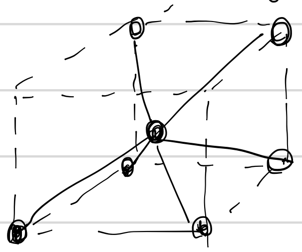
Inverse symmetry and $\chi^{(2)}$

Most materials have isotropic structure, like glass, water.....
It means if you rotate, reflect, inverse the material, the property of the material is identical to the original one.
Some crystal structures also have inverse symmetry.

Why inverse symmetry matters?



Suppose we choose our axis $\uparrow$ from the center, and we have.

Body center cubic : $\vec{P}_{NL} = \varepsilon_0 \vec{\vec{\chi}}^{(2)}_{\uparrow} \cdot \vec{E}\vec{E}$

Now if we reverse our axis to $\downarrow$, then

$\vec{E} \to -\vec{E}$ in the new coordinate, $\vec{P}_{NL} \to -\vec{P}_{NL}$

So: $-\vec{P}_{NL} = \varepsilon_0 \vec{\vec{\chi}}^{(2)}_{\downarrow} (-\vec{E})(-\vec{E}) \Rightarrow \vec{P}_{NL} = -\varepsilon_0 \vec{\vec{\chi}}^{(2)}_{\downarrow} \vec{E}\vec{E}$.

So in $\uparrow$ coordinate, $\vec{P}_{NL} = \vec{\vec{\chi}}^{(2)}_{\uparrow} \vec{E}\vec{E}$

$\downarrow$ coordinate, $\vec{P}_{NL} = -\vec{\vec{\chi}}^{(2)}_{\downarrow} \vec{E}\vec{E}$.

But as we know, the body center cubic has inverse symmetry, meaning its property (physics) does not depend on the direction of your axis.

So: $\vec{\vec{\chi}}^{(2)}_{\uparrow} = \vec{\vec{\chi}}^{(2)}_{\downarrow} = \vec{\vec{\chi}}^{(2)}$

So for inverse symmetry materials:

$$\vec{P}_{NL} = \varepsilon_0 \vec{\vec{\chi}}^{(2)} \vec{E}\vec{E} \quad \text{in } \uparrow \text{ coordinate}$$
$$\vec{P}_{NL} = -\varepsilon_0 \vec{\vec{\chi}}^{(2)} \vec{E}\vec{E} \quad \text{in } \downarrow \text{ coordinate.}$$

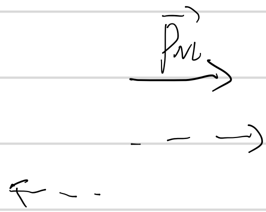
Only one solution: $\vec{\vec{\chi}}^{(2)} = 0$.

Deeper perspective:

$$\vec{P}_{NL} = \varepsilon_0 \vec{\vec{\chi}}^{(2)} \vec{E}\vec{E}$$

has different parity. $\vec{P}_{NL}$ has parity (-1), odd. meaning everytime you flip your coordinate,

$\vec{P}_{NL} \rightarrow -\vec{P}_{NL}$; So parity is -1.



The right-hand side: $\vec{E}\vec{E}$ has even parity. (1)

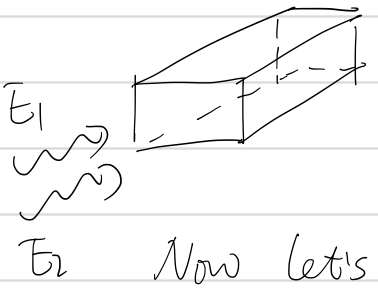
$(-\vec{E})(-\vec{E}) = \vec{E}\vec{E}$. flip coordinate does not matter.

So the only way that $\vec{P}_{NL} = \varepsilon_0 \vec{\vec{\chi}}^{(2)} \vec{E}\vec{E}$ has non-trivial solution, is that $\vec{\vec{\chi}}^{(2)}$ has odd parity.

$\vec{\vec{\chi}}^{(2)} \rightarrow -\vec{\vec{\chi}}^{(2)}$ when inverse coordinate.

Non-inverse symmetry $\rightarrow$ Non-zero $\vec{\vec{\chi}}^{(2)}$

Sum and difference frequency generation.



Previously, we noticed that in $\vec{P}_{NL} = \overleftrightarrow{\chi}^{(2)} \vdots \vec{E}\vec{E}$, $(\vec{E})^2 \rightarrow E_1 E_2$ term, and $E_1 E_2^*$ term.

Now let's suppose we have two input frequency; and three output

$$\vec{E}(z=0) = \vec{E}_1 e^{-i\omega_1 t + ik_1 z} + \vec{E}_2 e^{-i\omega_2 t + ik_2 z} + c.c.$$

We take sum frequency as an example:

$$\vec{E}(z) = \vec{e}_1 E_1(z) e^{-i\omega_1 t + ik_1 z} + \vec{e}_2 E_2(z) e^{-i\omega_2 t + ik_2 z} + \vec{e}_3 E_3(z) e^{-i\omega_3 t + ik_3 z} + c.c.$$

$\omega_3 = \omega_1 + \omega_2$.

General nonlinear equation gives:

$$\sum_j 2ik_j \vec{e}_j \frac{\partial E_j}{\partial z}(z) e^{-i\omega_j t + ik_j z} + c.c. = \mu_0 \frac{\partial^2}{\partial t^2} \vec{P}_{NL};$$

Let's again omit the complexity in polarization: $\vec{e}_j = \vec{e}_0$

take $\vec{P}_{NL} = \vec{e}_0 \varepsilon_0 \chi^{(2)} E^2$

$E^2 = (E_1 + E_2 + E_3 + E_1^* + E_2^* + E_3^*)^2$; We know phase-matching in a condition for $\chi^{(2)}$ process to happen. So this time, we quickly pick those combinition that has ω_1, ω_2 and $\omega_3 = \omega_1 + \omega_2$.

$$E^2 = (E_1 + E_2 + E_3 + E_1^* + E_2^* + E_3^*)^2$$

$$\sim \underbrace{2E_1E_2}_{\omega_1+\omega_2=\omega_3} + \underbrace{2E_3E_1^*}_{\omega_3-\omega_1=\omega_2} + \underbrace{2E_3E_2^*}_{\omega_3-\omega_2=\omega_1} + c.c.$$

$$\vec{P}_{NL} \to 2\vec{\varepsilon_0}\mu_0\varepsilon_0\chi^{(2)} \Big(E_1E_2\, e^{-i\omega_3 t + i(k_1+k_2)z} + E_3E_1^*\, e^{-i\omega_2 t + i(k_3-k_1)z} + E_3E_2^*\, e^{-i\omega_1 t + i(k_3-k_2)z} + c.c. \Big)$$

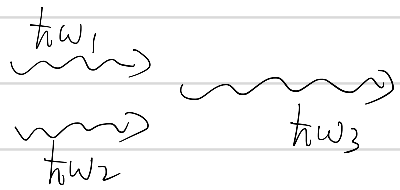
So nonlinear equation becomes:

$$\frac{\partial E_3(z)}{\partial z} = i\frac{\mu_0\varepsilon_0\chi^{(2)}\omega_3^2}{k_3} E_1(z)E_2(z)\, e^{i(k_1+k_2-k_3)z}$$

$$\frac{\partial E_2(z)}{\partial z} = i\frac{\mu_0\varepsilon_0\chi^{(2)}\omega_2^2}{k_2} E_3(z)E_1^*(z)\, e^{i(k_3-k_1-k_2)z}$$

$$\frac{\partial E_1(z)}{\partial z} = i\frac{\mu_0\varepsilon_0\chi^{(2)}\omega_1^2}{k_1} E_3(z)E_2^*(z)\, e^{i(k_3-k_1-k_2)z}$$

So again, phase matching condition $\Delta k = k_3 - k_1 - k_2 \to 0$

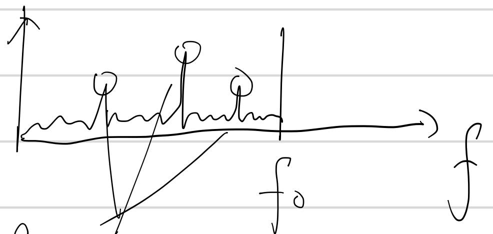


Sum frequency.

We now consider a particular case which is useful for information up-conversion.

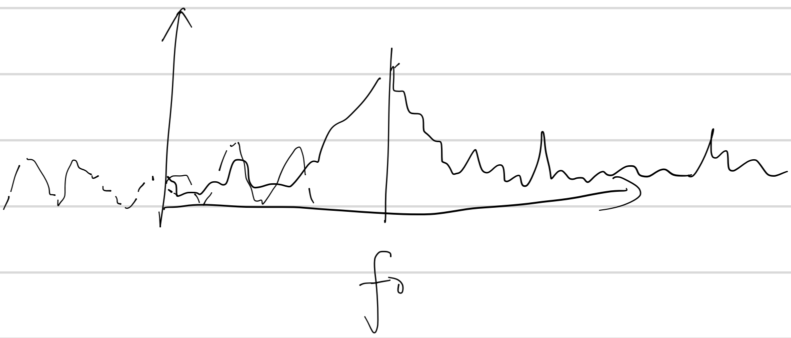
$$\boxed{\text{Possible Final exam: Verify energy conservation for sum frequency equations.}}$$

In information transportation, we always want the carrier to have higher frequency.



Information on sideband.

Because in general, the maximum bit rate is limited by the carrier frequency.



If the modulation rate is higher than carrier frequency, you will create base-band folding, and destroy the information!

Now assume $\omega_1 < \omega_2 < \omega_3$.
And we want to convert laser from ω_1 to ω_3.
ω_1 is small ; so is ω_3.

$|E_2| \gg |E_1|, |E_3|$, so take $E_2(z) = E_2(0)$.

Further assume $\Delta k = 0$, work in phase-matching condition:

we have:

$$\frac{\partial E_1}{\partial z} = i\mu_0\varepsilon_0\chi^{(2)}\frac{\omega_1^2}{k_1} E_2^*(0)\cdot E_3(z)$$

$$\frac{\partial E_3}{\partial z} = i\mu_0\varepsilon_0\chi^{(2)}\cdot\frac{\omega_3^2}{k_3} E_2(0)\cdot E_1(z)$$

Let:

$$i\mu_0\varepsilon_0\chi^{(2)}\frac{\omega_1^2}{k_1} E_2^*(0) = K_1$$

$$i\mu_0\varepsilon_0\chi^{(2)}\frac{\omega_3^2}{k_3} E_2(0) = K_3$$

It's relatively straightforward to solve this:

$$\frac{\partial^2 E_1}{\partial z^2} = \frac{i\mu_0 \varepsilon_0 \chi^{(2)} \omega_1^2}{k_1} E_2^*(0) \frac{\partial E_3}{\partial z} =$$

$$- (\mu_0 \varepsilon_0 \chi^{(2)})^2 \frac{\omega_1^2 \omega_3^2}{k_1 k_2} |E_2(0)|^2 E_1(z).$$

$$\therefore \frac{\partial^2 E_1}{\partial z^2} + \gamma^2 \cdot E_1(z) = 0. \quad ; \quad \gamma = -k_1 \cdot k_3$$

Oscillation equation: As $\gamma^2 > 0$, solution is:

$$E_1(z) = A_1 \cos\gamma z + B_1 \sin\gamma z ;$$

Similarly, we will have:

$$\frac{\partial^2 E_3}{\partial z^2} + \gamma^2 E_3(z) = 0 ;$$

$$E_3(z) = A_3 \cos\gamma z + B_3 \sin\gamma z ;$$

Boundary condition: $E_1(0) = E_1(0) = A_1$;

$E_3(0) = 0 = A_3$.

$$\therefore \begin{cases} A_1 = E_1(0) \\ A_3 = 0 \end{cases}$$

Second: as $E_3(z=0) = 0$; $\frac{\partial E_1(z=0)}{\partial z} = 0 \Rightarrow -A_1\gamma \sin\gamma z + B_1\gamma \cos\gamma z = 0$.

$\therefore B_1 = 0$.

Similarly:

$$\frac{\partial E_3(z=0)}{\partial z} = K_3 \cdot E_1(z=0)$$

$$= \gamma B_3 \cos\gamma z \Big|_{z=0} = B_3 \gamma .$$

$$\therefore B_3 = \frac{K_3}{\gamma} E_1(0)$$

Simplify: $K_3 = i \mu_0 \varepsilon_0 \chi^{(2)} \frac{\omega_3^2}{k_3} E_2(0)$

$$\gamma = \sqrt{-K_1 K_3} = \left(\mu_0^2 \varepsilon_0^2 \chi^{(2)2} \frac{\omega_3^2 \omega_1^2}{k_1 k_3} |E_2|^2 \right)^{1/2}$$

$$= \mu_0 \varepsilon_0 \chi^{(2)} \frac{\omega_1 \omega_3}{\sqrt{k_1 k_3}} |E_2(0)|$$

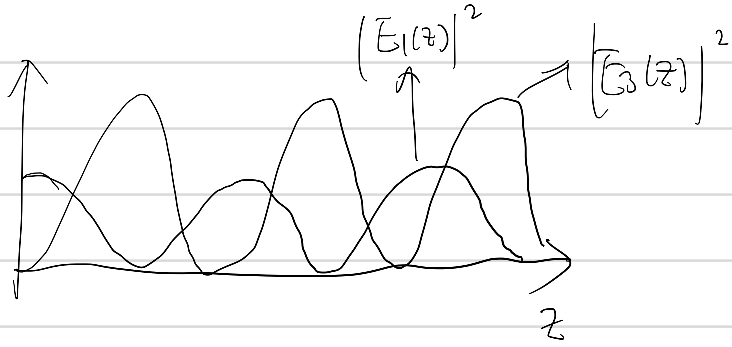
$$\therefore \frac{K_3}{\gamma} = i \frac{\omega_3^2 \sqrt{k_1 k_3}}{k_3 \cdot \omega_1 \omega_3} \frac{E_2(0)}{|E_2(0)|} = i \frac{\omega_3}{\omega_1} \sqrt{\frac{k_1}{k_3}} e^{i\varphi_2(0)};$$

phase

$$= i \frac{\omega_3}{\omega_1} \sqrt{\frac{n_1 \omega_1}{n_3 \omega_3}} e^{i\varphi_2(0)} = i \sqrt{\frac{n_1 \omega_3}{n_3 \omega_1}} e^{i\varphi_2(0)}$$

$$\therefore E_1(z) = E_1(0) \cos\gamma z$$

$$E_3(z) = i \sqrt{\frac{n_1 \omega_3}{n_3 \omega_1}} e^{i\varphi_2(0)} E_1(0) \sin\gamma z$$; $\omega_3 > \omega_1$. $|E_3|_{max} > |E_1|_{max}$.



Question 1: why can $|E_3|$ larger than $|E_1|$? Does this violate anything?

$$|E_3|^2_{max} = \frac{n_1 \omega_3}{n_3 \omega_1} |E_1(0)|^2.$$

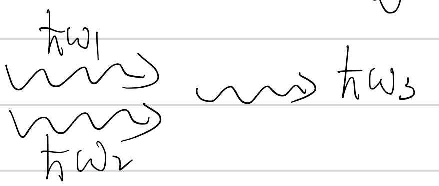
Notice: $\frac{\text{power}}{\hbar\omega} = \frac{\frac{1}{2} n \varepsilon_0 c A \cdot |E|^2}{\hbar \omega}$: photon per unit time.

$$\frac{P_{3,max}}{\hbar\omega_3} = \frac{\frac{1}{2} n_3 \varepsilon_0 c \cdot A \cdot |E_3|^2_{max}}{\hbar\omega_3} = \frac{\varepsilon_0 c \cdot A \cdot n_3 \cdot n_1 \omega_3}{2\hbar\omega_3 \, n_3 \omega_1} |E_1(0)|^2$$

$$= \frac{\varepsilon_0 c A n_1 |E_1(0)|^2}{2\hbar\omega_1}$$ = photon of $E_1(0)$ per unit time.

So all photon of $E_1(0)$ is converted to E_3, and because E_3 has higher energy per photon, so $|E_3|_{max} > |E_1|_{max}$.

Where does the energy comes from? from E_2



Question 2: why is the solution oscillating?

Consider when E_3 is max, and E_1 is zero.