36. Nonlinear optics: $\chi^{(2)}$ processes

The wave equation with nonlinearity

Second-harmonic generation: making blue light from red light

- approximations: SVEA, zero pump depletion
- phase matching
- quasi-phase matching
- surface SHG

When is it necessary to think about $\chi^{(3)}$?

The wave equation with nonlinearity

We have derived the wave equation in a medium, for the situation where the polarization is non-linear in E:

$$\frac{\partial^2 E}{\partial z^2} - \frac{n^2}{c^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P^{NL}}{dt^2}$$

linear optics

where $P^{NL} = \varepsilon_0 \left[\chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots \right]$

In order for this to make sense, this series must converge.

As a result, we must assume that $\chi^{(2)} >> \chi^{(3)} >> \chi^{(4)} >> \chi^{(5)} \ldots$

So the most important nonlinear term is the 2nd order term: the one involving $\chi^{(2)}$. To simply the problem, let's ignore all the other terms.

The wave equation with $\chi^{(2)}$ nonlinearity

So the wave equation can be written as:

$$\frac{\partial^2 E}{\partial z^2} - \frac{n^2}{c^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P^{(2)}}{dt^2}$$

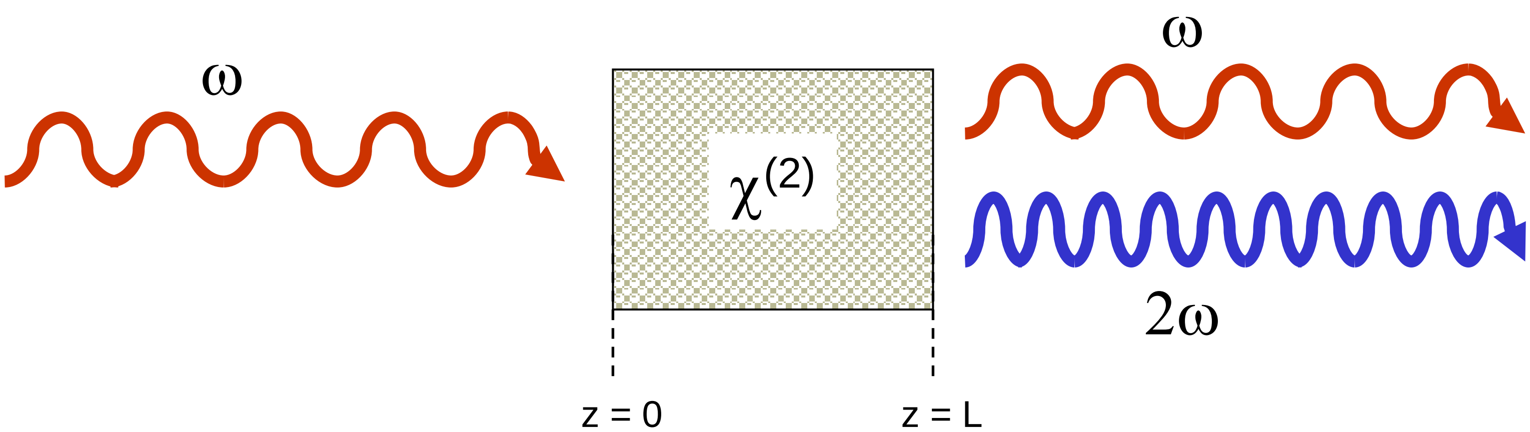
where the 2nd order polarization is given by:

$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} \left(E_{incident}\right)^2$$

As we saw in the last lecture, there are several non-linear processes that can occur, even if we restrict ourselves to $\chi^{(2)}$.

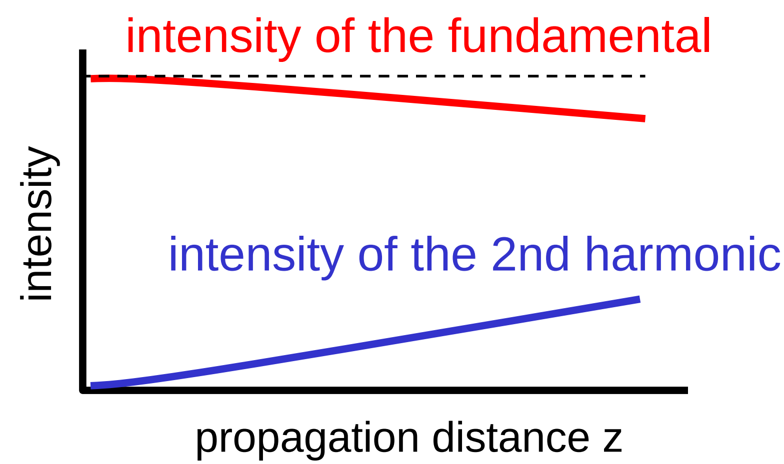
Pick one particularly interesting one: second harmonic generation (SHG) of a single incident wave at frequency ω.

Second Harmonic Generation: SHG



In this process, we imagine that one laser at frequency ω (the 'fundamental') is used to illuminate a nonlinear medium.

As this field propagates through the medium, its intensity will be depleted and the intensity of the 2nd harmonic wave (initially zero) will grow.



Describing the 2nd harmonic wave

We are interested in the behavior of the field that oscillates at 2ω; that is, the 2nd harmonic. We can assume that this field is of the form:

$$E_{2\omega}(z,t) = A_{2\omega}(z)\, e^{jk_{2\omega}z - j2\omega t} + c.c.$$

where we require that the amplitude $A_{2\omega}(z)$ is slowly varying, and also that it vanishes at the input facet of the nonlinear medium:

$$A_{2\omega}(z=0) = 0$$

Furthermore, the wave vector of this wave is related to the refractive index of the nonlinear medium at frequency 2ω:

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

Our goal is to determine $A_{2\omega}(z)$.

What equation must the 2nd harmonic obey?

The 2nd harmonic wave must obey the wave equation, of course.

$$\frac{\partial^2 E_{2\omega}}{\partial z^2} - \left(\frac{n(2\omega)}{c}\right)^2 \frac{\partial^2 E_{2\omega}}{\partial t^2} = \mu_0 \frac{\partial^2 P^{(2)}}{dt^2}$$

As we have seen, the 2nd-order polarization results from the field at frequency ω - the fundamental. Putting in the spatial dependence explicitly:

$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} \left(A_\omega e^{-j\omega t + jk_\omega z}\right)^2$$

the amplitude of the incident field (the one at frequency ω)

this is the k of the incident field:

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

$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)} A_\omega{}^2 e^{j[2k_\omega z - 2\omega t]}$$

Plugging in to the wave equation…

Plug our assumed forms for $E_{2\omega}(z,t)$ and $P^{(2)}$, to find:

$$\left(\underbrace{\cancel{\frac{\partial^2 A_{2\omega}}{\partial z^2}}}_{\text{SVEA}} + 2jk_{2\omega}\frac{\partial A_{2\omega}}{\partial z} \cancel{- k_{2\omega}^2 A_{2\omega}} + \cancel{n^2\left(\frac{2\omega}{c}\right)^2 A_{2\omega}} \right) e^{j\left(k_{2\omega}z - \cancel{2\omega t}\right)}$$

$$= -\frac{\chi^{(2)}\left(2\omega\right)^2}{c^2} A_\omega^{\ 2} e^{j\left[2k_\omega z - \cancel{2\omega t}\right]}$$

Slowly Varying Envelope Approximation (SVEA):

$$\left|\frac{\partial^2 A_{2\omega}}{\partial z^2}\right| << \left|k_{2\omega}\frac{\partial A_{2\omega}}{\partial z}\right|$$

So we neglect the second derivative of $A_{2\omega}$.

Solving the wave equation in second order

The nonlinear wave equation becomes:

$$2jk_{2\omega}\frac{\partial A_{2\omega}}{\partial z} = -\frac{4\chi^{(2)}\omega^2}{c^2} A_\omega(z)^2 \, e^{j2k_\omega z} e^{-jk_{2\omega} z}$$

At this point, we *could* find a similar first-order differential equation for A_ω, and then solve the two coupled equations.

But, instead of doing that, let's see if we can gain some physical insight by making another simplifying assumption:

Assume: The incident field is not significantly depleted by the conversion process. That is, A_ω does not decrease very much with increasing z.

$\longrightarrow$ A_ω is independent of z.

In this case, we can easily integrate both sides of this equation.

Integrate both sides

$$\int_0^z \frac{\partial A_{2\omega}}{\partial z'} dz' = \frac{2j\chi^{(2)}\omega^2}{k_{2\omega}c^2} A_\omega^{\ 2} \int_0^z e^{j[2k_\omega z' - k_{2\omega} z']} dz'$$

This is just $A_{2\omega}(z)$.

Define the 'phase mismatch' $\Delta k = 2k_\omega - k_{2\omega}$

Note, this is just:

$$2\frac{2\pi}{\lambda} n_\omega - \frac{2\pi}{\lambda/2} n_{2\omega}$$

$$= \frac{4\pi}{\lambda}\left(n_\omega - n_{2\omega}\right)$$

We can do the integral on the right side:

$$\int_0^z e^{j\Delta k \cdot z'} dz' = \frac{1}{j\Delta k}\left[e^{j\Delta k \cdot z} - 1\right]$$

Thus we've arrived at a result!

$$A_{2\omega}(z) \propto \chi^{(2)} A_\omega^{\ 2} \cdot \frac{\exp[j\Delta k z] - 1}{\Delta k}$$

The solution

The intensity of the second harmonic radiation is proportional to $|A_{2\omega}|^2$.

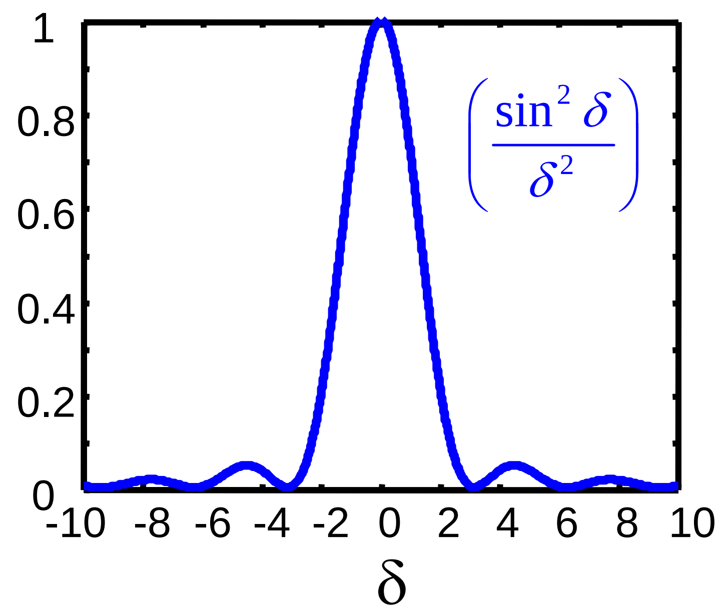
$$I_{2\omega}(z) \propto \left|A_{2\omega}(z)\right|^2 \propto I_\omega{}^2 \frac{\sin^2(\Delta k \cdot z/2)}{(\Delta k)^2}$$

δ = "dimensionless phase mismatch"

$$\boxed{I_{2\omega}(z) \propto I_\omega{}^2 z^2 \frac{\sin^2(\delta)}{\delta^2} \quad \text{where} \quad \delta = \Delta k \cdot z/2}$$

The intensity of the 2nd harmonic is proportional to the square of the intensity of the fundamental.

It also depends sensitively on the product of Δk and z.



Phase matching for a $\chi^{(2)}$ process

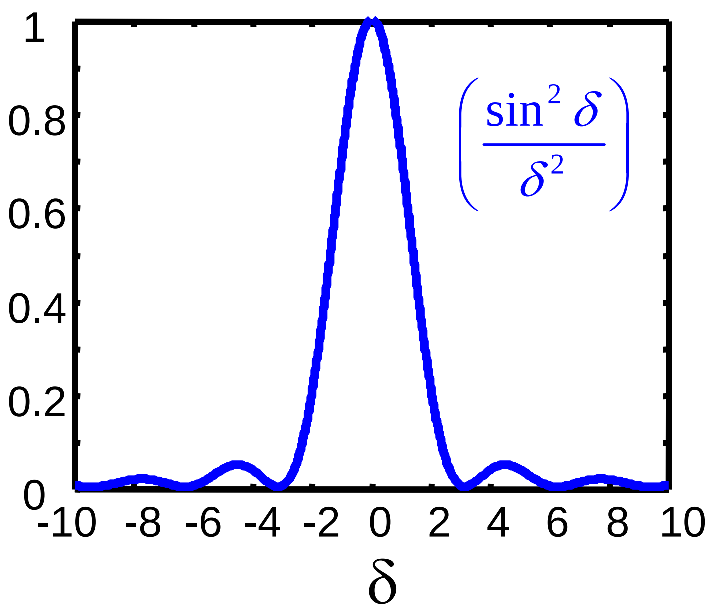
$$I_{2\omega}(z) \propto I_\omega{}^2 z^2 \left(\frac{\sin(\delta)}{\delta} \right)^2$$

where $\delta = \Delta k \cdot z/2$

$$= \frac{2\pi z}{\lambda_{input}} \left(n_\omega - n_{2\omega} \right)$$

To summarize:

- SVEA and zero-depletion approximations give lowest order solution.
- Intensity of SHG radiation is proportional to the square of the input intensity.
- In the limit $\delta << 1$, intensity of SHG radiation grows quadratically with propagation distance.
- Intensity of SHG is very sensitive to phase mismatch - maximum when $\Delta k = 0$



For example, how much does the SHG intensity drop if $|\delta| = 1$?

If $\delta = 1$, then $(\sin\delta/\delta)^2 = 0.71$.

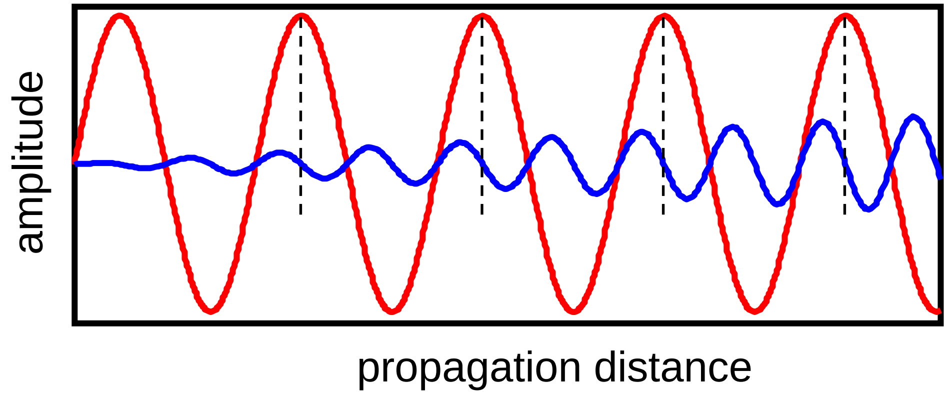
The condition $|\delta| < 1$ corresponds to $\left| \Delta k \right| < \frac{2}{L}$

$\rightarrow$ If the SHG medium is too thick for a given Δk, conversion efficiency suffers.

What does phase matching mean?

When $\Delta k = 0$, this means that $n(\omega) = n(2\omega)$. The phase velocity of the input and the 2nd harmonic are equal. $\lambda_\omega = 2\,\lambda_{2\omega}$.

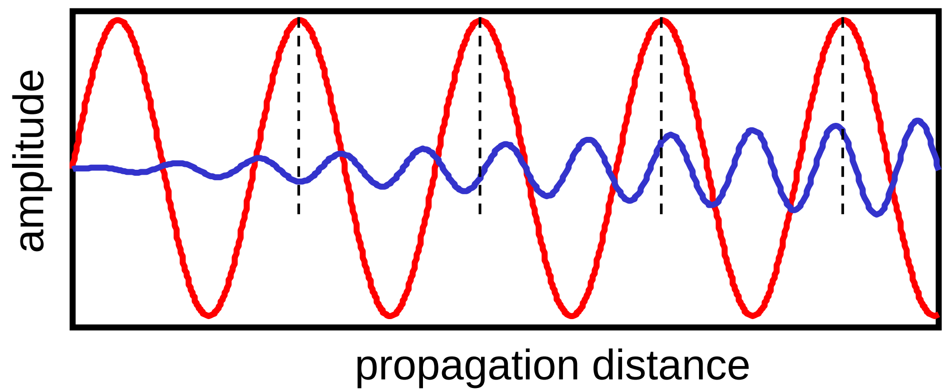
phase-matched:



The two waves maintain the same relative phase as they propagate.

When Δk is not zero, the phase velocity of the fundamental and 2nd harmonic are different, and $\lambda_\omega \neq 2\,\lambda_{2\omega}$. As z increases, the 2nd harmonic wave walks out of phase with the input wave.

not phase-matched:

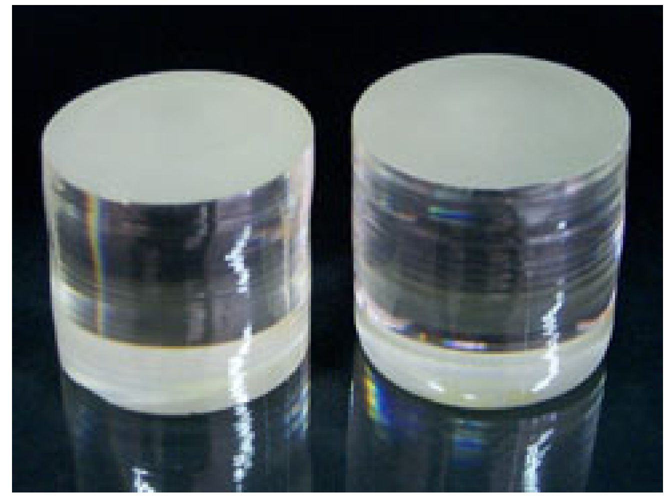


The condition $\Delta k\, L \ll 1$ ensures that the two waves don't walk too far out of phase with each other before reaching the end of the SHG crystal.

Materials and configurations for $\chi^{(2)}$ NLO

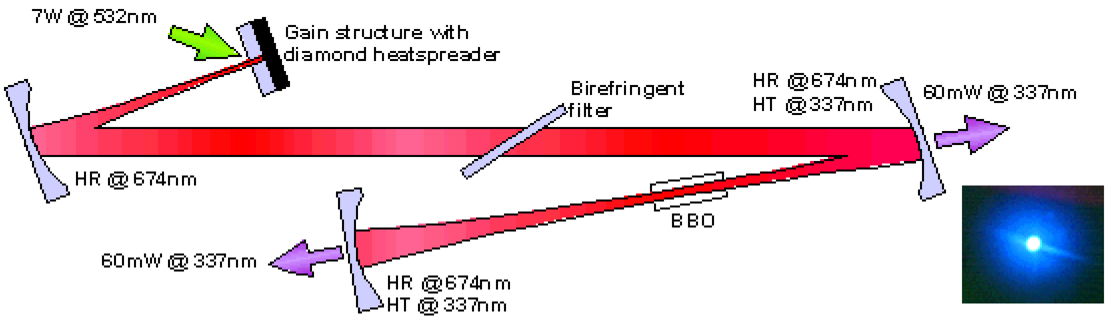
There are a number of materials commonly used for SHG or other frequency conversion effects based on $\chi^{(2)}$.

- KDP: potassium di-hydrogen phosphate
- BBO: beta-barium borate
- $LiNbO_3$: lithium niobate
- etc.



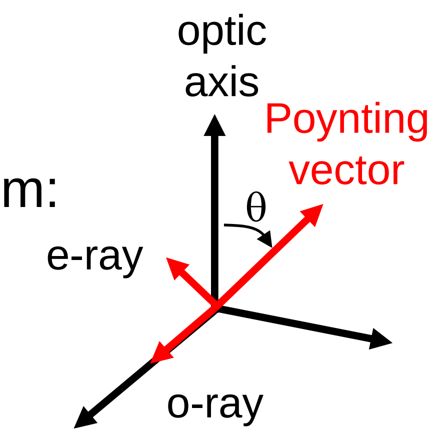
$LiNbO_3$ crystals

A non-linear crystal inside the laser cavity to produce UV light:

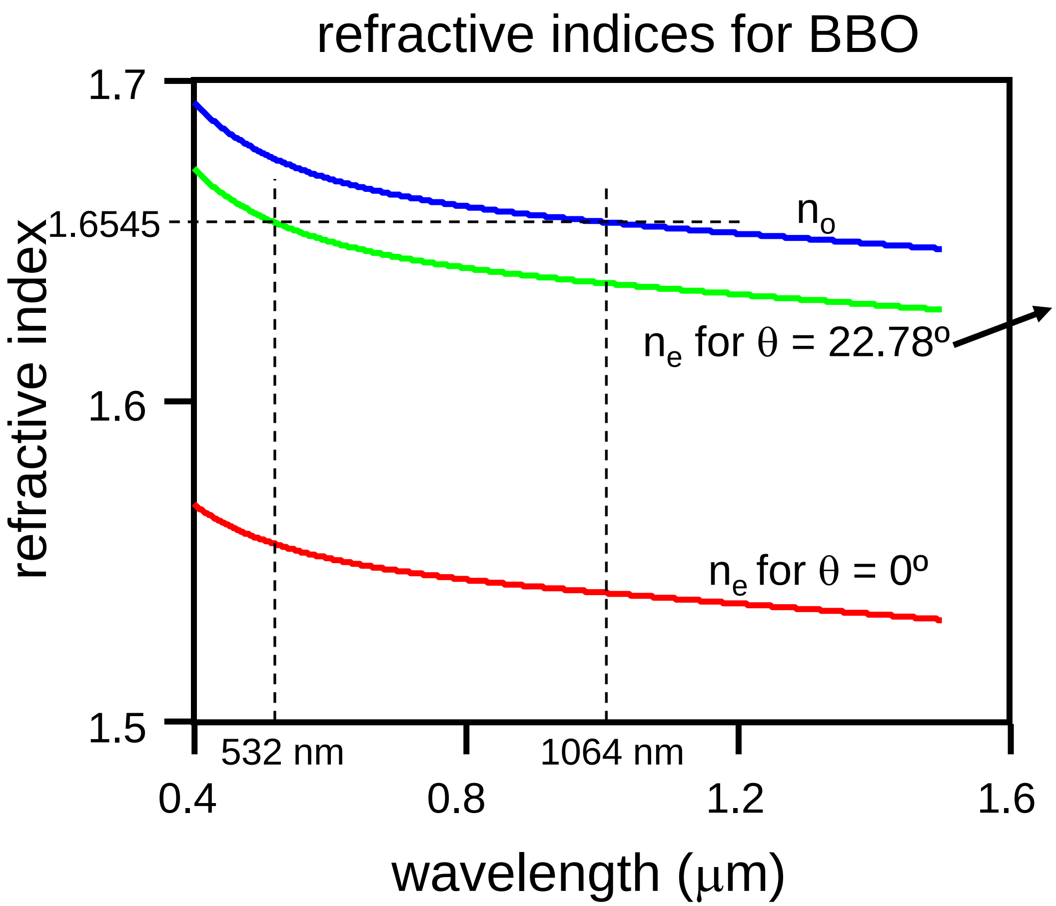


This is a “VECSEL”: a “vertical external cavity surface emitting laser”

SHG illustration



Example of matching $n(\omega)$ and $n(2\omega)$ in a nonlinear medium:



For λ = 1064 nm, at this angle, $n_o(\omega) = n_e(2\omega)$ and thus $\Delta k = 0$.

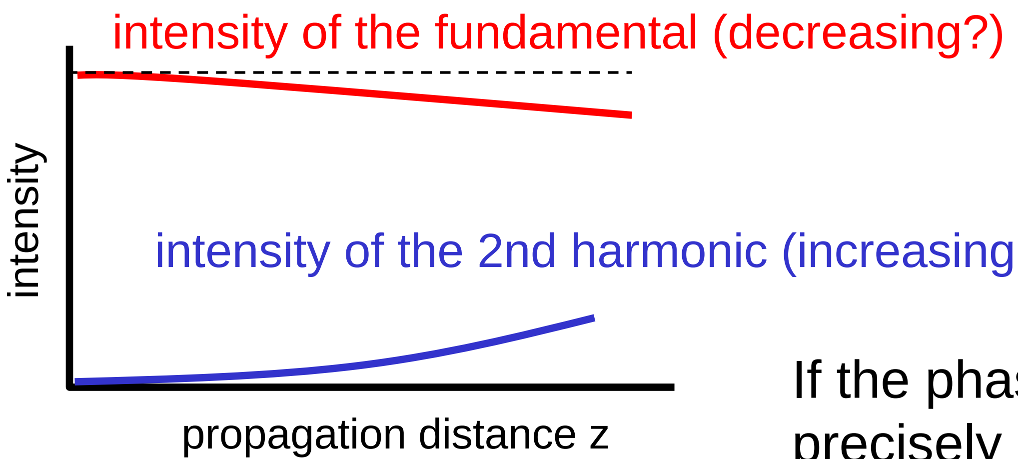
What if we changed the angle slightly? For example: 23º.

Then $n_o(\omega)$ is unchanged. But $n_e(2\omega) = 1.6542$. And thus:

$$\Delta k = \frac{4\pi}{\lambda}\left(n_\omega - n_{2\omega}\right) = 4150 \text{ m}^{-1}$$

For a crystal of thickness = 1 mm: $\delta = \Delta k \cdot z/2 = 2.1$ and so $\dfrac{\sin^2\delta}{\delta^2} = 0.18$

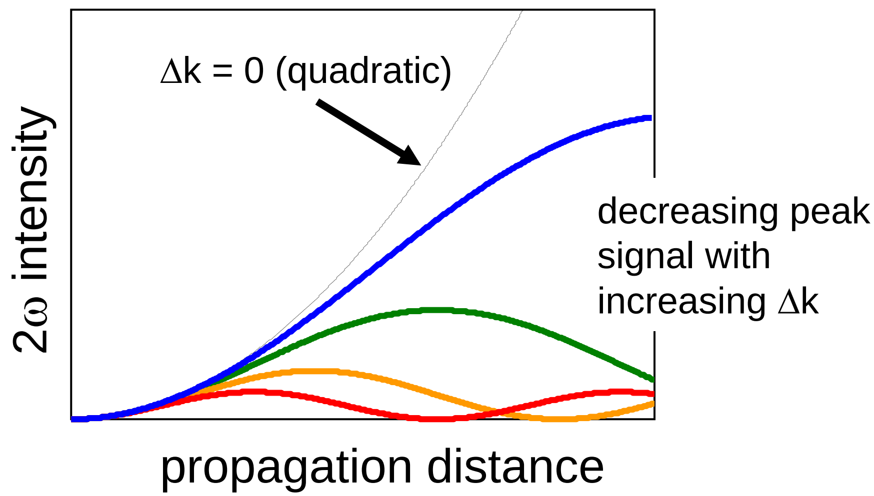
What if the phase matching is not perfect?

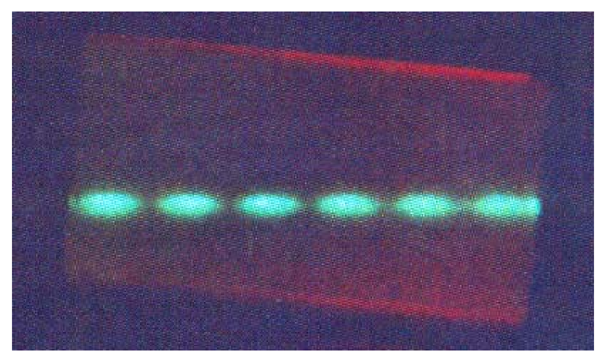


$$I_{2\omega}(z) \propto I_{\omega}{}^{2} z^{2} \frac{\sin^{2}(\Delta k \cdot z/2)}{(\Delta k \cdot z/2)^{2}}$$

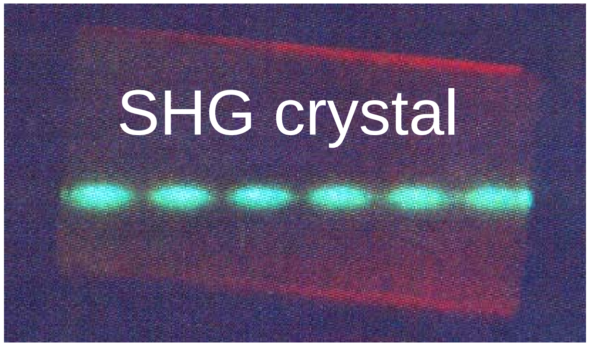
If the phase mismatch is not precisely zero, then how does the second harmonic intensity behave?

The SHG intensity oscillates as a function of propagation distance:





Another way to boost the SHG efficiency



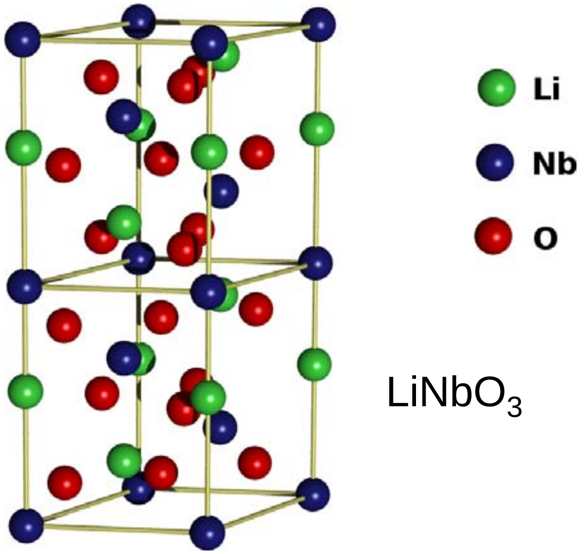
Why does the signal oscillate?

If phase matching condition is not perfect, then after a certain length (called the 'coherence length' L_{coh}), the fundamental and 2nd harmonic walk out of phase with each other.

At that point, the process reverses itself, and the fundamental grows while the 2ω beam diminishes. This process then oscillates.

What if, at $z = L_{coh}$, we could flip the sign of $\chi^{(2)}$? This would change the phase of $E_{2\omega}$ by π. Instead of cancelling out as it propagates beyond L_{coh}, $E_{2\omega}$ would be further enhanced.

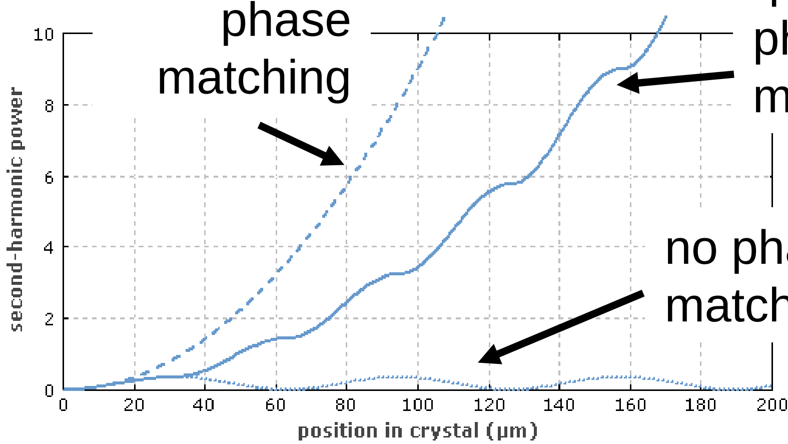
In some cases, we can control the sign of $\chi^{(2)}$ by changing the crystal structure.



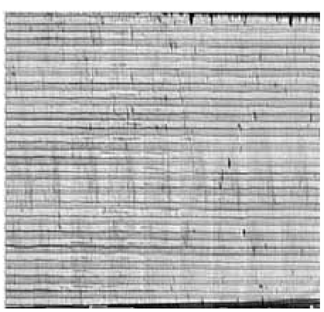
$LiNbO_3$

Quasi-phase matching

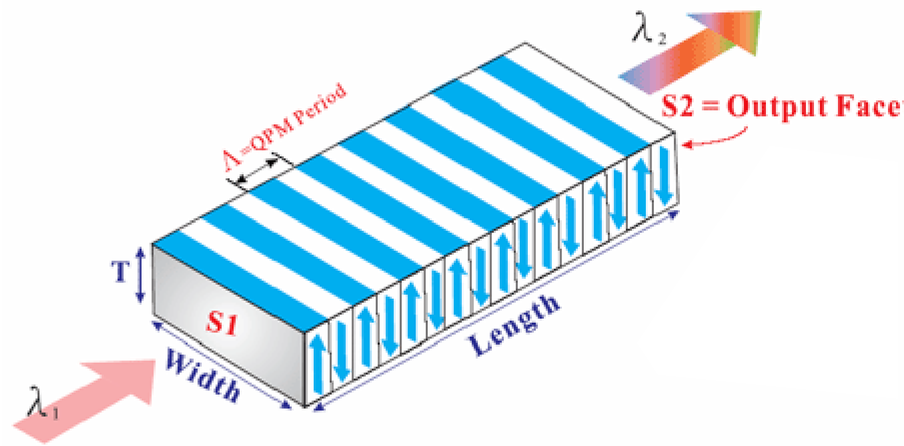
Flipping the sign of $\chi^{(2)}$ once each coherence length is known as "quasi-phase matching." It has become a critically important method for efficient second harmonic generation.



The process of fabricating a material where the sign of $\chi^{(2)}$ flips back and forth is known as "periodic poling".

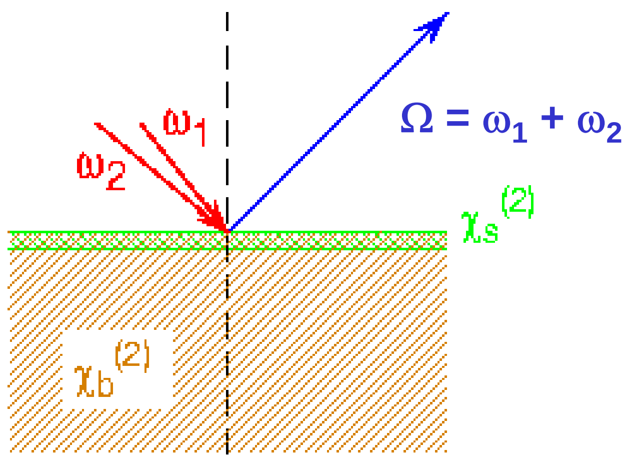


A photo of PPLN: periodically poled lithium niobate

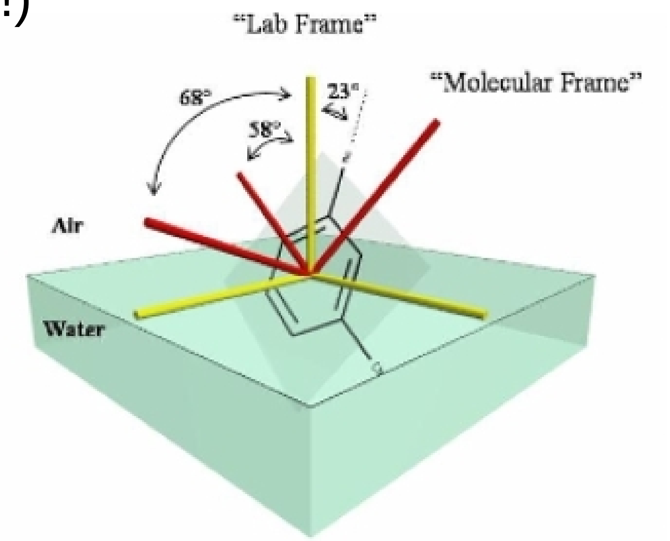


SHG at a surface

Another method of minimizing $\delta = \Delta k\, z\, /\, 2$: use a very small value of z. For example, at a surface or an interface.



"surface second harmonic generation"
- a very sensitive probe of surfaces
(but very weak!)



Applications:

- measuring the orientation of molecules at a liquid surface
- studying buried interfaces, e.g., silicon/insulator

Do we ever worry about $\chi^{(3)}$?

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

If the power series is to converge, then $| \chi^{(3)} | << | \chi^{(2)} |$

So when are $\chi^{(3)}$ effects important?

$\longrightarrow$ Usually, $\chi^{(3)}$ is only important when $\chi^{(2)}$ is equal to zero.

It is easy to argue that $\chi^{(2)}$ is zero most of the time…

Symmetry considerations

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

Consider a medium which exhibits inversion symmetry.

- many crystalline materials including all cubic crystals
- any amorphous material (glassy solid, liquid, gas)

In a material like that, reversing the sign of E must also reverse the sign of the induced polarization.

$$-P^{(2)} = \varepsilon_0 \chi^{(2)} [-E]^2$$

$P^{(2)} = -P^{(2)}$ can be true only if $\chi^{(2)} = 0$, as well as $\chi^{(4)}$, $\chi^{(6)}$, etc.

In this case, the largest non-linear effect is $\chi^{(3)}$.

Next lecture: $\chi^{(3)}$ effects