



Optical Waveform Synthesis

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AMO Summer School

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Why we are interested in it - motivation

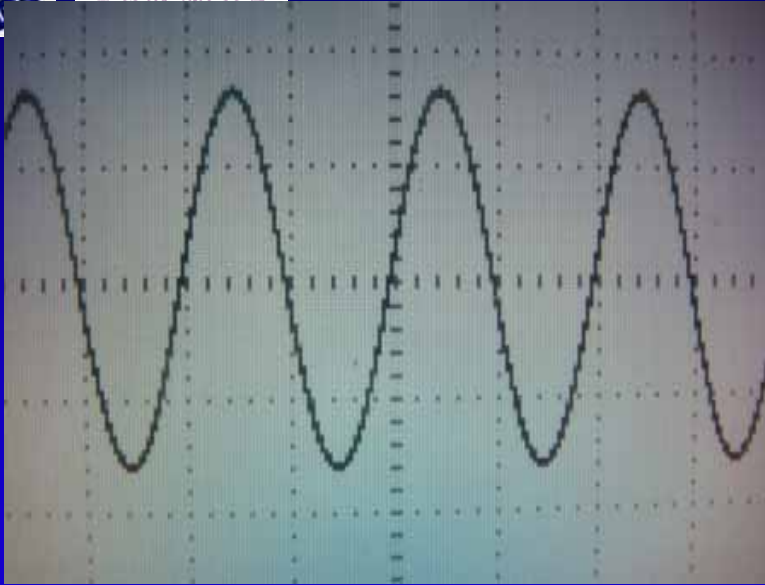
What is the approach - basic concept

How it is done and what have been done - results

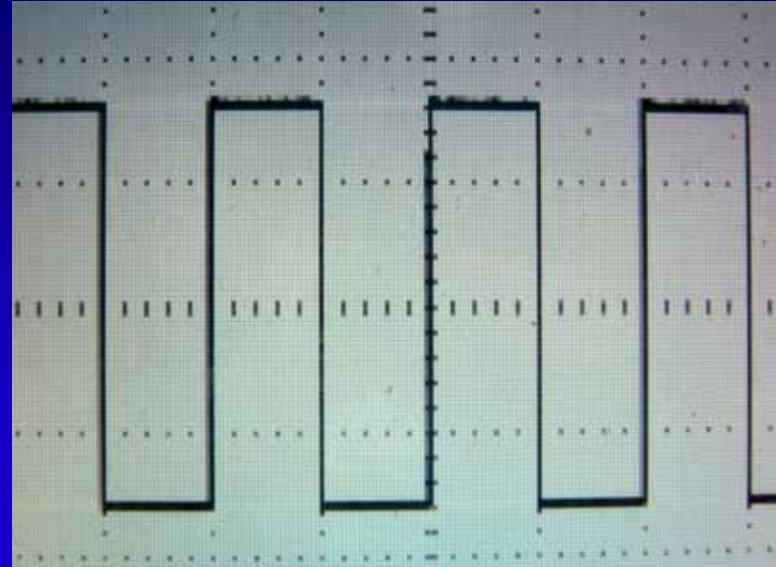
What can be done with these waveforms - application



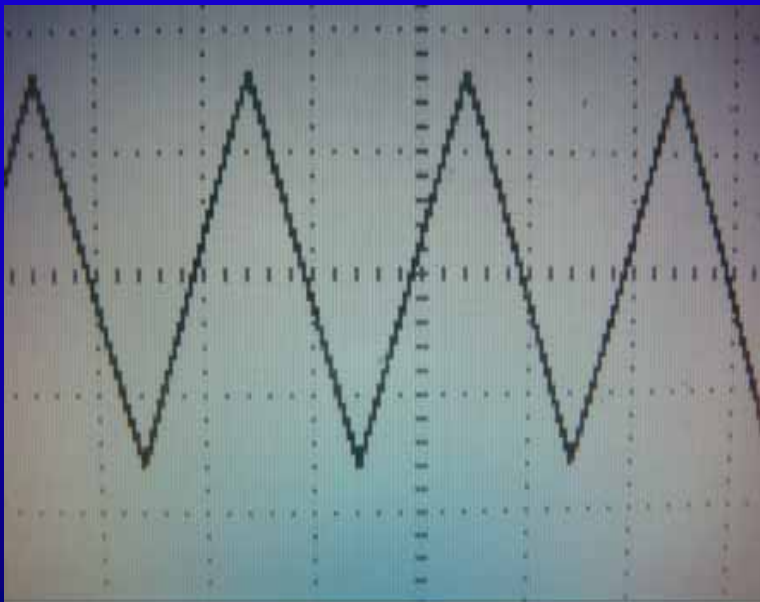
A function generator is used in many physical science laboratories to generate a wide variety of synthesized electrical signals such as the sine, square, triangle and ramp functions. These signals are used for testing and diagnostic applications.



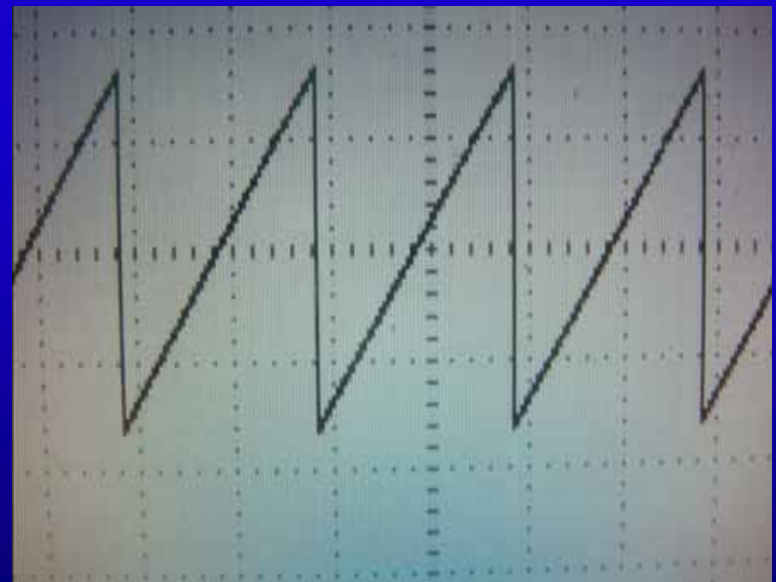
sine



square



triangle



sawtooth



Patented Jan. 6, 1942

2,268,872

UNITED STATES PATENT OFFICE

2,268,872

VARIABLE FREQUENCY OSCILLATION GENERATOR

William R. Hewlett, Palo Alto, Calif., assignor to Hewlett-Packard Company, Palo Alto, Calif., a co-partnership consisting of William R. Hewlett and David Packard

Application July 11, 1939, Serial No. 283,801

Jan. 6, 1942.

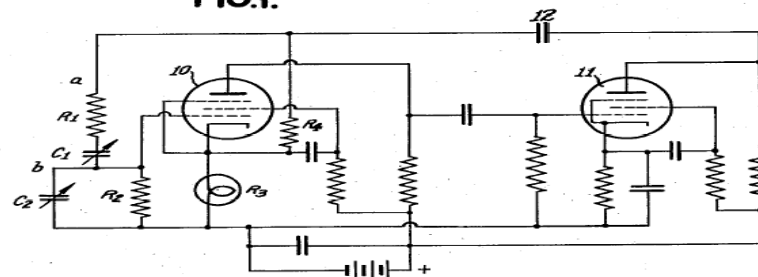
W. R. HEWLETT

2,268,872

VARIABLE FREQUENCY OSCILLATION GENERATOR

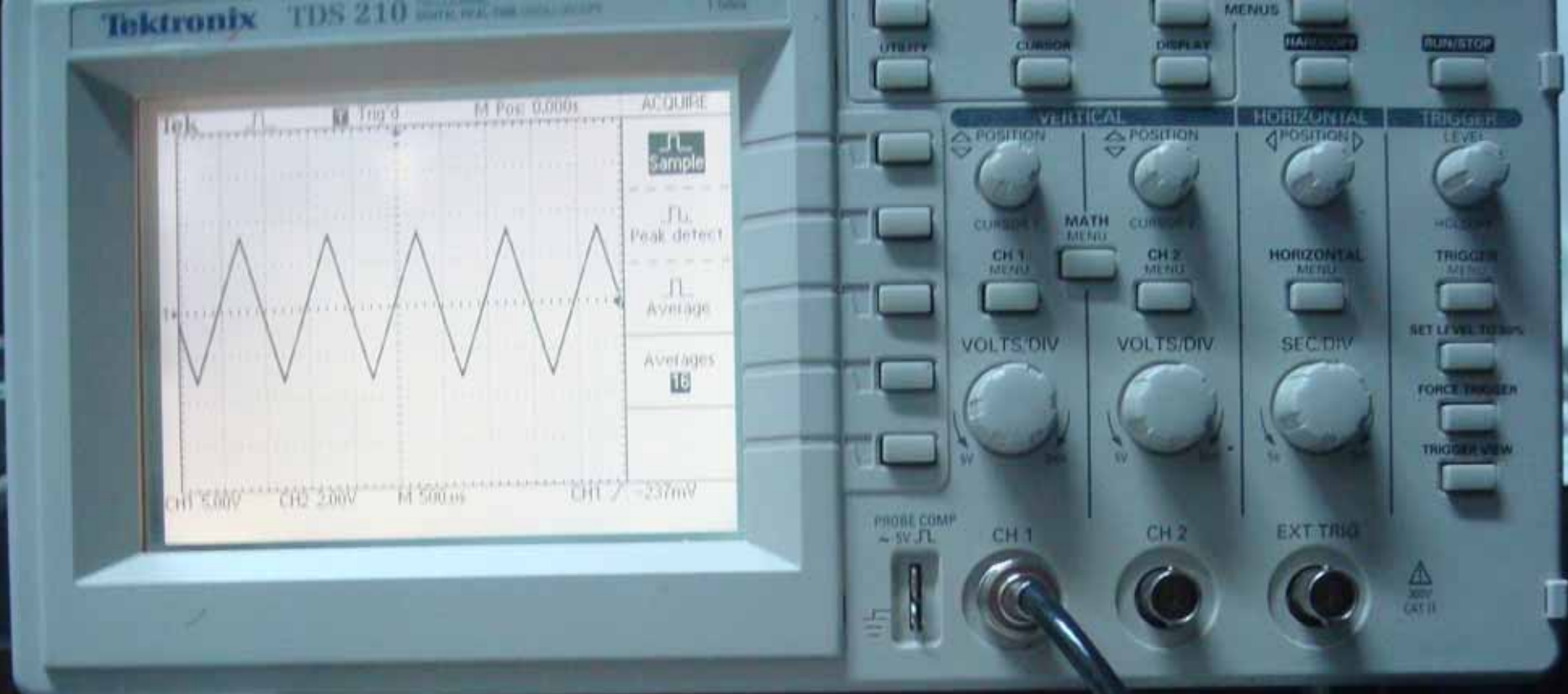
Filed July 11, 1939

FIG. 1.



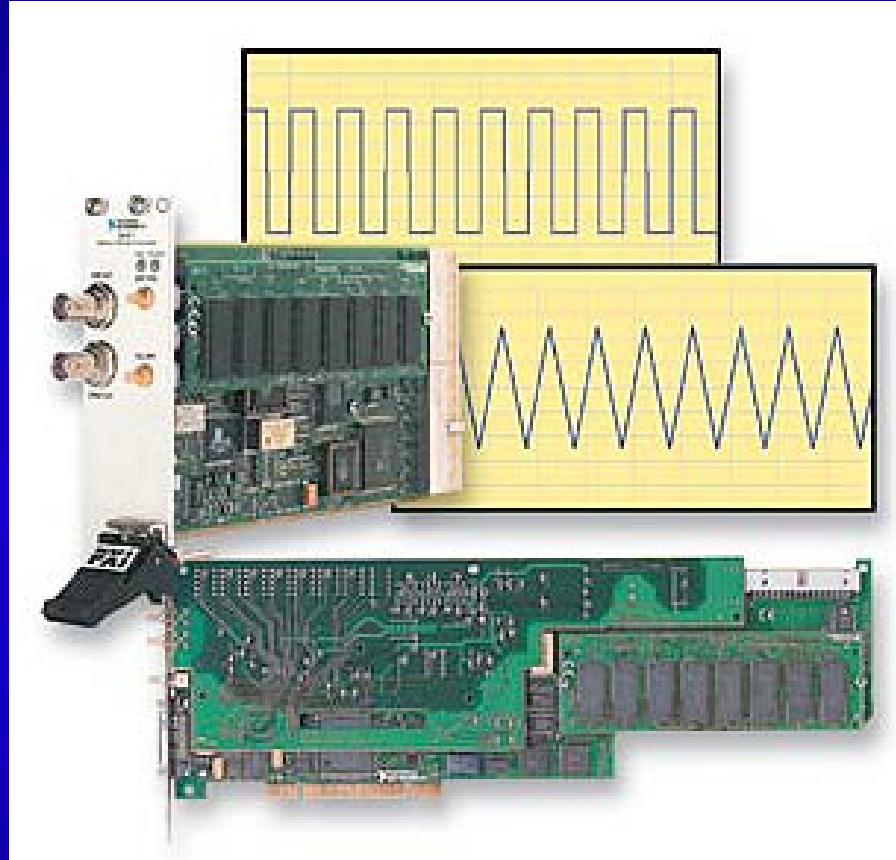
HP 200A







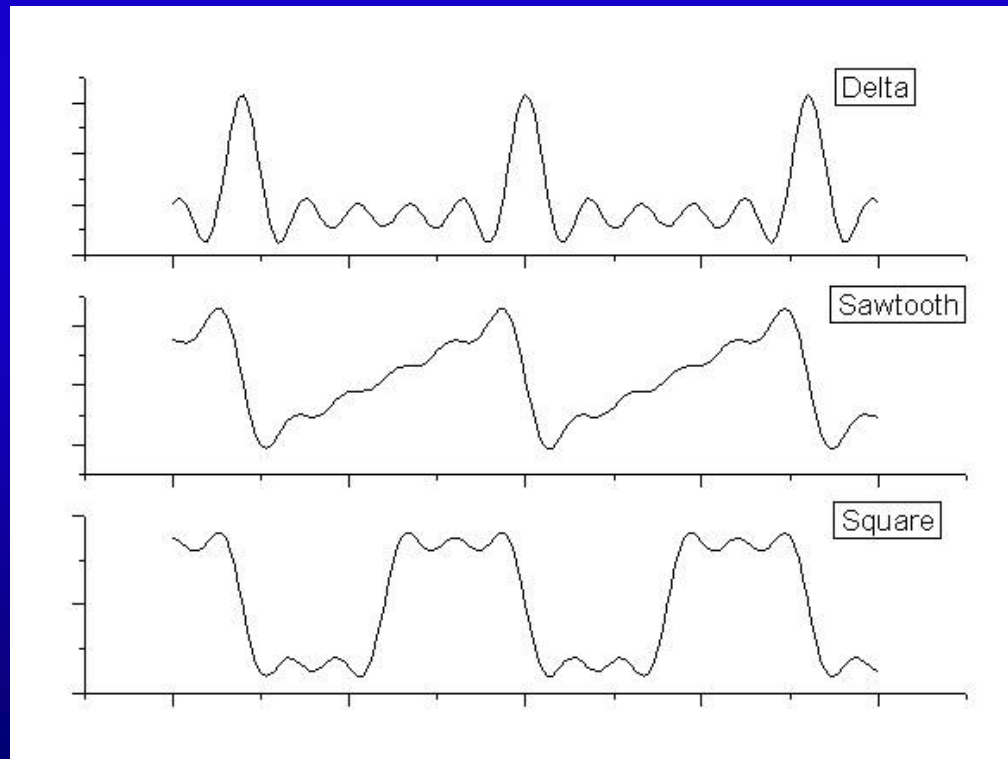
RF and Microwave function generators



Their speed are limited by the speed of IC circuits to $<100\text{GHz}$.



Synthesize various forms of ultrashort pulses in the optical frequency (10^{15} cps) regime





atto- [A toh]

(Danish or Norwegian: eighteen; a decimal prefix used in the international metric system for measurements)

This prefix is used in the metric [decimal] system as quintillionth [U.S.] and trillionth part [U.K.], 10^{-18} [0.000 000 000 000 000 001]. The metric symbol for **atto-** is **a**.

Prefix for small and large numbers:

micro 10^{-6}

mega 10^6

nano 10^{-9}

giga 10^9

pico 10^{-12}

tera 10^{12}

femto 10^{-15}

peta 10^{15}

atto 10^{-18}

exa 10^{18}

zepto 10^{-21}

zetta 10^{21}

yotta 10^{-24}

yocto 10^{24}





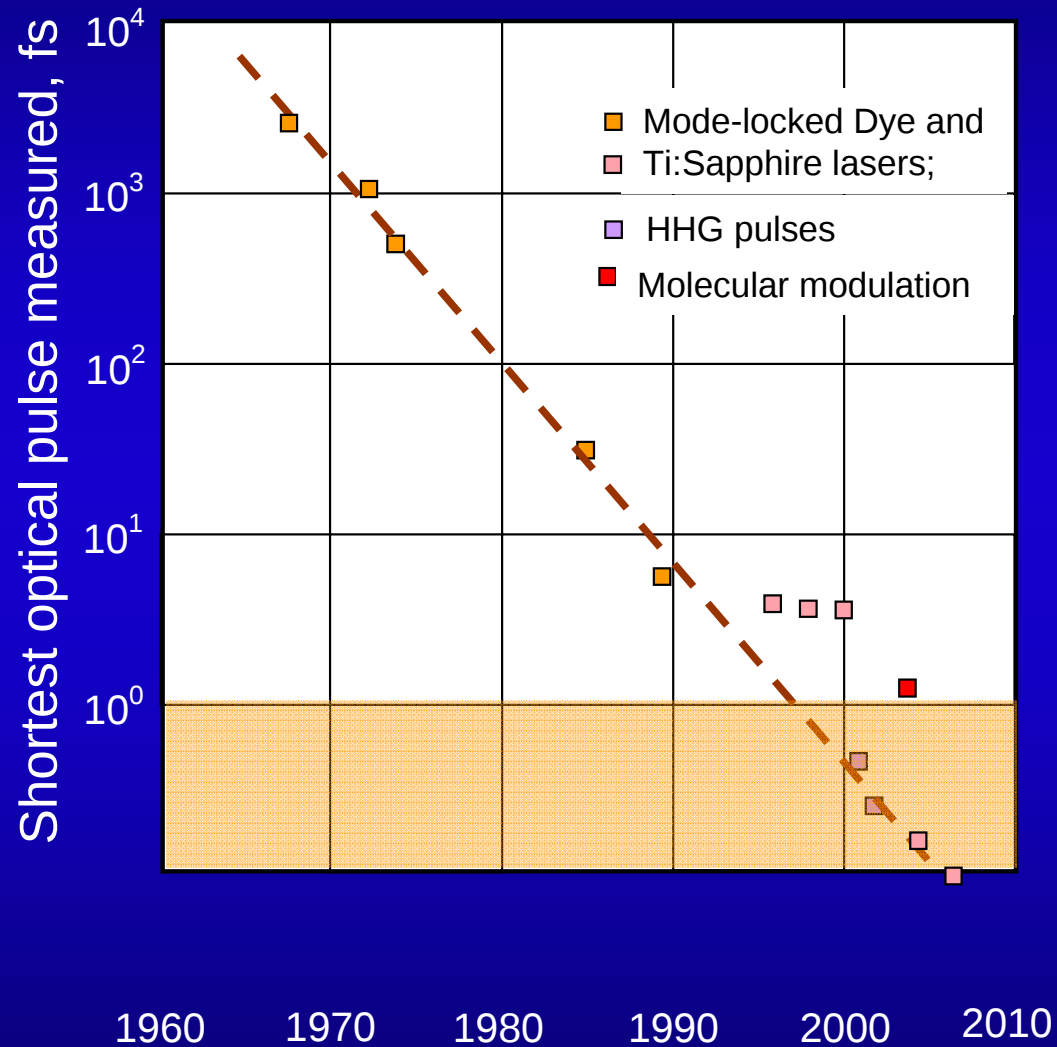
$$300 \text{ nm} = 1 \text{ fs}$$

single-cycle pulse $\equiv$ attosecond pulse

Frontier in optical physics

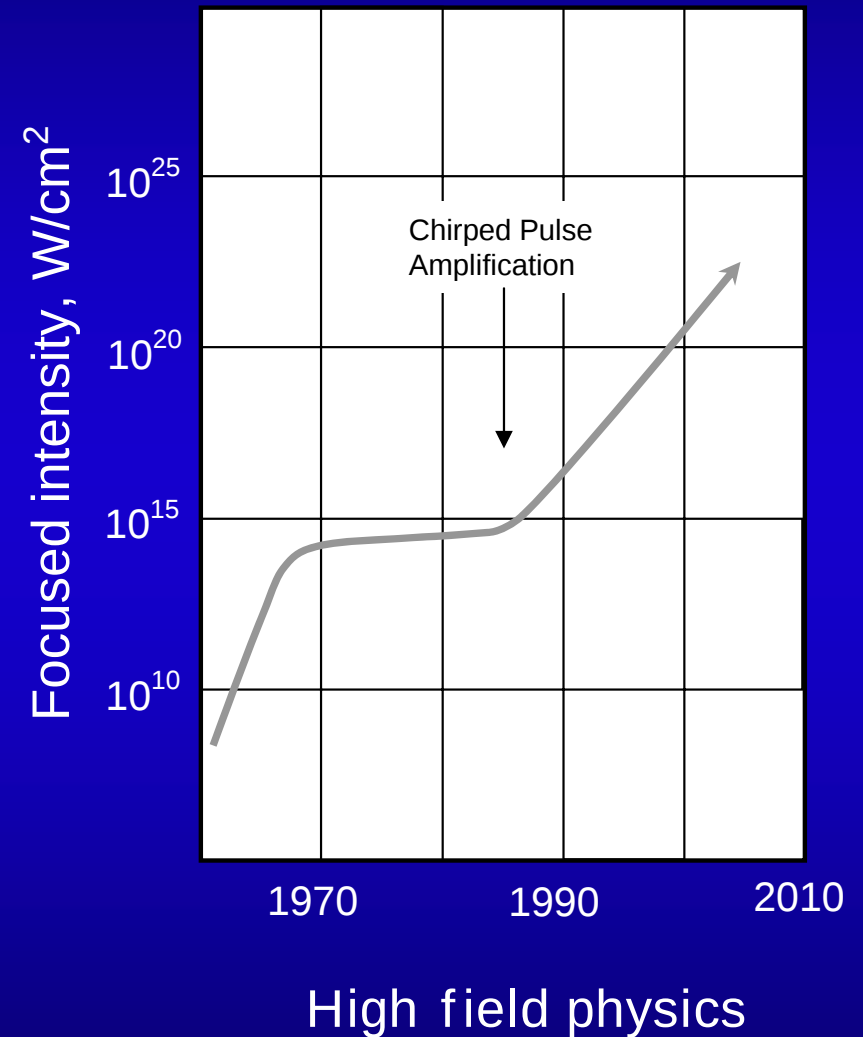


Laser pulses get shorter
over the years



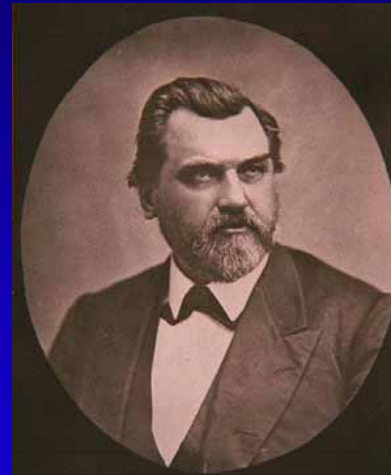
Ultrafast science

Peak intensity
increased

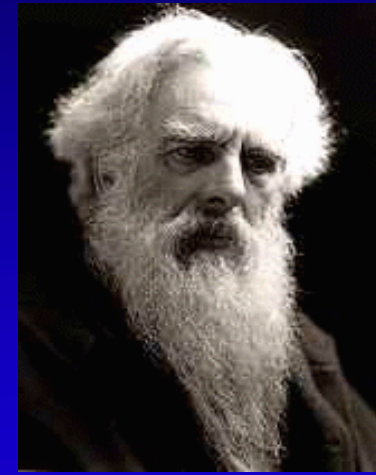




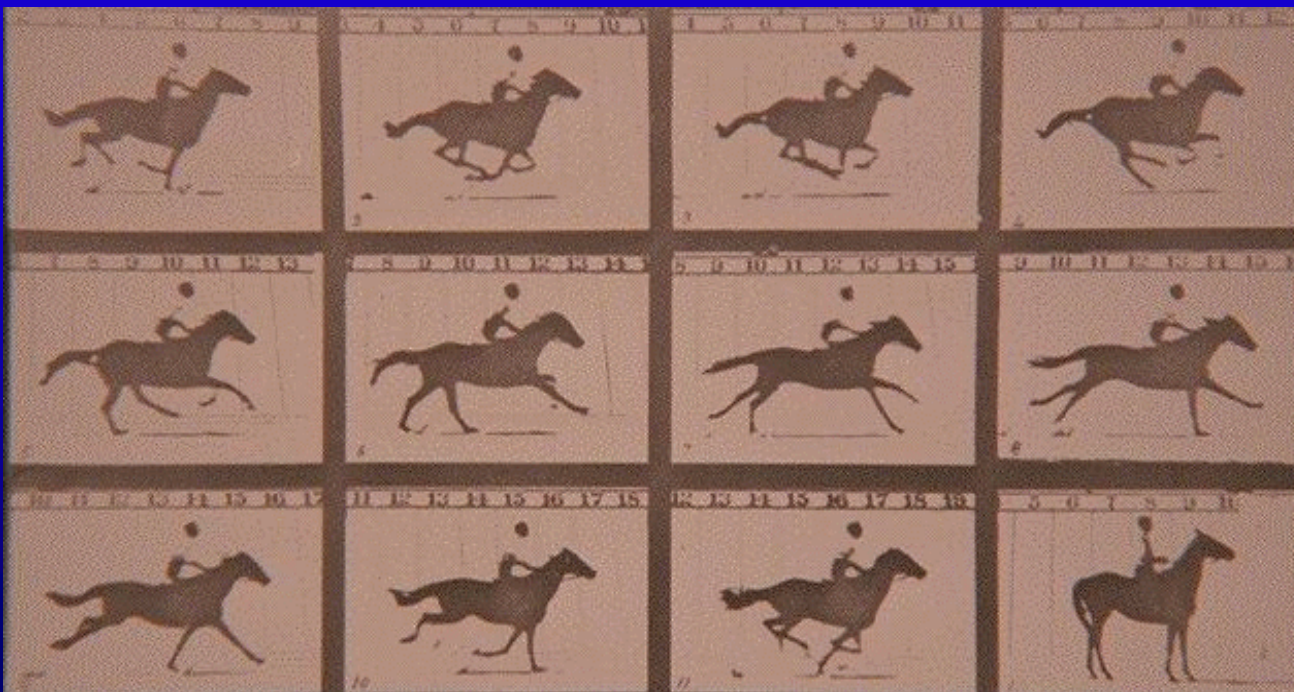
Bet: Do all four hooves
of a trotting horse ever
simultaneously leave
the ground?



Leland Stanford



Eadweard Muybridge



The "Trotting Horse"
Controversy
Palo Alto, CA 1872

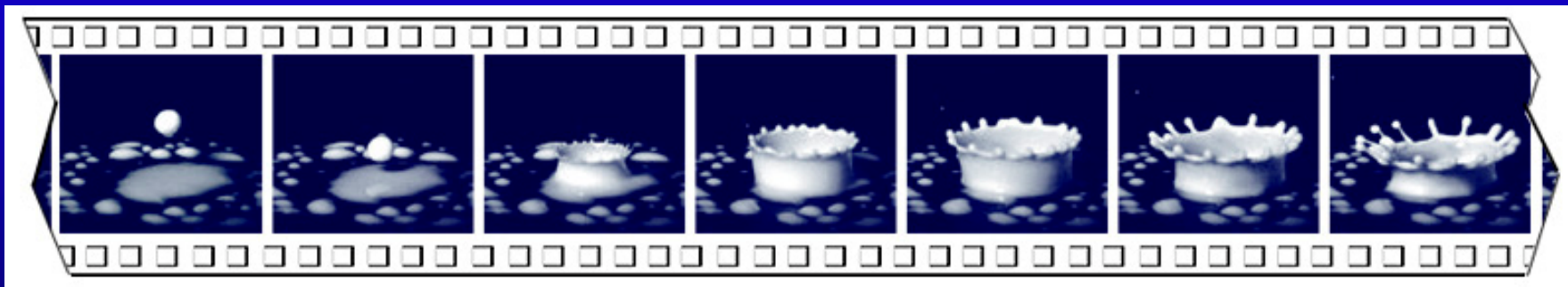
Time Resolution:
 $1/60$ th of a second





Birth of strobe photography

Harold
Edgerton
MIT, 1942



Time scale: a few microseconds

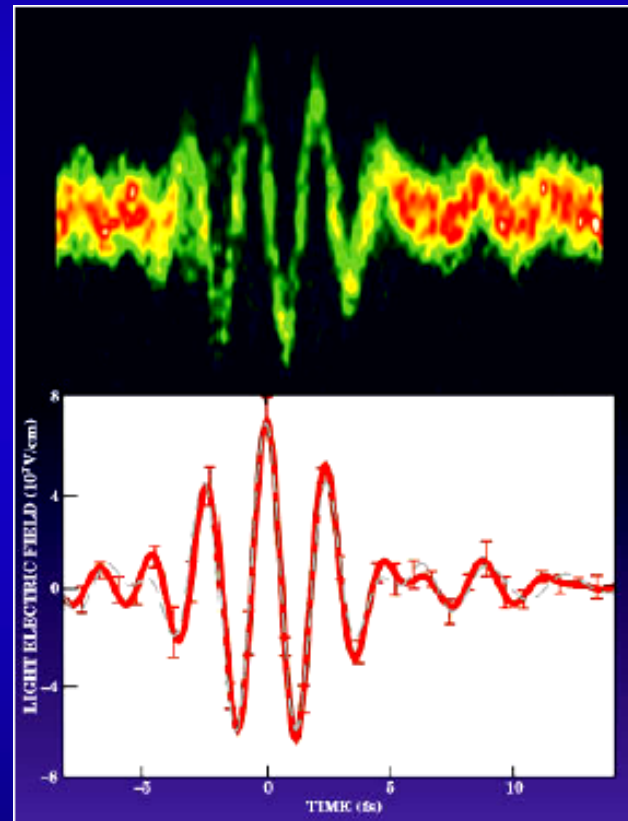
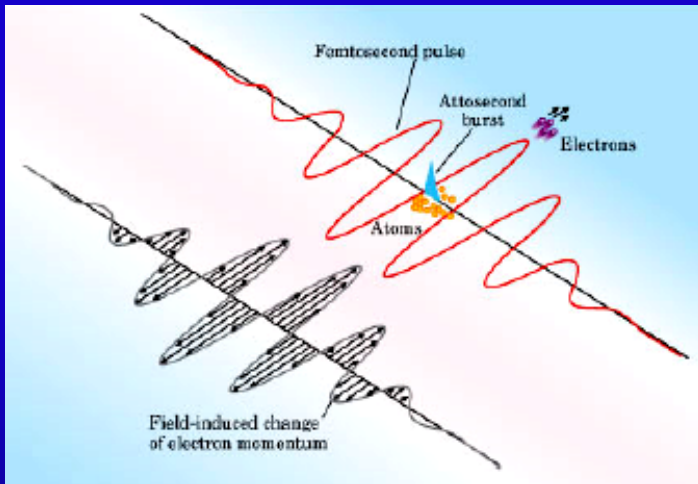




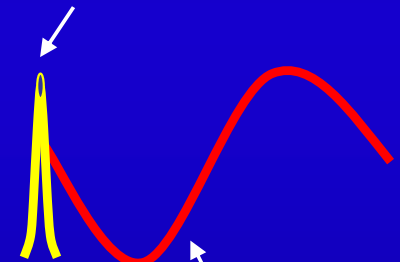
PHYSICS TODAY October 2004

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



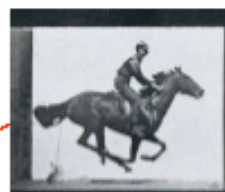
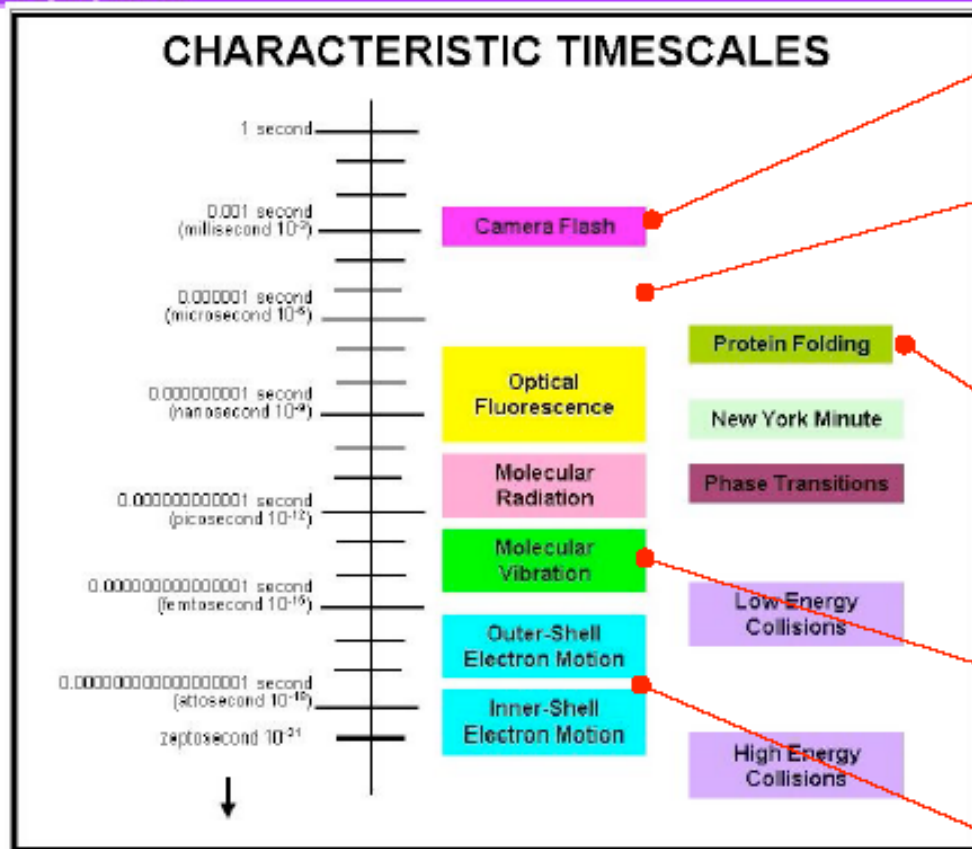
Sinusoidal pulse



EUV

An NSF Engineering Research Center

Capturing even faster events



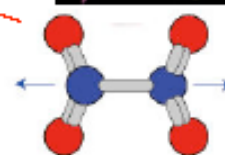
1887



1960



1980



1990



2000



Wouldn't it be nice if we can have complete control of the motion of **electrons** in space and in matter, or just about anywhere?

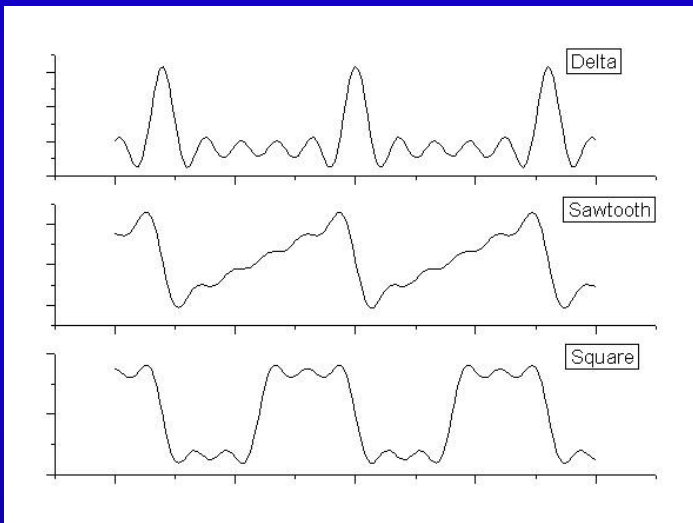
Electrons are charged particles. Their motion follows changes in the electric field. To control electron motion, we want electric fields of an arbitrary shape that we can produce at will.





GOAL

Develop an **Optical Function Generator**



With these waveforms we hope that we can do using **lightwaves** what others have done with microwaves:

Control free/bound electron motion with light

LIGHTWAVE ELECTRONICS



國立清華大學
光電工程研究所



How do we do it?

Basic Concepts



Joseph Fourier



Fourier was obsessed with the physics of heat and developed the Fourier series and transform to model heat-flow problems.

Joseph Fourier 1768 - 1830



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A revolutionary article

“
$$\varphi(y) = a \cos \frac{\pi y}{2} + a' \cos 3 \frac{\pi y}{2} + a'' \cos 5 \frac{\pi y}{2} + \dots$$

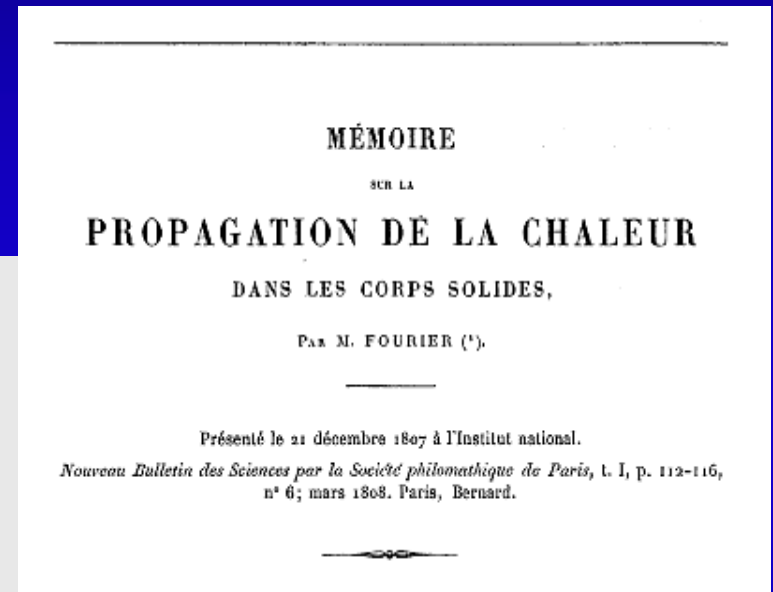
Multiplying both sides by $\cos(2i+1)\frac{\pi y}{2}$, and then integrating from $y = -1$ to $y = +1$ yields:

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

”

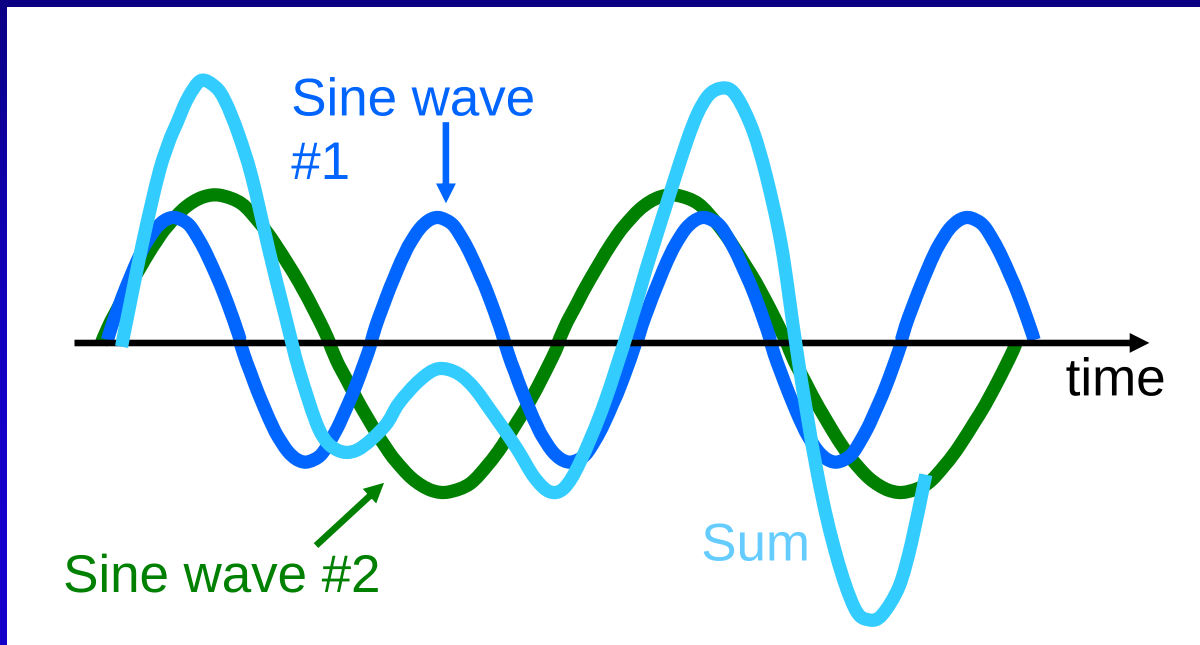
—Joseph Fourier, *Mémoire sur la propagation de la chaleur dans les corps solides*, pp. 218--219.^[1]

In these few lines, which are surprisingly close to the modern formalism used in Fourier series, Fourier unwittingly revolutionized both mathematics and physics. Although similar trigonometric series were previously used by Euler, d'Alembert, Daniel Bernoulli and Gauss,



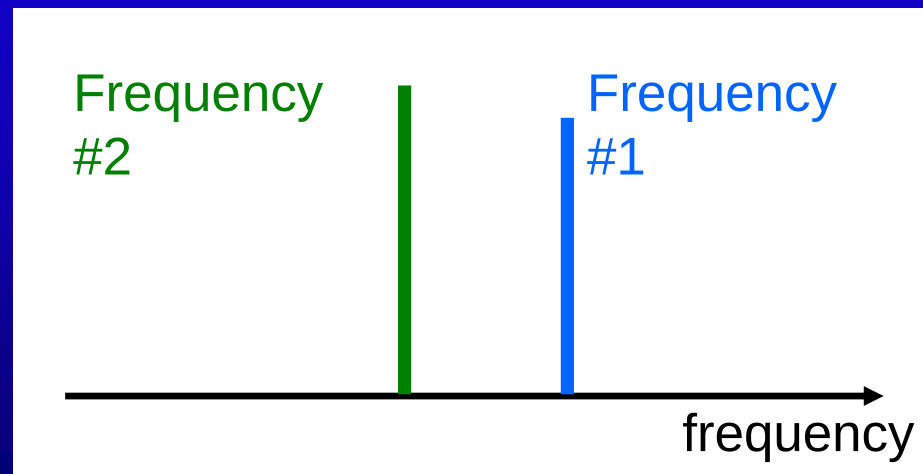


Many waves are sums of sinusoids.



- Consider the sum of two sine waves of different frequencies:

The spectrum of the sum of the two sine waves:



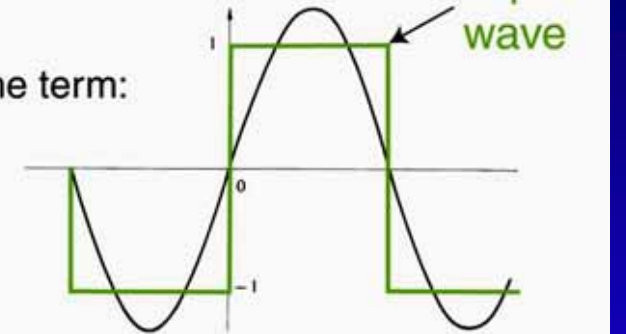


Fourier decomposing functions

- Here, we write a
- **square wave** as
- a sum of sine waves.

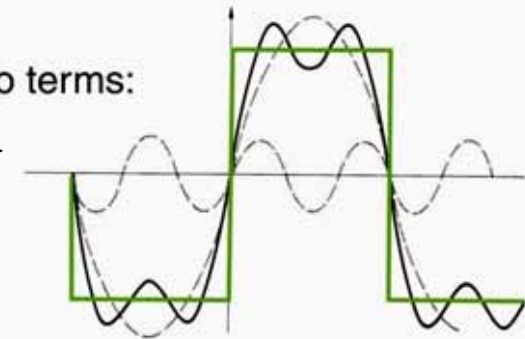
$$\sin(\omega t)$$

One term:



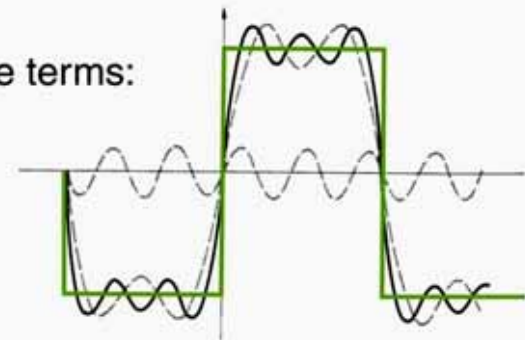
$$\sin(3\omega t)$$

Two terms:



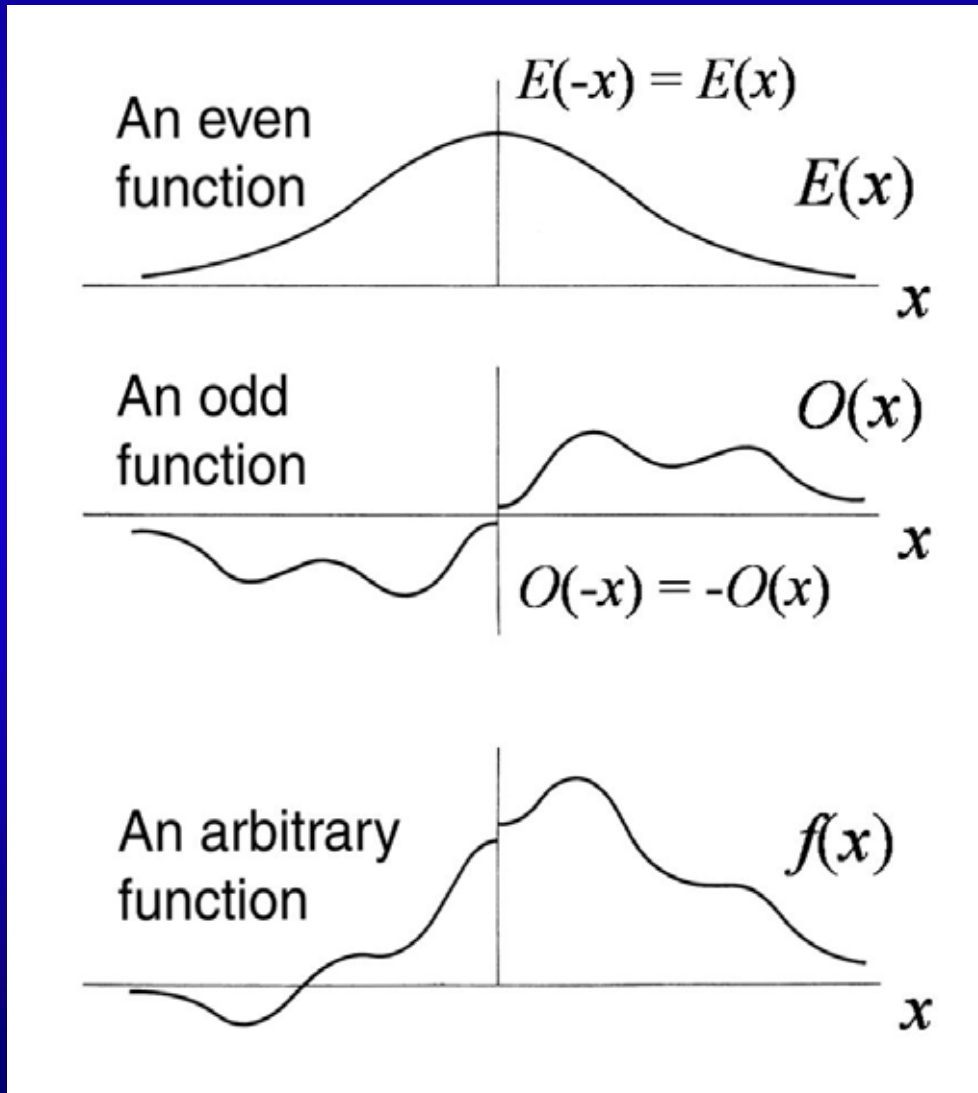
$$\sin(5\omega t)$$

Three terms:





Any function can be written as the sum of an even and an odd function.



$$F(x) = E(x) + O(x)$$



Fourier series

So if $f(t)$ is a general function, neither even nor odd, it can be written:

even component

odd component

$$f(t) = \frac{1}{\pi} \sum_{m=0}^{\infty} F_m \cos(mt) + \frac{1}{\pi} \sum_{m=0}^{\infty} F'_m \sin(mt)$$

where

$$F_m = \int f(t) \cos(mt) dt$$

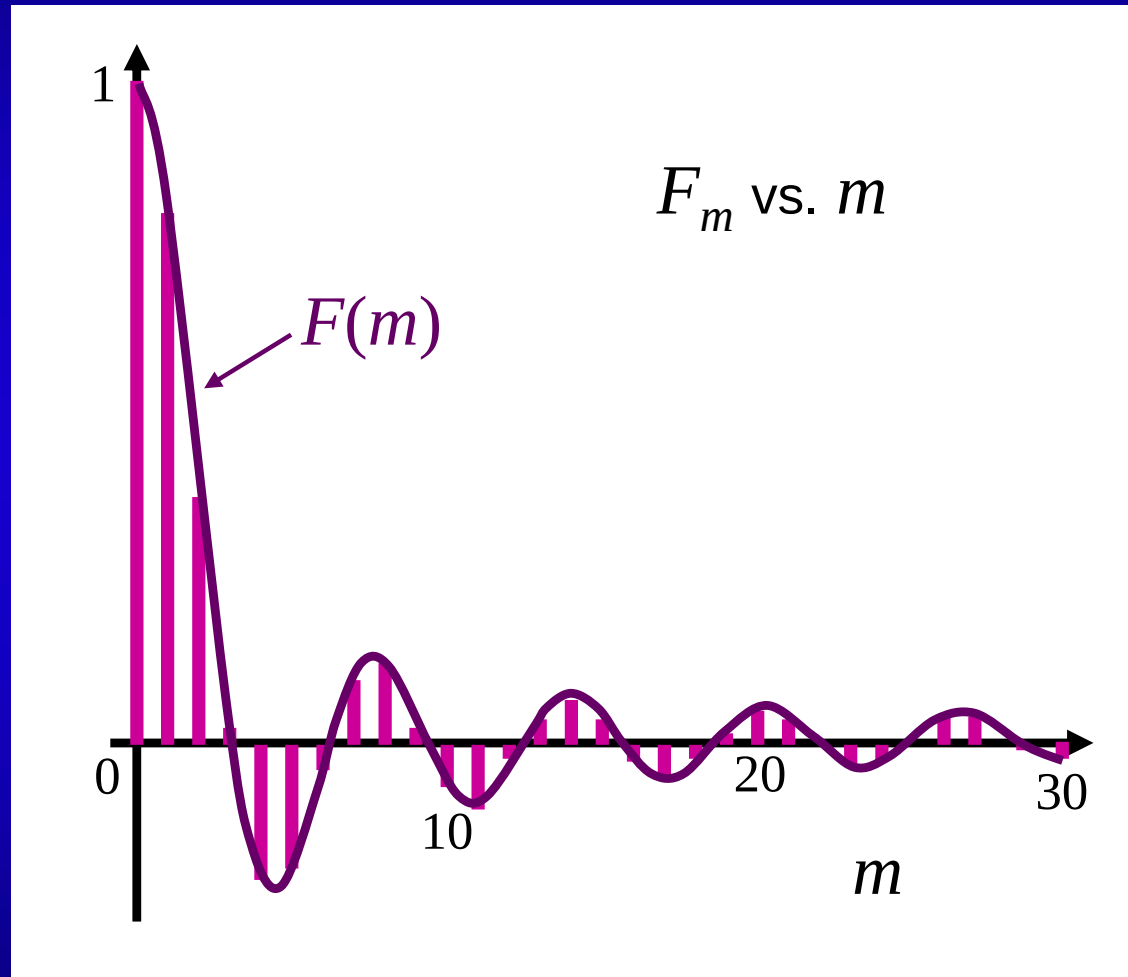
and

$$F'_m = \int f(t) \sin(mt) dt$$



We can plot the coefficients of a Fourier series.

Let the integer m become a real number and let the coefficients F_m become a function $F(m)$.



We really need two such plots, one for the cosine series and another for the sine series.





The Fourier transform

$$F(\omega) = \int_{-\infty}^{\infty} f(t) \exp(-i\omega t) dt$$

$F(\omega)$ is called the Fourier Transform of $f(t)$. **It contains equivalent information to that in $f(t)$.** We say that $f(t)$ lives in the **time domain**, and $F(\omega)$ lives in the **frequency domain**.

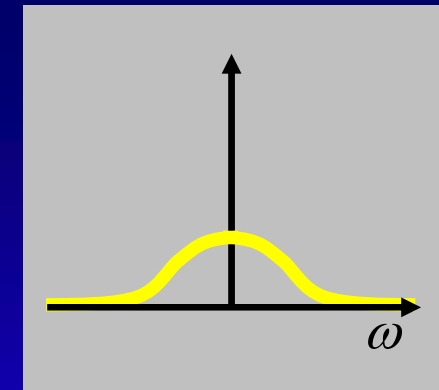
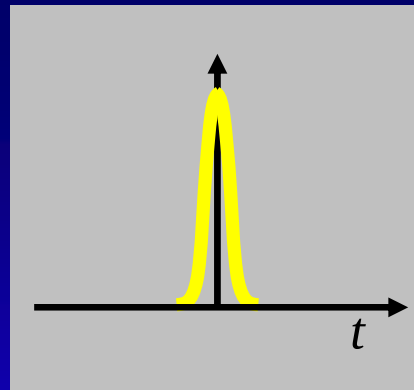
$F(\omega)$ is just another way of looking at a function or wave.



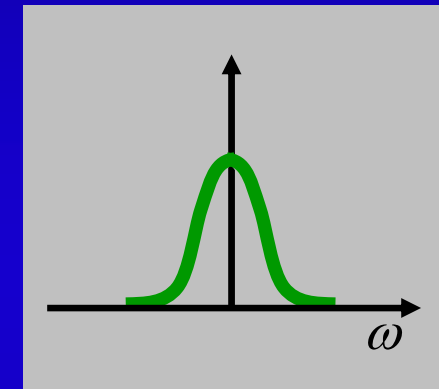
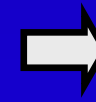
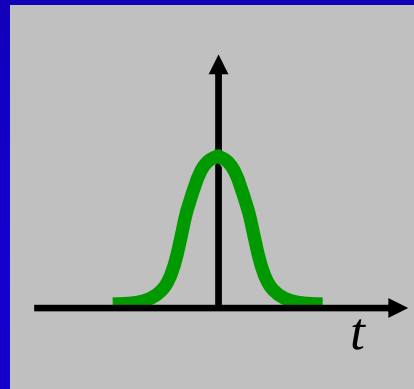
**The shorter
the pulse,
the broader
the spectrum!**

This is the essence
of the Uncertainty
Principle!

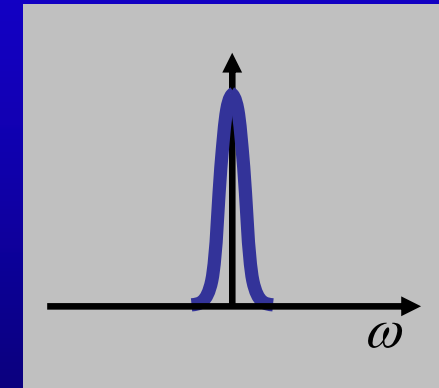
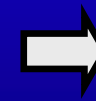
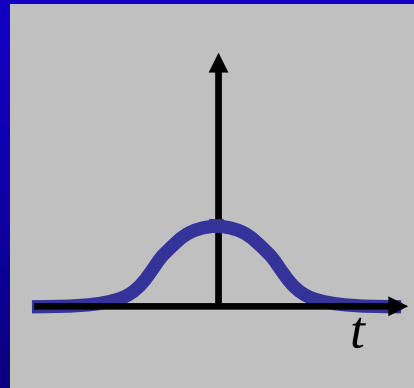
Short
pulse



Medium-
length
pulse



Long
pulse

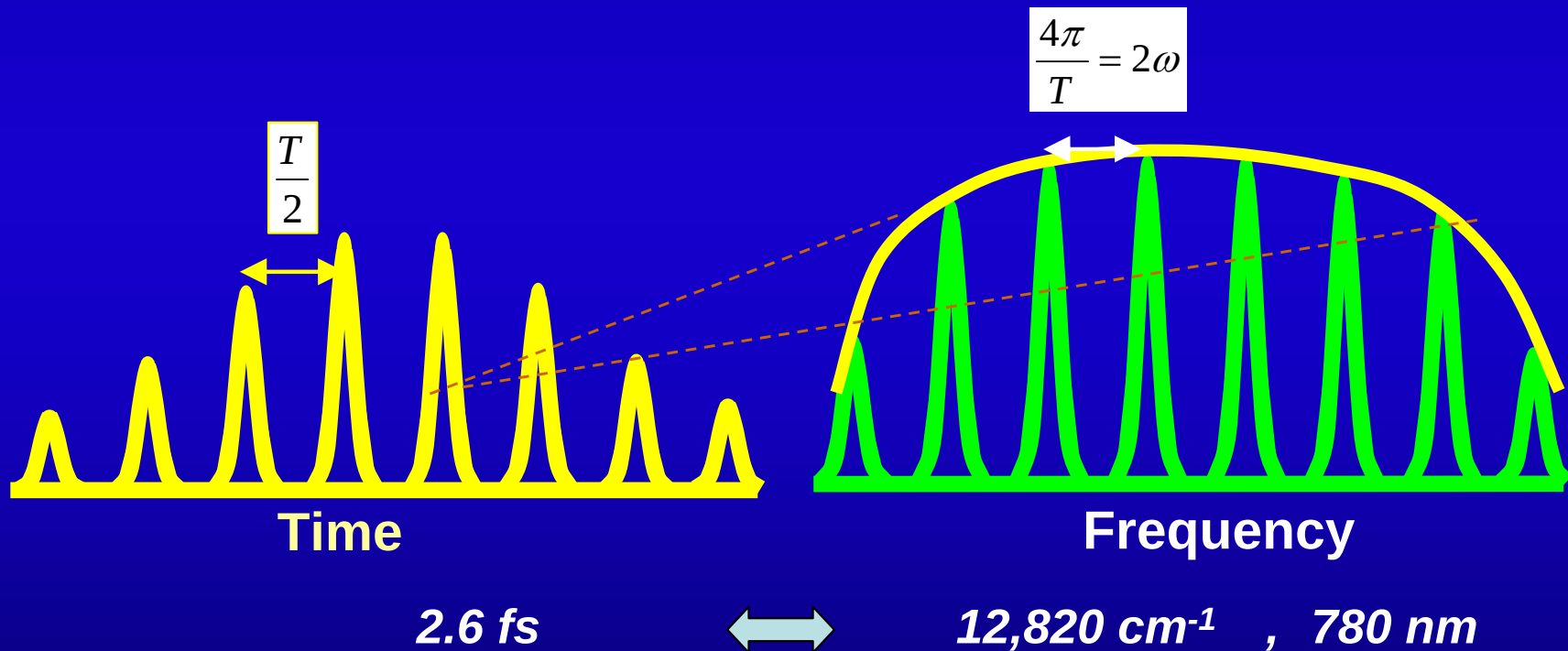




Correlation between time and frequency

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

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





Periodic waveforms are sums of sinusoidal waves

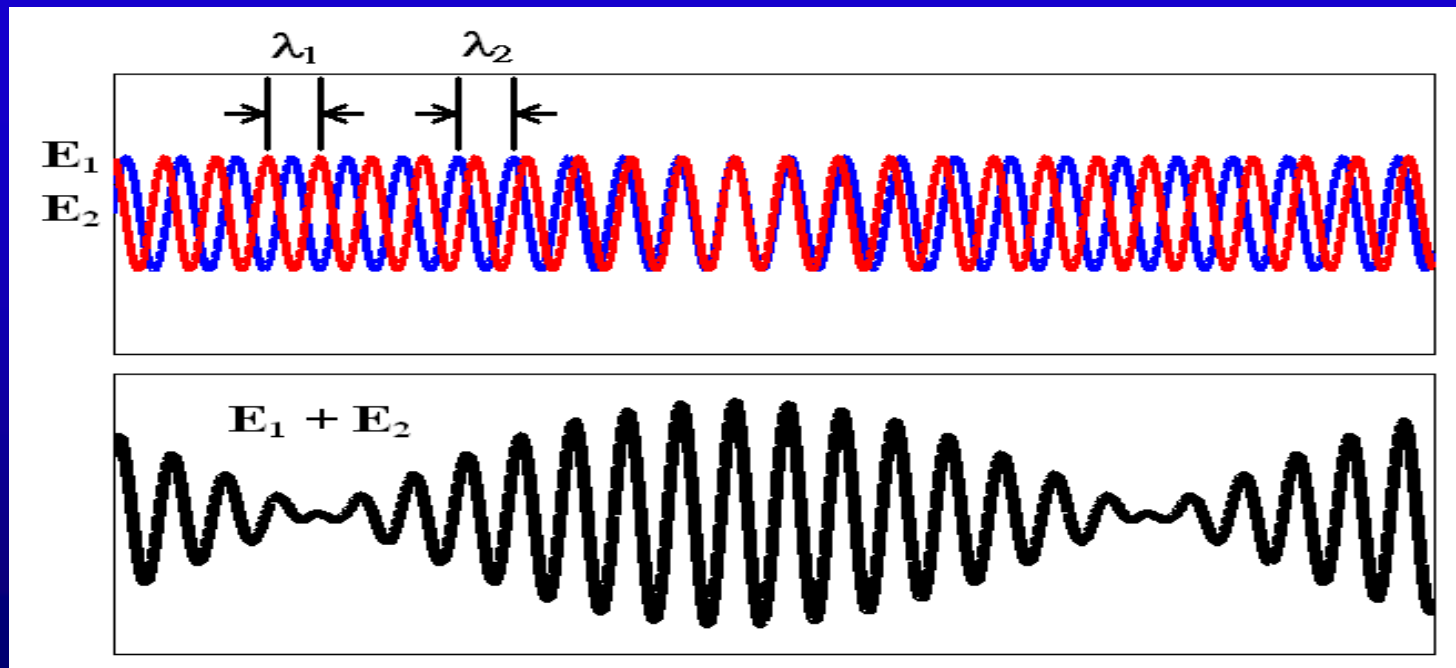
(a) Monochromatic light: sinusoidal wave

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

(b) Beating of two waves, ω_1 and ω_2

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

(b)



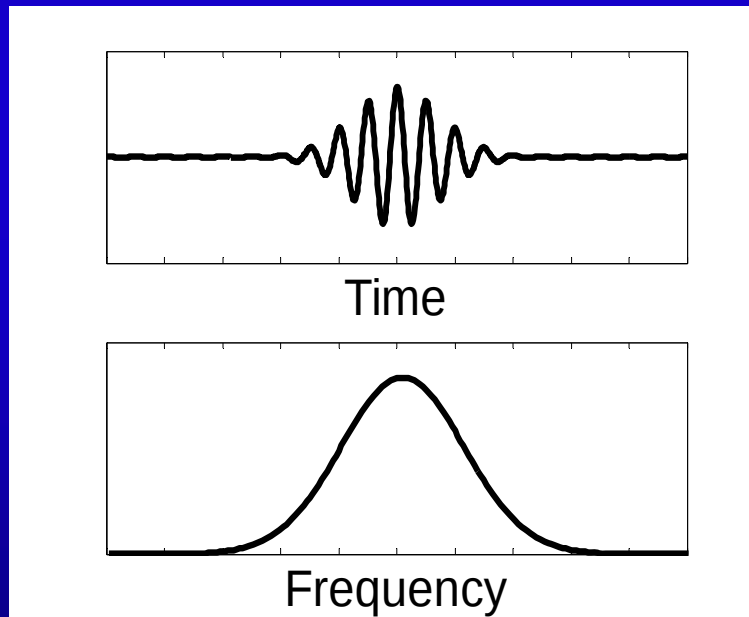


What is a single cycle optical pulse

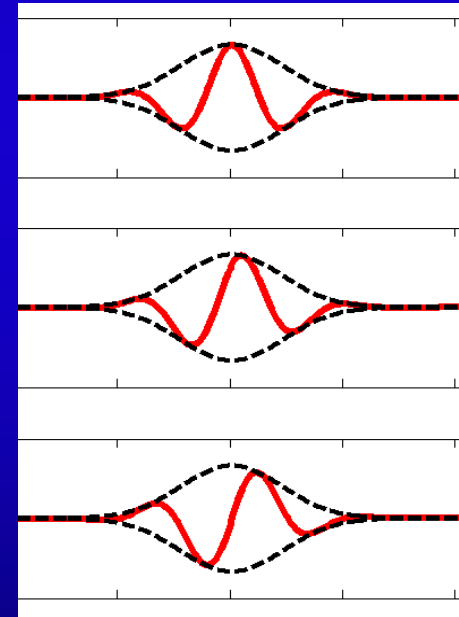
(c) Many waves propagating to form a wave packet (left)

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

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

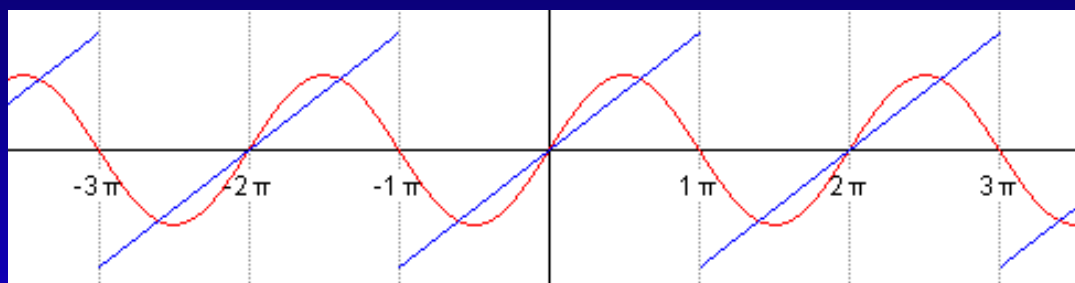


(c)

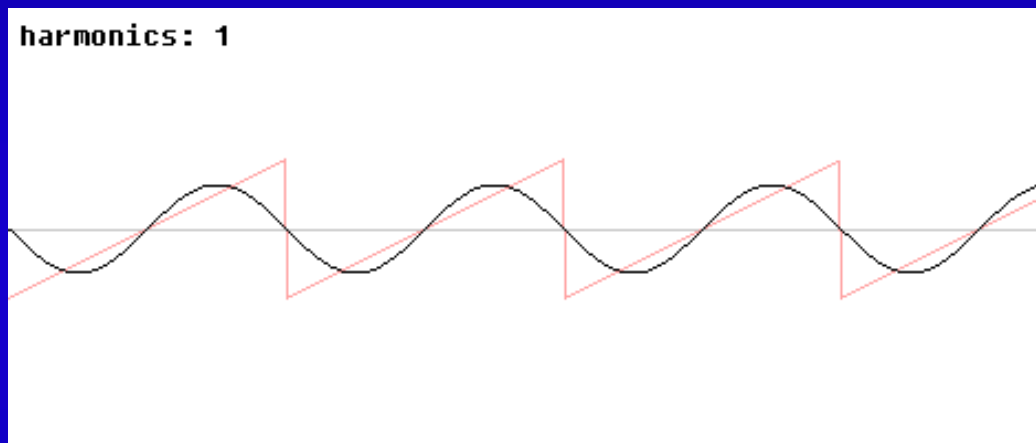


(d)

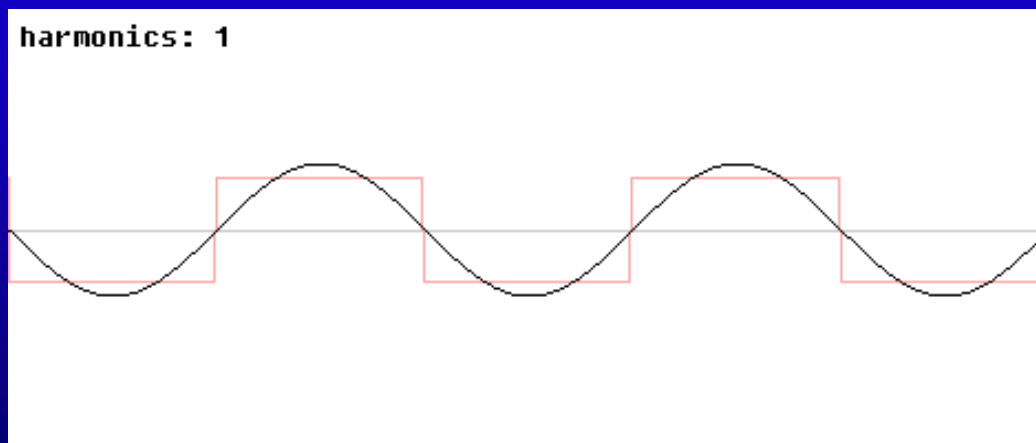




harmonics: 1

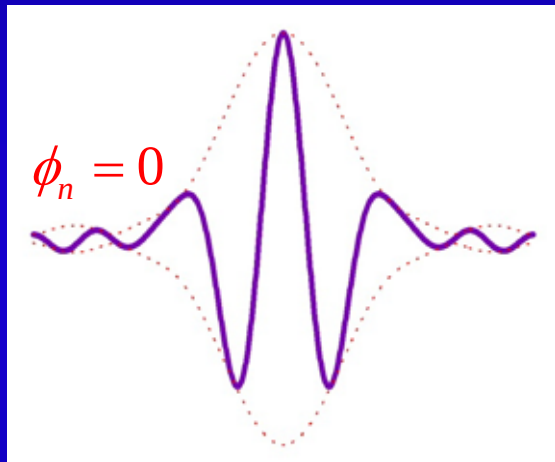


harmonics: 1

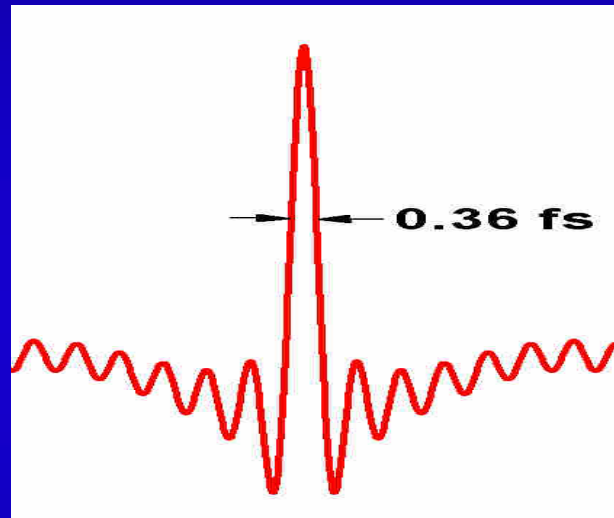




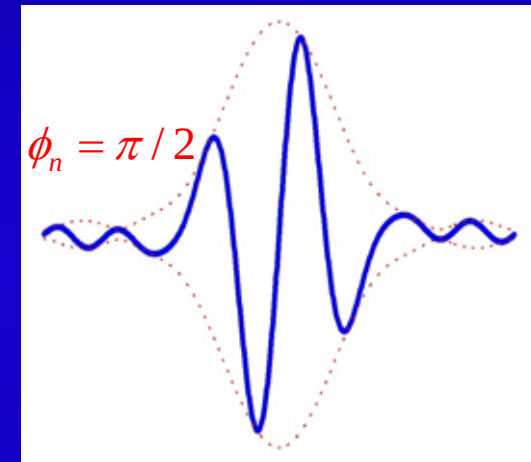
Single-cycle and sub-cycle waveforms



cosine pulse



half-cycle cosine pulse



sine pulse

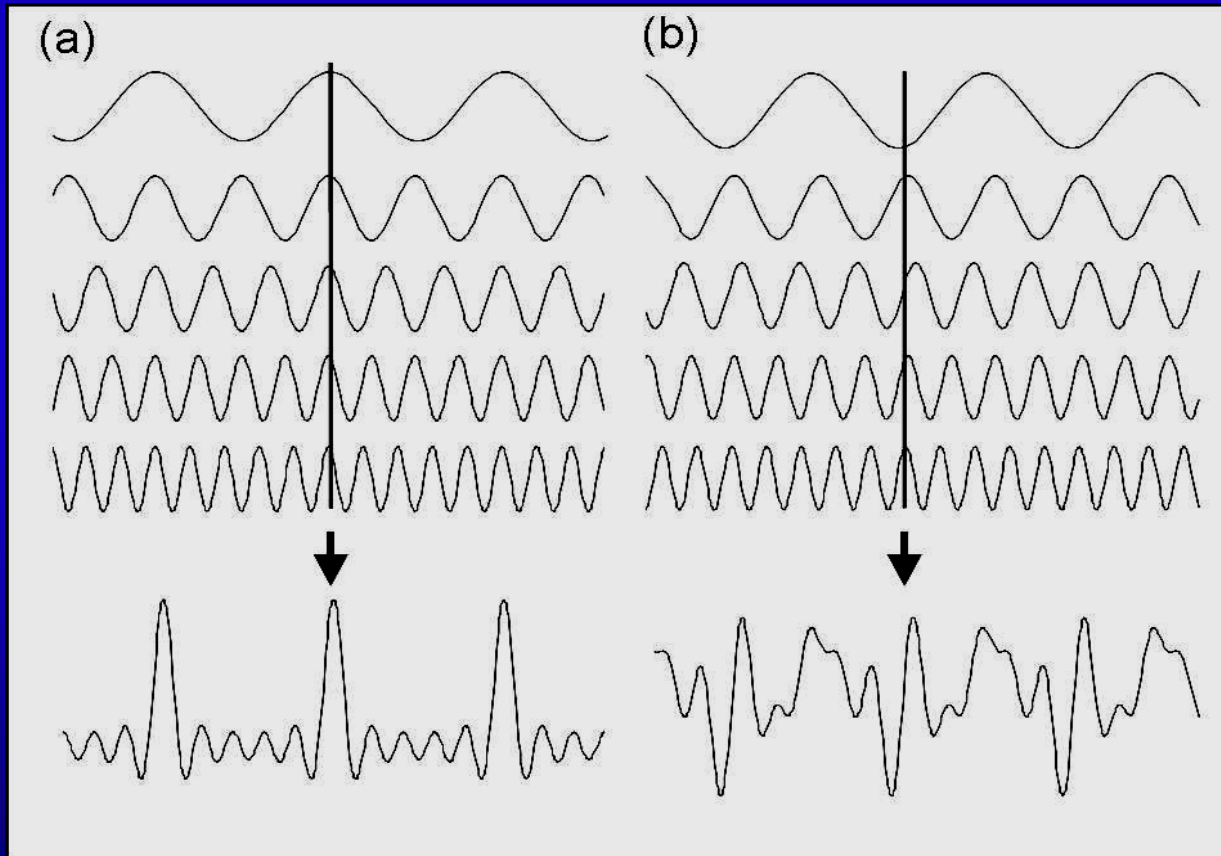


Phase coherence

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

(a) In phase $\phi_n = n\phi_0$

(b) Random phases





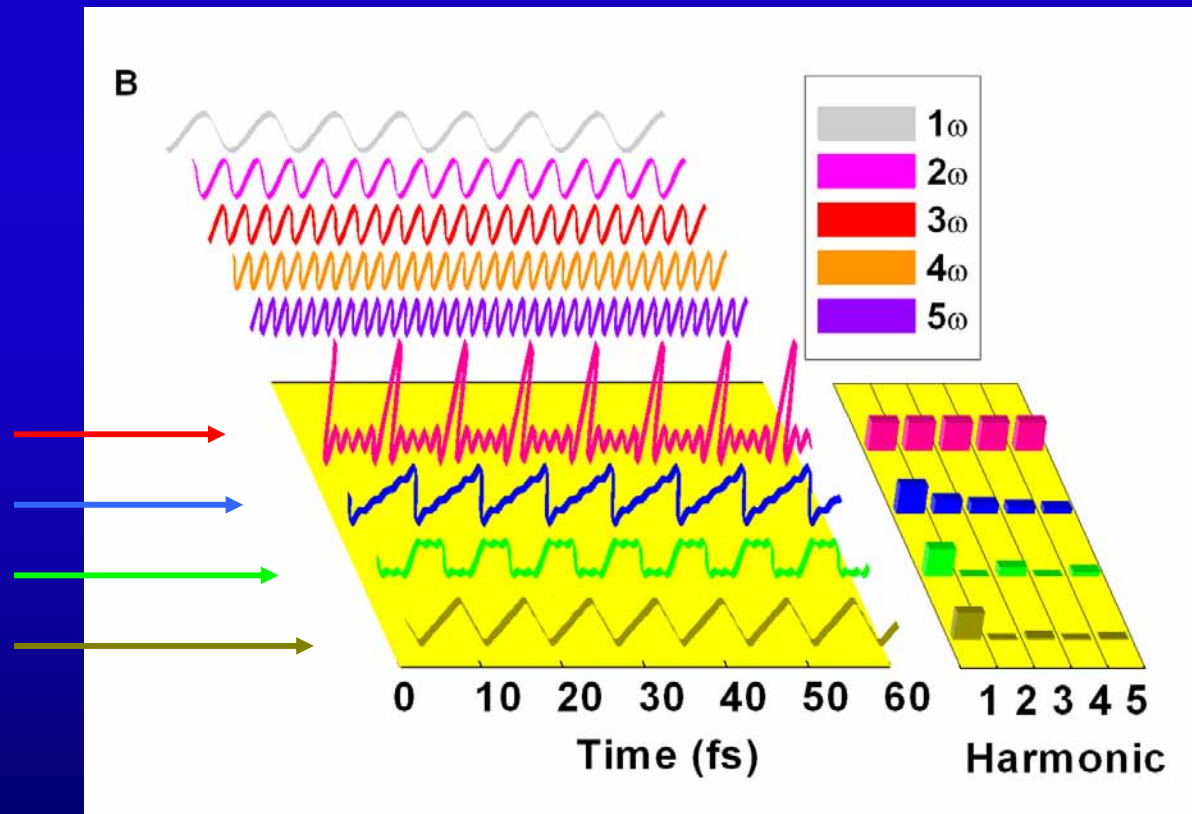
Our approach: Fourier Synthesis

Periodic waveforms are sums of sinusoidal waves

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

$$\omega_n = n \omega_m$$

single cycle cosine
saw tooth
square
triangle





Waveform	Formula	Normalized amplitude	CEP
Cosine	$\cos(n\omega t)$	1	NA
Sine	$\cos(n\omega t + \pi/2)$	1	NA
Single-cycle cosine	$\sum \cos(n\omega t), n=\text{integer}$	$\{1, 1, 1, 1, 1, \dots\}$	0
Single-cycle sine	$\sum \cos(n\omega t + \pi/2), n=\text{integer}$	$\{1, 1, 1, 1, 1, \dots\}$	$\pi/2$
Sawtooth	$(2/\pi) \sum \cos(n\omega t + \pi/2)/n, n=\text{integer}$	$\{1/1, 1/2, 1/3, 1/4, 1/5, \dots\}$	$\pi/2$
Triangular	$(8/\pi^2) \sum \cos(n\omega t)/n^2, n=\text{odd}$	$\{1/1, 0, 1/9, 0, 1/25, \dots\}$	0
Square	$(4/\pi) \sum (-1)^{(n-1)/2} \cos(n\omega t)/n, n=\text{odd}$	$\{1/1, 0, -1/3, 0, 1/5, \dots\}$	0





Tasks

1. Generate a broad (prefer octave spanning) spectrum or comb of frequencies (with equal spacing)
2. Align the phases of the comb frequencies
3. Determine that the synthesized field is correct

Challenge: these must be done in the optical domain

that is ~ 100 THz, ~ 1 femtosecond

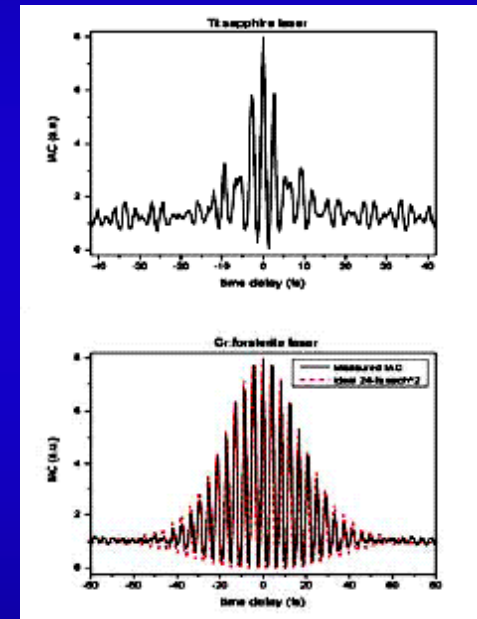
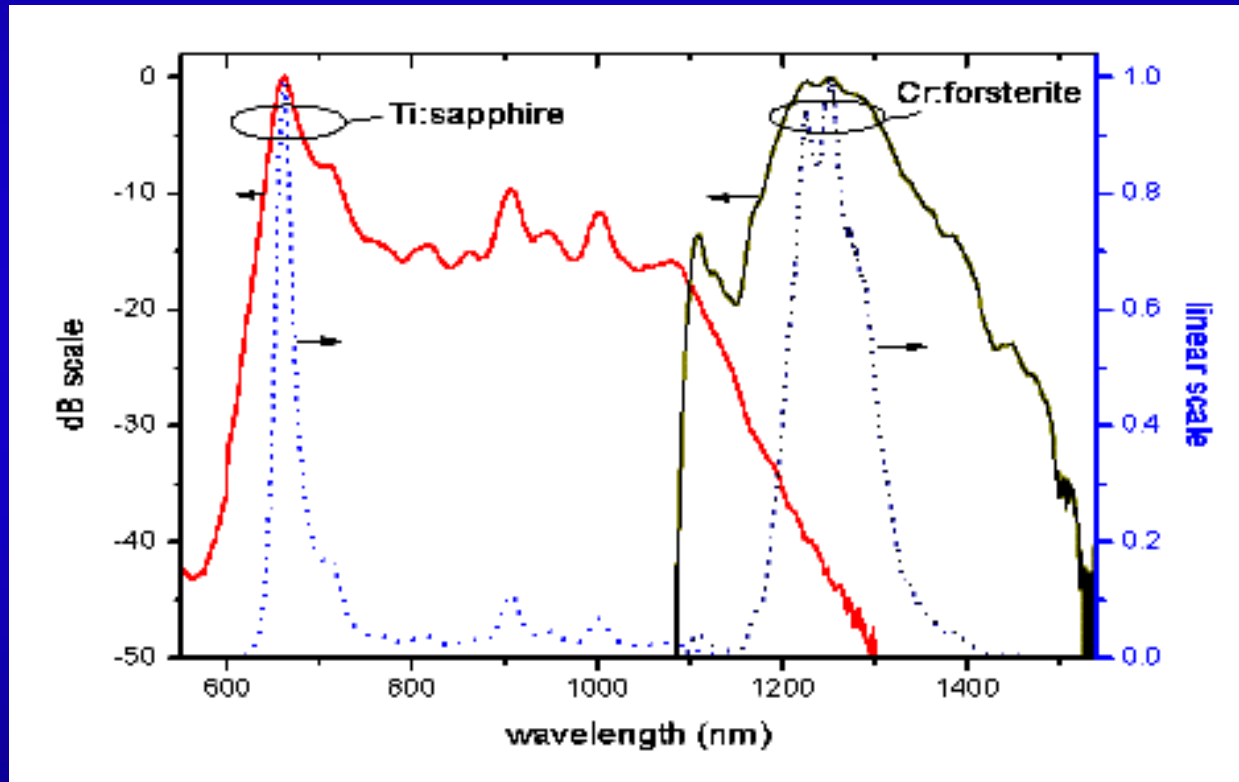


Methods to produce optical waveforms



Synthesize optical pulses

Isolated single cycle pulses -- extremely broadband lasers

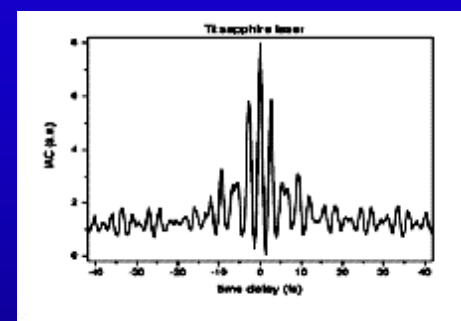
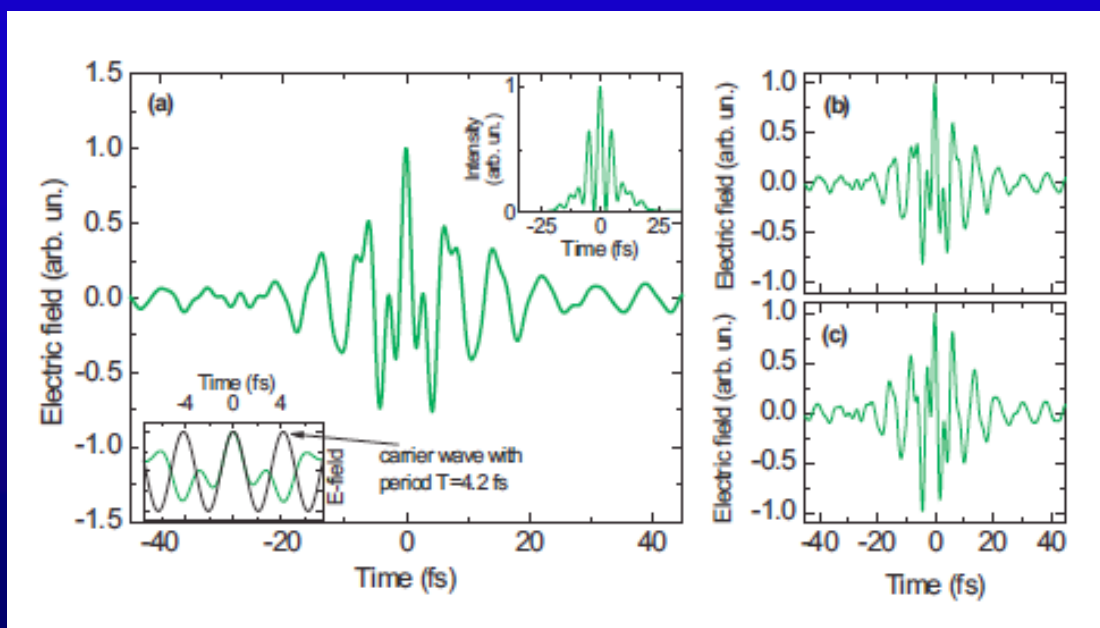
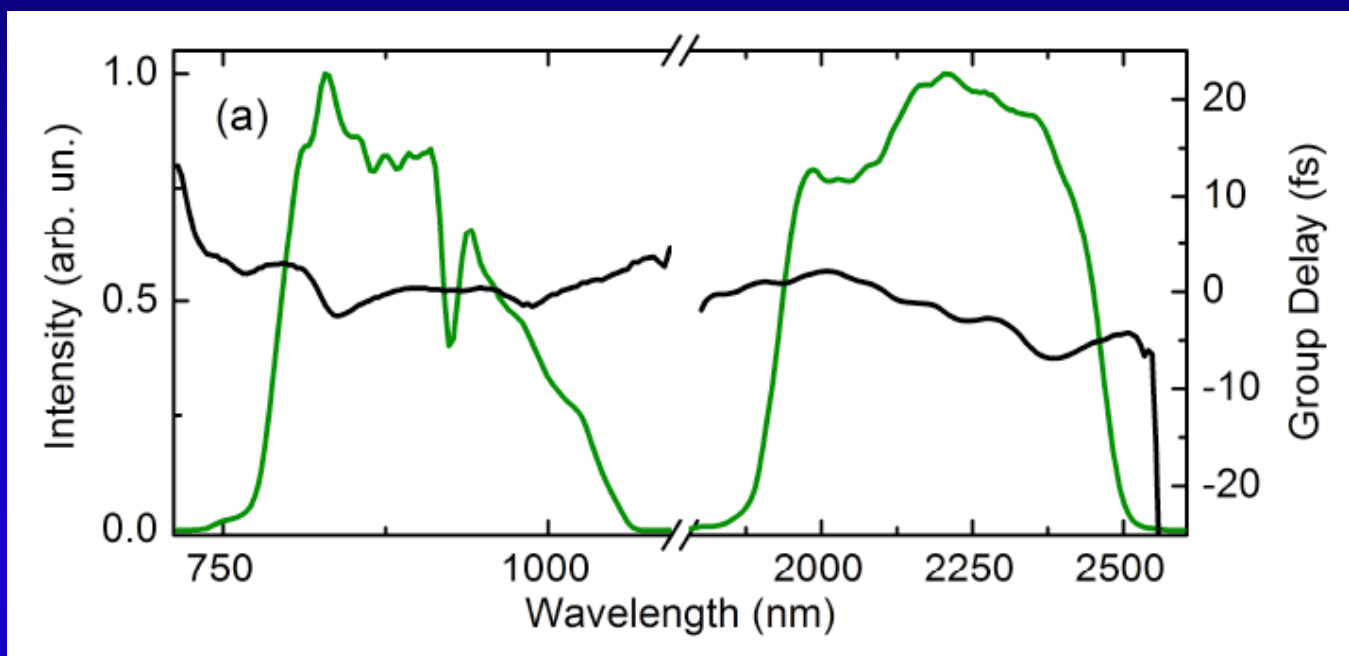


Franz Kartner, MIT



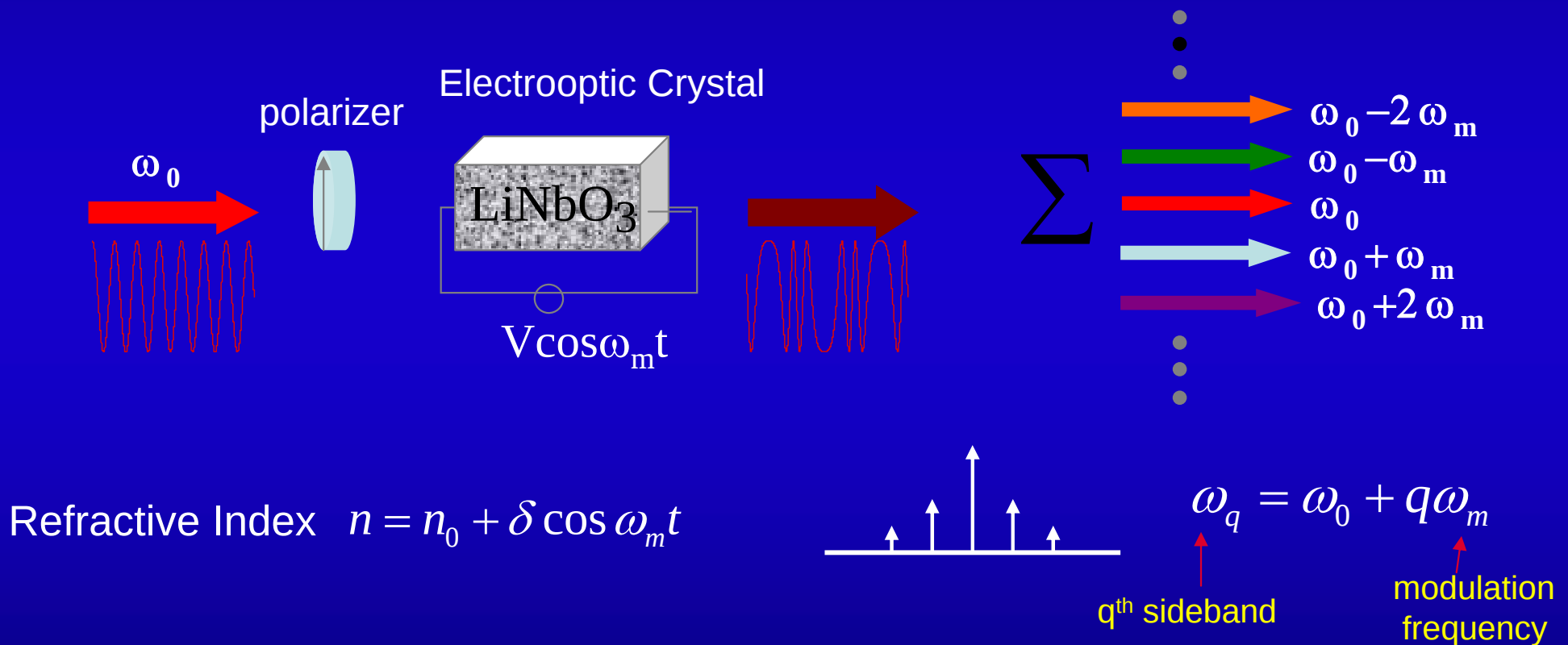


Huang et al., Nature Photonics, August, 2011



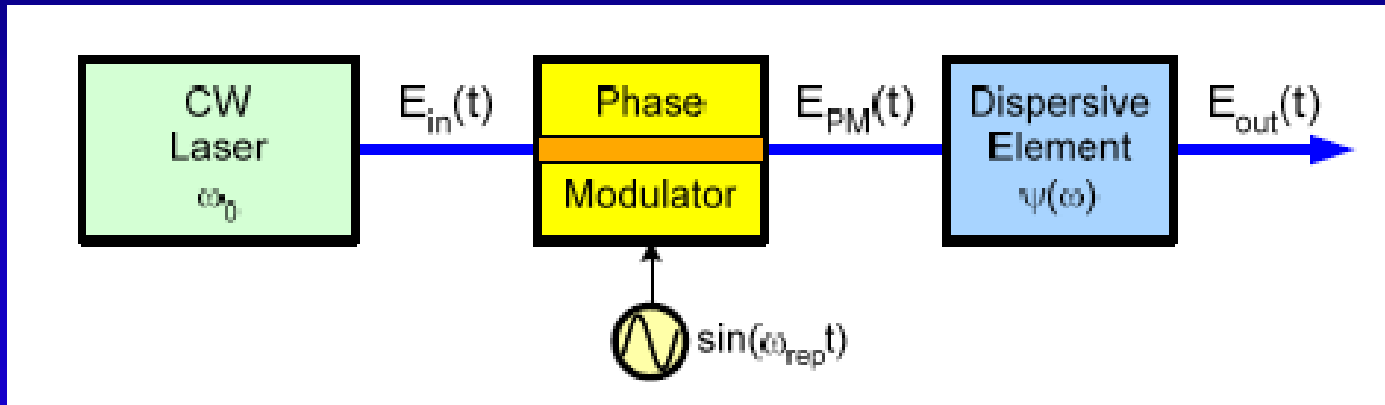


Frequency comb expansion by modulation

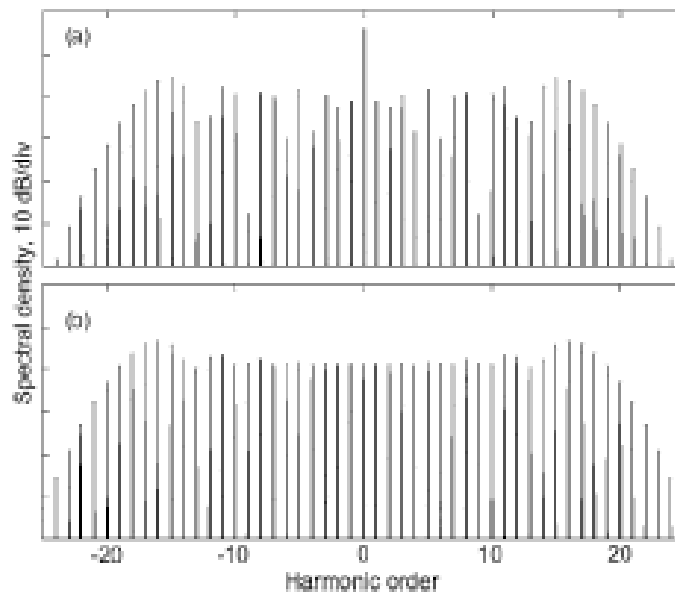




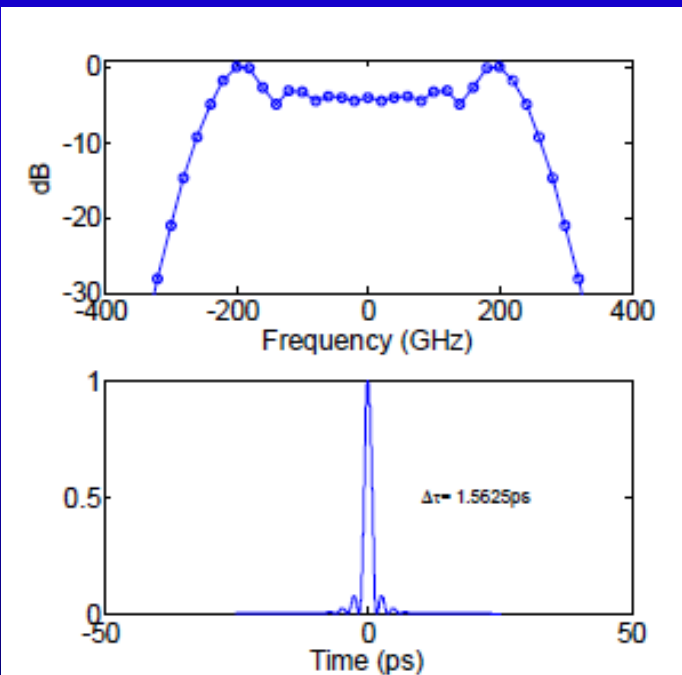
CW frequency comb produced by electro-optic modulation



Mode spacing
Tens of GHz

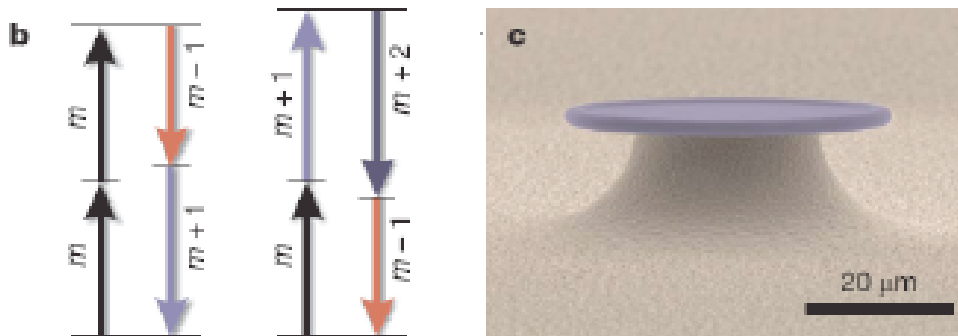
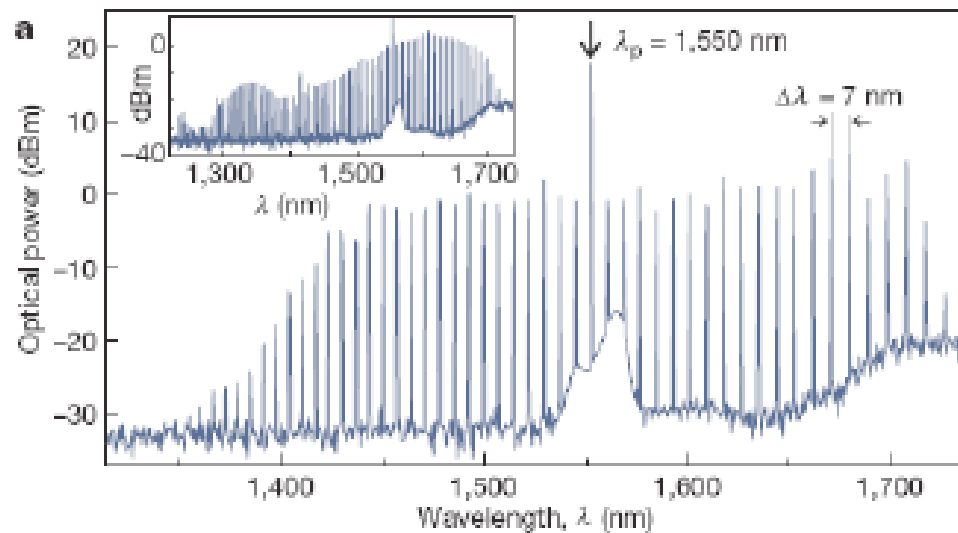


T. Sakamoto et.al, Opt. Lett. **32**, 1515 (2007).





CW comb produced by four wave Kerr mixing in a micro-toroidal ring

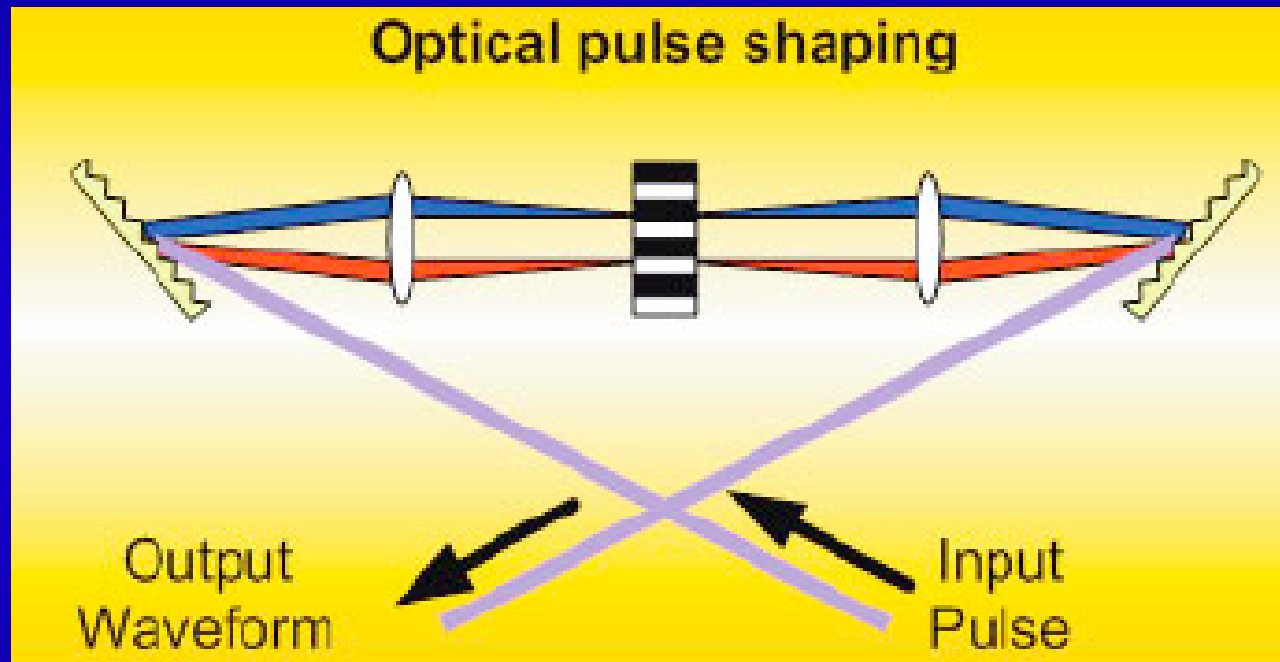


Mode spacing
 ~ 1 THz

P. Del'Haye et al., Nature, 2007



Waveform synthesis by line-by-line pulse shaping



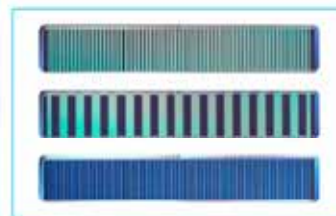
Andrew Weiner



Excellence through light.



Spatial Light Modulator SLM-S640d.



Examples for patterns.



SLM-S 640d with Reflexive Mirror.

The Liquid Crystal Modulator SLM-S640d is an excellent tool for modulation of femto second laser pulses in wave length ranges from 430 to 1600 nm.

The simultaneous modulation of phase and amplitude independent from each other is achieved by the alignment of two connected liquid crystal modulators in a 4f-arrangement or in a cpa-system (chirped pulse amplification). Any of the 1280 strips of the single masks can actively be controlled with 12 bit resolution.

The large active area of 64 x 10 mm offers the use of an increased laser pulse output power. An extensive LabView™ instruction set as well as several demo-programmes for calibration and control of the modulators facilitate the effective work.

Using the SLM-S640d the user is offered the possibility to optionally mount a mirror. Thus the same modulator can be operated both in transmissive and in reflexive configurations.



國立清華大學
光電工程研究所

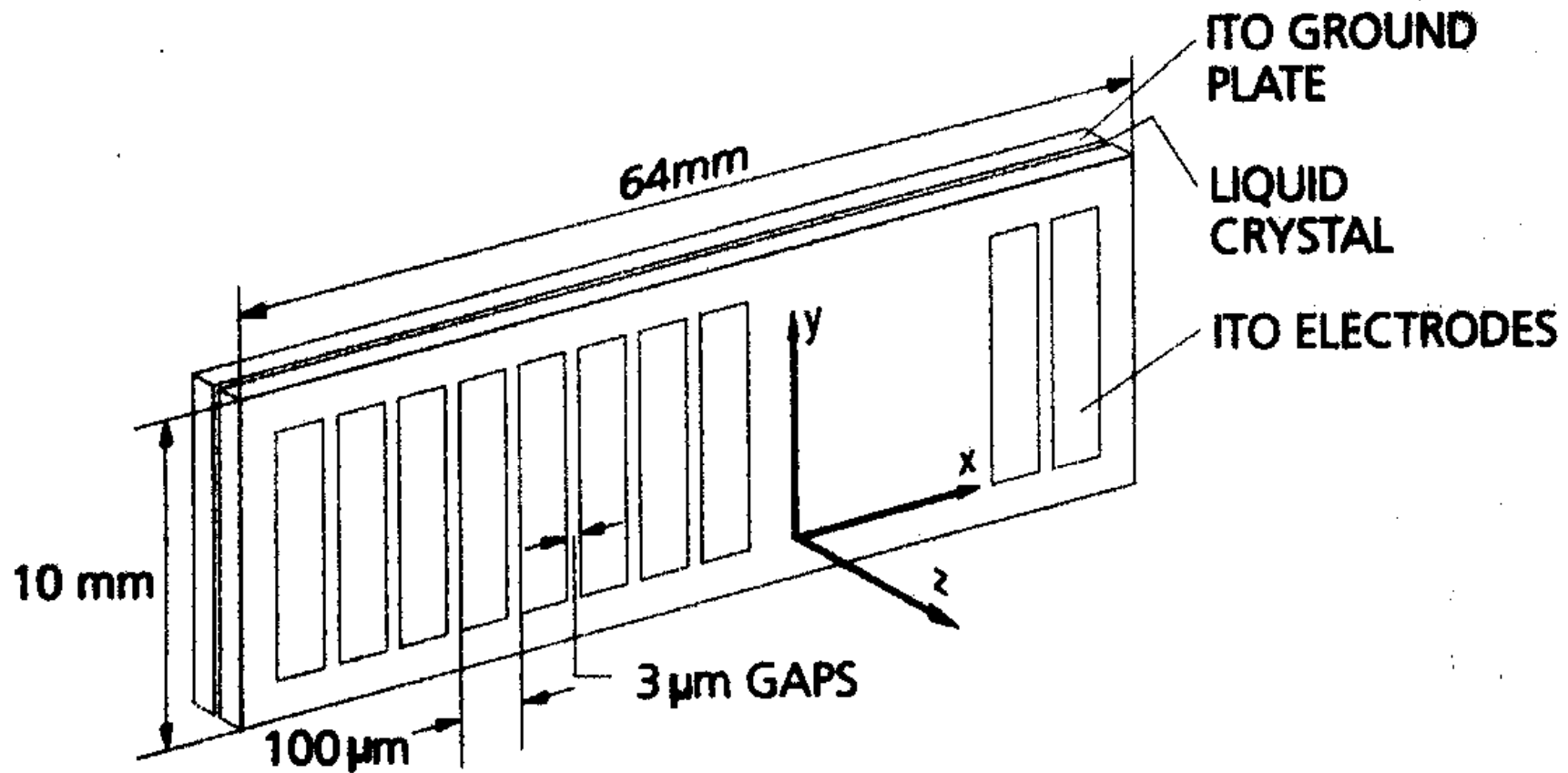


Fig.3.2 Layout of the optical active area



Shaped envelopes using CW frequency combs

Laser & Photon. Rev. 2, No. 4 (2008)

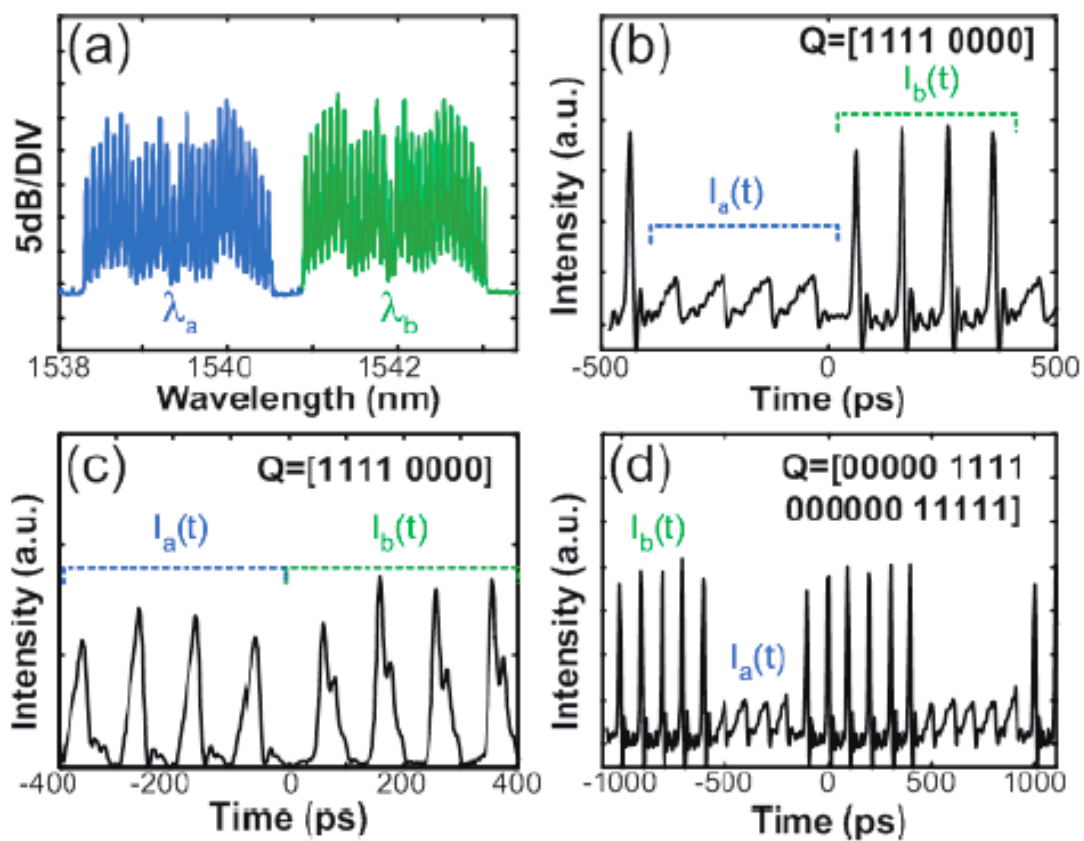
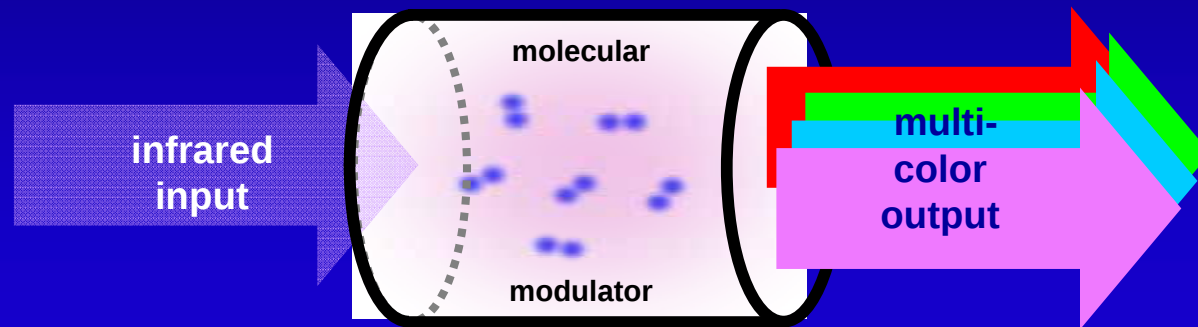


Figure 25 (online color at: www.lpr-journal.org) (a) 28-line



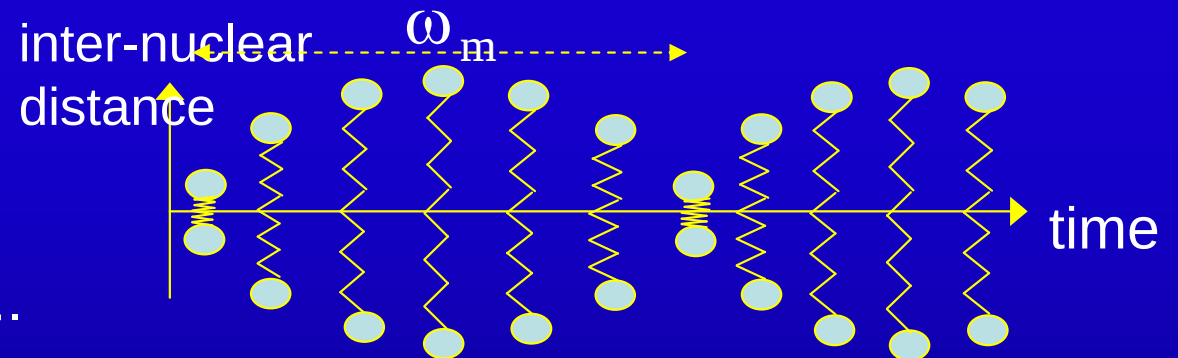
Bandwidth expansion by molecular modulation



Refractive Index

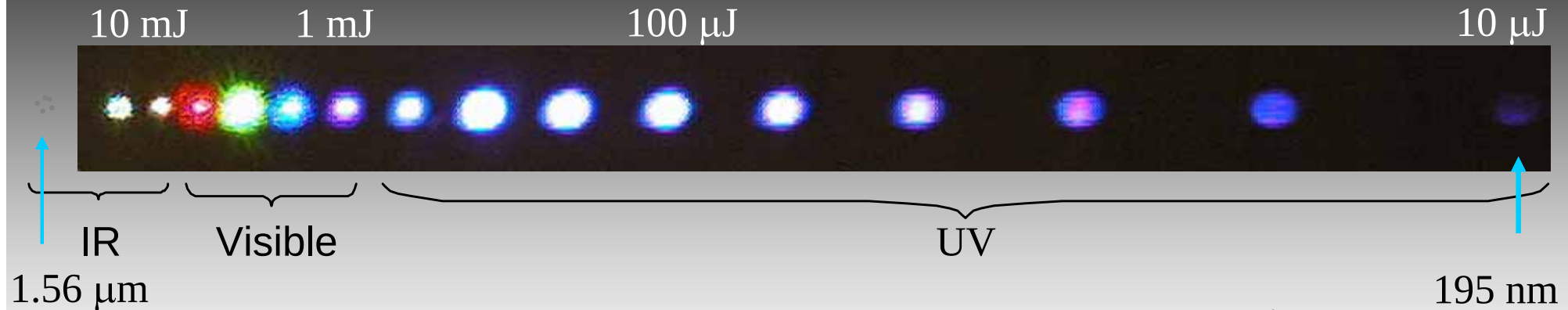
$$n = n_0 + \delta \cos \omega_m t$$

$$\omega_q = \omega_0 + q\omega_m \quad q = -2, -1, 0, 1, 2, 3, \dots$$



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).

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



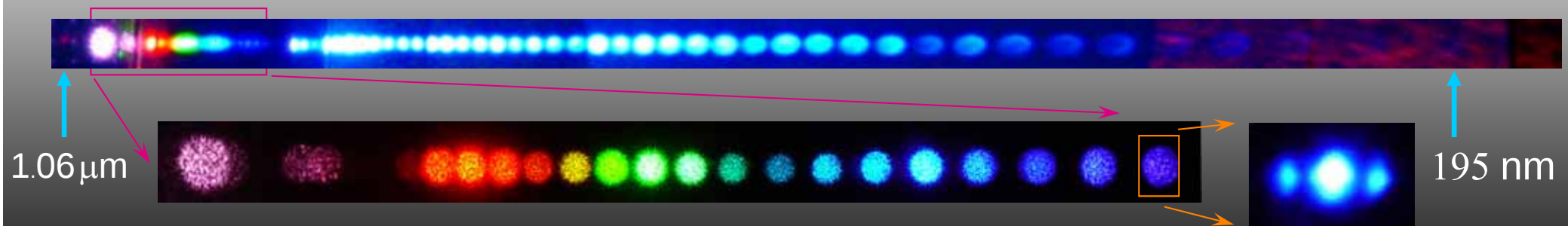
H_2 Rotation Spectra: 29 sidebands, spaced by 587 cm^{-1}



Harris group:

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 $< 587\text{ cm}^{-1}$



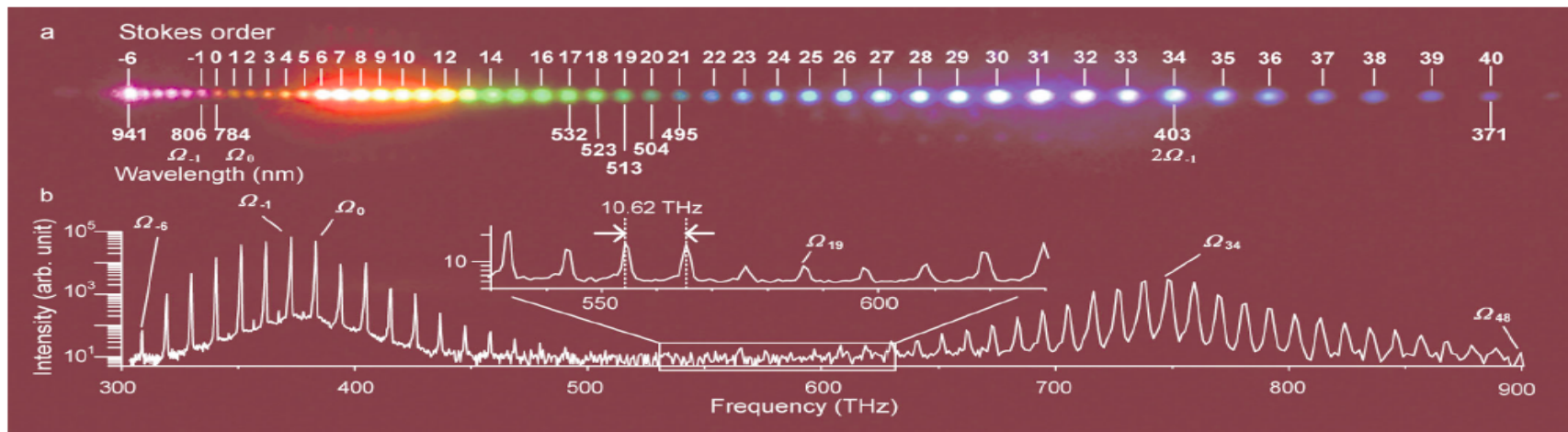
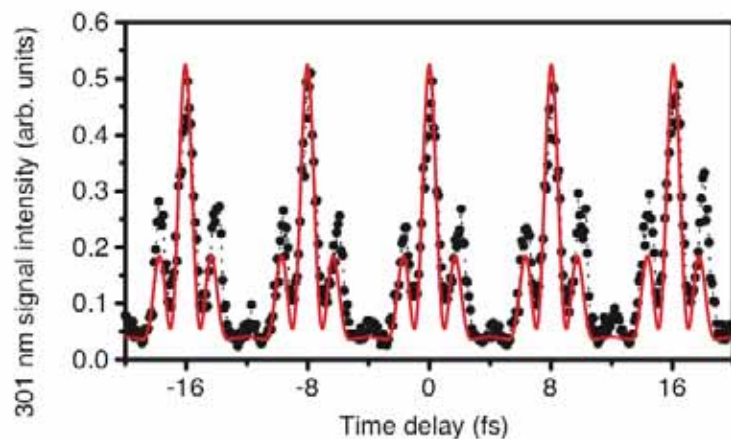


FIG. 2 (color). Spectra for octave-spanning Raman comb produced with three frequency laser radiations. (a) The photograph taken with a digital camera after dispersing the emission with a prism. (b) The spectrum taken with an OMA.

243602-3

Katsuragawa, PRL 101, 243602 (2008)



W.-J.Chen et al. PRL 100, 163906 (2008)

Commensurate comb
Constant CEP



Use the first two terms of the Fourier series

$$n = 1, 2 \quad \text{Then,} \quad \phi_m = \phi_2 - \phi_1$$
$$\phi_0 = 2\phi_1 - \phi_2$$

$$\text{since } \omega_2 = 2\omega_1 \quad \text{then, } \phi_2 = 2\phi_1 + \xi$$

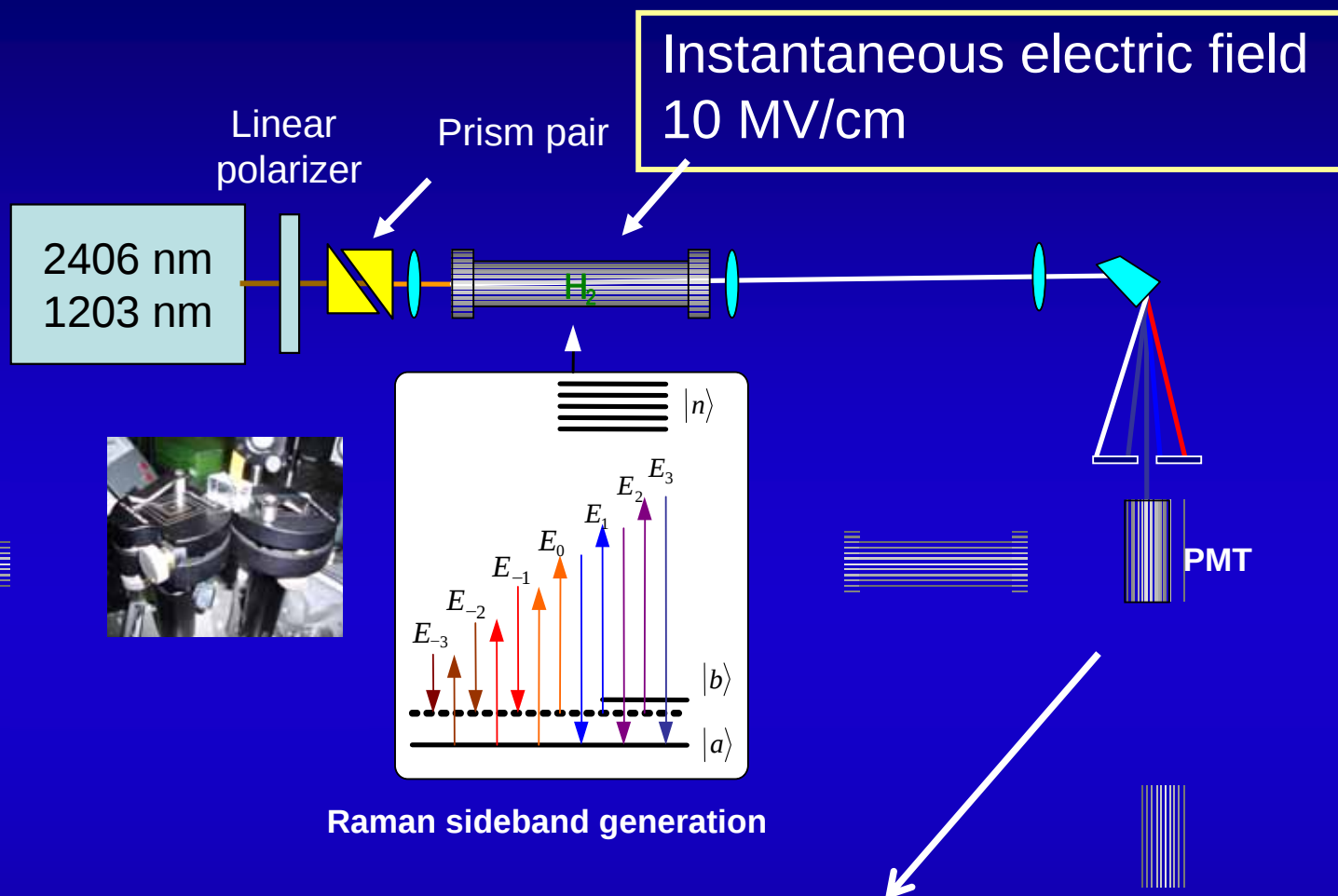
$$\begin{aligned} \phi_0 &= 2\phi_1 - \phi_2 \\ &= 2\phi_1 - (2\phi_1 + \xi) \\ &= \xi \end{aligned} \quad \text{a constant we can choose and control}$$

ω_1 is doubly significant. The period of the waveform is determined by the fundamental frequency and its shape is dominated by the first few terms of the Fourier series.





IR Laser at 2406 nm
Pulse duration $\sim 2-5$ ns
Peak power $\sim 2-5$ MW



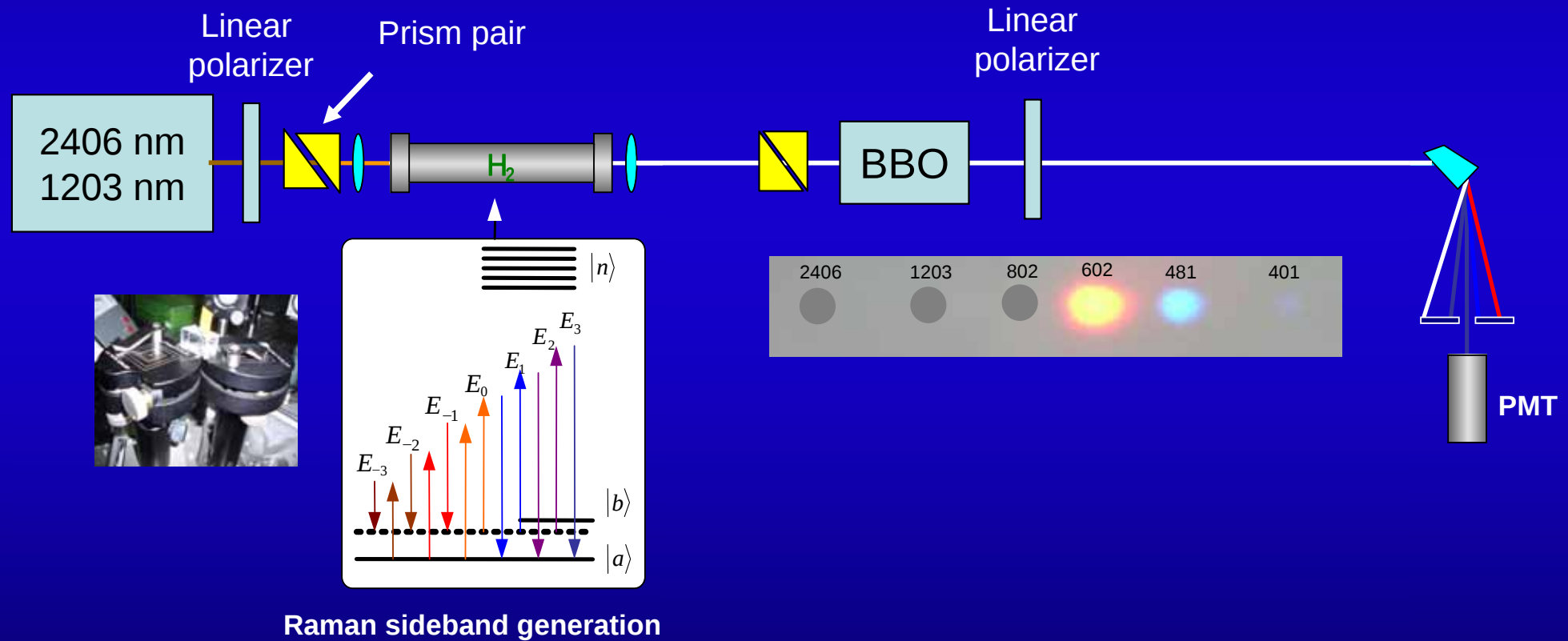
Total spectral span > 4 octaves ($70,000 \text{ cm}^{-1}$)
(~ 500 as)





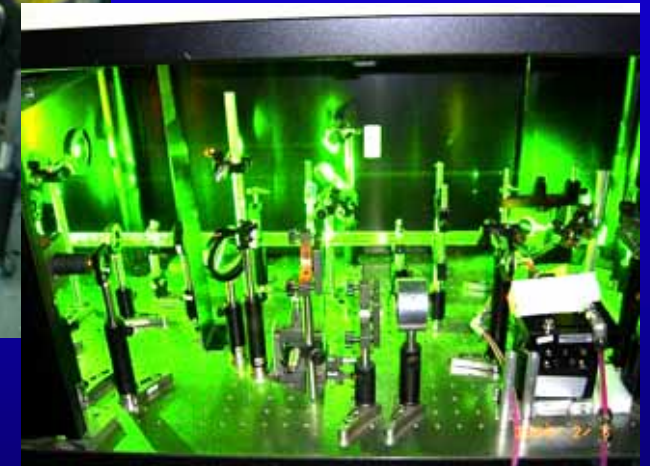
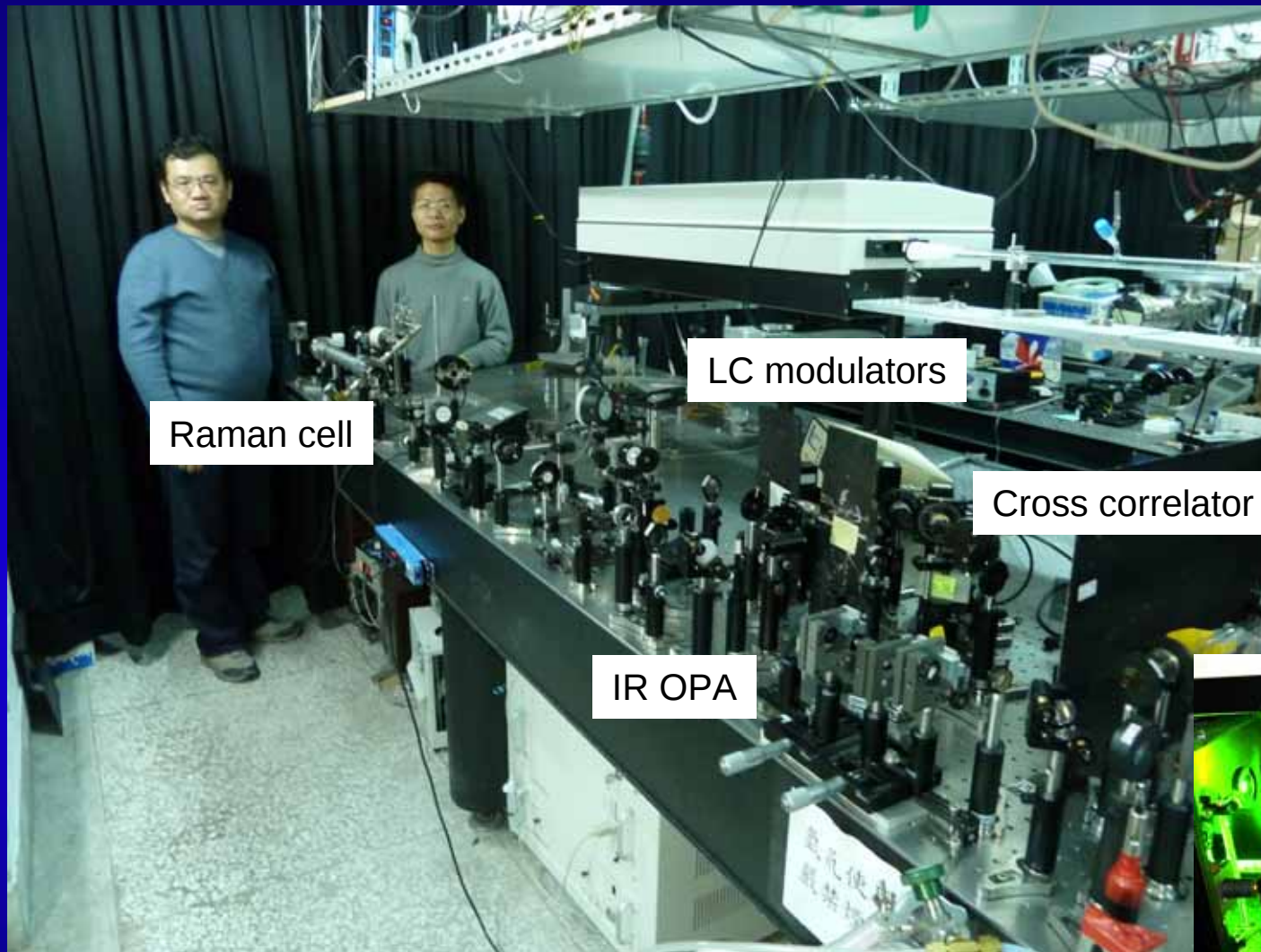
$$\lambda_1 = 2406 \text{ nm} = \text{Raman resonance} - \delta\lambda$$

$$\lambda_2 = \text{second harmonic} = 1203 \text{ nm}$$





Optical waveform synthesizer set up





Harmonic frequency combs contain the **fundamental frequency** component that sets the periodicity and have **multi-octave** spectral width with a mode spacing of tens to hundreds of THz. These are **ideal** conditions for **arbitrary optical waveform** synthesis.

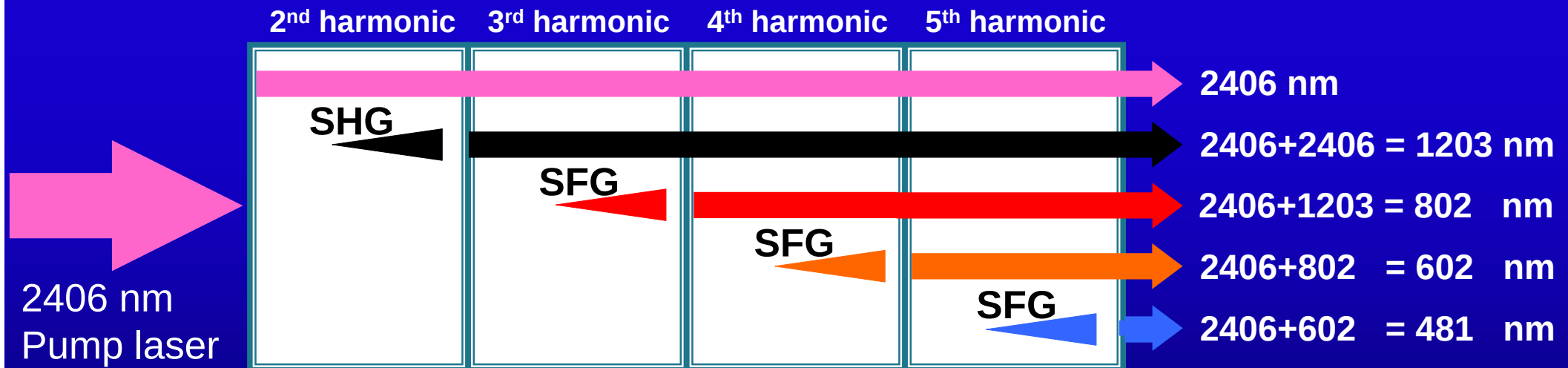




Multiple harmonic generation in a monolithic crystal

$$\omega_n = \omega_0 + n\omega_m$$

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



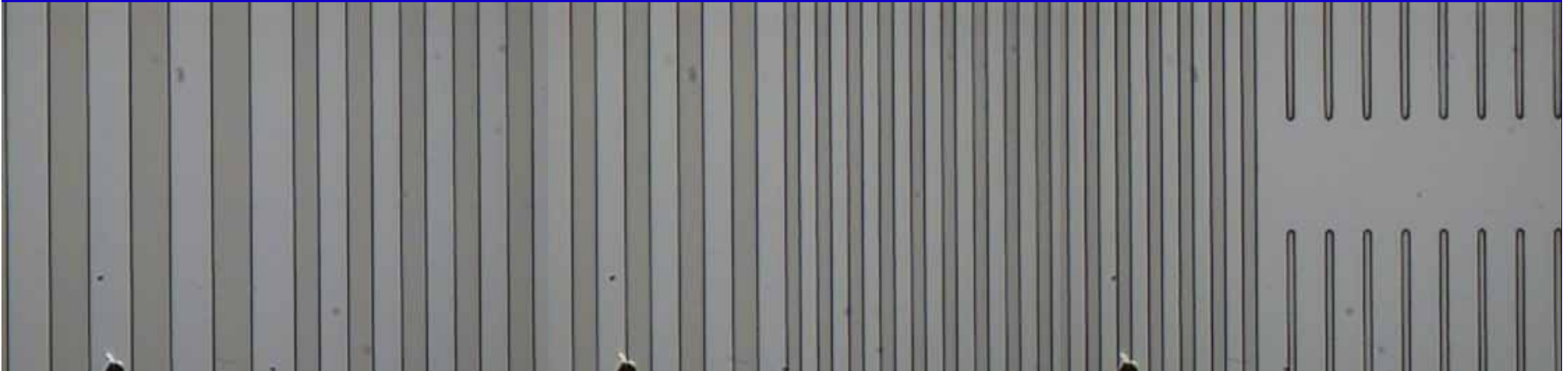


The poled domain periods in the crystal

 2406 nm pump laser	34.09	22.44	13.18	16.01	(μm)
				15.99	
				15.95	
				15.90	
			13.14	16.01	
				15.99	
				15.95	
				15.90	

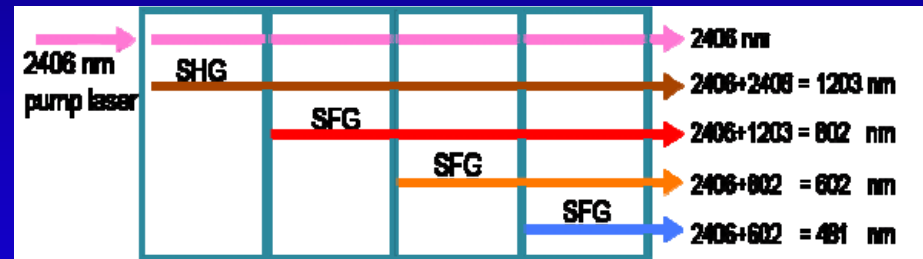
Cascaded QPM has a tight (10 nm) tolerance on domain widths

Photolithography

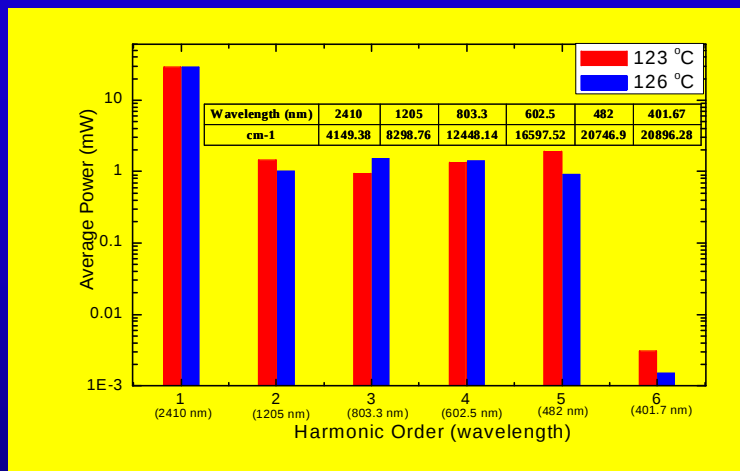




Multiple harmonics $\omega_n = n\omega_1$ are also generated in a QPM Xtal:



Cascaded QPM processes for multi-harmonics generation



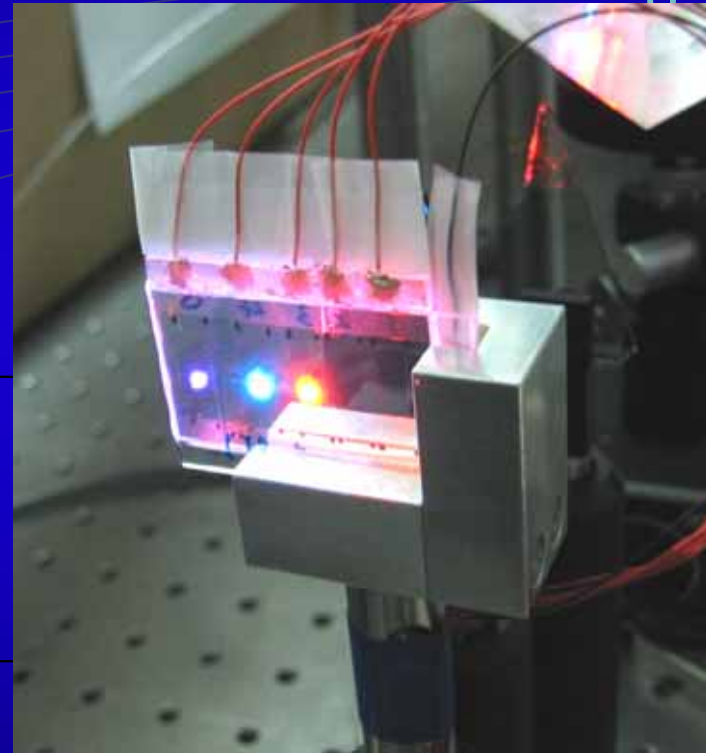
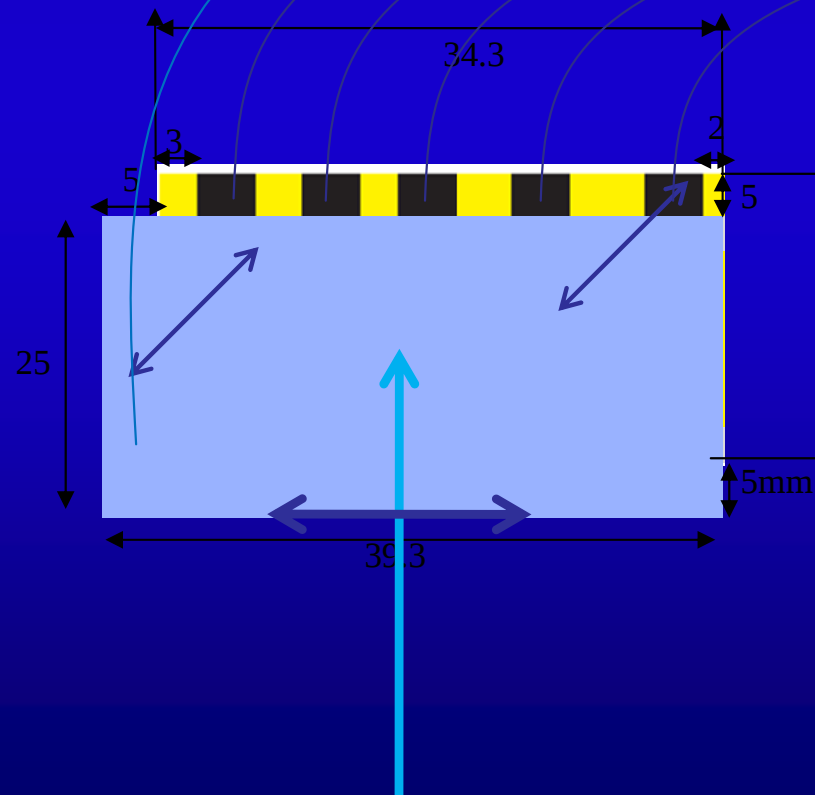
Up to the 7th harmonic has been observed

20% of the fundamental power is converted to the harmonics



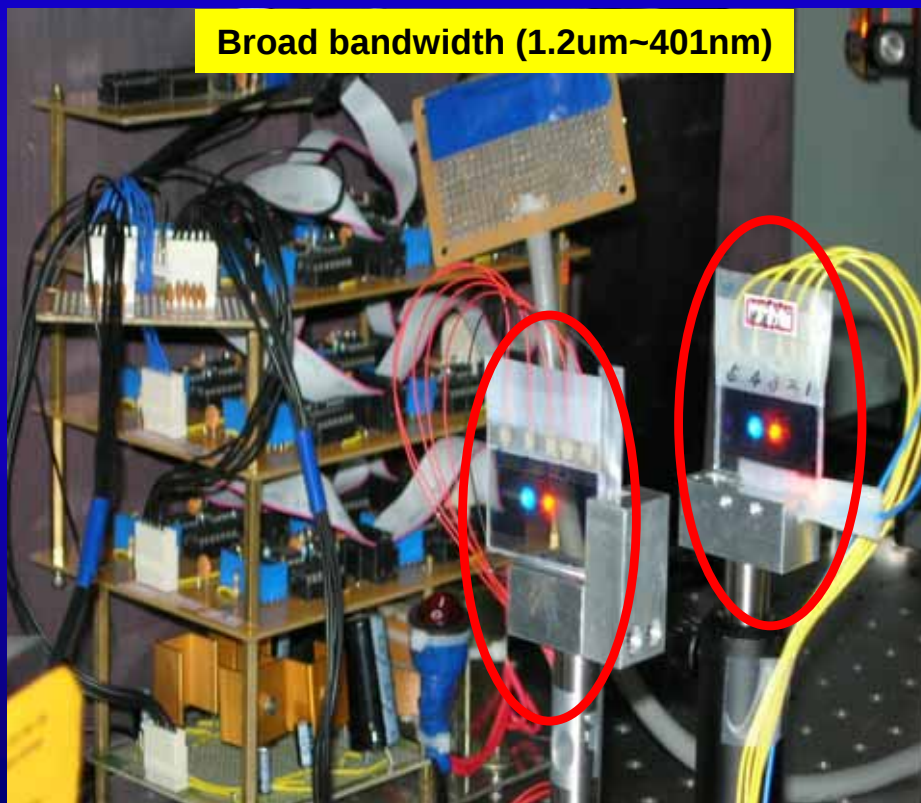


Liquid crystal spatial light modulator

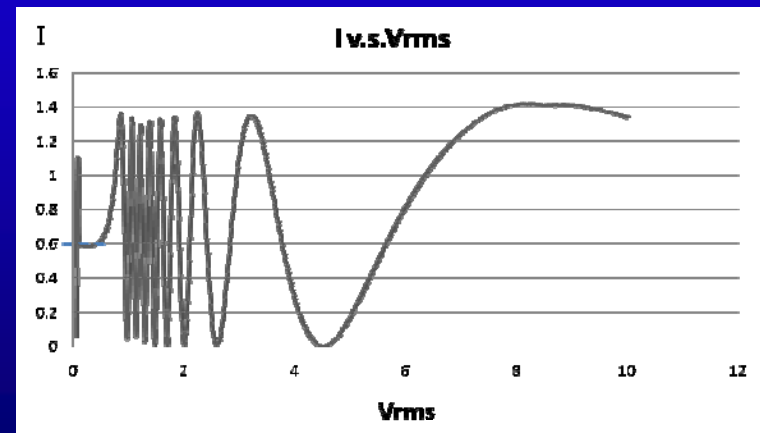




The essential component: spatial liquid crystal modulators
For this idea to work we must have modulators that are broadband and can be configured to modulate either the phase or the amplitude of the input beam. We custom-make these modulators to match the size and spacing of our beam components. The LC material is E7 from Merck.



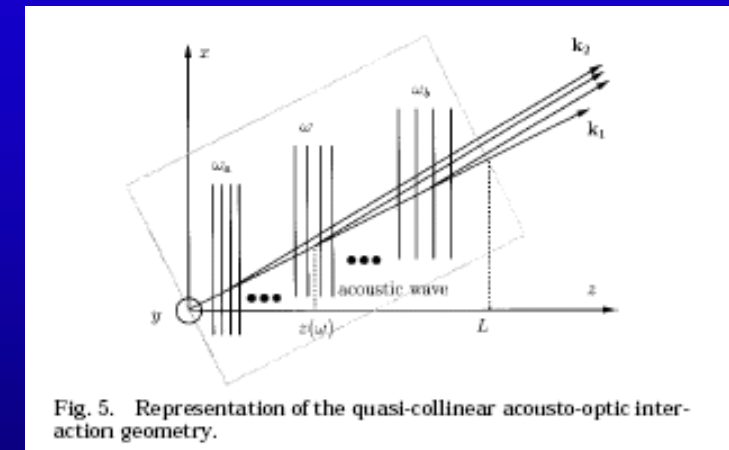
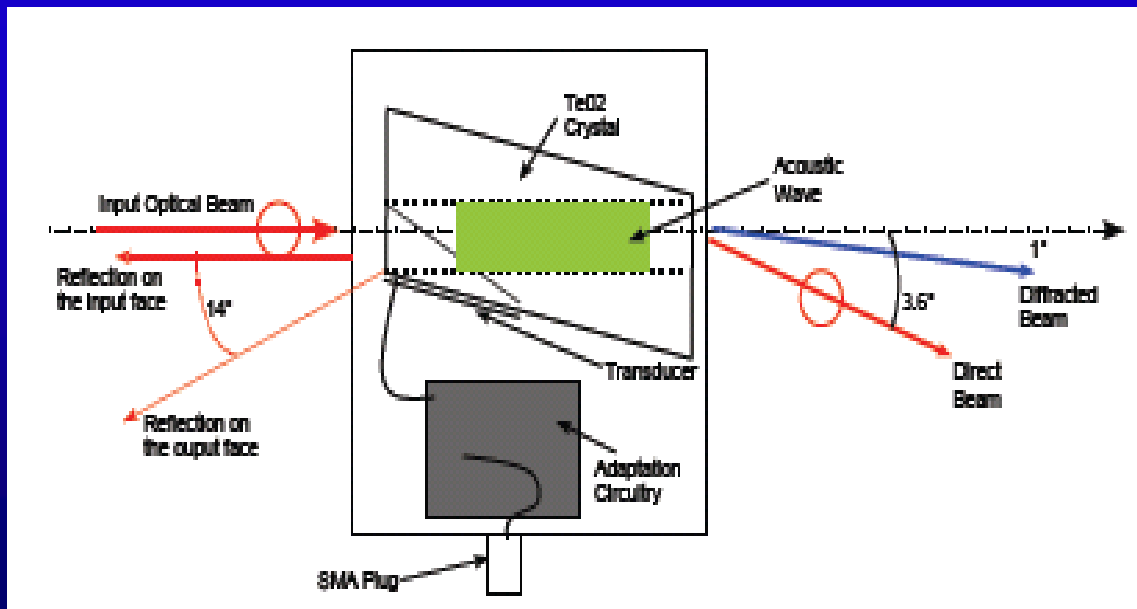
Now, our home-made liquid crystal modulator can cover wavelength from 2.4um to 401nm. Modulator for wavelength short than 344nm is now under testing.



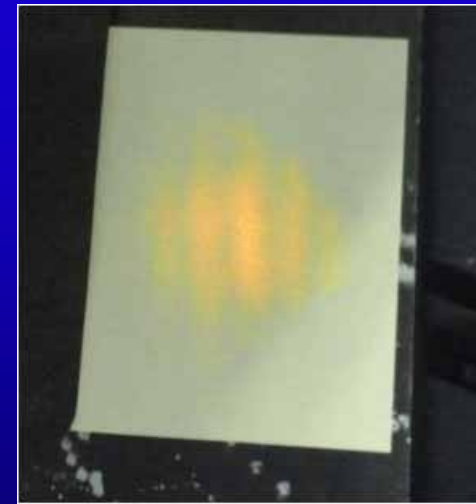
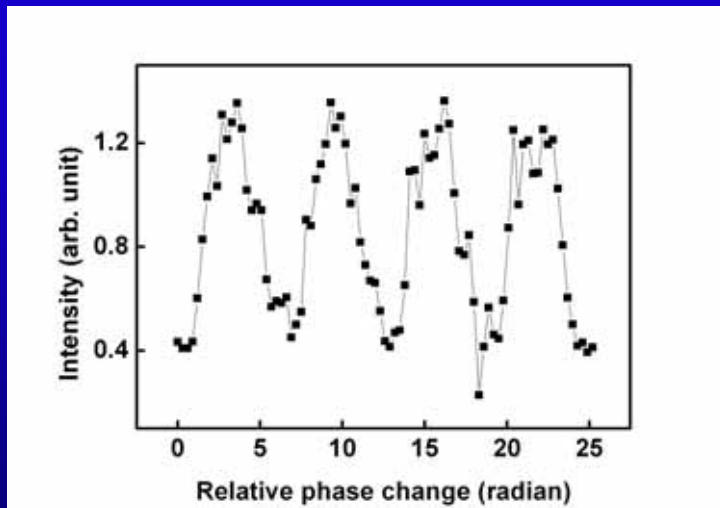
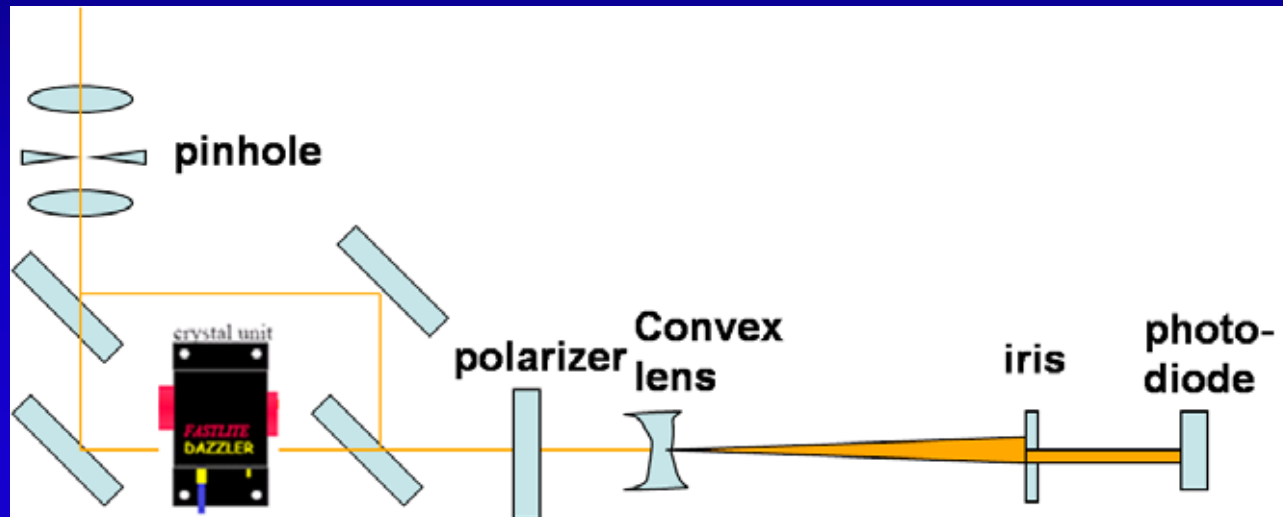
quasi-longitudinal vs transverse control

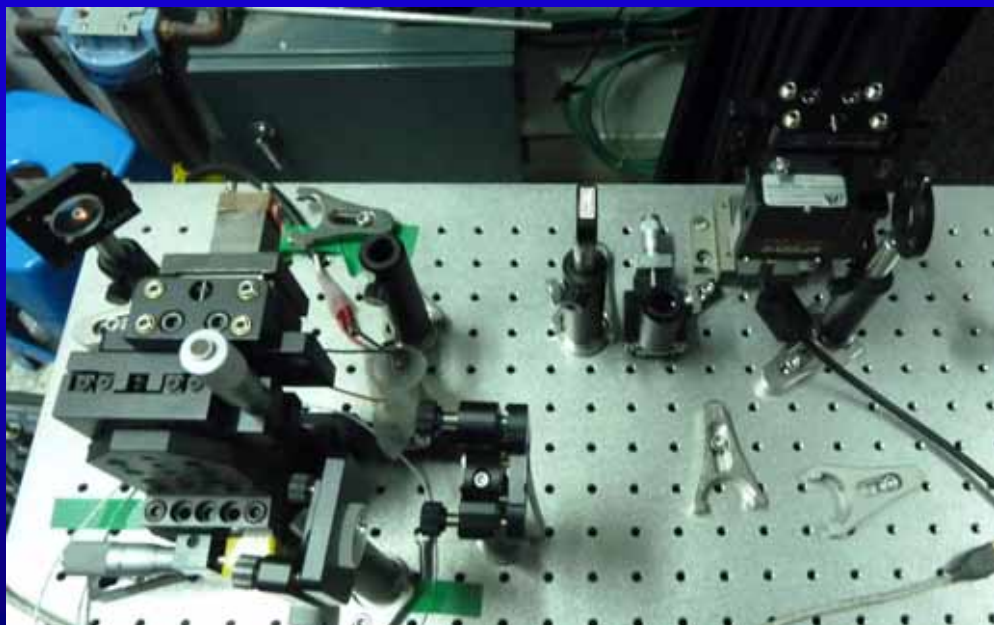
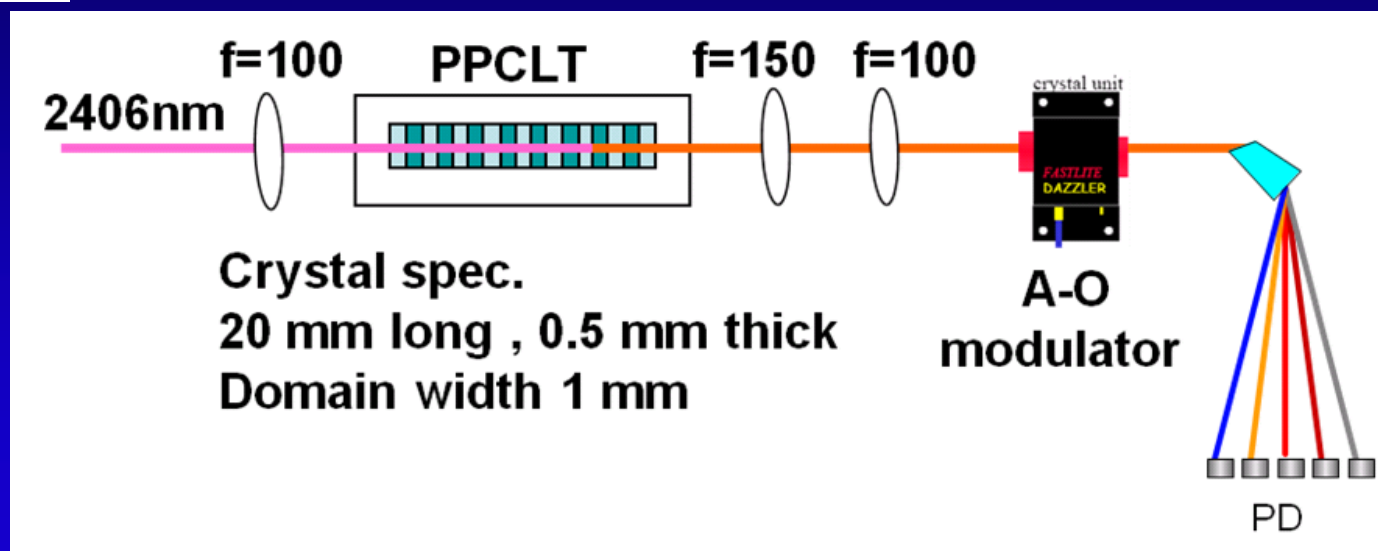
ACOUSTO OPTIC PROGRAMMABLE DISPERSIVE FILTER (AOPDF)

Acousto-optic amplitude and phase modulation



Mach-Zehnder interferometer to test and adjust phases





Breadboard set up



UB-AOM





Comparison of ways of generating a frequency comb for waveform synthesis

1. Broad continuous laser spectrum

isolated pulses -- important for pump-probe experiments

poor shaping resolution -- cannot get arbitrary shapes easily

2. Electro-optic modulation

tens to hundreds of comb lines -- high waveform shaping fidelity

insufficient bandwidth -- shaping of envelopes only

3. Harmonic comb

multi-octave comb

field shaping achieved

limited number of lines

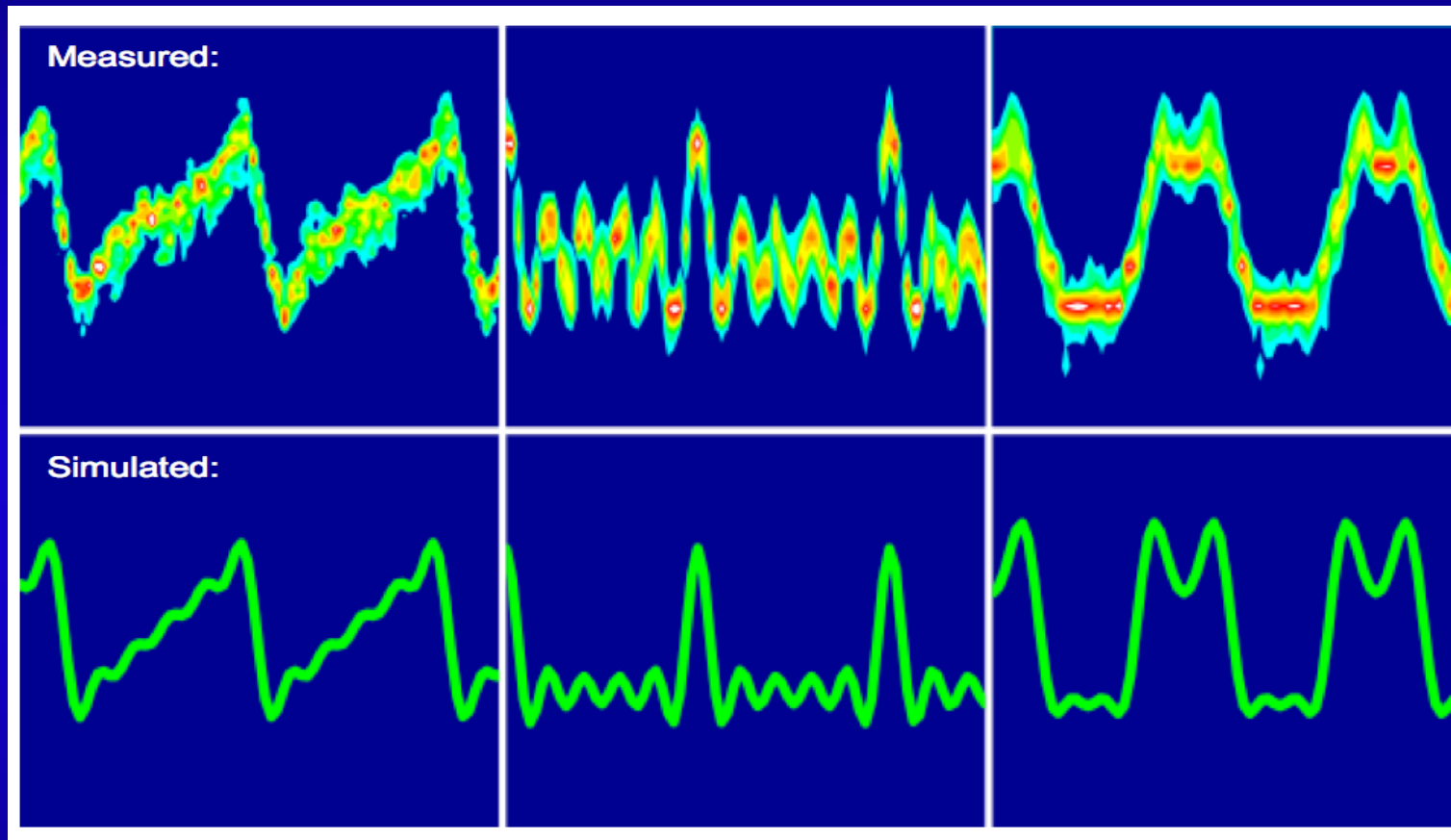




Synthesized waveforms



Instantaneous field waveforms synthesized with 5 harmonic components



Saw tooth

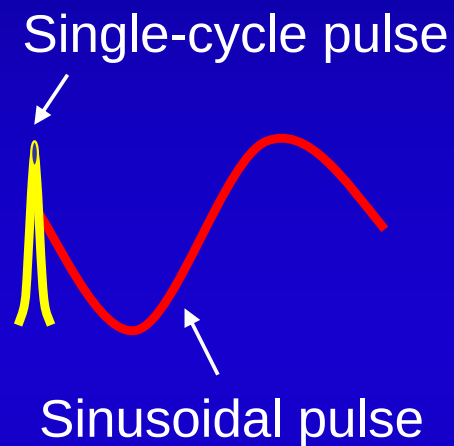
Single-cycle cosine

Square wave

Peak to peak spacing: 8.02 fs
Narrowest field FWHM: 843 as
Envelope width: 1.5 fs



All optical waveform characterization



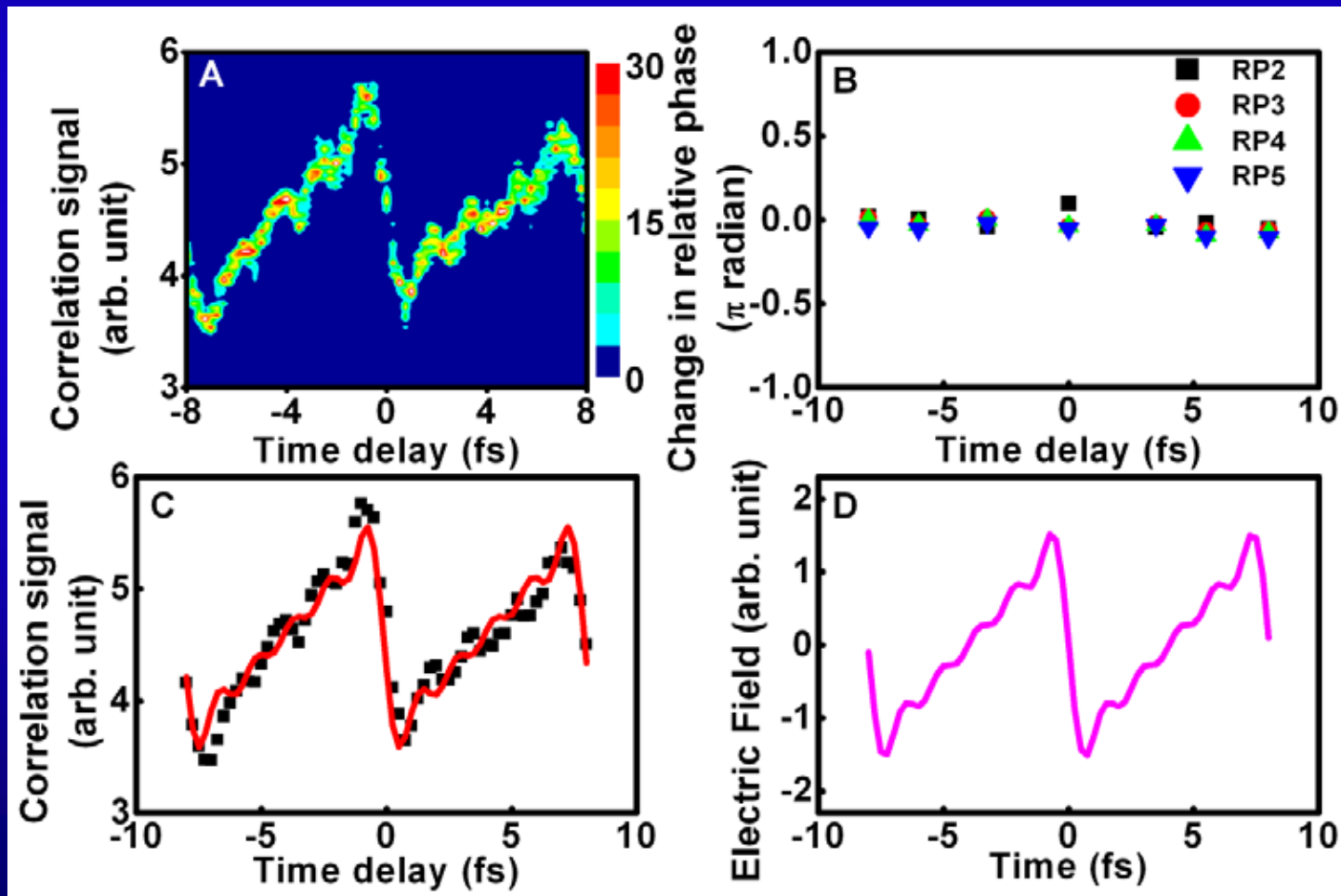
Shaper-Assisted Linear Cross-Correlation for optical field retrieval

SALC²



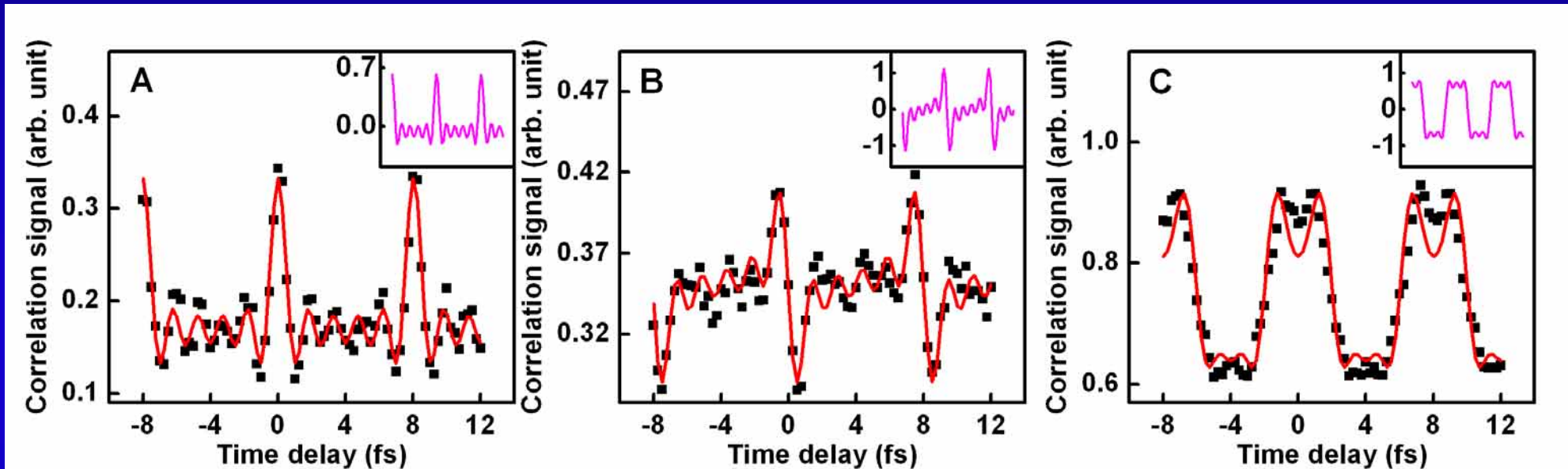
Sawtooth waveform synthesized with 5 harmonic components

relative amplitudes $\{1, 1/2, 1/3, 1/4, 1/5\}$ and CEP = $-\pi/2$





Instantaneous field waveforms synthesized with 5 harmonic components



Single-cycle cosine

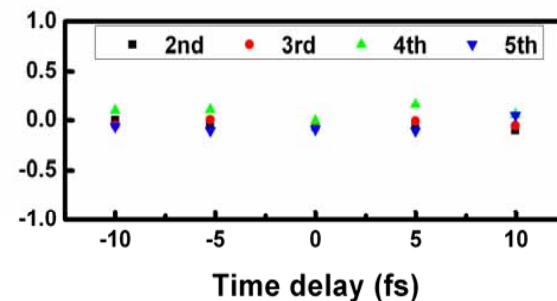
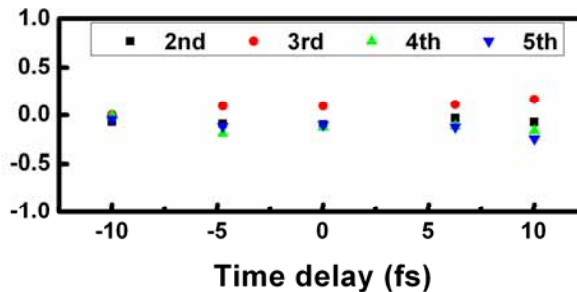
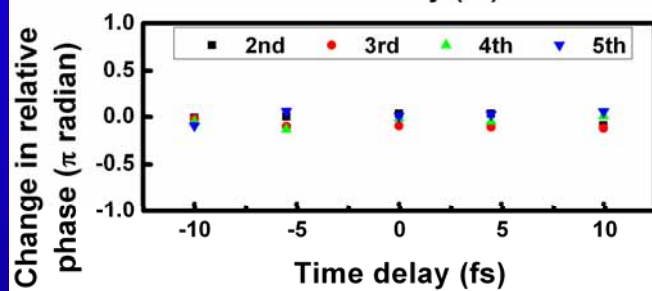
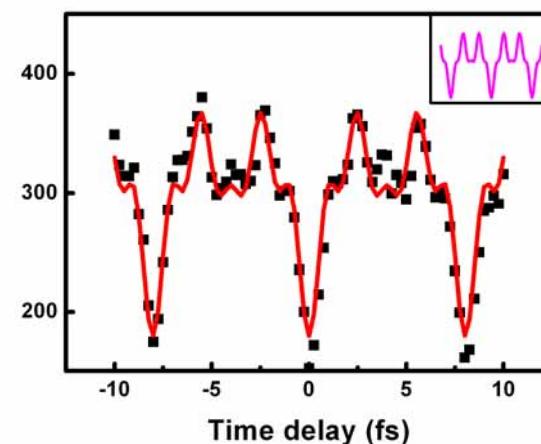
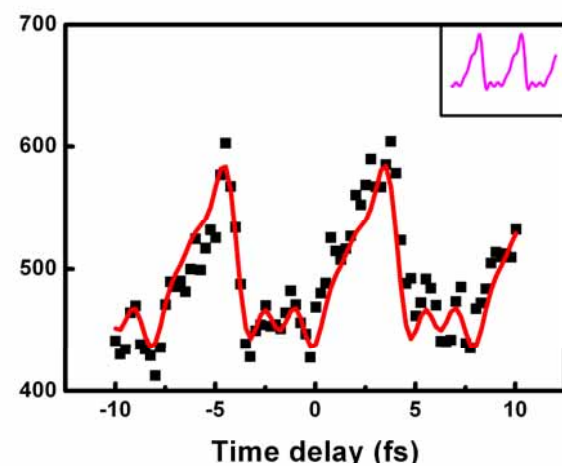
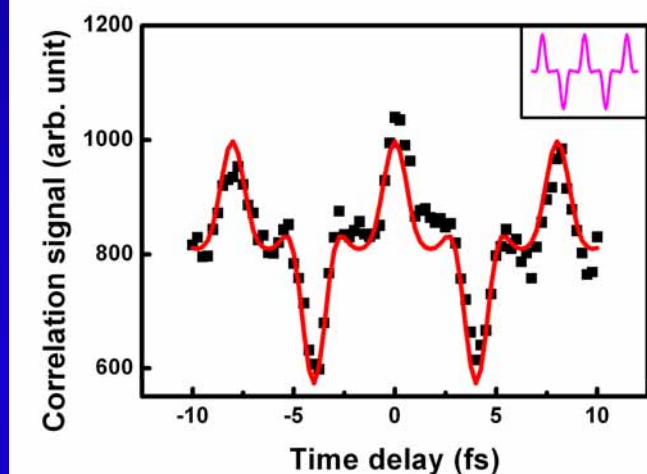
Single-cycle sine

Square wave

Peak to peak spacing: 8.02 fs
Narrowest field FWHM: 843 as
Envelope width: 1.5 fs



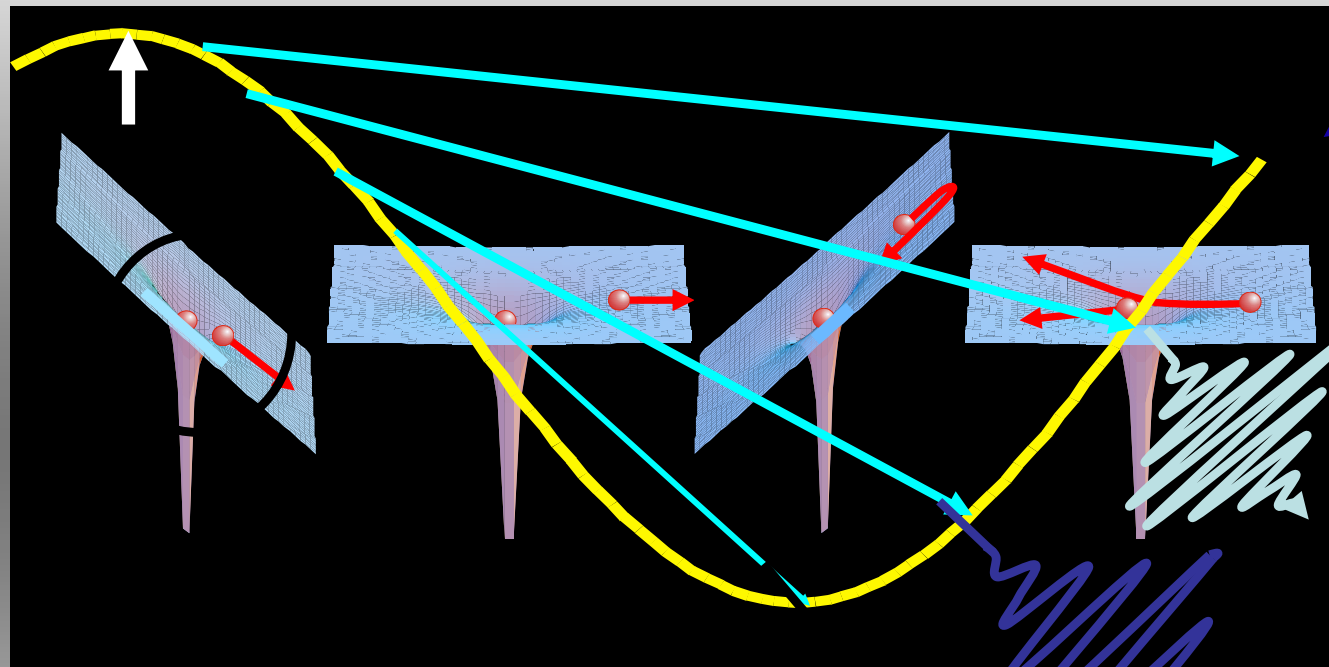
Arbitrary waveforms (correlation traces shown)





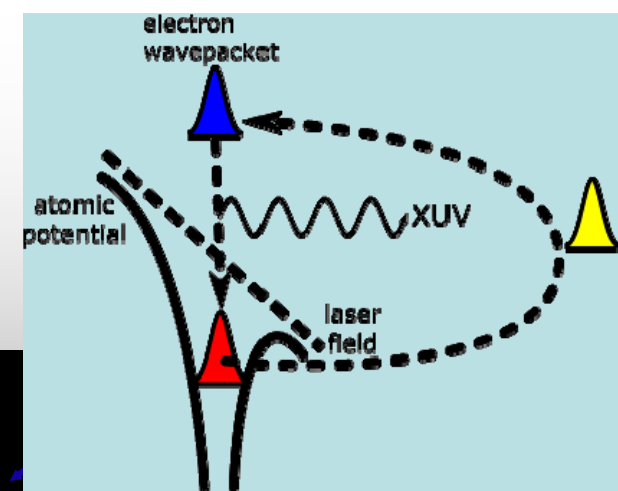
Application of arbitrary waveforms to high harmonic generation (HHG)

The key attosecond idea



Attoseconds
arise first
here

Coherence is transferred from
photon to electron to photon to.



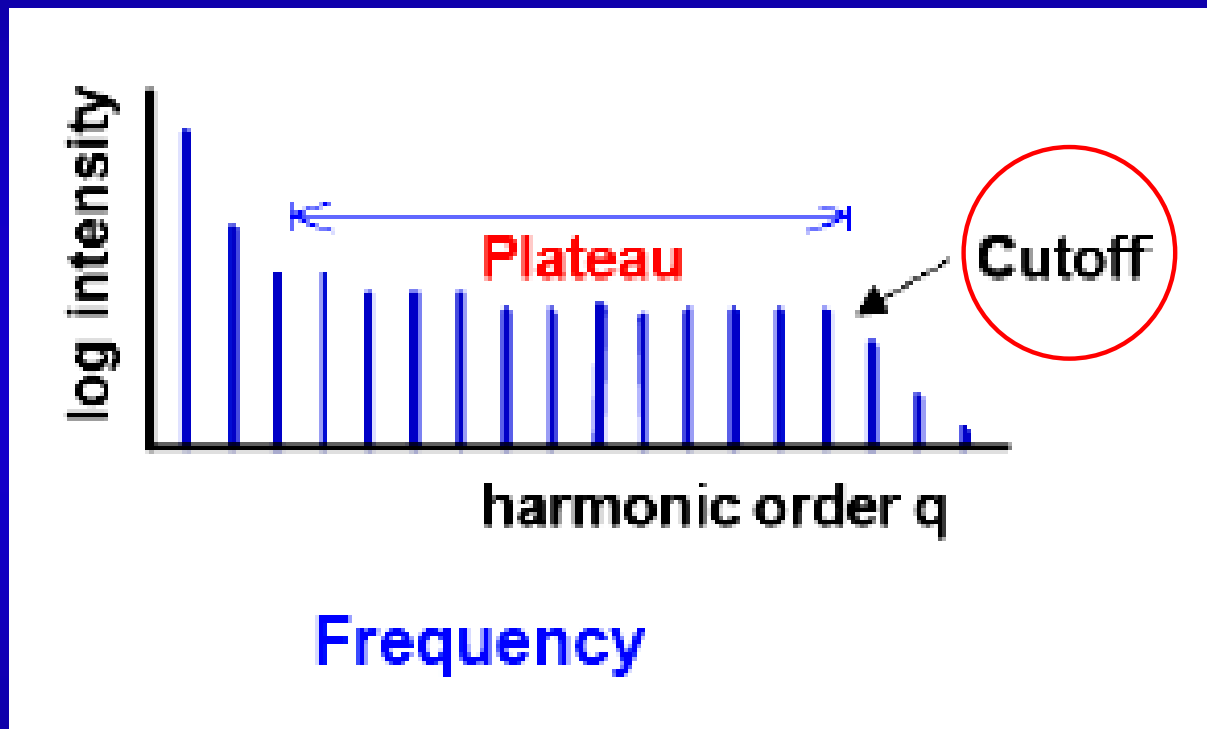
~200 eV

~10 eV



Generating attosecond pulses

High-order harmonic generation



$$h\nu_{\text{cutoff}} = I_p + 3.2U_p$$

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

Ponderomotive potential



Sinusoidal field is not optimized for HHG

PRL 102, 063003 (2009)

PHYSICAL REVIEW

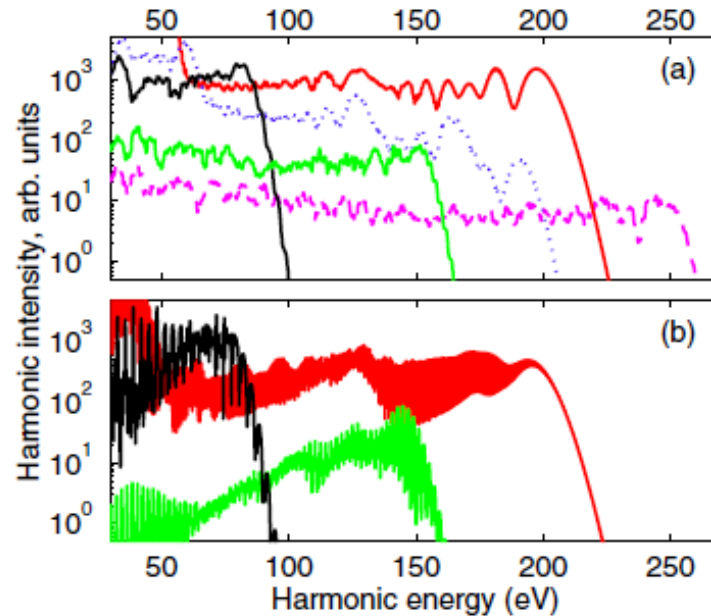
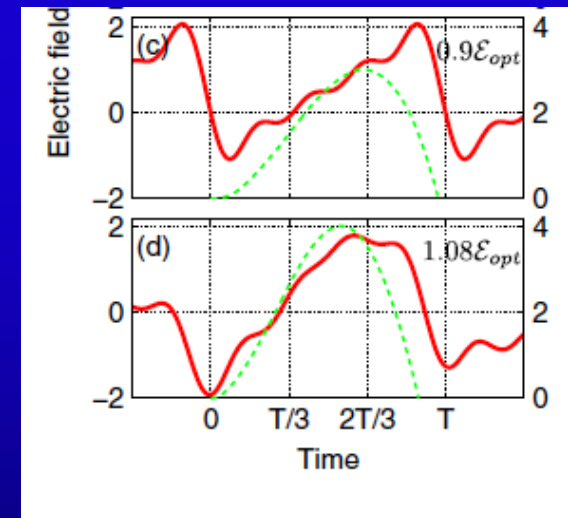


FIG. 2 (color online). Calculated HHG spectra generated by pure sinusoidal fields with wavelengths of 800 nm plotted as a solid black line, 1200 nm as a solid light gray (green) line, and 1600 nm as a dashed (pink) line; the optimized waveform plotted as solid dark gray (red) line; and a field composed solely of the 800 nm and 1600 nm components of the optimized field plotted in a dotted (blue) line. All spectra were generated using pulses of the same duration and pulse energy, normalized to a 16 fs FWHM 800 nm pulse with a peak intensity of $3 \times 10^{14} \text{ W cm}^{-2}$. (a) Single active electron calculations. For clarity a 5 eV wide moving average has been used. (b) Full propagation calculations after radial integration.

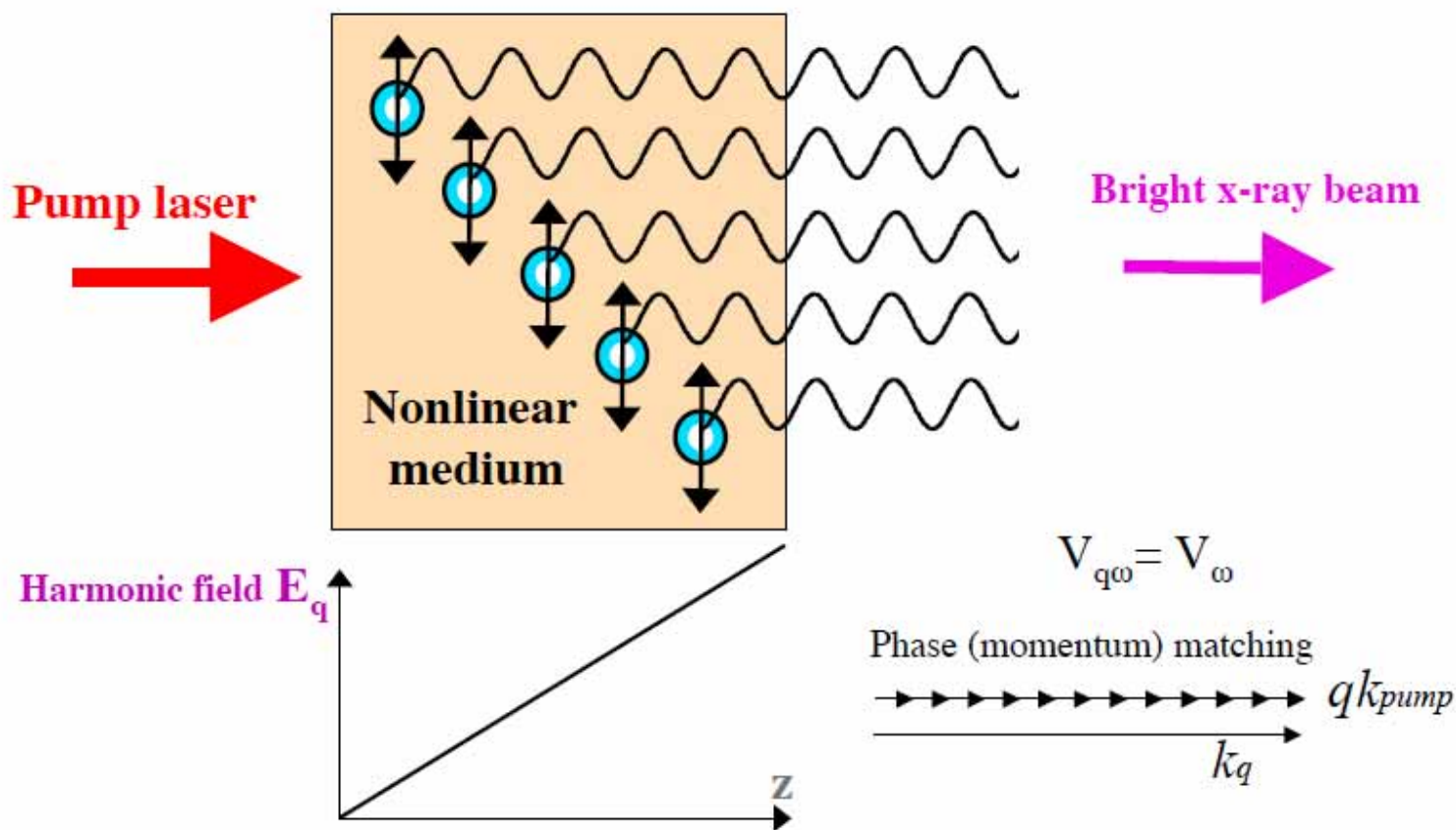
Black:
sinusoidal field

Red:
optimized field





Second challenge - phase matching for bright x-rays

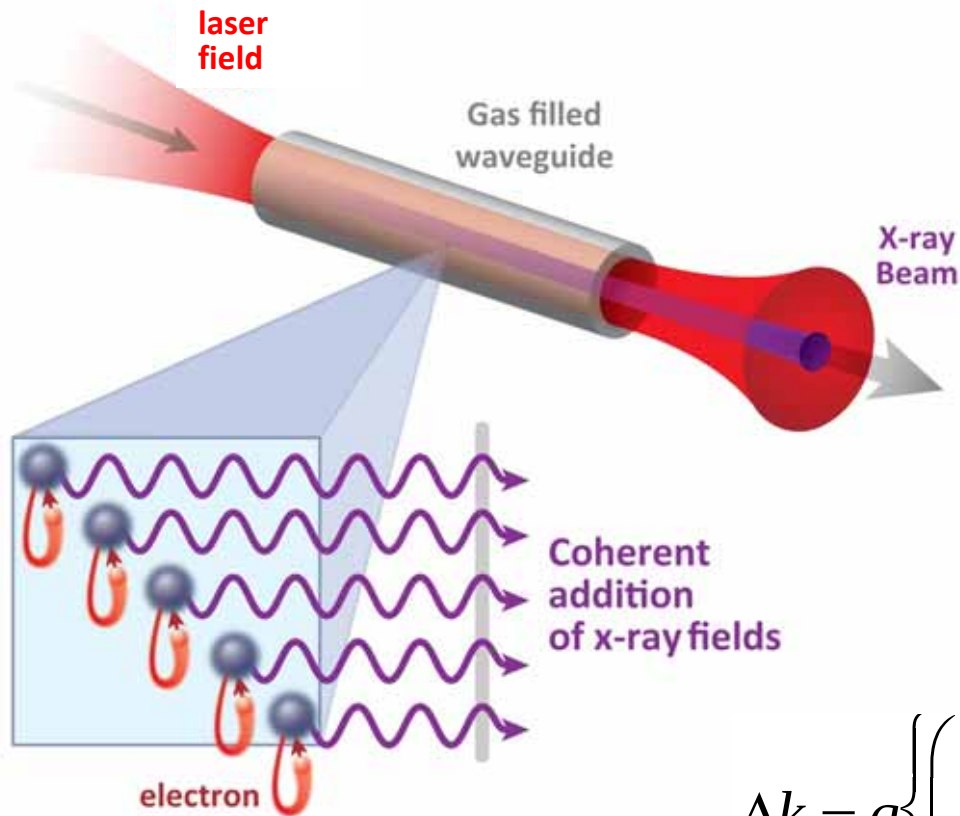


Phase matching - the emission from all atoms interfere constructively!

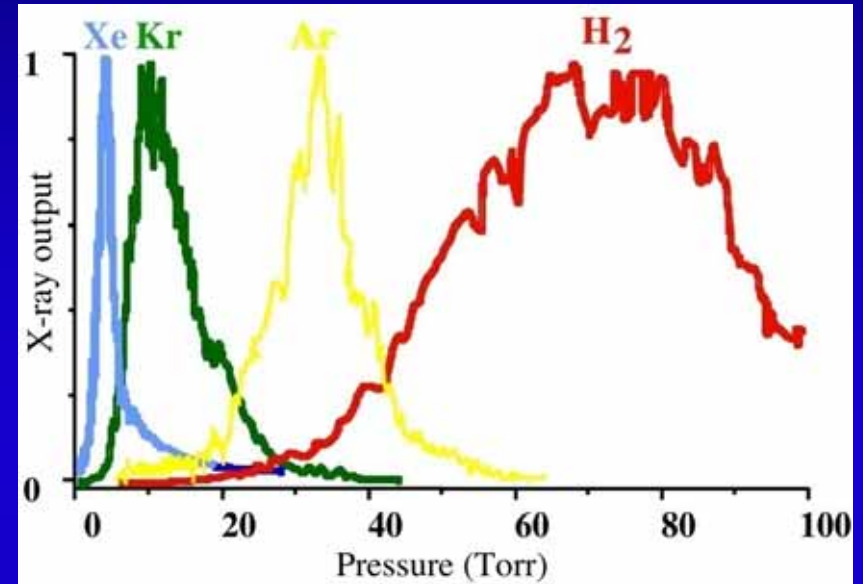




Phase matching of high harmonic beams



$$V_{\text{Laser}} = V_{\text{X-ray}} = c$$



$$\Delta k = qk_{\text{laser}} - k_{\text{HHG}} = 0$$

$$\Delta k = q \left\{ \left(\frac{u_{11}^2 \lambda_0}{4 \pi a^2} \right) - P \left((1 - \eta) \frac{2 \pi}{\lambda_0} \Delta \delta - \eta [N_{\text{atm}} r_e \lambda_0] \right) \right\}$$

Waveguide

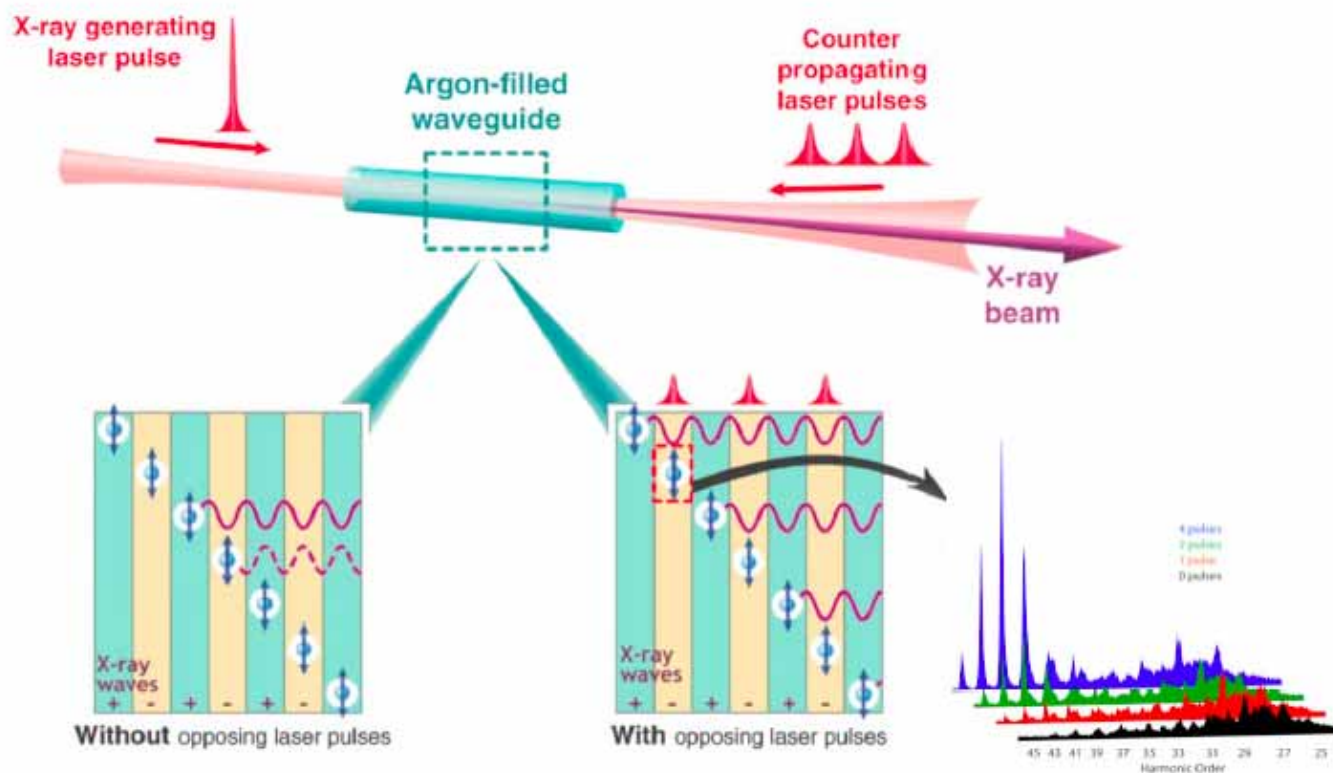
Neutrals

Plasma





All-optical quasi phase matching

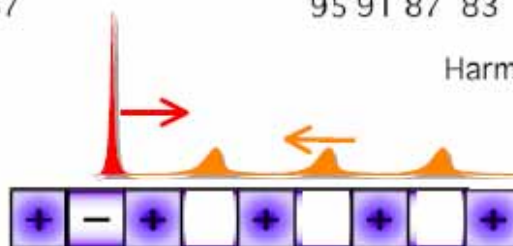
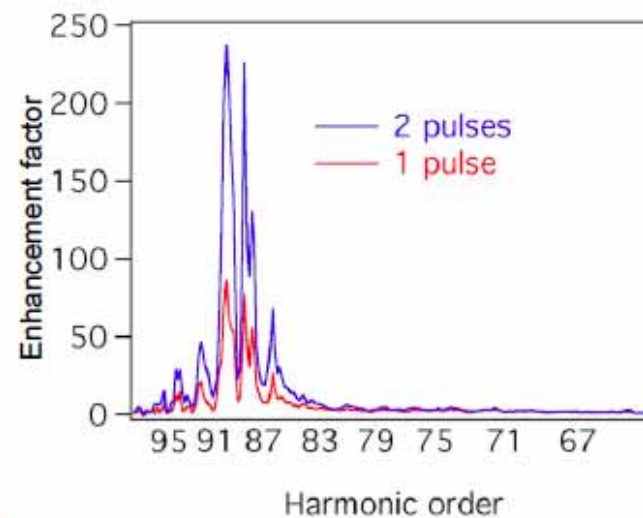
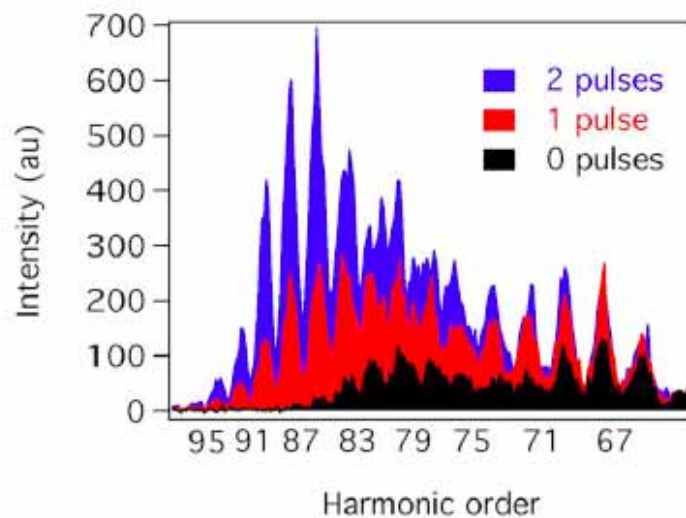


PRL 98, 123904 (2007); Nature Phys. 3, 270 (2007); Science 317, 775 (2007); CThX1





All-optical quasi phase matching at 70eV and 140 eV



- $L_c \sim 1/q$ so coherence length changes with harmonic order
- Observe *selective* enhancement HHG when $L_c \approx$ pulse separation!
- No enhancement where conventional phase matching possible

Lytle et al, *Optics Letters* 33, 174 (2008); Zhang et al, *Nature Physics* 3, 270 (2007)

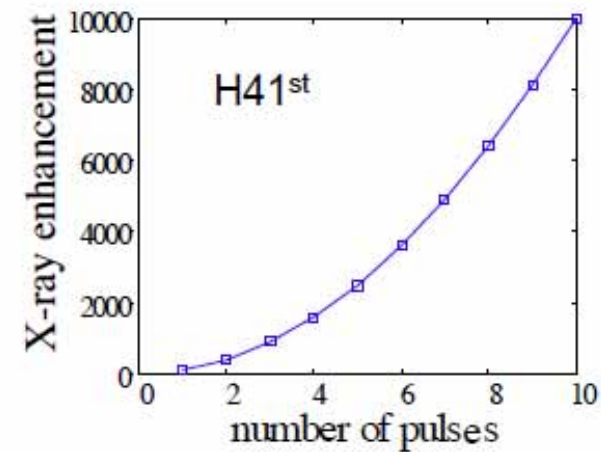
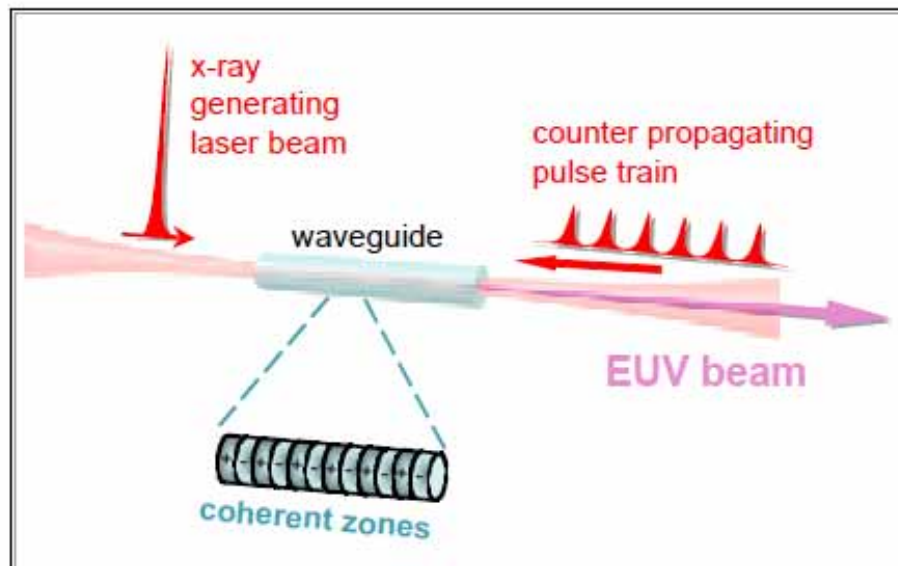


How far can we go?



- HHG in Helium in the water window
 - At a pressure of 5 torr, $L_c \sim 100 \mu\text{m}$
 - Absorption depth @ 300 eV: 10 meters
 - Possible enhancement:

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

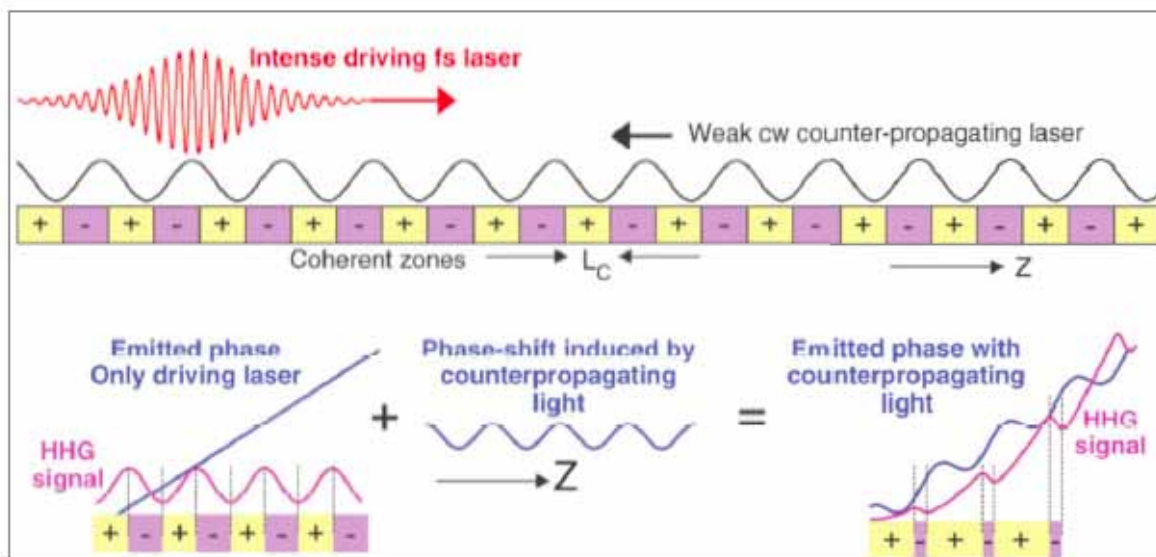




Phase matching at keV photon energies



- All-optical QPM solves phase matching problem even at keV energies!
- Use weak cw, mid-IR, counter-propagating beam in highly ionized regime where coherence lengths are very short ($\approx$ microns)
- Use capillary discharge to reduce defocusing loss at high laser intensity (*J. Rocca*)



**Grating-Assisted
Phase Matching**
Cohen et al., PRL
99, 053902 (2007)



SUMMARIZE

Why we are interested in it - **Frontier research in optical physics**

What is the approach - **Fourier synthesis**

How it is done and what have been done - **harmonic frequency comb + AM/PM + SALC²**

What can be done with these waveforms - **attosecond science and HHG process modification**