35. Introduction to nonlinear optics

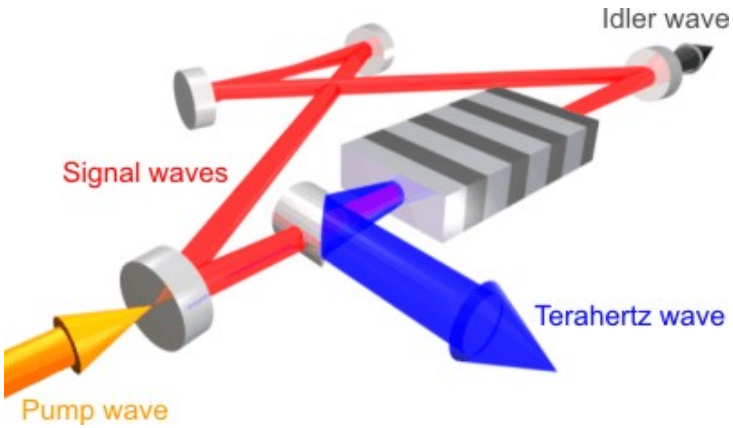
What are nonlinear-optical effects
and why do they occur?

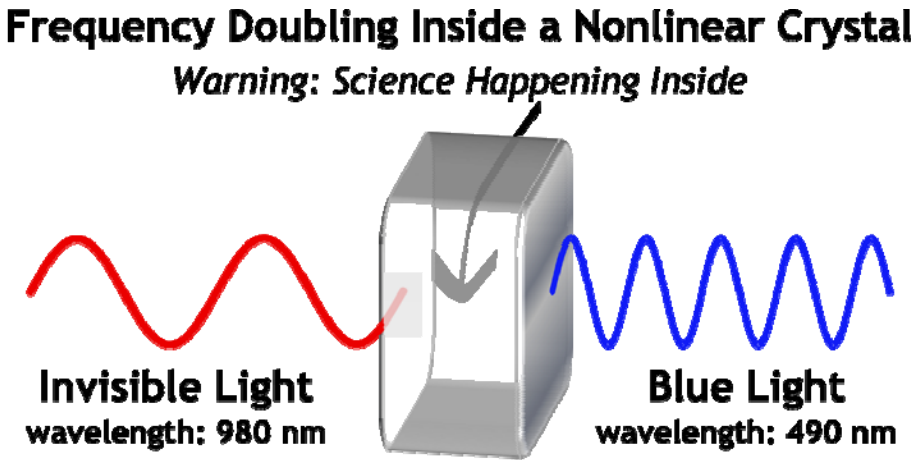
Maxwell's equations in a medium

Nonlinear-optical media

Second-harmonic generation

Conservation laws for photons
("Phase-matching")



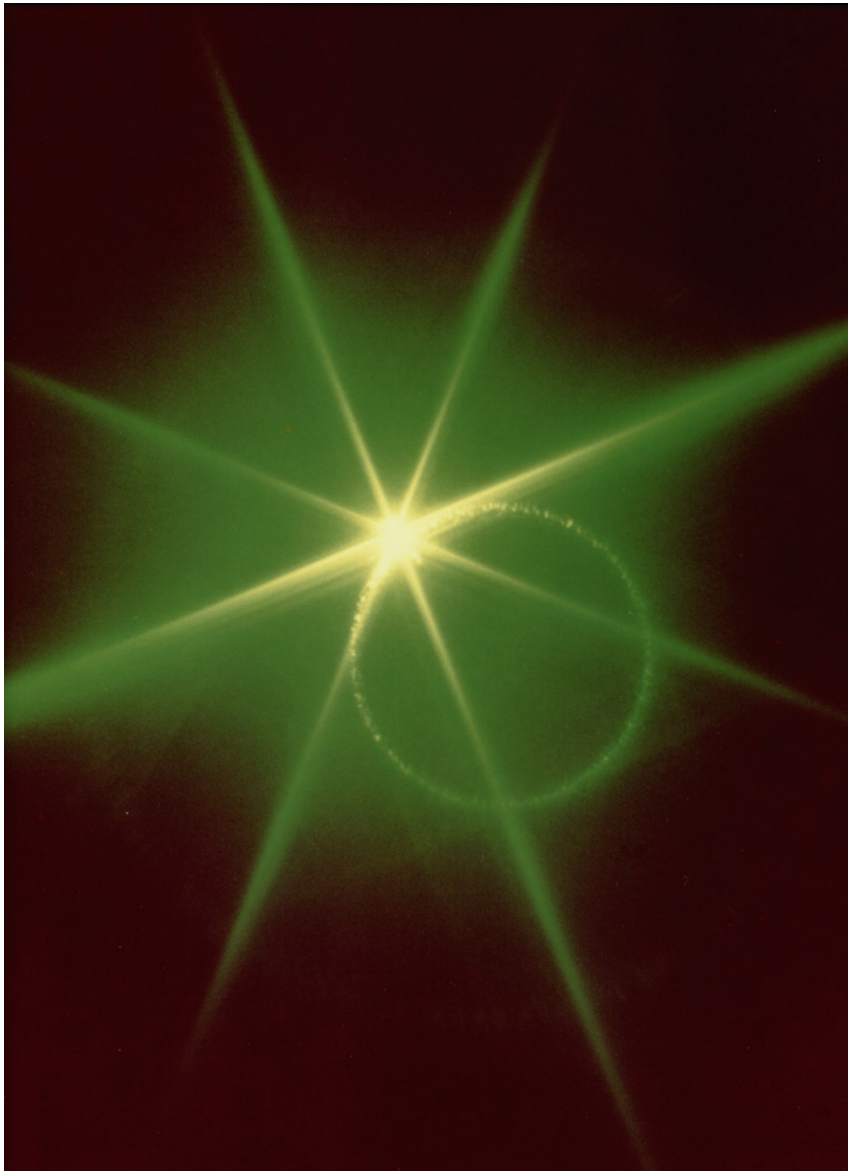


Nonlinear Optics can produce many exotic effects

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

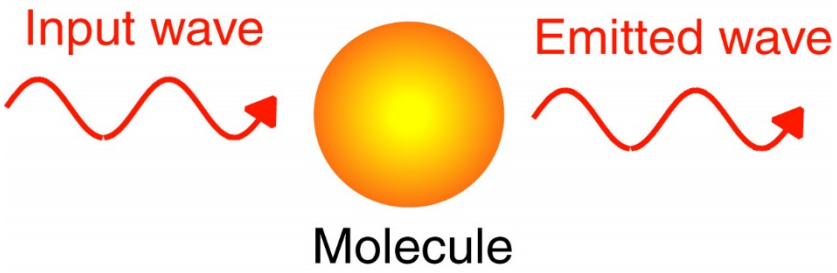
Nonlinear optics allows us to change the color of a light beam, to change its shape in space and time, and to create the shortest events ever made by humans.

Nonlinear optical phenomena are the basis of many components of optical communications systems, optical sensing, and materials research.

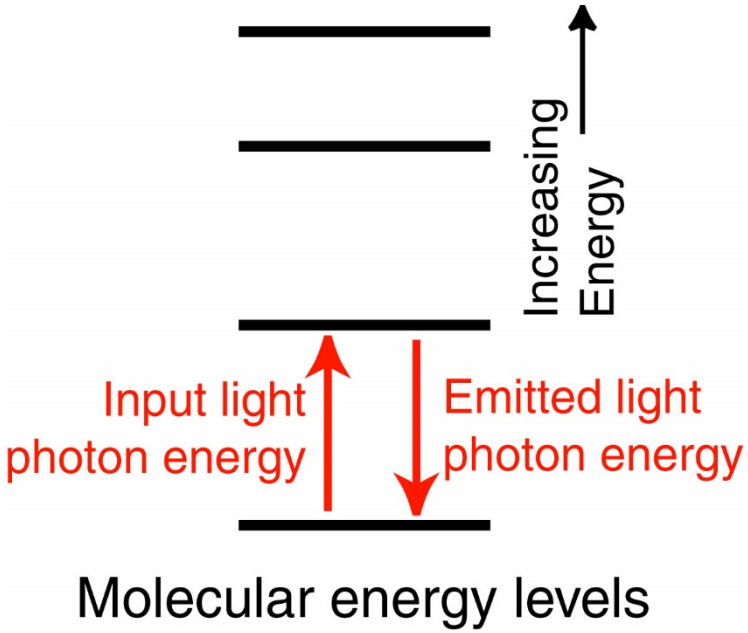


Why do nonlinear-optical effects occur?

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.

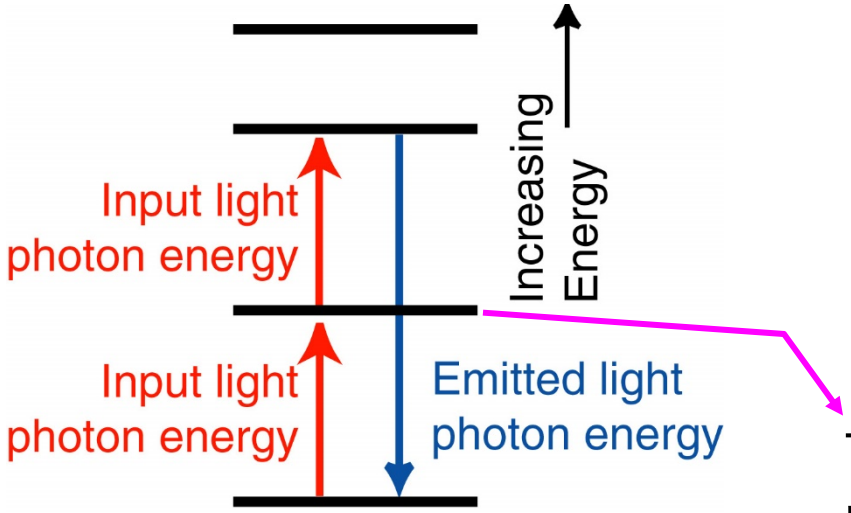


We can also imagine this process in terms of the molecular energy levels, using arrows for the photon energies:

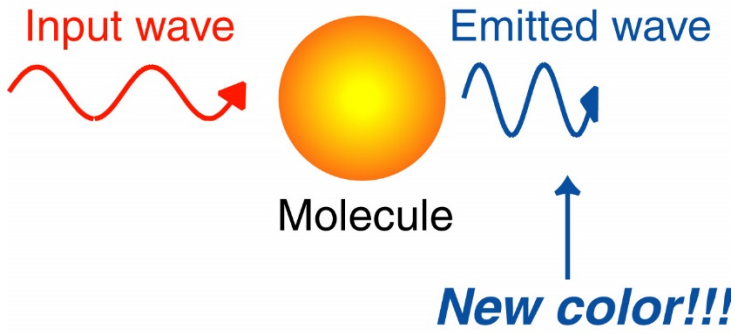


Why do nonlinear-optical effects occur? (continued)

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



This picture is a little bit misleading because it suggests that the intermediate energy level is necessary, which isn't true.

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 called the "susceptibility"

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

$$P(t) = \varepsilon_0 \frac{Ne^2 / \varepsilon_0 m_e}{2\omega_0 \left(\omega_0 - \omega - j\Gamma\right)} E(t)$$

In this particular model, this is $\chi(\omega)$.

Then, the wave equation becomes:

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

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

Reminder: Maxwell's Equations in a Medium

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

Of course this is the same equation as the usual homogeneous equation (waves in empty space), as long as we define:

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

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 (possibly complex) refractive index correction to the speed.

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 includes higher-order (nonlinear!) terms in the electric field:

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

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

Now instead of just one proportionality constant χ, we have a family of them: $\chi^{(1)}$, $\chi^{(2)}$, $\chi^{(3)}$, etc.

Then the wave equation looks 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 challenge…

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

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?

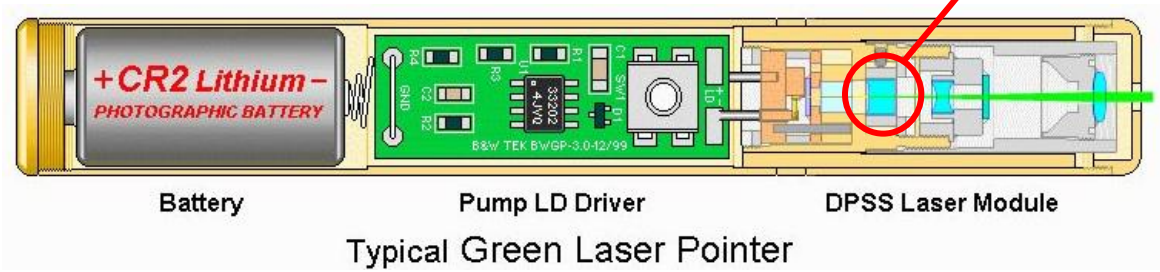
If we write the field as: $E(t) \propto E_0 \exp(j\omega t) + E_0^* \exp(-j\omega t)$

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

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

The birth of nonlinear optics

In 1961, P. Franken *et al.* focused a pulsed ruby laser (λ = 694 nm) into a quartz crystal. With about 3 joules of energy in the red pulse, they generated a few nanojoules of blue light (λ = 347 nm)

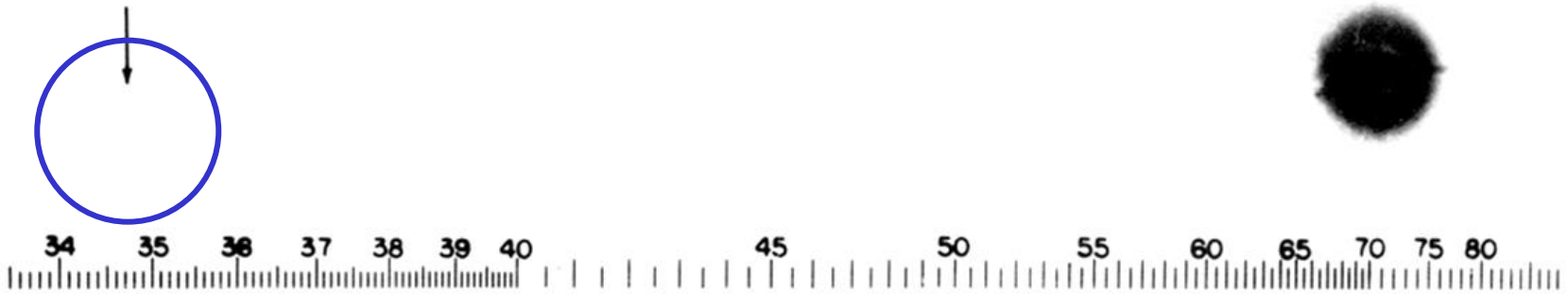


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

from Phys. Rev. Lett., 7, 118 (1961)

The copy editor thought it was a speck of dirt, and removed it….

Sum and difference frequency generation

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

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

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

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

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

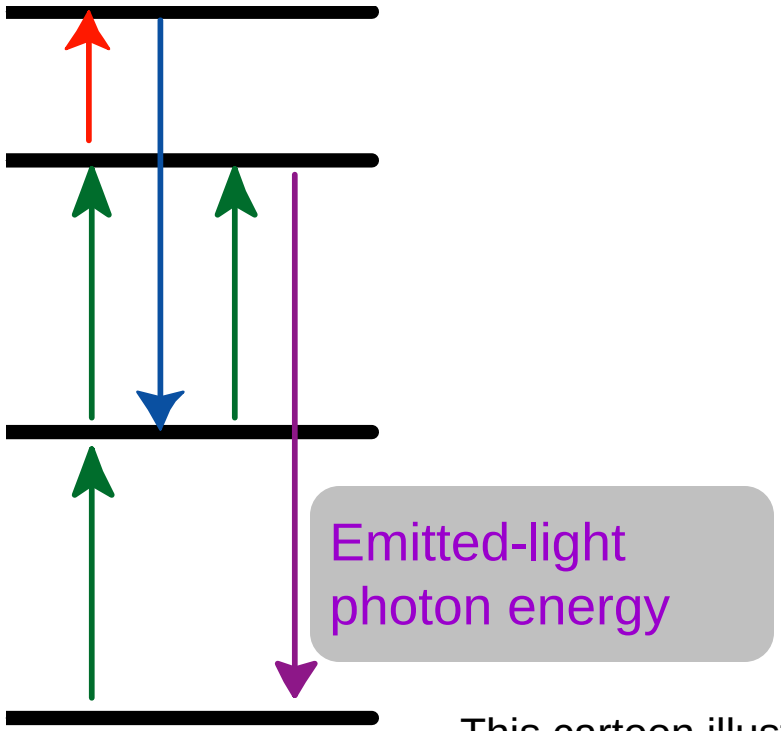
$$+ 2E_1 E_2 \exp(j[\omega_1 + \omega_2]t) + 2E_1^* E_2^* \exp(-j[\omega_1 + \omega_2]t)$$ sum frequency

$$+ 2E_1 E_2 \exp(j[\omega_1 - \omega_2]t) + 2E_1^* E_2^* \exp(-j[\omega_1 - \omega_2]t)$$ 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"

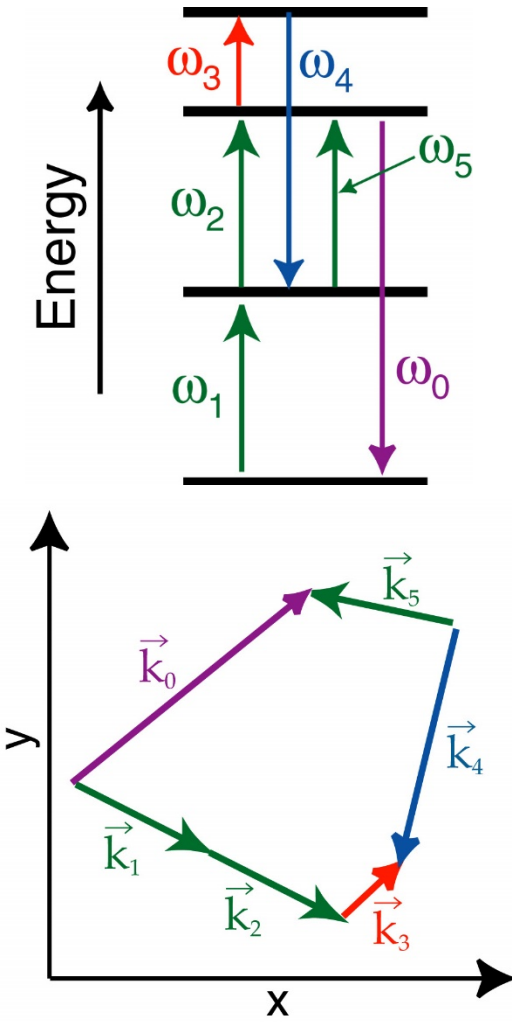
where N is the number of photons involved (including the emitted one).

Frequency doubling is a "three-wave mixing" process.

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:

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

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

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

No more than one of the many possible N-wave mixing processes can be phase-matched at any one time. Most of the time, none of them can.

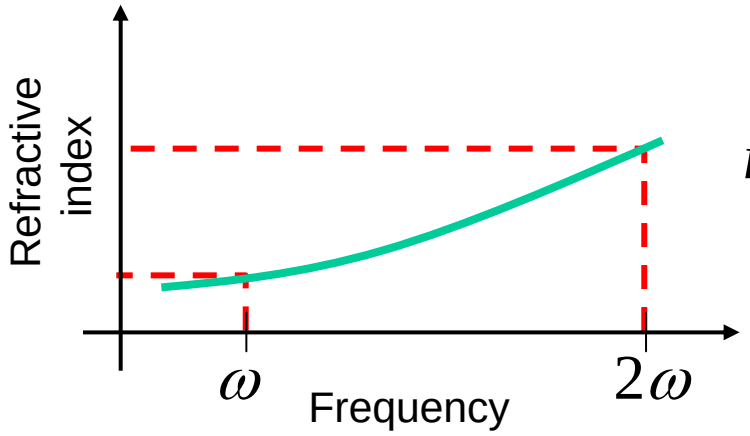
Phase-matching: an example

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



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

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

Unfortunately, dispersion prevents this from ever happening!

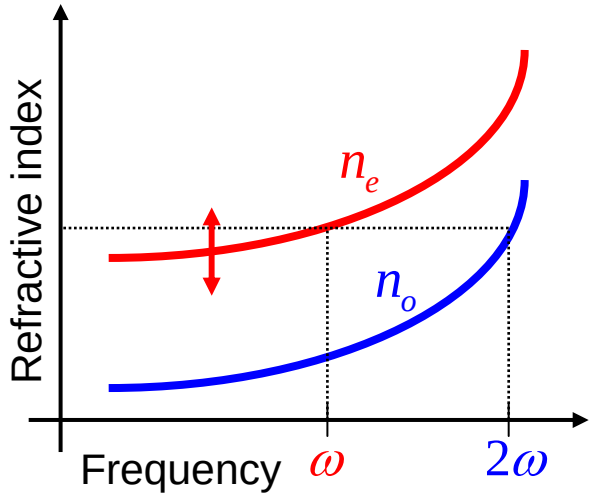
Phase-matching Second-Harmonic Generation using birefringence

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:

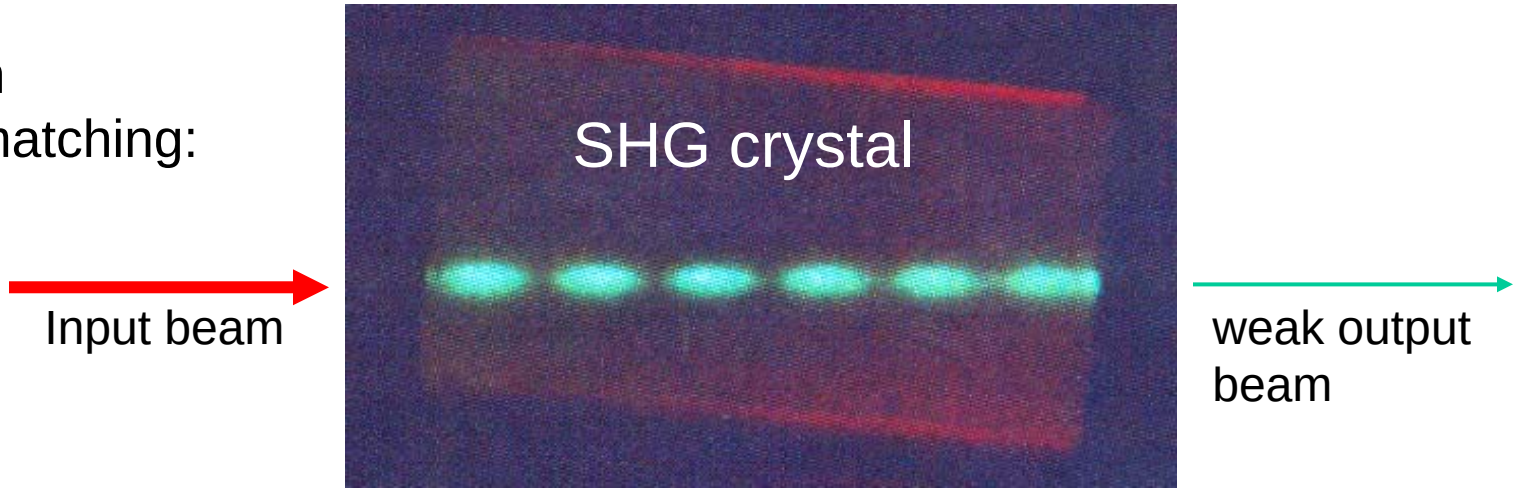


$$n_o(2\omega) = n_e(\omega)$$

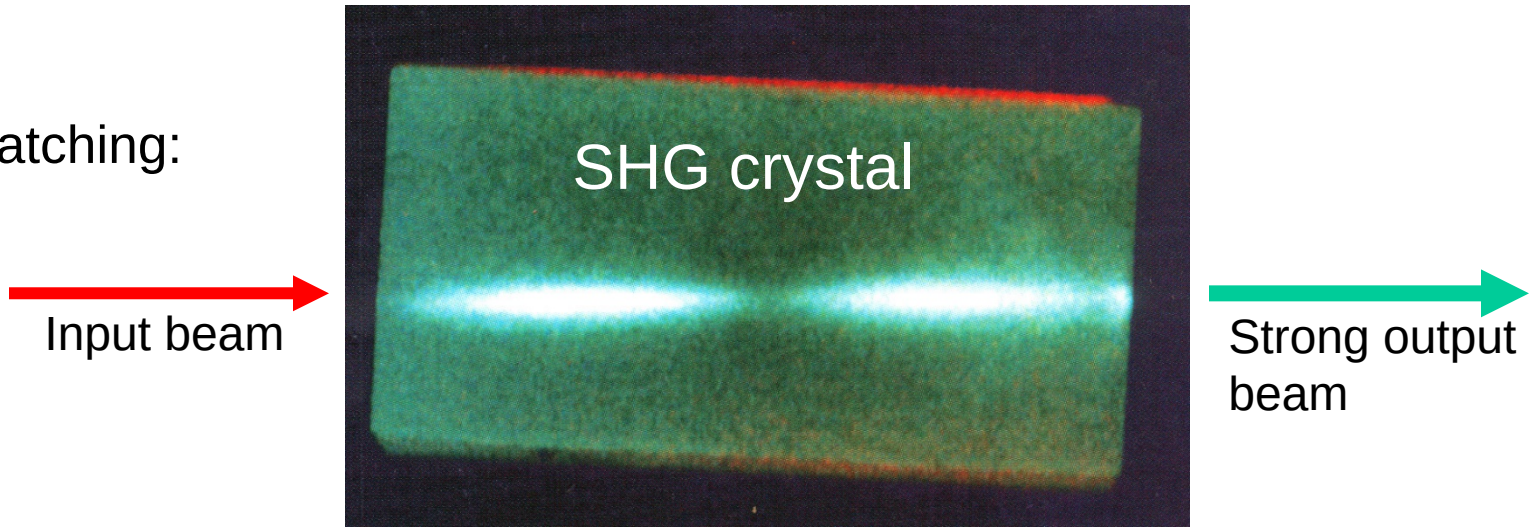
n_e depends on propagation angle, so by rotating the birefringent crystal, we can tune the condition precisely by moving the red curve up and down relative to the blue curve.

Light created in real crystals

Far from phase-matching:



Closer to phase-matching:



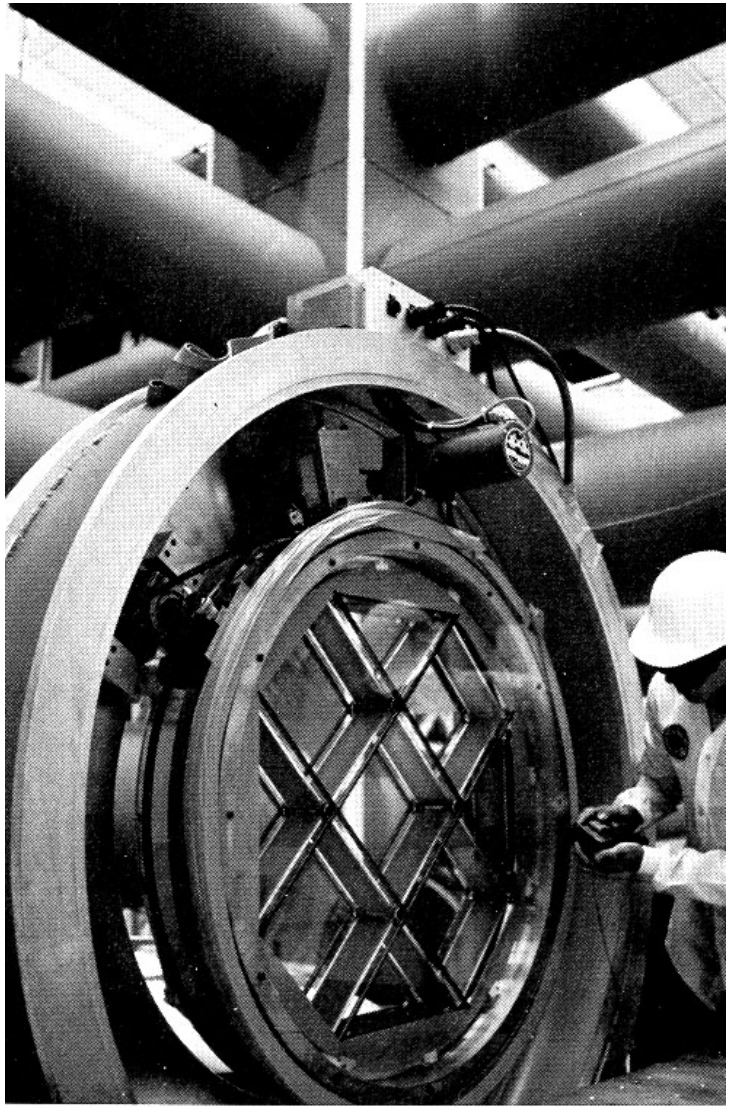
Note that SH beam is brighter as phase-matching is achieved.

Second-Harmonic Generation

SHG crystals at Lawrence Livermore National Laboratory

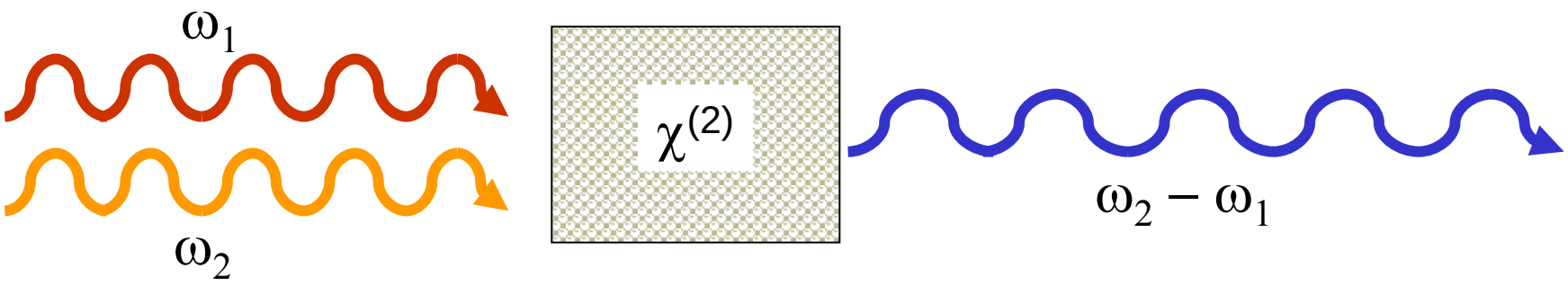
These crystals convert as much as 80% of the input light to its second harmonic. Then additional crystals produce the third harmonic with similar efficiency!

Cascading two second-order processes is usually more efficient than a single-step third-order process.

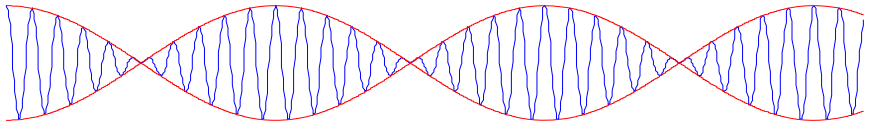


Another second-order process: difference-frequency generation

Example: consider two optical waves with similar frequencies ω_1 and ω_2:



$$\tilde{E}_{tot}(t) = \tilde{E}_0 \exp(j\omega_1 t) + \tilde{E}_0 \exp(j\omega_2 t)$$
$$= 2\tilde{E}_0 \cos(\omega_{ave} t)\cos(\Delta\omega t)$$

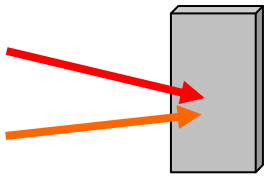


Electrons in the solid are unable to oscillate as rapidly as ω_{ave}, but they can oscillate as rapidly as $\Delta\omega$ if it is not too large.

Oscillating currents induced in the material produce radiation at $\Delta\omega$.

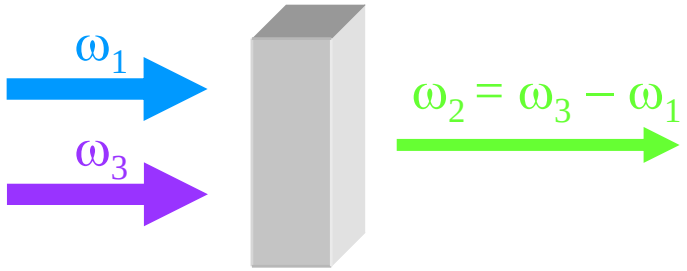
This is the origin of $\chi^{(2)}(\omega_2 - \omega_1)$ in semiconductors.

Suppose we illuminate a semiconductor with this superposition of two light waves, like this:

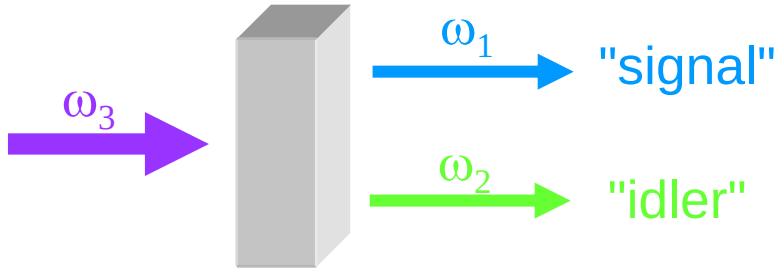


Difference-Frequency Generation: Optical Parametric Generation, Amplification, Oscillation

Difference-frequency generation takes many useful forms.

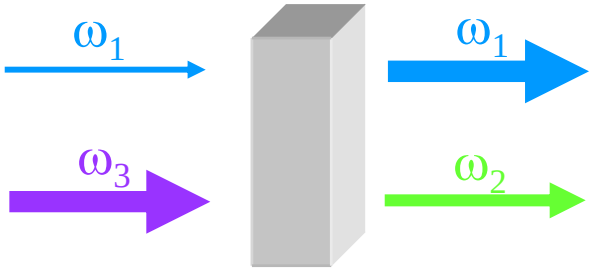


Difference-frequency generation
(also called Parametric Down-Conversion)

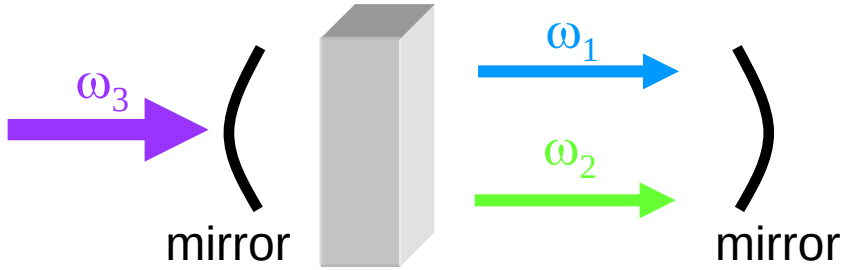


Optical Parametric
Generation (OPG)

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



Optical Parametric
Amplification (OPA)



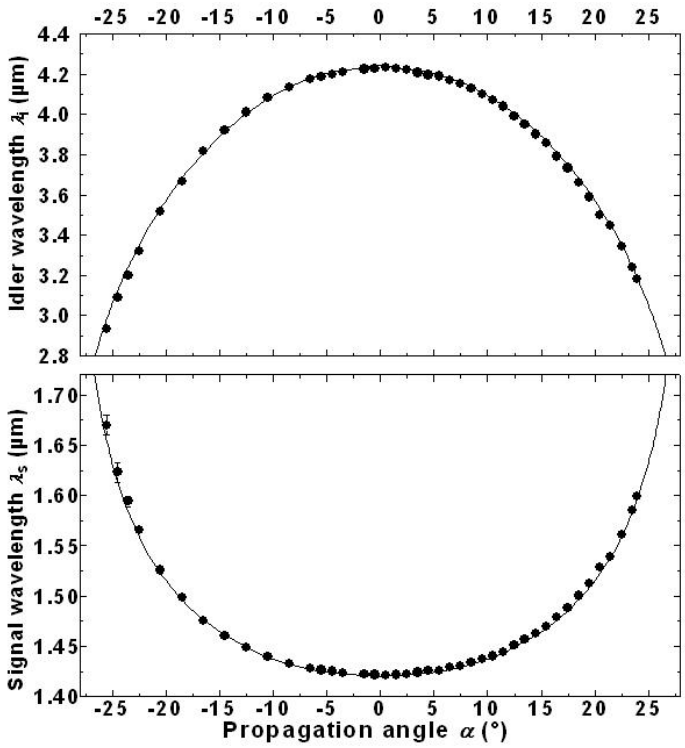
Optical Parametric
Oscillation (OPO)

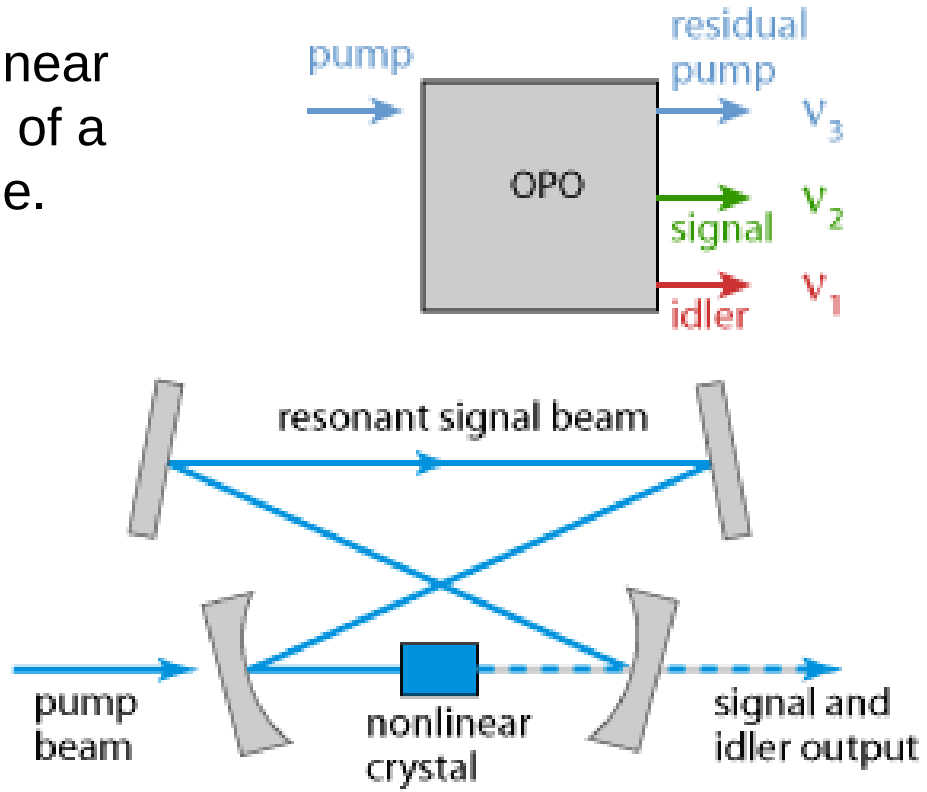
All of these are $\chi^{(2)}$ processes (three-wave mixing).

Optical Parametric Oscillator (OPO)

Like a laser, but much more widely tunable!

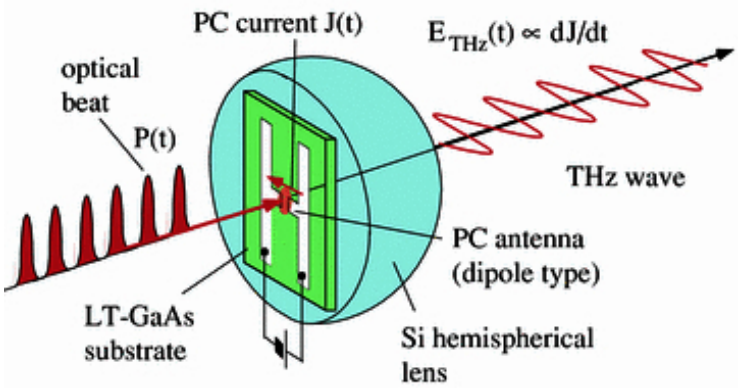
No energy is stored in the nonlinear crystal (unlike the gain medium of a laser), so heating is not an issue.



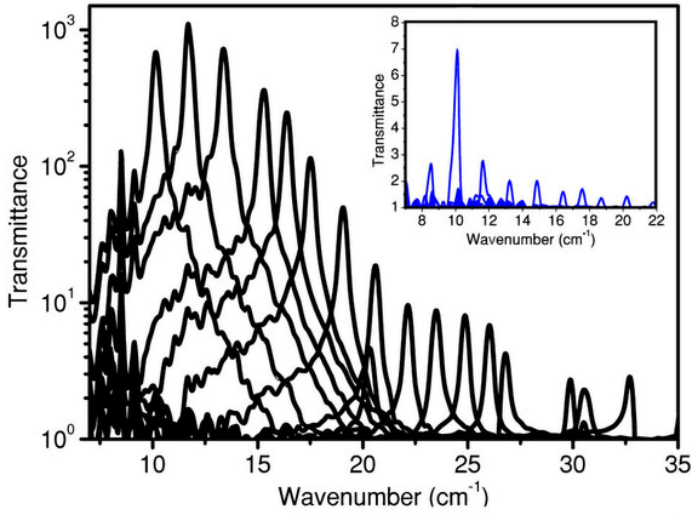


A commonly used and powerful method for generating tunable near- and mid-infrared light.

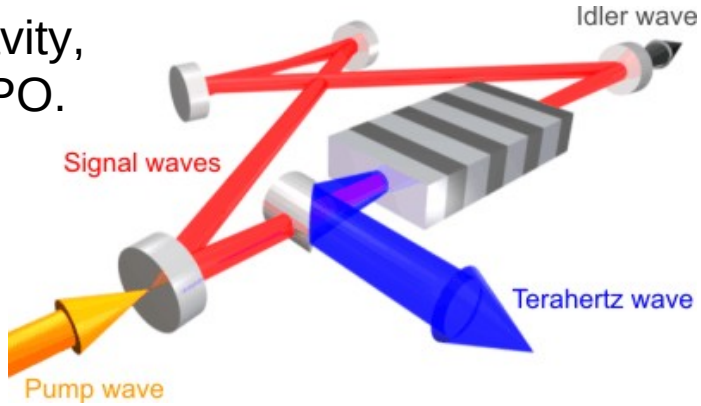
DFG is one common method for generation of terahertz radiation



Changing $\Delta\omega$ by tuning one laser results in tuning of the output THz frequency.

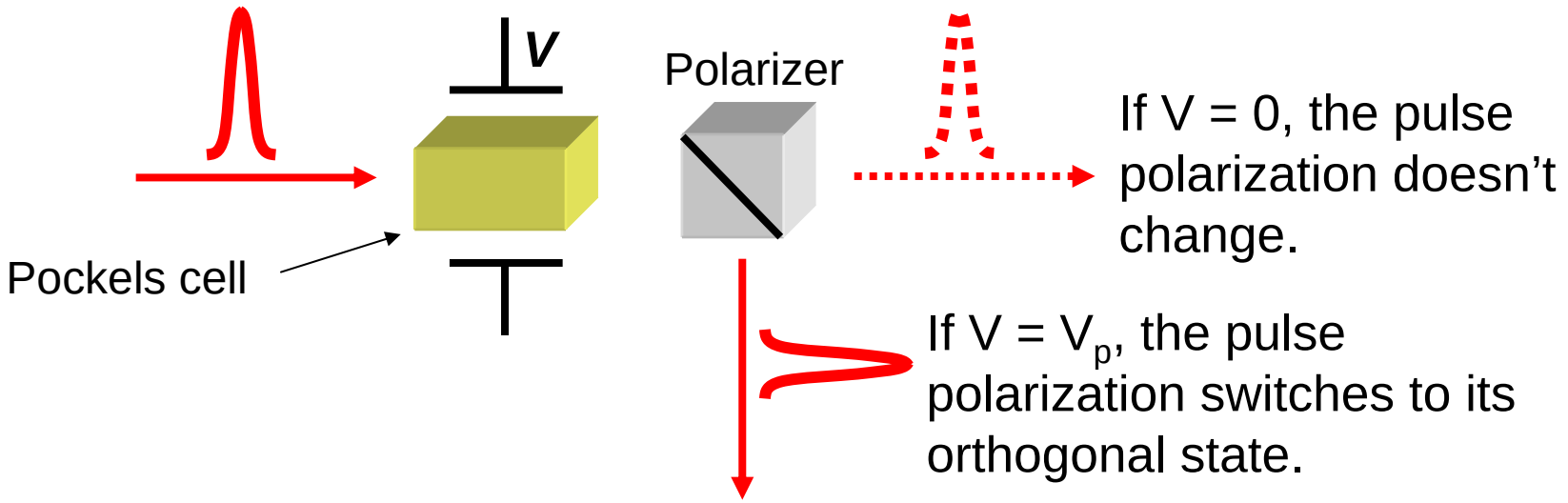


One can even put this DFG process into a laser cavity, creating a terahertz OPO.



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 $\omega + 0 = \omega$.

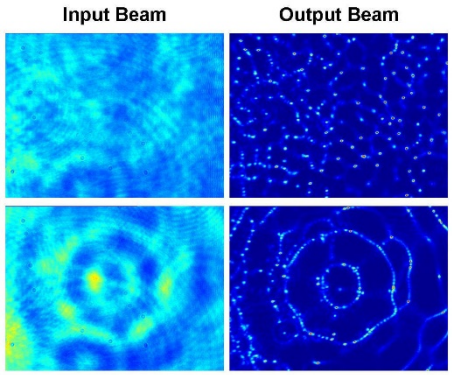
3rd order effects can also be important

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

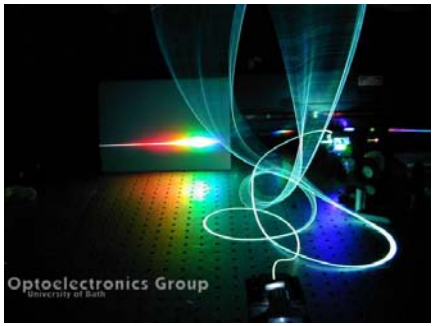
In certain specific situations, one can find that $\chi^{(2)}$ is identically zero.

In these cases, the largest non-linear contribution comes from the $\chi^{(3)}$ term.

- third harmonic generation
- other more unusual effects:

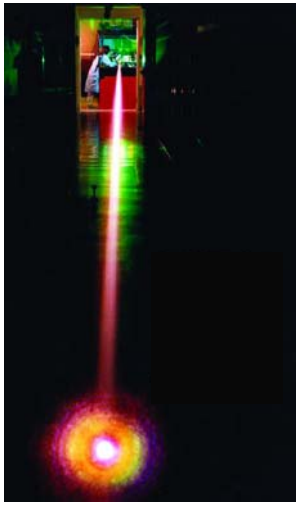


self focusing



supercontinuum generation


spatial solitons


optical filaments