Nonlinear optics

IMPRS-UFAST core course

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10-14 December 2018

Organizational notes

- CFEL/bldg. 99, SR O1.060
- 1- week course: 10-14 December 10:00-13:00
- break at 11:20-11:40
- I will show slides and refer to my notes
- I will propose few questions/exercises

Suggested material

- My notes and material
- Boyd, R. W., Nonlinear Optics
- Shen, Y. R., The principles of nonlinear optics
- Dmitriev, V. G., et al., Handbook of Nonlinear Optical Crystals
- Trebino, R., Frequency resolved optical gating
- Notes Franz X. Kärtner
  https://ufox.cfel.de/sites/sites_cfelgroups/site_cfel-ufox/content/e16281/e16715/e16716/e16717/UFST_V1final_2015.pdf
- Material from past courses https://ufox.cfel.de/teaching/

Contents

- Introduction and motivation
- Maxwell’s equations
- Nonlinear optics of 2nd and 3rd order
- Pump-probe technique
- High harmonic generation
- Pulse synthesis

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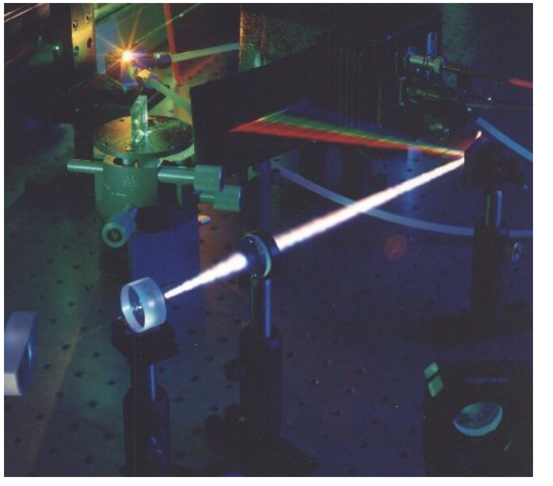
Nonlinear optics

- interaction of light with materials with nonlinear optical response
- photon energy (frequency, wavelength, color) may change
- nonlinear response of outer electrons in the atom potential
- Maxwell's equations with nonlinear polarization vs. the electric field



linear optics:

$$P = \varepsilon_0 \chi^{(1)} E$$

nonlinear optics:

$$P = \varepsilon_0 [\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots]$$

Discovery of nonlinear optics

- NLO started in 1961, with the discovery of SHG from a ruby laser
- not by chance, it started 1 year after the first laser was constructed
- but somebody at the journal did not get the point! ☺

VOLUME 7, NUMBER 4 PHYSICAL REVIEW LETTERS AUGUST 15, 1961

GENERATION OF OPTICAL HARMONICS*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan
(Received July 21, 1961)

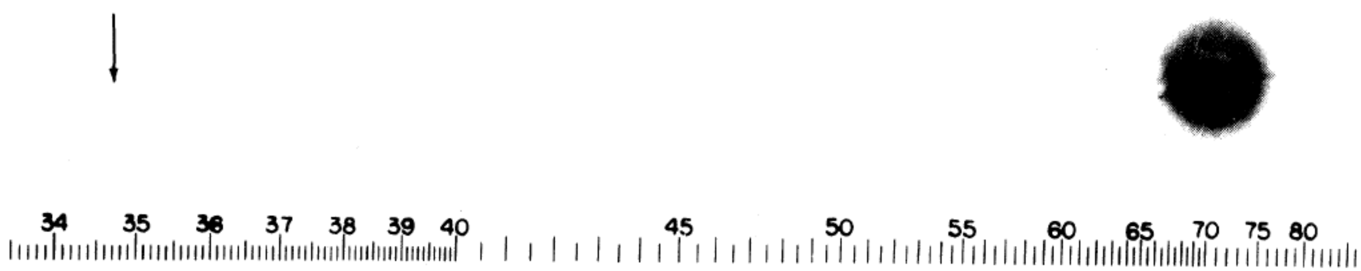
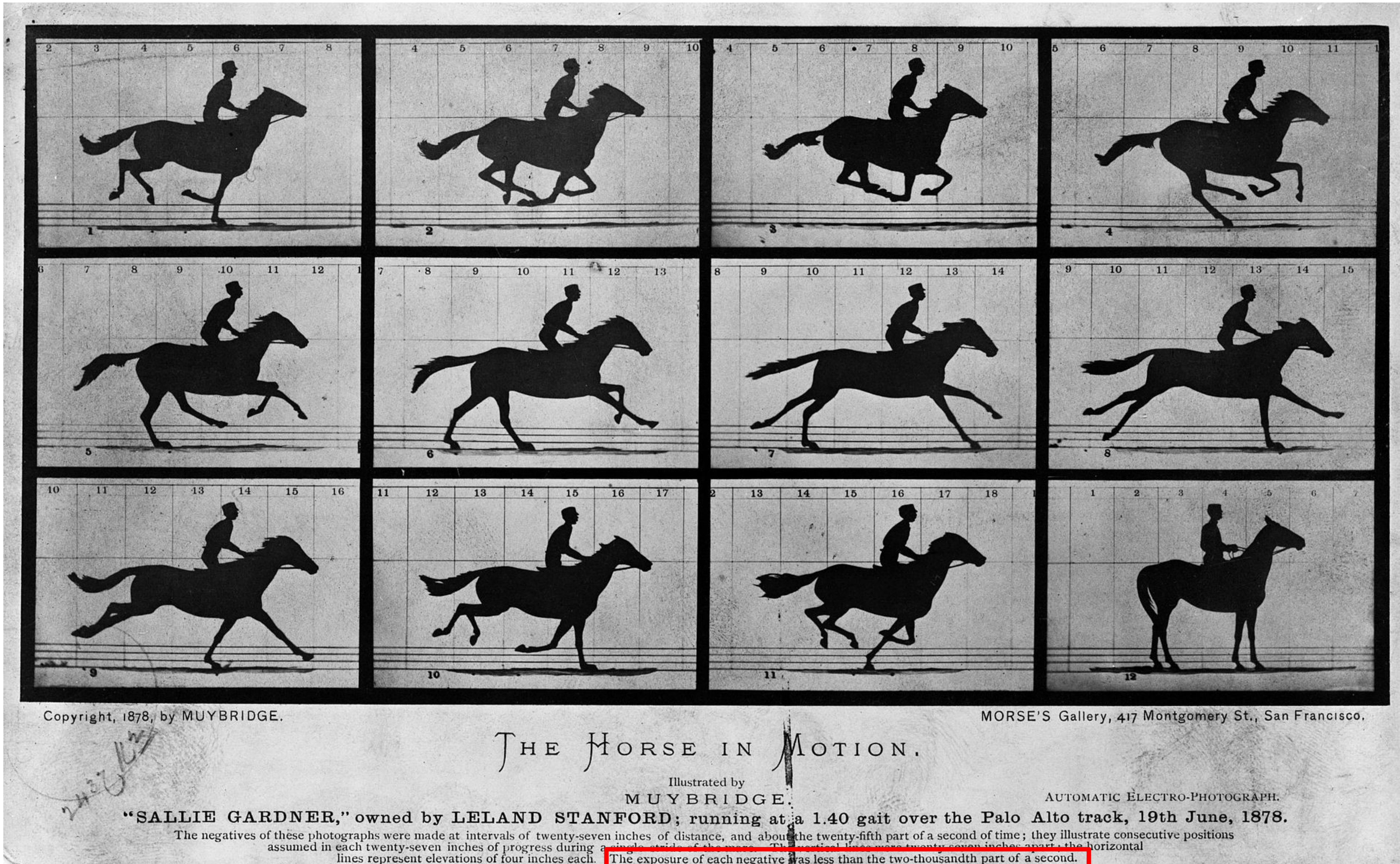


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

The galloping horse



Muybridge's *The Horse in Motion*, **1878**

Femtoseconds and attoseconds

- 1 femtosecond (fs) = 10^{-15} seconds
- 1 attosecond (as) = 10^{-18} seconds
- age of the Universe: 13.8 billion years = 0.435×10^{18} seconds
- there are as many attoseconds in only 2.3 seconds, as many seconds in the whole history of the Universe
- fs and as pulses can be generated through nonlinear optics

fs and as cameras

- How do electrons move?
- How does a laser work?
- How do chemical reactions occur?
- How do we see?
- How does photosynthesis occur?

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Maxwell's equations

Time domain

$$\nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t} + \mathbf{J}$$

Ampere's law

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$$

Faraday's law

$$\nabla \cdot \mathbf{D} = \rho$$

Gauss's law

$$\nabla \cdot \mathbf{B} = 0$$

no magnetic monopoles

electric displacement

$$\boxed{D=\varepsilon_0 E+P}$$

magnetic field

$$\boxed{B=\mu_0 H+M}$$

light speed in vacuum $\boxed{\varepsilon_0\mu_0=1/c^2}$

$\varepsilon_0=8.85\text{x}10^{-12}$ F/m

$\mu_0=4\pi\text{x}10^{-7}$ N/A^2

$c=3\text{X}10^8$ m/s

see notes page 5-9

Wave equation

- calculate curl of Faraday's law
- no free charges, no free currents, no magnetization
- neglect nonlinear polarization vs. linear
- plane waves

$$\underbrace{\overbrace{\frac{\partial^2 E}{\partial z^2} = \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2}}^{\text{in vacuum}} + \mu_0 \frac{\partial^2 P}{\partial t^2}}_{\text{in materials}}$$

solution in vacuum: $E = E(z \pm ct)$.   **-> demonstration (notes page 9)**

Sinusoidal waves in vacuum

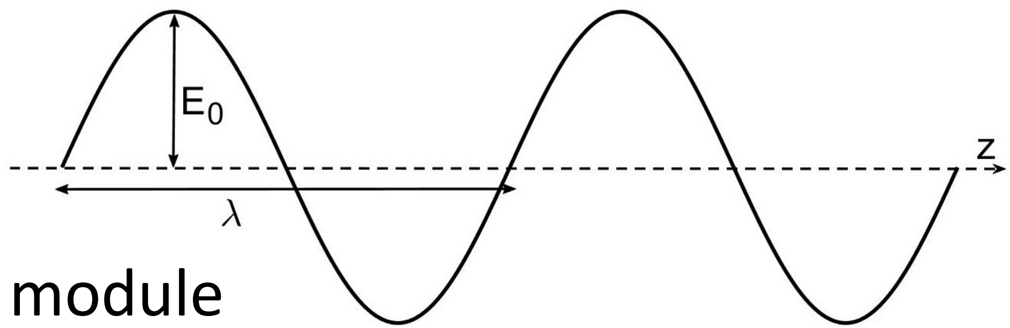
$$E = E_0 \cos\left(2\pi \frac{z}{\lambda} - 2\pi\nu t+\varphi\right) =$$

$$= E_0 \,\mathrm{Re}\left\{\exp\left[\, i\left(2\pi \frac{z}{\lambda} -2\pi\nu t+\varphi\right)\right]\right\}$$

- in space:

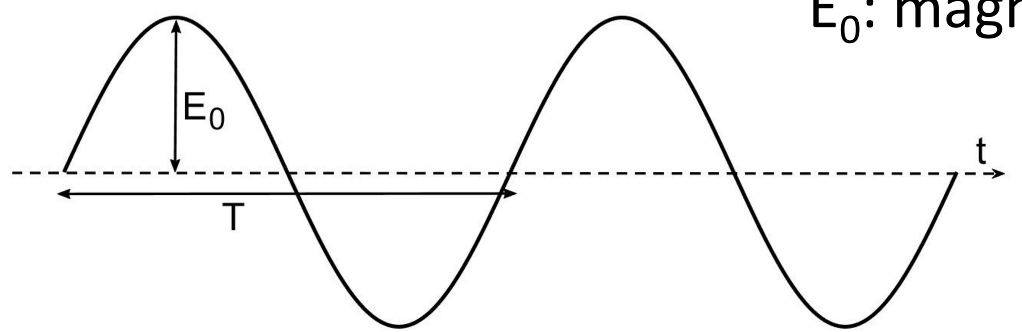
λ: wavelength

$k=2\pi/\lambda$: wavevector module



E_0: magnitude

- in time:



T: period

$\nu=1/T$: frequency

$\omega=2\pi\nu$: angular frequency

$$\boxed{\lambda\nu=c}$$

EM wave

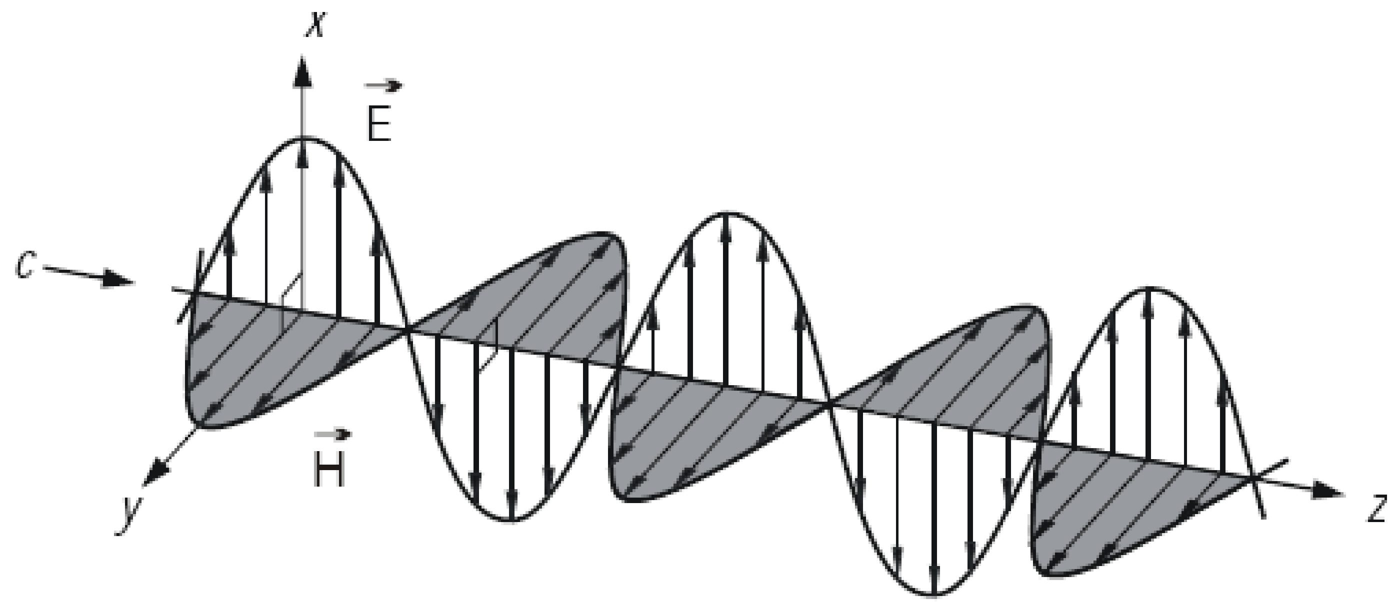


Figure 2.1: Transverse electromagnetic wave (TEM) [6]

H. A. Haus, ”Fields and Waves in Optoelectronics”, Prentice Hall 1984

Linear media

wave equation

$$\frac{\partial^2 E}{\partial z^2} = \frac{1}{c^2} n^2 \frac{\partial^2 E}{\partial t^2}$$

linear polarization

$$P = \varepsilon_0 \chi^{(1)} E$$

plane wave solution

$$E = E_0 \cos\left(2\pi\, \mathbf{n} \frac{z}{\lambda} - 2\pi\nu t + \varphi\right) =$$

$$= E_0 \,\mathrm{Re}\left\{\exp\left[i\left(2\pi\, \mathbf{n} \frac{z}{\lambda} - 2\pi\nu t + \varphi\right)\right]\right\}$$

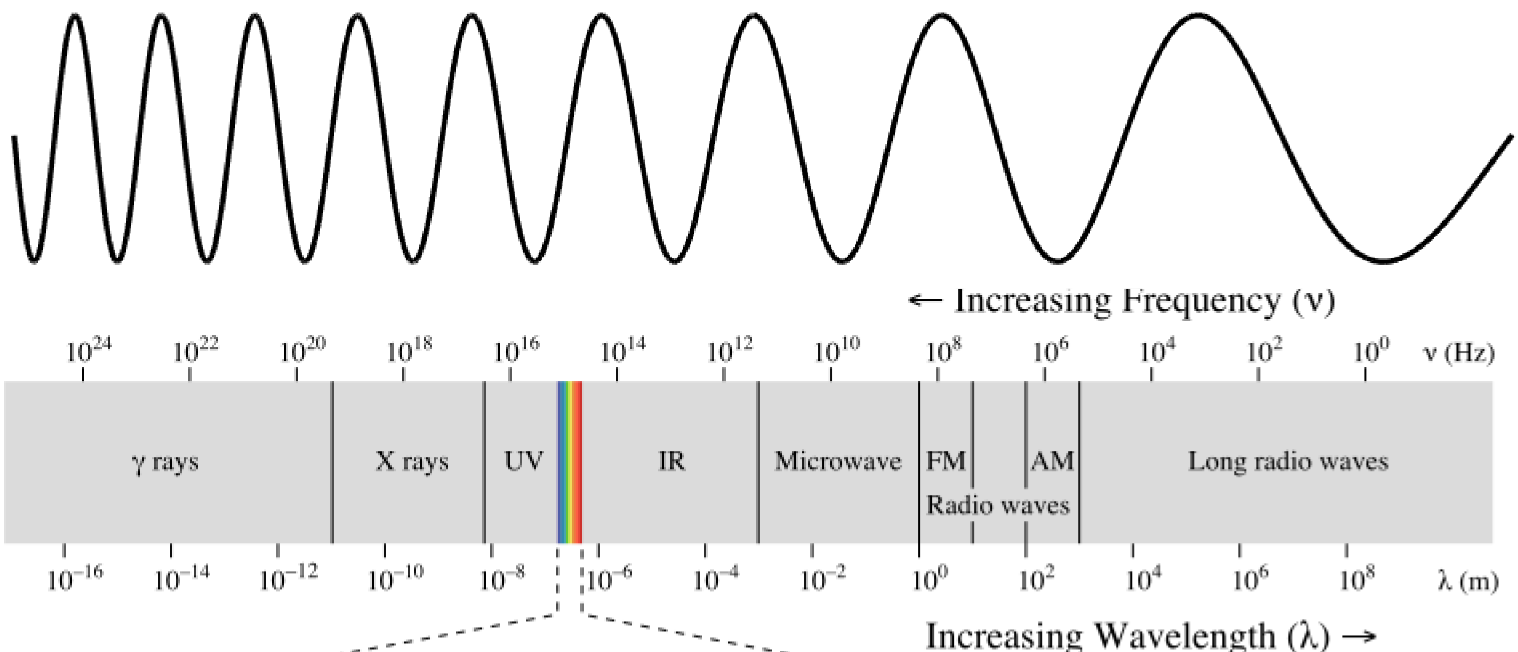
n: refractive index (Sellmeier equations)

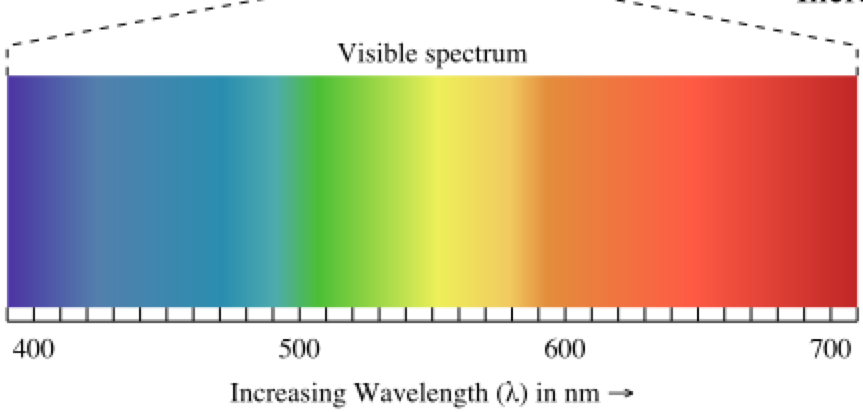
wavelength: λ/n

frequency: ν

speed: c/n

EM spectrum

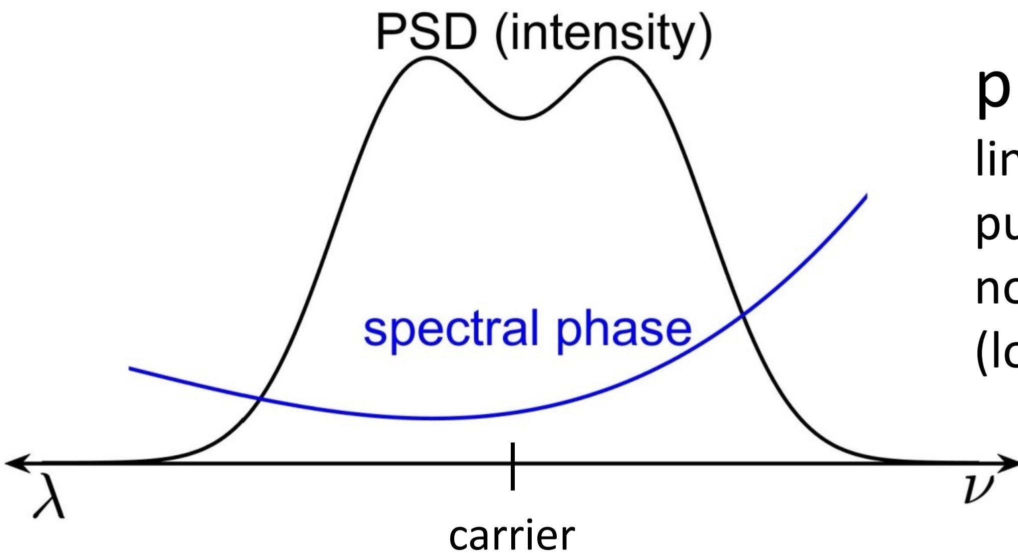




Color	Wavelength	Frequency	Photon energy
Violet	380–450 nm	668–789 THz	2.75–3.26 eV
Blue	450–495 nm	606–668 THz	2.50–2.75 eV
Green	495–570 nm	526–606 THz	2.17–2.50 eV
Yellow	570–590 nm	508–526 THz	2.10–2.17 eV
Orange	590–620 nm	484–508 THz	2.00–2.10 eV
Red	620–750 nm	400–484 THz	1.65–2.00 eV

https://www.khanacademy.org/science/physics/light-waves/introduction-to-light-waves/a/light-and-the-electromagnetic-spectrum

Optical pulses



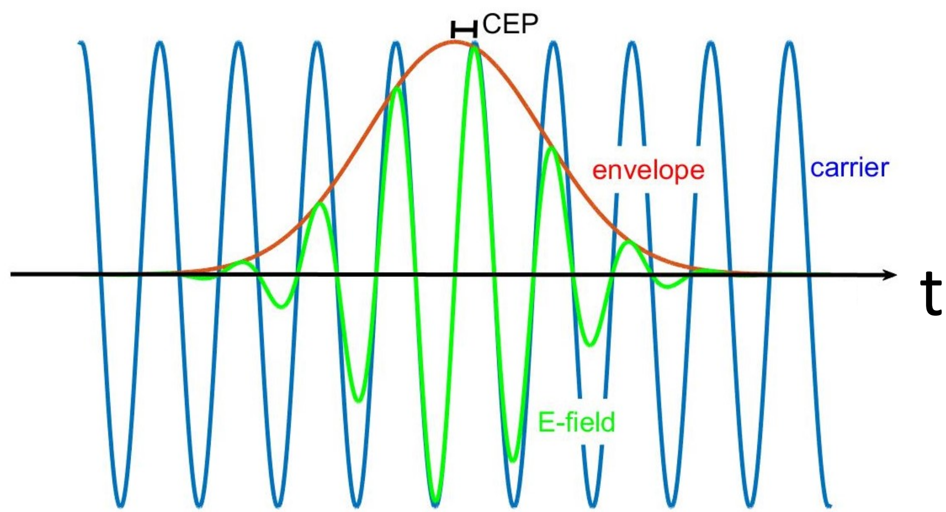
phase dispersion: n=n(ω)

linear phase: transform limited pulse (short)

nonlinear phase: chirped pulse (long)

intensity:

$$I=\frac{1}{2}\varepsilon_0 nc|\mathbf{E}|^2$$



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2^{nd} order nonlinear optics

noncentrosymmetric media

$$P = \varepsilon_0[\chi^{(1)}E + \boldsymbol{\chi^{(2)}E^2} + \cancel{\chi^{(3)}E^3 + \cdots}]$$

-> verify that $|P_{NL}| \ll |P_L|$ (notes page 21)

2^{nd} order nonlinear wave equation

$$\frac{\partial^2 E}{\partial z^2} = \frac{1}{c^2} n^2 \frac{\partial^2 E}{\partial t^2} + \frac{\chi^{(2)}}{c^2}\frac{\partial^2 E^2}{\partial t^2}$$

no analytic solution known

guess: bichromatic field $E = E_1 \exp(i\omega_1 t) + E_2 \exp(i\omega_2 t)$

see notes page 19-22

2^{nd} order nonlinear optics

generated new frequencies:

$$\underbrace{2\omega_1, 2\omega_2,}_{\text{SHG}} \overbrace{\omega_1+\omega_2,}^{\text{SFG}} \underbrace{\omega_1-\omega_2,}_{\text{DFG}} \overbrace{0}^{\text{OR}}$$

2^{nd} order phenomena:

- second harmonic generation (SHG)
- optical parametric amplification (OPA)
- difference frequency generation (DFG)
- sum frequency generation (SFG)
- optical rectification (OR)

Second harmonic generation (SHG)

- start from bichromatic field ($\omega_2 = 2\omega_1$)
- substitute in nonlinear wave equation
- neglect frequencies different than ω_1 and ω_2
- use slowly varying envelope approximation (SVEA)

-> derivation: notes from page 24

coupled equations for SHG

$$\frac{dE_1}{dz} = -i\frac{\omega_1 d}{n_1 c} E_1^{\,*} E_2 \exp(-i\Delta kz) \qquad \boxed{1}$$

$$\frac{dE_2}{dz} = -i\frac{\omega_1 d}{n_2 c} E_1^{\,2} \exp(i\Delta kz) \qquad \boxed{2}$$

$d = \frac{1}{2}\chi^{(2)}$

$\Delta k = k_2 - 2k_1$

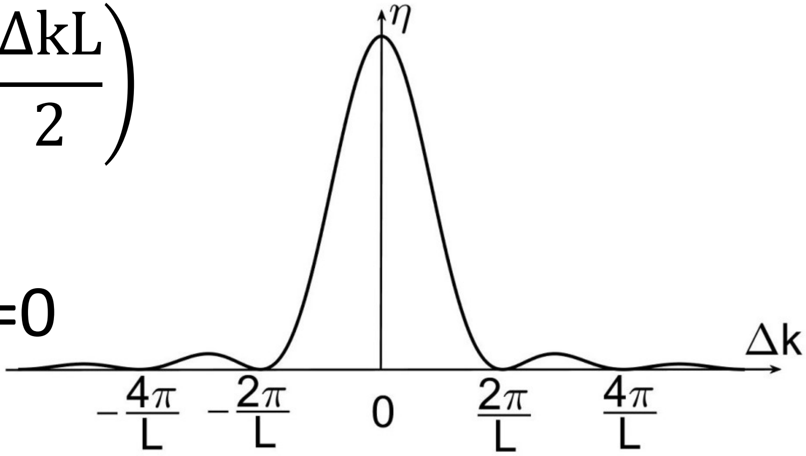
see notes page 24-27

Second harmonic generation

- no pump depletion (E_1=constant)
- integrate equation $\boxed{2}$ upon a distance z=L (crystal length)
- calculate intensity and efficiency

$$\eta = \frac{I_2}{I_1} = \frac{2\omega_1^{\,2} d^2 I_1}{n_2 n_1^{\,2} c^3 \varepsilon_0} L^2 \mathrm{sinc}^2\left(\frac{\Delta k L}{2}\right)$$

- maximum efficiency for Δk=0



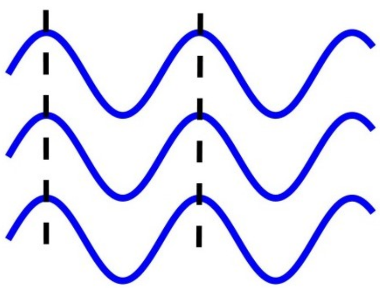
-> exercise: notes page 30. Required laser intensity for SHG

see notes page 27-31

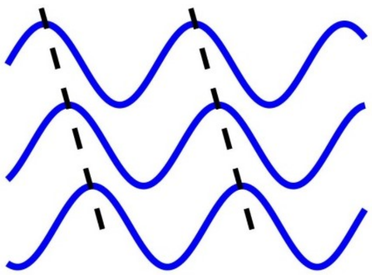
Momentum and energy in SHG

- $\Delta \mathbf{k}$=$\mathbf{k_2}$-2$\mathbf{k_1}$=$\mathbf{0}$ is the **phase matching** condition

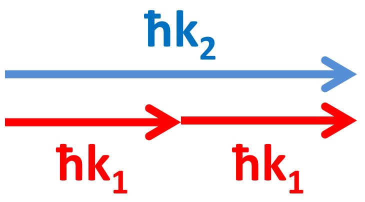
SHG waves in phase matching



SHG waves out of phase matching



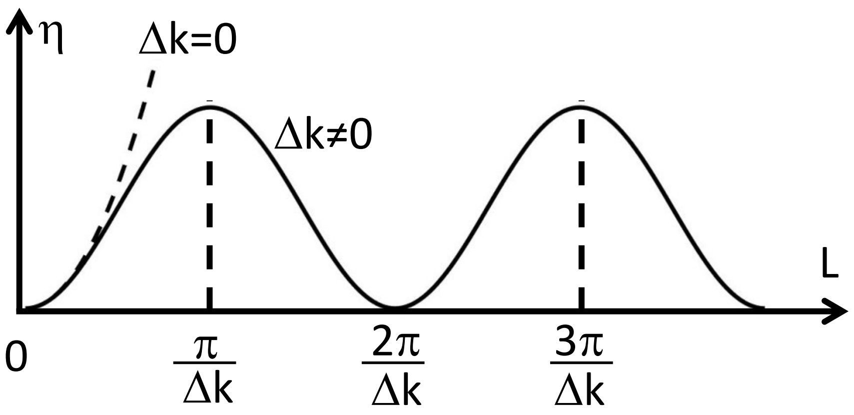
- momentum conservation between photons (not necessary but improves efficiency).



Momentum and energy in SHG

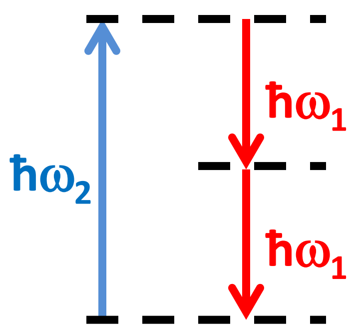
-> exercise: notes page 30. Required laser intensity for SHG

- coherence length in SHG:



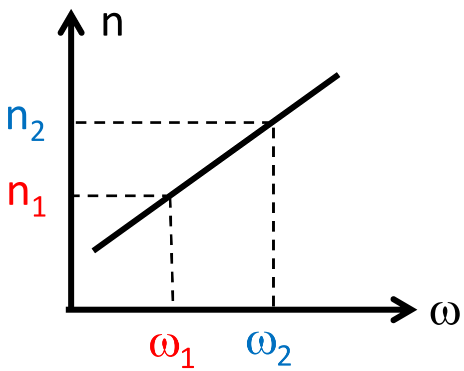
$$L_c = \frac{\pi}{\Delta k}$$

- photon energy is always conserved:

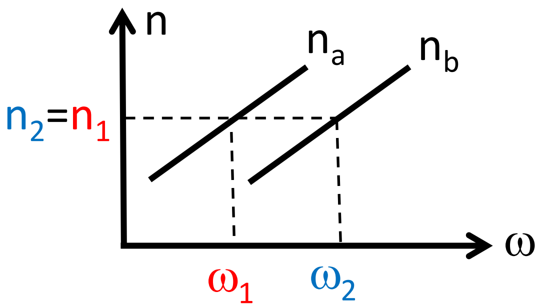


Phase matching in SHG

- $\Delta k = k_2 - 2k_1 = (\omega_2 n_2 - 2\omega_1 n_1)/c = 0$, $\omega_2 = 2\omega_1$ => $n_2 = n_1$!!!
- not possible in standard materials with normal dispersion

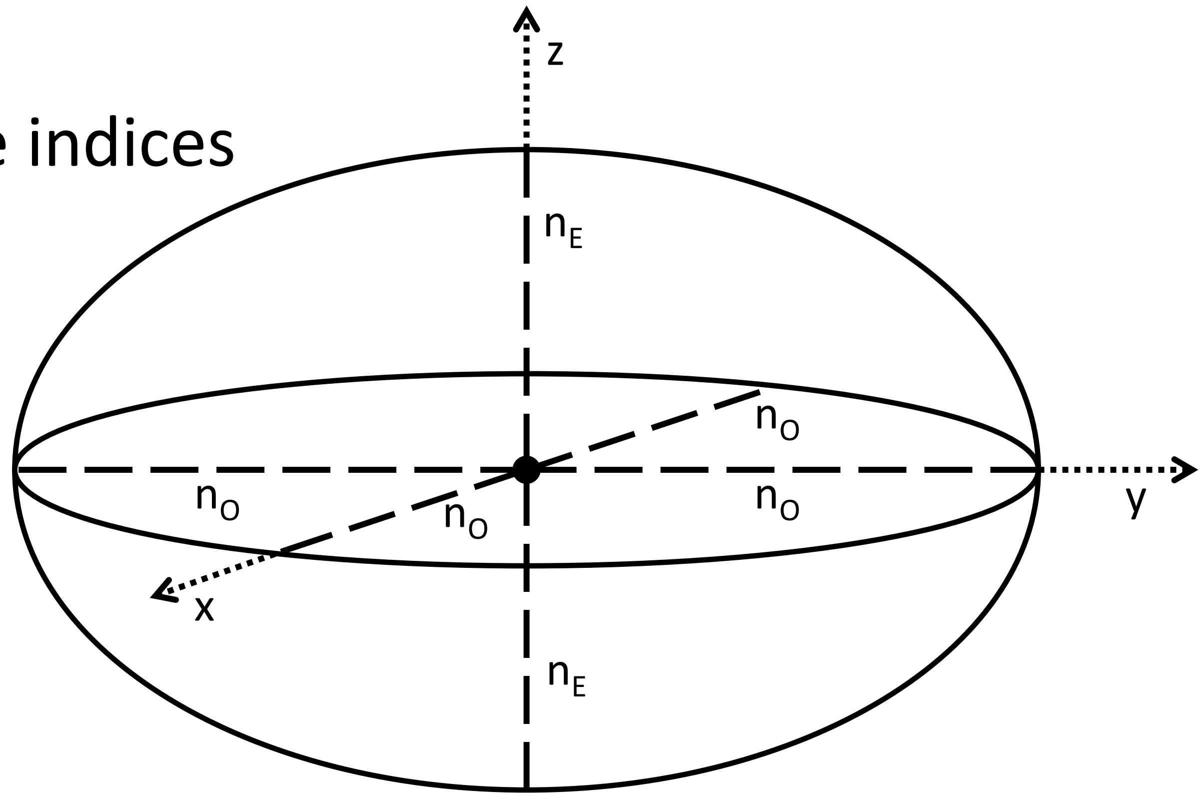


- one method exploits birefringent materials



Birefringent phase matching

- refractive indices depend on E-field direction (polarization)
- ellipsoid of the indices
- BBO ($n_E < n_O$):



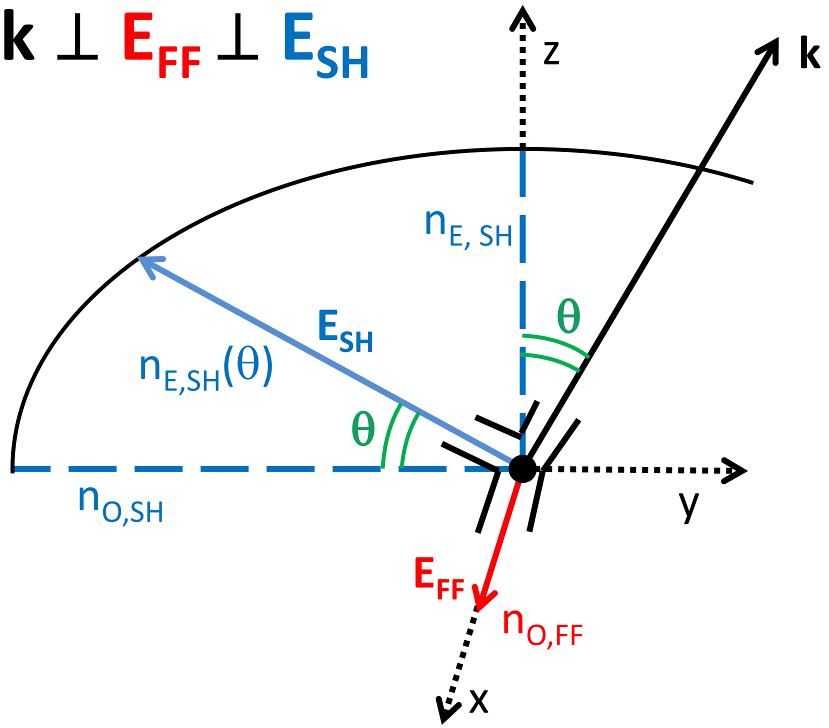
Birefringent phase matching

z: optical axis of BBO

k: propagation direction

$\mathbf{k} \perp \mathbf{E_{FF}} \perp \mathbf{E_{SH}}$

$n_{E,SH} < n_{E,SH}(\theta) < n_{O,SH}$

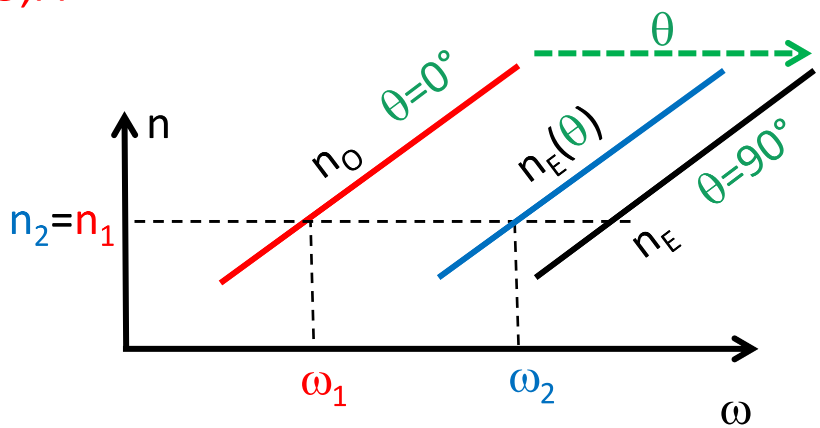


$$\frac{\cos^2(\theta)}{n_{O,SH}{}^2} + \frac{\sin^2(\theta)}{n_{E,SH}{}^2} = \frac{1}{n_{E,SH}{}^2(\theta)}$$

FF: fundamental frequency
SH: second harmonic

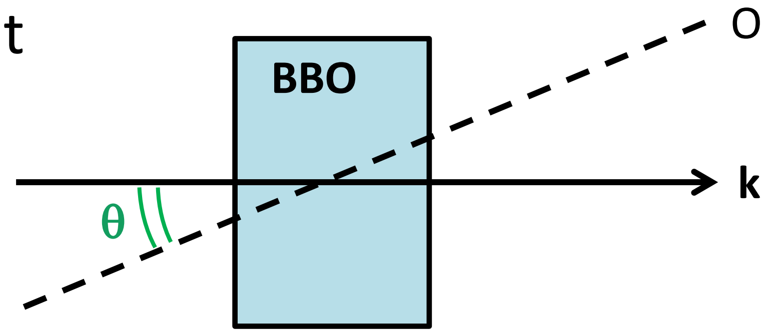
Birefringent phase matching (BPM)

if $n_{E,SH}(\theta) = n_{O,FF}$ then θ is the phase matching angle



crystals can be cut at

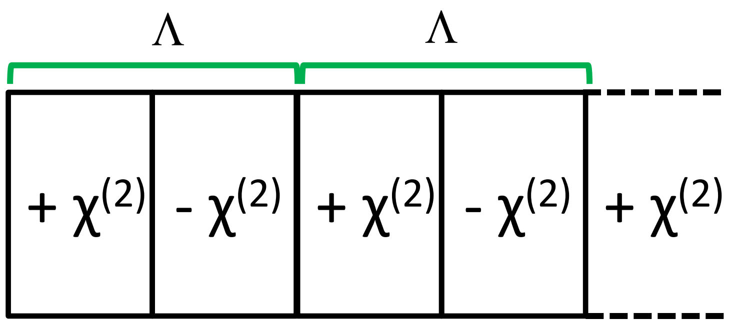
the desired angle θ



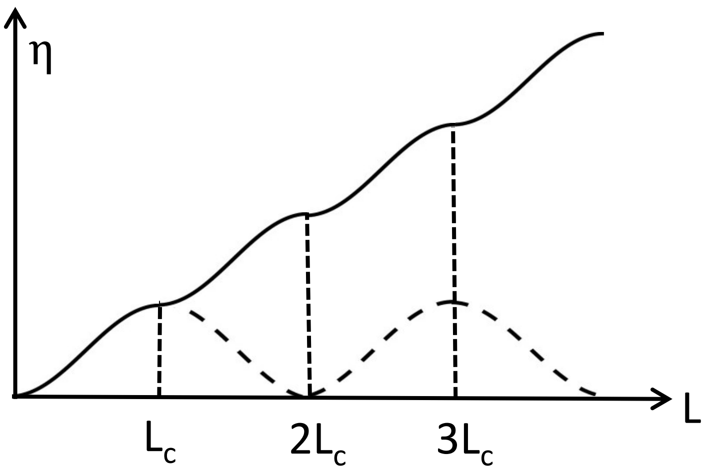
-> exercise: notes page 39. Calculate phase matching angle for SHG in BBO

Quasi phase matching (QPM)

- periodic modulation of $\chi^{(2)}$ in the crystal



- efficiency grows after coherence length $L_c = \frac{\pi}{\Delta k}$



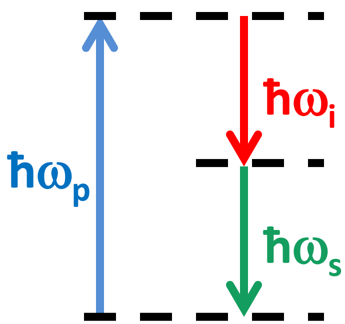
phase matching condition

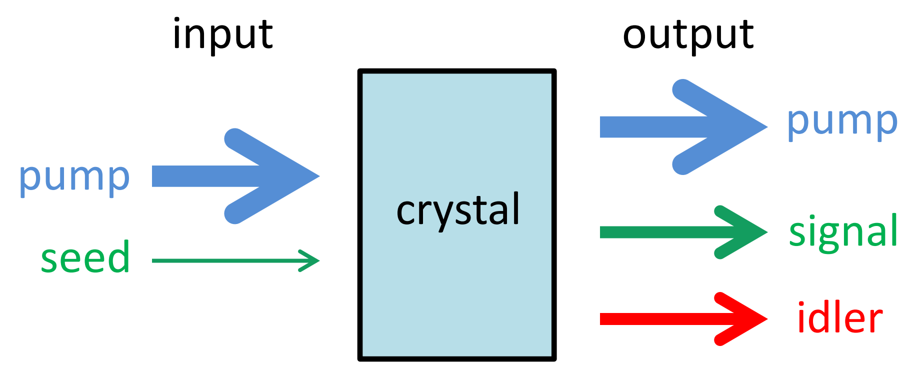
$$k_2 - 2k_1 - \frac{\pi}{\Lambda} = 0$$

Optical parametric amplifiers (OPA)

- 3 photons: pump, signal, idler. Pump amplifies signal and generates idler (DFG)

-> question: how many photons interact in SHG?





- $\hbar\omega_p = \hbar\omega_s + \hbar\omega_i$

- phase matching: $\Delta k = k_p - k_s - k_i = 0$

see notes page 44-48

Coupled equations for OPAs

- follow same steps as for SHG
- SVEA approximation

$$\begin{cases} \dfrac{dE_i}{dz} = -i\dfrac{\omega_i d}{n_i c} E_s^{\,*} E_p \exp(-i\Delta kz) & \boxed{1} \\[2ex] \dfrac{dE_s}{dz} = -i\dfrac{\omega_s d}{n_s c} E_i^{\,*} E_p \exp(-i\Delta kz) & \boxed{2} \\[2ex] \dfrac{dE_p}{dz} = -i\dfrac{\omega_p d}{n_p c} E_i E_s \exp(i\Delta kz) & \boxed{3} \end{cases}$$

G. Cerullo *et al.*, Rev. Sci. Instr. **74**,1 (2003)

see notes page 44-48

Signal and idler in OPAs

- perfect phase matching ($\Delta k=0$)
- no pump depletion (E_p=constant)

$$\begin{cases} I_s(L) = \frac{1}{4} I_{s0} \exp(2\Gamma L) \\ I_i(L) = \frac{1}{4} \frac{\omega_i}{\omega_s} I_{s0} \exp(2\Gamma L) \end{cases}$$

$$\Gamma^2 = \frac{2\omega_i \omega_s d^2 I_p}{n_i n_s n_p \varepsilon_0 c^3}$$

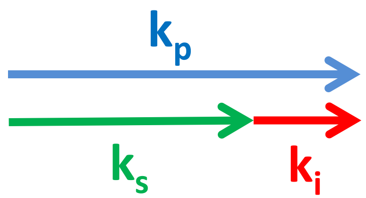
photons idler = # photons signal

$$\frac{I_i(L)}{\omega_i} = \frac{I_s(L)}{\omega_s}$$

gain: $G = \frac{I_s(L)}{I_{s0}} = \frac{1}{4}\exp(2\Gamma L)$

Collinear phase matching in OPAs

$$\Delta k = k_p - k_s - k_i = 0$$



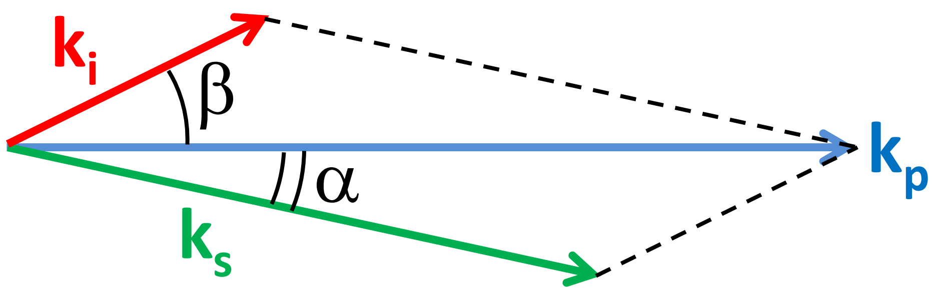
$$\begin{cases} \omega_p n_p - \omega_s n_s - \omega_i n_i = 0 & \text{momentum conservation (phase matching)} \\ \omega_p - \omega_s - \omega_i = 0 & \text{energy conservation} \end{cases}$$

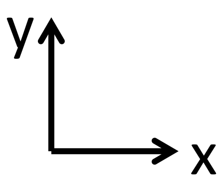
see notes page 44-48

Noncollinear phase matching in OPAs

- angle between pump and signal
- vectorial relationship

$$\Delta \mathbf{k}=\mathbf{k}_p-\mathbf{k}_s-\mathbf{k}_i=0$$





$$\begin{cases} k_p-k_s\cos\alpha-k_i\cos\beta=0 & \text{phase matching x-direction} \\ K_s\sin\alpha-k_i\sin\beta=0 & \text{phase matching y-direction} \\ \omega_p-\omega_s-\omega_i=0 & \text{energy conservation} \end{cases}$$

see notes page 48-52

Ultrashort pulses with OPAs

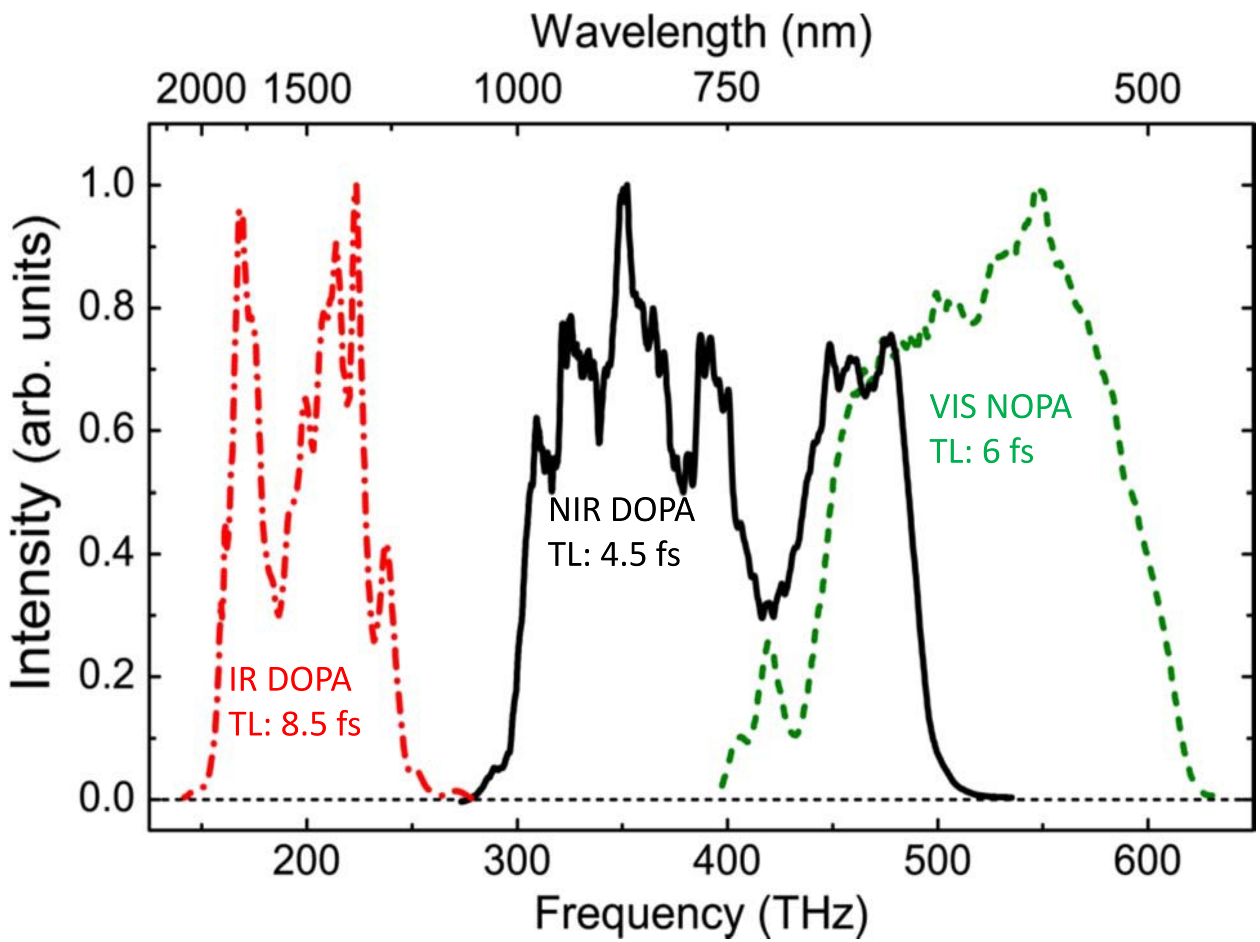
- OPAs can produce few-cycle pulses in 2 cases
  - degenerate (collinear) OPA (DOPA): $\omega_s = \omega_i = \omega_p/2$
    - ✓ λ_p = 400 nm, $\lambda_s = \lambda_i$ = 600-1000 nm in BBO

      -> Question: what is the PM angle?
    - ✓ λ_p = 800 nm, $\lambda_s = \lambda_i$ = 1200-2000 nm in BBO
  - noncollinear OPA (NOPA)
    - ✓ λ_p = 400 nm, λ_s = 500-700 nm, $\alpha = 3.7^o$ in BBO (visible NOPA)

- effects of crystal length:
  - ✓ increase gain
  - ✓ decrease bandwidth

see notes page 48-52

Ultrashort pulses with OPAs

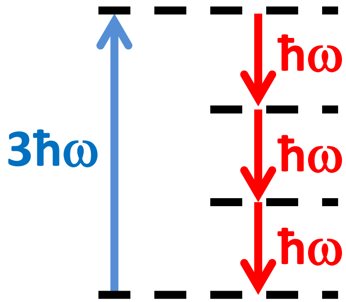


G. Cirmi *et al.*, JOSA B **25**, B62 (2008)

see notes page 53-55

3rd order nonlinear optics

- can occur in centrosymmetric media
  - third harmonic generation (THG)



  - intensity dependent refractive index

$$n=n_0+n_2I$$

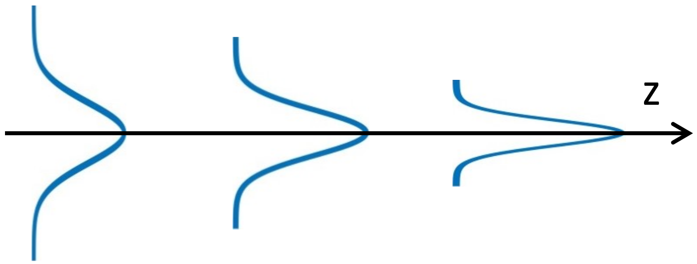
    - self focusing (SF)
    - self phase modulation (SPM)

$\left.\begin{array}{l}\text{self focusing (SF)}\\ \text{self phase modulation (SPM)}\end{array}\right\}$ white light generation (WLG)

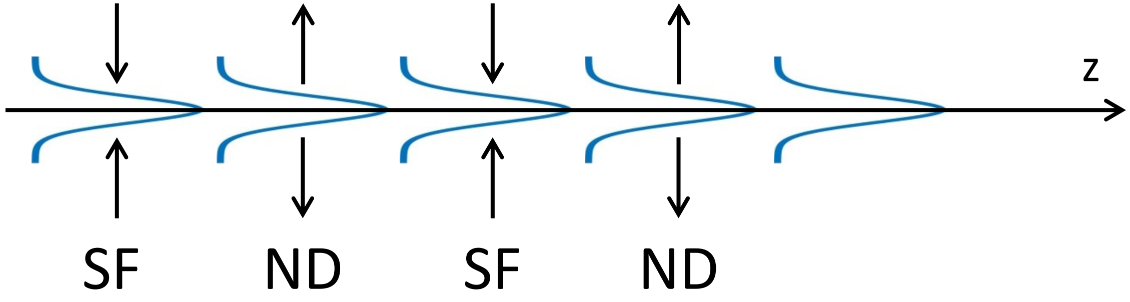
see notes page 56-57

Self focusing (SF)

- higher intensity in the center => higher refractive index => lens effect



- filament formation (SF vs. natural divergence ND)



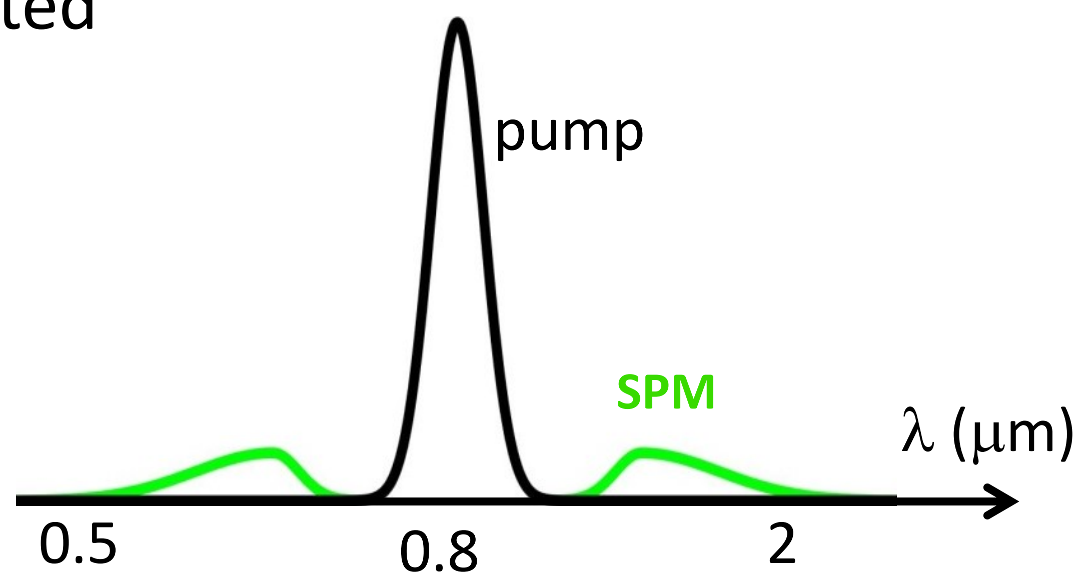
see notes page 58-59

Self phase modulation (SPM)

- n appears in the phase

$$E = E_0 \exp\left[i\left(2\pi\, \mathbf{n}\, \frac{z}{\lambda} - 2\pi\nu t + \varphi\right)\right]$$

- with n=n(I), new frequency components can be generated



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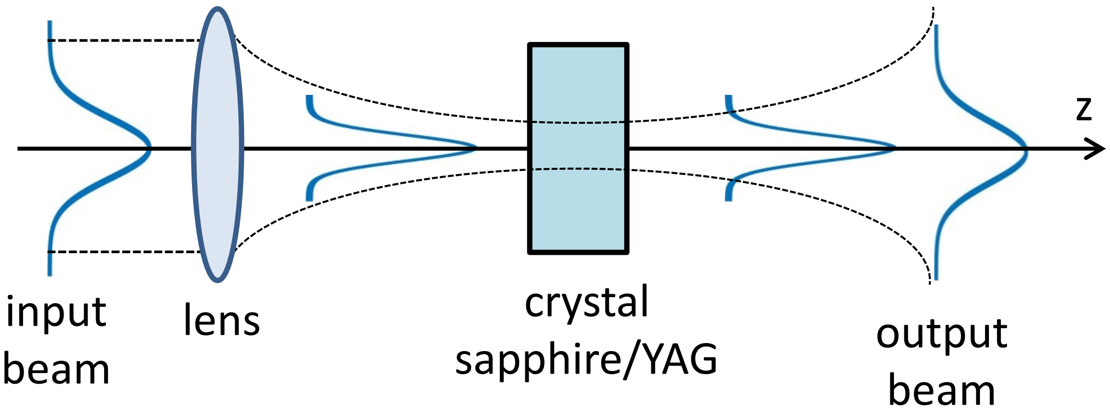
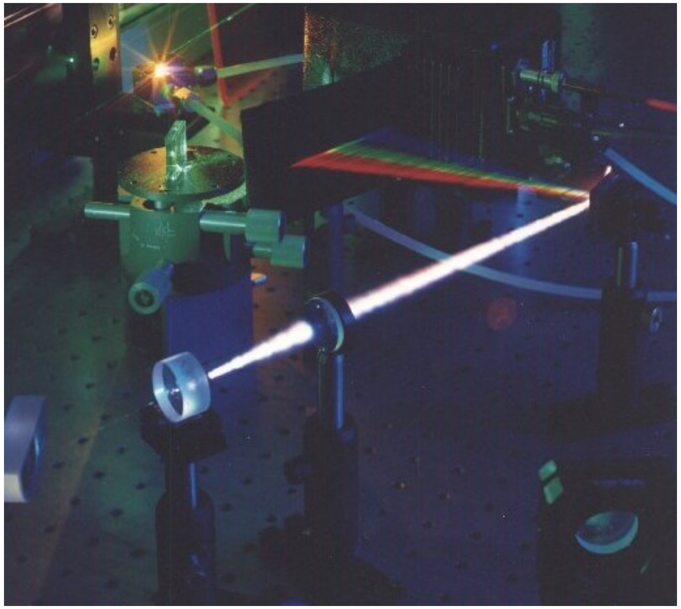
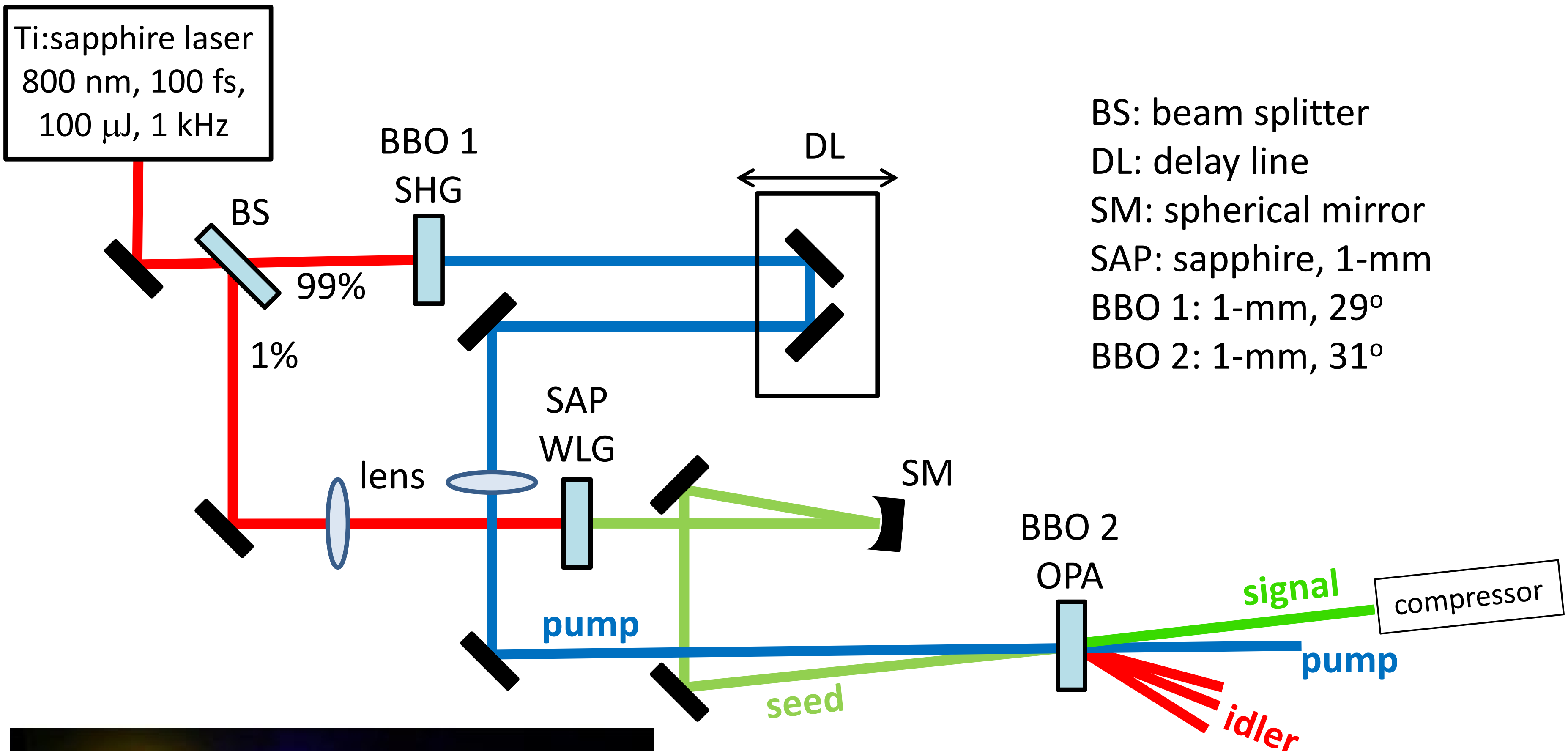
White light generation (WLG)

SPM + SF: WLG





Visible NOPA architecture



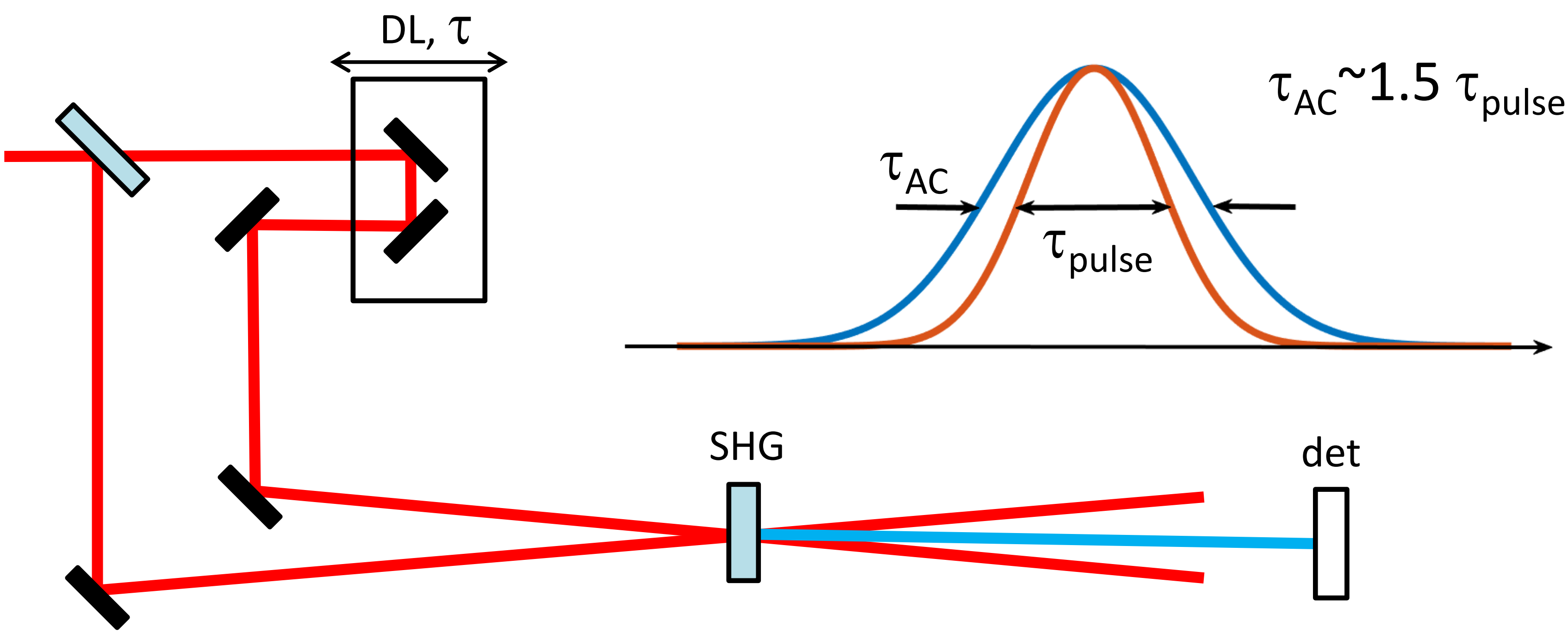
BS: beam splitter
DL: delay line
SM: spherical mirror
SAP: sapphire, 1-mm
BBO 1: 1-mm, 29°
BBO 2: 1-mm, 31°

pump (input): 40 μJ, 400 nm, 70 fs
signal: 5 μJ, 500-700 nm, 6 fs (after compressor)

see notes page 61-63

Pulse measurement

- how to measure the shortest possible events?
- with the same event -> autocorrelation



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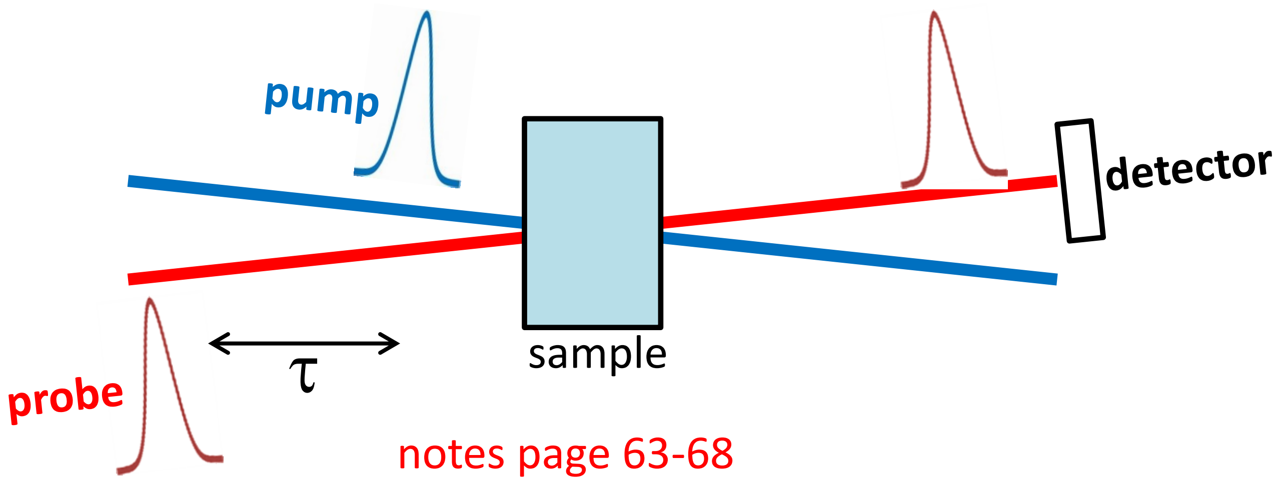
Contents

- Introduction and motivation
- Maxwell's equations
- Nonlinear optics of 2nd and 3rd order
- Pump-probe technique
- High harmonic generation
- Pulse synthesis

Applications of fs pulses
Pump-probe technique

- follow fs dynamics (physics, chemistry, biology, medicine)
- pump excites sample
- probe tests it
- pump-probe delay τ is controlled
- probe transmission or reflection is measured vs. τ
- dynamics can be followed



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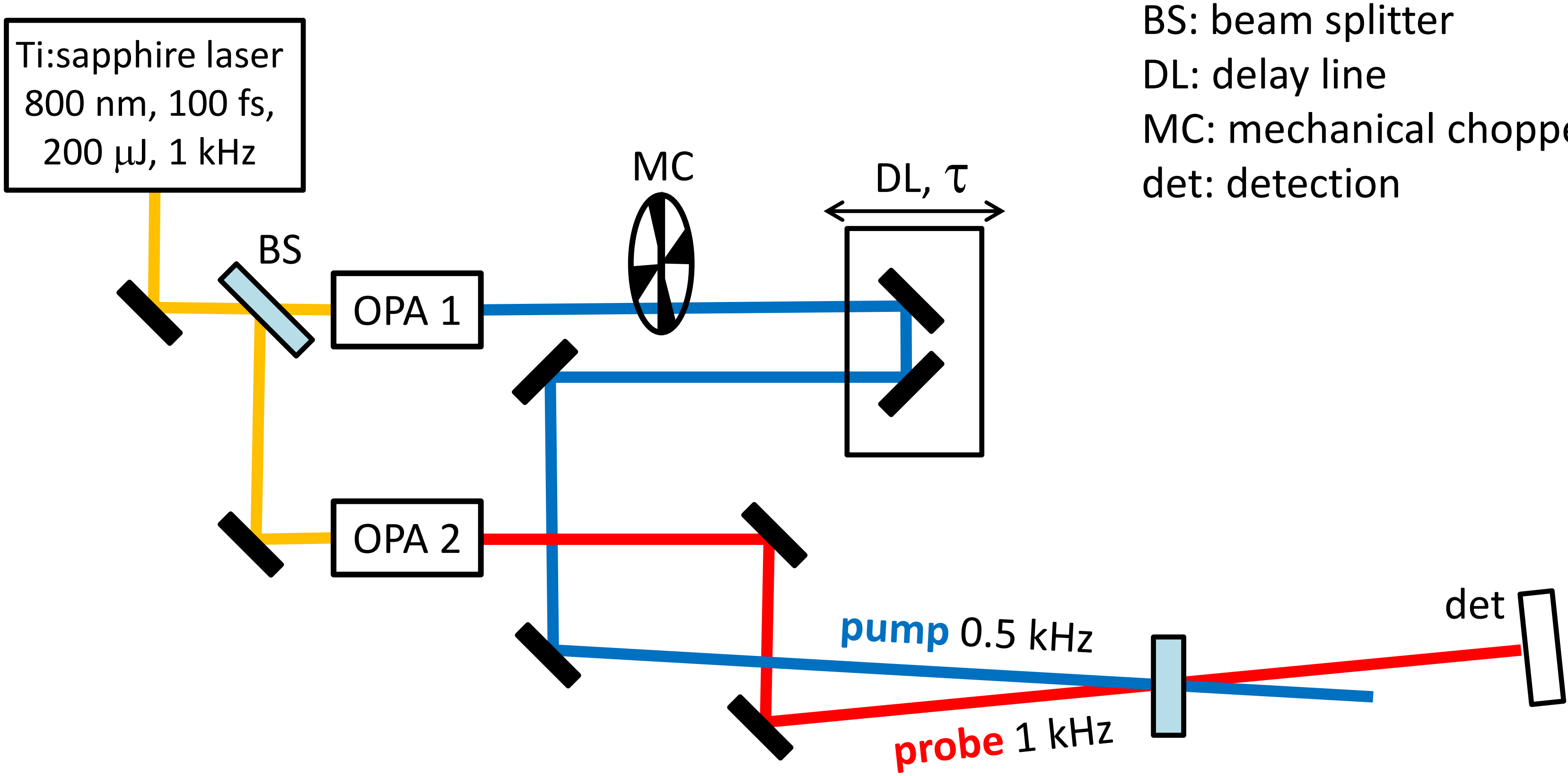
Pump-probe setup



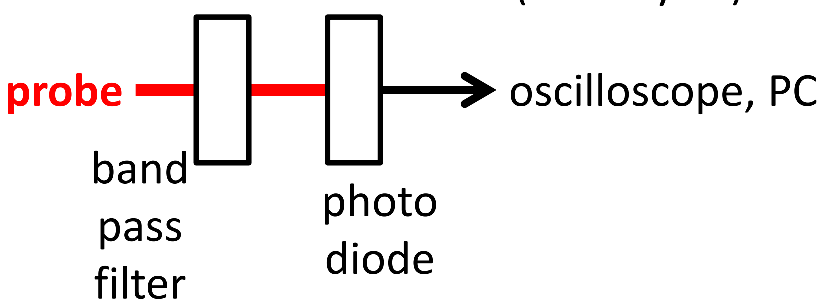
BS: beam splitter
DL: delay line
MC: mechanical chopper
det: detection

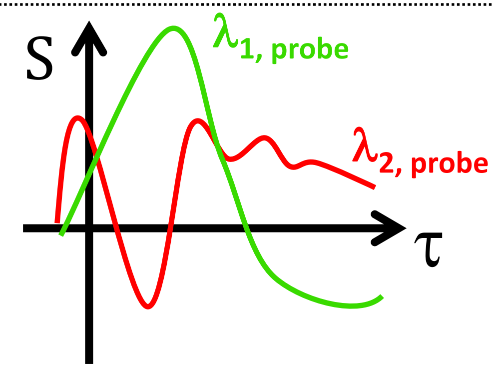
Pump-probe detection

- Differential probe transmission (pump on - pump off)

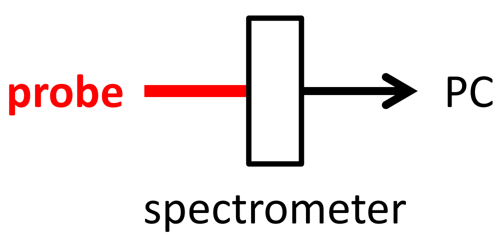
$$S = \frac{\Delta T}{T} = \frac{T_{P\,ON} - T_{P\,OFF}}{T_{P\,OFF}}$$

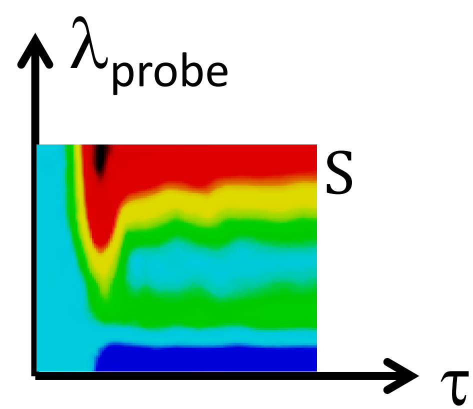
detection scheme 1 ("old style")



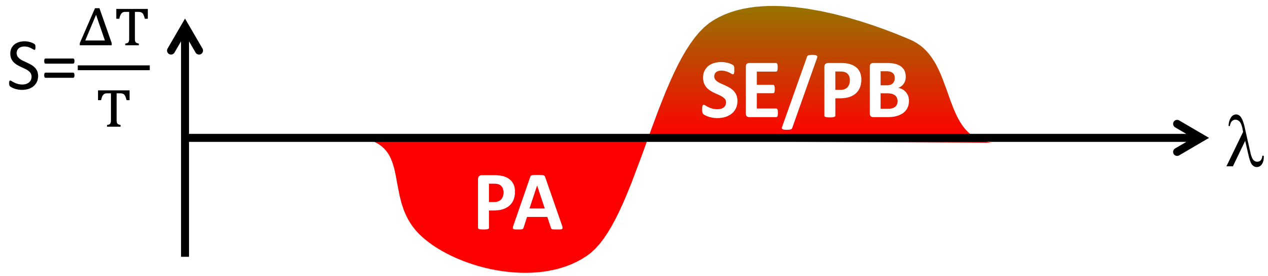


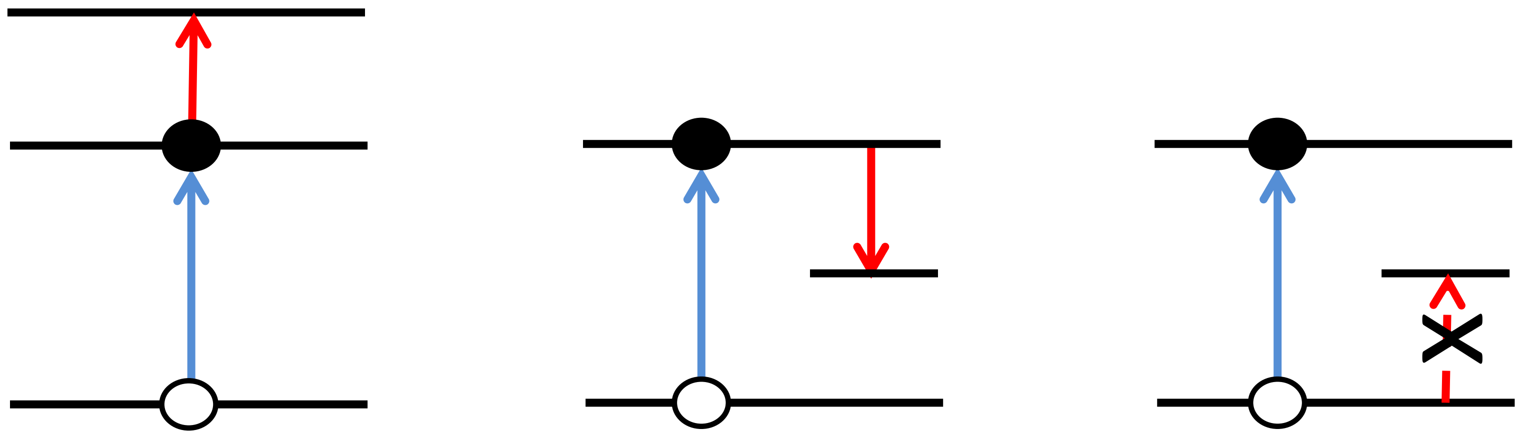
detection scheme 2 ("new style")





Pump-probe signals





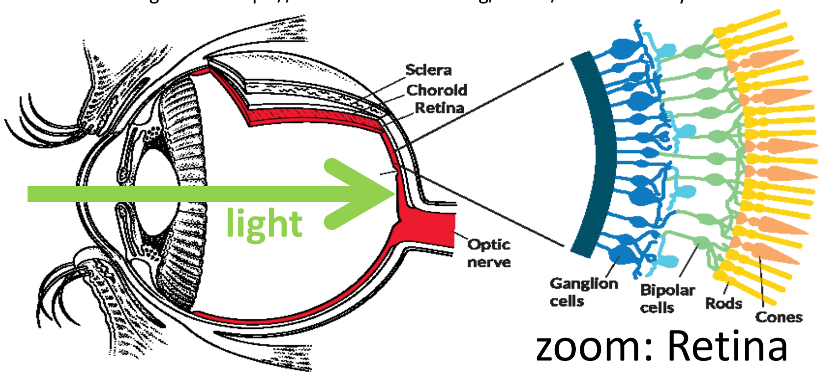
photoinduced absorption (PA) $S<0$

stimulated emission (SE) $S>0$

photo-bleaching (PB) $S>0$

Application of ultrafast fs spectroscopy: human vision

Image from: https://www.sciencenews.org/article/how-rewire-eye



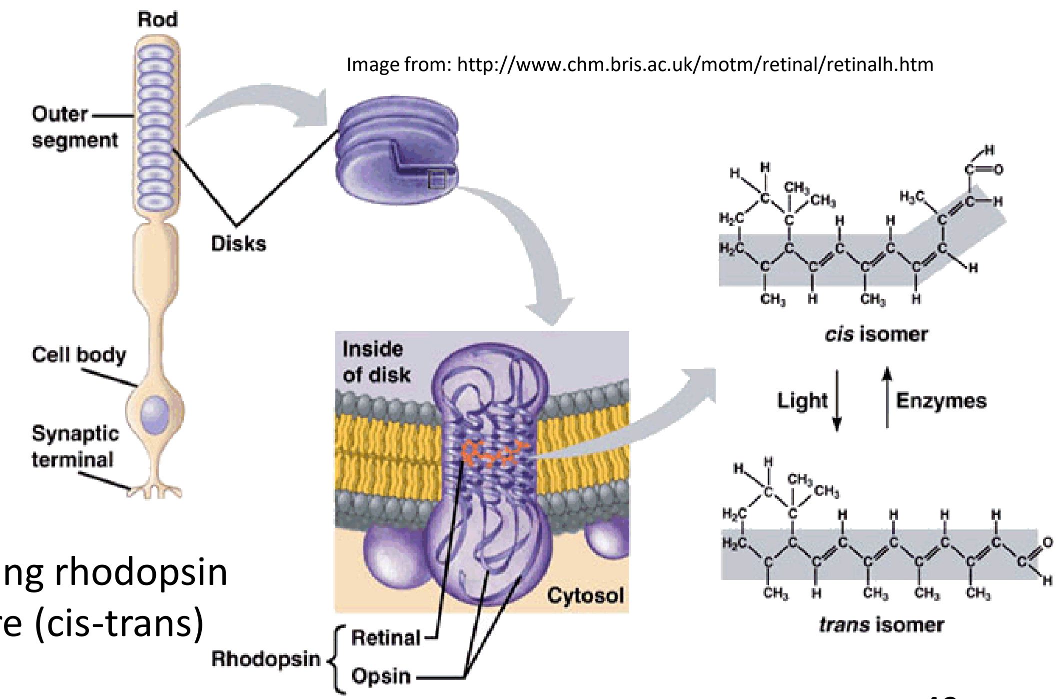
zoom: Retina

Our retina contains photoreceptors called cones and rods

Image from:
Eye clarity

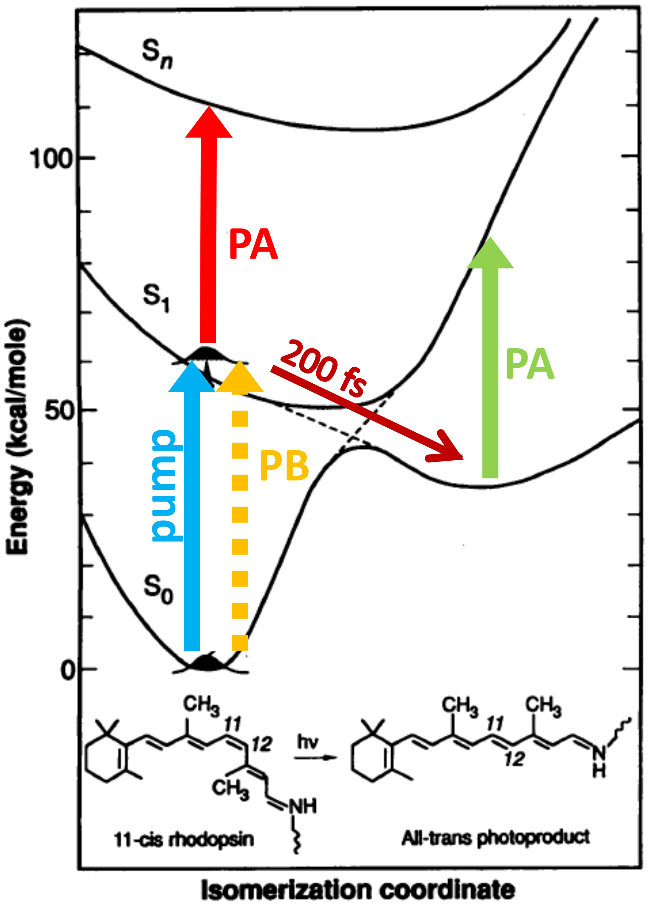
Cones: day vision
Rods: night vision

Image from: http://www.chm.bris.ac.uk/motm/retinal/retinalh.htm



Cones and rods are composed of disks containing rhodopsin
When light arrives, rhodopsin changes structure (cis-trans)
How fast is this process?

The primary step of vision



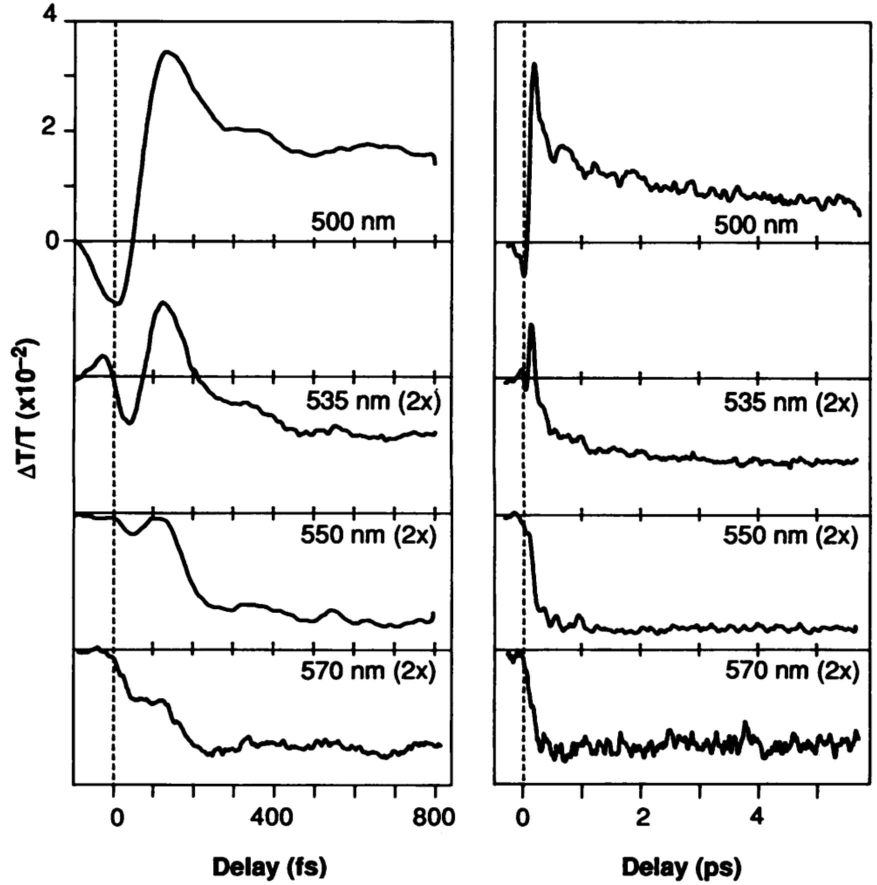
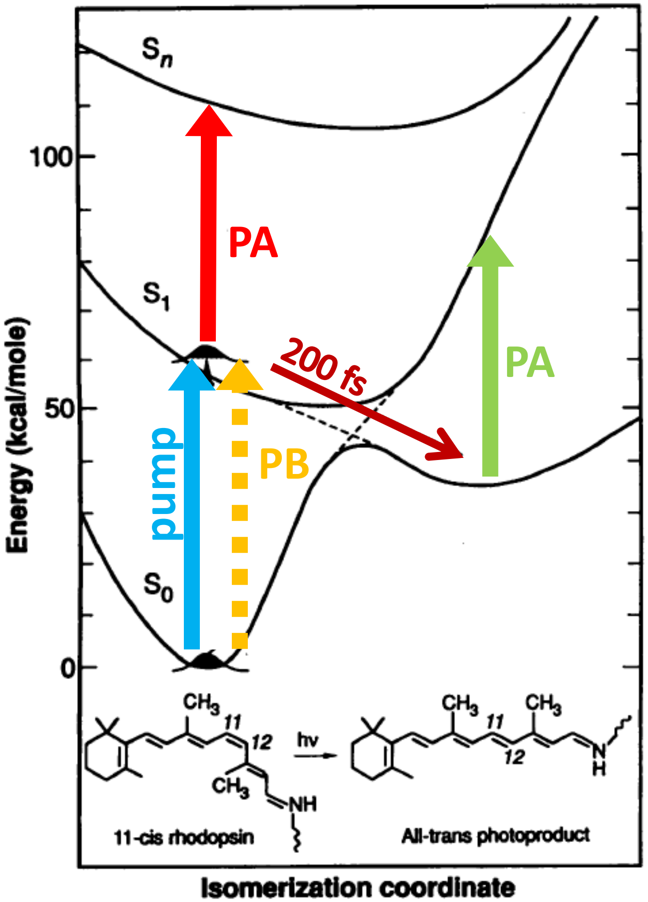


Fig. 2. Transient absorption measurements of 11-*cis* rhodopsin at various wavelengths following a 35-fs pump pulse at 500 nm (~10-fs probe).

Schoenlein R. W. *et al.*, "The first step in vision: femtosecond isomerization of rhodopsin", Science **254**, 412 (1991) DOI: 10.1126/science.1925597

The primary step of vision



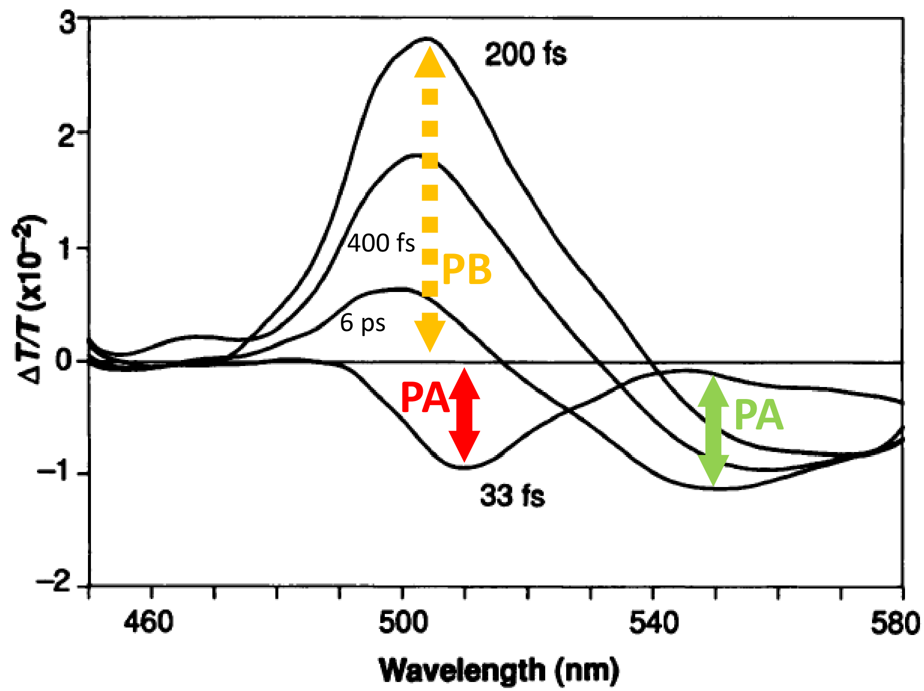


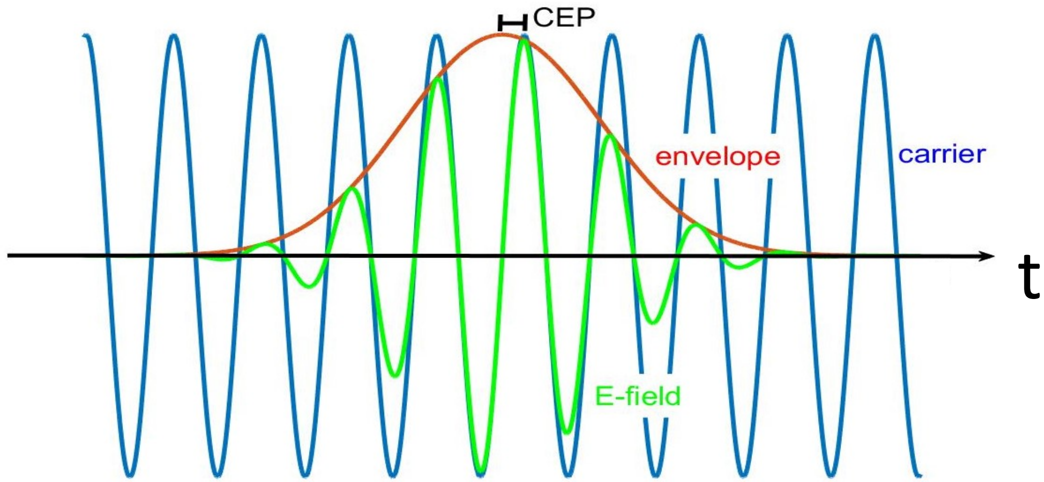
Fig. 3. Difference spectra measurements of 11-*cis* rhodopsin at various delays following a 35-fs pump pulse at 500 nm (~10-fs probe).

The kinetics of the primary event in vision have been resolved with the use of femtosecond optical measurement techniques. The 11-*cis* retinal prosthetic group of rhodopsin is excited with a 35-femtosecond pump pulse at 500 nanometers, and the transient changes in absorption are measured between 450 and 580 nanometers with a 10-femtosecond probe pulse. Within 200 femtoseconds, an increased absorption is observed between 540 and 580 nanometers, indicating the formation of photoproduct on this time scale. These measurements demonstrate that the first step in vision, the 11-*cis*→11-*trans* torsional isomerization of the rhodopsin chromophore, is essentially complete in only 200 femtoseconds.

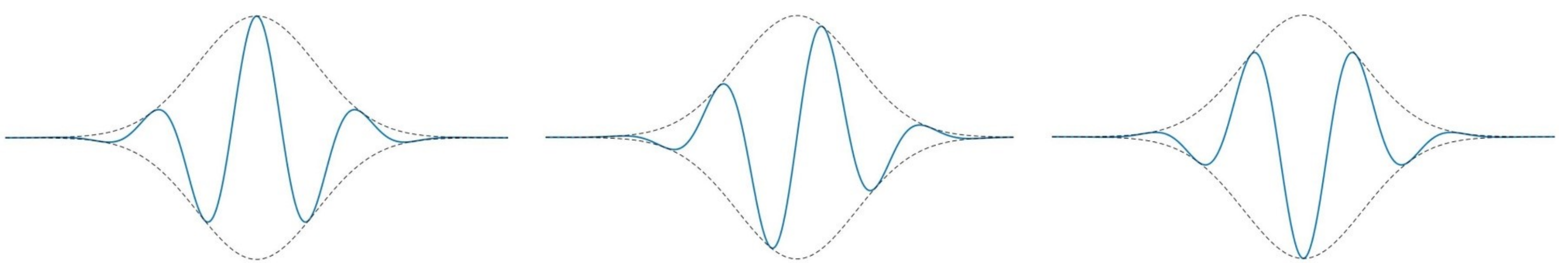
Schoenlein R. W. *et al.*, Science **254**, 412 (1991)

Carrier envelope phase (CEP)

- important for few-cycle pulses, for phenomena like HHG
- can be controlled in different ways



CEP=0 CEP=$\pi/2$ CEP=π



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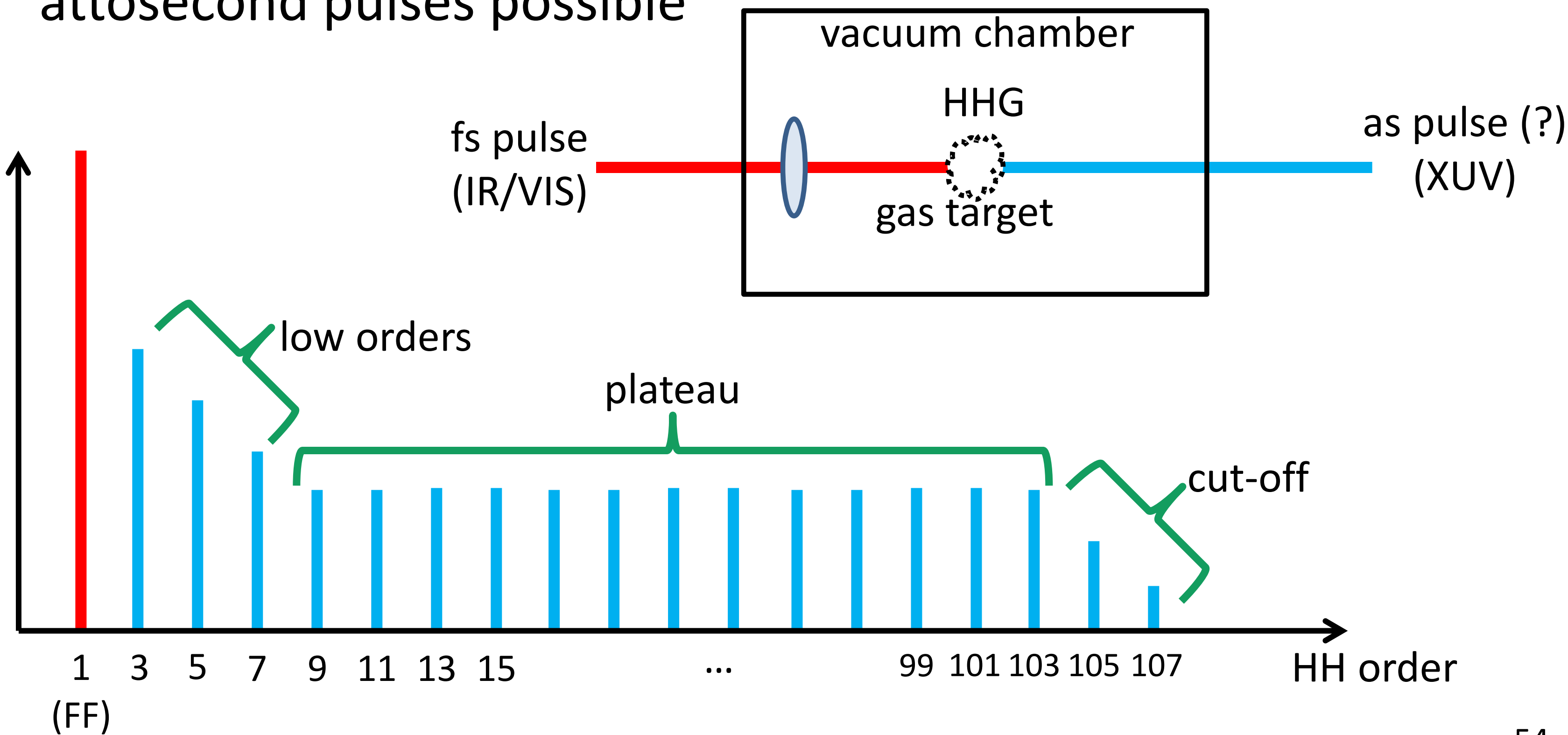
High harmonic generation (HHG)

- HHG can occur when focusing a fs pulse in gas (~10^{14} W/cm^2)
- radiation goes to UV and X-rays (up to keV)
- needs vacuum
- attosecond pulses possible



High harmonic generation

- cut-off energy: $E_{CO} = I_p + 3.17\ U_p$

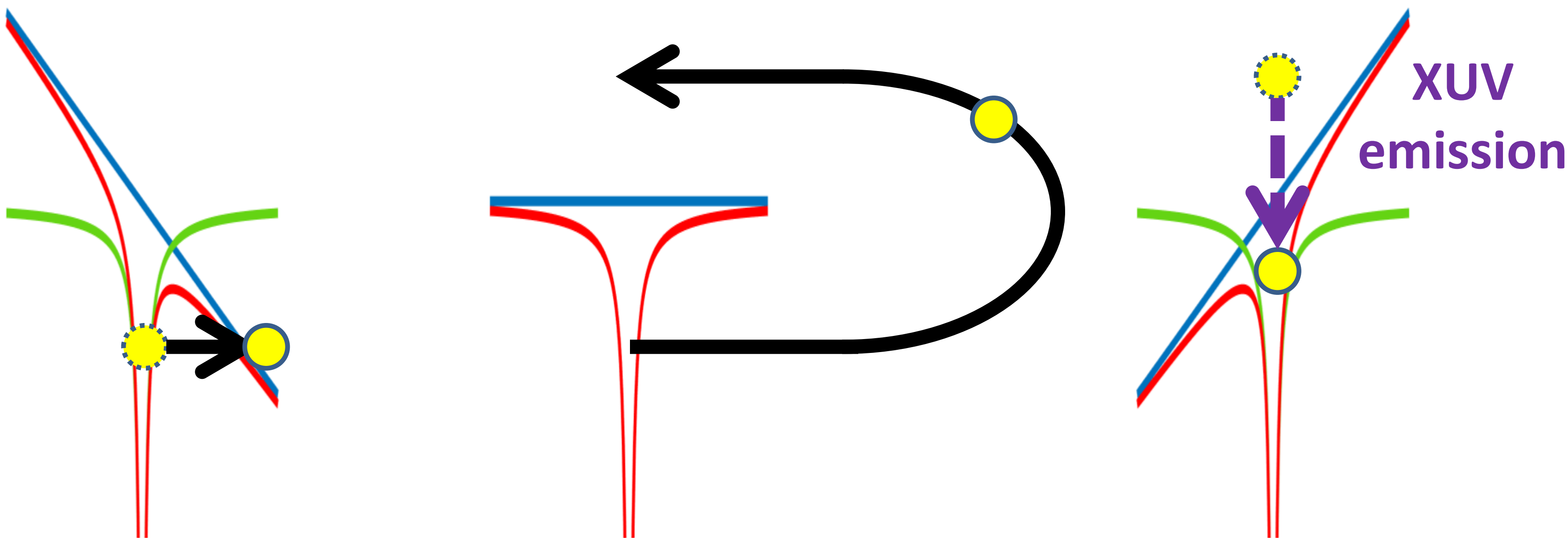
- ponderomotive energy: $U_p \propto \frac{I_D}{\omega_D{}^2}$

- scaling laws:
  - cut-off $\propto \omega_D{}^{-2}$
  - efficiency $\propto \omega_D{}^5$

3 step model

- semiclassical model of HHG
- **E-field** modifies **atomic potential** of electron

1. ionization (tunnel) **2. acceleration** **3. recombination**



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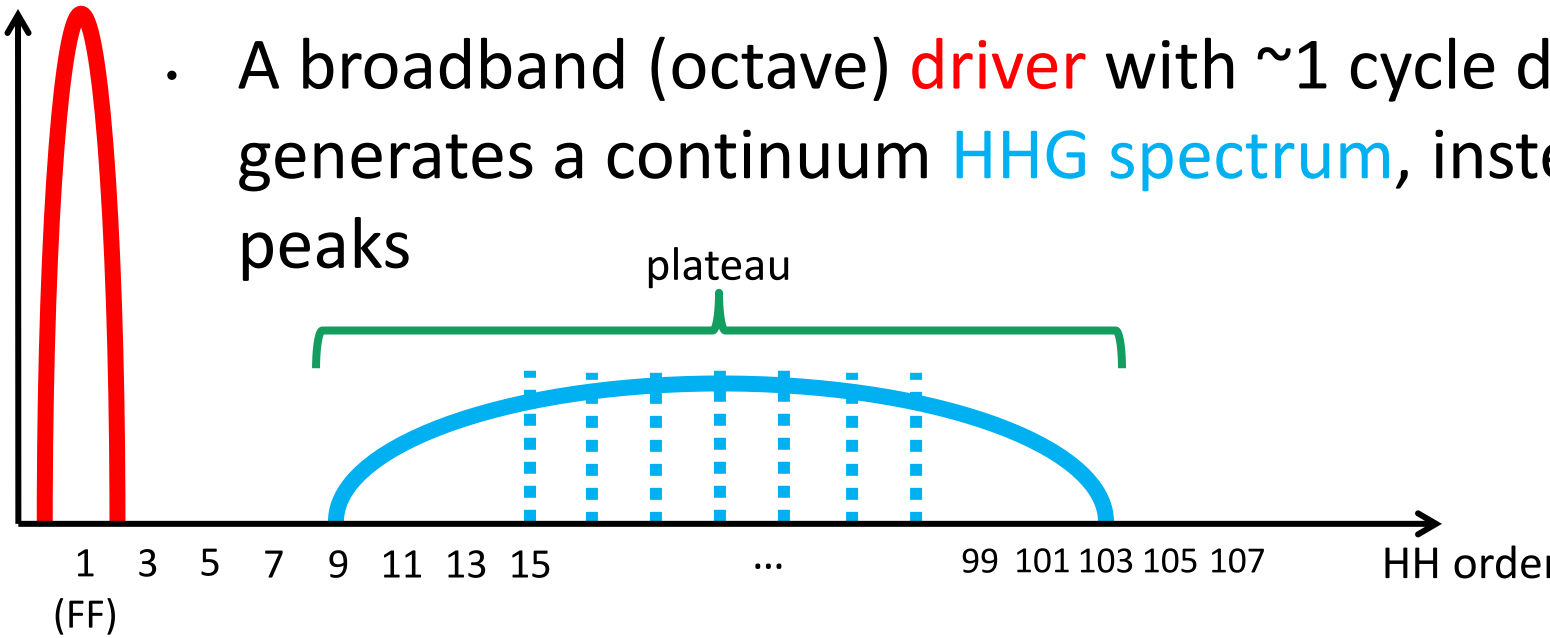
Attosecond pulses

- A broadband (octave) driver with ~1 cycle duration generates a continuum HHG spectrum, instead of peaks



- In time, this can give attosecond pulses

- attosecond science

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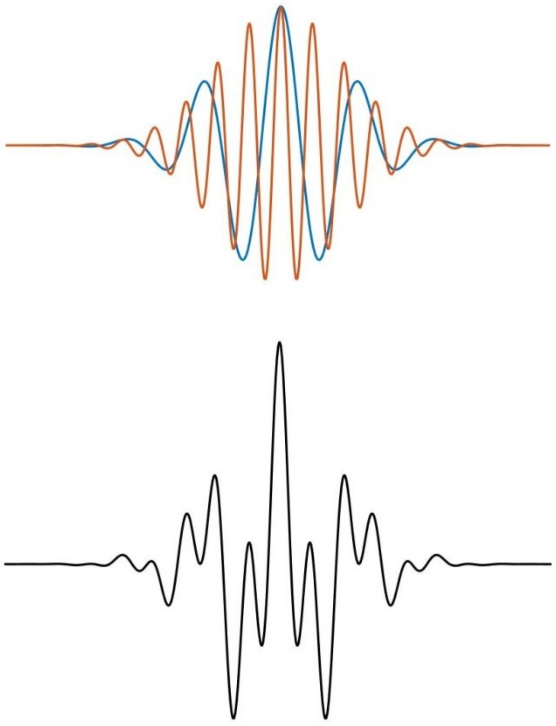
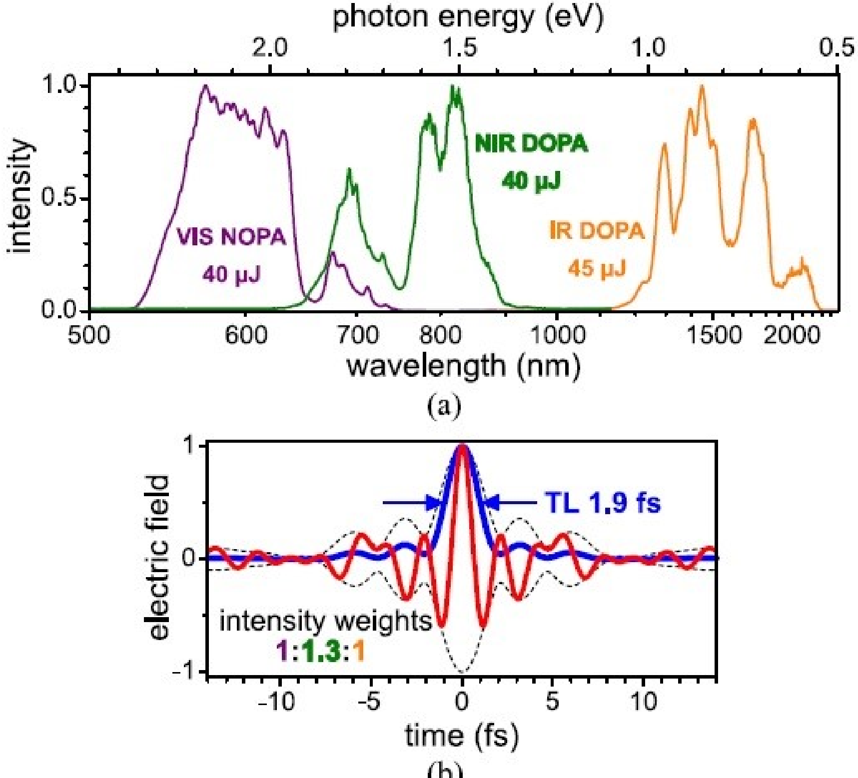
Pulse synthesis

- new frontier of NLO
- synthesize more pulses together





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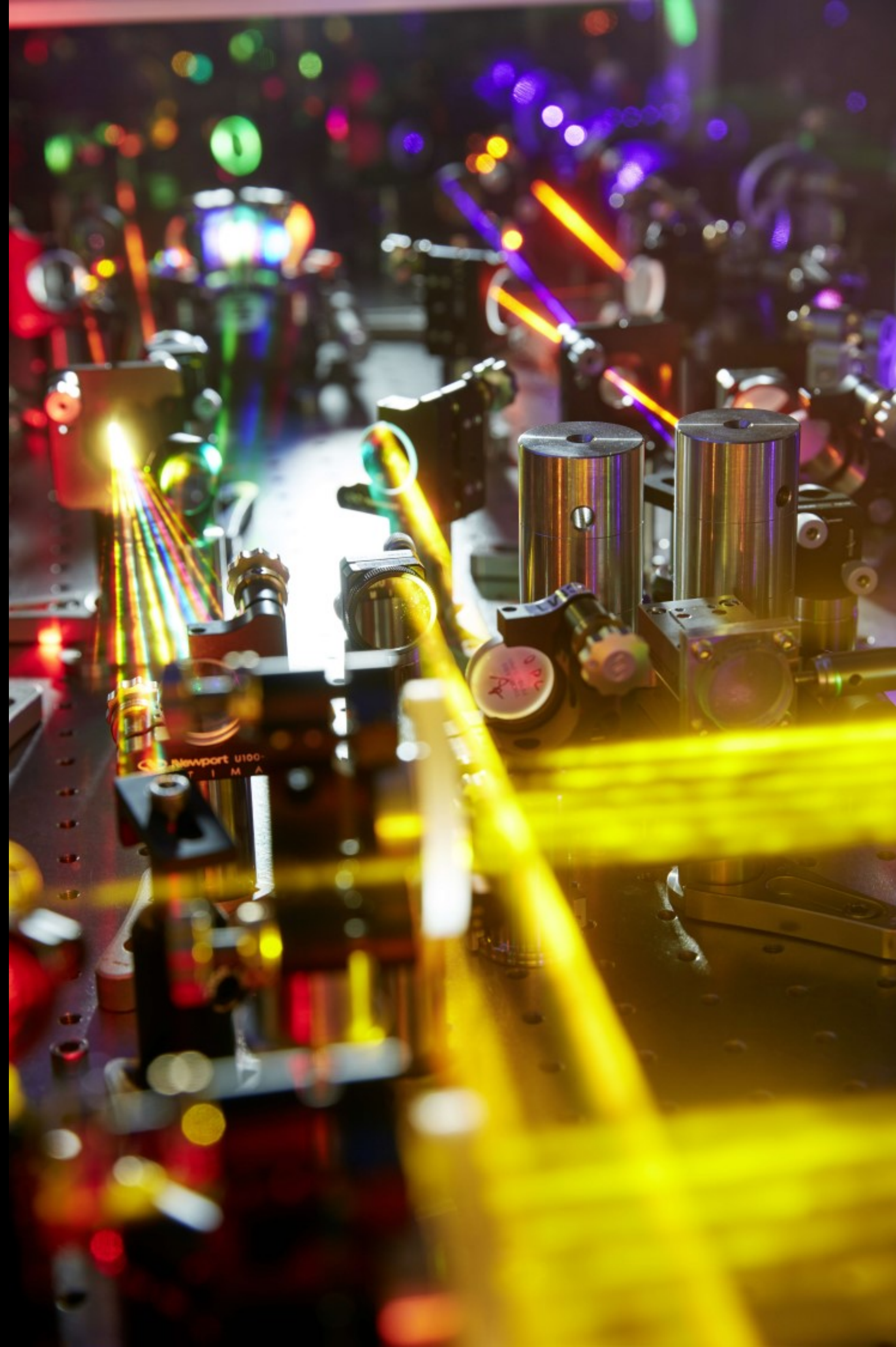
Mücke O. D. *et al.*, JSTQE **21**, 8700712 (201´5)

Pulse synthesis

- Long driver λ -> high HHG cut-off energy
- Short driver λ -> high HHG efficiency
- Efficient HHG at high energies?

- Isolated attosecond pulses
- Pump-probe fs-fs or as-fs or as-as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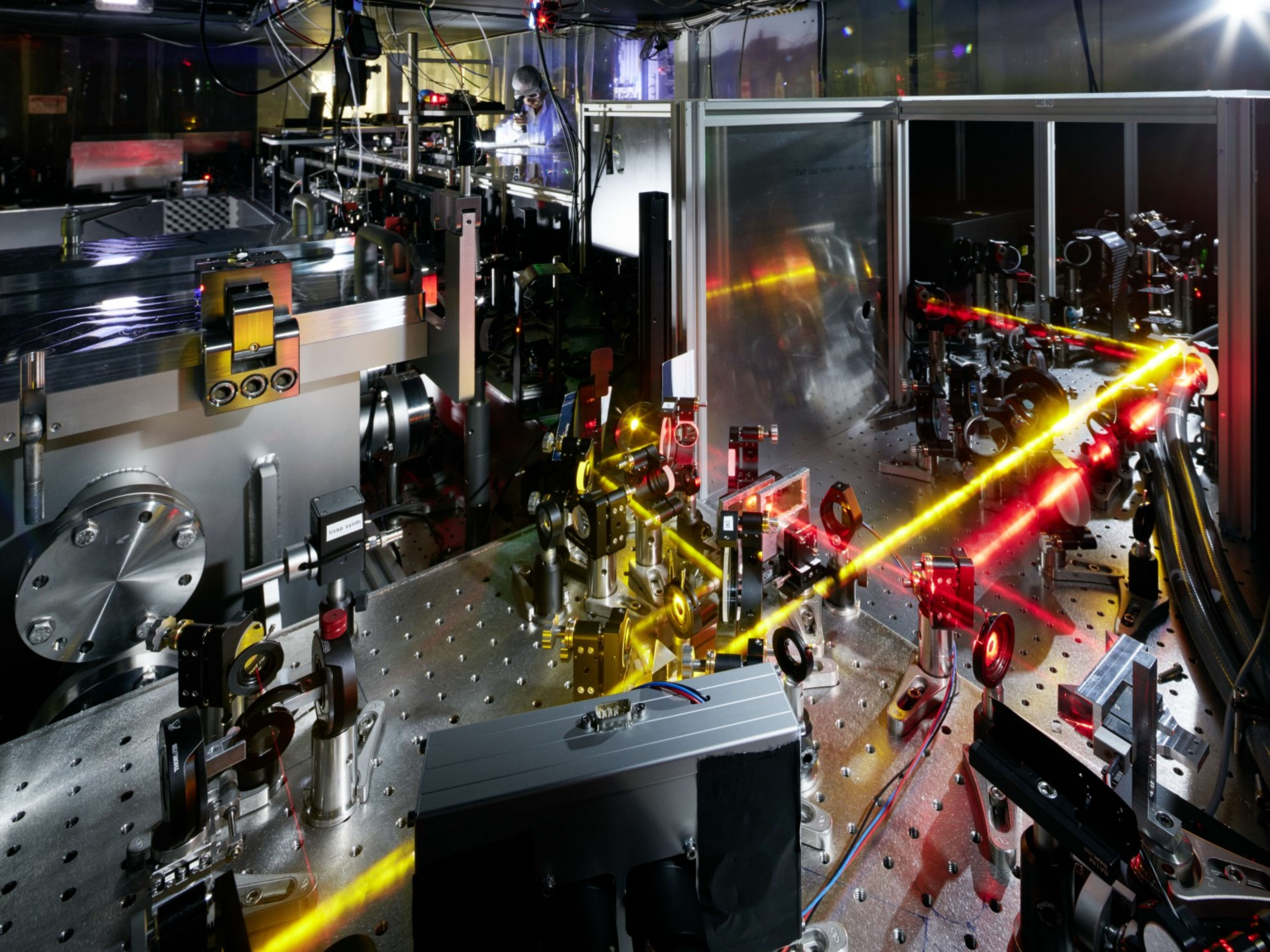
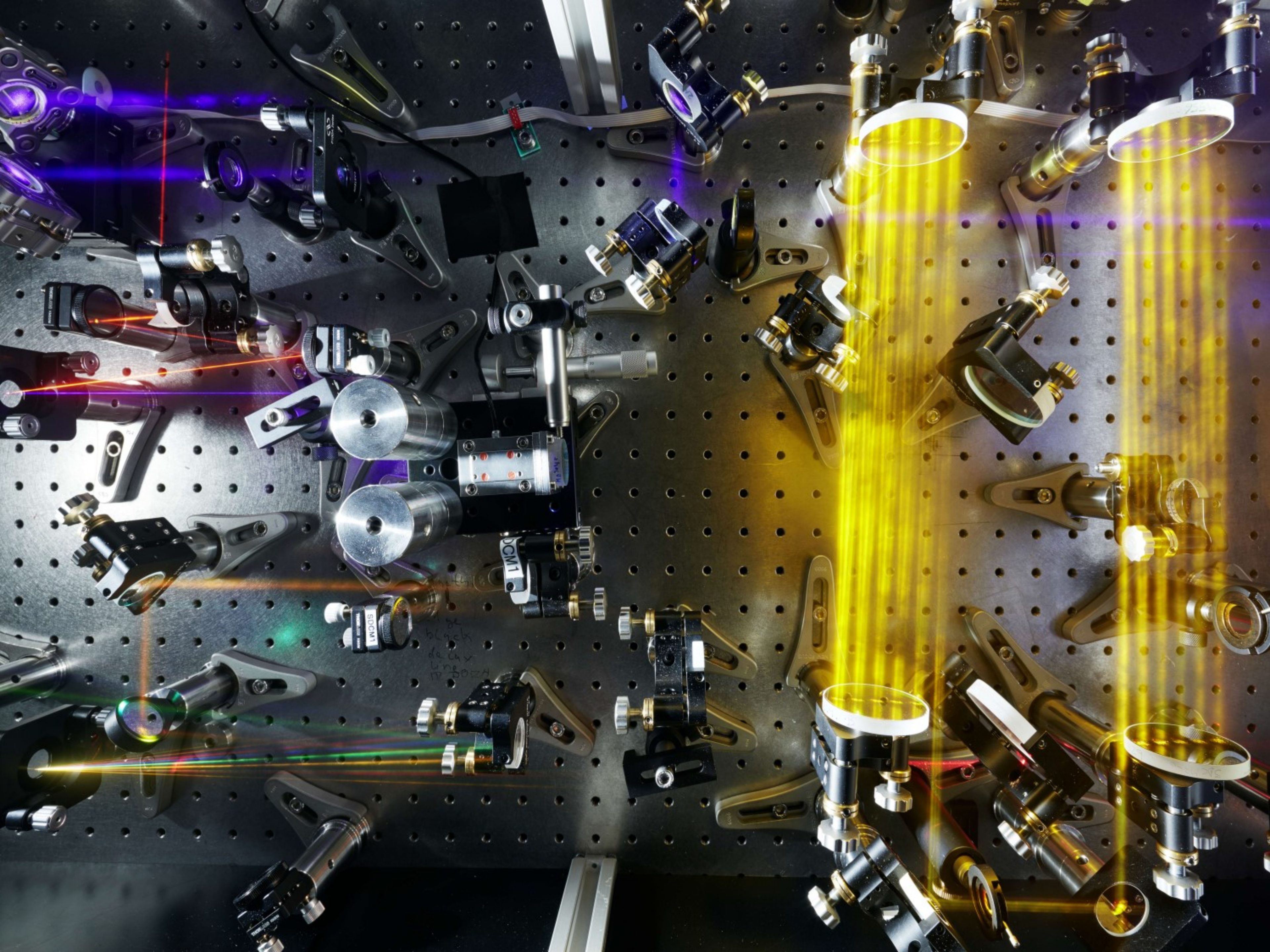
T I M A


tisap synth


DCM1
SDCM1

THANKS!