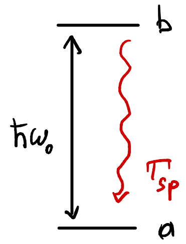
Non linear response of the TLS



1) <u>Hamiltonian</u> • $\hat{H}_0 = \hbar\omega_0 |b\rangle\langle b|$

Satisfies $\hat{H}_0 |a\rangle = 0\, |a\rangle$; $\hat{H}_0 |b\rangle = \hbar\omega_0 |b\rangle$

• Dipole interaction with light $\vec{E}(t) = E_0 \vec{e}_x \cos\omega t$ (classical)

$\hat{H}_I(t) = -\hat{\mu}_x E_0 \cos\omega t$

↳ $\langle a | H_I(t) | a\rangle = -\langle a|\hat{\mu}|a\rangle E_0 \cos\omega t = -\mu_{aa} E_0 \cos\omega t = 0$

↳ permanent dipole

$\langle b | H_I(t) | b\rangle = 0$

$\langle a | H_I(t) | b\rangle = -\mu_{ab} E_0 \cos\omega t$ with $\mu_{ab} = \mu_{ba}^*$ ($\hat{\mu}$ is an observ. ↳ Hermitian)

$= \langle a|\hat{\mu}|b\rangle$

Finally $\hat{H}_I(t) = \left(\hbar \underbrace{\Omega_1}_{} |a\rangle\langle b| + \hbar\Omega_1^* |b\rangle\langle a|\right) \cos\omega t \to \dfrac{e^{i\omega t} + e^{-i\omega t}}{2}$

($= \Omega_1$)

2) <u>Heisenberg equations</u> $[\hat{H}_I(t), \hat{\rho}] = \hat{H}_I \hat{\rho} - \hat{\rho}\hat{H}_I$ where $\hat{\rho} = \rho_{aa} |a\rangle\langle a| + \rho_{bb} |b\rangle\langle b| + \rho_{ab} |a\rangle\langle b| + \rho_{ba} |b\rangle\langle a|$

$$H_I \rho = \hbar\Omega_1 \cos\omega t \Big(|a\rangle\langle b|b\rangle\langle b| \rho_{bb} + |a\rangle\langle b|b\rangle\langle a| \rho_{ba} + |b\rangle\langle a|a\rangle\langle a| \rho_{aa} + |b\rangle\langle a|a\rangle\langle b| \rho_{ab} \Big)$$

$$= \hbar\Omega_1 \cos\omega t \Big(|a\rangle\langle b| \rho_{bb} + |a\rangle\langle a| \rho_{ba} + |b\rangle\langle a| \rho_{aa} + |b\rangle\langle b| \rho_{ab}$$

$$\rho H_I = \hbar\Omega_1 \cos\omega t \Big(\rho_{aa} |a\rangle\langle b| + \rho_{ba} |b\rangle\langle b| + \rho_{bb} |b\rangle\langle a| + \rho_{ab} |a\rangle\langle a| \Big)$$

$$H_I \rho - \rho H_I = \hbar\Omega_1 \cos\omega t \Big(|a\rangle\langle a| (\rho_{ba} - \rho_{ab}) + |b\rangle\langle b| (\rho_{ab} - \rho_{ba})$$
$$+ |a\rangle\langle b| (\rho_{bb} - \rho_{aa}) + |b\rangle\langle a| (\rho_{aa} - \rho_{bb}) \Big)$$

Similarly : $\hat{H}_0 \hat{\rho} - \hat{\rho}\hat{H}_0 = \hbar\omega_0 \left(|b\rangle\langle a| \rho_{ba} - |a\rangle\langle b| \rho_{ab} \right)$

$$\dot\rho = -\frac{i}{\hbar}[\hat H,\hat\rho] \rightarrow$$

$$\dot\rho_{aa} = -i\Omega_1\cos\omega t\,(\underbrace{\rho_{ba}-\rho_{ab}}_{\in i\mathbb{R}}) \quad (1)$$

$$\dot\rho_{bb} = -\dot\rho_{aa} \quad (2)$$

$$\dot\rho_{ab} = -i\Omega_1\cos\omega t\,(\rho_{bb}-\rho_{aa}) + i\omega_0\rho_{ab} \quad (3)$$

$$\dot\rho_{ba} = i\Omega_1\cos\omega t\,(\rho_{bb}-\rho_{aa}) - i\omega_0\rho_{ba} \quad (4)$$

3) We see that $\dot\rho_{bb}+\dot\rho_{aa} = 0 \Rightarrow \rho_{bb}+\rho_{aa} = c^{ste} = tr[\hat\rho] = 1$

4) When $\Omega_1 = 0$: $\rho_{aa} = c^{st}$, $\rho_{bb} = c^{st}$

$$\rho_{ab}(t) = \rho_{ab}^{(0)}e^{i\omega_0 t}$$

$$\rho_{ba}(t) = \rho_{ab}^{(0)*}e^{-i\omega_0 t}$$

coherences

5) $\cos\omega t = \frac{1}{2}(e^{+i\omega t}+e^{-i\omega t})$ For example in (3) :

$$\dot\rho_{ab} = -\frac{i\Omega_1}{2}(\rho_{bb}-\rho_{aa})(e^{i\omega t}+e^{-i\omega t}) + i\omega_0\rho_{ab} \approx -\frac{i\Omega_1}{2}e^{i\omega t}(\rho_{bb}-\rho_{aa}) + i\omega_0\rho_{ab}$$

$$\dot\rho_{ba} \approx \frac{i\Omega_1}{2}e^{-i\omega t}(\rho_{bb}-\rho_{aa}) - i\omega_0\rho_{ba}$$

$$\dot\rho_{aa} = -\frac{i\Omega_1}{2}(e^{i\omega t}+e^{-i\omega t})(\underbrace{\rho_{ba}}_{\sim e^{-i\omega_0 t}} - \underbrace{\rho_{ab}}_{\sim e^{+i\omega_0 t}}) \simeq -\frac{i\Omega_1}{2}(\rho_{ba}e^{+i\omega t} - \rho_{ab}e^{-i\omega t})$$

6) T_{sp} = spontaneous emission (spins : longitudinal relaxation / population deca

γ = dephasing $\geq \frac{T_{sp}}{2}$

7) $\rho'_{ab} = \rho_{ab}e^{-i\omega t} \rightarrow \dot\rho_{ab} = \dot\rho'_{ab}e^{i\omega t} + i\omega\rho'_{ab}e^{i\omega t} = -\frac{i\Omega_1}{2}e^{i\omega t}(\rho_{bb}-\rho_{aa}) + i\omega_0\rho'_{ab}e^{i\omega t}$

$$\Leftrightarrow \quad \dot\rho'_{ab} = -\frac{i\Omega_1}{2}(\rho_{bb}-\rho_{aa}) + i(\omega_0-\omega)\rho'_{ab} \quad (2')$$

$$\dot\rho_{bb} = +\frac{i\Omega_1}{2}(\rho'_{ba}-\rho'_{ab}) \quad (1')$$

No time-dependent coefficients

We now add relaxation terms :

$$\dot\rho_{bb} = \frac{i\Omega_1}{2}\left(\rho'_{ba} - \rho'_{ab}\right) - \Gamma_{sp}\,\rho_{bb} \quad ; \quad \dot\rho_{aa} = -\dot\rho_{bb} \qquad (1'')$$

$$\dot\rho'_{ab} = -\frac{i\Omega_1}{2}\left(\rho_{bb} - \rho_{aa}\right) + i(\omega_0 - \omega)\,\rho'_{ab} - \gamma\,\rho'_{ab} \qquad (2'')$$

8) Steady-state

$$\rho_{bb} = \frac{i\Omega_1}{2\Gamma_{sp}}\left(\rho'_{ba} - \rho'_{ab}\right) \qquad \rho_{aa} = 1 - \rho_{bb} \qquad (1ss)$$

$$(-i\,\delta\omega - \gamma)\,\rho'_{ab} = \frac{i\Omega_1}{2}\left(2\rho_{bb} - 1\right) \qquad (2ss)$$

2ss) $$\rho'_{ba} - \rho'_{ab} = \frac{1}{2}\left(2\rho_{bb} - 1\right)\left(\frac{-i\Omega_1}{+i\delta\omega - \gamma} - \frac{i\Omega_1}{-i\delta\omega - \gamma}\right) = \frac{\Omega_1}{2}\left(2\rho_{bb} - 1\right)\frac{-\delta\omega + i\gamma + \delta\omega + i\gamma}{\gamma^2 + \delta\omega^2}$$

$$= \frac{i\Omega_1\gamma}{\gamma^2 + \delta\omega^2}\left(2\rho_{bb} - 1\right)$$

1ss) $$\rho_{bb} = \frac{-\Omega_1^2\gamma}{2\Gamma_{sp}(\gamma^2 + \delta\omega^2)}\left(2\rho_{bb} - 1\right) \Rightarrow \frac{\Omega_1^2\gamma}{2\Gamma_{sp}(\gamma^2 + \delta\omega^2)} = \rho_{bb} \times \frac{\Gamma_{sp}(\gamma^2 + \delta\omega^2) + \Omega_1^2\gamma}{\Gamma_{sp}(\gamma^2 + \delta\omega^2)}$$

$$\boxed{\rho_{bb} = \frac{1}{2}\,\frac{\Omega_1^2\,\gamma/\Gamma_{sp}}{\delta\omega^2 + \gamma^2 + \Omega_1^2\frac{\gamma}{\Gamma_{sp}}}} \qquad \rho_{aa} = 1 - \rho_{bb}$$

$$\rho'_{ab} = \frac{i\Omega_1}{2(-i\delta\omega - \gamma)}\,\frac{-\delta\omega^2 - \gamma^2}{\delta\omega^2 + \gamma^2 + \Omega_1^2\frac{\gamma}{\Gamma_{sp}}} = \frac{i\Omega_1}{2}\,\frac{-i\delta\omega + \gamma}{\delta\omega^2 + \gamma^2 + \Omega_1^2\frac{\gamma}{\Gamma_{sp}}} \quad ; \quad \rho'_{ba} = \rho'^{*}_{ab}$$

9) $\rho_{ab} = \rho'_{ab}\,e^{i\omega t}$

$$\langle\hat\mu_x\rangle = Tr\left[\hat\mu_x\hat\rho\right] = \langle a|\hat\mu\hat\rho|a\rangle + \langle b|\hat\mu\hat\rho|b\rangle$$

$$= \langle a|\hat\mu|b\rangle\langle b|\hat\rho|a\rangle + \langle b|\hat\mu|a\rangle\langle a|\hat\rho|b\rangle$$

$$= \mu_{ab}\,\rho_{ba} + \mu_{ba}\,\rho_{ab}$$

$$\hookrightarrow \mu_{ab}^{*}\,\rho_{ba}^{*}$$

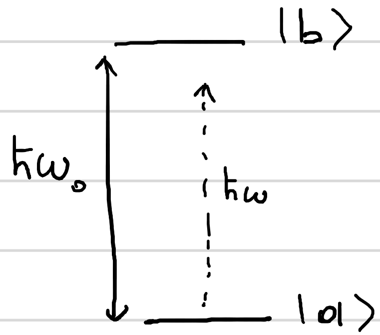
$$\langle\hat\mu_x\rangle = -\frac{i\Omega_1}{2}\,\mu_{ab}\,\frac{+i\delta\omega + \gamma}{\delta\omega^2 + \gamma^2 + \Omega_1^2\frac{\gamma}{\Gamma_{sp}}}\,e^{-i\omega t} + c.c. \qquad \text{but } \Omega_1 = -\frac{\mu_{ab}E_0}{\hbar}$$

$$\langle\hat\mu_x\rangle = E_0\,\frac{|\mu_{ab}|^2}{2\hbar}\,\frac{i\delta\omega + \gamma}{\delta\omega^2 + \gamma^2 + \Omega_1^2\frac{\gamma}{\Gamma_{sp}}}\,e^{-i\omega t} + c.c. \quad \to \Omega_1 \propto E_0 \;!$$

3a. 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 $\Omega_1 = \frac{-\mu_{ab} E}{\hbar}$; $\mu_{ab} \equiv \langle a | \hat{\mu} | b \rangle$

The expectation value of the dipole moment operator is :

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

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

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

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