3.7 Saturation parameters (T)

Consider an ensemble of atoms that are illuminated by a light field. Suppose we want to measure some property of the atoms with the light – for example, we are interested in determining the strength of a particular transition. In this situation, we need to be careful that the light field itself does not perturb the property of the atoms we are trying to measure. On the other hand, perhaps we are interested in observing some nonlinear optical process or maybe we want to optically pump all of the atoms into a particular Zeeman sublevel. In these cases, it is necessary that the light field strongly perturb the atomic system.

The crucial parameter that characterizes what regime we are in – whether or not the light field strongly perturbs the populations of the atomic states – is called the *saturation parameter* κ. The general form of the saturation parameter is

$$\kappa = \frac{\text{excitation rate}}{\text{relaxation rate}} . \tag{3.156}$$

The tricky part is that the exact form of κ and the behavior of the system as a function of κ depend on the specific system under consideration – the atomic level structure, the relaxation mechanisms, etc. In this problem, we consider a variety of systems in order to gain familiarity with calculating saturation parameters and understanding their implications.

In the following cases (a) and (b), assume that the light is tuned to resonance and that the optical depth is small, i.e.

$$n\sigma_{\text{abs}}\ell \ll 1 , \tag{3.157}$$

where ℓ is the length of the atomic sample, n is the atomic number density, and σ_{abs} is the appropriate absorption cross-section (see Problems 3.5 and 3.6). The quantity $\ell_0 = (n\sigma_{\text{abs}})^{-1}$ is commonly referred to as the absorption length. The condition (3.157) ensures that the intensity of the light field does not significantly change as the light propagates through the sample and, as long as all dimensions of the atomic sample are similarly small, that high atomic density effects such as *radiation trapping*[8] are not important. Additionally we assume that the average spacing between the atoms $n^{-1/3}$ is considerably larger than the wavelength of the light λ. This allows us to ignore effects that involve cooperative behavior of the atoms [such as Dicke superradiance (Dicke 1954), see Problem 3.14].

(a) Consider two-level stationary atoms for which the only source of line broadening is the spontaneous decay of the upper state $|e\rangle$ back to the lower state $|g\rangle$ (Fig. 3.5). Calculate the saturation parameter κ for the $|g\rangle \rightarrow |e\rangle$ transition

[8] If the atomic density is sufficiently high, there can be a significant probability that spontaneously emitted photons are re-absorbed. Thus the photons must diffuse out of the atomic sample, which affects, for example, measurements of excited state lifetimes. See, for example, Corney (1988).

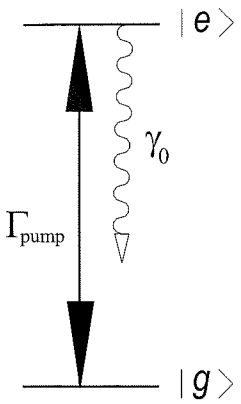

FIG. 3.5 Level diagram for the two-level system considered in part (a).

for narrow-band (monochromatic) incident light, and find the dependence of the fluorescence intensity on κ.

Solution

The excitation rate Γ_{pump} (we can think of the light effectively "pumping" the atoms into the excited state) is given by Eq. (3.132) from Problem 3.4:

$$\Gamma_{\text{pump}} = \frac{d^2 \mathcal{E}_0^2}{\gamma_0} , \tag{3.158}$$

where d is the dipole matrix element $\langle e|d|g\rangle$ between the states, $\mathcal{E}_0$ is the amplitude of the light electric field, γ_0 is the spontaneous decay rate of $|e\rangle$ to $|g\rangle$, and we have set $\hbar = 1$. The relaxation rate in this problem is γ_0, so from (3.156) we have

$$\boxed{\kappa = \frac{\Gamma_{\text{pump}}}{\gamma_0} = \frac{d^2 \mathcal{E}_0^2}{\gamma_0^2} .} \tag{3.159}$$

The fluorescence intensity I_F is proportional to the number of atoms in the excited state N_e multiplied by the spontaneous decay rate γ_0. To find the population of the upper state we can write rate equations for the number of atoms in the excited state N_e and the number of atoms in the ground state N_g:

$$\frac{dN_g}{dt} = -\Gamma_{\text{pump}} N_g + (\gamma_0 + \Gamma_{\text{pump}}) N_e , \tag{3.160}$$

$$\frac{dN_e}{dt} = +\Gamma_{\text{pump}} N_g - (\gamma_0 + \Gamma_{\text{pump}}) N_e . \tag{3.161}$$

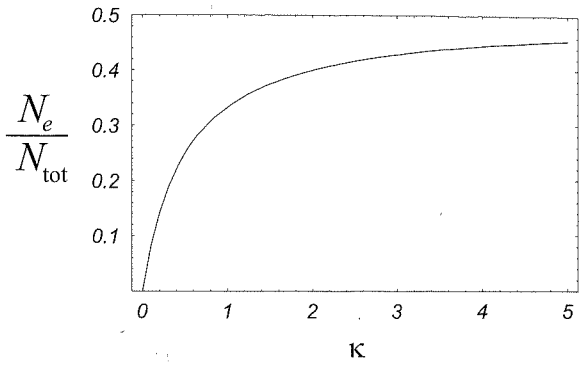

FIG. 3.6 Fractional population of excited state as a function of the saturation parameter κ for the case described in part (a). The fluorescence intensity I_F is proportional to $\gamma_0 N_e$.

We also know that $N_e + N_g = N_{\text{tot}}$ where N_{tot} is the total number of atoms in the sample. We have included the pumping rate for both the $|g\rangle \rightarrow |e\rangle$ transition and the $|e\rangle \rightarrow |g\rangle$ transition because at sufficiently high light powers ($\kappa \gtrsim 1$), stimulated emission from the upper state becomes important compared to spontaneous emission. It is clear that the stimulated emission and absorption rates should be the same from time-reversal symmetry [this can also be seen from Einstein's famous argument involving an atomic gas in thermal equilibrium with a photon gas, which was used to derive the A and B coefficients; see, for example, Griffiths (1995) or Bransden and Joachain (1989)]. In equilibrium, dN_g/dt and dN_e/dt are zero, and we find that

$$N_e = \frac{\kappa}{1 + 2\kappa} N_{\text{tot}} , \tag{3.162}$$

so the fluorescence intensity is proportional to $\kappa/(1 + 2\kappa)$ (Fig. 3.6).

(b) Now suppose we have a three-level system as shown in Fig. 3.7. The incident light is resonant with the $|g\rangle \rightarrow |e\rangle$ transition and the excited state $|e\rangle$ primarily decays to a metastable level $|m\rangle$ at a rate γ_0. There is a slow relaxation rate $\gamma_{\text{rel}} \ll \gamma_0$ of the metastable level back to the ground state. The states $|m\rangle$ and $|g\rangle$ could be, for example, different ground state hyperfine levels, and γ_{rel} could be the result of collisional relaxation. Again assume that Doppler broadening may be ignored and that the excitation light is monochromatic.

Calculate the saturation parameter κ for this situation, and find the dependence of the fluorescence intensity on κ.

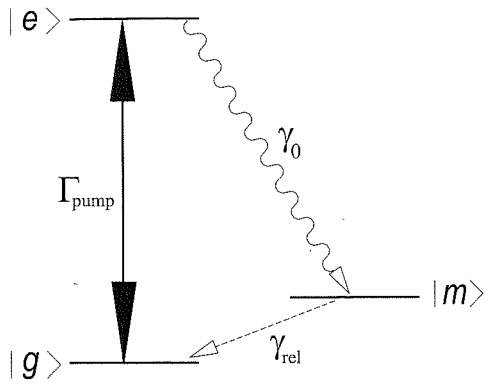

FIG. 3.7 Level diagram for the three-level system considered in part (b).

Solution

The relaxation rate referred to in Eq. (3.156) is generally the slowest relaxation rate in the system, since this process becomes a "bottleneck" for the incoherent return of atoms to the ground state. Therefore in this case the saturation parameter is given by

$$\boxed{\kappa = \frac{d^2\mathcal{E}^2}{\gamma_0\gamma_{\text{rel}}}}\ , \tag{3.163}$$

since γ_{rel} is the slowest rate in the problem.

To verify Eq. (3.163) and find the dependence of the spontaneous emission intensity on κ we again write down the appropriate rate equations as we did in part (a):

$$\frac{dN_g}{dt} = -\Gamma_{\text{pump}}N_g + \gamma_{\text{rel}}N_m\ , \tag{3.164}$$

$$\frac{dN_e}{dt} = +\Gamma_{\text{pump}}N_g - \gamma_0 N_e\ , \tag{3.165}$$

$$\frac{dN_m}{dt} = +\gamma_0 N_e - \gamma_{\text{rel}}N_m\ , \tag{3.166}$$

where we have neglected stimulated emission (since the transition saturates long before stimulated emission becomes important). We also have the condition $N_{\text{tot}} = N_g + N_e + N_m$. Setting the time derivatives of the populations equal to zero to obtain the steady state result, after some algebra (and making use of the fact that $\gamma_{\text{rel}} \ll \gamma_0$) we find for the excited state population (Fig. 3.8)

$$N_e = \frac{\kappa}{1+\kappa}\frac{\gamma_{\text{rel}}}{\gamma_0}N_{\text{tot}}\ . \tag{3.167}$$

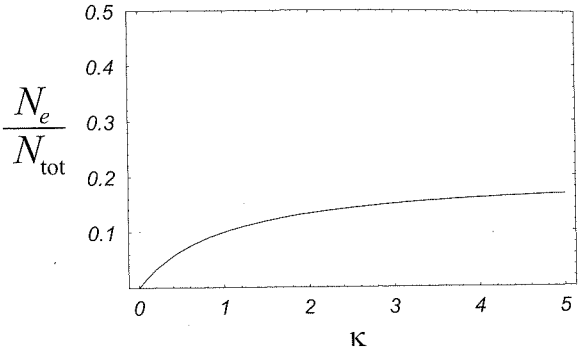

FIG. 3.8 Fractional population of excited state as a function of the saturation parameter κ for the case described in part (b). For the plot we have chosen $\gamma_{\text{rel}}/\gamma_0 = 0.2$.

Note that the maximum population in the upper state (obtained for $\kappa \gg 1$) is

$$N_e(\max) = \frac{\gamma_{\text{rel}}}{\gamma_0} N_{\text{tot}} \,. \tag{3.168}$$

Again the fluorescence intensity is proportional to $\gamma_0 N_e$, so the maximum fluorescence intensity is smaller than in the two-level case by a factor of $2\gamma_{\text{rel}}/\gamma_0$, since atoms tend to reside in the "bottleneck" state $|m\rangle$.

(c) Now we discuss the phenomenon of *power broadening*. Consider the atomic system discussed in part (b) of this problem (Fig. 3.7).

If one scans the frequency of a laser through the atomic resonance at low light powers [$\kappa \ll 1$, where κ is given by expression (3.163)], one finds that the fluorescence intensity measured as a function of detuning has a Lorentzian lineshape with width γ_0.

What is the dependence of the fluorescence intensity $I_F(\Delta)$ on detuning for large κ?

Solution

As the excitation light is tuned through resonance with the $|g\rangle \rightarrow |e\rangle$ transition, the pumping rate Γ_{pump} follows a Lorentzian dependence,[9] so we have an effective saturation parameter $\kappa_{\text{eff}}(\Delta)$ that depends on the detuning Δ of the light from resonance:

$$\kappa_{\text{eff}}(\Delta) = \kappa \frac{\gamma_0^2/4}{\Delta^2 + \gamma_0^2/4} \,, \tag{3.169}$$

[9] This can be seen by calculating the stimulated absorption rate as done in Problem 3.4 without assuming the excitation light is on resonance, but rather using the Lorentzian profile from Eq. (3.79).

where κ is the resonant saturation parameter [Eq. (3.163)] and the Lorentzian is normalized to unity on resonance. The effective saturation parameter $\kappa_{\text{eff}}(\Delta)$ can be used directly in the rate equations in place of κ, so we obtain from Eq. (3.167) the fluorescence intensity $I_F(\Delta) \propto \gamma_0 N_e$ as a function of detuning:

$$I_F(\Delta) \propto \frac{\kappa_{\text{eff}}(\Delta)}{1+\kappa_{\text{eff}}(\Delta)} \gamma_{\text{rel}} N_{\text{tot}} \tag{3.170}$$

$$= \kappa \frac{\gamma_0^2/4}{\Delta^2+\gamma_0^2/4} \frac{1}{1+\kappa\left(\frac{\gamma_0^2/4}{\Delta^2+\gamma_0^2/4}\right)} \gamma_{\text{rel}} N_{\text{tot}} \tag{3.171}$$

$$= \frac{\gamma_0^2/4}{\Delta^2+(1+\kappa)\gamma_0^2/4} \kappa \gamma_{\text{rel}} N_{\text{tot}} \,. \tag{3.172}$$

This is just a Lorentzian profile with a width

$$\boxed{\gamma = \gamma_0\sqrt{1+\kappa}} \tag{3.173}$$

known as the *power-broadened linewidth*.

(d) Finally, we consider how Doppler broadening affects our results. If the atoms in a sample have a thermal distribution of velocities, from the viewpoint of a moving atom the light frequency is shifted by an amount $\approx \vec{k}\cdot\vec{v}$, where $\vec{k}$ is the wavevector of the light and $\vec{v}$ is the atomic velocity. Averaging over all atomic velocities, as mentioned in Problem 3.6, we have for $I_F(\Delta)$ in the limit of large Doppler width $\Gamma_D \gg \gamma_0$:[10]

$$I_F(\Delta) = I_F(0) e^{-\Delta^2/\Gamma_D^2} \,. \tag{3.174}$$

In contrast to the previously discussed *homogeneous* broadening mechanisms such as spontaneous emission and power broadening, Doppler broadening is an example of *inhomogeneous* broadening – the probability for emission and absorption is not the same for all atoms.

Again consider atoms with the energy level structure shown in Fig. 3.7, but now assume that the atoms have a thermal distribution of velocities. If we tune the narrow-band excitation light to a particular frequency within the Doppler profile, the light primarily interacts with a group of atoms whose velocities are such that the Doppler shifts are less than the homogeneous linewidth. Such a set of atoms is commonly referred to as a *velocity group*, illustrated in Fig. 3.9.

What is the dependence of fluorescence intensity on κ for such a Doppler-broadened medium?

[10] A more accurate representation of the spectral profile, which takes into account both homogeneous and inhomogeneous broadening mechanisms is the *Voigt profile*, which is a convolution of Lorentzian and Gaussian profiles [see, for example, Demtröder (1996) and Khriplovich (1991)].

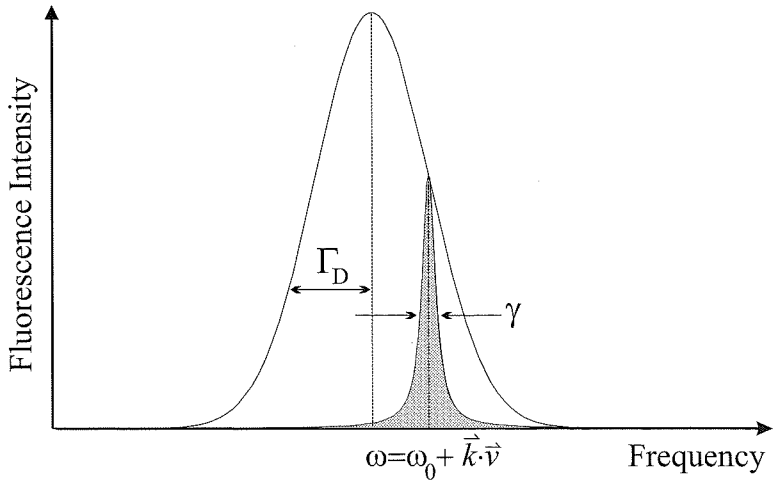

FIG. 3.9 When narrow-band excitation light is tuned to frequency ω within a Doppler-broadened profile, the fluorescence is due to a particular group of atoms with velocities $\vec{v}$ whose Doppler shifts are $\lesssim \gamma$.

Solution

The fraction δN of the total number of atoms N_{tot} with which the light interacts is

$$\delta N \sim \frac{\gamma}{\Gamma_D} N_{\text{tot}} , \tag{3.175}$$

where γ is the homogeneous linewidth. For the considered case, γ is the power-broadened linewidth given by Eq. (3.173). Otherwise, the rate equations for the resonant velocity group remain the same as those considered in part (b), and we have:

$$\boxed{I_F \propto \frac{\kappa}{1+\kappa}\delta N \propto \frac{\kappa}{\sqrt{1+\kappa}}\frac{\gamma_0}{\Gamma_D} N_{\text{tot}} .} \tag{3.176}$$

Note that in contrast to the Doppler-free case, the fluorescence intensity continues to increase ($\propto \sqrt{\kappa}$) even for $\kappa \gg 1$. This continues as long as $\gamma_0\sqrt{1+\kappa} \ll \Gamma_D$. In the opposite limit, $\gamma_0\sqrt{1+\kappa} \gg \Gamma_D$, Doppler broadening may be ignored.