

Nonlinear Optics (Yariv ch 8)

Recall classical electron oscillator model of atom

$$j_{nm} \leftarrow \text{nmf},$$

$$\mapsto x$$

linear spring restoring force



Response to applied field $E(t) = \text{Re}[\mathcal{E}e^{i\omega t}]$:



$$\text{polarization } P(\omega) = -NeX(\omega) = \epsilon_0 K(\omega) \mathcal{E} \quad \text{linear}$$

real atoms in crystals:

a) force not necessarily linear

b) restoring force not necessarily symmetric

$$F = -(m\omega_0^2 X + mDX^2 + \dots)$$

1st nonlinear term

modified eqn of motion:

$$\frac{d^2X}{dt^2} + \omega_0^2 X + DX^2 = -\frac{e\mathcal{E}}{2m}(e^{i\omega t} + e^{-i\omega t})$$

equation is no longer [↑] linear in X . Cannot use complex form and take real part at the end!

nonlinear term $\propto x^2$ gives rise to a component
in x oscillating at 2ω

$$x(t) = \frac{1}{2} (q_1 e^{i\omega t} + q_2 e^{2i\omega t} + cc)$$

plug into eqn of motion \Rightarrow see Tariq for
gory details

recover previous result

$$p^{(\omega)}(t) = \frac{\epsilon_0}{2} [\chi(\omega) \Sigma e^{i\omega t} + cc]$$

linear polarization

additional nonlinear polarization.

$$p^{(2\omega)}(t) = \frac{1}{2} [P^{(2\omega)} e^{2i\omega t} + cc]$$

$$= -\frac{Ne}{2} (q_2 e^{2i\omega t} + cc)$$

$$= \frac{1}{2} [d^{(2\omega)} \Sigma^2 e^{2i\omega t} + cc]$$

in crystals, d is a "3rd rank tensor"

$$P_i^{(2\omega)} = \sum_{j,k=x,y,z} d_{ijk}^{(2\omega)} \Sigma_i \Sigma_j$$

for most real crystals, most elements of d -tensor
are zero.

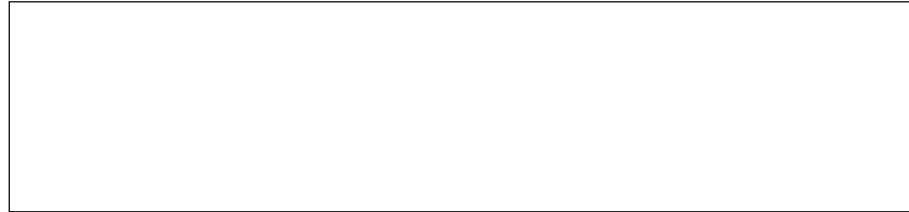
frequency mixing - by extension

$$\text{if } E = [\epsilon(\omega_1)e^{i\omega_1 t} + \epsilon(\omega_2)e^{i\omega_2 t} + cc]$$

polarization develops at not only $2\omega_1$, $2\omega_2$, but also sum + difference frequencies

$$P^{(\omega_1 + \omega_2)} = \sum_{jk} \left[d_{ijk}^{(\omega = \omega_1 + \omega_2)} E_j^{(\omega_1)} E_k^{(\omega_2)} e^{i(\omega_1 + \omega_2)t} + cc \right]$$

$$P^{(\omega_1 - \omega_2)} = \sum_{jk} \left[d_{ijk}^{(\omega = \omega_1 - \omega_2)} E_j^{(\omega_1)} (E_k^{(\omega_2)})^* e^{i(\omega_1 - \omega_2)t} + cc \right]$$

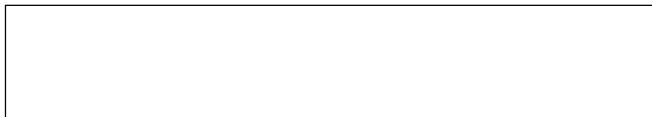


Consequences of nonlinear polarization for wave propagation in crystals

Recall wave equation:

$$\nabla^2 \vec{E} = \mu_0 \sigma \frac{\partial \vec{E}}{\partial t} + \mu_0 \epsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}$$

separate \vec{P} into linear, nonlinear parts



$$\text{then } \vec{E} = \epsilon_0 (1 + \chi^{(1)})$$

$$\nabla^2 \vec{E} = \mu_0 \sigma \frac{\partial \vec{E}}{\partial t} + \mu_0 \epsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \vec{P}_{NL}}{\partial t^2} \quad ①$$

Nonlinear 3-wave mixing!

Consider general case of coupling 3 waves together by nonlinear interaction

$$E^{(\omega_1)}(z, t) = \frac{1}{2} [E_1(z) e^{i(\omega_1 t - k_1 z)} + \text{cc}]$$

assume plane waves

$$\begin{aligned} E^{(\omega_2)}(z, t) &\text{ similar} \\ E^{(\omega_3)}(z, t) & \end{aligned}$$

$$E = E^{(\omega_1)}(z, t) + E^{(\omega_2)}(z, t) + E^{(\omega_3)}(z, t)$$

Subst into ① and separate into 3 eqns for each freq.
 P_{NL} will develop terms oscillating at all pairwise sum + diff frequencies

i.e.

$$\left[d^{(\omega_1+\omega_2)} E_1 E_2 e^{i[(\omega_1+\omega_2)t - (k_1+k_2)z]} + \text{cc} \right]$$

$$\text{or } \left[d^{(\omega_3-\omega_2)} E_3 E_2^* e^{i[(\omega_3-\omega_2)t - (k_3-k_2)z]} + \text{cc} \right]$$

if $\boxed{\quad}$, then $P_{NL}^{(\omega_1+\omega_2)}$ will act as a source for ω_3 wave in ω_3 eqn

look at ω_1 eqn

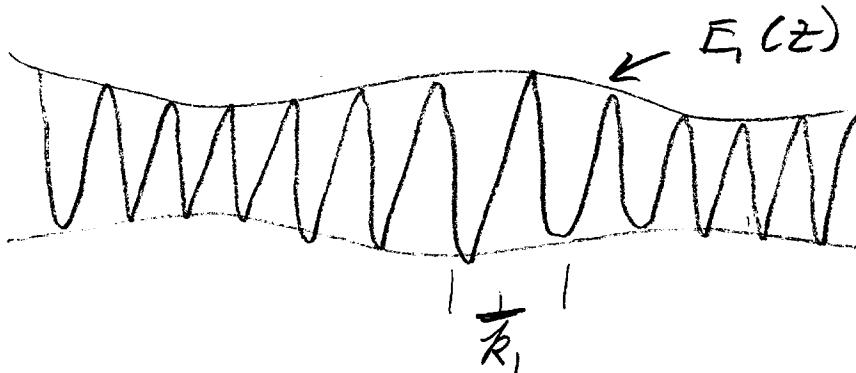
$$\textcircled{2} \quad \nabla^2 E^{(\omega_1)} = \mu_0 \nabla_1 \frac{\partial E^{(\omega_1)}}{\partial t} + \mu_0 E_1 \frac{\partial^2 E^{(\omega_1)}}{\partial t^2} + \mu_0 d \frac{\partial^2}{\partial t^2} \left[\frac{E_3(z) E_2^*(z)}{2} e^{i[(\omega_3-\omega_1)t - (k_3-k_1)z]} + \text{cc} \right]$$

Since $E^{(\omega)}$ is a plane wave

$$\textcircled{3} \quad \nabla^2 E^{(\omega_1)} = \frac{1}{2} \frac{\partial^2}{\partial z^2} [E_1(z) e^{i(\omega_1 t - k_1 z)} + \text{cc}] \approx -\frac{1}{2} [k_1^2 E_1(z) + 2ik_1 \frac{\partial E_1(z)}{\partial z} + \frac{\partial^2 E_1(z)}{\partial z^2}] e^{i(\omega_1 t - k_1 z)} + \text{cc}$$

slowly varying envelope approx:

$$\left| \frac{d^2 E_1(z)}{dz^2} \right| \ll \left| k_1 \frac{d E_1(z)}{dz} \right|$$



Combine ②, ③.

$$\begin{aligned} & -\frac{1}{2} \left[k_1^2 E_1(z) + 2ik_1 \frac{d E_1(z)}{dz} \right] e^{i(\omega_1 t - k_1 z)} + cc \\ &= [i\omega_1 \mu_0 - \omega_1^2 \mu_0 \epsilon_1] \left[\frac{E_1(z)}{z} e^{i(\omega_1 t - k_1 z)} + cc \right] \\ & \quad - \left[\frac{\omega_1^2 \mu_0 d}{2} E_3^*(z) E_2(z) e^{i[\omega_1 t - (k_3 - k_2)z]} + cc \right] \\ & \text{use } \boxed{\quad}, \text{ mult by } i/k_1, e^{-i(\omega_1 t - k_1 z)} \end{aligned}$$

$$\frac{d E_1(z)}{dz} = -\frac{\sigma_1}{2} \sqrt{\frac{\mu_0}{\epsilon_1}} E_1 - \frac{i\omega_1}{2} \sqrt{\frac{\mu_0}{\epsilon_1}} d E_3^*(z) E_2(z) e^{-i(k_3 - k_2 + k_1)z}$$

Similarly

$$\frac{d E_2^*(z)}{dz} = -\frac{\sigma_2}{2} \sqrt{\frac{\mu_0}{\epsilon_2}} E_2^* + \frac{i\omega_2}{2} \sqrt{\frac{\mu_0}{\epsilon_2}} d E_1 E_3^*(z) e^{-i(k_1 - k_3 + k_2)z}$$

$$\frac{d E_3(z)}{dz} = -\frac{\sigma_3}{2} \sqrt{\frac{\mu_0}{\epsilon_3}} E_3 + \frac{i\omega_3}{2} \sqrt{\frac{\mu_0}{\epsilon_3}} d E_1 E_2 e^{-i(k_1 + k_2 - k_3)z}$$

nonlinear coupled wave equations

Second harmonic generation $\omega_1 = \omega_2 = \omega; \omega_3 = 2\omega$

take $\sigma_1 = \sigma_2 = \sigma_3 = 0$

$$\frac{dE^{2\omega}}{dz} = -i\omega \sqrt{\frac{\mu_0}{\epsilon_2}} d[E^{(\omega)}(z)]^2 e^{i4kz} \quad \text{OK}$$



no pump depletion approx: conversion from $E^{(\omega)} \rightarrow E^{(2\omega)}$ is small

$$E^{(\omega)} \text{ approx const: } \cancel{\frac{dE^{(\omega)}}{dz}}$$

can simply integrate ④. Assume $E^{2\omega}(z=0) = 0$
crystal length L

$$E^{2\omega}(z=L) = -i\omega \sqrt{\frac{\mu_0}{\epsilon_2}} d[E^{(\omega)}]^2 \left(\frac{e^{i4kL} - 1}{i4k} \right)$$

$$\text{Output intensity } I(2\omega) = \frac{1}{2} \int \frac{\epsilon_2}{\mu_0} |E^{(2\omega)}|^2$$

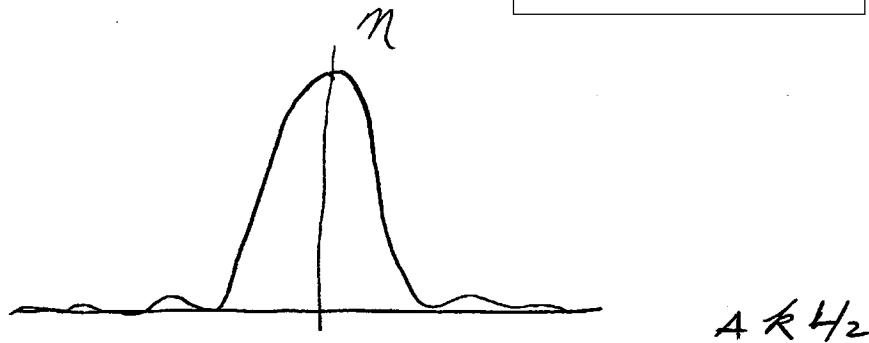
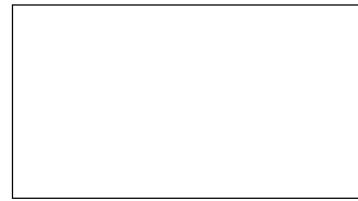
$$I(2\omega) = \frac{1}{2} \int \frac{\mu_0}{\epsilon_2} \omega^2 d^2 |E^{(\omega)}|^4 L^2 \frac{\sin^2(4kL/2)}{(4kL/2)^2}$$

conversion efficiency:

$$\eta_{\text{SHG}} = \frac{I(2\omega)}{I(\omega)} = 2 \sqrt{\frac{\mu_0}{\epsilon_2}} \frac{\mu_0}{\epsilon_1} \omega^2 d^2 L^2 \frac{\sin^2(4kL/2)}{(4kL/2)^2} I(\omega)$$

Phase matching

conversion efficiency α



$$\text{want } \Delta k = k^{(2\omega)} - 2k^{(\omega)} \approx 0$$

$$= \frac{2\omega}{c} n^{(2\omega)} - 2 \frac{\omega}{c} n^{(\omega)}$$



define coherence length $l_c \equiv \frac{2\pi}{4k}$

n is near peak when $l_c \gg L$

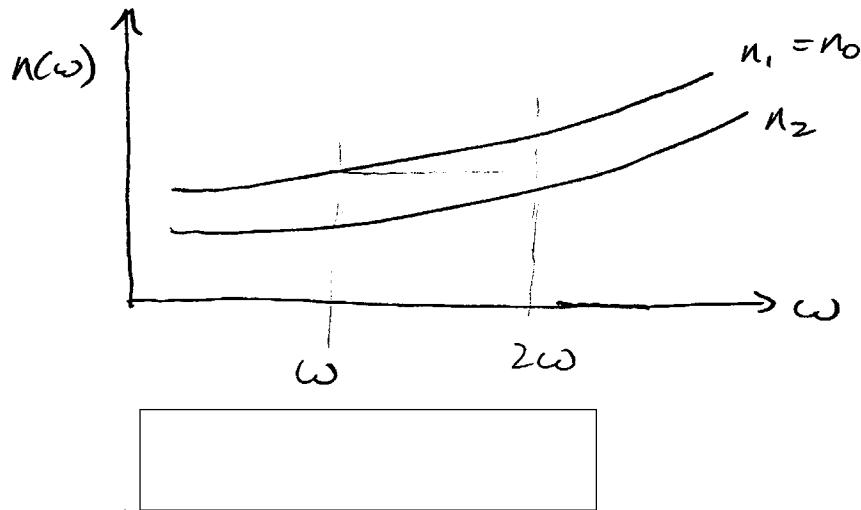
l_c - length over which $\omega, 2\omega$ waves remain in phase

How to achieve phase matching?

"Normal" dispersion



Solution: birefringent crystal



$$\text{recall} \quad n_e(2\omega) \leq n_e(\omega) \leq n_i(2\omega)$$

$$\frac{1}{n_e^2(2\omega)} = \frac{\cos^2 \Theta}{n_i^2(2\omega)} + \frac{\sin^2 \Theta}{n_2^2(2\omega)} = \frac{1}{n_0^2(\omega)}$$

Solve for Θ_m phase matching \neq

$$\sin^2 \Theta_m = \frac{n_0(\omega)^{-2} - n_0(2\omega)^{-2}}{n_2(2\omega)^{-2} - n_0(2\omega)^{-2}}$$

Send input wave, freq ω , into nonlinear, birefringent crystal at $\neq \Theta$ to optic axis, polarized as ordinary wave

\Rightarrow second harmonic comes out polarized as extraordinary wave!