Generation and Application of Carrier Envelope Phase Controlled Single-Cycle Optical Pulse Train

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Generation of sub-single-cycle pulses in the optical region

- Introduction
- Review of basic concepts
- Modeling molecular modulation
- Present status of experiments
- How to determine pulse duration
- Advance concepts

Prefix for small and large numbers:

micro	10^{-6}	mega	10^{6}
nano	10^{-9}	giga	10^{9}
pico	10^{-12}	tera	10^{12}
femto	10^{-15}	peta	10^{15}
atto	10^{-18}	exa	10^{18}
zepto	10^{-21}	zetta	10^{21}
yotta	10^{-24}	yocto	10^{24}

Man made today:

shortest time -- about 100 as

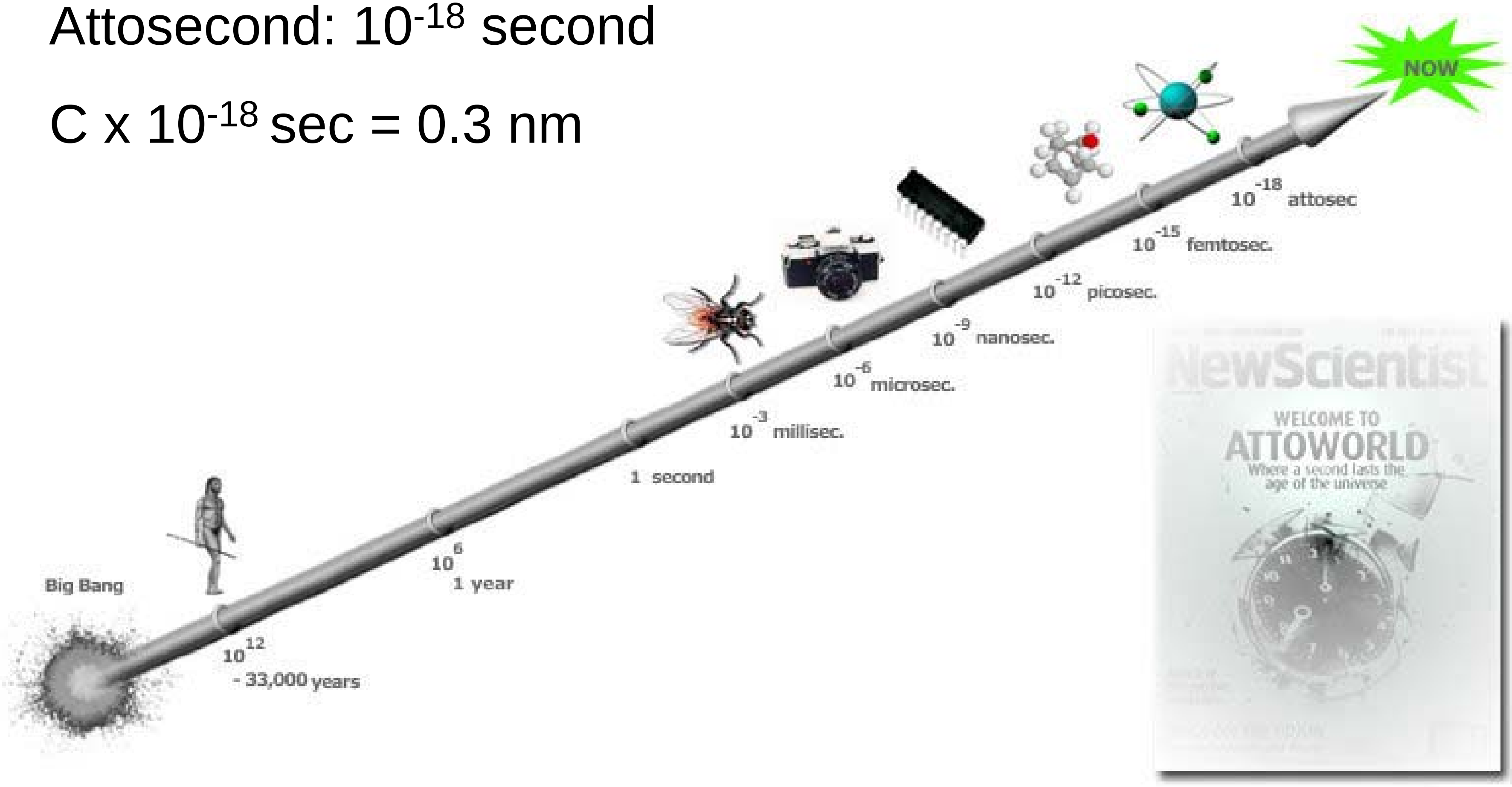
most intense light -- 10^{23} W/cm^{2}

Courtesy C. D. Lin, Kansa State U

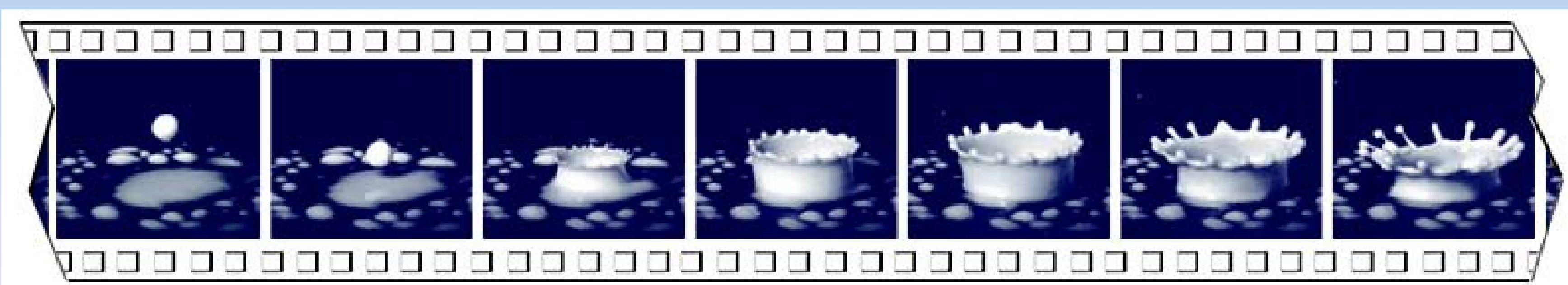
Attoworld

Attosecond: 10^{-18} second

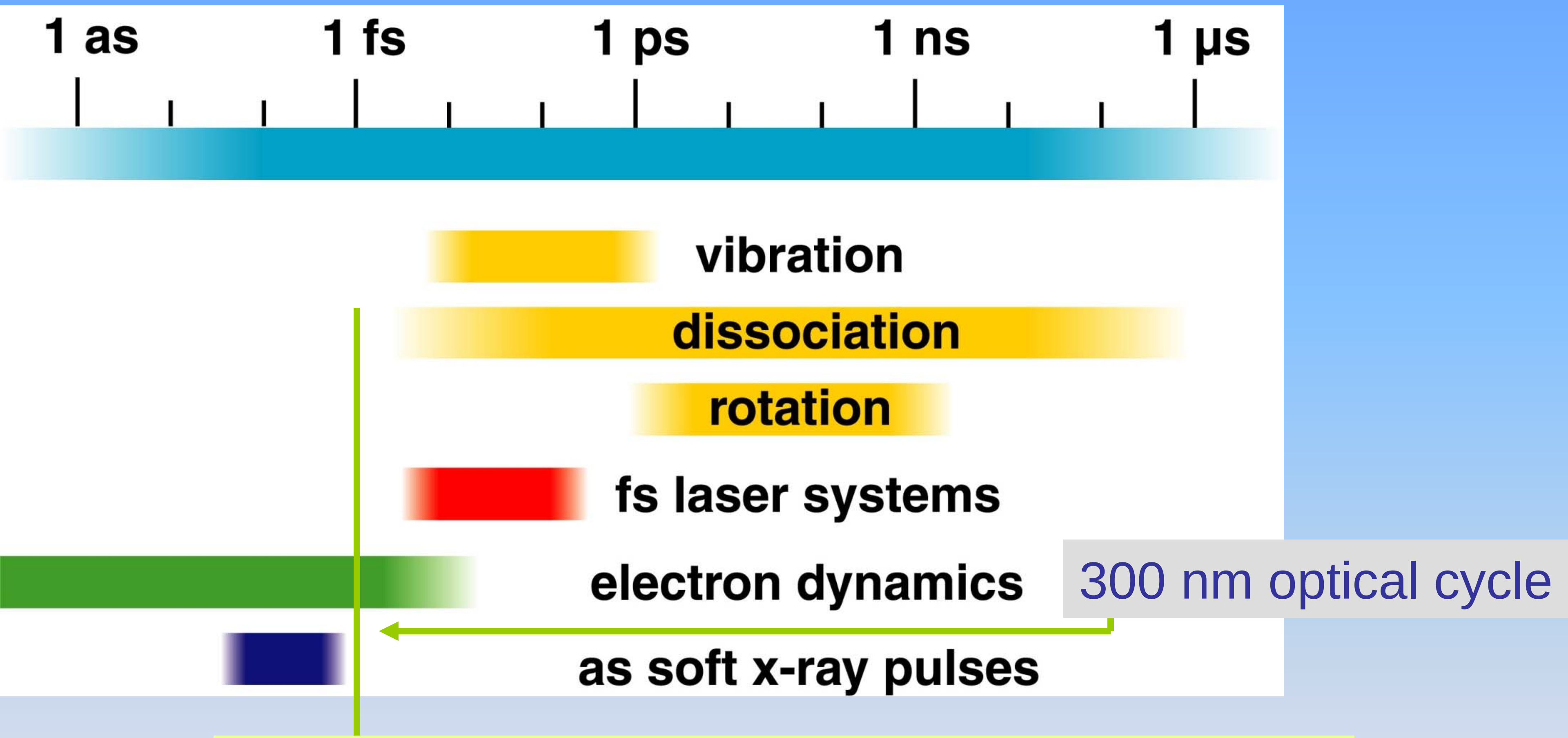
C x 10^{-18} sec = 0.3 nm



www.attoworld.de/attoworld.html

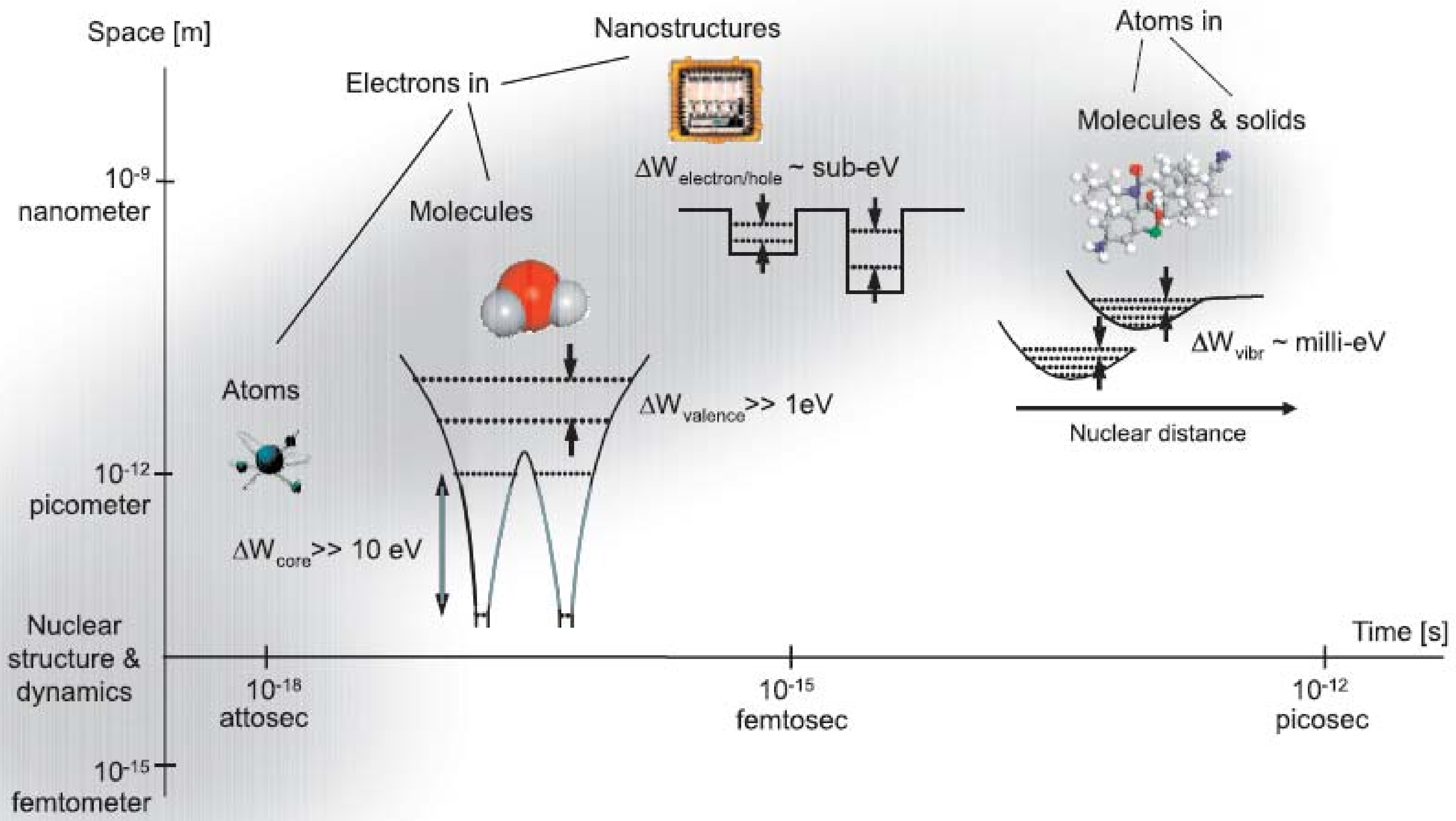


Time scales



Short pulses can be used to monitor and control molecular and electronic motion

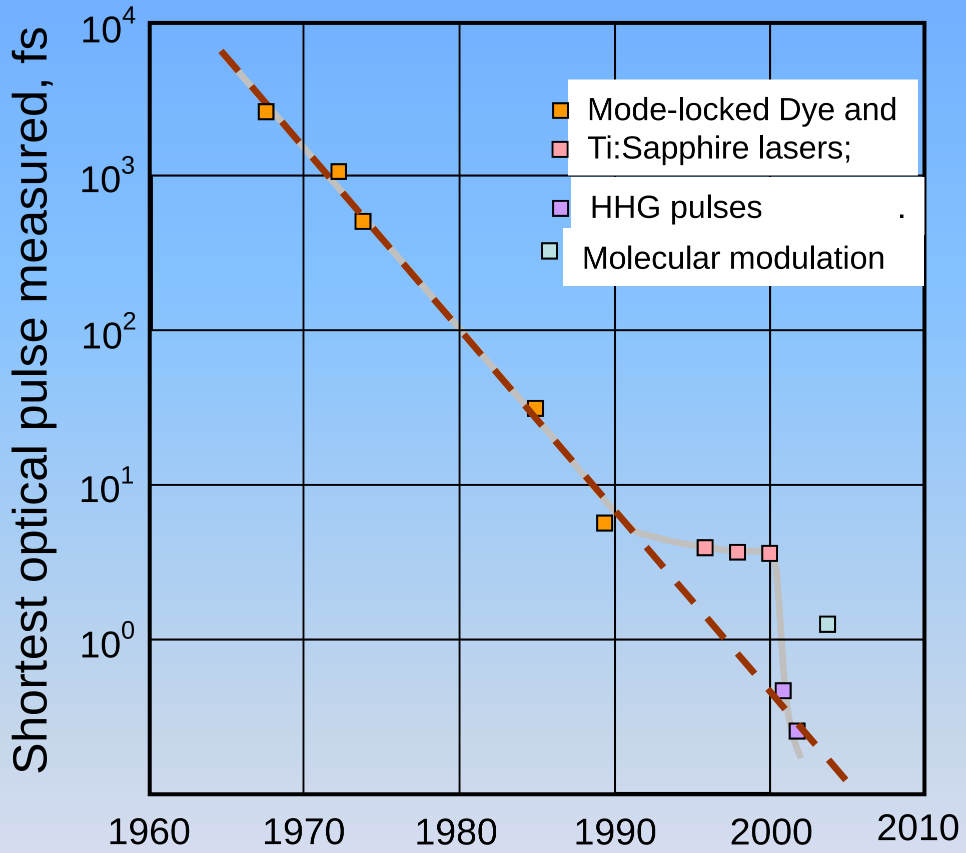
Characteristic length and time scales for structure and dynamics in the microcosm



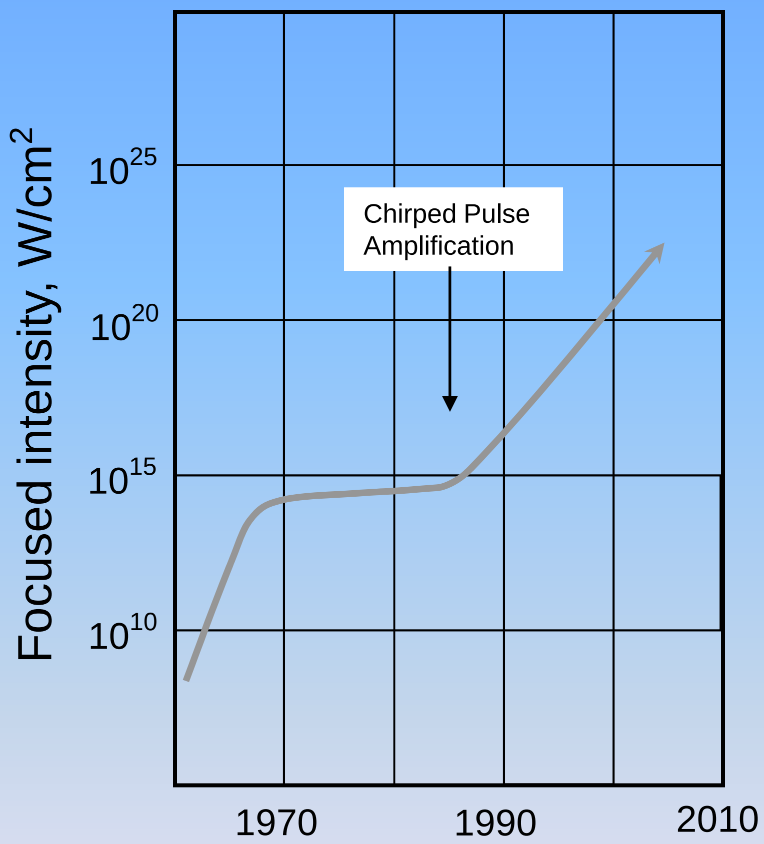
F. Krausz & M. Ivanov, Rev. Mod. Phys. 81 163 (2009)

Laser pulses got shorter over the years

Peak intensity increased



Ultrafast science

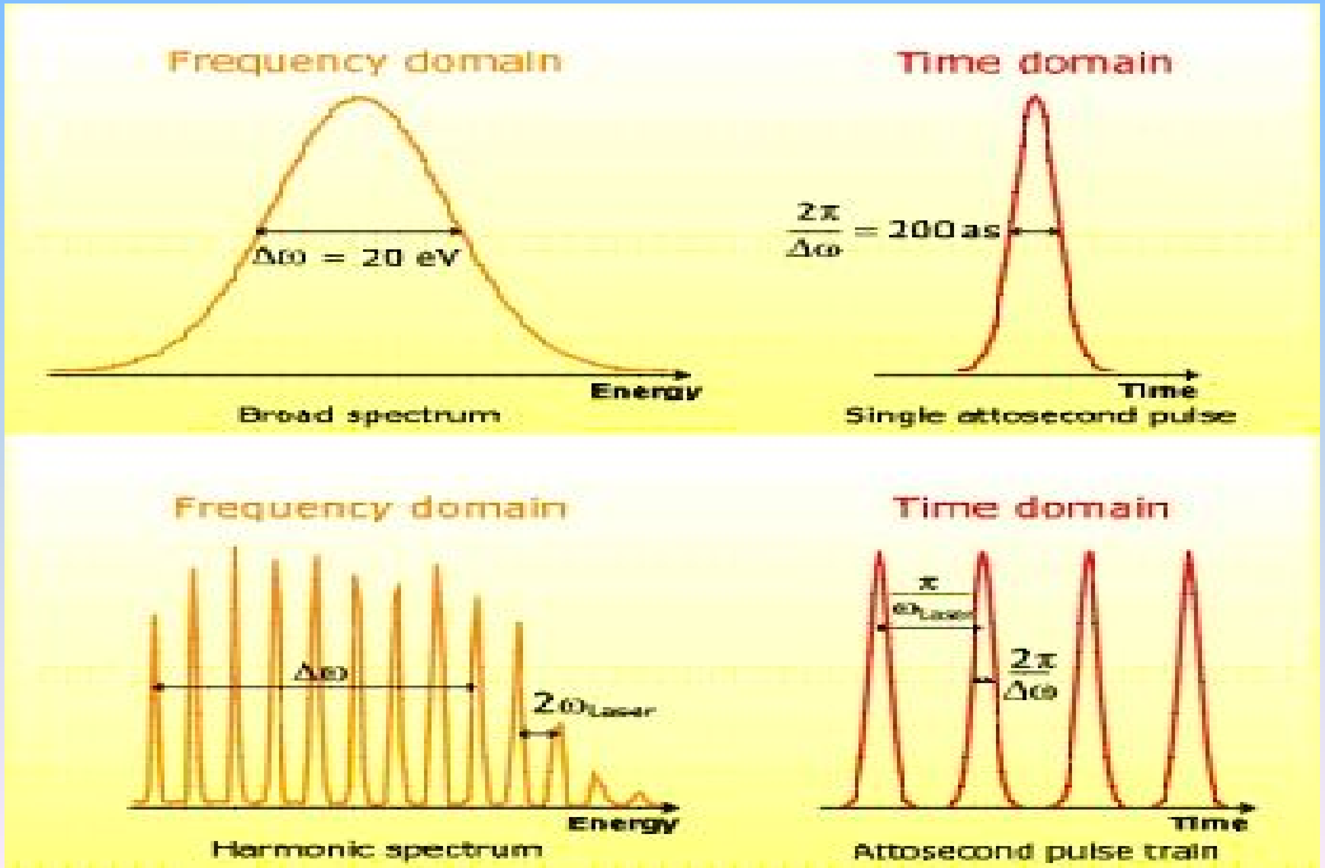


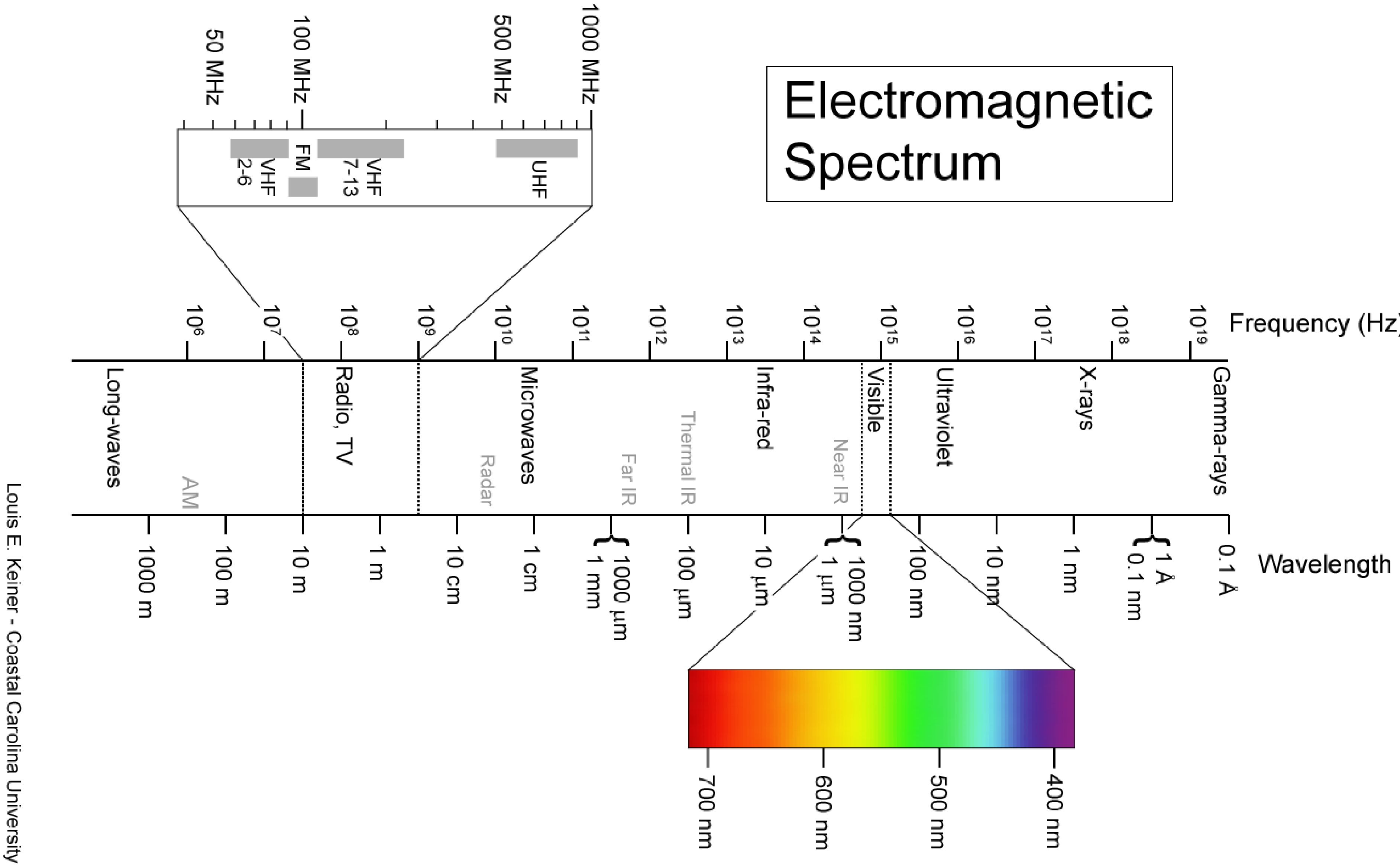
High field physics

Correlation between time and frequency

$$x(t-t_0) \xleftrightarrow{FT} e^{-j\omega t_0} X(\omega)$$

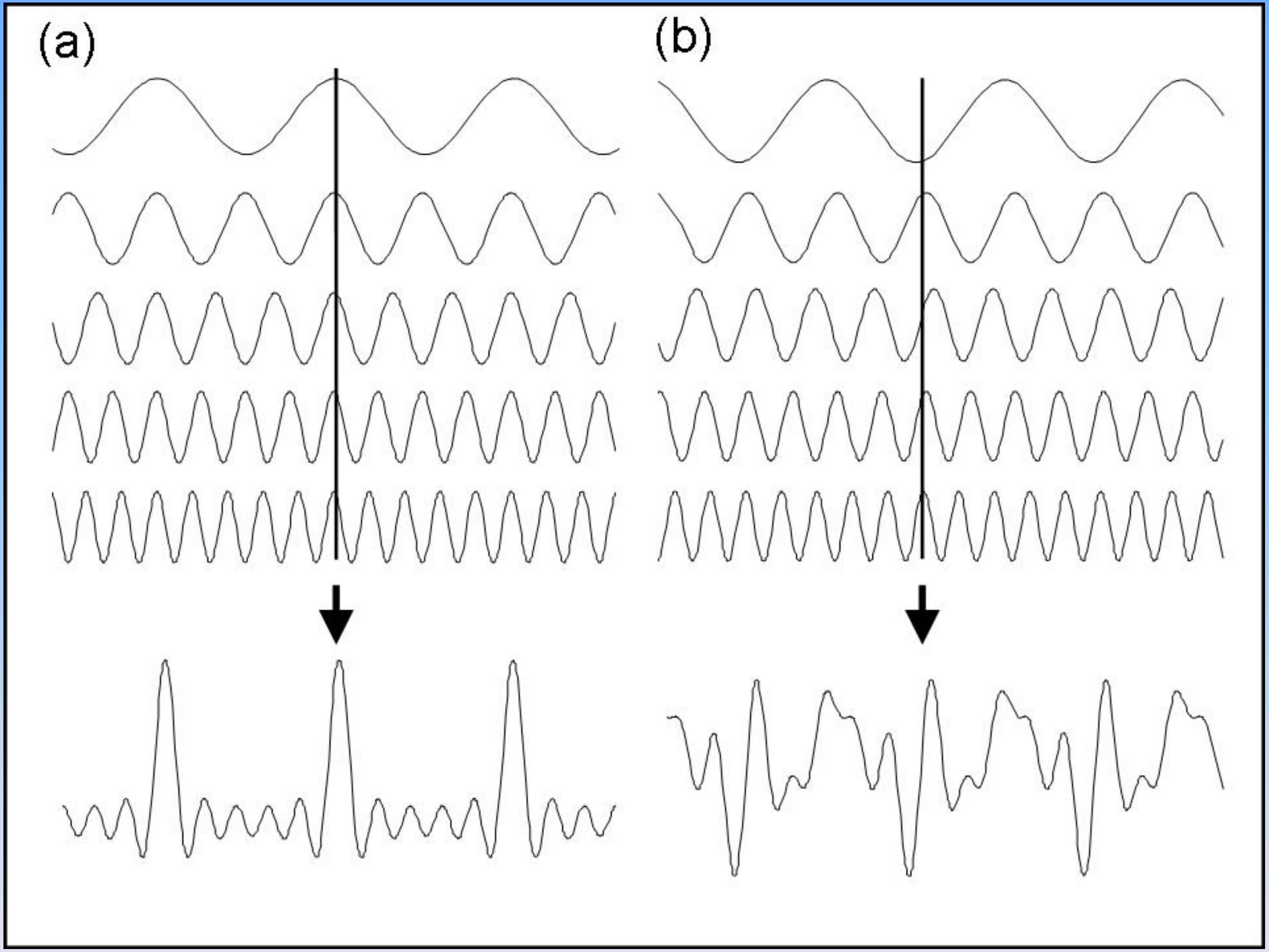
Fourier transform: $$X(\omega) = \int_{-\infty}^{\infty} x(t) e^{-i\omega t} d\omega$$





Electromagnetic Spectrum
Frequency (Hz)
10^6 10^7 10^8 10^9 10^{10} 10^{11} 10^{12} 10^{13} 10^{14} 10^{15} 10^{16} 10^{17} 10^{18} 10^{19}
50 MHz
100 MHz
500 MHz
1000 MHz
VHF 2-6
FM
VHF 7-13
UHF
Long-waves
AM
Radio, TV
Radar
Microwaves
Far IR
Thermal IR
Infra-red
Near IR
Visible
Ultraviolet
X-rays
Gamma-rays
Wavelength
1000 m
100 m
10 m
1 m
10 cm
1 cm
1 mm
1000 µm
100 µm
10 µm
1 µm
1000 nm
100 nm
10 nm
1 nm
0.1 nm
1 Å
0.1 Å
700 nm
600 nm
500 nm
400 nm
Louis E. Keiner - Coastal Carolina University

Effect of random phase
Principle of optical interference of coherent light fields



In phase

Random phase

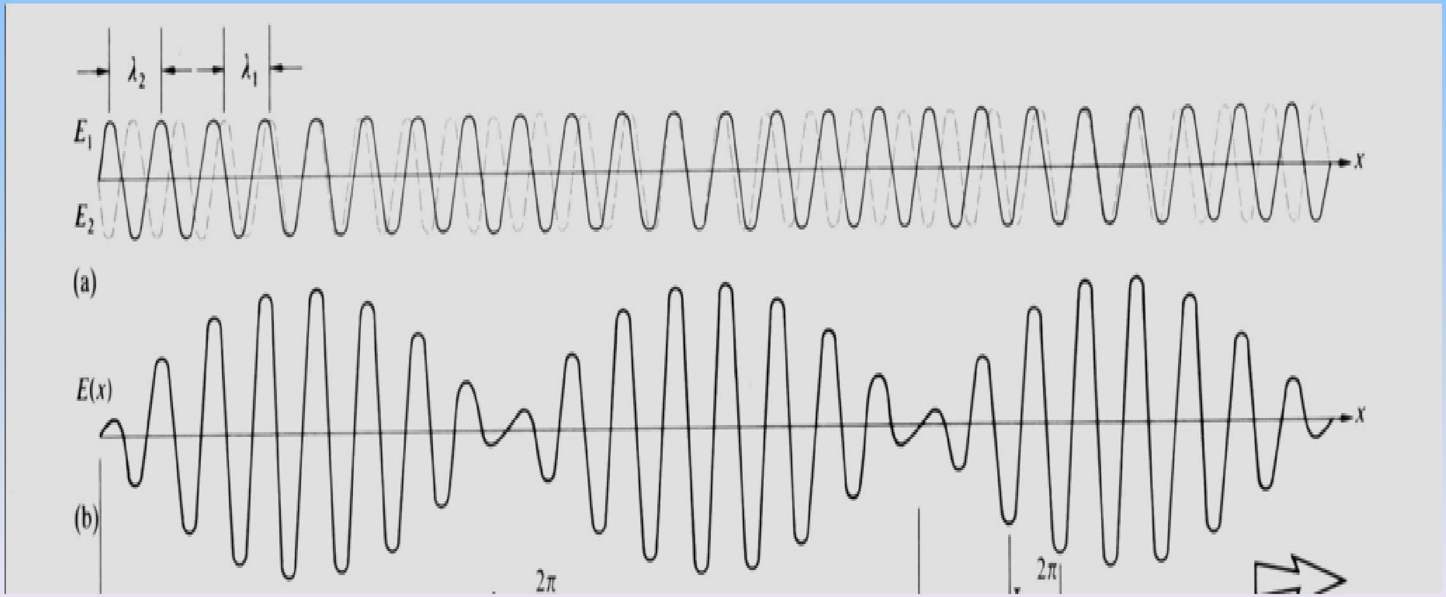
What is a single cycle optical pulse

(a) Monochromatic light: sinusoidal wave propagation

(b) Beating of two waves, ω_1 and ω_2

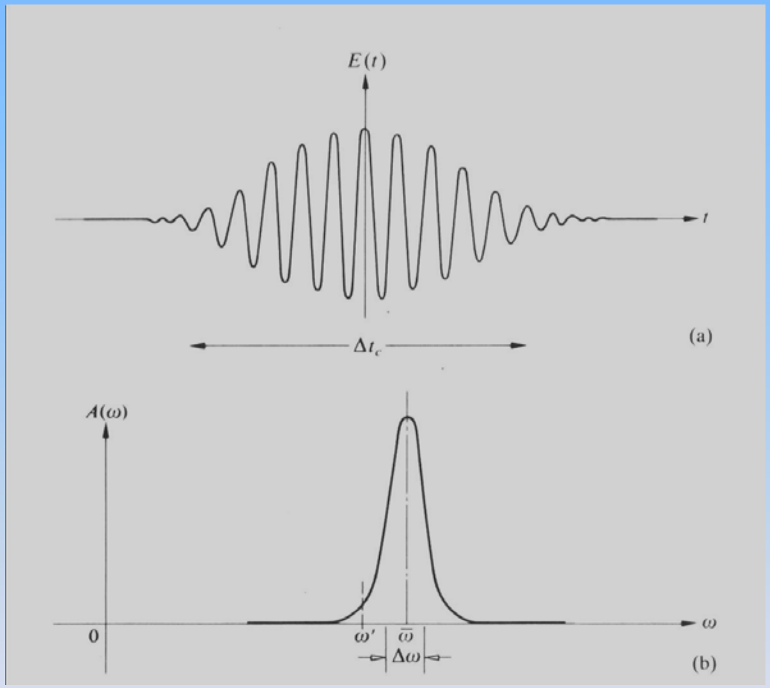
(a)

(b)

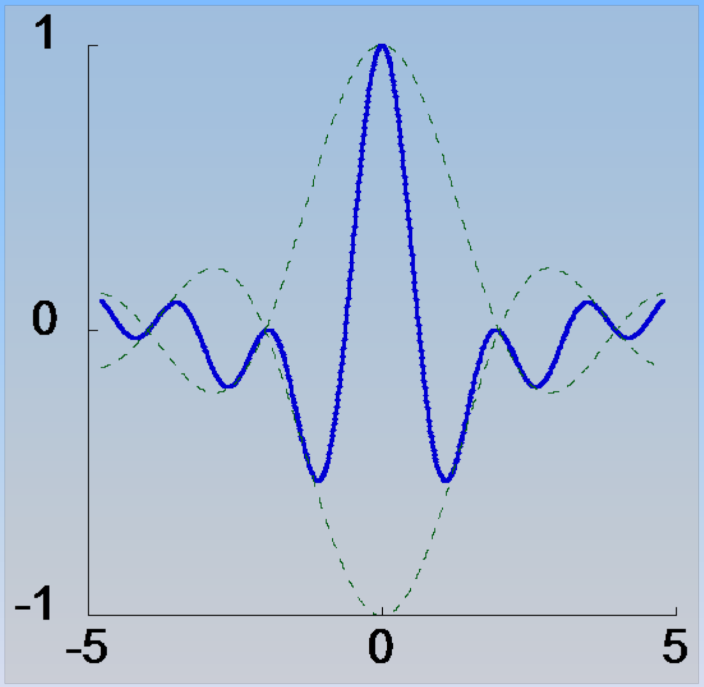


What is a single cycle optical pulse

(c) Many waves propagating to form a wave packet (left)
(d) Ultimate wavepacket is a single-cycle and sub-cycle pulse pulse (right)



(c)



(d)

Optical cycle

$$E(t) = \tilde{E}(t) + c.c. \qquad \tilde{E}(t) = A(t)e^{i(\omega_0 t + \phi)}$$

$$\omega_0 = \frac{\int_0^\infty \omega |E(\omega)|^2 d\omega}{\int_0^\infty |E(\omega)|^2 d\omega}$$ Carrier frequency

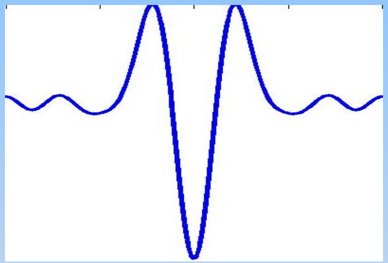
$E(\omega)$: Fourier transform of $E(t)$

T. Brabec and F. Krausz, Phys. Rev. Lett. 78, 3282 (1997)

Single cycle waveforms

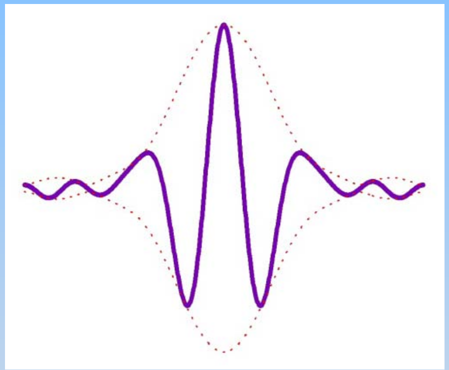
Inverted cosine

$\phi_n = \pi$



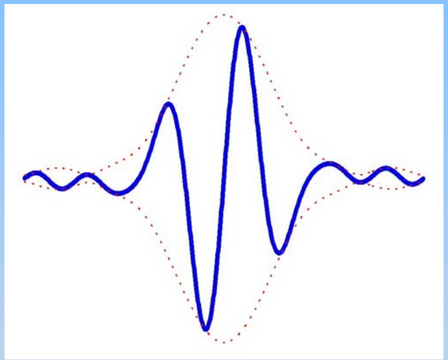
cosine pulse

$\phi_n = 0$



sine pulse

$\phi_n = \pi/2$

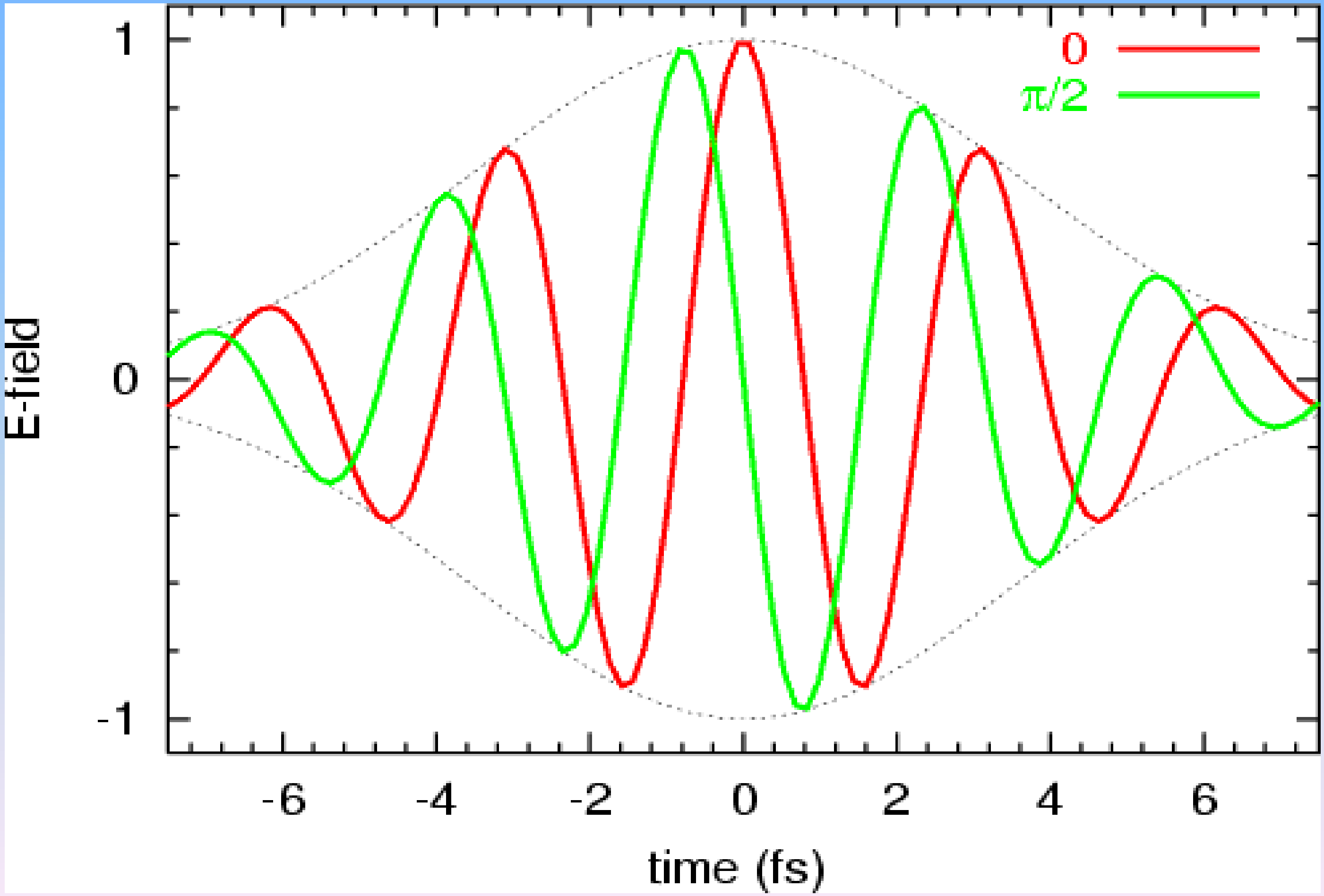


780 nm ⟺ ***12,820 cm^{-1}*** ⟺ ***2.6 fs***

200 nm ***50,000 cm^{-1}*** ***684 as***

Carrier envelope phase

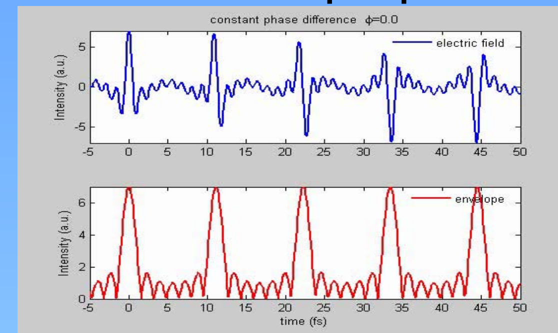
$$E(t) = E_0(t)\cos(\omega_0 t + \underline{\phi})$$



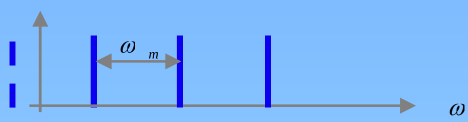
Constant carrier envelope phase

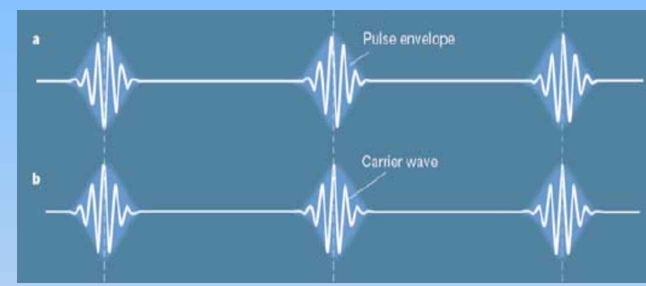
$$E(t)=\sum_n E_n(t)\cos(\omega_n t+\phi_n)$$

Φ_n = carrier envelope phase



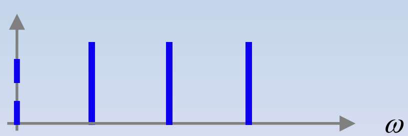
incommensurate $\omega_n = n\omega_m + \omega_{ceo}$ $\phi_n = \omega_{ceo} t + \phi_n'$

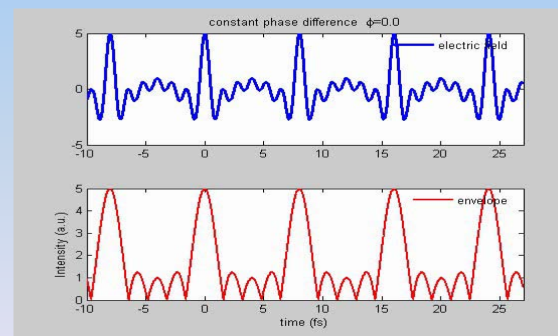




commensurate $\omega_q = n\omega_m$ $\phi_n = \phi_{CEP} + n\phi_m$

$$E(t)=\sum_n A_n(t)\cos(n\omega_m(t+\phi_m/\omega_m)+\phi_{CEP})$$





Constant CEP requires that the frequencies are commensurate and the relative phases form an arithmetic series

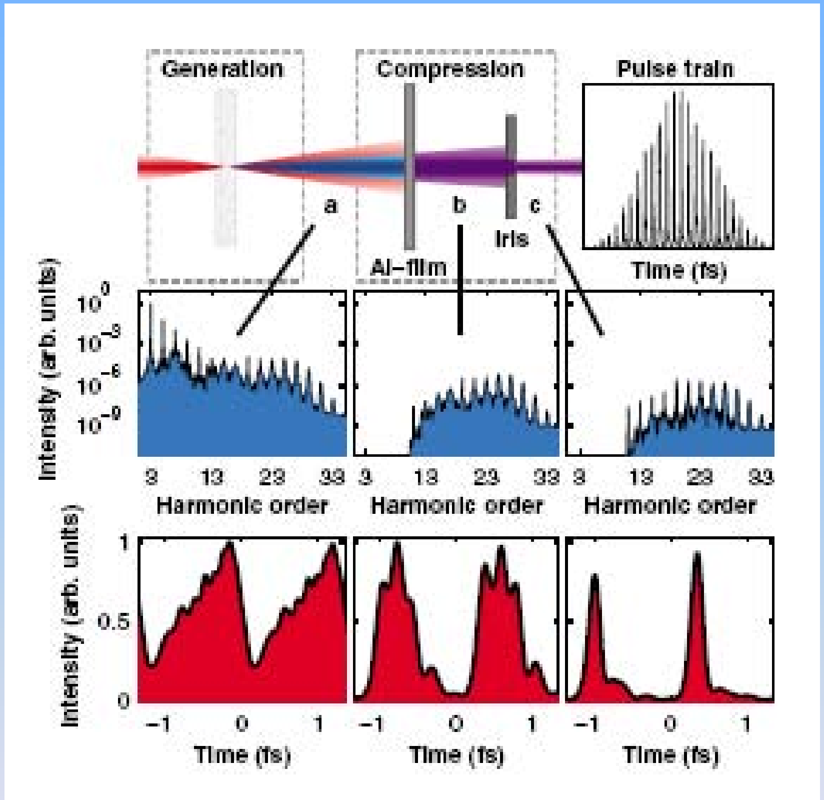
Ingredients of an attosecond single-cycle optical pulse:

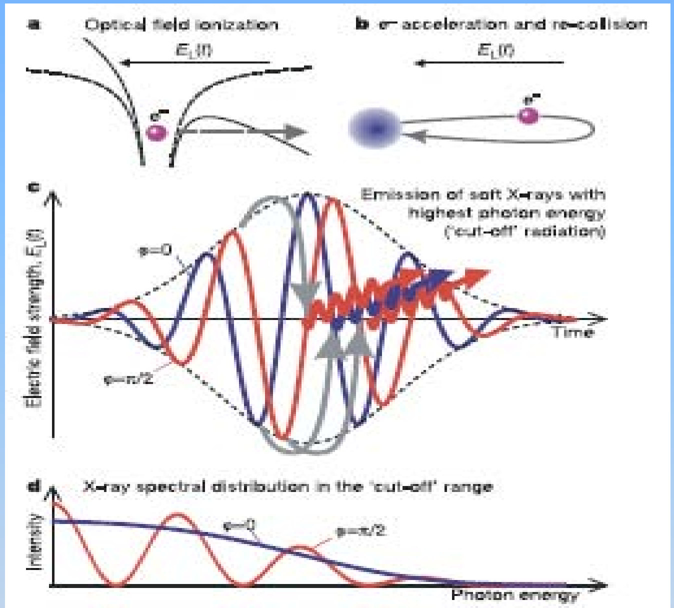
1. Broad spectrum – 2 or more octaves
2. In phase condition
3. Constant carrier envelope phase:
   - Commensurate frequencies
   - Constant phase difference between adjacent spectral components
4. Stable and controllable carrier envelope phase

Methods of generating attosecond pulses

A

High-order harmonic generation of phase-stabilized femtosecond pulse





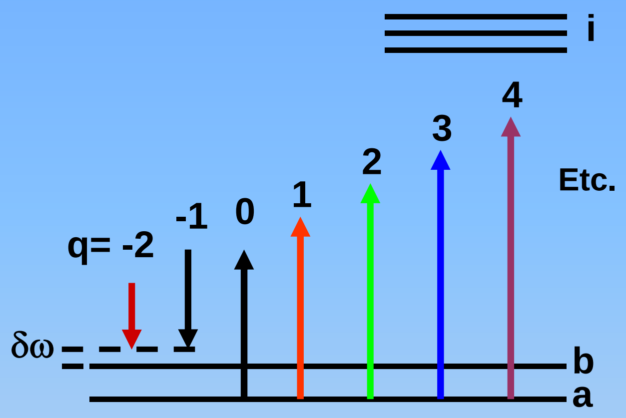
Advantages: single pulse
100 attosecond
Disadvantages: 30-100 eV photons
very low power
constitutes a few cycles

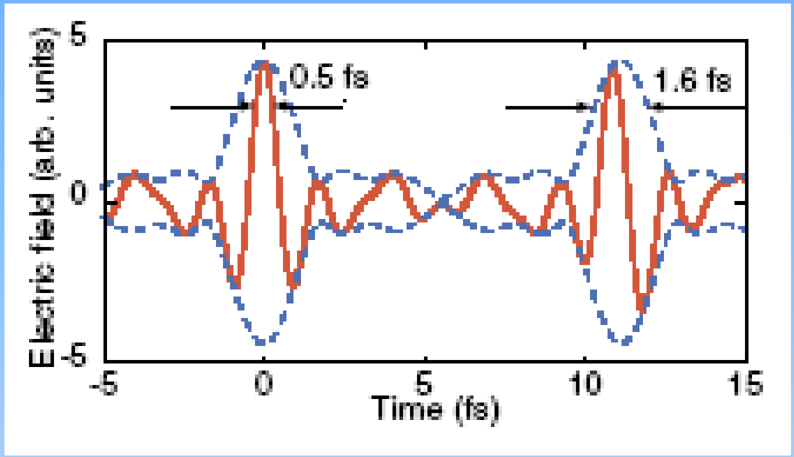
Krauze et.al., Nature 421, 611 (2003)
R. Lopez-Martens et. al., PRL 94, 033001 (2005)

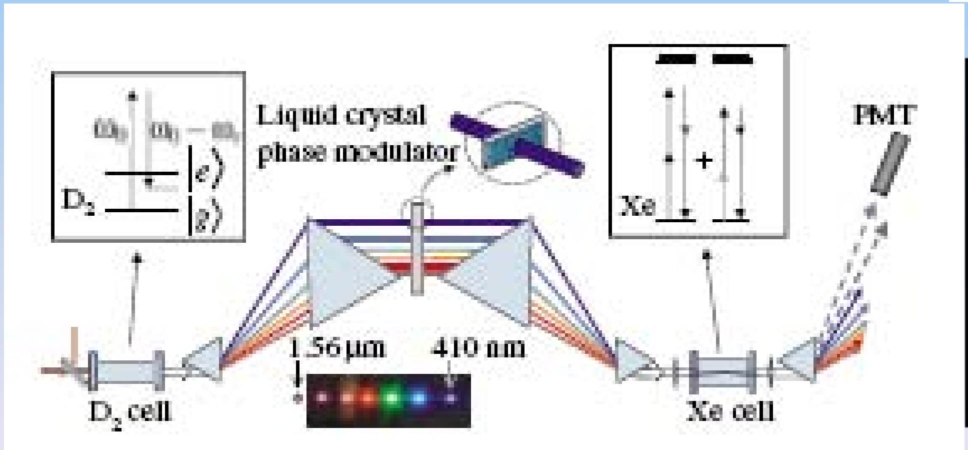
Methods of generating attosecond pulses

B

High-order stimulated Raman scattering using molecular modulation





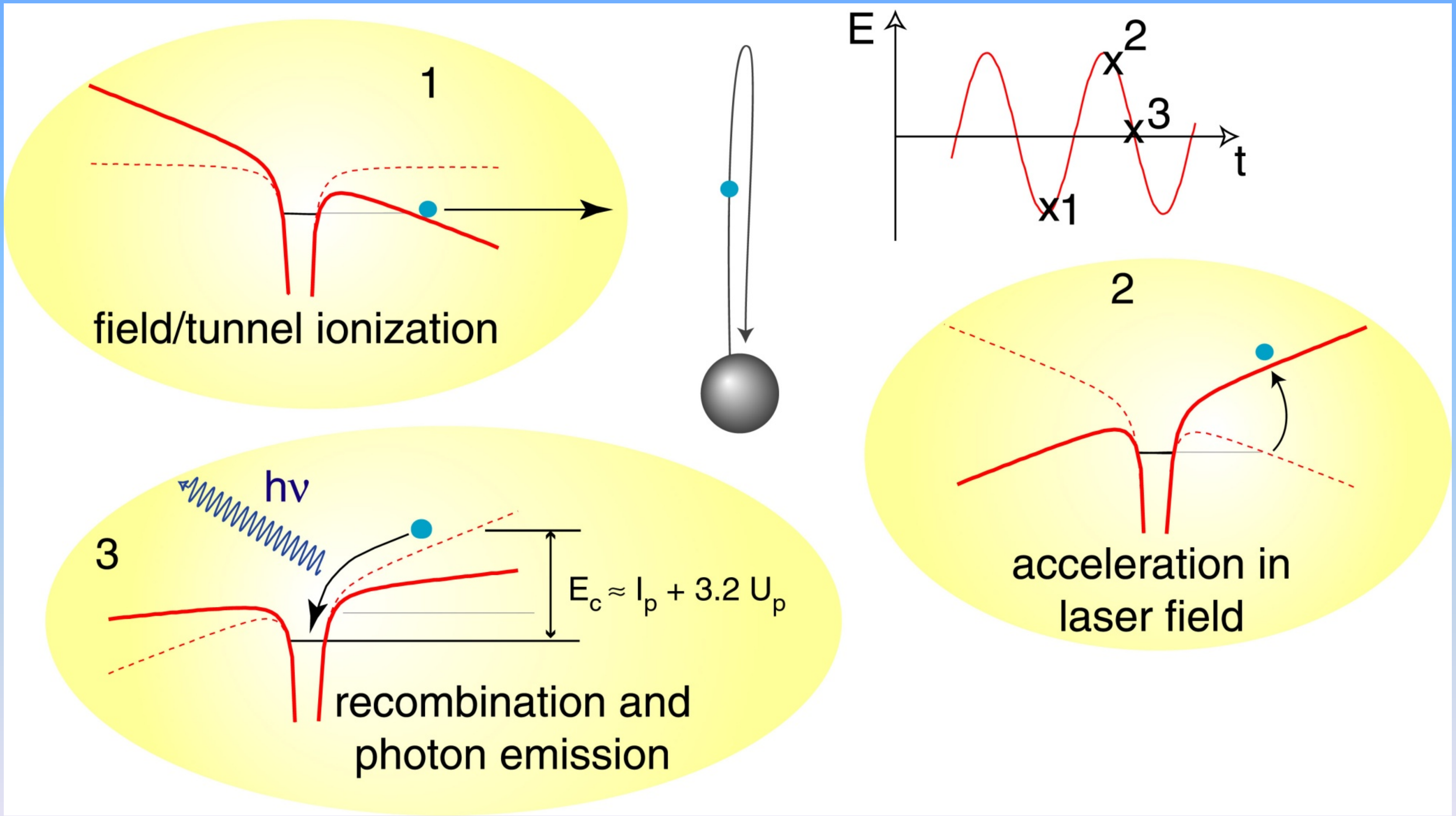


Advantages: IR-UV region
good power
single-cycle

Disadvantages: complex setup
8-50 fs pulse spacing
limited to ~ 300-500 as

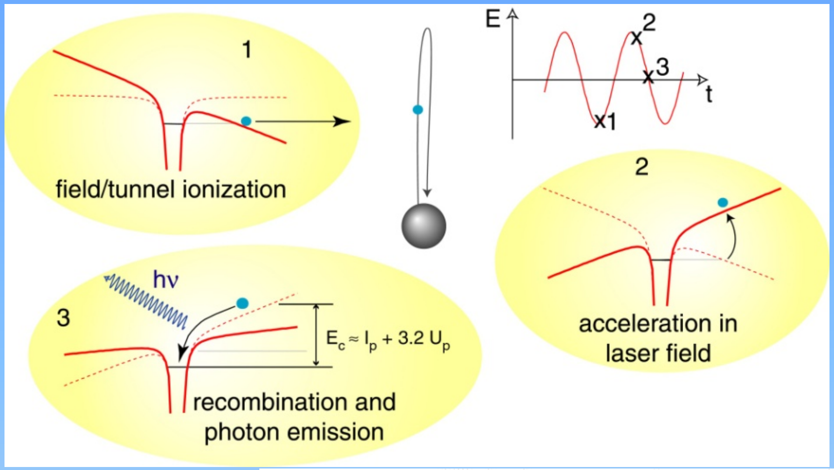
M.Y. Shverdin et.al., PRL 94, 033904 (2005)

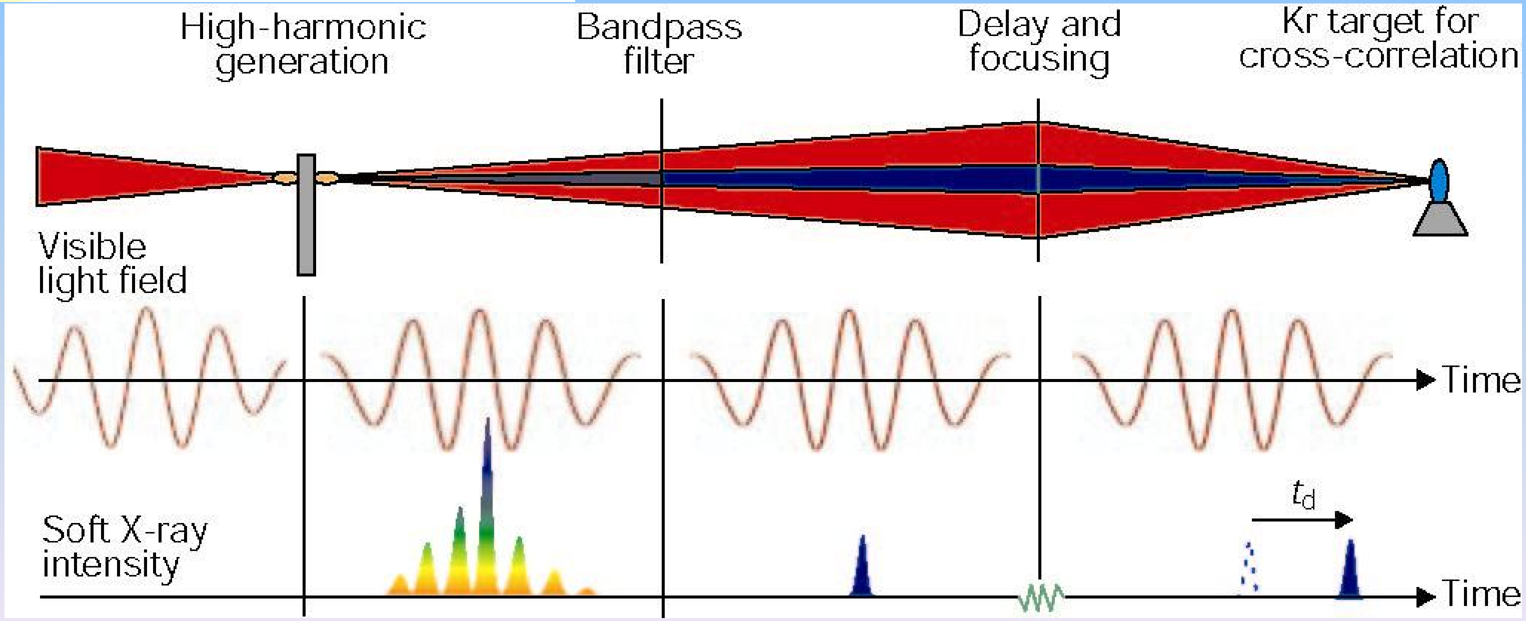
Three-step model



P. Corkum, Phys. Rev. Lett. **71**, 1994 (1993)

Isolated Attosecond-pulse production





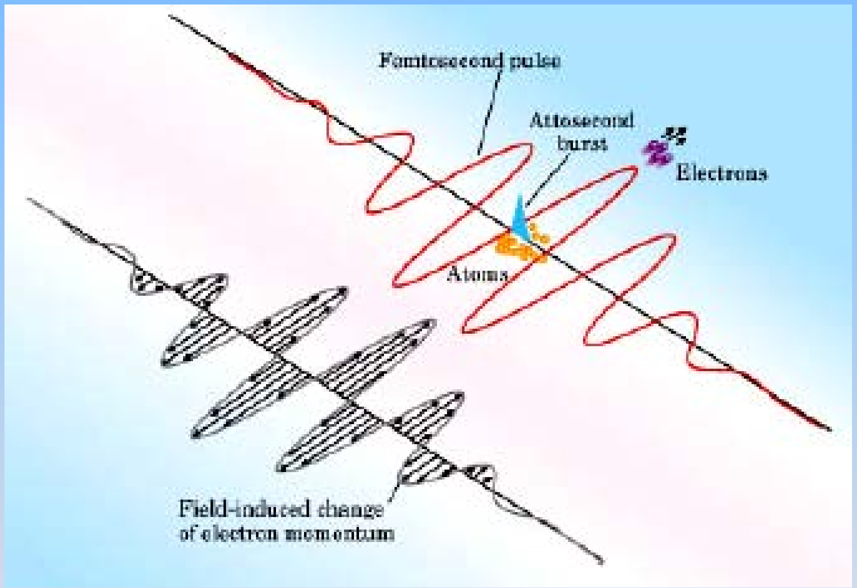
M. Hentschel et al, Nature **414**, 509 (2001)

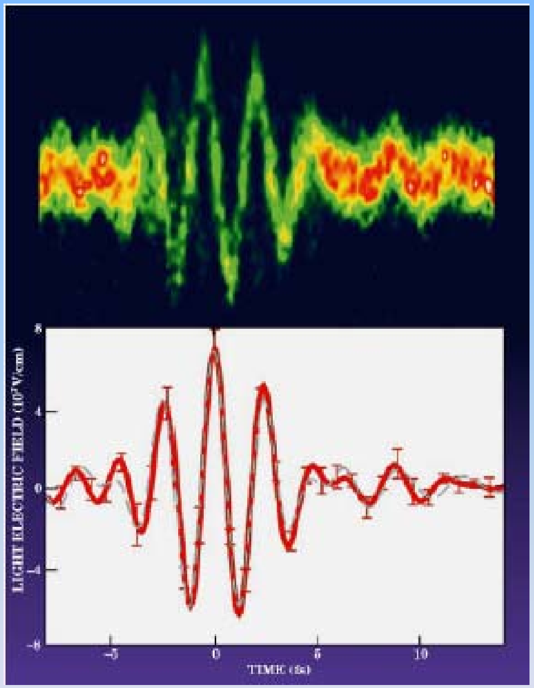
PHYSICS TODAY   October 2004

Search and Discovery

Attosecond Bursts Trace the Electric Field of Optical Laser Pulses

The familiar textbook sketch of light's oscillating electric field can now be drawn directly from measurements.

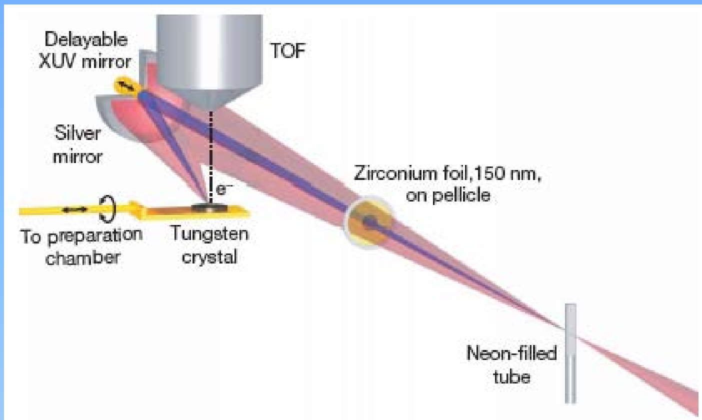


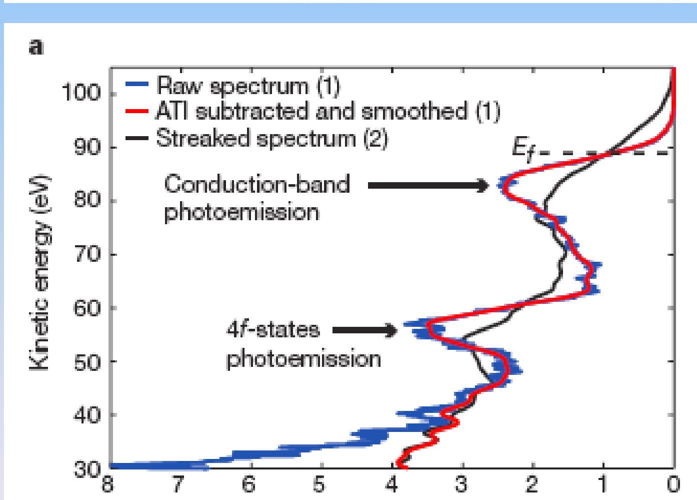


A. Baltuska et al., Nature **421**, 611 (2003)
E. Goulielmakis et al., Science **305**, 1267 (2004)

Attosecond spectroscopy in condensed matter

A. L. Cavalieri[1], N. Müller[2], Th. Uphues[1,2], V. S. Yakovlev[3], A. Baltuška[1,4], B. Horvath[1], B. Schmidt[5], L. Blümel[5], R. Holzwarth[5], S. Hendel[2], M. Drescher[6], U. Kleineberg[3], P. M. Echenique[7], R. Kienberger[1], F. Krausz[1,3] & U. Heinzmann[2]





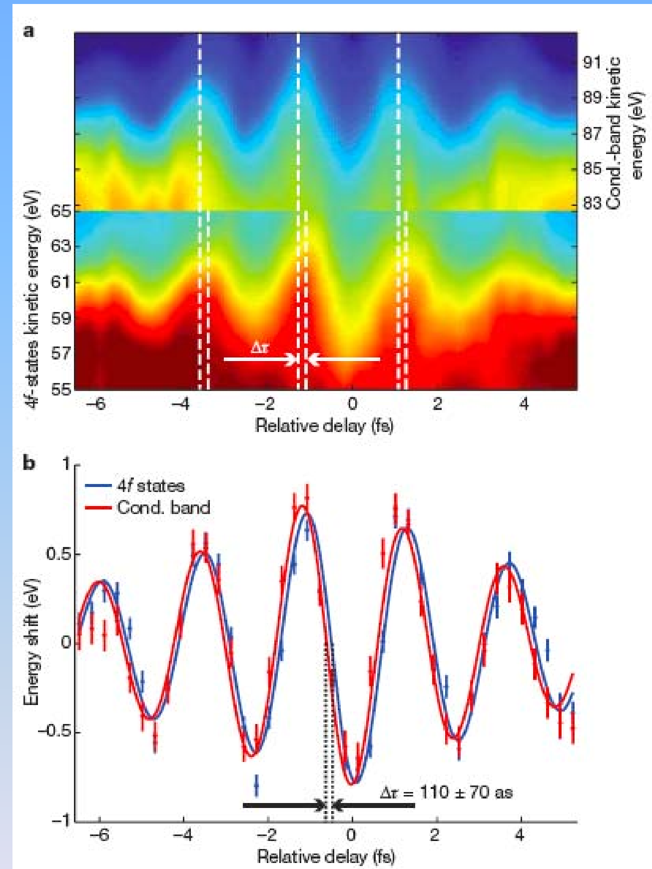


Figure 3 | Evidence of delayed photoemission. a, The 4*f* and conduction-band spectrograms, following cubic-spline interpolation of the measured data

Nature 449, 1029 (2007)

Common theme

Photon energies 30 eV to 100 eV (EUV to soft x-ray region)

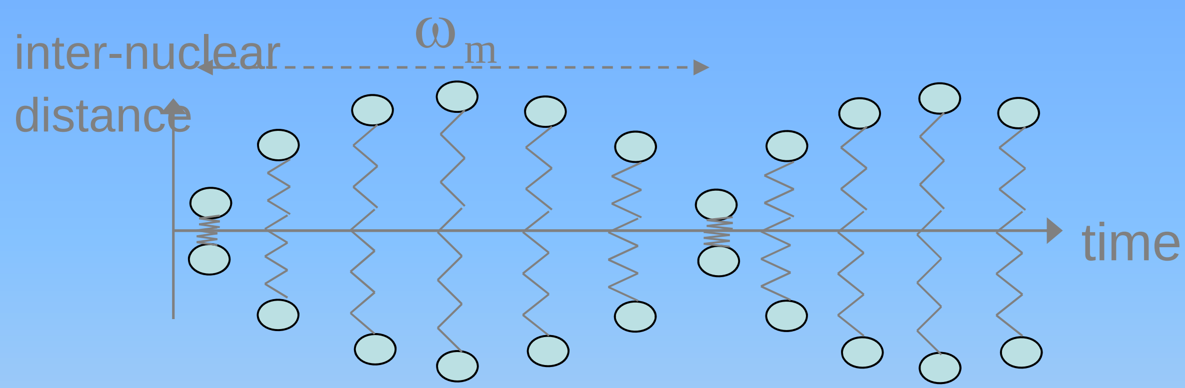
Traditional pump-probe measurements

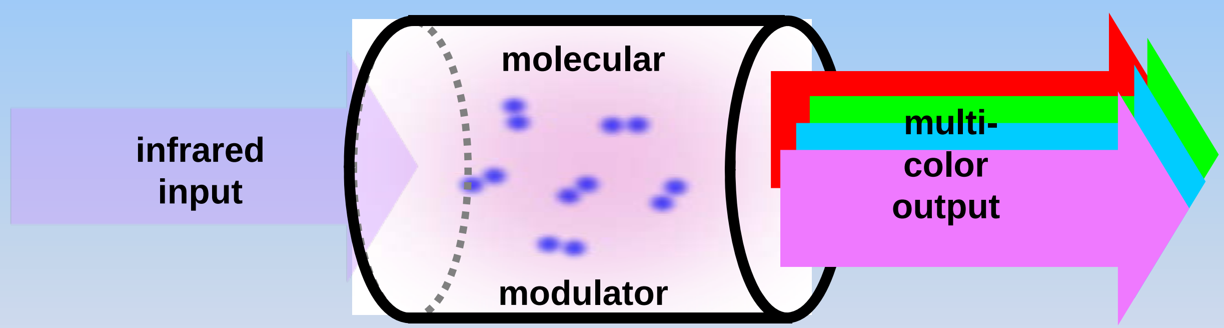
Photoelectron or photoion detection

Weak pulses - long signal acquisition time

Molecular Modulation

Molecular modulation is analogous to electro-optic modulation





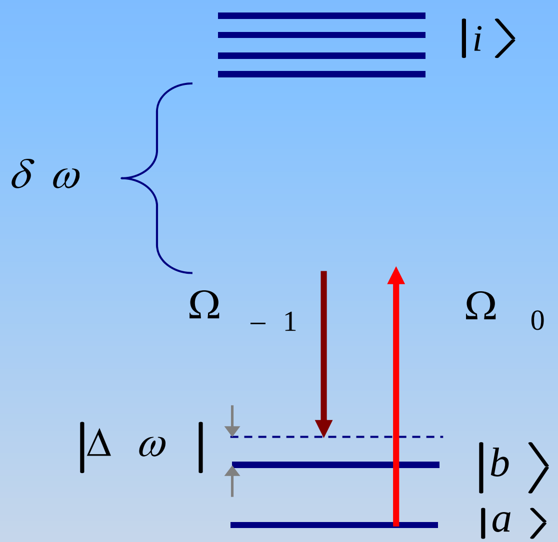
Refractive Index $n = n_0 + \delta\cos\omega_m t$

$\omega_q=\omega_0+q\omega_m$ q= -2,-1,0,1,2,3….

Alexei Sokolov
Steve Harris

Coherent Molecular Excitation

- Two strong laser fields adiabatically drive the molecules into a maximally coherent state.



$$\frac{\partial\rho_{aa}}{\partial\tau} = i\left(\Omega_{ab}\rho_{ba} - \Omega_{ba}\rho_{ab}\right) + \gamma_{\parallel}\rho_{bb}$$

$$\frac{\partial\rho_{bb}}{\partial\tau} = -i\left(\Omega_{ab}\rho_{ba} - \Omega_{ba}\rho_{ab}\right) - \gamma_{b}\rho_{bb}$$

$$\frac{\partial\rho_{ab}}{\partial\tau} = i\left(\Omega_{aa} - \Omega_{bb} + \delta + i\gamma_{\perp}\right)\rho_{ab} + i\Omega_{ab}\left(\rho_{bb} - \rho_{aa}\right)$$

$$\Omega_{aa} = \frac{1}{2}\sum_q a_q \left|E_q\right|^2$$

$$\Omega_{bb} = \frac{1}{2}\sum_q b_q \left|E_q\right|^2$$

$$\Omega_{ab} = \Omega_{ba}^{*} = \frac{1}{2}\sum_q d_q E_q E_{q+1}^{*}$$

Maximal coherence, $\rho_{ab} = 0.5$

Coherent Molecular Excitation

$$H_{eff} = -\hbar \begin{bmatrix} \Omega_{aa} & \Omega_{ab} \\ \Omega_{ba} & \Omega_{bb} - \delta \end{bmatrix}$$

$$\Omega_{aa} = \frac{1}{2}\sum_q a_q \left|E_q\right|^2$$

$$\Omega_{bb} = \frac{1}{2}\sum_q b_q \left|E_q\right|^2$$

$$\Omega_{ab} = \Omega_{ba}^* = \frac{1}{2}\sum_q d_q E_q E_{q+1}^*$$

$$a_q = \frac{1}{2\hbar^2}\sum_j \left(\frac{\left|\mu_{ja}\right|^2}{\omega_j - \omega_a - \omega_q} + \frac{\left|\mu_{ja}\right|^2}{\omega_j - \omega_a + \omega_q} \right)$$

$$b_q = \frac{1}{2\hbar^2}\sum_j \left(\frac{\left|\mu_{jb}\right|^2}{\omega_j - \omega_b - \omega_q} + \frac{\left|\mu_{jb}\right|^2}{\omega_j - \omega_b + \omega_q} \right)$$

$$d_q = \frac{1}{2\hbar^2}\sum_j \left(\frac{\mu_{aj}\mu_{jb}}{\omega_j - \omega_b - \omega_q} + \frac{\mu_{aj}\mu_{jb}}{\omega_j - \omega_a + \omega_q} \right)$$

Coherent Molecular Excitation

Solution to the Hamiltonion:

- Eigenfunctions $\left|\pm\right\rangle = \cos\theta^{(\pm)}\left|a\right\rangle + \sin\theta^{(\pm)}e^{-i\varphi}\left|b\right\rangle \qquad \Omega_{ab} = \left|\Omega_{ab}\right|e^{i\varphi}$

$$\tan\theta^{(\pm)} = \frac{2\left|\Omega_{ab}\right|}{\Omega_{aa} - \Omega_{bb} + \delta \pm \sqrt{\left(\Omega_{aa} - \Omega_{bb} + \delta\right)^2 + 4\left|\Omega_{ab}\right|^2}}$$

- Eigenvalues: $E_{eff} = -\hbar\lambda^{(\pm)} = \frac{\hbar}{2}\left(\Omega_{aa} + \Omega_{bb} - \delta\right) \pm \frac{\hbar}{2}\sqrt{\left(\Omega_{aa} - \Omega_{bb} + \delta\right)^2 + 4\left|\Omega_{ab}\right|^2}$

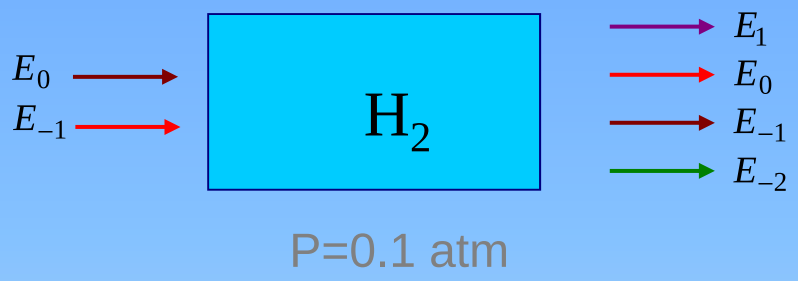
- Coherence:

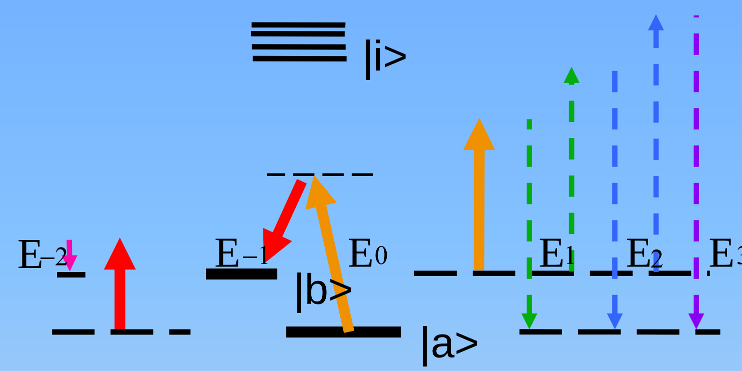
$$\rho_{ab}^{(\pm)} = \frac{1}{2}e^{i\varphi}\sin 2\theta^{(\pm)} = \pm\frac{\Omega_{ab}}{\sqrt{\left(\Omega_{aa} - \Omega_{bb} + \delta\right)^2 + 4\left|\Omega_{ab}\right|^2}}$$

$$\left|\rho_{ab}\right| = 0.5 \qquad \left|\Omega_{ab}\right| >> \left|\Omega_{aa} - \Omega_{bb} + \delta\right|$$

Sideband Generation and Propagation

- Adiabatically prepared molecules modulate the driving fields producing a wide comb





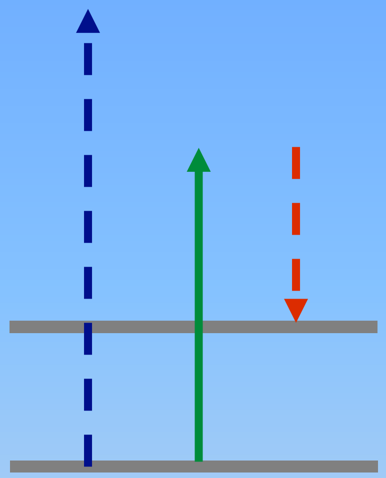
- Propagation equation for the *q*th sideband: $\omega_q = \omega_{q-1} + \omega_0 - \omega_{-1}$

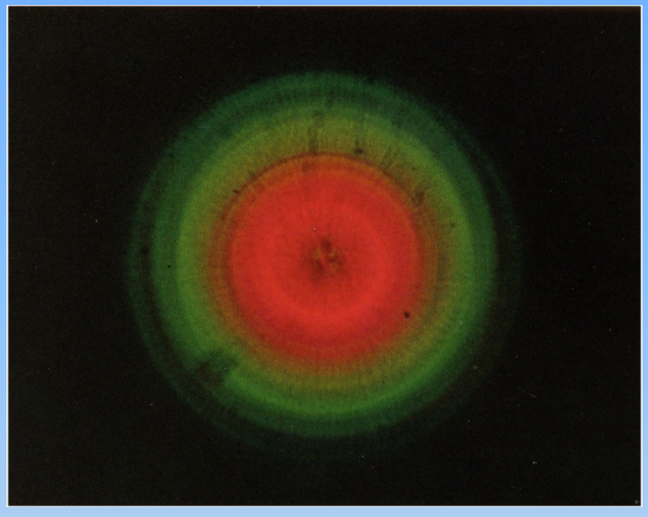
$$\frac{\partial E_q}{\partial z} = -j\eta\hbar\omega_q N\left(\underbrace{a_q\rho_{aa}E_q + d_q\rho_{bb}E_q}_{\text{dispersion}} + \underbrace{b_q^*\rho_{ab}E_{q-1} + c_q^*\rho_{ab}^*E_{q+1}}_{\text{coupling}}\right)$$

At maximum coherence, $\rho_{ab} = 0.5$ the dispersion and coupling terms become comparable. Phase-matching is then not important, and generation is collinear.

Courtesy M. Shverdin

Stimulated Raman Scattering





Traditional SRS:

- ★ Generation occurs at high gas pressure
- ★ Molecular excitation occurs on-resonance
- ★ Anti-Stokes generation occurs off-axis
- ★ Few Stokes and anti-Stokes orders are observed.

Courtesy M. Shverdin

Why low temperature

1. Put all molecules into one ro-vibrational state
   make all molecules contribute to the process

$$\frac{\partial E_q}{\partial \xi} = \frac{iN\hbar\omega_q}{\varepsilon_0 c}\left(a_q \rho_{aa} E_q + b_q \rho_{bb} E_q + d_{q-1} \rho_{ba} E_{q-1} + d_q^* \rho_{ab} E_{q+1}\right)$$

2. Reduce Doppler width to increase the coherence
   molecules with equal detuning but opposite in sign will
   off-set their contribution to the coherence build up

$$\Delta \nu_D = \frac{\nu_0}{c}\sqrt{\frac{2kT}{m}} = \frac{1}{\lambda_0}\sqrt{\frac{2kT}{m}} = 1.285\times 10^{11}\frac{1}{\lambda_0(nm)}\sqrt{\frac{T(K)}{\mu}}$$

$$a_q = \frac{1}{2\hbar^2}\sum_j\left(\frac{|\mu_{ja}|^2}{\omega_j - \omega_a - \omega_q} + \frac{|\mu_{ja}|^2}{\omega_j - \omega_a + \omega_q}\right)$$

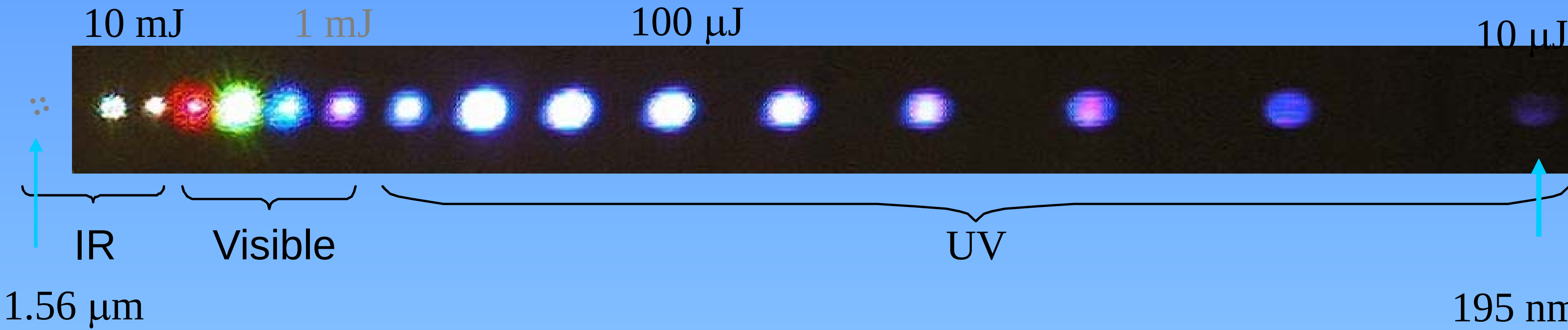
$$b_q = \frac{1}{2\hbar^2}\sum_j\left(\frac{|\mu_{jb}|^2}{\omega_j - \omega_b - \omega_q} + \frac{|\mu_{jb}|^2}{\omega_j - \omega_b + \omega_q}\right)$$

$$d_q = \frac{1}{2\hbar^2}\sum_j\left(\frac{\mu_{aj}\mu_{jb}}{\omega_j - \omega_b - \omega_q} + \frac{\mu_{aj}\mu_{jb}}{\omega_j - \omega_a + \omega_q}\right)$$

T = 300K $\Delta\nu_D$, D_2 = 778 MHz
T = 77K $\Delta\nu_D$, D_2 = 389 MHz

T = 300K $\Delta\nu_D$, H_2 = 1100 MHz

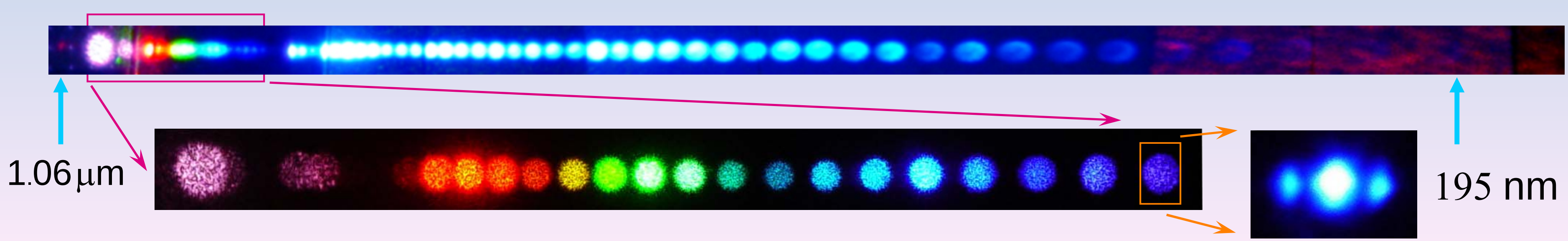
D_2 Vibration Spectra: 16 sidebands, spaced by 2994 cm^{-1}



H_2 Rotation Spectra: 29 sidebands, spaced by 587 cm^{-1}

Phys. Rev. A (R)(1997)
Phys. Rev. Lett. 81 (1998)
Opt. Lett. 24 (1999)
Phys. Rev. Lett. 84 (2000)
Phys. Rev. Lett. 85 (2000)
Phys. Rev. A 63 (2001)
Phys. Rev. Lett. 91 (2003)
Phys. Rev. Lett. 93 (2005)

Multiplicative Spectra: ~ 200 sidebands, spaced by < 587 cm^{-1}



Motivation

- **form subfemtosecond pulses**
- **produce THz pulse train**
- **generate tunable high power vacuum uv pulses**
- **arbitrary waveform synthesis**

Use gas phase hydrogen at room temperature

Pros:

- ***large* Raman transition of 4155 cm^{-1} for Q(1)**
- ***2/3* population in single quantum state (v=0, j=1) at room temperature**
- ***many parameters are known***
- ***nondestructible***
- ***room temperature is easy to operate***

Cons:

- ***large Doppler* width**
- ***requires two tunable high-power high- resolution lasers spaced 4155 cm^{-1} apart***
- ***smaller pulse train pulse-to-pulse spacing***

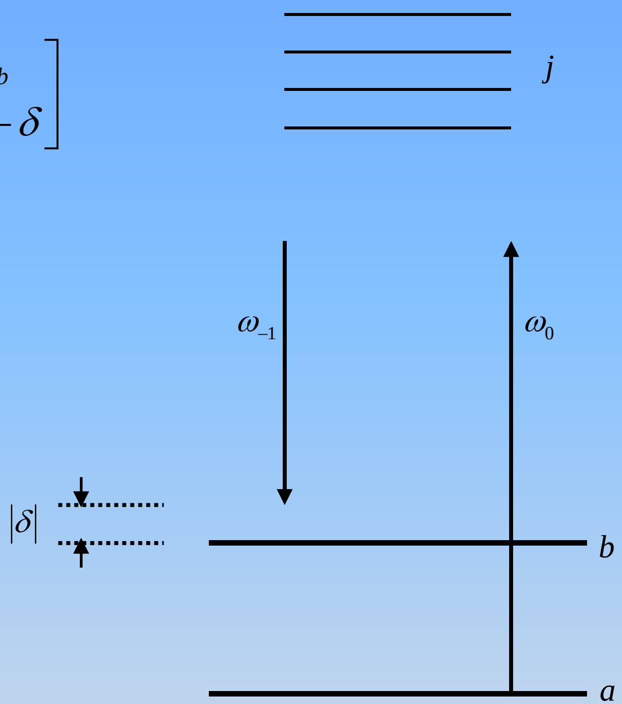
Simulation

$$\Omega_{aa} = \frac{1}{2}\sum_q a_q \left|E_q\right|^2$$

$$\Omega_{bb} = \frac{1}{2}\sum_q b_q \left|E_q\right|^2$$

$$\Omega_{ab} = \Omega_{ba}^* = \frac{1}{2}\sum_q d_q E_q E_{q+1}^*$$

$$H_{eff} = -\hbar\begin{bmatrix} \Omega_{aa} & \Omega_{ab} \\ \Omega_{ba} & \Omega_{bb} - \delta \end{bmatrix}$$



$$a_q = \frac{1}{2\hbar^2}\sum_j \left(\frac{\left|\mu_{ja}\right|^2}{\omega_j - \omega_a - \omega_q} + \frac{\left|\mu_{ja}\right|^2}{\omega_j - \omega_a + \omega_q} \right)$$

$$b_q = \frac{1}{2\hbar^2}\sum_j \left(\frac{\left|\mu_{jb}\right|^2}{\omega_j - \omega_b - \omega_q} + \frac{\left|\mu_{jb}\right|^2}{\omega_j - \omega_b + \omega_q} \right)$$

$$d_q = \frac{1}{2\hbar^2}\sum_j \left(\frac{\mu_{aj}\mu_{jb}}{\omega_j - \omega_b - \omega_q} + \frac{\mu_{aj}\mu_{jb}}{\omega_j - \omega_a + \omega_q} \right)$$

$$\frac{\partial \rho_{aa}}{\partial \tau} = i\left(\Omega_{ab}\rho_{ba} - \Omega_{ba}\rho_{ab}\right) + \gamma_{\parallel}\rho_{bb}$$

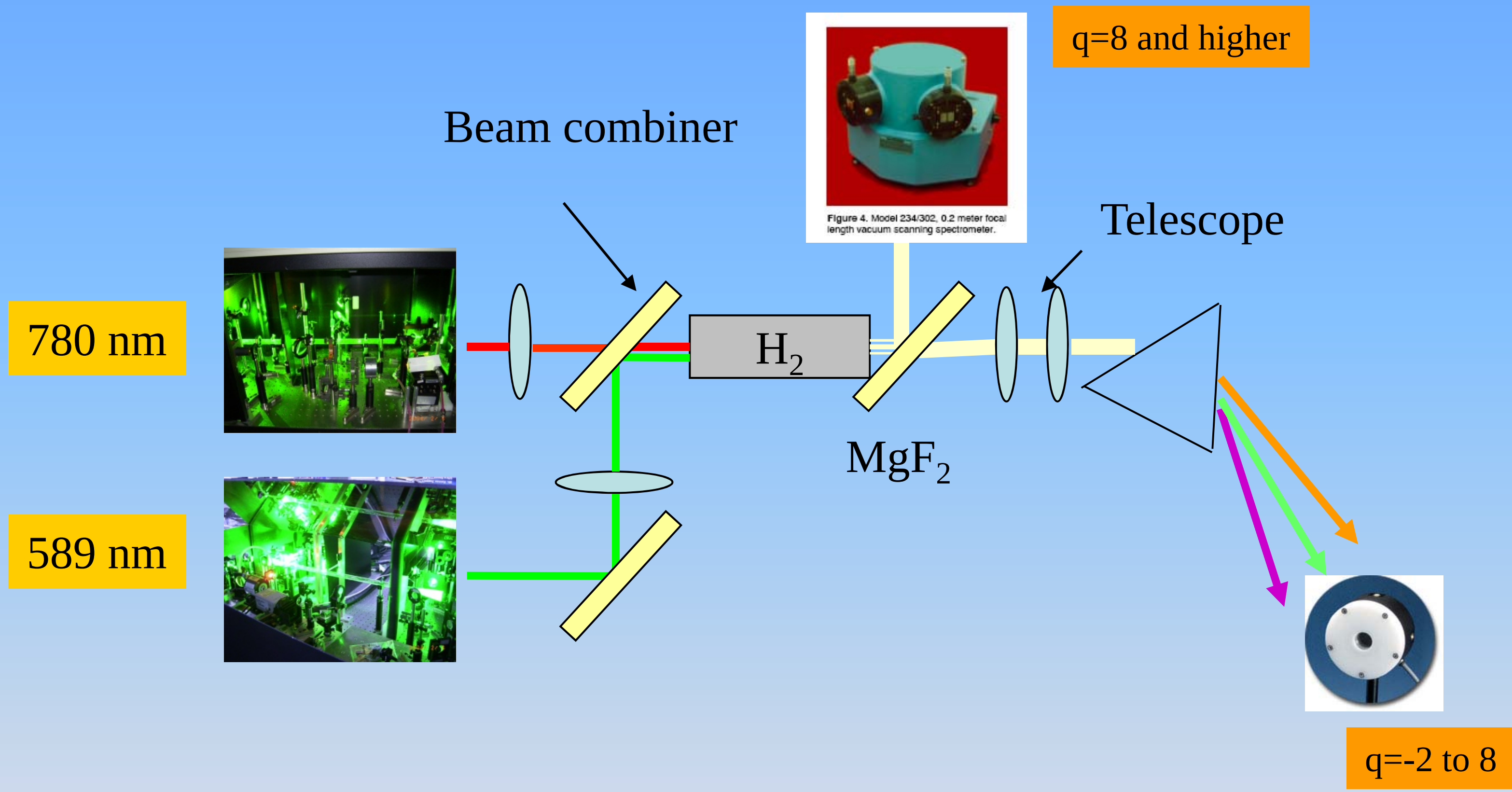
$$\frac{\partial \rho_{bb}}{\partial \tau} = -i\left(\Omega_{ab}\rho_{ba} - \Omega_{ba}\rho_{ab}\right) - \gamma_b\rho_{bb}$$

$$\frac{\partial \rho_{ab}}{\partial \tau} = i\left(\Omega_{aa} - \Omega_{bb} + \delta + i\gamma_{\perp}\right)\rho_{ab} + i\Omega_{ab}\left(\rho_{bb} - \rho_{aa}\right)$$

Thanks to Prof. Fam Le Kien for the full set of matrix element data

$$\frac{\partial E_q}{\partial z} = -j\eta\hbar\omega_q\left[N(a_q\rho_{aa}E_q + d_q\rho_{bb}E_q) + 0.666N(b_q^*\rho_{ab}E_{q-1} + b_{q+1}\rho_{ab}^*E_{q+1})\right]$$

Experimental Arrangement

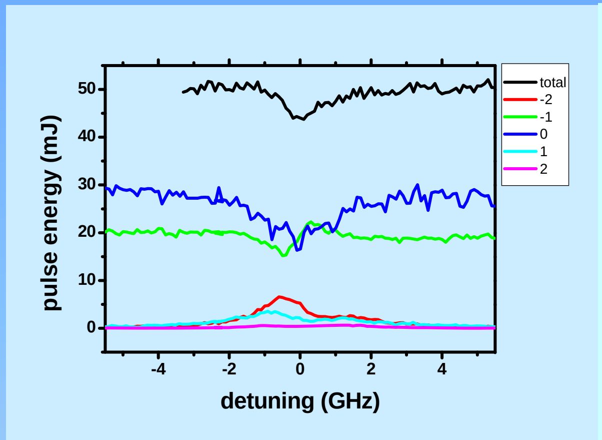


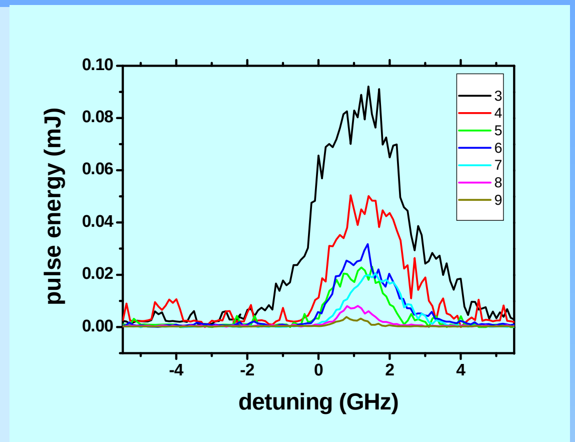
$\delta\omega_{laser}$ ~ 100 MHz
Δt_{laser} ~5 ns
λ_0=589 nm
λ_{-1}=780 nm

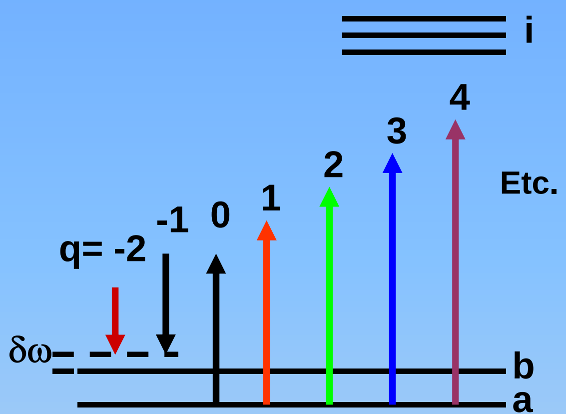
Intensity ~ 12 GW/cm^2
$\Omega_{ab,z=0}$~2 GHz
ρ_{ab} ~0.4
t_{delay}~0-1 ns

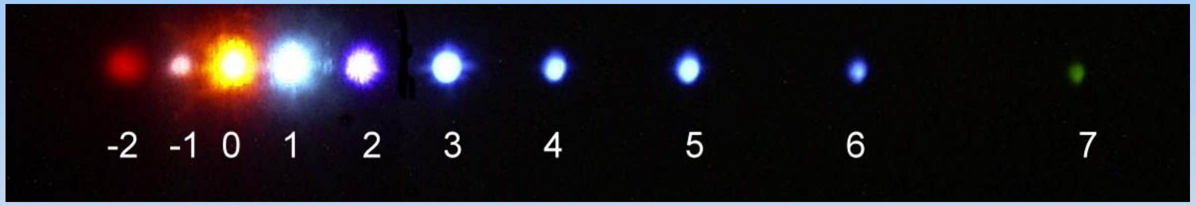
H_2 pressure ~1 atm.
H_2 Doppler width 250-750 MHz

Raman sidebands generated





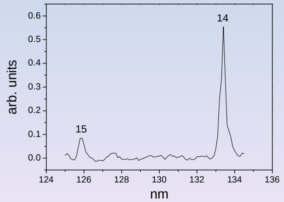




q	λ	
q = 3, λ =	339.6	nm
6	238.6	nm
9	183.9	nm
12	149.6	nm
14	133.0	nm

Total spectral span >70,000 cm^{-1}
(~500 as)

15^{th} order at 126 nm observed



$$\omega_q = n\omega_m$$

Note:

589 nm $\leftrightarrow$ 16978 cm^{-1} (4 x 4155.2 = 16621 cm^{-1})

780 nm $\leftrightarrow$ 12822.8 cm^{-1} (3 x 4155.2 cm^{-1} = 12465.6 cm^{-1})

The sidebands are not commensurate.

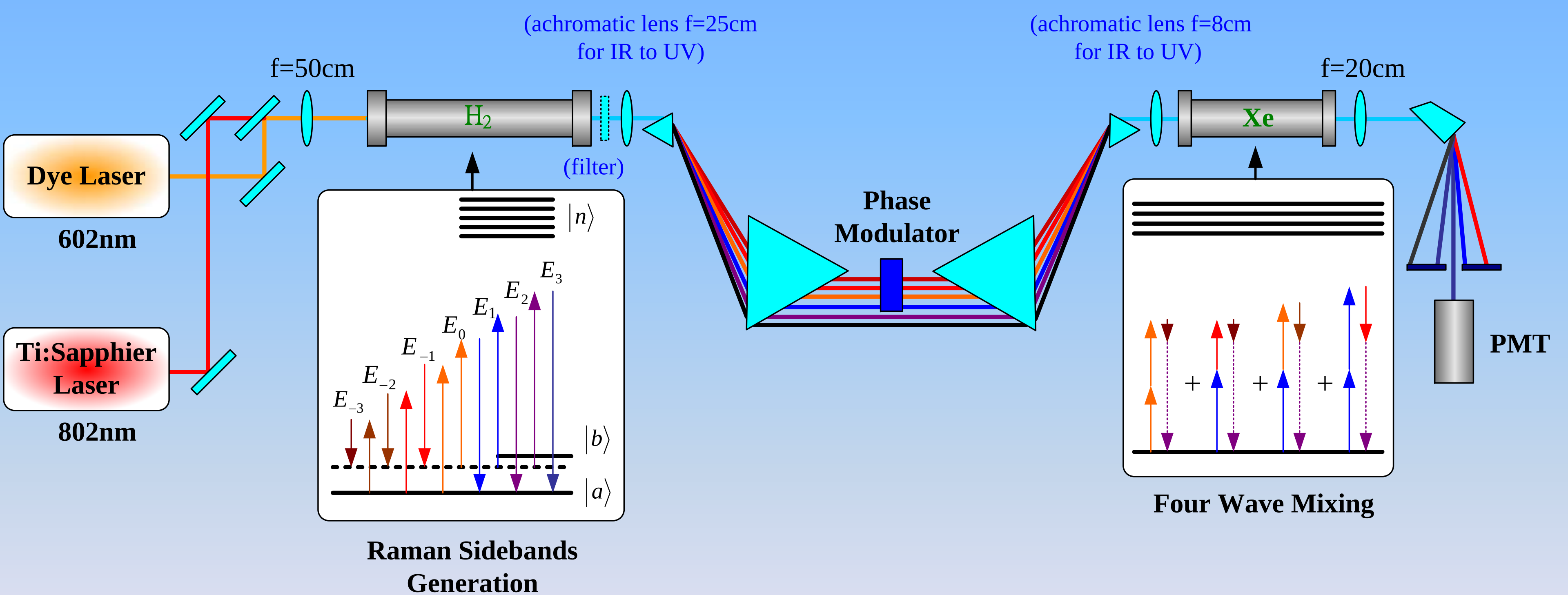
New input wavelengths:

ω_0 = 16621 cm^{-1} (602 nm)
ω_{-1} = 12465.6 cm^{-1} (802 nm)

These wavelengths produce a commensurate set of sidebands, as shown on the right:

Raman Order	nm	cm^{-1}	4 wave-mixing order
	∞	0	
-3	2407	4155	
-2	1203	8310	1
-1	802	12465	2
0	602	16620	3
1	481	20775	4
2	401	24930	5
3	344	29085	6
4	301	33240	7
5	267	37395	8
6	241	41550	9
7	219	45705	10
8	201	49860	11
9	185	54015	

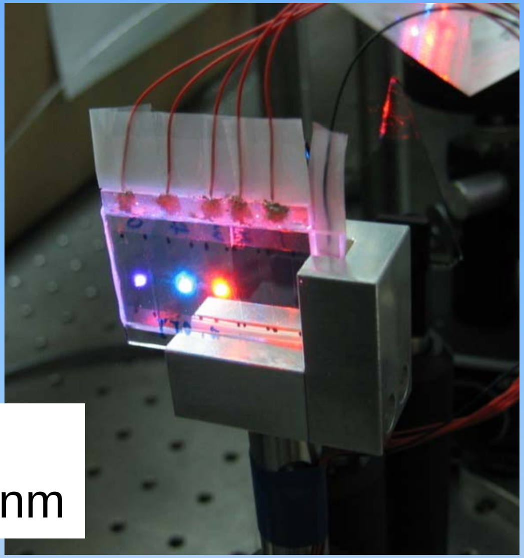
Phase adjustment Setup



M. Y. Shverdin et al. PRL **94**, 033904 (2005)

LC phase modulator

→

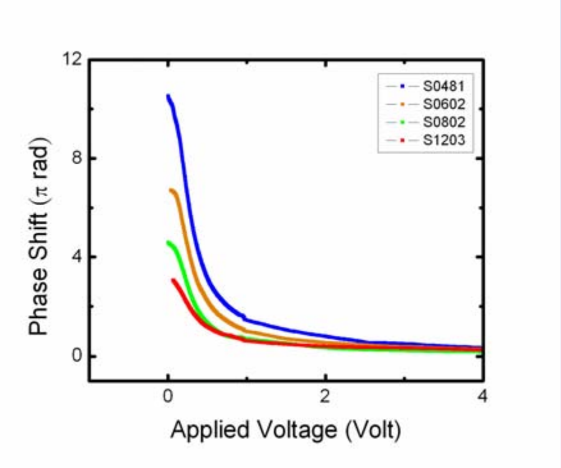


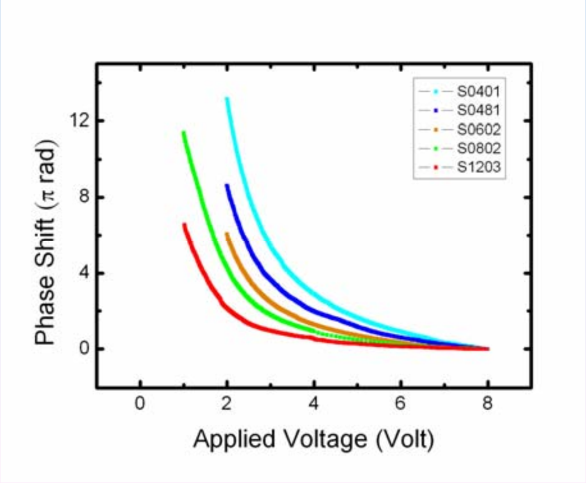
λ_{cutoff}

430 nm --> 380 nm

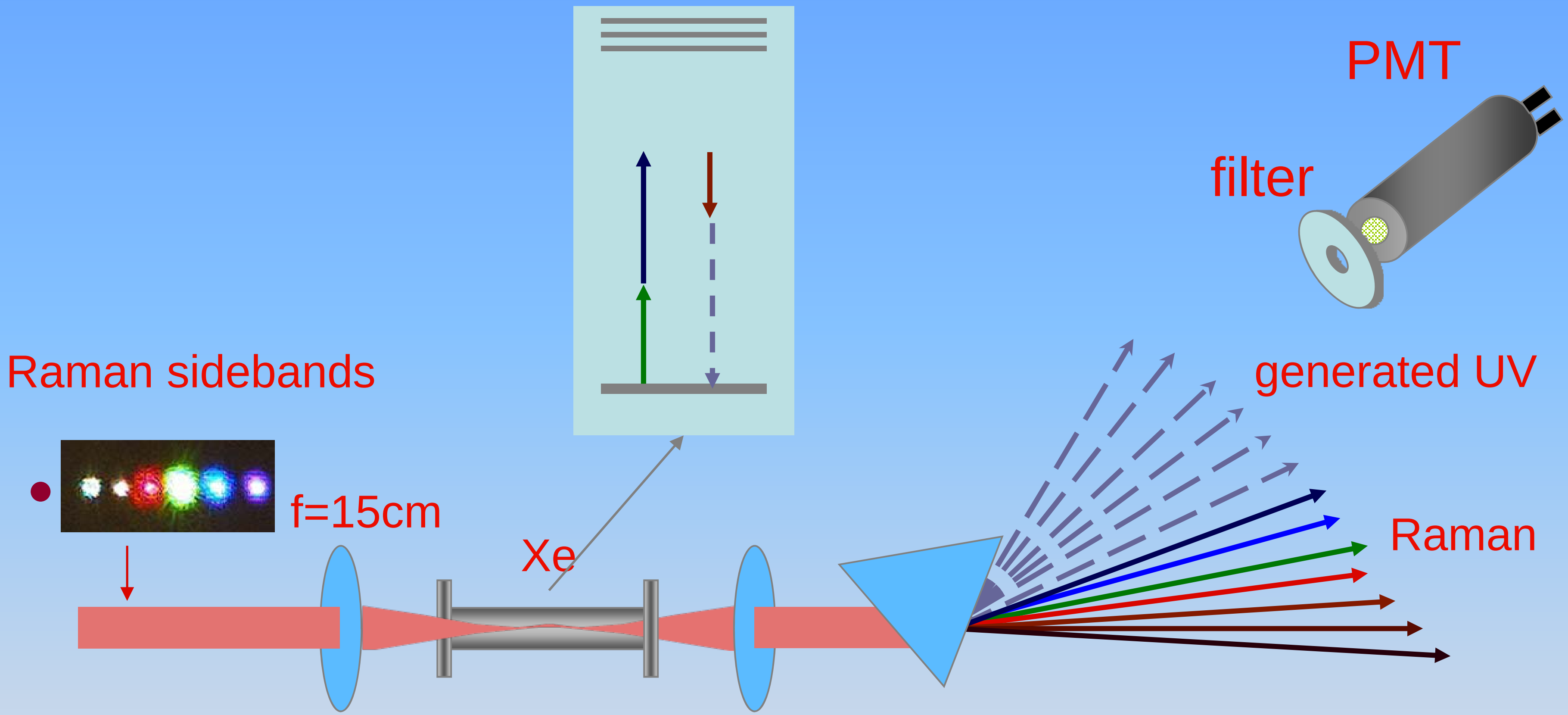
6 sidebands

7 sidebands





Four-wave Mixing



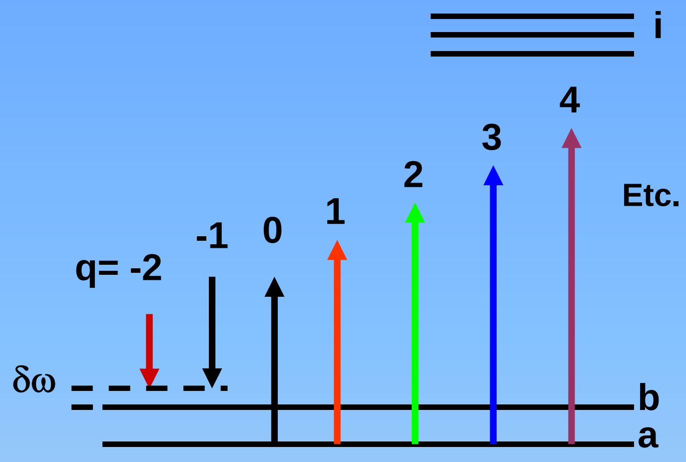
UV sidebands are generated at efficiencies 10^{-8} to 10^{-12} by a four-wave mixing process in a xenon cell at ~100 torr. Phasematching allows only the two photons up one photon down type of conversion. A total of n-1 different UV sidebands, beginning at the next short wavelength sideband, are generated.

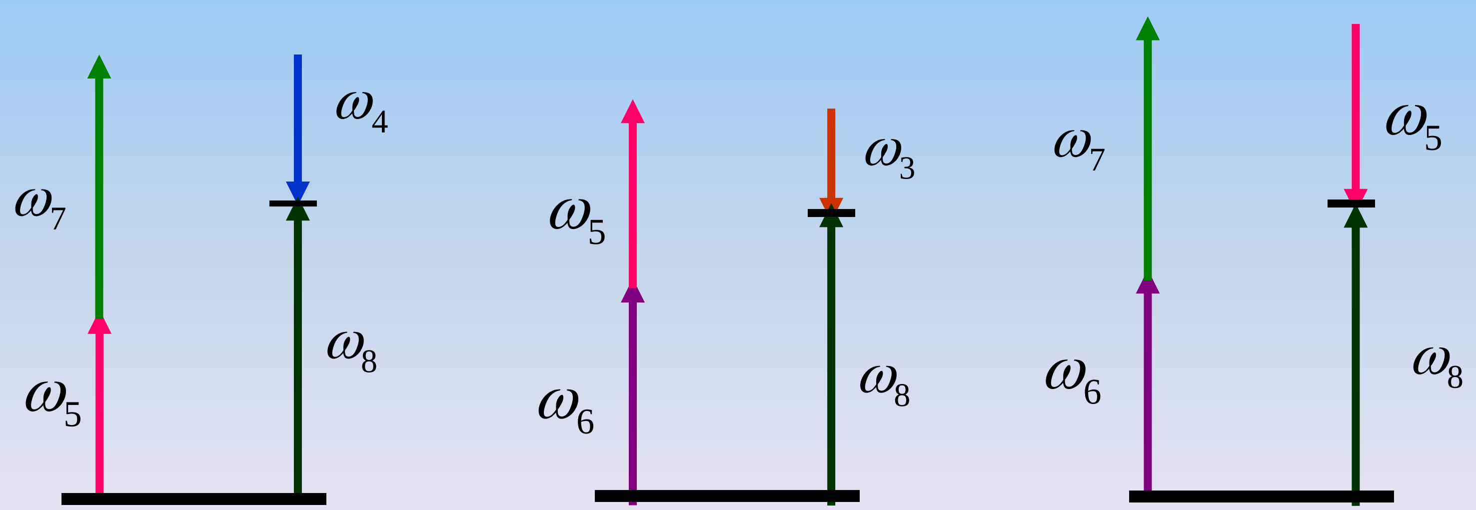
Multiple quantum paths interference

Four wave mixing: $\omega_5 + \omega_7 - \omega_4 = \omega_8$

$$\omega_6 + \omega_5 - \omega_3 = \omega_8$$

$$\omega_6 + \omega_7 - \omega_5 = \omega_8$$





Four Wave Mixing in Xe

$$E_\alpha = \chi E_i E_j E_k^* \qquad\qquad \phi_\alpha = \phi_i + \phi_j - \phi_k$$

7 :

$5+5-3$

$5+4-2$

$5+3-1$

$4+4-1$

$$\Rightarrow \begin{cases} \phi_5 - \phi_3 = \phi_4 - \phi_2 = \phi_3 - \phi_1 = 2s \\ \phi_5 - \phi_2 = \phi_4 - \phi_1 = 3t \end{cases}$$

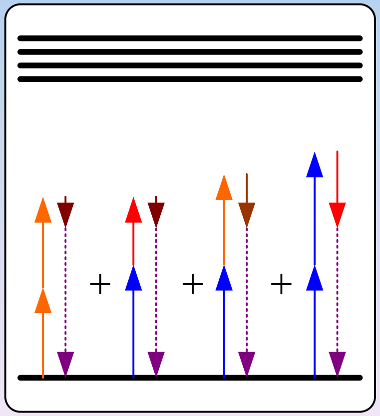
$$\phi_5 = \phi_1 + 4s \qquad\qquad \phi_4 = \phi_2 + 2s$$

$$-)\ \phi_5 = \phi_2 + 3t \qquad\qquad -)\ \phi_4 = \phi_1 + 3t$$

$$\Rightarrow \phi_1 - \phi_2 = 3t - 4s \qquad\qquad \phi_1 - \phi_2 = 2s - 3t$$

$$\Rightarrow 3t - 4s = 2s - 3t$$

$$t = s$$



$$\Rightarrow \begin{cases} \varphi_2 = \varphi_1 + \Delta \\ \varphi_3 = \varphi_1 + 2\Delta \\ \varphi_4 = \varphi_1 + 3\Delta \\ \varphi_5 = \varphi_1 + 4\Delta \end{cases}$$

In phase condition

1: 1203nm
2: 802nm
3: 602nm
4: 481nm
5: 401nm
6: 344nm
7: 301nm

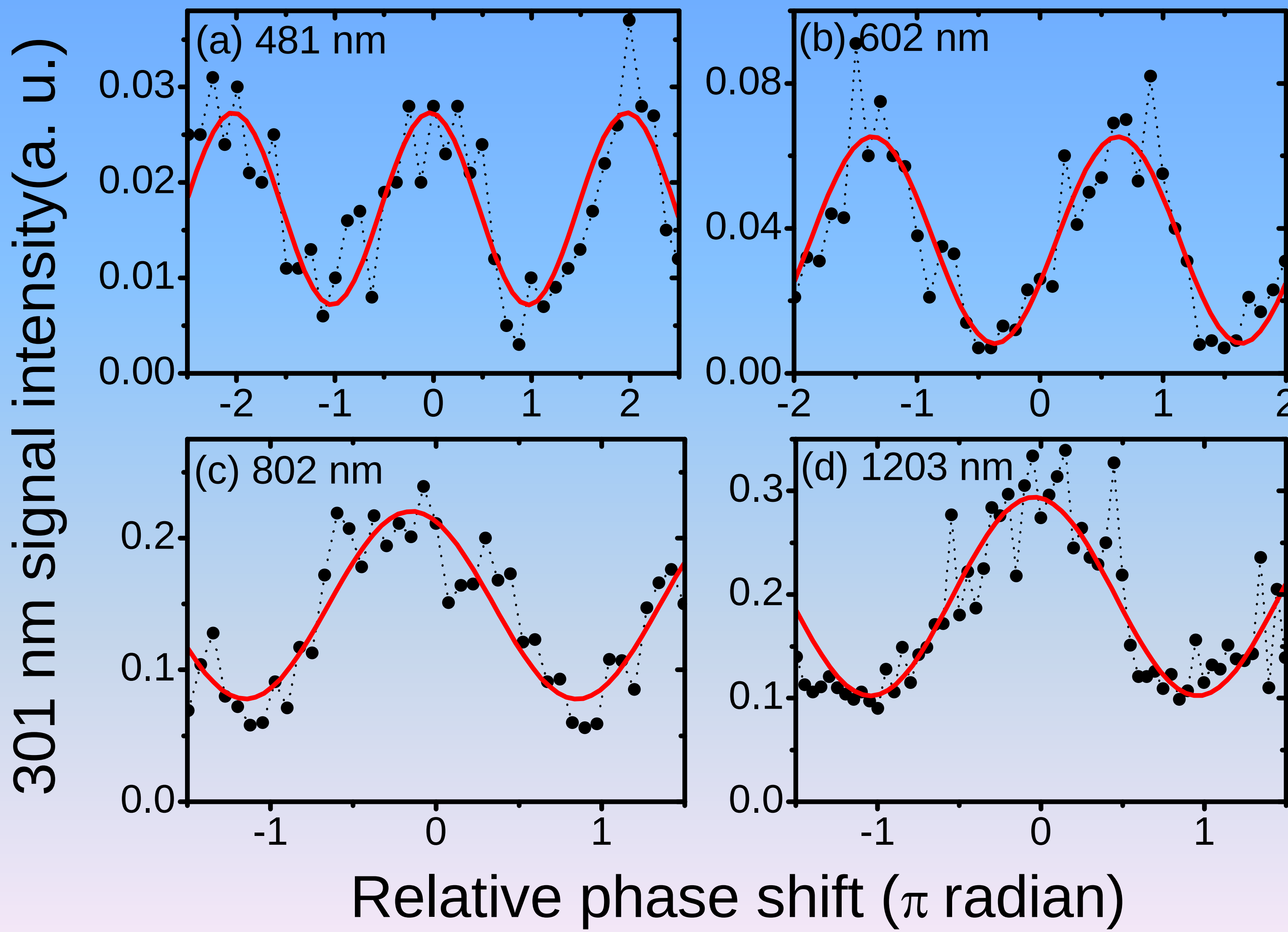
7=6+6-5, 6+5-4, 6+4-3, 6+3-2, 6+2-1.

5+5-3, 5+4-2, 5+3-1.

4+4-1

6, 5, 4	: 6+6-5, 6+5-4	$\Phi_{65}=\Phi_{54}$
+3	: 6+4-3, 5+5-3	$\Phi_{65}=\Phi_{54}=\Phi_{43}$
+2	: 6+3-2, 5+4-2	$\Phi_{65}=\Phi_{54}=\Phi_{43}=\Phi_{32}$
+1	: 6+2-1, 5+3-1, 4+4-1	$\Phi_{65}=\Phi_{54}=\Phi_{43}=\Phi_{32}=\Phi_{21}$

Searching in phase condition



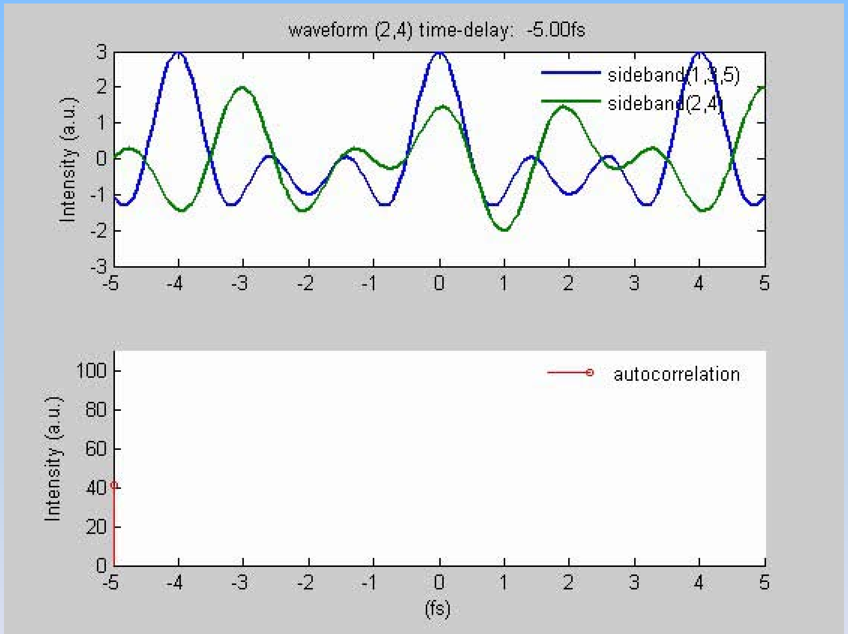
How to measure the pulse width?

Autocorrelation is standard way to measure ultrafast pulsewidth. However it could not be done here because of the wide bandwidth.

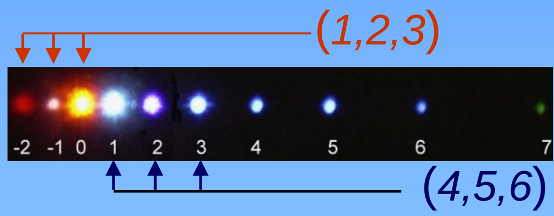
Solution: Correlation using pulses formed by the sidebands themselves.

Synthesize two pulses from the subsets of sidebands and electronically delay one pulse with respect to the other. Measure the resulting four-wave signal with a photomultiplier.

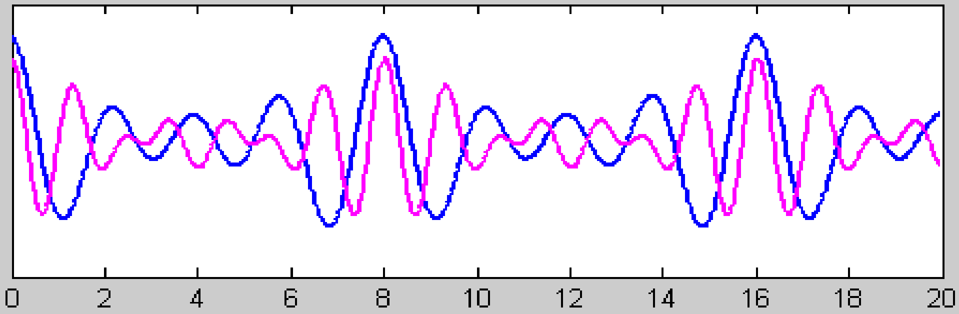
simulation



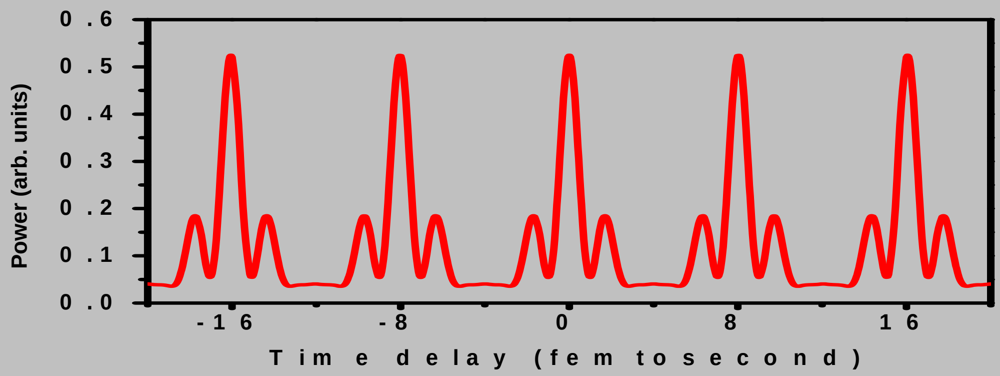
Cross Correlation of Single Cycle Pulse Train



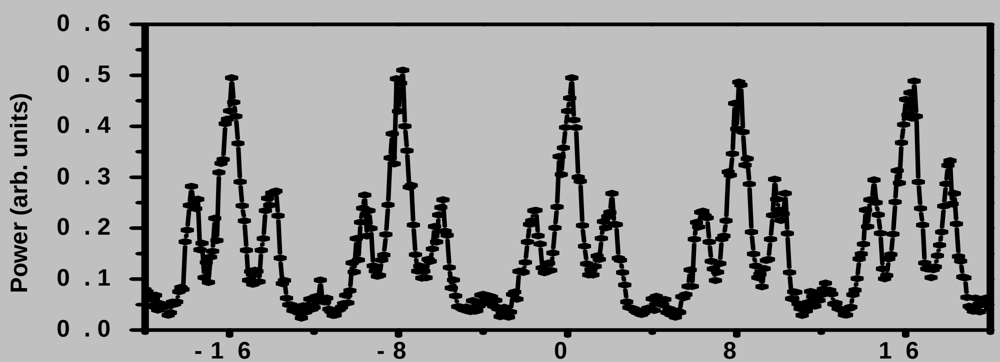
Sideband Orders



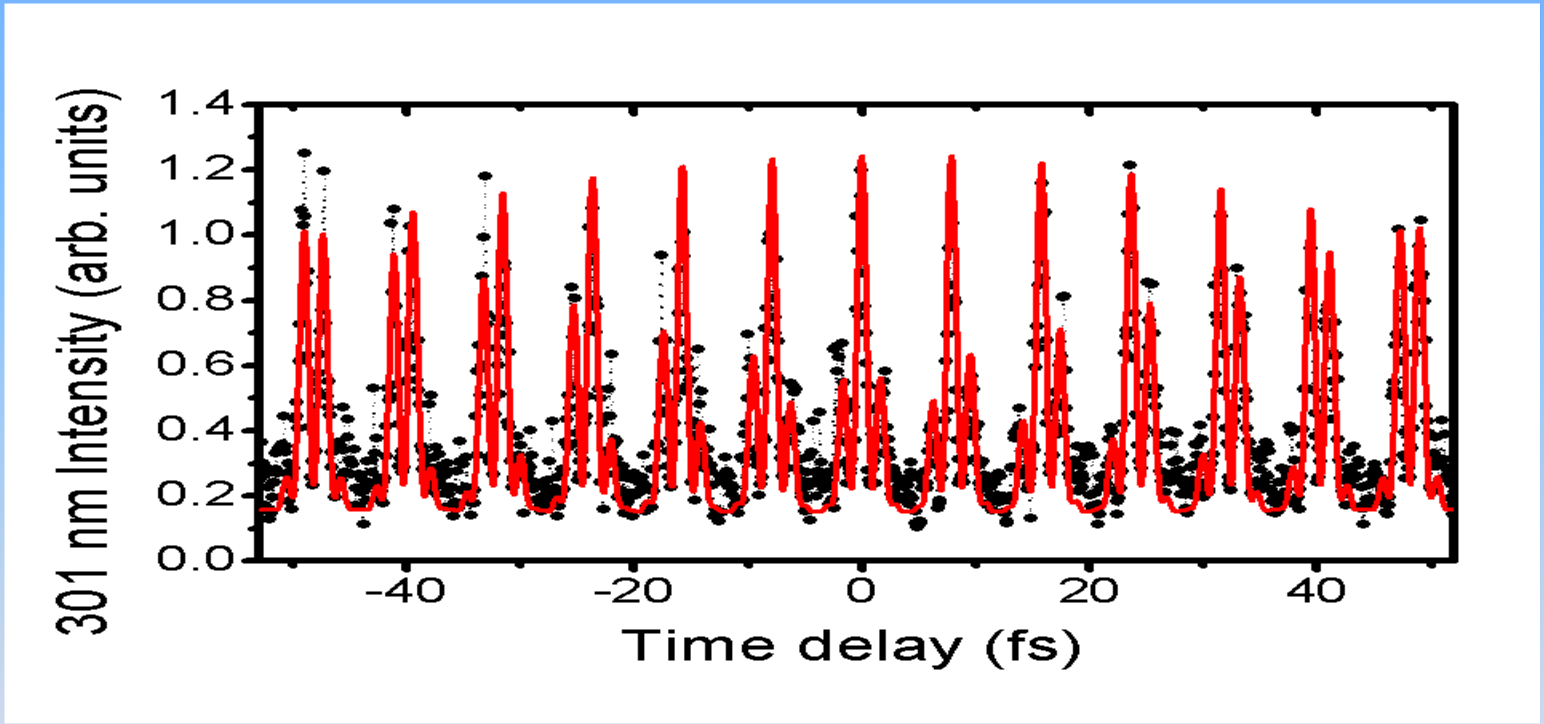
Simulation →



Experiment →

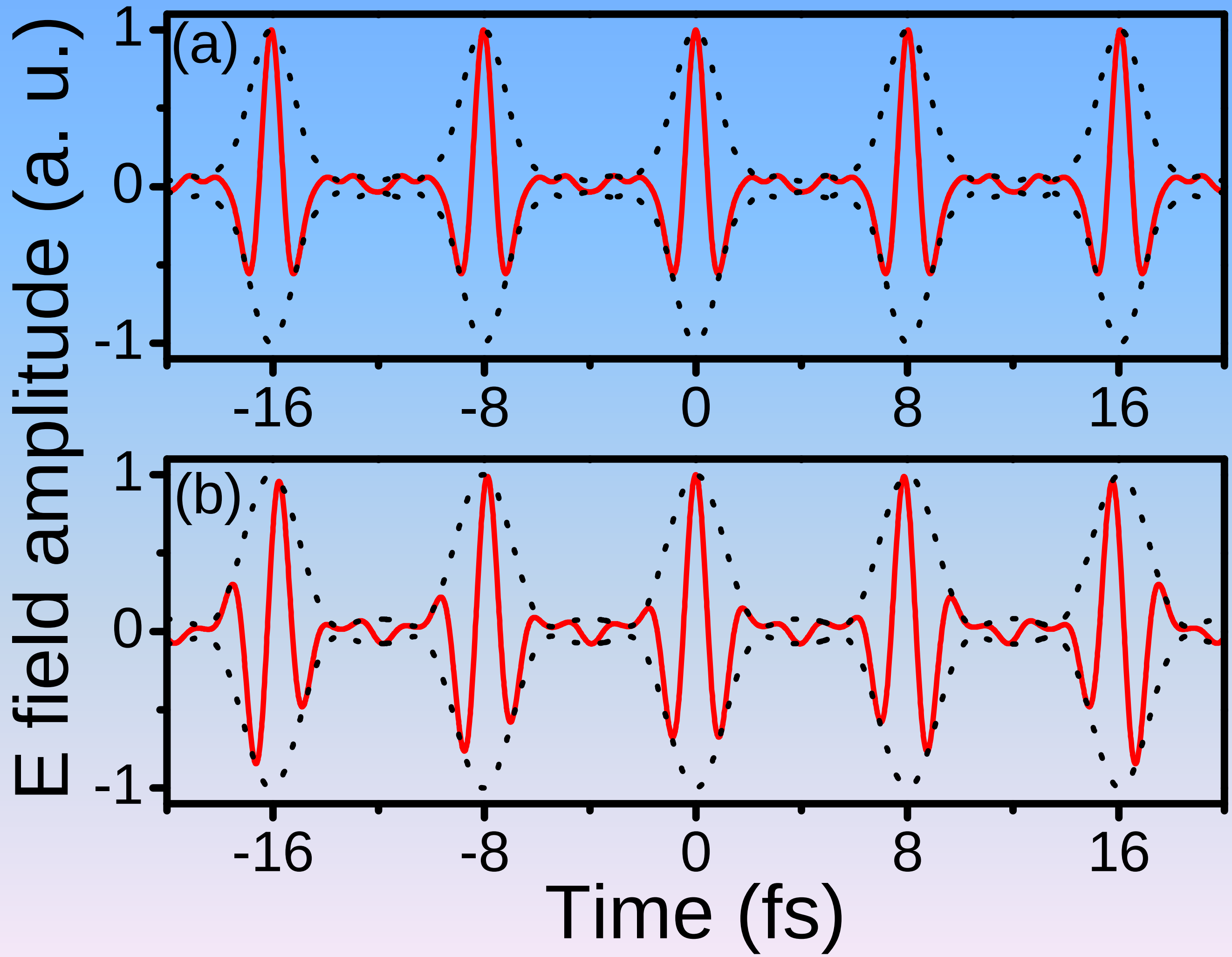


Cross correlation signal of incommensurate pulses



CEO frequency ~ 349 cm-1
Waveform repeats every 96 fs

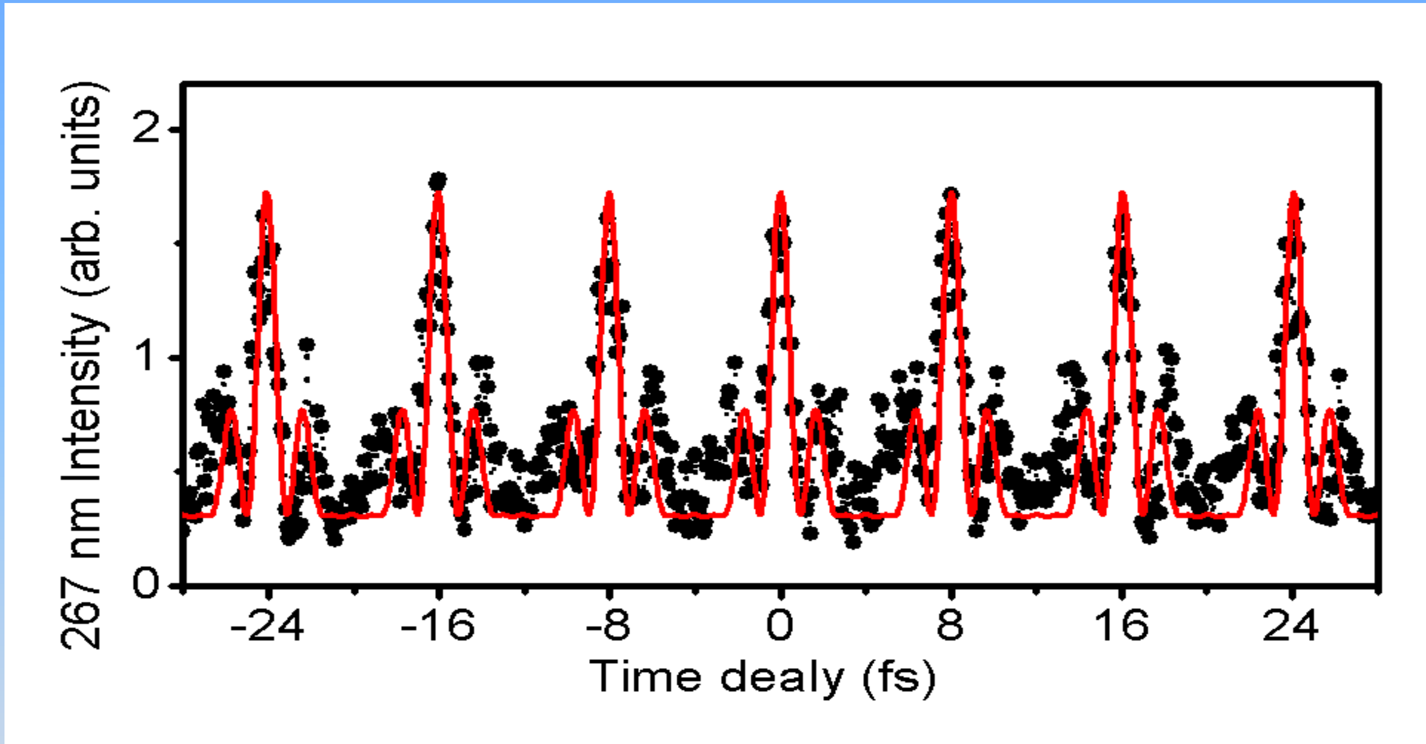
Pulse train



Commensurate

Incommensurate

7 beam correlation in Xe



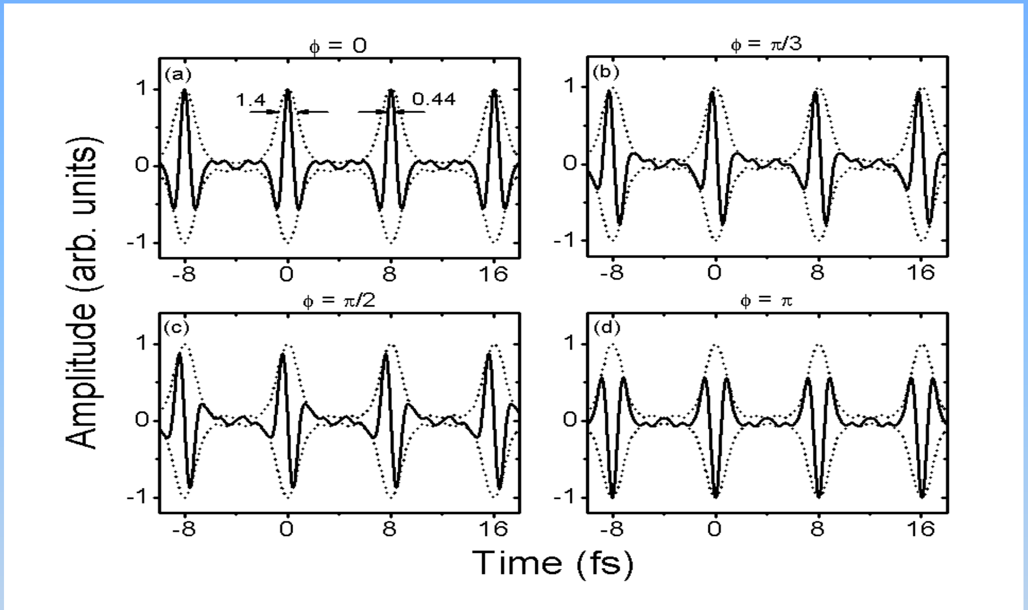
Carrier envelope phase is constant to ~2.5 part in 10^6

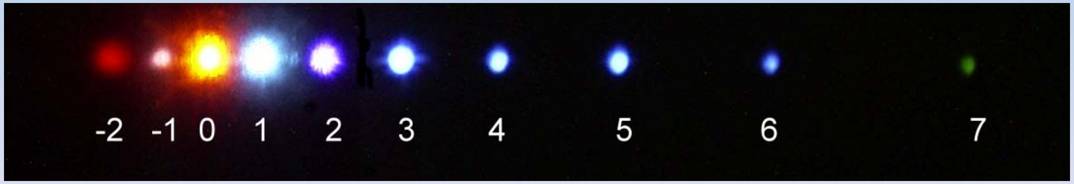
Total phase slip of <0.18 cycles over 1 million pulses

Status of sub-cycle optical pulse generation by molecular modulation

IAMS sub-cycle source

0.833 cycle per pulse
1.4 fs envelope
440 as cycle width
constant carrier envelope phase
2 ns pulse train duration
8.0 fs pulse spacing
~1 MW peak power





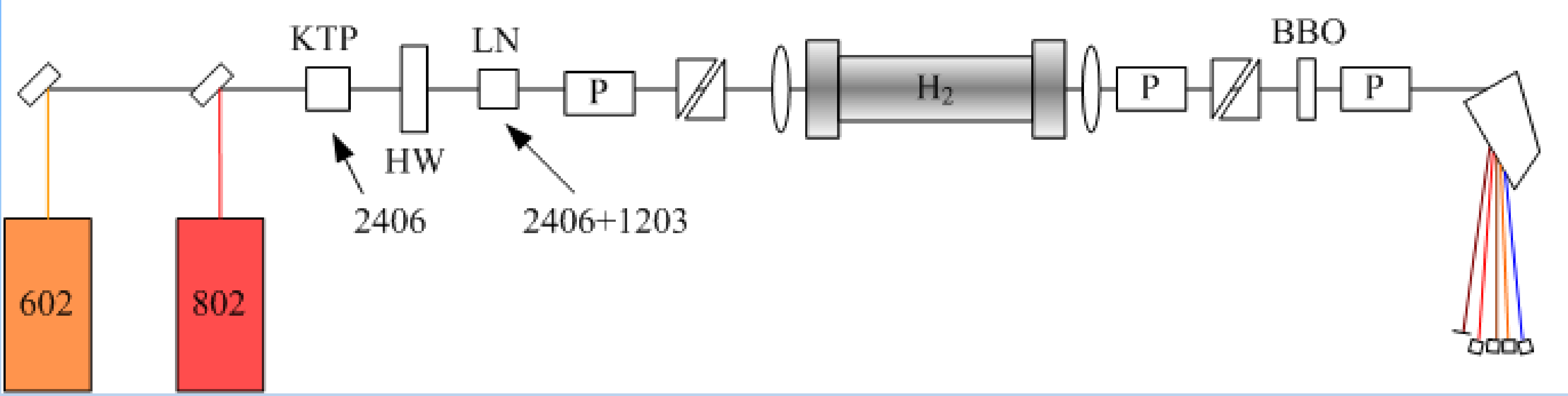
Total spectral span >70,000 cm^{-1}

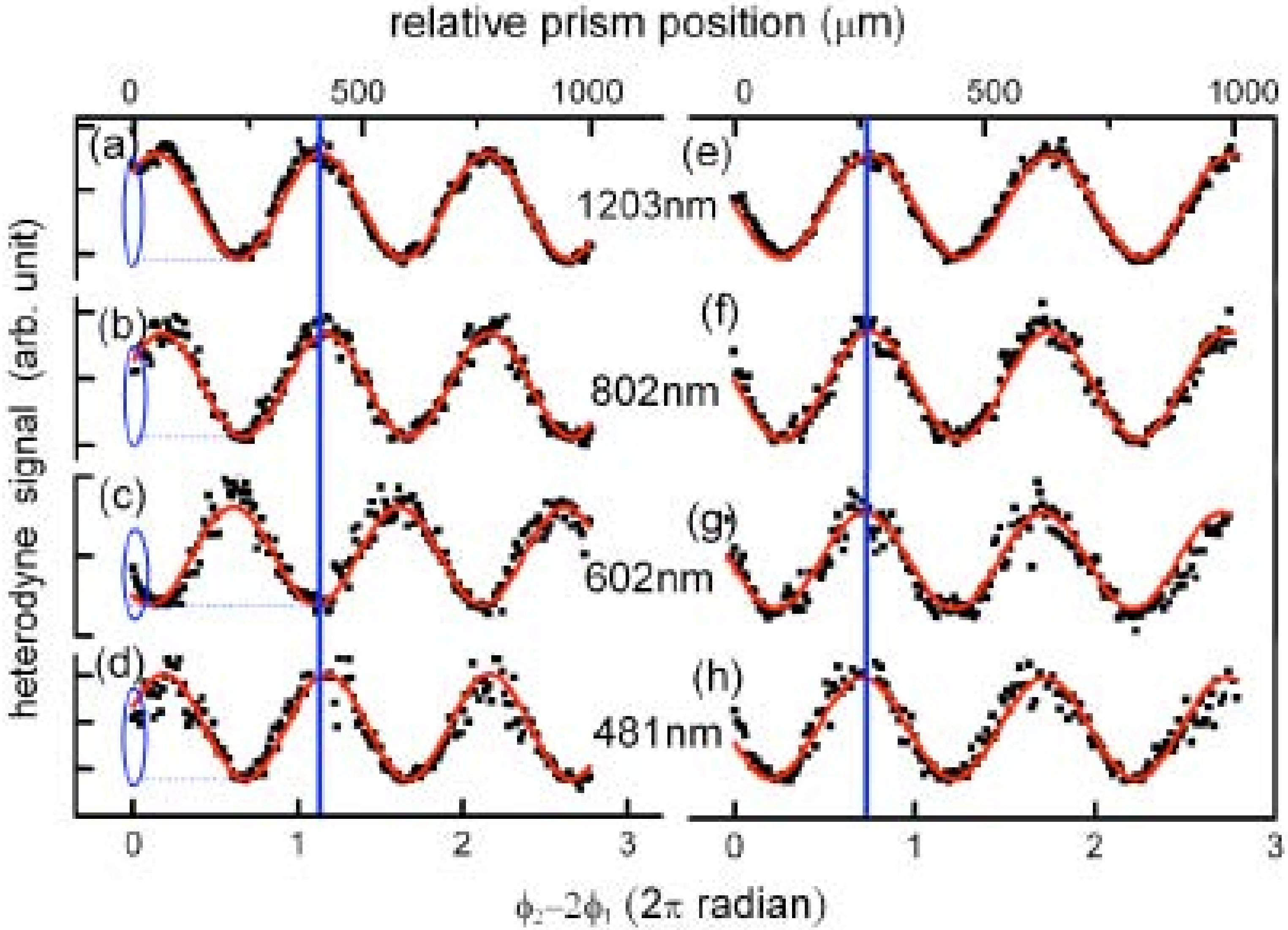
Phys. Rev. Lett. 100, 163906 (2008)

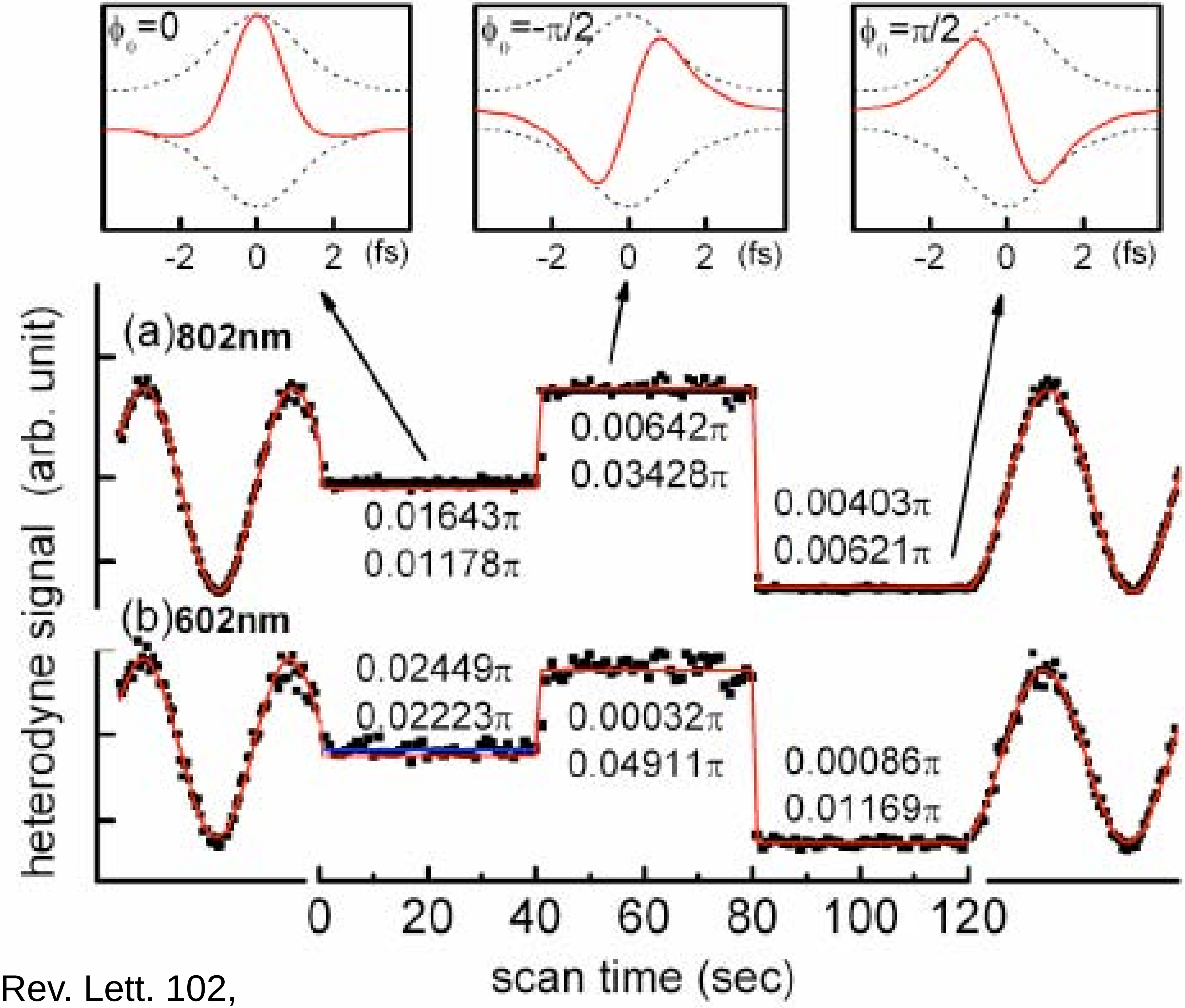
Ingredients of an attosecond single-cycle optical pulse:

1. Broad spectrum – 2 or more octaves
2. In phase condition
3. Constant carrier envelope phase:
    - Commensurate frequencies
    - Constant phase difference between adjacent spectral components
4. **Stable and controllable carrier envelope phase**

CEP control




relative prism position (μm)
0
500
1000
0
500
1000
(a)
(b)
(c)
(d)
(e)
(f)
(g)
(h)
1203nm
802nm
602nm
481nm
heterodyne signal (arb. unit)
0
1
2
3
0
1
2
3
φ2−2φ1 (2π radian)



Phys. Rev. Lett. 102, 213902 (2009)

Summary and Outlook

- Generated commensurate pulse train
- Single pulse duration 1.4fs
- Sub-single-cycle pulse: 0.8 cycles
- CEP (carrier-envelope phase) control

- Sub-femtosecond pulse generation
- Arbitrary waveform
- Application for ultra-fast dynamics

Harmonics

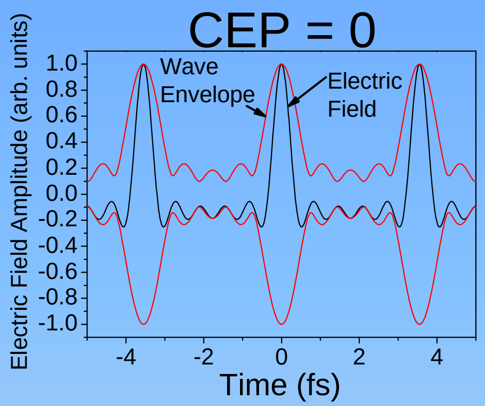
1203
802
602
481
401
344
301

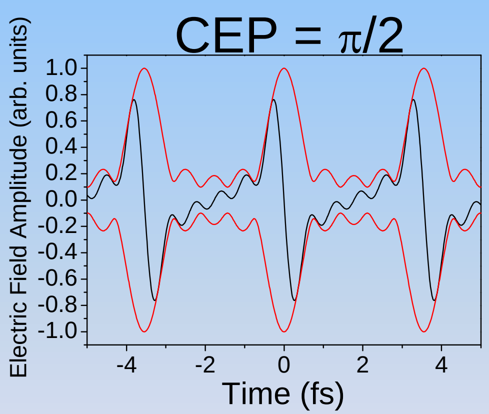
~25,000cm^{-1}

0.833 cycle per pulse
1.4 fs envelope
440 as cycle width
constant carrier envelope phase
2 ns pulse train duration
8.0 fs pulse spacing
~1 MW peak power

1064
532
355
266
213

~37,600cm^{-1}



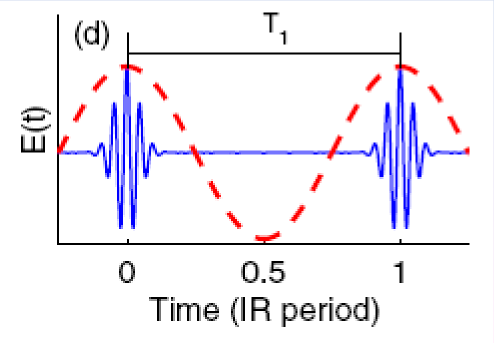


Attosecond pulse train

Raman Order	nm	cm^{-1}
1	2407	4155
2	1203	8310
3	802	12465
4	602	16620
5	481	20775
6	401	24930
7	344	29085
8	301	33240
9	267	37395
10	241	41550
11	219	45705
12	201	49860
13	185	54015

Nd:YAG Harmonics	nm	cm^{-1}
1	1064	9398
2	532	18796
3	355	28194
4	266	37592
5	213	46990

We can control the CEP and shape the pulse waveform.
We can synthesize a waveform which like the XUV-IR combination.



Advance Concepts

Technology

- Generate subfemtosecond pulses: add more sidebands and improve sideband power
- Increase pulse-to-pulse spacing
- Develop control of carrier envelope phase
- Modulate in photonic crystal fiber
- Arbitrary waveform synthesis

Science

- Optical-deep uv attosecond pump-probe
- Tracing molecular vibrational wavepacket
- Low energy electron dynamics in atoms
- Electron dynamics in semiconductors: direct bandgap vs indirect bandgap

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