Synthesis and Control of Single Cycle Optical Pulses

for Quantum Control and Attosecond Physics

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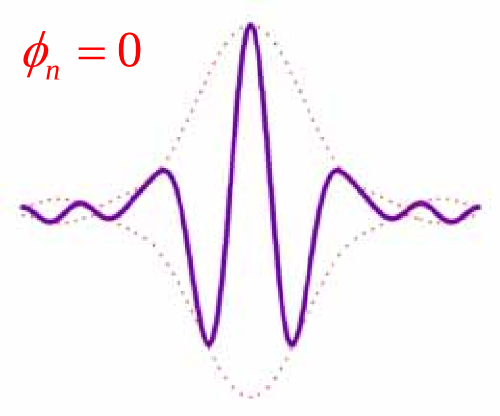
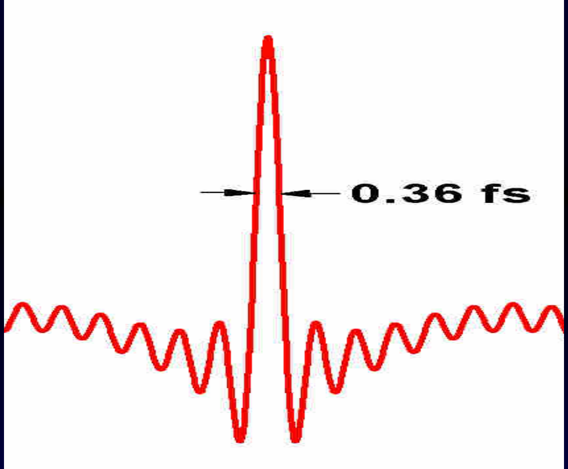
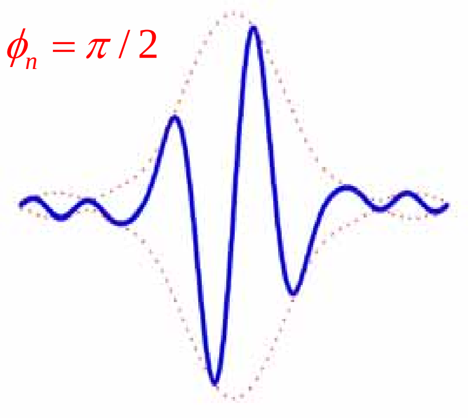
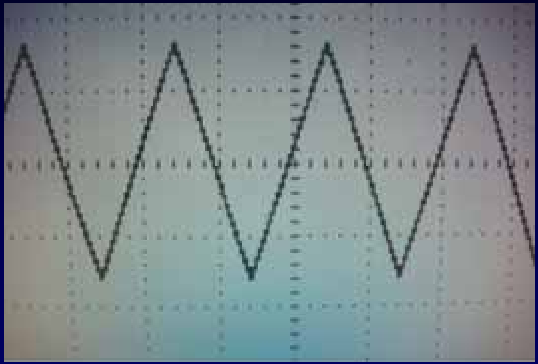
April 8, 2009

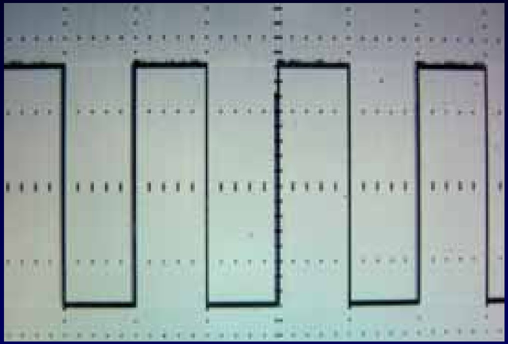
Single-cycle and sub-cycle pulses











300 nm = 1 fs

single-cycle pulse $\equiv$ attosecond pulse

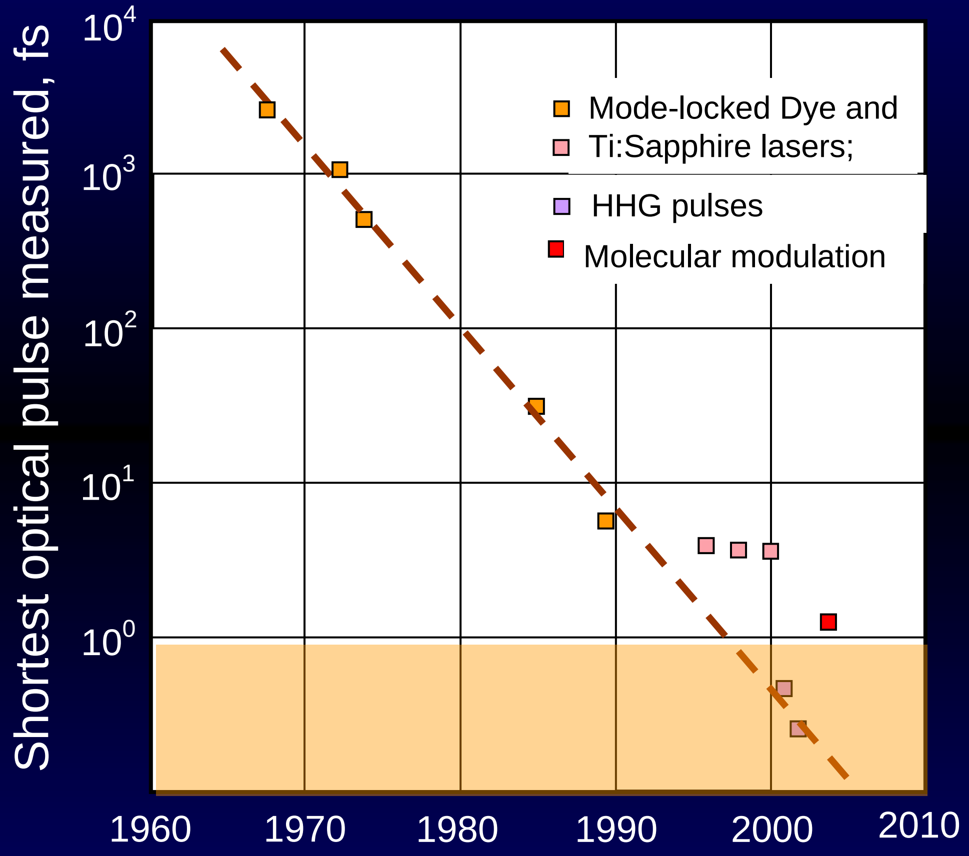
Why we are interested in it

How we do it

What we have done

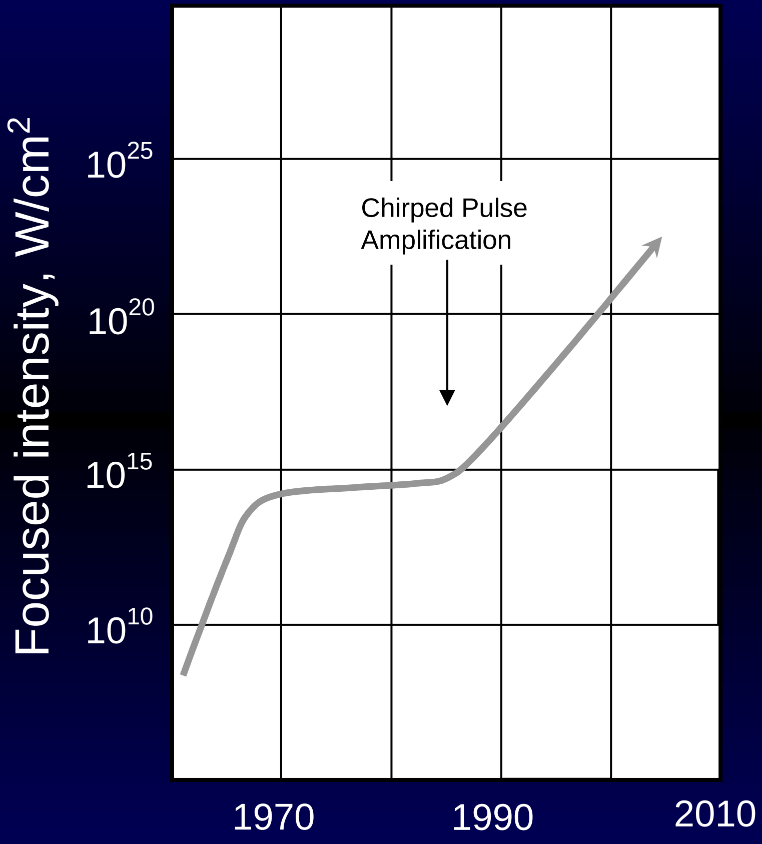
What could we do with these light pulses

Laser pulses got shorter over the years



Ultrafast science

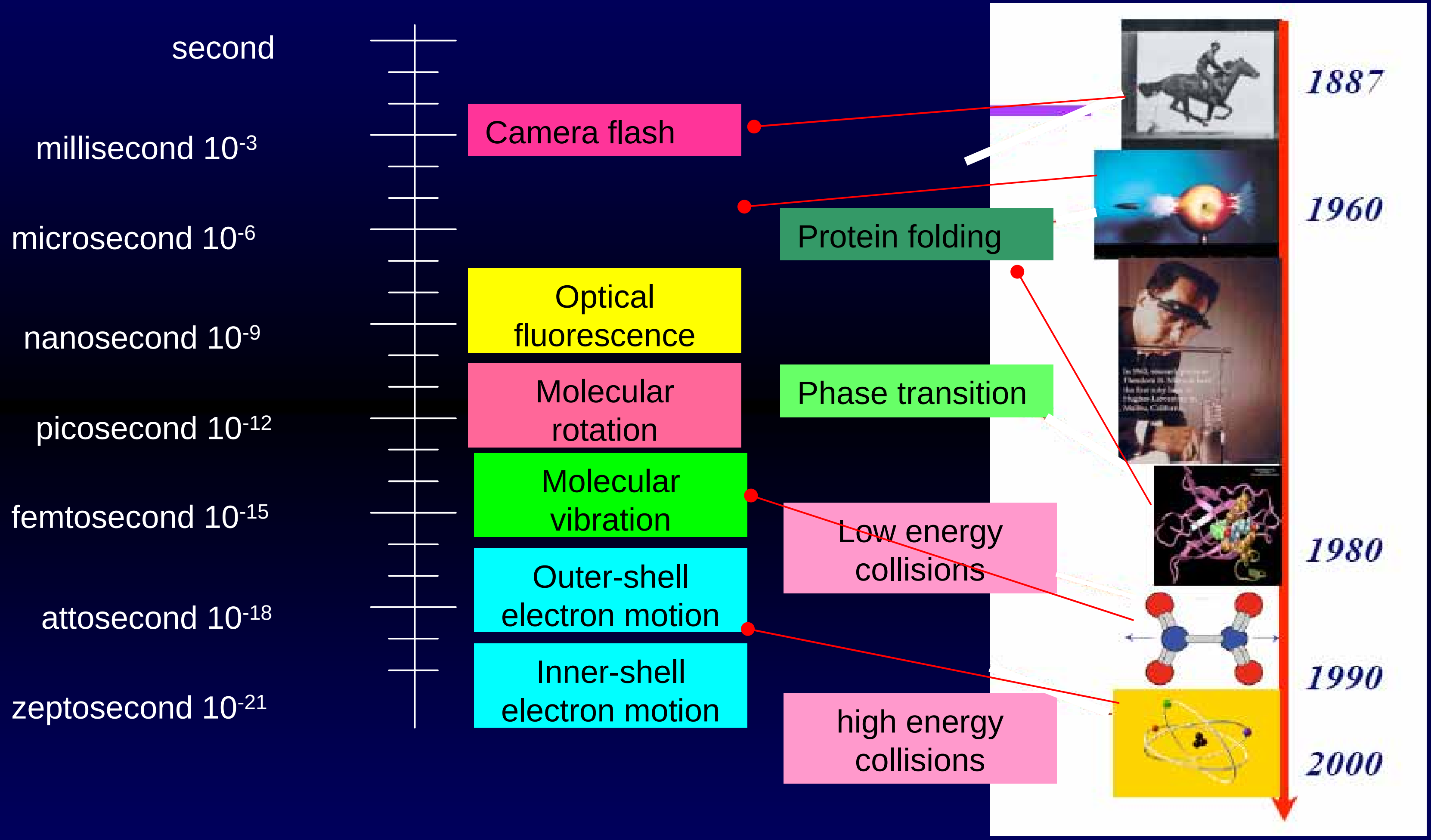
Peak intensity increased



High field physics

IAMS

Generating single-cycle pulses – in pursuit of attosecond pulse timing



second

millisecond 10^{-3}

microsecond 10^{-6}

nanosecond 10^{-9}

picosecond 10^{-12}

femtosecond 10^{-15}

attosecond 10^{-18}

zeptosecond 10^{-21}

Camera flash

Optical fluorescence

Molecular rotation

Molecular vibration

Outer-shell electron motion

Inner-shell electron motion

Protein folding

Phase transition

Low energy collisions

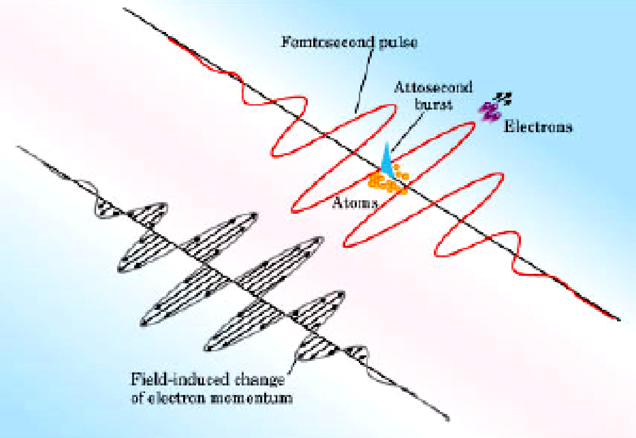
high energy collisions

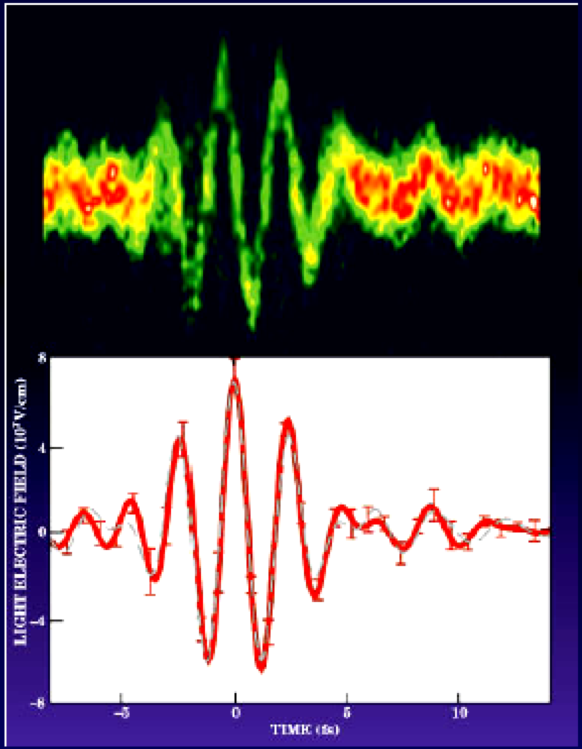
PHYSICS TODAY October 2004

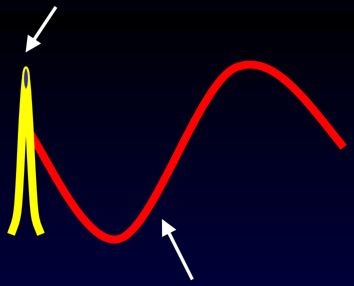
Search and Discovery

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.







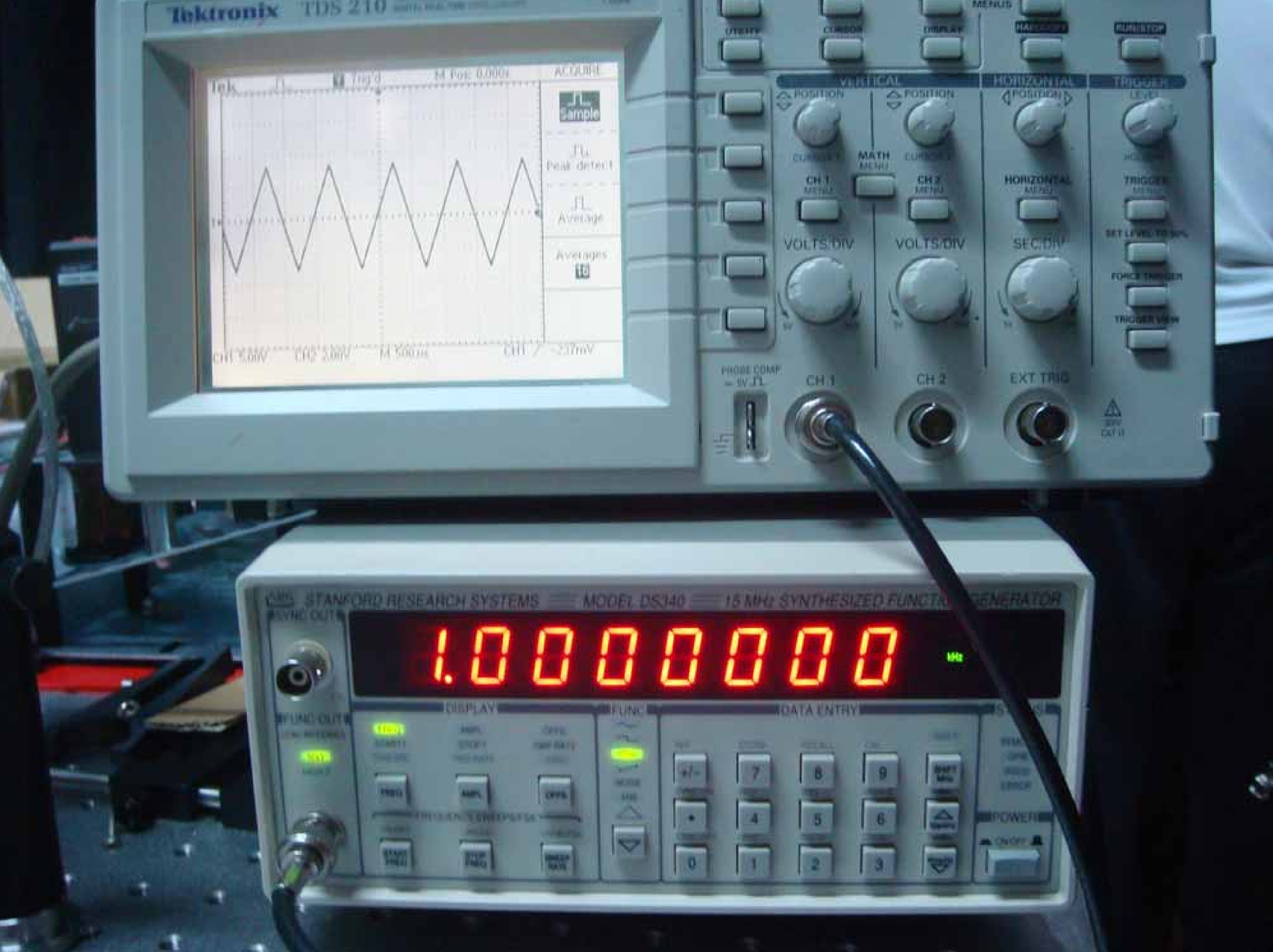
Single-cycle pulse

Sinusoidal pulse

GOAL

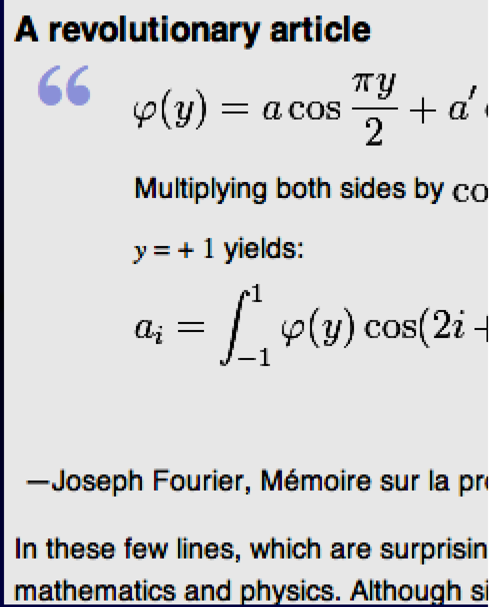
Synthesize various forms of ultrashort pulses

in the optical frequency (10^{15} cps) regime


Tektronix TDS 210
ACQUIRE
Sample
Peak detect
Average
Averages
16
CH1 5.00V
CH2 2.00V
M 500µs
CH1 ∕ -237mV
VERTICAL
HORIZONTAL
TRIGGER
POSITION
MATH MENU
CH 1 MENU
CH 2 MENU
HORIZONTAL MENU
TRIGGER MENU
VOLTS/DIV
SEC/DIV
SET LEVEL TO 50%
FORCE TRIGGER
TRIGGER VIEW
PROBE COMP
CH 1
CH 2
EXT TRIG
STANFORD RESEARCH SYSTEMS
MODEL DS340
15 MHz SYNTHESIZED FUNCTION GENERATOR
1.0000000
kHz
SYNC OUT
FUNC OUT
DISPLAY
FUNC
DATA ENTRY
POWER
ON/OFF

Basic Concepts

Jean Batiste Joseph Fourier (1768-1830)



A revolutionary article

$$\varphi(y) = a\cos\frac{\pi y}{2} + a'$$

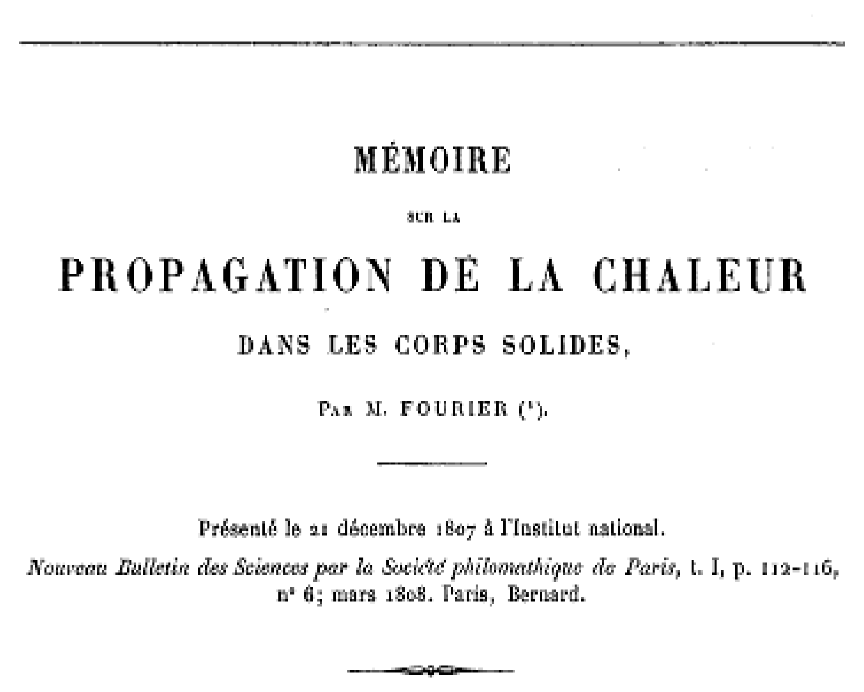
Multiplying both sides by co

$y = +1$ yields:

$$a_i = \int_{-1}^{1} \varphi(y)\cos(2i +$$

—Joseph Fourier, Mémoire sur la pr

In these few lines, which are surprisin
mathematics and physics. Although si



MÉMOIRE

SUR LA

PROPAGATION DE LA CHALEUR

DANS LES CORPS SOLIDES,

PAR M. FOURIER (1).

Présenté le 21 décembre 1807 à l'Institut national.

Nouveau Bulletin des Sciences par la Société philomathique de Paris, t. I, p. 112-116, n° 6; mars 1808. Paris, Bernard.

oseph Fourier initiated the
tudy of Fourier series in order
solve the heat equation.

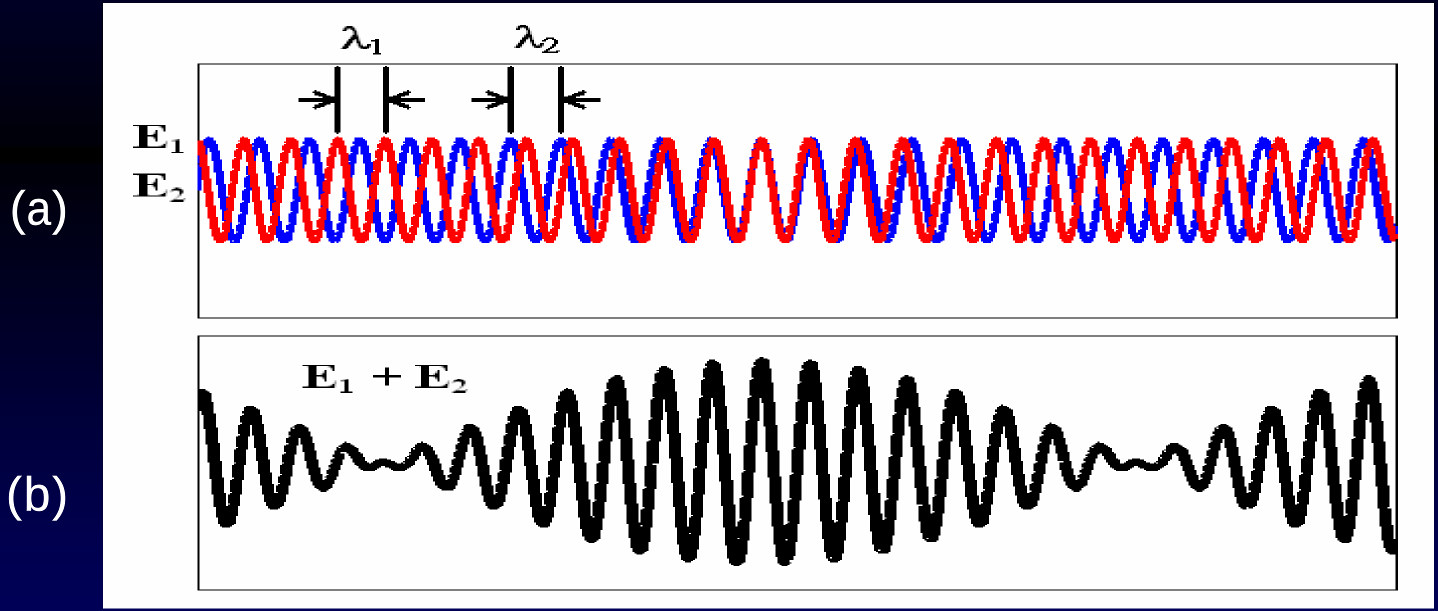
gly revolutionized both
iel Bernoulli and Gauss,

What is a single cycle optical pulse

(a) Monochromatic light: sinusoidal wave

(b) Beating of two waves, ω_1 and ω_2

$$\boxed{E(t) = A(t)\cos(\omega t + \phi)}$$



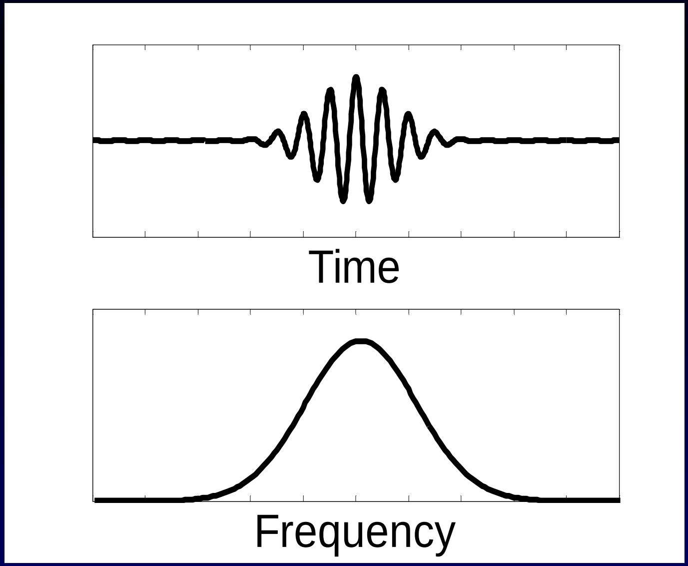
$$\boxed{E(t) = A_1(t)\cos(\omega_1 t + \phi_1) + A_2(t)\cos(\omega_2 t + \phi_2)}$$

What is a single cycle optical pulse

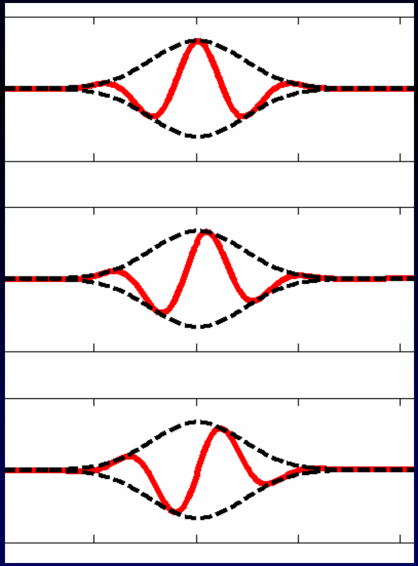
(c) Many waves propagating to form a wave packet (left)

(d) Ultimate wavepacket is a single-cycle and sub-cycle pulse train (right)

$$\boxed{E(t)=\sum_n E_n(t)=\sum_n A_n(t)\cos(\omega_n t+\phi_n)}$$



(c)

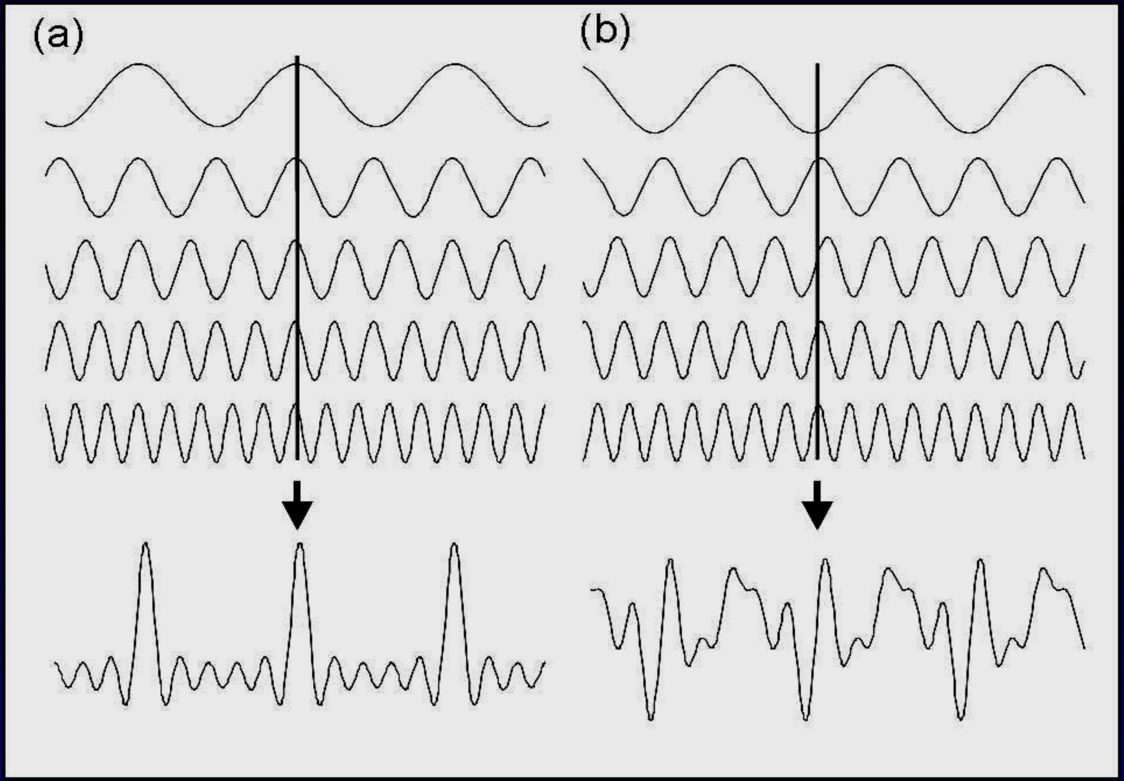


(d)

Phase coherence

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

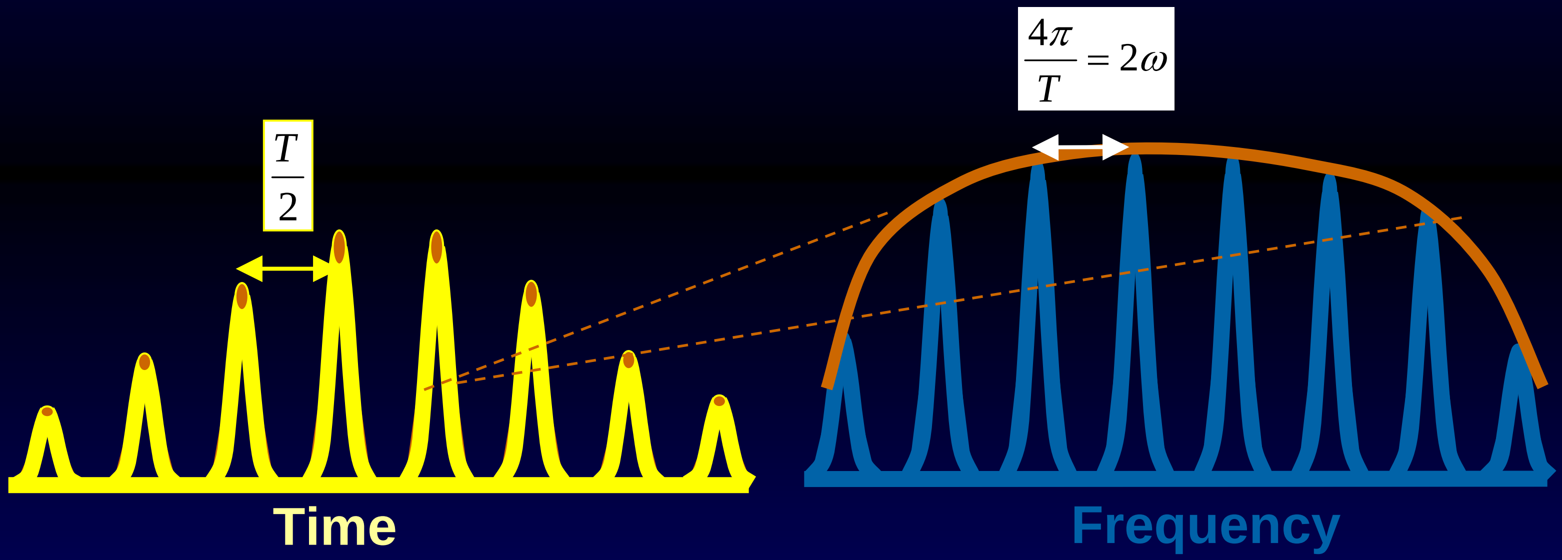
(a) In phase $\phi_n = n\phi_o$ (b) Random phases



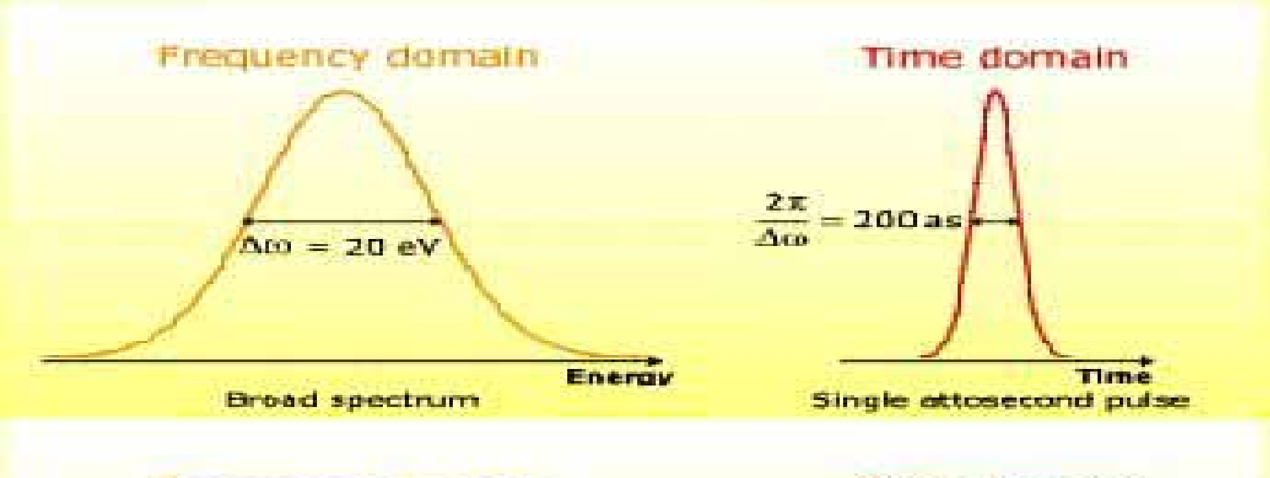
Correlation between time and frequency

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

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



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



Carrier frequency

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

single cycle

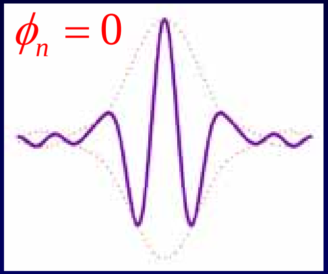
$$\Delta\omega \geq \omega_c$$

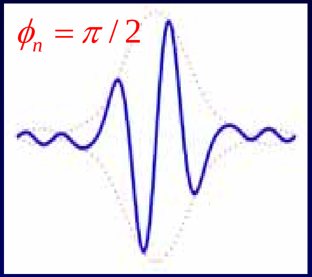
T. Brabec and F. Krausz, Phys. Rev. Lett. 78, 3282 (1997)

Things to remember

attosecond single-cycle optical pulses:

1. has a very broad spectrum – more than one octave
2. spectrum has perfectly phased spectral components
3. needs stable and controllable carrier-envelope phase





1 fs ↔ 300 nm

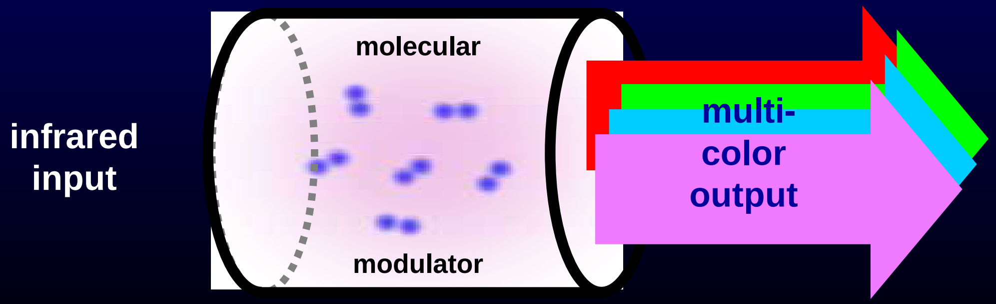
How to make a single-cycle pulse

A. Directly from a laser – phase-locking

B. From atoms – high harmonic generation

C. From molecules – molecular modulation

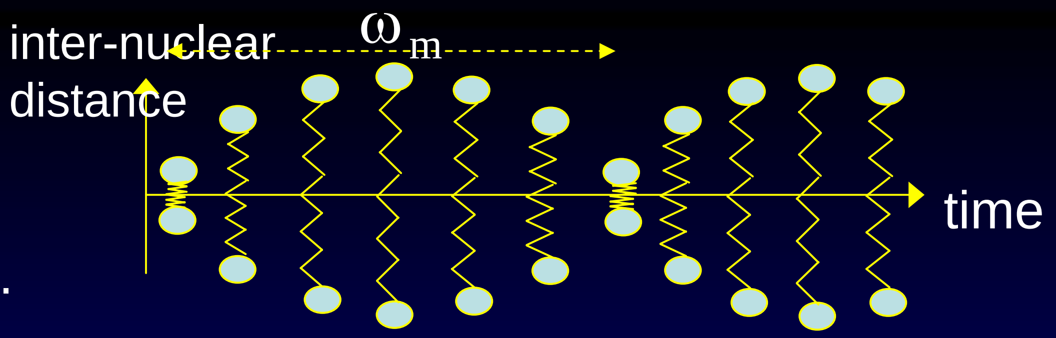
C Bandwidth expansion by molecular modulation



Refractive Index

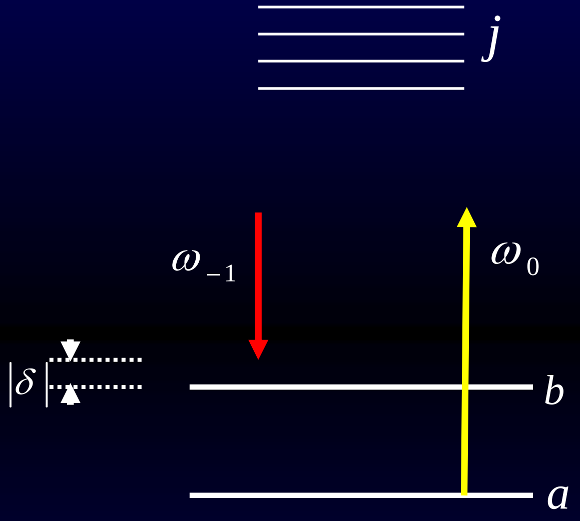
$$n = n_0 + \delta \cos \omega_m t$$

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



S. E. Harris and A. V. Sokolov, Phys. Rev. A **55**, R4019 (1997);
S. E. Harris and A. V. Sokolov, Phys. Rev. Lett. **81**, 2894 (1998).

C Bandwidth expansion by molecular modulation



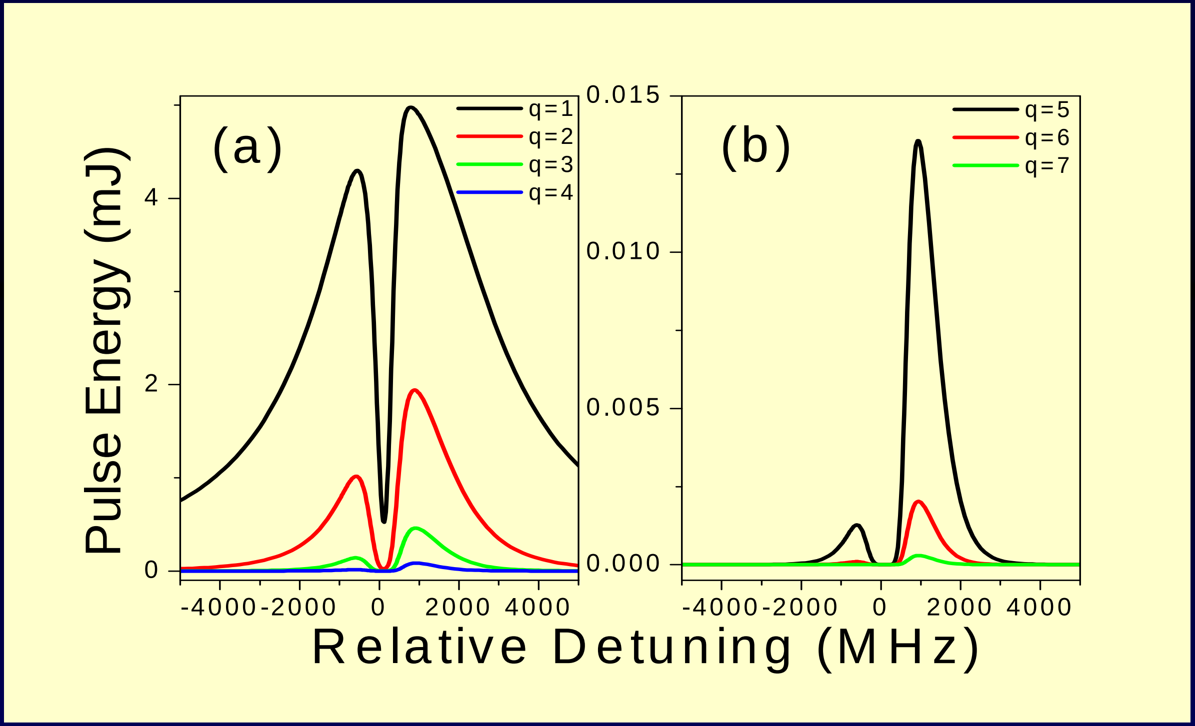
$$\frac{\partial \rho_{aa}}{\partial \tau} = i\left(\Omega_{ab}\rho_{ba} - \Omega_{ba}\rho_{ab}\right) + \gamma_{\parallel}\rho_{bb}$$

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

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

Detuning δ: adiabatic excitation, collinear sidebands

Bandwidth expansion by molecular modulation

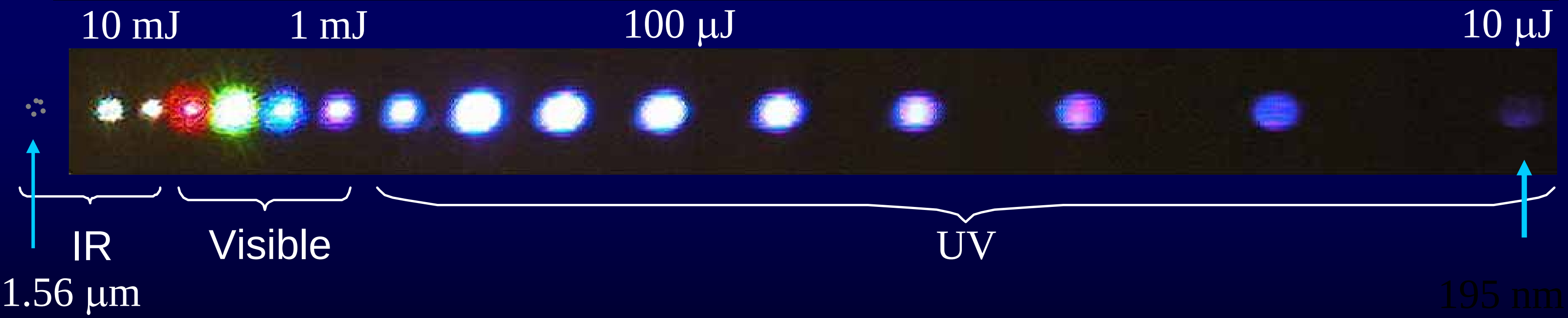


黃書偉

$$I = 10GW/cm^2$$

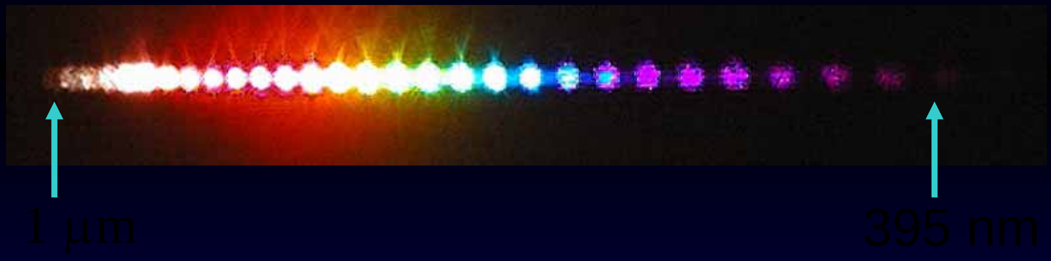
Huang et.al. PRA 74, 063825 (2006)

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

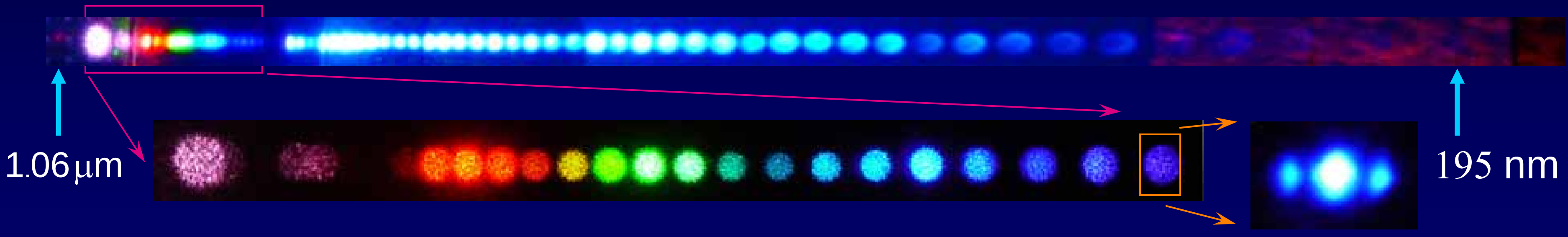


Phys. Rev. A (R)(1997)
Phys. Rev. Lett. 81 (1998)
Opt. Lett. 24 (1999)
Phys. Rev. Lett. 84 (2000)
Phys. Rev. Lett. 85 (2000)
Phys. Rev. A 63 (2001)
Phys. Rev. Lett. 91 (2003)
Phys. Rev. Lett. 93 (2005)
Phys. Rev. Lett. 100 (2008)

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



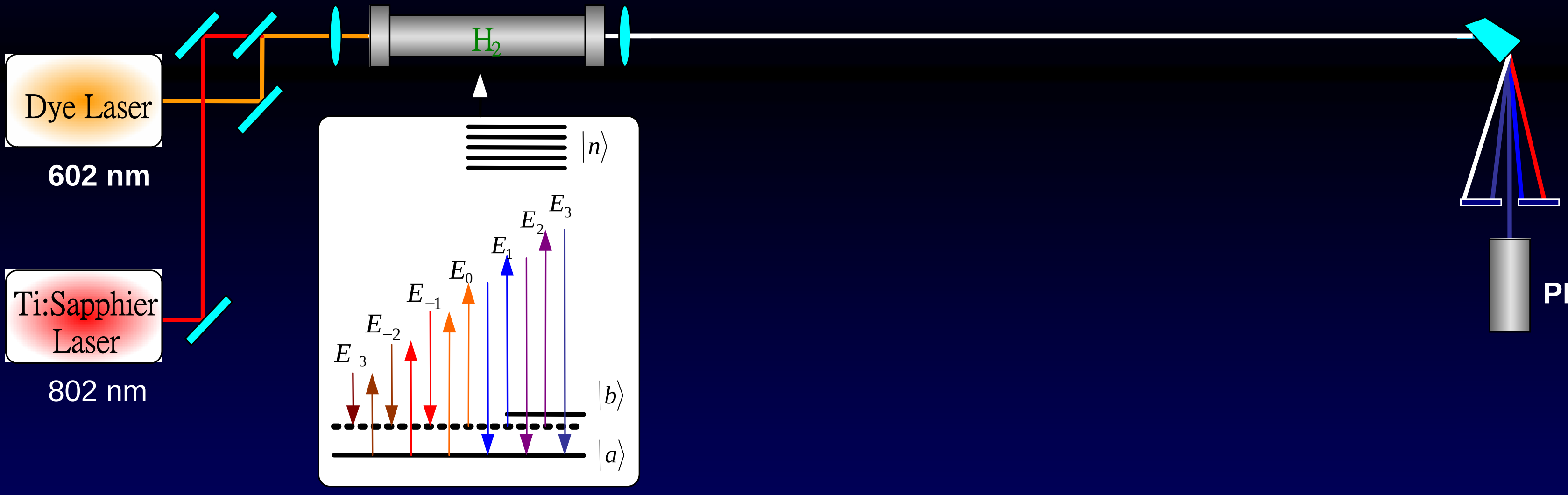
Multiplicative Spectra: ~ 200 sidebands, spaced by < 587 cm^{-1}



What we have done

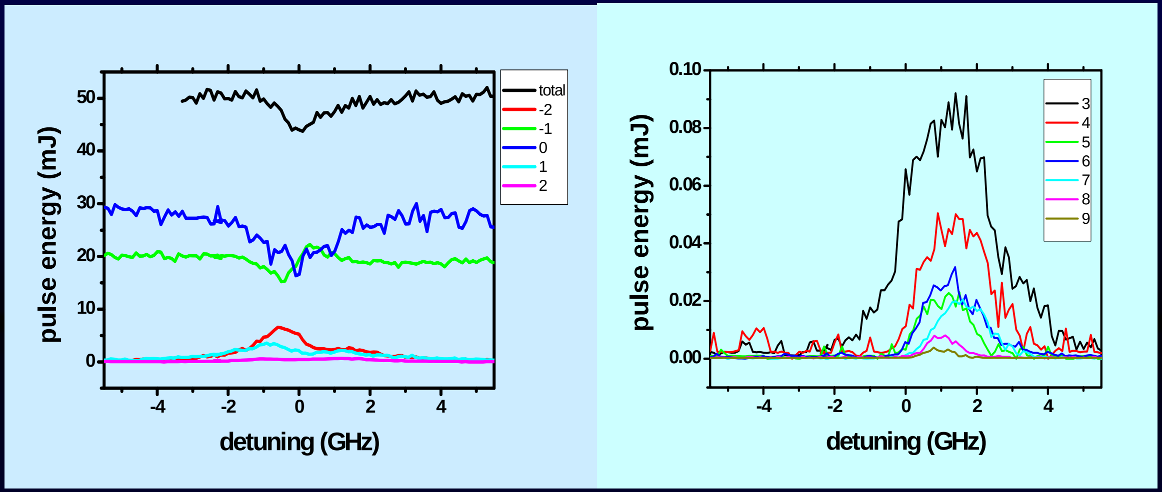
Bandwidth expansion by molecular modulation in room temperature H_2

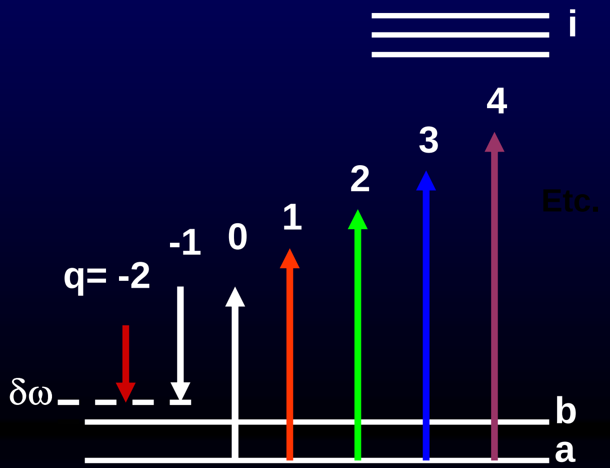
陳蔚然



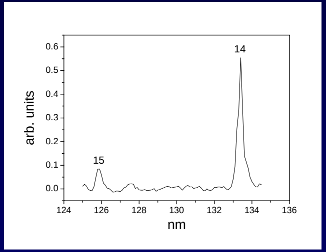
Raman sideband generation

Raman sidebands generated





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

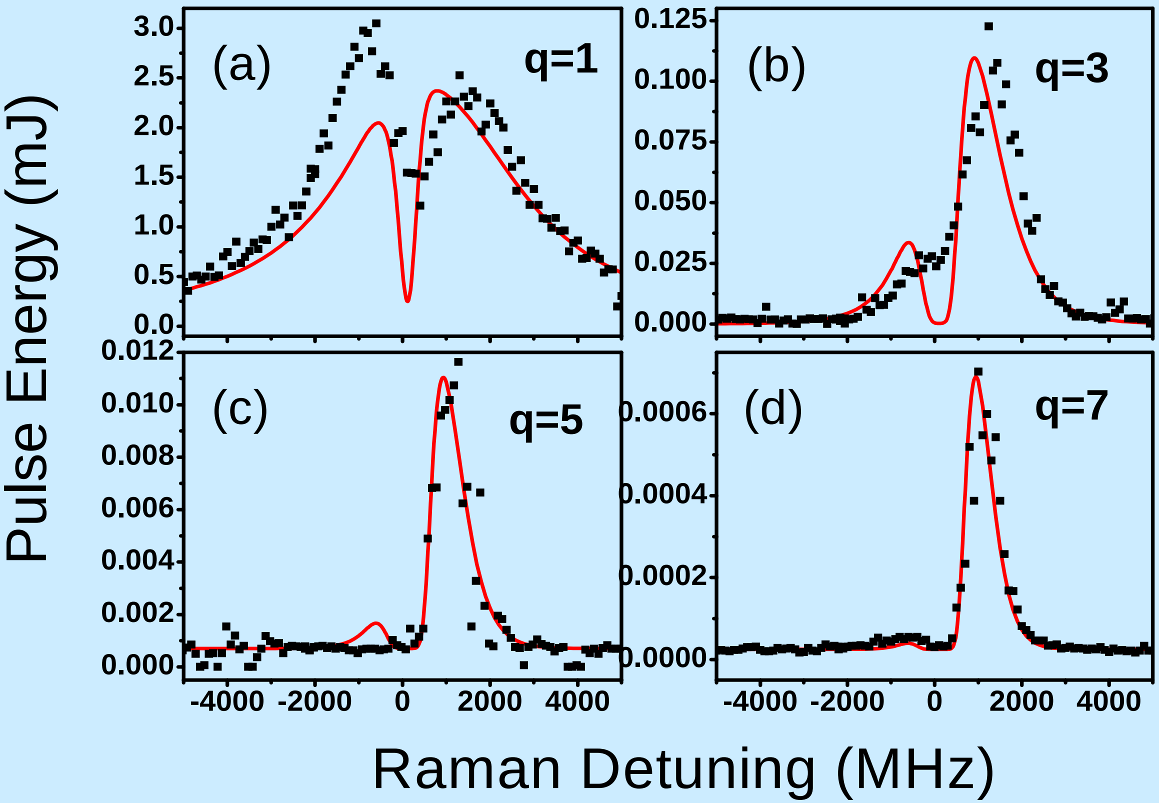


15th order at 126 nm observed

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

PRA 74, 063825 (2006)

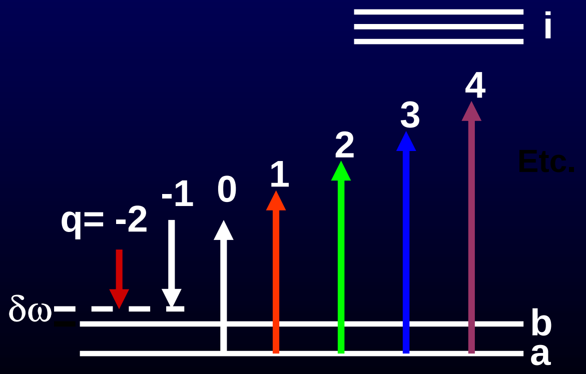
Comparison of experiment with simulation



H_2 pressure, 200 Torr

Multiple quantum paths interference

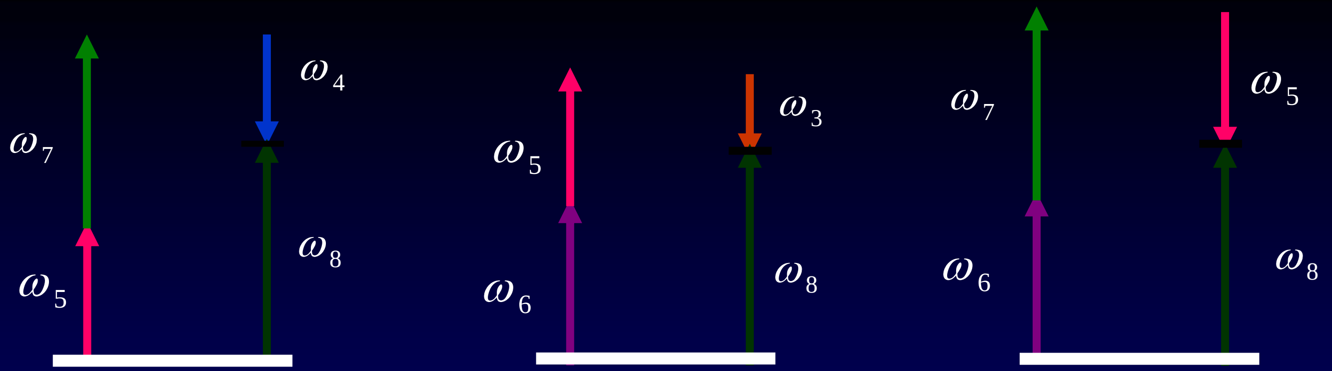
$\omega_q=\omega_0+q\omega_m$ q= -2,-1,0,1,2,3….



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

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

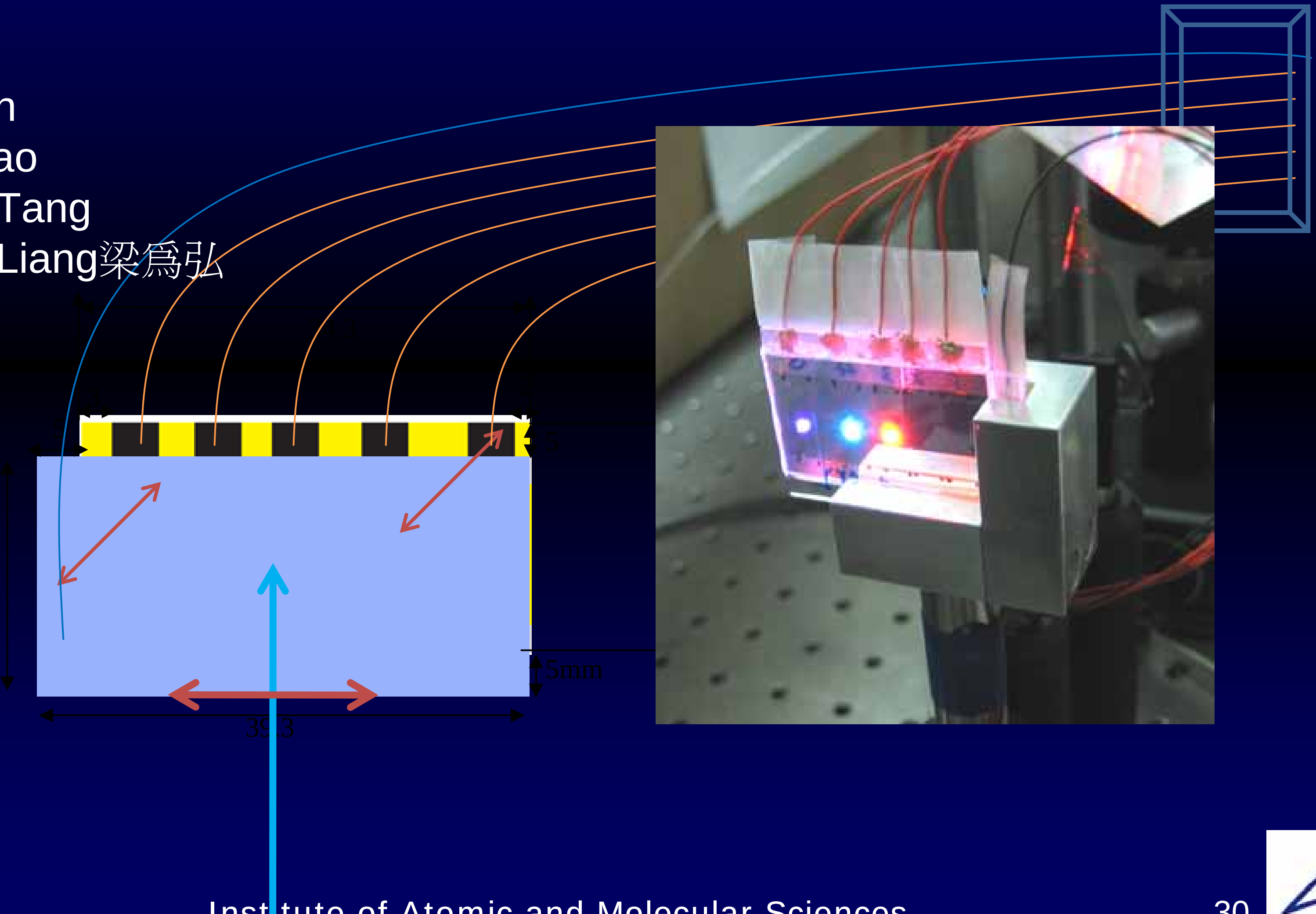
$\omega_6+\omega_7-\omega_5=\omega_8$



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

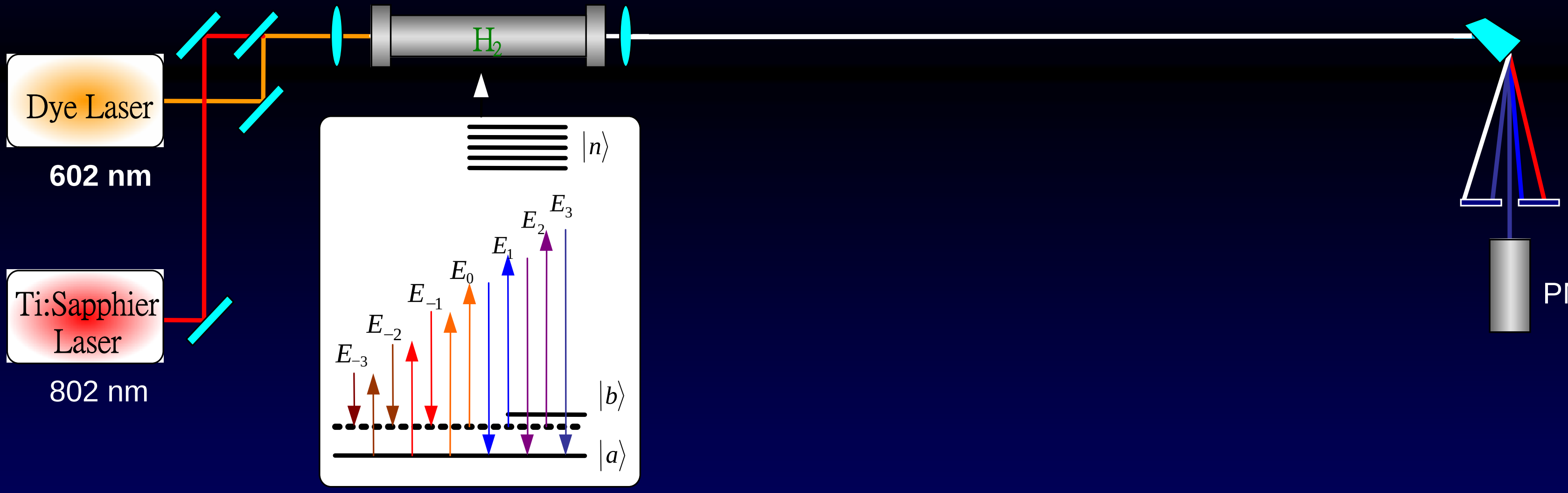
λ_{cutoff}
430 nm --> 380 nm

Ci-Ling Pan
Ru-Pin Chao
Chung-Ta Tang
Wei-Hong Liang梁爲弘



Generating attosecond pulses

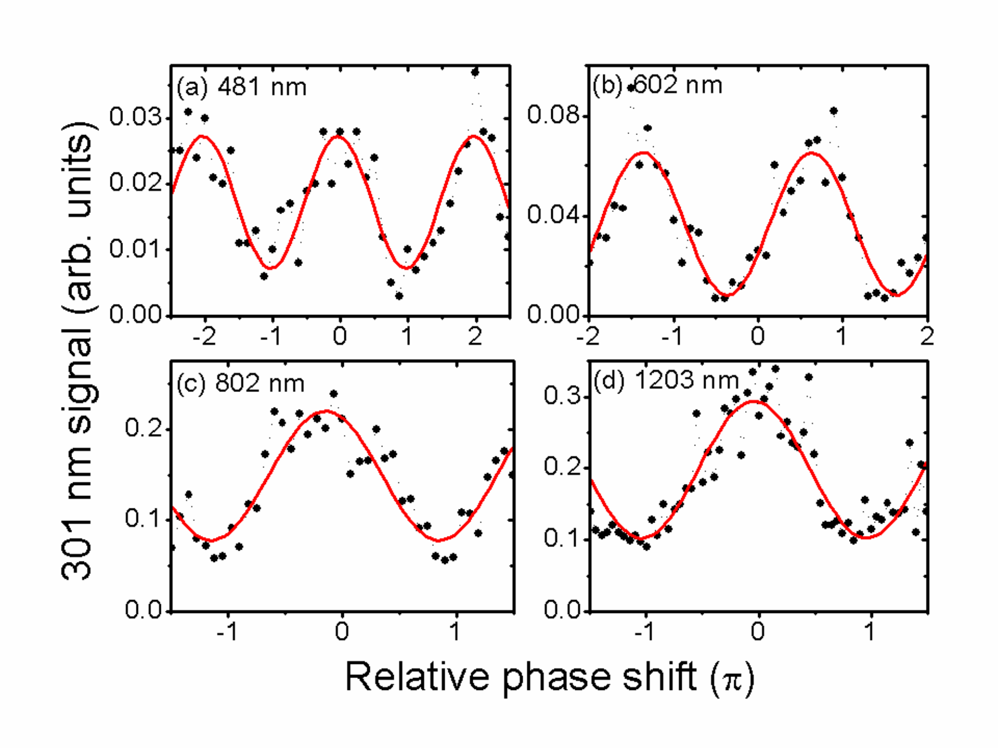
- Bandwidth expansion by molecular modulation



Raman sideband generation

Phase Optimization

Multiple quantum paths interference in four wave mixing



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

Verify single-cycle pulse train with constant CEP

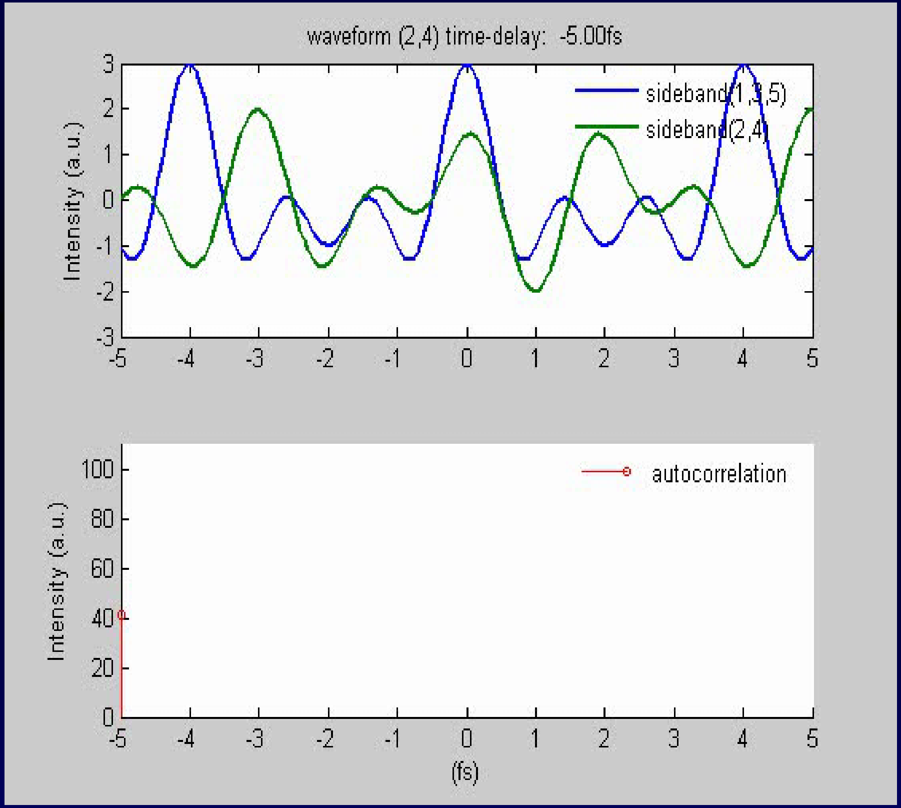
Cross Correlation of Single Cycle Pulse Train

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

Sideband Orders

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.



Carrier-envelope phase

$$E(t) = \sum_n E_n(t) = \sum_n A_n(t) e^{j(\omega_n t + \phi_n)}$$

temporal
envelope

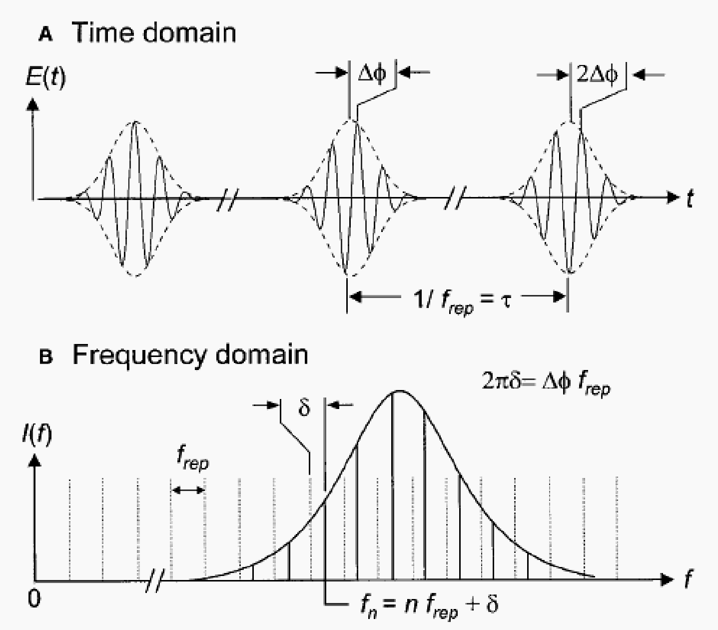
賴建任

In molecular modulation $\omega_n = \omega_{ceo} + n\omega_m$

$$\phi_n = \phi_0 + n\phi_m$$

$$E(t) = e^{j(\omega_{ceo} t + \phi_0)} \sum_n A_n(t) e^{jn\omega_m (t + \phi_m / \omega_m)}$$

$$CEP = \omega_{ceo} t + \phi_0$$



Carrier-Envelope Phase

$$CEP = \Delta\phi = \delta t + \phi_0$$

$$\Delta\phi = \phi_{CE} = absolute.phase$$

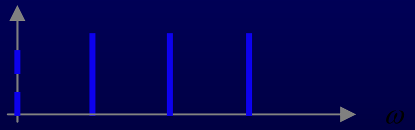
$$\delta t = \phi_{CEO} = CE.offset.phase$$

$$\phi_0 = static.phase$$

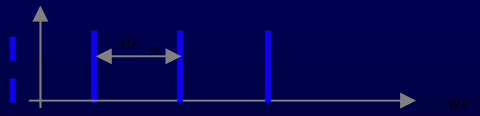
$$\omega_q = n\omega_m$$

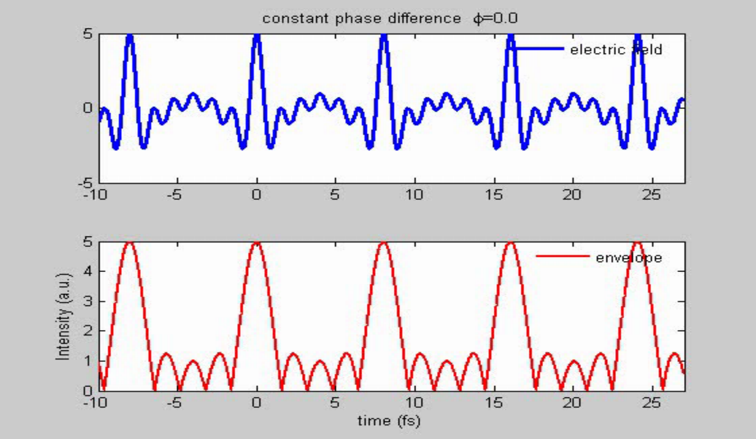
$$\omega_q = \omega_{ceo} + n\omega_m$$

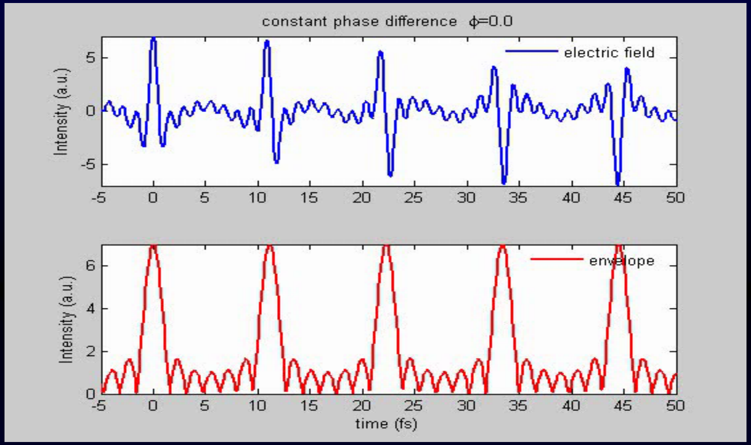
commensurate



incommensurate







Requires that the relative phase between adjacent sidebands be fixed:

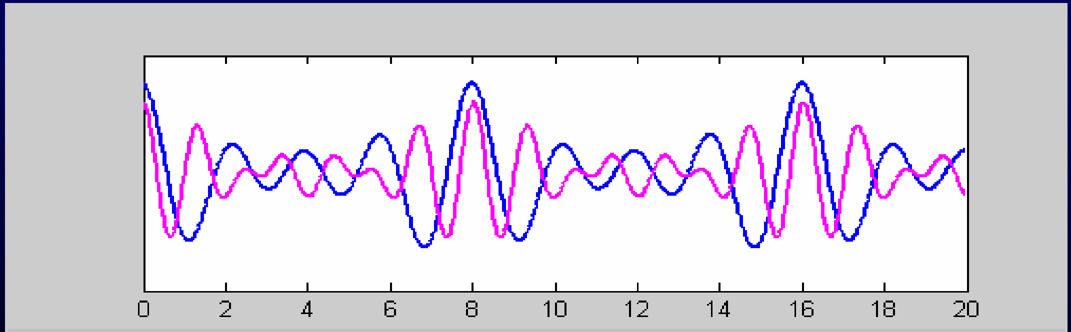
<u>Constant carrier envelope phase</u>

Institute of Atomic and Molecular Sciences
Academia Sinica, Taiwan

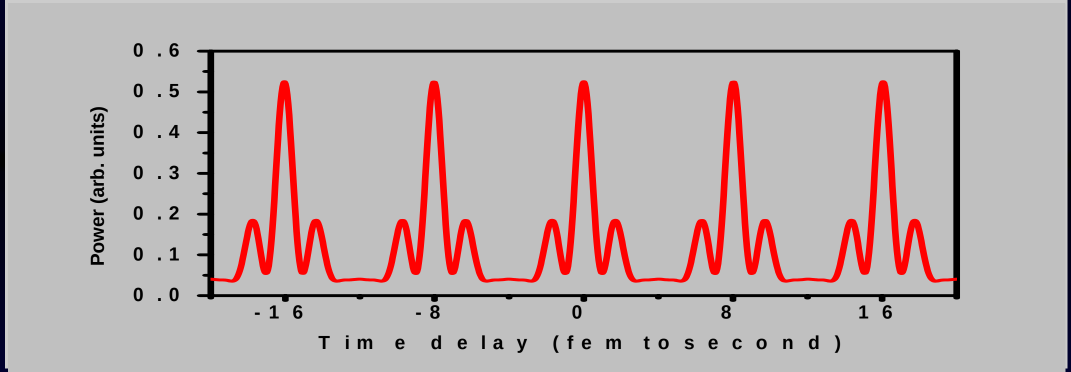
Raman Order	nm	cm^{-1}	4 wave-mixing order
	∞	0	
-3	2406	4155	
-2	1203	8310	1
-1	802	12465	2
0	602	16620	3
1	481	20775	4
2	401	24930	5
3	344	29085	6
4	301	33240	7
5	267	37395	8
6	241	41550	9
7	219	45705	10
8	201	49860	11
9	185	54015	

Cross Correlation of Single Cycle Pulse Train

Sideband Orders



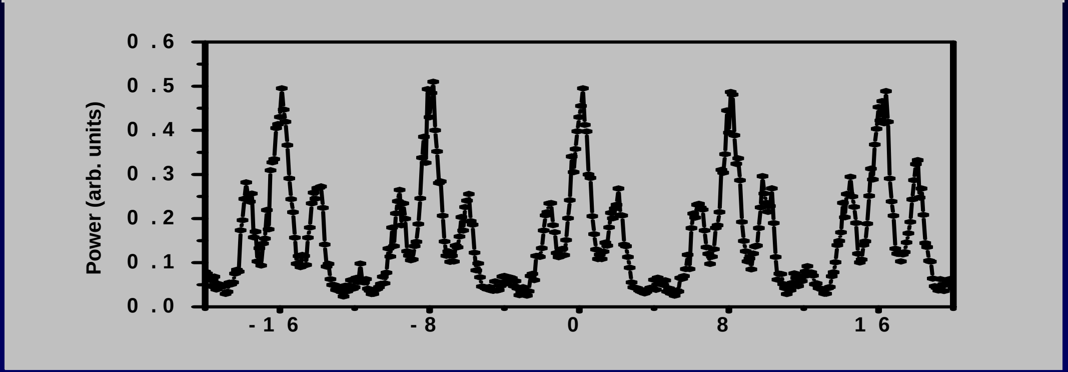
Simulation →



$$f_{rep} = 4155.2 cm^{-1}$$

$$\tau \approx 8\, fs$$

Experiment →



But, although the carrier-envelope offset phase ϕ_{ceo} is zero,

the static phase ϕ_0 varies from one nanosecond laser pulse to the next

In the Raman process, there are two input fields $E_p(t)$ and $E_{p+1}(t)$

$$E_p(t) = A_p(t)e^{j(\omega_p t + \phi_p)}$$

$$E_{p+1}(t) = A_{p+1}(t)e^{j(\omega_{p+1} t + \phi_{p+1})}$$

$$\boxed{CEP = \phi_0 = (p+1)\phi_p - p\phi_{p+1}}$$

For our case, 802 nm $E_3(t) = A_3(t)e^{j(\omega_3 t + \phi_3)}$

602 nm $E_4(t) = A_4(t)e^{j(\omega_4 t + \phi_4)}$

$$CEP = \phi_0 = 4\phi_3 - 3\phi_4$$

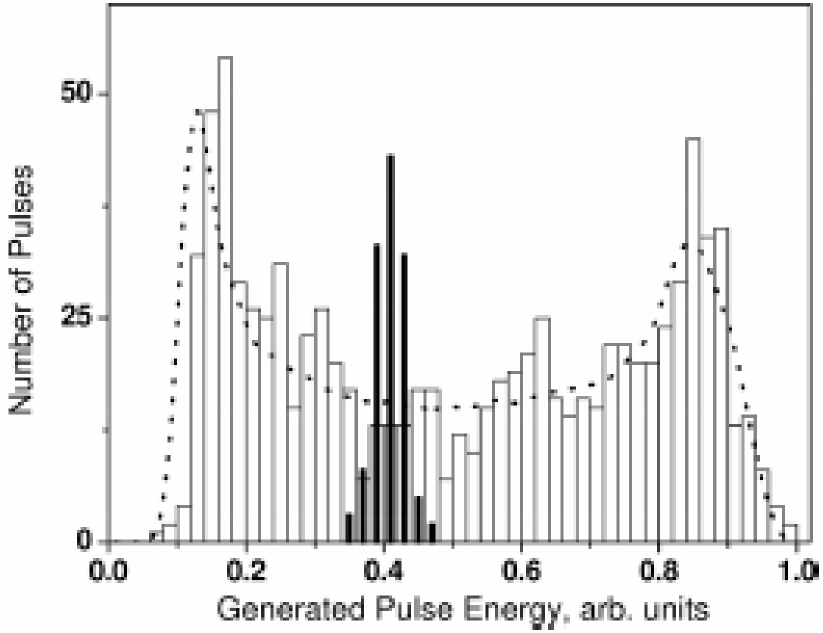


Fig. 3. Histograms of AS 5 pulse energy. Solid black bars: the number of pulses (out of 150) versus AS 5 pulse energy generated with red and IR input beams only. White bars: the histogram of AS 5 pulse energy (913 pulses total) with the addition of the third input beam. The dotted curve is a theoretical prediction obtained assuming perfect single-shot coherence of the two interfering fields and random shot-to-shot variation of their relative phase.

Miaochen Zhi and A. V. Sokolov, OL 32, 2251 (2007)

Our solution: Use the first two terms of the Fourier series

i.e. let p=1

Then, $\phi_m = \phi_2 - \phi_1$

$$\phi_0 = 2\phi_1 - \phi_2$$

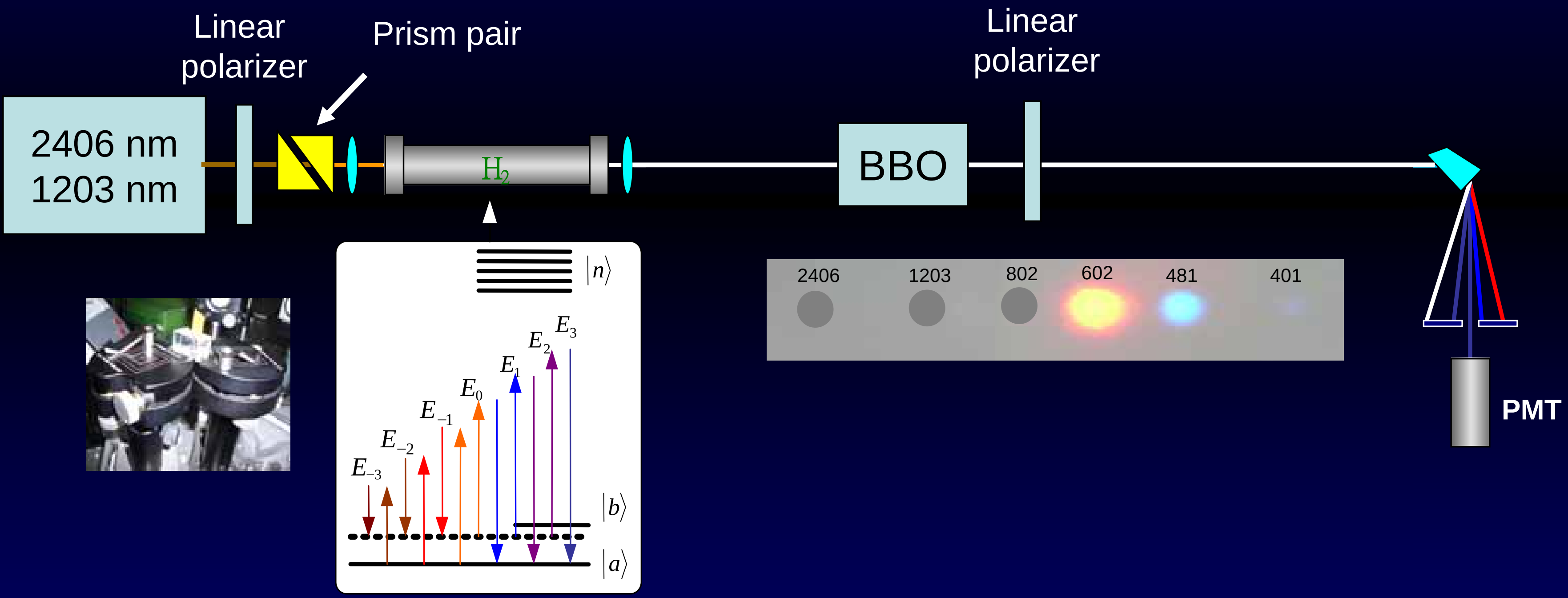
since $\omega_2 = 2\omega_1$ then, $\phi_2 = 2\phi_1 + \xi$

$$\boxed{\begin{aligned} \phi_0 &= 2\phi_1 - \phi_2 \\ &= 2\phi_1 - (2\phi_1 + \xi) \\ &= -\xi \end{aligned}}$$

a constant we can choose and control

need intense 2406 nm source

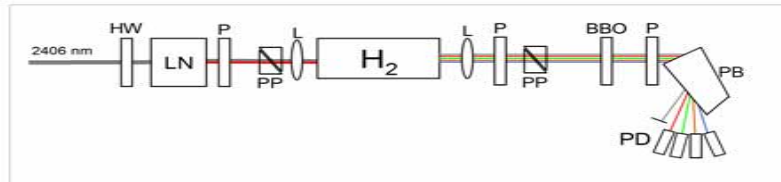
Bottom-up approach to single-cycle pulse generation

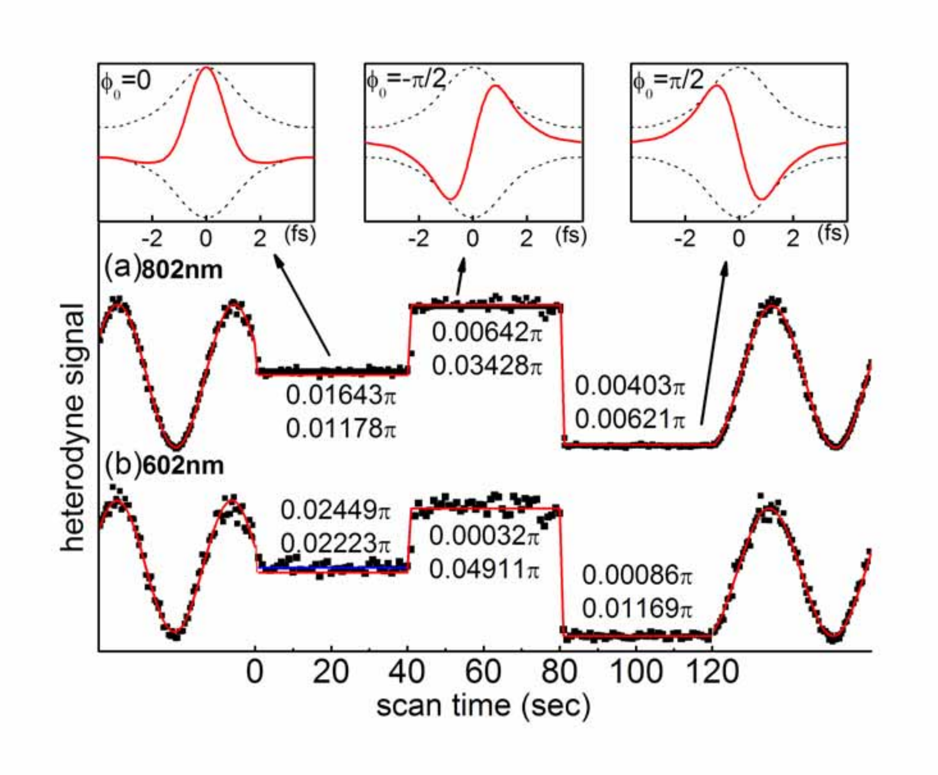


Raman sideband generatio

CEP controlled single-cycle pulses

謝智明
詹翰松

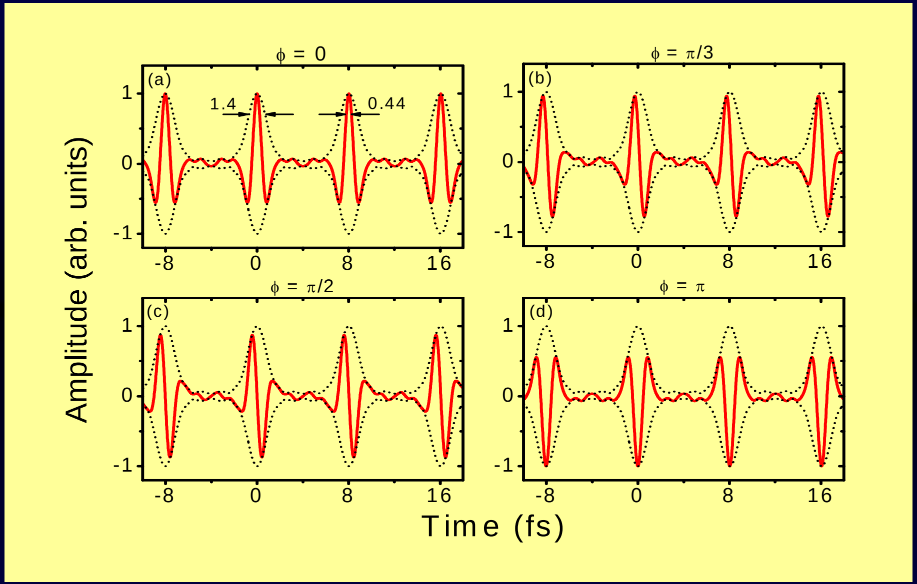


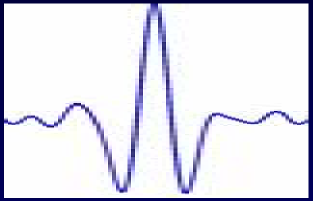


Status of sub-cycle optical pulse generation by molecular modulation

IAMS sub-cycle source

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





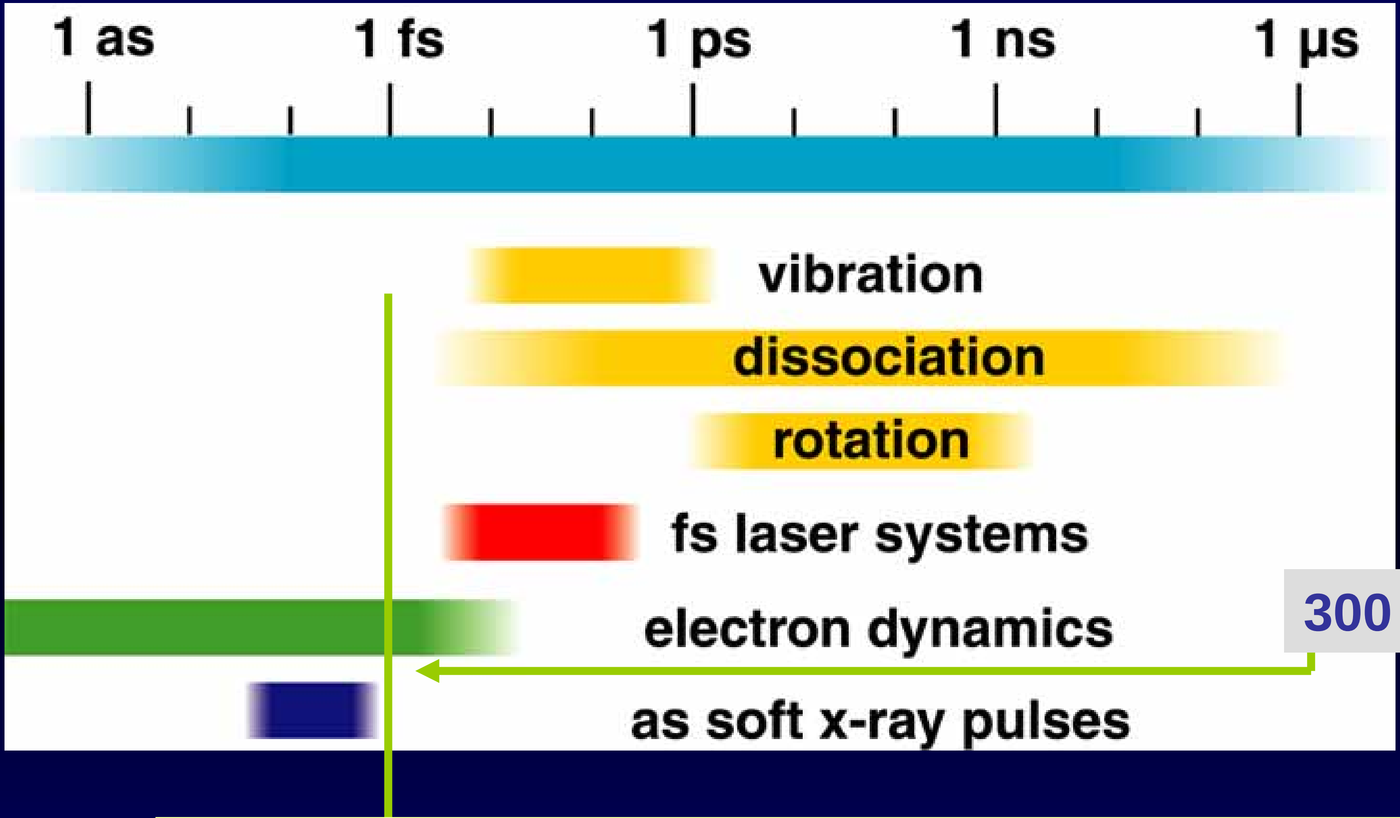
Single Cycle Pulse

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

Chen et.al. PRL 2008
Hsieh et al. submitted to PRL

What to do with these pulses

Time scales



300 nm optical cycle

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

CEP controlled single-cycle pulses: a new tool for science

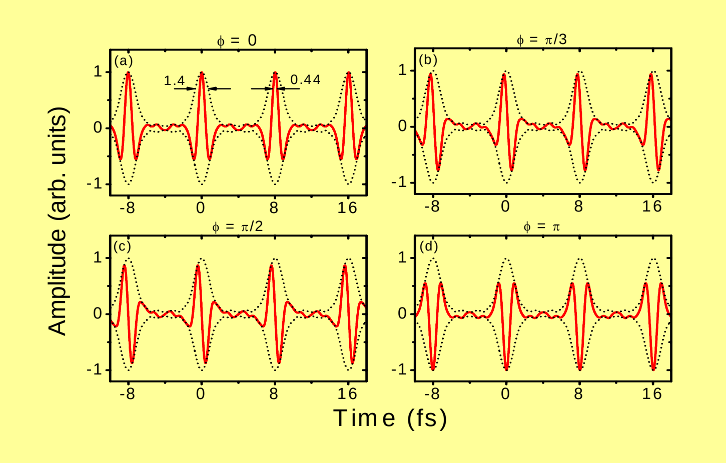
1. Nonlinearities dependent on the instantaneous E field
   “Single cycle physics

2. Quantum interference and coherent control using such

3. Electronic phase controlled excitation vs wavepacket control

4. Optical poling

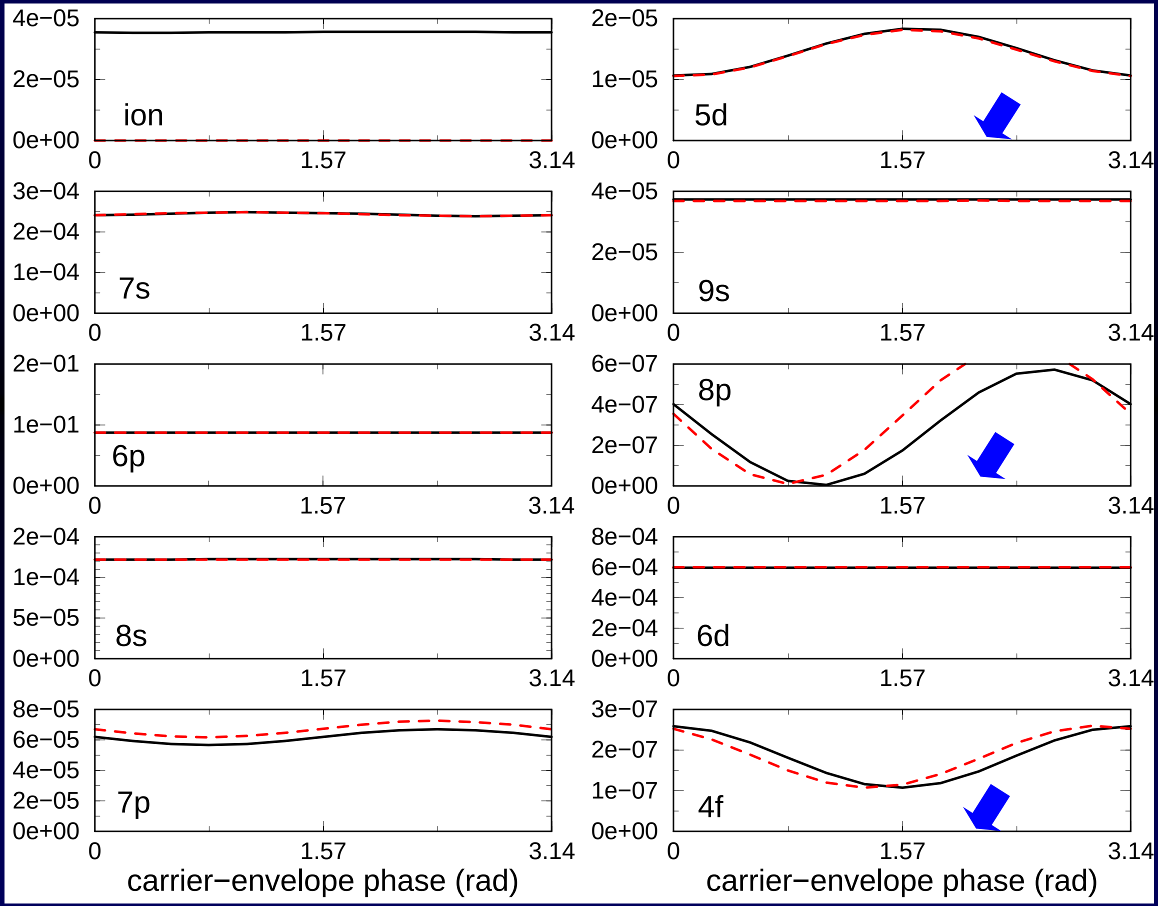
5. QPM x-ray generation

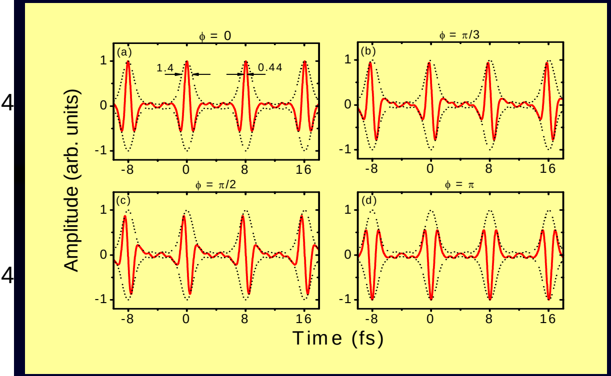


QM Interference on bound state population of Cs

——— **with ionization**
- - - - **without ionization**

1.0 cycle (FWHM) , I=10^{11} W/cm^2





Nakajima and Watanabe, *Phys. Rev. Lett.* **96, 213001 (2006)**

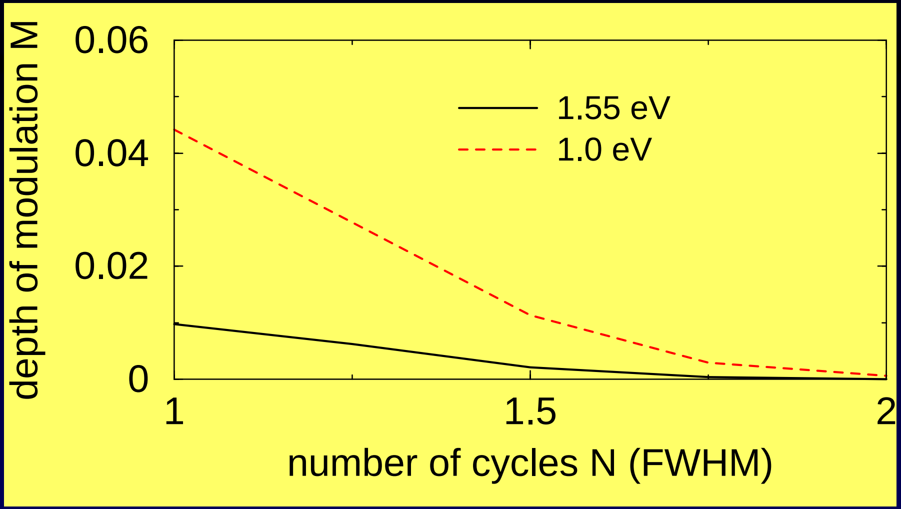
Effect on the Total ionization yield of Cs

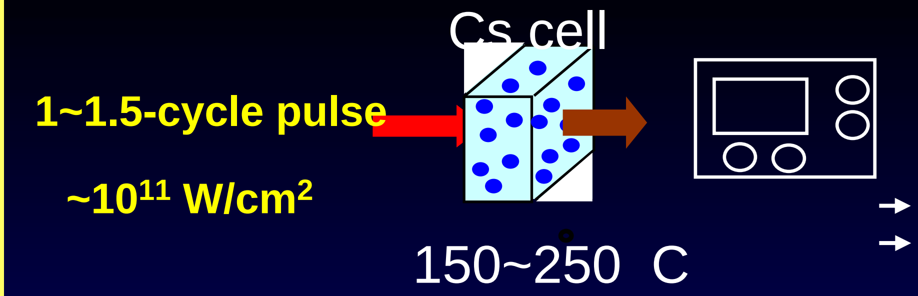
1.0 cycle (FWHM) , I=10^{11} W/cm^2 (γ =18)

ion yield

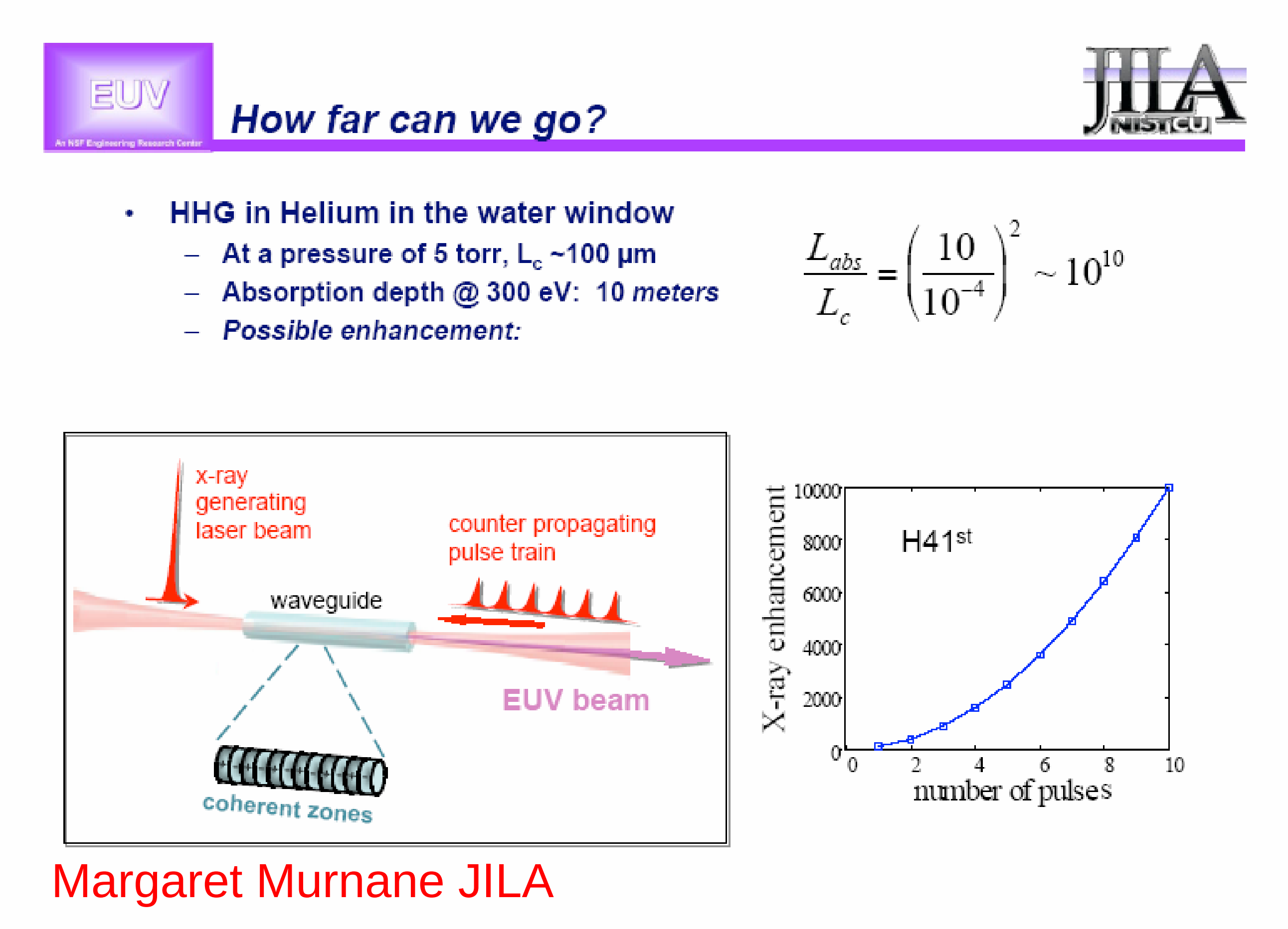
Depth of modulation $$M = \frac{Y(\phi_{\max}) - Y(\phi_{\min})}{Y(\phi_{\max}) + Y(\phi_{\min})}$$

$$0 \le M \le 1$$





QPM in x-ray generation



EUV

An NSF Engineering Research Center

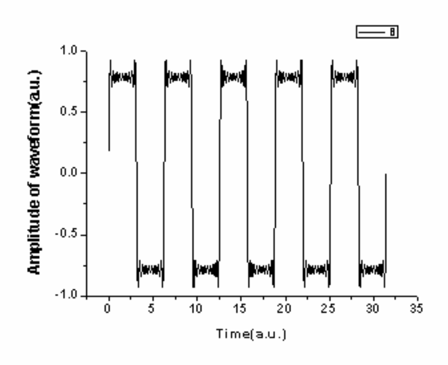
How far can we go?

JILA NIST CU

- **HHG in Helium in the water window**
  - **At a pressure of 5 torr, L_c ~100 μm**
  - **Absorption depth @ 300 eV: 10 *meters***
  - ***Possible enhancement:***

$$\frac{L_{abs}}{L_c} = \left(\frac{10}{10^{-4}}\right)^2 \sim 10^{10}$$

Margaret Murnane JILA



Collaborators

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Ru-Pin Chao-Pan NCTU
Chao-Kuei Lee NSYSU


陳蔚然


謝智明


賴建任


詹翰松

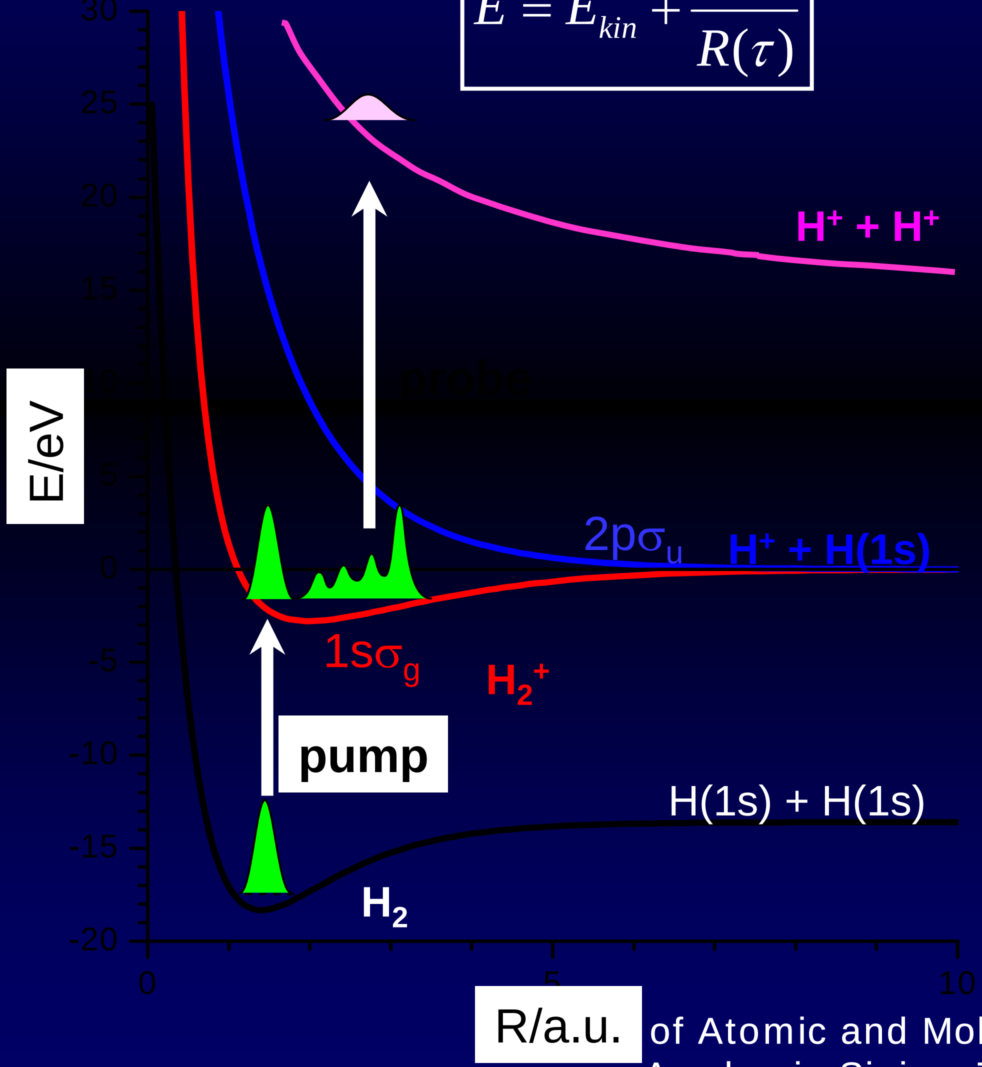

黃書偉

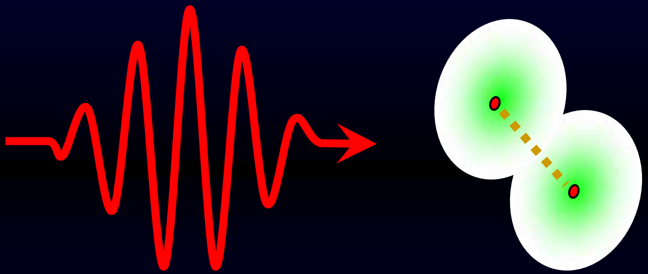

梁爲弘

Bound wave packets

$$E = E_{kin} + \frac{1}{R(\tau)}$$

Pulses shorter than the vibrational period are needed!

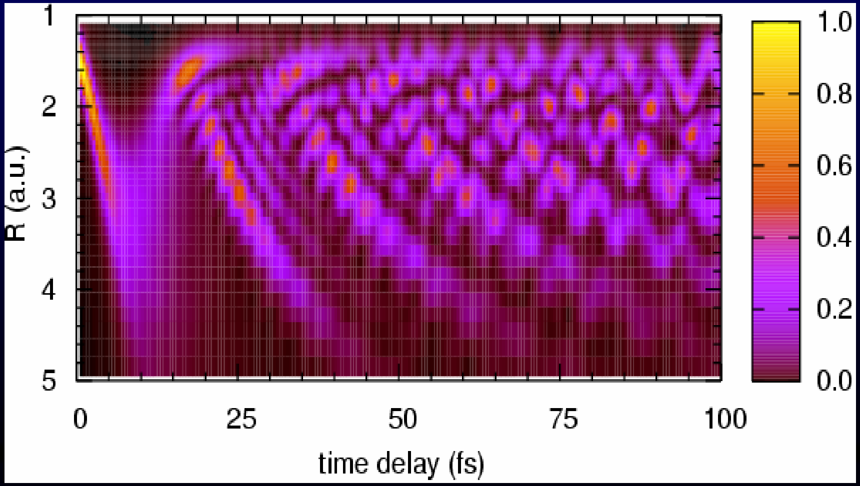




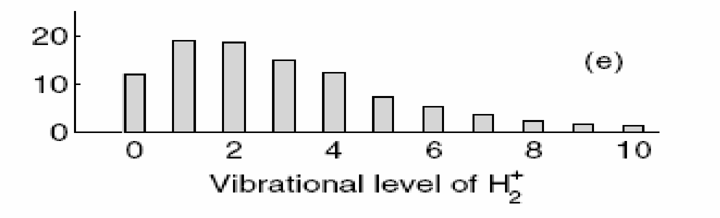
1) Single ionization

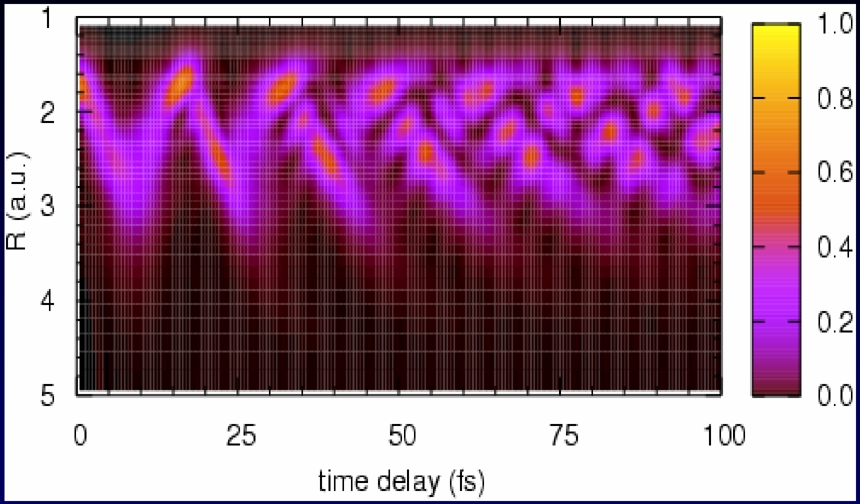
2b) Oscilations in the $1s\sigma_g$ potential

Time evolutions of two initial wave packets

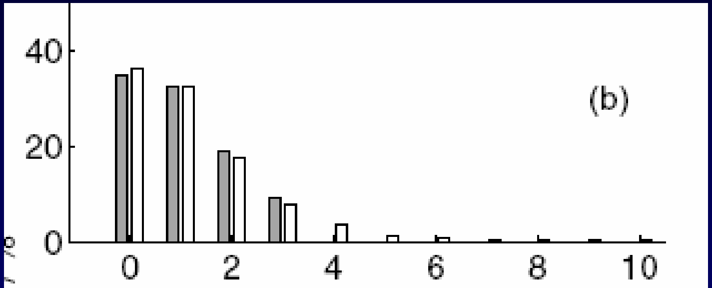


Franck-Condon (FC) initial vibrational distr.





Initial non-FC vibrational distribution



Molecular Orientation
CEP controlled photonics

Organic thin film

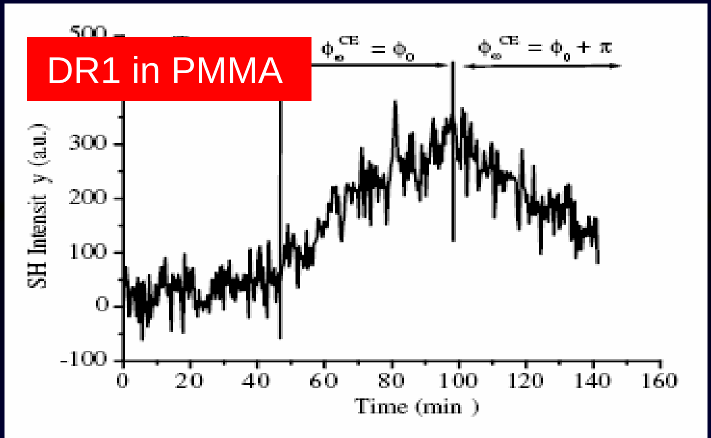
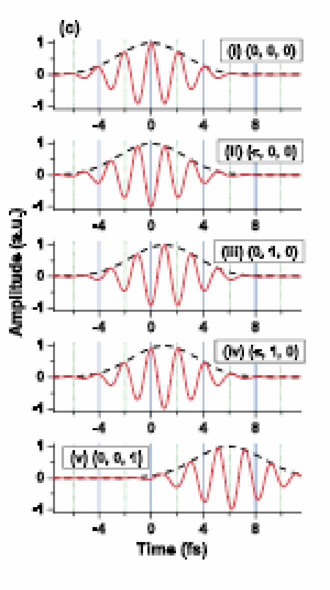

DR1 in PMMA

FIG. 3. Real-time evolution of the SH signal with fixed CE phases.



semiconductors

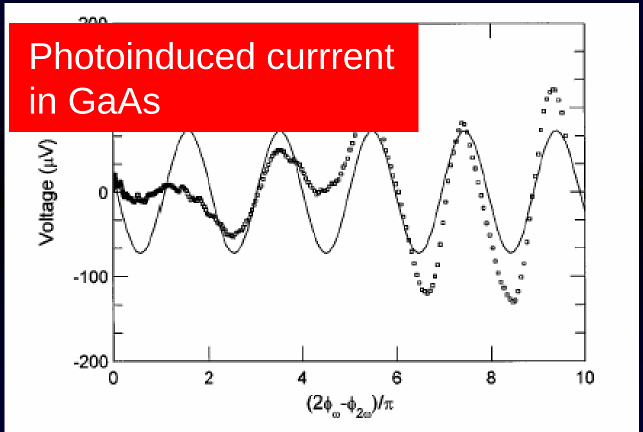

Photoinduced currrent in GaAs

FIG. 3. Induced coherently controlled current signature as a function of $\Delta\phi$ on a 10 μm gap MSM on normal GaAs using the OPG. The solid curve is the best fit for a constant amplitude sine function.

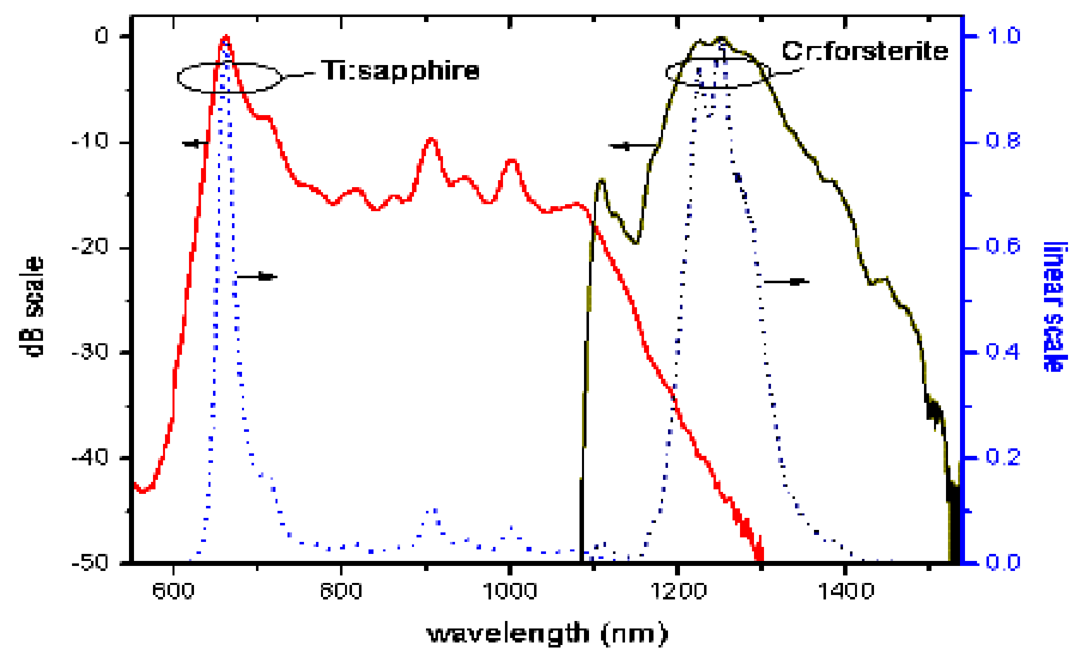
Kobayashi, PRL 94, 153903 (2005)

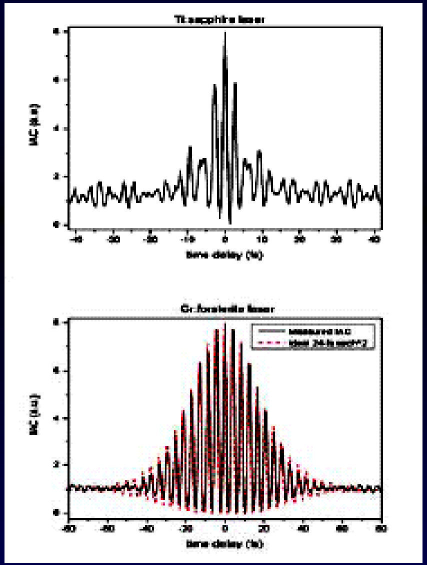
Van Dreel, PRL 78, 306 (1997)

Generating attosecond pulses

A

Modelocking extremely broadband lasers

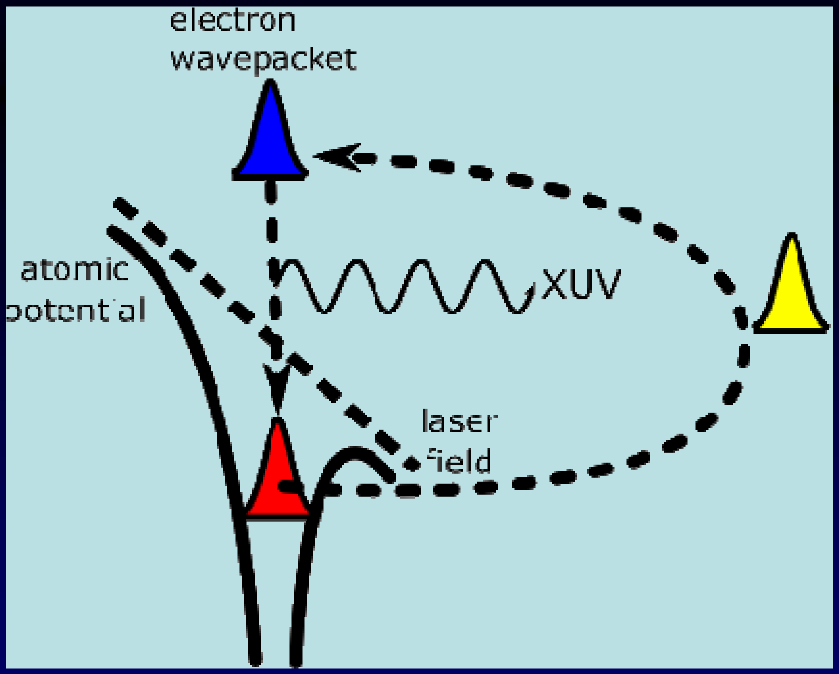




Franz Kartner, MIT

Generating attosecond pulses

B High-order harmonic generation



Tunnelling

Acceleration in the continuum

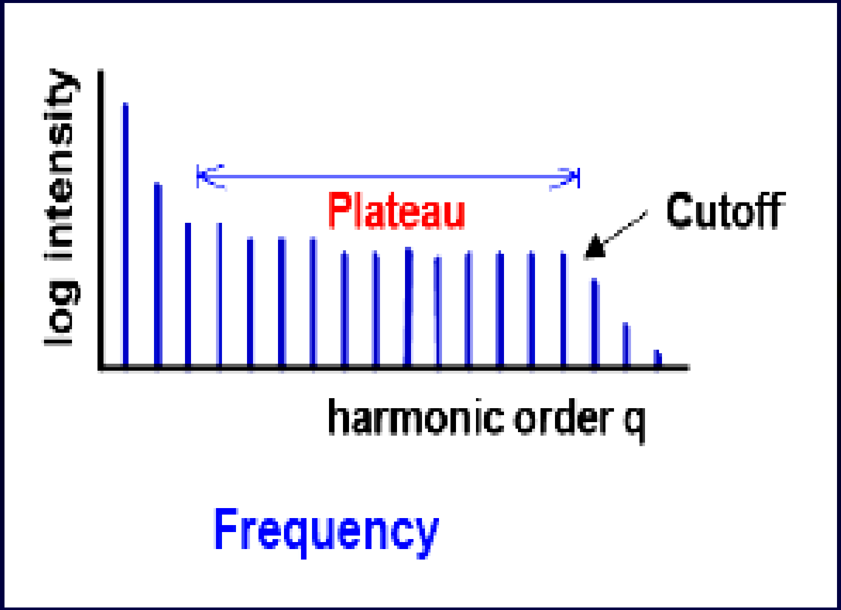
Recombination

Corkum, Kulander and Schafer, Becker, Muller, Lewenstein 1993

Generating attosecond pulses

B

High-order harmonic generation



$$h\upsilon_{cutoff} = I_p + 3.2U_p$$

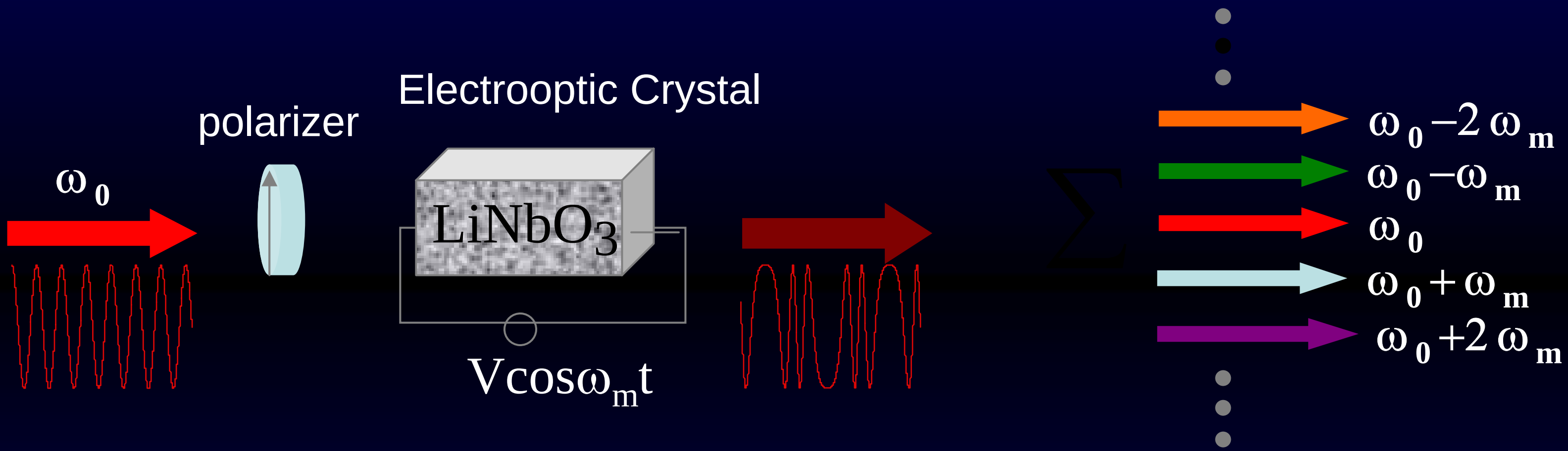
$$U_p = e^2 E_a{}^2 / 4m\omega^2$$

$$I \sim 3\times10^{14} W / cm^2$$

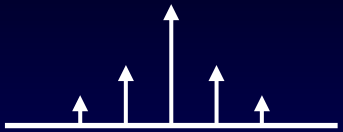
Advantages: shortest attosecond pulses
isolated pulse or pulse train
optics available

Disadvantages: 30 to >100 eV photons
pulse train spacing 2 fs.
low power

Bandwidth expansion by molecular modulation



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



$$\omega_q = \omega_0 + q\omega_m$$

q^th sideband

modulation frequency