* We are going to use now our knowledge of the Master equation and the quantum regression theorem to study some important averages and correlations. In this way we will see "in live" how these methods work in reality.

* We have seen already how $\langle 1|\hat{\rho}|1\rangle \equiv \rho_{11}$ and $\langle 2|\hat{\rho}|2\rangle \equiv \rho_{22}$ evolve in time (remember the rate equations above). In a similar way you can find the equation for $\langle 2|\hat{\rho}|1\rangle \equiv \rho_{21}$ and $\langle 1|\hat{\rho}|2\rangle \equiv \rho_{12}$

One gets in this way:

$$\dot{\rho}_{11} = -\gamma\bar{n}\rho_{11} + \gamma(\bar{n}+1)\rho_{22}$$

→ remember that $\rho_{11}+\rho_{22}=1$ at any time, and hence $\dot{\rho}_{22}=-\dot{\rho}_{11}$

$$\left.\begin{aligned}\dot{\rho}_{21} &= -\left[\frac{\gamma}{2}(2\bar{n}+1)+i\omega_A\right]\rho_{21}\\ \dot{\rho}_{12} &= -\left[\frac{\gamma}{2}(2\bar{n}+1)-i\omega_A\right]\rho_{12}\end{aligned}\right\} \quad \rho_{21}=\rho_{12}^{*} \text{ always.}$$

Note: we take $\omega_A' \equiv \omega_A$ to simplify notation)

* Now, remember that
  * $\hat{\sigma}_z \equiv |2\rangle\langle 2| - |1\rangle\langle 1| \longrightarrow \langle\hat{\sigma}_z\rangle = \rho_{22}-\rho_{11}$
  * $\hat{\sigma}_+ \equiv |2\rangle\langle 1| \longrightarrow \langle\hat{\sigma}_+\rangle = \rho_{12}$
  * $\hat{\sigma}_- \equiv |1\rangle\langle 2| \longrightarrow \langle\hat{\sigma}_-\rangle = \rho_{21}$

Then, from the previous equations one easily gets:

$$\frac{d}{dt}\langle\hat{\sigma}_z\rangle = -\gamma\left[(2\bar{n}+1)\langle\hat{\sigma}_z\rangle + 1\right]$$

$$\frac{d}{dt}\langle\hat{\sigma}_-\rangle = -\left[\frac{\gamma}{2}(2\bar{n}+1)+i\omega_A\right]\langle\hat{\sigma}_-\rangle$$

$$\frac{d}{dt}\langle\hat{\sigma}_+\rangle = -\left[\frac{\gamma}{2}(2\bar{n}+1)-i\omega_A\right]\langle\hat{\sigma}_+\rangle$$

* At optical frequencies and normal laboratory frequencies $\bar{n}$ is negligible, and the equations become of the form

$$\frac{d}{dt}\langle\hat{\sigma}_z\rangle = -\gamma\left[1+\langle\hat{\sigma}_z\rangle\right]$$

$$\frac{d}{dt}\langle\hat{\sigma}_-\rangle = -\left[\frac{\gamma}{2}+i\omega_A\right]\langle\hat{\sigma}_-\rangle$$

$$\frac{d}{dt}\langle\hat{\sigma}_+\rangle = -\left[\frac{\gamma}{2}-i\omega_A\right]\langle\hat{\sigma}_+\rangle$$

* let us now use the relations

- $\hat{\sigma}_+\hat{\sigma}_- + \hat{\sigma}_-\hat{\sigma}_+ = \hat{1}$
- $[\hat{\sigma}_+, \hat{\sigma}_-] = \hat{\sigma}_+\hat{\sigma}_- - \hat{\sigma}_-\hat{\sigma}_+ = \hat{\sigma}_z$

then $\hat{\sigma}_+\hat{\sigma}_- = (1 + \hat{\sigma}_z)/2$

$$\Downarrow$$

$$\frac{d}{dt}\langle\hat{\sigma}_+\hat{\sigma}_-\rangle = \frac{d}{dt}\left(\frac{\langle\hat{\sigma}_z\rangle}{2}\right)$$

- Hence, the 1st equation of the system of equations becomes $\frac{d}{dt}\langle\hat{\sigma}_+\hat{\sigma}_-\rangle = -\gamma\langle\hat{\sigma}_+\hat{\sigma}_-\rangle$

- We can then re-write the system of equations in an useful matrix form:

$$\frac{d}{dt}\begin{pmatrix}\langle\sigma_-\rangle\\ \langle\sigma_+\rangle\\ \langle\sigma_+\sigma_-\rangle\end{pmatrix} = \begin{bmatrix}-(\frac{\gamma}{2}+i\omega_A) & 0 & 0\\ 0 & -(\frac{\gamma}{2}-i\omega_A) & 0\\ 0 & 0 & -\gamma\end{bmatrix}\begin{bmatrix}\langle\sigma_-\rangle\\ \langle\sigma_+\rangle\\ \langle\sigma_+\sigma_-\rangle\end{bmatrix}$$

Let $\vec{S} \equiv \begin{pmatrix}\hat{\sigma}_-\\ \hat{\sigma}_+\\ \hat{\sigma}_+\hat{\sigma}_-\end{pmatrix}$; and $\hat{M} \equiv \begin{pmatrix}-(\frac{\gamma}{2}+i\omega_A) & 0 & 0\\ 0 & -(\frac{\gamma}{2}-i\omega_A) & 0\\ 0 & 0 & -\gamma\end{pmatrix}$

Then we can write the equation in the short form:

$$\frac{d}{dt}\vec{S}(t) = \hat{M}\cdot\vec{S}(t)$$

- We can now apply the quantum regression theorem to write equation for 2 time correlations. For example:

$$\frac{d}{d\tau}\langle\hat{\sigma}_-(t)\,\vec{S}(t+\tau)\rangle = \hat{M}\langle\hat{\sigma}_-(t)\,\vec{S}(t+\tau)\rangle$$

$$\frac{d}{d\tau}\langle\hat{\sigma}_+(t)\,\vec{S}(t+\tau)\rangle = \hat{M}\langle\hat{\sigma}_+(t)\,\vec{S}(t+\tau)\rangle$$

$$\frac{d}{d\tau}\langle\hat{\sigma}_+(t)\hat{\sigma}_-(t)\,\vec{S}(t+\tau)\rangle = \hat{M}\langle\hat{\sigma}_+(t)\hat{\sigma}_-(t)\,\vec{S}(t+\tau)\rangle$$

and many others.

* Let's consider a particular case.

Let's consider that the atom is initially in the excited state $|2\rangle$.

Then, initially $\langle\hat{\sigma}_+\rangle = \langle\hat{\sigma}_-\rangle = 0$

$$\langle\hat{\sigma}_+\hat{\sigma}_-\rangle = \langle\frac{\hat{\sigma}_z+1}{2}\rangle = 1$$

* From the differential equations for $\langle\hat{\sigma}_+\rangle, \langle\hat{\sigma}_-\rangle$, and $\langle\hat{\sigma}_+\hat{\sigma}_-\rangle$ we get easily that $\langle\vec{S}(t)\rangle = \begin{pmatrix} 0 \\ 0 \\ e^{-\gamma t}\end{pmatrix}$

* Hence $\langle\hat{\sigma}_-(t)\vec{S}(t)\rangle = \begin{pmatrix} 0 \\ 1-e^{-\gamma t} \\ 0\end{pmatrix}$; $\langle\hat{\sigma}_+(t)\vec{S}(t)\rangle = \begin{pmatrix} e^{-\gamma t} \\ 0 \\ 0\end{pmatrix}$; $\langle\hat{\sigma}_+(t)\hat{\sigma}_-(t)\vec{S}(t)\rangle = \begin{pmatrix} 0 \\ 0 \\ e^{-\gamma t}\end{pmatrix}$

using the properties of the Pauli matrices)

* Then, we can use these results as the initial conditions for the equations we got from the quantum regression theorem, to obtain

$$\langle\hat{\sigma}_-(t)\hat{\sigma}_+(t+\tau)\rangle = e^{i\omega_A\tau}e^{-\gamma\tau/2}(1-e^{-\gamma t})$$

$$\langle\hat{\sigma}_+(t)\hat{\sigma}_-(t+\tau)\rangle = e^{-i\omega_A\tau}e^{-\gamma\tau/2}e^{-\gamma t}$$

$$\langle\hat{\sigma}_+(t)\hat{\sigma}_-(t)\hat{\sigma}_+(t+\tau)\hat{\sigma}_-(t+\tau)\rangle = e^{-\gamma\tau}e^{-\gamma t}$$

* Of these expressions especially the second one has particular importance, because it provides the spontaneous emission spectrum, i.e. the probability of emitting a photon with a given frequency ω:

$$P(\omega) \propto \int_0^T dt\int_0^T dt'\, e^{-i\omega(t-t')}\langle\hat{\sigma}_+(t)\hat{\sigma}_-(t')\rangle$$

(this is the probability of detecting a photon with frequency ω in the interval between 0 and T) (we will see why later)

* Note that $\langle\hat{\sigma}_+(t+\tau)\hat{\sigma}_-(t)\rangle = \langle\hat{\sigma}_+(t)\hat{\sigma}_-(t+\tau)\rangle^*$

Hence for all t' and t:

$$\langle\hat{\sigma}_+(t)\hat{\sigma}_-(t')\rangle = e^{i\omega_A(t-t')}e^{-\frac{\gamma}{2}(t+t')}$$

Hence

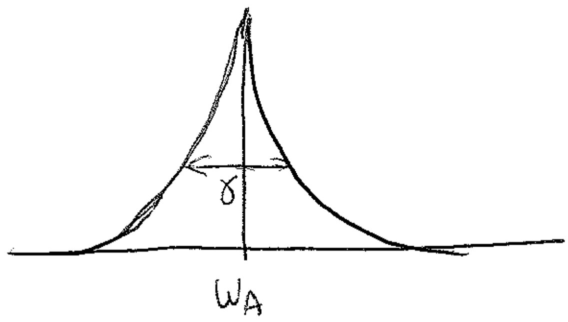
$$P(\omega) \propto \int_0^T dt\, e^{-\left[\frac{\gamma}{2} + i(\omega-\omega_A)\right]t} \int_0^T dt'\, e^{-\left[\frac{\gamma}{2} - i(\omega-\omega_A)\right]t'}$$

$$\propto \left[\frac{1 - e^{-\frac{\gamma}{2}T} e^{-i(\omega-\omega_A)T}}{\frac{\gamma}{2} + i(\omega-\omega_A)}\right]\left[\frac{1 - e^{-\frac{\gamma}{2}T} e^{i(\omega-\omega_A)T}}{\frac{\gamma}{2} - i(\omega-\omega_A)}\right]$$

For $T \gg 1/\gamma$:

$$\boxed{P(\omega) \propto \frac{1}{\left(\frac{\gamma}{2}\right)^2 + (\omega-\omega_A)^2}}$$

This gives the typical Lorentzian lineshape for the spectrum of the spontaneously emitted photons



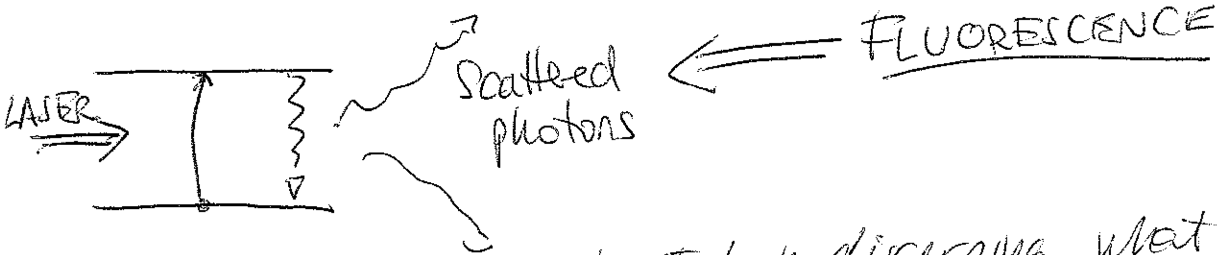
So, the spontaneously emitted photons do not have a defined frequency ω_A but a frequency $\omega_A \pm$ an uncertainty given by the line broadening γ.

• RESONANCE FLUORESCENCE

• Up to now we have employed the theory of Master equation to study the problem of spontaneous emission.

• We will now employ these techniques (and other ideas we have seen already) to study a paradigmatic problem of Quantum Optics, namely the so-called resonance fluorescence.

• In this problem we consider a 2-level atom irradiated by a strong monochromatic laser beam tuned to the atomic transition. Photons may be absorb from this beam and emitted to the many modes of the vacuum EM field as fluorescent scattering.



• As always we are first interested in discerning what is the system, what the reservoir, and what's the coupling.

* System:

• The incident laser mode is in a highly excited state that is essentially unaffected by its interaction with the atom.
• Our system is hence the atom interacting with the laser mode:

$$\hat{H}_S = \frac{\hbar\omega_A}{2}\hat{\sigma}_z - dE\left(e^{-i\omega_A t}\hat{\sigma}_+ + e^{i\omega_A t}\hat{\sigma}_-\right)$$

(remember that we consider the laser field as classical; the laser is in resonance with the transition)
($d \equiv$ Dipole; $E \equiv$ Electric field amplitude;

* Reservoir

• The reservoir is formed by the many modes of the EM field

$$\hat{H}_R = \sum_{k,\lambda} \hbar\omega_k \hat{r}^+_{k\lambda}\hat{r}_{k\lambda}$$

* Coupling

$$\hat{H}_{SR} = \sum_{k\lambda}\hbar\left(k^*_{k\lambda}\hat{r}^+_{k\lambda}\hat{\sigma}_- + k_{k\lambda}\hat{r}_{k\lambda}\hat{\sigma}_+\right)$$

where as previously $k_{k\lambda}$ are the coupling constants

* The master equation for the reduced density operator of the atom is identical to that for spontaneous emission, but adding a coherent part correspondent to the coherent driving term appearing in $\hat{H}_S$ now:

$$\frac{d\hat{\rho}}{dt} = -i\frac{\omega_A}{2}[\hat{\sigma}_z,\hat{\rho}] + \overbrace{i\frac{\Omega}{2}\left[e^{-i\omega_A t}\hat{\sigma}_+ + e^{i\omega_A t}\hat{\sigma}_-,\hat{\rho}\right]}^{\text{This is new now.}}$$
$$+\frac{\gamma}{2}\left(2\hat{\sigma}_-\hat{\rho}\hat{\sigma}_+ - \hat{\sigma}_+\hat{\sigma}_-\hat{\rho} - \hat{\rho}\,\hat{\sigma}_+\hat{\sigma}_-\right)$$

(note that now we neglect thermal effects, i.e. $\bar{n}=0$)

where $\Omega = dE/\hbar$ is the Rabi frequency (remember the discussion in page 37).

* From the master equation we can derive the equations of motion for the expectation values (exercise)

$$\frac{d}{dt}\langle\hat{\sigma}_-\rangle = -\left(\frac{\gamma}{2}+i\omega_A\right)\langle\hat{\sigma}_-\rangle - i\frac{\Omega}{2}e^{-i\omega_A t}\langle\hat{\sigma}_z\rangle$$

$$\frac{d}{dt}\langle\hat{\sigma}_+\rangle = -\left(\frac{\gamma}{2}-i\omega_A\right)\langle\hat{\sigma}_+\rangle + i\frac{\Omega}{2}e^{+i\omega_A t}\langle\hat{\sigma}_z\rangle$$

$$\frac{d}{dt}\langle\hat{\sigma}_z\rangle = i\Omega e^{-i\omega_A t}\langle\hat{\sigma}_+\rangle - i\Omega e^{i\omega_A t}\langle\hat{\sigma}_-\rangle - \gamma\left[\langle\hat{\sigma}_z\rangle + 1\right]$$

These equations are known as the optical Bloch equations and play a central role in the interaction of light with atoms.

* The solutions of these equations (initial condition: atom in the ground state) are of the form (exercise)

$$\langle\hat{\sigma}_z\rangle = -\frac{1}{\gamma^2+2\Omega^2}\left[\gamma^2 + 2\Omega^2 e^{-\frac{3\gamma}{4}t}\left[\cosh\kappa t + \frac{3\gamma}{4\kappa}\sinh\kappa t\right]\right]$$

$$\langle\hat{\sigma}_+\rangle = e^{i\omega_A t}\left[\begin{array}{l}\frac{\Omega\gamma}{\gamma^2+2\Omega^2}\left(1+e^{-\frac{3\gamma}{4}t}\left[\cosh\kappa t + \frac{3\gamma}{4\kappa}\sinh\kappa t\right]\right) \\ - i\frac{\Omega}{2\kappa}e^{-\frac{3\gamma}{4}t}\sinh\kappa t\end{array}\right] = \langle\hat{\sigma}_-\rangle^*$$

where $K = \sqrt{\left(\frac{\gamma}{4}\right)^2 - \Omega^2}$

* It's instructive to see what happens without spontaneous emission ($\gamma = 0$).

In that case $K = i\Omega \longrightarrow$ $\cosh Kt = \cos \Omega t$
$\sinh Kt = i \sin \Omega t$

Then:

$$\langle \hat{\sigma}_z \rangle = -\cos \Omega t$$

$$\left.\begin{aligned} \langle \hat{\sigma}_+ \rangle &= \frac{-i}{2} e^{i\omega_A t} \sin \Omega t \\ \langle \hat{\sigma}_- \rangle &= \frac{i}{2} e^{-i\omega_A t} \sin \Omega t \end{aligned}\right\} \begin{aligned} \langle \sigma_x \rangle &= \sin \omega_A t \sin \Omega t \\ \langle \sigma_y \rangle &= \cos \omega_A t \sin \Omega t \end{aligned}$$

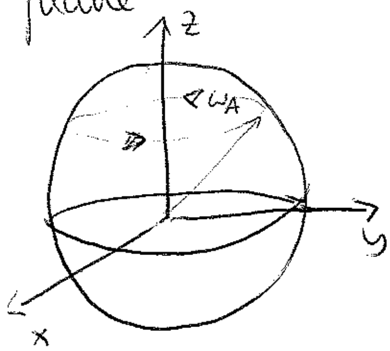
· Note that if we define a vector

$$\vec{v} = \langle \hat{\sigma}_x \rangle \hat{x} + \langle \hat{\sigma}_y \rangle \hat{y} + \langle \hat{\sigma}_z \rangle \hat{z}$$

This vector fulfills that $|\vec{v}|^2 = 1$, i.e. this vector has its tip at a spherical surface of radius 1.

This is the so-called <u>Bloch sphere</u>

. Note that the laser (ω_A) induces a fast rotation on the xy plane around the z axis.

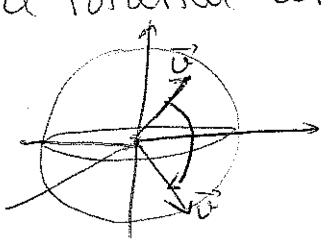


* We can define a rotating reference frame (rotating at frequency ω_A) such that we eliminate this fast rotation.

* Once we eliminate this, we can then take

$$\left.\begin{aligned} \langle \hat{\sigma}_x \rangle &= 0 \\ \langle \hat{\sigma}_y \rangle &= \sin \Omega t \\ \langle \hat{\sigma}_z \rangle &= \cos \Omega t \end{aligned}\right\}$$

So the Rabi oscillation induces a rotation around the x axis



* In the presence of spontaneous emission ($\gamma \neq 0$) it isn't anymore the case that $|\vec{v}|^2 = 1$ at any time.

One can actually prove (exercise) that

$$\frac{d}{dt}|\vec{v}|^2 = -\gamma\left[|\vec{v}|^2 - 1\right] - \gamma\left[\langle \sigma_z \rangle + 1\right]^2$$

- Note that after a given transient time (see the expression in page (101)) the system reaches a stationary state characterized by

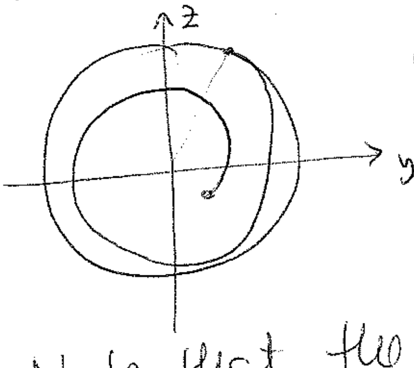
$$\langle \hat{\sigma}_z \rangle_{ss} = \frac{-\gamma^2}{\gamma^2 + 2\Omega^2}$$

$$\langle \hat{\sigma}_+ \rangle_{ss} = e^{i\omega_A t} \frac{2\gamma}{\gamma^2 + 2\Omega^2} = \langle \hat{\sigma}_- \rangle_s$$

In the stationary state $\quad |\vec{v}|_s^2 = 1 - \left[1 + \langle \hat{\sigma}_z \rangle_s\right]^2$

$$|\vec{v}|_{ss}^2 = 1 - \left[\frac{1}{1 + \frac{1}{2}\left(\frac{\gamma}{\Omega}\right)^2}\right]^2$$

Note that $|\vec{v}|_{ss} < 1$, and hence due to the spontaneous emission the vector $|\vec{v}|$ penetrates inside of the Bloch sphere.



(in the rotating frame)

- Note that the excited-state probability in the stationary state is $\quad P_{exc} = \frac{1 + \langle \hat{\sigma}_z \rangle}{2}$

$$P_{exc}^{ss} = \frac{\Omega^2}{\gamma^2 + 2\Omega^2}$$

* If $\gamma \gg \Omega$ (weak-coupling limit) $P_{exc}^{ss} \approx 0$ in the stationary limit (the system is completely overdamped)

On the contrary for very intense lasers $\gamma \ll \Omega$, then $P_{exc}^{ss} \cong \frac{1}{2}$ $\longrightarrow$ the atom becomes saturated with equal probability to be in the upper and in the lower levels.

• THE FLUORESCENCE SPECTRUM

• The master equation approach focuses on the dynamics of the atom. However, we will be also interested in the properties of the fluorescence. So, we will link now the scattered field to atomic operators.

• The scattered field is given as (Heisenberg picture)

$$\hat{E}(\vec{r},t) = \hat{E}^{(+)}(\vec{r},t) + \hat{E}^{(-)}(\vec{r},t)$$

where $\hat{E}^{(+)}(\vec{r},t) = i \sum_{k,\lambda} \sqrt{\frac{\hbar\omega_k}{2\epsilon_0 V}}\, \hat{e}_{k\lambda}\, \hat{r}_{k\lambda}(t)\, e^{i\vec{k}\vec{r}}$ (remember that $\hat{e}_{k\lambda}$ is the polarization vector)

$$\hat{E}^{(-)}(\vec{r},t) = \hat{E}^{(+)}(\vec{r},t)^{\dagger}$$

• We will be interested in correlation functions

$$G^{(1)}(t,t+\tau) = \langle \hat{E}^{(-)}(t)\, \hat{E}^{(+)}(t+\tau) \rangle$$

in particular because the Fourier transform of the $G^{(1)}$ function gives the spectrum of the scattered light

$$S(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-i\omega\tau} \langle E^{(-)}(t)\, E^{(+)}(t+\tau)\rangle\, d\tau$$

• let's relate $\hat{E}^{(\pm)}(\vec{r},t)$ with the atomic operators.

• From the Hamiltonian $\hat{H} = \hat{H}_S + \hat{H}_R + \hat{H}_{SR}$ we can obtain the Heisenberg equations of motion for the electromagnetic modes:

$$\dot{\hat{r}}_{k\lambda} = -i\omega_k \hat{r}_{k\lambda} - i\kappa^{*}_{k\lambda}\hat{\sigma}_-$$

let
$$\left.\begin{aligned} \hat{r}_{k\lambda} &= \hat{\tilde{r}}_{k\lambda}\, e^{-i\omega_k t} \\ \hat{\sigma}_- &= \hat{\tilde{\sigma}}_-\, e^{-i\omega_A t} \end{aligned}\right\} \frac{d}{dt}\hat{\tilde{r}}_{k\lambda} = -i\kappa^{*}_{k\lambda}\, \hat{\tilde{\sigma}}_-(t)\, e^{i(\omega_k-\omega_A)t}$$

Hence
$$\hat{\tilde{r}}_{k\lambda}(t) = \hat{\tilde{r}}_{k\lambda}(0) - i\kappa^{*}_{k\lambda}\int_0^t dt'\, \hat{\tilde{\sigma}}_-(t')\, e^{i(\omega_k-\omega_A)t'}$$

* Hence $\hat{E}(\vec{r},t)$ can be written as the contribution of 2 terms

$$\hat{E}(\vec{r},t) = \hat{E}_f(\vec{r},t) + \hat{E}_S(\vec{r},t)$$

• Free evolution of the electromagnetic field:

$$\hat{E}_f^{(+)}(\vec{r},t) = i \sum_{k\lambda} \sqrt{\frac{\hbar\omega_k}{2\epsilon_0 V}}\, \hat{e}_{k\lambda}\, \hat{r}_{k\lambda}(0)\, e^{-i(\omega_k t - \vec{k}\cdot\vec{r})}$$

• Source field radiated by the atom

$$\hat{E}_S^{(+)}(\vec{r},t) = i \frac{1}{2\epsilon_0 V} e^{-i\omega_A t} \sum_{k\lambda} \omega_k\, \hat{e}_{k\lambda}\, (\hat{e}_{k\lambda}\cdot\vec{d}_{12})\, e^{i\vec{k}(\vec{r}-\vec{r}_A)}$$

$$\times \int_0^t dt'\, \hat{\tilde{\sigma}}_-(t')\, e^{i(\omega_k - \omega_A)(t'-t)}$$

[Note: here we have used the explicit form of the coupling constant

$$\hbar_{2k\lambda} \equiv -i e^{i\vec{k}\cdot\vec{r}_A} \sqrt{\frac{\omega_k}{2\hbar\epsilon_0 V}}\; \hat{e}_{k\lambda}\cdot\vec{d}_{21}$$

with $\vec{d}_{21} = e\langle 2|\vec{r}|1\rangle$

$\vec{r}_A \equiv$ atom position (we take it as $\vec{r}_A = 0$)]

• Skipping the details, one can show that

$$\hat{E}_S^{(+)}(\vec{r},t) = -\frac{\omega_A^2}{4\pi\epsilon_0 c^2 r} (\vec{d}_{12}\times\vec{r})\times\vec{r}\;\; \hat{\sigma}_-(t - r/c)$$

This is actually the result for the radiation of a dipole with dipole-moment operator $\vec{d}_{12}\,\hat{\sigma}_-$.

* We are interested in correlations like $\langle E^{(-)}(t)\, E^{(+)}(t+\tau)\rangle$.
Note that the averages are taken over the vacuum electromagnetic field (we assume the temperature effects negligible). Hence $\hat{E}_f$ doesn't contribute to the averages. (Note: $\hat{E}_f^{(+)} \sim \hat{r}_{k\lambda}(0)$ which annihilates the vacuum).

· Hence only the source field contributes and

$$G^{(1)}\left(t+\frac{r}{c},\, t+\frac{r}{c}+\tau\right) = f(\vec{r}) \langle \hat{\sigma}_+(t)\,\hat{\sigma}_-(t+\tau)\rangle$$

where $f(\vec{r}) \equiv \left[\frac{\omega_A^2 d_{12}}{4\pi\epsilon_0 c^2}\right]^2 \frac{\sin^2\theta}{r^2}$ where θ = angle between $\vec{d}_{12}$ and $\vec{r}$

* We will be interested in the fluorescence spectrum in the stationary state

$$S(\omega) = f(\vec{r})\,\frac{1}{2\pi}\int_{-\infty}^{\infty} d\tau\, e^{i\omega\tau} \langle \hat{\sigma}_+(0)\,\hat{\sigma}_-(\tau)\rangle_{ss}$$

* Remember that in the stationary state

$$\langle \hat{\sigma}_\mp \rangle_{ss} = \pm i e^{\mp i\omega_A t} \frac{\Omega\gamma}{\gamma^2+2\Omega^2} \equiv e^{\mp i\omega_A t} \langle \hat{\tilde{\sigma}}_\mp \rangle_{ss}$$

$$\langle \hat{\sigma}_z \rangle_{ss} = \frac{-\gamma^2}{\gamma^2+2\Omega^2}$$

* However fluctuations about this steady state can occur, and they actually play a major role, as we will see in a moment.

$$\hat{\sigma}_z \equiv \langle \hat{\sigma}_z \rangle_{ss} + \Delta\hat{\sigma}_z$$ ← Fluctuations are intrinsic to quantum mechanics.

$$\hat{\tilde{\sigma}}_\pm \equiv \langle \hat{\tilde{\sigma}}_\pm \rangle_{ss} + \Delta\hat{\tilde{\sigma}}_\pm$$

Obviously $\langle \Delta\hat{\sigma}_z \rangle_{ss} = \langle \Delta\hat{\tilde{\sigma}}_\pm \rangle_{ss} = 0$ but $\langle \Delta\hat{\tilde{\sigma}}_+(0)\,\Delta\hat{\tilde{\sigma}}_-(\tau)\rangle \neq 0$

Hence $\langle \hat{\sigma}_+(0)\,\hat{\sigma}_-(\tau)\rangle_{ss} = e^{-i\omega_A\tau} \langle \hat{\tilde{\sigma}}_+(0)\,\hat{\tilde{\sigma}}_-(\tau)\rangle_{ss}$

$$= e^{-i\omega_A\tau}\left[\langle \hat{\tilde{\sigma}}_+(0)\rangle_{ss} \langle \hat{\tilde{\sigma}}_-(\tau)\rangle_{ss} + \langle \Delta\hat{\tilde{\sigma}}_+(0)\,\Delta\hat{\tilde{\sigma}}_-(\tau)\rangle\right]$$

* Hence the fluorescence spectrum decomposes into 2 parts

• Coherent component

$$S_{coh}(\omega) = f(\vec{r})\,\frac{1}{2\pi}\int_{-\infty}^{\infty} d\tau\; e^{i(\omega-\omega_A)\tau} \langle \hat{\tilde{\sigma}}_+ \rangle_{ss} \langle \hat{\tilde{\sigma}}_- \rangle_{ss}$$

$$= f(\vec{r})\,\frac{\Omega^2\gamma^2}{(\gamma^2+2\Omega^2)^2}\;\delta(\omega-\omega_A)$$

This is the part coming from the average stationary value of the operators. Note that the coherent component is a delta at exactly $\omega=\omega_A$.

• Incoherent component

$$S_{inc}(\omega) = f(\vec{r})\,\frac{1}{2\pi}\int_{-\infty}^{\infty} d\tau\; e^{i(\omega-\omega_A)\tau} \langle \Delta\hat{\tilde{\sigma}}_+(0)\,\Delta\hat{\tilde{\sigma}}_-(\tau) \rangle$$

• We need to evaluate $\langle \Delta\hat{\tilde{\sigma}}_+(0)\,\Delta\hat{\tilde{\sigma}}_-(\tau)\rangle$. For this we will use the quantum regression theorem.

Remember the Bloch equations (page 101)

$$\frac{d}{dt}\langle \Delta\hat{\tilde{\sigma}}_- \rangle = -i\frac{\Omega}{2}\langle \Delta\hat{\sigma}_z \rangle - \frac{\gamma}{2}\langle \Delta\hat{\tilde{\sigma}}_- \rangle$$

$$\frac{d}{dt}\langle \Delta\hat{\tilde{\sigma}}_+ \rangle = i\frac{\Omega}{2}\langle \Delta\hat{\sigma}_z \rangle - \frac{\gamma}{2}\langle \Delta\hat{\tilde{\sigma}}_+ \rangle$$

$$\frac{d}{dt}\langle \Delta\hat{\sigma}_z \rangle = i\Omega\langle \Delta\hat{\tilde{\sigma}}_+ \rangle - i\Omega\langle \Delta\hat{\tilde{\sigma}}_- \rangle - \gamma\langle \Delta\hat{\sigma}_z \rangle$$

let $\langle \Delta\vec{S} \rangle = \begin{pmatrix} \langle \Delta\hat{\tilde{\sigma}}_- \rangle \\ \langle \Delta\hat{\tilde{\sigma}}_+ \rangle \\ \langle \Delta\hat{\sigma}_z \rangle \end{pmatrix}$; $\hat{M} = \begin{bmatrix} -\gamma/2 & 0 & -i\Omega/2 \\ 0 & -\gamma/2 & i\Omega/2 \\ i\Omega & i\Omega & -\gamma \end{bmatrix}$

Then $\frac{d}{dt}\langle \Delta\vec{S}(t) \rangle = \hat{M}\langle \Delta\vec{S}(t) \rangle$

* The quantum regression theorem tells us that

$$\frac{d}{d\tau}\langle \Delta\hat{\sigma}_+(0)\,\Delta\vec{S}(\tau)\rangle_{ss} = \hat{M}\langle \Delta\hat{\sigma}_+(0)\,\Delta\vec{S}(\tau)\rangle_{ss}$$

The initial conditions are:

$\hat{\sigma}_+\hat{\sigma}_z = -\hat{\sigma}_+$

$\hat{\sigma}_-\hat{\sigma}_z = \hat{\sigma}_-$

$$\langle \Delta\hat{\sigma}_+(0)\,\Delta\vec{S}(0)\rangle_{ss} = \begin{pmatrix} \langle \hat{\sigma}_+\hat{\sigma}_-\rangle_{ss} - \langle\hat{\sigma}_+\rangle_{ss}\langle\hat{\sigma}_-\rangle_{ss} \\ \langle \hat{\sigma}_+^2\rangle_{ss} - \langle\hat{\sigma}_+\rangle_{ss}^2 \\ \langle \hat{\sigma}_+\hat{\sigma}_z\rangle_{ss} - \langle\hat{\sigma}_+\rangle_{ss}\langle\hat{\sigma}_z\rangle_{ss} \end{pmatrix} =$$

$$= \begin{bmatrix} \frac{1}{2}(1+\langle\hat{\sigma}_z\rangle_{ss}) - \langle\hat{\sigma}_+\rangle_{ss}\langle\hat{\sigma}_-\rangle_{ss} \\ -\langle\hat{\sigma}_+\rangle_{ss}^2 \\ -\langle\hat{\sigma}_+\rangle_{ss}\left[1+\langle\hat{\sigma}_z\rangle_{ss}\right] \end{bmatrix} = \frac{\Omega^2\gamma^2}{(\gamma^2+2\Omega^2)^2}\begin{bmatrix} 2\Omega^2/\gamma^2 \\ 1 \\ i\,2\Omega/\gamma \end{bmatrix}$$

* Solving the system of equations, and using this initial condition we reach to the following expression:

$$\langle \Delta\hat{\sigma}_+(0)\,\Delta\hat{\sigma}_-(\tau)\rangle_{ss} = \frac{1}{2}\,\frac{\Omega^2}{\gamma^2+2\Omega^2}\,e^{-\frac{\gamma}{2}\tau}$$

$$-\frac{1}{4}\,\frac{\Omega^2}{(\gamma^2+2\Omega^2)^2}\left[\gamma^2-2\Omega^2+\frac{\gamma}{4k}(\gamma^2-10\Omega^2)\right]e^{-\left[\frac{3\gamma}{4}-k\right]\tau}$$

$$-\frac{1}{4}\,\frac{\Omega^2}{(\gamma^2+2\Omega^2)^2}\left[\gamma^2-2\Omega^2-\frac{\gamma}{4k}(\gamma^2-10\Omega^2)\right]e^{-\left[\frac{3\gamma}{4}+k\right]\tau}$$

* Remember that the Fourier transform of an exponential is a Lorentzian, so we expect actually that the incoherent spectrum is composed by 3 Lorentzians.

Let's see this in more detail.

* For $\Omega \gg \gamma/4 \longrightarrow \kappa \simeq i\Omega$

and

$$\langle \Delta\hat{\sigma}_+(0)\Delta\hat{\sigma}_-(\tau)\rangle \simeq \frac{1}{4}e^{-\frac{\gamma}{2}\tau} + \frac{1}{8}e^{-\frac{3\gamma}{4}\tau}e^{i\Omega\tau} + \frac{1}{8}e^{-3\frac{\gamma}{4}\tau}e^{-i\Omega\tau}$$

Hence

$$S_{inc}(\omega) = \frac{f(\vec{r})}{2\pi}\int_{-\infty}^{\infty} d\tau \, \langle \Delta\hat{\sigma}_+(0)\Delta\hat{\sigma}_-(\tau)\rangle \, e^{+i(\omega-\omega_A)\tau}$$

$$= \frac{f(\vec{r})}{2\pi}\int_{-\infty}^{\infty} d\tau \left[\frac{1}{4}e^{-\frac{\gamma}{2}|\tau|}e^{i(\omega-\omega_A)\tau} + \frac{1}{8}e^{-\frac{3\gamma}{4}|\tau|}e^{i(\omega-\omega_A+\Omega)\tau} + \frac{1}{8}e^{-\frac{3\gamma}{4}|\tau|}e^{i(\omega-\omega_A-\Omega)\tau}\right]$$

Remember that

$$\int_{-\infty}^{\infty} d\tau \, e^{-\frac{\Gamma}{2}|\tau|}e^{i(\omega-\omega_0)\tau} = \frac{\Gamma}{(\Gamma/2)^2+(\omega-\omega_0)^2} \longleftarrow \text{Lorentzian shape.}$$

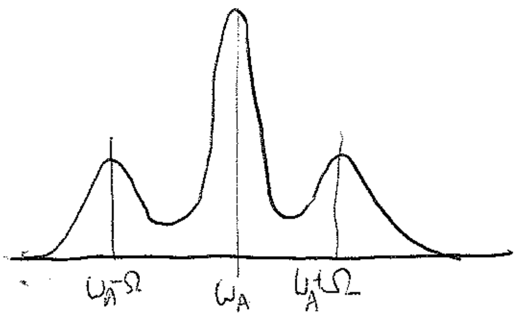
Hence:

$$S_{inc}(\omega) = \frac{f(\vec{r})}{2\pi}\left\{\frac{1}{4}\frac{\gamma}{\left(\frac{\gamma}{2}\right)^2+(\omega-\omega_A)^2} + \frac{1}{8}\frac{3\gamma/4}{\left(\frac{3\gamma}{8}\right)^2+(\omega-\omega_A+\Omega)^2} + \frac{1}{8}\frac{3\gamma/4}{\left(\frac{3\gamma}{8}\right)^2+(\omega-\omega_A-\Omega)^2}\right\}$$

So for a strong laser field the fluorescence spectrum splits into 3 peaks centered at ω_A and $\omega_A \pm \Omega$.

This is the so-called

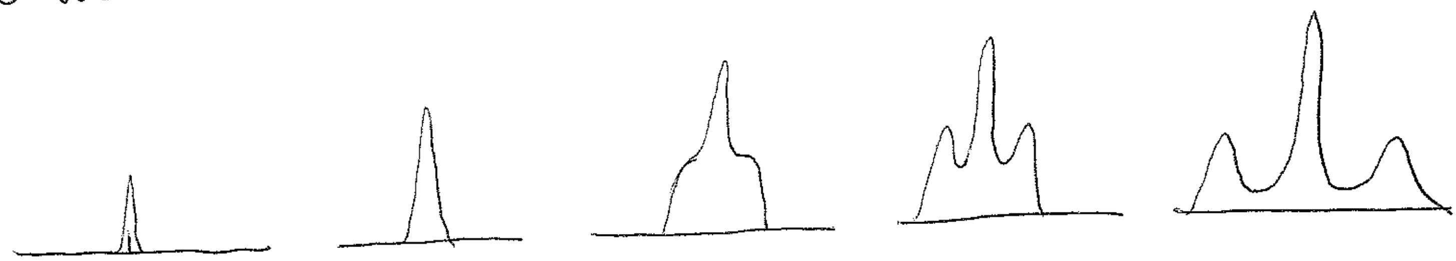
<u>Mollow Triplet</u>



* Note that for low intensities $\Omega \ll \gamma$ the incoherent component is negligible and hence

$$S(\omega) \cong S_{coh}(\omega) \propto \delta(\omega-\omega_A)$$

So we recover a sharp peak at $\omega=\omega_A$



GROWING Ω

* Let's try to understand the physics behind Mollow's triplet. A very intuitive picture is obtained from the so-called dressed-state formalism.

* Let's recall some ideas we have already seen when discussing in previous lectures the interaction of light with atoms. Let's forget for the moment the spontaneous damping.
* If you remember in the fully quantized form the interaction of a two level atom with a single mode is provided by the Jaynes-Cummings Hamiltonian (remember page 31; note that now I use a slightly different notation):

$$\hat{H} = \frac{1}{2}\hbar\omega_A\hat{\sigma}_z + \hbar\omega_A\hat{a}^\dagger\hat{a} + \hbar(K\hat{a}\hat{\sigma}_+ + K^*\hat{a}^\dagger\hat{\sigma}_-)$$

(Note that we consider that the laser frequency is exactly that of the atomic transition, and hence the detuning Δ (see eg. page 32) is zero)

* Remember that for a given number of photons n the states $\{|n+1,1\rangle, |n,2\rangle\}$ form a closed set, since

$$\hat{H}|n,2\rangle = (n+1/2)\hbar\omega_A |n,2\rangle + \sqrt{n+1}\,\hbar\kappa^* |n+1,1\rangle$$

$$\hat{H}|n+1,1\rangle = \sqrt{n+1}\,\hbar\kappa |n,2\rangle + (n+1/2)\hbar\omega_A |n+1,1\rangle$$

Solving the eigenproblem $E_{n,\pm}|\psi_{n,\pm}\rangle = \hat{H}|\psi_{n,\pm}\rangle$

we obtain [p. 32 at the bottom]

$$E_{n,\pm} = \hbar\omega_A(n+\tfrac{1}{2}) \pm \sqrt{n+1}\,\hbar|\kappa|$$

The eigenstates are the so-called dressed states

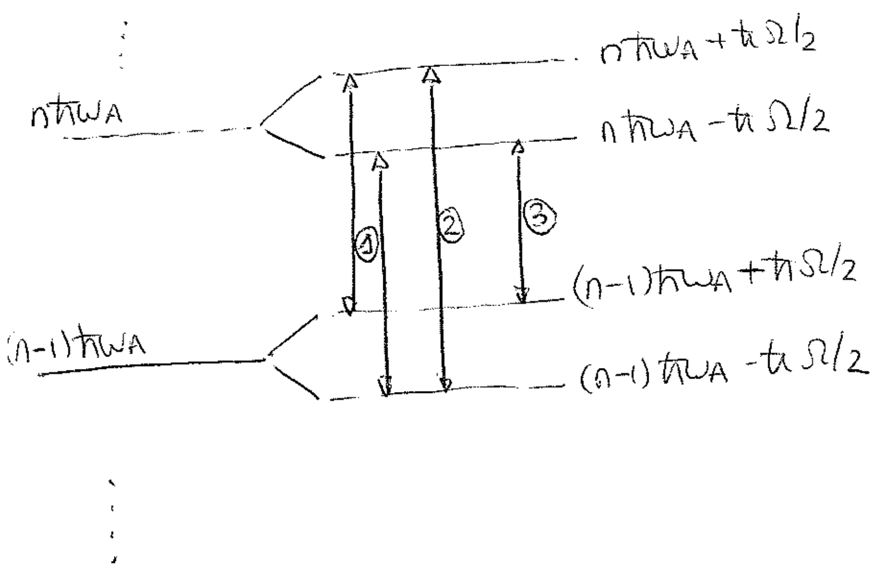
$$|\psi_{n,+}\rangle = \cos\phi\,|n+1,1\rangle + \sin\phi\,|n,2\rangle \qquad \text{with } \phi = \frac{\pi}{4}$$

$$|\psi_{n,-}\rangle = -\sin\phi\,|n+1,1\rangle + \cos\phi\,|n,2\rangle$$

* Remember that the laser field is assumed to be a coherent state with a very large mean photon number $\bar{n} \gg 1$. Hence (recall our discussion in page 37)

$$\sqrt{n+1}\,\hbar|\kappa| \simeq \hbar\Omega/2 \qquad \text{where } \Omega \equiv \text{Rabi frequency}$$

Hence $E_{n,\pm} \simeq n\hbar\omega_A \pm \hbar\Omega/2$



Since the dressed states contain a probability to be in $|2\rangle$, they (both) decay spontaneously. From the picture you can see that 3 possible processes occur.

* Processes ① involve transitions between
  * $(n\hbar\omega_A + \hbar\Omega/2) \longrightarrow [(n-1)\hbar\omega_A + \hbar\Omega/2]$
  * $[n\hbar\omega_A - \hbar\Omega/2] \longrightarrow [(n-1)\hbar\omega_A - \hbar\Omega/2]$

These processes involve an energy jump $\Delta E_{①} = \hbar\omega_A$

* Processes ② are of the form:
  * $[n\hbar\omega_A + \hbar\Omega/2] \longrightarrow [(n-1)\hbar\omega_A - \hbar\Omega/2]$

They therefore involve $\Delta E_{②} = \hbar\omega_A + \hbar\Omega$

* Processes ③ are:
  * $[n\hbar\omega_A - \hbar\Omega/2] \longrightarrow [(n-1)\hbar\omega_A + \hbar\Omega/2]$

Hence $\Delta E_{③} = \hbar\omega_A - \hbar\Omega$

* Therefore the three peaks of Mollow's triplet are directly related with the possible transitions between dressed states.
* Why the peaks dissapear when $\Omega \ll \gamma$? This is very easy to understand. Remember that the spontaneous emission at a given central frequency is not a delta-spectrum but a Lorentzian of width γ. Therefore if the peaks are separated by a frequency Ω, but the peak width is $\gamma \gg \Omega$, then simply one cannot resolve the peaks [remember the picture in p. 111]

- SECOND ORDER COHERENCE OF THE FLUORESCENCE

* Up to now we just had a look into the first-order correlation functions $G^{(1)}$.

* Remember that we split $G^{(1)}$ into 2 parts

$$G^{(1)}_{coh} = f(\vec{r}) \langle \hat{\sigma}_+ \rangle_{ss} \langle \hat{\sigma}_- \rangle_{ss}$$

(it's 1st order coherent because it factorizes, remember the discussion of p. 46)

and

$$G^{(1)}_{incoh} = f(\vec{r}) \langle \Delta \hat{\sigma}_+(0) \Delta \hat{\sigma}_-(\tau) \rangle_{ss}$$

* Note that at $\tau = 0$, $G^{(1)}_{incoh}(\tau=0) = 0$ (see p. 109) and hence $G^{(1)}(0) = G^{(1)}_{coh}$; and as a consequence $g^{(1)}(0) = 1$.

* However all this discussion just gives us information about first-order coherence. As we have seen already, important information about the quantum nature of light is obtained from the 2nd-order correlation functions:

$$G^{(2)}(t, t+\tau) \equiv \langle \hat{E}^{(-)}(t) \hat{E}^{(-)}(t+\tau) \hat{E}^{(+)}(t+\tau) \hat{E}^{(+)}(t) \rangle$$

As for the $G^{(1)}$ function we can relate $G^{(2)}$ with the atomic operators:

$$G^{(2)}\left(t+\frac{r}{c}, t+\frac{r}{c}+\tau\right) = f(\vec{r})^2 \langle \hat{\sigma}_+(t) \hat{\sigma}_+(t+\tau) \hat{\sigma}_-(t+\tau) \hat{\sigma}_-(t) \rangle$$

In the stationary state

$$G^{(2)}_{ss}(\tau) = f(\vec{r})^2 \langle \hat{\sigma}_+(0) \hat{\sigma}_+(\tau) \hat{\sigma}_-(\tau) \hat{\sigma}_-(0) \rangle$$

* Remember that $\hat{\sigma}_+ \hat{\sigma}_- = \frac{1}{2}(1 + \hat{\sigma}_z)$

Hence:

$$G_{ss}^{(2)}(\tau) = f(\vec{r})^2 \, \frac{1}{2} \left[\langle \hat{\sigma}_+ \hat{\sigma}_- \rangle_{ss} + \langle \hat{\sigma}_+(0)\, \hat{\sigma}_z(\tau)\, \hat{\sigma}_-(0) \rangle \right]$$

* We will find the expression for $G_{ss}^{(2)}(\tau)$ by means of the quantum regression theorem.

* Remember the optical Bloch equations, which we write in a matrix form: $\frac{d}{dt}\langle \vec{S} \rangle = \hat{M} \langle \vec{S} \rangle + \vec{b}$

Remember that $\langle \vec{S} \rangle = \begin{pmatrix} \hat{\tilde{\sigma}}_- \\ \hat{\tilde{\sigma}}_+ \\ \hat{\sigma}_z \end{pmatrix}$ (we employ the rotating frame at frequency ω_A : $\hat{\sigma}_\pm \to \hat{\tilde{\sigma}}_\pm$)

and $\hat{M} = \begin{bmatrix} -(\gamma/2) & 0 & -i\Omega/2 \\ 0 & -\gamma/2 & i\Omega/2 \\ -i\Omega & i\Omega & -\gamma \end{bmatrix}$ and $\vec{b} = \begin{pmatrix} 0 \\ 0 \\ -\gamma \end{pmatrix}$

* We can solve this equation in a formal way which will be later on useful:

$$\frac{d}{dt}\left[\langle \vec{S} \rangle + \hat{M}^{-1} \cdot \vec{b} \right] = \hat{M} \left[\langle \vec{S} \rangle + \hat{M}^{-1} \cdot \vec{b} \right]$$

Hence $\langle \vec{S}(t) \rangle = -\hat{M}^{-1}\vec{b} + e^{\hat{M}t} \left[\langle \vec{S}(0) \rangle + \hat{M}^{-1} \cdot \vec{b} \right]$

* Now we employ the quantum regression theorem:

$$\frac{d}{d\tau} \langle \hat{\sigma}_+(0)\, \vec{S}(\tau)\, \hat{\sigma}_-(0) \rangle_{ss} = \hat{M} \langle \hat{\sigma}_+(0)\, \vec{S}(\tau)\, \hat{\sigma}_-(0) \rangle_{ss} + \langle \hat{\sigma}_+ \hat{\sigma}_- \rangle_{ss}\, \vec{b}$$

Hence

$$\frac{d}{d\tau}\left[\langle \hat{\sigma}_+(0)\vec{S}(\tau)\hat{\sigma}_-(0) \rangle_{ss} + \langle \hat{\sigma}_+ \hat{\sigma}_- \rangle_{ss}\, \hat{M}^{-1}\vec{b} \right] =$$

$$= \hat{M} \left[\langle \hat{\sigma}_+(0)\vec{S}(\tau)\hat{\sigma}_-(0) \rangle_{ss} + \langle \hat{\sigma}_+ \hat{\sigma}_- \rangle_{ss}\, \hat{M}^{-1}\vec{b} \right]$$

and therefore:

$$\langle \hat{\sigma}_+(0)\vec{S}(\tau)\hat{\sigma}_-(0)\rangle_{ss} = -\langle\hat{\sigma}_+\hat{\sigma}_-\rangle_{ss}\,\hat{M}^{-1}\vec{b} + e^{\hat{M}\tau}\left[\langle\hat{\sigma}_+\vec{S}\hat{\sigma}_-\rangle_{ss} + \langle\hat{\sigma}_+\hat{\sigma}_-\rangle_{ss}\,\hat{M}^{-1}\vec{b}\right]$$

$$\langle\hat{\sigma}_+\vec{S}\hat{\sigma}_-\rangle_{ss} = \begin{pmatrix}\langle\hat{\sigma}_+\hat{\sigma}_-\hat{\sigma}_-\rangle_{ss}\\ \langle\hat{\sigma}_+\hat{\sigma}_+\hat{\sigma}_-\rangle_{ss}\\ \langle\hat{\sigma}_+\hat{\sigma}_z\hat{\sigma}_-\rangle_{ss}\end{pmatrix} = \langle\hat{\sigma}_+\hat{\sigma}_-\rangle_{ss}\begin{pmatrix}0\\0\\-1\end{pmatrix}$$

Hence:

$$\langle\hat{\sigma}_+(0)\vec{S}(\tau)\hat{\sigma}_-(0)\rangle_{ss} = \langle\hat{\sigma}_+\hat{\sigma}_-\rangle_{ss}\left\{-M^{-1}\vec{b} + e^{\hat{M}\tau}\left[\begin{pmatrix}0\\0\\-1\end{pmatrix} + \hat{M}^{-1}\vec{b}\right]\right\}$$

But $-\hat{M}^{-1}\vec{b} + e^{\hat{M}\tau}\left[\begin{pmatrix}0\\0\\-1\end{pmatrix} + \hat{M}^{-1}\vec{b}\right] = \langle\vec{S}(\tau)\rangle_{|1\rangle}$ when the initial condition is the atom in $|1\rangle$

Hence

$$G^{(2)}_{SS}(\tau) = f(\vec{r})^2\,\frac{1}{2}\langle\hat{\sigma}_+\hat{\sigma}_-\rangle_{ss}\left[1 + \langle\hat{\sigma}_z(\tau)\rangle_{|1\rangle}\right]$$

$$= f(\vec{r})^2\; P_2^{ss}\; P_2^{|1\rangle}(\tau)$$

where $P_2^{ss} \equiv$ Probability to be in $|2\rangle$ in the stationary state

$P_2^{|1\rangle}(\tau) \equiv$ Probability to be in $|2\rangle$ after a time τ ~~when~~ the atom starts at time $\tau=0$ in $|1\rangle$.

Using the results we obtain before we get

$$G^{(2)}_{SS}(\tau) = f(\vec{r})^2\left(\frac{\Omega^2}{\gamma^2+2\Omega^2}\right)^2\left[1 - e^{-3\gamma\tau/4}\left[\cosh kt + \frac{3\gamma}{4k}\sinh kt\right]\right]$$

Remember that $G^{(1)}_{SS}(0) = f(\vec{r})\,\frac{\Omega^2}{\gamma^2+2\Omega^2}$ (coming from the coherent contribution)

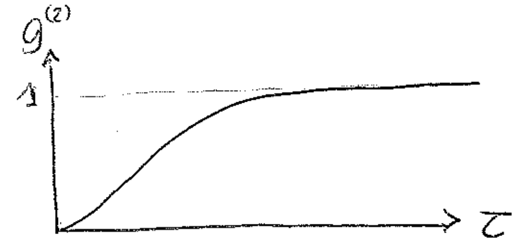
* Hence, the normalized 2nd order correlation function is finally

$$g^{(2)}(\tau) = \frac{G_{SS}^{(2)}(\tau)}{|G_{SS}^{(1)}(0)|^2} = 1 - e^{-3\gamma\tau/4}\left[\cosh k\tau + \frac{3\gamma}{4k}\sinh k\tau\right]$$

* One can see that $g^{(2)}(0) = 0$, i.e. the fluorescent light exhibits <u>photon antibunching</u>

* For low incident light intensities ($\Omega \ll \gamma/4$) $g^{(2)}(\tau)$ becomes a monotonic function:

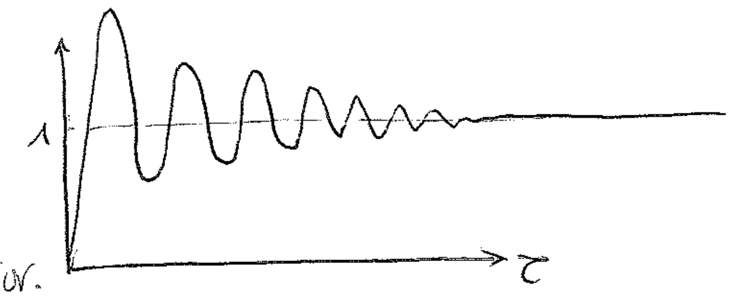
$$g^{(2)}(\tau) \cong \left(1 - e^{-\gamma\tau/2}\right)^2$$



* For strong coupling ($\Omega >> \gamma/4$)

$$g^{(2)}(\tau) \cong 1 - e^{-3\gamma\tau/4}\cos\Omega\tau$$

i.e. $g^{(2)}$ shows an oscillatory behavior.



* Therefore in resonance fluorescence we find a tendency of the photons to be separated (antibunching). It is very easy to understand why it is so.

Suppose that we detect a photon. This means that since the atom has decayed, right after the emission of the 1st photon the system is in $|1\rangle$. Any subsequent emission must begin with an excited atom. But there's a delay corresponding to the time taken for the atom to be re-excited. This is clear from the form of $G_{SS}^{(2)} \Rightarrow P_2^{SS} \; P_S^{|1\rangle}(\tau)$

we emit at time 0

we start in $|1\rangle$ and need some time to emit again.