## Ultratrumpųjų Lazerinių Impulsų Trukmės Matavimas

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Didžioji pranešimo dalis pavogta iš prof. R. Trebino paskaitų konspektų

# "Ultratrumpųjų" technologijų atsiradimas

Lažybos: Ar visos keturios bėgančio žirgo kojos pakyla į orą?





Leland Stanford

Eadweard Muybridge



The "Galloping Horse" Controversy Palo Alto, CA 1872

> Laikinė skyra: 1/60 sekundės

### Fotografavimas su blykste



"How to Make Apple sauce at MIT" 1964

Harold Edgerton MIT, 1942







"Splash on a Glass" Junior High School student 1996

#### Laikinė skyra: keletas mikrosekundžių

#### The 1999 Nobel Prize in Chemistry went to Professor Ahmed Zewail of Cal Tech for ultrafast spectroscopy.



Zewail used ultrafast-laser techniques to study how atoms in a molecule move during chemical reactions.

Laikinė skyra: šimtai femtosekundžių

# The Dilemma

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

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



Photograph taken by Harold Edgerton, MIT

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?

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

A laser pulse has the time-domain electric field:

$$E(t) = \operatorname{Re} \left\{ I(t)^{1/2} \exp \left[ i \omega_0 t - i \phi(t) \right] \right\}$$

$$\operatorname{Intensity} Phase$$

$$Equivalently, vs. frequency: \qquad (neglecting the negative-frequency component)$$

$$\widetilde{E}(\omega) = \operatorname{Re} \left\{ S(\omega - \omega_0)^{1/2} \exp \left[ -i \phi (\omega - \omega_0) \right] \right\}$$

$$\operatorname{Spectrum} Spectral Phase$$

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

#### Pulse Measurement in the Time Domain: Detectors

Detectors are devices that emit electrons in response to photons.

Examples: Photo-diodes, Photo-multipliers



Detectors have very **slow** rise and fall times: ~ 1 nanosecond.

As far as we're concerned, detectors have **infinitely slow** responses. They measure the time integral of the pulse intensity from  $-\infty$  to  $+\infty$ :

$$V_{detector} \propto \int_{-\infty}^{\infty} |E(t)|^2 dt$$

The detector output voltage is proportional to the pulse energy. By themselves, detectors tell us little about a pulse.

### Pulse Measurement in the Frequency Domain: The Spectrometer

The spectrometer measures the spectrum, of course. Wavelength varies across the camera, and the spectrum can be measured for a single pulse.



"Imaging spectrometers" allow many spectra to be measured simultaneously, one for each row of a 2D camera.

# Pulse Measurement in the Time Domain: The Michelson Interferometer

Measuring the interferogram is equivalent to measuring the spectrum.

### Okay, so how do we measure a pulse?

Result: Using only time-independent, linear filters, complete characterization of a pulse is **NOT** possible with a slow detector.

Translation: If you don't have a detector or modulator that is fast compared to the pulse width, you **CANNOT** measure the pulse intensity and phase with only linear measurements, such as a detector, interferometer, or a spectrometer.

V. Wong & I. A. Walmsley, Opt. Lett. **19**, 287-289 (1994) I. A. Walmsley & V. Wong, J. Opt. Soc. Am B, **13**, 2453-2463 (1996)

We need a shorter event, and we don't have one. But we do have the pulse itself, which is a start. And we can devise methods for the pulse to gate itself using optical nonlinearities.

## Pulse Measurement in the Time Domain: The Intensity Autocorrelator

Crossing beams in an SHG crystal, varying the delay between them, and measuring the second-harmonic (SH) pulse energy vs. delay yields the **Intensity Autocorrelation**:





### **Square Pulse and Its Autocorrelation**

Pulse

Autocorrelation

$$I(t) = \begin{cases} 1; |t| \le \Delta \tau_p^{FWHM} / 2 \\ 0; |t| > \Delta \tau_p^{FWHM} / 2 \end{cases}$$

$$A^{(2)}(\tau) = \begin{cases} 1 - \left| \frac{\tau}{\Delta \tau_A^{FWHM}} \right|; |\tau| \le \Delta \tau_A^{FWHM} \\ 0; |\tau| > \Delta \tau_A^{FWHM} \end{cases}$$



### **Gaussian Pulse and Its Autocorrelation**

Pulse

Autocorrelation

$$I(t) = \exp\left[-\left(\frac{2\sqrt{\ln 2}t}{\Delta \tau_p^{FWHM}}\right)^2\right]$$

$$A^{(2)}(\tau) = \exp\left[-\left(\frac{2\sqrt{\ln 2\tau}}{\Delta\tau_A^{FWHM}}\right)^2\right]$$



 $\Delta \tau_A^{FWHM} = 1.41 \Delta \tau_p^{FWHM}$ 

### Sech<sup>2</sup> Pulse and Its Autocorrelation



Since theoretical models for ideal ultrafast lasers usually predict sech<sup>2</sup> pulse shapes, people usually simply divide the autocorrelation width by 1.54 and call it the pulse width. Even when the autocorrelation is Gaussian...

### **Lorentzian Pulse and Its Autocorrelation**



$$\Delta \tau_A^{FWHM} = 2.0 \ \Delta \tau_p^{FWHM}$$

### **Autocorrelations of more complex intensities**

Autocorrelations nearly always have considerably less structure than the corresponding intensity.



An autocorrelation typically corresponds to more than one intensity. Thus the autocorrelation does not uniquely determine the intensity.

### Even nice autocorrelations have ambiguities.

These complex intensities have nearly Gaussian autocorrelations.



Conclusions drawn from an autocorrelation are unreliable.

# Autocorrelations of complex pulses: first consider a double pulse

Pulse

Autocorrelation

$$I(t) = I_0(t) + I_0(t + \tau_{sep})$$





### **Multi-shot Autocorrelations and "Wings"**

The delay is scanned over many pulses, averaging over any variations in the pulse shape from pulse to pulse. So results can be misleading.

Imagine a train of pulses, each of which is a double pulse. Suppose the double-pulse separation varies:



Wings also result if each pulse in the train has varying structure. And wings can result if each pulse in the train has the **same** structure! In this case, the wings actually yield the pulse width, and the central spike is called the **"coherence spike."** Be careful with such traces.

## **Autocorrelation of Very Complex Pulses**

As the intensity increases in complexity, its autocorrelation approaches a broad diffuse background with a coherence spike.



### **Third-Order Autocorrelation**

Third-order nonlinear-optical effects provide the 3rd-order intensity autocorrelation:

Polarization Gating (PG)  $\vec{k_2}$  $\vec{k_2}$  $\vec{k_3}$  $\vec{k_3}$ 

 $\omega_0 = \omega - \omega + \omega$  $\vec{k}_0 = \vec{k}_1 - \vec{k}_2 + \vec{k}_2$ 

 $A^{(3)}( au)$ 

 $\omega_0 = \omega - \omega + \omega$  $\vec{k}_0 = 2\vec{k}_1 - \vec{k}_2$ 

 $\omega_0 = \omega - \omega + \omega$ 

 $\vec{k}_0 = \vec{k}_1 - \vec{k}_2 + \vec{k}_2$ 

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

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

Note the 2

 $I^2(t)I(t-\tau)\,\mathrm{d}t$ 

 $E_{sig}^{TG}(t,\tau) \propto \begin{cases} E_{sig}^{PG}(t,\tau) \\ E_{sig}^{SD}(t,\tau) \end{cases}$ 

Third-harmonic generation (THG)  $\omega_0 = 3\omega$  $\vec{k}_0 = 2\vec{k}_1 + \vec{k}_2$ 

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

The third-order autocorrelation is not symmetrical, so it yields slightly more information, but not the full pulse. Third-order effects are weaker, so it's less sensitive and is used only for amplified pulses (> 1  $\mu$ J).



# Pulse Measurement in the Time Domain: The Interferometric Autocorrelator

What if we use a **collinear beam geometry**, and allow the autocorrelator signal light to interfere with the SHG from each individual beam?



Also called the "Fringe-Resolved Autocorrelation"

### **Interferometric Autocorrelation Math**

The measured intensity vs. delay is:

$$IA^{(2)}(\tau) = \int_{-\infty}^{\infty} \left[ E^{2}(t) + E^{2}(t-\tau) + 2E(t)E(t-\tau) \right] \left[ E^{*2}(t) + E^{*2}(t-\tau) + 2E^{*}(t)E^{*}(t-\tau) \right] dt$$

Multiplying this out:

$$IA^{(2)}(\tau) = \int_{-\infty}^{\infty} \left\{ \left| E^{2}(t) \right|^{2} + \left| E^{2}(t) E^{*2}(t-\tau) + 2E^{2}(t) E^{*}(t) E^{*}(t-\tau) + E^{2}(t-\tau) E^{*2}(t) + \left| E^{2}(t-\tau) \right|^{2} + 2E^{2}(t-\tau) E^{*}(t) E^{*}(t-\tau) + 2E(t)E(t-\tau) E^{*2}(t-\tau) + 2E(t)E(t-\tau) E^{*2}(t-\tau) + 4\left| E(t) \right|^{2} \left| E(t-\tau) \right|^{2} \right\} dt$$

$$= \int_{-\infty}^{\infty} \left\{ I^{2}(t) + E^{2}(t)E^{*2}(t-\tau) + 2I(t)E(t)E^{*}(t-\tau) + E^{2}(t-\tau)E^{*2}(t) + I^{2}(t-\tau) + 2I(t-\tau)E^{*}(t)E(t-\tau) + 2I(t-\tau)E^{*}(t)E(t-\tau) + 2I(t-\tau)E^{*}(t)E(t-\tau) + 2I(t-\tau)E^{*}(t-\tau) + 4I(t)I(t-\tau) \right\} dt$$

where  $I(t) \equiv |E(t)|^2$ 

### The Interferometric Autocorrelation is the sum of four different quantities.

$$= \int_{-\infty}^{\infty} I^{2}(t) + I^{2}(t-\tau) dt$$
 Constant (uninteresting)  
+  $4 \int_{-\infty}^{\infty} I(t)I(t-\tau) dt$  Intensity autocorrelation  
+  $2 \int_{-\infty}^{\infty} [I(t) + I(t-\tau)]E(t)E^{*}(t-\tau) dt + c.c$  Sum-of-intensities-weighted  
"interferogram" of  $E(t)$   
(oscillates at  $\omega$  in delay)  
Interferogram of the second harmonic

nic: +  $\int E^{2}(t)E^{2}(t-\tau) dt + c.c.$  equivalent to the spectrum of the SH (oscillates at 2 w in delay) (oscillates at  $2\omega$  in delay)

The interferometric autocorrelation simply combines several measures of the pulse into one (admittedly complex) trace. Conveniently, however, they occur with different oscillation frequencies:  $0, \omega$ , and  $2\omega$ .



### Interferometric Autocorrelation and Stabilization

To resolve the  $\omega$  and  $2\omega$  fringes, which are spaced by only  $\lambda$  and  $\lambda/2$ , we must actively stabilize the apparatus to cancel out vibrations, which perturb the delay by many  $\lambda$ .

Interferometric Autocorrelation Traces for a Flat-phase Gaussian pulse:



Fortunately, it's not always necessary to resolve the fringes.

## Interferometric Autocorrelation: Examples

The extent of the fringes (at  $\omega$  and  $2\omega$ ) indicates the approximate width of the interferogram, which is the coherence time. If it's the same as the width of the the low-frequency component, which is the intensity autocorrelation, then the pulse is near-Fourier-transform limited.



The interferometric autocorrelation nicely reveals the approximate pulse length and coherence time, and, in particular, their relative values.

# Does the interferometric autocorrelation yield the pulse intensity and phase?

**No.** The claim has been made that the Interferometric Autocorrelation, combined with the pulse interferogram (i.e., the spectrum), could do so (except for the direction of time).

Naganuma, IEEE J. Quant. Electron. 25, 1225-1233 (1989).

But the required iterative algorithm rarely converges.

The fact is that the interferometric autocorrelation yields little more information than the autocorrelation and spectrum.

We shouldn't expect it to yield the full pulse intensity and phase. Indeed, very different pulses have very similar interferometric autocorrelations.

#### **More Pulses with Similar Interferometric Autocorrelations**

Without trying to find ambiguities, we can try Pulses #3 and #4:



Interferometric Autocorrelations for Pulses #3 and #4



Chung and Weiner, IEEE JSTQE, 2001.

Despite very different pulse lengths, these pulses have nearly identical IAs.

# Nonlinear fluorescence and absorption are also used for autocorrelation, interferometric or not.



Resolving the sub- $\lambda$  fringes yields *interferometric* autocorrelation; otherwise not.

# 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.

# 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  $\tau$  is the delay.

Without  $g(t-\tau)$ ,  $\Sigma_E(\omega,\tau)$  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 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,  $\tau$ .

## **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:

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



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

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



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

### 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  $\Omega$  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.

### **Generalized Projections**

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.

# **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.

## **FROG Traces for More Complex Pulses**



### **FROG Measurements of a 4.5-fs Pulse!**



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

### Using FROG to align a pulse compressor

The grating separation must be correct, or the pulse will be chirped and long.



#### **FROG geometries:** Pros and Cons



# 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!



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 overlapping 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



# 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.



 $-\tau_0$ 

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

Frequency-Resolved Optical Gating: The Measurement of Ultrashort Laser Pulses

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**Rick Trebino**