12. Nonlinear optics I

What are nonlinear-optical effects and why do they occur?

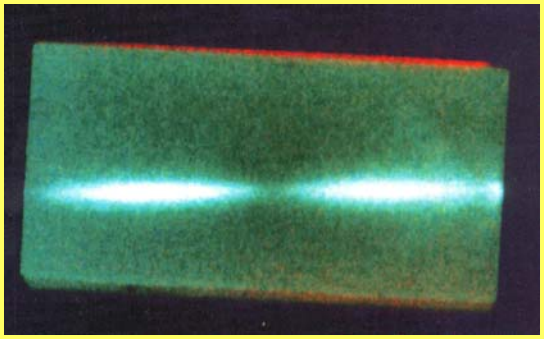
Maxwell's equations in a medium

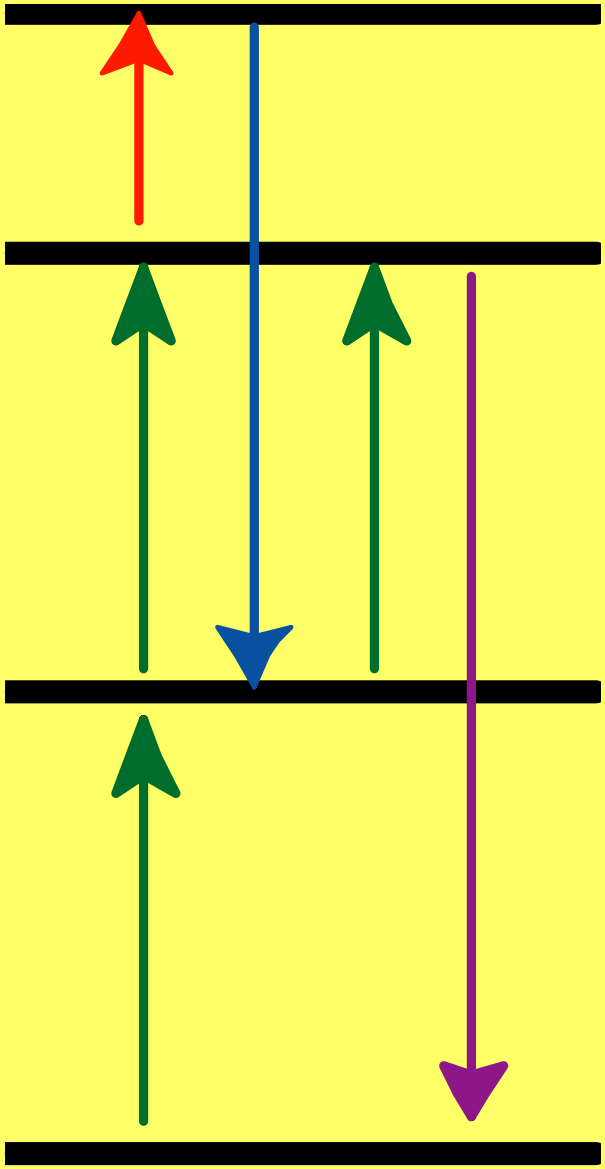
Nonlinear-optical media

Second-harmonic generation

Conservation laws for photons ("Phase-matching")

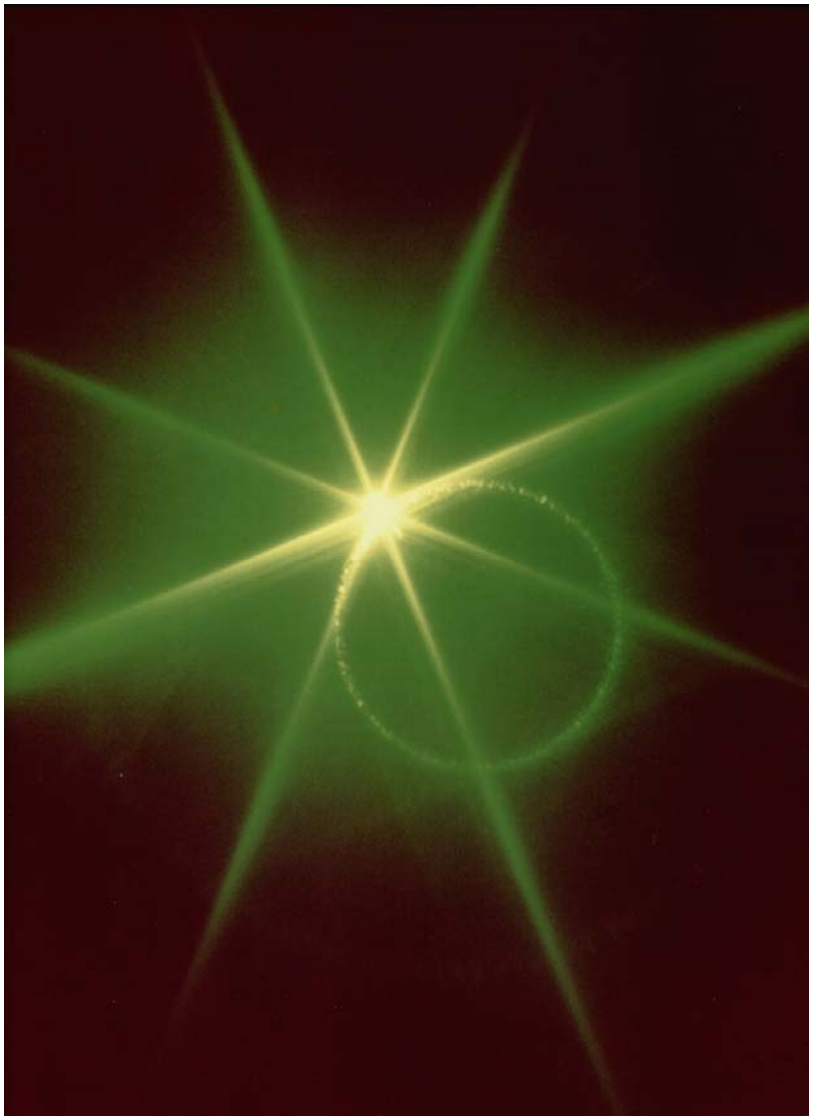
Quasi-phase-matching





Nonlinear Optics can produce many exotic effects.

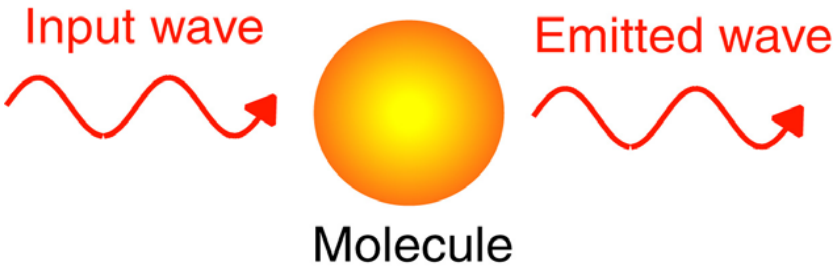
Sending infrared light into a crystal yielded this display of green light:



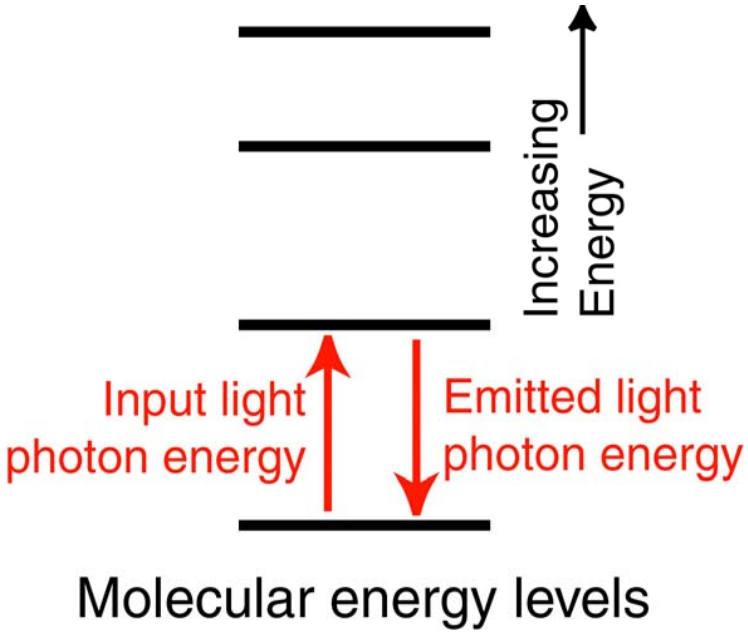
Nonlinear optics allows us to change the color of a light beam, to change its shape in space and time, to switch telecommunications systems, and to create the shortest events ever made by humans.

Why do nonlinear-optical effects occur?

Recall that, in normal linear optics, a light wave acts on a molecule, which vibrates and then emits its own light wave that interferes with the original light wave.

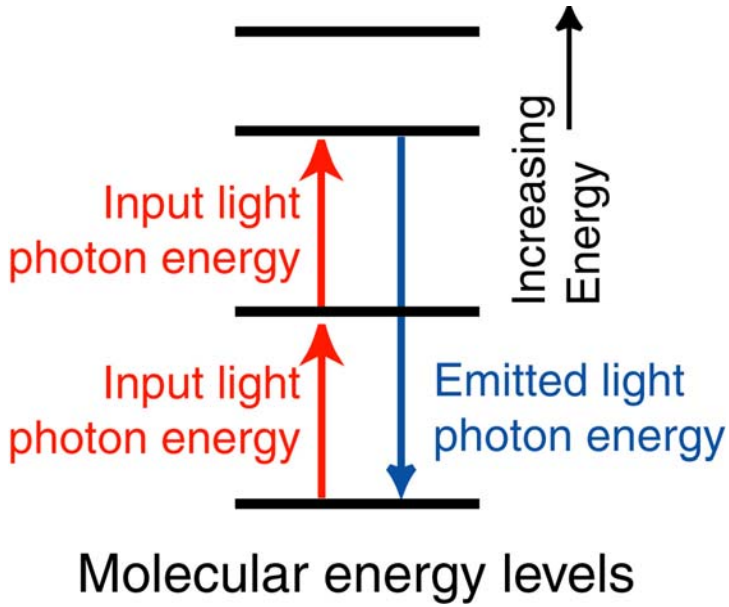


We can also imagine this process in terms of the molecular energy levels, using arrows for the photon energies:

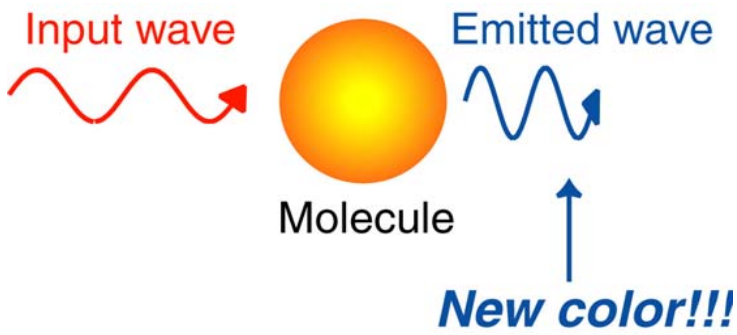


Why do nonlinear-optical effects occur? (continued)

Now, suppose the irradiance is high enough that many molecules are excited to the higher-energy state. This state can then act as the lower level for additional excitation. This yields vibrations at all frequencies corresponding to all energy differences between populated states.



Molecular energy levels



Reminder: Maxwell's Equations in a Medium

The induced polarization, P, contains the effect of the medium.
The inhomogeneous wave equation (in one dimension):

$$\frac{\partial^2 E}{\partial x^2} - \frac{1}{c_0^{\,2}} \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{dt^2}$$

The polarization is usually proportional to the electric field:

$$\vec{P} = \varepsilon_0 \chi \vec{E}$$ χ = unitless proportionality constant

Recall, for example, in the forced oscillator model, we found:

$$P(t) = \frac{Ne^2}{m} \frac{E(t)}{\omega_0^{\,2} - \omega^2 + i\gamma\omega}$$

Then, the wave equation becomes:

$$\frac{\partial^2 E}{\partial x^2} - \frac{1}{c_0^{\,2}} \frac{\partial^2 E}{\partial t^2} = \varepsilon_0 \mu_0 \chi \frac{\partial^2 E}{dt^2}$$

or $$\frac{\partial^2 E}{\partial x^2} - \frac{(1+\chi)}{c_0^{\,2}} \frac{\partial^2 E}{\partial t^2} = 0$$ since $$\frac{1}{c_0^{\,2}} = \varepsilon_0 \mu_0$$

Reminder: Maxwell's Equations in a Medium

$$\frac{\partial^2 E}{\partial x^2} - \frac{(1+\chi)}{c_0{}^2}\frac{\partial^2 E}{\partial t^2} = 0$$

But this is the same equation as the usual homogeneous equation, if we define a new constant c:

$$\frac{1}{c^2} = \frac{(1+\chi)}{c_0{}^2}$$

And, we call the quantity $\sqrt{1+\chi}$ the "refractive index".

So, we can describe light in a medium just like light in vacuum, as long as we take into account the refractive index correction.

But this only worked because P was proportional to E…

What if it isn't? Then P is a *non-linear* function of E!

Maxwell's Equations in a *Nonlinear* Medium

Nonlinear optics is what happens when the polarization is the result of higher-order terms in the field:

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

$$= P_{Linear} + P_{non-linear}$$

Then the wave equation must look like this:

$$\frac{\partial^2 E}{\partial x^2} - \frac{n^2}{c^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_{non-linear}}{dt^2}$$

The linear term can be treated in the same way as before, giving rise to the refractive index. But the non-linear term is a problem…

$$\frac{\partial^2 E}{\partial x^2} - \frac{n^2}{c^2}\frac{\partial^2 E}{\partial t^2} = \varepsilon_0 \mu_0 \chi^{(2)} \frac{\partial^2}{dt^2}\left(E^2\right) + \varepsilon_0 \mu_0 \chi^{(3)} \frac{\partial^2}{dt^2}\left(E^3\right) + \ldots$$

Usually, $\chi^{(2)}$, $\chi^{(3)}$, etc., are very small and can be ignored.
But not if E is big…

The effects of the non-linear terms

What sort of effect does this non-linear term have?

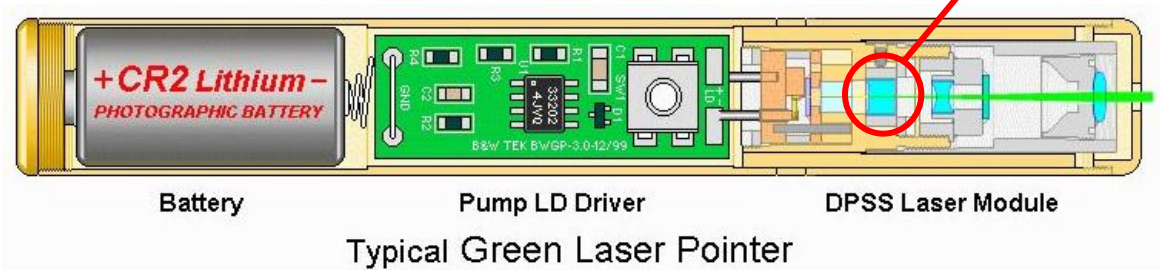
If we write the field as: $E(t) \propto E_0 \exp(i\omega t) + E_0^* \exp(-i\omega t)$

then $E(t)^2 \propto E_0^2 \exp(2i\omega t) + 2\left|E_0\right|^2 + E_0^{*2} \exp(-2i\omega t)$

terms that vary at a new frequency, the 2nd harmonic, 2ω!

Nonlinearity can lead to the generation of new frequency components.

This can be extremely useful:



Typical Green Laser Pointer

Frequency doubling crystal:
1064 nm ⟶ 532 nm

Sum and difference frequency generation

Suppose there are two different-color beams present, not just one:

$$E(t) = E_1 \exp(i\omega_1 t) + E_1^* \exp(-i\omega_1 t) + E_2 \exp(i\omega_2 t) + E_2^* \exp(-i\omega_2 t)$$

Then $E(t)^2$ has 16 terms:

$$E(t)^2 \propto E_1^2 \exp(2i\omega_1 t) + E_1^{*2} \exp(-2i\omega_1 t)$$ 2nd harmonic of ω_1

$$+ E_2^2 \exp(2i\omega_2 t) + E_2^{*2} \exp(-2i\omega_2 t)$$ 2nd harmonic of ω_2

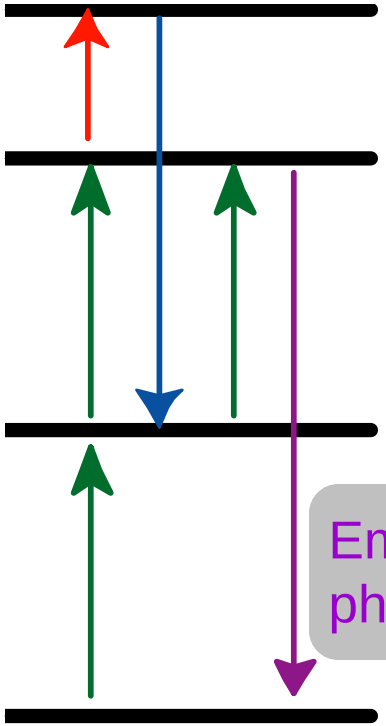
$$+ 2E_1 E_2 \exp(i[\omega_1 + \omega_2]t) + 2E_1^* E_2^* \exp(-i[\omega_1 + \omega_2]t)$$ sum frequency

$$+ 2E_1 E_2 \exp(i[\omega_1 - \omega_2]t) + 2E_1^* E_2^* \exp(-i[\omega_1 - \omega_2]t)$$ difference frequency

$$+ 2|E_1|^2 + 2|E_2|^2$$ zero frequency - known as “optical rectification”

This is an awful lot of processes - do they all occur simultaneously?
Which one dominates (if any)? What determines the efficiency?

Complicated nonlinear-optical effects can occur.



Nonlinear-optical processes are often referred to as:

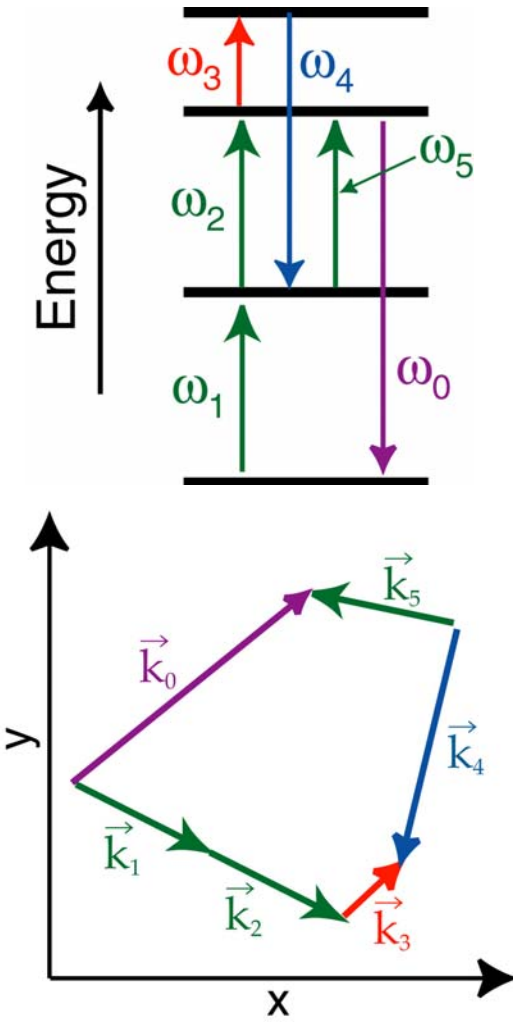
"N-wave-mixing processes"

where N is the number of photons involved (including the emitted one).

This cartoon illustrates a 6-wave mixing process.
It would involve the $\chi^{(5)}$ term in the wave equation.

The more photons (i.e., the higher the order) the weaker the effect, however. Very-high-order effects can be seen, but they require very high irradiance, since usually $\chi^{(2)} > \chi^{(3)} > \chi^{(4)} > \chi^{(5)}$ …

Conservation laws for photons in nonlinear optics



Energy must be conserved. Recall that the energy of a photon is $\hbar\omega$. Thus:

$$\omega_1 + \omega_2 + \omega_3 - \omega_4 + \omega_5 = \omega_0$$

Photon momentum must also be conserved. The momentum of a photon is $\hbar\vec{k}$, so:

$$\vec{k}_1 + \vec{k}_2 + \vec{k}_3 + \vec{k}_4 + \vec{k}_5 = \vec{k}_0$$

But $\vec{k}_0$ is related to ω_0: $\left|\vec{k}_0\right| = \frac{2\pi n}{\lambda} = \frac{n\omega_0}{c}$

So $\vec{k}_0$ may not correspond to a light wave at frequency ω_0!

Satisfying these two relations simultaneously is called "**phase-matching**."

Usually, only one (or zero) of the many possible N-wave mixing processes can be phase-matched at a time.

Phase-matching: an example

Consider the 2nd harmonic generation process:

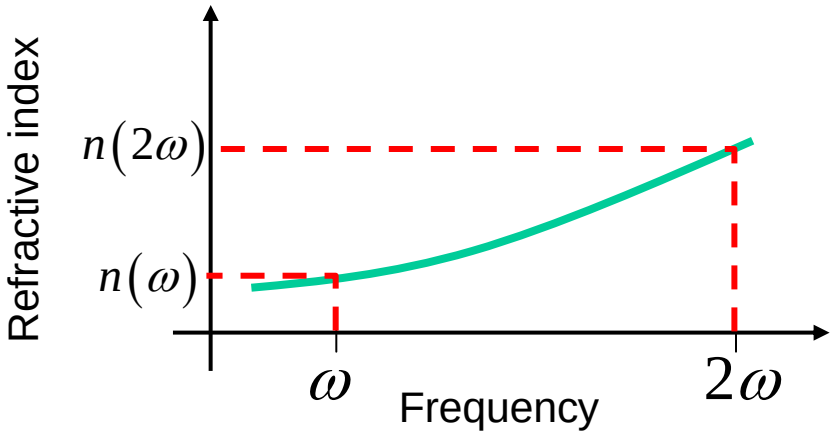
Energy conservation requires: ω in → nonlinear material → 2ω out

Momentum conservation requires: $\vec{k}(\omega)+\vec{k}(\omega)=\vec{k}(2\omega)$

2 red photons 1 blue photon

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

$$n(2\omega)=n(\omega)$$



Unfortunately, dispersion prevents this from ever happening!

Phase-matching Second-Harmonic Generation using birefringence

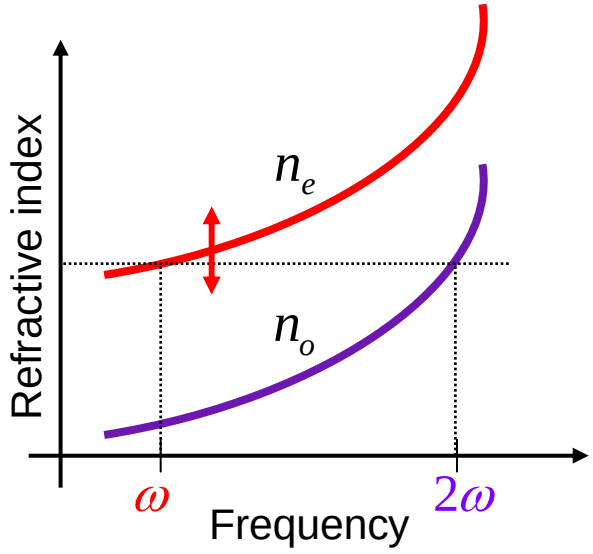
Birefringent materials have different refractive indices for different polarizations: the “Ordinary” and “Extraordinary” refractive indices!

Using this, we can satisfy the phase-matching condition.

For example:

Use the extraordinary polarization for ω and the ordinary for 2ω:

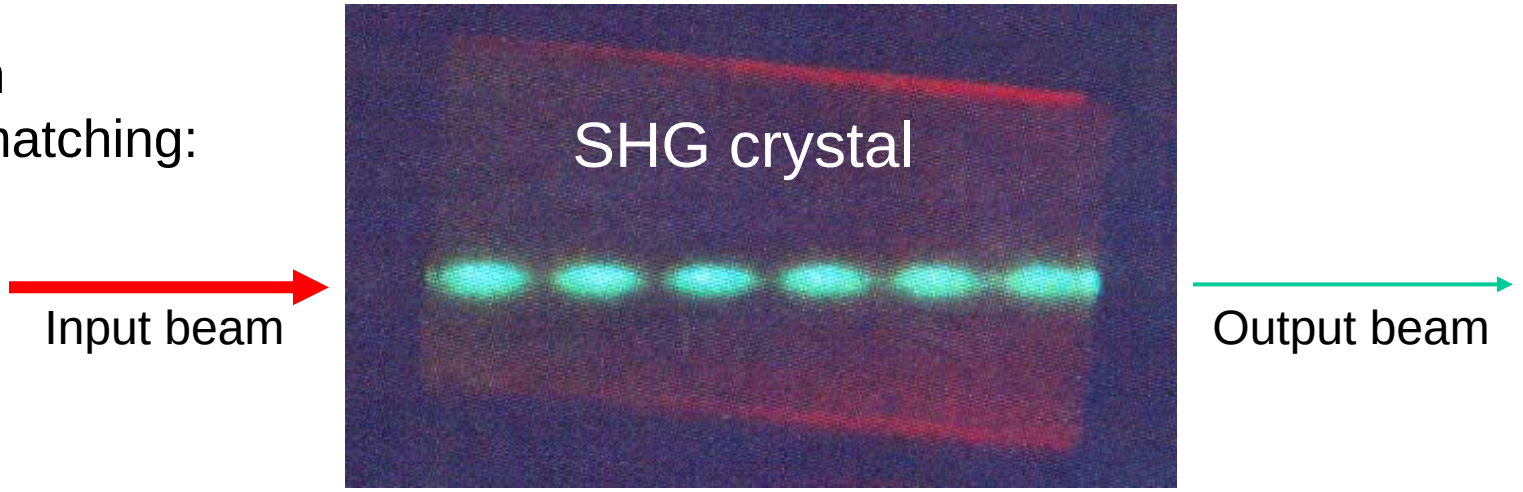
$$n_o(2\omega) = n_e(\omega)$$



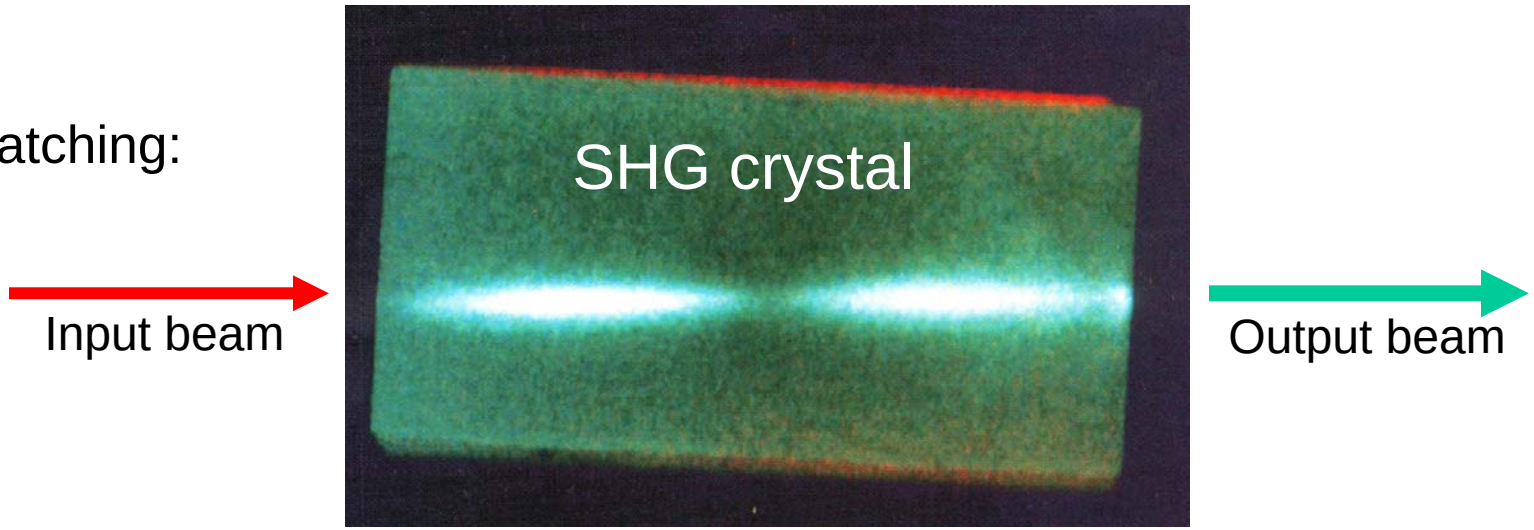
n_e depends on propagation angle, so by rotating the birefringent crystal, we can tune the condition precisely by moving the red curve up and down relative to the blue curve.

Light created in real crystals

Far from
phase-matching:



Closer to
phase-matching:

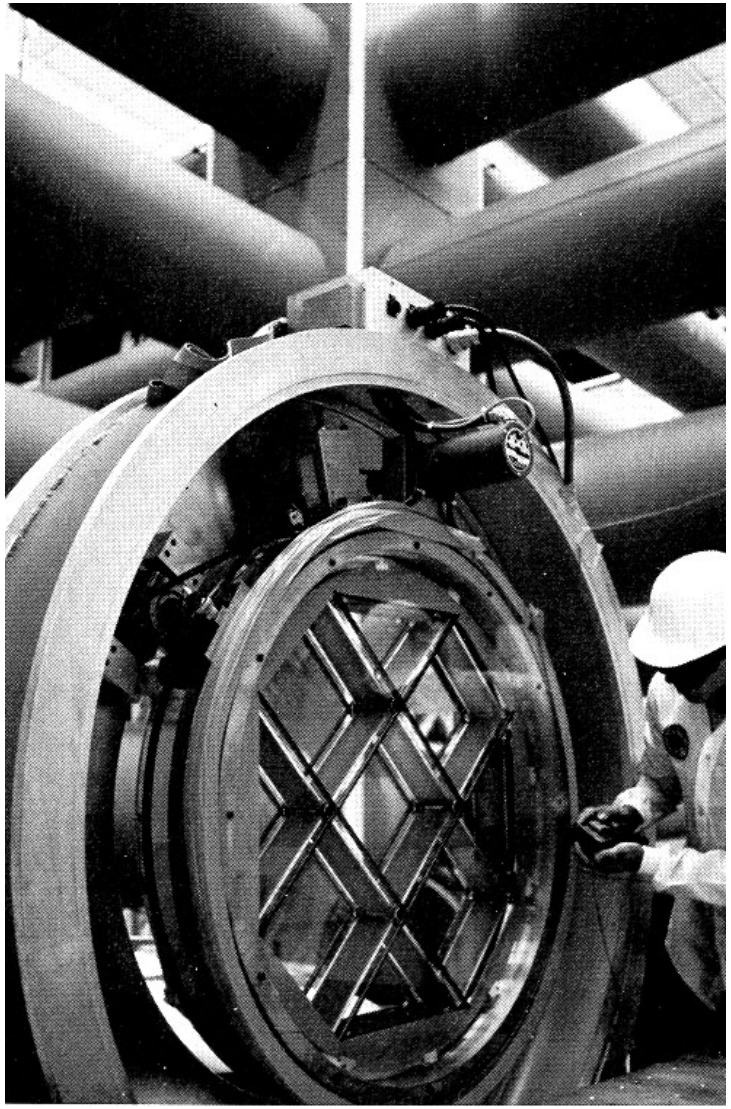


Note that SH beam is brighter as phase-matching is achieved.

Second-Harmonic Generation

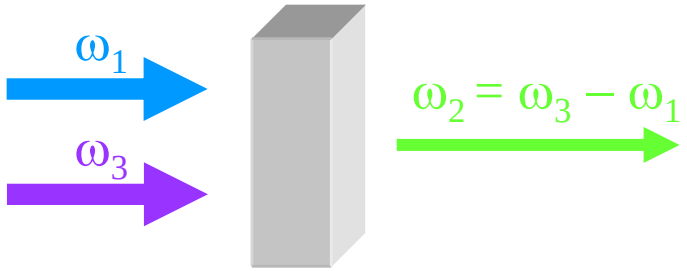
SHG KDP crystals at Lawrence Livermore National Laboratory

These crystals convert as much as 80% of the input light to its second harmonic. Then additional crystals produce the third harmonic with similar efficiency!

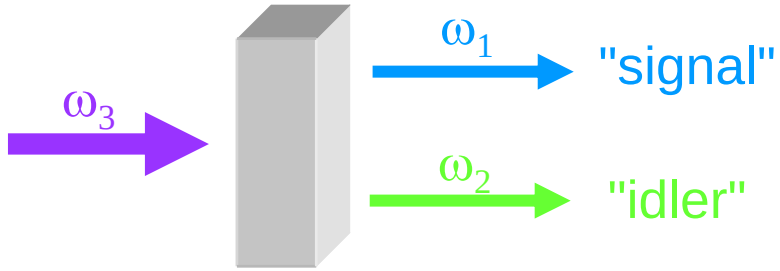


Difference-Frequency Generation: Optical Parametric Generation, Amplification, Oscillation

Difference-frequency generation takes many useful forms.

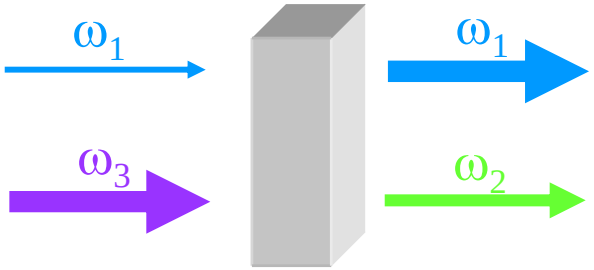


Parametric Down-Conversion
(Difference-frequency generation)

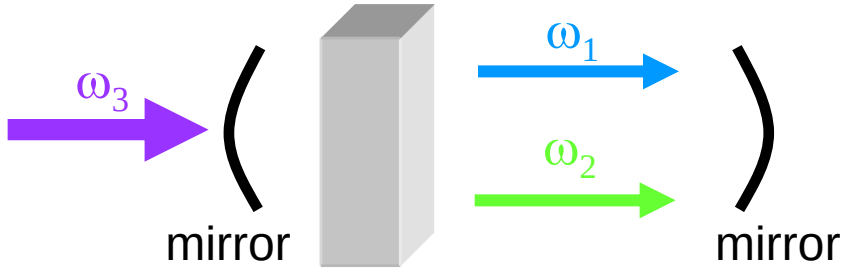


Optical Parametric
Generation (OPG)

By convention:
$\omega_{signal} > \omega_{idler}$



Optical Parametric
Amplification (OPA)

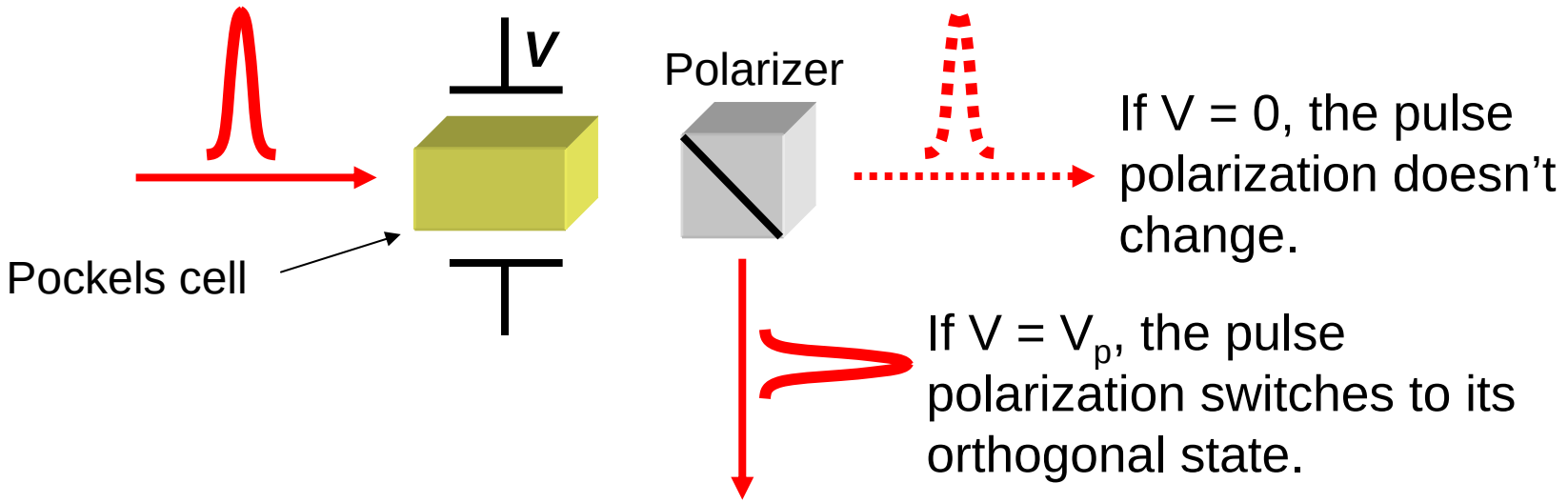


Optical Parametric
Oscillation (OPO)

All of these are $\chi^{(2)}$ processes (three-wave mixing).

Another 2nd-order process: Electro-optics

Applying a voltage to a crystal changes its refractive indices and introduces birefringence. In a sense, this is sum-frequency generation with a beam of zero frequency (but not zero field!).



The Pockels effect can be described as a $\chi^{(2)}$ nonlinear optical interaction, where $E^2 \rightarrow E(\omega)\, E(\omega = 0)$. Sum frequency is at $\omega + 0 = \omega$.

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:

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

linear optics

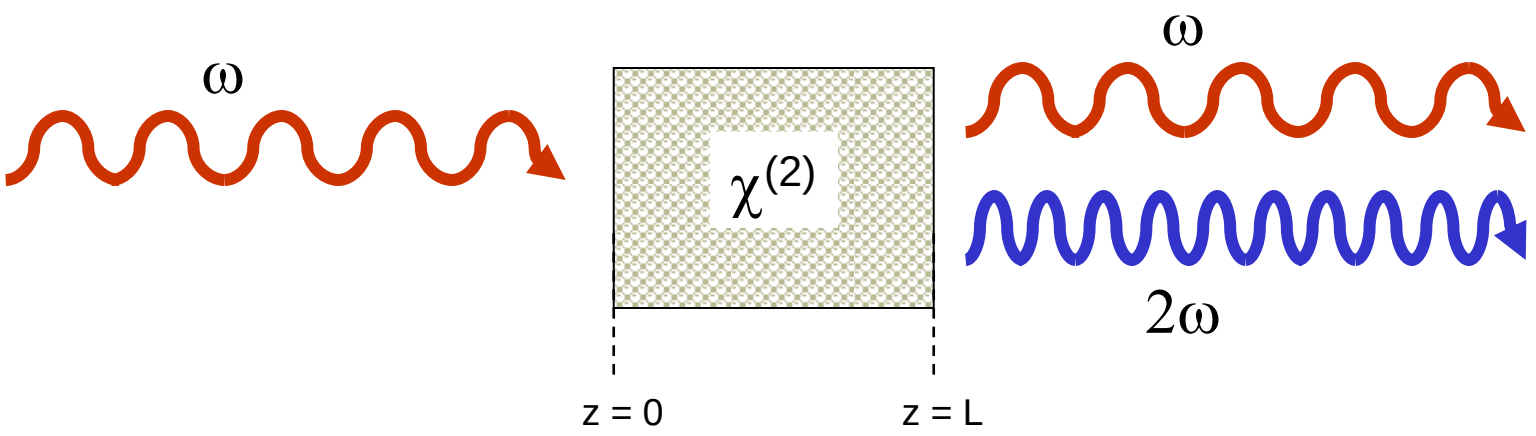
where $P^{NL} = \varepsilon_0 \left[\chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots \right]$

Usually, $\chi^{(2)} >> \chi^{(3)}$

In these cases, we neglect the third (and higher) orders.

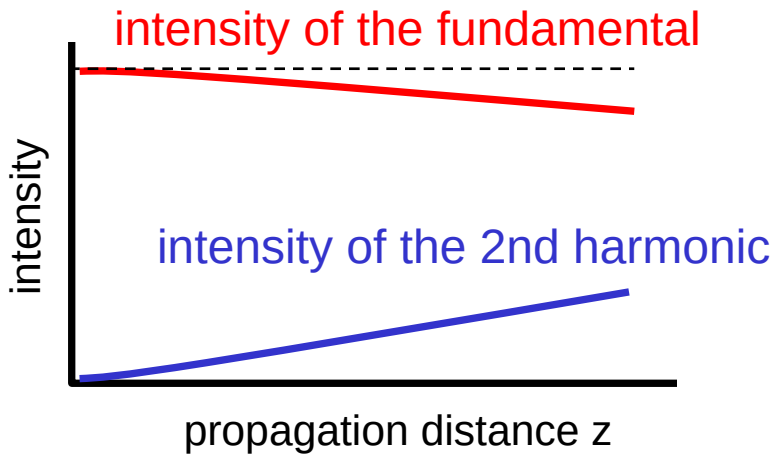
A good example: second harmonic generation

Second Harmonic Generation: SHG



In this process, we imagine that one laser (at frequency ω) 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:

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

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:

$$P^{(2)}(t) = 2\varepsilon_0 \chi^{(2)} \left(E_0 e^{-i\omega t + ik_\omega z}\right)^2$$

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

this is the k of the incident field:

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

$$P^{(2)}(t) = 2\varepsilon_0 \chi^{(2)} E_0{}^2 e^{i[2k_\omega z - 2\omega t]}$$

Plugging in to the wave equation…

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

$$\left(\frac{\partial^2 A_{2\omega}}{\partial z^2} + 2jk_{2\omega}\frac{\partial A_{2\omega}}{\partial z} - k_{2\omega}^2 A_{2\omega} + n^2\left(\frac{2\omega}{c}\right)^2 A_{2\omega} \right) e^{i\left(k_{2\omega}z - 2\omega t\right)}$$

$$= -\frac{2\chi^{(2)}(2\omega)^2}{c^2} E_0{}^2 e^{i\left[2k_\omega z - 2\omega t\right]}$$

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:

$$2ik_{2\omega}\frac{\partial A_{2\omega}}{\partial z} = -\frac{8\chi^{(2)}\omega^2}{c^2}E_0(z)^2 e^{i2k_\omega z}e^{-ik_{2\omega}z}$$

Now, we *could* find a similar first-order differential equation for E_0, 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, E_0 does not decrease very much with increasing z.

$\longrightarrow$ E_0 is independent of z.

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

Integrate both sides

$$\int_0^z \frac{\partial A_{2\omega}}{\partial z'} dz' = \frac{4i\chi^{(2)}\omega^2}{k_{2\omega}c^2} E_0{}^2 \int_0^z e^{i\left[2k_\omega z' - k_{2\omega} z'\right]} dz'$$

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

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

We can do the integral on the right side:

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

Note, this is just:

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

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

Thus we've arrived at a result!

$$A_{2\omega}(z) \propto E_0{}^2 \cdot \frac{\exp\left[i\Delta k z\right] - 1}{\Delta k}$$

The solution

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

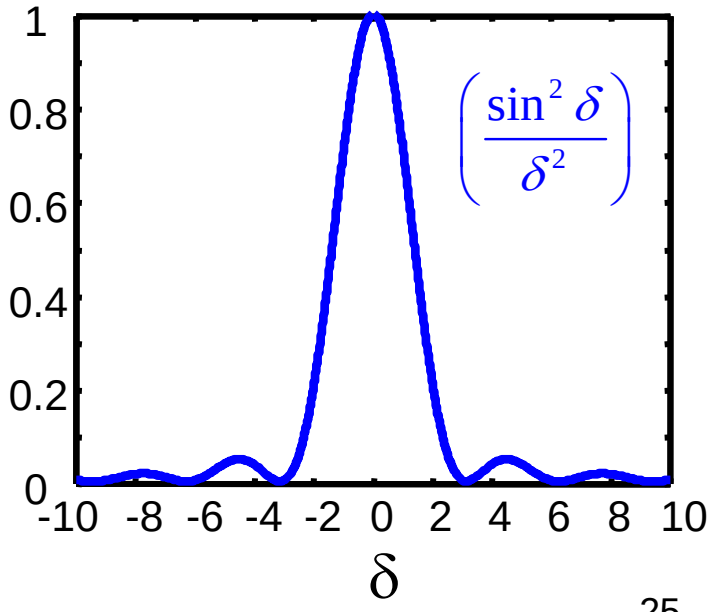
$$I_{2\omega}(z) \propto \left|A_{2\omega}(z)\right|^2 \propto I_0^{\,2} \frac{\sin^2(\Delta k \cdot z/2)}{(\Delta k)^2}$$

$$= I_0^{\,2} z^2 \frac{\sin^2(\delta)}{\delta^2} \quad \text{where} \quad \delta = \Delta k \cdot z/2$$

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.

δ = dimensionless phase mismatch

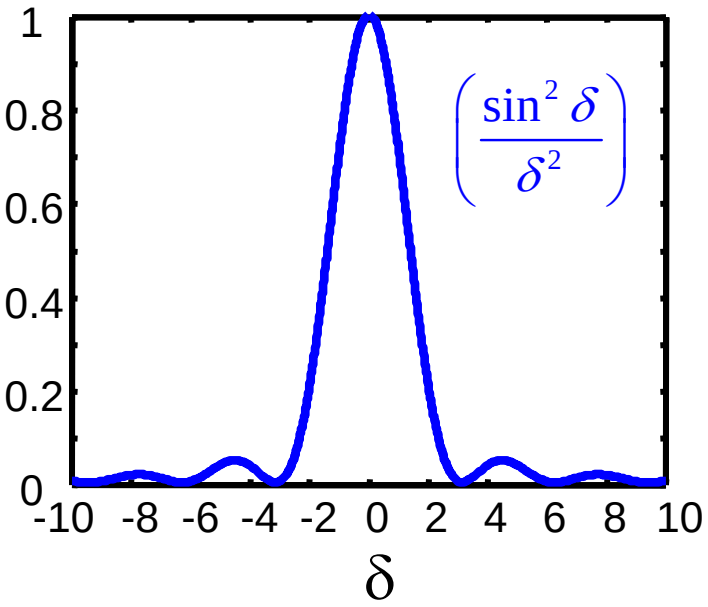


Phase matching for a $\chi^{(2)}$ process

$$I_{2\omega}(z) \propto I_0{}^2 z^2 \frac{\sin^2(\Delta k \cdot z/2)}{(\Delta k \cdot z/2)^2}$$

To summarize:

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



SHG intensity is most efficient for $|\delta| < 1$

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

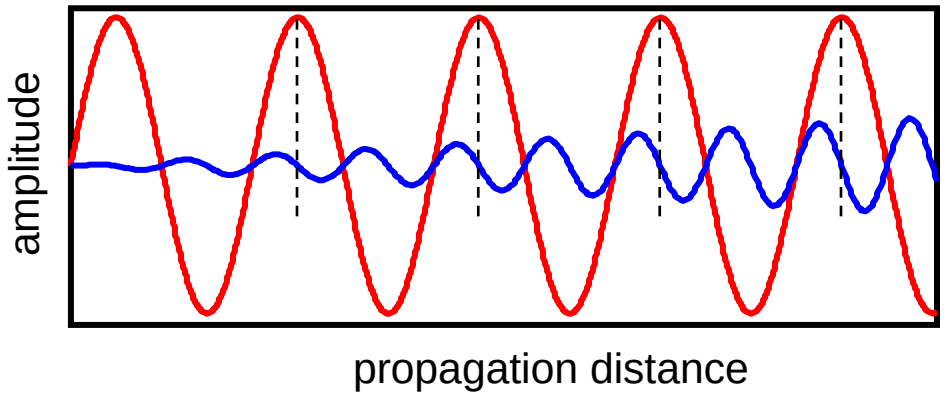
$|\delta| < 1$ corresponds to $|\Delta k| < \frac{2}{L}$

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

What does phase matching mean?

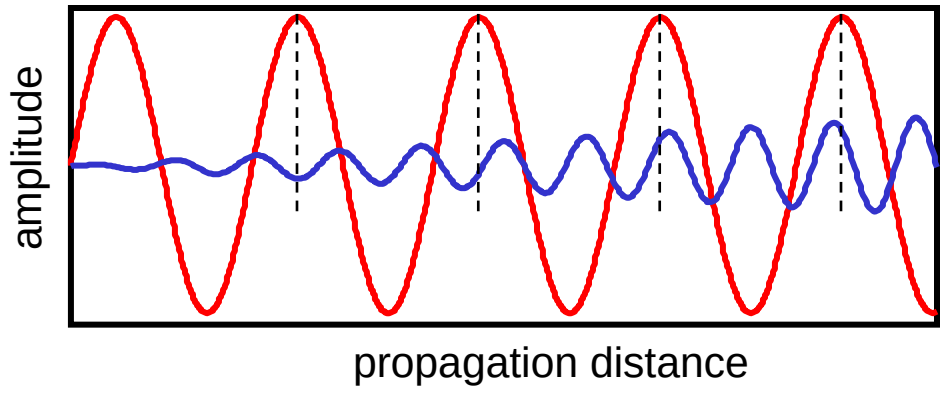
When $\Delta k = 0$, this means that $n(\omega) = n(2\omega)$. The phase velocity of the fundamental and 2nd harmonic are equal. $\lambda_\omega = 2\,\lambda_{2\omega}$.

phase-matched:



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 gets increasingly out of phase with the fundamental.

not phase-matched:

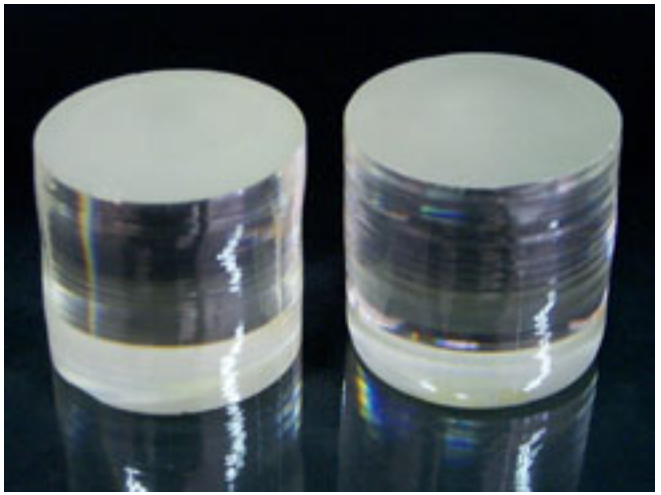


This is why $\Delta k\, L << 1$ is the important condition to satisfy.

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

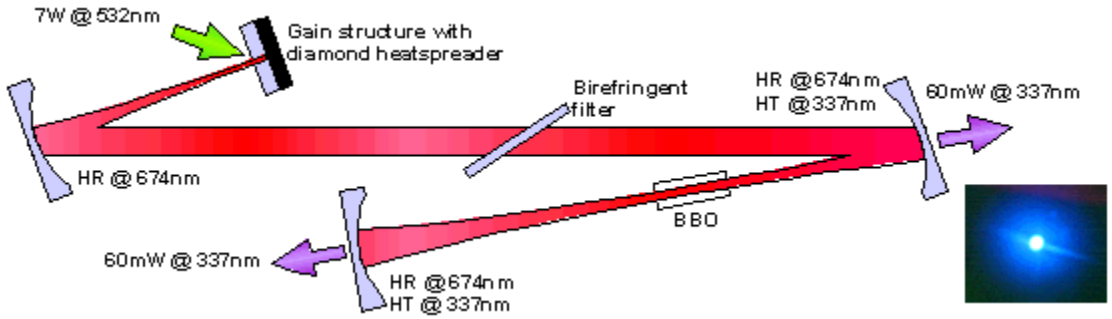
There are a number of materials commonly used for SHG or other frequency conversion effects based on $\chi^{(2)}$.

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



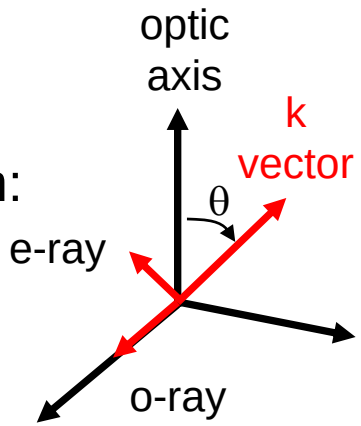
$LiNbO_3$ crystals

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

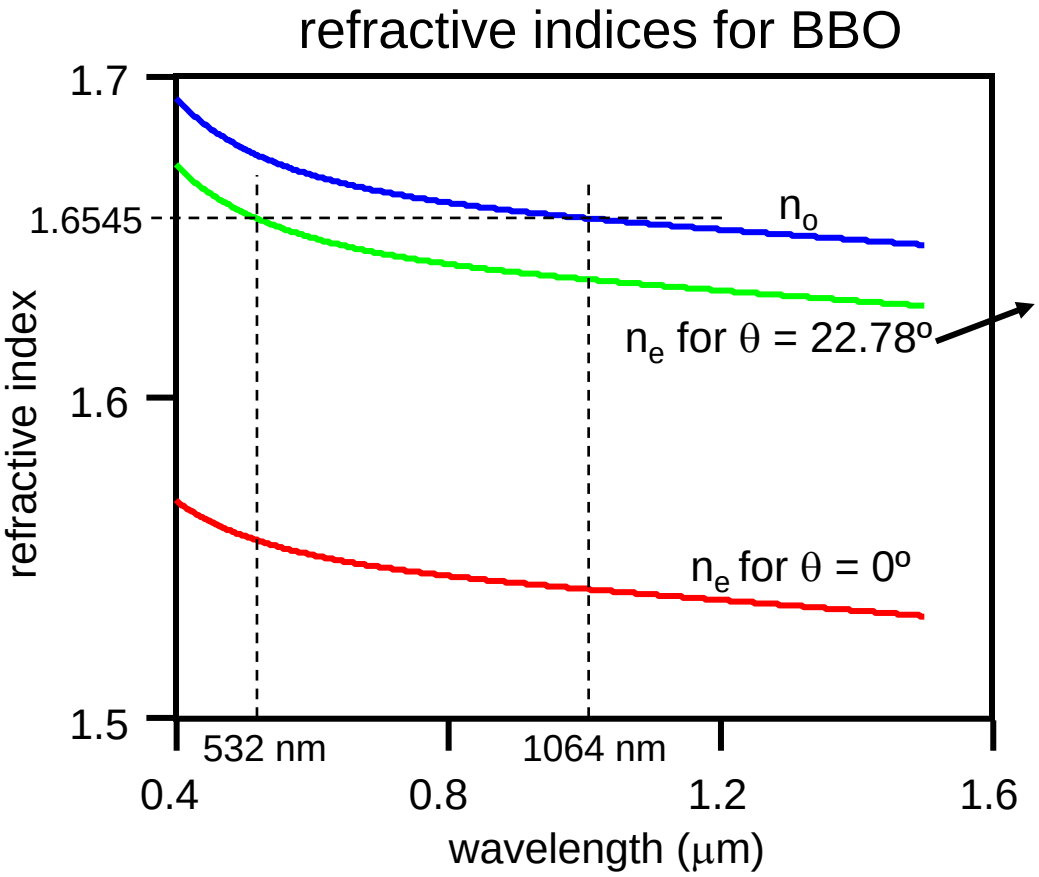


This is a “VECSEL”: a “vertical external cavity surface emitting laser”

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 $\Delta k = 0$.

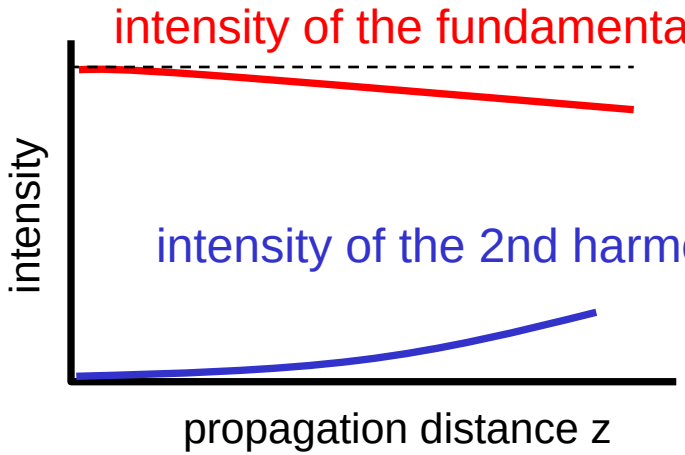
What if we changed the angle slightly? For example: 23°.

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 $\frac{\sin^2 \delta}{\delta^2} = 0.18$

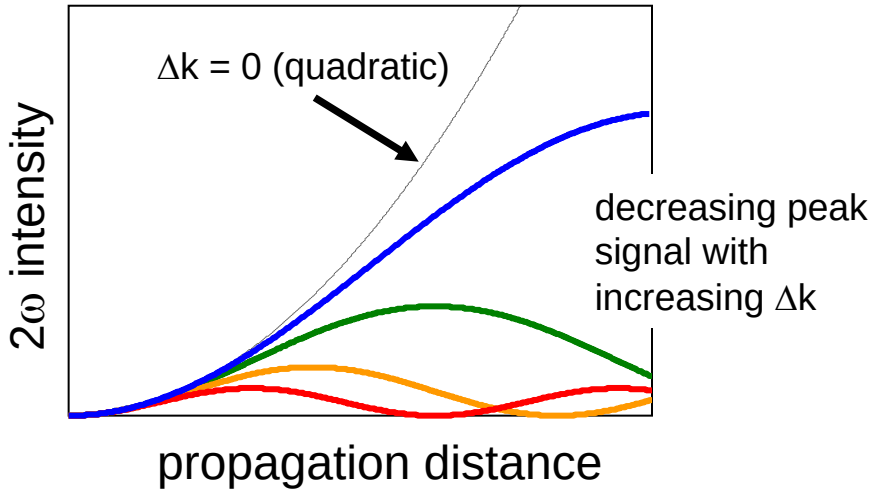
What if the phase matching is not perfect?

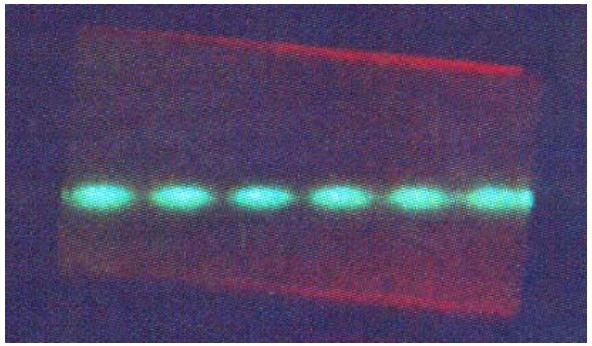


$$I_{2\omega}(z) \propto I_0^{\,2} z^2 \frac{\sin^2(\Delta k \cdot z/2)}{(\Delta k \cdot z/2)^2}$$

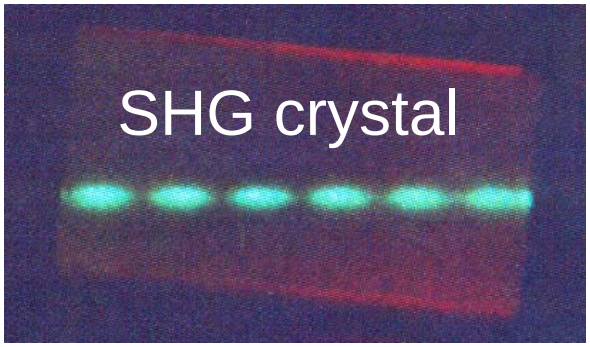
If the phase mismatch is not precisely zero, then how does the second harmonic intensity behave?

The SHG intensity oscillates as a function of propagation distance:





Another way to boost the SHG efficiency



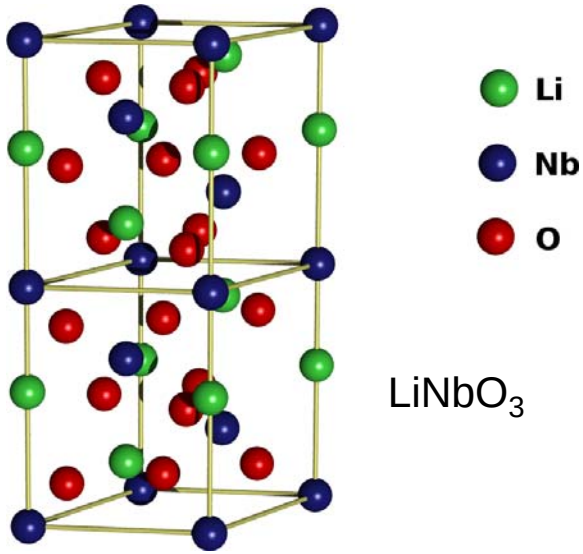
Why does the signal oscillate?

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.

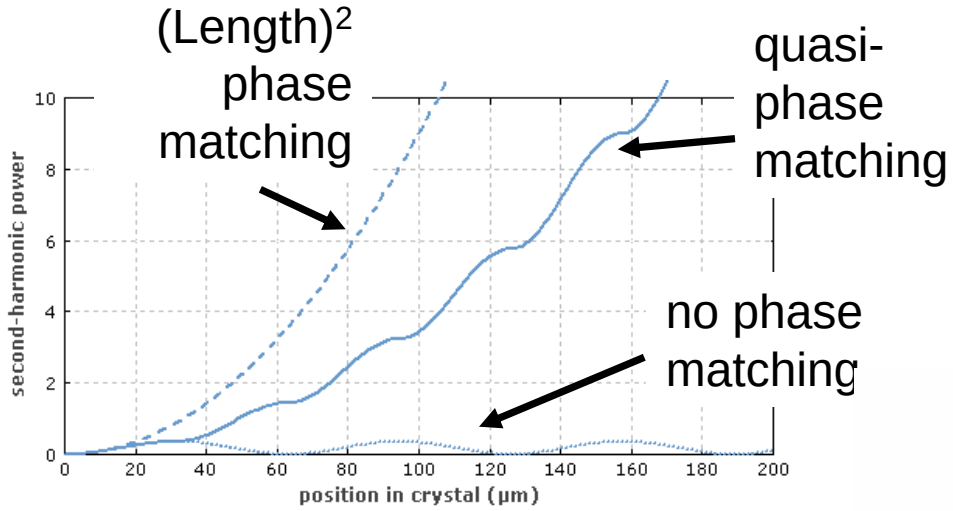
In some cases, we can control the sign of $\chi^{(2)}$ by changing the crystal structure.



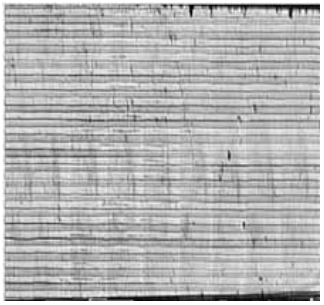
$LiNbO_3$

Quasi-phase matching

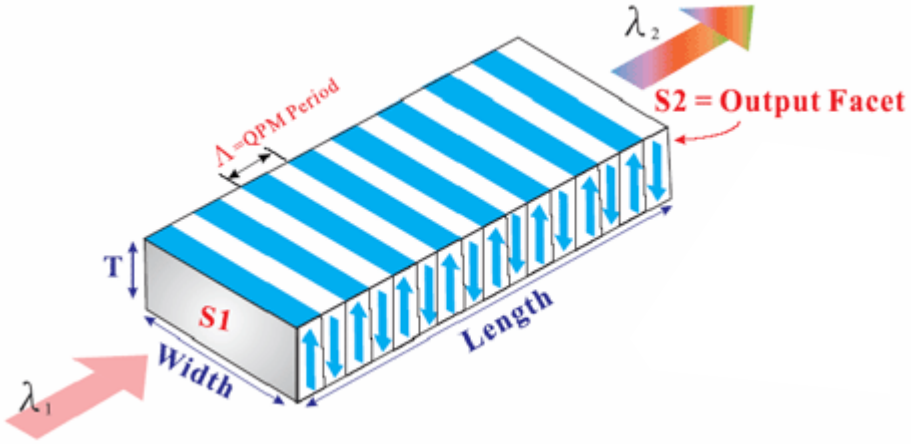
Flipping the sign of $\chi^{(2)}$ once each coherence length is known as "quasi-phase matching." It has recently become a critically important method for efficient second harmonic generation.



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

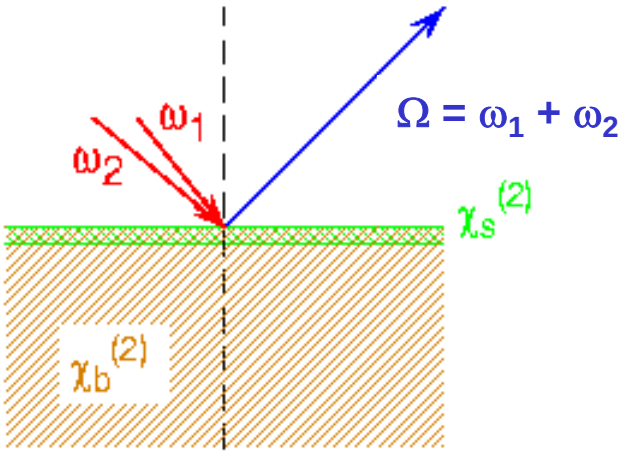


A photo of PPLN: periodically poled lithium niobate

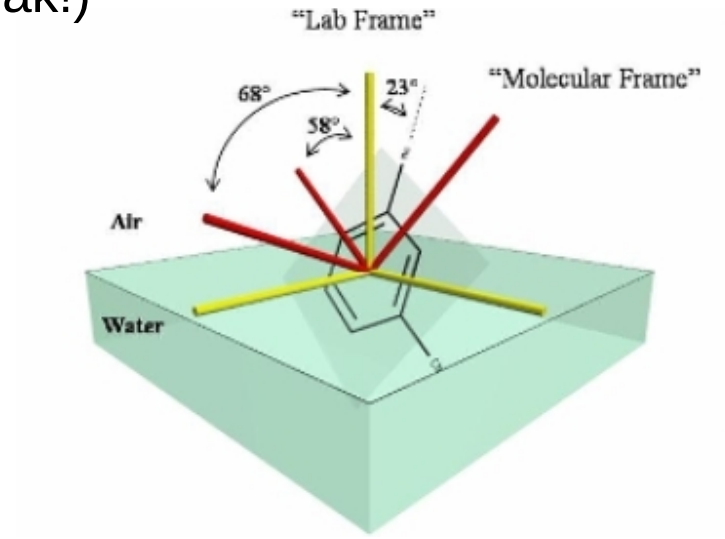


SHG at a surface

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