Lecture #17   27 Oct. 2005

Resonance Fluorescence from a driven 2-level atom

Consider a field $E(t)$ (later this will be the field radiated by the atom) and its Fourier transform

$$f(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} E(t)\, e^{-i\omega t}\, dt$$

the spectrum is

$$\mathcal{P}(\omega) = \langle |f(\omega)|^2 \rangle$$

but $E(t)$ may not be integrable on $-\infty \to +\infty$, and it may be hard to determine.

It turns out that we can define the correlation function

not the decay rate
$$\Gamma(\tau) = \lim_{T\to\infty} \frac{1}{2T}\int_{-T}^{T} E^*(t)\, E(t+\tau)\, dt$$

that is integrable

and it can be shown that

$$\mathcal{P}(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \Gamma(\tau)\, e^{-i\omega\tau}\, dt$$

(the Wiener-Khinchine theorem)   See, e.g. Loudon, Q. Th. of Light, 3rd ed., pp. 101-103

that is, the power spectrum is the F.T. of the field-field correlation function.

For a driven 2-level atom, the field is radiated by the induced, oscillating dipole moment. In steady state, the solutions of the optical Bloch equations give us the <u>average</u> value of this dipole moment. But it is the instantaneous value that radiates.

(Actually, it is $\ddot{d}(t)$ to which $E(t)$ is proportional. But the motion of $d(t)$ is mainly at frequencies close to ω, the driving frequency, so for $\delta, \Omega \ll \omega$ we can take $\ddot{d}(t) \simeq -\omega^2 d(t)$ for all frequencies of interest. That is, we can take $E(t) \propto d(t)$ to a good approximation.

To calculate the correlation function of the field we recall the steady state solution of the optical Bloch equations:

$$u = \frac{\Omega}{2} \frac{\delta}{\Gamma^2/4 + \delta^2 + \Omega^2/2}$$

$$v = \frac{\Omega}{2} \frac{\Gamma/2}{\Gamma^2/4 + \delta^2 + \Omega^2/2}$$

$$w + \tfrac{1}{2} = \sigma_{ee} = \tilde{\sigma}_{ee} = \frac{\Omega^2}{4} \frac{1}{\Gamma^2/4 + \delta^2 + \Omega^2/2}$$

Note that there is an <u>average</u> value of

$$\langle d \rangle \propto u\cos\omega t - v\sin\omega t$$ check sign

that oscillates locked to the driving frequency ω. Its correlation function has the form

$$\Gamma(\tau) \propto \cos(\omega\tau + \phi)$$ and its

Fourier transform is a δ-function

$$P(\omega')_{\text{coherent}} \propto \delta(\omega' - \omega)$$

Note also that as $\Omega \to \infty$, keeping δ, Γ finite, $\langle d \rangle \to 0$. This does <u>not</u> mean that the radiated field $\to 0$, only that the <u>coherent</u> part, locked to the driving laser frequency, goes to zero

Let's re-write u, v, w using the definition

$$s \equiv \frac{\Omega^2/2}{\delta^2 + \Gamma^2/4} = \frac{I/I_0}{1 + (2\delta/\Gamma)^2}$$

s = the saturation parameter

at $\delta = 0$ $\quad s = I/I_0$

with this definition, the steady state components of the Bloch vector are

$$u = \frac{\delta}{\Omega}\frac{s}{1+s}$$

$$v = \frac{\Gamma}{2\Omega}\frac{s}{1+s}$$

$$w + \frac{1}{2} = \sigma_{ee} = \frac{1}{2}\frac{s}{1+s}$$

as $\Omega \to \infty$ $s \to \infty$ and

$$u \to 0$$
$$v \to 0$$
$$\sigma_{ee} \to 1/2 \qquad \text{(saturation)}$$

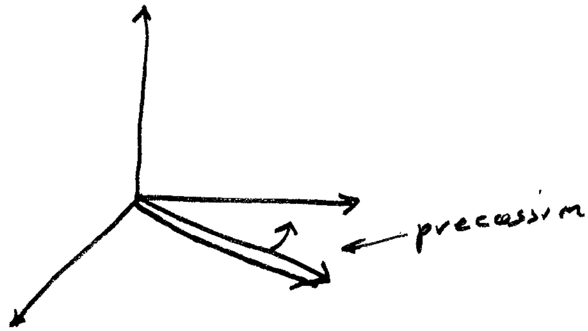
the total radiation is <u>always</u> proportional to σ_{ee}.

the total rate of radiating photons is $\Gamma\sigma_{ee}$ and has a maximum value $\Gamma/2$.

Only some of these are radiated by the <u>average</u> dipole moment oscillating in steady state at ω.

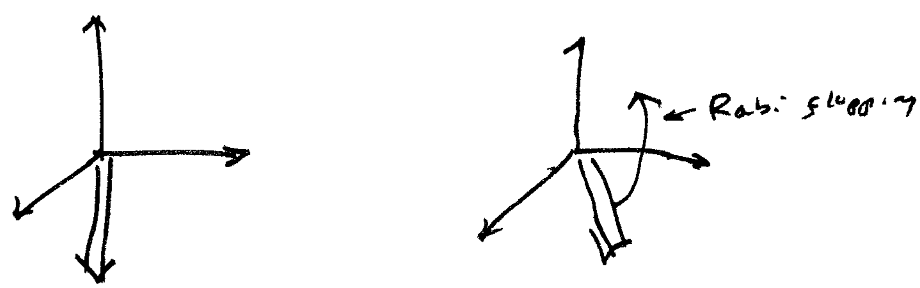
Where do the rest come from?

Consider a Bloch vector at some arbitrary time



qualitatively, let's think about the instantaneous state of the atom, not the average indicated by the density matrix treatment.

From time to time, the atom emits photons. After emission, the atom is in the ground state.



It doesn't radiate when in the ground state, but it begins Rabi flopping and radiates as it acquires excited state character. If $\Omega >> \Gamma$, it flops many times before emitting a photon, so its excited state character, and thus its emission rate is modulated at Ω. This makes the Rabi sidebands.

Each spont. emission re-sets the atom in the ground state. (This is the spirit of Quantum Monte-Carlo wavefunction trajectory calculations.)

This interrupts the emission and gives breadth to the emitted spectrum.

At the same time, the density matrix steady state solutions show that there is an average, non-zero value of the dipole, and this radiates a zero width spectrum that goes away as $\Omega \to \infty$

(Note that this is different from the radiation from a classical driven H.O. — the harmonic oscillator does not saturate, and this is a key difference.

Now let's do a quantitative calculation of the coherent and incoherent parts of the spectrum:

The radiated field, as we have seen, is proportional to the dipole moment.

the dipole moment operator is

$$\hat{d} = d_{ge}\left(|e\rangle\langle g| + |g\rangle\langle e|\right) \equiv \hat{d}_+ + \hat{d}_-$$

$\boxed{d_{ge} = d_{eg}^* \text{ real}}$

these "raising" and "lowering" parts of the dipole moment operator are related to positive and negative frequency components of the radiated field (since $E \propto d$)

define
$$S_+ = e^{-i\omega t}|e\rangle\langle g|$$
$$S_- = e^{i\omega t}|g\rangle\langle e|$$
these are the operators in the rotating frame.

ω = driving frequency

the correlation function of the electric field (whose Fourier transform is the power spectrum) is

$$\langle E^-(t+\tau)\, E^+(t)\rangle$$ where the average is over t

for a stationary process, this is independent of t - depends only on τ.

since $E = E^+ + E^-$, there are, mathematically, other terms in the correlation function $\langle E^*(t+\tau)\, E(t)\rangle$

but these other terms are small or zero

For a complete discussion, see Atom-Photon Interactions, Cohen-Tannoudji, Dupont-Roc, & Grynberg, 1992, pp. 134-136.

so the correlation function of the field is proportional to the correlation function of the dipole moment components

$$E(t) = \eta\, d(t)$$

of course, the $E(t)$ is retarded from $d(t)$, but we drop the retardation term.

since $\hat{d} \propto (|e\rangle\langle g| + |g\rangle\langle e|)$

$$\hat{d} \propto (S_+ e^{+i\omega t} + S_- e^{-i\omega t})$$

and $E^{\pm}(t) = \eta\, e^{\mp i\omega t} S_{\mp}$ because $e^{-i\omega t}$ is a "positive" frequency.

so the correlation function

$$\langle E^-(t+\tau)\, E^+(t)\rangle =$$

$$\eta^2 \langle S_+(t+\tau) e^{i\omega(t+\tau)} \cdot S_-(t) e^{-i\omega t}\rangle =$$

$$\eta^2 \langle S_+(t+\tau) e^{i\omega\tau} S_-(t)\rangle$$

the power spectrum $\mathcal{P}(\omega') = \frac{1}{2\pi}\eta^2 \int d\tau\, e^{i(\omega-\omega')\tau} \langle S_+(t+\tau) S_-(t)\rangle$

this is the formal result for the spectrum.

If we set $\gamma = 0$ then

$$\eta^2 \langle S_+(t) S_-(t) \rangle = I(t)$$, the intensity of the scattered light

but $$S_+ S_- = e^{-i\omega t} |e\rangle\langle g|g\rangle\langle e| e^{i\omega t} = |e\rangle\langle e|$$

and $$\langle |e\rangle\langle e| \rangle = \sigma_{ee}$$

so $$I_{tot} = \eta^2 \sigma_{ee}$$ — of course, because the excited state radiates.

$$I_{tot} = \eta^2 \cdot \frac{1}{2} \frac{s}{1+s}$$

now write $$S_\pm = \langle S_\pm \rangle + \delta S_\pm$$

where, by definition $\langle \delta S_\pm \rangle = 0$

$$I_{tot} = \eta^2 \langle S_+(t) S_-(t) \rangle = \eta^2 \Big(\underbrace{\langle S_+(t) \rangle \langle S_-(t) \rangle}_{1} + \underbrace{\langle \delta S_+ \delta S_- \rangle}_{2} \Big)$$

(the usual argument that the mean square fluctuation is the mean square minus the square mean)

Term 1 is the radiation of the mean value of the dipole moment (static in the rotating frame, because it is phase-coherent with the driving field)

this is the "coherent," monochromatic part of the spectrum.

$$I_{coh} = \eta^2 \langle s_+ \rangle \langle s_- \rangle = \eta^2 |\langle s_+ \rangle|^2$$

since S_+ & S_- are hermitian conjugates.

$$|\langle S_+ \rangle|^2 = |u_{st} + i v_{st}|^2$$

, the steady state value of α^2

recall $u_{st} = \frac{\delta}{\Omega} \frac{s}{1+s}$

$$v_{st} = \frac{\Gamma}{2\Omega} \frac{s}{1+s}$$

$$u_{st}^2 + v_{st}^2 = \frac{s^2}{(1+s)^2} \cdot \frac{\frac{1}{2}}{s} = \frac{1}{2} \frac{s}{(1+s)^2}$$

so $I_{coh} = \eta^2 \cdot \frac{1}{2} \frac{s}{(1+s)^2}$

but $I_{tot} = \eta^2 \frac{1}{2} \frac{s}{1+s}$

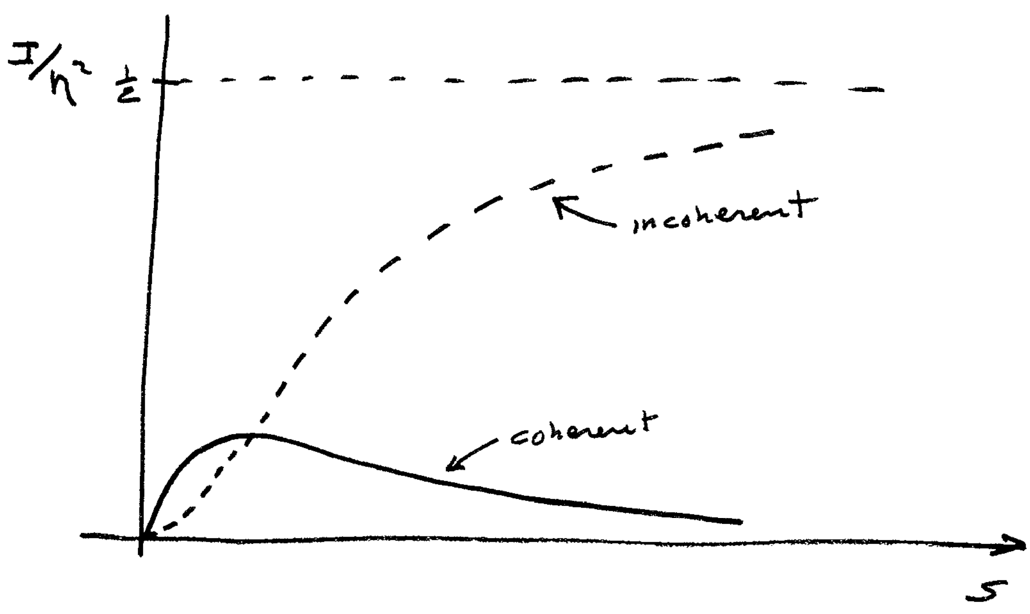
$$I_{incoh} = I_{tot} - I_{coh}$$

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

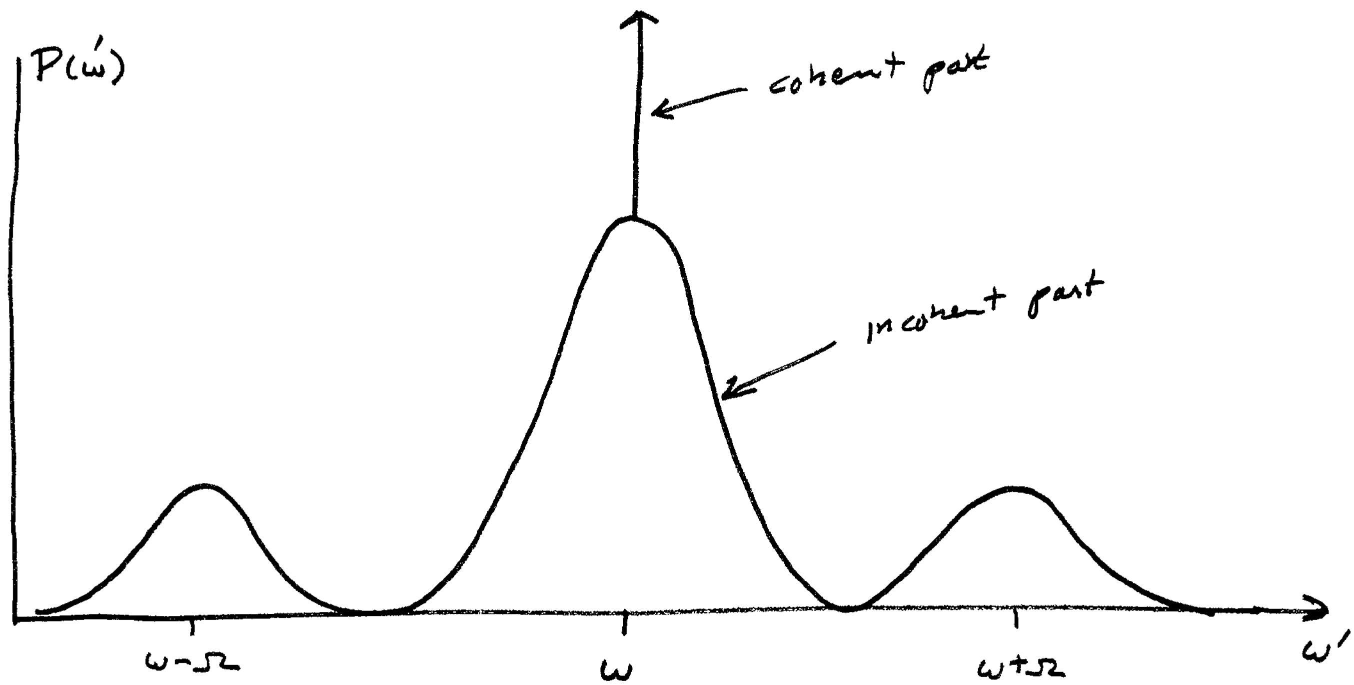
$$= \frac{n^2}{2}\left(\frac{s^2+s \quad - s}{(1+s)^2}\right)$$

$$I_{incoh} = n^2 \cdot \frac{1}{2}\frac{s^2}{(1+s)^2}$$

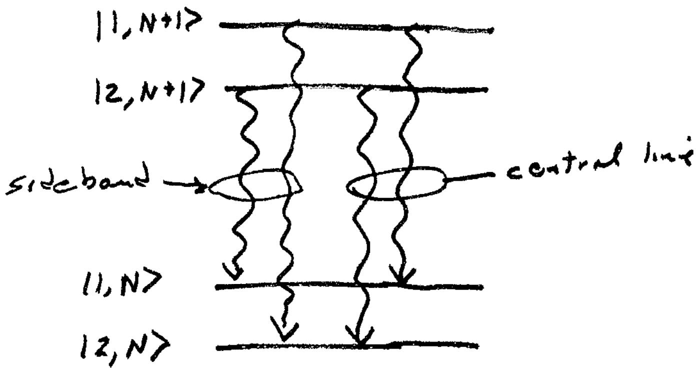
$$I_{coh} = n^2 \quad \frac{1}{2}\frac{s}{(1+s)^2}$$



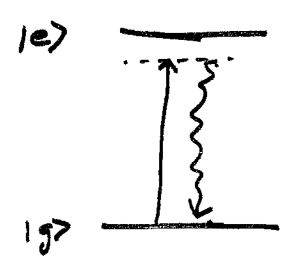
the spectrum. (which we did not explicitly calculate)



for $\quad \Omega \gg \Gamma \qquad \omega = \omega_0$

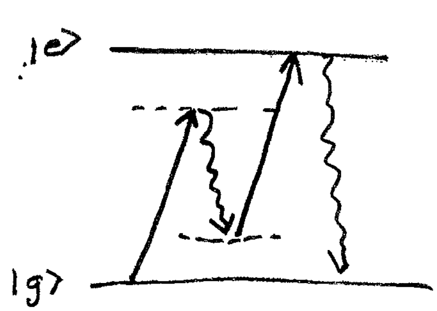


low intensity:



energy conservation $\Rightarrow$
$\omega' = \omega$

high intensity:



energy is conserved overall, but not photon by photon.