Nonlinear Optics

ECE 455 Optical Electronics

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ECE Illinois

Introduction

In this section, the following subjects will be covered:

- What is meant by 'nonlinear optics'
- Second and third order optical nonlinear effects
- Applications of nonlinear phenomenon

Linear Optics I

- To understand what is meant by **nonlinear**, it is first necessary to understand what is meant by **linear**
- The electric displacement field is most generally written as:

$$D_i(\omega) = \epsilon_0 E_i(\omega) + P_i(\omega) \tag{1}$$

- In linear optics, it is assumed that the dipole induced by an electric field is linearly proportional the the strength of that field:

$$P_i(\omega) = \epsilon_0 \chi_{ij}^{(1)}(\omega : \omega_1) E_j(\omega_1) \tag{2}$$

- This results from assuming the effect of the optical electric field on the medium is *perturbative*
- The net effect of the induced polarization is to change the effective speed of light in the material as:

$$n(\omega) = \sqrt{\chi^{(1)}(\omega) + 1} \tag{3}$$

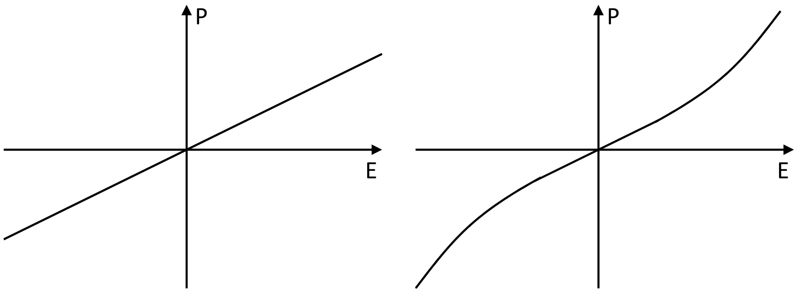
Linear Optics II

- Linear optical phenomenon are characterized by interactions of materials with a single photon
- All of the phenomenon discussed in this class so far are linear optic effects
- Pulses sent through linear materials may be amplified and phase distorted, but the output frequency is equal to the input frequency
- Linear optical phenomenon are independent of the intensity of the field
- Lenses, mirrors, prisms, gratings, optical fibers, and cavities can all be described by the theory of linear optics.

Nonlinear Optics

- Nonlinear optical phenomenon are characterized by interactions of materials with multiple photons
- Nonlinear optical phenomenon are in general a strong function of the intensity
- When any system is driven hard enough, it will exhibit nonlinear behavior.
- With ordinary light intensities, nonlinear optic effects are too small to be noticed

Linear and Nonlinear Behavior



Linear material response

Nonlinear material response

Generation of New Frequencies I

The polarization response can be written as a power series:

$$P = \epsilon_0 \left[\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots \right] \tag{4}$$

Suppose the input field is:

$$E_{in}(t) = E_0 cos(\omega_0 t) \tag{5}$$

We know the $\chi^{(1)} E$ term is the linear part of the polarization and is responsible for the index of refraction. The polarization from this linear term is:

$$P^{(1)}_{out}(t) = \epsilon_0 \chi^{(1)} E_0 cos(\omega_0 t) \tag{6}$$

Generation of New Frequencies II

The polarization from the $\chi^{(2)}$ term is:

$$\begin{aligned} P_{out}^{(2)}(t) &= \epsilon_0 \chi^{(2)} E_0^2 cos^2(\omega_0 t) \\ &= \epsilon_0 \chi^{(2)} E_0^2 \frac{1}{2} \left[1 + cos(2\omega_0 t)\right] \end{aligned} \quad (7)$$

There are two new frequencies which weren't present in the original signal: $2\omega_0$ and 0. Now consider the cubic term

$$\begin{aligned} P_{out}^{(3)}(t) &= \epsilon_0 \chi^{(3)} E_0^3 cos^3(\omega_0 t) \\ &= \epsilon_0 \chi^{(3)} E_0^3 \frac{1}{2} cos(\omega_0 t) \left[1 + cos(2\omega_0 t)\right] \\ &= \epsilon_0 \chi^{(3)} E_0^3 \frac{1}{2} \left[cos(\omega_0 t) + cos(\omega_0 t) cos(2\omega_0 t)\right] \\ &= \epsilon_0 \chi^{(3)} E_0^3 \left[\frac{3}{4} cos(\omega_0 t) + \frac{1}{4} cos(3\omega_0 t)\right] \end{aligned}$$

Example: New Frequencies

Problem: Two lasers with frequencies ω_1 and ω_2 pass through a material with a $\chi^{(3)}$ coefficient. Find all frequencies at which there is a polarization response.

Solution: If we were to continue as we have been doing, we would expand

$$P_{out}^{(3)} = \epsilon_0 \chi^{(3)} \left(E_0 cos(\omega_1 t) + E_0 cos(\omega_2 t) \right)^3 \tag{8}$$

and simplify it with sine and cosine identities.

However, we can recognize that when sines and cosines are multiplied, they generate frequencies with the sum and difference of the arguments. The polarization response frequencies will be:

$$\omega \in \{\omega_1, 3\omega_1, \omega_2, 3\omega_2, |2\omega_1 \pm \omega_2|, |\omega_1 \pm 2\omega_2|\} \tag{9}$$

General Rule for finding new frequencies

Our insight from the last example can be formalized with the aid of Fourier transform theory: Multiplication in the time domain is convolution in the frequency domain.

In general, if n frequencies are incident upon a medium with a k^{th} order nonlinearity, the frequencies at which the material will respond are:

$$\omega_{out} = |\omega_i \pm \omega_j ... \pm \omega_q| \tag{10}$$

where $\omega_i, \omega_j, ...\omega_q \in \{\omega_1, \omega_2, ..., \omega_n\}$.

In practice, most of these frequencies will not be observed because they will destructively interfere macroscopically.

Einstein Notation I

The linear response of a material to an optical field may in general be represented with the vector equation

$$\vec{P}^{(1)}(\omega) = \epsilon_0 \bar{\chi}^{(1)}(\omega)\vec{E}(\omega) \tag{11}$$

which is a compact way of writing

$$\begin{bmatrix} P_x(\omega) \\ P_y(\omega) \\ P_z(\omega) \end{bmatrix} = \epsilon_0 \begin{bmatrix} \chi_{xx}(\omega) & \chi_{xy}(\omega) & \chi_{xz}(\omega) \\ \chi_{yx}(\omega) & \chi_{yy}(\omega) & \chi_{yz}(\omega) \\ \chi_{zx}(\omega) & \chi_{zy}(\omega) & \chi_{zz}(\omega) \end{bmatrix} \begin{bmatrix} E_x(\omega) \\ E_y(\omega) \\ E_z(\omega) \end{bmatrix} \tag{12}$$

The above matrix equation doesn't seem so bad; The tensor has 9 terms (not all independent). However the tensor for the second-order nonlinear response contains 27 terms!

We need a compact way of representing these equations.

Einstein Notation II

Repeated subscript indices imply summation over spatial coordinates x, y and z. The following equation represents the Einstein notation for the linear polarization.

$$P_i(\omega) = \epsilon_0 \chi_{ij}^{(1)}(\omega : \omega_1) E_j(\omega_1) \tag{13}$$

For example, when considering the y coordinate, this becomes

$$\begin{aligned} P_y(\omega) &= \epsilon_0 \Big[\chi_{yx}^{(1)}(\omega : \omega_1) E_x(\omega_1) + \chi_{yy}^{(1)}(\omega : \omega_1) E_y(\omega_1) \\ &\quad + \chi_{yz}^{(1)}(\omega : \omega_1) E_z(\omega_1) \Big] \end{aligned} \tag{14}$$

Equations 11, 12, and 13 are all equivalent ways of expressing the same idea.

The $\chi^{(n)}$ Tensors

The expression

$$\chi^{(2)}_{ijk}(\omega : \omega_1, \omega_2) \tag{15}$$

should be interpreted in the following manner:

The coefficient of the material response at frequency ω and polarization i due to fields with frequencies ω_1 and ω_2 and polarizations j and k respectively.

The Nonlinear Wave Equation

In a sourceless material medium, Maxwell's four equations are

$$\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \quad (16)$$

$$\nabla \times \vec{H} = \frac{\partial \vec{D}}{\partial t} \quad (17)$$

$$\nabla \cdot \vec{B} = 0 \quad (18)$$

$$\nabla \cdot \vec{D} = 0 \quad (19)$$

If we take the curl of Equation 16 and assume a non-magnetic material ($\vec{B} = \mu_0 \vec{H}$), then we may substitute Equation 17 into Equation 16

$$\nabla \times \nabla \times \vec{E} = -\mu_0 \frac{\partial^2 \vec{D}}{\partial t^2} \quad (20)$$

The Nonlinear Wave Equation

In linear optics, the next step would be to substitute $\vec{D} = \bar{\epsilon}\vec{E}$ and go on our merry way, **but** the relationship between $\vec{D}$ and $\vec{E}$ is more complicated in nonlinear optics.

$$\begin{aligned} \vec{D} &= \epsilon_0 \vec{E} + \vec{P} && (21) \\ &= \epsilon_0 \vec{E} + \vec{P}^{(1)} + \vec{P}^{NL} && (22) \\ &= \epsilon_0 \left(1 + \bar{\chi}^{(1)}\right) \vec{E} + \vec{P}^{NL} && (23) \\ &= \epsilon_0 \bar{\epsilon}^{(1)} \vec{E} + \vec{P}^{NL} && (24) \end{aligned}$$

substituting this back into Equation 20

$$\nabla \times \nabla \times \vec{E} + \frac{\bar{\epsilon}^{(1)}}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = -\frac{1}{\epsilon_0 c^2}\frac{\partial^2 \vec{P}^{NL}}{\partial t^2} \qquad (25)$$

The Nonlinear Wave Equation

The next step in deriving the nonlinear wave equation is to apply the vector identity $\nabla \times \nabla \times \vec{E} = \nabla \cdot \left(\nabla \vec{E}\right) - \nabla^2 \vec{E}$. However, unlike linear optics, the equation $\nabla \cdot \left(\nabla \vec{E}\right) = 0$ is only approximate. However it is a good approximation as long as the pulses are not too 'short.' The resulting wave equation is

$$-\nabla^2 \vec{E} + \frac{\overline{\epsilon}^{(1)}}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = -\frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}^{NL}}{\partial t^2} \tag{26}$$

What would the corresponding linear wave equation look like?

$\chi^{(2)}$ Processes

- In order to possess even-ordered nonlinear coefficients, the material must lack inversion symmetry
- All nonlinear processes must obey the conservation of energy:

$$\omega_3 = \omega_1 + \omega_2 \tag{27}$$

- They must also obey the conservation of momentum:

$$\vec{k_3} = \vec{k_1} + \vec{k_2} \tag{28}$$

- By convention

$$\omega_3 > \omega_2 \geq \omega_1 \tag{29}$$

- In the context of frequency mixing, ω_3, ω_2 and ω_1 are referred to as the pump, signal, and idler respectively

Second Harmonic Generation

- The simplest $\chi^{(2)}$ interaction is second harmonic generation (SHG)
- Two photons from the source are combined into a single photon
- $\hbar\omega_1 + \hbar\omega_1 \rightarrow \hbar(2\omega_1)$

Second Harmonic Generation Math I

Let us examine the mathematics behind SHG. Let ω_1 be the fundamental optical frequency, and define $k_1 \equiv k(\omega_1) = \frac{\omega_1 n_1}{c}$ and $k_2 \equiv k(2\omega_1) = \frac{2\omega_1 n_2}{c}$. The electric field in the medium will consist of two parts: the fundamental frequency and its second harmonic.

$$E(z,t) = \frac{1}{2}\left[A_1(z)e^{\imath(\omega_1 t - k_1 z)} + A_2(z)e^{\imath(2\omega_1 t - k_2 z)} + c.c\right] \quad (30)$$

The nonlinear polarization will then be:

$$\begin{aligned} P_{NL}(z,t) &= 2\epsilon_0 d_{eff} E^2(z,t) \\ &= \frac{\epsilon_0 d_{eff}}{2}\left[A_1^2 e^{\imath(2\omega_1 t - 2k_1 z)} + 2A_2A_1^* e^{\imath(\omega_1 t - (k_2 - k_1)z)} + c.\right. \end{aligned}$$

Note some terms of the nonlinear polarization (such as $A_2^2 e^{\imath(4\omega_1 t - 2k_2 z)}$) have been discarded. We'll see why in a few slides

Second Harmonic Generation Math II

Substituting the above into the nonlinear wave equation (26), we obtain:

$$\frac{1}{2}\left[-\frac{d^2A_1}{dz^2} + \imath 2k_1\frac{dA_1}{dz} + k_1^2A_1 - \left(\frac{\omega_1 n_1}{c}\right)^2 A_1\right]\cdot e^{\imath(\omega_1 t - k_1 z)} +$$
$$\frac{1}{2}\left[-\frac{d^2A_2}{dz^2} + \imath 2k_2\frac{dA_1}{dz} + k_2^2A_2 - \left(\frac{\omega_2 n_2}{c}\right)^2 A_2\right]\cdot e^{\imath(2\omega_1 t - k_2 z)} +$$
$$+c.c. = \frac{d_{eff}}{c^2}\left[2\omega_1^2A_1^2e^{\imath(2\omega_1 t - 2k_1 z)} + \omega_1^2A_2A_1^*e^{\imath(\omega_1 t - (k_2-k_1)z)} + c.c.\right]$$

The above equation looks horrible, but fortunately, it can be simplified:

- First note that since $k_i \equiv \frac{\omega_i n_i}{c}$, the last two terms in the brackets both cancel.
- It will also be assumed that $\frac{d^2A_i}{dz^2} \ll k_i\frac{dA_i}{dz}$. This is commonly known as the **slowly varying envelope approximation**

Second Harmonic Generation Math III

- From phase matching considerations, the equation can be broken into two equations: the parts that vary as $e^{\imath\omega_1 t}$ and those which vary as $e^{\imath 2\omega_1 t}$

$$\frac{dA_1}{dz} = \frac{-\imath\omega_1 d_{eff}}{n_1 c} A_2 A_1^* e^{-\imath\Delta kz} \quad (32)$$

$$\frac{dA_2}{dz} = \frac{-\imath\omega_1 d_{eff}}{n_2 c} A_1^2 e^{\imath\Delta kz} \quad (33)$$

where $\Delta k \equiv k_2 - 2k_1$

Equations 41 and 42 are coupled nonlinear differential equations.

The Undepleted Pump Approximation

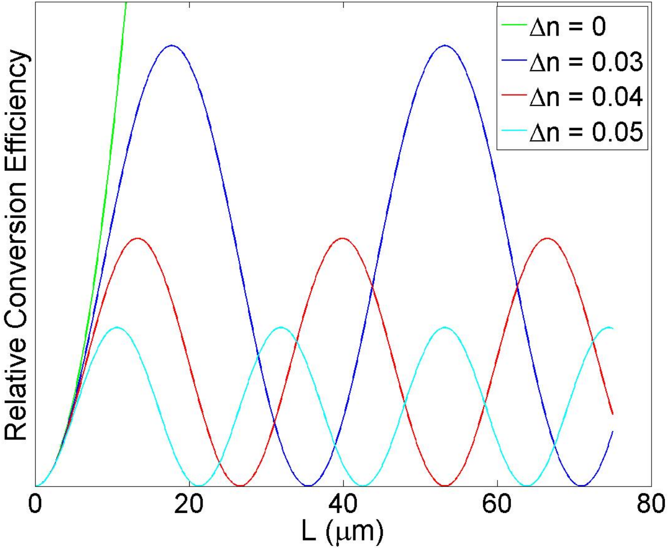
In the limit of low conversion efficiency A_1 can be considered constant. In this limit, Equation 42 can be integrated simply:

$$\begin{aligned} A_2(L) &= \frac{-\imath\omega_1 d_{eff}}{n_2 c} A_1^2 \int_0^L e^{\imath\Delta k z} dz \\ &= \frac{-\imath 2\omega_1 d_{eff}}{n_2 c \Delta k} A_1^2 e^{\frac{\imath \Delta k L}{2}} sin\left(\frac{\Delta k L}{2}\right) \end{aligned} \tag{34}$$

The conversion efficiency is then

$$\frac{I_{2\omega_1}}{I_{\omega_1}} = \frac{A_2^* A_2}{A_1^* A_1} = \left(\frac{2\omega_1 d_{eff}}{n_2 c \Delta k}\right)^2 |A_1|^2 sin^2\left(\frac{\Delta k L}{2}\right) \tag{35}$$

Second Harmonic Generation Picture



Final Thoughts on SHG

- Small index differences severely lower conversion efficiency: The $\Delta n = 0$ curve on the previous slide is far off the scale.
- $\Delta n = 0$ is known at the **phase matching** condition
- Processes which are not phase matched are not macroscopically observed because their efficiency is low
- With careful design, conversion efficiencies approaching 100% can be achieved
- For extremely short crystals, Δn has no effect!

Visual Image of Phase Matching Condition

Birefringent Media I

- Obeying the conservation of momentum is non-trivial because in general n is a function of ω

$$k(\omega) = \frac{\omega n(\omega)}{c} \tag{36}$$

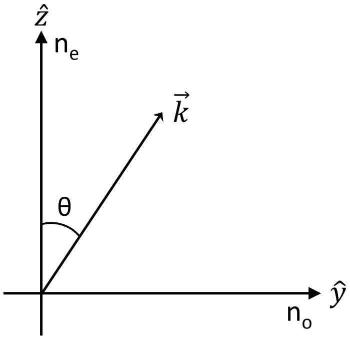
- Therefore in general $k(\omega_1 + \omega_2) \neq k(\omega_1) + k(\omega_2)$
- In order to overcome this difficulty, the properties of birefringent crystals are used
- Uniaxial media have indices of refraction of the form:

$$\bar{n} = \begin{bmatrix} n_o & 0 & 0 \\ 0 & n_o & 0 \\ 0 & 0 & n_e \end{bmatrix} \tag{37}$$

Birefringent Media II

- For a plane wave whose $\vec{k}$ vector makes an angle θ with the $\hat{z}$ axis, there will be two indices of refraction, depending on the polarization of the wave:
  - The ordinary index of refraction is n_o
  - The extraordinary index of refraction can be found with the following equation:

$$\frac{1}{n^2(\theta)} = \frac{sin^2(\theta)}{n_e^2} + \frac{cos^2(\theta)}{n_o^2} \tag{38}$$



Birefringent Media III

- **Type I Phasematching** involves the following two types of interactions
  - o + o → e
  - e + e → o
- **Type II Phasematching**
  - o + e → o
  - o + e → e

Second Harmonic Generation Example I

Problem: BBO is a negative uniaxial crystal ($n_e < n_o$). The indices of refraction for the ordinary and extraordinary axes are given by:

$$n_o^2(\lambda) = 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\lambda^2$$

$$n_e^2(\lambda) = 2.3753 + \frac{0.01224}{\lambda^2 - 0.01667} - 0.01516\lambda^2$$

where λ is expressed in μm. Find the phasematching angle to create the second harmonic of 780 nm light.

Solution: Because BBO is negative uniaxial, the 780 nm must be input as the ordinary wave. The frequency doubled output will be an extraordinary wave.

Second Harmonic Generation Example II

Evaluating the previous two equations, we find the indices of refraction to be:

$$n_o(780\text{ nm}) = 2.7595$$

$$n_o(390\text{ nm}) = 2.8741$$

$$n_e(390\text{ nm}) = 2.4634$$

Setting the ordinary index at 780 nm to be equal to the extraordinary index at 390 nm yields the following equation:

$$\frac{1}{n_o^2(780\text{ nm})} = \frac{1}{n^2(\theta_{pm})} = \frac{sin^2(\theta_{pm})}{n_e^2(390\text{ nm})} + \frac{cos^2(\theta_{pm})}{n_o^2(390\text{ nm})}$$

Solving this equation for θ_{pm} yields:

$$\theta_{pm} = 30^\circ$$

Non-critical Phase Matching

- Angle tuning a crystal to for phasematching purposes is known as critical phase matching
- Critical phase matching is sensitive to misalignment, and suffers from problems such as walk off.
- In special cases, the phasematching can be tuned by varying the temperature and composition of the crystal
- Periodically Poled Crystals

$$I_{2\omega} = \frac{2\omega_0^2 d_{eff}^2}{\epsilon_0 c^3 n^3} I_\omega^2 sinc^2\left(\frac{\Delta kL}{2}\right) \tag{39}$$

- Efficiency is not as good as angle phase matching, but this process is more versatile
- ADD PICTURE OF PPLN crystal and its response

Frequency Mixing

- When $\omega_1 \neq \omega_2$
- The propagation of

$$\frac{dA_1}{dz} = \frac{-\imath\omega_1 d_{eff}}{n_1 c} A_3 A_2^* e^{-\imath\Delta kz} \quad (40)$$

$$\frac{dA_2}{dz} = \frac{-\imath\omega_2 d_{eff}}{n_2 c} A_3 A_1^* e^{-\imath\Delta kz} \quad (41)$$

$$\frac{dA_2}{dz} = \frac{-\imath\omega_3 d_{eff}}{n_3 c} A_1 A_2 e^{\imath\Delta kz} \quad (42)$$

where $\Delta k = k_3 - k_1 - k_2$

- In sum frequency generation (SFG), two lower frequency photons combine to make a higher frequency photon
- In difference frequency generation (DFG), a high frequency photon is split into two lower frequency photons
- SHG and DFG can be used to amplify weak signals or convert a signal to from one center wavelength to another

$\chi^{(3)}$ Processes

- The general form of the $\chi^{(3)}$ tensor is:

$$P_i^{(3)}(\omega) = \epsilon_0 \chi_{ijkl}^{(3)}(\omega : \omega_1, \omega_2, \omega_3) E_j(\omega_1) E_k(\omega_2) E_l(\omega_3) \tag{43}$$

- $\chi^{(2)}$ effects are only observed in materials without inversion symmetry.
- Can be shown that all materials have nonzero $\chi^{(3)}$ coefficients

Intensity Dependent Index of Refraction

- The $\chi^{(3)}$ coefficient gives rise to an intensity dependent index of refraction

$$\begin{aligned} P &= \chi^{(1)}E + \chi^{(3)}E^3 \\ &= \left(\chi^{(1)} + \chi^{(3)}|E|^2\right)E \end{aligned} \tag{44}$$

- This can also be written as:

$$n = n_0 + n_2 I \tag{45}$$

Kerr Lensing

- In Gaussian laser beams, the intensity is highest in the center of the beam. Hence the index of refraction is highest in the center.
- The field turns the medium into a lens. The effect is known as **self-focusing**
- Because the only frequency present in these interactions is ω, dispersion is irrelevant and this interaction is always phase matched!

Self Phase Modulation (SPM)

- Temporal result of Kerr effect is known as self phase modulation
- A pulse can be written as:

$$E_{in}(t) = E(t)e^{\imath(\omega_0 t+\phi(t))} \tag{46}$$

where $E(t)$ is real.

- If this pulse is passed through a non-dispersive Kerr medium, the output is

$$E_{out}(t) = E(t)e^{\imath\left(\omega_0 t+\phi(t)+\frac{\omega n_0 L}{c}+\frac{\omega n_2 I(t) L}{c}\right)} \tag{47}$$

where $I(t)$ is the instantaneous intensity of the pulse

- Last term in exponential spectrally broadens pulse
- Because multiple frequencies are involved, dispersion and phase matching are important

Frequency Tripling

- The $\chi^{(3)}(3\omega : \omega, \omega, \omega)$ coefficient is typically very small.
- Intensities required to excite a third-harmonic response may be near or in excess of the damage threshold of the material
- Direct tripling is inefficient and rarely performed
- To obtain the third harmonic of a laser, first frequency double the beam and mix the output with the fundmental

Four Wave Mixing (FWM)

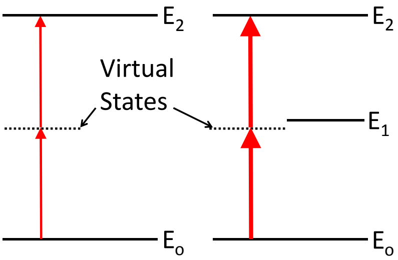
Common Nonlinear Crystals

	Crystal	Transparency
BBO	Beta Barium Borate	198-2600 nm
BiBO	Bismuth Triborate	286-2700 nm
KDP	Potassium Dihydrogen Phosphate	176-1550 nm
KTP	Potassium Titanyl Phosphate	352-4500 nm
LBO	Lithium Triborate	160-2300 nm
LN	Lithium Niobate	400-5500 nm

Non-Parametric Processes

In **non-parametric** nonlinear processes, energy can be transferred to the medium in which the light is propagating

Two Photon Absorption



- Two photons absorbed simultaneously to bring atom to high-lying energy level
- Absorption enhanced by enhanced by allowed transitions near single photon resonance
- Can limit the performance of high-powered lasers

Stimulated Raman Scattering (SRS)

- Interaction of light with an acoustic phonon in the material
-

Stimuated Brillioun Scattering (SBS)

- Interaction of light with an acoustic phonon in the material
- The scattered wave is always backwards propagating
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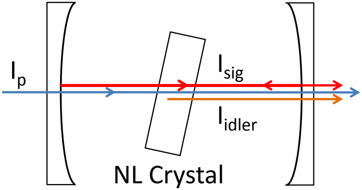
Frequency Conversion

Optical Parametric Oscillator (OPO)



- Place nonlinear crystal inside cavity
- When pumped with a high-intensity laser, crystal acts a gain medium
- Resulting device behaves very similar to a laser
- OPOs can angle or temperature tuned

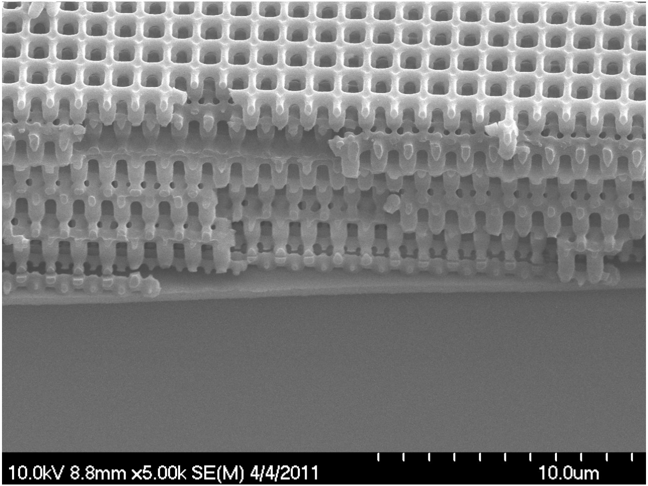
Continuum Generation

- Focus a femtosecond laser into a fiber or a sapphire plate
- Exact spectrum structure is chaotic
- Useful as a high intensity, broadband spectroscopic light source

Solitons

- Linear dispersion and self phase modulation balance each other
- Pulse propagates without changing shape
- Nonlinear propagation equation solutions have discrete energies and stable pulse profiles

Two Photon Photoresist



Two photon absorption permits high contrast 3D patterning
Courtesy Sidartha Gupta

Conclusions

- Nonlinear processes are those in which the output may contain frequencies of light not present in the input.
- Lasers are essential for observing nonlinear optical properties.
- Nonlinear optical interactions require a material medium (solid, liquid, gas, plasma)
- Photon energy must be conserved in **parametric** nonlinear processes
- Some photon energy is transferred to the medium in **non-parametric** nonlinear processes
- Momentum must be conserved in nonlinear processes (**phase-matching)** in order for them to be observable on a macroscopic scale