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Ultrafast laser technology; characterization of ultrafast pulses.

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Ultrafast laser pulses

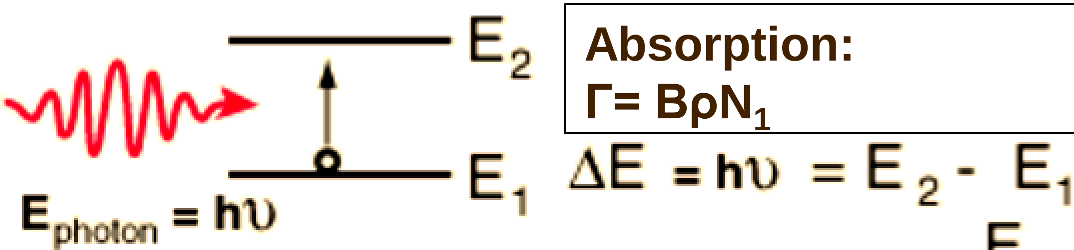
- **Outline**
  - How lasers produce ultrafast pulses, visible to soft x-rays
  - Characterizing ultrafast pulses
  - Shaping ultrafast pulses

- **Selected reviews:**
  - "The generation of ultrashort laser pulses," P.M.W. French, *Rep. Prog. Phys*. **58**, 169, (1995), http://iopscience.iop.org/0034-4885/58/2/001.
  - "Intense few-cycle laser fields: Frontiers of nonlinear optics," Thomas Brabec and Ferenc Krausz, *Reviews of Modern Physics,* **72**, 545 (2000), http://rmp.aps.org/pdf/RMP/v72/i2/p545_1
  - "Characterization of ultrashort electromagnetic pulses," I.A. Walmsley and C. Dorrer, *Advances in Optics and Photonics* **1**, 308–437 (2009) http://dx.doi.org/10.1364/AOP.1.000308
  - "A newcomer's guide to ultrashort pulse shaping and characterization," A. Monmayrant, S. Weber, and B. Chatel, *J. Phys. B* **43,** 103001, (2010), http://dx.doi.org/10.1088/0953-4075/43/10/103001

Ultrafast Laser Pulses: today's topics

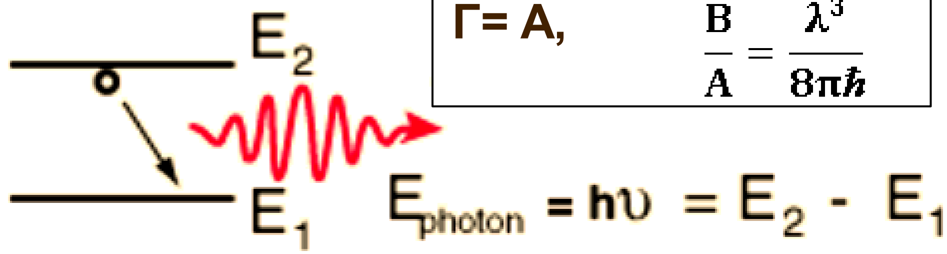
- **Lasers: What are they?**
- **Mode-locking and Dispersion control**
- **Chirped pulse amplification**
- **Self-phase-modulation and Few-cycle pulse generation**
- **High harmonic generation and attosecond pulses**
- **Measuring ultrafast processes and pulses**

Einstein's contribution to this subject: Three competing processes

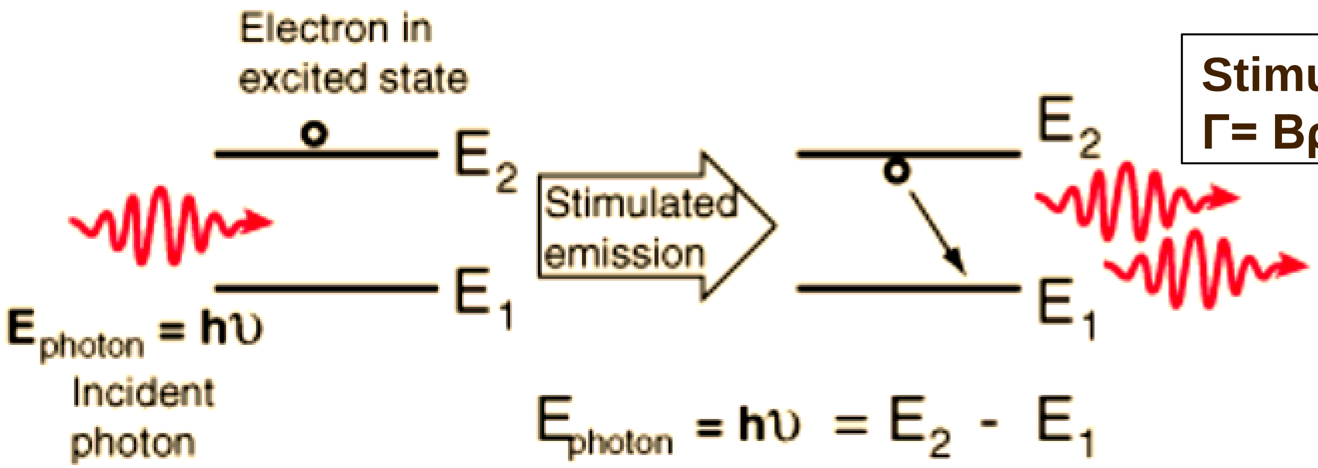


Absorption:
$\Gamma = B\rho N_1$

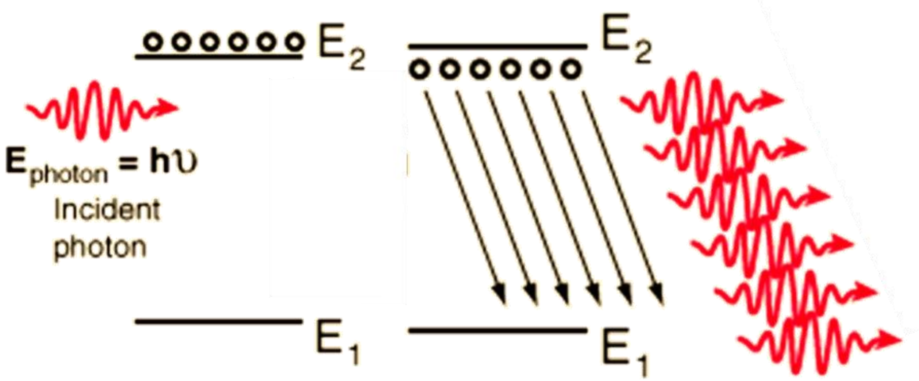
Fluorescence:
$\Gamma = A$, $\frac{B}{A} = \frac{\lambda^3}{8\pi\hbar}$



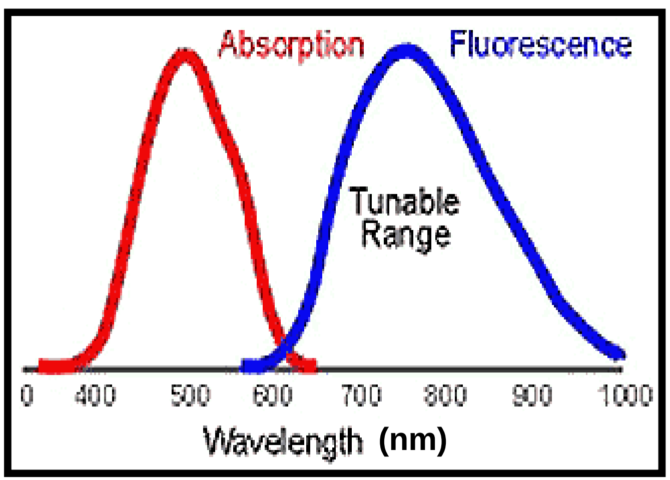
Stimulated emission:
$\Gamma = B\rho N_2$



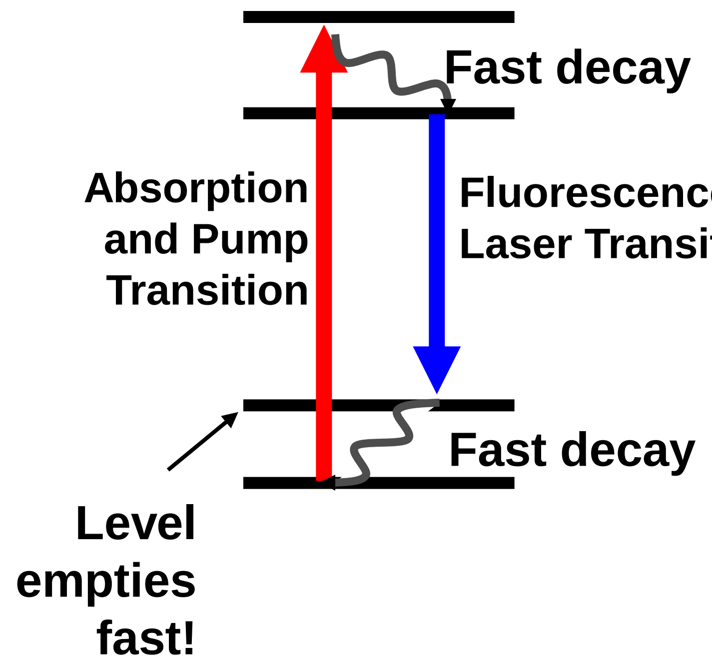
Population inversion and lasing



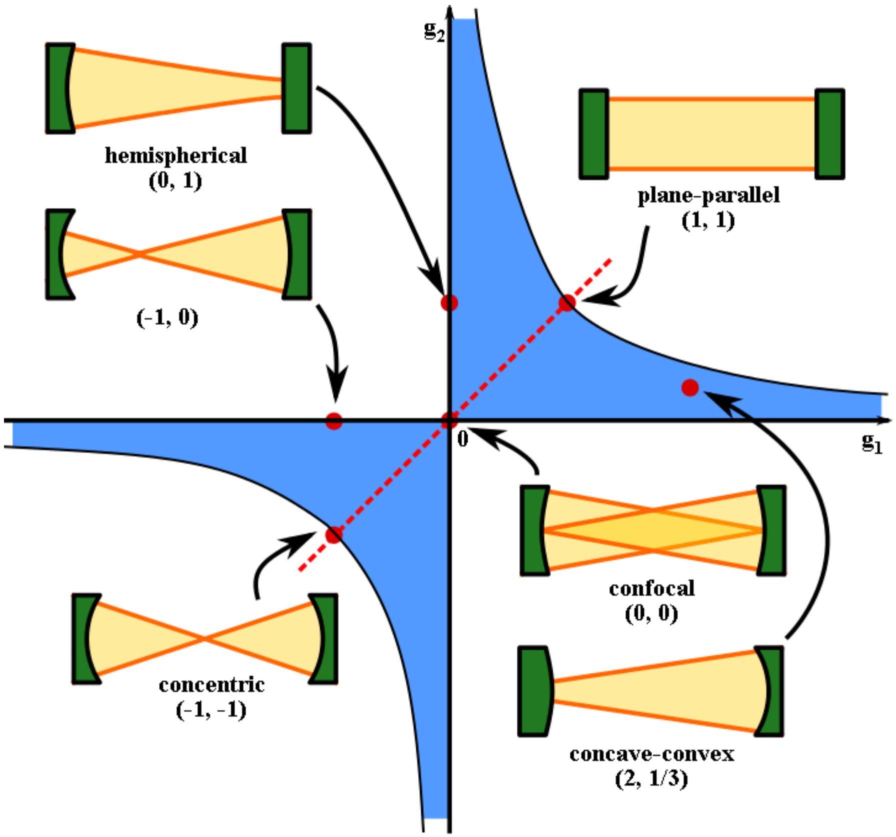
Absorption and emission spectra of Ti:Sapphire



Four-level system



Optical resonators confine the light



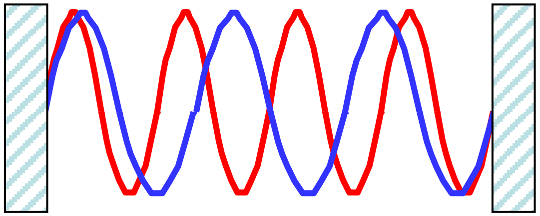
Stability criteria

$$0 \le \left(1 - \frac{L}{R_1}\right)\left(1 - \frac{L}{R_2}\right) \le 1.$$

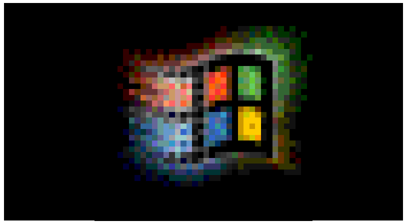
$$g_1 = 1 - \frac{L}{R_1}, \qquad g_2 = 1 - \frac{L}{R_2}.$$

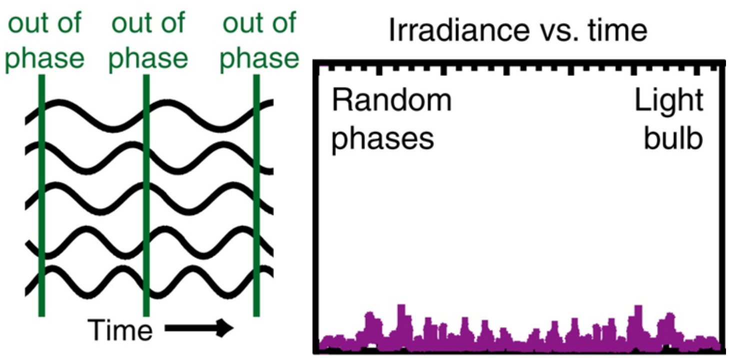
Generating short pulses: "mode-locking"

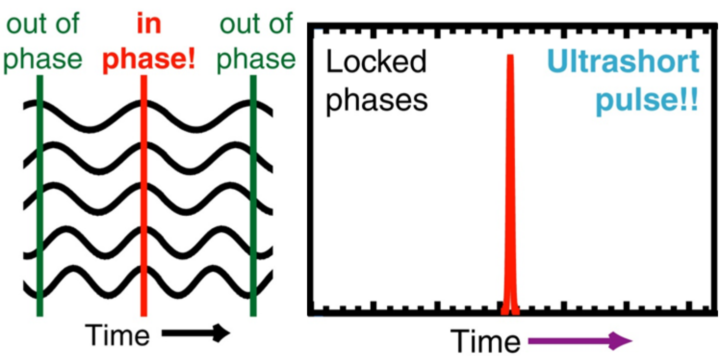
The spectrum of laser modes λ_n = *2L/n*



$$I(t) = \sum_{n=0}^{N} A_n \sin(\omega_n t + \varphi_n)$$

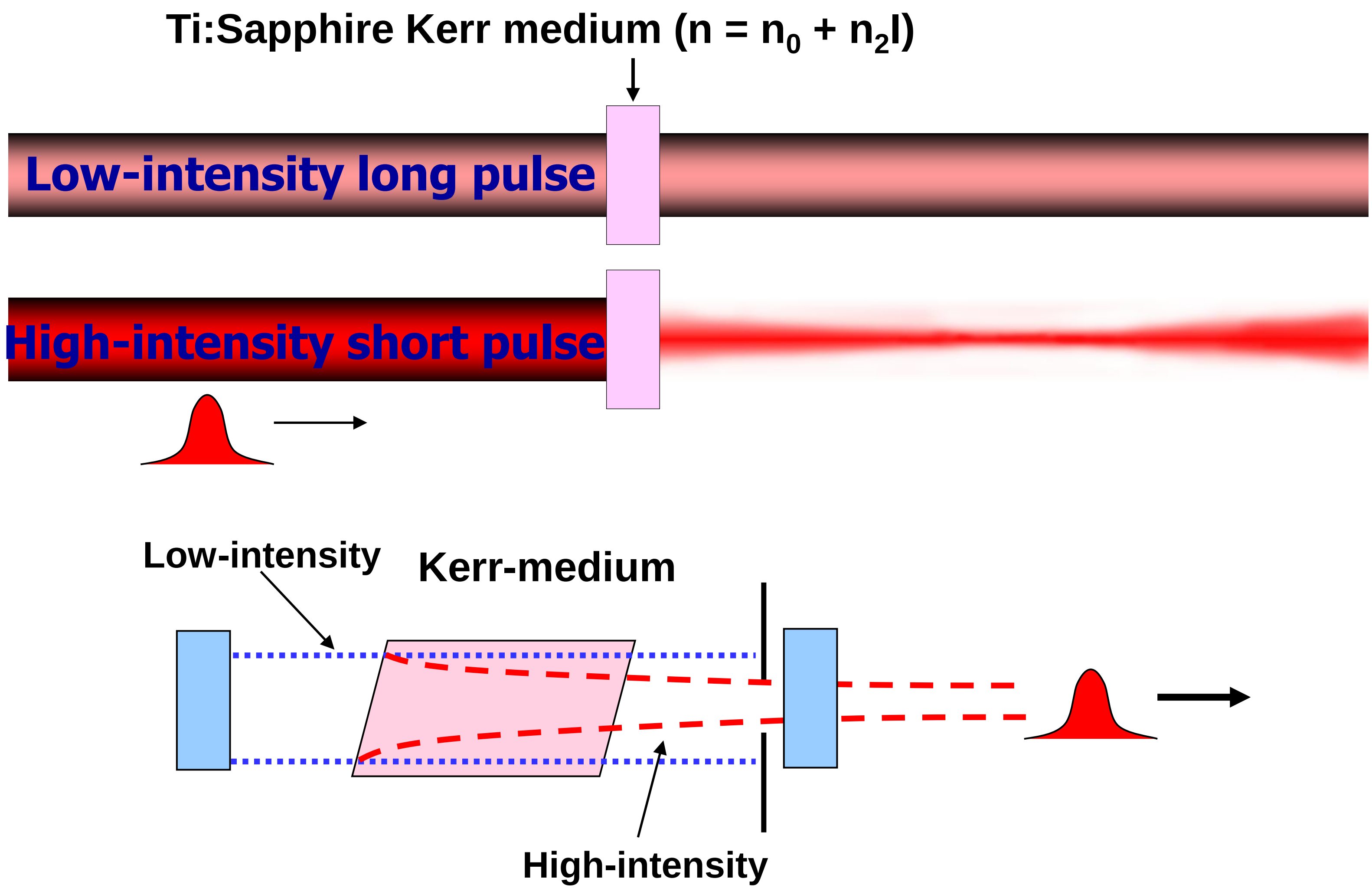






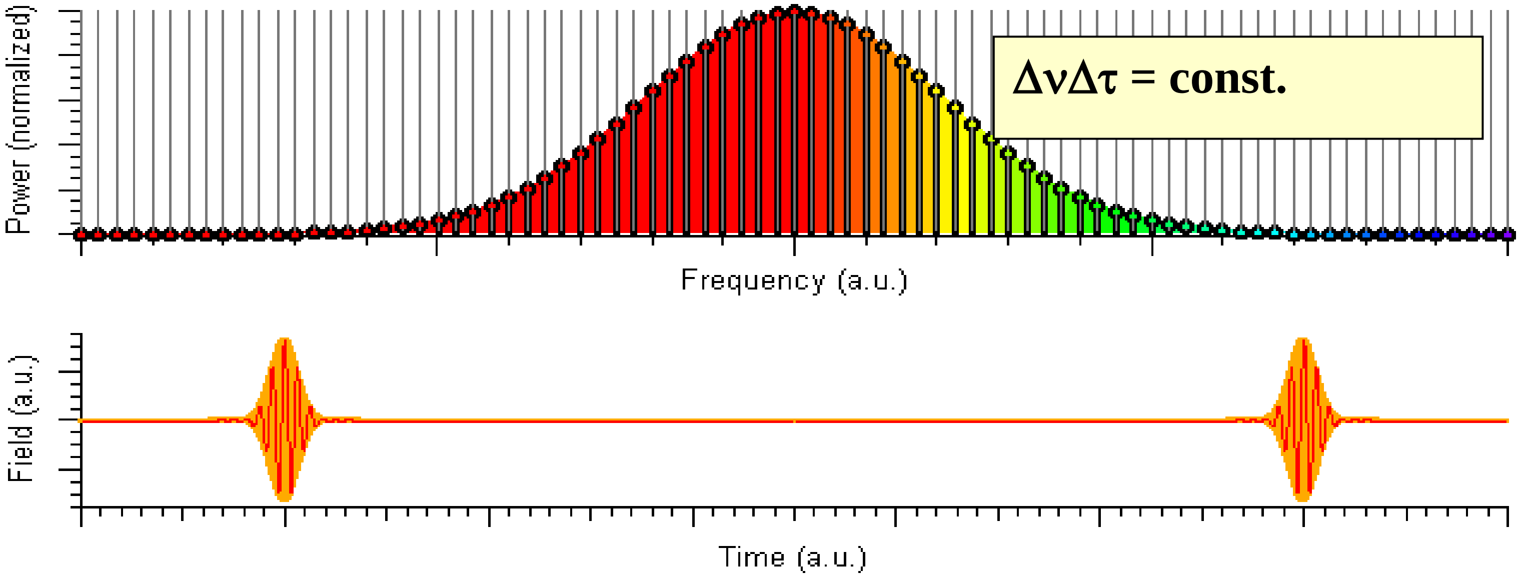
<u>Mode-locking is equivalent to a periodic shutter in the cavity with the round-trip time 2L/c and shutter speed 2L/Δnc</u>

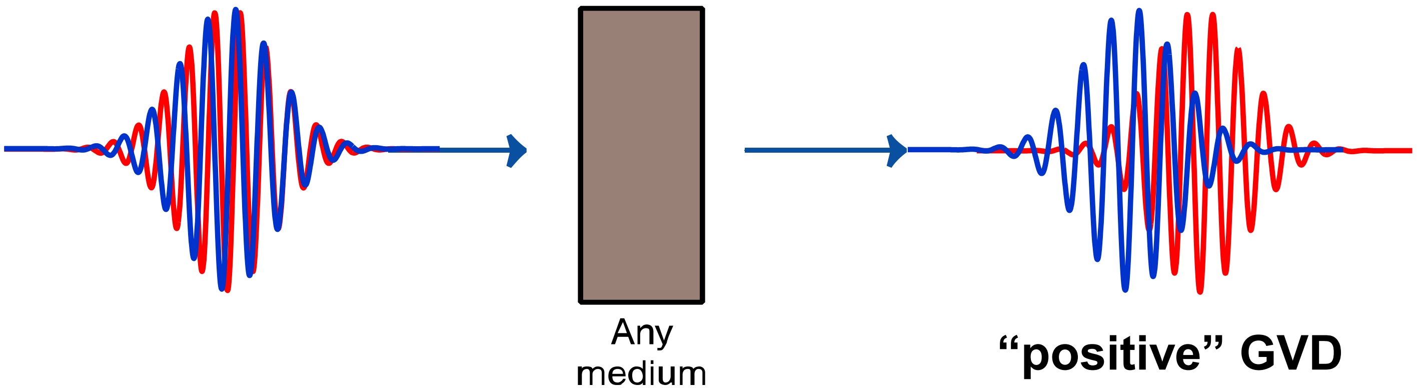
Kerr-lens mode-locking



Higher intensity short pulse is self-focused by the photoinduced lens.

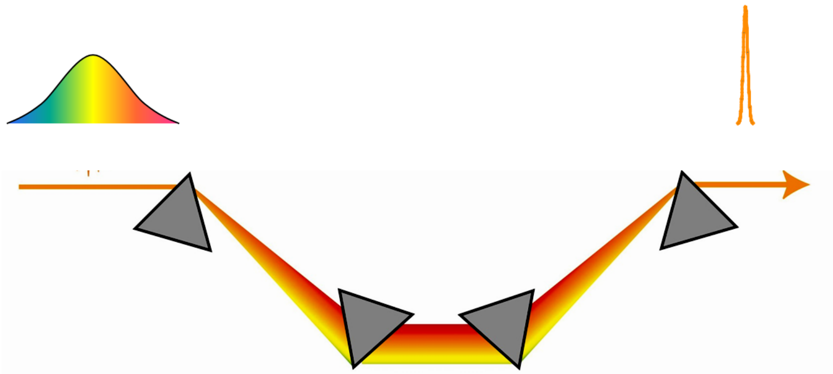
Short pulse = large bandwidth, dispersion



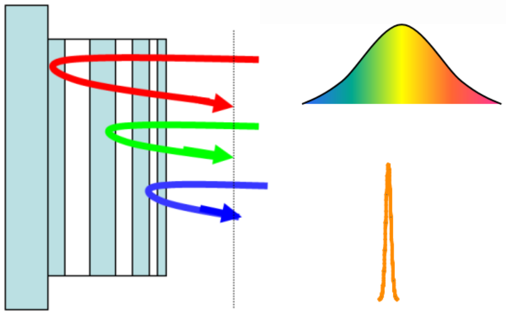


Dispersion compensation: add negative GVD

- Prism compensator

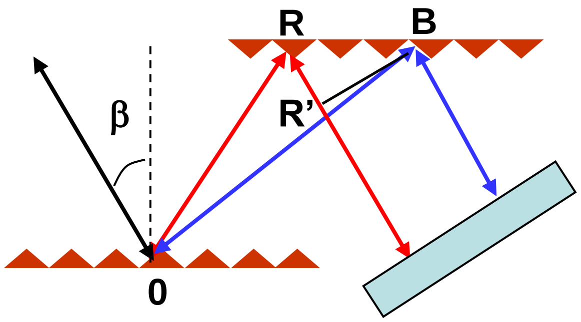


- Chirped mirror compensator

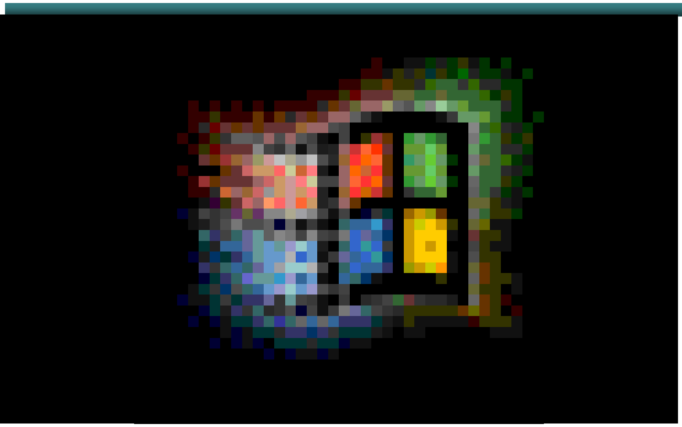


- Diffraction grating compensator

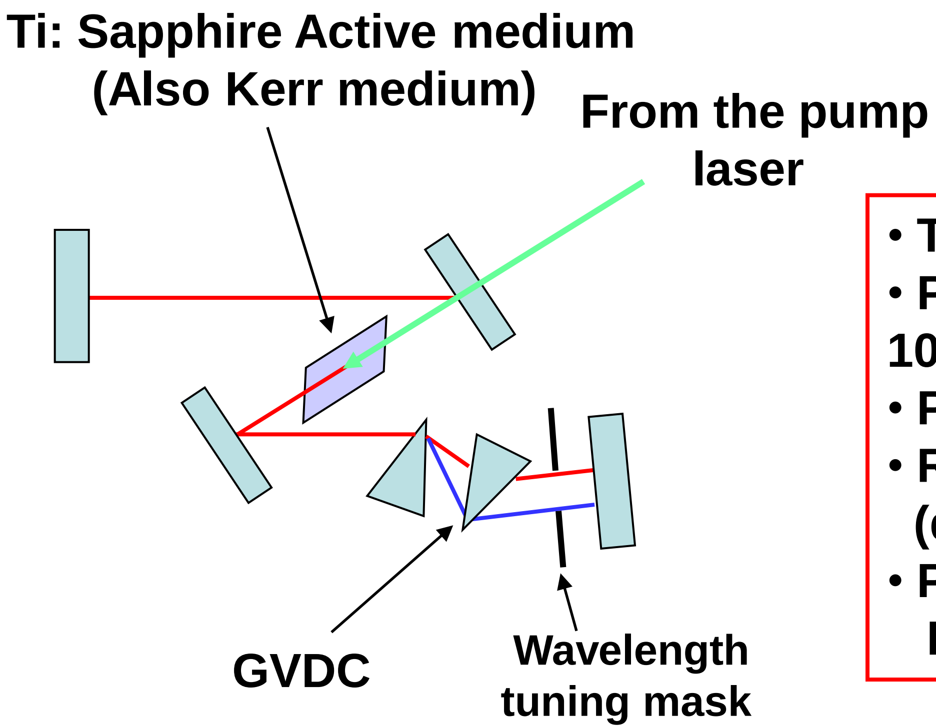
  If OR + RR' > OB,
  GVD < 0
  (can be + or - GVD



Grating Compressor

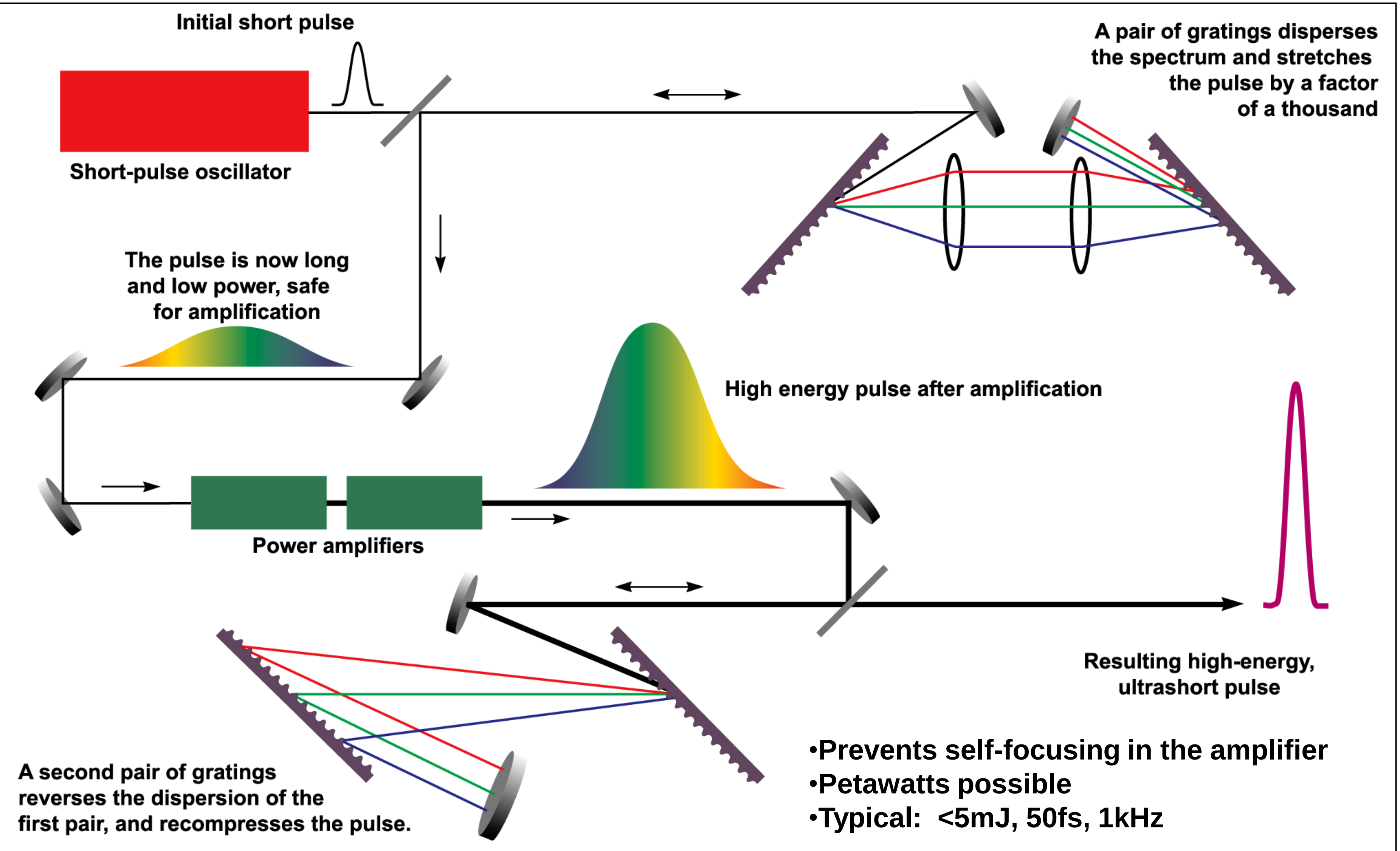


Typical fs Oscillator: KLM + GVDC = fs pulses



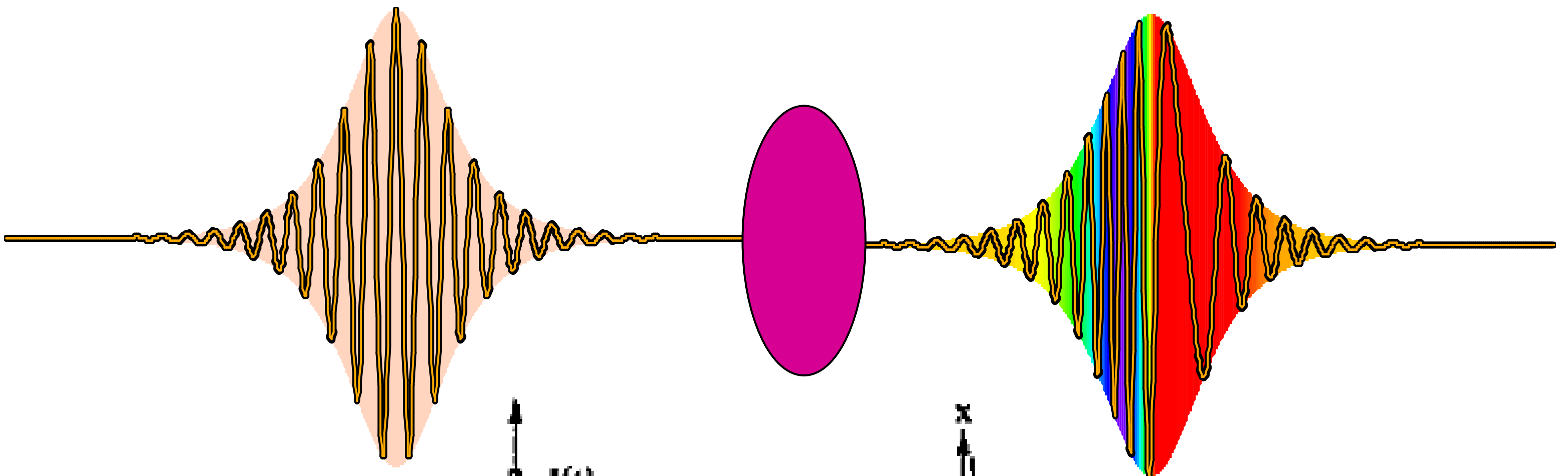
- **Tuning range 690-1050 nm**
- **Pulse duration > 10 fs (typically 50 - 100 fs)**
- **Pulse energy ~ 1 nJ**
- **Repetition rate 40 – 1000 MHz (determined by the cavity length)**
- **Pump source: DPSS CW YAG laser (532 nm, 5W)**

Chirped-pulse amplification

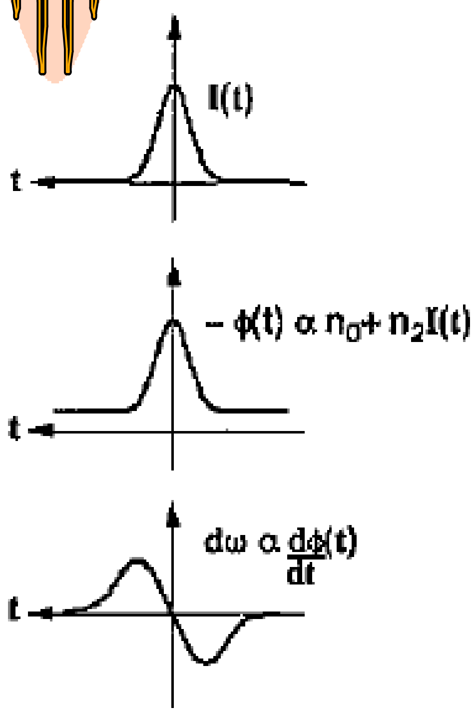


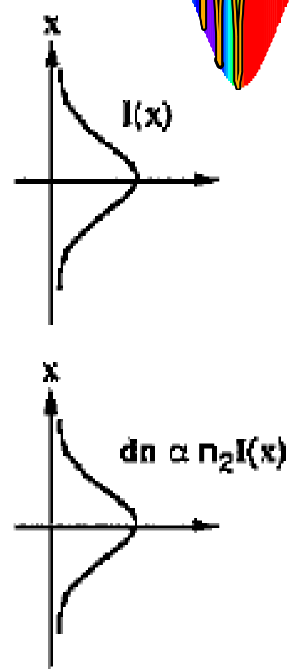
- Prevents self-focusing in the amplifier
- Petawatts possible
- Typical: <5mJ, 50fs, 1kHz

A temporal nonlinear index → self phase modulation



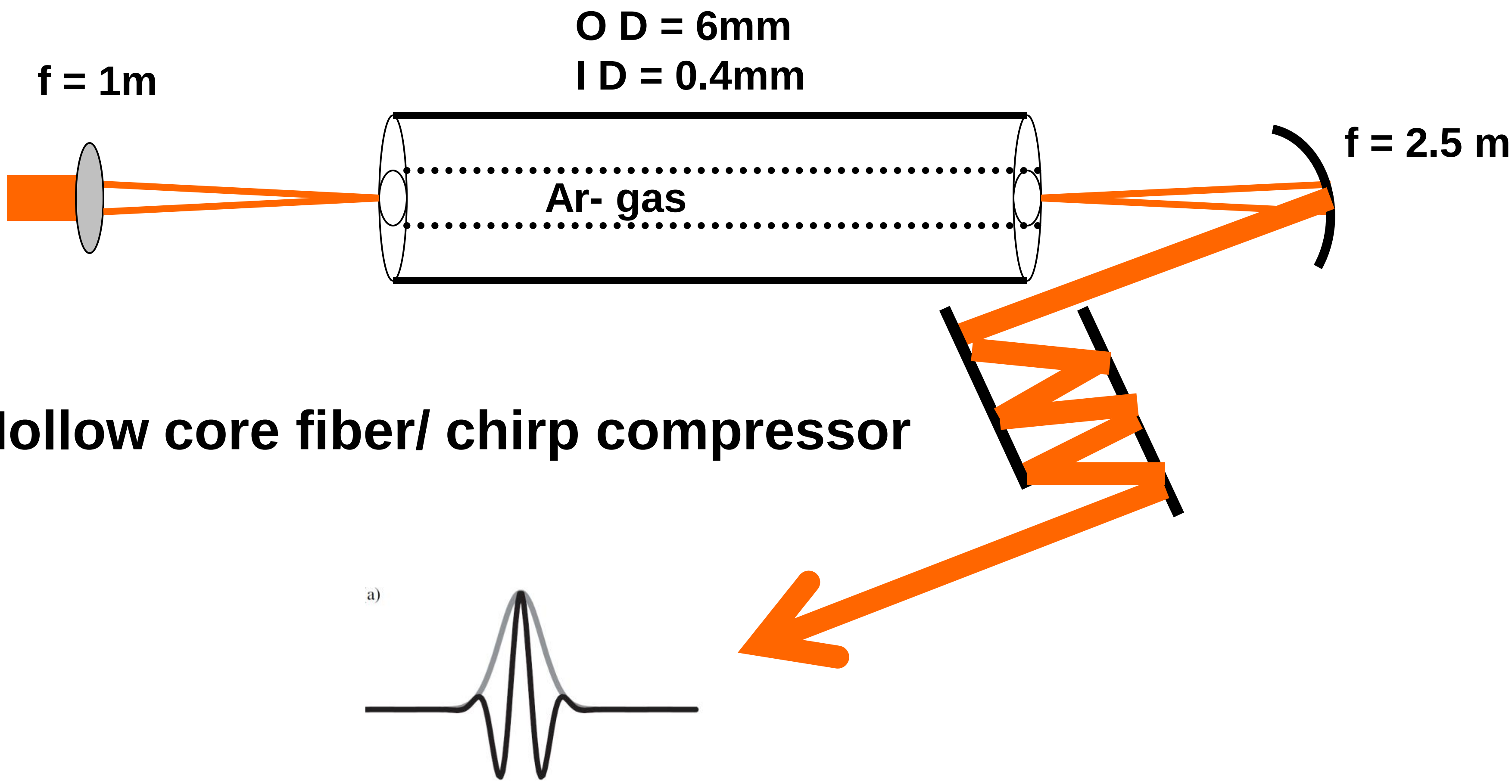
Self phase modulation





Self focusing

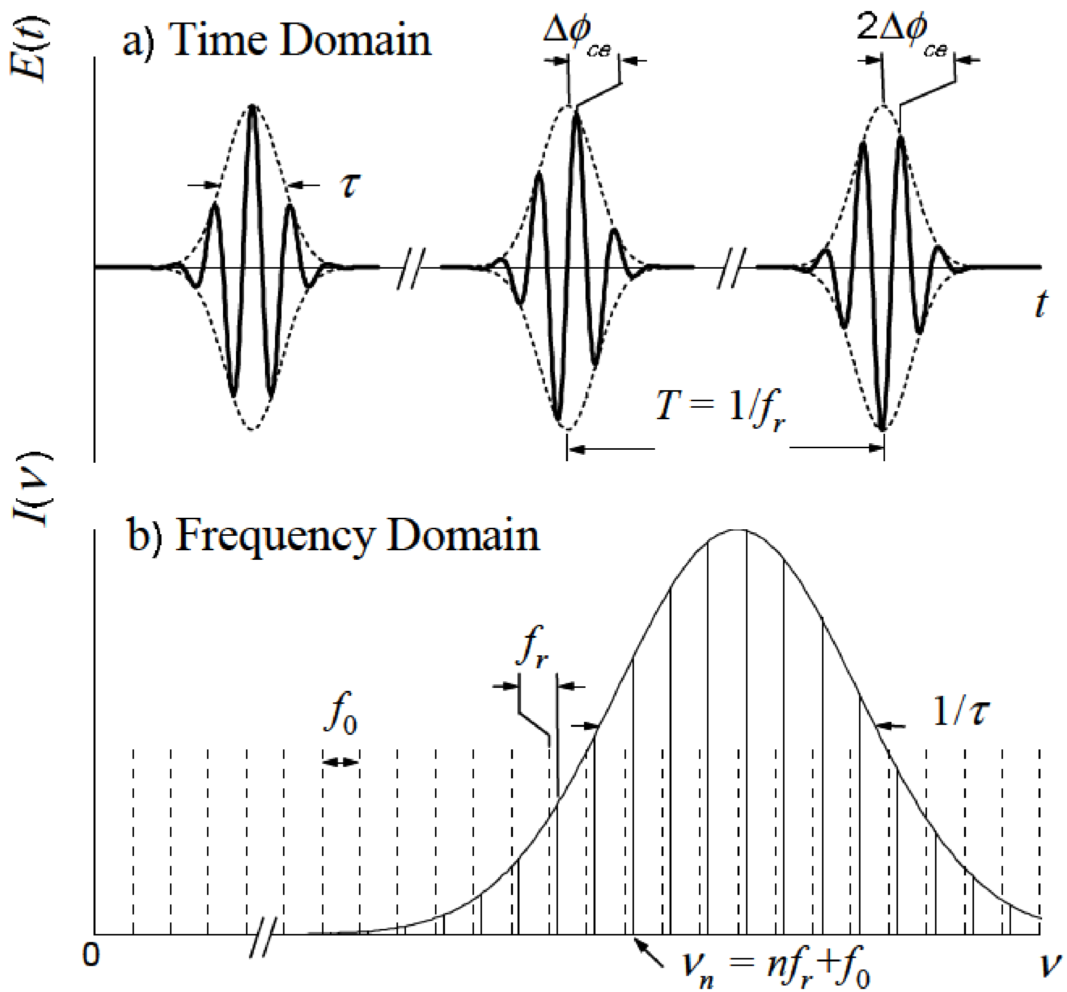
Generation of few-cycle pulses



Hollow core fiber/ chirp compressor

Carrier envelope offset

Optical Frequency Combs and their Applications, Jun Ye and Steven T. Cundiff,



- Stable only when the round trip phase is 2πn
- Phase shift can be measured and stabilized with an "f-2f" interferometer
- Stabilization leads to a direct link between rf and optical metrology, and a Nobel Prize for Hall and Haensch.
- Sub-femtosecond timing is therefore possible.

Measuring an optical pulse

- **All the information is in the electric field:**

$$\mathcal{E}(t) = \int_{-\infty}^{+\infty} \tilde{\mathcal{E}}(\omega)\exp(-\mathrm{i}\omega t)\frac{\mathrm{d}\omega}{2\pi}$$

$\mathcal{E}(t)$ **is a real function of all time**

$\tilde{\mathcal{E}}(\omega)$ **is the spectrum, and is generally a complex function**

Either is a complete description.

Added wrinkle: The field is a vector quantity because of polarization

Spectrograms and Wigner distributions

Wigner representation:

$$W(t,\omega) = \int E\left(t + \frac{t'}{2}\right) E^*\left(t - \frac{t'}{2}\right) e^{i\omega t'} dt'$$

$$= \frac{1}{2\pi} \int \tilde{E}\left(\omega + \frac{\omega'}{2}\right) \tilde{E}^*\left(\omega - \frac{\omega'}{2}\right) e^{-i\omega' t} d\omega'$$

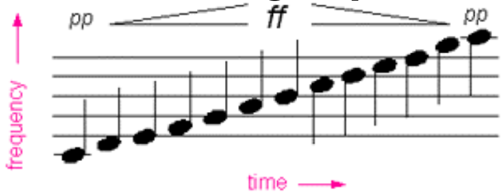
$$E(t)E^*(0) = \int W(t/2,\omega)\exp(i\omega t)d\omega$$

- **Complete description of the pulse up to global phase**
- **Real 2-D function (but can be negative)**

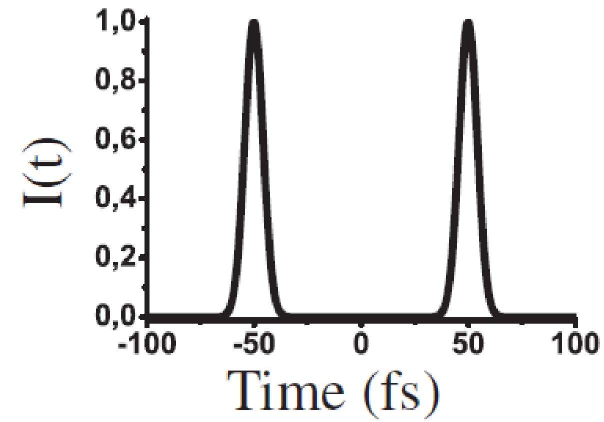
Spectrogram (Husimi) representation:

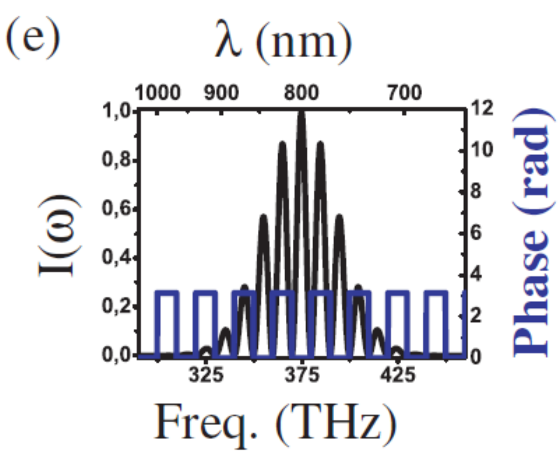
$$S(t,\omega) = \left| \int E(t') g(t'-t) e^{i\omega t'} dt' \right|^2$$

- **Incomplete: information lost by convolution integral**
- **Real and positive, the equivalent of a musical score for the light pulse**

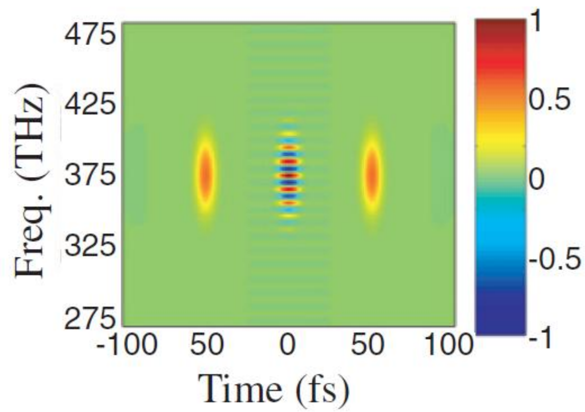


Example: two-pulse train

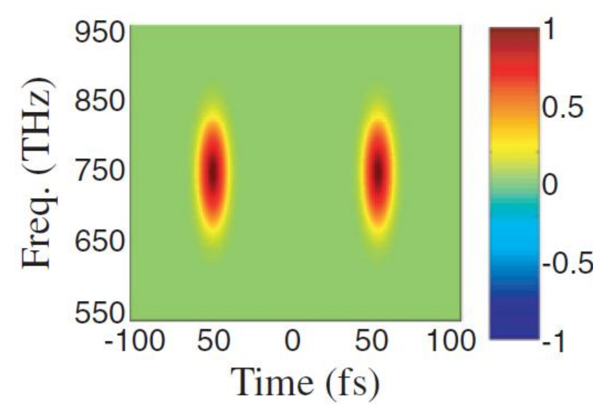




$\tilde{\mathcal{E}}(\omega)$ Amplitude and phase

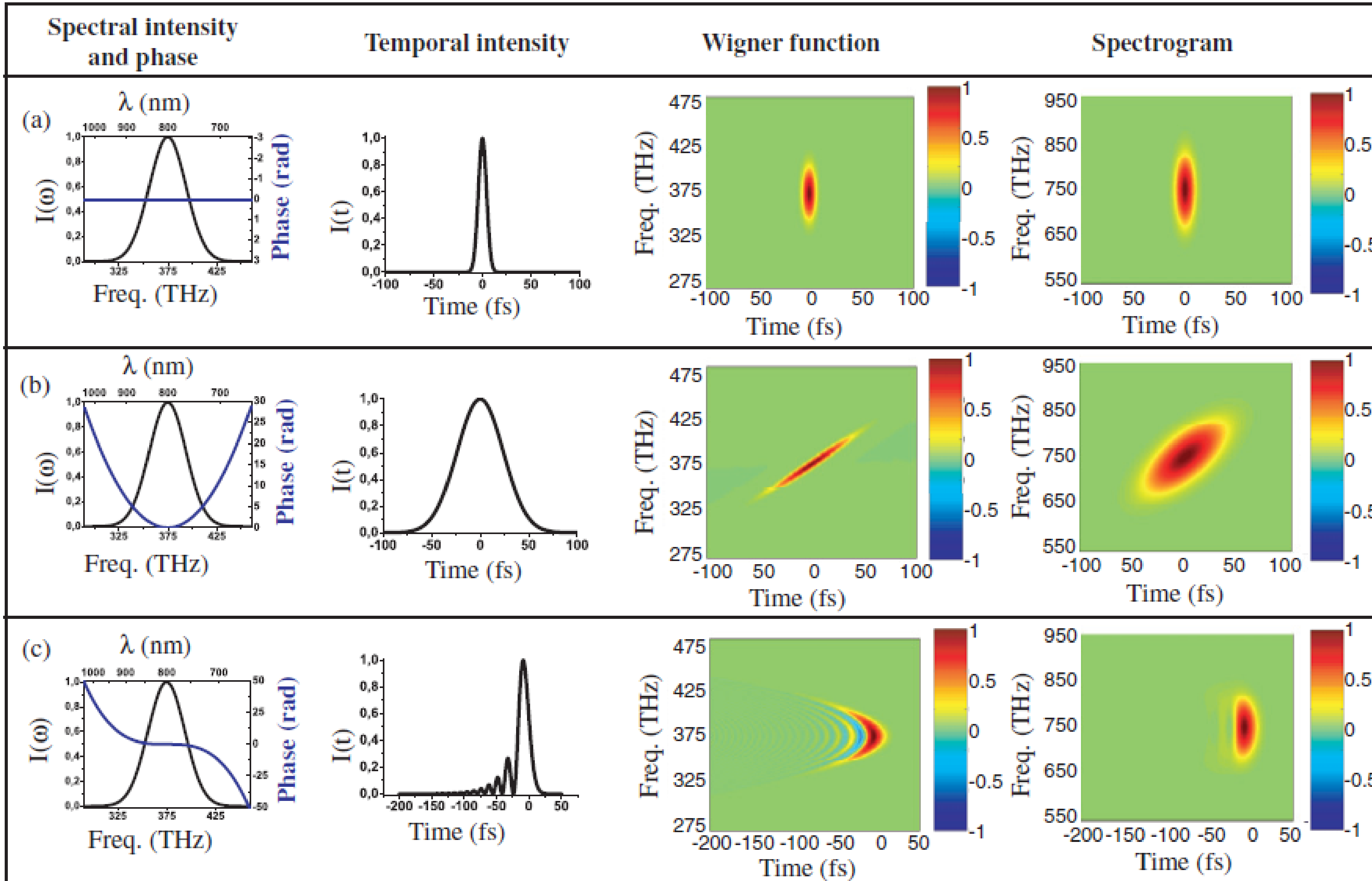


$W(t,\omega)$



$S(t,\omega)$

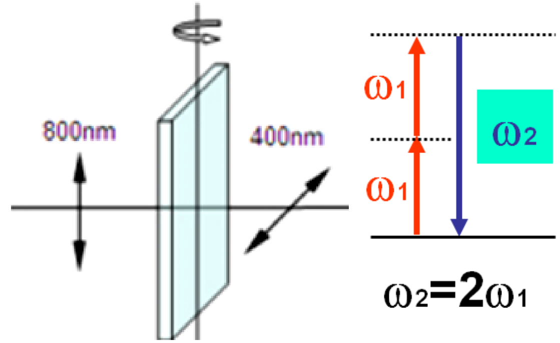
Important examples

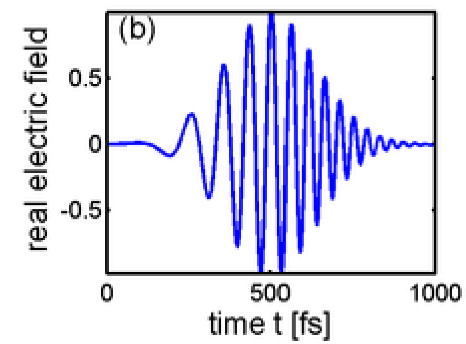


Using the pulse to measure itself: Autocorrelation using a doubler

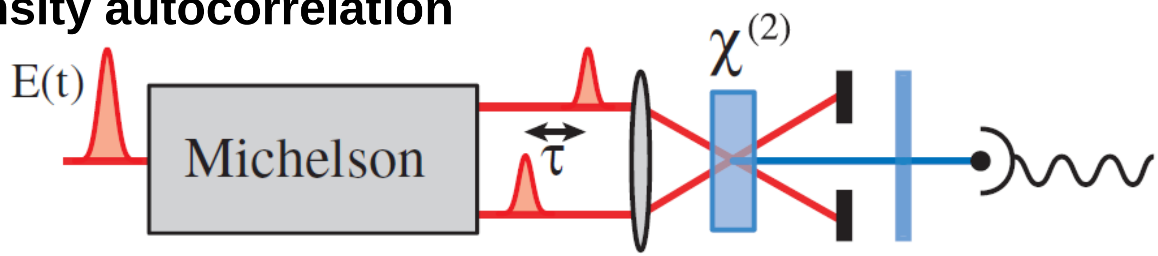
Second harmonic generation:

$$E_2(t,\tau) \propto E(t)E(t-\tau)$$

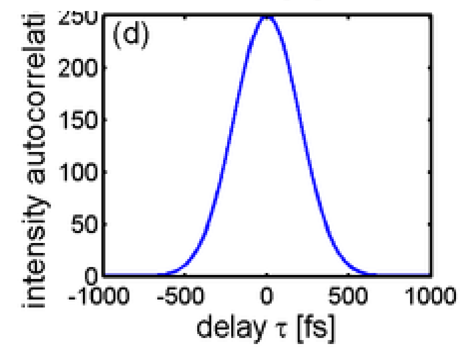




Intensity autocorrelation

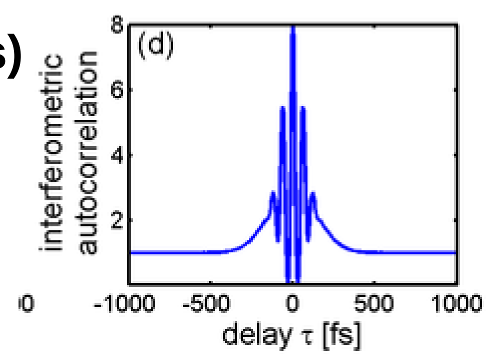


$$S(\tau) \propto \int |E_2(t,\tau)|^2 \, dt \propto \int I(t)I(t-\tau) \, dt$$



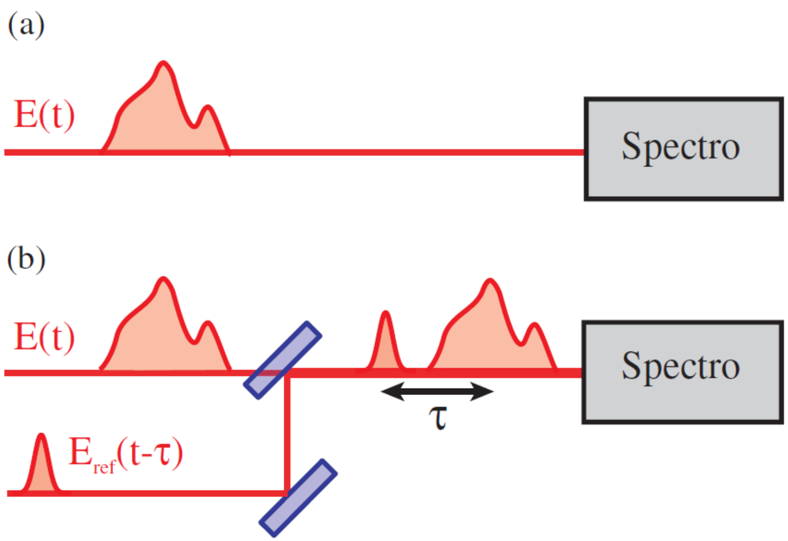
Interferometric nonlinear autocorrelation (collinear beams)

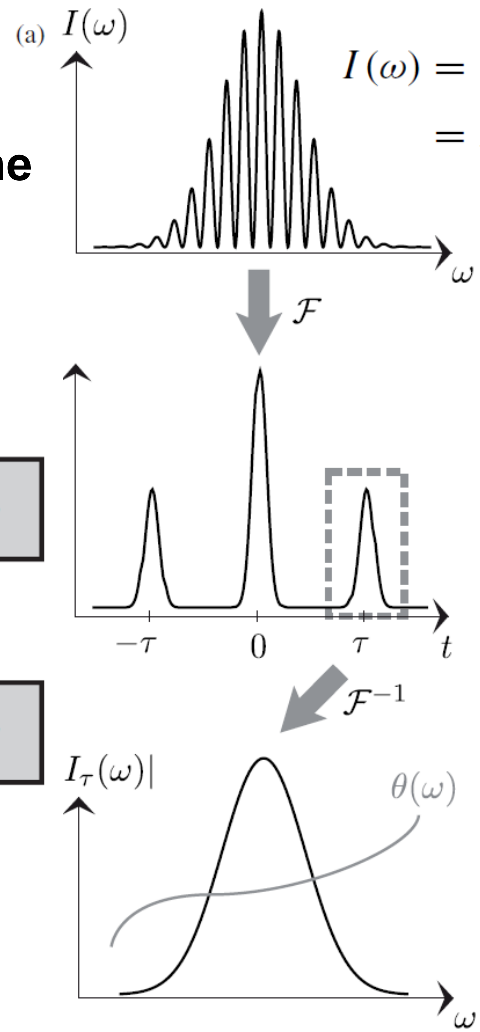
$$S(\tau) \propto \int |E(t) + E(t-\tau)|^4 \, dt$$



Cross correlation techniques

- **Uses reference pulse**
- **Reference must be flat-phase (“transform limited”) and cover the spectrum needed**
- **Full phase retrieval is possible**

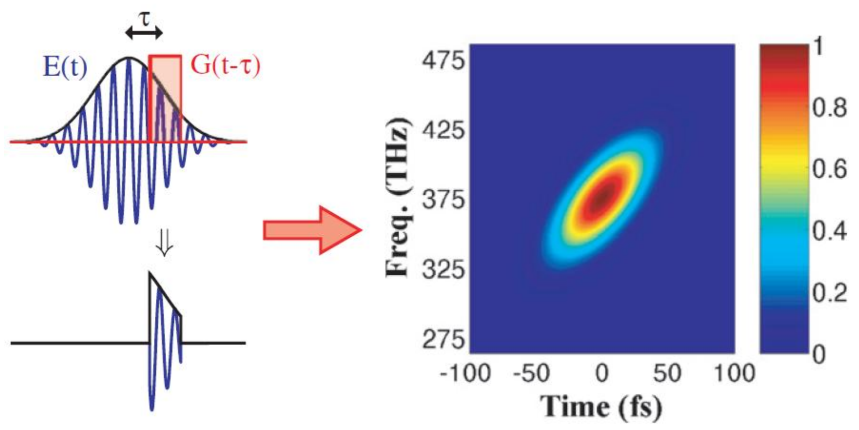


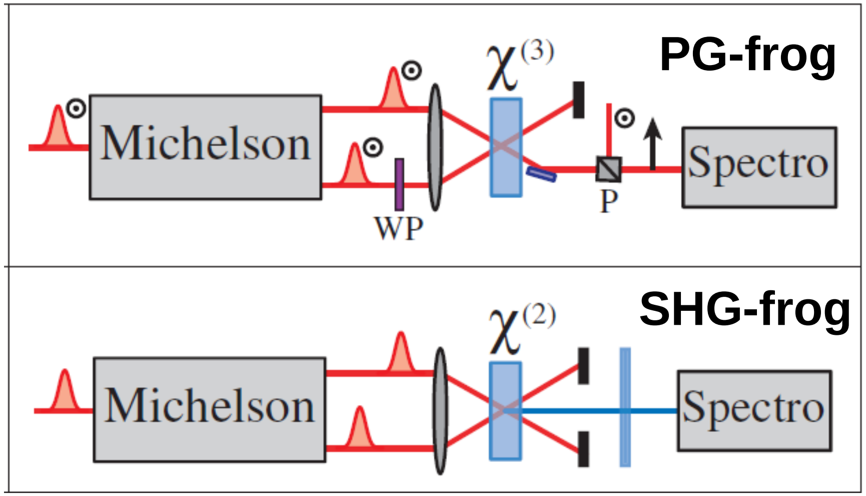


$$I(\omega) = \left|\tilde{E}(\omega) + \tilde{E}_{\mathrm{ref}}(\omega)\, e^{i\omega\tau}\right|^2$$
$$= A^2(\omega) + A^2_{\mathrm{ref}}(\omega) + 2A(\omega)A_{\mathrm{ref}}(\omega)$$
$$\times \cos\left[\varphi(\omega) - \varphi_{\mathrm{ref}}(\omega) - \omega\tau\right].$$

$$I_\tau(\omega) = A(\omega)A_{\mathrm{ref}}(\omega)\exp[i(\varphi(\omega) - \varphi_{\mathrm{ref}}(\omega) - \omega\tau)]$$

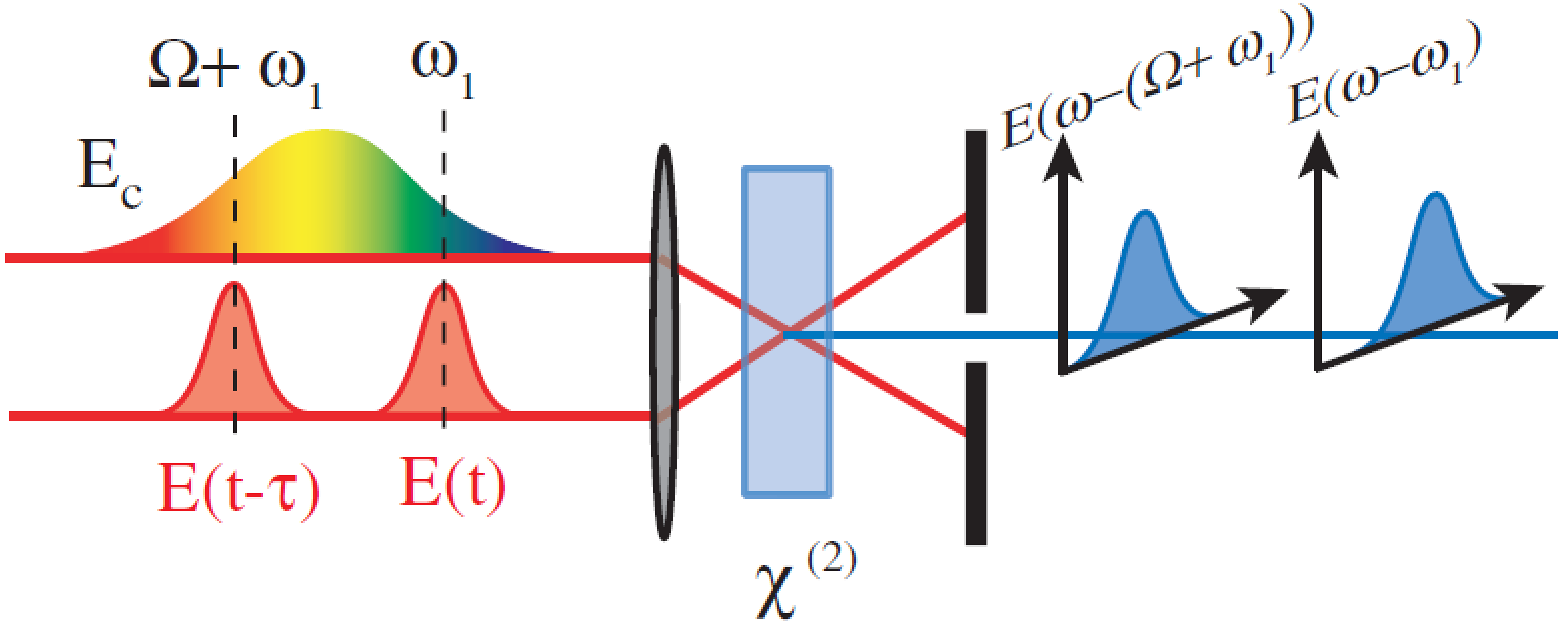
Frequency-resolved optical gate (FROG)





- Spectrally resolved cross correlation
- Gate pulse need not have spectral overlap
- Requires phase retrieval algorithm
- Full phase retrieval is possible

Spider

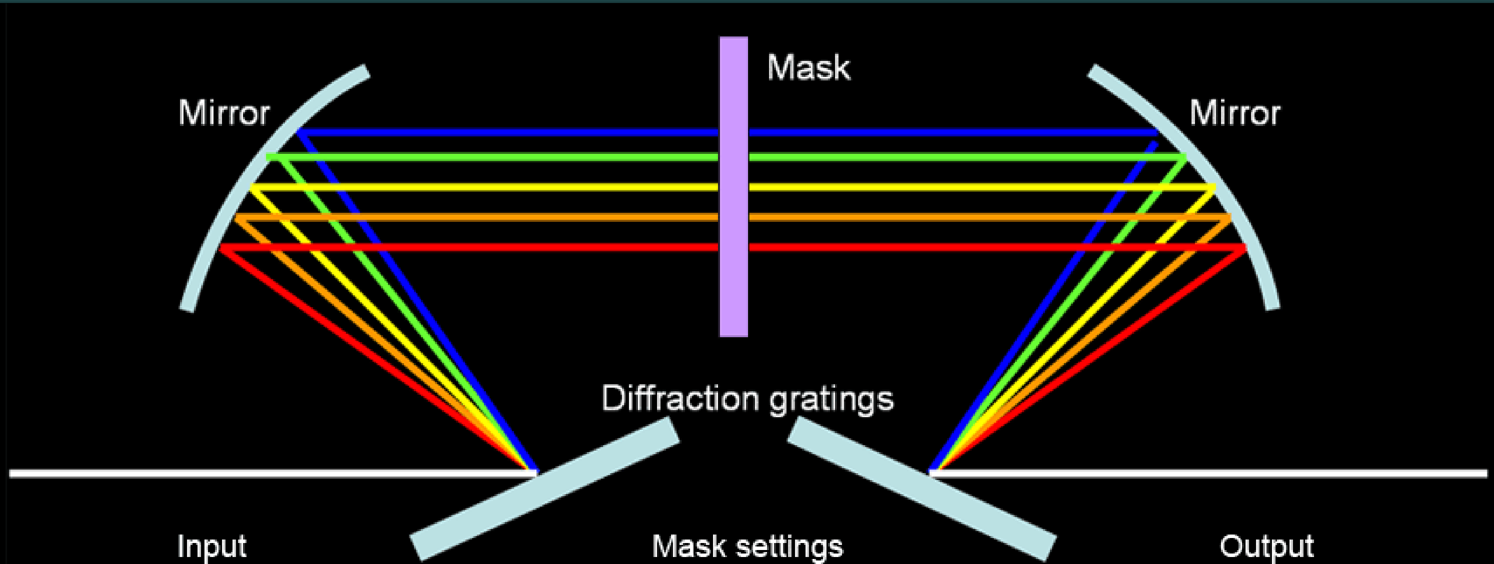


Spectral interferometry of upconverted pulse at two separated upconversion wavelengths

Direct phase retrieval without need for iterative algorithm

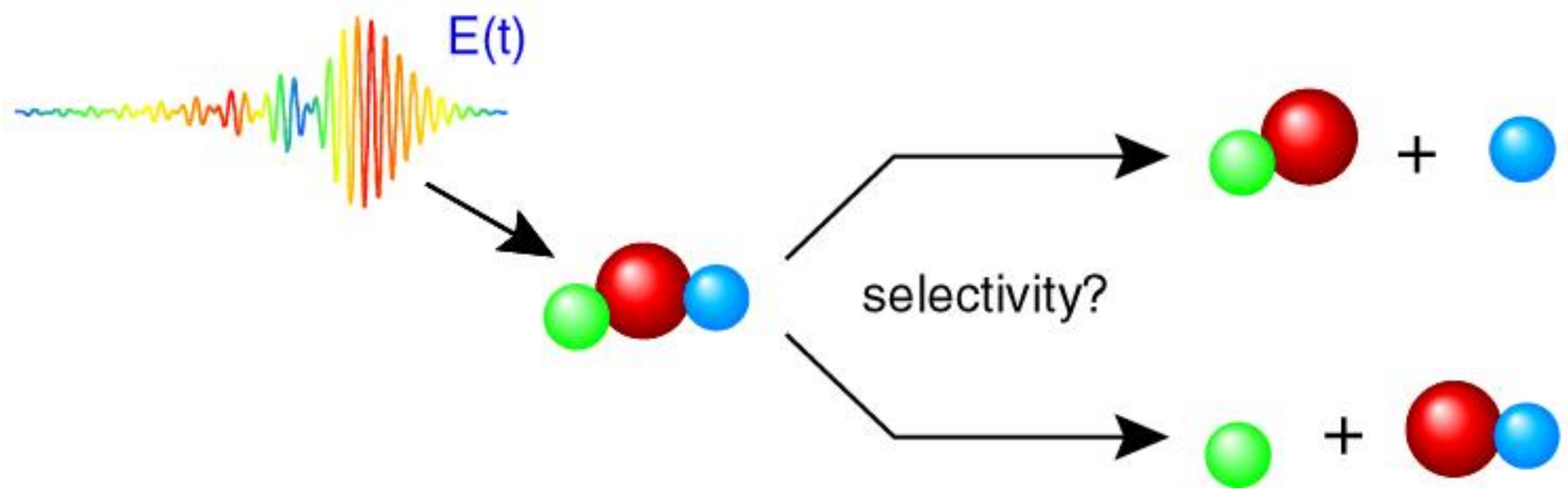
No need for a reference at the same wavelength as the signal

Pulse Shaper



Beyond ultrafast spectroscopy: controlling chemical reactions with ultrashort pulses

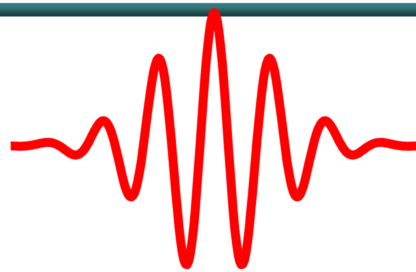
You can excite a chemical bond with the right wavelength, but the energy redistributes all around the molecule rapidly (“IVR”).

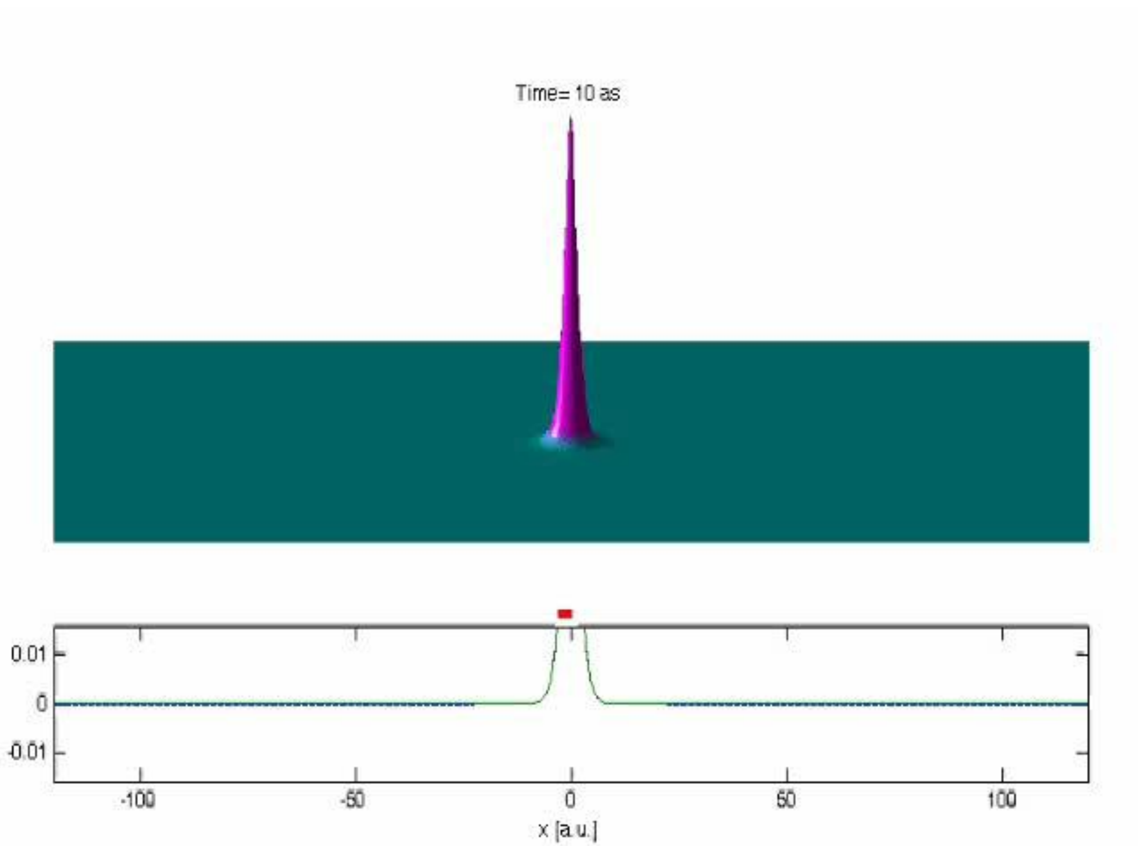


But exciting with an intense, shaped ultrashort pulse can control the molecule’s vibrations and produce the desired products.

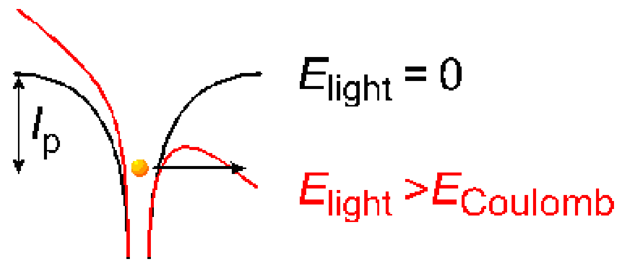
Extreme nonlinear conversion: ATI and HHG

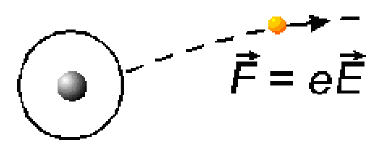
[Corkum, Kulander]





Laser-induced field ionization

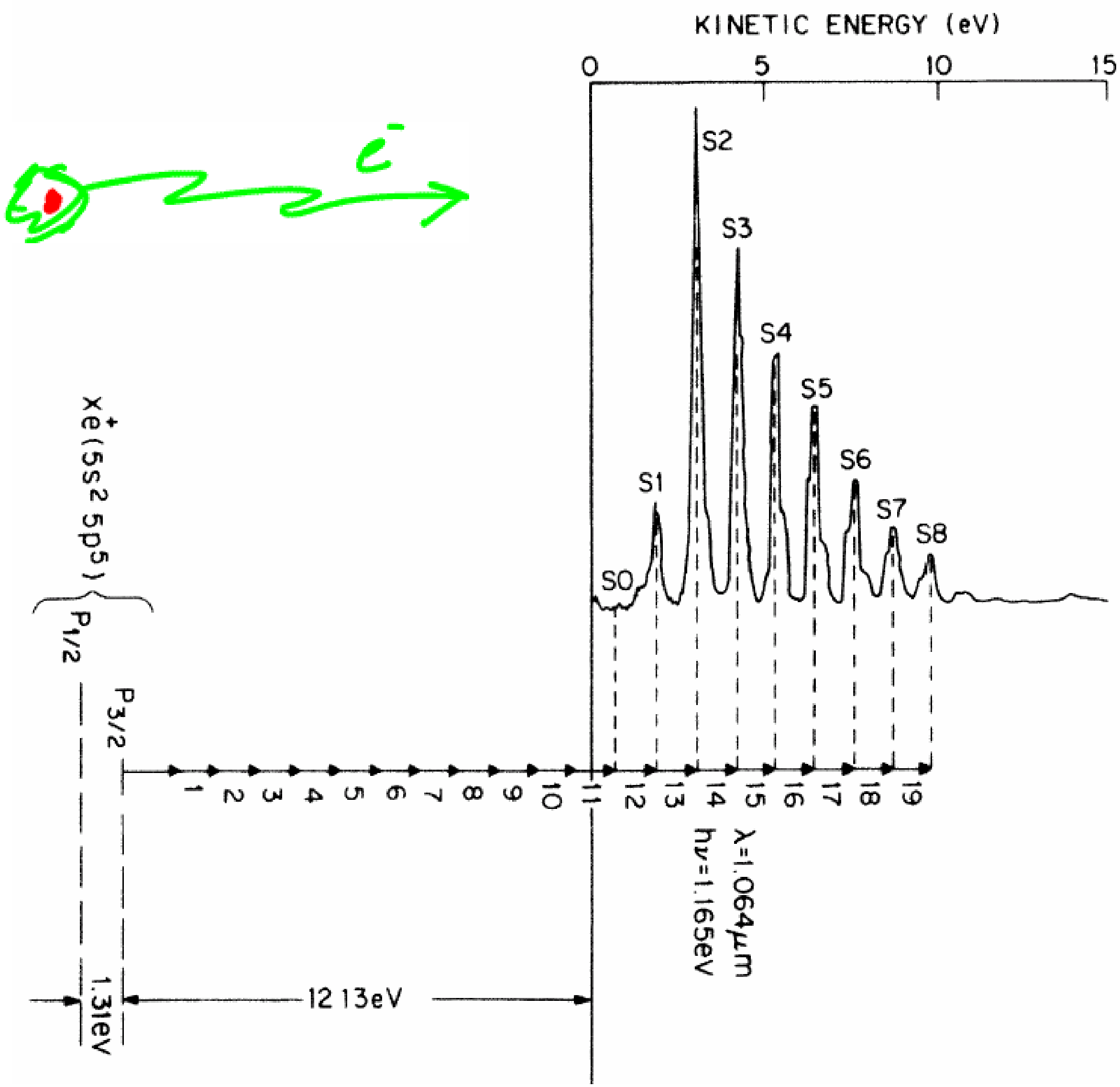




$$F_c = \frac{(I.P.)^2}{4} = 2.2 \times 10^8 v/cm \text{ for H}$$

$$I_c = \frac{I_a}{16^2} = 1.4 \times 10^{14} W/cm^2$$

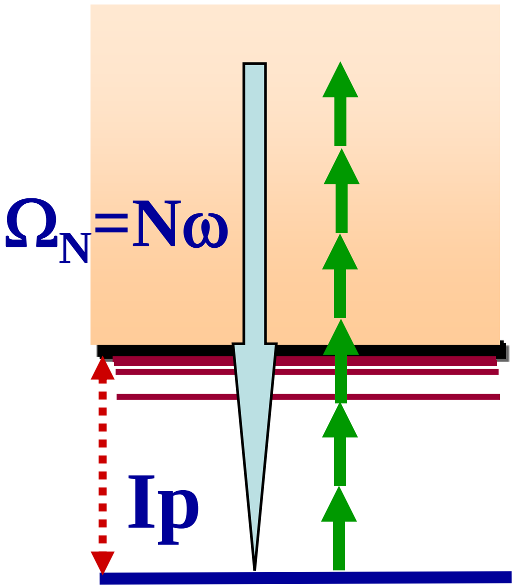
ATI: Tunnel ionization, wiggling electrons

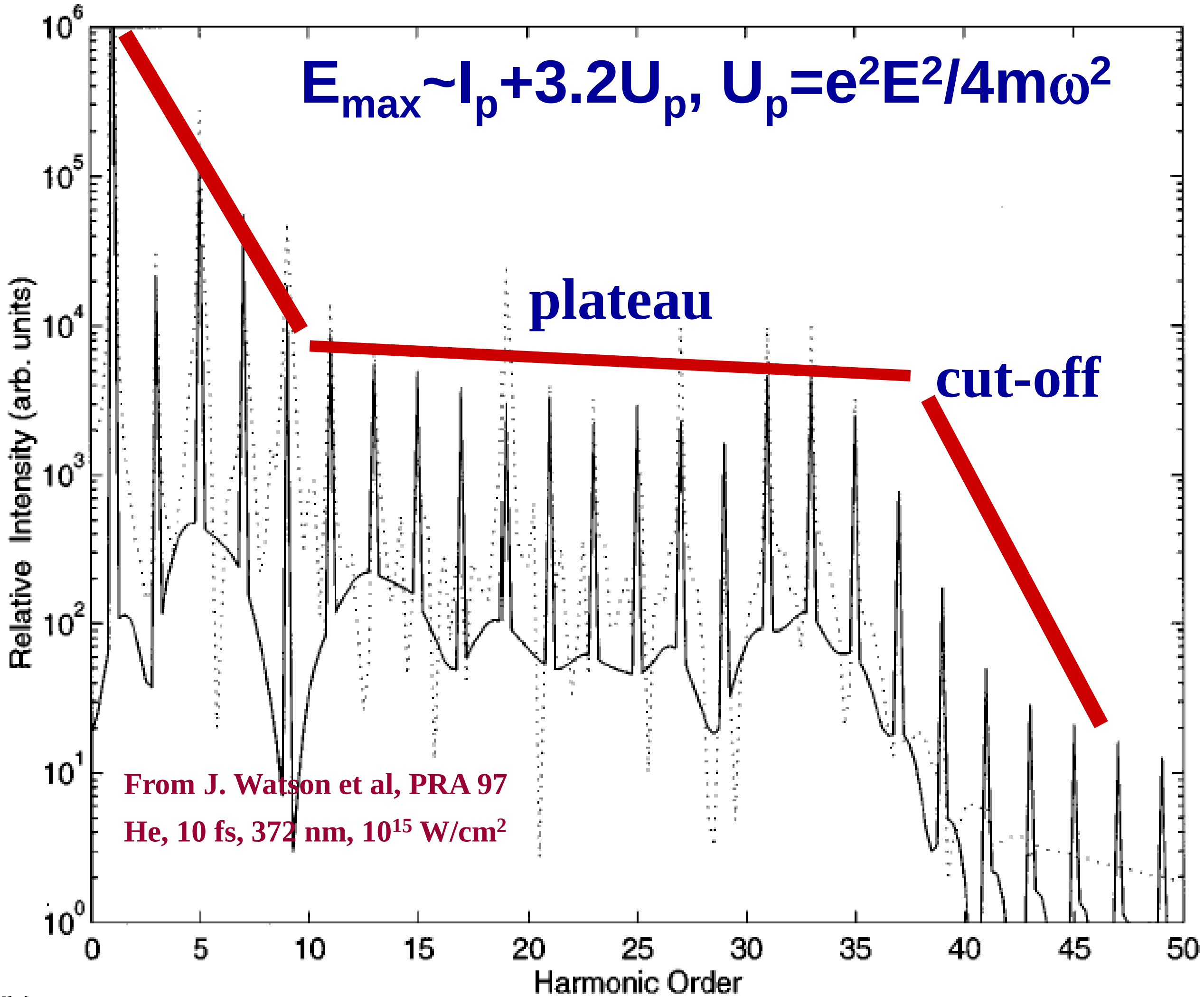


Wiggle energy

$U_p = e^2 E^2 / 4 m \omega^2$

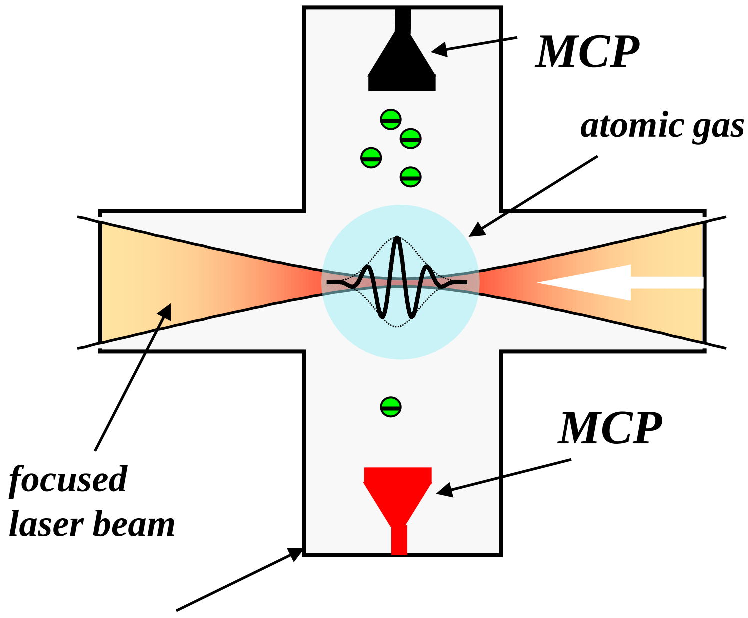
HHG: recombination

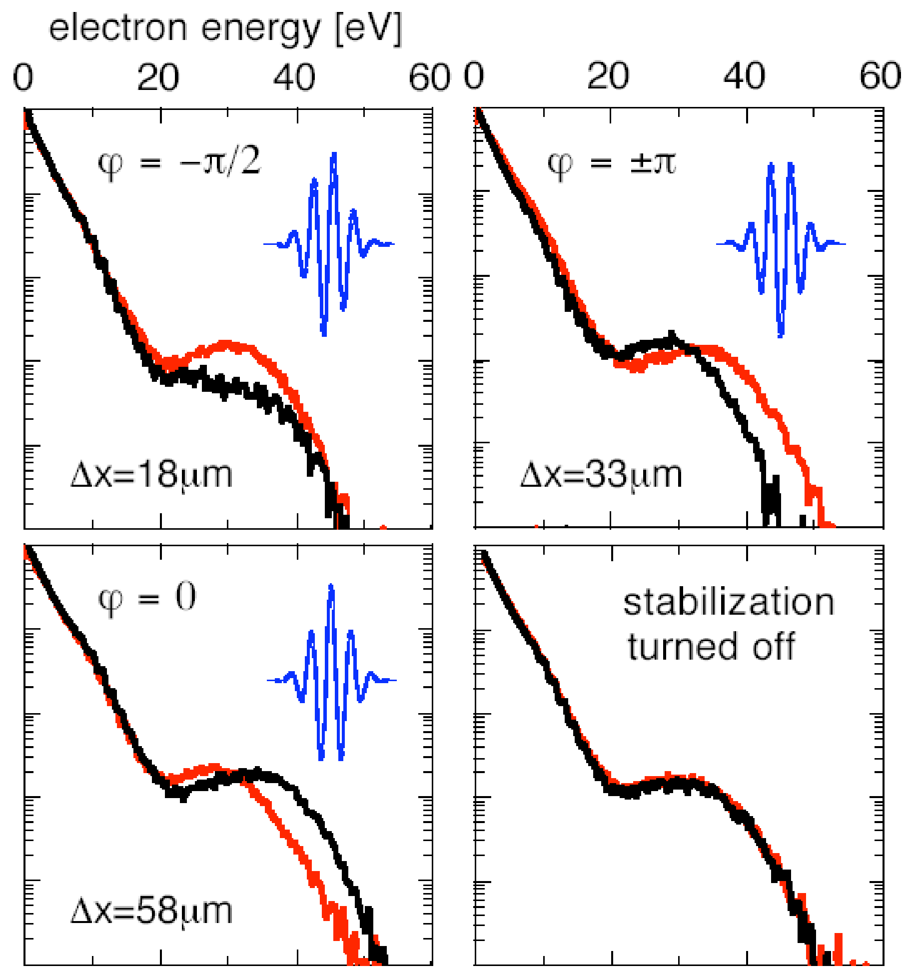




Ivanov, NRC

ATI is sensitive to carrier phase

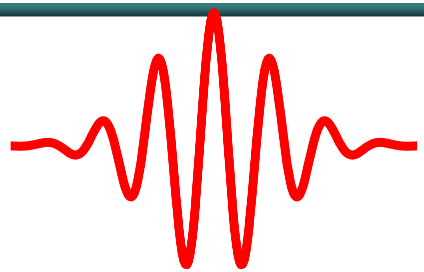




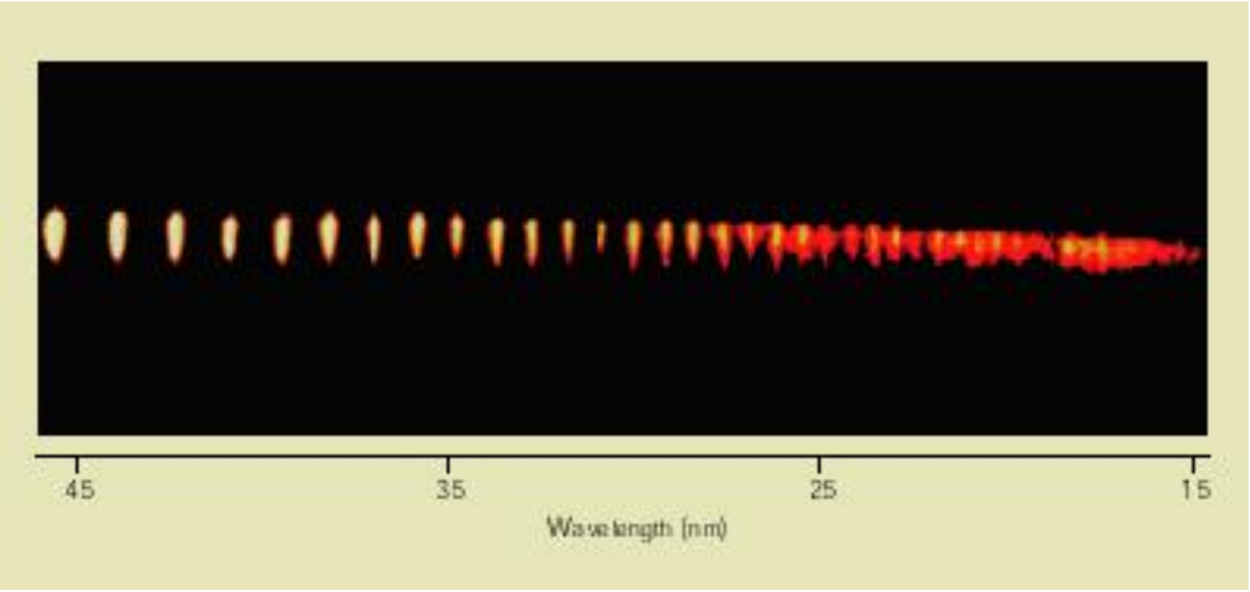
Paulus *et al*. Nature, 414 182 (2001)

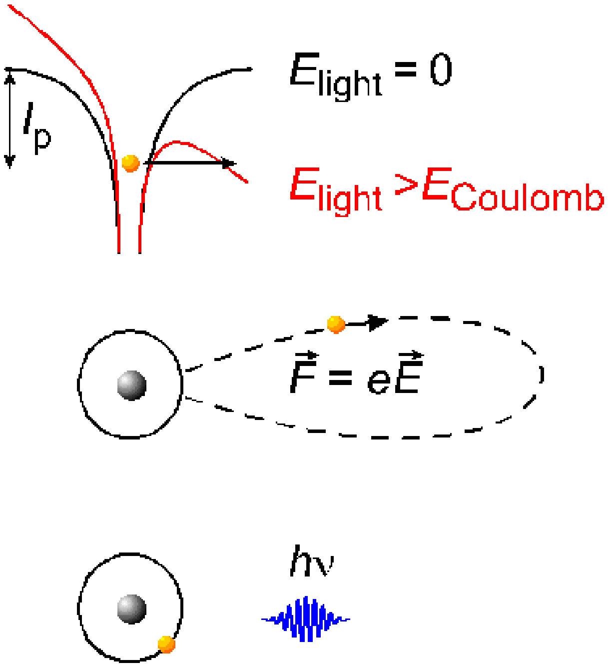
Acknowledgement: R. Kienberger

The spectra are high harmonics.



Laser-induced field ionization





RABBITT: measurement of the relative phase of adjacent harmonics

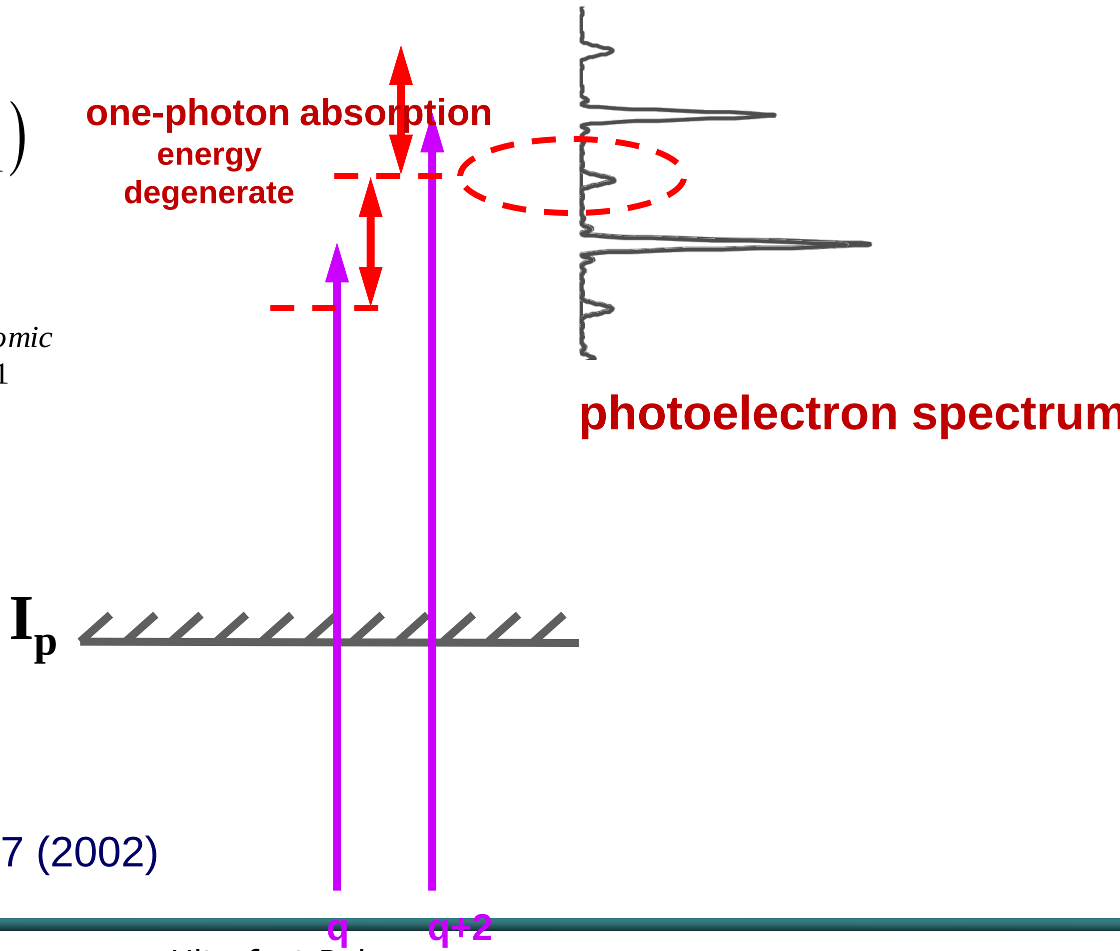
Intense optical field absorption & emission

sideband signal:

$$I_{q+1}^{SB} \propto \cos\left(2\omega_{IR}\tau - \Phi_{q+1}^{SB}\right)$$

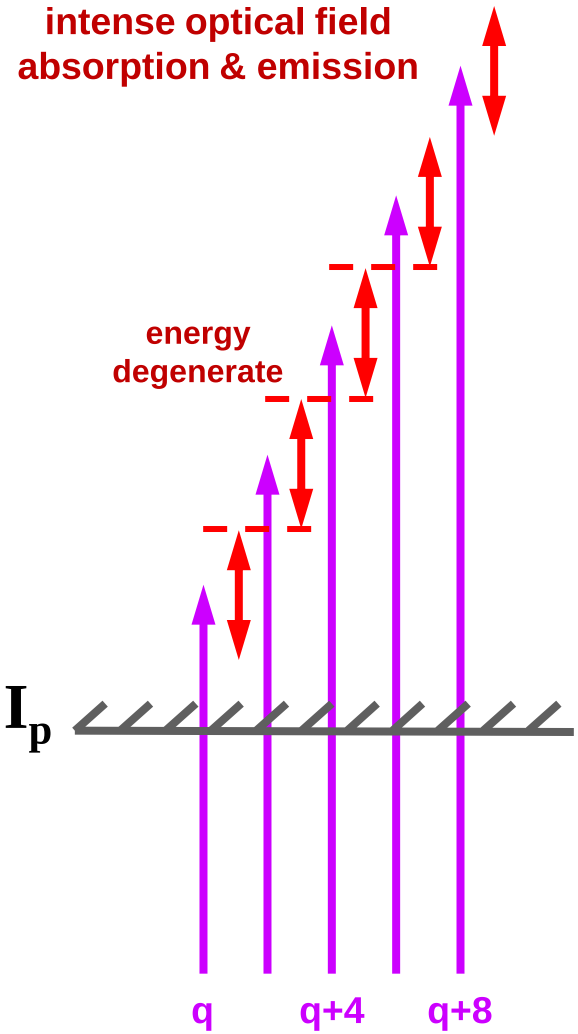
sideband phase:

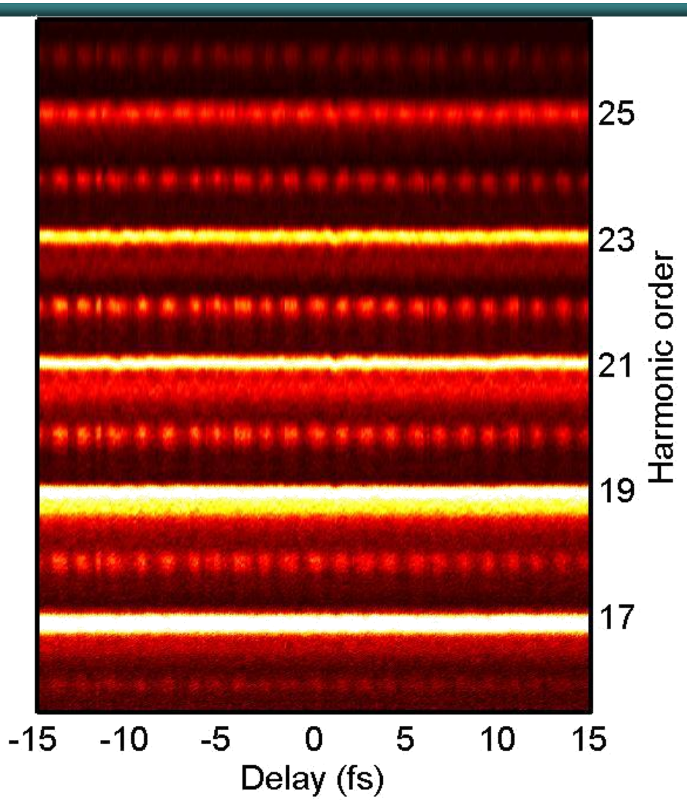
$$\Phi_{q+1}^{SB} = \underbrace{\varphi_{q+2} - \varphi_q}_{\Delta\varphi_{q+1}} - \Delta\varphi_{q+1}^{atomic}$$



H. Muller, *Appl. Phys. B,* 74, S17 (2002)

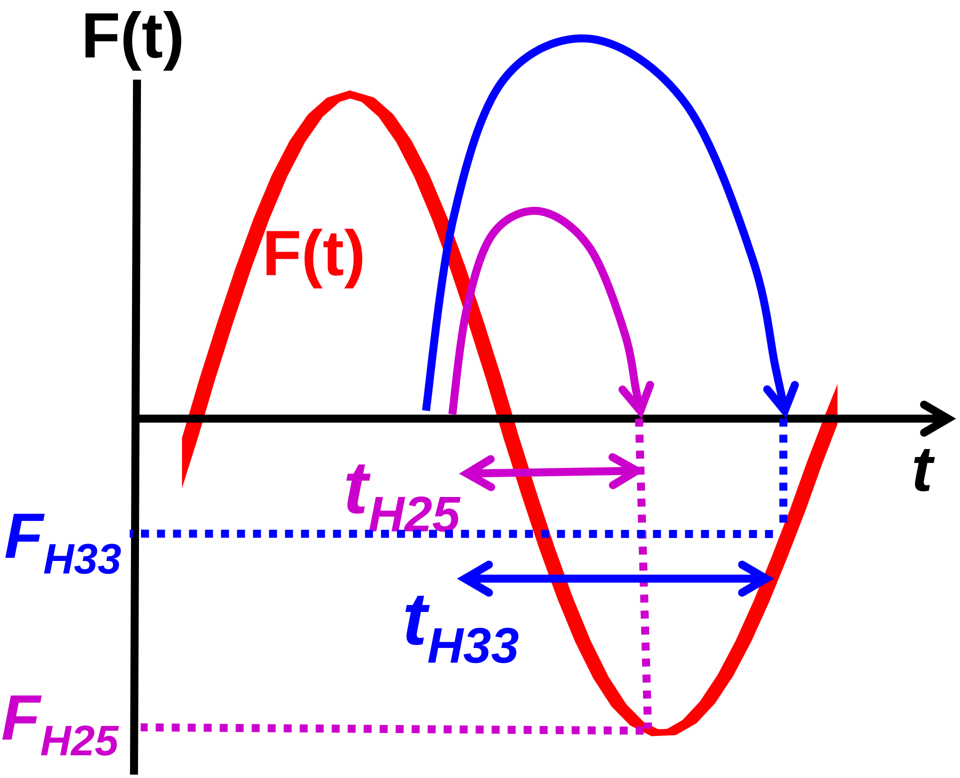
RABBITT: measurement of the relative phase of adjacent harmonic

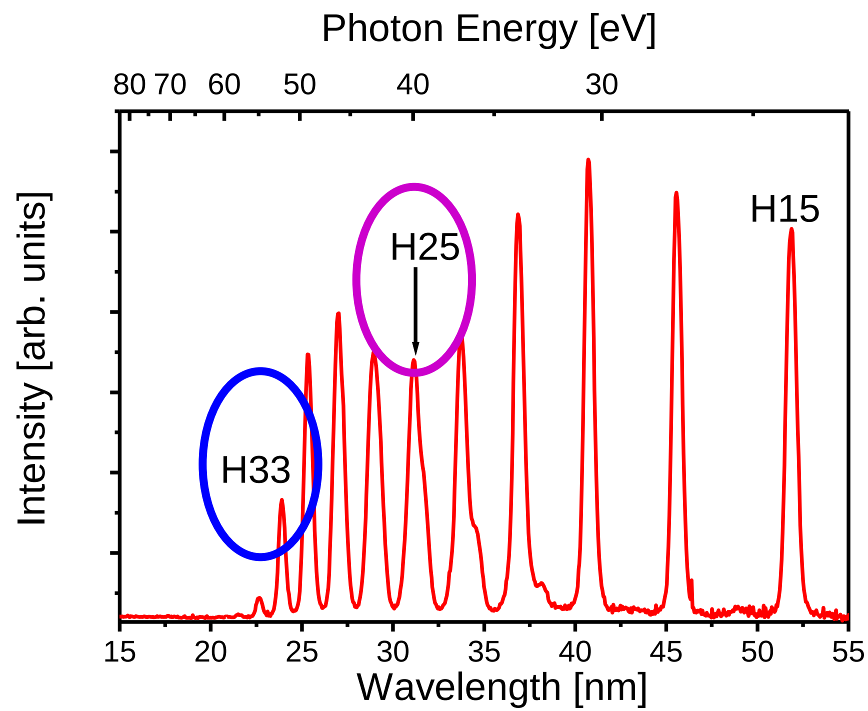




Y. Mairesse *et al.*, Science 302, 1540 (2003)
P. Johnsson *et al.*, PRL 95, 013001 (2005)

Each harmonic has its own return time and return field ("short trajectories" shown)

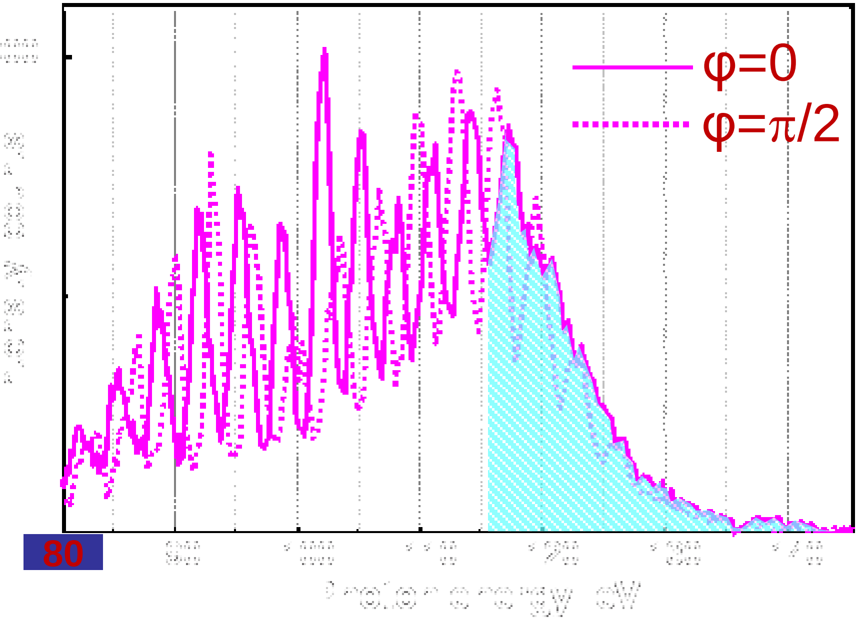




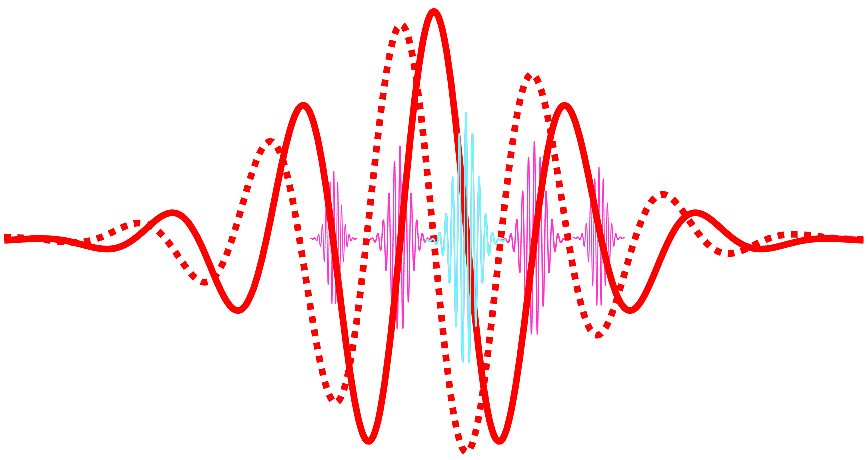
Emission from Strongly-Driven Atoms

Few - cycle driver : $\tau_p < 3T_o$

τ_p = 5 fs, T_o = 2.5 fs

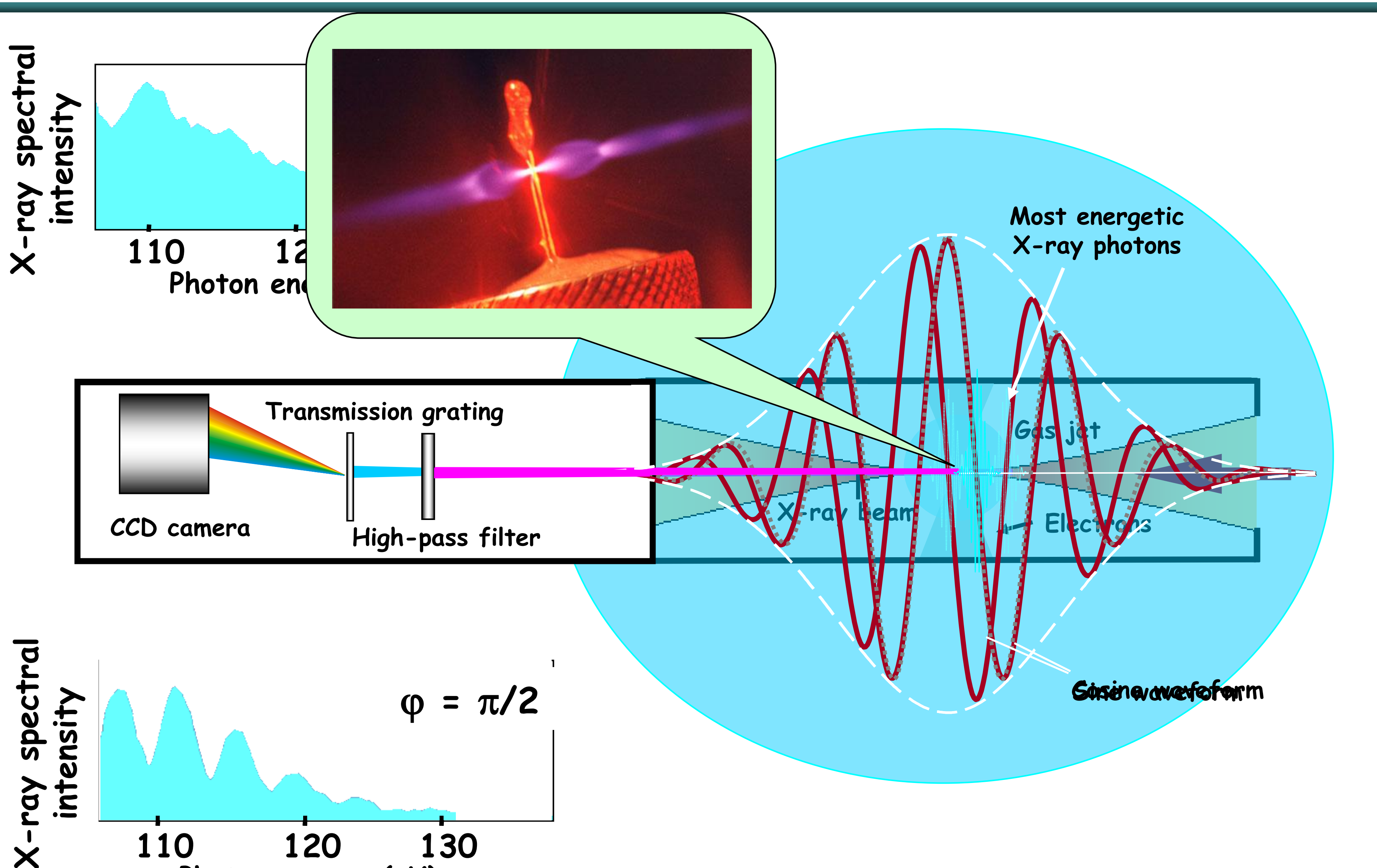


Kienberger, Baltuska, Krausz, 2003



- **No genuine harmonics of the laser radiation**
- **Cosine waveform with $\tau_p \sim 2T_o$ (5 fs @ 750 nm) offers the potential for single sub-femtosecond X-ray pulse generation**

HHG from a single attosecond burst

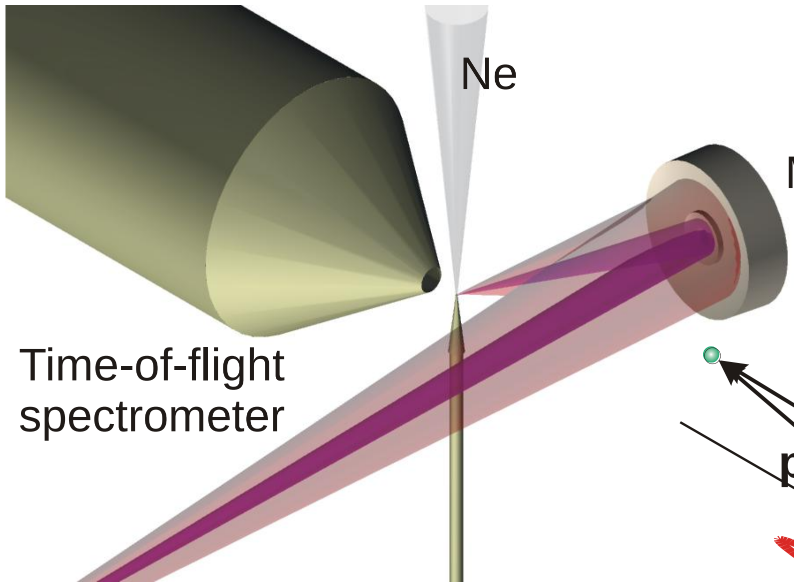


A. Baltuska *et al*, *Nature* 421, 611 (2003)

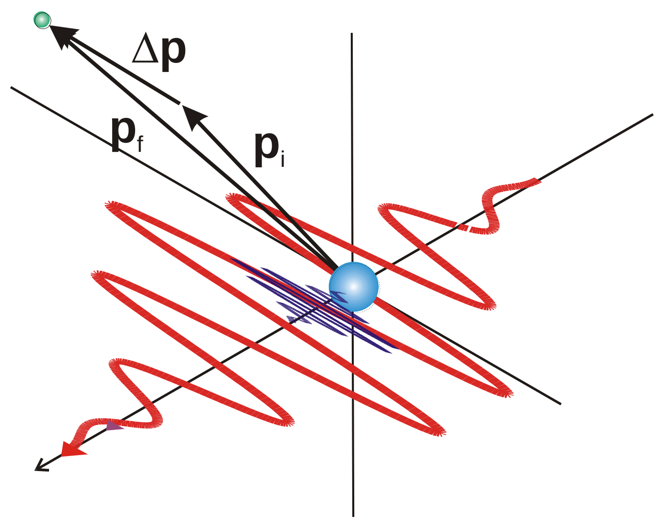
Acknowledgement: R. Kienberger

Ionization with an Isolated Attosecond Pulse

$$\Delta p(t_r) = e\int_{t_r}^{\infty} E_L(t')\,dt'$$



delay possible

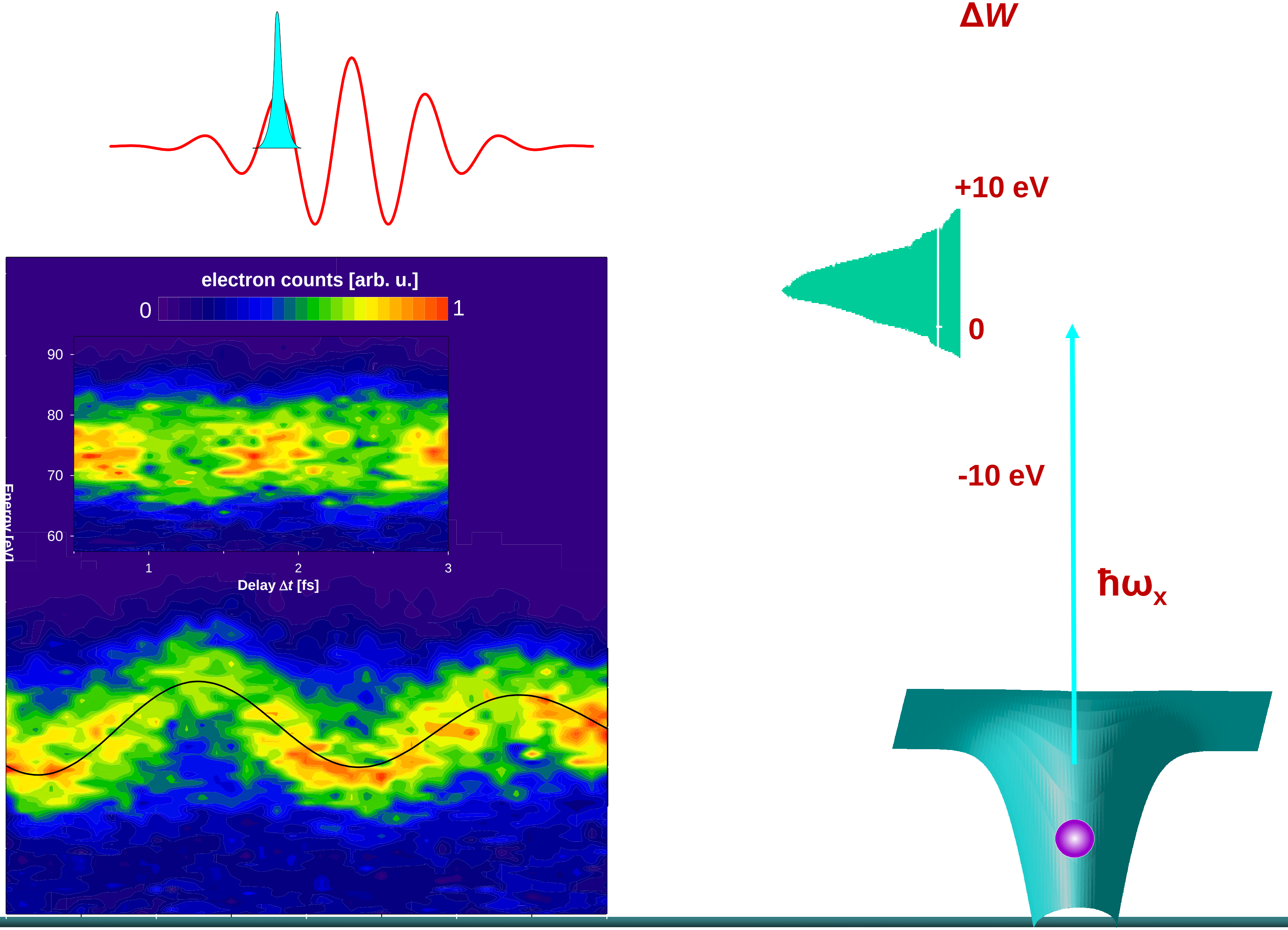


Detection as in:
Kienberger *et al*., Science **297**, 1144 (2002)

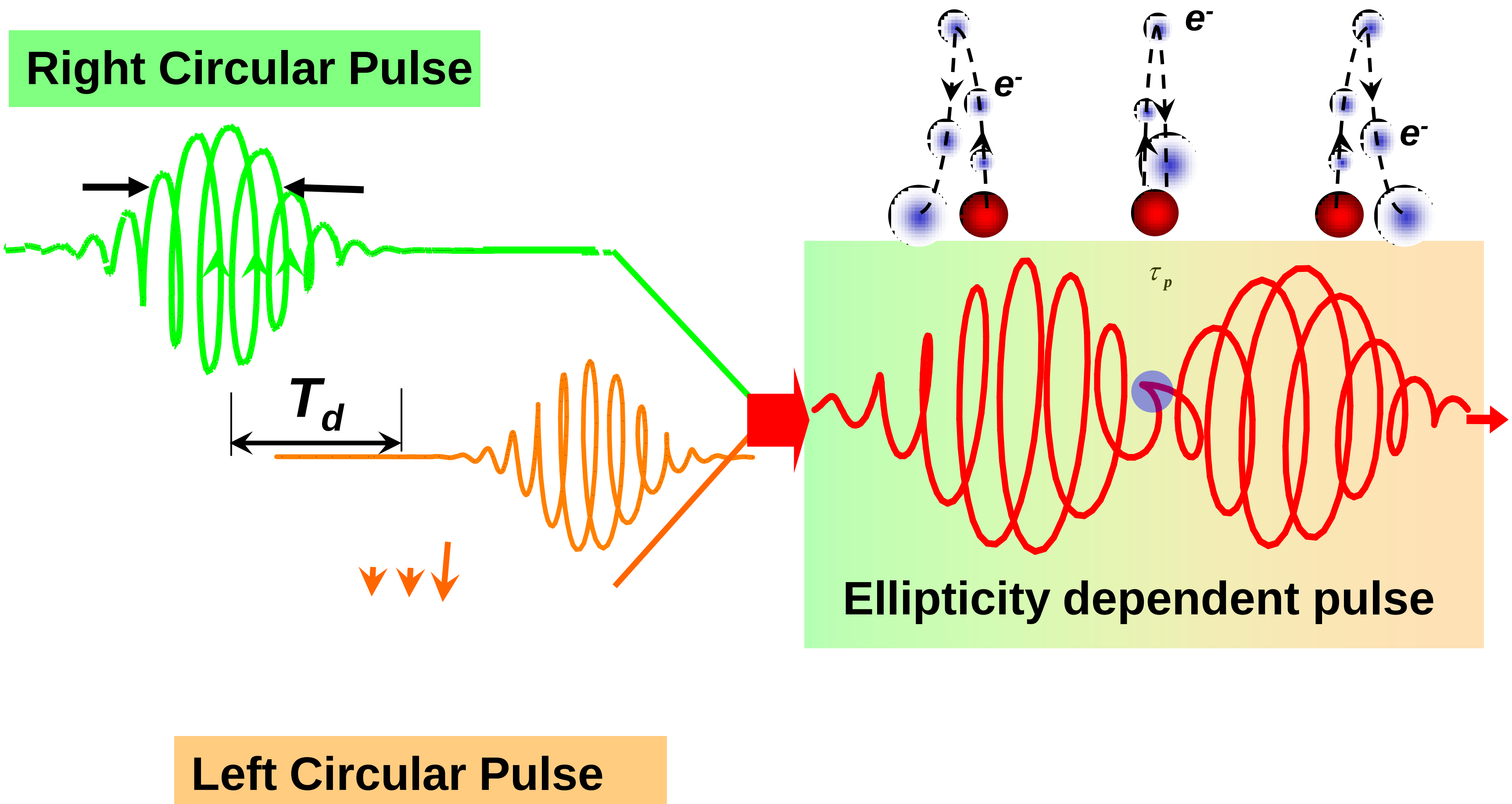
Gas: Ne
Electrons: 2p
W_b= 21.46 eV

XUV cut-off energy: ~93 eV
Mirror reflectivity bandwidth: ~9 eV (FWHM)

Mapping attosecond phenomena (attosecond streak camera)



Polarization gating for a single atto-pulse

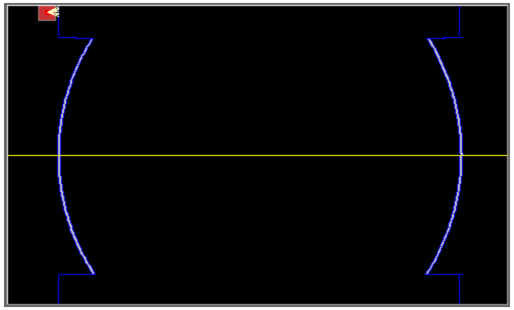


P. B. Corkum, N. H. Burnett, and M. Y. Ivanov, Opt. Lett. 19, 1870 (1994)

Summary

- **Good ultrafast sources and measurement techniques now exist from attoseconds to picoseconds**
- **Commercial sources are largely based on the properties of solid state laser media, especially Ti:Sapphire**
- **Attosecond science is pushing ultrafast into the VUV and soft x-rays**
- **Nonlinear optics is the key to ultrafast metrology**
- **Nonlinear processes become weaker at x-ray wavelengths, and this is a challenge for the field**

Q1: Which is stable?

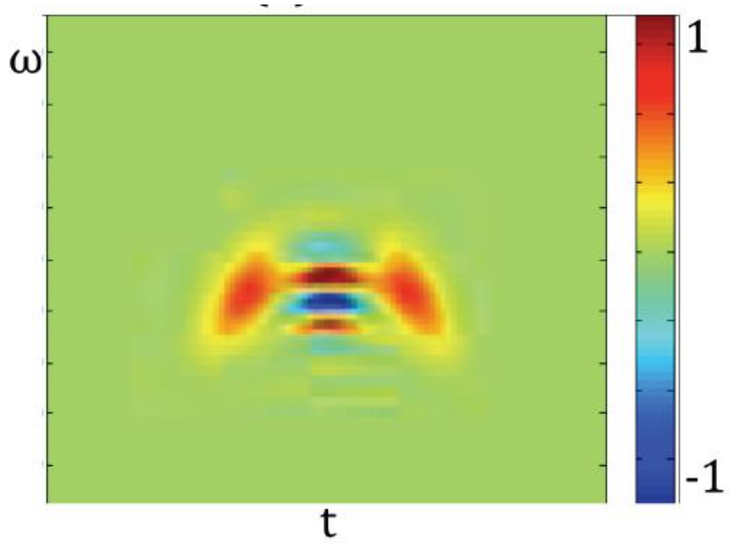


R1=R2=2.2
L=4

R1=R2=1.8
L=4

R1=R2=3
L=4

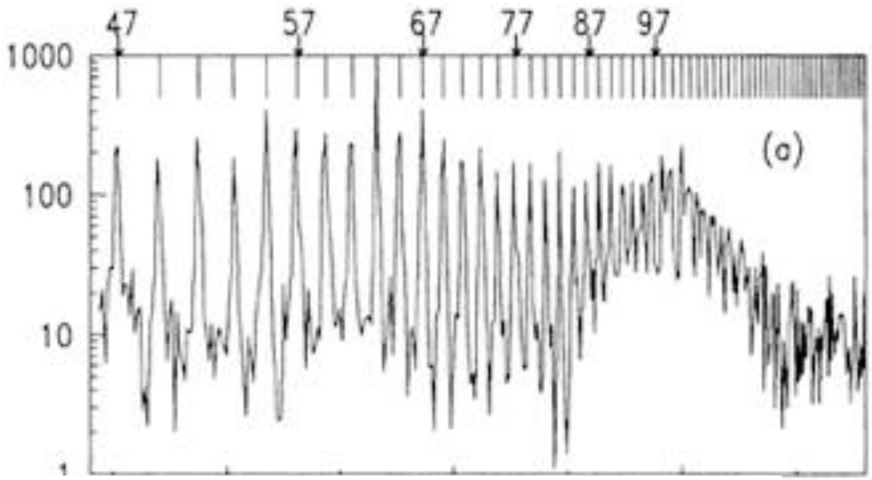
Q2: Describe the pulse that made this Wigner Distribution



Answer is beneath this card

High harmonics?

- **Why is the HHG spectrum split into discrete peaks?**
- **Why are the haronics odd?**
- **Why do longer wavelength drive lasers make higher harmonics?**



(from A. L'Huillier, Phys. Rev. Lett. 70, 774, 1993)