

Nonlinear optics

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

Maxwell's equations in a medium

Nonlinear-optical media

Second-harmonic generation

Sum- and difference frequency generation

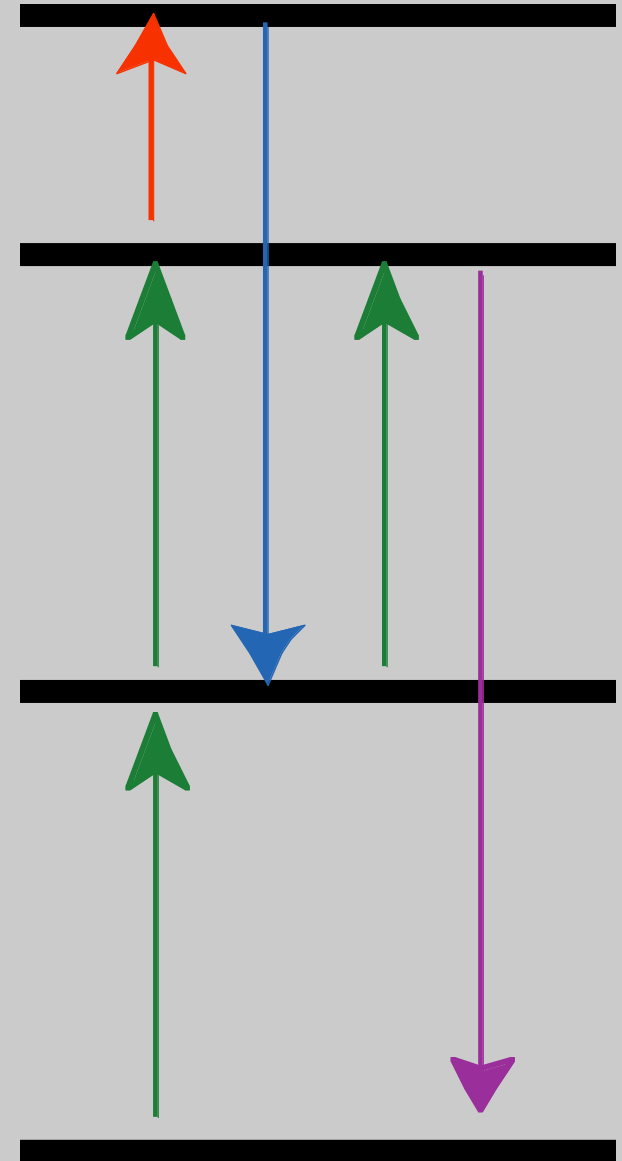
Conservation laws for photons ("Phase-matching")

Induced gratings

Phase conjugation and aberration cancellation

Holography

Self-phase modulation



Slides courtesy of R. Trebino (Ga Tech)

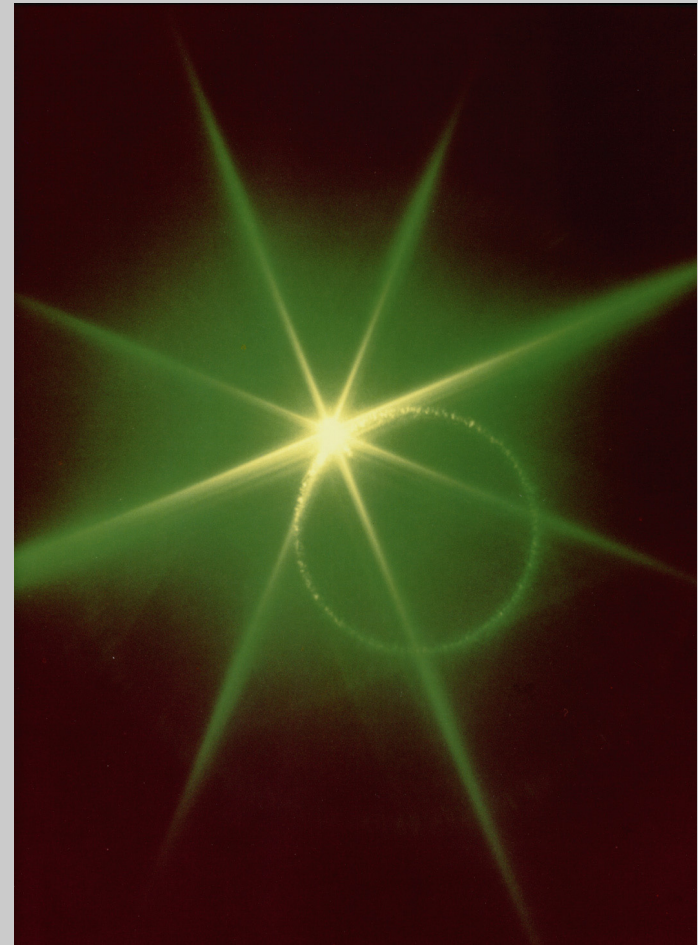
Nonlinear optics isn't something you see everyday.

Sending infrared light into a crystal yielded this display of green light (second-harmonic generation):

Nonlinear optics allows us to change the color of a light beam, to change its shape in space and time, and to create ultrashort laser pulses.

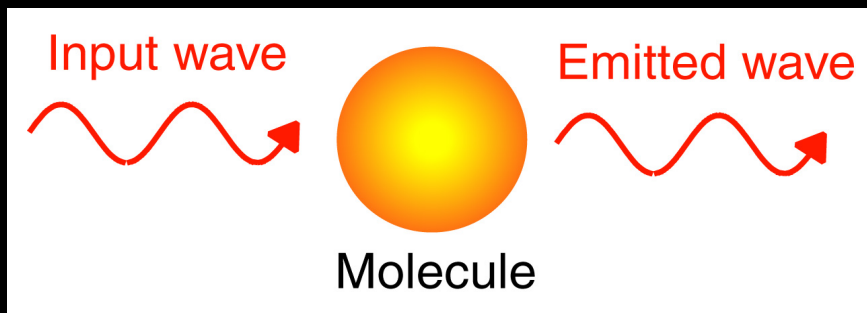
Why don't we see nonlinear optical effects in our daily life?

1. Intensities of daily life are too weak.
2. Normal light sources are incoherent.
3. The occasional crystal we see has the wrong symmetry (for SHG).
4. "Phase-matching" is required, and it doesn't usually happen on its own.

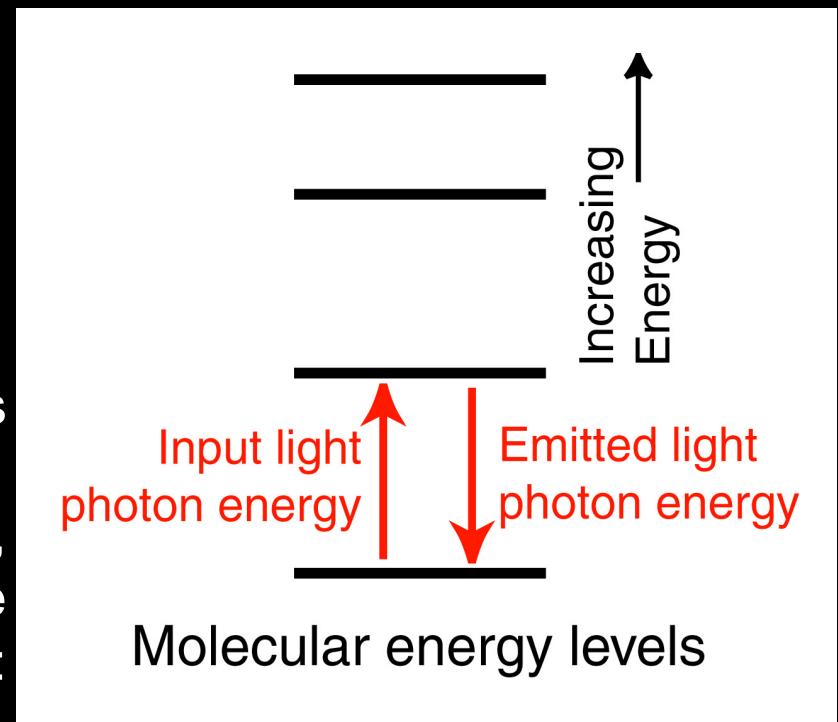


Why do nonlinear-optical effects occur?

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.

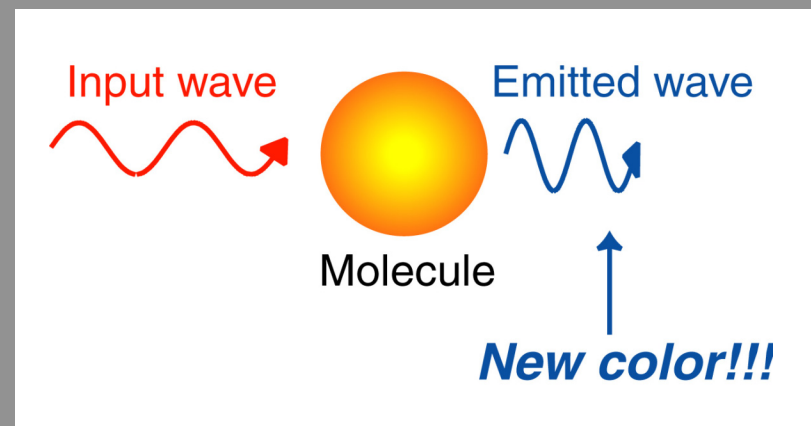
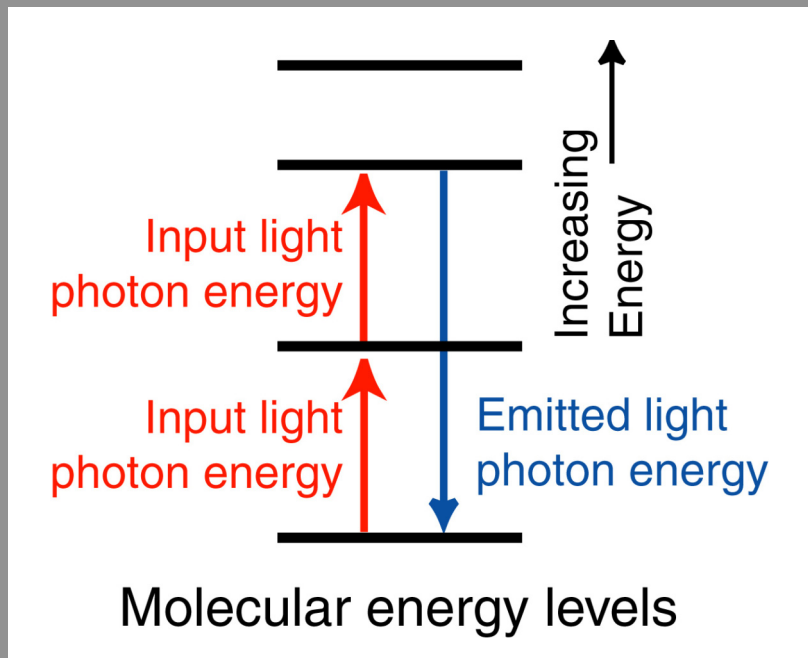


We can also imagine this process in terms of the molecular energy levels, using arrows for the photon energies:



Why do nonlinear-optical effects occur?

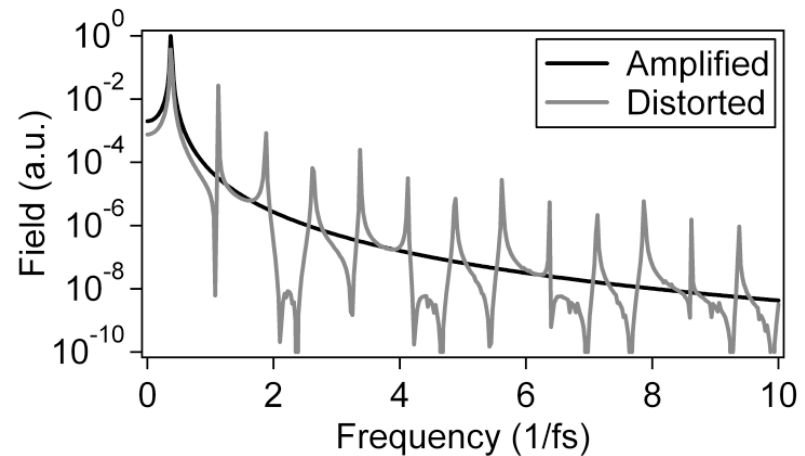
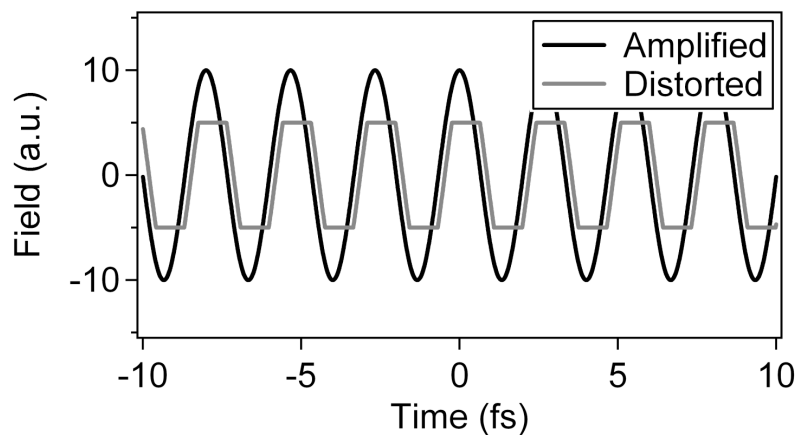
Now, suppose the irradiance is high enough that many molecules are excited to the higher-energy state. This state can then act as the lower level for additional excitation. This yields vibrations at all frequencies corresponding to all energy differences between populated states.



Nonlinear optics is analogous to nonlinear electronics, which we can observe easily.

Sending a high-volume sine-wave (“pure frequency”) signal into cheap speakers yields a truncated output signal, more of a square wave than a sine.

This square wave has higher frequencies: “harmonics”.

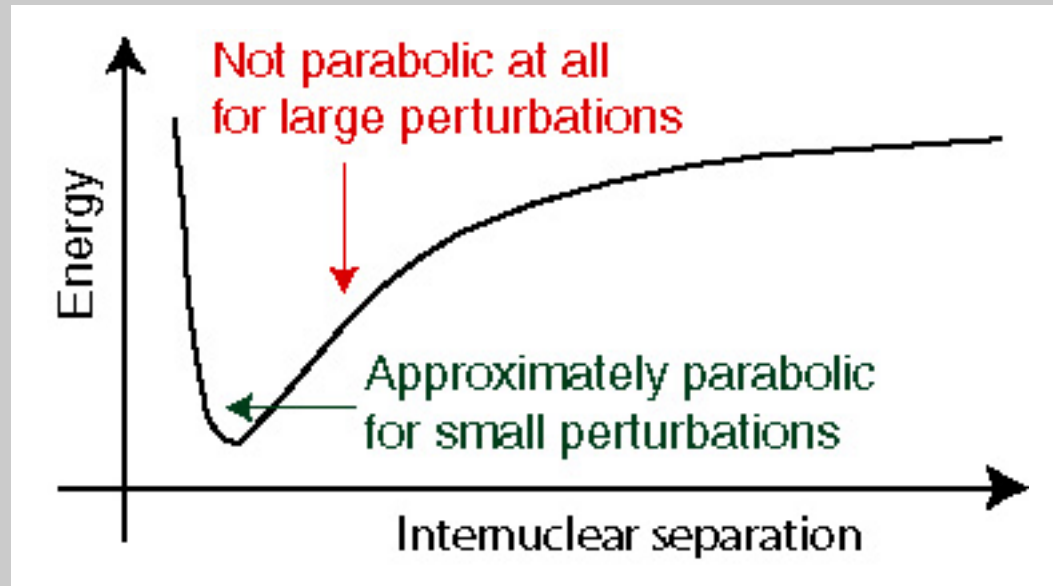


We hear this as distortion.

Nonlinear optics and anharmonic oscillators

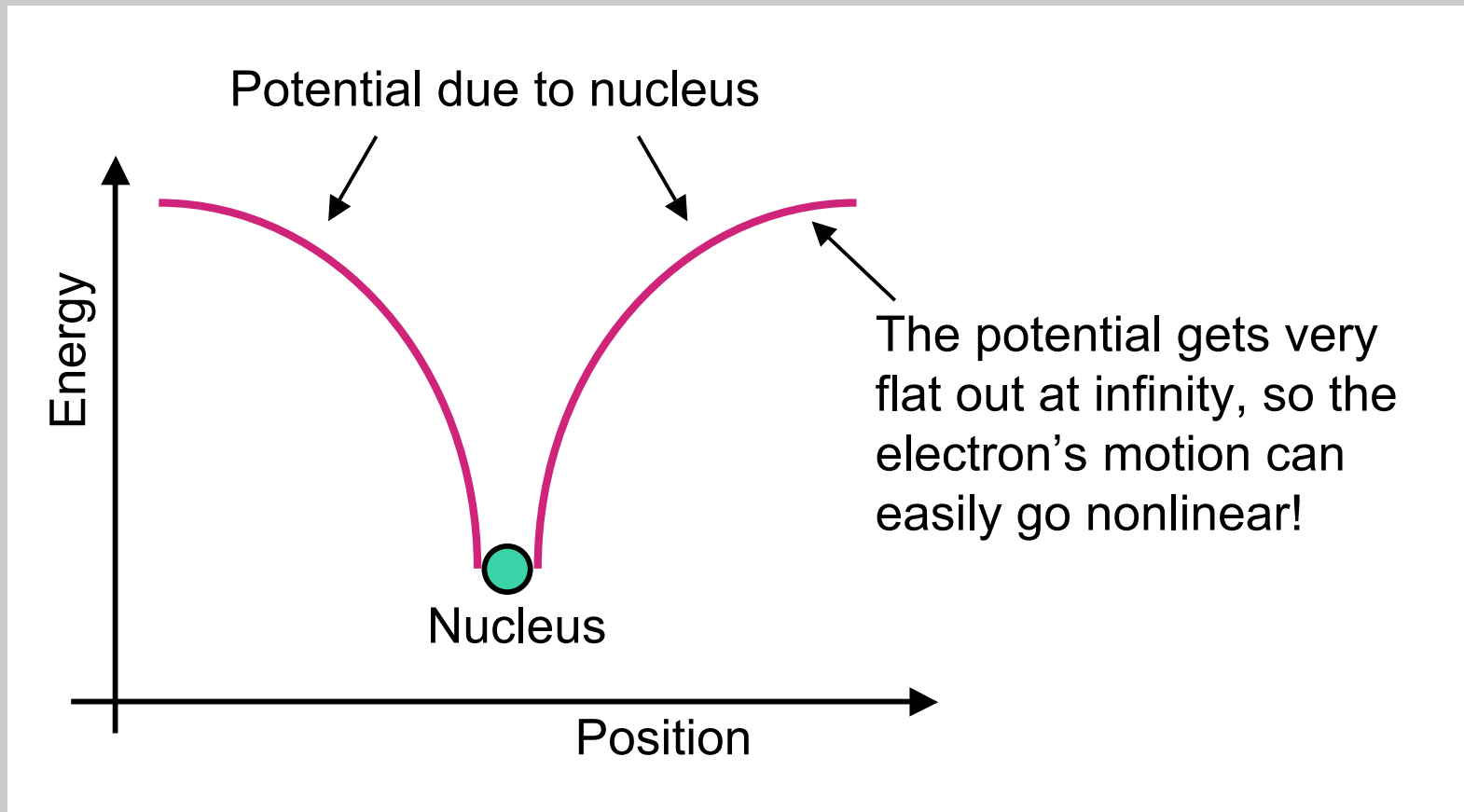
Another way to look at nonlinear optics is that the potential of the electron or nucleus (in a molecule) is not a simple harmonic potential.

Example: vibrational motion:



For weak fields, motion is harmonic, and linear optics prevails.
For strong fields (i.e., lasers), anharmonic motion occurs, and higher harmonics occur, both in the motion and the light emission.

Nonlinear effects in atoms and molecules



So an electron's motion will also depart from a sine wave.

Maxwell's Equations in a Medium

- The induced polarization, \mathcal{P} , contains the effect of the medium:

$$\begin{aligned}\vec{\nabla} \cdot \vec{\mathcal{E}} &= 0 & \vec{\nabla} \times \vec{\mathcal{E}} &= -\frac{\partial \vec{\mathcal{B}}}{\partial t} \\ \vec{\nabla} \cdot \vec{\mathcal{B}} &= 0 & \vec{\nabla} \times \vec{\mathcal{B}} &= \frac{1}{c_0^2} \frac{\partial \vec{\mathcal{E}}}{\partial t} + \mu_0 \frac{\partial \vec{\mathcal{P}}}{\partial t}\end{aligned}$$

These equations reduce to the (scalar) wave equation:

$$\frac{\partial^2 \mathcal{E}}{\partial z^2} - \frac{1}{c_0^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathcal{P}}{\partial t^2}$$

“Inhomogeneous
Wave Equation”

- Sine waves of all frequencies are solutions to the wave equation; it's the polarization that tells which frequencies will occur.
- The polarization is the driving term for the solution to this equation.

Solving the wave equation in the presence of linear induced polarization

For low irradiances, the polarization is proportional to the incident field:

$$\mathcal{P} = \epsilon_0 \chi \mathcal{E}$$

In this simple (and most common) case, the wave equation becomes:

$$\frac{\partial^2 \mathcal{E}}{\partial z^2} - \frac{1}{c_0^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = \frac{1}{c_0^2} \chi \frac{\partial^2 \mathcal{E}}{\partial t^2} \quad \text{Using the fact that:}$$
$$\epsilon_0 \mu_0 = 1/c_0^2$$

Simplifying:

$$\frac{\partial^2 \mathcal{E}}{\partial z^2} - \frac{1 + \chi}{c_0^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = 0$$

This equation has the solution: $\mathcal{E}(z, t) \propto E(0) \cos(\omega t - k z)$

where $\omega = c k$ and $c = c_0/n$ and $n = (1 + \chi)^{1/2}$

The induced polarization only changes the refractive index. Dull.
If only the polarization contained other frequencies...

Maxwell's Equations in a *Nonlinear* Medium

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

$$\mathcal{P} = \epsilon_0 \left[\chi^{(1)} \mathcal{E} + \chi^{(2)} \mathcal{E}^2 + \chi^{(3)} \mathcal{E}^3 + \dots \right]$$

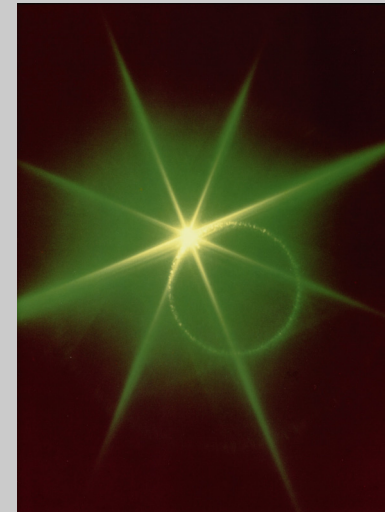
What are the effects of such nonlinear terms?
Consider the second-order term:

Since $\mathcal{E}(t) \propto E \exp(-i\omega t) + E^* \exp(i\omega t)$,

$$\mathcal{E}(t)^2 \propto E^2 \exp(-2i\omega t) + 2|E|^2 + E^{*2} \exp(2i\omega t)$$

$2\omega = 2\text{nd harmonic!}$

Harmonic generation is one of many exotic effects that can arise!



Sum- and difference-frequency generation

Suppose there are two different-color beams present:

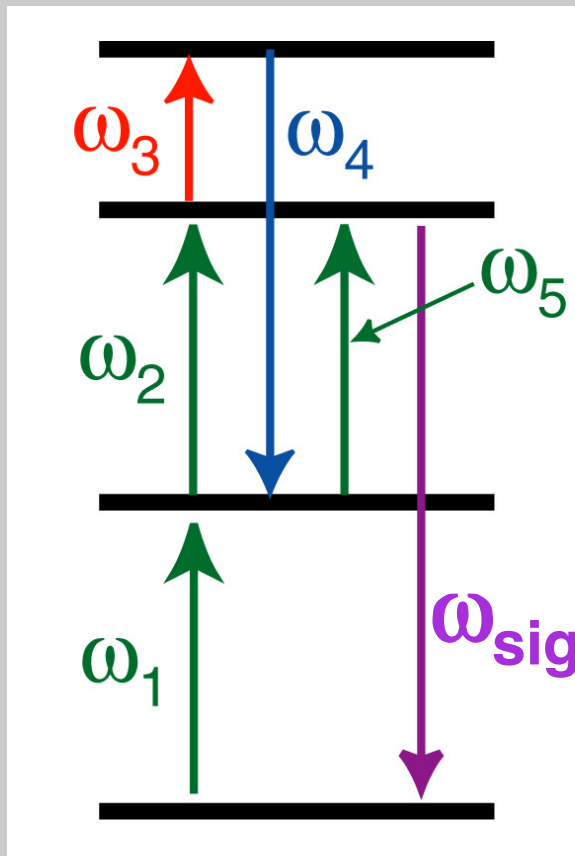
$$E(t) \propto E_1 \exp(-i\omega_1 t) + E_1^* \exp(i\omega_1 t) + E_2 \exp(-i\omega_2 t) + E_2^* \exp(i\omega_2 t)$$

So:

$$\begin{aligned} E(t)^2 \propto & E_1^2 \exp(-2i\omega_1 t) + E_1^{*2} \exp(2i\omega_1 t) && \text{2nd-harmonic gen} \\ & + E_2^2 \exp(-2i\omega_2 t) + E_2^{*2} \exp(2i\omega_2 t) && \text{2nd-harmonic gen} \\ & + 2E_1 E_2 \exp[-i(\omega_1 + \omega_2)t] + 2E_1^* E_2^* \exp[i(\omega_1 + \omega_2)t] && \text{Sum-freq gen} \\ & + 2E_1 E_2^* \exp[-i(\omega_1 - \omega_2)t] + 2E_1^* E_2 \exp[i(\omega_1 - \omega_2)t] && \text{Diff-freq gen} \\ & + 2|E_1|^2 + 2|E_2|^2 && \text{dc rectification} \end{aligned}$$

Note also that, when ω_i is positive inside the exp, the E in front has a $*$.

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 is a six-wave-mixing process.

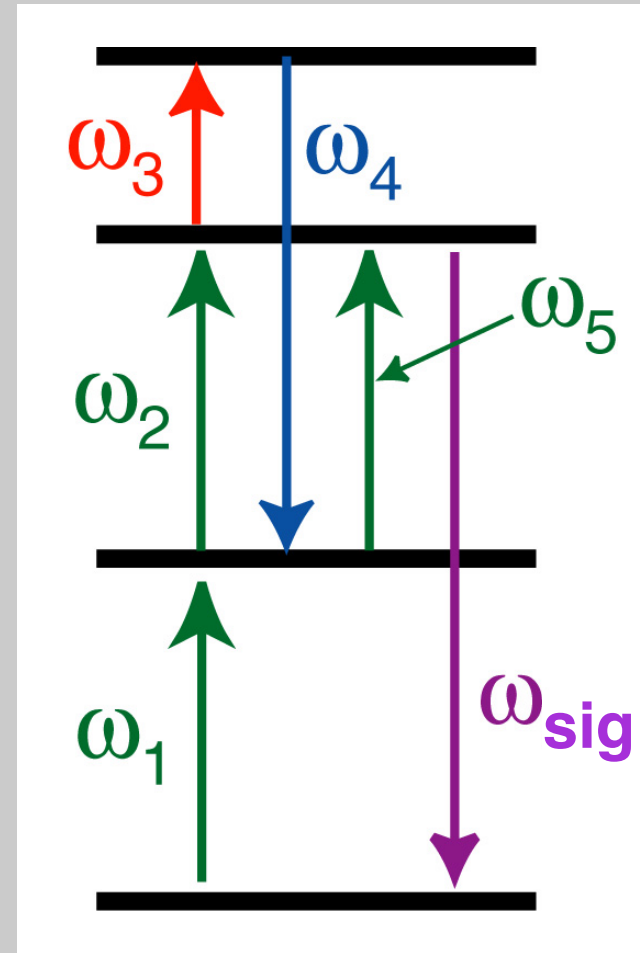
Emitted-light frequency

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. Also, if the photon energies coincide with the medium's energy levels as above, the effect will be stronger.

Induced polarization for nonlinear optical effects

Arrows pointing upward correspond to absorbed photons and contribute a factor of their field, E_i ; arrows pointing downward correspond to emitted photons and contribute a factor the complex conjugate of their field:

$$\mathcal{P} = \epsilon_0 \chi^{(5)} E_1 E_2 E_3 E_4^* E_5$$



Solving the wave equation in nonlinear optics

Recall the inhomogeneous wave equation:

$$\frac{\partial^2 \mathcal{E}}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathcal{P}}{\partial t^2}$$

Take into account the linear polarization by replacing c_0 with c .

Because it's second-order in both space and time, and \mathcal{P} is a nonlinear function of \mathcal{E} , we can't easily solve this equation. Indeed, nonlinear differential equations are really hard.

We'll have to make approximations...

- Slowly-varying envelope approximation:

$$\mathcal{E}(z, t) = A(z) B(t) \exp\left[i(kz - \omega_0 t)\right] + c.c.$$

Separation-of-frequencies approximation

The total E-field will contain several nearly discrete frequencies, ω_1, ω_2 , etc.

So we'll write separate (coupled) wave equations for each frequency, considering only the induced polarization at the given frequency:

$$\frac{\partial^2 \mathcal{E}_1}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 \mathcal{E}_1}{\partial t^2} = \mu_0 \frac{\partial^2 \mathcal{P}_1}{\partial t^2}$$

where \mathcal{E}_1 and \mathcal{P}_1 are the E-field and polarization at frequency ω_1 .

$$\frac{\partial^2 \mathcal{E}_2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 \mathcal{E}_2}{\partial t^2} = \mu_0 \frac{\partial^2 \mathcal{P}_2}{\partial t^2}$$

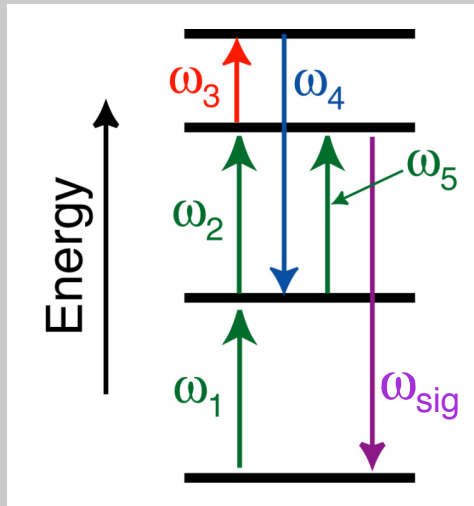
where \mathcal{E}_2 and \mathcal{P}_2 are the E-field and polarization at frequency ω_2 .

etc.

This will be a reasonable approximation even for relatively broadband ultrashort pulses

Phase-matching

Conservation laws for photons in nonlinear optics



Adding the frequencies:

$$\omega_1 + \omega_2 + \omega_3 - \omega_4 + \omega_5 = \omega_{sig}$$

is the same as energy conservation if we multiply both sides by \hbar :

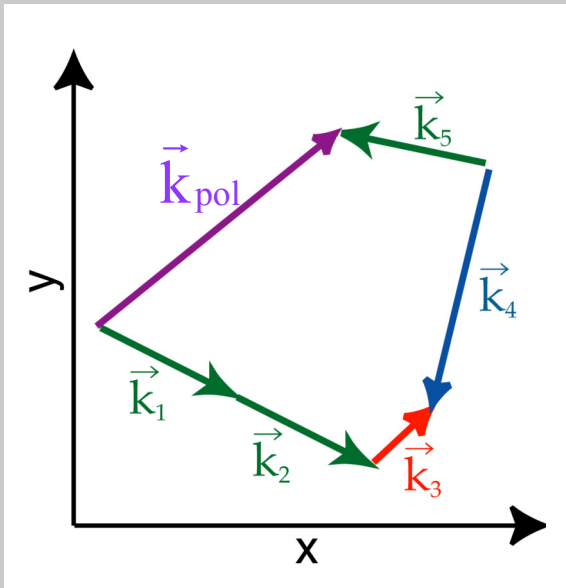
$$\hbar\omega_1 + \hbar\omega_2 + \hbar\omega_3 - \hbar\omega_4 + \hbar\omega_5 = \hbar\omega_{sig}$$

Adding the k 's conserves momentum:

$$\vec{k}_1 + \vec{k}_2 + \vec{k}_3 - \vec{k}_4 + \vec{k}_5 = \vec{k}_{sig}$$

$$\hbar\vec{k}_1 + \hbar\vec{k}_2 + \hbar\vec{k}_3 - \hbar\vec{k}_4 + \hbar\vec{k}_5 = \hbar\vec{k}_{sig}$$

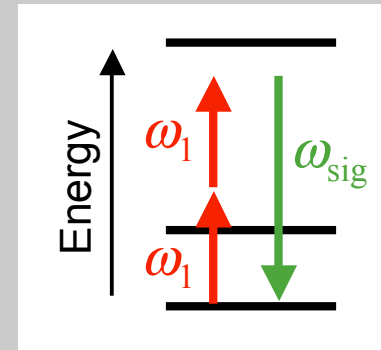
The second relation may not be satisfied. Ensuring that it is satisfied is called "**phase-matching.**"



Conservation laws for photons in SHG

Energy must be conserved:

$$\omega_1 + \omega_1 = \omega_{sig} \quad \Rightarrow \quad \omega_{sig} = 2\omega_1$$



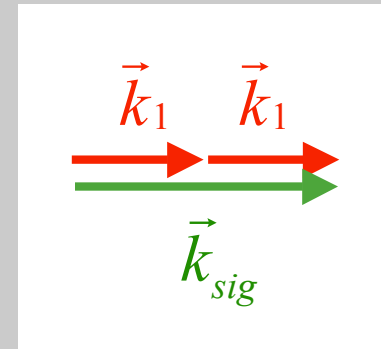
Momentum must also be conserved:

$$\vec{k}_1 + \vec{k}_1 = \vec{k}_{sig}$$

$$\Rightarrow \quad 2 \frac{\omega_1}{c_0} n(\omega_1) = \frac{2\omega_1}{c_0} n(2\omega_1)$$

$\omega_{sig} = 2\omega_1$

The equation above shows the conservation of momentum. The terms $2 \frac{\omega_1}{c_0} n(\omega_1)$ and $\frac{2\omega_1}{c_0} n(2\omega_1)$ are crossed out with pink 'X' marks. An arrow points from the $\omega_{sig} = 2\omega_1$ label to the $2\omega_1$ in the second term.



To simultaneously conserve energy and momentum:

$$\Rightarrow \quad n(\omega_1) = n(2\omega_1)$$

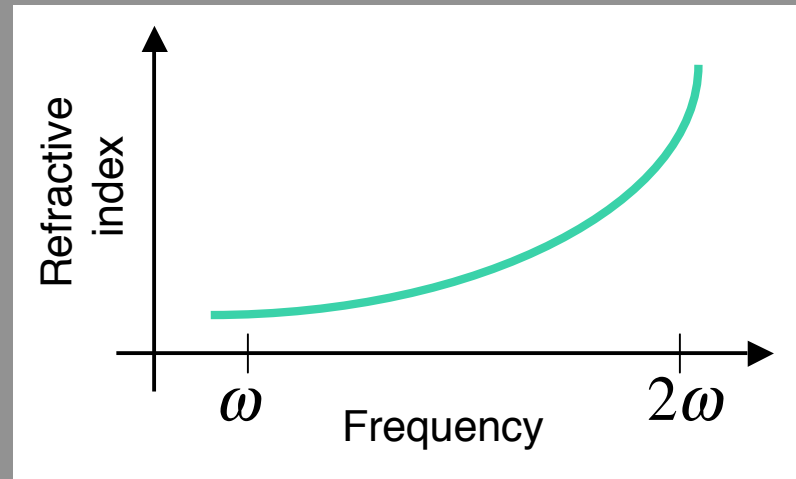
The phase-matching condition for SHG!

Phase-matching Second-Harmonic Generation

The phase-matching condition for SHG:

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

Unfortunately, dispersion prevents this from ever happening!



First Demonstration of Second-Harmonic Generation

P.A. Franken, et al, Physical Review Letters 7, p. 118 (1961)

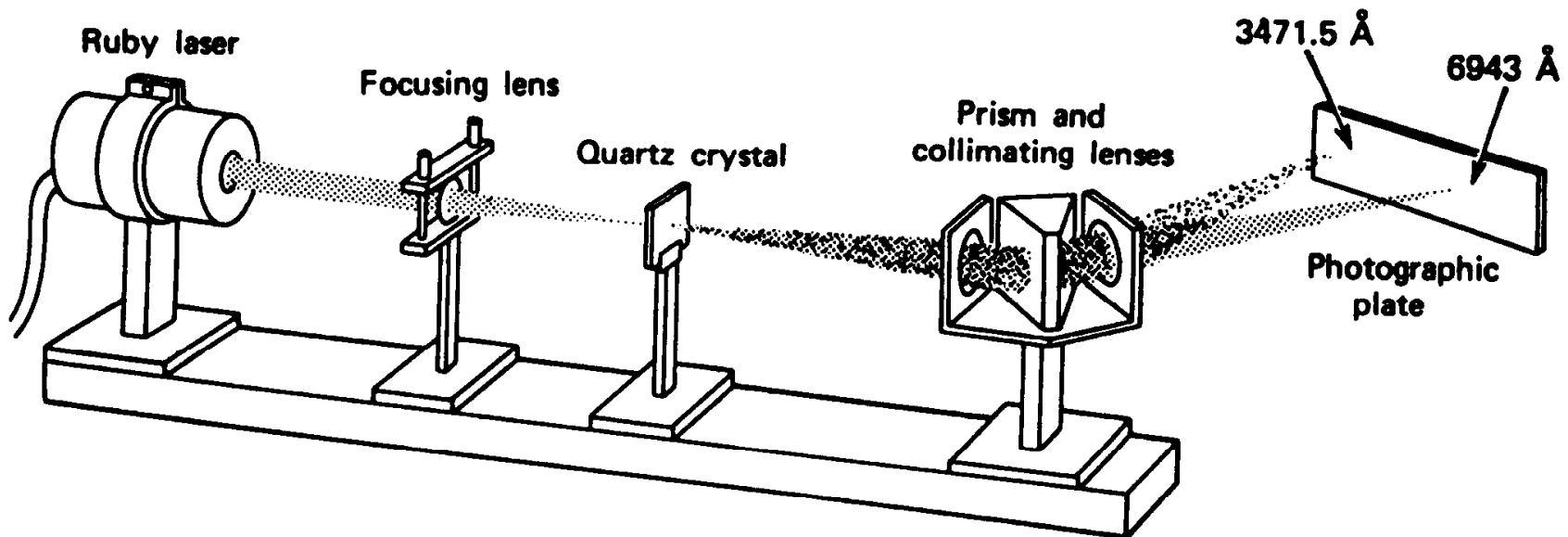
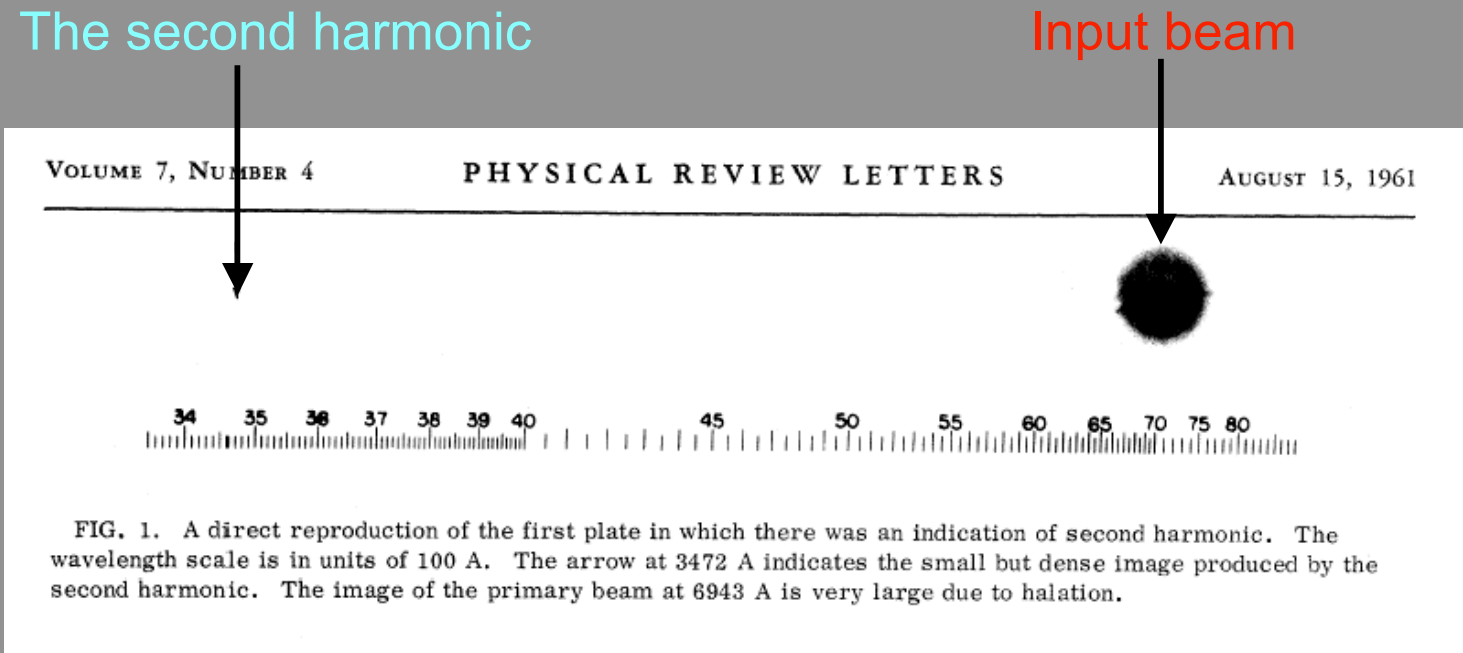


Figure 12.1. Arrangement used in the first experimental demonstration of second-harmonic generation [1]. A ruby-laser beam at $\lambda = 0.694 \mu\text{m}$ is focused on a quartz crystal, causing the generation of a (weak) beam at $\frac{1}{2}\lambda = 0.347 \mu\text{m}$. The two beams are then separated by a prism and detected on a photographic plate.

The second-harmonic beam was very weak because the process wasn't phase-matched.

First demonstration of SHG: The Data

The actual published result...



Note that the very weak spot due to the second harmonic is missing. It was removed by an overzealous Physical Review Letters editor, who thought it was a speck of dirt.

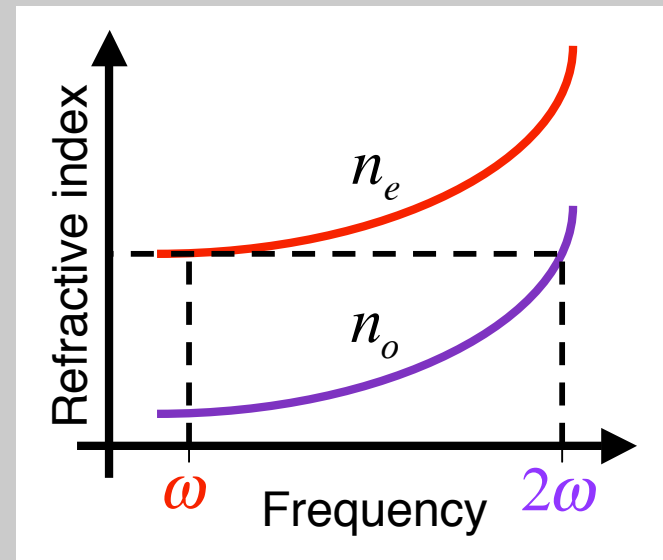
Phase-matching Second-Harmonic Generation using birefringence

Birefringent materials have different refractive indices for different polarizations. “Ordinary” and “Extraordinary” refractive indices can be different by up to 0.1 for SHG crystals.

We can now satisfy the phase-matching condition.

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

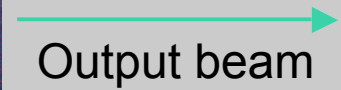
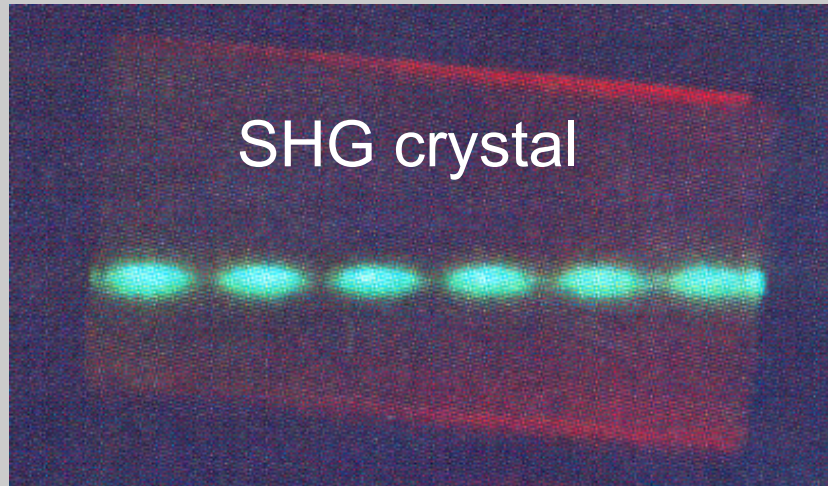
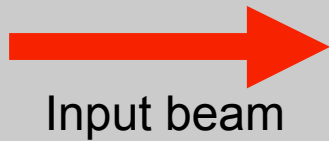
$$n_e(\omega) = n_o(2\omega)$$



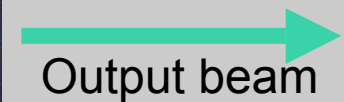
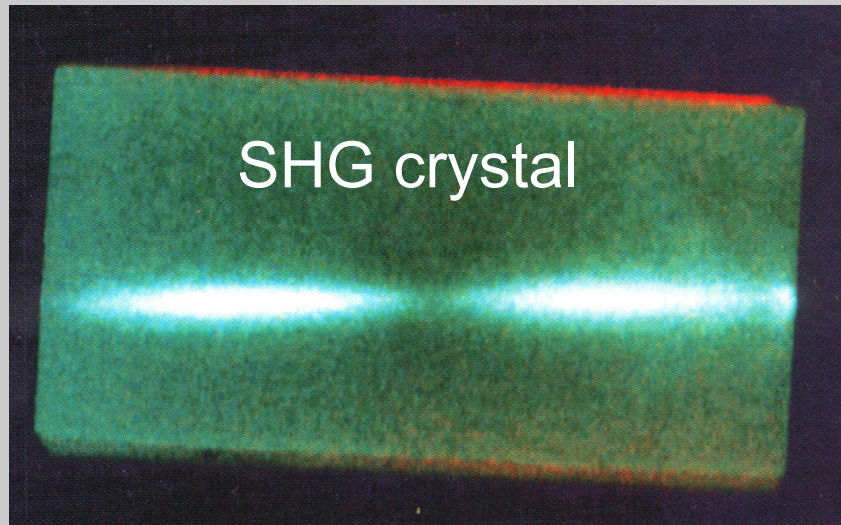
n_e depends on propagation angle, so we can tune for a given ω . Some crystals have $n_e < n_o$, so the opposite polarizations work.

Light created in real crystals

Far from
phase-matching:



Closer to
phase-matching:

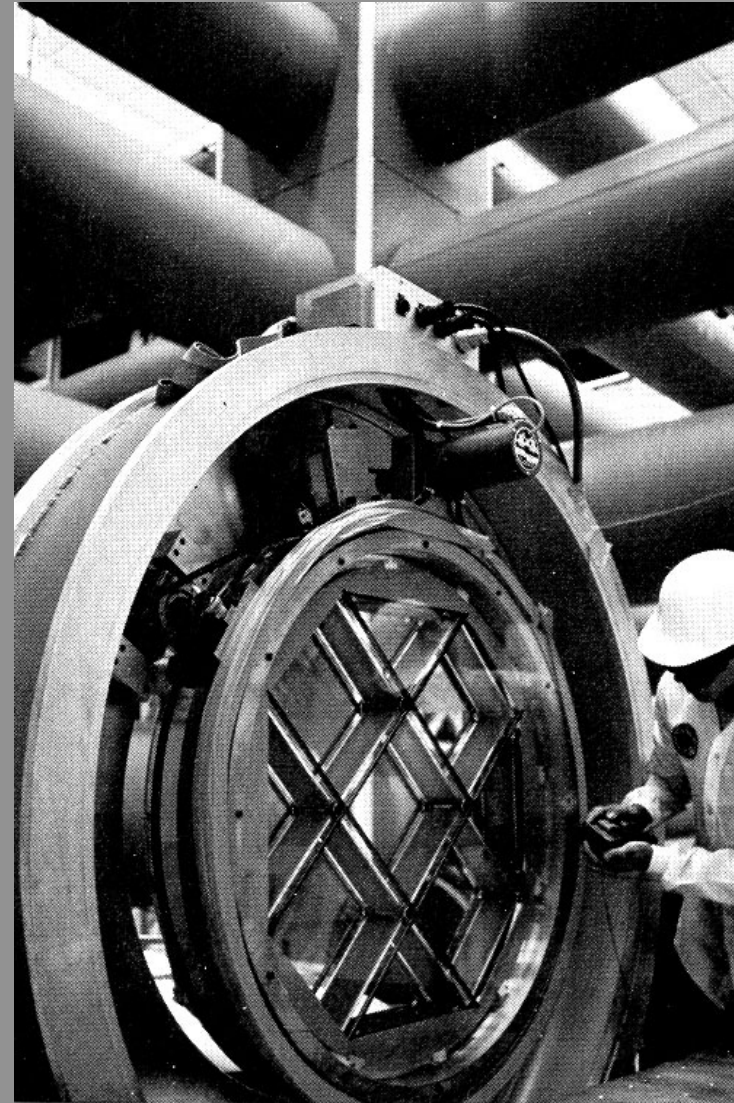


Note that SH beam is brighter as phase-matching is achieved.

Second-Harmonic Generation

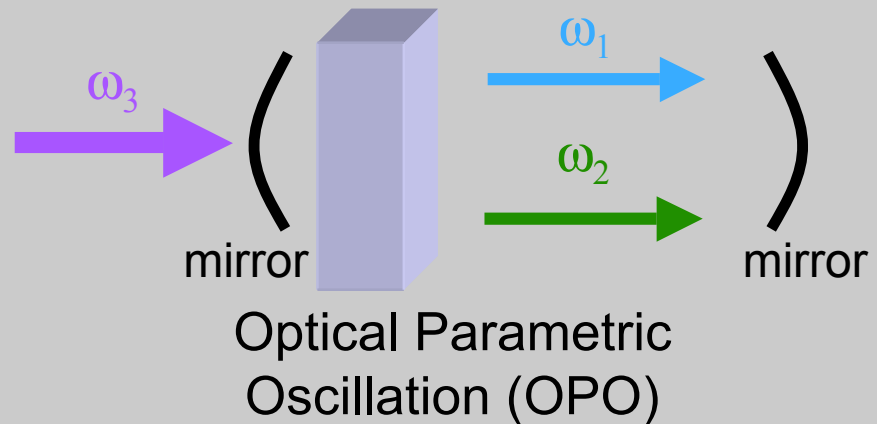
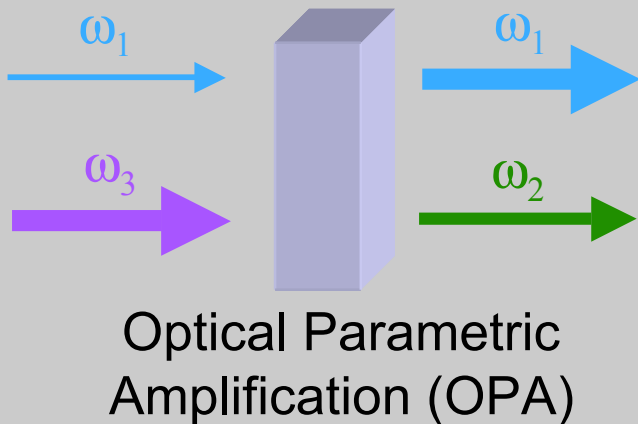
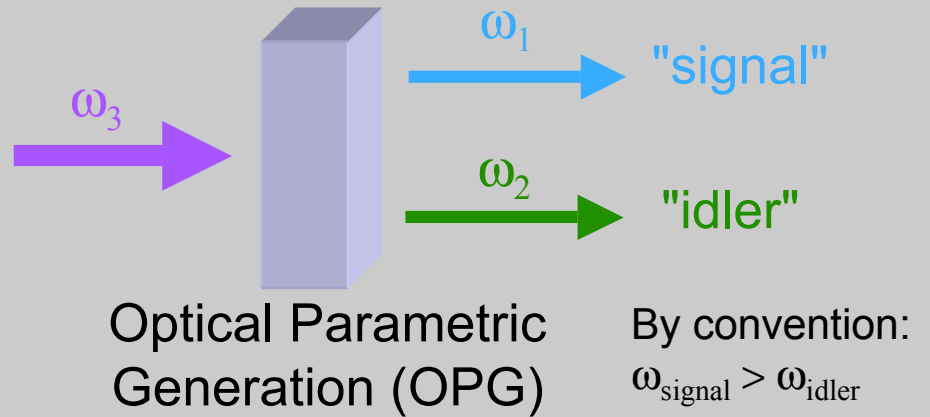
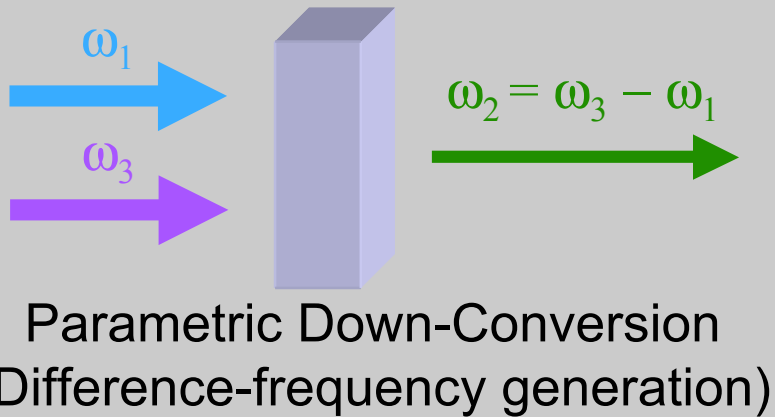
SHG KDP crystals at Lawrence Livermore National Laboratory

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

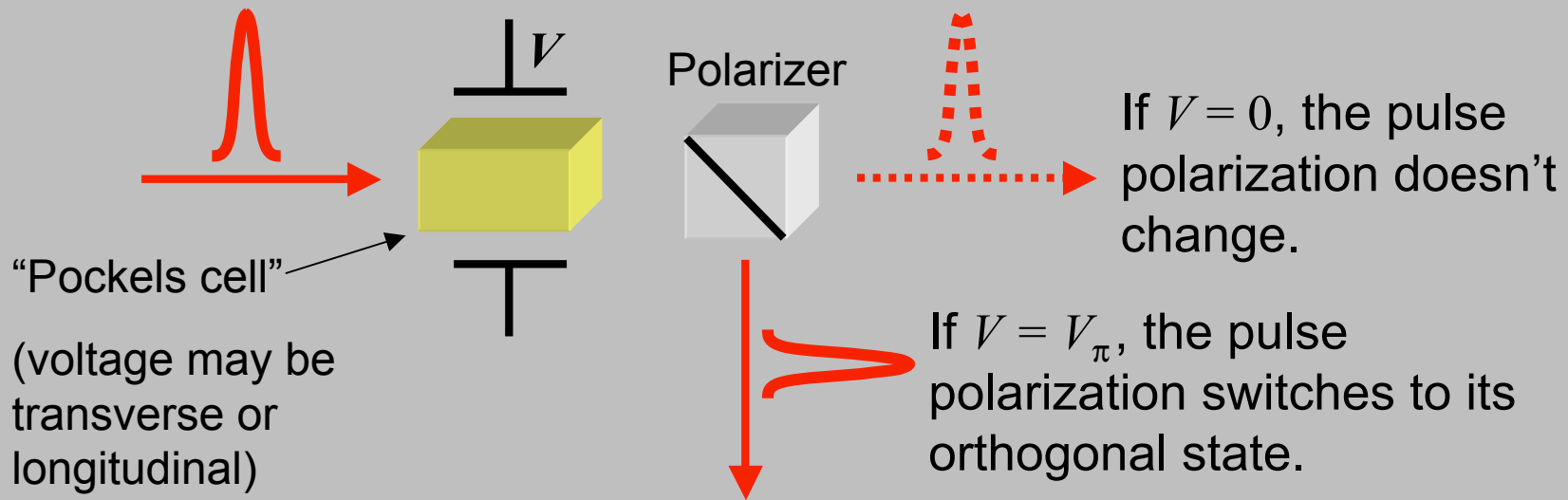
Difference-frequency generation takes many useful forms.



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!).

A few kV can turn a crystal into a half- or quarter-wave plate.



Abruptly switching a Pockels cell allows us to switch a pulse into or out of a laser.

Nonlinear Refractive Index

The refractive index in the presence of linear *and nonlinear* polarization:

$$n = \sqrt{1 + \chi^{(1)} + \chi^{(3)} |E|^2}$$

Now, the usual refractive index (which we'll call n_0) is: $n_0 = \sqrt{1 + \chi^{(1)}}$

$$\text{So: } n = \sqrt{n_0^2 + \chi^{(3)} |E|^2} = n_0 \sqrt{1 + \chi^{(3)} |E|^2 / n_0^2}$$

Assume that the nonlinear term $\ll n_0$:

$$\text{So: } n \approx n_0 \left[1 + \frac{1}{2} \chi^{(3)} |E|^2 / n_0^2 \right] \approx n_0 + \chi^{(3)} |E|^2 / 2n_0$$

Usually, we define a “nonlinear refractive index”: $n_2 \propto \chi^{(3)} / 2n_0$

$$n \approx n_0 + n_2 I$$

$$\text{since } I \propto |E|^2$$

Many nonlinear-optical effects can be considered as **induced gratings**.

The irradiance of two crossed beams is sinusoidal, inducing a sinusoidal absorption or refractive index in the medium—a diffraction grating!

An induced grating results from the cross term in the irradiance:

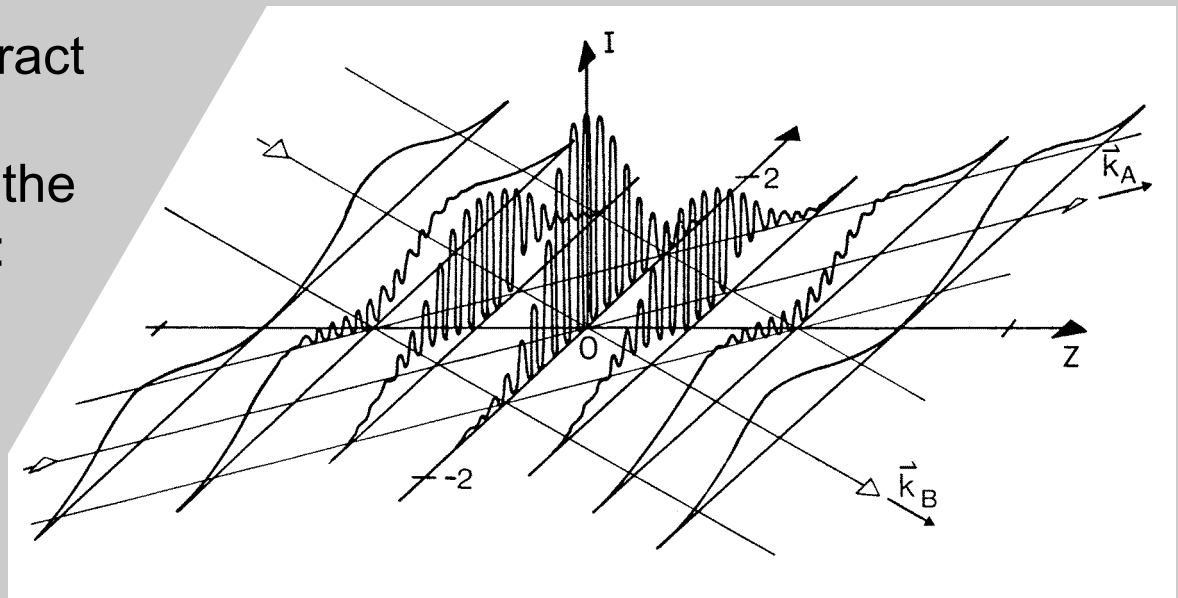
$$\begin{aligned} & \text{Re} \left\{ E_1 \exp [i(\omega t - kz \cos \theta - kx \sin \theta)] E_2^* \exp [-i(\omega t - kz \cos \theta + kx \sin \theta)] \right\} \\ & \propto \text{Re} \left\{ E_1 E_2^* \exp [-2ikx \sin \theta] \right\} \end{aligned}$$

A third beam will then diffract into a different direction.

This yields a beam that's the product of E_1 , E_2^* , and E_3 :

$$E_{sig} \propto (E_1 E_2^*) E_3$$

This is just a generic four-wave-mixing effect.

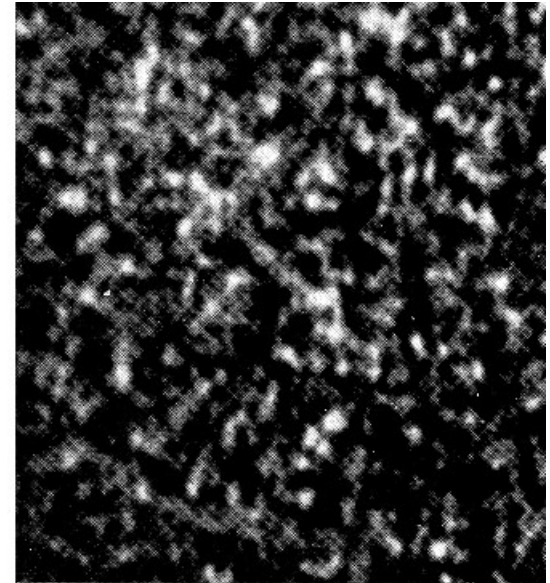
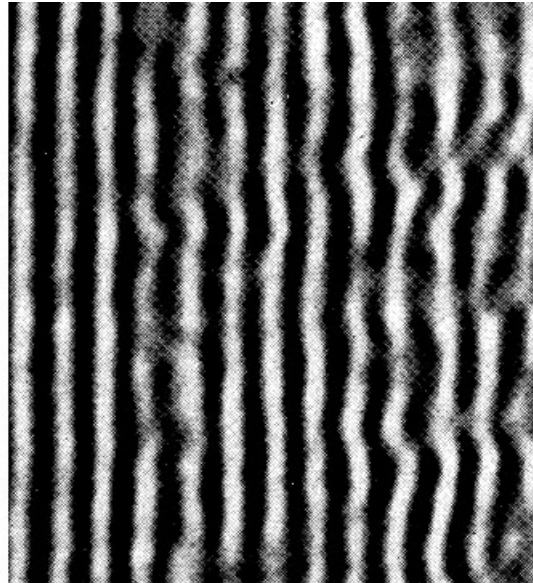
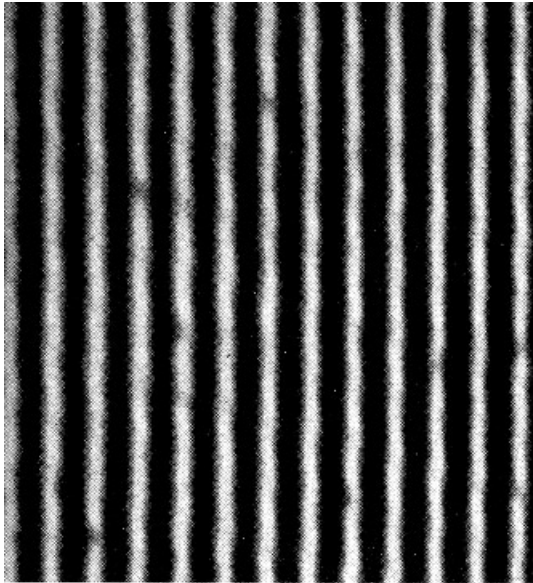


Induced gratings with plane waves and more complex beams

Two plane waves

A plane wave and a slightly distorted wave

A plane wave and a very distorted wave

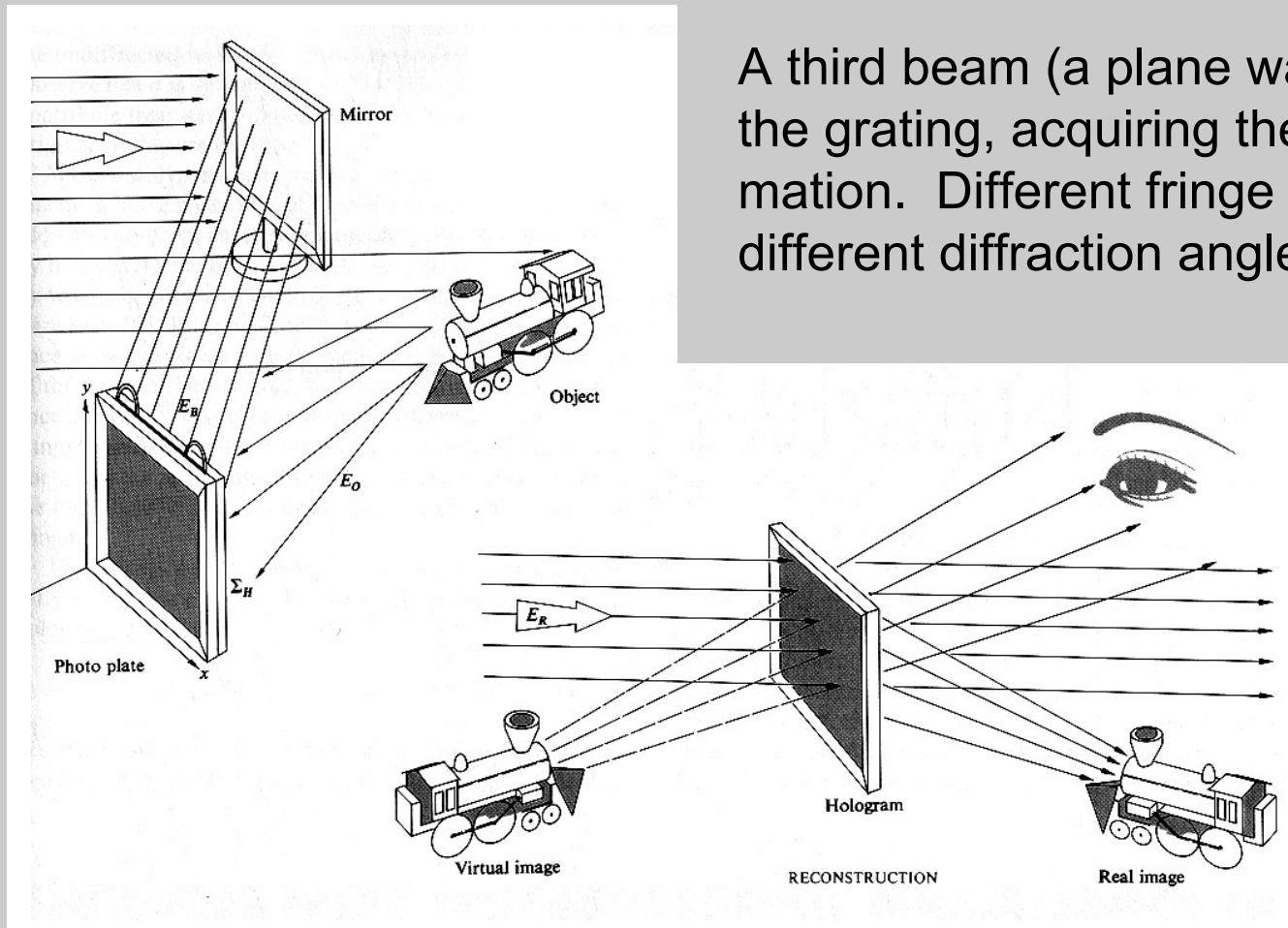


All such induced gratings will diffract a plane wave, reproducing the distorted wave.

Holography is an induced-grating process.

One of the write beams has a complex spatial pattern—the image. Different incidence angles correspond to different fringe spacings. Different object views are stored as different fringe spacings.

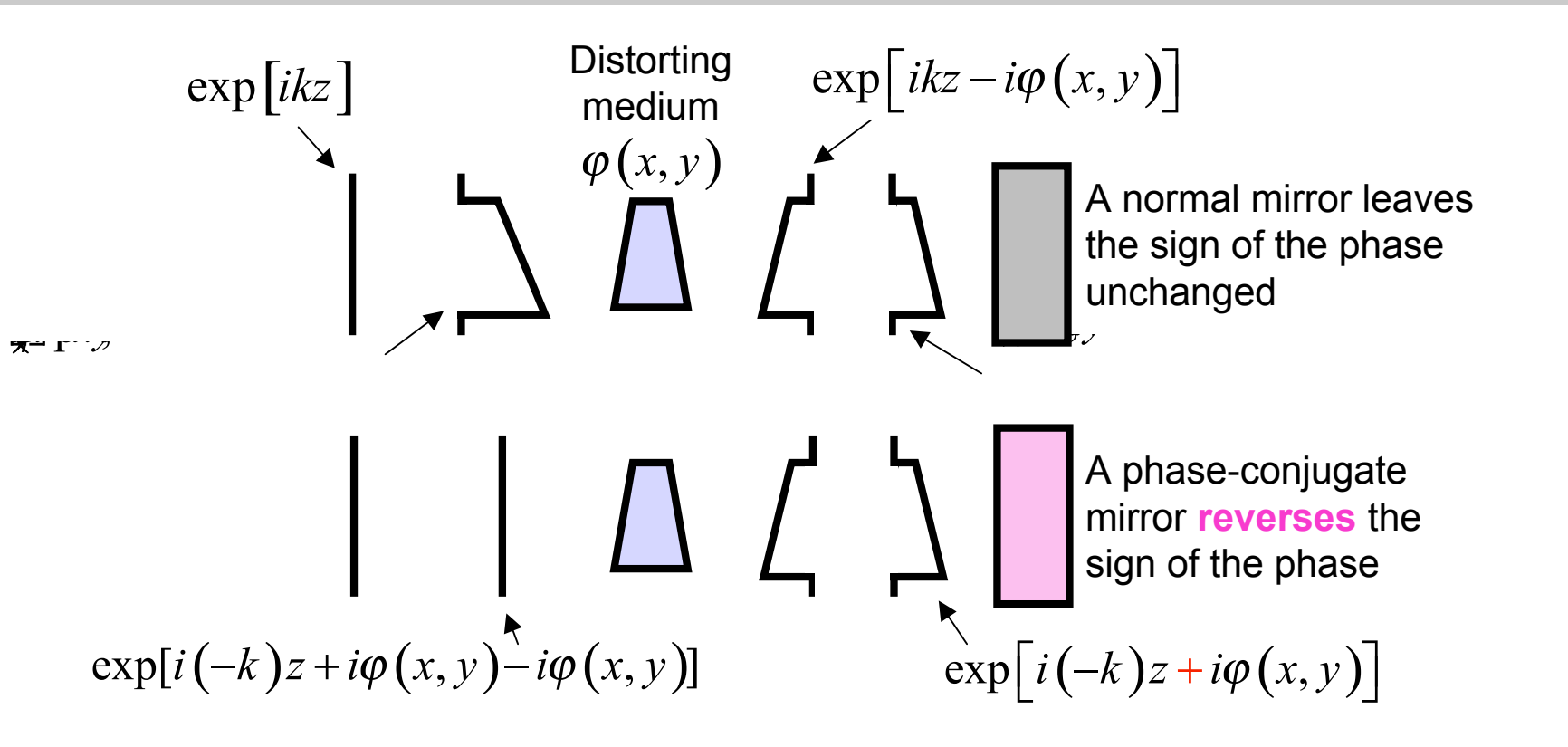
A third beam (a plane wave) diffracts off the grating, acquiring the image information. Different fringe spacings yield different diffraction angles—hence 3D!



The light phase stores the angular info.

Phase conjugation

When a nonlinear-optical effect produces a light wave proportional to E^* , the process is called a phase-conjugation process. Phase conjugators can cancel out aberrations.



The second traversal through the medium cancels out the phase distortion caused by the first pass!

Self-Phase Modulation & Continuum Generation

The self-modulation develops a phase vs. time proportional to the input pulse intensity vs. time.

$$E_{sig}(z, t) = E_{sig}(0, t) \exp[in_0 k z] = E_{sig}(0, t) \exp\{i[n_0 + n_2 I(t)]k z\}$$

$$\propto E_{sig}(0, t) \exp[in_2 k I(t) z]$$

Pulse Intensity vs. time

The further the pulse travels, the more modulation occurs.

That is:

$$\phi(z, t) = n_2 k z I(t)$$

A flat phase vs. time yields the narrowest spectrum. If we assume the pulse starts with a flat phase, then SPM broadens the spectrum.

This is not a small effect! A total phase variation of hundreds can occur! A broad spectrum generated in this manner is called a **Continuum**.

Experimental Continuum spectrum in a fiber

Continua created by propagating 500-fs 625-nm pulses through 30 cm of single-mode fiber.

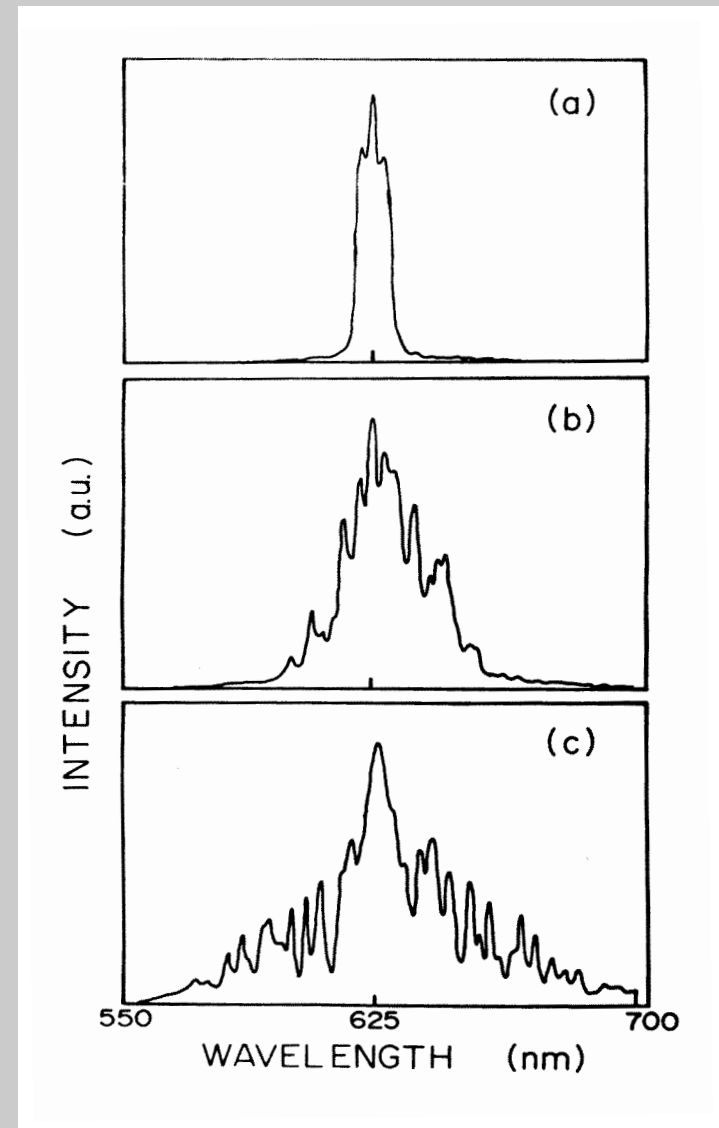
The Supercontinuum Laser Source, Alfano, ed.

Broadest spectrum occurs for highest energy.

Low Energy

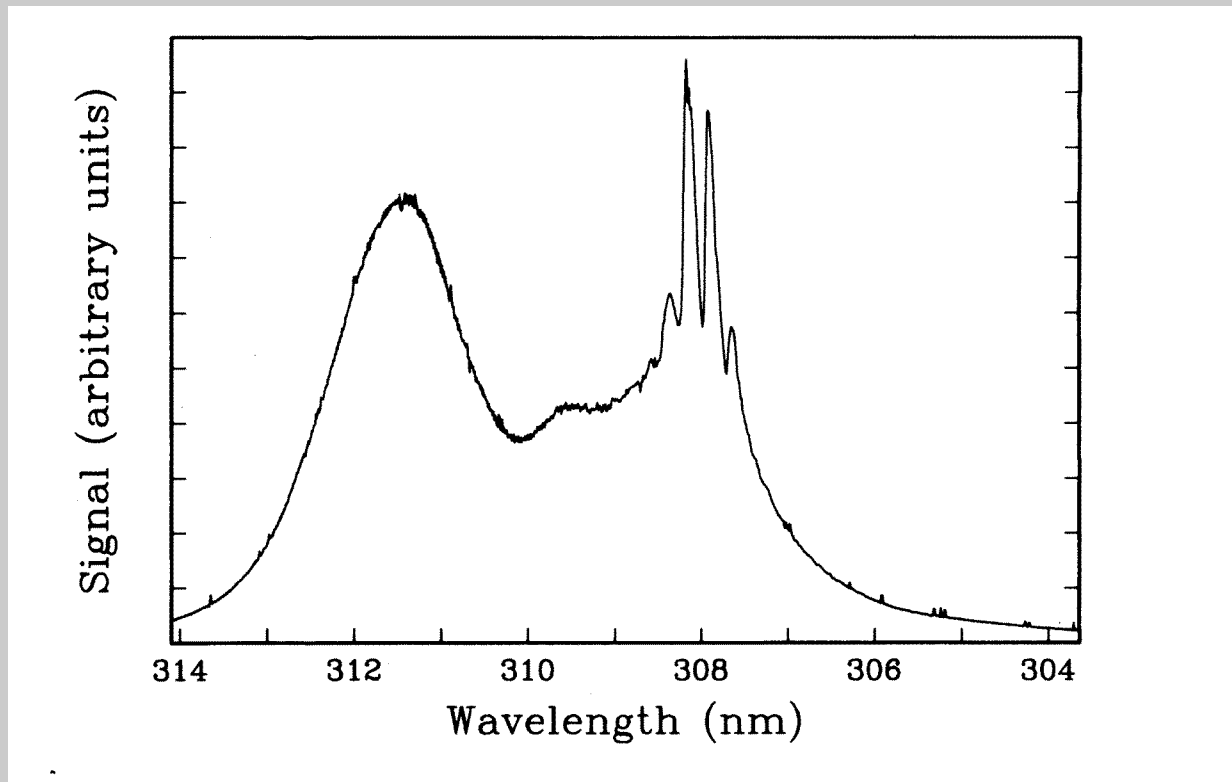
Medium Energy

High Energy



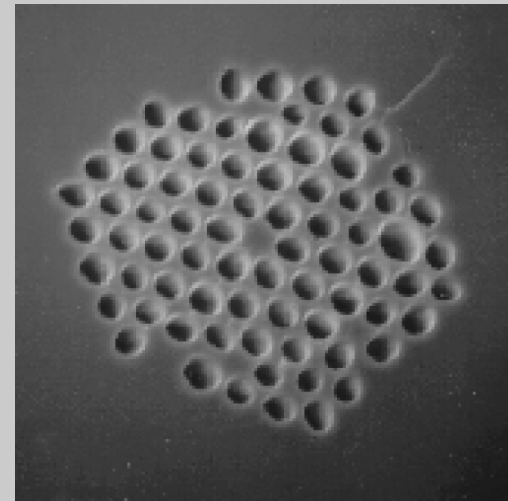
UV Continuum in Air!

308 nm input pulse; weak focusing with a 1-m lens.

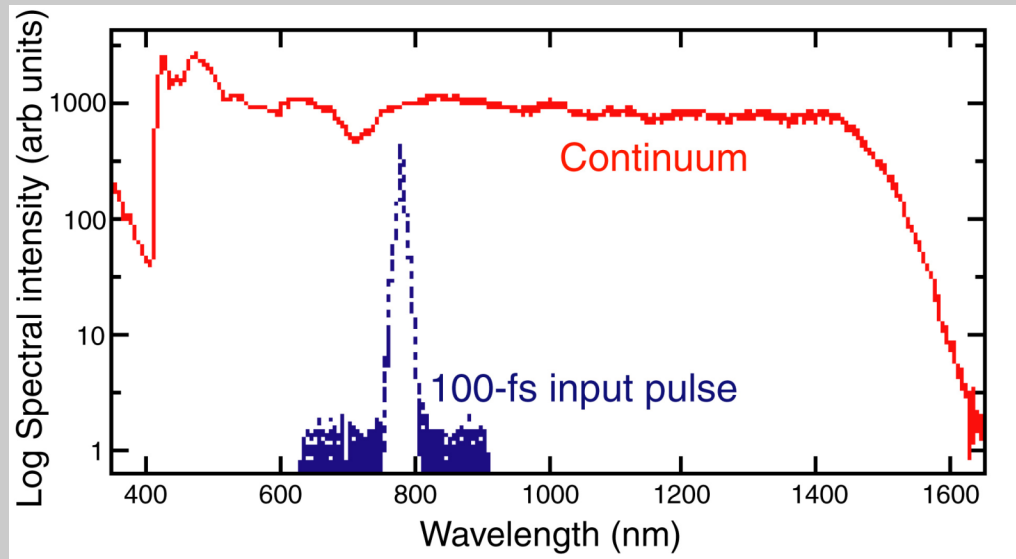


*The Supercontinuum
Laser Source, Alfano, ed.*

The continuum from microstructure optical fiber is ultrabroadband.



Cross section of the microstructure fiber.



The spectrum extends from ~400 to ~1500 nm and is relatively flat (when averaged over time).

This continuum was created using *unamplified* Ti:Sapphire pulses.

J.K. Ranka, R.S. Windeler, and A.J. Stentz, *Opt. Lett.* Vol. 25, pp. 25-27, 2000

Continuum is quite beautiful!

