# Nonlinear optics

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

Maxwell's equations in a medium

Nonlinear-optical media

Second-harmonic generation
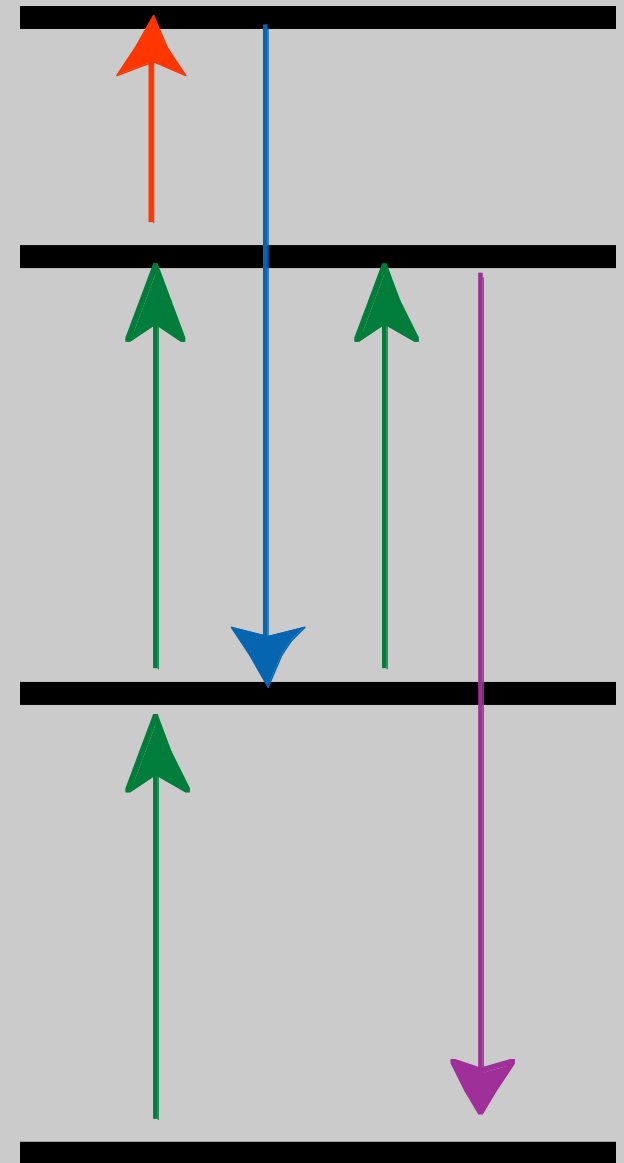
Sum- and difference frequency generation

Conservation laws for photons ("Phase-matching")

Induced gratings

Phase conjugation and aberration cancellation
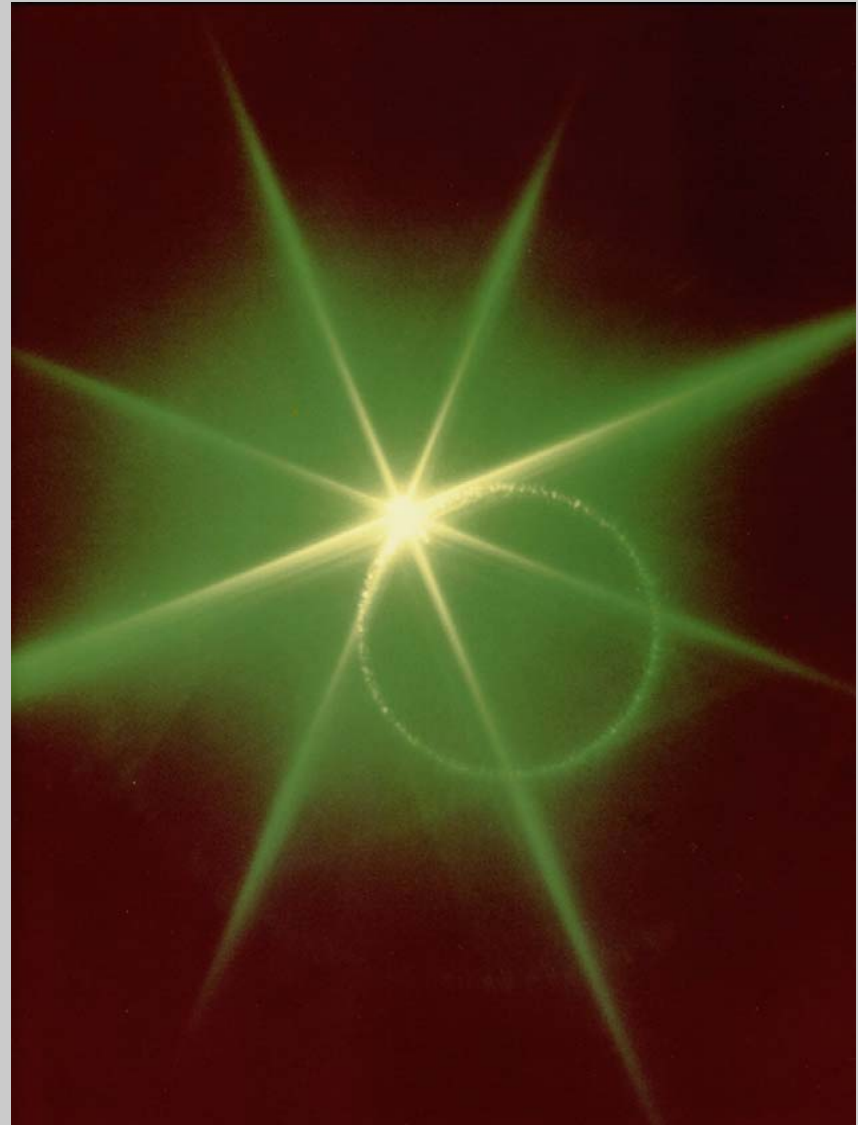
Holography

Self-phase modulation

# Nonlinear Optics produces 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 Man.
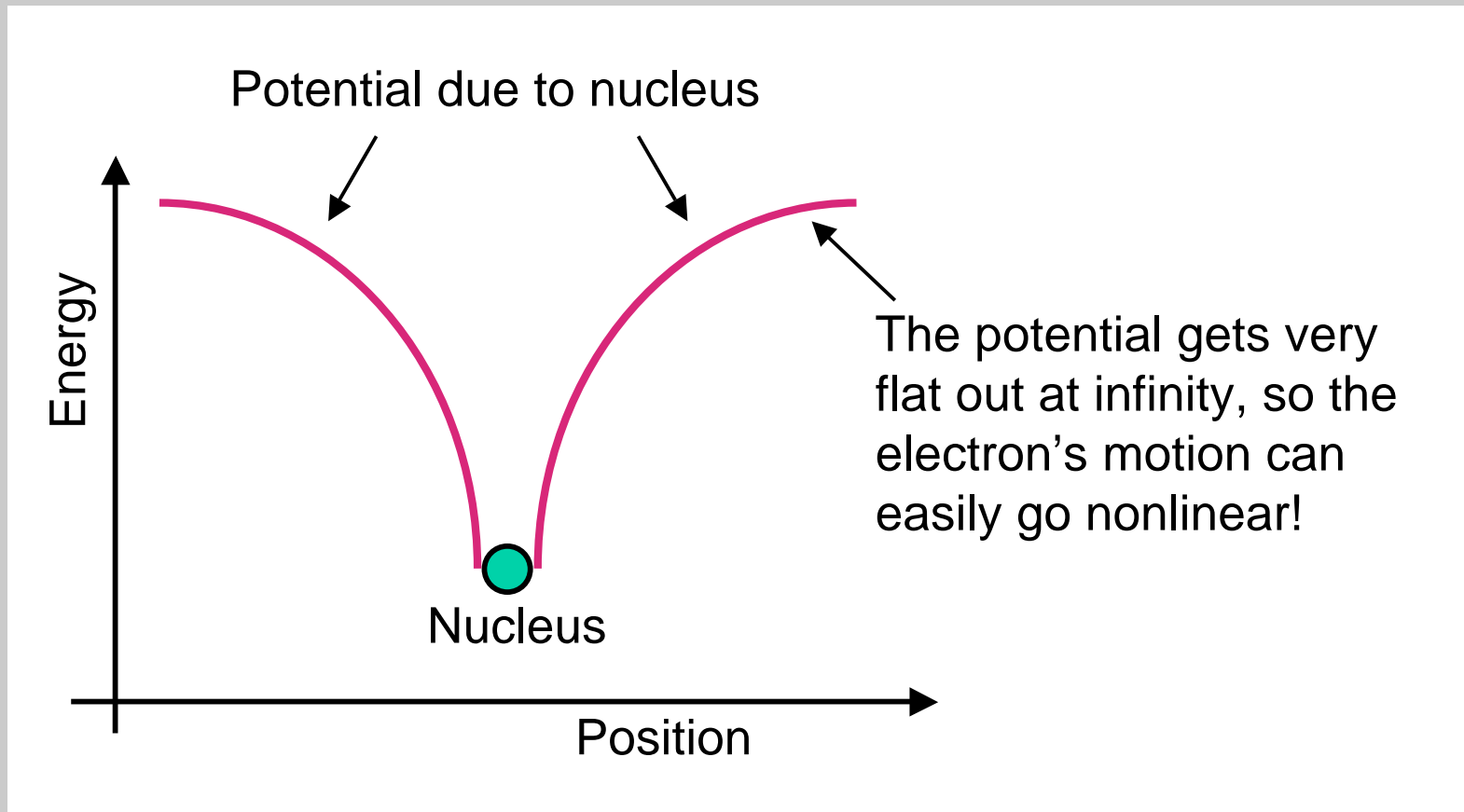
# Why do nonlinear effects occur, in general?

Imagine playing music through a cheap amplifier that just can't quite put out the power necessary to hit the loud notes.
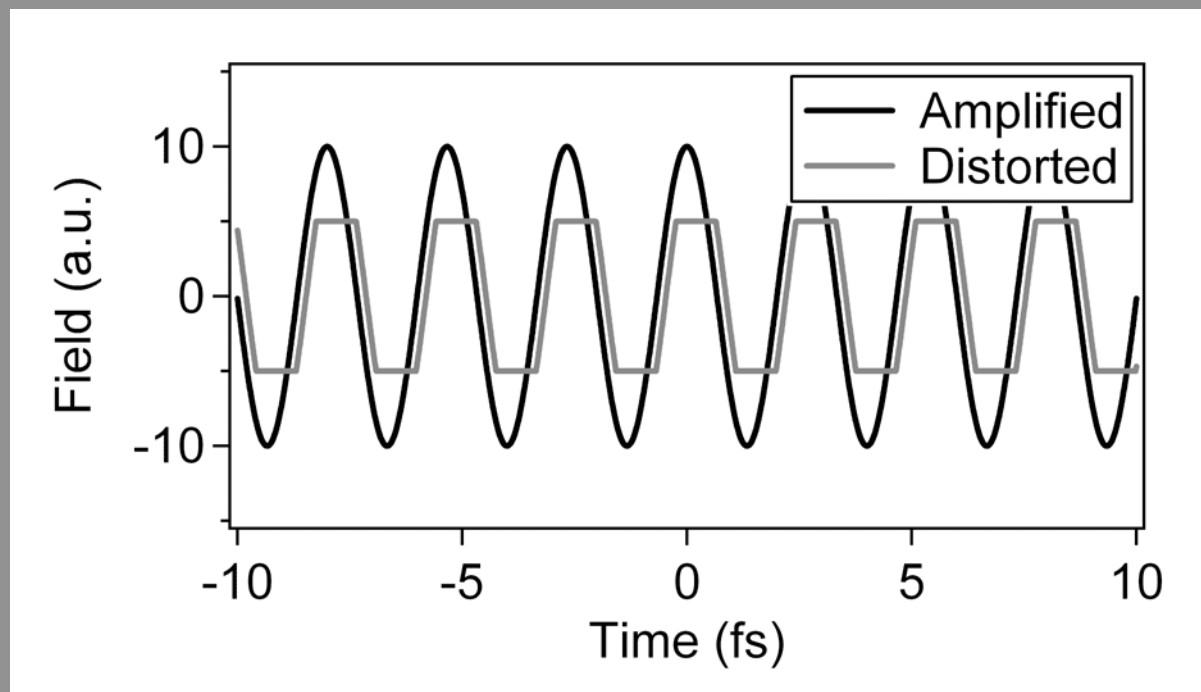


The sharp edges correspond to higher frequencies—harmonics!

# Nonlinear effects in atoms and molecules

Potential due to nucleus

Energy

Position

Nucleus

The potential gets very flat out at infinity, so the electron's motion can easily go nonlinear!

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

# 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.
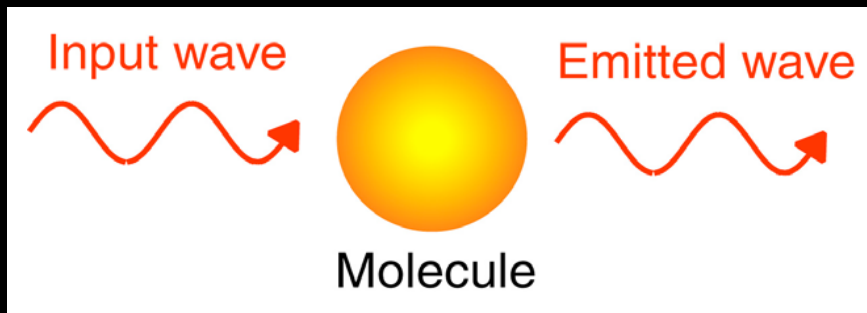


We can also imagine this process in terms of the molecular energy levels, using arrows for the photon energies:
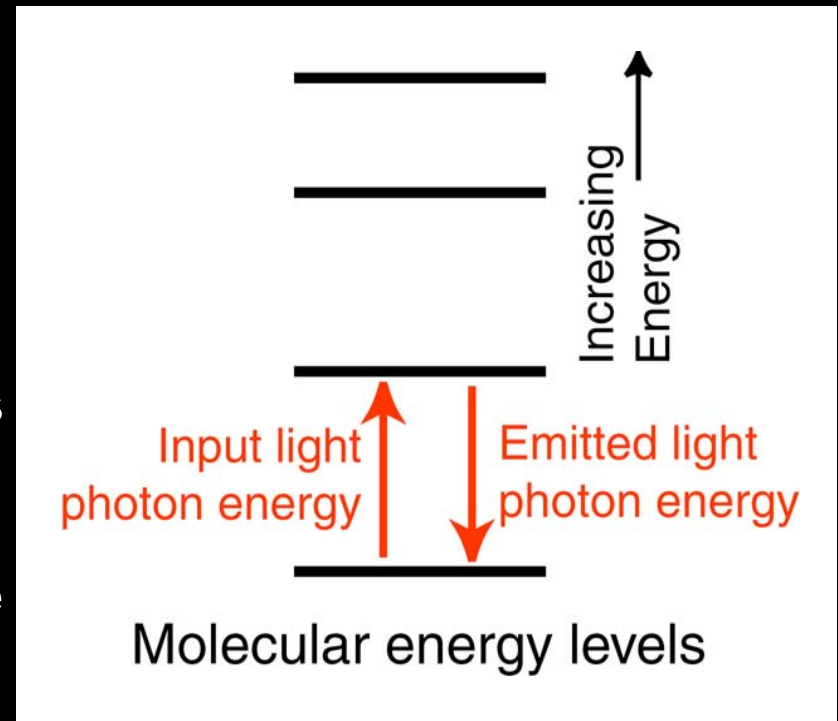
# Why do nonlinear-optical effects occur?

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.



Molecular energy levels
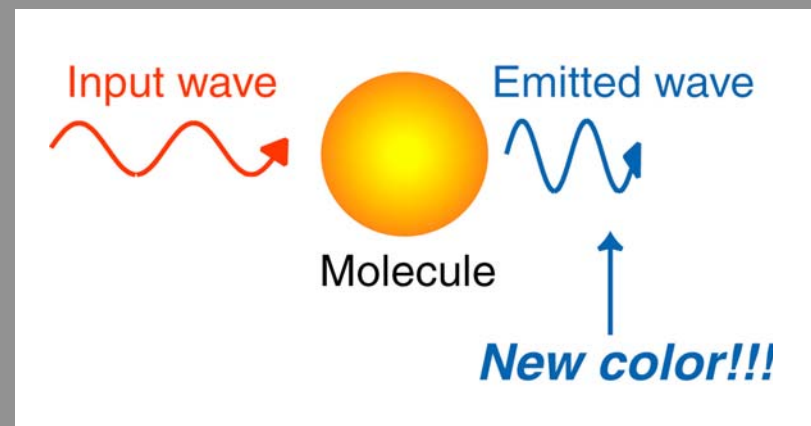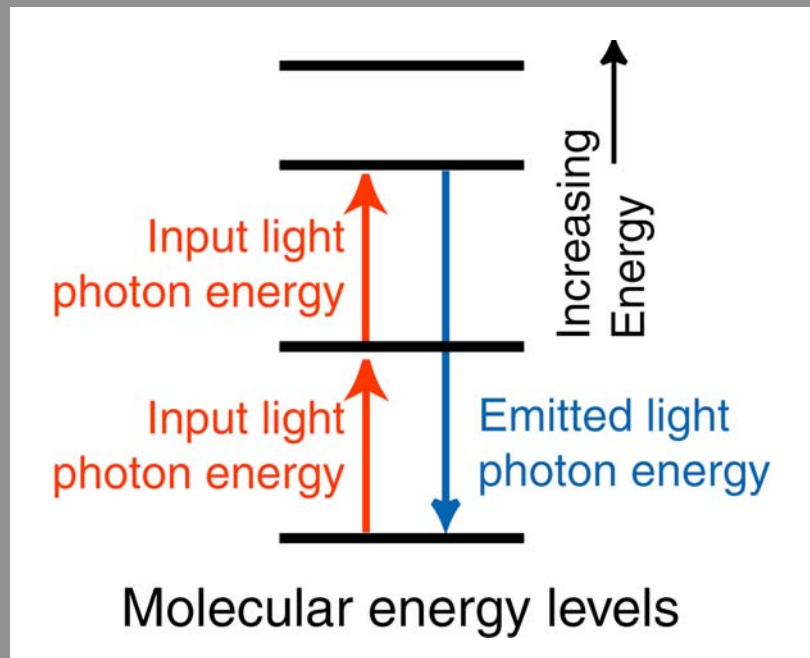
# Maxwell's Equations in a Medium

The induced polarization, $P$, contains the effect of the medium:

$$\vec{\nabla} \cdot \vec{E} = 0 \qquad \vec{\nabla} \times \vec{E} = -\frac{1}{c}\frac{\partial \vec{B}}{\partial t}$$

$$\vec{\nabla} \cdot \vec{B} = 0 \qquad \vec{\nabla} \times \vec{B} = \frac{1}{c}\frac{\partial \vec{E}}{\partial t} + \frac{4\pi}{c}\frac{\partial \vec{P}}{\partial t}$$

The polarization is proportional to the field:

$$\vec{P} = \chi \vec{E}$$

This has the effect of simply changing the dielectric constant:

$$\varepsilon = 1 + 4\pi\chi$$

# The effect of an induced polarization on a wave requires solving Maxwell's Equations.

The induced polarization in Maxwell's Equations yields another term in the wave equation:

$$\frac{\partial^2 E}{\partial x^2} - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P}{dt^2}$$

This is the "Inhomogeneous Wave Equation."

The polarization is the driving term for a new solution to this equation.

# 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:

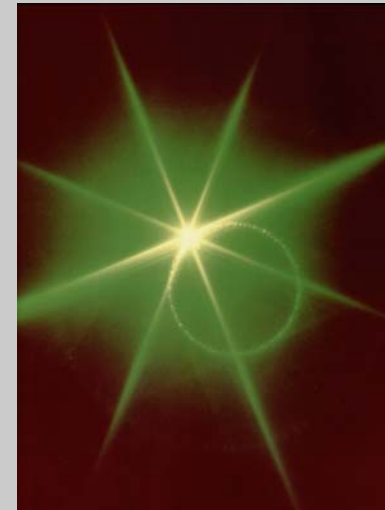$$\mathscr{P} = \chi^{(1)}\mathscr{E} + \chi^{(2)}\mathscr{E}^2 + \chi^{(3)}\mathscr{E}^3 + ...$$

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

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

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

**2ω = 2nd 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:

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

$$+ E_2^2 \exp(2i\omega_2 t) + E_2^{*2} \exp(-2i\omega_2 t) \qquad \textbf{2nd-harmonic gen}$$

$$+ 2E_1 E_2 \exp\left[i(\omega_1 + \omega_2)t\right] + 2E_1^* E_2^* \exp\left[-i(\omega_1 + \omega_2)t\right] \quad \textbf{Sum-freq gen}$$

$$+ 2E_1 E_2^* \exp\left[i(\omega_1 - \omega_2)t\right] + 2E_1^* E_2 \exp\left[-i(\omega_1 - \omega_2)t\right] \quad \textbf{Diff-freq gen}$$

$$+ 2\left|E_1\right|^2 + 2\left|E_2\right|^2 \qquad \textbf{optical rectification (dc)}$$

Note also that, when $\omega_i$ is negative inside the exp, the $E$ in front has a *.

# 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:
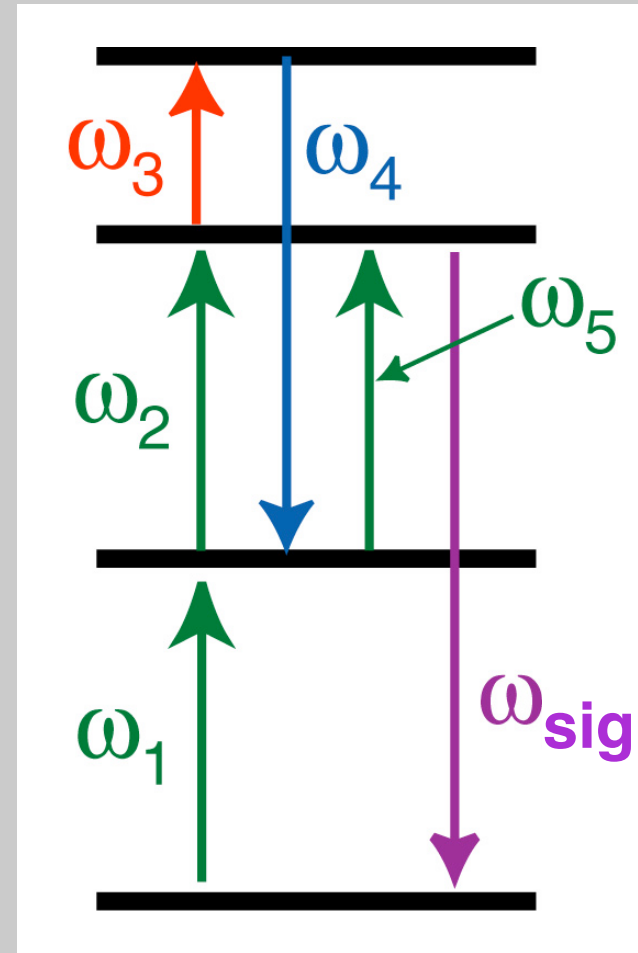
# Complicated nonlinear-optical effects can occur.



Emitted-light photon energy
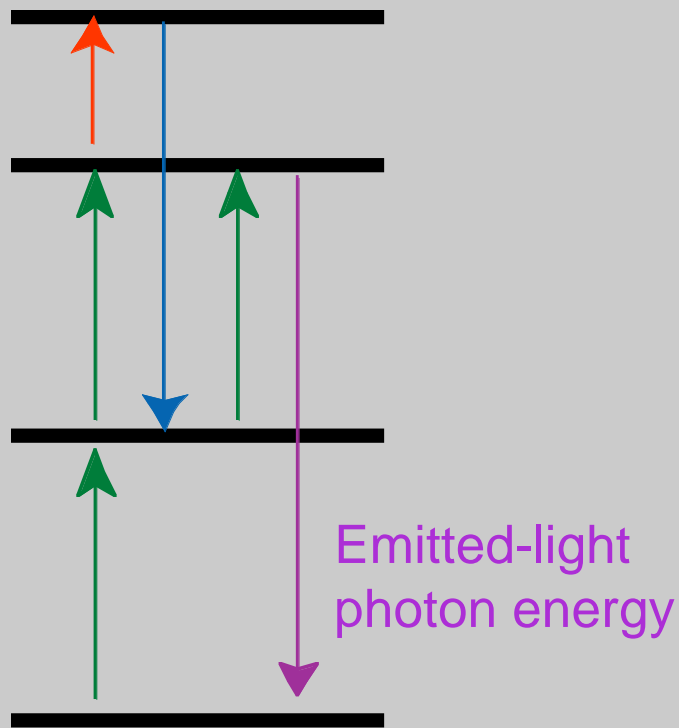
Nonlinear-optical processes are often referred to as:

"N-wave-mixing processes"

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

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.

# Conservation laws for photons in nonlinear optics
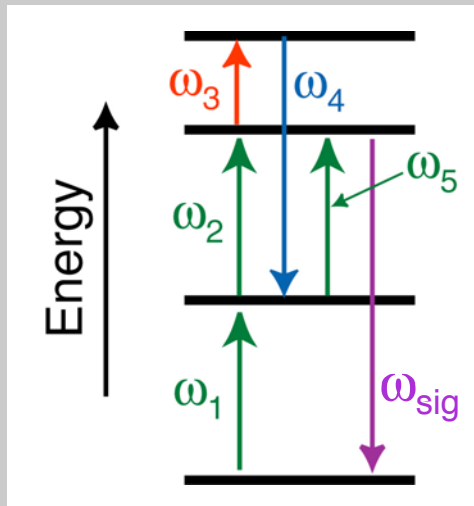




Energy must be conserved:

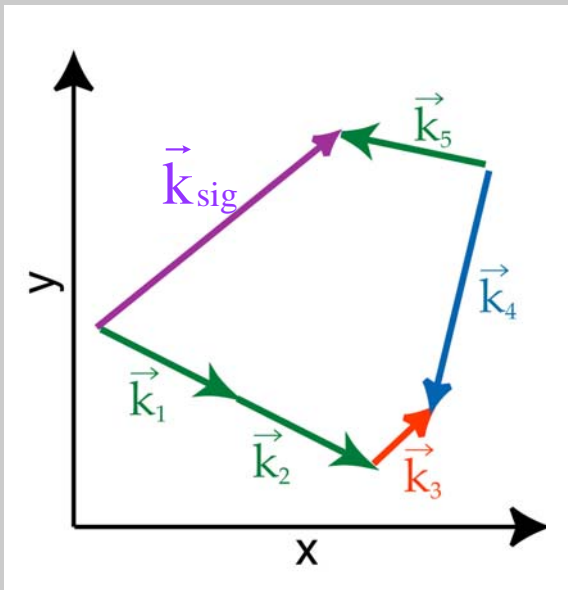$$\omega_1 + \omega_2 + \omega_3 - \omega_4 + \omega_5 = \omega_{sig}$$

(I've canceled the $\hbar$'s)

Momentum must also be conserved:

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

Unfortunately, $\vec{k}_{sig}$ may not correspond to a light wave at frequency $\omega_{sig}$!
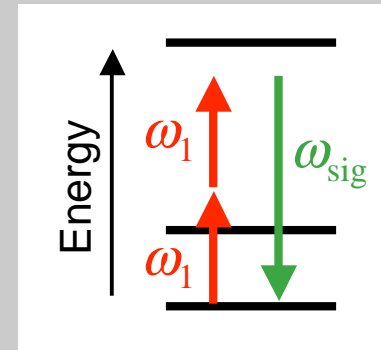
Satisfying these two relations simultaneously is called "**phase-matching**."

# Conservation laws for photons in SHG

Energy must be conserved:

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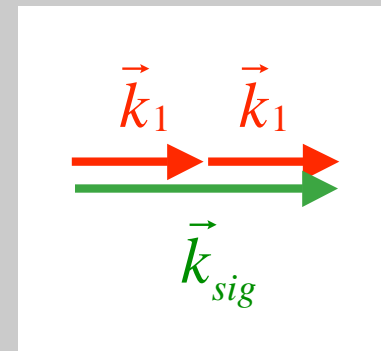


Momentum must also be conserved:

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

$$\omega_{sig} = 2\omega_1$$

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



To simultaneously conserve energy and momentum:

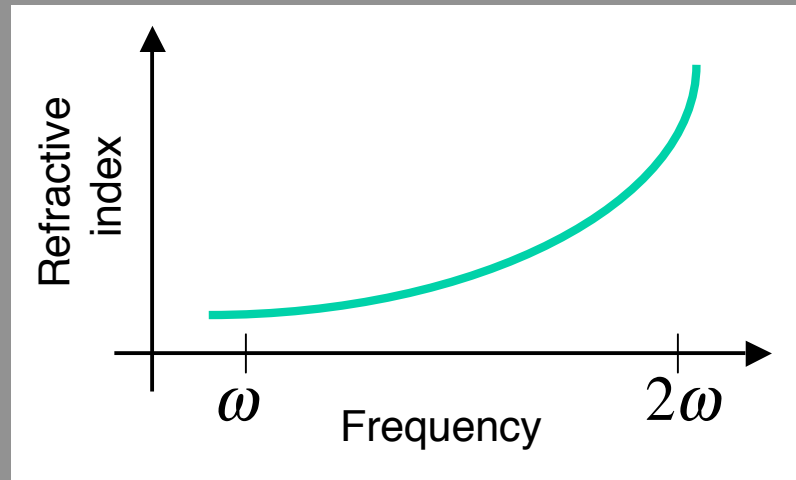$$\Rightarrow \quad \boxed{n(\omega_1) = n(2\omega_1)} \qquad \text{The phase-matching condition for SHG!}$$

# 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

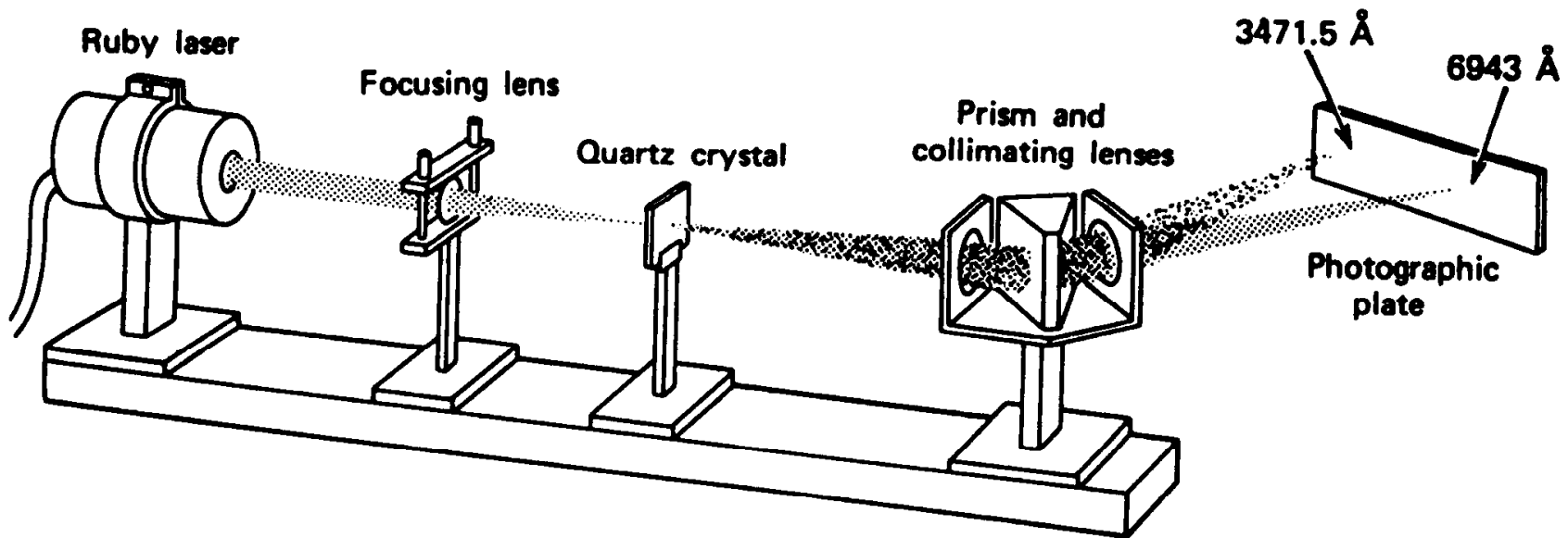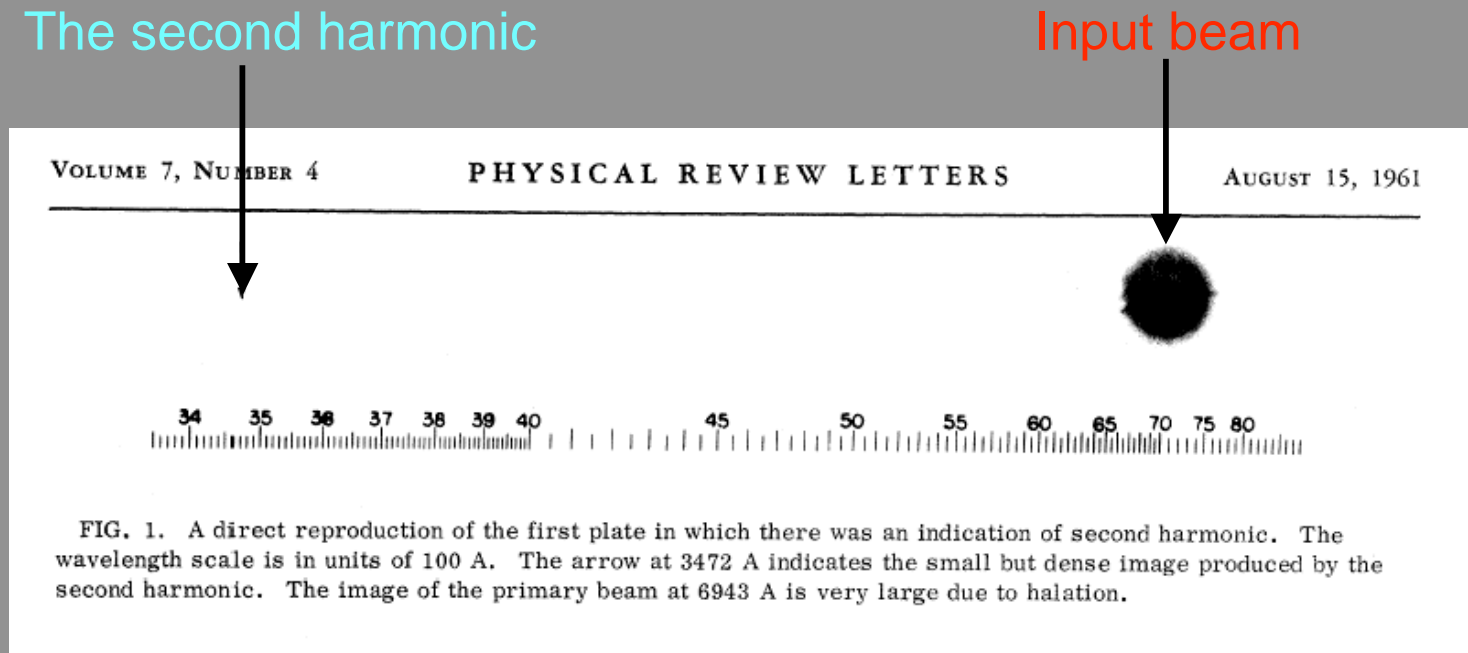P.A. Franken, et al, Physical Review Letters 7, p. 118 (1961)



**Figure 12.1.** Arrangement used in the first experimental demonstration of second-harmonic generation [1]. A ruby-laser beam at $\lambda = 0.694\ \mu m$ is focused on a quartz crystal, causing the generation of a (weak) beam at $\frac{1}{2}\lambda = 0.347\ \mu m$. 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…

The second harmonic                    Input beam



VOLUME 7, NUMBER 4          PHYSICAL REVIEW LETTERS          AUGUST 15, 1961

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.

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.
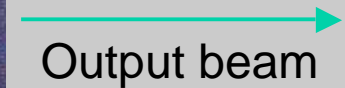
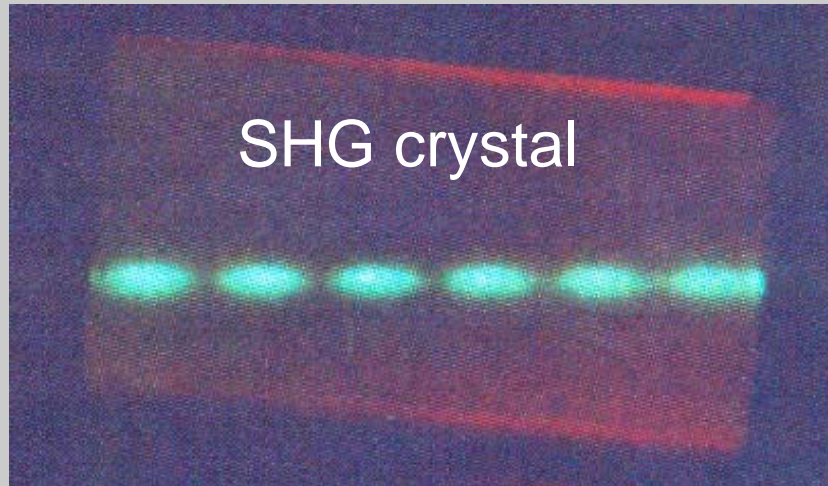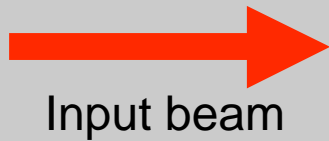Use the extraordinary polarization for $\omega$ and the ordinary for $2\omega$:

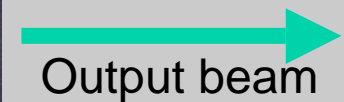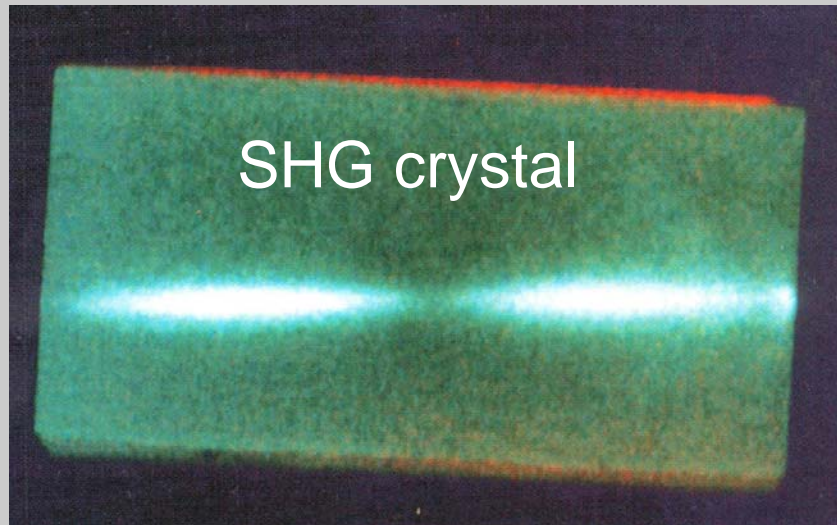$$n_e(\omega) = n_o(2\omega)$$
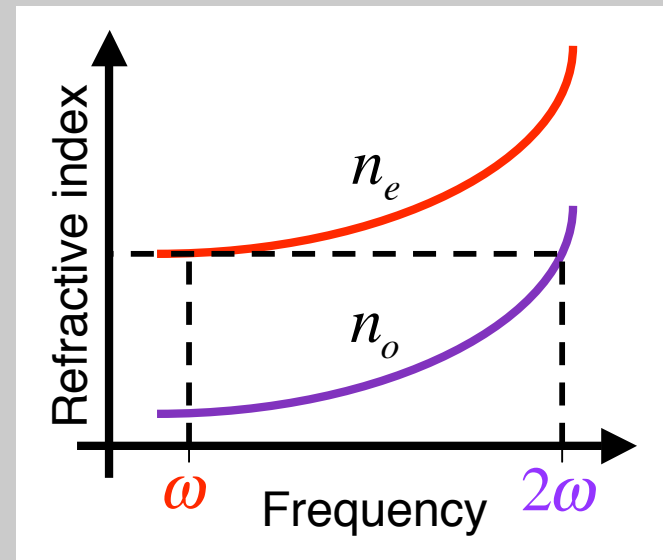


$n_e$ depends on propagation angle, so we can tune for a given $\omega$.

Some crystals have $n_e < n_o$, so the opposite polarizations work.

# Light created in real crystals

Far from
phase-matching:



SHG crystal

Input beam

Output beam

Closer to
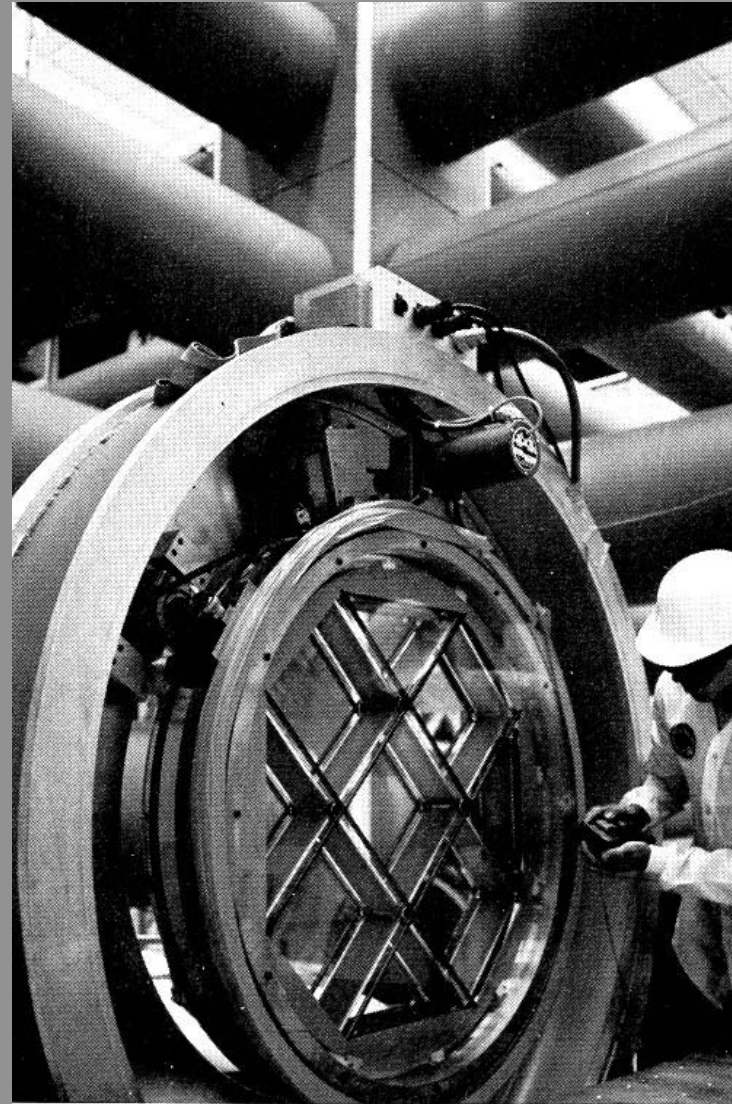phase-matching:



SHG crystal

Input beam

Output beam

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!
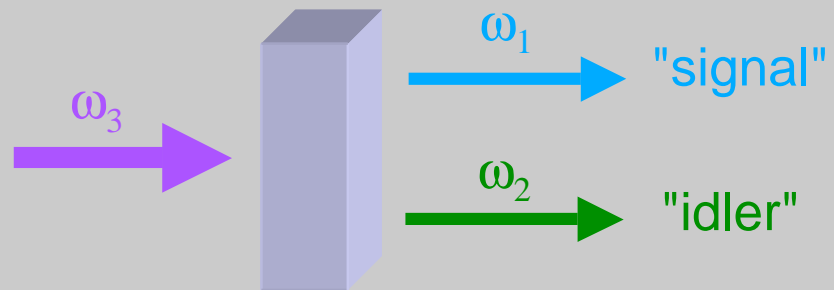
# Difference-Frequency Generation: Optical Parametric Generation, Amplification, Oscillation
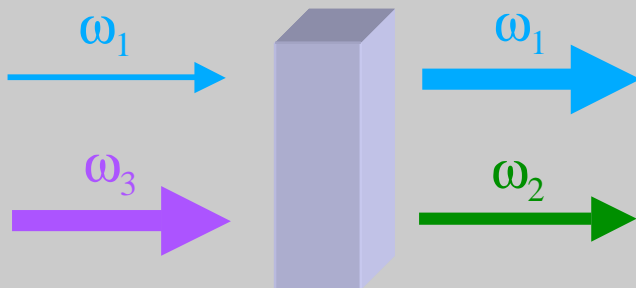
Difference-frequency generation takes many useful forms.

$\omega_1$

$\omega_3$

$\omega_2 = \omega_3 - \omega_1$
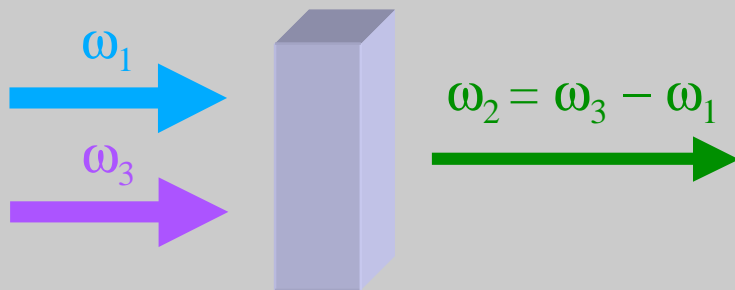
Parametric Down-Conversion
(Difference-frequency generation)

$\omega_3$

$\omega_1$  "signal"

$\omega_2$  "idler"

Optical Parametric
Generation (OPG)

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

$\omega_1$

$\omega_3$

$\omega_1$

$\omega_2$

Optical Parametric
Amplification (OPA)

$\omega_3$

$\omega_1$
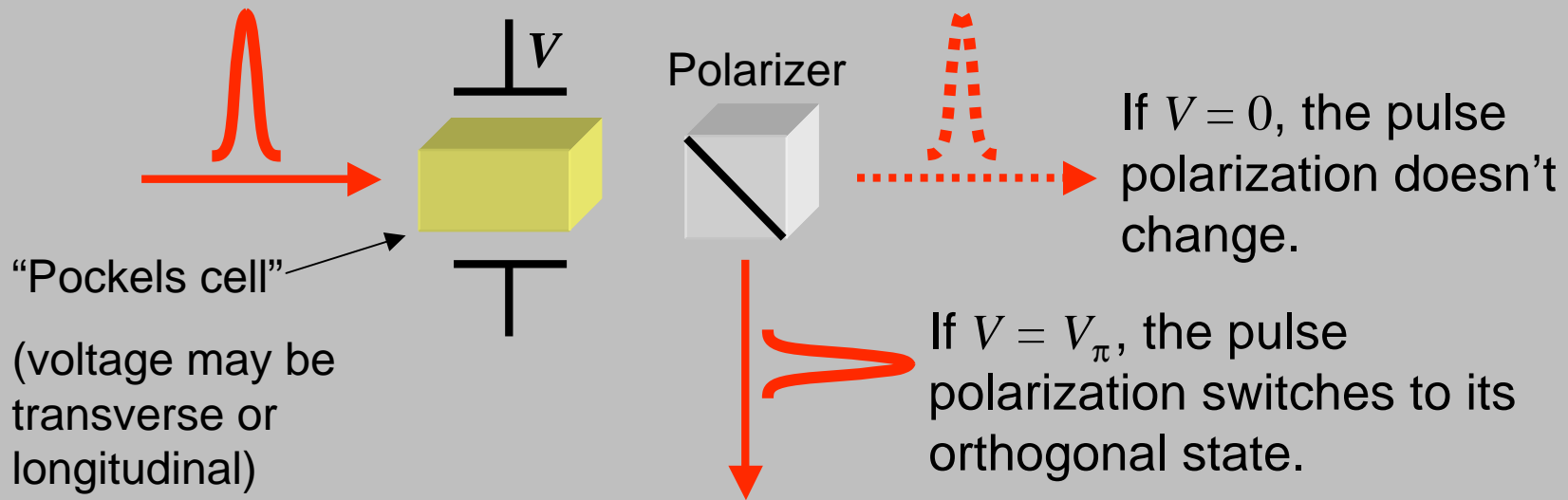
$\omega_2$

mirror

mirror
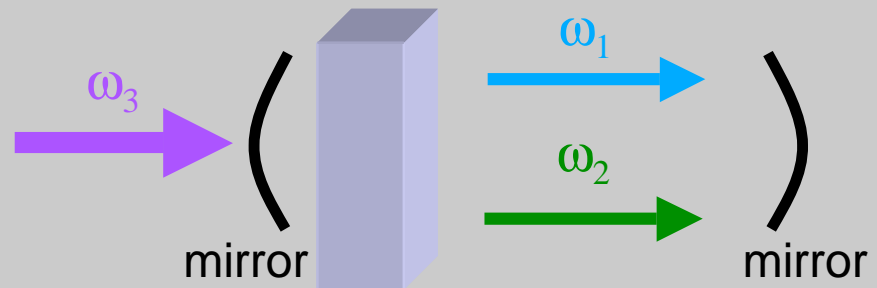
Optical Parametric
Oscillation (OPO)

# Another 2$^{nd}$-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.



"Pockels cell"

(voltage may be transverse or longitudinal)

Polarizer

If $V = 0$, the pulse polarization doesn't change.

If $V = V_\pi$, the pulse polarization switches to its orthogonal state.

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

# 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!

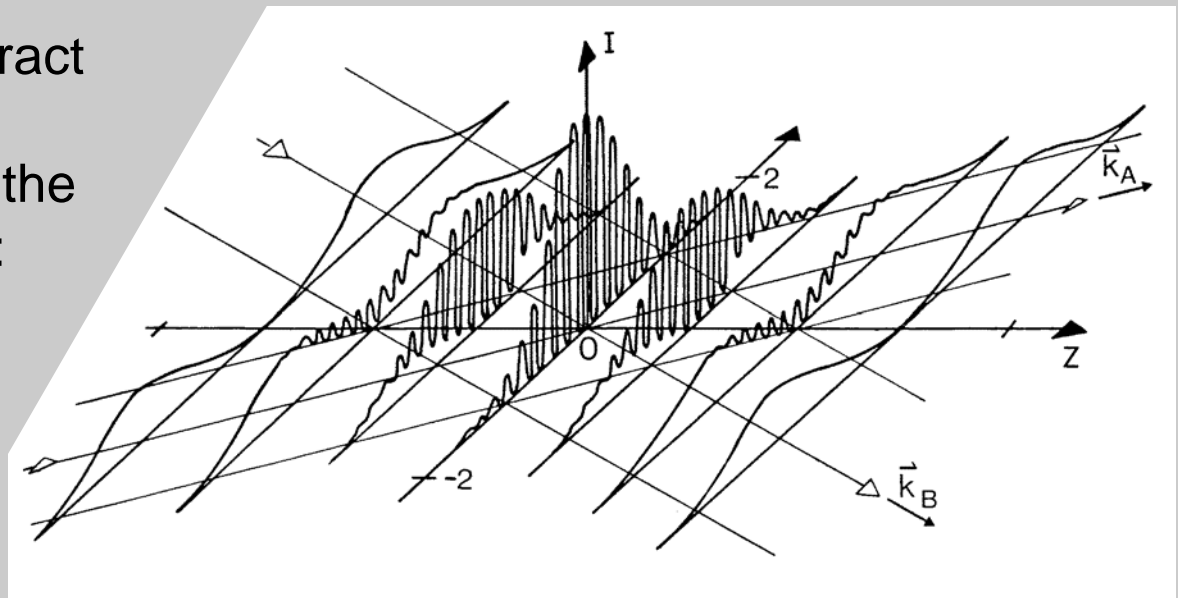An induced grating results from the cross term in the irradiance:

$$\mathrm{Re}\left\{E_1 \exp\left[i(\omega t - kz \cos\theta - kx \sin\theta)\right] E_2^* \exp\left[-i(\omega t - kz \cos\theta + kx \sin\theta)\right]\right\}$$

$$\propto \mathrm{Re}\left\{E_1 E_2^* \exp\left[-2ikx \sin\theta\right]\right\}$$

A third beam will then diffract into a different direction.
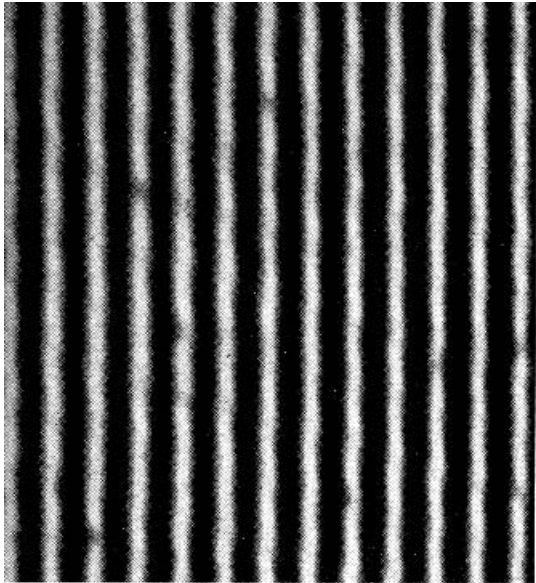This yields a beam that's the product of $E_1$, $E_2^*$, and $E_3$:

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

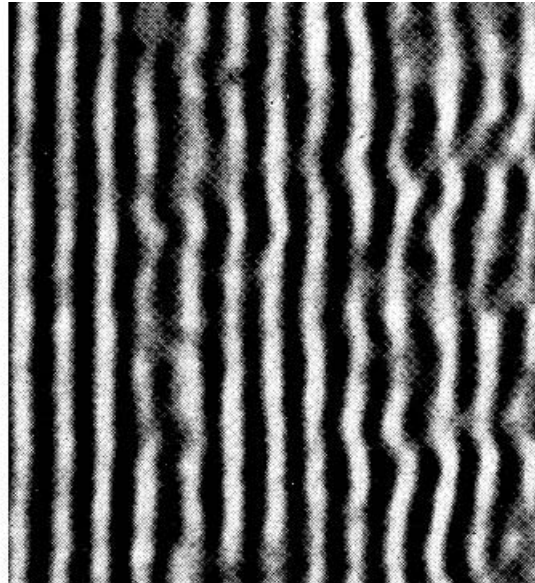This is just a generic four-wave-mixing effect.

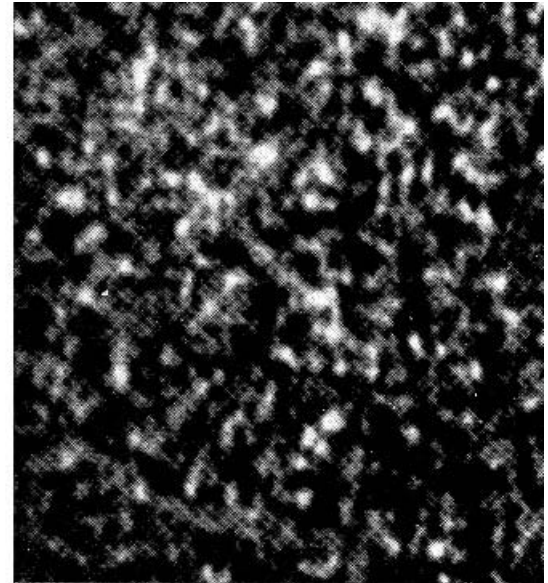# Induced gratings with plane waves and more complex beams

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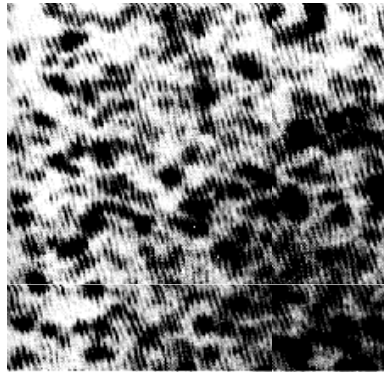
A third beam (a plane wave) diffracts off the grating, acquiring the image information.  Different fringe spacings yield different diffraction angles—hence 3D!
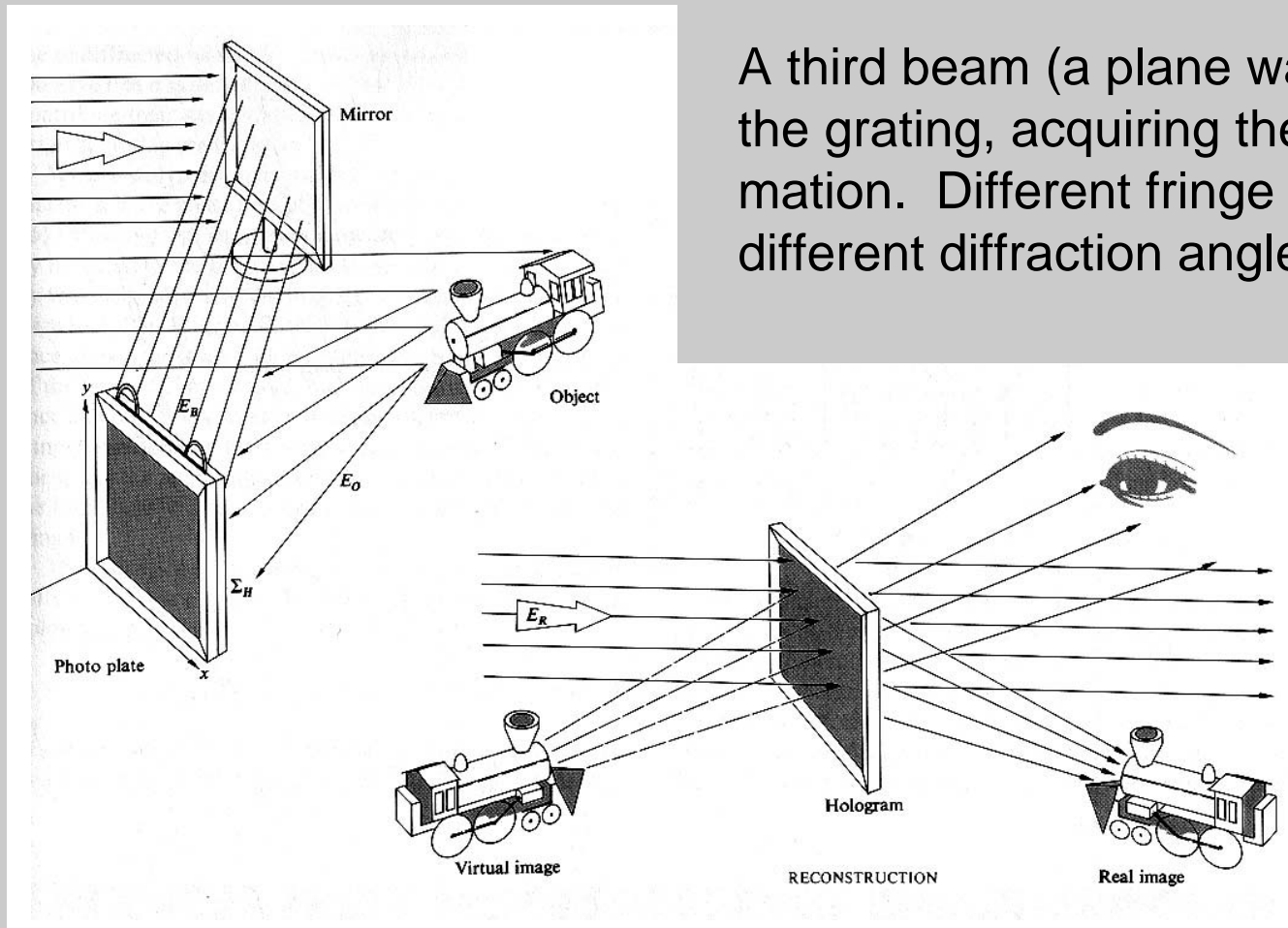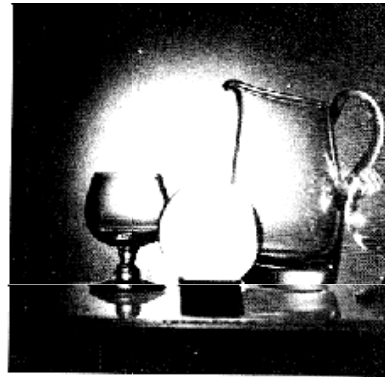


The light phase stores the angular info.
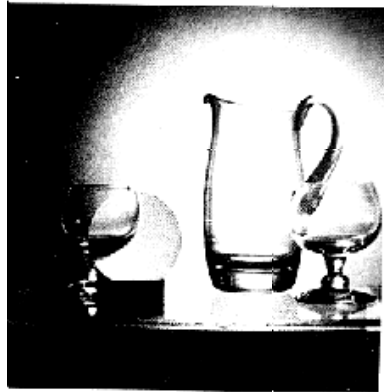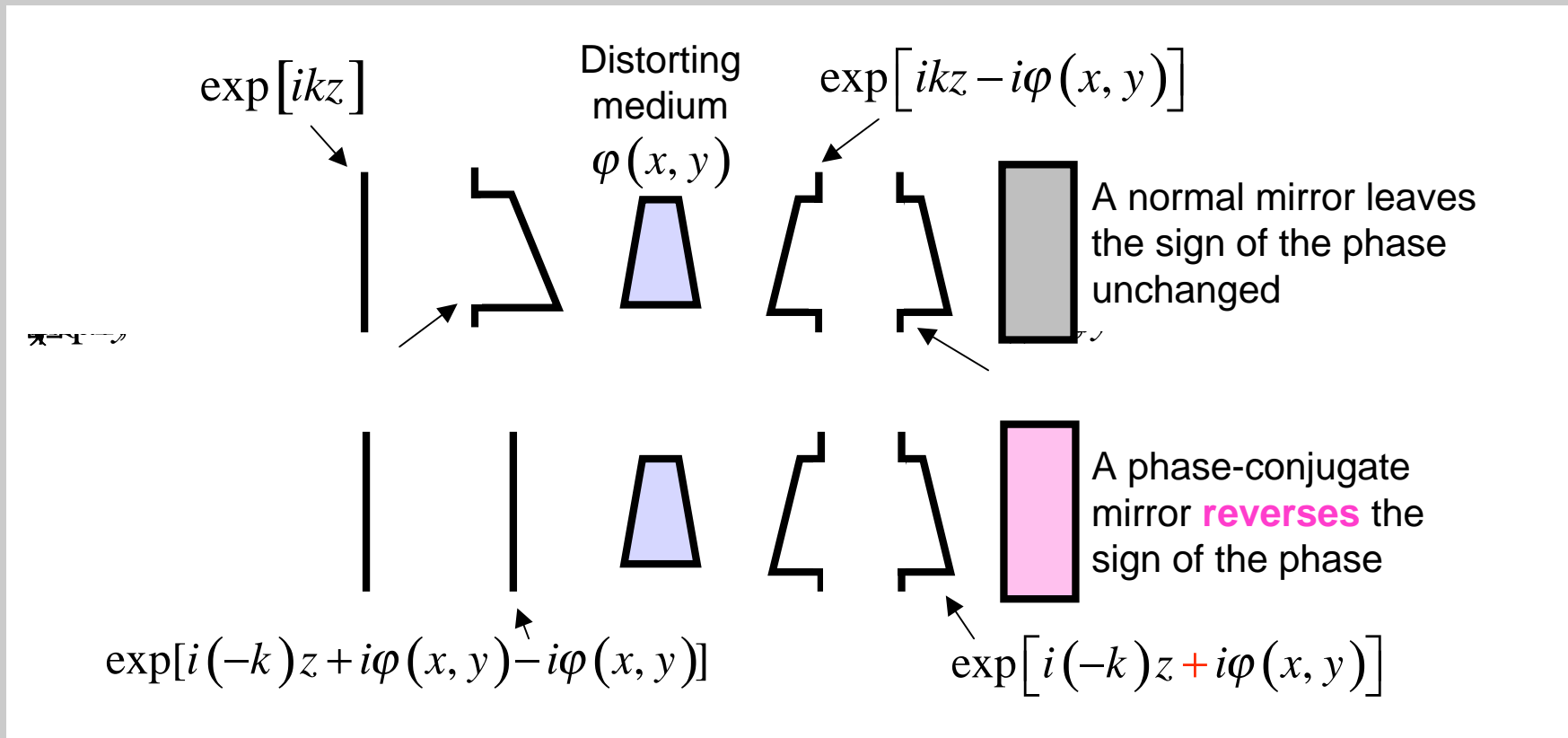
# A hologram and different views of it

The hologram

# 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.

$\exp[ikz]$

Distorting medium

$\varphi(x,y)$

$\exp[ikz - i\varphi(x,y)]$

A normal mirror leaves the sign of the phase unchanged

A phase-conjugate mirror **reverses** the sign of the phase

$\exp[i(-k)z + i\varphi(x,y) - i\varphi(x,y)]$

$\exp[i(-k)z + i\varphi(x,y)]$

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

# Nonlinear Refractive Index

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

$$n = \sqrt{1 + 4\pi\chi^{(1)} + 4\pi\chi^{(3)}\left|E\right|^2}$$

The usual refractive index (which we'll call $n_0$) is: $\quad n_0 = \sqrt{\varepsilon} = \sqrt{1 + 4\pi\chi^{(1)}}$

So: $\quad n = \sqrt{n_0^2 + 4\pi\chi^{(3)}\left|E\right|^2} = n_0\sqrt{1 + 4\pi\chi^{(3)}\left|E\right|^2 / n_0^2}$

Assume that the nonlinear term $<< n_0$ :

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

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

$$\boxed{n \approx n_0 + n_2 I}$$

since $I \propto \left|E\right|^2$

# Self-Phase Modulation & Continuum Generation

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

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

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

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

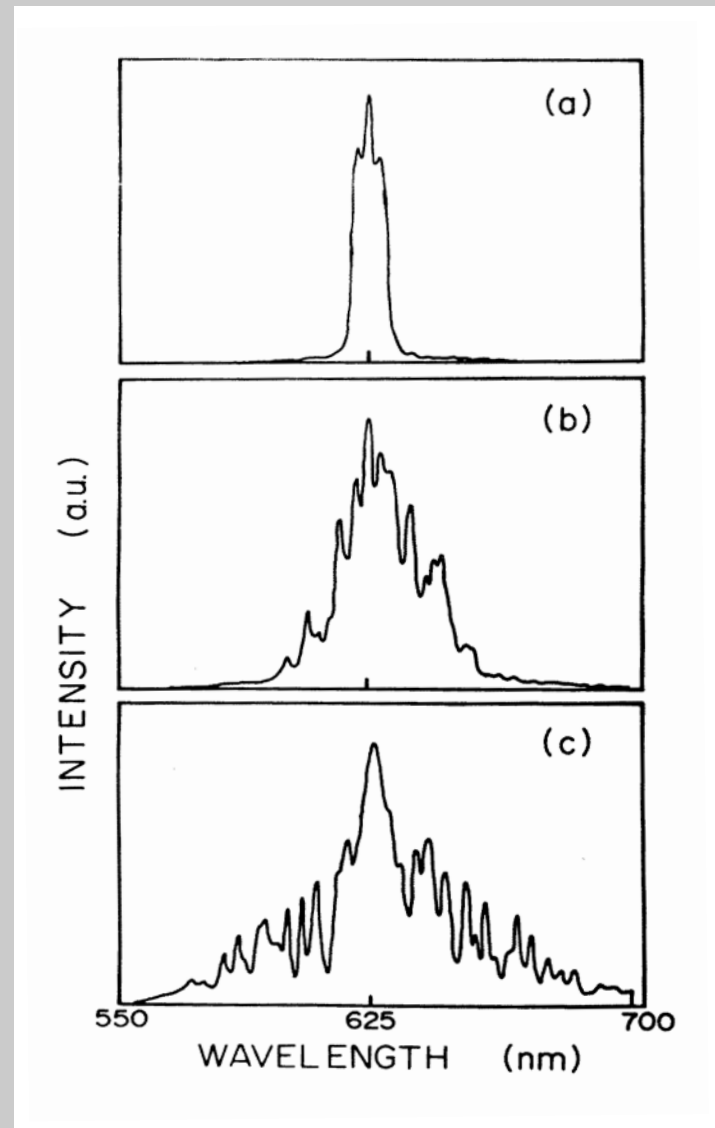Continua created by propagating 500-fs 625-nm pulses through 30 cm of single-mode fiber.
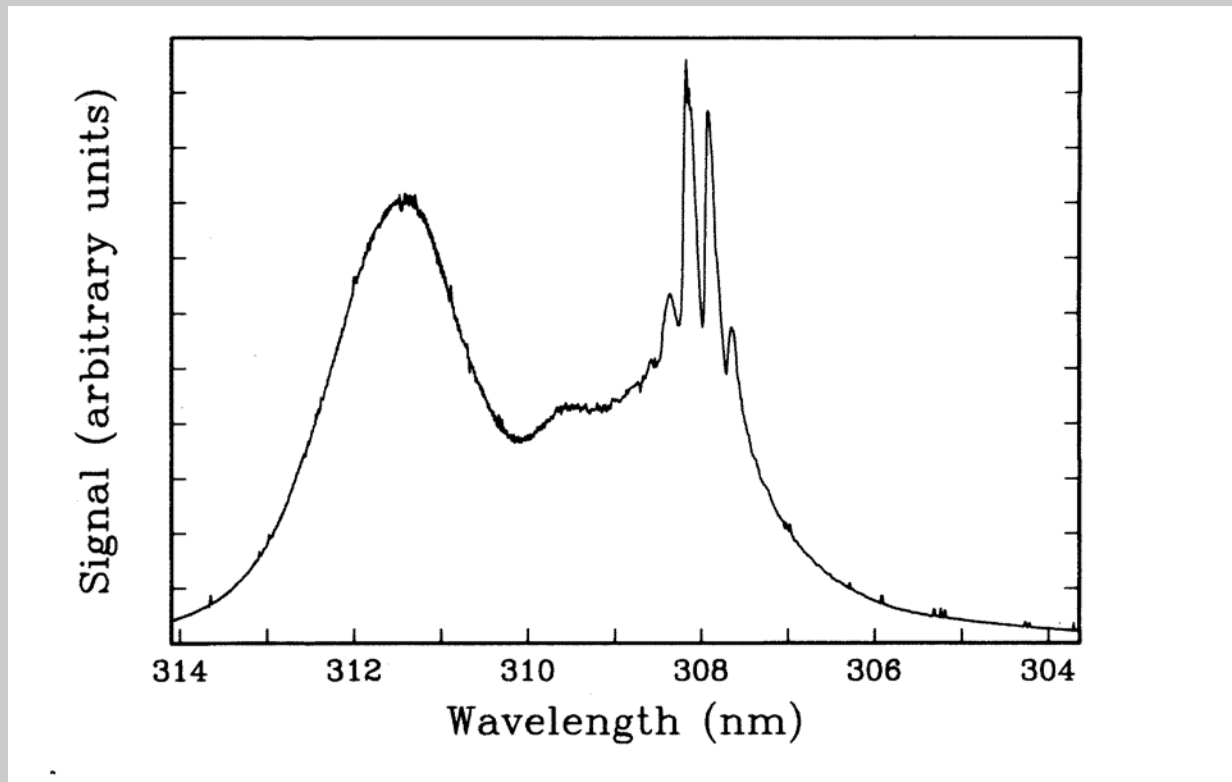
Low Energy

Medium Energy

Broadest spectrum occurs for highest energy.

High Energy

# UV Continuum in Air!

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

# The continuum from microstructure optical fiber is ultrabroadband.
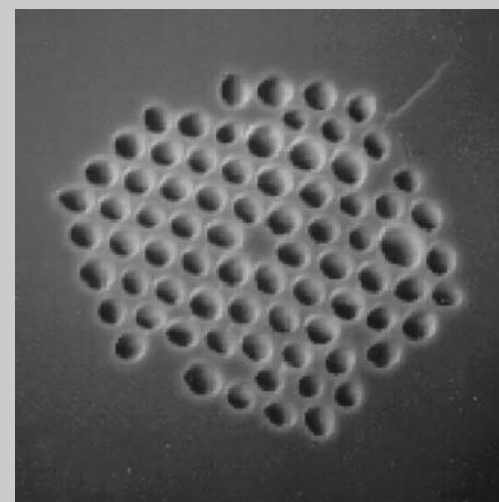


Cross section of the microstructure fiber.
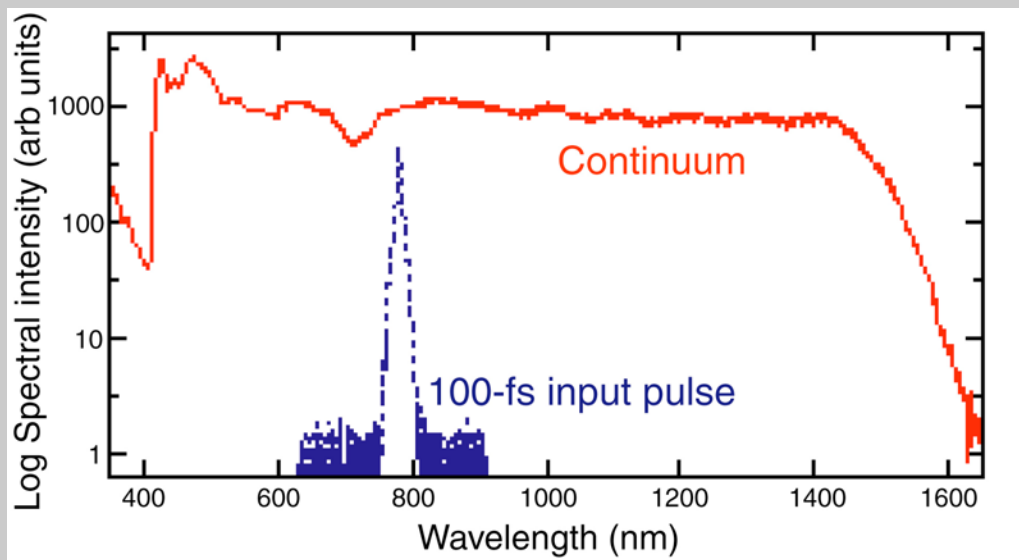


The spectrum extends from ~400 to ~1500 nm and is relatively flat (when averaged over time).

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!**