7 Interaction of Atoms With a Classical Light Field

7.1 The electron wavefunction and the two-level atom

The wavefunction of an electron $\Psi(x)$ can be decomposed with a complete set of eigenfunctions $\psi_i(x)$ which obey the Schroedinger equation:

$$H_0\psi_j(x) = \left(-\frac{\hbar^2}{2m}\nabla^2 + V\right)\psi_j(x) = E_j\psi_j(x)$$
(252)

In analogy to the quantization of the light field one can write:

$$\Psi(x) = \sum_{j} b_j^+ \psi_j(x) \tag{253}$$

with the *fermionic* creation operator b_i^+ .

The anti-commutation relation of the fermionic creation and annihilation operator are:

$$\{b_i, b_j\} = \{b_i^+, b_j^+\} = 0 \tag{254}$$

$$\left\{b_i, b_j^+\right\} = 1\tag{255}$$

An arbitrary state can thus be constructed by applying b_j^+ operators to the vacuum:

$$|\{j\}\rangle = b_{j1}^+ b_{j2}^+ \dots b_{jn}^+ |0\rangle \tag{256}$$

Due to the fermionic nature:

$$(b_j^+)^2 |0\rangle = 0$$
 or more general $(b_j^+)^2 |\varphi\rangle = 0$ (257)

The expectation value for the atomic Hamiltonian H_0

$$H_0 = \sum_j b_j^+ b_j E_j \tag{258}$$

is

$$\langle \psi | H_0 | \psi \rangle = \sum_j E_j \tag{259}$$

Since a lot of problems in quantum optics deal with the simplified case of *two-level* atoms it is convenient to limit the atomic Hilbert space to two dimensions and to introduce the Pauli spin operators $\sigma_j \in H^{\oplus 2}$ (similar as in a single spin system):

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}; \ \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}; \ \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$
(260)

Together with the raising and lowering operators

$$\sigma^{+} = \frac{1}{2} \left(\sigma_x + i \sigma_y \right); \quad \sigma^{-} = \frac{1}{2} \left(\sigma_x - i \sigma_y \right)$$
(261)

The latter operators have the following properties:

$$\left[\sigma^{+},\sigma^{-}\right] = 2\sigma_{z}; \quad \left[\sigma^{\pm},\sigma_{z}\right] = \mp\sigma^{\pm}; \quad \left\{\sigma^{+},\sigma^{-}\right\} = 1 \tag{262}$$

7.2 Bloch representation

If we assume a two-level system of two atomic states $|1\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$ and $|2\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$ then the following correspondence holds:

$$\begin{array}{ccc} \text{pseudo-spin operators} & \text{electron operators} \\ \sigma^+ & b_1^+ b_2 & |1\rangle \langle 2| \\ \sigma^- & b_2^+ b_1 & |2\rangle \langle 1| \end{array}$$

Any state of the two-level atom can be written as:

$$|\psi\rangle = c_1 |1\rangle + c_2 |2\rangle$$
 with $|c_1|^2 + |c_2|^2 = 1$ (263)

More generally (non-pure states) one has to write down the density operator ρ :

$$\rho^{(A)} = \rho_{11} |1\rangle \langle 1| + \rho_{22} |2\rangle \langle 2| + \rho_{12} |1\rangle \langle 2| + \rho_{21} |2\rangle \langle 1|$$
(264)

where
$$\rho_{ij} = \left\langle c_i c_j^* \right\rangle \quad i, j = 1, 2$$
(265)

 ρ has a representation in terms of a two-dimensional Hermitian covariant matrix.

The Bloch-representation has a very intuitive geometrical representation of the state.

Definition of the Bloch-vector \overrightarrow{r} :

$$r_1 = 2 \operatorname{Re}(\rho_{12})$$
 (266)

$$r_2 = 2 \operatorname{Im}(\rho_{12}) \tag{267}$$

$$r_3 = \rho_{22} - \rho_{11} \tag{268}$$

Therefore:

$$|1\rangle \triangleq (0, 0, -1) \tag{269}$$

$$|2\rangle \triangleq (0,0,1) \tag{270}$$

The Bloch-vector for a pure state lies on a sphere of radius |r| = 1.

Generally, it follows:

$$r_1^2 + r_2^2 + r_3^2 = 4 \left| \rho_{12} \right|^2 + \left| \rho_{22} - \rho_{11} \right|^2$$
(271)

$$= 1 - 4 \left(\rho_{22} \rho_{11} - \left| \rho_{12} \right|^2 \right) \tag{272}$$

from the Cauchy-Schwartz inequality on finds:

$$\rho_{22}\rho_{11} - |\rho_{12}|^2 = \left\langle |c_2|^2 \right\rangle \left\langle |c_1|^2 \right\rangle - \left| \left\langle c_1 c_2^* \right\rangle \right|^2 \ge 0$$
(273)

and thus

$$r_1^2 + r_2^2 + r_3^2 \le 1 \tag{274}$$



Figure 47: Bloch representation of a state of a two-level atom [from Mandel "Optical Coherence and Quantum Optics"]

7.3 Interaction of a two-level atom with a classical field

The interaction of a classical field E(t) with an atom can be described via the dipole interaction:

$$H_I = -\overrightarrow{\mu}(t) \cdot \overrightarrow{E}(t) \tag{275}$$

The time evolution of the density matrix $\rho(t)$ describing the state of the atom (Hamiltonian $H_A = \frac{1}{2}\hbar\omega_0\sigma_z$) follows from the Schroedinger equation with the Hamiltonian $H = H_A + H_I$:

$$\frac{\partial \rho(t)}{\partial t} = \frac{1}{i\hbar} \left[H_A + H_I, \rho(t) \right] \tag{276}$$

The general form of this equation of motion is:

$$\rho_{11} = \frac{1}{i\hbar} \left[\langle 1 | H_I | 2 \rangle \, \rho_{21} - c.c \right] \tag{277}$$

$$\dot{\rho}_{22} = -\frac{1}{i\hbar} \left[\langle 1 | H_I | 2 \rangle \, \rho_{21} - c.c \right] \tag{278}$$

$$\dot{\rho}_{12} = \frac{1}{i\hbar} \left[-\hbar\omega_0 \rho_{12} + \langle 1 | H_I | 2 \rangle \left(\rho_{22} - \rho_{11} \right) \right]$$
(279)

$$\dot{\rho}_{21} = \frac{1}{i\hbar} \left[\hbar \omega_0 \rho_{21} + \langle 2 | H_I | 1 \rangle \left(\rho_{11} - \rho_{22} \right) \right]$$
(280)

Obviously $(\dot{\rho}_{11} + \dot{\rho}_{22}) = 0.$

Remark: The link to classical or semiclassical physics is via the polarisation

$$P = \langle 1 | H_I | 2 \rangle \rho_{12} + c.c.$$
 (281)

These equations of motions can be expressed by the Bloch vector and are called $Bloch \ equations$:

$$\dot{r}_{1} = \frac{1}{\hbar} 2 \operatorname{Im} \left[\langle 1 | H_{I} | 2 \rangle \right] r_{3} - \omega_{0} r_{2}$$
(282)

$$\dot{r}_2 = -\frac{1}{\hbar} 2 \operatorname{Re} \left[\langle 1 | H_I | 2 \rangle \right] r_3 + \omega_0 r_1$$
(283)

$$\dot{r}_3 = -\frac{2}{\hbar} \operatorname{Im} \left[\langle 1 | H_I | 2 \rangle \right] r_1 + \frac{2}{\hbar} \operatorname{Re} \left[\langle 1 | H_I | 2 \rangle \right] r_2$$
 (284)

Obviously $d/dt(r_1^2 + r_2^2 + r_3^2) = 0!$

The motion of the Bloch vector can be described as a (complicated) precession around a vector Q(t):

$$\frac{d}{dt}\overrightarrow{r} = Q \times \overrightarrow{r} \tag{285}$$

with

$$Q = \begin{pmatrix} \frac{2}{\hbar} \operatorname{Re} \langle 1 | H_I | 2 \rangle \\ \frac{2}{\hbar} \operatorname{Im} \langle 1 | H_I | 2 \rangle \\ \omega_0 \end{pmatrix}$$
(286)

If the interaction with a classical single-mode field $E(t) = \hat{\epsilon} E_0(t) \exp(-i\omega_1 t) + c.c.$ is evaluated then the term $\langle 1|H_I|2 \rangle$ becomes:

$$\langle 1 | H_I | 2 \rangle = -\overrightarrow{\mu}_{12} \overrightarrow{E}(t) = - \langle 1 | \overrightarrow{\mu} | 2 \rangle \overrightarrow{E}(t)$$
(287)

The fast rotation of the Bloch-vector around the z-axis at the optical frequency ω_0 can be eliminated by transforming into a rotating frame:

$$\overrightarrow{r}' = \Theta \cdot \overrightarrow{r} \tag{288}$$

with

$$\Theta = \begin{pmatrix} \cos \omega_1 t & \sin \omega_1 t & 0\\ -\sin \omega_1 t & \cos \omega_1 t & 0\\ 0 & 0 & 1 \end{pmatrix}$$
(289)

This leads to the Bloch equations in the rotating frame:

$$\dot{r}_{1}' = (\omega_{1} - \omega_{0})r_{2}'$$
(290)

$$\dot{r}_{2}' = (\omega_{0} - \omega_{1})r_{1}' + \Omega r_{3}'$$
(291)

$$\dot{r}_3 = -\Omega r_2' \tag{292}$$

with the *Rabi frequency* Ω :

$$\Omega = 2\overrightarrow{\mu}_{12}\widehat{\epsilon} \left| E_0(t) \right| /\hbar \tag{293}$$

One can also write

$$\overrightarrow{r'} = Q' \times \overrightarrow{r'} \quad \text{with} \quad Q' = \begin{pmatrix} -\Omega \\ 0 \\ \omega_0 - \omega_1 \end{pmatrix}$$
(294)



Figure 48: Precession of Bloch vector vor $\delta = 0$ (a) and $\delta \neq 0$ (b) [from Scully "Quantum Optics"]

7.4 Ramsey fringes

If the field is in resonance with the atomic transition $(\omega_1 - \omega_0) = 0$ then it is:

$$r_1'(t) = 0 (295)$$

$$r_2'(t) = -\sin\Omega t \tag{296}$$

$$r_3'(t) = \cos\Omega t \tag{297}$$

A pulse which is applied to the atom initially in the ground state (r = (0, 0, 1)) which has the pulse area $\Omega t = \pi$, a so-called π -pulse, flips the atomic state to the excited state, whereas a pulse with area $\Omega t = \pi/2$, a $\pi/2$ -pulse, creates a coherent superposition of upper and lower atomic state of equal weight:

$$\Omega t = \pi \qquad \pi\text{-pulse} \qquad |2\rangle \longrightarrow |1\rangle \\ \Omega t = \pi/2 \qquad \pi/2\text{-pulse} \qquad |2\rangle \longrightarrow (|2\rangle + |1\rangle)/\sqrt{2}$$
(298)

A small detuning $\delta = \omega_1 - \omega_0$ leads to a rotation of the Bloch vector in the x-y-plane if there is a non-zero component of r_1 or r_2 .

A method to exploit this effect in order to perform precise measurements of a frequency ω was proposed by Ramsey, who was awarded the Nobel prize for this idea in 1989:

- First a $\pi/2$ -pulse is applied to an atom, which is initially in the ground state. This flips the Bloch vector into the x-y-plane.
- If there is no detuning (e.-mag. field in exact resonance with the atomic transition) then a second $\pi/2$ -pulse after some time T flips the Bloch vector exactly to the excited state, which can then be detected.
- If, however, there is some detuning then the Bloch vector rotates in the x-yplane by an angle $\delta \cdot T$. A second $\pi/2$ -pulse would then usually not tilt the Bloch vector exactly to the excited state (in the extreme case the Bloch vector may even be tilted back to the ground state).

This method can be used to compare the frequency of a field to an atomic transition and is called *Ramsey-method*. By changing T or δ the probability to detect the atom in the excited state oscillates. These oscillations are also called *Ramsey fringes*.

In a Ramsey interferometer the two pulses have to be separated in time as far as possible to obtain highest sensitivity. The sensitivity is not limited by the time-of-flight of the atom through a single interaction zone in the experiment.

Modern atomic clocks (e.g. Cs clocks) use the Ramsey method to stabilze an RF-field to a narrow atomic transition.



Figure 49: Principle setup for a Ramsey measurement.



Figure 50: Ramsey fringes

7.5 Rabi-Oscillation for pure states

Rabi-Oscillations are the typical dynamics for the coherent interaction of two-level atoms with light. In the following some aspects are described more explicitly for the case of pure states.

A general state of a two-level atom is of the form:

$$|\psi\rangle = c_e|e\rangle + c_q|g\rangle \tag{299}$$

We are interested in the time evolution of the coefficienty $c_e(t)$ and $c_g(t)$.

Following equation (265) for the density matrix it is possible to write:

$$\rho = \rho_{ee} \left| e \right\rangle \left\langle e \right| + \rho_{gg} \left| g \right\rangle \left\langle g \right| + \rho_{eg} \left| e \right\rangle \left\langle g \right| + \rho_{ge} \left| g \right\rangle \left\langle e \right| \tag{300}$$

where
$$\rho_{ij} = c_i c_j^*$$
 $i, j = e, g$ (301)

where we replaced the previously used subscript 1 and 2 by e and g, respectively, for clarity.

7.5.1 Resonant interaction

In the special case of *exact resonance* between the frequency of the light and the atomic transition frequency, i.e. $\delta = \omega_1 - \omega_0 = 0$, it is straightforward to derive an exact solution from the equation of motion (280) derived above:

$$c_e(t) = c_e(0) \cos\left(\frac{1}{2}\Omega t\right) - ic_g(0) \sin\left(\frac{1}{2}\Omega t\right)$$
(302)

$$c_g(t) = c_g(0) \cos\left(\frac{1}{2}\Omega t\right) - ic_e(0) \sin\left(\frac{1}{2}\Omega t\right)$$
(303)

with the Rabi-frequency $\Omega = 2 < e|H_I|g > /\hbar$.

With the special initial condition $c_e(0) = 0$, $c_g(0) = 1$ one finds:

$$c_e(t) = -i\sin\left(\frac{1}{2}\Omega t\right) \tag{304}$$

$$c_g(t) = \cos\left(\frac{1}{2}\Omega t\right) \tag{305}$$

and for the probability $P_g(t)$ and $P_e(t)$ to find the atom in the ground and excited state, respectively:

$$P_e(t) = |c_e(t)|^2 = \frac{1}{2} \left(1 - \cos \Omega t\right)$$
(306)

$$P_g(t) = |c_g(t)|^2 = \frac{1}{2} \left(1 + \cos \Omega t\right)$$
(307)

Figure 51 plots the time evolution which corresponds to the pictorial dynamics of the Bloch vector as described above.



Figure 51: Time evolution of the probability $P_g(t)$ and $P_e(t)$ to find the atom in the ground (solid) and excited (dashed) state, respectively. [from D.A. Steck *Quantum and Atom Optics*]

7.5.2 Near-resonant interaction

A more lengthy calculation provides an analytical solution for the case of *near-resonance* with $\delta \neq 0$. In the following the generalized Rabi-frequency $\tilde{\Omega}$ was introduced:

$$\tilde{\Omega} = \sqrt{\Omega^2 + (\omega_1 - \omega_0)^2} = \sqrt{\Omega^2 + \delta^2}$$
(308)

With this the time evolutions are:

$$c_e(t) = e^{i\delta t/2} \left[c_e(0) \cos\left(\frac{1}{2}\tilde{\Omega}t\right) + \frac{i}{\tilde{\Omega}} \left[\delta c_e(0) - \Omega c_g(0) \right] \sin\left(\frac{1}{2}\tilde{\Omega}t\right) \right]$$
(309)

$$c_g(t) = e^{i\delta t/2} \left[c_g(0) \cos\left(\frac{1}{2}\tilde{\Omega}t\right) - \frac{i}{\tilde{\Omega}} \left[\delta c_g(0) + \Omega c_e(0)\right] \sin\left(\frac{1}{2}\tilde{\Omega}t\right) \right]$$
(310)

Again, with the special initial condition $c_e(0) = 0$, $c_g(0) = 1$ one finds:

$$c_e(t) = -ie^{i\delta t/2} \frac{\Omega}{\tilde{\Omega}} \sin\left(\frac{1}{2}\tilde{\Omega}t\right)$$
(311)

$$c_g(t) = e^{i\delta t/2} \left[\cos\left(\frac{1}{2}\tilde{\Omega}t\right) - i\frac{\delta}{\tilde{\Omega}}\sin\left(\frac{1}{2}\tilde{\Omega}t\right) \right]$$
(312)

and for the probability $P_e(t)$ to find the atom in the excited state:

$$P_e(t) = \frac{\Omega^2}{\tilde{\Omega}^2} \left(\frac{1}{2} - \frac{1}{2} \cos \tilde{\Omega} t \right)$$
(313)

Figure 52 plots the time evolution of $P_e(t)$ for different detunings. For non-vanishing detuning the state will never be fully excited. This corresponds to a rotation of the Bloch vector on a non-maximum circle.



Figure 52: Time evolution of the probability $P_e(t)$ to find the atom in the excited state for different detunings Δ . [from D.A. Steck *Quantum and Atom Optics*]

7.6 Phenomenological treatment of damping

In the previous chapter the Bloch-vector was introduced. We pointed out that the Bloch-vector under *coherent* time evolution always has unity length. In case of *incoherent* evolution this is no longer the case.

7.6.1 Generalized Bloch-equation

How can we describe an incoherent evolution, e.g. damping? The starting points are the Bloch-equation as provided in equation 280

$$\dot{\rho}_{11} = \frac{1}{i\hbar} \left[\langle 1 | H_I | 2 \rangle \, \rho_{21} - c.c \right] \tag{314}$$

$$\dot{\rho}_{22} = -\frac{1}{i\hbar} \left[\langle 1 | H_I | 2 \rangle \rho_{21} - c.c \right]$$
(315)

$$\dot{\rho}_{12} = \frac{1}{i\hbar} \left[-\hbar\omega_0 \rho_{12} + \langle 1 | H_I | 2 \rangle \left(\rho_{22} - \rho_{11} \right) \right]$$
(316)

$$\dot{\rho}_{21} = \frac{1}{i\hbar} \left[\hbar \omega_0 \rho_{21} + \langle 2 | H_I | 1 \rangle \left(\rho_{11} - \rho_{22} \right) \right]$$
(317)

We will now introduce damping terms in a phenomenological way:

- Γ as a damping rate of the population (diagonal elements of the density matrix)
- γ_⊥ as a damping rate of the coherence (off-diagonal elements of the density matrix)
 The rate Γ also damps the off-diagonal elements. In order to include more

general cases we introduce an additional rate γ_c which describes pure dephasing (e.g. due to collisions of two-level atoms in a gas). Therefore:

$$\gamma_{\perp} = \Gamma/2 + \gamma_c \tag{318}$$

With the damping rates we can formulate the generalized version of the **optical Bloch equation** including damping (in the interaction picture):

$$\dot{\rho}_{ee} = i\frac{\Omega}{2} \left(\rho_{eg} - \rho_{ge}\right) - \Gamma \rho_{ee} \tag{319}$$

$$\dot{\rho}_{gg} = -i\frac{\Omega}{2} \left(\rho_{eg} - \rho_{ge}\right) + \Gamma \rho_{ee} \tag{320}$$

$$\dot{\rho}_{ge} = -\left(\gamma_{\perp} + i\delta\right)\rho_{ge} - i\frac{\Omega}{2}\left(\rho_{ee} - \rho_{gg}\right) \tag{321}$$

$$\dot{\rho}_{eg} = -\left(\gamma_{\perp} - i\delta\right)\rho_{eg} + i\frac{\Omega}{2}\left(\rho_{ee} - \rho_{gg}\right)$$
(322)

where we have again introduced the Rabi-frequency $\Omega = 2 \langle 1 | H_I | 2 \rangle / \hbar$.

In a similar way the equations of motion for the Bloch-vector in the interaction picture (see (290) to (292)) can be generalized as:

$$\dot{r}_1' = \delta r_2' - \gamma_\perp r_1' \tag{323}$$

$$\dot{r}_{2}' = -\delta r_{1}' + \Omega r_{3}' - \gamma_{\perp} r_{2}'$$
(324)

$$\dot{r}'_{3} = -\Omega r'_{2} - \Gamma \left(r'_{3} + 1 \right)$$
 (325)

with $\delta = \omega_1 - \omega_0$

7.6.2 Steady-state-solution of the Bloch-equation with damping

It is straightforward to derive the steady-state solution of the Bloch-equation by setting the time derivatives to zero.

As a result for the population of excited state ρ_{ee} we find:

$$\rho_{ee} = \frac{\Omega^2}{2\Gamma\gamma_{\perp}} \frac{1}{1 + \frac{\delta^2}{\gamma_{\perp}^2} + \frac{\Omega^2}{\Gamma\gamma_{\perp}}}$$
(326)

And for the steady state coherence ρ_{eq} :

$$\rho_{eg} = -i \frac{\Omega}{2\gamma_{\perp}} \frac{1 + \frac{i\delta}{\gamma_{\perp}}}{1 + \frac{\delta^2}{\gamma_{\perp}^2} + \frac{\Omega^2}{\Gamma\gamma_{\perp}}}$$
(327)

Often the so-called *saturation parameter* S is introduced to simplify the notation.

$$S = \frac{\Omega^2 / \Gamma \gamma_\perp}{1 + \delta^2 / \gamma_\perp^2} \tag{328}$$

With this parameter the expression for the steady-state values of ρ_{ee} and $|\rho_{eg}|^2$ are:

$$\rho_{ee} = \frac{S/2}{1+S} \tag{329}$$

$$|\rho_{eg}|^2 = \frac{\Gamma}{4\gamma_{\perp}} \frac{S}{(1+S)^2}$$
(330)

Finally, we give the analytic form for the time evolution of the excited state probability $\rho_{ee}(t)$ for the special case $\delta = 0$, $\gamma_{\perp} = \Gamma/2$ and the atom initially in the ground state.

$$\rho_{ee}(t) = -1 + \frac{\Omega^2}{\Omega^2 + \Gamma^2/2} \left[1 - e^{-(3\Gamma/4)t} \left(\cos \Omega_{\Gamma} t + \frac{\Omega^2 - \Gamma^2/4}{\Gamma \Omega_{\Gamma}} \sin \Omega_{\Gamma} t \right) \right]$$
(331)
where $\Omega_{\Gamma} = \sqrt{\Omega^2 + (\Gamma/4)^2}$

with $\Omega_{\Gamma} = \sqrt{\Omega^2 + (\Gamma/4)^2}$

Note that after some initial oscillation the atomic inversion will relax to a value below 0, i.e. the population in the upper state is always less than the population in the lower state. The following figure plots time evolutions for different values of the damping Γ :



Figure 53: Time evolution of the probability $\rho_{ee}(t)$ to find the atom in the excited state for different damping rates Γ . [from D.A. Steck *Quantum and Atom Optics*]