# Chapter 6

# Interaction of Light and Matter

Atomic or molecular gases in low concentration show sharp energy eigenspectra. 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

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

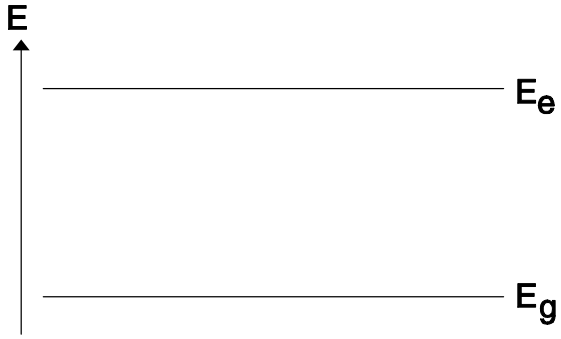


Figure 6.1: Two-level atom

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

$$\mathbf{H}_A = E_e \left|e\right\rangle \left\langle e\right| + E_g \left|e\right\rangle \left\langle g\right| . \tag{6.2}$$

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

$$\begin{aligned} \mathbf{1} &= \left|e\right\rangle \left\langle e\right| + \left|g\right\rangle \left\langle g\right| , && (6.3)\\ \boldsymbol{\sigma}_z &= \left|e\right\rangle \left\langle e\right| - \left|g\right\rangle \left\langle g\right| , && (6.4)\\ \boldsymbol{\sigma}^+ &= \left|e\right\rangle \left\langle g\right| , && (6.5)\\ \boldsymbol{\sigma}^- &= \left|e\right\rangle \left\langle e\right| . && (6.6) \end{aligned}$$

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

$$\begin{aligned} \boldsymbol{\sigma}_x &= \boldsymbol{\sigma}^+ + \boldsymbol{\sigma}^-, && (6.7)\\ \boldsymbol{\sigma}_y &= -\mathrm{j}\boldsymbol{\sigma}^+ + \mathrm{j}\boldsymbol{\sigma}^- . && (6.8) \end{aligned}$$

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

$$\left|\psi\right\rangle = c_g \left|g\right\rangle + c_e \left|e\right\rangle . \tag{6.9}$$

We obtain

$$\begin{aligned} \boldsymbol{\sigma}^+ \left|\psi\right\rangle &= c_g \left|e\right\rangle , && (6.10)\\ \boldsymbol{\sigma}^- \left|\psi\right\rangle &= c_e \left|g\right\rangle , && (6.11)\\ \boldsymbol{\sigma}_z \left|\psi\right\rangle &= c_e \left|e\right\rangle - c_g \left|g\right\rangle . && (6.12) \end{aligned}$$

The operator $\boldsymbol{\sigma}^+$ generates a transition from the ground to the excited state, and $\boldsymbol{\sigma}^-$ does the opposite. In contrast to $\boldsymbol{\sigma}^+$ and $\boldsymbol{\sigma}^-$, $\boldsymbol{\sigma}_z$ is a Hermitian operator, and its expectation value is an observable physical quantity with expectation value

$$\langle\psi|\,\boldsymbol{\sigma}_z\,|\psi\rangle = |c_e|^2 - |c_g|^2 = w, \tag{6.13}$$

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 $W = N\,\langle\psi|\,\boldsymbol{\sigma}_z\,|\psi\rangle$. 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

$$\mathbf{H}_A = \frac{1}{2}\hbar\omega_{eg}\boldsymbol{\sigma}_z, \tag{6.14}$$

with the transition frequency

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

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

$$\begin{aligned}
[\boldsymbol{\sigma}^+, \boldsymbol{\sigma}^-] &= \boldsymbol{\sigma}_z, && (6.16)\\
[\boldsymbol{\sigma}^+, \boldsymbol{\sigma}_z] &= -2\boldsymbol{\sigma}^+, && (6.17)\\
[\boldsymbol{\sigma}^-, \boldsymbol{\sigma}_z] &= 2\boldsymbol{\sigma}^-, && (6.18)
\end{aligned}$$

and anti-commutator relations, respectively

$$\begin{aligned}
[\boldsymbol{\sigma}^+, \boldsymbol{\sigma}^-]_+ &= \mathbf{1}, && (6.19)\\
[\boldsymbol{\sigma}^+, \boldsymbol{\sigma}_z]_+ &= \mathbf{0}, && (6.20)\\
[\boldsymbol{\sigma}^-, \boldsymbol{\sigma}_z]_+ &= \mathbf{0}, && (6.21)\\
[\boldsymbol{\sigma}^-, \boldsymbol{\sigma}^-]_+ &= [\boldsymbol{\sigma}^+, \boldsymbol{\sigma}^+]_+ = \mathbf{0}. && (6.22)
\end{aligned}$$

The operators $\boldsymbol{\sigma}_x$, $\boldsymbol{\sigma}_y$, $\boldsymbol{\sigma}_z$ fulfill the angular momentum commutator relations

$$\begin{aligned}
[\boldsymbol{\sigma}_x, \boldsymbol{\sigma}_y] &= 2\mathrm{j}\boldsymbol{\sigma}_z, && (6.23)\\
[\boldsymbol{\sigma}_y, \boldsymbol{\sigma}_z] &= 2\mathrm{j}\boldsymbol{\sigma}_x, && (6.24)\\
[\boldsymbol{\sigma}_z, \boldsymbol{\sigma}_x] &= 2\mathrm{j}\boldsymbol{\sigma}_y. && (6.25)
\end{aligned}$$

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

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

The operators are then represented by matrices

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

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

$$\boldsymbol{\sigma}_z \quad \rightarrow \quad \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{6.29}$$

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

## 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

$$\vec{\mathbf{d}} = -e_0\vec{\mathbf{x}}. \tag{6.31}$$

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

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

For simplicity, we may assume that the medium is an atomic gas. The atoms posses 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

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

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 consituting 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

$$\vec{M} = e_0 \langle g|\,\vec{\mathbf{x}}\,|e\rangle \tag{6.34}$$

the expectation value for the dipole moment can be written as

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

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

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

The energy of an electric dipole in an electric field is

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

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

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

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 transistion frequency $\omega \approx \omega_{eg}$. The atom-field interaction Hamiltonian operator is then

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

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 $\boldsymbol{\sigma}^+(t) = \boldsymbol{\sigma}^+(0) e^{\mathrm{j}\omega_{eg} t}$, thus terms proportional to the products $\boldsymbol{\sigma}^+ e^{\mathrm{j}\omega t}$ rotate at twice the optical frequency and will be neglected in the following

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

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

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

Written in the energy representation, we obtain

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

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

with the Rabi-frequency defined as

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

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 $c_{\in,g}$ 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

$$\Delta = \frac{\omega_{eg} - \omega}{2} \tag{6.45}$$

and the new probability amplitudes

$$C_e = c_e e^{\mathrm{j}\frac{\omega}{2}t}, \tag{6.46}$$

$$C_g = c_g e^{-\mathrm{j}\frac{\omega}{2}t}. \tag{6.47}$$

This leads to the new system of equations with constant coefficients

$$\frac{d}{dt}C_e = -\mathrm{j}\Delta C_e - \mathrm{j}\Omega_r C_g, \tag{6.48}$$

$$\frac{d}{dt}C_g = +\mathrm{j}\Delta C_g - \mathrm{j}\Omega_r C_e. \tag{6.49}$$

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

$$\frac{d^2}{dt^2}C_e = -\Omega_r^2 C_e \tag{6.50}$$

$$\frac{d^2}{dt^2}C_g = -\Omega_r^2 C_g. \tag{6.51}$$

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

$$C_g(t) = \cos(\Omega_r t) \tag{6.52}$$

$$C_e(t) = -\mathrm{j}\sin(\Omega_r t). \tag{6.53}$$

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

$$|c_b(t)|^2 = \cos^2(\Omega_r t) \tag{6.54}$$

$$|c_a(t)|^2 = \sin^2(\Omega_r t), \tag{6.55}$$

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

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

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

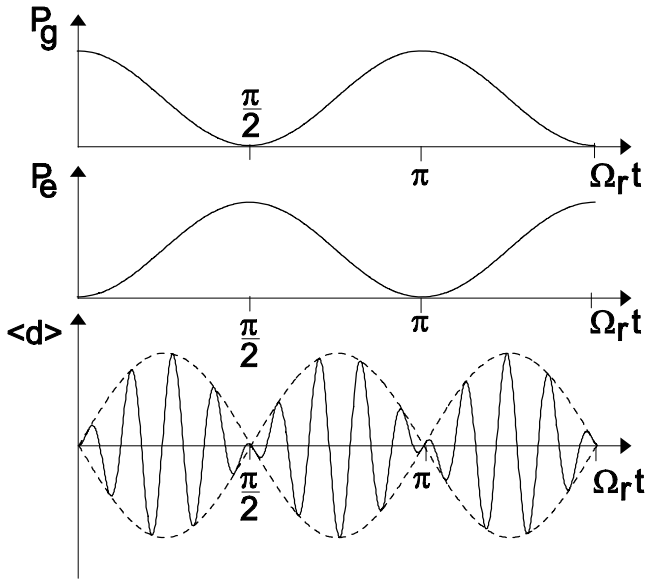


Figure 6.2: Evolution of occupation probabilities of ground and excited state and the average dipole moment of a two-level atom in resonant interaction with a coherent classical field.

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.