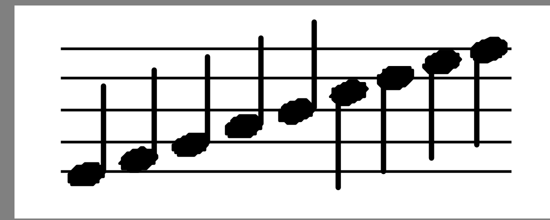
The Musical Score,



the Fundamental Theorem of Algebra,

$$z^n + a_{n-1} z^{n-1} + \ldots + a_0 \; = \; (z - z_n)(z - z_{n-1}) \ldots (z - z_1)$$

and the Measurement of the Shortest Events Ever Created

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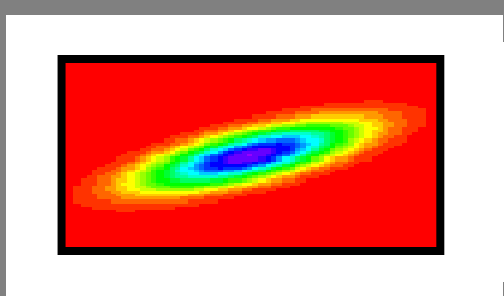


Funding: DOE, NSF, Georgia Tech

The Dilemma

In order to measure an event in time, you need a *shorter* one.

Photograph taken by Harold Edgerton, MIT

To study this event, you need a strobe light pulse that's shorter.

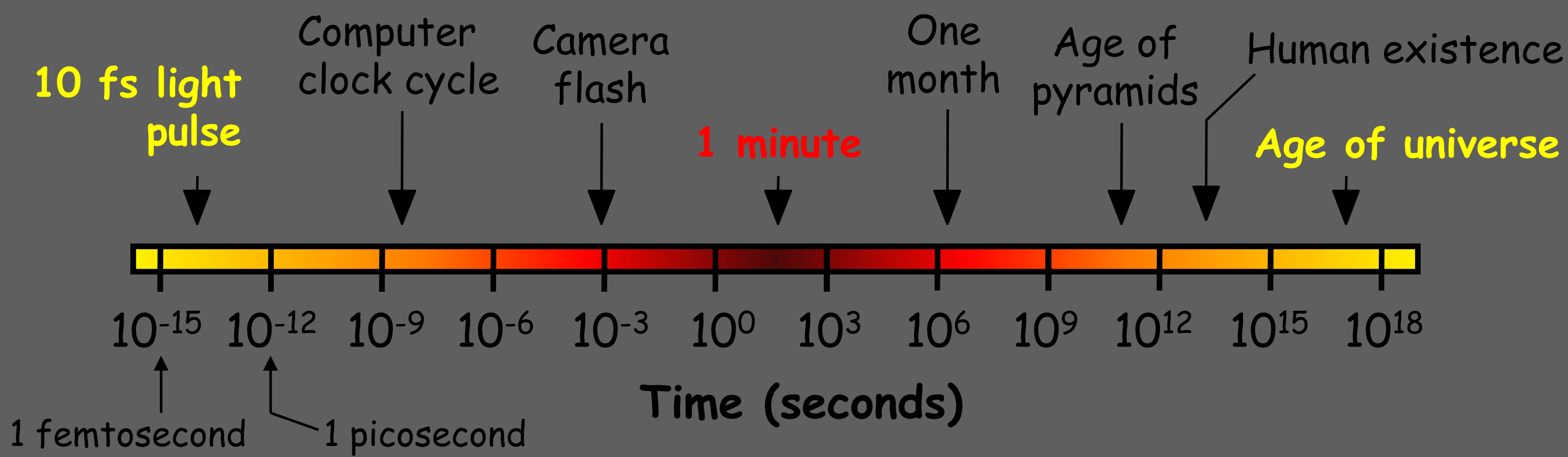
But then, to measure the strobe light pulse,
you need a detector whose response time is even shorter.

And so on…

So, now, how do you measure the *shortest* event?

Ultrashort laser pulses are the shortest technological events ever created by humans.

It's routine to generate pulses < 1 picosecond (10^{-12} s).
Researchers generate pulses a few femtoseconds (10^{-15} s) long.



Such a pulse is to one second as 5 cents is to the US national debt.

Such pulses have many applications in physics, chemistry, biology, and engineering. You can measure any event—if you have a pulse that's shorter.

So how do you measure the pulse itself?

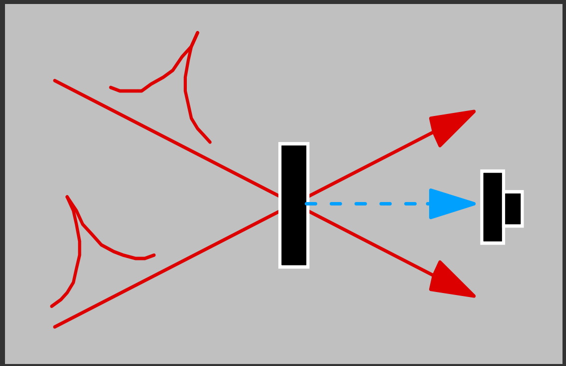
You must use the pulse to measure itself.

But that isn't good enough. It's only as short as the pulse.
It's not shorter.

Example: **Intensity Autocorrelation**

$$\int_{-\infty}^{\infty} I(t)I(t-\tau)\,dt$$

where $I(t)$ = pulse intensity



Techniques based on using the pulse to measure itself have not sufficed.

We must measure an ultrashort laser pulse's intensity and phase vs. time or frequency.

A laser pulse has the time-domain electric field:

$$E(t) = \mathrm{Re}\left\{\sqrt{I(t)}\exp\left[i(\omega_0 t - \phi(t))\right]\right\}$$

Intensity — $I(t)$; Phase — $\phi(t)$

Equivalently, vs. frequency:

$$\tilde{E}(\omega) = \sqrt{S(\omega)}\exp\left[-i\varphi(\omega)\right]$$

(neglecting the negative-frequency component)

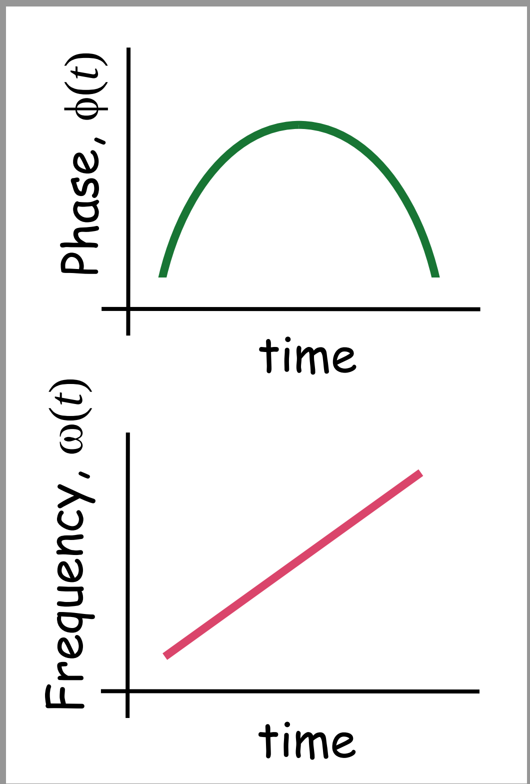
Spectrum — $S(\omega)$; Spectral Phase — $\varphi(\omega)$

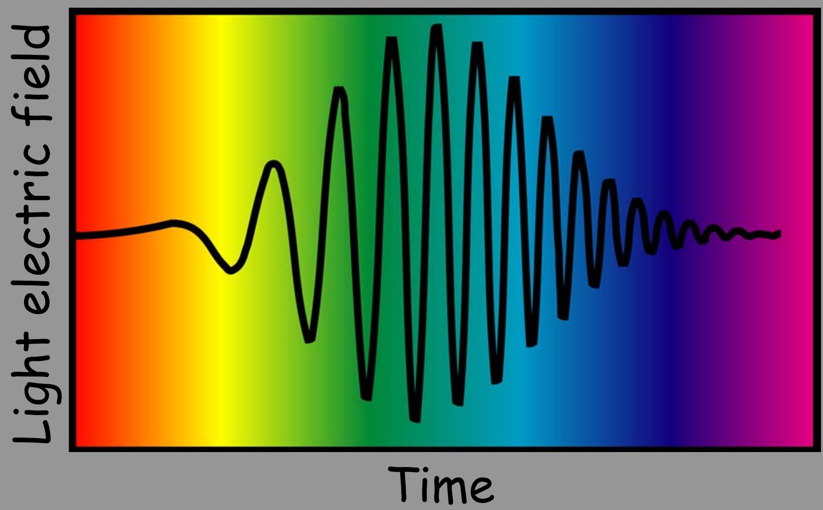
Knowledge of the intensity and phase or the spectrum and spectral phase is sufficient to determine the pulse.

The phase determines the pulse's frequency (i.e., color) vs. time.

The instantaneous frequency: $\omega(t) = \omega_0 - d\phi / dt$

Example: "Linear chirp"

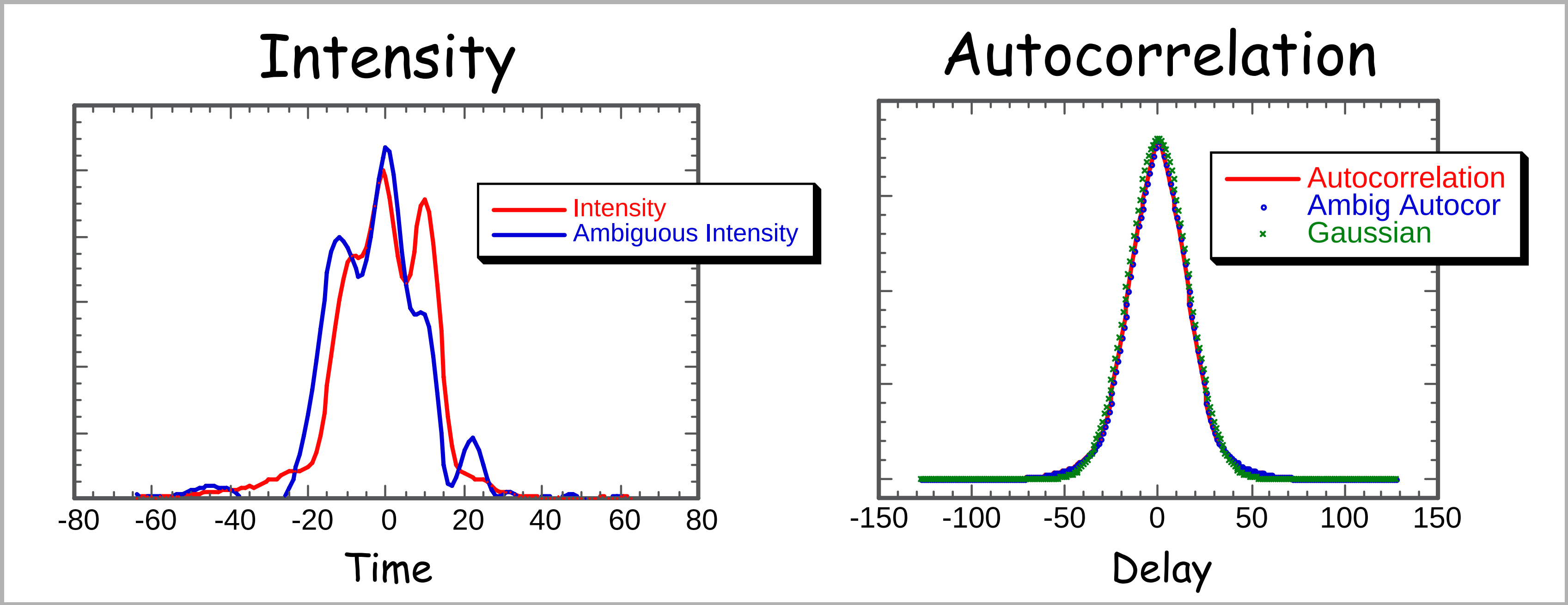




We'd like to be able to measure,
not only linearly chirped pulses,
but also pulses with arbitrarily complex phases and frequencies vs. time.

Autocorrelations have ambiguities.

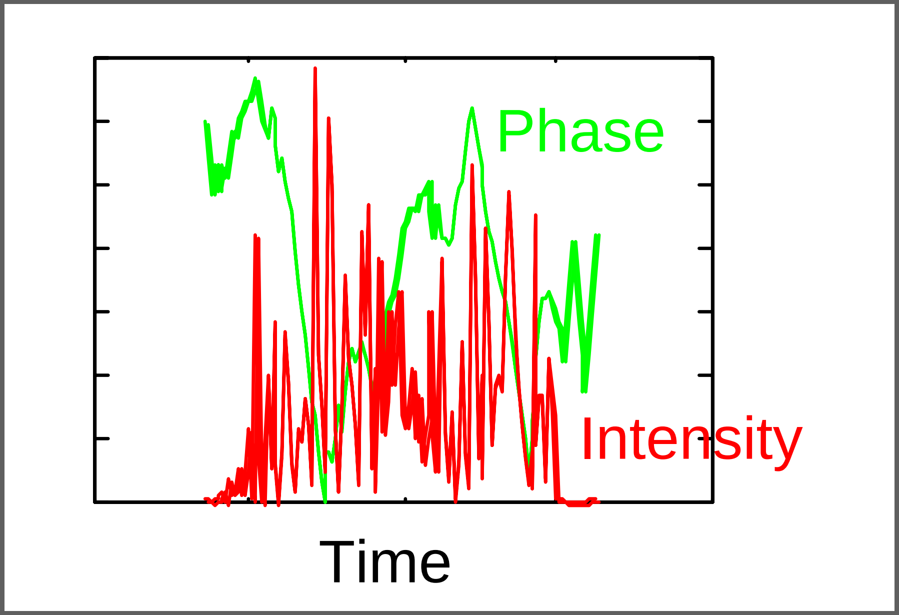
These intensities have the same, nearly Gaussian, autocorrelations.



Retrieving the intensity from the autocorrelation is equivalent to the 1D Phase-Retrieval Problem, a well-known unsolvable problem.

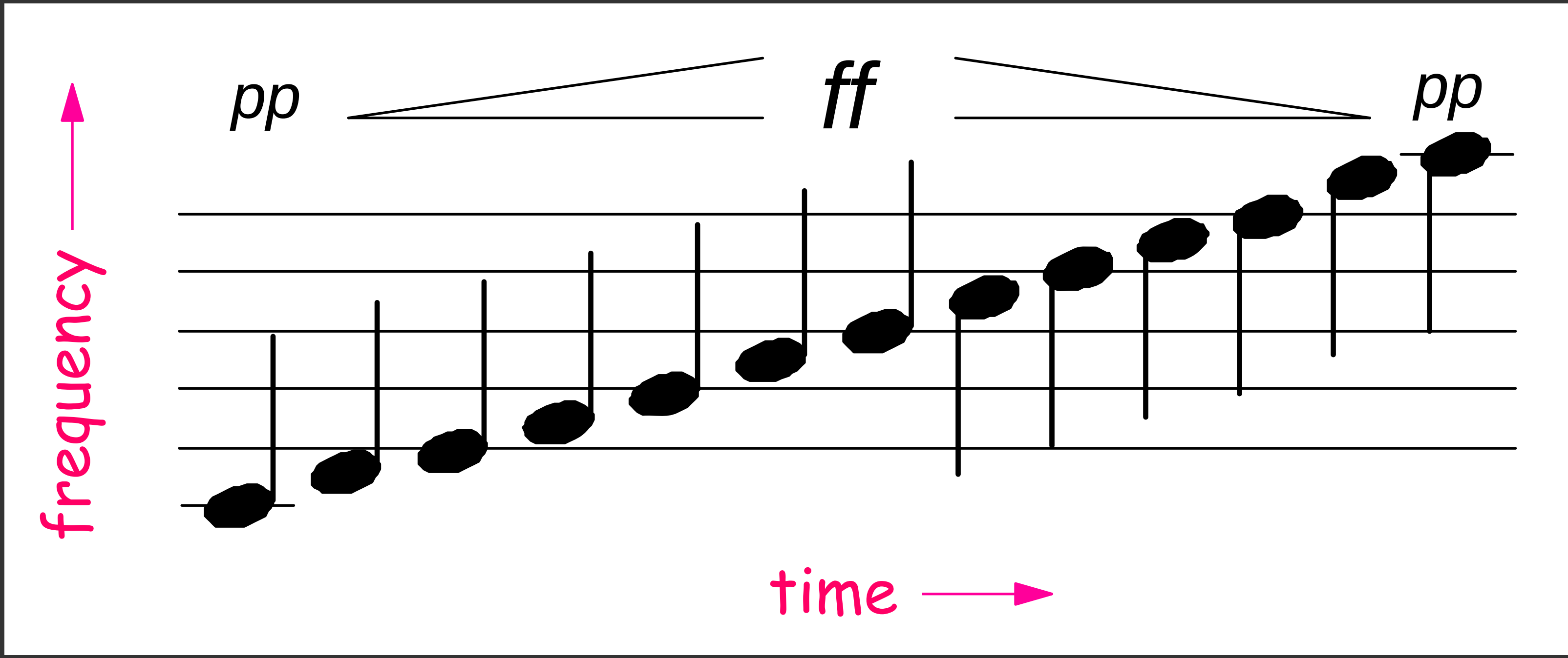
Autocorrelation and related techniques yield little information about the pulse.

Perhaps it's time to ask how researchers in other fields deal with their waveforms…



Consider, for example, acoustic waveforms.

Most people think of acoustic waves in terms of a musical score.



It's a plot of frequency vs. time, with info on top about intensity.

The musical score lives in the "time-frequency domain."

A mathematically rigorous form of the musical score is the "spectrogram."

If $E(t)$ is the waveform of interest, its spectrogram is:

$$\Sigma_E(\omega,\tau) \equiv \left| \int_{-\infty}^{\infty} E(t)\, g(t-\tau)\, \exp(-i\omega t)\, dt \right|^2$$

where $g(t-\tau)$ is a variable-delay gate function and τ is the delay.

Without $g(t-\tau)$, $\Sigma_E(\omega,\tau)$ would simply be the spectrum.

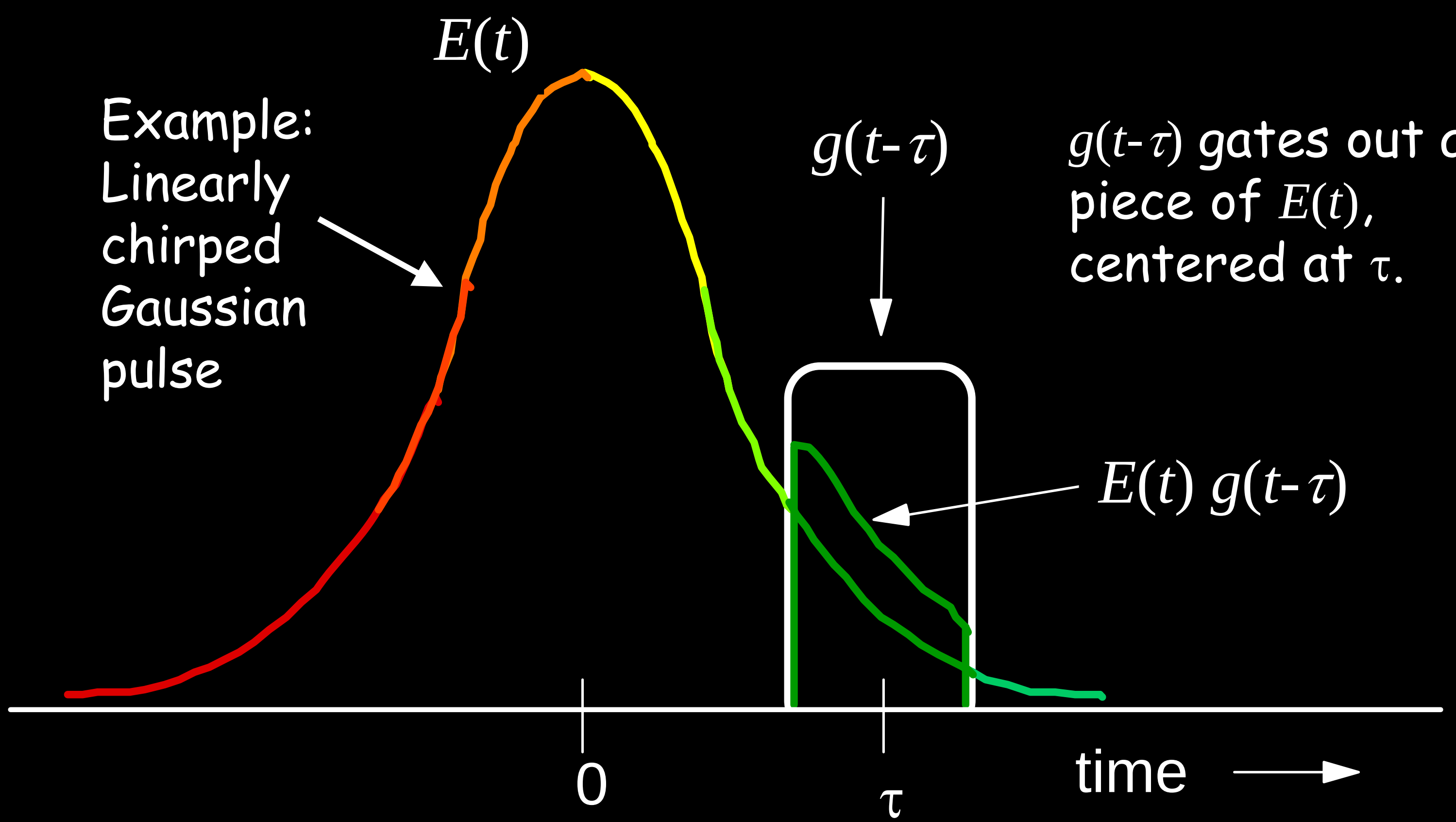
The spectrogram is a function of ω and τ.

It is the set of spectra of all temporal slices of $E(t)$.

The spectrogram is one of many time-frequency quantities, such as the Wigner Distribution, Wavelet Transform, and others.

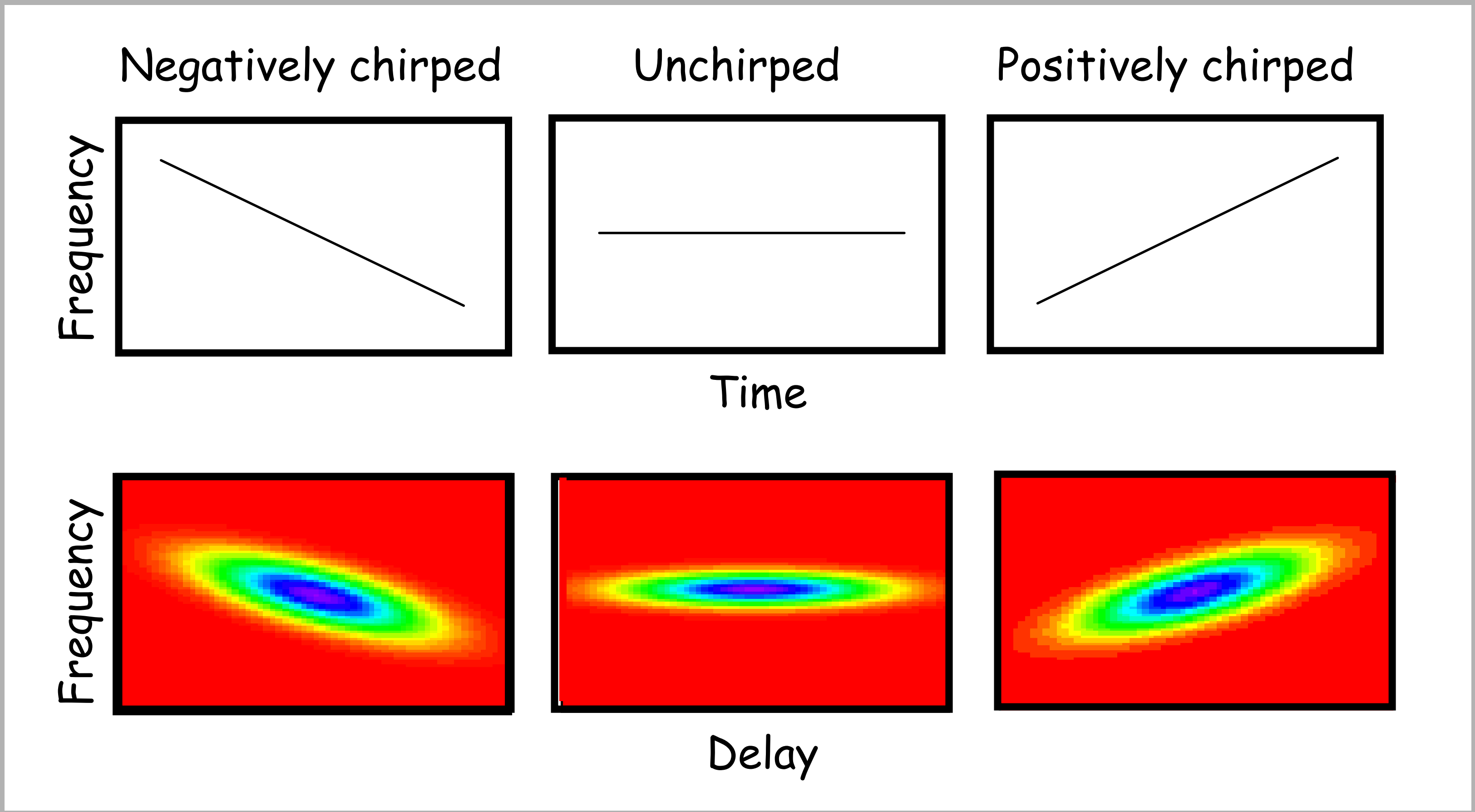
The Spectrogram of a waveform $E(t)$

We must compute the spectrum of the product: $E(t)\, g(t-\tau)$



The spectrogram tells the color and intensity of $E(t)$ at the time, τ.

Spectrograms for Linearly Chirped Pulses



Like a musical score, the spectrogram visually displays the frequency vs. time (and the intensity, too).

Properties of the Spectrogram

Algorithms exist to retrieve $E(t)$ from its spectrogram.

The spectrogram essentially uniquely determines the waveform intensity, $I(t)$, and phase, $\phi(t)$.

There are a few ambiguities, but they're "trivial."

The gate need not be—and should not be—much shorter than $E(t)$.

Suppose we use a delta-function gate pulse:

$$\left| \int_{-\infty}^{\infty} E(t)\, \delta(t-\tau) \exp(-i\omega t)\, dt \right|^2 = \left| E(\tau) \exp(-i\omega\tau) \right|^2$$

$$= \left| E(\tau) \right|^2$$ = **The Intensity.**

No phase information!

The spectrogram resolves the dilemma! It doesn't need the shorter event! It temporally resolves the slow components and spectrally resolves the fast components.

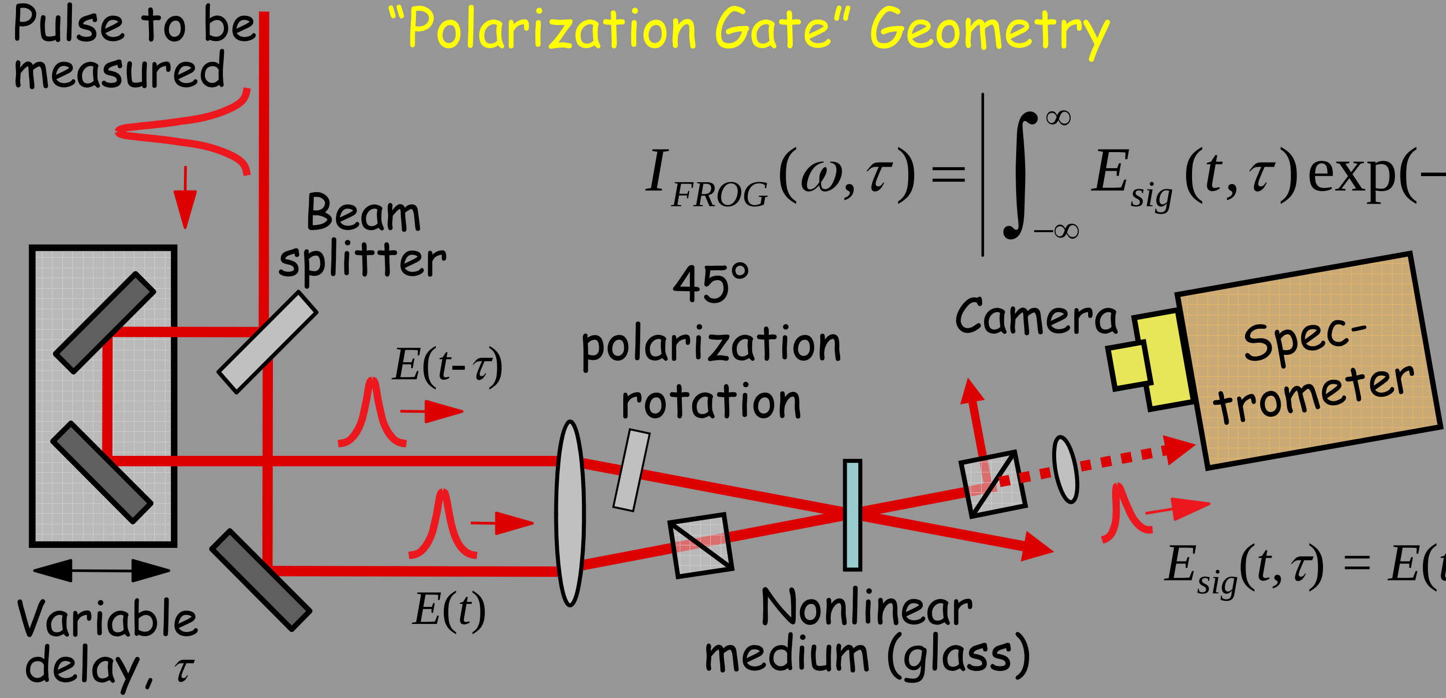
Frequency-Resolved Optical Gating (FROG)

FROG involves gating the pulse with a variably delayed replica of itself in an instantaneous nonlinear-optical medium and then spectrally resolving the gated pulse vs. delay.

Collaborator:
Dan Kane

"Polarization Gate" Geometry

$$I_{FROG}(\omega,\tau) = \left| \int_{-\infty}^{\infty} E_{sig}(t,\tau) \exp(-i\omega t)\, dt \right|^2$$



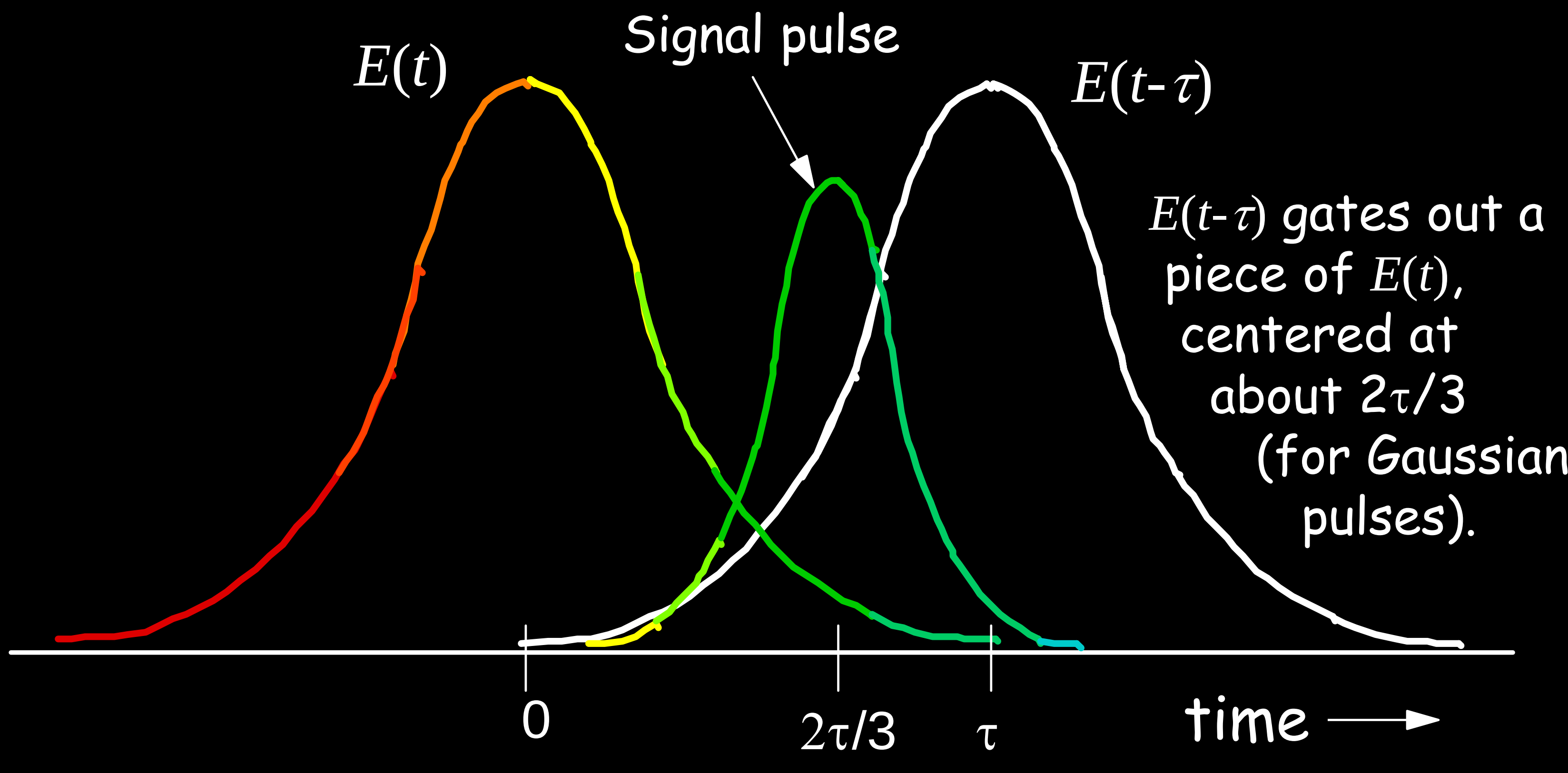
$E_{sig}(t,\tau) = E(t)\,|E(t-\tau)|^2$

Use any ultrafast nonlinearity: Second-harmonic generation, etc.

R. Trebino, *Frequency-Resolved Optical Gating: The Measurement of Ultrashort Laser Pulses*, Kluwer

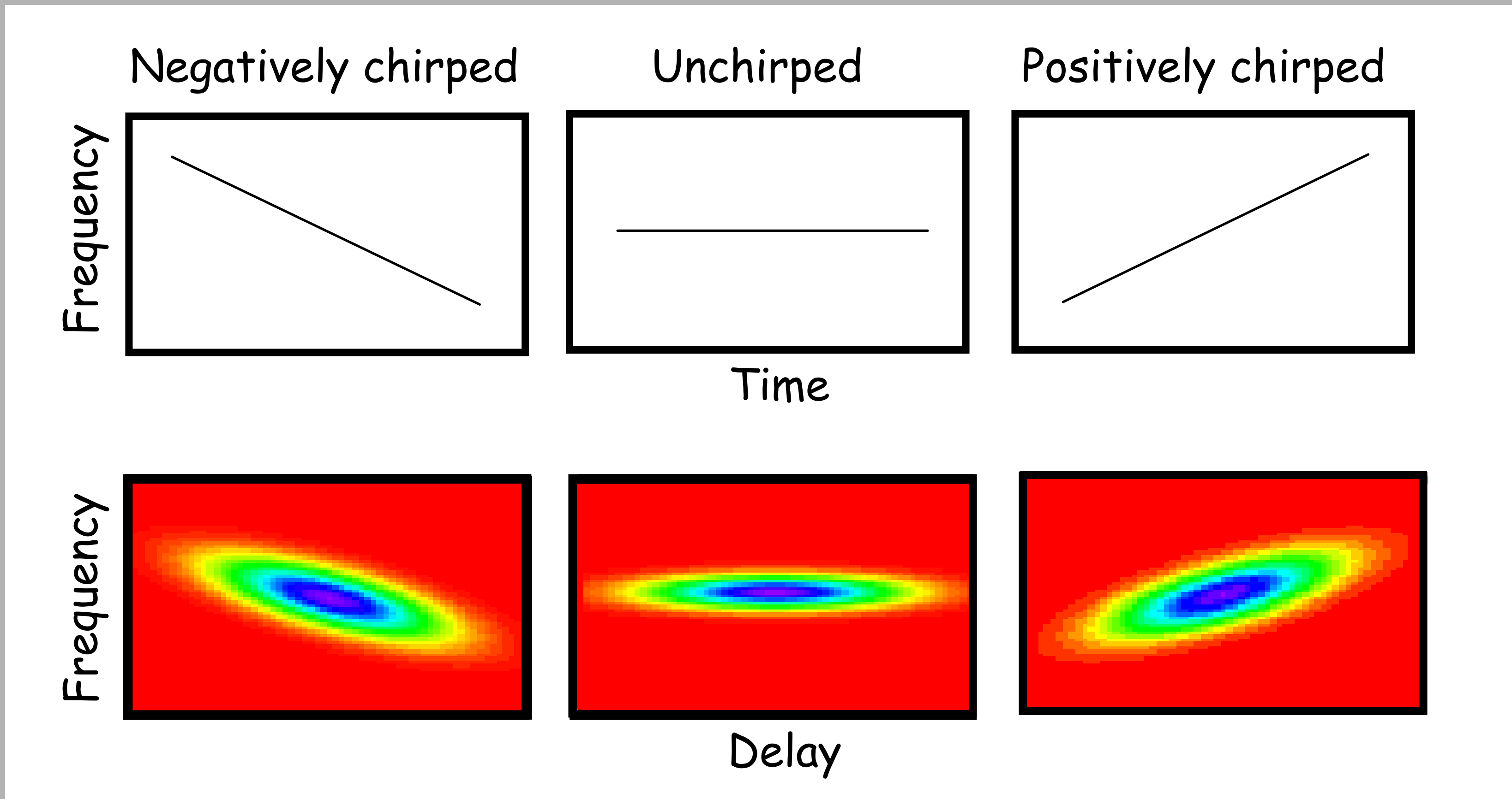
FROG

$$E_{sig}(t,\tau) \propto E(t)\left|E(t-\tau)\right|^2$$



The gating is more complex for complex pulses, but it still works.
And it also works for other nonlinear-optical processes.

FROG Traces for Linearly Chirped Pulses



Like a musical score, the FROG trace visually reveals the pulse frequency vs. time—for simple and complex pulses.

The FROG trace is a spectrogram of $E(t)$.

Substituting for $E_{sig}(t,\tau)$ in the expression for the FROG trace:

$E_{sig}(t,\tau) \propto E(t)\,|E(t-\tau)|^2$

$$I_{FROG}(\omega,\tau) \propto \left| \int E_{sig}(t,\tau)\exp(-i\omega t)\,dt \right|^2$$

yields:

$$I_{FROG}(\omega,\tau) \propto \left| \int E(t)\,g(t-\tau)\exp(-i\omega t)\,dt \right|^2$$

where:

$g(t-\tau) = |E(t-\tau)|^2$

Unfortunately, spectrogram inversion algorithms require that we know the gate function, and that's what we're trying to find!

Consider FROG as a two-dimensional phase-retrieval problem.

If $E_{sig}(t,\tau)$, is the 1D Fourier transform with respect to Ω of some new signal field, $\hat{E}_{sig}(t,\Omega)$, then:

The input pulse, $E(t)$, is easily obtained from $\hat{E}_{sig}(t,\Omega)$: $E(t) \propto \hat{E}_{sig}(t,0)$

and

$$I_{FROG}(\omega,\tau) = \left| \int E_{sig}(t,\tau) \exp(-i\omega t)\, dt \right|^2$$

So we must invert this integral equation and solve for $\hat{E}_{sig}(t,\Omega)$.

This integral-inversion problem is the 2D phase-retrieval problem, for which the solution exists and is (essentially) unique.
And simple algorithms exist for finding it.

1D vs. 2D Phase Retrieval

1D Phase Retrieval: Suppose we measure $S(\omega)$ and desire $E(t)$, where:

$$S(\omega) = \left| \int_{-\infty}^{\infty} E(t) \exp(-i\omega t)\, dt \right|^2$$

Given $S(\omega)$, there are **infinitely many solutions** for $E(t)$. *We lack the spectral phase.*

We assume that $E(t)$ and $E(x,y)$ are of finite extent.

2D Phase Retrieval: Suppose we measure $S(k_x,k_y)$ and desire $E(x,y)$:

$$S(k_x,k_y) = \left| \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} E(x,y) \exp(-ik_x x - ik_y y)\, dx\, dy \right|^2$$

Given $S(k_x,k_y)$, there is **essentially one solution** for $E(x,y)$!!!
It turns out that it's possible to retrieve the 2D spectral phase!

Stark, Image Recovery, Academic Press, 1987.

These results are related to the Fundamental Theorem of Algebra.

Phase Retrieval and the Fundamental Theorem of Algebra

The Fundamental Theorem of Algebra states that all polynomials can be factored:

$$f_{N-1}\, z^{N-1} + f_{N-2}\, z^{N-2} + \ldots + f_1\, z + f_0 = f_{N-1}\,(z - z_1)\,(z - z_2) \ldots (z - z_{N-1})$$

The Fundamental Theorem of Algebra fails for polynomials of two variables. Only a set of measure zero can be factored.

$$f_{N-1,M-1}\, y^{N-1}\, z^{M-1} + f_{N-1,M-2}\, y^{N-1}\, z^{M-2} + \ldots + f_{0,0} = ?$$

Why does this matter?

The existence of the 1D Fundamental Theorem of Algebra implies that 1D phase retrieval is impossible.

The **non-existence** of the 2D Fundamental Theorem of Algebra implies that 2D phase retrieval is **possible**.

1D Phase Retrieval & the Fundamental Theorem of Algebra

The Fourier transform $\{F_0, \ldots, F_{N-1}\}$ of a discrete 1D data set, $\{f_0, \ldots, f_{N-1}\}$, is:

$$F_k \equiv \sum_{m=0}^{N-1} f_m e^{-imk} = \sum_{m=0}^{N-1} f_m z^m \qquad \text{where } z = e^{-ik}$$

polynomial!

The Fundamental Theorem of Algebra states that any polynomial, $f_{N-1} z^{N-1} + \ldots + f_0$, can be factored to yield: $f_{N-1}(z-z_1)(z-z_2)\ldots(z-z_{N-1})$

So the magnitude of the Fourier transform of our data can be written:

$$|F_k| = |f_{N-1}(z-z_1)(z-z_2)\ldots(z-z_{N-1})| \qquad \text{where } z = e^{-ik}$$

Complex conjugation of any factor(s) leaves the magnitude unchanged, but changes the phase, yielding an ambiguity! So 1D phase retrieval is impossible!

2D Phase Retrieval and the Fundamental Theorem of Algebra

The Fourier transform $\{F_{0,0}, \ldots, F_{N\text{-}1,N\text{-}1}\}$ of a discrete 2D data set, $\{ f_{0.0}, \ldots, f_{N\text{-}1,N\text{-}1}\}$, is:

$$F_{k,q} \equiv \sum_{m=0}^{N-1} \sum_{p=0}^{N-1} f_{m,p}\, e^{-imk} e^{-ipq} = \sum_{m=0}^{N-1} \sum_{p=0}^{N-1} f_{m,p}\, y^m z^p$$

Polynomial of 2 variables!

where $y = e^{-ik}$
and $z = e^{-iq}$

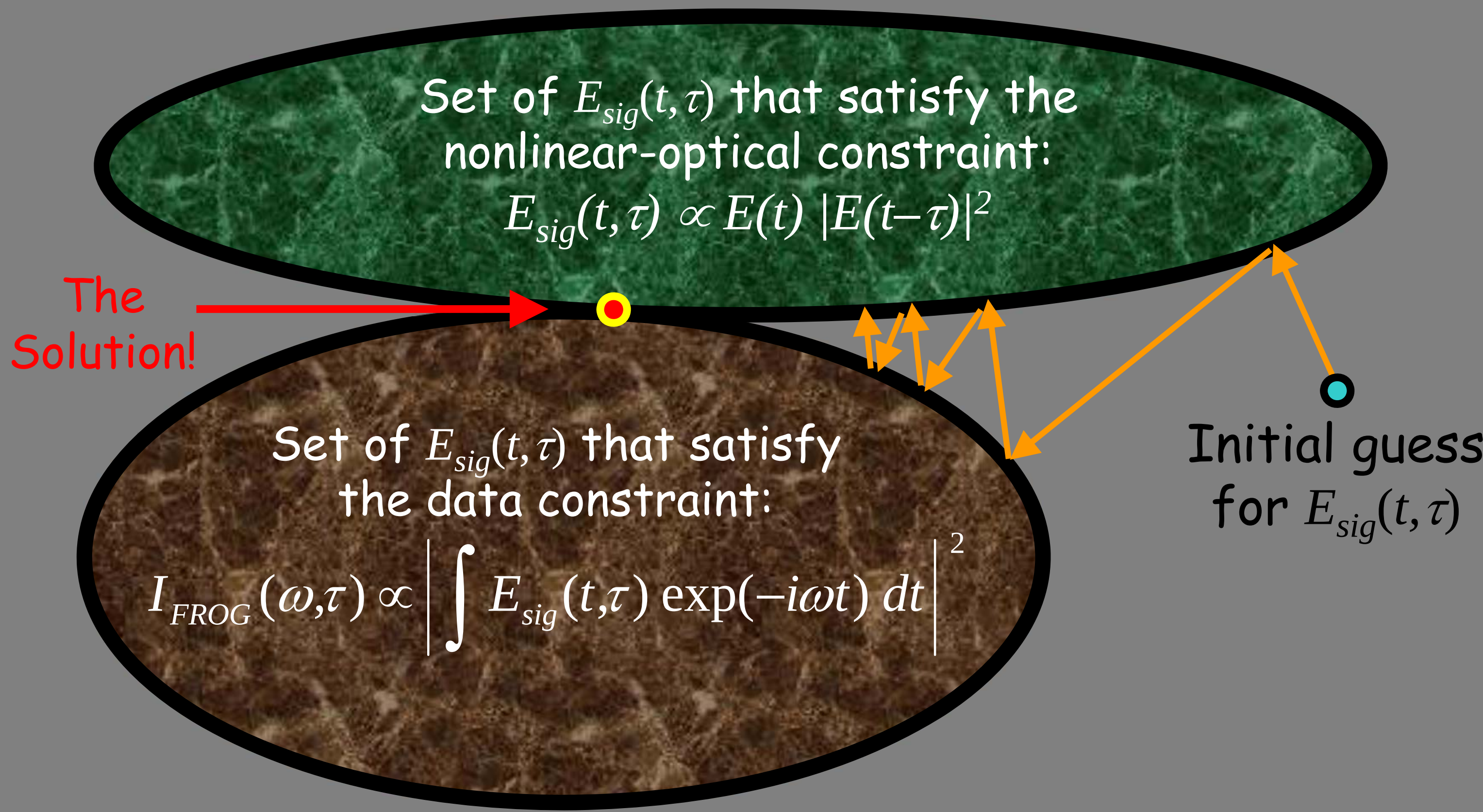
But we cannot factor polynomials of two variables. So we can only complex-conjugate the entire expression (yielding a trivial ambiguity).

Only a set of polynomials of measure zero can be factored.
So 2D phase retrieval is possible! And the ambiguities are very sparse.

Generalized Projections

Collaborator: Ken DeLong,
Femtosoft Technologies

A projection maps the current guess for the waveform to the closest point in the constraint set.

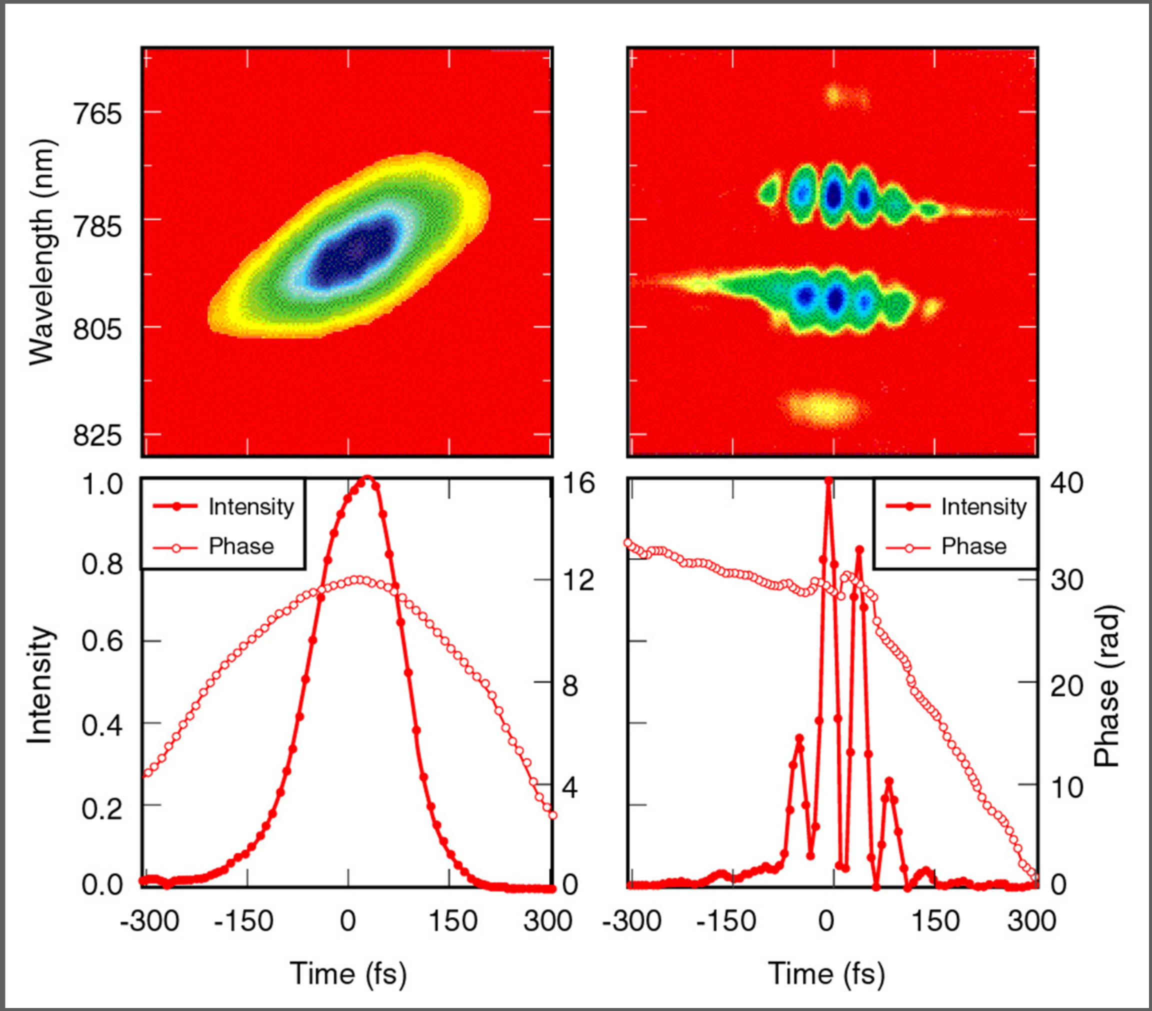


Convergence is guaranteed for convex sets, but generally occurs even with non-convex sets and in particular in FROG.

Ultrashort pulses measured using FROG

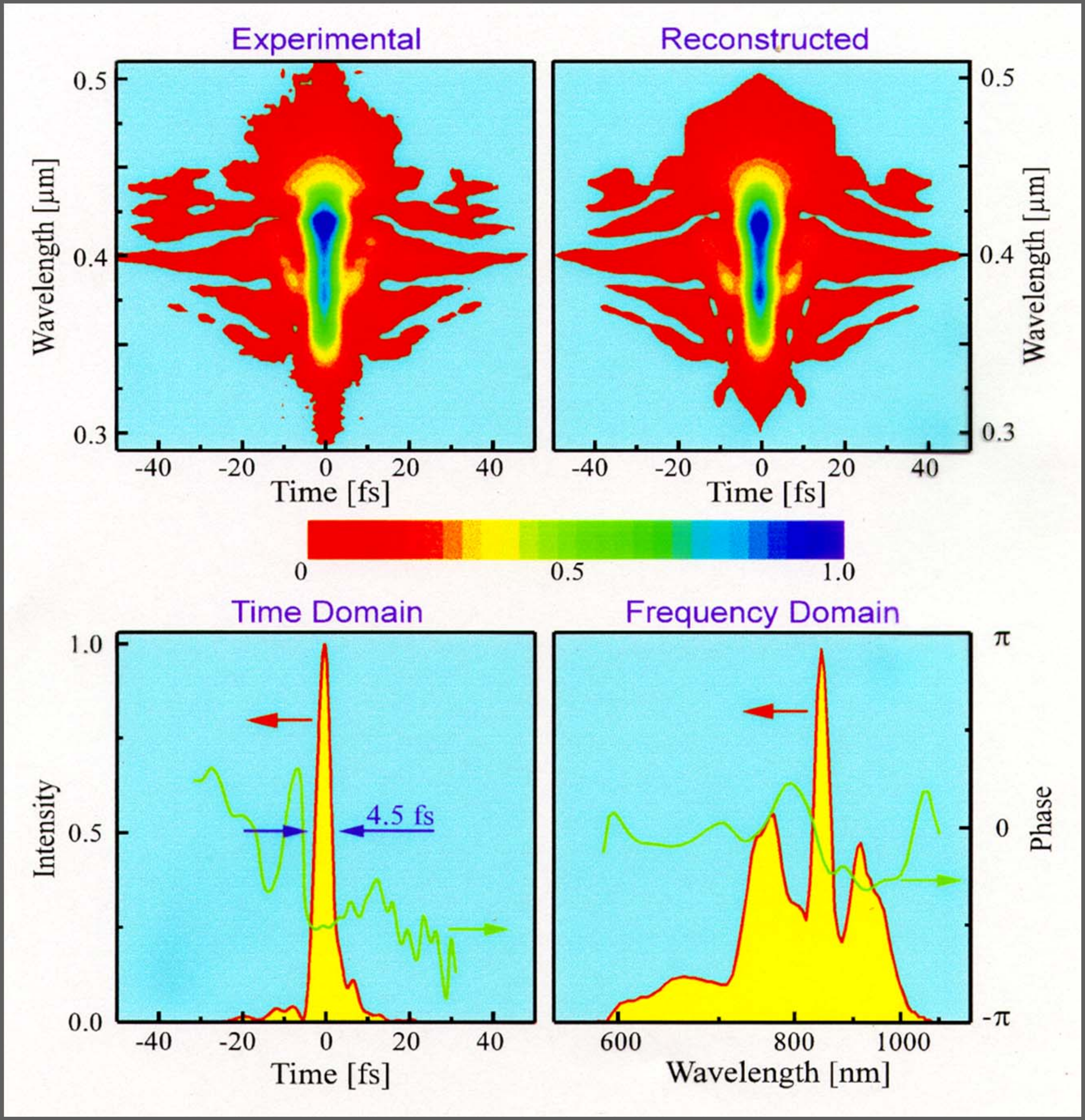
FROG Traces

Retrieved pulses



Data courtesy of Profs. Bern Kohler and Kent Wilson, UCSD.

FROG Measurements of a 4.5-fs Pulse!

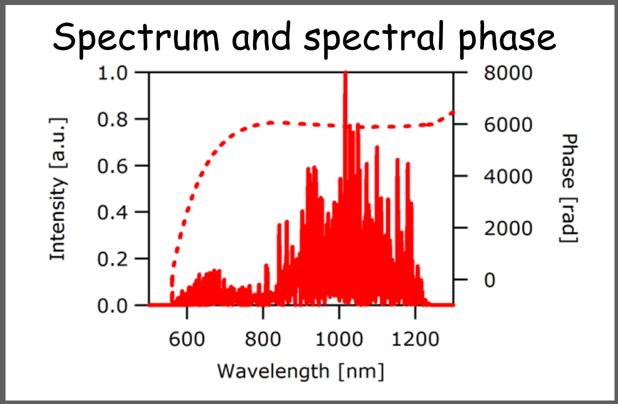


Baltuska, Pshenichnikov, and Weirsma, J. Quant. Electron., 35, 459 (1999).

Frontiers in ultrashort-pulse measurement

Measurement of very complex pulses (continuum)

Measurement of noisy trains of pulses (continuum)



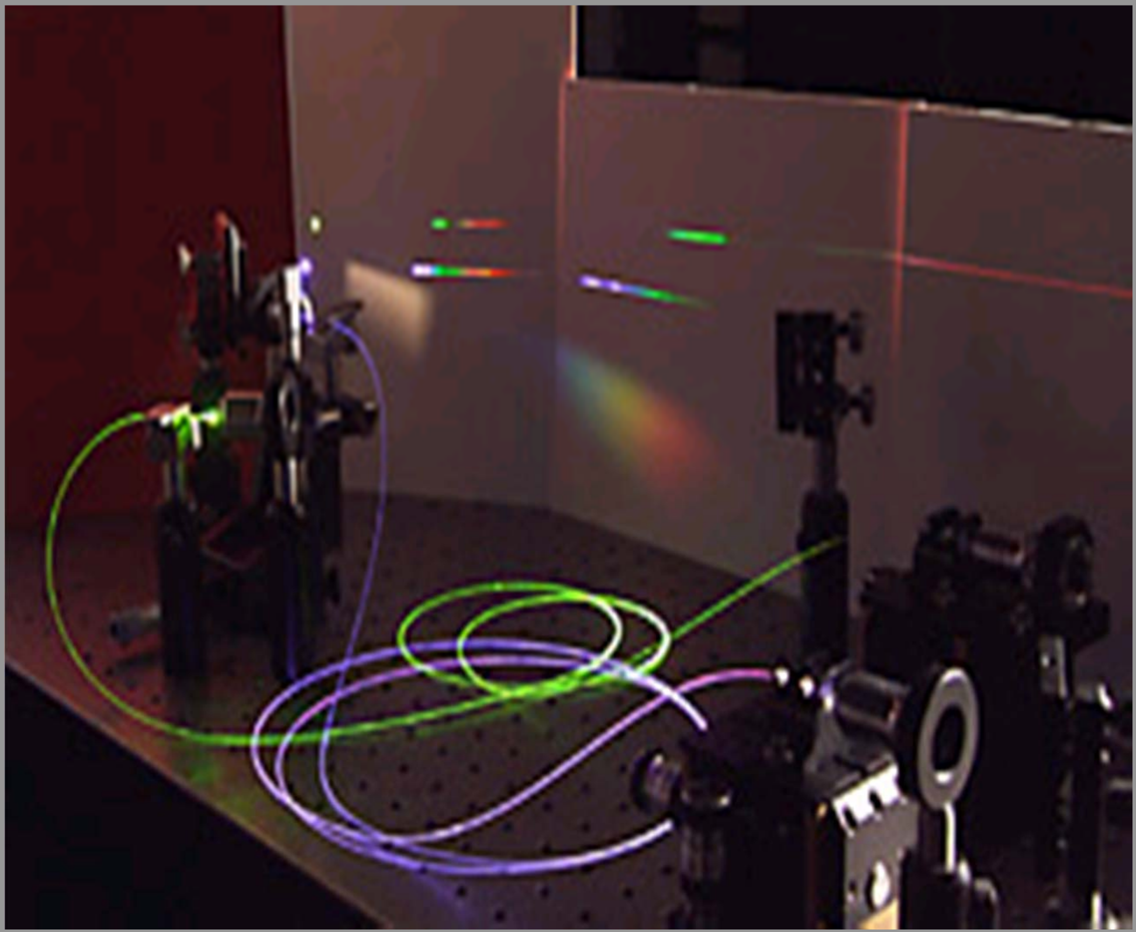
Measurement of ultraweak, spatially incoherent pulses with random absolute phase (sub-ps fluorescence)

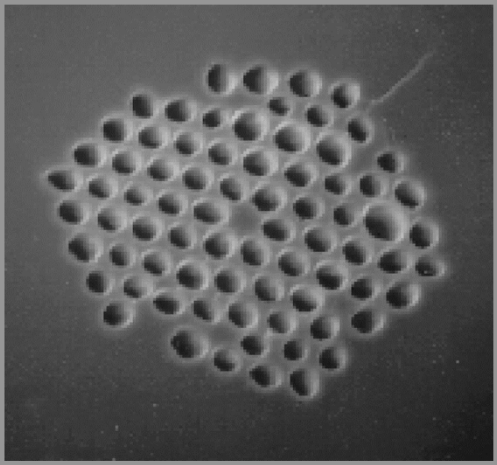
Development of a practical alignment-free pulse-measurement device (GRENOUILLE)

Measurement of spatio-temporal pulse distortions* (e.g., spatial chirp and pulse-front tilt)

*This device should not itself introduce these distortions!

Microstructure fiber yields ultrabroadband continuum.



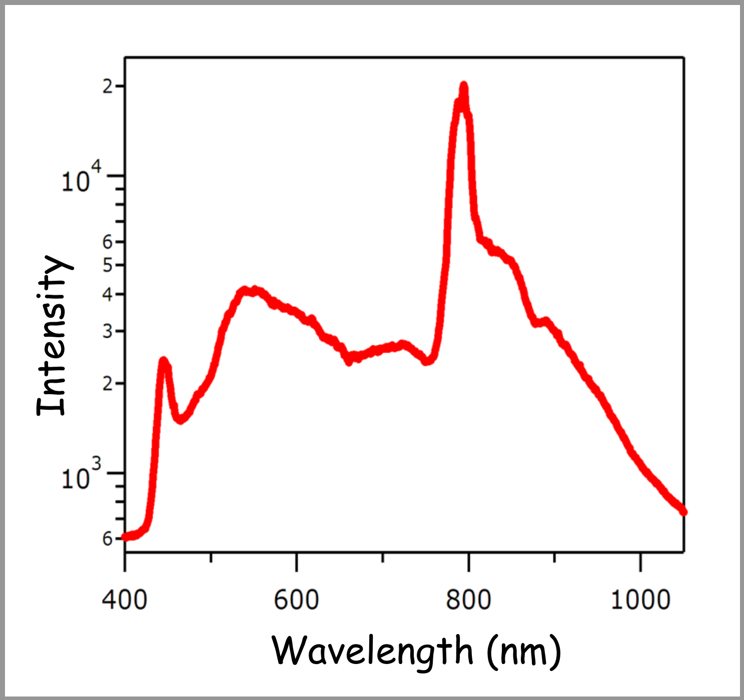


The continuum has many applications, from medical imaging to metrology.

It's a important to measure it.

Photographs courtesy of Jinendra Ranka, Lucent

Measurements of the microstructure-fiber continuum have yielded a **broad**, **smooth**, and **stable** spectrum.

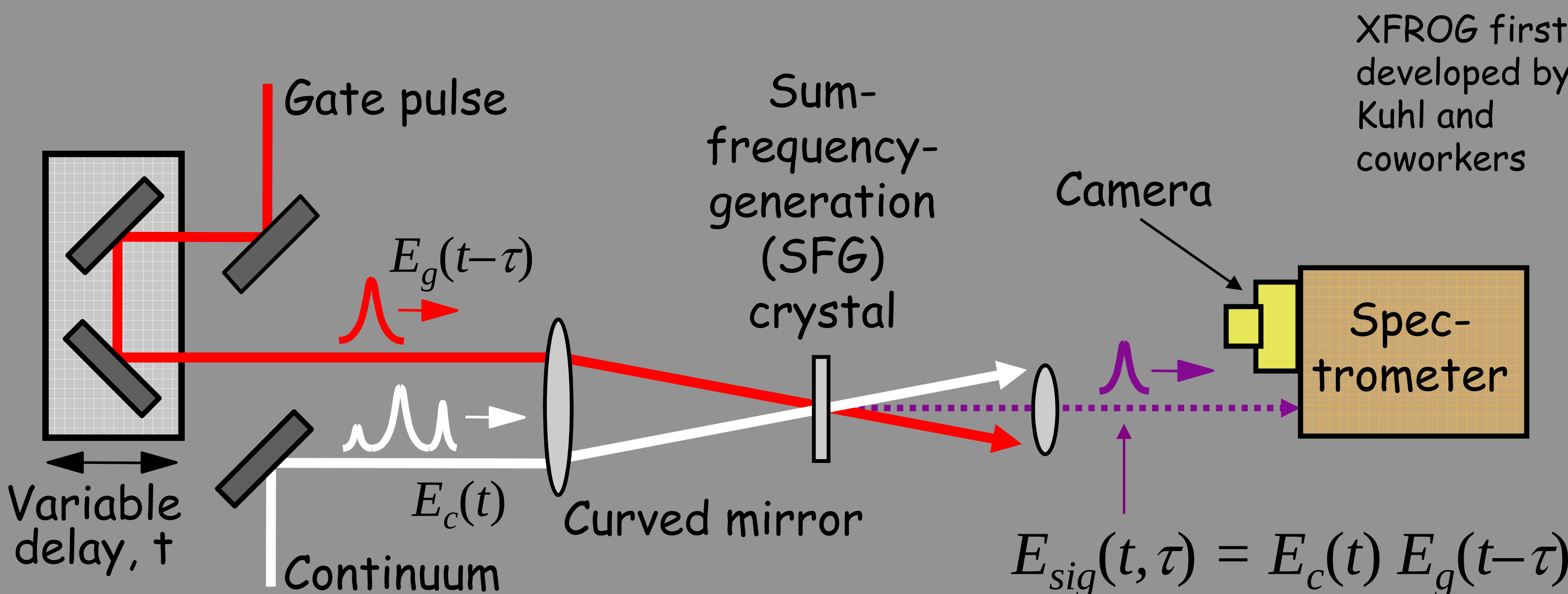


A typical microstructure-fiber continuum spectrum generated in our lab by a train of 30-fs Ti:Sapphire oscillator pulses.

Unfortunately, only **one** of these adjectives is in fact true!

XFROG: Gating a pulse with another pulse

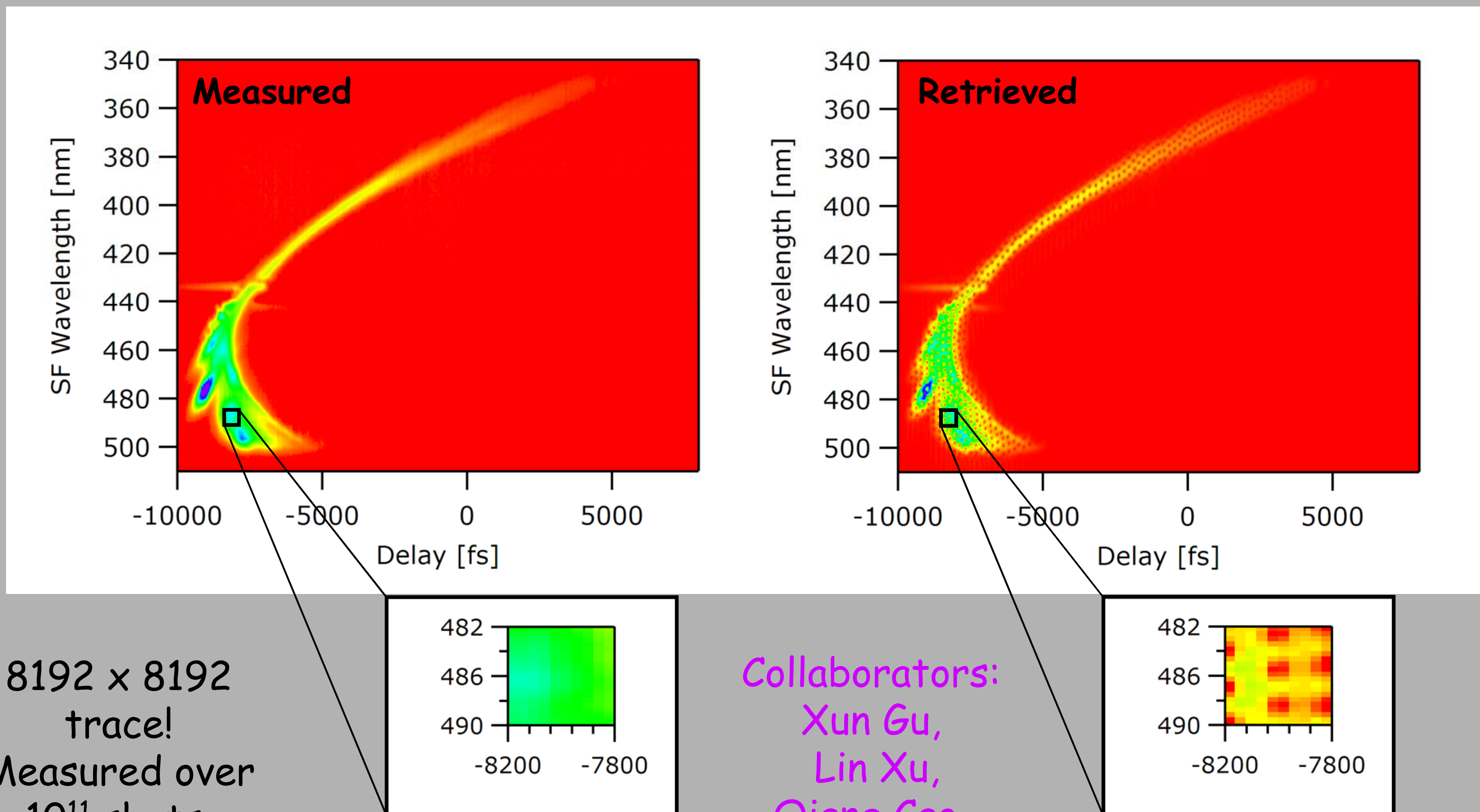
It's better to gate a complicated pulse with a simple (known) one.



$$I_{XFROG}(\omega,\tau) = \left| \int_{-\infty}^{\infty} E_c(t)\, E_g(t-\tau) \exp(-i\omega t)\, dt \right|^2$$

The XFROG trace is the usual spectrogram.

XFROG measurement of the continuum

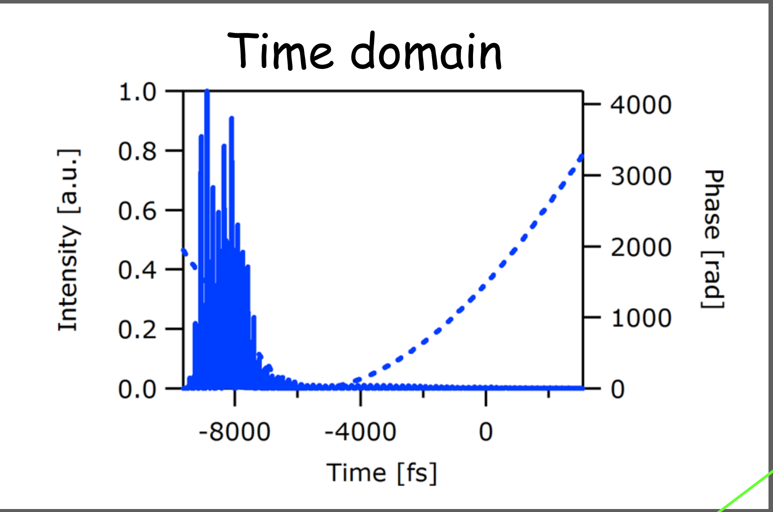


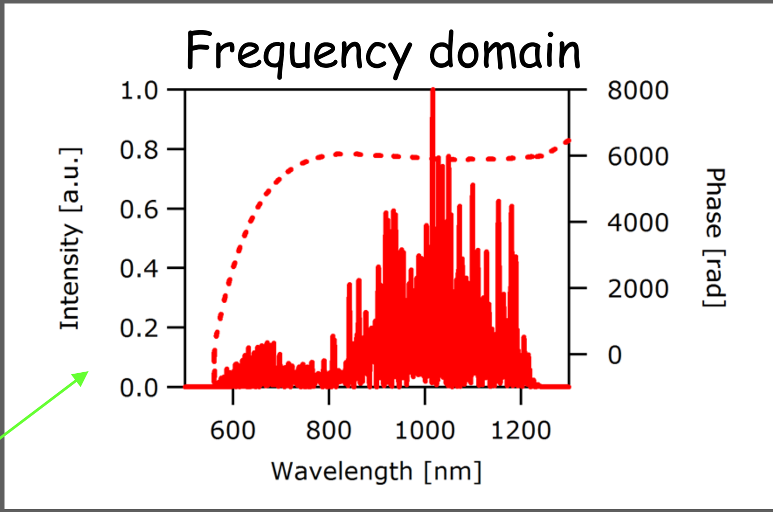
8192 x 8192 trace! Measured over 10^{11} shots.

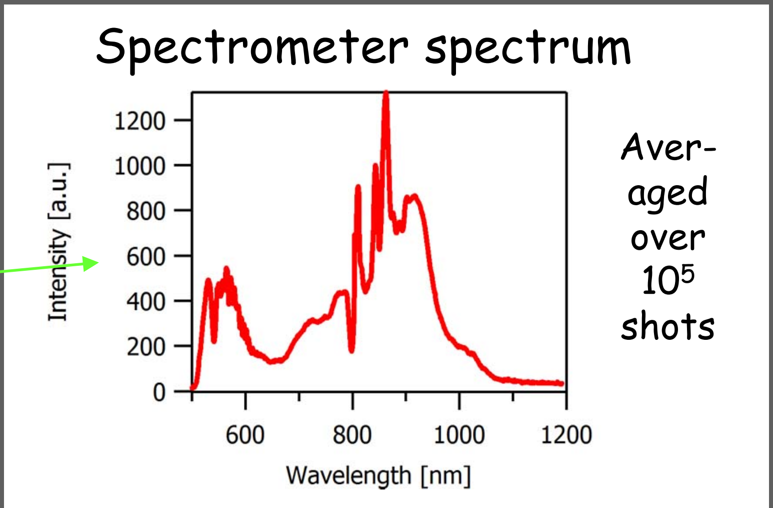
Collaborators:
Xun Gu,
Lin Xu,
Qiang Cao,
Erik Zeek

While the large-scale structure of each trace is identical, the measured trace lacks the fine-scale structure of the retrieved trace.

XFROG-measured intensity and phase of the microstructure-fiber continuum





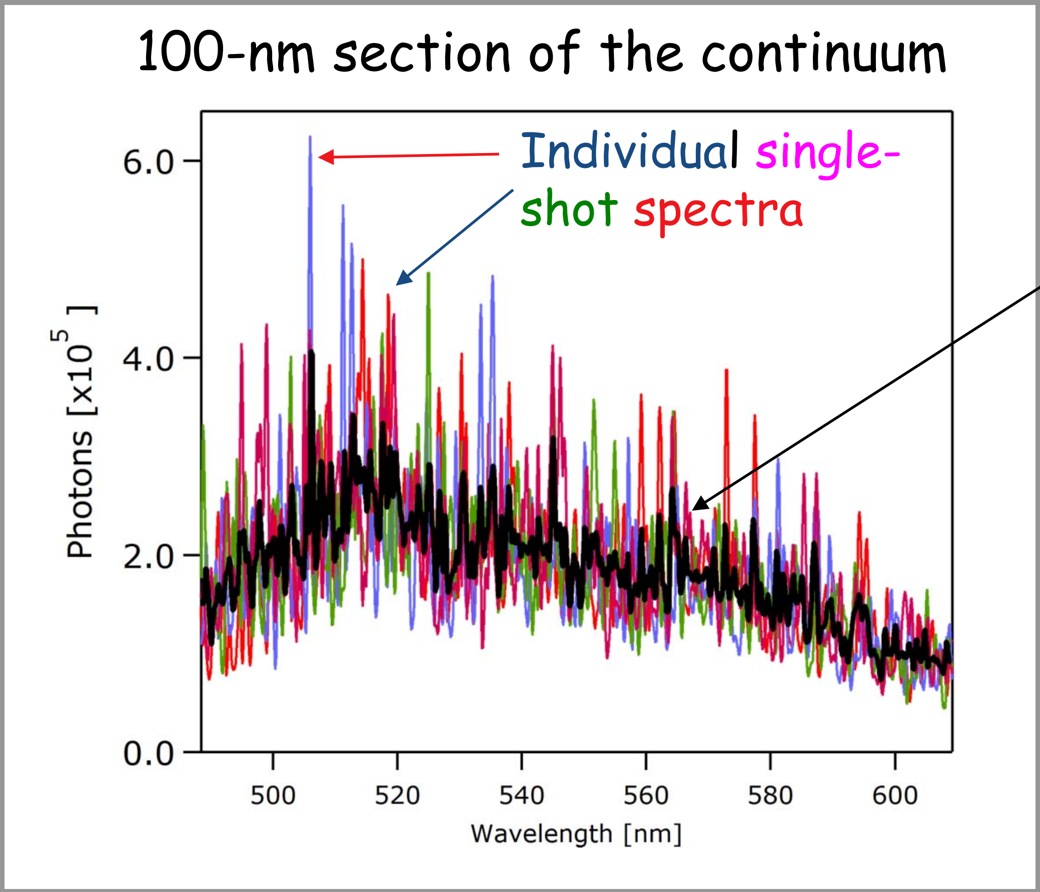


The XFROG-measured spectrum contains much more structure than the spectrum-measured with a spectrometer.

Which spectral measurement is correct?

Single-shot spectra reveal fine structure!

Sending a single continuum pulse into a spectrometer, yields its true spectrum.



Manual average of *four* consecutive single-shot spectrum measurements

Despite averaging over 10^{11} shots, FROG still sees the structure!

Why does FROG see the spectral structure when even the few-shot spectrum doesn't?

FROG sees the missing structure because it operates in the time-frequency domain.

Frequency structure is tagged by its time and so is less likely to wash out.

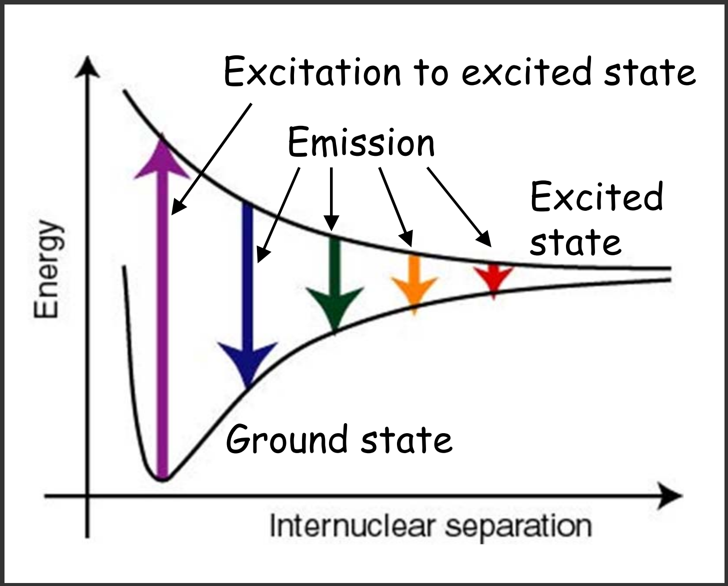
Even when it does, the trace area yields the time-bandwidth product, so it still indicates a complex pulse.

Measurement of Ultraweak Fluorescence

Not all ultrashort pulses are generated by lasers.

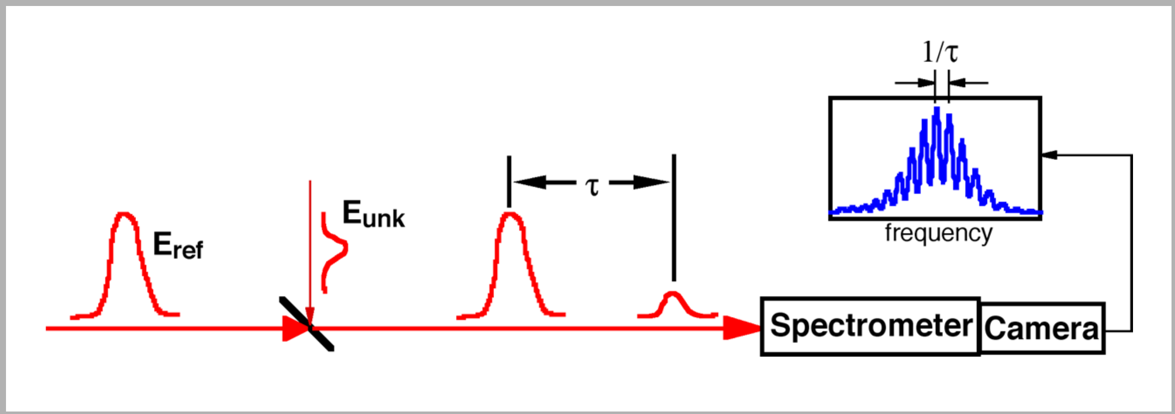
Biologically important fluorescence is necessarily weak and ultrafast.

Knowledge of the fluorescence intensity and phase vs. time would yield important information about molecular dynamics in the fluorescing molecule.



Existing techniques cannot measure the phase evolution of weak fluorescence.

Spectral Interferometry cannot measure weak fluorescence.



Previously, we showed that SI could measure a train of pulses with less than one photon each.

$$S_{SI}(\omega) = S_{ref}(\omega) + S_{unk}(\omega) + 2\sqrt{S_{ref}(\omega)}\,\sqrt{S_{unk}(\omega)}\cos[\phi_{unk}(\omega) - \phi_{ref}(\omega) - \omega\tau]$$

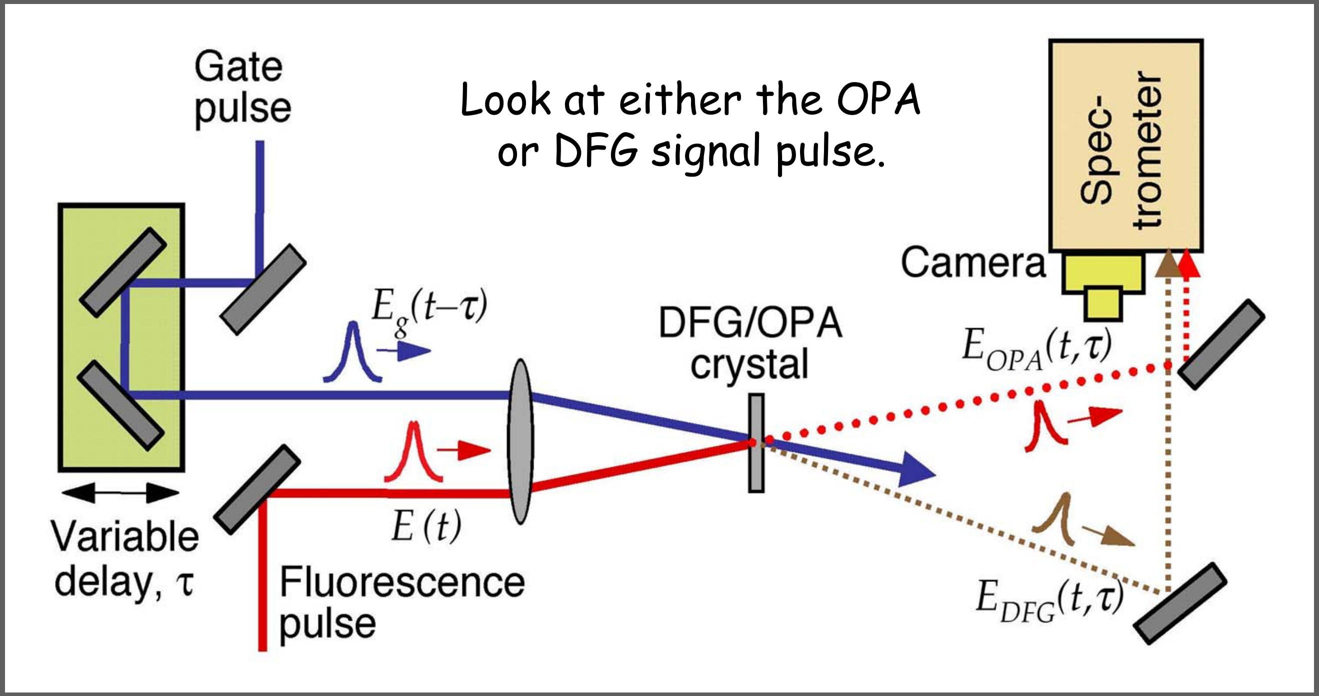
The **absolute phase** (the constant-phase term) in fluorescence is **random**, washing out the SI fringes and preventing multi-shot measurements.

Spatial incoherence severely limits the number of spatially coherent photons generated by an incoherent (i.e., fluorescent) source.

Gating with gain in XFROG

Optical parametric amplification (OPA) and Difference-Frequency Generation (DFG) have **exponential gain** (up to ~10^6). This doesn't distort the phase, and huge bandwidths are also possible. Perfect!

Collaborators: Stephan Link, Aparna Shreenath, Jing Zhang, and Xuan Liu



Absolute phase and spatial coherence don't affect FROG measurements. We must, however, modify the FROG algorithm for these processes: the gate function is now: $\exp[g|E_{gate}(t)|]$. But this is easy.

But what pulse will we use for the XFROG gate pulse?

Requirements for the OPA or DFG XFROG Gate Pulse <u>(relative to the fluorescence):</u>

Shorter

Synchronized

Bluer

Brighter

Requirements for the Fluorescence Excitation Pulse <u>(relative to the fluorescence):</u>

Shorter

Synchronized

Bluer

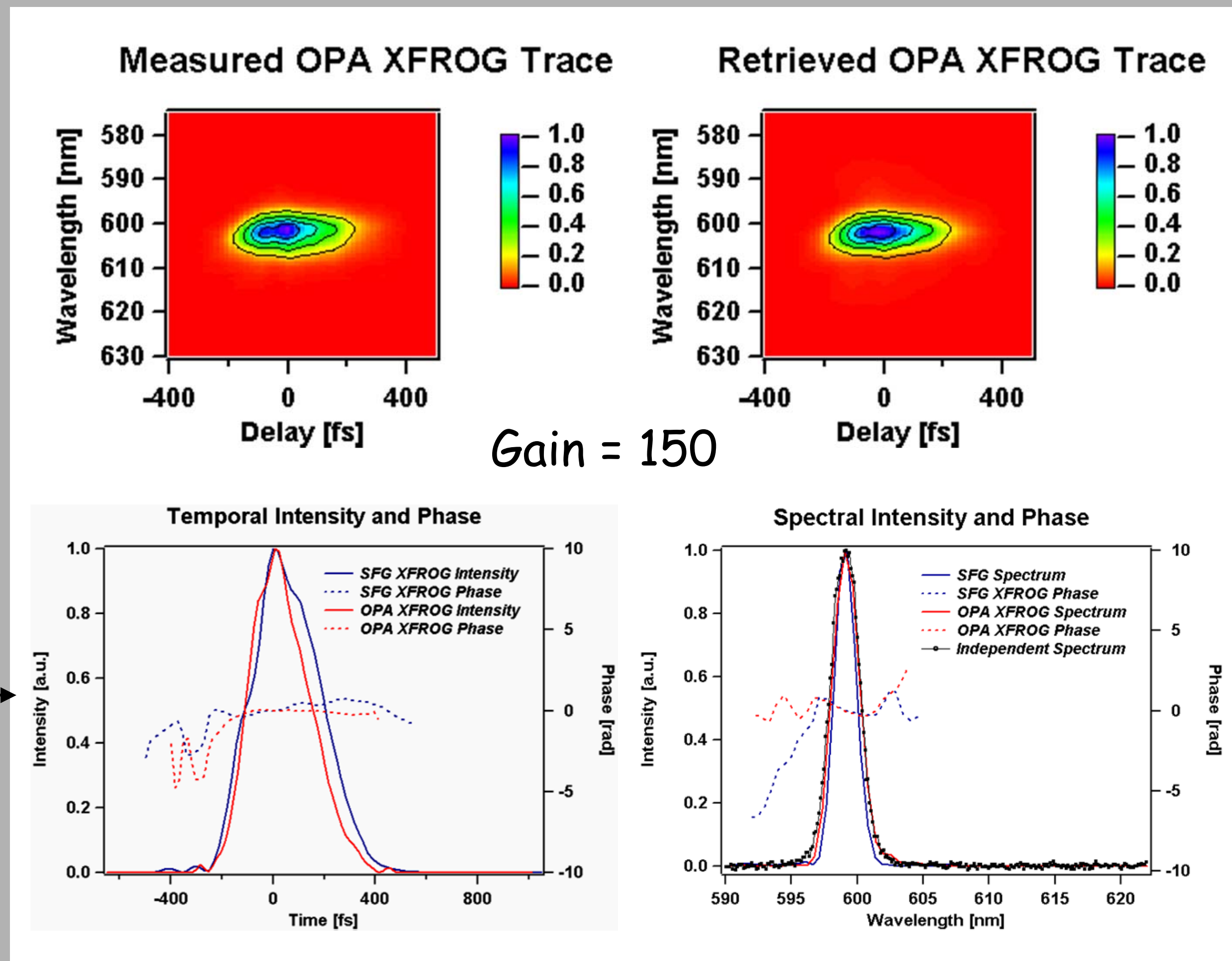
Brighter

Remarkably, the fluorescence excitation pulse will essentially always provide an ideal XFROG gate pulse.

OPA XFROG measurements of a weak (80 fJ) fluorescence-like pulse

Fluorescence-like test pulse: continuum created in bulk sapphire (spectrally filtered and attenuated)

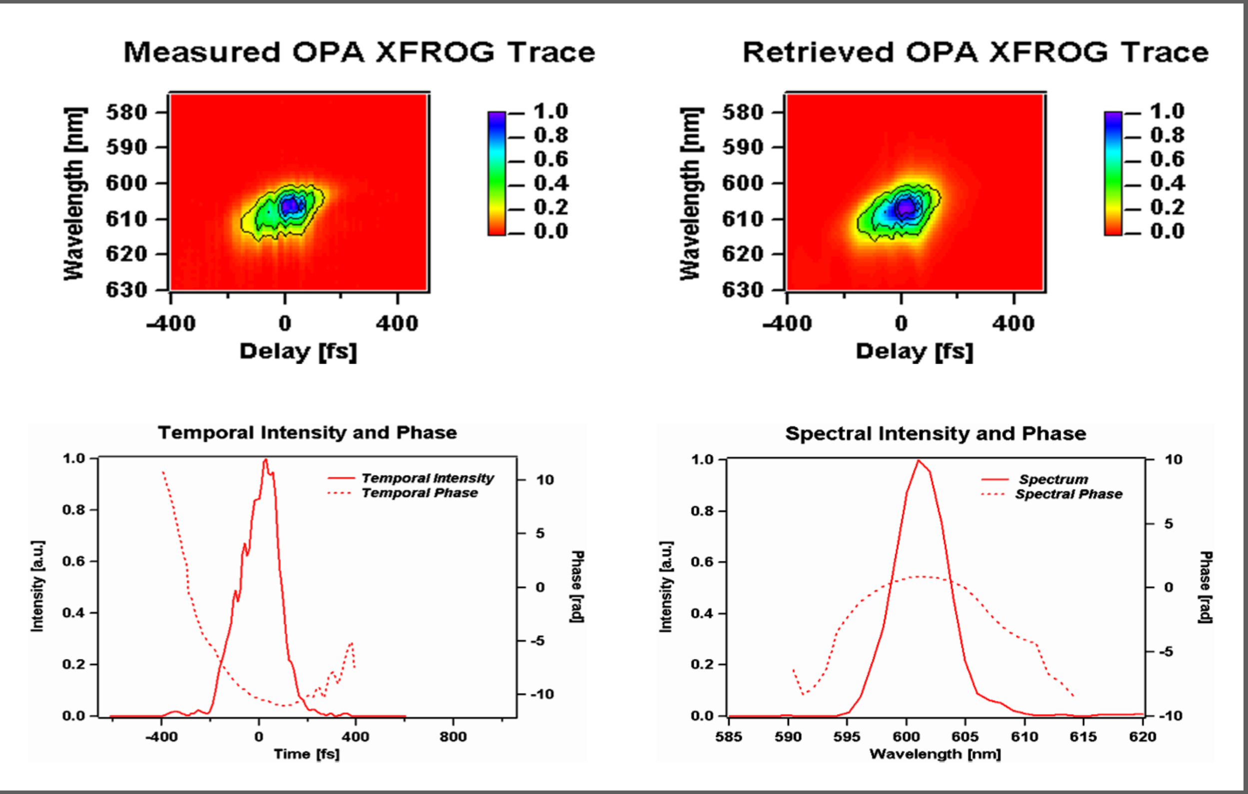
Comparison with an already well-established technique, SFG XFROG (measuring the same pulse, but less attenuated).



OPA XFROG measurements of a really weak (50 aJ) fluorescence-like pulse

The same continuum generated in bulk sapphire (filtered and now **heavily** attenuated).

Gain = 1000



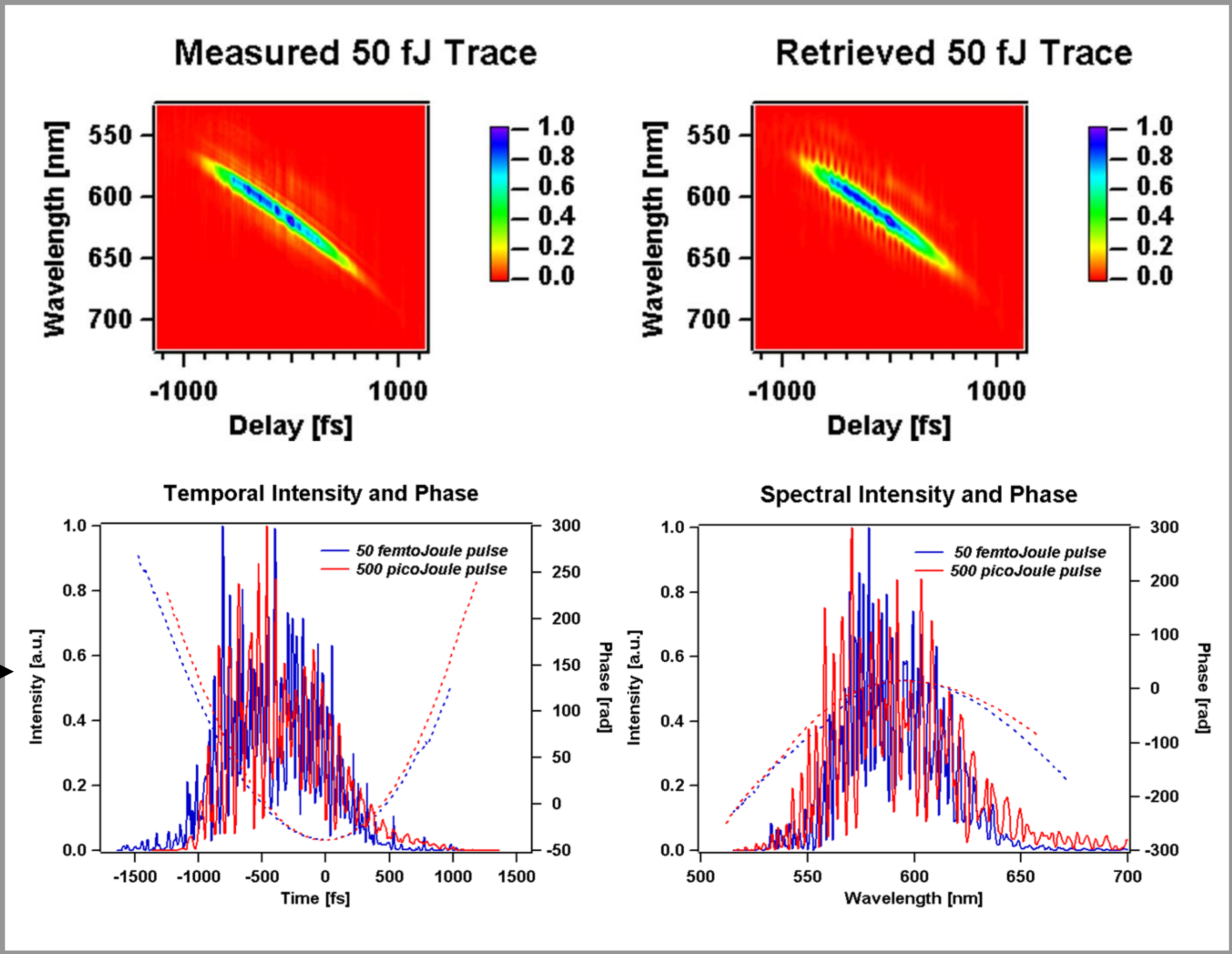
This measurement involved fewer input photons than our SI measurement of less than one photon per pulse.

OPA XFROG measurement of a broadband (100 nm), weak (50 fJ) pulse

Gain = 1000

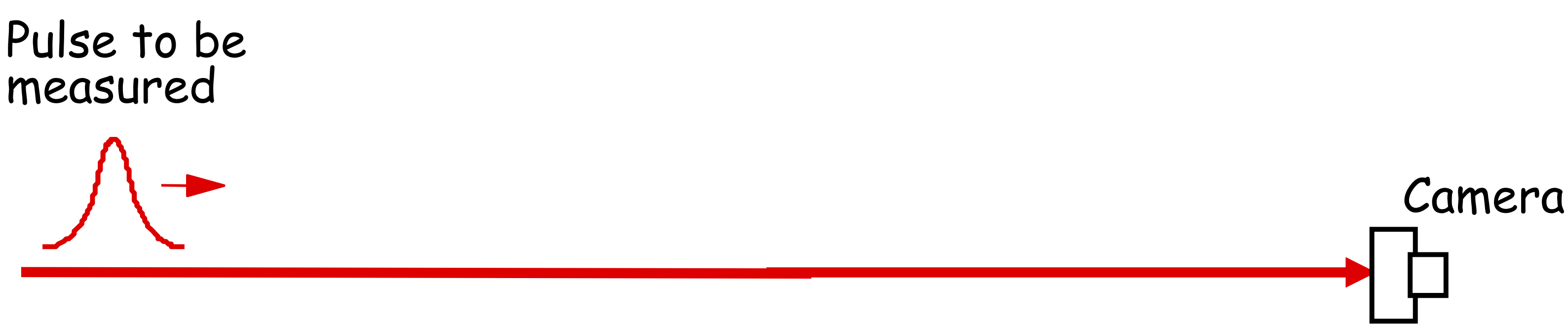
2-mm thick BBO

Comparison with a measurement of the bulk continuum at higher pulse energy (recall that continuum is unstable, so the structure will be different)



Can we simplify FROG?

Collaborators:
Mark Kimmel, Selcuk Akturk, and Patrick O'Shea

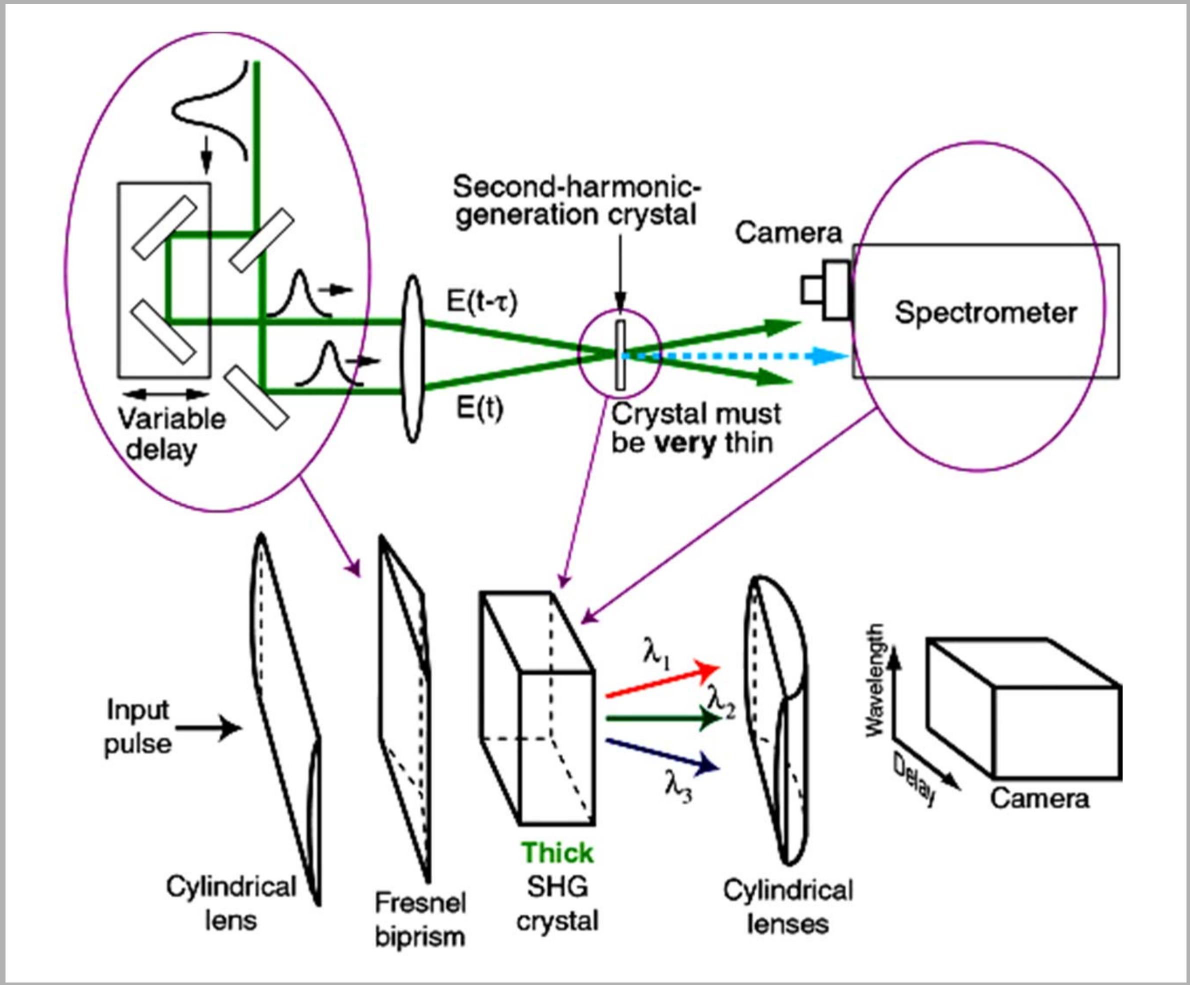


Remarkably, we can design a FROG without these components!

We can greatly simplify FROG!

FROG:
Frequency-
Resolved
Optical
Gating

GRENOUILLE:
GRating-
Eliminated
No-nonsense
Observation of
Ultrafast
Incident
Laser
Light
E-fields

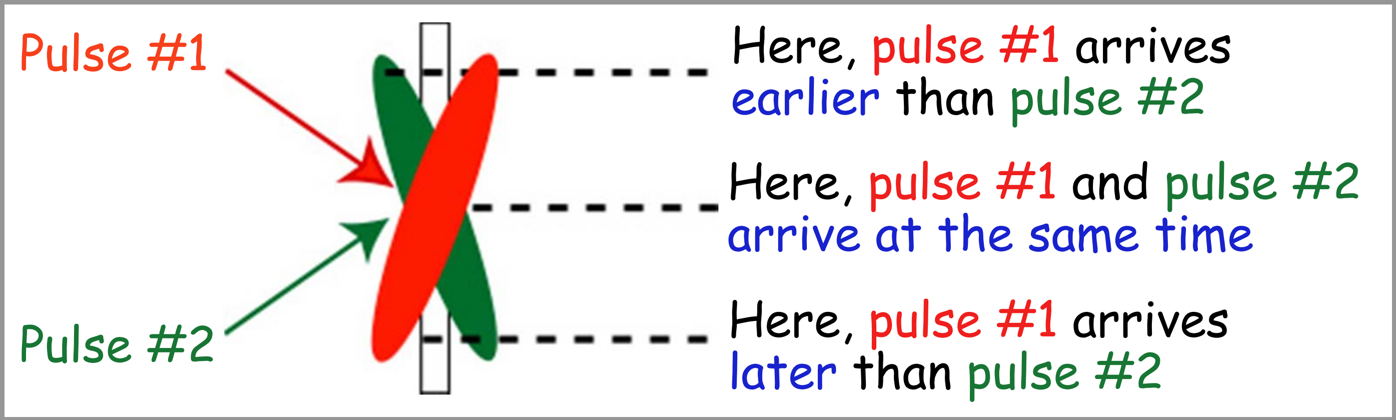


Winner, 2003 R&D100 award

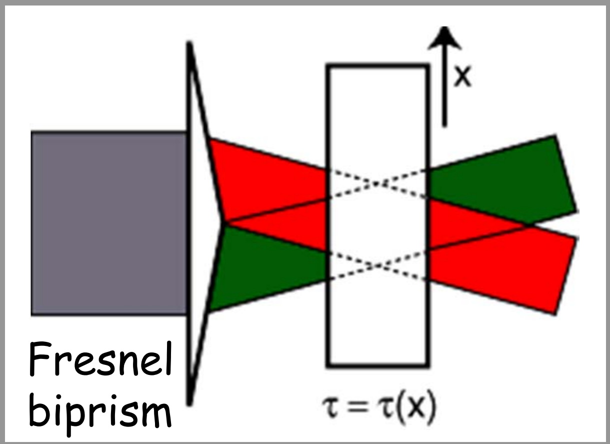
A *single* optic (a Fresnel biprism) replaces the *entire* delay line, and a *thick* SHG crystal replaces *both* the thin crystal *and* spectrometer.

Single-Shot FROG and the Fresnel biprism

Crossing beams at a large angle maps delay onto transverse position.



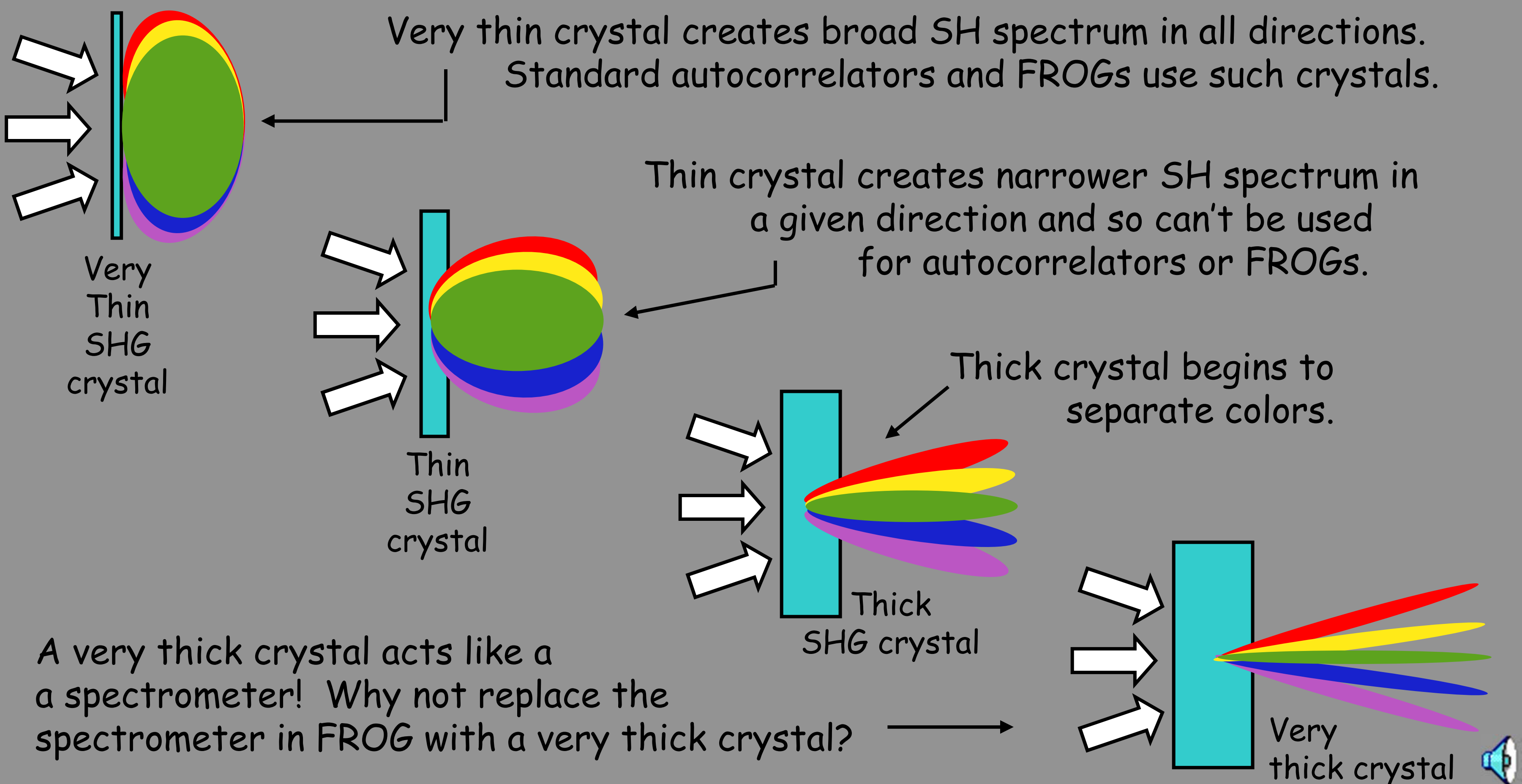
This avoids manually scanning the delay. But it still requires over-lapping the beams in space (and time). Here's how we avoid even that:



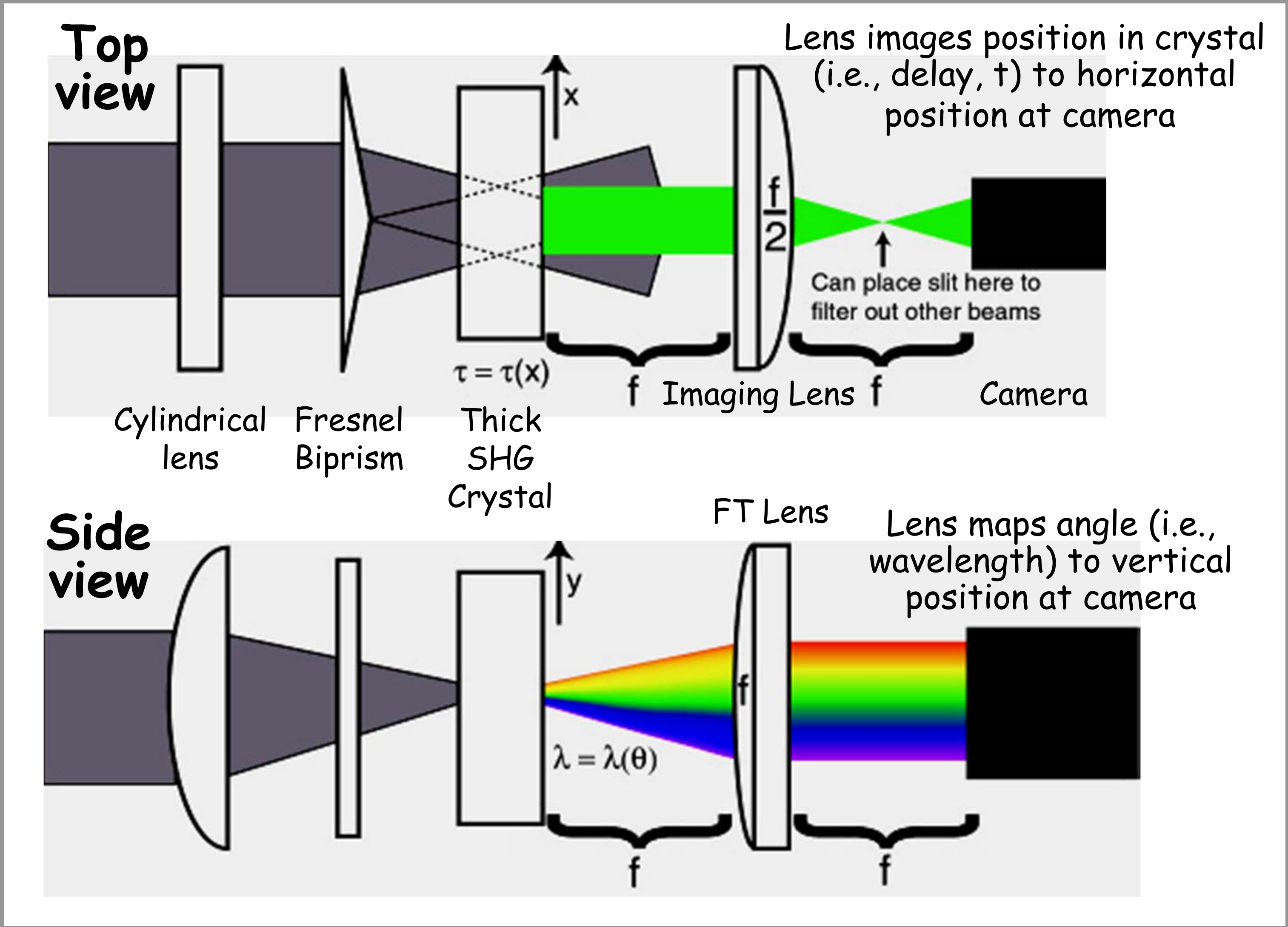
Even better, this design is amazingly compact and easy to use, and it never misaligns!

The angular width of second harmonic varies inversely with the crystal thickness.

Suppose white light with a large divergence angle impinges on an SHG crystal. The SH generated depends on the angle. And the angular width of the SH beam created varies inversely with the crystal thickness.



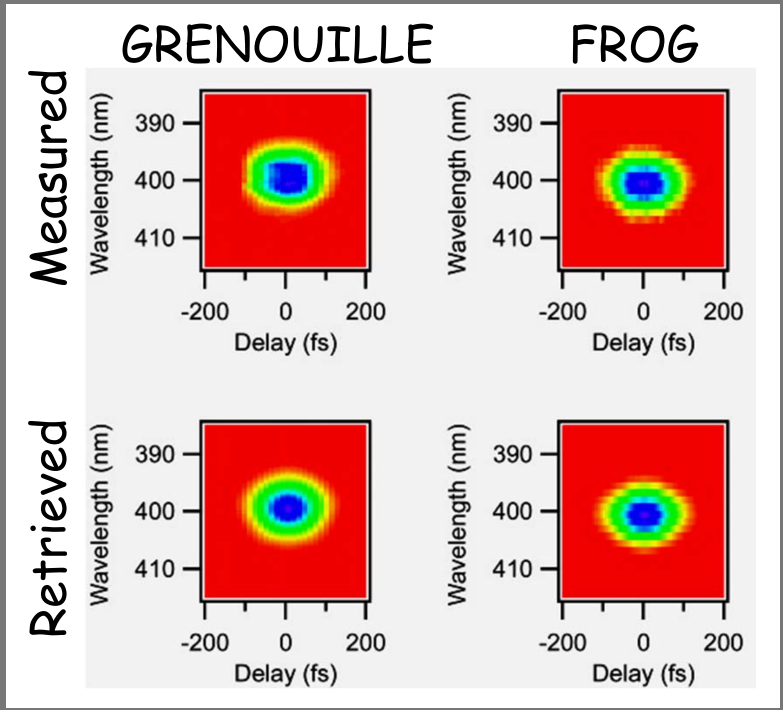
GRENOUILLE Beam Geometry



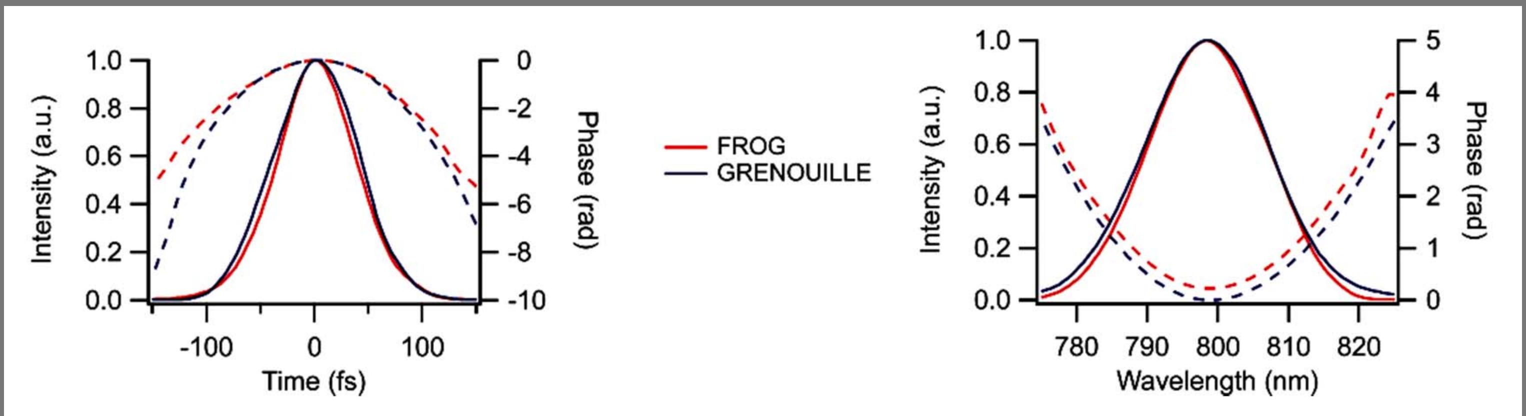
Yields a complete single-shot FROG. Uses the standard FROG algorithm.
Never misaligns. Is more sensitive. Measures spatio-temporal distortions!

Testing GRENOUILLE

Compare a GRENOUILLE measurement of a pulse with a tried-and-true FROG measurement of the same pulse:

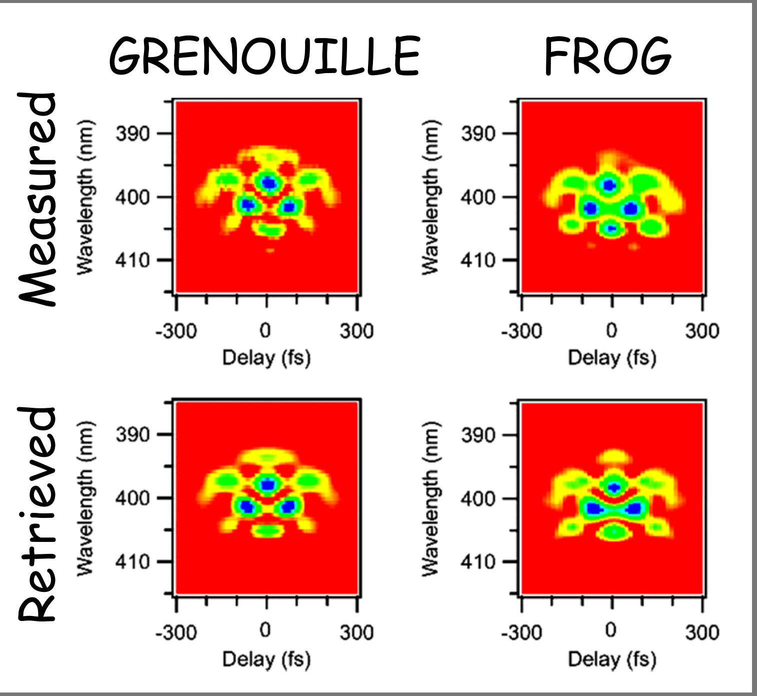


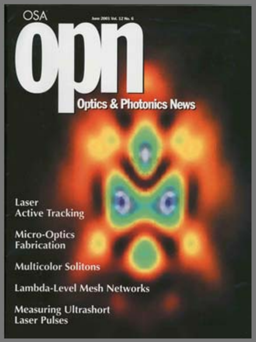
Retrieved pulse in the time and frequency domains



Really Testing GRENOUILLE

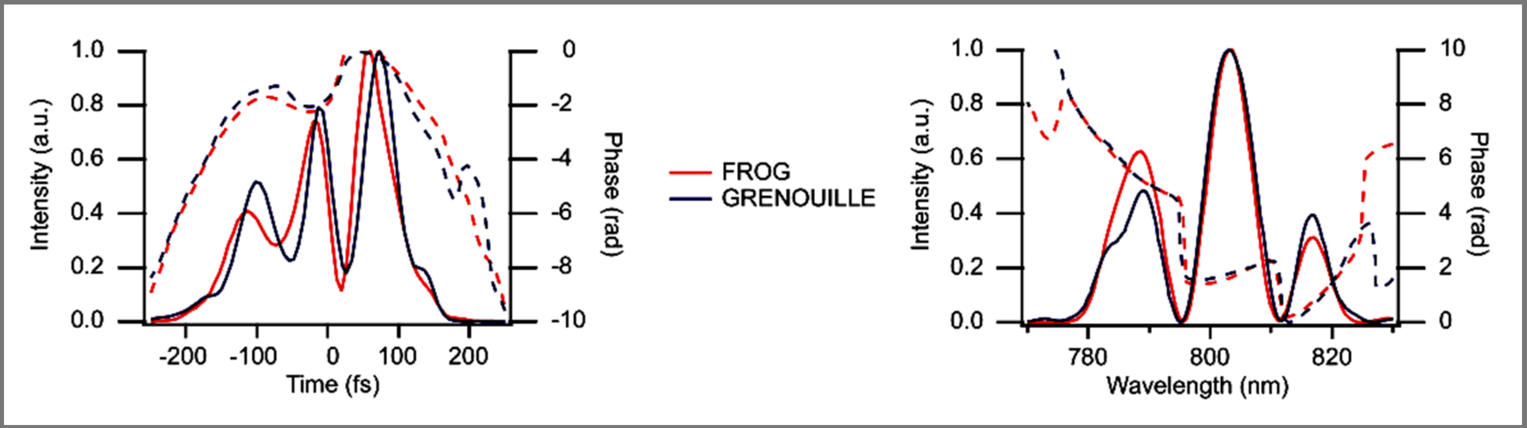
GRENOUILLE accurately measures even complex pulses.





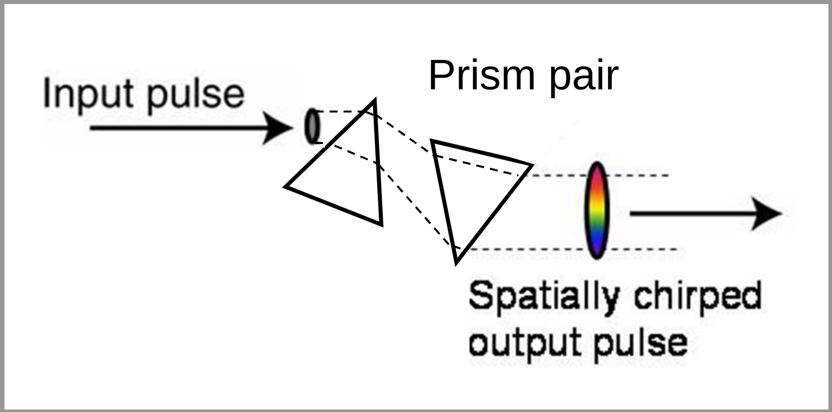
Read more about GRENOUILLE in the cover story of OPN, June 2001

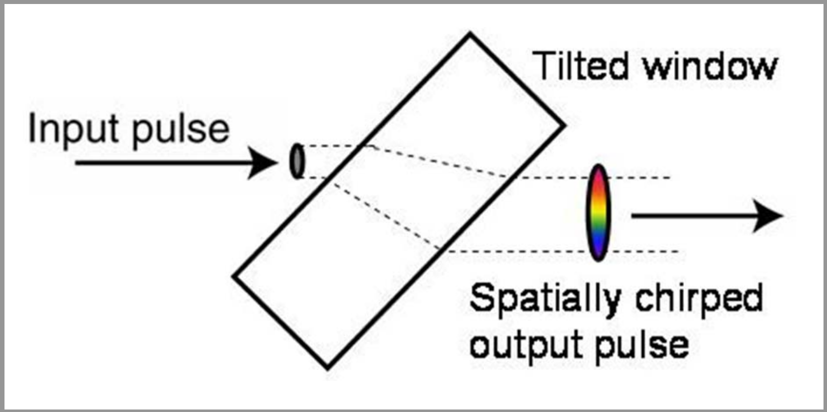
Retrieved pulse in the time and frequency domains



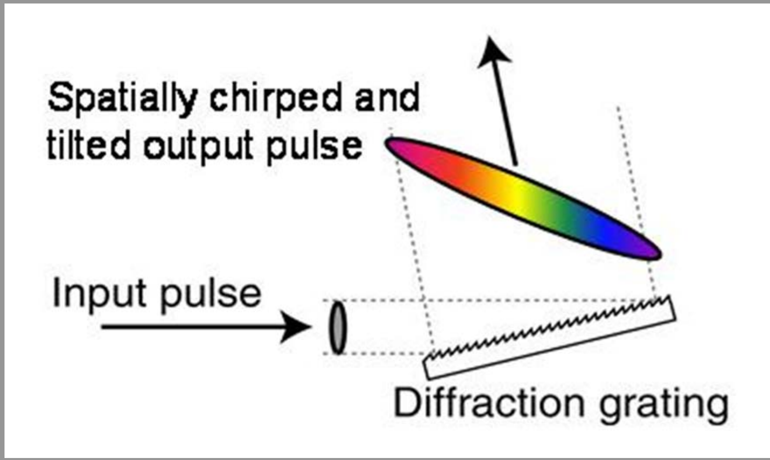
Spatio-temporal distortions in pulses

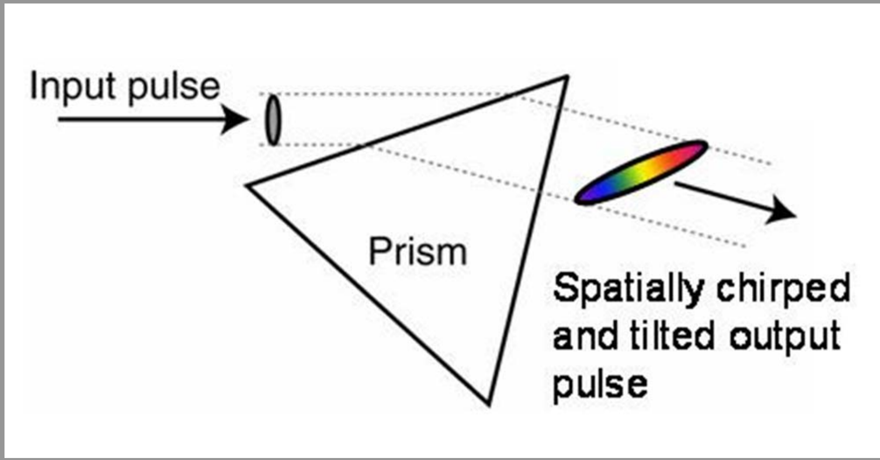
Prism pairs and simple tilted windows cause "spatial chirp."



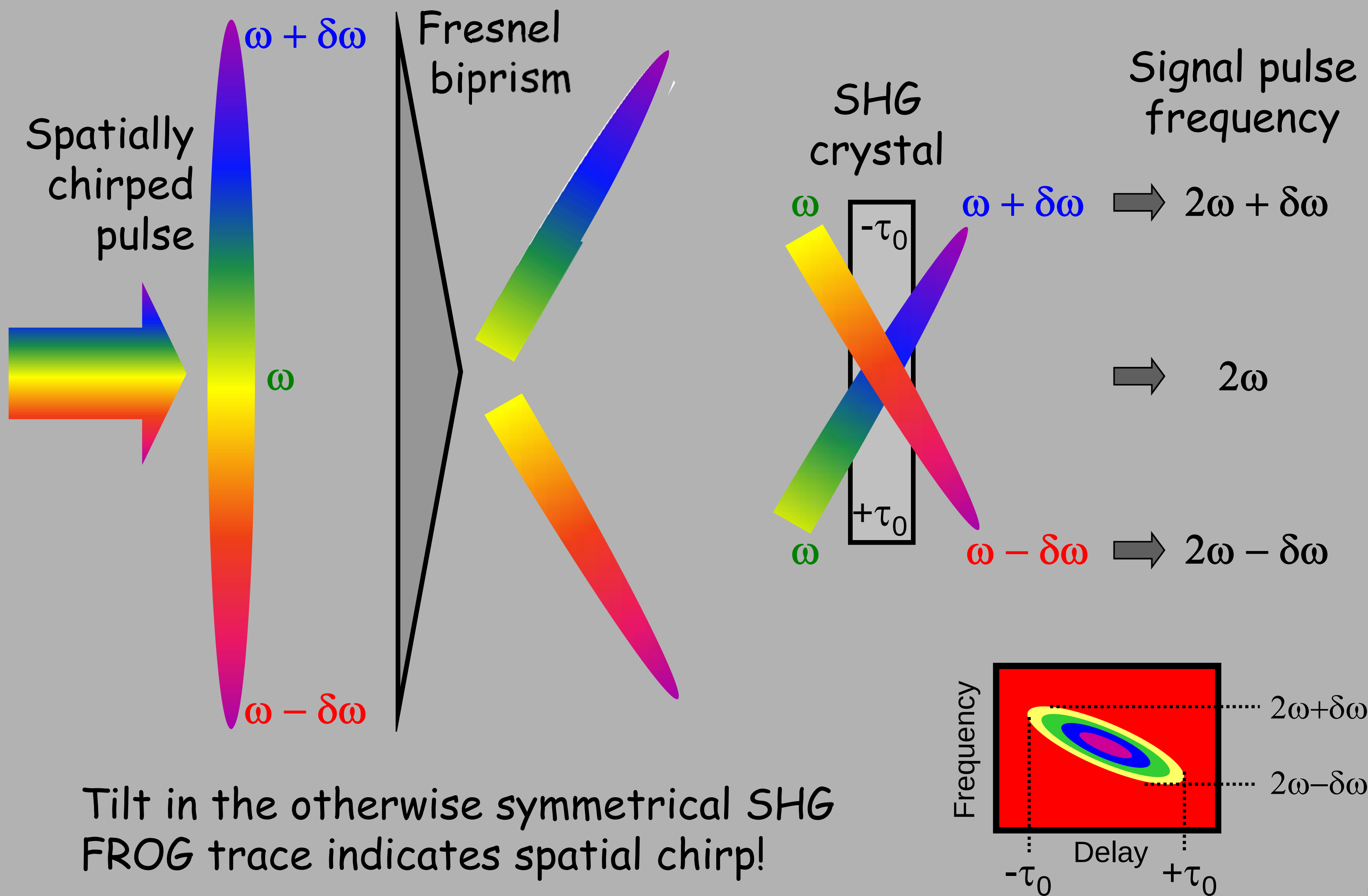


Gratings and prisms cause both spatial chirp and "pulse-front tilt."



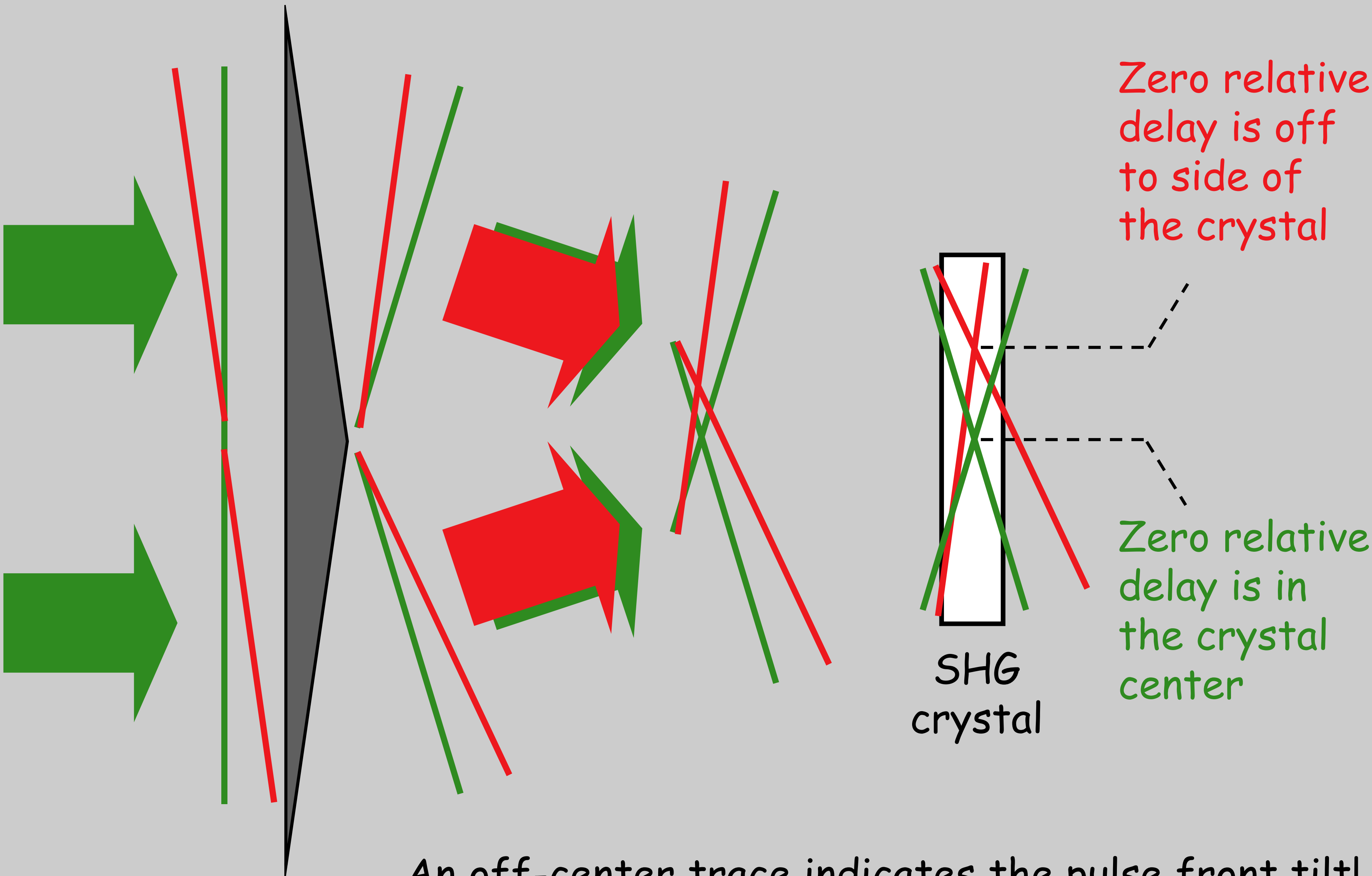


GRENOUILLE measures spatial chirp.



Tilt in the otherwise symmetrical SHG FROG trace indicates spatial chirp!

GRENOUILLE measures pulse-front tilt.



An off-center trace indicates the pulse front tilt!

To learn more, visit our web sites…

www.physics.gatech.edu/frog

www.swampoptics.com

And if you read only one ultrashort-pulse-measurement book this year, make it this one!