Single-cycle and attosecond light pulses

Bandwidth, bandwidth, bandwidth…

UV wavelength…

Possible routes:
  - Raman scattering
  - High-harmonic generation

Measurement of attosecond pulses

Sources:
The physics of attosecond light pulses, Pierre Agostini and Louis F DiMauro
Chang Hee Nam and Kyung Taec Kim
Reinhard Kienberger\textsuperscript{1}, M. Hentschel\textsuperscript{1}, M. Drescher\textsuperscript{1,2}, G. Reider\textsuperscript{1}, Ch. Spielmann\textsuperscript{1}, Ferenc Krausz\textsuperscript{1}
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Why try to make attosecond pulses?

Faster is better. And yes, some things really are that fast.

The Bohr orbit time in hydrogen: 152 attoseconds

Molecular vibrations can also be very fast. The vibrational oscillation period of the H₂ molecule: ~ 7 fs
X-ray Pump-Probe Spectroscopy

With sub-fs x-ray pulses, we could trace inner shell relaxation processes.

Sub-fs X-ray pump

Sub-fs X-ray probe
Bandwidth

The uncertainty principle requires that $\Delta \tau \Delta \nu > 1$.

So for a sub-fs pulse, $\Delta \nu > 10^{15}$ Hz. This is a lot!
Imagine a **half-cycle pulse**. Its **electric field** looks like this:

![Electric field graph](image1)

As it propagates to the far field, such a pulse evolves into a **single-cycle pulse**:

![Electric field graph](image2)

In the far field, the shortest possible pulse of a given wavelength is one cycle long.

**Single-cycle pulses**

Because this pulse field has nonzero area, it has an \( \omega = 0 \) **component**—which has infinite wavelength! And this wavelength diffracts away immediately:

\[
z_R = \frac{\pi w^2}{\lambda} \to 0
\]
Attosecond pulses are short-wavelength pulses.

A single-cycle red pulse:

Compressing a single-cycle red pulse:

A single-cycle 800-nm pulse has a period of 2.7 fs.

To achieve a period of 1 fs requires a wavelength of 300 nm.

If you compress a single-cycle pulse of one wavelength, you necessarily reduce its wavelength!
Two methods for generating many equally spaced modes with $10^{15}$ Hz bandwidth

1. Cascaded Raman scattering (does not start with short pulses)

2. High-harmonic generation (starts with short optical pulses)

But do these methods phase-lock the modes?
Raman effect: the inelastic scattering of photons from molecules

The Raman process is a $\chi^{(3)}$ nonlinearity, so it is weak.

But it can be stimulated by illuminating the molecule with two lasers, spaced in frequency by $\nu_t$. 

![Diagram of Raman effect]
Raman scattering: broadband output

Input two frequencies nearly resonant with a Raman resonance.

At high intensity, the process cascades many times.

Raman processes can cascade many times, yielding a series of equally spaced modes!

S. E. Harris and A. V. Sokolov PRL 81, 2894
Cascaded Raman generation

A. V. Sokolov et al.
PRL 85, 85 562

\[ \Delta \omega_{ba} = 2994 \text{ cm}^{-1} \]

This can be done with high-power \textit{nanosecond} laser pulses!
Experimental demonstration of cascaded Raman scattering

75,000 cm$^{-1}$ (2.3 x 10$^{15}$ Hz) of bandwidth has been created!

A. V. Sokolov et al. PRL 85, 562
Numerical simulation of Raman scattering in $D_2$

Propagation distance

$z$ (cm)

Harris and Sokolov. PRL 81, 2894
Is the light produced using cascaded Raman scattering an attosecond pulse?

Unfortunately, no one has reliably measured the spectral phase of this light, so we don’t know its pulse length…

And if you haven’t measured it, you haven’t made it!

The measurements are suggestive, but not conclusive.
Is the light produced using cascaded Raman scattering an attosecond pulse?

- Generate 7 sidebands using the cascaded Raman process
- Vary their relative phases using a liquid crystal
- Optimize the generation of 4-wave-mixing in a Xe cell to obtain the shortest pulse

Two methods for generating many equally spaced modes with $10^{15}$ Hz bandwidth

1. Cascaded Raman scattering (does not start with short pulses)
2. High-harmonic generation (starts with short optical pulses)

In order to understand harmonic generation, we need to characterize the pulse train being used to generate them.
High Harmonic Generation in a gas

HHG produces equally spaced harmonics out to as much as the 300th harmonic, potentially as short as 10 attoseconds!
How to explain high harmonic generation?

We cannot use a perturbative approach, i.e., the $\chi^{(n)}$ picture.

We must resort to an atomic picture of the dynamics:
1. ionization of an atom
2. acceleration of a free electron
3. impact with the parent ion

If all of the x-ray harmonics are in phase, it could be used to generate the world’s shortest light pulses.

“attosecond physics”
High-harmonic generation has all the features needed for attosecond pulse generation.

- XUV region.
- Equally spaced frequencies and lots of ‘em.
- Overall very broad bandwidth (> $10^{15}$ Hz).
- Spatial coherence.
- Reasonable stability.

The physics seems to imply that the XUV pulse should be really short…

Maybe HHG just naturally produces attosecond pulses without even trying…
Are the high harmonics actually in phase?

Using a ~100 fs input pulse, the answer is NO!

Measuring the relative phases of adjacent harmonics:

The resulting intensity vs. time:

P. Antoine et al., PRL 77, 1234 (1996)
The highest and most intense harmonics are generated by the shortest pulses.

Measured XUV spectral intensity from neon

Note the broader harmonic spectrum from the 7-fs pulse.

M. Schnürer et al., PRL 83, 722 (1999)
Ch. Spielmann et al., Science 278, 661 (1997)
Z. Chang et al., PRL 79, 2967 (1997)
Theory says many-fs input pulses yield many-fs XUV pulses, but few-fs input pulses could yield attosecond XUV pulses...

In theory, a 5-fs input pulse yields an attosecond XUV pulse...

Theory predicts much more intense XUV for a zero (or $\pi$) absolute phase.
HHG and the absolute phase (carrier-offset phase)

HHG is very sensitive to the peak intensity, which is higher for a 0°-absolute phase (cos) than for 90°-absolute phase (sin).

\[ \mathcal{E}(t) = E(t) \cos(\omega_L t) \]

\[ \mathcal{E}(t) = E(t) \sin(\omega_L t) \]

Input pulse instantaneous intensity

Threshold for HHG

Multi cycle pulse

Few cycle pulse
Zero\textsuperscript{th}-order phase: the absolute phase

- The absolute phase is the same in both the time and frequency domains.
  \[ f(t)\exp(i\phi_0) \supseteq F(\omega)\exp(i\phi_0) \]
- An absolute phase of $\pi/2$ will turn a cosine carrier wave into a sine.
- It’s usually irrelevant, unless the pulse is only a cycle or so long.

Notice that the two four-cycle pulses look alike, but the two single-cycle pulses are quite different.
Zero\textsuperscript{th}-order phase: the absolute phase

What do we know about the absolute phase of each pulse in our pulse train?

Here, every pulse in the train is identical – all have zero absolute phase.

Here, the absolute phase increases from pulse to pulse.

This absolute phase is sometimes known as the ‘carrier envelope phase’, since it represents the phase shift between the carrier wave and the peak of the pulse envelope.
To understand this, remember mode-locking

Locking vs. not locking the phases of the laser modes (frequencies)

Note: if all the modes are in phase at a particular time (as shown), then it is guaranteed that they will all come into phase periodically, at the commensurate period of all the modes.

Mode-locked lasers produce trains of equally spaced pulses.
The Shah Function

The Shah function, \( \text{III}(x) \), is an infinitely long train of equally spaced delta-functions.

\[
\text{III}(x) = \sum_{n=-\infty}^{\infty} \delta(x - n)
\]

The symbol \( \text{III} \) is pronounced \textit{shah} after the Cyrillic character \( \mathfrak{w} \), which is said to have been modeled on the Hebrew letter \( \mathfrak{v} \) (shin) which, in turn, may derive from the Egyptian \( \text{𓊨𓊪𓊨} \) a hieroglyph depicting papyrus plants along the Nile.
The Fourier Transform of the Shah Function

\[
F\{III(t)\} = \int_{-\infty}^{\infty} \sum_{m=-\infty}^{\infty} \delta(t - m) \exp(-j\omega t) dt
\]

\[
= \sum_{m=-\infty}^{\infty} \int_{-\infty}^{\infty} \delta(t - m) \exp(-j\omega t) dt
\]

\[
= \sum_{m=-\infty}^{\infty} \exp(-j\omega m)
\]

If \( \omega = 2n\pi \), where \( n \) is an integer, the sum diverges; otherwise, cancellation occurs, and the sum vanishes.

So: \( F\{III(t)\} \propto III(\omega/2\pi) \)

The Fourier transform of the Shah function is another Shah function.
The Shah Function in ultrafast optics

An infinite train of identical pulses (for example, from a mode-locked laser) can be written as a convolution:

\[ E(t) = \Pi(t/T) \ast f(t) \]

\[ = \sum_{m=-\infty}^{\infty} f(t/T - m) \]

where \( f(t) \) is the shape of each pulse and \( T \) is the time between pulses.
The Fourier transform of an infinite train of pulses

An infinite train of identical pulses can be written:

\[ E(t) = \text{III}(t/T) \ast f(t) \]

where \( f(t) \) represents a single pulse and \( T \) is the time between pulses. The Convolution Theorem states that the Fourier Transform of a convolution is the product of the Fourier Transforms. So:

\[ \tilde{E}(\omega) \propto \text{III}(\omega T / 2\pi) F(\omega) \]

The spectrum of a train of pulses consists of a series of equally spaced modes, with an envelope given by the spectrum of a single pulse.
But that analysis ignored the phase of the pulse!

\[ E(t) = \text{III}(t/T) \ast f(t) \]

We assumed that the intensity of the pulse, \(|f(t)|^2\), was invariant from pulse to pulse. But this doesn’t require that the phase is invariant.

Duration is invariant → there is no net GVD in the laser cavity. Therefore:

\[
\frac{\partial k}{\partial \omega} = \frac{1}{v_g} = \text{const.}; \quad \frac{\partial^m k}{\partial \omega^m} = 0 \quad \text{for } m > 1
\]

This is satisfied by

\[ k = k_c + \frac{1}{v_g} \omega \]

which corresponds to

\[ n = c_0 \left( \frac{1}{v_g} + \frac{k_c}{\omega} \right) \]

Slope gives \( v_g \)

That is, the net GVD is zero, but the phase velocity can still exhibit dispersion:

\[ v_p = \frac{\omega}{k} = \frac{v_g}{\frac{v_g k_c}{\omega} + 1} \]

the effective index of the laser cavity
Dispersion of the phase velocity: implications for the spectrum

Recall the formula for the cavity modes inside a laser:

$$\omega_m = \frac{2\pi c}{nL} m \quad (m = \text{integer})$$

If we put in the effective index: $$n = c \left( \frac{1}{v_g} + \frac{k_c}{\omega} \right)$$ and solve for $$\omega$$,

then we immediately find:

$$\omega_m = \frac{2\pi v_g}{L} m - k_0 v_g$$

i.e., the cavity modes are still an equally spaced comb in the frequency domain, but they are shifted by a constant offset.

$$f_m = m f_{rep} - f_0$$
Carrier envelope phase: time and frequency domain

The spectrum of the pulse train consists of a series of delta functions corresponding to the cavity modes.

These modes do not lie at multiples of the laser rep rate. They are shifted by a constant offset.

$$f_m = m f_{\text{rep}} - f_0$$

where

$$f_0 = \frac{\omega_c v_g}{2\pi} \left( \frac{1}{v_g} - \frac{1}{v_p} \right)$$

In the time domain, this is manifested as a non-zero carrier envelope phase $\Delta \phi$. 
Carrier envelope phase: time and frequency domain

The $m^{th}$ laser mode:

$$\omega_m = m\omega_L + 2\pi f_0$$

$f_0 \neq 0$

How to measure the absolute phase

If the spectrum is broad enough…
(spanning at least an octave)

…then comb lines on the low-frequency end of the fundamental spectrum will frequency-double to overlap comb lines on the high-frequency end of the fundamental spectrum.

The heterodyne beat note between these two comb lines will be:

\[ 2\left( \frac{m}{2} f_{rep} + f_0 \right) - (mf_{rep} + f_0) \]

direct measurement of \( f_0 \)!
How to control the absolute phase

Stabilizing the SH spectrum stabilizes the absolute phase at all frequencies.

Apolonski et al. PRL 85 740
How to measure an attosecond pulse?

Attosecond autocorrelation is too difficult.

Using the relatively narrowband 800-nm pump pulse as the gate yields an output electron energy width about half as broad.

The 800-nm beam is easier to work with.

There’s a wider choice of nonlinear effects.

But the 800-nm pulse is a lot longer!

Fortunately, femtosecond measurements using FROG have taught us that this isn’t a problem. All attosecond-pulse methods are versions of XFROG.
As with any long-sought milestone, false claims abound.

“Observation of attosecond light localization in HHG”

N. A. Papadogiannis et al. PRL 83, 4289

These guys basically performed an interferometric autocorrelation measurement, but with HHG as the nonlinear process instead of SHG.
A false claim...

These authors saw peaks, more indicative of the broad spectrum than a short pulse. They also saw unphysical asymmetries, without question due to noise.

Don’t believe everything you read, even if it’s in a peer-reviewed scientific journal!

“Observation of attosecond light localization in HHG” N. A. Papadogiannis, et al. PRL 83, 4289

Harmonic autocorrelation
Energy shift of an electron wave-packet

As we vary the relative delay between the XUV pulse and the 800-nm field, the **added energy** of the emitted electron packet will vary.

There's an angular shift, too, but it's too small to measure.

FROG-CRAB: “frequency-resolved optical gating for complete reconstruction of attosecond bursts”
FROG measurement of an attosecond pulse

Measure photoelectron spectra, rather than light spectra.

F. Krausz and coworkers, Science, 2008
FROG measurement of the current world record shortest atto-second pulse from K. Zhao, et al., *Optics Letters* (2012)

67 attoseconds!
Attosecond pulses: conclusions

This is just the beginning of attosecond science.

In 1980, the shortest pulses were picoseconds long. Femtosecond pulses were only a dream.

Now, not much later, attosecond pulses are real.

Moral: it’s tough to make predictions, especially about the future.