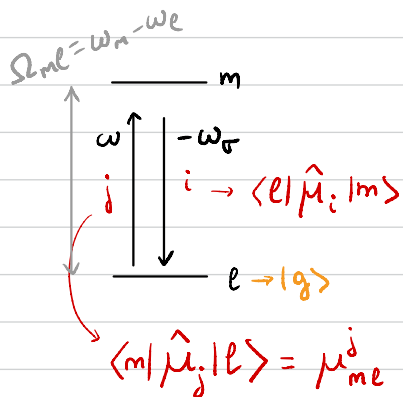


Week 05

I) Interpretation of the perturbative expressions

1) Linear suscept.

$$\chi_{ij}^{(1)}(-\omega_\sigma; \omega) = \frac{N/V}{\epsilon_0 \hbar} \sum_{lm} \rho_{ee}^{(0)} \left(\frac{\mu_{em}^i \mu_{me}^j}{\Omega_{me} - \omega - i\gamma_{me}} + \overset{\text{anti-resonant} \uparrow}{\frac{\mu_{em}^j \mu_{me}^i}{\Omega_{me} + \omega + i\gamma_{me}}} \right)$$



• upward arrow: photon annihilation
positive frequency

• downward — photon creation
negative frequency
amplification / stimulated emission

2) Perturbative expression for $\chi^{(2)}$

* We consider all the population in the ground state

* We look at sum-frequency generation (SFG): $\omega_\sigma = \omega_1 + \omega_2$, $\omega_1, \omega_2 > 0$

Only two terms can become near-resonant:

$$\chi_{ijk}^{(2)}(-\omega_\sigma; \omega_1, \omega_2) = \frac{N/V}{2\epsilon_0 \hbar^2} \sum_{mn} \left\{ \frac{\mu_{gn}^i \mu_{nm}^j \mu_{mg}^k}{(\Omega_n - \omega_\sigma)(\Omega_m - \omega_2)} \right. \quad (a_1) \rightarrow$$

$\mu_{gn}^i \mu_{nm}^k \mu_{mg}^j$ (a₂)

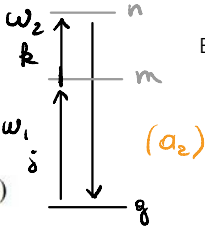
$(\Omega_n - \omega_\sigma)(\Omega_m - \omega_1)$

General expression for $\chi^{(2)}$

$\chi_{ijk}^{(2)}(\omega_p + \omega_q, \omega_q, \omega_p)$

$\omega_r = \omega_1 + \omega_2$

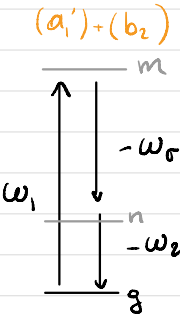
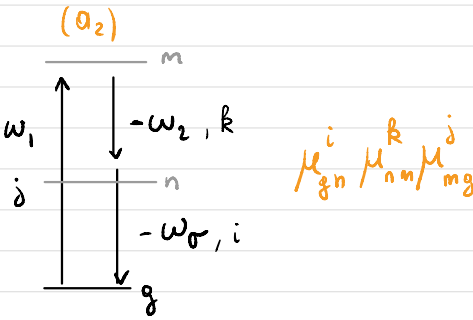
$$\begin{aligned} &= \frac{N}{2\epsilon_0\hbar^2} \sum_{lmn} \rho_{ll}^{(0)} \left\{ \frac{\mu_{ln}^i \mu_{nm}^j \mu_{ml}^k}{[(\omega_{nl} - \omega_p - \omega_q) - i\gamma_{nl}][(\omega_{ml} - \omega_p) - i\gamma_{ml}]} \right. \\ &+ \frac{\mu_{ln}^i \mu_{nm}^k \mu_{ml}^j}{[(\omega_{nl} - \omega_p - \omega_q) - i\gamma_{nl}][(\omega_{ml} - \omega_q) - i\gamma_{ml}]} \\ &+ \frac{\mu_{ln}^k \mu_{nm}^i \mu_{ml}^j}{[(\omega_{mn} - \omega_p - \omega_q) - i\gamma_{mn}][(\omega_{nl} + \omega_p) + i\gamma_{nl}]} \\ &+ \frac{\mu_{ln}^j \mu_{nm}^i \mu_{ml}^k}{[(\omega_{mn} - \omega_p - \omega_q) - i\gamma_{mn}][(\omega_{nl} + \omega_q) + i\gamma_{nl}]} \\ &+ \frac{\mu_{ln}^j \mu_{nm}^i \mu_{ml}^k}{[(\omega_{nm} + \omega_p + \omega_q) + i\gamma_{nm}][(\omega_{ml} - \omega_p) - i\gamma_{ml}]} \\ &+ \frac{\mu_{ln}^k \mu_{nm}^i \mu_{ml}^j}{[(\omega_{nm} + \omega_p + \omega_q) + i\gamma_{nm}][(\omega_{ml} - \omega_q) - i\gamma_{ml}]} \\ &+ \frac{\mu_{ln}^k \mu_{nm}^j \mu_{ml}^i}{[(\omega_{ml} + \omega_p + \omega_q) + i\gamma_{ml}][(\omega_{nl} + \omega_p) + i\gamma_{nl}]} \\ &\left. + \frac{\mu_{ln}^j \mu_{nm}^k \mu_{ml}^i}{[(\omega_{ml} + \omega_p + \omega_q) + i\gamma_{ml}][(\omega_{nl} + \omega_q) + i\gamma_{nl}]} \right\}. \end{aligned}$$



- (a2) far off-resonance (lossless) **DFG**
- (a'1) $\frac{\mu_{ln}^k \mu_{nm}^i \mu_{ml}^j}{(\omega_{nl} + \omega_p)(\omega_{ml} - \omega_q)}$
- (a'2) $\frac{\mu_{ln}^j \mu_{nm}^i \mu_{ml}^k}{(\omega_{nl} + \omega_q)(\omega_{ml} - \omega_p)}$
- (b1)
- (b2)
- (b'1)
- (b'2)

* For DFG $\omega_1 > \omega_2 > 0$ $\omega_r = \omega_1 - \omega_2 > 0$

$\chi_{ijk}^{(2)}(-\omega_r: \omega_1, -\omega_2) \rightarrow$ Two terms : (a2) and (a'1)+(b2)



II) Wave propagation in a non linear medium

$$\vec{\nabla}_x \vec{E} = -\mu_0 \dot{\vec{H}}$$

$$\vec{\nabla}_x \vec{H} = \epsilon_0 \dot{\vec{E}} + \dot{\vec{P}} \rightarrow \vec{\nabla}_x (\vec{\nabla}_x \vec{E}) = -\frac{1}{c^2} \ddot{\vec{E}} - \frac{1}{\epsilon_0 c^2} \ddot{\vec{P}}(\vec{E})$$

1) We neglect beam walk-off : $\vec{\nabla} \cdot \vec{E} = 0 \Rightarrow \vec{\nabla}_x (\vec{\nabla}_x \vec{E}) = -\nabla^2 \vec{E}$

2) We expand $\vec{P}(\vec{E}) = \vec{P}^{(1)}(\vec{E}) + \vec{P}^{(2)}(\vec{E}) \dots$

$$-\nabla^2 \vec{E}(\vec{r}, t) + \frac{1}{c^2} \ddot{\vec{E}}(\vec{r}, t) + \frac{1}{\epsilon_0 c^2} \ddot{\vec{P}}^{(1)}(\vec{r}, t) = -\frac{1}{\epsilon_0 c^2} \ddot{\vec{P}}^{(n)}(\vec{r}, t)$$

Fourier transform $t \rightarrow \omega$:

$$(1) \quad \nabla^2 \vec{E}_\alpha(\vec{r}) + \frac{\omega_\alpha^2 (1 + \chi_\alpha^{(1)})}{c^2} \vec{E}_\alpha(\vec{r}) = \frac{-\omega_\alpha^2}{\epsilon_0 c^2} \vec{P}_\alpha^{(n)}(\vec{r})$$

$\downarrow$ $\vec{E}(\omega_\alpha)$ $\uparrow$ $\vec{P}_\alpha^{(1)} = \epsilon_0 \chi_\alpha^{(1)} \vec{E}_\alpha$

3) We consider plane waves polarized along $\vec{u}_\alpha$ propagating along z .

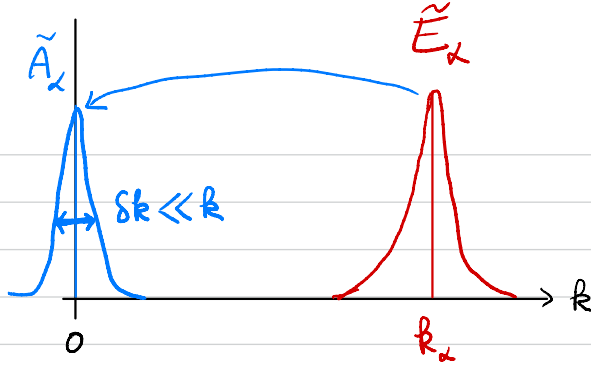
4) Slowly varying envelope approximation (SVEA):

$$\vec{E}_\alpha(z) = A_\alpha(z) \vec{u}_\alpha e^{i k_\alpha z} \quad \text{with} \quad k_\alpha = \frac{\omega_\alpha n_\alpha}{c}$$

$$\downarrow$$

$$\vec{E}_\alpha(k)$$

$$n_\alpha = \sqrt{1 + \chi_\alpha^{(1)}}$$



$$\frac{\partial \tilde{A}_\alpha}{\partial \delta} \approx |\delta k \tilde{A}_\alpha| \ll k_\alpha A_\alpha$$

$$\frac{\partial^2 A_\alpha}{\partial \delta^2} \ll k_\alpha \frac{\partial A_\alpha}{\partial \delta}$$

SVEA

Now: $\nabla^2 \vec{E}_\alpha(\hat{n}) = \left(\cancel{\frac{\partial^2 A_\alpha}{\partial \delta^2}} + 2ik_\alpha \frac{\partial A_\alpha}{\partial \delta} - k_\alpha^2 A_\alpha \right) e^{ik_\alpha \delta} \vec{u}_\alpha$

↓
inject in (1) : cancel with $k_\alpha \vec{E}_\alpha(\delta)$

$$\hookrightarrow \frac{\partial A_\alpha}{\partial \delta} = \frac{i\omega_\alpha c}{2\varepsilon_0 n_\alpha} \vec{P}_\alpha^{(n)}(\delta) \cdot \vec{u}_\alpha e^{-ik_\alpha \delta} \quad (2)$$

$\vec{E}(-\omega_n) = \vec{E}_n^*$

where: $\vec{P}_\alpha^{(n)} = \varepsilon_0 K \chi^{(n)}(-\omega_\alpha; \omega_1, \dots, \omega_n) : \vec{E}(\omega_1) \dots \vec{E}(\omega_n)$

$$= \varepsilon_0 K \left(\chi^{(n)}(\dots) : \vec{u}_1 \dots \vec{u}_n \right) A(\omega_1) \dots A(\omega_n) e^{i(k_1 + \dots + k_n)\delta}$$

$$\frac{\partial A_\alpha}{\partial \delta} = \frac{i\omega_\alpha c}{2n_\alpha} \chi_{\text{eff}}^{(n)} A_1^{(*)}(\delta) \dots A_n^{(*)}(\delta) e^{i\Delta k \delta}$$

where

$$\Delta k = k_1 + \dots + k_n - k_\alpha = \frac{1}{c} (n_1 \omega_1 + \dots + n_n \omega_n - n_\alpha \omega_\alpha)$$

phase mismatch

$$\chi_{\text{eff}}^{(n)} = K \vec{u}_\alpha \cdot \chi^{(n)}(\dots) : \vec{u}_1 \dots \vec{u}_n$$

effective nonlinear suscept.