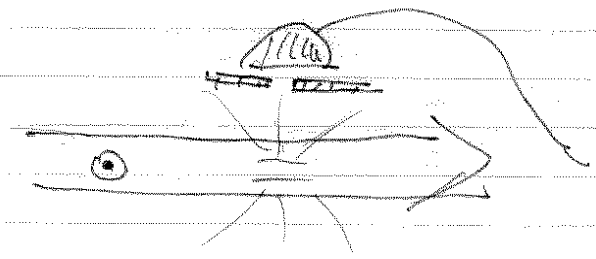
Lecture 23: Resonance Fluorescence

Historically, the phenomenon of (near) resonance fluorescence has played an important role in the development of quantum optics, including nonclassical spectrum, photon antibunching, squeezing, and quantum jumps. We have seen some of these earlier in the semester in our discussions of nonclassical light. We now revisit this phenomenon with our newly developed formalism of the master equation and Heisenberg-Langevin equations.

The problem

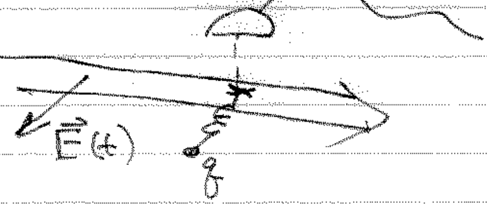
An atom (two-level) is driven "coherently" near resonance. The atom "fluoresces", scattering light into other directions. For simplicity we will take the driving field to be a linearly polarized (out of the page) plane wave, and we detect the fluorescence in the third orthogonal direction



Our goal is to understand the properties of the scattered light, most particular here, its spectrum.

Classical Problem

Let us first consider the classical analog : Scattering by Lorentz oscillator



$$\vec{E}(t) = \vec{E}_0 \cos\omega_L t = \mathrm{Re}\, \vec{E}_0 e^{-i\omega_L t}$$

In the dipole approximation, the radiated field

$$\vec{E}_{rad}(t) \propto \ddot{\vec{d}}_{\perp}(t_{ret}) \qquad t_{ret} = t - \frac{|\vec{x}-\vec{x}'|}{c}$$

(transverse component)

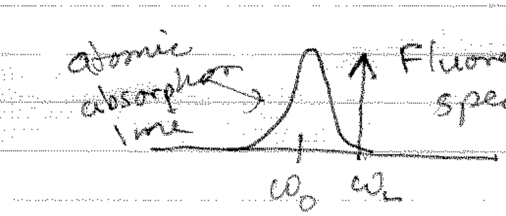
We take the Lorentz oscillator to have a natural resonance ω_0 and decay rate Γ. In steady state, the oscillator oscillates at the driving frequency ω_L not the natural resonance.

The amplitude of that oscillation (and its phase) will depend on the detuning. Taking the detector $\perp$ to the incident polarization $\vec{d}_\perp = \vec{d}$

$$\Rightarrow \quad \vec{E}_{rad}(t) \propto \mathrm{Re}\left(k_L^2\, \underset{\sim}{d}\, e^{-i\omega t}\right)$$

$$\underset{\sim}{d} = \underset{\sim}{\alpha} E_0 \propto \frac{1}{-\Delta - i\frac{\Gamma}{2}} E_0$$

The spectrum of the scattered light is a delta function, centered at ω_L, equal to spectrum of the incident light

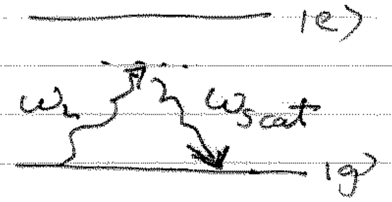


This is Rayleigh scattering. It explain why the sky is blue.

Now let us return to the <u>quantum problem</u>

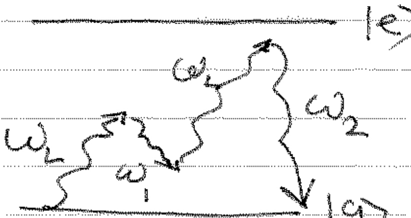
We can think of scattering as arising in perturbation theory.

- For weak fields, the lowest order perturbation process



The scattering is elastic $\omega_{scat} = \omega_L$

- For stronger fields, nonlinear multi-photon processes are possible



ω_1 and ω_2 are "sidebands"

$\omega_1 + \omega_2 = 2\omega_L$

At very high intensities perturbation theory breaks down all together, and we see quite a different spectrum from the elastic scattering expected in first order.

This picture agrees with what we know already about the driven, two-level atom. At low intensities such that the <u>saturation parameter</u> is small, the atom responds like a Lorentz oscillator. For higher intensities, the atom responds nonlinearly, leading to Rabi flopping, etc.

Quantum Problem

Atom driven by monochromatic laser and vacuum

$$\hat{H}_{total} = \hat{H}_A + \hat{H}_{AL} + \hat{H}_V + \hat{H}_{AV}$$

In the rotating frame, and rotating wave approximation

$$\hat{H}_A = -\frac{\hbar\Delta}{2}\hat{\sigma}_z \qquad \Delta_L = \omega_L - \omega_{eg}$$

$$\hat{H}_{AL} = -\frac{\hbar\Omega}{2}(\hat{\sigma}_+ + \hat{\sigma}_-) \qquad \Omega = d_{eg}E_0/\hbar$$

$$\hat{H}_V = \sum_j{}' \hbar\omega_j \hat{b}_j^\dagger \hat{b}_j = \int d^3x \left(\frac{\hat{E}_V^2 + \hat{B}_V^2}{8\pi}\right)$$

$$\hat{H}_{AV} = -\hat{\vec{d}}\cdot\vec{E}_V \cong -d_{eg}\left(e^{i\omega_L t}\hat{\sigma}_+\hat{E}_V^{(+)} + e^{-i\omega_L t}\hat{\sigma}_-\hat{E}_V^{(-)}\right)$$

The vacuum is a zero temperature reservoir for atom.

- Schrödinger Picture $\Rightarrow$ Master equation

$$\frac{d\hat{\rho}}{dt} = -\frac{i}{\hbar}[\hat{H},\rho] + \mathcal{L}_{relax}[\hat{\rho}]$$

$$\hat{H} = \hat{H}_A + \hat{H}_{AL} \qquad \mathcal{L}_{relax}[\hat{\rho}] = -\frac{\Gamma}{2}\left(\{\hat{a}^\dagger\hat{a},\hat{\rho}\} - 2\hat{a}\rho\hat{a}^\dagger\right)$$

- Heisenberg picture $\Rightarrow$ Langevin equation (Optical-Bloch plus noise)

$$\frac{d\hat{\sigma}_+}{dt} = -\frac{i}{\hbar}[\hat{\sigma}_+, \hat{H}_{tot}] = -i\Delta_L\hat{\sigma}_+ - i\frac{\Omega}{2}\hat{\sigma}_z + i\frac{d_{eg}}{\hbar}\hat{E}_V^{(-)}\hat{\sigma}_z$$

$$\frac{d\hat{\sigma}_z}{dt} = -\frac{i}{\hbar}[\hat{\sigma}_z, \hat{H}_{tot}] = i\Omega(\hat{\sigma}_- - \hat{\sigma}_+) + 2i\frac{d_{eg}}{\hbar}\left(\hat{E}_V^{(+)}\hat{\sigma}_+ e^{-i\omega_L t} - \hat{E}_V^{(-)}\hat{\sigma}_- e^{i\omega_L t}\right)$$

$$\frac{d\hat{a}_k}{dt} = -\frac{i}{\hbar}[\hat{a}_k, \hat{H}_{tot}] = -i\omega_k\hat{a}_k - g_k\hat{\sigma}_- e^{i\omega_L t}$$

$$\left(\hbar g_k = \sqrt{\frac{2\pi\hbar\omega}{V}}\, d_{eg}\right)$$

~~Off to "interaction picture" $\hat{\sigma}_+ \to \hat{\sigma}_+ e^{-i\omega t}$~~

Formal integration of vacuum modes

$$\Rightarrow \hat{b}_k(t) = \hat{b}_{vac}(t) + \hat{b}_{Source}(t)$$

$$\hat{b}_{vac}(t) = \hat{b}_k(0)\, e^{-i\omega_k t}$$

$$\hat{b}_{Source}(t) = -g_k \int_0^t dt'\, \hat{\sigma}_-(t')\, e^{-i\Delta_k(t-t')} \qquad (\Delta_k = \omega_L - \omega_k)$$

$$\hookrightarrow \hat{E}^{(+)}_{Source}(\vec{x},t) \propto e^{-i\omega_L(t-\frac{r}{c})}\, \hat{\sigma}_-(t-\tfrac{r}{c})$$

Plugging $\hat{b}_k(t)$ into atomic e.o.f. and tracing over vac

In Heisenberg picture

$$\boxed{\begin{aligned} \Rightarrow \frac{d\hat{\sigma}_+}{dt} &= \left(-i\Delta_L - \frac{\Gamma}{2}\right)\hat{\sigma}_+ - i\frac{\Omega}{2}\hat{\sigma}_z + \hat{\mathcal{F}}_+ \\ \frac{d}{dt}\hat{\sigma}_z &= -i\Omega(\hat{\sigma}_+ - \hat{\sigma}_-) - \Gamma\hat{\sigma}_z + \hat{\mathcal{F}}_z \\ \hat{\mathcal{F}}_+ = i\frac{d_{eg}}{\hbar}\hat{E}^{(+)}_v\hat{\sigma}_z \quad &, \quad \hat{\mathcal{F}}_z = i\frac{d_{eg}}{\hbar}(\hat{\sigma}_+ \underset{e^{-i\omega_L t}}{\hat{E}^{(-)}} + h.c.) \end{aligned}}$$

Of course, mean values give the familiar Optical Bloch equation $\langle\hat{\sigma}_+\rangle = u + iv = \rho_{ge}$ $\quad\langle\hat{\sigma}_z\rangle = w = \rho_{ee} - \rho_{gg}$

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

$$\frac{d}{dt}\langle\hat{\sigma}_z\rangle = -i\Omega(\hat{\sigma}_+ - \hat{\sigma}_-) - \Gamma\langle\hat{\sigma}_z\rangle$$

Coherent vs incoherent fluorescence

The radiated field (up to constants)

$$\hat{E}^{(+)}(r,t) = e^{-i\omega_L(t-\frac{r}{c})}\,\hat{\sigma}_-(t-\tfrac{r}{c})$$

(shift origin of time) $t \Rightarrow t-\frac{r}{c}$

Intensity $\hat{I} = \hat{E}^{(-)}(t)\,\hat{E}^{(+)}(t) = \hat{\sigma}_+(t)\,\hat{\sigma}_-(t) = (|e\rangle\langle e|)(t)$

$\Rightarrow$ Total radiation rate $\propto$ total population in excited state

The "coherent" part of scatter field

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

$$\Rightarrow \hat{\sigma}_-(t) = \underbrace{\langle\hat{\sigma}_-(t)\rangle}_{\text{coherent}} + \underbrace{\delta\hat{\sigma}_-}_{\text{fluctuation}}$$

$$\Rightarrow \langle \hat{I}\rangle = \langle \hat{\sigma}_+(t)\,\hat{\sigma}_-(t)\rangle = \underbrace{\langle\hat{\sigma}_+(t)\rangle\langle\hat{\sigma}_-(t)\rangle}_{I_{coh}} + \underbrace{\langle\delta\hat{\sigma}_+(t)\,\delta\hat{\sigma}_-(t)\rangle}_{I_{incoh}}$$

$$I_{coh} = |\langle\hat{\sigma}_+\rangle|^2 = |u+iv|^2$$

$$I_{incoh} = I - I_{coh} = \langle |e\rangle\langle e|(t)\rangle - |\langle\hat{\sigma}_+\rangle|^2$$

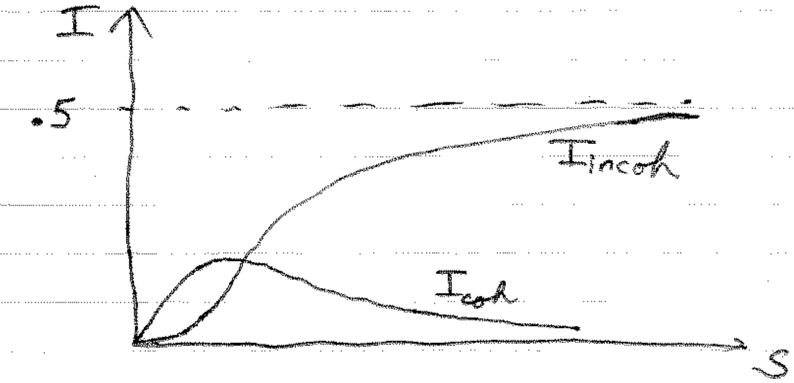
$$= P_e(t) - |u+iv|^2$$

In steady state
$$\begin{cases} u+iv = \dfrac{s}{1+s}\left(\dfrac{\Delta}{\Omega} + i\dfrac{\Gamma}{2\Omega}\right) \\[2ex] P_e = \dfrac{s/2}{1+s} \end{cases}$$

$s = \dfrac{\Omega^2/2}{\Delta^2+\Gamma^2/4}$ sat param

$$\Rightarrow \; I_{coh} = \frac{s^2}{(1+s)^2}\left(\frac{\Delta^2 + \Gamma^2/4}{\Omega^2}\right) = \frac{1}{2}\,\frac{s}{(1+s)^2}$$

$$I_{incoh} = \frac{1}{2}\frac{s}{(1+s)} - \frac{1}{2}\frac{s}{(1+s)^2} = \frac{1}{2}\left(\frac{s}{1+s}\right)^2$$



Weak field $s \ll 1 \Rightarrow$ Coherent (Rayleigh) scattering

$s \gg 1 \Rightarrow$ Incoherent

Spectrum of fluorescence

Need two-time correlation function

$$\langle \hat{E}^{(-)}(t+\tau)\,\hat{E}^{(+)}(t)\rangle = \langle \hat{\sigma}_+(t+\tau)\,\hat{\sigma}_-(t)\rangle\, e^{+i\omega_L \tau}$$

$$S(\omega) = \int d\tau \,\langle \hat{E}^{(-)}(t+\tau)\,\hat{E}^{(+)}(t)\rangle\, e^{-i\omega\tau}$$

$$= \int d\tau\, \langle \hat{\sigma}_+(t+\tau)\,\hat{\sigma}_-(t)\rangle\, e^{-i(\omega-\omega_L)\tau}$$

$$= S_{coh}(\omega) + S_{incoh}(\omega)$$

$\cancel{S_{coh} = \int d\tau \,\langle \hat{\sigma}_+ \rangle}$

We seek the spectrum after mean values reach steady state

$$\hat{\sigma}_+(t) = \langle\hat{\sigma}_+\rangle + \delta\hat{\sigma}_+(t)$$

$$\boxed{S_{coh}(\omega) = \int d\tau\, |\langle\hat{\sigma}_+\rangle|^2 e^{-i(\omega-\omega_L)\tau} = |\langle\hat{\sigma}_+\rangle|^2 \delta(\omega-\omega_L)}$$

Elastic scattering at driving frequency

$$\boxed{S_{incoh}(\omega) = \int_{-\infty}^{\infty} d\tau \langle \delta\hat{\sigma}_+(t+\tau)\,\delta\hat{\sigma}_-(t)\rangle e^{-i(\omega-\omega_L)\tau}}$$

Quantum Regression theorem

We seek two time correlation function.
Let us thus consider some collection of system operators $\{\hat{A}_i\}$ satisfy Langevin equations

$$\frac{d\hat{A}_i}{dt} = \sum_j \mathcal{L}_{ij}\hat{A}_j + \hat{\mathcal{F}}_i(t)$$

where $\mathcal{L}_{ij}$ contains both Hamiltonian and relaxation and $\hat{\mathcal{F}}_i$ are the noise operators

$$\Rightarrow \frac{d}{dt}\langle\hat{A}_i(t)\rangle = \sum_j \mathcal{L}_{ij}\langle\hat{A}_j(t)\rangle \qquad \text{since} \quad \langle\hat{\mathcal{F}}_i(t)\rangle = 0$$

Now consider $\langle\hat{A}_i(t+\tau)\hat{A}_l(t)\rangle$

$\tau > t$ $$\frac{d}{d\tau}\langle\hat{A}_i(t+\tau)\hat{A}_l(t)\rangle = \sum_j \mathcal{L}_{ij}\langle\hat{A}_j(t+\tau)\hat{A}_l(t)\rangle$$

$$+ \langle\hat{\mathcal{F}}_i(\tau)\hat{A}_l(t)\rangle$$

In the Markov approximation $\langle \hat{F}_i(\tau)\, \hat{A}_\ell(t) \rangle = 0$ $\tau > t$

$$\Rightarrow \boxed{\frac{d}{d\tau} \langle \hat{A}_i(t+\tau)\, \hat{A}_\ell(t) \rangle = \sum_j \mathcal{L}_{ij} \langle \hat{A}_j(t+\tau)\, \hat{A}_\ell(t) \rangle}$$

This is known as the "Quantum Regression Theorem".

It states that the two-time correlation functions "regress" to the same equation of motion as the first.

Stated in other terms:

$$\langle \hat{A}_i(\tau) \rangle = \mathrm{Tr} \langle \hat{A}_i\, \hat{\rho}(\tau) \rangle$$

$$\langle \hat{A}_i(t+\tau)\, \hat{A}_\ell(t) \rangle = \mathrm{Tr} \langle \hat{A}_i\, \hat{\rho}^{(\ell)}_{eff}(\tau; t) \rangle$$

where $\dfrac{d\hat{\rho}^{(\ell)}_{eff}}{d\tau} = \mathcal{L}\left[\hat{\rho}^{(\ell)}_{eff}\right]$ $\quad \hat{\rho}^{(\ell)}_{eff}(t) = \hat{A}_\ell\, \hat{\rho}(t)$

Using this we can calculate the spectrum. The calculation is tedious (see texts).

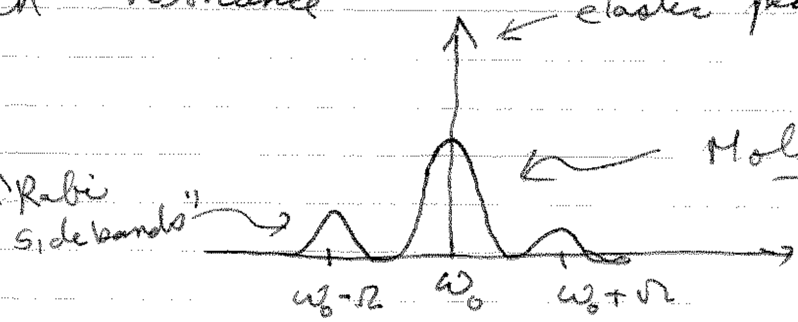
The solution for $\Delta = 0$ (resonance) $\Omega \gg \frac{\Gamma}{2}$ (high saturation)

$$S = \frac{I_0}{2\pi} \left(\frac{\pi\, \Gamma^2/4}{\frac{\Gamma^2}{4} + \frac{\Omega^2}{2}}\, \delta(\omega - \omega_L) \right\} \text{coherent}$$

Incoherent part
$$\left\{ \begin{array}{l} + \dfrac{1}{2} \dfrac{\Gamma/2}{\omega^2 + \frac{\Gamma^2}{4}} \\[2ex] + \dfrac{1}{4} \dfrac{\frac{3}{4}\Gamma}{(\omega - \Omega)^2 + \left(\frac{3\Gamma}{4}\right)^2} + \dfrac{1}{4} \dfrac{\frac{3}{4}\Gamma}{(\omega + \Omega)^2 + \left(\frac{3\Gamma}{4}\right)^2} \end{array} \right.$$

Historically, this spectrum was first calculated by P. R. Mollow (Phy Rev. 188, 1969 (1969))

On resonance



An intuitive picture can be see using the Dressed states (Cohen-Tannoudji 1976)