15. Measuring Ultrashort Laser Pulses II: FROG

The Musical Score and the Spectrogram

Frequency-Resolved Optical Gating (FROG)

1D vs. 2D Phase Retrieval

FROG as a 2D Phase-retrieval Problem

Second-harmonic-generation (SHG) FROG (and other geometries)

Measuring the shortest event ever created

Single-shot FROG, XFROG, TREEFROG, and GRENouille

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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 information on top about the intensity.

The musical score lives in the “time-frequency domain.”
A mathematically rigorous form of a musical score is the “spectrogram.”

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

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

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

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

The spectrogram is a function of $\omega$ and $\tau$. It is the set of spectra of all temporal slices of $E(t)$.
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 $\tau$.
Like a musical score, the spectrogram visually displays the frequency vs. time.
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 are “trivial.”

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

Suppose we use a delta-function gate pulse:

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

No phase information!

The spectrogram resolves the measurement 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 the pulse in an instantaneous nonlinear-optical medium, and then spectrally resolving the gated pulse.

Use any fast nonlinear-optical interaction: SHG, self-diffraction, etc.

Frequency-Resolved Optical Gating

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

The signal pulse reflects the color of the gated pulse, \( E(t) \), at the time \( 2\tau/3 \).

\( E(t) \) contributes phase (i.e., color), to the signal pulse.

\( \left| E(t-\tau) \right|^2 \) contributes only intensity, not phase (i.e., color), to the signal pulse.
FROG Traces for Linearly Chirped Pulses

Negatively chirped pulse
Unchirped pulse
Positively chirped pulse

The FROG trace visually displays the frequency vs. time.
FROG Traces for More Complex Pulses

Self-phase-modulated pulse

Cubic-spectral-phase pulse

Double pulse

Frequency

Time

Frequency

Delay

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

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

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

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

yields:

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

where: $g(t - \tau) = \left| E(t - \tau) \right|^2$

Unfortunately, spectrogram inversion algorithms require that we know the gate function.
Instead, consider FROG as a two-dimensional phase-retrieval problem.

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

$$I_{\text{FROG}}(\omega, \tau) = \left| \int \int \hat{E}_{\text{sig}}(t, \Omega) \exp(-i\omega t - i\Omega \tau) \, dt \, d\Omega \right|^2$$

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

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

This integral-inversion problem is the 2D phase-retrieval problem, for which the solution exists and is unique. And simple algorithms exist for finding it. Stark, Image Recovery, Academic Press, 1987.
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.

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!

These results are related to the Fundamental Theorem of Algebra.

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

Phase Retrieval and the Fundamental Theorem of Algebra

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

\[ f_{N-1}z^{N-1} + f_{N-2}z^{N-2} + \ldots + f_1z + f_0 = f_{N-1}(z-z_1)(z-z_2) \ldots (z-z_{N-1}) \]

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 and 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:

\[
F_k \equiv \sum_{m=0}^{N-1} f_m e^{-i mk} = \sum_{m=0}^{N-1} f_m z^m \quad \text{where} \quad z = e^{-ik}
\]

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:

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

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 \( \{F_{0,0}, \ldots, F_{N-1,N-1}\} \) of a discrete 2D data set, \( \{f_{0,0}, \ldots, f_{N-1,N-1}\} \), is:

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

where \( y = e^{-ik} \) and \( z = e^{-iq} \)

Polynomial of 2 variables!

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.
An iterative Fourier-transform algorithm finds the pulse intensity and phase

Constraint #1: mathematical form of optical nonlinearity

Start with noise

\[ E_{\text{sig}}(t, \tau) \]

Find \( E'_{\text{sig}}(t, \tau) \) such that

\[ E'_{\text{sig}}(t, \tau) \propto E(t) |E(t - \tau)|^2 \]

and is as close as possible to \( E_{\text{sig}}(t, \tau) \)

Inverse FFT with respect to \( \omega \)

\[ \tilde{E}_{\text{sig}}(\omega, \tau) \]

FFT with respect to \( t \)

\[ E'(t, \tau) \]

\[ E(t) \]

Replace the magnitude of \( E_{\text{sig}}(\omega, \tau) \) with \( \sqrt{\text{FROG}(\omega, \tau)} \)

Constraint #2: FROG trace data

Generalized Projections

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

Set of waveforms that satisfy nonlinear-optical constraint:

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

Set of waveforms that satisfy data constraint:

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

Convergence is guaranteed for convex sets, but generally occurs even with nonconvex sets.
Applying the Signal Field Constraint

We must find $E'_\text{sig}(t, \tau)$ such that $E'_\text{sig}(t, \tau) \propto E(t) |E(t - \tau)|^2$
and is as close as possible to $E_{\text{sig}}(t, \tau)$.

The way to do this is to find the field, $E(t)$, that minimizes:

$$Z \equiv \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \left| E_{\text{sig}}(t, \tau) - E(t) |E(t - \tau)|^2 \right|^2 dt \, d\tau$$

Once we find the $E(t)$ that minimizes $Z$, we write the new signal field as:

$$E'_\text{sig}(t, \tau) \propto E(t) |E(t - \tau)|^2$$

This is the new signal field in the iteration.
Ultrashort Laser Pulse Electric Fields Measured Using FROG

FROG Traces

FROG-derived Electric Fields

Data courtesy of Prof. Bern Kohler and Kent Wilson, Dept. of Chemistry, UCSD
Second-Harmonic-Generation FROG

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

We can use a second-harmonic-generation crystal for the nonlinear-optical medium.

\[ I_{FROG}(\omega, \tau) = \left| \int E_{\text{sig}}(t, \tau) e^{-i\omega t} \, dt \right|^2 \]

Second-harmonic generation (SHG) is the strongest NLO effect.

Kane and Trebino, JQE, 29, 571 (1993).
SHG FROG traces are symmetrical with respect to delay.

Negatively chirped pulse

Un chirped pulse

Positively chirped pulse

The direction of time is ambiguous in the retrieved pulse.
SHG FROG traces for complex pulses

- Self-phase-modulated pulse
- Cubic-spectral-phase pulse
- Double pulse

SHG FROG traces are symmetrized PG FROG traces.
SHG FROG Measurements of a Free-Electron Laser

SHG FROG works very well, even in the mid-IR and for difficult sources.

Shortest pulse vs. year

Plot prepared in 1994, reflecting the state of affairs at that time.

In the mid-1990's, the shortest pulse from a Ti:Sapphire laser was 10 fs. But its spectrum was broad enough to support a much shorter one.
The measured pulse spectrum had two humps, and the measured autocorrelation had wings.

Two different theories emerged, and both agreed with the data.

Despite different predictions for the pulse shape, both theories were consistent with the data.
FROG distinguishes between the theories.

SHG FROG Measurements of a 4.5-fs Pulse

FROG is simply a frequency-resolved nonlinear-optical signal that is a function of time and delay.

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

Many interactions have been used, e.g., polarization rotation in a fiber.
FROG traces of the same pulse for different geometries

SHG FROG traces are the least intuitive; TG, PG, and SD FROG traces are the most.
# FROG geometries: Pros and Cons

<table>
<thead>
<tr>
<th>Method</th>
<th>Diagram</th>
<th>Sensitivity</th>
<th>Ambiguities</th>
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<tr>
<td>Second-harmonic generation</td>
<td><img src="image" alt="SHG" /></td>
<td>0.001 nJ</td>
<td>Direction of time; Rel. phase of multiple pulses</td>
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<td>Third-harmonic generation</td>
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<td>1 nJ</td>
<td>Relative phase of multiple pulses; tightly focused beams</td>
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<td>Polarization-gate</td>
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<td>100 nJ</td>
<td>None</td>
</tr>
<tr>
<td>Self-diffraction</td>
<td><img src="image" alt="SD" /></td>
<td>1000 nJ</td>
<td>None</td>
</tr>
</tbody>
</table>
Single-shot FROG

Crossing beams at a large angle produces a range of delays across the nonlinear-optical medium and maps delay onto transverse position.

- Here, pulse #1 arrives earlier than pulse #2.
- Here, pulse #1 and pulse #2 arrive at the same time.
- Here, pulse #1 arrives later than pulse #2.

This effect allows us to measure a pulse on a single laser shot by using a large beam and a large beam angle.
Single-Shot Polarization-Gate FROG

FROG allows a very simple imaging spectrometer.

We can use the focus of the beam in the nonlinear medium as the entrance slit for a home-made imaging spectrometer (in multi-shot and single-shot FROG measurements).

This eliminates the need for a bulky expensive spectrometer as well as the need to align the beam through a tiny entrance slit (which would involve three sensitive alignment parameters)!
When a known reference pulse is available: 

**Cross-correlation FROG (XFROG)**

If a known pulse is available (it need not be shorter), then it can be used to fully measure the unknown pulse. In this case, we perform sum-frequency generation, and measure the spectrum vs. delay.

\[
E_{SF}(t, \tau) \propto E(t)E_g(t - \tau)
\]

The XFROG trace (a spectrogram):

\[
I_{XFROG}(\omega, \tau) \equiv \left| \int_{-\infty}^{\infty} E(t) E_g(t - \tau) \exp(-i \omega t) dt \right|^2
\]

XFROG completely determines the intensity and phase of the unknown pulse, provided that the gate pulse is not too long or too short. If a reasonable known pulse exists, use XFROG, not FROG.

Example of XFROG measurement: microstructure-fiber ultrabroadband continuum.

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

It’s important to measure it.
Ultrabroadband Continuum

Ultrabroadband continuum was created by propagating 1-nJ, 800-nm, 30-fs pulses through 16 cm of Lucent microstructure fiber. The 800-nm pulse was measured with FROG, so it made an ideal known gate pulse. This pulse has a time-bandwidth product of ~ 4000, and is the most complex ultrashort pulse ever measured.

Measuring two pulses simultaneously

It is not sufficient to be able to measure one ultrashort pulse. Most experiments that involve one ultrashort pulse also involve another; one as input, the other as output.

**TREEFROG: Twin Recovery of E-field Envelopes FROG**

Can we recover both pulses from a single trace?

The problem is equivalent to "Blind Deconvolution."
1D vs. 2D Blind Deconvolution

1D blind deconvolution:

\[ h(t) = \int_{-\infty}^{+\infty} f(t') g(t - t') \, dt' \quad \text{(h is of finite extent)} \]

- There are infinitely many solutions for \( f(t) \) and \( g(t) \).

2D blind deconvolution:

\[ h(x,y) = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} f(x',y') g(x - x', y - y') \, dx' \, dy' \quad \text{(h is of finite extent)} \]

- There is essentially one solution for \( f(x,y) \) and \( g(x,y) \)!

*Phase retrieval is a special case of blind deconvolution.*
TREETFROG is still under active study, and many variations exist. It will be useful in excite-probe spectroscopic measurements, which involve crossing two pulses with variable relative delay at a sample.

Can we simplify FROG?

FROG has 3 sensitive alignment degrees of freedom ($\theta$, $\phi$ of a mirror and also delay).

The thin crystal is also a pain.

Remarkably, we can design a FROG without these components!
GRating-Eliminated No-nonsense Observation of Ultrafast Incident Laser Light E-fields (GRENOUILLE)

2 key innovations:
A single optic that replaces the entire delay line, and a thick SHG crystal that replaces both the thin crystal and spectrometer.

The Fresnel biperism

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

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

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.

- **Very thin crystal** creates broad SH spectrum in all directions. Standard autocorrelators and FROGs use such crystals.

- **Thin crystal** creates narrower SH spectrum in a given direction and so can’t be used for autocorrelators or FROGs.

- **Thick crystal** begins to separate colors.

- **Very thick crystal** acts like a spectrometer! Why not replace the spectrometer in FROG with a very thick crystal?
**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
Disadvantages of GRENOUILLE

Its low spectral resolution limits its use to pulse lengths between ~ 20 fs and ~ 1 ps.

Like other single-shot techniques, it requires good spatial beam quality.

Improvements on the horizon:

Inclusion of GVD and GVM in FROG code to extend the range of operation to shorter and longer pulses.