Attosecond Radiation Sources - PHSY761

Lecture 06

Summary:

Amplifiers of ultrashort Pulses
1. Short pulse propagation

Nonlinear optics
1. Perturbative nonlinear optics
2. Parametric processes, SFG, DFG

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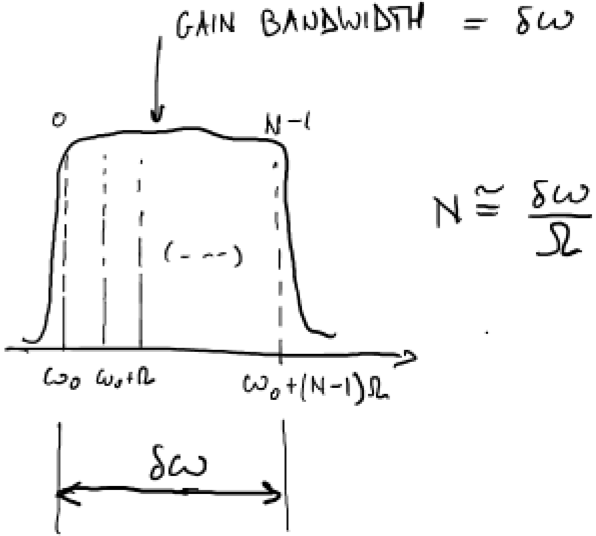
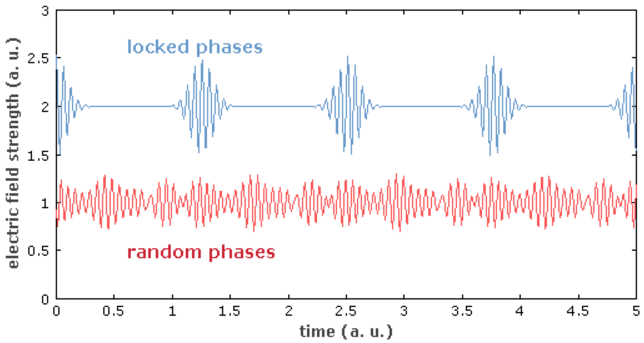
28 October 2025

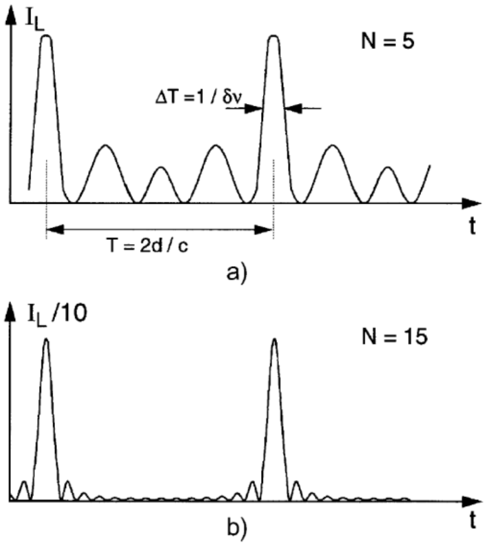
Last week: laser amplifiers, missing ultrashort laser amplifiers

Reminder: Mode-locked lasers



$$N \cong \frac{\delta\omega}{\Omega}$$





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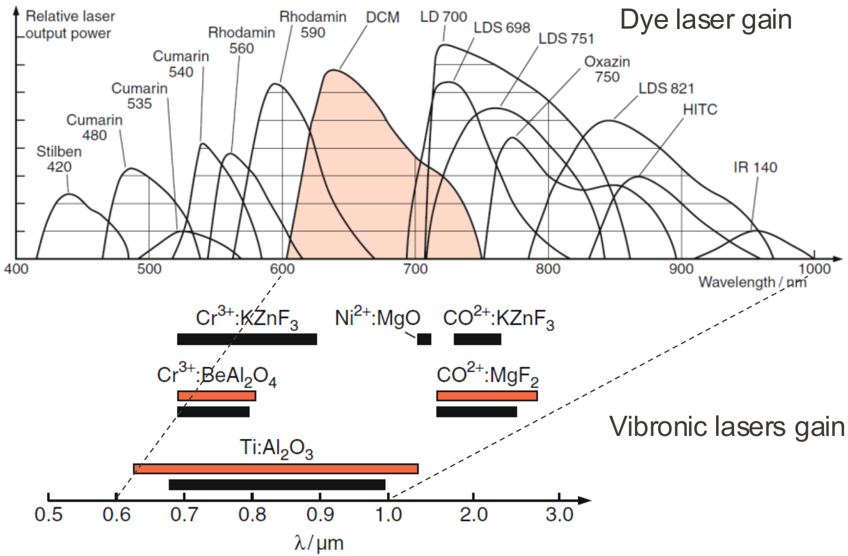
EPFL

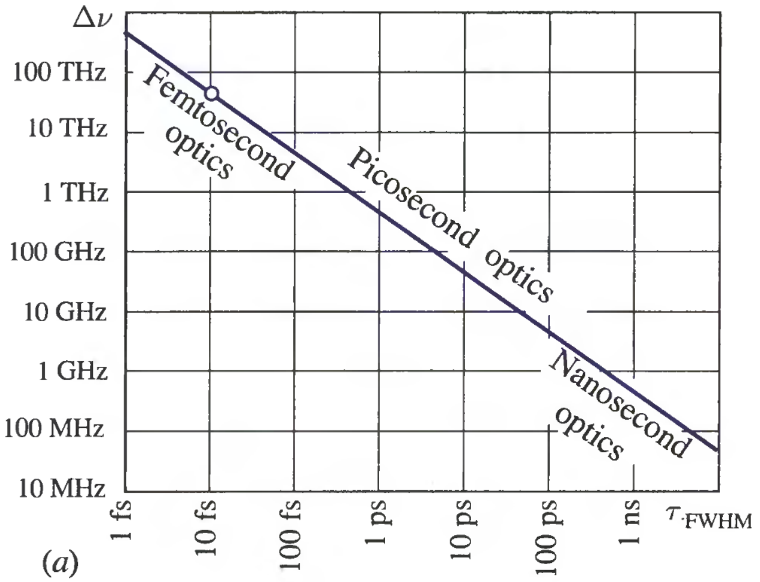
Time-bandwidth product

$$\Delta\nu \cdot \tau_p \geq k_{TBP} \sim 1$$

	$I(t)$ $(x \equiv t/\tau)$	τ_p/τ	$\Delta\nu_p \times \tau_p$
Gaussian	$I(t) = e^{-x^2}$	$2\sqrt{\ln 2}$	0.4413
Hyperbolic secant (soliton pulse)	$I(t) = \mathrm{sech}^2 x$	1.7627	0.3148



Freq. Bandwith vs time duration



Below 1 ps, we speak of ultrashort pulses: the spectral bandwidth is necessarily high!
Ti:Sapphire solid state material with the largest known bandwidth (230 nm)

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Nonlinear optical effects

Under a strong electric field, the polarization of a medium is no longer proportional to the fied:

Assuming a small deviation one can attempt to write a power series expansion for **P**: (FREQ. DOMAIN)

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

$\chi^{(q)}$ effects

$\chi^{(2)}$ is zero in media with inversion symmetry, typically the first non-zero term is $\chi^{(3)}$

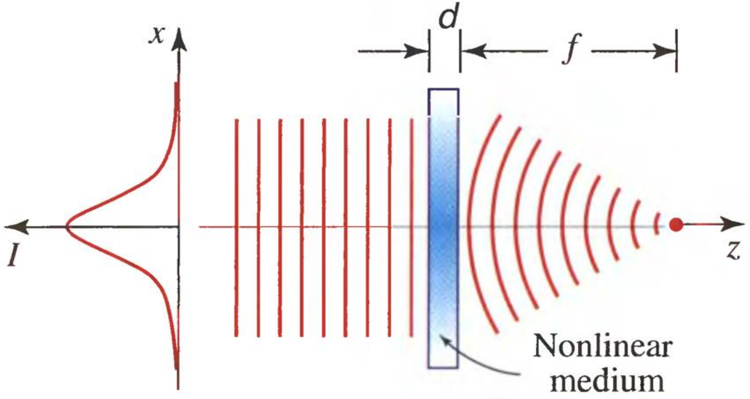
$\chi^{(3)}$ effects: the nonlinear index of refraction

$$\tilde{P}^{(3)}(t) = \chi^{(3)}\tilde{E}(t)^3$$

$$n(\omega, I) = n_0(\omega) + n_2(\omega) I.$$

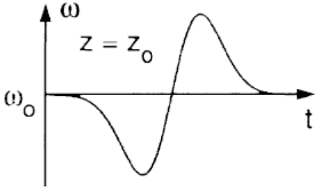
In space: Self-focusing

The radial variation of intensity translate into a radially varying optical path: this acts as a lens!



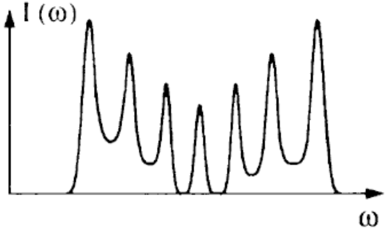
In time: Self-phase modulation and spectral broadening

$$\omega_{inst} = \frac{d\phi}{dt} = \omega_0 - A\frac{dI}{dt}$$



b)

spectral broadening



B integral

- The peak intensity during amplification increases as the pulse energy grows
- How to quantify the impact of n_2 ?

$$B = \frac{2\pi}{\lambda} \int_0^L \underbrace{n_2}_{\text{NONLINEAR INDEX OF REFRACTION}} I(z)\, dz$$

Remember, $\chi^{(3)}$ in time = self-phase modulation.

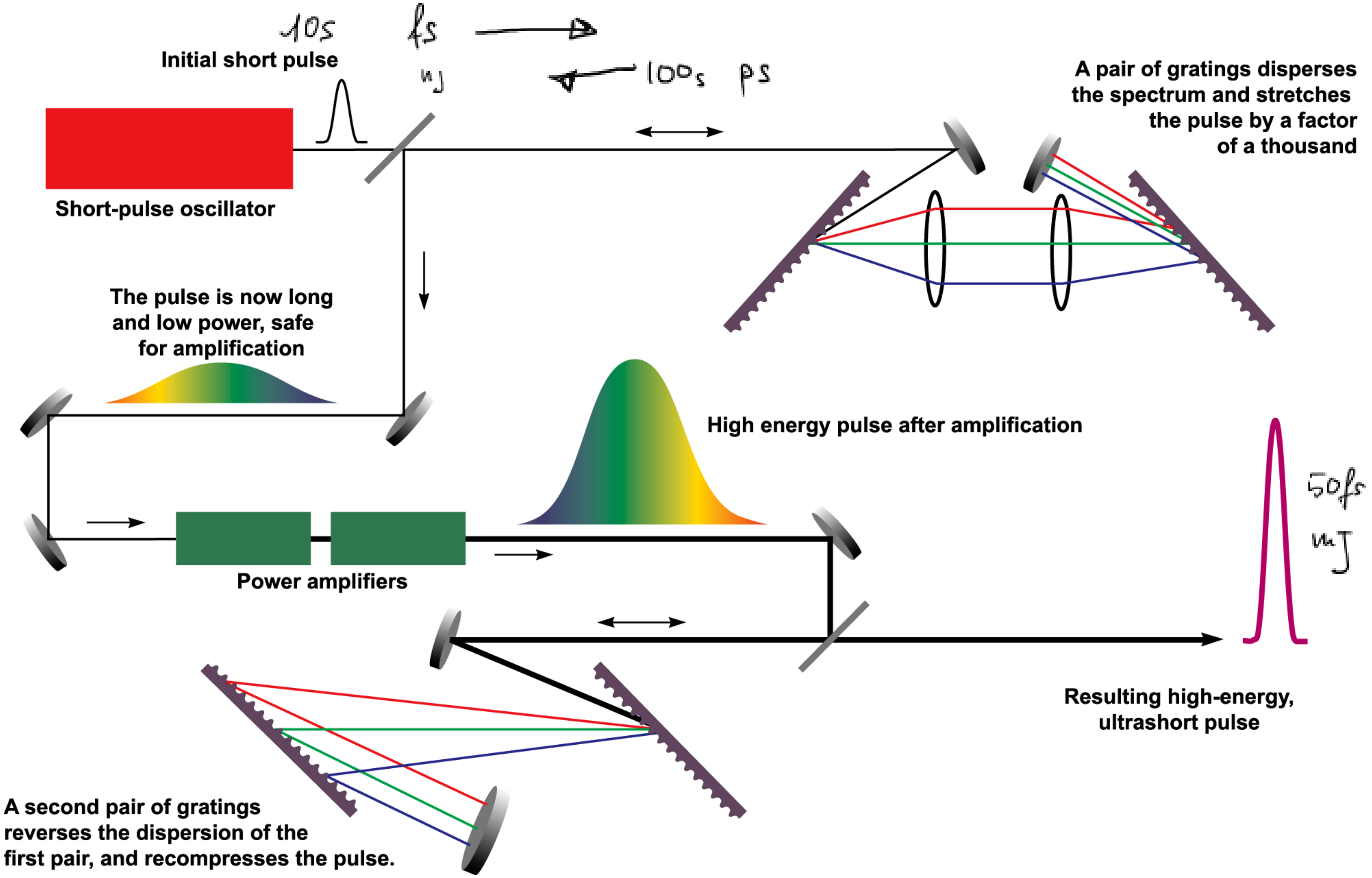
B-integral is a measure of the overall nonlinear phase accumulated. If B is too high, it is almost impossible to recompress the pulse.

Self-focusing might lead to damage of the optics

1. To further improve the laser intensities, the best route is to shorted the pulse duration
2. Self-effects become a fundament limit when amplifying short (picosecond and femtosecond) pulses

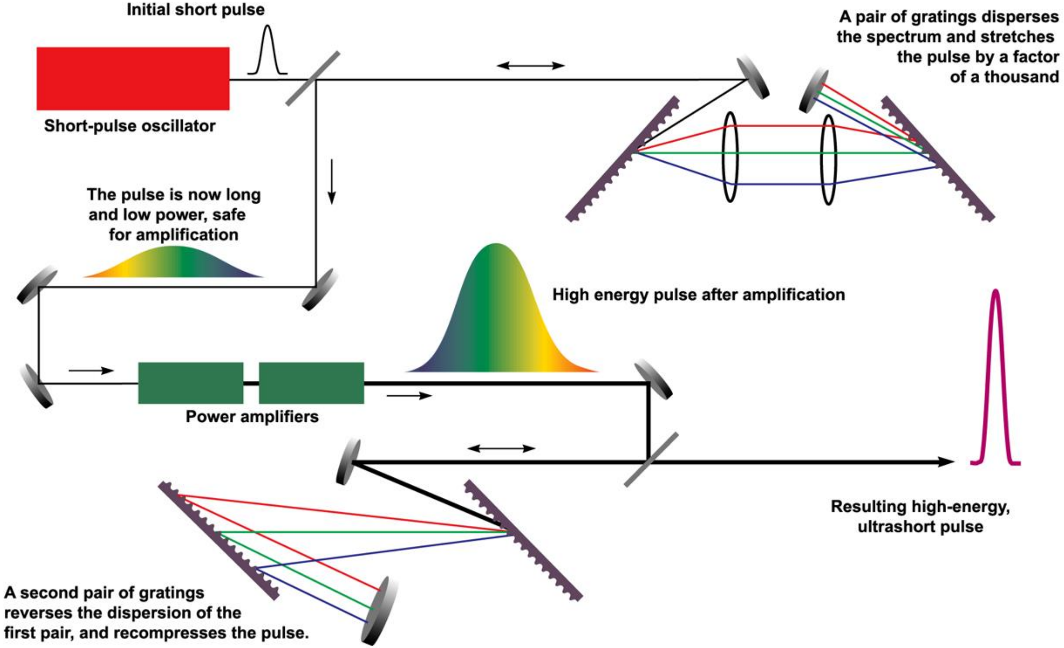
CHIRPED PULSE AMPLIFICATION

Chirped pulse amplification



Ultrashort pulse propagation

- How do we describe a femtosecond pulse?
- How does propagation the pulse duration?
- How can we control the pulse duration?



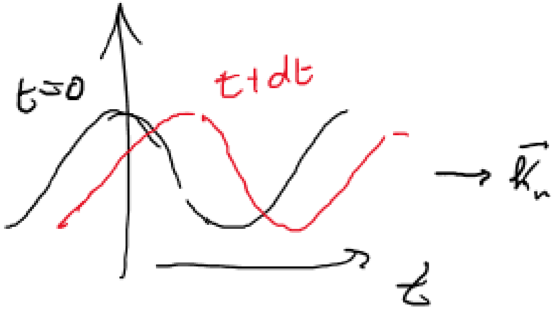
Pulse propagation some concepts: 1) Monochromatic wave, phase velocity

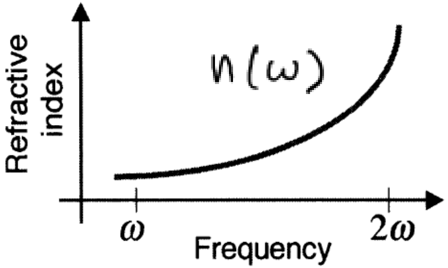
$$E(z,t) = A_\omega \exp(i\phi(z,t)) = A_\omega \exp[i(\omega t - \underbrace{k_n(\omega)}_{\text{MEDIUM DISPERSION RELATION}} z)]$$

$$k_n(\omega) = \frac{\omega}{c} n(\omega)$$

Phase front $d\phi = 0 \rightarrow \omega dt - k_n dz = 0$

$$v_{ph} = \frac{dz}{dt} = \frac{\omega}{k_n} = \frac{c}{n(\omega)}$$



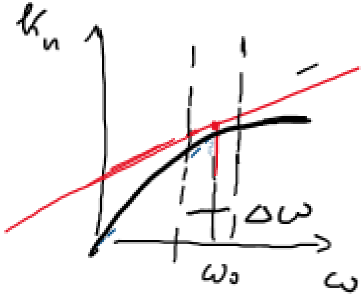


Phase velocity of the phase fronts of a plane wave in a material

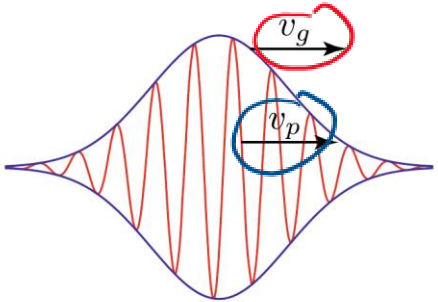
2) Quasi-monochromatic wavepackets

Wave packet: $E(t,z) = \int_{\Delta\omega} d\omega \, A_\omega \, e^{i(\omega t - k_n(\omega) z)}$

For frequency narrow pulses (e.g. ps) -> small $\Delta\omega$ around ω_0



LINEAR APPROXIMATION



Phase vs Group velocity

$$k_n(\omega - \omega_0) = k_n(\omega_0) + k_n'(\omega_0)\Delta\omega$$

$$E(t,z) \sim A\left(t - z/v_g\right) \exp\left(i\omega_0 t - k_n(\omega_0) z\right)$$

SAME SHAPE, TIME OFFSET

DIFFERENT

$$v_p = \frac{\omega}{k_n}$$

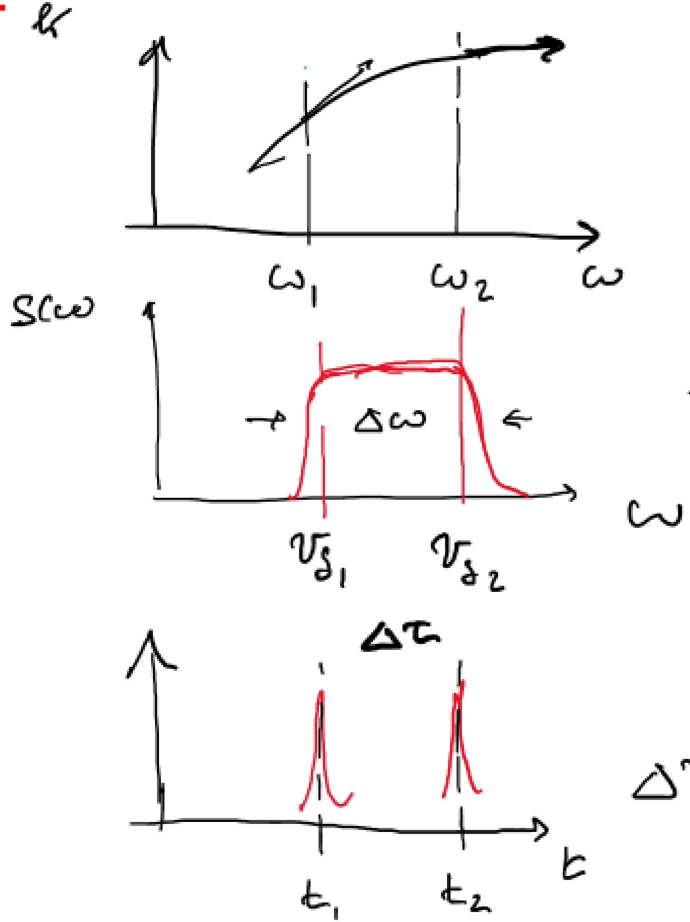
$$1/v_g = \frac{\partial k_n}{\partial \omega} \qquad v_g = \frac{\partial \omega}{\partial k_n} \qquad \text{" GROUP VELOCITY "}$$

$$t_g = z/v_g = \frac{\partial k_n}{\partial \omega} z \qquad \text{" GROUP DELAY "}$$

$$\varphi(\omega) = k_n(\omega) z$$

" SPECTRAL PHASE "

2) 2 x Quasi-monochromatic wavepackets



BANDWIDTH IS LARGE :
AS THE PULSE PROPAGATES
DIFFERENT PORTIONS OF THE SPECTRUM
HAVE DIFFEREN GROUP VELOCITIES.

→ ESTIMATE OF τ AFTER PROP. OF DISTANCE ℓ ?

$$\Delta\tau \sim \ell \, \Delta v_g^{-1} =$$

$$\varphi'(\omega_2) - \varphi'(\omega_1) \simeq \underbrace{\varphi''(\omega_0)}_{\text{GDD}} (\omega_2 - \omega_1)$$

GROUP DELAY DISPERSION

$$\Delta\tau \sim \varphi''(\omega_0)\,\Delta\omega$$

$$\ell \left| \frac{d^2 k_n}{d\omega^2} \right|_{\omega=\omega_0} \Delta\omega$$

→ GROUP VELOCITY DISPERSION $= \left. \frac{d}{d\omega}\left(\frac{1}{v_g}\right) \right|_{\omega=\omega_0}$

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$$\Delta\tau \sim \varphi''(\omega_0)\,\Delta\omega$$

- The formula works only in the limit of these frequency components to be well separated in time
- In the case of a femtosecond pulse this is the case only for very large GDD (large l or large GVD)

I need to properly to do a calculation based on Fourier transformation of the “spectral envelope” A(ω)

General idea: - expand the wavevector k(ω) of the pulse around the central frequency
- calculate the temporal shape by inverse Fourier transformation

Pulse propagation in an isotropic, linear medium

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1) Maxwell equations → 2) Wave equation → 3) Fourier transform (frequency domain)

HELMOLZ EQUATION

$$\frac{\partial^2 \tilde{E}(z,\omega)}{\partial z^2} + \frac{\omega^2}{c^2}\tilde{E}(z,\omega) = -\mu_0 \omega^2 \underbrace{\tilde{P}(z,\omega)}_{\varepsilon_0 \chi(\omega)\tilde{E}(z,\omega)}$$

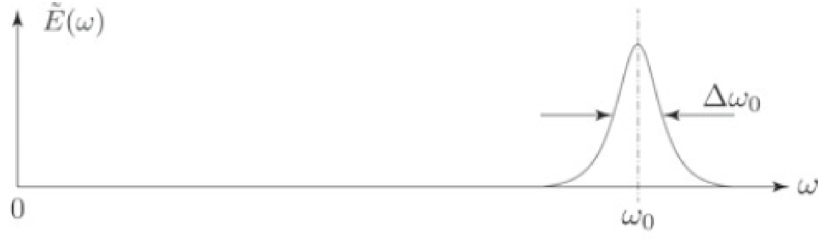
EXPRESSION FOLLOWS FROM THE ASSUMPTION OF LINEAR RESPONSE!

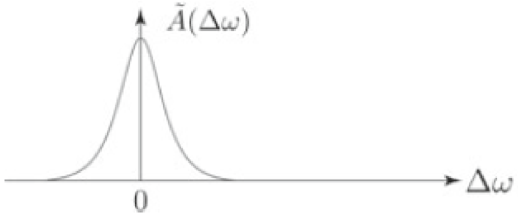
$$\frac{\partial^2 \tilde{E}(z,\omega)}{\partial z^2} + \frac{\omega^2}{c^2}\Big[\overbrace{1+\chi(\omega)}^{n^2(\omega)}\Big]\tilde{E}(z,\omega) = 0$$

$$k_n(\omega) = \frac{\omega}{c} n(\omega)$$

Wavepacket description

$$E(t) = \frac{1}{2\pi}\int \tilde{E}(\omega)e^{i\omega t}d\omega$$





$$E(t) = \frac{1}{2\pi}\int \underbrace{\tilde{E}(\omega_0+\Delta\omega)}\, e^{i(\omega_0+\Delta\omega)t}d\Delta\omega = \frac{1}{2\pi}e^{i\omega_0 t}\int \underbrace{\tilde{A}(\Delta\omega)}\,e^{i\Delta\omega t}d\Delta\omega$$

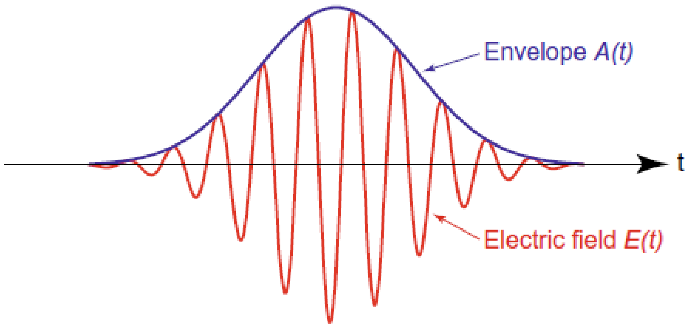
Frequency-shifted spectrum

$$E(t) = A(t)e^{i\omega_0 t}\ , \quad \text{where}\ \ A(t) = \frac{1}{2\pi}\int \tilde{A}(\Delta\omega)e^{i\Delta\omega t}d\Delta\omega$$

Temporal envelope

$$E(t) = A(t)\mathrm{e}^{\mathrm{i}\omega_0 t}\ , \quad \text{where}\ \ A(t) = \frac{1}{2\pi}\int \tilde{A}(\Delta\omega)\mathrm{e}^{\mathrm{i}\Delta\omega t}\,\mathrm{d}\Delta\omega$$

Temporal envelope

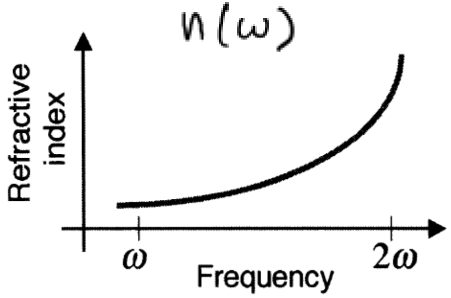


The propagation of a wave packet can be considerably simplified in the frequency domain:

$$\tilde{E}\left(z, \omega_0 + \Delta\omega\right) = \tilde{A}\left(z, \Delta\omega\right)\mathrm{e}^{-\mathrm{i}k_n(\omega_0)z}$$

Ultrashort pulse propagation in a **linear,** dispersive medium

- **linear** medium: no nonlinear effects (e.g. gain or saturable absorption)
- SVEA : Slowly Varying Envelope Approximation



$$\left| \frac{\partial^2 \tilde{A}}{\partial t^2} \right| \ll \left| k_n(\omega_0) \frac{\partial \tilde{A}}{\partial z} \right|$$

ENVELOPE DOES NOT CHANGE MUCH OVER DISTANCE

$dz \sim \lambda_n$

Small variation of the wavevector (can be treated as Taylor expansion)

$$k_n(\omega) = k_n(\omega_0) + \Delta k_n(\omega)$$

$$\tilde{A}(z,\Delta\omega) = \tilde{A}(0,\Delta\omega)\exp\left[i\Delta k(\omega) z\right] \leftarrow$$

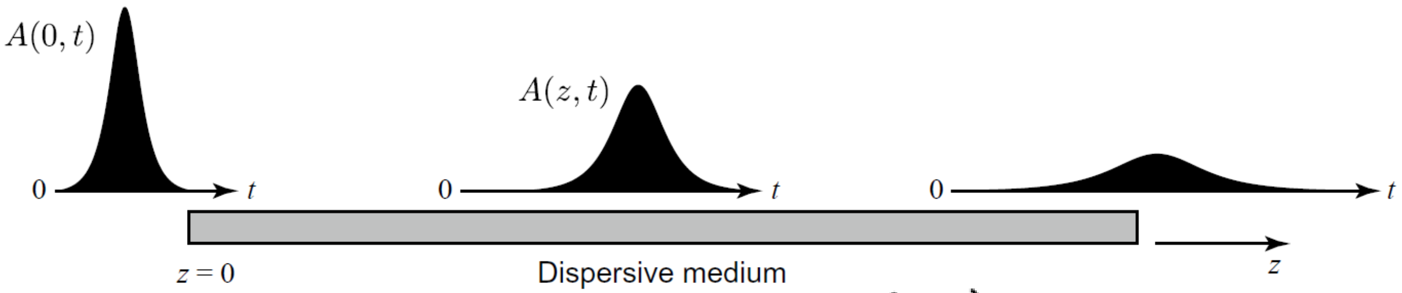
SIMPLE SOLUTION FOR THE SPECTRAL ENVELOPE

$$k_n(\omega) = k_n(\omega_0) + \Delta k_n(\omega)$$

TAYLOR EXPANSION OF THE SPECTRAL PHASE

$$\boxed{\varphi(\omega) = \varphi_0 + (\omega-\omega_0)\,\varphi_1 + (\omega-\omega_0)^2\,\varphi_2/2 + \cdots}$$

$\frac{\partial\varphi}{\partial\omega}$ GD $\quad \frac{\partial^2\varphi}{\partial\omega^2}$ GDD -- $\frac{\partial^3\varphi}{\partial\omega^3}$ TOD



A(z,t) DETERMINED BY INVERSE FT

$$A(z,t) = \frac{1}{2\pi}\int d\Delta\omega \; \tilde{A}(z,\Delta\omega)\, e^{+i\Delta\omega t}$$

TEMPORAL ENVELOPE

Temporal intensity envelope

REAL E-FIELD

TIME DEP. PHASE

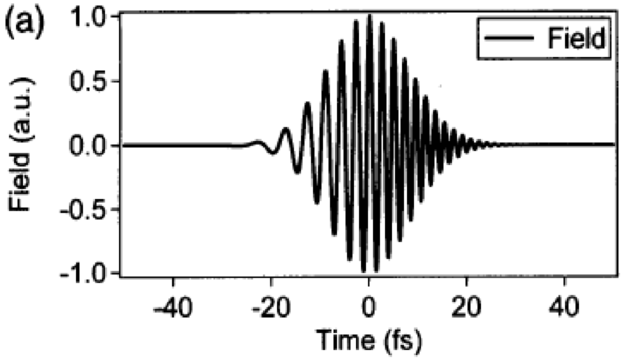
$$\boxed{\mathcal{E}(t) = \frac{1}{2}\sqrt{I(t)}\,\exp\{i\,[\omega t - \phi(t)]\} + c.c.}$$

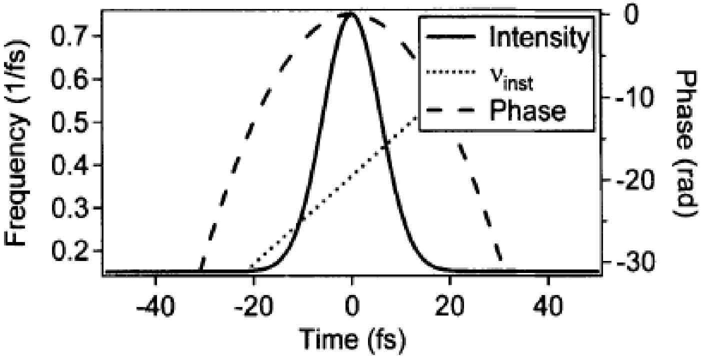
FAST OSCILLATING CARRIER

COMPLEX AMPLITUDE (NO CARRIER)

$$\boxed{E(t) \equiv \sqrt{I(t)}\,\exp[-i\phi(t)]}$$

Pulse duration: FWHM I(t)





$$\omega_{\text{inst}}(t) \equiv \omega_0 - \mathrm{d}\phi/\mathrm{d}t$$

$\varphi(t)$ is the TEMPORAL PHASE: information of frequency vs time

TIME vs FREQUENCY DOMAIN DESCRIPTIONS

TEMPORAL PHASE $\phi(t)$

$$\mathcal{E}(t) = \tfrac{1}{2}\sqrt{I(t)}\,\exp\{i\,[\omega t - \phi(t)]\} + c.c.$$

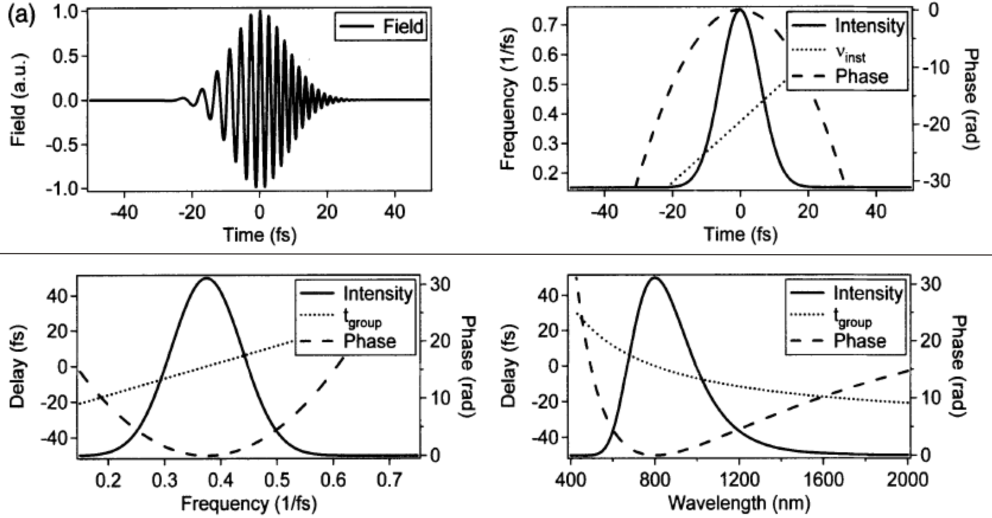
$$\tilde{\mathcal{E}}(\omega) = \int_{-\infty}^{\infty} \mathcal{E}(t)\exp(-i\omega t)\,dt$$

FOURIER TRANSFORM

Spectral description:

$$\tilde{\mathcal{E}}(\omega) = \sqrt{S(\omega)}\exp[-i\varphi(\omega)]$$

COMPLEX AMPLITUDE

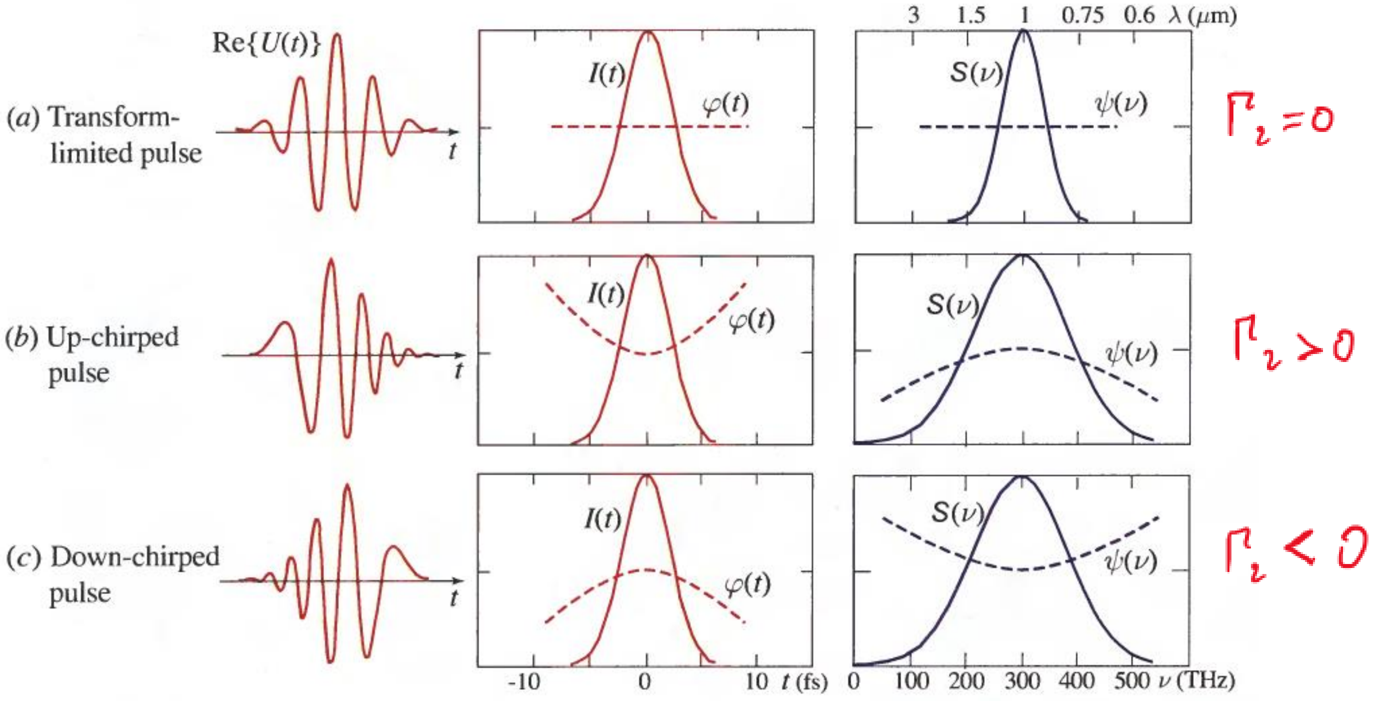


$\varphi(\omega)$ is the SPECTRAL PHASE: information of time vs frequency

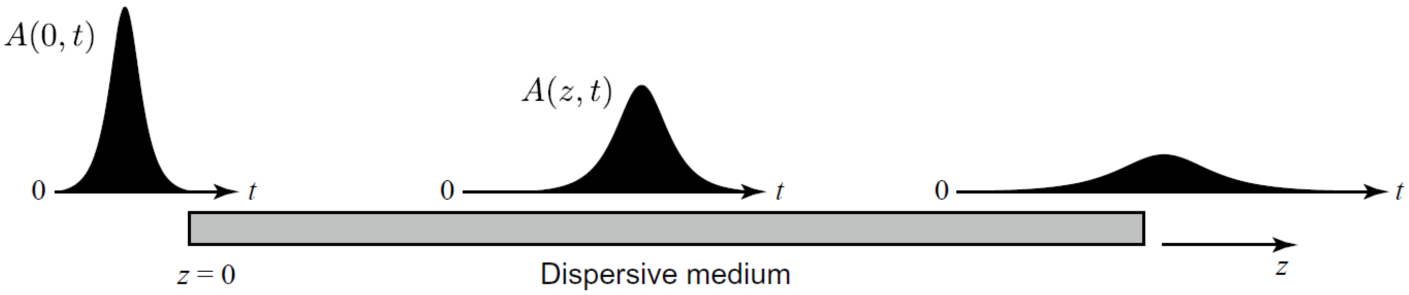
$\varphi(\omega)$

ϕ and φ DIFFERENT FUNCTIONS

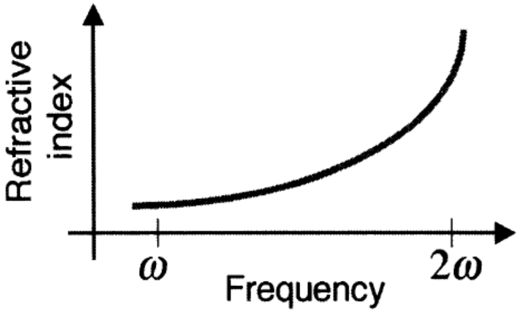
Chirped pulses



- A non-flat phase is the result of a non-transform limited pulse: by compensating the phase a shorted pulse could be produced.
- By controlling the phase one can stretch/compress the pulse.

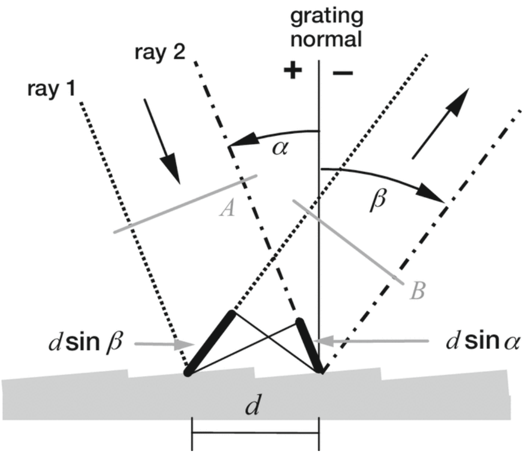


- Most material have “normal” dispersion at VIS-NIR wavelengths
- To compensate linear propagation temporal broadening, we need optical devices with opposite (“anomalous”) dispersion.
- Multiple materials in an amplifier, multiple passes.



Calculate the cumulative GDD (and TOD, and higher orders) -> Compensate with a suitable compressor (e.g. prism compressor, grating compressor..)

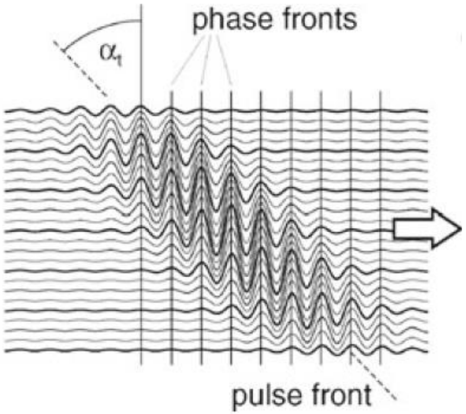
DIFFRACTION GRATING



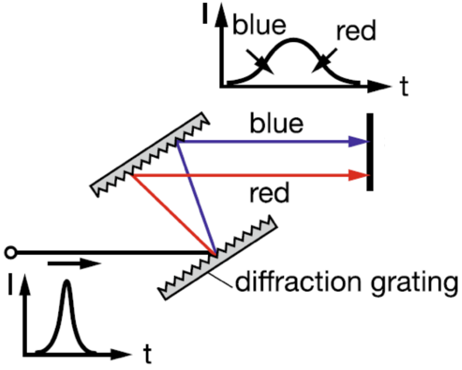
$$n\lambda = d(\sin\beta - \sin\alpha)$$

$$\Downarrow$$

$$\beta = \sin^{-1}\left[\frac{n\lambda}{d} + \sin\alpha\right]$$



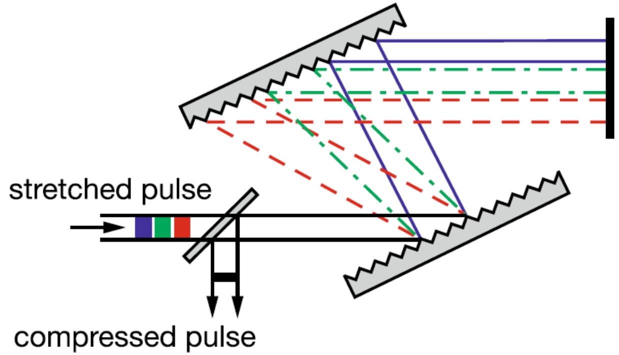
Periodic array of scatterers with spacing d = 1/Λ

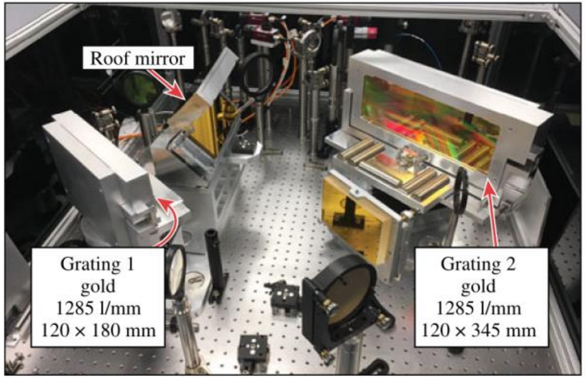


Angular dispersion: wavelength-dependent propagation angle.

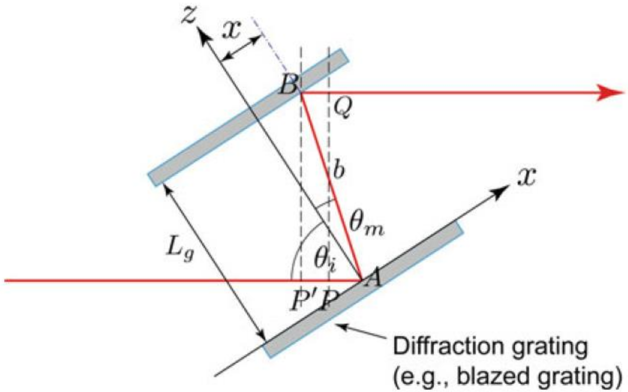
Pulse front tilt/spatial chirp: multiple grating configuration

TREACY COMPRESSOR





- 4 gratings are necessary for avoiding pulse front tilt and spatial chirp
- High efficiency is fundamental : transmission η^4
- Blazed gratings, near Littrow condition -> diffraction efficiency in the first order can approach 90%



2x contributions

$$\phi = \frac{\omega}{c} L + \phi_g(x)$$

$$x = L_g \tan\theta_m$$

1. Optical path

$$\frac{L_g}{\cos\theta_m}\left[1 + \cos(\theta_m + \theta_i)\right]$$

2. Spatial dependent phase shift due to diffraction

$$\phi_g(x) = \pi - m\frac{2\pi}{\Lambda}x$$

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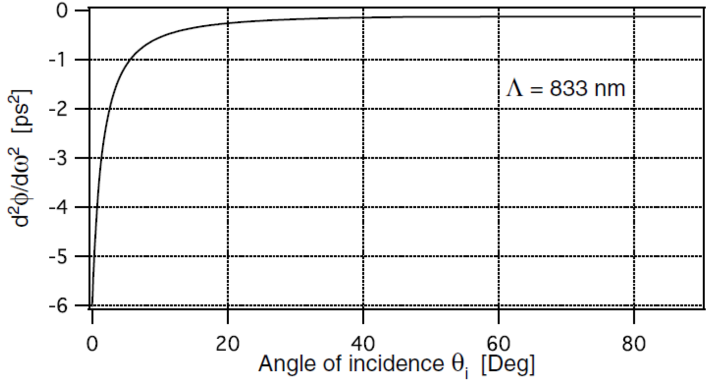
$$\frac{d^2\phi}{d\omega^2} = -\frac{\lambda^3 L_g}{\pi c^2 \Lambda^2}\left[1-\left(\frac{\lambda}{\Lambda}-\sin\theta_i\right)^2\right]^{-3/2}$$

Sapphire	n (800 nm) = 1.76019	
	$\left.\frac{\partial n}{\partial \lambda}\right\|_{800\text{ nm}} = -0.0268\frac{1}{\mu\text{m}}$	$\left.\frac{\partial k_n}{\partial \omega}\right\|_{800\text{ nm}} = 5.87\times10^{-9}\frac{\text{s}}{\text{m}}$
	$\left.\frac{\partial^2 n}{\partial \lambda^2}\right\|_{800\text{ nm}} = 0.064\frac{1}{\mu\text{m}^2}$	$\left.\frac{\partial^2 k_n}{\partial \omega^2}\right\|_{800\text{ nm}} = 5.80\times10^{-26}\frac{\text{s}^2}{\text{m}} = 58\frac{\text{fs}^2}{\text{mm}}$
	$\left.\frac{\partial^3 n}{\partial \lambda^3}\right\|_{800\text{ nm}} = -0.377\frac{1}{\mu\text{m}^3}$	$\left.\frac{\partial^3 k_n}{\partial \omega^3}\right\|_{800\text{ nm}} = 4.21\times10^{-41}\frac{\text{s}^3}{\text{m}} = 42.1\frac{\text{fs}^3}{\text{mm}}$



Example: Reg. Ti:Sapphire, 5 mm Crystal, 10 passes -> 58 x 5 x10 = 2.9 x 10^3 fs^2

Grating compressor, 1200 lines/mm, d= 5 cm, 20° incidence -> -0.2 ps^2 = -0.2 x 10^6 fs^2

- Gratings allow for a very large amount of GVD compared to propagation in materials
- In a compact setup from 10 fs -> 500 ps (would require 50 m of fused silica)

Grating compressor -> Negative dispersion: pulse compression during amplification!

GRATING STRETCHER

$$\boxed{\frac{d^2\phi}{d\omega^2} = -\frac{\lambda^3 L_g}{\pi c^2 \Lambda^2}\left[1-\left(\frac{\lambda}{\Lambda}-\sin\theta_i\right)^2\right]^{-3/2}}$$

$$\frac{d^2\phi}{d\omega^2} = -\frac{m^2\lambda^3 M^2 L_{\text{eff}}}{2\pi c^2 \Lambda^2}\left[1-\left(-m\frac{\lambda}{\Lambda}-\sin\theta_i\right)^2\right]^{-3/2}$$

$L < f$ positive GDD

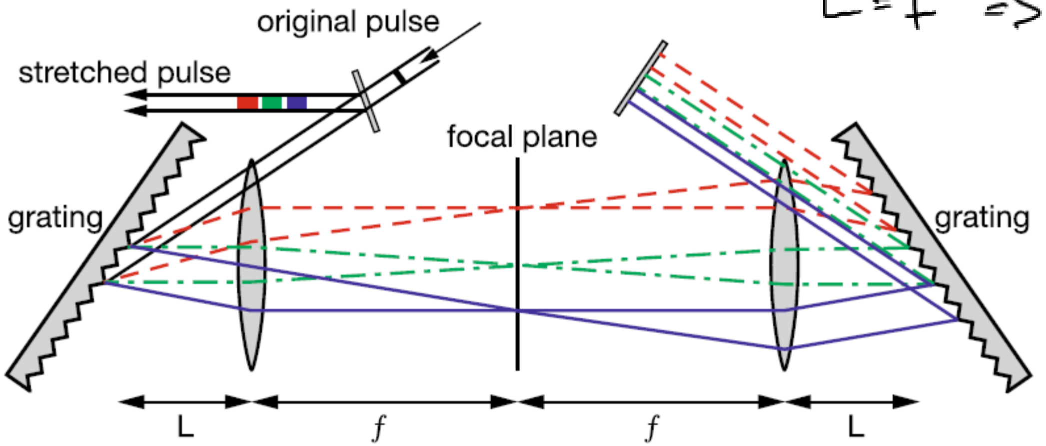
$L \geq f$ negative GDD

$L = f \Rightarrow$ 4f system

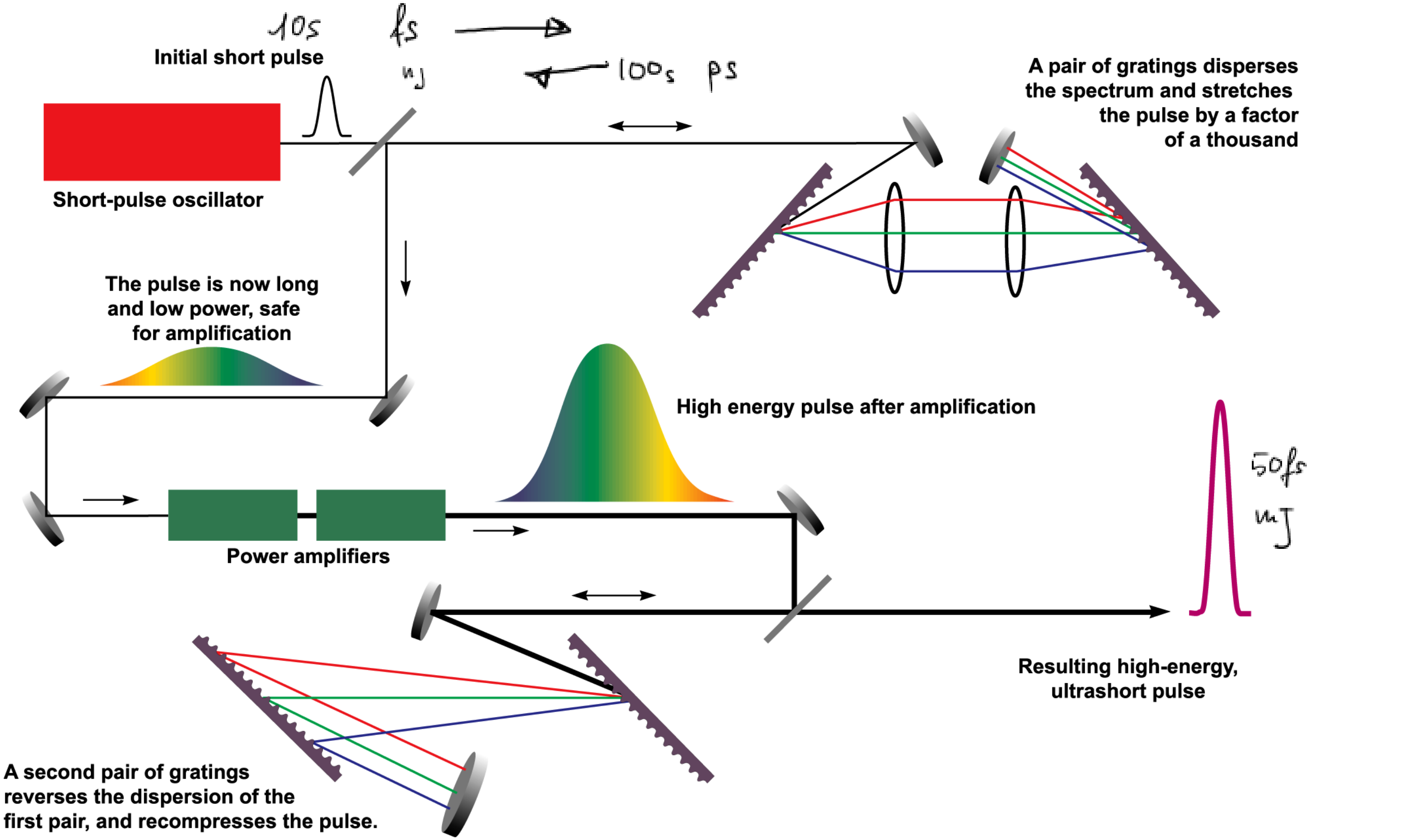
GDD = 0

CAN USE TO "SHAPE" THE PHASE

- "Oeffner" stretcher



Chirped pulse amplification



CPA and GAIN NARROWING

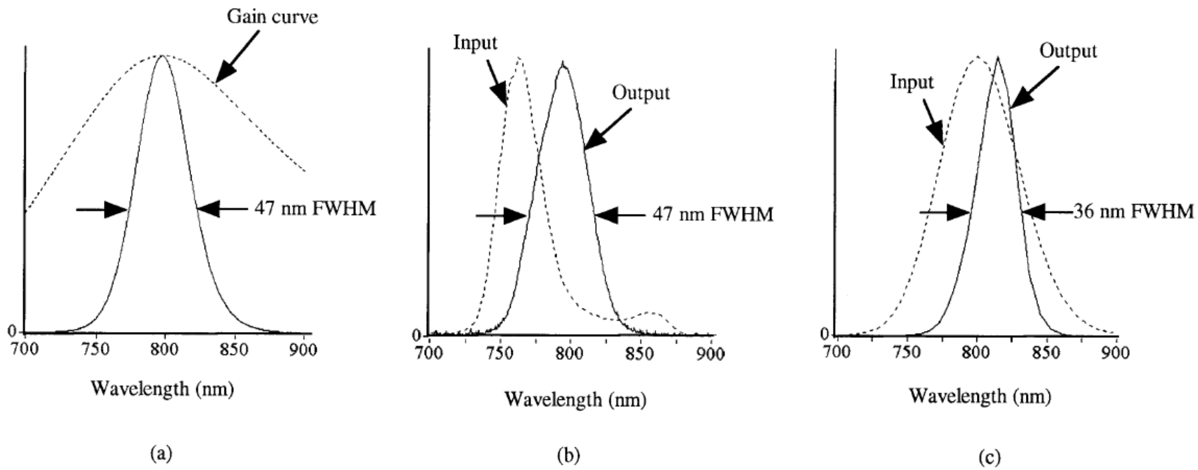


FIG. 5. Gain narrowing for the case of (a) an infinitely broad and flat input spectrum, (b) an optimally offset and shaped input spectrum, and (c) a nonoptimum input spectrum.

GAIN NARROWING IN TI:SAPPHIRE LIMITS THE ACHIEVABLE AMPLIFIED PULSE DURATION

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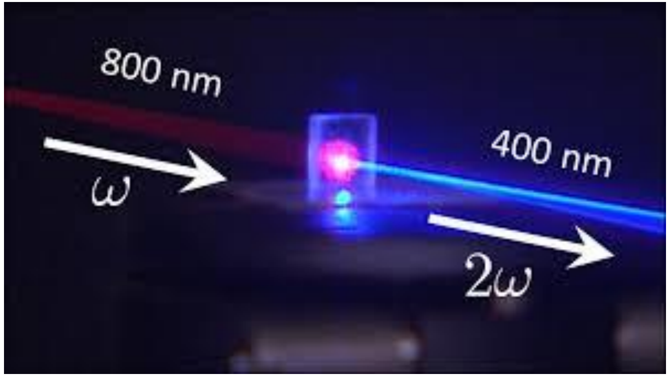
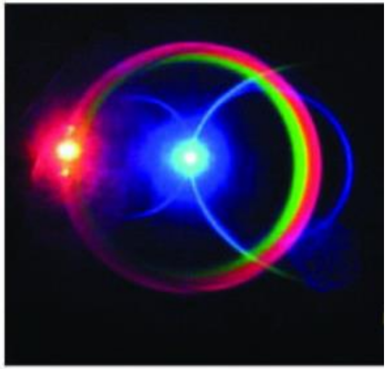
PERTURBATIVE NONLINEAR OPTICS

CPA of femtosecond pulse:

- Very high peak intensity
- Breakdown of linear response of the material -> Nonlinear optics



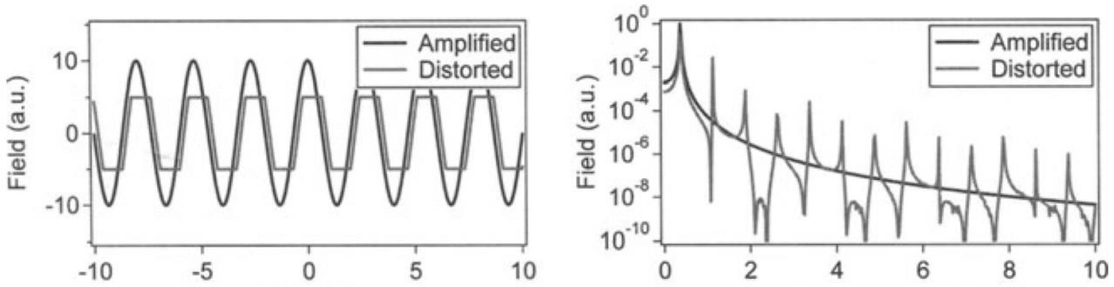
Second harmonic generation



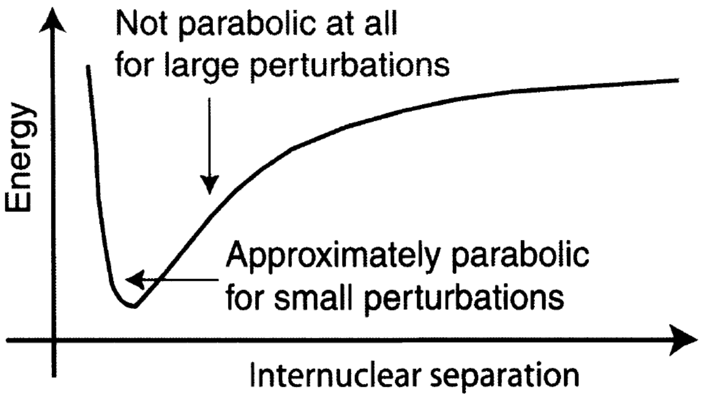
Parametric amplification

Nonlinear effects some examples

Example 1: distortion in an electronic amplifier driven by a sin wave: the clipping results in harmonics of the driver



Example 1: nuclear vibrations anharmonicity



Due to the deviations from a perfect parabolic potential (harmonic oscillator), a system driven at a certain ω, will oscillates also at other frequencies!

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Nonlinear optical effects

Under a strong electric field, the polarization of a medium is no longer proportional to the fied:

Assuming a small deviation one can attempt to write a power series expansion for **P**:

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

Wave equation: **P** act as a source term:

$$\frac{\partial^2 \mathcal{E}}{\partial z^2} - \frac{1}{c_0^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathcal{P}}{\partial t^2}$$

«Perturbative» non-linear optics:

$\chi^{(q)}$ effects

$$\mathcal{E}(t) \propto \cos(\omega t) \longrightarrow \wp(t) \propto \cos^q(\omega t)$$

$$\cos^{2n-1} A = \frac{1}{2^{2n-2}} \left\{ \cos(2n-1)A + \binom{2n-1}{1} \cos(2n-3)A + \cdots + \binom{2n-1}{n-1} \cos A \right\}$$

When do nonlinear optical effects appear?

ELECTRONIC nonlinearities physical origin: strong distortion of the valence orbitals -> breakdown of anharmonic response

At which E the linear and quadratic term become comparable?

$$\varepsilon_0 \chi^{(1)} E \sim \varepsilon_0 \chi^{(2)} E^2$$

$$\hookrightarrow E \sim \frac{\chi^{(1)}}{\chi^{(2)}} \quad \chi^{(1)} \rightarrow \mathcal{O}(1)$$

Order-magnitude electric field acting on a valence electron:

$$E \sim \frac{1}{4\pi\varepsilon_0} \frac{e}{a_0^2}$$

$e \rightarrow$ e^- charge

$a_0 \rightarrow$ Bohr radius

$E_{at} = 5 \times 10^{11}$ V/m

$$\frac{\chi^{(n)}}{\chi^{(n-1)}} \sim \frac{1}{E_{at}}$$

E_{at}: INTERATOMIC ELECTRIC FIELD

(J.Mod.Opt., 1999, VOL. 46, NO. 3, 367) the magnitude of the response of order n is related to the (n-1)th

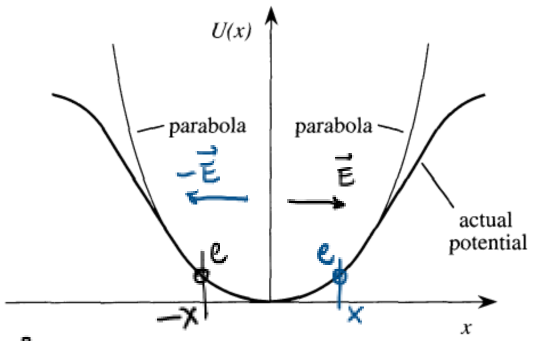
Electric field of solar radiation on the earth surface:

$E_{sun} \approx 3$ V/m (in a 1 nm bandwidth at 500 nm)

Nonlinear effects typically require laser light to be observed: nonlinear optics became widespread only after the ruby laser. The $\chi^{(2)}$ nonlinear response should dominate.

Problem: vanishing $\chi^{(2)}$

Given these estimates, the $\chi^{(2)}$ response should dominate:



$P(t) = \chi^{(2)} E^2(t)$ $p \sim -ex$ $p \sim ex$

$-P(t) = \chi^{(2)} (-E(t))^2 \overset{!}{=} \chi^{(2)} E^2(t)$

$\Longrightarrow \chi^{(2)} = 0$!

FOR A SYMMETRIC POTENTIAL A REVERSAL OF $\vec{E}$ ⟹ REVERSAL OF $\vec{P}$

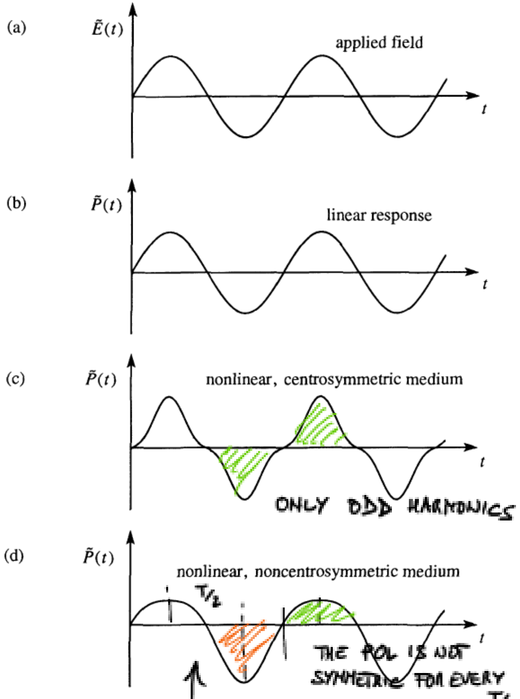


FIGURE 1.5.1 Waveforms associated with the atomic response.

EVEN HARMONICS

ODD ORDERS cannot be observed in centro-symmetric media: many crystal classes lack inversion symmetry and exhibit non-vanishing $\chi^{(2)}$.
Example silica (SiO_2): $\chi^{(2)} = 0$ in glass, $\chi^{(2)} \neq 0$ in quartz single crystals

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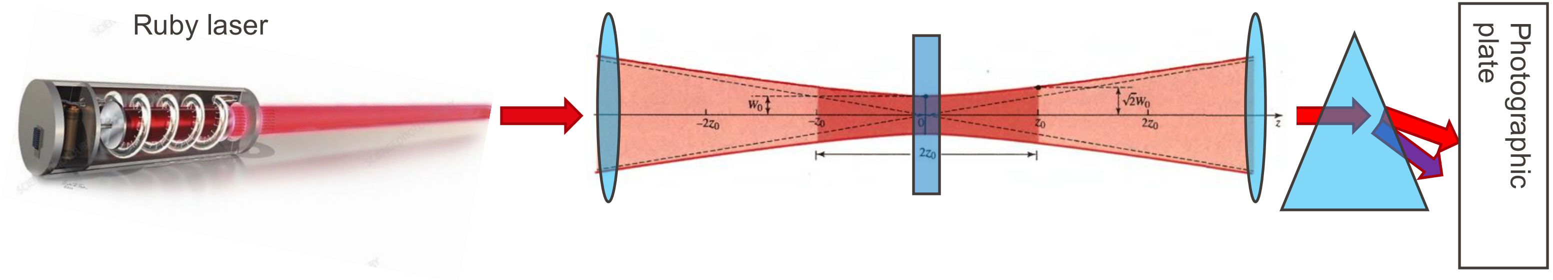
The first nonlinear optics experiment with a ruby laser:



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

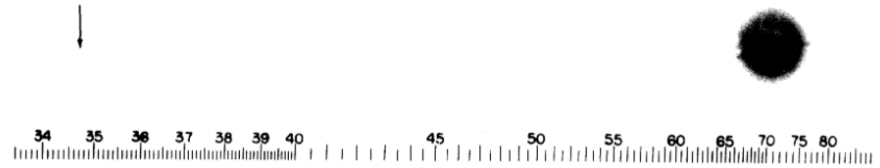


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

Can you see the spot?

See APS Landmarks: Ruby Red Laser light become ultraviolet https://physics.aps.org/articles/v7/112#

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Second harmonic generation now:

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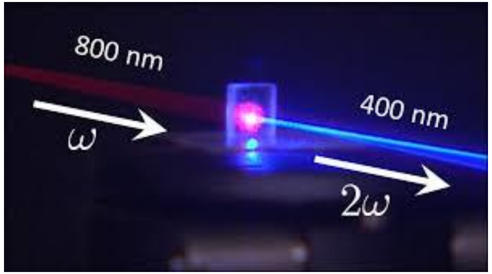
- Non-linear crystals: materials with very high $\chi^{(2)}$ (example BBO, LBO ..)
- Higher intensities are routinely produced (GW pulses readily available in fs lasers)
- Phase matching: for the beam to grow over macroscopic distances, microscopic dipoles must radiate in phase and interfere constructively over the crystal length ($L_{coh} > L_{crystal}$)



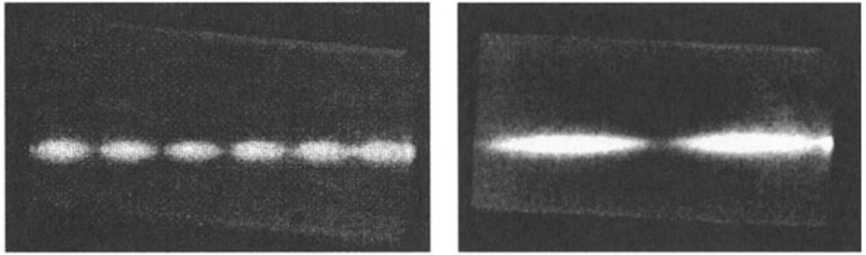
PHASE MATCHING CONDITION

$$\Delta k = 2k_{FH} - k_{SH} = 0$$

FUNDAMENTAL BEAM WAVEVECTOR

SH WAVEVECTOR

Far from phase-matching | Closer to phase-matching



Six coherence lengths | Two coherence lengths

- Can reach high efficiencies (> 50% is not atypical, in some cases close to unity)

$\chi^{(2)}$ effects couple two interacting waves:

- Driving field: $$\tilde{E}(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + \text{c.c.}$$
- Resulting polarization: $$\tilde{P}^{(2)}(t) = \chi^{(2)} \tilde{E}(t)^2$$

$$\tilde{P}^{(2)}(t) = \chi^{(2)}\left[E_1^2 e^{-2i\omega_1 t} + E_2^2 e^{-2i\omega_2 t} + 2E_1E_2 e^{-i(\omega_1+\omega_2)t} + 2E_1E_2^* e^{-i(\omega_1-\omega_2)t} + \text{c.c.}\right] + 2\chi^{(2)}\underbrace{[E_1E_1^* + E_2E_2^*]}_{\text{DC TERM}}.$$

HIGH FREQUENCY A.C.

DC TERM

CORRESPOND TO A STATIC ELECTRIC FIELD

$$\tilde{P}^{(2)}(t) = \sum_n P(\omega_n) e^{-i\omega_n t}$$

The polarization can also be expressed in terms of its frequency components

$$P(2\omega_1) = \chi^{(2)} E_1^2 \quad \text{(SHG)},$$

$$P(2\omega_2) = \chi^{(2)} E_2^2 \quad \text{(SHG)},$$

SECOND HARMONIC GENERATION

$$P(\omega_1 + \omega_2) = 2\chi^{(2)} E_1 E_2 \quad \text{(SFG)},$$

Sum Frequency Generation

$$P(\omega_1 - \omega_2) = 2\chi^{(2)} E_1 E_2^* \quad \text{(DFG)},$$

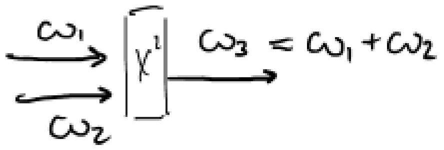
Difference Frequency Generation

$$P(0) = 2\chi^{(2)} (E_1 E_1^* + E_2 E_2^*) \quad \text{(OR)}.$$

Optical rectification

*For every component P(ω), there is also P(-ω), however P(-ω)=P(ω)**

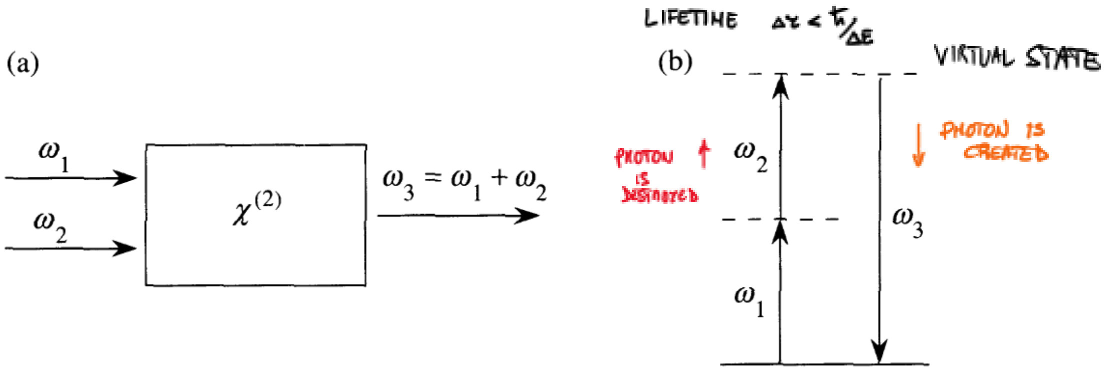
Three wave mixing : in $\chi^{(2)}$ processes three waves interact thanks to the nonlinear susceptibility



Parametric processes

- A nonlinear optical process which leave the quantum state unchanged is called parametric process
- No energy is deposited in the material! Photon energy conservation is always satisfied
- Energy level diagrams with «virtual states»: *example SFG*

Two photons of energy $\hbar\omega_1$ and $\hbar\omega_2$ are absorbed via virtual states, the end state decays emitting a photon of energy $\hbar\omega_3 = \hbar\omega_1 + \hbar\omega_2$



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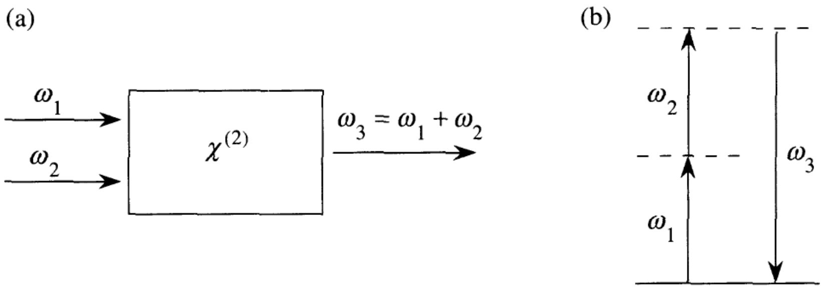
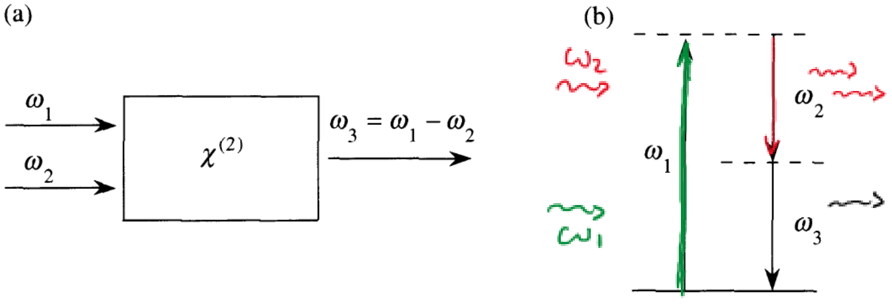
SFG and DFG

- Sum Frequency generation (SFG):



- Two photons are destroyed and a photon at the sum energy is created

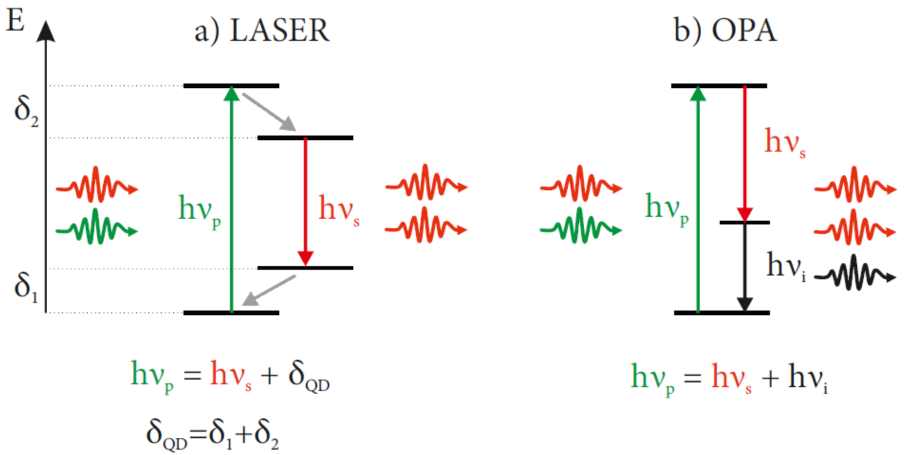
- Difference frequency generation (DFG):



- Here a virtual state, excited by the highest energy photon decay by emitting two photons,
- Beam ω_2 is amplified in the process

OPTICAL PARAMETRIC AMPLIFICATION (OPA)

Chirped pulse amplification and OPA: OPCPA



Improved Average Power scalability

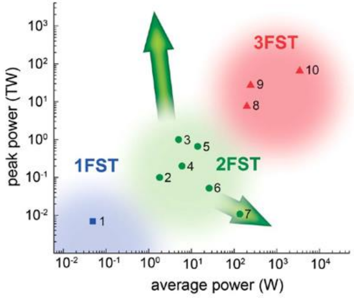
Bandwidth

Promising technology for the next generation of femtosecond lasers:

1st generation: Dye laser -> Short pulse

2nd generation: Ti:Sapphire -> Shorter pulses, high pulse power

3rd generation: Ytterbium-based OPCPAs -> Shorter pulses, higher pulse power, higher average power, frequency tunability



- Needs high power sub-picosecond Ytterbium lasers (kWs average power, and GW pulse power at 100s of KHz).

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Nonlinear non-parametric processes:

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- Saturable absorption/amplification:

$$\alpha = \frac{\alpha_0}{1 + I/I_s}$$

$$I_{sat} = \frac{h\nu}{\sigma\tau}$$

- Two-photon absorption:

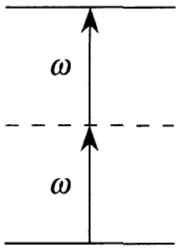
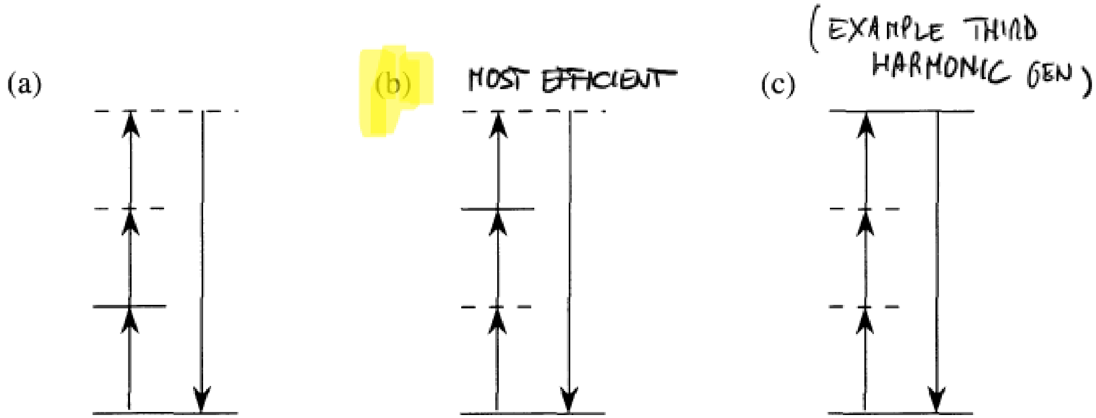


FIGURE 1.2.10 Two-photon absorption.

- Resonant enhancement of nonlinear processes:



- One or more step of a diagram corresponds to one of the system resonances
- Strong «resonant enhancement» of nonlinear effects, but also absorption of the beam with population transfer might occur

Classical electrodynamics treatment of non-resonant $\chi^{(2)}$ effects

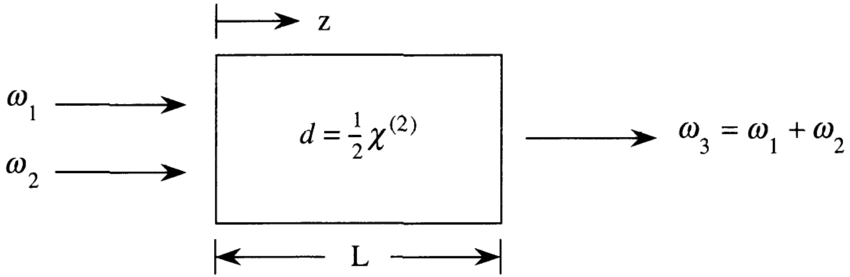


FIGURE 2.2.1 Sum-frequency generation.

- Perfectly monochromatic, plane-waves.
- Perfectly lossless medium
- For every frequency component one must solve a wave equation:

$$-\nabla^2 \mathbf{E}_n(\mathbf{r}) - \frac{\omega_n^2}{c^2}\epsilon^{(1)}(\omega_n) \cdot \mathbf{E}_n(\mathbf{r}) = \frac{4\pi\omega_n^2}{c^2}\mathbf{P}_n^{\mathrm{NL}}(\mathbf{r})$$

COUPLES E_3 WITH E_1 AND E_2

- The various equation are coupled through P^{NL}

Wave equation for Sum Frequency Generation (SFG)

$$\tilde{E}_i(z,t) = E_i e^{-i\omega_i t} + \text{c.c.}, \qquad i = 1, 2,$$

SPATIALLY OSCILLATING (EVERY λ_i)

$$E_i = A_i e^{ik_i z}, \qquad i = 1, 2,$$

$$\tilde{P}_3(z,t) = P_3 e^{-i\omega_3 t} + \text{c.c.},$$

$$P_3 = 4 d_{\text{eff}} E_1 E_2$$

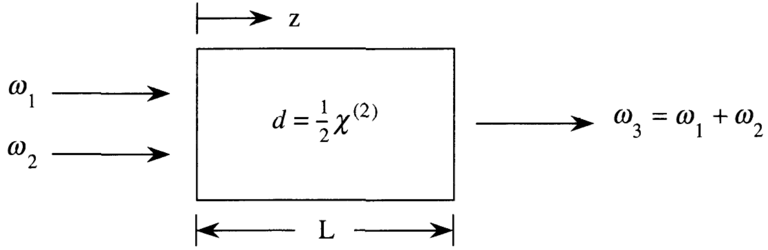


FIGURE 2.2.1 Sum-frequency generation.

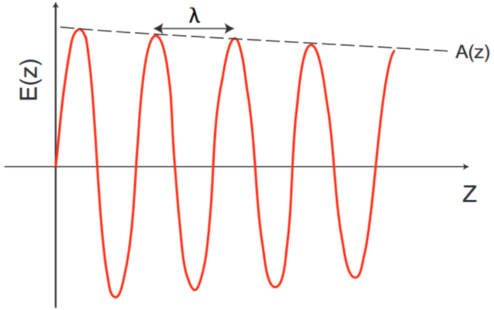
- Let’s write the amplitude for A_3 (the field generated at the sum-frequency field)

$$\frac{d^2 A_3}{dz^2} + 2ik_3 \frac{dA_3}{dz} = \frac{-16\pi d_{\text{eff}} \omega_3^2}{c^2} A_1 A_2 e^{i(k_1+k_2-k_3)z}.$$

- Slowly Varying Amplitude Approximation:

The amplitude does not change considerably over distances comparable with the light wavelength

In absence of non-linear polarization it would be a constant -> Nonlinear effects are typically small..



$$\left|\frac{d^2A_3}{dz^2}\right| \ll \left|k_3\frac{dA_3}{dz}\right|$$

- The second derivative term is dropped

$$\frac{dA_3}{dz} = \frac{8\pi i d_{\text{eff}}\omega_3^2}{k_3c^2}A_1A_2e^{i\Delta kz}$$

$$\frac{dA_1}{dz} = \frac{8\pi i d_{\text{eff}}\omega_1^2}{k_1c^2}A_3A_2^*e^{-i\Delta kz}$$

$$\frac{dA_2}{dz} = \frac{8\pi i d_{\text{eff}}\omega_2^2}{k_2c^2}A_3A_1^*e^{-i\Delta kz}$$

SIGN DIFFERENCE!

$$\Delta k = k_1 + k_2 - k_3$$

PHASE MISMATCH

Undepleted wave limit:

- In the case of low efficiencies, the deplections of the initial two beams (1 and 2) can be neglected, the equations for A_3 is decoupled and can be integrated.
- One obtains:

"CONDENSED" $\chi^{(2)}$ PRODUCT OF INTENSITIES !

$$I_3 = \frac{512\pi^5 d_{\text{eff}}^2 \overbrace{I_1 I_2}}{n_1 n_2 n_3 \lambda_3^2 c} L^2 \operatorname{sinc}^2(\Delta k L/2)$$

- In the case of Δk=0 the SFG beam intensity grows quadratically along the medium!

- Solution valid until the beam does not grow significantly, afterwards the approximation fails and the coupling needs to be accounted: back-conversion from 3 to 1 and 2 can be observed!

Phase matching

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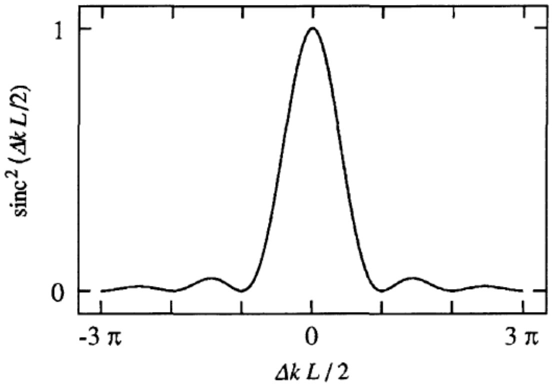
- The quadratic growth occurs if the wavevector mismatch is approximately zero:

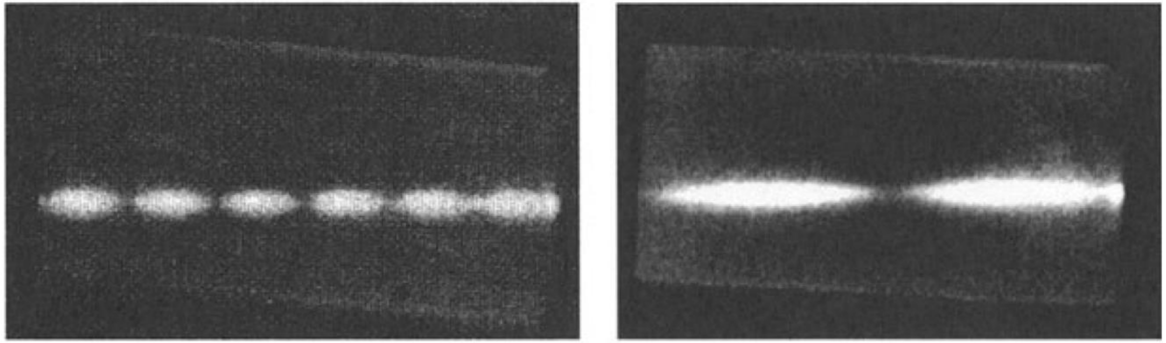
$$\Delta k = k_1 + k_2 - k_3 \approx 0$$

- Coherence length: $\pi/\Delta k$



Far from phase-matching

Closer to phase-matching



Six coherence lengths

Two coherence lengths

Perfect phase-matching seems a very special condition:

$$\Delta k = 2k_\omega - k_{2\omega} = 0$$

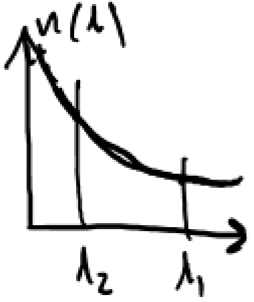
$$\Updownarrow \qquad \| $$

$$= \frac{2\omega}{c} n(\omega) - \frac{2\omega}{c} n(2\omega) = 0$$

SAME PHASE VEL. $\quad n(2\omega) = n(\omega)$

NORMAL DISPERSION

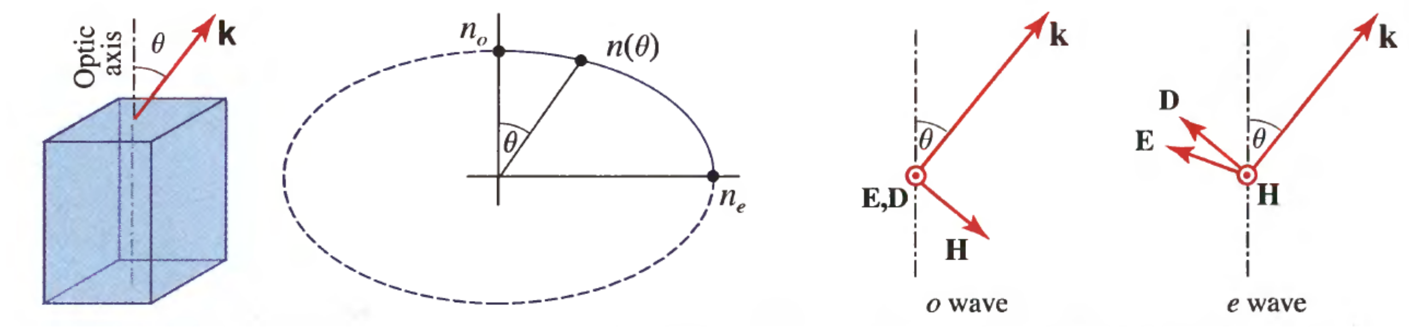
$n(\omega) \neq n(2\omega)$



Typically $n(2\omega) > n(\omega)$

Refractive index wavelength dependence

Critical phase matching in uniaxial crystals



- The refractive index of waves is determined entirely by the angle between **k** and the optical axis.
- ordinary-wave **E** ortogonal to the optical axis.
- Positive uniaxial (n_o> n_e) , negative uniaxial (n_e> n_o)

$$\frac{1}{n^2(\theta)} = \frac{\cos^2\theta}{n_o^2} + \frac{\sin^2\theta}{n_e^2}$$

- This relation can be used to achieve phase matching!
- Type I: the signal and idler waves have the same polarization, orthogonal to the pump oo->e or ee->o
- Type II: oe->o or oe->e

WAVE PROPAGATION AND WAVE EQUATION IN NON-ISOTROPIC MEDIUM HAS TO BE CONSIDERED

$\chi^{(2)}$ Nonlinear optical effects in 3d

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i, j, k CARTESIAN COORDINATES

$$P_i(\underbrace{\omega_n + \omega_m}_{\text{FREQ. OF POLARIZ.}}) = \sum_{jk} \sum_{(nm)} \chi^{(2)}_{ijk}(\overbrace{\omega_n + \omega_m}^{\text{GEN. FREQ.}}, \overbrace{\omega_n, \omega_m}^{\text{DRIVING FREQ.}}) E_j(\omega_n) E_k(\omega_m)$$

(nm): SUMMATION PERFORMED FOR FIXED $\omega_n + \omega_m$

- To include all possible polarizations generated by three interacting waves 6 tensor (x 2 counting the negative ω) have to be determined: each one has 3^3=27 components:

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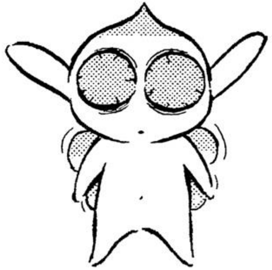
$$\chi^{(2)}_{ijk}(\omega_1, \omega_3, -\omega_2), \quad \chi^{(2)}_{ijk}(\omega_1, -\omega_2, \omega_3), \quad \chi^{(2)}_{ijk}(\omega_2, \omega_3, -\omega_1),$$
$$\chi^{(2)}_{ijk}(\omega_2, -\omega_1, \omega_3), \quad \chi^{(2)}_{ijk}(\omega_3, \omega_1, \omega_2), \quad \text{and} \quad \chi^{(2)}_{ijk}(\omega_3, \omega_2, \omega_1)$$



- In practice often the problem is simplified:
  - symmetry selection rules
  - NON-RESONANT ELECTRONIC PROCESSES:
    - the three interacting waves are very far from the lowest resonance in the crystal ($\chi^{(2)}$ independent of ω)

Voigt notation

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$$d_{ijk} = \frac{1}{2}\chi^{(2)}_{ijk}$$

jk:	11	22	33	23, 32	31, 13	12, 21
l:	1	2	3	4	5	6

- The d_{ij} are tabulated for most crystals

$$d_{il} = \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix}$$

- Example polarization for SFG:

$$\begin{bmatrix} P_x(\omega_3) \\ P_y(\omega_3) \\ P_z(\omega_3) \end{bmatrix} = 4 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \times \begin{bmatrix} E_x(\omega_1)E_x(\omega_2) \\ E_y(\omega_1)E_y(\omega_2) \\ E_z(\omega_1)E_z(\omega_2) \\ E_y(\omega_1)E_z(\omega_2) + E_z(\omega_1)E_y(\omega_2) \\ E_x(\omega_1)E_z(\omega_2) + E_z(\omega_1)E_x(\omega_2) \\ E_x(\omega_1)E_y(\omega_2) + E_y(\omega_1)E_x(\omega_2) \end{bmatrix}$$

The effective nonlinear coefficient

- Depending on the crystal symmetry class (there are 32 x classes) several elements are redundant or identically zero! Example class 3m (BBO, ..)

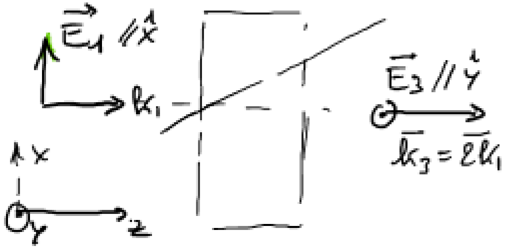
$$d_{il} = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{31} & \boxed{-d_{22}} \\ \boxed{-d_{22}} & \boxed{d_{22}} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix}$$

- Typically the geometry is very well defined! The polarization of the interacting waves is linear, and well known relative to the crystal orientation (phase matching): the net effect is summarized in an effective nonlinear coefficient (with a smart choice of x,y,z ..)

POLARIZATION ONLY ALONG CERTAIN DIRECTIONS

$$\begin{bmatrix} P_x(2\omega) \\ P_y(2\omega) \\ P_z(2\omega) \end{bmatrix} = 2 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \begin{bmatrix} E_x(\omega)^2 \\ \cancel{E_y(\omega)^2} \\ \cancel{E_z(\omega)^2} \\ \cancel{2E_y(\omega)E_z(\omega)} \\ \cancel{2E_x(\omega)E_z(\omega)} \\ \cancel{2E_x(\omega)E_y(\omega)} \end{bmatrix}$$

ONLY SOME COMPONENT CAN ACHIEVE PHASE MATCHING !



$$P(\omega_3) = 4d_{\text{eff}} E(\omega_1) E(\omega_2)$$

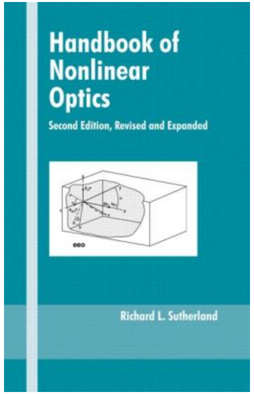
$$d_{\text{eff}} = d_{31} \sin\theta - d_{22} \cos\theta \sin 3\phi$$

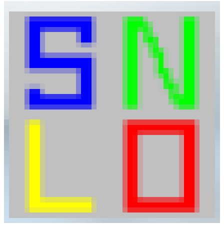
Properties of nonlinear optical materials, resources :

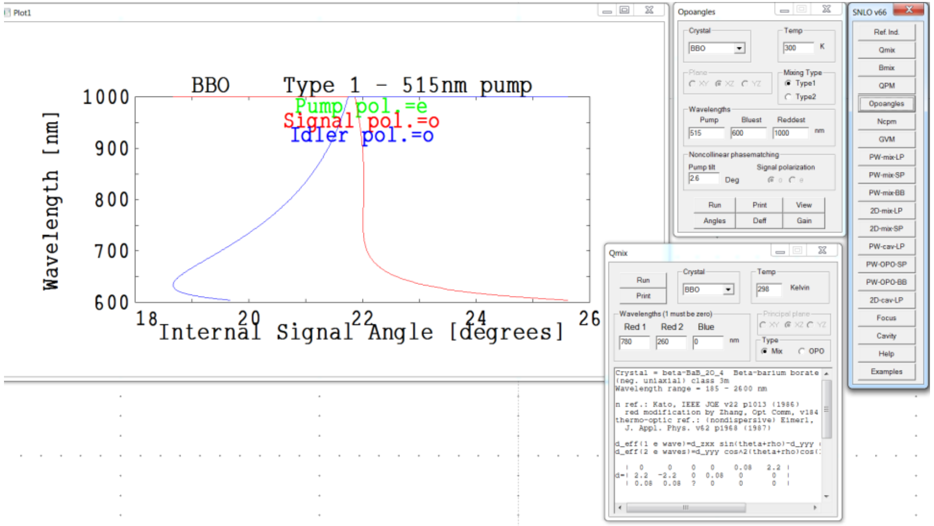
Nonlinear optics is nowadays well-established

Sutherland - Handbook of nonlinear optics

Free software: SNLO
http://www.as-photonics.com/products/snlo





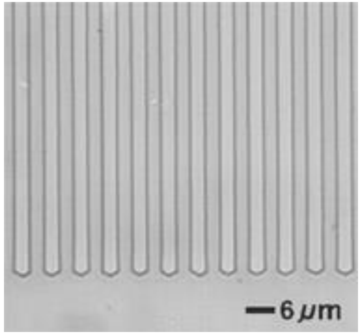


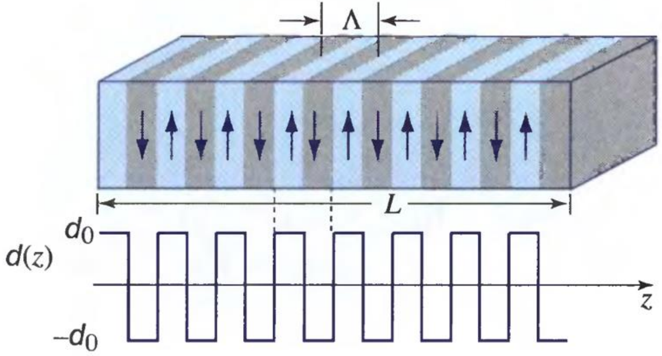
Temperature phase matching:

AKA Noncritical phase matching (or *90° phase matching*)

The crystal is held in oven at well defined T: the refractive index change with T , n(T) is different for different crystal axis, and phase matching can be achieved.

Quasi-phase matching:





Example: SEM picture of a periodically poled lithium niobate (PPLN) cystal showing the periodically inverted non-linear optical coefficient