n-wave mixing process

$$\vec{P}(t) \approx \vec{P}^{(1)}(t)$$

In general:

$$\vec{P}(t) = \underline{\vec{P}^{(1)}(t)} + \underbrace{\vec{P}^{(2)}(t) + \vec{P}^{(3)}(t) + \dots}_{\vec{P}^{(NL)}(t)}$$

n-wave mixing

n+1 wave mixing

n - incoming waves

1 new field

Incoming field

$$E(\vec{r},t) = \sum_{j=1}^{n} E_j(\vec{r},t)\, e^{i\vec{k}_j\cdot\vec{r} - i\omega_j t} + c.c.$$

We neglect absorption of new field

$$\varepsilon(\omega_j) = n_j^2$$

$$k_j = \frac{\omega_j}{c} n_j$$

simplification

$$\omega_j = \omega$$

$$n_j = n$$

Wave equation

$$-\nabla^2 E(\vec{r},t) + \frac{n^2}{c^2}\frac{\partial^2}{\partial t^2} E(\vec{r},t) = -\frac{1}{c^2\varepsilon_0}\frac{\partial^2}{\partial t^2} \underbrace{P^{(NL)}(t,\vec{r})}_{\text{induced by } \vec{E}}$$

We will justify later

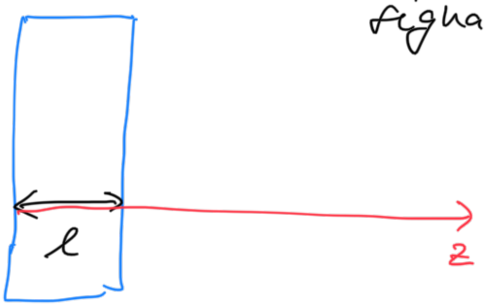
$$\vec{k}_s = \pm\vec{k}_1 \pm \vec{k}_2 \pm \vec{k}_3 \pm \ldots \pm \vec{k}_m$$ ← order of response

↑ incoming wave vectors

$$\omega_s = \pm\omega_1 \pm \omega_2 \pm \omega_3 \pm \ldots \pm \omega_m$$

$$\boxed{P^{(NL)}(\vec{r},t) = \sum_{m=2,3,\ldots} \sum_s P_s^{(m)}(t)\, e^{i\vec{k}_s\cdot\vec{r} - i\omega_s t}}$$

Geometry of the experiment



signal $\vec{k}_s$ → along the direction of the z-axis

$k_s l \gg 1$

many wavelengths in l

<u>Single component</u> of $P^{(NL)}(t)$

$$P^{(NL)}(\vec{r},t) = P_s(t)\, e^{i\vec{k}_s\cdot\vec{r} - i\omega_s t}$$

$\vec{k}_s = (0, 0, k_s)$

↑ length of $\vec{k}_s$

Solution of the wave-equation

$$E(\vec{r},t) = E_s(z,t)\, e^{-i\omega_s t + \vec{k}_s{}'\cdot\vec{r}}$$

z-direction

$$k_s' = \frac{\omega_s}{c} n_s$$

$\vec{k}_s{}'$ can be different from $\vec{k}_s$

we work with amplitudes or envelopes $P_s(t)$

Example: Time-derivative

$$\frac{\partial}{\partial t} P_s(t)\, e^{-i\omega_s t} = \underbrace{\left(\frac{\partial}{\partial t} P_s(t)\right)}_{\approx 0} e^{-i\omega_s t} \left(-i\omega_s P_s(t)\right) e^{-i\omega_s t}$$

$$\approx -i\omega_s P_s(t)\, e^{-i\omega_s t}$$

$$\left|\frac{\partial}{\partial t} P_s(t)\right| \ll \left|\omega_s P_s(t)\right|$$

Simplification of the wave-equation

spatial change of electric field

$$\frac{\partial^2}{\partial z^2} E_s(z)\, e^{i\vec{k}_s{}'\cdot\vec{r}} = ?$$

$$\frac{\partial}{\partial z} E_s(z)\, e^{i\vec{k}\cdot\vec{r}} = \left(\frac{\partial}{\partial z} E_s(z)\right) e^{i\vec{k}\cdot\vec{r}} + i k_z E_s(z)\, e^{i\vec{k}\cdot\vec{r}}$$

$$\frac{\partial}{\partial z}\left(\frac{\partial}{\partial z} E_s(z)\, e^{i\vec{k}\cdot\vec{r}}\right) = \underbrace{\cancel{\left(\frac{\partial^2}{\partial z^2} E_s(z)\right)}}_{\approx 0} e^{i\vec{k}\cdot\vec{r}} \cancel{+ i k_z \left(\frac{\partial}{\partial z} E_s(z)\right)} e^{i\vec{k}\cdot\vec{r}}$$

$$+2ik_z\left(\frac{\partial}{\partial z}E_s(z)\right)e^{i\vec{k}\cdot\vec{r}} - k_z^2 E_s(z)\, e^{i\vec{k}\cdot\vec{r}}$$

$$\underline{\nabla^2 E_s(\vec{r},t)\, e^{i\vec{k}_s'\cdot\vec{r}}} \approx 2i|\vec{k}_s'|\left(\frac{\partial}{\partial z}E_s(z)\right)e^{i\vec{k}'\cdot\vec{r}} - |\vec{k}_s'|^2 E_s(z)\, e^{i\vec{k}'\cdot\vec{r}}$$

$$-2i|\vec{k}_s'|\left(\frac{\partial}{\partial z}E_s(z)\right)e^{i\vec{k}_s'\cdot\vec{r}} + \cancel{|\vec{k}_s'|^2 E_s(t)\, e^{i\vec{k}_s'\cdot\vec{r}}}$$

$$\cancel{-\frac{n^2\omega_s^2}{c^2}E_s(z)}\, e^{i\vec{k}_s'\cdot\vec{r}} = \frac{\omega_s^2}{c^2\varepsilon_0}P_s(t)\, e^{i\vec{k}_s\cdot\vec{r}}$$

$$k_s' = \frac{n\omega_s}{c}$$

$$ik_s'\frac{\partial}{\partial z}E_s(z) = -\frac{\omega_s^2}{2c^2\varepsilon_0}P_s(t)\, e^{i(\vec{k}_s-\vec{k}_s')_z z}$$

$$\Delta k = (\vec{k}_s - \vec{k}_s')_z$$

Integrate:

$$E_s(z,t) = +i\frac{\omega_s^2}{k_s' c^2\varepsilon_0}P_s(t)\int_0^z dz'\, e^{i\Delta k z'}$$

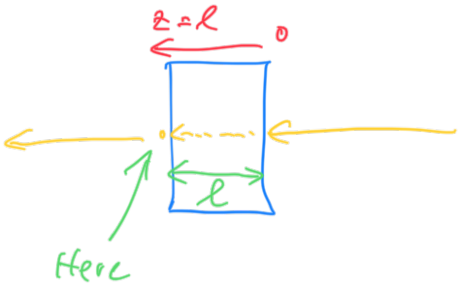
$$\boxed{E_s(z,t)} = i\frac{\omega_s}{c n_s\varepsilon_0}P_s(t)\frac{1}{i\Delta k}\left(e^{i\Delta k z} - 1\right)$$

$$= i\frac{\omega_s}{c n_s\varepsilon_0}P_s(t)\, e^{i\frac{\Delta k z}{2}}\, z\,\frac{\left(e^{i\frac{\Delta k z}{2}} - e^{-i\frac{\Delta k z}{2}}\right)}{2i\frac{\Delta k}{2}z}$$

$$\boxed{= i \frac{\omega_s}{c n_s \varepsilon_0} P_s(t)\, e^{i\frac{\Delta k z}{2}}\, z \,\mathrm{sinc}\left(\frac{\Delta k z}{2}\right)}$$

$\Delta k = 0$

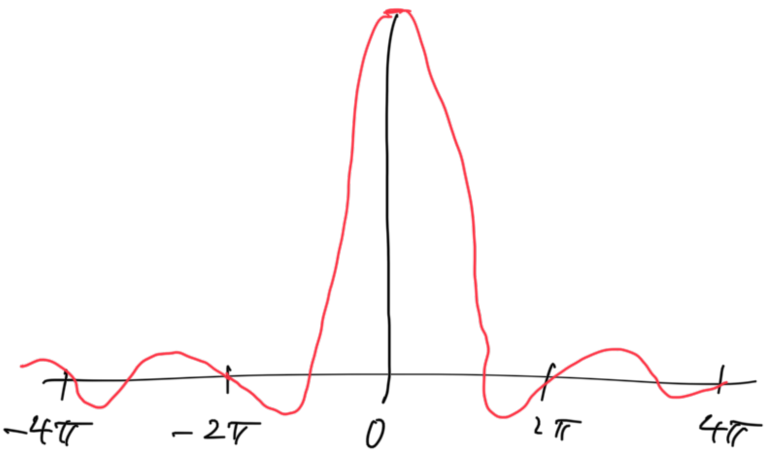
$$E_s^{(\Delta k = 0)}(l,t) = i \frac{\omega_s}{c n_s \varepsilon_0} P_s(t)\, e^{i\frac{\Delta k l}{2}}\, l$$



Intensity of the signal

$$I_s(t,l) \simeq |E_s(t,l)|^2 = \frac{\omega_s^2}{c^2 n_s^2 \varepsilon_0^2} |P_s(t)|^2 l^2 \,\mathrm{sinc}^2\left(\frac{\Delta k l}{2}\right)$$

$$\mathrm{sinc}(x) = \frac{\sin x}{x}$$



for $l \to \infty$

$$\mathrm{sinc}\left(\frac{\Delta k l}{2}\right) \longrightarrow \delta(\Delta k l)$$

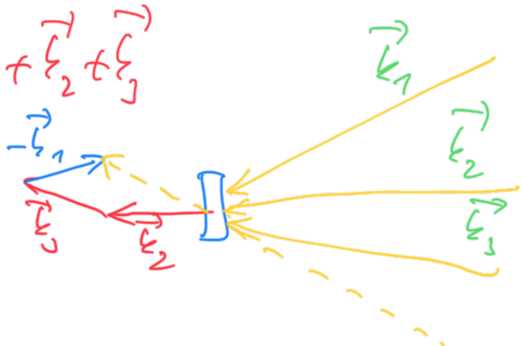
$|\Delta k l| \ll \pi \;\to\; \Delta k$ is small

Signal appears only in directions

$$\vec{k}_s = \pm \vec{k}_1 \pm \vec{k}_2 \pm \ldots$$

$\boxed{\text{Phase-matching condition}}$

$\vec{k}_s = -\vec{k}_1 + \vec{k}_2 + \vec{k}_3$



$\Leftarrow$ 4-wave mixing process

<u>Detection of non-linear signal</u>

$$I \sim |E|^2$$

Trick: <u>heterodyne dection</u>

Mix the weak signal E_s with a strong E_{Lo}

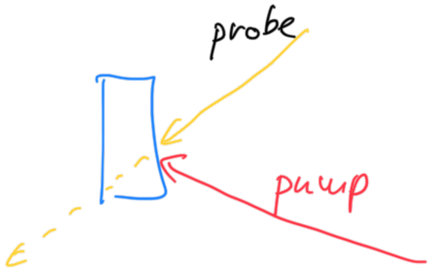
Local oscillator

$$I_T(t) \simeq |E_{Lo} + E_s|^2 = \underbrace{|E_{Lo}|^2}_{\text{well-known}} + \underbrace{\cancel{|E_s|^2}}_{\text{very small}} + \underline{2\,\mathrm{Re}(E_{Lo}^* E_s)}$$

$$I_{HET} \approx 2\,\mathrm{Re}(E_{Lo}^* E_s)$$

depends only linearly on E_s

Pump-probe experiment



$$E_s \sim E_{pump}^2 E_{probe}$$

$$\vec{k}_s = \underbrace{-\vec{k}_{pu} + \vec{k}_{pu}}_{=0} + \vec{k}_{pr}$$

$$E_s \approx i\omega_s P_s l$$

$$\boxed{I_{HET}^{probe} \approx \omega_s l \, \mathrm{Im}\left[E_{probe}^*(t) \, P_s(t) \right]}$$

Absorption of the probe

Signal is self-heterodyned!

The method is homodyne detection