

Chapter 6

Interaction of Light and Matter

Atomic or molecular gases in low concentration show sharp energy eigen-spectra. This was shown for the hydrogen atom. Usually, there are infinitely many energy eigenstates in an atomic, molecular or solid-state medium and the spectral lines are associated with allowed transitions between two of these energy eigenstates. For many physical considerations it is already sufficient to take only two of these possible energy eigenstates into account, for example those which are related to the laser transition. The pumping of the laser can be later described by phenomenological relaxation processes into the upper laser level and out of the lower laser level. The resulting simple model is often called a two-level atom, which is mathematically also equivalent to a spin 1/2 particle in an external magnetic field, because the spin can only be parallel or anti-parallel to the field, i.e. it has two energy levels and energy eigenstates [4]. The interaction of the two-level atom with the electric field of an electromagnetic wave is described by the Bloch equations.

6.1 The Two-Level Model

An atom with only two energy eigenvalues is described by a two-dimensional state space spanned by the two energy eigenstates $|e\rangle$ and $|g\rangle$. The two states constitute a complete orthonormal system. The corresponding energy eigenvalues are E_e and E_g , see Fig. 6.1. In the position-, i.e. x -representation, these states correspond to the wave functions

$$\psi_g(x) = \langle x | g \rangle, \quad \text{and} \quad \psi_e(x) = \langle x | e \rangle. \quad (6.1)$$

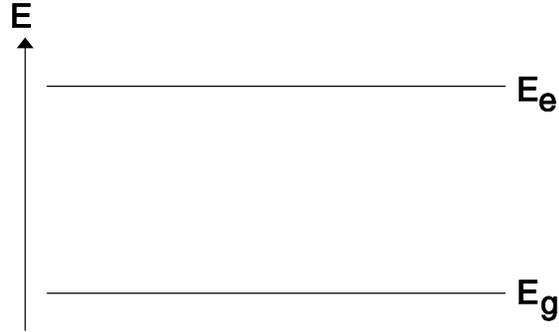


Figure 6.1: Two-level atom

The Hamiltonian operator of the two-level atom is in the energy representation

$$\mathbf{H}_A = E_e |e\rangle \langle e| + E_g |g\rangle \langle g|. \quad (6.2)$$

In this two-dimensional state space only $2 \times 2 = 4$ linearly independent linear operators are possible. A possible choice for an operator base in this space is

$$\mathbf{1} = |e\rangle \langle e| + |g\rangle \langle g|, \quad (6.3)$$

$$\boldsymbol{\sigma}_z = |e\rangle \langle e| - |g\rangle \langle g|, \quad (6.4)$$

$$\boldsymbol{\sigma}^+ = |e\rangle \langle g|, \quad (6.5)$$

$$\boldsymbol{\sigma}^- = |g\rangle \langle e|. \quad (6.6)$$

The non-Hermitian operators $\boldsymbol{\sigma}^\pm$ could be replaced by the Hermitian operators $\boldsymbol{\sigma}_{x,y}$

$$\boldsymbol{\sigma}_x = \boldsymbol{\sigma}^+ + \boldsymbol{\sigma}^-, \quad (6.7)$$

$$\boldsymbol{\sigma}_y = -j\boldsymbol{\sigma}^+ + j\boldsymbol{\sigma}^-. \quad (6.8)$$

The physical meaning of these operators becomes obvious, if we look at the action when applied to an arbitrary state

$$|\psi\rangle = c_g |g\rangle + c_e |e\rangle. \quad (6.9)$$

We obtain

$$\boldsymbol{\sigma}^+ |\psi\rangle = c_g |e\rangle, \quad (6.10)$$

$$\boldsymbol{\sigma}^- |\psi\rangle = c_e |g\rangle, \quad (6.11)$$

$$\boldsymbol{\sigma}_z |\psi\rangle = c_e |e\rangle - c_g |g\rangle. \quad (6.12)$$

The operator σ^+ generates a transition from the ground to the excited state, and σ^- does the opposite. In contrast to σ^+ and σ^- , σ_z is a Hermitian operator, and its expectation value is an observable physical quantity with expectation value

$$\langle \psi | \sigma_z | \psi \rangle = |c_e|^2 - |c_g|^2 = w, \quad (6.13)$$

the inversion w of the atom, since $|c_e|^2$ and $|c_g|^2$ are the probabilities for finding the atom in state $|e\rangle$ or $|g\rangle$ upon a corresponding measurement. If we consider an ensemble of N atoms the total inversion would be $W = N \langle \psi | \sigma_z | \psi \rangle$. If we separate from the Hamiltonian (6.1) the term $(E_e + E_g)/2 \cdot \mathbf{1}$, where $\mathbf{1}$ denotes the unity matrix, we rescale the energy values correspondingly and obtain for the Hamiltonian of the two-level system

$$\mathbf{H}_A = \frac{1}{2} \hbar \omega_{eg} \sigma_z, \quad (6.14)$$

with the transition frequency

$$\omega_{eg} = \frac{1}{\hbar} (E_e - E_g). \quad (6.15)$$

This form of the Hamiltonian is favorable. There are the following commutator relations between operators (6.4) to (6.6)

$$[\sigma^+, \sigma^-] = \sigma_z, \quad (6.16)$$

$$[\sigma^+, \sigma_z] = -2\sigma^+, \quad (6.17)$$

$$[\sigma^-, \sigma_z] = 2\sigma^-, \quad (6.18)$$

and anti-commutator relations, respectively

$$[\sigma^+, \sigma^-]_+ = \mathbf{1}, \quad (6.19)$$

$$[\sigma^+, \sigma_z]_+ = \mathbf{0}, \quad (6.20)$$

$$[\sigma^-, \sigma_z]_+ = \mathbf{0}, \quad (6.21)$$

$$[\sigma^-, \sigma^-]_+ = [\sigma^+, \sigma^+]_+ = \mathbf{0}. \quad (6.22)$$

The operators σ_x , σ_y , σ_z fulfill the angular momentum commutator relations

$$[\sigma_x, \sigma_y] = 2j\sigma_z, \quad (6.23)$$

$$[\sigma_y, \sigma_z] = 2j\sigma_x, \quad (6.24)$$

$$[\sigma_z, \sigma_x] = 2j\sigma_y. \quad (6.25)$$

The two-dimensional state space can be represented as vectors in \mathbb{C}^2 according to the rule:

$$|\psi\rangle = c_g |g\rangle + c_e |e\rangle \quad \rightarrow \quad \begin{pmatrix} c_e \\ c_g \end{pmatrix}. \quad (6.26)$$

The operators are then represented by matrices

$$\sigma^+ \quad \rightarrow \quad \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}, \quad (6.27)$$

$$\sigma^- \quad \rightarrow \quad \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}, \quad (6.28)$$

$$\sigma_z \quad \rightarrow \quad \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad (6.29)$$

$$\mathbf{1} \quad \rightarrow \quad \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}. \quad (6.30)$$

6.2 The Atom-Field Interaction In Dipole Approximation

The dipole moment of an atom $\vec{\mathbf{d}}$ is determined by the position operator $\vec{\mathbf{x}}$ via

$$\vec{\mathbf{d}} = -e_0 \vec{\mathbf{x}}. \quad (6.31)$$

Then the expectation value for the dipole moment of an atom in state (6.9) is

$$\begin{aligned} \langle \psi | \vec{\mathbf{d}} | \psi \rangle &= -e_0 (|c_e|^2 \langle e | \vec{\mathbf{x}} | e \rangle + c_e c_g^* \langle g | \vec{\mathbf{x}} | e \rangle \\ &+ c_g c_e^* \langle e | \vec{\mathbf{x}} | g \rangle + |c_g|^2 \langle g | \vec{\mathbf{x}} | g \rangle). \end{aligned} \quad (6.32)$$

For simplicity, we may assume that the medium is an atomic gas. The atoms possess inversion symmetry, therefore, energy eigenstates must be symmetric or anti-symmetric, i.e. $\langle e | \vec{\mathbf{x}} | e \rangle = \langle g | \vec{\mathbf{x}} | g \rangle = 0$, see problem set 8. We obtain

$$\langle \psi | \vec{\mathbf{d}} | \psi \rangle = -e_0 (c_e c_g^* \langle g | \vec{\mathbf{x}} | e \rangle + c_g c_e^* \langle g | \vec{\mathbf{x}} | e \rangle^*). \quad (6.33)$$

Note, this means there is no permanent dipole moment in an atom, which is in an energy eigenstate. This might not be the case in a solid. The

atoms constituting the solid are oriented in a lattice, which may break the symmetry. If so, there are permanent dipole moments and consequently the matrix elements $\langle e | \vec{\mathbf{x}} | e \rangle$ and $\langle g | \vec{\mathbf{x}} | g \rangle$ would not vanish.

An atom does only exhibit a dipole moment, if the product $c_e c_g^* \neq 0$, i.e. the state of the atom is in a superposition of states $|e\rangle$ and $|g\rangle$. With the dipole matrix elements

$$\vec{M} = e_0 \langle g | \vec{\mathbf{x}} | e \rangle \quad (6.34)$$

the expectation value for the dipole moment can be written as

$$\langle \psi | \vec{\mathbf{d}} | \psi \rangle = -(c_e c_g^* \vec{M} + c_g c_e^* \vec{M}^*) = -\langle \psi | (\boldsymbol{\sigma}^- \vec{M}^* + \boldsymbol{\sigma}^+ \vec{M}) | \psi \rangle. \quad (6.35)$$

Since this is true for an arbitrary state, the dipole operator (6.31) is represented by

$$\vec{\mathbf{d}} = -(\boldsymbol{\sigma}^- \vec{M}^* + \boldsymbol{\sigma}^+ \vec{M}). \quad (6.36)$$

The energy of an electric dipole in an electric field is

$$\mathbf{H}_{A-F} = -\vec{\mathbf{d}} \cdot \vec{E}(\vec{x}_A, t). \quad (6.37)$$

We assume that the electric field is due to a monochromatic electromagnetic wave. Then the electric field at the position of the atom, \vec{x}_A , can be written as

$$\vec{E}(\vec{x}_A, t) = \frac{1}{2} (\underline{E}_0 e^{j\omega t} \vec{e}_p + \underline{E}_0^* e^{-j\omega t} \vec{e}_p^*), \quad (6.38)$$

where \underline{E}_0 denotes the complex electric field amplitude at the position of the atom and \vec{e}_p is the polarization vector of the wave. As we will see shortly, when there is a strong interaction of the wave with the atomic levels, the frequency of the electromagnetic wave is close to the atomic transition frequency $\omega \approx \omega_{eg}$. The atom-field interaction Hamiltonian operator is then

$$\mathbf{H}_{A-F} = -\vec{\mathbf{d}} \cdot \vec{E}(\vec{x}_A, t) = (\boldsymbol{\sigma}^- \vec{M}^* + \boldsymbol{\sigma}^+ \vec{M}) \frac{1}{2} (\underline{E}_0 e^{j\omega t} \vec{e}_p + \underline{E}_0^* e^{-j\omega t} \vec{e}_p^*) \quad (6.39)$$

In the Rotating-Wave Approximation (RWA)[3], we only keep the slowly varying components in the interaction Hamiltonian. If there is no field, the operator $\boldsymbol{\sigma}^+$ evolves in the Heisenberg picture of the atom according to $\boldsymbol{\sigma}^+(t) = \boldsymbol{\sigma}^+(0) e^{j\omega_{eg} t}$, thus terms proportional to the products $\boldsymbol{\sigma}^+ e^{j\omega t}$ rotate at twice the optical frequency and will be neglected in the following

$$\mathbf{H}_{A-F} \approx \mathbf{H}_{A-F}^{RWA} = \frac{1}{2} (\vec{M} \cdot \vec{e}_p^*) \underline{E}_0^* e^{-j\omega t} \boldsymbol{\sigma}^+ + h.c.. \quad (6.40)$$

The Schrödinger Equation for a two-level atom in a classical field is then

$$\begin{aligned} j\hbar \frac{d}{dt} |\psi\rangle &= (\mathbf{H}_A + \mathbf{H}_{A-F}) |\psi\rangle \\ &\approx (\mathbf{H}_A + \mathbf{H}_{A-F}^{RWA}) |\psi\rangle. \end{aligned} \quad (6.41)$$

Written in the energy representation, we obtain

$$\frac{d}{dt} c_e = -j \frac{\omega_{eg}}{2} c_e - j\Omega_r e^{-j\omega t} c_g, \quad (6.42)$$

$$\frac{d}{dt} c_g = +j \frac{\omega_{eg}}{2} c_g - j\Omega_r^* e^{+j\omega t} c_e, \quad (6.43)$$

with the Rabi-frequency defined as

$$\Omega_r = \frac{\vec{M} \cdot \vec{e}_p^*}{2\hbar} \underline{E}_0. \quad (6.44)$$

For the time being, we assume that the the Rabi-frequency is real. If this is not the case, a transformation including a phase shift in the amplitudes $c_{e,g}$ would be necessary to eliminate this phase. As expected the field couples the energy eigenstates.

6.3 Rabi-Oscillations

If the incident light has a constant field amplitude, \underline{E}_0 , Eqs. (6.42) and (6.43) can be solved and we observe an oscillation in the population difference, the Rabi-oscillation [1]. To show this we introduce the detuning between field and atomic resonance

$$\Delta = \frac{\omega_{eg} - \omega}{2} \quad (6.45)$$

and the new probability amplitudes

$$C_e = c_e e^{j\frac{\omega}{2}t}, \quad (6.46)$$

$$C_g = c_g e^{-j\frac{\omega}{2}t}. \quad (6.47)$$

This leads to the new system of equations with constant coefficients

$$\frac{d}{dt} C_e = -j\Delta C_e - j\Omega_r C_g, \quad (6.48)$$

$$\frac{d}{dt} C_g = +j\Delta C_g - j\Omega_r C_e. \quad (6.49)$$

Note, these equations are identical to the coupled mode equations between two waveguide modes as studied in section 2.7.4. But now the coupling is between modes in time, i.e. resonances. The modes are electronic ones instead of photonic modes. But otherwise what has been said in section 2.7.4 applies in the same way. For the case of vanishing detuning it is especially easy to eliminate one of the variables and we arrive at

$$\frac{d^2}{dt^2}C_e = -\Omega_r^2 C_e \quad (6.50)$$

$$\frac{d^2}{dt^2}C_g = -\Omega_r^2 C_g. \quad (6.51)$$

The solution to this set of equations are the oscillations we are looking for. If the atom is at time $t = 0$ in the ground-state, i.e. $C_g(0) = 1$ and $C_e(0) = 0$, respectively, we arrive at

$$C_g(t) = \cos(\Omega_r t) \quad (6.52)$$

$$C_e(t) = -j \sin(\Omega_r t). \quad (6.53)$$

Then, the probabilities for finding the atom in the ground or excited state are

$$|c_b(t)|^2 = \cos^2(\Omega_r t) \quad (6.54)$$

$$|c_a(t)|^2 = \sin^2(\Omega_r t), \quad (6.55)$$

as shown in Fig. 6.2. For the expectation value of the dipole operator under the assumption of a real dipole matrix element $\vec{M} = \vec{M}^*$ we obtain

$$\langle \psi | \vec{\mathbf{d}} | \psi \rangle = -\vec{M} c_e c_g^* + c.c. \quad (6.56)$$

$$= -\vec{M} \sin(2\Omega_r t) \sin(\omega_{eg} t). \quad (6.57)$$

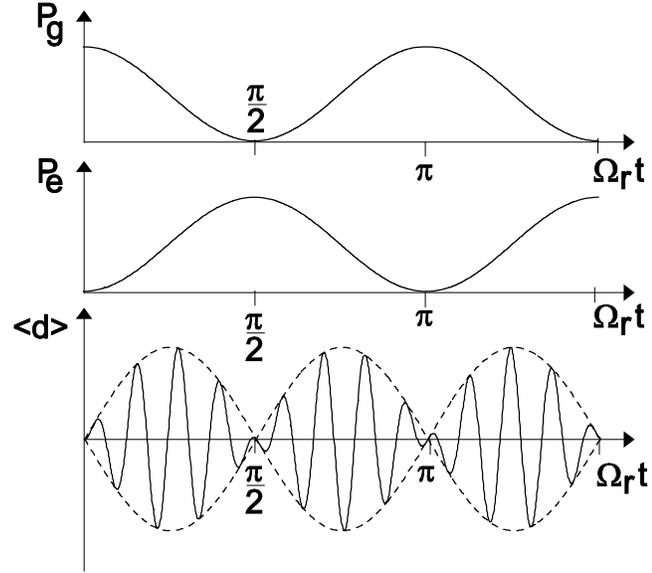


Figure 6.2: Evolution of occupation probabilities of ground and excited state and the average dipole moment of a two-level atom in resonant interaction with a coherent classical field.

The coherent external field drives the population of the atomic system between the two available states with a period $T_r = \pi/\Omega_r$. Applying the field only over half of this period leads to a complete inversion of the population. These Rabi-oscillations have been observed in various systems ranging from gases to semiconductors. Interestingly, the light emitted from the coherently driven two-level atom is not identical in frequency to the driving field. If we look at the Fourier spectrum of the polarization according to Eq.(6.57), we obtain lines at frequencies $\omega_{\pm} = \omega_{eg} \pm 2\Omega_r$. This is clearly a nonlinear output and the sidebands are called Mollow-sidebands [2]. Most important for the existence of these oscillations is the coherence of the atomic system over at least one Rabi-oscillation. If this coherence is destroyed fast enough, the Rabi-oscillations cannot happen and it is then impossible to generate inversion in a two-level system by interaction with light. This is the case for a large class of situations in light-matter interaction and especially for typical laser materials. So we are interested in finding out what happens in the case of loss of coherence in the atomic system due to additional interaction of the atoms with its environment.

6.4 The Density Operator

To study incoherent or dissipative processes it is necessary to switch to a statistical description. That is, we investigate not only the interaction of the atoms with the light field, via the Schrodinger Equation, leading to Rabi-oscillations but rather the interaction of an atomic ensemble with the light field. This is achieved by using the density operator instead of deterministic wave functions, similar to classical statistical mechanics, where the deterministic trajectories of particles are replaced by probability distributions.

The density operator of a pure state is defined by the dyadic product of the state with itself

$$\rho = |\psi\rangle \langle\psi| \quad (6.58)$$

or in the energy representation by a 2×2 -matrix

$$\rho = \begin{pmatrix} \rho_{ee} & \rho_{eg} \\ \rho_{ge} & \rho_{gg} \end{pmatrix}. \quad (6.59)$$

. In the case of a pure state (6.9) this is

$$\rho = \begin{pmatrix} c_e c_e^* & c_e c_g^* \\ c_g c_e^* & c_g c_g^* \end{pmatrix}. \quad (6.60)$$

For the rather simple case of a two-level system, each element of the density matrix corresponds to a physical quantity. The main diagonal contains the population probabilities for the levels; the off-diagonal element is the expectation value of the positive or negative frequency component of the dipole moment of the atom, i.e. its contribution to the polarization in the medium.

However, the concept of a density operator can be applied to any quantum mechanical system, not just the two-level atom. If an ensemble is described by a density operator, the expectation value of an arbitrary operator \mathbf{A} can be computed using the trace formula

$$\langle \mathbf{A} \rangle = Tr\{\rho \mathbf{A}\}. \quad (6.61)$$

The trace of an operator is defined as

$$Tr\{\mathbf{O}\} = \sum_n \langle n | \mathbf{O} | n \rangle. \quad (6.62)$$

where $|n\rangle$ can be any complete orthonormal base (ONB) in the Hilbert space. For example for the density matrix of the pure state (6.58) we find

$$\langle \mathbf{A} \rangle = \text{Tr}\{\rho \mathbf{A}\} = \sum_n \langle n | \rho \mathbf{A} | n \rangle \quad (6.63)$$

$$= \sum_n \langle n | \psi \rangle \langle \psi | \mathbf{A} | n \rangle = \langle \psi | \mathbf{A} \sum_n | n \rangle \langle n | \psi \rangle \quad (6.64)$$

$$= \langle \psi | \mathbf{A} | \psi \rangle. \quad (6.65)$$

The advantage of the density operator is that it can also be applied to a statistical mixture of pure states. For example, if the atom is in state $|e\rangle$ with probability p_e and in state $|g\rangle$ with probability p_g a density operator

$$\rho = p_e |e\rangle \langle e| + p_g |g\rangle \langle g| \quad (6.66)$$

is defined, which can be used to compute the average values of observables in the proper statistical sense

$$\langle \mathbf{A} \rangle = \text{Tr}\{\rho \mathbf{A}\} = p_e \langle e | \mathbf{A} | e \rangle + p_g \langle g | \mathbf{A} | g \rangle. \quad (6.67)$$

Since the matrices (6.27) to (6.30) build a complete base in the space of 2×2 -matrices, we can express the density matrix as

$$\rho = \rho_{ee} \frac{1}{2}(\mathbf{1} + \sigma_z) + \rho_{gg} \frac{1}{2}(\mathbf{1} - \sigma_z) + \rho_{eg} \sigma^+ + \rho_{ge} \sigma^- \quad (6.68)$$

$$= \frac{1}{2} \mathbf{1} + \frac{1}{2}(\rho_{ee} - \rho_{gg}) \sigma_z + \rho_{eg} \sigma^+ + \rho_{ge} \sigma^-, \quad (6.69)$$

since the trace of the density matrix is always one (normalization). Choosing the new base $\mathbf{1}, \sigma_x, \sigma_y, \sigma_z$, we obtain

$$\rho = \frac{1}{2} \mathbf{1} + \frac{1}{2}(\rho_{ee} - \rho_{gg}) \sigma_z + d_x \sigma_x + d_y \sigma_y, \quad (6.70)$$

with

$$d_x = \frac{1}{2}(\rho_{eg} + \rho_{ge}) = \Re\{\langle \sigma^{(+)} \rangle\}, \quad (6.71)$$

$$d_y = \frac{j}{2}(\rho_{eg} - \rho_{ge}) = \Im\{\langle \sigma^{(+)} \rangle\}. \quad (6.72)$$

The expectation value of the dipole operator is given by (6.36)

$$\langle \vec{\mathbf{d}} \rangle = \text{Tr}\{\rho \vec{\mathbf{d}}\} = -\vec{M}^* \text{Tr}\{\rho \sigma^+\} + c.c. = -\vec{M}^* \rho_{ge} + c.c. \quad (6.73)$$

From the Schrödinger equation for the wave function $|\psi\rangle$ we can easily derive the equation of motion for the density operator called the von Neumann equation

$$\begin{aligned}\dot{\rho} &= \frac{d}{dt} |\psi\rangle \langle\psi| + h.c. = \frac{1}{j\hbar} \mathbf{H} |\psi\rangle \langle\psi| - \frac{1}{j\hbar} |\psi\rangle \langle\psi| \mathbf{H} \\ &= \frac{1}{j\hbar} [\mathbf{H}, \rho].\end{aligned}\quad (6.74)$$

Due to the linear nature of this equation, this is also the correct equation for a density operator describing an arbitrary mixture of states. In case of a two-level atom, the von Neumann equation is

$$\dot{\rho} = \frac{1}{j\hbar} [\mathbf{H}_A, \rho] = -j \frac{\omega_{eg}}{2} [\sigma_z, \rho].\quad (6.75)$$

Using the commutator relations (6.16) - (6.18), the result is

$$\dot{\rho}_{ee} = 0, \quad (6.76)$$

$$\dot{\rho}_{gg} = 0, \quad (6.77)$$

$$\dot{\rho}_{eg} = -j\omega_{eg}\rho_{eg} \quad \rightarrow \quad \rho_{eg}(t) = e^{-j\omega_{eg}t}\rho_{eg}(0), \quad (6.78)$$

$$\dot{\rho}_{ge} = j\omega_{eg}\rho_{ge} \quad \rightarrow \quad \rho_{ge}(t) = e^{j\omega_{eg}t}\rho_{ge}(0). \quad (6.79)$$

Again the isolated two-level atom has rather simple dynamics. The populations are constant. If there is a dipole moment induced at $t = 0$, i.e. the system is in a superposition state, then this dipole moment oscillates with the transition frequency ω_{eg} .

6.5 Energy- and Phase-Relaxation

In reality one has to work very hard to isolated an atom from its environment. Indeed in the case of laser active media, we are interested at radiating atoms, i.e. atoms that have a dipole interaction with the field. The coupling with the infinitely many modes of the free field leads already to spontaneous emission, an irreversible process. We could treat this process by using the Hamiltonian

$$\mathbf{H} = \mathbf{H}_A + \mathbf{H}_F + \mathbf{H}_{A-F}. \quad (6.80)$$

Here, \mathbf{H}_A is the Hamiltonian of the atom, \mathbf{H}_F of the free field in thermal equilibrium at temperature T , and \mathbf{H}_{A-F} describes the interaction between

them. A complete treatment along these lines would be straight forward using the techniques we learned so far, however it is beyond the scope of this class. The result of this calculation leads to the von Neumann equation of the reduced density matrix, i.e. the density matrix of the atom alone. In fact the result of such a calculation gives for the diagonal elements of the density operator, i.e. the state population probabilities, equations identical to those in section 3.3 involving Einstein's A and B coefficients. With the spontaneous emission rate $A = 1/\tau_{sp}$, i.e. the inverse spontaneous life time τ_{sp} , the populations change due to the induced and spontaneous emission processes and the absorption processes

$$\frac{d}{dt}|c_e(t)|^2 = \frac{d}{dt}\rho_{ee} = -\Gamma_e\rho_{ee} + \Gamma_a\rho_{gg} \quad (6.81)$$

with the abbreviations

$$\Gamma_e = \frac{1}{\tau_{sp}}(n_{th} + 1), \quad (6.82)$$

$$\Gamma_a = \frac{1}{\tau_{sp}}n_{th}. \quad (6.83)$$

see Figure 6.3.

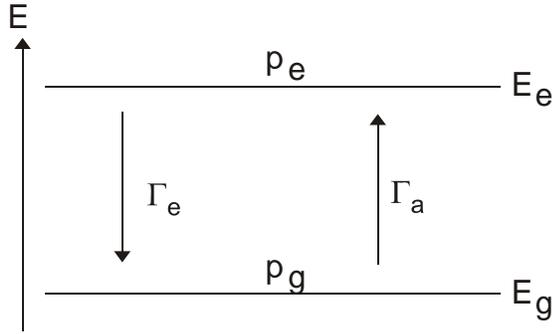


Figure 6.3: Two-level atom with transition rates due to induced and spontaneous emission and absorption.

Here n_{th} is the number of thermally excited photons in the modes of the free field with frequency ω_{eg} , $n_{th} = 1/(\exp(\hbar\omega_{eg}/kT) - 1)$, at temperature T .

The total probability of being in the excited or the ground state has to be maintained, that is

$$\frac{d}{dt}\rho_{gg} = -\frac{d}{dt}\rho_{ee} = \Gamma_e\rho_{ee} - \Gamma_a\rho_{gg}. \quad (6.84)$$

If the populations decay, the polarization does as well, since $\rho_{ge} = c_e^*c_g$. It turns out that the polarization dynamics according to Eq.(6.78), besides the coherent oscillation, also acquires a decay process due to the finite lifetime of the excited state

$$\frac{d}{dt}\rho_{ge} = j\omega_{eg}\rho_{ge} - \frac{\Gamma_e + \Gamma_a}{2}\rho_{ge}. \quad (6.85)$$

Thus the absorption as well as the emission processes are destructive to the phase. Therefore, the corresponding rates add up in the phase decay rate.

Taking the coherent (6.76)-(6.79) and incoherent processes (6.84-6.85) into account results in the following equations for the normalized average dipole moment $d = d_x + jd_y$ and the inversion w

$$\dot{d} = \dot{\rho}_{ge} = (j\omega_{eg} - \frac{1}{T_2})d, \quad (6.86)$$

$$\dot{w} = \dot{\rho}_{ee} - \dot{\rho}_{gg} = -\frac{w - w_0}{T_1}, \quad (6.87)$$

with the time constants

$$\frac{1}{T_1} = \frac{2}{T_2} = \Gamma_e + \Gamma_a = \frac{2n_{th} + 1}{\tau_{sp}} \quad (6.88)$$

and the equilibrium inversion w_0 , due to the thermal excitation of the atom by the thermal field

$$w_0 = \frac{\Gamma_a - \Gamma_e}{\Gamma_a + \Gamma_e} = \frac{-1}{1 + 2n_{th}} = -\tanh\left(\frac{\hbar\omega_{eg}}{2kT}\right). \quad (6.89)$$

The time constant T_1 denotes the energy relaxation in the two-level system and T_2 the phase relaxation. T_2 is the correlation time between amplitudes c_e and c_g . The coherence between the excited and the ground state described by ρ_{ge} is destroyed by the interaction of the two-level system with the environment.

In this basic model, the energy relaxation is half the phase relaxation rate or

$$T_2 = 2T_1. \quad (6.90)$$

The atoms in a laser medium do not only interact with the electromagnetic field, but also with phonons, i.e. acoustic vibrations of the host lattice in solid state laser material. Atoms might collide with each other in a gas laser and so on. All these processes must be considered when determining the energy and phase relaxation rates. Thus it might be not only radiative transitions that lead to a finite energy relaxation time T_1 . Some of the processes are elastic, i.e. there is no energy relaxation but only the phase is influenced during the collision. Therefore, these processes reduce T_2 but have no influence on T_1 . In real systems the phase relaxation time is most often much shorter than twice the energy relaxation time.

$$T_2 \leq 2T_1. \quad (6.91)$$

If the inversion deviates from its equilibrium value, w_0 , it relaxes back into equilibrium with a time constant T_1 . Eq. (6.89) shows that for all temperatures $T > 0$ the inversion is negative, i.e. the lower level is stronger populated than the upper level. Thus with incoherent thermal light, inversion in a two-level system cannot be achieved. Inversion can only be achieved by pumping with incoherent light, if there are more levels and subsequent relaxation processes into the upper laser level. Due to these relaxation processes the rate Γ_a deviates from the equilibrium expression (6.83), and it has to be replaced by the pump rate Λ . If the pump rate Λ exceeds Γ_e , the inversion corresponding to Eq. (6.89) becomes positive,

$$w_0 = \frac{\Lambda - \Gamma_e}{\Lambda + \Gamma_e}. \quad (6.92)$$

If we allow for artificial negative temperatures, we obtain with $T < 0$ for the ratio of relaxation rates

$$\frac{\Gamma_e}{\Gamma_a} = \frac{1 + \bar{n}}{\bar{n}} = e^{\frac{\hbar\omega_{eg}}{kT}} < 1. \quad (6.93)$$

Thus the pumping of the two-level system drives the system far away from thermal equilibrium. Now, we have a correct description of an ensemble of atoms in thermal equilibrium with its environment, which is a much more realistic description of media especially of typical laser media.

6.6 The Bloch Equations

If there is a coherent additional field in addition to the coupling to the environment, the Hamiltonian has to be extended by the dipole interaction with

that field,

$$\mathbf{H}_E = -\vec{\mathbf{d}} \cdot \vec{E}(\vec{x}_A, t). \quad (6.94)$$

Again we use the interaction Hamiltonian in RWA according to Eq.(6.40) for a time harmonic field Eq.(6.38) with polarization vector \vec{e}_p

$$\mathbf{H}_E = \frac{1}{2} \left(\vec{M} \cdot \vec{e}_p^* \right) \underline{E}_0^* e^{-j\omega t} \boldsymbol{\sigma}^+ + h.c.. \quad (6.95)$$

In the von Neumann equation this leads to the additional term

$$\dot{\rho}|_E = \frac{1}{j\hbar} [\mathbf{H}_E, \rho] \quad (6.96)$$

$$= -j\Omega_r e^{-j\omega t} [\boldsymbol{\sigma}^+, \rho] + j\Omega_r^* e^{j\omega t} [\boldsymbol{\sigma}^-, \rho]. \quad (6.97)$$

With the density operator expressed as

$$\rho = \frac{1}{2} \mathbf{1} + \frac{1}{2} (\rho_{ee} - \rho_{gg}) \boldsymbol{\sigma}_z + \rho_{eg} \boldsymbol{\sigma}^+ + \rho_{ge} \boldsymbol{\sigma}^-, \quad (6.98)$$

and the commutation relations (6.16) - (6.18) we find

$$\begin{aligned} \dot{\rho}|_E &= \frac{1}{2} (\dot{\rho}_{ee} - \dot{\rho}_{gg}) \boldsymbol{\sigma}_z + \dot{\rho}_{eg} \boldsymbol{\sigma}^+ + \dot{\rho}_{ge} \boldsymbol{\sigma}^- \quad (6.99) \\ &= -j\Omega_r e^{-j\omega t} \left\{ \frac{1}{2} (\rho_{ee} - \rho_{gg}) [\boldsymbol{\sigma}^+, \boldsymbol{\sigma}_z] + \rho_{ge} [\boldsymbol{\sigma}^+, \boldsymbol{\sigma}^-] \right\} + \\ &\quad + j\Omega_r^* e^{j\omega t} \left\{ \frac{1}{2} (\rho_{ee} - \rho_{gg}) [\boldsymbol{\sigma}^-, \boldsymbol{\sigma}_z] + \rho_{eg} [\boldsymbol{\sigma}^-, \boldsymbol{\sigma}^+] \right\} \\ &= j\Omega_r e^{-j\omega t} \{ (\rho_{ee} - \rho_{gg}) \boldsymbol{\sigma}^+ + \rho_{ge} \boldsymbol{\sigma}_z \} \\ &\quad + j\Omega_r^* e^{j\omega t} \{ (\rho_{ee} - \rho_{gg}) \boldsymbol{\sigma}^- - \rho_{eg} \boldsymbol{\sigma}_z \} \end{aligned}$$

or expressed by the components of the density operator

$$(\dot{\rho}_{ee} - \dot{\rho}_{gg})|_E = 2j\Omega_r e^{-j\omega t} \rho_{ge} + c.c., \quad (6.100)$$

$$\dot{\rho}_{ge}|_E = j\Omega_r^* e^{j\omega t} (\rho_{ee} - \rho_{gg}), \quad (6.101)$$

The interaction with the external field leads to the following contributions in the dynamics of the dipole moment and the inversion

$$\dot{d}|_E = \dot{\rho}_{ge}|_E = j\Omega_r^* e^{j\omega t} w, \quad (6.102)$$

$$\dot{w}|_E = \dot{\rho}_{ee}|_E - \dot{\rho}_{gg}|_E = 2j\Omega_r e^{-j\omega t} d + c.c. \quad (6.103)$$

Thus, the total dynamics of the two-level system including the pumping and dephasing processes from Eqs.(6.86) and (6.87) is given by

$$\dot{d} = -\left(\frac{1}{T_2} - j\omega_{eg}\right)d + j\Omega_r^* e^{j\omega t} w, \quad (6.104)$$

$$\dot{w} = -\frac{w - w_0}{T_1} + 2j\Omega_r e^{-j\omega t} d - 2j\Omega_r^* e^{j\omega t} d^*. \quad (6.105)$$

These equations are called the Bloch Equations. They describe the dynamics of a statistical ensemble of two-level atoms interacting with a classical electric field. Together with the Maxwell-Equations, where the polarization of the medium is related to the expectation value of the dipole moment of the atomic ensemble these result in the Maxwell-Bloch Equations.

6.7 Dielectric Susceptibility and Saturation

We have assumed that the external field is time harmonic

$$\vec{E}(\vec{x}_A, t) = \frac{1}{2} (\underline{E}_0 e^{j\omega t} \vec{e}_p + \underline{E}_0^* e^{-j\omega t} \vec{e}_p^*). \quad (6.106)$$

The Bloch Equations are nonlinear. However, for moderate field intensities, i.e. the magnitude of the Rabi-frequency is much smaller than the optical frequency, $|\Omega_r| \ll \omega$, the inversion does not change much within an optical cycle of the field. We assume that the inversion w of the atom will only be slowly changing and it adjusts itself to a steady state value w_s . If the inversion can be assumed time independent, $w = w_s$ the equation for the dipole moment is linear and the dipole moment will oscillate with the same frequency as the driving field

$$\underline{d} = \underline{d}_0 e^{j\omega t}. \quad (6.107)$$

With the time harmonic solution (6.107) we find from Eqs. (6.104) and (6.105) for the dipole amplitude and the steady state inversion

$$\underline{d}_0 = \frac{j \left(\vec{M}^* \cdot \vec{e}_p \right) w_s}{2\hbar \left(1/T_2 + j(\omega - \omega_{eg}) \right)} \underline{E}_0 \quad (6.108)$$

$$w_s = \frac{w_0}{1 + \frac{T_1}{\hbar^2} \frac{1/T_2 |\vec{M}^* \cdot \vec{e}_p|^2}{(1/T_2)^2 + (\omega_{eg} - \omega)^2} |\underline{E}_0|^2}. \quad (6.109)$$

We introduce the normalized lineshape function, which is in this case a Lorentzian,

$$L(\omega) = \frac{(1/T_2)^2}{(1/T_2)^2 + (\omega_{eg} - \omega)^2}, \quad (6.110)$$

and connect the square of the field $|\underline{E}_0|^2$ to the intensity I of a propagating plane wave, according to Eq. (2.38), $I = \frac{1}{2Z_F} |\underline{E}_0|^2$,

$$w_s = \frac{w_0}{1 + \frac{I}{I_s} L(\omega)}. \quad (6.111)$$

Thus the stationary inversion depends on the intensity of the incident light. Therefore, w_0 is called the unsaturated inversion, w_s the saturated inversion and I_s , with

$$I_s = \left[\frac{2T_1 T_2 Z_F}{\hbar^2} |\vec{M}^* \cdot \vec{e}_p|^2 \right]^{-1}, \quad (6.112)$$

is the saturation intensity. The expectation value of the dipole operator (6.31) is then given by

$$\langle \tilde{\mathbf{d}} \rangle = -(\vec{M}^* \underline{d} + \vec{M} \underline{d}^*). \quad (6.113)$$

Multiplication with the number of atoms per unit volume, N , relates the dipole moment of the atom to the complex polarization \vec{P}_0 of the medium, and therefore to the susceptibility according to

$$\vec{P}_0 = -2N \vec{M}^* \underline{d}_0, \quad (6.114)$$

$$\vec{P}_0 = \epsilon_0 \chi(\omega) \vec{e}_p \underline{E}_0. \quad (6.115)$$

From the definitions (6.114), (6.115) and Eq. (6.108) we obtain for the linear susceptibility of the medium

$$\chi(\omega) = \vec{M}^* \vec{M}^T \frac{jN}{\hbar \epsilon_0} \frac{w_s}{1/T_2 + j(\omega - \omega_{eg})}, \quad (6.116)$$

which is a tensor. In the following we assume that the direction of the atom is random, i.e. the alignment of the atomic dipole moment, \vec{M} , and the electric field is random. Therefore, we have to average over the angle enclosed

between the electric field of the wave and the atomic dipole moment, which results in

$$\overline{\begin{pmatrix} M_x M_x & M_x M_y & M_x M_z \\ M_y M_x & M_y M_y & M_y M_z \\ M_z M_x & M_z M_y & M_z M_z \end{pmatrix}} = \begin{pmatrix} \overline{M_x^2} & 0 & 0 \\ 0 & \overline{M_y^2} & 0 \\ 0 & 0 & \overline{M_z^2} \end{pmatrix} = \frac{1}{3} |\vec{M}|^2 \mathbf{1}. \quad (6.117)$$

Thus, for homogeneous and isotropic media the susceptibility tensor shrinks to a scalar

$$\chi(\omega) = \frac{1}{3} |\vec{M}|^2 \frac{jN}{\hbar\epsilon_0} \frac{w_s}{1/T_2 + j(\omega - \omega_{eg})}. \quad (6.118)$$

Real and imaginary part of the susceptibility

$$\chi(\omega) = \chi'(\omega) + j\chi''(\omega) \quad (6.119)$$

are then given by

$$\chi'(\omega) = -\frac{|\vec{M}|^2 N w_s T_2^2 (\omega_{eg} - \omega)}{3\hbar\epsilon_0} L(\omega), \quad (6.120)$$

$$\chi''(\omega) = \frac{|\vec{M}|^2 N w_s T_2}{3\hbar\epsilon_0} L(\omega). \quad (6.121)$$

If the incident radiation is weak, i.e.

$$\frac{I}{I_s} L(\omega) \ll 1 \quad (6.122)$$

we obtain $w_s \approx w_0$. For optical transitions there is no thermal excitation of the excited state and $w_0 = -1$. For an inverted system, $w_0 > 0$, the real and imaginary parts of the susceptibility are shown in Fig. 6.4.

The shape of the susceptibility computed quantum mechanically compares well with the classical susceptibility (2.43) derived from the harmonic oscillator model close to the transition frequency ω_{eg} for a transition with reasonably high $Q = T_2\omega_{eg}$. Note, the quantum mechanical susceptibility is identical to the complex Lorentzian introduced in Eq.(2.90). There is an appreciable deviation, however, far away from resonance. Far off resonance the rotating wave approximation should not be used.

The physical meaning of the real and imaginary part of the susceptibility is of course identical to section 2.1.8. The propagation constant k of a TEM-wave in such a medium is related to the susceptibility by

$$k = \omega \sqrt{\mu_0 \epsilon_0 (1 + \chi(\omega))} \approx k_0 \left(1 + \frac{1}{2} \chi(\omega) \right), \quad \text{with } k_0 = \omega \sqrt{\mu_0 \epsilon_0} \quad (6.123)$$

for $|\chi| \ll 1$. Under this assumption we obtain

$$k = k_0 \left(1 + \frac{\chi'}{2} \right) + j k_0 \frac{\chi''}{2}. \quad (6.124)$$

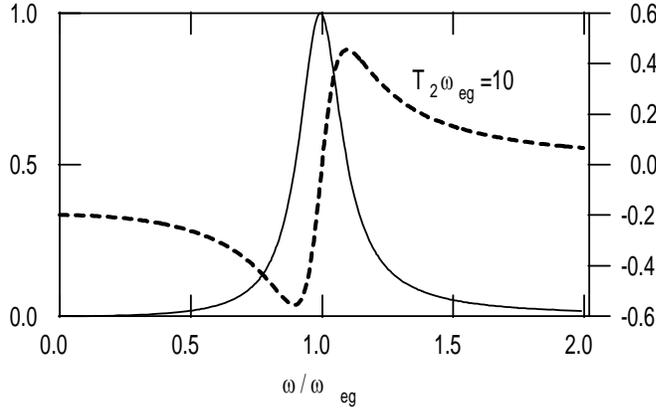


Figure 6.4: Real and imaginary part of the complex susceptibility for an inverted medium $w_s > 0$. The positive imaginary susceptibility indicates exponential growth of an electromagnetic wave propagating in the medium.

The real part of the susceptibility contributes to the refractive index $n = 1 + \chi'/2$. In the case of $\chi'' < 0$, the imaginary part leads to an exponential damping of the wave. For $\chi'' > 0$ amplification takes place. Amplification of the wave is possible for $w_0 > 0$, i.e. an inverted medium.

The phase relaxation rate $1/T_2$ of the dipole moment determines the width of the absorption line or the bandwidth of the amplifier. The amplification can not occur forever, because the amplifier saturates when the intensity reaches the saturation intensity. This is a strong deviation from the linear susceptibility we derived from the classical oscillator model. The reason for this saturation is two fold. First, the light can not extract more energy from the atoms then there is energy stored in them, i.e. energy conservation holds. Second the induced dipole moment in a two-level atom is limited by the maximum value of the matrix element. In contrast the induced dipole moment in a classical oscillator grows proportionally to the applied field without limits.

6.8 Rate Equations and Cross Sections

In many cases the fastest process in the atom-field interaction dynamics is the dephasing of the dipole moment, i. e. $T_2 \rightarrow 0$. For example, in semi-conductors $T_2 < 50fs$. In those cases the magnitude of the dipole moment relaxes instantaneously into the steady state and follows the magnitude of the intensity envelope $I(t)$ of the electromagnetic field, which evolves on a much slower time scale. From Eq.(6.104) we obtain with the steady state solutions for the dipole moment (6.107) and (6.108) for the time dependent inversion in the atomic system after adiabatic elimination of dipole moment

$$\dot{w} = -\frac{w(t) - w_0}{T_1} - \frac{w(t)}{T_1 I_s} L(\omega) I(t), \quad (6.125)$$

where $I(t)$ is the intensity of the electromagnetic wave interaction with the two-level atom. In this limit the Bloch Equations became simple rate equations. We only take care of the counting of population differences due to spontaneous and stimulated emissions.

The interaction of an atom with light at a given transition with the stream of photons on resonance, i.e. $\omega = \omega_{eg}$ is often described by the mass action law, that is that the number of induced transitions, for example from the excited to the ground state, is proportional to the product of the number of atoms in the excited state and the photon flux density $I_{ph} = I/\hbar\omega_{eg}$

$$\dot{w}|_{induced} = -\sigma w I_{ph} = -\frac{w}{T_1 I_s} I. \quad (6.126)$$

Thus the interaction cross section can be expressed in terms of the saturation intensity as

$$\sigma = \frac{\hbar\omega_{eg}}{T_1 I_s} \quad (6.127)$$

$$= \frac{2\omega_{eg} T_2 Z_F}{\hbar} |\vec{M}^* \cdot \vec{e}_p|^2. \quad (6.128)$$

In this chapter, we have introduced the most important spectroscopic quantities that characterize an atomic transition, which are the lifetime of the excited state or often called upper-state lifetime or longitudinal lifetime T_1 , the phase relaxation time or transverse relaxation time T_2 which is the inverse half-width at half maximum of the line and the interaction cross-section that only depends on the dipole matrix element and the linewidth of the transition.

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