Laser Physics 12.
Interaction of light with matter

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Light-matter interactions (summary)

Interactions in a volume V with a selected mode of frequency ν:
spontaneous emission, absorption, stimulated emission

Probability densities of the processes ~ transition cross section $\sigma(\nu)$ [cm²]

$$p_{sp} = \frac{c}{V}\sigma(\nu), \quad p_{ab} = p_{ie} = \frac{c}{V}\sigma(\nu), \quad P_{ab} = P_{ie} = W_i = n\frac{c}{V}\sigma(\nu) \quad [s^{-1}]$$

Strength of interactions, lineshape function:

$$S = \int_0^\infty \sigma(\nu)d\nu \quad [cm^2 s^{-1}], \quad g(\nu) = \frac{\sigma(\nu)}{S}.$$

Total spontaneous emission into all modes: $P_{sp} = \frac{8\pi}{\lambda^2}S \quad [s^{-1}]$

Spontaneous lifetime: $P_{sp} = \frac{1}{t_{sp}}, \quad S = \frac{\lambda^2}{8\pi t_{sp}}, \quad \sigma(\nu) = S\,g(\nu) = \frac{\lambda^2}{8\pi t_{sp}}g(\nu)$

Interaction with a photon beam of frequency ν travelling in a selected direction:

Φ photon-flux density (photons / cm² ·s) $\quad W_i = \Phi\sigma(\nu).$

effective interaction area

S can be determined from measurement, $g(\nu)$?

Light-matter interactions

<u>Line-broadening mechanism</u>

The frequency dependence of light-matter interactions is governed by the normalized lineshape function $g(\nu)$. Materials can be classified into two basically different groups:

homogeneous

All atoms, molecules behave similarly in the light-matter interaction, they have the same individual lineshape function.

inhomogeneous

Group of atoms and molecules behave differently in the light-matter interaction, the whole system can be characterized by an average lineshape function.

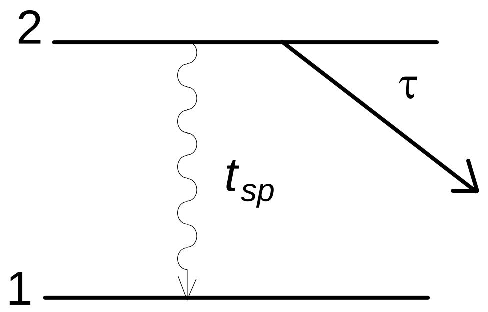
The reality is always a mixture of the two properties!

Light-matter interactions

<u>Homogeneous broadening – lifetime or natural broadening</u>

It is always present, the questions is how much dominant is?

Excited energy levels have finite lifetime. If level 2 is an excited level, its lifetime τ represents the inverse of the rate at which the population of that level decays to level 1 and to all other lower energy levels radiatively (t_{sp}) or nonradiatively, therefore $\tau \leq t_{sp}$.



The population of level 2 decays exponentially, therefore the amplitude of emitted electromagnetic field decays also exponentially

$$E = e^{-t/2\tau} e^{j2\pi\nu_0 t}, \quad E_2 - E_1 = h\nu_0.$$

The energy decays with τ, therefore the factor of ½!!

The spectral dependence can be calculated by the Fourier-transform of the exponentially decaying harmonic function.

Light-matter interactions

Homogeneous broadening – lifetime or natural broadening (cont)

Time dependent field amplitude: $E(t) = e^{-t/2\tau} e^{j2\pi\nu_0 t}, \quad \text{ha} \quad t > 0$

$E(t) = 0, \quad \text{ha} \quad t < 0.$

the Fourier-transform of *f*(t):

$$f(t) \rightarrow F(\nu) = \int_{-\infty}^{\infty} f(t)\, e^{-j2\pi\nu t} dt \rightarrow |F(\nu)|^2 \rightarrow \text{normalizing} \rightarrow \text{g}(\nu)$$

$$g(\nu) = \frac{\Delta\nu / 2\pi}{(\nu - \nu_o)^2 + (\Delta\nu / 2)^2}, \quad \Delta\nu = \frac{1}{2\pi\tau}, \quad g(\nu_0) = \frac{2}{\pi\,\Delta\nu}.$$

$\Delta\nu$ is the FWHM of the Lorentz-function. We could start from the uncertainty principle of Heisenberg:

$$\Delta E\, \Delta t \approx \hbar, \quad \Delta E\, \tau \approx \hbar \rightarrow 2\pi\, \Delta\nu\, \tau \approx 1.$$

Light-matter interactions

Homogeneous broadening – lifetime or natural broadening (cont)

General case: both selected energy levels are excited levels, both have lifetime broadenings:

$$\Delta E_2 = \frac{h}{2\pi\tau_2}, \quad \Delta E_1 = \frac{h}{2\pi\tau_1},$$

the broadening of the transfer is:

$$\Delta E = \Delta E_1 + \Delta E_2 = \frac{h}{2\pi}\left(\frac{1}{\tau_1} + \frac{1}{\tau_2}\right) = \frac{h}{2\pi}\frac{1}{\tau},$$

The reciprocal of the characteristic time τ is the sum of the reciprocal lifetimes and the broadening $\Delta\nu$ can be calculated:

$$\boxed{\frac{1}{\tau} = \frac{1}{\tau_1} + \frac{1}{\tau_2} \quad \rightarrow \quad \Delta\nu = \frac{1}{2\pi}\left(\frac{1}{\tau_1} + \frac{1}{\tau_2}\right).}$$

$$\tau_{typ} \sim 10^{-8}s \quad \Delta\nu_{nat} = \frac{1}{2\pi\tau_{typ}} = \frac{1}{2\pi}10^{8} \sim 16 \text{ MHz}.$$

Light-matter interactions

Homogeneous broadening – lifetime or natural broadening (cont)

$$g(\nu_o)_{Lorentz} = \frac{2}{\pi\,\Delta\nu},$$

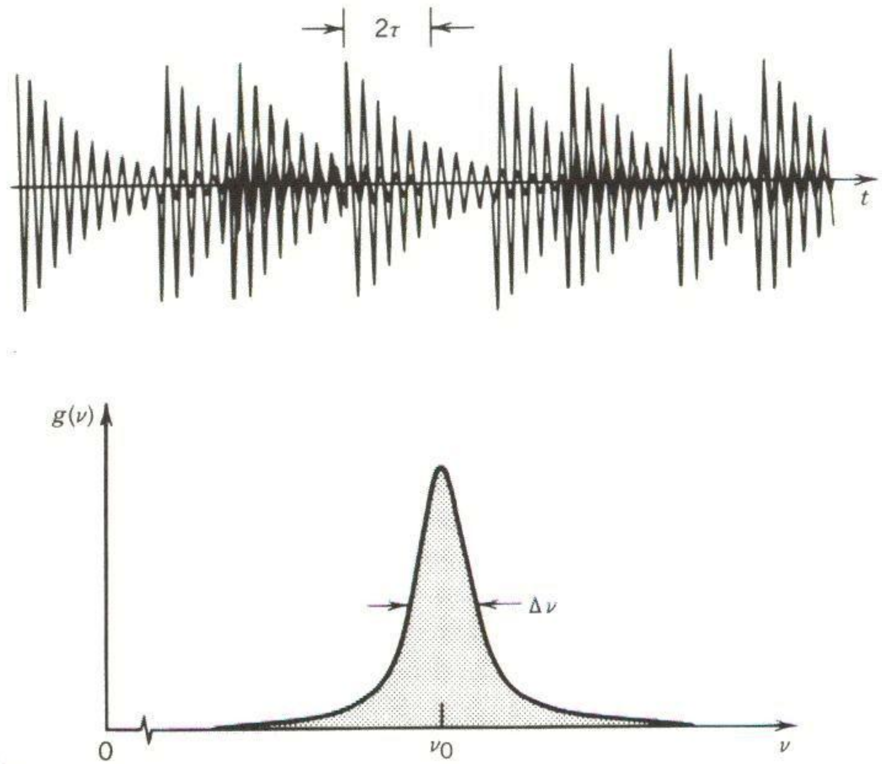
$$\sigma(\nu_o) = \frac{\lambda^2}{2\pi}\frac{1}{2\pi\, t_{sp}\Delta\nu}.$$

In ideal case $\tau_2 = t_{sp}$ and $1/\tau_1 = 0$ (lower level is stationary):

$$\frac{1}{2\pi\, t_{sp}} = \Delta\nu, \quad \sigma(\nu_o) = \frac{\lambda^2}{2\pi}.$$

10^{-11}-10^{-7} cm^2 in 0.1 – 10 µm wavelength range.

$\sigma(\nu_0)$ has a typical order of magnitude of 10^{-20}-10^{-11}cm^2 (small overlap)



Wavepacket emissions at random time and the Lorentz-function

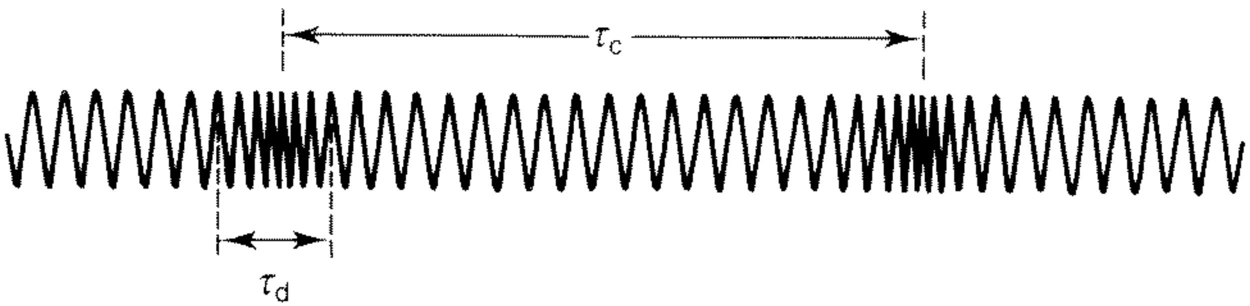
Light-matter interactions

<u>Homogeneous broadening – collision broadening</u>

In gas and fluid can be important with increasing the density of particle (with increasing the pressure). All particle suffer from the same effect, therefore it is a homogeneous effect.

Collision will disturb the light emission process. Two different collisions:

> inelastic – particle leaves the excited level $\rightarrow$ lifetime of the excited level decreases $\rightarrow$ similar to the previous case (lifetime broadening)
>
> elastic, there is no transfer between energy levels, there is a disturbance in the mechanism of light emission $\rightarrow$ random phase shift



$\tau_d \sim 10^{-13}$ s

$\tau_c \geq 10^{-8}$ s

Light-matter interactions

Homogeneous broadening – collision broadening (cont.)

Harmonic function with random phase shift $\rightarrow$ frequency spectra by Fourier transform $\rightarrow$ again Lorentz-function.

If τ_c is the average time between collisions, the normalized lineshape function:

$$g_c(\nu) = \frac{2\tau_c}{1+(\nu-\nu_0)^2 4\pi^2\tau_c^2}$$

$$g_c^{\max}(\nu) = 2\tau_c(\nu=\nu_0)$$

$$\boxed{\Delta\nu_c = \frac{1}{\pi\tau_c}}$$

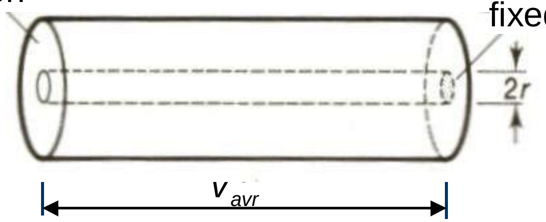
$\Delta\nu_c$ depends on the pressure, estimation with drastic simplifications:

„ideal" monatomic gas of radius *r*, hard spheres, we fix one particle, the others are moving toward the fix particle with v_{avr}^{rel} .

Light-matter interactions

Homogeneous broadening – collision broadening (cont.)

Suppose that the movement of particles toward the fixed particle takes place in a unit cross section cylinder of length v_{avr}^{rel} :



The probability of collision is proportional with the area of the fixed particle: $4r^2\pi$

The number of collisions in unit time is: $4r^2\pi \cdot N \cdot v_{avr}^{rel}$, N is the atomic density.

The average collision time is: $\tau_c = \dfrac{1}{4\pi r^2 v_{avr}^{rel} N}$. If there is m mol gas in V volume

$$PV = m \underset{\substack{\uparrow \\ N_A k_B}}{R} T = m N_A k_B T = N V k_B T \qquad (N = \frac{mN_A}{V}) \qquad N = \frac{P}{k_B T}$$

Light-matter interactions

Homogeneous broadening – collision broadening (cont.)

$$\tau_c = \frac{k_B T}{4\pi r^2 v_{avr}^{rel} P} \qquad \Delta\nu_c = \frac{1}{\pi\tau_c} = \frac{4r^2 v_{avr}^{rel} P}{kT}$$

Collision broadening is proportional to the pressure, $\Delta\nu_c \sim P$.

Rough guide suitable for estimation: $\frac{\Delta\nu_c}{P} \sim 5-10\frac{MHz}{torr}.$

Different homogeneous broadening together

The sum of Lorentzian distributions is again a Lorentzian function:

$$\Delta\nu_{L_1} + \Delta\nu_{L_2} = \Delta\nu_L \qquad \Delta\nu_{nat+c} = \frac{1}{2\pi}\left(\frac{1}{\tau_1} + \frac{2}{\tau_c}\right)$$

Crystal field interaction is a ”non conventional” collision process, interaction with phonons in solids, but similarly homogeneous effect → the lineshape function is Lorentzian.

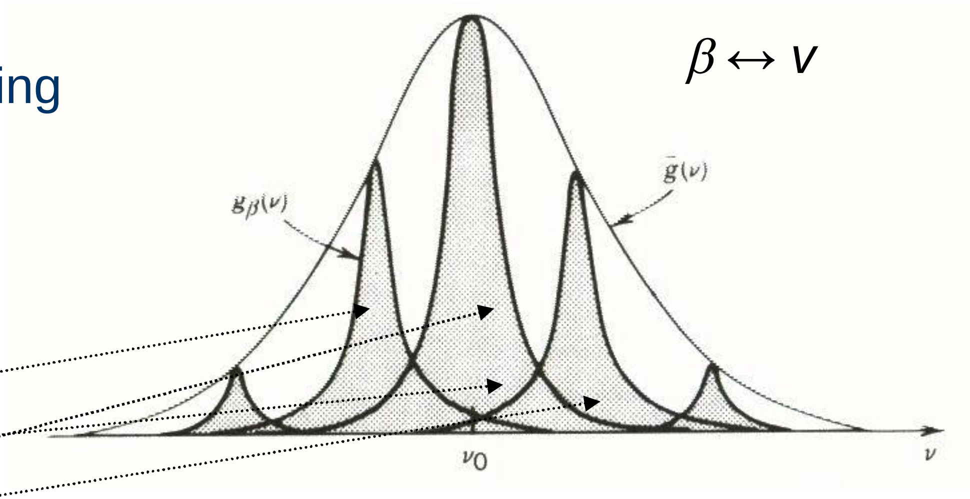
Light-matter interactions

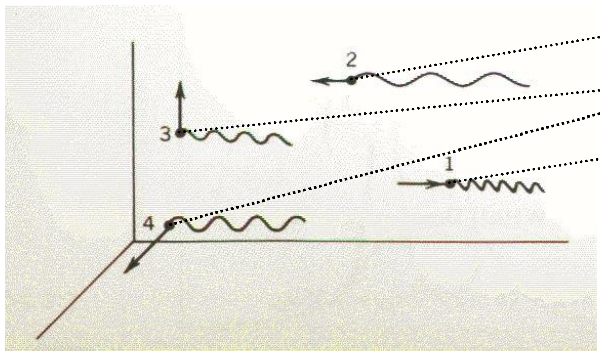
<u>Inhomogeneous broadening — Doppler-broadening</u>

Different atoms have different lineshape functions or different center of frequency → average lineshape function $\bar{g}(\nu) = \langle g_\beta(\nu) \rangle$, average with respect to the variable β.

E.g. light emission of atoms moving with velocity *v*, frequency shift because of the Doppler-effect.

$\beta \leftrightarrow v$





Direction of observation

$$\nu_0 \rightarrow \nu_0 \pm \left(\frac{v}{c}\right)\nu_0,$$

Temperature dependent velocity distribution in the gas

Light-matter interactions

Inhomogeneous broadening — Doppler-broadening (cont.)

$p(v)dv$ is the probability that the velocity of an atom is in the interval of [v, $v+dv$] , the average lineshape function is:

$$\bar{g}(\nu)=\int_{-\infty}^{\infty} g\left(\nu-\nu_0\frac{v}{c}\right)p(v)\,dv.$$

In equilibrium the velocity distribution of atoms with mass M at temperature T is the Maxwell - Boltzmann distribution. The probability that the velocity component of the atom is in the range of [v, $v+dv$] in a given direction (e.g. in the direction of the resonator axis)

$$p_v dv=\left(\frac{M}{2\pi kT}\right)^{1/2}\exp\left(-\frac{Mv^2}{2kT}\right)dv$$

Gaussian-distribution.

maximum at v=0

FWHM: $$\left|v_{1/2}\right|=\left(\frac{2\ln(2)kT}{M}\right)^{1/2}.$$

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Light-matter interactions

Inhomogeneous broadening — Doppler-broadening (cont.)

In case the homogeneous broadening $\Delta\nu_L << \nu_0 \left|\frac{v_{1/2}}{c}\right|$, its effect can be neglected:

$$p_v dv = g(\nu)d\nu = \underbrace{\frac{c}{\nu_o}\left(\frac{M}{2\pi kT}\right)^{1/2}}_{\max imum\ \nu=\nu_o} \exp\left\{-\frac{Mc^2}{2k_BT}\frac{(\nu-\nu_o)^2}{\nu_o^2}\right\}d\nu,$$

where

$$|v| = \frac{c(\nu_0-\nu)}{\nu_0}, \quad v^2 = \frac{c^2(\nu_0-\nu)^2}{\nu_0^2}, \quad dv = \frac{c}{\nu_0}d\nu.$$

$$\frac{1}{2} = exp\left\{-\frac{Mc^2}{2kT}\frac{(\nu_{1/2}-\nu_0)^2}{\nu_0^2}\right\}, \quad \nu_{1/2}-\nu_0 = \nu_0\left(\frac{2kT\ln 2}{Mc^2}\right)^{1/2},$$

$$\Delta\nu_D = \underbrace{2(\nu_{1/2}-\nu_0)}_{because\ of\ the\ symmetry} = \frac{2\nu_0}{c}\left(\frac{2kT\ln 2}{M}\right)^{1/2}$$

the FWHM or Doppler broadening.

($\frac{\nu_0}{c} = \frac{1}{\lambda}$)

Light-matter interactions

Different inhomogeneous broadenings together

Inhomogeneous distribution of doping materials in solids causes inhomogeneous broadening $\rightarrow$ Gaussian distribution. The superposition of two different Gaussian distribution:

$$\sqrt{\left(\Delta\nu_{1G}{}^{2}\right)+\left(\Delta\nu_{2G}\right)^{2}}=\Delta\nu_{G}.$$

Homogeneous and inhomogeneous distributions together

Convolution of the Lorentz and the Gauss functions $\rightarrow$ Voight-integral (can be numerically calculated).

Numerical examples - He-Ne laser

$\Delta\nu_D$=? $T = 400\ K$, $\lambda = 633\ nm$, $M_{Ne}=\dfrac{20\text{g}}{6\cdot 10^{23}}$, $k = 1.38\cdot 10^{-23}\ \text{JK}^{-1}$

$\Delta\nu_D^{Ne} = 1.5\cdot 10^{9}\ \text{Hz} = 1.5\ \text{GHz}.$ $h = 6.63\cdot 10^{-34}\ \text{Js}$

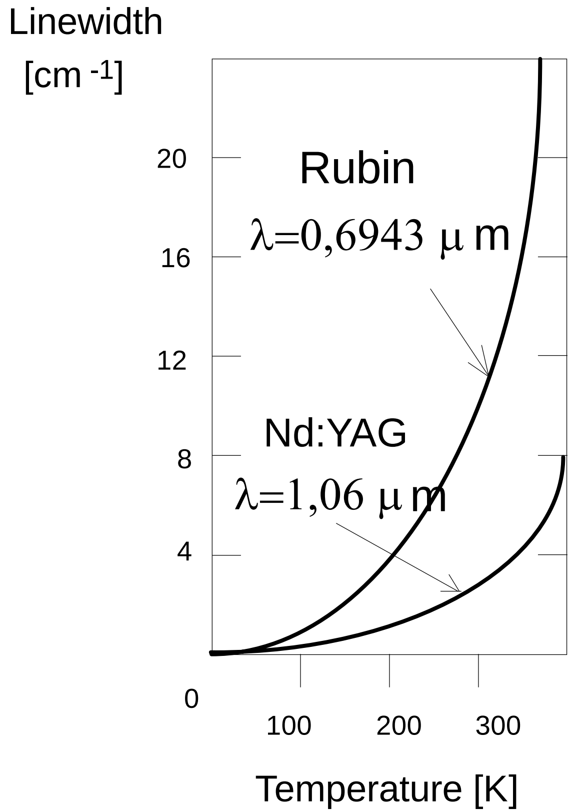
Collision broadening? Can be neglected at 3-4 *torr* pressure! Typical inhomogeneously broadened laser material.

Light-matter interactions

Numerical examples - ruby és Nd:YAG laser

T-dependent broadening, because the lattice vibration increases with *T*!
Typical systems with homogeneous broadening.
Inhomogeneous effect at $T \sim 0$ because of impurities.

ν/c_0 ($1/\lambda$, cm^{-1}) data,



e.g. at 300 *K*	$\Delta\nu_L$
Nd:YAG	120 GHz
Ruby	330 GHz

Light-matter interactions

Typical broadenings of different laser materials

	Type	Gas	Liquid	Solid
Homogeneous	natural	1 kHz - 10 MHz	can be neglected	can be neglected
	collision	5 - 10 MHz/torr	9 THz	-
	crystal field interaction	-	-	300 GHz (300K)
Inhomogeneous	Doppler	50 MHz - 1 GHz	can be neglected	-
	local field	-	15 THz	30 GHz - 15 THz

Light-matter interactions

Problem

Possible decays of E_2 and E_1 in an atom, t_{sp} = 5 ms, τ_{nr} = 50 µs, τ_{20} = 10 ps, τ_1 = 15 µs. Calculate τ_{21} and $\Delta\nu_{nat}$ of the transition! Is it an ideal choice to use that transition for a laser transition?