Physics 4261: Lectures for week 9

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9.1 Large excited state fractions

Now we will consider the situation when the excited state is not necessarily a small amount of the total. Again, we work in a rotating-wave approximation (RPA). The equations are

$$\begin{aligned} i\dot{c_1} &= \frac{\Omega}{2} c_2 e^{i\delta t}, \\ i\dot{c_2} &= \frac{\Omega^*}{2} c_1 e^{-i\delta t}, \end{aligned}$$

where $\delta = \omega - \omega_0$. At this point we can make a clever substitution and define $\widetilde{c_1} = c_1 e^{-i\delta t}$ and $\widetilde{c_2} = c_2 e^{i\delta t}$. Then the equations become

$$\begin{pmatrix} \dot{\widetilde{c_1}} \\ \dot{\widetilde{c_2}} \end{pmatrix} = -\frac{i}{2} \begin{pmatrix} \delta & \Omega \\ \Omega^* & -\delta \end{pmatrix} \begin{pmatrix} \widetilde{c_1} \\ \widetilde{c_2} \end{pmatrix},$$

which is easily solved. Let us write things in terms of the density matrix, as that is where we are ultimately going.

$$\rho = \begin{pmatrix} |\widetilde{c_1}|^2 & \widetilde{c_1}\widetilde{c_2}^* e^{i\omega t} \\ \widetilde{c_1}^* \widetilde{c_2} e^{-i\omega t} & |\widetilde{c_2}|^2 \end{pmatrix}.$$

Now, I can define my basis states in terms of $\widetilde{c_1}$ and $\widetilde{c_2}$, in which case the density matrix loses the phase factor

$$\widetilde{\rho} = \begin{pmatrix} |\widetilde{c_1}|^2 & \widetilde{c_1}\widetilde{c_2}^* \\ \widetilde{c_1}^* \widetilde{c_2} & |\widetilde{c_2}|^2 \end{pmatrix}.$$

Since this phase does not affect the dissipative part of the density matrix evolution, I will use this convention. Let me also assume Ω is real from here. From this I deduce that

$$\begin{aligned} \dot{u} + i\dot{v} &= 2\left(\dot{\widetilde{c_1}}\widetilde{c_2}^* + \widetilde{c_1}\dot{\widetilde{c_2}}^*\right) \\ &= -i\delta\widetilde{c_1}\widetilde{c_2}^* - i\Omega\widetilde{c_2}\widetilde{c_2}^* - i\delta\widetilde{c_1}\widetilde{c_2}^* + i\Omega\widetilde{c_1}\widetilde{c_1}^*, \\ &= \delta v - i\delta u + i\Omega w, \end{aligned}$$

and

$$\begin{aligned}\dot{w} &= 2\left(\dot{\widetilde{c}}_1\widetilde{c}_1^{\,*} + \widetilde{c}_1\dot{\widetilde{c}}_1^{\,*}\right)\\ &= -i\delta\widetilde{c}_1\widetilde{c}_1^{\,*} - i\Omega\widetilde{c}_2\widetilde{c}_1^{\,*} + i\delta\widetilde{c}_1\widetilde{c}_1^{\,*} + i\Omega\widetilde{c}_1\widetilde{c}_2^{\,*},\\ &= -\Omega v,\end{aligned}$$

Finally, putting everything together and including dissipative terms gives the OBE,

$$\begin{aligned}\dot{u} &= \delta v - \frac{\Gamma}{2}u,\\ \dot{v} &= -\delta u + \Omega w - \frac{\Gamma}{2}v,\\ \dot{w} &= -\Omega v - \Gamma(w-1).\end{aligned}$$

9.2 Fixed points of the OBE

Recall the OBE:

$$\begin{aligned}\dot{u} &= \delta v - \frac{\Gamma/2}{u},\\ \dot{v} &= -\delta u + \Omega w - \frac{\Gamma}{2}v,\\ \dot{w} &= -\Omega v - \Gamma(w-1).\end{aligned}$$

We want to set this system of equations equal to zero. This is a matrix equation:

$$\begin{pmatrix}-\Gamma/2 & \delta & 0\\ -\delta & -\Gamma/2 & \Omega\\ 0 & -\Omega & -\Gamma\end{pmatrix}\begin{pmatrix}u\\ v\\ w\end{pmatrix} = \begin{pmatrix}0\\ 0\\ -\Gamma\end{pmatrix}.$$

On your homework, you will find the solutions:

$$\begin{pmatrix}u\\ v\\ w\end{pmatrix} = \frac{1}{\delta^2 + \Omega^2/2 + \Gamma^2/4}\begin{pmatrix}\Omega\delta\\ \Omega\Gamma/2\\ \delta^2 + \Gamma^2/4\end{pmatrix}.$$

Let's have some fun with these. First we are going to calculate the scattering rate, which is just A_{21} times the population of the excited state $1/2 - w/2$. A note about A_{21} and Γ here: for the two-level system, they will be equal, but in anticipation multi-level systems, we will allow them to be different. The scattering rate is then

$$R_{\text{scat}} = \frac{(\Gamma/2)\Omega^2/2}{\delta^2 + \Omega^2/2 + \Gamma^2/4}.$$

Next we look for the optical absorption cross section. By this, we mean the scattering rate as a function of photon flux per unit area, or $I/\hbar\omega$, where $I = \frac{\epsilon_0 E^2}{2}$. This will be

$$\sigma(\omega) = \frac{\Gamma\hbar\omega}{2I}\frac{\Omega^2/2}{\delta^2 + \Omega^2/2 + \Gamma^2/4}.$$

Now, the big issue is to detangle the intensity of light I from Ω^2, because both depend on electric field squared. Here are some relations (taking $A_{21} = \Gamma$)

$$\begin{aligned}
A_{21} &= \frac{4\alpha\omega_0^3}{3c^2}|X_{12}|^2, \\
X_{12} &= \frac{3\pi\epsilon_0 c^3\hbar}{e^2\omega_0^3}\Gamma, \\
\Omega^2 &= \frac{e^2E^2|X_{12}|^2}{\hbar^2} = \frac{3\pi\epsilon_0 c^3}{\hbar\omega_0^3}\Gamma, \\
I &= \frac{\epsilon_0 c}{2}E^2, \\
E^2 &= \frac{2I}{\epsilon_0 c}, \\
\Omega^2 &= \frac{6\pi c^2}{\hbar\omega_0^3}\Gamma I.
\end{aligned}$$

Putting all of this together (and taking $\omega \approx \omega_0$) gives me

$$\begin{aligned}
\sigma(\omega) &= \frac{6\pi}{c^2\omega_0^2}\frac{\Gamma^2/4}{\delta^2 + \Omega^2/2 + \Gamma^2/4}, \\
\sigma(\omega) &= \frac{3\lambda^2}{2\pi}\frac{1}{1 + 4\frac{\delta^2}{\Gamma^2} + \frac{2\Omega^2}{\Gamma^2}}
\end{aligned}$$

We notice that the cross section still depends on the intensity, in the term

$$\begin{aligned}
\frac{2\Omega^2}{\Gamma^2} &= \frac{12\pi c^2}{\hbar\omega_0^3\Gamma}I = \frac{I}{I_{\text{sat}}}, \\
I_{\text{sat}} &= \frac{\hbar\omega_0^3\Gamma}{12\pi c^2} = \frac{\pi}{3}\frac{hc\Gamma}{\lambda^3} = \frac{\pi}{3}\frac{hc}{\lambda^3\tau},
\end{aligned}$$

where I_{sat} is called the saturation intensity, because above this value the cross section begins to go down, and the transition is said to be saturated. Then the cross section is

$$\begin{aligned}
\sigma(\omega) &= \frac{3\lambda^2}{2\pi}\frac{1}{1 + 4\frac{\delta^2}{\Gamma^2} + \frac{I}{I_{\text{sat}}}}, \\
\sigma(\omega) &\propto \frac{1}{1 + 4\frac{\delta^2}{\Delta\omega^2}}, \\
\Delta\omega^2 &= \Gamma^2\left(1 + \frac{I}{I_{\text{sat}}}\right).
\end{aligned}$$

The tendency of $\Delta\omega$ (the FWHM) to increase with power is called "power broadening". Finally, we can reexpress the scattering rate as

$$R_{\text{scat}} = \frac{I}{I + I_{\text{sat}} + I_{\text{sat}}4\frac{\delta^2}{\Gamma^2}}\frac{\Gamma}{2}.$$

So sometimes we define a frequency-dependent saturation intensity

$$I_s(\omega) = I_{\text{sat}}\left(1 + 4\frac{\delta^2}{\Gamma^2}\right).$$

9.2.1 Light shift

Finally, we consider the evolution of the system with no dissipation, and compute a quantity we call the "light shift". This is how much the addition of the radiation field causes the atomic levels to shift. Classically, it is analogous to the refractive index of the atoms. Recall that the addition of the field to the equations lead things to oscillate at the frequency $W/2 = \sqrt{(\delta/2)^2 + (\Omega/2)^2}$. Now, if $\Omega \to 0$ this oscillates at $\delta/2$ as expected from being off resonance. Therefore the difference

$$\frac{W-\delta}{2} = \frac{1}{2}\left(\delta^2 - \sqrt{\delta^2+\Omega^2}\right) \approx \frac{\Omega^2}{4\delta} = \frac{\Gamma^2}{8\delta}\frac{I}{I_{\text{sat}}},$$

represents the amount that the "dressed atoms" have different energy levels from the atoms ordinarily.

9.3 Spectroscopy

In this section we are going to cover the basics of actual physically realized spectrscopy, having a good understanding of single atoms in radiation fields under our belts. So far, our atomic theory has told us that intrinsic properties of the atoms determine the center wavelength and the width of our transitions. The main question that we want to ask here is: what effects can lead to broadening or shifts in experiments. We begin with the Doppler effect

9.4 Doppler broadening

Doppler broadening is typically the most significant source of broadening, but fortunately it can be overcome. From your homework, you derived that the Doppler effect leads to an additional broadening

$$g_D(\omega) = \frac{c}{u\omega_0\sqrt{\pi}} \exp\left\{-\frac{c^2}{u^2}\frac{\delta^2}{\omega_0^2}\right\},$$

where $u = \sqrt{2k_BT/M}$.

9.4.1 The crossed-beam method

Here we summarize what happens in the crossed-beam method. The atoms leave the oven with a thermal velocity distribution, but then only certain angles are allowed into the chamber. Thus the transverse velocity is reduced by the spread angle α (or $\sin\alpha$) from the expected value of the thermal velocity (plus some prefactors, which we will ignore). By putting the beam in the direction perpendicular to the atomic beam, the transverse velocity will be small, and therefore the Doppler broadening reduced. An consideration here is the transit-time broadening, although in these experiments this is typically small. This is the frequency time uncertainty relation $\Delta f_{\text{tt}} = 1/T = v/d$.

9.4.2 Saturated absorption spectroscopy (Foot 8.3)

I would call this the workhorse of modern spectroscopy. Note that here we will use the notation $N(v)$ to denoted the density of atoms with velocity v (relative to the beam). Our starting point is

the number of atoms in the ground state (i.e. $|c_1|^2 = (1+w)/2$), which is given by

$$N_1(v) - N_2(v) = Nf(v) \times \frac{1}{1 + (I/I_{\text{sat}}) \frac{x^2}{x^2 + \Gamma/4}},$$

where x is the Doppler-included detuning $x = \omega - \omega_0 + kv$. Let us sketch out what $N(v)$ looks like with no intensity. But here is where things get tricky: when the intensity is significant compared to the saturation value, the atoms which are on-resonant with the laser beam will exist in the upper state with significant probability and absorption will be saturated. Now, let me take another beam (weak beam, probe beam), and run it in the opposite direction. Note the Doppler shift of this beam is in the opposite direction! Now, if the beam is at true resonance, then both beams will be resonant with stopped atoms, and there will be low absorption of the probe, as the transition is saturated by the pump. That being said however, if the laser if within the Doppler profile, but not exactly on resonance, the pump and probe will see different velocity groups, and hence the probe will be absorbed at the normal amount. So the net effect is to carve a hole in the distribution. On your homework you will work out how wide this "hole" is.

When there are multiple excited states (for example, multiple hyperfine levels) another phenomenon can happen when the frequency is exactly halfway between two transitions. In this case, one transition from the pump beam will be resonant with a velocity class which is resonant with the *other* transition for the probe beam. This is called a "crossover".

Note that a few more things can happen if there are multiple hyperfine or fine structure levels in the ground state. In this case, in addition to saturating transitions by populating the upper level of the optical transition, the population can also be "depumped" into another ground state level which is off-resonant, and this is similarly reduces the absorption on resonance. However, if two transitions involving different ground states are on resonance (this requires large Doppler width and small hyperfine splitting in the ground state), then the population can be "repumped" into the original state, reducing the natural depumping effect and *increasing* the absorption strength.