Ultrashort pulse train synthesized by light waveform control

Wei-Jan Chen 陳蔚然

AMO Seminar,
National Tsing Hua University
Sep. 27, 2010

Outline

- Introduction
- Review of basic concepts
- High Harmonic Generation
- Molecular Modulation
- Multicolor synthesized
- Outlook

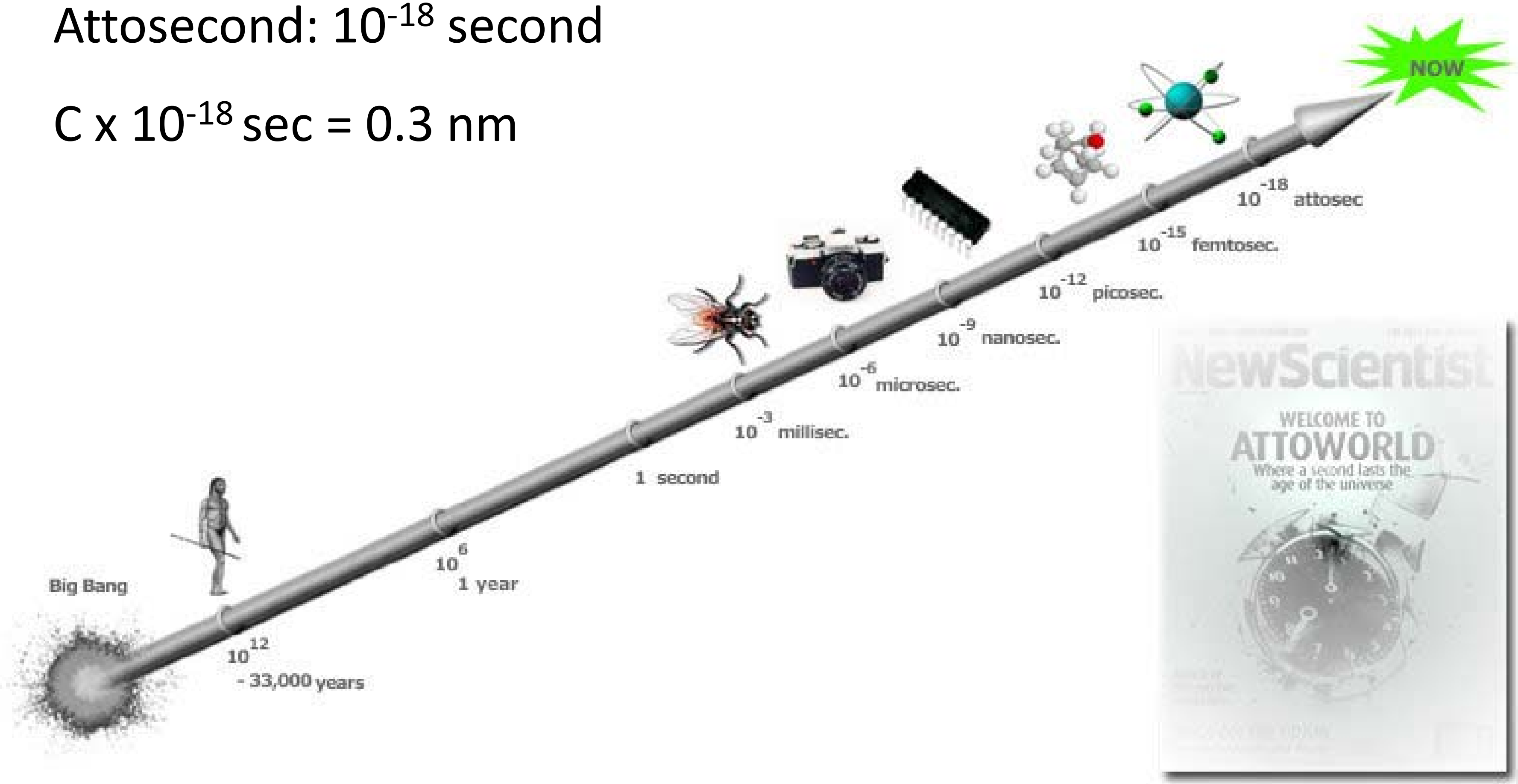
Outline

- Introduction
- Review of basic concepts
- High Harmonic Generation
- Molecular Modulation
- Multicolor synthesized
- Outlook

Attoworld

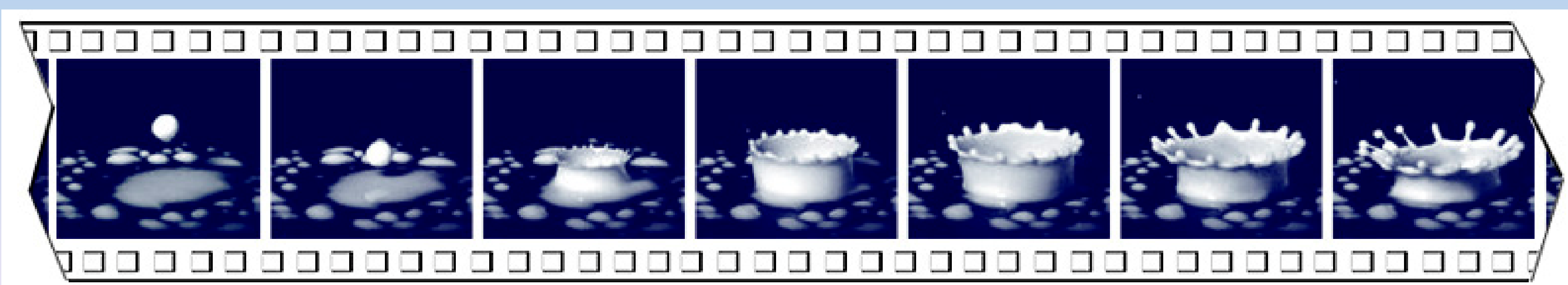
Attosecond: 10^{-18} second

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

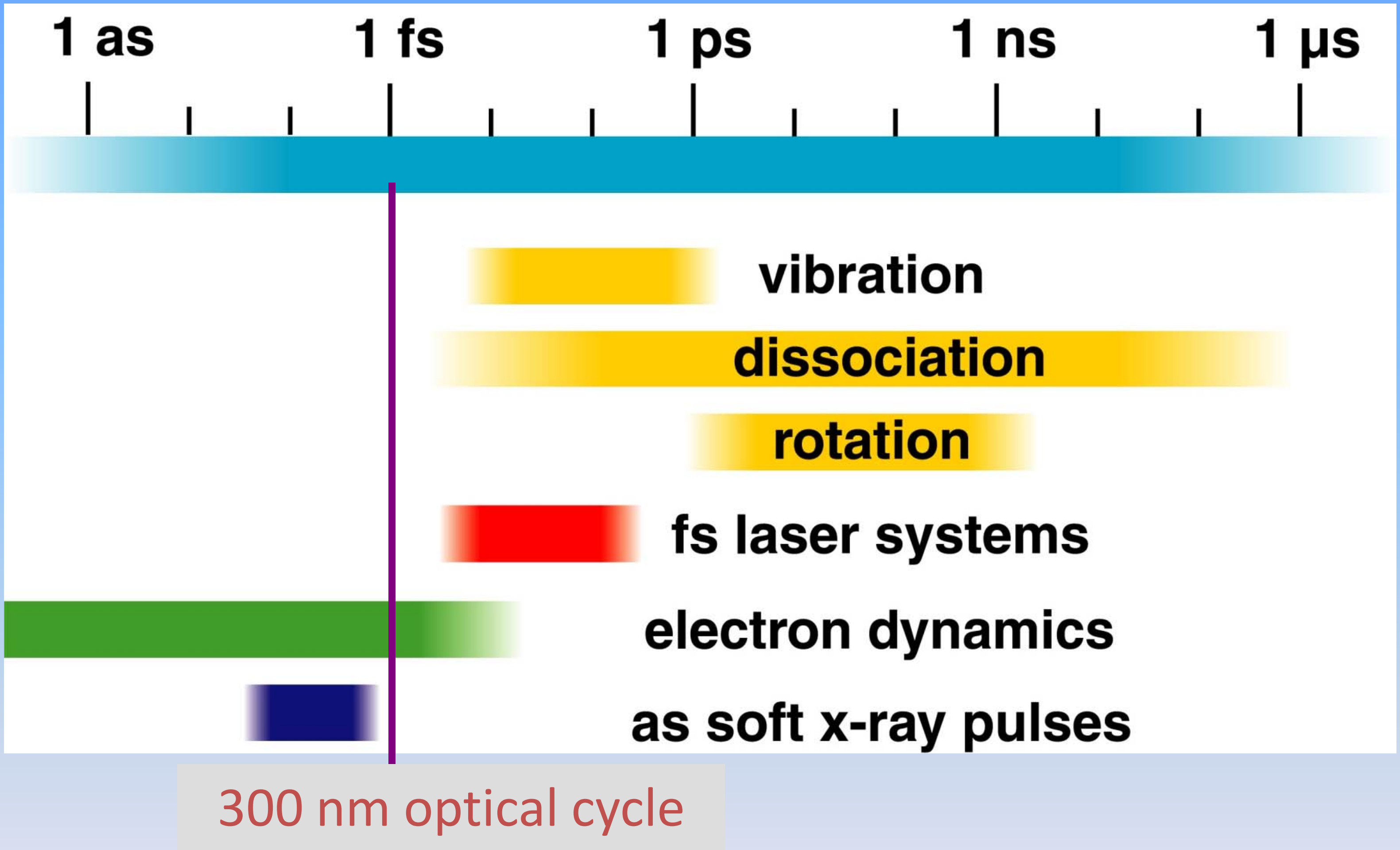


www.attoworld.de/attoworld.html



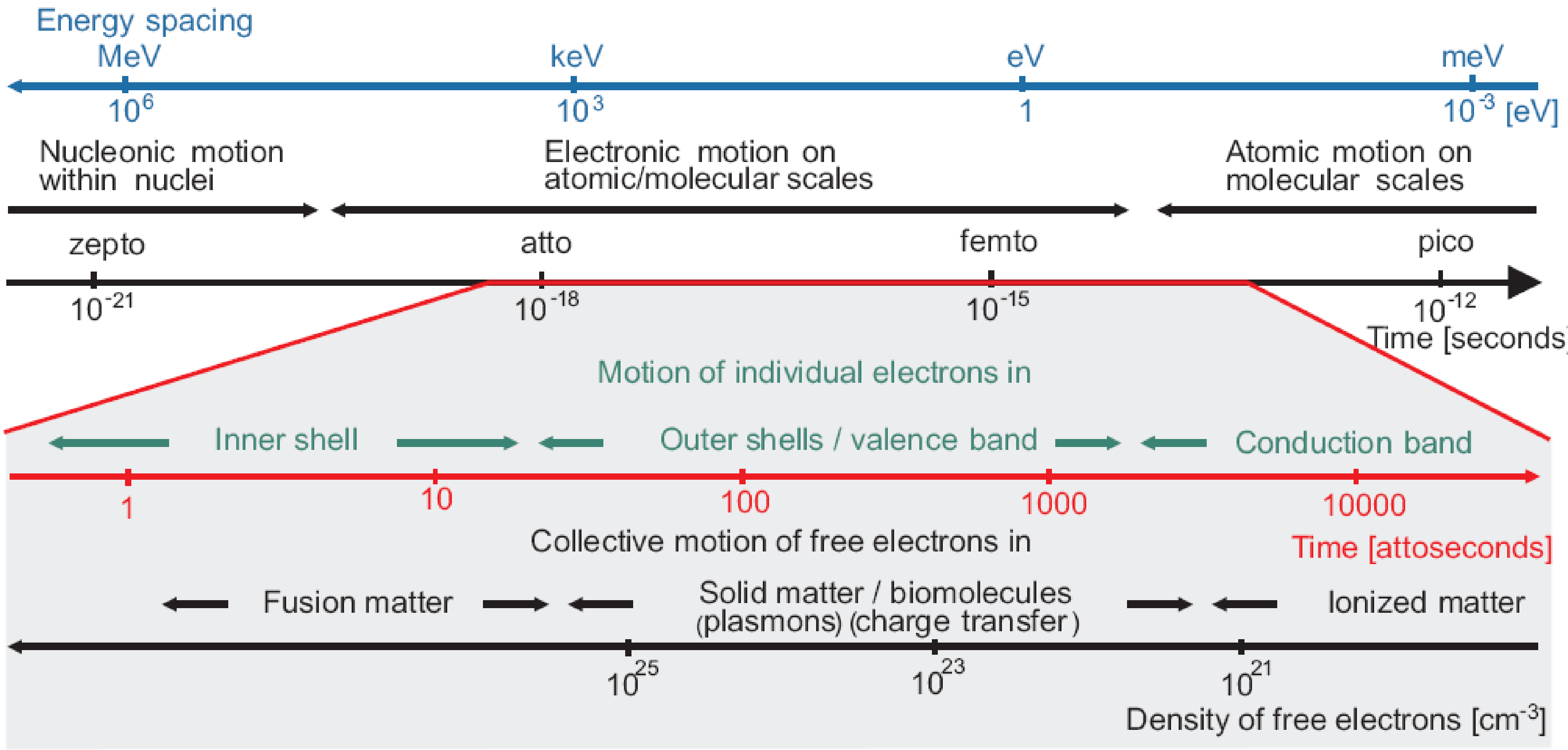


Request for Ultrashort Pulse



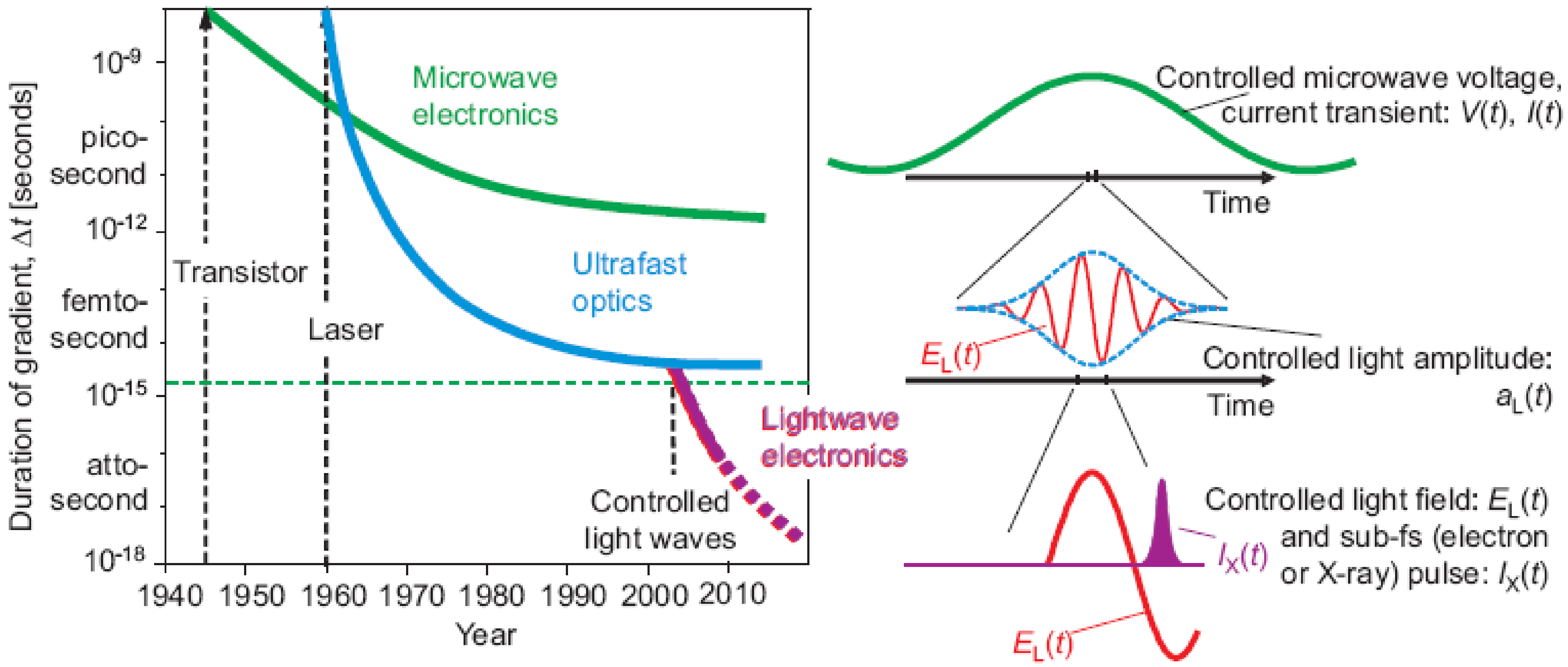
Short pulses can be used to monitor and control molecular and electronic motion

Electronic Motion



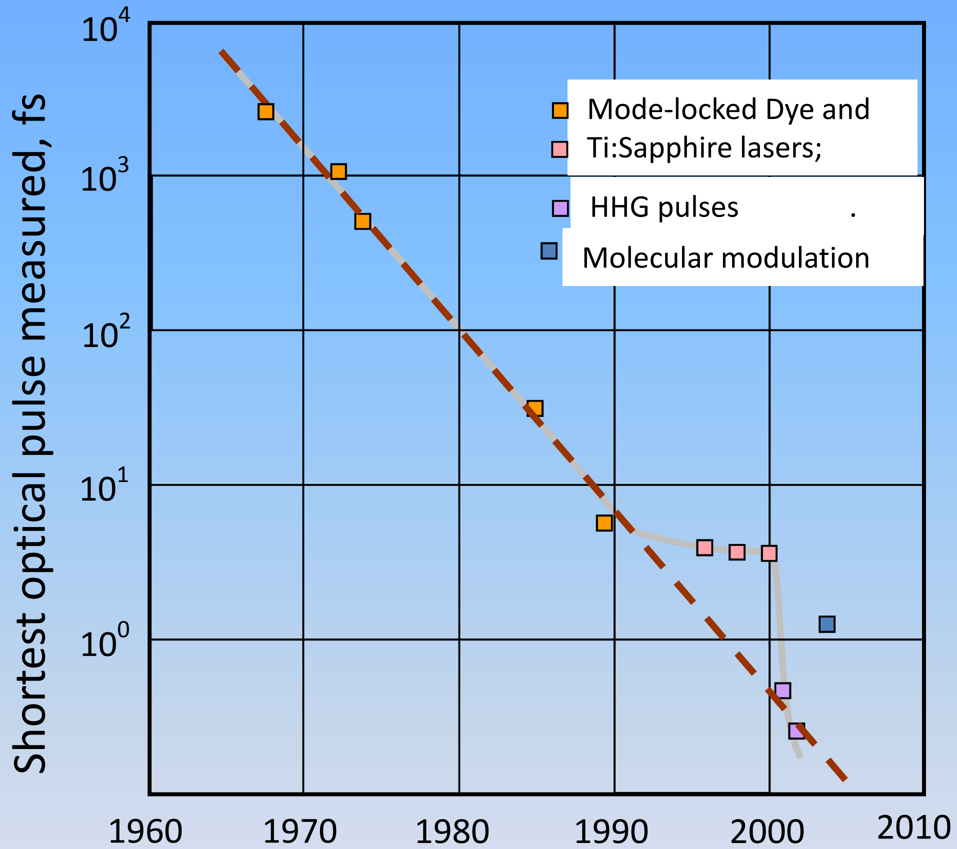
F. Krausz & M. Ivanov, Rev. Mod. Phys. 81 163 (2009)

Evolution of Ultrafast Science



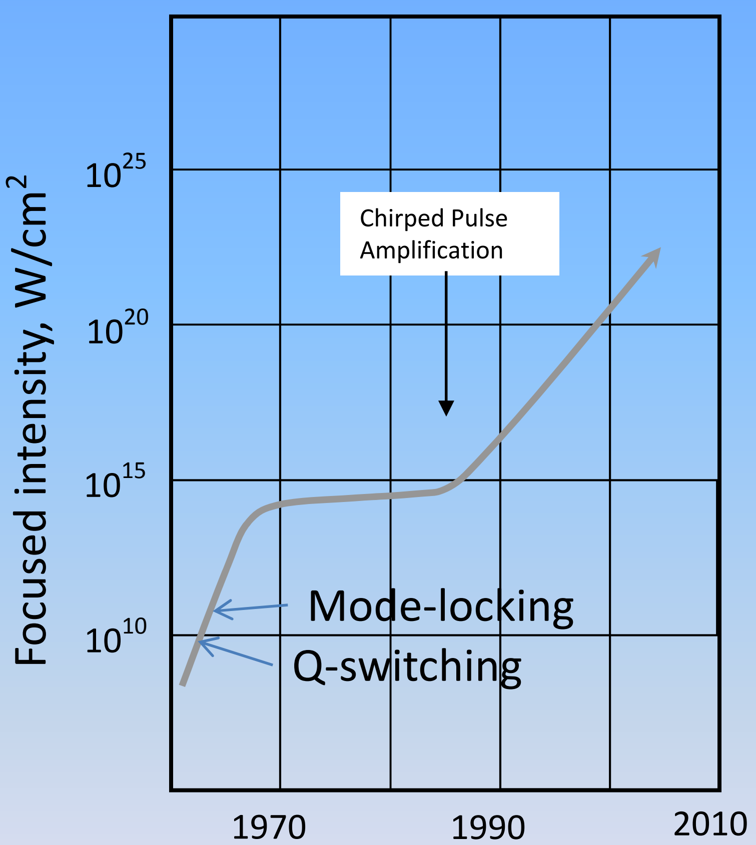
F. Krausz & M. Ivanov, Rev. Mod. Phys. 81 163 (2009)

Laser pulses got shorter over the years



Ultrafast science

Peak intensity increased



High field physics

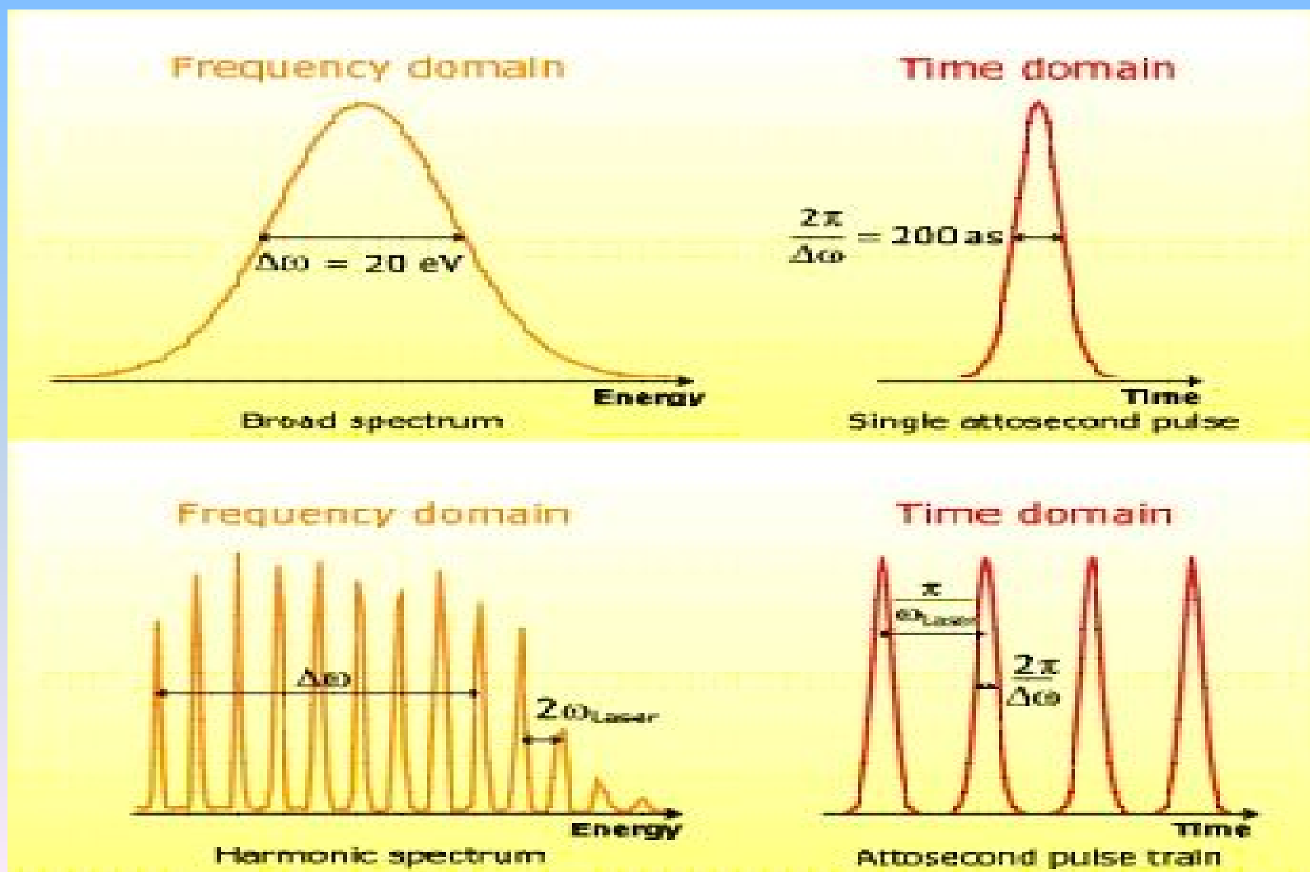
Outline

- Introduction
- Review of basic concepts
- High Harmonic Generation
- Molecular Modulation
- Multicolor synthesized
- Outlook

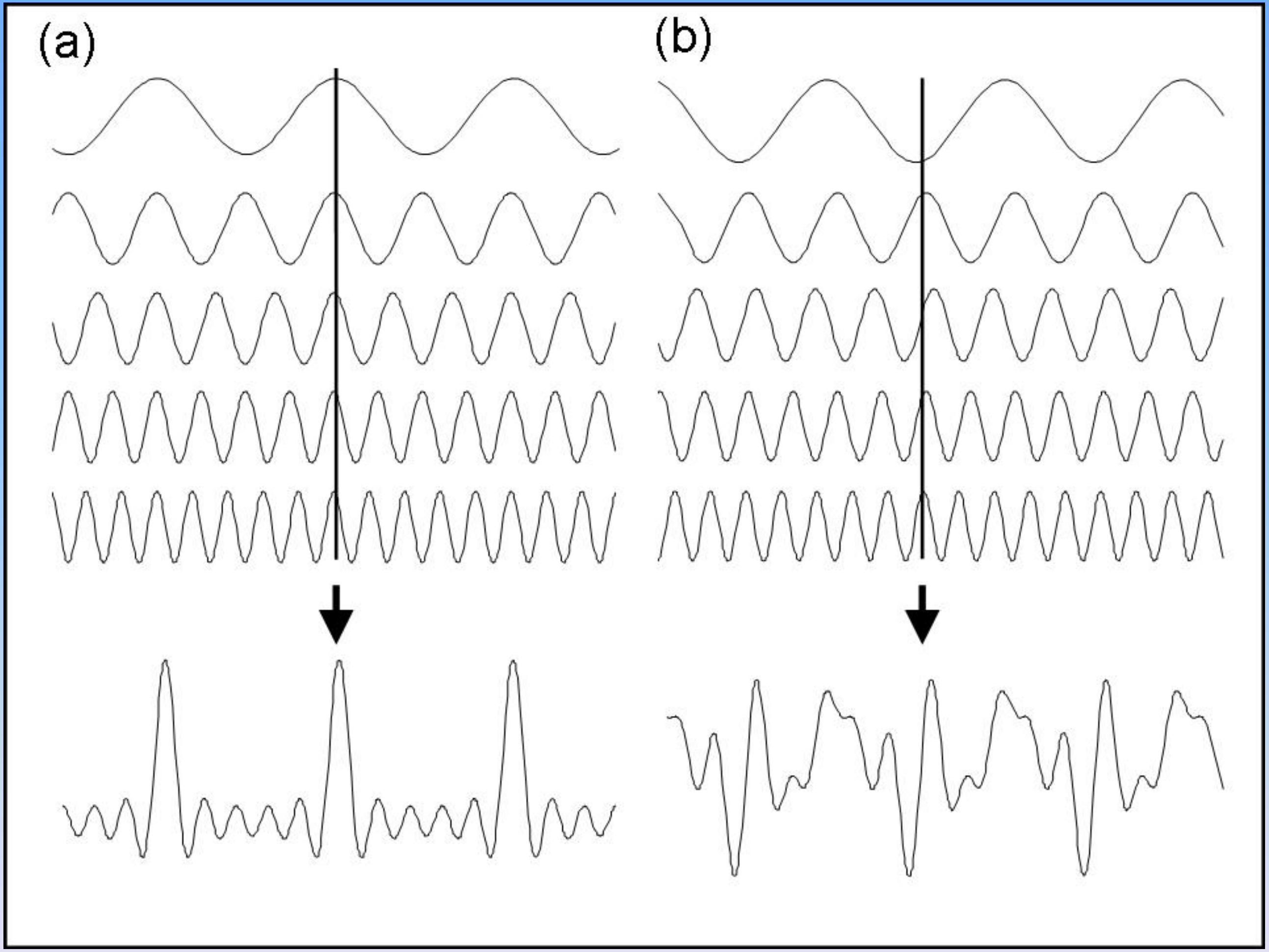
Correlation Between Time and Frequency

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

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



Principle of optical interference of coherent light fields

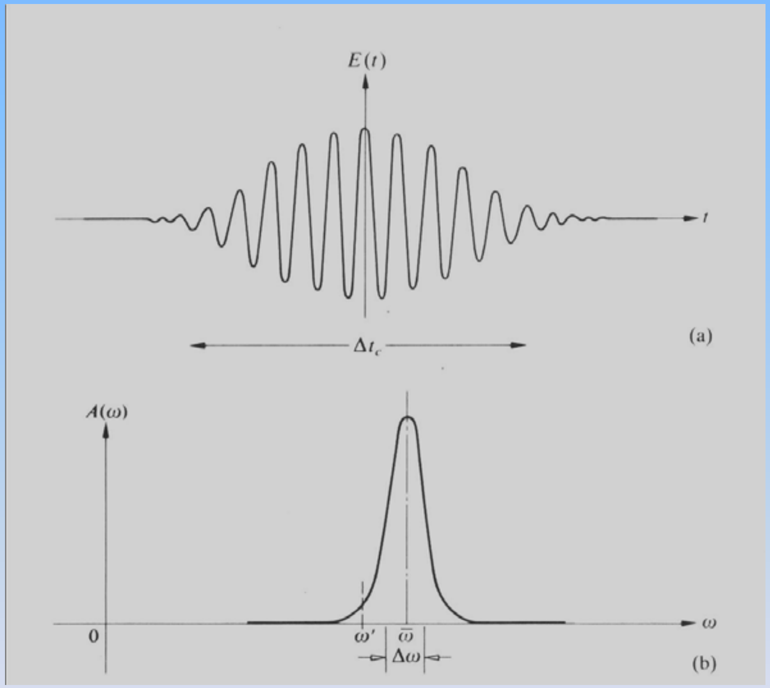


In phase

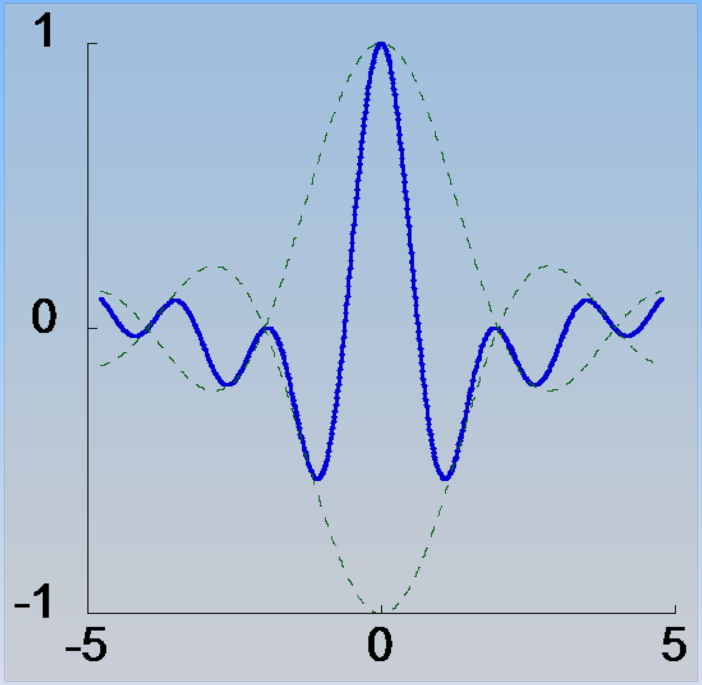
Random phase

What is a single cycle optical pulse

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



(a)



(b)

Optical cycle

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

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

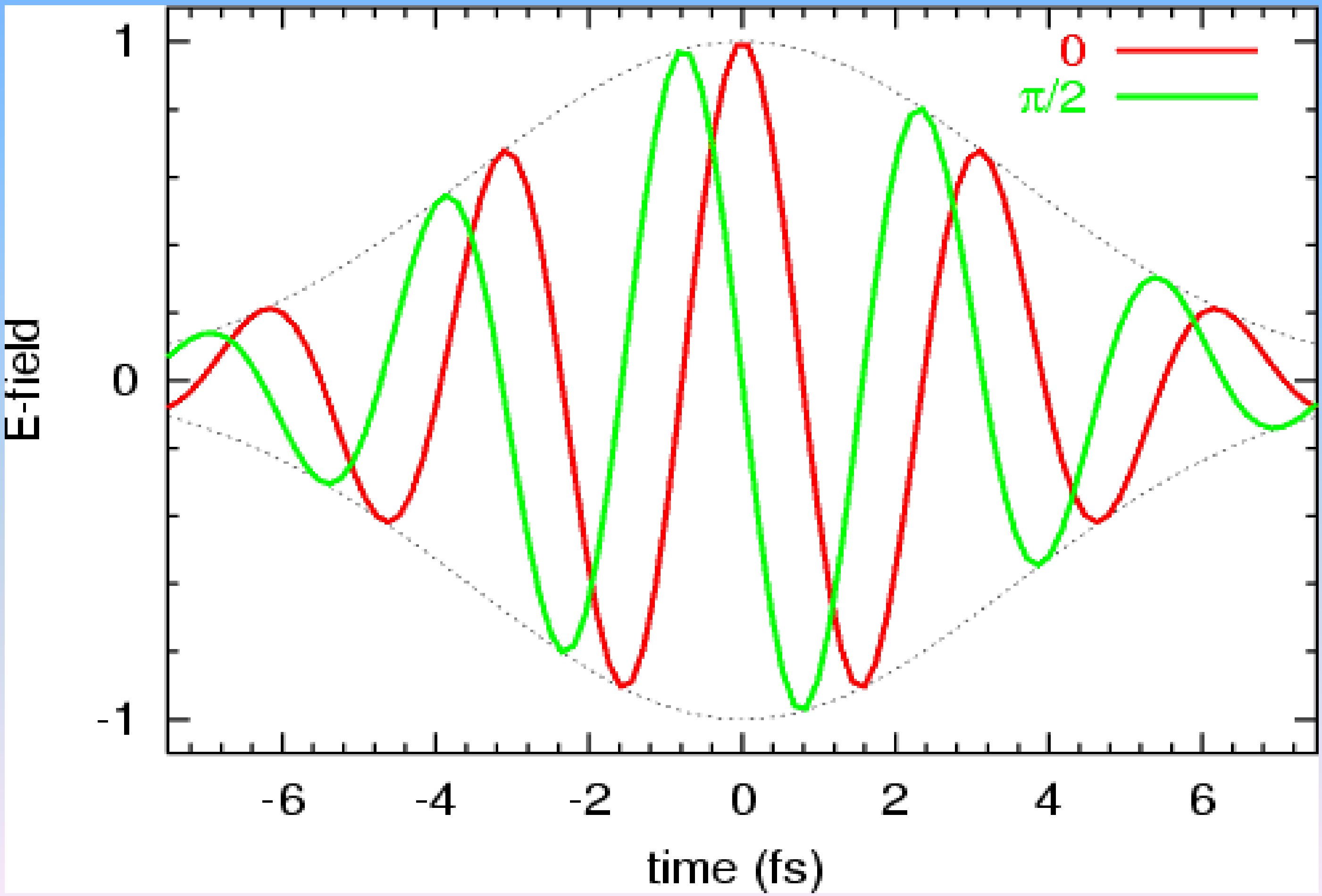
Carrier frequency

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

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

Carrier envelope phase

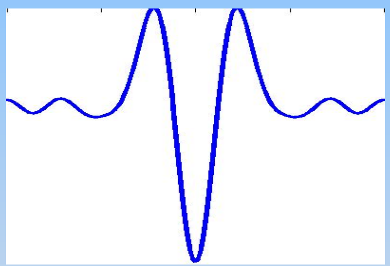
$$E(t) = E_0(t)\cos(\omega_0 t + \underline{\phi})$$



Single cycle waveforms

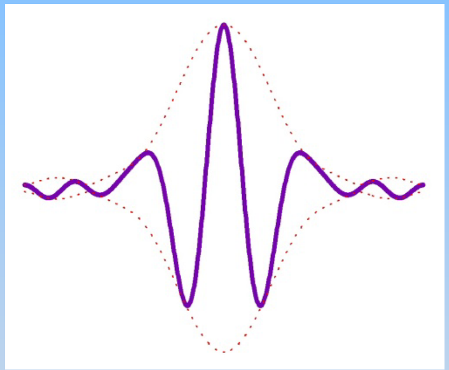
Inverted cosine

$\phi_n = \pi$



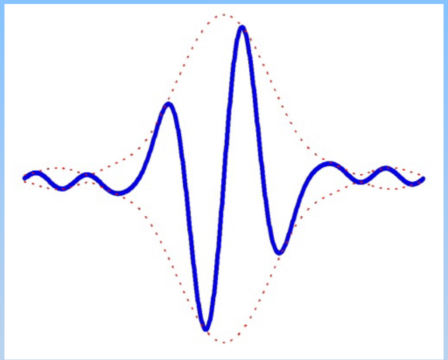
cosine pulse

$\phi_n = 0$



sine pulse

$\phi_n = \pi/2$



780 nm

200 nm

↔

12,820 cm^{-1}

50,000 cm^{-1}

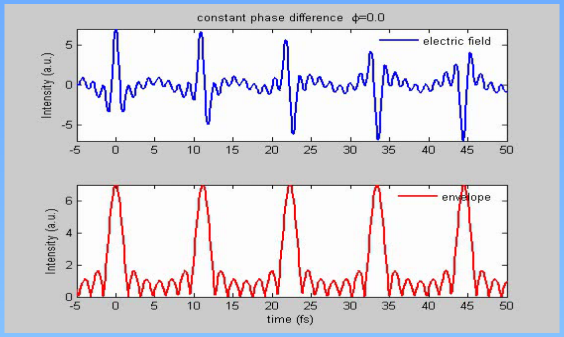
↔

2.6 fs

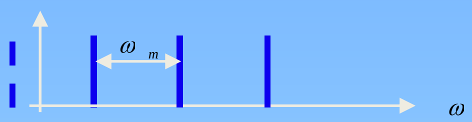
684 as

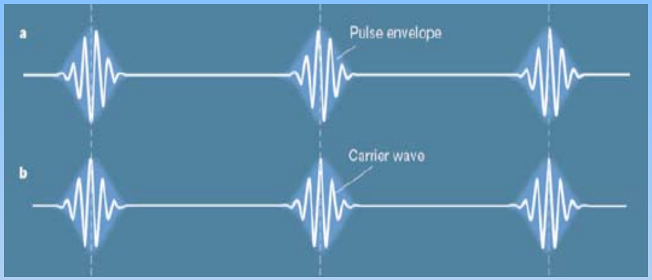
Constant carrier envelope phase

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



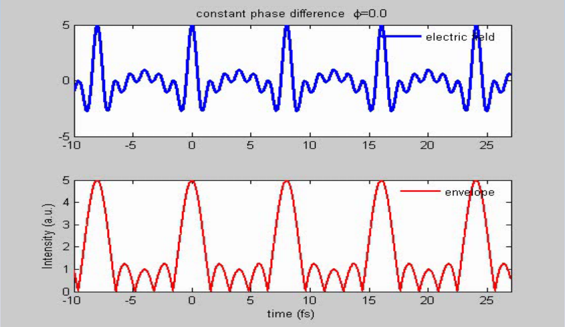
incommensurate $\omega_n = n\omega_m + \omega_{ceo}$ $\phi_n = \omega_{ceo} t + \phi_n^{'}$

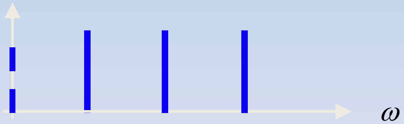




commensurate $\omega_q = n\omega_m$ $\phi_n = \phi_{CEP} + n\phi_m$

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





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

Light Waveform Control

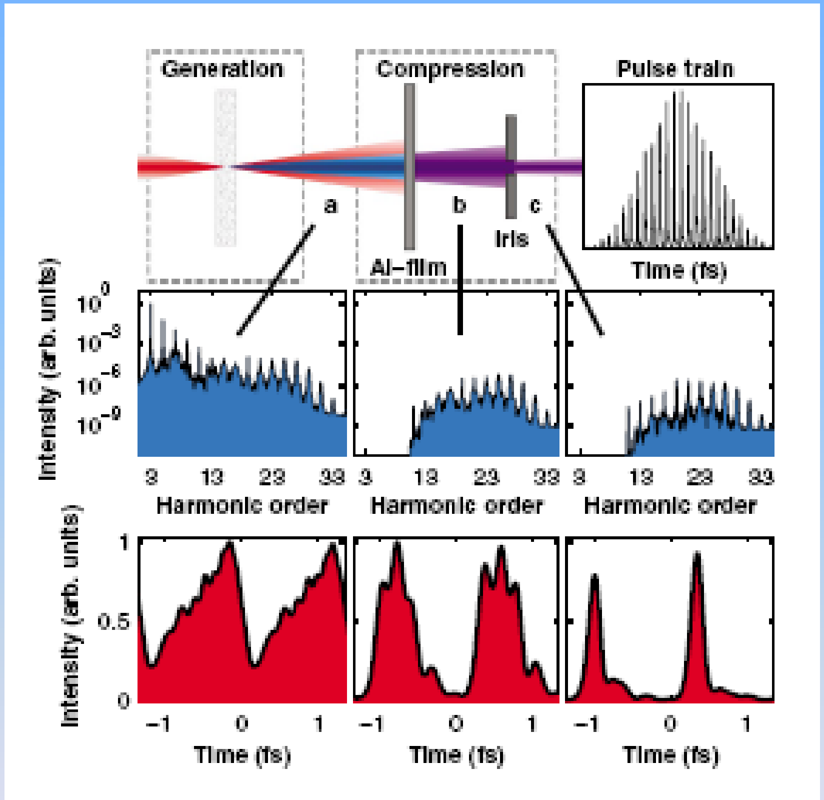
Outline

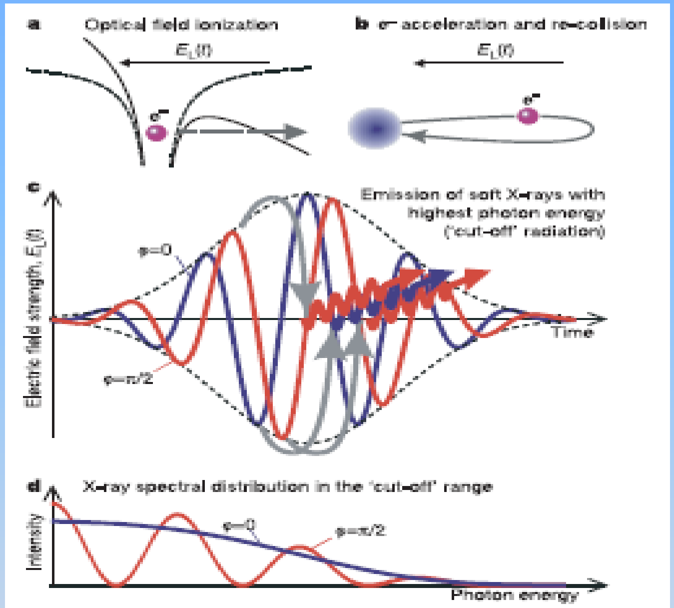
- Introduction
- Review of basic concepts
- High Harmonic Generation
- Molecular Modulation
- Multicolor synthesized
- Outlook

Methods of Generating Attosecond Pulses

A

High-order harmonic generation of phase-stabilized femtosecond pulse

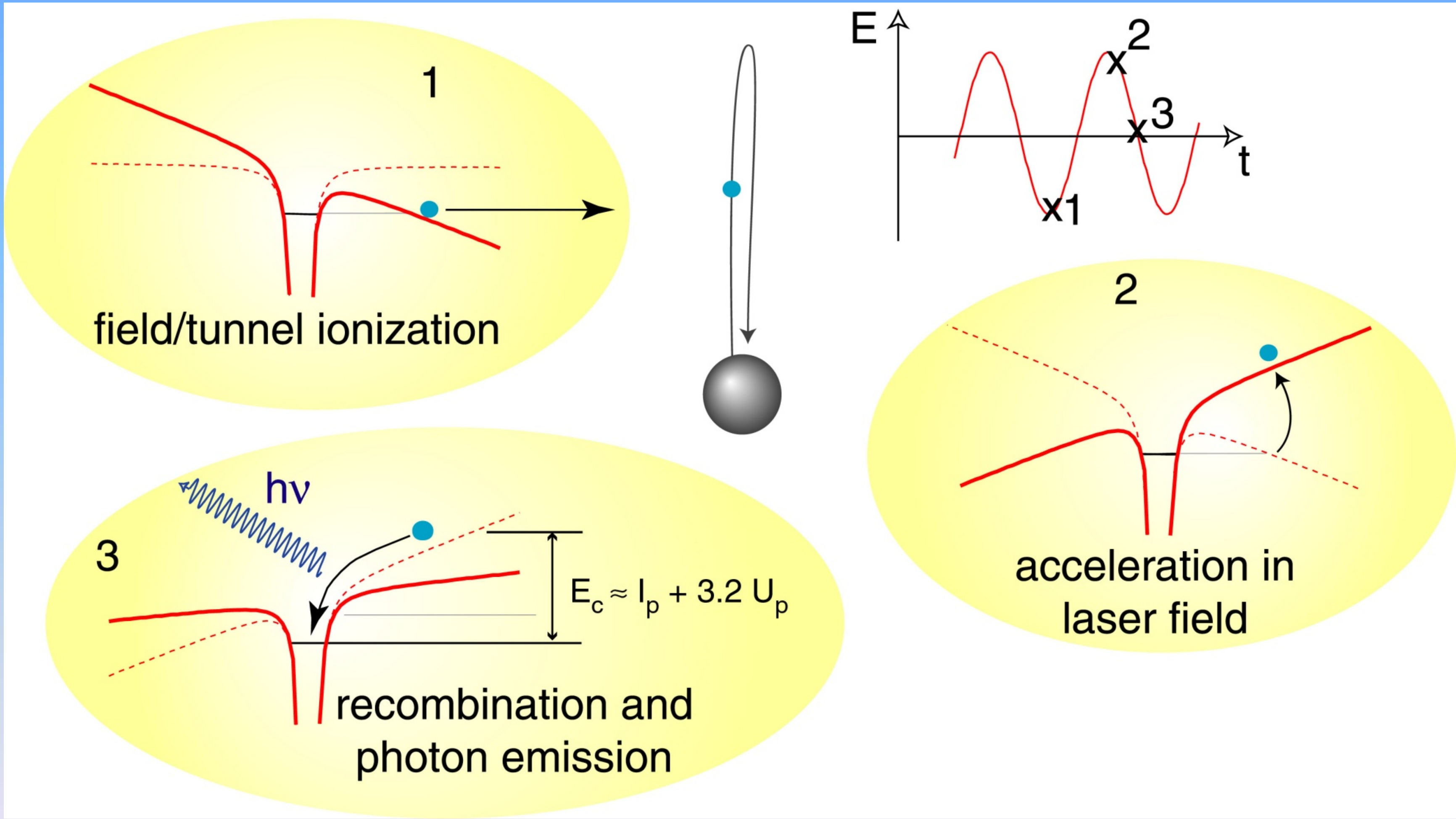




single pulse 100 attosecond
30-100 eV photons
very low power
constitutes a few cycles

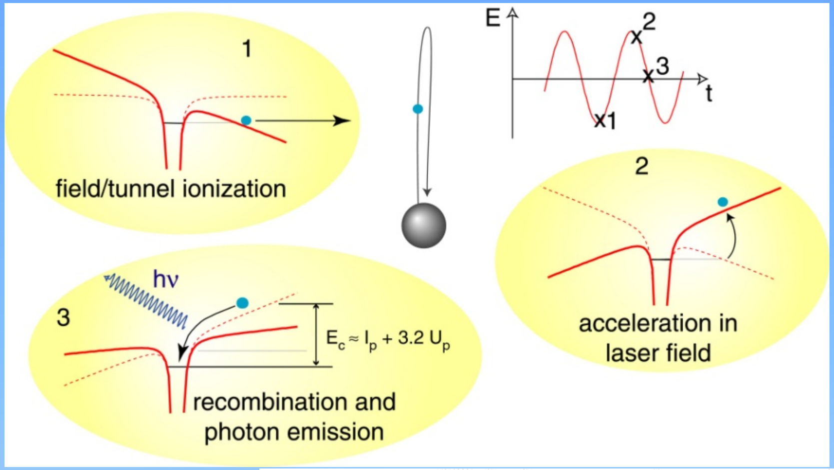
Krauze et.al., Nature 421, 611 (2003)
R. Lopez-Martens et. al., PRL 94, 033001 (2005)

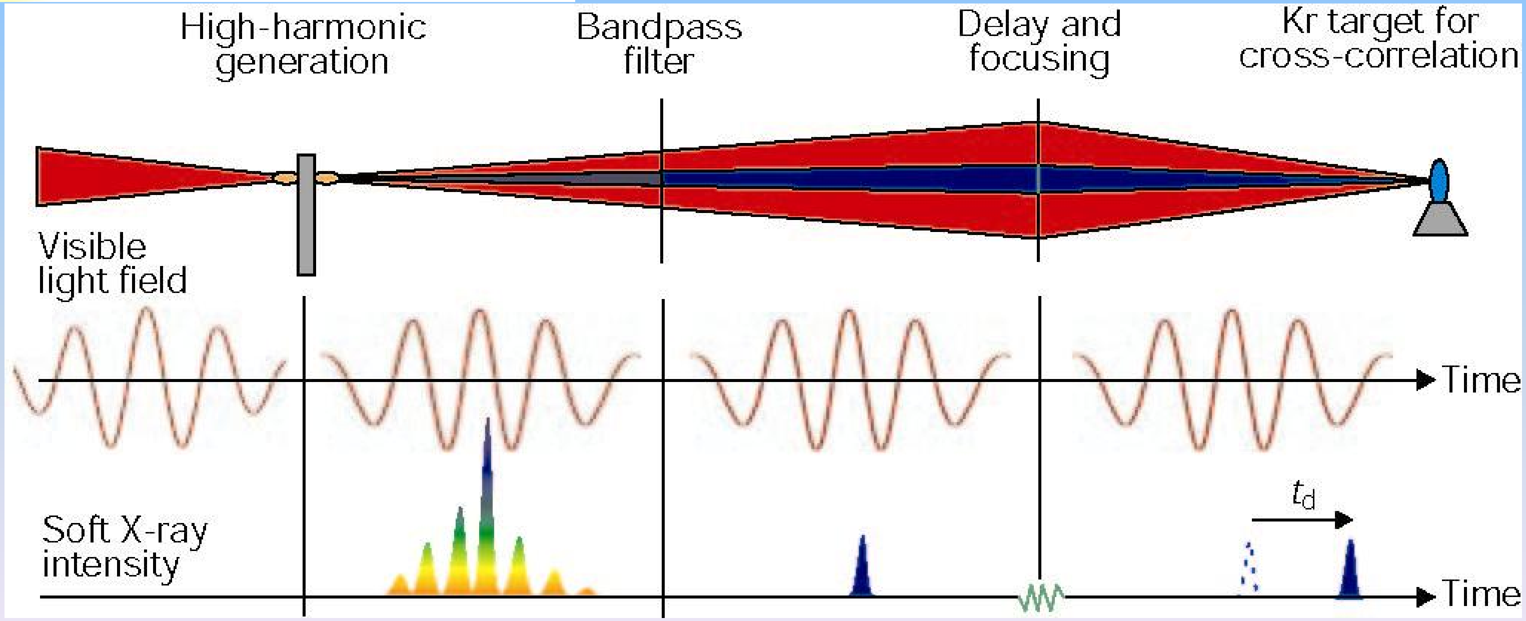
Three-step model



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

Isolated Attosecond-pulse production

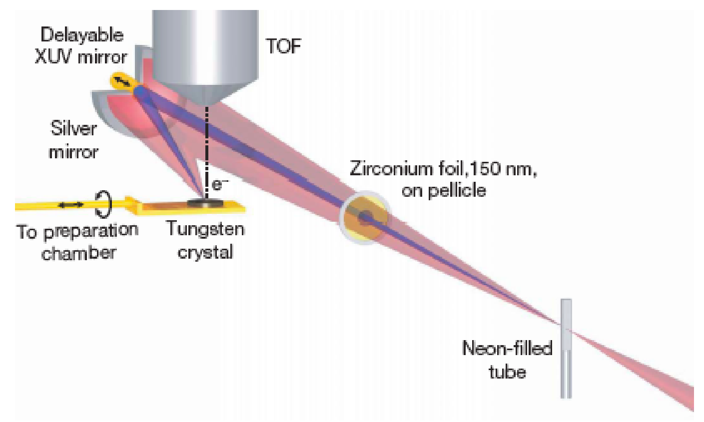


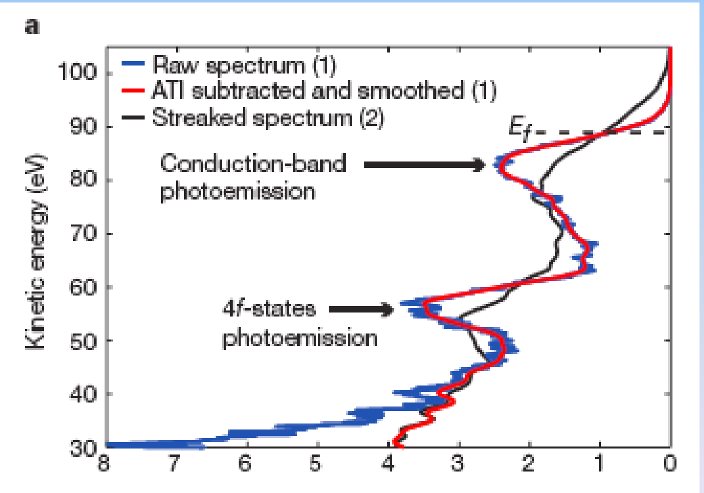


M. Hentschel et al, Nature **414**, 509 (2001)

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]





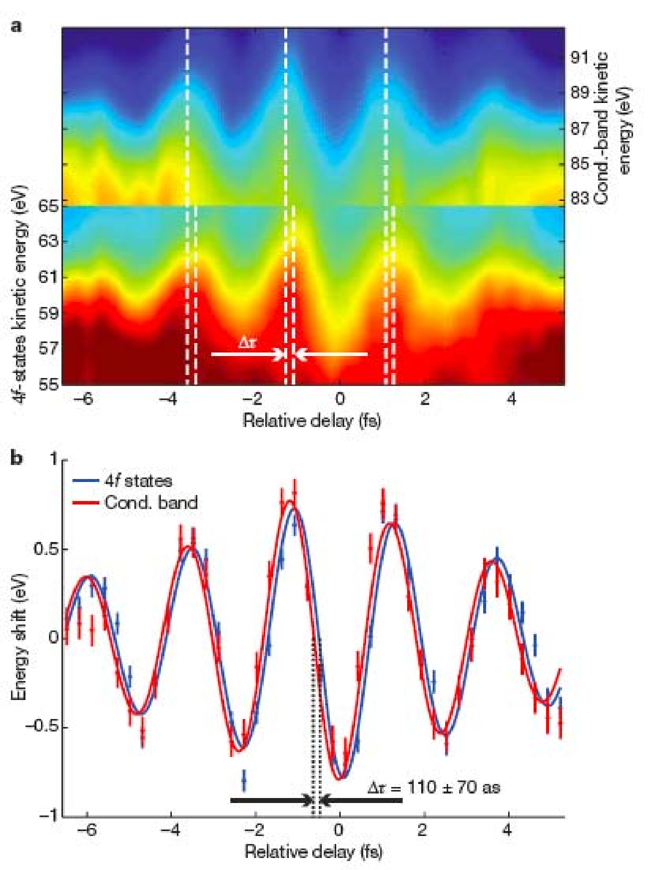


Figure 3 | Evidence of delayed photoemission. **a**, The 4*f* and conduction-band spectrograms, following cubic-spline interpolation of the measured data

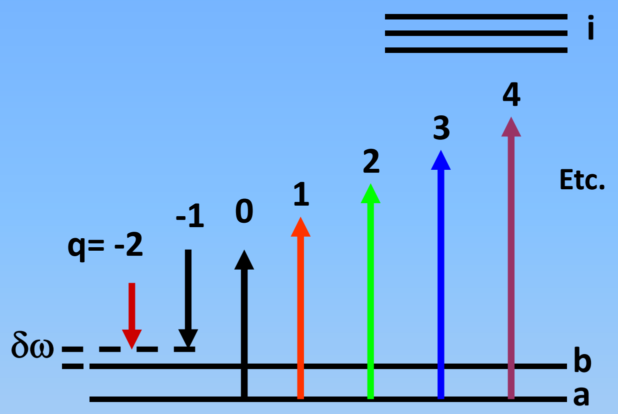
Nature 449, 1029 (2007)

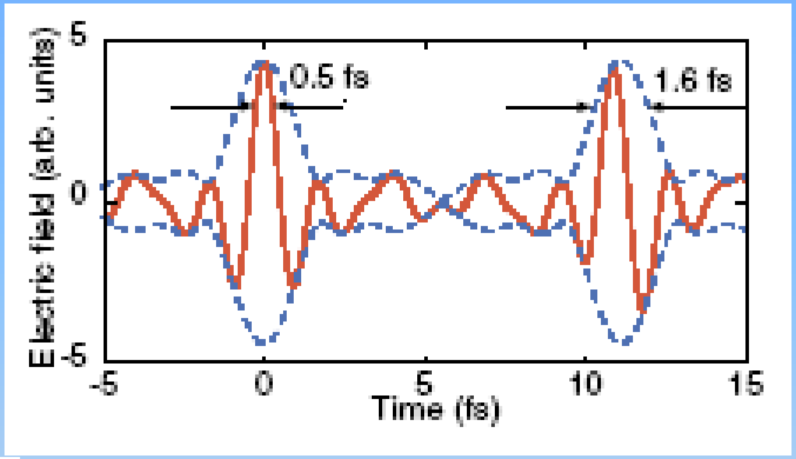
Outline

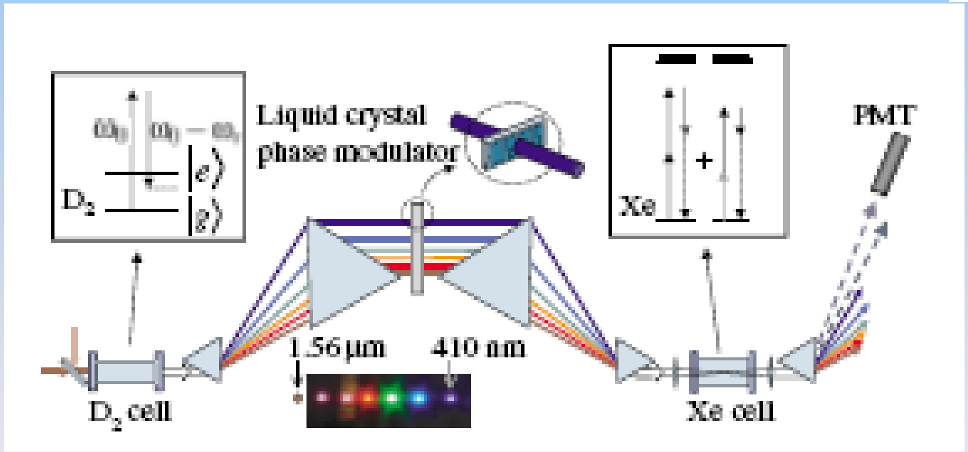
- Introduction
- Review of basic concepts
- High Harmonic Generation
- Molecular Modulation
- Multicolor synthesized
- Outlook

Methods of Generating Attosecond Pulses

B High-order stimulated Raman scattering using molecular modulation





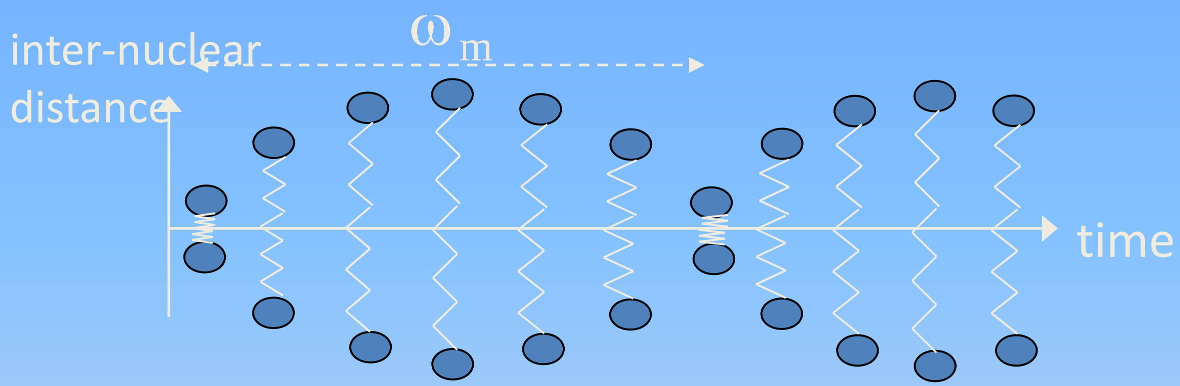


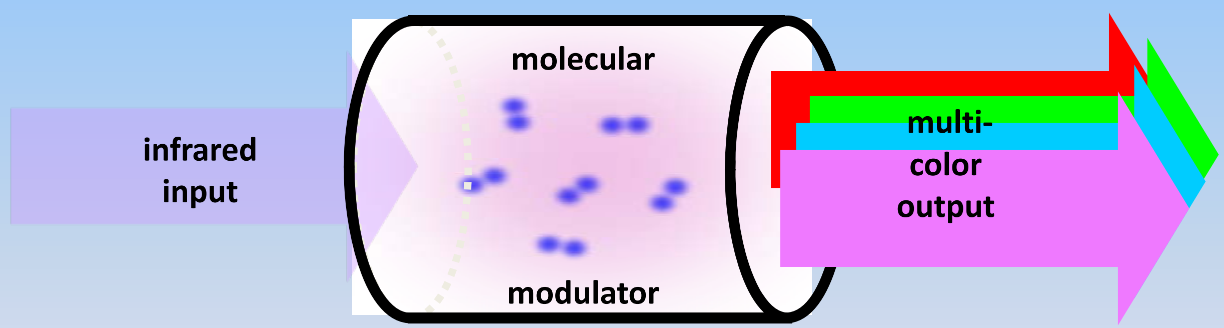
IR-UV region
good power
single-cycle
8-50 fs pulse spacing
limited to ~ 300-500 as

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

Molecular Modulation

Molecular modulation is analogous to electro-optic modulation





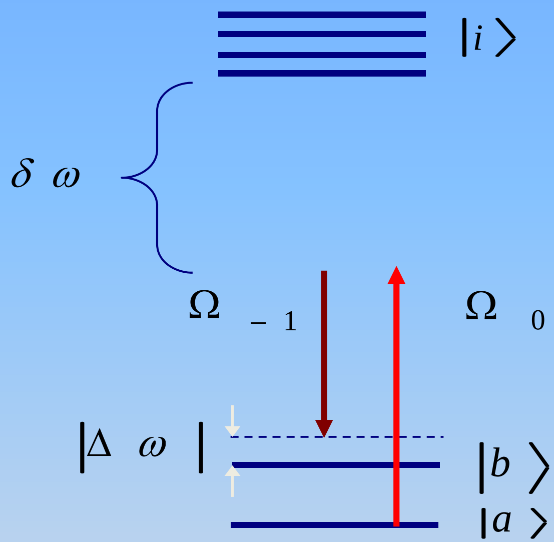
Refractive Index $$n = n_0 + \delta \cos \omega_m t$$

$\omega_q = \omega_0 + q\omega_m$ 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, $\rho_{ab} = 0.5$

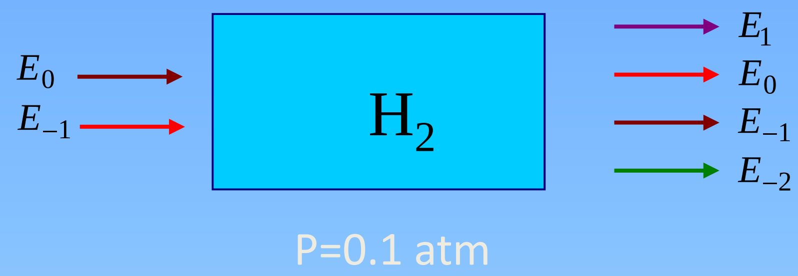
Coherence:

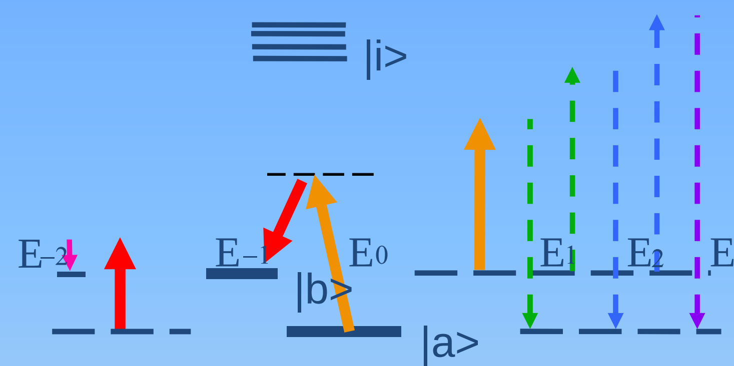
$$\rho_{ab}^{(\pm)} = \frac{1}{2}e^{i\varphi}\sin 2\theta^{(\pm)} = \pm\frac{\Omega_{ab}}{\sqrt{\left(\Omega_{aa} - \Omega_{bb} + \delta\right)^2 + 4\left|\Omega_{ab}\right|^2}}$$

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

Sideband Generation and Propagation

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





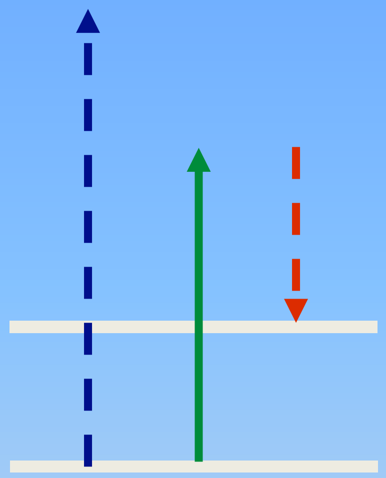
- Propagation equation for the *q*th sideband: $\omega_q = \omega_{q-1} + \omega_0 - \omega_{-1}$

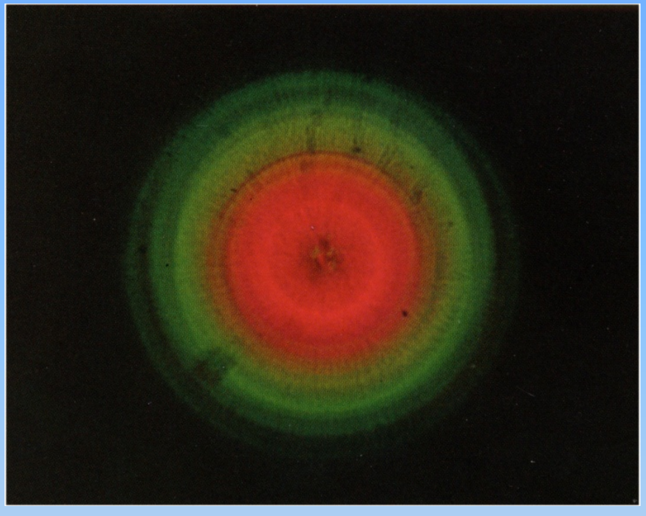
$$\frac{\partial E_q}{\partial z} = -j\eta\hbar\omega_q N\left(\underbrace{a_q\rho_{aa}E_q + d_q\rho_{bb}E_q}_{\text{dispersion}} + \underbrace{b_q^*\rho_{ab}E_{q-1} + c_q^*\rho_{ab}^*E_{q+1}}_{\text{coupling}}\right)$$

At maximum coherence, ρ_{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



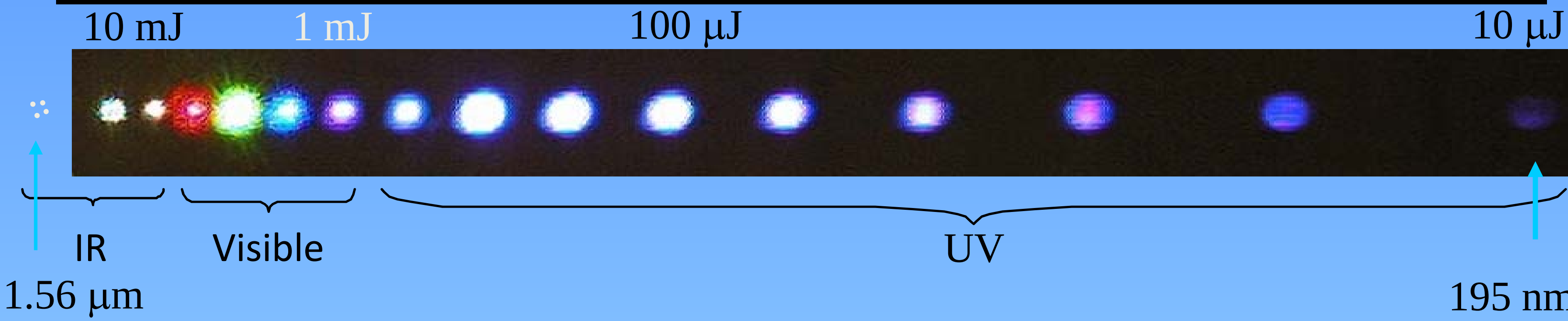


Traditional SRS:

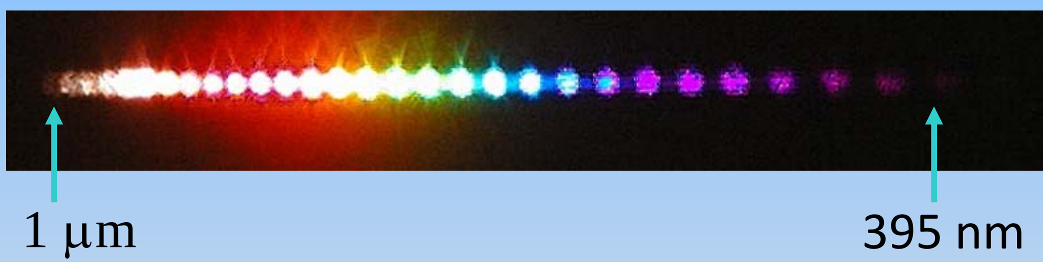
- ★ Generation occurs at high gas pressure
- ★ Molecular excitation occurs on-resonance
- ★ Anti-Stokes generation occurs off-axis
- ★ Few Stokes and anti-Stokes orders are observed.

Courtesy M. Shverdin

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

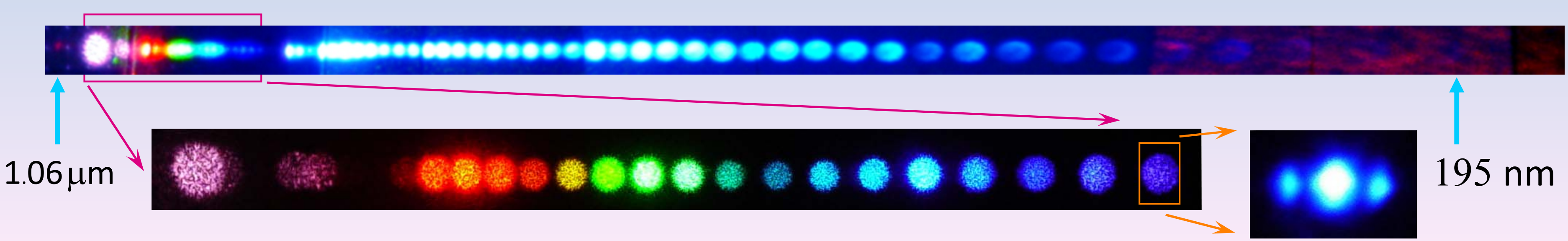


H_2 Rotation Spectra: 29 sidebands, spaced by 587 cm^{-1}

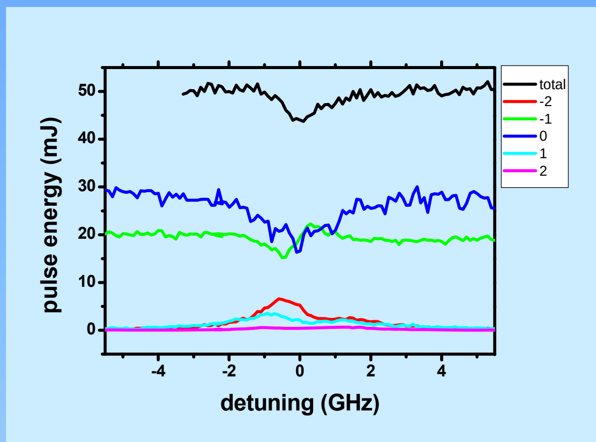


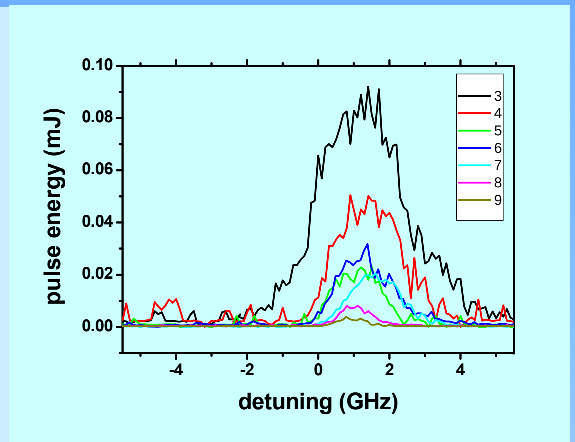
Phys. Rev. A (R)(1997)
Phys. Rev. Lett. 81 (1998)
Opt. Lett. 24 (1999)
Phys. Rev. Lett. 84 (2000)
Phys. Rev. Lett. 85 (2000)
Phys. Rev. A 63 (2001)
Phys. Rev. Lett. 91 (2003)
Phys. Rev. Lett. 93 (2005)

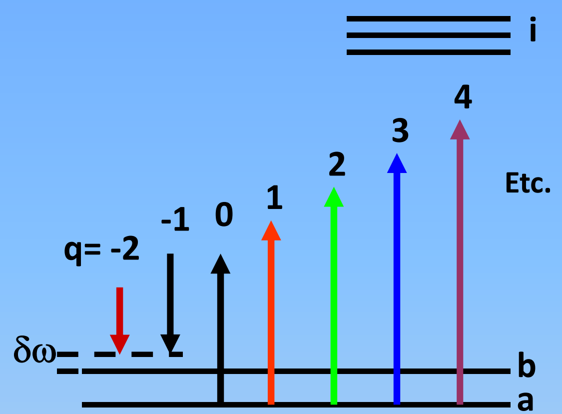
Multiplicative Spectra: ~ 200 sidebands, spaced by < 587 cm^{-1}

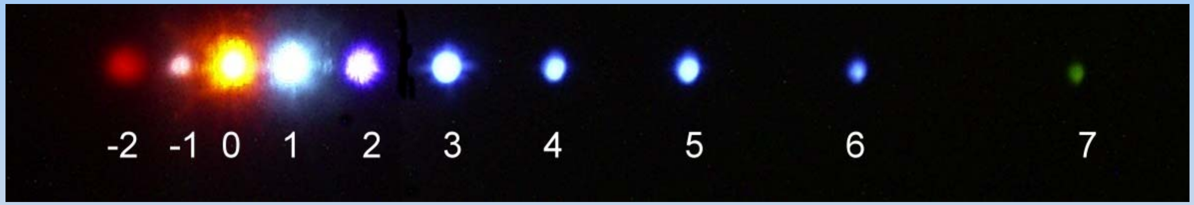


Raman Spectrum





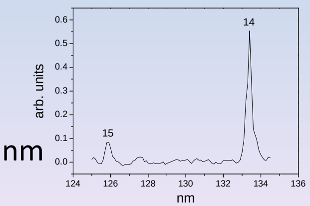




q = 3, λ =	339.6	nm
6	238.6	nm
9	183.9	nm
12	149.6	nm
14	133.0	nm

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

15th order at 126 nm observed



$$\omega_q = n\omega_m$$

Note:

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

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

The sidebands are not commensurate.

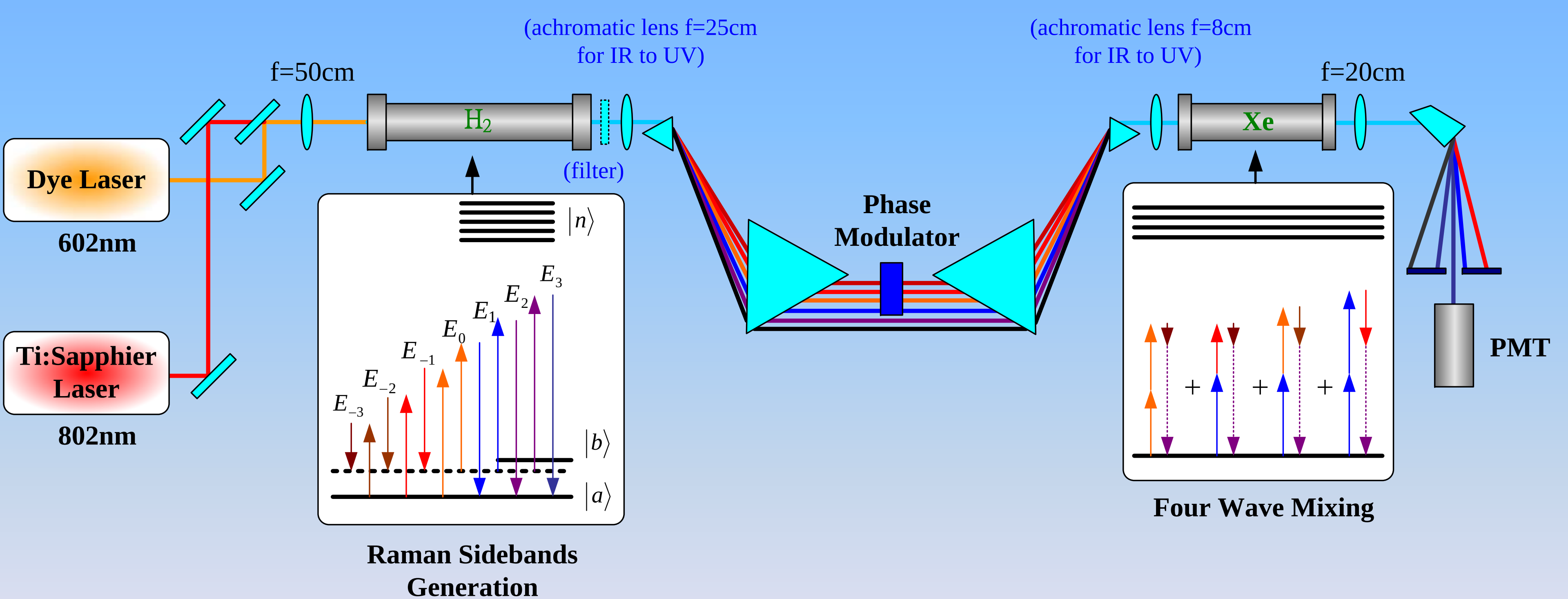
New input wavelengths:

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	

Experiment Setup



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

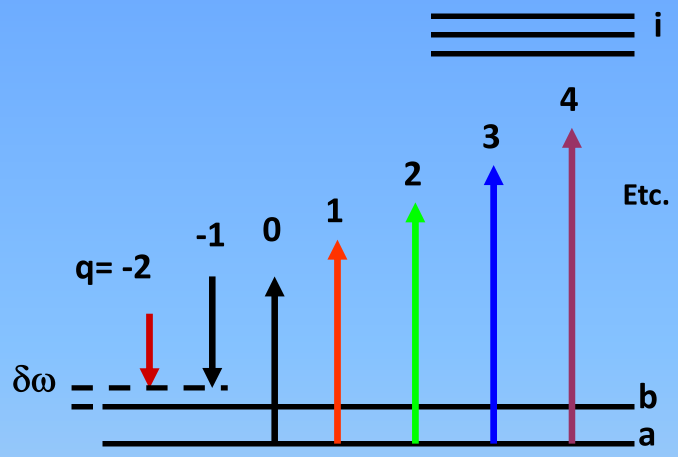
Multiple quantum paths interference

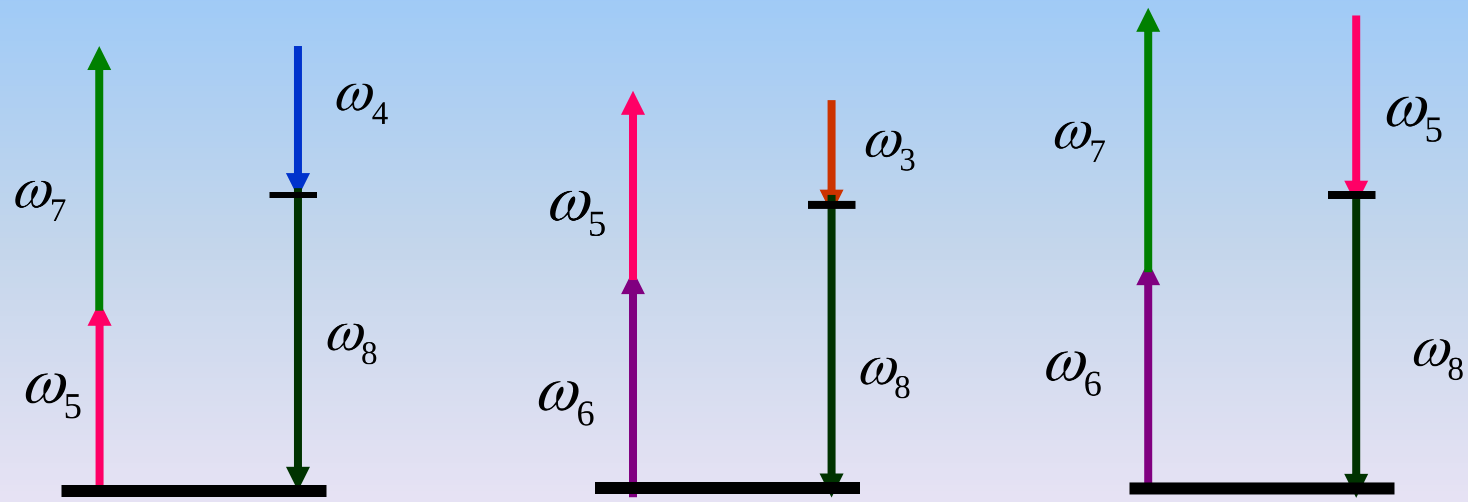
Four wave mixing:

$$\omega_5 + \omega_7 - \omega_4 = \omega_8$$

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

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





In phase condition

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

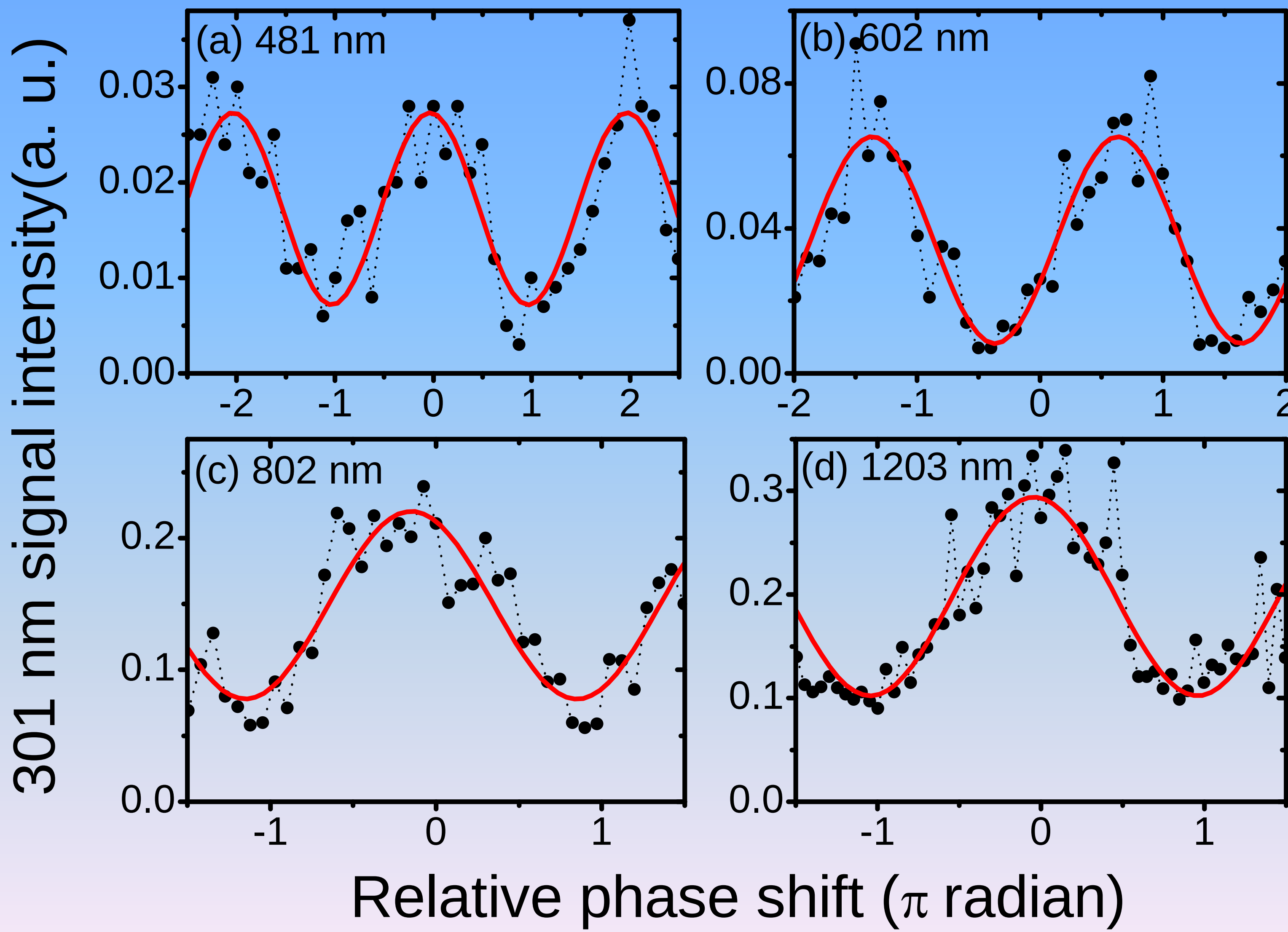
5+5-3, 5+4-2, 5+3-1.

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	$\Phi_{65}=\Phi_{54}=\Phi_{43}=\Phi_{32}=\Phi_{21}$

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

Searching in phase condition



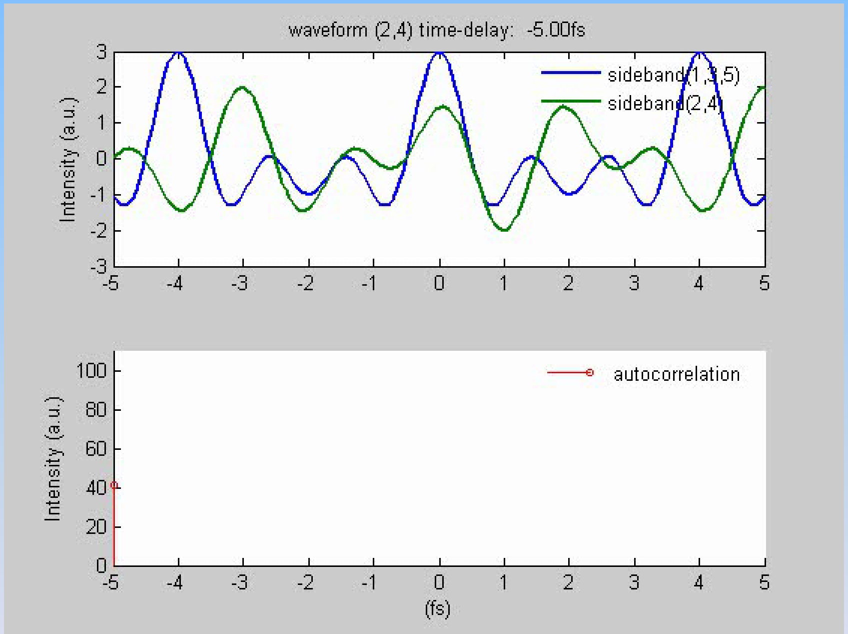
How to check the pulse shape?

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

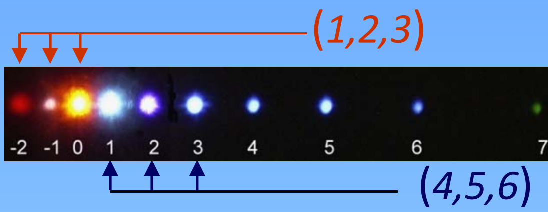
Solution: Correlation using pulses formed by the sidebands themselves.

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.

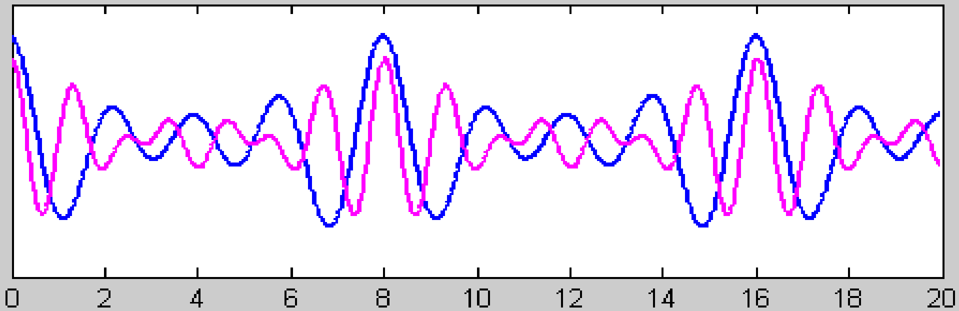
simulation



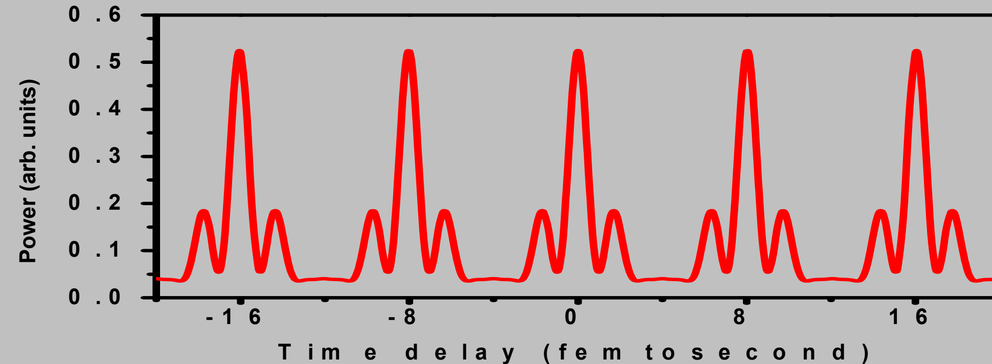
Cross Correlation of Single Cycle Pulse Train



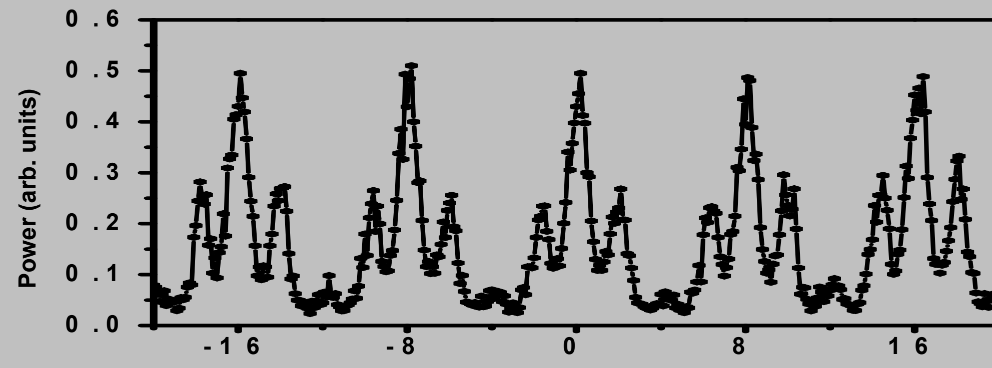
Sideband Orders



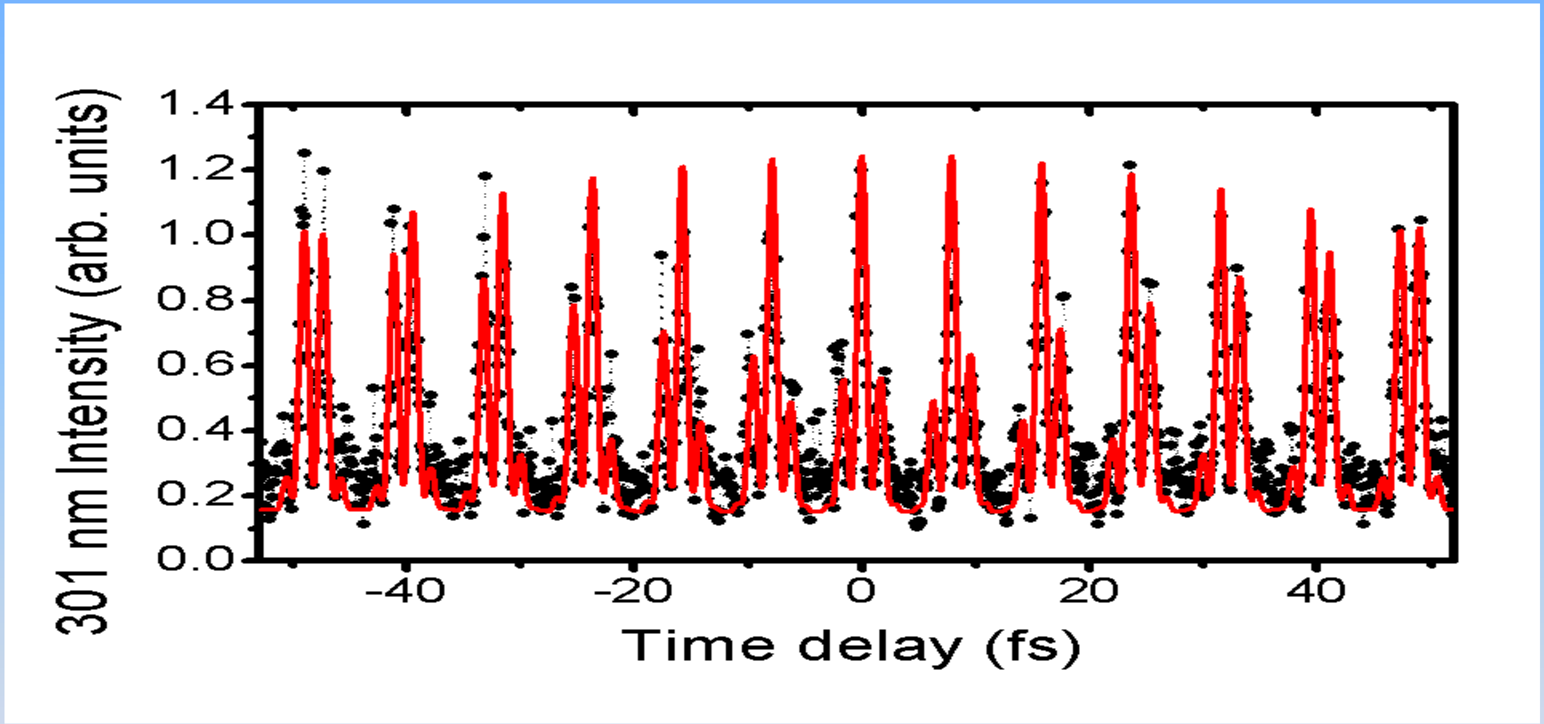
Simulation →



Experiment →

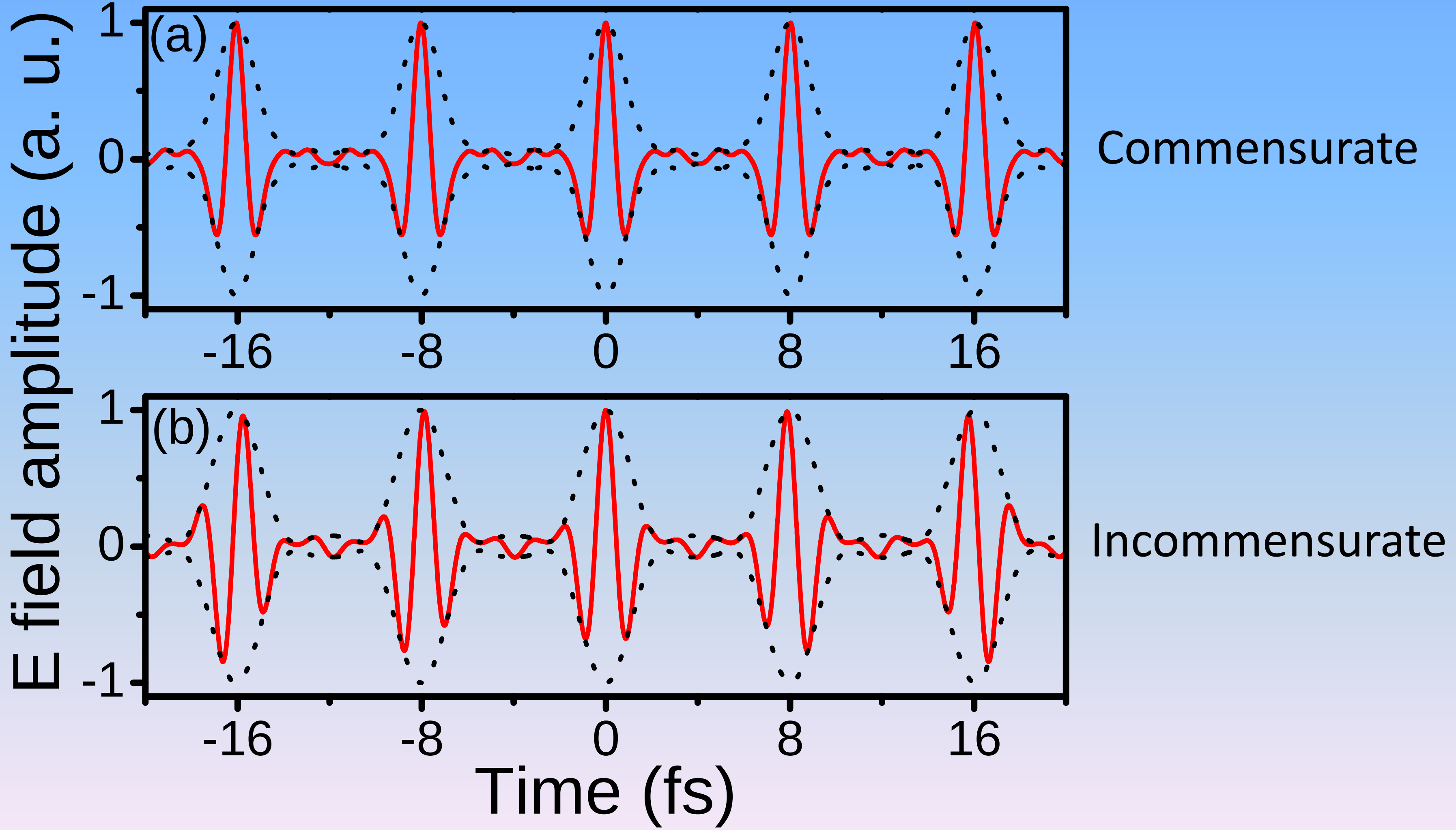


Cross correlation signal of incommensurate pulses



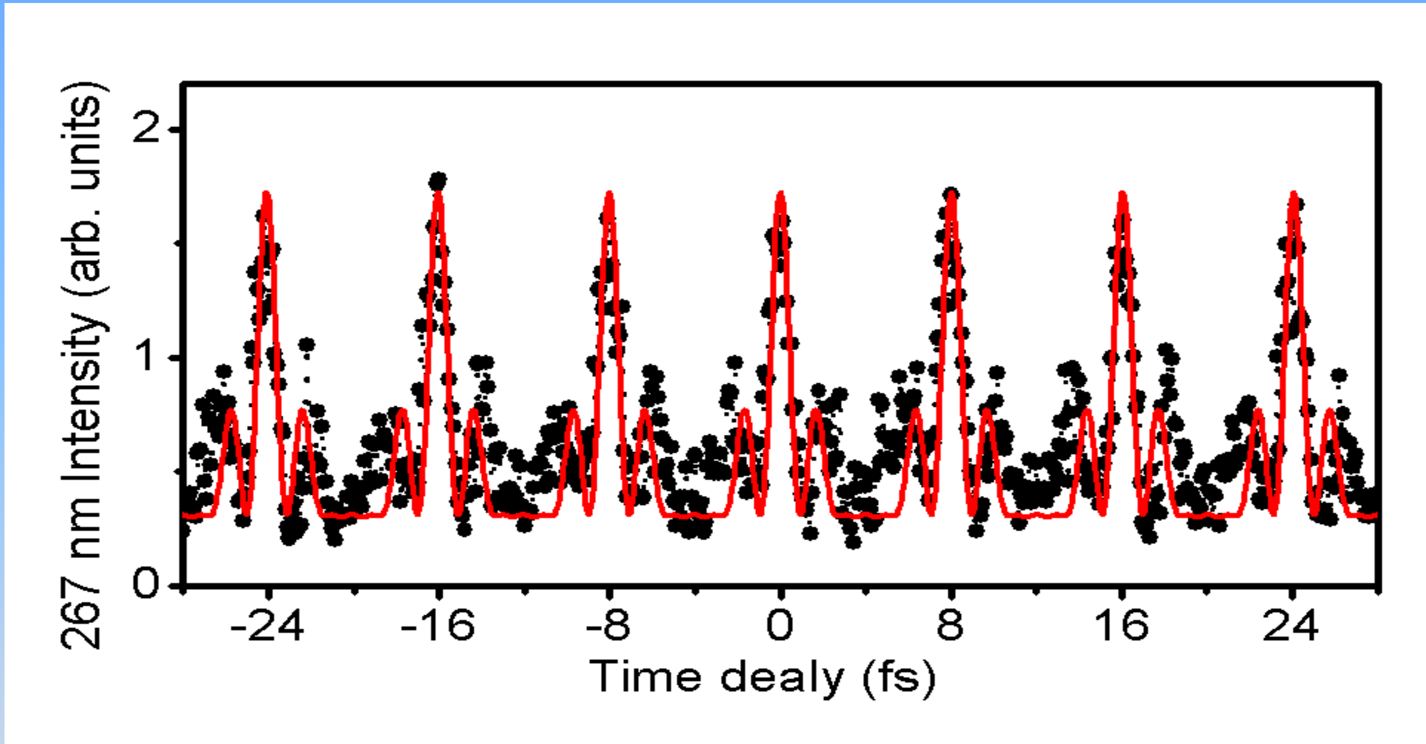
CEO frequency ~ 349 cm-1
Waveform repeats every 96 fs

Pulse train



E field amplitude (a. u.)
1
0
-1
(a)
-16
-8
0
8
16
(b)
Time (fs)
Commensurate
Incommensurate

7 beam correlation in Xe



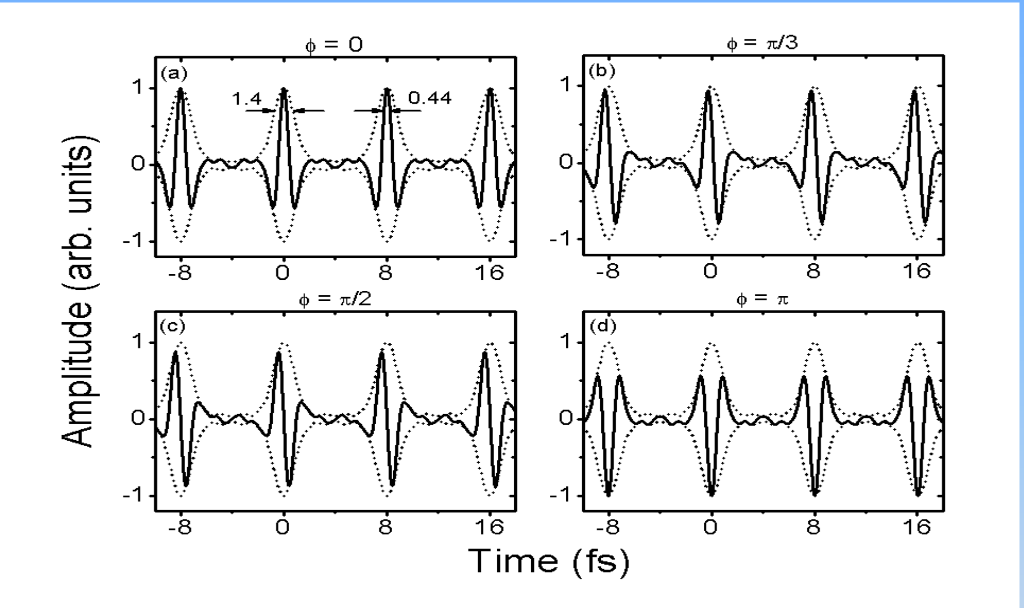
Carrier envelope phase is constant to ~2.5 part in 10^6

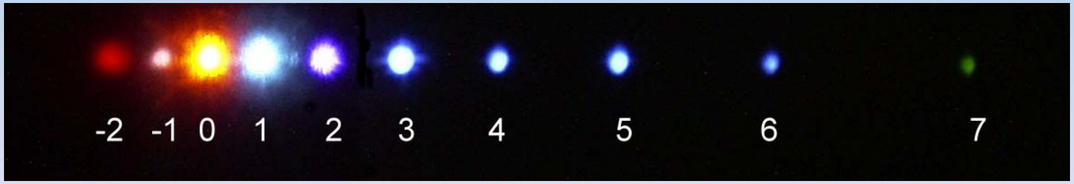
Total phase slip of <0.18 cycles over 1 million pulses

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





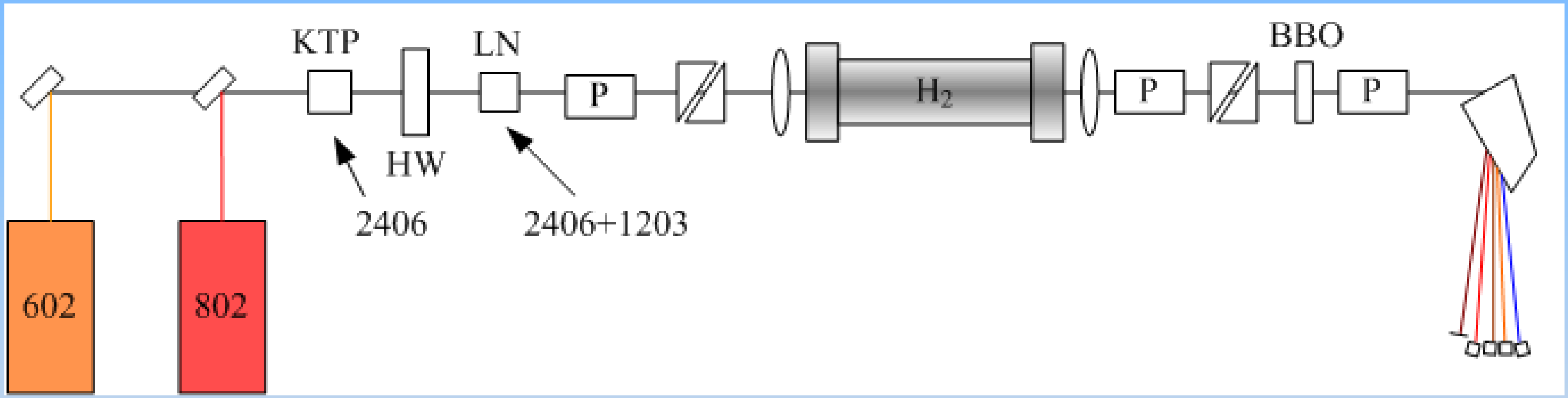
Total spectral span >70,000 cm^{-1}

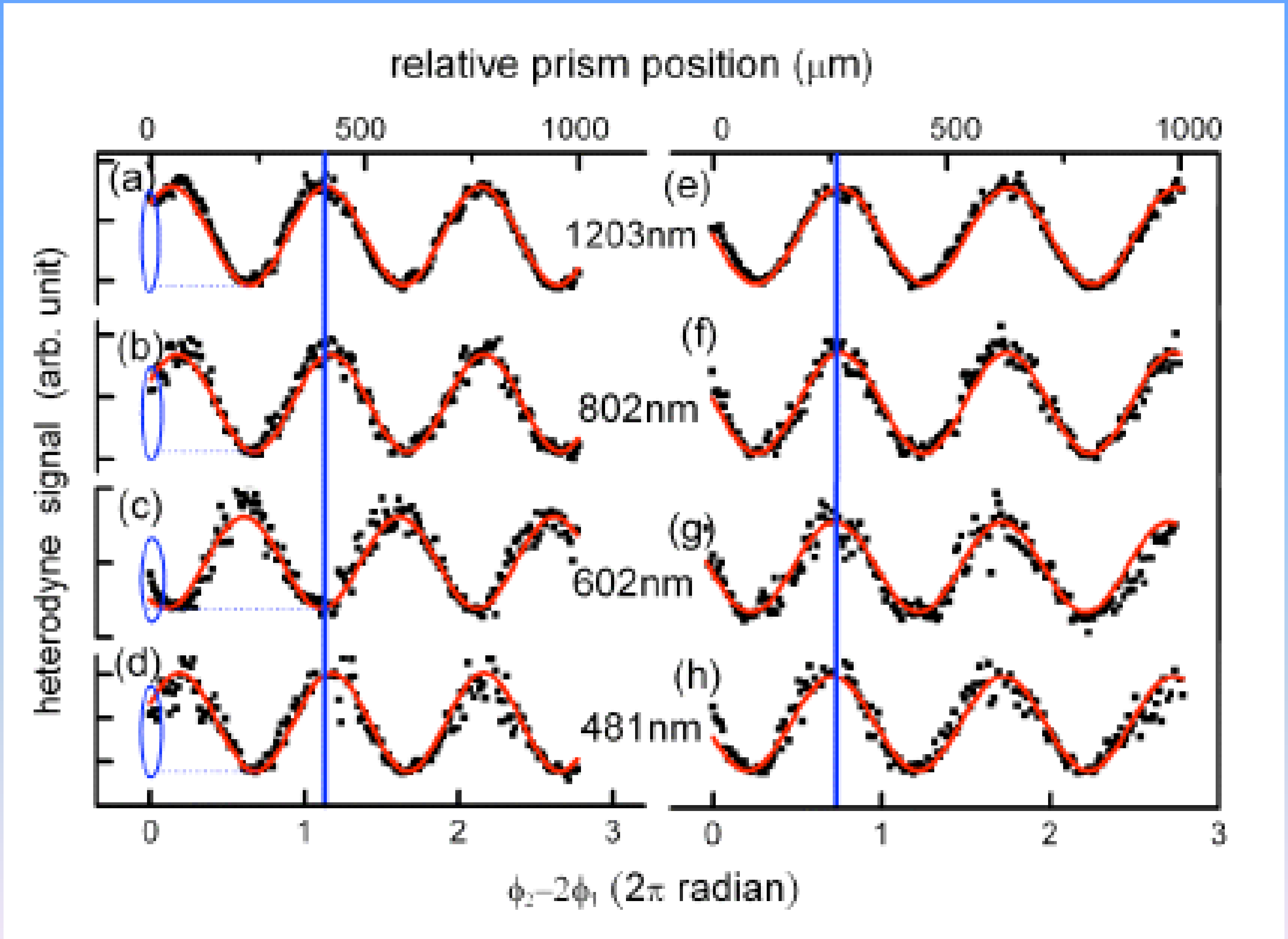
Wei-Jan Chen et al., Phys. Rev. Lett. 100, 163906 (2008)
Zhi-Ming Hsieh et al., Phys. Rev. Lett. 102, 213902 (2009)

Ingredients of an attosecond single-cycle optical pulse:

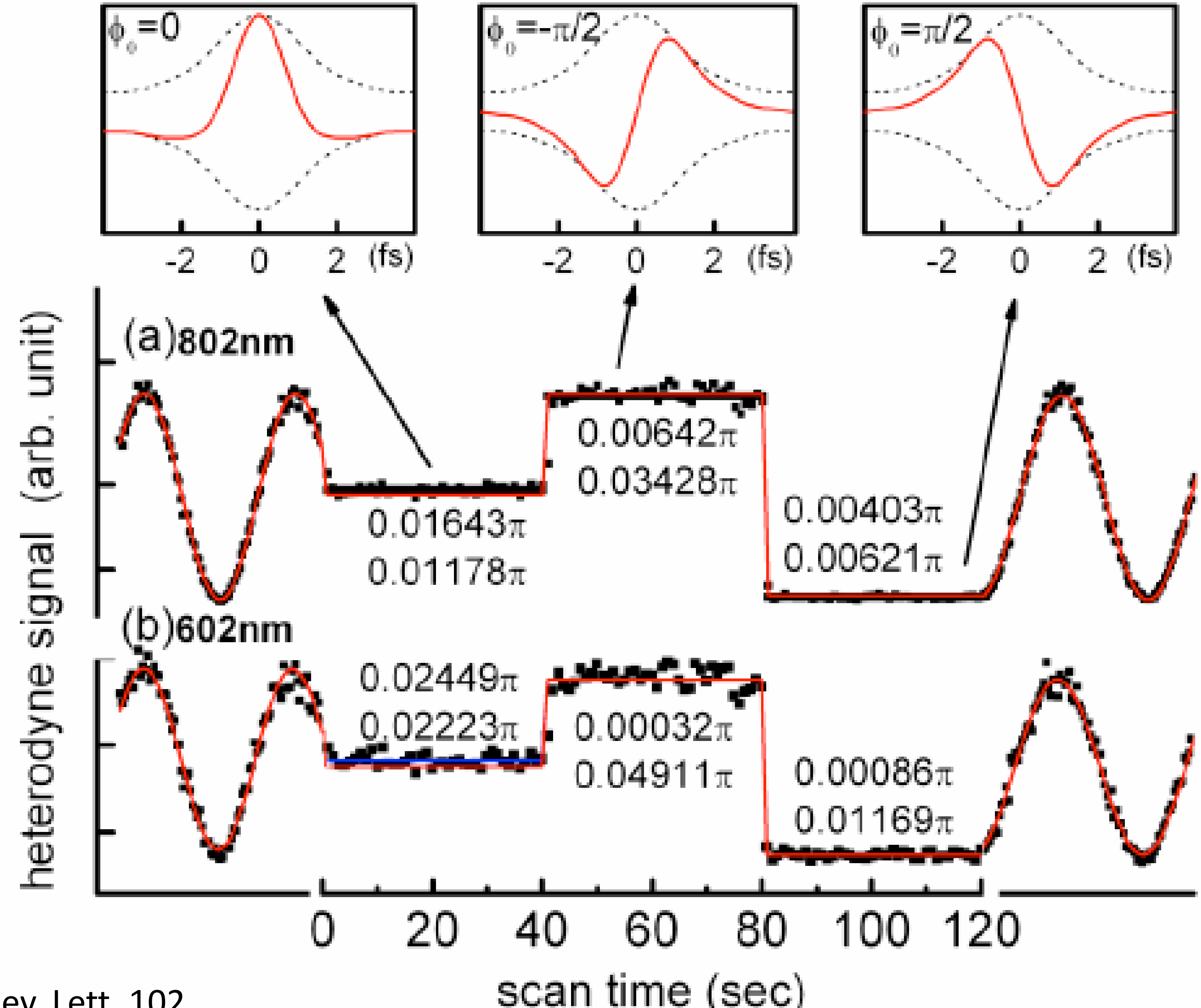
1. Broad spectrum – 2 or more octaves
2. In phase condition
3. Constant carrier envelope phase:
   - Commensurate frequencies
   - Constant phase difference between adjacent spectral components
4. **Stable and controllable carrier envelope phase**

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
$\phi_2-2\phi_1$ (2π radian)



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

Brief 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

Outline

- Introduction
- Review of basic concepts
- High Harmonic Generation
- Molecular Modulation
- Multicolor synthesized
- Outlook

Motivation

- Broadband source
  coherent and commensurate
- High peak power enough
  10^{13}-10^{14} W/cm^2
- Simple experiment setup
- Light waveform control

Methods of Generating Attosecond Pulses

C Harmonics Generation

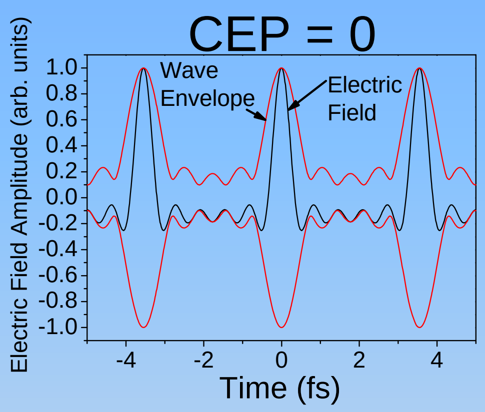
1203
802
602
481
401
344
301

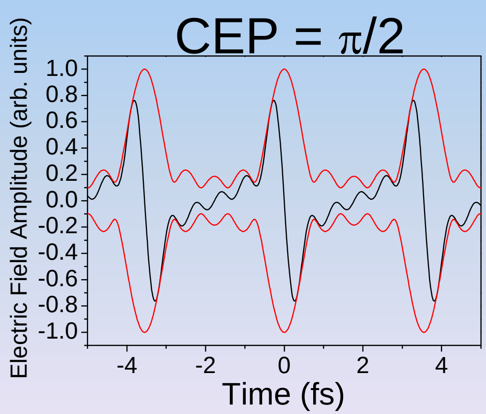
~25,000cm^{-1}

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

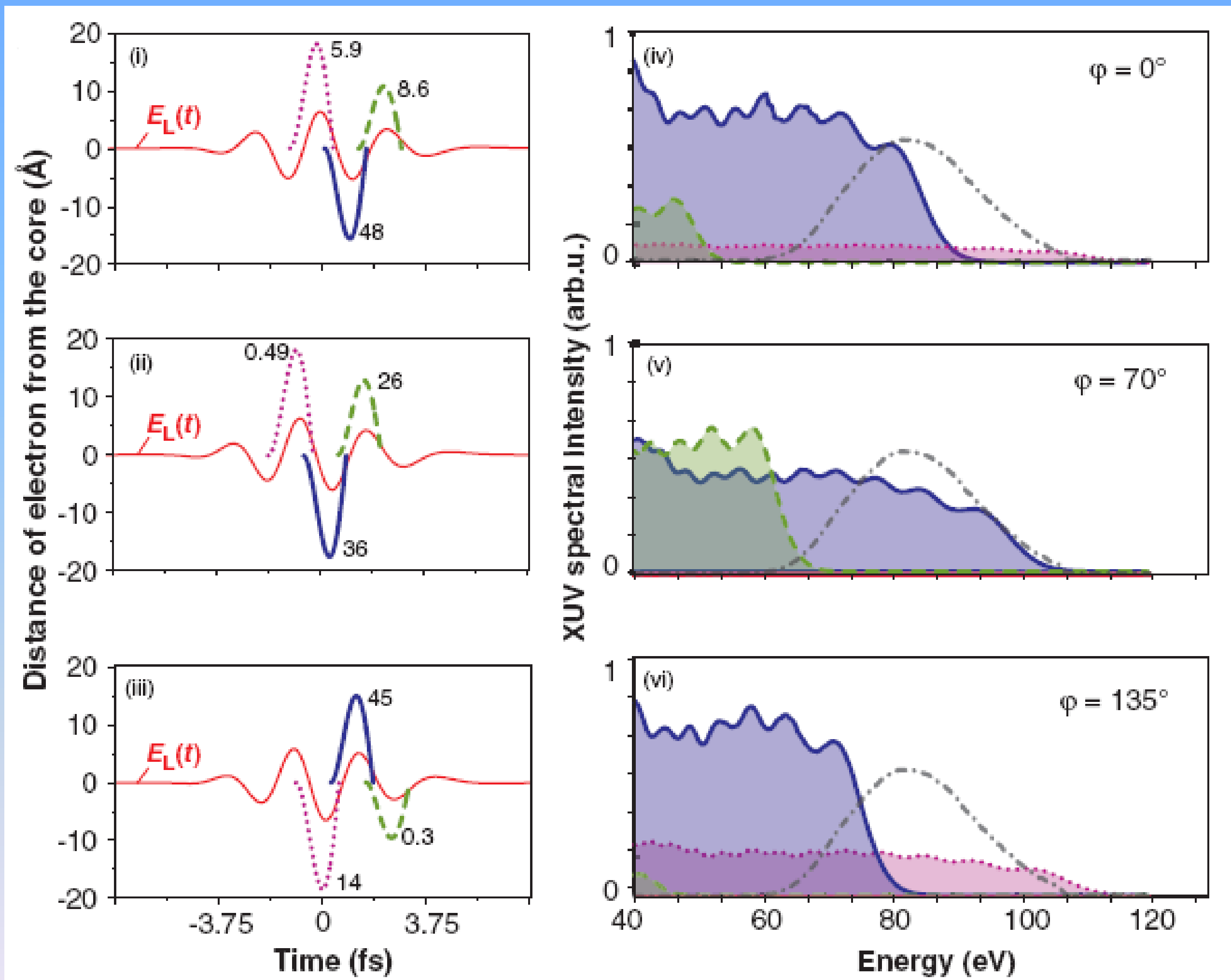
~37,600cm^{-1}





Single-cycle Nonlinear Optics

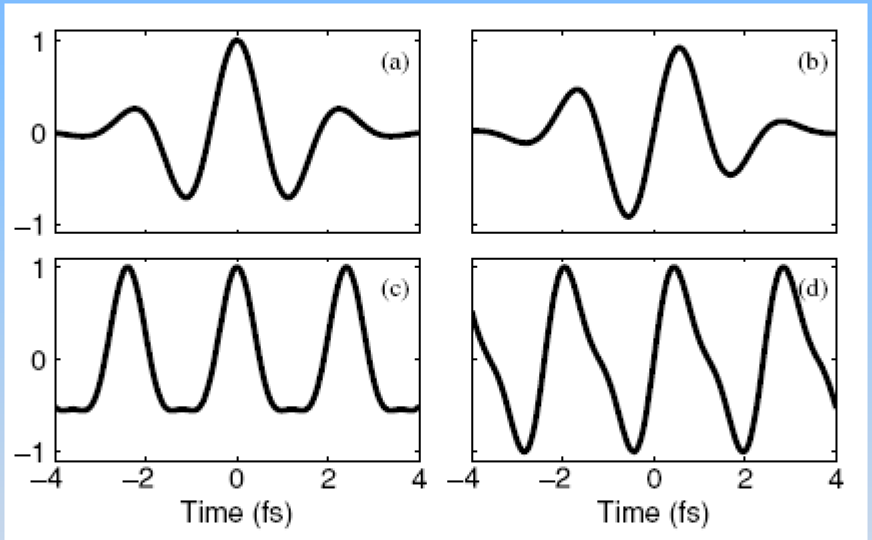
Simulation of sub-femtosecond XUV emission from neon atoms ionized by a linearly polarized, sub-1.5-cycle, 720 nm laser field.



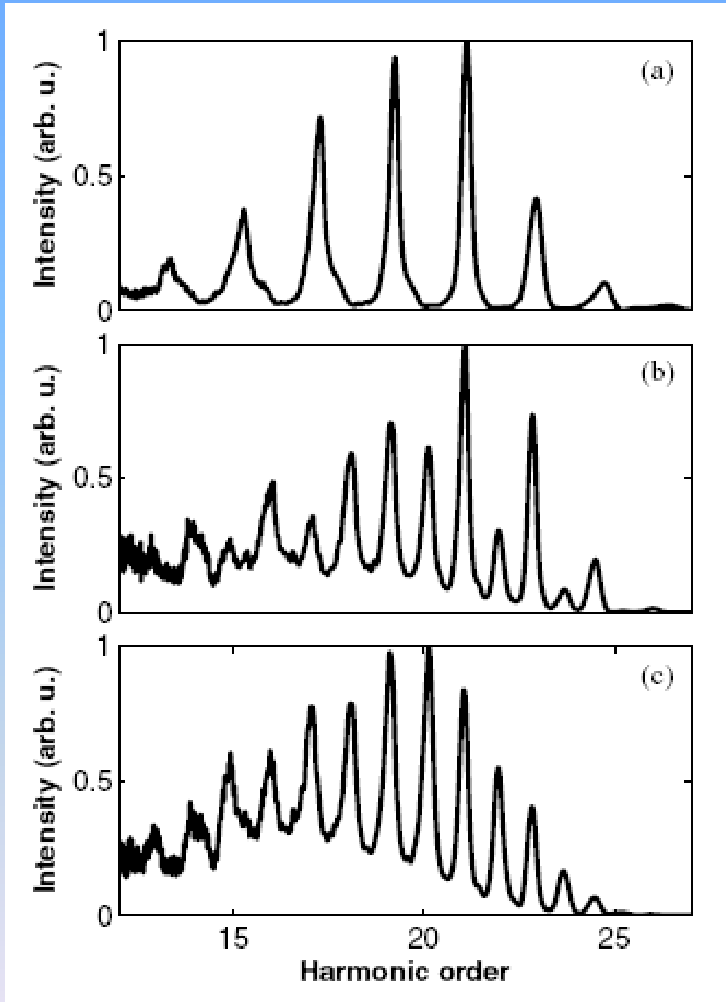
E. Goulielmakis, et al., Science 320, 1614 (2008)

Two-color multi-cycle field

Few-cycle pulse

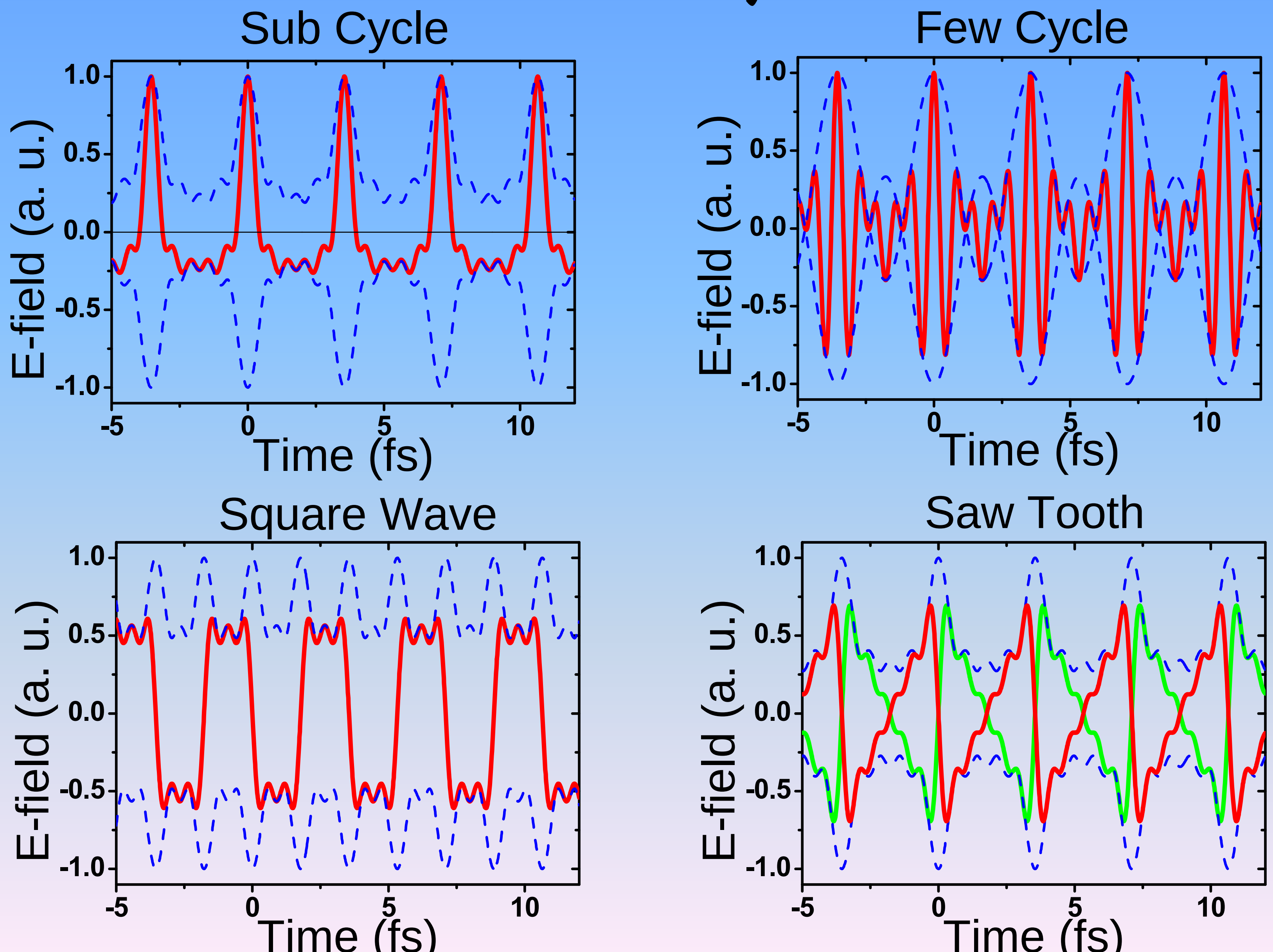


Two-color pulse train

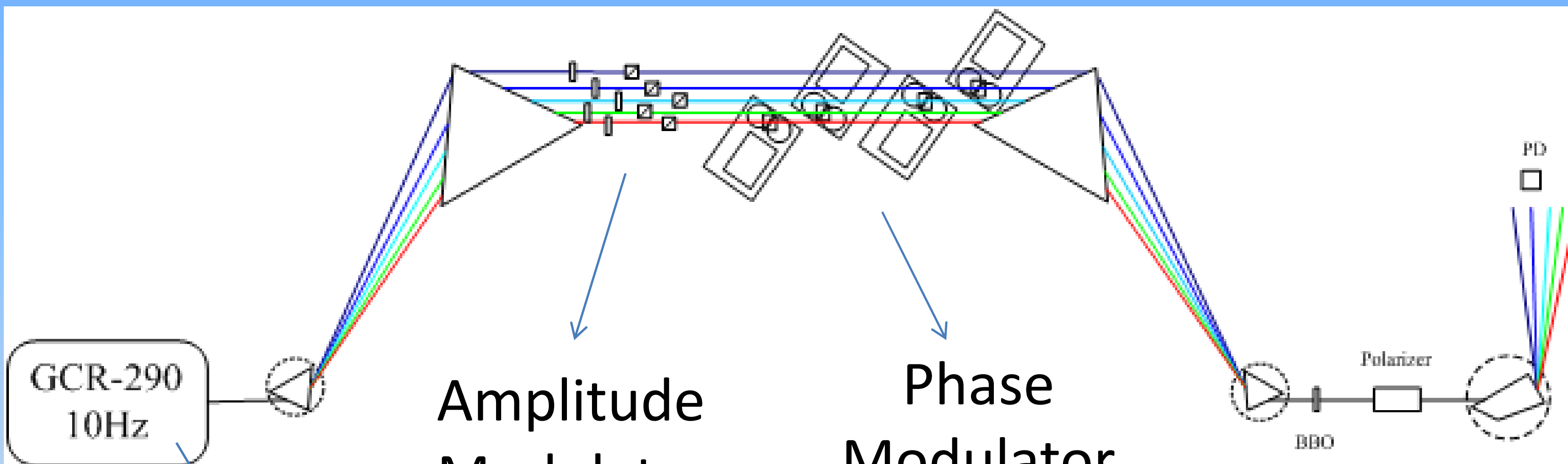


J Mauritsson et al., J. Phys. B: At. Mol. Opt. Phys. 42, 134003 (2009)

Waveform by 5 E-field

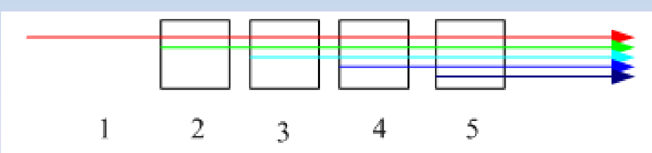


Experimental Setup



Amplitude Modulator

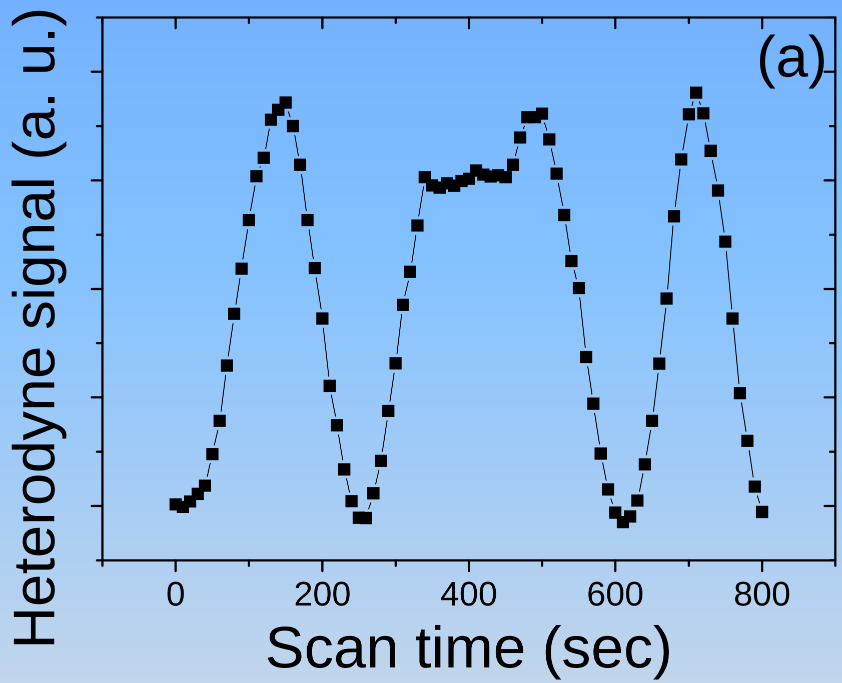
Phase Modulator



Harmonics generation

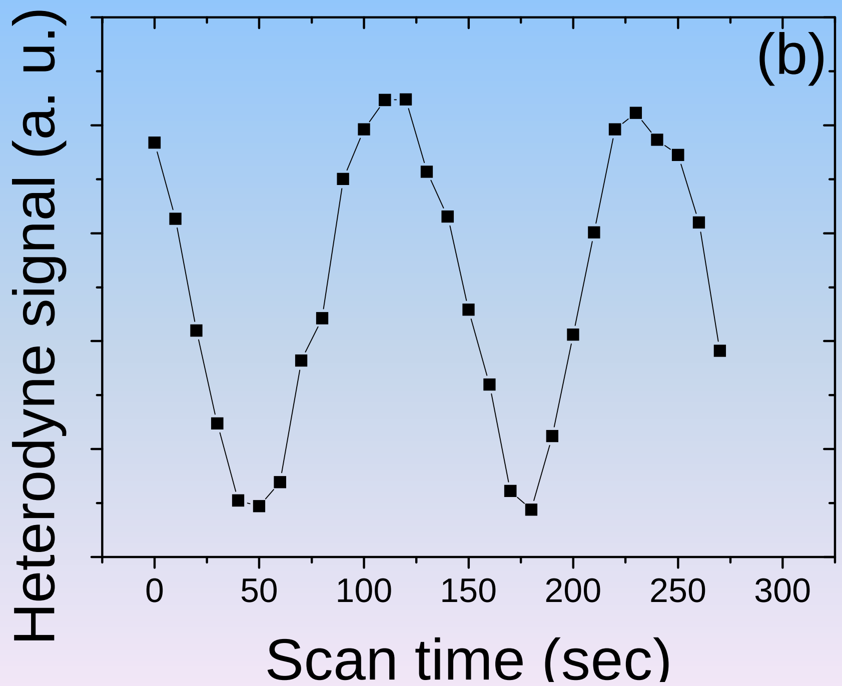
Relative Phase Measurement

Relative Phase Measurement

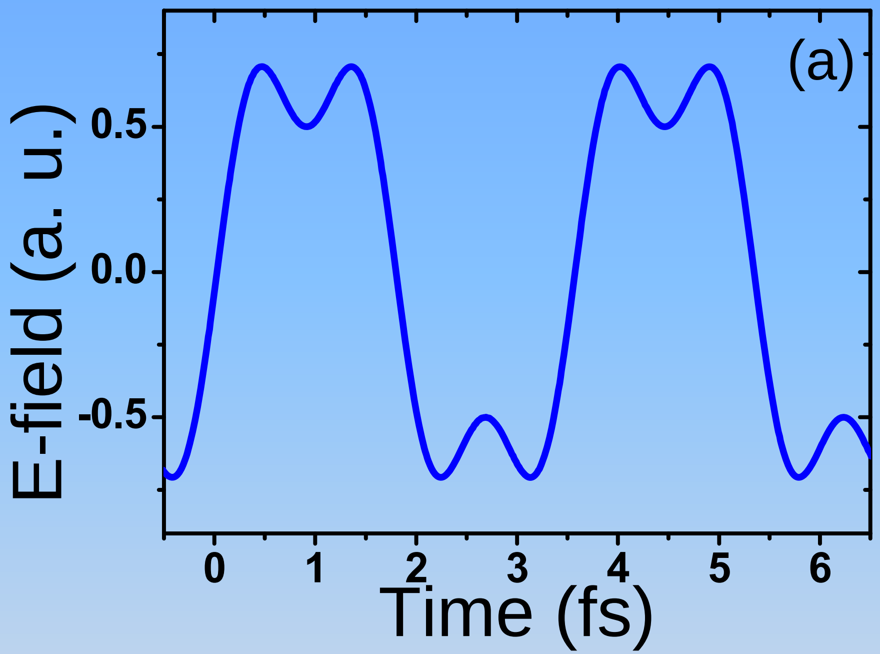


(a) The relative phase between 1064 nm and 532 nm

(b) The relative phase between 1064 nm and 355 nm

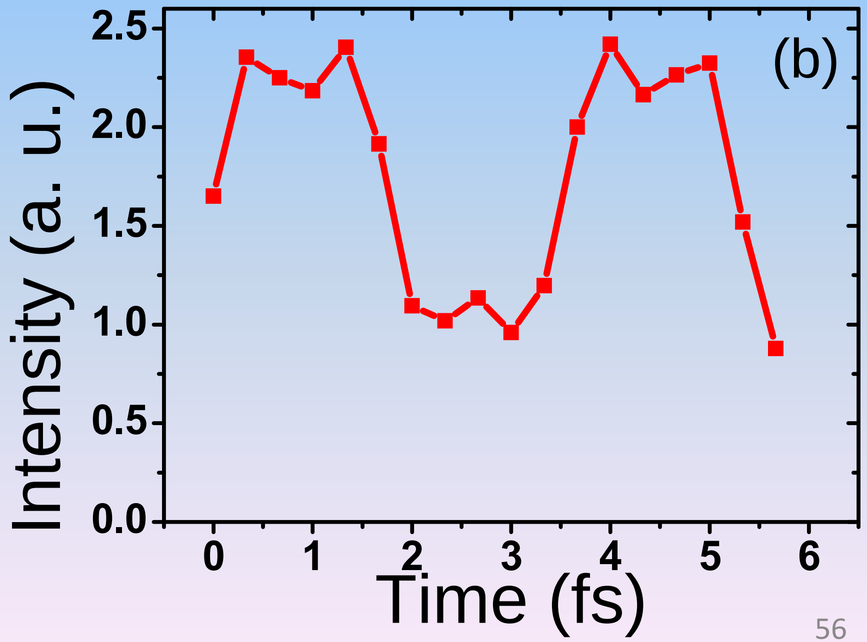


Waveform by linear cross-correlation



(a) Square wave synthesized by 3 harmonics

(b) Square wave shown by linear cross-correlation process



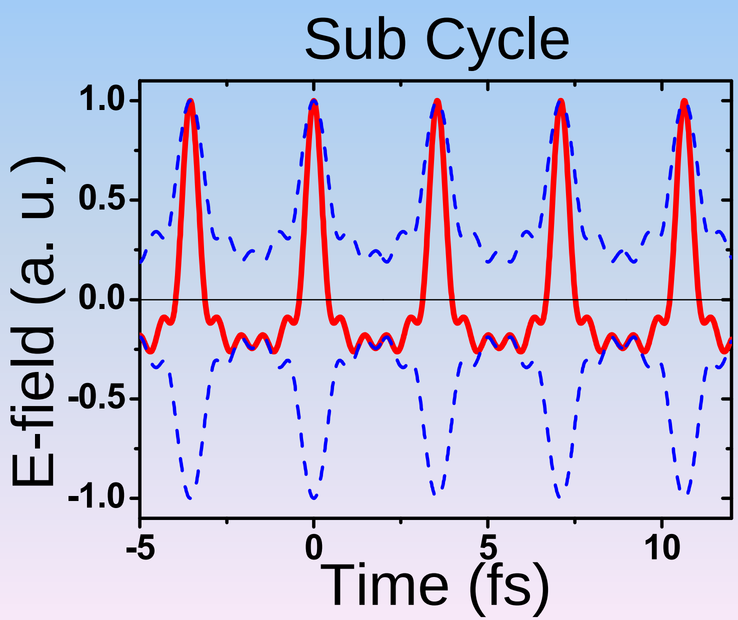
Outline

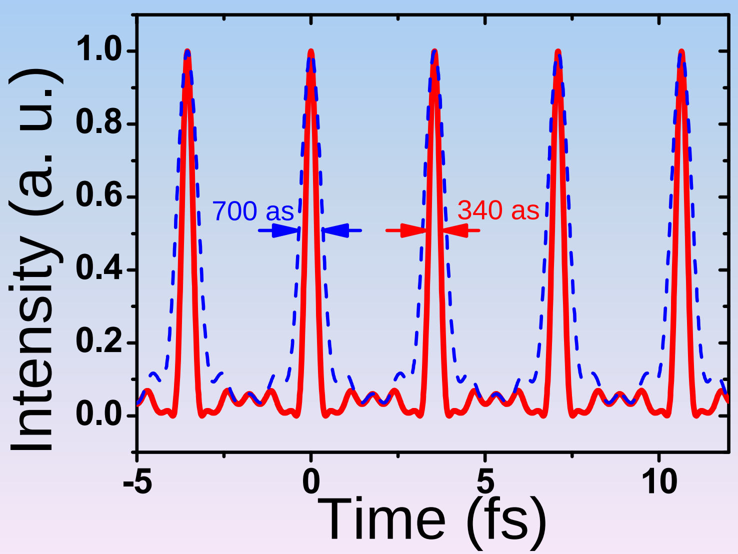
- Introduction
- Review of basic concepts
- High Harmonic Generation
- Molecular Modulation
- Multicolor synthesized
- Outlook

The characteristics of the waveform

Pulse train: period 3.55 fs
Pulse energy
1064 nm: 380 mJ
532 nm: 178 mJ
355 nm: 70 mJ
266 nm: 41 mJ
213 nm: 22 mJ

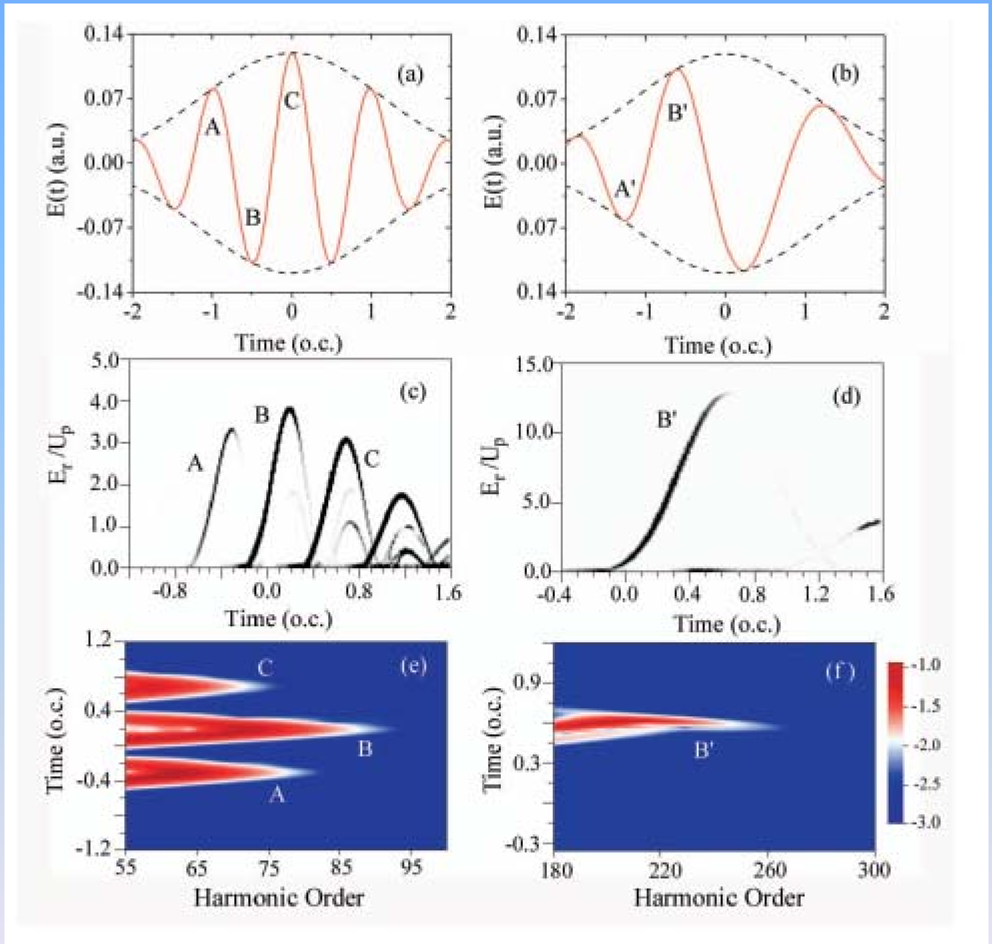
When CEP=0
After these modulator, assume each effective pulse energy is half of original energy
=>The pulse width ~ 340 as
Focusing to a Φ20μm spot, the intensity will reach 10^{14} W/cm^2.





Outlook

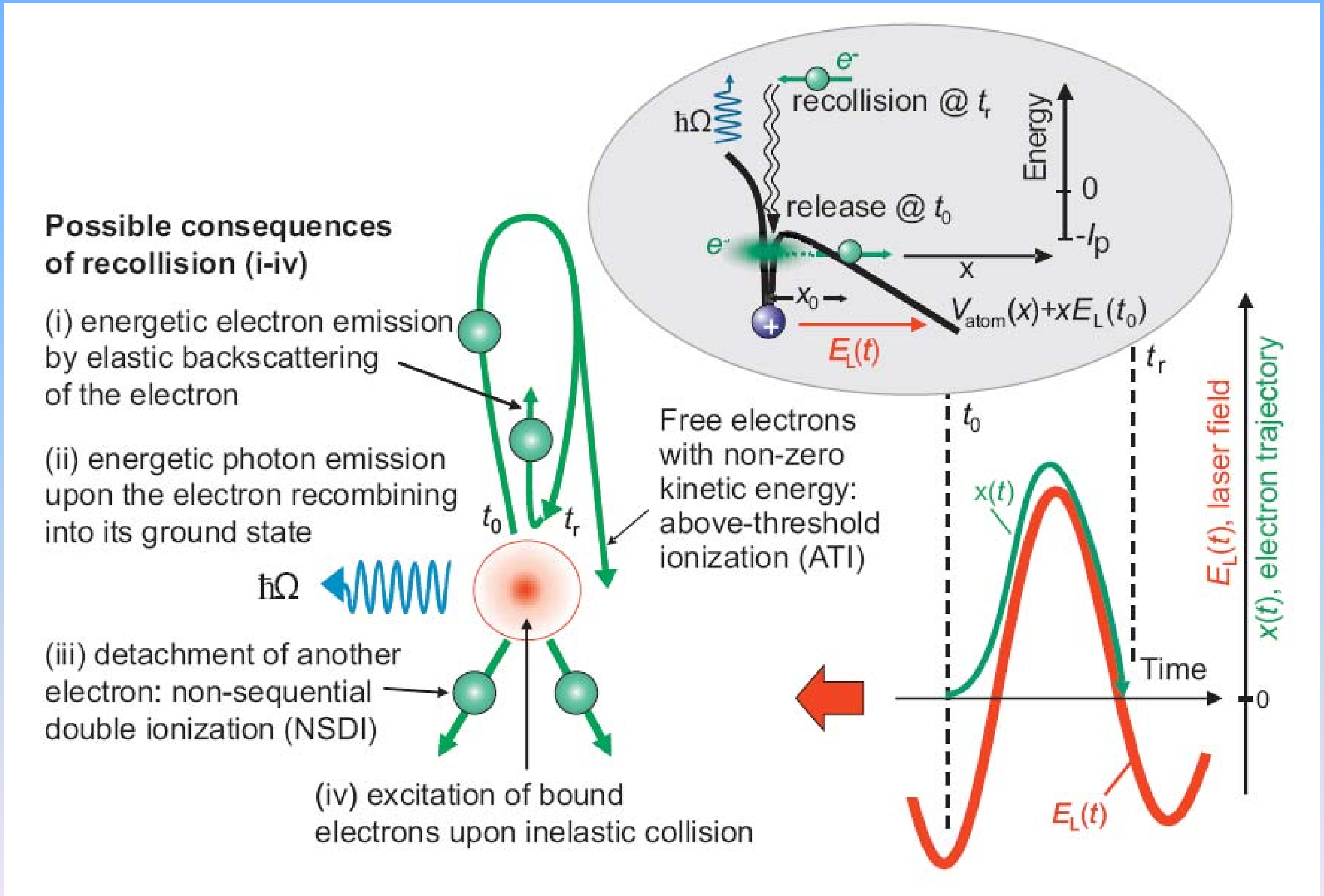
- The further waveform control
- The HHG setup and measurement



Juan J. Carrera and Shih-I Chu, Phys. Rev. A 75, 033807 (2007)

Outlook

- Photoelectron and/or ion measurement



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

Collaboraters

National Tsing Hua University

Andy Kung(孔慶昌) Ci-Ling Pan(潘犀靈) Wei-Jan Chen(陳蔚然)
Han-Sung Chan(詹翰松) Hong-Zhe Wang(王宏哲)
Wei-Hsiang Wang(王韋翔)

IAMS Zhi-Ming Hsieh(謝智明) Shu Wei Huang(黃書偉)
Hao-Yu Su(蘇皓瑜) Chien-Jen Lai(賴建任) Sih-Ying Wu(吳思螢)

National Chiao Tung University:

Ru-Pin Pan(趙如蘋) Tsung-Ta Tang(湯宗達) Ruei-Yin Lin(林睿茵)

National Sun-Yat-Sen University

Chao-Kuei Lee(李晁逵)

Acknowledgement

Stanford: Steve Harris and his students

TAMU: Alexei Sokolov

Berkeley: Ron Shen

Research supported by Academia Sinica and the NSC

Thanks for your attention