# Modules of the 2024 Course

| **Topics covered** | **No** | **Lecture/Date** |
|---|---|---|
| Introductory presentation; Basic of laser operation I: dispersion theory, atoms | 1 | 11. 09. 2024 |
| Basic of laser operation II: dispersion theory, atoms | 2 | 18. 09. 2024 |
| Laser systems I: 3 and 4 level lasers, gas lasers, solid state lasers, applications | 3 | 25. 09. 2024 |
| Laser systems II: semi-conductor lasers, external cavity lasers, applications | 4 | 02. 10. 2024 |
| Noise characteristics of lasers: linewidth, coherence, phase and amplitude noise, OSA (1) | 5 | 09. 10. 2024 |
| Noise characteristics of lasers: linewidth, coherence, phase and amplitude noise, OSA (2) | 6 | 16. 10. 2024 |
| Optical detection | 7 | 30. 10. 2024 |
| Optical fibers: light propagation in fibers, specialty fibers and dispersion (GVD) | 8 | 06. 11. 2024 |
| Ultrafast lasers I.: Passive mode locking and ultrafast lasers | 9 | 13. 11. 2024 |
| Ultrafast lasers II: mode locking, optical frequency combs / frequency metrology | 10 | 20. 11. 2024 |
| Ultrafast lasers III: pulse characterization, applications | 11 | 27. 11. 2024 |
| Nonlinear frequency conversion I: theory, frequency doubling, applications | 12 | 04. 12. 2024 |
| Nonlinear frequency conversion II: optical parametric amplification (OPA) | 13 | 11. 12. 2024 |
| Laboratory visits (lasers demo) | 14 | 20. 12. 2024 |

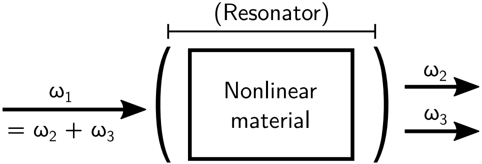
# What is nonlinear frequency conversion?

The conversion of light to other frequencies (wavelengths), using optical nonlinearities.

Examples:

Sum-frequency generation

Second-harmonic generation



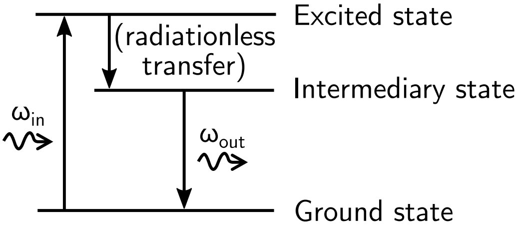
Optical parametric oscillation

# What is nonlinear frequency conversion?

Differences:

### Stimulated emission

Example: Three-level system



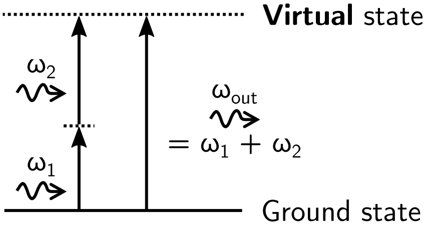
Gain occurs by first absorbing the input, and then re-emitting to the output frequency.

- *Non-parametric:* The material transitions to various quantum states.
- Photon energy is not necessarily conserved:

$$\hbar\omega_{out} \neq \hbar\omega_{in}$$

### Non-linear frequency conversion

Example: Sum-frequency generation



Gain occurs by “instantaneous” transfer of energy from input to output frequency.

- *Parametric:* The quantum state of the material remains unchanged.
- Photon energy is conserved:

$$\hbar\omega_{out} = \hbar\omega_1 + \hbar\omega_2$$

# Extra information: The term “parametric”

**Avoid confusion:**

“Nonlinear frequency conversion is a parametric process,”
however, “nonlinear” and “parametric” do not always go together.

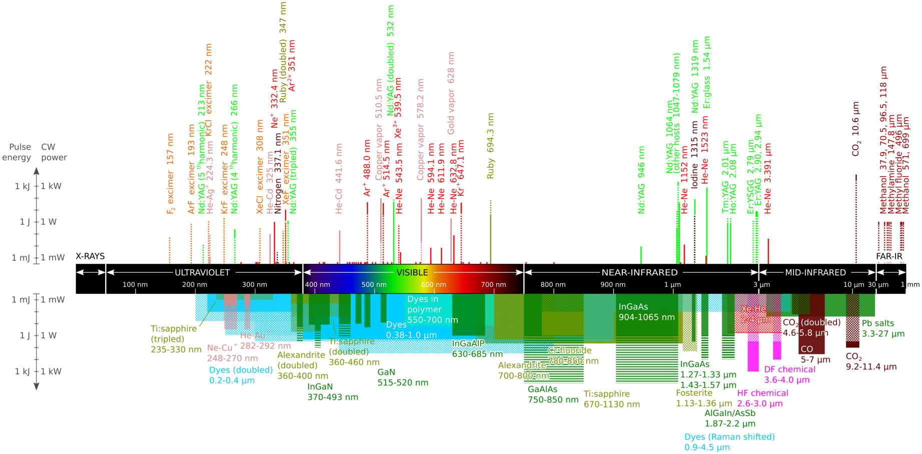
Examples of other processes:

- Nonlinear and non-parametric: saturable absorption, 2-photon absorption
- Linear and parametric: refraction, Rayleigh scattering

Also, “parametric” here is not synonymous for “tunable” (even if many nonlinear processes are in fact tunable!).

# Why is nonlinear frequency conversion used?

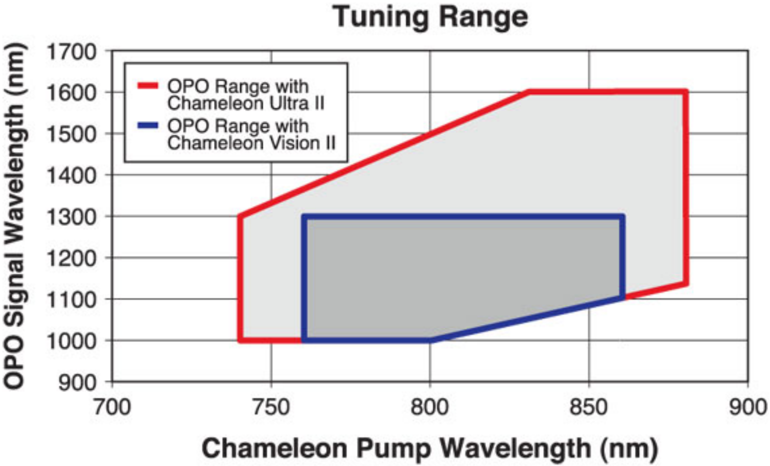
There isn't a gain medium for every wavelength.



Nonlinear conversion allows to create wavelengths that are inaccessible with known gain media.

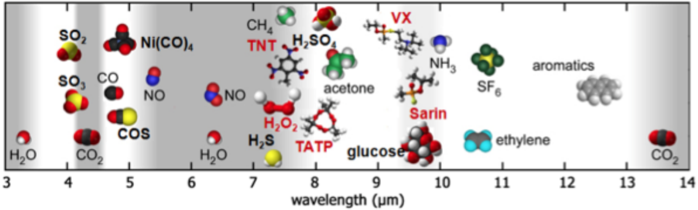
# Why is nonlinear frequency conversion used?

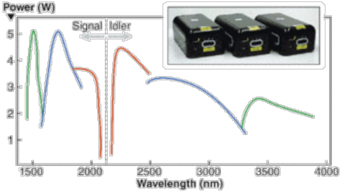
With one pump frequency, it is possible to generate a range of output frequencies:



# Why is nonlinear frequency conversion used?

Example of application: **Molecular spectroscopy**
(to probe molecular resonances in the infrared)





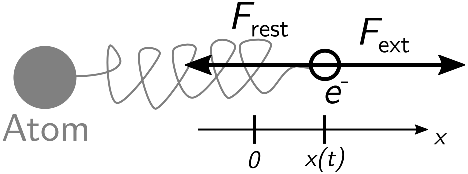
# Electron oscillator model

**Model:** Assume an electron subjected to:

- A force due to an external electric field: $F_{\text{ext}} = eE(t)$
- A restoring force, function the electron's position: $F_{\text{r}} = F(x)$

(For simplicity, here we neglect damping and the electron's inertia.)

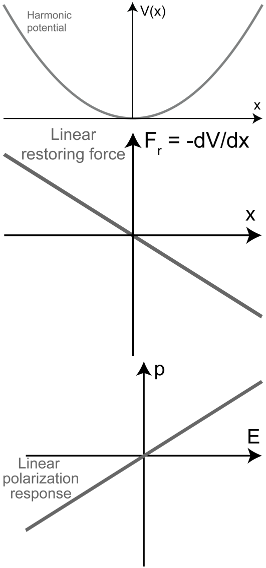
→ The balance of the forces determines the electron's position:



→ The electron's displacement gives rise to polarization:

$$p(t) = -Nex(t)$$

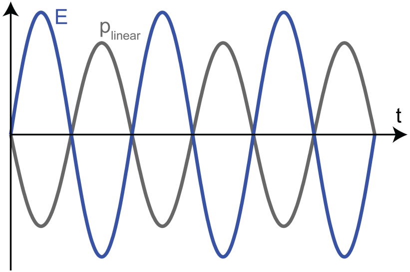
# Electron oscillator model



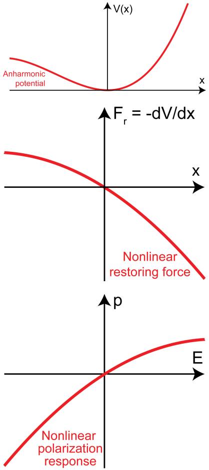
At small field intensities, one usually approximates the potential energy of this electron as a parabola (harmonic potential).

→ This leads to a linear restoring force, and a linear polarization response.

Response in time:



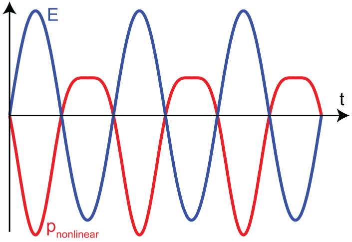
# Electron oscillator model



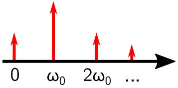
Real potentials are not perfectly parabolic (**anharmonic potentials**).

→ This leads to a nonlinear restoring force, and a **nonlinear polarization response**.

Response in time:



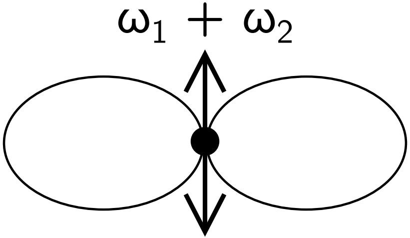
→ Spectrum of $p(t)$:



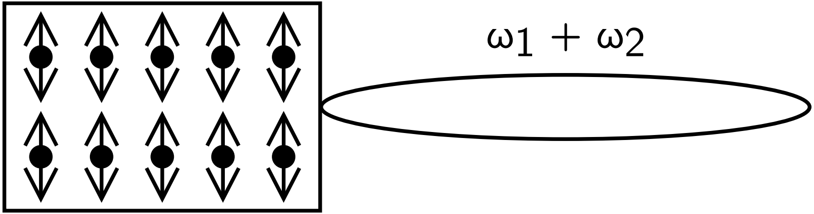
(New harmonics are created)

# Electron oscillator model

Each oscillating electron in the material radiates these new harmonics in a typical dipole pattern:



In nonlinear frequency conversion, we are not interested in the radiation of only one single dipole.
The objective is to superpose all dipoles of the material coherently to create a strong directional beam:



# Wave equation

We will now study frequency conversion using the wave equation:

$$\frac{\delta^2 E}{\delta z^2} - \frac{1}{c^2}\frac{\delta^2 E}{\delta t^2} = \frac{1}{\varepsilon_0 c^2}\frac{\delta^2 P}{\delta t^2}$$

Derivation of the wave equation: see Boyd, “Nonlinear optics”, Chapter 2.

Objectives:

- Understand the nonlinear energy transfer (*coupled wave equation*)
- Derive the condition for efficient conversion (*phase matching condition*)

# Wave equation

$$\frac{\delta^2 E}{\delta z^2} - \frac{1}{c^2}\frac{\delta^2 E}{\delta t^2} = \frac{1}{\varepsilon_0 c^2}\frac{\delta^2 P}{\delta t^2}$$

Expand the components of polarization:

$$P = \underbrace{\varepsilon_0 \chi^{(1)} E}_{P_L \ \text{(linear)}} + \underbrace{\varepsilon_0 \chi^{(2)} E^2 + \varepsilon_0 \chi^{(3)} E^3 + \dots}_{P_{NL} \ \text{(nonlinear)}}$$

$$\frac{\delta^2 E}{\delta z^2} - \frac{1}{c^2}\frac{\delta^2 E}{\delta t^2} = \frac{1}{c^2}\chi^{(1)}\frac{\delta^2 E}{\delta t^2} + \frac{1}{\varepsilon_0 c^2}\frac{\delta^2 P_{NL}}{\delta t^2}$$

Introduce the linear refractive index: $n^2 = 1 + \chi^{(1)}$

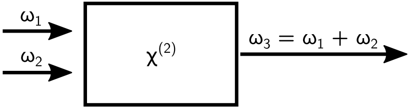
(Note that $n$ and $\chi$ are wavelength-dependent. This will be important when we discuss phase-matching.)

$$\frac{\delta^2 E}{\delta z^2} - \frac{n^2}{c^2}\frac{\delta^2 E}{\delta t^2} = \underbrace{\frac{1}{\varepsilon_0 c^2}\frac{\delta^2 P_{NL}}{\delta t^2}}_{\text{Nonlinear source term}}$$

To continue, we need to define $E$ more precisely.

# Sum Frequency Generation

Let us define the various electric fields in the case of **sum frequency generation (SFG)**.



Note: For SFG, we focus on $\chi^{(2)}$, and assume $\chi^{(3)}, \chi^{(4)}, \cdots = 0$

$$\left\{\begin{array}{l}
E_1(z,t) = \overbrace{A_1(z)}^{\text{Varying amplitude factor, used to describe nonlinear interactions}} e^{ik_1z}e^{-i\omega_1t} \text{ (+ complex conjugate)} \\
E_2(z,t) = A_2(z)e^{ik_2z}e^{-i\omega_2t} \text{ (+ complex conjugate)} \\
E_3(z,t) = A_3(z)\underbrace{e^{ik_3z}e^{-i\omega_3t}}_{\text{Solution we expect in the linear case}} \text{ (+ complex conjugate)}
\end{array}\right.$$

These can now be plugged into the wave equation:

$$E = E_1 + E_2 + E_3 \text{ (+ complex conjugates)}$$

$$P_{NL} = \varepsilon_0\chi^{(2)}E^2$$

$$\frac{\delta^2 E}{\delta z^2} - \frac{n^2}{c^2}\frac{\delta^2 E}{\delta t^2} = \frac{1}{\varepsilon_0 c^2}\frac{\delta^2}{\delta t^2}\left(\varepsilon_0\chi^{(2)}E^2\right)$$

# Sum Frequency Generation

This creates many terms. Fortunately, we see that the equation must hold for each frequency component separately.

**More explanation:** An equation of the form $Ae^{-i\omega_a t} + Be^{-i\omega_b t} + \cdots = 0$ can only be valid for all values of $t$ if all the coefficients are all zero, i.e. $A = 0$, $B = 0$, $\ldots$ We have such an equation here: the wave equation must be valid for every $t$, and each field we plug into it has an $e^{-i\omega_i t}$ dependence. This dependence is preserved by the operators $\frac{\delta^1}{\delta z^2}$ and $\frac{\delta^2}{\delta t}$. Therefore, we can just consider all terms with the same frequency, and set their sum equal to $0$. The terms we are interested in here are the ones with frequency $e^{-i\omega_3 t}$ or equivalently $e^{-i(\omega_1+\omega_2)t}$.

We consider only the terms with a factor $e^{-i\omega_3 t}$ or $e^{-i(\omega_1+\omega_2)t}$:

$$\frac{\delta^2 E_3}{\delta z^2} - \frac{n^2}{c^2}\frac{\delta^2 E_3}{\delta t^2} = \frac{1}{c^2}\chi^{(2)}\frac{\delta^2}{\delta t^2}\left(2E_1E_2\right)$$

Fill in the field definitions, and calculate the derivatives.

$$\left(\frac{\delta^2 A_3}{\delta z^2} + 2ik_3\frac{\delta A_3}{\delta z} - \underbrace{k_3^2 A_3 - \frac{n^2\omega_3^2}{c^2}A_3}_{=0,\text{ because }\omega_3 = \frac{ck_3}{n}}\right)e^{i\left(k_3 z - \omega_3 t\right)} = \frac{-2\chi^{(2)}\omega_3^2}{c^2}A_1A_2e^{i\left(\left(k_1+k_2\right)z-\omega_3 t\right)}$$

# Sum Frequency Generation

$$\longrightarrow \left(\frac{\delta^2 A_3}{\delta z^2} + 2ik_3 \frac{\delta A_3}{\delta z}\right) e^{i\left(k_3 z - \omega_3 t\right)} = \frac{-2\chi^{(2)}\omega_3^2}{c^2} A_1 A_2 e^{i\left((k_1+k_2)z - \omega_3 t\right)}$$

Group exponentials

$$\left(\frac{\delta^2 A_3}{\delta z^2} + 2ik_3 \frac{\delta A_3}{\delta z}\right) = \frac{-2\chi^{(2)}\omega_3^2}{c^2} A_1 A_2 e^{i\left(k_1+k_2-k_3\right)z}$$

Slowly-varying envelope approximation: $\left|\frac{\delta^2 A_3}{\delta z^2}\right| \ll \left|k_3 \frac{\delta A_3}{\delta z}\right|$

Valid if $A_3$ does not vary much over a distance of the order of the wavelength.

$$\frac{\delta A_3}{\delta z} = \frac{i\chi^{(2)}\omega_3^2}{k_3 c^2} A_1 A_2 e^{i\left(k_1+k_2-k_3\right)z}$$

$\Delta k = k_1 + k_2 - k_3$ (this is called the phase mismatch)

$d_{\text{eff}} = \frac{1}{2}\chi^{(2)}$ (nonlinear coefficient, used for historical reasons)

## Coupled wave equation:

$$\frac{\delta A_3}{\delta z} = \frac{2id_{\text{eff}}\omega_3^2}{k_3 c^2} A_1 A_2 e^{i\Delta k z}$$

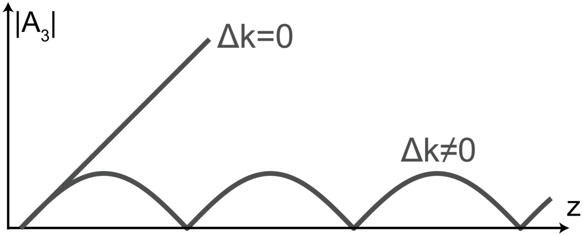
# Sum Frequency Generation

**Coupled wave equation:**

$$\frac{\delta A_3}{\delta z} = \frac{2 i d_{\mathsf{eff}} \omega_3^2}{k_3 c^2} A_1 A_2 e^{i\Delta k z}$$

Notice that:

- $\Delta k = 0 \Rightarrow A_3$ gradually builds up,
- $\Delta k \neq 0 \Rightarrow A_3$ just oscillates up and down.



# Sum Frequency Generation

We can also see this by integrating the coupled wave equation:

$$A_3 = \frac{2id_{\text{eff}}\omega_3^2}{k_3c^2}A_1A_2\int_0^L e^{i\Delta kz}dz$$

$$= \frac{2id_{\text{eff}}\omega_3^2}{k_3c^2}A_1A_2e^{i\frac{\Delta kL}{2}}L\,\text{sinc}\,\frac{\Delta kL}{2}$$

Note: We assume here that $A_1$ and $A_2$ are constant, for simplicity. In reality they decrease as $A_3$ builds up.

Take the magnitude square, and convert amplitudes to intensities:

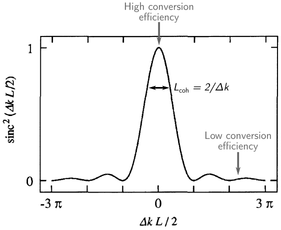
$$I_i = \frac{1}{2}n_i\varepsilon_0 c\,|A_i|^2$$

(we write $n_i$ because the refractive index depends on the frequency)

$$I_3 = \frac{8d_{\text{eff}}^2\omega_3^2I_1I_2L^2}{n_1n_2n_3\varepsilon_0c^2}\,\text{sinc}^2\left(\frac{\Delta kL}{2}\right)$$

The term $\text{sinc}^2\left(\frac{\Delta kL}{2}\right)$ is large ($> 70\%$) over a distance of the order of $L_{\text{coh}} = \frac{2}{\Delta k}$.

The conversion efficiency is maximum when $\Delta k = 0$.

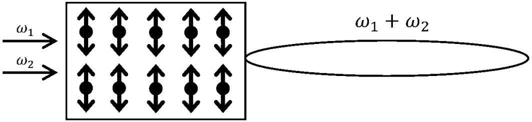

High conversion efficiency

Low conversion efficiency

# Phase matching

Efficient frequency conversion occurs only near $\Delta k = 0$ (phase matching condition).

Microscopically, this corresponds to the case where all dipoles are in phase and add up coherently.

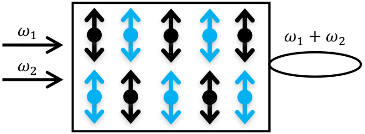


$\text{Power} \sim N^2$

↓

Number of dipoles

In case of phase mismatch, some dipole emissions counteract others, and power fluctuates instead of building up.



# Normal dispersion

Taking phase matching into account, we need to satisfy two equations at the same time:

$$\omega_1 + \omega_2 = \omega_3$$

(energy conservation)

$$n_1\omega_1 + n_2\omega_2 = n_3\omega_3$$

(phase matching; we used $k_i = n_i\omega_i/c$ and $n_i$ denotes the refractive index at $\omega_i$)

This is not possible in the usual case of normal dispersion (i.e. when $n(\omega)$ increases with $\omega$):

$$n_1\omega_1 + n_2\omega_2 = n_3\omega_3 \xrightarrow[\omega_1+\omega_2=\omega_3]{} n_1\omega_1 + n_2\left(\omega_3 - \omega_1\right) = n_3\omega_3$$

$$\xrightarrow[\text{rearrange}]{} \underbrace{\left(n_1 - n_2\right)}_{<0}\omega_1 = \underbrace{\left(n_3 - n_2\right)}_{>0}\omega_3$$

With $\omega_1 < \omega_2 < \omega_3$ and $n_1 < n_2 < n_3$

The problem is even more evident for second harmonic generation:

$$2n_1\omega_1 = n_3\omega_3 \xrightarrow[\omega_3=1\omega_1]{} n_1 = n_3$$

We need the refractive indices to be *equal* at $\omega_1$ and $\omega_3$!

# Birefringent crystals

In practice, one way to obtain phase matching is by using *birefringent crystals*.
Crystals having different refractive indices depending on their orientation.

To show how, we will study the case of frequency doubling in a *BBO* crystal.
BBO = "$\beta$-barium borate"

For this particular case of birefringent phase matching, we assume:

- Second harmonic generation (i.e. we need $n_3 = n_1$)
- Negative uniaxial crystal

  *Negative*: $n_e < n_o$

  *unaxial*: One crystal axis has an "extraordinary" refractive index $n_e$, and all perpendicular directions have an "ordinary" index $n_o$.
- Type I phase matching

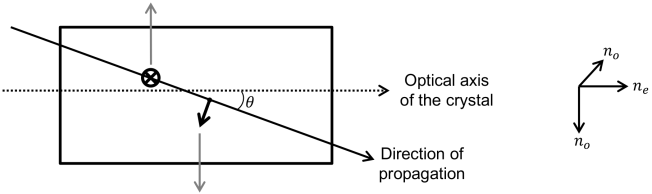
  *Type I*: both input photons have the same polarization

  *Type II*: input photons have orthogonal polarizations

Note: You can find more explanation about other cases of'birefringent phase matching in the book chapter on Moodle.

# Birefringent crystals

If we propagate at an angle $\theta$ of the optical axis of a uniaxial crystal, one polarization experiences $n_o$ while the other experiences a mixture of $n_o$ and $n_e$:



Light with this polarization experiences a refractive index $n_o$.

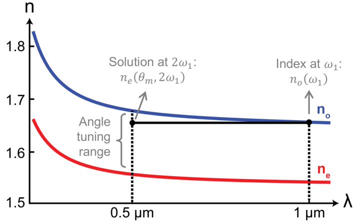
Light with this polarization experiences an index $n_e(\theta)$ that is between $n_o$ and $n_e$.

So, the "effective" extraordinary index $n_e(\theta)$ depends on the angle:

$$\frac{1}{\underbrace{n_e(\theta)^2}_{\text{Extraordinary index at angle } \theta}} = \frac{\cos^2\theta}{n_o^2} + \frac{\sin^2\theta}{\underbrace{n_e^2}_{\text{Principal extraordinary index}}}$$

# Birefringent crystals

By varying the angle $\theta$, we can now satisfy $n_e(\theta, 2\omega_1) = n_o(\omega_1)$:



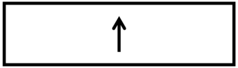
Example: BBO crystal, second harmonic generation with input $\lambda_1 = 1\ \mu\text{m}$. $\rightarrow \theta_m \approx 24^\circ$

Note that birefringent phase matching is not always possible:

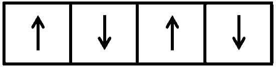
- Not all crystals are birefringent
- Certain nonlinear effects require identical input & output polarizations
- Birefringence may be too small for compensation
  → This is especially the case at shorter wavelengths: while the refractive index changes rapidly, the difference between $n_o$ and $n_e$ stays approximately the same.

# Quasi phase matching

Quasi phase matching relies on a crystal where one of the crystalline axes is periodically inverted (= *periodic poling*):



Homogeneous crystal



Periodically poled crystal

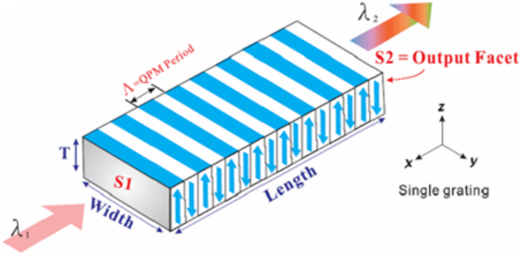
First, remember that in case of phase mismatch, the field amplitude oscillates sinusoidally with distance:

$$A_3 \sim d_{\text{eff}} L \,\mathrm{sinc}\,\frac{L}{\Delta k/2}$$

$$\Big\downarrow$$

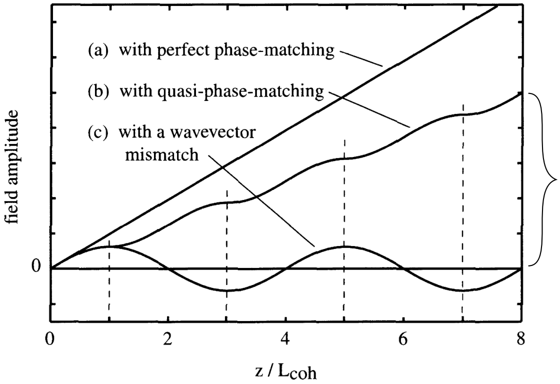
Set $L_{\text{coh}} = \frac{2}{\Delta k}$
Assume $\Delta k$ is constant

$$A_3 \sim d_{\text{eff}} \sin\frac{L}{L_{\text{coh}}}$$



# Quasi phase matching

Periodic poling allows us to invert the sign of $d_{\text{eff}}$ each time the oscillation is about to go down. Therefore, even if $\Delta k \neq 0$, the amplitude keeps building up:



Note that the amplitude builds up more slowly than with perfect phase matching. More precisely:

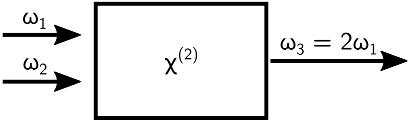
$$E_{\text{quasi}} = \frac{2}{\pi} E_{\text{perfect}}$$

The optimal poling period is $\Lambda = \pi L_{\text{coh}} = \frac{2\pi}{\Delta k}$.

Example: Periodically Poled Lithium Niobate (PPLN), second harmonic generation with input $\lambda_1 = 1.06\ \mu\text{m}$.
$\rightarrow L_{\text{coh}} = 3.4\ \mu\text{m}$

# Second harmonic generation

Let us define the electric fields in the case of **second harmonic generation (SHG)**.



$$\begin{cases} \mathrm{E}_1(z,t) = \mathrm{A}_1(z) e^{ik_1 z} e^{-i\omega_1 t} \text{ (+ complex conjugate)} \\ \mathrm{E}_3(z,t) = \mathrm{A}_3(z) e^{ik_3 z} e^{-i\omega_3 t} \text{ (+ complex conjugate)} \end{cases}$$

Note: SHG is also an $\chi^{(2)}$-effect, so we assume $\chi^{(3)}, \chi^{(4)}, \dots = 0$

We plug this into the wave equation, and collect the terms by frequency:

$$\frac{\delta^2 E}{\delta z^2} - \frac{n^2}{c^2}\frac{\delta^2 E}{\delta t^2} = \frac{1}{\varepsilon_0 c^2}\frac{\delta^2}{\delta t^2}\left(\varepsilon_0 \chi^{(2)} E^2\right)$$

$$\xrightarrow[\text{(+ complex conjugates)}]{E=E_1+E_3} \begin{cases} \frac{\delta^2 E_1}{\delta z^2} - \frac{n^2}{c^2}\frac{\delta^2 E_1}{\delta t^2} = \frac{1}{\varepsilon_0 c^2}\frac{\delta^2}{\delta t^2} \overbrace{\left(\sum_0 \chi^{(2)} \cdot 2E_1^* E_3\right)}^{P_1^{NL}} \\ \frac{\delta^2 E_3}{\delta z^2} - \frac{n^2}{c^2}\frac{\delta^2 E_3}{\delta t^2} = \frac{1}{\varepsilon_0 c^2}\frac{\delta^2}{\delta t^2} \underbrace{\left(\varepsilon_0 \chi^{(2)} \cdot E_1^2\right)}_{P_3^{NL}} \end{cases}$$

Terms with $e^{i\omega_1 t}$ or $e^{i(\omega_3-\omega_1)t}$

Terms with $e^{i\omega_3 t}$ or $e^{i2\omega_1 t}$

# Second harmonic generation

From this, we can calculate the coupled wave equations (as before):

$$\begin{cases} \frac{\delta A_1}{\delta z} = \frac{2id_{\text{eff}}\omega_1^2}{k_1c^2}A_1^*A_3e^{i\Delta kz} & \text{(coupled wave equation for the source)} \\ \frac{\delta A_3}{\delta z} = \frac{id_{\text{eff}}\omega_3^2}{k_3c^2}A_1^2e^{i\Delta kz} & \text{(coupled wave equation for the output)} \end{cases}$$

where the phase mismatch is:

$$\Delta k = 2k_1 - k_3.$$

It is possible to derive explicit formulas for both source and output intensities.
We highlight here only the main points of this calculation.

If you're interested, the full derivation can be found in Boyd, Nonlinear Optics, Chapter 2. Beware that in the book they use Gaussian units (this changes some formulas slightly).

First, one can prove there is conservation of energy at every location $z$:

$$I_1(z) + I_3(z) = I$$

(or $u_1(z) + u_2(z) = 1$ if we use normalized intensities)

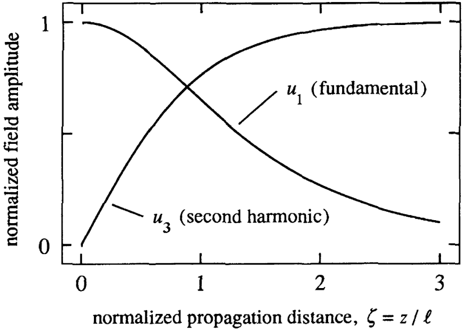
# Second harmonic generation

Second, the conversion happens over a characteristic length scale $\ell$:

$$\ell = \sqrt{\frac{2\varepsilon_0 c^3 n_1^2 n_3}{I}}\frac{1}{2\omega_1 d_{\text{eff}}}$$

(Note the dependencies on $I$ and $d_{\text{eff}}$)

The intensities are then as follows for perfect phase matching ($\Delta k = 0$):

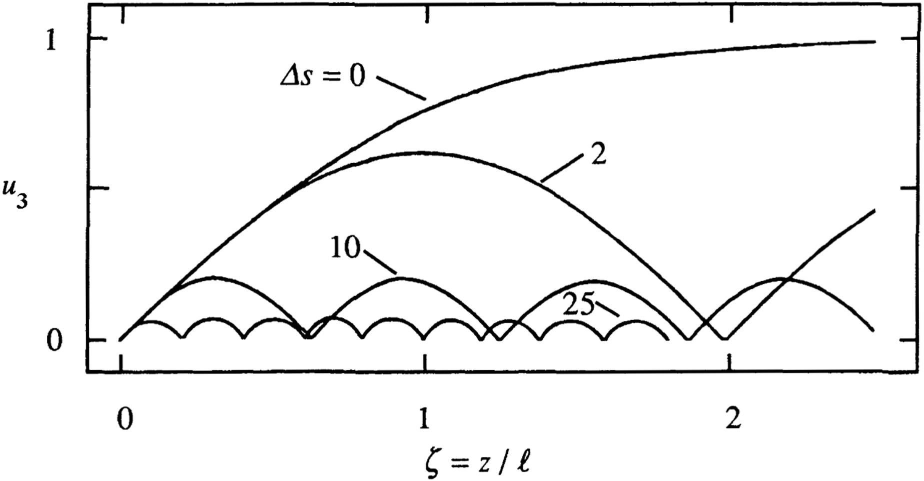


$$\begin{cases} u_1(z) = \tanh \frac{z}{\ell} \\ u_3(z) = \operatorname{sech} \frac{z}{\ell} \end{cases}$$

(These are the normalized intensities,
so $I_1 = I u_1$ and $I_3 = I u_3$.)

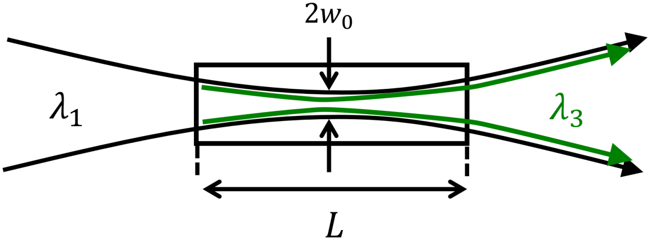
# Second harmonic generation

In case of phase mismatch ($\Delta k \neq 0$), the intensity does not build up but oscillates:



# Second harmonic generation

To illustrate the use of these formulas, we calculate the conversion efficiency for a Gaussian beam with waist $w_0$ focused in a crystal of length $L$:



Assume:

- a crystal length of $L = 1\ \mathrm{cm}$,
- a source wavelength $\lambda_1 = 1064\ \mathrm{nm}$,
- an input power $P = 1\ \mathrm{W}$,
- a nonlinear coefficient $d_{\text{eff}} = 12.5\ \frac{\mathrm{pm}}{\mathrm{V}}$,
- perfect phase matching,
- we focus the Gaussian beam so that the depth of focus $b$ (which is twice the Rayleigh range $z_R$) is equal to the crystal length $L$.

# Second-order Autocorrelation

The condition on the depth of focus of the Gaussian beam allows us to calculate the waist, and therefore the beam intensity at the waist:

$$L = b = 2z_R \quad \longrightarrow \quad L = \frac{2\pi w_0^2}{\lambda_1/n_1} \quad \longrightarrow \quad w_0 \approx 29\ \mu\text{m}$$

$$I = \frac{P}{\pi w_0^2} \quad \longrightarrow \quad I \approx 376\ \text{MW}/\text{m}^2$$

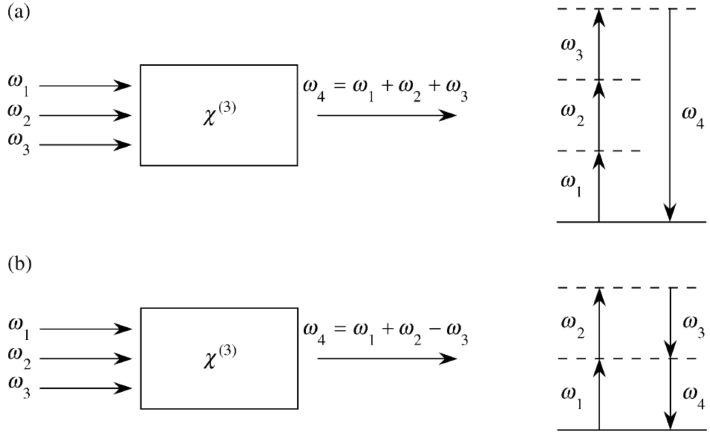
We can now calculate the characteristic length $\ell$ and the efficiency $\eta$:

$$\ell = \sqrt{\frac{2\varepsilon_0 c^3 n_1^2 n_3}{I}}\frac{1}{2\omega_1 d_{\text{eff}}} \quad \longrightarrow \quad \ell \approx 7.2\ \text{cm}$$

$$\eta = \frac{u_3^2(z=L)}{u_1^2(z=0)} = \frac{\tanh^2 \frac{L}{\ell}}{1} \quad \longrightarrow \quad \eta \approx 2\%$$

# Second-order Autocorrelation

Third-order (Kerr) nonlinear contribution to the nonlinear polarization:



$$P_{NL}^{(3)} = \varepsilon_0 \chi^{(3)} E(t)^3$$

**Intensity dependent refractive index:**

$$n = n_0 + n_2 I$$

**Kerr nonlinearity and intensity-dependent refractive index**

$$n_2 = \frac{3}{2 n_0^2 \varepsilon_0 c} \chi^{(3)}$$