Nonlinear Optics.

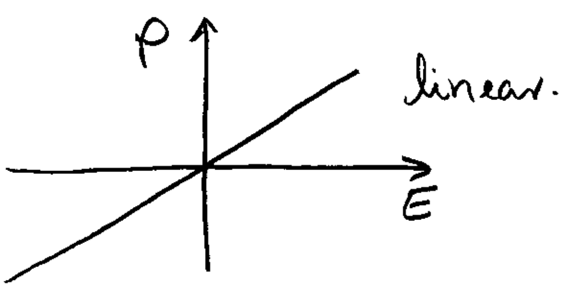
Light interacts with light via the medium.
light - modifies medium - modifies light.

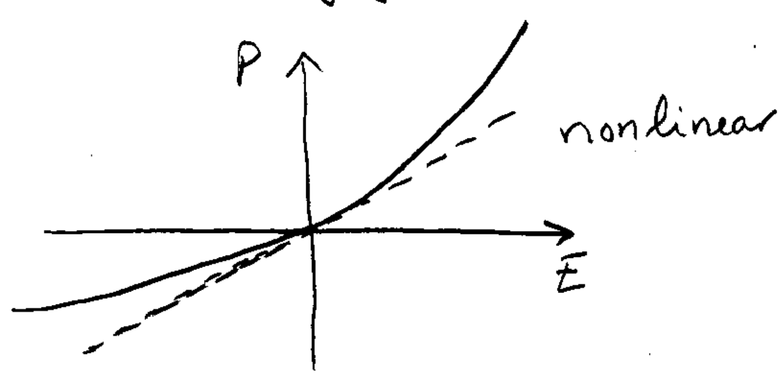
Light propagation in a media charactized by second-order (quadratic) or third order. P-E relation.

2nd order Nonlinear P & E. relation
- frequency doubling (second harmonic generat')
- wave mixing of two waves to generate a third
  $\omega_3 = \omega_1 + \omega_2$ or $\omega_3 = \omega_1 - \omega_2$.
- two waves amplify a third wave
  (parametric amplification).
  feedback $\Rightarrow$ parametric oscillation.

3rd order nonlinear P & E relation
- third-harmonic generation
- self-phase modulation
- self focusing.
- four-wave mixing.
- optical amplification.
- optical phase conjugation.

Nonlinear Optical Media.





In general we write $P = \epsilon_0 \chi E + 2dE^2 + 4\chi^{(3)}E^3 + \cdots$

or $P = \epsilon_0 (\chi E + \chi^{(2)}E^2 + \chi^{(3)}E^3)$ alternate.

d – coefficient describing the second order nonlinear effect
$\chi^{(3)}$ – " " " third " " "

$d = 10^{-24}$ to 10^{-21} (MKS units A-s/V²).
$\chi^{(3)} \approx 10^{-34}$ to 10^{-29} (MKS units).

Nonlinear Wave Equation

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

$$P = \epsilon_0 \chi E + P_{NL}$$

$$P_{NL} = 2d\,E^2 + 4\chi^{(3)}E^3 + \cdots \quad \text{(nonlinear polarizat).}$$

$n^2 = 1+\chi \quad c_0 = \frac{1}{\sqrt{\mu_0 \epsilon_0}} \quad c = \frac{c_0}{n}$

$$\Rightarrow \quad \nabla^2 E - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = -\mu_0 \frac{\partial^2 P_{NL}}{\partial t^2} = \underbrace{S}_{\substack{\text{source term.} \\ \text{(driving force).}}}$$

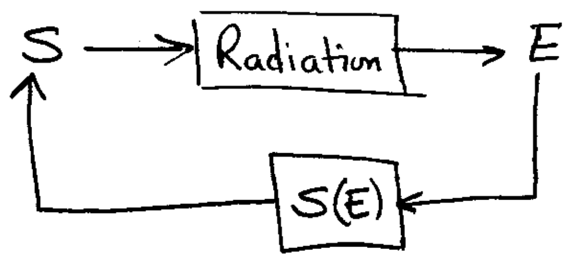
Notice, this is a wave equation in which $S = -\mu_0 \frac{\partial^2 P_{NL}}{\partial t^2}$ acts as a source radiating in a linear medium of refractive index n. P_{NL} (and S) is a nonlinear funct of E
$\Longrightarrow$ above equat is a nonlinear differential equat of E.

$$\boxed{\nabla^2 E - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = S}$$

Basic equation that underlies the theory of nonlinear optics.

Two approaches to solving this.
1. Born approximation
2. Coupled wave theory.

<u>Born Approximation</u>



E_0 (optical field) incident on nonlinear medium.

$\Rightarrow S(E_0) \rightarrow E_1$.

$S(E_1) \rightarrow E_2$ and so on.

First step of this is known as the First Born Approximation.
Second Born Approximat : second step.

First Born : light intensity sufficiently weak so nonlinearity is small.

$E_0 \rightarrow P_{NL} \rightarrow S(E_0) \rightarrow E_1$

If E_0 has one or several monochromatic waves of different frequencies.

$\Rightarrow P_{NL} \Rightarrow S(E_0)$ is nonlinear and new frequencies are created.

Second Order Nonlinear Optics

$$P_{NL} = 2dE^2$$

- Consider E comprising one or two harmonic components
  $\Rightarrow$ spectral components of P_{NL}

Second Harmonic Generation and Rectification

Response of nonlinear medium to harmonic electric field of angular frequency ω (wavelength $\lambda_0 = \frac{2\pi c_0}{\omega}$) and complex amplitude $E(\omega)$.

$$E(t) = \mathrm{Re}\{E(\omega)\exp(j\omega t)\}.$$

$$P_{NL} = 2dE^2 = 2d\,EE^* = \frac{2d}{4}\left(E(\omega)\exp(j\omega t) + E^*(\omega)\exp(-j\omega t)\right) * \left(E^*(\omega)\exp(-j\omega t) + E(\omega)\exp(j\omega t)\right).$$

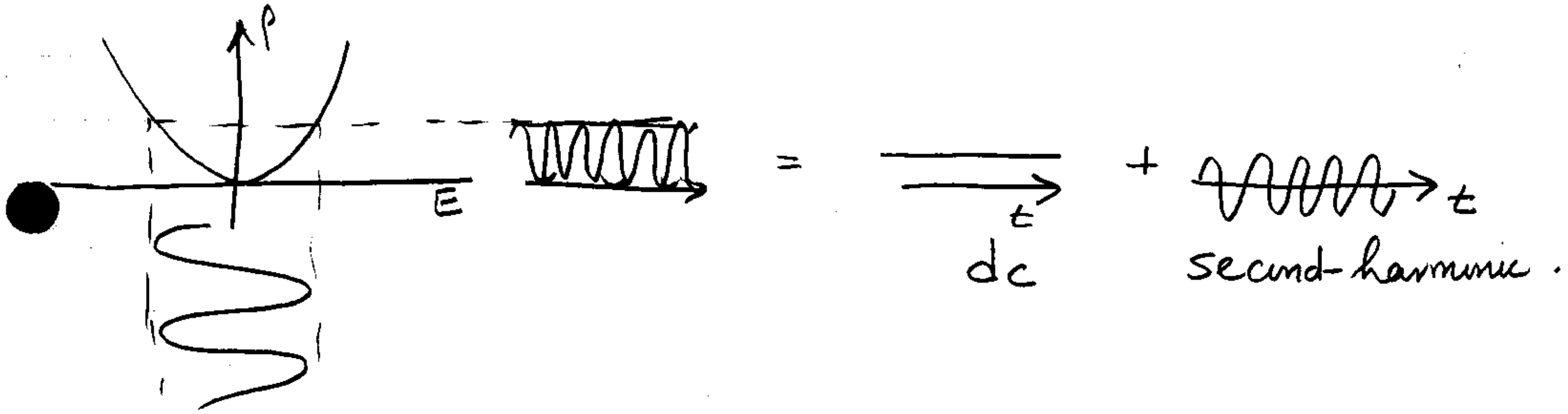
$$= \frac{2d}{4}\Big(E(\omega)E^*(\omega) + E(\omega)E^*(\omega) + E(\omega)E(\omega)\exp(j2\omega t) + E^*(\omega)E^*(\omega)\exp(-j2\omega t)\Big).$$

$$= d\,E(\omega)E^*(\omega) + \frac{d}{2}\Big(E(\omega)E(\omega)\exp(j2\omega t) + E^*(\omega)E^*(\omega)\exp(-j2\omega t)\Big)$$

$$= P_{NL}(0) + d\,\mathrm{Re}\{E(\omega)E(\omega)\exp(j2\omega t)\}.$$

$$= P_{NL}(0) + \mathrm{Re}\{P_{NL}(2\omega)\exp(j2\omega t)\}.$$

$$P_{NL}(0) = d\,E(\omega)E^*(\omega) \qquad P_{NL}(2\omega) = d\,E(\omega)E(\omega).$$



$$S(t) = -\mu_0 \frac{\partial^2 P_{NL}}{\partial t^2}.$$

$$S(2\omega) = 4\mu_0 \omega^2 d\, E(\omega)E(\omega). \longrightarrow$$ radiates field at 2ω. $\left(\frac{\lambda_0}{2}\right)$.

$$I(2\omega) \propto |S(2\omega)|^2 \propto \omega^4 d^2 I^2$$

$$I = \frac{|E(\omega)|^2}{2\eta}$$

Second Harmonic proportional to d^2, $\frac{1}{\lambda_0^4}$ and to I^2.

Consequently, 2^{nd} HG $\propto I = \frac{P}{A}$ P: incident Power, A: cross-sectional area.

$\Rightarrow$ want high intensity.

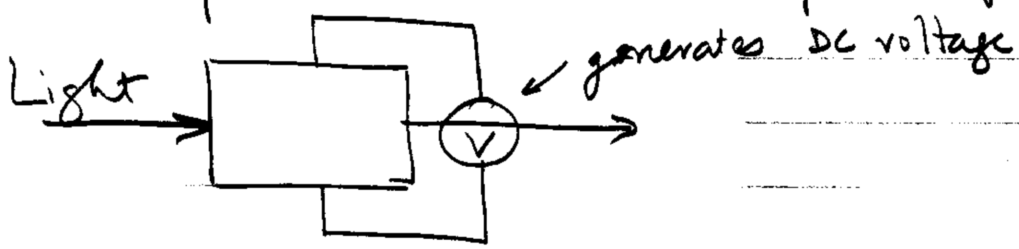
Enhance SHG. we want long interaction length.

Ruby Laser (694 nm (red).) $\xrightarrow{\omega}$ KDP Crystal $\xrightarrow{2\omega}$ 347 nm (UV).

Ti: Sapphire (700 – 1050 nm (IR)) $\xrightarrow{\omega}$ BBO $\xrightarrow{2\omega}$ 350–525 nm (visible).

Optical Rectification. $P_{NL}(0)$ corresponds to a steady (non-time varying) polarization density.

$\Rightarrow$ dc potential across the plates of a capacit.



MW peak power $\longrightarrow$ several hundred μV.

Electro-Optic Effect.

Suppose we have

$$E(t) = \underbrace{E(0)}_{\text{DC bias}} + Re\{\underbrace{E(\omega)\exp(j\omega t)}_{\text{optical field}}\}.$$

$$\Rightarrow P_{NL}(t) = P_{NL}(0) + Re\{P_{NL}(\omega)\exp(j\omega t)\} + Re\{P_{NL}(2\omega)\exp(j2\omega t)\}.$$

$$P_{NL}(0) = d[2E^2(0) + |E(\omega)|^2]. \quad \text{— 0 frequency.}$$

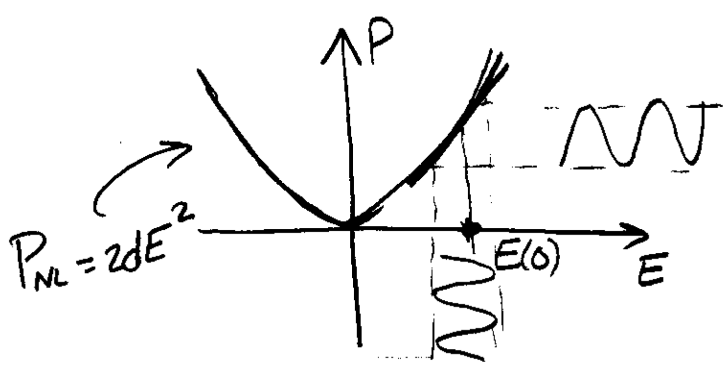
$$P_{NL}(\omega) = 4dE(0)E(\omega) \quad \text{— } \omega$$

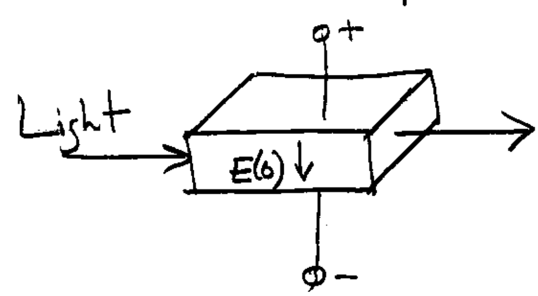
$$P_{NL}(2\omega) = dE(\omega)E(\omega) \quad \text{— } 2\omega.$$

If the optical field is much smaller in magnitude than the electric field.

$$|E(\omega)|^2 << |E(0)|^2$$

$\Rightarrow$ $P_{NL}(2\omega)$ is small compared to $P_{NL}(\omega)$ & $P_{NL}(0)$.





We can write $P_{NL}(\omega) = \epsilon_0 \Delta\chi E(\omega)$. $\Delta\chi = \left(\frac{4d}{\epsilon_0}\right)E(0)$.

$\Delta\chi$ - increase in susceptibility proportional to electric field $E(0)$.

$$n^2 = 1+\chi \Rightarrow 2n\Delta n = \Delta\chi$$

$$\text{or} \quad \Delta n = \frac{2d}{n\epsilon_0} E(0).$$

Medium is linear with refractive index $n+\Delta n$ that is controlled by $E(0)$.

$\Rightarrow$ $E(0)$ & $E(\omega)$ are coupled, one field controls the other, the medium exhibits the linear electro-optic effect (Pockels Effect).

$$\Delta n = -\frac{1}{2} n^3 r E(0)$$

$$\Rightarrow \boxed{r \simeq -\frac{4}{\epsilon_0 n^4} d}$$

not quite true because medium is dispersive response at $E(0)$ is different from $E(\omega)$.

Three Wave Mixing.

Frequency Conversion.

$$\mathcal{E}(t) = \mathrm{Re}\left\{E(\omega_1)\exp(j\omega_1 t) + E(\omega_2)\exp(j\omega_2 t)\right\}.$$

$$P_{NL} = 2d\,\mathcal{E}^2$$

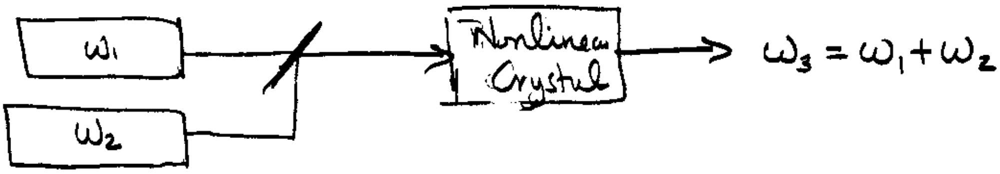
Frequencies at 0, $2\omega_1$, $2\omega_2$, $\omega_+ = \omega_1+\omega_2$, and $\omega_- = \omega_1-\omega_2$.

$$P_{NL}(0) = d\left[|E(\omega_1)|^2 + |E(\omega_2)|^2\right]. \quad P_{NL}(2\omega_1) = d\,E(\omega_1)E(\omega_1)$$

$$P_{NL}(2\omega_2) = d\,E(\omega_2)E(\omega_2). \quad P_{NL}(\omega_+) = 2d\,E(\omega_1)E(\omega_2)$$

$$P_{NL}(\omega_-) = 2d\,E(\omega_1)E^*(\omega_2)$$

From previous page, it is obvious that a second-order nonlinear medium can be used to mix two optical waves of different frequencies to generate a third wave at the difference or sum frequencies.



Notice: Although the incident waves at ω_1 & ω_2 produce polarization densities at 0, $2\omega_1$, $2\omega_2$, $\omega_1+\omega_2$ and $\omega_1 - \omega_2$ all waves not necessarily generated.

Phase Matching. Wave 1 — $\vec{k}_1$

Wave 2 → $\vec{k}_2$

$$E(\omega_1) = A_1 \exp(-j\vec{k}_1\cdot\vec{r}) \qquad E(\omega_2) = A_2 \exp(-j\vec{k}_2\cdot\vec{r})$$

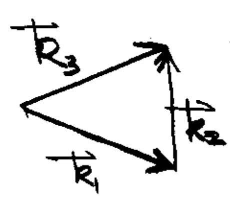
$$\Rightarrow P_{NL}(\omega_3) = P_{NL}(\omega_2+\omega_1) = 2d\, E(\omega_1)E(\omega_2) = 2d A_1 A_2 \exp(-j\vec{k}_3\cdot\vec{r}).$$

$$\boxed{\omega_3 = \omega_1 + \omega_2.} \qquad \text{frequency-matching condition}$$

and $$\boxed{\vec{k}_3 = \vec{k}_1 + \vec{k}_2} \qquad \text{Phase matching condition.}$$

Medium: light source of frequency $\omega_3 = \omega_1 + \omega_2$, $\vec{k}_3 = \vec{k}_1 + \vec{k}_2$.

$\Rightarrow$ radiates wave in $\vec{k}_3$ direction.



Notice argument of E_3 is $\omega_3 t - \vec{k}_3\cdot\vec{r}$. We need to ensure frequency matching AND phase matching for this wave.

Temporal and Spatial phase matching.

$\Rightarrow$ interaction over time and space

<u>Example</u> suppose $\cdot k_1 = \frac{n\omega_1}{c_0}$ $\cdot k_2 = \frac{n\omega_2}{c_0}$

Then $k_3 = \frac{n\omega_1}{c_0} + \frac{n\omega_2}{c_0} = \frac{n\omega_3}{c_0} \Rightarrow \omega_3 = \omega_1 + \omega_2.$

That is $\cdot$ frequency matching insures phase matching!

However, all materials are dispersive $\Rightarrow$ n_1, n_2, n_3.

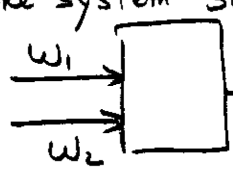
$$\frac{n_3\omega_3}{c_0} = \frac{n_1\omega_1}{c_0} + \frac{n_2\omega_2}{c_0} \Rightarrow \boxed{n_3\omega_3 = n_1\omega_1 + n_2\omega_2}.$$

$\hookrightarrow$ phase matching.

$\boxed{\omega_3 = \omega_1 + \omega_2}$ - frequency matching.

Three wave Mixing.

Make system so that only ω_3 is phase & frequency matched



$\omega_3 = \omega_1 + \omega_2$.

Once ω_3 is generated $\rightarrow$ $\omega_2 = \omega_3 - \omega_1$ is generated.
AND is also phase-matched.

$\Rightarrow \omega_3 - \omega_2 = \omega_1$

mix to form ω_1

This complicated process is called <u>Three Wave Mixing</u>.

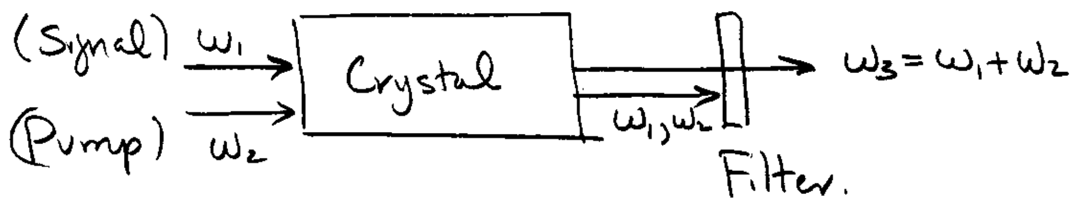
Two-wave Mixing — not possible in general.

ω_1 & ω_2 cannot be coupled without the help of a third wave.

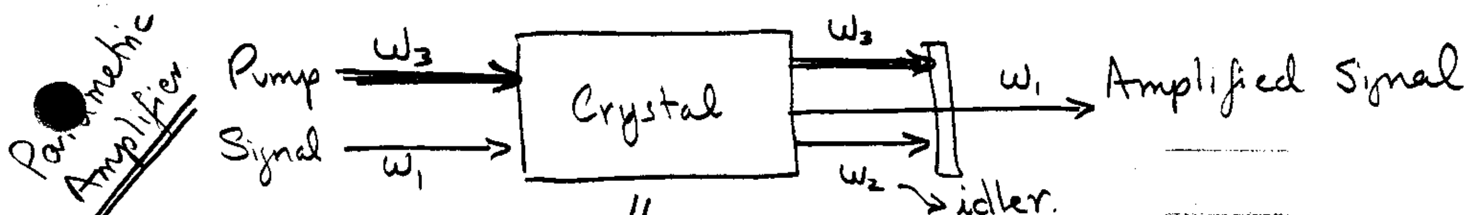
Only case: degenerate case $\omega_2 = 2\omega_1$

$\rightarrow \frac{\omega_2}{2} = \omega_2 - \omega_1$ contributes to ω_1.

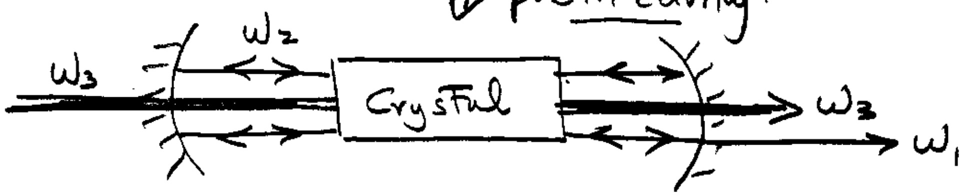
Three wave mixing is a parametric interaction.



Up-converted signal.



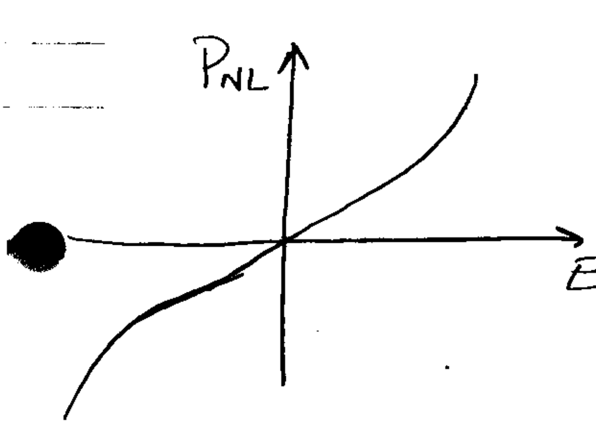
$\Downarrow$ put in cavity.



Parametric Oscillator. (Parametric Amplifier w/ feedback).

Down-converter: $\omega_2 = \omega_3 - \omega_1$.

Third-Harmonic Generation and Self-Phase Modulation.



$$E(t) = \mathrm{Re}\{E(\omega)\exp(j\omega t)\}.$$

$$P_{NL}(\omega) = 3\chi^{(3)}|E(\omega)|^2 E(\omega).$$

$$\underbrace{P_{NL}(3\omega) = \chi^{(3)} E^{(3)}(\omega)}_{\text{very low conversion efficiency}}.$$

Optical Kerr Effect

$$P_{NL}(\omega) = 3\chi^{(3)} |E(\omega)|^2 E(\omega).$$

$$\epsilon_0 \Delta\chi = \frac{P_{NL}(\omega)}{E(\omega)} = 3\chi^{(3)} |E(\omega)|^2 = 6\chi^{(3)} \eta I.$$

$$I = \frac{|E(\omega)|^2}{2\eta}.$$

Recall:

$$P(\omega) = \epsilon_0 \chi E(\omega) + \epsilon_0 \Delta\chi E(\omega).$$

$$\Rightarrow P(\omega) = \epsilon_0 (\chi + \Delta\chi) E(\omega).$$

$$D(\omega) = \epsilon_0 E(\omega) + P(\omega)$$

$$D(\omega) = \epsilon_0 \underbrace{(1+\chi+\Delta\chi)}_{\epsilon_r} E(\omega).$$

$$D(\omega) = \epsilon_0 \epsilon_r E(\omega).$$

index of refract $\rightarrow n = \sqrt{\epsilon_r}$.

$$n^2 = 1+\chi$$

$$\Rightarrow \Delta n = \frac{\partial n}{\partial \chi} \Delta\chi.$$

$$= \frac{\Delta\chi}{2n}.$$

or $$\boxed{\Delta n = \frac{3\eta}{\epsilon_0 n} \chi^{(3)} I = n_2 I}$$

$n(I) = n + n_2 I \Rightarrow$ Refractive index is a linear function of the optical intensity!

$$\boxed{n_2 = \frac{3\eta_0}{n^2 \epsilon_0} \chi^{(3)}.}$$

$n(I) = n + n_2 I$ – Optical Kerr Effect. (similar to electro-optic Kerr Effect, $\Delta n \propto E^2$).

$\therefore$ Optical Kerr Effect is a self-induced effect – phase velocity of the wave depends on the wave's own intensity!

n_2 (cm^2/W) $10^{-16} \rightarrow 10^{-14}$ in glasses.

10^{-14} to 10^{-7} in doped glasses.

10^{-10} to 10^{-8} in organic materials.

$n_2 \cdot 10^{-10}$ to 10^{-2} in semiconductors.

* Sensitive to wavelength and polarization.

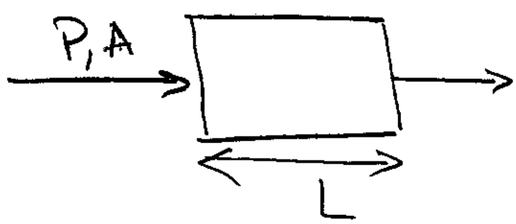
Sometimes we write:

$$n(I) = n + \frac{n_2 |E|^2}{2} \Rightarrow n_2 \text{ is different by a factor of } \eta.$$

Self-Phase modulation.

Optical Kerr Effect in Third Order Medium.

Optical beam of Power P, cross-sectional area A $\left(I = \frac{P}{A}\right)$.

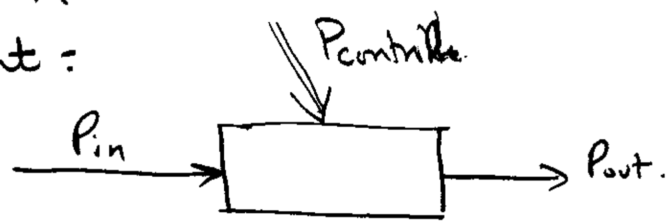


$$\phi = \frac{2\pi n(I) L}{\lambda_0}$$

$$= 2\pi\left(n + n_2 \frac{P}{A}\right)\frac{L}{\lambda_0}.$$

$$\Rightarrow \Delta\phi = \frac{2\pi n_2 L}{\lambda_0 A} P.$$

Useful for light controlling light:

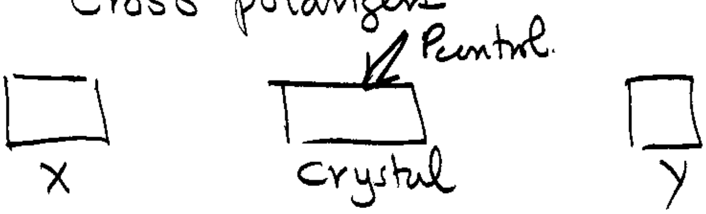


To maximize this effect, we want L large and A small.

$$\Delta\phi = \pi \Rightarrow P_\pi = \frac{\lambda_0 A}{2 L n_2} \quad \text{(half-wave power)}.$$

Phase modulation → intensity modulation.

Cross polarizers



n_2 - depends on polarization

All optical Modulator.