Lecture 18 — 11/8

Announce:

Next week (11/13 + 15), I'll be gone ⇒

11/13, class will be at 9 o'clock (instead of 3)

11/15: class will be taught by Abigail

Program:

- Line shapes & Broadening
- Many-electron atoms

7) Line shapes

a) Recap / Motivation

- Ideal lineshape for measurement: Rabi transition

$$P = \frac{\Omega^2}{\Omega^2+\delta^2} \sin^2 \frac{\sqrt{\Omega^2+\delta^2}}{2} t \qquad \text{transition probability}$$

- General approach

V: coupling between $|a\rangle, |b\rangle$, $\omega_0 = \omega_b - \omega_a$

$|\psi\rangle = a|a\rangle + b|b\rangle$

$i\dot{a} = \langle a|V|b\rangle e^{-i\omega_0 t}\, b$

$i\dot{b} = \langle b|V|a\rangle e^{i\omega_0 t}\, a$

assume $a(0) = 1$,

$$W_{ba} \underset{|a\rangle\to|b\rangle}{:\ \text{rate}} \equiv \frac{d}{dt}|b(t)|^2$$

1^{st} order perturbation theory: $a(t) \approx a(0) = 1$

$$W_{ba} = \int_0^t dt' \underbrace{\langle a|V(t)|b\rangle\langle b|V(t')|a\rangle}_{\equiv G_{ba}(t,t') \underset{\text{usually}}{=} G_{ba}(\underbrace{t-t'}_{\tau})} e^{-i\omega_0(t-t')} + c.c.$$

$$= \int_{-\infty}^{t} d\tau\, G_{ba}(\tau)\, e^{-i\omega_0 \tau}$$

(gives same short-time ($t \ll \frac{1}{\Omega}$) monochromatic limit as Rabi)

b) Homogeneous & inhomogeneous broadening

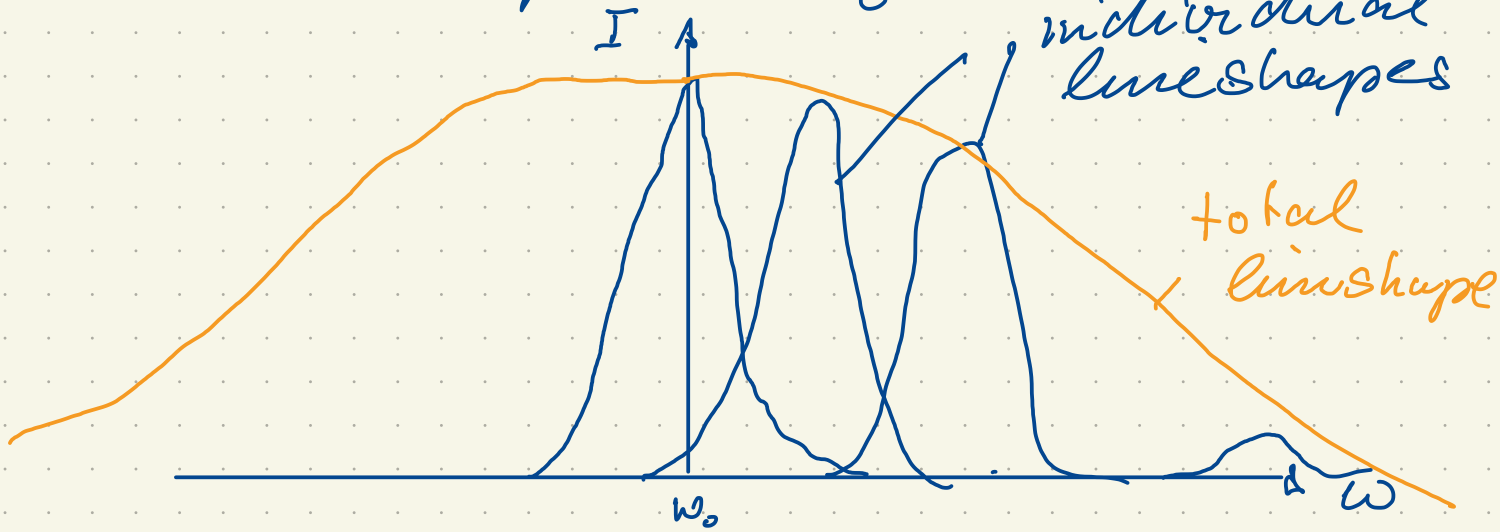
"Homogeneous": all atoms (or molecules) in the system have identical line shape functions.

<u>Examples</u>: natural lineshape ("lifetime br."), pressure broadening

line shape: typically Lorentzian

"<u>inhomogeneous</u>": different (micro-) environment for different atoms.

$\Rightarrow$ total lineshape is average!



most important example: Doppler

typical lineshape: Gaussian

c) Examples

- <u>Natural linewidth</u>

$$G_{ba}(\tau) = \frac{|\wp|^2}{4} e^{-\frac{\Gamma}{2}\tau} e^{i\omega\tau} \qquad (\Gamma = \gamma_b + \gamma_a)$$

$$\Rightarrow W_{ba} = \frac{|\wp|^2}{2} \frac{\Gamma/2}{(\Gamma/2)^2 + \delta^2} \qquad \text{Lorentzian}$$

(Note: this is the limit of $\Omega \to 0$!)

- Doppler broadening

Doppler shift of atom with velocity $\vec{v}$ and resonance frequency ω_0:

$$\omega' = \omega_0 + \vec{k}\cdot\vec{v}$$

$\omega - kv \quad \circ \quad \omega + kv$, $\vec{v}$

$$\left.\right\} \quad \frac{\Delta\omega}{\omega_0} = \frac{v}{c} \qquad (1)$$

Fraction of atoms with velocity between v and $v + dv$:

$$(2) \qquad f(v)\,dv = \frac{1}{\sqrt{\pi}\,\alpha} e^{-\frac{v^2}{\alpha^2}} \qquad \left(\alpha = \sqrt{\frac{2kT}{M}} \text{ most probable velocity}\right)$$

Maxwell - Boltzmann distribution

$$(1)+(2) \Rightarrow \boxed{g_{Doppler}(\omega') = \frac{c}{\sqrt{\pi}\,\alpha\,\omega_0} e^{-\left(\frac{c\,\Delta\omega}{\alpha\,\omega_0}\right)^2}} \qquad \text{with } \Delta\omega = \omega' - \omega_0$$

"spectral density", "form factor"

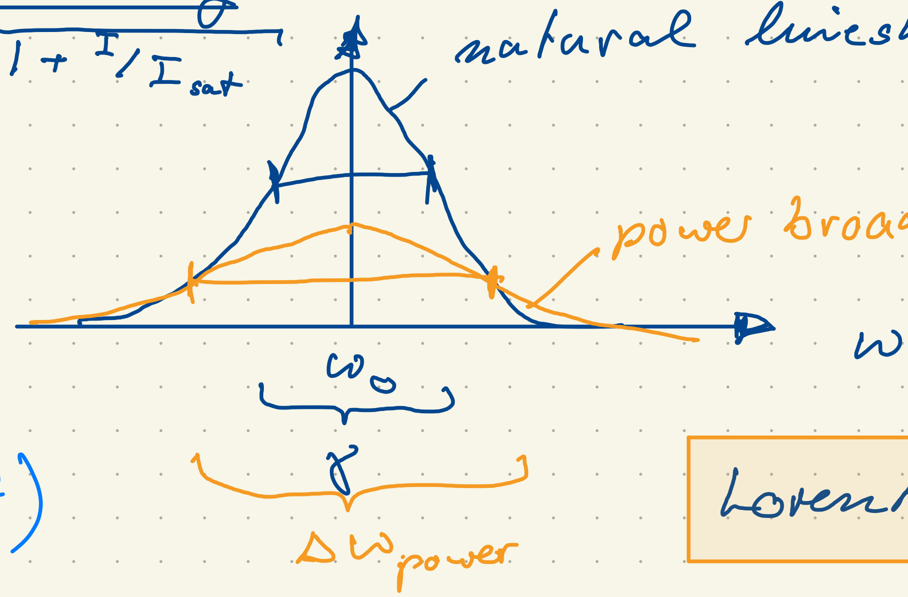
$$\Rightarrow \text{FWHM}: \Delta\omega_{Doppler} = 2\sqrt{\ln 2}\,\frac{\alpha\,\omega_0}{c} = 2\sqrt{\ln 2}\,\alpha k$$

for Hydrogen @ RT: $\alpha = 2230\ \frac{m}{s}$

$$\frac{\Delta\omega_{Dopp}}{2\pi} \approx 6\,\text{GHz} \Big|_{\lambda = 600\,nm}$$

- Power Broadening

$$\Delta\omega_{power} = \Gamma\sqrt{1 + I/I_{sat}}$$



(see Budker 3.7)

$\boxed{\text{Lorentzian}}$

- Pressure broadening:

colliding atoms (elastic) ⇒ motion is governed by diffusion.

⇒ collision time $\hat{=}$ correlation (coherence) time T_2

$$\Delta\omega_{pressure} = \frac{1}{\pi T_2}$$

Lorentzian (because diffusion affects all atoms equally)

- Dicke narrowing

some elastic collisions don't disturb coherence of atoms

e.g. with buffer gas (e.g. He)

⇒ Doppler broadening is <u>limited</u>

Diffusing atoms:

$$P(z,z',t) \equiv \text{probability}\left[(z,0) \rightarrow (z',t)\right]$$

$$P(z,z',t) = \frac{1}{\sqrt{4\pi D t}}\, e^{-\frac{(z-z')^2}{4Dt}}$$

$$G_{ba} = \frac{|\beta|^2}{4}\, \overline{e^{ik(z-z')}} = \frac{|\beta|^2}{4\sqrt{4\pi D t}} \int ds\, e^{-\frac{s^2}{4Dt}}\, e^{iks}$$

$$= \frac{|\beta|^2}{4}\, e^{-k^2 D t}$$

$$\boxed{W_{ba} = \frac{|\beta|^2}{2}\, \frac{k^2 D}{(k^2 D)^2 + \delta^2}}$$

Lorentzian

$$\Delta\omega_{Dicke} = 2 k^2 D$$

Ideal gas: $D = \frac{\alpha\, l}{3}$ $\quad$ l: mean free path

$$\Rightarrow \Delta\omega_{Dicke} = 2\, \frac{2\pi}{\lambda}\, k\, \frac{\alpha\, l}{3} = \frac{2\pi}{3\sqrt{2\ln 2}}\, \frac{l}{\lambda}\, \Delta\omega_{Doppler}$$

for $l \ll \lambda$: narrowing!

- transit time broadening

atoms transit through interaction region (e.g. laser beam) during time window τ:

$$\overline{E(\nu)} = \frac{E_0}{2} \int_{-\tau/2}^{\tau/2} dt \left(e^{i\omega_0 t} + e^{-i\omega_0 t}\right) e^{-i\nu t}$$

$$= E_0 \frac{\sin \frac{\omega_0 - \nu}{2}\tau}{\omega_0 - \nu}$$

$$\Rightarrow g_{transit}(\delta) = \frac{2}{\pi\tau} \frac{\sin^2 \delta/2\, \tau}{\delta^2}$$

$$\Rightarrow \Delta\omega_{transit} \approx \frac{5.6}{\tau} .$$

For $v = 500 \frac{m}{s}$, laser beam width 1mm

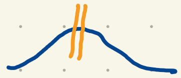
$$\Rightarrow \tau \approx 2 \cdot 10^{-6} s \longrightarrow \Delta\omega_{transit} \approx 2.8 \text{ MHz}$$

(Budker 3.13)

typically Gaussian

d) Mitigation of broadening

Often easy / possible to make line width narrower:

- measure longer (often: $\delta\omega \propto \frac{1}{T}$ or similar)
- "split the line" 
- for inhomogeneous: single out atoms

  example: Doppler – select single velocity element ("hole burning")

  example: spin echo

  in general: inhomogeneous line broadening can *always* be mitigated
- homogeneous: often only made better by increasing measuring time T ⇒ Ramsey !

Life times:

T_1 : population life times (typically $T_1 = \frac{1}{\gamma}$)

T_2 : coherence life time (i.e. decay rate of off-diagonal density-matrix elements in the ideal case, e.g. single-atom)

T_2^*: real measured decoherence time (inverse of inhomogeneous broadening) – often orders of magnitude larger than T_1, T_2

Ramsey interferometry

Remember: probability of (transition into) excited state:

$$P(t) = \frac{\Omega^2}{\Delta^2+\Omega^2}\sin^2\frac{\sqrt{\Omega^2+\Delta^2}}{2}t$$

(ideal case w/ decay neglected)

⇒ linewidth of this determines precision, measuring time for π-pulse:

$\tau = \frac{\pi}{\sqrt{\Omega^2 + \Delta^2}}$ ⇒ linewidth ideally Ω ⇒

small Ω ⇒ narrow line ⇒ long τ

compounded / limited by decoherence

Ramsey's idea: two $\frac{\pi}{2}$-pulses replace π-pulse

⇒ $\frac{\pi}{2}$ interaction → free evolution (no technical noise) → $\frac{\pi}{2}$ interaction

then (for $\Delta \to 0$):

$P(t) \propto \cos^2 \frac{\Delta\omega \cdot T}{2}$

⇒ $\Delta\omega = \frac{2\pi}{T}$ for one fringe, T is only limited by (natural) decoherence!

(see Cs clock example)