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Temporal Characterization of Ultrafast Laser Pulses

Francesca Calegari

Center For Free Electron Laser Science (CFEL) Deutsches Elektronen-Synchrotron (DESY) Hamburg Universität francesca.calegari@desy.de



Time scale in matter



A journey in time...

10⁻¹⁸ s **Electron dynamics** Nuclear dynamics 10⁻¹² - 10⁻¹⁵ s 10⁻⁶ - 10⁻⁹ s Protein folding Heart beat 1 s

Atomic unit of time: 24 attoseconds

Electron orbit time around the nucleus: 150 attoseconds

Attosecond Science for following and controlling electron dynamics in matter!

Time resolved measurement

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

We need a strobe light pulse short enough!

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

How can we measure the shortest events?





Time resolved measurement



G. Cerullo et al., Photochem. Photobiol. Sci. 6, 135 (2007)

PROBE: a second delayed laser pulse probe the dynamics

How fast can we measure?





With **pico/femto**second laser pulses: real-time observation of nuclear dynamics & breakage of a chemical bond

With **atto**second laser pulses: real-time observation of electron dynamics

F. Krausz Phys. Scr. 91, 063011 (2016)

Summary of the lecture

- Pulse characterization
- Intensity autocorrelation
- Interferometric Autocorrelation (IAC)
- Frequency Resolved Optical Gating (FROG)
- Spectral Phase Interferometry for Direct Electric-field Reconstruction (SPIDER)
- Attosecond pulse characterization

Ultrafast lasers





- Pulse duration T (fs-ns)
- Pulse energy E (pJ-mJ) •
- Peak power $P_p \approx E/T$ (kW-PW) Repetition rate f_R (Hz-MHz)
- Average power $P=E^{*}f_{R}$ (mW-W) •
- Center wavelength λ_0 (infrared-UV)

Measurement of pulse "physical quantities"



Full characterization of an optical pulse

Electric field of a laser pulse in time domain:

 $E(t) \sim \text{Re} \left\{ I(t)^{1/2} \exp \left[j\omega_0 t - j\phi(t) \right] \right\}$ Intensity
Temporal phase

...& in frequency domain:

 $\tilde{E}(\omega) \sim |(\omega - \omega_0)^{1/2}| \exp[-j\phi(\omega - \omega_0)]$ Intensity $\int Spectral phase$ Can be measured with a spectrometer

Measurement of the spectrum



- Transform-limited pulse can be obtained from the measured spectrum
- Spectral phase is missing!

The spectral phase

$$\widetilde{\mathsf{E}}(\omega) \sim |(\omega - \omega_0)^{1/2} \exp[-j\varphi(\omega - \omega_0)]$$
Intensity Spectral phase

The **instantaneous frequency** (frequency vs time) can be retrieved from the spectral phase



$$\boldsymbol{\omega}(t) = \boldsymbol{\omega}_0 - d\boldsymbol{\phi} / dt$$



The spectral phase

$$\stackrel{\sim}{\mathsf{E}}(\omega) \sim |(\omega - \omega_0)^{1/2}| \exp\left[-j\phi(\omega - \omega_0)\right]$$
Intensity Spectral phase

The group delay can be retrieved from the spectral phase



$$\tau_g(\omega) = d\varphi / d\omega$$

The group delay vs. frequency is approximately the inverse of the instantaneous frequency vs. time

We should be able to measure, pulses with arbitrarily complex phases and frequencies vs. time!

Measurement in the time domain

Is there a device to measure the duration of the pulse?

Photo-detectors: photodiodes & photomultipliers

- Photo-detectors are devices that emit electrons in response to photons
- The detector output voltage is proportional to the pulse energy



Photo-detectors measure the time integral of the pulse intensity:

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

The detector response is too slow for ultrafast pulses (typically nanoseconds)!

Measurement in the time domain



Fast photo-detectors allow the laser pulse train to be observed on the oscilloscope:



Measurement in the time domain

Photo-detectors tell us only a very little about the pulse



The best way to temporally characterize a laser pulse is to use the pulse itself (or a reference pulse)

All-optical methods!

Field autocorrelation



Field autocorrelation

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

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

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

\propto Pulse energy



Field autocorrelation (interferogram)

- Measuring the interferogram is equivalent to measuring the spectrum
- Field autocorrelation measurement gives no information about the spectral phase
- Field autocorrelation measurement cannot distinguish a transform-limited pulse from a longer chirped pulse with the same bandwidth

Intensity autocorrelation



Intensity Autocorrelation:

- create a delayed replica of the pulse
- cross beams in an second-harmonic generation (SHG) crystal
- vary the delay between the two pulses
- measure the second-harmonic (SH) pulse energy vs. delay

Intensity autocorrelation



$$E_{SH}(t,\tau) \propto E(t)E(t-\tau)$$

$$\implies I_{AC}(\tau) \propto \int_{-\infty}^{\infty} \left| E(t) E(t-\tau) \right|^2 dt$$

$$I_{AC}(\tau) \propto \int_{-\infty}^{\infty} I(t)I(t-\tau)dt$$

Intensity autocorrelation: squared pulse

Pulse

Autocorrelation

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

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



Intensity autocorrelation: gaussian pulse

Pulse

Autocorrelation



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

Intensity autocorrelation: sech² pulse



Intensity autocorrelation: Lorentzian pulse

Pulse

Autocorrelation

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

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



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

Intensity autocorrelation:

• It is always symmetric, and assumes its maximum value at $\tau = 0$.

$$I_{AC}(\tau) \propto \int_{-\infty}^{\infty} I(t)I(t-\tau)dt \qquad I_{AC}(\tau) = I_{AC}(-\tau)$$

- Width of the correlation peak gives information about the pulse width
- Pulse phase information is missing
- To get the pulse duration, it is necessary to assume a pulse shape, and to use the corresponding deconvolution factor
- For short pulses, very thin crystals must be used to guarantee enough phase- matching bandwidth
- The intensity autocorrelation is **not** sufficient to determine the pulse intensity profile

Autocorrelations of more complex intensities

Autocorrelations nearly always have considerably less structure than the corresponding intensity



An autocorrelation typically corresponds to many different intensities \longrightarrow the autocorrelation does not uniquely determine the intensity

Autocorrelations of more complex intensities

These complex intensities have nearly Gaussian autocorrelations



Autocorrelation has many nontrivial ambiguities!

Geometrical distortions in autocorrelation

When crossing beams at an angle, the delay varies across the beam



This effect causes a range of delays to occur at a given time and could cause geometrical smearing with a broadening of the autocorrelation width

Single-shot autocorrelation

Crossing beams at an angle also maps delay onto transverse position



Large beams and a large angle allows to achieve the desired range of delays in a single-shot. No-need for delay scan! Single-shot SHG AC has no geometrical smearing



An alternative approach is to use a collinear beam geometry, and allow the autocorrelator signal light to interfere with the SHG from each individual beam

$$IA^{(2)}(\tau) \equiv \int_{-\infty}^{\infty} \left| \left[E(t) - E(t - \tau) \right]^2 \right|^2 dt$$
$$IA^{(2)}(\tau) \equiv \int_{-\infty}^{\infty} \left| \frac{E^2(t) + E^2(t - \tau)}{1 - 2E(t)E(t - \tau)} \right|^2 dt$$
New terms Autocorrelation term

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

$$\begin{split} IA^{(2)}(\tau) &= \int_{-\infty}^{\infty} \Big\{ \left| E^{2}(t) \right|^{2} + E^{2}(t) E^{*2}(t-\tau) - 2E^{2}(t) E^{*}(t) E^{*}(t-\tau) + \\ & E^{2}(t-\tau) E^{*2}(t) + \left| E^{2}(t-\tau) \right|^{2} - 2E^{2}(t-\tau) E^{*}(t) E^{*}(t-\tau) + \\ & -2E(t)E(t-\tau)E^{*2}(t) - 2E(t)E(t-\tau)E^{*2}(t-\tau) + 4 \left| E(t) \right|^{2} \left| E(t-\tau) \right|^{2} \Big\} dt \\ &= \int_{-\infty}^{\infty} \Big\{ I^{2}(t) + E^{2}(t)E^{*2}(t-\tau) - 2I(t)E(t)E^{*}(t-\tau) + \\ & E^{2}(t-\tau) E^{*2}(t) + I^{2}(t-\tau) - 2I(t-\tau)E^{*}(t)E(t-\tau) + \\ & -2I(t)E(t-\tau)E^{*}(t) - 2I(t-\tau)E(t)E^{*}(t-\tau) + 4I(t)I(t-\tau) \Big\} dt \end{split}$$

Where: $I(t) \equiv |E(t)|^2$

Interferometric autocorrelation From the math we can extract 4 terms:

$$= \int_{-\infty}^{\infty} I^{2}(t) + I^{2}(t-\tau) dt = I_{back} \quad \text{Background}$$

$$+ 4 \int_{-\infty}^{\infty} I(t)I(t-\tau) dt = I_{int} \quad \text{Intensity} \\ \text{autocorrelation}$$

$$- 2 \int_{-\infty}^{\infty} [I(t) + I(t-\tau)]E(t)E^{*}(t-\tau) dt + c.c = I_{\omega} \quad \text{Interferogram} \\ \text{of } E(t), \\ \text{oscillating at } \omega$$

$$+ \int_{-\infty}^{\infty} E^{2}(t)E^{2*}(t-\tau) dt + c.c. = I_{2\omega} \quad \text{Interferogram of the} \\ \text{SH oscillating at } 2\omega$$

 $IA^{(2)}(\tau = 0) = 8$ $IA^{(2)}(\tau \rightarrow \infty) = 1$





Interferometric autocorrelation also have ambiguities

- It is always symmetric 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.
- Using optical spectrum and background-free intensity autocorrelator can determine the presence or absence of strong chirp. The interferometric autocorrelation serves as a clear visual indication of moderate to large chirp.
- It is difficult to distinguish between different pulse shapes and, especially, different phases from interferometric autocorrelations.
- Like the intensity autocorrelation, it must be curve-fit to an assumed pulse shape and so should only be used for rough estimates.

How to measure both pulse intensity profile and phase?

- A pulse can be represented by two arrays of data with length N, one for the amplitude/intensity and the other for the phase. Totally we have 2N degrees of freedom (corresponding to the real and imaginary parts for the electric field)
- Intensity autocorrelator provides only one array of data with length N.
 Optical spectrum measurement can provide another array of data with length N. However some information, especially about phase, is missing from both measurements
- Need to have more data, providing enough redundancy to recover the both the amplitude and phase

How about measuring the spectrum of the autocorrelation pulse at each delay? NxN data points

How to measure both pulse intensity profile and phase?

Frequency vs Time \rightarrow SPECTROGRAM A spectrogram can be seen as a musical score!



How about measuring the spectrum of the autocorrelation pulse at each delay? NxN data points

The spectrogram

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

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

where g(t-t) is a variable-delay gate function and t is the delay

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

The spectrogram is a function of ω and t

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

The spectrogram

We must compute the spectrum of the product: $E_{sig}(t,\tau) = E(t) g(t-\tau)$



The spectrogram contains the color and intensity of E(t) at each time t

The spectrogram





The spectrogram yields the color and intensity of E(t) at the time, t.

Frequency-Resolved Optical Gating (FROG): SHG-FROG

Background-free intensity autocorrelator + optical spectrum analyzer



FROG provides N X N data points. With an iterative algorithm it is possible to retrieve both the amplitude and phase of the measured optical pulse.

Frequency-Resolved Optical Gating (FROG): SHG-FROG

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



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E_{sig}(t,\tau) = E(t) g(t-\tau)g(t-\tau) = E(t-\tau)
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Frequency-Resolved Optical Gating (FROG): SHG-FROG

SHG FROG traces are symmetrical with respect to delay



Frequency

SHG FROG has an ambiguity in the direction of time, but it can be removed

Generalized projections algorithm

E(t) can be fully retrieved from the measured spectrogram by applying iterative reconstruction algorithms



FROG algorithm



SHG FROG measurement of a 4.5-fs pulse



Agreement between the experimental and reconstructed FROG traces provides a nice check on the measurement.

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

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



FROG using arbitrary nonlinear-optical interactions

FROG is simply a frequency-resolved nonlinear-optical signal that's a function of time and delay (or another variable).

Pulse retrieval remains equivalent to the 2D phase-retrieval problem.

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

Spectral interferometry

Measure the spectrum of the sum of a known and unknown pulse Retrieve the unknown pulse from the spectral fringes



 $S_{SI}(\omega) = S_{ref}(\omega) + S_{unk}(\omega) + 2\sqrt{S_{ref}(\omega)}\sqrt{S_{unk}(\omega)}\cos[\varphi_{unk}(\omega) - \varphi_{ref}(\omega) + \omega T]$

Spectral interferometry



This retrieval algorithm is quick, direct, and reliable

A reference pulse is usually not available!

If we perform spectral interferometry between a pulse and itself, the spectral phase cancels out. Perfect sinusoidal fringes always occur:

$$S_{SI}(\omega) = S_{unk}(\omega) + S_{unk}(\omega) + 2\sqrt{S_{unk}(\omega)}\sqrt{S_{unk}(\omega)}\cos[\varphi_{unk}(\omega) - \varphi_{unk}(\omega) + \omega T]$$

However if we frequency shift one pulse replica compared to the other:

$$S_{SI}(\omega) = S(\omega) + S(\omega + \delta\omega) + 2\sqrt{S(\omega)}\sqrt{S(\omega + \delta\omega)}\cos[\varphi(\omega + \delta\omega) - \varphi(\omega) + \omega T]$$

 $\phi_{SPIDER} = \varphi(\omega + \delta\omega) - \varphi(\omega) + \omega T = \delta\omega \frac{d\varphi}{d\omega} + \omega T \quad \text{Time delay}$

This measures the derivative of the spectral phase (the group delay)



1) Make a very chirped pulse

2) Create two replicas of the pulse

3) Frequency shift the 2 replicas by SFG with the broadband pulse and perform SI



ZAP-SPIDER



Extraction of the spectral phase







Many more methods exist...

- 2DSI: Two Dimensional Spectral Shearing Interferometer
- STRUT: Spectrally and Temporally Resolved Upconversion Technique
- TURTLE: Tomographic Ultrafast Retrieval of Transverse Light *E* fields Reconstruction
- TADPOLE: Temporal Analysis by Dispersing a Pair Of Light E-fields

Use a second gas jet and photoionization to produce a crosscorrelation with the input pulse



Energy-resolve the photoelectrons to generate a spectrally resolved cross-correlation. This generates a type of XFROG trace, which yields the intensity and phase of the attosecond pulse.

As the relative delay between the XUV pulse and the 800nm field varies, the **added energy** (ΔW) of the emitted **electron packet** will vary.





time

$$S(\vec{v},\tau) = \left| \int \exp(i\Phi(t)) \vec{d}_{\vec{p}-\vec{A}(t)} \cdot \vec{E}_{XUV}(t-\tau) \exp(i(W+I_p)t) dt \right|^2$$

$$\Phi(t) = -\int_t^{+\infty} dt' \left[\vec{v} \cdot \vec{A}(t') + \vec{A}^2(t') / 2 \right] =$$

$$= -\int_t^{+\infty} dt' U_p(t') + \frac{\sqrt{8WU_p}}{\omega} \cos\theta \cos\omega t - (U_p / 2\omega) \sin 2\omega t$$

In FROG CRAB the gate function is a modulation of the phase of the electronic wave packet: phase gate!

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

$$S(\vec{v},\tau) = \left| \int \exp(i\Phi(t)) \vec{d}_{\vec{p}-\vec{A}(t)} \cdot \vec{E}_{XUV}(t-\tau) \exp(i(W+I_p)t) dt \right|^2$$

$$\Phi(t) = -\int_t^{+\infty} dt' \left[\vec{v} \cdot \vec{A}(t') + \vec{A}^2(t') / 2 \right] =$$

$$= -\int_t^{+\infty} dt' U_p(t') + \frac{\sqrt{8WU_p}}{\omega} \cos\theta \cos\omega t - (U_p / 2\omega) \sin 2\omega t$$

 $S(\mathbf{v}, \tau) = I_{FROG-CRAB}$ spectrogram $\Phi(t) = phase of electron wavepacket modulated by the external IR field$ $<math>\mathbf{A}(t) = vector potential of the IR pulse$ W = kinetic energy of the ejected electron $\omega = frequency of the IR field$ $U_p = ponderomotive potential of the IR pulse$ $\theta = angle between the electron velocity <math>\mathbf{v}$ and the vector potential \mathbf{A}

FROG CRAB





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Lecture slides from R. Trebino on the following website: http://frog.gatech.edu/lectures.html