

EE236A notes

① Electromagnetic energy density

- Start with Maxwell's equations

$$\vec{\nabla} \times \vec{H} = \vec{J} + \frac{\partial \vec{D}}{\partial t} = \vec{J} + \frac{\partial}{\partial t} (\epsilon_0 \vec{E} + \vec{P})$$

$$\vec{\nabla} \times \vec{E} = - \frac{\partial}{\partial t} \mu_0 (\vec{H} + \vec{M})$$

$$\hookrightarrow \vec{E} \cdot \vec{\nabla} \times \vec{H} = \vec{E} \cdot \vec{J} + \epsilon_0 \vec{E} \cdot \frac{\partial \vec{E}}{\partial t} + \vec{E} \cdot \frac{\partial \vec{P}}{\partial t}$$

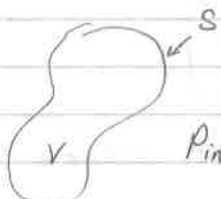
$$\text{and} \quad = \frac{1}{2} \frac{\partial}{\partial t} (\vec{E} \cdot \vec{E})$$

$$\vec{H} \cdot \vec{\nabla} \times \vec{E} = - \frac{\mu_0}{2} \frac{\partial}{\partial t} (\vec{H} \cdot \vec{H}) - \mu_0 \vec{H} \cdot \frac{\partial \vec{M}}{\partial t}$$

These expressions help us evaluate the divergence of the Poynting vector:

$$\begin{aligned} - \vec{\nabla} \cdot (\vec{E} \times \vec{H}) &= - \vec{H} \cdot \vec{\nabla} \times \vec{E} + \vec{E} \cdot \vec{\nabla} \times \vec{H} \\ &= \vec{E} \cdot \vec{J} + \frac{1}{2} \epsilon_0 \frac{\partial}{\partial t} (\vec{E} \cdot \vec{E}) + \frac{1}{2} \mu_0 \frac{\partial}{\partial t} (\vec{H} \cdot \vec{H}) \\ &\quad + \vec{E} \cdot \frac{\partial \vec{P}}{\partial t} + \mu_0 \vec{H} \cdot \frac{\partial \vec{M}}{\partial t} \\ &= \vec{E} \cdot \vec{J} + \frac{\partial}{\partial t} \left(\frac{1}{2} \epsilon_0 |\vec{E}|^2 + \frac{1}{2} \mu_0 |\vec{H}|^2 \right) + \vec{E} \cdot \frac{\partial \vec{P}}{\partial t} + \mu_0 \vec{H} \cdot \frac{\partial \vec{M}}{\partial t} \end{aligned}$$

Use Stokes' theorem to find the power flux into a surface



$$- \int_V \vec{\nabla} \cdot (\vec{E} \times \vec{H}) dV = - \int_S (\vec{E} \times \vec{H}) \cdot \hat{n} dA$$

$$P_{in}(S) = \int_V \left[\underbrace{\vec{E} \cdot \vec{J}}_{\text{Work done on currents}} + \underbrace{\frac{\partial}{\partial t} \left(\frac{\epsilon_0}{2} \mathcal{E}^2 + \frac{\mu_0}{2} \mathcal{H}^2 \right)}_{\text{energy stored in the fields}} + \underbrace{\vec{E} \cdot \frac{\partial \vec{P}}{\partial t}}_{\text{work done on electric polarization of medium}} + \underbrace{\mu_0 \vec{H} \cdot \frac{\partial \vec{M}}{\partial t}}_{\text{work done on magnetic polarization of medium}} \right] dV$$

- Let's concentrate on the $\vec{E} \cdot \frac{\partial \vec{P}}{\partial t}$ term

$$\cdot \text{let } \mathcal{E}(t) = \mathcal{E}(\omega) e^{i\omega t}$$

$$= \mathcal{E}(\omega) \cos \omega t + i \mathcal{E}(\omega) \sin \omega t$$

$$= \text{Re } \mathcal{E}(\omega) \cos \omega t + i \text{Im } \mathcal{E}(\omega) \cos \omega t + i \text{Re } \mathcal{E}(\omega) \sin \omega t$$

$$- \text{Im } \mathcal{E}(\omega) \sin \omega t$$

$$\text{Re } \mathcal{E}(t) = \text{Re } \mathcal{E}(\omega) \cos \omega t - \text{Im } \mathcal{E}(\omega) \sin \omega t$$

$$= \frac{1}{2} (\mathcal{E} + \mathcal{E}^*) \frac{1}{2} (e^{i\omega t} + e^{-i\omega t}) - \frac{1}{2i} (\mathcal{E} - \mathcal{E}^*) \frac{1}{2i} (e^{i\omega t} - e^{-i\omega t})$$

$$= \frac{1}{4} (\mathcal{E} + \mathcal{E}^*) (e^{i\omega t} + e^{-i\omega t}) + \frac{1}{4} (\mathcal{E} - \mathcal{E}^*) (e^{i\omega t} - e^{-i\omega t})$$

$$= \frac{1}{4} \mathcal{E} e^{i\omega t} + \frac{1}{4} \mathcal{E} e^{-i\omega t} + \frac{1}{4} \mathcal{E}^* e^{i\omega t} + \frac{1}{4} \mathcal{E}^* e^{-i\omega t}$$

$$+ \frac{1}{4} \mathcal{E} e^{i\omega t} - \frac{1}{4} \mathcal{E} e^{-i\omega t} - \frac{1}{4} \mathcal{E}^* e^{i\omega t} + \frac{1}{4} \mathcal{E}^* e^{-i\omega t}$$

$$\text{Re } \mathcal{E}(t) = \frac{1}{2} \mathcal{E} e^{i\omega t} + \frac{1}{2} \mathcal{E}^* e^{-i\omega t}$$

So we have $\text{Re}(\mathcal{E}) = \frac{1}{2}(\mathcal{E}e^{i\omega t} + \mathcal{E}^*e^{-i\omega t})$

we also have $P(t) = P(\omega)e^{i\omega t}$ where $P(\omega) = \epsilon_0 \chi \mathcal{E}(\omega)$

$\text{Re}(P) = \frac{1}{2}(Pe^{i\omega t} + P^*e^{-i\omega t})$

For some reason, Eli uses only the real part of \mathcal{E} and P in all calculations:

$$\frac{\partial P}{\partial t} = \frac{1}{2}(i\omega P(\omega)e^{i\omega t} - i\omega P^*(\omega)e^{-i\omega t})$$

Then:

$$\begin{aligned} \mathcal{E} \cdot \frac{\partial P}{\partial t} &= \frac{1}{4}(\mathcal{E}e^{i\omega t} + \mathcal{E}^*e^{-i\omega t})(i\omega P e^{i\omega t} - i\omega P^* e^{-i\omega t}) \\ &= \frac{1}{4}(i\omega \mathcal{E}P e^{i2\omega t} - i\omega \mathcal{E}P^* + i\omega \mathcal{E}^*P - i\omega \mathcal{E}^*P^* e^{-2i\omega t}) \end{aligned}$$

Take the time average: oscillating terms go to zero

$$\begin{aligned} \langle \mathcal{E} \cdot \frac{\partial P}{\partial t} \rangle &= \frac{1}{4}(-i\omega \mathcal{E} \cdot P^* + i\omega \mathcal{E}^* \cdot P) \\ &= \frac{1}{4}(-i\omega \mathcal{E} \cdot \epsilon_0 \chi^* \mathcal{E}^* + i\omega \mathcal{E}^* \cdot \epsilon_0 \chi \mathcal{E}) \\ &= \frac{1}{4} \epsilon_0 (-i\omega \chi^* \mathcal{E} \cdot \mathcal{E}^* + i\omega \chi \mathcal{E}^* \cdot \mathcal{E}) \\ &= \frac{1}{4} \epsilon_0 \underbrace{(i\omega \chi - i\omega \chi^*)}_{= -2\omega \text{Im} \chi} \mathcal{E} \cdot \mathcal{E}^* \end{aligned}$$

Thus, the average power dissipated per unit volume is:

$$\langle \mathcal{E} \cdot \frac{\partial P}{\partial t} \rangle = -\frac{1}{2} \epsilon_0 \omega \text{Im} \chi |\mathcal{E}(\omega)|^2$$

high frequency waves dissipate more power

- A naive formulation of the stored EM energy density

$$\begin{aligned} \text{Power: } &= \frac{\partial}{\partial t} \left(\frac{\epsilon_0}{2} \mathcal{E}^2 \right) + \mathcal{E} \cdot \frac{\partial P}{\partial t} = \frac{\partial}{\partial t} \left(\frac{\epsilon_0}{2} \mathcal{E}^2 \right) + \mathcal{E} \cdot \epsilon_0 \chi \frac{\partial \mathcal{E}}{\partial t} \\ &= \frac{\partial}{\partial t} \left(\frac{\epsilon_0}{2} \mathcal{E}^2 \right) + \frac{\partial}{\partial t} \left(\frac{\epsilon_0 \chi}{2} \mathcal{E}^2 \right) \\ &= \frac{\partial}{\partial t} \left(\frac{1}{2} \epsilon_0 (1 + \chi) \mathcal{E}^2 \right) = \frac{\partial}{\partial t} \left(\frac{1}{2} \epsilon(\omega) \mathcal{E}^2 \right) \end{aligned}$$

stored energy = $\frac{1}{2} \epsilon(\omega) |\mathcal{E}(\omega)|^2$ However, this does not properly account for the dispersive property of ϵ

- Electromagnetic energy density in dispersive media $\chi = \chi(\omega)$
- Pure frequencies do not really exist since the electric field must build up from $\mathcal{E} = 0$

$$\text{FT: } \mathcal{E}(t) = \int_{-\infty}^{\infty} \mathcal{E}(\omega) e^{i\omega t} d\omega$$

- let $\omega = \omega_0 + \alpha$

$$\begin{aligned} \Rightarrow \mathcal{E}(t) &= \int_{-\infty}^{\infty} \mathcal{E}(\omega_0 + \alpha) e^{i(\omega_0 + \alpha)t} d\alpha \\ &= e^{i\omega_0 t} \underbrace{\int_{-\infty}^{\infty} \mathcal{E}(\omega_0 + \alpha) e^{i\alpha t} d\alpha}_{\mathcal{E}_0(t)} = \mathcal{E}_0(t) e^{i\omega_0 t} \end{aligned}$$

If most frequency components are near ω_0 , this is the slowly varying amplitude to the wave $e^{i\omega_0 t}$

- Polarization:

$$\begin{aligned} P(t) &= \int_{-\infty}^{\infty} \epsilon_0 \chi(\omega) \mathcal{E}(\omega) e^{i\omega t} d\omega \\ &= \epsilon_0 e^{i\omega_0 t} \int_{-\infty}^{\infty} \chi(\omega_0 + \alpha) \mathcal{E}(\omega_0 + \alpha) e^{i\alpha t} d\alpha \end{aligned}$$

Taylor expand $\chi(\omega_0 + \alpha) = \chi(\omega_0) + \alpha \frac{\partial \chi}{\partial \omega}(\omega_0)$

$$\begin{aligned} &= \epsilon_0 e^{i\omega_0 t} \chi(\omega_0) \int_{-\infty}^{\infty} \mathcal{E}(\omega_0 + \alpha) e^{i\alpha t} d\alpha \\ &\quad + \epsilon_0 e^{i\omega_0 t} \frac{\partial \chi}{\partial \omega} \Big|_{\omega_0} \int_{-\infty}^{\infty} \mathcal{E}(\omega_0 + \alpha) \alpha e^{i\alpha t} d\alpha \end{aligned}$$

$$= \epsilon_0 e^{i\omega_0 t} \chi(\omega_0) \mathcal{E}_0(t) + \epsilon_0 e^{i\omega_0 t} \frac{\partial \chi}{\partial \omega} \Big|_{\omega_0} \left(-i \frac{\partial \mathcal{E}_0}{\partial t} \right)$$

$$P(t) = \epsilon_0 \left[\chi(\omega_0) \mathcal{E}_0(t) - i \frac{\partial \chi}{\partial \omega} \Big|_{\omega_0} \frac{\partial \mathcal{E}_0}{\partial t} \right] e^{i\omega_0 t}$$

- The power density is $\mathcal{E} \cdot \frac{\partial P}{\partial t}$, so find $\frac{\partial P}{\partial t}$ small

$$\frac{\partial P}{\partial t} = \epsilon_0 \left[\chi \frac{\partial \mathcal{E}_0}{\partial t} e^{i\omega_0 t} + \chi \mathcal{E}_0 i\omega_0 e^{i\omega_0 t} - i \frac{\partial \chi}{\partial \omega} \Big|_{\omega_0} \frac{\partial^2 \mathcal{E}_0}{\partial t^2} e^{i\omega_0 t} - i \frac{\partial \chi}{\partial \omega} \Big|_{\omega_0} \frac{\partial \mathcal{E}_0}{\partial t} i\omega_0 e^{i\omega_0 t} \right]$$

$$= \epsilon_0 \left[\chi \frac{\partial \mathcal{E}_0}{\partial t} + i\omega_0 \chi \mathcal{E}_0 + \omega_0 \frac{\partial \chi}{\partial \omega} \Big|_{\omega_0} \frac{\partial \mathcal{E}_0}{\partial t} \right] e^{i\omega_0 t}$$

valid?

$$= \epsilon_0 \left[\frac{\partial \mathcal{E}_0}{\partial t} \frac{\partial(\omega_0 \chi)}{\partial \omega_0} + i\omega_0 \chi \mathcal{E}_0 \right] e^{i\omega_0 t}$$

- Then using the real part of \mathcal{E} and P in the analysis as we did previously, and assuming χ is real, we have (for time average)

$$\begin{aligned} \text{power density} &= \frac{1}{4} \left(\mathcal{E}^* \frac{\partial P}{\partial t} + \mathcal{E} \frac{\partial P^*}{\partial t} \right) \\ &= \frac{1}{4} \left(\mathcal{E}_0^* e^{-i\omega_0 t} \epsilon_0 \left(\frac{\partial \mathcal{E}_0}{\partial t} \frac{\partial(\omega_0 \chi)}{\partial \omega_0} + i\omega_0 \chi \mathcal{E}_0 \right) e^{i\omega_0 t} \right. \\ &\quad \left. + \mathcal{E}_0 e^{i\omega_0 t} \epsilon_0 \left(\frac{\partial \mathcal{E}_0}{\partial t} \frac{\partial(\omega_0 \chi)}{\partial \omega_0} - i\omega_0 \chi \mathcal{E}_0 \right) e^{-i\omega_0 t} \right) \end{aligned}$$

more careful needed here
 this at least work if ϵ_0 is real

$$\begin{aligned} \text{power density} &= \frac{1}{4} \left(\epsilon_0 \dot{\epsilon}_0 \left(\frac{\partial \epsilon_0}{\partial t} \frac{\partial (\omega_0 X)}{\partial \omega_0} + i \omega_0 X / \epsilon_0 \right) + \epsilon_0 \dot{\epsilon}_0 \left(\frac{\partial \epsilon_0}{\partial t} \frac{\partial (\omega_0 X)}{\partial \omega_0} - i \omega_0 X / \epsilon_0 \right) \right) \\ &= \frac{1}{2} \epsilon_0 \frac{\partial \epsilon_0}{\partial t} \frac{\partial (\omega_0 X)}{\partial \omega_0} \epsilon_0 = \frac{1}{4} \epsilon_0 \frac{\partial \epsilon_0^2}{\partial t} \frac{\partial (\omega_0 X)}{\partial \omega_0} \end{aligned}$$

↑ should be 1/2

Integrate:

$$\begin{aligned} \text{energy density} &= \frac{1}{2} \epsilon_0 \frac{\partial (\omega_0 X)}{\partial \omega_0} \epsilon_0^2 + \frac{1}{2} \epsilon_0 \dot{\epsilon}_0^2 \\ &= \frac{1}{2} \epsilon_0 \frac{\partial (\omega_0 (1+X))}{\partial \omega_0} \epsilon_0^2 \end{aligned}$$

vacuum energy contribution

- Also equate $\epsilon = \epsilon_0$ in this step

The corrected expression for the stored EM energy is:

$$\text{energy density} = \frac{\epsilon_0}{2} \frac{d(\omega \epsilon(\omega))}{d\omega} |\vec{E}|^2 + \frac{\mu_0}{2} \frac{d(\omega \mu(\omega))}{d\omega} |\vec{H}|^2$$

Reduces to the naive expression if $\frac{\partial \epsilon}{\partial \omega} = 0$

Group velocity

- Maxwell's equations give the wave equation $\nabla^2 \vec{E} = \mu \epsilon \frac{\partial^2 \vec{E}}{\partial t^2}$
- Using $\vec{E} = \vec{E}_0 e^{i(\vec{k} \cdot \vec{r} - \omega t)}$ as a solution,

$$\text{phase velocity } \frac{\omega}{k} = \sqrt{\frac{1}{\mu \epsilon}} = \frac{1}{\sqrt{\epsilon_r \mu_r}} \frac{1}{\sqrt{\epsilon_0 \mu_0}} = \frac{c}{n}$$

$$\begin{aligned} \text{group velocity } \frac{\partial \omega}{\partial k} &= \left(\frac{\partial}{\partial \omega} \left(\frac{\omega n}{c} \right) \right)^{-1} = \left(\frac{\omega}{c} \frac{\partial n}{\partial \omega} + \frac{n}{c} \right)^{-1} \\ &= \frac{c}{\left(\omega \frac{\partial n}{\partial \omega} + n \right)} \end{aligned}$$

$$\boxed{v_g = \frac{c}{\partial(n\omega)/\partial \omega}}$$

- We can show that these two expressions for power flux are equivalent:

$$v_g \times \text{energy density} = |\vec{E} \times \vec{H}| = \mathcal{E} H$$


Poynting vector magnitude

- At the condition where $\frac{d(\omega n)}{d\omega}$ is constant with frequency, the group velocity is constant and pulses centered at that frequency are resistant to dispersion. This occurs when $\frac{d^2(\omega n)}{d\omega^2} = 0$

- For glass, this is $\sim \lambda = 1.3 \mu\text{m}$
- Does not coincide with minimum absorption $\sim \lambda = 1.55 \mu\text{m}$
- Does not mean material is nondispersive ($n(\omega)$ constant)

② Lorentz-Lorenz Oscillator Model for Dielectric Constant

- Begin with a harmonic oscillator spring model for electrons in a dielectric material


 • Apply Newton's 2nd law

$$F = m\ddot{x} + m\frac{\dot{x}}{\tau} + m\omega_0^2 x = q\mathcal{E}$$

\uparrow acceleration \uparrow damping \uparrow spring restoring force \swarrow driving field

- Let the solution be $x = x_0 e^{i\omega t}$

$$\Rightarrow \frac{q\mathcal{E}}{m} = -\omega^2 x + \frac{i\omega}{\tau} x + \omega_0^2 x = (\omega_0^2 - \omega^2)x + \frac{i\omega}{\tau} x$$

$$x = \frac{q\mathcal{E}/m}{(\omega_0^2 - \omega^2) + i\omega/\tau}$$

This is the electron's displacement from equilibrium, which forms a dipole moment. The polarization is the dipole moment volume density:

$$P(\omega) = Nq x \quad N = \# \text{ atoms/vol.}$$

$$P(\omega) = \underbrace{\frac{Nq^2/m}{\omega_0^2 - \omega^2 + i\omega/\tau}}_{\epsilon_0 \chi(\omega)} \cdot \mathcal{E}(\omega)$$

Thus, we have:

$$\chi(\omega) = \frac{Nq^2/m\epsilon_0}{\omega_0^2 - \omega^2 + i\omega/\tau} \quad \begin{array}{l} \text{single oscillator at } \omega_0 \\ \text{damping term: } 1/\tau \end{array}$$

- Separate the real and imaginary parts:

$$\chi'(\omega) = \frac{Nq^2}{m\epsilon_0} \frac{\omega_0^2 - \omega^2}{(\omega_0^2 - \omega^2)^2 + \omega^2/\tau^2}$$

$$\chi''(\omega) = \frac{Nq^2}{m\epsilon_0} \frac{\omega/\tau}{(\omega_0^2 - \omega^2)^2 + \omega^2/\tau^2}$$

- Consider the case of weak damping, and look at the real part

$$\epsilon_0 \epsilon'(\omega) = (1 + \chi'(\omega)) \epsilon_0 \approx \epsilon_0 \left(1 + \frac{Nq^2}{m\epsilon_0} \frac{1}{\omega_0^2 - \omega^2} \right)$$

$$\text{Limits: } \omega \ll \omega_0 : \epsilon'(\omega) = 1 + \frac{Nq^2}{m\epsilon_0 \omega_0^2}$$

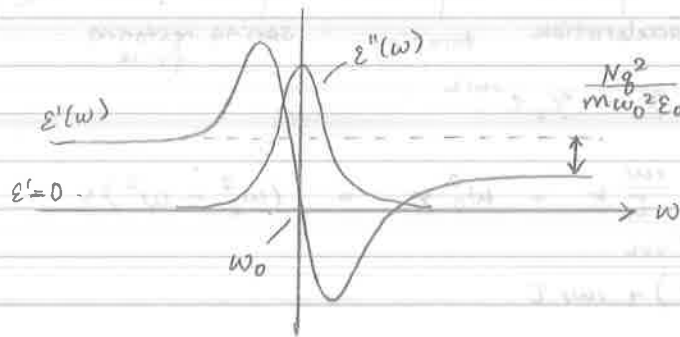
$$\omega \gg \omega_0 : \epsilon'(\omega) = 1$$

This is the step-down in ϵ' passing through a resonance

Near resonance:

$$\epsilon'(\omega) \approx 1 + \frac{N_0^2}{m\epsilon_0} \frac{1}{\omega_0^2 - \omega^2} = 1 + \frac{N_0^2}{m\epsilon_0} \frac{1}{(\omega_0 - \omega)(\omega_0 + \omega)} \quad \leftarrow \approx 2\omega_0$$

$$\epsilon'(\omega) \approx 1 + \frac{N_0^2}{2m\epsilon_0\omega_0} \frac{1}{\omega_0 - \omega}$$



⇒ For optical communication, two frequencies are of interest:

- $d\epsilon'/d\omega$ minimum $\rightarrow 1.3\mu\text{m}$
- ϵ'' minimum $\rightarrow 1.55\mu\text{m}$

near resonance
 $\omega \approx \omega_0$

Now look at the imaginary part:

$$\epsilon''(\omega) = \frac{N_0^2}{m\epsilon_0} \frac{\omega/\tau}{(\omega_0^2 - \omega^2)^2 + \omega^2/\tau^2} \approx \frac{N_0^2}{m\epsilon_0} \frac{\omega_0/\tau}{(\omega_0 - \omega)^2 (2\omega_0)^2 + \omega_0^2/\tau^2}$$

$$\epsilon''(\omega) = \frac{N_0^2}{m\epsilon_0} \frac{1/(4\omega_0\tau)}{(\omega_0 - \omega)^2 + 1/(2\tau)^2}$$

This frequency profile is famously known as a Lorentzian.

The integral of the Lorentzian has a simple expression:

$$\int_{-\infty}^{\infty} d\omega \epsilon''(\omega) = \frac{N_0^2}{m\epsilon_0} \frac{\pi}{2\omega_0} \int_{-\infty}^{\infty} \frac{1}{\pi} \frac{(1/2\tau) d\omega}{(\omega_0 - \omega)^2 + 1/(2\tau)^2} = 1$$

$$\int_{-\infty}^{\infty} d\omega \epsilon''(\omega) = \frac{N_0^2}{m\epsilon_0\omega_0} \cdot \frac{\pi}{2} = 1$$

This is dubious since the χ'' expression (Lorentzian) was found under the $\omega \ll \omega_0$ assumption, but here we integrate the Lorentzian from $\omega = -\infty$ to ∞

To make the model more realistic, introduce multiple oscillators:

$$\chi(\omega) = \sum_j \frac{N_0^2}{m\epsilon_0} \frac{1}{\omega_j^2 - \omega^2 - i\omega/\tau} \times S_j \quad \text{where } \sum_j S_j = 1$$

↖ resonance frequency of i th oscillator
↖ oscillator strength

The step-down in ϵ'

is reduced to:

$$\Delta\epsilon' = \frac{N_0^2}{m\epsilon_0\omega_j^2} S_j$$

- Absorption coefficient

• Start with dimensional analysis:

$$\frac{\text{dissipated power}}{\text{volume}} = \alpha(\omega) \times \frac{\text{power}}{\text{area}} \text{ (flux)}$$

$$\hookrightarrow \frac{1}{2} \epsilon_0 \omega \chi'' |\mathcal{E}|^2 = \alpha(\omega) \times \left(\frac{1}{2} \epsilon_0 \epsilon_r |\mathcal{E}|^2 \right) \times \frac{c}{\sqrt{\epsilon_r}}$$

↑
↑
↑
see §1
EM energy density
"naive" form
wave speed

$$\omega \chi'' = \alpha(\omega) \times c \sqrt{\epsilon_r}$$

$$\Rightarrow \boxed{\alpha(\omega) = \frac{\omega}{c \sqrt{\epsilon_r(\omega)}} \chi''(\omega)}$$

Absorption tends to increase with frequency

• Integrate the absorption spectrum:

$$\begin{aligned} \int_0^\infty \alpha(\omega) d\omega &= \int_0^\infty \frac{\omega}{c \sqrt{\epsilon_r}} \cdot \sum_i \frac{N_B^2}{m \epsilon_0} \frac{\omega/\tau \times S_i}{(\omega_i^2 - \omega^2)^2 + \omega^2/\tau^2} d\omega \\ &= \sum_i \frac{N_B^2}{m \epsilon_0} \frac{1}{c} \int_0^\infty \frac{\omega}{\sqrt{\epsilon_r}} \times \left[\frac{\omega/\tau d\omega}{(\omega_i^2 - \omega^2)^2 + \omega^2/\tau^2} \right] S_i \end{aligned}$$

Assume a narrow Lorentzian linewidth $\omega \rightarrow \omega_i$

* Assume $\sqrt{\epsilon_r}$ is constant at each resonance (dubious!)

integrated Lorentzian

$$\begin{aligned} \int_0^\infty \alpha(\omega) d\omega &= \sum_i \frac{N_B^2}{m \epsilon_0} \frac{\omega_i}{c \sqrt{\epsilon_r}} \times \frac{\pi}{2} \frac{1}{\omega_i} \times S_i \\ &= \frac{N_B^2}{m \epsilon_0} \frac{\pi}{2} \frac{1}{c \sqrt{\epsilon_r}} \sum_i S_i \end{aligned}$$

Express as: $\boxed{\int_0^\infty \alpha(\omega) d\omega = \frac{2\pi^2}{\sqrt{\epsilon_r}} \cdot \frac{N_B^2}{4\pi m \epsilon_0 c}}$

This is also called the oscillator strength "sum rule"

- Problems with the model

- 1) The index of refraction $\sqrt{\epsilon_r}$ should never have been taken out of the integral in the above derivation. It is unclear what index should be used for the sum rule
- 2) In writing the expression for $\alpha(\omega)$, we used the non-dispersive form of the EM energy density. But we then considered cases near resonance (where dispersion is maximum!)

→

3) Local fields were not taken into account

- The internal field (or local field) at an atom is not exactly the external field since the surrounding dipole moments will influence the local field.
- e.g. for a spherical hole, $E_i = E_0 + \frac{p}{3\epsilon_0}$

- Consider the case of no restoring force: free electron gas

$$\epsilon_r = 1 + \frac{Nq^2}{m\epsilon_0} \sum_i \frac{S_i}{\omega_i^2 - \omega^2 - i\omega/\tau}$$

$$= 1 + \frac{Nq^2}{m\epsilon_0} \frac{1}{-\omega^2 - i\omega/\tau} \quad \omega_i = 0$$

$$\epsilon_r = 1 - \frac{Nq^2}{m\epsilon_0} \frac{1}{\omega^2 + i\omega/\tau}$$

This is the dielectric constant for a plasma, or free electron gas.

- Assuming no damping, $\epsilon_r \rightarrow 0$ when:

$$0 > \epsilon_r = 1 - \frac{Nq^2}{m\epsilon_0} \frac{1}{\omega^2} \Rightarrow \omega^2 < \frac{Nq^2}{m\epsilon_0}$$

in the UV range in most metals

This is called the plasma frequency

$$\omega_p = \sqrt{\frac{Nq^2}{m\epsilon_0}}$$

- When $\epsilon_r < 0$, the refractive index n is pure imaginary so no waves can propagate in the medium
- The condition of $\epsilon_r < 0$ can also occur for a dielectric near resonance if the damping is weak. This is called the polariton condition.

- Coupled mode formulation of the oscillator model

- There is an alternative, equivalent method of deriving this model. It involves simultaneously solving two coupled equations: one for the material response \vec{P} , the other for the electromagnetic wave \vec{E} :

$$\textcircled{1} \text{ matter equation } [(\omega_0^2 - \omega^2) - \frac{i\omega}{\tau}] P = \frac{Nq^2}{m} \epsilon_0 E e^{-i\omega t}$$

Maxwell's equations in the time-harmonic, source-free formulation give:

$$-\nabla^2 \vec{E} + \mu_0 \epsilon_0 \omega^2 \vec{E} = -\mu_0 \omega^2 \vec{P}$$

$$\textcircled{2} \text{ EM equation: } (-k^2 + \mu_0 \epsilon_0 \omega^2) \vec{E} = -\mu_0 \omega^2 \vec{P}$$

From the matter equation we have $\rho = \frac{Nq^2/m \cdot \mathcal{E}}{(\omega_0^2 - \omega^2) - i\omega/\tau}$
 Insert this into the EM equation

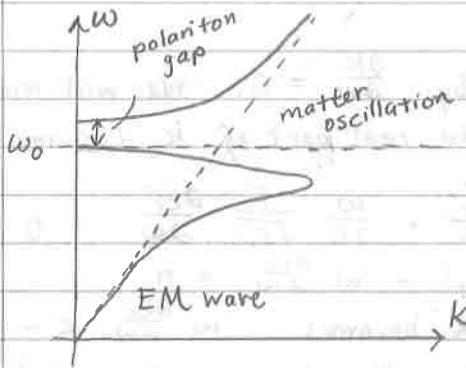
$$-k^2 \mathcal{E} + \mu_0 \omega^2 (\epsilon_0 \mathcal{E} + \rho) = -k^2 \mathcal{E} + \mu_0 \omega^2 \epsilon_r \epsilon_0 \mathcal{E} = 0$$

where $\epsilon_r = 1 + \frac{Nq^2/m\epsilon_0}{(\omega_0^2 - \omega^2) - i\omega/\tau}$
 exactly what we previously derived.

• Solve for the wave-vector k :

$$k^2 = \mu_0 \omega^2 \epsilon_r \epsilon_0 \Rightarrow k = \frac{\omega}{c} \sqrt{\epsilon_r}$$

The dispersive shape of $\epsilon_r(\omega)$ is known.
 Below we plot $k(\omega)$, which is what is more commonly called the dispersion relation



- When ω is small, $\epsilon_r = \text{constant}$, $k \rightarrow \frac{\omega}{c} n$
 - When ω is large, $\epsilon_r \rightarrow 1$, $k \rightarrow \frac{\omega}{c}$
- These are like normal EM waves. But what happens near the middle?

Polaritons

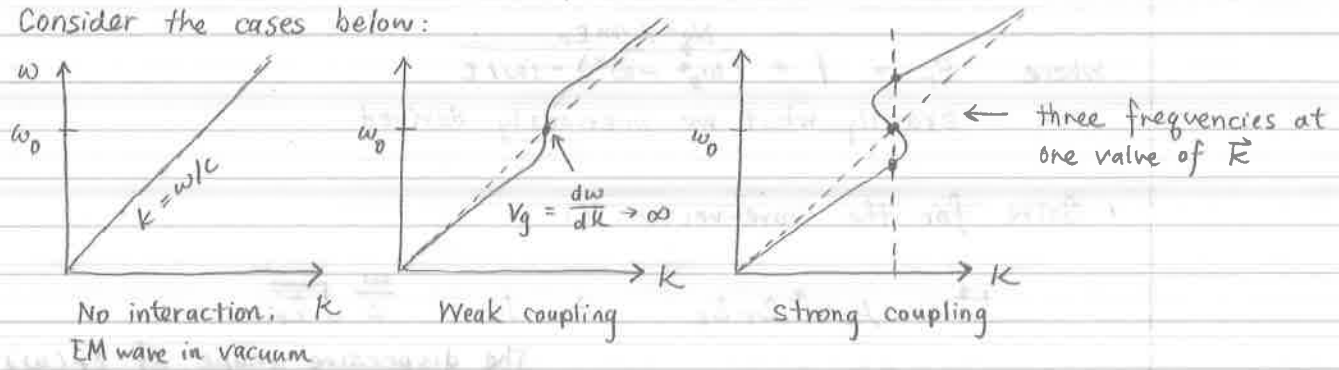
- Although the coupled mode formulation did not really tell us anything new, it does shed light on the nature of polaritons: these are coupled electromagnetic/matter oscillations
 - At very low or high frequencies, it behaves as expected
 - Near resonance, $\epsilon_r(\omega)$ changes rapidly, and ϵ_r' can be negative. This causes $k \propto \sqrt{\epsilon_r}$ to become imaginary, and a band is created wherein no waves can propagate \Rightarrow a polariton gap
- The existence of a polariton gap is the strongest statement of the so-called "polariton condition"
 - It requires 1) the oscillator density N to be high
 - 2) the damping τ^{-1} to be small

These conditions maximize the degree of anomalous dispersion at resonance ω_0 , which makes it more likely to achieve $\epsilon_r < 0$

 - In the absence of any damping ($\tau \rightarrow \infty$), the upper and lower bounds on the gap are ω_0 and $\sqrt{\omega_0^2 + \omega_p^2}$ where $\omega_p^2 = Nq^2/m\epsilon_0$

The necessary condition for a polariton gap to exist ($\epsilon_r' \rightarrow 0$) can be found to be approximately: $\omega_p^2 > 2\omega_0/\tau$

There are weaker statements of the polariton condition. Consider the cases below:



The weak coupling condition is given by $\frac{\partial k}{\partial \omega} = 0$. We will make a simplification by considering only the real part of k (assume $k'' = 0$):

Then:

$$\frac{\partial k}{\partial \omega} = \frac{\partial}{\partial \omega} \left(\frac{\omega}{c} \sqrt{\epsilon_r'} \right) = \frac{\sqrt{\epsilon_r'}}{c} + \frac{\omega}{2c} \frac{1}{\sqrt{\epsilon_r'}} \frac{\partial \epsilon_r'}{\partial \omega} = 0$$

$$2\epsilon_r' + \omega \frac{\partial \epsilon_r'}{\partial \omega} = 0$$

Condition becomes $\omega \frac{\partial \epsilon_r'}{\partial \omega} < -2\epsilon_r'$

If we evaluate this expression using the known relation for $\epsilon_r'(\omega)$ near resonance $\omega \approx \omega_0$, the condition can be rewritten as: $\omega_p^2 > 1/\tau^2$

Thus, weak coupling does not require as large of a value for ω_p than the condition for a polariton gap. (since $\omega_0 \gg 1/\tau$ in most cases of interest)

In practice this means that polariton response is seen in materials with a high density of oscillators (N) at a single resonance

③ Nonlinear Optical Susceptibility

- Nonlinear optical response:

$$P(t) = \epsilon_0 \left[\chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \dots \right]$$

linear
2nd order
3rd order

↑
↑
↑

only occurs in media without inversion symmetry
occurs in all media

- How do you know you have an anharmonic (nonlinear) oscillator?

- Order of magnitude analysis: nonlinear effects appear when the driving field is comparable to the electric field holding the atom together: $E_{\text{atom}} \approx 2V/0.1\text{nm} \sim 2 \times 10^8 \text{V/cm}$

- Thus for most materials the nonlinear susceptibilities will have magnitudes around:

$$\chi^{(2)} \approx \frac{\chi^{(1)}}{E_{\text{atom}}}, \quad \chi^{(3)} \approx \frac{\chi^{(1)}}{E_{\text{atom}}^2}$$

which gives: $P = \epsilon_0 \chi^{(1)} E \left[1 + \left(\frac{E}{E_{\text{atom}}} \right) + \left(\frac{E}{E_{\text{atom}}} \right)^2 + \dots \right]$

- Anharmonic Oscillator analysis (Eli & Boyd 1.4.3)

Extend the Lorentz model:

Non-centrosymmetric

$$\Rightarrow m\ddot{x} + m\frac{\dot{x}}{\tau} + \underbrace{m\omega_0^2 x + mDx^2}_{\text{restoring force}} = qE$$

Anharmonic component

The corresponding potential is:

$$U = -\int F dx = + \frac{1}{2} m\omega_0^2 x^2 + \frac{1}{3} m a x^3$$

harmonic
anharmonic

Assume electric field has the form $E = E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t}$

$$\Rightarrow \text{Re } E = \frac{1}{2} E_1 e^{i\omega_1 t} + \frac{1}{2} E_1^* e^{-i\omega_1 t} + \frac{1}{2} E_2 e^{i\omega_2 t} + \frac{1}{2} E_2^* e^{-i\omega_2 t}$$

Then, we solve the equation using perturbation theory assuming that $mDx^2 \ll m\omega_0^2 x$. Using the parameter λ ($0 < \lambda < 1$) to describe the perturbation, we rewrite the equation as:

$$\ddot{x} + \frac{\dot{x}}{\tau} + \omega_0^2 x + Dx^2 = \lambda \frac{qE}{m}$$

and we now expand x as a power series: $x = \lambda x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)} + \dots$

Substitute:

$$(\lambda \ddot{x}^{(1)} + \lambda^2 \ddot{x}^{(2)} + \lambda^3 \ddot{x}^{(3)}) + \frac{1}{\tau} (\lambda \dot{x}^{(1)} + \lambda^2 \dot{x}^{(2)} + \lambda^3 \dot{x}^{(3)}) + \omega_0^2 (\lambda x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)}) + D(\lambda x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)})^2 = \lambda g \mathcal{E} / m$$

Match powers of λ :

$$\begin{aligned} \lambda: \quad \ddot{x}^{(1)} + \frac{1}{\tau} \dot{x}^{(1)} + \omega_0^2 x^{(1)} &= \frac{g \mathcal{E}}{m} \longrightarrow x^{(1)} \text{ satisfies the lowest order equation} \\ \lambda^2: \quad \ddot{x}^{(2)} + \frac{1}{\tau} \dot{x}^{(2)} + \omega_0^2 x^{(2)} + D(x^{(1)})^2 &= 0 \\ \lambda^3: \quad \ddot{x}^{(3)} + \frac{1}{\tau} \dot{x}^{(3)} + \omega_0^2 x^{(3)} + D x^{(1)} x^{(2)} &= 0 \end{aligned}$$

- We have already solved the first equation: $x^{(1)} = \sum_n \frac{g \mathcal{E}_n / m}{\omega_0^2 - \omega_n^2 + i \omega_n / \tau} e^{-i \omega_n t}$

Introduce shorthand: $\mathcal{D}(\omega) = \omega_0^2 - \omega^2 - \frac{i \omega}{\tau}$: $x^{(1)} = \sum_n \frac{g \mathcal{E}_n / m}{\mathcal{D}(\omega_n)} e^{-i \omega_n t}$

- Substitute $x^{(1)}$ into the λ^2 equation to find $x^{(2)}$:

$$\ddot{x}^{(2)} + \frac{1}{\tau} \dot{x}^{(2)} + \omega_0^2 x^{(2)} = -D \left(\frac{g \mathcal{E} / m}{\mathcal{D}(\omega)} \right)^2$$

* Caution: when the electric field is squared, there are various possible frequency combinations:

$$\begin{aligned} (\text{Re } \mathcal{E})^2 &= \left(\frac{1}{2} \mathcal{E}_1 e^{i \omega_1 t} + \frac{1}{2} \mathcal{E}_1^* e^{-i \omega_1 t} + \frac{1}{2} \mathcal{E}_2 e^{i \omega_2 t} + \frac{1}{2} \mathcal{E}_2^* e^{-i \omega_2 t} \right)^2 \\ &= \frac{1}{4} \left[\mathcal{E}_1^2 e^{i 2 \omega_1 t} + 2 \mathcal{E}_1 \mathcal{E}_1^* e^{0 t} + 2 \mathcal{E}_1 \mathcal{E}_2 e^{i(\omega_1 + \omega_2)t} + 2 \mathcal{E}_1 \mathcal{E}_2^* e^{i(\omega_1 - \omega_2)t} \right. \\ &\quad \left. + \mathcal{E}_1^* \mathcal{E}_2 e^{-i(\omega_1 + \omega_2)t} + 2 \mathcal{E}_1^* \mathcal{E}_2 e^{i(\omega_2 - \omega_1)t} + 2 \mathcal{E}_1^* \mathcal{E}_2^* e^{i(\omega_1 - \omega_2)t} \right. \\ &\quad \left. + \mathcal{E}_2^2 e^{i 2 \omega_2 t} + 2 \mathcal{E}_2 \mathcal{E}_2^* e^{0 t} + \mathcal{E}_2^* \mathcal{E}_1 e^{-i(\omega_1 + \omega_2)t} \right] \end{aligned}$$

ignore negative freq. $\omega_1 > \omega_2$

$$(\text{Re } \mathcal{E})^2 = \frac{1}{4} \mathcal{E}_1^2 e^{i 2 \omega_1 t} + \frac{1}{4} \mathcal{E}_2^2 e^{i 2 \omega_2 t} + \frac{1}{2} \mathcal{E}_1 \mathcal{E}_2 e^{i(\omega_1 + \omega_2)t} + \frac{1}{2} \mathcal{E}_1 \mathcal{E}_2^* e^{i(\omega_1 - \omega_2)t} + \frac{1}{2} (\mathcal{E}_1 \mathcal{E}_1^* + \mathcal{E}_2 \mathcal{E}_2^*) e^{i \cdot 0 \cdot t}$$

Now solve for $x^{(2)}$ in the same way, separately for each combination.

$$x^{(2)}(2\omega_1) = -D \left(\frac{g \mathcal{E}_1 / m}{\mathcal{D}(\omega_1)} \right)^2 \frac{1}{\mathcal{D}(2\omega_1)}$$

$$x^{(2)}(2\omega_1) = -D \frac{(g^2 / m) (1/4) \mathcal{E}_1^2}{\mathcal{D}(\omega_1)^2 \cdot \mathcal{D}(2\omega_1)}$$

Then the polarization is:

$$p^{(2)}(2\omega_1) = N q x^{(2)}(2\omega_1) = \epsilon_0 x^{(2)}(2\omega_1) \mathcal{E}^2(\omega_1)$$

$$x^{(2)}(2\omega_1) = \frac{N q}{\epsilon_0} \frac{x^{(2)}(2\omega_1)}{\mathcal{E}^2(\omega_1)} \leftarrow \frac{1}{4} \mathcal{E}_1^2$$

The final frequency must satisfy $\omega_1 + \omega_2 + \omega_3 = 0$, since the overall free energy $F = e^{i(\omega_1 + \omega_2 + \omega_3)t}$ cannot be time-averaged to zero...

So:

$$\begin{aligned}\chi^{(2)}(-2\omega_1, \omega_1, \omega_1) &= \frac{Ng^3}{m^2\epsilon_0} D \cdot \frac{1}{2(\omega_1)^2 2(2\omega_1)} \\ &= D \cdot \frac{Ng^3}{m^2\epsilon_0} \left(\frac{Ng^2}{m\epsilon_0}\right)^{-3} \chi^{(1)}(\omega_1)^2 \chi^{(1)}(2\omega_1) \\ \chi^{(2)}(-2\omega_1, \omega_1, \omega_1) &= D \cdot \frac{m\epsilon_0^2}{N^2g^3} \chi^{(1)}(\omega_1)^2 \chi^{(1)}(2\omega_1)\end{aligned}$$

In general, it can be shown that:

$$\chi^{(2)}(-(\omega_1 + \omega_2), \omega_1, \omega_2) = D \cdot \frac{m\epsilon_0^2}{N^2g^3} \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_2) \chi^{(1)}(\omega_1 + \omega_2)$$

Miller's rule: observe that the $\chi^{(2)}/(\chi^{(1)})^3$ quotient will be a constant for every combination:

$$\frac{\chi^{(2)}(-(\omega_1 + \omega_2), \omega_1, \omega_2)}{\chi^{(1)}(\omega_1) \chi^{(1)}(\omega_2) \chi^{(1)}(\omega_1 + \omega_2)} = \frac{D \cdot m\epsilon_0^2}{N^2g^3}$$

- $\chi^{(2)} \sim (\chi^{(1)})^3$: higher index materials have greater nonlinear susceptibility.
- $\chi^{(2)}$ is greatest near resonances where $\chi^{(1)}$ is high. But absorption also tends to be high.

- Second-order nonlinear effects:

- 1) Second harmonic generation: $\chi^{(2)}(-2\omega, \omega, \omega)$
- 2) Sum/frequency generation: $\chi^{(2)}(-(\omega_1 \pm \omega_2), \omega_1, \omega_2)$
- 3) Optical rectification: $\chi^{(2)}(0, \omega, -\omega)$
Optically induced static field

- 4) Electro-optic effect: $\chi^{(2)}(-\omega, \omega, 0)$

$$\begin{aligned}P &= \epsilon_0 \chi^{(1)}(\omega) \mathcal{E} + \epsilon_0 \chi^{(2)}(-\omega, \omega, 0) \mathcal{E}^2 \\ &= \epsilon_0 \left(\chi^{(1)}(\omega) + \underbrace{\chi^{(2)}(-\omega, \omega, 0)} \right) \mathcal{E}\end{aligned}$$

Field-dependent refractive index change

- Relative strengths of nonlinear effects depends on expansion of \mathcal{E}^2

$$P^{(2)} = \epsilon_0 \chi^{(2)} \mathcal{E}^2$$

$$\begin{aligned}\frac{1}{\epsilon_0} P^{(2)} &= \chi^{(2)}(-2\omega_1, \omega_1, \omega_1) \left(\frac{1}{4} \mathcal{E}_1^2\right) e^{i2\omega_1 t} \\ &+ \chi^{(2)}(-2\omega_2, \omega_2, \omega_2) \left(\frac{1}{4} \mathcal{E}_2^2\right) e^{i2\omega_2 t} \\ &+ \chi^{(2)}(-(\omega_1 + \omega_2), \omega_1, \omega_2) \left(\frac{1}{2} \mathcal{E}_1 \mathcal{E}_2\right) e^{i(\omega_1 + \omega_2)t} \\ &+ \chi^{(2)}(-(\omega_1 - \omega_2), \omega_1, -\omega_2) \left(\frac{1}{2} \mathcal{E}_1 \mathcal{E}_2^*\right) e^{i(\omega_1 - \omega_2)t} \\ &+ \chi^{(2)}(0, \omega_1, -\omega_1) \left(\frac{1}{2} \mathcal{E}_1 \mathcal{E}_1^*\right) + \chi^{(2)}(0, \omega_2, -\omega_2) \left(\frac{1}{2} \mathcal{E}_2 \mathcal{E}_2^*\right)\end{aligned}$$

- To find $\chi^{(3)}$ for this case, plug in $\chi^{(2)}$ into the χ^3 equation. \mathcal{E}^3 will need to be found, quickly complicating the math...

Centrosymmetric

: For this case, use as the restoring force $F_{res} = -m\omega_0^2 x + mFx^3$

The corresponding potential is

$$U = - \int F_{res} dx = \frac{1}{2} m \omega_0^2 x^2 - \frac{1}{4} m F x^4$$

Notice that this is an even function: the potential is symmetric under $x \rightarrow -x$. So this corresponds to systems with inversion symmetry, such as isotropic / centrosymmetric media

- Equation of motion:

$$\ddot{\vec{r}} + \frac{1}{\tau} \dot{\vec{r}} + \omega_0^2 \vec{r} - F(\vec{r} \cdot \vec{r}) \vec{r} = \frac{q\vec{\mathcal{E}}}{m}$$

with

$$\vec{\mathcal{E}} = \vec{\mathcal{E}}_1 e^{-i\omega_1 t} + \vec{\mathcal{E}}_2 e^{-i\omega_2 t} + \vec{\mathcal{E}}_3 e^{-i\omega_3 t} = \sum_n \vec{\mathcal{E}}(\omega_n) e^{-i\omega_n t}$$

Again, use perturbation theory: $\vec{r} = \lambda \vec{r}^{(1)} + \lambda^2 \vec{r}^{(2)} + \lambda^3 \vec{r}^{(3)} + \dots$

$$\lambda: \ddot{\vec{r}}^{(1)} + \frac{1}{\tau} \dot{\vec{r}}^{(1)} + \omega_0^2 \vec{r}^{(1)} = \frac{q\vec{\mathcal{E}}}{m}$$

$$\lambda^2: \ddot{\vec{r}}^{(2)} + \frac{1}{\tau} \dot{\vec{r}}^{(2)} + \omega_0^2 \vec{r}^{(2)} = 0$$

$$\lambda^3: \ddot{\vec{r}}^{(3)} + \frac{1}{\tau} \dot{\vec{r}}^{(3)} + \omega_0^2 \vec{r}^{(3)} - F(\vec{r}^{(1)} \cdot \vec{r}^{(1)}) \vec{r}^{(1)} = 0$$

- As before, the solution to the first equation is the linear response:

$$\vec{r}^{(1)} = \sum_n \frac{q\vec{\mathcal{E}}(\omega_n)/m}{\omega(\omega_n)} e^{-i\omega_n t}$$

The linear susceptibility is given by:

$$\vec{p}^{(1)}(\omega_n) = Nq\vec{r}^{(1)}(\omega_n)$$

tensor

$$P_i^{(1)}(\omega_n) = \epsilon_0 \sum_j \chi_{ij}^{(1)}(\omega_n) \mathcal{E}_j(\omega_n) = \epsilon_0 \sum_j \chi^{(1)}(\omega_n) \delta_{ij} \mathcal{E}_j(\omega_n)$$

isotropic medium

$$\Rightarrow \epsilon_0 \chi^{(1)}(\omega_n) \vec{\mathcal{E}}(\omega_n) e^{-i\omega_n t} = Nq \frac{q\vec{\mathcal{E}}(\omega_n)}{m\omega(\omega_n)} e^{-i\omega_n t}$$

$$\chi^{(1)}(\omega_n) = \frac{Nq^2}{m\epsilon_0} \frac{1}{\omega(\omega_n)} \quad \text{Linear susceptibility for centrosymmetric crystal}$$

Tensor form: $\chi^{(1)}(\omega_n) = \frac{Nq^2}{m\epsilon_0} \frac{1}{\omega(\omega_n)} \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}$

- Consider the λ^2 equation:

$$\ddot{\vec{r}}^{(2)} + \frac{1}{\tau} \dot{\vec{r}}^{(2)} + \omega_0^2 \vec{r}^{(2)} = 0$$

This oscillator is damped but not driven!

Therefore $\vec{r}^{(2)} = 0$ in the steady state: $\chi^{(2)} = 0$

for an centrosymmetric systems

• Tensor form: $P_k = \epsilon_0 \sum_{ij} \chi_{kij}^{(2)} E_i E_j$ (indices are directions)

In materials with inversion symmetry, flipping all the directions has no effect:

$$P_k = \sum_{ij} \chi_{kij}^{(2)} E_i E_j$$

$$-P_k = \sum_{ij} \chi_{kij}^{(2)} (-E_i)(-E_j) = \sum_{ij} \chi_{kij}^{(2)} E_i E_j$$

This is consistent only if $\chi_{kij}^{(2)} = 0$

- Now consider the λ^3 equation, substituting $\vec{r}^{(1)}$

$$\ddot{\vec{r}}^{(3)} + \frac{1}{\tau} \dot{\vec{r}}^{(3)} + \omega_0^2 \vec{r}^{(3)} = F(\vec{r}^{(1)}, \vec{r}^{(1)}) \vec{r}^{(1)}$$

harmonic \downarrow

$$\vec{r}^{(3)}(\omega_g) \delta(\omega_g) = F \sum_{mnp} \frac{q^3}{m^3} (\vec{E}(\omega_m) \cdot \vec{E}(\omega_n)) \vec{E}(\omega_p) \frac{e^{-i(\omega_m + \omega_n + \omega_p)t}}{\delta(\omega_m) \delta(\omega_n) \delta(\omega_p)}$$

$$\vec{r}^{(3)}(\omega_g) = F \sum_{mnp} \frac{q^3}{m^3} \frac{(\vec{E}(\omega_m) \cdot \vec{E}(\omega_n)) \vec{E}(\omega_p)}{\delta(\omega_m) \delta(\omega_n) \delta(\omega_p) \delta(\omega_g)} e^{-i(\omega_m + \omega_n + \omega_p)t}$$

Write the polarization as $\overset{\text{frequencies of driving fields}}{\chi^{(3)}}$

$$P_i^{(3)}(\omega_g) = \epsilon_0 \sum_{jkl} \sum_{mnp} \overset{\text{frequencies of driving fields}}{\chi_{ijkl}^{(3)}}(\omega_g, \omega_m, \omega_n, \omega_p) \overset{\text{components of driving fields}}{E_j(\omega_m) E_k(\omega_n) E_l(\omega_p)}$$

\nwarrow component of polarization \nearrow components of driving fields

Using $P^{(3)} = \epsilon_0 \chi^{(3)} = Nq^4 \vec{r}^{(3)}$, we have

$$\chi_{ijkl}^{(3)}(\omega_g, \omega_m, \omega_n, \omega_p) = F \frac{Nq^4}{\epsilon_0 m^3} \frac{\delta_{jk} \delta_{il}}{\delta(\omega_m) \delta(\omega_n) \delta(\omega_p) \delta(\omega_g)}$$

- In this expression, the deltas originate from:

• δ_{il} : the polarization component (i) must match the vector component of $\vec{r}^{(3)}$ (l)

• δ_{jk} : the dot product $\vec{E}(\omega_m) \cdot \vec{E}(\omega_n) = \sum_{jkl} E_j(\omega_m) E_k(\omega_n) \delta_{jk}$ this arises from the isotropic nature of the crystal, assumed in the original expression for the restoring force

- Return to the polarization expression:

$$P_i^{(3)}(\omega_g) = \epsilon_0 \sum_{jkl} \sum_{mnp} \frac{FN_0^4}{m^3 \epsilon_0} \frac{E_j(\omega_m) E_k(\omega_n) E_l(\omega_p)}{\omega(\omega_m) \omega(\omega_n) \omega(\omega_p) \omega(\omega_g)} (\delta_{jk} \delta_{il})$$

Several terms in the sum will be identical:

e.g.

$$\begin{aligned} \textcircled{1} E_j(\omega_m) E_k(\omega_n) E_l(\omega_p) \delta_{jk} \delta_{il} &= \textcircled{2} E_l(\omega_p) E_k(\omega_n) E_j(\omega_m) \delta_{lk} \delta_{ij} \\ &\xrightarrow{j \leftrightarrow l, m \leftrightarrow p} \\ &= \textcircled{3} E_j(\omega_m) E_l(\omega_p) E_k(\omega_n) \delta_{jl} \delta_{ik} \\ &\xrightarrow{n \leftrightarrow p, k \leftrightarrow l} \end{aligned}$$

Each of these combinations will appear twice in the sum

To display this symmetry explicitly, write each $\chi_{ijkl}(\omega_m, \omega_n, \omega_p, \omega_g)$ component as $1/6$ the total sum:

$$\chi_{ijkl}^{(3)}(\omega_g, \omega_m, \omega_n, \omega_p) = \frac{FN_0^4}{3m^3 \epsilon_0} \frac{\delta_{jk} \delta_{il} + \delta_{lk} \delta_{ij} + \delta_{jl} \delta_{ik}}{\omega(\omega_m) \omega(\omega_n) \omega(\omega_p) \omega(\omega_g)}$$

Rewrite this in terms of the linear susceptibilities: $\chi^{(1)}(\omega_m) = \frac{N_0^2}{m \epsilon_0 \omega(\omega_m)}$

$$\Rightarrow \chi_{ijkl}^{(3)}(\omega_g, \omega_m, \omega_n, \omega_p) = \frac{FN_0^4}{3m^3 \epsilon_0} \left(\frac{N_0^2}{m \epsilon_0} \right)^{-4} \chi^{(1)}(\omega_m) \chi^{(1)}(\omega_n) \chi^{(1)}(\omega_p) \chi^{(1)}(\omega_g) \cdot (\delta_{jk} \delta_{il} + \delta_{lk} \delta_{ij} + \delta_{jl} \delta_{ik})$$

Final result is:

$$\chi_{ijkl}^{(3)}(\omega_g, \omega_m, \omega_n, \omega_p) = \frac{FN_0^3}{3N^3 \epsilon_0^4} \chi^{(1)}(\omega_m) \chi^{(1)}(\omega_n) \chi^{(1)}(\omega_p) \chi^{(1)}(\omega_g) (\delta_{jk} \delta_{il} + \delta_{lk} \delta_{ij} + \delta_{jl} \delta_{ik})$$

This defines the third-order susceptibility tensor

IF crystal is centrosymmetric

So far, we have invoked the ^① symmetry of an isotropic material, and ^② intrinsic permutation symmetry, which is merely the statement that the multiplication order of fields in $E_j(\omega_m) E_k(\omega_n) E_l(\omega_p)$ does not matter

• In centro-symmetric crystals, the lowest-order nonlinearity is $\chi^{(3)}$, since $\chi^{(2)}$ does not exist

- Also valid for all liquids, which are isotropic

• Tensor forms $\checkmark 3 \times 3^2$

- $\chi^{(2)}$ has 27 elements in its general form, and for second harmonic generation this is reduced to 18 (since $E_x(\omega) E_y(\omega) = E_y(\omega) E_x(\omega)$, etc.)

• Cubic crystals are isotropic but may not have inversion symmetry

- Kleinman's Symmetry: works under the assumption that away from any resonances, the material is non-dispersive

• Normally, the indices of χ cannot be permuted without also permuting the frequencies

$$\chi_{ijk}^{(2)}(\omega_3, \omega_1, \omega_2) = \chi_{ikj}^{(2)}(\omega_3, \omega_2, \omega_1) = \chi_{jik}^{(2)}(\omega_1, \omega_3, \omega_2)$$

intrinsic permutation symmetry

$$\downarrow \text{Kleinman's symmetry}$$

$$\chi_{ijk}^{(2)}(\omega_3, \omega_1, \omega_2) = \chi_{ikj}^{(2)}(\omega_3, \omega_1, \omega_2) = \chi_{jik}^{(2)}(\omega_3, \omega_2, \omega_1)$$

- If this holds, the elements of $\chi^{(2)}$ which share a permutation of indices will be symmetric, e.g. $\chi_{xyz}^{(2)} = \chi_{zxy}^{(2)} = \chi_{zyx}^{(2)}$, etc.
- This reduces the number of independent elements in $\chi^{(2)}$ from 18 to 10! (for the SHG case of $\omega_1 = \omega_2$)

- $\chi^{(3)}$ has 81 possible elements including intrinsic permutation symmetry. This comes from:

$$\# \text{ elements} = 3 \times 3^3 = 81 \text{ elements}$$

components in \mathbb{P}

permutations of field components in \mathbb{E}^3

Reductions in the number of independent elements in a cubic crystal:

- 1) Diagonal elements are equal due to isotropy: $\chi_{xxxx}^{(3)} = \chi_{yyyy}^{(3)} = \chi_{zzzz}^{(3)}$
- 2) Terms with an odd number of subscripts in any one direction are zero due to inversion symmetry:

$$\begin{aligned} \text{e.g. } P_x &= \chi_{xxyy}^{(3)} E_x(\omega_m) E_x(\omega_n) E_y(\omega_p) \\ P_x &= \chi_{xxyy}^{(3)} E_x(\omega_m) E_x(\omega_n) (-E_y(\omega_p)) \end{aligned} \quad \begin{array}{l} \downarrow y \rightarrow -y \\ \Rightarrow \chi_{xxyy} = 0 \end{array}$$

This also means that any element of $\chi^{(3)}$ with three unique directions is zero.

- 3) Isotropy further allows any pair of the same indices to be swapped for a pair with a different index, since the coordinate axes are equivalent

$$\begin{aligned} \text{e.g. } \chi_{xxyy} &= \chi_{xxzz} = \chi_{yyxx} = \dots \\ \chi_{xyxy} &= \chi_{xzyz} = \chi_{yzzy} = \dots \end{aligned}$$

With these three conditions, there are only 4 independent terms in $\chi^{(3)}$

$$\chi_{xxxx}^{(3)}, \chi_{xxyy}^{(3)}, \chi_{xyxy}^{(3)}, \chi_{xyyx}^{(3)}$$

Use the result for $\chi^{(3)}$ derived for a centrosymmetric crystal:

$$\begin{aligned} \chi_{xxyy}^{(3)}(\omega_g, \omega_m, \omega_n, \omega_p) &= \frac{Fm\epsilon_0^3}{3N^3g^4} \chi^{(1)}(\omega_m) \chi^{(1)}(\omega_n) \chi^{(1)}(\omega_p) \chi^{(1)}(\omega_g) \\ &= \chi_{xyxy}^{(3)}(\omega_g, \omega_n, \omega_m, \omega_p) \\ &= \chi_{xyyx}^{(3)}(\omega_g, \omega_p, \omega_n, \omega_m) \end{aligned}$$

This leads to 2 additional facts of interest

1) For $\chi_{xxxx}^{(3)}$, $\delta_{jk}\delta_{ie} + \delta_{ek}\delta_{ij} + \delta_{je}\delta_{ik} = 3$ and the order of the frequencies does not matter. Therefore it will always be true that:

$$\chi_{xxxx}^{(3)} = \chi_{xxyy}^{(3)} + \chi_{xyxy}^{(3)} + \chi_{xyyx}^{(3)}$$

So for a centrosymmetric medium there are only 3 independent elements in $\chi^{(3)}$.

2) If Kleinman's symmetry holds, the frequencies can be swapped freely, so

$$\chi_{xxyy}^{(3)} = \chi_{xyxy}^{(3)} = \chi_{xyyx}^{(3)} = \frac{1}{3} \chi_{xxxx}^{(3)}$$

↑
from 1)

So in this case there is only a single independent element!

This condition will also automatically hold for third-harmonic generation ($\omega = \omega_m = \omega_n = \omega_p$) without requiring non-dispersion.

- Third-order nonlinear effects:

1) Third-harmonic generation: $\chi^{(3)}(-3\omega, \omega, \omega, \omega)$

2) Intensity-dependent index change: $\chi^{(3)}(-\omega, \omega, -\omega, \omega)$

$$\begin{aligned} \rho &= \epsilon_0 \chi^{(1)} \mathcal{E}(\omega) + \epsilon_0 \chi^{(3)}(-\omega, \omega, -\omega, \omega) \mathcal{E}^3(\omega) \\ &= \epsilon_0 \left[\chi^{(1)}(\omega) + \underbrace{\chi^{(3)}(-\omega, \omega, -\omega, \omega)}_{\text{intensity-dependent change in } n} |\mathcal{E}(\omega)|^2 \right] \mathcal{E}(\omega) \end{aligned}$$

3) Refractive index change caused by a second frequency: $\chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \omega_2)$

$$\Rightarrow \rho = \epsilon_0 \left[\chi^{(1)}(\omega) + \underbrace{\chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \omega_2)}_{\text{pump}} |\mathcal{E}(\omega_2)|^2 \right] \mathcal{E}(\omega_1)_{\text{probe}}$$

4) Raman gain caused by a second frequency: $\text{Im} [\chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \omega_2)]$

Pump at ω_2 : intensity $|\mathcal{E}(\omega_2)|^2$ is proportional to the gain in $\mathcal{E}(\omega_1)$. The gain is $\propto [\text{Im} \chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \omega_2)] \cdot |\mathcal{E}(\omega_2)|^2$

5) Four-wave mixing: $\chi^{(3)}(-(2\omega_1 - \omega_2), \omega_1, \omega_1, -\omega_2)$

Various combinations are possible, generating many new frequencies

6) Two-photon absorption: $\text{Im} [\chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \omega_2)]$

↳ The absorption is $\propto \text{Im} [\chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \omega_2)] \mathcal{E}(\omega_1) \mathcal{E}^*(\omega_2)$

7) Gain or absorption saturation: $\text{Im} [\chi^{(3)}(-\omega, \omega, -\omega, \omega)]$

Absorption coefficient $\propto \text{Im} [\chi^{(3)}(-\omega, \omega, -\omega, \omega)] |\mathcal{E}(\omega)|^2$

- Quantum mechanically, a high intensity beam can deplete the electronic population in the upper or lower states, leading to saturated gain or absorption
- If $\text{Im} \chi > 0 \Rightarrow$ absorption
 $\text{Im} \chi < 0 \Rightarrow$ gain

④ Wave Equation for Nonlinear Optics

- Write the polarization as: $\vec{P} = \epsilon_0 \chi^{(1)} \vec{E} + \vec{P}_{NL}$
 where we consider 2nd order nonlinearity $P_{NL} = \epsilon_0 \chi_{ijk}^{(2)} E_j E_k$
 (Einstein notation)

- Using Maxwell's equations with $\vec{J} = 0, \rho = 0$, we have

$$\vec{\nabla} \times \vec{H} = \frac{\partial}{\partial t} (\epsilon \vec{E}) + \frac{\partial}{\partial t} \vec{P}_{NL}$$

$$\vec{\nabla} \times \vec{E} = - \frac{\partial}{\partial t} (\mu_0 \vec{H})$$

Take curl of Faraday's law: $\vec{\nabla} \times (\vec{\nabla} \times \vec{E}) = \vec{\nabla} (\vec{\nabla} \cdot \vec{E}) - \nabla^2 \vec{E}$

$$\begin{aligned} \Rightarrow \nabla^2 \vec{E} &= - \vec{\nabla} \times \left(- \frac{\partial}{\partial t} (\mu_0 \vec{H}) \right) \\ &= \frac{\partial}{\partial t} (\mu_0 \vec{\nabla} \times \vec{H}) = \mu_0 \epsilon \frac{\partial^2 \vec{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \vec{P}_{NL}}{\partial t^2} \end{aligned}$$

$$\text{So: } \nabla^2 \vec{E} - \mu_0 \epsilon \frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}_{NL}}{\partial t^2}$$

This is the nonlinear wave equation

- Consider three waves $\omega_1, \omega_2, \omega_3$ propagating in the z direction

$$E_{1i} = \text{Re} (E_{1i}(z) e^{ik_1 z - i\omega_1 t}) \quad i, j, k: \text{ x or y polarization}$$

$$E_{2j} = \text{Re} (E_{2j}(z) e^{ik_2 z - i\omega_2 t})$$

$$E_{3k} = \text{Re} (E_{3k}(z) e^{ik_3 z - i\omega_3 t})$$

↑ slowly varying amplitudes

- Consider E_1 to be the wave, then the nonlinear polarization is

$$P_{NL}(\omega_1) = \epsilon_0 \chi_{ijk} E_{3j}(z) E_{2k}^*(z) e^{i(k_3 - k_2)z - i(\omega_3 - \omega_2)t}$$

ω_1

- Meanwhile, using the slowly varying amplitude approximation,

$$\begin{aligned} \nabla^2 E_{1i}(z, t) &= \left[-k_1^2 E_{1i}(z) + 2ik_1 \frac{dE_{1i}(z)}{dz} + \frac{d^2 E_{1i}(z)}{dz^2} \right] e^{ik_1 z - i\omega_1 t} \\ \frac{\partial^2}{\partial t^2} E_{1i}(z, t) &= -\omega_1^2 E_{1i}(z) e^{ik_1 z - i\omega_1 t} \end{aligned}$$

- The wave equation becomes:

$$\left[(-k_1^2 + \cancel{\mu_0 \epsilon \omega_1^2}) E_{1i}(z) + 2ik_1 \frac{dE_{1i}(z)}{dz} \right] e^{i(k_1 z - \omega_1 t)} = -\epsilon_0 \mu_0 \omega_1^2 \chi_{ijk} E_{3j}(z) E_{2k}^*(z) e^{i(k_3 - k_2)z - i(\omega_3 - \omega_2)t}$$

↑ The linear wave equation requires this

* The same susceptibility $\chi^{(2)}$ is used throughout these expressions by Kleinman's symmetry

- Dropping the time dependence, this becomes:

$$2ik_1 \frac{d\mathcal{E}_{1i}(z)}{dz} e^{ik_1 z} = -\epsilon_0 \mu_0 \omega_1^2 \chi_{ijk}^{(2)} \mathcal{E}_{3j}(z) \mathcal{E}_{2k}^*(z) e^{i(k_3 - k_2)z}$$

$$\frac{d\mathcal{E}_{1i}(z)}{dz} = \frac{i\epsilon_0 \mu_0 \omega_1^2}{2k_1} \chi_{ijk}^{(2)} \mathcal{E}_{3j}(z) \mathcal{E}_{2k}^*(z) e^{i(k_3 - k_2 - k_1)z}$$

$$\textcircled{1} \frac{d\mathcal{E}_{1i}(z)}{dz} = \frac{i}{\sqrt{\epsilon_1}} \frac{\omega_1}{2c} \chi_{ijk}^{(2)} \mathcal{E}_{3j}(z) \mathcal{E}_{2k}^*(z) e^{i \underbrace{(k_3 - k_2 - k_1)}_{\Delta K} z}$$

- We could also have treated \mathcal{E}_2 or \mathcal{E}_3 as the propagating wave, which results in:

$$\textcircled{2} \frac{d\mathcal{E}_{2k}(z)}{dz} = \frac{i}{\sqrt{\epsilon_2}} \frac{\omega_2}{2c} \chi_{ijk}^{(2)} \mathcal{E}_{1i}^*(z) \mathcal{E}_{3j}(z) e^{i(k_3 - k_2 - k_1)z}$$

$$\textcircled{3} \frac{d\mathcal{E}_{3j}(z)}{dz} = \frac{i}{\sqrt{\epsilon_3}} \frac{\omega_3}{2c} \chi_{ijk}^{(2)} \mathcal{E}_{1i}(z) \mathcal{E}_{2k}(z) e^{-i(k_3 - k_2 - k_1)z}$$

- Notice that all three expressions share $\Delta K = k_3 - k_2 - k_1$.

$\Delta K \neq 0$ even though $\Delta\omega = 0$, because $n = n(\omega)$

- Consider the case of second harmonic generation: $\omega_3 = 2\omega$, $\omega_1 = \omega_2 = \omega$

Use the last equation:

$$\frac{d\mathcal{E}_{3j}(z)}{dz} = \frac{i}{\sqrt{\epsilon_3}} \frac{\omega_3}{2c} \chi_{ijk}^{(2)} \mathcal{E}_{1i} \mathcal{E}_{2k} e^{-i\Delta K z}$$

We approximate these to be fixed with z : $\chi^{(2)}$ is small enough not to significantly perturb the applied fields, which continue to travel as plane waves

- Integrate along length of crystal.

$$\mathcal{E}_{3j}(z) = \frac{i\omega_3}{2c\sqrt{\epsilon_3}} \chi_{ijk}^{(2)} \mathcal{E}_{1i} \mathcal{E}_{2k} \int_0^L dz e^{-i\Delta K \cdot z}$$

$$= \frac{i\omega_3}{2c\sqrt{\epsilon_3}} \chi_{ijk}^{(2)} \mathcal{E}_{1i} \mathcal{E}_{2k} \frac{e^{-i\Delta K \cdot L} - 1}{-i\Delta K}$$

$$|\mathcal{E}_{3j}(z)|^2 = \frac{\omega_3^2}{4c^2\epsilon_3} |\mathcal{E}_{1i}|^2 |\mathcal{E}_{2k}|^2 |\chi^{(2)}|^2 \left(\frac{e^{-i\Delta K \cdot L} - 1}{-i\Delta K} \right) \left(\frac{e^{i\Delta K \cdot L} - 1}{i\Delta K} \right)$$

$$= \frac{\omega_3^2}{4c^2\epsilon_3} |\mathcal{E}_{1i}|^2 |\mathcal{E}_{2k}|^2 |\chi^{(2)}|^2 \left(\frac{1 - \frac{1}{2}(e^{i\Delta K \cdot L} + e^{-i\Delta K \cdot L})}{\Delta K^2/2} \right)$$

$$|\mathcal{E}_{3j}(z)|^2 = \frac{\omega_3^2}{4c^2\epsilon_3} |\mathcal{E}_{1i}|^2 |\mathcal{E}_{2k}|^2 |\chi^{(2)}|^2 \frac{\sin^2(\Delta K \cdot L/2)}{(\Delta K/2)^2}$$

- The relative strength of the sum wave is:

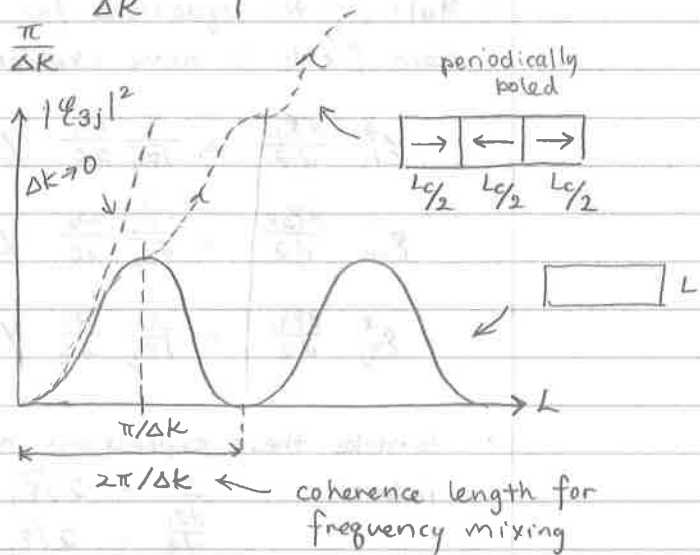
$$\left| \frac{\mathcal{E}_{3j}(z)}{\mathcal{E}_{1i}} \right|^2 = \left| \frac{\omega_3}{c\sqrt{\epsilon_3}} \cdot \chi^{(2)} \cdot \frac{\sin(\Delta k \cdot L/2)}{\Delta k} \right|^2$$

This is maximized when $L = \frac{\pi}{\Delta k}$

$$\Rightarrow \left| \frac{\mathcal{E}_{3j}}{\mathcal{E}_{1i}} \right|^2 = \left| \frac{\omega_3}{c\sqrt{\epsilon_3}} \frac{\chi^{(2)}}{\Delta k} \right|^2$$

- The efficiency of the nonlinear process is increased by having $\Delta k \rightarrow 0$. This condition is called phase matching

\Rightarrow Also increases the coherence length $L_c = \frac{2\pi}{\Delta k}$ of crystal



- When Δk is small, the wave and the fields driving the polarization are in phase over a long distance. For second harmonic generation, the coherence length is:

$$L_c = 2\pi / (k(2\omega) - 2k(\omega)) = 2\pi / \left[\frac{2\omega}{c} (n(2\omega) - n(\omega)) \right]$$

Typically $n(2\omega) - n(\omega) \approx 0.01$, so $L_c \approx 100 \mu\text{m}$

- One method to achieve better phase matching is to use an anisotropic crystal which has the property $n_e(\omega) = n_o(2\omega)$ for some incident direction
 - However, this response is very sensitive to angle and is not that robust
- Alternatively, the efficiency can be increased by stacking crystals along z , each with size $L_c/2$ and having alternating crystal orientations
 - This allows the sum wave's power to increase continually along the chain of crystals (shown above)
 - Poled LiNbO_3 can be made to have this property: apply a field across each section of the crystal to decide its orientation (requires high temperature to set crystal)
 - This is called "quasi-phase-matching"

Manley-Rowe relations

- Multiply the equations for $\frac{d\mathcal{E}_{1i}}{dz}$, $\frac{d\mathcal{E}_{2k}}{dz}$, $\frac{d\mathcal{E}_{3j}}{dz}$ by their respective wave fields to more explicitly display their symmetry

$$\mathcal{E}_{1i}^* \frac{d\mathcal{E}_{1i}}{dz} = \frac{i}{\sqrt{\epsilon_1}} \frac{\omega_1}{2c} \chi_{ijk}^{(2)} \mathcal{E}_{3j} \mathcal{E}_{2k}^* \mathcal{E}_{1i}^* e^{i\Delta k \cdot z}$$

$$\mathcal{E}_{2k}^* \frac{d\mathcal{E}_{2k}}{dz} = \frac{i}{\sqrt{\epsilon_2}} \frac{\omega_2}{2c} \chi_{ijk}^{(2)} \mathcal{E}_{3j} \mathcal{E}_{2k}^* \mathcal{E}_{1i}^* e^{i\Delta k \cdot z}$$

$$\mathcal{E}_{3j}^* \frac{d\mathcal{E}_{3j}}{dz} = \frac{i}{\sqrt{\epsilon_3}} \frac{\omega_3}{2c} \chi_{ijk}^{(2)} \mathcal{E}_{3j}^* \mathcal{E}_{2k} \mathcal{E}_{1i} e^{-i\Delta k \cdot z}$$

$\chi^{(2)}$ equal by Kleinman's symmetry (no dispersion)

- To make these expressions more intuitive, express in terms of intensity:

$$I_i = 2\sqrt{\epsilon_i} \epsilon_0 c \mathcal{E}_i \mathcal{E}_i^* \\ \frac{dI_i}{dz} = 2\sqrt{\epsilon_i} \epsilon_0 c \left[\mathcal{E}_i^* \frac{d\mathcal{E}_i}{dz} + \mathcal{E}_i \frac{d\mathcal{E}_i^*}{dz} \right] \\ = 2\sqrt{\epsilon_i} \epsilon_0 c \left[\mathcal{E}_i^* \frac{d\mathcal{E}_i}{dz} + (\mathcal{E}_i^* \frac{d\mathcal{E}_i}{dz})^* \right]$$

$$\text{e.g. } \frac{dI_1}{dz} = \frac{2\sqrt{\epsilon_1} \epsilon_0 c \omega_1}{\sqrt{\epsilon_1} \cdot 2c} \chi^{(2)} \left[i \mathcal{E}_{3j} \mathcal{E}_{2k}^* \mathcal{E}_{1i}^* e^{i\Delta k \cdot z} - i \mathcal{E}_{3j}^* \mathcal{E}_{2k} \mathcal{E}_{1i} e^{-i\Delta k \cdot z} \right] \\ = \epsilon_0 \omega_1 \chi^{(2)} i \left[\mathcal{E}_{3j} \mathcal{E}_{2k}^* \mathcal{E}_{1i}^* e^{i\Delta k \cdot z} - (\mathcal{E}_{3j} \mathcal{E}_{2k}^* \mathcal{E}_{1i}^* e^{i\Delta k \cdot z})^* \right]$$

$$\Rightarrow \frac{dI_1}{dz} = -2\epsilon_0 \omega_1 \chi^{(2)} \text{Im} \left[\mathcal{E}_{3j} \mathcal{E}_{2k}^* \mathcal{E}_{1i}^* e^{i\Delta k \cdot z} \right]$$

$$\frac{dI_2}{dz} = -2\epsilon_0 \omega_2 \chi^{(2)} \text{Im} \left[\mathcal{E}_{3j} \mathcal{E}_{2k}^* \mathcal{E}_{1i}^* e^{i\Delta k \cdot z} \right]$$

$$\frac{dI_3}{dz} = +2\epsilon_0 \omega_3 \chi^{(2)} \text{Im} \left[\mathcal{E}_{3j} \mathcal{E}_{2k}^* \mathcal{E}_{1i}^* e^{i\Delta k \cdot z} \right]$$

$\text{Im} A^* = -\text{Im} A$

- From this we see that the spatial variation of total intensity is:

$$\frac{dI}{dz} = \frac{dI_1}{dz} + \frac{dI_2}{dz} + \frac{dI_3}{dz}$$

$$= -2\epsilon_0 \chi^{(2)} \text{Im} \left[\mathcal{E}_{3j} \mathcal{E}_{2k}^* \mathcal{E}_{1i}^* e^{i\Delta k \cdot z} \right] (\omega_1 + \omega_2 - \omega_3) = 0$$

- The equation also imply a conserved quantity:

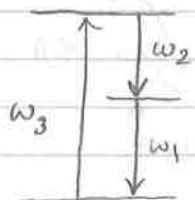
$$\frac{1}{\omega_1} \frac{dI_1}{dz} = \frac{1}{\omega_2} \frac{dI_2}{dz} = -\frac{1}{\omega_3} \frac{dI_3}{dz}$$

These are the Manley-Rowe relations.

$$\frac{I}{\omega} = \frac{\text{photon number}}{\text{area} \cdot \text{time}}$$

- Slightly rewrite the relations:

$$\frac{d}{dz} \left(\frac{I_1}{\omega_1} + \frac{I_3}{\omega_3} \right) = \frac{d}{dz} \left(\frac{I_2}{\omega_2} + \frac{I_3}{\omega_3} \right) = \frac{d}{dz} \left(\frac{I_1}{\omega_1} - \frac{I_2}{\omega_2} \right) = 0$$



This states that the rate at which ω_1 (or ω_2) is created is equal to the rate at which ω_3 is created.

⇒ conservation of energy

• the change in photon number is the same at all 3 frequencies

Absorption

• First, what is the effect on index of a complex ϵ_r ?

$$n^2 = (n' + in'')^2 = \epsilon' + i\epsilon''$$

$$n'^2 + 2in'n'' - n''^2 = \epsilon' + i\epsilon''$$

$$\Rightarrow \epsilon' = n'^2 - n''^2$$

$$\epsilon'' = 2n'n''$$

Complex ϵ affects both parts of n

• Consider a wave with complex index:

$$E(z) = E_0 e^{i \frac{n'\omega}{c} z - \frac{n''\omega}{c} z} \Rightarrow I(z) = |E_0|^2 e^{-\frac{2n''\omega}{c} z}$$

If $n'' > 0$, the wave attenuates

• Contrary to what Eli claims in lecture, the attenuation is exactly the same as absorption!!

$$\alpha = \frac{\omega}{n'c} \chi''(\omega) = \frac{\omega}{n'c} (2n'n'')$$

$$\text{absorption } \alpha = \frac{2\omega n''}{c} = \text{attenuation}$$

* This equivalence is made by Yaniv!

The only case where attenuation does not correspond to absorption is in total internal reflection, where the decay constant of the evanescent wave comes from a complex angle (from Snell's law) rather than from n''

- Absorption and nonlinear susceptibility

• Consider:

$$P = \epsilon_0 (\chi^{(1)} + \chi^{(3)} |\mathcal{E}|^2) \mathcal{E}$$

$$\text{Re}(\chi^{(3)} |\mathcal{E}|^2)$$

Intensity-dependent refractive index

$$\text{Im}(\chi^{(3)} |\mathcal{E}|^2)$$

Intensity-dependent absorption

> 0 : absorption

< 0 : gain

• Two-photon absorption:

$$\alpha(\omega_2) = \frac{\omega_1}{n(\omega_1) \cdot c} \text{Im}[\chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \omega_2)] |\mathcal{E}(\omega_2)|^2$$

• Through $\chi^{(3)}$, the $\hbar\omega_2$ photons modulate the absorption of $\hbar\omega_1$ photons

(brief discussion)

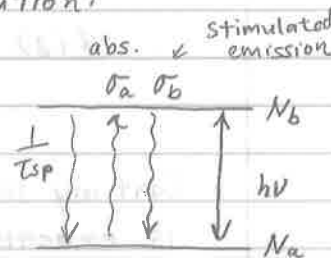
- Detailed balance: in thermal equilibrium, absorption of light is always balanced by spontaneous emission

See §6 for more rigorous treatment

• Introduce B = brightness: # photons/area/Hz/s/4π str

This is given by the Planck blackbody distribution:

$$B(\nu, T) = \frac{8\pi n^2 \nu^2}{c^2} \frac{1}{e^{h\nu/kT} - 1}$$



• Consider a two-level system with populations N_a, N_b and energy difference $h\nu$.

• Detailed balance requires (over a frequency slice $\Delta\nu$)

$$\underbrace{\frac{8\pi n^2 \nu^2}{c^2} \Delta\nu}_{\text{electromagnetic energy density}} \underbrace{\left(N_a \sigma_a - N_b \sigma_b \right)}_{\substack{\uparrow \\ \text{absorption}} \quad \substack{\uparrow \\ \text{stimulated emission}}} = \underbrace{\frac{N_b}{T_{sp}}}_{\substack{\uparrow \\ \text{spontaneous emission}}}$$

$$\frac{8\pi n^2 \nu^2 \Delta\nu}{c^2} \sigma_a \left(1 - \frac{N_b \sigma_b}{N_a \sigma_a} \right) = \frac{N_b}{N_a} \frac{1}{T_{sp}} (e^{h\nu/kT} - 1)$$

In equilibrium, $\frac{N_b}{N_a} = e^{-h\nu/kT}$ by Boltzmann statistics

$$\frac{8\pi n^2 \nu^2 \Delta\nu}{c^2} \sigma_a \left(1 - \frac{\sigma_b}{\sigma_a} e^{-h\nu/kT} \right) = \frac{1}{T_{sp}} (1 - e^{-h\nu/kT})$$

* This is only possible to satisfy when $\sigma_a = \sigma_b \neq 0$ otherwise, detailed balance cannot hold.

$$\Rightarrow \frac{8\pi n^2 \nu^2 \Delta \nu}{c^2} \sigma_a (1 - e^{-h\nu/kT}) = \frac{1}{T_{sp}} (1 - e^{-h\nu/kT})$$

absorption coefficient $\alpha = \sigma_a (N_a - N_b) = \frac{N_a}{T_{sp}} \frac{c^2}{8\pi n^2 \nu^2 \Delta \nu} \frac{(N_a - N_b)}{N_a}$

$$\Rightarrow \alpha = \frac{c^2}{T_{sp} 8\pi n^2 \nu^2 \Delta \nu} (N_a - N_b)$$

- This expression tells us that the absorption coefficient is:

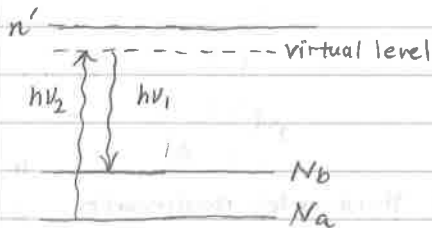
• $\alpha > 0$ if $N_a > N_b$: net absorption

• $\alpha < 0$ if $N_b > N_a$: net gain

↳ this is called population inversion

Raman gain

- Spontaneous Raman effect (weak): emission is not directed ↑ isotropic



• A virtual intermediate level assists the generation of $h\nu_f < h\nu_i$ (Stokes) or $h\nu_f > h\nu_i$ (anti-Stokes)

• Anti-Stokes emission in this way is inefficient because of the relative thermal populations of the initial states N_a and N_b

- Stimulated Raman effect

• This effect arises from the molecular polarizability of molecules in the medium:

$$P = \epsilon_0 N \gamma \cdot \mathcal{E} = \epsilon_0 N \left(\gamma_0 + \frac{\partial \gamma}{\partial x} \Big|_0 x \right) \cdot \mathcal{E}$$

↳ displacement of the e^- from molecule

• To solve for the displacement x , find the restoring force:

- First find the electrical energy, given the electric field consisting of the input beam (ω_2) and Stokes-shifted light (ω_1)

$$\mathcal{E} = \mathcal{E}_1 e^{-i\omega_1 t} + \mathcal{E}_2 e^{-i\omega_2 t} \quad \text{where } \omega_2 > \omega_1$$

$$\Rightarrow |\mathcal{E}|^2 = \mathcal{E}_2^* \mathcal{E}_1 e^{-i(\omega_2 - \omega_1)t} + \mathcal{E}_1 \mathcal{E}_2^* e^{+i(\omega_2 - \omega_1)t}$$

Prop this term for simplicity

- The restoring force is:

$$F = \frac{\partial}{\partial x} \left(\frac{1}{2} \epsilon |\mathcal{E}|^2 \right) = \epsilon_0 \frac{\partial}{\partial x} \left(\frac{1}{2} N \left(\gamma_0 + \frac{\partial \gamma}{\partial x} \Big|_0 x \right) |\mathcal{E}|^2 \right)$$

$$F \approx \frac{\epsilon_0}{2} N \left(\frac{\partial \gamma}{\partial x} \Big|_0 \right) \cdot \mathcal{E}_2^* \mathcal{E}_1 e^{i(\omega_2 - \omega_1)t}$$

force per oscillator

- The equation of motion becomes:

$$\ddot{x} + \frac{\dot{x}}{\tau} + \omega_0^2 x = \frac{\epsilon_0}{2m} \left(\frac{\partial^2 \chi}{\partial x^2} \Big|_0 \right) \epsilon_2^* \epsilon_1 e^{-i(\omega_2 - \omega_1)t}$$

↓ harmonic oscillator

$$x e^{i\omega t} = \frac{\frac{\epsilon_0}{2m} \left(\frac{\partial^2 \chi}{\partial x^2} \Big|_0 \right) \epsilon_2^* \epsilon_1 e^{-i(\omega_2 - \omega_1)t}}{\omega_0^2 - \omega^2 + i\omega/\tau}$$

- If the molecular displacement oscillates at the same frequency as the overall electric field, this means

$$x = \frac{\epsilon_0 \left(\frac{\partial^2 \chi}{\partial x^2} \Big|_0 \right) \epsilon_2^* \epsilon_1}{2m [\omega_0^2 - (\omega_2 - \omega_1)^2 + i(\omega_2 - \omega_1)/\tau]}$$

- The polarization is then:

$$P = \epsilon_0 N \left[\chi_0 + \left(\frac{\partial^2 \chi}{\partial x^2} \Big|_0 \right) x \right] \cdot \mathcal{E}$$

$\chi^{(1)}$ $\chi^{(3)}$ $\chi^{(3)}$ Raman : a third-order nonlinearity

$$\Rightarrow P_{\text{Raman}}^{(3)} = \frac{\epsilon_0 N \left(\frac{\partial^2 \chi}{\partial x^2} \Big|_0 \right)^2 \epsilon_2^* \epsilon_1}{2m [\omega_0^2 - (\omega_2 - \omega_1)^2 + i(\omega_2 - \omega_1)/\tau]} \cdot \mathcal{E}$$

other terms lead to frequency mixing

Various frequency combinations arise when multiplied by \mathcal{E} .

The polarization term with a sum frequency of ω_1 arises:

$$P_{\text{Raman}}^{(3)}(\omega_1) = \frac{\epsilon_0 N \left(\frac{\partial^2 \chi}{\partial x^2} \Big|_0 \right)^2 |\epsilon_2|^2 \epsilon_1}{2m [\omega_0^2 - (\omega_2 - \omega_1)^2 + i(\omega_2 - \omega_1)/\tau]}$$

- The corresponding nonlinear susceptibility is:

$$\chi_{\text{Raman}}^{(3)}(-\omega_1, \omega_1, -\omega_2, \omega_2) = \frac{\left(\frac{\epsilon_0 N}{2m} \right) \left(\frac{\partial^2 \chi}{\partial x^2} \Big|_0 \right)^2}{\omega_0^2 - (\omega_2 - \omega_1)^2 + i(\omega_2 - \omega_1)/\tau}$$

$$\chi_{\text{Raman}}^{(3)} = \frac{\left(\frac{\epsilon_0 N}{2m} \right) \left(\frac{\partial^2 \chi}{\partial x^2} \Big|_0 \right)^2}{(\omega_0 - (\omega_2 - \omega_1))(\omega_0 + (\omega_2 - \omega_1)) + i(\omega_2 - \omega_1)/\tau}$$

Take $\omega_2 - \omega_1 \approx \omega_0$ since we expect the Stokes shift to be roughly equal to the vibrational mode frequency

$$\chi_{\text{Raman}}^{(3)}(\omega_1) = \frac{\left(\frac{\epsilon_0 N}{4m\omega_0}\right) \left(\frac{\partial \chi}{\partial x}\bigg|_0\right)^2}{(\omega_0 - (\omega_2 - \omega_1)) + i(1/2\tau)}$$

Defining $\chi_{\text{Raman}} = \chi'_{\text{Raman}} + i\chi''_{\text{Raman}}$, we have

$$\chi'_{\text{Raman}}(\omega_1) = \frac{\left(\frac{\epsilon_0 N}{4m\omega_0}\right) \left(\frac{\partial \chi}{\partial x}\bigg|_0\right)^2 (\omega_0 - (\omega_2 - \omega_1))}{(\omega_0 - (\omega_2 - \omega_1))^2 + 1/(4\tau^2)}$$

$$\chi''_{\text{Raman}}(\omega_1) = \frac{-\left(\frac{\epsilon_0 N}{4m\omega_0}\right) \left(\frac{\partial \chi}{\partial x}\bigg|_0\right)^2 \frac{1}{2\tau}}{(\omega_0 - (\omega_2 - \omega_1))^2 + 1/(4\tau^2)}$$

May be off by one or more factors of 2

The absorption coefficient is related to χ'' :

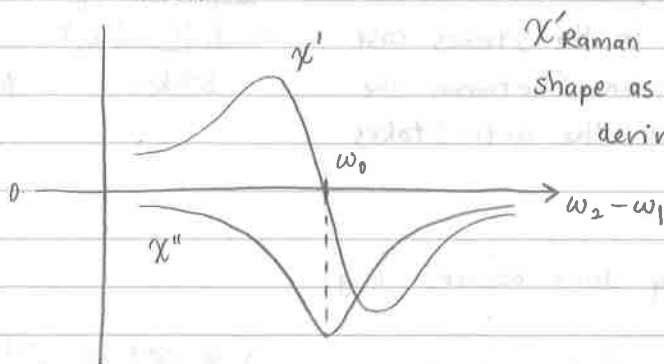
$$\alpha_{\text{Raman}}(\omega_1) = \frac{\omega_1}{n_1 c} \chi''_{\text{Raman}}(\omega_1) \cdot |\mathcal{E}_2|^2$$

Since $\chi'' < 0$, this is actually a gain coefficient!

$$\Rightarrow \boxed{g_{\text{Raman}}(\omega_1) = \frac{\left(\frac{\epsilon_0 N \omega_1}{8n_1 c m \omega_0 \tau}\right) \left(\frac{\partial \chi}{\partial x}\bigg|_0\right)^2 |\mathcal{E}_2|^2}{(\omega_0 - (\omega_2 - \omega_1))^2 + 1/4\tau^2}} \quad \text{Raman gain}$$

$I(z) = I_0 e^{g \cdot z}$

* Stimulated Raman emission: there is gain in the Stokes-shifted frequency as the pump intensity $|\mathcal{E}_2|^2$ increases. Also, since this is a stimulated emission process, the ω_1 light is emitted in a narrow cone.



χ'_{Raman} and χ''_{Raman} have the same overall shape as the $\chi^{(1)}$ and $\chi^{(1)}$ linear susceptibilities derived from the harmonic oscillator model.

- The greatest Stokes scattering occurs when exactly $\omega_2 - \omega_1 = \omega_0$
- This model does not directly predict gain for anti-Stokes scattering

- The Anti-Stokes gain can be predicted with a third frequency $\omega_3 = \omega_2 + \omega_1$, replacing ω_1 in the previous equations with ω_3 . Then we have $\omega_3 - \omega_2 \approx \omega_0$.

$$\chi_{\text{Raman}}^{(3)}(\omega_3) = \frac{\left(\frac{\epsilon_0 N}{2m}\right) \left(\frac{\partial^2 \chi}{\partial x^2}\bigg|_0\right)^2}{(\omega_0 - (\omega_2 - \omega_3))(\omega_0 + (\omega_2 - \omega_3)) + i(\omega_2 - \omega_3)/\tau}$$

$\approx 2\omega_0$ $\approx -\omega_0$

$$= \frac{\left(\frac{\epsilon_0 N}{4m\omega_0}\right) \left(\frac{\partial^2 \chi}{\partial x^2}\bigg|_0\right)^2}{(\omega_0 + (\omega_2 - \omega_3)) - i(1/2\tau)}$$

The imaginary part then becomes:

$$\chi''_{\text{Raman}}(\omega_3) = \frac{\left(\frac{\epsilon_0 N}{8m\omega_0\tau}\right) \left(\frac{\partial^2 \chi}{\partial x^2}\bigg|_0\right)^2}{(\omega_0 + (\omega_2 - \omega_3))^2 + 1/4\tau^2} \quad \text{Anti-stokes}$$

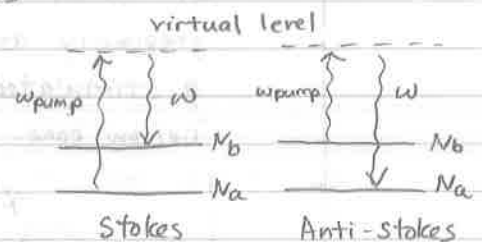
- * Observe that $\chi''_{\text{Raman}}(\omega_3) > 0$: therefore $\alpha_{\text{Raman}}(\omega_3) > 0$ and the anti-stokes emission attenuates in the presence of the lower frequency pump beam.

* Summary: stimulated Raman emission gives

1) Gain at $\omega = \omega_{\text{pump}} - \omega_0$

2) Loss at $\omega = \omega_{\text{pump}} + \omega_0$

- The reason for this is simple: there is population inversion in the Stokes case and no population inversion (between the virtual level and N_a) in the anti-Stokes case.



- However, frequency mixing does occur, e.g.

$$P(2\omega_2 - \omega_1) = \chi_{\text{Raman}}^{(3)}(-2\omega_2 + \omega_1, \omega_2, \omega_2, -\omega_1) \mathcal{E}_2 \mathcal{E}_1^* \mathcal{E}_2 e^{-i(2\omega_2 - \omega_1)t}$$

If $\omega_2 - \omega_1 = \omega_0$, we have:

$$P(\omega_2 + \omega_0) = \chi_{\text{Raman}}^{(3)}(-(\omega_2 + \omega_0), \omega_2, \omega_2, -\omega_1) \mathcal{E}_2 \mathcal{E}_1^* \mathcal{E}_2 e^{-i(\omega_2 + \omega_0)t}$$

↳ This resembles anti-Stokes scattering, but it is really just simple frequency mixing coming from the real part of $\chi^{(3)}$, not the imaginary part.

- Insert the spatial dependence on this sum frequency wave:

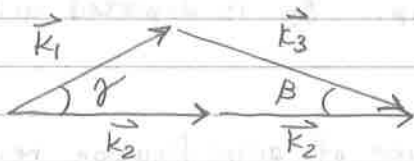
$$P(\omega_3) = \chi^{(3)} \mathcal{E}_2 \mathcal{E}_1^* \mathcal{E}_2 e^{i(2\vec{k}_2 - \vec{k}_1) \cdot \vec{r}} e^{-i\omega_3 t}$$

If we insert this into the wave equation (as we did in the beginning of this chapter for second-harmonic generation), the resulting wave will have a spatial dependence that goes as $e^{i(2\vec{k}_2 - \vec{k}_1 - \vec{k}_3) \cdot \vec{r}}$

- As before, to have efficient frequency mixing at ω_3 , phase matching is required:

$$2\vec{k}_2 - \vec{k}_1 - \vec{k}_3 = 0$$

It is not possible to satisfy this while keeping the three vectors parallel. In practice, \vec{k}_3 will be emitted in a direction that satisfies the above vector sum.



Introduce an angle γ between \vec{k}_1 and \vec{k}_2 , and the anti-Stokes radiation is emitted at an angle β .

- Parametric Amplification

- Start with the equations of propagation of sum frequency generation waves, involving $\mathcal{E}_1, \mathcal{E}_2, \mathcal{E}_3$. This time, regard ω_3 as a high-intensity pump wave, so \mathcal{E}_3 is constant:

$$\frac{d\mathcal{E}_1}{dz} = \frac{i}{\sqrt{\epsilon_1}} \frac{\omega_1}{2c} \chi^{(2)} \mathcal{E}_3 \mathcal{E}_2^*$$

$$\text{where } \omega_3 = \omega_2 + \omega_1,$$

$$\frac{d\mathcal{E}_2^*}{dz} = \frac{-i}{\sqrt{\epsilon_2}} \frac{\omega_2}{2c} \chi^{(2)} \mathcal{E}_3^* \mathcal{E}_1$$

and perfect phase matching is assumed

$$\Delta k = k_3 - k_2 - k_1 = 0$$

- Solve the coupled equations using matrix methods:

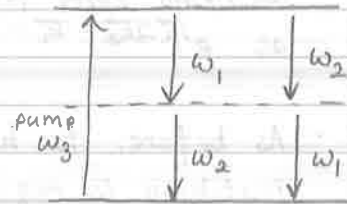
$$\frac{d}{dz} \begin{bmatrix} \mathcal{E}_1 \\ \mathcal{E}_2^* \end{bmatrix} = g \begin{bmatrix} \mathcal{E}_1 \\ \mathcal{E}_2^* \end{bmatrix} = \begin{bmatrix} 0 & \frac{i}{\sqrt{\epsilon_1}} \frac{\omega_1}{2c} \chi^{(2)} \mathcal{E}_3 \\ -\frac{i}{\sqrt{\epsilon_2}} \frac{\omega_2}{2c} \chi^{(2)} \mathcal{E}_3^* & 0 \end{bmatrix} \begin{bmatrix} \mathcal{E}_1 \\ \mathcal{E}_2^* \end{bmatrix}$$

gain/absorption if imag
propagation constant if
real

- This is solved in the same way as any eigenvalue equation; by taking the determinant

$$\Rightarrow g^2 = \frac{\omega_1 \omega_2}{4\sqrt{\epsilon_1 \epsilon_2} c^2} |\chi^{(2)}|^2 |\mathcal{E}_3|^2$$

- There is gain for both waves ω_1 and ω_2 , so that both $|\mathcal{E}_1|$ and $|\mathcal{E}_2|$ increase monotonically in space
- This is because the presence of ω_1 stimulates the emission of ω_2 , and vice versa. Therefore there is exponential growth for each wave, called parametric amplification



- * Thus, difference frequency generation is qualitatively dissimilar from sum frequency generation, where the intensity of the sum frequency oscillates with z .

- Eventually, as \mathcal{E}_1 and \mathcal{E}_2 build up, \mathcal{E}_3 is depleted and amplification does not proceed

- Quadrature squeezing: a special type of amplification results in the degenerate case, $\omega_1 = \omega_2 = \omega$, $\omega_3 = 2\omega$

$$\Rightarrow \frac{d\mathcal{E}_1}{dz} = \frac{\omega}{2\sqrt{\epsilon_1} c} \chi^{(2)} i \mathcal{E}_3 \mathcal{E}_1^* = \frac{\omega}{2\sqrt{\epsilon_1} c} |\chi^{(2)} \mathcal{E}_3| \mathcal{E}_1^*$$

choose $t=0$ such that \mathcal{E}_3 is pure imaginary (?)

- Now break up the wave $\mathcal{E}_1 = \mathcal{E}_1' + i \mathcal{E}_1''$

$$\begin{aligned} \text{Re}(\mathcal{E}_1 e^{i\omega t}) &= \frac{1}{2} \mathcal{E}_1 e^{i\omega t} + \frac{1}{2} \mathcal{E}_1^* e^{-i\omega t} \\ &= \frac{1}{2} \mathcal{E}_1' e^{i\omega t} + \frac{1}{2} \mathcal{E}_1'' e^{i\omega t} + \frac{1}{2} \mathcal{E}_1' e^{-i\omega t} - \frac{1}{2} \mathcal{E}_1'' e^{-i\omega t} \\ &= \mathcal{E}_1' \cos \omega t - \mathcal{E}_1'' \sin \omega t \end{aligned}$$

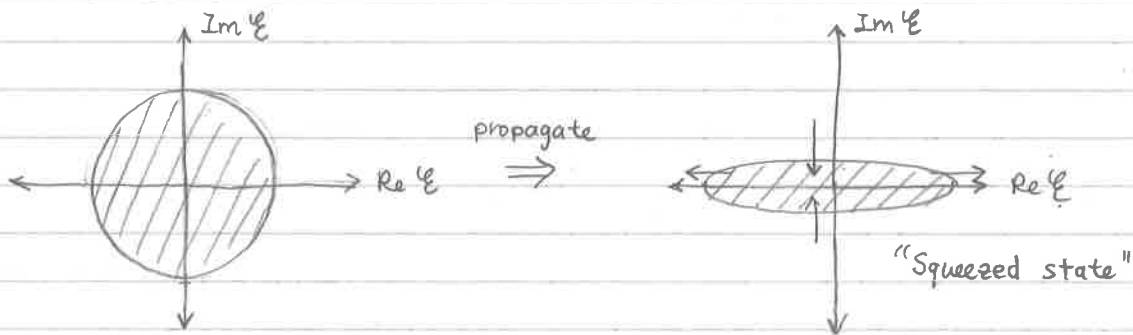
Then: Two parts with different phase

$$\text{Re}\left(\frac{d\mathcal{E}_1'}{dz}\right) = \frac{\omega}{2\sqrt{\epsilon_1} c} |\chi^{(2)} \mathcal{E}_3| \mathcal{E}_1' \Rightarrow \text{cosine is amplified}$$

$$\text{Re}\left(\frac{d\mathcal{E}_1''}{dz}\right) = -\frac{\omega}{2\sqrt{\epsilon_1} c} |\chi^{(2)} \mathcal{E}_3| \mathcal{E}_1'' \Rightarrow \text{sine is de-amplified}$$

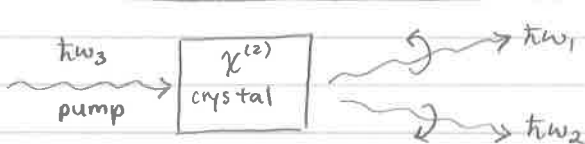
Thus, the amplification for the degenerate case is phase sensitive!
The direction of energy transfer also depends on the phase of \mathcal{E}_3 , which in our case was chosen to make \mathcal{E}_3 imaginary

- This type of amplification has quantum mechanical significance; depicted below



- The phenomenon of compressing the electric field in the complex plane is called quadrature squeezing, and the resultant state is called a squeezed electromagnetic state
- The uncertainty principle states that the product of the uncertainties in $\text{Re } \mathcal{E}$ and $\text{Im } \mathcal{E}$ has a minimum value
 - States where this uncertainty is exactly equal to the quantum noise limit are known as coherent states
 - Typically, the uncertainty is balanced between $\text{Re } \mathcal{E}$ and $\text{Im } \mathcal{E}$.
 - Otherwise, we have a squeezed state where the uncertainty in one of the components is lower than the quantum noise in that component in vacuum (but the uncertainty in the other component is correspondingly greater)

- Spontaneous parametric downconversion



- If only a pump beam is present, parametric amplification can still occur: the field that is amplified is zero-point noise (vacuum EM fluctuations)

- To preserve energy and momentum, the pump photon must simultaneously produce two photons satisfying:
 - 1) Energy conservation $\rightarrow \hbar\omega_3 = \hbar\omega_1 + \hbar\omega_2$
 - 2) Momentum conservation $\rightarrow \vec{k}_3 = \vec{k}_1 + \vec{k}_2$
 - 3) Angular momentum conservation \rightarrow the two photons have reciprocal polarizations summing to the pump polarization
- \Rightarrow The two photons are quantum mechanically entangled!
If $\omega_1 = \omega_2$, then they can be indistinguishable.

⑤ Nonlinear Optics in the Two-Level Approximation

- When transitions in the medium are resonantly excited, perturbation techniques become inadequate

- For instance, these methods cannot predict absorption or gain saturation
- To predict these phenomena, it is sufficient to treat just two quantum mechanical energy levels connected by an optical field

- Two-level system using density matrix

• The general expression for a wavefunction is

$$\psi = c_1 \psi_1 + c_2 \psi_2$$

$$|\psi|^2 = |c_1|^2 |\psi_1|^2 + |c_2|^2 |\psi_2|^2 + \underbrace{c_1^* c_2 \psi_1^* \psi_2 + c_1 c_2^* \psi_1 \psi_2^*}_{\text{interference terms}}$$

↑ $\hbar\omega_0 = E_2 - E_1$

• Formulate this using the density matrix for two levels:

$$\rho = \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{bmatrix} = \begin{bmatrix} |c_1|^2 & c_1 c_2^* \\ c_1^* c_2 & |c_2|^2 \end{bmatrix}$$

• The time evolution of the density matrix is:

$$\frac{d\rho}{dt} = \frac{d}{dt}(\psi\psi^*) = \left(\frac{d\psi}{dt}\right)\psi^* + \psi\left(\frac{d\psi^*}{dt}\right)$$

By Schrödinger's equation, $\frac{d\psi}{dt} = -\frac{i}{\hbar} \hat{H}\psi$, $\frac{d\psi^*}{dt} = \frac{i}{\hbar} \psi^* \hat{H}$ Hermitian
↓

$$\begin{aligned} \frac{d\rho}{dt} &= -\frac{i}{\hbar} \hat{H}\psi\psi^* + \frac{i}{\hbar} \psi(\psi^* \hat{H}) \\ &= -\frac{i}{\hbar} (\hat{H}\psi\psi^* - \psi\psi^* \hat{H}) = -\frac{i}{\hbar} [H, \psi\psi^*] \\ \frac{d\rho}{dt} &= -\frac{i}{\hbar} [\hat{H}, \rho] \end{aligned}$$

This can be used in place of Schrödinger's equation

• Consider ψ_1 and ψ_2 to be eigenstates of the original Hamiltonian \hat{H}_0 and now include the perturbation due to an electric field:

$$\hat{H}_0 \rightarrow \hat{H}_0 - \vec{p} \cdot \vec{E} = \hat{H}_0 - g \hat{x} \cdot \vec{E}$$

The new Hamiltonian including this interaction is:

$$\hat{H} = \begin{bmatrix} E_1 + \langle 1 | g \hat{x} \cdot \vec{E} | 1 \rangle & \langle 1 | g \hat{x} \cdot \vec{E} | 2 \rangle \\ \langle 2 | g \hat{x} \cdot \vec{E} | 1 \rangle & E_2 + \langle 2 | g \hat{x} \cdot \vec{E} | 2 \rangle \end{bmatrix}$$

Assume these terms are small compared to E_1, E_2

An alternative formulation using $\hat{p} \rightarrow \hat{p} - g\hat{A}$ can be shown to be equivalent under the Coulomb gauge

- Furthermore, make the dipole approximation: the electric field varies over a much larger length scale than the dipole moment of the atomic wavefunction, so $\langle 1 | \hat{q} \hat{x} \cdot \vec{E} | 2 \rangle = \langle 1 | \hat{q} \hat{x} | 2 \rangle \cdot \vec{E} \stackrel{\text{def}}{=} \varphi_{12}$

$$\Rightarrow \hat{H} = \begin{bmatrix} E_1 & -\varphi_{12} \mathcal{E} \\ -\varphi_{21} \mathcal{E} & E_2 \end{bmatrix}$$

- Now the equation of motion is fully specified:

$$\begin{bmatrix} \frac{d\rho_{11}}{dt} & \frac{d\rho_{12}}{dt} \\ \frac{d\rho_{21}}{dt} & \frac{d\rho_{22}}{dt} \end{bmatrix} = -\frac{i}{\hbar} \begin{bmatrix} E_1 & -\varphi_{12} \mathcal{E} \\ -\varphi_{21} \mathcal{E} & E_2 \end{bmatrix} \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{bmatrix} + \frac{i}{\hbar} \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{bmatrix} \begin{bmatrix} E_1 & -\varphi_{12} \mathcal{E} \\ -\varphi_{21} \mathcal{E} & E_2 \end{bmatrix}$$

$$\Rightarrow \frac{d\rho_{11}}{dt} = -\frac{i}{\hbar} (E_1 \rho_{11} - \rho_{21} \varphi_{12} \mathcal{E}) + \frac{i}{\hbar} (E_1 \rho_{11} - \rho_{12} \varphi_{21} \mathcal{E})$$

$$= \frac{i}{\hbar} (-\rho_{12} \varphi_{21} \mathcal{E} + \rho_{21} \varphi_{12} \mathcal{E})$$

$$\frac{d\rho_{11}}{dt} = \frac{i}{\hbar} \mathcal{E} (\rho_{21} \varphi_{12} - \rho_{12} \varphi_{21})$$

Similarly, $\frac{d\rho_{22}}{dt} = -\frac{i}{\hbar} \mathcal{E} (\rho_{21} \varphi_{12} - \rho_{12} \varphi_{21}) = -\frac{d\rho_{11}}{dt}$

$$\frac{d\rho_{12}}{dt} = +\frac{i}{\hbar} (\rho_{11} \varphi_{12} \mathcal{E} + E_2 \rho_{12} - \rho_{12} E_1 + \rho_{22} \varphi_{12} \mathcal{E})$$

$$= \frac{i}{\hbar} (E_2 - E_1) \rho_{12} + \frac{i}{\hbar} \varphi_{12} \mathcal{E} (\rho_{11} - \rho_{22})$$

$$= -i\omega_0 \rho_{12} + \frac{i}{\hbar} \varphi_{12} \mathcal{E} (\rho_{22} - \rho_{11})$$

$$\frac{d\rho_{21}}{dt} = +\frac{i}{\hbar} (E_1 \rho_{21} - \varphi_{21} \mathcal{E} \rho_{22} + \varphi_{21} \mathcal{E} \rho_{11} - E_2 \rho_{21})$$

$$= -i\omega_0 \rho_{21} - \frac{i}{\hbar} \varphi_{21} \mathcal{E} (\rho_{22} - \rho_{11})$$

Notice that since $\rho_{12} = \rho_{21}^*$, we must have $\frac{d\rho_{12}}{dt} = \left(\frac{d\rho_{21}}{dt}\right)^*$ which is satisfied if $\varphi_{12} = \varphi_{21}^*$. Thus, we need only be concerned with the following rates:

$$\frac{d}{dt} (\rho_{11} - \rho_{22}) = 2 \frac{i}{\hbar} \mathcal{E} (\rho_{21} \varphi_{12} - \rho_{12} \varphi_{21})$$

$$\frac{d\rho_{21}}{dt} = -i\omega_0 \rho_{21} - \frac{i}{\hbar} \varphi_{21} \mathcal{E} (\rho_{22} - \rho_{11})$$

(signs agree with Eli's notes)

- When the two-level system is not perfectly isolated, two types of relaxation can occur:

1) Energy relaxation: after a finite time T_1 , the excited state tends to decay to the lower state. The density matrix is modified to:

$$\begin{aligned} \text{excited state: } \frac{d\rho_{22}}{dt} &= \left(\frac{d\rho_{22}}{dt}\right)_0 \overset{\text{relaxation}}{\leftarrow} - \frac{1}{T_1} (\rho_{22} - \rho_{22}^0) \\ \text{ground state: } \frac{d\rho_{11}}{dt} &= \left(\frac{d\rho_{11}}{dt}\right)_0 + \frac{1}{T_1} (\rho_{22} - \rho_{22}^0) \\ &= \left(\frac{d\rho_{11}}{dt}\right)_0 + \frac{1}{T_1} (\rho_{11}^0 - \rho_{11}) \end{aligned}$$

2) Phase relaxation: since the excited state has a finite linewidth, after time T_2 it is no longer purely in state $|2\rangle$ and the interference terms vanish (decoherence)

⇒ Rates are modified to:

$$\textcircled{1} \quad \frac{d}{dt} (\rho_{11} - \rho_{22}) = \frac{2i}{\hbar} \mathcal{E} (\rho_{21} \rho_{12} - \rho_{12} \rho_{21}) - \frac{(\rho_{11} - \rho_{22}) - (\rho_{11} - \rho_{22})_0}{T_1}$$

$$\textcircled{2} \quad \frac{d}{dt} \rho_{21} = -i\omega_0 \rho_{21} - \frac{i}{\hbar} \mathcal{E} \rho_{21} (\rho_{22} - \rho_{11}) - \frac{\rho_{21}}{T_2}$$

- What are the steady-state solutions?

Let $\rho_{21}(t)$ have a slowly varying amplitude component:

$$\rho_{21}(t) = \sigma_{21}(t) e^{-i\omega t} \quad \leftarrow \text{frequency of } \vec{\mathcal{E}}$$

Plug this into $\textcircled{1}$ and $\textcircled{2}$, and let the LHS equal zero for the steady state solution:

$$\textcircled{1} \Rightarrow 0 = \frac{2i}{\hbar} \mathcal{E} (\sigma_{21} \rho_{21}^* - \sigma_{21}^* \rho_{21}) - \frac{(\rho_{11} - \rho_{22}) - (\rho_{11} - \rho_{22})_0}{T_1}$$

$$\textcircled{2} \Rightarrow 0 = -i(\omega_0 - \omega) \sigma_{21} - \frac{\sigma_{21}}{T_2} - \frac{i}{\hbar} \mathcal{E} \rho_{21} (\rho_{22} - \rho_{11})$$

- If we solve these coupled equations, we get:

$$\rho_{22} - \rho_{11} = (\rho_{22} - \rho_{11})_0 \frac{1 + (\omega - \omega_0)^2 T_2^2}{1 + (\omega - \omega_0)^2 T_2^2 + 4\Omega_S^2 T_2 T_1}$$

where $\Omega_S \equiv \frac{\mathcal{E} \rho_{21}}{2\hbar}$ is the Rabi frequency

* Note that if Ω_S is large (by pumping very hard), we get $\rho_{11} = \rho_{22}$. The up and down transitions have equal rates (saturation)

- The solution for σ_{21} is:

$$\sigma_{21} = \frac{\varphi_{21} \mathcal{E} (\rho_{22} - \rho_{11})}{\hbar (\omega - \omega_0 + i/T_2)} = \frac{\varphi_{21} \mathcal{E} (\rho_{22} - \rho_{11})_0 (1 + (\omega - \omega_0)^2 T_2^2)}{\hbar (\omega - \omega_0 + i/T_2) (1 + (\omega - \omega_0)^2 T_2^2 + 4\Omega_S^2 T_1 T_2)}$$

$$\bar{\sigma}_{21} = \frac{2\Omega_S (\rho_{22} - \rho_{11})_0}{\omega - \omega_0 + i/T_2} \cdot \frac{1 + (\omega - \omega_0)^2 T_2^2}{1 + (\omega - \omega_0)^2 T_2^2 + 4\Omega_S^2 T_1 T_2}$$

(By taking the real and imaginary parts of this, we can match the solution in Eli's notes)

- The polarization is:

$$P(t) = N \underbrace{(\varphi_{21}^* \sigma_{21} + \varphi_{21} \sigma_{21}^*)}_{\text{mean dipole moment}} e^{i\omega t} = P e^{i\omega t} = \epsilon_0 \chi \mathcal{E} e^{i\omega t}$$

For simplicity, we'll discard the second term

$$\begin{aligned} \Rightarrow \chi(\omega) &= \frac{N \varphi_{21}^* \sigma_{21}}{\epsilon_0 \mathcal{E}} = \frac{N |\varphi_{21}|^2 (\rho_{22} - \rho_{11})}{\epsilon_0 \hbar (\omega - \omega_0 + i/T_2)} \\ &\equiv \Delta N_0 \\ &= \frac{N (\rho_{22} - \rho_{11})_0 |\varphi_{21}|^2 (1 + (\omega - \omega_0)^2 T_2^2)}{\epsilon_0 \hbar (\omega - \omega_0 + i/T_2) (1 + (\omega - \omega_0)^2 T_2^2 + 4\Omega_S^2 T_1 T_2)} \end{aligned}$$

Find the real and imaginary parts:

$$\chi'(\omega) = \frac{\Delta N_0 |\varphi_{21}|^2}{\epsilon_0 \hbar} \cdot \frac{1 + (\omega - \omega_0)^2 T_2^2}{1 + (\omega - \omega_0)^2 T_2^2 + 4\Omega_S^2 T_1 T_2} \cdot \frac{\omega - \omega_0}{(\omega - \omega_0)^2 + 1/T_2^2}$$

$$\chi'(\omega) = \frac{\Delta N_0 |\varphi_{21}|^2 T_2}{\epsilon_0 \hbar} \cdot \frac{(\omega - \omega_0) T_2}{1 + (\omega - \omega_0)^2 T_2^2 + 4\Omega_S^2 T_1 T_2}$$

and likewise

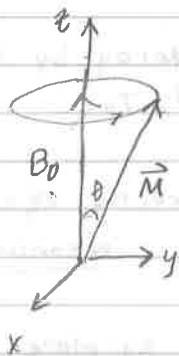
$$\chi''(\omega) = \frac{\Delta N_0 |\varphi_{21}|^2 T_2}{\epsilon_0 \hbar} \cdot \frac{1}{1 + (\omega - \omega_0)^2 T_2^2 + 4\Omega_S^2 T_1 T_2}$$

$\omega - \omega_0$ is the detuning

- These are the susceptibilities associated with an optical field interacting with a two-level system.
- The term containing $\Omega_S^2 \propto |\mathcal{E}|^2$ accounts for absorption or gain saturation. As $|\mathcal{E}|^2$ increases, $\alpha \rightarrow 0$.
- A strong driving field also leads to broadening of the absorption line

Magnetic Resonance

- A classical magnetic moment is a famous example of a two-level system; it contains all the dynamics of the quantum mechanical two-level system.



$\vec{\tau} = \vec{M} \times \vec{B}$: magnet tries to line up with the applied magnetic field.

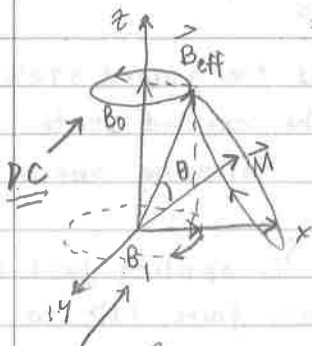
- The magnetic moment \vec{M} is proportional to the angular momentum \vec{J} by the gyromagnetic ratio: $\gamma \vec{J} = \vec{M}$

$$\vec{\tau} = \frac{d\vec{J}}{dt} = \vec{M} \times \vec{B} \Rightarrow \boxed{\frac{d\vec{M}}{dt} = \gamma \vec{M} \times \vec{B}}$$

magnetic moment equation of motion

- The angular frequency of Larmor precession is γB_0
- The angle θ between \vec{M} and \vec{B} remains at its initial value.

- Now introduce an x-y component of $\vec{B} = B_1 \cos \omega t \hat{x} + B_1 \sin \omega t \hat{y} + B_0 \hat{z}$

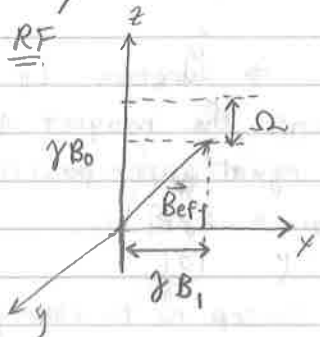


\Rightarrow Then the magnetic moment can be induced

to precess around a new magnetic field vector, and \vec{M} can explore the sphere of directions (Bloch sphere)

- But with \vec{B} rotating and \vec{M} rotating around \vec{B} , this quickly gets complicated

- Switch to a rotating reference frame where \vec{B} is stationary, e.g. choose \vec{B}_1 to lie along \hat{x}



$$\left. \frac{d\vec{M}}{dt} \right|_{\text{rot}} = \frac{d\vec{M}}{dt} + \vec{\Omega} \times \vec{M} \quad \text{by vector calculus}$$

angular frequency vector of rotation about \hat{z} : $\vec{\Omega} = \omega \hat{z}$

$$\Rightarrow \left. \frac{d\vec{M}}{dt} \right|_{\text{rot}} = \gamma \vec{M} \times \vec{B} + \vec{\Omega} \times \vec{M} = \gamma \vec{M} \times \vec{B} - \vec{M} \times \vec{\Omega} = \vec{M} \times \gamma \vec{B} - \vec{M} \times \vec{\Omega}$$

$$\boxed{\left. \frac{d\vec{M}}{dt} \right|_{\text{rot}} = \vec{M} \times (\gamma \vec{B} - \vec{\Omega})}$$

effective magnetic field $\vec{B}_{\text{eff}} = \vec{B} - \frac{\omega}{\gamma} \hat{z}$

- Damping also exists in magnetic resonance

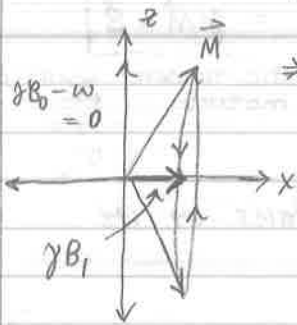
- The energy in the magnetic precession is $\vec{M} \cdot \vec{B}$, so the component of \vec{M} along \vec{B} can decay by energy relaxation:

$$\frac{d}{dt} (\vec{M} \cdot \vec{B}) = \gamma (\vec{M} \times \vec{B}) \cdot \vec{B} - \frac{(\vec{M} \cdot \vec{B}) - (\vec{M} \cdot \vec{B})_0}{T_1}$$

\uparrow M_z in normal frame

- The components in the other directions can decay by the process of phase relaxation, at the rate $1/T_2$.

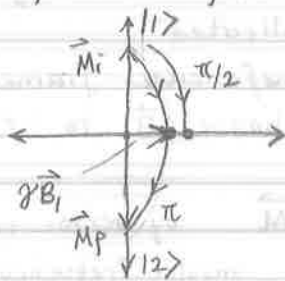
- Consider the case where the RF frequency is exactly equal to the Larmor precession frequency: $\omega = \gamma B_0$: resonance



\Rightarrow In this condition \vec{B}_{eff} lies in the xy -plane, and \vec{M} precesses about the x -axis. This precession of the medium's magnetic dipole moments emits radiation which can be detected: this is the principle of magnetic resonance imaging (MRI)

- How does this relate to population inversion?

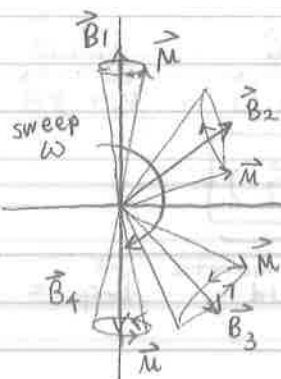
- Consider the case of \vec{M} aligned to $B_0 \hat{z}$ as the ground state $|1\rangle$, and the case of \vec{M} aligned with $-B_0 \hat{z}$ the excited state $|2\rangle$
- If the magnet begins at $|1\rangle$, consider the following operations:



1) π pulse: the RF field is applied just long enough for \vec{M} to precess from $|1\rangle$ to $|2\rangle$. The duration is:

$$\gamma B_1 T_\pi = \pi \Rightarrow \text{duration } T_\pi = \frac{\pi}{\gamma B_1}$$

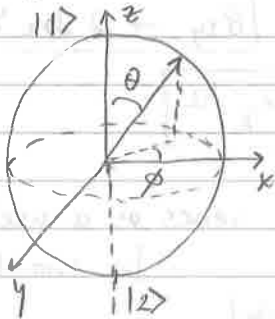
2) $\frac{\pi}{2}$ pulse: the pulse sends the magnet to the x -axis, which is an equal superposition of $|0\rangle$ and $|1\rangle$: $T_{\pi/2} = \frac{\pi}{2\gamma B_1}$



3) Adiabatic fast passage: sweep ω to slowly rotate \vec{B}_{eff} from \hat{z} to $-\hat{z}$, starting with $\omega = 0$ and ending at $\omega = 2\gamma B_0$. Then abruptly turn off the magnetic field.
 $\Rightarrow \vec{M}$ remains at $|1\rangle$

- When the RF frequency is not on resonance ($\omega \neq \gamma B_0$), the magnetic moment \vec{M} does not precess about the \hat{x} axis but rather about $\gamma B_1 \hat{x} + (\gamma B_0 - \omega) \hat{z}$. The precession frequency is $\sqrt{(\gamma B_1)^2 + (\gamma B_0 - \omega)^2}$.

- Magnetic Resonance: correspondence with 2-level electronic system



- The states of any 2-level system can be represented on a Bloch sphere if we specify

$$|\psi\rangle = c_1 |\psi_1\rangle + c_2 |\psi_2\rangle$$

$$= e^{i\phi/2} \cos \frac{\theta}{2} |\psi_1\rangle + e^{-i\phi/2} \sin \frac{\theta}{2} |\psi_2\rangle$$

Automatically normalized

Then:

$$\rho_{11} - \rho_{22} = \cos^2 \frac{\theta}{2} - \sin^2 \frac{\theta}{2} = \cos \theta = M_z$$

$$2\rho_{21} = 2 \cos \frac{\theta}{2} \sin \frac{\theta}{2} e^{-i\phi} = \sin \theta e^{-i\phi}$$

$$= M_x - iM_y$$

$$\rho_{21} = \frac{1}{2} (M_x - iM_y)$$

- From this we see that the magnetic moment \vec{M} corresponds to the density matrix, which like \vec{M} has three degrees of freedom

$$\Rightarrow \boxed{M_z \rightarrow \rho_{11} - \rho_{22}}, \quad \boxed{M_x \rightarrow 2 \operatorname{Re}(\rho_{21})}, \quad \boxed{M_y \rightarrow 2 \operatorname{Im}(\rho_{21})}$$

- To complete the correspondence, we make an analogy of the magnetic moment's equation of motion with the dynamics of the two-level system:

$$\frac{d\vec{M}}{dt} = \gamma \vec{M} \times \vec{B} \quad \Leftrightarrow \quad \frac{d\vec{r}}{dt} = \vec{\Omega} \times \vec{r}$$

where $\vec{r} = 2 \operatorname{Re} \rho_{21} \hat{x} + 2 \operatorname{Im} \rho_{21} \hat{y} + (\rho_{11} - \rho_{22}) \hat{z}$. From Schrödinger's equation using $\hat{H} = \begin{bmatrix} E_1 & \rho_{21}^* \mathcal{E} \\ \rho_{21} \mathcal{E} & E_2 \end{bmatrix}$ as the Hamiltonian, the right

side equation is satisfied if:

$$\vec{\Omega} = -2 \operatorname{Re} \left(\frac{\rho_{12} \mathcal{E}}{\hbar} \right) \hat{x} - 2 \operatorname{Im} \left(\frac{\rho_{12} \mathcal{E}}{\hbar} \right) \hat{y} - \omega_0 \hat{z}$$

$$\text{where } \hbar \omega_0 \equiv E_2 - E_1$$

→

Then, to make the correspondence between the equations of motion, we must have: $\vec{\Omega} \rightarrow -\gamma \vec{B}$

$$\Rightarrow \gamma \begin{bmatrix} B_{1x} \\ B_{1y} \\ B_0 \end{bmatrix} = \frac{\mu_{12}}{\hbar} \begin{bmatrix} 2 \operatorname{Re} \mathcal{E} \\ 2 \operatorname{Im} \mathcal{E} \\ \hbar \omega_0 / \mu_{12} \end{bmatrix}$$

The factors of 2 are somewhat dubious...

The correspondence is: $B_{1x} \rightarrow 2 \operatorname{Re} \mathcal{E}(t)$, $B_{1y} \rightarrow 2 \operatorname{Im} \mathcal{E}(t)$

$$\gamma \rightarrow \frac{\mu_{12}}{\hbar} \quad \text{and} \quad \gamma B_0 \rightarrow \frac{E_2 - E_1}{\hbar}$$

This completes the analogy! Magnetic resonance serves as a useful model for two-level systems.

- The two-level system vs. harmonic oscillator model

- The expressions for χ obtained using these two methods are actually remarkably similar:

$$\text{Harmonic oscillator: } \sum_i \omega_i \chi''_{HO}(\omega) = \sum_i \frac{N g^2}{m \epsilon_0} \frac{1/4\tau}{(\omega_i - \omega)^2 + (1/2\tau)^2} \times S_i$$

$$\text{Two-level system: } \omega_{12} \chi''_{2L}(\omega) = \frac{\Delta N_0 \mu_{12}^2}{\epsilon_0 \hbar} \frac{1/T_2}{\omega_{12} [(\omega_{12} - \omega)^2 + 1/T_2^2 + 4\Omega_S^2 T_1/T_2]}$$

* This term is the main difference between the two, and accounts for the fact that when there are only two levels, absorption saturates.

- We can ignore saturation if we extend the two-level system to a ladder of energy levels $|i\rangle$ with energy $\hbar\omega_i$ above $\hbar\omega_1$. Then:

$$\sum_i \omega_i \chi''_{2L}(\omega) = \sum_i \frac{\Delta N_0 g^2 |\langle 1 | \hat{x} | i \rangle|^2}{\epsilon_0 \hbar} \frac{\omega_i (1/T_2)}{(\omega_i - \omega)^2 + 1/T_2^2}$$

$$\Rightarrow \sum_i \int \omega_i \chi''_{2L}(\omega) d\omega = \sum_i \frac{\Delta N_0 g^2 |\langle 1 | \hat{x} | i \rangle|^2}{\epsilon_0 \hbar} \omega_i \int d\omega \frac{1/T_2}{(\omega_i - \omega)^2 + 1/T_2^2}$$

$$\sum_i \int \omega_i \chi''_{2L}(\omega) d\omega = \sum_i \frac{\Delta N_0}{\epsilon_0 \hbar} g^2 |\langle 1 | \hat{x} | i \rangle|^2 \omega_i \pi$$

Lorentzian

To make a correspondence with the harmonic oscillator model, we must find a simplification for $\sum_i |\langle 1 | \hat{x} | i \rangle|^2$. For the harmonic oscillator model, we previously showed:

$$\sum_i \int \omega_i \chi''_{HO}(\omega) d\omega = \frac{N g^2}{2m \epsilon_0} \pi \sum_i S_i = \frac{N g^2}{2m \epsilon_0} \pi$$

- The term $w_i |\langle 1 | \hat{x} | i \rangle|^2$ can indeed be simplified.

Start by finding the commutator $[\hat{x}, \hat{H}]$ using Ehrenfest's theorem:

$$\begin{aligned} \frac{d}{dt} \langle \hat{x} \rangle &= \frac{1}{i\hbar} \langle [\hat{x}, \hat{H}] \rangle + \left\langle \frac{\partial \hat{x}}{\partial t} \right\rangle \\ \frac{1}{m} \langle \hat{p} \rangle &= \frac{1}{i\hbar} \langle [\hat{x}, \hat{H}] \rangle \\ \Rightarrow \frac{i\hbar}{m} \hat{p} &= [\hat{x}, \hat{H}] \end{aligned}$$

• Then the nested commutator $[[\hat{x}, \hat{H}], \hat{x}]$ is:

$$\begin{aligned} [[\hat{x}, \hat{H}], \hat{x}] &= [\hat{x}, \hat{H}] \hat{x} - \hat{x} [\hat{x}, \hat{H}] \\ &= \frac{i\hbar}{m} (\hat{p} \hat{x} - \hat{x} \hat{p}) = \frac{i\hbar}{m} [\hat{p}, \hat{x}] = \frac{\hbar^2}{m} \end{aligned}$$

• However, there is another way to write this commutator:

$$\begin{aligned} \frac{1}{2} [[\hat{x}, \hat{H}], \hat{x}] &= \frac{1}{2} (xH - Hx)x - \frac{1}{2} x(xH - Hx) \\ &= xHx - \frac{1}{2} Hx^2 - \frac{1}{2} x^2 H \\ \frac{1}{2} \langle 1 | [[\hat{x}, \hat{H}], \hat{x}] | 1 \rangle &= \langle 1 | xHx | 1 \rangle - \frac{1}{2} (\langle 1 | Hx^2 | 1 \rangle + \langle 1 | x^2 H | 1 \rangle) \\ &= \langle 1 | xHx | 1 \rangle - E_1 \langle 1 | x^2 | 1 \rangle \end{aligned}$$

Use spectral decomposition to expand the wavefunction:

$$\begin{aligned} \psi: |1\rangle &= \sum_i |i\rangle \langle i|1\rangle \\ x|1\rangle &= \sum_i |i\rangle \langle i|x|1\rangle \end{aligned}$$

$$\Rightarrow \frac{1}{2} \langle 1 | [[\hat{x}, \hat{H}], \hat{x}] | 1 \rangle = \sum_i \langle 1 | x | i \rangle \langle i | H | i \rangle \langle i | x | 1 \rangle - E_1 \sum_i \langle 1 | x | i \rangle \langle i | x | 1 \rangle$$

previous result

$$\frac{\hbar^2}{2m} = \sum_i (E_i - E_1) |\langle 1 | x | i \rangle|^2$$

$$\frac{\hbar}{2m} = \sum_i w_i |\langle 1 | x | i \rangle|^2$$

$$1 = \frac{2m}{\hbar} \sum_i w_i |\langle 1 | x | i \rangle|^2 \rightarrow$$

A surprising identity!

For the case of a Bloch wavefunctions (such as those found in crystals) which have the form $\psi(x) = u(x)e^{ikx}$, an additional term is added as a perturbation on the Hamiltonian. Using second order perturbation theory, we can show that the above relation still holds, but with the electron mass replaced by the band effective mass:

$$\frac{1}{m_{\text{eff}}} = \frac{2w_{12}}{\hbar} |\langle 1 | \hat{x} | 2 \rangle|^2$$

transition electric dipole length

where we have treated the crystal as a two-level system defined by the conduction and valence band edges.

- Now we can match the quantum mechanical absorption to that of the harmonic oscillator model

$$\begin{aligned}
 \text{QM: } \sum_j \int \omega_j \chi''_{\text{QM}}(\omega) d\omega &= \sum_j \frac{\Delta N_0 q^2}{2m\epsilon_0} \frac{2m}{\hbar} |\langle 1 | \hat{x} | 1 \rangle|^2 \omega_j \pi \\
 &= \frac{\Delta N_0 q^2}{2m\epsilon_0} \cdot \pi \cdot \underbrace{\sum_j \frac{2m}{\hbar} \omega_j |\langle 1 | \hat{x} | 1 \rangle|^2}_{=1}
 \end{aligned}$$

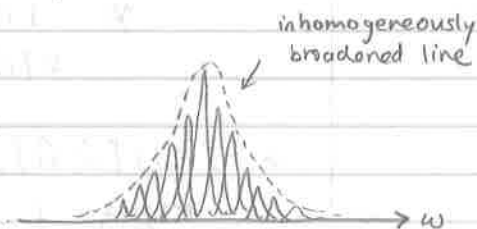
$$\text{HO: } \sum_j \int \omega_j \chi''_{\text{HO}}(\omega) d\omega = \frac{N_0 q^2}{2m\epsilon_0} \cdot \pi \cdot \underbrace{\sum_j S_j}_{=1}$$

The correspondence is complete, with the logical associations given by:

- $\Delta N_0 = N(\rho_{22} - \rho_{11}) \leftrightarrow N$; number of states/oscillators available to make an energy transition
- The oscillator strengths: $\frac{2m}{\hbar} \omega_j |\langle 1 | \hat{x} | 1 \rangle|^2 \leftrightarrow S_j$
Both sum up to 1.

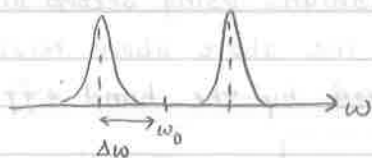
- **Motional Narrowing**: a useful concept in spectroscopy

- In the presence of inhomogeneities (e.g. spatially varying magnetic field in an inhomogeneous medium), the resonance frequency is broadened



- However, if atoms/molecules are moving around, they will each explore more of the material's inhomogeneities, and the time-averaged resonance frequency of each atom has less variation than the instantaneous resonance frequency; this is motional narrowing

- Consider a simpler case: two possible resonance frequencies arising from a molecule jumping between 2 inhomogeneities (thus 2 resonance frequencies): this is a simplified form of inhomogeneous broadening



- As the molecule jumps around, its phase changes: $\xi(t) = e^{-i\omega(t)t}$

- The correct way to find the spectrum in the presence of molecular motion is by taking a Fourier transform:

$$\text{Spectrum } F(\omega) = \int e^{i \int_{-\infty}^t \omega(t') dt'} e^{-i\omega t} dt$$

In the case of jumping between two energy levels, we have:

$$e^{i\omega(t) \cdot t} = e^{i\omega_0 t} \cdot e^{\underbrace{\pm i\Delta\omega t}_{\text{phase}}}$$

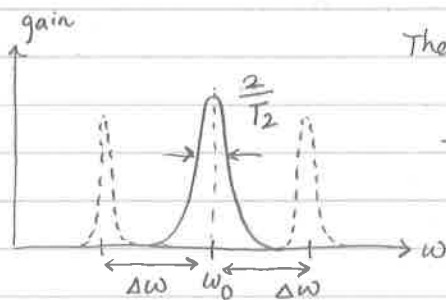
- A simple analysis is sufficient to determine what happens. On a single random jump, the phase changes by $\delta\phi = \Delta\omega \cdot \tau$ where τ is the average time between jumps
- After N jumps, the RMS change in phase is:

$$\begin{aligned} \sqrt{\langle \phi \rangle^2} &= \sqrt{\langle (-1-1+1+1-\dots)^2 \cdot \delta\phi^2 \rangle} = \sqrt{N \cdot \delta\phi^2} \\ &= \sqrt{N} \cdot \delta\phi \end{aligned}$$

When this accumulated phase change goes to ~ 1 rad, the original phase is lost. We can use this to define the phase relaxation time T_2

$$\sqrt{\langle \phi \rangle^2} = 1 \quad \Rightarrow \quad \sqrt{N} \delta\phi = \sqrt{N} \cdot \Delta\omega \tau = 1$$

jumps $N = \frac{1}{\Delta\omega^2 \tau^2}$



Then we have $T_2 = N \cdot \tau$ so

$$T_2 = \frac{1}{\Delta\omega^2 \tau}$$

- Therefore, in the case of short correlation times $\tau \ll 1/\Delta\omega$, a narrow line at the center frequency is produced if the molecular motion is fast

- If the jumping is slow $\tau \gg 1/\Delta\omega$, we have the original inhomogeneous broadening situation, and (for this example) the two lines at $\omega_0 \pm \Delta\omega$ are recovered with no signal at the center frequency

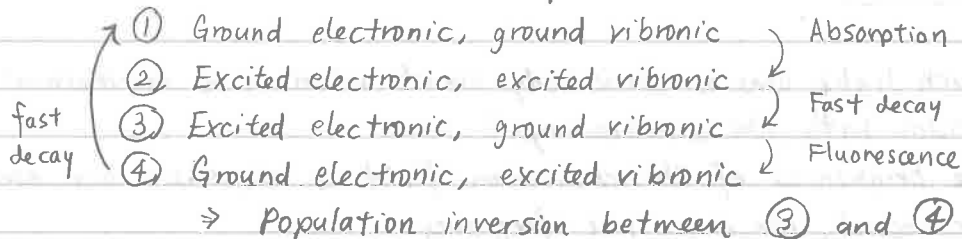
⑥ Stimulated Emission

- An optoelectronically active material is one in which an electronic excitation lingers for a relatively long time ($\approx 1\text{ns}$ or more) rather than degrading rapidly to heat (within $\approx 1\text{ps}$ or less). These are the materials used for light emission.
 - Examples:
 - atoms in a gas: Na, Ne, Hg, etc.
 - atoms in a crystal: Cr^{3+} in Al_2O_3
 Na^{3+} in glass

- Stimulated emission becomes significant if a large population inversion can be achieved. Common methods are:

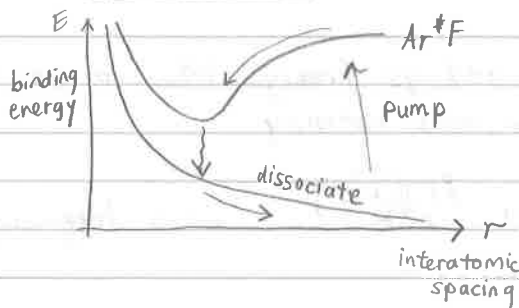
- Three-level pumping: external pump excites electrons from a ground state ① to an excited state ③. Electrons decay slowly from ③ to a lower state ② but ② is quickly depopulated
 - Population inversion between ③ and ②
 - Ex: Cr^{3+} in $\text{Al}_2\text{O}_3 \rightarrow$ ruby laser
- Four-level pumping: similar to above, but electrons are pumped from ① to ④. ④ decays rapidly to a lower state ③. Another lower state decays rapidly from ② to ①
 - More efficient population inversion is possible between ③ and ②
 - Ex: Nd:YAG laser, He-Ne laser

- Franck-Condon principle: electronic transitions are much faster than nuclear motions, so electronic-vibronic transitions that involve the least nuclear motion to be favored:



- Ex: dye molecules

- Excimer lasers: involves a noble gas (e.g. Ar) and halogen (e.g. F)



- Excited Ar^* atoms induced by electric discharge pumping bond readily with F atoms
- On emission, Ar^*F relaxes to ArF , after which the two atoms are rapidly repelled and dissociated

- Lasers can also be categorized by the physical mechanism by which the gain medium is pumped

1) Optical pumping: ruby laser, Nd:YAG

2) Electrical discharge: excimer lasers

3) Inelastic atomic collisions: electric discharge excites gas atoms of type A, which coincide with excited energy levels of gas B, and energy is transferred between the gas species by collisions

• Ex: He-Ne laser, CO₂ laser (N₂ → CO₂)

4) Direct current injection: semiconductor diode lasers

- Semiconductor diode lasers: most common type of laser

Al_{0.3}Ga_{0.7}As
 GaAs
Al_{0.3}Ga_{0.7}As

- Population inversion between a large number of states in the conduction band and the valence band
- Emitted photon has $h\nu = E_g$

- A double heterostructure guides the beam, forming an optical cavity, while also creating a potential well for confining electrons and holes

- More efficient than dye lasers:

- Conduction band states are delocalized, and electrons and holes can move independently w.r.t. each other

- To be contrasted with dyes in which electrons and holes are bound together as excitons, which quickly recombine

- Long recombination lifetimes (~μs) facilitate stimulated emission

- Detailed balance

- How much light does a black body emit when it is in thermal equilibrium with its surroundings?

- The brightness of the radiation field is the # photons emitted per second, per area, per frequency:

$$B(\nu, T) = \frac{8\pi n^2 \nu^2}{c^2} \frac{1}{e^{h\nu/kT} - 1} d\nu$$

- In equilibrium, photons come and go freely. When a photon is absorbed, the radiation field loses entropy:

$$\Delta S_{\text{rad}} = \frac{h\nu}{T} = -k \ln \left(1 + \frac{8\pi n^2 \nu^2}{c^2 B} \right) \quad \text{at a frequency } \nu$$

- Meanwhile, the material gains entropy when it absorbs a photon

$$\text{Free energy per molecule} = \mu = U - T\Delta S_{\text{mat}} \Rightarrow \Delta S_{\text{mat}} = \frac{h\nu - \mu}{T}$$

The change in entropy must be zero by the second law of thermodynamics: $\Delta S_{\text{rad}} = \Delta S_{\text{mat}}$

$$\frac{h\nu - \mu}{T} = k \ln \left(1 + \frac{8\pi n^2 \nu^2}{c^2 B} \right)$$

$$\hookrightarrow B(\nu, \mu, T) = \frac{8\pi n^2 \nu^2}{c^2} \frac{1}{e^{(h\nu - \mu)/kT} - 1}$$

This is the brightness of the photon field in equilibrium with a semiconductor with chemical potential μ

- Now apply detailed balance:

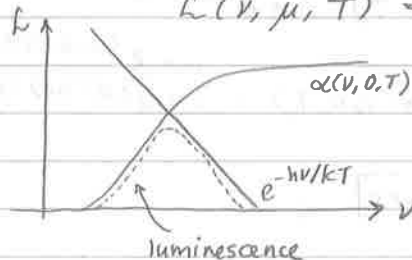
$$\text{emission } L(\nu, \mu, T) = \alpha(\nu, \mu, T) B(\nu, \mu, T) \quad \text{absorption}$$

$$L(\nu, \mu, T) = \alpha(\nu, \mu, T) \frac{8\pi n^2 \nu^2}{c^2} e^{(\mu - h\nu)/kT}$$

assuming $(h\nu - \mu) \gg kT$

- If we assume simply that $\alpha(\nu, \mu, T) = \alpha(\nu, 0, T)$, then the luminescence is:

$$L(\nu, \mu, T) = \alpha(\nu, 0, T) e^{-h\nu/kT} \cdot \left(\frac{8\pi n^2 \nu^2}{c^2} e^{\mu/kT} \right)$$



- Indeed, for many materials (e.g. Ge, Si, GaAs), the luminescence is found to obey $L \sim \alpha \cdot e^{-h\nu/kT}$

- Integrated over the spectrum, this is written as:

$$B_{\text{np}} = \int d\nu \alpha(\nu) \frac{8\pi n^2 \nu^2}{c^2} e^{(\mu - h\nu)/kT} \quad \leftarrow \text{absorption}$$

luminescence = radiative recombination

law of mass action

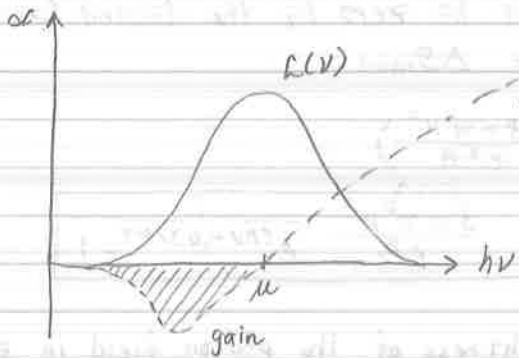
$$B_{\text{ni}}^2 e^{\mu/kT} = e^{\mu/kT} \int d\nu \alpha(\nu) \frac{8\pi n^2 \nu^2}{c^2} e^{-h\nu/kT}$$

$$\Rightarrow B = \frac{1}{n_i^2} \int d\nu \alpha(\nu) \frac{8\pi n^2 \nu^2}{c^2} e^{-h\nu/kT}$$

This is the Shockley-van Roosbroeck relation, which gives the radiative coefficient B .

- Go back to the exact expression $L(\nu, \mu) = \alpha(\nu, \mu) B(\nu, \mu)$ and rearrange:

$$\alpha(\nu, \mu, T) = L(\nu, \mu, T) \frac{c^2}{8\pi n^2 \nu^2} \times \left(e^{(h\nu - \mu)/kT} - 1 \right)$$



this can be negative

- This expression suggests that if $\mu > h\nu$, $\alpha < 0$ and there is gain!

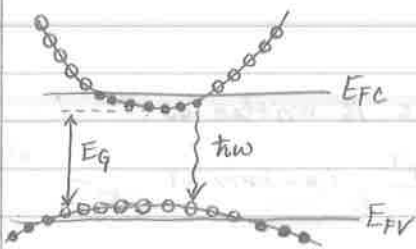
- Find an improved approximation to $\alpha(\nu, \mu, T)$ in terms of $\alpha(\nu, 0, T)$:

$$\begin{aligned} L(\nu, \mu, T) &= \frac{8\pi n^2 \nu^2}{c^2} \alpha(\nu, \mu, T) \cdot \frac{1}{e^{(h\nu - \mu)/kT} - 1} \\ &= \frac{8\pi n^2 \nu^2}{c^2} \alpha(\nu, \mu, T) \cdot \frac{e^{(\mu - h\nu)/kT}}{1 - e^{(\mu - h\nu)/kT}} \end{aligned}$$

Approximation: luminescence saturates (?)

$$\begin{aligned} L(\nu, \mu, T) &= \frac{8\pi n^2 \nu^2}{c^2} \alpha(\nu, 0, T) \frac{e^{(\mu - h\nu)/kT}}{1 + e^{(\mu - h\nu)/kT}} \\ \rightarrow \alpha(\nu, \mu, T) &= \alpha(\nu, 0, T) \cdot \frac{1 - e^{(\mu - h\nu)/kT}}{1 + e^{(\mu - h\nu)/kT}} \end{aligned}$$

Stimulated emission in semiconductors



- Under illumination or the application of voltage, two Fermi levels exist in the semiconductor: one for electrons (E_{fc}) and one for holes (E_{fv}). Their separation is the chemical potential: $h\nu = E_{fc} - E_{fv}$

- The emission rate is given by Fermi's golden rule:

$$\frac{1}{\tau} = \frac{2\pi}{\hbar} |M|^2 \frac{dN}{dE_j} \leftarrow \begin{array}{l} \text{CB / VB} \\ \text{joint density of} \\ \text{states} \end{array}$$

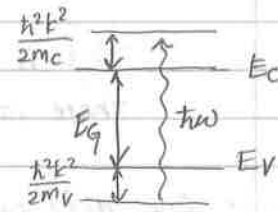
dipole matrix element

- Photons have large energy but little momentum relative to the charge carriers in the material. Therefore transitions of electrons tend to preserve the momentum (or k), and the dispersion relation is:

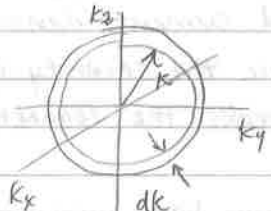
$$\hbar\omega = E_g + \frac{\hbar^2 k^2}{2m_c} + \frac{\hbar^2 k^2}{2m_v}$$

$$\hbar\omega = E_g + \frac{\hbar^2 k^2}{2} \left(\frac{1}{m_c} + \frac{1}{m_v} \right)$$

↑ photon
electron
reduced mass
 $\frac{1}{m_r}$



The joint density of states is calculated using the reduced effective mass:



$$dN = 4\pi k^2 dk \times \left(\frac{2\pi}{L} \right)^{-3} \times 2 \leftarrow \text{spin}$$

$$dN = \frac{V}{\pi^2} k^2 dk$$

Electron dispersion: $E = \frac{\hbar^2 k^2}{2m_r}$ $dE = \frac{\hbar^2 k}{m_r} dk$

$$\Rightarrow \frac{dN}{dE} = \frac{V}{\pi^2} \left(\frac{2m_r E}{\hbar^2} \right) \frac{m_r}{\hbar^2} \frac{\hbar}{\sqrt{2m_r E}} = \frac{V}{\pi^2} \frac{m_r^{3/2} (2E)^{1/2}}{\hbar^3}$$

The net rate of electronic excitation is:

$$\frac{1}{\tau} = \frac{2\pi}{\hbar^2} |M|^2 \frac{dN}{dE} \left[\underbrace{f_v (1 - f_c)}_{\text{upward transitions}} - \underbrace{f_c (1 - f_v)}_{\text{downward transitions}} \right]$$

$f_{c,v}$ = Fermi-Dirac occupancy

$$= \frac{2\pi}{\hbar^2} |M|^2 \frac{dN}{dE} (f_v - f_v f_c - f_c - f_c f_v)$$

$$\frac{1}{\tau} = \frac{2\pi}{\hbar^2} |M|^2 \frac{dN}{dE} \left[\frac{1}{e^{(E_v - E_{Fv})/kT} + 1}} - \frac{1}{e^{(E_c - E_{Fc})/kT} + 1}} \right] \rightarrow \alpha$$

- This rate directly gives the absorption coefficient α .

The sign of the absorption coefficient is determined by the bracketed term.

- Gain occurs if: $\frac{1}{e^{(E_v - E_{Fv})/kT} + 1}} - \frac{1}{e^{(E_c - E_{Fc})/kT} + 1}} < 0$

$$\Rightarrow E_c - E_{Fc} < E_v - E_{Fv}$$

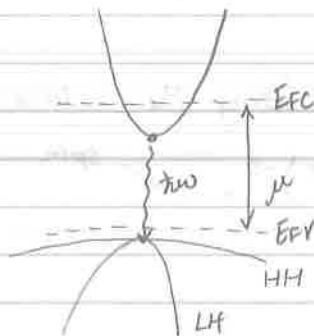
$$E_c - E_v < E_{Fc} - E_{Fv}$$

Since $E_g = E_c - E_v$ and $\mu = E_{FC} - E_{FV}$, the gain condition becomes:

$$E_g = \hbar\omega < \mu$$

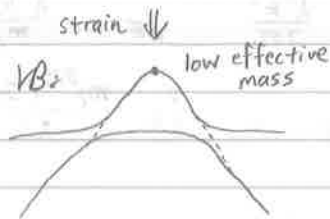
This is the Bernard Duroufour condition. If this is satisfied, there is sufficient population inversion to provide gain.

- One difficulty is that there is asymmetry in the effective mass of electrons and holes, so the Fermi levels are not symmetric about midgap.



- Leads to large electron density at threshold
 - Large Auger recombination
 - Large threshold current density

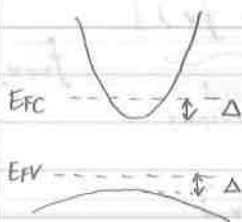
- It is possible to reduce the severity of this issue using strain, which breaks the degeneracy at the valence band minimum.



- Holes at the valence band minimum tend to be light holes under strain, making the bands more symmetric

(aka transparency)

- Consider a quantum well at threshold, with $m_c < m_v$:



Electron density:

$$n_T = \int_0^{\infty} f_c(E) \left(\frac{dN}{dE} \right)_c dE$$

$$= \int_0^{\infty} \frac{1}{1 + e^{(E_c - \Delta)/kT}} \frac{m_c}{\pi \hbar^2} dE$$

$$\Rightarrow n_T = \frac{m_c}{\pi \hbar^2} kT \ln(1 + e^{\Delta/kT}) \approx \frac{m_c}{\pi \hbar^2} \Delta$$

Similarly, the hole density is:

$$p_T = \int_0^{\infty} \frac{1}{1 + e^{(E_v + \Delta)/kT}} \frac{m_v}{\pi \hbar^2} dE \approx \frac{m_v}{\pi \hbar^2} kT e^{-\Delta/kT}$$

At transparency, $n_T = p_T$ is required. If $\frac{m_c}{m_v} = \frac{1}{6}$, $\Delta \approx 1.43 kT$ and the carrier density at threshold is

$$n_T \approx 10^{12}/\text{cm}^2 \text{ (2D)} \rightarrow$$

$$n_T \approx 2 \times 10^{18}/\text{cm}^2 \text{ (3D)}$$

This leads to large Auger recombination
Thus, increasing $\frac{m_c}{m_v}$ improves laser efficiency

⑦ Zero-point Fluctuations and Spontaneous Emission

- Quantum harmonic oscillator: prelude to discussion of second quantization. Consider an electron (mass m) in a potential well $V = \frac{1}{2} m \omega_0^2 x^2$. The Hamiltonian is

$$\mathcal{H} = \frac{1}{2m} p^2 + \frac{1}{2} m \omega_0^2 x^2$$

- Introduce new variables:

$$a^\dagger \equiv \sqrt{\frac{m\omega_0}{2\hbar}} x - i \sqrt{\frac{1}{2m\hbar\omega_0}} p, \quad a \equiv \sqrt{\frac{m\omega_0}{2\hbar}} x + i \sqrt{\frac{1}{2m\hbar\omega_0}} p$$

$$\text{Then: } x = \sqrt{\frac{\hbar}{2m\omega_0}} (a^\dagger + a), \quad p = i \sqrt{\frac{m\hbar\omega_0}{2}} (a^\dagger - a)$$

- Rewrite the Hamiltonian:

$$\begin{aligned} \mathcal{H} &= \frac{1}{2m} \left(-\frac{m\hbar\omega_0}{2} (a^\dagger - a)^2 \right) + \frac{1}{2} m \omega_0^2 \frac{\hbar}{2m\omega_0} (a^\dagger + a)^2 \\ &= -\frac{\hbar\omega_0}{4} (a^{\dagger 2} - a^\dagger a - a a^\dagger + a^2) + \frac{\hbar\omega_0}{4} (a^{\dagger 2} + a^\dagger a + a a^\dagger + a^2) \end{aligned}$$

$$\mathcal{H} = \frac{\hbar\omega_0}{2} (a a^\dagger + a^\dagger a) = \hbar\omega_0 \left(a^\dagger a + \frac{1}{2} \right)$$

Easy to derive that $[a, a^\dagger] = 1$

- Now call $\hat{N} \equiv a^\dagger a$ the number operator with eigenvalue n

$$\hat{N} |n\rangle = n |n\rangle$$

↑
number state

↑
number eigenvalue

$$\begin{aligned} \text{Then } \hat{N} a^\dagger |n\rangle &= a^\dagger a a^\dagger |n\rangle = a^\dagger (a^\dagger a + 1) |n\rangle \\ &= a^\dagger (a^\dagger a |n\rangle + |n\rangle) \\ &= a^\dagger (n+1) |n\rangle \end{aligned}$$

$$\hat{N} a^\dagger |n\rangle = (n+1) a^\dagger |n\rangle$$

- $a^\dagger |n\rangle$ has eigenvalue $n+1$. Therefore we say that a^\dagger is the raising operator

$$\text{Similarly, } \hat{N} a |n\rangle = (n-1) a |n\rangle$$

- so a is the lowering operator

• What if $n < 0$?

$$\langle n | \hat{N} | n \rangle = \int \psi_n^* \hat{N} \psi_n d^3\vec{r}$$

$$= \int \psi_n^* \left(\frac{\mathcal{H}}{\hbar\omega_0} - \frac{1}{2} \right) \psi_n d^3\vec{r}$$

$$n = \frac{1}{\hbar\omega_0} \int \psi_n^* \mathcal{H} \psi_n d^3\vec{r} - \frac{1}{2} \geq 0$$

Therefore n cannot be negative!

• The state $n=0$ represents the bottom of the ladder:
 $\hat{N} | n=0 \rangle = 0$. This must mean $\langle 0 | \hat{N} | 0 \rangle = 0$.

$$\Rightarrow 0 = \frac{1}{\hbar\omega_0} \int \psi_0^* \mathcal{H} \psi_0 d^3\vec{r} - \frac{1}{2} = \frac{E_0}{\hbar\omega_0} - \frac{1}{2}$$

$$\Rightarrow E_0 = \frac{1}{2} \hbar\omega_0$$

This is the zero-point energy!

• In general, $\mathcal{H} = \hbar\omega_0 \left(\hat{N} + \frac{1}{2} \right) \Rightarrow \boxed{E_n = \hbar\omega_0 \left(n + \frac{1}{2} \right)}$
 $n = 0, 1, 2, 3, \dots$

- Analogy to electromagnetic fields: second quantization

Consider Maxwell's equations:

$$\vec{\nabla} \times \vec{\mathcal{E}} = - \frac{\partial \vec{B}}{\partial t} \quad \rightarrow \quad \vec{k} \times \vec{\mathcal{E}} = - \frac{\partial \vec{B}}{\partial t}$$

$$\vec{\nabla} \times \vec{B} = \frac{1}{c^2} \frac{\partial \vec{\mathcal{E}}}{\partial t} \quad \rightarrow \quad c^2 \vec{k} \times \vec{B} = \frac{\partial \vec{\mathcal{E}}}{\partial t}$$

Make the comparison to electron position and momentum

Newton's 2nd law $\rightarrow k_0 \vec{x} = - \frac{d\vec{p}}{dt} \quad \leftrightarrow \quad \vec{k} \times \vec{\mathcal{E}} = - \frac{\partial \vec{B}}{\partial t}$

$$\frac{1}{m} \vec{p} = \frac{d\vec{x}}{dt} \quad \leftrightarrow \quad c^2 \vec{k} \times \vec{B} = \frac{\partial \vec{\mathcal{E}}}{\partial t}$$

Express the second pair of equations in terms of the resonance frequency $\omega_0 = \sqrt{k_0/m}$ (electron), $\omega = kc$ (photon):

$$1) \quad k_0 \vec{x} = - \frac{d\vec{p}}{dt} \quad \leftrightarrow \quad \vec{k} \times \vec{\mathcal{E}} = - \frac{\partial \vec{B}}{\partial t}$$

$$2) \quad (\omega_0^2/k_0) \vec{p} = \frac{d\vec{x}}{dt} \quad \leftrightarrow \quad (\omega^2/k) \vec{B} = \frac{\partial \vec{\mathcal{E}}}{\partial t}$$

Thus there is an analogy between the quantum harmonic oscillator and the electromagnetic fields, with

$\vec{\mathcal{E}} \leftrightarrow \vec{x}$, $\vec{B} \leftrightarrow \vec{p}$: the analogy between fields and operators is called second quantization

$n = \#$ photons in the field



- Thus, the energy in the electromagnetic field must be quantized in the same way: $E_n = \hbar\omega (n + \frac{1}{2})$

This implies that even in vacuum there is a finite amount of zero-point energy: $\frac{1}{2}\hbar\omega = E_0$ (not useful for doing work)

- The zero-point electric field inside of a volume V is:

$$\epsilon_0 \langle E_0^2 \rangle = \frac{1}{V} \frac{\hbar\omega}{2} \Rightarrow \boxed{\langle E_0 \rangle = \sqrt{\frac{\hbar\omega}{2\epsilon_0 V}}} \text{ (RMS)}$$

- Quantum noise

- The common expression for Johnson noise in a resistor is $\langle \Delta V \rangle^2 = 4KTR \Delta\nu$ where $\Delta\nu$ is the bandwidth.

But if $\hbar\nu \approx KT$ or $\hbar\nu \gg KT$, the expression is modified to:

$$\boxed{\langle \Delta V \rangle^2 = 4 \left[\frac{1}{2}\hbar\nu + \frac{\hbar\nu}{e^{\hbar\nu/KT} - 1} \right] R \Delta\nu}$$

↑
Zero-point fluctuation (QM)

← Standard Johnson-Nyquist (classical)

- To observe quantum noise, either go to high frequencies or low temperature to satisfy $\hbar\nu \gg KT$

- Spontaneous emission

- Second quantization implies that the electromagnetic field is quantized into number states $n = 0, 1, 2, \dots$ where n is the number of photons and $n = 0$ is the EM ground state
- Transitions from an electronic (e.g. atomic) excited state to lower state will therefore be coupled with upward transitions of the electromagnetic field from the ground state (no photons) to an excited state (≥ 1 photons)
 - Therefore, vacuum fluctuations in the electromagnetic field give rise to electronic transitions: this is spontaneous emission
 - The infinitely many directions in which a photon can be emitted means this is an irreversible process

- Methods of calculating the spontaneous emission rate

1) Detailed balancing

- The electromagnetic energy density is $\epsilon \mathcal{E}^2$, and the power flux is $\frac{c}{n} \times \epsilon \mathcal{E}^2 = \frac{c}{n} \times n^2 \epsilon_0 \mathcal{E}^2 = c n \epsilon_0 \mathcal{E}^2$
- Detailed balance dictates that the rate of absorption of this electromagnetic power is equal to the rate of spontaneous emission.

Spontaneous emission arises from:

$$\mathcal{E}_p \cos \omega t = \underbrace{\frac{1}{2} \mathcal{E}_p e^{i\omega t}}_{\text{upward}} + \underbrace{\frac{1}{2} \mathcal{E}_p e^{-i\omega t}}_{\text{downward}} \quad \rightarrow \quad \text{RMS field: } \mathcal{E} = \frac{1}{\sqrt{2}} \mathcal{E}_p$$

- The rate is given by Fermi's golden rule:

$$\text{rate} = \frac{2\pi}{\hbar} |\langle g_x \cdot \frac{1}{2} \mathcal{E}_p \rangle|^2 \frac{dN}{dE}$$

- Now impose detailed balance,

absorption coefficient

$$\rightarrow \alpha \times c n \epsilon_0 \mathcal{E}^2 = \left(\frac{2\pi}{\hbar} |\langle g_x \cdot \frac{1}{2} \mathcal{E}_p \rangle|^2 \frac{dN}{dE} \right) \hbar \omega$$

$$= \frac{1}{3} \frac{2\pi}{\hbar} |\langle g_x \rangle|^2 \left(\frac{\mathcal{E}_p}{2} \right)^2 \frac{dN}{dE} \hbar \omega$$

$$\alpha c n \epsilon_0 \mathcal{E}^2 = \frac{1}{3} \frac{2\pi}{\hbar} \left(\frac{\mathcal{E}^2}{2} \right) |\langle g_x \rangle|^2 \frac{dN}{dE} \hbar \omega$$

$$\alpha = \frac{\pi}{c n} \frac{\omega |\langle g_x \rangle|^2}{3 \epsilon_0} \frac{dN}{dE}$$

- Integrate over the spectrum, using the electromagnetic density of states:

$$\begin{aligned} \mathcal{L} &= \int \alpha \frac{8\pi n^2 \nu^2 d\nu}{c^2} \frac{1}{e^{h\nu/kT} - 1} \\ \text{Spontaneous emission } \left(\frac{\# \text{ photons}}{\text{cm}^3 \cdot \text{s}} \right) &\approx \int \frac{\pi}{c n} \frac{\omega |\langle g_x \rangle|^2}{3 \epsilon_0} \frac{dN}{dE} \frac{8\pi n^2}{c^2} \left(\frac{\omega}{2\pi} \right)^2 d\nu \cdot e^{-h\nu/kT} \\ &= \int \frac{2}{3} \frac{\omega^3 |\langle g_x \rangle|^2}{\epsilon_0 c^3} \cdot n \left(\frac{d\nu}{dE} \right) e^{-h\nu/kT} dN \end{aligned}$$

- * Consider a two-level system, so $dN = 1$ (eliminate integral). Also, ignore the exponential factor. This is dubious...

$$\mathcal{L} = \frac{2}{3} \frac{n \omega^3}{\epsilon_0 c^3} |\langle g_x \rangle|^2 \cdot \frac{1}{2\pi\hbar}$$

$$\Rightarrow \boxed{\mathcal{L} = \frac{4}{3} \frac{|\langle g_x \rangle|^2 n \omega^3}{4\pi \epsilon_0 \hbar c^3}} \quad \text{between states } i \text{ and } j$$

2) Second quantization method

- If we rigorously go through the second quantization of the electromagnetic fields (operator fields), we find:

dubious →
$$\hat{\mathcal{E}} = -i \sqrt{\frac{\hbar \omega}{2V\epsilon_0}} \times (a_k^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}} - a_k e^{i\mathbf{k}\cdot\mathbf{r}}) \hat{\mathbf{x}} \leftarrow \text{arbitrary polarization}$$

$$\hat{\mathcal{E}} = -\frac{i}{n} \sqrt{\frac{\hbar \omega}{2V\epsilon_0}} \times (a_k^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}} - a_k e^{i\mathbf{k}\cdot\mathbf{r}}) \hat{\mathbf{x}}$$

- Again use Fermi's golden rule to find the transition rate

$$\mathcal{L} = \frac{2\pi}{\hbar} |\langle 1, k | g \hat{\mathbf{x}} \cdot \hat{\mathcal{E}} | 0, n \rangle|^2 \frac{dN}{dE}$$

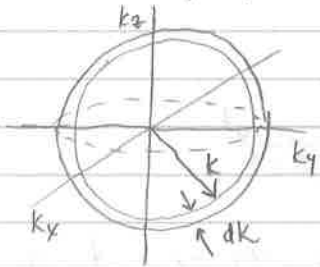
↑
↑
 final state: 1 photon in EM field initial state: 0 photons
 Ground electronic state K Excited electronic state n

- Insert expression for \mathcal{E} :

$$\mathcal{L} = \frac{2\pi}{\hbar} |\langle 1, k | g \mathbf{x} (a^\dagger - a) | 0, n \rangle|^2 \frac{\hbar \omega}{n^2 2V\epsilon_0} \cdot \frac{dN}{dE} \cdot \frac{1}{3}$$

↙ polarization averaging

The density of electromagnetic modes is:



$$dN = \left[4\pi k^2 dk / \left(\frac{2\pi}{L}\right)^3 \right] \times 2$$

↙ k-space volume per state ↘ polarization

$$dN = 4\pi k^2 dk \cdot \frac{V}{4\pi^3}$$

Use $\omega = \frac{c}{n} k$, $E = \hbar \omega$

$$\Rightarrow dN = 4\pi \left(\frac{n\omega}{c}\right)^2 \frac{n}{c} \frac{dE}{\hbar} \frac{V}{4\pi^3}$$

$$\frac{dN}{dE} = \frac{n^3 \omega^2 V}{\hbar c^3 \pi^2} \leftarrow \text{optical DOS}$$

- The spontaneous emission rate is:

$$\mathcal{L} = \frac{2\pi}{\hbar} |g \chi_{nk}|^2 \frac{\hbar \omega}{2n^2 V \epsilon_0} \cdot \frac{n^3 \omega^2 V}{\hbar c^3 \pi^2} \cdot \frac{1}{3}$$

$$\mathcal{L} = \frac{4}{3} \frac{n\omega^3}{4\pi \hbar \epsilon_0 c^3} |g \chi_{nk}|^2$$

This is the same answer!

$$\mathcal{L} = \frac{4}{3} \frac{|g \chi_{nk}|^2 n \omega^3}{4\pi \epsilon_0 \hbar c^3}$$

3) Dipole fluctuations: simply start from the movement of charge

The general expression for the retarded vector potential is:

$$A(\vec{x}) = \int \frac{\mu_0 J(\vec{x}') d^3 \vec{x}'}{4\pi |\vec{x} - \vec{x}'|} \quad \text{current at } \vec{x}', \text{ observe potential at } \vec{x}$$

where $\int J d^3 \vec{x}' = \int \rho v d^3 \vec{x}' = (\int \rho d^3 \vec{x}') v = qv$ for a single electron

• Therefore the potential from an accelerating electron is

$$A = \frac{\mu_0 q v}{4\pi r} \xrightarrow{\text{Coulomb gauge}} \mathcal{E} = -\frac{\partial A}{\partial t} = \frac{\mu_0 q \dot{v}}{4\pi r}$$

↑ distance from electron
electric field from accelerating electron

• Radiated power is: $\frac{P_{\text{rad}}}{\text{area}} = c n \epsilon_0 \mathcal{E}^2 = \frac{c \epsilon_0 \mu_0^2 q^2 \dot{v}^2}{(4\pi)^2 r^2} \times \sin^2 \theta \leftarrow \text{dipole directivity}$

Integrate over sphere of area $4\pi r^2$:

Eli claims $\sin^2 \theta$ averages to this...

$$P_{\text{rad}} = 4\pi r^2 \frac{c \epsilon_0 \mu_0^2 q^2 \dot{v}^2}{\epsilon_0 (4\pi)^2 r^2} \times \frac{2}{3}$$

$$= \frac{m q^2 \dot{v}^2}{\epsilon_0 c^3 4\pi} \cdot \frac{2}{3} = \frac{2}{3} \frac{m q^2 \ddot{x}^2}{4\pi \epsilon_0 c^3} = \frac{2}{3} \frac{m \omega^4 \langle |g_x|^2 \rangle}{4\pi \epsilon_0 c^3}$$

• Spontaneous emission rate is:

$$\mathcal{L} = \frac{P_{\text{rad}}}{\hbar \omega} \Rightarrow \mathcal{L}_{\text{DF}} = \frac{2}{3} \frac{\langle |g_x|^2 \rangle m \omega^3}{4\pi \epsilon_0 \hbar c^3}$$

* This differs from the previous results by a factor of 2!

To account for the missing factor, we must realize that the ground state is a fluctuating dipole, which must radiate.

• The Zero-point radiation must be absorbed, and leads to additional spontaneous emission ("stimulated" by zero-point radiation)

$$\begin{aligned} \# \text{ Absorbed photons / time / area} &= \alpha \cdot \frac{n}{n} \cdot \text{energy density} \cdot \frac{1}{\hbar \omega} \\ &= \left(\frac{\pi \omega}{\epsilon_0 n} \frac{1}{3} \frac{\langle |g_x|^2 \rangle}{dE} \right) \cdot \frac{c}{n} \cdot \frac{\hbar \omega}{2V} \cdot \frac{1}{\hbar \omega} \end{aligned}$$

↑ see method 1
↑ see method 2
↑ zero-point energy density

By detailed balance, this absorption gives rise to spontaneous emission at the same rate:

$$L_{zp} = \frac{\pi \omega}{c n \epsilon_0} \frac{|\langle g_x \rangle|^2}{3} \frac{n^3 \omega^2 V}{\hbar c^3 \pi^2} \cdot \frac{c}{n} \frac{\hbar \omega}{2V} \frac{1}{\hbar \omega}$$

$$= \frac{2}{3} \frac{n \omega^3}{c^3 \hbar \epsilon_0} \frac{1}{4\pi} |\langle g_x \rangle|^2 = \frac{2}{3} \frac{|\langle g_x \rangle|^2 n \omega^3}{4\pi \epsilon_0 \hbar c^3}$$

Thus, the combined rate of spontaneous emission is:

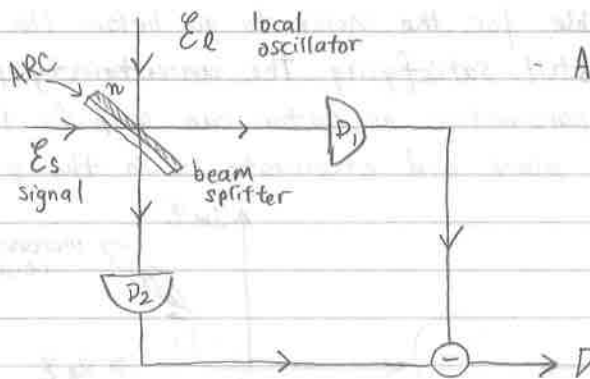
$$L = L_{DF} + L_{zp} \Rightarrow \boxed{L = \frac{4}{3} \frac{|\langle g_x \rangle|^2 n \omega^3}{4\pi \epsilon_0 \hbar c^3}}$$

↑ "classical" ↑ quantum

This is consistent with the other methods

- Coherent Detection: this is a way of detecting very small electric field, including vacuum fluctuations

• Consider a double balanced mixer:



At the detectors, the power is

$$D_1 \sim \frac{|E_s - E_l|^2}{2}$$

$$D_2 \sim \frac{|E_s + E_l|^2}{2}$$

Beamsplitter:

There is a 180° phase change in E_l due to reflection. There is no such sign change on E_s due to an anti-reflection coating

The final signal is:

$$D = D_2 - D_1 = \frac{|E_s + E_l|^2}{2} - \frac{|E_s - E_l|^2}{2}$$

$$= \frac{E_s^2 + 2E_s E_l \cos \phi + E_l^2}{2} - \frac{E_s^2 - 2E_s E_l \cos \phi + E_l^2}{2}$$

$$\text{Output: } \boxed{D_2 - D_1 = 2 E_s E_l \cos \phi}$$

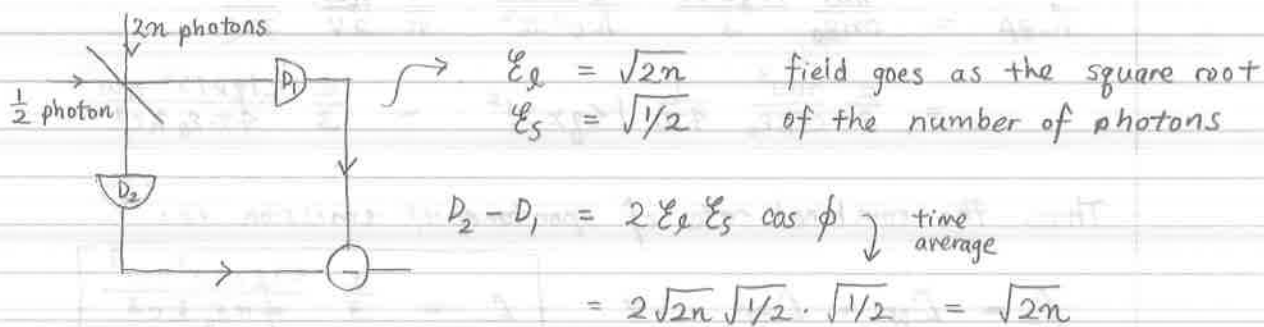
signal
(weak)

local oscillator
(strong, controlled)

relative phase

* A weak signal E_s can be made readily detectable with a large local oscillator field E_l

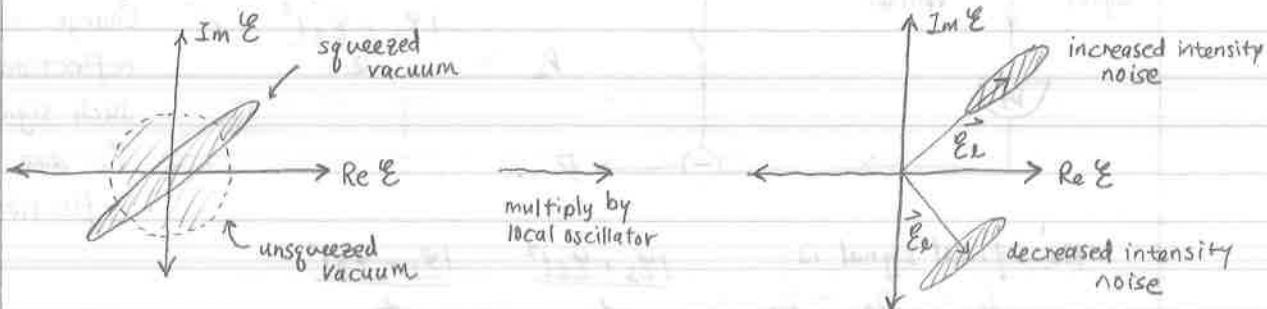
- Consider the case where we do not send a signal \mathcal{E}_s into the signal port. In this case it is the zero-point fluctuation that mixes with the local oscillator



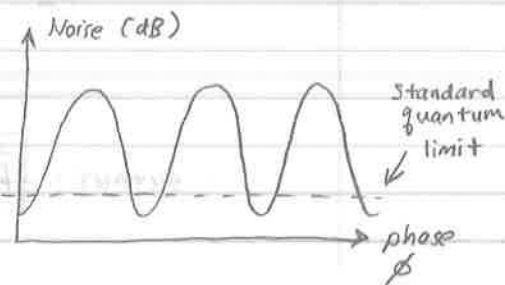
* Even if the local oscillator ($2n$ photons) has no shot noise, we still expect a noise at the output of $\sqrt{2n}$ photons as the zero-point noise enters the system

* The noise of $\sqrt{2n}$ defines the standard quantum limit of noise in an optical signal

- Squeezed vacuum: it is possible for the noise to go below the standard quantum limit while still satisfying the uncertainty principle. As covered in Section 4, a parametric oscillator can amplify the field along one axis of the complex plane and attenuate it in the other



- In changing the phase of the local oscillator, the quantum noise in the field intensity (i.e. amplitude uncertainty) can go above and below the standard quantum limit



- In a detection scheme such as the double balanced mixer, noise suppression is further limited by:

- Quantum efficiency of detectors
 - Wavefront matching (imperfect transmission)
 - Absorption processes - Reflections
- couples unsqueezed vacuum into system

- Expressing the electromagnetic field in terms of photon number states

- A coherent state is one that has minimum uncertainty in the components of the electromagnetic field

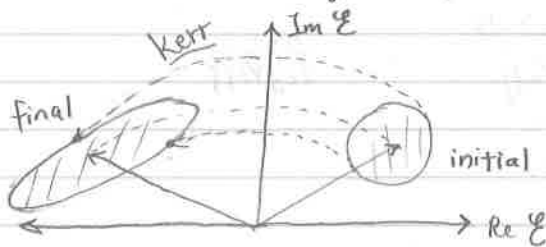
$$|\alpha\rangle = e^{-\frac{1}{2}|\alpha|^2} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle$$

↑ coherent state field
 ← Photon number state, or Fock state

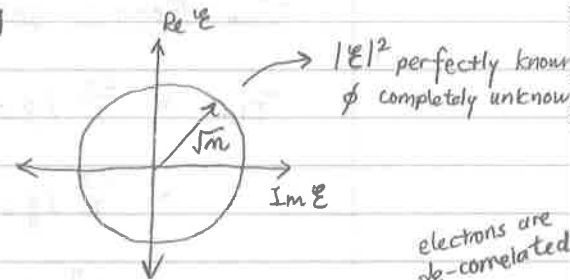
- Therefore, a coherent state is a superposition of infinitely many excited photon states
- The mean number of photons $\langle \alpha | n | \alpha \rangle = |\alpha|^2$
 Variance in photon number is $\langle \Delta n^2 \rangle = |\alpha|^2$
 \Rightarrow Obeys Poisson statistics!
- The coherent state with amplitude $|\alpha|$ is a displaced version of the ground photon number state $|0\rangle$ in the complex plane

• A squeezed state is obtained from applying the squeeze operator \hat{S} to the vacuum state $|0\rangle$, then displacing it by the field amplitude

- The squeezed state is a superposition of only the even-numbered Fock states $|n\rangle$
- Can be generated with a parametric amplifier
- Can also be generated via the Kerr effect

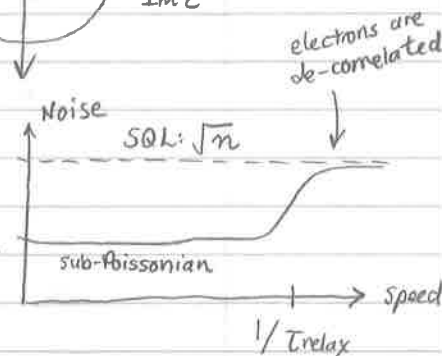


\rightarrow Medium imparts an intensity-dependent phase shift to the beam, which causes squeezing



- A pure number state can be considered a squeezed state, but may not be at the limit of uncertainty

- Eli claims that light emission with noise below the standard quantum limit \sqrt{n} is possible if fed by metallic resistors, which have sub-Poissonian shot noise if the emission is not fast enough to de-correlate the electrons



sub-Poissonian shot noise

$$(\Delta i)^2 = 2 \left(\frac{\lambda}{L} \right) q i \Delta f$$

Purcell effect

- The Purcell effect (first articulated in 1946) is conventionally defined as the enhancement of a molecule's rate of spontaneous emission by the effect of its optical environment
 - However, it may in fact be a much more general property of circuits

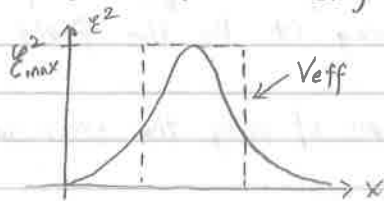
- In free space, the spontaneous emission rate has been derived to be:

$$\frac{1}{\tau_{sp}} = \frac{4}{3} \frac{|g \times|^2 \omega^3}{4\pi \epsilon_0 \hbar c^3}$$

Is it different inside a cavity?

- Consider the zero-point field in a cavity. The total energy is half of a photon: $\int \epsilon_0 \mathcal{E}^2(x) dV = \frac{1}{2} \hbar \omega$

- The peak field in the cavity is required to calculate the spontaneous emission rate using Fermi's golden rule. To obtain this value, define



the effective volume:

$$V_{eff} \equiv \frac{\int \epsilon_0 \mathcal{E}^2(x) dV}{\epsilon_0 \mathcal{E}_{max}^2}$$

This means that a box with size V_{eff} in which the field is uniformly \mathcal{E}_{max} contains all the energy in the cavity.

Thus:

$$\mathcal{E}_{max}^2 = \frac{\int \epsilon_0 \mathcal{E}^2(x) dV}{\epsilon_0 V_{eff}} = \frac{\frac{1}{2} \hbar \omega}{2 \epsilon_0 V_{eff}}$$

- Then Fermi's golden rule gives:

$$\frac{1}{\tau_{sp}} = \frac{2\pi}{\hbar} |g \times \mathcal{E}_{max}|^2 \frac{dN}{dE} \quad \downarrow \text{bandwidth of cavity}$$

$$= \frac{2\pi}{\hbar} |g \times|^2 \frac{\frac{1}{2} \hbar \omega}{2 \epsilon_0 V_{eff}} \cdot \frac{1}{\hbar \Delta \nu}$$

$$= \frac{2\pi^2}{\epsilon_0 V_{eff}} |g \times|^2 \frac{1}{\hbar} \frac{V}{\Delta \nu} = \frac{\pi}{\epsilon_0 \hbar} |g \times|^2 \frac{Q}{V_{eff}} = \frac{1}{\tau_{sp}}$$

Spontaneous emission rate in a cavity

- The resulting enhancement factor is:

$$F_p = \frac{1/T_{sp, cavity}}{1/T_{sp, free space}} = \frac{\left(\frac{\pi}{\epsilon_0 \hbar} |g x|^2 \frac{Q}{V_{eff}} \right)}{\left(\frac{4}{3} \frac{|g x|^2}{4\pi \epsilon_0} \frac{\omega^3}{\hbar c^3} \right)}$$

$$= \frac{\pi}{\epsilon_0 \hbar} \frac{Q}{V_{eff}} \cdot \frac{3}{4} \frac{4\pi \epsilon_0 \hbar c^3}{\omega^3}$$

$$F_p = \frac{3\pi^2 c^3}{\omega^3} \frac{Q}{V_{eff}} = 3\pi^2 c^3 \frac{\lambda^3}{8\pi^3 c^3} \frac{Q}{V_{eff}}$$

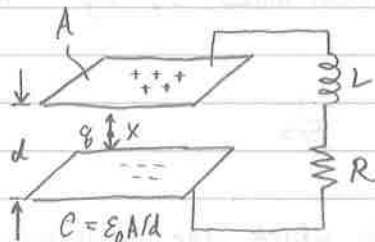
$$\Rightarrow \text{Purcell factor} = \frac{3}{8\pi} \left(\frac{\lambda^3}{V_{eff}} \right) Q$$

This is close, but not precisely equal to Purcell's originally published enhancement factor, which is:

$$\boxed{\text{Purcell factor} = \frac{3}{4\pi^2} \left(\frac{\lambda^3}{V} \right) Q}$$

The error (by a factor of $\frac{\pi}{2}$) is probably due to the definition of effective volume V_{eff} in the quantum mechanical treatment, whereas Purcell's original derivation was classical.

- The Purcell effect is arguably nothing more than a simple circuit property. An LC circuit is an electromagnetic cavity, and the effective volume is the volume between the capacitor plates.



• A single charge q oscillates (with dipole moment qx) between the capacitor plates. If $x < d$, the charge induced on the plates is fractional, in a statistical sense: qx/d

• The Q is $\frac{1}{\omega_0 RC} \Rightarrow \frac{1}{R} = \omega_0 C Q$

• The power lost in the cavity is:

$$P_{loss} = \frac{V^2}{R} = \left(\frac{qx/d}{C} \right)^2 \cdot \omega C Q = \frac{q^2 x^2}{Cd^2} \omega C Q = \frac{(qx)^2}{d^2} \cdot \frac{d}{\epsilon_0 A} \omega C Q$$

$$P_{loss} = \frac{(qx)^2}{\epsilon_0 \cdot V} \omega C Q$$

- The dissipated power can be thought of as the power that is "radiated" into the LC circuit:

$$P = 4\pi \cdot \frac{(q_x)^2 \omega}{4\pi \epsilon_0} \frac{Q}{V}$$

- If we divide this by $\hbar\omega$, we get the same expression for the cavity spontaneous emission rate as we found using Fermi's golden rule (up to a factor of π)
- However, the point is that the Purcell effect is an enhancement of power loss in any type of electromagnetic cavity, not necessarily by spontaneous emission; quantum mechanics is not necessary to describe it!

- Strong coupling: cavity polaritons

- The derivation of $\frac{1}{\tau_{sp}}$ from Fermi's golden rule implicitly assumes that time-dependent perturbation theory is valid. However, in cases of strong coupling between light and matter, time-dependent perturbation theory does not hold.
- The strong coupling condition is the same as the condition for polariton formation (8.2): $\frac{N g^2}{m \epsilon_0} > \frac{1}{\tau^2}$

If we now insert our established correspondence between the classical oscillator model and the quantum mechanical model of light-matter interactions, we have:

$$\frac{N g^2}{m \epsilon_0} \rightarrow \frac{N g^2}{\epsilon_0} \frac{|<11|X|0>|^2 2\omega_0}{\hbar} > \frac{1}{\tau^2}$$

- Now consider a cavity of volume V_{eff} in which the zero-point field amplitude is \mathcal{E}_{zp} . The oscillator density in the cavity can be rewritten as:

$$\begin{array}{l} \# \text{ molecules} \\ \text{in cavity} \end{array} \quad N = \frac{Nm}{V_{eff}} = Nm \left(\frac{\epsilon_0 |\mathcal{E}_{zp}|^2}{\frac{1}{2} \hbar \omega_0} \right)$$

- Assuming $N_m = 1$ for simplicity, the polaron condition becomes:

$$\frac{2\varepsilon_0 |\mathcal{E}_{zp}|^2}{\hbar \omega_0} \frac{g^2 |\langle x \rangle|^2 2\omega_0}{\varepsilon_0 \hbar} = \frac{4g^2}{\hbar^2} |\langle x \rangle|^2 |\mathcal{E}_{zp}|^2 > \frac{1}{\tau^2}$$

Strong coupling: $\boxed{|g \langle x \rangle \mathcal{E}_{zp}| > \frac{\hbar}{2\tau}}$

↑
vacuum Rabi splitting energy: dipole
matrix element of the zero-point field

* The strong coupling condition is satisfied if the vacuum Rabi splitting energy is greater than the damping $\hbar/2\tau$.

- The emission rate in the two regimes are summarized as:

1) Strong coupling: $|g \langle x \rangle \mathcal{E}_{zp}| > \frac{\hbar}{2\tau}$

$$\Rightarrow \frac{1}{\tau_{sp}} = \frac{|g \langle x \rangle \mathcal{E}_{zp}|}{\hbar} \quad (\text{why?})$$

2) Weak coupling: $|g \langle x \rangle \mathcal{E}_{zp}| \leq \frac{\hbar}{2\tau}$: Fermi's golden rule holds

$$\Rightarrow \frac{1}{\tau_{sp}} = \frac{2\pi}{\hbar} |g \langle x \rangle \mathcal{E}_{zp}|^2 \frac{1}{\hbar \Delta V} \leftarrow \text{cavity bandwidth}$$

• Consider the weak coupling case in which the vacuum Rabi splitting matches the bandwidth of the cavity: $\frac{1}{\hbar} |g \langle x \rangle \mathcal{E}_{zp}| = \Delta V$

Then we have: $\frac{1}{\tau_{sp}} = \frac{|g \langle x \rangle \mathcal{E}_{zp}|}{\hbar}$

This is as fast as the strong coupling case.

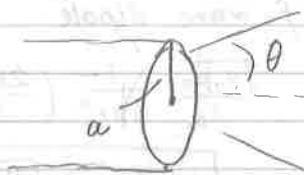
(Note: if $N_m > 1$, the strong coupling condition is modified to $N_m |g \langle x \rangle \mathcal{E}_{zp}| > \hbar/2\tau$)

⑧ Antennas and Metal Optics

- **Antennas** can convert electrical power into electromagnetic radiation, and vice versa. While most commonly used in (and invented for) radio waves, it can also be used at optical frequencies

- Capture-cross section

- Ideal isotropic antenna: $\sigma = \lambda^2 / 4\pi$
 - Directional antenna: $\sigma = \lambda^2 / \Omega$ ← acceptance solid angle ⇒ $P_{in} = \sigma \cdot \text{Intensity}$
- e.g. consider antenna emission



- Emission angle is:

$$\theta = \frac{1/a}{2\pi/\lambda} = \frac{\lambda}{2\pi a}$$

• Solid angle $\Omega = \int_0^{2\pi} d\phi \int_0^\theta \sin\theta' d\theta'$

$$= 2\pi (1 - \cos\theta') \Big|_0^\theta$$

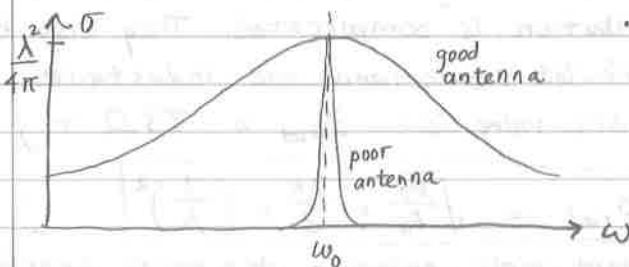
$$= 4\pi \sin^2 \frac{\theta'}{2} \Big|_0^\theta = 4\pi \left(\frac{\lambda}{2\pi a}\right)^2$$

$$\Omega = \frac{1}{\pi} \left(\frac{\lambda}{a}\right)^2$$

Emission Cross-section $\sigma = \pi a^2 = \lambda^2 / \Omega$

By reciprocity, this is also the capture cross-section

- An antenna should have low Q!



- A low Q antenna has a good capture cross-section for a wide band of frequencies: this is desirable
- The penalty for high Q is in the bandwidth, not the capture cross section at the center frequency

- Radiation resistance

- Recall the radiated power from dipole fluctuations (§7):

In vacuum,

$$P_{rad} = \frac{2}{3} \frac{1}{4\pi\epsilon_0 c^3} |Nq\dot{v}|^2 \quad \text{where } N = \# \text{ electrons}$$

$$= \left(\frac{1}{6\pi\epsilon_0 c^3}\right) \left(Nq \frac{dv}{dt}\right)^2 = \left(\frac{1}{6\pi\epsilon_0 c^3}\right) (\omega Nq v)^2$$

$$P_{rad} = \left(\frac{\omega^2}{6\pi\epsilon_0 c^3}\right) \left(a \frac{N}{a} q v\right)^2 = \left(\frac{\omega^2 a^2}{6\pi\epsilon_0 c^3}\right) \times I^2$$

• Radiated power $P_{rad} = \left(\frac{\omega^2 a^2}{6\pi\epsilon_0 c^3} \right) \times I^2$

Radiation resistance $R_{rad} = \frac{\omega^2 a^2}{6\pi\epsilon_0 c^3}$

By reciprocity, the captured power is:

$$\frac{1}{\Omega} \times \text{Intensity} = I^2 R_{rad} = \frac{V^2}{R_{rad}} = P_{in}$$

• For fixed voltage, longer wavelengths produce more power

• Express this quantity for a typical half-wave dipole:

$$R_{rad} = \frac{\omega^2 a^2}{6\pi\epsilon_0 c^3} = \frac{1}{\epsilon_0 c} \frac{\omega^2}{6\pi c^2} a^2 = \frac{\sqrt{\epsilon_0 \mu_0}}{\epsilon_0} \frac{1}{6\pi c^2} \left(\frac{2\pi c}{\lambda} \right)^2 \left(\frac{\lambda}{2} \right)^2$$

$$= \sqrt{\frac{\mu_0}{\epsilon_0}} \frac{1}{6\pi c^2} \frac{4\pi^2 c^2}{4} \Rightarrow R_{rad} = \sqrt{\frac{\mu_0}{\epsilon_0}} \times \frac{\pi}{6} \approx 197 \Omega$$

Impedance of free space: 377Ω

• However, it turns out this is not actually the correct answer!

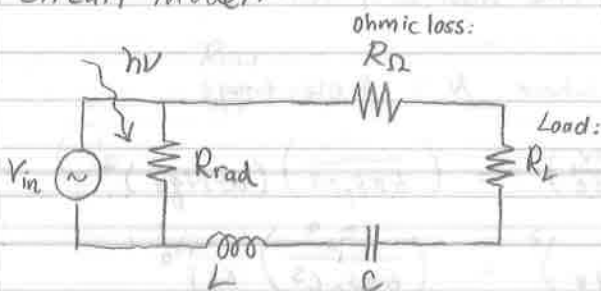


In reality, the current is not uniformly distributed along the antenna, and the true distribution is complicated. They also have distributed capacitance and inductance. A more realistic value is: $Z_{rad} = 73 \Omega + j 42 \Omega$

• For an arbitrary size, $R_{rad} = \sqrt{\frac{\mu_0}{\epsilon_0}} \times \frac{2\pi}{3} \times \left(\frac{a}{\lambda} \right)^2$

• Cannot make antenna too much shorter than λ . Radiation resistance will suffer!

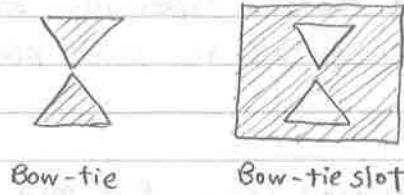
- Circuit model:



- Receiver voltage: $V_{in} = \sqrt{P_{in} \cdot R_{rad}}$

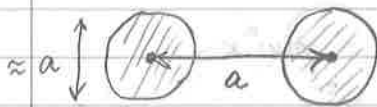
- For maximum transfer of power to the load, impose impedance matching: $R_L = R_{rad} + R_{\Omega}$

- Babinet's principle: a slot antenna with a shape etched from a slab of metal produces the same radiation pattern as a piece of metal with that shape



→ The slot antenna has less ohmic resistance because it has more metal

- Wheeler's Limit: a low Q is desirable, but there is a fundamental lower limit to the achievable Q for a small antenna
- Consider a model of an antenna as two metal spheres:



Capacitance $C \approx \epsilon_0 a$

Stored electrical energy $= q^2 / 2C$
 $= (I/\omega)^2 / \epsilon_0 a$ Dropped a factor of 2.

- The Q is:

$$Q = 2\pi \times \frac{\text{stored energy}}{\text{radiated energy in 1 period}}$$

$$= \frac{2\pi \times (I^2 / \omega^2 \epsilon_0 a)}{\left(\frac{\omega^2 a^2}{6\pi \epsilon_0 c^3} I^2 \cdot \frac{2\pi}{\omega} \right)} = \frac{I^2 \cdot 6\pi \epsilon_0 c^3}{\omega^2 \epsilon_0 a \cdot \omega a^2 I^2}$$

$$= \frac{6\pi c^3}{\omega^3 a^3} = 6\pi c^3 \left(\frac{2\pi c}{\lambda} \right)^3 \frac{1}{a^3} = 6\pi \frac{\lambda^3}{8\pi^3} \frac{1}{a^3}$$

$$\Rightarrow \boxed{Q_{ml} = \frac{3}{4\pi^2} \left(\frac{\lambda}{a} \right)^3}$$

This is a lower bound on antenna Q .
 A small antenna may have a high Q .

- It is possible to deduce R_{rad} from the wheeler limit

$$Q = \frac{1}{\omega R_{rad} C} \Rightarrow R_{rad} = \frac{1}{\omega Q C} = \frac{1}{\omega C} \frac{4\pi^2}{3} \left(\frac{a}{\lambda} \right)^3$$

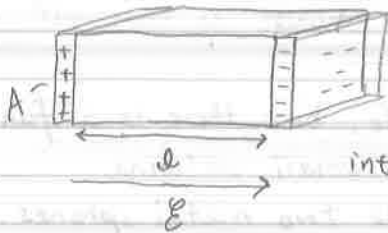
$$R_{rad} = \frac{\lambda}{2\pi c \epsilon_0 a} \frac{4\pi^2}{3} \frac{a^3}{\lambda^3} = \sqrt{\frac{\mu_0}{\epsilon_0}} \frac{2\pi}{3} \left(\frac{a}{\lambda} \right)^2$$

Same prediction from before!

* The same limit can be derived using magnetic dipole radiation (with a factor of 4 difference...)

Kinetic inductance in metals

- Antennas are generally constructed from metals. Now let's consider the motion of electric charge in a metal, especially at optical frequencies. Waves of electrons in metals are called plasmons
- Consider bulk plasmons:



Apply Newton's law, $F = ma$

$$qE = m\ddot{x}$$

The electric field is given by Gauss's law:

integral form: $\epsilon_0 E \cdot A = -Ng \cdot Ax$

$$\Rightarrow E = -\frac{Ngx}{\epsilon_0}$$

We then have:

$$-\frac{Ng^2x}{\epsilon_0} = m\ddot{x} = -m\omega^2x$$

$$\omega^2x = \frac{Ng^2}{m\epsilon_0}x = \omega_p^2x$$

Bulk plasmon frequency is $\omega_p = \sqrt{\frac{Ng^2}{m\epsilon_0}}$

This is the rate of collective charge oscillation in a metal

- Try treating the problem in terms of circuit elements

$$\begin{aligned} qE &= m\dot{v} \\ \text{potential} \rightarrow -q \frac{\partial \phi}{\partial z} &= \frac{m}{N \cdot A \cdot q} \times N \cdot A \cdot q \cdot \dot{v} = \left(\frac{m}{NAq} \right) \cdot \frac{dI}{dt} \end{aligned}$$

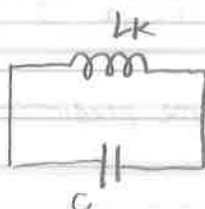
$$\phi = - \underbrace{\frac{m}{Ng^2} \cdot \frac{l}{A}}_{L_k} \cdot \frac{dI}{dt} = -L_k \frac{dI}{dt}$$

This quantity resembles an inductance. It is called the kinetic inductance L_k :

$$L_k = \frac{m}{Ng^2} \frac{l}{A}$$

- If we include the capacitance associated with this charge distribution, we have an LC circuit (ignoring Faraday inductance)

Resonance frequency is:



$$\omega^2 = \frac{1}{L_k C} = \frac{Ng^2 A}{m l} \cdot \frac{l}{\epsilon_0 A} = \frac{Ng^2}{m\epsilon_0} = \omega_p^2$$

Thus, this circuit adequately describes bulk plasmons.

- The kinetic inductance is an inductance that is attributed to the inertial mass of electrons. This results in a delayed response of a metal to very high frequency electromagnetic excitation
- The kinetic inductance can also be derived from the expression for AC conductivity of a metal:

$$\sigma = \frac{Nq^2\tau}{m} \left(\frac{1}{1+i\omega\tau} \right)$$

(DC) \uparrow frictional term

The impedance of a metal wire is:

$$Z = \frac{l}{\sigma} \cdot \frac{\rho}{A} = \frac{m}{Nq^2\tau} (1+i\omega\tau) \frac{\rho}{A}$$

$$Z = \underbrace{\frac{m}{Nq^2\tau} \frac{\rho}{A}}_{\text{Ohmic resistance } R} + i\omega \underbrace{\frac{m}{Nq^2} \frac{\rho}{A}}_{\text{kinetic inductance } L_k}$$

- A metal contains both free and bound charges. In experiments, the effects of these charges cannot be separated, making it problematic to define a relative permittivity ϵ_r . Instead, what is done is assume $\epsilon_r = 1$ (dubious?) and measure a lumped parameter known as the effective permittivity ϵ_m

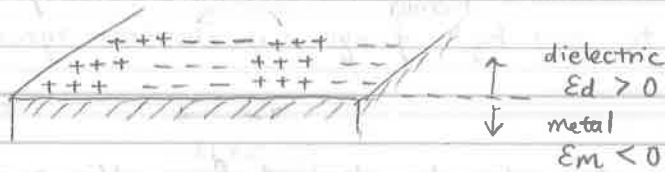
$$\vec{\nabla} \times \vec{H} = \vec{J} + \frac{1}{\epsilon_f \epsilon_0} \frac{\partial \vec{P}}{\partial t} = \sigma \vec{E} + \epsilon_0 (i\omega \vec{E})$$

$$= \left(\frac{1}{\rho} + i\omega \epsilon_0 \right) \vec{E} = i\omega \epsilon_m \epsilon_0 \vec{E}$$

Thus, we have $\epsilon_m = \epsilon_0 + \frac{1}{i\omega \epsilon_0 \rho}$ or $\rho = \frac{i}{\omega \epsilon_0 (\epsilon_m - 1)}$
 This relates the resistivity to the measured "permittivity"

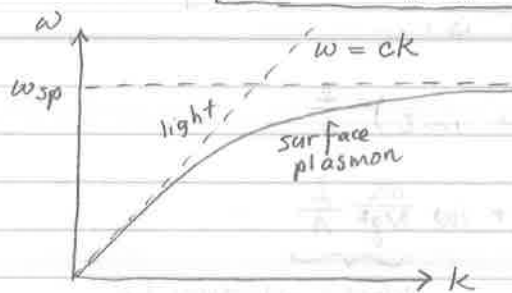
- In the Drude model, $\epsilon_m = 1 - \frac{\omega_p^2}{\omega^2}$. Using this, the kinetic inductance can be derived from ρ .
- The permittivity of a metal will henceforth be taken to mean the effective quantity ϵ_m . Care must be taken not to use this in place of ϵ_r in Maxwell's equations

Surface plasmons



- If it can be shown that for a wave that propagates along the boundary of a metal and a dielectric, Maxwell's equations yield the following dispersion relation:

$$k = \frac{\omega}{c} \sqrt{\frac{\epsilon_m(\omega)}{\epsilon_m(\omega) + \epsilon_d(\omega)}}$$



- At low frequencies, ϵ_m is large (and negative) according to the Drude model.

Therefore: $k = \frac{\omega}{c}$

- What happens at high frequency? Surface plasmons do not exist when the square root argument is negative

The surface plasmon frequency is:

$$0 = \frac{\epsilon_m(\omega)}{\epsilon_m(\omega) + \epsilon_d} \Rightarrow \epsilon_m(\omega) = -\epsilon_d \Rightarrow \omega_p^2 = \omega^2 = -\epsilon_d$$

(If $\epsilon_d = 1$, $\omega_{sp} = \frac{\omega_p}{\sqrt{2}}$)

$$\omega_{sp} = \frac{\omega_p}{\sqrt{1 + \epsilon_d}}$$

Surface plasmon resonance

- Orthogonal to the interface, the surface plasmon is an exponentially decaying wave on both the dielectric and metal side. The extinction coefficient on the metal side is

$$k_m = \sqrt{k^2 - \frac{\omega^2}{c^2} \epsilon_m} = \frac{\omega}{c} \sqrt{\frac{\epsilon_m^2}{1 + \epsilon_m}} \quad (\text{If } \epsilon_d = 1)$$

Therefore, the skin depth of the surface wave is

$$\delta_m = \frac{1}{k_m} = \frac{1}{\frac{\omega}{c} \sqrt{k^2 - (\omega^2/c^2) \epsilon_m}}$$

This is purely a result from Maxwell's equations, and does not depend on collision effects

ϵ_m' = real
 ϵ_m'' = imaginary

- There are multiple types of skin depth associated with surfaces of metals:

Normal incidence waves

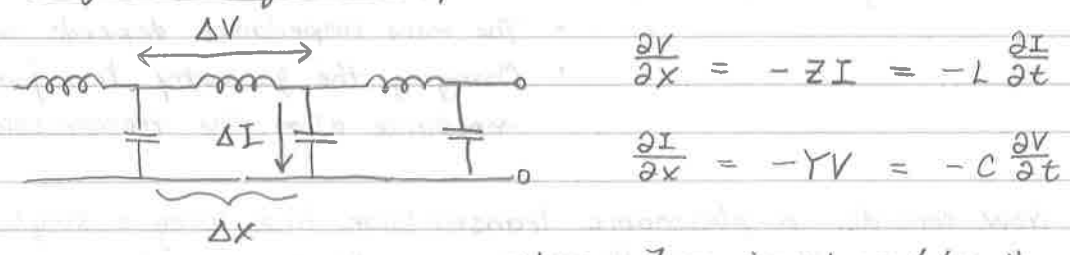
- 1) Collisional skin depth: $\delta_m = \sqrt{\frac{2\rho'(\omega)}{\omega\mu_0}} = \frac{c\sqrt{2\epsilon_m''(\omega)}}{\omega|1-\epsilon_m|}$
 - This is the extinction coefficient of surface current, most relevant in the microwave regime: $\omega\tau \ll 1$
- 2) Collisionless skin depth: $\delta_m = \frac{c}{\omega\sqrt{1-\epsilon_m}} = \frac{c}{\omega p}$
 - This pertains to waves of high frequency, above the collision frequency of electrons: $\omega\tau > 1$ (~ 10 THz)

Surface waves

- 3) Surface wave skin depth: $\delta_m = \frac{1}{\sqrt{k^2 - (\omega^2/c^2)\epsilon_m}}$
 - This is the penetration depth of surface waves.

Plasmonic transmission lines

• It is often proposed that surface plasmons should be used for metal transmission lines. To study this, start with the telegrapher's equations for transmission lines:



where $Z \equiv$ impedance / length
 $Y \equiv$ admittance / length

• The coupled equations can be solved:

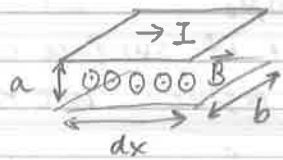
$$\frac{\partial^2 V}{\partial x^2} = -Z \frac{\partial I}{\partial x} = ZYV \Rightarrow V = Ae^{-\sqrt{ZY}x} + Be^{\sqrt{ZY}x}$$

- From this equation we have the propagation constant = \sqrt{ZY}
- We can also relate V to I :

$$\frac{\partial V}{\partial x} = -\sqrt{ZY}V = -ZI \Rightarrow V = \sqrt{\frac{Z}{Y}}I$$

Therefore, the wave impedance is $\sqrt{\frac{Z}{Y}}$

- parallel plate
- Consider the case of a lossless metal transmission line at microwave frequencies (ignore kinetic inductance)
 - The impedance comes from Faraday inductance: $Z = i\omega L$



- Magnetic field $B = \frac{\mu_0 I}{b}$ (Ampere's law)
- By Faraday's law, the generated voltage is:

$$V = -\frac{d}{dt}(B \cdot A) = -\underbrace{(a \, dx)}_{L_f} \frac{\mu_0}{b} \frac{dI}{dt}$$

L_f : Faraday inductance

- The admittance arises from capacitance
- $$C = \epsilon_0 (b \, dx) / a$$

⇒ From this, the transmission line parameters are:

--- Propagation constant

$$k = \sqrt{ZY} = \sqrt{(i\omega L_f')(i\omega C')} = \omega \sqrt{\left(\frac{a}{b} \mu_0\right) \left(\frac{b}{a} \epsilon_0\right)}$$

per unit length

$$k = \omega \sqrt{\epsilon_0 \mu_0} = \frac{\omega}{c}$$

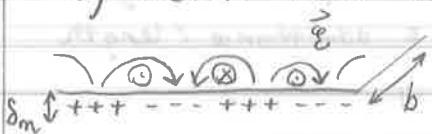
Signal travels at the speed of light!

- Wave impedance

$$\sqrt{\frac{Z}{Y}} = \sqrt{i\omega \left(\frac{a}{b} \mu_0\right) / i\omega \left(\frac{b}{a} \epsilon_0\right)} = \sqrt{\frac{\mu_0}{\epsilon_0}} \cdot \frac{a}{b}$$

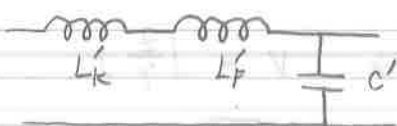
- The wave impedance depends on geometry
- Changing the geometry transforms the wave impedance along the transmission line

- Now consider a plasmonic transmission line using a single piece of metal. There are three components to impedance and



admittance to consider. These can be found to have the following forms (per length):

- Kinetic inductance: $L_k' = \frac{m}{Nq^2} \frac{1}{8\pi b} \cdot \frac{1}{l} = \frac{1}{\epsilon_0 \omega_p^2} \frac{1}{b \, 8\pi}$
- Capacitance: $C' = 2\epsilon_0 k b$
- Faraday inductance: $L_f' = \mu_0 / 2k b$



- Using a circuit model with these parameters, a dispersion relation close to the Maxwell solution (previously shown) can be derived.

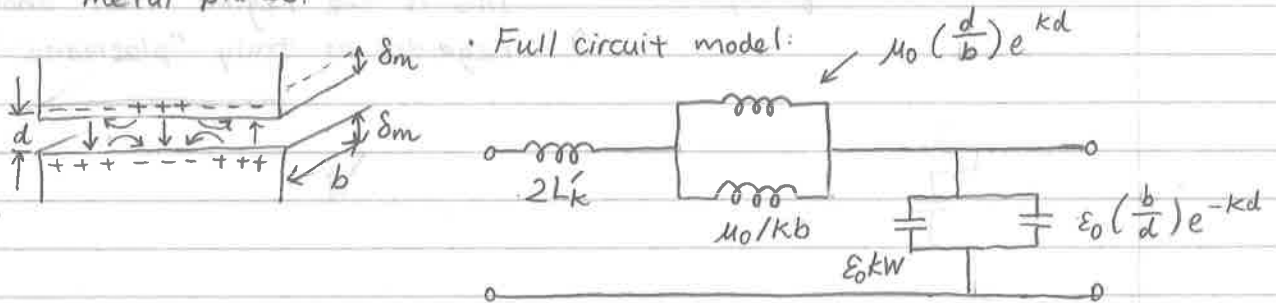
- The parameters of the single-plate plasmonic transmission line are:

- Propagation constant: $k^2 = \omega^2 \left(\frac{1}{c^2} + \frac{2k}{\omega^2(1-\epsilon_m)\delta_m} \right)$

- Wave impedance: $Z = \sqrt{\frac{L'}{C'}} \rightarrow \frac{1}{b} \frac{\lambda_p}{2\pi} \sqrt{\frac{\mu_0}{2\epsilon_0}}$

for $k > \omega_p/c$, when kinetic inductance dominates

- The analysis can also be done for a parallel plate plasmonic transmission line, but is more complicated because of additional capacitance and Faraday inductance between the two metal plates.

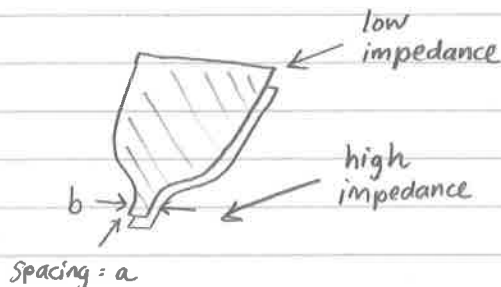


• This is also close to the exact Maxwell solution.

The wave impedance at high frequency is twice that of the previous case:

$$Z = \frac{1}{b} \frac{\lambda_p}{2\pi} \sqrt{\frac{2\mu_0}{\epsilon_0}} \quad (\text{kinetic inductance dominates})$$

• This is the expected behavior: at the nanoscale, where b is very small, the impedance is very high. This suggests the possibility of making a nano-transformer for optical frequency signals: $Z \propto 1/b$

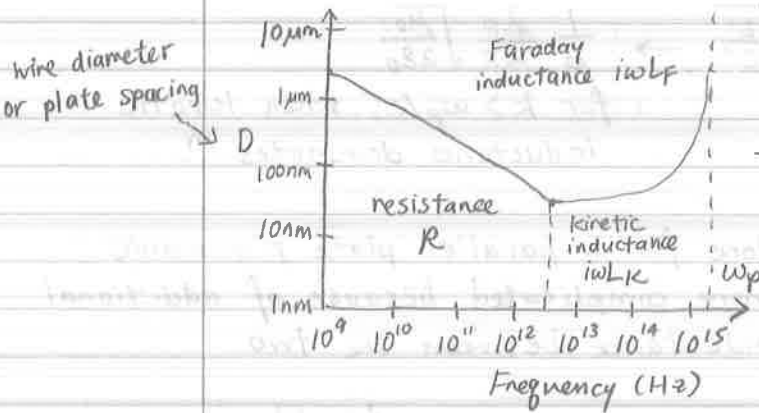


• By tapering the transmission line, the impedance Z dramatically increases:

- Voltage ΔV increases, current ΔI decreases, reducing I^2R resistive losses in the metal

- Increase in voltage is accompanied by an enhancement in the electric field

- For both metal wires and parallel plate metal transmission lines, the following general rule applies to the relative importance of the different circuit components contributing to the impedance:



$$Z = R + i\omega L_F + i\omega L_K$$

- Kinetic inductance becomes the dominant contribution only for very small geometries and very high frequencies (optical frequencies and above)

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This is the regime that should be regarded as truly "plasmonic"