Attosecond Physics

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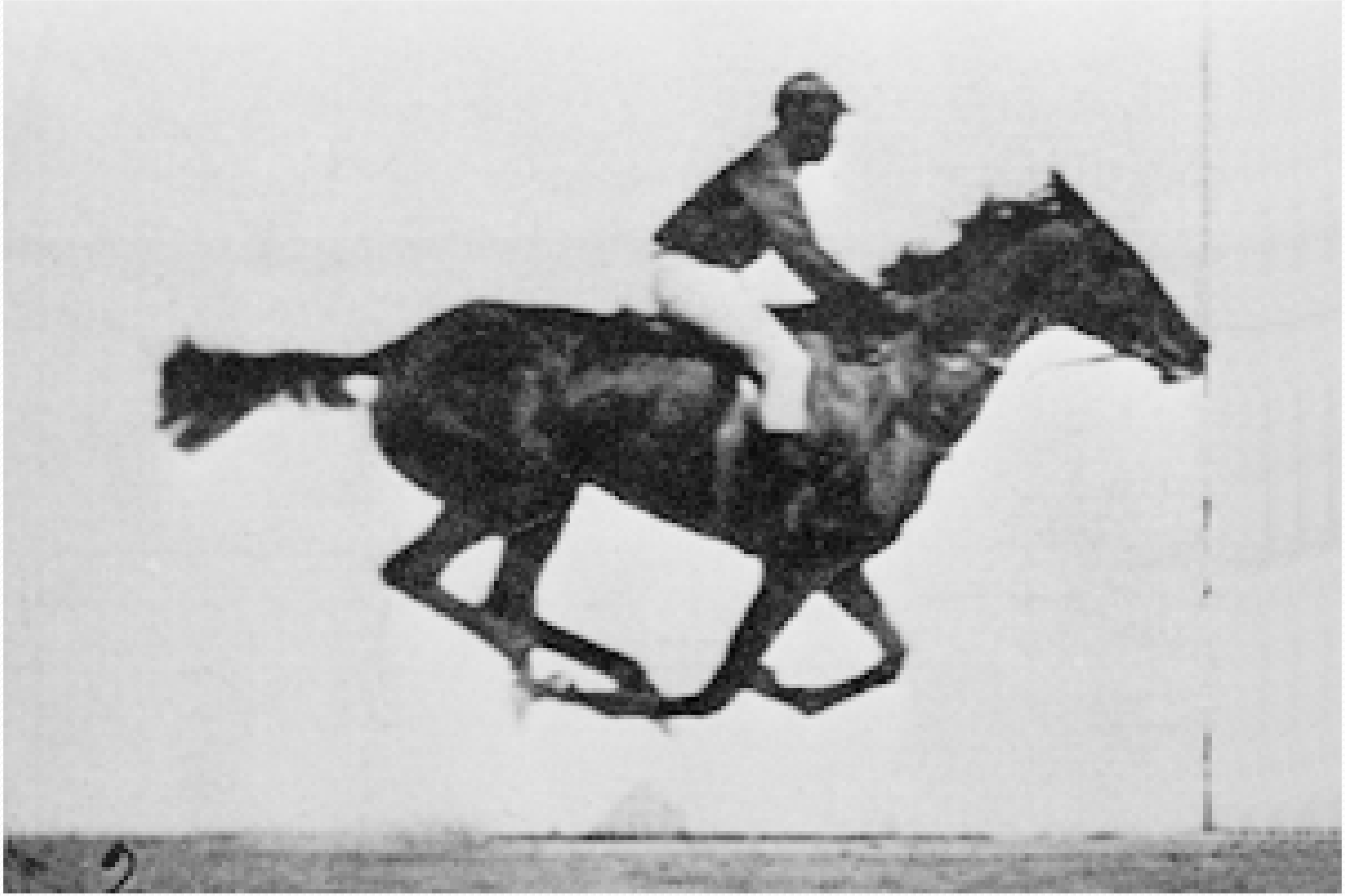
2015 AMO summer school

Aug. 27, Hsinchu

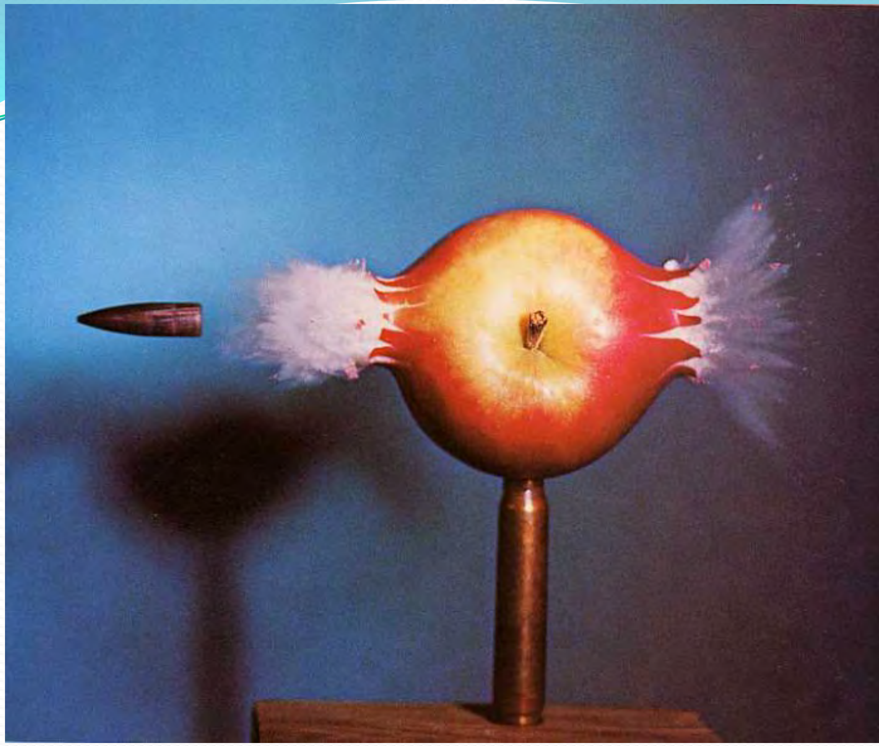
Nobel Prize in Chemistry

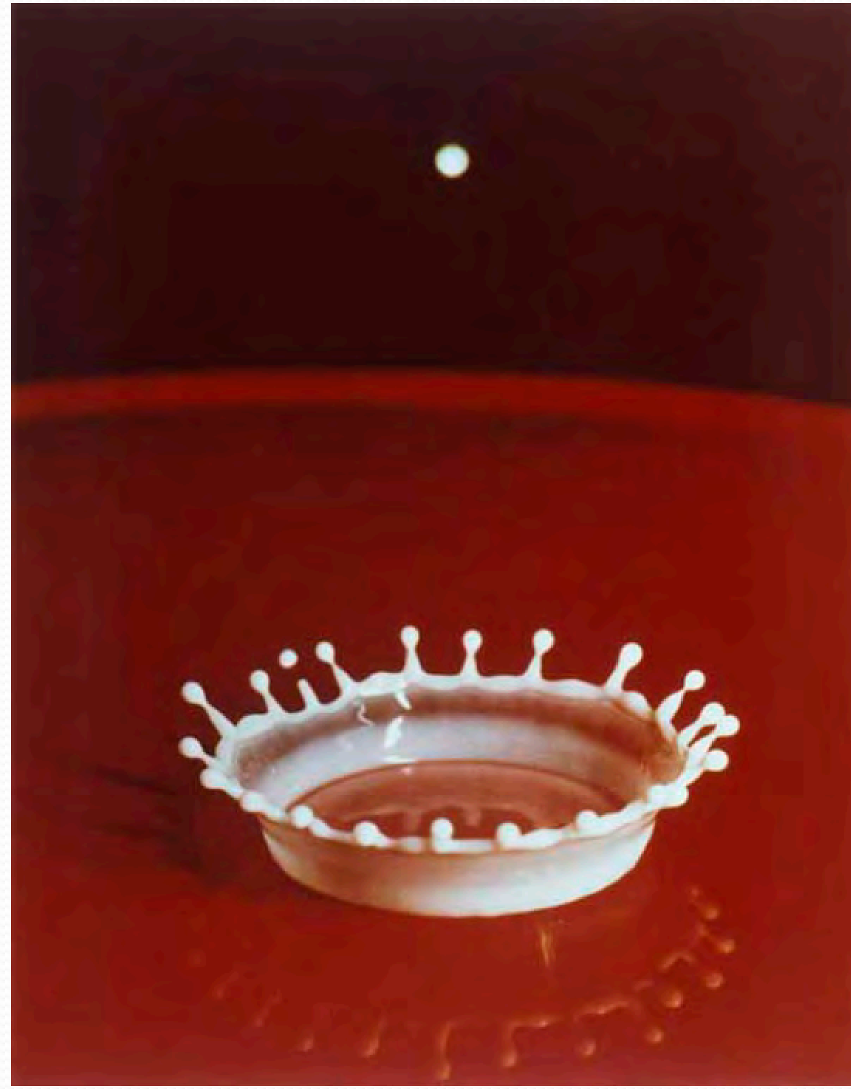
- **1999: Ahmed Zewail**
  Zewail successfully used a rapid laser technique called femtosecond spectroscopy to observe how atoms in a molecule move during a chemical reaction. In femtosecond spectroscopy, a pump-probe experiment "photographs" chemical reactions as they happen, using an ultrafast laser as "flash".

Femtochemistry



Eadweard Muybridge





Harold Edgerton

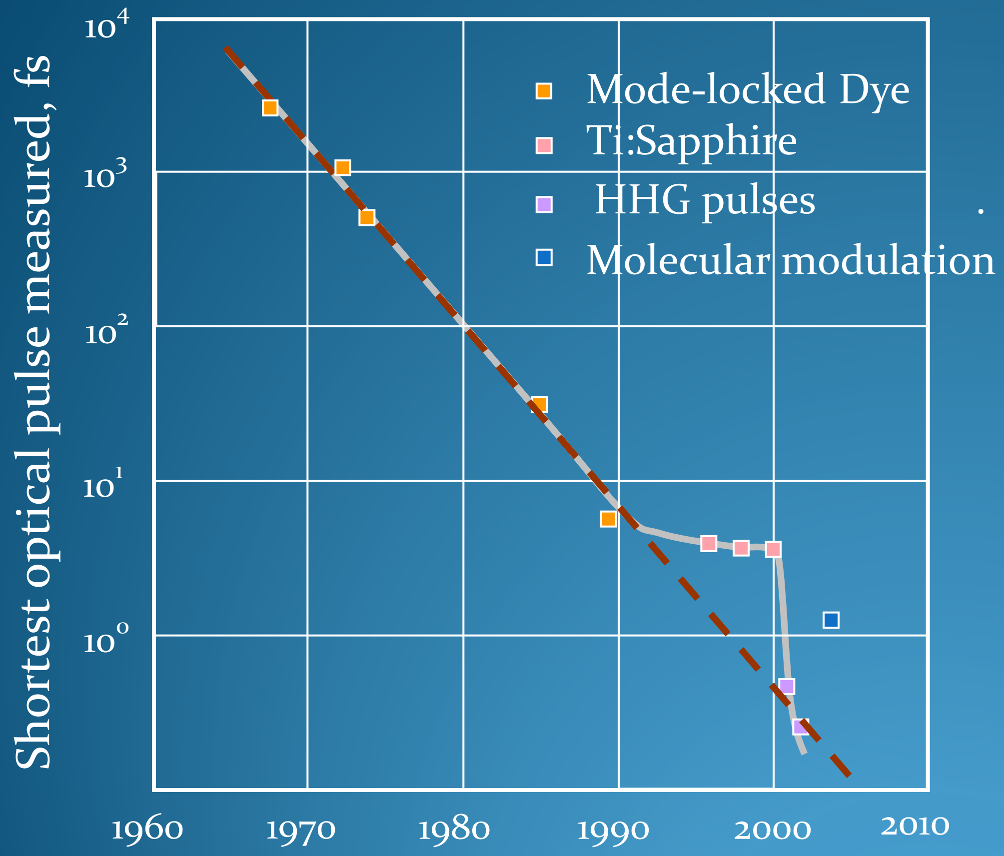
What is Attosecond Physics

- Ultrafast optics
- Strong field physics

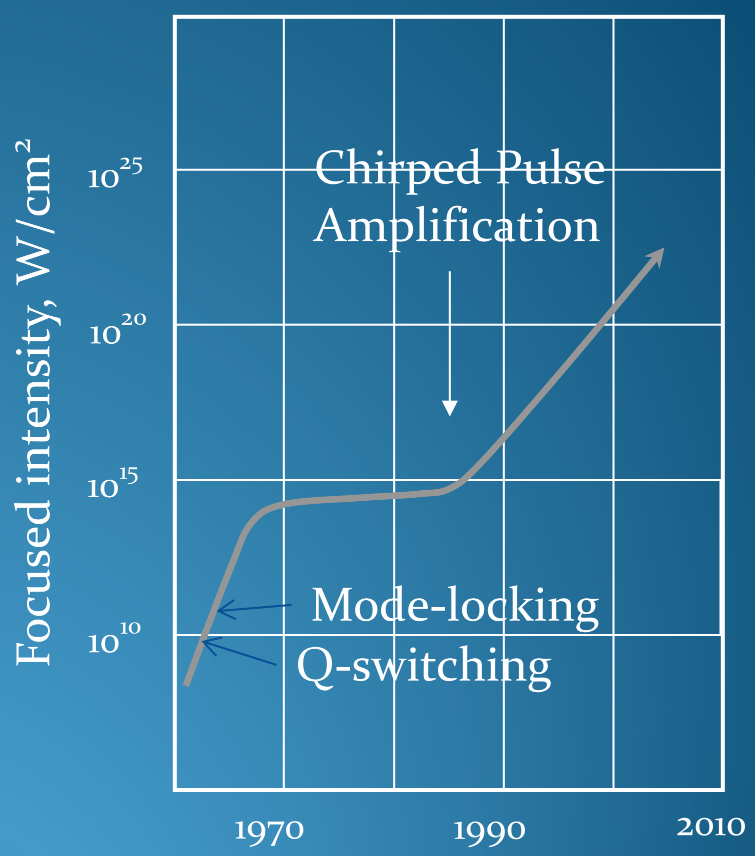
One Atomic unit of length a_0=0.0529 nm
One Atomic unit of electric field E_H = 5.142 x 10^9 V/cm
One Atomic unit of intensity E_0 = 3.55 x 10^{16} W/cm^2
One Atomic unit of time T_a = 24.2 as

Laser pulses got shorter over the years

Peak intensity increased



Ultrafast science

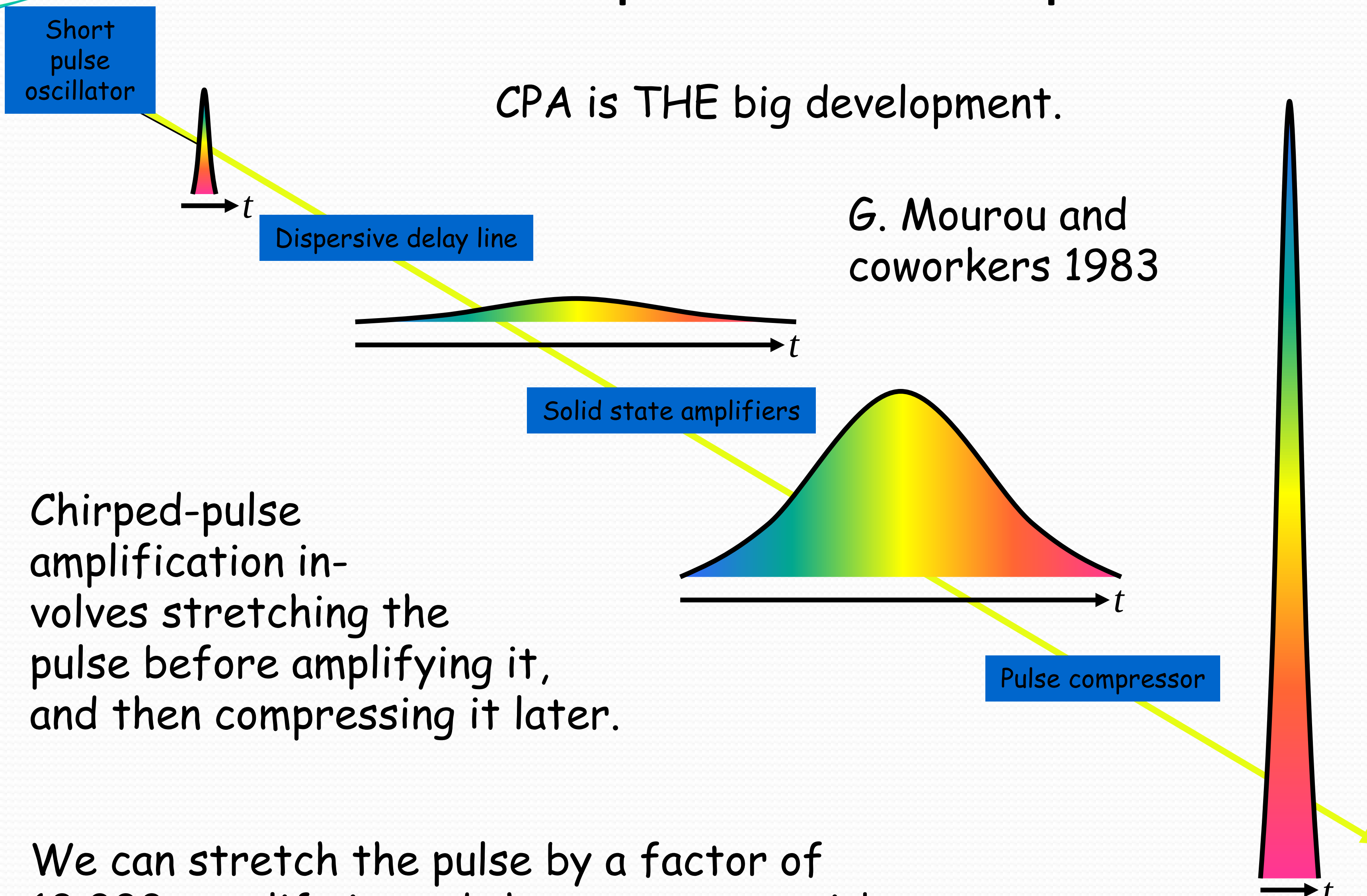


High field physics

Chirped-Pulse Amplification

CPA is THE big development.

G. Mourou and coworkers 1983



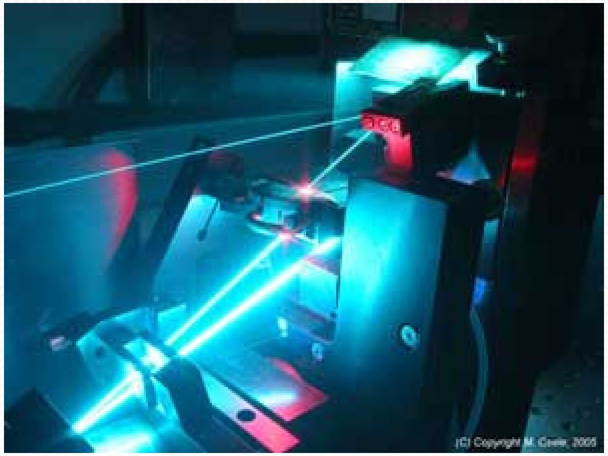
Chirped-pulse amplification involves stretching the pulse before amplifying it, and then compressing it later.

We can stretch the pulse by a factor of 10,000, amplify it, and then recompress it!

R. Trebino's lecture

Femtosecond Laser

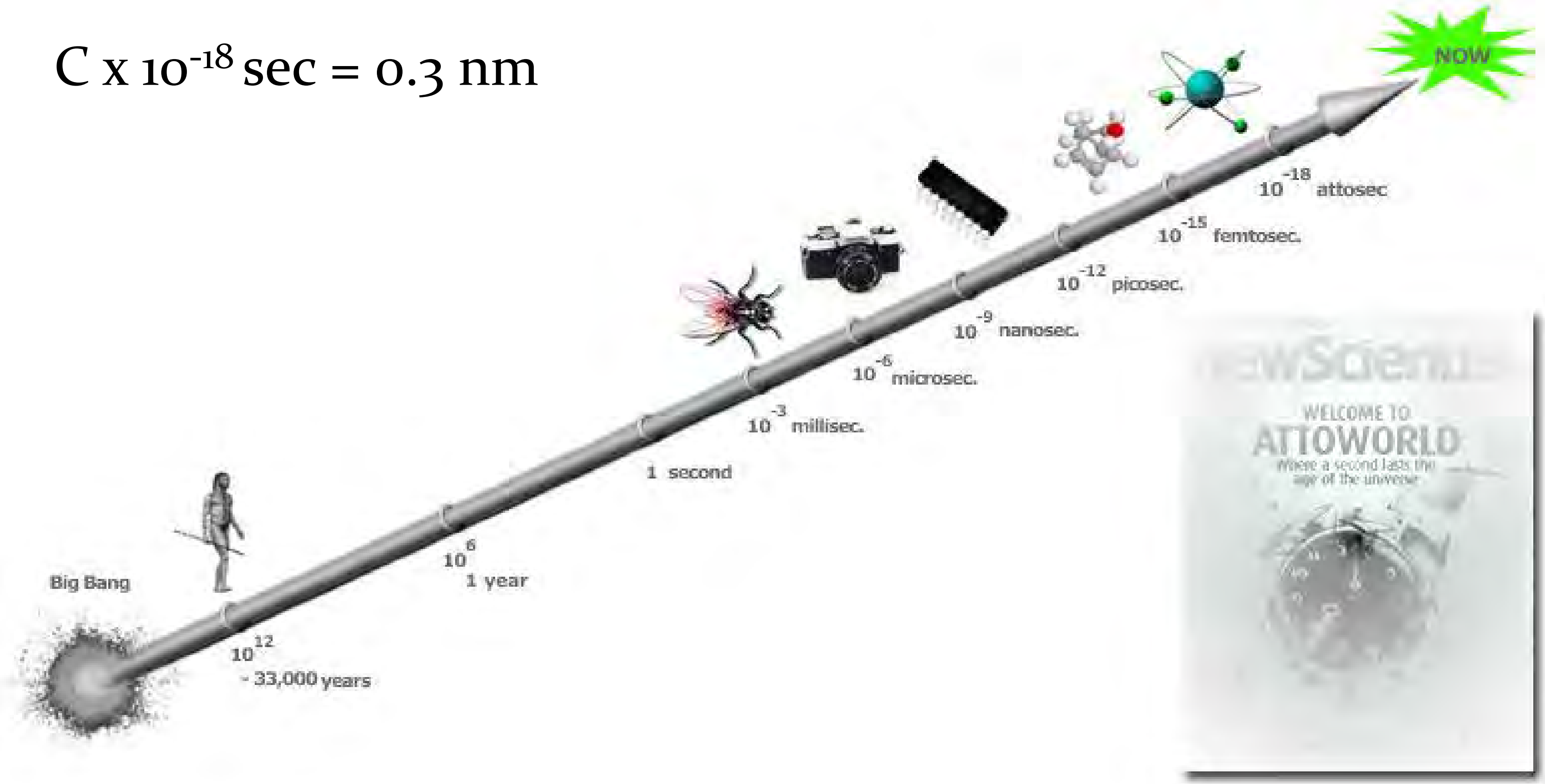
- 1974: E. P. Ippen and C. V. Shank develop the sub-picosecond mode-locked CW dye laser, establishing ultrafast optical science.
- 1982: P. F. Moulton develops titanium -sapphire laser. The titanium -sapphire laser replaces the dye laser for tunable and ultrafast laser applications.



Attoworld

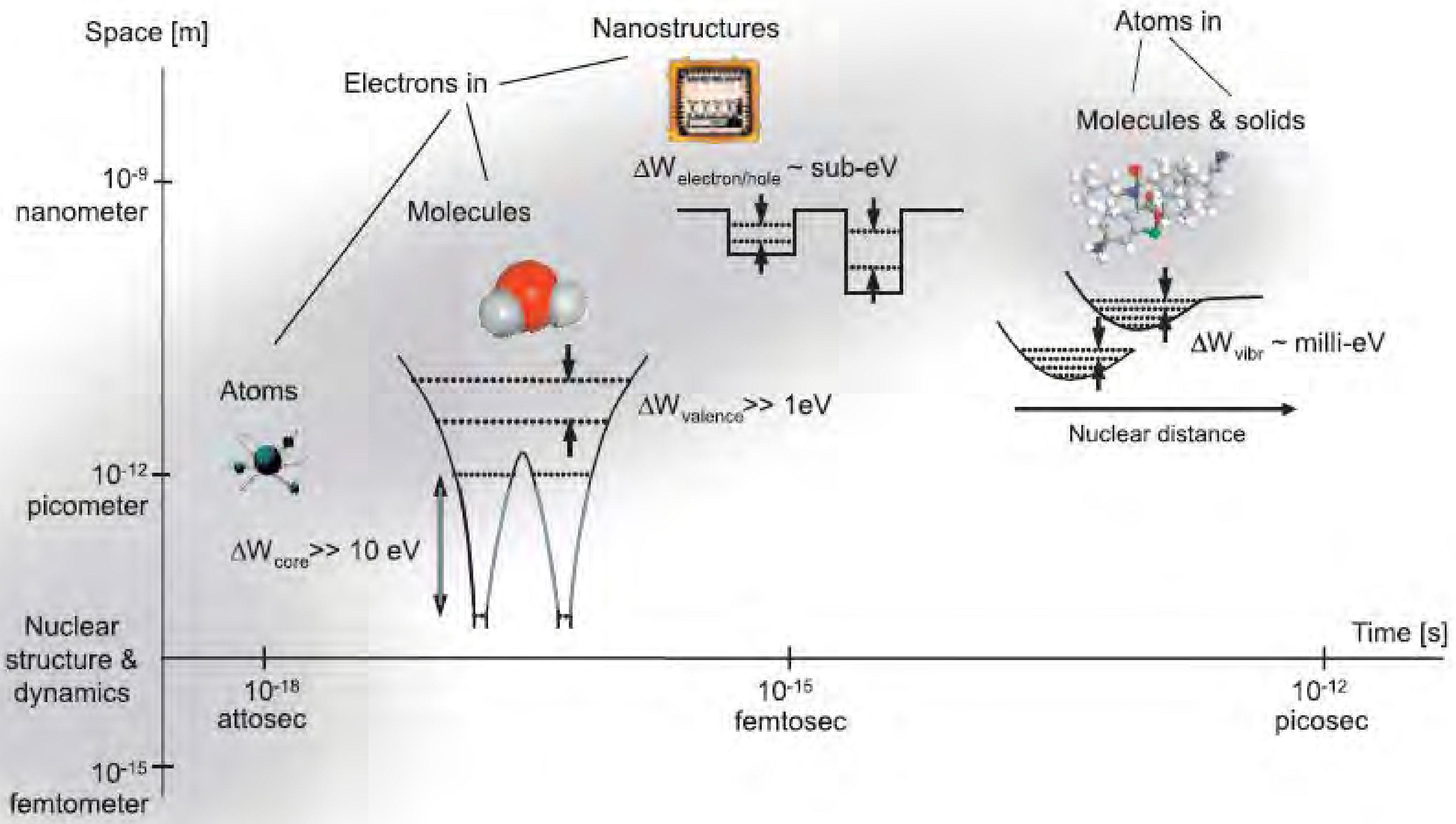
Attosecond: 10^{-18} second

$C \times 10^{-18}$ sec = 0.3 nm



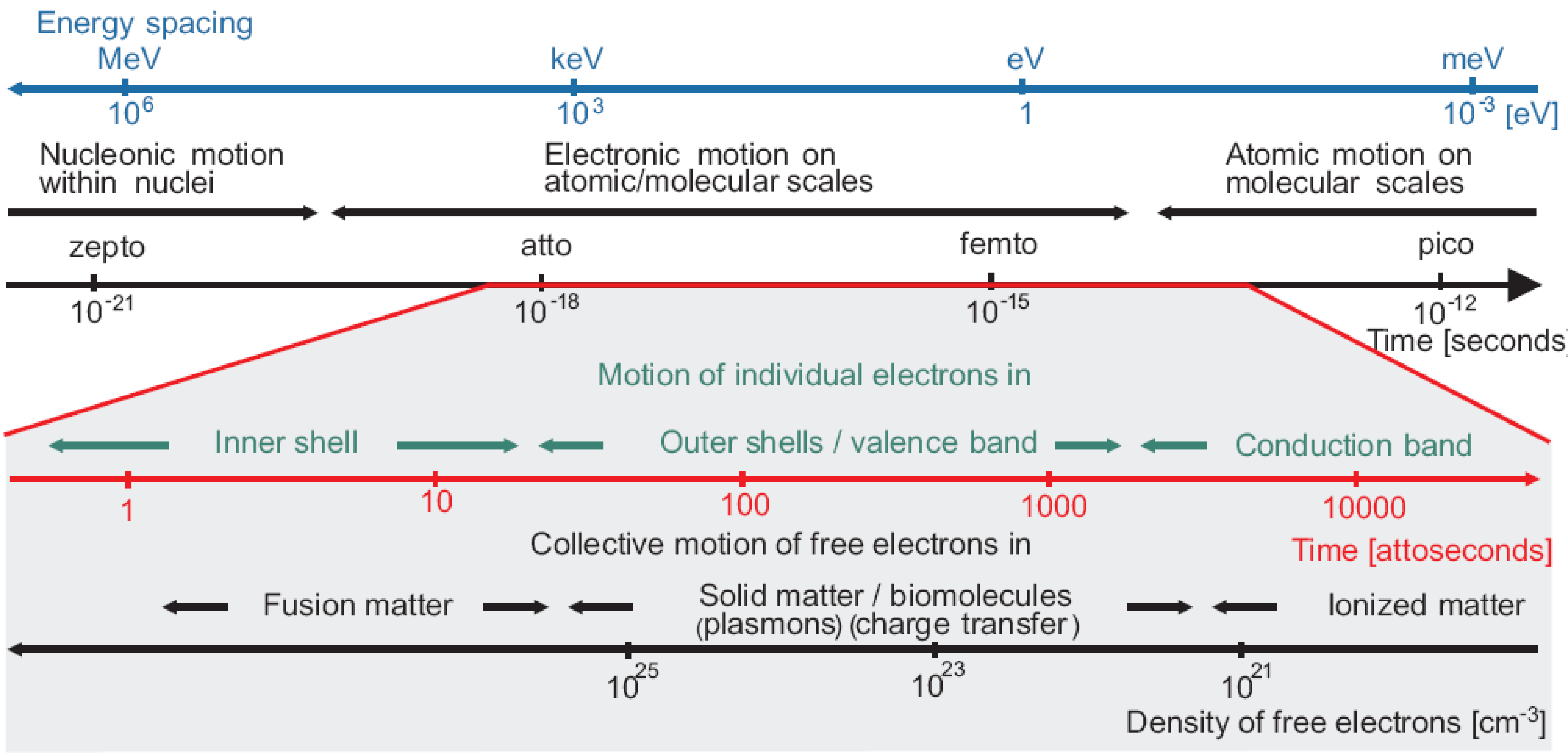
www.attoworld.de/attoworld.html

Characteristic length and time scales in the microcosm



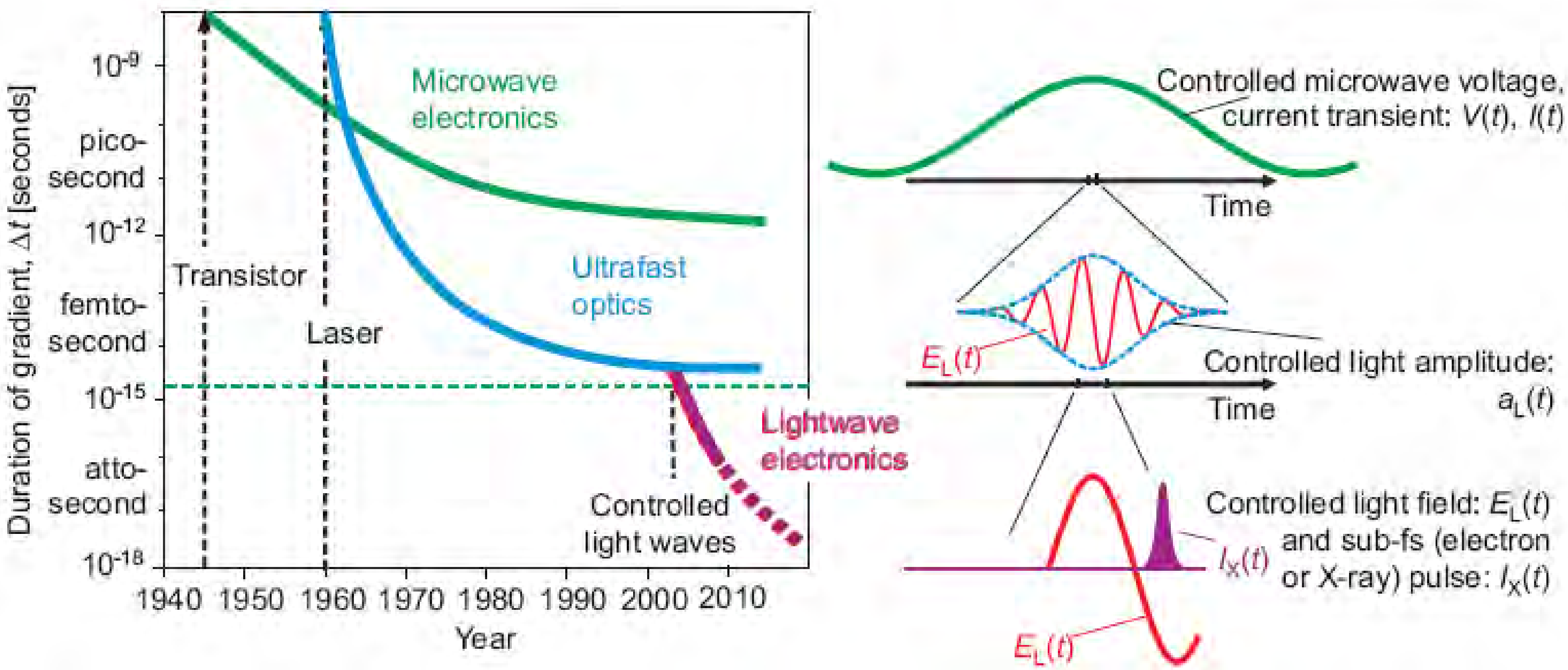
F. Krausz & M. Ivanov, Rev. Mod. Phys. 81 163 (2009)

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)

Attosecond physics

- Attosecond Pulse Generation and Characterization
- Broadband High-Harmonics Generation
- High Harmonic Spectroscopy
- Ultrafast Phenomena
- Strong Field Electronic and Nuclear Dynamics
- New Ultrafast Sources and Applications
- …

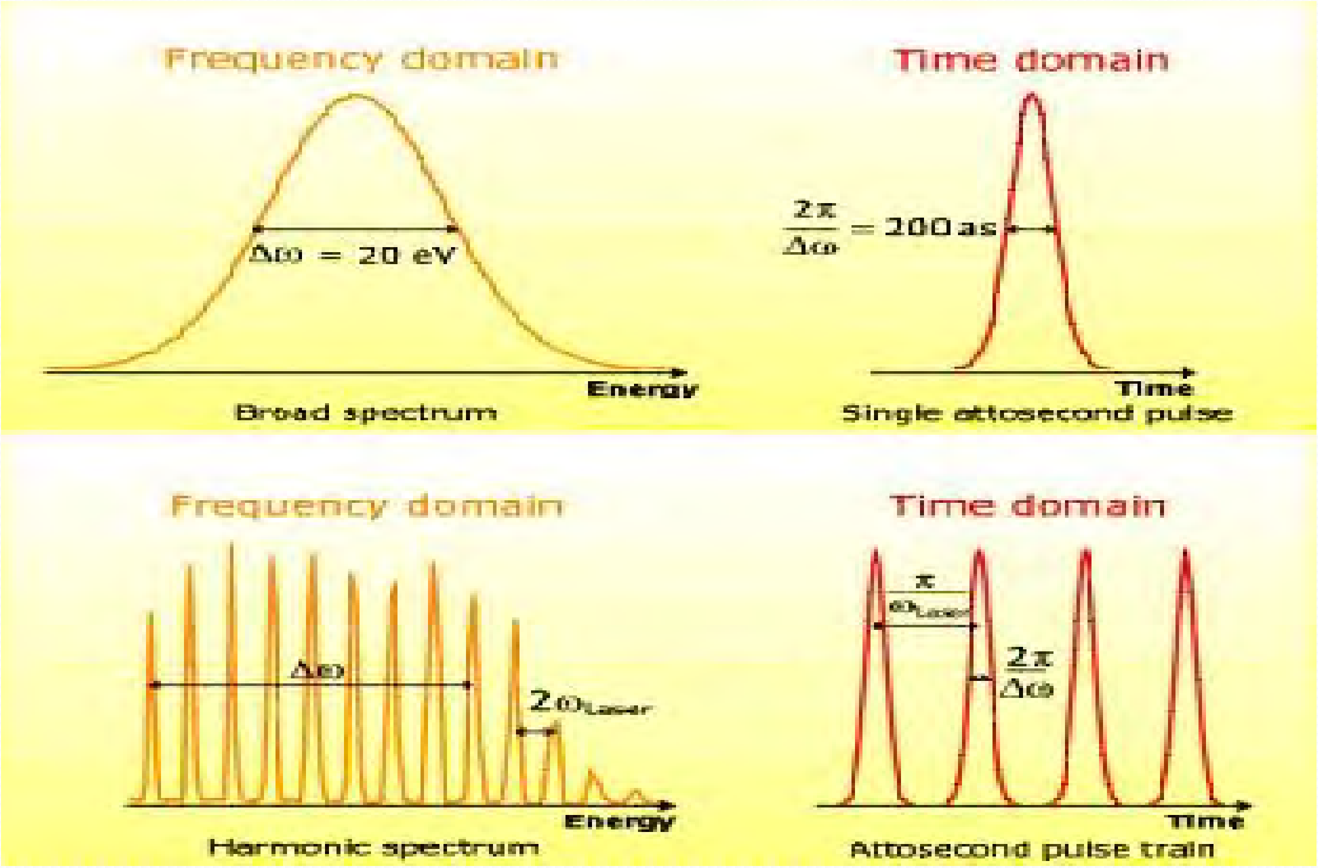
The experimental tools and techniques for electronic dynamics

- Few-cycle pulse with controllable CEP in NIR-VIS optical range
- Attosecond XUV pulse (isolated pulse, pulse train)
- Attosecond electron pulse
- Detectors for ion, electron, or photon

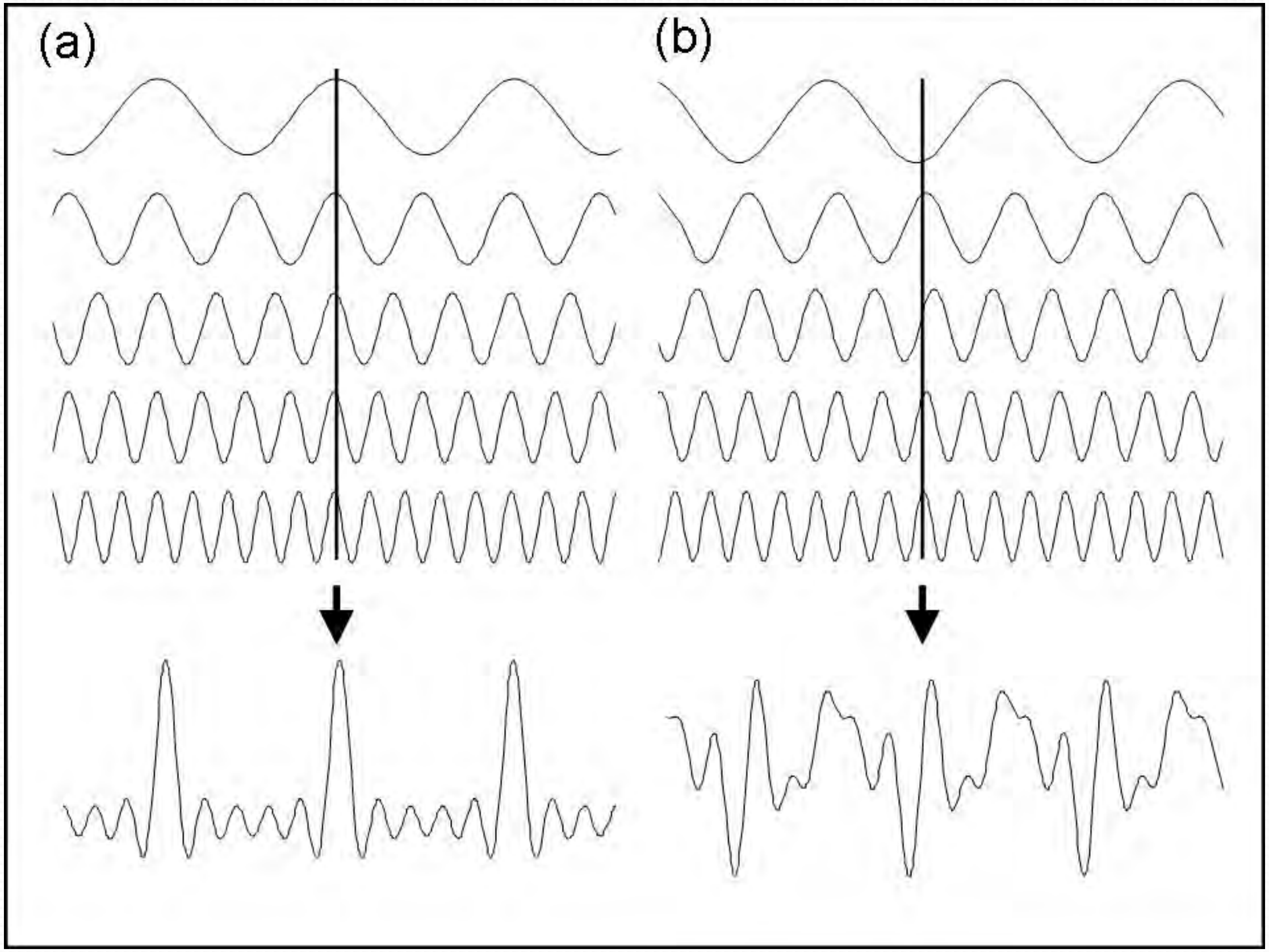
Correlation Between Time and Frequency

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

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



Principle of optical interference of coherent light fields



In phase Random phase

Optical cycle

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

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

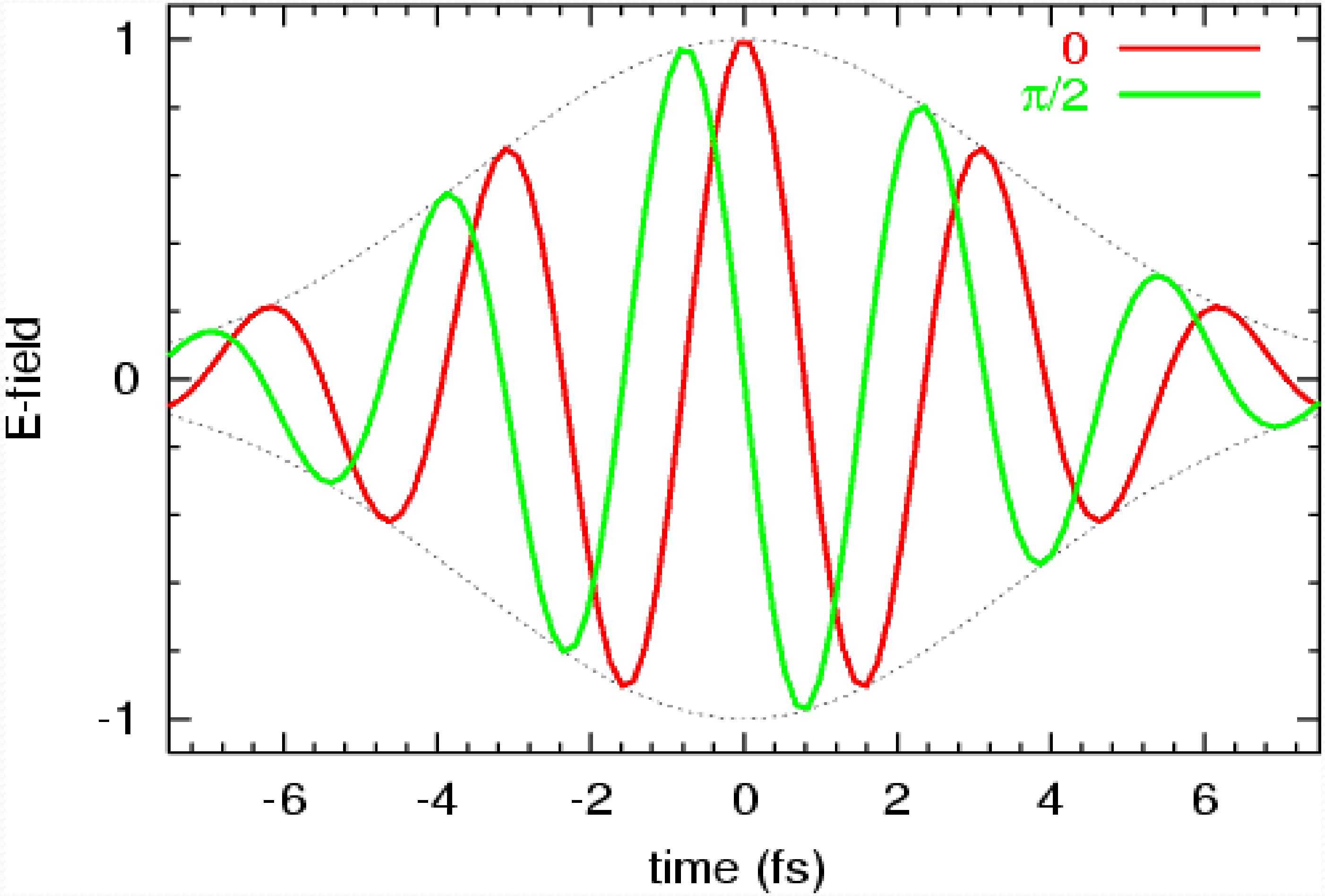
Carrier frequency

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

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

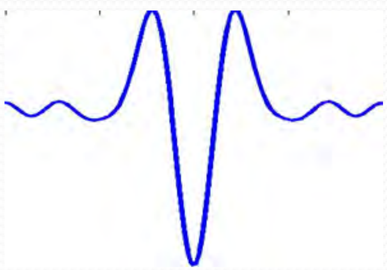
Carrier envelope phase

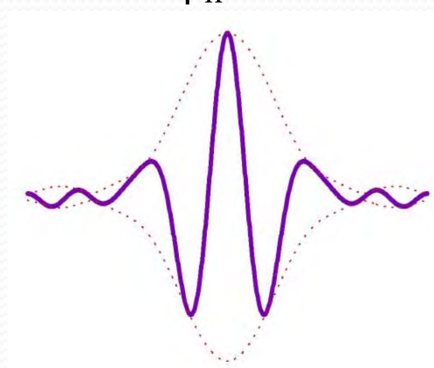
$$E(t) = E_0(t)\cos(\omega_0 t + \underline{\phi})$$

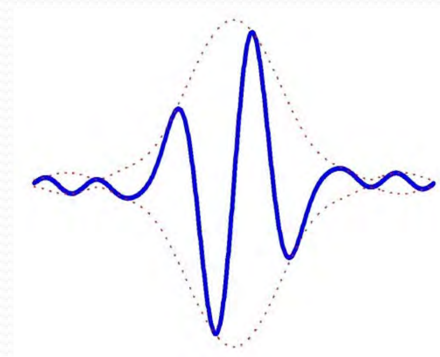


Single cycle waveforms

Inverted cosine	cosine pulse	sine pulse
$\phi_n = \pi$	$\phi_n = 0$	$\phi_n = \pi/2$





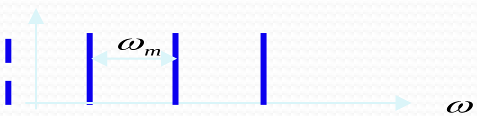


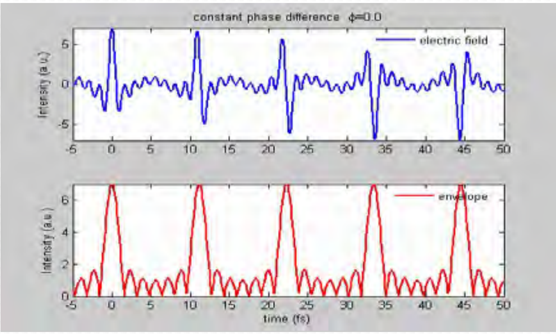
780 nm	⟺	***12,820 cm^{-1}***	⟺	***2.6 fs***
200 nm		***50,000 cm^{-1}***		***684 as***

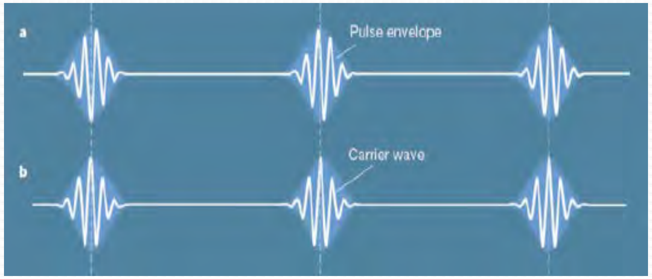
Constant carrier envelope phase

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

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

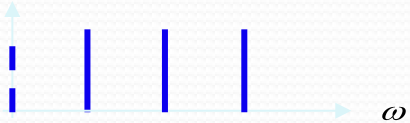


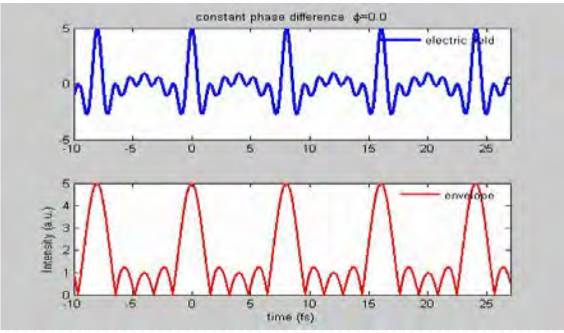




commensurate $\omega_q = n\omega_m \qquad \phi_n = \phi_{CEP} + n\phi_m$

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

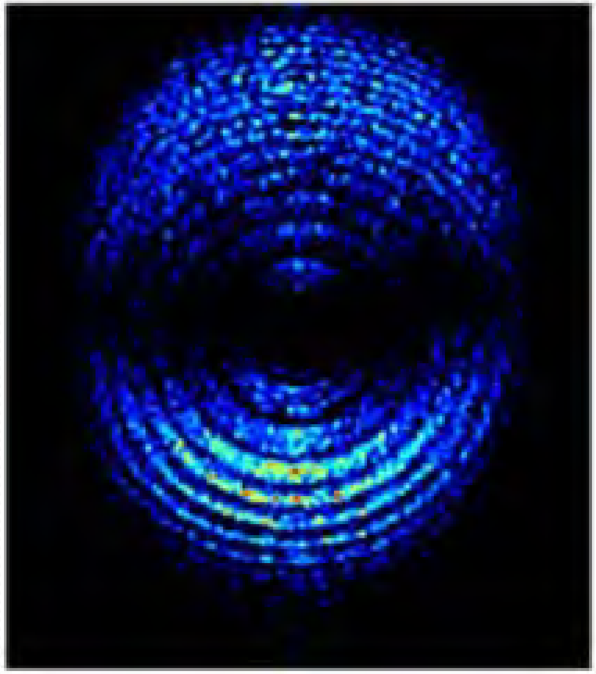




Constant CEP requires that the frequencies are commensurate and the relative phases form an arithmetic series

Attosecond pulse train for quantum stroboscope

Hummingbird wing



Electron velocity mapping

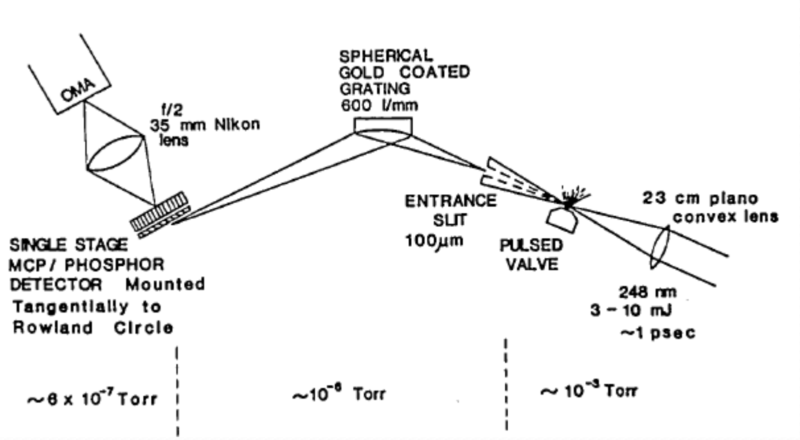
Ingredients of an attosecond single-cycle optical pulse:

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

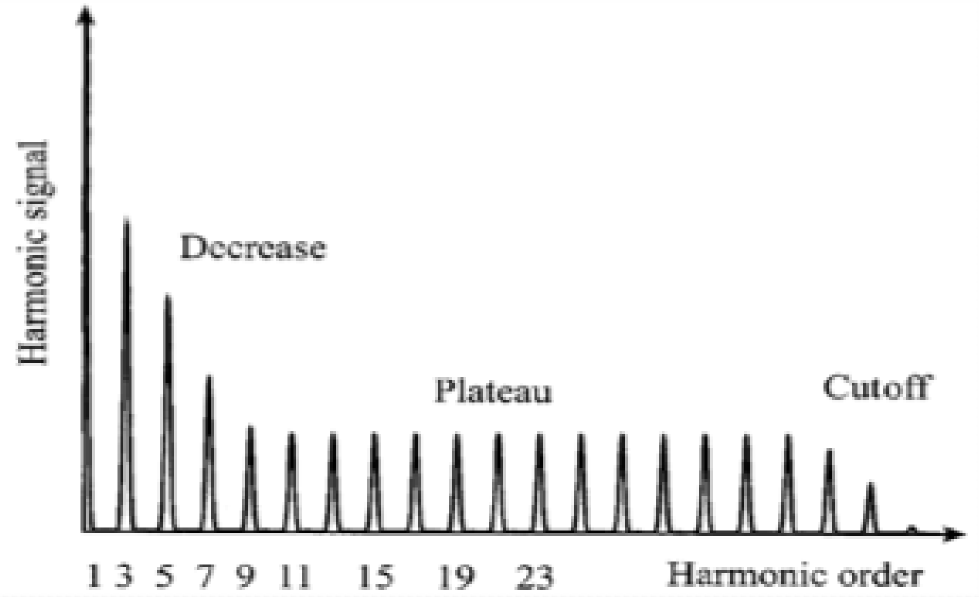
Lightwave control

High Harmonic Generation

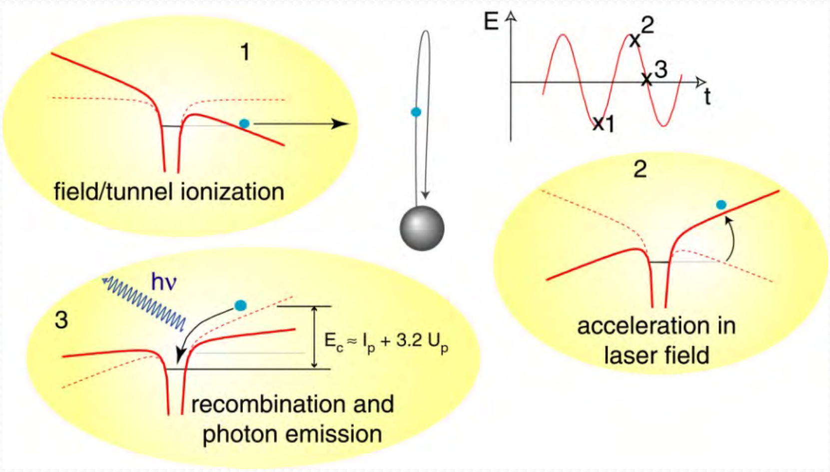
High-order Harmonic Generation has been a regular process to generate attosecond pulse and coherent soft X-ray.



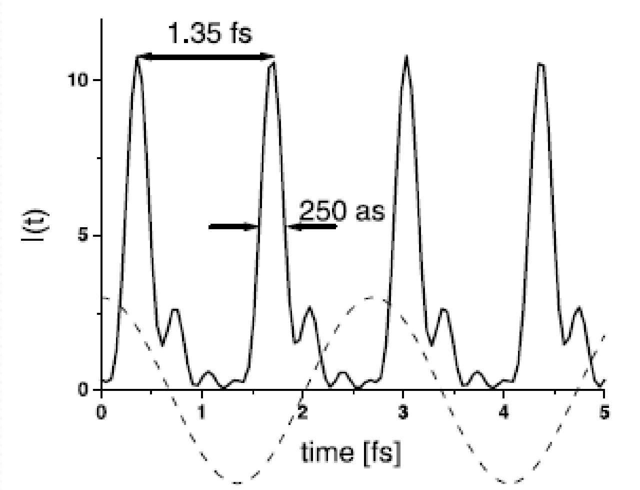
J. Opt. Soc. Am. B 4, 595 (1987)



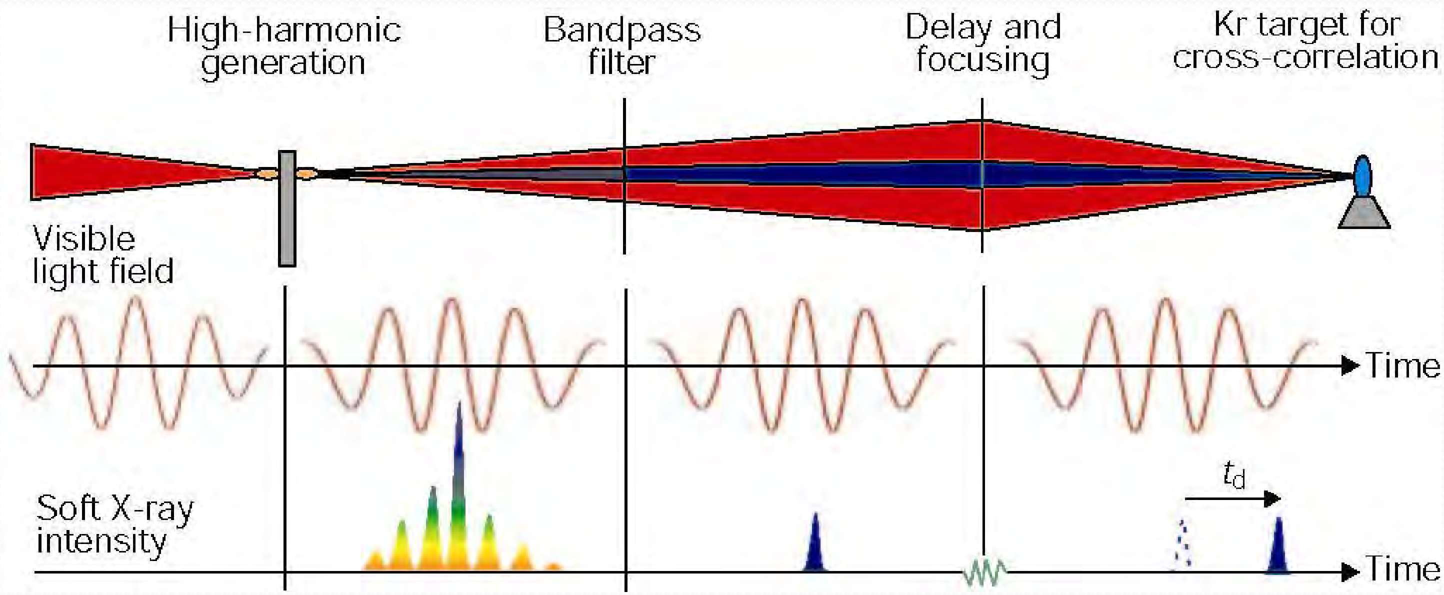
Attosecond Pulse Generation



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



P. M. Paul, *et al.*, Science **292**, 1689 (2001)



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

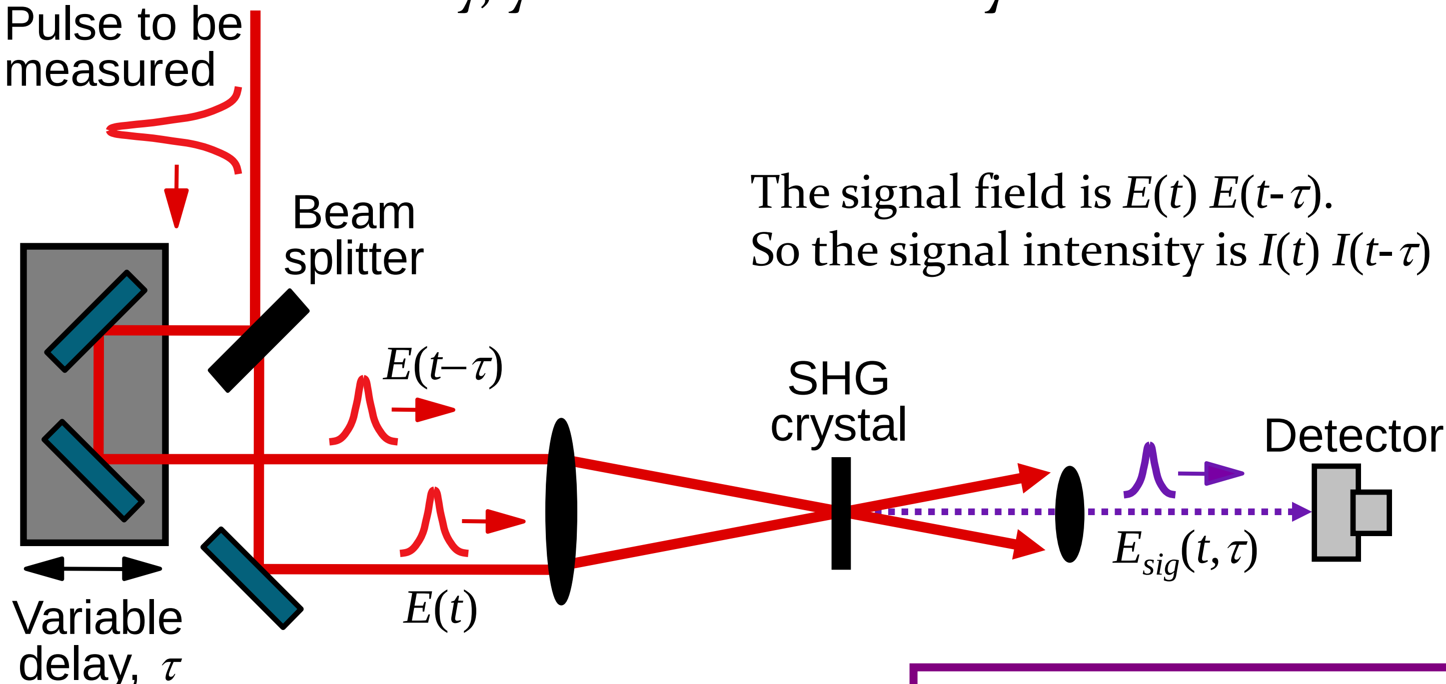
How to measure pulses

- Autocorrelation
- Frequency-Resolved Optical Gating (FROG)
- Spectral Phase Interferometry for Direct Electric-field Reconstruction (SPIDER)
- Reconstruction of Attosecond Beating by Interference of Two-Photon Transition (RABITT)
- Complete Reconstruction of Attosecond Bursts (CRAB)

Pulse Measurement in the Time Domain:

The Intensity Autocorrelator

Crossing beams in a nonlinear-optical crystal, varying the delay between them, and measuring the signal pulse energy vs. delay, yields the **Intensity Autocorrelation, $A^{(2)}(\tau)$**.



The signal field is $E(t)\,E(t-\tau)$.
So the signal intensity is $I(t)\,I(t-\tau)$

The Intensity Autocorrelation:

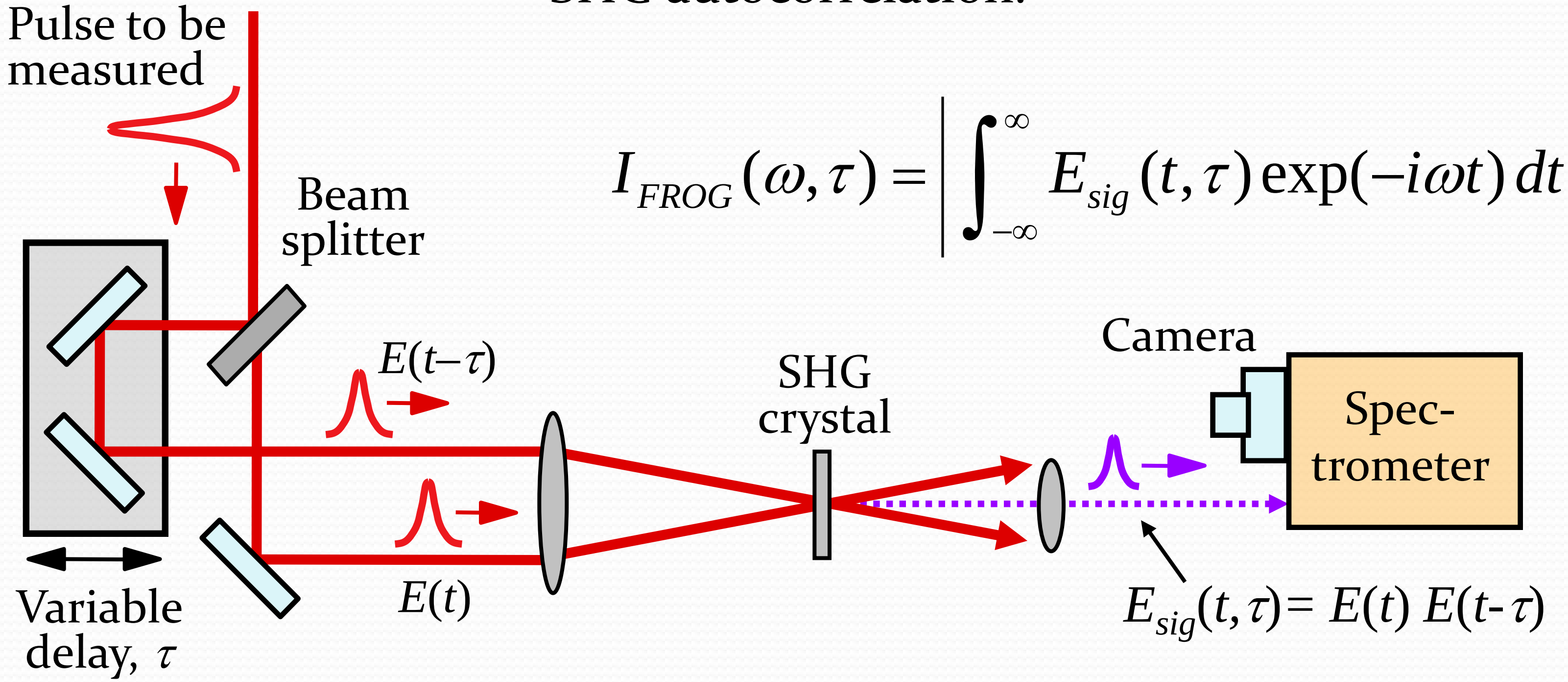
$$A^{(2)}(\tau) \equiv \int_{-\infty}^{\infty} I(t)\,I(t-\tau)\,dt$$

R. Trebino's lecture

Second-harmonic-generation FROG

SHG FROG is simply a spectrally resolved SHG autocorrelation.

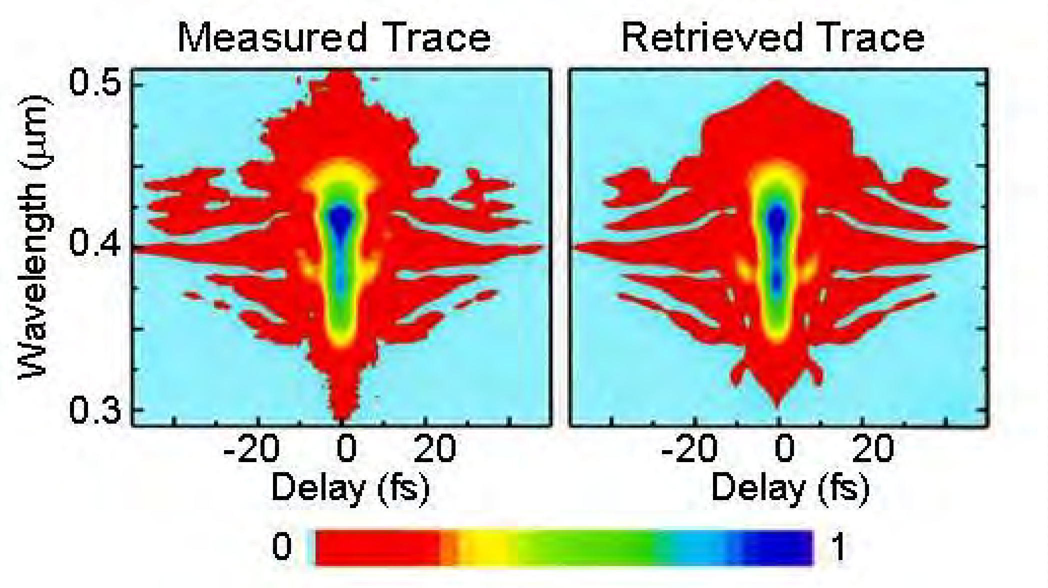
$$I_{FROG}(\omega,\tau) = \left| \int_{-\infty}^{\infty} E_{sig}(t,\tau)\exp(-i\omega t)\,dt \right|^2$$



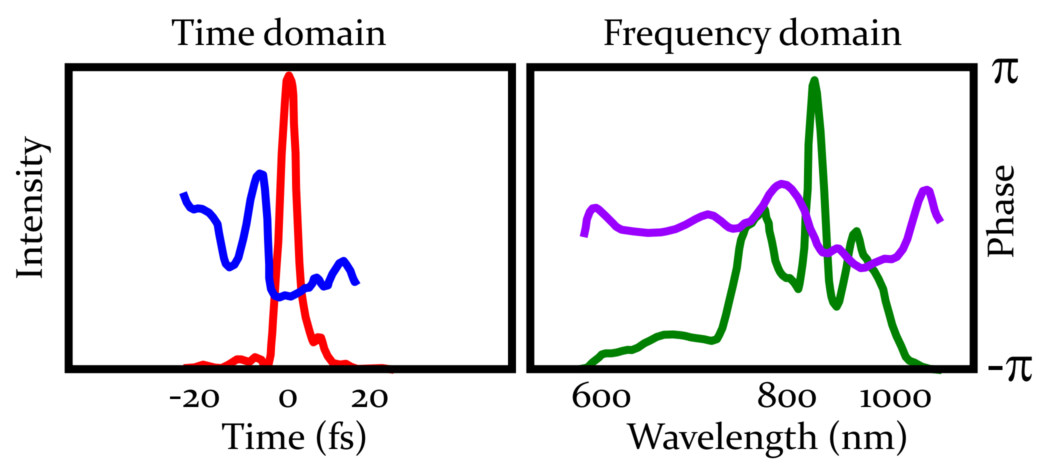
SHG FROG is the most sensitive version of FROG.

R. Trebino's lecture

SHG FROG measurements of a 4.5-fs pulse!



Agreement between the experimental and reconstructed FROG traces provides a nice check on the measurement.



Baltuska, Pshenichnikov, and Weirsma, J. Quant. Electron., 35, 459 (1999).

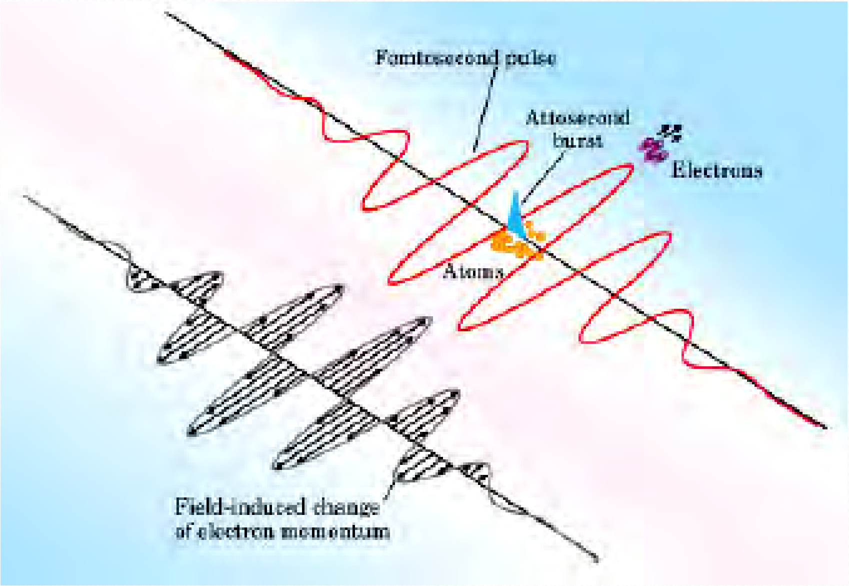
R. Trebino's lecture

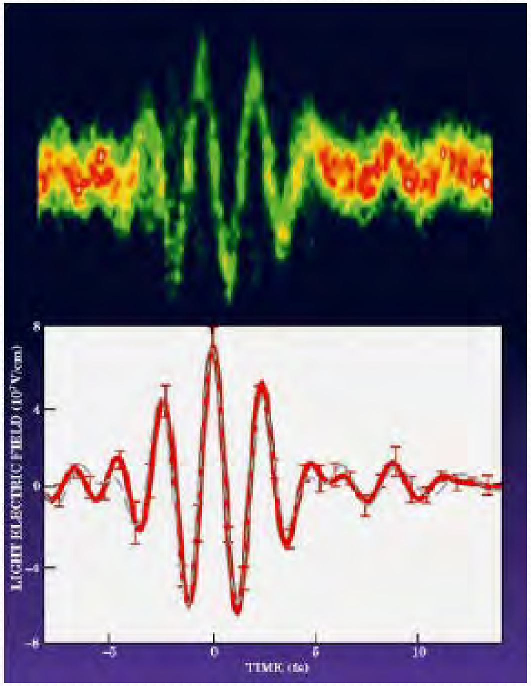
PHYSICS TODAY October 2004

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.

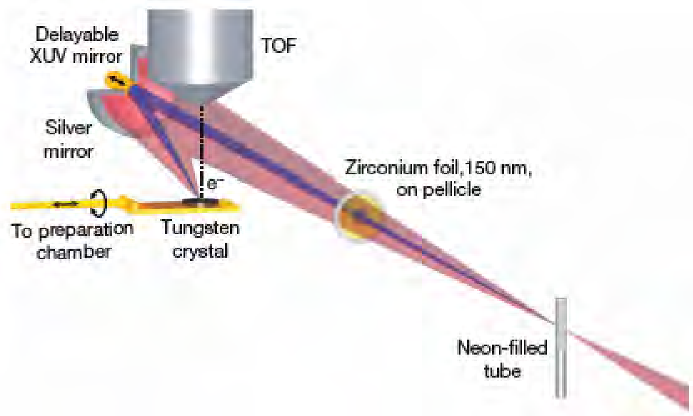


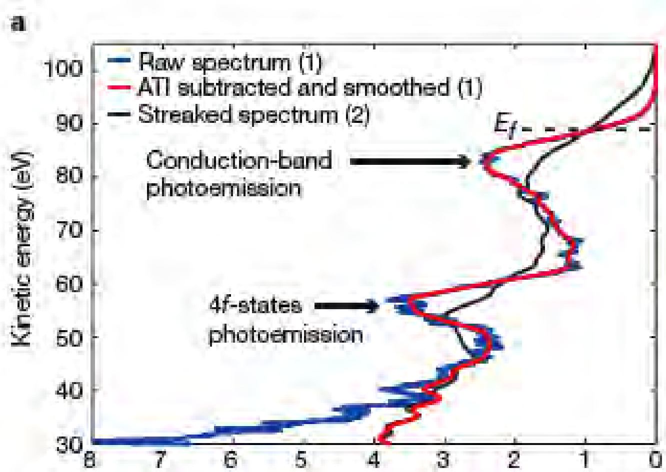


A. Baltuska et al., Nature **421**, 611 (2003)
E. Goulielmakis et al., Science **305**, 1267 (2004)

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]





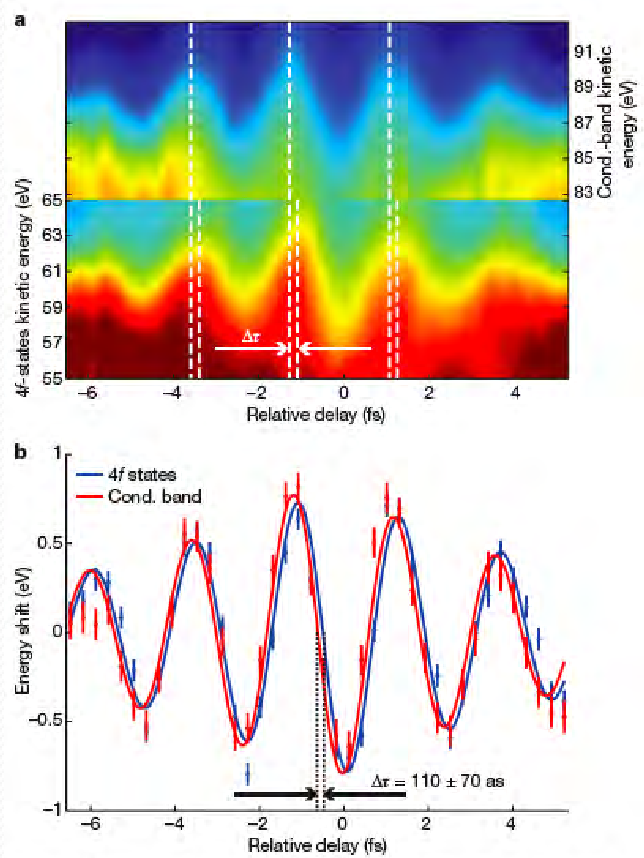
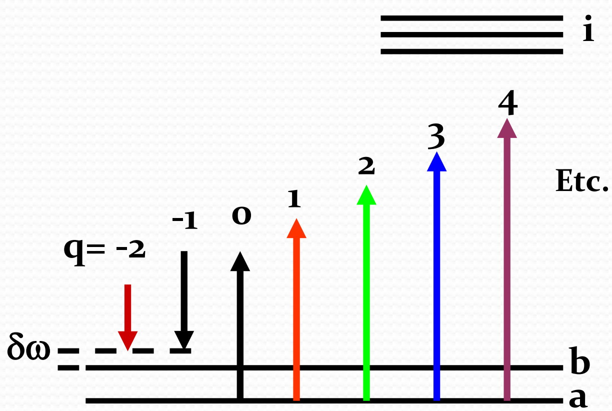


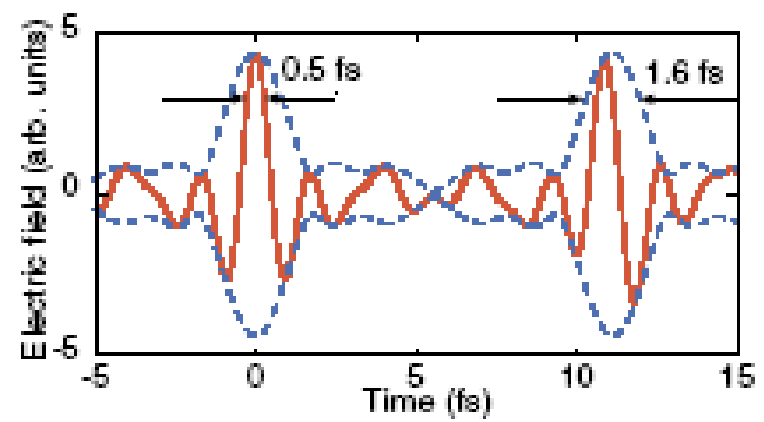
Figure 3 | Evidence of delayed photoemission. a, The 4*f* and conduction-band spectrograms, following cubic-spline interpolation of the measured data

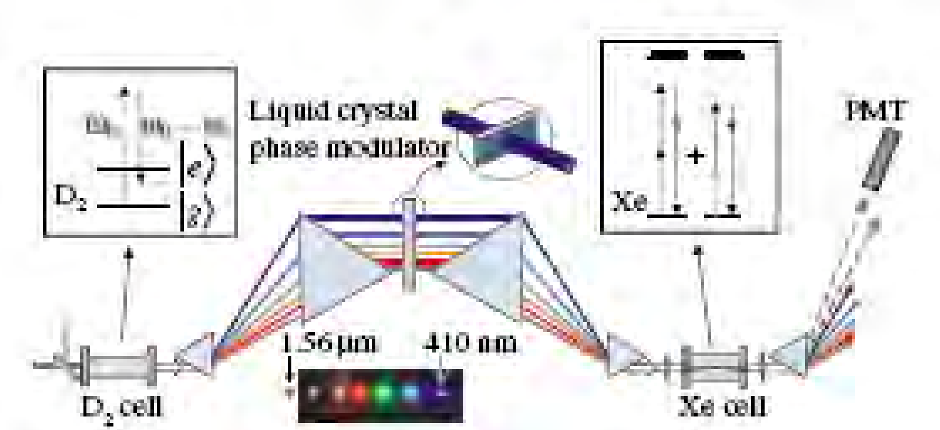
Nature 449, 1029 (2007)

Methods of generating attosecond pulses

High-order stimulated Raman scattering using molecular modulation





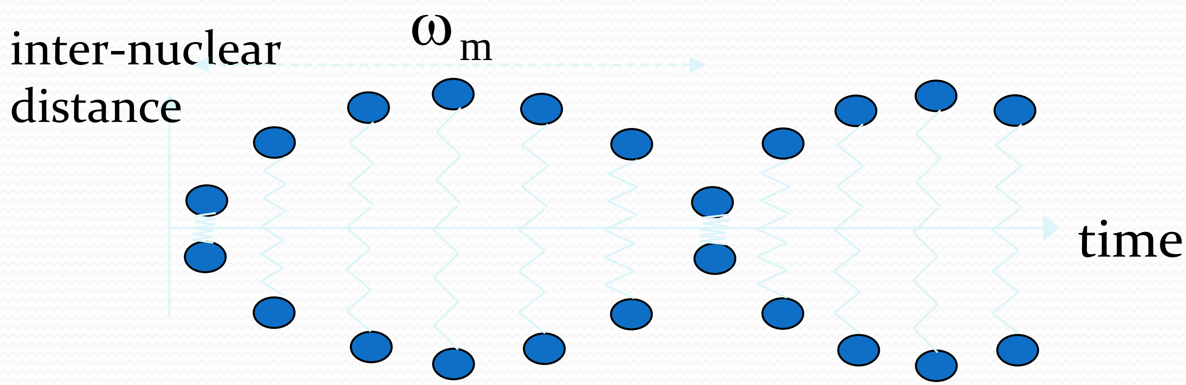


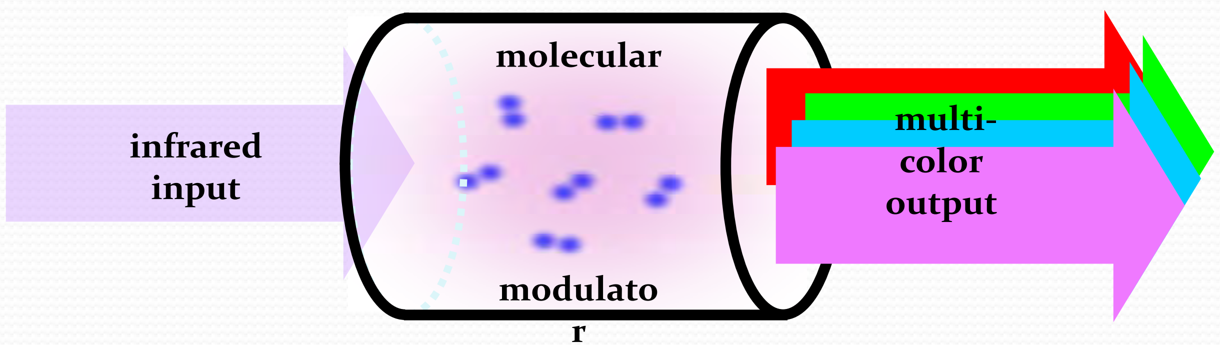
Advantages: IR-UV region
good power
single-cycle
Disadvantages: complex setup
8-50 fs pulse spacing
limited to ~ 300-500 as

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

Molecular Modulation

Molecular modulation is analogous to electro-optic modulation



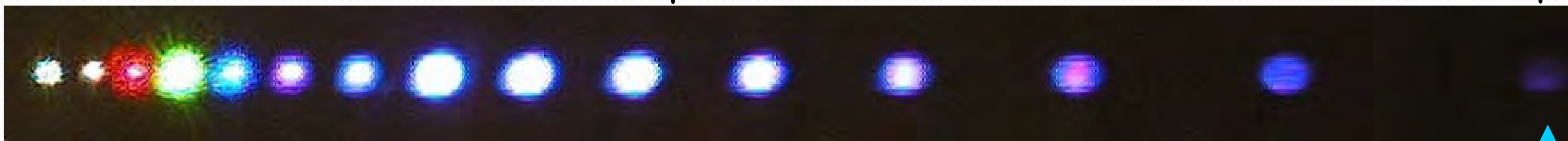


Refractive Index $n = n_0 + \delta \cos \omega_m t$

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

Alexei Sokolov
Steve Harris

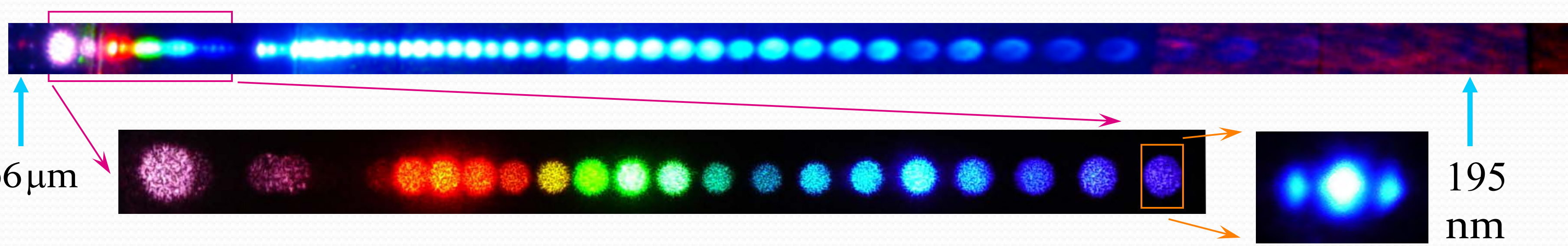
D_2 Vibration Spectra: 16 sidebands, spaced by 2994 cm^{-1}



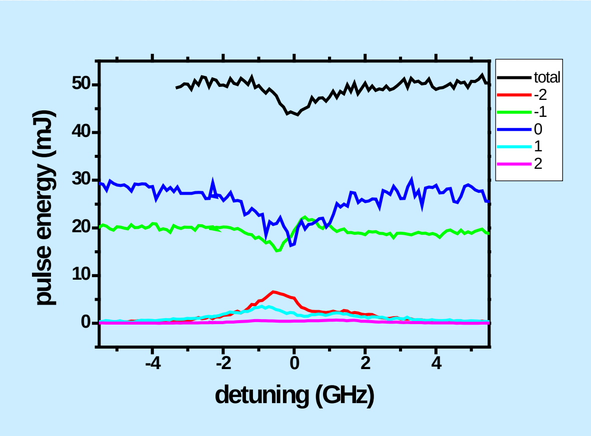
H_2 Rotation Spectra: 29 sidebands, spaced by 587 cm^{-1}

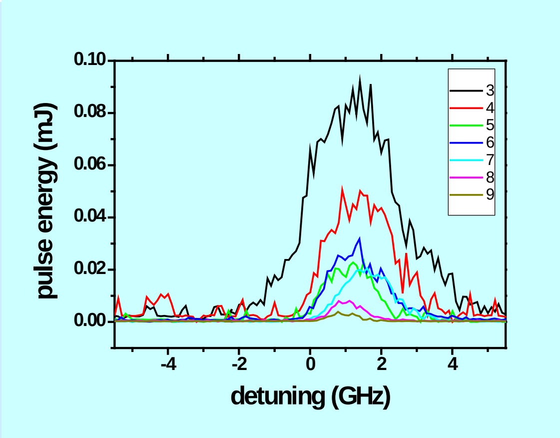
Phys. Rev. A (R)(1997)
Phys. Rev. Lett. 81 (1998)
Opt. Lett. 24 (1999)
Phys. Rev. Lett. 84 (2000)
Phys. Rev. Lett. 85 (2000)
Phys. Rev. A 63 (2001)
Phys. Rev. Lett. 91 (2003)
Phys. Rev. Lett. 93 (2005)

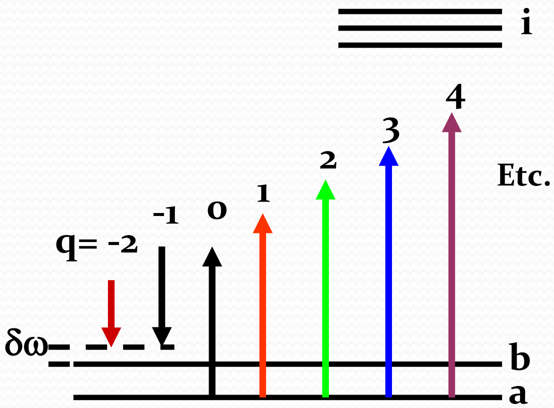
Multiplicative Spectra: ~ 200 sidebands, spaced by < 587 cm^{-1}

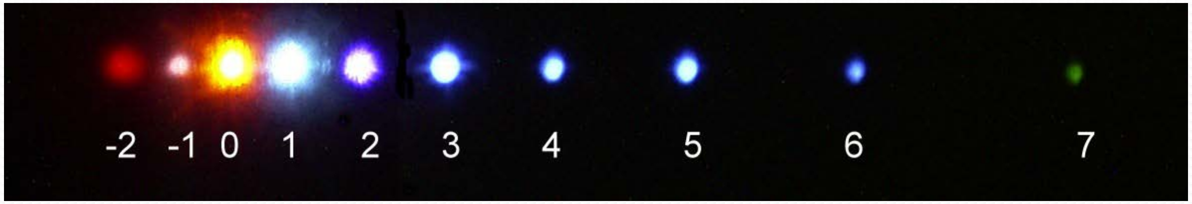


Raman sidebands generated



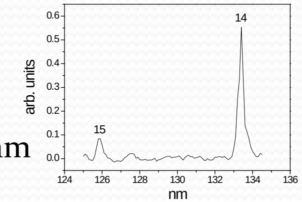






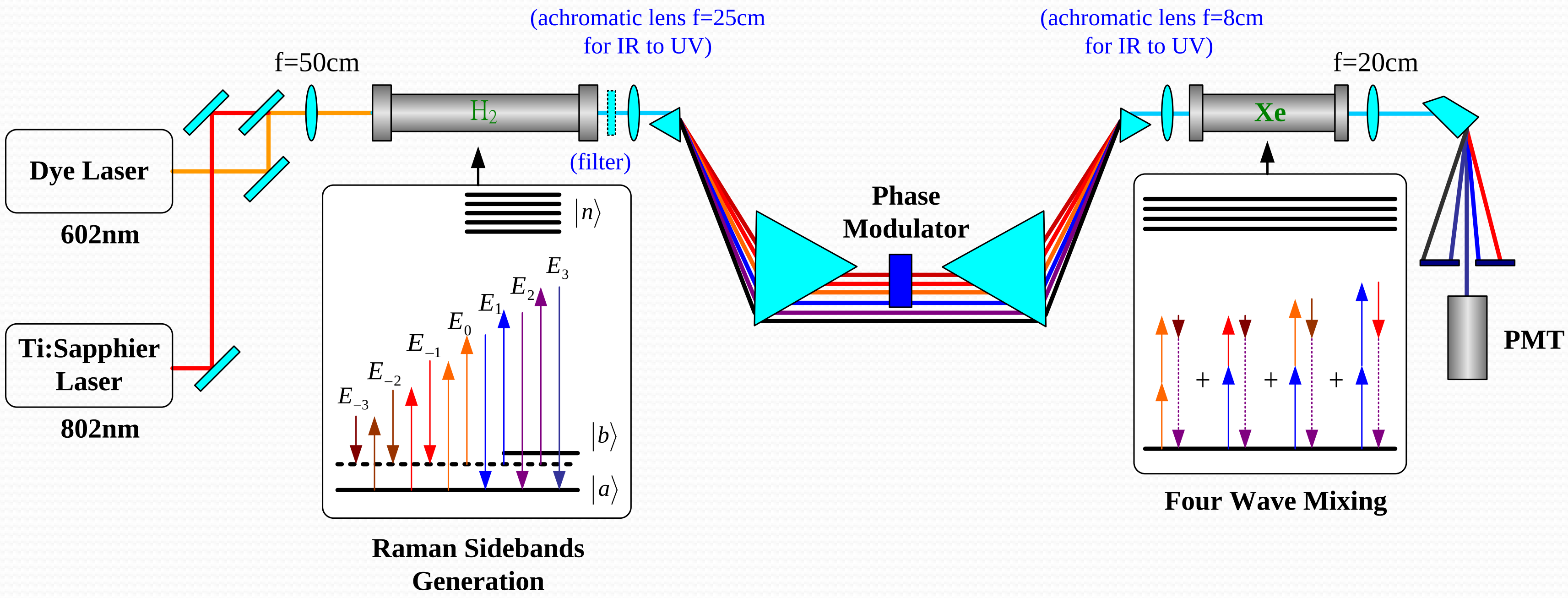
$q = 3, \lambda =$ 339.6 nm
6 238.6 nm
9 183.9 nm
12 149.6 nm
14 133.0 nm

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



15th order at 126 nm observed

Experiment Setup

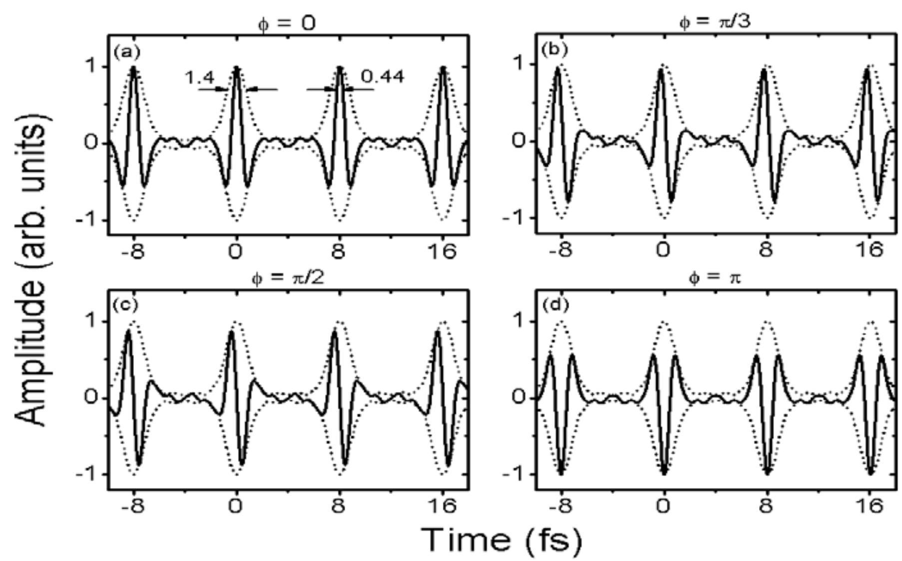


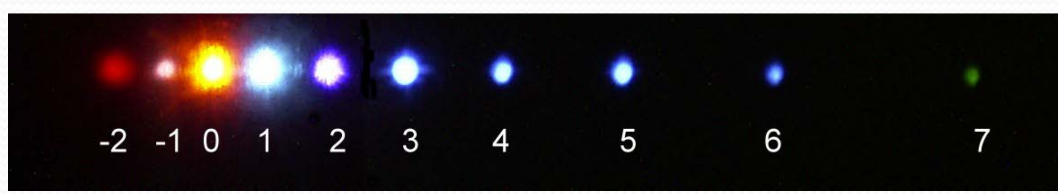
M. Y. Shverdin et al. PRL **94**, 033904 (2005)

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

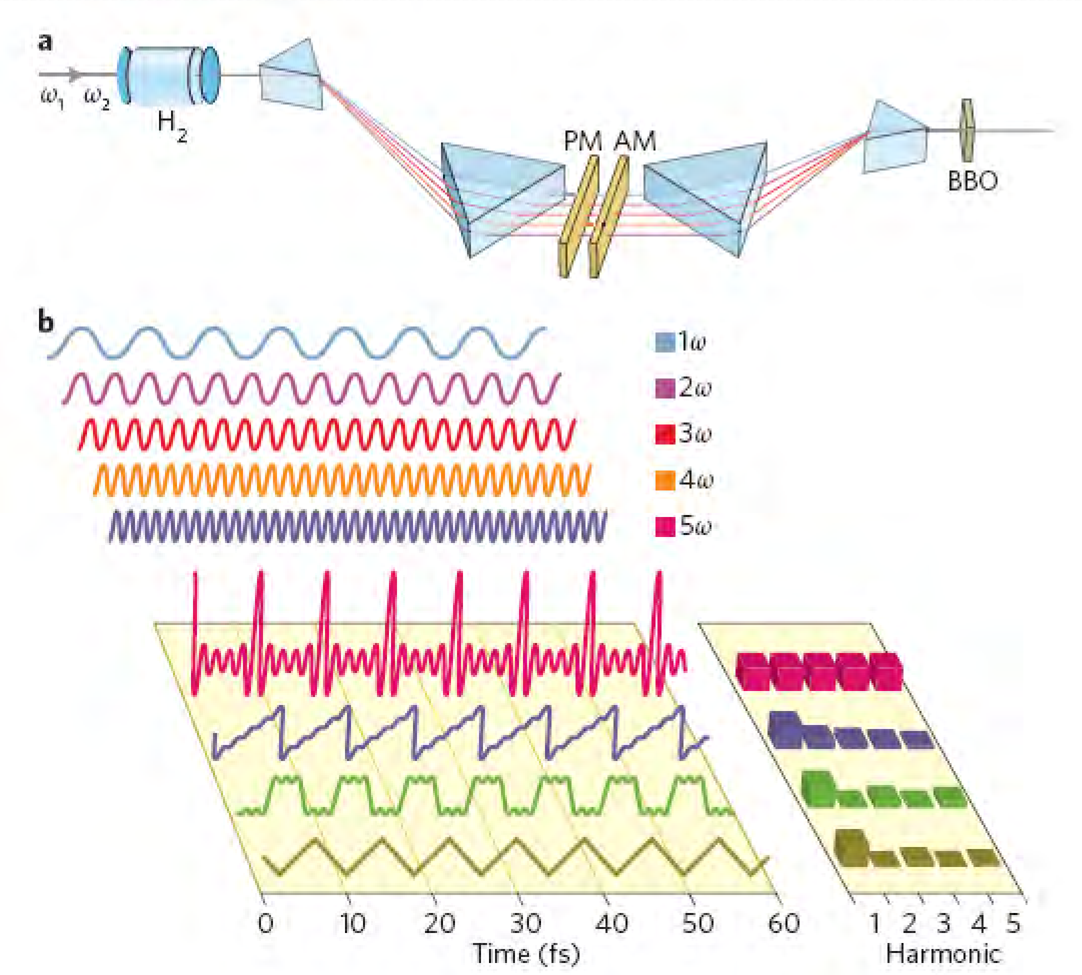


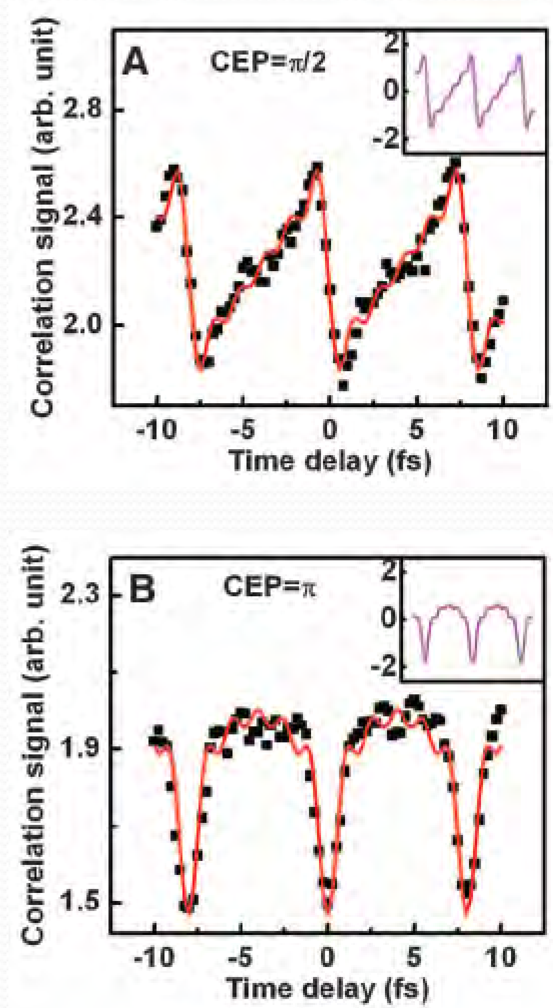


Total spectral span >70,000 cm^{-1}

Phys. Rev. Lett. 100, 163906 (2008)
Phys. Rev. Lett. 102, 213902 (2009)

Synthesis and measurement of ultrafast waveform

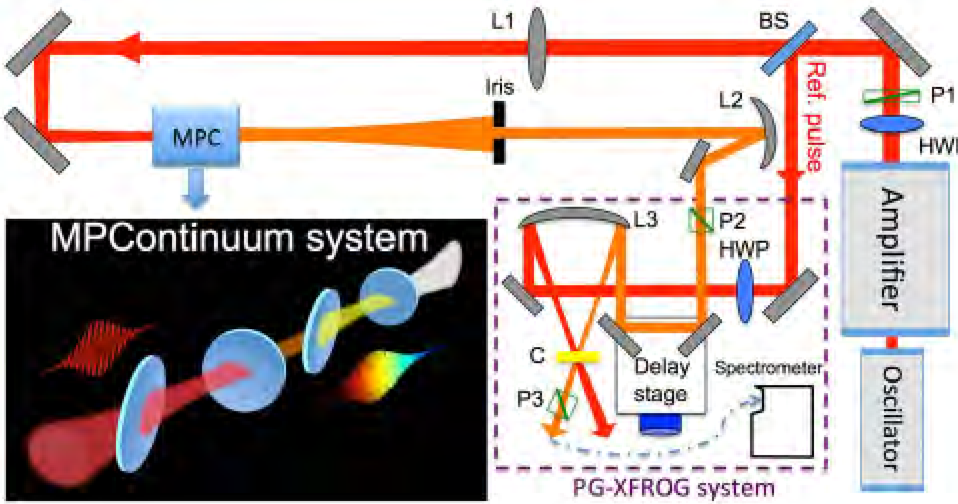


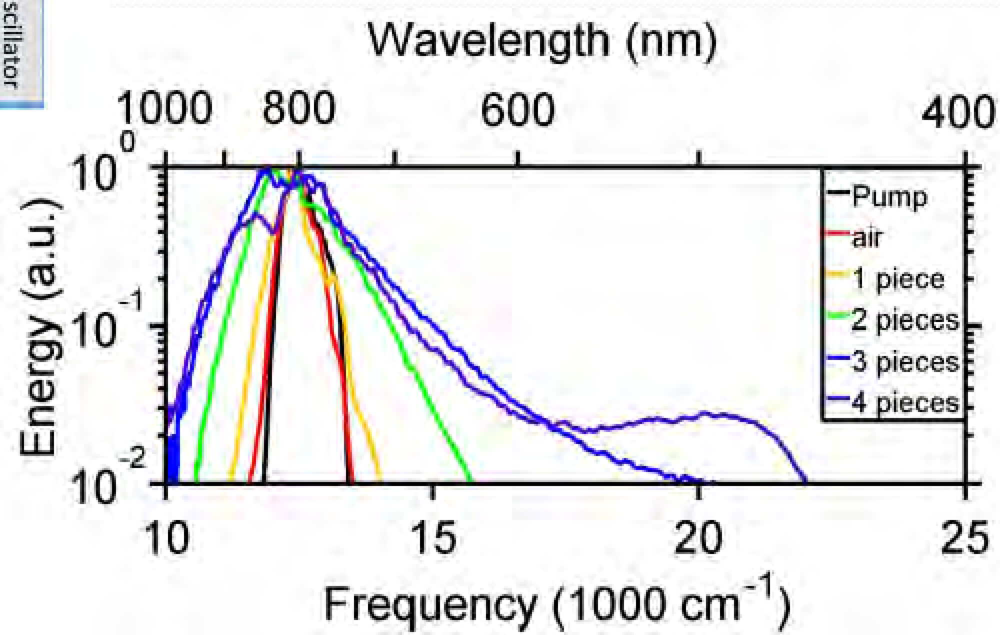


Han-Sung Chan, et al.,
Science 331, 1165 (2011)

Generation of intense supercontinuum in condensed media

Chih-Hsuan Lu,[1] Yu-Jung Tsou,[1] Hong-Yu Chen,[1] Bo-Han Chen,[1] Yu-Chen Cheng,[2] Shang-Da Yang,[1] Ming-Chang Chen,[1] Chia-Chen Hsu,[3] and A. H. Kung[1,2,*]

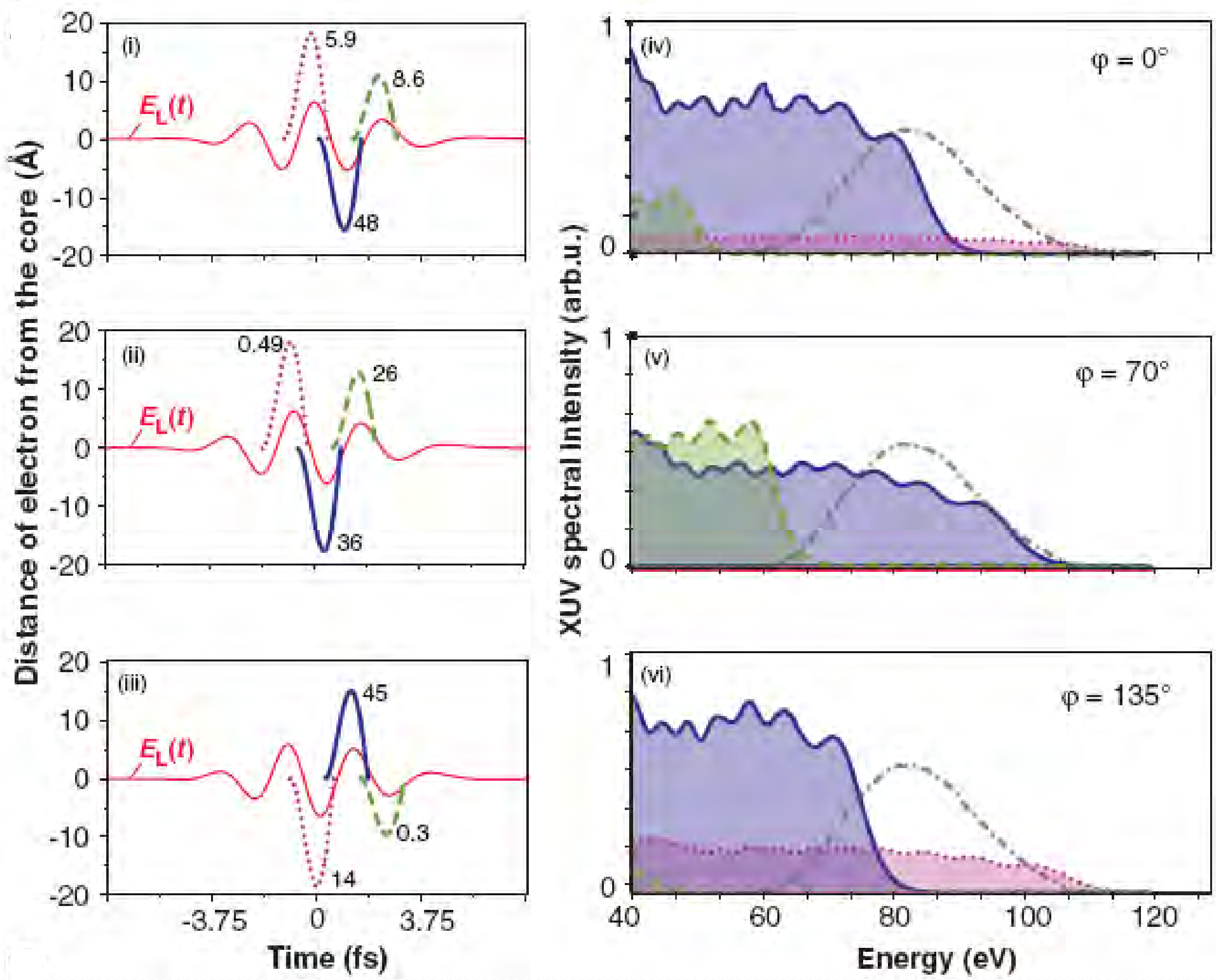




Optica 1, 400 (2014)

Single-cycle Nonlinear Optics

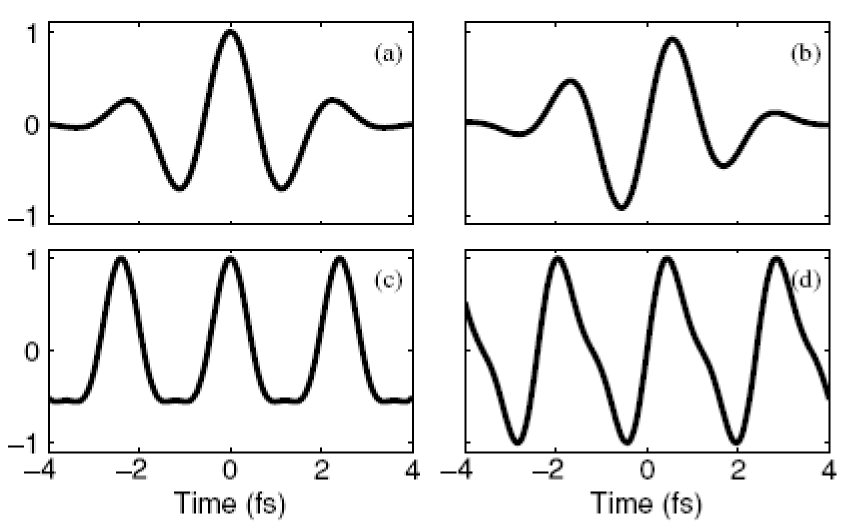
Simulation of sub-femtosecond XUV emission from neon atoms ionized by a linearly polarized, sub-1.5-cycle, 720 nm laser field.

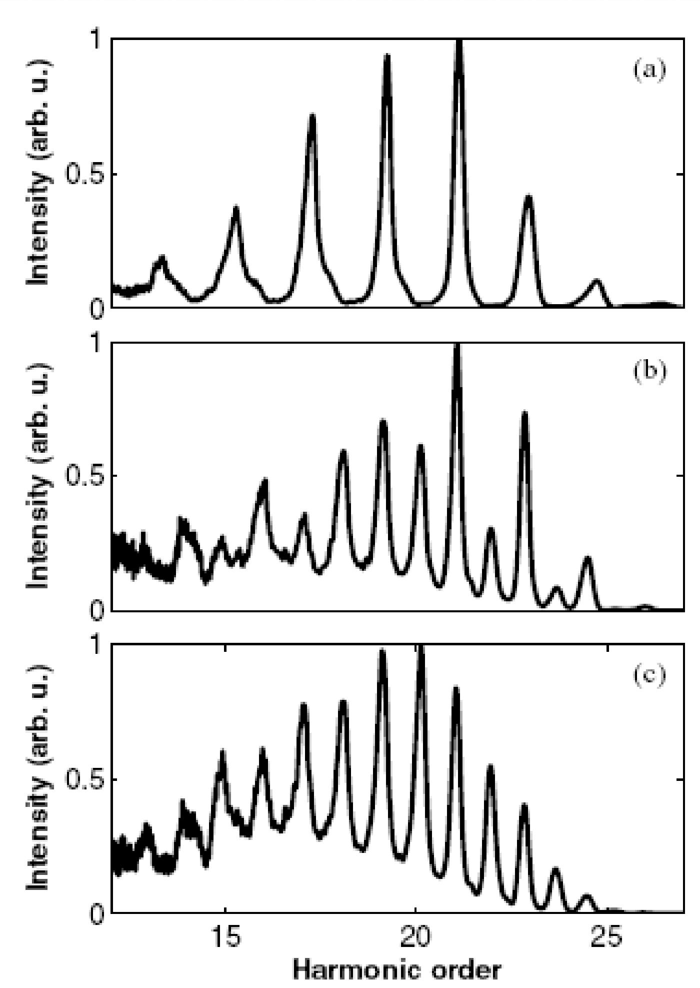


E. Goulielmakis, et al., Science 320, 1614 (2008)

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)

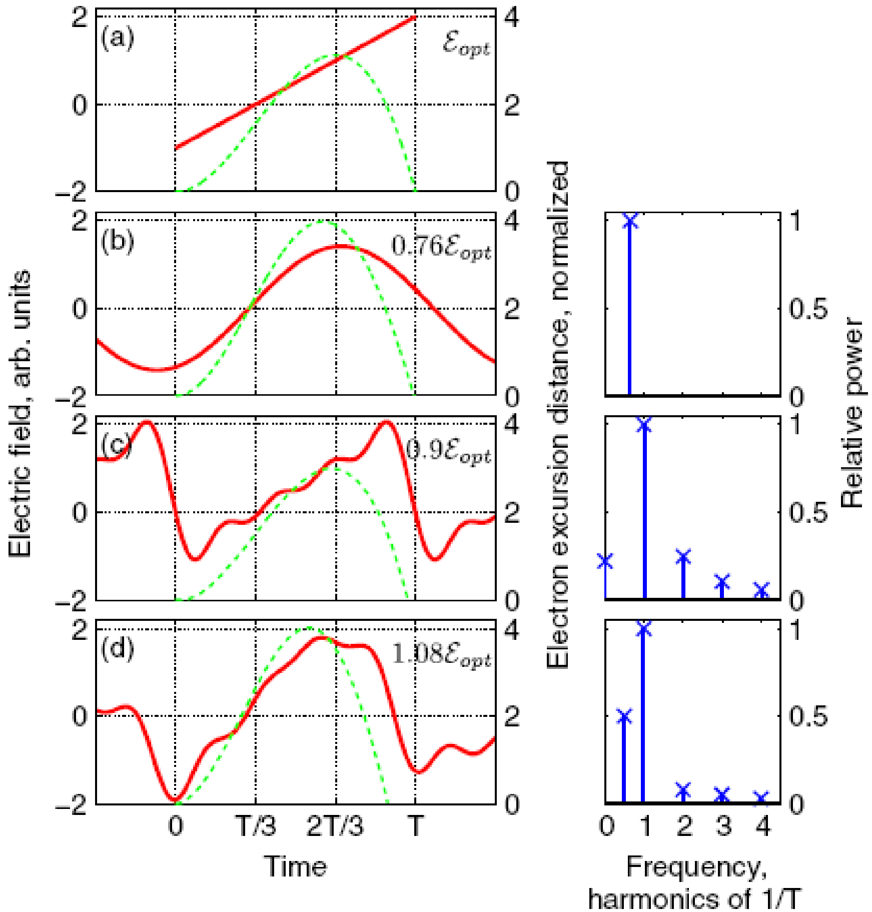
PRL 102, 063003 (2009) PHYSICAL REVIEW LETTERS week ending 13 FEBRUARY 2009

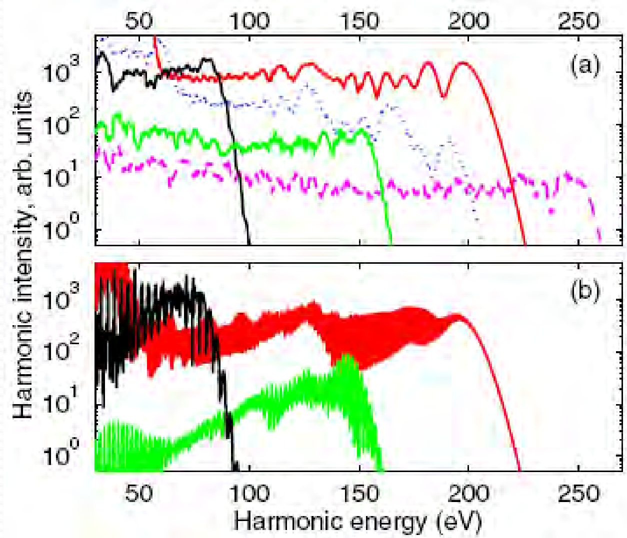
Ideal Waveform to Generate the Maximum Possible Electron Recollision Energy for Any Given Oscillation Period

L. E. Chipperfield,* J. S. Robinson, J. W. G. Tisch, and J. P. Marangos

Imperial College London, London SW7 2BW

(Received 25 April 2008; published 12 February 2009)

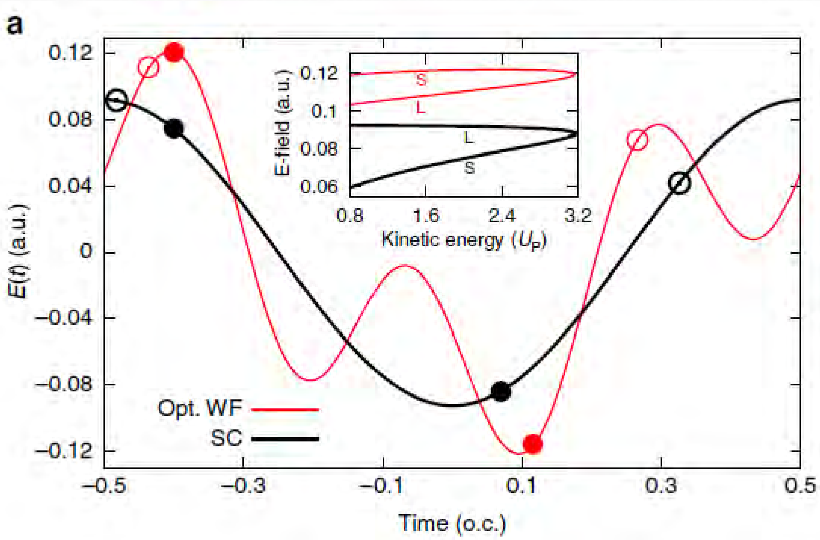


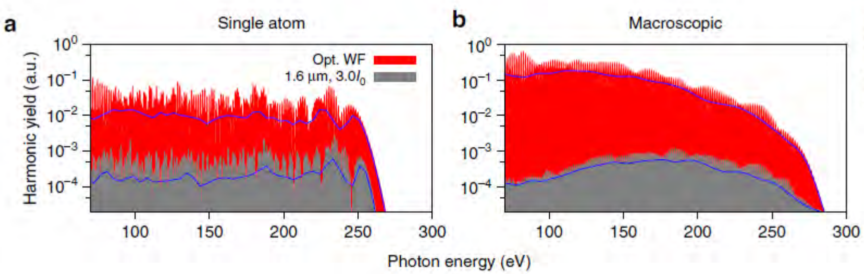


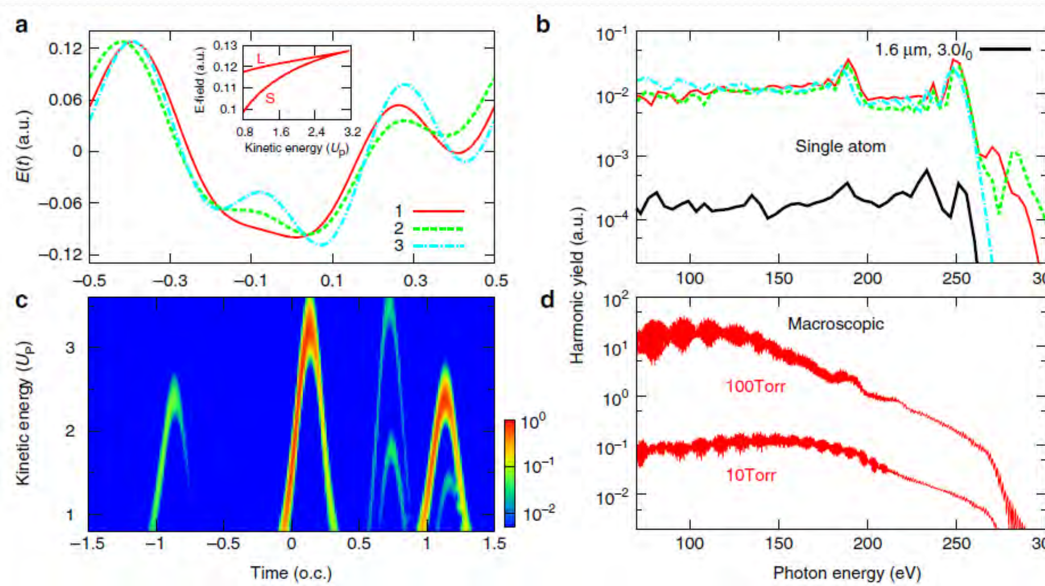
L. E. Chipperfield, et al.,
Phys. Rev. Lett. 102, 063003 (2009)

Waveforms for optimal sub-keV high-order harmonics with synthesized two- or three-color laser fields

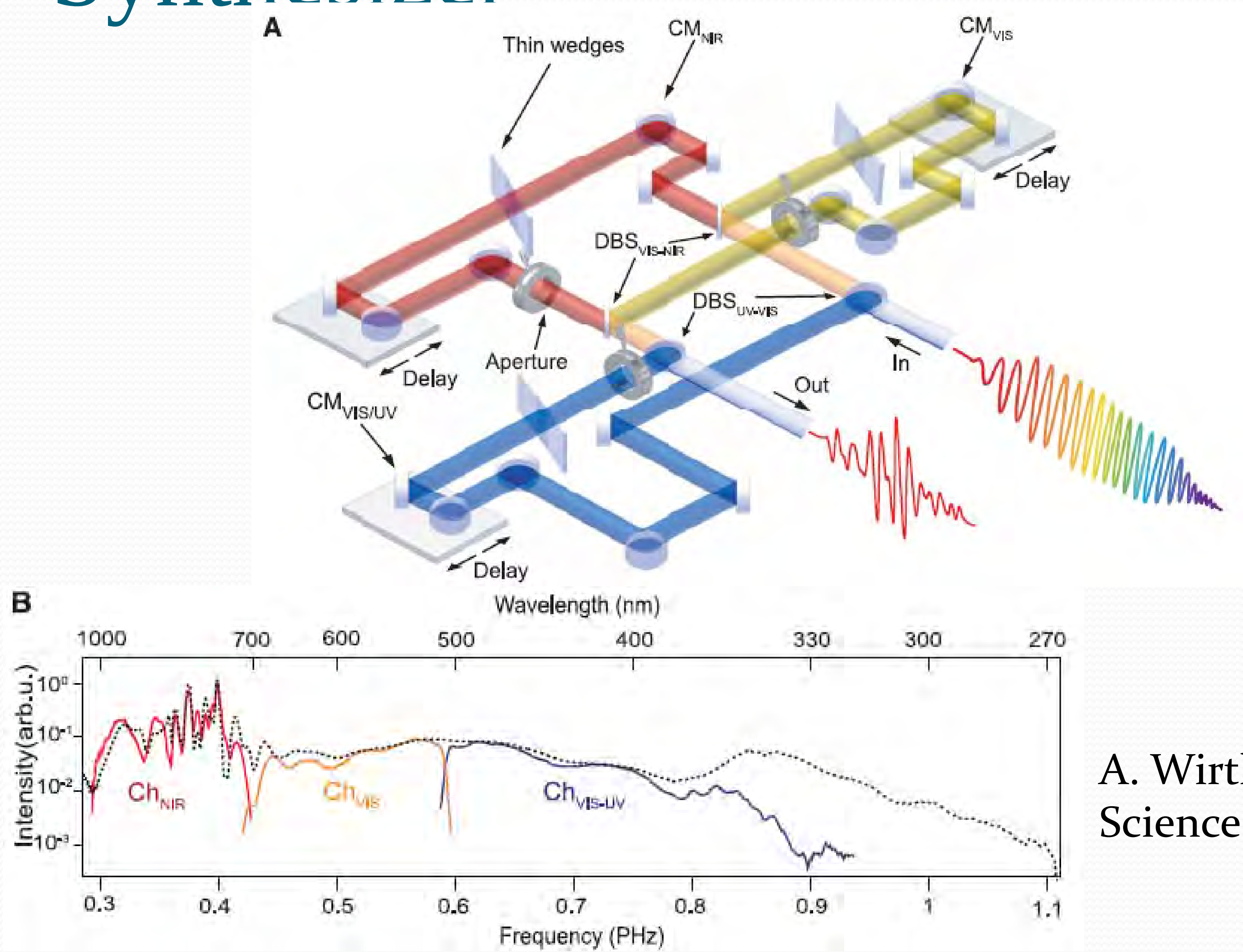
Cheng Jin, et al.,
Nat. Commun. 5, 4003 (2014)







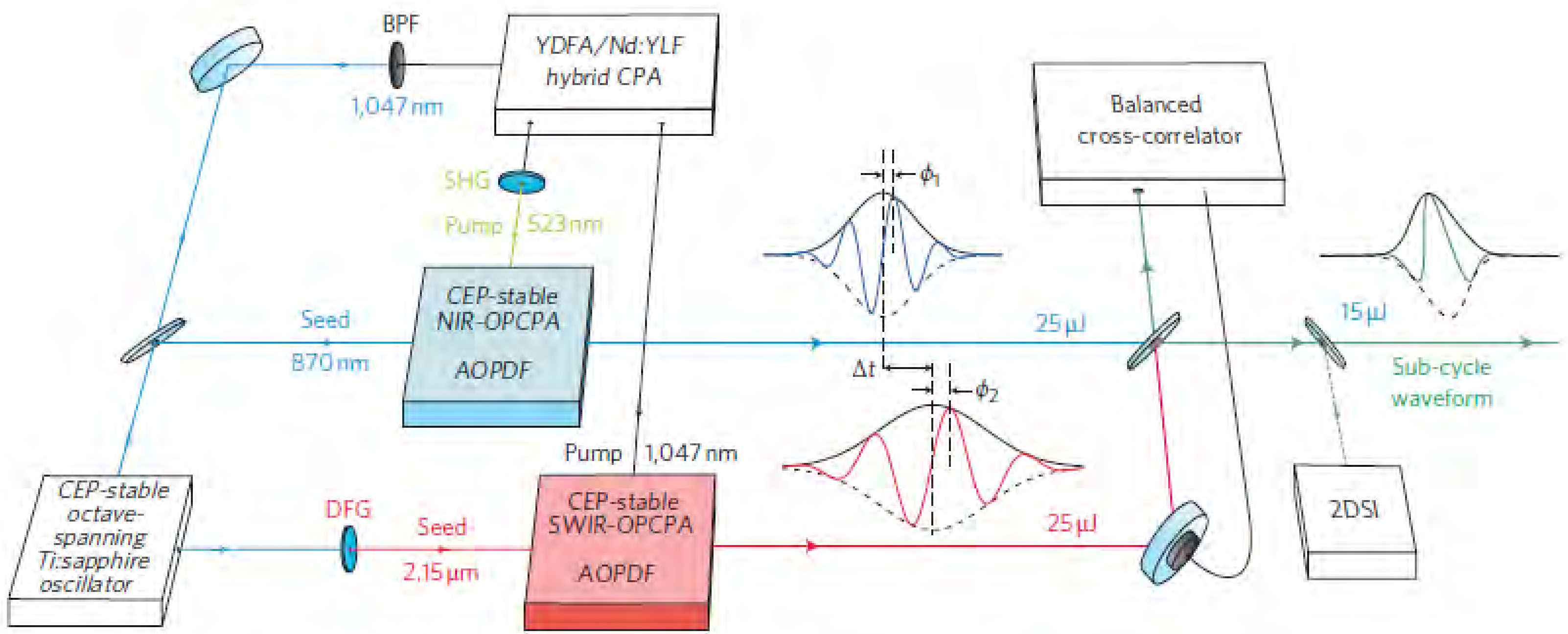
Three-channel Optical Field Synthesizer



A. Wirth et al.
Science 334, 195 (2011)

High-energy pulse synthesis with sub-cycle waveform control for strong-field physics

Shu-Wei Huang[1], Giovanni Cirmi[1], Jeffrey Moses[1], Kyung-Han Hong[1], Siddharth Bhardwaj[1], Jonathan R. Birge[1], Li-Jin Chen[1], Enbang Li[2], Benjamin J. Eggleton[2], Giulio Cerullo[3] and Franz X. Kärtner[1,4]*



Nat. Photon. 5, 475 (2011)

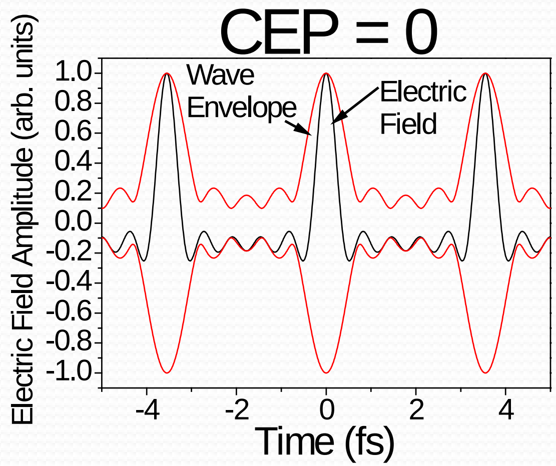
Multi-color laser field

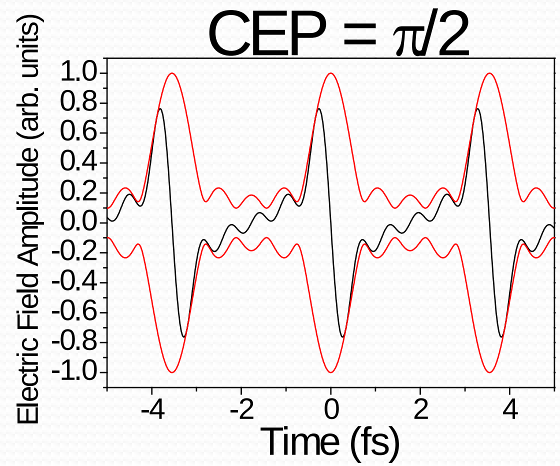
- Broadband source: larger than 2 octaves
  coherent and commensurate
- High peak power enough
  10^{13}-10^{14} W/cm^2
- Simple experiment setup
- Light waveform controllable

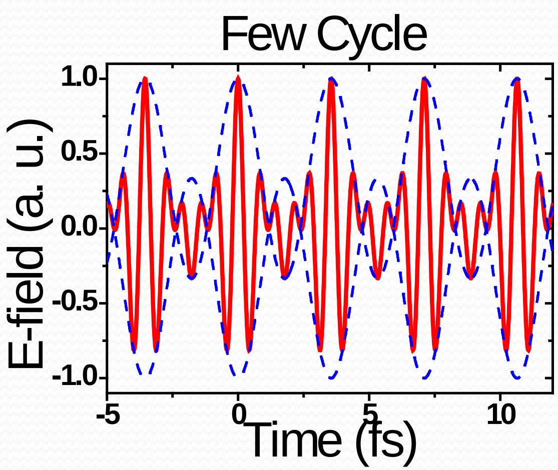
Waveform by Harmonics

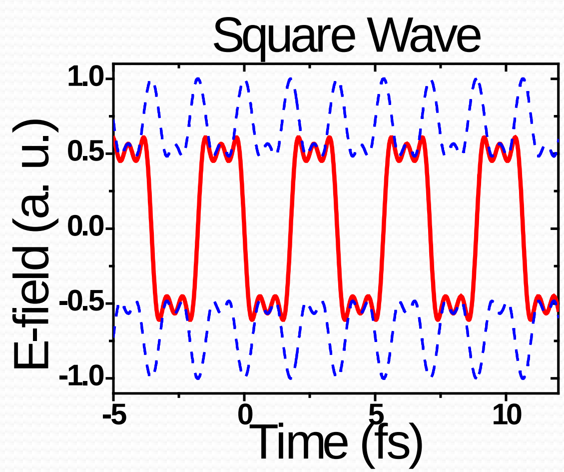
1064
532
355
266
213

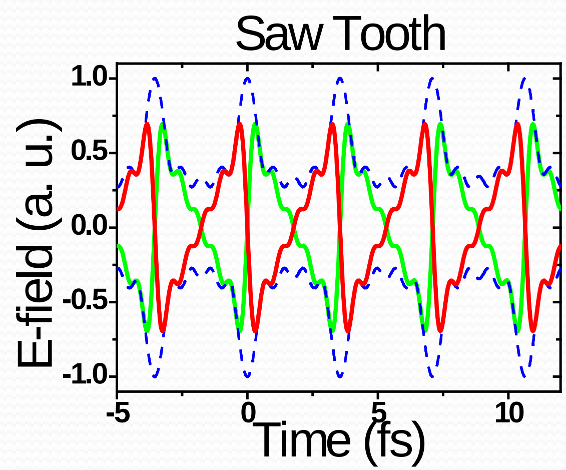
~37,600cm^{-1}



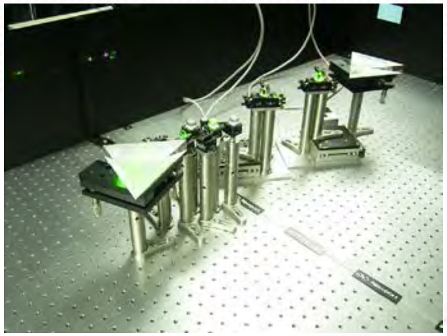


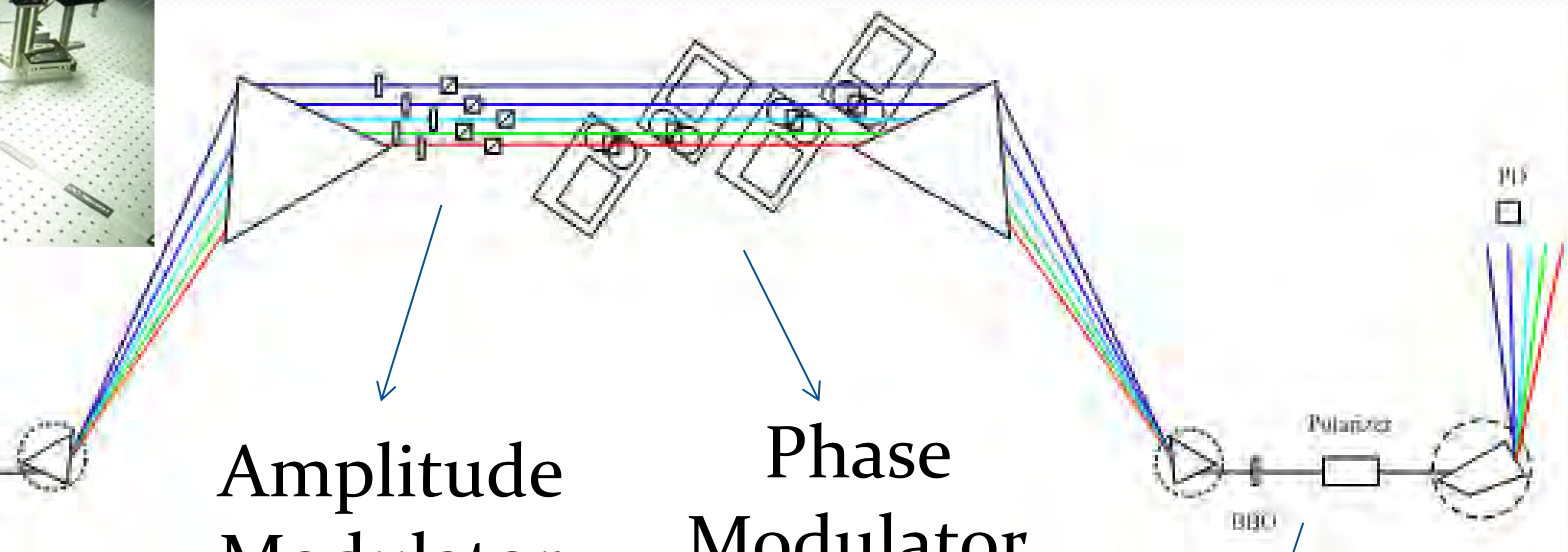






Experimental Setup

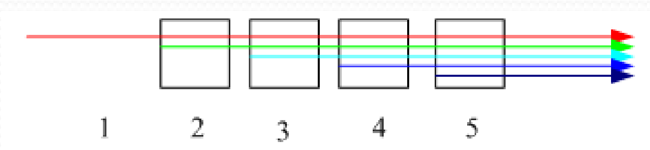




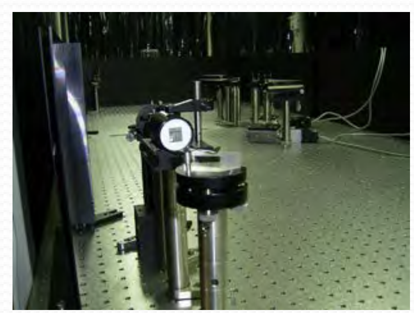
Amplitude Modulator

Phase Modulator

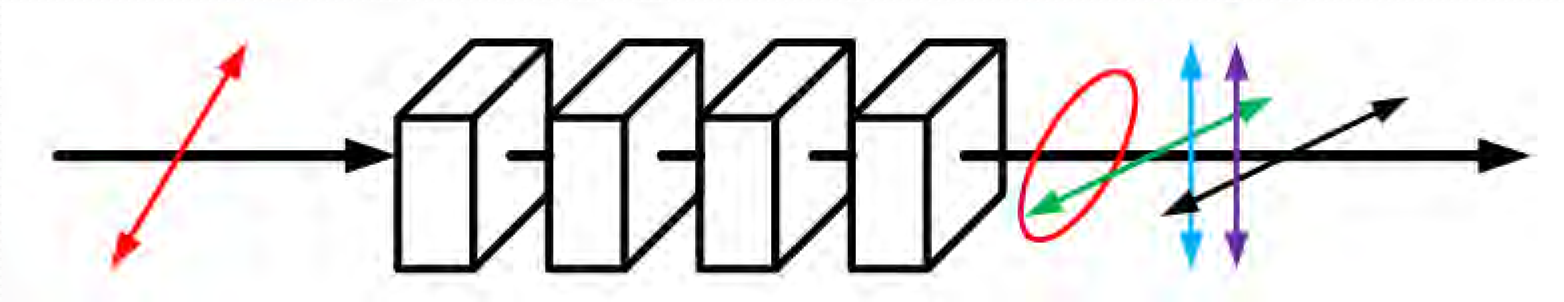
Relative Phase Measurement

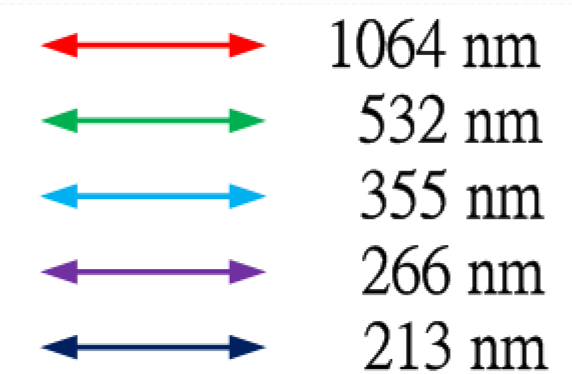


Harmonics generation (collinearly)



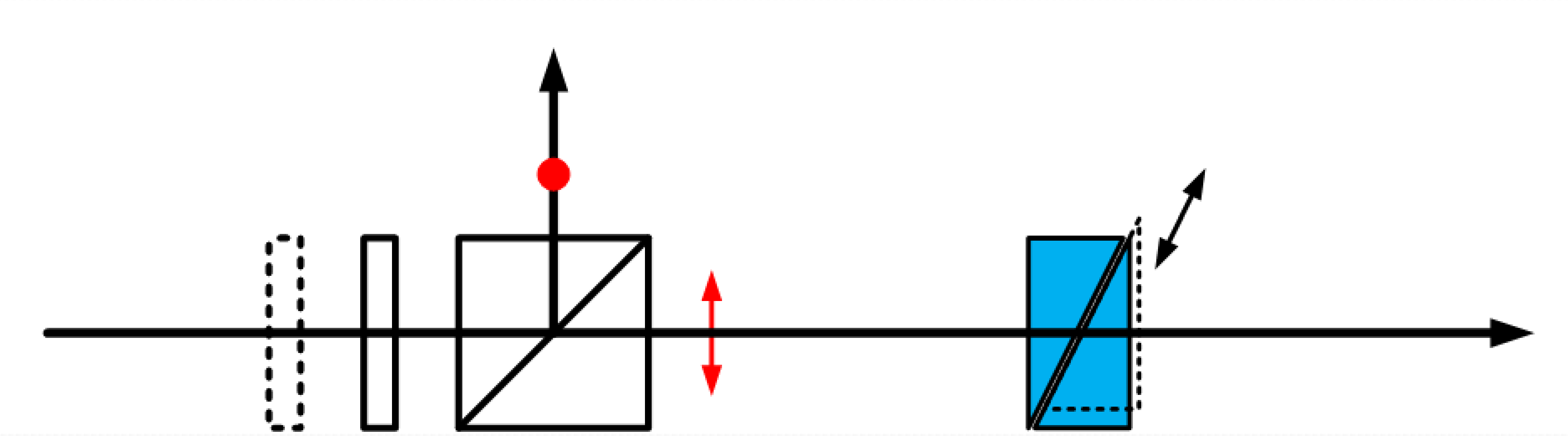
Harmonic Generation





		1064:	380 mJ
KD*P (II)	1064+1064 →	532:	178 mJ
KD*P (I)	1064+532 →	355:	70 mJ
BBO (I)	532+532 →	266:	41 mJ
BBO (I)	1064+266 →	213:	22 mJ

Amplitude and Phase modulator



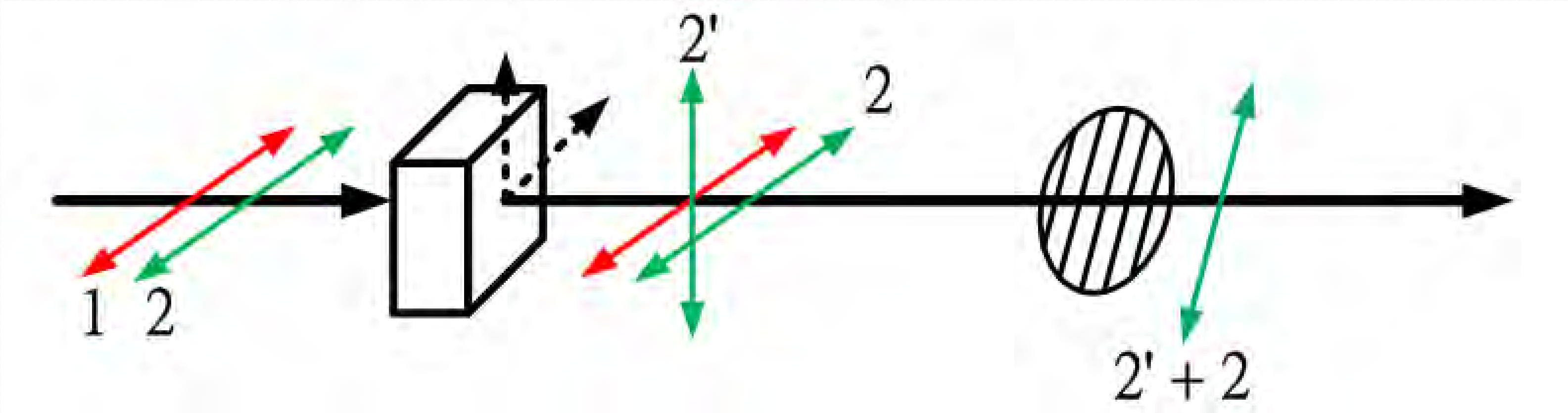
λ/4 λ/2 PBC

Prism Pair

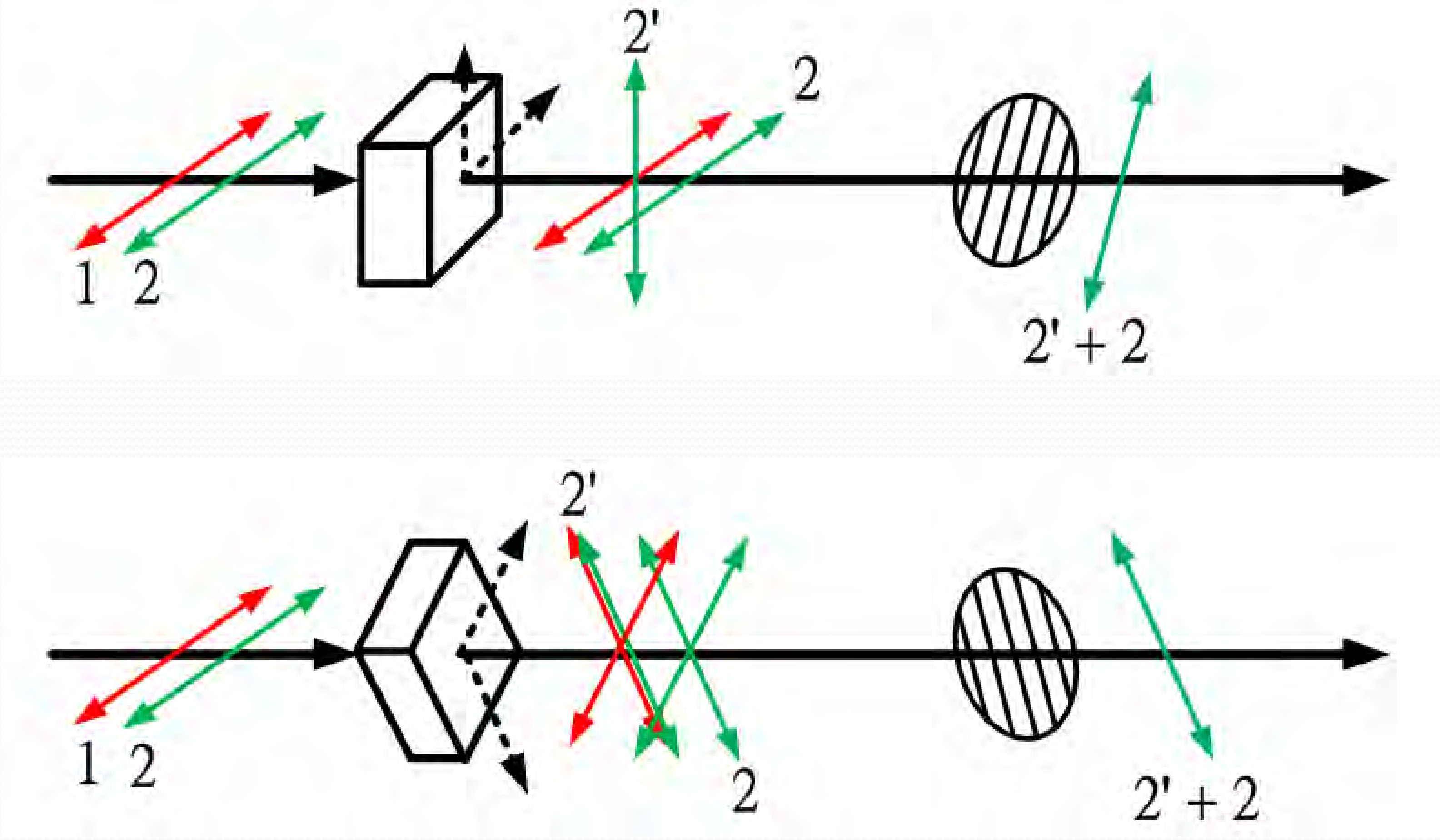
Amplitude
Modulator
For 1064
532
355
266

Phase Modulator
For 1064
532
355
266

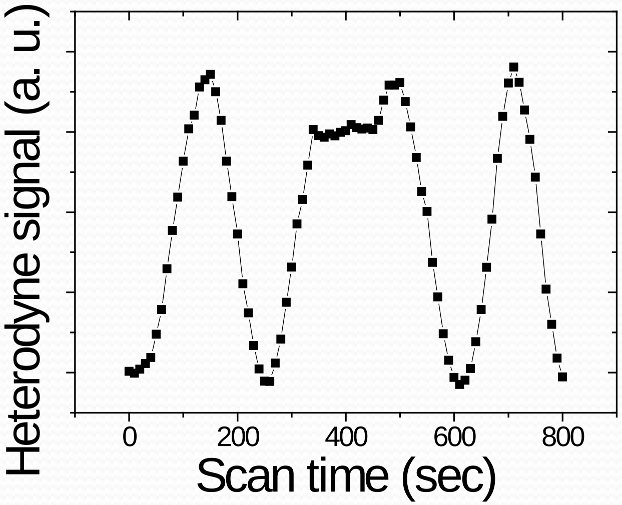
Relative Phase Measurement

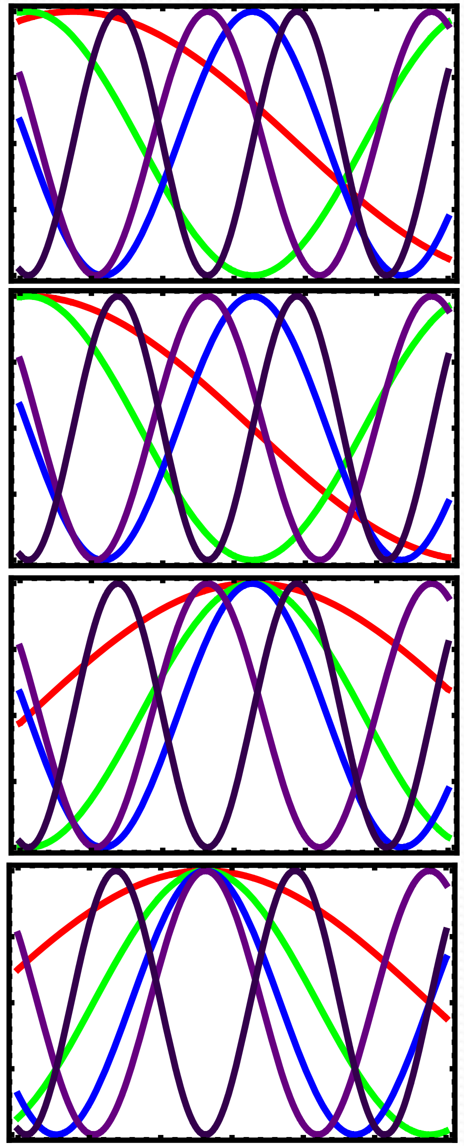


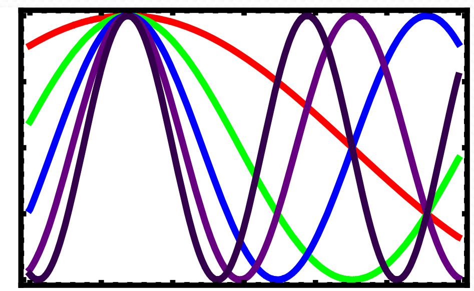
Relative Phase Measurement

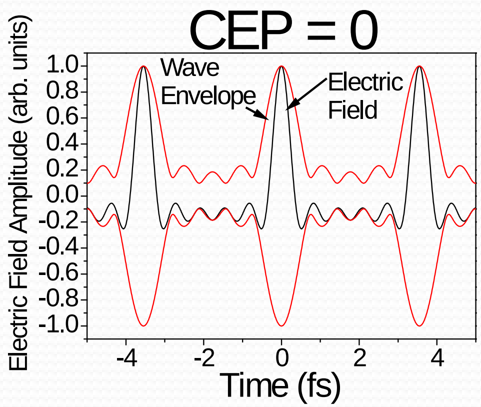


Relative Phase Measurement









The relative phase between harmonics

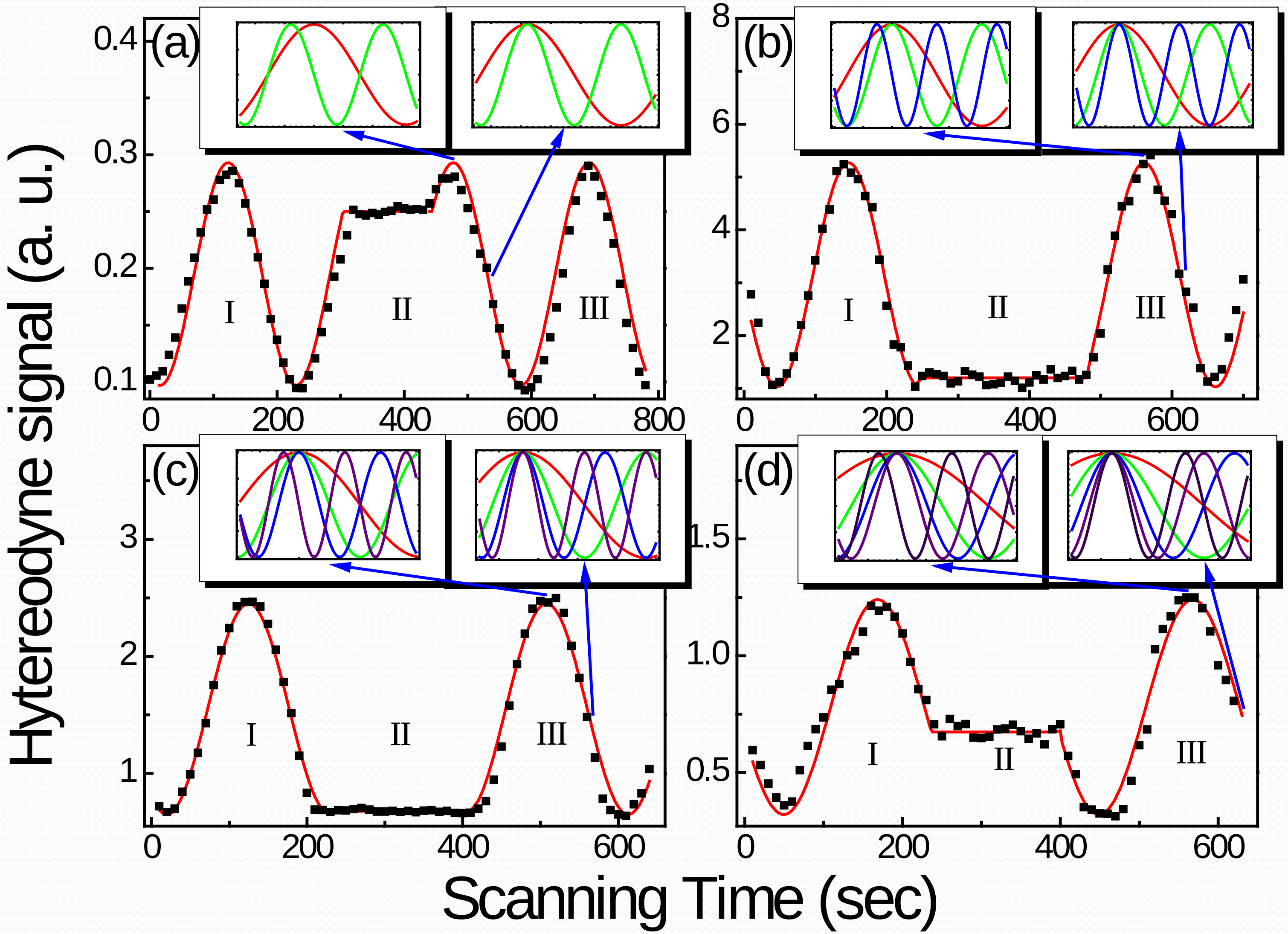
SHG: $\Phi_{532} = \Phi_{1064} + \pi/2$

SFG: $\Phi_{355} = \Phi_{1064} + \Phi_{532} + \pi/2$

SHG: $\Phi_{266} = \Phi_{532} + \pi/2$

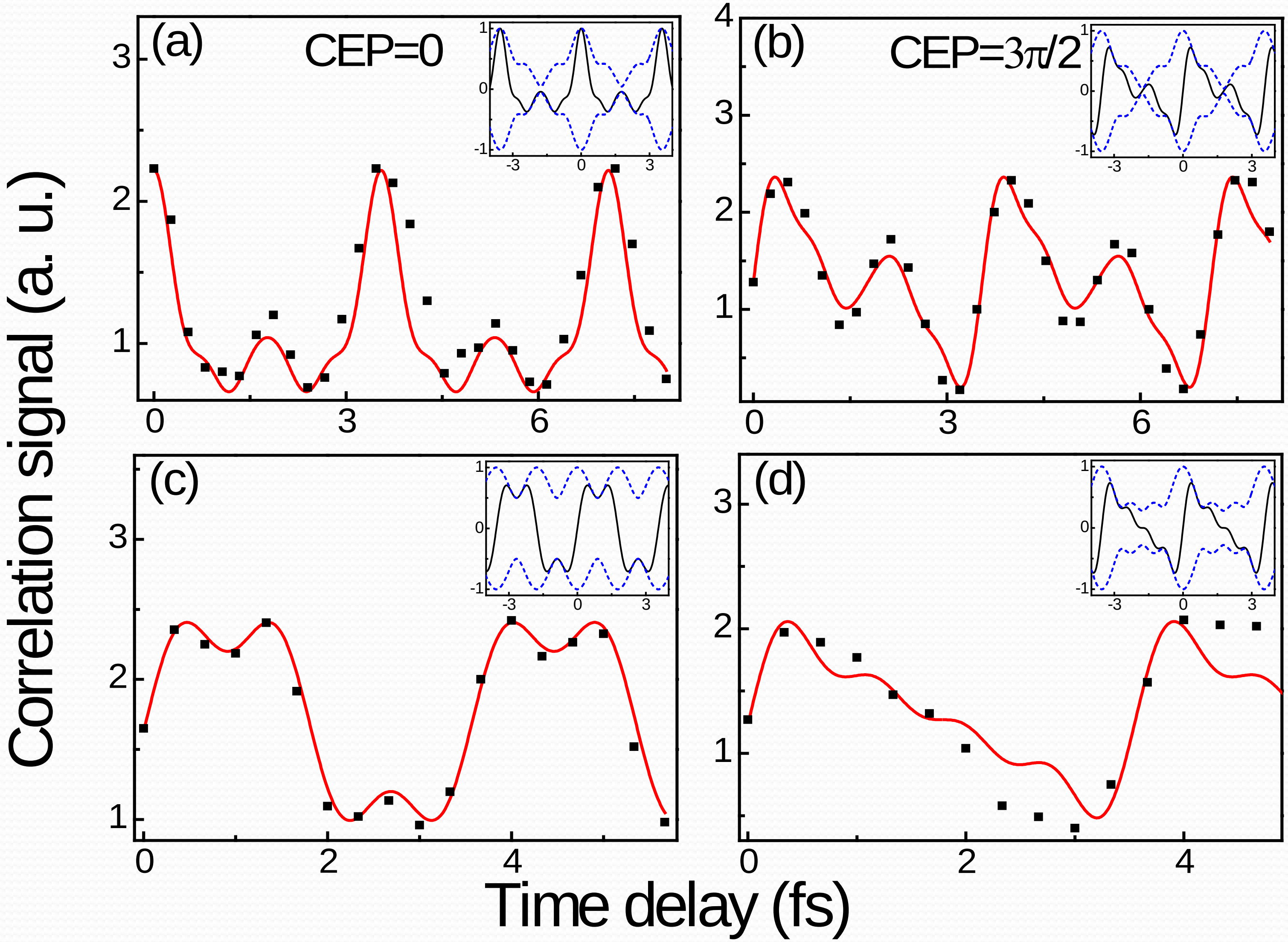
SFG: $\Phi_{213} = \Phi_{1064} + \Phi_{266} + \pi/2$

Relative Phase Measurement



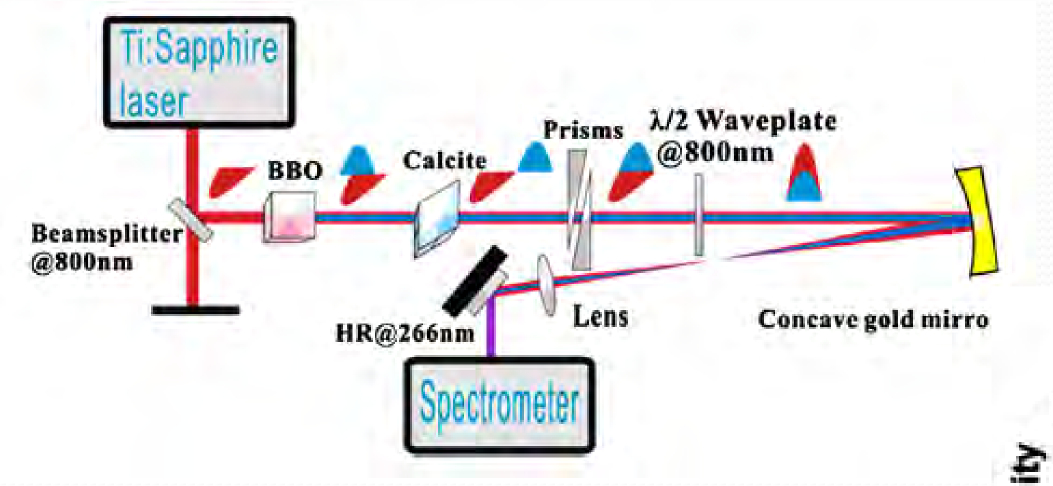
The heterodyne signal: (a) 532 nm, (b) 355 nm, (c) 266 nm, (d) 213 nm

Waveform Synthesized by five harmonics



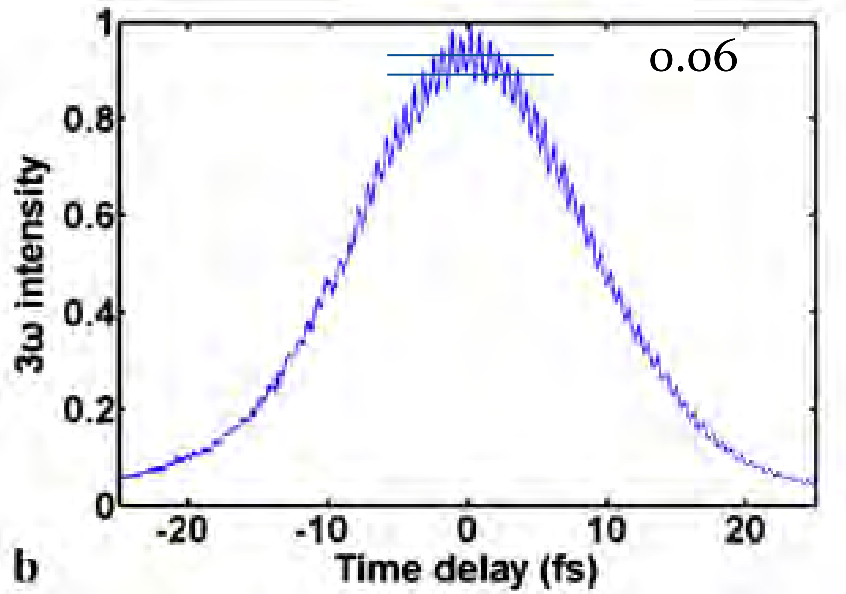
Laser Phys. Lett. 9, 212 (2012)

Third-harmonic generation in relative-phase-controlled two-color laser field



H. Xu, et al.,
Appl. Phys. B 104, 909 (2011)

$$S_{3\omega}(\tau) \propto a^2 A_\omega^6 \sqrt{\frac{\pi}{6}} + 9b^2 A_{2\omega}^4 A_\omega^2 \sqrt{\frac{\pi}{6}} \cdot \exp\left(-\frac{4\tau^2}{3\tau_\omega^2}\right)$$
$$+ 6ab A_\omega^4 A_{2\omega}^2 \sqrt{\frac{\pi}{6}} \cos 4\omega\tau \, \exp\left(-\frac{4\tau^2}{3\tau_\omega^2}\right)$$



Third-Harmonic Generation by two-color

$$\tilde{E}_i(z,t) = E_i e^{-i\omega_i t} + c.c. \qquad E_i = A_i e^{i(k_i z + \phi_i)}$$

$$\begin{cases} \tilde{P}^{(3)}(z,t) = \varepsilon_0 \chi^{(3)} \tilde{E}^3(z,t) \\ \tilde{E}(z,t) = \tilde{E}_1(z,t) + \tilde{E}_2(z,t) \qquad \phi_1 = 0,\ \Delta\phi = \phi_2 - \phi_1 = \phi_2 \end{cases}$$

$$\tilde{P}_3^{(3)}(z,t) = \varepsilon_0 (a\tilde{E}_1^3(z,t) + 3b\tilde{E}_2^2(z,t)\tilde{E}_1^*(z,t) + c.c.$$

$$a = \chi^{(3)}(\omega_3; \omega_1, \omega_1, \omega_1) \qquad b = \chi^{(3)}(\omega_3; \omega_2, \omega_2, -\omega_1)$$

Third-Harmonic Generation by two-color

$$\tilde{E}_3(t) = \varepsilon_0 (a e^{-i3\omega_1 t} \int_{-L/2}^{L/2} A_1^3 e^{-i\Delta k_{13} z} dz + 3b e^{-i3\omega_1 t} \int_{-L/2}^{L/2} A_2^2 A_1^* e^{-i(\Delta k_{123} z + 2\Delta\phi)} dz) + c.c.$$

$$= \varepsilon_0 (a A_1^3 \sin c(\frac{\Delta k_{13} L}{2}) \sin(3\omega_1 t) + 3b A_2^2 A_1^* \sin c(\frac{\Delta k_{123} L}{2}) \sin(3\omega_1 t + 2\Delta\phi)$$

$$I_3(\Delta\phi) \propto \int \tilde{E}_3^2(t) dt$$

$$= a^2 A_1^6 \sin c^2 (\frac{\Delta k_{13} L}{2}) + 9b^2 A_2^4 A_1^2 \sin c^2 (\frac{\Delta k_{123} L}{2})$$

$$+ 6ab A_1^4 A_2^2 \sin c(\frac{\Delta k_{13} L}{2}) \sin c(\frac{\Delta k_{123} L}{2}) \cos(2\Delta\phi)$$

Relative phase measurement for multi-color waveform synthesis

$$E(t) = \sum_n A_n \cos(n\omega t + \phi_n)$$

$$E(t) = \sum_n A_n \cos(n\omega(t - \phi_1/\omega) + \phi_n) = \sum_n A_n \cos(n\omega t + \phi_n - n\phi_1)$$

$\phi'_1 \rightarrow 0$, $\phi'_n \rightarrow \phi_n - n\phi_1$.

$\phi_n - \phi_{n-1} = \phi_{n-1} - \phi_{n-2} = \ldots = \phi_2 - \phi_1$

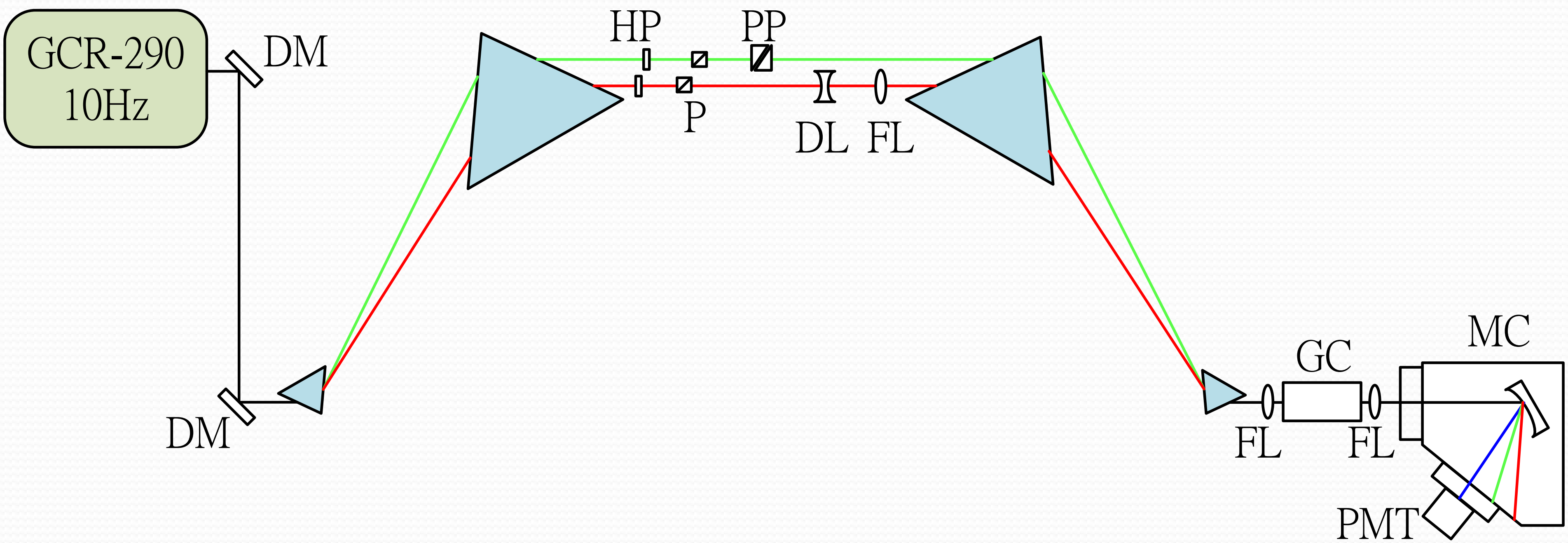
Interference of FWMs

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

$\Delta\phi = \phi_2 - \phi_1$

Interference of FWM and THG

Experiment Setup for two-color THG



DM: dichroic mirror; HP: half-wave plate; P: polarizer; PP: prism pair; DL: defocus lens; FL: focus lens; GC: gas cell;
MC: monochromator; PMT: photomultiplier tube

Third-Harmonic Generation by 2-color

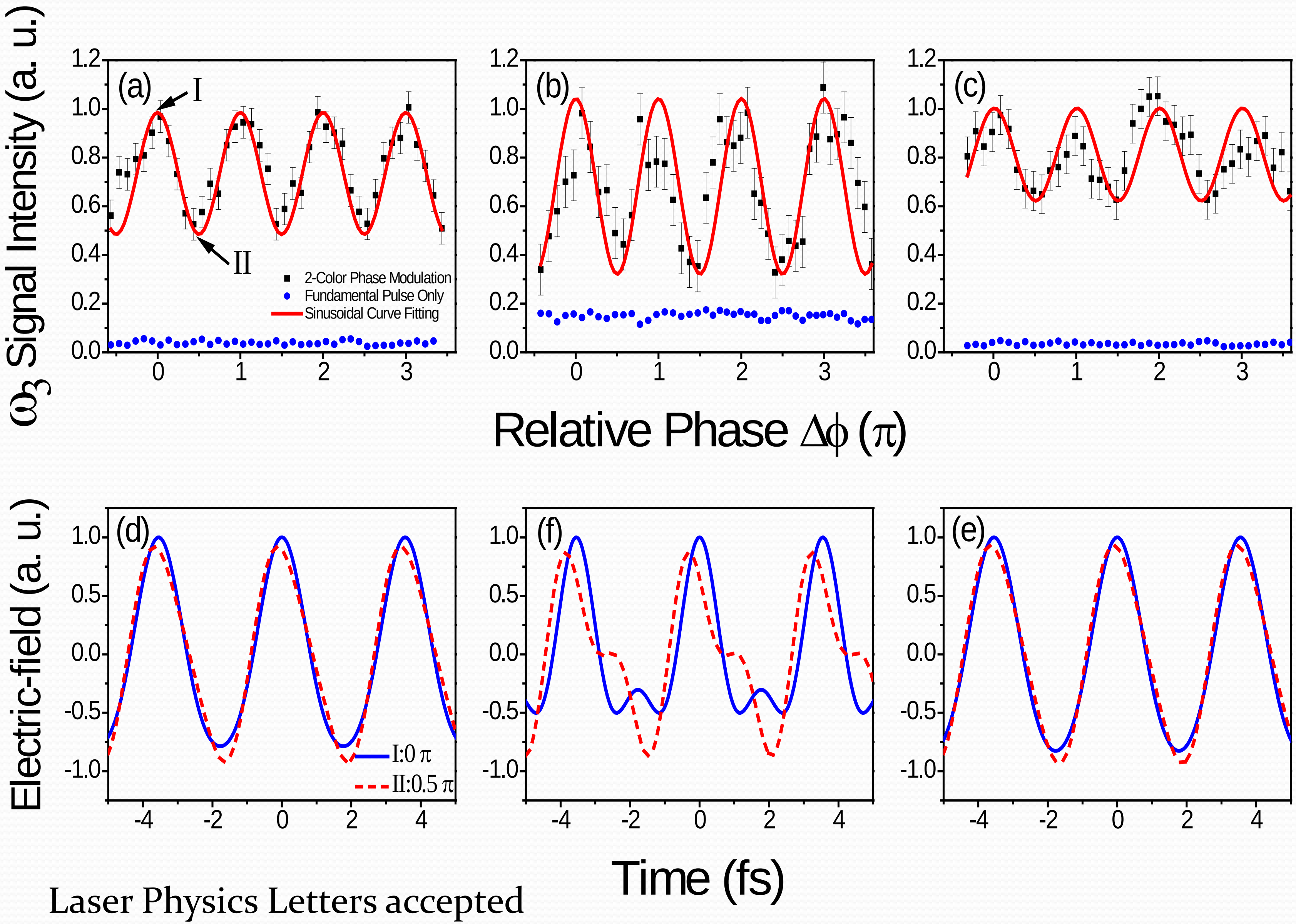
$$I_3(\Delta\phi) \propto A_1^6 \sin c^2(\frac{\Delta k_{13}L}{2}) + 9A_2^4 A_1^2 \sin c^2(\frac{\Delta k_{123}L}{2})$$

$$+6A_1^4 A_2^2 \sin c(\frac{\Delta k_{13}L}{2}) \sin c(\frac{\Delta k_{123}L}{2}) \cos(2\Delta\phi) \qquad (6)$$

$$= \alpha^2 A_1^6 + 9\beta^2 A_2^4 A_1^2 + 6\alpha\beta A_1^4 A_2^2 \cos(2\Delta\phi)$$

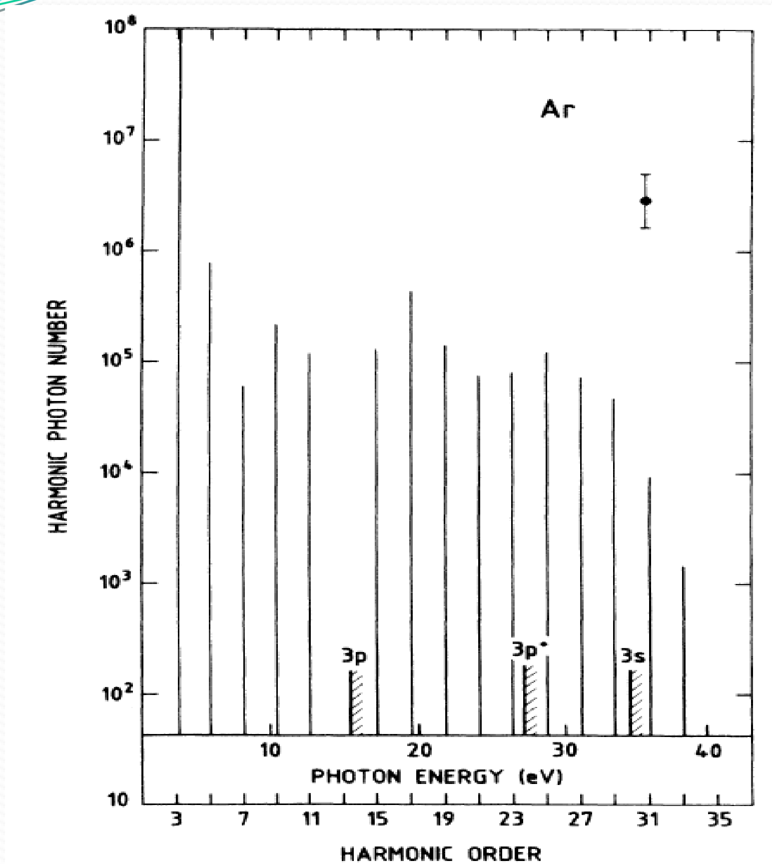
where $\alpha = \sin c(\frac{\Delta k_{13}L}{2}), \beta = \sin c(\frac{\Delta k_{123}L}{2})$

Third-Harmonic Signal



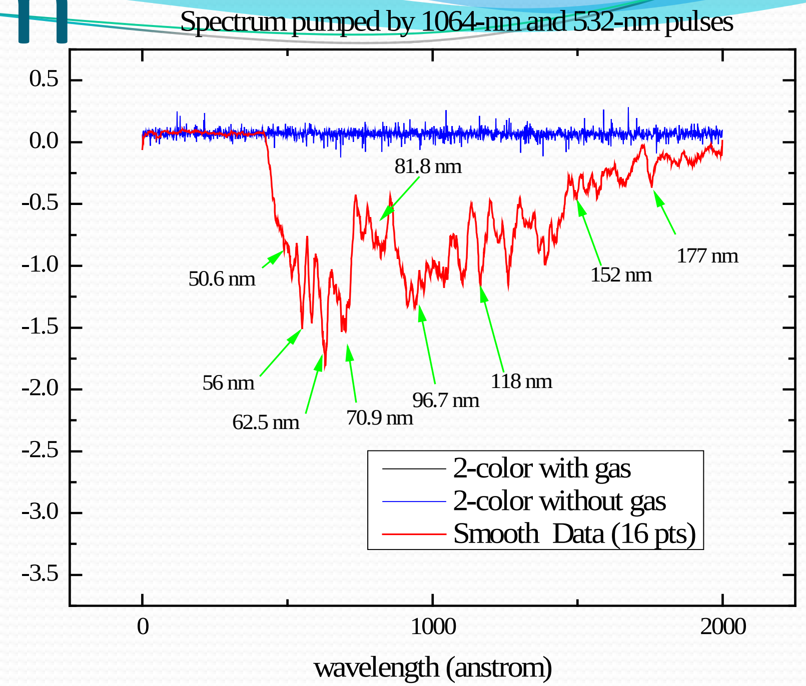
Laser Physics Letters accepted

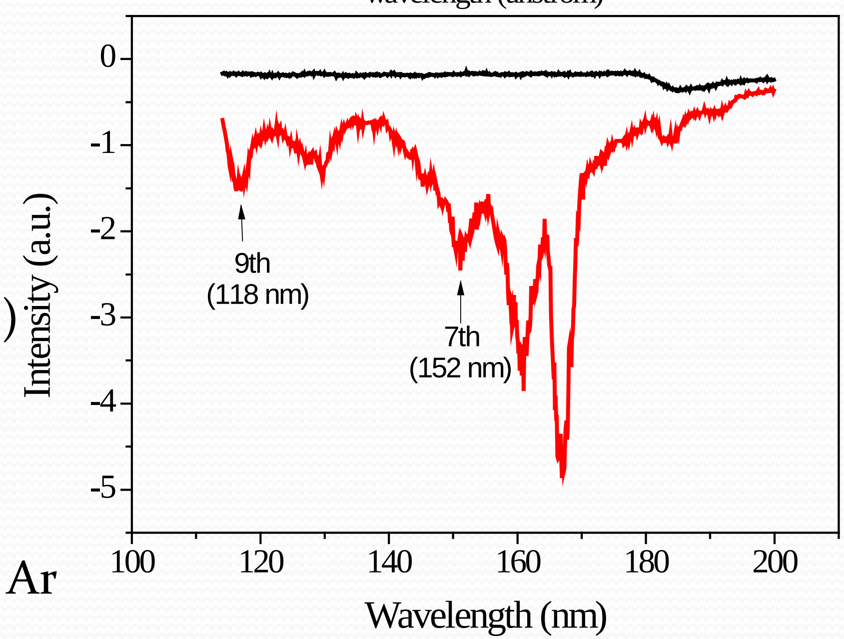
HHG by 1064 nm



X. F. Li, et al. Phys. Rev. A 39, 5751 (1989)

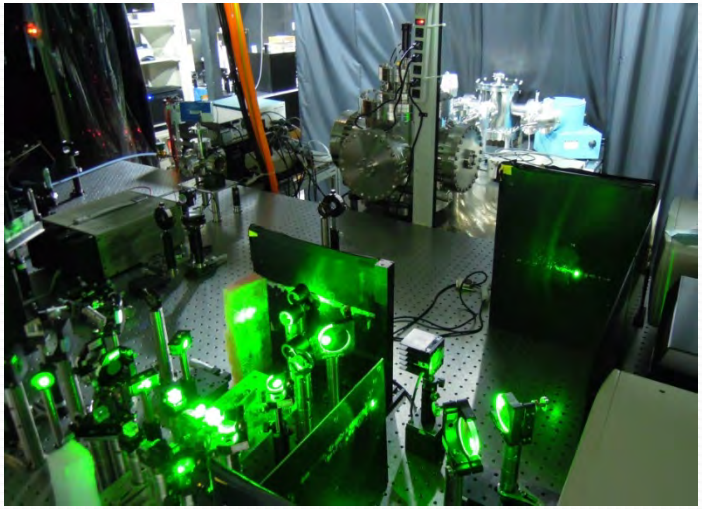
$3x10^{13}$ W/cm², 15 Torr Ar

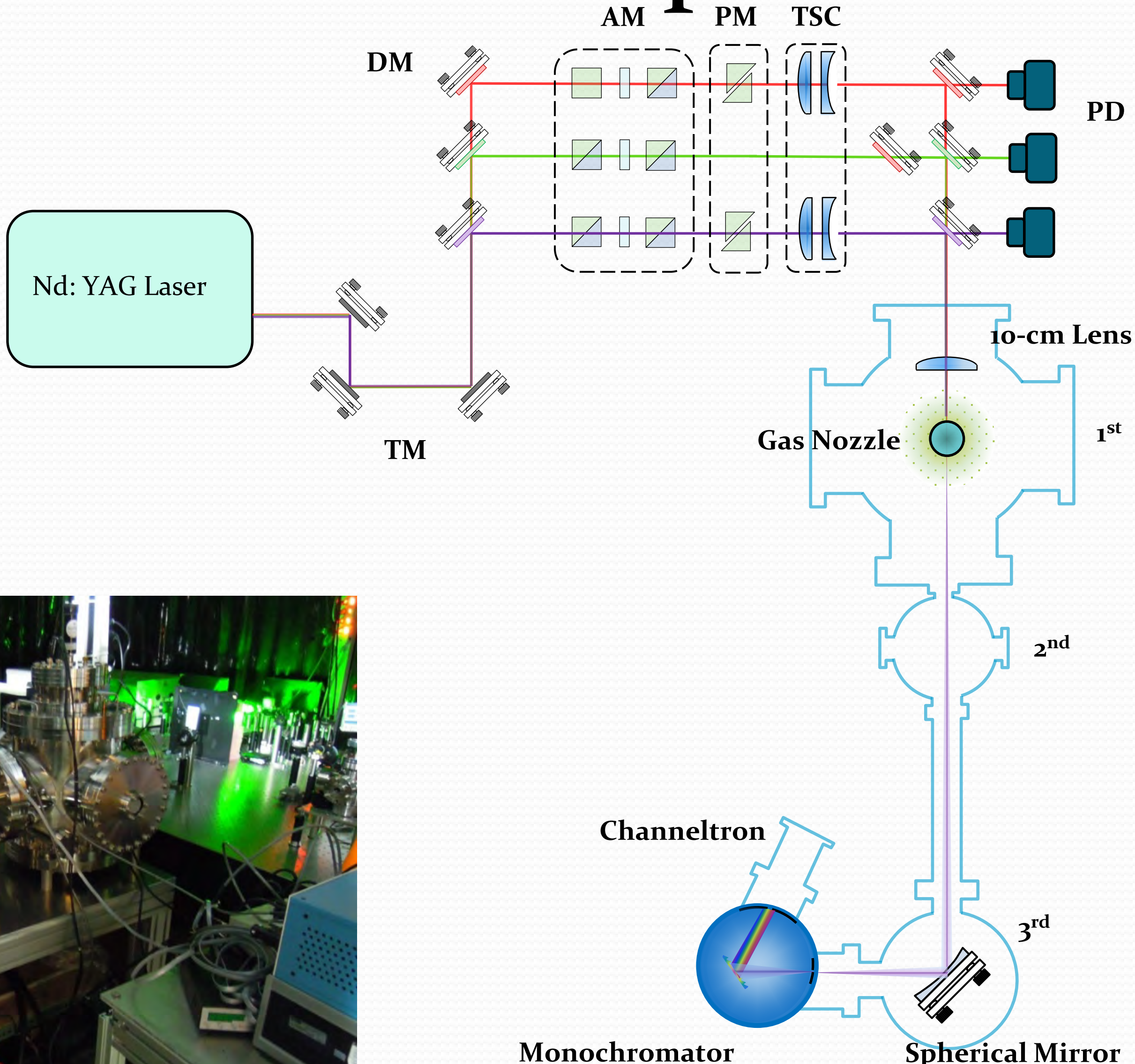


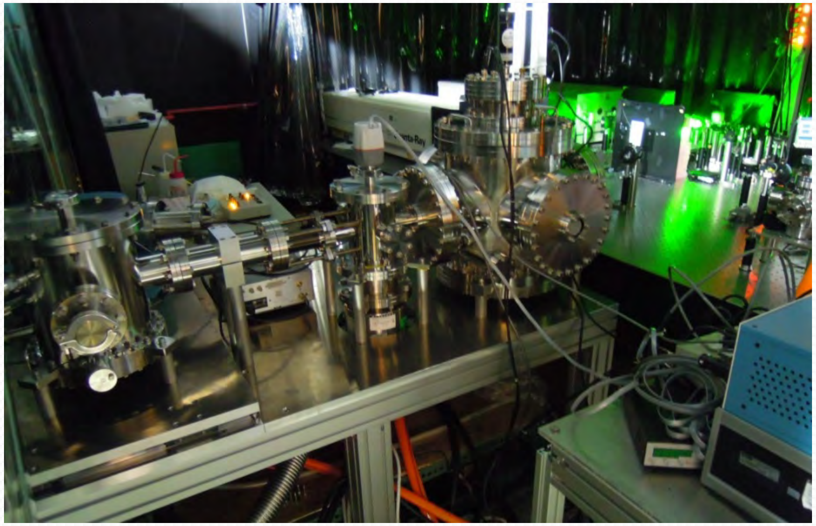


$3.66x10^{13}$ W/cm², 12 Torr Ar

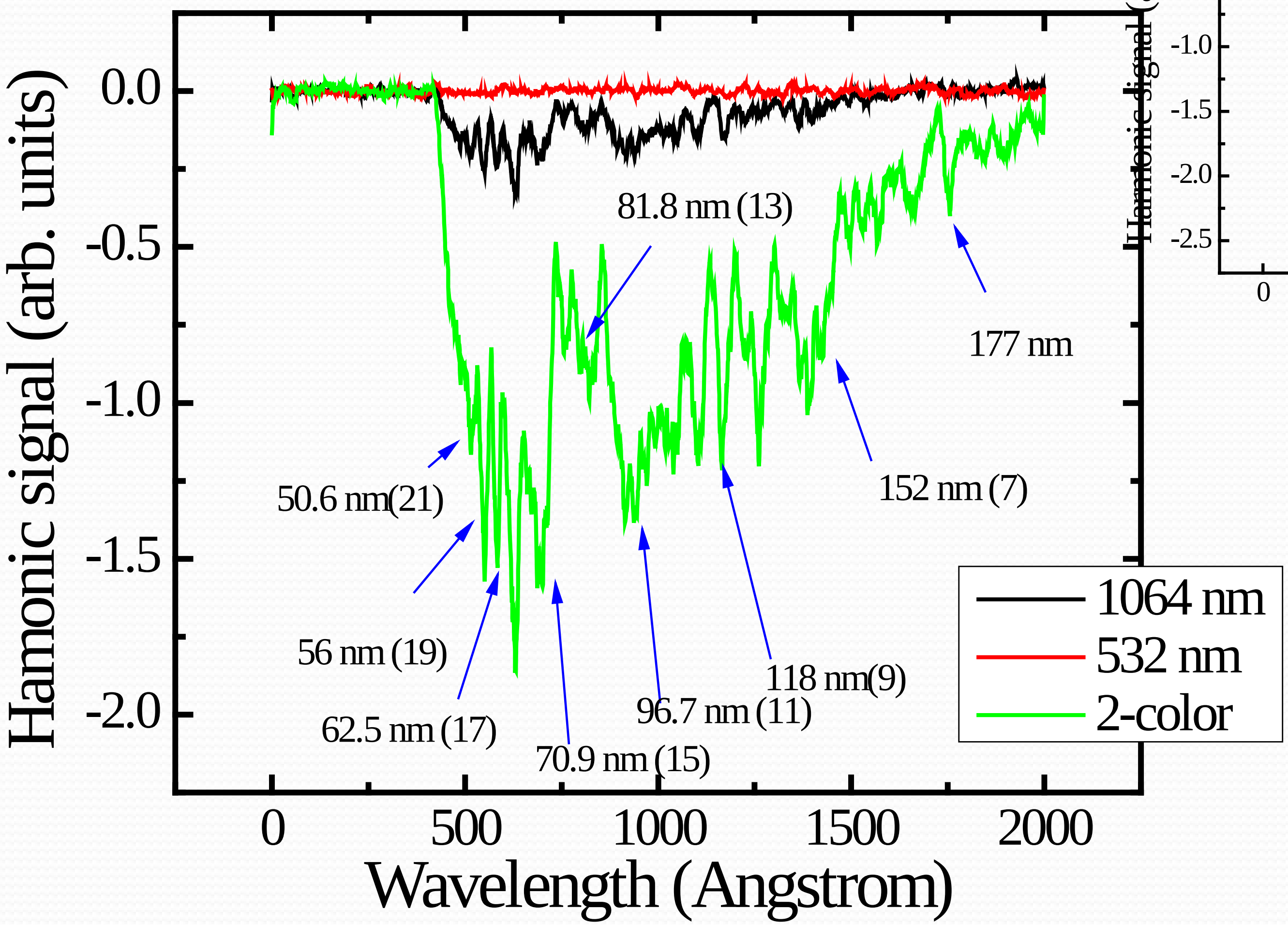
Experiment setup

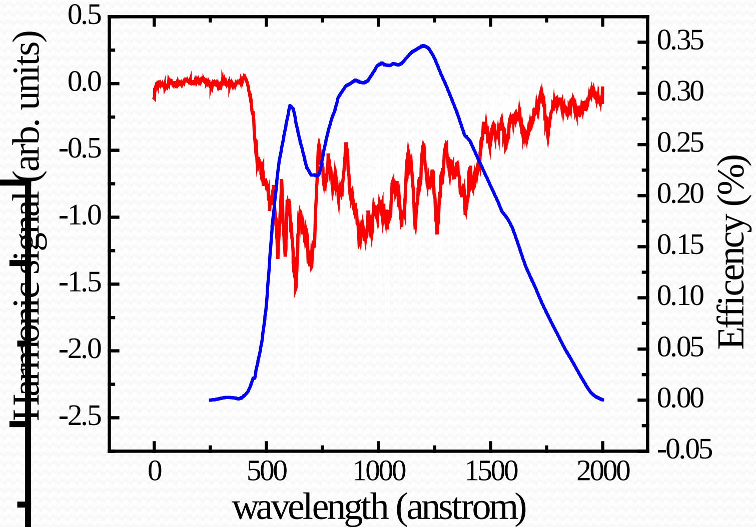




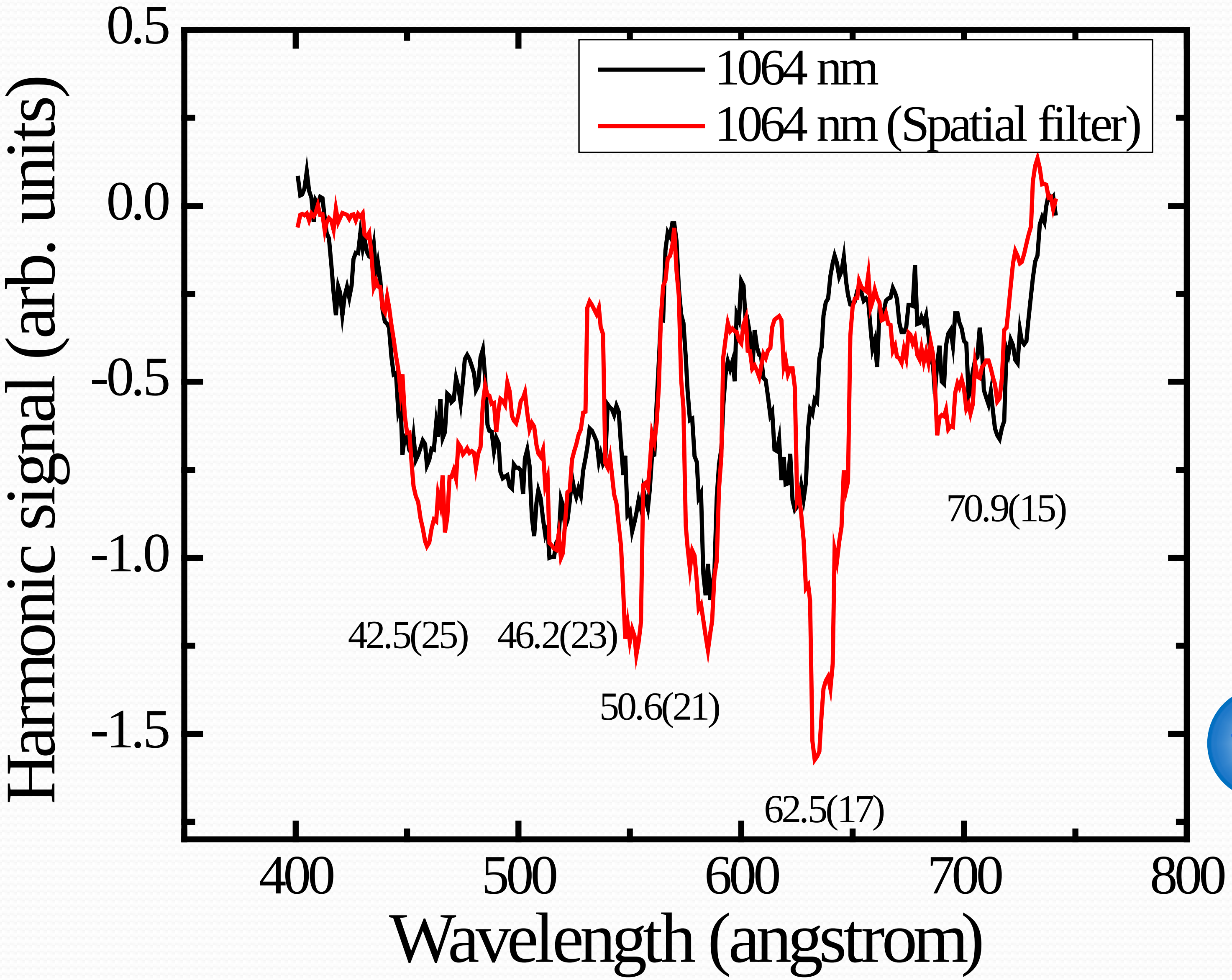


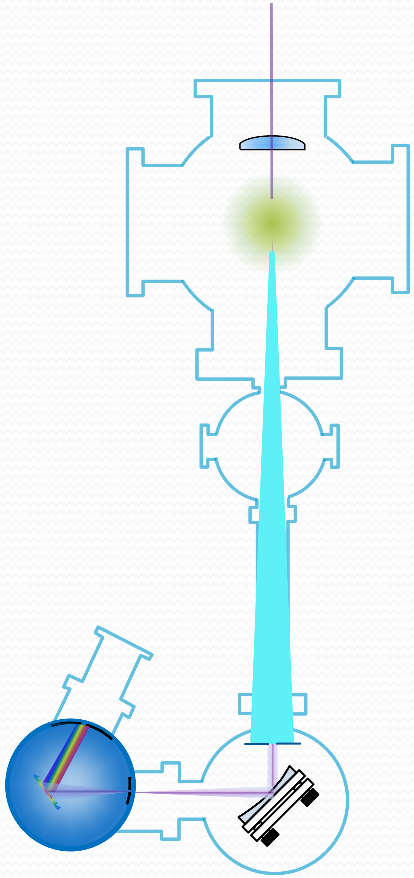
Two-color Harmonic Generation



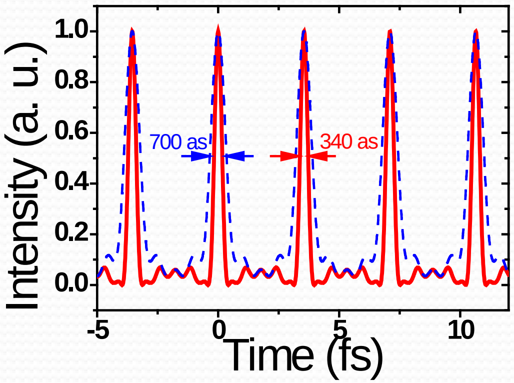


Harmonics and Fluorescence





Summary



When CEP=0
=>The pulse width ~ 340 as
Focusing to a Φ20μm spot, the intensity will reach 10^{14} W/cm^2.

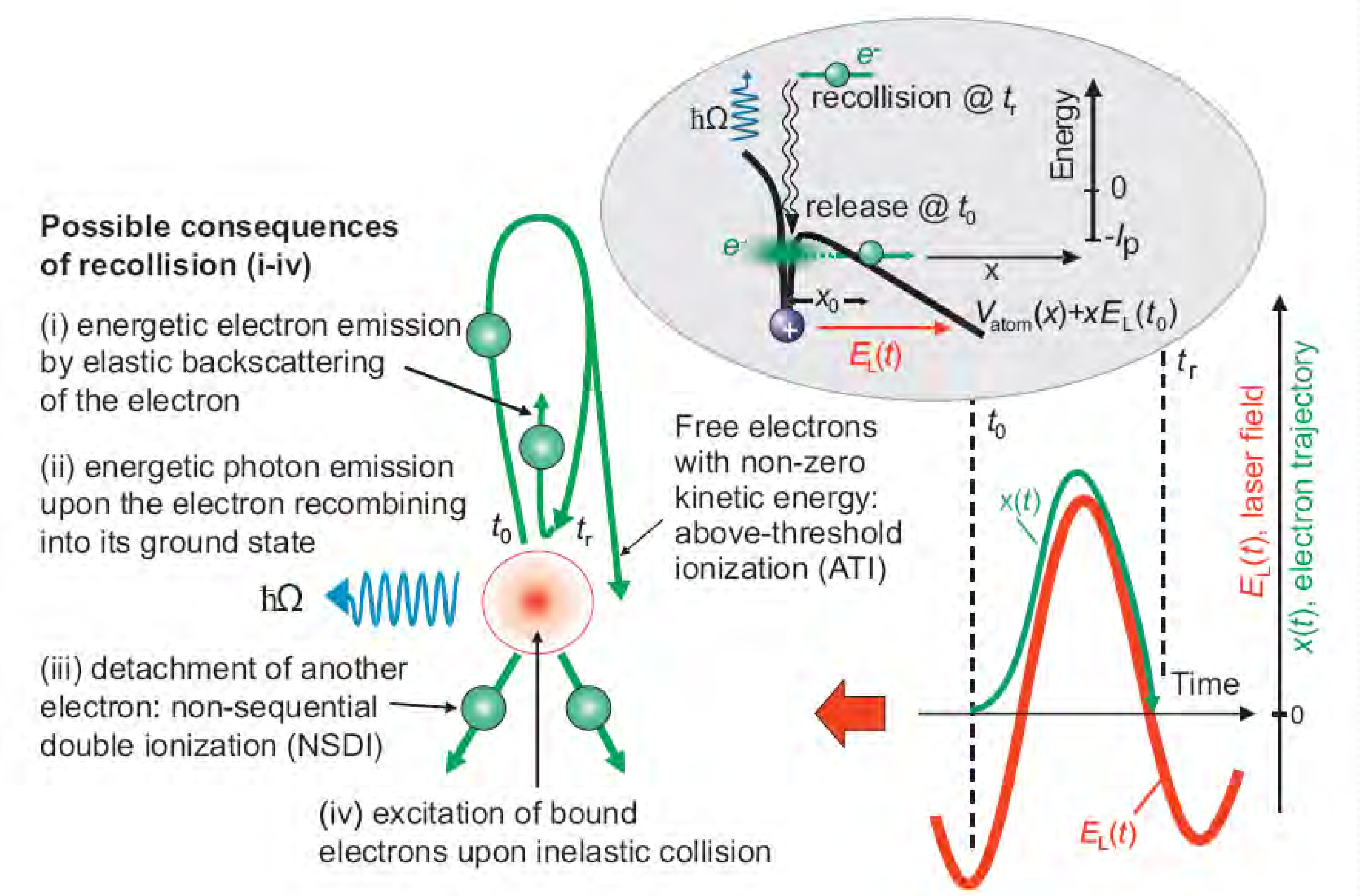
We have investigated the generation of TH on the influence of relative phases and amplitudes of the two-color fields. A modulation depth as high as 0.35 has been observed.

We present a promising way for *in situ* determination of the relative phase in multi-color laser field.

Waveform synthesis in the VUV spectral range by higher harmonics generation using waveform-controlled multi-colour quasi-single-frequency laser fields is feasible.

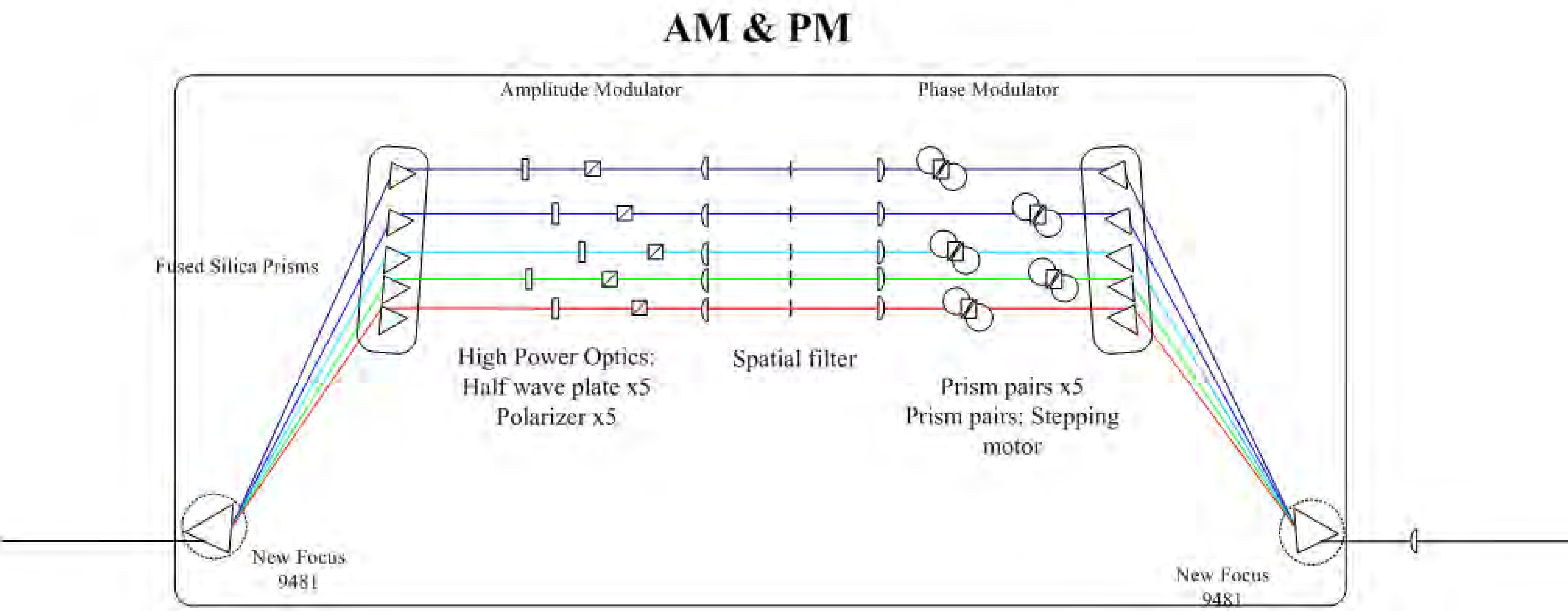
The plasma induced by the two-color laser field is shown to have a significant effect on generation of the harmonic signal.

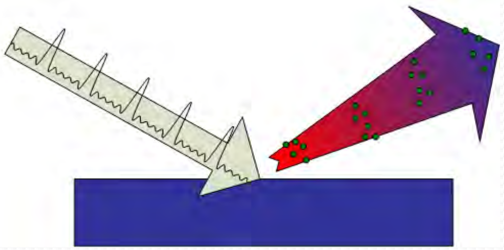
Photoelectron and/or ion measurement

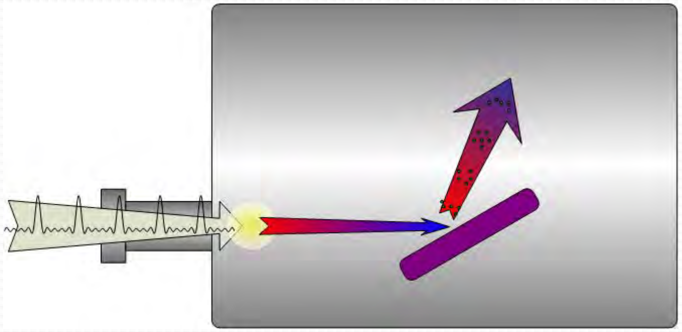


F. Krausz & M. Ivanov, Rev. Mod. Phys. 81 163 (2009)

Quantum Stroboscope for Electron Motion







Photoelectron emission &
Nonlinear optics

Thanks for your attention