The homogeneous equation

$$\ddot{D}+\gamma_{ab}\dot{D}+(\omega_{ba}^2+\gamma_{ab}^2/4)D=0\,, \tag{2.81b}$$

which describes the atomic dipoles without the driving field ($\Omega_{ab}=0$), has the solution for weak damping ($\gamma_{ab}\ll\omega_{ba}$)

$$D(t)=D_0\,\mathrm{e}^{(-\gamma_{ab}/2)t}\cos\omega_{ba}t\,. \tag{2.82}$$

The inhomogeneous equation (2.81a) shows that the induced dipole moment of the atom interacting with a monochromatic radiation field behaves like a driven damped harmonic oscillator with $\omega_{ba}=(E_b-E_a)/\hbar$ for the eigenfrequency and $\gamma_{ab}=(\gamma_a+\gamma_b)$ for the damping constant oscillating at the driving field frequency ω.

Using the approximation $(\omega_{ba}+\omega)\simeq 2\omega$ and $\gamma_{ab}\ll\omega_{ba}$, which means weak damping and a close-to-resonance situation, we obtain solutions of the form

$$D=D_1\cos\omega t+D_2\sin\omega t\,, \tag{2.83}$$

where the factors D_1 and D_2 include the frequency dependence,

$$D_1=\frac{\Omega_{ab}(\omega_{ba}-\omega)}{(\omega_{ba}-\omega)^2+(\gamma_{ab}/2)^2}\,, \tag{2.84a}$$

$$D_2=\frac{\frac{1}{2}\Omega_{ab}\gamma_{ab}}{(\omega_{ab}-\omega)^2+(\gamma_{ab}/2)^2}\,. \tag{2.84b}$$

These two equations for D_1 and D_2 describe dispersion and absorption of the EM wave. The former is caused by the phase lag between the radiation field and the induced dipole oscillation, and the latter by the atomic transition from the lower level E_a to the upper level E_b and the resultant conversion of the field energy into the potential energy (E_b-E_a).

The macroscopic polarization $\boldsymbol{P}$ of a sample with N atoms/cm^3 is related to the induced dipole moment $\boldsymbol{D}$ by $\boldsymbol{P}=N\boldsymbol{D}$.

2.7.6 Interaction with Strong Fields

In the previous sections we assumed weak-field conditions where the probability of finding the atom in the initial state was not essentially changed by the interaction with the field. This means that the population in the initial state remains approximately constant during the interaction time. In the case of broadband radiation, this approximation results in a *time-independent transition probability*. Also the inclusion of weak-damping terms with $\gamma_{ab}\ll\omega_{ba}$ did not affect the assumption of a constant population in the initial state.

When intense laser beams are used for the excitation of atomic transitions, the weak-field approximation is no longer valid. In this section, we therefore consider the "strong-field case." The corresponding theory, developed by

Rabi, leads to a time-dependent probability of the atom being in either the upper or lower level. The representation outlined below follows that of [2.21].

We consider a monochromatic field of frequency ω and start from the basic equations (2.68) for the probability amplitudes in the rotating wave approximation with $\omega_{ba} = -\omega_{ab}$

$$\dot{a}(t) = \frac{\mathrm{i}}{2}\Omega_{ab}\,\mathrm{e}^{-\mathrm{i}(\omega_{ba}-\omega)t}b(t)\,, \tag{2.85a}$$

$$\dot{b}(t) = \frac{\mathrm{i}}{2}\Omega_{ab}\,\mathrm{e}^{+\mathrm{i}(\omega_{ba}-\omega)t}a(t)\,. \tag{2.85b}$$

Inserting the trial solution

$$a(t) = \mathrm{e}^{\mathrm{i}\mu t} \;\Rightarrow\; \dot{a}(t) = \mathrm{i}\mu\,\mathrm{e}^{\mathrm{i}\mu t}\,,$$

into (2.85a) yields

$$b(t) = \frac{2\mu}{\Omega_{ab}}\,\mathrm{e}^{\mathrm{i}(\omega_{ba}-\omega+\mu)t} \;\Rightarrow\; \dot{b}(t) = \frac{2\mathrm{i}\mu(\omega_{ba}-\omega+\mu)}{\Omega_{ab}}\,\mathrm{e}^{(\omega_{bu}-\omega+\mu)t}\,.$$

Substituting this back into (2.85b) gives the relation

$$2\mu(\omega_{ba}-\omega+\mu) = \Omega_{ab}^2/2\,.$$

This is a quadratic equation for the unknown quantity μ with the two solutions

$$\mu_{1,2} = -\frac{1}{2}(\omega_{ba}-\omega) \pm \frac{1}{2}\sqrt{(\omega_{ba}-\omega)^2+\Omega_{ab}^2}\,. \tag{2.86}$$

The general solutions for the amplitudes a and b are then

$$a(t) = C_1\,\mathrm{e}^{\mathrm{i}\mu_1 t} + C_2\,\mathrm{e}^{\mathrm{i}\mu_2 t}\,, \tag{2.87a}$$

$$b(t) = (2/\Omega_{ab})\,\mathrm{e}^{\mathrm{i}(\omega_{ba}-\omega)t}(C_1\mu_1\,\mathrm{e}^{\mathrm{i}\mu_1 t} + C_2\mu_2\,\mathrm{e}^{\mathrm{i}\mu_2 t})\,. \tag{2.87b}$$

With the initial conditions $a(0) = 1$ and $b(0) = 0$, we find for the coefficients

$$C_1 + C_2 = 1 \quad \text{and} \quad C_1\mu_1 = -C_2\mu_2\,,$$
$$\Rightarrow\; C_1 = -\frac{\mu_2}{\mu_1-\mu_2} \quad C_2 = +\frac{\mu_1}{\mu_1-\mu_2}\,.$$

From (2.86) we obtain $\mu_1\mu_2 = -\Omega_{ab}^2/4$. With the shorthand

$$\Omega = \mu_1 - \mu_2 = \sqrt{(\omega_{ba}-\omega)^2+\Omega_{ab}^2}\,,$$

we get the probability amplitude

$$b(t) = \mathrm{i}(\Omega_{ab}/\Omega)\,\mathrm{e}^{\mathrm{i}(\omega_{ba}-\omega)t/2}\sin(\Omega t/2)\,. \tag{2.88}$$

The probability $|b(t)|^2 = b(t)b^*(t)$ of finding the system in level E_b is then

$$|b(t)|^2 = (\Omega_{ab}/\Omega)^2\sin^2(\Omega t/2)\,, \tag{2.89}$$

where

$$\Omega = \sqrt{(\omega_{ba} - \omega)^2 + (\boldsymbol{D}_{ab} \cdot \boldsymbol{E}_0/\hbar)^2} \tag{2.90}$$

is called the general *"Rabi flopping frequency"* for the nonresonant case $\omega \neq \omega_{ba}$. Equation (2.89) reveals that the transition probability is a periodic function of time. Since

$$|a(t)|^2 = 1 - |b(t)|^2 = 1 - (\Omega_{ab}/\Omega)^2 \sin^2(\Omega t/2) , \tag{2.91}$$

the system oscillates with the frequency Ω between the levels E_a and E_b, where the level-flopping frequency Ω depends on the detuning $(\omega_{ba} - \omega)$, on the field amplitude E_0, and the matrix element D_{ab} (Fig. 2.20b).

The general Rabi flopping frequency Ω gives the frequency of population oscillation in a two-level system in an electromagnetic field with amplitude E_0.

Note: In the literature often the term "Rabi frequency" is restricted to the resonant case $\omega = \omega_{ba}$.

At resonance $\omega_{ba} = \omega$, and (2.89) and (2.91) reduce to

$$|a(t)|^2 = \cos^2(\boldsymbol{D}_{ab} \cdot \boldsymbol{E}_0 t/2\hbar) , \tag{2.92a}$$

$$|b(t)|^2 = \sin^2(\boldsymbol{D}_{ab} \cdot \boldsymbol{E}_0 t/2\hbar) . \tag{2.92b}$$

After a time

$$T = \pi\hbar/(\boldsymbol{D}_{ab} \cdot \boldsymbol{E}_0) = \pi/\Omega_{ab} , \tag{2.93}$$

the probability $|b(t)|^2$ of finding the system in level E_b becomes unity. This means that the population probability $|a(0)|^2 = 1$ and $|b(0)|^2 = 0$ of the initial system has been inverted to $|a(T)|^2 = 0$ and $|b(T)|^2 = 1$ (Fig. 2.22).

Radiation with the amplitude A_0, which resonantly interacts with the atomic system for exactly the time interval $T = \pi\hbar/(\boldsymbol{D}_{ab} \cdot \boldsymbol{E}_0)$, is called a π-*pulse* because it changes the phases of the probability amplitudes $a(t)$, $b(t)$ by π, see (2.87, 2.88).

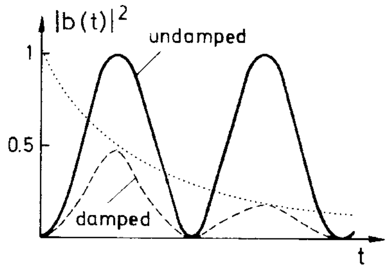

Fig. 2.22. Population probability $|b(t)|^2$ of the levels E_b altering with the Rabi flopping frequency due to the interaction with a strong field. The resonant case is shown without damping and with damping due to decay channels into other levels. The decaying curve represents the factor $\exp[-(\gamma_{ab}/2)t]$

We now include the damping terms γ_a and γ_b, and again insert the trial solution

$$a(t) = \mathrm{e}^{\mathrm{i}\mu t},$$

into (2.80a, 2.80b). Similar to the procedure used for the undamped case, this gives a quadratic equation for the parameter μ with the two complex solutions

$$\mu_{1,2} = -\frac{1}{2}\left(\omega_{ba} - \omega - \frac{\mathrm{i}}{2}\gamma_{ab}\right) \pm \frac{1}{2}\sqrt{\left(\omega_{ba} - \omega - \frac{\mathrm{i}}{2}\gamma\right)^2 + \Omega_{ab}^2},$$

where

$$\gamma_{ab} = \gamma_a + \gamma_b \quad \text{and} \quad \gamma = \gamma_a - \gamma_b\,. \tag{2.94}$$

From the general solution

$$a(t) = C_1\,\mathrm{e}^{\mathrm{i}\mu_2 t} + C_2\,\mathrm{e}^{\mathrm{i}\mu_2 t},$$

we obtain from (2.80a) with the initial conditions $|a(0)|^2 = 1$ and $|b(0)|^2 = 0$ the transition probability

$$|b(t)|^2 = \frac{\Omega_{ab}^2\,\mathrm{e}^{(-\gamma_{ab}/2)t}[\sin(\Omega/2)t]^2}{(\omega_{ba} - \omega)^2 + (\gamma/2)^2 + \Omega_{ab}^2}\,. \tag{2.95}$$

This is a damped oscillation (Fig. 2.22) with the damping constant $\frac{1}{2}\gamma_{ab} = (\gamma_a + \gamma_b)/2$, the Rabi flopping frequency

$$\Omega = \mu_1 - \mu_2 = \sqrt{\left(\omega_{ba} - \omega + \tfrac{\mathrm{i}}{2}\gamma\right)^2 + \Omega_{ab}^2}, \tag{2.96}$$

and the envelope $\Omega_{ab}^2\,\mathrm{e}^{-(\gamma_{ab}/2)t}/[(\omega_{ba} - \omega)^2 + (\gamma/2)^2 + \Omega_{ab}^2]$. The spectral profile of the transition probability is Lorentzian (Sect. 3.1), with a half-width depending on $\gamma = \gamma_a - \gamma_b$ and on the strength of the interaction. Since $\Omega_{ab}^2 = (\boldsymbol{D}_{ab} \cdot \boldsymbol{E}_0/\hbar)^2$ is proportional to the intensity of the electromagnetic wave, the linewidth *increases* with increasing intensity (saturation broadening, Sect. 3.5). Note, that $|a(t)|^2 + |b(t)|^2 < 1$ for $t > 0$, because the levels a and b can decay into other levels.

In some cases the two-level system may be regarded as isolated from its environment. The relaxation processes then occur only between the levels $|a\rangle$ and $|b\rangle$, but do not connect the system with other levels. This implies $|a(t)|^2 + |b(t)|^2 = 1$. Equation (2.80) then must be modified as

$$\dot{a}(t) = -\frac{1}{2}\gamma_a a(t) + \frac{1}{2}\gamma_b b(t) + \frac{\mathrm{i}}{2}\Omega_{ab}\,\mathrm{e}^{-\mathrm{i}(\omega_{ba}-\omega)t} b(t)\,, \tag{2.97a}$$

$$\dot{b}(t) = -\frac{1}{2}\gamma_b b(t) + \frac{1}{2}\gamma_a a(t) + \frac{\mathrm{i}}{2}\Omega_{ab}\,\mathrm{e}^{+\mathrm{i}(\omega_{ba}-\omega)t} a(t)\,. \tag{2.97b}$$

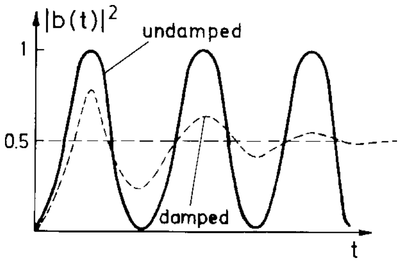

Fig. 2.23. Population of level $|b\rangle$ for a closed two-level system where the relaxation channels are open only for transitions between $|a\rangle$ and $|b\rangle$

The trial solution $a = \exp(\mathrm{i}\mu t)$ yields, for the resonance case $\omega = \omega_{ba}$, the two solutions

$$\mu_1 = \frac{1}{2}\Omega_{ab} + \frac{\mathrm{i}}{2}\gamma_{ab}\,, \quad \mu_2 = -\frac{1}{2}\Omega_{ab}\,,$$

and for the transition probability $|b(t)|^2$, one obtains with $|a(0)|^2 = 1$, $|b(0)|^2 = 0$ a damped oscillation that approaches the steady-state value

$$|b(t=\infty)|^2 = \frac{1}{2}\frac{\Omega_{ab}^2 + \gamma_a\gamma_b}{\Omega_{ab}^2 + (\frac{1}{2}\gamma_{ab})^2}\,. \tag{2.98}$$

This is illustrated in Fig. 2.23 for the special case $\gamma_a = \gamma_b$ where $|b(\infty)|^2 = 1/2$, which means that the two levels become equally populated.

For a more detailed treatment see [2.21–2.24].

2.7.7 Relations Between Transition Probabilities, Absorption Coefficient, and Line Strength

In this section we will summarize important relations between the different quantities discussed so far.

The absorption coefficient $\alpha(\omega)$ for a transition between levels $|i\rangle$ and $|k\rangle$ with population densities N_i and N_k and statistical weights g_i, g_k is related to the absorption cross section $\sigma_{ik}(\omega)$ by

$$\alpha(\omega) = [N_i - (g_i/g_k)N_k]\sigma_{ik}(\omega)\,. \tag{2.99}$$

The Einstein coefficient for absorption B_{ik} is given by

$$B_{ik} = \frac{c}{\hbar\omega}\int_0^\infty \sigma_{ik}(\omega)\,\mathrm{d}\omega = \frac{c\,\overline{\sigma}_{ik}}{\hbar\omega}\int_0^\infty g(\omega-\omega_0)\,\mathrm{d}\omega \tag{2.100}$$

where $g(\omega-\omega_0)$ is the line profile of the absorbing transition at center frequency ω_0. The transition probability per second according to (2.15) is then

$$P_{ik} = B_{ik}\cdot\varrho = \frac{c}{\hbar\omega\cdot\Delta\omega}\int \varrho(\omega)\cdot\sigma_{ik}(\omega)\,\mathrm{d}\omega\,, \tag{2.101}$$

where $\Delta\omega$ is the spectral linewidth of the transition.

The line strength S_{ik} of a transition is defined as the sum

$$S_{ik} = \sum_{m_i, m_k} \left|D_{m_i, m_k}\right|^2 = |D_{ik}|^2 , \tag{2.102}$$

over all dipole-allowed transitions between all subcomponents m_i, m_k of levels $|i\rangle$, $|k\rangle$. The oscillator strength f_{ik} gives the ratio of the power absorbed by a molecule on the transition $|i\rangle \to |k\rangle$ to the power absorbed by a classical oscillator on its eigenfrequency $\omega_{ik} = (E_k - E_i)/h$.

Some of these relations are compiled in Table 2.2.

Table 2.2. Relations between the transition matrix element D_{ik} and the Einstein coefficients A_{ik}, B_{ik}, the oscillator strength f_{ik}, the absorption cross section σ_{ik}, and the line strength S_{ik}. The numerical values are obtained, when λ [m], B_{ik} [$\mathrm{m^3 s^{-2} J^{-1}}$], D_{ik} [As m], m_c [kg]

$A_{ki} = \frac{1}{g_k} \frac{16\pi^3 \nu^3}{3\varepsilon_0 h c^3} \|D_{ik}\|^2$	$B_{ik}^{(\nu)} = \frac{1}{g_i} \frac{2\pi^2}{3\varepsilon_0 h^2} \|D_{ik}\|^2$	$B_{ik}^{(\omega)} = \frac{1}{g_i} \frac{\pi}{3\varepsilon_0 \hbar^2} \|D_{ik}\|^2$
$= \frac{2.82 \times 10^{45}}{g_k \cdot \lambda^3} \|D_{ik}\|^2\, \mathrm{s}^{-1}$	$= 6 \times 10^{31}\, \lambda^3 \frac{g_i}{g_k} A_{ki}$	$= \frac{g_k}{g_i} B_{ki}$
$f_{ik} = \frac{1}{g_i} \frac{8\pi^2 m_e \nu}{e^2 h} \|D_{ik}\|^2$	$S_{ik} = \|D_{ik}\|^2$	$\sigma_{ik} = \frac{1}{\Delta\nu} \frac{2\pi^2 \nu}{3\varepsilon_0 c h g_i} \cdot S_{ik}$
$= \frac{g_k}{g_i} \cdot 4.5 \times 10^4 \lambda^2 A_{ki}$	$= (7.8 \times 10^{-21} g_i \lambda)\, f_{ik}$	$B_{ik} = \frac{c}{h\nu} \int_0^\infty \sigma_{ik}(\nu)\, \mathrm{d}\nu$

2.8 Coherence Properties of Radiation Fields

The radiation emitted by an extended source S generates a total field amplitude A at the point P that is a superposition of an infinite number of partial waves with the amplitudes A_n and the phases ϕ_n emitted from the different surface elements dS (Fig. 2.24), i.e.,

$$A(P) = \sum_n A_n(P)\, \mathrm{e}^{\mathrm{i}\phi n(P)} = \sum_n \left[A_n(0)/r_n^2\right] \mathrm{e}^{\mathrm{i}(\phi_{n0} + 2\pi r_n/\lambda)} , \tag{2.103}$$

where $\phi_{n0}(t) = \omega t + \phi_n(0)$ is the phase of the nth partial wave at the surface element dS of the source. The phases $\phi_n(r_n, t) = \phi_{n,0}(t) + 2\pi r_n/\lambda$ depend on the distances r_n from the source and on the angular frequency ω.

If the phase differences $\Delta\phi_n = \phi_n(P, t_1) - \phi_n(P, t_2)$ at a *given point* P between two different times t_1, t_2 are nearly the same for all partial waves, the radiation field at P is *temporally coherent*. The maximum time interval $\Delta t = t_2 - t_1$ for which $\Delta\phi_n$ for all partial waves differ by less than π is termed the *coherence time* of the radiation source. The path length $\Delta s_c = c\Delta t$ traveled by the wave during the coherence time Δt is the *coherence length*.