Nonlinear optics

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

Nonlinear-optical media

Maxwell's equations in a medium

Second-harmonic generation

Sum- and difference frequency generation

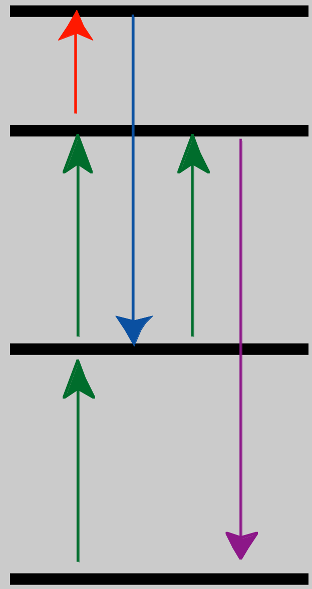
Conservation laws for photons ("Phase-matching")

Induced gratings

Holography

Phase conjugation and aberration cancellation

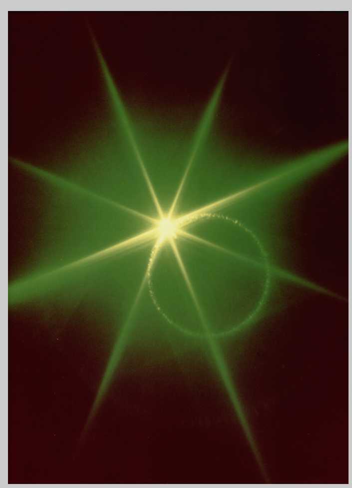
Self-phase modulation



Some 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?

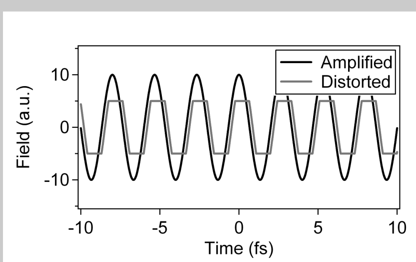
1. Intensities of daily life are too weak.
2. Normal light sources are incoherent.
3. The occasional crystal we see has the wrong symmetry (for SHG).
4. "Phase-matching" is required, and it doesn't usually happen on its own.

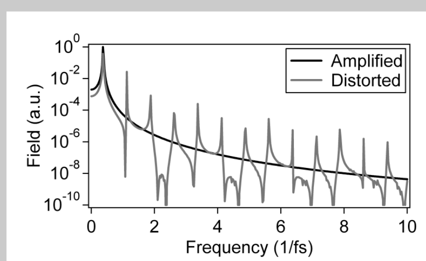
Aspects of NLO

Microscopic response How light couples to the medium	EM propagation How medium affects propagation	Applications/devices How to design systems using NLO
Classical: anharmonic potential	Phase matching	Harmonic generation, sum or difference mixing
Quantum: perturbation	Saturated conversion	Optical parametric amp
Quantum: density matrix	Dispersive propagation	Optical parametric oscill
Molecular rotation, vibrational response	Induced gratings	Optical switching
Semiconductors	Soliton effects,	Pulse compression, cleaning
Free electrons	Self-focusing dynamics	NL response for image contrast

Classical picture for harmonic generation

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”.



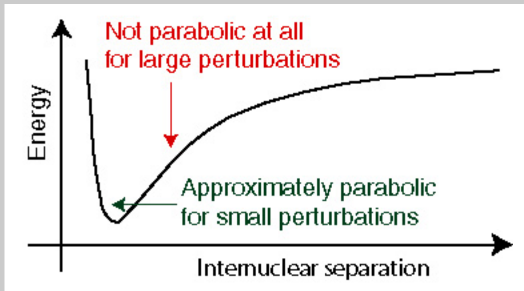


We hear this as distortion.

Nonlinear optics and anharmonic oscillators

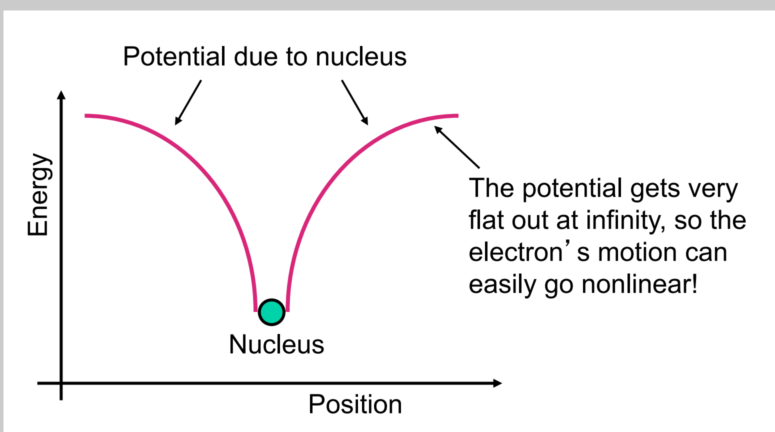
Another way to look at nonlinear optics is that the potential of the electron or nucleus (in a molecule) is not a simple harmonic potential.

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.

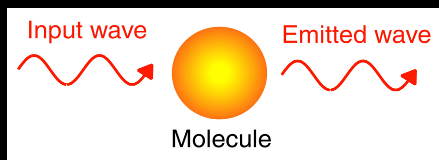
Nonlinear effects in atoms and molecules



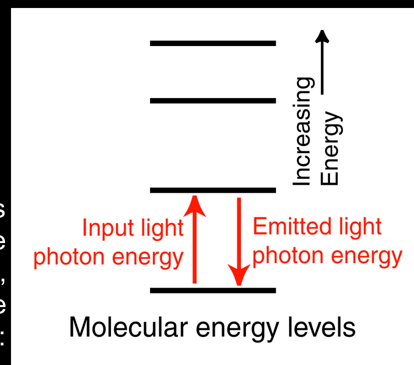
So an electron's motion will also depart from a sine wave.

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.

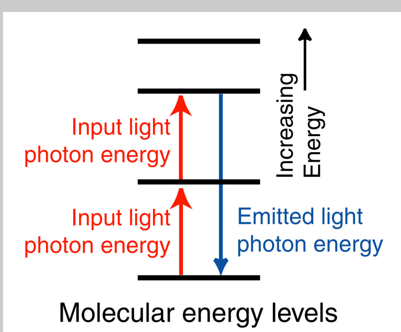


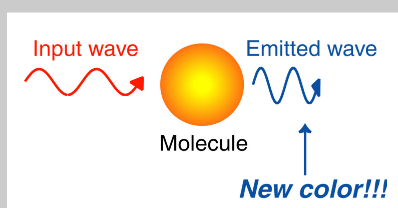
We can also imagine this process in terms of the molecular energy levels, using arrows for the photon energies:



Photon/quantum picture of NLO

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.





Maxwell's Equations in a Medium

- The induced polarization, **P**, contains the effect of the medium:

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

$$\vec{\nabla} \cdot \mathbf{B} = 0 \qquad \vec{\nabla} \times \mathbf{B} = \frac{1}{c^2}\frac{\partial \mathbf{E}}{\partial t} + \mu_0 \frac{\partial \mathbf{P}}{\partial t}$$

These equations reduce to the wave equation:

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

"Inhomogeneous Wave Equation"

- Sinusoidal waves of all frequencies are solutions to the wave equation
- The polarization (**P**) can be thought of as the driving term for the solution to this equation, so the polarization determines which frequencies will occur.

Solving the wave equation in the presence of *linear* induced polarization

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

$$\mathbf{P}(\mathbf{E}) = \varepsilon_0 \chi \mathbf{E}, \quad \mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P} = \varepsilon_0 (1+\chi)\mathbf{E} = \varepsilon\, \mathbf{E} = n^2 \mathbf{E}$$

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

$$\frac{\partial^2 \mathbf{E}}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{c^2}\chi \frac{\partial^2 \mathbf{E}}{\partial t^2}$$

Using the fact that: $\varepsilon_0 \mu_0 = 1/c^2$

Simplifying: $$\frac{\partial^2 \mathbf{E}}{\partial z^2} - \frac{n^2}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = 0$$

This equation has a linearly polarized solution:

$$\mathbf{E}(z,t) = \hat{\mathbf{x}}\, E(0)\cos(kz - \omega t)$$

Where $\omega = k\,c, \quad k = 2\pi n/\lambda, \quad v_{ph} = c/n$

The induced polarization only changes the refractive index.

Linear propagation

- Two waves can propagate independently:

$$\left[\frac{\partial^2}{\partial z^2} - \frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\right]\left(\mathbf{E}_1 + \mathbf{E}_2\right) = 0$$

- This is just like

$$\left[\frac{\partial^2}{\partial z^2} - \frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\right]\mathbf{E}_1 = 0 \qquad \left[\frac{\partial^2}{\partial z^2} - \frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\right]\mathbf{E}_2 = 0$$

So:

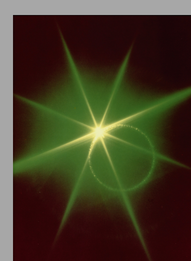
- One wave doesn't affect the other
- Any input frequency stays at that frequency (freq and photon energy are conserved)
- Medium can be non-uniform (gradients, waveguides, …)
- Medium can be birefringent:

$$\ddot{\chi} = \begin{pmatrix} \chi_{xx} & 0 & 0 \\ 0 & \chi_{yy} & 0 \\ 0 & 0 & \chi_{zz} \end{pmatrix} \qquad \ddot{\varepsilon} = \begin{pmatrix} \varepsilon_{xx} & 0 & 0 \\ 0 & \varepsilon_{yy} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{pmatrix}$$

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 (scalars here for now):

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



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

$$E(t) \propto E\exp(-i\omega t) + E^*\exp(i\omega t),$$

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

2w = 2nd harmonic!

In general, all components are tensors: allow for different input, output polarizations

Second-order response with 2 input frequencies

Calculate $P^{(2)} \propto E(t)^2$ with real fields $E = E_1 + E_1^* + E_2 + E_2^*$

Then let $E_1 \to E_1 e^{-i\omega_1 t}$ $E_2 \to E_2 e^{-i\omega_2 t}$

Group terms according to their frequency (including both conjugates) …

and draw arrow energy diagrams for each process.

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:

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

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

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

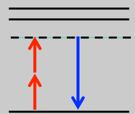
$$+ 2E_1 E_2^* \exp\left[-i(\omega_1 - \omega_2)t\right] + 2E_1^* E_2 \exp\left[i(\omega_1 - \omega_2)t\right]$$ **Diff-freq gen**

$$+ 2\left|E_1\right|^2 + 2\left|E_2\right|^2$$ **dc rectification**

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

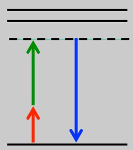
Energy diagrams for second order processes

Second harmonic generation
SHG

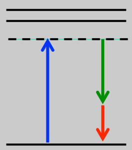


Dashed line: process is non-resonant
= “virtual level”

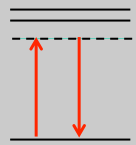
Sum frequency mixing



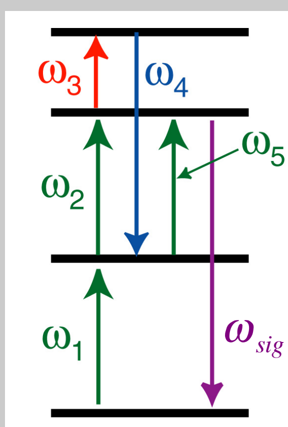
Difference frequency mixing



Optical rectification
e.g. THz generation



More complicated nonlinear-optical effects can occur



Emitted-light frequency

Nonlinear-optical processes are often referred to as:

"N-wave-mixing processes"

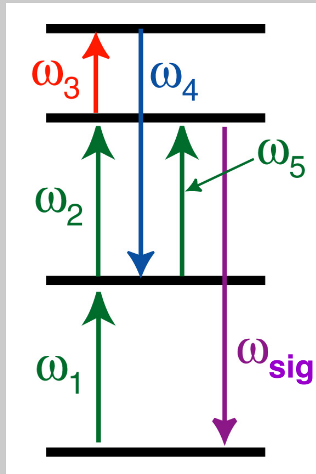
where N is the number of photons involved (including the emitted one). This is a six-wave-mixing process.

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:
 photons are used up,
 contribute a factor of the field, E_i to P
Arrows pointing downward:
 photons are produced
 contribute a factor of the complex conjugate of the field:

$$P = \varepsilon_0 \chi^{(5)} E_1 E_2 E_3 E_4^* E_5$$



Solving the wave equation in nonlinear optics

Recall the inhomogeneous wave equation:

$$\boxed{\frac{\partial^2 \mathbf{E}}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{\partial^2}{\partial t^2}\mathbf{P}(\mathbf{E}) \approx \mu_0 \frac{\partial^2}{\partial t^2}\left(\varepsilon_0 \chi^{(1)}\mathbf{E} + \mathbf{P}^{(2)}\right)}$$

$$\boxed{\rightarrow \frac{\partial^2 \mathbf{E}}{\partial z^2} - \frac{n^2}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{\partial^2}{\partial t^2}\mathbf{P}^{(2)} = \frac{1}{c^2}\chi^{(2)}\frac{\partial^2}{\partial t^2}\mathbf{E}^2}$$

Because it's second-order in both space and time, and **P** is a nonlinear function of **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:

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

Separation-of-frequencies approximation

The total E-field will contain several nearly discrete frequencies, w_1, w_2, etc.

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

$$\frac{\partial^2 E_1}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E_1}{\partial t^2} = \mu_0 \frac{\partial^2 P_1^{(2)}}{\partial t^2}$$

Where E_1 and P_1 are the E-field and polarization at frequency w_1.

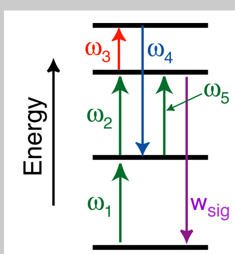
$$\frac{\partial^2 E_2}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E_2}{\partial t^2} = \mu_0 \frac{\partial^2 P_2^{(2)}}{\partial t^2}$$

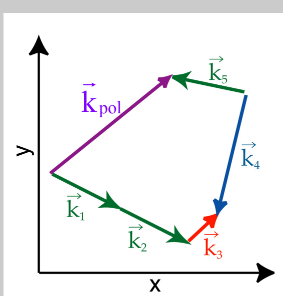
Where E_2 and P_2 are the E-field and polarization at frequency w_2.

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:

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

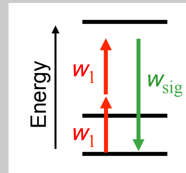
$$\hbar\vec{k}_1 + \hbar\vec{k}_2 + \hbar\vec{k}_3 - \hbar\vec{k}_4 + \hbar\vec{k}_5 = \hbar\vec{k}_{sig}$$

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:

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

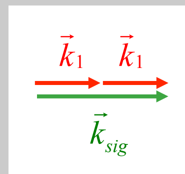


Momentum must also be conserved:

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

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

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



To make the process efficient

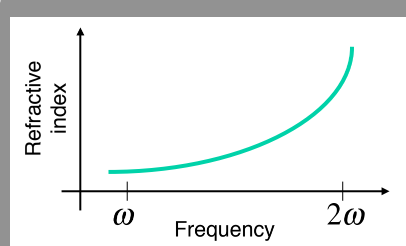
$$\Rightarrow \boxed{n(\omega_1) = n(2\omega_1)}$$ The phase-matching condition for SHG!

Phase-matching Second-Harmonic Generation

The phase-matching condition for SHG:

$$\boxed{n(\omega) = n(2\omega)}$$

Unfortunately, dispersion normally prevents this from ever happening!



First Demonstration of Second-Harmonic Generation

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

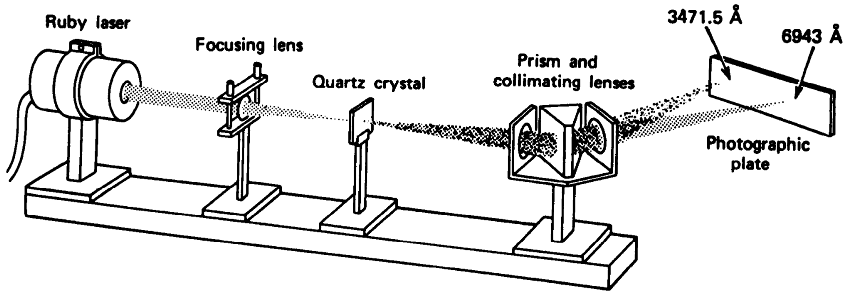


Figure 12.1. Arrangement used in the first experimental demonstration of second-harmonic generation [1]. A ruby-laser beam at $\lambda = 0.694\ \mu m$ is focused on a quartz crystal, causing the generation of a (weak) beam at $\frac{1}{2}\lambda = 0.347\ \mu m$. The two beams are then separated by a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

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.

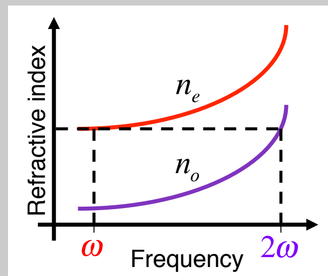
Phase-matching Second-Harmonic Generation using birefringence

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 w and the ordinary for $2w$:

$$\boxed{n_e(\omega) = n_o(2\omega)}$$

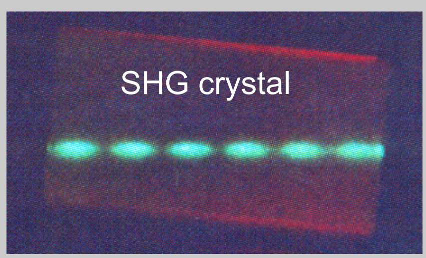


n_e depends on propagation angle, so we can tune for a given w. Some crystals have $n_e < n_o$, so the opposite polarizations work.

Light created in real crystals

Far from phase-matching:

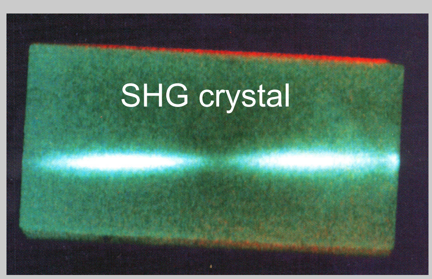
Input beam



Output beam

Closer to phase-matching:

Input beam



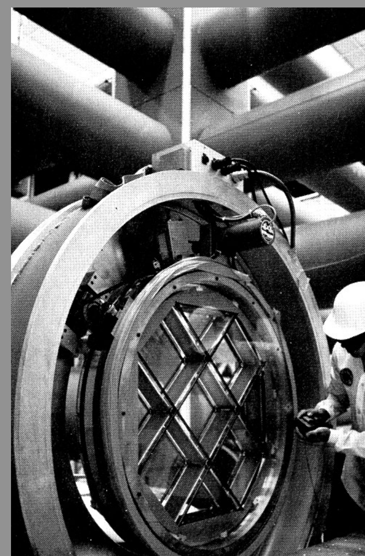
Output beam

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

Second-Harmonic Generation engineered for high conversion

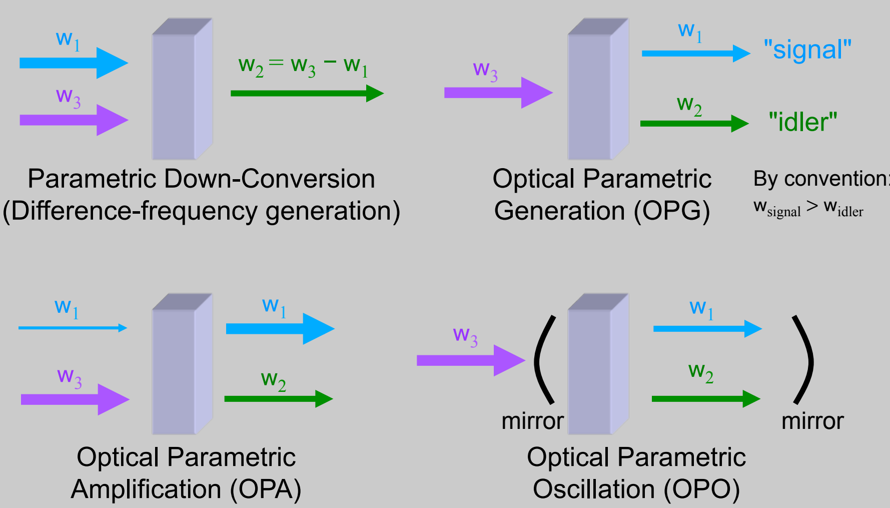
SHG KDP crystals at Lawrence Livermore National Laboratory

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.

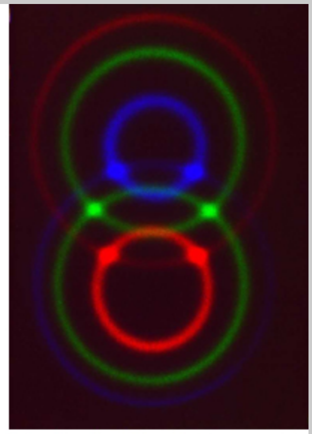


Spontaneous parametric down conversion

Crystal "splits" a photon into two.

The quantum properties of the two new photons are entangled.

Source for quantum optics experiments.



This image of light from a down-coversion crystal shows the spatial emission directions of the entangled photons. Photons emitted at the intersection of the two green rings are entangled in polarization, as well as energy.

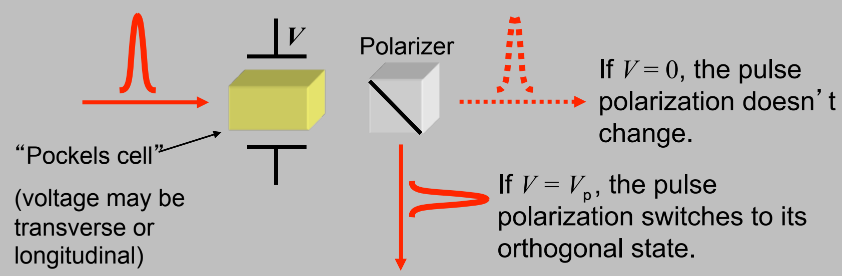
OPCPA: optical parametric chirped pulse amplification

OPA can be an alternative to laser amplification: more bandwidth, no crystal heating, flexibility on output wavelength….

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: $n_0 = \sqrt{1 + \chi^{(1)}}$

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 << n_0:

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

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

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

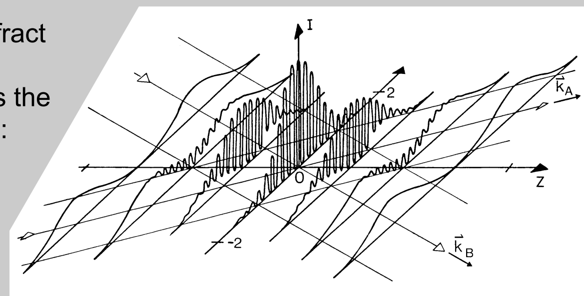
An induced grating results from the cross term in the irradiance:

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

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:

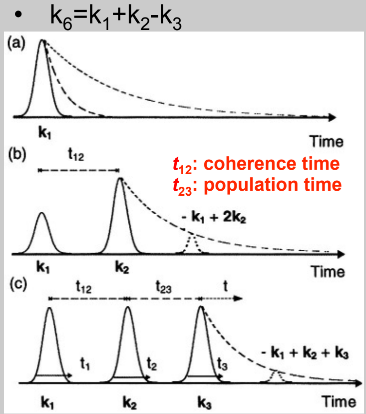
$$E_{sig} \propto \left(E_1 E_2^*\right)E_3$$

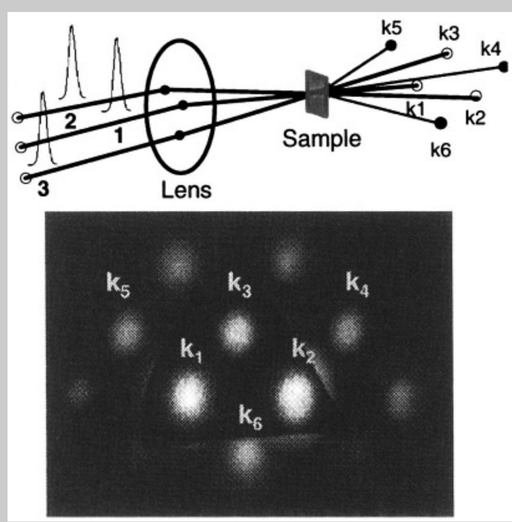
This is just a generic four-wave-mixing effect.



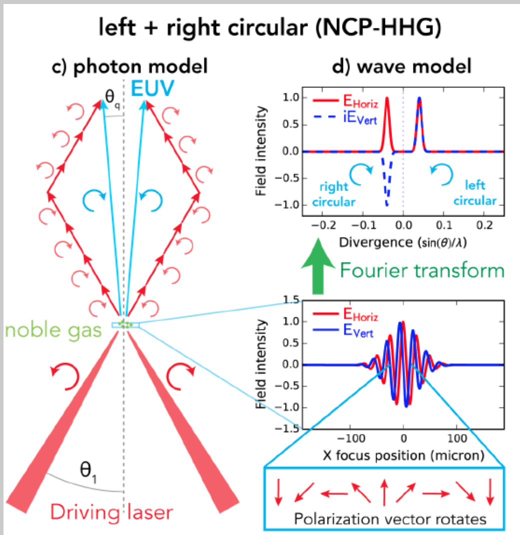
Transient gratings and coherent spectrosopy

- Pump-probe experiments can investigate properties of materials
- Three main output directions
- $k_4 = -k_1+k_2+k_3$
- $k_5=k_1-k_2+k_3$
- $k_6=k_1+k_2-k_3$





Photon vs wave model of noncollinear circularly polarized high-order harmonic geneneration

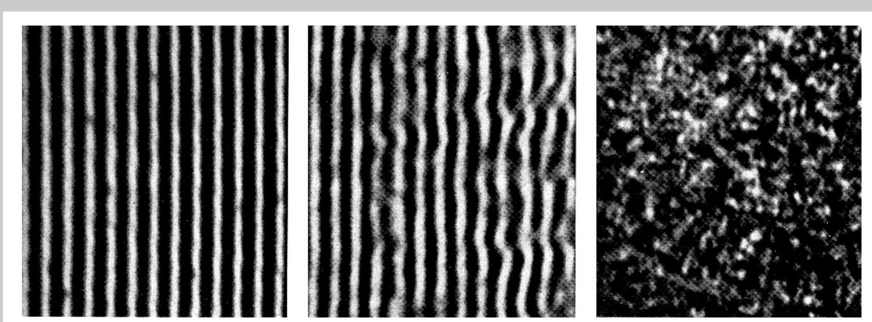


- "orthogonal polarizations don't interfere"
- But R+L = linear, and orientation depends on relative phase
- Pump is *linear* at the atomic level

Hickstein et. al., *Nature Photonics 9, 743–750 (2015)*

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



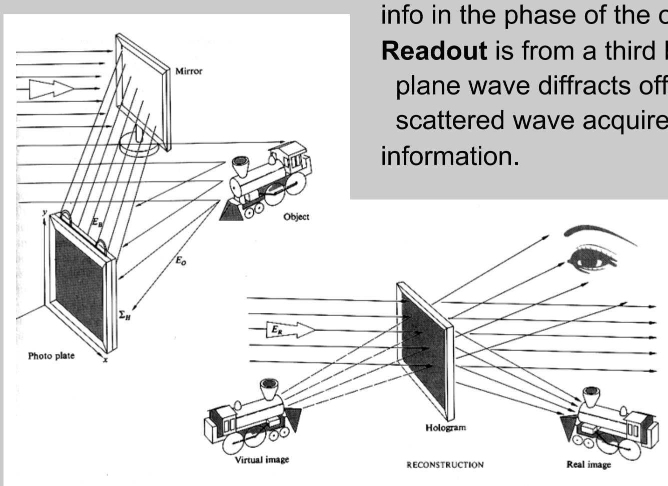
All such induced gratings will diffract a plane wave, reproducing the distorted wave.

Holography is an induced-grating process.

Recording is interference of
smooth reference beam +
scattered object beam

Interferogram = hologram records 3D info in the phase of the object field

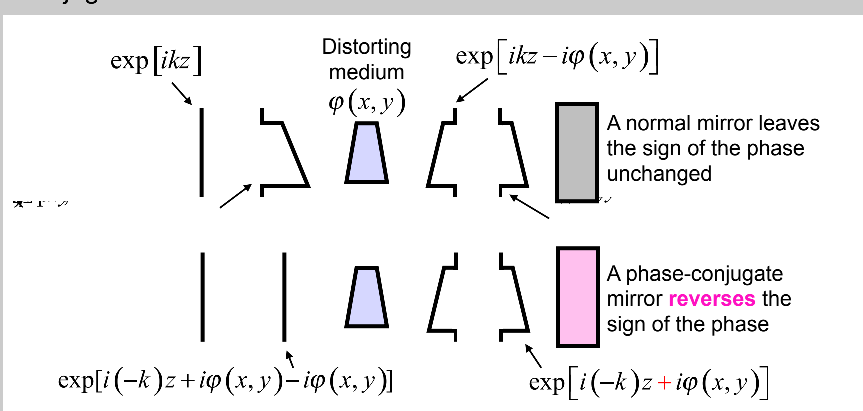
Readout is from a third beam
plane wave diffracts off the grating
scattered wave acquires the image information.



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.

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

$$\propto E_{sig}(0,t)\exp\left[i n_2 k I(t)\, z\right]$$

Pulse Intensity vs. time

The further the pulse travels, the more modulation occurs.

That is: $\boxed{\phi(z,t) = n_2 k\,z\,I(t)}$

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.

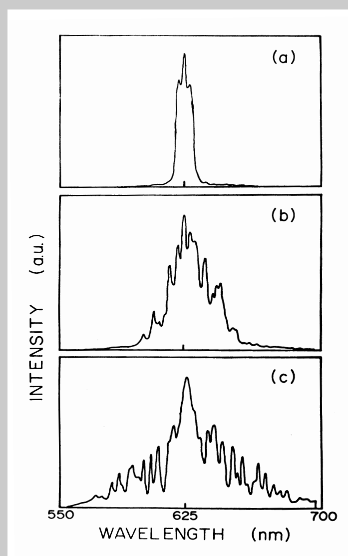
The Supercontinuum Laser Source, Alfano, ed.

Broadest spectrum occurs for highest energy.

Low Energy

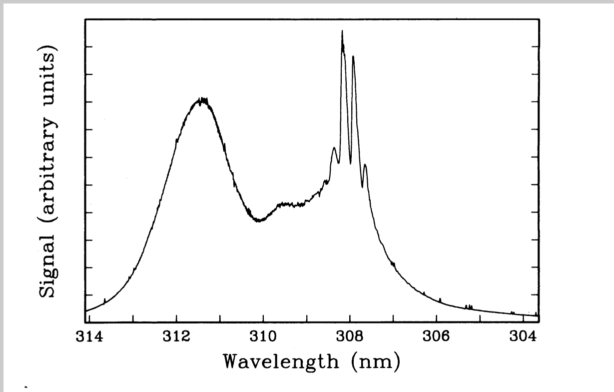
Medium Energy

High Energy



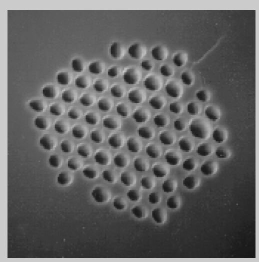
UV Continuum in Air!

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

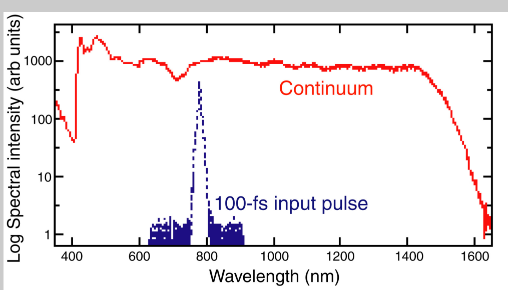


The Supercontinuum Laser Source, Alfano, ed.

The continuum from microstructure optical fiber is ultrabroadband.



Cross section of the microstructure fiber.

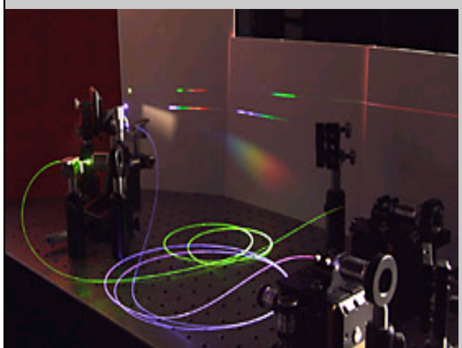


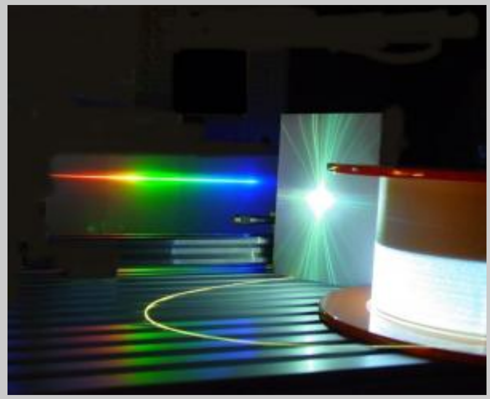
The spectrum extends from ~400 to ~1500 nm and is relatively flat (when averaged over time).

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!





Nonlinear wave equation with degenerate FWM

- With degenerate frequencies, NL equation is

$$\frac{\partial^2 \mathbf{E}}{\partial z^2} - \frac{n^2}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{c^2}\chi^{(3)}\frac{\partial^2}{\partial t^2}\left(\left|\mathbf{E}\right|^2 \mathbf{E}\right)$$

- With slowly-varying envelope equation and dispersion, we get the nonlinear Schrodinger equation:

$$\frac{\partial A}{\partial z} + i\frac{\beta_2}{2}\frac{\partial^2 A}{\partial t^2} = i\gamma\left|A\right|^2 A$$