Single-cycle and attosecond light pulses

Bandwidth, bandwidth, bandwidth…

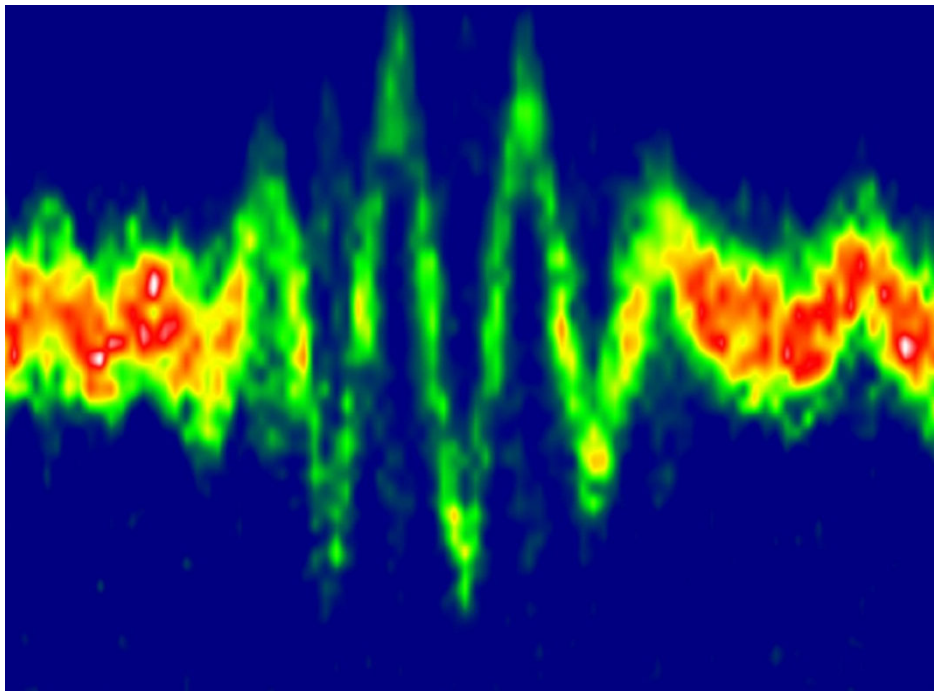
UV wavelength…

Possible routes:

- Raman scattering
- High-harmonic generation

Carrier-envelope phase

Measurement of attosecond pulses

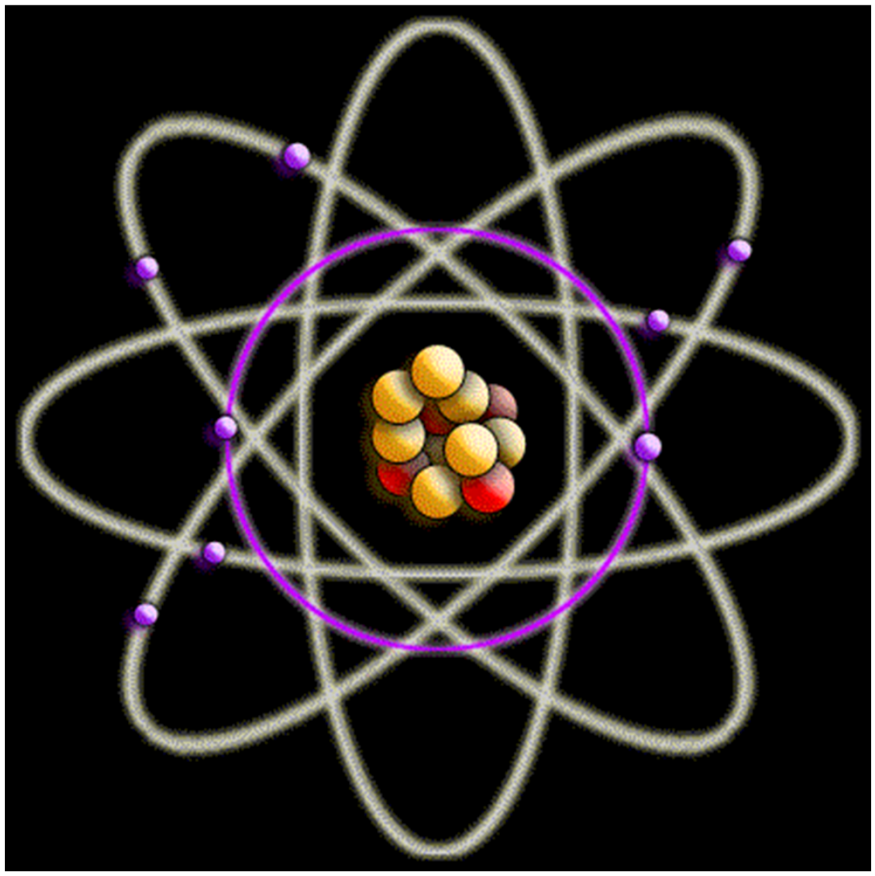


Sources:

The physics of attosecond light pulses, Pierre Agostini and Louis F DiMauro
Rep. Prog. Phys. 67 (2004) 813.
Chang Hee Nam and Kyung Taec Kim

Reinhard Kienberger[1], M. Hentschel[1], M. Drescher[1,2], G. Reider[1], Ch. Spielmann[1], Ferenc Krausz[1]
[1]Institut für Photonik, Technische Universität Wien, AUSTRIA
[2]Fakultät für Physik, Universität Bielefeld, GERMANY
...with input from: Rick Trebino, Ga Tech; Dan Mittleman, Rice; Steve Cundiff, JILA

Why try to make attosecond pulses?



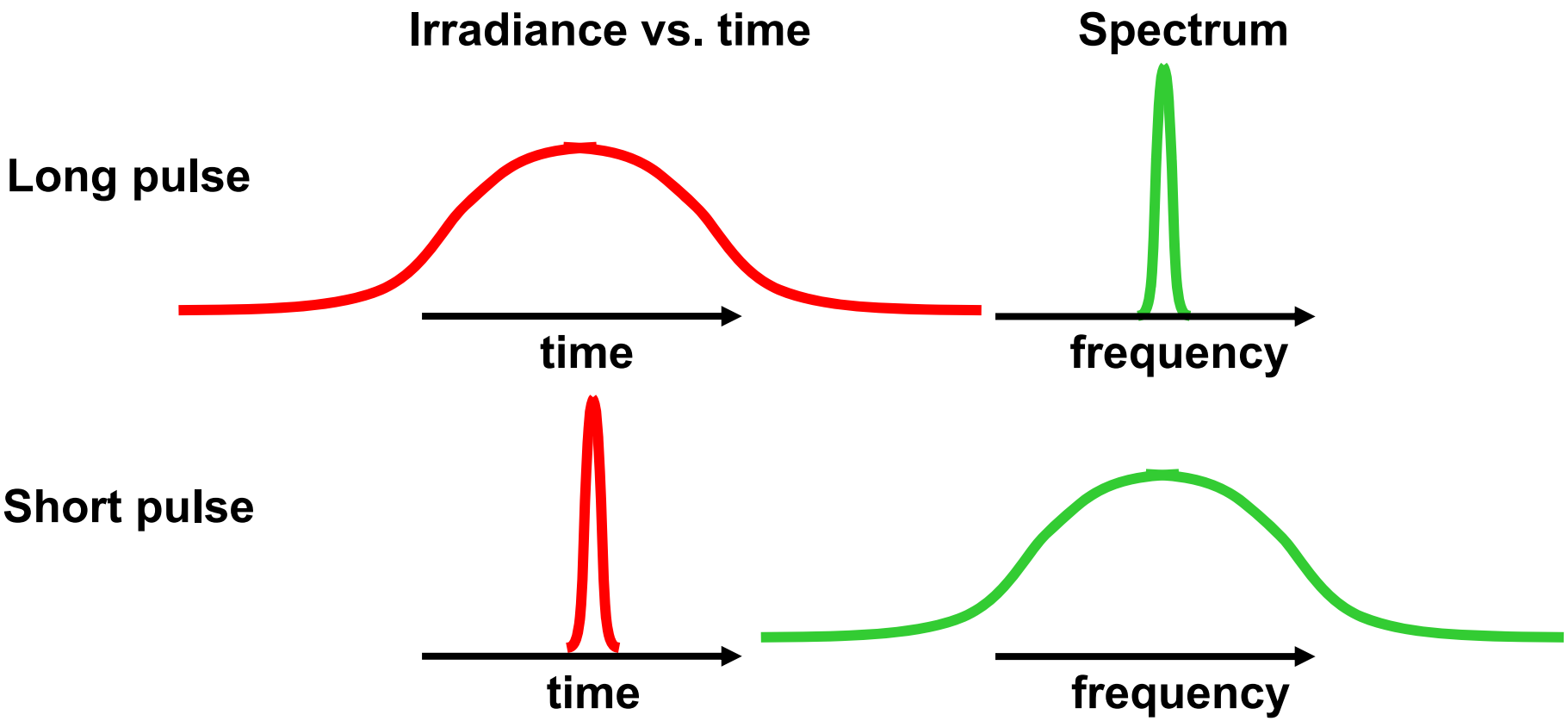
Faster is better. And yes, some things really are that fast.

The Bohr orbit time in hydrogen: 152 attoseconds

Molecular vibrations can also be very fast. The vibrational oscillation period of the H_2 molecule: ~ 7 fs

Bandwidth

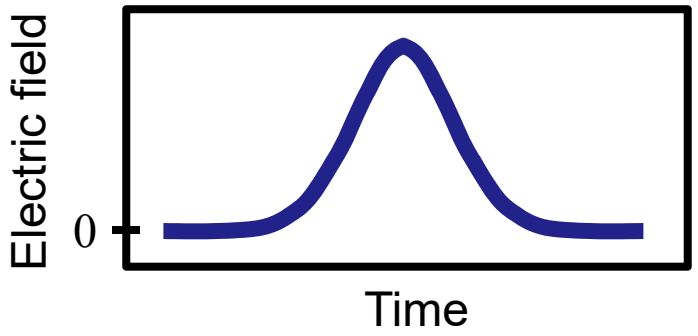
The uncertainty principle requires that $\Delta\tau\ \Delta\nu > 1$.



So for a sub-fs pulse, $\Delta\nu > 10^{15}$ Hz. This is a lot!

Single-cycle pulses

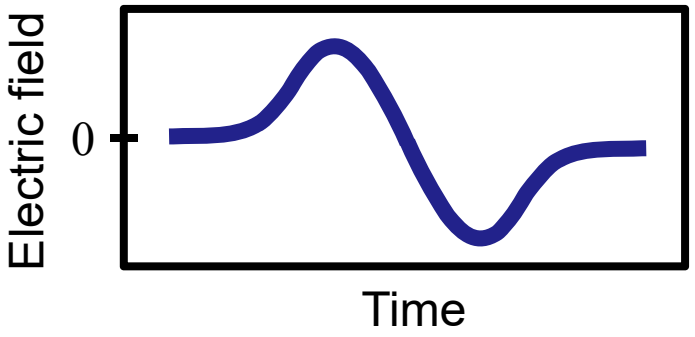
Imagine a **half-cycle pulse**. Its **electric field** looks like this:



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

$$z_R = \pi w^2 / \lambda \to 0$$

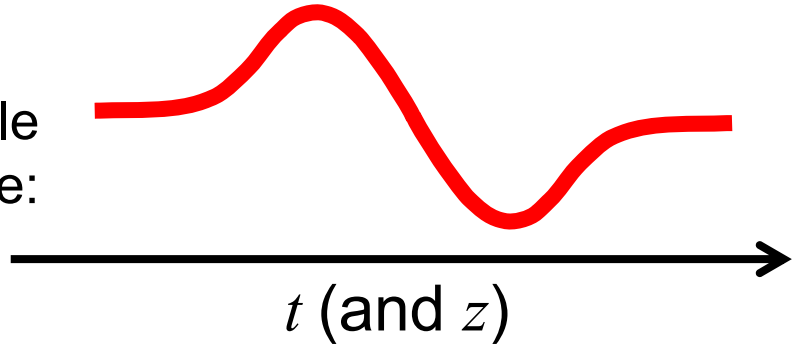
As it propagates to the far field, such a pulse evolves into a **single-cycle pulse**:



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

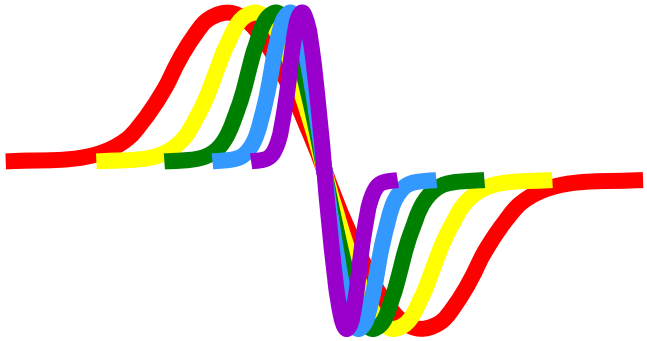
Attosecond pulses are short-wavelength pulses.

A single-cycle red pulse:



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

Compressing a single-cycle red pulse:



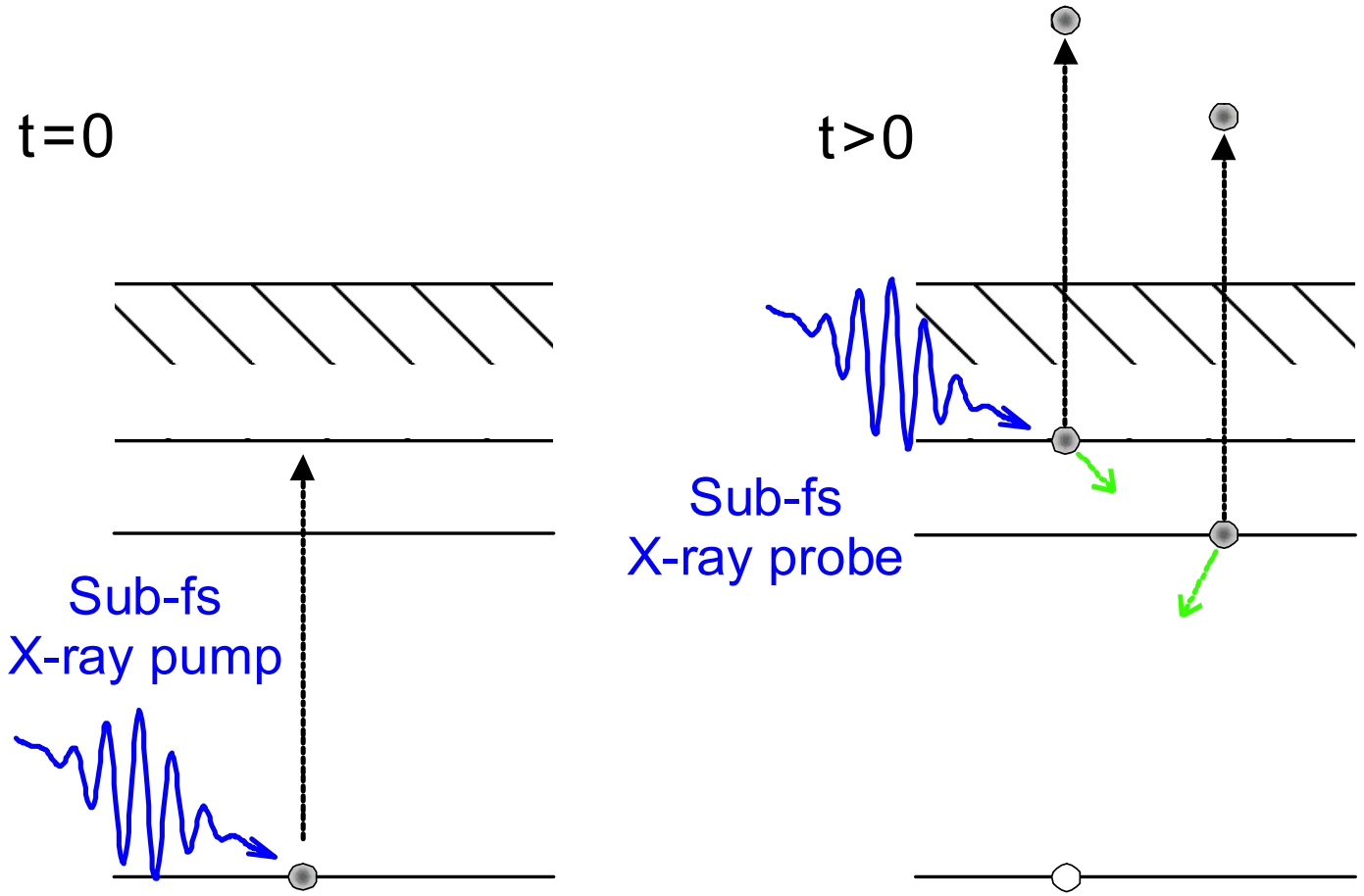
To achieve a period of 1 fs requires a wavelength of 300 nm.

If you compress a single-cycle pulse of one wavelength, you necessarily reduce its wavelength!

X-ray Pump-Probe Spectroscopy

Sub-femtosecond pulses necessarily have high-photon-energy (UV or x-ray)

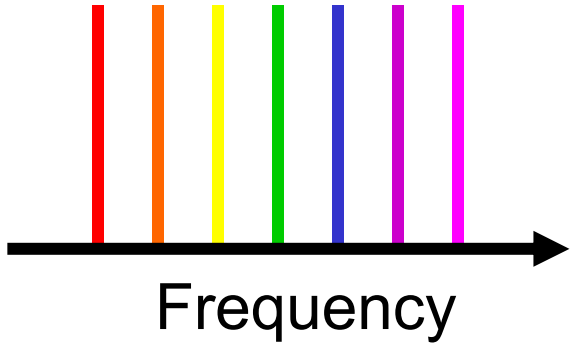
With such pulses, we could trace inner shell relaxation processes.



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)

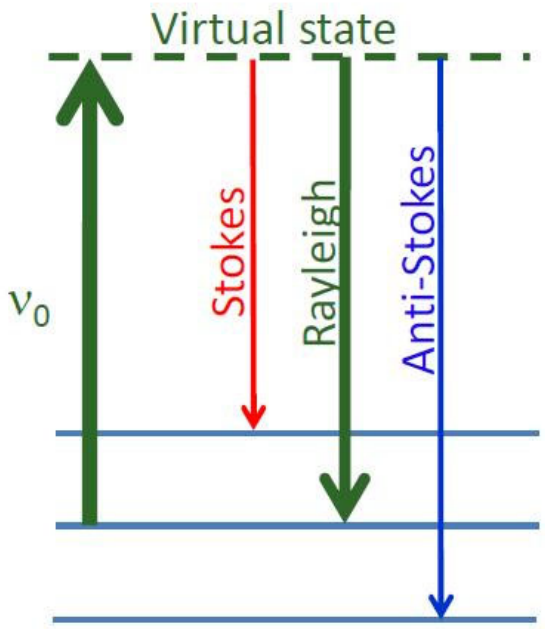


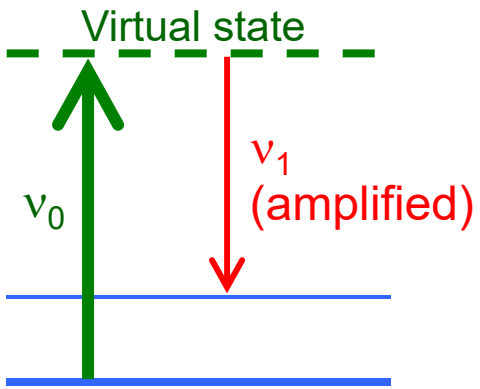
But do these methods phase-lock the modes?

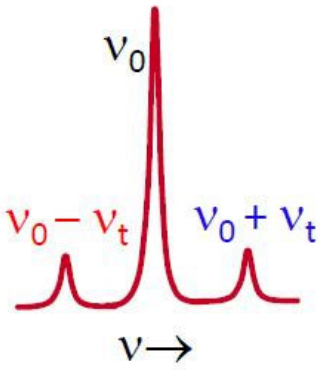
Raman effect: the inelastic scattering of photons from molecules

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 ν_t.



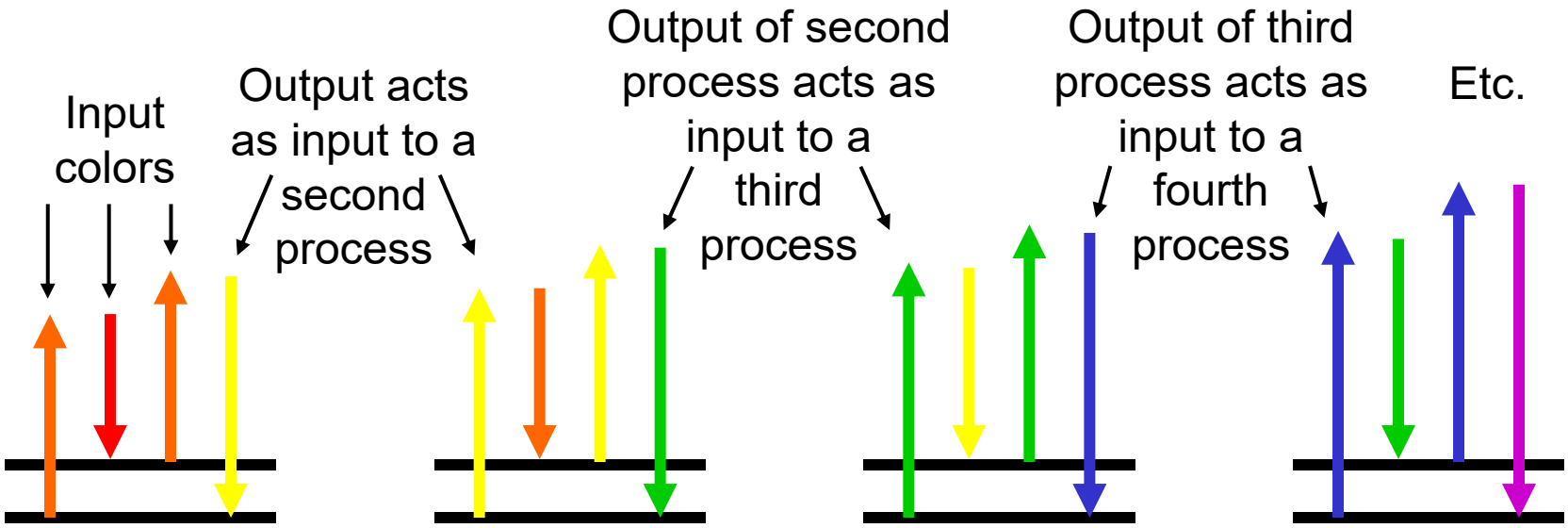




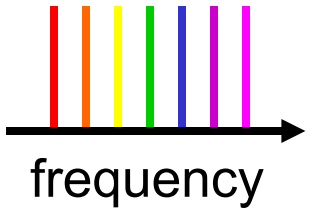
Raman scattering: broadband output

Input two frequencies nearly resonant with a Raman resonance.

At high intensity, the process cascades many times.



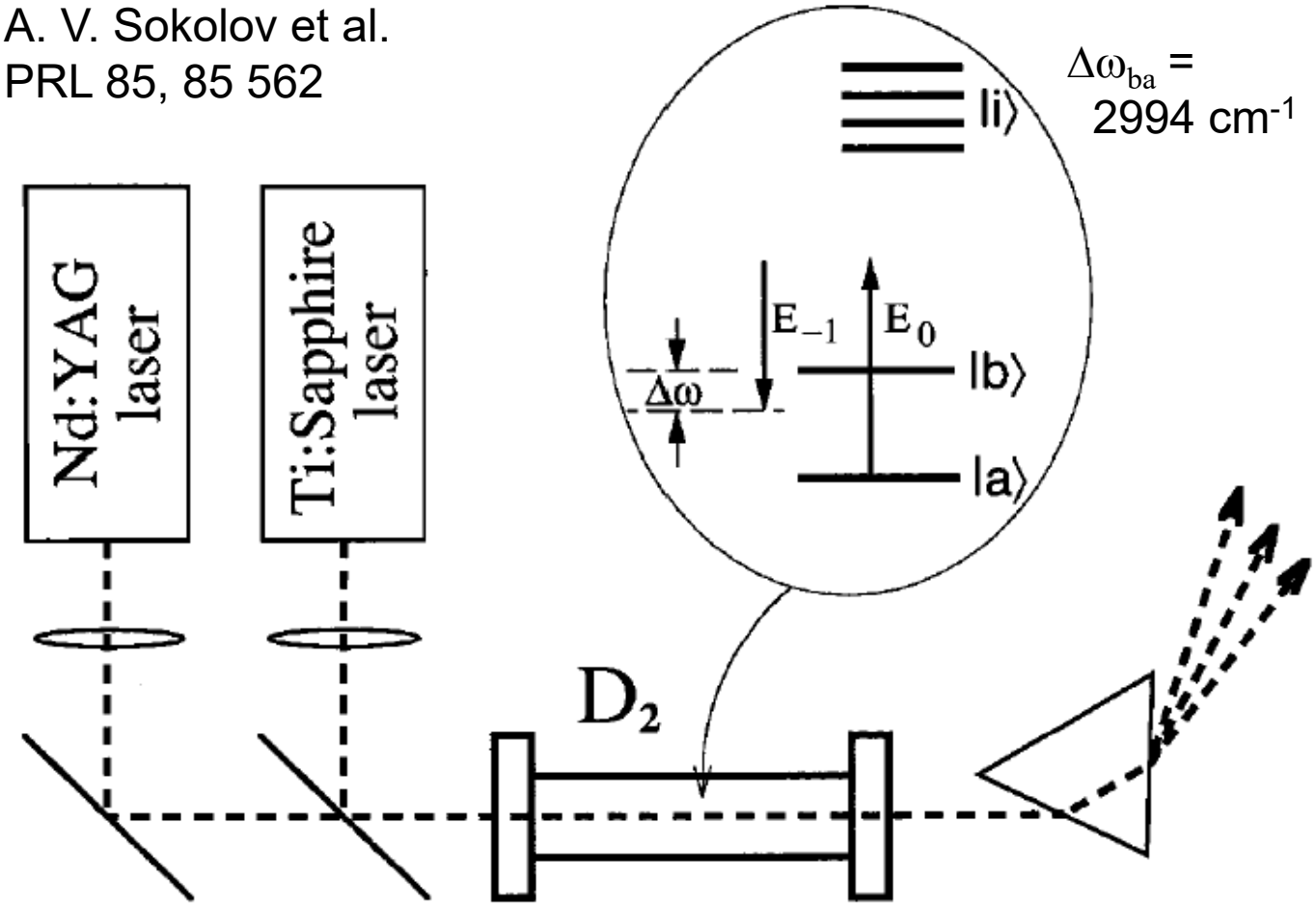
Raman processes can cascade many times, yielding a series of equally spaced modes!

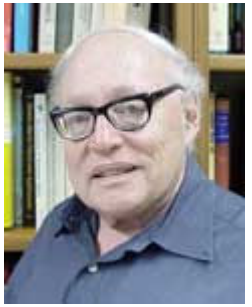


S. E. Harris and A. V. Sokolov PRL 81, 2894

Cascaded Raman generation

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

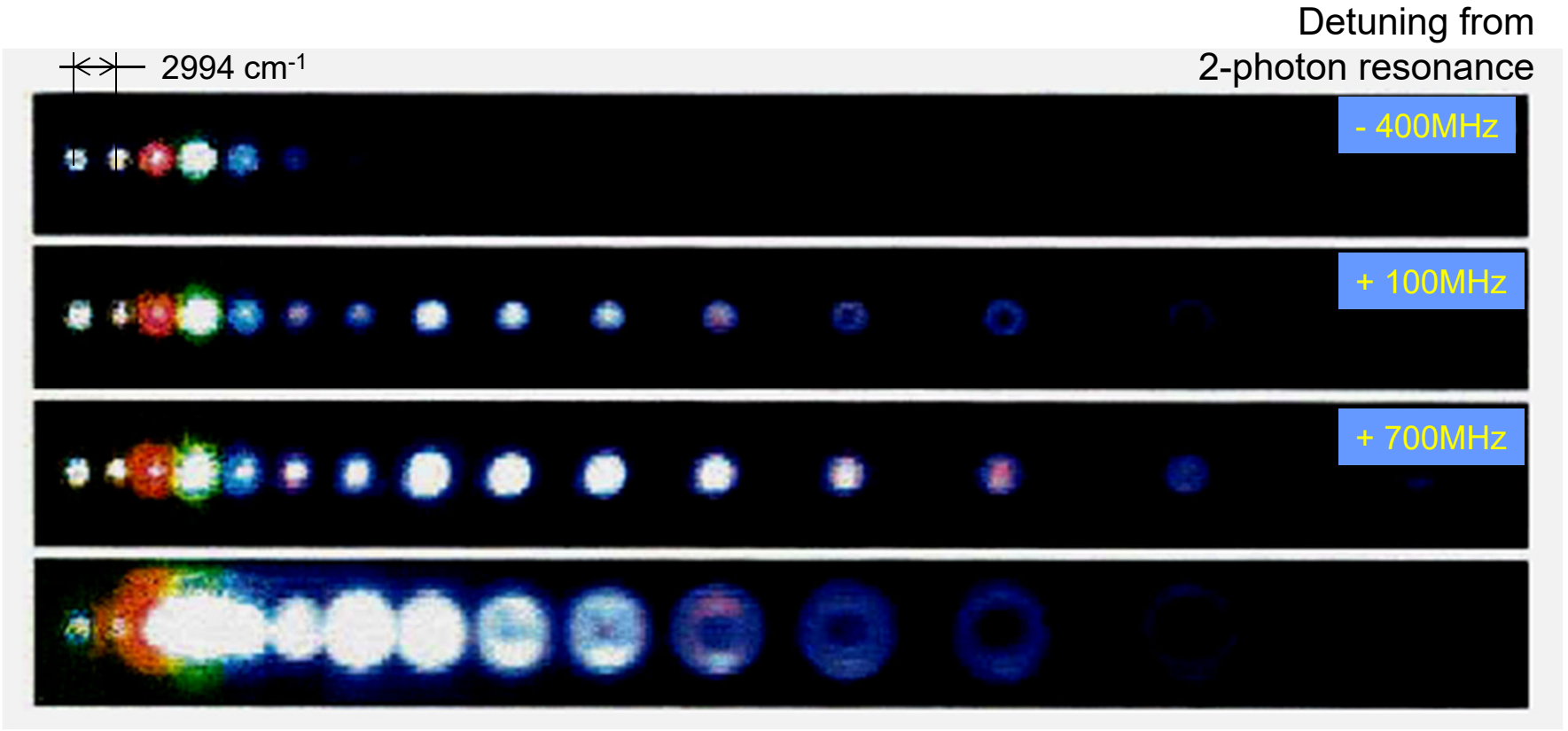




Steve Harris, Stanford Univ.

This can be done with high-power **nanosecond** laser pulses!

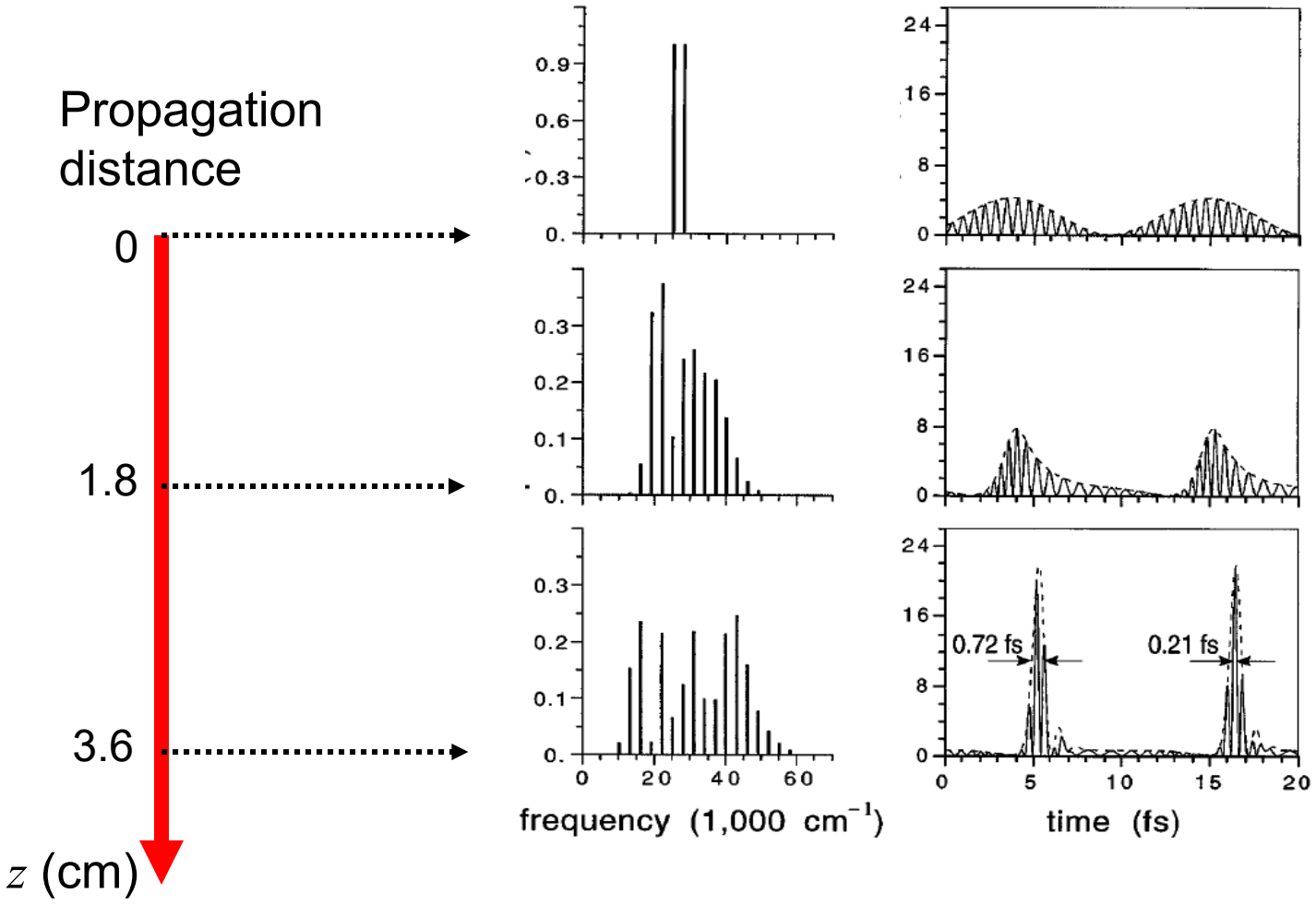
Experimental demonstration of cascaded Raman scattering



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



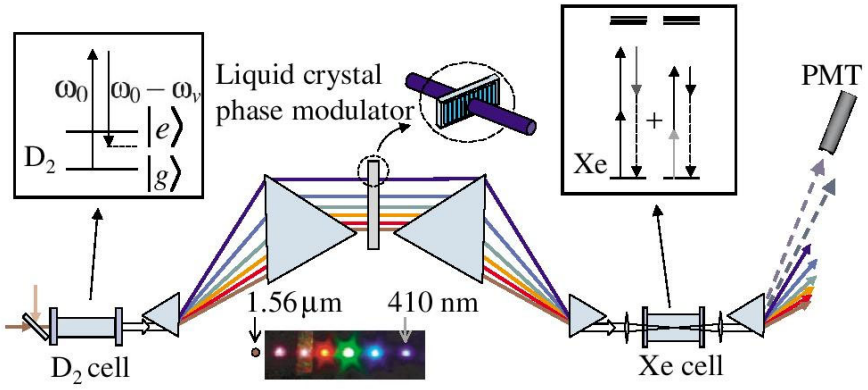
Harris and Sokolov. PRL 81, 2894

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.



Shverdin et al., *Phys. Rev. Lett.* **94**, 033904 (2005)

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

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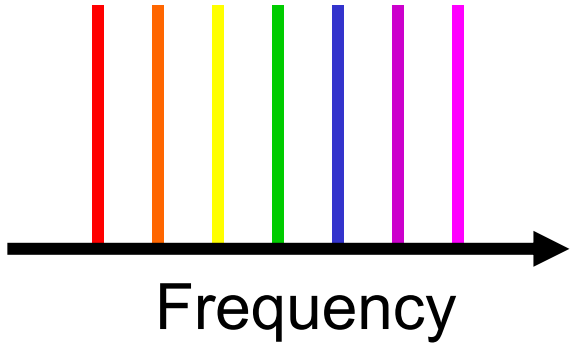
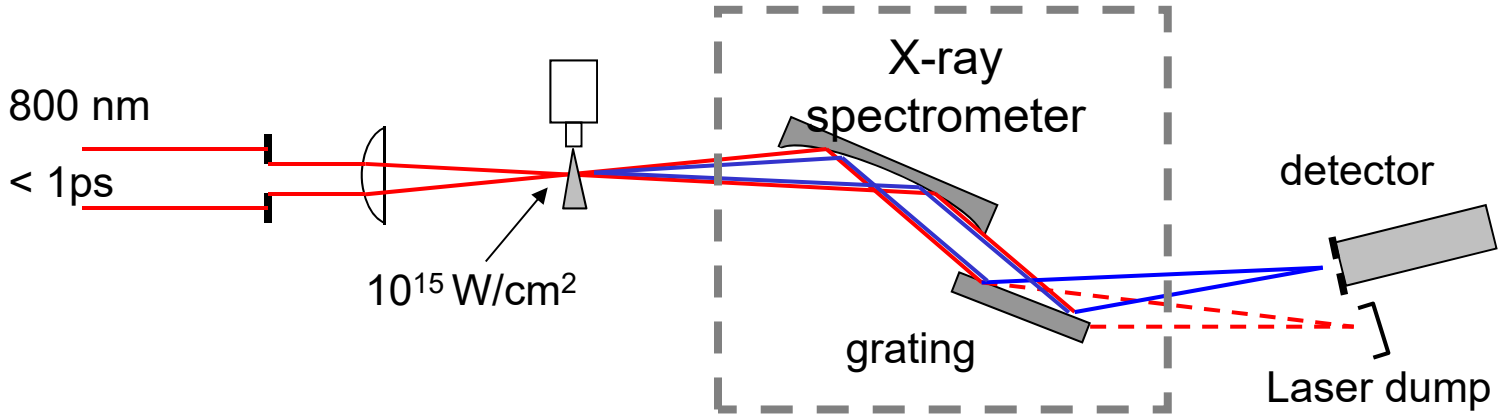
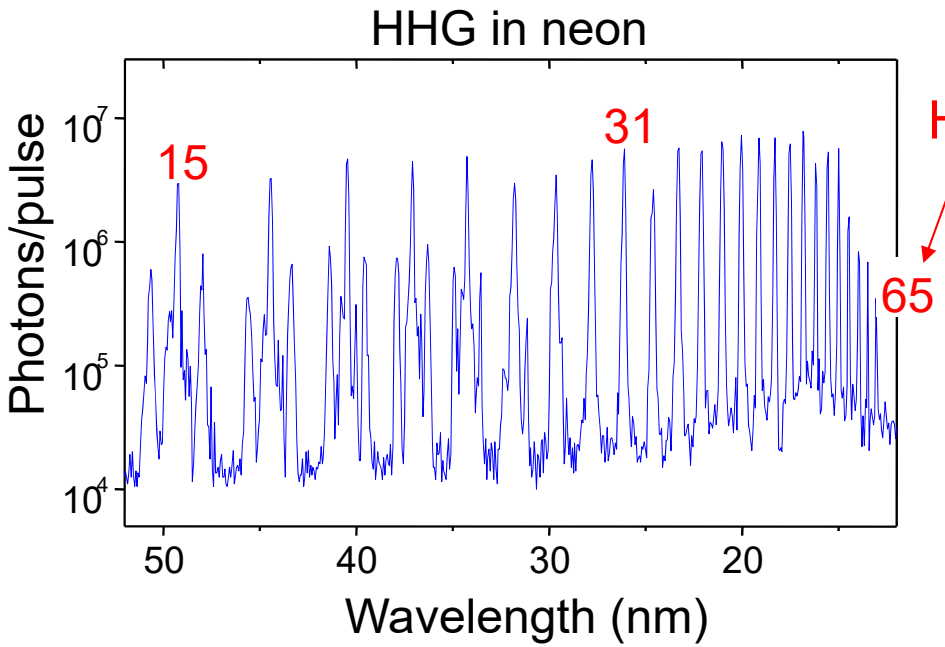
2. High-harmonic generation
(starts with short optical pulses)



More recent work has focused on the second option, which relies on femtosecond pulses.

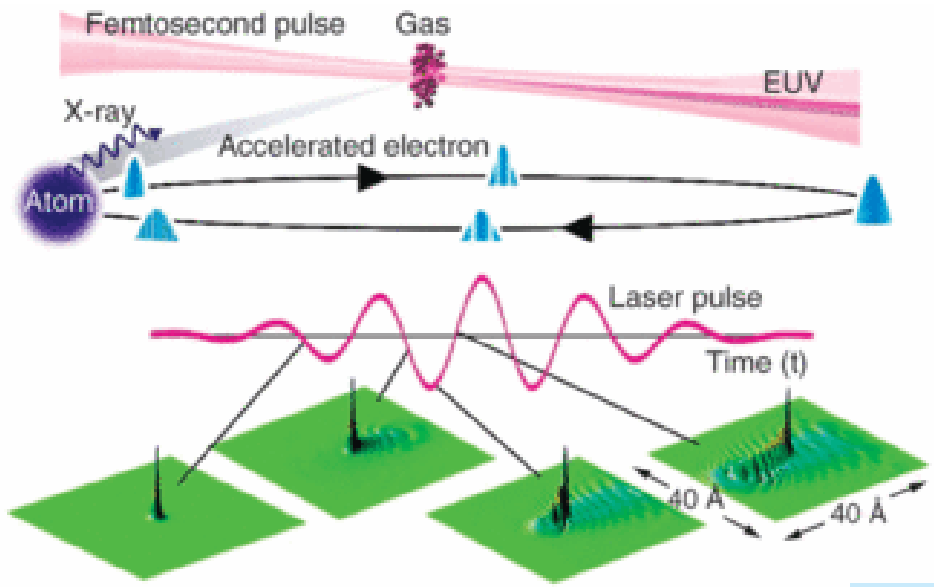
High Harmonic Generation in a gas





HHG produces equally spaced harmonics out to as much as the 300th harmonic, potentially as short as 10 attoseconds!

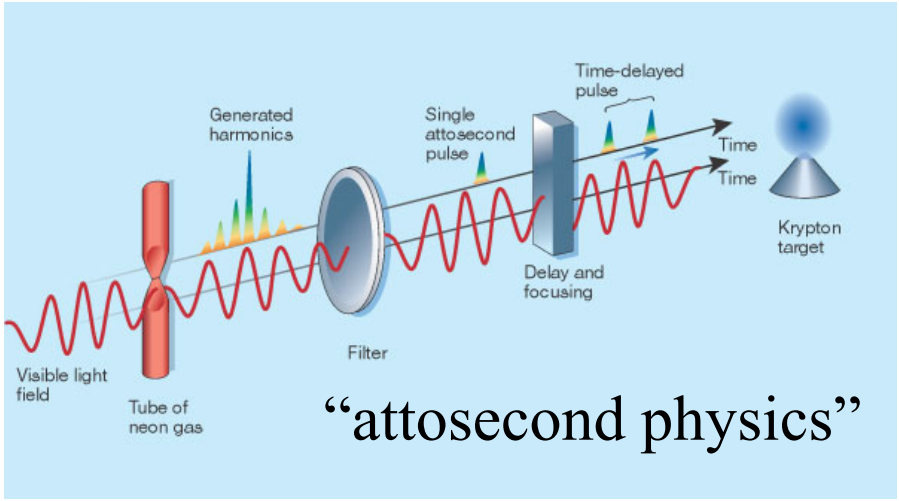
How to explain high harmonic generation?



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.



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.

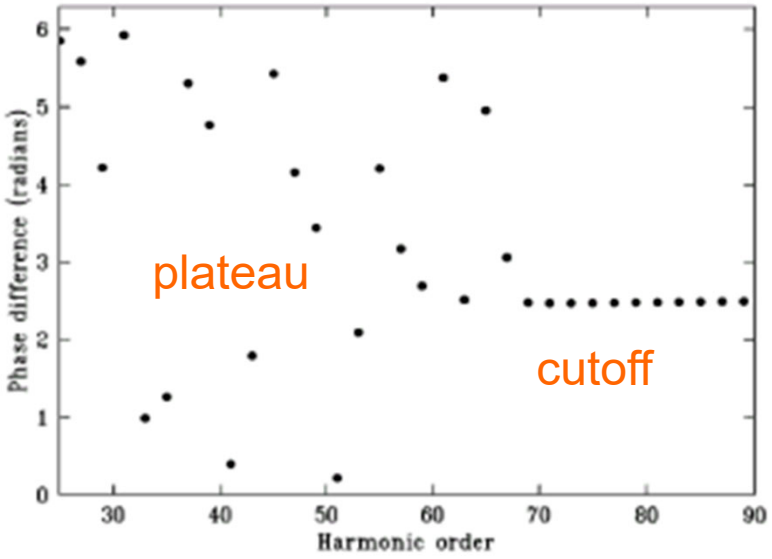
The physics seems to imply that the XUV pulse should be really short…

Maybe HHG just naturally produces attosecond pulses without even trying…

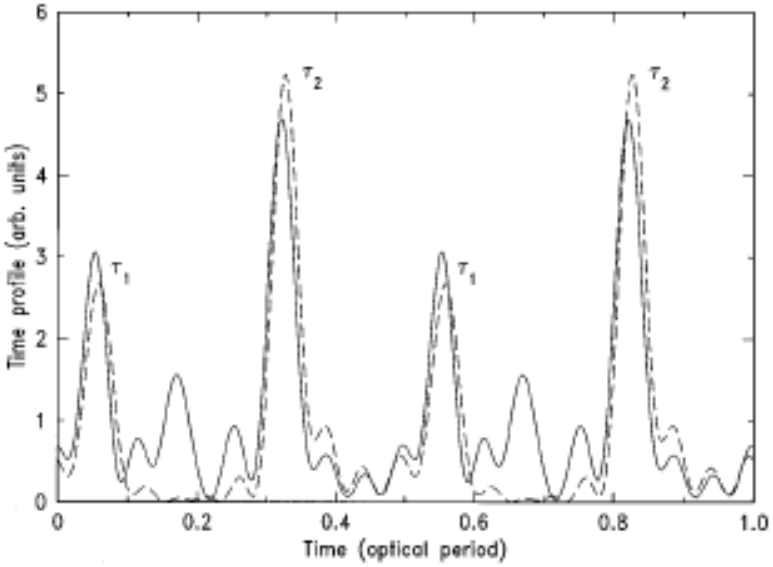
Are the high harmonics actually in phase?

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

Measuring the relative phases of adjacent harmonics:



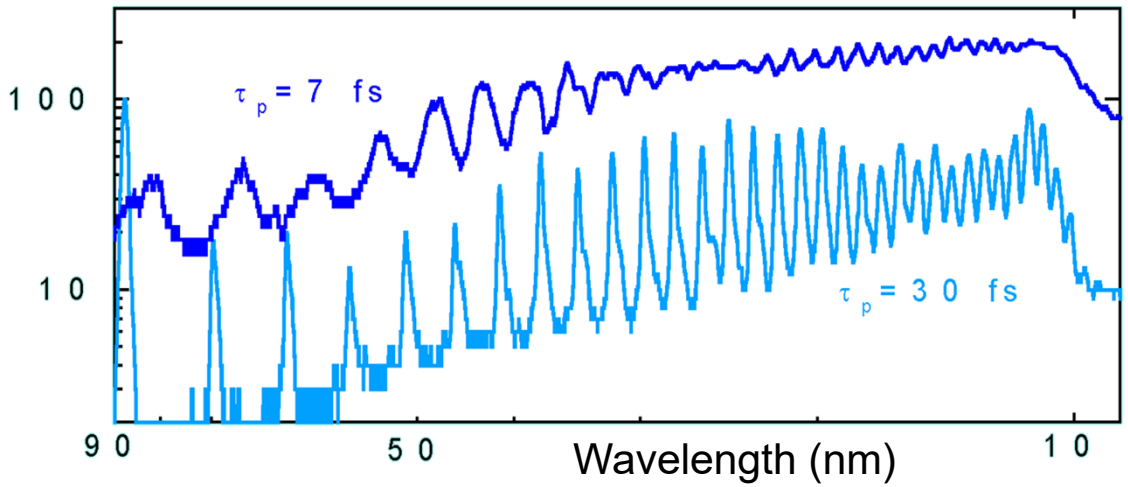
The resulting intensity vs. time:



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



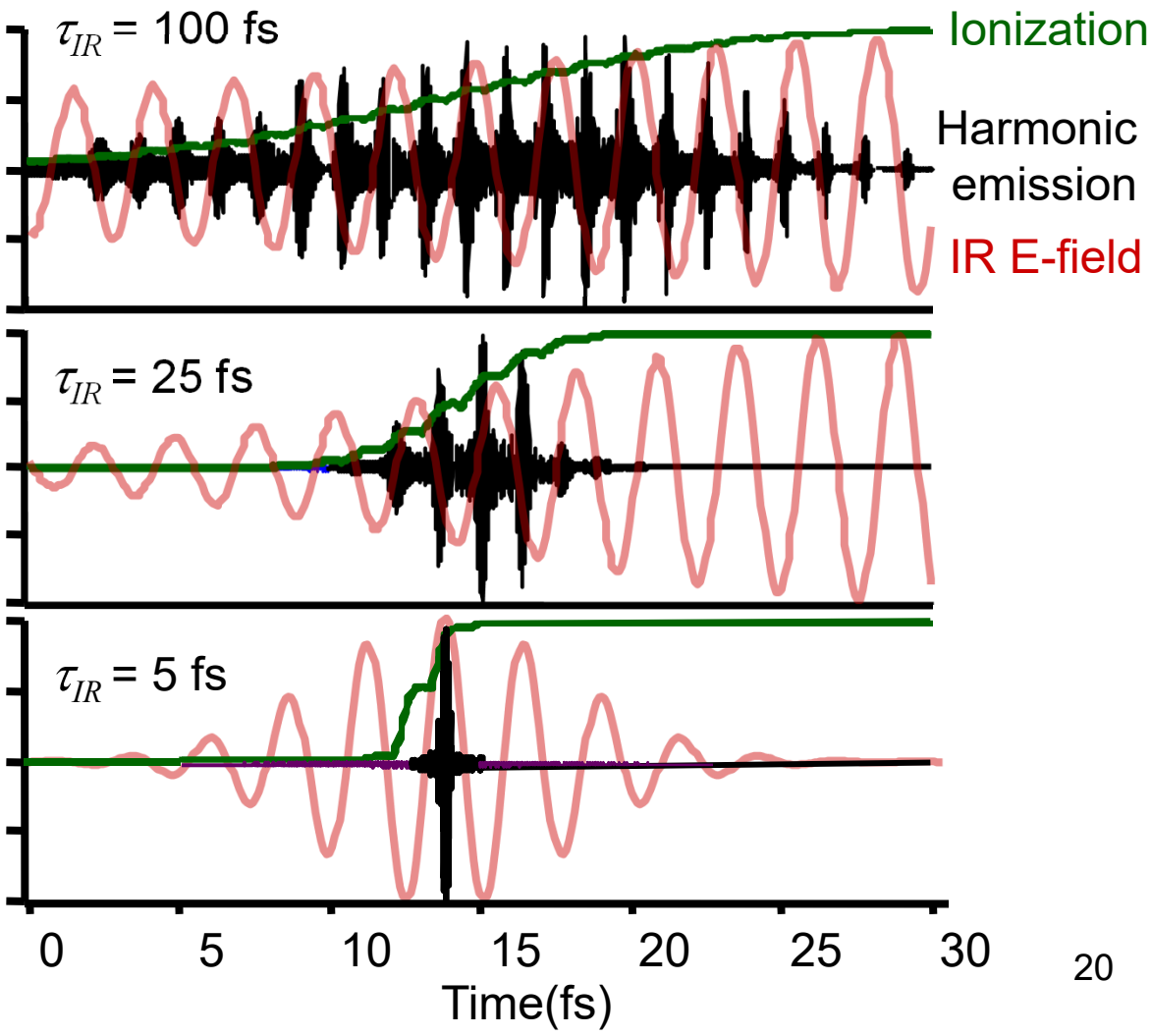
Note the broader harmonic spectrum from the 7-fs pulse.

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 predicted 100-fs input pulses yield many-fs XUV pulses, but few-fs input pulses could yield attosecond XUV pulses…

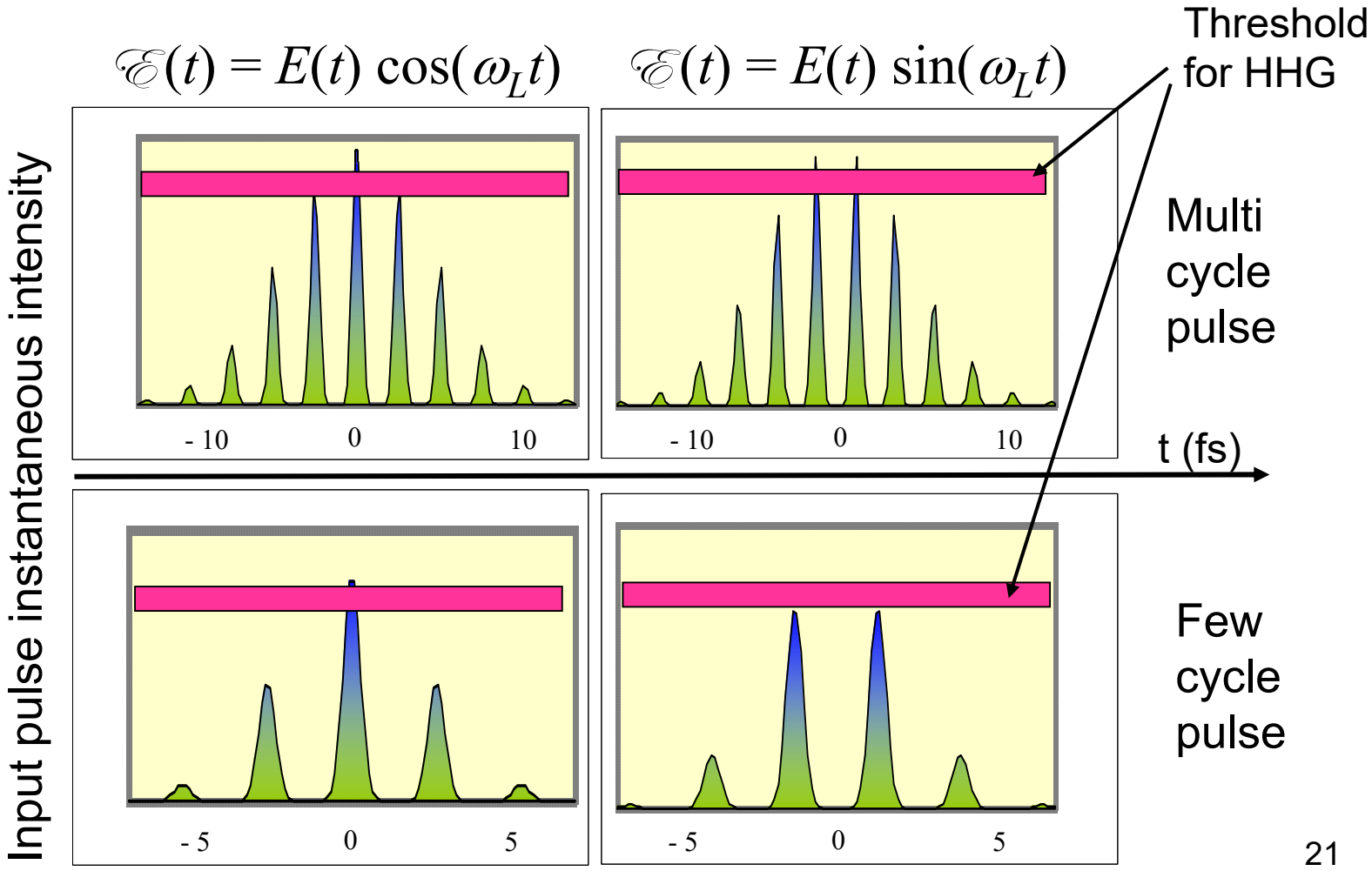
I. P. Christov, M. M. Murnane, and H. C. Kapteyn, PRL 78, 1251, (1997)

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



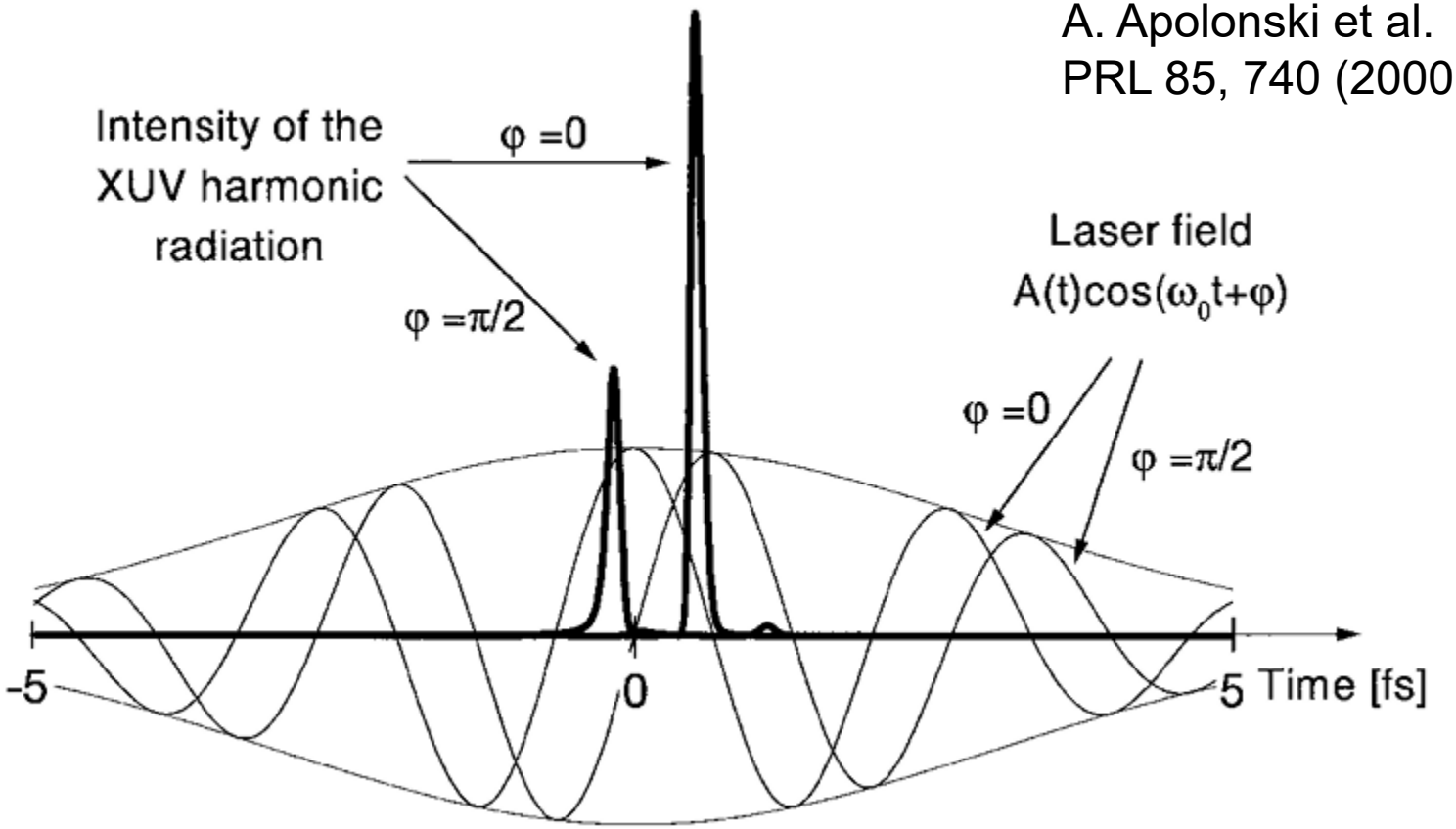
HHG and the absolute phase (carrier-offset phase)

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



Theory predicted much more intense XUV for a zero (or π) absolute phase.

A. Apolonski et al.
PRL 85, 740 (2000)



So the absolute phase of the pulse matters. But what do we know about it?

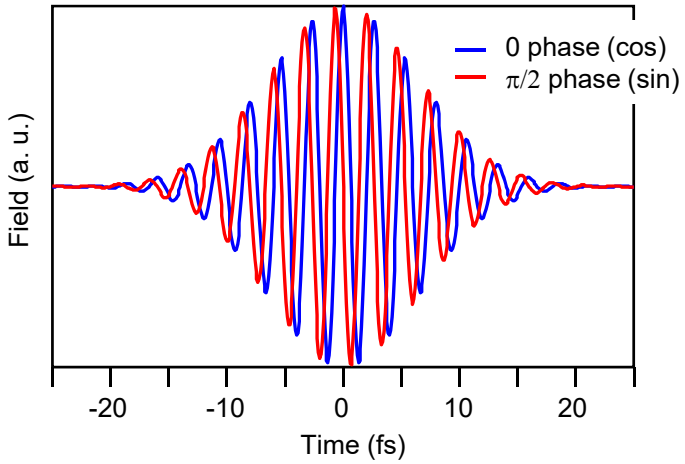
Zeroth-order phase: the absolute phase

- The absolute phase is the same in both the time and frequency domains.

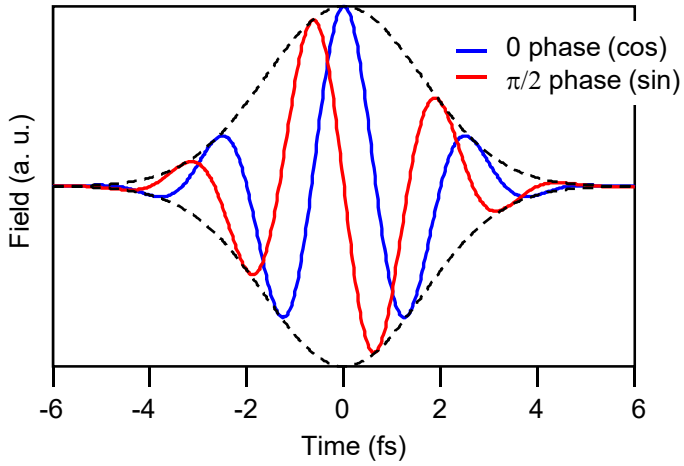
$$f(t)\exp(i\phi_0) \supset F(\omega)\exp(i\phi_0)$$

- 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.

Different absolute phases for a four-cycle pulse



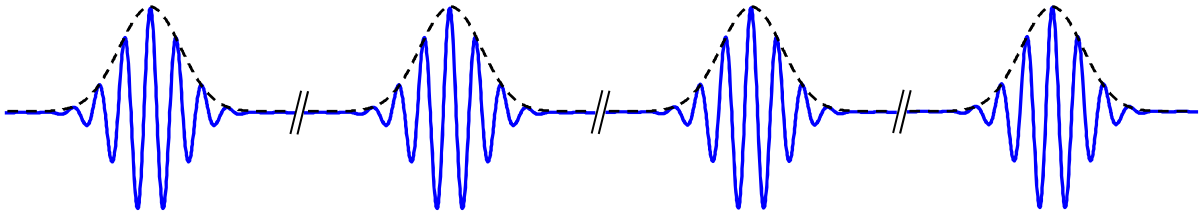
Different absolute phases for a single-cycle pulse



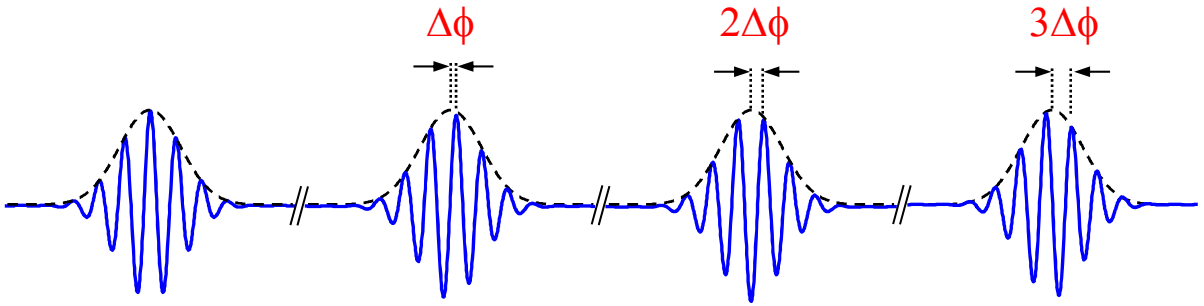
Notice that the two four-cycle pulses look alike, but the two single-cycle pulses are quite different.

Zeroth-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.

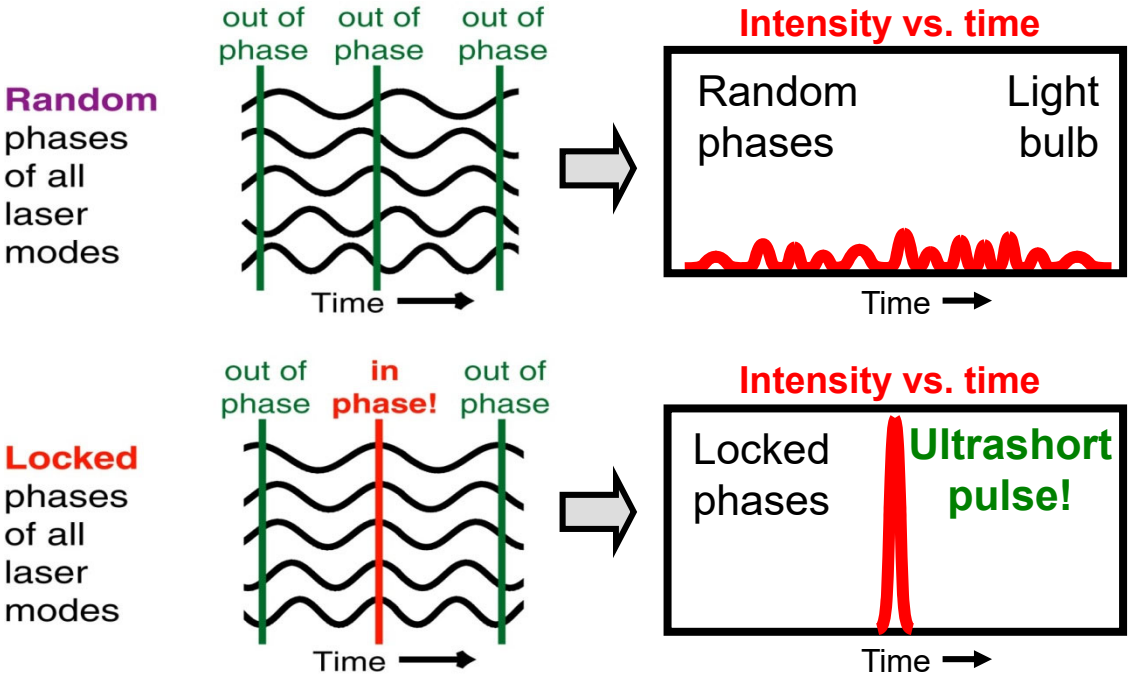


Here, the absolute phase increases from pulse to pulse.

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)



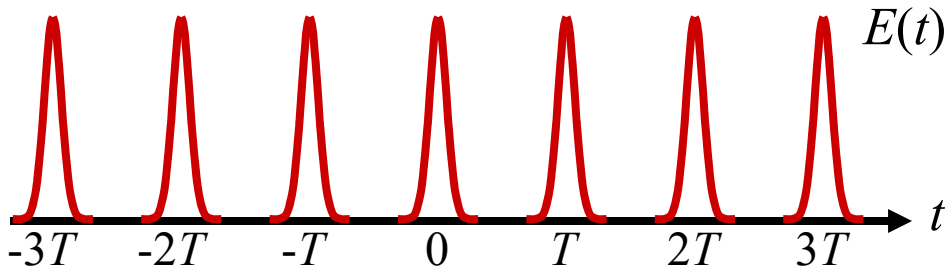
Note: if all the modes are in phase at a particular time (as shown), then it is guaranteed that they will all come into phase periodically, at the commensurate period of all the modes.

Mode-locked lasers produce trains of equally spaced pulses.

Reminder: an infinite train of pulses

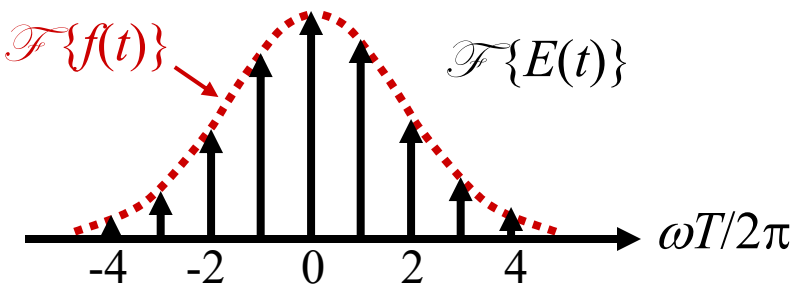
An infinite train of identical pulses can be written as a convolution with the Shah function:

$$E(t) = \mathrm{III}(t/T) * f(t)$$



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:

$$\tilde{E}(\omega) \propto \mathrm{III}(\omega T / 2\pi) F(\omega)$$



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!

$$E(t) = \mathrm{III}(t/T) * f(t)$$

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.

If the duration is invariant, then we know that there is no net GVD in the laser cavity. Therefore:

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

In other words, the group velocity could vary with frequency at certain points in the laser (e.g., inside the gain medium, or in between the prisms), but when added up over one round trip, the value is the same for all frequencies.

This *could* be satisfied if the effective refractive index of the laser cavity is dispersionless: $\frac{\partial k}{\partial \omega} = \frac{\partial}{\partial \omega}\left(\frac{n\omega}{c_0}\right) = \frac{n}{c_0} + \frac{\omega}{c_0}\frac{\partial n}{\partial \omega}$

A stable pulse train in a dispersive laser cavity

But it also could be satisfied if the effective index of the laser cavity is given by this:

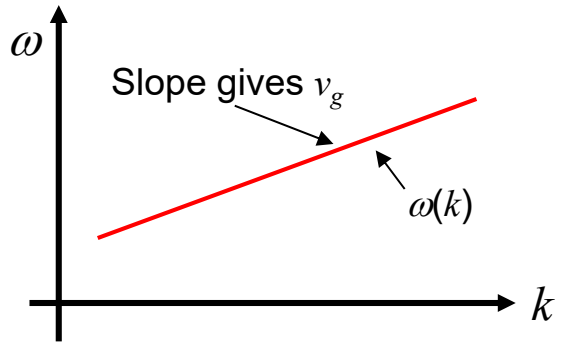
$$n(\omega) = c_0\left(A + \frac{B}{\omega}\right)$$

in which case: $\dfrac{\partial k}{\partial \omega} = \dfrac{n}{c_0} + \dfrac{\omega}{c_0} c_0\left(-\dfrac{B}{\omega^2}\right) = A$ which is v_g^{-1}, a constant.

That is, the net GVD is zero, so the pulses still appear as a periodic train with identical intensities.

But the phase velocity can still exhibit dispersion:

$$k = B + \frac{1}{v_g}\omega$$



$$v_P = \frac{\omega}{k} = \frac{v_g}{1 + B\dfrac{v_g}{\omega}}$$

Dispersion of the phase velocity: implications for the spectrum

Now, recall the formula for the cavity modes inside a laser:

$$\omega_m = \frac{2\pi c}{nL_{RT}} m \qquad (m = \text{integer})$$

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

then we immediately find: $\omega_m = v_g\left(\frac{2\pi}{L_{RT}} m - B\right)$

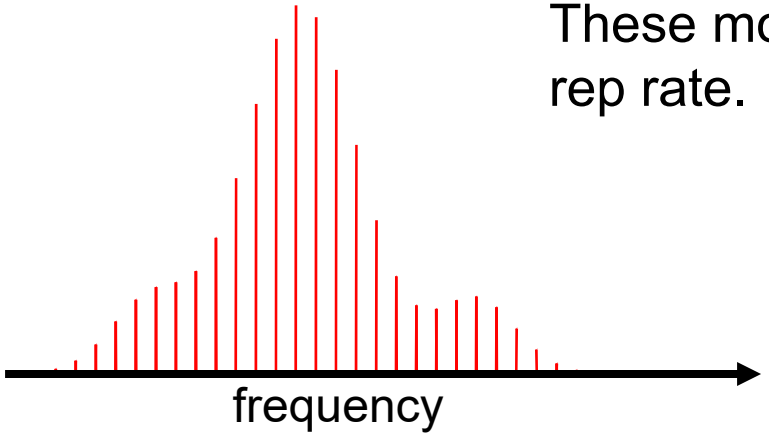
i.e., the cavity modes are still an equally spaced comb in the frequency domain, but they are shifted by a constant offset:

$$f_m = m f_{rep} - f_{offset}$$

where $f_{rep} = v_g/L_{RT}$ and $f_{offset} = v_g B/2\pi$

Carrier envelope phase: time and frequency domain

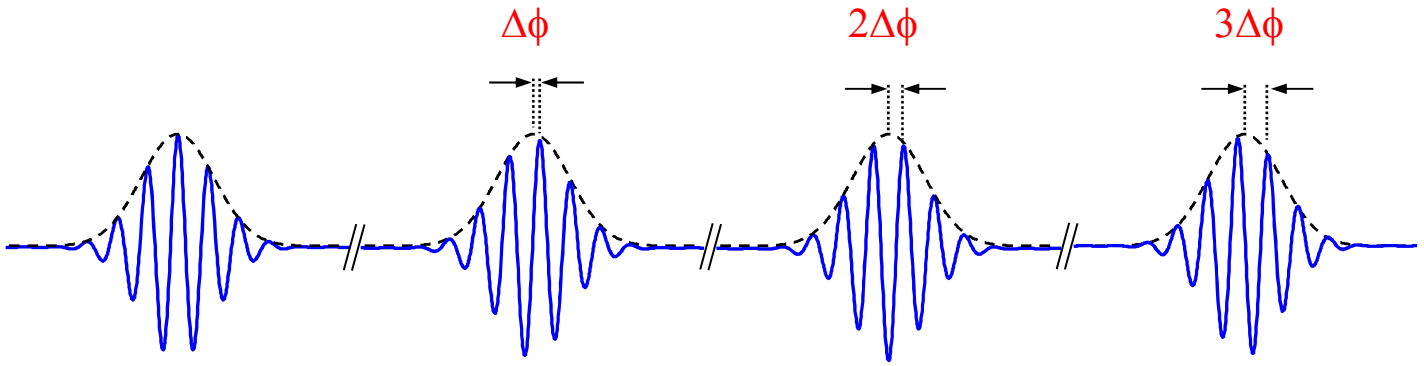
The spectrum of the pulse train consists of a series of delta functions corresponding to the cavity modes.



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

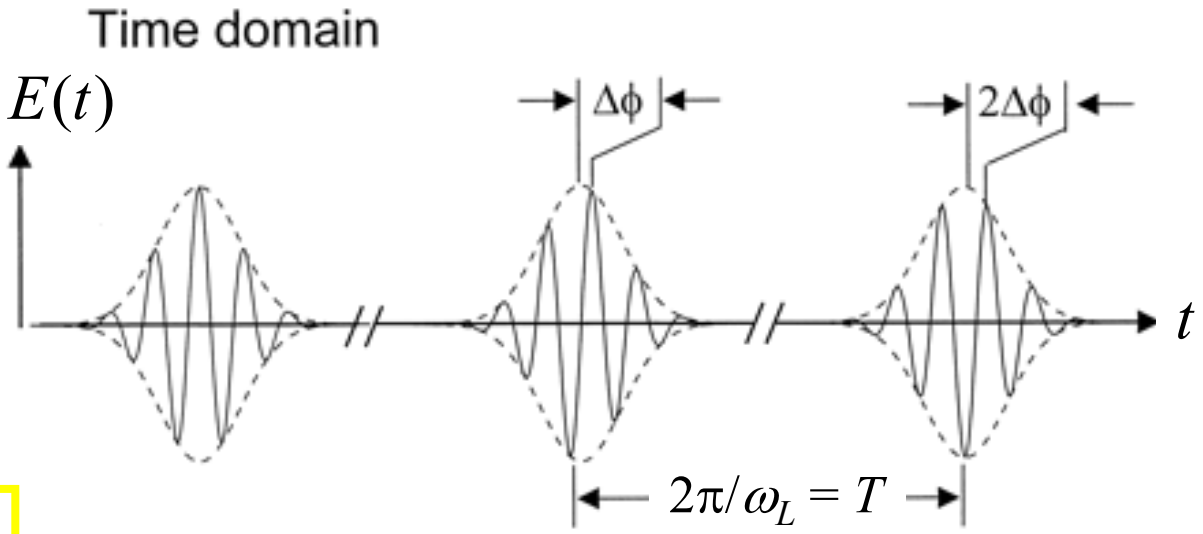
$$f_m = mf_{rep} - f_{offset}$$

where $f_{offset} = \frac{Bv_g}{2\pi}$



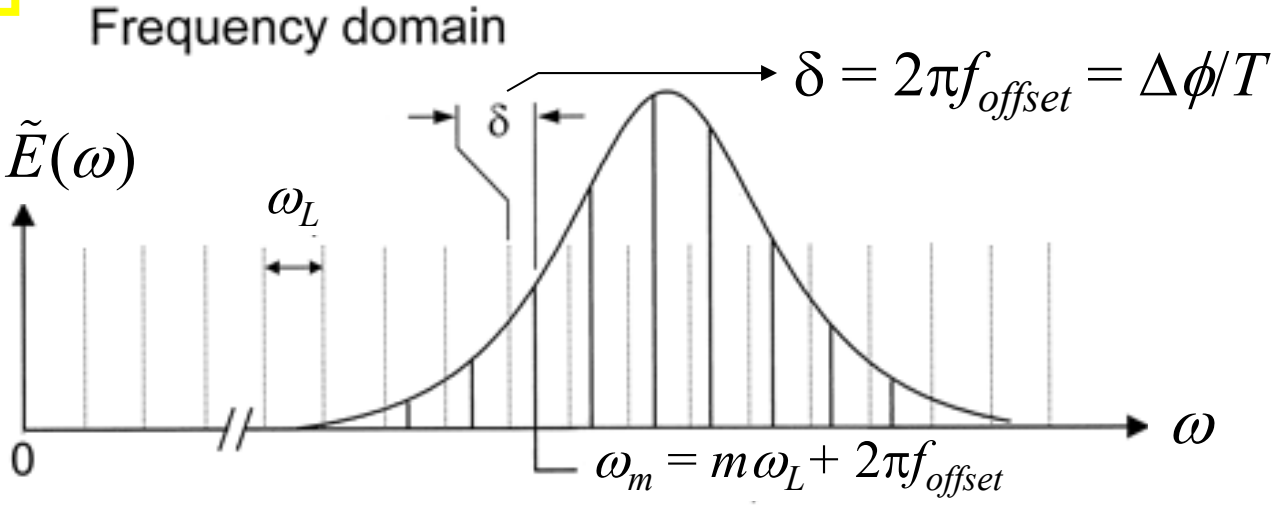
In the time domain, this is manifested as a non-zero carrier envelope phase $\Delta\phi$. The pulses are not identical! Their intensities are the same, but their phases change.

Carrier envelope phase



The m^{th} laser mode:

$$\omega_m = m\omega_L + 2\pi f_{offset}$$



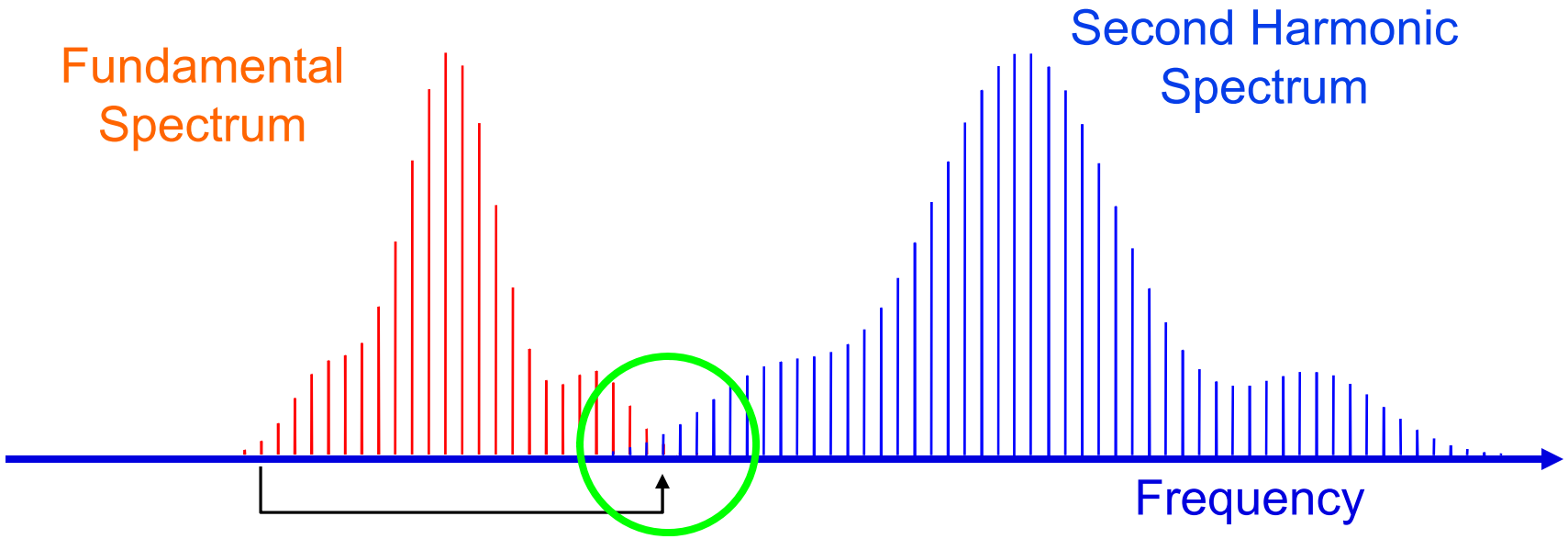
The amount of phase shift from pulse to pulse is related to the offset frequency:

$$2\pi f_{offset} = \Delta\phi/T$$

"Carrier-envelope phase control of femtosecond mode-locked lasers and direct optical frequency synthesis," D. J. Jones, et al., Science 288, 635.

How to measure the absolute phase

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

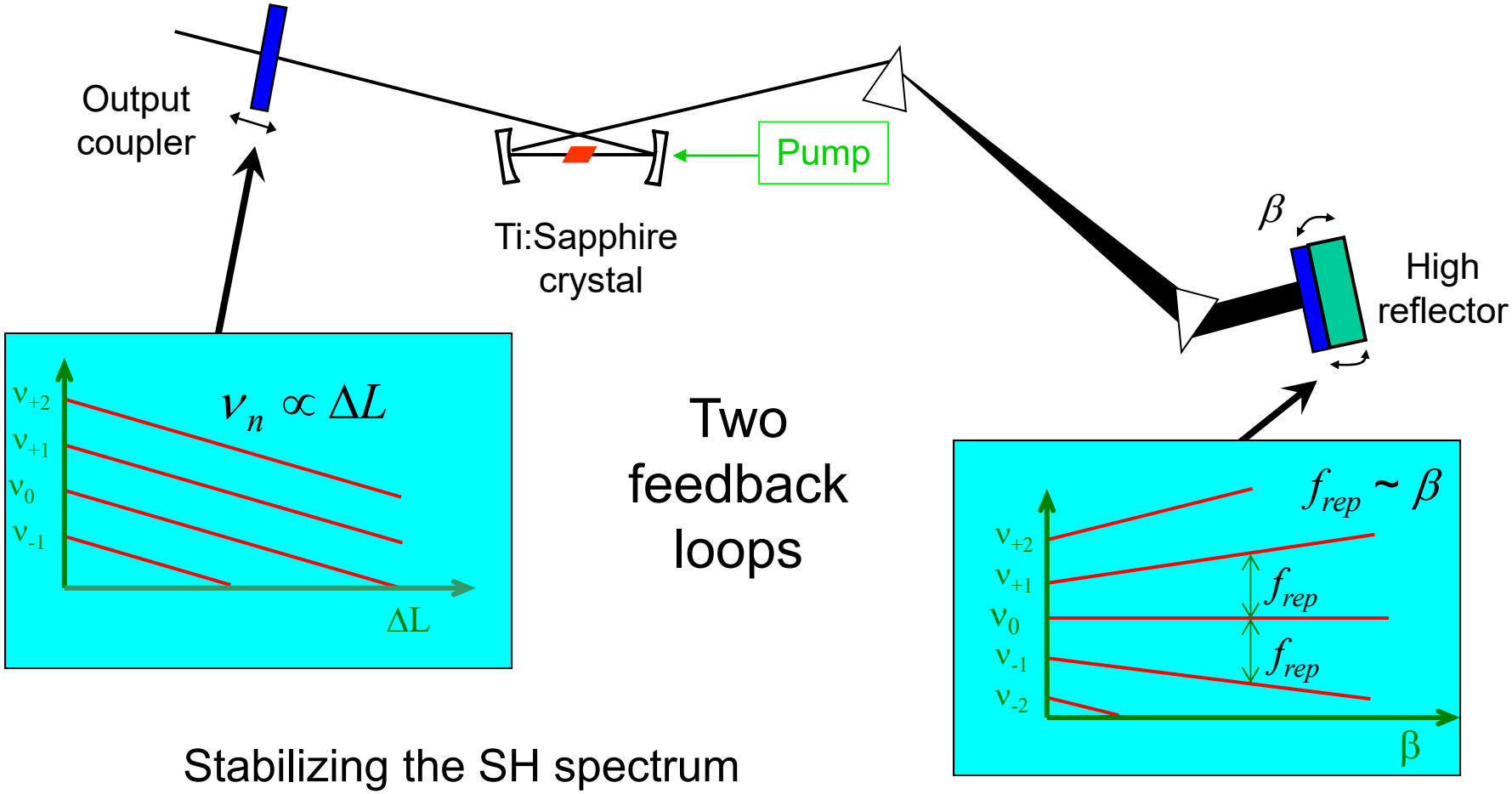


…then comb lines on the low-frequency end of the fundamental spectrum will frequency-double to overlap comb lines on the high-frequency end of the fundamental spectrum.

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

$$2\left(\frac{m}{2}f_{rep}+f_{offset}\right)-\left(mf_{rep}+f_{offset}\right)$$ a direct measurement of f_{offset}!

How to control the absolute phase



Two feedback loops

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

This is a Really Big Deal.

Apolonski et al.
PRL 85 740

Optical frequency combs

The Nobel Prize in Physics 2005

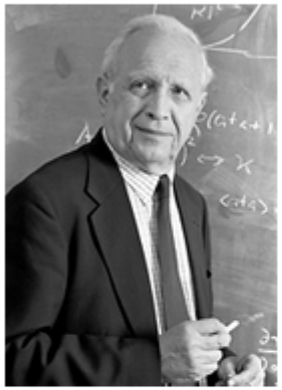


Photo: J.Reed
Roy J. Glauber
Prize share: 1/2

Photo: Sears.P.Studio
John L. Hall
Prize share: 1/4

Photo: F.M. Schmidt
Theodor W. Hänsch
Prize share: 1/4

The Nobel Prize in Physics 2005 was divided, one half awarded to Roy J. Glauber *"for his contribution to the quantum theory of optical coherence"*, the other half jointly to John L. Hall and Theodor W. Hänsch *"for their contributions to the development of laser-based precision spectroscopy, including the optical frequency comb technique"*.

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.

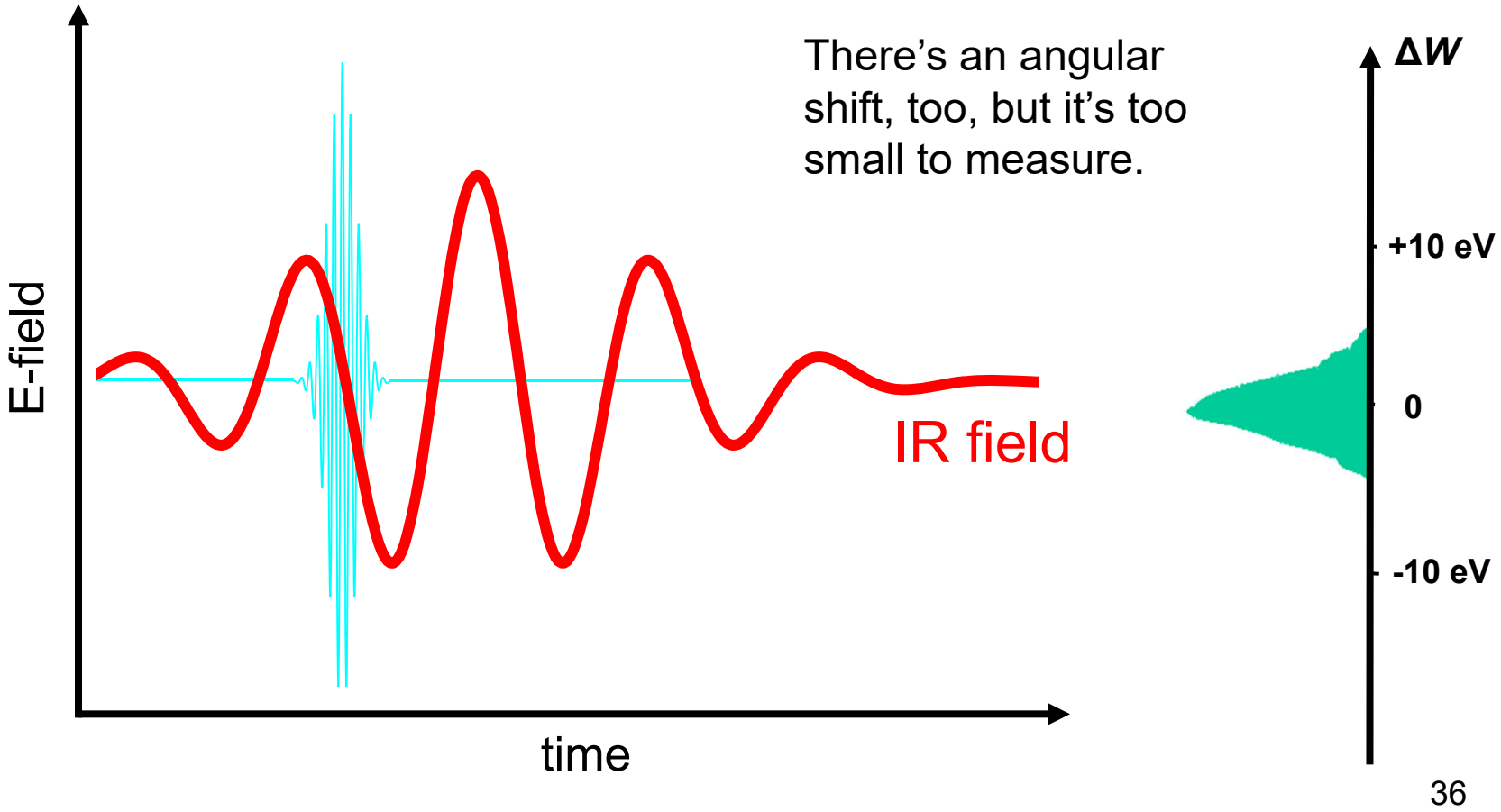
There's a wider choice of nonlinear effects.

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.

Energy shift of as 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.

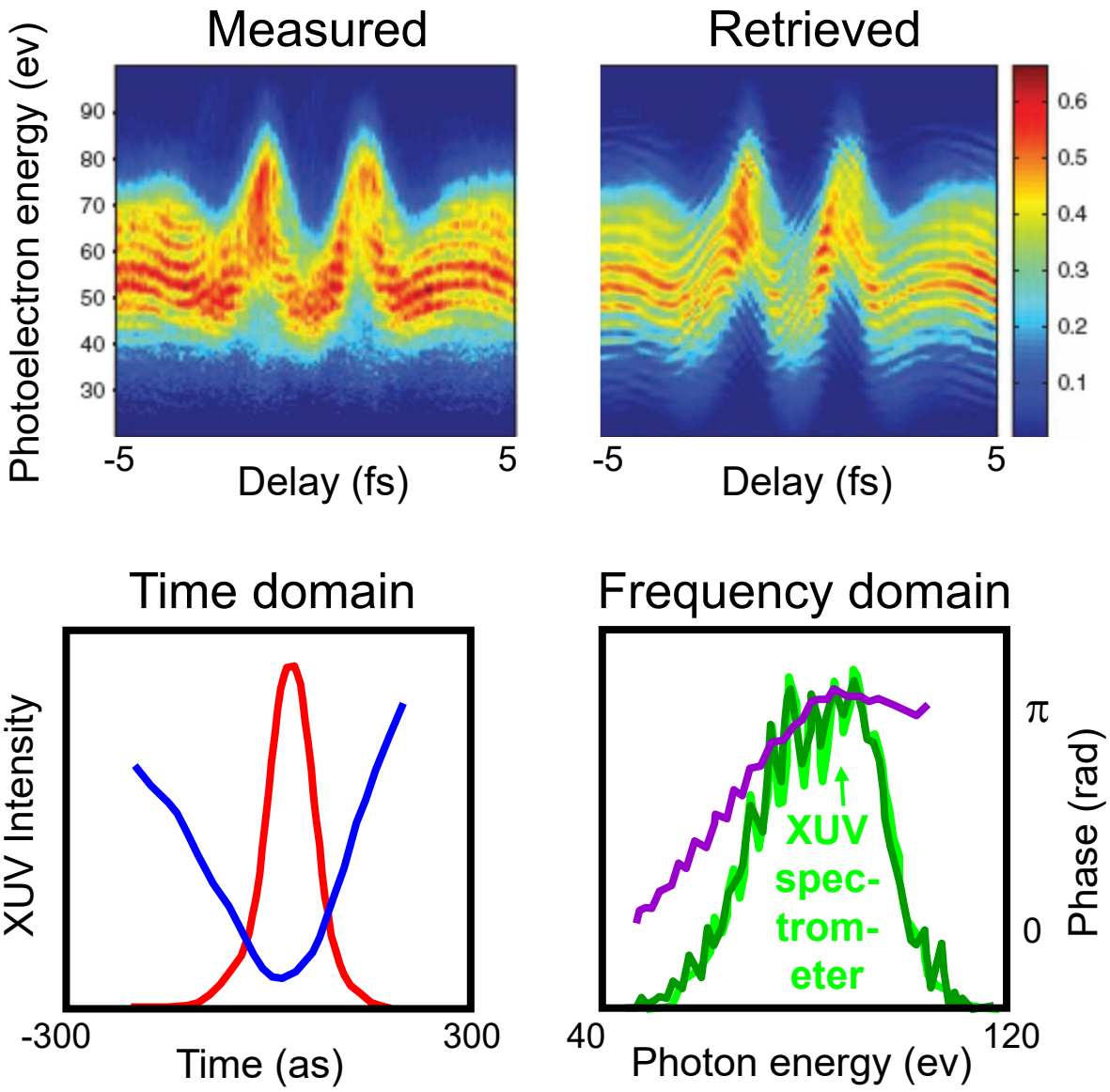


FROG-CRAB: "frequency-resolved optical gating for complete reconstruction of attosecond bursts"

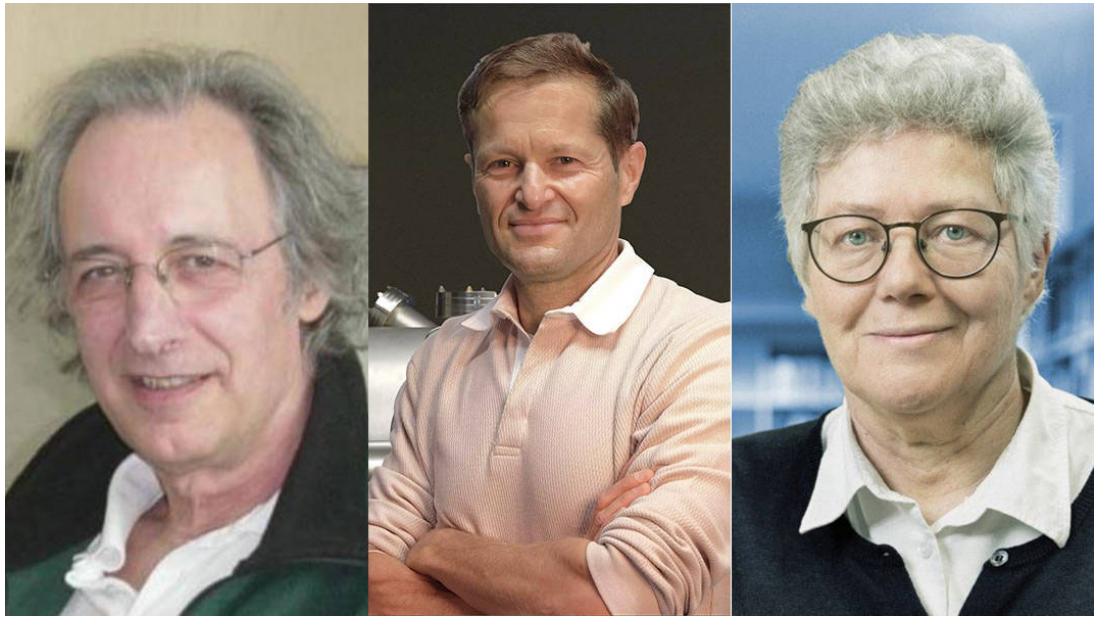
FROG measure-ment of an atto-second pulse

Measure photoelectron spectra, rather than light spectra.

F. Krausz and coworkers, Science, 2008



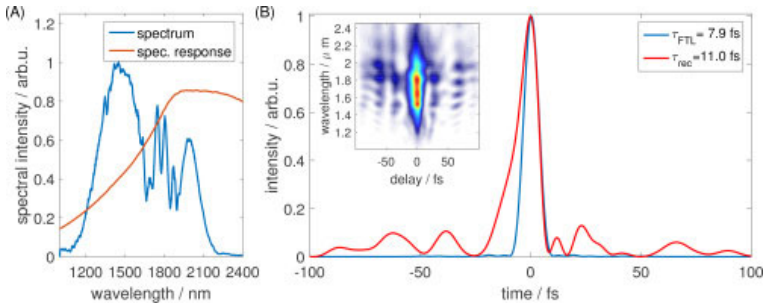
2023 Nobel Prize in physics



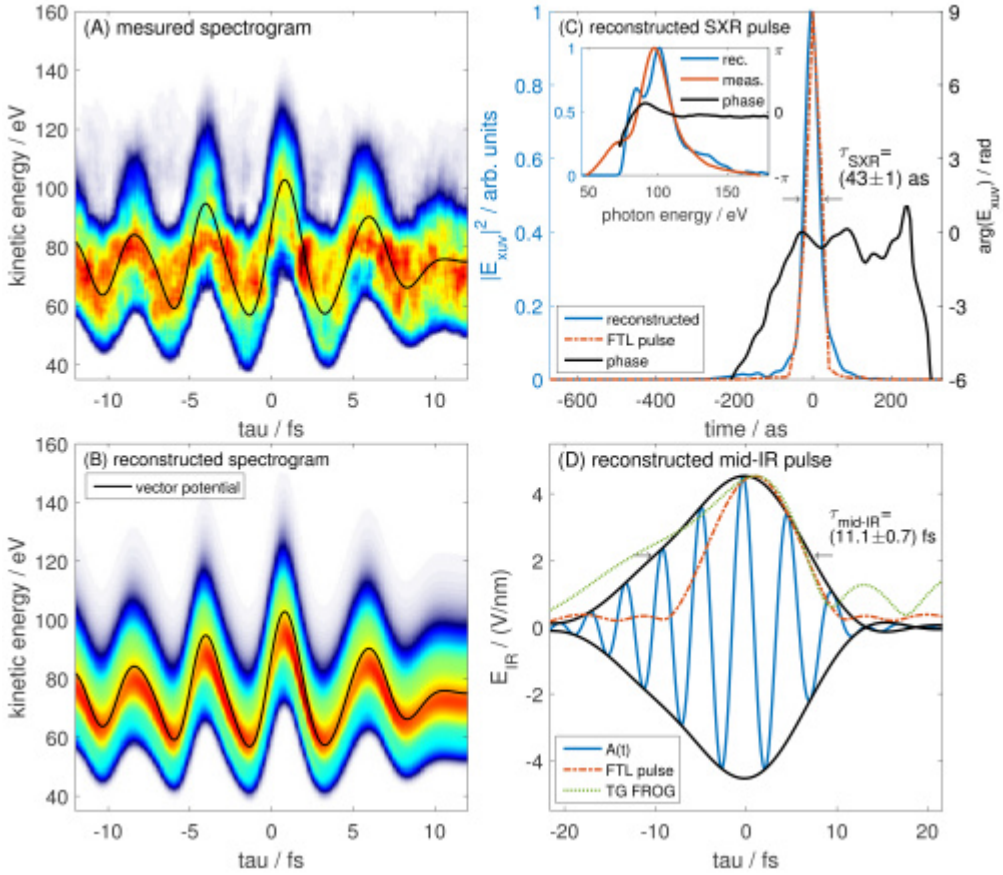
Pierre Agostini Ferenc Krausz Anne L'Huillier

"For experimental methods that generate attosecond pulses of light"

The current world record shortest atto-second pulse



Input pulse:
11 fs duration
λ_{center} = 1.8 μm
480 μJ energy
1 kHz rep rate

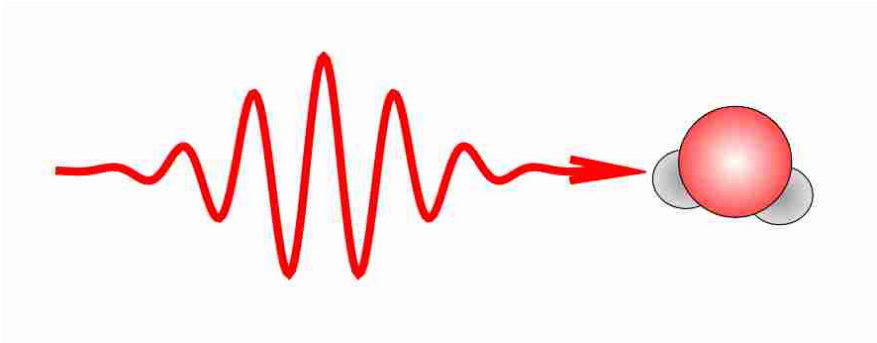


"Streaking of **43-attosecond** soft-X-ray pulses generated by a passively CEP-stable mid-infrared driver"

Thomas Gaumnitz, Arohi Jain, Yoann Pertot, Martin Huppert, Inga Jordan, Fernando Ardana-Lamas, and Hans Jakob Wörner, *Optics Express* 25, 27506-27518 (2017)

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. And attosecond pulses weren't even a dream.

Now, attosecond pulses are real.

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