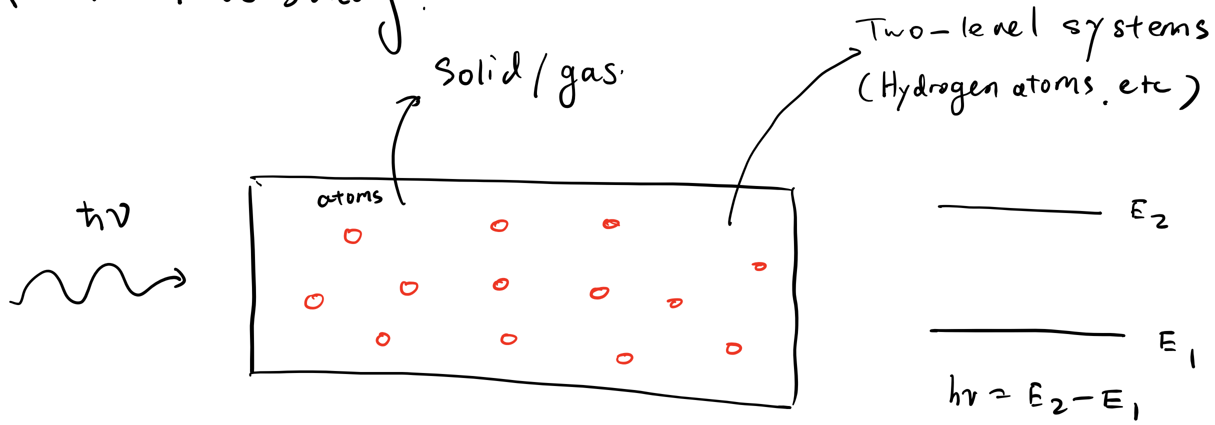
Lecture 13. Interaction of radiation and atomic systems.

Learning objectives

1. Atomic transitions
2. Complex refractive index of atomic systems
3. Homogeneous and inhomogeneous broadening

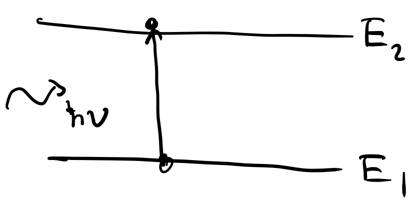
1. Atomic transitions (P211~216. Yariv)

Problem to study:

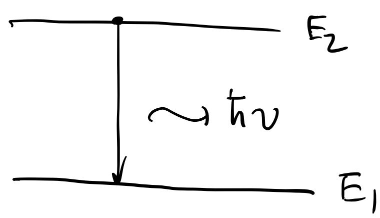


Three different scenarios:

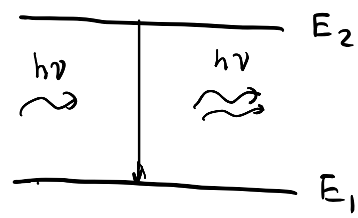
① Absorption



② Spontaneous emission



③ Stimulated emission



How to model this system?

$$\xrightarrow{\hbar\nu} \boxed{\chi(\omega) = \chi'(\omega) - i\chi''(\omega)}$$

induced dipole moment

$p = \alpha E$

polarization $P = N\alpha E$

$= \epsilon_0 \chi E$

Comments:

① $\chi(\omega)$ should be derived by time-dependent pertabation theory in QM, but can be approximated by classical harmonic oscillator model.

② In this case, the 2-level transitions are regarded as "dipoles". They are known as "Rabi oscillators".

Recall classical harmonic osillator model:

equation of motion:

$$m\frac{d^2x}{dt^2} + m\gamma\frac{dx}{dt} + m\omega_0^2 x = -eE$$

where x is the position of electron relative to the atom.

m is the electron mass, $\omega_0 = \frac{E_2 - E_1}{\hbar}$ is oscillation freq. of e^-

. γ is damping coefficient.

Let $E = E_0 e^{i\omega t}$,

$$x = \frac{-eE_0}{m(\omega_0^2-\omega^2+i\gamma\omega)} \exp(i\omega t)$$

induced dipole moment:

$$p = -ex = \frac{e^2}{m(\omega_0^2-\omega^2+i\gamma\omega)} E_0 \exp(i\omega t) = \alpha E$$

So atomic polarizability:

$$\boxed{\alpha = \frac{e^2}{m(\omega_0^2-\omega^2+i\gamma\omega)}}$$

If there are N atoms per unit volume:

$$\tilde{n}^2 = 1+\chi = 1+\frac{N\alpha}{\epsilon_0} = 1+\frac{Ne^2}{m\epsilon_0(\omega_0^2-\omega^2+i\gamma\omega)}$$

where $\chi = \chi' - i\chi'' = \dfrac{Ne^2}{m\epsilon_0(\omega_0^2-\omega^2+i\gamma\omega)}$

when $\chi \ll 1$, $n = 1+\frac{1}{2}\chi = 1+\dfrac{Ne^2}{2m\epsilon_0(\omega_0^2-\omega^2+i\gamma\omega)}$

When $\omega \approx \omega_0$

$$\alpha = \frac{e^2}{2\omega_0 m(\omega_0-\omega+i\gamma/2)}$$

$$\tilde{n}^2 = 1+\chi = 1+\frac{Ne^2}{2\omega_0 m\epsilon_0(\omega_0-\omega+i\gamma/2)}$$

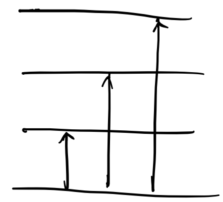
$$\chi = \chi' - i\chi = \frac{Ne^2}{2\omega_0 m\epsilon_0(\omega_0-\omega+i\gamma/2)}$$

$$\tilde{n} = 1 + \frac{1}{2}\chi = 1 + \frac{Ne^2}{4\omega_0 m \epsilon_0 (\omega_0 - \omega + i\gamma/2)} \quad (\chi \ll 1) \quad (1)$$

Comments:

① Here, we only assumed single resonant freq. In real atoms, there are many resonant freqs.

real atoms:



② Using time-dependent theory in QM.

$$\boxed{\alpha = \frac{f_{21} e^2}{2\omega_0 m (\omega_0 - \omega + i\frac{\gamma}{2})}}$$

where γ is the linewidth of the transition, f_{21} is the oscillator strength.

$$f_{21} = \frac{2m\omega_0}{\hbar} |\langle u_2 | x | u_1 \rangle|^2 = \frac{2m\omega_0}{\hbar} \iiint u_2^* x u_1 \, dx \cdot dy \cdot dz,$$

where u_2, u_1 are wavefunctions of eigenstates.

$$\sum_{m=2}^{\infty} f_{m1} = 1$$

2. Complex refractive index and dispersion

Complex refractive index:

$$\tilde{n} = n - ik = 1 + \frac{Ne^2(\omega_0^2-\omega^2)}{2m\epsilon_0\left[(\omega_0^2-\omega^2)^2+\gamma^2\omega^2\right]} - i\,\frac{Ne^2\gamma\omega}{2m\epsilon_0\left[(\omega_0^2-\omega^2)^2+\gamma^2\omega^2\right]}$$

$$k' = \tilde{n}\frac{\omega}{c} = \frac{2\pi}{\lambda}n' = \frac{2\pi}{\lambda}(n-ik)$$

Electric field inside the medium.

$$E = A\exp\left[i(\omega t - k'z)\right] = A\exp\left[i\left(\omega t - \frac{2\pi}{\lambda}(n-ik)\right)\right]$$

$$= A\exp\left[i\left(\omega t - \frac{2\pi}{\lambda}nz\right)\right]\underbrace{\exp\left(-\frac{2\pi}{\lambda}kz\right)}_{\text{decay}}$$

Intensity: $I(z) = I(0)\exp(-\alpha z)$

$$I = E^* \cdot E$$

So attenuation coefficient: $\alpha = \frac{4\pi}{\lambda}\cdot k$

example: $k = 0.0001$, at $\lambda = 500$ nm, $\alpha = 25\ \text{cm}^{-1}$

When $\omega \simeq \omega_0$

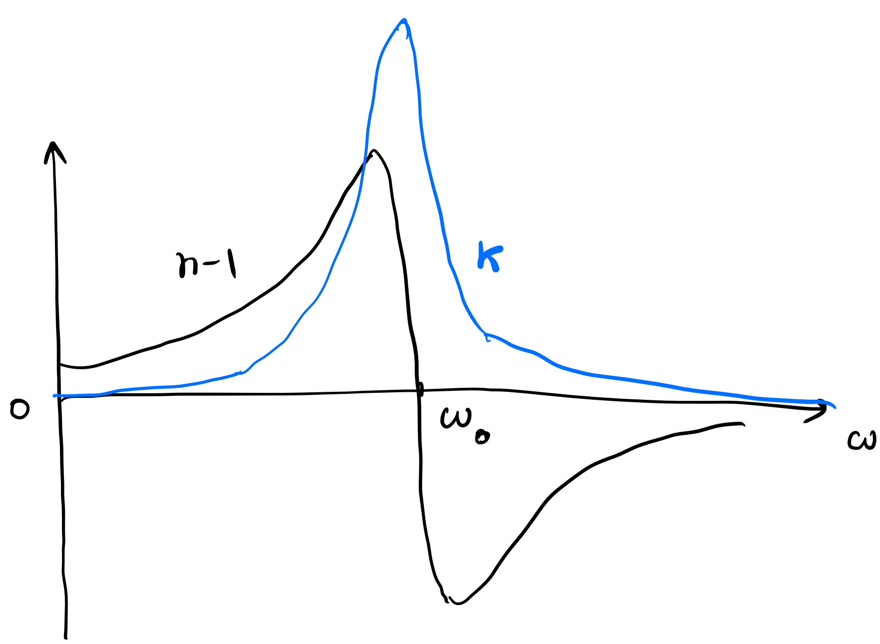
$$\tilde{n} = n - ik = 1 + \frac{Ne^2(\omega_0-\omega)}{4m\omega_0\epsilon_0\left[(\omega_0-\omega)^2+\left(\frac{\Gamma}{2}\right)^2\right]} - i\,\frac{Ne^2(\Gamma/2)}{4m\omega_0\epsilon_0\left[(\omega_0-\omega)^2+(\Gamma/2)^2\right]}$$

$$\chi = \chi' - i\chi'' = \frac{Ne^2(\omega_0-\omega)}{2m\omega_0\epsilon_0\left[(\omega_0-\omega)^2+(\Gamma/2)^2\right]} - i\,\frac{Ne^2(\Gamma/2)}{2m\omega_0\epsilon_0\left[(\omega_0-\omega)^2+(\Gamma/2)^2\right]}$$

$$\xrightarrow{\text{convert } \omega \text{ to } \nu}$$

$$\chi' = \frac{Ne^2(\nu_0-\nu)}{4\pi m\omega_0\epsilon_0\left[(\nu_0-\nu)^2+\left(\frac{\Delta\nu}{2}\right)^2\right]}$$

$$\chi'' = \frac{Ne^2(\Delta\nu/2)}{4\pi m\omega_0\epsilon_0\left[(\nu_0-\nu)^2+\left(\frac{\Delta\nu}{2}\right)^2\right]}$$

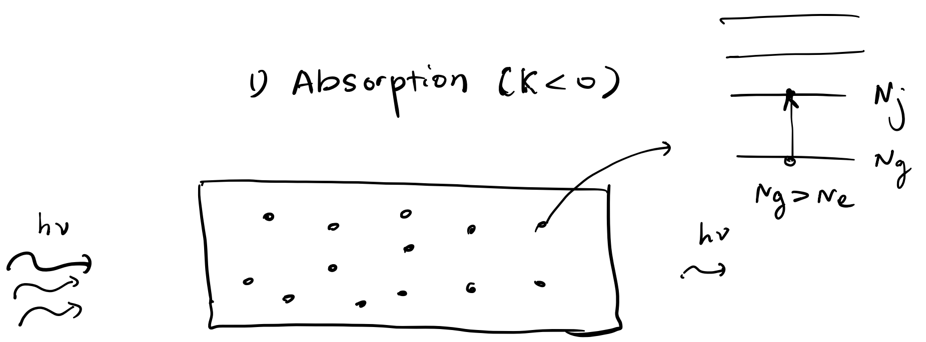


Comments:

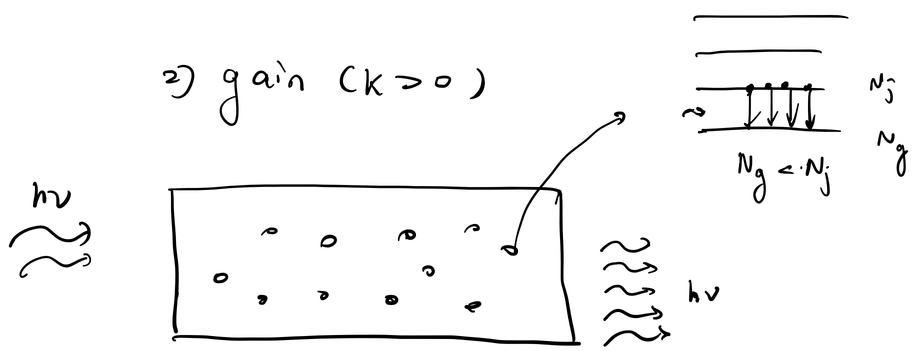
① Strong dispersion around atomic transition

② k can be positive (absorption) or negative (gain)

1) Absorption $(K < 0)$



2) gain $(K > 0)$

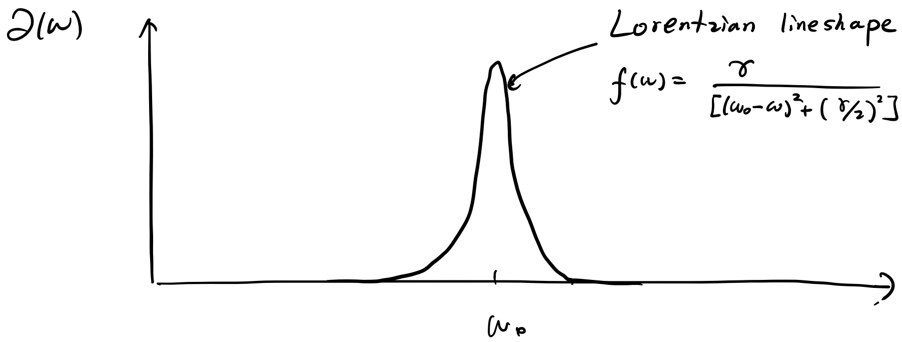


③ Normally, $N_g > N_j$ $(K > 0)$, to realize gain, we need some "pump" to inverse the population $(N_g < N_j)$

3. Homogeneous and inhomogeneous broadening

Absorption coefficient of two-level system:

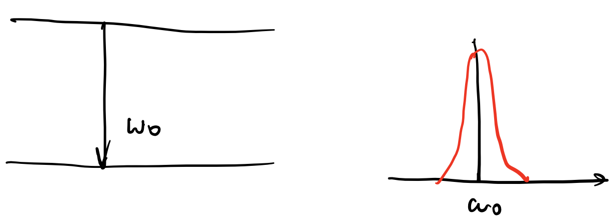
$$\alpha(\omega) = \frac{4\pi}{\lambda} \cdot \frac{Ne^2\gamma}{8m\omega_0\epsilon_0\left[(\omega_0-\omega)^2 + (\gamma/2)^2\right]}$$



Comments

① Absorption spectrum is the same as the spontaneous emission spectrum.

② This means that absorption/radiation is not strictly monochromatic! The absorption/spon. emission spectrum is **broadened** around ω_0



Define $g(\nu) = \frac{1}{2\pi} \frac{\gamma/2\pi}{(\nu-\nu_0)^2 + (\gamma/4\pi)^2}$ as the lineshape function,

and $\int_{-\infty}^{\infty} g(\nu)\, d\nu = 1$. $g(\nu)d\nu$: probability of spon. emission

from level 2 to 1 will resnit in a photon whose freqnency is

between ν and $\nu + d\nu$.

$$g(\nu) = \frac{\Delta\nu}{2\pi\left[(\nu-\nu_0)^2 + \left(\frac{\Delta\nu}{2}\right)^2\right]} \quad , \quad \Delta\nu = \frac{\gamma}{2\pi} = \frac{1}{2\pi\tau} \quad (\text{FWHM})$$

where τ is the lifetime of the emitting state.

<u>Homogeneous broadening</u>: finite lifetime of electrons in upper or lower states, or collisions

$$\text{So} \quad \Delta\nu = \frac{1}{2\pi}\left(\tau_u^{-1} + \tau_l^{-1} + \tau_{co}^{-1}\right)$$

upper lower Collision.

<u>Three possible reasons</u>:

① Spontaneous lifetime of the excited state

② Collision of an atom embeded in a crystal with phonon.

③ Collisions beween atoms.

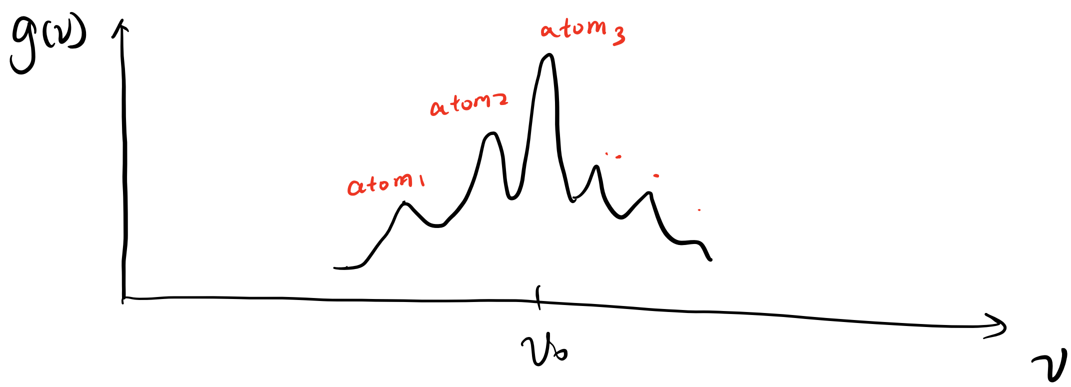
Comments:

① For homogeneous brodening, spread $\delta\nu$ is characteristic of each atom in the sample. $g(\nu)$ describe the response of any atoms. ⇒ Atoms are indistinguishable

② For homogeneous brodening, each atom has a same ν_0

Inhomogeneous brodening:

individual atoms are distinguishable, each having a slightly different transition freq. ν_0

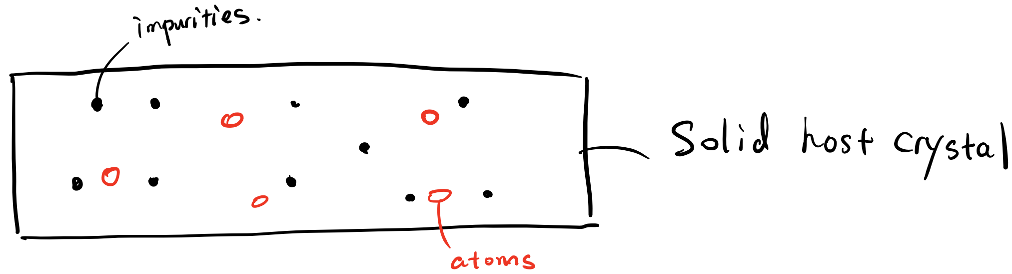


Comments:

① Spectrum of spontaneous emission reflects the spread in each individul transition frequencies, not the broadening due to the finite lifetime of the excited states.

Two possible reasons:

① Impurities or random strain in the host crystal.



② For gaseou atom (or molecule), transition freq. ν is doppler shifted.

$$\nu = \nu_0 + \frac{v_x}{c}\nu_0$$

moving velocity.

transition freq of a stationary atom