14. Measuring Ultrashort Laser Pulses I: Autocorrelation

The dilemma

The goal: measuring the intensity and phase vs. time (or frequency)

Why?

The Spectrometer and Michelson Interferometer

1D Phase Retrieval

Autocorrelation

1D Phase Retrieval

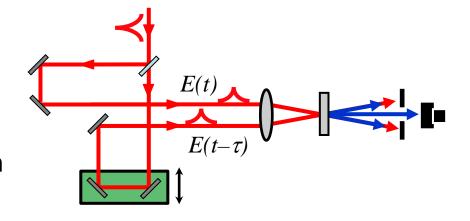
Single-shot autocorrelation

The Autocorrelation and Spectrum

Ambiguities

Third-order Autocorrelation

Interferometric Autocorrelation



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?

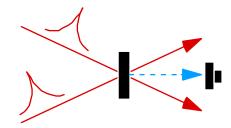
Ultrashort laser pulses are the shortest technological events ever created by humans.

It's routine to generate pulses shorter than 10⁻¹³ seconds in duration, and researchers have generated pulses only a few fs (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—as long as you've got 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.

Why measure an ultrashort laser pulse?

To determine the temporal resolution of an experiment using it.

To determine whether it can be made even shorter.

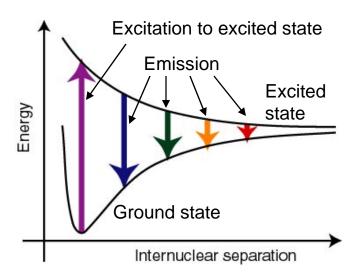
To better understand the lasers that emit them and to verify models of ultrashort pulse generation.

To better study media: the better we know the light in and light out, the better we know the medium we study with them.

To use pulses of specific intensity and phase vs. time to control chemical reactions: "Coherent control."

To understand pulse-shaping efforts for telecommunications, etc.

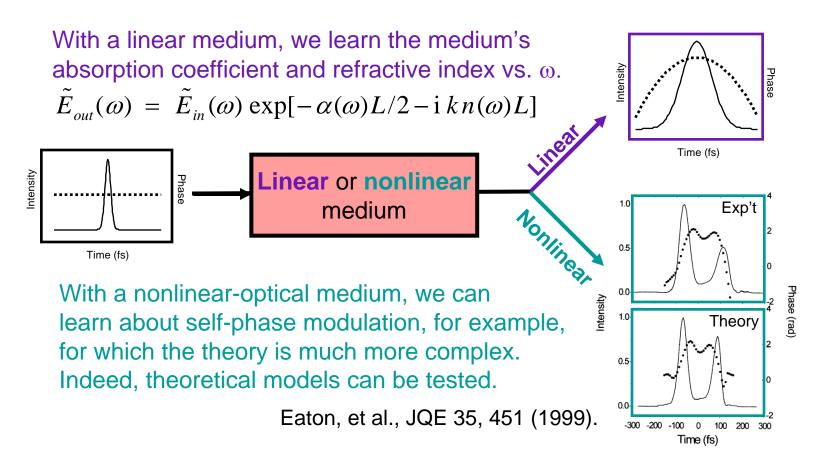
Because it's there.



As a molecule dissociates, its emission changes color (i.e., the phase changes), revealing much about the molecular dynamics, not available from the mere spectrum, or even the intensity vs. time.

Studying Media by Measuring the Intensity and Phase of Light Pulses

Measuring the intensity and phase of the pulses into and out of a medium tells us as much as possible about the linear and nonlinear effects in the medium.



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

A laser pulse has the time-domain electric field:

$$E(t) = Re \{ \underbrace{I(t)^{1/2}} exp [i\omega_0 t - i\phi(t)] \}$$
Intensity
Phase

Equivalently, vs. frequency:

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

$$\uparrow \qquad \qquad \uparrow$$
Spectrum
Spectral
Phase

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

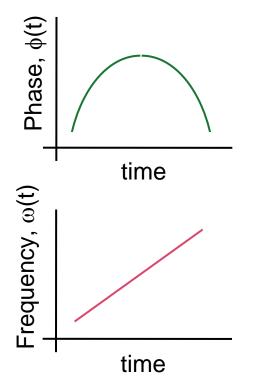
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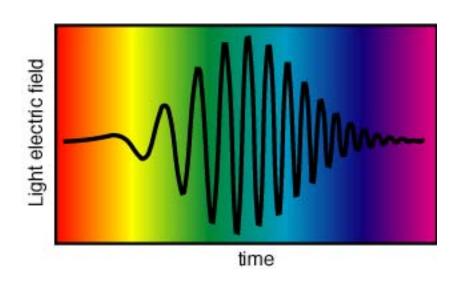
The phase determines the pulse's frequency (i.e., color) vs. time.

The instantaneous frequency:

$$\omega(\mathbf{t}) \equiv \omega_0 - \mathbf{d}\phi/\mathbf{d}\mathbf{t}$$

Example: "Linear chirp"

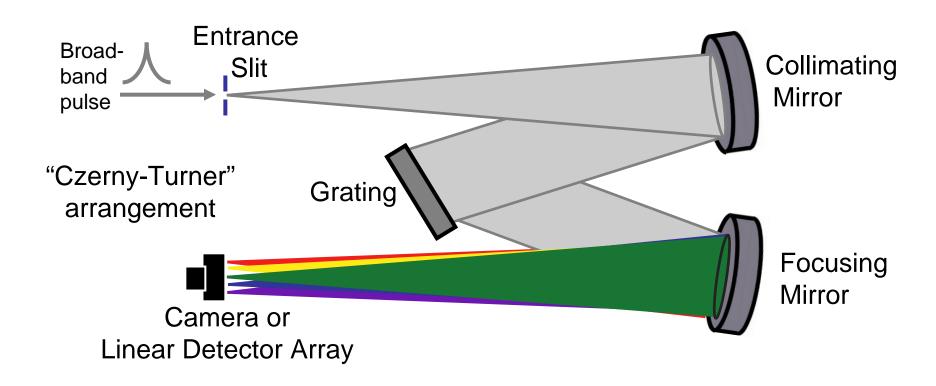




We'd like to be able to measure, not only linearly chirped pulses, but also pulses with arbitrarily complex phases and frequencies vs. time.

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.



There are numerous different arrangements for the optics of a spectrometer. This is just one example.

One-dimensional phase retrieval

It's more interesting than it appears to ask what information we lack when we know only the pulse spectrum.

Recall:
$$\tilde{E}(\omega) = \int_{-\infty}^{\infty} E(t)e^{-i\omega t}dt \qquad \text{and} \qquad \frac{S(\omega)}{\varphi(\omega)} = \frac{|\tilde{E}(\omega)|^2}{|\tilde{E}(\omega)|^2}$$
 Spectrum
$$\varphi(\omega) = \frac{|\tilde{E}(\omega)|^2}{|\tilde{E}(\omega)|^2}$$

Clearly, what we lack is the **spectral phase**.

But can we somehow retrieve it?

Mathematically, this problem is called the 1D phase retrieval problem.

Obviously, we cannot retrieve the spectral phase from the mere spectrum.

But what if we have some **additional** information? What if we know we have a **pulse**, which is, say, finite in duration?

There are still infinitely many solutions for the spectral phase.

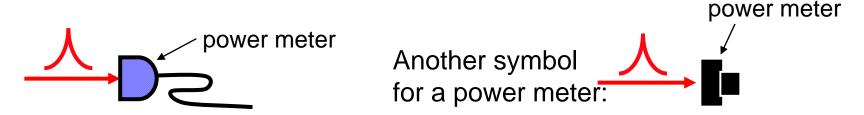
The 1D Phase Retrieval Problem is unsolvable.

E.J. Akutowicz, Trans. Am. Math. Soc. **83**, 179 (1956) E.J. Akutowicz, Trans. Am. Math. Soc. **84**, 234 (1957)

Pulse Measurement in the Time Domain: Power meters

Power meters are devices that emit electrons in response to photons.

Examples: Photo-diodes, Photo-multipliers



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

As far as we're concerned, they 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, power meters tell us little about a pulse.

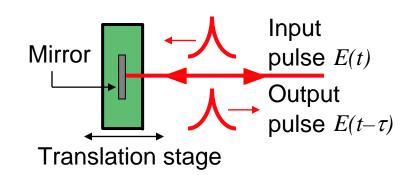
Pulse Measurement in the Time Domain: Varying the pulse delay

Since detectors are essentially infinitely slow, how do we make timedomain measurements on or using ultrashort laser pulses?

We'll delay a pulse in time.

And how will we do that?

By simply moving a mirror!



Moving a mirror backward by a distance L yields a delay of:

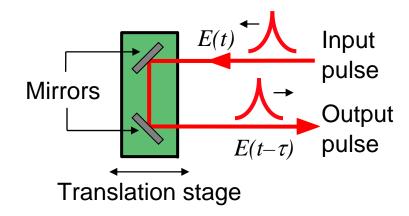
$$\tau = 2L/c$$

Do not forget the factor of 2!
Light must travel the extra distance
to the mirror—and back!

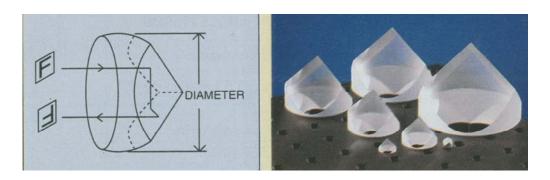
Since light travels 300 µm per psec, 300 µm of mirror displacement yields a delay of 2 ps. Controllable delay steps of less than 1 fs are not too difficult to implement.

We can also vary the delay using a mirror pair or corner cube.

Mirror pairs involve two reflections and displace the return beam in space:
But out-of-plane tilt yields a nonparallel return beam.



Corner cubes involve three reflections and also displace the return beam in space. Even better, they always yield a parallel return beam:



[&]quot;Hollow corner cubes" avoid propagation through glass.

Pulse Measurement in the Time Domain:

The Michelson Interferometer

$$V_{MI}(\tau) \propto \int_{-\infty}^{\infty} \left| E(t) + E(t - \tau) \right|^{2} dt$$

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

Beam-splitter

detector

Delay

$$\Rightarrow V_{MI}(\tau) \propto 2 \int_{-\infty}^{\infty} |E(t)|^2 dt + 2 \operatorname{Re} \int_{-\infty}^{\infty} E(t) E^*(t-\tau) dt$$

 ∑ Pulse energy (boring)

 ∑ Field autocorrelation (maybe interesting, but...)

 ∑ The FT of the field autocorrelation is just the spectrum!

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.

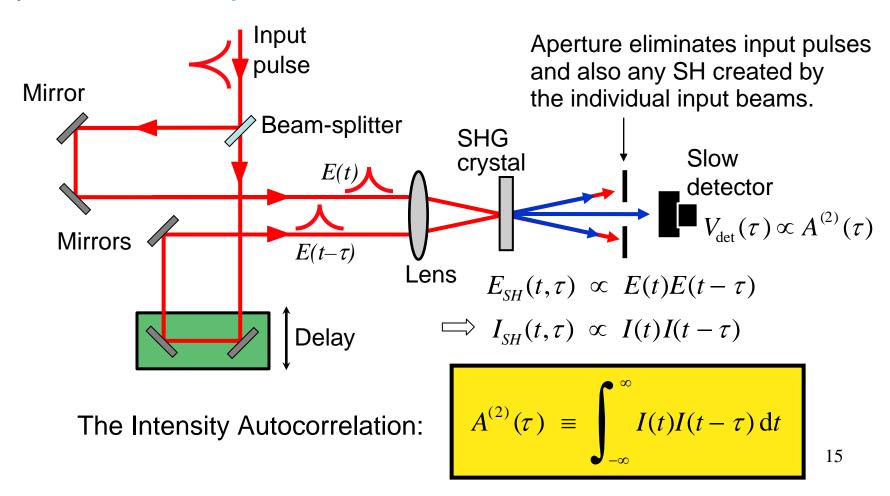
Translation: If you don't have a detector or modulator that is fast compared to the pulse width, you **CANNOT** measure the pulse intensity and phase with only linear measurements, such as a detector, interferometer, or a spectrometer.

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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)
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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**:



Practical Issues in Autocorrelation

Group-velocity mismatch must be negligible, or the measurement will be distorted. Equivalently, the phase-matching bandwidth must be sufficient. So very thin crystals (<100 µm!) must be used. This reduces the efficiency and hence the sensitivity of the device.

Conversion efficiency must be kept low, or distortions due to "depletion" of input light fields will occur.

The beam overlap in space must be maintained as the delay is scanned.

Minimal amounts of glass must be used in the beam before the crystal to minimize the GVD introduced into the pulse by the autocorrelator.

It's easy to introduce systematic error. The only feedback on the measurement quality is that it should be maximal at $\tau = 0$ and symmetrical in delay:

$$A^{(2)}(-\tau) = A^{(2)}(\tau) \qquad \text{because} \qquad \int I(t)I(t-\tau) \, dt = \int I(t'+\tau)I(t') \, dt'$$
$$t' = t - \tau$$

Square Pulse and Its Autocorrelation

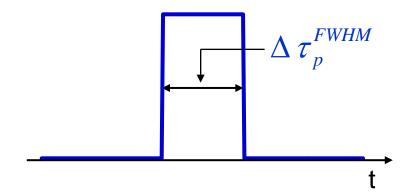
Pulse

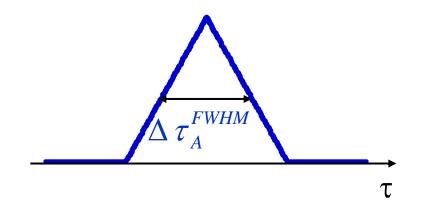
Autocorrelation

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

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

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





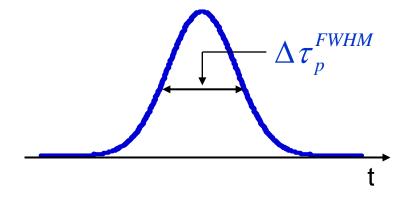
$$\Delta \tau_A^{FWHM} = \Delta \tau_p^{FWHM}$$

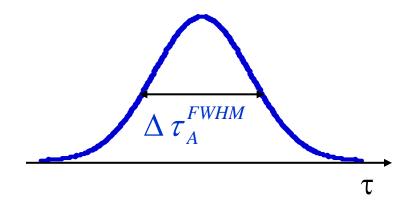
Gaussian Pulse and Its Autocorrelation

Pulse

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

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





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

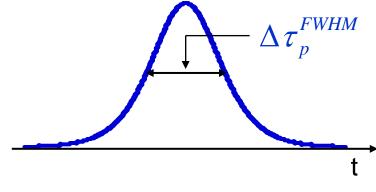
Sech² Pulse and Its Autocorrelation

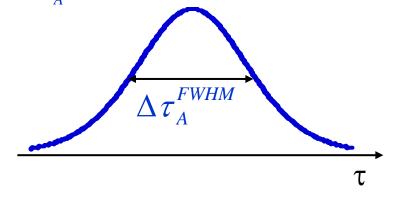
Pulse

Autocorrelation

$$I(t) = \operatorname{sech}^{2} \left[\frac{1.7627 t}{\Delta t_{p}^{FWHM}} \right]$$

$$A^{(2)}(\tau) = \frac{3}{\sinh^2\left(\frac{2.7196\,\tau}{\Lambda\,\tau_A^{FWHM}}\right)} \left[\frac{2.7196\,\tau}{\Delta\,\tau_A^{FWHM}} \coth\left(\frac{2.7196\,\tau}{\Delta\,\tau_A^{FWHM}}\right) - 1\right]$$

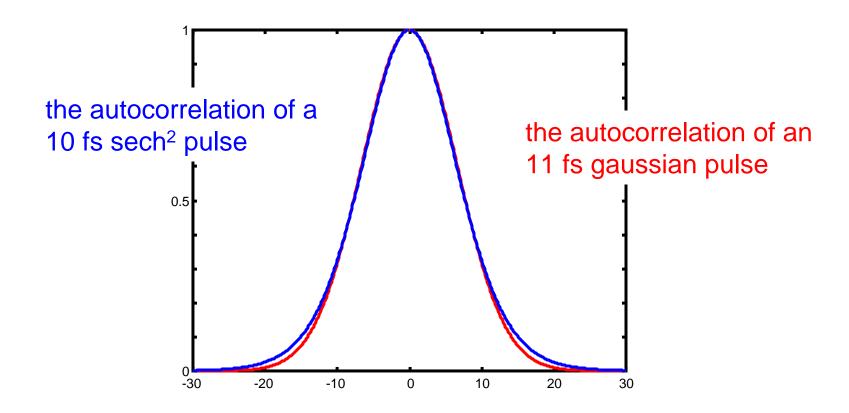




$$\Delta \tau_A^{FWHM} = 1.54 \ \Delta \tau_p^{FWHM}$$

Since theoretical models for ideal ultrafast lasers often predict sech² pulse shapes, people used to (and some still do) simply divide the autocorrelation width by 1.54 and call it the pulse width. Even when the autocorrelation is Gaussian...

Sech² pulse vs Gaussian pulse



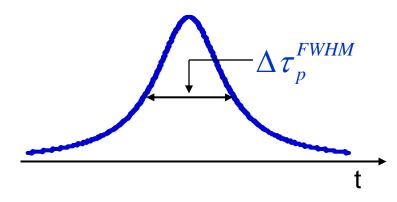
Lorentzian Pulse and Its Autocorrelation

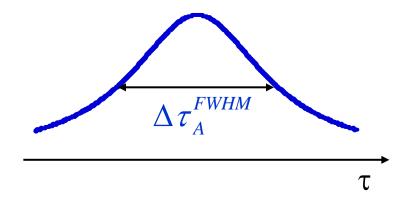
Pulse

Autocorrelation

$$I(t) = \frac{1}{1 + \left(2t/\Delta \tau_p^{FWHM}\right)^2}$$

$$A^{(2)}(\tau) = \frac{1}{1 + \left(2\tau/\Delta \tau_A^{FWHM}\right)^2}$$





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

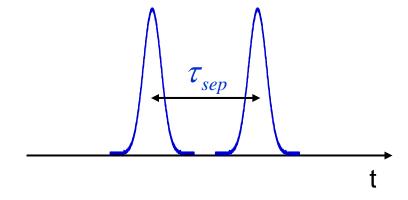
A Double Pulse and Its Autocorrelation

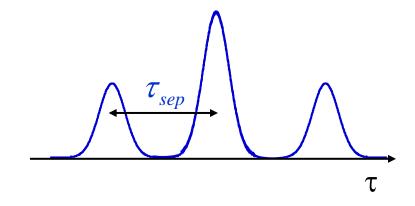
Pulse

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

Autocorrelation

$$A^{(2)}(\tau) = A_0^{(2)}(\tau + \tau_{sep}) + 2A_0^{(2)}(\tau) + A_0^{(2)}(\tau - \tau_{sep})$$



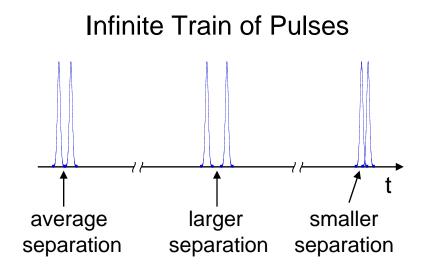


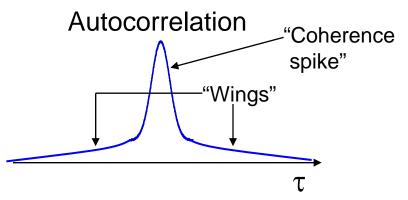
where:
$$A_0^{(2)}(\tau) = \int I_0(t) I_0(t-\tau) dt$$

Multi-shot Autocorrelation 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:



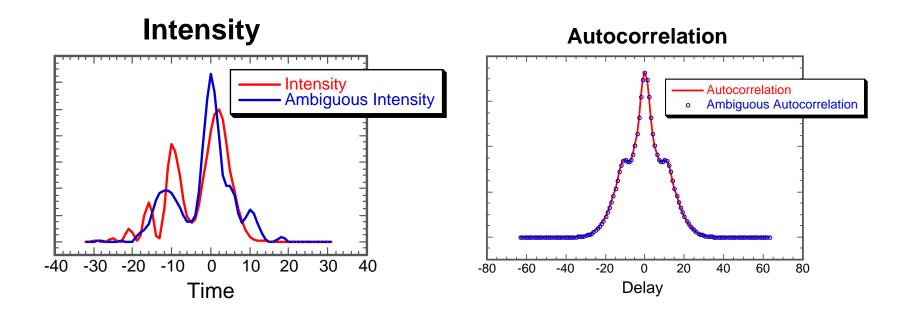


The locations of the side pulses in the autocorrelation vary from pulse to pulse. The result is "wings."

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.

Autocorrelations of more complex intensities

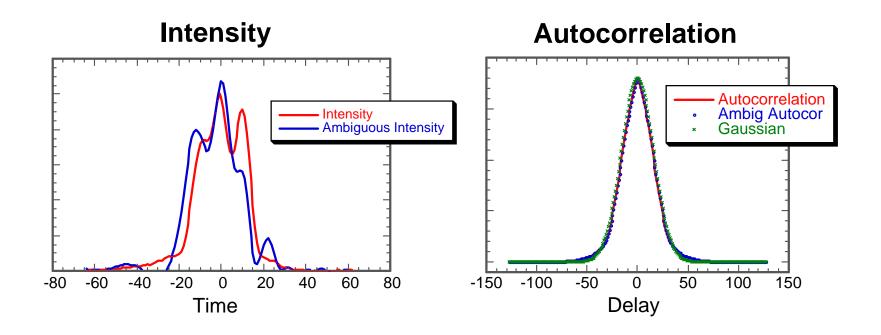
Autocorrelations nearly always have considerably less structure than the corresponding intensity.



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.

Retrieving the Intensity from the Intensity Autocorrelation is also equivalent to the 1D Phase-Retrieval Problem!

$$A^{(2)}(\tau) = \int I(t)I(t-\tau) dt$$

Applying the Autocorrelation Theorem:

$$\mathscr{F}\{A^{(2)}(au)\} \propto \left| \mathscr{F}\{I(t)\} \right|^2$$

Thus, the autocorrelation yields only the magnitude of the Fourier Transform of the Intensity. It says nothing about its phase! It's the 1D Phase-Retrieval Problem again!

We do have additional information: I(t) is always positive. The positivity constraint reduces the ambiguities dramatically, but still, it rarely eliminates them all.

The Intensity Autocorrelation is **not** sufficient to determine the intensity of the pulse vs. time.

Pulse Measurement in Both Domains: Combining the Spectrum and Autocorrelation

Perhaps the combined information of the autocorrelation and the spectrum could determine the pulse intensity and phase.

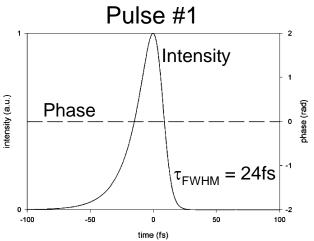
This idea has been called: "Temporal Information Via Intensity (TIVI)"

J. Peatross and A. Rundquist, J. Opt. Soc. Am B 15, 216-222 (1998)

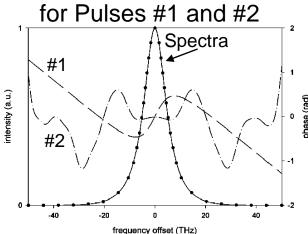
It involves an iterative algorithm to find an intensity consistent with the autocorrelation. Then it involves another iterative algorithm to find the temporal and spectral phases consistent with the intensity and spectrum.

Neither step has a unique solution, so this doesn't work.

Ambiguities in TIVI: Pulses with the Same Autocorrelation and Spectrum

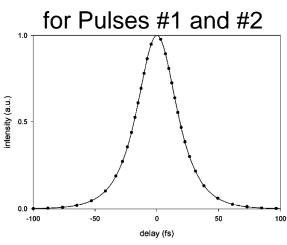


Spectra and spectral phases



Pulse #2 Phase $\tau_{\text{FWHM}} = 21 \text{fs}$ Intensity time (fs)

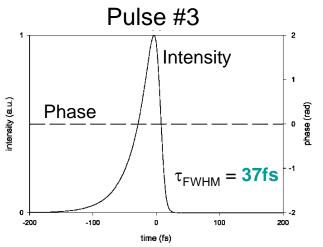
Autocorrelations

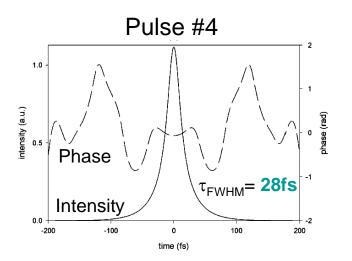


Chung and Weiner, IEEE JSTQE, 2001.

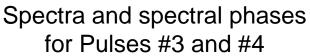
These pulses—especially the phases—are very different.

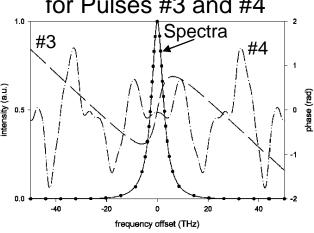
Ambiguities in TIVI: More Pulses with the Same Autocorrelation and Spectrum

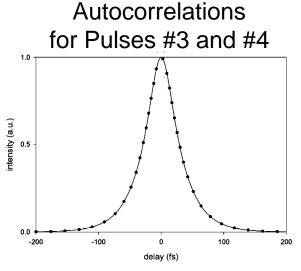




Chung and Weiner, IEEE JSTQE, 2001.







Despite having very different lengths, these pulses have the same autocorrelation and spectrum!

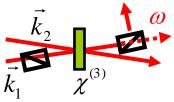
There's no way to know all the pulses having a given autocorrelation and spectrum.

Third-Order Autocorrelation

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



Polarization Gating (PG)



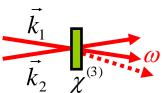
$$\omega_0 = \omega - \omega + \omega$$

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

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

Note the 2

Self-diffraction (SD)

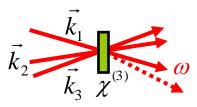


$$\omega_0 = \omega - \omega + \omega$$

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

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

Transient Grating (TG)



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

$$\vec{k}_0 = \vec{k}_1 - \vec{k}_2 + \vec{k}_3$$

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

Third-harmonic generation (THG)

$$\vec{k}_1$$
 \vec{k}_2 $\chi^{(3)}$

$$\omega_0 = 3\omega$$

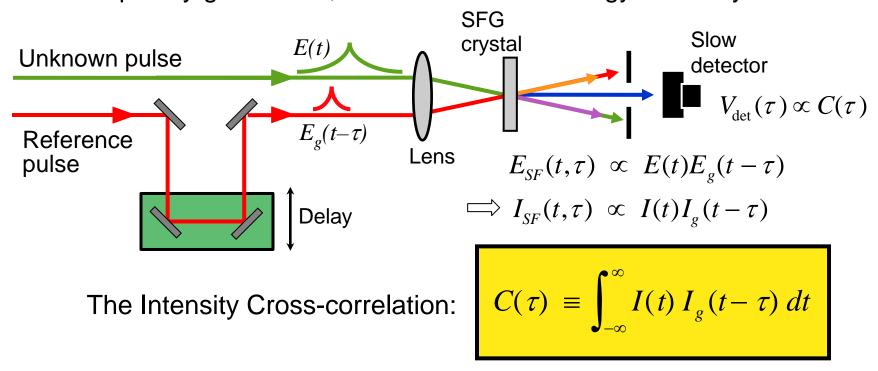
$$\vec{k}_0 = 2\vec{k}_1 + \vec{k}_2$$

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

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 μ J).

When a shorter reference pulse is available: The Intensity Cross-Correlation

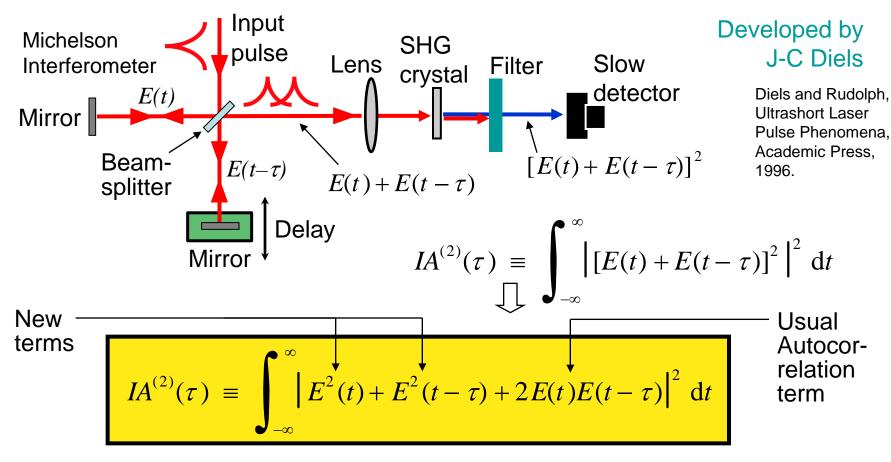
If a shorter reference pulse is available (*it need not be known*), then it can be used to measure the unknown pulse. In this case, we perform sum-frequency generation, and measure the energy vs. delay.



If the reference pulse is much shorter than the unknown pulse, then the intensity cross-correlation fully determines the unknown pulse intensity.

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:

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

$$\left. - \left| E^{2}(t - \tau) E^{*2}(t) \right| + \left| E^{2}(t - \tau) \right|^{2} + 2E^{2}(t - \tau) E^{*}(t) E^{*}(t - \tau) + \left. 2E(t) E(t - \tau) E^{*2}(t) + 2E(t) E(t - \tau) E^{*2}(t - \tau) + 4 \left| E(t) \right|^{2} \left| E(t - \tau) \right|^{2} \right\} dt$$

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

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 ω in delay)

(oscillates at ω in delay)

$$+ \int_{-\infty}^{\infty} E^{2}(t)E^{2*}(t-\tau) dt + c.c.$$

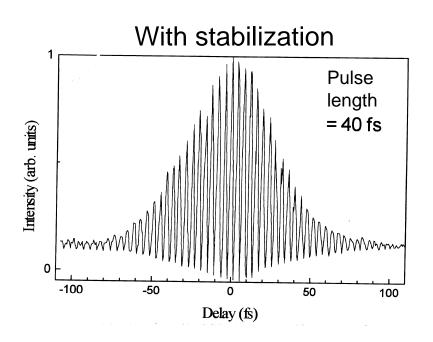
+ $\int_{-\infty}^{\infty} E^2(t)E^{2*}(t-\tau) dt + c.c.$ Interferogram of the second harmonic; equivalent to the spectrum of the SH (oscillates at 2ω in delay)

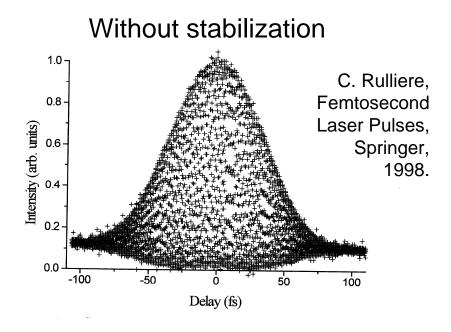
The interferometric autocorrelation simply combines several measures of the pulse into one (admittedly complex) trace. Conveniently, however, they occur with different oscillation frequencies: θ , ω , and 2ω . 34

Interferometric Autocorrelation and Stabilization

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

Interferometric Autocorrelation Traces for a Flat-phase Gaussian pulse:

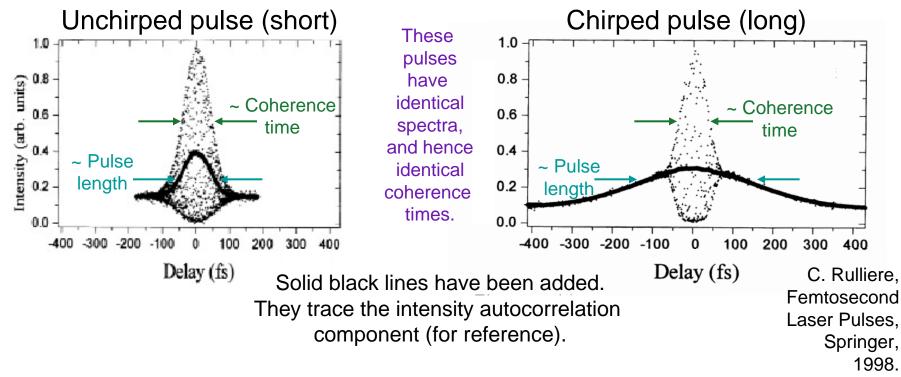




Fortunately, it's not always necessary to resolve the fringes.

Interferometric Autocorrelation: Examples

The extent of the fringes (at ω and 2ω) indicates the approximate width of the interferogram, which is the coherence time. If it's the same as the width of 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. 36

Interferometric Autocorrelation: Practical Details

A good check on the interferometric autocorrelation is that it should be symmetrical, and the peak-to-background ratio should be 8.

This device is difficult to align; there are five very sensitive degrees of freedom in aligning two collinear pulses.

Dispersion in each arm must be the same, so it is necessary to insert a compensator plate in one arm.

The typical ultrashort pulse is still many wavelengths long. So many fringes must typically be measured: data sets are large, and scans are slow.

Like the intensity autocorrelation, it must be curve-fit to an assumed pulse shape.

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).

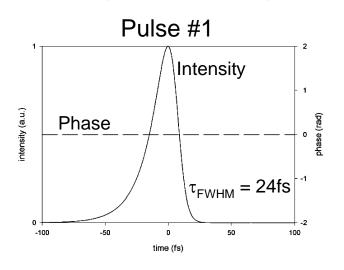
But the required iterative algorithm rarely converges.

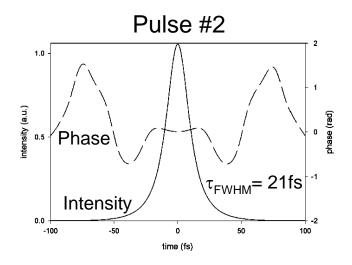
The fact is that the interferometric autocorrelation yields little more information than the autocorrelation and spectrum.

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

Pulses with Very Similar Interferometric Autocorrelations

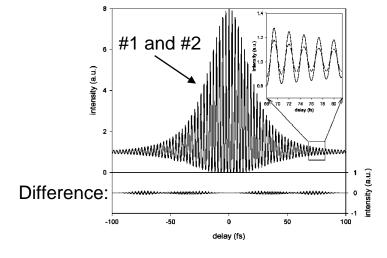
Without trying to find ambiguities, we can just try Pulses #1 and #2:





Interferometric Autocorrelations for Pulses #1 and #2

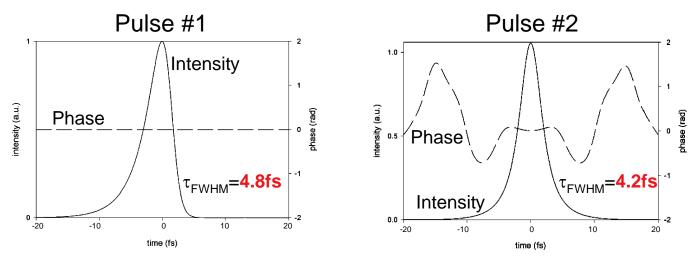
Despite the very different pulses, these traces are nearly identical!



Chung and Weiner, IEEE JSTQE, 2001.

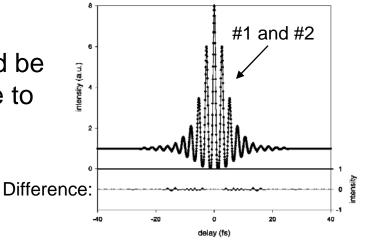
Pulses with Very Similar Interferometric Autocorrelations

It's even harder to distinguish the traces when the pulses are shorter, and there are fewer fringes. Consider Pulses #1 and #2, but 1/5 as long:



Interferometric Autocorrelations for Shorter Pulses #1 and #2

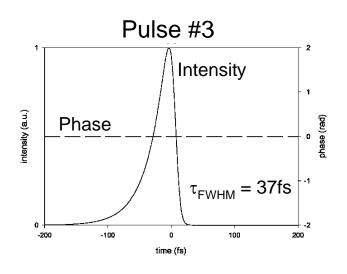
In practice, it would be virtually impossible to distinguish these.

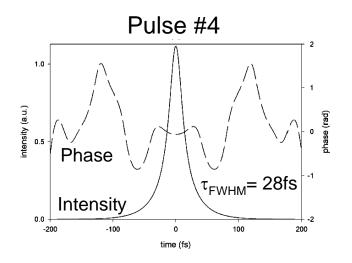


Chung and Weiner, IEEE JSTQE, 2001.

More Pulses with Similar Interferometric Autocorrelations

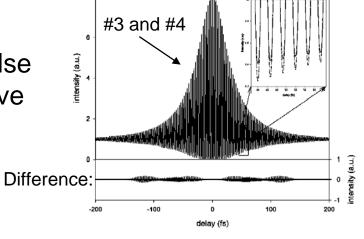
Without trying to find ambiguities, we can try Pulses #3 and #4:





Interferometric Autocorrelations for Pulses #3 and #4

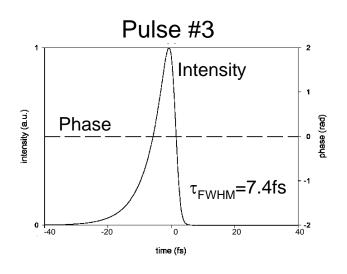
Despite very different pulse lengths, these pulses have nearly identical IAs.

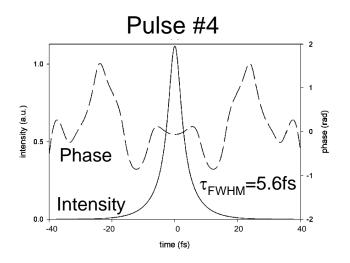


Chung and Weiner, IEEE JSTQE, 2001.

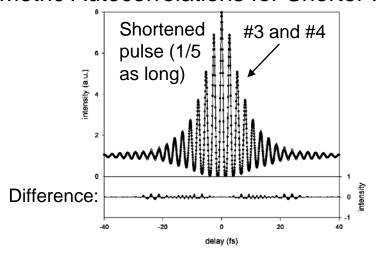
More Pulses with Similar Interferometric Autocorrelations

Shortening Pulses #3 and #4 also yields very similar IA traces:





Interferometric Autocorrelations for Shorter Pulses #3 and #4



Chung and Weiner, IEEE JSTQE, 2001.

It is dangerous to derive a pulse length from the IA.