Chapter 3
Field-Matter Coupling and Two-Level Systems

With this chapter, we start the applications part of this book by considering the interaction between lasers and matter. Lasers have already been discussed in Chap. 1. Therefore, we begin immediately with the theoretical description of the coupling of a given classical light field realized, e.g., by a laser, to a quantum mechanical system.[1] After the discussion of different gauges or frames, related by unitary transformations, the Volkov solution for the laser-driven free particle is reviewed.

Due to their simplicity and the fact that they serve as paradigms for many phenomena observed in more complex systems, some analytically solvable two-level systems will be discussed in the remainder of this chapter. We will first look at Rabi oscillations mediated by a static electric field and after the introduction of the rotating wave approximation, the laser-driven case will be reviewed.

3.1 Light-Matter Interaction

The interaction of a single quantum particle with an electromagnetic field shall be considered in the following. Starting from the principle of minimal coupling and using several unitary transformations, some commonly used ways of setting up a field driven Hamiltonian will be presented.

[1] In the literature this is frequently called semiclassical laser matter interaction [1]. We have, however, used the expression "semiclassics" already differently in Chap. 2.

F. Grossmann, *Theoretical Femtosecond Physics*, Graduate Texts in Physics,
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3.1.1 Minimal Coupling

The most straightforward approach to the coupling of a charged particle with charge q to an electromagnetic field is given by the principle of "minimal coupling". In classical mechanics this principle aims at producing Newton's equation with the Lorentz force by constructing a corresponding Lagrangian.

3.1. *Study classical minimal coupling by answering the following questions:*

(a) *Under which conditions for the potentials* $\boldsymbol{A}$ *and* Φ *does the classical Lagrangian*

$$L(\dot{\boldsymbol{r}}, \boldsymbol{r}, t) = \frac{m}{2}\dot{\boldsymbol{r}}^2 - q\Phi(\boldsymbol{r}, t) + q\dot{\boldsymbol{r}} \cdot \boldsymbol{A}(\boldsymbol{r}, t)$$

lead to Newton's equation of motion with the Lorentz force?

(b) *Give explicit expressions for the canonical momentum* $\boldsymbol{p} = \partial L/\partial\dot{\boldsymbol{r}}$ *and for the mechanical momentum* $\boldsymbol{p}_{\mathrm{m}} = m\dot{\boldsymbol{r}}$.

(c) *What is the explicit form of the Hamiltonian* $H(\boldsymbol{p}, \boldsymbol{r}, t) = \dot{\boldsymbol{r}} \cdot \boldsymbol{p} - L(\dot{\boldsymbol{r}}, \boldsymbol{r}, t)$*?*

To arrive at the quantum version of minimal coupling, we could just use the classical result and invoke the correspondence principle. More instructive is a direct approach to quantum minimal coupling, however, which shall be discussed in some detail now.

Let us first consider the effect of a local unitary transformation with the scalar field $\chi(\boldsymbol{r}, t)$

$$\Psi'(\boldsymbol{r}, t) = \mathrm{e}^{\mathrm{i}\frac{q}{\hbar}\chi(\boldsymbol{r},t)}\Psi(\boldsymbol{r}, t) \tag{3.1}$$

on the time-dependent Schrödinger equation [2]. For the transformed wavefunction the transformed equation

$$\mathrm{i}\hbar\dot{\Psi}'(\boldsymbol{r}, t) = \hat{H}'\Psi'(\boldsymbol{r}, t) \tag{3.2}$$

holds, where the primed Hamiltonian is given by

$$\hat{H}' = \mathrm{e}^{\mathrm{i}\frac{q}{\hbar}\chi(\boldsymbol{r},t)}\hat{H}\mathrm{e}^{-\mathrm{i}\frac{q}{\hbar}\chi(\boldsymbol{r},t)} - q\dot{\chi}(\boldsymbol{r}, t), \tag{3.3}$$

with

$$\hat{H} = \frac{\hat{\boldsymbol{p}}^2}{2m} + V(\boldsymbol{r}). \tag{3.4}$$

Shifting the momentum operator $\hat{\boldsymbol{p}} = \frac{\hbar}{\mathrm{i}}\nabla$ twice past the exponential factor of the unitary transformation, we get the identity

$$\mathrm{e}^{\mathrm{i}\frac{q}{\hbar}\chi(\boldsymbol{r},t)}\hat{\boldsymbol{p}}^2\mathrm{e}^{-\mathrm{i}\frac{q}{\hbar}\chi(\boldsymbol{r},t)}\Psi'(\boldsymbol{r}, t) = (\hat{\boldsymbol{p}} - q\nabla\chi)^2\Psi'(\boldsymbol{r}, t), \tag{3.5}$$

and therefore

$$\hat{H}' = \frac{1}{2m}\left(\frac{\hbar}{\mathrm{i}}\nabla - q\nabla\chi\right)^2 + V(\boldsymbol{r}) - q\frac{\partial\chi}{\partial t} \tag{3.6}$$

holds for the primed Hamiltonian.

Following Weyl [3], the time-dependent Schrödinger equation has to be invariant under the unitary transformation (3.1) introduced above. To satisfy this requirement, the original time-dependent Schrödinger equation has to be modified slightly, however, according to

$$\mathrm{i}\hbar\dot{\Psi}(\boldsymbol{r},t) = \left[\frac{1}{2m}\left(\frac{\hbar}{\mathrm{i}}\nabla - q\boldsymbol{A}(\boldsymbol{r},t)\right)^2 + V(\boldsymbol{r}) + q\Phi(\boldsymbol{r},t)\right]\Psi(\boldsymbol{r},t). \tag{3.7}$$

This equation is now formally equivalent to the transformed time-dependent Schrödinger equation

$$\mathrm{i}\hbar\dot{\Psi}'(\boldsymbol{r},t) = \left[\frac{1}{2m}\left(\frac{\hbar}{\mathrm{i}}\nabla - q\boldsymbol{A}'(\boldsymbol{r},t)\right)^2 + V(\boldsymbol{r}) + q\Phi'(\boldsymbol{r},t)\right]\Psi'(\boldsymbol{r},t) \tag{3.8}$$

if the relations

$$\boldsymbol{A}' = \boldsymbol{A} + \nabla\chi, \qquad \Phi' = \Phi - \dot{\chi} \tag{3.9}$$

hold. These, however, are the gauge transformations of the potentials $\boldsymbol{A}(\boldsymbol{r},t)$ and $\Phi(\boldsymbol{r},t)$ of classical electrodynamics. The electromagnetic fields

$$\boldsymbol{\mathcal{E}} = -\frac{\partial\boldsymbol{A}}{\partial t} - \nabla\Phi, \tag{3.10}$$

$$\boldsymbol{B} = \nabla\times\boldsymbol{A} \tag{3.11}$$

are unchanged by such transformations.

Summarizing, minimal coupling amounts to replacing the canonical momentum $\hat{\boldsymbol{p}}$ by the kinetic momentum $\hat{\boldsymbol{p}} - q\boldsymbol{A}(\boldsymbol{r},t)$ and shifting the potential by $q\Phi(\boldsymbol{r},t)$ in the Hamiltonian. The probability current density in the equation of continuity (2.4) of Chap. 2 has to be changed accordingly, as can be seen by solving Exercise 3.2.

3.2. *Find the modified expression for the probability current density $\boldsymbol{j}$ in the case of coupling of the motion of a charged particle to an external field. Show that the expression you gained is gauge invariant.*

Expanding the square of the kinetic momentum, cross terms of the form $\hat{\boldsymbol{p}}\cdot\boldsymbol{A}$ and $\boldsymbol{A}\cdot\hat{\boldsymbol{p}}$ appear. In the Coulomb gauge, which, for sources at infinity, is defined by

$$\Phi(\boldsymbol{r},t) = 0, \qquad \nabla\cdot\boldsymbol{A}(\boldsymbol{r},t) = 0\,, \tag{3.12}$$

we can conclude that $\hat{\boldsymbol{p}}$ commutes with $\boldsymbol{A}$. Therefore the two cross-terms are identical as in classical mechanics.

Furthermore, the energy obviously is not conserved any more in the presence of a time-dependent external field. The question, which operator could be considered as the energy operator, does not have a straightforward answer, however. Studying Exercise 3.3 and the literature given in the solutions sheds more light on this question.

3.3. *Let $\hat{\Theta}(\boldsymbol{A}, \Phi)$ be an operator that depends on the potentials of the electromagnetic field.*

(a) *Show that for the operator $\hat{\Theta}$ to have a gauge invariant expectation value*

$$\mathrm{e}^{\mathrm{i}\frac{q}{\hbar}\chi}\hat{\Theta}(\boldsymbol{A}, \Phi)\mathrm{e}^{-\mathrm{i}\frac{q}{\hbar}\chi} = \hat{\Theta}(\boldsymbol{A}', \Phi')$$

has to hold.

(b) *Show that $\hat{H} = \frac{(\hat{\boldsymbol{p}}-q\boldsymbol{A})^2}{2m} + V(\boldsymbol{r}) + q\Phi$ is not a gauge invariant operator and its expectation value cannot be the energy. Discuss an alternative, that may be considered as the energy operator.*

In general, the case of a system of many charged particles which are coupled to a laser field has to be studied. As we will see in Chap. 5, the motion of the center of mass and the relative motion without a laser can be separated. With the laser they do not necessarily separate any more [3]. We will deal with the coupling of an electromagnetic field to a many particle system in more detail in Chap. 5.

3.1.2 Dipole Approximation and Length Gauge

Another well-known form of light matter interaction rests on the dipole approximation, in which case the vector potential is assumed to be independent of position.[2] For an atom of typical size of the order of Angstroms in a field of optical wavelength of several hundred nanometers this is a well founded approximation, as depicted in Fig. 3.1.

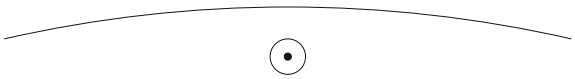

Fig. 3.1 An atom in the field of a light wave with wavelength much longer than the typical extension of the atom

[2]Therefore, due to (3.11), the magnetic induction vanishes in dipole approximation.

The effect of the vector potential in the minimal coupling Hamiltonian of (3.7) in Coulomb gauge and in dipole approximation is a time-dependent shift of the momentum leading to

$$\hat{H}_{\mathrm{v}} = \frac{[\hat{\boldsymbol{p}} - q\boldsymbol{A}(t)]^2}{2m} + V(\boldsymbol{r}). \tag{3.13}$$

Due to the corresponding Lagrangian (vector potential couples to velocity), the term velocity gauge is therefore frequently used.[3] Applying a gauge transformation with the scalar field

$$\chi(\boldsymbol{r}, t) = -\boldsymbol{r} \cdot \boldsymbol{A}(t) \tag{3.14}$$

leads to transformed potentials of the form

$$\boldsymbol{A}' = 0, \qquad \Phi' = -\frac{\partial \chi}{\partial t} = \boldsymbol{r} \cdot \dot{\boldsymbol{A}} = -\boldsymbol{r} \cdot \boldsymbol{\mathcal{E}}(t), \tag{3.15}$$

where the last step follows from (3.10) in Coulomb gauge. The present gauge is thus also called length gauge (electric field couples to the position). The corresponding time-dependent Schrödinger equation then reads

$$\mathrm{i}\hbar\dot{\Psi}_{\mathrm{l}}(\boldsymbol{r}, t) = \left[\frac{\hat{\boldsymbol{p}}^2}{2m} + V(\boldsymbol{r}) - q\,\boldsymbol{r} \cdot \boldsymbol{\mathcal{E}}(t)\right] \Psi_{\mathrm{l}}(\boldsymbol{r}, t) \tag{3.16}$$

and contains the laser-matter interaction in terms of the dipole operator $q\,\hat{\boldsymbol{r}}$. Historically it has been introduced by Göppert-Mayer [5] by using the fact that the Lagrangians in the length as well as in the velocity gauge only differ by a total time-derivative.

3.4. *Switch from the velocity to the length gauge by adding a total time derivative to the Lagrangian.*

(a) *Show first that adding a total time-derivative* $\frac{\mathrm{d}}{\mathrm{d}t} f(\boldsymbol{r}, t)$ *to the Lagrangian does not alter the equations of motion.*
(b) *In the dipole approximation and the Coulomb gauge* ($\Phi = 0, \boldsymbol{A} = \boldsymbol{A}(t)$) *add* $-q\frac{\mathrm{d}}{\mathrm{d}t}(\boldsymbol{r} \cdot \boldsymbol{A})$ *to the velocity gauge Lagrangian and simplify the resulting expression.*

We stress that in dipole approximation and under the Coulomb gauge, the vector potential as well as the electric field are independent of the position vector and the magnetic induction is zero. A coupling to the magnetic field by going beyond the

[3] Synonymously, some authors [3, 4] use the expression $\boldsymbol{A} \cdot \boldsymbol{p}$ gauge.

dipole approximation would become necessary for a large electron quiver velocity (see Sect. 3.1.4) on the order of the speed of light.[4]

Velocity and length gauge are related by a unitary transformation and therefore any measurable quantity may not depend on the gauge used. If one uses approximations during the solution process, however, there may be orders of magnitude and even qualitative [7] differences between the results predicted in the different gauges. A recent investigation of a gauge independent strong field approximation is given in [8]. A more technical question is, in which gauge numerical calculations should be performed. For the investigation of high-order harmonic generation using laser irradiated hydrogen atoms, to be discussed in Sect. 4.5, it was found that for high laser intensities, the velocity gauge seems to be favorable from a numerical perspective [9]. This fact was corroborated in a recent publication on the ionization of hydrogen atoms, see the supplemental material of [10]. The case of very short (down to half cycle) pulses is discussed with respect to gauge invariance in [11].

Finally, it is worthwhile to note that, as shown in Appendix 3.A, the notion of parity, well-known in autonomous Hamiltonian systems, can be generalized to the case of periodically, dipolarly driven systems.

3.1.3 Kramers-Henneberger Transformation

In the case of strong fields, another unitary transformation will turn out to be very useful. We start again from the minimally coupled time-dependent Schrödinger equation (3.7) in the Coulomb gauge and in dipole approximation, leading to

$$\begin{aligned} \mathrm{i}\hbar\dot{\Psi}_{\mathrm{v}}(\boldsymbol{r},t) &= \left[\frac{1}{2m}\left(\frac{\hbar}{\mathrm{i}}\boldsymbol{\nabla}-q\boldsymbol{A}(t)\right)^2+V(\boldsymbol{r})\right]\Psi_{\mathrm{v}}(\boldsymbol{r},t) \\ &= \left[-\frac{\hbar^2}{2m}\boldsymbol{\nabla}^2+\frac{\mathrm{i}q\hbar}{m}\boldsymbol{A}(t)\cdot\boldsymbol{\nabla}+\frac{q^2}{2m}\boldsymbol{A}^2(t)+V(\boldsymbol{r})\right]\Psi_{\mathrm{v}}(\boldsymbol{r},t). \end{aligned} \tag{3.17}$$

Successively performing two unitary transformations

$$\Psi_{\mathrm{a}}(\boldsymbol{r},t)=\hat{U}_2\hat{U}_1\Psi_{\mathrm{v}}(\boldsymbol{r},t), \tag{3.18}$$

with

$$\hat{U}_1=\exp\left\{\frac{\mathrm{i}q^2}{2m\hbar}\int_0^t \mathrm{d}t'\boldsymbol{A}^2(t')\right\}, \tag{3.19}$$

$$\hat{U}_2=\exp\left\{-\frac{q}{m}\int_0^t \mathrm{d}t'\boldsymbol{A}(t')\cdot\boldsymbol{\nabla}\right\} \tag{3.20}$$

[4] For a charged particle in a plane electromagnetic wave, the magnetic part of the Lorentz force is smaller by a factor v/c than the electric one [6].

defines a wavefunction in the Kramers-Henneberger frame [12, 13]. The use of the subscript "a" will become apparent below. The first transformation eliminates the squared vector potential, whereas the displacement operator (3.20) moves the coupling into the argument of the potential, as can be seen by solving Exercise 3.5. The time-dependent Schrödinger equation in the Kramers-Henneberger frame is then given by

$$\mathrm{i}\hbar\dot{\Psi}_\mathrm{a}(\boldsymbol{r},t) = \left[-\frac{\hbar^2}{2m}\Delta + V[\boldsymbol{r}+\boldsymbol{\alpha}(t)]\right]\Psi_\mathrm{a}(\boldsymbol{r},t)\ , \tag{3.21}$$

where

$$\boldsymbol{\alpha}(t) = -\frac{q}{m}\int_0^t \mathrm{d}t'\boldsymbol{A}(t'). \tag{3.22}$$

3.5. *Show that the two unitary transformations into the Kramers-Henneberger frame eleminate the terms proportional to* $\boldsymbol{A}^2$ *and* $\boldsymbol{A}$ *in the Hamiltonian. Due to the fact that the first transformation is a global phase transformation, it just remains to calculate*

$$\hat{U}_2\hat{V}\hat{U}_2^{-1},$$

to prove the shift in the argument of the potential.
Hint: Use the operator relation known as Baker-Haussdorff (or Hadamard) lemma $\mathrm{e}^{\hat{L}}\hat{M}\mathrm{e}^{-\hat{L}} = \sum_{n=0}^{\infty}\frac{1}{n!}[\hat{L},\hat{M}]_n$*, where* $[\hat{L},\hat{M}]_n = [\hat{L},[\hat{L},\hat{M}]_{n-1}]$ *and* $[\hat{L},\hat{M}]_0 = \hat{M}$*.*

Differentiating (3.22) twice and using (3.10) in the Coulomb gauge, we find that

$$m\ddot{\boldsymbol{\alpha}}(t) = q\boldsymbol{\mathcal{E}} \tag{3.23}$$

holds. The Kramers-Henneberger transformation thus is characterized by a spatial translation into an accelerated frame, corresponding to the oscillatory quiver motion of the charged particle in the electric field. The present case is therefore also frequently referred to as the "acceleration gauge", although the use of the term gauge is misleading, since no gauge transformations of the potentials can be given here [11].

In the high-frequency limit, the TDSE in the Kramers-Henneberger frame can be averaged over a (short) period of the external field. This has, e.g., been done in the calculations in [14] for the case of a periodically driven double-well potential, to be discussed in more detail in Sect. 5.5.1. This way, analytical predictions of the influence of high-frequency driving on the system dynamics can be given.

3.1.4 Volkov Wavepacket and Ponderomotive Energy

To fill the presented formalism with life, we now turn to an important, exactly solvable model. For the case of 1D free motion, i.e., $V(x) = 0$ of an electron with mass m_e and charge $q = -e$ in a cw laser field $\mathcal{E} = \mathcal{E}_0 \cos(\omega t)$, the time-dependent Schrödinger equation in length gauge (3.16) can be solved exactly analytically under the assumption of a Gaussian initial state.

Due to the fact that the total time-dependent potential

$$V_\mathrm{L}(x,t) = ex\mathcal{E}_0 \cos(\omega t) \tag{3.24}$$

is linear, the resulting Volkov wavepacket with initial phase space center $(0, q_0)$ is given by using the GWD of Sect. 2.1.4 for $\alpha_0 = \gamma/2$ according to[5]

$$\Psi(x,t) = \left(\frac{\gamma}{\pi}\right)^{1/4} \sqrt{\frac{1}{1+\mathrm{i}\gamma\hbar t/m_\mathrm{e}}} \exp\left\{\frac{\mathrm{i}}{\hbar}\left[\frac{U_\mathrm{p}}{2\omega}\sin(2\omega t) - U_\mathrm{p} t + x p(t)\right]\right\}$$
$$\exp\left\{-\frac{\gamma}{2(1+\mathrm{i}\gamma\hbar t/m_\mathrm{e})}[x-q(t)]^2\right\}, \tag{3.25}$$

where the general solutions

$$p(t) = p_0 - e\mathcal{E}_0 \sin(\omega t)/\omega, \tag{3.26}$$
$$q(t) = q_0 + \frac{p_0 t}{m_\mathrm{e}} + e\mathcal{E}_0[\cos(\omega t) - 1]/(m_\mathrm{e}\omega^2) \tag{3.27}$$

of the classical equations of motion for position and momentum have been used with the initial conditions $p(0) = p_0 = 0, q(0) = q_0$.

The amplitude of oscillations of position $e\mathcal{E}_0/(m_e\omega^2)$ is the so-called quiver amplitude. We can convince ourselves of the analytic form of the Volkov solution by solving Exercise 3.6. As a side result it will turn out that the derivative of the kinetic energy averaged over a period of the external field vanishes. A free particle can therefore not accumulate energy from the field.

3.6. *Using Gaussian Wavepacket Dynamics calculate the wavefunction of a free electron in a laser field with the potential*

$$V_\mathrm{L}(x,t) = ex\mathcal{E}_0 \cos(\omega t)$$

in length gauge.

[5] The gauge index will be mostly suppressed in the remainder of the book, as we will explicitly state which gauge is used.

(a) *Determine the solutions* (p_t, q_t) *of the classical equations of motion with the initial conditions* $(0, q_0)$*. Then calculate the classical kinetic energy and its derivative and average the results over one period* $T = 2\pi/\omega$ *of the external field. Interprete the results.*
(b) *Use the result for* α_t *from the free particle case (why is this possible?).*
(c) *Employing integration by parts, show that*

$$\int_0^t \mathrm{d}t' L = -\int_0^t \mathrm{d}t' \frac{p_{t'}^2}{2m_\mathrm{e}} + q_t p_t$$

holds. Use this result to determine the phase $\delta_t = \int_0^t \mathrm{d}t'(L - \alpha_{t'})$ *and insert everything in the GWD expression. Why is the final result exact?*

As could be seen by working through the previous exercise, there is an important quantity hidden in the Volkov solution. This is the average of the kinetic energy over one period, which is given by

$$U_\mathrm{p} := \frac{1}{T}\int_0^T \mathrm{d}t \frac{p^2}{2m_\mathrm{e}} = \frac{e^2\mathcal{E}_0^2}{4m_\mathrm{e}\omega^2}, \tag{3.28}$$

as can be shown by using (3.26). This quantity is called ponderomotive energy (or ponderomotive potential). $2U_\mathrm{p}$ is the maximal kinetic energy that an electron may have at a certain time. It is important to keep in mind that low frequency fields lead to high ponderomotive energies.

A generalization of the results to 3D can be performed in several ways. Alternatively to replacing all occurences of position and momentum in the solution above by the corresponding 3D vectors, we start from the TDSE in velocity gauge with the Hamiltonian of (3.13) in the case of $V(\boldsymbol{r}) = 0$. After a Fourier transformation of the wavefunction to the momentum representation (see also Sect. 2.3.2) via

$$\Psi(\boldsymbol{p}) = \frac{1}{(2\pi\hbar)^{3/2}}\int \mathrm{d}^3 r \exp\left\{-\frac{\mathrm{i}}{\hbar}\boldsymbol{p}\cdot\boldsymbol{r}\right\}\Psi(\boldsymbol{r}), \tag{3.29}$$

we get

$$\mathrm{i}\hbar\dot{\Psi}(\boldsymbol{p}, t) = \frac{1}{2m_\mathrm{e}}\left[\boldsymbol{p} + e\boldsymbol{A}(t)\right]^2\Psi(\boldsymbol{p}, t) \tag{3.30}$$

for the time-dependent Schrödinger equation. There is no operator hat on the momentum any more because we have used the corresponding eigenvalue equation.

A solution to the ordinary differential equation above is given by

$$\Psi(\boldsymbol{p}, t) = \Psi(\boldsymbol{p}, 0)\exp\left\{-\frac{\mathrm{i}}{\hbar}\int_0^t \mathrm{d}t' \frac{\left[\boldsymbol{p} + e\boldsymbol{A}(t')\right]^2}{2m_\mathrm{e}}\right\}, \tag{3.31}$$

as can be proven by separation of variables and checked by differentiation with respect to time. Inverse Fourier transformation back to position space now gives the Volkov wavepacket

$$\Psi(\boldsymbol{r},t) = \frac{1}{(2\pi\hbar)^{3/2}} \int \mathrm{d}^3 p \ \exp\left\{\frac{\mathrm{i}}{\hbar}\boldsymbol{p}\cdot\boldsymbol{r}\right\} \exp\left\{-\frac{\mathrm{i}}{\hbar}\int_0^t \mathrm{d}t' \frac{\left[\boldsymbol{p}+e\boldsymbol{A}(t')\right]^2}{2m_\mathrm{e}}\right\} \Psi(\boldsymbol{p},0). \tag{3.32}$$

If we use as a special case a delta function centered around $\boldsymbol{p}_0$,

$$\Psi(\boldsymbol{p},0) = \delta(\boldsymbol{p}-\boldsymbol{p}_0), \tag{3.33}$$

for the initial $\boldsymbol{p}$ state, then the Volkov state

$$\Psi_\mathrm{v}(\boldsymbol{r},t) = \frac{1}{(2\pi\hbar)^{3/2}} \exp\left\{\frac{\mathrm{i}}{\hbar}\boldsymbol{p}_0\cdot\boldsymbol{r} - \frac{\mathrm{i}}{\hbar}\int_0^t \mathrm{d}t' \frac{\left[\boldsymbol{p}_0+e\boldsymbol{A}(t')\right]^2}{2m_\mathrm{e}}\right\} \tag{3.34}$$

in velocity gauge emerges.

Starting from the velocity gauge result and using the gauge transformation from (3.14) leads to

$$\Psi_\mathrm{l}(\boldsymbol{r},t) = \frac{1}{(2\pi\hbar)^{3/2}} \exp\left\{\frac{\mathrm{i}}{\hbar}[\boldsymbol{p}_0+e\boldsymbol{A}(t)]\cdot\boldsymbol{r} - \frac{\mathrm{i}}{\hbar}\int_0^t \mathrm{d}t' \frac{\left[\boldsymbol{p}_0+e\boldsymbol{A}(t')\right]^2}{2m_\mathrm{e}}\right\}, \tag{3.35}$$

which is the Volkov state in length gauge.

In the Kramers-Henneberger frame, starting again from (3.34), the dependence on $\boldsymbol{A}(t)$ cancels out and we arrive at the corresponding Volkov state

$$\Psi_\mathrm{a}(\boldsymbol{r},t) = \frac{1}{(2\pi\hbar)^{3/2}} \exp\left\{\frac{\mathrm{i}}{\hbar}\boldsymbol{p}_0\cdot\boldsymbol{r} - \frac{\mathrm{i}}{\hbar}\frac{\boldsymbol{p}_0^2}{2m_\mathrm{e}}t\right\}, \tag{3.36}$$

which is just a plane wave.

A more general initial state, e.g., a Gaussian centered around $\boldsymbol{p}_0$, will lead to a generalization of the GWD solution to 3D. It is a nice exercise in Gaussian integration to rederive the 1D result (3.25) from (3.32).

3.2 Analytically Solvable Two-Level Problems

Driven two-level systems are the easiest realizations of the field-matter coupling formalism just reviewed. Several paradigms in the theory of laser induced dynamics can be found already in the solutions of these simple systems. They shall therefore now be studied in some detail. We will concentrate on analytically solvable cases, which can either be solved exactly or under some approximations.

3.2.1 Dipole Matrix Element

First of all, the Hamilton matrix has to be set up. To this end, we consider two energy levels with the unperturbed orthogonal states $|\psi_1\rangle, |\psi_2\rangle$ and the corresponding energies $E_1 = -\hbar\epsilon$, $E_2 = \hbar\epsilon$, which are the diagonal elements of the Hamilton matrix.

To write down an expression for the off-diagonal elements of the Hamilton matrix in the case of an external perturbation, we assume that it is due to the coupling to an electric field of the form

$$\boldsymbol{\mathcal{E}}(\boldsymbol{r}, t) = \boldsymbol{\mathcal{E}}_0 \cos(\boldsymbol{k} \cdot \boldsymbol{r} - \omega t). \tag{3.37}$$

We now turn to the dipole approximation of Sect. 3.1.2, i.e., $\lambda = 2\pi/k$ shall be much larger than the size of the quantum system, as depicted in Fig. 3.1. In the argument of the cosine, $\boldsymbol{r}$ can then be replaced by $\boldsymbol{r}_0$ which can be set to zero without loss of generality. The electric field is then purely time-dependent

$$\boldsymbol{\mathcal{E}}(t) = \boldsymbol{\mathcal{E}}_0 \cos(\omega t) \tag{3.38}$$

and the coordinate independent force

$$\boldsymbol{F}(t) = -e\,\boldsymbol{\mathcal{E}}(t) \tag{3.39}$$

acts on the electron. The corresponding potential energy is given by

$$V_{\mathrm{L}}(\boldsymbol{r}, t) = e\,\boldsymbol{r} \cdot \boldsymbol{\mathcal{E}}(t). \tag{3.40}$$

Adding this potential energy to the Hamiltonian leads to the length gauge form of the Hamiltonian in (3.16).

If the two levels under consideration have real eigenfunctions with different parity (see Exercise 3.8) then

$$\hbar\nu_{12}(t) \equiv \boldsymbol{\mathcal{E}}(t) \cdot \int \mathrm{d}^3 r\; \psi_1\, e\,\boldsymbol{r}\,\psi_2 = \boldsymbol{\mu}_{12} \cdot \boldsymbol{\mathcal{E}}(t) = \hbar\nu_{21}(t) \tag{3.41}$$

follows for the non-vanishing off-diagonal elements of the Hamilton matrix, which are proportional to the matrix element

$$\boldsymbol{\mu}_{12} \equiv \int \mathrm{d}^3 r \; \psi_1 \, e \, \boldsymbol{r} \psi_2 \tag{3.42}$$

of the (negated) dipole operator. The diagonal matrix elements due to the laser potential are zero.

3.2.2 Rabi Oscillations Induced by a Constant Perturbation

For the following, we assume that the perturbation is time-independent, i.e., we set $\omega \to 0$ and define $\nu := \lim_{\omega \to 0} \nu_{12}(t)$. As an Ansatz for the solution of the time-dependent Schrödinger equation (2.22), a superposition of the unperturbed eigenstates with time-dependent coefficients

$$|\Psi(t)\rangle = c_1(t)|\psi_1\rangle + c_2(t)|\psi_2\rangle \tag{3.43}$$

can be chosen. For the vector $\boldsymbol{c}^{\mathrm{T}} = (c_1, c_2)$ of coefficients, the linear system of coupled ordinary differential equations

$$\mathrm{i}\hbar \dot{\boldsymbol{c}} = \mathbf{H}\boldsymbol{c} \; , \tag{3.44}$$

with the two by two Hamilton matrix

$$\mathbf{H} = \hbar \begin{pmatrix} -\epsilon & \nu \\ \nu & \epsilon \end{pmatrix} \tag{3.45}$$

emerges. This Hamiltonian can be expressed in terms of the Pauli spin matrices, which are discussed in Appendix 3.B.

As mentioned in Sect. 2.3.1, the time-evolution can be determined by solving the eigenvalue problem. The eigenvalues of the matrix in (3.45) are

$$E_{\pm} = \pm\hbar\sqrt{\epsilon^2 + \nu^2} \tag{3.46}$$

and the corresponding eigenstates are given by

$$|\psi_+\rangle = \sin(\Theta)|\psi_1\rangle + \cos(\Theta)|\psi_2\rangle, \tag{3.47}$$

$$|\psi_-\rangle = \cos(\Theta)|\psi_1\rangle - \sin(\Theta)|\psi_2\rangle, \tag{3.48}$$

where the definition

$$\Theta \equiv \frac{1}{2} \arctan\left(\frac{\nu}{\epsilon}\right) \tag{3.49}$$

has been used.[6] In the case of degeneracy of the unperturbed states ($\epsilon = 0$) $\Theta = \pi/4$, and the eigenstates are the symmetric, respectively antisymmetric combination of the two unperturbed states.

The spectral representation of the time-evolution operator for the solution of the Schrödinger equation is given by

$$\hat{U}(t,0) = \sum_{\pm} |\psi_{\pm}\rangle \exp\left\{-\frac{\mathrm{i}}{\hbar} E_{\pm} t\right\} \langle\psi_{\pm}|, \tag{3.50}$$

as can be seen by comparison with (2.39). In the basis of the eigenvectors (1,0) and (0,1) of the unperturbed Hamilton matrix, the matrix

$$\begin{aligned} \mathbf{U}(t,0) = &\begin{pmatrix} \sin^2(\Theta) & \sin(\Theta)\cos(\Theta) \\ \sin(\Theta)\cos(\Theta) & \cos^2(\Theta) \end{pmatrix} \exp\left\{-\frac{\mathrm{i}}{\hbar} E_{+} t\right\} \\ &+ \begin{pmatrix} \cos^2(\Theta) & -\sin(\Theta)\cos(\Theta) \\ -\sin(\Theta)\cos(\Theta) & \sin^2(\Theta) \end{pmatrix} \exp\left\{-\frac{\mathrm{i}}{\hbar} E_{-} t\right\} \end{aligned} \tag{3.51}$$

for the time-evolution operator can be derived. This matrix allows us to calculate

$$P_{21}(t) = |\langle\psi_2|\hat{U}(t,0)|\psi_1\rangle|^2 = |U_{21}(t,0)|^2, \tag{3.52}$$

which is the probability to find the system in state $|\psi_2\rangle$ at time t, if it was in state $|\psi_1\rangle$ at time zero (in terms of the coefficients this corresponds to the initial conditions $c_1(0) = 1, c_2(0) = 0$). From the matrix representation of $\hat{U}$ by using $\sin\Theta\cos\Theta = \frac{1}{2}\sin(2\Theta)$, we get

$$P_{21}(t) = \frac{\nu^2}{\nu^2 + \epsilon^2} \sin^2(\Omega_{\mathrm{R}} t/2), \tag{3.53}$$

where

$$\Omega_{\mathrm{R}} \equiv 2\sqrt{\epsilon^2 + \nu^2} \tag{3.54}$$

is the definition of the so-called Rabi frequency. $P_{21}(t)$ performs Rabi oscillations with the amplitude

[6] The identities $\arctan(x) = \arccos(1/\sqrt{1+x^2})$ and $\arctan(x) = \arcsin(x/\sqrt{1+x^2})$ can be used to resolve the cosine and sine terms in (3.47) and (3.48).

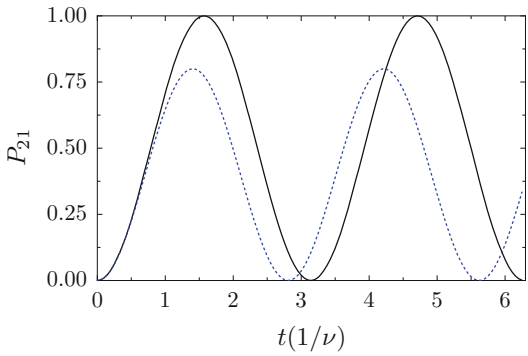

Fig. 3.2 Rabi oscillations of the probability to be in the upper state, starting from the lower state, induced by the perturbation $\nu = 1$. The degenerate $\epsilon = 0$ (*solid black line*), as well as the non-degenerate case $\epsilon = 0.5$ (*dashed blue line*) are depicted as a function of time in units of $1/\nu$; all energies in arbitrary units

$$A = \frac{\nu^2}{\nu^2 + \epsilon^2}, \tag{3.55}$$

which are depicted in Fig. 3.2. Only in case of degeneracy, $\epsilon = 0$, do the oscillations have an amplitude of 1. Furthermore, for non-degenerate systems the oscillations are faster than for degenerate unperturbed levels.

Rabi oscillations are analogous to the tunneling dynamics of the probability amplitude in a symmetric double well, which will be considered in Chap. 5. There the eigenstates of the unperturbed problem are the symmetric, respectively antisymmetric superposition of *localized* states in the left and right well and thus for a localized initial condition $c_1 = \pm c_2 = 1/\sqrt{2}$ has to be chosen.

3.2.3 *Time-Dependent Perturbations and Rotating Wave Approximation*

In the presence of a time-dependent perturbation $\hat{V}(t) = \hbar\hat{\nu}(t)$, and with $E_{1,2} = \hbar\epsilon_{1,2}$, the time-dependent Schrödinger equation for the coefficients is

$$\mathrm{i}\dot{c}_1 = c_1\epsilon_1 + c_2\nu_{12}(t), \tag{3.56}$$
$$\mathrm{i}\dot{c}_2 = c_2\epsilon_2 + c_1\nu_{21}(t). \tag{3.57}$$

In the "strong-coupling" limit, i.e., for $\nu_{21} \gg \epsilon_2 - \epsilon_1$, these coupled differential equations can be solved perturbatively [15]. There exists, however, another approximate approach to solve the differential equations, starting from the Ansatz

$$c_1(t) = d_1(t)\exp[-\mathrm{i}\epsilon_1 t], \tag{3.58}$$
$$c_2(t) = d_2(t)\exp[-\mathrm{i}\epsilon_2 t], \tag{3.59}$$

which leads to

$$\mathrm{i}\dot{d}_1 = d_2 v_{12}(t)\exp[-\mathrm{i}\omega_{21}t], \tag{3.60}$$

$$\mathrm{i}\dot{d}_2 = d_1 v_{21}(t)\exp[\mathrm{i}\omega_{21}t], \tag{3.61}$$

where the abbreviation $\omega_{21} = \epsilon_2 - \epsilon_1$ has been introduced. Note that the transformation from the vector $\boldsymbol{c}$ to the vector $\boldsymbol{d}$ of coefficients is equivalent to a unitary transformation into the interaction picture.

In the case of a monochromatic (coherent) perturbation $v_{12}(t) \sim \cos(\omega t)$, the system of differential equations can be solved analytically by using the so-called rotating wave approximation, as will be shown in the following. In the case of interaction with incoherent radiation (a random superposition of monochromatic laser fields) we can use perturbation theory and in this way give a microscopic derivation of the B-coefficient of Chap. 1. This last case will be dealt with in Appendix 3.C.

3.2.3.1 Rotating Wave Approximation

For the monochromatic field in (3.38), the Schrödinger equation in the interaction picture (3.60) and (3.61) can be written as

$$\mathrm{i}\dot{d}_1 = d_2 \frac{\boldsymbol{\mu}_{12}\cdot\boldsymbol{\mathcal{E}}_0}{2\hbar}\left\{\exp[\mathrm{i}(\omega-\omega_{21})t] + \exp[-\mathrm{i}(\omega+\omega_{21})t]\right\}, \tag{3.62}$$

$$\mathrm{i}\dot{d}_2 = d_1 \frac{\boldsymbol{\mu}_{21}\cdot\boldsymbol{\mathcal{E}}_0}{2\hbar}\left\{\exp[-\mathrm{i}(\omega-\omega_{21})t] + \exp[\mathrm{i}(\omega+\omega_{21})t]\right\}. \tag{3.63}$$

In order to introduce the rotating wave approximation (RWA), we define the detuning between the field and the external frequency

$$\Delta_\mathrm{d} \equiv \omega - \omega_{21}. \tag{3.64}$$

For $\Delta_\mathrm{d} \ll \omega_{21}$, the terms that oscillate at about twice the frequency of the external field are the so-called counter-rotating terms. In the differential equations above they can be neglected, if we assume that the coefficients $d_{1,2}$ change on a much longer time scale. To prove this procedure mathematically, one has to average the differential equations over times much larger than $1/(\omega+\omega_{21})$, see Exercise 3.7.

The differential equations in RWA are now dramatically simplified and read

$$\mathrm{i}\dot{d}_1 = d_2 \frac{\mu\mathcal{E}_0}{2\hbar}\exp[\mathrm{i}\Delta_\mathrm{d}t], \tag{3.65}$$

$$\mathrm{i}\dot{d}_2 = d_1 \frac{\mu\mathcal{E}_0}{2\hbar}\exp[-\mathrm{i}\Delta_\mathrm{d}t], \tag{3.66}$$

where we have assumed in addition that the polarization of the field is in the direction of the dipole matrix element, which has the absolute value $\mu = \mu_{12} = \mu_{21}$. The

solution of the two coupled differential equations can be found by differentiating (3.65) with respect to time and inserting (3.66). The second order differential equation that emerges can be solved, and one gets

$$d_1(t) = \frac{\hbar}{\mu\mathcal{E}_0}\exp[\mathrm{i}\Delta_\mathrm{d}t/2]\{(\Delta_\mathrm{d}-\Omega_\mathrm{R})C\exp[\mathrm{i}\Omega_\mathrm{R}t/2] + (\Delta_\mathrm{d}+\Omega_\mathrm{R})D\exp[-\mathrm{i}\Omega_\mathrm{R}t/2]\}, \tag{3.67}$$

$$d_2(t) = \exp[-\mathrm{i}\Delta_\mathrm{d}t/2]\{C\exp[\mathrm{i}\Omega_\mathrm{R}t/2] + D\exp[-\mathrm{i}\Omega_\mathrm{R}t/2]\} \tag{3.68}$$

with the Rabi frequency in the time-dependent case

$$\Omega_\mathrm{R} = \sqrt{\Delta_\mathrm{d}^2 + \left(\frac{\mu\mathcal{E}_0}{\hbar}\right)^2}\,. \tag{3.69}$$

The parameters C and D have to be determined from the initial conditions. In the case of non-resonance (corresponding to the non-degenerate case for constant perturbations) the oscillations are again faster than on resonance.

3.7. *Consider a two-level system interacting with a monochromatic laser field.*

(a) *Average the TDSE over times long in comparison to* $1/(\omega+\omega_{21})$ *in order to motivate neglecting the counter-rotating terms.*

(b) *Using the initial conditions* $d_1(0)=1$ *and* $d_2(0)=0$*, give explicit expressions for* C *and* D *and for* $d_1(t)$ *and* $d_2(t)$*. Depict* $|d_2(t)|^2$ *for resonance as well as for off-resonance.*

Furthermore, the quality of the RWA depends on the soundness of the approximation of neglecting the counter-rotating terms. The validity of this assumption can be studied explicitly for a specific example in Exercise 3.8.

3.8. *An electron shall move in an inversion symmetric potential* $V(x)=V(-x)$ *in one spatial dimension.*

(a) *Show that the eigenfunctions of the TISE must fulfill either*

$$\psi_{2n}(x)=\psi_{2n}(-x) \quad \text{or} \quad \psi_{2n+1}(x)=-\psi_{2n+1}(-x),$$

and that diagonal dipole matrix elements $\mu_{nn}=\langle\psi_n|e\hat{x}|\psi_n\rangle$ *therefore always vanish.*

(b) *Calculate the dipole matrix element between the ground and the first excited state of the harmonic oscillator*

$$V(x)=\frac{1}{2}m_\mathrm{e}\omega_\mathrm{e}^2x^2$$

with a frequency in the visible range, $\omega_e = 3.14 \times 10^{15}\,s^{-1}$. Determine the Rabi frequency in the resonance case for 3 different field strengths $\mathcal{E}_0 = 1, 10^6, 10^{10}\,V\,m^{-1}$. Is the condition for the applicability of the RWA fulfilled for all field strengths?

3.2.3.2 Area Theorem

Finally, we consider the case of resonance, $\Delta_d = 0$, in which $\hbar$ times the external frequency equals the level spacing. Furthermore, we assume that the external field shall be of finite duration, i.e., it shall consist of a laser pulse with an envelope, so that we have to replace $\mathcal{E}_0$ by $\mathcal{E}_0 f(t)$ in the time-dependent Schrödinger equation. According to (3.69), a time-dependent Rabi frequency

$$\Omega_R(t) = \frac{\mu \mathcal{E}_0 f(t)}{\hbar} \tag{3.70}$$

emerges, with the help of which the coupled differential equations can be written as

$$i\dot{d}_1 = \frac{\Omega_R(t)}{2} d_2, \tag{3.71}$$

$$i\dot{d}_2 = \frac{\Omega_R(t)}{2} d_1. \tag{3.72}$$

For the initial conditions $d_1(0) = 1, d_2(0) = 0$ the solutions are given by

$$d_1(t) = \cos\left(\int_0^t dt' \frac{\Omega_R(t')}{2}\right), \tag{3.73}$$

$$d_2(t) = -i \sin\left(\int_0^t dt' \frac{\Omega_R(t')}{2}\right), \tag{3.74}$$

as can be verified by insertion. In RWA the population transfer in the resonance case does not depend on the specific shape of the pulse, but only on the area below the pulse. This is the so-called area theorem. A π-pulse, for which $\int_0^t dt' \Omega_R(t') = \pi$, allows for a complete transfer of population.

3.2.4 Exactly Solvable Time-Dependent Cases

In very few special cases, also in the case of a time-dependent perturbation an exact analytical solution of the time-dependent two-level Schrödinger equation can be found [16]. As our starting point we use (3.44) in the case of time-dependent ϵ and ν. After elimination of c_1, the second order differential equation

$$\ddot{c}_2 - \frac{\dot{\nu}}{\nu}\dot{c}_2 + \left(\epsilon^2 + \nu^2 - \mathrm{i}\dot{\epsilon} + \mathrm{i}\epsilon\frac{\dot{\nu}}{\nu}\right)c_2 = 0 \tag{3.75}$$

for the other coefficient can be derived, see also Exercise 3.9. Reasons for the time-dependence of the diagonal as well as for the off-diagonal elements of the Hamiltonian may be the coupling to a light field or nuclear motion in a molecule, which will be considered in detail in Chap. 5.

3.2.4.1 The Rosen-Zener Model

In the case of coupling to a pulsed laser field and in RWA[7] we can choose

$$\epsilon = \Delta/2, \qquad \nu(t) = \nu_0 \mathrm{sech}(t/T_\mathrm{p}), \tag{3.76}$$

defining the Rosen-Zener model with a pulse length parameter T_p, see Fig. 3.3. As found by these authors, the solution of the time-dependent Schrödinger equation for this choice can be determined exactly analytically. With the initial condition $c_1(-\infty) = 1$ and for $t \to \infty$ it is given by [17]

$$|c_2(\infty)|^2 = \sin^2(\pi\nu_0 T_\mathrm{p})\mathrm{sech}^2(\pi\Delta T_\mathrm{p}/2). \tag{3.77}$$

For the resonance case, $\Delta = 0$, this solution is proven in Exercise 3.9. In the resonance case it is also rewarding to note that in the argument of the sine, the pulse area $\nu_0 \int_{-\infty}^{\infty} \mathrm{d}t\ \mathrm{sech}(t/T_\mathrm{p}) = \nu_0 \pi T_\mathrm{p}$ appears. This is yet another manifestation of the area theorem discussed at the end of Sect. 3.2.3.

3.9. *Consider the TDSE for the two-level Rosen-Zener model.*

(a) *Prove the equation for c_2 that can be gained by the elimination of c_1.*
(b) *Transform the independent variable with the help of*

$$z = \frac{1}{2}(\tanh\frac{t}{T_\mathrm{p}} + 1).$$

What is the differential equation for $c_2(z)$?
(c) *Consider the special case $\epsilon = 0$ and determine $c_2(t = \infty)$ for the initial conditions $c_2(t = -\infty) = 0$ and $c_1(-\infty) = 1$.*
Hint: Use the hypergeometric function (see I. S. Gradshteyn and I. M. Rhyzik, Tables of Integrals Series and Products (Academic Press, San Diego, 1994), Sect. 9.1, or http://dlmf.nist.gov/15) and
$F(a, b; c; 1) = \frac{\Gamma(c)\Gamma(c-a-b)}{\Gamma(c-a)\Gamma(c-b)}$, $\Gamma(1-x)\Gamma(x) = \frac{\pi}{\sin(\pi x)}$, $\Gamma(3/2) = \sqrt{\pi/2}$

[7] This is an approximation and therefore the notion of exact solubility refers to the final equation and not the initial problem.

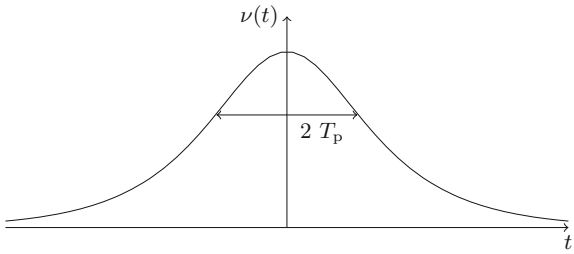

Fig. 3.3 Laser pulse envelope and width parameter T_p for the Rosen-Zener model

3.2.4.2 The Landau-Zener Model

Apart from the Rosen-Zener case, an exact analytic solution is available in the case of a linear time-dependence

$$\epsilon(t) = \lambda t, \qquad \nu(t) = \nu_0 \tag{3.78}$$

of the diagonal terms. This kind of time-dependence can, e.g., be induced in a scattering experiment, by nuclear motion in a molecule, or by time-dependent magnetic fields applied to atoms. The model has been investigated by Landau [18], as well as by Zener [19], Stückelberg [20], and Majorana [21]. An asymptotic solution of (3.75) for the initial condition $c_1(-\infty) = 1$ is given by

$$|c_2(\infty)|^2 = 1 - \exp[-2\pi\gamma], \tag{3.79}$$

where $\gamma = \nu_0^2/|2\lambda|$. The expression for γ can be further specified in molecular theory and leads to the celebrated Landau-Zener formula [22].

We note that for an infinitely fast change of the energy, i.e., $\lambda \to \infty$, it follows that $\gamma \to 0$ and no population will be transferred, i.e., $|c_2(\infty)|^2 \to 0$. On the contrary in the case of $\lambda \to 0$, one finds $\gamma \to \infty$, yielding complete population transfer, i.e., $|c_2(\infty)|^2 \to 1$.

3.3 Notes and Further Reading

Minimal Coupling and Gauge Transformations
Unitary (gauge) transformations of the wavefunction are discussed in depth in [2, 3]. The theory of minimal coupling and the different gauges or frames for field-matter interaction are at least partly covered in the books just mentioned as well as in many other quantum theoretical textbooks, see, e.g. [4]. Schleich's book [3] focuses on the subtleties arising from the inclusion of center of mass motion and contains an appendix, dealing with terms beyond the dipole approximation.

The inclusion of magnetic field coupling is discussed in [23] and in [24]. In [23] an intensity/wavelength diagram is presented that delineates regions, where a fully relativistic treatment (including order $(v/c)^2$ terms) is needed from those, where (magnetic) terms of order v/c have to be taken into account and those, where the inclusion of the electric field only is sufficient. A similar discussion can be found on page 868 of [4]. For the example of a Ti:sapphire laser with 800 nm wavelength, the intensity above which magnetic field effects can become important is $10^{16}\mathrm{W/cm}^2$. An insightful discussion of the gauge invariant calculation of expectation values and probabilities is given in [25].

Two-Level Systems

Our formulation of the interaction of two-level systems with coherent and incoherent (see Appendix 3.C) light is based on the presentation in Haken's book [26]. A landmark paper in this field is the one by Shirley [27], treating the periodically driven two-level problem in Floquet theory. The Rosen-Zener model is a special case of the first Demkov-Kunike model, which is discussed in the appendix of [28].

The theory of two-level systems interacting with magnetic fields has not been dealt with here but is covered in the book on photon-atom interactions by Weissbluth [29]. This book is also a treasure-house, if one is interested in the effect of damping on the dynamics of a two-level system. The wide field of dissipative quantum systems is usually described in a density matrix formulation (see also Appendix 3.B). More details on that exciting field can be found in the books by Weiss [30] and by Breuer and Petruccione [31].

The laser field is considered to be a classical field throughout the rest of this book. In quantum optics, where the light field is treated quantum mechanically, the RWA can also be performed, and if applied to a driven two-level system, this is known as the Jaynes-Cummings model, which is treated in detail in the book by Schleich [3].

3.A Generalized Parity Transformation

In the case of a symmetric static potential $V(x) = V(-x)$ and in length gauge, with a sinusoidal laser potential of the form $e\mathcal{E}_0 x \sin(\omega t)$, the extended Hamiltonian $\hat{\mathcal{H}}$ in (2.138) is invariant under the generalized parity transformation

$$\hat{\mathcal{P}}: \qquad x \to -x, \quad t \to t + \frac{T}{2} . \tag{3.80}$$

The Floquet functions thus transform according to

$$\hat{\mathcal{P}}\psi_{\alpha'}(x,t) = \pm\psi_{\alpha'}(x,t) , \tag{3.81}$$

i.e., they have either positive or negative generalized parity. With the help of (2.150) it follows that $\psi_{\alpha'}(x,t)$, $\psi_{\beta'}(x,t)$ have the same or different generalized parity, depending on $(\alpha - k) - (\beta - l)$ being even or odd.

As we will see in Chap. 5, exact crossings of the quasienergies as a function of external parameters are of utmost importance for the quantum dynamics of periodically driven systems. For stationary systems, the possibility of exact crossings has been studied in the heyday of quantum theory by von Neumann and Wigner [32]. These authors found that eigenvalues of eigenfunctions with different parity may approach each other arbitrarily closely and may thus cross exactly. This is in contrast to eigenvalues of the same parity, which always have to be at a finite distance, a fact which is sometimes referred to as the non-crossing rule. The corresponding behavior in the spectrum as a function of external parameters is called allowed, respectively avoided crossing. In the Floquet case, the Hamiltonian can also be represented by a Hermitian matrix, see e.g. (2.183), and therefore the same reasoning applies, with parity replaced by generalized parity.

For the investigations to be presented in Sect. 5.5.1 it is decisive if these *exact* crossings are singular events in parameter space or if they can occur by variation of just a single parameter. In [32] it has been shown that for Hermitian matrices (of finite dimension) with complex (real) elements, the variation of three (two) free parameters is necessary in order for two eigenvalues to cross. Using similar arguments, it can be shown that for a real Hermitian matrix with alternatingly empty off-diagonals (as it is e.g., the case for the Floquet matrix of the periodically driven, quartic, symmetric, bistable potential) the variation of a single parameter is enough to make two quasienergies cross.

In the case of *avoided* crossings an interesting behavior of the corresponding eigenfunctions can be observed. There is a continuous change in the structure in position space if one goes through the avoided crossing [33]. Pictorially this is very nicely represented in the example of the driven quantum well, depicted in Fig. 3.4, taken out of [34], where for reasons of better visualization the Husimi transform of the quasi-eigenfunctions as a function of action angle variables (J, Θ) [35] is shown.

3.B Pauli Spin Matrices and the Two-Level Density Matrix

The Pauli spin matrices

$$\boldsymbol{\sigma}_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}; \ \boldsymbol{\sigma}_y = \begin{pmatrix} 0 & -\mathrm{i} \\ \mathrm{i} & 0 \end{pmatrix}; \ \boldsymbol{\sigma}_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{3.82}$$

together with the 2×2 unit matrix, form a complete basis in the space of complex hermitian 2×2 matrices. In their terms our Hamiltonian (3.45) reads

$$\mathbf{H} = \hbar v \boldsymbol{\sigma}_x - \hbar \epsilon \boldsymbol{\sigma}_z. \tag{3.83}$$

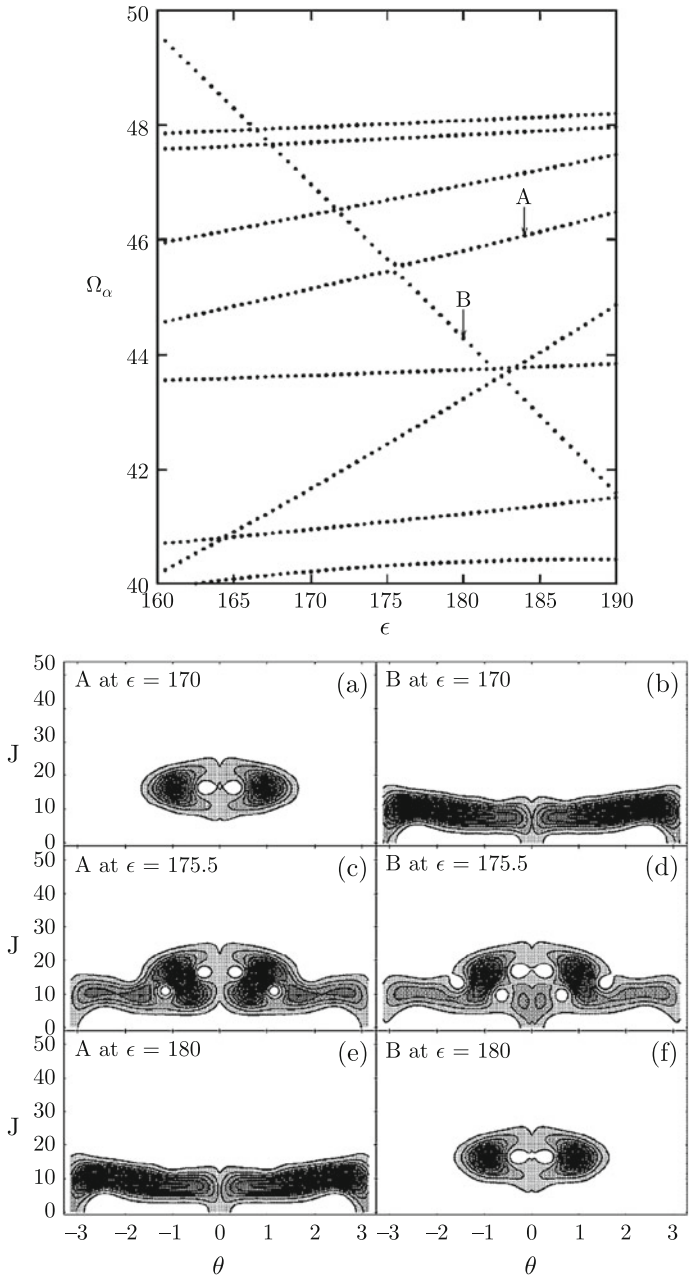

Fig. 3.4 Avoided crossing of Floquet energies (here denoted by Ω_α) as a function of field amplitude (*upper panel*) and associated change of character of the Floquet functions, corresponding to the two levels labelled by A and B in the driven quantum well (*lower panels* (**a–f**)); from [34]

Furthermore, a general density operator can be written as

$$\hat{\rho} = \frac{1}{2}\left(\hat{1} + \boldsymbol{r}\cdot\hat{\boldsymbol{\sigma}}\right), \tag{3.84}$$

with a vector $\boldsymbol{r}$ that is of unit length for all times in the case of pure state dynamics, and a vector-operator $\hat{\boldsymbol{\sigma}}$, composed of the Pauli operators. This then allows for a geometrical interpretation of two-level dynamics by going to the Feynman-Vernon-Hellwarth (or Bloch sphere) representation, discussed in the book by Tannor [36].

The pure state density matrix, in the case of a two-level system with energies E_1, E_2, in the basis of the corresponding eigenstates is given by

$$\boldsymbol{\rho} = \begin{pmatrix} |d_1|^2 & d_1 d_2^* \exp\{-\mathrm{i}(E_1 - E_2)t/\hbar\} \\ d_1^* d_2 \exp\{\mathrm{i}(E_1 - E_2)t/\hbar\} & |d_2|^2 \end{pmatrix}, \tag{3.85}$$

with the *populations* of the different energy levels on the diagonal and where the off-diagonal elements are sometimes called *coherences*.

A frequently considered mixed state is the thermal density matrix at temperature T with only diagonal elements

$$\rho_{mn} = \frac{\mathrm{e}^{-\beta E_n}}{Q}\delta_{mn}, \tag{3.86}$$

where $\beta = 1/(k_\mathrm{B}T)$ with Boltzmann constant k_B and where $Q = \sum_{n=1}^{2} \mathrm{e}^{-\beta E_n}$ is the partition function. An initial pure state evolves into a thermal mixed state by relaxation (due to coupling to an environment) which is governed by the time scale for population decay T_1 and the dephasing or coherence decay time scale T_2, which are related via

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T_2^*}, \tag{3.87}$$

with the pure dephasing time T_2^* [36].

3.C Two-Level System in an Incoherent Field

As the starting point of the perturbative treatment of a two-level system in an incoherent external field, we use the Schrödinger equation in the interaction representation (3.60) and (3.61) with the initial conditions $d_1(0) = 1$ and $d_2(0) = 0$. For very small perturbations, the coefficient d_1 is assumed to remain at its initial value, leading to

$$\mathrm{i}\dot{d}_2 = v_{21}(t)\exp[\mathrm{i}\omega_{21}t]\ . \tag{3.88}$$

This equation can be integrated immediately to yield

$$d_2(t) = -\mathrm{i}\int_0^t \mathrm{d}t' v_{21}(t') \exp[\mathrm{i}\omega_{21}t'] \,, \tag{3.89}$$

analogous to the first order iteration in (2.28). The field shall consist of a superposition of waves with uniformly distributed, statistically independent phases ϕ_j

$$\boldsymbol{\mathcal{E}}(t) = \frac{1}{2}\sum_{\omega_j>0} \boldsymbol{\mathcal{E}}_j \exp[\mathrm{i}\phi_j - \mathrm{i}\omega_j t] + \text{c.c.} \,. \tag{3.90}$$

If we insert this into the equation above, we get

$$\begin{aligned} d_2(t) &= -\frac{\mathrm{i}}{2\hbar}\sum_j \boldsymbol{\mathcal{E}}_j \cdot \boldsymbol{\mu}_{21} \exp[\mathrm{i}\phi_j] \int_0^t \mathrm{d}t' \exp[\mathrm{i}(\omega_{21} - \omega_j)t'] \\ &= -\frac{\mathrm{i}}{2\hbar}\sum_j \boldsymbol{\mathcal{E}}_j \cdot \boldsymbol{\mu}_{21} \exp[\mathrm{i}\phi_j] S_j \,, \end{aligned} \tag{3.91}$$

where the definition

$$S_j = [\mathrm{i}(\omega_{21} - \omega_j)]^{-1} \left\{\exp[\mathrm{i}(\omega_{21} - \omega_j)t] - 1\right\} \tag{3.92}$$

has been introduced. The occupation probability of the second level is then given by the double sum

$$|d_2(t)|^2 = (2\hbar)^{-2} \sum_j \sum_{j'} \exp[\mathrm{i}(\phi_j - \phi_{j'})] \boldsymbol{\mathcal{E}}_j \cdot \boldsymbol{\mu}_{21} \boldsymbol{\mathcal{E}}_{j'} \cdot \boldsymbol{\mu}_{21}^* S_j S_{j'}^* \,. \tag{3.93}$$

Averaging over the phases is now performed and denoted by $<>$, yielding

$$< \exp[\mathrm{i}(\phi_j - \phi_k)] >= \delta_{jk} \,. \tag{3.94}$$

One of the sums in (3.93) therefore collapses and

$$< |d_2(t)|^2 >= \left|\frac{\boldsymbol{e} \cdot \boldsymbol{\mu}_{21}}{\hbar}\right|^2 \sum_j |\mathcal{E}_j|^2 (\omega_{21} - \omega_j)^{-2} \sin^2[(\omega_{21} - \omega_j)t/2] \tag{3.95}$$

follows for identical polarization, $\boldsymbol{e}$, of the light waves.

Now we have to sum over the distribution of frequencies. To this end we consider the time derivative of the expression above[8]

$$\frac{\mathrm{d}}{\mathrm{d}t} < |d_2(t)|^2 >= \left| \frac{\boldsymbol{e} \cdot \boldsymbol{\mu}_{21}}{\sqrt{2}\hbar} \right|^2 \sum_j |\mathcal{E}_j|^2 (\omega_{21} - \omega_j)^{-1} \sin[(\omega_{21} - \omega_j)t] \ . \quad (3.96)$$

With the definition of an energy density per *angular* frequency interval $\rho(\omega_j) = \frac{1}{2}\varepsilon_0 |\mathcal{E}_j|^2 / \Delta\omega_j$, assuming that the frequencies are distributed continuously, and replacing $\rho(\omega_j)$ by its resonance value $\rho(\omega_{21})$, due to

$$\int_{-\infty}^{\infty} \mathrm{d}\omega \sin(\omega t)/\omega = \pi \ , \quad (3.97)$$

we get

$$\frac{\mathrm{d}}{\mathrm{d}t} < |d_2(t)|^2 >= \frac{\pi}{\varepsilon_0} \left| \frac{\boldsymbol{e} \cdot \boldsymbol{\mu}_{21}}{\hbar} \right|^2 \rho(\omega_{21}) \ . \quad (3.98)$$

The right hand side of this expression is a constant and therefore consistent with the assumptions made in the derivation of Planck's radiation law in Chap. 1.

Comparing the equation above with (1.2) for N_1=1 and after switching from the *angular* to the *linear* frequency case [37]

$$B = \frac{2\pi^2}{\varepsilon_0} \left| \frac{\boldsymbol{e} \cdot \boldsymbol{\mu}_{21}}{h} \right|^2 \quad (3.99)$$

is found for Einstein's B coefficient.

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[8] Note that $2\cos(x/2)\sin(x/2) = \sin(x)$.

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