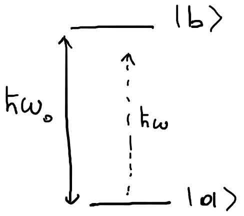
# Non-perturbative susceptibility

Solving the equations of motion for the density matrix under monochromatic drive we find for the coherence:

$$\boxed{\rho_{ba}(t) = -i\frac{\Omega_1^*}{2} \times \frac{\gamma + i(\omega-\omega_0)}{(\omega_0-\omega)^2+\gamma^2+\Omega_1^2\frac{\gamma}{T_{sp}}} e^{-i\omega t}}$$




with $\Omega_1 = \frac{-\mu_{ab} E}{\hbar}$ ; $\mu_{ab} \equiv \langle a|\hat{\mu}|b\rangle$

$\Delta\omega = \omega - \omega_0$

The expectation value of the dipole moment operator is:

$$\langle\hat{\mu}\rangle = Tr\{\hat{\rho}\hat{\mu}\} = \mu_{ab}\,\rho_{ba}(t) + c.c.$$

$$= \frac{i|\mu_{ab}|^2}{2\hbar} \times \frac{\gamma + i(\Delta\omega)}{(\Delta\omega)^2+\gamma^2+\Omega_1^2\frac{\gamma}{T_{sp}}} E\, e^{-i\omega t} + c.c.$$

Polarisation: $P(\omega) = \frac{N}{V}\langle\hat{\mu}\rangle = \chi(\omega, E)\, E(\omega)$ with $E(\omega) = \frac{E}{2}$

- Grynber, Aspect, Fabre - Complement 2C density matrix and optical Bloch equations
- Boyd – Ch. 6 Nonlinear Optics in the Two-Level Approximation



$$\chi(\omega, E_0) = \frac{N}{V}\frac{i|\mu_{ab}|^2}{\varepsilon_0\hbar} \times \frac{\gamma + i(\Delta\omega)}{(\Delta\omega)^2+\gamma^2+\Omega_1^2\frac{\gamma}{T_{sp}}} = \frac{-N|\mu_{ab}|^2}{V\varepsilon_0\hbar\gamma} \times \frac{\Delta\omega/\gamma - i}{1+\left(\frac{\Delta\omega}{\gamma}\right)^2+\frac{\Omega_1^2}{\gamma T_{sp}}}$$

For a dilute gaz $n(\omega, E_0) = \sqrt{1+\chi(\omega,E)} \approx 1 + \frac{1}{2}\chi(\omega, E)$ as $|\chi| \ll 1$

↳ The absorption coefficient is: $\alpha = \frac{2\omega}{c}\mathrm{Im}[n] \approx \frac{\omega}{c}\mathrm{Im}[\chi]$

After defining $\alpha_0 = \frac{\omega_0}{c} \times \frac{N}{V} \cdot \frac{|\mu_{ab}|^2}{\varepsilon_0\hbar\gamma}$ we can write:

$$\alpha(\Delta\omega) = \frac{\alpha_0}{1+\frac{\Omega_1^2}{\gamma T_{sp}}}\left(1 + \frac{(\Delta\omega)^2}{\gamma^2\left(1+\frac{\Omega_1^2}{\gamma T_{sp}}\right)}\right)^{-1}$$

↳ Lorentzian of FWHM: $\Delta\omega_{1/2} = 2\gamma\sqrt{1+\frac{\Omega_1^2}{\gamma T_{sp}}}$

and peak value: $\alpha_{max} = \frac{\alpha_0}{1+\frac{\Omega_1^2}{\gamma T_{sp}}}$

We may define $E_S^2 = \frac{\hbar^2 \gamma T_{sp}}{|\mu_{ab}|^2}$ so that $\frac{\Omega_1^2}{\gamma T_{sp}} = \frac{E^2}{E_S^2}$ (saturation field)

and then: $$\chi(\omega, E) = \frac{-\alpha_0 c}{\omega_0} \times \frac{\Delta\omega/\gamma - i}{1 + \left(\frac{\Delta\omega}{\gamma}\right)^2} \left(1 + \frac{E^2/E_s^2}{1+\left(\frac{\Delta\omega}{\gamma}\right)^2}\right)^{-1}$$

Expanding in Taylor series to first order:

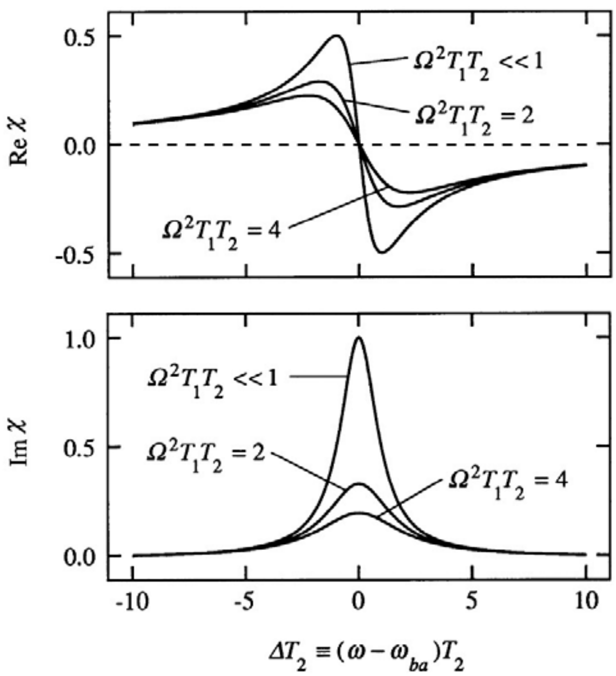
$$\chi(\omega, E) \simeq \frac{-\alpha_0 c}{\omega_0} \times \frac{\Delta\omega/\gamma - i}{1 + \left(\frac{\Delta\omega}{\gamma}\right)^2} \times \left[1 - \frac{1}{1+\left(\frac{\Delta\omega}{\gamma}\right)^2} \times \frac{E^2}{E_S^2}\right] + O(E^4)$$

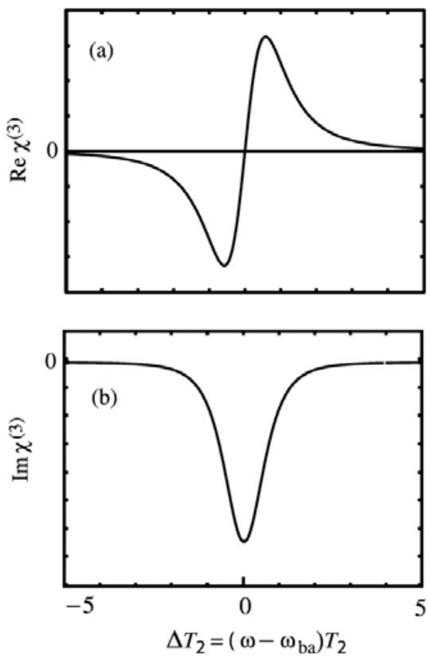
$$= \chi^{(1)}(\omega) + K\, \chi^{(3)}(-\omega_s; \omega, \omega, -\omega)\, E^2$$

with $K = 3/4$ (cf. Week 04)



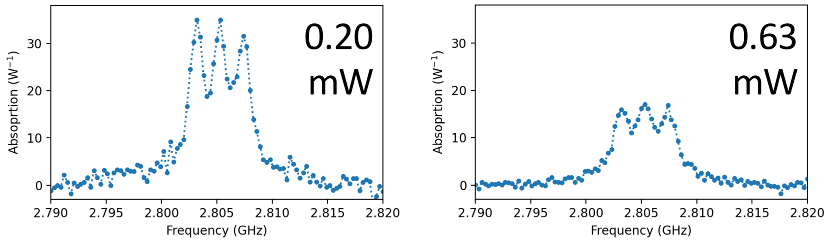
# Power dependent susceptibility

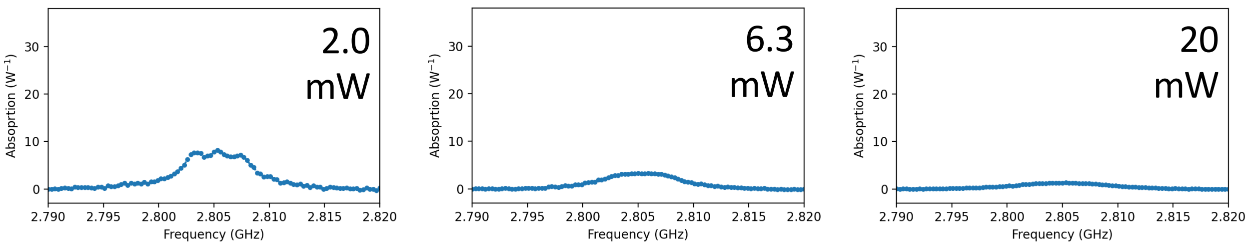


# Example from our lab: microwave absorption of NV centers in diamond (© Valentin Goblot)

- Absorption normalized by nominal power in MW antenna





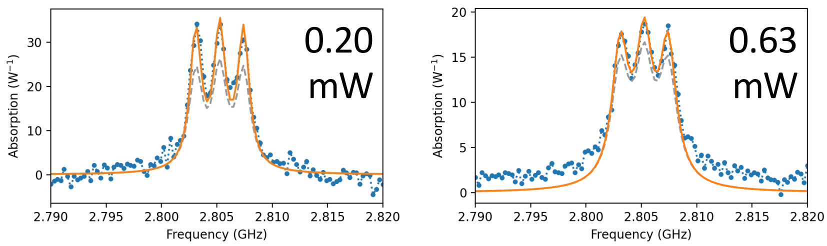

# Example from our lab: microwave absorption of NV centers in diamond

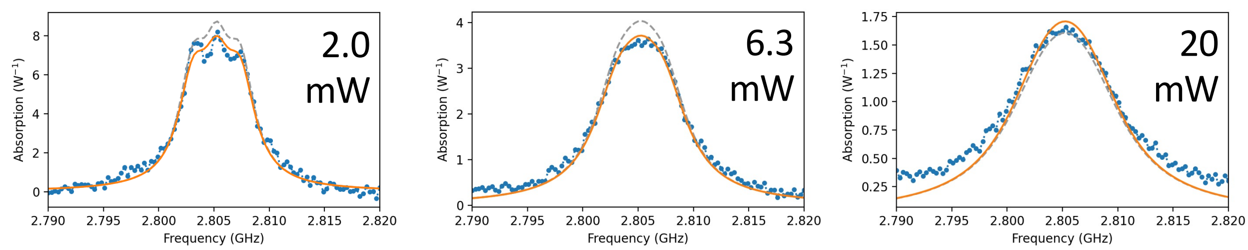
$$\alpha(\Delta\omega) = \frac{\alpha_0}{1+\frac{\Omega_1^2}{\gamma T_{sp}}}\left(1+\frac{(\Delta\omega)^2}{\gamma^2\left(1+\frac{\Omega_1^2}{\gamma T_{sp}}\right)}\right)^{-1}$$

- Result of the fit:

  $\gamma = 0.60$ MHz

  Nominal $\Omega_1$
  Fitted $\Omega_1$



Conclusion $\chi^{(1)}(\omega) = \frac{-\alpha_0 c}{\omega_0} \frac{\frac{\Delta\omega}{\gamma} - i}{1+\left(\frac{\Delta\omega}{\gamma}\right)^2}$

$$\chi^{(3)}(-\omega_s;\omega,\omega,-\omega) = -\frac{4\chi^{(1)}(\omega)}{3(E_S^\Delta)^2} \qquad \text{with } (E_S^\Delta)^2 = E_S^2\left(1+\left(\frac{\Delta\omega}{\gamma}\right)^2\right)$$

The saturation intensity for a given detuning is : $I_S^\Delta = \frac{1}{2}\varepsilon_0 c (E_S^\Delta)^2$

Typical values for atomic systems (ex: $3s \to 3p$ of sodium):

$|\mu_{ab}| = 2.5\, e\, a_0 \simeq 2.0 \times 10^{-29}$ C.m

↳ Rabi frequency : $\Omega = \frac{|\mu_{ab}|}{\hbar}\left(\frac{2I}{\varepsilon_0 c}\right)^{1/2} = 2\pi \times 10^9 \left[\frac{I\,[W/cm^2]}{127}\right]^{1/2}$ $[rad.s^{-1}]$

With $\frac{1}{T_{sp}} = 16$ ns and $\gamma = \frac{T_{sp}}{2}$ we get : $I_S^{\Delta=0} = 52.7\,\frac{W}{m^2} = 52.7\,\frac{pW}{\mu m^2}$



# Absorption cross-section of a single atom

We define the absorption cross section $\sigma$ by : $\underset{[m^{-1}]}{\alpha} = \underset{[m^{-3}]}{\frac{N}{V}}\sigma \Rightarrow \sigma\,[m^2]$

↳ Effective area of a single atom interacting with monochromatic light.

On resonance, far below saturation : $\sigma_0 = \frac{2\pi}{\lambda}\frac{|\mu_{ab}|^2}{\varepsilon_0\hbar\gamma}$

$\sigma_0$ is maximum if $\gamma = \frac{T_{sp}}{2}$ (no pure dephasing). But $\boxed{T_{sp} = \frac{\omega_0^3}{c^2} \times \frac{|\mu_{ab}|^2}{3\pi\varepsilon_0\hbar}}$

("Einstein A coefficient")

Therefore $\sigma_0^{max} = \frac{2\pi}{\lambda} \times 6\pi\left(\frac{\lambda}{2\pi}\right)^3 = \frac{3\lambda^2}{2\pi}$

↳ The "optical size" of a single atom is comparable to the wavelength at resonance

But not true if $\gamma \gg T_{sp}$ ; or if $\Delta\omega \gg \gamma$ .