

The optical activity induced in a material by a magnetic field in the direction of light propagation is known as the Faraday effect described by the Verdet constant (better material parameter  $V(\omega)$ ) via

$$S_+ = V(\omega)lB \quad (3.35e)$$

In magnetic dichroism one investigates the difference in the absorption spectra for  $\sigma^+$  and  $\sigma^-$  polarized light induced by the B-field.

## 3.2 Microscopic Aspects

In contrast to the preceding section, we present now the basic interaction processes between light and matter from a microscopic point of view. We use here the perturbative or weak coupling approach for the interaction between light and matter, which is in most cases sufficient for dilute systems such as gases. For solids the strong coupling approach is often necessary, which leads to the concept of polaritons and which is introduced in Chap. 5. We describe first in words the basic interaction mechanisms between light and matter, namely absorption, spontaneous and stimulated emission (Sect. 3.2.1), then we proceed to the treatment of linear optical properties in the framework of perturbation theory (Sect. 3.2.2). Since these topics are also treated in many textbooks (e.g. References [81M1, 90K1, 93H1, 96Y1] of Chap. 1 or [55S1, 71F1, 73H1, 76H1, 92M1] of Chap. 2) it is not necessary to go into too much detail here.

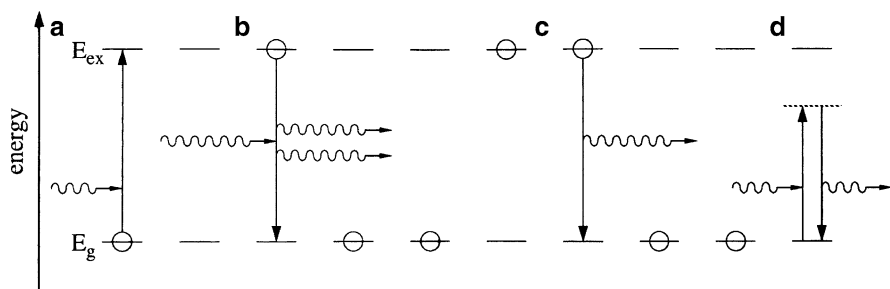
### 3.2.1 Absorption, Stimulated and Spontaneous Emission, Virtual Excitation

For simplicity we assume that we have a certain number of two-level “atoms” as shown in Fig. 3.16. Every atom has one electron which can be either in the ground or in the excited state. Later we will extend the model from a two-level system to bands in semiconductors, but the basic interaction processes remain the same.

In Fig. 3.16a an incident photon hits an atom in its ground state. With a certain probability the photon is annihilated and the electron gains enough energy to reach the excited state. For reasons of energy conservation, the photon has to fulfill the condition

$$\hbar\omega = E_{\text{ex}} - E_{\text{g}} , \quad (3.36)$$

where  $E_{\text{ex}} - E_{\text{g}}$  is the energy difference between the ground and excited states. We call this process absorption in agreement with the definition in Sect. 3.1.5 if the energy of the photon is soon converted into other forms of energy, that is, if the electron undergoes some scattering processes, which destroy its coherence



**Fig. 3.16** Schematic representations of various interaction processes of light with matter namely absorption (a), stimulated emission (b), spontaneous emission (c) and virtual excitation (d)

or more precisely the coherence of the electric polarization connected with this transition to the incident light field. See Chap. 23. The electron eventually returns to its ground state and loses its energy e.g., as phonons i.e., as heat or as a photon which is not coherent with the incident one. The first process is called non-radiative recombination, the second one resonance fluorescence.

If an incident photon hits an atom with its electron in the excited state it can induce with a certain probability a transition of the electron from the excited to the ground state. In this process a second photon is created which is identical in momentum, energy, polarization and phase to the incident one. This process is called induced or stimulated emission. This process can be used to amplify a photon field. It is therefore the basic mechanism for all lasers (Light Amplification by Stimulated Emission of Radiation). Absorption and stimulated emission are closely related events (Fig. 3.16b).

An electron in the excited state can also with a certain probability reenter the ground state by itself, either by emitting a photon (Fig. 3.16c) or by losing the transition energy through phonons or collisions. In the present context, the first mechanism is of interest. It is called spontaneous emission or spontaneous radiative recombination, while the second possibility is known as non-radiative recombination. Spontaneous emission can also be understood in another way. In Sect. 2.5 we saw in connection with (2.54) that photons are similar to harmonic oscillators and have consequently a zero-point energy. This zero-point energy exists for all photon modes. It cannot be absorbed because a harmonic oscillator does not have any states below the zero-point energy, but it can induce a transition in the way as discussed in connection with Fig. 3.16b. So we can consider spontaneous emission as a process induced by the zero-point vibrations of the electro-magnetic field, which are also called vacuum fluctuations (Fig. 3.16c) or zero point fluctuations of the electro-magnetic field.

The last process presented here is virtual excitation. Understanding this phenomenon often causes some problems for students. So we develop this topic slowly and try to explain it from various points of view in context with Fig. 3.16d. Virtual excitation means the creation of a state with the same wave function as the

excited state, but with an energy which is different from the eigenenergy of this excited state. This process becomes possible through the uncertainty principle of quantum mechanics which can be written in space and momentum coordinates.

$$\Delta x_i \Delta p_i \geq h \text{ for } i = 1, 2, 3 . \quad (3.37a)$$

A similar relation exists for energy and time

$$\Delta E \Delta t \approx \hbar . \quad (3.37b)$$

We need here (3.37b). It says that it is possible to violate energy conservation by an amount  $\Delta E$  up to a maximum time  $\Delta t$  which fulfills the above condition. Or, in other words, if we want to define the energy with a certain precision  $\Delta E$ , the state has to exist at least for a time  $\Delta t$ . In principle (3.37b) is valid also in simple classical wave theory (e.g., acoustics) and is very well known from Fourier transformations. A harmonic oscillation with central frequency  $\omega$  which lasts only for a time  $\Delta t$ , has a spectral width  $\Delta\omega$  given by

$$\Delta\omega \Delta t \geq 1 . \quad (3.37c)$$

The connection between (3.37b) and (3.37c) comes simply from the relation

$$E = \hbar\omega . \quad (3.37d)$$

It should be mentioned that a similar set of arguments holds for (3.37a), too. If we send now a photon with energy  $\hbar\omega'$  to the atom, we can excite the electron for a maximum time  $\Delta t$  given by (3.37b) or for our specific case by

$$\Delta t \approx \hbar [ (E_{\text{ex}} - E_{\text{g}}) - \hbar\omega' ]^{-1} . \quad (3.38)$$

At the latest after the time  $\Delta t$  has elapsed, the excited state must collapse. The simplest way to collapse is to emit a photon identical to the one which caused the virtual excitation. This “new” photon has however a certain phase delay with respect to the incident photon, because the energy was stored for a maximum time  $\Delta t$  in the atom. As a consequence, an electromagnetic wave propagates with a lower phase-velocity through an ensemble of atoms than through vacuum. The same effect is described phenomenologically by the refractive index  $n(\omega)$ . (See (2.15) and (2.41).) So we get a first hint of how  $n(\omega)$  can be understood and calculated in quantum mechanics. Obviously  $\Delta t$  increases if we approach the resonance condition in (3.38) and consequently wave propagation through a material will deviate more strongly from that through vacuum. This is indeed the case, as we shall see in Sect. 4.3.

If the virtually excited state emits a photon  $\hbar\omega'$  in a direction different from the incident one, we have a scattering process as discussed in connection with (3.26). If  $\hbar\omega'$  approaches the resonance energy, this scattering process is also known as resonance fluorescence as mentioned already above.

In connection with this scattering, we may ask how can light propagate at all in a clear or transparent medium? The answer is that we have in dense media many scattering atoms or centers in the coherence volume of light, independent of whether it is light from an incandescent lamp, a laser or another source. As a consequence, every scattered wave finds another one which has a phase difference, resulting in destructive interference. The only way that all scattered waves interfere constructively is just the usual propagating wave. The explanation for blue sky, which we gave in Sect. 3.1.5 fulfills, apart from the condition that the diameter of the scattering centers is small compared to the wavelength of the scattered radiation, another condition, namely that there are only a few scattering centers i.e., gas molecules, density fluctuations or other inhomogenities per coherence volume of sunlight, so that the mutually destructive interference of the scattered waves is not complete. For a detailed discussion of spatial and temporal coherence see e.g. [07M1, 07S1] of Chap. 1 and [73C1, 93S1] of Chap. 2.

If the virtually excited state disappears under simultaneous emission of a photon and the creation or annihilation of a phonon (i.e., a quantum of the lattice vibrations, as in Sect. 11.1), energy conservation for the emitted photon  $\hbar\omega_R$  implies

$$\hbar\omega_R = \hbar\omega' \pm \hbar\Omega_{\text{phonon}} . \quad (3.39)$$

This phenomenon is called Raman scattering for optical phonons, and Brillouin scattering for acoustic phonons. The “−” sign gives the Stokes and the “+” sign the anti-Stokes emission. Similar processes are also possible with more than one phonon or with excitations in the electronic or spin system of the semiconductor.

From the few phenomena outlined briefly above which involve virtual excitation, it is obvious that this mechanism is of some importance for the optical properties of matter. Therefore we want to examine it from another point of view and outline the well-known classical analog of the virtually excited states. In addition this analogy gives some justification for the calculation of the dielectric function used in Chap. 4.

Virtual excitation in quantum mechanics corresponds to a driven or forced oscillation in classical mechanics. If we have an oscillator of eigen frequency  $\omega_0$  (corresponding to the energetic differences  $E_{\text{ex}} - E_{\text{g}}$  in quantum mechanics) and if we excite it with an external frequency  $\omega$ , it will oscillate with frequency  $\omega$  after a short damped transient feature of oscillations with  $\omega_0$ . The amplitude of these steady oscillations increases with decreasing detuning  $|\omega - \omega_0|$  depending on the properties of the oscillator, e.g., its damping. This increase of the amplitude of the classical oscillator corresponds to the increase of  $\Delta t$  in (3.39) in the picture of virtual excitation, and it is qualitatively understandable that we will get the strongest deviations of  $\varepsilon(\omega)$  or  $\tilde{n}(\omega)$  from the vacuum value  $\varepsilon = \tilde{n} = 1$  in the vicinity of the resonance  $\omega_0$ . We elaborate this concept in detail in Chap. 4.

However, before doing so, we shall demonstrate how the various transitions shown in Fig. 3.16 and some others can be treated quantitatively in quantum mechanics by perturbation theory.

### 3.2.2 *Perturbative Treatment of the Linear Interaction of Light with Matter*

In this section we present first the Hamiltonian of the system elaborating the perturbation terms. Then we outline shortly how a perturbation causes transitions between various eigenstates. Finally we join these two things together ending with an understanding of the theoretical description of absorption and stimulated or spontaneous transitions.

The Hamiltonian of the total system consisting of the electron states in Fig. 3.16 of the two level atoms (or of the bands of the semiconductor), the radiation field and the interaction of these two systems can be written as

$$H = H_{\text{el}} + H_{\text{rad}} + H_{\text{interac}} . \quad (3.40)$$

In the picture of second quantization outlined in Sect. 2.5  $H_{\text{interac}}$  contains terms which describe, e.g., the annihilation of a photon and of an electron in the ground state and the creation of an electron in the excited state for the process shown in Fig. 3.16a, weighted with a factor which contains the transition matrix element.

The exact solution of the total Hamiltonian leads to the polariton concept (Chap. 5) in the field of linear optics and describes among other things the changes of the electronic states introduced by the presence of the radiation in nonlinear optics for which we will see some examples in Sects. 20.1–20.4.

For our present purposes we follow an approach which is widely used and which treats the radiation field as a small perturbation, that is, we assume that the eigenstates  $\varphi$  and the eigenenergies  $E_n$  of  $H_{\text{el}}$ , do not change much in the presence of the electromagnetic field, and that the eigenstates of  $H_{\text{rad}}$  are the photons described already in Sect. 2.5. The approximation which we use now is known as the semiclassical treatment of radiation. It consists of replacing the canonical conjugate momentum  $\mathbf{p}$  in the Hamilton function by

$$\mathbf{p} \rightarrow \mathbf{p} - e\mathbf{A} , \quad (3.41)$$

where  $\mathbf{A}$  is the vector potential (2.48).

If we replace  $\mathbf{p}$  by its operator

$$\mathbf{p} = \frac{\hbar}{i} \nabla \quad (3.42)$$

the single particle Hamiltonian reads

$$H = \frac{1}{2m} \left( \frac{\hbar}{i} \nabla - e\mathbf{A} \right)^2 + V(\mathbf{r}) \quad (3.43)$$

including any electrostatic potential into  $V(\mathbf{r})$ . Making use of the Coulomb gauge (2.49) we can evaluate (3.43) to obtain

$$H = -\frac{\hbar^2}{2m} \nabla^2 + V(\mathbf{r}) - \frac{e}{m} \mathbf{A} \frac{\hbar}{i} \nabla + \frac{e^2}{2m} \mathbf{A}^2 , \quad (3.44)$$

$$H = H_{\text{el}} - \frac{e}{m} \mathbf{A} \frac{\hbar}{i} \nabla + \frac{e^2}{2m} \mathbf{A}^2 , \quad (3.45)$$

$$H = H_{\text{el}} + H^{(1)} + H^{(2)} . \quad (3.46)$$

In (3.46) there are two perturbation terms  $H^{(1)}$  and  $H^{(2)}$ . If we assume that  $A$  and thus the light intensity are small, and in the regime of linear optics they are small by definition, then  $H^{(1)}$  is a perturbation term of first order and  $H^{(2)}$  is small of second order. Consequently  $H^{(1)}$  must be used in first-order perturbation theory. In the second-order approximation we have to use  $H^{(1)}$  in second-order perturbation theory and  $H^{(2)}$  in first-order perturbation theory, etc. We shall come back to this latter aspect in Chap. 19.

In order to arrive at Fermi's golden rule for the transition rate  $w_{ji}$  from an initial state  $i$  (e.g., the ground state  $g$  in Fig. 3.16) to another state  $j$  (e.g., the excited state  $ex$  in Fig. 3.16) one uses the time-dependent Schrödinger equation

$$H\psi = i\hbar \frac{\partial \psi}{\partial t} \quad (3.47a)$$

with the stationary solutions when  $H = H_0$

$$\psi_n(\mathbf{r}, t) = \varphi_n(\mathbf{r}) e^{-i(E_n/\hbar)t} . \quad (3.47b)$$

For the solution in the presence of a perturbation  $H^{(1)}$  we make the ansatz

$$\psi(\mathbf{r}, t) = \sum_n a_n(t) \varphi_n(\mathbf{r}) e^{-i(E_n/\hbar)t} . \quad (3.48)$$

We assume that the perturbation is switched on at  $t = 0$ . Before the system is in state  $i$ , i.e.,

$$\left. \begin{array}{l} a_i(t) = 1 \\ a_{n \neq i}(t) = 0 \end{array} \right\} \quad \text{for } t \leq 0 . \quad (3.49)$$

For  $t > 0$  the  $a_{n \neq i}(t)$  start to grow and under these conditions the transition rate  $w_{ij}$  of Fermi's golden rule becomes

$$w_{ij} = \frac{2\pi}{\hbar} \left| H_{ij}^{(1)} \right|^2 D(E) , \quad (3.50a)$$

where  $D(E)$  is the density of the final states modified by momentum conservation if applicable.  $H_{ij}^{(1)}$  is the transition matrix element given by

$$H_{ij}^{(1)} = \int \psi_j^*(\mathbf{r}) H^{(1)} \psi_i(\mathbf{r}) d\tau =: \langle \psi_j | H^{(1)} | \psi_i \rangle \quad (3.50b)$$

For a non-degenerate two level system  $D(E)$  is simply one per atom. The square of the transition matrix element  $|H_{ij}|^2$  is known as the transition probability. Later on we will assume, for simplicity of writing, that some constant factors as the term  $2\pi/\hbar$  are incorporated in this  $|H_{ij}|^2$ . Transition probabilities are given apart from some coefficients by the square of the respective transition matrix elements of (3.50) in the case of first order perturbation and by the terms (3.51) for second order.

The transition rate  $w_{ij}$  is proportional to the transition probability multiplied by the square of the amplitude of the perturbation  $H^{(1)}$ , i.e., here by  $|A_0|^2 \sim I$  where  $I$  is the light intensity, i.e. as already mentioned the energy flux per unit of area and time.

If the first-order perturbation term (3.50b) vanishes, then according to what we stated above the second-order contribution reads

$$w_{ij}^{(2)} = \frac{2\pi}{\hbar} \left| \sum_{k \neq i,j} \frac{H_{jk}^{(1)} H_{ki}^{(1)}}{E_i - E_k} + H_{ij}^{(2)} \right|^2 D(E) . \quad (3.51)$$

We restrict ourselves for the moment to the first order according to (3.50) and discuss the term  $H_{ij}^{(1)}$  in some more detail for the perturbations of (3.45). With the vector potential  $\mathbf{A}$

$$\mathbf{A} = \mathbf{A}_0 e^{i(\mathbf{k}\mathbf{r} - \omega t)} \quad (3.52a)$$

we find, e.g., for the absorption process (3.50)

$$\begin{aligned} w_{g \rightarrow \text{ex}} &= \frac{2\pi}{\hbar} \left| \frac{-e\hbar}{im} \mathbf{A}_0 \cdot \int \varphi_{\text{ex}}^*(\mathbf{r}) e^{i(E_{\text{ex}}/\hbar)t} \mathbf{e}_A e^{i(\mathbf{k}\mathbf{r} - \omega t)} \nabla \varphi_g(\mathbf{r}) e^{-i(E_g/\hbar)t} d\tau \right|^2 D(E_g + \hbar\omega) \\ &\propto |A_0 < \varphi_{\text{ex}} | H^{(1)} | \varphi_g >|^2 D(E_g + \hbar\omega) =: \\ &A_0^2 |H_{\text{eg}}^{(1)}|^2 D(E_g + \hbar\omega), \end{aligned} \quad (3.52b)$$

where  $\mathbf{e}_A$  is the unit vector in the direction of  $\mathbf{A}$ . We also give in (3.52) a generally used abbreviation for the integral. A significant transition rate occurs only if the time dependent exponential functions vanish, or, mathematically

$$E_{\text{ex}} - E_g - \hbar\omega = 0. \quad (3.53a)$$

This is again the law of energy conservation. If the  $\varphi_i$  and  $H^{(1)}$  have plane-wave character and are described by a wave vector  $\mathbf{k}$ , a similar argument results in  $\mathbf{k}$  conservation.

$$\hbar\mathbf{k}_{\text{ex}} - \hbar\mathbf{k}_g - \hbar\mathbf{k} = 0 \quad (3.53b)$$

This is not the case for the two-level atoms discussed here but is true for most of the eigenstates of crystalline semiconductors.

We see that the transition rate is proportional to  $A_0^2$ , and thus to the light intensity  $I = \langle S \rangle$  or the density of photons  $N_{\text{ph}}(\omega)$  in a certain mode:

$$w_{ij} \propto A_0^2 \left| H_{\text{eg}}^{(1)} \right|^2 \propto I \left| H_{\text{eg}}^{(1)} \right|^2 \propto N_{\text{ph}} \left| H_{\text{eg}}^{(1)} \right|^2 . \quad (3.54)$$

By partial integration using the fact that the eigenfunctions form an orthonormal set, or that the  $H^{(1)}$  is Hermitian adjoint, or by the argument of the microscopic reversibility of a transitions from state  $i \rightarrow j$  and from  $j \rightarrow i$  induced by some perturbation  $H^{(1)}$ , we find that

$$\left| \int \varphi_j^* H^{(1)} \varphi_i d\tau \right|^2 = \left| \int \varphi_i^* H^{(1)} \varphi_j d\tau \right|^2 . \quad (3.55)$$

We see that the probabilities for induced emission and absorption are the same and that the rates differ only by factors containing the number of atoms in the upper and lower states. This fact is the basis for the relation between the Einstein coefficients. See Problem 10 in Sect. 3.3. Spontaneous emission has to be treated in the sense mentioned above as emission stimulated by the zero field. We return to this aspect in a moment. First the interaction operator  $H^{(1)}$  should be simplified to reach the so-called dipole approximation.

We note that the radius of an atom ( $r \simeq 0.1 \text{ nm}$ ) and the distance between neighboring atoms in a solid ( $a \simeq 0.3 \text{ nm}$ ) are small compared to the wavelength in the visible ( $\lambda \simeq 500 \text{ nm}$ ). Therefore there is practically no phase shift of the electromagnetic radiation over one atom or between one atom and its neighbors. Thus we can expand the term  $e^{i\mathbf{k}\mathbf{r}}$  in (3.52) in a power series and stop after the constant term

$$e^{i\mathbf{k}\mathbf{r}} = 1 + \frac{i\mathbf{k}\mathbf{r}}{1!} + \frac{(i\mathbf{k}\mathbf{r})^2}{2!} + \dots \simeq 1 . \quad (3.56)$$

This is the first step towards the dipole approximation. It means, that the momentum of the photon  $\hbar\mathbf{k}$  in (3.53b) is neglected.

The matrix element  $H_{ij}^{(1)}$  still contains the momentum operator  $\mathbf{p} = \frac{\hbar}{i}\nabla$

$$H_{ij}^{(1)} \sim \langle \varphi_i | \mathbf{p} | \varphi_j \rangle =: \langle \mathbf{p}_{i,j} \rangle .$$

With the semiclassical relation

$$\frac{\hbar}{i}\nabla = \mathbf{p} = m\dot{\mathbf{r}} \quad (3.57)$$

and some arguments of plausibility (See e.g. [55S1] of Chap. 2), we find that

$$\int \varphi_j^* \frac{\hbar}{i}\nabla \varphi_i d\tau = m \frac{i}{\hbar} (E_i - E_j) \int \varphi_j^* \mathbf{r} \varphi_i d\tau = m\omega \int \varphi_j^* \mathbf{r} \varphi_i d\tau . \quad (3.58)$$

For a detailed derivation of this relation actually some knowledge of the wave functions is required. (For details see e.g. [55S1, 85G1, 92M1] of Chap. 2).

We note that in this so-called dipole approximation (3.56) to (3.59) the transition rate is given by



$$w_{ij} \sim I\omega^2 |\mathbf{e}_A \cdot \mathbf{e} \mathbf{r}_{ij}|^2 D(E) = I\omega^2 |H_{ij}^D|^2 D(E). \quad (3.59)$$

Where  $\mathbf{e}_A$  is the unit vector in the direction of  $\mathbf{A}$  and  $|H_{ij}^D|^2$  is just the expectation value of the dipole moment  $\mathbf{e} \mathbf{r}$  squared since the field amplitude squared appears already in the intensity of the light field  $I$ .

To avoid confusion we note that  $|H_{ij}^{(1)}|$  contains still the field strengths  $\mathbf{A}$  or  $\mathbf{E}$  and has the dimension of an energy while  $|H_{ij}^D|$  contains the dipole operator only.

This result can also be obtained in a more intuitive way if we remember that the energy of a dipole  $\mathbf{e} \mathbf{r}$  in an electric field  $\mathbf{E} = \dot{\mathbf{A}}$  is given by

$$\mathbf{e} \mathbf{r} \cdot \mathbf{E} = H^{(1)} \quad (3.60)$$

Using this approach in combination with (3.50) yields directly (3.59).

Alternatively it is shown with similar arguments as in (3.57) in Ref. [07M1] of Chap. 1 that the perturbation term  $-e \mathbf{p} \mathbf{A} / m$  can be replaced by  $\mathbf{e} \mathbf{r} \mathbf{E}$ . Compare the perturbation term of first order in (3.45) with (3.60).

From now on we will call  $|H_{ij}^D|^2$  the dipole-transition probability and the operator  $\mathbf{e} \mathbf{r}$  the dipole operator  $H^D$ .

This result can be obtained still in another way using for  $\mathbf{A}$  a suitable gauge different from the Coulomb gauge ( $\nabla \cdot \mathbf{A} = 0$ ) (see (2.49)) via  $\mathbf{A}' = \mathbf{A} + \nabla \chi$  and  $\Phi' = \Phi - \dot{\chi}$  where  $\chi$  is a scalar field with existing second derivative. It can be shown that  $\mathbf{A}'$  results in identical electric and magnetic fields as  $\mathbf{A}$ . The choice  $\chi(\mathbf{r}, t) = i\omega^{-1} \mathbf{r} \cdot \mathbf{E} = i\omega^{-1} \mathbf{r} \cdot \mathbf{E}_0 \exp[i(\mathbf{k} \mathbf{r} - \omega t)]$  results for  $H^{(1)}$  directly in (3.60).

Transitions using higher terms in (3.44), (3.51) and (3.56) correspond to quadrupole, octupole and higher order transitions.

To conclude this section, we calculate the net rate of the transitions shown in Fig. 3.16a–c.

We assume that we have a density of photons  $N_{\text{ph}}$  which populate only one mode in the sense used for the calculation of the density of states in Sect. 2.6 that is, all photons have the same wave vector  $\mathbf{k}$ , polarization  $\mathbf{e}_A$  and energy  $\hbar\omega$ .

Furthermore  $\hbar\omega$  fulfills the energy conservation law according to (3.54). The density of identical two-level atoms is  $N_A$  where a fraction  $\alpha_g$  is in the ground state and correspondingly  $(1 - \alpha_g)$  are in the excited state. The net rate of the change of  $N_{\text{ph}}$  with time is then given using ((3.54) and (3.59))

$$\frac{\partial N_{\text{ph}}}{\partial t} = -N_A \alpha_g N_{\text{ph}} |H_{g \rightarrow e}^D|^2 + N_A (1 - \alpha_g) (1 + N_{\text{ph}}) |H_{g \rightarrow e}^D|^2. \quad (3.61)$$

The first term on the r.h.s. describes the absorption of photons, the second one the spontaneous and stimulated emission in the factor  $(1 + N_{\text{ph}})$ .

From (3.55) we see that

$$\left| H_{g \rightarrow e}^D \right|^2 = \left| H_{e \rightarrow g}^D \right|^2 = |H^D|^2 \quad (3.62)$$

and hence

$$\frac{1}{|H^D|^2} \frac{\partial N_{\text{ph}}}{\partial t} = N_{\text{ph}} \cdot N_A(1 - 2\alpha_g) + N_A(1 - \alpha_g) . \quad (3.63)$$

The first term on the right-hand side depends linearly on  $N_{\text{ph}}$  and describes the net rate of absorption and stimulated emission. The second term gives the spontaneous emission since it is independent of  $N_{\text{ph}}$ .

There is net absorption for  $\alpha_g > 1/2$  (absorption coefficient  $\alpha(\omega) > 0$  or  $\partial N_{\text{ph}}/\partial t < 0$ ) and amplification or optical gain for  $\alpha_g < 1/2$ , i.e., for gain, more than half of the atoms have to be in the upper state. This situation cannot be reached in thermal equilibrium, but only under the influence of a suitable source of pump power. Usually one or more additional energy levels are required (three- and four-level lasers). This fact can be easily elucidated with the following argument. We start with a situation where all atoms are in the ground state i.e.,  $\alpha_g(t = 0) = 1$ . If we send for  $t > 0$  a photon field with frequency  $\omega$  fulfilling (3.52a) into the system we initially have absorption since  $\alpha(\omega)$  is given by

$$-\alpha(\omega) \sim N_A(1 - 2\alpha_g) . \quad (3.64)$$

With increasing time and pump power  $\alpha(\omega)$  decreases because  $\alpha_g$  decreases. For the situation  $\alpha_g = 1/2$  and an incoherently excited ensemble of atoms the absorption vanishes and the material becomes transparent. This means no more pumping is possible under (quasi-) stationary excitation to reach  $\alpha_g > 1/2$ . In Chaps. 21–23 we will reexamine the above considerations using the proper statistics introduced in Sect. 2.6. In Chaps. 23 and 27 we shall see further that without dephasing processes or for sufficiently intense pulses shorter than the dephasing time it is possible to drive the system periodically from the ground state to the completely inverted state ( $\alpha_g = 0$ ) and back again. This process is known as Rabi flopping.

### 3.3 Problems

1. Consider the interface between vacuum (or air) and glass ( $n = 1.45$ ) at a wavelength  $\lambda_{\text{vac}}$  of  $0.5 \mu\text{m}$ . Calculate for an angle of incidence  $\alpha_i = 45^\circ$  the incident, reflected, and transmitted wave vectors, and the transmitted and reflected intensities for both polarizations. Calculate Brewster's angle for the transition air→glass and glass→air and the angle for the onset of total internal reflection.
2. Find a piece of polarizing material (polaroid) and observe the light reflected from a nicely polished floor or scattered from the blue sky using different

orientations of the light propagation and of the polarization. Do not look into the sun! Try to explain your findings.

3. Play with a piece of clear calcite and the polaroid.
4. From Figs. 3.11 and 3.12 one can understand that a birefringent crystal can be used as a polarizer if the lateral diameter of the beams is smaller than their lateral displacement after the passage. Usually one uses slightly more complex arrangements known as Glan–Thomson or Taylor polarizers. Make yourself familiar with the way of operation of these optical components. What can happen if you use them under oblique incidence or with a di- or convergent light beam with large angle of aperture?
5. Derive the laws of reflection and refraction from the principle of Maupertins (see Fig. 3.4) and from momentum conservation (3.16). Does this law also hold if you kick a soccer ball against a wall?
6. Consider Fig. 3.9c. Explain in words why only in this diagram the three curves intersect, i.e., why zero absorption gives both the highest maxima and the lowest minima in reflectance.
7. Verify the energy conservation law (3.23a) with the help of (3.23b) and (3.18).
8. What is a quarter ( $\lambda/4$ ) or half ( $\lambda/2$ ) wave plate? How thick is it? What is the state of polarisation of the transmitted light beam, when the incident beam is linearly polarized? Does it depend on the orientation of the incident linear polarization with respect to the crystallographic axis. Make a simple sketch. (Generally one uses a uniaxial material and normal incidence for these devices.) What is a low order  $\lambda/4$  plate? Why are achromatic  $\lambda/4$  plates rather expensive?
9. Make a simple sketch to make yourself familiar with optical activity. Can you imagine, that the absorption can be different for  $\sigma^+$  and  $\sigma^-$  polarized light?
10. Find in a textbook the definition and meaning of Einstein's coefficients.
11. Inform yourself on time independent perturbation theory. The perturbed wave function contains in first order virtually excited states and the perturbed energy in second order. Inspect time dependent perturbation theory for virtual excitations. Derive Fermi's golden rule.
12. Verify some of the prominent features in Fig. 3.6 with (3.18), e.g., normal and grazing incidence,  $\alpha_B$  or  $\alpha_{\text{tot}}$ .

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