# Chapter 2

# **Maxwell-Bloch Equations**

## 2.1 Maxwell's Equations

Maxwell's equations are given by

$$\vec{\nabla} \times \vec{H} = \vec{j} + \frac{\partial \vec{D}}{\partial t},$$
 (2.1a)

$$\vec{\nabla} \times \vec{E} = -\frac{\partial B}{\partial t},$$
 (2.1b)

$$\vec{\nabla} \cdot \vec{D} = \rho, \qquad (2.1c)$$

$$\vec{\nabla} \cdot \vec{B} = 0. \tag{2.1d}$$

The material equations accompanying Maxwell's equations are:

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P}, \qquad (2.2a)$$

$$\vec{B} = \mu_0 \vec{H} + \vec{M}.$$
 (2.2b)

Here,  $\vec{E}$  and  $\vec{H}$  are the electric and magnetic field,  $\vec{D}$  the dielectric flux,  $\vec{B}$  the magnetic flux,  $\vec{j}$  the current density of free carriers,  $\rho$  is the free charge density,  $\vec{P}$  is the polarization, and  $\vec{M}$  the magnetization. By taking the curl of Eq. (2.1b) and considering  $\vec{\nabla} \times (\vec{\nabla} \times \vec{E}) = \vec{\nabla} (\vec{\nabla} \vec{E}) - \Delta \vec{E}$ , we obtain

$$\Delta \vec{E} - \mu_0 \frac{\partial}{\partial t} \left( \vec{j} + \epsilon_0 \frac{\partial \vec{E}}{\partial t} + \frac{\partial \vec{P}}{\partial t} \right) = \frac{\partial}{\partial t} \vec{\nabla} \times \vec{M} + \vec{\nabla} \left( \vec{\nabla} \cdot \vec{E} \right)$$
(2.3)

and hence

$$\left(\Delta - \frac{1}{c_0^2} \frac{\partial^2}{\partial t^2}\right) \vec{E} = \mu_0 \left(\frac{\partial \vec{j}}{\partial t} + \frac{\partial^2}{\partial t^2} \vec{P}\right) + \frac{\partial}{\partial t} \vec{\nabla} \times \vec{M} + \vec{\nabla} \left(\vec{\nabla} \cdot \vec{E}\right).$$
(2.4)

The vacuum velocity of light is

$$c_0 = \sqrt{\frac{1}{\mu_0 \epsilon_0}}.$$
(2.5)

## 2.2 Linear Pulse Propagation in Isotropic Media

For dielectric non magnetic media, with no free charges and currents due to free charges, there is  $\vec{M} = \vec{0}$ ,  $\vec{j} = \vec{0}$ ,  $\rho = 0$ . We obtain with  $\vec{D} = \epsilon(\vec{r}) \vec{E} = \epsilon_0 \epsilon_r(\vec{r}) \vec{E}$ 

$$\vec{\nabla} \cdot (\epsilon \left( \vec{r} \right) \vec{E}) = 0. \tag{2.6}$$

In addition for homogeneous media, we obtain  $\vec{\nabla} \cdot \vec{E} = 0$  and the wave equation (2.4) greatly simplifies

$$\left(\Delta - \frac{1}{c_0^2} \frac{\partial^2}{\partial t^2}\right) \vec{E} = \mu_0 \frac{\partial^2}{\partial t^2} \vec{P}.$$
(2.7)

This is the wave equation driven by the polarization in the medium. If the medium is linear and has only an induced polarization described by the susceptibility  $\chi(\omega) = \epsilon_r(\omega) - 1$ , we obtain in the frequency domain

$$\widehat{\vec{P}}(\omega) = \epsilon_0 \chi(\omega) \widehat{\vec{E}}(\omega).$$
(2.8)

Substituted in (2.7)

$$\left(\Delta + \frac{\omega^2}{c_0^2}\right)\hat{\vec{E}}(\omega) = -\omega^2 \mu_0 \epsilon_0 \chi(\omega)\hat{\vec{E}}(\omega), \qquad (2.9)$$

where  $\hat{\vec{D}} = \epsilon_0 \epsilon_r(\omega) \hat{\vec{E}}(\omega)$ , and thus

$$\left(\Delta + \frac{\omega^2}{c_0^2} (1 + \chi(\omega)) \hat{\vec{E}}(\omega) = 0, \qquad (2.10)$$

22

with the refractive index n and  $1+\chi(\omega)=n^2$ 

$$\left(\Delta + \frac{\omega^2}{c^2}\right)\hat{\vec{E}}(\omega) = 0, \qquad (2.11)$$

where  $c = c_0/n$  is the velocity of light in the medium.

## 2.2.1 Plane-Wave Solutions (TEM-Waves)

The complex plane-wave solution of Eq. (2.11) is given by

$$\hat{\vec{E}}^{(+)}(\omega,\vec{r}) = \hat{\vec{E}}^{(+)}(\omega)e^{-j\vec{k}\cdot\vec{r}} = E_0e^{-j\vec{k}\cdot\vec{r}}\cdot\vec{e}$$
(2.12)

with

$$\vec{k}|^2 = \frac{\omega^2}{c^2} = k^2.$$
 (2.13)

Thus, the dispersion relation is given by

$$k(\omega) = \frac{\omega}{c_0} n(\omega). \tag{2.14}$$

From  $\vec{\nabla} \cdot \vec{E} = 0$ , we see that  $\vec{k} \perp \vec{e}$ . In time domain, we obtain

$$\vec{E}^{(+)}(\vec{r},t) = E_0 \vec{e} \cdot e^{j\omega t - j\vec{k}\cdot\vec{r}}$$
(2.15)

with

$$k = 2\pi/\lambda, \tag{2.16}$$

where  $\lambda$  is the wavelength,  $\omega$  the angular frequency,  $\vec{k}$  the wave vector,  $\vec{e}$  the polarization vector, and  $f = \omega/2\pi$  the frequency. From Eq. (2.1b), we get for the magnetic field

$$-\mathbf{j}\vec{k} \times E_0 \vec{e} e^{\mathbf{j}(\omega t - \vec{k}\vec{r})} = -\mathbf{j}\mu_0 \omega \vec{H}^{(+)}, \qquad (2.17)$$

or

$$\vec{H}^{(+)} = \frac{E_0}{\mu_0 \omega} e^{j(\omega t - \vec{k}\vec{r})} \vec{k} \times \vec{e} = H_0 \vec{h} e^{j(\omega t - \vec{k}\vec{r})}$$
(2.18)

with

$$\vec{h} = \frac{\vec{k}}{|k|} \times \vec{e} \tag{2.19}$$

and

$$H_0 = \frac{|k|}{\mu_0 \omega} E_0 = \frac{1}{Z_F} E_0.$$
 (2.20)

The natural impedance is

$$Z_F = \mu_0 c = \sqrt{\frac{\mu_0}{\epsilon_0 \epsilon_r}} = \frac{1}{n} Z_{F_0}$$
(2.21)

with the free space impedance

$$Z_{F_0} = \sqrt{\frac{\mu_0}{\epsilon_0}} = 377 \,\Omega.$$
 (2.22)

For a backward propagating wave with  $\vec{E}^{(+)}(\vec{r},t) = E_0 \vec{e} \cdot e^{j\omega t + j\vec{k}\cdot\vec{r}}$  there is  $\vec{H}^{(+)} = H_0 \vec{h} e^{j(\omega t - \vec{k}\vec{r})}$  with

$$H_0 = -\frac{|k|}{\mu_0 \omega} E_0.$$
 (2.23)

Note that the vectors  $\vec{e}$ ,  $\vec{h}$  and  $\vec{k}$  form an orthogonal trihedral,

$$\vec{e} \perp \vec{h}, \quad \vec{k} \perp \vec{e}, \quad \vec{k} \perp \vec{h}.$$
 (2.24)

#### 2.2.2 Complex Notations

Physical  $\vec{E}$ ,  $\vec{H}$  fields are real:

$$\vec{E}(\vec{r},t) = \frac{1}{2} \left( \vec{E}^{(+)}(\vec{r},t) + \vec{E}^{(-)}(\vec{r},t) \right)$$
(2.25)

with  $\vec{E}^{(-)}(\vec{r},t) = \vec{E}^{(+)}(\vec{r},t)^*$ . A general temporal shape can be obtained by adding different spectral components,

$$\vec{E}^{(+)}(\vec{r},t) = \int_0^\infty \frac{d\omega}{2\pi} \hat{\vec{E}}^{(+)}(\omega) e^{\mathbf{j}(\omega t - \vec{k} \cdot \vec{r})}.$$
(2.26)

Correspondingly, the magnetic field is given by

$$\vec{H}(\vec{r},t) = \frac{1}{2} \left( \vec{H}^{(+)}(\vec{r},t) + \vec{H}^{(-)}(\vec{r},t) \right)$$
(2.27)

with  $\vec{H}^{(-)}(\vec{r},t) = \vec{H}^{(+)}(\vec{r},t)^*$ . The general solution is given by

$$\vec{H}^{(+)}(\vec{r},t) = \int_0^\infty \frac{d\omega}{2\pi} \hat{\vec{H}}^{(+)}(\omega) e^{j(\omega t - \vec{k} \cdot \vec{r})}$$
(2.28)

with

$$\widehat{\vec{H}}^{(+)}(\omega) = \frac{E_0}{Z_F} \vec{h}.$$
(2.29)

## 2.2.3 Poynting Vectors, Energy Density and Intensity for Plane Wave Fields

Quantity	Real fields	Complex fields $\langle \rangle_t$
Energy density	$w = \frac{1}{2} \left( \epsilon_0 \epsilon_r \vec{E}^2 + \mu_0 \mu_r \vec{H}^2 \right)$	$w = \frac{1}{4} \begin{pmatrix} \epsilon_0 \epsilon_r \left  \vec{E}^{(+)} \right ^2 \\ +\mu_0 \mu_r \left  \vec{H}^{(+)} \right ^2 \end{pmatrix}$
Poynting vector	$\vec{S} = \vec{E}  imes \vec{H}$	$\vec{T} = \frac{1}{2}\vec{E}^{(+)} \times \left(\vec{H}^{(+)}\right)^*$
Intensity	$I = \left  \vec{S} \right  = cw$	$I = \left  \vec{T} \right  = cw$
Energy Cons.	$\frac{\partial w}{\partial t} + \vec{\nabla} \vec{S} = 0$	$\frac{\partial w}{\partial t} + \vec{\nabla} \vec{T} = 0$

For  $\vec{E}^{(+)}(\vec{r},t) = E_0 \vec{e}_x e^{j(\omega t - kz)}$  we obtain the energy density

$$w = \frac{1}{2}\epsilon_r\epsilon_0|E_0|^2, \qquad (2.30)$$

the poynting vector

$$\vec{T} = \frac{1}{2Z_F} |E_0|^2 \vec{e_z}$$
(2.31)

and the intensity

$$I = \frac{1}{2Z_F} |E_0|^2 = \frac{1}{2} Z_F |H_0|^2.$$
(2.32)

#### 2.2.4 Dielectric Susceptibility

The polarization is given by

$$\vec{P}^{(+)}(\omega) = \frac{\text{dipole moment}}{\text{volume}} = N \cdot \langle \vec{p}^{(+)}(\omega) \rangle = \epsilon_0 \chi(\omega) \vec{E}^{(+)}(\omega), \qquad (2.33)$$

where N is density of elementary units and  $\langle \vec{p} \rangle$  is the average dipole moment of unit (atom, molecule, ...).

#### Classical harmonic oscillator model

The damped harmonic oscillator driven by an electric force in one dimension, x, is described by the differential equation

$$m\frac{d^2x}{dt^2} + 2\frac{\omega_0}{Q}m\frac{dx}{dt} + m\omega_0^2 x = e_0 E(t), \qquad (2.34)$$

where  $E(t) = \hat{E}e^{j\omega t}$ . By using the ansatz  $x(t) = \hat{x}e^{j\omega t}$ , we obtain for the complex amplitude of the dipole moment  $p = e_0 x(t) = \hat{p}e^{j\omega t}$ 

$$\hat{p} = \frac{\frac{e_0^2}{m}}{(\omega_0^2 - \omega^2) + 2j\frac{\omega_0}{Q}\omega}\hat{E}.$$
(2.35)

For the susceptibility, we get

$$\chi(\omega) = \frac{N \frac{e_0^2}{m} \frac{1}{\epsilon_0}}{(\omega_0^2 - \omega^2) + 2j\omega \frac{\omega_0}{Q}}$$
(2.36)

and thus

$$\chi(\omega) = \frac{\omega_p^2}{(\omega_0^2 - \omega^2) + 2j\omega\frac{\omega_0}{Q}},$$
(2.37)

with the plasma frequency  $\omega_p$ , determined by  $\omega_p^2 = N e_0^2 / m \epsilon_0$ . Figure 2.1 shows the real part and imaginary part of the classical susceptibility (2.37).



Figure 2.1: Real part and imaginary part of the susceptibility of the classical oscillator model for the electric polarizability.

Note, there is a small resonance shift due to the loss. Off resonance, the imaginary part approaches very quickly zero. Not so the real part, it approaches a constant value  $\omega_p^2/\omega_0^2$  below resonance, and approaches zero for above resonance, but slower than the real part, i.e. off resonance there is still a contribution to the index but practically no loss.

## 2.3 Bloch Equations

Atoms in low concentration show line spectra as found in gas-, dye- and some solid-state laser media. 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 the possible energy eigenstates into account, for example those which are related to the laser transition. The pumping of the laser can be 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 antiparallel to the field, i.e. it has two energy levels and energy eigenstates. The interaction of the two-level atom or the spin with the electric or magnetic field is described by the Bloch equations.

#### 2.3.1 The Two-Level Model

An atom having 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  $E_g$  (Fig. 2.2).



Figure 2.2: Two-level atom

In the position-, i.e. x-representation, these states correspond to the wave

functions

$$\psi_e(x) = \langle x | e \rangle$$
, and  $\psi_g(x) = \langle x | g \rangle$ . (2.38)

The Hamiltonian of the atom is given by

$$H_A = E_e |e| < e| + E_g |g| < g|.$$
(2.39)

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

$$1 = |e| < e| + |g| < g|, \qquad (2.40)$$

$$\boldsymbol{\sigma}_z = |e\rangle \langle e| - |g\rangle \langle g|, \qquad (2.41)$$

$$\boldsymbol{\sigma}^+ = |e\rangle \langle g|, \qquad (2.42)$$

$$\boldsymbol{\sigma}^{-} = |g\rangle \langle e|. \tag{2.43}$$

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

$$\boldsymbol{\sigma}_x = \boldsymbol{\sigma}^+ + \boldsymbol{\sigma}^-, \qquad (2.44)$$

$$\sigma_y = -j\sigma^+ + j\sigma^-.$$
 (2.45)

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

$$|\psi\rangle = c_g |g\rangle + c_e |e\rangle.$$
 (2.46)

We obtain

$$\boldsymbol{\sigma}^+ |\psi\rangle > = c_g |e\rangle, \tag{2.47}$$

$$\boldsymbol{\sigma}^{-}|\psi\rangle > = c_{e}|g\rangle, \qquad (2.48)$$

$$\boldsymbol{\sigma}_{z}|\psi\rangle > = c_{e}|e\rangle - c_{g}|g\rangle. \tag{2.49}$$

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

$$<\psi|\sigma_z|\psi> = |c_e|^2 - |c_g|^2 = w,$$
 (2.50)

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.

28

#### 2.3. BLOCH EQUATIONS

If we consider an ensemble of N atoms the total inversion would be  $\sigma =$  $N < \psi | \boldsymbol{\sigma}_z | \psi >$ . If we separate from the Hamiltonian (2.38) the term  $(E_e +$  $E_q)/2 \cdot \mathbf{1}$ , where **1** denotes the unity matrix, we rescale the energy values correspondingly and obtain for the Hamiltonian of the two-level system

$$\mathbf{H}_A = \frac{1}{2}\hbar\omega_{eg}\boldsymbol{\sigma}_z,\tag{2.51}$$

with the transition frequency

$$\omega_{eg} = \frac{1}{\hbar} (E_e - E_g). \tag{2.52}$$

This form of the Hamiltonian is favorable. There are the following commutator relations between operators (2.41) to (2.43)

$$[\boldsymbol{\sigma}^+, \boldsymbol{\sigma}^-] = \boldsymbol{\sigma}_z, \qquad (2.53)$$

$$[\boldsymbol{\sigma}^+, \boldsymbol{\sigma}_z] = -2\boldsymbol{\sigma}^+, \qquad (2.54)$$

$$[\boldsymbol{\sigma}^{-}, \boldsymbol{\sigma}_{z}] = 2\boldsymbol{\sigma}^{-}, \qquad (2.55)$$

and anti-commutator relations, respectively

$$[\boldsymbol{\sigma}^+, \boldsymbol{\sigma}^-]_+ = \mathbf{1}, \qquad (2.56)$$

$$[\sigma^{+}, \sigma_{z}]_{+} = 0, \qquad (2.57)$$
  
$$[\sigma^{-}, \sigma_{z}]_{+} = 0, \qquad (2.58)$$

$$[\boldsymbol{\sigma}^{-}, \boldsymbol{\sigma}_{z}]_{+} = \mathbf{0}, \qquad (2.58)$$

$$[\sigma^{-}, \sigma^{-}]_{+} = [\sigma^{+}, \sigma^{+}]_{+} = \mathbf{0}.$$
 (2.59)

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

$$[\boldsymbol{\sigma}_x, \boldsymbol{\sigma}_y] = 2j\boldsymbol{\sigma}_z, \qquad (2.60)$$

$$[\boldsymbol{\sigma}_y, \boldsymbol{\sigma}_z] = 2j\boldsymbol{\sigma}_x, \qquad (2.61)$$

$$[\boldsymbol{\sigma}_z, \boldsymbol{\sigma}_x] = 2j\boldsymbol{\sigma}_y. \tag{2.62}$$

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

$$|\psi\rangle = c_e|e\rangle + c_g|g\rangle \rightarrow \begin{pmatrix} c_e \\ c_g \end{pmatrix}.$$
 (2.63)

The operators are then represented by matrices

$$\sigma^+ \rightarrow \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix},$$
 (2.64)

$$\sigma^- \rightarrow \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix},$$
 (2.65)

$$\sigma_z \rightarrow \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix},$$
 (2.66)

$$\mathbf{1} \quad \rightarrow \quad \left(\begin{array}{cc} 1 & 0\\ 0 & 1 \end{array}\right). \tag{2.67}$$

## 2.3.2 The Atom-Field Interaction In Dipole Approximation

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

$$\vec{\mathbf{p}} = -e_0 \, \vec{\mathbf{x}}.\tag{2.68}$$

Then the expectation value for the dipole moment of an atom in state (2.46) is

$$<\psi|\mathbf{\vec{p}}|\psi> = -e_0(|c_e|^2 < e|\mathbf{\vec{x}}|e> + c_e c_g^* < g|\mathbf{\vec{x}}|e> (2.69) + c_g c_e^* < e|\mathbf{\vec{x}}|g> + |c_g|^2 < g|\mathbf{\vec{x}}|g>).$$

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

$$<\psi|\mathbf{\vec{p}}|\psi> = -e_0 (c_e c_g^* < g|\mathbf{\vec{x}}|e> + c_g c_e^* < g|\mathbf{\vec{x}}|e>^*).$$
 (2.70)

(Note, this means, there is no permanent dipole moment in an atom, which is in an energy eigenstate. Note, 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. If so, there are also crystal fields, which then imply level shifts, via the linear Stark effect.) Thus an atom does only exhibit a dipole moment in the average, 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  $|g \rangle$ .

#### 2.3. BLOCH EQUATIONS

With the dipole matrix elements

$$\vec{M} = e_0 < g |\vec{\mathbf{x}}| e > \tag{2.71}$$

the expectation value for the dipole moment can be written as

$$<\psi|\vec{\mathbf{p}}|\psi> = -(c_e c_g^* \vec{M} + c_g c_e^* \vec{M}^*) = -<\psi|(\sigma^+ \vec{M}^* + \sigma^- \vec{M})|\psi>.$$
 (2.72)

Since this is true for an arbitrary state, the dipole operator (2.68) is represented by

$$\vec{\mathbf{p}} = \vec{\mathbf{p}}^+ + \vec{\mathbf{p}}^- = -\vec{M}^* \boldsymbol{\sigma}^+ - \vec{M} \boldsymbol{\sigma}^-.$$
(2.73)

Therefore, the operators  $\sigma^+$  and  $\sigma^-$  are proportional to the complex dipole moment operators  $\vec{\mathbf{p}}^+$  and  $\vec{\mathbf{p}}^-$ , respectively.

The energy of an electric dipole in an electric field is

$$\mathbf{H}_{A-F} = -\vec{\mathbf{p}} \cdot \vec{E}(\vec{x}_A, t). \tag{2.74}$$

The electric field at the position of the atom,  $\vec{x}_A$ , can be written as

$$\vec{E}(\vec{x}_A, t) = \frac{1}{2} \left( \vec{E}(t)^{(+)} + \vec{E}(t)^{(-)} \right) = \frac{1}{2} \left( \hat{\vec{E}}(t)^{(+)} e^{j\omega t} + \hat{\vec{E}}(t)^{(-)} e^{-j\omega t} \right), \quad (2.75)$$

where  $\hat{\vec{E}}(t)^{(+)}$  denotes the slowly varying complex field envelope with  $\omega \approx \omega_{eg}$ . In the Rotating-Wave Approximation (RWA), we only keep the slowly varying components in the interaction Hamiltonian. As we will see later, if there is no field the operator  $\sigma^+$  evolves like  $\sigma^+(t) = \sigma^+(0)e^{j\omega_{eg}t}$ , thus we obtain in RWA

$$\mathbf{H}_{A-F} = -\vec{\mathbf{p}} \cdot \vec{E}(\vec{x}_A, t) \approx \tag{2.76}$$

$$\approx \mathbf{H}_{A-F}^{RWA} = \frac{1}{2}\vec{M}^*\vec{E}(t)^{(-)}\boldsymbol{\sigma}^+ + h.c..$$
 (2.77)

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

$$j\hbar \frac{d}{dt}|\psi\rangle > = (\mathbf{H}_A + \mathbf{H}_{A-F})|\psi\rangle$$
(2.78)

$$\approx (\mathbf{H}_A + \mathbf{H}_{A-F}^{RWA}) | \psi > . \tag{2.79}$$

Written in the energy representation, we obtain

$$\frac{d}{dt}c_e = -j\frac{\omega_{eg}}{2}c_e - j\Omega_r e^{-j\omega t}c_g, \qquad (2.80)$$

$$\frac{d}{dt}c_g = +j\frac{\omega_{eg}}{2}c_g - j\Omega_r e^{+j\omega t}c_e, \qquad (2.81)$$

with the Rabi-frequency defined as

$$\Omega_r = \frac{\vec{M}^* \hat{\vec{E}}}{2\hbar}.$$
(2.82)

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

#### 2.3.3 Rabi-Oscillations

If the incident light has a constant field amplitude  $\hat{\vec{E}}$  Eqs. (2.80) and (2.81) 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

$$\Delta = \frac{\omega_{ab} - \omega}{2} \tag{2.83}$$

and the new probability amplitudes

$$C_e = c_e e^{j\frac{\omega}{2}t}, \qquad (2.84)$$

$$C_g = c_g e^{-j\frac{\omega}{2}t}.$$
 (2.85)

This leads to the new system of equations with constant coefficients

$$\frac{d}{dt}C_e = -j\Delta C_e - j\Omega_r C_g, \qquad (2.86)$$

$$\frac{d}{dt}C_g = +j\Delta C_g - j\Omega_r C_e.$$
(2.87)

Note, these are coupling of mode equations in time. Now, the modes are electronic ones instead of photonic modes. But otherwise everything is the same. For the case of vanishing detuning it is especially easy to eliminate one of the variables and we arrive at

$$\frac{d^2}{dt^2}C_e = -\Omega_r^2 C_e \tag{2.88}$$

$$\frac{d^2}{dt^2}C_g = -\Omega_r^2 C_g. aga{2.89}$$

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

$$C_q(t) = \cos\left(\Omega_r t\right) \tag{2.90}$$

$$C_e(t) = -j\sin\left(\Omega_r t\right). \tag{2.91}$$

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

$$|c_b(t)|^2 = \cos^2(\Omega_r t)$$
(2.92)
$$|c_r(t)|^2 = \sin^2(\Omega_r t)$$
(2.93)

$$|c_a(t)|^2 = \sin^2(\Omega_r t), \qquad (2.93)$$

as shown in Fig. 2.3. For the expectation value of the dipole operator under the assumption of a real dipole matrix element  $\vec{M} = \vec{M}^*$  we obtain

$$<\vec{\mathbf{p}}> = -\vec{M}c_ec_g^* + c.c.$$
 (2.94)

$$= -\vec{M}\sin\left(2\Omega_r t\right)\sin\left(\omega_{eg}t\right). \tag{2.95}$$



Figure 2.3: 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.(2.95), 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. So we are interested what happens in the case of loss of coherence due to additional interaction of the atoms with a heat bath.

#### 2.3.4 The Density Operator

To study incoherent or dissipative processes it is useful to switch to a statistical description 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

$$\boldsymbol{\rho} = |\psi\rangle \langle \psi| \tag{2.96}$$

or in coordinate representation by a  $2 \times 2$ -matrix

$$\boldsymbol{\rho} = \begin{pmatrix} \rho_{ee} & \rho_{eg} \\ \rho_{ge} & \rho_{gg} \end{pmatrix}.$$
(2.97)

In case of a pure state (2.46) this is

$$\boldsymbol{\rho} = \begin{pmatrix} c_e c_e^* & c_e c_g^* \\ c_g c_e^* & c_g c_g^* \end{pmatrix}.$$
(2.98)

It is obvious, that, 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 and 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 medium polarization.

The expectation value of an arbitrary operator  $\mathbf{A}$  can be computed using the trace formula

$$\langle \mathbf{A} \rangle = Tr\{\boldsymbol{\rho}\mathbf{A}\} = \langle \psi | \mathbf{A} | \psi \rangle.$$
 (2.99)

The advantage of the density operator is, that mixtures of pure states can also be treated in a statistical sense. 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

$$\rho = p_e |e\rangle \langle e| + p_g |g\rangle \langle g|$$
(2.100)

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

$$\langle \mathbf{A} \rangle = Tr\{\boldsymbol{\rho}\mathbf{A}\} = p_e \langle e|\mathbf{A}|e\rangle + p_g \langle g|\mathbf{A}|g\rangle.$$
(2.101)

Since the matrices (2.64) to (2.67) build a complete base in the space of  $2 \times 2$ -matrices, we can express the density matrix as

$$\boldsymbol{\rho} = \rho_{ee} \frac{1}{2} (\mathbf{1} + \boldsymbol{\sigma}_z) + \rho_{gg} \frac{1}{2} (\mathbf{1} - \boldsymbol{\sigma}_z) + \rho_{eg} \boldsymbol{\sigma}^+ + \rho_{ge} \boldsymbol{\sigma}^- \qquad (2.102)$$

$$= \frac{1}{2}\mathbf{1} + \frac{1}{2}(\rho_{ee} - \rho_{gg})\boldsymbol{\sigma}_z + \rho_{eg}\boldsymbol{\sigma}^+ + \rho_{ge}\boldsymbol{\sigma}^-, \qquad (2.103)$$

since the trace of the density matrix is always one (normalization). Choosing the new base  $\mathbf{1}, \boldsymbol{\sigma}_x, \boldsymbol{\sigma}_y, \boldsymbol{\sigma}_z$ , we obtain

$$\boldsymbol{\rho} = \frac{1}{2} \mathbf{1} + \frac{1}{2} (\rho_{ee} - \rho_{gg}) \boldsymbol{\sigma}_z + d_x \boldsymbol{\sigma}_x + d_y \boldsymbol{\sigma}_y, \qquad (2.104)$$

with

$$d_x = \frac{1}{2} \left( \rho_{eg} + \rho_{ge} \right) = \Re\{\langle \boldsymbol{\sigma}^{(+)} \rangle\}, \qquad (2.105)$$

$$d_y = \frac{1}{2} \left( \rho_{eg} - \rho_{ge} \right) = \Im\{<\boldsymbol{\sigma}^{(+)}>\}.$$
 (2.106)

The expectation value of the dipole operator is given by (2.73)

$$\langle \vec{\mathbf{p}} \rangle = Tr\{\boldsymbol{\rho}\vec{\mathbf{p}}\} = -\vec{M}^*Tr\{\boldsymbol{\rho}\,\boldsymbol{\sigma}^+\} + c.c. = -\vec{M}^*\rho_{ge} + c.c.$$
(2.107)

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

$$\dot{\boldsymbol{\rho}} = \frac{d}{dt} |\psi\rangle \langle \psi| + h.c. = \frac{1}{j\hbar} \mathbf{H} |\psi\rangle \langle \psi| - \frac{1}{j\hbar} |\psi\rangle \langle \psi| \mathbf{H} \quad (2.108)$$
$$= \frac{1}{j\hbar} [\mathbf{H}, \boldsymbol{\rho}].$$

Due to the linear nature of the 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

$$\dot{\boldsymbol{\rho}} = \frac{1}{\mathrm{j}\hbar} [\mathbf{H}_A, \boldsymbol{\rho}] = -\mathrm{j} \frac{\omega_{\in g}}{2} [\boldsymbol{\sigma}_z, \boldsymbol{\rho}].$$
(2.109)

Using the commutator relations (2.53) - (2.55), the result is

$$\dot{\rho}_{\epsilon e} = 0, \qquad (2.110)$$

$$\dot{\rho}_{gg} = 0,$$
 (2.111)

$$\dot{\rho}_{eg} = -j\omega_{eg}\rho_{eg} \rightarrow \rho_{eg}(t) = e^{-j\omega_{eg}t}\rho_{eg}(0),$$
 (2.112)

$$\dot{\rho}_{ge} = j\omega_{eg}\rho_{ge} \rightarrow \rho_{ge}(t) = e^{j\omega_{eg}t}\rho_{ge}(0).$$
 (2.113)

Again the isolated two-level atom has a rather simple dynamics, the populations are constant, only the dipole moment oscillates with the transition frequency  $\omega_{\in g}$ , if there has been a dipole moment induced at t = 0, i.e. the system is in a superposition state.

#### 2.3.5 Energy- and Phase-Relaxation

In reality, there is no isolated atom. Indeed in our case we are interested with a radiating atom, i.e. it has 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

$$\mathbf{H} = \mathbf{H}_A + \mathbf{H}_F + \mathbf{H}_{A-F}.$$
 (2.114)

Here,  $\mathbf{H}_A$  is the Hamiltonian of the atom,  $\mathbf{H}_F$  of the free field and  $\mathbf{H}_{A-F}$  describes the interaction between them. A complete treatment along these lines is beyond the scope of this class and is usually done in classes on Quantum Mechanics. But the result of this calculation is simple and leads in the von Neumann equation of the reduced density matrix, i.e. the density matrix of the atom. With the spontaneous emission rate  $1/\tau_{sp}$ , i.e. the inverse spontaneous life time  $\tau_{sp}$ , the populations change according to

$$\frac{d}{dt}|c_e(t)|^2 = \frac{d}{dt}\rho_{ee} = -\Gamma_e\rho_{ee} + \Gamma_a\rho_{gg}$$
(2.115)

with the abbreviations

$$\Gamma_e = \frac{1}{\tau_{sp}} (n_{th} + 1),$$
(2.116)

$$\Gamma_a = \frac{1}{\tau_{sp}} n_{th}. \tag{2.117}$$

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

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

$$\frac{d}{dt}\rho_{gg} = -\frac{d}{dt}\rho_{ee} = \Gamma_e \rho_{ee} - \Gamma_a \rho_{gg}.$$
(2.118)

If the populations decay, so does the polarization too, since  $\rho_{ge} = c_e^* c_g$ , i.e.

$$\frac{d}{dt}\rho_{ge} = j\omega_{eg}\rho_{eg} - \frac{\Gamma_e + \Gamma_a}{2}\rho_{ge}.$$
(2.119)

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

Taking the coherent (??-2.113) and incoherent processes (2.118-2.119) into account results in the following equations for the normalized average dipole moment  $d = d_x + jd_y$  and the inversion w

$$\dot{d} = \dot{\rho}_{ge} = (j\omega_{eg} - \frac{1}{T_2})d,$$
 (2.120)

$$\dot{w} = \dot{\rho}_{ee} - \dot{\rho}_{gg} = -\frac{w - w_0}{T_1},$$
(2.121)

with the time constants

$$\frac{1}{T_1} = \frac{2}{T_2} = \Gamma_e + \Gamma_a = \frac{2n_{th} + 1}{\tau_{sp}}$$
(2.122)

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

$$w_0 = \frac{\Gamma_a - \Gamma_e}{\Gamma_a + \Gamma_e} = \frac{-1}{1 + 2n_{th}} = -\tanh\left(\frac{\hbar\omega_{eg}}{2kT}\right).$$
 (2.123)

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  $c_g$ . This coherence is destroyed by the interaction of the two -level system with the environment. In this model the energy relaxation is half the phase relaxation rate or

$$T_2 = 2T_1. (2.124)$$

The atoms in a laser medium do not only interact with the electromagnetic field, but in addition also with phonons of the host lattice, they 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. Some of these processes are only destroying the phase, but do actually not lead to an energy loss in the system. 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,

$$T_2 \le 2T_1.$$
 (2.125)

If the inversion deviates from its equilibrium value  $w_0$  it relaxes back into equilibrium with a time constant  $T_1$ . Eq. (2.123) 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  $\Gamma_a$  deviates from the equilibrium expression (2.117), and it has to be replaced by the pump rate  $\Lambda$ . If the pump rate  $\Lambda$  exceeds  $\Gamma_e$ , the inversion corresponding to Eq. (2.123) becomes positive,

$$w_0 = \frac{\Lambda - \Gamma_e}{\Lambda + \Gamma_e}.$$
(2.126)

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

$$\frac{\Gamma_e}{\Gamma_a} = \frac{1+\bar{n}}{\bar{n}} = e^{\frac{\hbar\omega_{eg}}{kT}} < 1.$$
(2.127)

Thus the pumping of the two-level system drives the system far away from thermal equilibrium, which has to be expected.

## 2.3.6 The Two-Level Atom with a Coherent Classical External Field

If there is in addition to the coupling to an external heat bath, which models the spontaneous decay, pumping, and other incoherent processes, a coherent external field, the Hamiltonian has to be extended by the dipole interaction with that field,

$$\mathbf{H}_E = -\vec{\mathbf{p}}\vec{E}(\vec{x}_A, t). \tag{2.128}$$

Again we use the interaction Hamiltonian in RWA

$$\mathbf{H}_E = \frac{1}{2}\vec{M}^*\vec{E}(t)^{(-)}\sigma^+ + h.c..$$
(2.129)

This leads in the von Neumann equation to the additional term

$$\dot{\boldsymbol{\rho}}|_E = \frac{1}{j\hbar} [H_E, \boldsymbol{\rho}] \tag{2.130}$$

$$= \frac{1}{2j\hbar} \vec{M}^* \vec{E}(t)^{(-)} [\boldsymbol{\sigma}^+, \boldsymbol{\rho}] + h.c. \qquad (2.131)$$

or

$$\dot{\rho}_{ee}|_E = \frac{1}{2j\hbar} \vec{M}^* \vec{E}^{(-)} \rho_{ge} + c.c.,$$
 (2.132)

$$\dot{\rho}_{ge}|_E = \frac{1}{2j\hbar} \vec{M} \vec{E}^{(+)} (\rho_{ee} - \rho_{gg}),$$
(2.133)

$$\dot{\rho}_{gg}|_E = -\frac{1}{2j\hbar} \vec{M}^* \vec{E}^{(-)} \rho_{ge} + c.c..$$
 (2.134)

The evolution of the dipole moment and the inversion is changed by

$$\dot{d}|_E = \dot{\rho}_{ge}|_E = \frac{1}{2j\hbar} \vec{M} \vec{E}^{(+)} w,$$
 (2.135)

$$\dot{w}|_E = \dot{\rho}_{ee}|_E - \dot{\rho}_{gg}|_E = \frac{1}{j\hbar} (\vec{M}^* \vec{E}^{(-)} d^* - \vec{M} \vec{E}^{(+)} d).$$
 (2.136)

Thus, the total dynamics of the two-level system including the pumping and dephasing processes from Eqs.(2.120) and (2.121) is given by

$$\dot{d} = -(\frac{1}{T_2} - j\omega_{eg})d + \frac{1}{2j\hbar}\vec{M}\vec{E}^{(+)}w, \qquad (2.137)$$

$$\dot{w} = -\frac{w - w_0}{T_1} + \frac{1}{j\hbar} (\vec{M}^* \vec{E}^{(-)} d - \vec{M} \vec{E}^{(+)} d^*).$$
(2.138)

These equations are called Bloch-equations. They describe the dynamics of an atom interacting with a classical electric field. Together with Eq. (2.7) they build the Maxwell-Bloch equations.

## 2.4 Dielectric Susceptibility

If the incident field is monofrequent, i.e.

$$\vec{E}(t)^{(+)} = \vec{E}e^{j\omega t},$$
 (2.139)

and assuming that the inversion w of the atom will be well represented by its time average  $w_s$ , then the dipole moment will oscillate with the same frequency in the stationary state

$$d = \hat{d}e^{\mathbf{j}\omega t},\tag{2.140}$$

and the inversion will adjust to a new stationary value  $w_s$ . With ansatz (2.139) and (2.140) in Eqs. (2.137) and (2.138), we obtain

$$\hat{d} = \frac{-j}{2\hbar} \frac{w_s}{1/T_2 + j(\omega - \omega_{eg})} \vec{M} \hat{\vec{E}},$$
 (2.141)

$$w_s = \frac{w_0}{1 + \frac{T_1}{\hbar^2} \frac{1/T_2 |\vec{M}\vec{E}|^2}{(1/T_2)^2 + (\omega_{eq} - \omega)^2}}.$$
(2.142)

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

$$L(\omega) = \frac{(1/T_2)^2}{(1/T_2)^2 + (\omega_{eg} - \omega)^2},$$
(2.143)

and connect the square of the field  $|\hat{\vec{E}}|^2$  to the intensity I of a propagating plane wave, according to Eq. (2.32),  $I = \frac{1}{2Z_F} |\hat{\vec{E}}|^2$ ,

$$w_s = \frac{w_0}{1 + \frac{I}{I_s} L(\omega)}.$$
 (2.144)

Thus the stationary inversion depends on the intensity of the incident light, therefore,  $w_0$  can be called the unsaturated inversion,  $w_s$  the saturated inversion and  $I_s$ , with

$$I_{s} = \left[\frac{2T_{1}T_{2}Z_{F}}{\hbar^{2}} \frac{\left|\vec{M}\vec{E}\right|^{2}}{\left|\vec{E}\right|^{2}}\right]^{-1}, \qquad (2.145)$$

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

$$\langle \vec{p} \rangle = -(\vec{M}^*d + \vec{M}d^*).$$
 (2.146)

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

$$\hat{\vec{P}}^{(+)} = -2N\vec{M}^*\hat{d},$$
 (2.147)

$$\hat{\vec{P}}^{(+)} = \epsilon_0 \chi(\omega) \hat{\vec{E}}.$$
(2.148)

From the definitions (2.147), (2.148) and Eq. (2.141) we obtain for the linear susceptibility of the medium

$$\chi(\omega) = \vec{M}^* \vec{M}^T \frac{\mathrm{j}N}{\hbar\epsilon_0} \frac{w_s}{1/T_2 + \mathrm{j}(\omega - \omega_{eg})},\tag{2.149}$$

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

$$\begin{pmatrix} M_x M_x & M_x M_y & M_x M_z \\ M_y M_x & M_y M_y & M_y M_z \\ M_z M_x & M_z M_y & M_z M_z \end{pmatrix} = \begin{pmatrix} \overline{M_x^2} & 0 & 0 \\ 0 & \overline{M_y^2} & 0 \\ 0 & 0 & \overline{M_z^2} \end{pmatrix} = \frac{1}{3} |\vec{M}|^2 \mathbf{1}. \quad (2.150)$$

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

$$\chi(\omega) = \frac{1}{3} |\vec{M}|^2 \frac{jN}{\hbar\epsilon_0} \frac{w_s}{1/T_2 + j(\omega - \omega_{eg})}.$$
 (2.151)

Real and imaginary part of the susceptibility

$$\chi(\omega) = \chi'(\omega) + j\chi''(\omega) \tag{2.152}$$

are then given by

$$\chi'(\omega) = -\frac{|\vec{M}|^2 N w_s T_2^2(\omega_{eg} - \omega)}{3\hbar\epsilon_0} L(\omega), \qquad (2.153)$$

$$\chi''(\omega) = \frac{|\dot{M}|^2 N w_s T_2}{3\hbar\epsilon_0} L(\omega).$$
(2.154)

#### 2.4. DIELECTRIC SUSCEPTIBILITY

If the incident radiation is weak enough, i.e.

$$T_1 T_2 \frac{|\vec{M}^* \vec{E}|^2}{\hbar^2} L(\omega) \ll 1$$
 (2.155)

we obtain  $w_s \approx w_0$ . Since  $w_0 < 0$ , and especially for optical transitions  $w_0 = -1$ , real and imaginary part of the susceptibility are shown in Fig. 2.4.



Figure 2.4: Real and imaginary part of the complex susceptibility.

The susceptibility computed quantum mechanically compares well with the classical susceptibility derived from the harmonic oscillator model close to the transistion frequency for a transition with reasonably high  $Q = T_2 \omega_{ab}$ . Note, there is an appreciable deviation 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 becomes obvious, when the propagation of a plane electro-magnetic wave through this medium is considered,

$$\vec{E}(z,t) = \Re\left\{\hat{\vec{E}}e^{\mathbf{j}(\omega t - kz)}\right\},\qquad(2.156)$$

which is propagating in the positive z-direction. The propagation constant k is related to the susceptibility by

$$k = \omega \sqrt{\mu_0 \epsilon_0 (1 + \chi(\omega))} \approx k_0 \left( 1 + \frac{1}{2} \chi(\omega) \right), \quad \text{with} \quad k_0 = \omega \sqrt{\mu_0 \epsilon_0} \quad (2.157)$$

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

$$k = k_0 \left(1 + \frac{\chi'}{2}\right) + j k_0 \frac{\chi''}{2}.$$
 (2.158)

The real part of the susceptibility contributes to the refractive index  $n = 1 + \chi'/2$ . In 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.

## 2.5 Rate Equations

With the wave equation Eq.(2.7) and the expression for the polarization induced by the electric field of the wave, we end up with the complete Maxwell-Bloch equations describing an electromagnetic field interacting with a statistical ensemble of atoms that are located at postions  $z_i$ 

$$\left(\Delta - \frac{1}{c_0^2} \frac{\partial^2}{\partial t^2}\right) \vec{E}^{(+)}(z,t) = \mu_0 \frac{\partial^2}{\partial t^2} \vec{P}^{(+)}(z,t), \qquad (2.159)$$

$$\vec{P}^{(+)}(z,t) = -2N\vec{M}^*d(z,t)$$
(2.160)

$$\dot{d}(z,t) = -(\frac{1}{T_2} - j\omega_{eg})d + \frac{1}{2j\hbar}\vec{M}\vec{E}^{(+)}w, \qquad (2.161)$$

$$\dot{w}(z,t) = -\frac{w-w_0}{T_1} + \frac{1}{j\hbar} (\vec{M}^* \vec{E}^{(-)} d - \vec{M} \vec{E}^{(+)} d^2).162)$$

In the following we consider a electromagnetic wave with polarization vector  $\vec{e}$ , frequency  $\omega_{eg}$  and wave number  $k_0 = \omega_{eg}/c_0$  with a slowly varying envelope propagating to the right

$$\vec{E}(z,t)^{(+)} = \sqrt{2Z_{F_0}}A(z,t)e^{j(\omega_{eg}t-k_0z)}\vec{e},$$
(2.163)

with

$$\left|\frac{\partial A(z,t)}{\partial t}\right|, \left|c\frac{\partial A(z,t)}{\partial z}\right| \ll \left|\omega_{eg}A(z,t)\right|.$$
(2.164)

Note, we normalized the complex amplitude A(t) such that its magnitude square is proportional to the intensity of the wave. This will also excite a wave of dipole moments in the atomic medium according to

$$d(z,t) = \hat{d}(z,t)e^{j(\omega_{eg}t - k_0 z)},$$
 (2.165)

that is also slowly varying. In that case, we obtain from Eq.(2.159-d) in leading order

$$\left(\frac{\partial}{\partial z} + \frac{1}{c_0}\frac{\partial}{\partial t}\right)A(z,t) = jN\vec{e}^T\vec{M}^*\sqrt{\frac{Z_{F_0}}{2}}\hat{d}(z,t), \qquad (2.166)$$

$$\frac{\partial}{\partial t}d(z,t) = -\frac{1}{T_2}\hat{d} + \frac{\sqrt{2Z_{F_0}}}{2j\hbar}\left(\vec{M}\vec{e}\right)A(t)w \qquad (2.167)$$

$$\frac{\partial}{\partial t}w(z,t) = -\frac{w-w_0}{T_1} + \frac{\sqrt{2Z_{F_0}}}{j\hbar} \left(\left(\vec{M}^*\vec{e}^*\right)A^*(t)\hat{d} - \left(\vec{M}\vec{e}\right)A(t)\hat{d}^*\right)$$

Furthermore, in the limit, where the dephasing time  $T_2$  is much faster than the variation in the envelope of the electric field, one can adiabatically eliminate the rapidly decaying dipole moment, i.e.

$$\hat{d} = T_2 \frac{\sqrt{2Z_{F_0}}}{2j\hbar} \left(\vec{M}\vec{e}\right) A(t)w, \qquad (2.169)$$

$$\dot{w} = -\frac{w - w_0}{T_1} + \frac{|A(t)|^2}{E_s}w,$$
 (2.170)

where  $E_s = I_s T_1$ , is called the saturation fluence,  $[J/cm^2]$ , of the medium.

Note, now we don't have to care anymore about the dipole moment and we are left over with a rate equation for the population difference of the medium and the complex field amplitude of the wave.

$$\left(\frac{\partial}{\partial z} + \frac{1}{c_0}\frac{\partial}{\partial t}\right)A(z,t) = \frac{N\hbar}{4T_2E_s}w(z,t)A(z,t), \qquad (2.171)$$

$$\dot{w} = -\frac{w - w_0}{T_1} + \frac{|A(z,t)|^2}{E_s}w(z,t)$$
 (2.172)

Equation (2.171) clearly shows that we obtain gain for an inverted medium and that the gain saturates with the electromagnetic power density flowing through the medium.

## 2.6 Pulse Propagation with Dispersion and Gain

In many cases, mode locking of lasers can be most easily studied in the time domain. Then mode locking becomes a nonlinear, dissipative wave propaga-

45

tion problem. In this chapter, we discuss the basic elements of pulse propagation in linear and nonlinear media, as far as it is necessary for the following chapters. A comprehensive discussion of nonlinear pulse propagation can be found in [6].

We consider the electric field of a monochromatic electromagnetic wave with frequency  $\Omega$ , which propagates along the z-axis, and is polarized along the x-axis, (Fig. 2.5).



Figure 2.5: Transverse electro-magnetic wave. Figure by MIT OCW.

In a linear, isotropic, homogeneous, and lossless medium the electric field of that electromagnetic wave is given by

$$E(z,t) = \vec{e}_{x}E(z,t),$$

$$E(z,t) = \Re \left\{ \tilde{E}(\Omega)e^{j(\Omega t - Kz)} \right\}$$

$$= |\tilde{E}|\cos(\Omega t - Kz + \varphi),$$
(2.173)

where  $\tilde{E} = |\tilde{E}|e^{j\varphi}$  is the complex wave amplitude of the electromagnetic wave at frequency  $\Omega$  and wave number  $K = \Omega/c = n\Omega/c_0$ . Here, *n* is the refractive index, *c* the velocity of light in the medium and  $c_0$  the velocity of light in vacuum, respectively. The planes of constant phase propagate with the phase velocity *c* of the wave. Usually, we have a superposition of many frequencies with spectrum shown in Fig. 2.6



Figure 2.6: Electric field and pulse envelope in frequency domain.

In general, the refractive index is a function of frequency and one is interested in the propagation of a pulse, that is produced by a superposition of monochromatic waves grouped around a certain carrier frequency  $\omega_0$  (Fig. 2.6)

$$E(z,t) = \Re\left\{\frac{1}{2\pi}\int_0^\infty \tilde{E}(\Omega)e^{\mathbf{j}(\Omega t - K(\Omega)z)}d\Omega\right\}.$$
(2.174)

We can always separate the complex electric field in Eq. (2.174) into a carrier wave and an envelope A(z,t)

$$E(z,t) = \Re \left\{ A(z,t) e^{j(\omega_0 t - K(\omega_0)z)} \right\}.$$
 (2.175)

The envelope is given by

$$A(z,t) = \frac{1}{2\pi} \int_{-\omega_0 \to -\infty}^{\infty} \tilde{A}(\omega) e^{j(\omega t - k(\omega)z)} d\omega, \qquad (2.176)$$

where we introduced the offset frequency, offset wave vector and spectrum of

the envelope

$$\omega = \Omega - \omega_0, \qquad (2.177)$$

$$k(\omega) = K(\omega_0 + \omega) - K(\omega_0), \qquad (2.178)$$

$$\tilde{A}(\omega) = \tilde{E}(\Omega = \omega_0 + \omega),$$
 (2.179)

(see Fig. 2.8).

Depending on the dispersion relation, the pulse will be reshaped during propagation.



Figure 2.7: Electric field and pulse envelope in time domain.

#### 2.6.1 Dispersion

If the spectral width of the pulse is small compared to the carrier frequency, the envelope is only slowly varying with time. Additionally, if the dispersion relation  $k(\omega)$  is only slowly varying over the pulse spectrum, it is useful to represent the dispersion relation,  $K(\Omega)$ , see Fig. 2.8, by its Taylor expansion

$$k(\omega) = k'\omega + \frac{k''}{2}\omega^2 + \frac{k^{(3)}}{6}\omega^3 + O(\omega^4).$$
(2.180)

If the refractive index depends on frequency, the dispersion relation is no longer linear with respect to frequency, see Fig. 2.8.

48



Figure 2.8: Taylor expansion of dispersion relation.

For the moment, we keep only the first term, the linear term, in Eq.(2.180). Then we obtain for the pulse envelope from (2.176) by definition of the group velocity  $v_q = 1/k'$ 

$$A(z,t) = A(0,t-z/v_q).$$
(2.181)

Thus the derivative of the dispersion relation at the carrier frequency determines the velocity of the corresponding wave packet. We introduce the local time  $t' = t - z/v_g$ . With respect to this local time the pulse shape is invariant during propagation

$$A(z,t') = A(0,t').$$
(2.182)

If the spectrum of the pulse becomes broad enough, so that the second order term in (2.180) becomes important, wave packets with different carrier frequencies propagate with different group velocities and the pulse spreads. When keeping in the dispersion relation terms up to second order it follows from (2.176)

$$\frac{\partial A(z,t')}{\partial z} = -j \frac{k''}{2} \frac{\partial^2 A(z,t')}{\partial t'^2}.$$
(2.183)

This is equivalent to the Schrödinger equation for a nonrelativistic free particle. Like in Quantum Mechanics, it describes the spreading of a wave packet. Here, the spreading is due to the first nontrivial term in the dispersion relation, which describes spreading of an electromagnetic wave packet via group velocity dispersion (GVD). Of course, we can keep all terms in the dispersion relation, which would lead to higher order derivatives in the equation for the envelope

$$\frac{\partial A(z,t')}{\partial z} = j \sum_{n=2}^{\infty} \frac{k^{(n)}}{n!} \left( j \frac{\partial}{\partial t'} \right)^n A(z,t').$$
(2.184)

Therefore, one usually calls the first term dispersion and the higher order terms higher order dispersion. In the following, we always work in the local time frame to get rid of the trivial motion of the pulse. Therefore, we drop the prime to simplify notation. Figure 2.9 shows the evolution of a Gaussian wave packet during propagation in a medium which has no higher order dispersion and k'' = 2 is given in normalized units. The pulse spreads continuously.



Figure 2.9: Amplitude of the envelope of a Gaussian pulse, |A(z, t')|, in a dispersive medium.



Figure 2.10: (a) Phase and (b) instantaneous frequency of a Gaussian pulse during propagation through a medium with positive or negative dispersion.

As shown in Fig. 2.10(a), during propagation in the dispersive medium, the pulse acquires a linear chirp, i.e. its phase becomes parabolic. The derivative of the phase with respect to time is the instantaneous frequency Fig. 2.10(b). It indicates, that the low frequencies are in the front of the pulse, whereas the high frequencies are in the back of the pulse. This is due to the positive dispersion k'' > 0, which causes, that wave packets with lower frequencies travel faster than wave packets with higher frequencies.

#### 2.6.2 Loss and Gain

If the medium considered has loss, we can incorporate this loss into a complex refractive index

$$n(\Omega) = n_r(\Omega) + jn_i(\Omega). \tag{2.185}$$

The refractive index is determined by the linear response,  $\chi(\Omega)$ , of the polarization in the medium onto the electric field induced in the medium

$$n(\Omega) = \sqrt{1 + \chi(\Omega)}.$$
(2.186)

For an optically thin medium, i.e.  $|\chi(\Omega)| \ll 1$  we obtain approximately

$$n(\Omega) \approx 1 + \frac{\chi(\Omega)}{2}.$$
 (2.187)

For a two level atom with an electric dipole transition, the susceptibility is given, in the rotating wave approximation, by the complex Lorentzian lineshape

$$\chi(\Omega) = \frac{2j\alpha}{1 - j\frac{\Omega - \Omega_0}{\Delta\Omega}},$$
(2.188)

where  $\alpha$  will turn out to be the peak absorption or gain of the transition, which is proportional to the density of the atomic inversion,  $\Omega_0$  is the center frequency of the optical transition and  $\Delta\Omega$  is the HWHM linewidth of the transition. Figure 2.11 shows the normalized real and imaginary part of the complex Lorentzian

$$\chi_r(\Omega) = \frac{-2\alpha \frac{(\Omega - \Omega_0)}{\Delta \Omega}}{1 + \left(\frac{\Omega - \Omega_0}{\Delta \Omega}\right)^2},$$
(2.189)

$$\chi_i(\Omega) = \frac{2\alpha}{1 + \left(\frac{\Omega - \Omega_0}{\Delta\Omega}\right)^2}, \qquad (2.190)$$

which are the real- and imaginary part of the complex susceptibility for a noninverted optical transition, i.e. loss.



Figure 2.11: Normalized real and imaginary part of the complex Lorentzian.

The real part of the transition modifies the real part of the refractive index of the medium, whereas the imaginary part leads to loss in the case of a noninverted medium.

In the derivation of the wave equation for the pulse envelope (2.184) in section 2.6.1, there was no restriction to a real refractive index. Therefore, the wave equation (2.184) also treats the case of a complex refractive index. If we assume a medium with the complex refractive index (2.187), then the wave number is given by

$$K(\Omega) = \frac{\Omega}{c_0} \left( 1 + \frac{1}{2} \left( \chi_r(\Omega) + j\chi_i(\Omega) \right) \right).$$
 (2.191)

Since we introduced a complex wave number, we have to redefine the group velocity as the inverse derivative of the real part of the wave number with respect to frequency. At line center, we obtain

$$v_g^{-1} = \left. \frac{\partial K_r(\Omega)}{\partial \Omega} \right|_{\Omega_0} = \frac{1}{c_0} \left( 1 - \alpha \frac{\Omega_0}{\Delta \Omega} \right).$$
(2.192)

Thus, for a narrow absorption line,  $\alpha > 0$  and  $\frac{\Omega_0}{\Delta\Omega} \gg 1$ , the absolute value of the group velocity can become much larger than the velocity of light in

vacuum. The opposite is true for an inverted, and therefore, amplifying transition,  $\alpha < 0$ . There is nothing wrong with it, since the group velocity only describes the motion of the peak of a Gaussian wave packet, which is not a causal wave packet. A causal wave packet is identical to zero for some earlier time  $t < t_0$ , in some region of space. A Gaussian wave packet fills the whole space at any time and can be reconstructed by a Taylor expansion at any time. Therefore, the tachionic motion of the peak of such a signal does not contradict special relativity.

The imaginary part in the wave vector (2.191), due to gain and loss, has to be completely treated in the envelope equation (2.184). In the frequency domain this leads for a wave packet with a carrier frequency at line center,  $\omega_0 = \Omega_0$  and  $K_r(\Omega_0) = k_0$ , to the term

$$\frac{\partial \tilde{A}(z,\omega)}{\partial z}\bigg|_{(loss)} = \frac{-\alpha k_0}{1 + \left(\frac{\omega}{\Delta\Omega}\right)^2} \tilde{A}(z,\omega).$$
(2.193)

In the time domain, we obtain up to second order in the inverse linewidth

$$\frac{\partial A(z,t)}{\partial z}\Big|_{(loss)} = -\alpha k_0 \left(1 + \frac{1}{\Delta\Omega^2} \frac{\partial^2}{\partial t^2}\right) A(z,t), \quad (2.194)$$

which corresponds to a parabolic approximation of the Lorentzian line shape at line center, (Fig. 2.11). For an inverted optical transition, we obtain a similar equation, we only have to replace the loss by gain

$$\frac{\partial A(z,t)}{\partial z}\Big|_{(gain)} = g\left(1 + \frac{1}{\Omega_g^2}\frac{\partial^2}{\partial t^2}\right)A(z,t),$$
(2.195)

where  $g = -\alpha k_0$  is the peak gain at line center per unit length and  $\Omega_g$  is the HWHM linewidth of the gain transition. The gain is proportional to the inversion in the atomic system, see Eq.(2.149), which also depends on the field strength or intensity according to the rate equation (2.172)

$$\frac{\partial g(z,t)}{\partial t} = -\frac{g - g_0}{\tau_L} - g \frac{|A(z,t)|^2}{E_L}.$$
(2.196)

Here,  $E_L$  is the saturation fluence of the gain medium and  $\tau_L$  the life time of the inversion, i.e. the upper-state life time of the gain medium.

	Fused Quartz	Sapphire
$a_1$	0.6961663	1.023798
$a_2$	0.4079426	1.058364
$a_3$	0.8974794	5.280792
$\lambda_1^2$	$4.679148 \cdot 10^{-3}$	$3.77588 \cdot 10^{-3}$
$\lambda_1^2 \ \lambda_2^2 \ \lambda_3^2$	$1.3512063 \cdot 10^{-2}$	$1.22544 \cdot 10^{-2}$
$\lambda_3^2$	$0.9793400 \cdot 10^2$	$3.213616 \cdot 10^2$

Table 2.1: Table with Sellmeier coefficients for fused quartz and sapphire.

## 2.7 Kramers-Kroenig Relations

The linear susceptibility is the frequency response of a linear system to an applied electric field, which is causal, and therefore real and imaginary parts obey Kramers-Kroenig Relations

$$\chi_r(\Omega) = \frac{2}{\pi} \int_0^\infty \frac{\omega \chi_i(\omega)}{\omega^2 - \Omega^2} d\omega = n^2(\Omega) - 1, \qquad (2.197)$$

$$\chi_i(\Omega) = -\frac{2}{\pi} \int_0^\infty \frac{\Omega \chi_r(\omega)}{\omega^2 - \Omega^2} d\omega.$$
(2.198)

In transparent media one is operating far away from resonances. Then the absorption or imaginary part of the susceptibility can be approximated by

$$\chi_i(\Omega) = \sum_i A_i \delta\left(\omega - \omega_i\right) \tag{2.199}$$

and the Kramers-Kroenig relation results in a Sellmeier Equation for the refractive index

$$n^{2}(\Omega) = 1 + \sum_{i} A_{i} \frac{\omega_{i}}{\omega_{i}^{2} - \Omega^{2}}$$
 (2.200)

$$= 1 + \sum_{i} a_i \frac{\lambda}{\lambda^2 - \lambda_i^2}.$$
 (2.201)

For an example Table 2.1 shows the sellmeier coefficients for fused quartz and sapphire.

A typical situation for a material having resonances in the UV and IR, such as glass, is shown in Fig. 2.12



Figure 2.12: Typcial distribution of absorption lines in a medium transparent in the visible.

#### Figure by MIT OCW.

The regions where the refractive index is decreasing with wavelength is usually called normal dispersion range and the opposite behavior anormal dispersion

 $\frac{dn}{d\lambda} < 0 : \text{normal dispersion (blue refracts more than red)} \\ \frac{dn}{d\lambda} > 0 : \text{abnormal dispersion}$ 

Fig.2.13 shows the transparency range of some often used media.



Figure 2.13: Transparency range of some materials.

Figure by MIT OCW.

## 2.8 Pulse Shapes and Time-Bandwidth Products

The following table 2.2 shows pulse shape, spectrum and time bandwidth products of some often used pulse forms.

a(t)	$\hat{a}(\omega) = \int_{-\infty}^{\infty} a(t) e^{-j\omega t} dt$	$\Delta t$	$\Delta t \cdot \Delta f$
Gauss: $e^{-\frac{t^2}{t\tau^2}}$	$\sqrt{2\pi}\tau e^{-\frac{1}{t}\tau^2\omega^2}$	$2\sqrt{\ln 2}\tau$	0.441
Hyperbolicsecant: $\operatorname{sech}(\frac{t}{\tau})$	$\frac{\tau}{2} \operatorname{sech}(\frac{\pi}{2}\tau\omega)$	1.7627 $\tau$	0.315
Rect-function: = $\begin{cases} 1, &  t  \le \tau/2 \\ 0, &  t  > \tau/2 \end{cases}$	$ au rac{\sin( au\omega/2)}{ au\omega/2}$	τ	0.886
Lorentzian: $\frac{1}{1+(t/\tau)^2}$	$2\pi\tau e^{- \tau\omega }$	1.287 $\tau$	0.142
Double-Exponential: $e^{-\left \frac{t}{\tau}\right }$	$\frac{\tau}{1+(\omega\tau)^2}$	ln2 $\tau$	0.142

Table 2.2: Pulse shapes, corresponding spectra and time bandwidth products.



Figure 2.14: Fourier relationship to table above. Figure by MIT OCW.



Figure 2.15: Fourier relationships to table above. Figure by MIT OCW.

60

# Bibliography

- I. I. Rabi: "Space Quantization in a Gyrating Magnetic Field,". Phys. Rev. 51, 652-654 (1937).
- [2] B. R. Mollow, "Power Spectrum of Light Scattered by Two-Level Systems," Phys. Rev 188, 1969-1975 (1969).
- [3] P. Meystre, M. Sargent III: Elements of Quantum Optics, Springer Verlag (1990).
- [4] L. Allen and J. H. Eberly: Optical Resonance and Two-Level Atoms, Dover Verlag (1987).
- [5] G. B. Whitham: "Linear and Nonlinear Waves," John Wiley and Sons, NY (1973).

### BIBLIOGRAPHY

62