \sqrt{|\phi''_0|}
\]
1. Propagation of ultrashort pulses

b) Resonant (two level system)

- Total absorption negligible:
  \[ \Gamma \ll \Delta_D \ll \Delta \omega \]

- Dispersion:
  \[ \phi(\omega) = \frac{\alpha_0 L \Delta_D}{\omega - \omega_0} \]

\( \omega_0 \)

Optical depth

\[ \Delta \omega \sim THz \]

\[ \Delta_D \sim GHz \ll \Delta \omega \]
Rubidium, $4S_{1/2} \rightarrow 4P_{1/2}$, $\tau_0=75\text{fs}$; $\alpha_0|\Delta_d|=3\text{THz}$

**Top Diagram:**
- Real part of the spectrum
- Axes: $\nu - \nu_0$ (THZ)
- Graphs: entrance and exit
- Arrow: $\alpha_0|\Delta_d$

**Bottom Diagram:**
- Electric field (arb. units)
- Axes: time (fs)
- Graphs: entrance and exit
- Range: 0 to 2000 fs
Compensation?

\[ \phi(\omega) \approx \frac{\alpha_0 L \Delta_D}{\omega - \omega_0} \]

Cannot be developed around central laser frequency

Second order no longer representative
All order are involved

Compensation with standard devices
2. Compensation with a pulse shaper

initial pulse  
distorted pulse  
recovered pulse

\[ e^{i\phi_{\text{disp}}} \quad \text{linear response} \quad e^{-i\phi_{\text{disp}}} \]

640 pixels phase-amplitude SLM : 0.06 nm

\[ T(\omega) \]

\[ E_{\text{IN}}(\omega) \]

\[ E_{\text{OUT}}(\omega) = E_{\text{IN}}(\omega) T(\omega) \]
Compensation with a pulse shaper

Cell Only

SLM Only

SLM + Cell

\[ \tau_{FWHM} = 120 \text{ fs}, \lambda = 794,76 \text{ nm} \]

\[ Rb: 5^2S_{1/2} \rightarrow 5^2P_{1/2}, \alpha_0 L \approx 21500 \]

Efficient Compensation

Up to 85% of the incident energy recovered below the initial pulse envelop
Origin of Limitations:

1. Pixelisation: under-sampling (0.06 nm)

2. Diffraction:

\[ \phi(n) \neq \phi(n+1) \]

Finite spot size for each spectral component
When Laser spot covers 2 Pixels

\[ \phi(n+1) \neq \phi(n) : \text{Interferences} \]

\[ \Rightarrow \text{Spectral hole around } \lambda ! \]
Compensation with a pulse shaper

- Asymptotic part well reproduced
- Unable to reproduce exact behaviour near the resonance
- Spectrum intensity is affected
Flat Phase

\[ \sin(\phi) \]

\[ \nu - \nu_0 \text{(THz)} \]

Spectrum (arb. units)

\[ \lambda (\text{nm}) \]

Crosscorrelation (arb. units)

\[ \text{time (fs)} \]
3. Case of an ultrashort pulse train

- **Independent pulses**: Intensity superposition.
- **Mutually coherent pulses**: Field superposition.

\[ \phi \quad \text{(phase shift/} \omega_0 \text{)} \]

\[ \begin{array}{c}
\text{CELL} \\
\text{[Diagram showing pulse train and phase shift]} \\
\end{array} \]

\[ \sum \text{waves} = ? \]

Depends on both \( \phi \) and \( T \)
\[ \tau_0 = 75 \text{ fs}, \quad \Phi = 0, \quad T = 577 \text{ fs}, \quad \alpha_0 | = 520 \]

\[ \tau_0 = 75 \text{ fs}, \quad \Phi = \pi, \quad T = 577 \text{ fs}, \quad \alpha_0 | = 520 \]

\[ \frac{c}{\lambda^2} \alpha_0 | \Delta_d \]

\[ \frac{c}{\lambda^2} \frac{1}{T} \]

Negligible if \( \alpha_0 | \Delta_d < T^{-1} \); \( \alpha_0 | \) max \( \sim \) \( \tau_0^{-1} / \Delta_d \)
PHASE CONTROL OF DISPERSION EFFECTS

\[ \phi = \pi \Rightarrow \text{High sensitivity} \]

\[ \phi = 0 \Rightarrow \text{Robust propagation} \]
4. Propagation in an atomic system driven by a strong field

Modifications due to the Strong Field (effect of Relative Phase and Intensity)

On the energy and temporal Profile of the propagating pulse

Rb atom $4s\,S_{1/2} \rightarrow 4p\,P_{1/2}$
Coherent Control of the Gain

Relative phase $\phi=\omega_L t_A$

Propagating Pulse Intensity (mV)

- Crossed polarisation!
- Interference at $2\omega_L$ in one photon transition!!!
Interpretation

1 - "Ordinary" interference in one photon transition (Temporal Ramsey fringes)

Looking at the population the excited state:

\[ n_f = 4n \cos^2 \phi / 2 \]

Interference between two absorption paths phase-shifted by \( \phi = \omega_L t_a \)
2- Our situation

Absorption Path

Emission Path

Interference phase $2\phi$

1- Interf. between absorp. and emis. paths connecting two linear superp. of states
2- Interference phase $2\phi$, $\phi = \omega_L t_a$ the phase with respect to the strong field.
3- The two paths are « synchronous » (phase shifted but not delayed!)
Dressed state analysis

\[(|a\rangle, |b\rangle) \leftrightarrow |\pm\rangle = \frac{\mp |a\rangle + e^{-i\omega_0 t} |b\rangle}{\sqrt{2}}\]

\(|a\rangle \rightarrow |+\rangle \quad \text{Rabi frequency} \quad |b\rangle \rightarrow |\pm\rangle \]

\(|+\rangle \quad \Omega(t)\]
Action of the weak field

Two situations

$\phi = 0$

$\phi = \frac{\pi}{2}$

Transparency window
Non-resonant

Absorption/amplification
Resonant
Dependence vs Strong Field energy at Zero Delay

\[ \phi = \frac{\pi}{2} \]

\[ \phi = 0 \]

**THEORY**

\[ \frac{E_{\text{out}}}{E_{\text{in}}} \]

**EXPERIMENT**

\[ \frac{E_{\text{out}}}{E_{\text{in}}} \]
Control of the Shape

\[ \theta_F = 1.2\pi \]
Phase $\phi$

Control Field Strength $\theta_F$

Energy

Shape

Phase $\phi$
5. Towards « active » pulse shaping

Classical devices (pulse shaper)

Passive: no amplification
no creation of spectral components

Strongly driven system

Active: create new frequencies
(light-shift)
Transient Light Shift in a 3 level $\Xi$ system

$E_{out} = E_{in} + \propto e^{-i \omega_L t} + \propto e^{-\int_{-\infty}^{t} \omega - d t'}$

$\text{Fixed Laser Frequency}$ + $\text{Varying Dipole Frequency}$
\[ \Delta \phi = \int_{-\infty}^{t} (\omega_0(t') - \omega_L) dt' \]
**Varying** Dipole Frequency

+ 

**Fixed** Laser Frequency

\[ \Delta \phi = \int_{-\infty}^{t} \left( \omega_0 (t') - \omega_L \right) dt' \]

**Fixed** Dipole Frequency

+ 

**Varying** Laser Frequency

\[ \Delta \phi = \int_{-\infty}^{t} \left( \omega_0 - \omega_L (t') \right) dt' \]
Chirped pulse propagation: principle

- Self-induced heterodyne field

- Mapping of the incident field phase on the intensity profile

\[ \Delta \phi = \int_{-\infty}^{t} (\omega_0 - \omega_L(t')) dt' \]

- Direct basic temporal shaping
Chirped pulse propagation: experiment

Depth of modulation increases with the density
Conclusion

- **Atomic system at equilibrium:**
  Compensation of Dispersion for a weak pulse and a pulse train

- **Strongly driven atomic system:**
  $2\omega$ Oscillations on one photon transition
    - Coherent Control of Energy
    - Coherent Control of the pulse Shape and possibility of active pulse shaping