The Birth of Ultrafast Technology

Bet: Do all four hooves of a galloping horse ever simultaneously leave the ground?

Leland Stanford  
Eadweard Muybridge  
Palo Alto, CA  1872

Time Resolution: 1/60th of a second
If you think you know fast, think again.

Ultrashort laser pulses are the shortest events ever created.
Ultrafast Optics vs. Electronics

No one expects electronics to ever catch up.
Time Resolution: a few microseconds

“How to Make Apple sauce at MIT”
1964

“Splash on a Glass”
Curtis Hurley
Junior High School student
1996
The Metric System

We’ll need to really know the metric system because the pulses are incredibly short and the powers and intensities can be incredibly high.

Prefixes:

<table>
<thead>
<tr>
<th>Small</th>
<th>Big</th>
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<tbody>
<tr>
<td>Milli (m)</td>
<td>Kilo (k)</td>
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<tr>
<td>Micro (µ)</td>
<td>Mega (M)</td>
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<tr>
<td>Nano (n)</td>
<td>Giga (G)</td>
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<tr>
<td>Pico (p)</td>
<td>Tera (T)</td>
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<tr>
<td>Femto (f)</td>
<td>Peta (P)</td>
</tr>
<tr>
<td>Atto (a)</td>
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| 10^{-3}     | 10^{+3}    |
| 10^{-6}     | 10^{+6}    |
| 10^{-9}     | 10^{+9}    |
| 10^{-12}    | 10^{+12}   |
| 10^{-15}    | 10^{+15}   |
| 10^{-18}    |            |
**Timescales**

- **10 fs light pulse**
- **Computer clock cycle**
- **Camera flash**
- **1 minute**
- **One month**
- **Age of pyramids**
- **Age of universe**

Time (seconds):

\[
\begin{align*}
10^{-14} & \quad 10^{-9} & \quad 10^{-4} & \quad 10^1 & \quad 10^6 & \quad 10^{11} & \quad 10^{16}
\end{align*}
\]

10 fs is to 1 minute as 1 minute is to the age of the universe.

Alternatively, 10 fs is to 1 sec as 5 cents is to the US national debt.
Ultrafast Lasers

A 4.5-fs pulse...

Current record: 4.0 fsec
Baltuska, et al. 2001

Reports of attosecond pulses, too!

Ultrafast Ti:sapphire laser
The Shortest Pulses at Different Wavelengths

![Graph showing pulse duration vs. frequency for different wavelengths.](image)
Short Pulses at Short Wavelengths

90 degree relativistic Thompson scattering
Lawrence Berkeley National Laboratory
Ultrafast set-ups can be very sophisticated.
The Highest Intensities Imaginable

0.2 TW = 200,000,000,000 watts!

1 kHz “Chirped-Pulse Amplification (CPA)” system at the University of Colorado (Murnane and Kapteyn)
Even Higher Intensities!

National Ignition Facility (under construction)

192 shaped pulses
1.8 MJ total energy
Continuous vs. ultrashort pulses of light

A constant and a delta-function are a Fourier-Transform pair.
Long vs. short pulses of light

The uncertainty principle says that the product of the temporal and spectral pulse widths is greater than ~1. 
Ultrafast laser media

Solid-state laser media have broad bandwidths and are convenient.
A generic ultrashort-pulse laser

A generic ultrafast laser has a broadband gain medium, a pulse-shortening device, and two or more mirrors:

Pulse-shortening devices include:
- Saturable absorbers
- Phase modulators
- Dispersion compensators
- Optical-Kerr media
One way to make short pulses: the saturable absorber

Like a sponge, an absorbing medium can only absorb so much. High-intensity spikes burn through; low-intensity light is absorbed.
Generating short pulses = “mode-locking”

Locking the phases of the laser frequencies yields an ultrashort pulse.

Random phases of all laser modes

Locked phases of all laser modes

Irradiance vs. time

Random phases

Light bulb

Locked phases

Ultrashort pulse!!
Group velocity dispersion broadens ultrashort laser pulses

Different frequencies travel at different group velocities in materials, causing pulses to expand to highly "chirped" (frequency-swept) pulses.

![Diagram showing input ultrashort pulse, any medium, and chirped output not-so-ultrashort pulse]

Longer wavelengths almost always travel faster than shorter ones.
The Linearly Chirped Pulse

Group velocity dispersion produces a pulse whose frequency varies in time.

This pulse increases its frequency linearly in time (from red to blue).

In analogy to bird sounds, this pulse is called a "chirped" pulse.
Pulse Compressor

This device has negative group-velocity dispersion and hence can compensate for propagation through materials (i.e., for positive chirp).

The longer wavelengths traverse more glass.

It’s routine to stretch and then compress ultrashort pulses by factors of >1000.
Ultrafast optics is nonlinear optics.

At high intensities, nonlinear-optical effects occur.

All mode-locking techniques are nonlinear-optical.

Creating new colors of laser light requires nonlinear optics.

Second-harmonic-generation of infrared light yields this beautiful display of intense green light.
Continuum Generation

Continuum Generation: focusing a femtosecond pulse into a clear medium turns the pulse white.

A clear medium, such as water, quartz, or ethylene glycol

Generally, small-scale self-focusing occurs, causing the beam to break up into filaments.

This process can be as much as 100% efficient, with wavelengths ranging from the mid-UV to the mid-IR.

Recently developed techniques involving optical fibers, hollow fibers, and microstructure fibers produce very broadband continuum, over 500 THz (1000 nm) in spectral width!
The continuum from microstructure optical fiber is ultrabroadband.

This continuum was created using $nJ$ ultrashort pulses.


Cross section of the microstructure fiber.

The spectrum extends from $\sim 400$ to $\sim 1500$ nm and is relatively flat (when averaged over time).
The Dilemma

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

To study a soap bubble popping, 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.

So, now, how do you measure the *shortest* event?
Using the pulse to measure itself: The Intensity Autocorrelator

Crossing beams in a nonlinear-optical crystal, varying the delay between them, and measuring the signal pulse energy vs. delay, yields the Intensity Autocorrelation, $A^{(2)}(\tau)$.

The signal field is $E(t)E(t-\tau)$. So the signal intensity is $I(t)I(t-\tau)$.

The Intensity Autocorrelation:

$$A^{(2)}(\tau) \equiv \int_{-\infty}^{\infty} I(t)I(t-\tau)dt$$
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.

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

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

“Polarization Gate” Geometry

Pulse to be measured

Beam splitter

Variable delay, \( t \)

E\((t-\tau)\)

E\(t\)

45° polarization rotation

Nonlinear medium

Camera

Spec-trometer

\[ E_{\text{sig}}(t, \tau) = E(t) \left| E(t-\tau) \right|^2 \]
The FROG trace visually displays the frequency vs. time.
One of the shortest events ever created!

FROG traces

A 4.5 fs pulse!
FROG Measurement of the Ultrabroadband Continuum

Ultrabroadband continuum was created by propagating 1-nJ, 800-nm, 30-fs pulses through 16 cm of Lucent microstructure fiber.

Spectrogram

This pulse has a time-bandwidth product of ~4000, and is the most complex ultrashort pulse ever measured.
Spatio-temporal characteristics of ultrashort laser pulses

Ultrashort laser pulses are broadband, so the tendency of different colors to propagate differently can cause the pulse to have **spatio-temporal distortions**.

Beam divergence angle $\theta$ depends on $\lambda$: $\theta = \frac{2\lambda}{\pi w}$, where $w = \text{beam spot size}$

So, if $\lambda$ ranges from 500 nm to 1000 nm, $\theta$ varies by a factor of 2. And, in the far-field, the beam spot size and intensity will vary significantly with color!
Dispersion causes pulse fronts to tilt.

Phase fronts are perpendicular to the direction of propagation.

Because the group velocity is usually less than the phase velocity, pulse fronts tilt when light traverses a prism. With gratings, it’s a simple light-travel-distance issue.

This effect can be useful (for measuring pulses), but it can also be a pain.
We can shape ultrashort pulses.

This usually occurs in the frequency domain.
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.
Ultrafast Laser Spectroscopy: Why?

Most events that occur in atoms and molecules happen on fs and ps time scales. The length scales are very small, so very little time is required for the relevant motion.

Fluorescence occurs on a ns time scale, but competing non-radiative processes only speed things up because relaxation rates add:

\[
\frac{1}{\tau_{ex}} = \frac{1}{\tau_{fl}} + \frac{1}{\tau_{nr}}
\]

Biologically important processes utilize excitation energy for purposes other than fluorescence and hence must be very fast.

Collisions in room-temperature liquids occur on a few-fs time scale, so nearly all processes in liquids are ultrafast.

Semiconductor processes of technological interest are necessarily ultrafast or we wouldn’t be interested.
The simplest ultrafast spectroscopy method is the Excite-Probe Technique.

This involves exciting the sample with one pulse, probing it with another a variable delay later, and measuring the change in the transmitted probe pulse average power vs. delay:

\[ E_{ex}(t-\tau) \]

Excite pulse

\[ E_{pr}(t) \]

Probe pulse

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

Sample medium

Detector

Variable delay, \( \tau \)

The excite and probe pulses can be different colors. This technique is also called the “Pump-Probe” Technique.
Ultrafast Excite-Probe Measurements in DNA

DNA bases undergo photo-oxidative damage, which can yield mutations. Understanding the photo-physics of these important molecules may help to understand this process.

Transient absorption at 600 nm of protonated guanosine in acidic (pH 2) and basic (pH 11) aqueous solution.
Beyond ultrafast spectroscopy: controlling chemical reactions with ultrashort pulses

You can excite a chemical bond with the right wavelength, but the energy redistributes all around the molecule rapidly ("IVR").

But exciting with an intense, shaped ultrashort pulse can control the molecule’s vibrations and produce the desired products.
Ultrashort in time is also ultrashort in space

Novel imaging techniques yield ~1-µm resolution, emphasizing edges of objects. They include optical coherence tomography and multi-photon imaging.

Example for application:

2-photon microscopy of pollen grains using an ultrashort pulse