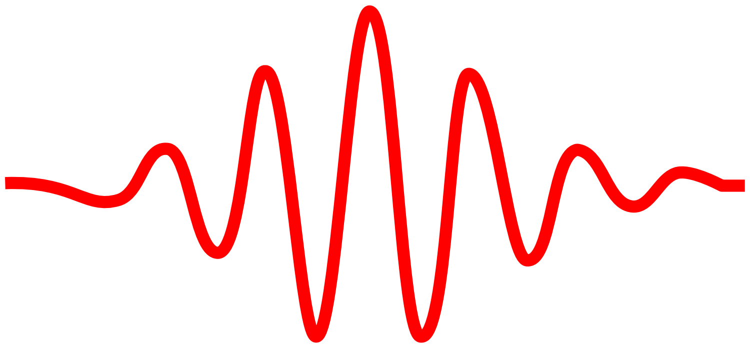
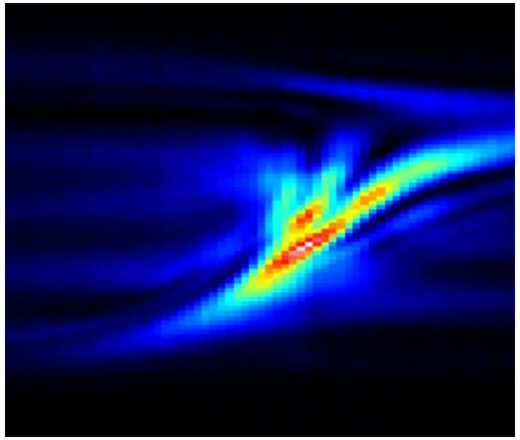
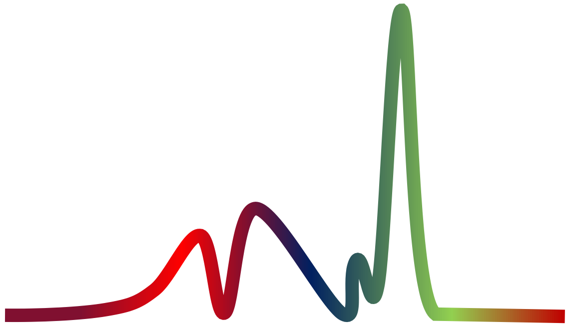
Journées thématiques : « Techniques de caractérisation des impulsions ultrabrèves »
9-10 novembre 2023

Diagnostics fondamentaux II







V. Loriot

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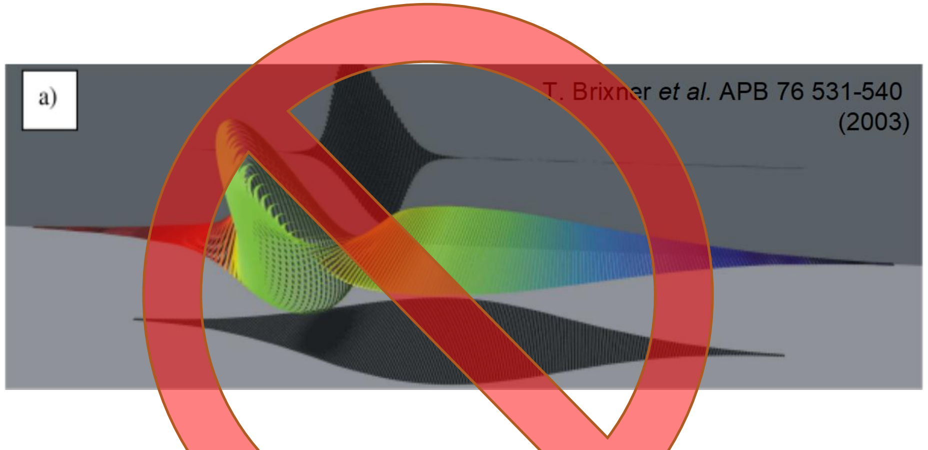
Outline

- Assumption on the pulse to characterize
- Non-linearity of the 2^{nd} and 3^{rd} order (SHG-THG)
- Time scanning techniques:
  - From autocorrelation to FROG
  - Tip-Toe
  - FROST
- Chirp-Scans - *d*-scans
- SSIR (Wizzler)

Assumption on the pulse to characterize (in this lecture)

Need to be linearly polarized

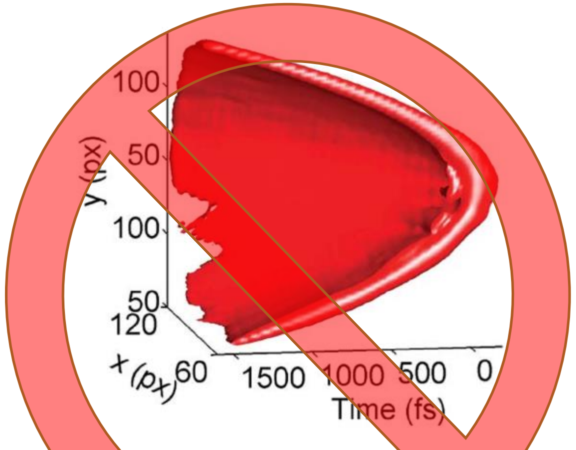
$$\vec{E}(x,y,t) = E(x,y,t)\,\vec{e}_x$$



The two polarization can have a different temporal profile
Application: temporal gating for HHG

Need to be spatially homogeneous

$$\vec{E}(x,y,t) = E(x,y) \times E(t)\,\vec{e}_x$$



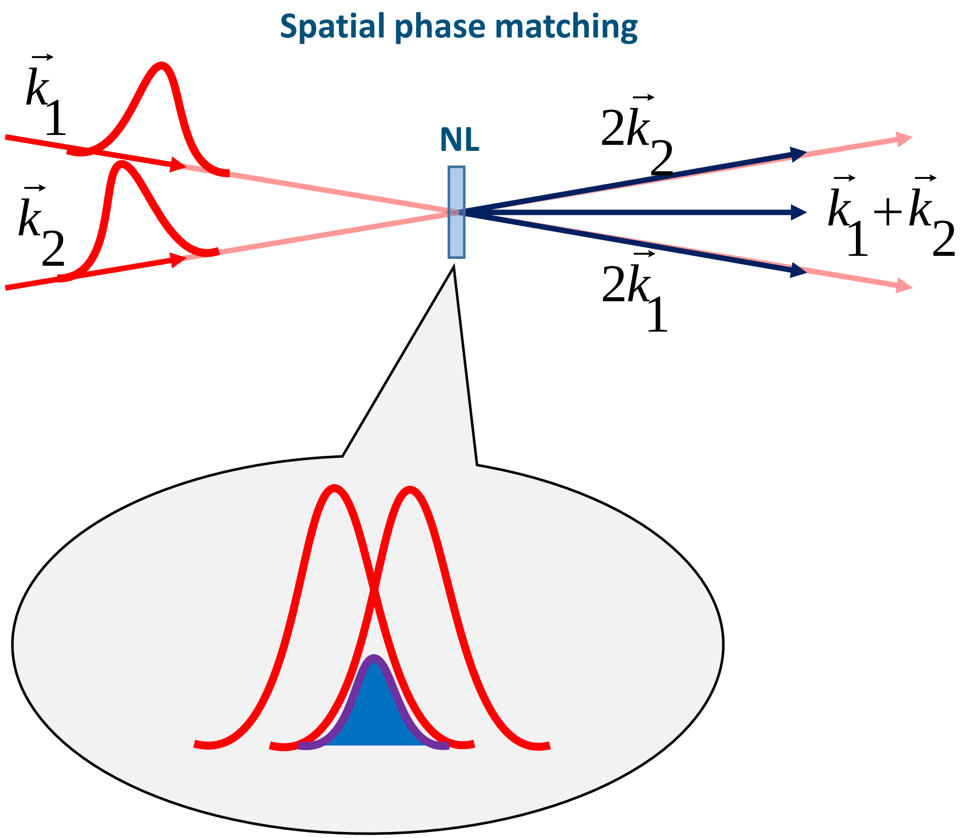
The pulse duration is 100 fs everywhere
but the temporal profile is centered along a spatial parabola

For such pulses see the spatio-temporal characterization tomorrow afternoon

V. Loriot *et al.* Photon. Tech. Letts. **24** 273-275 (2012)

Non-linearity of the 2nd and 3rd order (SHG-THG)

Non-collinear SHG in the temporal/spatial domain



$$I_{2\omega}^{(2k_2)}(t) = I_{\omega}^{(k_2)}(t) \times I_{\omega}^{(k_2)}(t) = \left|I_{\omega}^{(k_2)}(t)\right|^2$$

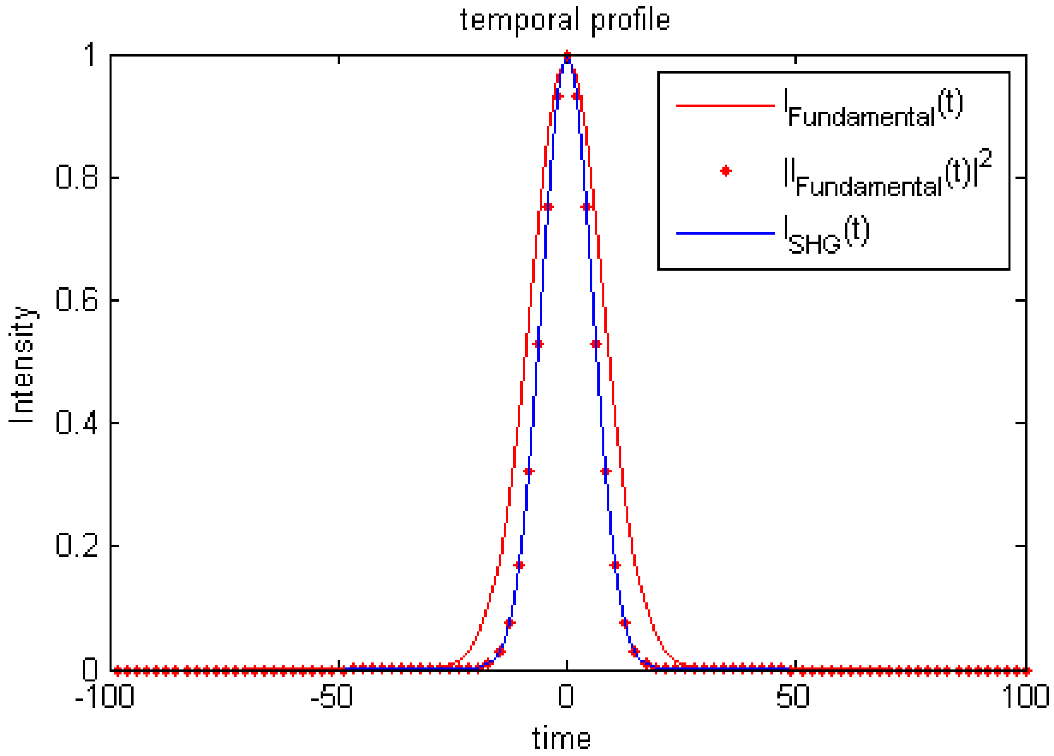
$$I_{2\omega}^{(k_1+k_2)}(t) = I_{\omega}^{(k_1)}(t) \times I_{\omega}^{(k_2)}(t)$$

$$I_{2\omega}^{(2k_1)}(t) = I_{\omega}^{(k_1)}(t) \times I_{\omega}^{(k_1)}(t) = \left|I_{\omega}^{(k_1)}(t)\right|^2$$

A standard detector would measure the power of the doubled frequency

$$S(t) = R(t) \times \int_{-\infty}^{\infty} I(t)dt$$

SHG and pulse duration with perfect phase matching



$$I_\omega(t) \propto e^{\left(-4\ln(2)\frac{t^2}{\Delta t_\omega^2}\right)}$$

$$I_{2\omega}(t) \propto |I_\omega(t)|^2$$

$$I_{2\omega}(t) \propto e^{2\left(-4\ln(2)\frac{t^2}{\Delta t_\omega^2}\right)}$$

$$I_{2\omega}(t) \propto e^{\left(-4\ln(2)\frac{t^2}{\left(\Delta t_\omega/\sqrt{2}\right)^2}\right)}$$

$$\boxed{\Delta t_{2\omega} = \Delta t_\omega/\sqrt{2}}$$

SHG shorten the pulse duration

This is the case for ideal SHG crystal:

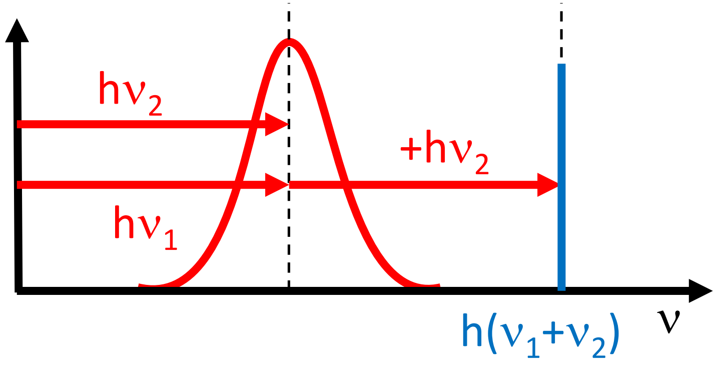
- Perfect angular phase matching
- The frequency can be doubled with the same efficiency on the whole spectrum

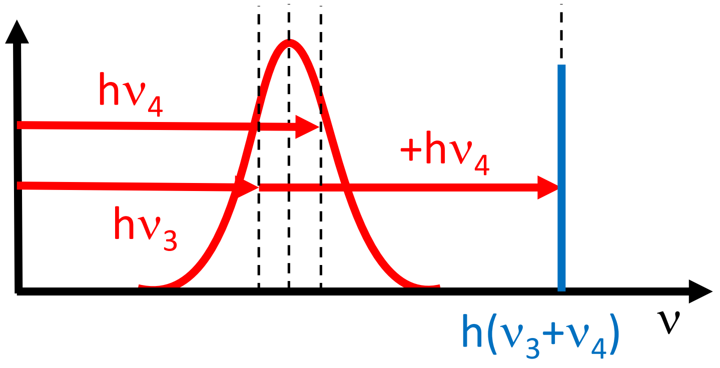
Works for infinitely thin SHG crystal

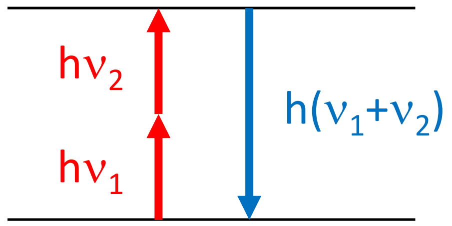
SHG in the spectral domain

Non-lineal crystal

- When the intensity is high enough (~GW.cm^{-2})
- Non-centrosymmetric crystal
- Non linear crystal sum the frequencies (energy E=hν)







Pathways interferences

- It exists many pathways to generate a given 2nd harmonic frequency
- Each pathway creates a wave with a given amplitude and phase
- All the pathways interfere together
- The electric field at 2ω depends on the amplitude and phase of all possible pathways

$$\tilde{E}(2\omega_0) = \int_{-\infty}^{\infty} \tilde{E}\left(\omega_0 - \frac{\delta}{2}\right) \times \tilde{E}\left(\omega_0 + \frac{\delta}{2}\right) d\delta$$

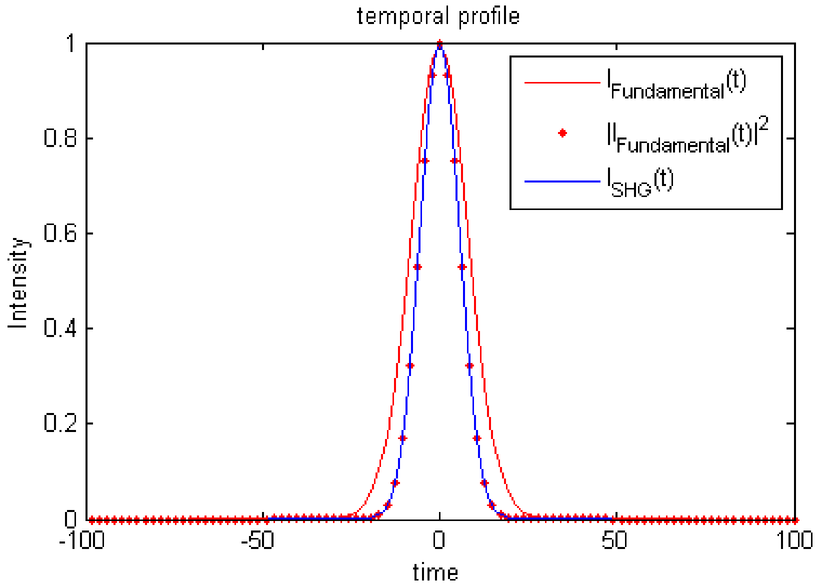
$$\tilde{E}_{2\omega}(\omega) = \tilde{E}_{\omega}(\omega) \otimes \tilde{E}_{\omega}(\omega)$$

Spectral Auto-convolution

SHG spectral width in LTF with perfect phase matching

<u>Gaussian width spectral-temporal relation (FWHM)</u>

$$\Delta t \Delta \omega = 4 \ln 2$$
$$\omega = 2\pi\nu$$
$$\lambda\nu = c$$
$$\omega = \frac{2\pi c}{\lambda}$$
$$\Delta\omega = \frac{2\pi c \Delta\lambda}{\lambda_0^2}$$
$$\Delta t \Delta\lambda = 4 \ln 2 \frac{\lambda_0^2}{2\pi c}$$
$$\Delta t = 4 \ln 2 \frac{\lambda_0^2}{2\pi c \Delta\lambda} = \frac{2 \ln(2) \lambda_0^2}{\pi \; c \; \Delta\lambda}$$



@800 nm 20 fs -> $\Delta\lambda$ =47 nm - $\Delta\nu$ = 22 THz

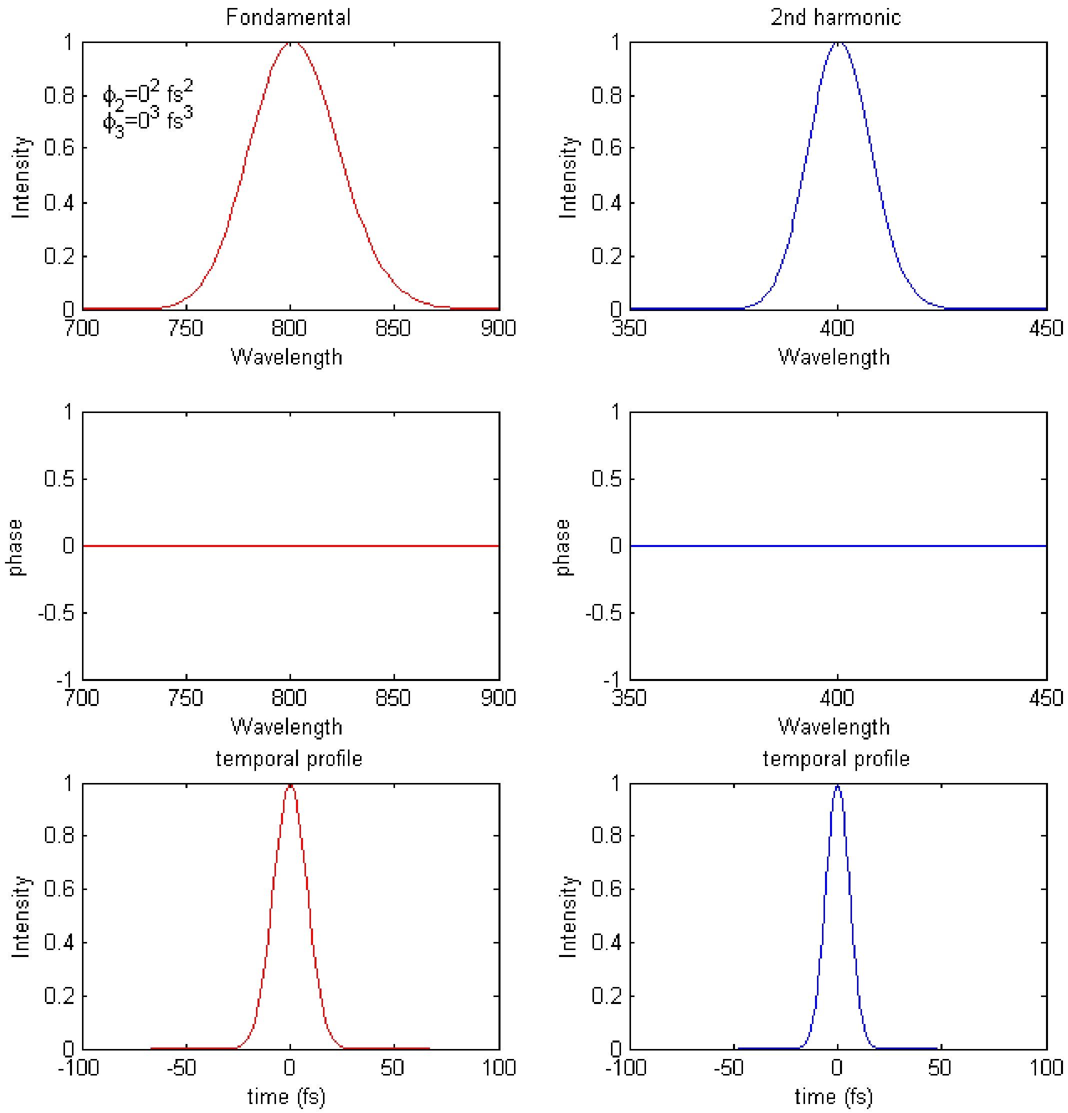
@400nm 20 fs/$\sqrt{2}$=14.14 fs

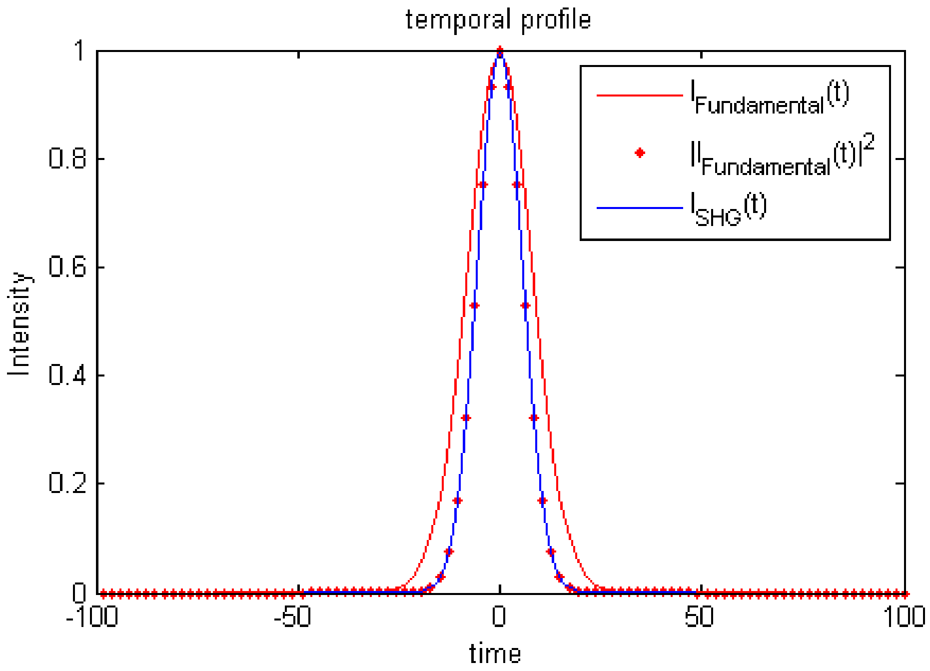
@400 nm 14 fs -> $\Delta\lambda$ =47/2$\sqrt{2}$= 16 nm

@400nm $\Delta\nu$=22$\sqrt{2}$=30 THz

<u>SHG broaden **$\Delta\nu$** and shorten **$\Delta\lambda$**</u>

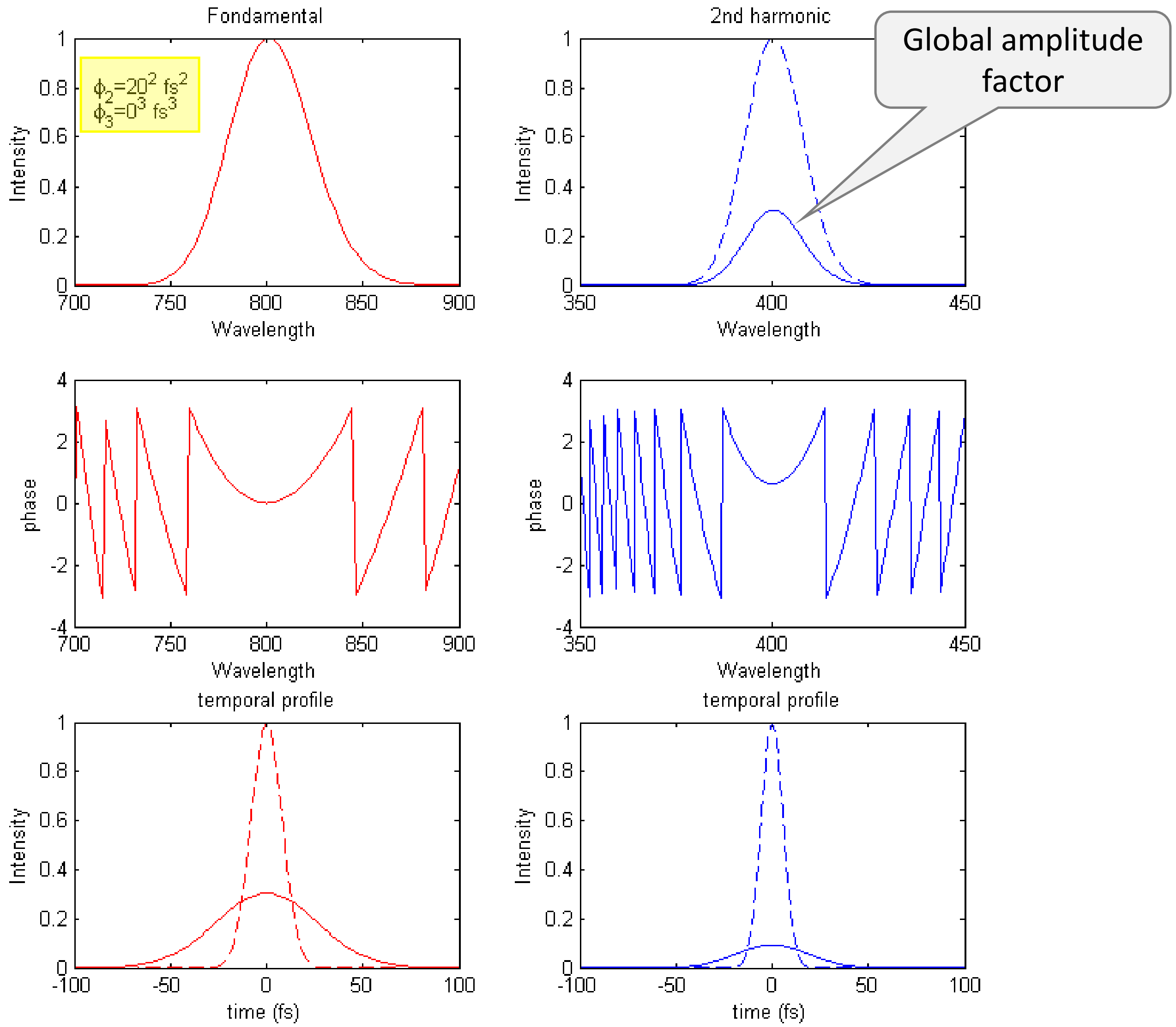
SHG in the temporal v.s. spectral domain

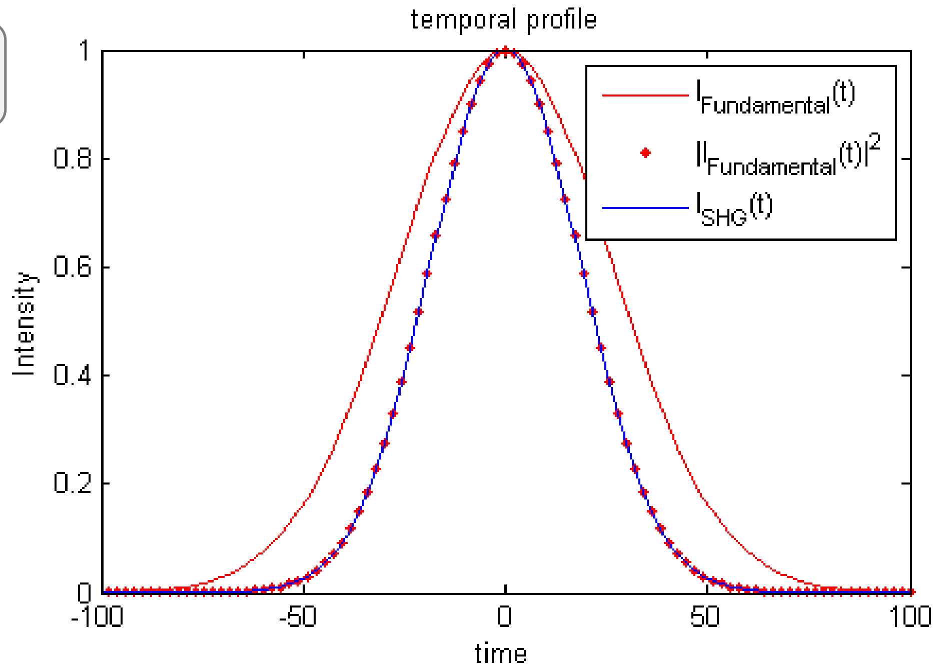




LTF

SHG in the temporal v.s. spectral domain



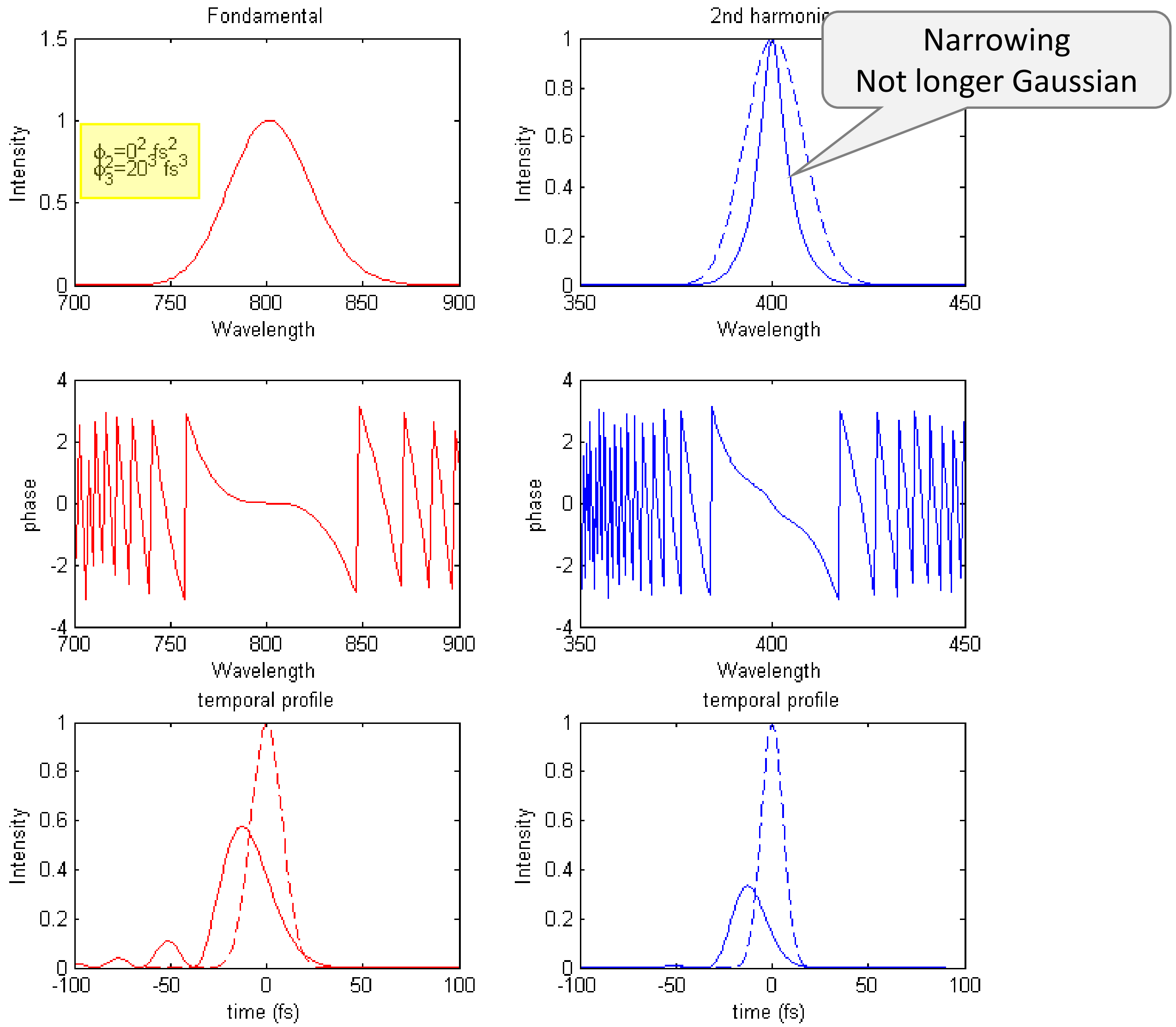


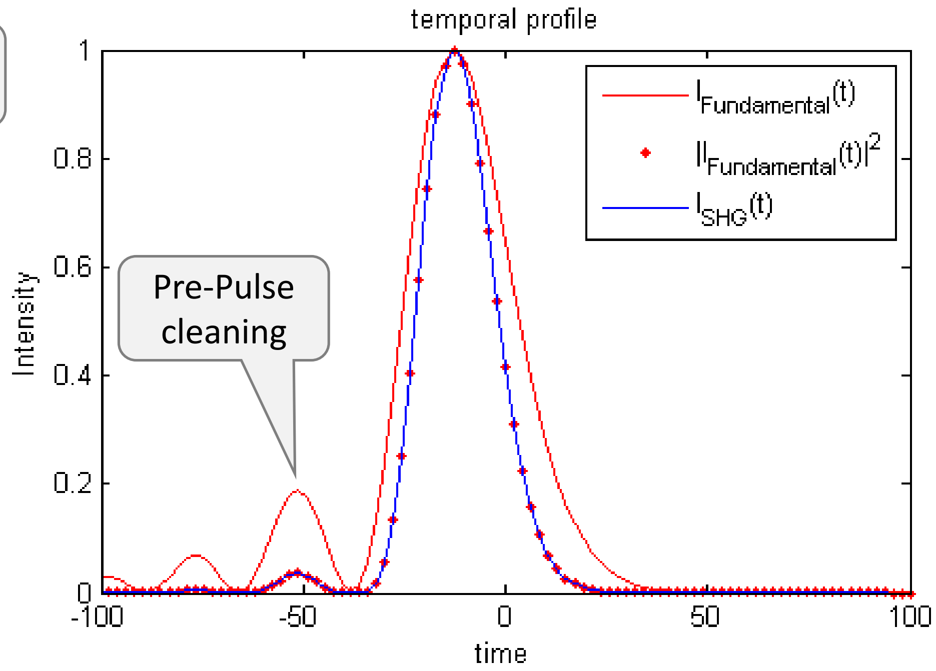
<u>With a chirp</u>

(20 fs)² = 400 fs²

Affects significantly a 20 fs pulse

SHG in the temporal v.s. spectral domain



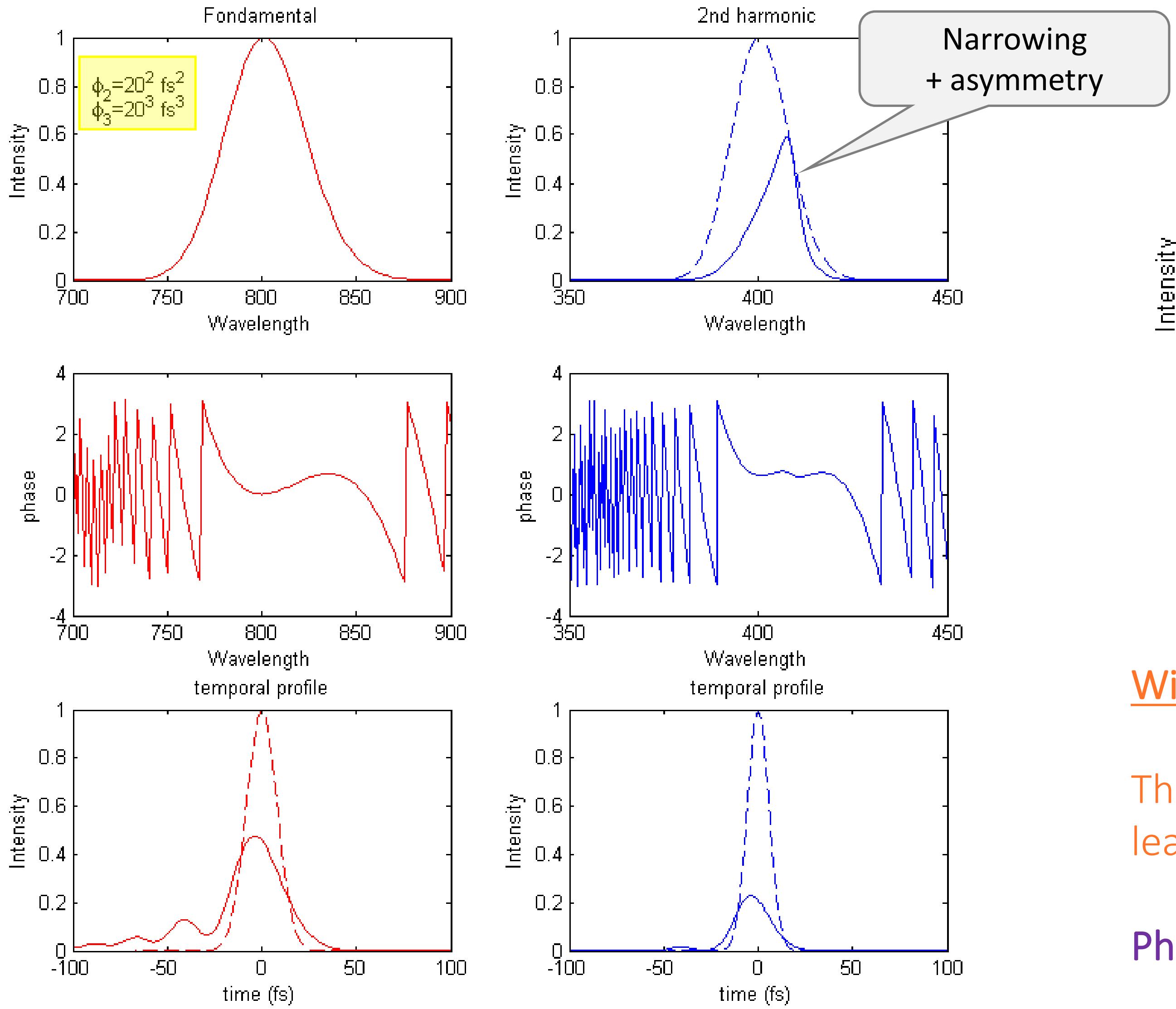


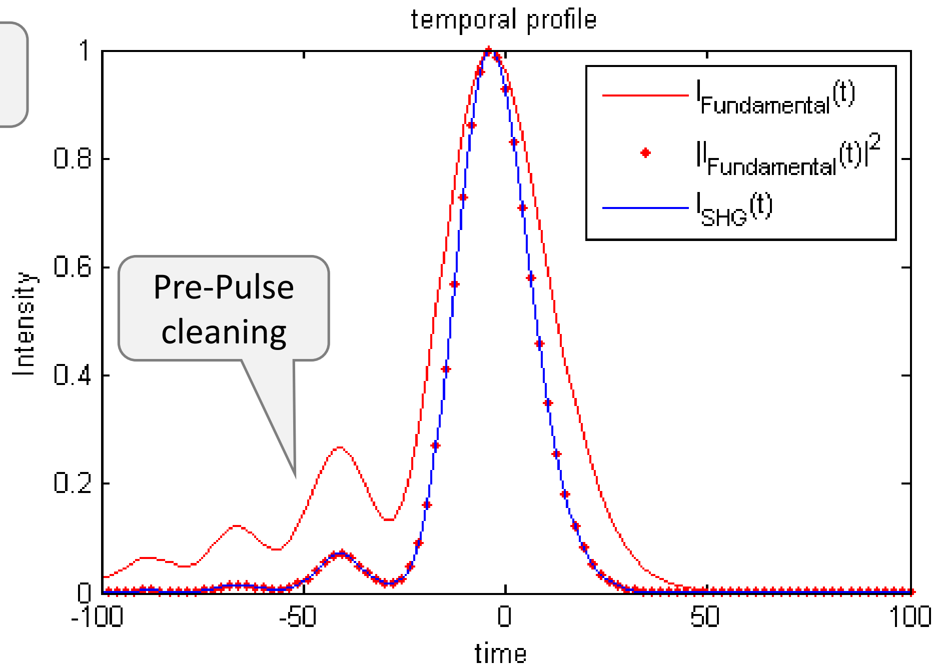
With a TOD (third order dispersion)

$(20\ fs)^3 = 8\ 000\ fs^3$

Affects significantly a 20 fs pulse

SHG in the temporal v.s. spectral domain



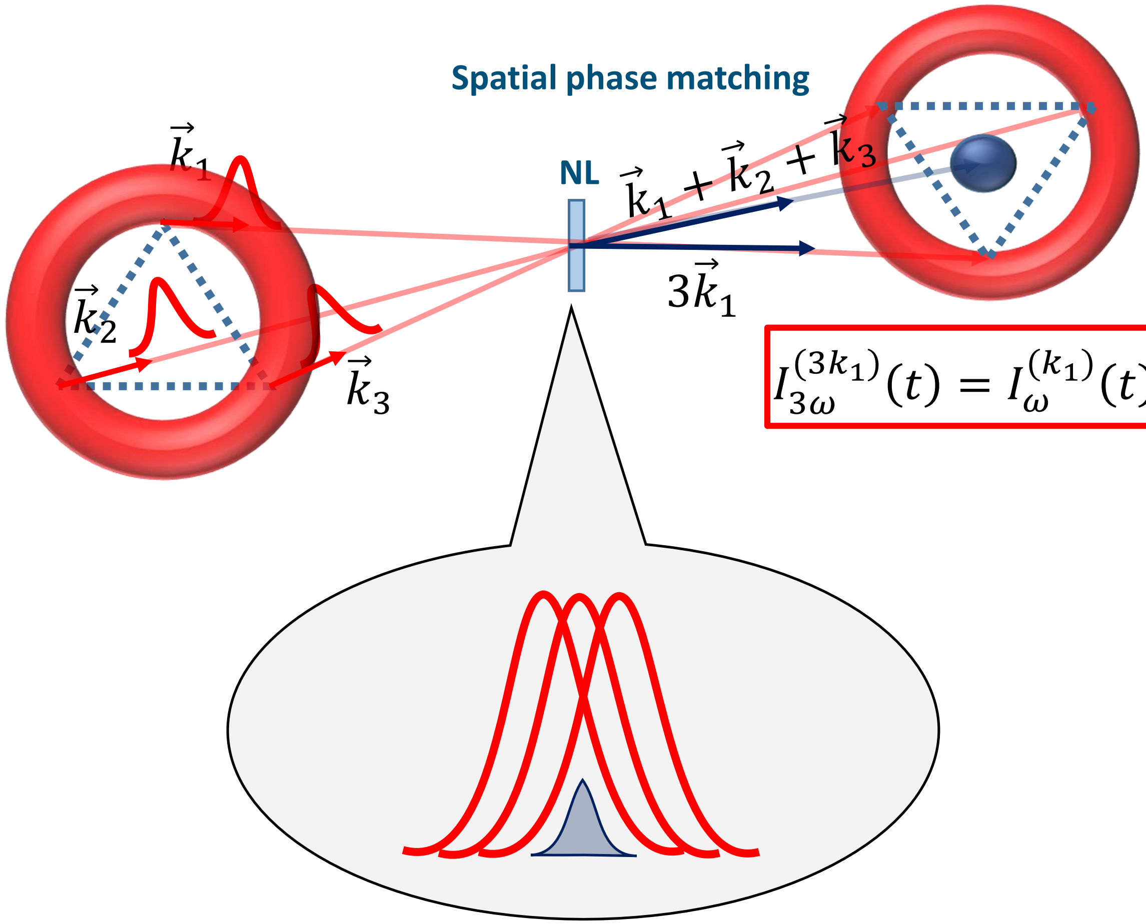


With chirp and TOD

The SHG of a Gaussian spectrum does not necessary lead to a spectrum with a Gaussian shape

Phase – Amplitude coupling

Direct non-collinear THG in the temporal/spatial domain



$$I_{3\omega}^{(3k_1)}(t) = I_{\omega}^{(k_1)}(t) \times I_{\omega}^{(k_1)}(t) \times I_{\omega}^{(k_1)}(t) = \left| I_{\omega}^{(k_1)}(t) \right|^3$$

$$I_{3\omega}^{(k_1+k_2+k_3)}(t) = I_{\omega}^{(k_1)}(t) \times I_{\omega}^{(k_1)}(t) \times I_{\omega}^{(k_1)}(t)$$

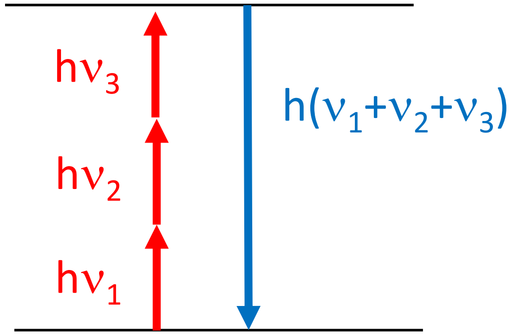
A standard detector would measure the power of the doubled frequency

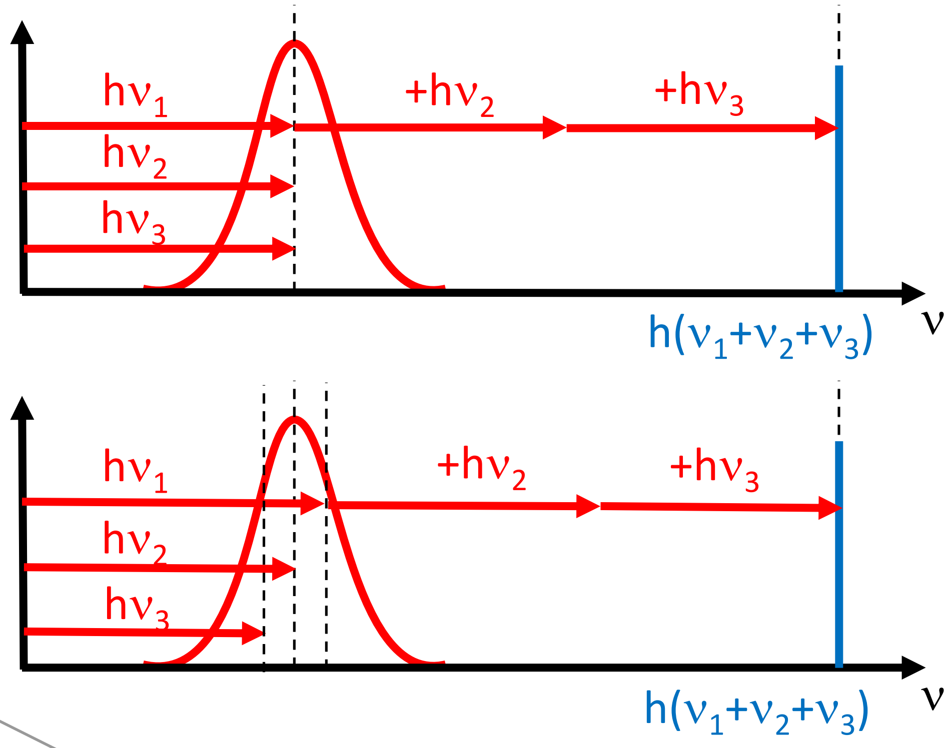
$$S(t) = R(t) \times \int_{-\infty}^{\infty} I(t)dt$$

Direct THG in the spectral domain

Non-lineal crystal

- When the intensity is high enough (~GW.cm^{-2})
- All materials allowed
- Non linear crystal sum the frequencies (energy E=hν)





Pathways interferences

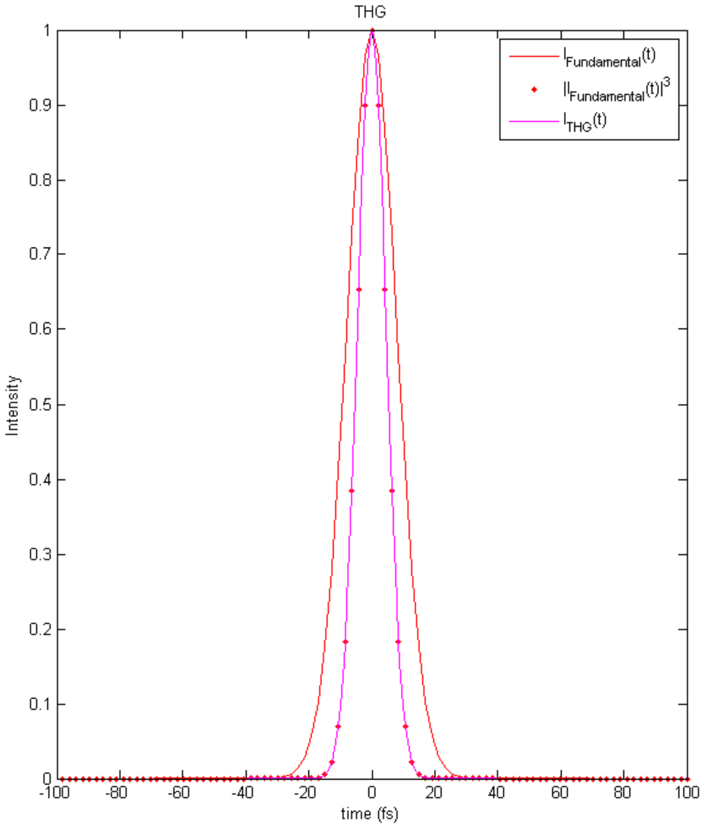
- It exists many pathways to generate a given 3rd harmonic frequency
- Each pathway creates a wave with a given amplitude and phase
- All the pathways interfere together
- The electric field at 3ω depends on the amplitude an phase of all possible pathways

$$\tilde{E}_{3\omega}(\omega) = \tilde{E}_{\omega}(\omega) \otimes \tilde{E}_{\omega}(\omega) \otimes \tilde{E}_{\omega}(\omega)$$

$$\tilde{E}_{3\omega}(\omega) = \tilde{E}_{2\omega}(\omega) \otimes \tilde{E}_{\omega}(\omega)$$

Double Spectral Auto-convolution

THG Temporal & spectral properties in LTF and perfect phase matching



@800 nm 20 fs -> $\Delta\lambda$ =47 nm - $\Delta\nu$ = 22 THz

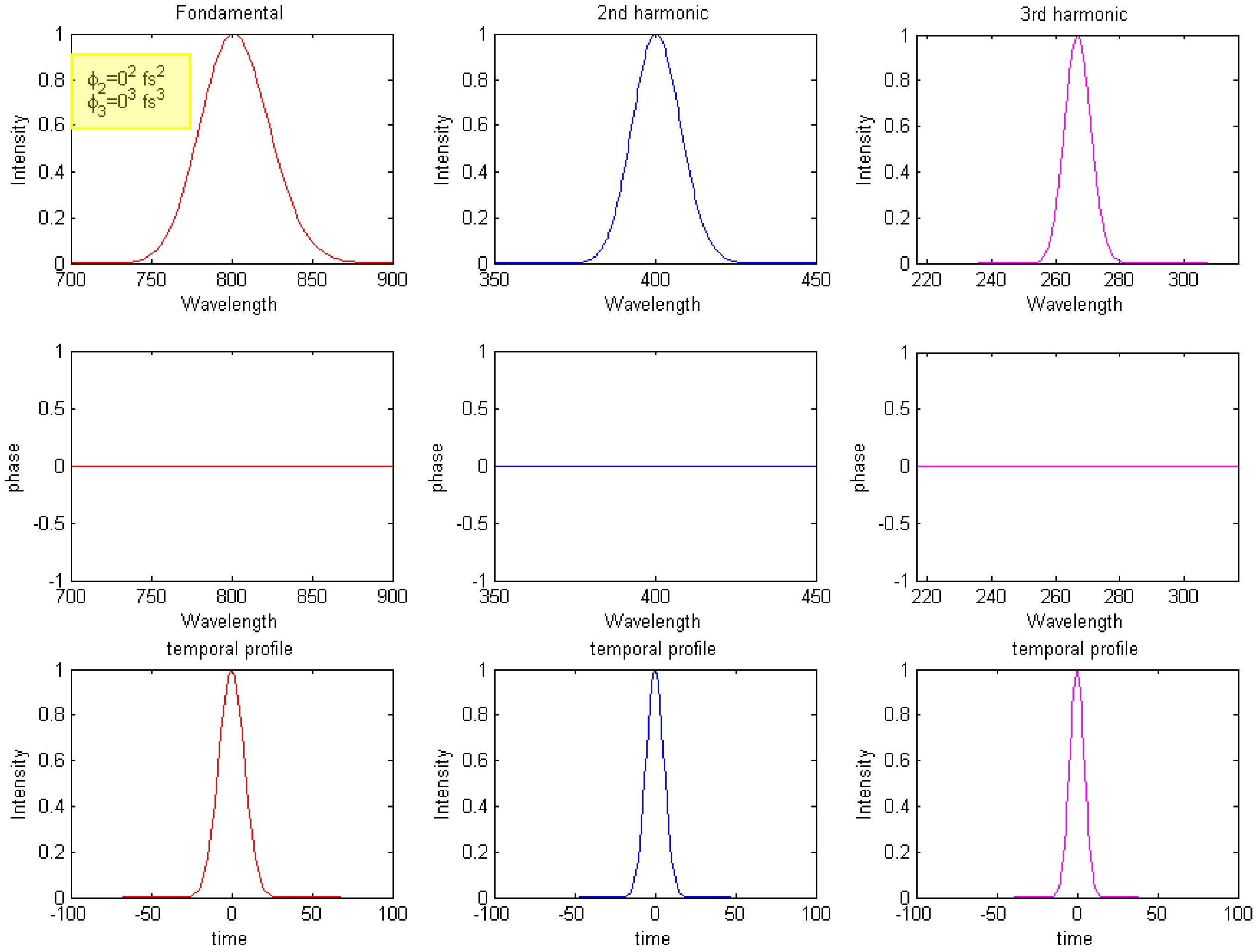
$$I_{3\omega}(t) = |I_{\omega}(t)|^3$$

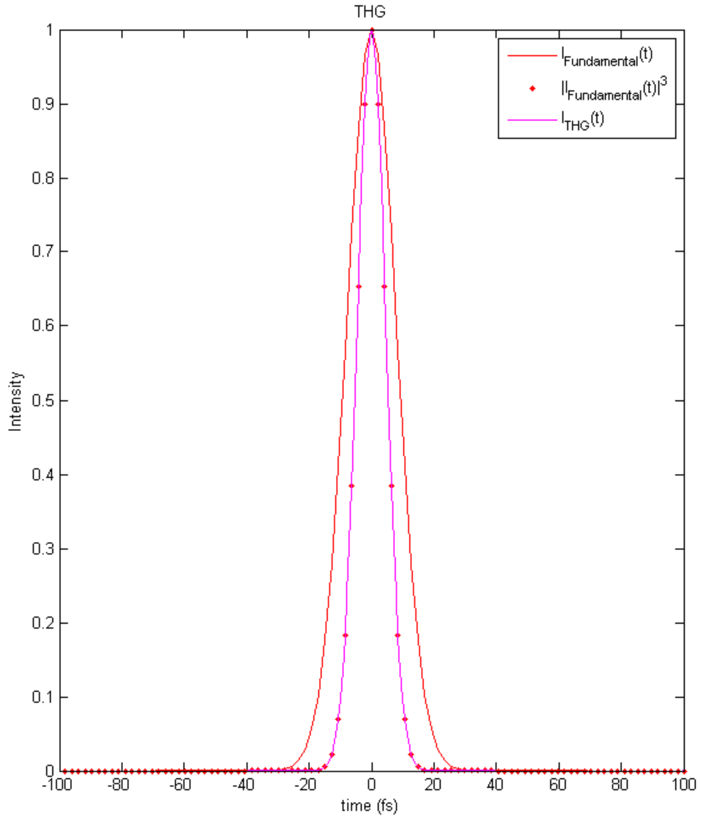
@266nm 20 fs/$\sqrt{3}$=11.5 fs

@266 nm 11.5 fs -> $\Delta\lambda = 47/(3\sqrt{3})$= 9 nm

@266 nm $\Delta\nu$=22x$\sqrt{3}$ = 38 THz

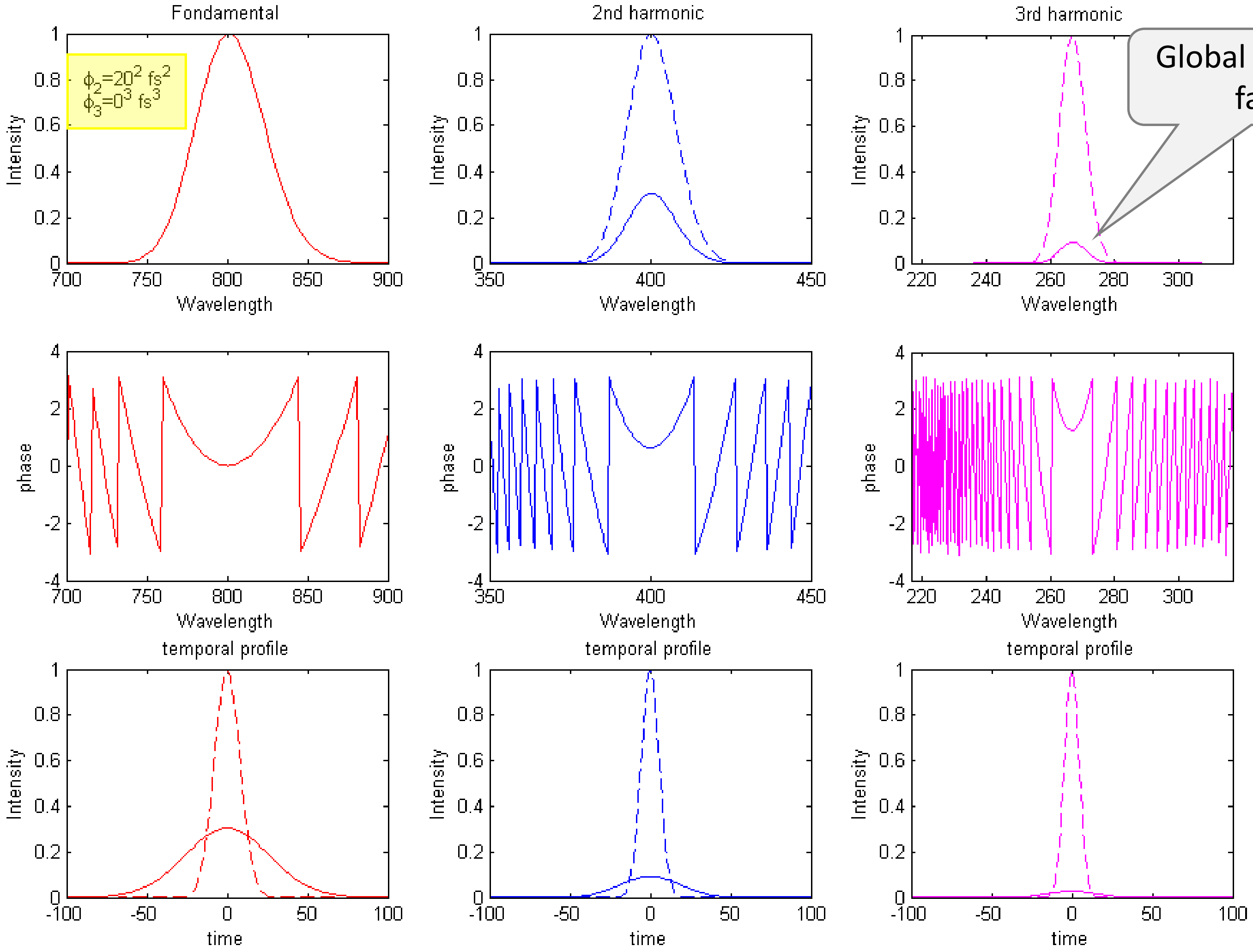
SHG-THG in the temporal v.s. spectral domain



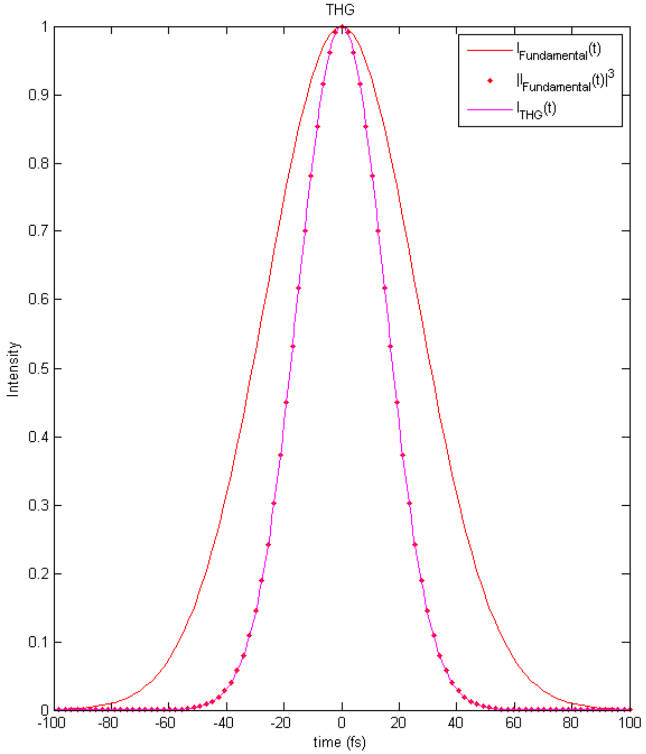


LTF

SHG-THG in the temporal v.s. spectral domain

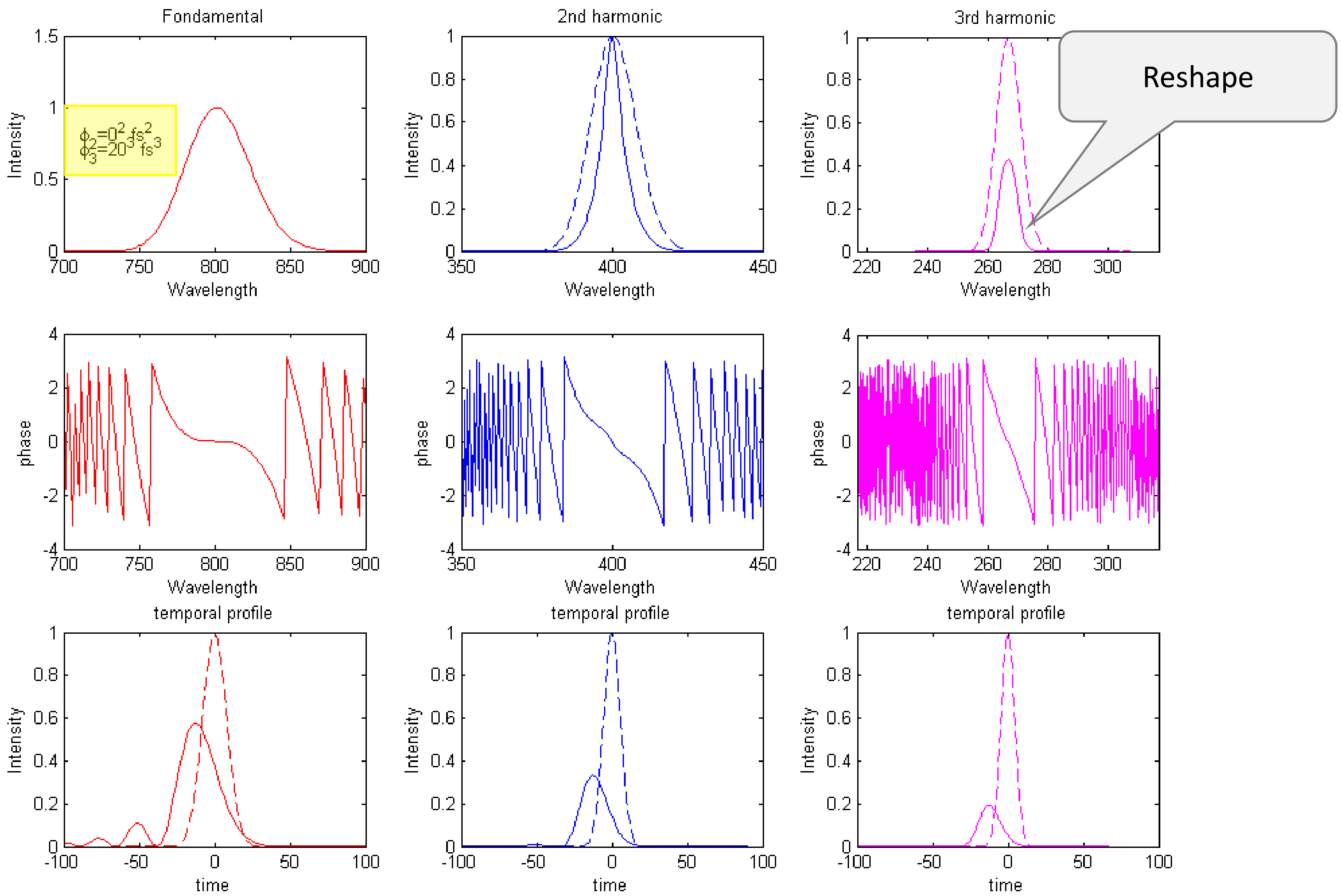


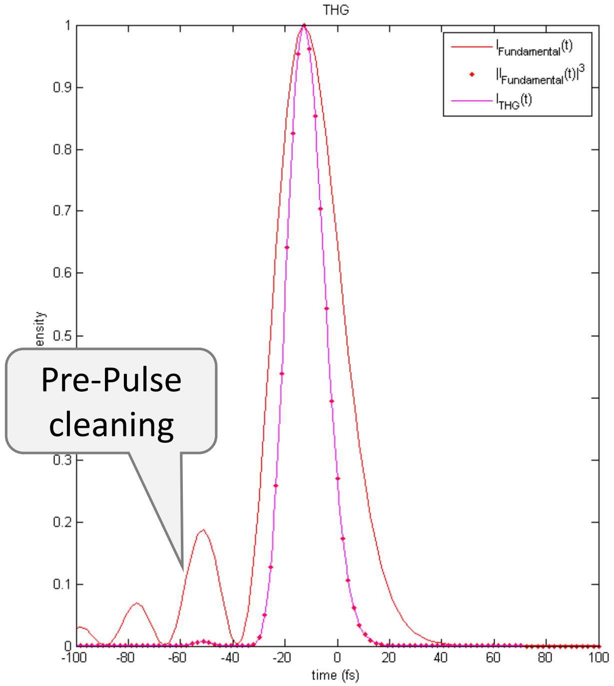
Global amplitude factor



Chirp

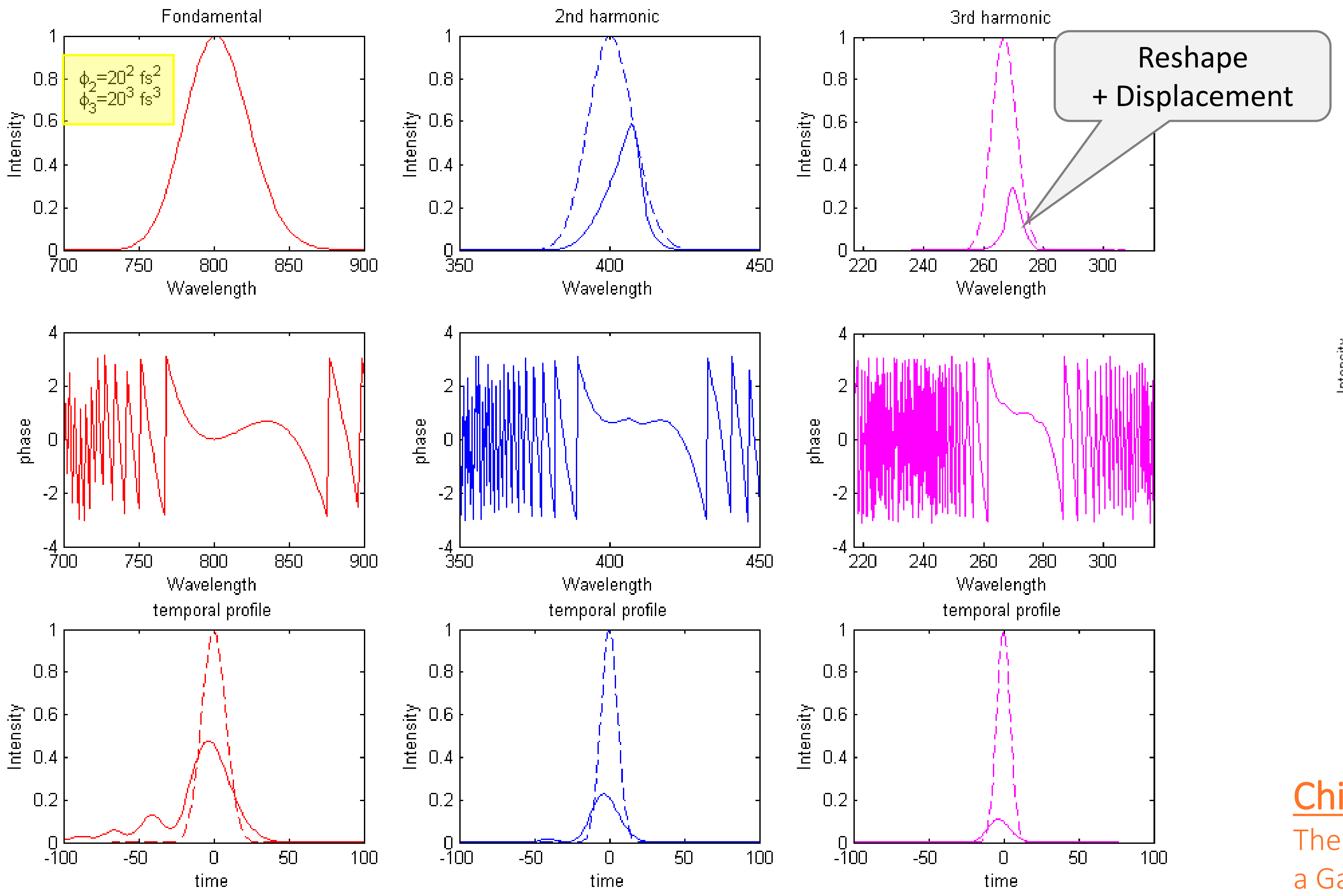
SHG-THG in the temporal v.s. spectral domain

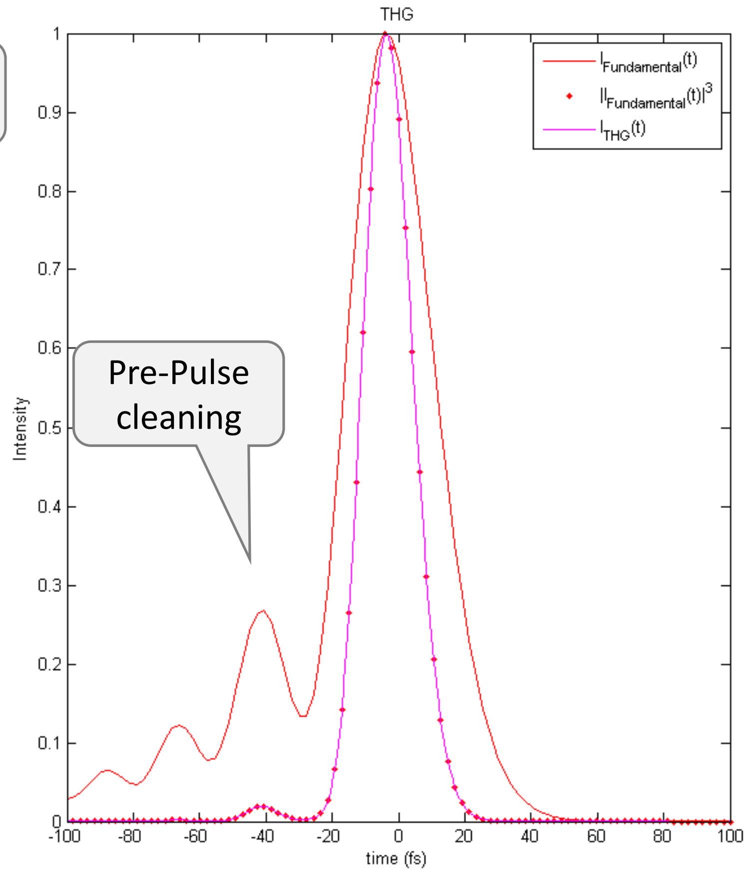




TOD

SHG-THG in the temporal v.s. spectral domain





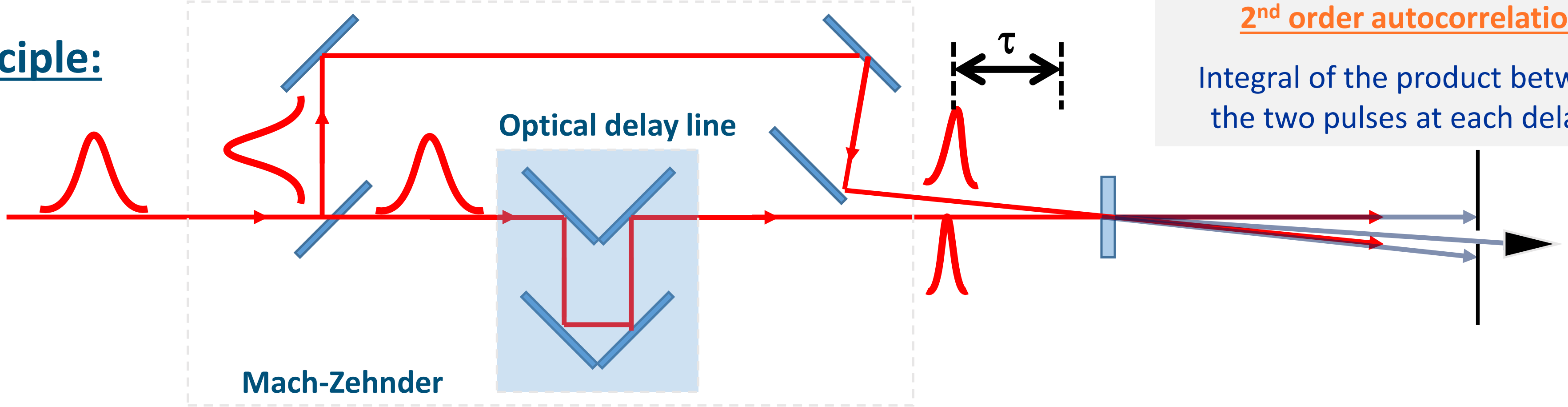
Chirp & TOD

The THG of a Gaussian spectrum is not necessary a Gaussian spectrum

Time scanning techniques:
From autocorrelation to FROG & Grenouille

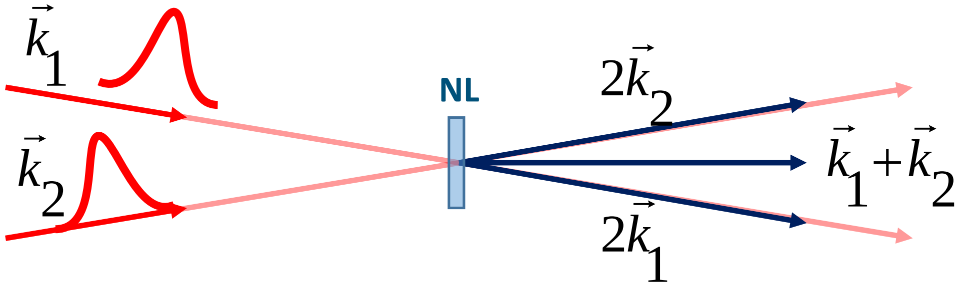
Pulse measurement : 2nd order Autocorrelation in intensity

Principle:



2nd order autocorrelation

Integral of the product between the two pulses at each delay τ



$$I_{2\omega}(t) = I_{\omega}^{k_1}(t) \times I_{\omega}^{k_2}(t-\tau)$$

$$S(\tau) = \int I_{2\omega}(t)dt = \int I_{\omega}^{k_1}(t) \times I_{\omega}^{k_2}(t-\tau)dt$$

$$S(\tau) = I_{\omega}^{k_1}(t) \star I_{\omega}^{k_2}(t)$$

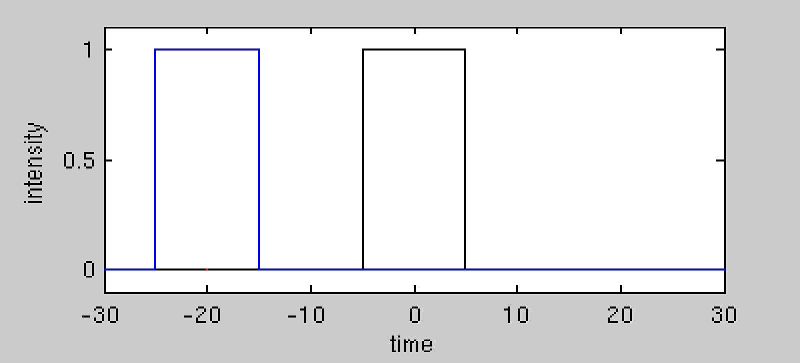
Correlation product

Using a small angle

- No optical cycle interferences
- Separate spatially the contributions

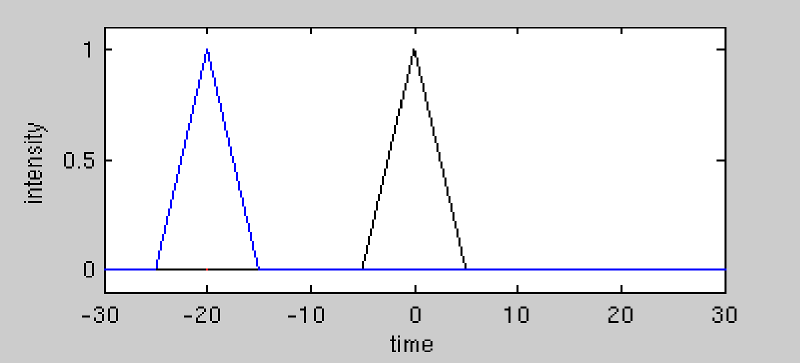
Pulse shape and 2nd order Autocorrelation

Square shape:



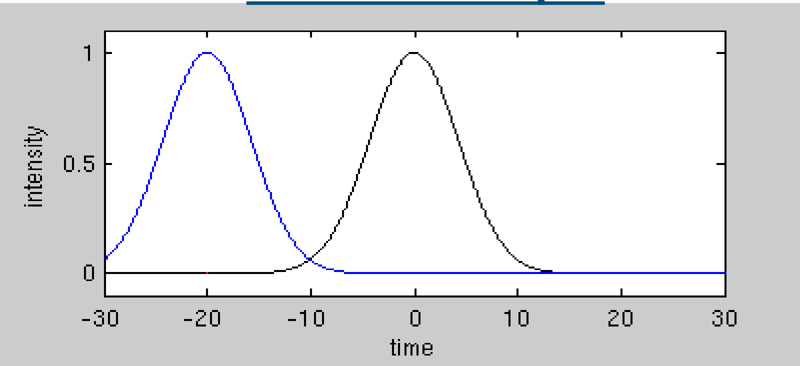
A square shape pulse make a triangular autocorrelation

Triangular shape:



A triangular shape makes a smooth curve

Gaussian shape:

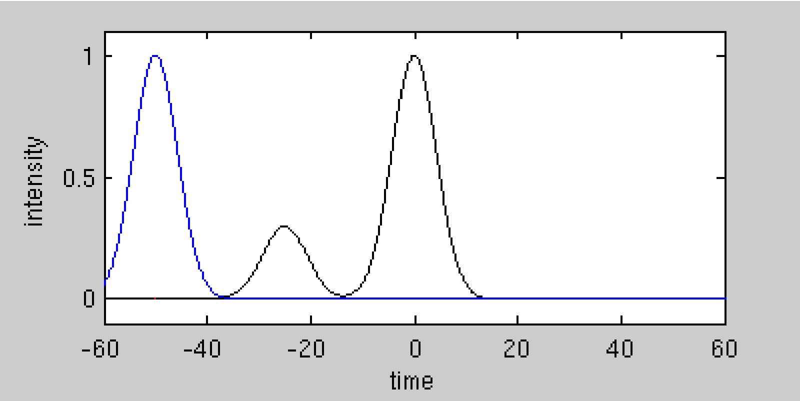


Shape information

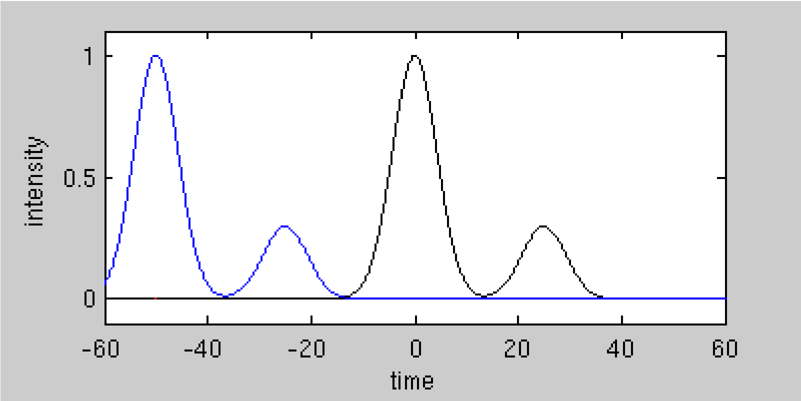
Not easy to distinguish between a Gaussian and a triangular shape with 2nd order autocorrelation

Temporal indetermination

With a pre-pulse



With a post -pulse



- Unable to distinguish between a pre-pulse and a post pulse
- 2 pulses generate 3 peaks
- Peaks broader ($\sqrt{2}$)
- Maintain position of the pulses
- Direct idea of the relative amplitudes

Limitation on the pulse measurement

The measurement *is/need to be* always symmetric

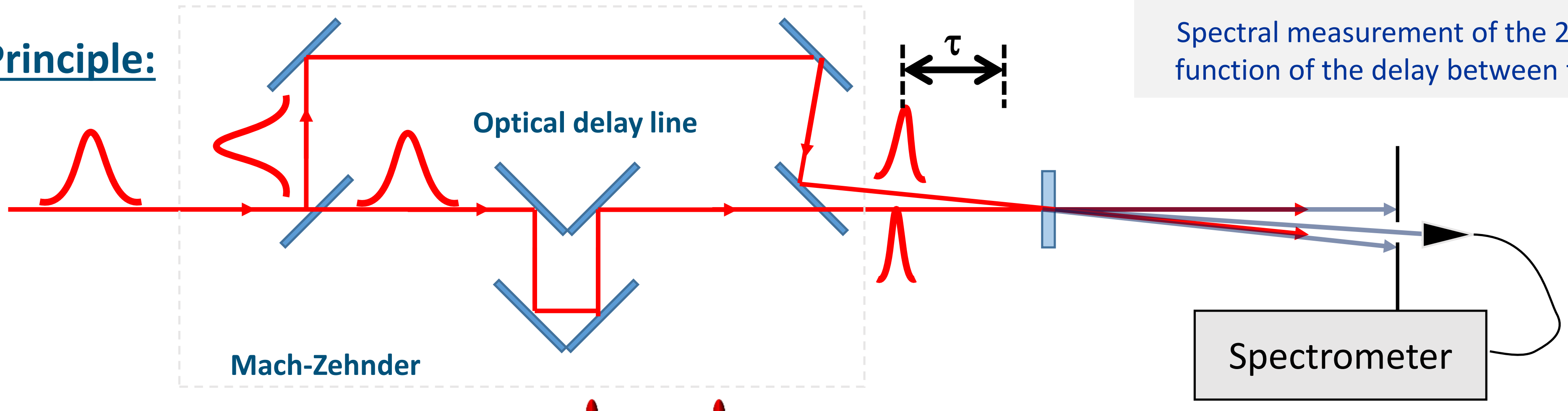
Need to suppose a pulse shape

Need to suppose the sign of the pre/post-pulses

Give an idea of the pulse duration and shape

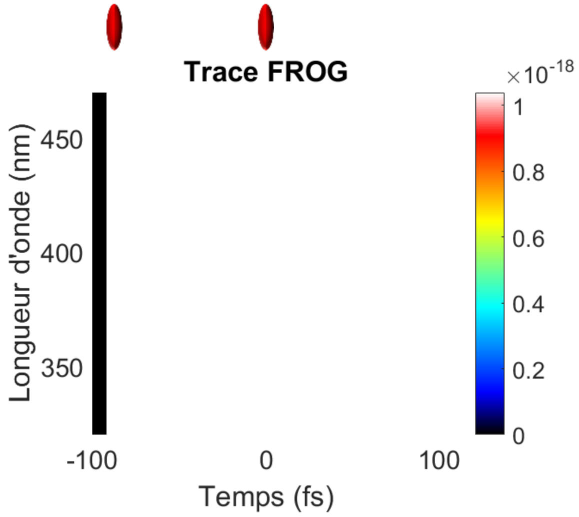
Frequency Resolved Optical Gating - FROG

Principle:



FROG

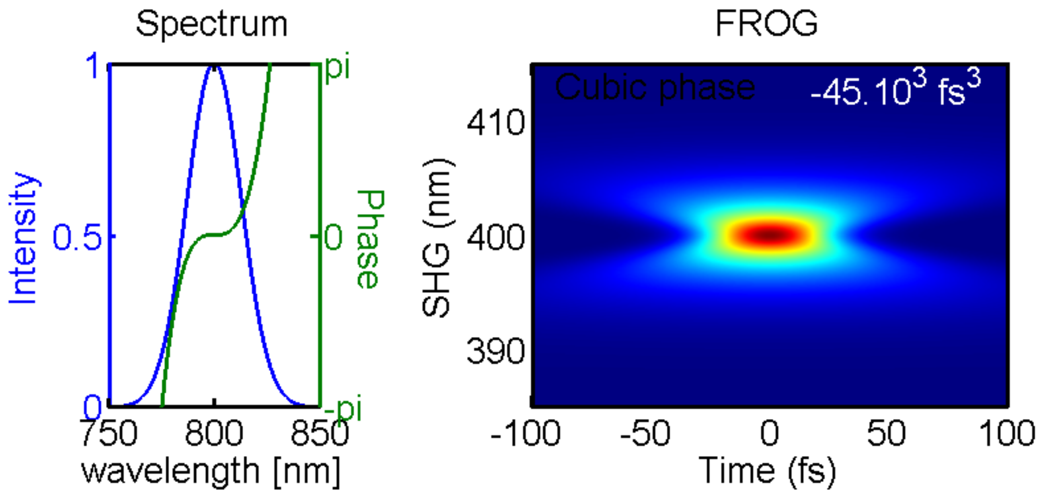
Spectral measurement of the 2nd harmonic in function of the delay between the two pulses



Rick Trebino "Frequency Resolved Optical Gating: The measurement of ultrafast laser pulses", Norwell, MA, Kluwer Academic Publishers, 2000

R. Trebino *et al.* Rev. Sci. Instrum. **69** 3277 (1997) http://frog.gatech.edu

FROG for different pulses



Movie

$$S(\tau,\omega) \propto \left| \int E(t)E(t-\tau)\,e^{i\omega t}\,dt \right|^2$$

Dimensional analysis

• We want to retrieve 2 dimensions (Amplitude & Phase)

+ We have a 2D signal (Delay & Frequency)

+ Fourier Transform is bijective

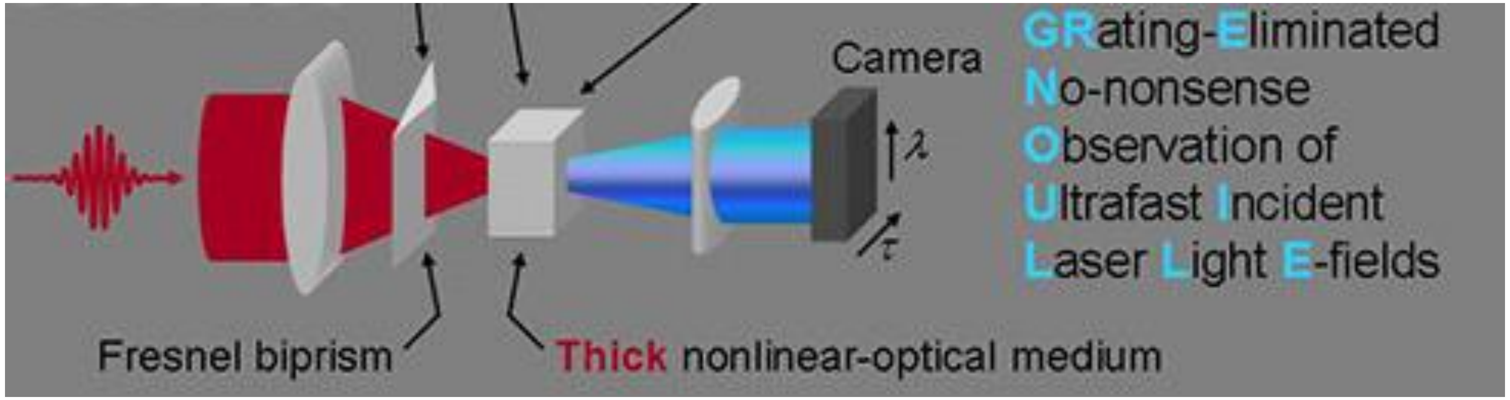
All the information is *in principle* contained into the map

!!! One dimension of FROG is symmetrized !!!

• The sign of the temporal axis is not defined

• Hence it is not possible to reconstruct the sign of the spectral phase

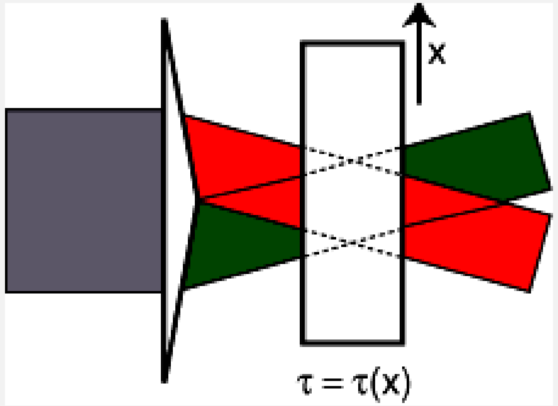
Single shot FROG : Grenouille

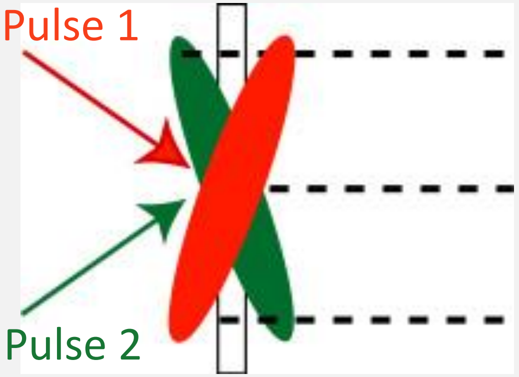


Grenouille

- Fresnel biprism introduces a delay between the right-left components.
  - The delay can be read horizontally
- The tick SHG crystal select the wavelength that can be doubled (phase matching)
  - The SHG wavelength can be read vertically

Fresnel Biprism





pulse 1 earlier than pulse 2

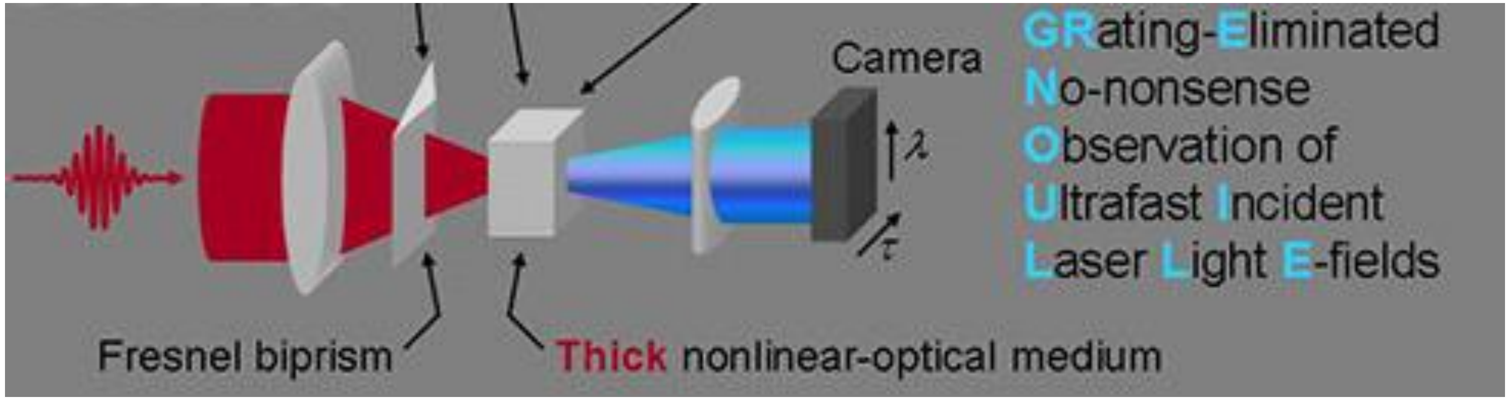
pulse 1 and pulse 2 at the same time

pulse 1 later than pulse 2

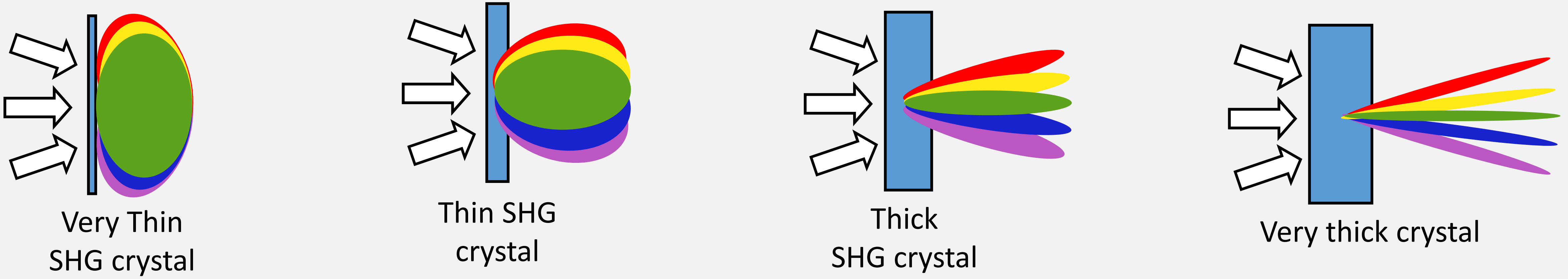
Fresnel biprism :
scan of the delay between the pulses

https://frog.gatech.edu/tutorial.html

Single shot FROG : Grenouille



Angular acceptance of tick NL crystal

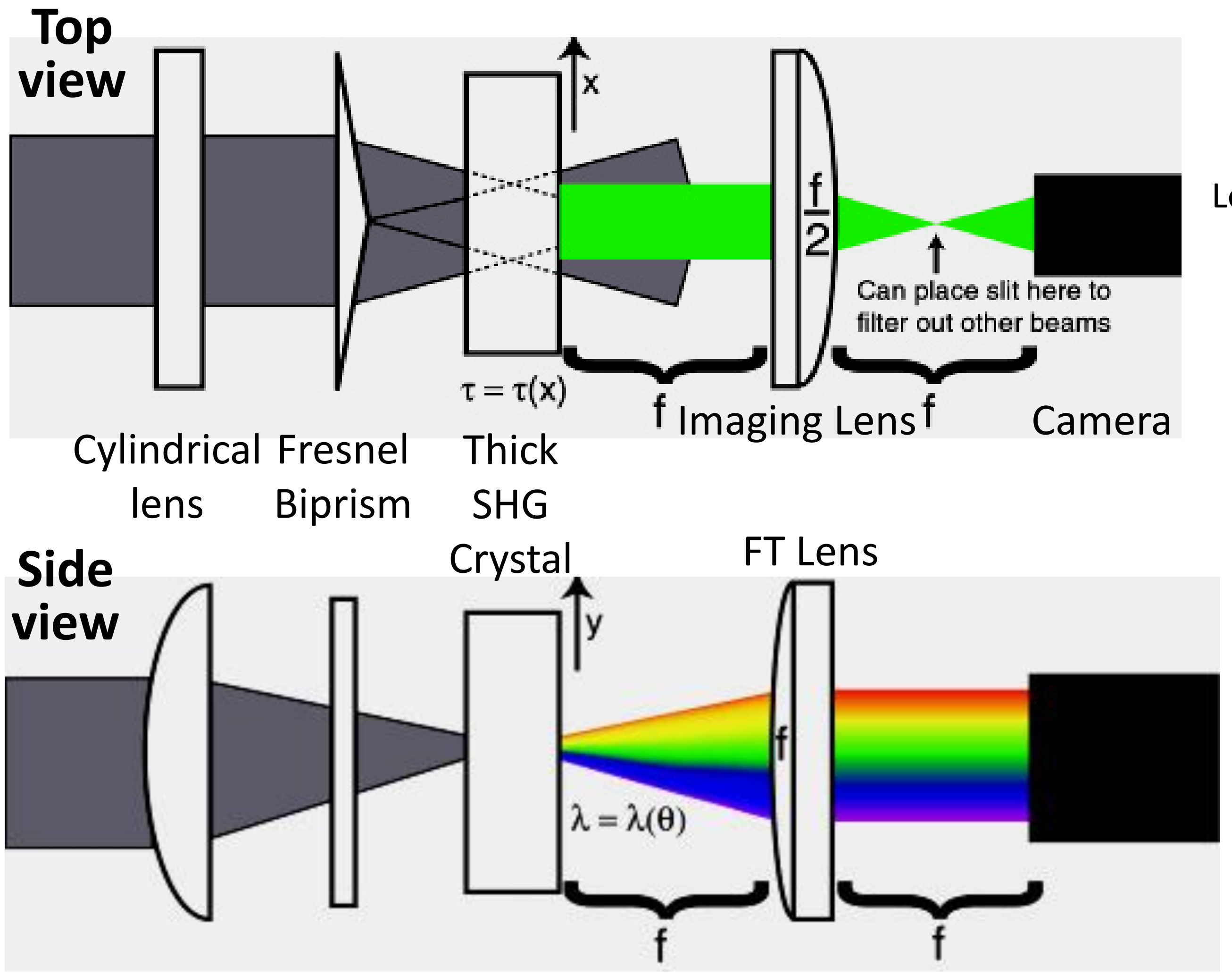


Very thin crystal can create broad SH spectrum in all directions.
Standard autocorrelators and FROGs use such crystals.

A very thick crystal acts like an angular spectrometer!
The use of a tick crystal naturally select the wavelength to be doubled

https://frog.gatech.edu/tutorial.html

Grenouille geometry



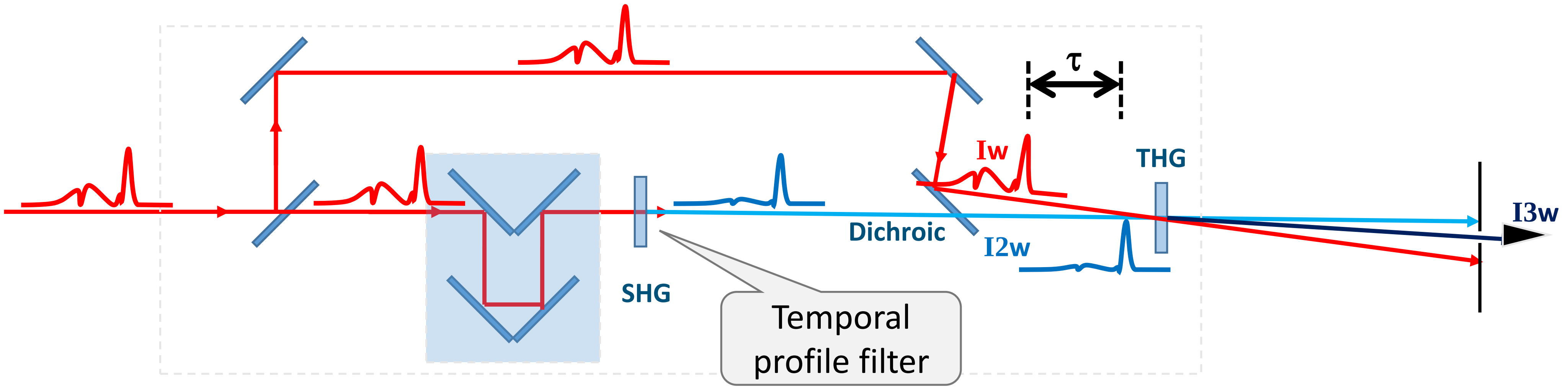
Lens images position in crystal (i.e., delay) to horizontal position at camera

Lens maps angle (i.e., wavelength) to vertical position at camera

Grenouille needs a very uniform spatio-temporal profile

https://frog.gatech.edu/tutorial.html

Non-symmetric FROG method : THG FROG



THG FROG

- FROG between a pulse and its second harmonics
- The SHG cleans the temporal profile and shorten the pulse
- The measured signal closer to the main initial pulse
- No phase sign indetermination

Not easy to perform:

- The SHG step has to be very well controlled (spectral acceptance, chirp difference, uniformity)
- The natural non-linearity of the process reduces the working energy range
- The analysis strongly depends on the parameters of the interferometer
- The spectrometer has to be sensitive at around 3ω

General X-FROG method

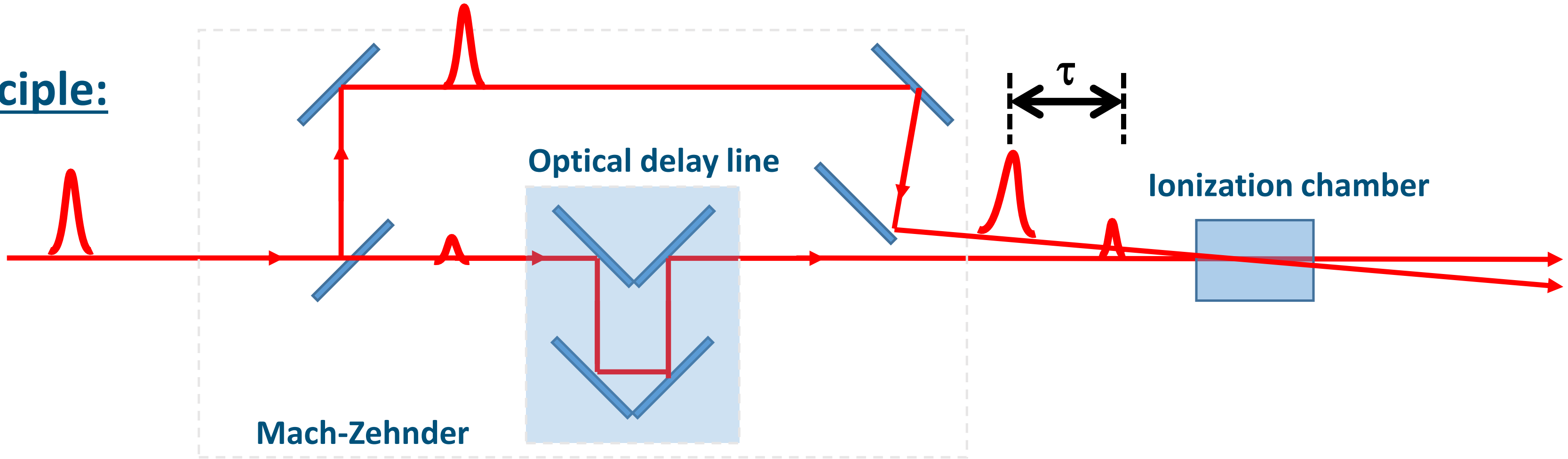
Cross FROG (X-FROG)

- Can be applied when the two pulses are temporally stable each other
- 4 unknown : Amplitude and phase of the two pulses ($A_1(\omega), \phi_1(\omega), A_2(\omega), \phi_2(\omega)$)
- X-FROG measure two “independent” dimensions (time, spectrum)
- By measuring at the spectrometer ($A_1(\omega),\ A_2(\omega)$) the phases ($\phi_1(\omega), \phi_2(\omega)$) can be retrieved
- *In situ* measurements in femtochemistry
- Hard to perform in practice because of the non-uniformity of the pulses after frequency conversion

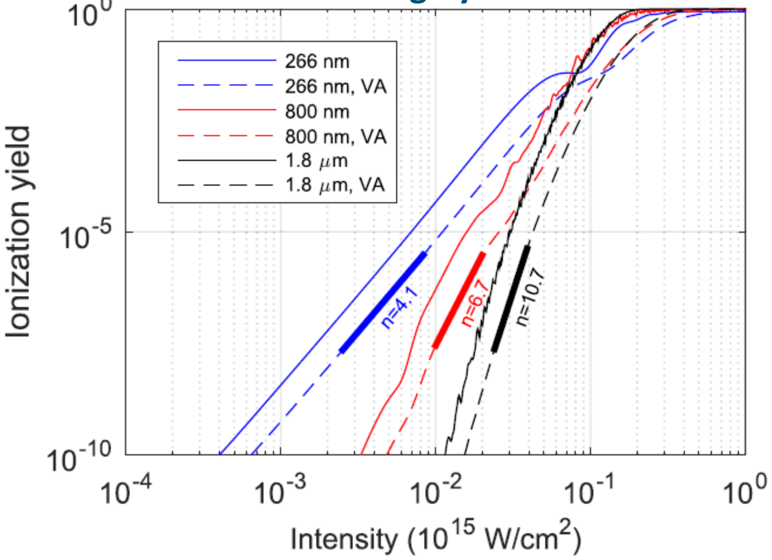
Time scanning techniques:
Tip-Toe

Tip-Toe (Tunneling Ionization with a Perturbation for the Time-domain Observation of an Electric field)

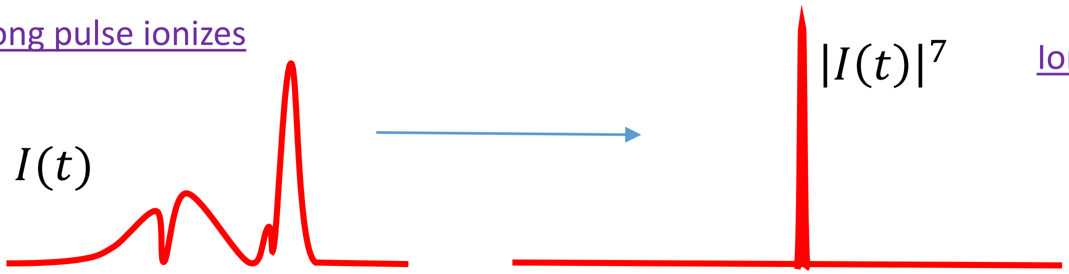
Principle:



Ionization is highly non-linear



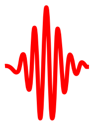
A strong pulse ionizes



Ionization makes an ultrashort gate

$$ions \propto |I(t)|^7$$

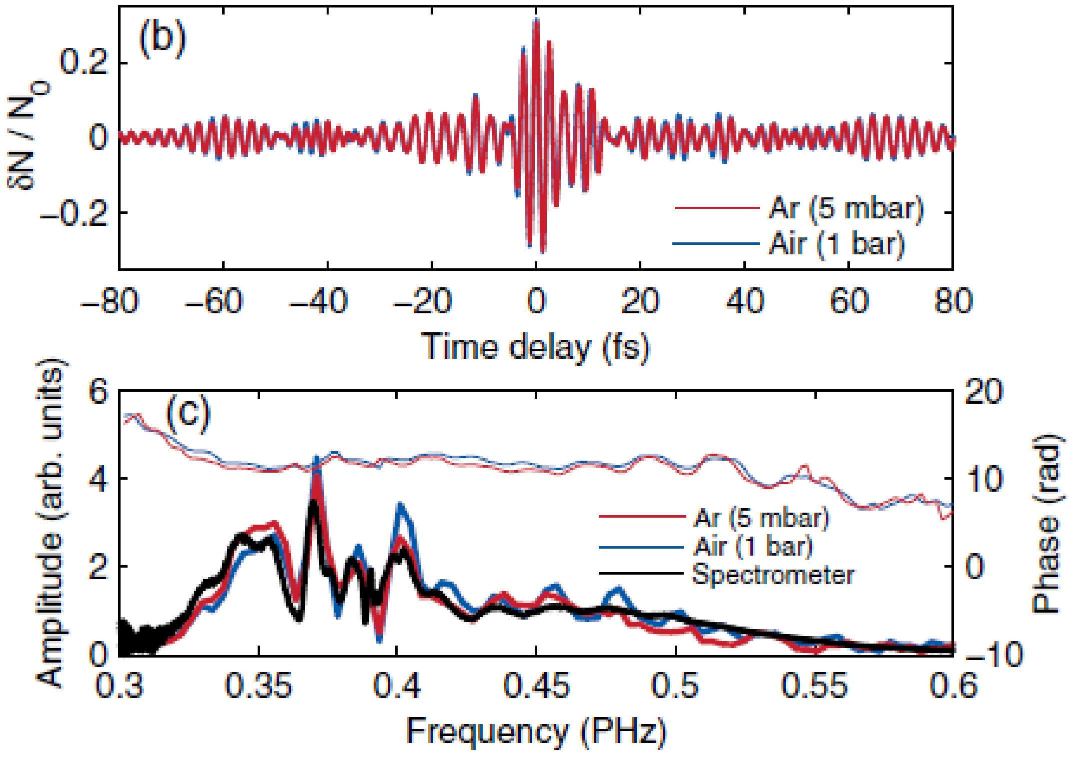
A weak replica modulates the ionization

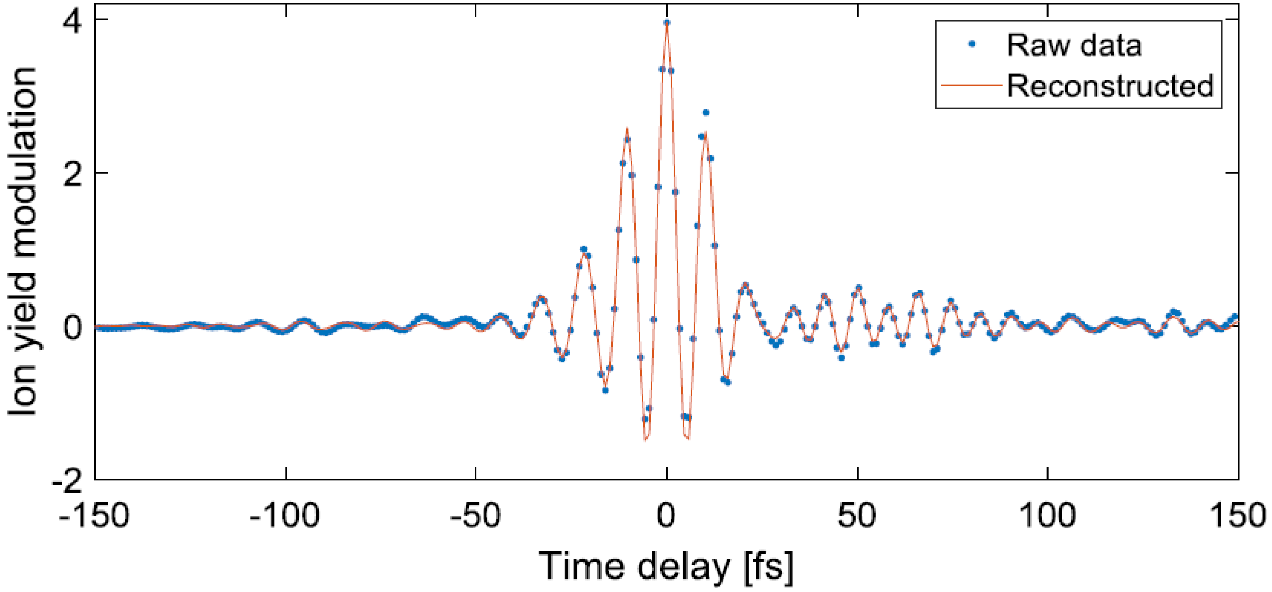


$$ions \propto \left|E_{strong}(t) + E_{weak}(t-\tau)\right|^{14}$$

S. B. Park *et al.* Optica **5** 402 (2018)
W. Cho *et al.* Sci. Rep. **9** 16067 (2019)

Tip-Toe Direct imaging of the electric field





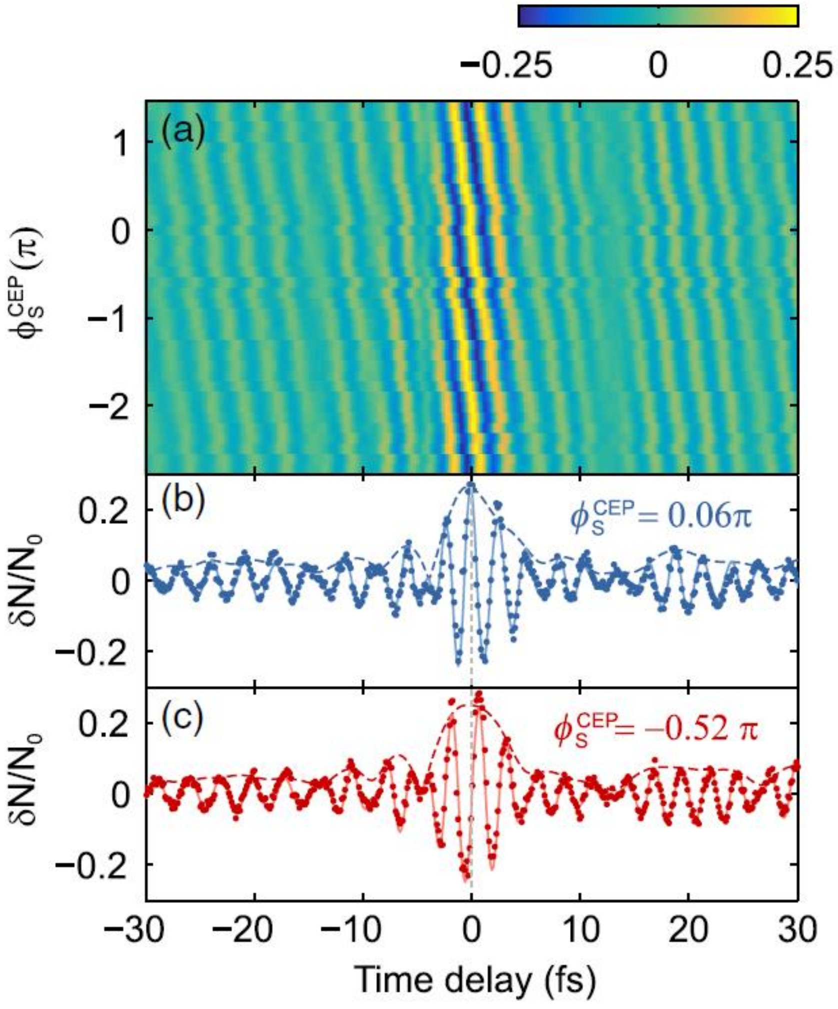
<u>Direct measurement of the electric field:</u>

- Spectral intensity and phase from Fourier Transform
- Well suited with mass/photoelectron spectroscopy

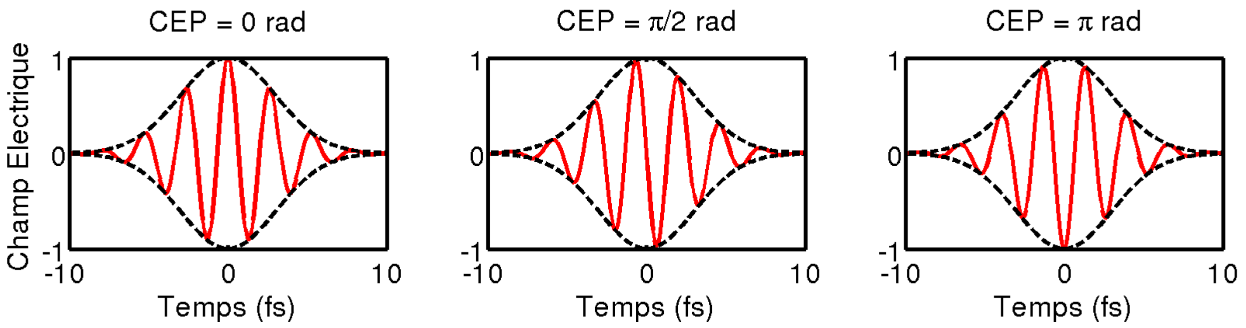
- Need high laser stability (non-linearity)
- Need very stable delay line

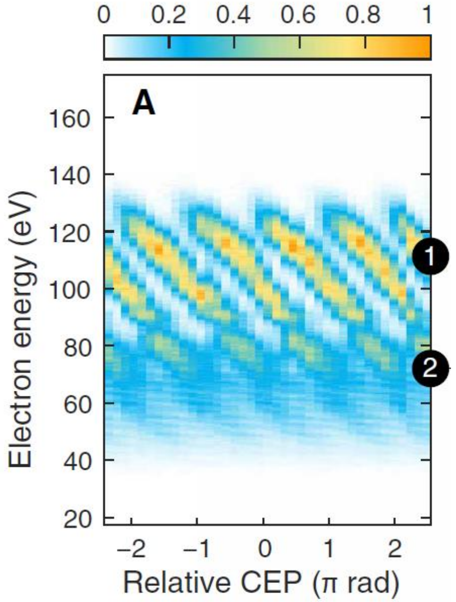
S. B. Park *et al.* Optica **5** 402 (2018)
M. Kurucz *et al.* Opt. Com. 472 126035 (2020)

Tip-Toe CEP sensitive



The direct reconstruction of the electric field allows the visualization of the Carrier Envelope Phase

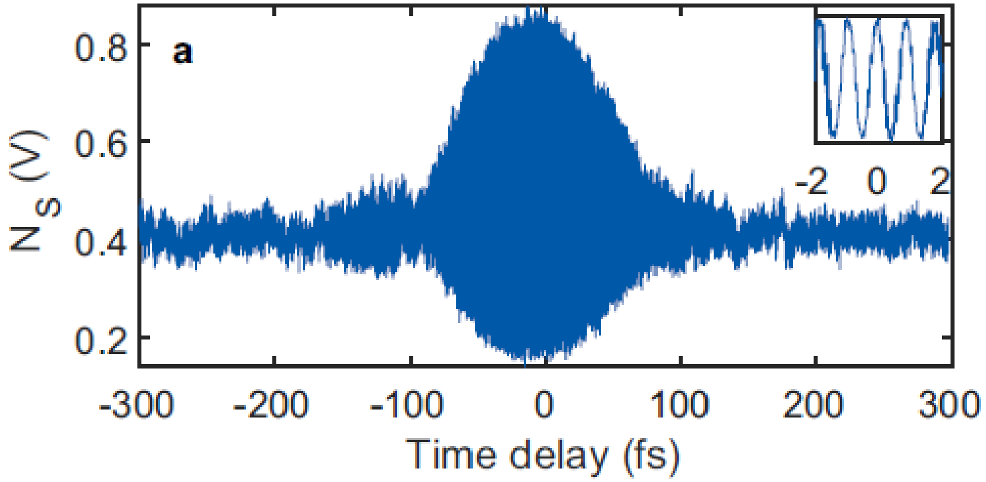


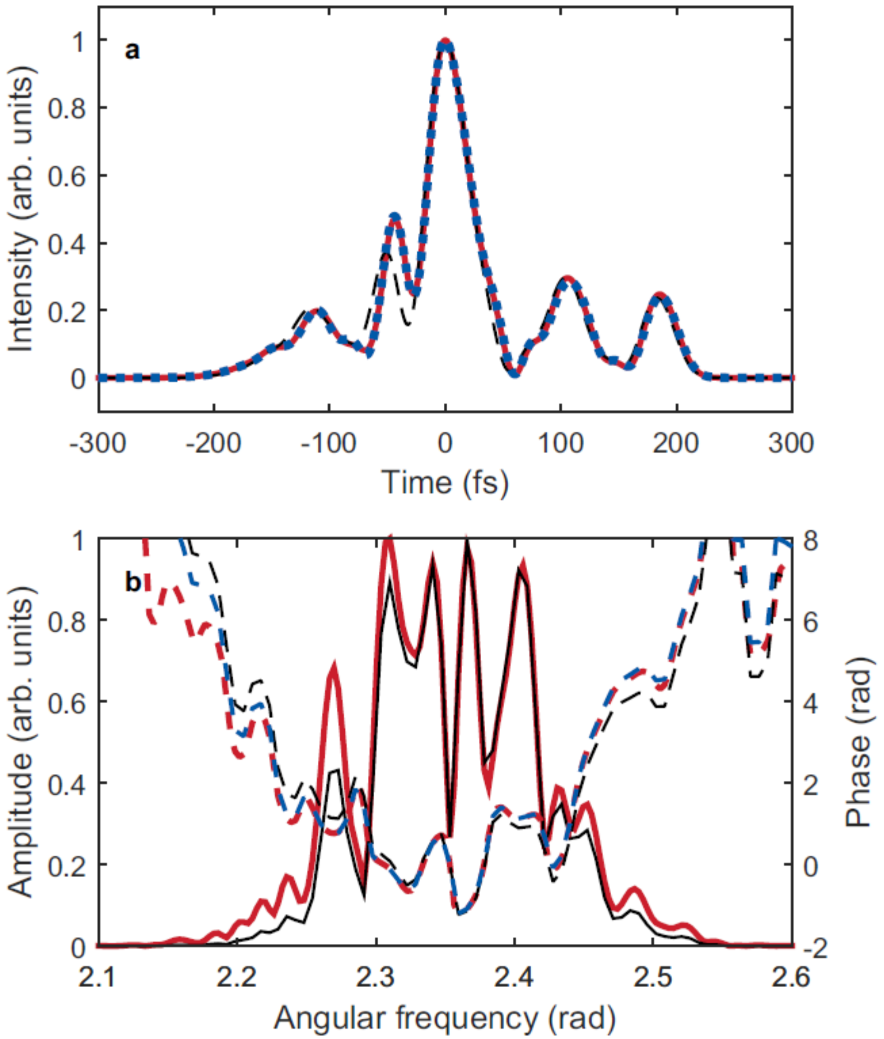


As many highly non-linear effect, high harmonic generation is (HHG) highly dependent on the CEP

S. B. Park *et al.* Optica **5** 402 (2018)
W. Cho *et al.* Sci. Rep. **9** 16067 (2019)

Tip-Toe long pulses



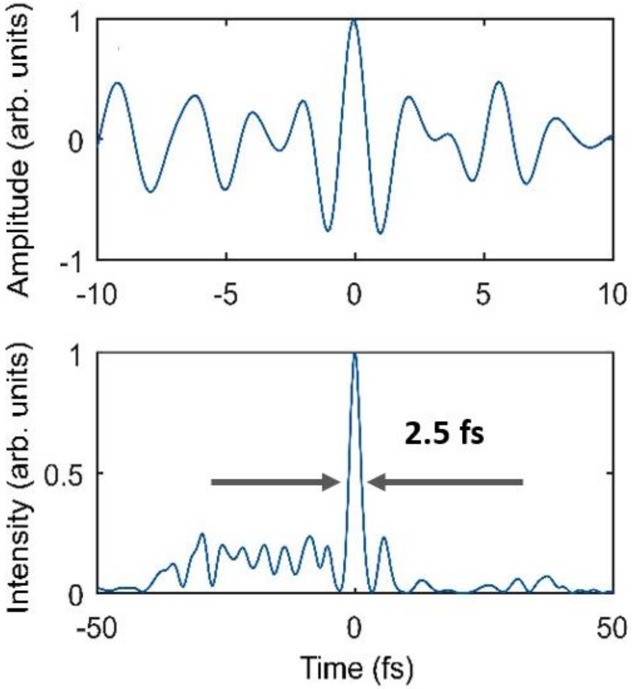


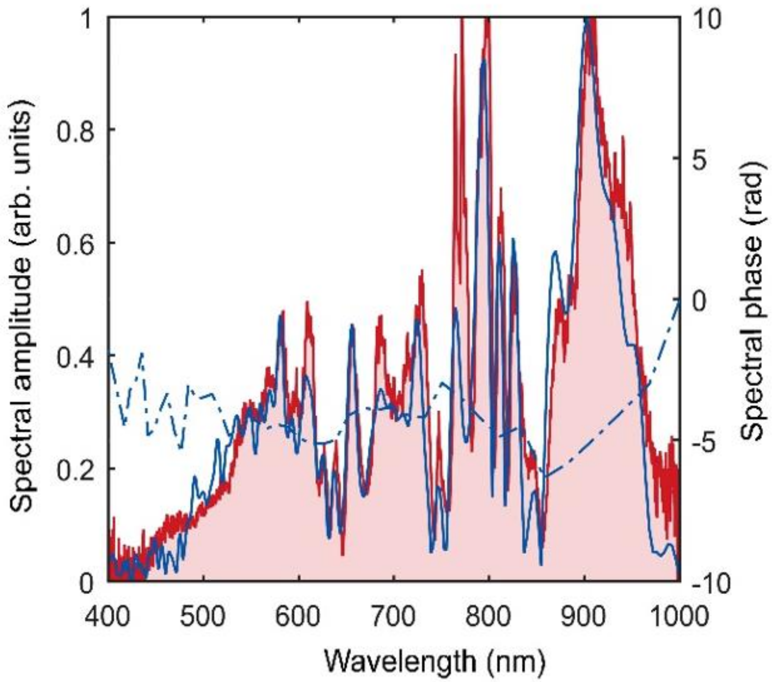
Tip-Toe measurement is not limited to few-cycle pulses. Pulses of tens of fs can also be measured

W. Cho *et al.* Sci. Rep. **9** 16067 (2019)

Measurement of complex pulses

Operation:
Pulse duration: 0.5 fs – 2 ps
Wavelength: 266nm-10µm
Min pulse energy (@50fs):
2µJ @ 400nm
10µJ @ 800nm
60µJ @ 10µm
Scan 2-10s @1 kHz

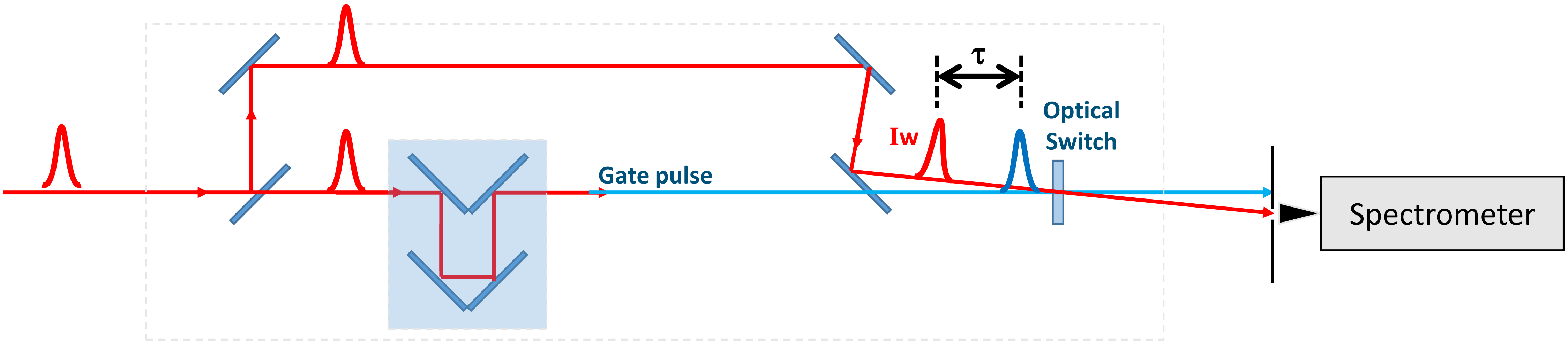




S. I. Hwang *et al.* Sci. Rep. **9** 1613 (2019)

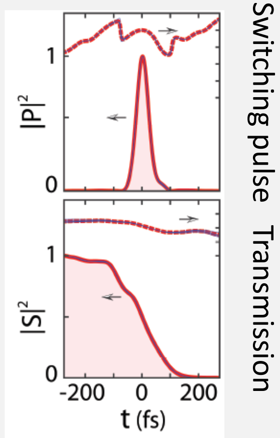
Time scanning techniques:
FROST

Principle of FROST (FRequency Resolved Optical SwiTching)

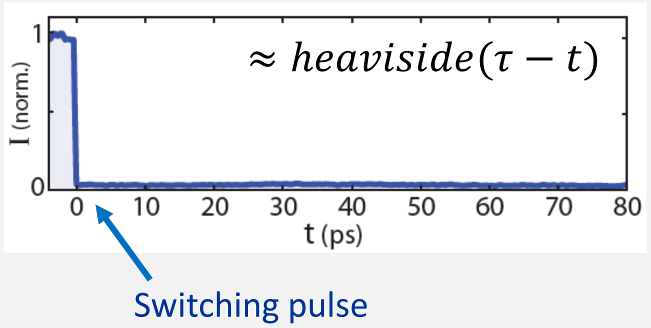


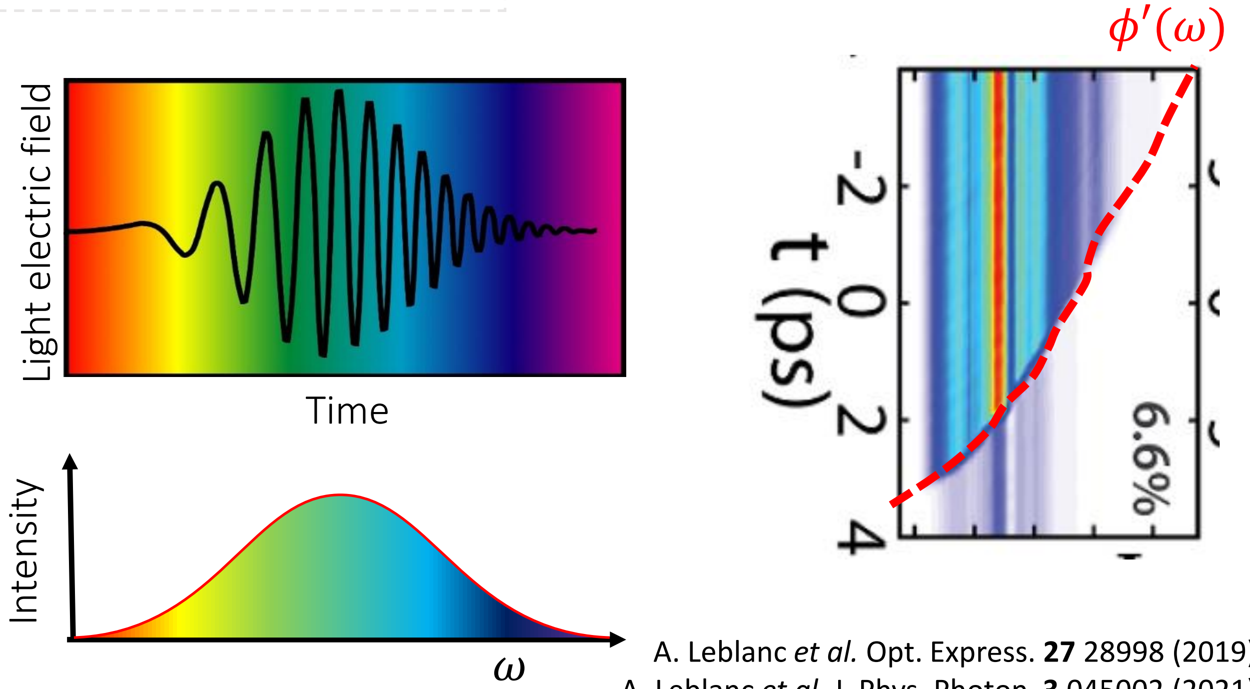
Optical switch

- The optical transmission suddenly vanishes after absorption of a pulse producing a temporal gate ("Heaviside-like")



Transparency :

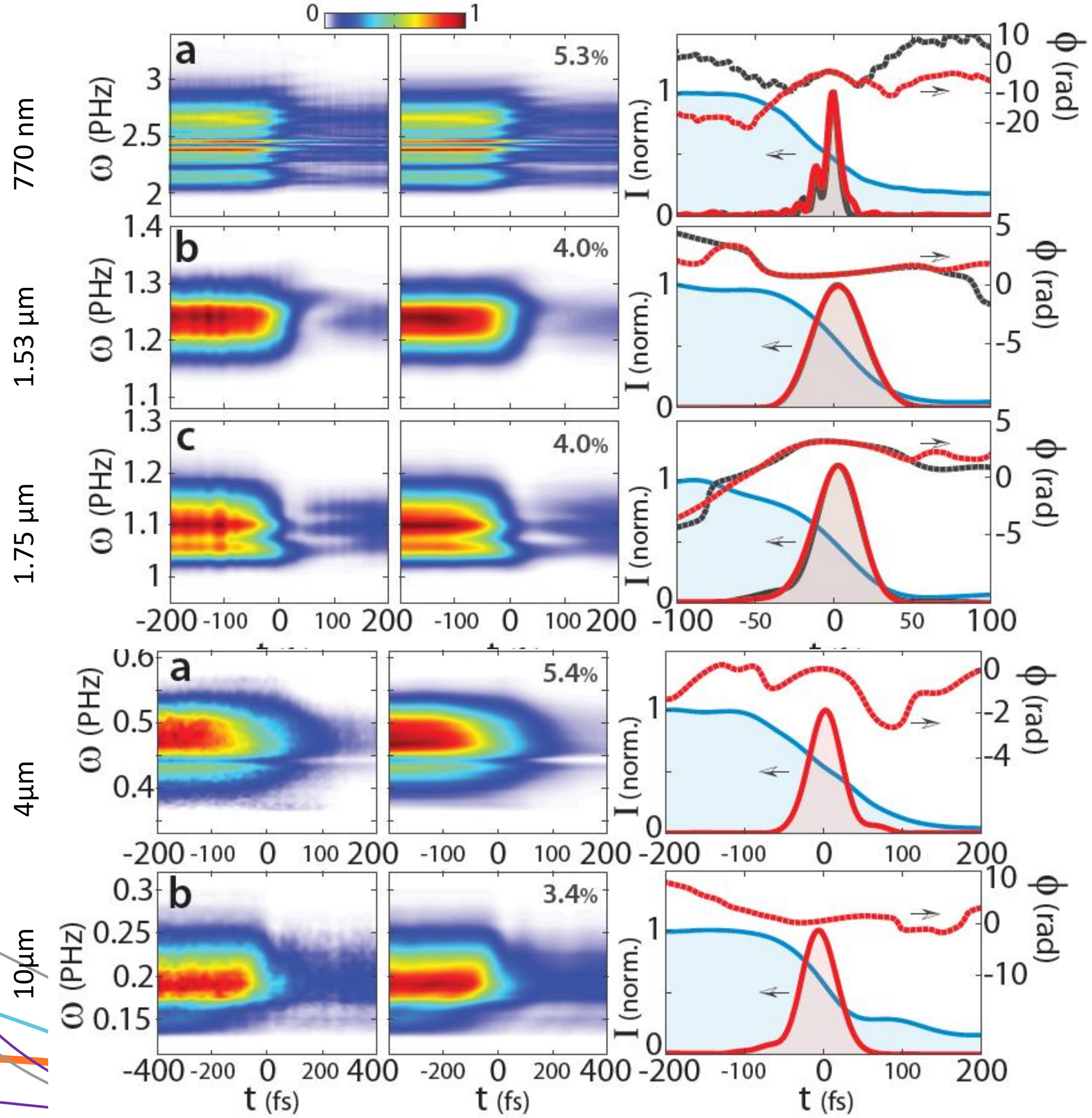




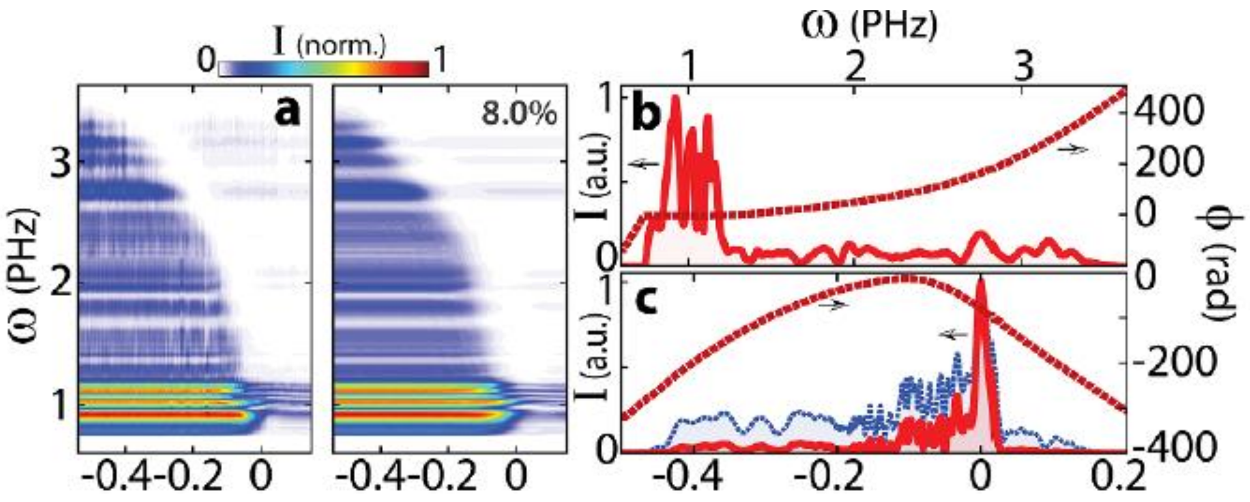
A. Leblanc *et al.* Opt. Express. **27** 28998 (2019)
A. Leblanc *et al.* J. Phys. Photon. **3** 045002 (2021)

Performances of FROST

Central wavelength work principle

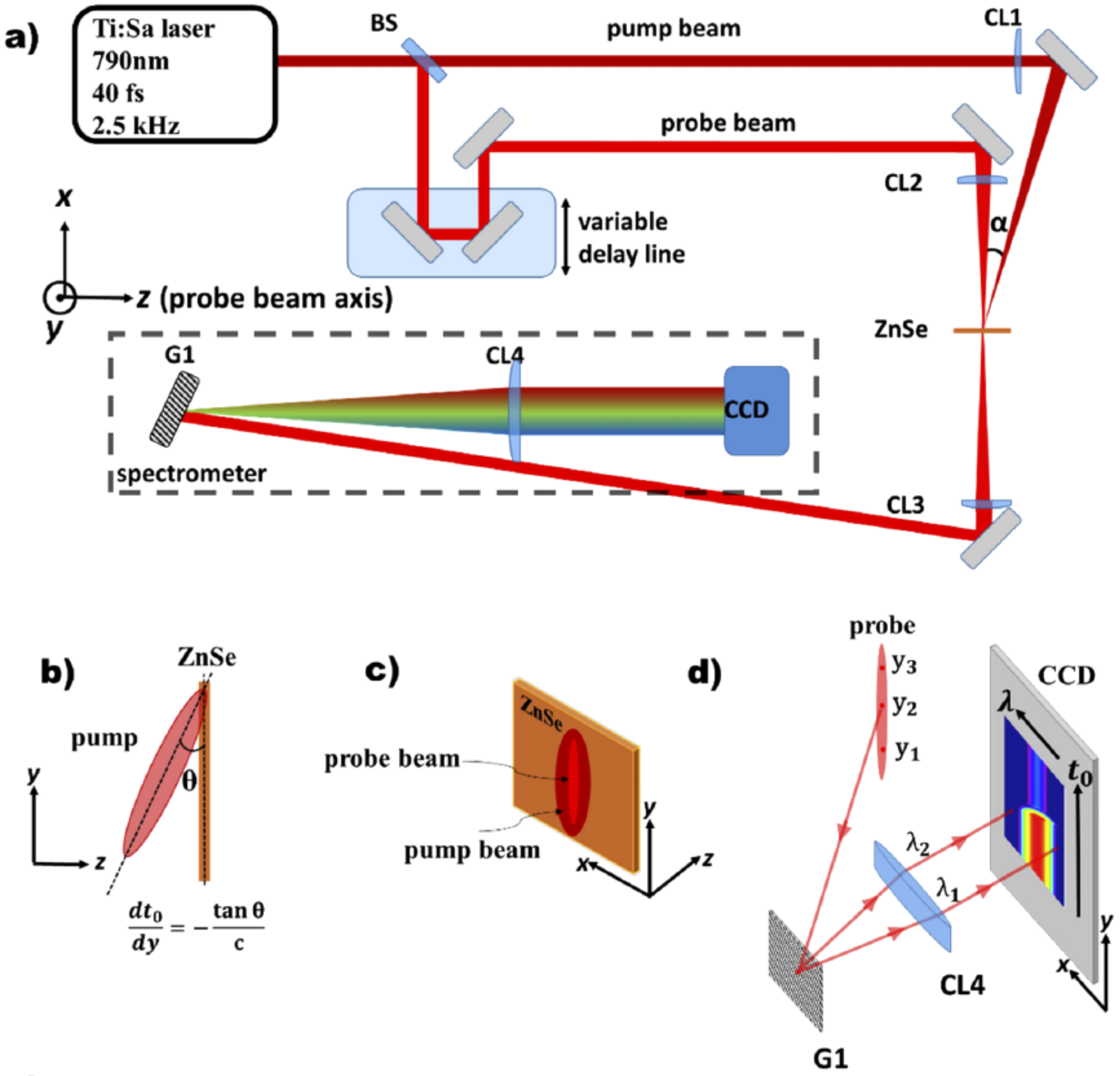


Two octave spanning pulse characterization



A. Leblanc *et al.* Opt. Express. **27** 28998 (2019)
A. Leblanc *et al.* J. Phys. Photon. **3** 045002 (2021)

Single shot FROST



Delay introduced the incident angle

Using a pump pulse that has a tilt, the gate would close with a time that linearly depends on the position on the crystal

More information on FROST

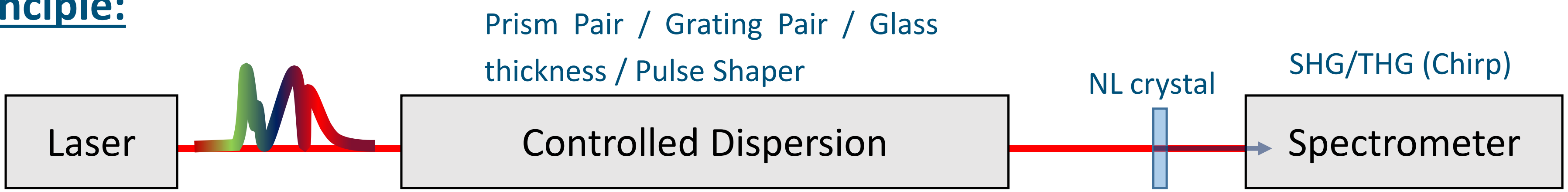
Tomorrow by A Leblanc

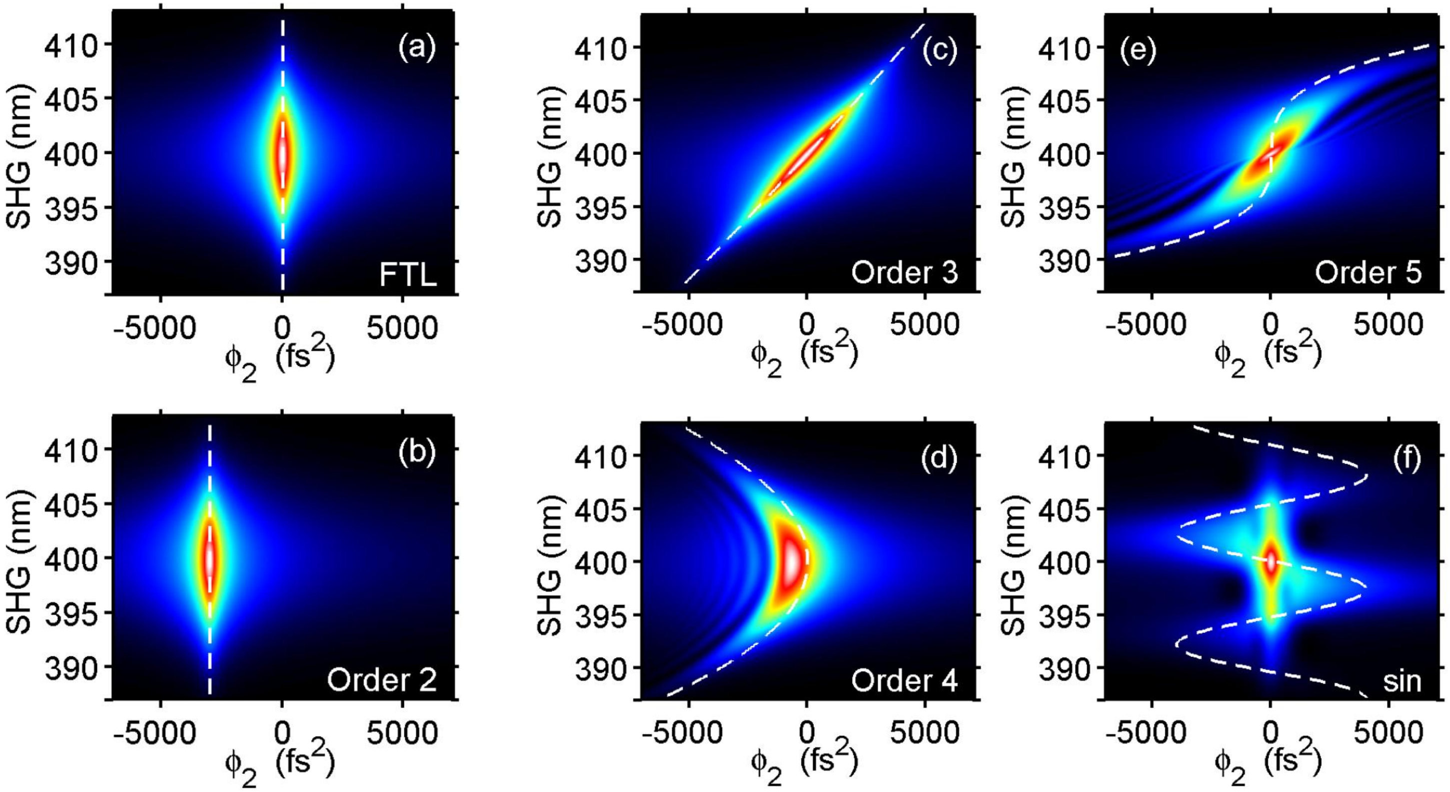
A. Leblanc *et al.* Opt. Express. **28** 35807 (2020)

Chirp Scans
D-Scans

Chirp Scans

Principle:





Movie

Easy to implement

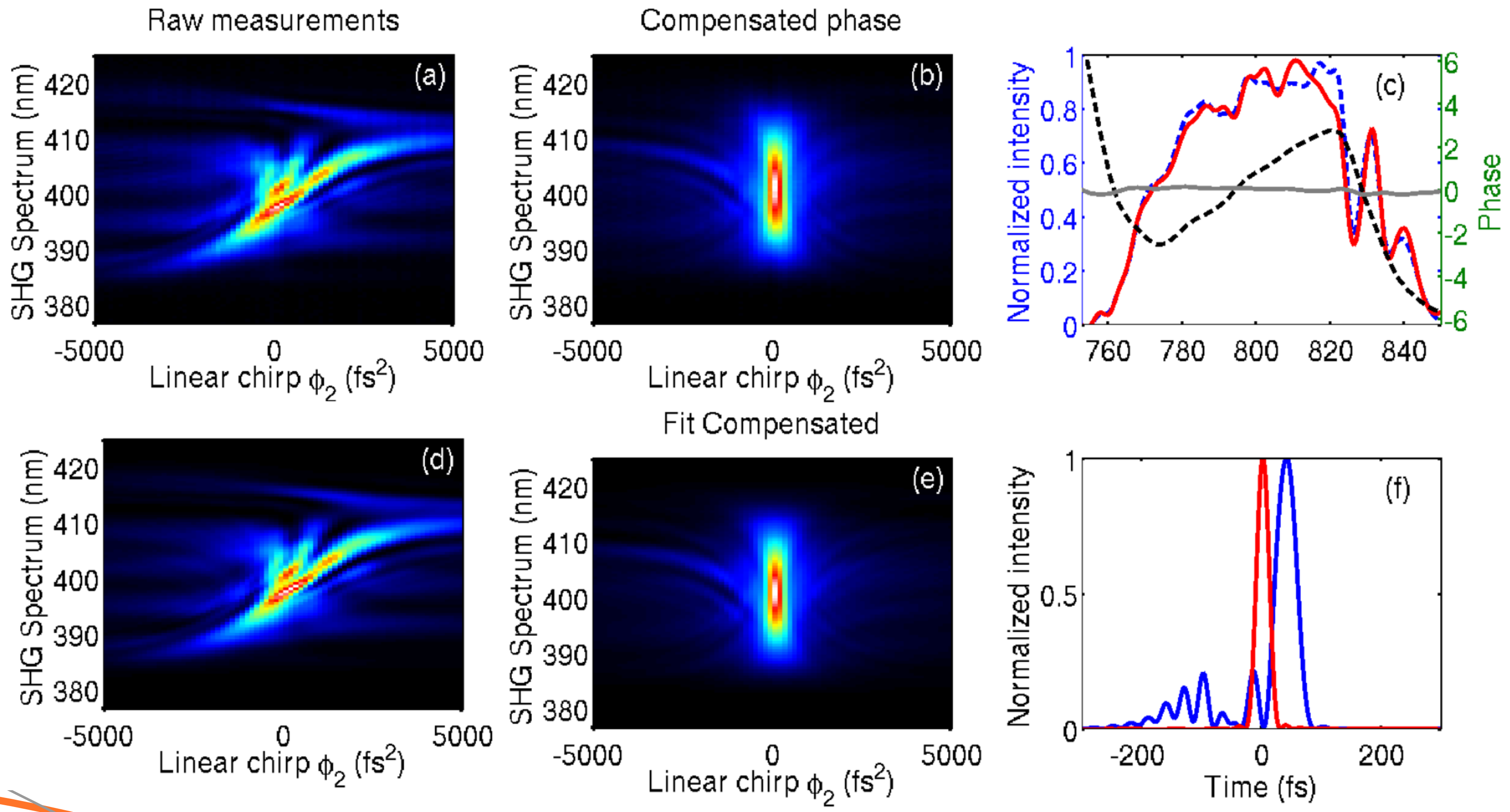
- No beam splitting
- No recombinaiton

Intuitive

- $\phi_2(\omega) \approx -\max\left(I_{2\omega}(\omega)\right)$
- Easy to identify the Taylor phase order

V. Loriot *et al.* Opt. Express. **21** 24879 (2013)
M. Miranda *et al.* Opt. Express. **20** 688 (2012)
M. Hoffmann *et al.* Opt. Express. **22** 5234 (2014)
V. V. Lozovoy *et al.* Opt Express. **16** 592 (2008)

Chirp Scans – 25 fs strong distortion

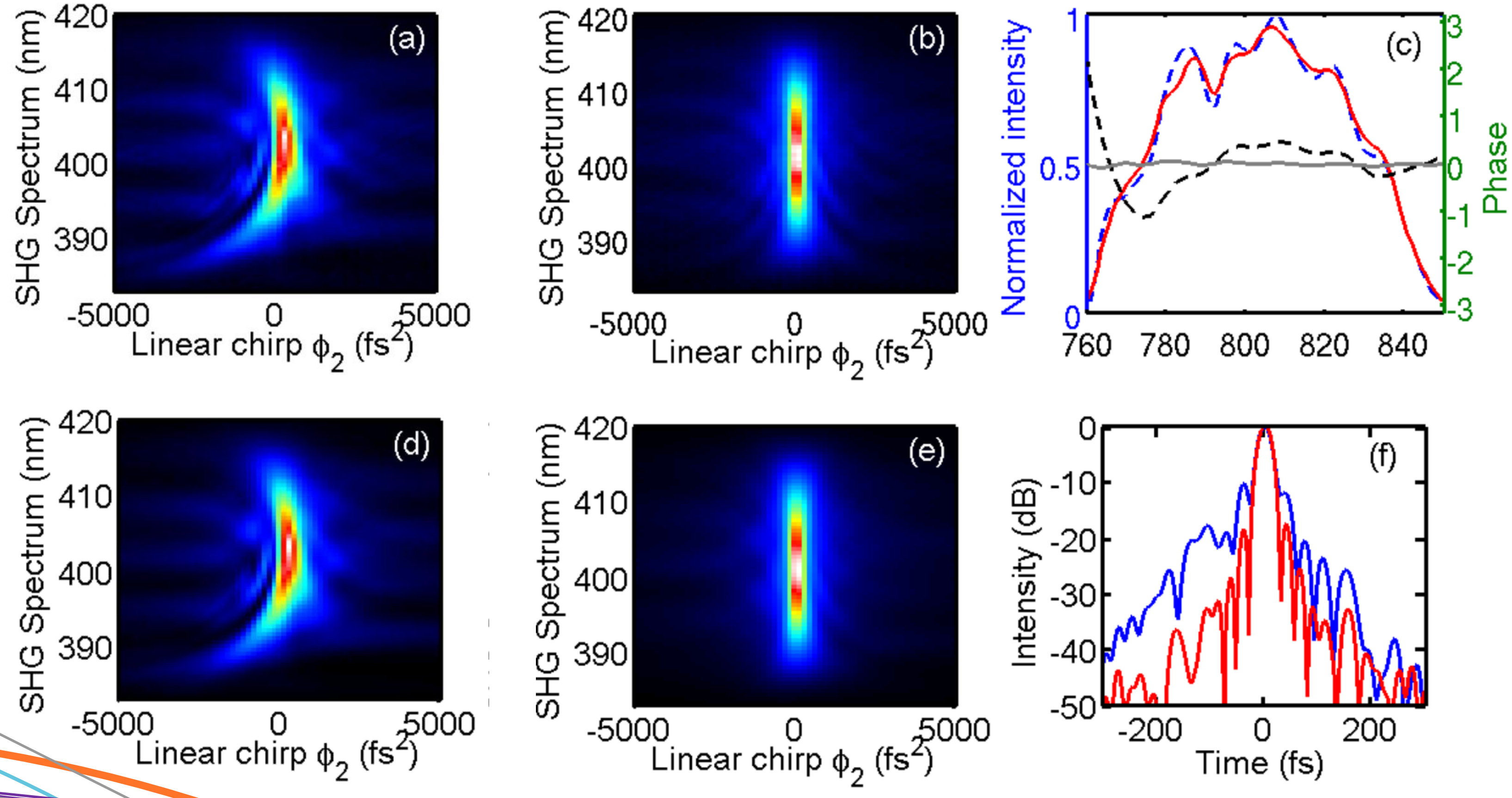


No iteration

- Pulse optimized in a single measurement
- **Amplitude and phase reconstruction**
- If phase can be compensated (dazzler), the pulse can be LTF iteration-less from a big distortion

V. Loriot *et al.* Opt. Express. **21** 24879 (2013)

Chirp Scans – 25 fs small distortion



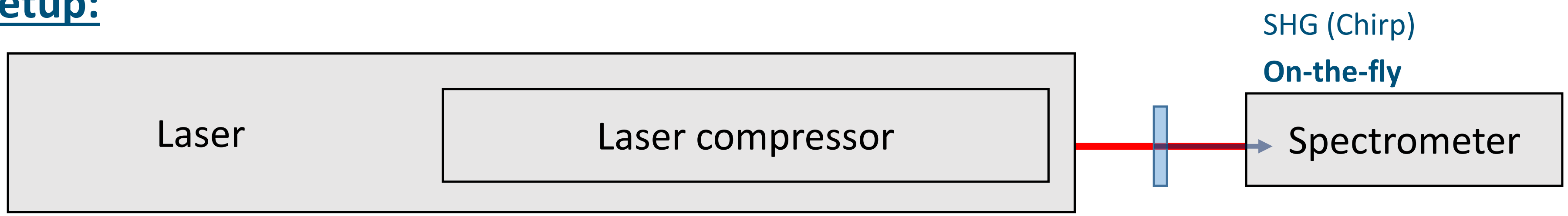
Accuracy

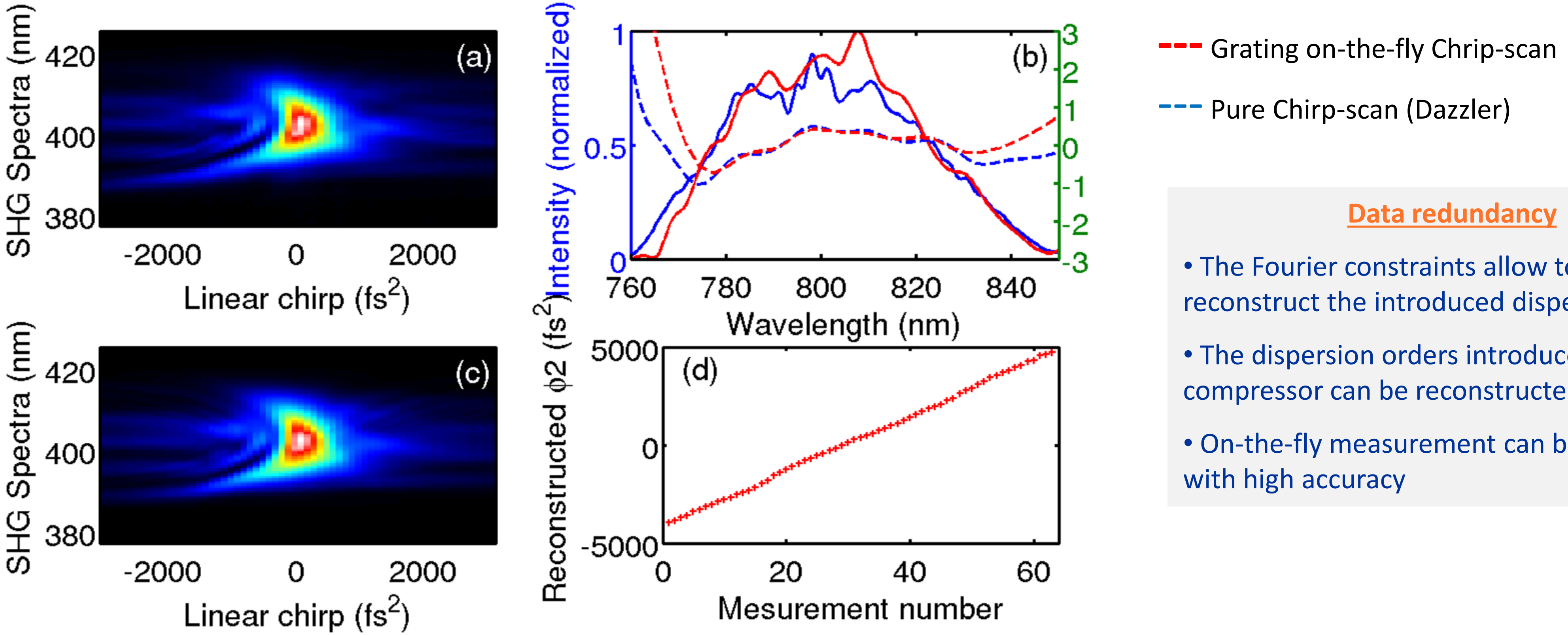
- Even small distorsion makes strong image changes.
- Well suited for pulse optimization

V. Loriot *et al.* Opt. Express. **21** 24879 (2013)

Chirp Scans – 25 fs Chirp retrieval

Setup:





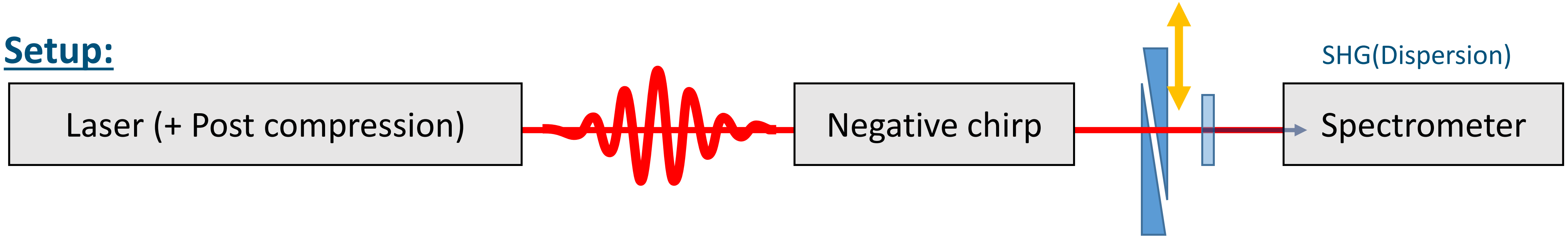
Data redundancy

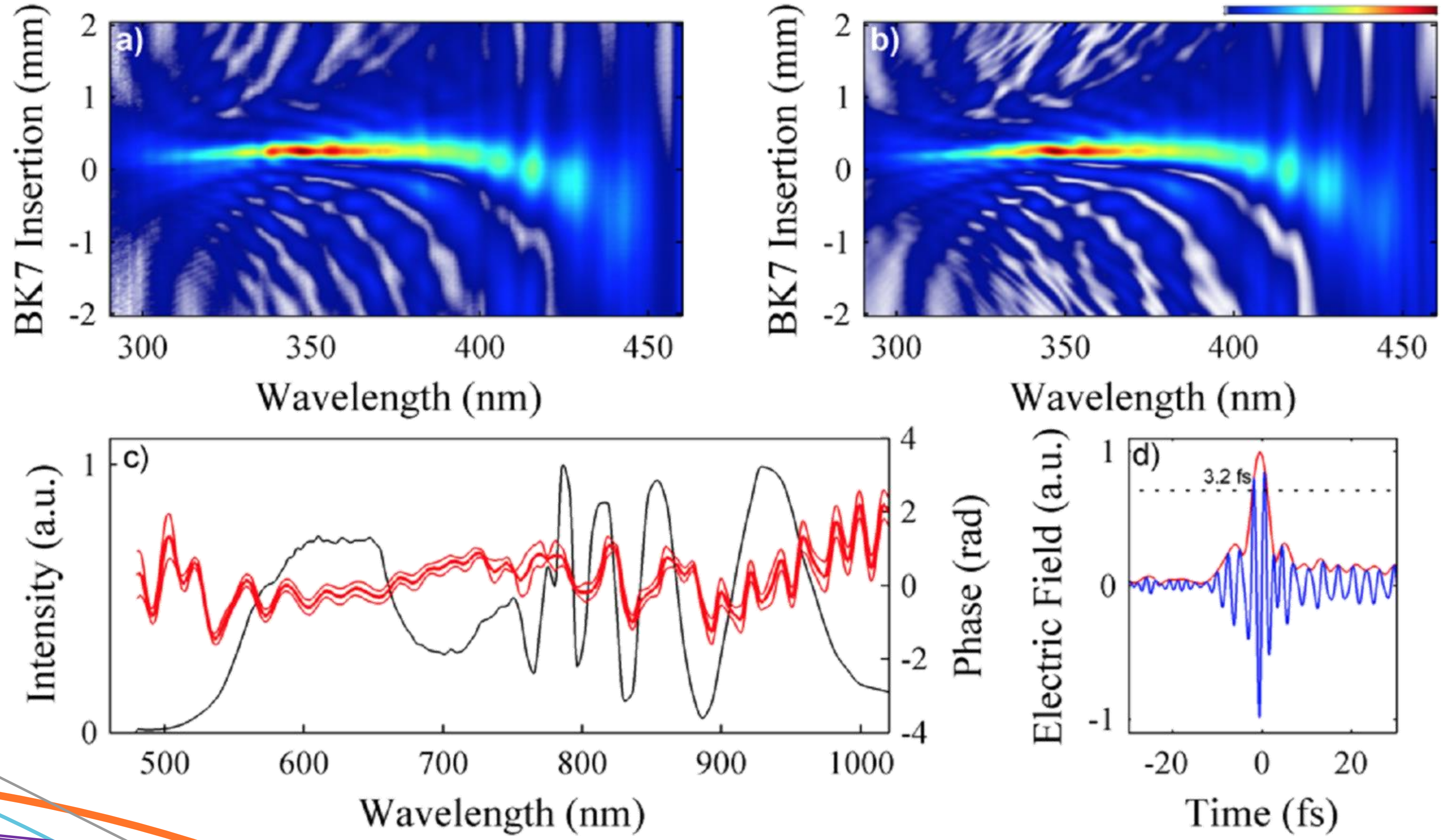
- The Fourier constraints allow to reconstruct the introduced dispersion
- The dispersion orders introduced by the compressor can be reconstructed
- On-the-fly measurement can be performed with high accuracy

V. Loriot *et al.* Opt. Express. **21** 24879 (2013)

Chirp Scans – 1.4 optical cycle

Setup:





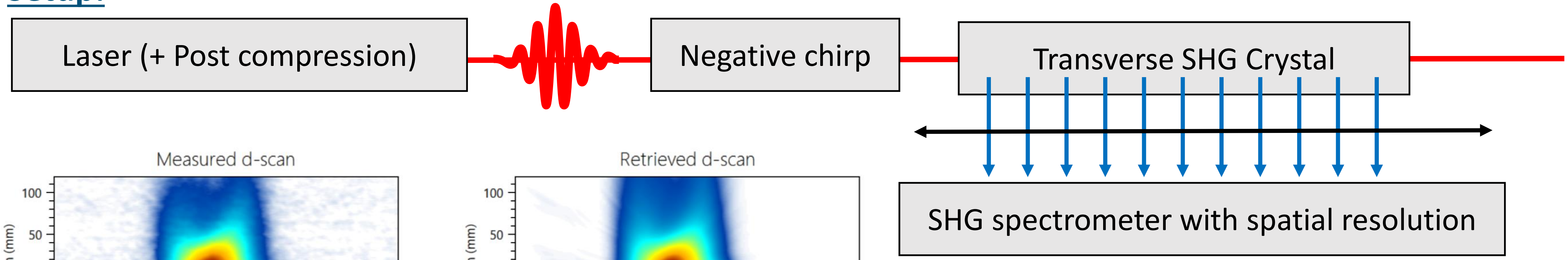
Few cycle regime

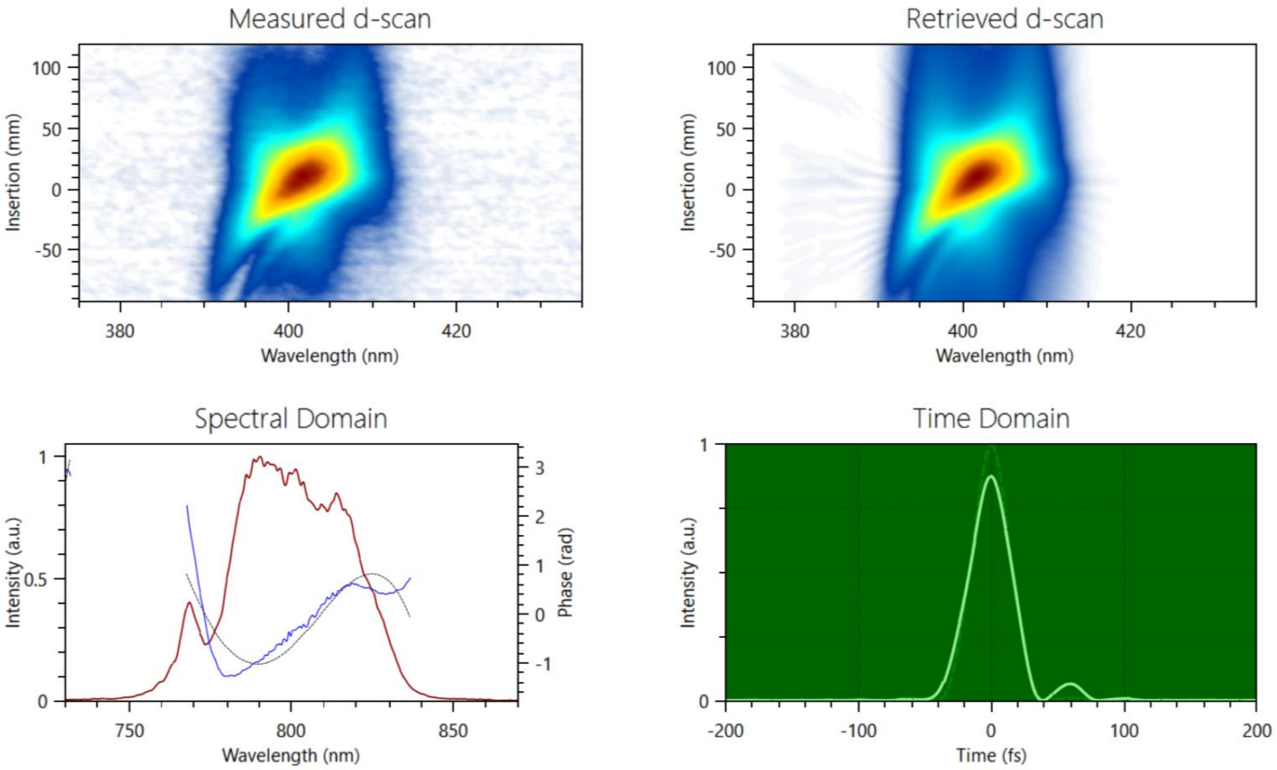
- Well suited on the few cycle regime
- Dual pulse measurement and optimization for the experiment
- Dispersion can be introduced with glass dispersion

F. Silva *et al.* Opt. Express. **22** 10181 (2014)

Single shot Chirp Scans

Setup:





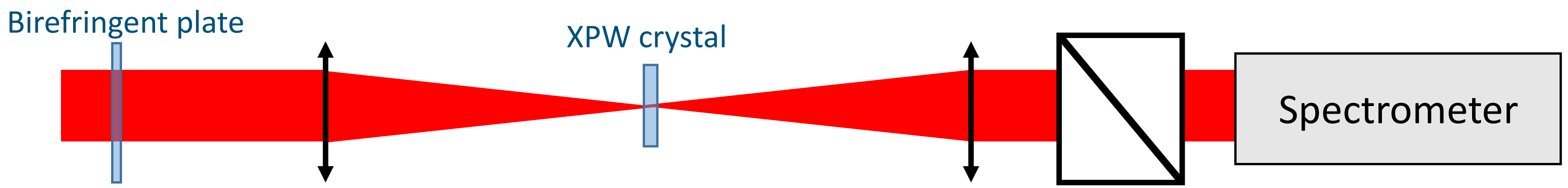
Dual Chirp and SHG crystal

- Transverse SHG produces SHG at 90° from the propagation axis
- The NL-crystal introduces a chirp by itself through its propagation
- Spatially resolved spectrometer allows to capture the SHG as a function of the chirp

F. J. Salgado-Remacha *et al.* Opt. Letts. 45 3925 (2020)

SRSI - Wizzler

Self Referenced Spectral Interferometry (SRSI)



<u>Tick birefringent plate</u>

- Create a pulse replica shifted in time on the perpendicular polarization
- Controlled chirp

<u>XPW (cross-polarized wave generation)</u>

$$\chi^{(3)}(-\omega_0; \omega_0, -\omega_0, \omega_0)$$

- BaF_2 for 800 nm
- Broaden the spectra
- Clean-up the pulse
- Generate on the perpendicular polarization

<u>Polarizer and spectrometer</u>

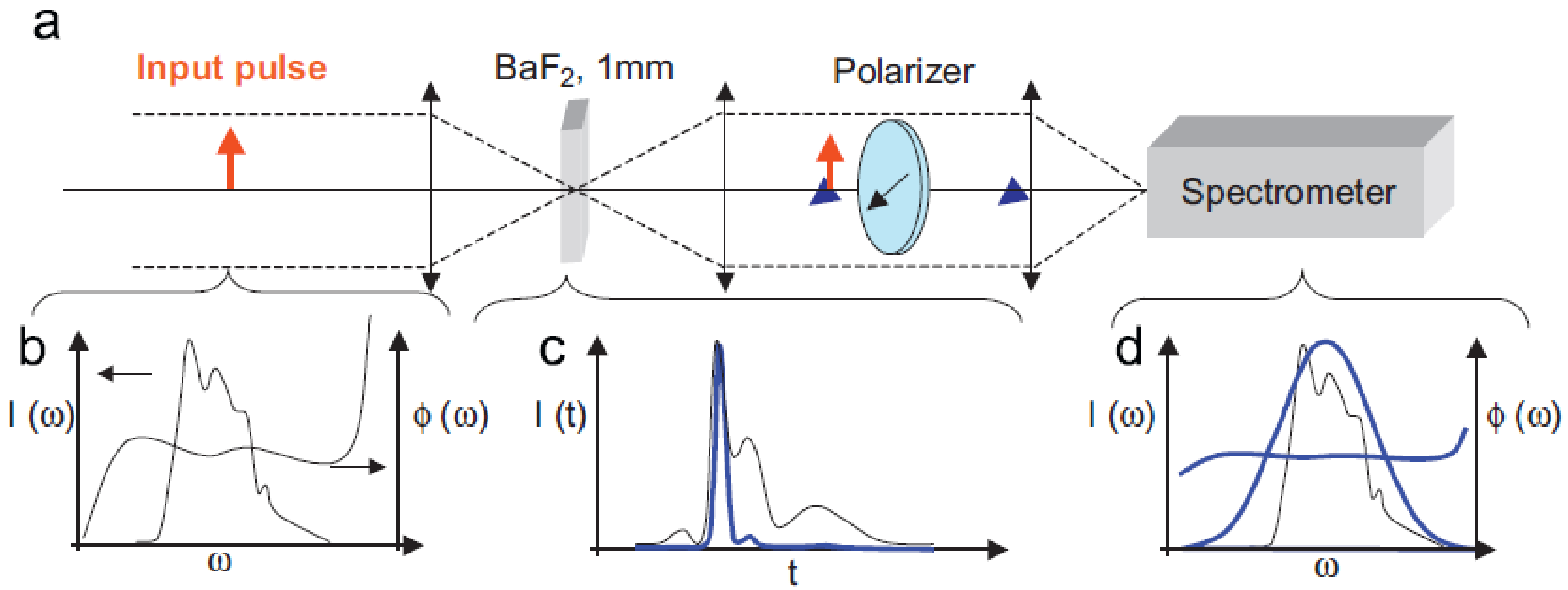
Measure the interference between:

- Pulse replica from the birefringent plate
- Pulse cleaned by the XPW

- In line operation (easy alignment)
- Non-iterative reconstruction algorithm
- Need well polarized incoming pulse (add polarizer)

T. Oksenhendler *et al.* Appl. Phys. B (2010)

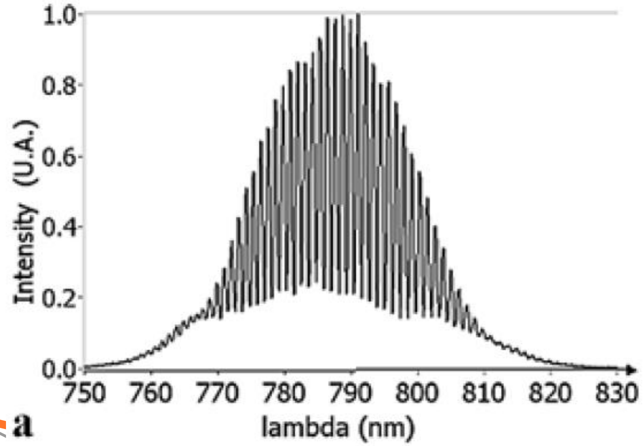
SRSI principle



Initial spectrum

Temporal cleaning

Spectral broadening and phase flattening
Linear interferometry between the pulses

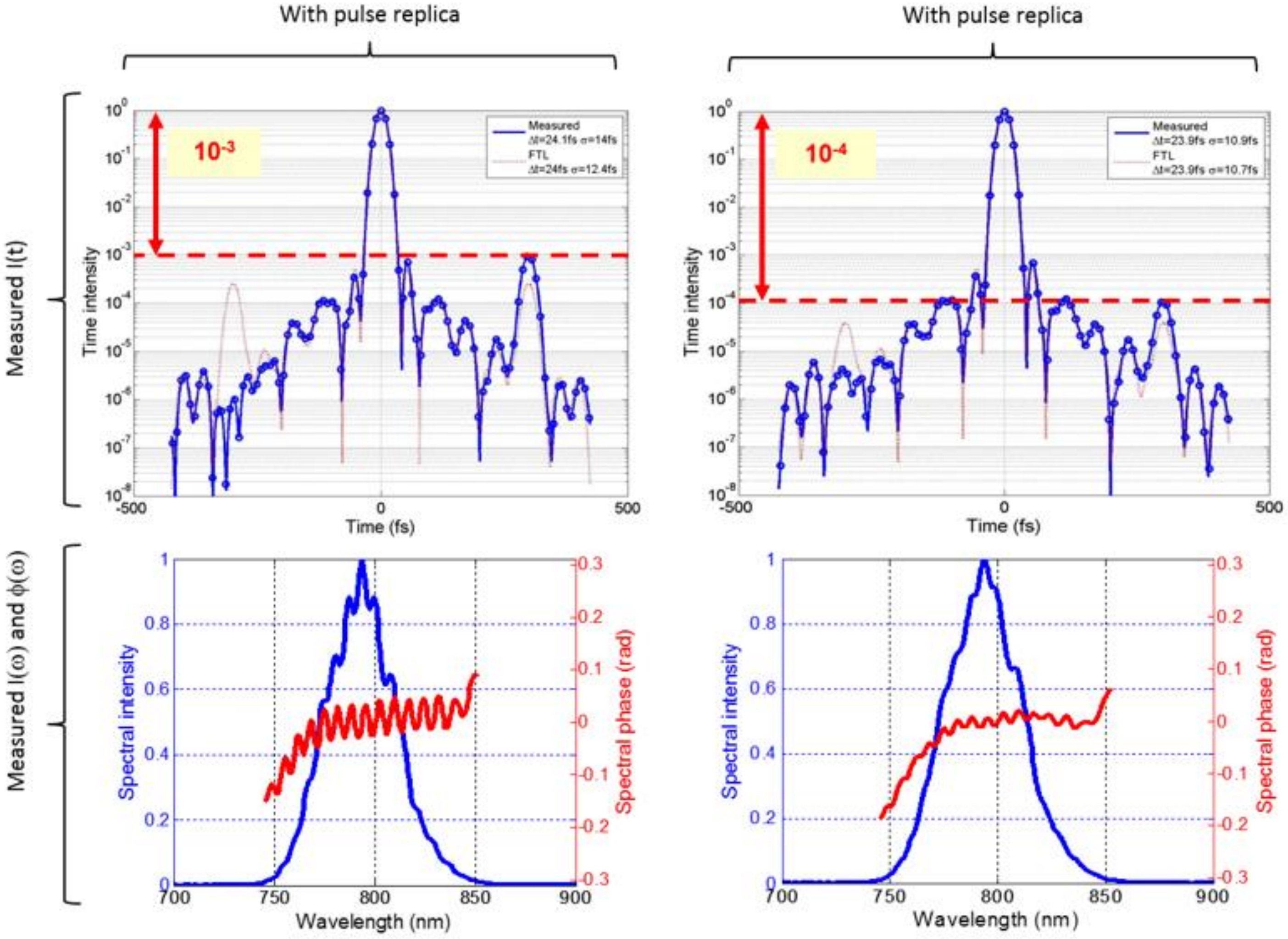


Interference between the pulse $I(t)$ and its XPW cleaned-up version $|I(t+t_0)|^3$ (almost Fourier transform)

Twice the dynamic range of the spectrometer

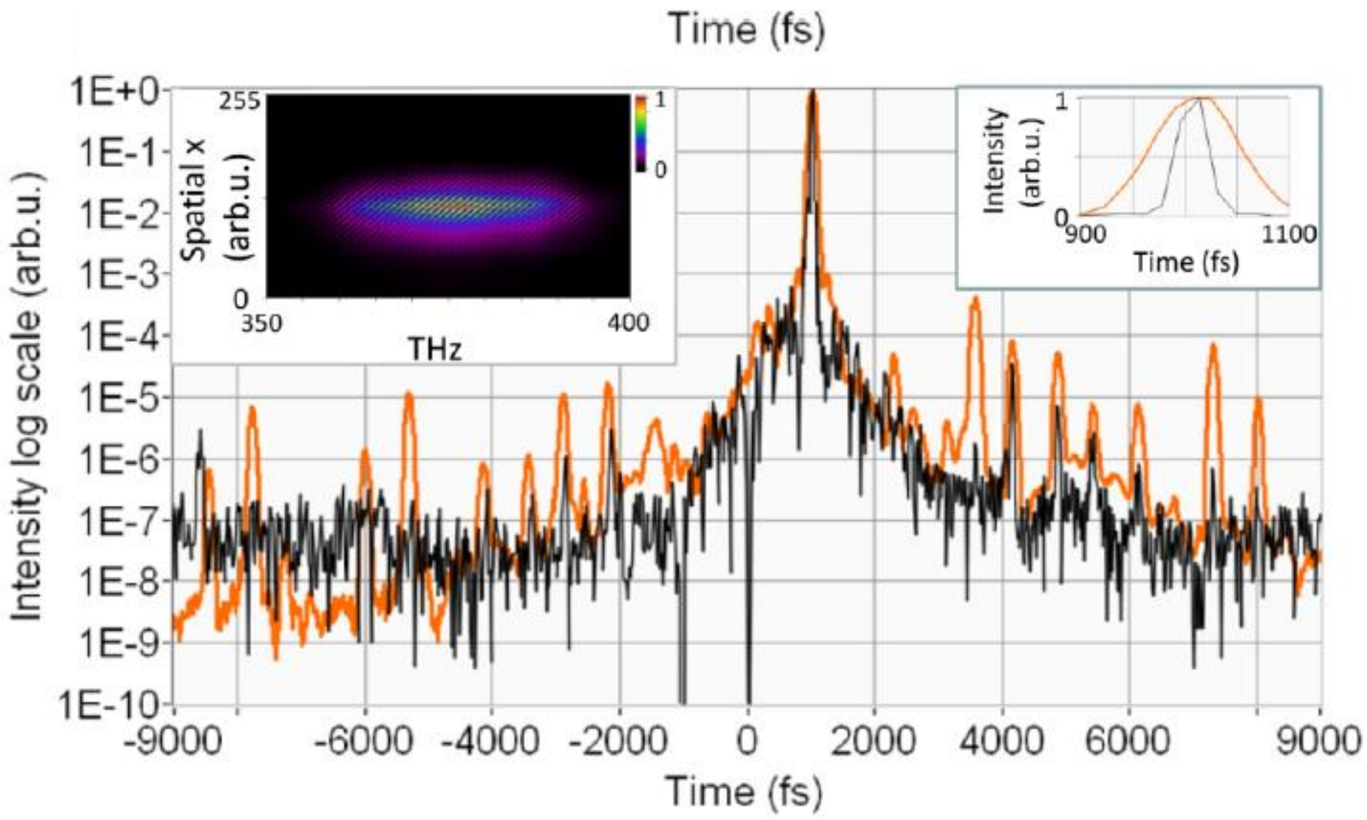
S. Grabielle *et al.* Nu. Instru. Meth. Phys. Rch. A **653** 121 (2011)

Standard SSRI (Wizzler)



High dynamic range and large temporal windows SRSI

18 ps temporal range
10^8 dynamic range
20 fs shortest pulse



--- Ultrashort laser pulse

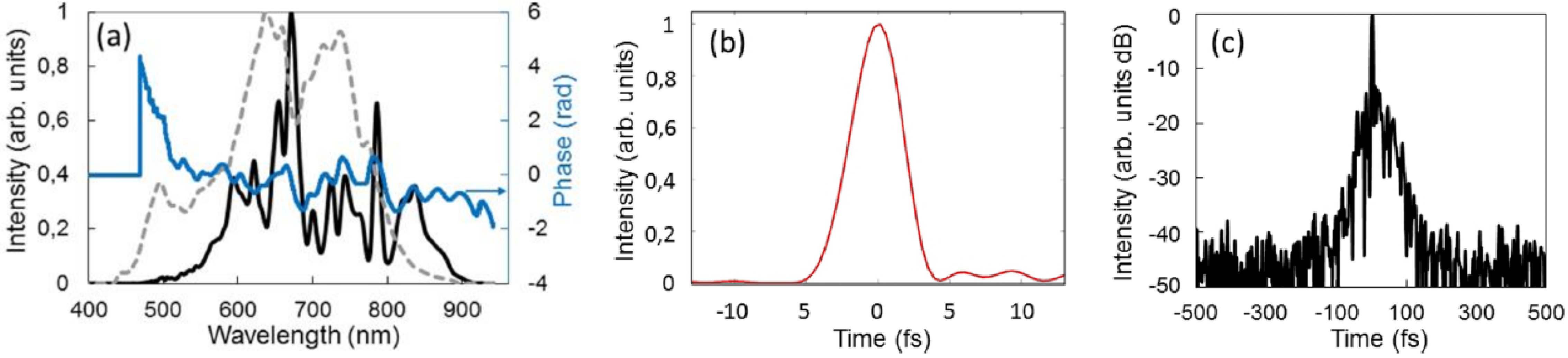
--- XPW cleaned

Tuning the SRSI properties

- The temporal range is given by the delay introduced by the birefringent plate
- A broad temporal range requires high resolving spectrometer $\Delta\nu = 1/(t_{max} - t_{min})$
- The dynamic range is limited by the spectrometer dynamic range

T. Oksenhendler *et al.* Opt. Express **25** 12588 (2017)

Few cycle SRSI



Few cycle pulses

- Few cycle pulses have almost an octave bandwidth
- Perpendicular polarization prevent the overlap between the initial and final pulse
- Calibration of the XPW, calcite, and spectrometer is a key for few-cycle regime

A. Trabattoni, *et al.* Rev. Sci. Instrum. **86**, 113106 (2015)

Conclusions

- Photodiodes based systems are too slow to resolve the temporal profile of a femtosecond pulse

- There is no ideal technique that measure all parameters at once :
  - From few cycle to tens of picosecond pulses
  - All the spectral ranges (SHG-THG crystal, spectrometers available,...)
  - From nJ to tens of J (detection threshold, optical damage)
  - Dynamic range independent of the optical detector
  - Resolve the temporal profile over the spatial profile
  - Resolves the space-time coupling
  - Resolve the polarization between the two axis with the phase relation
  - Measure the CEP
  - Single short measurement
  - On line measurement (no beam splitting)

- Most of the exiting techniques fulfill more than one criteria
- Not all the parameters are always relevant for the desired application
- Important to chose the method that matches at the best the experimental requirement

- Usual tools : Fourier transform, Temporal or Spectral convolution, Delay lines, Spectrometer, Imaging spectrometer, Spectral fringes, SHG, THG, Optical switch, known phase insertion, single short solution, Fresnel biprism, pulse shaper, use of spatial coordinate as temporal or spectral coordinate, polarization filtering,.....

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Thanks for your attention