Summary

Amplifiers of ultrashort Pulses
1. Short pulse propagation

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

Advanced Radiation Sources - PHSY761

Lecture 06

15 October 2024

Michele Puppin

RECAP: TIME vs FREQUENCY DOMAIN

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

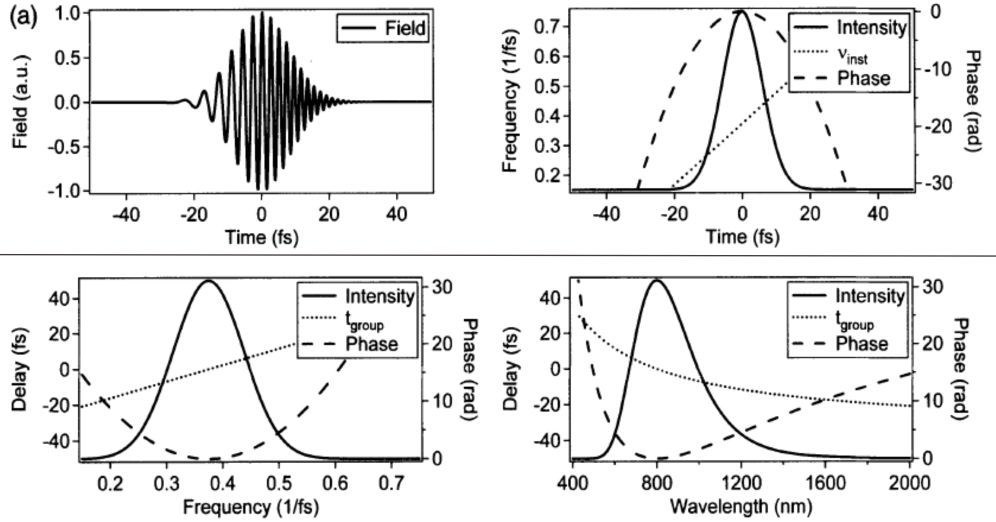
$$\tilde{\mathcal{E}}(\omega) = \int_{-\infty}^{\infty} \mathcal{E}(t)\exp(-\mathrm{i}\omega t)\,\mathrm{d}t$$

FOURIER TRANSFORM

Spectral description:

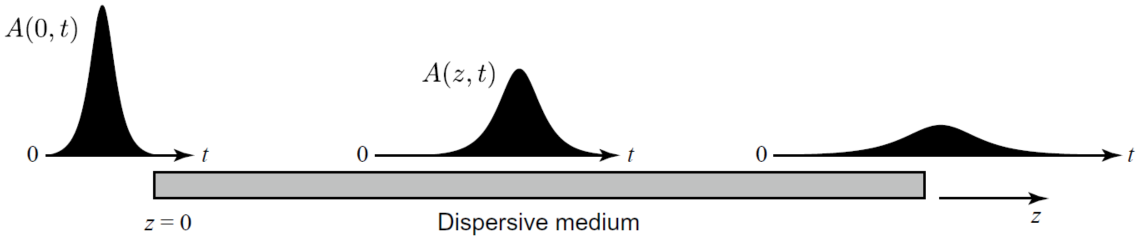
$$\tilde{\mathcal{E}}(\omega) = \sqrt{S(\omega)}\exp[-\mathrm{i}\varphi(\omega)]$$

COMPLEX AMPLITUDE



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

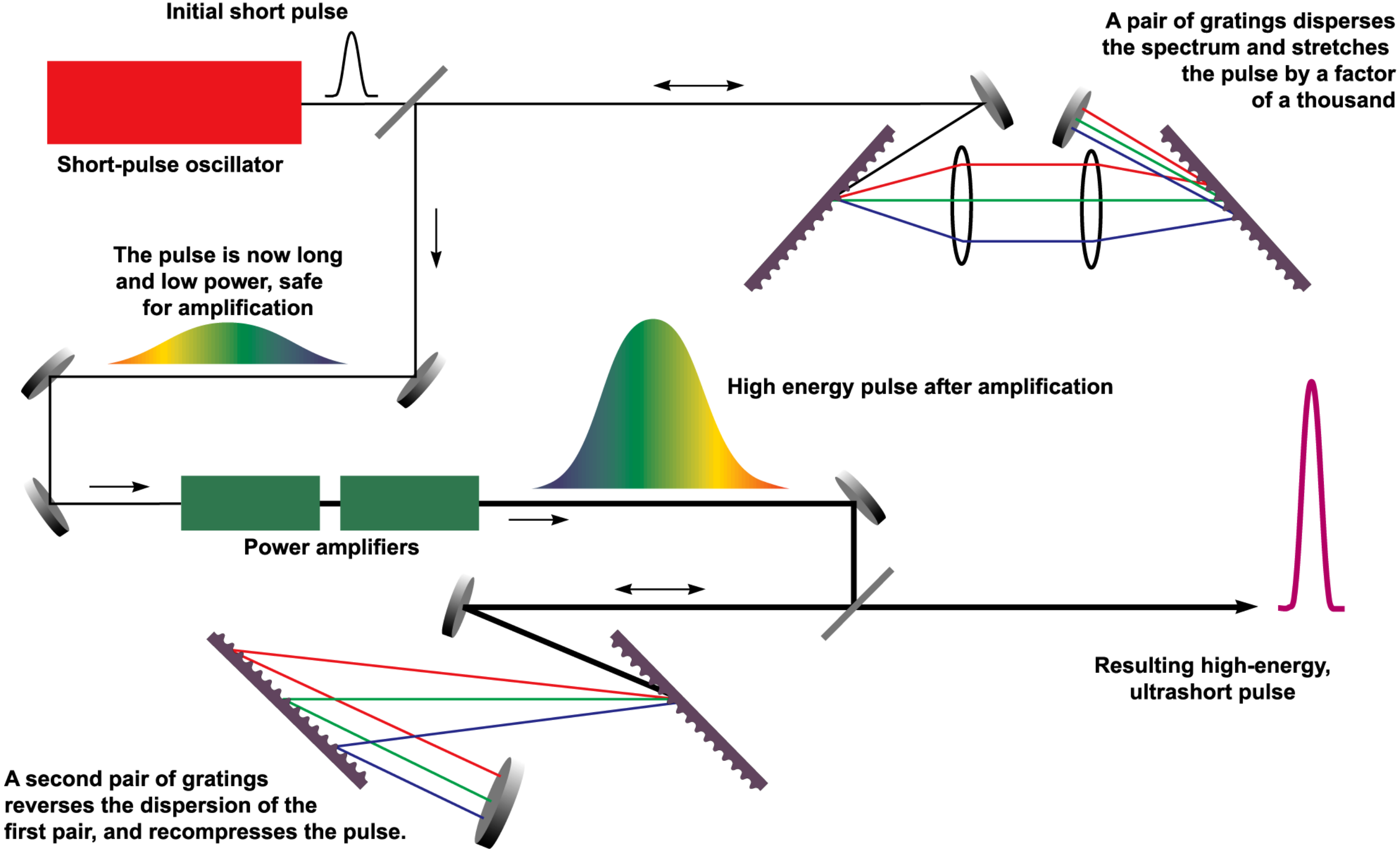
RECAP: Linear pulse propagation



$$\tilde{A}(z, \Delta\omega) = \tilde{A}(0, \Delta\omega)\, e^{-i\Delta k_n z}$$

$$k_n(\omega) \approx k_n(\omega_0) + k_n' \Delta\omega + \frac{1}{2} k_n'' \Delta\omega^2 + \ldots$$

RECAP 2: CPA


Initial short pulse
Short-pulse oscillator
A pair of gratings disperses the spectrum and stretches the pulse by a factor of a thousand
The pulse is now long and low power, safe for amplification
High energy pulse after amplification
Power amplifiers
Resulting high-energy, ultrashort pulse
A second pair of gratings reverses the dispersion of the first pair, and recompresses the pulse.

GRATING COMPRESSOR/STRETCHER

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- Limited in producing NEGATIVE GDD, we need to find an instrument with inverted (and matched GDD) ->

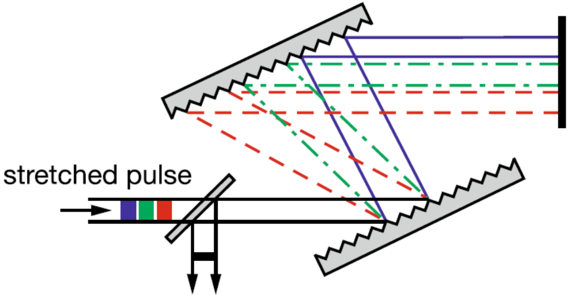
$$\frac{\mathrm{d}^2\phi}{\mathrm{d}\omega^2} = -\frac{\lambda^3 L_g}{\pi c^2 \Lambda^2}\left[1-\left(\frac{\lambda}{\Lambda}-\sin\theta_i\right)^2\right]^{-3/2}$$

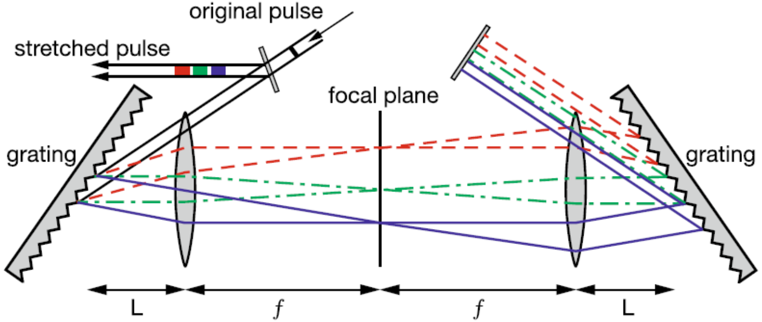
$$\frac{\mathrm{d}^2\phi}{\mathrm{d}\omega^2} = -\frac{m^2\lambda^3 M^2 L_{\mathrm{eff}}}{2\pi c^2 \Lambda^2}\left[1-\left(-m\frac{\lambda}{\Lambda}-\sin\theta_i\right)^2\right]^{-3/2}$$

$L < f$ positive GDD

$L > f$ negative GDD

$L = f$ => 4f system





MAXWELL EQUATIONS in matter:

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$$
\begin{cases}
\vec{\nabla}\times\vec{H} = \vec{J}_f + \dfrac{\partial \vec{D}}{\partial t} \\
\vec{\nabla}\times\vec{E} = -\dfrac{\partial \vec{B}}{\partial t} \\
\vec{\nabla}\cdot\vec{D} = \rho_f \\
\nabla\cdot B = 0
\end{cases}
$$

$$\vec{H} = \frac{\vec{B}}{\mu_0} + \vec{M}$$

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

MATERIAL EQUATIONS

LINEARITY

$$\vec{P} = \varepsilon_0 \chi \vec{E} \Rightarrow \vec{D} = \varepsilon\varepsilon_0 \vec{E}$$

$$\chi = \varepsilon - 1$$

$$\vec{M} = \frac{1}{\mu_0}\chi_m \vec{B} \qquad \vec{H} = \frac{\vec{B}}{\mu\mu_0}$$

Susceptibilities

$$\vec{J}_f = 0 \quad \rho_f = 0$$

$$\vec{M} = 0 \longrightarrow \mu \sim 1$$

- homogenous dielectric material
- non magnetic

WAVE EQUATION:

$$\vec{\nabla}\times(\vec{\nabla}\times\vec{E})$$

$$= \nabla^2\vec{E} - \vec{\nabla}(\vec{\nabla}\cdot\vec{E})$$

LINEAR + ISOTROPIC

$$= -\frac{\partial}{\partial t}\left[\vec{\nabla}\times\vec{B}\right]$$

$$= -\frac{\partial}{\partial t}\left[\mu_0 \frac{\partial \vec{D}}{\partial t}\right]$$

$$= -\frac{\partial^2}{\partial t^2}\left[\mu_0\varepsilon_0 \vec{E} + \mu_0 \vec{P}\right]$$

WAVE EQ.

$$\nabla^2 \vec{E} - \frac{1}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2} \longrightarrow$$

- MAXWELL EQ.
- HOMOGENEOUS DIELECTRIC MEDIUM

WAVE EQUATION

$$c = \frac{1}{\sqrt{\mu_0 \varepsilon_0}}$$ LIGHT SPEED IN VACUUM

↓ 1d PROP. ALONG z

LINEAR, DISPERSIONLESS MEDIUM

$$\vec{P}(t) = \varepsilon_0 \chi \vec{E}(t)$$

$$\frac{\partial^2 E(z,t)}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \chi \varepsilon_0 \frac{\partial^2 E}{\partial t^2}$$

~

$$\frac{\partial^2 E}{\partial z^2} - \mu_0 \varepsilon_0 \frac{\partial^2 E}{\partial t^2} - \mu_0 \varepsilon_0 \chi \frac{\partial^2 E}{\partial t^2} = 0$$

$P \rightarrow \varepsilon_0 \chi E$

PHASE VELOCITY

$$c^2 = \frac{1}{\mu_0 \varepsilon_0} \qquad \frac{\chi + 1}{\mu_0 \varepsilon_0} \rightarrow \frac{n^2}{c^2} = \frac{1}{v_p^2}$$

$$n = \sqrt{\varepsilon} = \sqrt{1 + \chi} \qquad \frac{\partial^2 E}{\partial z^2} - \frac{1}{v_p^2} \frac{\partial^2 E}{\partial t^2} = 0$$

SOLUTION → PLANE WAVE WITH PHASE VELOCITY $v_p = \frac{c}{n}$

$$E = E^{(+)} e^{i(\omega t - kz)} + E^{(-)} e^{i(\omega t + kz)}$$

LINEAR RESPONSE REGIME:

$$\vec{P}(t) = \varepsilon_0 \chi \vec{E}(t)$$

"IMPULSE RESPONSE FUNCTION"

$$\vec{P}(t) = \varepsilon_0 \int_{-\infty}^{t} R(t-t')\, E(t')\, dt'$$

Account for non-instantaneous response, phase shift ...

TIME DOMAIN DESCRIPTION

$$= \varepsilon_0 \underbrace{R(t) * E(t)}_{\text{CONVOLUTION}}$$

FREQUENCY DOMAIN SIMPLER!

$$\vec{P}(\omega) = \underbrace{\varepsilon_0 \chi(\omega) \vec{E}(\omega)}_{\text{PRODUCT}}$$

Frequency-dependent susceptibility

$$\chi(\omega) = \mathcal{F}[R(t)]$$

IF χ DEPENDS ON ω $(n(\omega)) \rightarrow$ CONVENIENT TO WORK IN THE ω DOMAIN

$$\tilde{E}(\omega) = \mathcal{F}[E(t)] = \int_{-\infty}^{+\infty} E(t)\, e^{-i\omega t}\, d\omega$$

$$E(t) = \mathcal{F}^{-1}[\tilde{E}(\omega)] = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \tilde{E}(\omega)\, e^{i\omega t}\, d\omega$$

$$\frac{\partial E(t,z)}{\partial t} = \frac{\partial}{\partial t}\mathcal{F}^{-1}[E(\omega,z)] = \mathcal{F}^{-1}[-i\omega E(\omega)]$$

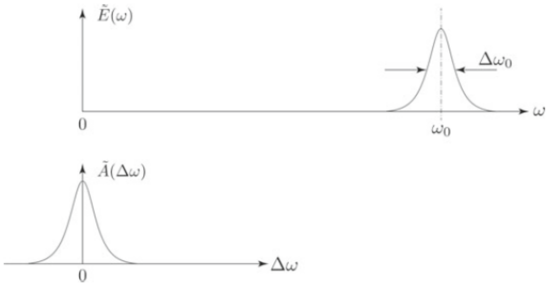
$\downarrow$

HELMOLZ EQUATION

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

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

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



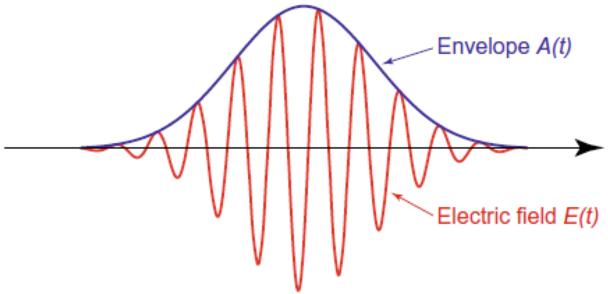
$$E(t) = \frac{1}{2\pi}\int \tilde{E}(\omega)\mathrm{e}^{\mathrm{i}\omega t}\mathrm{d}\omega$$

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

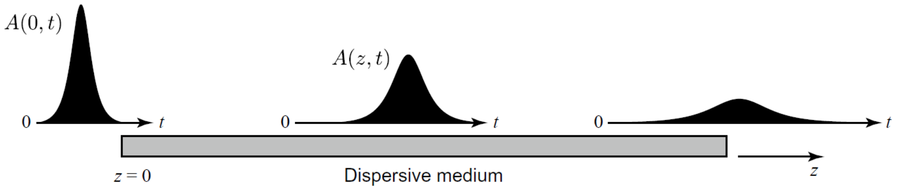
Frequency-shifted spectrum

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

Temporal envelope



Ultrashort pulse propagation in a linear dispersive medium



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WAVE EQ. IN THE FREQ. DOMAIN

$$\frac{\partial^2 \tilde{E}(z,\omega)}{\partial z^2} + k_n^2(\omega)\,\tilde{E}(z,\omega) = 0$$

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

$$\frac{\partial^2}{\partial z^2}\tilde{A}(z,\Delta\omega) - 2 i k_n(\omega_0)\frac{\partial}{\partial z}\tilde{A}(z,\Delta\omega) - [k_n(\omega_0)]^2\tilde{A}(z,\Delta\omega) + [k_n(\omega_0+\Delta\omega)]^2\tilde{A}(z,\omega) = 0$$

(1) SVEA

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

(2) $k_n(\omega_0+\Delta\omega) \approx k_n(\omega_0) + \Delta k_n$

$\Delta k_n \ll k_n$

$$\frac{\partial}{\partial z}\tilde{A}\left(z,\Delta\omega\right)+\mathrm{i}\Delta k_n\tilde{A}\left(z,\Delta\omega\right)=0$$

$$\tilde{A}\left(z,\Delta\omega\right)=\tilde{A}\left(0,\Delta\omega\right)\mathrm{e}^{-\mathrm{i}\Delta k_n z}$$

$S \propto |E(\omega)|^2 \rightarrow$ SPECTRUM UNCHANGED

TAYLOR EXPANSION OF THE SPECTRAL PHASE

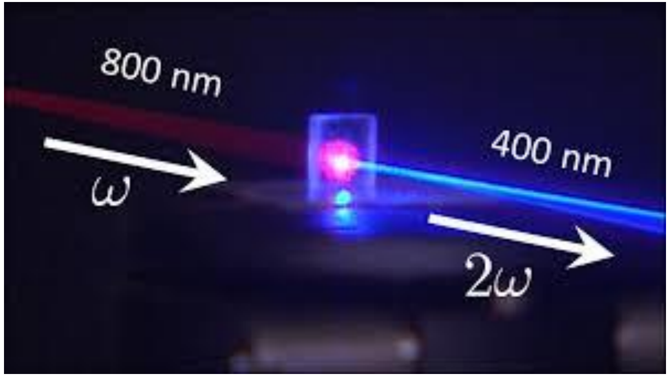
$$\varphi(\omega)=\varphi_0+(\omega-\omega_0)\,\varphi_1+(\omega-\omega_0)^2\,\varphi_2/2+\cdots$$

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

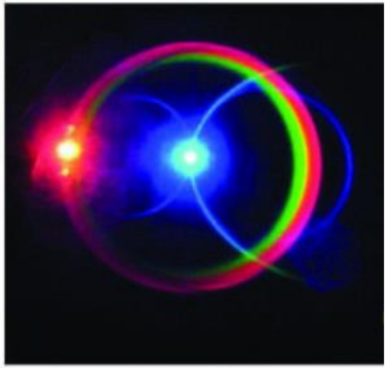
PERTURBATIVE NONLINEAR OPTICS

CPA of femtosecond pulse:

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



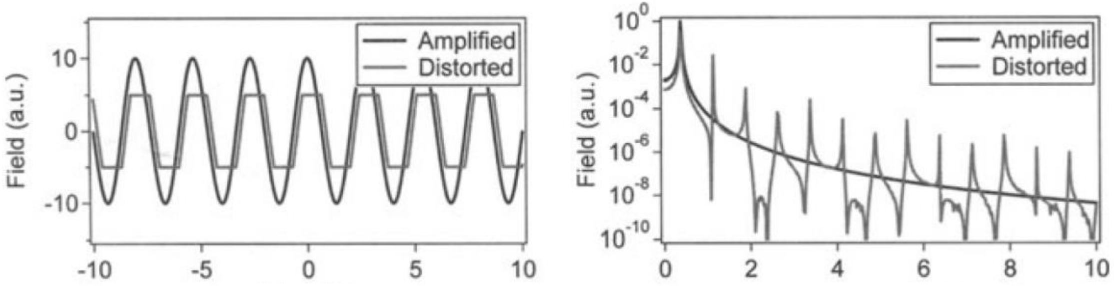
Second harmonic generation



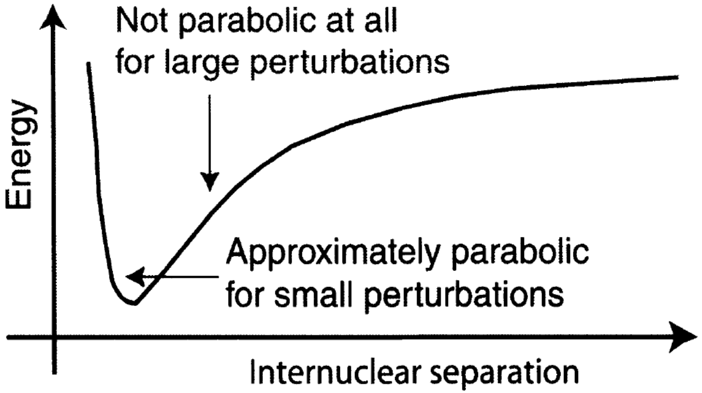
Parametric amplification

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?

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

At which E the linear and quadratic term become comparable?

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

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

$\chi^{(1)} \to \mathcal{O}(1)$

Order-magnitude electric field acting on a valence electron:

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

$e \to e^-$ charge; $a_0 \to$ 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

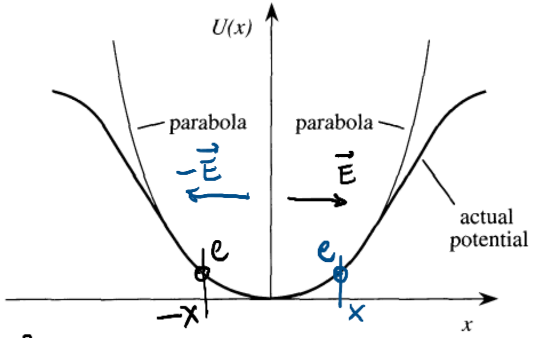
Electric field of solar radiation on the earth surface:

$E_{sun} \approx 3$ 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:



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

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

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

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

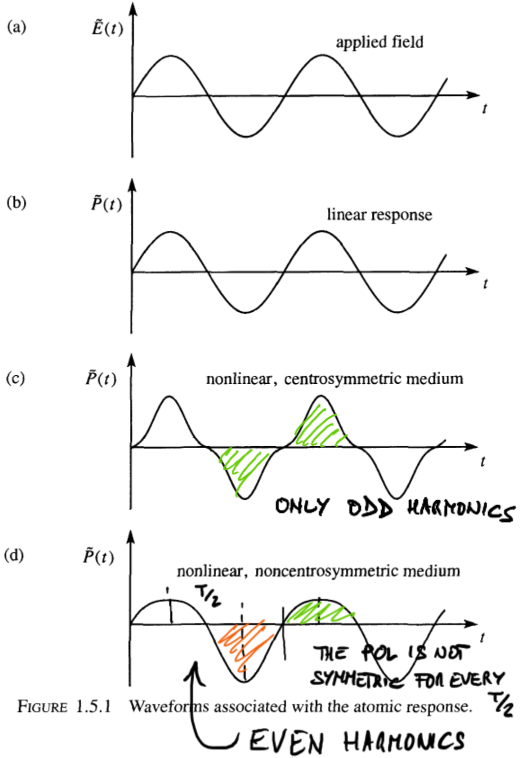
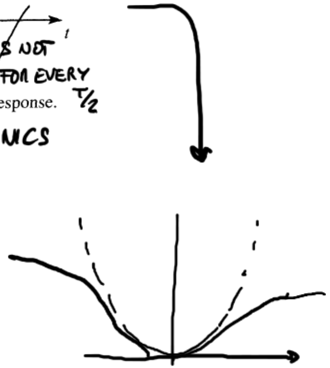


FIGURE 1.5.1 Waveforms associated with the atomic response.



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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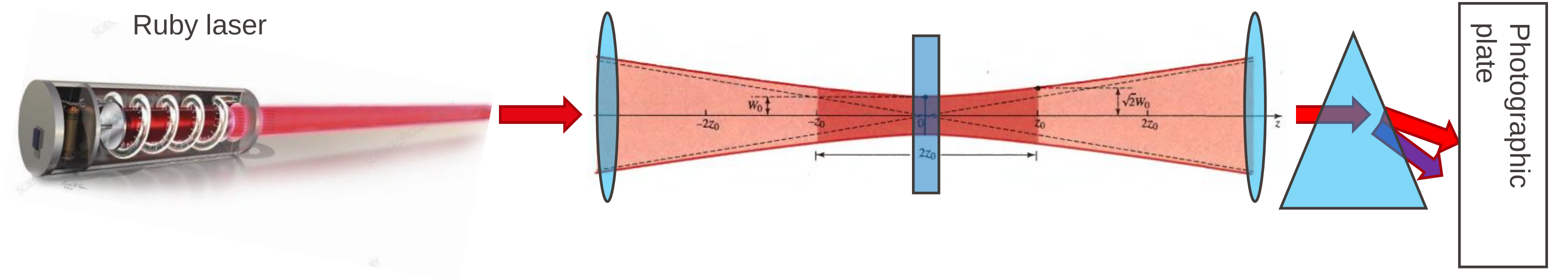
The first nonlinear optics experiment with a ruby laser:



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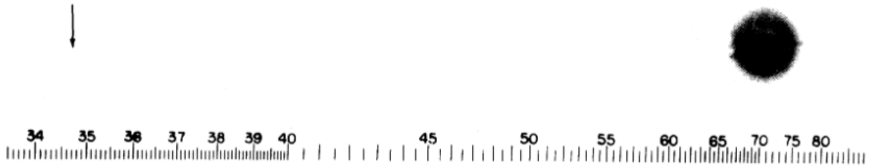


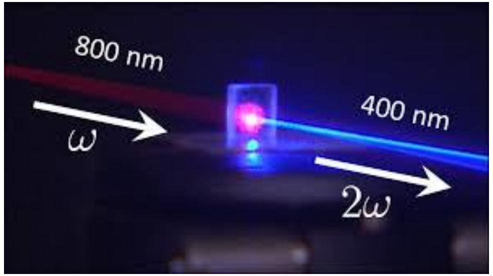
FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

Can you see the spot?

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

Second harmonic generation now:

- 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 ($L_{coh} > L_{crystal}$)

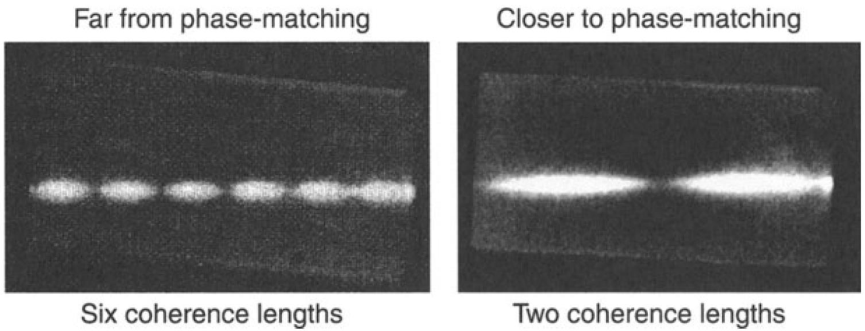


PHASE MATCHING CONDITION

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

k_{FH}: FUNDAMENTAL BEAM WAVEVECTOR

k_{SH}: SH WAVEVECTOR



- 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:

$$\tilde{P}^{(2)}(t) = \chi^{(2)} \tilde{E}(t)^2$$

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

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

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

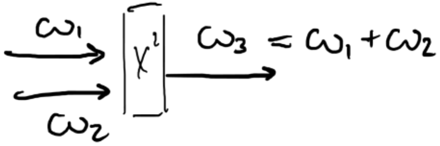
Difference Frequency Generation

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

Optical rectification

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

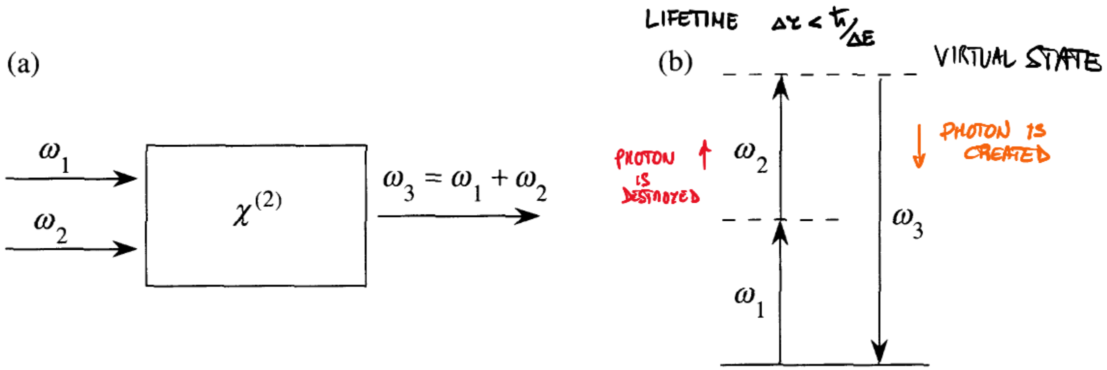
Three wave mixing : in $\chi^{(2)}$ processes three waves interact thanks to the nonlinear susceptibility



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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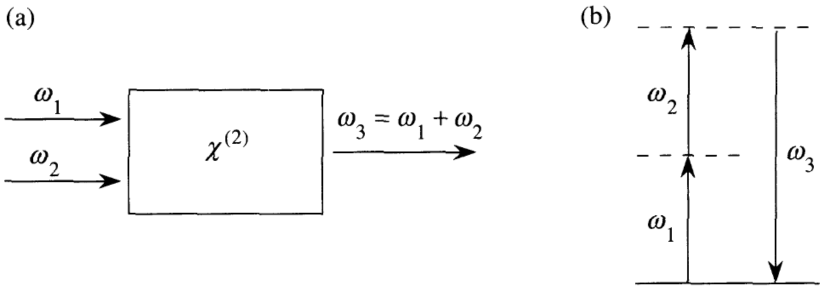
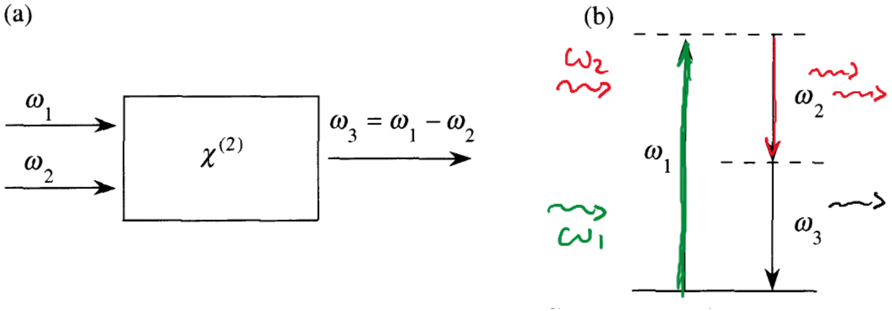
SFG and DFG

- Sum Frequency generation (SFG):



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

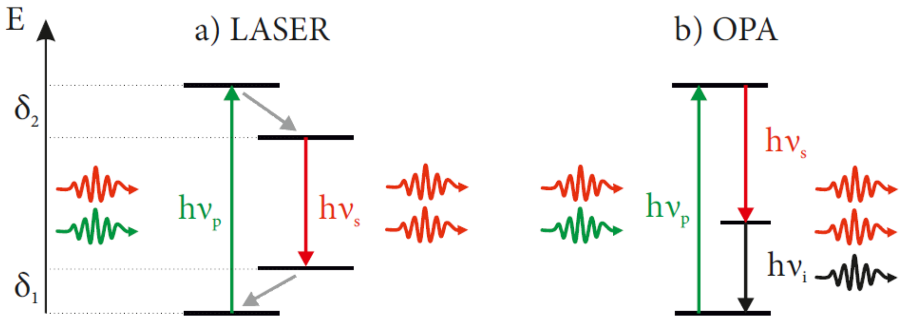
- Difference frequency generation (DFG):



- 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



$h\nu_p = h\nu_s + \delta_{QD}$

$\delta_{QD} = \delta_1 + \delta_2$

$h\nu_p = h\nu_s + h\nu_i$

Improved Average Power scalability

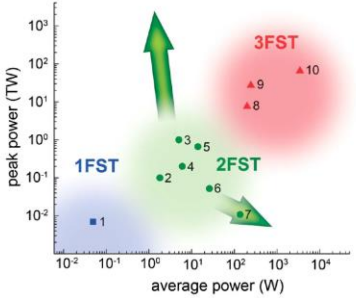
Bandwidth

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:

- Saturable absorption/amplification:

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

- Two-photon absorption:

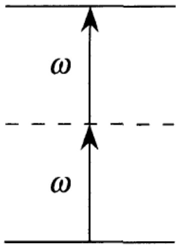
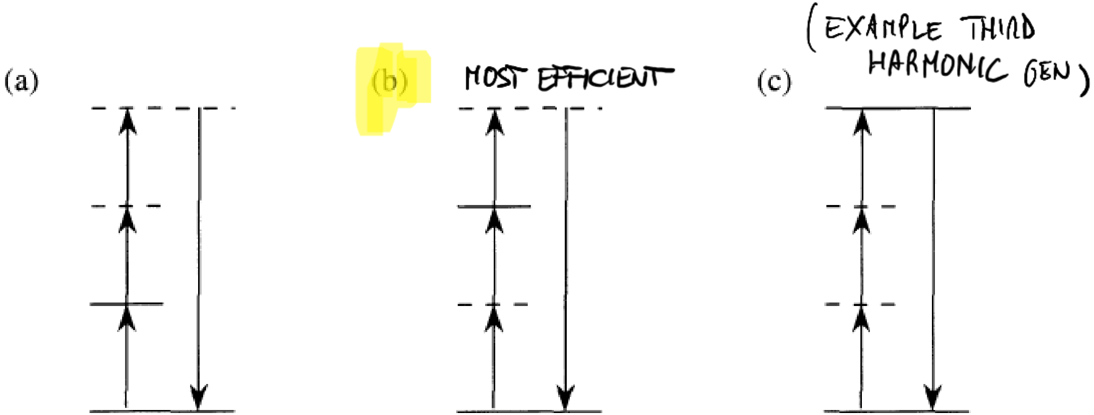


FIGURE 1.2.10 Two-photon absorption.

- 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

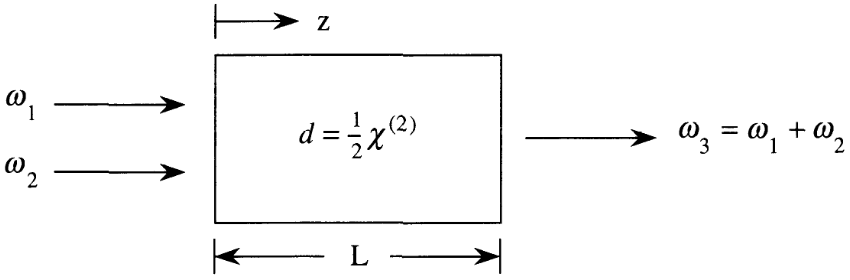


FIGURE 2.2.1 Sum-frequency generation.

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

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

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)

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

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

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

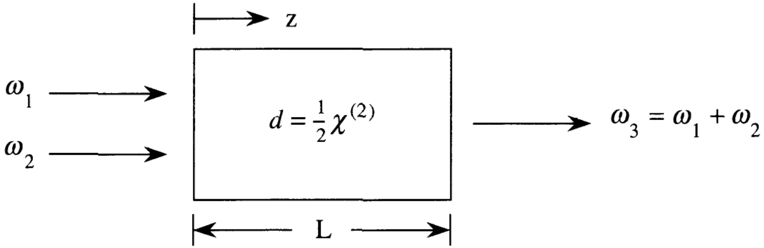


FIGURE 2.2.1 Sum-frequency generation.

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

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

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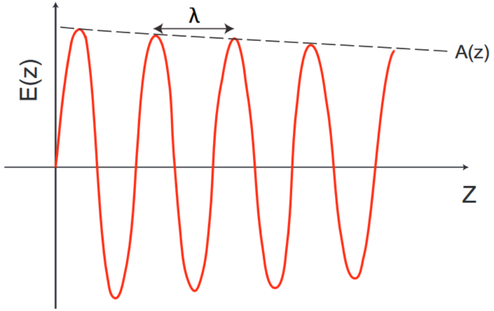
- Slowly Varying Amplitude Approximation:

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..



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

- The second derivative term is dropped

$$\frac{dA_3}{dz} = \frac{8\pi i d_{\text{eff}} \omega_3^2}{k_3 c^2} A_1 A_2 e^{i\Delta k z}$$

$$\frac{dA_1}{dz} = \frac{8\pi i d_{\text{eff}} \omega_1^2}{k_1 c^2} A_3 A_2^* e^{-i\Delta k z}$$

$$\frac{dA_2}{dz} = \frac{8\pi i d_{\text{eff}} \omega_2^2}{k_2 c^2} A_3 A_1^* e^{-i\Delta k z}$$

SIGN DIFFERENCE!

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

PHASE MISMATCH

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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 !

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

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

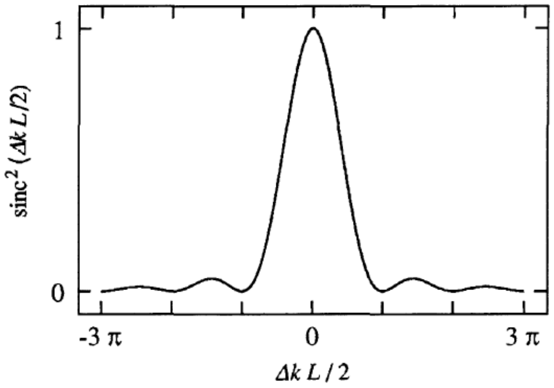
- Solution valid until the beam does not grow significantly, afterwards the approximation fails and the coupling needs to be accounted: back-conversion from 3 to 1 and 2 can be observed!

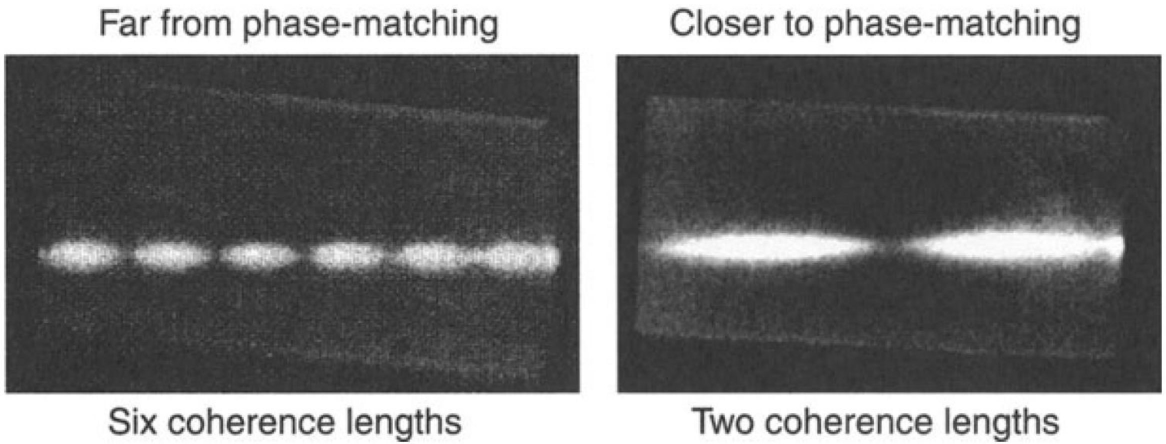
Phase matching

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

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

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





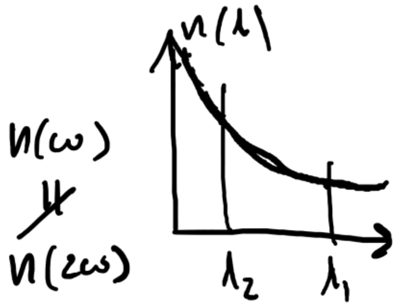
Perfect phase-matching seems a very special condition:

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

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

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

NORMAL DISPERSION



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

Refractive index wavelength dependence

Critical phase matching in uniaxial crystals

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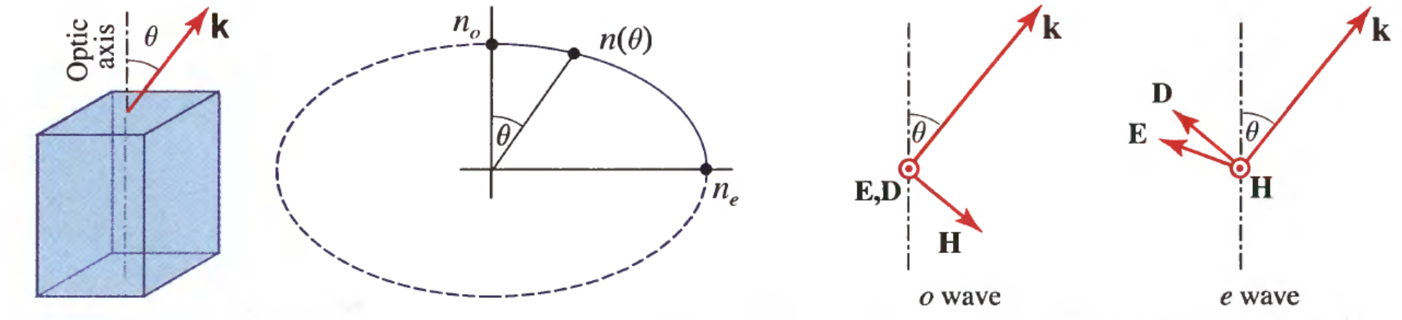

- The refractive index of waves is determined entirely by the angle between **k** and the optical axis.
- ordinary-wave **E** ortogonal to the optical axis.
- Positive uniaxial (n_o> n_e) , negative uniaxial (n_e> n_o)

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

- 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

i, j, k CARTESIAN COORDINATES — GEN. FREQ. — DRIVING FREQ.

$$P_i(\omega_n+\omega_m)=\sum_{jk}\sum_{(nm)}\chi^{(2)}_{ijk}(\omega_n+\omega_m,\omega_n,\omega_m)E_j(\omega_n)E_k(\omega_m)$$

FREQ. OF POLARIZ. — SUMMATION PERFORMED FOR FIXED $\omega_n+\omega_m$

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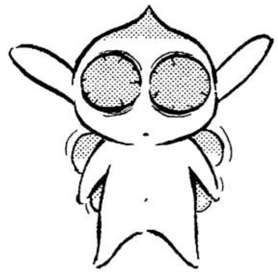
- To include all possible polarizations generated by three interacting waves 6 tensor (x 2 counting the negative ω) have to be determined: each one has 3^3=27 components:

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



- 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)

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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

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

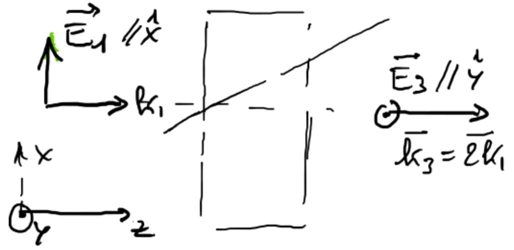
$$d_{il} = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix}$$

- 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

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

ONLY SOME COMPONENT CAN ACHIEVE PHASE MATCHING !



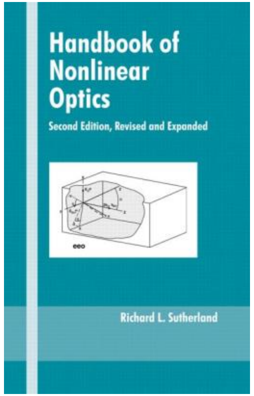
$$P(\omega_3) = 4d_{\text{eff}} E(\omega_1) E(\omega_2)$$

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

Properties of nonlinear optical materials, resources :

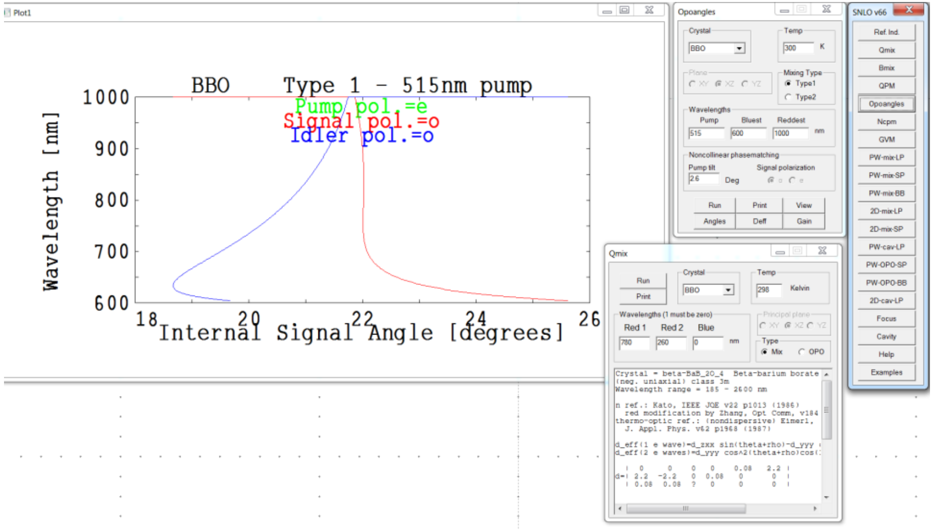
Nonlinear optics is nowadays well-established: you don't need to calculate everything yourself.

Sutherland - Handbook of nonlinear optics



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

SNLO

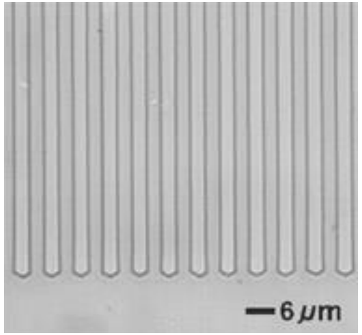


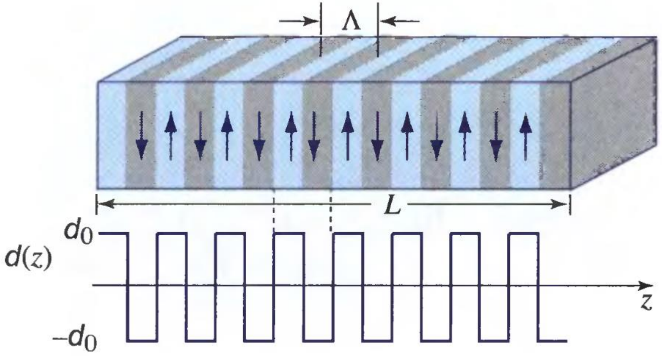
Temperature phase matching:

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