Parametric processes – pure nonlinear optics!

We are already familiar with nonlinear effects

1. Saturation in a two-level system

$$\chi = -\frac{i\wp_{ab}}{\varepsilon_0 \hbar}\frac{\Delta\rho_0}{(\gamma - i\Delta)}\frac{1}{1+\underline{\underline{I/I_s}}\,L(\Delta)}$$

χ depends on the laser intensity
=> changes in the refractive index and absorption coefficient are ~~not~~ also intensity-dependent, and propagation is non-linear (doubled initial intensity $\neq$ doubled final intensity)
In saturation case the non-linearity is due to "self" action of the laser field

2. EIT / Raman resonanes

$$\chi_p = \frac{i\wp_{13}^2}{\varepsilon_0 \hbar}\frac{\Gamma_{12}}{\Gamma_{12}\Gamma_{13}+|\Omega_2|^2}$$

χ_p does not depend on the probe intensity (so it is linear in probe field), but is affected by the intensity of the strong control field, so it is a nonlinear interaction.

Atoms are "easy", so we can calculate χ from fundamental principles. This is impossible for more complex systems (crystals)

$\vec{P} = \chi^{(1)}\vec{E}$ linear effect

$\vec{P} = \chi^{(1)}\vec{E} + \chi^{(2)}\vec{E}^2 + \chi^{(3)}\vec{E}^3 + \dots$

in general, $\chi^{(n)}$ can be a tensor (in anisotropic crystals) but it is not common.

Most common (strong) nonlinearities : $\chi^{(2)}$ second order susceptibility
$\chi^{(3)}$ third order susceptibility

For most nonlinear crystals we tend to work far from any material's absorption resonances (to avoid absorption), so the nonlinear terms are weak

Crude example: saturation in a two-level scheme

$$\chi = \chi_0 \frac{1}{1 + I/I_s\, L(\Delta)} \approx \chi_0 \Big(\underset{\chi^{(1)}}{\uparrow 1} - \underset{\chi^{(3)}}{\uparrow \frac{I}{I_s} L(\Delta)} + \underset{\chi^{(5)}}{\uparrow \frac{1}{2}\left(\frac{I}{I_s}\right)^2 L^2(\Delta)} - \dots \Big)$$

$$\chi^{(1)} \gg \chi^{(3)} \gg \chi^{(5)}$$

Second-order nonlinearity

$$P^{(2)} = \chi^{(2)} E^2 \qquad E = \frac{1}{2} E_0 e^{-i\omega t} + \frac{1}{2} E_0^* e^{i\omega t}$$

$$P^{(2)} = \frac{1}{4}\chi^{(2)} \left\{ E_0^2 e^{-2i\omega t} + E_0^{*2} e^{2i\omega t} + 2|E_0|^2 \right\}$$

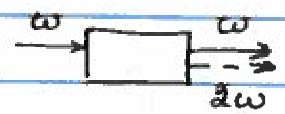
Two contributions : 1. zero frequency (DC term) → optical rectification, creates DC electric field

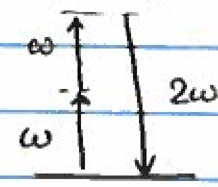
2. Second harmonics (2ω) → results in generation of an optical field on the doubled frequency

SHG – second harmonics generation

If we write the Maxwell wave equation for the frequency 2ω, we'll get

$$\nabla^2 E_{2\omega} - \frac{n_{2\omega}^2}{c^2}\frac{\partial^2 E_{2\omega}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P^{(2)}}{\partial t^2}$$



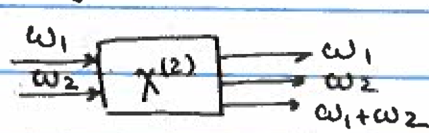


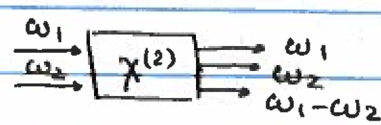
Sum and difference frequency generation

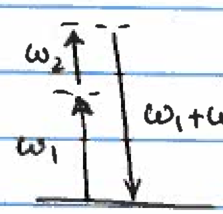
$$E = \tfrac{1}{2}E_1 e^{-i\omega_1 t} + \tfrac{1}{2}E_2 e^{-i\omega_2 t} + c.c. \qquad (\omega_1 > \omega_2)$$

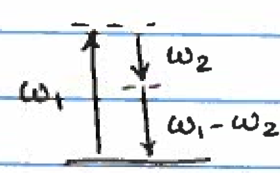
$$P^{(2)} = \chi^{(2)}E^2 = \frac{1}{4}\chi^{(2)}\underbrace{\left(|E_1|^2 + |E_2|^2\right)}_{\text{optical rectification}} + \frac{1}{4}\chi^{(2)}\underbrace{\left(E_1^2 e^{-2i\omega t} + E_2^2 e^{-2i\omega t} + c.c.\right)}_{\text{SHG}}$$

$$+ \frac{\chi^{(2)}}{4}\Big[\underbrace{E_1E_2\, e^{-i(\omega_1+\omega_2)t}}_{\text{sum-frequency generation}} + \underbrace{E_1E_2^{*}\, e^{-i(\omega_1-\omega_2)t}}_{\text{frequency-difference generation}} + c.c.\Big]$$

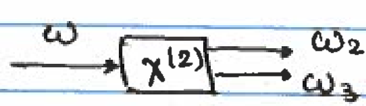


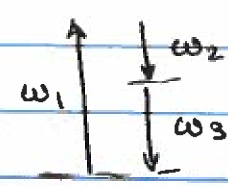






Parametric down conversion: pair of field is generated





Very important process for quantum information applications!

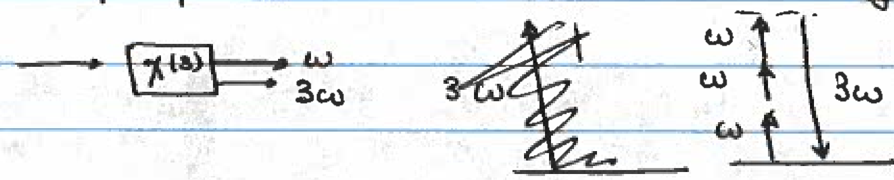
Second-order nonlinearity is the strongest, but less common — requires breaking the symmetry $\vec{E} \to -\vec{E}$ $\vec{P}^{(2)} \to \vec{P}^{(2)}$

Can occure only in anisotropic crystals (favourite $\chi^{(2)}$ materials — lithium niobate)

Third-order nonlinearity is much more common but weaker

$$\vec{P}^{(3)} = \chi^{(3)} \vec{E}^{3}$$

One pump field — third-harmonics generation

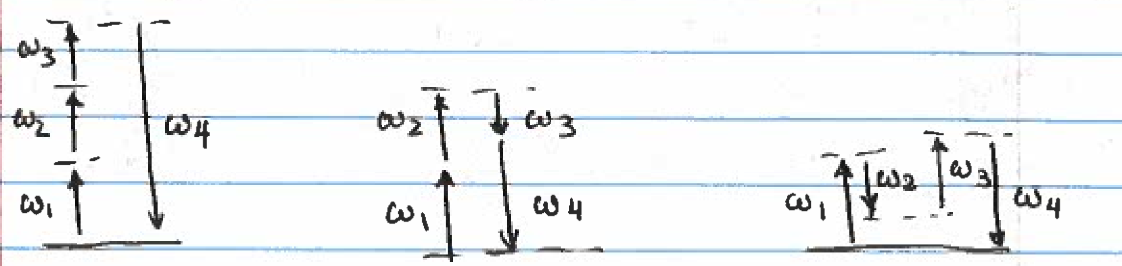


Four-wave mixing: three waves in, four waves out

$$\vec{E} = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_3 e^{-i\omega_3 t} + \text{c.c.}$$

If all three frequencies are different, the resulting polarization has 22 different frequency contributions

Examples



$\omega_4 = \omega_1 + \omega_2 + \omega_3$ $\omega_4 = \omega_1 + \omega_2 - \omega_3$ $\omega_4 = \omega_1 - \omega_2 + \omega_3$

Can we realize all possible frequencies?

What determines what process dominates?

Propagation equations for the sum-freq. generation in a $\chi^{(2)}$ medium (example)

$$E_{1,2} = \frac{1}{2} \mathcal{E}_{1,2}\, e^{ik_{1,2}z - i\omega_{1,2}t} + c.c.$$

newly generated field $\qquad \omega_1 + \omega_2 = \omega_3$

$$E_3 = \frac{1}{2} \mathcal{E}_3\, e^{ik_3 z - i\omega_3 t} + c.c$$

$$P^{(3)}_{sum} = \chi^{(2)} E_1 E_2 = \frac{1}{4} \chi^{(2)} \mathcal{E}_1 \mathcal{E}_2\, e^{i(k_1+k_2)z}\, e^{-i(\omega_1+\omega_2)t}$$

$$P_{\omega_3} = \mathcal{P}_3\, e^{ik_3 z - i\omega_3 t}$$

$$\mathcal{P}^{(3)} = \frac{1}{4} \chi^{(2)} \mathcal{E}_1 \mathcal{E}_2\, e^{i(k_1+k_2-k_3)z}$$

slowly varying amplitude

Propagation equation for the slowly-varying amplitudes

$$\frac{\partial \mathcal{E}_3}{\partial z} + \cancel{\frac{n_3}{c} \frac{\partial \mathcal{E}_3}{\partial t}}^{\,0} = \frac{ik_3}{2\varepsilon_0} \mathcal{P}^{(3)} \qquad \text{steady-state case}$$

$$\frac{d\mathcal{E}_3}{dz} = \frac{ik_3}{2\varepsilon_0} \cdot \frac{1}{4} \chi^{(2)} \mathcal{E}_1 \mathcal{E}_2\, e^{i\Delta k z} \qquad \Delta k = k_1 + k_2 - k_3$$

momentum mismatch

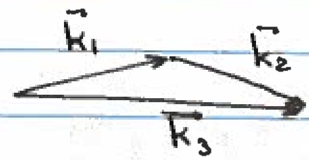
If $\Delta k = 0$ — phase matching conditions: $\frac{d\mathcal{E}_3}{dz} = \text{const}$

$k_{1z} + k_{2z} - k_{3z} = 0$ momentum conservation

$$\frac{n(\omega_1)\,\omega_1}{c} \cos\theta_1 + \frac{n(\omega_2)\,\omega_2}{c} \cos\theta_2 = \frac{n(\omega_3)\,\omega_3}{c} \cos\theta_3$$

If we can control / adjust $n(\omega_i)$, we may be able to achieve phase-matching in collinear geometry. Very convenient and desirable → hard to find such conditions and not always possible!

Typically, we have to find proper relative orientation



How bad it is if $\Delta k \neq 0$?

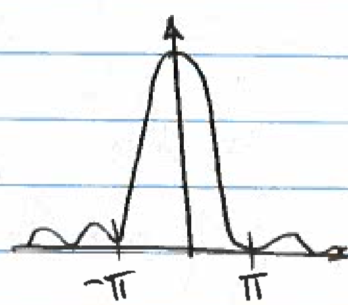
$$\frac{d\mathcal{E}_3}{dz} = \frac{ik_3}{8\varepsilon_0}\mathcal{E}_1\mathcal{E}_2\, e^{i\Delta k z}$$

If $\mathcal{E}_1$ and $\mathcal{E}_2$ do not change much at the distance L (undepleted pump approximation)

$$\mathcal{E}_3 \approx \frac{ik_3}{8\varepsilon_0}\mathcal{E}_1\mathcal{E}_2\chi^{(2)}\int_0^L e^{i\Delta k z}\,dz = \frac{k_3}{8\varepsilon_0}\mathcal{E}_1\mathcal{E}_2\chi^{(2)}\frac{e^{-i\Delta kL}-1}{\Delta k}$$

$$I_3 \propto |\mathcal{E}_3|^2$$

$$I_3 \propto I_1 I_2 (\chi^{(2)})^2 L^2 \underbrace{\frac{\sin^2 \Delta kL/2}{(\Delta kL/2)^2}} \quad \text{sinc}\,(\Delta kL/2)$$



Efficient energy conversion only for $\Delta kL \lesssim 2\pi$ (small Δk or small L)

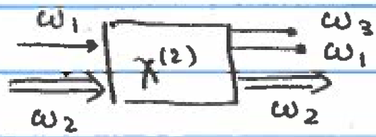
~~For the~~ For the phase-matched conditions

$$\frac{d\mathcal{E}_3}{dz} = \frac{ik_3}{8\varepsilon_0}\mathcal{E}_1\mathcal{E}_2\chi^{(2)} = g_3 \qquad \mathcal{E}_3(z) = g_3 \cdot z$$

linear growth

This valid for the undepleted pump approx, if $\mathcal{E}_3$ becomes comparable with $\mathcal{E}_1$ & $\mathcal{E}_2$ $\rightarrow$ more complex, need to solve for all three fields

Up-conversion (sum-frequency generation with one strong & one weak field)



$\omega_3 = \omega_1 + \omega_2$

photon exchange b/w ω_1 & ω_3

Strong ω_2 – pump, ω_1 – signal, ω_3 – idler

In this case $\mathcal{E}_1$ and $\mathcal{E}_3$ are weak and changing, and $\mathcal{E}_2$ is strong and constant

$$\frac{d\mathcal{E}_3}{dz} = \frac{ik_3}{8\varepsilon_0}\chi^{(2)}_{(\omega_1,\omega_2,-\omega_3)}\mathcal{E}_1\mathcal{E}_2\, e^{i\Delta k z}$$

$$\frac{d\mathcal{E}_1}{dz} = \frac{ik_1}{8\varepsilon_0}\chi^{(2)}_{(+\omega_1,-\omega_2,+\omega_3)}\mathcal{E}_2^*\mathcal{E}_3\, e^{-i\Delta k z}$$

If $\Delta k = 0$ (perfect phase matching)

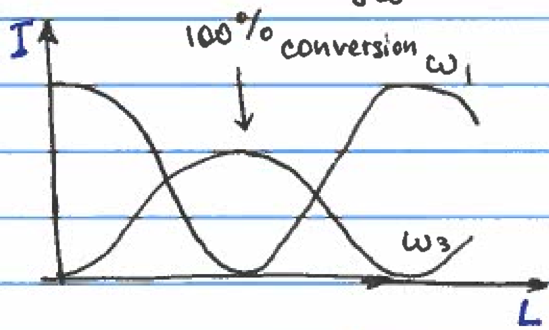
$$\frac{d^2\mathcal{E}_3}{dz^2} = \frac{ik_3}{8\varepsilon_0}\chi^{(2)}\mathcal{E}_2\frac{d\mathcal{E}_1}{dz} = -\underbrace{\frac{k_1k_3}{64\varepsilon_0}|\chi^{(2)}|^2|\mathcal{E}_2|^2}_{\varkappa^2}\mathcal{E}_3$$

similar eqn for $\mathcal{E}_1$

$$\mathcal{E}_1(z) = \mathcal{E}_1(0)\cos\varkappa z$$

$$\mathcal{E}_3(z) = -\mathcal{E}_1(0)\frac{\varkappa}{\frac{ik_1}{8\varepsilon_0}\chi^{(2)}\mathcal{E}_2^*}\sin\varkappa z \simeq -\mathcal{E}_1(0)\sqrt{\frac{k_3}{k_1}}\sin\varkappa z$$

$\times$ 'phase' factor



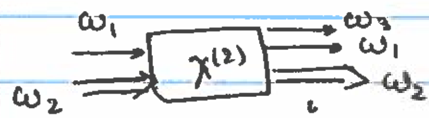
$$I_1(z) = I_1(0)\cos^2\varkappa z$$

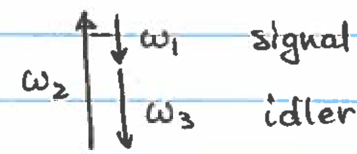
$$I_3(z) = I_1(0)\left(\frac{k_3}{k_1}\right)^* \sin^2\varkappa z$$

energy mismatch

$\omega_1 \neq \omega_3$

Frequency difference generation





$$\begin{cases} \dfrac{d\mathcal{E}_3}{dz} = \dfrac{ik_3}{8\varepsilon_0}\chi^{(2)}(-\omega_1,\omega_2,-\omega_3)\,\mathcal{E}_2\mathcal{E}_1^* \,\cancel{e^{i\Delta kz}}^{=1} \\ \dfrac{d\mathcal{E}_1}{dz} = \dfrac{ik_1}{8\varepsilon_0}\chi^{2}(-\omega_1,\omega_2,-\omega_3)\,\mathcal{E}_2\mathcal{E}_3^* \,\cancel{e^{i\Delta kz}}^{=1} \end{cases}$$

$$\frac{d\mathcal{E}_1^*}{dz} = -\frac{ik_1}{8\varepsilon_0}\chi^{(2)}\,\mathcal{E}_2^*\mathcal{E}_3$$

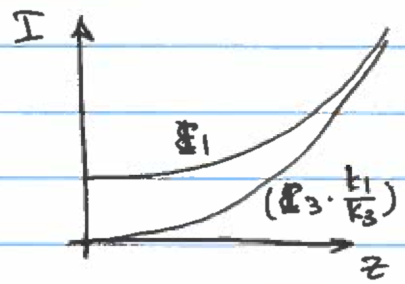
$$\begin{cases} \dfrac{d^2\mathcal{E}_3}{dz^2} = \dfrac{k_1k_3}{64\varepsilon_0^2}|\chi^{(2)}|^2\,|\mathcal{E}_2|^2\mathcal{E}_3 = \varkappa^2\mathcal{E}_3 \\ \dfrac{d^2\mathcal{E}_1}{dz^2} = \varkappa^2\mathcal{E}_1 \end{cases}$$

$$\mathcal{E}_1 = \mathcal{E}_1(0)\cosh\varkappa z$$

$$\mathcal{E}_3 = \mathcal{E}_1^*(0)\left(i\frac{\mathcal{E}_2}{|\mathcal{E}_2|}\sqrt{\frac{k_3}{k_1}}\right)\sinh\varkappa z$$

$$I_1(z) = I_1(0)\cosh^2\varkappa z$$

$$I_3(z) = I_3(0)\frac{k_3}{k_1}\cancel{\cosh^2}\sinh^2\varkappa z$$



Both fields grow exponentially and simultaneously until the pump is depleted

Parametric amplification