Influence of phonons on light emission and propagation in semiconductor nano optics

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Matthias-René Dachner

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Vorsitzende/r: Prof. Dr. rer. nat. Stephan Reitzenstein, TU Berlin
1. Gutachter: Prof. Dr. rer. nat. Andreas Knorr, TU Berlin
2. Gutachter: Priv.-Doz. Dr. rer. nat. Uwe Bandelow, WIAS

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In this work the optical properties of semiconductor quantum dots (QDs) with regard to their application in optical amplifiers are investigated. Quantum dot semiconductor optical amplifiers (SOAs) can be operated as non-linear optical devices. In this regime the propagation of an optical pulse through an SOA may result in a reshaping of the pulse determined by the interplay of effects such as saturable gain and loss, external pumping and dispersion.

In a solid state environment, phonons introduce losses, but can be also responsible for the carrier capture and thus for the pumping of the QDs. In this thesis, the pure dephasing, decoherence without a change of the levels’ occupations, is investigated on the basis of the independent boson model. The influence of phonons on the shape of linear optical spectra with respect to the wave functions as well as the phonon dispersion relation is examined. Therefore, the resulting spectra of microscopically calculated wave functions as well as simple model wave functions are compared. The impact of the wave function parameters size and internal dipole moment, i.e. displacement between the centers of mass of electron and hole wave function, are investigated for the typical coupling mechanisms deformation potential coupling, piezoelectric coupling for acoustic phonons and polar coupling to optical phonons. For optical spectra of QDs the phonon dispersion relation is important. Due to the spatial confinement, higher phonon wavenumbers gain importance, thus the typical Debye and Einstein assumptions are compared to more realistic model dispersion relations.

Next, the phonon assisted carrier capturing is discussed. The capture process is described via an effective multiphonon Hamiltonian, that is perturbatively derived within a projection operator formalism. This leads to a rate equation with temperature dependent scattering rates.

The optical response of QDs is determined by the carrier dynamics. To calculate the dynamics, the non-resonant QD optical Bloch equations are investigated. With the aim of an analytical expression for the adiabatic response, the microscopical polarization is described by a perturbative series for the regimes: conservative (negligible losses), slow pumping (dephasing much stronger than relaxation processes) and fast pumping (dephasing and relaxation of the same magnitude).

Finally, an electric field propagating through a QD medium is described using the microscopically derived adiabatic response. The propagation is formulated in terms of a generalized Ginzburg-Landau equation with parameters on a microscopic footing for the conservative as well as the dissipative regime. Those wave equations are investigated for the existence of (dissipative) solitons.
Zusammenfassung


Die optische Antwort der Quantenpunkte wird durch ihre Ladungsträgerdynamik bestimmt. Zur Berechnung dieser Dynamik werden die nicht-resonanten optischen Blochgleichungen verwendet. Mit dem Ziel die adiabatische optische Antwort analytisch zu beschreiben, werden die Blochgleichungen störungstheoretisch, in den verschiedenen Bereichen: konservatives Regime (vernachlässigbare Verluste), Regime des langsamen Pumpens (signifikante Dephasierung, viel schwächere Relaxation), sowie Regime des schnellen Pumpens (Dephasierung und Relaxation sind in der gleichen Größenordnung), entwickelt.

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Prologue

1.1 Introduction

Our modern society is based on the transfer of information. Optical data transmission is the backbone of long-range transmission and makes its way to shorter ranges as faster transmission rates are required. In electronics the speed of information processing is limited, since the electrons’ mass introduces resistance that is even enhanced by the skin effect, that pushes the electric current to a conductor’s surface for higher frequencies [Mit10]. For photons these specific solid state restrictions do not apply. However, the realization of fast optical devices has to overcome other limitations. In particular, optical pulses in a medium suffer dispersion, i.e. temporal broadening after a propagated distance due to different phase velocities of the frequencies the pulse is composed of. This limits the interval between two optical bits and thus the data transmission rate. Therefore, a deep understanding of propagation of optical pulses and the interaction with the host medium is of crucial importance to improve data transmission. In fiber optics the concept of optical solitons was embraced, dealing specifically with this problem. Here the dispersion in the fiber is exploited, as it cancels the non-linear optical response of the material leading to stable optical pulse shapes over long distances [Mit10].

The fundamental concept of a soliton was introduced when John Scott Russell observed a water wave in a channel in 1834 that did not change in shape or velocity over a distance of 2-3 km. Korteweg and De Vries presented their famous equation for the wave propagation in a shallow water channel and the corresponding “solitary wave” solution in 1895 [KV95]. In 1965 Zabusky and Kruskal investigated waves in a collisionless plasma, which can be also described via the Korteweg-DeVries equation, and coined the name “soliton” for numerically found solutions, that did not change their shape during propagation and showed elastic scattering [ZK65]. Gardner et.
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al. discovered in 1967 an analytical method to obtain such soliton solutions including interaction [GGK+67]. The inverse scattering theory, which can be used to obtain soliton solutions for various non-linear equations is based on this work [ZS74; NML+84]. In 1973 Hasegawa and Tappert described the light in fibers via the non-linear Schrödinger equation and proposed the existence of optical solitons in dielectric materials [HT73].

Despite that, over the years many non-linear equations having soliton solutions were intensively studied. The term soliton is restricted to conservative (closed, integrable) systems and in many practical applications dissipation or coupling to the environment is not negligible. However, also in dissipative systems localized structures can occur, leading to the concept of the “dissipative soliton” [AA05a; AA08a]. In the same way as solitons they are stable localized structures in a variety of physical, biological or chemical systems.

This work will shed some light on a host material that consists of semiconductor quantum dots (QDs) providing the basis for dissipative optical solitons. In QDs the electrons are spatially confined below their DeBroglie wavelength and thus have discrete energy levels, rendering them a highly efficient optical material for lasers or optical amplifiers [BGL99; Gru06].

The thesis is divided into five chapters. This first chapter introduces or revisits the nomenclature and general properties of electric fields, carriers in semiconductor quantum dots, phonons, as well as the respective interactions. The second chapter is focused on the dissipative influence of phonons on the dynamics of electrons. Specifically, Sec. 2.1 shows the influence of pure dephasing on optical spectra, whereas Sec. 2.2 focuses on the phonon assisted coupling to an electronic reservoir. The electron dynamics are described in a rate equation limit via the QD Bloch equations. In Chap. 3 we explore the Bloch equations in various limits to yield analytical expressions for the QDs’ optical response. This non-linear optical response is then used in wave equations in Chap. 4, that consequently describe the propagation of optical pulses through QD media. The properties of those wave equations are investigated especially in connection with soliton solutions.

1.2 Dramatis personae

This work describes the dynamics of electrons in semiconductor quantum dots, the influence of phonons on them as well as the properties and evolution of optical fields interacting with this solid state system. In this section the basics of the framework shall be briefly introduced. Section 1.2.1 discusses wave functions and energies of the eigenstates of the electronic system, Sec. 1.2.2 comments on the phonon modes and the interaction mechanisms to the electronic system and in Sec. 1.2.3 the wave equations for the electric field are treated and linked with the electronic system.

We describe the electronic dynamics within the framework of second quantization. In practice this means the systems wave functions obtain operator character and a commutation relation:

\[ [\psi^\dagger(\vec{r}, t), \psi(\vec{r}', t)]_{\pm} = \delta(\vec{r} - \vec{r}') \]

\( \psi \) denotes the full wave function of the electronic (anti-commutator) or bosonic system (commutator). Those operators are creation and annihilation operators that act on the vacuum state. This wave
function operator is decomposed into the eigenstates of the systems where the coefficients \( c_i \) are in fact operators.

\[
\psi = \sum_i c_i \phi_i,
\]

with the eigenfunctions of the systems Hamiltonian \( \phi_i \). In the next section we discuss the eigenfunctions. The operators of the new systems are constructed by “sandwiching” the first quantization operators with wave function creators and annihilators.

**1.2.1 Electronic Hamiltonian**

The foundation of microscopic calculations presented in this work are the electronic wave functions. They contain the information of the material and the geometry of the structure and thus define the strength of coupling elements and time scales of dynamics. We deal with heterostructures that are composed of different semiconductor materials, such that electrons are confined in a potential due to different band gaps (cf. Fig. 1.1a) [Gru06].

The (self-organized) fabrication of such a structure starts with the bulk medium, where electrons are not confined below their DeBroglie wavelength and thus have defined momenta and corresponding energies in a quasi-continuous bandstructure. On top another material is deposited and due to a mismatch of the lattice constants stress is generated. Depending on the materials and growing conditions first a few monolayer thick film, the wetting layer (WL) is formed before the stress is alleviated by forming small islands, the quantum dots (QDs) [BGL99]. Typical QDs are flat and have a typical diameter of 10-20 nm.

Due to the three-dimensional confinement, electrons in QDs have discrete energies and are thus called “artificial atoms”. They have advantageous optical properties as a distinguished and controllable emission wavelength and a high quantum yield [Bim05]. Furthermore, they are prosperous candidates for single and entangled photon sources [BSP+00; SYA+06; Bim08; SWO+10]. Wetting layer and bulk have quasi-continuous energies that can be used as a carrier reservoir for the QDs [FCA+01; MUB+02; GBDT+08; WDM+09; Wil13].

**Effective mass and envelope approximation**

There are several microscopic approaches to calculate wave functions of semiconductor heterostructures as \( 8\vec{k} \cdot \vec{p} \) theory [SWL+09; SWB07; SBL+03; WSB06] or tight binding calculations [SMS+11; SLS+11] beginning with the position of every atom.

It is however of advantage to get an analytical hold of the wave functions. This is the purpose of the envelope function approximation [HK04; YC05] for heterostructures. Here, the overall lattice potential is approximated by a microscopic, periodic lattice \( V_g \) with an additional slowly varying envelope potential \( U \), (cf. Fig. 1.1b). The full systems Hamiltonian reads:

\[
H |\psi\rangle = (H_0 + V_g + U) |\psi\rangle = \epsilon |\psi\rangle.
\]
\[ H_0 = \text{the quasi-kinetic energy part of the electron Hamiltonian and } \varepsilon \text{ the eigenenergy of the eigenstate } |\psi\rangle. \]

Without envelope potential \( U \) there are the eigenstates \( |i, \lambda\rangle \) with eigenenergies \( E_i^\lambda \) satisfying Bloch's theorem.

\[
(H_0 + V_g) |i, \lambda\rangle = E_i^\lambda |i, \lambda\rangle, \quad |i, \lambda\rangle = \frac{1}{\sqrt{V}} u_i^\lambda (\vec{r}) e^{i\vec{k}_i \cdot \vec{r}}, \quad u_i^\lambda (\vec{r} + \vec{R}) = u_i^\lambda (\vec{r}). \quad (1.2)
\]

Here, \( \lambda \) describes the bands and \( i \) is the intraband index, denoting the energies within the bands. \(^1\)

Since \( |i, \lambda\rangle \) are a complete orthonormal system \( |\psi\rangle \) can be decomposed with coefficients \( c_i^\lambda \).

\[
|\psi\rangle = \sum_{\lambda} \sum_i c_i^\lambda |i, \lambda\rangle, \quad (1.3)
\]

\[
\langle j, \lambda' | H | i, \lambda \rangle = c_j^{\lambda'} E_j^{\lambda'} + \sum_{\lambda} \sum_i c_i^\lambda \langle j, \lambda' | U | i, \lambda \rangle = \varepsilon c_j^{\lambda'}. \quad (1.4)
\]

The envelope potential changes much slower than the atomic potentials, thus we can assume it to be constant over the range of a unit cell \( U(\vec{R} + \vec{r}) \approx U(\vec{R}) \).

\[
\langle j, \lambda | U | i, \lambda' \rangle \approx \frac{1}{N} \sum_n U(\vec{R}_n) e^{-i (\vec{k}_j - \vec{k}_i) \cdot \vec{R}_n} \frac{1}{V_{uc}} \int_{V_{uc}} d\vec{r} u_j^{\lambda^*} (\vec{r}) u_i^\lambda (\vec{r}) e^{i (\vec{k}_i - \vec{k}_j) \cdot \vec{r}}, \quad (1.5)
\]

\(^1\)In the typical III-V semiconductors there are four relevant bands: one conduction band and the three valence bands light hole, heavy hole and split-off bands. We will however restrict ourselves to the heavy holes, since there are energetic offsets between the valence bands in semiconductor-nano structures \([YC05]\).
with the typical approximation of small wave numbers $u_k^\lambda (\vec{r}) \approx u_0^\lambda (\vec{r})$ (i.e. the resulting wave functions should have a larger spatial extent) the integral over the unit cell: $\frac{1}{V_{uc}} \int_{V_{uc}} d\vec{r} u_0^\lambda (\vec{r}) u_0^{\lambda'} (\vec{r}) = \delta_{\lambda,\lambda'}$ yields band diagonality. This means within this approximation there are no band mixing effects for the wave functions.

$$\langle j, \lambda | U | i, \lambda' \rangle \approx \frac{1}{N} \sum_n U(\vec{R}_n) e^{-i(\vec{k}_j - \vec{k}_i) \cdot \vec{R}_n} \delta_{\lambda,\lambda'}.$$ (1.6)

The next typical approximation is the effective mass approximation, where the band structure of the host material is expanded for small wave numbers yielding a parabolic behavior:

$$E_j^\lambda = E_0^\lambda + \frac{\hbar^2 \vec{k}_j^2}{2m_j^\lambda}.$$ (1.7)

$m_j^\lambda$ is an effective, band and material depending mass. Equation (1.4) is multiplied with $e^{i\vec{k}_j \cdot \vec{r}}$ and the sum over all $\vec{k}_j$ is taken, then it reads

$$\sum_{\vec{k}_j} c_{\vec{k}_j}^\lambda \frac{\hbar^2 \vec{k}_j^2}{2m_j^\lambda} e^{i\vec{k}_j \cdot \vec{r}} + \sum_{\vec{k}_i} c_{\vec{k}_i}^\lambda \frac{1}{N} \sum_n U(\vec{R}_n) \sum_{\vec{k}_j} e^{-i\vec{k}_j \cdot (\vec{R}_n - \vec{r})} e^{i\vec{k}_i \cdot \vec{R}_n} \delta_{\lambda,\lambda'} = (\epsilon - E_0^\lambda) \sum_{\vec{k}_j} c_{\vec{k}_j}^\lambda e^{i\vec{k}_j \cdot \vec{r}}.$$ (1.8)

This is a Schrödinger equation for the envelope wave functions $\phi = \sum_{\vec{k}} c_{\vec{k}}^\lambda e^{i\vec{k} \cdot \vec{r}}$, where only the envelope is the potential.

$$\left(-\frac{\hbar^2 \Delta}{2m_j^\lambda} + U(\vec{r})\right) \phi_\lambda (\vec{r}) = \mathcal{E}_\lambda \phi_\lambda (\vec{r}).$$ (1.9)

The Eigenfunctions $\phi_\lambda$ yield the coefficients for the full wave function $| \psi \rangle$ by the definition in Eq. (1.3)

$$\psi_\lambda (\vec{r}) = \frac{1}{\sqrt{V}} \sum_i u_i^\lambda (\vec{r}) c_i^\lambda e^{i\vec{k}_i \cdot \vec{r}} \approx u_0^\lambda (\vec{r}) \phi_\lambda (\vec{r}),$$ (1.10)

$$\epsilon^\lambda = E_0^\lambda + \mathcal{E}_\lambda.$$ (1.11)

The wave equation of a semiconductor heterostructure is composed of the eigenfunction of the envelope Schrödinger equation and a Bloch factor. The eigenenergies are the ones from the envelope problem plus the band edge.

The following section presents the specific wave functions for this work.
Wave functions of the system

To describe the system, a convenient model potential $U$ has to be chosen. For the sake of simplicity this is done independently for QD and WL states, yielding an unintended wave function overlap. This can be corrected by the orthogonalized plane waves technique [Nie05; Mal08; Wil13].

A typical choice of the model potential describes lens shaped QDs via an in-plane harmonic oscillator potential and an one dimensional potential well in growth direction[WHF+96; Nie05; Mal08].

The lowest state $\phi_s$ is the product of the two dimensional harmonic oscillator ground state $\varphi^\lambda_s(x, y)$ and the ground state of an one dimensional infinite well $\xi_0^\lambda(z)$. This wave function has no nodes and is thus called $s$ state in analogy to the hydrogen atom:

$$\phi^\lambda_s(\vec{r}) = \varphi^\lambda_s(x, y)\xi_0^\lambda(z),$$

(1.12)

$$\varphi^\lambda_s(x, y) = \sqrt{\frac{m\lambda\omega^\lambda_s}{\hbar\pi}} e^{-\frac{m\lambda\omega^\lambda_s}{2\hbar}(x^2+y^2)},$$

(1.13)

$$\xi_0^\lambda(z) = \begin{cases} \sqrt{\frac{2}{L}} \cos(\frac{\pi z}{L}), & |z| \leq L/2, \\ 0, & \text{else}, \end{cases}$$

(1.14)

$$E_{0}^\lambda = \frac{\hbar^2}{2m\lambda} \omega^\lambda_s + \frac{\hbar^2\pi^2}{2m\lambda L^2} + E_{\text{InAs}}.$$  

(1.15)

$L$ describes the height of the QDs with an effective width of the infinite quantum well. Alternatively the $x$-$y$-part is written as

$$\varphi^\lambda_s(x, y) = e^{-\frac{x^2+y^2}{2l^2_s}} \ln \frac{2}{\lambda},$$

(1.16)

with $l^\lambda_s = \sqrt{\frac{\hbar\ln 2}{m\lambda\omega^\lambda_s}}$ being the full width at half maximum of the probability density. This size can be measured [MBM+03] or calculated by e.g. 8 band $\vec{k} \cdot \vec{p}$ calculations [SGB99; SWB07].

To describe the next higher states, the first excited state of the harmonic oscillator is used:

$$\varphi^\lambda_p = x\sqrt{2} \left( \frac{m\lambda\omega^\lambda_s}{\pi\hbar} \right)^{\frac{1}{2}} e^{-\frac{m\lambda\omega^\lambda_s}{2\hbar}(x^2+y^2)}.$$

Those, so called, $p$ states are two fold degenerated, one node in $x$ direction, one in $y$ direction, where $x$, $y$ have to be chosen according to the crystal symmetry.

The wetting layer is described as one dimensional potential well in $z$ direction, in the $x$-$y$-plane...
the states are described as plane waves:

\begin{equation}
\phi_l = \varphi_l(x, y) \xi_l(z), \quad l \neq 0,
\end{equation}

\begin{equation}
\varphi_l(x, y) = e^{i(k_l^x x + k_l^y y)},
\end{equation}

\begin{equation}
\xi_l(z) = \begin{cases} 
\sqrt{\frac{2}{L_z}} \cos \frac{\pi z}{L_z}, & |z| \leq L_z/2, \\
0, & \text{else},
\end{cases}
\end{equation}

\begin{equation}
E_{\lambda}^\lambda = \frac{\hbar^2 (k_l^x)^2 + (k_l^y)^2}{2m_{\lambda}^2} + \frac{\hbar^2 \pi^2}{2m_{\lambda} L_z^2} + E_{\text{InGaAs}}.
\end{equation}

$L_z$ denotes the effective width of the WL. Although multiple subbands with continuous energies exist, we restrict ourselves to the lowest subband.

Figure 1.2: Energies of the QD–WL system (for a InAs/GaAs example) in space (a) and reciprocal space (b). The QD states have discrete energies, whereas the WL states are a continuum. The binding energy $\Delta E$ is a characteristic quantity, especially for capture processes.

An important quantity for the phonon assisted relaxation (cf. Sec. 2.2) is the energetic distance between band edge of the WL and the QD energies, the binding energy $\Delta \epsilon^\lambda$ (cf. Fig. 1.2b). With the presented wave functions there is the following dependence

\begin{equation}
\Delta \epsilon^\lambda = E_{\text{InGaAs}}^\lambda - E_{\text{InAs}}^\lambda + \frac{\hbar^2 \pi^2 (L_z^2/L_{\lambda}^2 - 1)}{2m_{\lambda} L_z^2} - \frac{\hbar^2 \ln 2}{2m_{\lambda} (l_{\lambda}^\lambda)^2}.
\end{equation}

It depends on the size of the nanostructures as well as on the material parameters: band edges and effective masses. For electrons, the binding energy is larger than for holes, due to the greater hole
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mass. Further, smaller QDs have a smaller binding energy [SGB99] due to their larger ground state energy. However, since model wave functions are always an approximation, we will use the binding energy as a free parameter, with the dependencies above in mind.

For the calculation of optical spectra in Sec. 2.1 we will use an even simpler approach to the QD states, where the wave functions are Gaussian and isometric in all three spatial directions [WRR+08].

1.2.2 Phonon Hamiltonian

Having discussed the eigenstates of the electronic system we turn next to the quantized lattice vibrations, the phonons. In solid state systems the dynamics of the lattice vibrations, the phonons, are of importance. The ions of the periodic lattice $V_g(\vec{r}) = \sum_n v(\vec{r} - \vec{R}_n)$ (cf. Eq. (1.2.1)) are in motion, described as coupled harmonic oscillators around the positions $\vec{R}_n = \vec{R}_n^{(0)} + \vec{u}_n$. The displacements $\vec{u}_n$ can be described via a continuous displacement field $\vec{u}(\vec{r})$. To describe phonons the system of oscillators is diagonalized leading to a system of uncoupled oscillators. The phonons are described by creation and annihilation operators $b^\dagger_{\vec{q},\kappa}, b_{\vec{q},\kappa}$ with eigenenergies $\hbar \omega_{\kappa}(\vec{q})$ that depend on the lattice symmetry. Figure 1.3 shows the dispersion relations of phonons in GaAs and AlN. There are two categories of branches with qualitatively different dispersions: acoustic and optical phonons. For acoustic phonons, ions in one unit cell oscillate in phase, which leads to a linear dispersion at the $\Gamma$ point ($q = 0$) [Czy00]. For optical phonons the ions of a unit cell oscillate in opposite directions leading to a non-vanishing dispersion at $q = 0$. The small displacements are described via phonon operators by the relation [Hak73; Czy00]

$$\vec{u}(\vec{R}_n^{(0)}) = \sum_{\vec{q}} \vec{u}_{\vec{q}}(\vec{R}_n^{(0)}) = \frac{1}{\sqrt{N}} \sum_{\vec{q}} \sum_{\kappa} \sqrt{\frac{\hbar}{2M_n \omega_{\kappa}(\vec{q})}} \hat{e}_{\kappa}^{\text{ph}}(\vec{q}) e^{i\vec{q} \cdot \vec{R}_n^{(0)}} (b_{\vec{q},\kappa} + b^\dagger_{-\vec{q},\kappa}), \quad (1.22)$$

with $M_n$ being the mass of the ions, $N$ the number of unit cells, $\kappa$ denotes the dispersion branches, $\omega_{\kappa}(\vec{q})$ is the dispersion relation of the respective modes. Finally, $\hat{e}_{\kappa}^{\text{ph}}(\vec{q})$ denotes the unit vector of the phonon polarization.

The next important part of the Hamiltonian is the electron-phonon interaction. On the following pages we want to recap the most important of the various interaction mechanisms.\(^2\)

Deformation potential coupling

The deformation potential coupling directly takes into account the energetic changes of the electronic band structure for the displaced lattice. This short derivation follows Ref. [YC05]. The lattice potential is decomposed into the resting lattice and a correction from the displacement:

$$v(\vec{r} - \vec{R}) = v(\vec{r} - \vec{R}_0 - \vec{u}) \approx v(\vec{r} - \vec{R}_0) - \nabla v(\vec{r} - \vec{R}_0) \cdot \vec{u}.$$

\(^2\)A compilation of properties and derivations of those and other mechanisms can be found in Ref. [Hau70]
Figure 1.3: Dispersion relation of GaAs [GGP+91] and AlN [SBMP+99] and visualization of dispersion relation approximations. Acoustic phonons start linearly at the $\Gamma$ point ($\vec{q} = 0$) and in Debye approximation (not shown) they are linearly treated over the whole Brillouin zone. As an improvement we use a sinusoidal description as the approximation. Optical phonons have a constant value near the $\Gamma$ point ($\vec{q} = 0$) and in Einstein approximation they are assumed to have a constant dispersion over the whole Brillouin zone. We approximate the dispersion by a cosine. The high symmetry points of the Brillouin zone are $\vec{q}_\Gamma = (0, 0, 0)$, i.e. the center, the edges of the Zincblende Brillouin zone $\vec{q}_K = \frac{3\pi}{2a}(1, 1, 0)$, $\vec{q}_X = \frac{2\pi}{a}(0, 1, 0)$, $\vec{q}_L = \frac{\pi}{a}(1, 1, 1)$ and the edges of a Wurtzit Brillouin zone $\vec{q}_K = \frac{4\pi}{3a}(1, 0, 0)$, $\vec{q}_M = \frac{\pi}{\sqrt{3}a}(\sqrt{3}, 1, 0)$, and $\vec{q}_A = \frac{\pi}{2a}(0, 0, 1)$. The Brillouin zone high symmetry points for various lattices can be found in [SC10]. For all approximations we use an isotropic description, i.e. assuming the Brillouin zone to be spherical.

Since the lattice potential dictates the electronic energy, the interaction Hamiltonian is expressed in terms of the electronic Hamiltonian.

\[
H_{e-ion} = \frac{\partial H_e}{\partial \vec{R}(0)} \vec{u} \approx \sum_{\lambda_i} \left( \sum_{\lambda_i} \langle \lambda_i | \frac{\partial E_{\lambda_i}}{\partial \vec{R}(0)} | \lambda_i \rangle a_{\lambda_i}^\dagger a_{\lambda_i} \right)
\]

The energetic change $\delta E_{\lambda_i}$ with respect to the lattice displacement is typically accessed via a volume dilatation $\delta V$[YC05]

\[
\delta E_{\lambda_i} = D_{\lambda_i} \frac{\delta V}{V}.
\]

with the volume deformation potential $D_{\lambda_i}$.\footnote{Note that in the literature there is a, for non-experts confusing, amount of deformation potentials. For the coupling to LA phonons, typically the volume deformation potential is used.} To relate this with the phonons, i.e. lattice displacement, linear deformation theory is used. The displacement field is described via a deformation matrix $d_{ij}$:

\[
u_i(\vec{R}) = \sum_j d_{ij} R_j.
\]
For example, $d_{ij} = \delta_{xi}\delta_{ij}$ would be stretching of the solid along the $x$-axis up to the double width. For a linear theory, i.e., $d_{ij} = \text{const}$., the definition can be inverted and divided into symmetric and asymmetric parts.

$$
d_{ij} = \frac{\partial u_i}{\partial R_j} = 0.5 \left[ \left( \frac{\partial u_i}{\partial R_j} + \frac{\partial u_j}{\partial R_i} \right) + \left( \frac{\partial u_i}{\partial R_j} - \frac{\partial u_j}{\partial R_i} \right) \right] = s_{ij} + f_{ij} \quad (1.23)
$$

The strain tensor $s_{ij}$ describes deformation, while the asymmetric $f_{ij}$ describes rotations and does not change electronic energies. For small deformations the volume dilatation can be expressed as the trace of the strain tensor:

$$
\frac{\delta V}{V} = s_{xx} + s_{yy} + s_{zz} + s_{xx}s_{yy} + s_{yy}s_{zz} + s_{zz}s_{xx} + s_{xx}s_{yy}s_{zz} \approx s_{xx} + s_{yy} + s_{zz}.
$$

The displacement field is expressed with phonon operators (cf. Eq. (1.22)):

$$
s_{ij} = \frac{1}{\sqrt{N}} \sum_{\vec{q}} 0.5(q_j u_i^0 + q_i u_j^0) \sum_{\kappa} \sqrt{\frac{\hbar}{2M_i \omega_\kappa(\vec{q})}} e^{i\vec{q} \cdot \vec{R}(0)} n_{\kappa} \epsilon_{\kappa \lambda} \epsilon_{\lambda \mu} (\vec{q}) e^{i\vec{q} \cdot \vec{e}_{\kappa \mu}(\vec{q})},
$$

(1.24)

To be consistent and have a spatially constant strain tensor, this approximation is only valid in the long wavelength limit $\vec{q} \rightarrow 0$. This yields the energy change per lattice displacement due to deformation potential coupling

$$
\delta E_{\lambda i} = D_{\lambda i} \vec{q} \cdot \vec{u} \Rightarrow \frac{\partial E_{\lambda i}}{\partial \vec{R}} = D_{\lambda i} \vec{q},
$$

$$
H_{\text{el-ph}}|_{\text{DF}} = D_{\lambda i} \sum_{\vec{q}} \langle \lambda j | \vec{q} \cdot \vec{u} | \lambda i \rangle a_{\lambda j}^{\dagger} a_{\lambda i},
$$

and thus the interaction Hamiltonian can be written formally

$$
H_{\text{el-ph}}|_{\text{DF}} = \sum_{\vec{q}, \kappa} \sum_{\lambda ij} g_{ji}^{\kappa \lambda} a_{\lambda j}^{\dagger} a_{\lambda i} (b_{\vec{q}, \kappa} + b_{-\vec{q}, \kappa}^{\dagger}),
$$

with the deformation potential coupling element:

$$
g(\vec{q})_{\kappa} = \sqrt{\frac{\hbar}{2q V \omega_{\kappa}(\vec{q})}} \left( \int d^3r |\varphi_{\lambda}(r)^2 e^{i\vec{q} \cdot \vec{r}} \right) \vec{q} \cdot \vec{e}_{\kappa \lambda}(\vec{q}). \quad (1.25)
$$

Since $\vec{q} \cdot \vec{e}_{LA}(\vec{q}) = q$ and $\vec{q} \cdot \vec{e}_{TA}(\vec{q}) = 0$ there is only coupling to longitudinal acoustic phonons.
Piezoelectric coupling

In the case of piezoelectric coupling the displacement induced strain $s_{ij}$ creates an electric field $\vec{E}$. This effect is strong in polar material and is accounted by an electro-mechanical tensor $e_{\text{mech}}^{ij}$. The electric displacement field from Maxwell’s equations reads [Mah72; YC05]

$$D_k = \sum_{i,j} e_{\text{mech}}^{ij} s_{ji} + \varepsilon_0 \varepsilon_r E_k,$$

with the relative permittivity of the material $\varepsilon_r$ (excluding piezoelectric effects) which is assumed to be isometric. This means a deformation of the $i - j$ plane results in an electric field in the $k$ direction. $\varepsilon_r$ is composed of the response from the electrons, which is commonly assumed to be the high frequency permittivity $\varepsilon^\infty$ and of the permittivity from the ions, which is the static permittivity without the electronic contribution $\varepsilon^\text{stat} = \varepsilon^\infty$ thus $\varepsilon_r = \varepsilon^\text{stat}$ [Mah72].

---

**Figure 1.4:** Visualization of the strain tensor and the piezoelectric fields created by deformation.

Using the source equation from Maxwell’s equations $\nabla \cdot \vec{D} = 0$ in Fourier domain yields the relation between the strain to a longitudinal electric field that is created:

$$\sum_k q_k D_k = 0 = \sum_k \sum_{ij} q_k e_{\text{mech}}^{ij} s_{ji}(\vec{q}) + \varepsilon_0 \varepsilon_r \sum_k q_k E_k(\vec{q}).$$

---

4 Actually, one has to use the static relative permittivity without piezoelectric contributions.
5 Sometimes the literature states $\varepsilon_r = \varepsilon^\infty$ [YC05], which would neglect the influence of the ions on the permittivity.
1.2. DRAMATIS PERSONÆ

To incorporate the energy into the Hamiltonian the induced electric field is taken into account via its scalar potential $\phi$:

$$E_k(\vec{q}) = -i q_k \phi(\vec{q}) \Rightarrow \phi(\vec{q}) = \frac{\sum_k \sum_{ij} q_k e_{k,ij}^m s_{ij}(\vec{q})}{\varepsilon_0 \varepsilon_r \sum_k q_k}, \quad \phi(\vec{r}) = \sum_{\vec{q}} e^{i\vec{q}\cdot\vec{r}} \phi(\vec{q}).$$

This potential leads to the electron-phonon interaction $H_{el-ph} = -e_0 \phi(\vec{r})$. Symmetry properties allow the $3 \times 3 \times 3$ tensor $e_{k,ij}^m$ to be described as a $3 \times 6$ tensor as well as the strain tensor as a vector with 6 elements.\(^6\)

The phonon operators enter again via the strain tensor (cf. Eq. (1.24)):

$$H_{el-ph} = -|e_0| \sum_{mn} \langle m|\phi(\vec{r})|n\rangle a_{m}^\dagger a_{n} = -i \sum_{\vec{q}} 0.5 \frac{|e_0|}{\varepsilon_0 q^2} \sum_{ij} q_k e_{k,ij}^{mech} q_i e_j(\vec{q}) \left( \frac{\hbar}{2N \varepsilon \omega} \right)^{1/2} \langle m|e^{i\vec{q}\cdot\vec{r}}|n\rangle \left( b_{-\vec{q},\kappa}^\dagger + b_{\vec{q},\kappa} \right) a_{m}^\dagger a_{n}.$$  

The quantity $\mathcal{F}_\kappa(\vec{q})$ does not depend on the value of $\vec{q}$, but on the angle and is thus influenced by the crystal symmetry. For a Zincblende system one number defines the piezoelectric interaction $e_{14} = e_{25} = e_{36}$, i.e. the induced electric field is transverse to the strain. The coupling in this case reads:

$$\left( \sum_{k} \sum_{ij} q_k e_{k,ij}^{mech} q_i e_j(\vec{q}) \right) = 2(q_x q_y e_z^* + q_x q_z e_y^* + q_y q_z e_x^*).$$

In a Wurtzite system there are three piezoelectric effects $e_{31} = e_{32} = e_{24}, e_{33}$. Deformation of the c-plane ($x-y$ plane) leading to a field along the (0001) (i.e. $z$) direction; deformation along the (0001) direction leading also to a field along (0001); and an askew deformation leading to fields in the c-plane.

**Polar coupling to optical phonons**

Optical phonons, i.e. lattice vibrations where the ions in an unit cell do not oscillate in the same direction, create polarization fields, that change the energy of the electronic states. The derivation is similar to the piezoelectric coupling, because the polarization between the displaced ions creates an electric field. In contrast to the coupling mechanisms discussed before, detailed derivations can be found in nearly every solid state text book (e.g. [Czy00; Hak73; YC05; Mah72]). The Hamiltonian reads

$$H_{el-ph} = \sum_{m,n} \sum_{\vec{q}} g_{mn}^{\vec{q}} \left( b_{-\vec{q},\text{LO}}^\dagger + b_{\vec{q},\text{LO}} \right) a_{m}^\dagger a_{n},$$

\(^6\)The convention is: $1 = xx, 2 = yy, 3 = zz, 4 = yz = zy, 5 = xz = zx, 6 = xy = yx$ and for the first coordinate $1 = x, 2 = y, 3 = z.$
with the Fröhlich coupling element
\begin{equation}
g_{mn}^{\vec{q}} = -i \sqrt{\frac{\hbar \omega_{\text{LO}}}{2V}} \sqrt{\frac{e^2}{\epsilon_0}} \left( \frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon_{\text{stat}}} \right) \frac{1}{q} \left| m \right| e^{i \vec{q} \cdot \vec{r}} \left| n \right>.
\end{equation}

A widespread formulation of this coupling element uses the Fröhlich polaron constant \( \alpha \) as a material parameter [Frö54]:
\begin{equation}
g_{mn}^{\vec{q}} = \sqrt{\frac{1}{\epsilon_0 4\pi}} \left( \frac{(\hbar \omega_{\text{LO}})^2}{2\mu} \right)^{\frac{1}{4}} \sqrt{\alpha} \frac{1}{\sqrt{\epsilon}} q \left| m \right| e^{i \vec{q} \cdot \vec{r}} \left| n \right> \quad \alpha = e^2 \sqrt{\frac{\mu}{2\hbar^3 \omega_{\text{LO}}}} \left( \frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon_{\text{stat}}} \right). \tag{1.26}
\end{equation}

with the reduced mass \( \mu = \frac{m_h m_e}{m_h + m_e} \).

**Form factor**

All the phonon coupling Hamiltonians consist of the form factor \( \langle j | e^{i \vec{q} \cdot \vec{r}} | i \rangle \). For the QD states in envelope function approximation (cf. Sec. 1.2.1) this means:
\begin{equation}
\langle j | e^{i \vec{q} \cdot \vec{r}} | i \rangle = \int d^3 r \phi_{\lambda_j}^* \phi_{\lambda_i} u_{0_j}^{\lambda_j} u_{0_i}^{\lambda_i} e^{i \vec{q} \cdot \vec{r}}
\end{equation}

For long wavelength compared to the size of the unit cell, the phonon coupling is band diagonal.

The form factor has typical Fourier transform properties, i.e. the smaller the extent of the wave functions is, the higher are the wave numbers that contribute.

**1.2.3 Electric fields, Maxwell’s equations, Wave equations**

To investigate the dynamics of electromagnetic waves, Maxwell’s equations are always a good starting point.
\begin{align}
\nabla \cdot \vec{E} &= \rho_{\text{micr}}, \\
\nabla \times \vec{B} &= \partial_t \vec{E} + j_{\text{micr}},
\end{align}

We start with the microscopic Maxwell’s equations, with \( \rho_{\text{micr}} \) including all charges, i.e. electrons and atoms. However, assuming that in the ground state all charges are bound, we will only calculate the microscopic dynamics of the electrons. The wave functions of the QD electrons reads:
\begin{equation}
\Psi^{(1)}(\vec{r}) = \sum_i \sum_l \psi_l^{(s)}(\vec{r} - \vec{r}_i) a_{i,l}^{(1)},
\end{equation}

---

The original definition is in cgs units, the prefactor in red converts the expression to SI units.
with the states \( l \) of the \( i \)th quantum dot and \( a_{i,l}^{(\dagger)} \) being the electronic annihilation (creation) operators as introduced in Sec. 1.2.1.

The electronic charge density of the electrons is defined by

\[
\varrho_{\text{micr}} = -e_0 \left\langle \psi^\dagger \psi \right\rangle = -e_0 \sum_{ij} \sum_{kl} \psi_k^*(\vec{r} - \vec{r}_i) \varphi_l(\vec{r} - \vec{r}_i) \left\langle a_{i,k}^{\dagger} a_{i,l} \right\rangle .
\] (1.28)

The QD wave functions are assumed to have no overlap between different QDs, thus we introduce a Kronecker delta \( \delta_{ij} \). The overall charge is obtained by spatial integration and yields a result \( \sim \delta_{kl} \). In a neutral solid, those diagonal terms are canceled out by the charge of the ions thus the non-diagonal contributions are left \( \sim \bar{\delta}_{kl} = (1 - \delta_{kl}) \), because the wave functions are orthonormal.

The residual part is used to define the macroscopic polarization with \( \nabla \cdot \vec{P} = \varrho_{\text{micr}} \). Thus this quantum mechanical charge expression has to be integrated.

\[
\vec{P}(\vec{r}) = -e_0 \sum_{kl} \int_{-\infty}^\vec{r} d\vec{r}' \sum_i \varphi_k^*(\vec{r}' - \vec{r}_i) \varphi_l(\vec{r}' - \vec{r}_i) \left\langle a_{i,k}^{\dagger} a_{i,l} \right\rangle \delta_{kl}
\]

\[
= -e_0 \sum_{kl} \sum_i \left( \vec{r}' - \vec{r}_i \right) \varphi_k^*(\vec{r}' - \vec{r}_i) \varphi_l(\vec{r}' - \vec{r}_i) \left\langle a_{i,k}^{\dagger} a_{i,l} \right\rangle \delta_{kl}
\]

\[
- \int_{-\infty}^\vec{r} d\vec{r}' \left( \vec{r}' - \vec{r}_i \right) \frac{\partial}{\partial \vec{r}} \varphi_k^*(\vec{r}' - \vec{r}_i) \varphi_l(\vec{r}' - \vec{r}_i) \left\langle a_{i,k}^{\dagger} a_{i,l} \right\rangle \delta_{kl} .
\] (1.29)

The correction \( \nabla \varphi_k^*(\vec{r}' - \vec{r}_i) \varphi_l(\vec{r}' - \vec{r}_i) \) is neglected, and therefore the polarization yields:

\[
\vec{P} = -e \sum_{kl} \sum_i \left( \vec{r}' - \vec{r}_i \right) \varphi_k^*(\vec{r}' - \vec{r}_i) \varphi_l(\vec{r}' - \vec{r}_i) \left\langle a_{i,k}^{\dagger} a_{i,l} \right\rangle (\vec{r}_i).
\]

Since the spatial dependence is microscopic and the QDs have an extent below the spatial resolution of optical fields, thus the polarization of the whole QD is concentrated in one point and can be approximated by a delta distribution.

\[
-e \varphi_k^*(\vec{r}' - \vec{r}_i) \varphi_l(\vec{r}' - \vec{r}_i) \left( \vec{r}' - \vec{r}_i \right) \approx \delta_{kl}(\vec{r}_i) \delta(\vec{r}' - \vec{r}_i) ; \quad \bar{d}_{kl}(\vec{r}_i) = -e \int d\vec{r}' \varphi_k^*(\vec{r}' - \vec{r}_i) \varphi_l(\vec{r}' - \vec{r}_i) \delta(\vec{r}' - \vec{r}_i)
\]

For practical reasons we pool \( N \) quantum dots in a volume \( V \) at position \( \vec{R} \) together and assume they have similar attributes and behave alike

\[
\vec{P}(\vec{R}) = \sum_{kl} \bar{d}_{kl}(\vec{R}) \left\langle a_{k}^{\dagger} a_{l} \right\rangle (\vec{R}) \sum_{\vec{r}_i \in \vec{R}} \delta(\vec{r} - \vec{r}_i) ,
\]

\[
= \frac{N}{V} = n_{\text{QD}}
\]
with the quantum dot density $n_{\text{QD}}$. The microscopic current can be treated similar. From this we get the macroscopic Maxwell’s equations

$$\nabla \cdot \vec{D} = \varrho_{\text{free}}, \quad \nabla \cdot \vec{B} = 0, \quad (1.30a)$$

$$\nabla \times \vec{H} = \partial_t \vec{D} + \vec{j}_{\text{free}}, \quad \nabla \times \vec{E} = -\partial_t \vec{B}. \quad (1.30b)$$

The material properties are included in

$$\vec{D} = \varepsilon_0 \vec{E} + \vec{P} = \varepsilon \vec{E}; \quad \vec{P}(\omega) = \varepsilon_0 \chi(\omega) \vec{E}(\omega); \quad \varepsilon = \varepsilon_0 (1 + \chi),$$

$$\vec{H} = \frac{1}{\mu_0} \vec{B} - \vec{M} = \frac{1}{\mu} \vec{B}; \quad \vec{M} = \chi_m \vec{H}; \quad \mu = \mu_0 (1 + \chi_m).$$

We only consider the common case without free charges or currents $\varrho_{\text{free}} = \vec{j}_{\text{free}} = 0$, which yields the wave equation:

$$\Delta \vec{E} - \frac{1}{c^2} \frac{d^2}{dt^2} \vec{E} = \nabla(\nabla \cdot \vec{E}) + \mu_0 \frac{d^2}{dt^2} \vec{P}. \quad (1.31)$$

The part $(\nabla \cdot \vec{E}) = (\nabla \cdot \vec{P})$ is neglected because we assume transversality of the electric field, that is not spoiled by the polarization.

$$\Delta \vec{E} - \frac{1}{c^2} \frac{d^2}{dt^2} \vec{E} = \mu_0 \frac{d^2}{dt^2} \vec{P}. \quad (1.32)$$

This is the common wave equation for optical fields, which is able to describe the various phenomena discussed in this work. The spatial composition of $\vec{P}(\vec{r})$ defines the optical modes of the system. The behavior of $\vec{P}$ in linear optics is presented in Sec. 2.1, a non-linear description is found in Chap. 3.

**Chirped pulses**

In non-linear optics chirped pulses, i.e. pulses with a time dependent frequency can be of importance. Typically, we decompose an electric field $\vec{E}$ into a slow envelope and a carrier wave (cf. left panel of Fig. 1.5):

$$\vec{E} = \vec{E} e^{i \omega_l t} + \vec{E}^* e^{-i \omega_l t}. \quad (1.33)$$

However, this carrier frequency can time dependent (cf. middle and right upper panel of Fig. 1.5). In the most simple case this time dependence is linear:

$$\omega_l(t) = \omega_l(t_0) + \kappa(t - t_0). \quad (1.34)$$

The time dependent part $\kappa$ is called chirp and for rising frequency $\kappa > 0$ it is called upchirp, whereas a falling frequency $\kappa < 0$ is denoted downchirp. In this work we will make use of rotating
Figure 1.5: Left panel: temporal profile of an optical pulse is decomposed into carrier frequency and the slow envelope. Upper panels: a chirped pulse has a time dependent frequency, rising frequencies (left) are called upchirp; falling frequencies (right) downchirp. Lower panels: The absolute value and imaginary part (corresponds to phase) of the envelopes of the chirped pulses.

frames, where all quantities are rotated by the carrier frequency of the electric field. For chirped pulses this rotating frame is done with the constant frequency $\omega_l(t_0)$, where $t_0$ is the position of the maximum of the pulse. The lower panels of Fig. 1.5 show the resulting envelope of an up- and down-chirp. The chirp adds a complex behavior to the envelope $E = |E|e^{i\kappa t}$. The absolute value of the envelope is independent of the chirp, while the imaginary part represents the phase $\text{Im}(E) = |E| \sin(\kappa t)$ and is showing rising frequencies for positive and negative times. Upchirp and downchirp can be discriminated by the sign of the phase. We will handle with chirped pulses in Chapters 3 and 4.

**Electric field Hamiltonian**

The electric field enters also in the Hamiltonian for the interaction with electrons. The derivation can be thoroughly done via the combined Maxwell–Schrödinger Lagrangian for the electro-magnetic potentials [Mil10]. From there to the widely used electric field coupling Hamiltonian that resembles the energy of a dipole in an electric field [HK04]:

$$H_{\text{el-field}} = \sum_{kl} \vec{d}_{kl} \cdot \vec{E} a_k^\dagger a_l.$$  \hspace{1cm} (1.35)

The dipole coupling element is defined in the same way as above.

This approach is called semi-classical, since the electric field is not quantized. Quantum optics with quantum dots [Jah12] is very interesting field of research, but out of scope of this work. For the description of the excitation of a quantum system the semi-classical is mostly sufficient.
1.3 Interactions – full systems’ Hamiltonian and equations of motion

The full Hamiltonian of the system consists of

\[ H = H_{\text{el}} \rightarrow \sum_i \epsilon_i a_i^\dagger a_i \] electronic eigenenergies, cf. Sec. 1.2.1

\[ + H_{\text{ph}} \rightarrow \sum_{\vec{q},\kappa} \hbar \omega_{\vec{q},\kappa} b_{\vec{q},\kappa}^\dagger b_{\vec{q},\kappa} \] phonon eigenenergies, i.e. phonon dispersion

\[ + H_{\text{el-ph}} \rightarrow \sum_{ij} \sum_{\vec{q},\kappa} g_{ij}^{\vec{q},\kappa} a_i^\dagger a_j^\dagger \left( b_{\vec{q},\kappa} + b_{-\vec{q},\kappa}^\dagger \right) \] electron-phonon interaction

\[ + H_{\text{el-light}} \rightarrow \sum_{ij} \vec{d}_{ij} \cdot \vec{E} a_i^\dagger a_j \] semi-classical interaction with an optical field

Using this Hamiltonian, dynamical equations for the quantum mechanical values can be derived via Heisenberg equations of motion.

\[ i\hbar \partial_t \hat{O} = [\hat{O}, H]. \] (1.36)

1.3.1 Electron–light system

For the electron light system \( \hat{H} = H_{\text{el}} + H_{\text{el-light}} \) the Heisenberg equation of motion approach results in the Bloch equations

\[ i\hbar \frac{\partial}{\partial t} a_i^\dagger a_j = (\epsilon_j - \epsilon_i) a_i^\dagger a_j + \sum_k \vec{E} \cdot (\vec{d}_{ij} a_i^\dagger a_k - \vec{d}_{ki} a_k^\dagger a_j), \] (1.37)

which is a closed system of coupled equations. In our scope, \( d_{ij} \) is non-diagonal. For two level systems, polarizations \( i \neq j \) couple to densities \( i = j \) via the electric field and vice versa. Those equations are deeply discussed in Chap. 3. In Chap. 2 we will also make use of them in a linear response theory.

1.3.2 Electron–phonon system

The equations for the electron-phonon system \( H = H_{\text{el}} + H_{\text{el-ph}} + H_{\text{ph}} \) are rather similar to the Bloch equations on first sight, but due to the quantized nature of the phonons, the hierarchy problem occurs:

\[ i\hbar \partial_t a_i^\dagger a_j = \hbar \omega_{ij} a_i^\dagger a_j + \sum_k (g_{ij}^{\vec{q},\kappa} a_i^\dagger a_k^\dagger - g_{kj}^{\vec{q},\kappa} a_k^\dagger a_j^\dagger) \left( b_{\vec{q},\kappa} + b_{-\vec{q},\kappa}^\dagger \right). \] (1.38)
1.3. INTERACTIONS – FULL SYSTEMS’ HAMILTONIAN AND EQUATIONS OF MOTION

The equation couples to phonon assisted operators of the kind $a_k^\dagger a_j b_{\vec{q},\kappa}^{(1)}$, whose dynamics would have to be calculated and couple to even higher order terms.

$$i\hbar \partial_t \langle a_k^\dagger a_j b_{\vec{q},\kappa} \rangle = (\hbar \omega_{ji} + \hbar \omega_{\vec{q},\kappa}) a_k^\dagger a_j b_{\vec{q},\kappa} + \sum_{k,k',\kappa'} (g_{jk}^{\vec{q},\kappa'} a_k^\dagger a_k - g_{kj}^{\vec{q},\kappa'} a_k^\dagger a_j) (b_{\vec{q},\kappa'} b_{\vec{q},\kappa} + b_{\vec{q},\kappa'}^\dagger b_{\vec{q},\kappa})$$

$$+ \sum_{kl} g_{lk}^{-\vec{q},\kappa} (a_k^\dagger a_k \delta_{ij} - a_k^\dagger a_j a_k) . \quad (1.39)$$

That means the dynamics are described by an infinite system of coupled equations. The hierarchy can be broken by factorization of higher order terms, thus neglecting correlations. Typical approximations are

- the one electron assumption, where $a_k^\dagger a_j a_j a_l$ vanishes exactly; An alternative treatment of the term is the Hartree-Fock factorization: $\langle a_k^\dagger a_j a_j a_l \rangle = \langle a_k^\dagger a_j \rangle \langle a_k^\dagger a_j \rangle - \langle a_k^\dagger a_j \rangle \langle a_k^\dagger a_l \rangle$, which is a mean field approach.

- factorization of electrons and phonons \( \langle a_k^\dagger a_j b_{\vec{q},\kappa'}^\dagger b_{\vec{q},\kappa} \rangle = \langle a_k^\dagger a_j \rangle \langle b_{\vec{q},\kappa'}^\dagger b_{\vec{q},\kappa} \rangle \)

- bath approximation for the phonons $\langle b_{\vec{q},\kappa'}^\dagger b_{\vec{q},\kappa} \rangle = \langle b_{\vec{q},\kappa'}^\dagger \rangle \langle b_{\vec{q},\kappa} \rangle = 0$.

We will now use those approximations to derive a rate equation for the densities in second order Born-Markovian approximation. The dynamics equation is written with the definitions $\rho_{ij} = \langle a_i^\dagger a_j \rangle$ and $n_{\vec{q},\kappa} = \langle b_{\vec{q},\kappa}^\dagger b_{\vec{q},\kappa} \rangle$ as

$$i\hbar \partial_t \langle a_i^\dagger a_j b_{\vec{q},\kappa} \rangle e^{i(\omega_{ji} + \omega_{\vec{q},\kappa})t} = \sum_k \left( g_{ij}^{\vec{q},\kappa'} \rho_{ik} (n_{\vec{q},\kappa} + 1) - g_{ki}^{\vec{q},\kappa'} \rho_{kj} n_{\vec{q},\kappa} \right)$$

$$- \sum_l g_{lk}^{-\vec{q},\kappa'} (\rho_{lk} \rho_{lj} + \rho_{lj} \rho_{lk} ) e^{i(\omega_{ji} + \omega_{\vec{q},\kappa})t} \quad (1.40)$$

which can be formally solved. To describe relaxation phenomena, it is convenient to neglect polarizations (non-diagonal terms) on the right side, since they represent spatial inhomogeneities.

$$i\hbar \langle a_i^\dagger a_j b_{\vec{q},\kappa} \rangle = \int_{-\infty}^{t} dt' \left( g_{ji}^{\vec{q},\kappa'} \rho_{ii} (n_{\vec{q},\kappa} + 1) - g_{jj}^{\vec{q},\kappa'} \rho_{jj} n_{\vec{q},\kappa} \right) e^{-i(\omega_{ji} + \omega_{\vec{q},\kappa})(t-t')} \approx g_{ji}^{\vec{q},\kappa'} \left( \rho_{ii} (n_{\vec{q},\kappa} + 1) - \rho_{jj} - \rho_{jj} n_{\vec{q},\kappa} \right) \int_{-\infty}^{t} dt' e^{-i(\omega_{ji} + \omega_{\vec{q},\kappa})(t-t')} \quad (1.41)$$

$$\pi \delta(\omega_{ji} + \omega_{\vec{q},\kappa}) + \frac{1}{\omega_{ji} + \omega_{\vec{q},\kappa}}$$

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Inserting this into Eq. (1.38) yields a closed equation, e.g. for the occupation of level \(i\):

\[
\imath \hbar \partial_t \rho_{ii} = \sum_{\vec{q}, \kappa} \sum_l \text{Im}(g_{li}^{\vec{q}, \kappa} \langle a_i^\dagger a_i b_{\vec{q}, \kappa} \rangle) - \sum_{\vec{q}, \kappa} \sum_l \text{Im}(g_{li}^{\vec{q}, \kappa} \langle a_i^\dagger a_i b_{\vec{q}, \kappa} \rangle)
\]

\[
= -\frac{i\pi}{\hbar} \sum_{\vec{q}, \kappa} \sum_l |g_{li}^{\vec{q}, \kappa}|^2 \left( \rho_{ii} (1 - \rho_{ii}) (n_{\vec{q}, \kappa} + 1) - \rho_{ii} (1 - \rho_{ii}) n_{\vec{q}, \kappa} \right) \delta(\omega_{li} + \omega_{\vec{q}, \kappa})
\]

\[
+ \frac{i\pi}{\hbar} \sum_{\vec{q}, \kappa} \sum_{lk} |g_{li}^{\vec{q}, \kappa}|^2 \left( \rho_{ll} (1 - \rho_{ii}) (n_{\vec{q}, \kappa} + 1) - \rho_{ii} (1 - \rho_{ii}) n_{\vec{q}, \kappa} \right) \delta(\omega_{il} + \omega_{\vec{q}, \kappa}).
\]

(1.42)

This rate equation is the Fermi golden rule. Both lines consist of an out-scattering part (\(\sim -\rho_{ii}\)) and an in-scattering part (\(\sim \rho_{ii}\)). The many electron contributions create blocking terms (1 - \(\rho\)) that inhibit scattering into occupied states. The delta distributions make the scattering processes energy conserving. The upper line can only be non-zero for \(\omega_{li} < 0\), i.e. \(\epsilon_i > \epsilon_l\), correspondingly the out-scattering from \(i\) is proportional to phonon emission (the emission term can be identified by the +1 due to spontaneous emission). The lower line represents cases, where \(\epsilon_l > \epsilon_i\), thus the in-scattering is related to the phonon emission.

In this work we will deal with the independent boson model (IBM) in Sec. 2.1.1, with only diagonal coupling to phonons \(g_{ki}^{\vec{q}, \kappa} \sim \delta_{ki}\), which has an exact solution for the polarization in linear optics, i.e. every order of the many particle problem is included in the solution. In Sec. 2.2 we derive an effective higher order electron-phonon Hamiltonian where the non-diagonal coupling plays the major role, and the hierarchy can be interrupted sooner.
Phonon influence on carrier dynamics in QDs

In this work the optical response of QDs plays a major role. This response is determined by the microscopic dynamics of the carriers in the system. In the semiconductor environment phonons, the quantized lattice vibrations, have a significant influence on the carrier dynamics. This chapter describes the impact of electron-phonon interaction on the dynamics and resulting effects. On one hand interaction with phonons yields decoherence, i.e. the loss of microscopic transitions probabilities, which can be directly seen in optical spectra. Section 2.1 discusses how luminescence or absorption spectra are influenced by the electron-phonon interaction and discusses the impact of phonon modes, carrier wave functions and phonon coupling strength. Here, calculations can be easily compared to experiments. On the other hand the phonon system takes excess energy of relaxing carriers (or gives energy to excite carriers in the opposite process). Section 2.2 discusses how carriers from the reservoir are captured, i.e. relax into the QD states, via interaction with optical phonons. For that aim we present an effective multi-phonon Hamiltonian derived via a projection operator technique. This capture process and the dependence on temperature and carrier reservoir density are described.

2.1 Linear absorption and luminescence spectra

Optical spectra are a traditional and versatile way to obtain information about the structure and properties of matter. One of the most simple approaches are absorption spectra. They are commonly described by the linear\(^1\) response of matter to optical light fields defined via the absorption coefficient

\(^1\)In general optical response is a non-linear function of the optical field. Typically, weak probing fields are used, that can be approximated to not excite the system and thus yields only a linear response.
\( \alpha(\omega) \) from Beer-Lambert law \( |E(z, \omega)|^2 = |E(0, \omega)|^2 e^{-\alpha(\omega)z} \). We consider an optical field \( E(\omega, z) = E_0(\omega)e^{ikz - \frac{\omega}{c}z} \) traveling through a material with linear response \( P(\omega) = \varepsilon_0\chi(\omega)E(\omega) \). The typical wave equation Eq. (1.32) in frequency domain reads

\[
\Delta E(\omega) + \frac{\omega^2}{c^2} E(\omega) = -\frac{\omega^2}{\varepsilon_0 c^2} P(\omega) = -\frac{\omega^2}{c^2} \chi(\omega)E(\omega).
\]

By inserting the ansatz mentioned above, the wave number \( k \) as well as for the absorption \( \alpha \) are related to the susceptibility \( \chi \):

\[
0 = (ik - \frac{\alpha}{2})^2 + \frac{\omega^2}{c^2} (1 + \chi(\omega)).
\]

Expressions for \( k \) and \( \alpha \) are derived via splitting the equation into real and imaginary part

\[
\alpha(\omega) = \frac{\omega^2}{c^2}\text{Im}(\chi(\omega)) \cdot \frac{1}{k(\omega)}, \quad k^2 = +\frac{\alpha^2}{4} + \frac{\omega^2}{c^2} (1 + \text{Re}(\chi(\omega))),
\]

\[
\Rightarrow \alpha^2 = 2\frac{\omega^2}{c^2} (1 + \text{Re}(\chi)) \left[ \sqrt{1 + \frac{\text{Im}(\chi)^2}{(1 + \text{Re}(\chi))^2}} - 1 \right]. \tag{2.1}
\]

To understand this further it is convenient to investigate the typical behavior of susceptibilities. In Sec. 1.2.3 we showed the polarization is composed of all microscopic transition probabilities.

\[
\varepsilon_0\chi(\omega)E(\omega) = P(\omega) = \sum_{ij} d_{ij} \rho_{ij}(\omega) + c.c. \tag{2.2}
\]

The most simple approximation for semiconductors treat every transition as independent two level system and investigate their linear response. Here, indirect transitions are neglected, i.e. every point in the band structure is a free two level system with the dynamics in frequency domain (assuming constant occupations):

\[
-i\omega \rho_{ij}(\omega) = -i(\omega_i - \omega_j - i\gamma) \rho_{ij}(\omega) + \frac{d_{ij} E(\omega)}{\hbar} (\rho_{jj} - \rho_{ii}), \tag{2.3}
\]

\[
P = \sum_{ij} \frac{|d_{ij}|^2 (\rho_{jj} - \rho_{ii})}{\hbar} \left( \frac{\omega - (\omega_j - \omega_i) - i\gamma}{(\omega - (\omega_j - \omega_i))^2 + \gamma^2} - \frac{\omega + (\omega_j - \omega_i) + i\gamma}{(\omega + (\omega_j - \omega_i))^2 + \gamma^2} \right) E. \tag{2.4}
\]

The imaginary part contains Lorentzian peaks at the systems resonances, while the real part has discontinuities and decays only slowly. The dipole elements determine the strength and include the density of states. \( \rho_{jj} - \rho_{ii} \) is the inversion and decides whether the response absorbs or enhances incoming light. Figure 2.1 shows the susceptibilities of a two level system as well as of the GaAs band structure, both in the ground state.
CHAPTER 2. PHONON INFLUENCE ON CARRIER DYNAMICS IN QDS

Figure 2.1: Susceptibilities of a two level system and of the semiconductor GaAs (data from Ref. [Ref]). The real part represents the refractive index and describes the scattering of light, whereas the imaginary part determines absorption/gain. For a bulk system the susceptibilities add up, since the real part decreases only slowly away from the resonance there is a significant contribution below the band gap.

Our primary interest is the response from QDs, with transition frequencies below the band gap of the host bulk semiconductor. Thus, the response is divided \( \chi = \chi_{\text{active}} + \chi_{\text{bg}} \). The imaginary part of the response of the background material is off-resonant, when investigating the active material, and does not contribute, whereas the real part response of the background almost always gives a contribution and is larger than the real part response of the smaller active system. Hence, \( \text{Im}(\chi) \approx \text{Im}(\chi_{\text{active}}) \) and \( \text{Re}(\chi) \approx \text{Re}(\chi_{\text{bg}}) \). Typically, \( (1 + \text{Re}(\chi_{\text{bg}})) \gg \text{Im}(\chi_{\text{active}}) \), then from Eq. (2.1) follows the absorption coefficient can be calculated via

\[
\alpha(\omega) \approx \frac{\omega}{c} \text{Im}(\chi_{\text{active}}(\omega)) \frac{1}{\sqrt{1 + \text{Re}(\chi_{\text{bg}}(\omega))}}.
\]  

(2.5)

The wave number of the traveling light field is independent of absorption contributions:

\[
k(\omega) \approx \frac{\omega}{c} \sqrt{1 + \text{Re}(\chi_{\text{bg}}(\omega))}.
\]  

(2.6)

The linear optical response to an electric field is derived from the polarization \( \chi(\omega) = \frac{P(\omega)}{E(\omega)} \), which is obtained from the dynamics of the microscopic polarization.

The temporal behavior of the polarization does not only have a signature in absorption spectra, but also in luminescence spectra. Generally, to describe luminescence one needs a quantum optical

\[\lambda = \frac{2\pi c}{\omega}; \quad \frac{\partial \lambda}{\partial \omega} = -\frac{\lambda}{\omega}; \quad k(\omega) = \frac{\omega}{c} n(\omega).\]  

2The wave number describes the refractive behavior of the material. The optical properties are often described in terms of the refractive index and the wave length. For comparison with the literature we list here some useful relations:
2.1. LINEAR ABSORPTION AND LUMINESCENCE SPECTRA

treatment, since essentially spontaneous emission dominates this effect. The luminescence spectrum is [SZ97]:
\[ S(r, \omega) = \frac{1}{\pi} \Re \int_0^{\infty} d\tau \left( E^-(r, t) E^+(r, t + \tau) \right) e^{i\omega \tau}, \]

with the quantum optical electric field operators \( E^+, E^- \), that describe the emission. The spectrum is the inverse Fourier transform with respect to \( \tau \). The expectation value from the field operators relates to the dynamics of the microscopic polarizations
\[ \left\langle E^-(r, t) E^+(r, t + \tau) \right\rangle = I_0 \left\langle \sigma_+(t) \sigma_-(t + \tau) \right\rangle. \]

The polarization \( \sigma_+ = a_\uparrow^i a_i \) defines the initial condition at \( t \) and the evolution of \( \sigma_- = a_\downarrow^i a_i \) is responsible for the spectrum. For a two level system, that is prepared in the pure state \( |2\rangle \langle 2| \) this would mean the spectrum is defined by the temporal evolution of \( \sigma_- = a_\downarrow^2 a_1 \). As we will see, absorption and luminescence spectra show the same behavior, but with mirrored frequency.

Next, we will show absorption/luminescence spectra of a QD system with phonon influence by calculating the polarization dynamics under the influence of a phonon bath.

2.1.1 Independent Boson model

QDs interacting with phonons are commonly modeled by the so called “Independent Boson Model”(IBM), which describes the polarization of an electronic two level system without change of the occupation under interaction with a bath of phonons [Mah90]. Within this approximations the model has an exact solution. The model is widely used for systems with discrete states like QDs (as in this work) [KAK02; KAK+05; Web08] or molecules [RAK+06]. This section briefly derives the well known solution of the IBM,\(^3\) while Sec. 2.1.2 shows the properties of the phonon interaction in QDs with the help of the IBM solutions and Sec. 2.1.3 applies this to real systems and compares to experimental data.

Start with the Hamiltonian from Sec. 1.3:
\[ H = \sum_i \epsilon_i a_\uparrow^i a_i + \sum_q \hbar \omega_q b_q^\dagger b_q + \sum_{ij} V_{ij} a_\uparrow^i a_j, \]
\[ V_{ij} = \sum_q g_{ij}^q (b_q + b_q^\dagger). \]

The Heisenberg equation of motion approach yields the dynamics equation for the microscopic polarization:
\[ i\hbar \partial_t a_k^\dagger a_l = (\epsilon_l - \epsilon_k) a_k^\dagger a_l + \sum_i \left( V_{li} a_k^\dagger a_i - V_{ik} a_k^\dagger a_i \right) \]

\(^3\)The derivation of the IBM solution can be done in a more general way as presented here using propagators [Muk95], however we restrict ourselves to a simple derivation stressing the most important assumptions.
The equations for all polarizations are decoupled. We define the effective coupling element
\[ V_{kl} = V_{kl}(t) = a_k(t)e^{i(\omega t - \omega_k)t}. \]

\[ p_{kl} = p_{kl}(0) - \frac{i}{\hbar} \int_0^t dt_1 \sum_k (V_{ki}(t_1)e^{i(\omega t - \omega_i)t_1}p_{ki}(t_1) - V_{ik}(t_1)e^{i(\omega t - \omega_k)t_1}p_{lk}(t_1)) \tag{2.12} \]

Here, we restrict ourselves to diagonal coupling \( V_{ij} \sim \delta_{ij} \). Further, to take into account initial conditions later on, we have to investigate the effect on the statistical operator \( \rho \).

\[ p_{kl}(t)\rho = p_{kl}(0)\rho - \frac{i}{\hbar} \int_0^t dt_1 (V_{kl}(t_1) - V_{kk}(t_1)) p_{kl}(t_1)\rho. \tag{2.13} \]

The equations for all polarizations are decoupled. We define the effective coupling element \( V_{kl} = V_{ll} - V_{kk} \) which is composed of the diagonal coupling elements of the interacting states. The dynamic equation for the polarization can be formally solved via a Dyson series, which means the iterative replacement of \( p_{kl} \) on the right side with the formal solution. This results in a sum over every order of \( V \). Since the phonons are assumed to be a thermalized bath, odd orders of \( V \) vanish.

\[ \langle p_{kl} \rangle (t) = \langle p(0) \rangle \sum_{n=0}^{\infty} \left( \frac{-i}{\hbar} \right)^n \int_0^t dt_1 \int_0^{t_1} dt_2 \cdots \int_0^{t_{2n-1}} dt_{2n} \langle V_{kl}(t_1)V_{lk}(t_2) \cdots V_{kl}(t_{2n}) \rangle = \langle p_{kl}(0) \rangle \sum_{n=0}^{\infty} p_{kl}^{(n)}(t) \tag{2.14} \]

For this simple problem with diagonal coupling only, we can use the solid state variant of Wick’s theorem [Mah90], where

\[ p^{(n)} = \frac{1}{n!} \left( p^{(1)} \right)^n. \]

In conclusion, using Wick’s theorem leads exactly to \( p(t) = p(0)e^{p^{(1)}(t)} \). For a more complex problem, one would use the cumulant expansion [Ric07].

To get grasp of the experimentally accessible absorption, the polarization and the exciting light field have to be processed \( \chi(\omega) = \frac{P(\omega)}{E(\omega)} \). For practical use the interaction of electrons with the light field on the one hand and all other interactions (in this work: phonons) on the other are temporally separated [Web08]. In order to do so the light field is modeled as a delta peak \( E(t) = E_0\delta(t) \) preparing the electronic system, before the other interactions kick in at \( t = 0 \).

\[ p(t \leq 0) = iE_0\theta(t), \quad p(t > 0) = p(0)e^{p^{(1)}(t)}. \tag{2.15} \]

This leads to the following expression for the macroscopic polarization:

\[ P(t) = \sum_{ij} d_{ij} \text{Tr} \left( a_i^\dagger a_j \rho \right) = d_{12}e^{i(\omega_2 - \omega_1)t}p(0)e^{p^{(1)}(t)} = iE_0d_{12}e^{i(\omega_2 - \omega_1)t}e^{p^{(1)}(t)}. \]

\[ \text{This starting condition can be expressed in a more elegant way using a description based on propagators.} \]
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Thus, the absorption can be calculated from the polarization dynamics via

$$\alpha(\omega) \sim \frac{\omega}{e} \text{Re}(\mathcal{F}(e^{\nu(t)})) = f((\omega_2 - \omega_1) - \omega).$$

In the same way the luminescence is described by

$$S(\omega) \sim \text{Re}(\mathcal{F}^{-1}(e^{\nu(t)})) = f((\omega_2 - \omega_1) + \omega),$$

according to Eq. (2.7). We see that luminescence and absorption have nearly the same behavior, but with mirrored frequencies around the transition energies $\hbar(\omega_2 - \omega_1)$.

For the first order term one needs to evaluate:

$$\langle V(t_1)V(t_2) \rangle = \sum_{qq'} g(q,\kappa) g(q',\kappa) \langle (b_q(t_1) + b^\dagger_{-q}(t_1))(b_q(t_2) + b^\dagger_{-q}(t_2)) \rangle. \tag{2.16}$$

In order two work with the two-time operators we need their temporal evolution. The dynamics can be derived again via Heisenberg equation of motion. Here it is important to note, that the evolution equation for the phonon operators is fed by electronic occupations, which leads to the problem, that even if the system is in its ground state, the phonon dynamics would be driven. This problem is overcome by the Weyl transformation [Web08] or using the electron-hole picture. However, we are using the bath approximation for the phonons via assuming the coupling in the phonon equations of motion vanishes anyways.

$$i\hbar \partial_t b_{q,\kappa} = \hbar \omega_{q,\kappa} b_{q,\kappa} \Rightarrow b_{q,\kappa}(t) = b_{q,\kappa}(0)e^{-i\omega_{q,\kappa}t}, b^\dagger_{q,\kappa}(t) = b^\dagger_{q,\kappa}(0)e^{i\omega_{q,\kappa}t}. \tag{2.17}$$

Additionally we insert a phenomenological decay $\gamma$ of the phonon dynamics.\footnote{This decay represents phonon-phonon interaction, where a phonon decays into phonons of other modes. How such influence is incorporated microscopically can be found in [SZ97] as "Heisenberg-Langevin" approach.} Temperature dependent decay rates can be found for example in Refs. [BKS94; RRB06]. The dynamics of the phonons is inserted and the time integral can be solved:

$$\langle V(t_1)V(t_2) \rangle \approx \sum_q |g(q,\kappa)|^2 \left\{ \langle b_{q,\kappa} b^\dagger_{q,\kappa} \rangle e^{(-i\omega_{q,\kappa}-\gamma)(t_1-t_2)} + \langle b^\dagger_{q,\kappa} b_{q,\kappa} \rangle e^{(i\omega_{q,\kappa}-\gamma)(t_1-t_2)} \right\}. \tag{2.18}$$

The temporal integrations now include only exponential functions and are thus easy to solve:

$$\int_0^t dt_1 \int_0^{t_1} dt_2 e^{(\pm i\omega_{q,\kappa}-\gamma)(t_1-t_2)} = -\int_0^t dt_1 \frac{1}{\pm i\omega_{q,\kappa} - \gamma} (1 - e^{(\pm i\omega_{q,\kappa}-\gamma)t_1})$$

$$= -\frac{1}{\pm i\omega_{q,\kappa} - \gamma} + \frac{1}{(\pm i\omega_{q,\kappa} - \gamma)^2} e^{(\pm i\omega_{q,\kappa}-\gamma)t} - 1. \tag{2.19}$$
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This yields the solutions for the cumulant of the phonon dynamics

$$p^{(1)}(t) = -\sum_q |g_q|^2 \left\{ \langle b_q b_q^\dagger \rangle \left[ \frac{t}{\omega_q + \gamma} + \frac{1}{(i\omega_q + \gamma)^2} (e^{-i\omega_q t - \gamma t} - 1) \right] + \langle b_q^\dagger b_q \rangle \left[ -\frac{t}{i\omega_q - \gamma} + \frac{1}{(i\omega_q - \gamma)^2} (e^{i\omega_q t - \gamma t} - 1) \right] \right\}.$$  \hfill (2.20)

The solution can be decomposed into various parts with different meaning:

$$p(t) = p(0) \exp \{ i(-\omega_g - \Delta_1)t - \Delta_2 t + R(t) - S_1 - iS_2 \}$$  \hfill (2.21)

with the two contributions linear to time

$$\Delta_1 = -\sum_{\vec{q}\kappa} \frac{|g_{\vec{q}\kappa}|^2}{\hbar^2} \frac{\omega_{\vec{q}\kappa}}{\omega_{\vec{q}\kappa}^2 + \gamma_{\vec{q}\kappa}^2} \quad \text{Polaron shift,} \quad \hfill (2.22a)$$

$$\Delta_2 = -\sum_{\vec{q}\kappa} \frac{|g_{\vec{q}\kappa}|^2}{\hbar^2} \frac{\gamma_{\vec{q}\kappa}}{\omega_{\vec{q}\kappa}^2 + \gamma_{\vec{q}\kappa}^2} (2n_{\vec{q}\kappa} + 1) \quad \text{Zero-phonon line broadening.} \quad \hfill (2.22b)$$

Those terms shift the spectrum (a) or broaden the else delta shaped zero phonon line temperature dependent (b), respectively. However the latter term is absent, if the phonon decay $\gamma$ is not taken into account. The next term is the only one with a complex temporal behavior and depending on the phonon occupation and thus responsible for the shape of phonon side bands:

$$R(t) = -\sum_{\vec{q}\kappa} \frac{|g_{\vec{q}\kappa}|^2}{\hbar^2} \left[ \frac{(n_{\vec{q}\kappa} + 1)}{(i\omega_{\vec{q}\kappa} + \gamma_{\vec{q}\kappa})^2} e^{-(i\omega_{\vec{q}\kappa} + \gamma_{\vec{q}\kappa}) t} + \frac{n_{\vec{q}\kappa}}{(i\omega_{\vec{q}\kappa} - \gamma_{\vec{q}\kappa})^2} e^{(i\omega_{\vec{q}\kappa} - \gamma_{\vec{q}\kappa}) t} \right]. \quad \hfill (2.22c)$$

The part proportional to $n_{\vec{q}\kappa} + 1$ represents phonon emission, while the other term corresponds to phonon absorption. Its meaning gets clearer by investigation of its role in the polarization, for that the exponential is expanded into a series [MK00]:

$$p(\omega - \omega_g - \Delta_1) \sim \mathcal{F} \left( e^{R(t)} \right) = \sum_{n=0}^{\infty} \frac{1}{n!} \mathcal{F}(R(t)^n) .$$

The counter $n$ has the meaning of the number of interactions with the phonon system. This can be nicely seen for optical phonons in Einstein approximation $\omega_{\vec{q}LO} = \omega_{0,LO}$ in the zero temperature limit $n_{\vec{q}\kappa} = 0$:

$$R(t)^n|_{LO} = R(0)^n e^{-(i\omega_{\vec{q}\kappa} + n\gamma_{\vec{q}\kappa}) t} ,$$

$$\mathcal{F}(R(t)^n e^{-\Delta_2 t}) = R(0)^n \frac{1}{\sqrt{2\pi}} \frac{i(\omega - n\omega_{\vec{q}\kappa}) + \Delta_2 + n\gamma_{\vec{q}\kappa}}{(\omega - n\omega_{\vec{q}\kappa})^2 + (\Delta_2 + n\gamma_{\vec{q}\kappa})^2} .$$
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The spectrum in this case consists of Lorentzian lines (or delta shaped lines for the $\gamma \to 0$ case, which is consistent with zero temperature) with the distance of an LO phonon energy, starting with the zero phonon line at $\omega_g + \Delta_1$. For the $\gamma \to 0$ case they have a size ratio of

$$\frac{p(n\omega_{\text{LO}})}{p(0)} = \frac{1}{n!} R(0)^n; \quad T = 0 \text{ K},$$

which means they are Poisson distributed with the mean value of $R(0)$, i.e. the highest line in the spectrum is the order $n$ closest to $R(0)$. This so called Huang-Rhys factor $R(0)$ [HR50; MK00] is a measure for the coupling strength and easily accessible in experiments. The last terms of Eq. (2.21) represent the Huang-Rhys factor $R(t = 0) = S_1 + iS_2$ and ensure the correct starting condition.

$$S_1 = \sum_{\vec{q}_c} \left| g_{\vec{q}_c}^\text{g} \right|^2 \frac{\omega_{\vec{q}_c}^2}{h^2} \left( \frac{\omega_{\vec{q}_c}^2 + \gamma_{\vec{q}_c}^2}{\omega_{\vec{q}_c}^2 + \gamma_{\vec{q}_c}^2} \right)^2 (2n_{\vec{q}_c} + 1) \quad \text{Huang-Rhys factor, (2.22d)}$$

$$S_2 = \sum_{\vec{q}_c} \left| g_{\vec{q}_c}^\text{g} \right|^2 \frac{2\omega_{\vec{q}_c} \gamma_{\vec{q}_c}}{h^2} \left( \frac{\omega_{\vec{q}_c}^2 + \gamma_{\vec{q}_c}^2}{\omega_{\vec{q}_c}^2 + \gamma_{\vec{q}_c}^2} \right)^2. \quad \text{(2.22e)}$$

Often the IBM solution is presented/used without the phonon decay rate $\gamma$.

$$p^{(1)}(t) = \sum_{q} \left| g_{q}^\text{g} \right|^2 \frac{1}{h^2 \omega_q} \left\{ (n_q + 1) \left( e^{-i\omega_q t} - 1 \right) + n_q \left( e^{i\omega_q t} - 1 \right) + i\omega_q t \right\}. \quad \text{(2.23)}$$

Especially for this case it is convenient to define a spectral density [Muk95; RAK+06]: 6

$$J_\kappa(\omega) = \sum_{\vec{q}} \left| g_{\vec{q}}^\kappa \right|^2 \frac{1}{h^2 \omega_{\vec{q}}} \delta(\omega - \omega_\kappa(\vec{q})) = \frac{V}{2 \pi^3} \int \frac{d\vec{q}}{h^2 \omega_{\vec{q}}} \left| g_{\vec{q}}^\kappa \right|^2 \delta(\omega - \omega_\kappa(\vec{q})). \quad \text{(2.24)}$$

This quantity links the coupling strength for every phonon wave vector $\vec{q}$ to a frequency in the spectrum depending on the phonon mode’s dispersion relation. The spectral density contains all information but the temperature (respective the phonon occupation). In the $T = 0 \text{ K}$ limit $R(t)$ can be directly expressed as the Fourier transform of the spectral density:

$$R(t) = \sum_{\kappa} \int d\omega J_\kappa(\omega) e^{-(i\omega)t} = \mathcal{F}^{-1}(J(-\omega)). \quad \text{(2.25)}$$

That means in the low temperature limit the spectrum is defined by the spectral density.

$$p(\omega) \sim \mathcal{F} \left( e^{i(-\omega_g - \Delta_1)t - S_1} e^{\mathcal{F}^{-1}(f(\omega ))} \right) = e^{-S_1} \mathcal{F} \left( e^{\mathcal{F}^{-1}(J_\kappa(\omega - \omega_g - \Delta_1))} \right)$$

$$\omega' = \omega - \omega_g - \Delta_1 = e^{-S_1} \mathcal{F} \left( \sum_i \frac{1}{i!} (\mathcal{F}^{-1}(J_\kappa(\omega')))^i \right) \approx e^{-S_1} \delta(\omega') + e^{-S_1} J_\kappa(\omega'). \quad \text{(2.26)}$$

6Note, that in the literature the power of $\omega$ in the definition can vary.
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The absorption spectrum is approximative the delta like zero phonon line plus the spectral density from the phonons (one-phonon contribution). For optical phonons that means again \( J_{\text{LO}}(\omega) = S_1 \delta(\omega - \omega_{\text{LO}}) \). For acoustic phonons it is a continuous band showing the coupling strength depending on the phonon energy. We will dive deeper into the shape and meaning of the spectral density on the next pages.

In the high temperature limit \( n_{qk} \gg 1 \) the spectrum becomes symmetric around the zero phonon line \( p(\omega') = p(-\omega') \).

2.1.2 Diagonal coupling due to various interaction mechanisms

In this part we will investigate the spectral density (Eq. (2.24)) caused by deformation potential and piezoelectric coupling to acoustic phonons, as well as polar coupling to optical phonons. Those coupling mechanisms are introduced in Sec. 1.2.2. One major influence to the spectral density are the QD’s wave functions. Here, we choose Gaussian QD model wave functions

\[
\varphi_v(\vec{r}) = \sqrt{\frac{1}{a_v^2 \pi^3}} e^{-\frac{\vec{r}^2}{2a_v^2}}, \quad \varphi_c(\vec{r}) = \sqrt{\frac{1}{a_c^2 \pi^3}} e^{-\frac{\vec{r}^2}{2a_c^2}}. \tag{2.27}
\]

Where \( a_v (a_c) \) denote the extent of the valence band (conduction band) wave functions. We introduce the distance of the centers of mass of valence and conduction band wave functions \( \vec{r}_0 \) and thus describes a dipole moment of the QD. Those model wave functions can be further generalized to an anisotropic form. The advantage of the chosen simple wave functions is the possible analytical treatment which displays fundamental properties of the coupling.

**Deformation potential coupling of acoustic phonons** According to Sec. 1.2.2 the effective deformation potential coupling element for Gaussian wave functions reads

\[
g(\vec{q})_{\text{LA}} = \sqrt{\frac{h \vec{q}^2}{2D_V \omega_{\text{LA}}(\vec{q})}} \left( D_v e^{-\frac{a_v^2 \vec{q}^2}{4}} e^{i \vec{q} \cdot \vec{r}_0} - D_c e^{-\frac{a_c^2 \vec{q}^2}{4}} \right). \tag{2.28}
\]

For the spectral density (Eq. (2.24)) we need the absolute square of this function

\[
|g(\vec{q})|_{\text{LA}}^2 := \frac{h \vec{q}^2}{2D_V \omega_{\text{LA}}(\vec{q})} \left( D_v^2 e^{-\frac{a_v^2 \vec{q}^2}{4}} + D_c^2 e^{-\frac{a_c^2 \vec{q}^2}{4}} - 2D_v D_c e^{-\frac{(a_v^2 + a_c^2) \vec{q}^2}{4}} \cos(|\vec{q}| |\vec{r}_0| \cos \theta_{\vec{q}}) \right). \tag{2.29}
\]

Here, we can see that the coupling is induced by the energy change due to deformation in valence and conduction band and a mixed term that depends on the distance of the localized conduction and valence electron density. The \( \cos \theta \) describes, the part of the phonon wave that travels between electron and hole wave function. For long wavelength the difference to the case \( \vec{r}_0 = 0 \) is not significant. However, for small QDs larger wave numbers \( \vec{q} \) are important. This is due to the Fourier character of the coupling element. Later on we will assume isotropic dispersion relations.
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\( \omega_{LA}(\vec{q}) = \omega_{LA}(q) \) and do not investigate the angular dependence of phonons or light, thus we are only interested in the angular average of this quantity:

\[
|g(q)_{LA}|^2_{DF} := \frac{1}{4\pi} \int_0^{2\pi} d\phi \int_0^{\pi} d\theta \sin \theta |g(\vec{q})_{LA}|^2 = \frac{\hbar}{2gV} \frac{q}{\omega_{LA}(q)} q \left( D_v^2 e^{-\frac{a_v^2\vec{q}^2}{2}} + D_c^2 e^{-\frac{a_c^2\vec{q}^2}{2}} - 2D_vD_c e^{-\frac{(a_v^2+a_c^2)\vec{q}^2}{4}} \sin q|\vec{r}_0| \right).
\]

This already contains the most important properties of the typical phonon interaction strength.

![Figure 2.2: Electron LA phonon interaction strength via deformation potential coupling. Left panel: influence of the wave function size on the coupling strength. Small QDs have a large extent in the \( q \) domain; due to the prefactor linear in \( q \) such couplings have a higher interaction strength. The right panel shows the influence of a built in dipole due to a distance between the wave functions, which has a significant influence for \( q \approx \frac{\pi}{r_0} \).](image)

In Debye approximation \( \omega_{LA}(q) = c_{LA}q \) the \( q \) dependence of the prefactor is linear. Without loss of generality, assume \( a_v \leq a_c \) (for the other case the indices \( c, v \) have to be switched). The \( q \) dependence can be written as

\[
|g(q)_{LA}|^2_{DF} \sim D_v^2 q e^{-\frac{a_v^2\vec{q}^2}{2}} \left( 1 - \frac{D_c}{D_v} e^{-\frac{a_v^2\vec{q}^2}{2}} \right)^2 + 2D_cD_v e^{-\frac{a_v^2\vec{q}^2}{2}} \left( 1 - \frac{\sin qr_0}{qr_0} \right).
\]

The behavior for different QD wave function sizes and distances between the wave functions is show in Fig. 2.2. In general one has the properties:

- For \( q = 0 \) (displacement of the whole solid) the coupling vanishes

- For large \( q \) the coupling decays as \( q e^{-\frac{a_v^2\vec{q}^2}{2}} \), \( a = \min(a_v, a_c) \), i.e. the smallest parts of the wave functions define the short wave length behavior.
For medium q there is one or two maxima (in the case \( r_0 = 0 \)) depending on the size of wave functions and deformation potential in valence and conduction band. The two maxima case (as presented in Ref. [Web08]) can occur if the deformation potentials have the same sign \( \text{sgn}(D_v) = \text{sgn}(D_c) \), \( a_v < a_c \), and \( |D_c| > |D_v| \) (or \( a_v > a_c \) and \( |D_c| < |D_v| \)). For equally sized wave functions \( a = a_v = a_c \) the maximum is at \( q_{\text{max}} = \frac{1}{a} \). For unequal sizes this maximum shifts towards zero. The size of the maximum \( |g(q_{\text{max}})|^2 \) is proportional to \( \frac{1}{a} \), i.e. smaller QDs have a larger phonon interaction.

A distance \( r_0 \) between the wave functions introduces a correction, that can lead to additional oscillations in a range dictated by the extent of the larger wave function. The influence of the correction is large for \( q \approx \frac{\pi}{r_0} < \frac{1}{a} \), which means the correction is important if the wave functions are strongly confined and separated, which can occur in a polar material.

### Piezoelectric coupling to acoustic phonons

The other interaction mechanism with acoustic phonons covered in this work is the piezoelectric coupling, cf. Sec. 1.2.2.

\[
\begin{align*}
g_\kappa(q) &= \sqrt{\frac{\hbar}{2\varrho V \omega_\kappa(q)}} \left( e^{-\frac{a_v^2 q^2}{4}} e^{i\vec{q}\cdot\vec{r}_0} - e^{-\frac{a_c^2 q^2}{4}} \right) F_\kappa(q), \\
|g_\kappa(q)|^2_{\text{PZ}} &= \frac{\hbar F_\kappa^2(q)}{2\varrho V \omega_\kappa(q)} \left( e^{-\frac{a_v^2 q^2}{2}} + e^{-\frac{a_c^2 q^2}{2}} - 2 e^{-\frac{(a_v^2+a_c^2)q^2}{4}} \cos(q|r_0| \cos \theta_q) \right).
\end{align*}
\]

Here, the coupling strength is defined by the piezoelectric factors \( F_\kappa \), which depend on the orientation but not on the absolute value of \( \vec{q} \). This angular dependence is different for different lattice symmetries, thus the typical lattice symmetries Zincblende and Wurtzit are two independent cases. An angular dependence of the part containing the wave function is introduced by a non-vanishing \( \vec{r}_0 \) (for the chosen spherical wave functions). Thus, the coupling can be divided in a part where the wave function influence is angular independent and the angular dependent part.

\[
\begin{align*}
|g_\kappa(q)|^2_{\text{isometric}} &= \frac{\hbar F_\kappa^2(q)}{2\varrho V \omega_\kappa(q)} q \left( e^{-\frac{a_v^2 q^2}{2}} + e^{-\frac{a_c^2 q^2}{2}} - 2 e^{-\frac{(a_v^2+a_c^2)q^2}{4}} \right), \\
|g_\kappa(q)|^2_{\text{angular}} &= \frac{\hbar F_\kappa^2(q)}{2\varrho V \omega_\kappa(q)} q e^{-\frac{(a_v^2+a_c^2)q^2}{4}} \left( 1 - \cos(q|r_0| \cos \theta_q) \right).
\end{align*}
\]

\( F_\kappa^2 \) denotes the angular average of the piezoelectric coupling factor. First, we investigate the part of the coupling where only the piezoelectric factors carry the angular dependence (corresponding to the \( r_0 = 0 \) case). The angular average will later on turn them into numbers that define the coupling strength to the different modes. The QD size dependence of the piezoelectric coupling is shown in Fig. 2.3. Here, the following general properties are valid:

- For equal sizes of the wave functions, the coupling vanishes
- For \( q = 0 \) the coupling vanishes
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Figure 2.3: Piezoelectric coupling strength for different QD wave functions. Left panel: wave functions with a larger spatial extent, have a maximum at small \( q \) and decay fast. Due to the \( 1/q \) prefactor, small wave functions have a small coupling strength. The right panel shows the influence of the dipole moment (distance between electron and hole center of mass) on the coupling strength. A dipole moment enhances the piezoelectric phonon coupling and can lead to oscillations for high \( q \).

- For large \( q \) the coupling decays as \( \frac{1}{q} e^{-\frac{a}{q^2}} \), \( a = \min(a_v, a_c) \), i.e. the smallest parts of the wave functions define the short wave length behavior.

- The coupling has one maximum between zero and \( 2 \sqrt{\ln\left(\frac{a_c^2}{a_v^2}\right) a_v^2 - a_c^2} \) (which is where the maximum of the terms in brackets without \( 1/q \) dependence occurs).

- For a global scaling \( a_v \sim a_{\text{scale}}, a_c \sim a_{\text{scale}} \) the position of the maximum has a dependence \( q_{\text{max}} \sim \frac{1}{a_{\text{scale}}} \), i.e. for smaller QDs the maximum shifts to larger \( q \). Subsequently the value of the maximum becomes smaller for smaller QDs, \( |g(q_{\text{max}})|^2 \sim a_{\text{scale}} \).

The angular dependent part, which is introduced by a non-vanishing dipole moment \( r_0 \), is rather complicated, first the growth direction (defining the direction of the dipole) of the QD in respect to the lattice symmetry has to be taken into account, second the result is different for every mode and every crystal symmetry, then the result of angular averaging are composed from products of polynomials times sine and cosine functions of \( qr_0 \). The results are shown in App. A. Since it is not as easy to show general properties with those analytical results, we want to show numerically the typical behavior in Fig. 2.3b. Generally the term can enhance the phonon coupling for higher \( q \).

**mixed contributions to LA phonon coupling** For \( |g_{\text{LA}}(q)|^2 \) there is also the mixed deformation/piezoelectric contribution. In the Zincblende case these contributions vanish at the case with isotropic wave functions, as discussed here, when taking the angular average by integrating over the in plane angle \( \int_0^{2\pi} d\phi F_z = 0 \) (valid for in plane symmetric wave functions). It also vanishes also in case of Wurtzit structure.
Fröhlich coupling to LO phonons

The last coupling mechanism we cover is polar coupling to LO phonons.

$$g_{LO}(\vec{q}) = -i \sqrt{\frac{\hbar \omega_{LO}(\vec{q})}{2V}} \left( e^2 \frac{1}{\epsilon_0} - \frac{1}{\epsilon_{stat}} \right) \frac{1}{q} \left( e^{-\frac{a^2 \vec{q}^2}{4}} e^{i\vec{q} \cdot \vec{r}_0} - e^{-\frac{a^2 \vec{q}^2}{4}} \right),$$

(2.34)

$$|g_{LO}(\vec{q})|^2 = \frac{\hbar \omega_{LO}(\vec{q})}{2V} \frac{e^2}{\epsilon_0} \left( \frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon_{stat}} \right) \frac{1}{q^2} \left( e^{-\frac{a^2 \vec{q}^2}{2}} + e^{-\frac{a^2 \vec{q}^2}{2}} - 2e^{-\frac{(a^2 + a^2) \vec{q}^2}{4}} \cos(q|\vec{r}_0| \cos \theta) \right).$$

(2.35)

Compared to the cases before, the investigation is rather simple, since there are neither different coupling strength to valence and conduction band, nor any angular dependent prefactors. Hence, it is easy to carry out the angular average. Since the spectral density is a $\vec{q}$ integration (cf. Eq. (2.24)) it depends on the coupling strength times $q^2$ (spherical coordinates). For the acoustic phonon cases this $q^2$ is compensated by $\omega^{-1}(q)$, in the LO case it is convenient to define $q^2$ as part of the coupling strength:

$$q^2 |g_{LO}(q)|^2 = \frac{\hbar \omega_{LO}(q)}{2V} \frac{e^2}{\epsilon_0} \left( \frac{1}{\epsilon_{\infty}} - \frac{1}{\epsilon_{stat}} \right) \left( e^{-\frac{a^2 \vec{q}^2}{2}} + e^{-\frac{a^2 \vec{q}^2}{2}} - 2e^{-\frac{(a^2 + a^2) \vec{q}^2}{4}} \sin(q|\vec{r}_0|) \right).$$

(2.36)

As before the expression can be split into the part for $r_0 = 0$ and the correction for taking into account the dipole moment of the wave functions. The following properties hold and are visualized in Fig. 2.4:

- The coupling vanishes for $q = 0$ as well as for equally sized wave functions
- Also as before, the $r_0$ independent part has one maximum at $q_{\text{max}}^2 = 2 \sqrt{\frac{\ln(a^2/a^2)}{a^2-a^2}}$. The strength of the maximum is independent of the QD size. The width of the coupling scales roughly as $\frac{1}{a^2}$. Since for the calculation of the spectral density the area of the coupling strength matters, small QDs will show stronger LO phonon signatures.
- The correction term for non-vanishing $r_0$ enhances the coupling and introduces a coupling, even if the sizes of the wave functions are equal

Influence of the phonon dispersion

After inspecting the wave number dependent interaction strength we turn next to the dispersion relation in order to calculate the spectral density as a function of the phonon frequency. Typically the acoustic phonon dispersion relation is described within Debye approximation $\omega_\kappa(q) = c_\kappa q$, whereas optical phonons are described in Einstein approximation $\omega_{LO}(q) = \omega_{LO}$. The problem is, that those approximations only hold for small wave numbers (cf. Fig. 1.3 and especially for small QDs relevant wave numbers can be easily higher than those approximations are reasonable). We
improve those approximations by describing acoustic phonons with a sinusoidal dispersion relation and optical phonons with a cosine dispersion. An advantage of those approximations is that both are invertible. We restrict ourselves to isotropic dispersion relations.\footnote{For LO phonons in nitride systems this is no good approximation.}

\begin{equation}
J_{\kappa}(\omega) = \sum_{q} \frac{|g_{q\kappa}|^{2}}{\hbar^{2}c_{\kappa}} \delta(\omega - \omega_{\kappa}(q)) = \frac{V}{2\pi^{2}} \int_{-\infty}^{\infty} dq \frac{1}{\partial_{q}\omega_{\kappa}(q)} \frac{q^{2}|g_{q\kappa}|^{2}}{\hbar^{2}\omega^{2}} \delta(\omega_{\kappa}^{-1}(\omega) - q). \tag{2.37}
\end{equation}

For acoustic Phonons in Debye approximation $\omega_{\kappa}^{-1}(\omega) = \frac{\omega}{c_{\kappa}}$ the spectral densities read (here we only show the deformation potential coupling; piezoelectric coupling behaves analogously):

\begin{equation}
J_{\kappa}(\omega) = \frac{1}{g} \frac{1}{4\pi^{2}} \frac{1}{\hbar c_{\kappa}} \frac{\omega}{\hbar c_{\kappa}} \left( D_{v}^{2} e^{-\frac{a_{v}^{2}\omega^{2}}{2c_{\kappa}^{2}}} + D_{c}^{2} e^{-\frac{a_{c}^{2}\omega^{2}}{2c_{\kappa}^{2}}} - 2D_{v}D_{c} e^{-\frac{(a_{v}^{2}+a_{c}^{2})\omega^{2}}{2c_{\kappa}^{2}}} \sin \frac{\omega}{c_{\kappa}} \frac{r_{0}}{\omega_{\kappa}^{-1}(\omega)} \right). \nonumber
\end{equation}

The spectral density in Debye approximation shows qualitatively the same behavior as the coupling strength $|g(q)|^{2}$ had in the $q$ domain. The most simple improvement is to take into account a sinusoidal dispersion $\omega_{\kappa}(q) = \omega_{\kappa}^{\text{max}} \sin(a_{\text{latt}}q)$, with $a_{\text{latt}} = \pi/2/\omega_{\kappa}^{\text{max}}$ and $\omega_{\kappa}^{\text{max}}$ is the dispersion at the Brillouin zone edge $\omega_{\kappa}^{\text{max}}$, e.g. at the K,X,M point or a respective mean value. To be more consistent with the Debye approximation for small $q$, $\omega_{\kappa}^{\text{max}} = 2c_{\kappa}q_{\text{max}}/\pi$ is a good choice, rather than the actual value. In the following expression you need to insert the wave number as a function of the frequency $q(\omega) = \arcsin \left( \frac{\omega}{\omega_{\kappa}^{\text{max}}} \right) a_{\text{latt}}$

\begin{equation}
J_{\kappa}(\omega) = \frac{1}{2\pi^{2}} \frac{1}{\hbar} \frac{1}{a_{\text{latt}}} \frac{q(\omega)^{4}}{\sqrt{\omega_{\kappa}^{\text{max}}^{2} - \omega^{2}}} \omega \left( D_{v}^{2} e^{-\frac{a_{v}^{2}\omega^{2}}{2\omega_{\kappa}^{\text{max}}^{2}}} + D_{c}^{2} e^{-\frac{a_{c}^{2}\omega^{2}}{2\omega_{\kappa}^{\text{max}}^{2}}} - 2D_{v}D_{c} e^{-\frac{(a_{v}^{2}+a_{c}^{2})\omega^{2}}{4\omega_{\kappa}^{\text{max}}^{2}}} \sin \frac{qr_{0}}{\omega_{\kappa}^{\text{max}}} \right). \nonumber
\end{equation}
In comparison to the Debye approximation this changes the spectral density especially for larger energies, cf. Fig. 2.5. For very small QDs there can even occur a discontinuity at $\omega = \omega_{\text{max}}$ due to the divergent phonon density of states at the band edge.

![Figure 2.5: Comparison of spectral densities using different approximations for the phonon dispersion. The upper figures show InAs/GaAs QDs, while the lower ones depict a GaN/AlN system. The left pictures show the longitudinal acoustic phonons' spectral density. For larger QDs Debye and sinusoidal description of the dispersion coincide, while for smaller QDs they behave differently, especially at the Brillouin zone edge. The right panels compare the optical phonon spectral density. In Einstein approximation there is only a delta line at the $\Gamma$ point LO energy, while with the cosine description the peak gets an asymmetric shape. For smaller QDs the maximum shifts to lower energies.](image)

Optical phonons in Einstein approximation consist of the $q$ integration over the interaction strength times a delta function at the LO energy:

$$J_{\kappa}(\omega) = \frac{V}{2\pi^2} \left( \int_{-\infty}^{\infty} dq q^2 |g_{\kappa q}|^2 \right) \frac{1}{\hbar^2 \omega_{LO}^2} \delta(\omega - \omega_{LO}).$$

That is, QD geometry and wave functions enter in this limit only via the total coupling strength compared to the zero phonon line, i.e. the Huang-Rhys factor Eq. (2.22d). Introducing the $q$ dependence of the dispersion is here even more important. We do this by approximating it with a cosine $\omega(q) = \frac{\omega_{LO} + \omega_q}{2} + \sqrt{\frac{\omega_{LO} - \omega_q}{2}} \cdot \cos \pi \frac{q}{q_{\text{lat}}} = \omega_0 + \Omega_0 \cos(2a_{\text{lat}}q)$ with the inverse $q(\omega) = \ldots$
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\[ \frac{1}{2a_{\text{latt}}} \arccos \left( \frac{\omega - \omega_0}{\Omega} \right). \]

Thus

\[ J_\kappa(\omega) = \frac{1}{4\pi^2 \epsilon_0} \left( \frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_{\text{stat}}} \right) \frac{1}{2a_{\text{latt}}} \sqrt{\Omega_0^2 - (\omega - \omega_0)^2} \frac{1}{\hbar \omega} \]

\[ \left( e^{-\frac{a^2 y^2}{2}} + e^{-\frac{a^2 q^2}{2}} - 2e^{-\frac{(a^2 + a^2)q^2}{4}} \sin(qr_0) \right). \]

Now also frequencies below the LO gamma point frequency contribute to the spectrum and the maximum is shifted towards lower energies. As the maximum of the coupling strength shifts to larger \( q \) for smaller QDs, the frequency shift is also larger for smaller QDs, cf. Fig. 2.5.

2.1.3 Numerical investigation of optical spectra

Spectra can be numerically evaluated for any wave function. Thus using realistic \( 8 \vec{k} \cdot \vec{p} \) wave functions comparison to experimental spectra. Real quantum dots do not match the wave function approximations made in the previous section. Hence, for comparing theoretical models to measured spectra it is convenient to use more realistic wave functions. However, the insights from the previous section will help to understand the following observations:

InAs/GaAs QDs

In Reference [SDW+11] we compared the experimental electro-luminescence spectrum of a single QD emitter [LST+09] with simulations within the IBM framework. Figure 2.6 shows the respective data. The simulations were done using the presented Gaussian wave functions as well as wave functions microscopically calculated via \( 8 \vec{k} \cdot \vec{p} \) theory [SWB07]. For the spectrum of the acoustic phonons we could see the above discussed properties of a spectrum, e.g. the asymmetry of the spectrum for low temperatures due to spontaneous phonon emission. An interesting finding is that for larger \( q \) the coupling via Gaussian wave functions underestimates the coupling strength compared to the experiment and \( 8 \vec{k} \cdot \vec{p} \) wave functions. That means the ansatz with Gaussian wave functions underestimates the spatial confinement. Further, we could observe that the spectral density calculated with \( 8 \vec{k} \cdot \vec{p} \) wave functions did not vanish at the edge of the Brillouin zone, thus the Debye approximation is not good for InAs/GaAs QDs. However, since the acoustic phonon interaction is rather weak in this material system, the experiment could not make a statement about behavior at the Brillouin zone edge (at about 12 meV) of the spectrum. Piezoelectric coupling to acoustic phonons plays no rule here. For the LO part of the spectrum the experimental data shows a broad and asymmetric line incompatible to the usual Einstein approximation. As discussed above, using a cosine dispersion relation leads to such a line shape. Further, taking into account a finite phonon lifetime further broadens the spectrum to match the experimental data.
CHAPTER 2. PHONON INFLUENCE ON CARRIER DYNAMICS IN QDS

Figure 2.6: Spectra of InAs/GaAs single QD emitter as in Ref. [SDW+11]. Comparison of experimental data with simulations. In the left panel acoustic phonon sidebands give a prediction of the wave functions extent. Gaussian wave functions cannot describe the behavior as good as microscopically calculated wave functions, since a Gaussian description underestimates the spatial confinement. The right panel shows the LO phonon peak is asymmetric and broadened, which can be described by a cosine dispersion relation and an additional 5 ps phonon lifetime.

GaN/AlN QDs

Nitride based QDs are a promising basis for the development of light emitters for a broad spectral region from visible to UV. Carriers in nitride hetero-structures are strongly confined making them candidates for room temperature devices for quantum information applications. In those systems piezoelectric and pyroelectric (temperature dependence of the dipoles) effects have a significant influence, because the lattice lacks inversion symmetry. This leads to a strong piezoelectric electron-phonon interaction on one hand and a large build-in dipole moment in the QDs on the other. Additionally, nitride QDs and their wave functions are rather small compared to the arsenide system. All in all, knowing the rules of thumb from above, this leads to a strong electron-phonon interaction in nitride QDs. In Ref. [OHR+12] we investigate the cathodoluminescence spectrum of GaN/AlN QDs. The data is shown in Fig. 2.7. Especially compared to the arsenide system the acoustic phonon sidebands are huge as is the broadening of the zero phonon line. In the nitride system the piezoelectric coupling to acoustic phonons is in the same order of magnitude as the deformation potential coupling. Such a strong signature is the ideal basis for comparison to simulations with different wave functions. Simulations with $8\vec{k} \cdot \vec{p}$ wave functions and Gaussian wave functions underline the influence of the intrinsic dipole in nitride quantum dots. The comparison of the experimental spectra for different QDs yielded a broader spectrum, i.e. stronger acoustic phonon interaction for QDs with lower emission energy, i.e. larger QDs [Ost12]. As we saw in Sec. 2.1.2 this can be attributed to the enhanced piezoelectric coupling for QDs with a larger intrinsic dipole moment. For possible applications, where it is mostly favorable to have small coupling to phonons, this means it is important to find the QD right size since too small QDs have strong deformation
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**Figure 2.7:** Spectra of nitride QDs from cathodoluminescence. In the left panel experimental cathodoluminescence spectra of a single QD for different temperatures are shown. The spectra are normalized with respect to the area beneath them, since the area corresponds to the total emitted power. The right figure is Fig. 2 from Ref. [OHR+12] and shows calculated spectra for various temperatures as well as the respective spectral density decomposed into the different coupling mechanisms. The lower right panel compares the calculated spectrum to the measured data.

Potential coupling as too large QDs have a strong piezoelectric coupling.

A thorough investigation of the optical phonons remains to be done in the material system.
2.2 Coupling to carrier reservoir – multi-phonon relaxation

Phonons play a significant role in intra-band and inter-subband transitions in semiconductors due to the possibility of exchanging momentum and energy. The relaxation process can be attacked via Heisenberg equation of motion approach and in Born-and Markovian approximation leading to first order relaxation processes as described in Sec. 1.3.

2.2.1 The phonon bottleneck

In a QD–WL system the description of capturing and relaxation via LO phonons suffers a problem: QD energies as well as phonon energies are discrete, which gives rise to an expected inefficient scattering mechanism, due to the required energy conservation from Markovian approximation (as in Fermi’s golden rule, cf. Sec. 1.3). This creates the necessity of taking into account higher order processes, to allow for the violation of energy conservation on short timescales. There is a long history of experiments trying to confirm this so called “phonon bottleneck” [HBG+01; UNS+01], because other scattering channels as Coulomb scattering are efficient in relaxations [Wil13; Maj12]. On the other side theories try to include mechanisms, that circumvent the strict energy conservation, e.g. by non-Markovian approaches as higher order density matrix formalism [FWD+03] or Green’s functions approaches using the polaron description [LNS+06; SNG+05]. Those approaches solve the full dynamics of all quantities and need a high numerical effort. A perturbative multi-phonon treatment is another approach to the problem [MUB+02; IS94; IS92]. Such an approach can be considered as higher order Markovian, i.e. higher numbers of one-phonon interactions that violate energy conservation are included implicitly, leading to an energy conserving process. The advantage of such an approach is that it leads to a rate equation description, which is often desired for further use as the calculation of QD based devices [KDW+11] or for more complex systems with more interaction as the quantum optical treatment of the biexciton cascade where the interaction with phonons influences the degree of entanglement of the photons [CMD+10].

In this section we present a perturbative multi-phonon approach based on a projection operator technique. Within this approach it is easily possible to take into account many electron contributions, which is vital for solid state system. In the end we present the resulting rate equation and discuss the corresponding multi-phonon rates.

2.2.2 Effective multi-phonon Hamiltonian

We present an approach where an effective multi-phonon Hamiltonian is derived. Effective Hamiltonians are often used to reduce the complexity of a problem. The approach we use is based on a multi photon approach from [Fai87, p. 158ff], and delivers the possibility to treat relaxations on a rate equation limit.

The system is divided into a resonant subsystem where all (many particle) states have the same energy and a non-resonant subsystem consisting of all other states. Within a perturbation theory the influence of the non-resonant subsystem is taken into account implicitly for transitions within
the resonant subsystem. The Hamiltonian of the full system \( \hat{H}' \) is split into a free part \( \hat{H}'_0 \) and the interaction part \( \hat{V} \), which has the role of a perturbation:

\[
\hat{H}' |\Psi\rangle = E |\Psi\rangle, \tag{2.39}
\]

\[
\hat{H}' = \hat{H}'_0 + \hat{V}. \tag{2.40}
\]

Additionally we need a constant energy \( W \) which has to be assigned later on and decides on the kind of perturbation theory.\(^8\)

\[
(\hat{H}' - W \cdot \text{Id}) |\psi\rangle = \hat{H} |\psi\rangle = E |\psi\rangle; \quad |\psi\rangle = e^{iWt} |\Psi\rangle, \quad \hat{H} = \hat{H}_0 + \hat{V}; \quad \hat{H}_0 = \hat{H}'_0 - W \cdot \text{Id}. \tag{2.41}
\]

The eigenstates of \( \hat{H}_0 \), \( |i\rangle \) are divided into resonant and non-resonant ones to define the two projection operators

\[
P = \sum_i |i\rangle \langle i| \tag{2.43}
\]

\[
Q = \text{Id} - P = \sum_i |i\rangle \langle i|. \tag{2.44}
\]

where \( \sum_i \) sums up resonant states, \( \sum_{\bar{i}} \) the rest. For the application to the solid state system \( |i\rangle \) will be a product state of electronic and phonon states.

The projection operators are applied to Eq. (2.41) using the projector properties \( 1 = P + Q \) and \( P^2 = P \).

\[
EP |\psi\rangle =: E |\psi\rangle_P = P \hat{H} |\psi\rangle + P \hat{H} Q |\psi\rangle =: \hat{H}_{PP} |\psi\rangle_P + \hat{H}_{PQ} |\psi\rangle_Q, \tag{2.45}
\]

\[
EQ |\psi\rangle = \hat{H}_{QP} |\psi\rangle_P + \hat{H}_{QQ} |\psi\rangle_Q \Leftrightarrow |\psi\rangle_Q = (E \cdot \text{Id} - \hat{H}_{QQ})^{-1} \hat{H}_{QP} |\psi\rangle_P. \tag{2.46}
\]

The non resonant states \( |\psi\rangle_Q \) are eliminated by inserting Eq. (2.46) into Eq. (2.45). Since the projectors commute with \( \hat{H}_0 \) and \( PQ = 0 \) the interaction between the subsystems is solely the perturbation part \( \hat{H}_{QP} = \hat{V}_{QP}. \) The inverse operator is decomposed via a Neumann series:

\[
(Id - \hat{O})^{-1} = \sum_{n=0}^{\infty} \hat{O}^n, \tag{2.47}
\]

for a general operator \( \hat{O} \). The identity holds if \( \sum_{n=0}^{\infty} \hat{O}^n \) converges, especially for \( ||\hat{O}|| < 1 \) [Wer05; RS80]. Thus, Eq. (2.45) reads:

\[
E |\psi\rangle_P = \hat{H}_{PP} |\psi\rangle_P - \hat{V}_{QP}(\hat{H}_{QQ} - E)^{-1} \hat{V}_{QP} |\psi\rangle_P
\]

\[
= \left( \hat{H}_{PP} - \hat{V}_{QP} \hat{H}_{QQ}^{-1} (1 - E \hat{H}_{QQ}^{-1})^{-1} \hat{V}_{QP} \right) |\psi\rangle_P
\]

\[
= \left( \hat{H}_{PP} - \hat{V}_{QP} \hat{H}_{QQ}^{-1} \sum_{n=0}^{\infty} \left( \hat{H}_{QQ}^{-1} \hat{V}_{QP} E^n \right) \right) |\psi\rangle_P. \tag{2.48}
\]

\(^8\)Such a constant can also be introduced in the perturbative treatment of the Schrödinger equations, its choice decides for example between Schrödinger and Brillouin–Wigner perturbation series.[Nol06]
\[ H_{QQ}^{-1} = (Q\hat{H}Q + Q\hat{V}Q)^{-1} = \sum_{n=0}^{\infty} (-1)^n \hat{H}_0^{-1}(\hat{V}_{QQ}\hat{H}_0^{-1})^n \quad \text{with} \quad \hat{H}_0 = Q\hat{H}Q. \quad (2.49) \]

Inserting into Eq. (2.48) leads to:

\[ E|\psi\rangle_P = \left( \hat{H}_{PP} - \hat{V}_{PP} \sum_{n'=0}^{\infty} \left( \sum_{n=0}^{\infty} (-1)^n \hat{H}_0^{-1}(\hat{V}_{QQ}\hat{H}_0^{-1})^n \right)^{(n'+1)} \right) \hat{V}_{QP}E^{n'} |\psi\rangle_P. \quad (2.50) \]

\[ \hat{A} \] is nearly an effective Hamiltonian except for the self consistent dependence on its eigenvalue \( E \). To make it a Hamilton operator \( E^n \) is iteratively replaced by \( \hat{A}^n \) (valid due to \( E^n |\psi_P\rangle = A^n |\psi_P\rangle \)). Subsequently, the terms can be ordered by their order of \( \hat{V} \). This is, where we assign \( W \) in a way, that \( \hat{H}_{PP} = \hat{V}_{PP} \). Else all higher terms would contain orders of \( \hat{H}_0 \).

\[ \hat{H}_{PP} = \hat{V}_{PP} \Leftrightarrow P\hat{H}_0P = 0 \Leftrightarrow W = \left\langle \psi | P\hat{H}_0'P |\psi \right\rangle. \quad (2.51) \]

By construction all transitions within the resonant subsystem are energy conserving.

Eventually, the effective Hamiltonian is explicitly given by

\[ \hat{H}_\text{eff} = \hat{V}_{PP} - \hat{V}_{PP} \hat{H}_0^{-1} \hat{V}_{QP} \]
\[ + \hat{V}_{PQ} \hat{H}_0^{-1} \hat{V}_{QQ} \hat{H}_0^{-1} \hat{V}_{QP} - \hat{V}_{PQ} \hat{H}_0^{-2} \hat{V}_{QP} \hat{V}_{PP} \]
\[ - \hat{V}_{PQ} \hat{H}_0^{-1} \hat{V}_{QQ} \hat{H}_0^{-1} \hat{V}_{QP} + \hat{V}_{PQ} \hat{H}_0^{-1} \hat{V}_{QQ} \hat{H}_0^{-2} \hat{V}_{QP} \hat{V}_{PP} + \hat{V}_{PQ} \hat{H}_0^{-2} \hat{V}_{QQ} \hat{H}_0^{-1} \hat{V}_{QP} \hat{V}_{PP} \]
\[ + \hat{V}_{PQ} \hat{H}_0^{-2} \hat{V}_{QP} \hat{V}_{PP} \hat{H}_0^{-1} \hat{V}_{QP} - \hat{V}_{PQ} \left( \hat{H}_0^{-1} \right)^3 \hat{V}_{QP} \hat{V}_{PP}^2 \]
\[ \cdots. \quad (2.52) \]

The free energy part of the effective Hamiltonian is contained in \( W/\hbar \):

\[ \hat{H}_0' |\psi\rangle_P = W |\psi\rangle_P \Rightarrow \hat{H}_\text{eff} = \hat{H}_0' + \hat{H}_\text{eff}. \quad (2.53) \]

The first order term just represents the interaction from the initial system restricted to the resonant subsystem. Higher order terms represent interactions within the resonant system, assisted by the non-resonant states. This can be seen with the aid of the second order:

\[ -\hat{V}_{PQ} (Q\hat{H}_0Q - W)^{-1} \hat{V}_{QP} = \sum_{i,j} P\hat{V} |i\rangle \langle j| (\hat{H}_0' - W)^{-1} |j\rangle \langle i| \hat{V} P \]
\[ = \sum_{i} \left( |i\rangle \langle \hat{H}_0' |i\rangle - W)^{-1} P\hat{V} |i\rangle \langle i| \hat{V} P. \quad (2.54) \]
This describes an interaction within the resonant subsystem, assisted by the non resonant states \( |i\rangle \) via an implicit transition with an weighting term \( \left( \langle i | \hat{H}_0' | i \rangle - W \right)^{-1} \) that is inverse proportional to the energetic difference of the resonant states and the energy of \( |i\rangle \). This is an energy conserving interaction composed of two transitions, that do not conserve energy. This can be seen as a higher order Markovian process. Another point of view is to call \( |i\rangle \) a virtual state. Figure 2.8 shows some processes from the effective two-phonon Hamiltonian we will explicitly derive in a minute. The interpretation of higher order terms follows analogously.

**Figure 2.8:** Two phonon relaxation between two states with energetic difference of \( 2\hbar \omega_{LO} \). a) processes in a resonant two level system: The relaxation takes place via non-resonant states with a non-matching number of phonons. Such states are typically called virtual. The two phonon transition is composed of one diagonal interaction and a non-diagonal. b) a system with an additional continuum of non-resonant states. Here, the relaxation includes intermediate transitions into those states and is such composed of two non-diagonal interactions.
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Application to the electron–phonon system

The next step is to apply this general technique to the specific QD/WL electron and phonon problem. The electron phonon Hamiltonian reads

\[ H'_0 = \sum_i \epsilon_ia_i^\dagger a_i + \hbar \omega \sum_q b_q^\dagger b_q, \]

\[ V = \sum_{ij} \sum_q g_{ijq}a_i^\dagger a_j (b_q + b_{-q}^\dagger). \]

Now it is important to define the resonant states. The system states are composed of the electronic states \(|\epsilon\rangle\) and the phonon states \(|n\rangle\) each being many particle states.

\[ |\psi\rangle = \sum_{\epsilon, n} |\epsilon, n\rangle. \]

We define one state \(|\epsilon_0, n_0\rangle\) to be the reference for the energy conservation. This is the state where QD level is occupied and there is a phonon number state \(|n_0\rangle\). Everything has to be done for a general initial condition, where when the electron is occupied there are \(N_0\) Phonons in the system and all electrons in the WL have an overall energy of \(\epsilon_{WL}\). The phonon number is lowered by \(n\) for a transition into other electronic states \(|\epsilon_r\rangle\) under energy conservation within an \(n\) phonon process.

\[ W = \epsilon_0 + \epsilon_{WL}^0 + \hbar \omega N_0 = \epsilon_r + \epsilon_{WL}^0 + \hbar \omega N_r, \]

\[ N_0 = \sum_q \langle n_0 | b_q^\dagger b_q | n_0 \rangle, \quad N_r = \sum_q \langle n_r | b_q^\dagger b_q | n_r \rangle. \]

The effective Hamiltonian shall only operate on the QD states and the electronic states being integer times the phonon energy away with energies \(\epsilon_r = \epsilon_0 + n\hbar \omega_{LO}\). Thus the projector \(P\) contains all states fulfilling this energy condition. We want to deal with a system, where the one phonon transition is energetically forbidden. Thus there are no resonant states for which a transition \(PVP\) is allowed, which means \(VPP\) vanishes. We will directly turn to the two phonon case.

The effective two phonon Hamiltonian, already written in Eq. (2.54) yields for the actual interaction Hamiltonian Eq. (2.55)

\[ P\hat{H}^{(2)}P = \sum_{ij} \sum_{q\pm} \sum_{k\pm'} g_{ijqk}g_{k\pm'} (\langle \psi | \hat{H}_0 | \psi \rangle - W)^{-1} Pa_i^\dagger a_j b_q^\dagger b_{q'}^\dagger P \]

with \(b_q^+ = b_{-q}^\dagger\) and \(b_q^- = b_q\). The energy of all states in \(P\) is \(W\). The operators left and right of the projector lead to the conditions

\[ \langle \psi | \hat{H}_0 | \psi \rangle = W - \epsilon_i + \epsilon_k \pm' \hbar \omega_{LO} = W - \epsilon_i + \epsilon_j \mp \hbar \omega_{LO}. \]

\[ ^9 \text{Note, that we use the electron-hole picture, where conduction and valence band are symmetric. Later on we will restrict ourselves to one band, when calculating the rates, the derivation here is, however, done in general.} \]
This introduces an important energy relation. Now, the inverse term is not depending on the actual initial condition \( P \) anymore. The sum over the non-resonant states can be removed via

\[
P\tilde{H}^{(2)} = \sum_{ij} \sum_{kl} \sum_{q\pm} \sum_{q'\pm'} g_{ij}^{qq'} g_{kl}^{qq'} (-\epsilon_i + \epsilon_k) (1 - \delta_{k\ell}) \sum_{\alpha} a_i^\dagger a_j (1 - \delta_{k\ell}) \sum_{\alpha} a_k^\dagger a_l \frac{1}{\hbar \omega_{\text{LO}}} (b_{q}^\pm b_{q'}^\mp - b_{q'}^\mp b_{q}^\pm) \]

The second line removes possible resonant one phonon processes. Assuming there are no electronic states resonant to a single phonon transition, it vanishes as stated above. Hence, in normal ordering the effective Hamiltonian reads

\[
\tilde{H}^{(2)} = \sum_{ij} \sum_{kl} \sum_{q\pm} \sum_{q'\pm'} g_{ij}^{qq'} g_{kl}^{qq'} (-\epsilon_l + \epsilon_k) (1 - \delta_{k\ell}) \sum_{\alpha} a_i^\dagger a_j (1 - \delta_{k\ell}) \sum_{\alpha} a_k^\dagger a_l \frac{1}{\hbar \omega_{\text{LO}}} (b_{q}^\pm b_{q'}^\mp - b_{q'}^\mp b_{q}^\pm).
\]

This effective Hamiltonian contains one and two electron transitions with interaction with two phonons. The one particle term represents the transition from level \( l \) to level \( k \) composed of the transitions \( l \to k \) and \( k \to i \) without taking into account possible occupations of the level \( k \). The effectiveness of the transitions is weighted by how close the intermediate level is to a single phonon resonance. In terms of Fig. 2.8 it contains virtual and intermediate transitions. For the intermediate transitions the occupation of the intermediate levels has in general to be taken into account. For a one electron assumption, i.e. a completely empty WL, the term vanishes. Since we are interested in an effective one particle phonon interaction the term is treated via the Hartree-Fock factorization [Czy00]:

\[
a_i^\dagger a_k^\dagger a_j a_l = \langle a_i^\dagger a_j \rangle a_i^\dagger a_l + \langle a_i^\dagger a_l \rangle a_i^\dagger a_j - \langle a_i^\dagger a_j \rangle a_i^\dagger a_l - \langle a_l^\dagger a_j \rangle a_i^\dagger a_l.
\]

Note, that this term contains the fermionic property \( a_i^\dagger a_k^\dagger a_j a_l \sim (1 - \delta_{ik})(1 - \delta_{jl}) = \delta_{ik}\delta_{jl} \). We will explicitly use these inverse Kronecker deltas for bookkeeping reasons. The first two terms describe one particle operators in the mean field of other one-particle operators, the last two terms, called exchange interaction, make the difference to classical mean field methods. Putting this into Eq. (2.60) one can re-index the sums to get the two contributions:

\[
\tilde{H}^{(2)}_{\text{two particle}} = -\sum_{ij} \sum_{kl} \sum_{q\pm} \sum_{q'\pm'} g_{ij}^{qq'} g_{kl}^{qq'} \langle a_i^\dagger a_j \rangle a_k^\dagger a_l \frac{1}{\hbar \omega_{\text{LO}}} (b_{q}^\pm b_{q'}^\mp - b_{q'}^\mp b_{q}^\pm) + \sum_{ij} \sum_{kl} \sum_{q\pm} \sum_{q'\pm'} g_{ij}^{qq'} g_{kl}^{qq'} \langle a_i^\dagger a_j \rangle a_l^\dagger a_i \frac{1}{\hbar \omega_{\text{LO}}} (b_{q}^\pm b_{q'}^\mp - b_{q'}^\mp b_{q}^\pm). 
\]

\[10\]Note that the denominator is equal to \((-\epsilon_i + \epsilon_j \pm \hbar \omega_{\text{LO}}) \), according to Eq. (2.59).

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The first term has the same weighting as the one particle process in Eq. (2.60), but now includes the occupation of the intermediate levels. Note, that in terms of Fig. 2.8 those processes are only intermediate processes (b). The exchange term however is weighted by the energetic difference of the resonant or the intermediate levels, respectively. This Hamiltonian is restricted to only operate on the interesting resonant electronic states via tracing out non-resonant states. This only leaves operators acting on resonant energies as operators.

The next step is to explicitly treat the $\pm, \pm'$ sums, i.e. the single phonon absorption and emission processes, which leads to two general cases:

- First case is both phonon operators are creators/annihilators $\pm = \pm'$. Those parts of the Hamiltonian are the transition terms, where two phonons are absorbed/emitted. Here, the overall phonon number changes by two via the transitions (cf. Fig. 2.8). From Eq. (2.59) we get the energy relation for the states:

\[
(\epsilon_l - \epsilon_k) - (\epsilon_i - \epsilon_j) = \pm 2\hbar\omega_{\text{LO}}
\]  

(2.63)

Since $l, i$ are resonant electronic states the possible energetic differences are $(\epsilon_l - \epsilon_i) \in \{-2\hbar\omega_{\text{LO}}, 0, 2\hbar\omega_{\text{LO}}\}$. Thus, the assisting states need to obey $(\epsilon_k - \epsilon_j) \in \{0, \pm 2\hbar\omega_{\text{LO}}, 0\}$, respectively. In the two phonon emission/absorption case the many particle correction Eq. (2.62) vanishes because $[b_{i\pm}^\dagger, b_{k\pm}^\dagger] = 0$. Hence, the two phonon transition part of the effective Hamiltonian is the one electron contribution and reads:

\[\hat{H}^{(2)}|_{\pm=\pm'} = \sum_{ilk} \sum_{qq'} g_{ik}^a g_{kl}^d \frac{\hbar\omega_{\text{LO}}}{\pm} a_i^\dagger b_{i\pm}^\dagger b_{k\pm}.\]  

(2.64)

The energy conservation Eq. (2.59) results in the constraint $i \neq l$, i.e. only non-diagonal interaction. The vanishing many particle contribution Eq. (2.62) can be understood as follows: There are two parts canceling each other; one part acts as a Pauli blocking correction to the one electron part, i.e. $1 - \langle a_k^\dagger a_k \rangle$, whereas the other part is proportional to the occupation of the intermediate level, i.e. an electron from the intermediate level scatters into the QD and then the intermediate level is filled again. Comparing this to Fig. 2.8b both processes represent different time ordering of the left and the right side.

- In the second case $\pm = \mp'$ there is one phonon creation as well as one annihilation, which means the overall phonon number is not changed in the process. The energy relation Eq. (2.59) then reads:

\[
(\epsilon_l - \epsilon_i) - (\epsilon_k - \epsilon_j) = 0.
\]  

(2.65)

For the case of a diagonal interaction $i = l$ (i.e. the electronic state does not change) it simply means the carrier is virtually/intermediately occupying a non-resonant level and then jumps back to its original state, or for the exchange part that diagonal interactions of two electrons compensate each other. A transition $\epsilon_l - \epsilon_i = \pm \hbar\omega_{\text{LO}}$ would be compensated by an opposite two phonon transition of other states. We neglect this, since it would couple to fast
oscillating terms in the reservoir. Further, the intermediate resonance $\langle a_k^\dagger a_j \rangle$ for $\epsilon_k = \epsilon_j$ can be assumed to be diagonal $\langle a_k^\dagger a_k \rangle \delta_{kj}$ for homogeneous/isotropic systems \cite{Ric07}.

$$\hat{H}^{(2)}_{|\pm=\mp} = \sum_{il} \sum_{k} \sum_{qq'} g^{il}_{ik} g^{kl}_{qk} \left( b_{il}^+ b_{qk}^+ - \langle a_k^\dagger a_k \rangle \delta_{kl} \delta_{qq'} (\mp \delta_{qq'}) \right) a_i^\dagger a_l. \quad (2.66)$$

Eventually, we obtain an effective two phonon Hamiltonian with effective coupling elements. We will use only processes in one band since inter-band have weak contribution due to their large energy separation on one hand and band diagonal phonon coupling elements on the other. \footnote{Note that due to the exchange terms however densities of the other band would contribute.}

We assume one QD state $\epsilon_0$ and a quasi-continuum $\epsilon_r$ in the carrier reservoir.

$$\hat{H}^{(2)}_{\text{el-ph}} = g_{00} a_0^\dagger a_0 + g_{rr} a_r^\dagger a_r$$

$$+ \sum_{q} g^{q}_{00} a_0^\dagger a_q b_{-q}^\dagger b_q + \sum_{q} g^{q}_{rr} a_r^\dagger b_{-q}^\dagger b_q$$

$$+ \sum_{q} g^{q}_{0r} a_0^\dagger a_r b_{-q}^\dagger b_q.$$  \hspace{1cm} (2.67)

With the six effective electron-two-phonon coupling elements, that include densities from the carrier reservoir, which we denote $f_k = \langle a_k^\dagger a_k \rangle$ and treat as equilibrium distribution. The first line does not include phonon operators, thus those terms renormalize the electronic energies temperature independent:

$$g_{ii} = \sum_q g^q_{ii} \frac{\overline{g}^q_{ii}}{\hbar \omega_{\text{LO}}} + \sum_{k \neq i} \sum_q \left( g^q_{ik} \overline{g}^q_{ki} \left( \frac{f_k}{-\epsilon_i + \epsilon_k - \hbar \omega_{\text{LO}}} + \frac{1 - f_k}{-\epsilon_i + \epsilon_k + \hbar \omega_{\text{LO}}} \right) \right), \quad (2.68a)$$

This are processes, where a phonon is spontaneously emitted and absorbed again and thus create a new quasi-particle, the polaron. The first term is the polaron shift from the pure diagonal interaction we already know from Sec. 2.1.1 (Eq. (2.22a)). The next represents an intermediate transition, where the relaxation from $k$ to $i$ and emission of the phonon is the first process, thus it is proportional to the density of the level $k$. This process is most effective if $\epsilon_k > \epsilon_i$, i.e. at coupling to a higher energy level. Then there is the process, where the electron is excited from $i$ to $k$ first under emission of a phonon, proportional to the blocking of level $k$. This process is effective, if $k$ has a lower energy than $i$.

The next effective coupling elements are diagonal in the electronic operators and do not change the phonon number. They also represent polarons. This would correspond to re normalization of both, electrons and phonons, especially they lead to temperature dependent energy shifts of the
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electronic levels.

\[ g_{ii}^{qq'} = \sum_{k \neq i} \frac{g_k^q g_{ki}^{q'}}{-\epsilon_i + \epsilon_k - \hbar \omega_{LO}} + \frac{g_{ik}^{q'} g_k^q}{-\epsilon_i + \epsilon_k + \hbar \omega_{LO}}. \]  \hspace{1cm} (2.68b)

Those two term arise from intermediate transitions, where the first favors intermediate levels above \( \epsilon_k > \epsilon_i \) and the second favors intermediate levels below.

Finally there are the relaxation/excitation matrix elements. The two phonon emission with the capturing of the carrier from the WL.

\[ g_{0r}^{qq'} = \sum_{k \neq r} \frac{g_0^q g_{kr}^{q'}}{-\epsilon_0 + \epsilon_k + \hbar \omega_{LO}}. \]  \hspace{1cm} (2.68c)

The first two contributions are direct transitions under the additional diagonal emission of a phonon.

The last process covered is the two phonon absorption with excitation of the QD electron into the WL.

\[ g_{r0}^{qq'} = \sum_{k \neq r} \frac{g_k^q g_{k0}^{q'}}{-\epsilon_0 + \epsilon_k - \hbar \omega_{LO}}. \]  \hspace{1cm} (2.68d)

This process is the complex conjugate of the two phonon emission. Next, we will use the effective Hamiltonian to derive rate equations for the QD occupations.

2.2.3 Multiphonon relaxation rates

Using the effective Hamiltonian Eq. (2.67), we derive Heisenberg equations of motion for the QD occupations \( \langle a_0^\dagger a_0 \rangle \). The derivation of the dynamics equations is made just as with the conventional Hamiltonian, cf. Sec. 1.3. The dynamic equation for the QD occupation in one band follows from the commutator with \( H_{el} + H_{ph} + H_{el-ph}^{(2)} \) and reads

\[ i\hbar \partial_t a_0^\dagger a_0 = \sum_{q'q''r} g_{0r}^{q'q''} a_0^\dagger a_r b_{-q'}^\dagger b_{-q''}^\dagger - \sum_{q'q''r} g_{r0}^{q'q''} a_r^\dagger a_0 b_{q'} b_{q''}. \]  \hspace{1cm} (2.69)
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The dynamics couple to transitions assisted by two phonon operators. The equations of motion for those higher order terms read:

\[ i\hbar \partial_t a_{0\alpha} b_{-q_1} b_{-q_2} = (\epsilon_r - \epsilon_0 - g_{00} - 2\hbar \omega_{LO}) a_{0\alpha} b_{-q_1} b_{-q_2} - \sum_{q',q''} g_{00q''} a_{0\alpha} b_{-q'} b_{-q''} b_{-q_1} b_{-q_2} + \sum_{q',q''} g_{0q'q''} a_{0\alpha} b_{-q_1} b_{-q_2} b_{-q'} b_{-q''} - \sum_{q',q''} \sum_{r'} g_{r'q''} a_{0\alpha} a_{r} a_{r'} \left( -b_{-q_1} b_{-q_2} \delta_{q',q''} - b_{-q'} b_{-q_1} \delta_{q''-q_2} \right) + \sum_{q',q''} \sum_{r'} g_{r'q''} \left( a_{0\alpha} - a_{r\alpha} \right) b_{-q_1} b_{-q_2} b_{-q'} b_{-q''} + \sum_{q',q''} \sum_{r'} (g_{0q'q''} + g_{r'q''}) (a_{0\alpha} a_{r} a_{q_1} - a_{r\alpha} a_{q_1}) \left( b_{-q_1} b_{-q_2} \delta_{q',q''} + b_{-q'} b_{-q_1} \delta_{q''-q_2} + \delta_{q_1,q'} \delta_{q_2,q''} \right). \]

The free part not only consist of the bare energy of the electrons and phonons but also of the polaron shifts of the levels. The next terms resemble also the two-phonon assisted transition, with additional assistance of the phonon number operator, thus they lead to a further, temperature dependent, polaronic energy shift. The next part is also a polaronic energy shift, assisted by the occupations of the electronic levels. Taking into account the polaronic terms however would need the calculation of higher orders in the hierarchy. The last two contributions are the desired couplings to electron and phonon densities.

At this point we stop the expansion and try to close the system of equations with the methods we introduced in Sec. 1.3. The energy renormalizations are included in effective level energies \( \tilde{\epsilon}_r, \tilde{\epsilon}_0 \) and are no longer in the focus of our investigation. The scattering will happen self consistently between states that obey the energy conservation. Those terms do not directly contribute to scattering rates.

Now for the relevant part we apply Born factorization to separate electron and phonon dynamics, the four operator electron values are factorized via Hartree-Fock \( \langle a_{0\alpha} a_{r} a_{q_1} a_{q_2} \rangle = \langle a_{0\alpha} a_{0} \rangle \langle a_{r} a_{r} \rangle \), but the last part is omitted, rendering it effectively only a Hartree (classical mean field) factorization. The four operator phonon terms are factorized assuming a thermalized bath \( \langle b_{-q_1} b_{-q_2} b_{-q'} b_{-q''} \rangle = \langle b_{-q_1} b_{q} \rangle \langle b_{-q_2} b_{-q'} \rangle + \langle b_{-q_2} b_{q} \rangle \langle b_{-q_1} b_{-q'} \rangle + \langle b_{-q_1} b_{q} \rangle \langle b_{-q_2} b_{-q'} \rangle = n_{q_1} \delta_{q_2,q'}, \) where \( n \) denotes the Bose distribution. Finally, integration and Markovian approximation yields

\[ \langle a_{0\alpha} b_{-q_1} b_{-q_2} b_{-q'} b_{-q''} \rangle = -i \frac{\pi}{\hbar} \delta(\tilde{\epsilon}_r - \tilde{\epsilon}_0 - 2\hbar \omega_{LO}) \left( (g_{r_0} n_{q_1} - g_{r_0} n_{q_2}) \left( \langle a_{0\alpha} a_{0} \rangle \langle a_{r} a_{r} \rangle - \langle a_{r\alpha} a_{r\alpha} \rangle \right) + (g_{r_0} n_{q_1} - g_{r_0} n_{q_2}) \left( \langle a_{0\alpha} a_{0} \rangle \langle a_{r} a_{r} \rangle - \langle a_{r\alpha} a_{r\alpha} \rangle \right) \right). \]

The rate equation for the QD occupation follows directly as

\[ \hbar \partial_t \rho_{00} = \sum_{q_1,q_2} \sum_r \frac{2\pi}{\hbar} \delta(\tilde{\epsilon}_r - \tilde{\epsilon}_0 - 2\hbar \omega_{LO}) g_{0q_{1r}} g_{r_{0q_1} - q_2} + g_{r_{0q_2} - q_{1r}} \left( (1 - \rho_{00}) \rho_{rr} (n_{q_1} + 1)(n_{q_2} + 1) - \rho_{00} (1 - \rho_{rr}) (n_{q_1} n_{q_2}) \right). \]
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Since the energy of the resonant states of the reservoir is set and the phonon distribution does not depend on the phonon wave number in Einstein approximation, a rate independent of the phonon and electron densities can be defined. This leads to a simple equation consisting of in- and out-scattering rates:

$$\partial_t \rho_{00} = \frac{\Gamma_{rr}(n + 1)^2(1 - \rho_{00}) - \Gamma(1 - \rho_{rr})n^2 \rho_{00}}{S_{in} - S_{out}}$$

(2.71)

The rates define the long time limit (steady state) of the QD occupation $\rho^\infty$, i.e. the equilibrium occupation. Since the rates depend on the carrier distribution in the reservoir, pumping of the reservoir directly affects this steady state. A convenient description is the relaxation time picture, that is also used in Chap. 3.

$$\partial_t \rho_{00} = -\Gamma_{\text{relax}}(\rho_{00} - \rho^\infty); \quad \Gamma_{\text{relax}} = S_{in} + S_{out}, \quad \rho^\infty = \frac{S_{in}}{S_{in} + S_{out}}.$$  (2.72)

An non-equilibrium occupation is changed with the rate $\Gamma_{\text{relax}}$ until it reaches the equilibrium defined by the pumping rates. The order of magnitude of $\Gamma_{\text{relax}}$ is defined by the system dependent $\Gamma$, which is multiplied with $n^2 + \rho_{rr}(2n + 1)$ which has as rather simple behavior on the temperature and the carrier density in the reservoir. $\Gamma$ depends mainly on the binding energy of the QD levels, i.e. the energetic distance between QD state and WL band edge. The equilibrium occupation does not depend on $\Gamma$, but is defined via the occupation in the resonant level and the phonon distribution. We assume the WL carrier density to be thermalized, i.e. following a Fermi-Dirac distribution.

$$\rho_{rr} = \frac{1}{1 + e^{\beta(\epsilon_r - \mu)}}$$

with $\beta = k_B T$ and the chemical potential $\mu$ which is determined by the carrier density in the WL $n_{WL}$. For a two dimensional reservoir this can be expressed analytically[CK99]:

$$\mu_{eh} = \frac{1}{\beta} \left( \exp \left( \frac{\hbar^2 \pi}{2m_e h} n_{WL} - 2\bar{\rho}_{00} n_{QD} \right) - 1 \right)$$

$2\bar{\rho}_{00} n_{QD}$ presents a correction taking into account the carriers that are captured on average with the QD density $n_{QD}$ and the average occupation of a QD level $\bar{\rho}_{00}$. The carrier density in the reservoir is affected by pumping, e.g. via an applied current or optical excitation. The strength of $\Gamma$ does depend on the wave functions of the system on one hand and on the QD binding energy, i.e. the energetic distance between QD state and WL band edge of the WL continuum on the other, cf. Sec. 1.2.1. Fig. 2.9 shows the dependence of the rates on this binding energy. For energies below $\hbar \omega_{LO}$ our assumptions of no possible one phonon processes fails, above $2\hbar \omega_{LO}$ energy conservation is not possible anymore.

The in- and out-scattering is determined by the pumping strength, which affects the carrier density. Figure 2.10 shows that for no carriers in the reservoir there are no in-scattering processes, but out-scattering is non-vanishing. This is in contrast to rates based on Coulomb interaction, where for an empty reservoir also the out-scattering vanishes [Wil13]. At high pumping strength,
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**Figure 2.9:** Rate $\Gamma$ over binding energy in units $\hbar\omega_{\text{LO}}$ for InAs/GaAs QD material parameters. Near $\Delta = \hbar\omega_{\text{LO}}$ the rate is large due to many continuum states near a one phonon transition, for larger binding energies the number of such states is lowered, thus the rate decreases.

**Figure 2.10:** In- and out-scattering rate as a function of the carrier density in the carrier reservoir. The in-scattering depends mainly on the occupation of the resonant level. For low temperatures it resembles a Fermi function, which is broadened for higher temperatures. Additionally higher temperatures enhance the rates. The out-scattering starts at a non-vanishing contribution proportional to $n^2$ for low carrier densities and decreases for higher carrier densities, due to the blocking in the resonant reservoir state.

However, the resonant levels are completely occupied, hence the in-scattering saturates and the out-scattering vanishes. The most interesting influence is the temperature, which affects the occupation in the resonant states, but more over the phonon density. In Fig. 2.11 the temperature influence is shown. For low temperatures there are no phonons, hence only the in-scattering, proportional to the spontaneous phonon emission $\sim (n + 1)$ is present. For higher temperatures the phonon density is monotonously growing.

The rate equations derived in this chapter can be used to introduce losses or pumping terms in the optical Bloch equations that are the topic of the next chapter.
Figure 2.11: Two phonon rates as a function of the temperature. For high carrier densities the in scattering is decreased due to the decreasing occupation of the resonant level and increases for even higher temperatures due to the increasing phonon occupation. For lower carrier densities the in scattering rate is only increasing. The out scattering rates follow the temperature behavior of $n^2$. 
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3

Non-linear optical response of quantum dots

As described in Sec. 1.2.1, quantum dots (QDs) can be described by a discrete level system, similar to atomic systems, hence in the most simple approximation as a two level system or taking into account higher states in the most simple approximation two uncoupled two level systems. Such two level systems coupled to an electric field semi-classically can described via the so called Bloch equations [AE87]. The Bloch equations are linear differential equations for the electronic quantities, but non-linear in the electric field, which leads to a non-linear optical response and hence a non-linear system in combination with the wave equation. This chapter deals with various approaches to describe the response, which is then used in Chap. 4 to yield wave equations that are capable of describing the propagation of optical pulses through QD media. Section 3.1 deals with the conservative Bloch equations. In Sec. 3.1.1 its well known analytic limits for pulses resonant to the two level system as well as non-resonant constant excitations are revisited, while Sec. 3.1.2 presents a perturbative approach for non-resonant pulses. In Sec. 3.2 dissipative Bloch equations according to the losses and pumping introduced in Chap. 2 are discussed. Here, Sec. 3.2.1 generalizes the perturbative description for the dissipative case. The chapter ends with the description of the Bloch equations in the strong dissipative regime in Sec. 3.2.2.

3.1 Conservative quantum dot Bloch equations

The optical response of a QD is defined by the microscopic polarization. Hence, its dynamics has to be calculated. Using the electronic and semi-classical electric field Hamiltonian $\hat{H} = H_{\text{el}} + H_{\text{el-light}}$ in the Heisenberg equation of motion approach (cf. Sec. 1.3) leads to the well known optical Bloch
3.1. CONSERVATIVE QUANTUM DOT BLOCH EQUATIONS

equations [AE87] (cf. Sec. 1.3) for the microscopic polarizations\(^1\)

\[
\begin{align*}
\hbar \frac{\partial \rho_{12}}{\partial t} &= (\epsilon_2 - \epsilon_1) \rho_{12} + \vec{d}_{21} \cdot \vec{E} (\rho_{11} - \rho_{22}), \\
\hbar \frac{\partial \rho_{21}}{\partial t} &= -\hbar \omega_{21} \rho_{21} - \vec{d}_{12} \cdot \vec{E} u_3,
\end{align*}
\]

(3.1a)

(3.1b)

as well as the equations for the occupations

\[
\begin{align*}
\hbar \frac{\partial \rho_{11}}{\partial t} &= (\vec{E} \cdot \vec{d}_{12} \rho_{12} - \vec{E} \cdot \vec{d}_{21} \rho_{21}), \\
\hbar \frac{\partial \rho_{22}}{\partial t} &= -(\vec{E} \cdot \vec{d}_{12} \rho_{12} - \vec{E} \cdot \vec{d}_{21} \rho_{21}).
\end{align*}
\]

(3.1c)

(3.1d)

The occupations of the levels are from now on described via the inversion \(u_3 = \rho_{22} - \rho_{11}\) which is one when the electron is in the upper level and \(-1\), if the lower level is fully occupied. It is convenient to transform them into a rotating frame with the frequency of the external electric field \(\omega_l\). The field is decomposed into a slow envelope times the oscillation \(\vec{E} = E e^{i\omega_l t} + E^* e^{-i\omega_l t}\) and the microscopic polarizations are treated in the same way \(\tilde{\rho}_{12} = \rho_{12} e^{i\omega_l t} + \rho_{21} e^{-i\omega_l t}\):

\[
\begin{align*}
\partial_t \rho_{12} &= -i\Delta \omega \rho_{12} + i\Omega u_3, \\
\partial_t u_3 &= -4\text{Im}(\Omega^* \rho_{12}),
\end{align*}
\]

(3.2)

with \(\Omega(t) = \frac{d}{dt} E(t)\) being the so called Rabi field and the important system parameter \(\Delta \omega = \omega_{21} - \omega\) labels the detuning between the two level system and the frequency of the light. Here, we additionally applied the rotating wave approximation, where occurring terms with the frequency \(2\omega_l\) in the equation for the inversion are neglected. Also neglected is the dynamics of \(\rho_{21}\), since it is governed by the oscillation \(-\Delta \omega - 2\omega_l\).\(^2,\(^3\) Without any further interactions the phase space is

\[
\begin{align*}
\rho_{22} &\quad \rho_{12} \quad \rho_{11} \\
\epsilon_2 &\quad \varepsilon_1
\end{align*}
\]

\(\vec{E}(t) = E(t) \cos(\omega_l t)\)

Figure 3.1: Two level system of the states 1 and 2. The level occupations are observed via the number operators \(\rho_{11} = \langle a_1^\dagger a_1 \rangle, \rho_{22} = \langle a_2^\dagger a_2 \rangle\). The transition probabilities between the levels are described via the microscopic polarization \(\rho_{12} = \langle a_1^\dagger a_2 \rangle\) and its complex conjugate.

\(^1\)Throughout this chapter quantities with a tilde (\(\tilde{\rho}_{12}\)) represent the full quantities, while those without a tilde (\(\rho_{12}\)) will represent the slowly varying envelope of \(\tilde{\rho}_{12}\).

\(^2\)Note, that for the discrimination between near resonant and far resonant contributions one needs to constitute which level has the higher energy.

\(^3\)Subsequently the macroscopic polarization (cf. Sec. 1.2.3) \(\tilde{P} = P e^{i\omega_l t} + P^* e^{-i\omega_l t}\) is given by \(P = \cdots d_{12} \rho_{12}\).
conserved, i.e. all solutions \((2\Re(\rho_{12}), 2\Im(\rho_{12}), u_3)\) (the so called Bloch vector) lie on a sphere of radius \(|\rho_{12}|^2 + \frac{1}{4}u_3^2\), which is a conserved quantity. Those equations can be solved numerically. However, in the two limits of resonant and non-resonant excitation, with completely different dynamics, analytical solutions can be obtained.

### 3.1.1 Exact solutions of the optical Bloch equations

To recap the exact limits of the conservative Bloch equations we follow Ref. [AE87]. In the regime of resonant excitation \(\Delta \omega = 0\) this system is exactly solvable for real pulse envelopes

\[
\partial_t \rho_{12} = i\Omega u_3, \quad \partial_t u_3 = -4\Im(\rho_{12})\Omega.
\]

Their solutions depend on the pulse area

\[
\rho_{12} = i \sin \left( \int_{-\infty}^{t} \Omega(t')dt' \right), \quad u_3 = -\cos \left( \int_{-\infty}^{t} \Omega(t')dt' \right).
\]

The shown solutions represent the typical initial conditions of no polarization \(\rho_{12}(-\infty) = \rho_{12}(0) = 0\) and the electron being in the lower level \(u_3(-\infty) = u_3(0) = -1\). The state of the system after an excitation does not depend on the shape of the envelope, but only on its integral. Figure 3.2a shows the solution to Eq. (3.3) for various pulse areas. The initial state of the electronic system is only restored for pulse areas with a multiple of \(2\pi\). The optical response in this case results in the sine-Gordon equation briefly discussed in Sec. 4.1.

For continuous wave excitation \(\Omega(t) = \Omega_0\) well known so called Rabi oscillations with the Rabi frequency \(\Omega_0\) occur:

\[
\rho_{12} = i \sin \Omega_0 t, \quad u_3 = -\cos \Omega_0 t.
\]

In the case of such a stationary field the Bloch equations can be analytically solved also for a non-vanishing detuning [AE87]. We start by writing Eqs. 3.2 in a matrix representation

\[
\frac{\partial}{\partial t} \begin{pmatrix} \rho_{12} \\ \rho_{21} \\ u_3 \end{pmatrix}(t) = i \begin{pmatrix} -\Delta \omega & 0 & \Omega \\ 0 & \Delta \omega & -\Omega^* \\ 2\Omega^* & -2\Omega & 0 \end{pmatrix} \begin{pmatrix} \rho_{12} \\ \rho_{21} \\ u_3 \end{pmatrix}(t).
\]

The dynamics of this equation can be calculated by diagonalizing the time independent matrix which has the eigenvalues \(0, \pm \sqrt{\Delta \omega^2 + 4|\Omega|^2}\). The non-zero terms correspond to a renormalized Rabi frequency. In order to solve the system, one needs to know the unitary transformations \(T\) leading to the diagonalization [AE87]:

\[
\begin{pmatrix} 0 & 0 & 0 \\ 0 & \sqrt{\Delta \omega^2 + 4|\Omega|^2} & 0 \\ 0 & 0 & -\sqrt{\Delta \omega^2 + 4|\Omega|^2} \end{pmatrix} = T^{-1} \begin{pmatrix} -\Delta \omega & 0 & \Omega \\ 0 & \Delta \omega & -\Omega^* \\ 2\Omega^* & -2\Omega & 0 \end{pmatrix} T.
\]
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Since the field is time independent, knowing the transformation matrices yields the solution:

\[
\begin{pmatrix}
\rho_{12} \\
\rho_{21} \\
u_3
\end{pmatrix}(t) = T \begin{pmatrix}
1 & 0 & 0 \\
0 & e^{i\sqrt{\Delta\omega^2 + 4|\Omega|^2} t} & 0 \\
0 & 0 & e^{-i\sqrt{\Delta\omega^2 + 4|\Omega|^2} t}
\end{pmatrix} T^{-1} \begin{pmatrix}
\rho_{12} \\
\rho_{21} \\
u_3
\end{pmatrix}(0).
\]

Using the typical starting condition \(u_3 = -1, \rho_{12} = 0\) results in the exact solution:

\[
\rho_{12} = \frac{\Omega \left(i\sqrt{\Delta\omega^2 + 4|\Omega|^2} \sin \left( t\sqrt{\Delta\omega^2 + 4|\Omega|^2} \right) + \Delta\omega \left(1 - \cos \left(t\sqrt{\Delta\omega^2 + 4|\Omega|^2}\right)\right)\right)}{\Delta\omega^2 + 4|\Omega|^2},
\]

\[u_3(t) = -\frac{\Delta\omega^2 + 4|\Omega|^2 \cos \left(t\sqrt{\Delta\omega^2 + 4|\Omega|^2}\right)}{\Delta\omega^2 + 4|\Omega|^2}.
\]

All quantities oscillate with the renormalized Rabi frequency \(\sqrt{\Delta\omega^2 + 4|\Omega|^2}\). Inversion and real

![Graphs showing the behavior of \(\rho_{12}\) and \(u_3\) over time for different detunings.](a) Pulsed excitation on resonance for different pulse areas (b) Non-resonant continuous excitation for different detunings.

**Figure 3.2:** Exact solutions of the optical Bloch equations for a) resonant pulsed optical fields with different pulse areas and b) continuous optical fields for different detunings. The final state of the system is determined solely by the area of the interacting pulse. For areas that are a multiple of \(2\pi\), the final state recovers the initial state. For a temporally constant field (b) polarization and inversion are oscillations with the (renormalized) Rabi frequency. With higher detuning the effective frequency of the oscillations rises, whereas the amplitude and the mean value decreases.

Part of the polarization are in phase, while the imaginary part is shifted by \(\pi/2\). The important quantity characterizing the behavior is the detuning in relation to the excitation strength \(x = 2|\Omega|/\Delta\omega\). Figure 3.2(b) shows the dynamics depending on the detuning. The inversion oscillates around the
value \(- (1 + x^2)^{-1}\) with an amplitude \(x^2(1 + x^2)^{-1}\). The real part of the polarization has a mean as well as an amplitude of \(0.5x(1 + x^2)^{-1}\), while the imaginary part oscillates around zero with an amplitude \(0.5x(1 + x^2)^{-1/2}\).

There are certain properties of the non-resonant response to pulses we can see already here. For weak fields the oscillations grow faster, as the amplitude becomes smaller. For this case \(|2\Omega| < |\Delta\omega|\) the mean values can be described as geometric series of the field:

\[
\bar{\rho}_{12} = 0.5 \sum_{n=0}^{\infty} (-1)^n \left( \frac{2\Omega}{\Delta\omega} \right)^{2n+1}, \quad \bar{u}_3 = \sum_{n=0}^{\infty} (-1)^n \left( \frac{2\Omega}{\Delta\omega} \right)^{2n}.
\]  

(3.4a)

The mean value of the polarization is composed of odd orders of the field, whereas the inversion consists of even orders. The series are alternating and every order becomes smaller, hence every order presents a correction for the order before.

3.1.2 Perturbative approach for non-resonant excitation

In the last section we have revisited the well known (e.g. from [AE87]) solutions for arbitrary pulse widths and no detuning as well as for temporally constant pulses with arbitrary detuning. Here, we present a perturbative approach for the cases in between. Perturbative treatment of the Bloch equations is an approach, where the number of interactions with the field is counted and thus convenient for the description of four-wave-mixing techniques [DMR+10; MRH+10; Muk95; DWE+12]. For our approach the constraint is that the pulse is slow, or spectrally thin in comparison to the detuning: \(\frac{\partial \Omega}{\partial t} \ll \Delta\omega\). We start by integrating the Bloch equations (3.2) formally:

\[
\rho_{12}(t) = i \int_{-\infty}^{t} dt' e^{i\Delta\omega(t' - t)}\Omega(t')u_3(t'), \quad (3.5a)
\]

\[
u_3 = u_3^{(0)} - 4 \int_{-\infty}^{t} dt' \text{Im}(\Omega^*(t')\rho_{12}(t')). \quad (3.5b)
\]

Equation (3.5a) shall be expanded with respect to the detuning, which can be done via multiple integration by parts, or a treatment in Fourier space:

\[
\rho_{12}(\omega) = i \mathcal{F}(e^{-i\Delta\omega t}) \mathcal{F}(\Omega(t)u_3(t)),
\]

\[
\mathcal{F}(e^{-i\Delta\omega t}) = \frac{i}{\omega + \Delta\omega} \sum_{n=0}^{\infty} \left( -\frac{\omega}{\Delta\omega} \right)^n \approx \frac{i}{\Delta\omega} - \frac{i\omega}{\Delta\omega^2}.
\]

This expansion is the better in the lower orders the smaller \(\omega\) is compared to the detuning \(\Delta\omega\). Thus, this is a good description for spectrally thin pulses \(\Omega(\Delta\omega) \ll \Omega(0)\), which vanish for larger

\[
\frac{\partial \Omega}{\partial t} \sim \frac{1}{T} \quad \text{is inversely proportional to the width of a pulse } T, \quad \text{e.g. for a Gaussian } \frac{\partial \Omega}{\partial t} = \frac{1}{T^2} \quad \text{and } \frac{1}{T} \text{ can be considered a constant.}
\]

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![Graph](image)

**Figure 3.3:** Visualization of the slowly envelope approximation in Fourier space by the exactly solved polarization for different pulse widths. Spectrally broad pulses (upper figure) excite the system’s resonance at \( \Delta \omega \) and thus create oscillations with the detuning frequency. For longer pulses, i.e. spectrally thin ones, the resonance is not as strongly excited, but the polarization follows the field adiabatically and the oscillations in time domain vanish.

\( \omega \) (cf. Fig. 3.3), i.e. \( \frac{\omega}{\Delta \omega} \ll 1 \). Back in time domain this results in a series of differential operators, with the aforementioned constraint \( \partial_t \Omega \ll \Delta \omega \Omega \):

\[
\rho_{12}(t) = -\frac{1}{\Delta \omega} \sum_{n=0}^{\infty} \left( -\frac{i}{\Delta \omega} \right)^n \Omega(t) u_3(t) \partial_t \Omega \ll \Delta \omega \Omega \approx \frac{1}{\Delta \omega} \left( \Omega(t) u_3(t) + i \frac{1}{\Delta \omega} \partial_t \Omega(t) u_3(t) \right).
\]

(3.6)

The polarization follows the electric field adiabatically, this can be thought of oscillations of the polarization being averaged out.

However, the dynamics of the polarization and the inversion are still coupled. In order to arrive at a set of uncoupled differential equations an expansion in the order of the electric field is applied:

\[
\rho = \sum_i \rho^{(i)}; \quad \rho^{(i)} \sim |\Omega|^i; \quad u_3 = \sum_i u_3^{(i)}; \quad u_3^{(i)} \sim |\Omega|^i.
\]

Equations (3.5a) then couple a quantity of the order \( n + 1 \) to the complete dynamics of the electric field times a quantity of the order \( n \).

\[
\rho_{12}^{(n+1)}(t) = i \int_{-\infty}^{t} dt' e^{i \Delta \omega(t'-t)} \Omega(t') u_3^{(n)}(t'), \tag{3.7a}
\]

\[
u_3^{(n+1)} = -4 \int_{-\infty}^{t} dt' \text{Im}(\Omega^*(t') \rho_{12}^{(n)}(t')). \tag{3.7b}
\]
Now the dynamics can be treated iteratively, assuming that the zeroth order is constant, i.e. no other interactions than the field, and inserting this value into the first order:

\[ u_3^{(0)} = \text{const.,} \quad \rho_{12}^{(0)} = 0, \quad (3.8) \]

\[ \rho_{12}^{(1)}(t) \approx \frac{u_3^{(0)}}{\Delta \omega} \left( \Omega(t) + i \frac{1}{\Delta \omega} \partial_t \Omega(t) \right). \quad (3.9) \]

Here, the first order inversion \( u_3^{(1)} \) vanishes due to the starting condition \( \rho_{12}^{(0)} = 0 \), which leads to the property that the inversion exists only in even orders and the polarization only in odd orders (for two level systems without permanent dipoles). Subsequently, the second order inversion can be derived from the first order polarization, third order polarization from the second order inversion and so on. This procedure can be taken to arbitrary orders and written in general:

\[
\begin{align*}
 u_3(t) &= \sum_{n=0}^{\infty} (-1)^n \frac{1}{\Delta \omega^{2n}} \frac{(2n)!}{(n!)^2} |\Omega|^{2n} \left[ \rho_{12}^{(0)} \right], \quad (3.10a) \\
 \rho_{12}(t) &= \sum_{n=0}^{\infty} (-1)^n \frac{1}{\Delta \omega^{2n+1}} \frac{(2n)!}{(n!)^2} \left( \Omega(t)|\Omega|^{2n}(t) + i \frac{1}{\Delta \omega} \partial_t |\Omega|^{2n}(t')\Omega(t') \right) \left[ u_3^{(0)} \right]. \quad (3.10b)
\end{align*}
\]

We proof this via mathematical induction in App. B. Figure 3.4 compares the perturbative solution up to \( n = 2 \) to the numerically exact solution. For larger fields the perturbative description is not accurate in low orders, since saturating effects are missing. For too short pulses oscillations of the polarization reside in the system which cannot be described due to the slowly varying envelope approximation in Eq. 3.6, where we omitted oscillating behavior of the inversion. The oscillating behavior of the exact non-resonant solution is not reproduced, but the mean values can be approximated. The exact solution of the Bloch equations for a detuned steady field (cf. Eqs. (3.4)) cannot be reproduced since it relies on the oscillations of the quantities, that are not present in the perturbative, adiabatic treatment. However the presented solution for pulses is close to the mean value of the steady field solution.

For further use, i.e. Chap. 4, we omit the time derivative consistent with the slowly varying envelope approximation.\(^5\) We will use the solution up to fifth order, namely up to \( n = 2 \):

\[ \rho_{12}(t) \approx \left[ \frac{1}{\Delta \omega} \Omega(t) - \frac{2}{\Delta \omega^3} \Omega(t)|\Omega|^2(t) + \frac{6}{\Delta \omega^5} \Omega(t)|\Omega|^4(t) \right] u_3^{(0)}. \quad (3.11) \]

The non-linear response presented here corresponds to the Kerr effect and leads to the Nonlinear Schrödinger equation, if taken into account as optical response to a wave equation. This equation is discussed in detail in Sec. 4.33.

\(^5\)For real pulse amplitudes this also omits the imaginary part of the polarization. In contrast to the real part it has a small amplitude that even weakens with slower pulses.
3.2 Dissipation in QD Bloch equations in relaxation time approximation

The last section covered conservative Bloch equations. However, the aim of this work is to deal with dissipative wave equations derived from QD dynamics (cf. Sec. 4.3.3). There are two ways to turn on dissipation:

i) input and output of carriers (relaxation/capturing) due to interaction with a carrier reservoir as presented in Sec. 2.2. We will refer to this process as pumping.

ii) On the other hand there is loss due to decoherence, i.e. loss of quantum mechanical information. Such decoherence is caused by capture and relaxation processes as in i), but also without change of occupations as so called pure dephasing as discussed in Sec. 2.1.1.

As in the previous section one can derive the equations of motion from the Hamiltonian

\[ \hat{H} = \hat{H}_{el} + \hat{H}_{el-field} \]

via Heisenberg’s equation of motion \( \mathcal{i} \hbar \frac{\partial}{\partial t} O = [O, \hat{H}] \). This time we additionally use a phenomenological treatment of the diagonal interaction with a phonon bath \( \hat{H}_{ph,di} \) like in Sec. 2.1.1 as well as phonon mediated interaction with carrier reservoirs \( \hat{H}_{ph,nd} \) as in Sec. 2.2, to introduce relaxation.
and dephasing rates.

\[ \begin{align*}
    i\hbar \frac{\partial}{\partial t} \tilde{\rho}_{12} &= (\epsilon_2 - \epsilon_1) \tilde{\rho}_{12} - \vec{d}_{21} \cdot \vec{E} u_3 - i\hbar \gamma \tilde{\rho}_{12}, \\
    i\hbar \frac{\partial}{\partial t} \rho_{11} &= (\vec{E} \cdot \vec{d}_{12} \tilde{\rho}_{12} - \vec{E} \cdot \vec{d}_{21} \tilde{\rho}_{21}) - i\hbar \Gamma_1 (\rho_{11} - \rho_{11}^\infty), \\
    i\hbar \frac{\partial}{\partial t} \rho_{22} &= -(\vec{E} \cdot \vec{d}_{12} \tilde{\rho}_{12} - \vec{E} \cdot \vec{d}_{21} \tilde{\rho}_{21}) - i\hbar \Gamma_2 (\rho_{22} - \rho_{22}^\infty).
\end{align*} \]

(3.12a) \hfill (3.12b) \hfill (3.12c)

The polarization dephases with a rate \( \gamma \), and the densities in the states \( 1, 2 \) are driven to their steady state occupations \( \rho_{\infty} \) with rates \( \Gamma_{1/2} \) due to the interaction with a respective reservoir. Those rates or time constants are calculated in Sec. 2.2. To create similar equations as in the conservative case in Sec. 3.1 we have to use the inversion as well as the total carrier density and thus relaxation rates in center of mass and relative notation.

\[ \Gamma = \frac{1}{2} (\Gamma_1 + \Gamma_2), \quad \Delta \Gamma = \frac{1}{2} (\Gamma_1 - \Gamma_2). \]

(3.13)

In comparison an additional equation for the total carrier number \( u_4 = \rho_{11} + \rho_{22} \) shows up. This yields the dissipative Bloch equations, where neither energy, nor occupation is conserved.

\[ \begin{align*}
    \dot{\rho}_{12} &= -i \Delta \omega \rho_{12} + i \Omega u_3 - \gamma \rho_{12}, \\
    \dot{u}_3 &= -4 \text{Im}(\Omega^* \rho_{12}) - \Gamma (u_3 - u_3^\infty) - \Delta \Gamma (u_4 - u_4^\infty), \\
    \dot{u}_4 &= -\Delta \Gamma (u_3 - u_3^\infty) - \Gamma (u_4 - u_4^\infty).
\end{align*} \]

(3.14a) \hfill (3.14b) \hfill (3.14c)

Here, the phase space is not conserved, but in the long time limit the Bloch sphere \( (2 \text{Re}(\rho_{12}), 2 \text{Im}(\rho_{12}), u_3) \) will shrink to one point \( (0, 0, u_3^\infty) \). We will concentrate on \( \Delta \Gamma = 0 \), i.e. the symmetric case where both levels have similar relaxation rates from the reservoir, which leads to a quasi carrier number conservation.

\[ \begin{align*}
    \dot{\rho}_{12} &= -i \Delta \omega \rho_{12} + i \Omega u_3 - \gamma \rho_{12}, \\
    \dot{u}_3 &= -4 \text{Im}(\Omega^* \rho_{12}) - \Gamma (u_3 - u_3^\infty).
\end{align*} \]

(3.15a) \hfill (3.15b)

Note that this approximation corresponds to the usual treatment in atomic optics, where \( \Gamma \) corresponds rather to non-radiative recombination or direct pumping by another optical field, than to a coupling to a carrier reservoir. Hence we deal with the well known Bloch equations in relaxation time approximation [AE87].

### 3.2.1 Perturbative approach

As in the conservative case in Sec. 3.1 we can solve the Bloch equations formally and expand in orders of the field. The approach is indeed the same, but the steps are more elaborate in the...
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dissipative case due to additional terms. Here, we do not give a general formula, but explicitly derive the solution up to the fifth order. Equations (3.15) are integrated formally.

\[
\rho_{12}(t) = i \int_{-\infty}^{t} dt' e^{i(\Delta \omega - i \gamma)(t' - t)} \Omega(t') u_3(t'),
\]

\[
u_3 = u_3^\infty - 4 \int_{-\infty}^{t} dt' e^{\Gamma(t' - t)} \left( \text{Im}(\Omega^*(t') \rho_{12}(t')) \right).
\]

As in Eq. (3.6), we use the approximation of a slowly varying pulse envelope. This time the linear spectra of the pulse are not delta like, but also broadened by $\gamma$. Now, the prerequisite of the slowly varying envelope is $\frac{d \Omega}{dt} \ll \sqrt{\Delta \omega^2 + \gamma^2}$ which is, as before, the spectral distance from the two level system’s resonance, which is now shifted on the imaginary axis by $\gamma$, cf. Fig. 3.5. By applying the approximation we get:

\[
\rho_{12}(t) = -\frac{1}{\Delta \omega - i \gamma} \sum_{n=0}^{\infty} \left( -\frac{i \partial t}{\Delta \omega - i \gamma} \right)^n \Omega(t) u_3(t)
\]

\[
\approx \frac{\Delta \omega + i \gamma}{\Delta \omega^2 + \gamma^2} \left( \Omega(t) u_3(t) + \frac{1}{\Delta \omega + i \gamma} \partial_t \Omega(t) u_3(t) \right)
\]

\[
= \frac{1}{a_+} \left[ (\Delta \omega + i \gamma) \Omega(t) u_3(t) + \frac{1}{a_+} (ia_- - 2 \Delta \omega \gamma) \partial_t \Omega(t) u_3(t) \right],
\]

with $a_\pm = \Delta \omega^2 \pm \gamma^2$.

As is Sec. 3.1 we choose a perturbative approach by iteratively solving the polarization in orders of the electric field:

\[
u_3^{(0)} = \text{const.} = u_3^\infty,
\]

\[
\rho_{12}^{(1)}(t) = u_3^{(0)} \left[ (\Delta \omega + i \gamma) \Omega(t) + \frac{1}{a_+} (ia_- - 2 \Delta \omega \gamma) \partial_t \Omega(t) \right],
\]

\[
u_3^{(2)} = -4 \int_{-\infty}^{t} dt' e^{\Gamma(t' - t)} \left( \text{Im}(\Omega^*(t') \rho_{12}^{(1)}(t')) \right).
\]
Figure 3.5: Visualization of the approximation Eq. (3.17) in Fourier space. The resonance of the two level system is expanded around the center frequency of the pulse \( \omega = 0 \). a) the resonance of the two level system is shifted by \( \gamma \) on the imaginary axis. For the quality of the approximation \( \omega \) is compared to the distance of the resonance, which is a circle with the radius \( \sqrt{\Delta \omega^2 + \gamma^2} \). b) a short (spectrally broad) pulse excites the system. In the upper panel there is no dephasing such that the resonance of the system is excited and oscillations occur in the time domain. With a small dephasing (middle panel) the same pulse results in a weaker (and broadened) excitation of the system and a more significant adiabatic excitation, in the time domain the oscillations are damped out. For strong dephasing (lower panel) the pulse excites the system adiabatically and exists only on the timescale of the pulse.

to the fifth order. We use again Eq. (B.5): \( \Omega \partial_t \Omega^* = \frac{1}{2} \partial_t |\Omega|^2 - i |\Omega|^2 \partial_t \phi \), yielding the second order inversion explicitly

\[
\begin{align*}
  u_3^{(2)} &= -4 \frac{u_3^{(0)}}{a_+} \left( \gamma \int_{-\infty}^{t} dt' e^{\Gamma(t'-t)}|\Omega|^2 + \frac{a_-}{2a_+} \int_{-\infty}^{t} dt' e^{\Gamma(t'-t)} \partial_t |\Omega|^2 \\
  &\quad+ \frac{2 \Delta \omega \gamma}{a_+} \int_{-\infty}^{t} dt' e^{\Gamma(t'-t)} |\Omega|^2 \partial_{t'} \phi(t') \right). \tag{3.21}
\end{align*}
\]

In comparison to the conservative case an additional memory integral \( M^{(1)}(t) = \int_{-\infty}^{t} dt' e^{\Gamma(t'-t)} |\Omega|^2 \) enters proportional to \( \gamma \) and has the property that \( \partial_t M^{(1)}(t) = |\Omega|^2 (t) - \Gamma M^{(1)}(t) \). This term represents the damped pulse energy. The second integral can be treated via integration by parts:

\[
\int_{-\infty}^{t} dt' e^{\Gamma(t'-t)} \partial_{t'} |\Omega|^2 = |\Omega|^2 - \Gamma \int_{-\infty}^{t} dt' e^{\Gamma(t'-t)} |\Omega|^2 = |\Omega|^2 - \Gamma M^{(1)}(t) = \partial_t M^{(1)}(t). \tag{3.22}
\]

Luckily, this type of memory has the general property that integration and differentiation interchange, which will be advantageous for higher orders:

\[
\int_{-\infty}^{t} dt' e^{\Gamma(t'-t)} \partial_{t'} f(t') = f(t) - \Gamma \int_{-\infty}^{t} dt' e^{\Gamma(t'-t)} f(t') = \partial_t \int_{-\infty}^{t} dt' e^{\Gamma(t'-t)} f(t'). \tag{3.23}
\]
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For the further procedure we will neglect the chirp term $\partial_t \varphi$, in consistence to the slowly varying envelope approximation. This leads to a compact description of the second order inversion:

$$u_3^{(2)} = -4 \frac{u_3^{(0)}}{a_+} \left( \frac{a_+ - 1}{a_+} \partial_t M^{(1)} + \gamma M^{(1)} \right). \tag{3.24}$$

Note, that this $\partial_t M$ corresponds to the adiabatic following behavior from the conservative solution Eq. (3.10a). Hence, explicitly written, the inversion now consists of an adiabatically following term, as well as a memory term, that includes the relaxation via the rate $\Gamma$:

$$u_3^{(2)} = -4 \frac{u_3^{(0)}}{a_+} \left( \frac{a_+ - 1}{a_+} \left( \frac{\Omega^2}{2} + \left( \gamma - \frac{\Gamma a_+}{2} \right) M^{(1)} \right) \right). \tag{3.25}$$

The third order polarization is acquired straight forward, inserting the second order inversion Eq. (3.24) into Eq. (3.17) leads to the compact representation:

$$\rho_{12}^{(3)} = -4 \frac{u_{12}^{(0)}}{a_+^2} \left[ (\Delta \omega + i \gamma) \left( \frac{a_+ - 1}{2a_+} \Omega(t) \partial_t M^{(1)} + \gamma \Omega(t) M^{(1)}(t) \right) \right. \left. + \left( \frac{a_+ - 2 \Delta \omega \gamma}{a_+} \right) \left( \frac{a_+ - 1}{2a_+} \partial_t \Omega(t) \partial_t M^{(1)} + \gamma \partial_t \Omega(t) M^{(1)} \right) \right]. \tag{3.26}$$

To distinguish the adiabatic and memory contributions, the derivatives $\partial_t M^{(1)} = |\Omega|^2 - \Gamma M$ are explicitly applied, leading to:

$$\rho_{12}^{(3)} = -4 \frac{u_{12}^{(0)}}{a_+^2} \left[ \left( \frac{a_+ - 1}{2a_+} \left( 1 + 2 \Gamma \frac{a_+ - 1}{a_+} \right) - 2 \frac{\gamma^2}{a_+} \right) + i \frac{a_+}{2a_+} \left( 3 \gamma - 2 \Gamma \frac{a_+ - 1}{2a_+} \right) \Omega(t) |\Omega|^2 
+ \left( \frac{a_+ - 2 \Delta \omega \gamma}{a_+} \right) \left( \frac{a_+ - 1}{2a_+} \partial_t \Omega(t) |\Omega|^2 + \left( \gamma - \Gamma \frac{a_+}{2a_+} \right) M^{(1)}(t) \partial_t \Omega(t) \right) \right] 
+ \left[ \Delta \omega + 2 \Gamma \frac{\Delta \omega \gamma}{a_+} + i \left( \gamma - \Gamma \frac{a_+}{a_+} \right) \right] \left( \gamma - \Gamma \frac{a_+}{2a_+} \right) M^{(1)}(t) \Omega(t) \right]. \tag{3.27}$$

The third order polarization is composed of four components: an adiabatically following part, a part following the derivative of the field, terms depending on the memory as well as a mixture of memory and derivative terms. Every component contributes to real and imaginary part of the polarization.

By means of those lowest orders we can describe already the influence of dephasing on relaxation rates. As in Sec. 3.1 the higher orders are a corrective for the perturbation theory. Third order will be of major importance in Sec. 4.3.3. The effect of dephasing can be investigated in the two limits of fast and slow pumping.
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Figure 3.6: Components of inversion, and polarization, Eqs. (3.28), up to second or third order, respectively, for a Gaussian pulse in the slow pumping limit \( \Gamma = 0 \). The solid line is the perturbative description, whereas the shaded area represents the full numerical solution of the Bloch equations. The first column represents the conservative case, cf. Sec. 3.1.2: For vanishing dephasing the real part of the polarization as well as the inversion follow the pulse adiabatically, while the imaginary part of the polarization follows the derivative of the pulse. With increasing dephasing, the influence of memory terms (dash-dotted lines) increases until the dephasing reaches the value of the detuning. For higher dephasing the leading prefactor \( a_{-1}^{-1} \) affects the overall response: The dephasing destroys the coherence of the polarization and as such diminishes the interaction with the electric field.

**Slow pumping limit** \( \Gamma \rightarrow 0 \). Here, \( M^{(1)} \) becomes the pulse energy \( \int_{-\infty}^{t} dt' |\Omega|^2 \), whose derivative is just the absolute square of the pulse \( |\Omega|^2 \). In this case the inversion does not return to its steady state after the pulse passed through. Similar to the conservative case its value after the pulse depends on the pulse’s energy. This influence is large if the dephasing rate is faster, since the polarization vanishes and cannot assist the decay of the inversion. The influence of dephasing is shown in
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Fig. 3.6. The slow pumping limit of the polarization \( \rho_{12} = \rho_{12}^{(1)} + \rho_{12}^{(3)} \) is:

\[
\text{Re}(\rho_{12}) \approx \frac{u_3^{(0)}}{a_+} \Delta \omega \left[ \left( 1 - \frac{2 \Delta \omega^2}{a_+^2} |\Omega|^2 + 6 \frac{\gamma^2}{a_+^2} |\Omega|^2 - 4 \frac{\gamma}{a_+} M^{(1)}(t) \right) \Omega(t) \right.
\]
\[
- 2 \frac{\gamma}{a_+} \left( 1 - \frac{\gamma}{a_+} \left( \frac{a_-}{a_+} |\Omega|^2 - 4 \frac{\gamma}{a_+} M^{(1)}(t) \right) \partial_t \Omega(t) \right] ,
\]

(3.28a)

\[
\text{Im}(\rho_{12}) \approx \frac{u_3^{(0)}}{a_+} \left[ \left( 1 - \frac{6a_-}{a_+^2} |\Omega|^2 - 4 \frac{\gamma}{a_+} M^{(1)}(t) \right) \left( \gamma \Omega(t) + \frac{a_-}{a_+} \partial_t \Omega(t) \right) \right] ,
\]

(3.28b)

\[
u_3 = \nu_3^{\infty} \left[ 1 - \frac{4}{a_+} \left( \frac{a_-}{a_+} |\Omega|^2 \right) + \gamma M^{(1)} \right] .
\]

(3.28c)

We can see for non-vanishing \( \gamma \) (specifically \( \gamma > \frac{a_-}{a_+} \frac{\gamma^2}{a_+^2} > \frac{1-\gamma}{\gamma+1} \)) the inversion is dominated by the memory contributions, which has its maximum at \( \gamma = \Delta \omega \) (because \( \frac{\gamma}{a_+} \) is maximal there). The imaginary part of the polarization is dominated by the adiabatic behavior for larger \( \gamma \) also due to the same relation. Since the behavior can be mostly characterized with the parameter \( \frac{\gamma}{a_+} \), the effective dephasing is influenced by the detuning. Figure 3.6 also shows that, as stated before, the approach gets better with increasing \( \gamma \), since the slowly varying envelope approximation gets better. The conservative limit (cf. Eq. (3.11)) is reproduced for vanishing dephasing \( \gamma \to 0 \).
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Figure 3.7: Components of inversion, and polarization, Eqs. (3.30), up to second or third order, respectively, for a Gaussian pulse in the fast pumping limit $\Gamma = 0.5 \times \gamma$. The solid line is the perturbative description, whereas the shaded area represents the full numerical behavior of the Bloch equations. The first column represents the conservative case, cf. Sec. 3.1.2: For vanishing dephasing the real part of the polarization as well as the inversion follow the pulse adiabatically, while the imaginary part of the polarization follows the derivative of the pulse. The adiabatic description of the polarization approximates the full solution well, getting better with increasing dephasing. The inversion is described by the memory terms, that also show adiabatic behavior, cf. Fig. 3.7.

**Fast pumping** $\partial_t |\Omega|^2 \ll \Gamma |\Omega|^2$ Now the relaxation rate of the inversion is much faster, then the pulse duration, and the memory integrals can be approximated by the intensity:

$$M^{(n)} = \Gamma^{-1} |\Omega|^{2n} - \Gamma^{-1} \partial_t M^{(n)} \approx \Gamma^{-1} |\Omega(t)|^{2n}. \quad (3.29)$$

Polarization and inversion in the fast pumping limit consist only of adiabatic terms:

$$\rho_{12} = \frac{u_3^{(0)}}{a_+} \left[ (\Delta \omega + i\gamma) \left( 1 - \frac{4}{a_+} \frac{\gamma |\Omega|^2}{\Gamma} \right) \Omega(t) + \left( \frac{\gamma \Gamma}{a_+} - 2 \frac{\Delta \omega \gamma}{a_+} \right) \left( 1 - 12 \frac{\gamma}{\Gamma} |\Omega|^2 \right) \partial_t \Omega(t) \right],$$

$$u_3 = u_3^{\infty} \left[ 1 - \frac{4}{a_+} \frac{\gamma}{\Gamma} |\Omega|^2 \right].$$

(3.30a)  
(3.30b)
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The strength of the higher order terms is given by the ratio $\frac{\gamma}{\Gamma}$. Since relaxation ($\Gamma$) and dephasing rate ($\gamma$) are correlated $\gamma = 2\Gamma + \gamma_{\text{pure}}$ (cf. Sec. 2.2.3), in the fast pumping limit the dephasing is completely defined by the relaxation rates $\gamma \approx 2\Gamma$. The fast pumping limit up to second/third order is shown in Fig. 3.7 for different values of dephasing. All quantities show clear adiabatic behavior. As in the slow pumping case the real part polarization in monotonously decreasing with increasing dephasing and the imaginary part has the maximal strength at $\gamma = \Delta \omega$ because $\frac{\gamma}{\pi}$ is maximal there. Also as in the slow pumping case the approximate description gets better with stronger dephasing. The main difference is the now adiabatic dispersion, which is clear, since in the fast pumping limit the influence of the memory becomes adiabatic.

Higher order corrections. To calculate the fourth order inversion the next step is again multiplying Eq. (3.26) by $\Omega$ and bringing the derivative in front, then write the memory terms of the next order using

$$\Omega^* \partial_t \Omega M_0^{(1)} = \frac{1}{2} |\Omega|^2 M_0^{(1)} + \frac{1}{2} |\Omega|^2 \partial_t M_0^{(1)} - i |\Omega|^2 M_0^{(1)} \partial_t \varphi. \quad (3.31)$$

Again the time dependence of a pulse’s phase $\partial_t \varphi$ is neglected, leading to the fourth order inversion in the compact form:

$$u_3^{(4)} = (-4)^2 \frac{u_3^{(0)}}{a_+^2} \left[ \gamma \left( \frac{a_-}{2a_+} N_1^{(2)} + \gamma N_0^{(2)} \right) + \frac{a_-}{a_+} \left( \frac{a_-}{2a_+} \partial_t N_1^{(2)} + \frac{a_-}{2a_+} N_2^{(2)} + \gamma \partial_t N_0^{(2)} + \gamma N_1^{(2)} \right) \right], \quad (3.32)$$

with the generalized memory integrals

$$N_i^{(j)} = \int_{-\infty}^{t} dt' e^{\Gamma(t-t')} |\Omega|^2 \partial_t^j N_i^{(j-1)}$$

containing derivatives of lower order generalized memory integrals. For $i = 1$ it is just the memory integral we used before $N^{(1)} = M^{(1)}$. As we will see shortly they can be expressed as a sum of memory integrals without derivatives. Here, especially $N_j^{(2)} = \int_{-\infty}^{t} dt' e^{\Gamma(t-t')} |\Omega|^2 \partial_t^2 M^{(1)}$ is valid. For further use the explicit form can be acquired via extracting the derivatives in the memory integrals:

$$N_0^{(2)} = N_0^{(2)}, \quad \partial_t N_0^{(2)} = |\Omega|^2 M^{(1)} - \Gamma N_0^{(2)},$$
$$N_1^{(2)} = M^{(2)} - \Gamma N_0^{(2)}, \quad \partial_t N_1^{(2)} = \left( |\Omega|^4 - \Gamma M^{(2)} \right) - \Gamma \left( |\Omega|^2 M^{(1)} - \Gamma N_0^{(2)} \right),$$
$$N_2^{(2)} = \frac{1}{2} \partial_t M^{(2)} - \Gamma N_1^{(2)} = \frac{1}{2} \left( |\Omega|^4 - 3\Gamma M^{(2)} \right) + \Gamma^2 N_0^{(2)}.$$

The fourth order inversion consists of adiabatic following terms $\sim |\Omega|^4$, memory of the pulse’s energy $\sim M^{(1)}$ and the corresponding square $\sim M^{(2)}$ as well as generalized memory $\sim N_0^{(2)}$. It
reads explicitly written:

\[
\begin{align*}
u_3^{(4)} &= (-4)^2 \frac{u_3^{(0)}}{a_+^2} \frac{a_-}{2a_+} \left[ \frac{a_-}{2a_+} \frac{3}{2} \Omega^4 + \left( 2 \gamma - \frac{a_-}{2a_+} \frac{5}{2} \Gamma \right) M^{(2)} \right] \\
& \quad + \left( \gamma - \frac{a_-}{2a_+} \Gamma \right) \left[ \Omega^2 M^{(1)} + \left( \frac{2a_+}{a_-} \gamma^2 - 3 \Gamma \gamma + 2 \frac{a_-}{2a_+} \Gamma^2 \right) N_0^{(2)} \right]. \quad (3.33)
\end{align*}
\]

Higher orders are proportional to \(a_+^{-(n+1)/2}\), which means for increasing \(\gamma\) the higher orders become less important. The polarization of fifth order is acquired by inserting Eq. (3.33) and its derivative

\[
\partial_t u_3^{(4)} = (-4)^2 \frac{u_3^{(0)}}{a_+^2} \frac{a_-}{2a_+} \left[ \frac{a_-}{2a_+} \frac{6}{2}\Omega^2 \partial_t |\Omega| + \left( 2 \gamma - \frac{a_-}{2a_+} \frac{5}{2} \Gamma \right) \left( |\Omega|^4 - \Gamma M^{(2)} \right) \right] \\
& \quad + \left( \gamma - \frac{a_-}{2a_+} \Gamma \right) \left( M^{(1)} |\Omega| \partial_t |\Omega| + |\Omega|^4 - \Gamma |\Omega|^2 M^{(1)} \right) \\
& \quad + \left( \frac{2a_+}{a_-} \gamma^2 - 3 \Gamma \gamma + 2 \frac{a_-}{2a_+} \Gamma^2 \right) \left( |\Omega|^2 M^{(1)} - \Gamma N_0^{(2)} \right). \quad (3.34)
\]

into Eq. (3.17) to yield:

\[
\begin{align*}
\rho_{12}^{(5)} &= (-4)^2 \frac{u_3^{(0)}}{a_+^2} \frac{a_-}{2a_+} \left\{ (\Delta \omega + i \gamma) \left[ \frac{a_-}{2a_+} \frac{3}{2} \Omega^4 + \left( 2 \gamma - \frac{a_-}{2a_+} \frac{5}{2} \Gamma \right) M^{(2)} \Omega \right] \\
& \quad + \left( \gamma - \frac{a_-}{2a_+} \Gamma \right) \left[ \Omega^2 \Omega M^{(1)} + \left( \frac{2a_+}{a_-} \gamma^2 - 3 \Gamma \gamma + 2 \frac{a_-}{2a_+} \Gamma^2 \right) N_0^{(2)} \Omega \right] \right\} \\
& \quad + \frac{i a_- - 2 \Delta \omega \gamma}{a_+} \left[ \frac{a_-}{2a_+} \frac{15}{2} |\Omega|^4 \partial_t \Omega + \left( 3 \gamma - \frac{a_-}{2a_+} \frac{7}{2} \Gamma \right) |\Omega|^4 \right] \\
& \quad - \left( 2 \gamma - \frac{a_-}{2a_+} \frac{5}{2} \Gamma \right) \Gamma M^{(2)} \Omega - \left( \gamma - \frac{a_-}{2a_+} \Gamma \right) \left( |\Omega|^2 \Omega M^{(1)} \right) \\
& \quad + \left( \frac{2a_+}{a_-} \gamma^2 - 3 \Gamma \gamma + 2 \frac{a_-}{2a_+} \Gamma^2 \right) \left( |\Omega|^2 \Omega M^{(1)} - \Gamma N_0^{(2)} \Omega \right) \\
& \quad + \left( 2 \gamma - \frac{a_-}{2a_+} \frac{5}{2} \Gamma \right) M^{(2)} \partial_t \Omega \\
& \quad + 2 \left( \gamma - \frac{a_-}{2a_+} \Gamma \right) |\Omega|^2 M^{(1)} \partial_t \Omega + \left( \frac{2a_+}{a_-} \gamma^2 - 3 \Gamma \gamma + 2 \frac{a_-}{2a_+} \Gamma^2 \right) N_0^{(2)} \partial_t \Omega \right\}. \quad (3.35)
\end{align*}
\]

To describe non-linear response for Chap. 4 we are interested in the adiabatic terms. As for the third order those terms give the main contribution to the solution of the polarization. We refrain from describing every term in the lengthy expression, but show, as for the third order, the slow and
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the fast pumping limit of Eq. (3.35). In the slow pumping limit $\Gamma \to 0$ the polarization reads:

$$
\rho_{12}^{(5)} = (-4)^2 \frac{u_3^{(0)}}{a_3} \frac{a_-}{2a_+} \left\{ (\Delta \omega + i\gamma) \frac{a_-}{2a_+} \frac{3}{\Omega} |\Omega|^4 \Omega + \frac{\gamma}{a_+} \frac{2\Delta \omega \gamma}{3\gamma} |\Omega|^4 \Omega 
+ (\Delta \omega + i\gamma) \left[ 2\gamma M^{(2)} \Omega + \gamma|\Omega|^2 \Omega M^{(1)} + \left( \frac{2a_+}{a_-} \gamma^2 \right) N_0^{(2)} \Omega \right] 
+ \frac{\gamma}{a_+} \left( \frac{2a_+}{a_-} \gamma^2 \right) |\Omega|^2 \Omega M^{(1)} \right\}.
$$

(3.36)

Whereas in the fast pumping limit the memory terms follow again adiabatically $M^{(n)} = \Gamma^{-1} |\Omega|^{2n}$ as well the generalized memories $N^{(2)} = \Gamma^{-1} M^{(2)} = \Gamma^{-2} |\Omega|^4$.

$$
\rho_{12}^{(5)} = (-4)^2 \frac{u_3^{(0)}}{a_3} \left( \Delta \omega + i\gamma \right) \frac{\gamma^2}{\Gamma^2} |\Omega|^4 \Omega.
$$

(3.37)

For use in Chap. 4 we will use the adiabatic parts of Eq. (3.35), that includes the adiabatic part of the slow pumping limit, as well as relaxation rate contributions, but not the fast pumping limit. The real part reads:

$$
\text{Re}(\rho_{12}) = \Delta \omega \frac{u_3^\infty}{a_+} \left\{ 1 - 4 \left( \frac{a_-}{2a_+} \left( 1 + \frac{2\gamma}{a_+} \right) \frac{3}{a_+} - \frac{\gamma^2}{a_+} \right) |\Omega|^2 \Omega + 
+ (4)^2 \frac{a_-}{2a_+} \left[ \frac{a_-}{2a_+} \left( \frac{3}{2} + \frac{7}{a_+} \frac{\Gamma}{\gamma} \right) - 6 \frac{\gamma^2}{a_+} \right] |\Omega|^4 \Omega \right\},
$$

(3.38a)

whereas the imaginary part is:

$$
\text{Im}(\rho_{12}) = \gamma \frac{u_3^\infty}{a_+} \left\{ 1 - 4 \frac{a_-}{2a_+} \left( 3 - \frac{2\Gamma}{\gamma} \frac{a_-}{a_+} \right) \frac{|\Omega|^2}{a_+} + 
+ (4)^2 \left( \frac{a_-}{2a_+} \right)^2 \left[ \frac{15}{2} - 7 \frac{\Gamma}{\gamma} \frac{a_-}{a_+} \right] \frac{|\Omega|^4}{a_+} \right\} \Omega.
$$

(3.38b)
3.2.2 Saturable absorber – adiabatic Ansatz for the dissipative Bloch equations

Another possible treatment of the dissipative Bloch equations Eq. (3.15) is similar to the case presented in the last section, but uses the adiabatic following of the polarization and inversion right from the start, i.e. \((\partial_t \rho)/\rho \sim \Delta \omega \ll \gamma, \Gamma\). In this case the Bloch equations are in the quasi-steady state [AE87].

\[
0 \frac{\partial \rho}{\partial t} = -i \Delta \omega \rho_{12} + i \Omega u_3 - \gamma \rho_{12},
\]

\[
0 \frac{\partial u_3}{\partial t} = -4 \text{Im}(\Omega^* \rho_{12}) - \Gamma (u_3 - u_3^\infty).
\]

The steady state solution in dependence on the field \(\Omega\) are given by:

\[
\rho_{12} = \frac{\Omega u_3}{\Delta \omega - i \gamma} = (\Delta \omega + i \gamma) \frac{\Omega u_3}{\Delta \omega^2 + \gamma^2},
\]

\[
u_3 = u_3^\infty - \frac{4}{\Gamma} \text{Im}(\Omega \rho_{12}).
\]

Indeed Eq. (3.40a) resembles the zeroth order of the slowly varying envelope approximation of Eq. (3.17) and Eq. (3.40b) is the corresponding inversion in the fast pumping limit (cf. Eq. (3.16b)). That means the description from Sec. 3.2.1 is just between this regime and the conservative case. Those coupled equations can be consistently solved to yield

\[
u_3 = \left(1 + 4 \frac{\gamma}{\Gamma} \frac{|\Omega|^2}{\Delta \omega^2 + \gamma^2}\right)^{-1} u_3^\infty,
\]

\[
\rho_{12} = \frac{\Delta \omega + i \gamma}{\Delta \omega^2 + \gamma^2} \left(1 + 4 \frac{\gamma}{\Gamma} \frac{|\Omega|^2}{\Delta \omega^2 + \gamma^2}\right)^{-1} u_3^\infty \Omega.
\]

The behavior can be boiled down to two parameters

\[
a = \frac{\Delta \omega + i \gamma}{\Delta \omega^2 + \gamma^2} u_3^\infty\quad \text{and} \quad b = 4 \frac{\gamma}{\Gamma} \frac{1}{\Delta \omega^2 + \gamma^2}.
\]

\[
\frac{u_3}{u_3^\infty} = \frac{1}{1 + b|\Omega|^2}, \quad \rho_{12} = \frac{a}{1 + b|\Omega|^2} \Omega.
\]

The description is again adiabatically with intensity dependent coefficients. In contrast to Sec. 3.2.1 all orders of th field are contained. The polarization has a maximum at \(|\Omega| = \sqrt{\frac{a}{b} - 1}\). The inversion decays towards zero. This response describe a saturable absorber/gain depending on the sign of the steady state inversion. For small optical fields, \(|\Omega|^2 \ll b^{-1}\) the response is linear, whereas for higher fields it saturates and vanishes, cf. Fig. 3.8. We will use the non-linear response for wave equations in Sec. 4.3.3.
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\[ \frac{a}{1 + b|\Omega|^2} \approx a \sum_{k=0}^{\infty} (-b)^k |\Omega|^{2k} \]

\[ = u^3 \Delta \omega + i\gamma \sum_{k=0}^{\infty} \left( -4 \frac{\gamma}{\Gamma} \frac{1}{a_+} \right)^k |\Omega|^{2k}, \quad (3.44) \]

Figure 3.8: Left panel: behavior of the saturable absorber non-linearity as a function of the field amplitude. For small intensities the polarization grows linearly, until it decreases again after a saturation intensity. The inversion decreases with increasing field strength. Right panels: behavior of the quantities for a light pulse, the shaded areas represent the numerically exact solution. For too slow rates (upper panel) the exact solution is asymmetric, i.e. memory effects are important. For fast enough rates (middle panel) the saturable absorber treatment agrees with the exact solution: the polarization is excited for small fields and diminished for the larger parts of the pulse, for the decreasing flank of the pulse the situations is the same. For small pulse amplitudes, i.e. where saturation does not occur, polarization and inversion follow the pulse adiabatically.

**Polynomial limit**

In the regime without saturation, \( b|\Omega|^2 \ll 1 \), the solution can be treated via a Taylor expansion or geometric series, respectively, and becomes comparable to the perturbative treatment:
which contains the perturbative solutions in the fast pumping limit we presented in the last section, cf. Eqs. (3.30a) and (3.37). The value of the polarization and inversion is determined by the ratio of the dephasing $\gamma$ and relaxation $\Gamma$. For the assumption of vanishing pure dephasing this ratio is constant $\gamma/\Gamma = 2$, so we can now investigate the conservative limit of those equations, $b = \frac{2}{\Delta \omega^2}$.\footnote{This limit seems of course inconsistent since we assumed fast rates, however it shows the quality of the perturbative approach, which is a description between conservative and fast pumping limit.} The difference between the conservative solution Eq. (3.10) and the series in Eq. (3.44) in order $n$ is $b^n - \frac{1}{\Delta \omega^2 n} \frac{2n!}{(n!)^2} = \frac{1}{\Delta \omega^2 n} \left( 2^n - \frac{2n!}{(n!)^2} \right)$, which is small for low orders.

### 3.3 Conclusion

In this chapter we started with the Bloch equations with and without dephasing and relaxation rates. The Bloch equations are a coupled systems of differential equations with a non-linear dependence on the optical field. We showed derivations for the polarization in the different cases to use them in the following Chap. 4 as non-linearity. To summarize, we used the following limits for a closed description:

- The conservative limit (Sec. 3.1.2) for negligible dephasing and relaxation yielded a perturbation series of arbitrary order (Eqs. (3.10)).

- With dephasing and relaxation rates (Sec. 3.2.1) also a perturbative approach for slow fields was possible. Within this treatment there were two limits yielding an adiabatic polarization:
  - Slow pumping limit, with a negligible relaxation rate $\Gamma$
  - In the fast pumping limit, where we assumed a relaxation rate that is faster than the pulse length $\Gamma \gg \frac{\partial |\Omega|^2}{|\Omega|^2}$

In both limits for higher dephasing rates the behavior of the polarization became more adiabatic and thus the shown description more accurate.

- Saturable absorber treatment (Sec. 3.2.2) for fast relaxation rates (fast pumping limit) results in a non-perturbative expression, which can be expanded for small fields to result in the fast pumping limit of the perturbative approach.
Non-linear wave equations describing QD devices – The search for dissipative solitons

In the previous chapter the material response of QDs to pulsed excitation was studied. We progress in this chapter by including the back-action of the QD response to the external pulse and investigate how optical pulses can be shaped, when propagating through QD media. Therefore, the general wave equation for pulses in semiconductors is briefly derived in Sec. 4.2. In the next section the non-linear response from Chap. 3 is taken into account. This leads to well known non-linear equations, whose general features are discussed in Sec. 4.3. Further, those equations are known to support stable solutions, so called solitary waves and solitons, in the conservative as well as the dissipative regime. First, analytical solutions in the conservative regime are discussed, and finally, in Sec. 4.3.3 the formation of solitons in the dissipative regime is explored.

4.1 Introduction to non-linear wave equations and solitons

In this work we restricted ourselves to wave equations of the generalized Ginzburg-Landau type. Of course there are many other non-linear equations with soliton solutions and interesting properties, we want to review briefly in this section.

Historically, the soliton story begins in 1834, when John Scott Russell observed a single water wave, later called a solitary wave, that was constant in shape and velocity over a long distance in an English channel. The theoretical description of the phenomenon followed in 1895 by Korteweg and
De Vries [KV95] describing the small elevation $\eta$ on the plane water surface with the depth $l$:

$$\frac{\partial \eta}{\partial t} = \frac{3}{2} \sqrt{\frac{g}{l}} \frac{\partial}{\partial x} \left( \frac{1}{2} \eta^2 + \frac{2}{3} \alpha \eta + \frac{1}{3} \sigma \frac{\partial^2 \eta}{\partial x^2} \right). \tag{4.1}$$

with $\alpha$ being an arbitrary parameter and $\sigma = \frac{4}{3} l^2 - \frac{T l}{\rho g}$ depending on the water depth as well as the surface tension $T$ and the density $\rho$. They show the solitary wave solution

$$\eta = h \text{sech}^2 \left( x \sqrt{\frac{h}{4\sigma}} \right).$$

Nowadays the Korteweg-deVries (KdV) equation is probably the most known non-linear wave equation. The term soliton was coined in 1965 by Zabusky and Kruskal, when they numerically solved the KdV equation to describe a collisionless plasma and found the solitons were left unchanged after collisions with each other [ZK65]. The next big event in the history of solitons was the inverse scattering theory by Gardner et al in 1967 [GGK+67], which provides a general method to analytically derive the fundamental and higher order solutions of the KdV equation. Higher order solutions can be interpreted as multiple solitons scattering with each other. The inverse scattering method can be also used to obtain soliton solutions of various non-linear wave equations as the non-linear Schrödinger equation (NLSE) (which is featured in Sec. 4.3.2) as well as the sine-Gordon equation which we will shortly discuss below.

The NLSE plays a major role for the description of optical pulses in a fiber [Has90; Mit10]. Here, the non-linear response, also referred to as the intensity dependent refractive index, modulates the phase of an optical pulse. As predicted by Hasegawa and Tappert in 1973 this self-phase modulation in combination with the dispersion in fibers can lead to the existence of solitons.

The last well known non-linear equation, which was also found to have soliton solutions in this section is the sine-Gordon equation. This equation is especially noteworthy for the description of the resonant response from discrete level systems, as QDs:

In Sec. 3.1.1 we showed, that the resonant response of QDs to an optical field is the sine of the pulse area, which leads just to the sine-Gordon equation:

$$\frac{\partial}{\partial z} \left( \frac{k_z'}{k_z} \frac{\partial}{\partial t} \right) E = \frac{d_{21} \omega_i^2}{2 k_z \epsilon_0 c^2} \sin \left( \frac{d_{12}}{\hbar} \int_{-\infty}^{\tau} dt' E(t') \right), \quad \xi = z - k_z^{-1} t; \quad \tau = t \tag{4.2}$$

$$\Rightarrow \frac{\partial}{\partial \xi} \frac{\partial}{\partial \tau} \Theta = \left| \frac{d_{21}}{4 \hbar \epsilon_0 c} \right|^2 \sin \Theta. \tag{4.3}$$

This is a conservative wave equation for the pulse area. To find the fundamental soliton solution the coordinate frame $\xi' = z - v_{\text{soliton}} t; \quad \tau = t$ is useful. Here, the velocity of the soliton solution enters as a free parameter. Those coordinates yield the equation

$$\frac{\partial^2}{\partial \xi^2} \Theta = \left( \frac{1}{v_{\text{soliton}}} - k_z' \right)^{-1} \left| \frac{d_{21}}{4 \hbar \epsilon_0 c} \right|^2 \sin \Theta. \tag{4.4}$$
CHAPTER 4. THE SEARCH FOR DISSIPATIVE SOLITONS

This can be easily solved to yield:

\[ \Theta = 4 \tan^{-1} e^{\tau/T} \Rightarrow E = \frac{1}{T} \sech \left( \frac{t - z/v_{\text{soliton}}}{T} \right) \]  

(4.5)

This is a soliton solution where the electric field is a hyperbolic secant with a pulse area of \( 2\pi \). As in the NLSE case the height and the width of the pulse are inversely proportional. In the sine-Gordon case additionally the relative velocity \( v_{\text{soliton}} = k_{\gamma}^{-1} \left( 1 + \frac{k_{\gamma}^4 |d_{\alpha}|^2}{4|\sigma|\epsilon_0 c} \right) \) depends on the width of the pulse. Short pulses have a slow velocity whereas for long pulses the velocity approaches the group velocity. The phenomenon is called self induced transparency and was also investigated on the influence of relaxation and dephasing rates on the propagation [MH69]. The sine-Gordon equation is well studied and a description of the fundamental and higher order solutions can be found in [Man10]. In Ref. [ACM97] a generalized sine-Gordon equation with dissipation is investigated for different properties in the conservative and the dissipative regime and the transition between the regimes.

The soliton concept can be generalized to dissipative systems, i.e. open systems that allow an energy exchange with the environment [AA05a; AA08a]. The field of dissipative solitons covers the description physical, chemical and biological systems. One category, interesting in the semiconductor optics context, but not covered by this thesis, are cavity solitons [Lug03]. Here, the active material inside a cavity is pumped and has to offer to bistable operation, i.e. two different lasing intensities. The solitons are then spatial spots with a higher intensity on a darker background. The formation of those solitons is a two dimensional spatial problem, the velocity can be influenced by external fields and they offer a rich interaction behavior [BMP+07; GSS+02; PGA+08; PBM+05].

4.2 Wave equation – slowly varying envelope approximation

To describe the propagation of optical pulses a wave equation for a full electric field \( \vec{E} \) was derived from Maxwell’s equations in Sec. 1.2.3. To obtain a wave equation for optical pulses with a slowly varying envelope, we start with the typical wave equation Eq. (1.32):

\[ \Delta \vec{E} - \frac{1}{c^2} \frac{d^2}{dt^2} \vec{E} = \mu_0 \frac{d^2}{dt^2} \vec{P}, \]

with the macroscopic polarization \( \vec{P} \). As stated in Sec. 1.2.3 the polarization connects the results from Chap. 3 to the propagation of electric fields since it is the sum of all microscopic polarizations, which form the response of the QD medium to the external field. Both, the electric field \( \vec{E} \) and polarization \( \vec{P} \) are divided into a slowly varying envelope \( \vec{E}, \vec{P} \) and a fast wave part with the carrier frequency of the light \( \omega_l \) and a corresponding wave vector \( \vec{k} \).

\[ \vec{E}(t) = \vec{E}(t)e^{-i \vec{k} \cdot \vec{r} + i \omega_l t} + c.c., \quad \vec{P}(t) = \vec{P}(t)e^{-i \vec{k} \cdot \vec{r} + i \omega_l t} + c.c., \]  

(4.6a)

\[ \vec{E}(\omega) = \vec{E}(\omega - \omega_l)e^{-i \vec{k} \cdot \vec{r}} + \vec{E}^*(\omega + \omega_l)e^{i \vec{k} \cdot \vec{r}}, \quad \vec{P}(\omega) = \vec{P}(\omega - \omega_l)e^{-i \vec{k} \cdot \vec{r}} + \vec{P}^*(\omega + \omega_l)e^{i \vec{k} \cdot \vec{r}}. \]  

(4.6b)
4.2. WAVE EQUATION – SLOWLY VARYING ENVELOPE APPROXIMATION

The slow envelopes \( \vec{E}, \vec{P} \) are complex vectors. In this work we will restrict ourselves to one-dimensional propagation in \( z \) direction, hence we define \( \vec{k} = k_z \vec{e}_z \) without loss of generality.

The wave equation of the envelope \( E \) can be written in frequency domain\(^1\):

\[
\left( \Delta_{x,y} + \partial_z^2 - 2i k_z \partial_z - k_z^2 + \frac{\omega^2}{c^2} \right) \vec{E}(\Delta \omega) = -\mu_0 \omega^2 \vec{P}(\Delta \omega), \quad \Delta \omega = \omega - \omega_l. \tag{4.7}
\]

The macroscopic polarization is now split into the part of the passive background medium as well as the active QD medium \( \vec{P} = \vec{P}_{\text{bg}} + \vec{P}_{\text{QD}} \). Where \( \vec{P}_{\text{bg}} \) is assumed to be a linear response: \( \vec{P}_{\text{bg}}(\omega) = \epsilon_0 \chi(\omega) \vec{E}(\omega) \).\(^2\) As already described in Sec 2.1 \( \chi \) is a complex number with the real part being the index of refraction and the imaginary part corresponding to the absorption.

\[
\left( \Delta_{x,y} + \partial_z^2 - 2i k_z \partial_z - k_z^2 + \frac{\omega^2}{c^2} \left( 1 + \text{Re}(\chi(\omega)) \right) \right) \vec{E}(\Delta \omega) = -\mu_0 \omega^2 \vec{P}_{\text{QD}}(\Delta \omega) - i \frac{\omega^2 \text{Im}(\chi)}{c^2} \vec{E}. \tag{4.8}
\]

The term \( K(\omega) = \frac{\omega}{c} \sqrt{1 + \text{Re}(\chi(\omega))} \) represents the dispersion of the background material (cf. Sec 2.1). The envelope of a propagating pulse is influenced by the difference of \( k_z \) from the ansatz in Eq. 4.6a can be any function of \( \omega_l \) (with the constraint of obeying boundary conditions). Here, we have the freedom to choose the pulse’s dispersion relation, which affects the solutions for envelopes. We choose \( k_z \) to be the materials dispersion at the frequency of the light \( k_z = K(\omega_l) \) (Compare to Eq. (2.1) with vanishing \( \text{Im}(\chi) \)) and expand the materials dispersion \( K(\omega) \) around \( \omega_l\):

\[
K(\omega) \approx k_z + k_z' \Delta \omega + \frac{1}{2} k_z'' \Delta \omega^2.
\]

This ansatz is inserted into Eq. (4.8) and higher orders than \( \Delta \omega^2 \) are neglected. Thus, a wave equation is obtained, where the dispersion of the background medium is included in the derivative, while the active medium enters as a non-linear source term.

\[
\left( \Delta_{x,y} + \partial_z^2 - 2i k_z \partial_z + 2 k_z \frac{\partial k_z}{\partial \omega_l} \Delta \omega + \left( \frac{\partial k_z}{\partial \omega_l} \right)^2 \Delta \omega^2 + k_z \frac{\partial^2 k_z}{\partial \omega_l^2} \Delta \omega \right) \vec{E}(\Delta \omega) = -\mu_0 \omega^2 \vec{P}_{\text{QD}}(\Delta \omega) - i \frac{\omega^2 \text{Im}(\chi)}{c^2} \vec{E}(\Delta \omega). \tag{4.9}
\]

Next, we divide the whole equation by \( 2k_z \),\(^3\) and assume the term \( \frac{1}{2k_z} \left( \frac{\partial k_z}{\partial \omega_l} \right)^2 \) can be neglected.\(^4\)

\(^1\)This equation only represents the part proportional to \( e^{-i k \cdot r} \), since it is linearly independent of \( e^{i k \cdot r} \).

\(^2\)Note that for the Fourier transforms of the envelopes this means, that although the quantities are functions of \( \Delta \omega \) due to the rotating frame, the connecting susceptibility is still a function of \( \omega \), i.e. \( \vec{P}_{\text{bg}}(\Delta \omega) = \epsilon_0 \chi(\omega) \vec{E}(\Delta \omega) \), because it is defined for the full quantities, that are linear superpositions of the envelopes shifted by \( \pm \omega_l \), cf. Eq. (4.6b).

\(^3\)which is of course only possible for \( k_z \neq 0 \), which is reasonable for semiconductors below the band gap, cf. Fig. 2.1.

\(^4\)Alternatively one can define \( k'' = \frac{1}{k_z} \partial_{\omega_l} (k_z \partial_{\omega_l} k_z) \) instead of \( k'' = \partial_{\omega_l}^2 k_z \) to arrive at the same wave equation.
Finally, the back transformation to the time domain is made in relation to \( \Delta \omega \) which yields the wave equation for the pulse envelope

\[
\left( \frac{1}{2k_z} \Delta_{x,y} + \frac{1}{2k_z} \partial_z^2 - \frac{i}{2} k' \partial_t \right) \vec{E} = -\frac{\omega_0^2}{2k_z c^2 \epsilon_0} \vec{P}_{QD} - i\alpha E,
\]

(4.10)

with the absorption coefficient \( \alpha = \frac{\omega l}{2c \sqrt{1 + \text{Re}(\chi)}} \).

Next, the “Slowly varying envelope” approximation is applied by neglecting \( \partial_z E \ll k_z E \) and thus the second derivative after \( z \).

The spatial behavior perpendicular to the propagating direction can be calculated for specific geometries of wave guides or cavities. For the phenomenon of cavity solitons the Laplacian \( \Delta_{x,y} \) is important. However, as we constrict ourselves to a one dimensional problem it can be neglected.

This wave equation is typically written in the co-moving frame \( \tau = t - k' z, \xi = z \), which means the observer at some point \( z \) takes into account the group velocity \( k' - \frac{1}{2} \) for the time dependence of the propagating pulse. Then the derivatives in those new coordinates are \( \partial_\xi = \partial_z + k' \partial_t \) and \( \partial_\tau = \partial_t \), which leads to the typical partial differential equation with a first order derivative in the propagation coordinate \( \xi \) and a second order derivative in the progression coordinate \( \tau \):

\[
\left( i \partial_\xi - \frac{1}{2} k'' \partial_\tau^2 \right) \vec{E} = -\frac{\omega_0^2}{2c^2 \epsilon_0} \vec{P}_{QD} - i\alpha E.
\]

(4.11)

### 4.2.1 Pulse broadening due to group velocity dispersion

A prominent term in Eq. (4.11) is the group velocity dispersion (GVD) \( k'' \) which leads to a broadening of the optical pulse due to different phase velocities of the frequency components of the pulse. The regime where \( k'' > 0 \) is called normal dispersion, while \( k'' < 0 \) is referred as anomalous dispersion [Has90]. The pulse broadening effect of the GVD can be seen with from the wave equation without a non-linear response:

\[
0 = i \frac{\partial E}{\partial \xi} - \frac{k''}{2} \frac{\partial^2 E}{\partial \tau^2}.
\]

(4.12)

This equation can be formally solved in frequency domain for known initial conditions via

\[
E(\xi, \omega) = E(0, \omega) e^{-\frac{1}{2} k'' \omega^2 \xi},
\]

(4.13)

\[
E(\xi, \tau) = \frac{1}{2\pi} \int d\omega E(\xi, \omega) e^{i\omega \tau}.
\]

(4.14)

In particular, the effect of broadening due to GVD can be nicely shown on a Gaussian pulse since it stays Gaussian under Fourier transform and Eq. (4.13) is also of Gaussian shape. Since the product

\[\text{ Sometimes the refractive index } n(\omega) = \frac{c}{\omega} n(\omega) \text{ is used to describe those regimes with } \partial_\omega n(\omega) > 0 \text{ being the normal dispersion. Both descriptions are related via } k'' = \frac{1}{2} \left( 2 \partial_\omega n + \omega^2 \partial_\omega^2 n \right).\]
of two Gaussian pulses results in a Gaussian, the broadening is fully analytically accessible. We start with a Gaussian pulse with an inverse width of $\gamma_0$ and a chirp $\kappa_0$:

$$E(0, \tau) = e^{-(\gamma_0 - i\kappa_0)\tau^2},$$  

$$E(0, \omega) = \sqrt{\pi} (\gamma_0 - i\kappa_0)^{-\frac{1}{2}} e^{-\frac{1}{4}(\gamma_0 - i\kappa_0)^{-1}\omega^2}. \tag{4.15}$$

The Fourier transform has an inverse width of $\hat{\gamma} = \frac{1}{4} \frac{\gamma_0}{\gamma_0^2 + \kappa_0^2}$ and a “chirp” of $\hat{\kappa} = -\frac{1}{4} \frac{\gamma_0 \kappa_0}{\gamma_0^2 + \kappa_0^2}$. A chirp does not affect the width in time, but broadens the width in frequency domain. After the propagation of a length $\xi$ the pulse is still Gaussian, but with different properties due to the additionally introduced “chirp” $\frac{k''}{2} \xi$ from Eq. (4.13).

$$E(\xi, \omega) = \sqrt{\pi} (\gamma_0 - i\kappa_0)^{-\frac{1}{2}} e^{-\frac{1}{4}(\gamma_0 - i\kappa_0)^{-1}\omega^2} e^{-\frac{1}{2}k''\omega^2\xi}, \tag{4.17}$$

$$E(\xi, \tau) = (1 + 2i (\gamma_0 - i\kappa_0) k''\xi)^{-1/2} e^{-\frac{1}{2} \left(\frac{\gamma_0^2}{\gamma_0^2 + \kappa_0^2} + \kappa_0^2\right) \gamma_0^2 \tau^2} e^{-i \frac{1}{2} \left(\frac{\gamma_0^2}{\gamma_0^2 + \kappa_0^2} + \kappa_0^2\right) k'' \xi^2} \tag{4.18}.$$

In the frequency domain the inverse width is still $\hat{\gamma} = \frac{1}{4} \frac{\gamma_0}{\gamma_0^2 + \kappa_0^2}$, but $\hat{\kappa}$ is now modified to $-\frac{1}{4} \frac{\gamma_0 \kappa_0}{\gamma_0^2 + \kappa_0^2} + \frac{k''}{2} \xi$, which yields a new width $\gamma(\xi)$ in time domain depending on the propagated distance:

$$\frac{\gamma_0}{\gamma(\xi)} = 1 + 2|k''L_D\kappa_0|((1 - \xi/L_D)^2 - 1); \quad L_D = -\frac{\kappa_0}{2\left(\frac{\gamma_0^2}{\gamma_0^2 + \kappa_0^2}\right) k''}. \tag{4.19}$$

The distance dependent chirp, i.e. the imaginary part of the exponential, of the pulse reads:

$$\kappa(\xi) = \kappa_0 \frac{(1 - \xi/L_D)(\gamma_0^2 + \kappa_0^2)}{\gamma_0^2 + \kappa_0^2 (1 - \xi/L_D)^2}. \tag{4.20}$$

With the dispersion length $L_D$ [Kno11], which is the maximal range for a possible compression of a pulse, in the case $\kappa_0 < 0$ (down-chirp) for normal dispersion $k'' > 0$ (or in the opposite case with up-chirp and anomalous dispersion). After $L_D$ the “chirp” in the frequency domain $\hat{\kappa}$ vanishes. In this case a pulse recovers its starting width after a distance of $2L_D$, because the absolute value of the chirp is restored (cf. 4.1). Without a starting chirp the broadening of a Gaussian pulse after a distance $\xi$ reads:

$$\frac{\gamma_0}{\gamma(\xi)} = 1 + (2\gamma_0 k'' \xi)^2 \tag{4.21}$$

The broadening depends on the square of the initial width and the distance traveled, hence group velocity dispersion is especially important for ultra short pulses and long distances.

---

6 A chirp is the time dependent frequency of a pulse and briefly discussed in Sec. 1.2.3.

7 In a more technical context, the term dispersion length is reserved for the distance until a non-chirped pulse has broadened by a factor of $\sqrt{2}$ [Mit10], namely $L_D' = 2\gamma_0 k''$ for a Gaussian.
4.3 Generalized complex Ginzburg-Landau type equations

The wave equation Eq. (4.11) derived in the last section has the structure of a so-called generalized complex Ginzburg-Landau equation. In Sec. 4.3.1 we briefly comment on some general properties, then the non-linearities derived from the semiconductor QDs in Chap. 3 are introduced and the resulting wave equations are discussed. In Sec. 4.3.2 conservative wave equations in general and especially the non-linear Schrödinger equation are treated. Section 4.3.3 addresses dissipative wave equations, especially the complex Ginzburg-Landau equation and the saturable absorber equation. The transition from dissipative to the conservative regime is presented in Sec. 4.3.4.

4.3.1 General properties

The generalized Ginzburg-Landau equation reads [Ros05]:

\[
0 = i \frac{\partial E}{\partial \xi} - k'' \frac{\partial^2 E}{\partial \tau^2} + f(|E|^2)E
\] (4.22)

with the non-linearity \(f(|E|^2)E\) which relates to the polarization of the QDs (active material) presented in Chap. 3. This implies restrictions to the cases that can be described with this wave equation:

i) the polarization has to vanish for vanishing fields, i.e. we cannot describe cases with significant memory or dependence on the pulse area or the derivative of a pulse. The description can be used for adiabatic following hence for non-resonant interaction.

ii) \(f\) shall only depend on the absolute value of the field, which is the case for the QD dynamics discussed in Chap. 3, due to the rotating wave approximation and no permanent dipoles.

Now we want to show some general properties of solutions to Eq. (4.22). First, we consider a formal solution \(E_0(\tau, \xi)\) to show its invariances.

It is easy to see that Eq. (4.22) is invariant to translations and constant phases, i.e. if \(E_0(\tau, \xi)\) is a solution of Eq. (4.22) then \(E(\tau, \xi) = E_0(\tau - \tau_0, \xi)e^{i\phi_0}\) is also a solution.

Galilean invariance

The next general invariance we want to discuss is the one to constant velocities within the co-moving frame. Inserting the ansatz \(E(\tau, \xi) = E_0(\tau - A\xi, \xi)e^{iB'\xi + iC'\tau}\) into Eq. (4.22) leads to

\[-iA\partial_\tau E_0 + i\partial_\xi E_0 - B'E_0 - \frac{k''}{2} \left( \frac{\partial^2 E_0}{\partial \tau^2} + 2iC'\partial_\tau E_0 - C'^2 E_0 \right) = -f(|E_0|^2)E_0, \] (4.23)

which leaves the following equation for the parameters of the moving solution:

\[-i \left( A + k''C' \right) \frac{\partial E_0}{\partial \tau} - \left( B' - \frac{k''}{2} C'^2 \right) E_0 = 0. \] (4.24)
4.3. GENERALIZED COMPLEX GINZBURG-LANDAU TYPE EQUATIONS

The latter equation is fulfilled for $C' = -\frac{A}{k^2}$ and $B' = \frac{k'^2}{2} - 2\frac{A^2}{k^3}$. A pulse with a velocity $A$ relative to the group velocity and a phase linear in $\tau$ and $\xi E(\tau, \xi) = E_0(\tau - A\xi, \xi)e^{i\frac{A^2}{2(k^2)} - 2\frac{A^2}{k^3}\tau}$ is also a solution of the generalized complex Ginzburg-Landau equation. This means if we want to find a solution it is only necessary to find the solution with the relative velocity $A = 0$ to know a whole family of solutions. We also see, that the starting condition $E(\tau, 0) = E_0(\tau, 0)e^{-i\frac{2A^2}{k^2}\tau}$ which is a pulse with a center frequency other than $\omega_l$, leads to a velocity differing from the group velocity. Hence, two pulses with a different detuning will have a mutual distance depending on the distance they traveled.

Scaling: normalized equations

A common technique to discuss wave equations is to switch into a normalized description [Has90]. $\xi$ and $\tau$ coordinate can be rescaled $\xi' = \frac{1}{\lambda}\xi$, $\tau' = \frac{1}{\sqrt{k''}}\tau$ thus the non-linearity is rescaled and an equivalence class of non-linearities can be described via the same equation

$$i\frac{\partial}{\partial \xi'} E - \frac{1}{2}\frac{\partial^2}{\partial \tau'^2} E = -\lambda f(|E|^2)E.$$ (4.25)

Which yields for a field $\psi \sim E$ with $f(|\psi|^2) = \lambda f(|E|^2)$ the same solutions in the scaled system. This works well in a limited number of non-linearities like the non-linear Schrödinger equation. Normalized equations typically normalize the leading term of $f$ to 1. Correspondingly the problem is described by one parameter less than in the unnormalized description. Thus, the parameter space is reduced. In Sec. 4.3.3 we present the microscopically derived non-linearities and show the respective normalized equations.

Energy flux

From Eq. (4.22) follows the continuity equation for the pulse energy [AA05b]

$$\partial_\xi |E|^2 + \partial_\tau \frac{ik''}{2} (E^* \partial_\tau E - E \partial_\tau E^*) = -\text{Im} f(|E|^2) |E|^2. \quad (4.25)$$

This introduces the energy flux $j = \frac{ik''}{2} (E^* \partial_\tau E - E \partial_\tau E^*)$ as well as the energy generating density $P_{\text{gen}} = -\text{Im} f(|E|^2) |E|^2$. The energy flux is a measure for the deformation of a pulse. A positive (negative) sign means parts of the pulse are transported towards the negative (positive) direction of the $\tau$ axis. That is for a Gaussian pulse exposed to group velocity dispersion (cf. Eq. (4.18)):

$$j(\tau, \xi) = -2|E|^2 k'' \left( \frac{\gamma_0^2 + \kappa_0^2}{\gamma_0^2 + \kappa_0^2 (1 - \xi/L_D)} \right) \kappa_0 \left( 1 - \xi/L_D \right)^2 \tau = -2|E|^2 \kappa(\xi). \quad (4.26)$$

Figure 4.1 shows the energy flux of a Gaussian pulse with GVD for various distances. For $\xi < L_D$ the pulse’s energy is transported towards the center, leading to a narrowing of the pulse, whereas for $\xi > L_D$ the energy is transported to the wings, which broadens the pulse. As we can see, the
energy generation is depending on the intensity, while the flux is sensitive to the phase of a pulse. For the case with an unchirped Gaussian pulse the energy flux always broadens the pulse:

$$ j(\tau, \xi) = -|E|^2 \frac{\xi}{4(\sigma^2)^2} + \xi^2 \tau. $$

(4.27)

In the regime of dissipative wave equations, $P_{\text{gen}}$ is a non-vanishing function of the intensity, and an important quantity: for stable solutions it has to be in equilibrium with the energy flux $j$. We will discuss this further in Sec. 4.3.3.

**Dispersion and Dissipation**

Since $E$ is a complex field it is convenient to decompose it into its absolute value and its phase $E = q e^{i \phi'}$ which leads to equations for the real and imaginary part of Eq. (4.22) (restricting ourselves to $k'' \in \mathbb{R}$).
4.3. GENERALIZED COMPLEX GINZBURG-LANDAU TYPE EQUATIONS

Inserting the ansatz into Eq. (4.22), the real part of the equation corresponds to the dispersion:

\[ 0 = -q \frac{\partial \varphi}{\partial \xi} - \frac{k''}{2} \left[ \frac{\partial^2 q}{\partial \tau^2} - q \left( \frac{\partial \varphi}{\partial \tau} \right)^2 \right] + \text{Re} f(q^2)q. \]  

(4.28a)

It is mainly the propagation equation for the phase and is driven by the real part of the polarization. The imaginary part directly includes the propagation of the absolute value and contains the imaginary part of the polarization, which we already saw corresponding to absorption/luminescence in Sec. 2.1. Thus the imaginary part of Eq. (4.22) resembles the continuity equation (4.25):

\[ 0 = \frac{\partial q}{\partial \xi} - \frac{k''}{2} \left[ 2 \frac{\partial \varphi}{\partial \tau} \frac{\partial q}{\partial \tau} + q \frac{\partial^2 \varphi}{\partial \tau^2} \right] + \text{Im} f(q^2)q. \]  

(4.28b)

Both equations are coupled via the dispersion. In the next sections we show some conservative and dissipative non-linearities \( f(q^2) \) and solutions to the corresponding wave equations.

4.3.2 Conservative cases

In the conservative regime there is no energy consumption or generation, i.e. \( \text{Im}(f(q^2)) = 0 \). We define \( f_R = \text{Re} f \). In this section we want to derive the most simple localized solutions \( q(\tau) \), the so-called fundamental solutions, which have one maximum. Higher order solutions that are able to describe situations with multiple pulses and where solitons can collide etc. are not topic of this thesis and can be learned from the literature [NML+84; Has90; Man10].

General approach to solitary solutions

There are many approaches to find the fundamental solitary solution for different specific non-linear equations, that work with similar schemes [KV95; Has90; Kno11]. Here, we show the generalized procedure for an arbitrary conservative non-linearity \( f(q^2) \) and apply it on the 3-5 non-linear Schrödinger equation (NLSE). We search for the solitary pulse solution \( E_0(\tau, \xi) = q(\tau)e^{i\varphi(\tau, \xi)} \), i.e. a pulse that does not change its shape during propagation \( \partial_\xi q = 0 \). According to Sec. 4.3.1 we can derive any solution with a constant velocity from this solution. In the conservative case Eqs. (4.28) read

\[ 0 = -q \frac{\partial \varphi}{\partial \xi} - \frac{k''}{2} \left[ \frac{\partial^2 q}{\partial \tau^2} - q \left( \frac{\partial \varphi}{\partial \tau} \right)^2 \right] + f_R(q^2)q, \]  

(4.29a)

\[ 0 = -\frac{k''}{2} q^{-1} \frac{\partial}{\partial \tau} \left( q^2 \partial_\tau \varphi \right) \Leftrightarrow (\partial_\tau \varphi = 0 \wedge q \neq 0). \]  

(4.29b)

From the second equation we can see, that the phase has to be constant in time \( \varphi(\xi) \) for a non-moving\(^8\) solitary pulse. Specifically, this means there is no energy flux for a stable conservative

\(^8\)Remember that non-moving means stationary in \( \tau \) within the co-moving frame, i.e. in the \( t, z \) coordinate system this is a pulse, that moves with the group velocity \( k'^{-1} \).
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solution. The first equation yields the pulse shape, with the definition \( C_0 = -2/k'' \partial_\xi \psi \) and multiplication with \( \partial_\tau q \) it reads:

\[
0 = -\frac{k''}{4} \partial_\tau \left( \frac{\partial}{\partial \tau} q \right)^2 + \frac{k''}{4} C_0 \frac{\partial^2 q}{\partial \tau^2} + \frac{1}{2} \frac{\partial}{\partial \tau} F(q^2); \quad \partial_\tau F(x) = f(x),
\]

which can be integrated to yield:

\[
\left( \frac{\partial}{\partial \tau} q \right)^2 = C_0 q^2 + \frac{2}{k''} F(q^2) + C_1.
\]

For a pulsed solution the integration constant has to be \( C_1 = -\frac{2}{k''} F(0) \), for the boundary condition \( q = 0 \). This equation defines the maximum of \( q \) as a function of the free parameter \( C_0 \) connecting the phase \( \partial_\xi \psi \) and the amplitude \( q \) of the solution. That means a general solution of a conservative generalized Ginzburg-Landau equation we have to solve the integral for

\[
\pm \int_{q_0}^{q(\tau)} dq' \frac{1}{\sqrt{C_0 q'^2 - \frac{2}{k''} (F(q'^2) - F(0))}} = \int_{\tau_0}^\tau d\tau = \tau - \tau_0.
\]

The left side is a line integral where \( q \) is the curve, i.e. we search for the curve \( q(\tau) \) on which the integrand yields an integration linear in time (cf. Fig. 4.2). The sign of the integrals depends on the

\[ q_0 \]

\[ \tau_0 \]

\[ q_{\text{max}} \]

\[ 14 \]

\[ 12 \]

\[ 10 \]

\[ 8 \]

\[ 6 \]

\[ 4 \]

\[ 2 \]

\[ 0 \]

\[ -20 -15 -10 -5 0 5 10 15 20 \]

\[ -20 -15 -10 -5 0 5 10 15 20 \]

\[ \text{Time (norm. coordinates)} \]

\[ q \]

\[ \text{Figure 4.2: Left panel: Visualization of the approach described in the text. The color map represents the integrand of the left hand side of Eq. (4.32) (or specifically Eq. (4.34) for the solution of the 3-5NLSE). The solution is obtained as the line integral that results in a linear growth with the time coordinate } \tau. \text{ Right panel: Solutions to the 3-5NLSE (the color code only discriminates different solutions). There is a continuum of solutions for every initial height with a corresponding width.} \]

sign of \( \partial_\tau q \). Hence, four cases would have to be taken into account \( \tau \leq \tau_0 \) and \( q \leq q_0 \). Thus, it is convenient to choose \( q_0 = q(\tau_0) \) to be the maximum of the pulsed solution, given by:

\[
C_0 q_0^2 = \frac{2}{k''} (F(q_0^2) - F(0)).
\]

The solution is symmetric around \( \tau_0 \). Note that this is an equation for \( q_0^2 \) but not \( q_0 \), however only the positive solution \( q_0 \) is needed.

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4.3. GENERALIZED COMPLEX GINZBURG-LANDAU TYPE EQUATIONS

Application: cubic-quintic non-linear Schrödinger equation

In Sec. 3.1.2 we derived the perturbative response of the conservative Bloch equations (Eq. (3.2)). This resulted in a non-linear response that leads to the non-linear Schrödinger equation (NLSE).

\[ i \frac{\partial E}{\partial \xi} - \frac{k''}{2} \frac{\partial^2 E}{\partial \tau^2} = -a|E|^2 E + b|E|^4 E. \quad (a, b \in \mathbb{R}) \] (4.33)

The NLSE is one of the best known conservative, non-linear wave equations. In particular it is famous for describing the propagation of pulses through optical fibers [Has90; Mit10]. Mostly the cubic NLSE is discussed in the literature. Here, we want to focus on its, also well known, generalization with an additional quintic term. The left side of Eq. (4.33) describes the broadening of a pulse due to GVD, while the (cubic) non-linearity on the right side, which is sometimes described as intensity dependent refractive index, affects the phase depending on the pulse amplitude \( \varphi = \varphi_0 + aq^2 \xi \). This is called self-phase modulation [Mit10] and is comparable to a chirp. The quintic non-linearity is a saturation for the cubic part. Solitary solutions are pulse shapes for which the GVD chirp and the chirp from the non-linearity compensate each other. To find the fundamental solution to Eq. (4.33) with the general method we introduced above this we have to assign the non-linearity \( f(q^2) \) with:

\[ f(x) = -ax + bx^2; \quad F(x) = -\frac{a}{2}x^2 + \frac{b}{3}x^3; \quad C_1 = 0, \]
\[ \frac{\partial}{\partial \tau} q = \pm q \sqrt{C_0 - Aq^2 + Bq^4}; \quad A = \frac{a}{k''}, \quad B = \frac{2b}{3k''}. \]

As described in Eq. (4.3.2), the maximum of the solution \( q_0 \) obeys \( C_0 = -Bq_0^4 + q_0^2 A \). The solution of the integral yields

\[ \tau - \tau_0 = \int_{q_0}^{q_1} \frac{dq}{q \sqrt{C_0 - Aq^2 + Bq^4}} \]
\[ = -\frac{1}{\sqrt{C_0}} \log \left( \frac{2\sqrt{C_0} \sqrt{-Aq^2 + Bq^4 + C_0} - Aq^2 + 2C_0}{-Aq^2 + 2q^2 C_0/q_0^2} \right). \] (4.34)

This equation has to be resolved after \( q \) as a function of \( \tau \), starting with exponentiating the whole expression.

\[ e^{-2\sqrt{C_0}(\tau-\tau_0)} = \frac{1}{q^2} \left( 2q_0 \sqrt{-Bq_0^4 + A\sqrt{-A(q^2 - q_0^2)} + B(q^4 - q_0^4) - Aq^2 + 2(Aq_0^2 - Bq_0^4)} \right) \]
\[ \frac{A - 2Bq_0^2}{A - 2Bq_0^2} \] (4.35)

Obviously, the left hand side of this expression is a monotonously falling function. Since we search for a symmetric, pulsed solution we add the rising flank \( e^{2\sqrt{C_0}(\tau-\tau_0)} \) and resolve the sum after \( q \).
This leads to
\[
\cosh \left( 2\sqrt{C_0} (\tau - \tau_0) \right) = \frac{(2Aq_0^2 - 2Bq_0^4 - Aq^2)}{A - 2Bq_0^2} \frac{1}{q^2}.
\] (4.36)

Finally, the fundamental soliton solution for the 3-5-NLSE can be obtained as
\[
q^2 = \frac{2q_0^2 \left( a - \frac{2}{3} bq_0^2 \right)}{a + \left( a - \frac{4}{3} bq_0^2 \right) \cosh \left( 2q_0 / \sqrt{k''} \sqrt{a - \frac{2}{3} bq_0^2} (\tau - \tau_0) \right)},
\] (4.37)
\[
\varphi = -\frac{1}{2} k'' C_0 z = -\left( a - \frac{2}{3} bq_0^2 \right) \frac{q_0^2}{2} \xi.
\] (4.38)

This is a continuous family of solutions, where every member has a maximum \(q_0\) and a corresponding width proportional to \(\frac{1}{q_0}\).

The number of fundamental solutions is even larger taking into account the phase, translational and Galilean invariance of the NLSE, which means the general fundamental solution:
\[
q(\xi, \tau) = \sqrt{\frac{2q_0^2 \left( a - \frac{2}{3} bq_0^2 \right)}{a + \left( a - \frac{4}{3} bq_0^2 \right) \cosh \left( 2q_0 / \sqrt{k''} \sqrt{a - \frac{2}{3} bq_0^2} (\tau - \tau_0) \right)}},
\] (4.39)
\[
\varphi(\xi, \tau) = \frac{v^2 \xi - 2v \tau}{2k''} + \varphi_0 - \left( a - \frac{2}{3} bq_0^2 \right) \frac{q_0^2}{2} \xi.
\] (4.39)

Note, that the more commonly used limit of the cubic NLSE is of course included for \(b \to 0\)
\[
q^2 = \frac{2q_0^2}{1 + \cosh \left( 2q_0 / \sqrt{k''} \sqrt{a} \right)} = q_0^2 \sech^2 \left( q_0 \sqrt{\frac{a}{k''}} \right).
\] (4.39)

### 4.3.3 Dissipative cases

When dissipative non-linearities occur in the wave equation, typically no analytical solution or closed form expression exists. However there are stable solutions which can be found numerically. In this subsection we want to cover solitary waves in the dissipative regime. They are called dissipative or auto solitons in analogy to the conservative case [AA05b; Ros05]. The most important difference is that there is no continuum of solutions, but discrete ones playing the role of fixed points for the dynamics. Additionally, there are also periodically changing solutions, corresponding to limit cycles [ASCT01; Akh98] depending on the parameter regime. Further, the scattering properties of dissipative solitons show a wealth of variants, e.g. the soliton-typical preservation of the pulses, but also annihilation [ACM97], or the formation of stable bound states [BMA00; VKR01].

To find stable solutions the continuity equation Eq. (4.28b) is of importance:
\[
0 = q\frac{\partial q}{\partial \xi} - k'' \frac{\partial}{\partial \tau} \left( q^2 \frac{\partial \varphi}{\partial \tau} \right) + \text{Im} f(q^2)q^2.
\]
4.3. GENERALIZED COMPLEX GINZBURG-LANDAU TYPE EQUATIONS

For stable pulsed solutions ($\partial_t q = 0$) the energy flux $j = k'' \left( q^2 \frac{\partial \psi}{\partial \tau} \right)$ has to balance the energy generation $P_{\text{gen}} = \text{Im} f(q^2)q^2$,\footnote{With the equilibrium condition $\partial_t j = P_{\text{gen}}$.} while the energy generation and drain has to cancel out over the whole regime, i.e. $\int_{-\infty}^{\infty} d\tau P_{\text{gen}}(\tau)$ has to vanish, this means it has to yield positive and negative areas of the same size [AA08b]. Thus it is convenient to investigate the roots of $\text{Im} f(q^2)q^2$, which are also homogeneous solutions (also known as continuous wave solution), i.e. $q(\tau) = \text{const.}$. Physically those points are where gain and absorption just compensate each other and gain changes to absorption with increasing intensity (stable fixed point) or the other way around (unstable fixed point). One root is always the solution $q = 0$ which should be stable for the existence of pulsed solutions.

The following sections will look in more detail on different special equations and their solutions, as well as regimes where they exist and how they can be numerically obtained.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4.3.png}
\caption{Schematic of a stable solution in the dissipative regime. The horizontal lines show the roots of the energy generating function $P_{\text{gen}}$. At the points where the intensity crosses those levels the response is alternated between gain and absorption. The areas between $P_{\text{gen}}$ and 0 visualize the generation/absorption of energy, for a stable solution the positive and negative areas have to be of the same size, while the energy flux has to balance energy generation and drain. The inset is a magnification of the graph for the temporal range it is shown.}
\end{figure}
CHAPTER 4. THE SEARCH FOR DISSIPATIVE SOLITONS

Complex Ginzburg-Landau equation

The complex Ginzburg-Landau equation is the big brother of the non-linear Schrödinger equation Eq. (4.33) from the last section. For the complex Ginzburg-Landau equation every real parameters of the NLSE become complex numbers, i.e. $a \rightarrow a + i\epsilon$ and $b \rightarrow b - i\mu$. The Ginzburg-Landau equation originates from the description of second order phase transitions in hydrodynamics and superconductivity and is nowadays one of the most investigated non-linear equations in physics [AK02]. With the non-linear response from Sec. 3.2.1 we also obtain a complex Ginzburg-Landau equation describing the propagation of an optical pulse through a medium consisting of two level systems. That is the parameters of the non-linearity are microscopically derived from the dissipative Bloch equations in relaxation time approximation under the non-resonant excitation with a weak and slow optical pulse.

From the last paragraph we learned, that in order to obtain stable solutions, the response of the system has to yield regimes of absorption and gain depending on the amplitude of the pulse. However, this behavior cannot be achieved by the adiabatic, non-linear response of one two level system since it shows either gain or absorption, independent on the intensity of the optical pulse. That is for a stable solution as shown in Fig. 4.3 we need the response of two two level systems as well as an additional linear absorption. QDs typically consist of more than one electronic transition. We take into account s shell and p shell excitation in a regime, where the s shell exhibits gain, while the p shell absorbs. It is our aim to find soliton solutions for QD parameters from Sec. 3.2.1.

$$i\frac{\partial E}{\partial \xi} - \frac{k''}{2} \frac{\partial^2 E}{\partial \tau^2} = -i\alpha E - (a + i\epsilon)|E|^2 E + (b - i\mu)|E|^4 E. \quad (4.40)$$

We can obtain this equation with the results from Sec. 3.1.2 with the parameters in Table 4.1. All QD contributions show the common prefactor $c_{pre} = \frac{\omega^2}{2\epsilon\epsilon_0} \frac{|d_{12}|^2 u_n}{\hbar \omega} \phi$ stemming from the wave equation (4.11) and the common prefactors of every order of the polarization.

Note that often in studies there is also a complex GVD $k'' \rightarrow k'' + i\beta$, with the spectral filtering $\beta$. Taking this into account can change a lot to the particular properties of the solutions. In this work, we will not discuss this parameter.

Normalized notation: Typically, non-linear equations are depicted in their normalized form, to make general statements (cf. Sec. 4.3.1). For the cGLE this means the amplitude is renormalized to let the renormalized $\tilde{a} = 1$ and $\tau$ to renormalize $k''$ to 1.

$$i\frac{\partial \psi}{\partial \xi'} - \frac{D}{2} \frac{\partial^2 \psi}{\partial \tau'^2} = -i\tilde{a}\psi - (1 + i\tilde{\epsilon})|\psi|^2 \psi + (\tilde{b} - i\tilde{\mu})|\psi|^4 \psi. \quad (4.41)$$

Homogeneous solutions of the cGLE: As noted before it is of advantage to know the homogeneous or continuous wave solutions $q(\tau) = \text{const.}$ of a dissipative wave equation since they mark the intensities, where gain and absorption alternate and thus define the energy generating density $P_{\text{gen}}$. According to Eq. (4.28b) a homogeneous solution follows:

$$0 = \partial_\xi q + \text{Im} f(q^2) q. \quad (4.42)$$
4.3. GENERALIZED COMPLEX GINZBURG-LANDAU TYPE EQUATIONS

Coefficients of the complex Ginzburg Landau equation derived with the perturbative approach in Table 4.1:

<table>
<thead>
<tr>
<th>Coefficient</th>
<th>adiabatic part (slow pumping limit)</th>
<th>fast pumping limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>linear absorption $\alpha$</td>
<td>$\gamma c_{\text{pre}}$</td>
<td>$\gamma c_{\text{pre}}$</td>
</tr>
<tr>
<td>Kerr coefficient $a$</td>
<td>$\Delta \omega \frac{4</td>
<td>d_{12}</td>
</tr>
<tr>
<td>Cubic gain $\epsilon$</td>
<td>$\gamma \frac{4</td>
<td>d_{12}</td>
</tr>
<tr>
<td>quintic phase $b$</td>
<td>$\Delta \omega \left( \frac{4</td>
<td>d_{12}</td>
</tr>
<tr>
<td>quintic absorption $\mu$</td>
<td>$\gamma \left( \frac{4</td>
<td>d_{12}</td>
</tr>
</tbody>
</table>

Table 4.1: Parameters of the complex Ginzburg Landau equation derived with the perturbative approach in Sec. 3.1.2. All coefficients have the common prefactor $c_{\text{pre}} = \frac{\omega_0^2}{2c^2 \epsilon_0} \frac{|d_{12}|^2}{\hbar} \frac{u_0^2}{a_+}$.

One steady state solution is $q_S = q_0 = 0$. For the non-trivial steady state homogeneous solutions $q_S$ of the cGLE one has to find the roots of

$$0 = \frac{1}{i} \text{Im} f(q_S^2) = \alpha + \epsilon q_S^2 + \mu q_S^4.$$  \hspace{1cm} (4.43)

Which yields to solutions for the square of the absolute value of the field:

$$q_S^2 = q_\pm^2 = -\frac{1}{2} \frac{\epsilon}{\mu} \left( 1 \pm \sqrt{1 - 4 \frac{\alpha \mu}{\epsilon^2}} \right).$$  \hspace{1cm} (4.44)

For the existence of such solutions the term has to be real and positive which means

- $\text{sgn}(\epsilon) = -\text{sgn}(\mu)$: a non-linear gain has to be compensated by a non-linear absorption, because at least in one case the term in brackets is positive.

- $0 < \frac{\alpha \mu}{\epsilon^2}$ for both solutions being positive. That means $\alpha$ and $\mu$ have the same sign. Absorption and gain alternate with the orders.

- Subsequently $4 \frac{\alpha \mu}{\epsilon^2} < 1 \iff \frac{2|\epsilon|}{|\alpha|} < \frac{|\epsilon|}{2|\mu|} \iff \frac{|\epsilon|}{2|\mu|} > \sqrt{\frac{2}{\mu}}$ has to be valid to yield real values. This implies for the coefficients if $|\alpha| > \frac{|\epsilon|}{2}$ then $|\mu| < \frac{|\epsilon|}{2}$ and vice versa, i.e. they have to be monotonous (compared to $\epsilon/2$)!

Depending on the parameters there are two non-trivial solutions or none, which is the typical behavior of a saddle-node-bifurcation.\footnote{The normal form of the saddle-node bifurcation reads $\partial_t x = r + x^2$, which also describes our case since Eq. (4.43) resembles this quadratic behavior for $q^2$.} The homogeneous solutions depending on the systems parameters are plotted in Fig. 4.4. The stability of the solutions can be observed for small perturbations around
the steady state solutions $q_S$: $q(\xi) = q_S + \delta(\xi)$, with the deviation $\delta \sim e^{-\lambda \xi}$ growing or vanishing during propagation. A solution is stable if those perturbations are damped out if the ansatz is inserted into Eq. (4.42), i.e. $0 < \lambda = \frac{1}{2} \text{Im} f \left((q_S + \delta)^2\right) (q_S + \delta)$. For the linear stability analysis the non-linearity is expanded linearly in $\delta$ around $q_S$.

$$\lambda \approx \frac{1}{\delta} \frac{\text{Im} f \left(q_S^2\right) q_S + \text{Im} f \left(q_S^2\right) + 2q_S^2 \text{Im} f' \left(q_S^2\right)}{=0^{11}}.$$ 

The zero solution is stable for $0 < \lambda = \text{Im} f(0) = \alpha$, i.e. for linear absorption ($\alpha > 0$). Consequently, for the existence of the two non-trivial homogeneous solutions the third order is non-linear gain ($\epsilon < 0$) and fifth order non-linear absorption ($\mu > 0$). The stability of the non-trivial solutions follows from $\lambda/\left((2q_\pm^2) = f'(q_\pm^2) = \epsilon + 2\mu q_\pm^2 > 0$, where inserting Eq. (4.44) yields:

$$0 > \mp \sqrt{1 - 4\frac{\alpha \mu}{\epsilon^2}}.$$ 

Thus $q_+$ is always stable, while $q_-$ is always unstable.

**Pulsed solutions** To find a pulsed solution, first of all a solution has to exist. Existence of various solutions with different properties is done elsewhere [ASCT01; SCAA+97; AASC96; AA08b; AA05b]. Secondly, to find a specific solution the initial conditions have to be in the proximity of it. For example, since the zero solution is a stable attractor, an initially too small pulse would rather evolve into the zero solution (i.e. vanish) than into a stable pulsed solution. Hence, to find reasonable initial conditions we take a close look at homogeneous solutions and thus on the energy generating density.

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11 The term vanishes for $q_S = 0$ as well as for $q_S = q_\pm$ since $\text{Im} f(q_\pm^2) = 0$. 

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Numerically we find solutions with a Fourier split step algorithm, where the discrete advancing step $\Delta \xi$ is done in the Fourier domain for the temporal derivative and in time domain for the non-linear part. As used for the introduction of the GVD (cf. Eq. (4.13)) the derivative part of the wave equation has the exact solution in Fourier domain. Hence, for the propagation from $\xi = \Delta \xi \times j$ to $\xi = \Delta \xi \times (j+1)$ the field reads:

$$
\tilde{E}(j+1, \omega) = \tilde{E}(j, \omega) e^{i k''(\omega^2 d\xi)},
$$

$$
\tilde{E}(j+1, \omega) = \tilde{E}(j, \omega) + \tilde{E}(j, \omega) \left( e^{i k''(\omega^2 d\xi)} - 1 \right).
$$

(4.45)

The non-linear part can be solved via the Euler or a Runge-Kutta method [PTV92; PTV+96]:

$$
E(j+1, \tau) = E(j, \tau) + \Delta \xi f(|E(j, \tau)|^2) E(j, \tau).
$$

(4.46)

Thus the combined propagated field reads

$$
E(j+1, \tau) = E(j, \tau) + \Delta \xi f(|E(j, \tau)|^2) E(j, \tau) + \mathcal{F}^{-1} \left( \mathcal{F} \left( E(j, \tau) \right) \left( e^{i k''(\omega^2 d\xi)} - 1 \right) \right).
$$

(4.47)

Figure 4.5 shows various initial conditions evolving into those two stable solutions. Initial conditions with intensities below the $E_-$ level are completely absorbed during propagation and strive towards the zero solution. For initial conditions with higher intensities, the propagation leads to a transformation into the same solution for all cases. The form of this solution depends on the system parameters. In the right panel of Fig. 4.5 the pulse energy as a function of the traveled distance is plotted. One can see that the energy of the stable solutions is approached. The fixed pulse energy is the necessary condition for the arrival at the stable solution.
CHAPTER 4. THE SEARCH FOR DISSIPATIVE SOLITONS

Saturable absorber non-linearity

In Sec. 3.2.2 we showed the fast pumping limit of the Bloch equations leading to the saturable absorber description of the response. As discussed before, to obtain stable solutions we need to take into account two two level systems: the s states of the QD that offer gain as well as the p states that are responsible for a non-linear absorption. This yields the wave equation that is intensely discussed in Ref. [Ros05]:

\[
\frac{1}{i} \frac{\partial E}{\partial \xi} - k'' \frac{\partial^2 E}{\partial \tau^2} = -\alpha' E - \frac{a}{1 + b_a|E|^2} E + \frac{g}{1 + b_g|E|^2} E. \tag{4.48}
\]

Analog to finding the solution to the cGLE we start by investigating the homogeneous solutions to Eq. (4.48):

\[
0 = \text{Im} f(q^2) = \alpha' + \frac{a}{1 + b_a q^2} - \frac{g}{1 + b_g q^2},
\]

\[
q^2_\pm = \left(\frac{(g - \alpha') b_a - (a + \alpha') b_g}{2 \alpha' b_a b_g}\right) \left(1 \pm \sqrt{4 \left(\frac{-\alpha' + g - a} {(g - \alpha') b_a - (a + \alpha') b_g} \right)^2 + 1}\right).
\]

Formally one can see an analogue to the homogeneous solutions of the cGLE:

\[
q^2_\pm = -\frac{1}{2} \frac{\epsilon}{\mu} \left(1 \pm \sqrt{1 - 4 \frac{\alpha' \epsilon}{\mu^2}}\right),
\]

\[
\alpha = \alpha' - g + a, \quad \epsilon = -(g b_a - a b_g) + (b_a + b_g) \alpha', \quad \mu = \alpha' b_a b_g.
\]

In the same way as for the cGLE there are parameter restrictions for the existence of real, positive solutions:

- \( \text{sgn} \left((g - \alpha') b_a - (a + \alpha') b_g\right) = \text{sgn}(\alpha') \),
- \((-\alpha' + g - a)\alpha' b_a b_g < 0\),
- \(0 < 4 \frac{(-\alpha' + g - a)\alpha' b_a b_g} {(g - \alpha') b_a - (a + \alpha') b_g} + 1\). \\

While the homogeneous solutions have the same qualitative behavior as those of the cGLE, the stability of those solutions is quite different. The stability of the zero solutions is ensured for \(0 < \alpha' + a - g = \alpha\). The non-trivial solutions are stable for \( f'(q^2) = -\frac{a b_a}{(1 + b_a q^2)^2} + \frac{g b_g}{(1 + b_g q^2)^2} > 0\):

\[
0 < \frac{\epsilon}{\alpha'} \left[\frac{\alpha'}{\epsilon} (b_a + b_g)(\alpha' - \alpha) + \alpha - \left(1 - \frac{\alpha'(b_a + b_g)}{\epsilon}\right) \frac{1}{2} \frac{\epsilon^2}{\mu}\right].
\]
Figure 4.6: Optical pulse propagating through QD material described in the saturable absorber (fast pumping) limit. The pulse excites the homogeneous solutions piecewise: initial conditions below $E_-$ lead to the zero solutions, whereas initial conditions above lead to the $E_+$ solution. The dispersion is weak, such instead of a pulse shape two fronts are created leading to a rectangular structure.

Table 4.2: Material parameters used in numerical simulations in the saturable absorber description

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N_{QD}$</td>
<td>$5 \times 10^{13}$ cm$^{-2}$</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>$4.00 \times 10^{-4}$ nm$^{-1}$</td>
</tr>
<tr>
<td>$k''_l$</td>
<td>$2.91 \times 10^{-3}$ fs$^2$/nm</td>
</tr>
<tr>
<td>$n_l$</td>
<td>$3.40$ nm$^{-1}$</td>
</tr>
<tr>
<td>$d_{s1}$</td>
<td>$0.440 \varepsilon_0$ nm</td>
</tr>
<tr>
<td>$d_{p1}$</td>
<td>$0.658 \varepsilon_0$ nm</td>
</tr>
<tr>
<td>$\Delta \omega_s$</td>
<td>$-9.12 \times 10^{-2}$ fs$^{-1}$</td>
</tr>
<tr>
<td>$\Delta \omega_p$</td>
<td>$6.08 \times 10^{-2}$ fs$^{-1}$</td>
</tr>
<tr>
<td>$\gamma_s$</td>
<td>$2.03$ ps$^{-1}$</td>
</tr>
<tr>
<td>$\Gamma_s$</td>
<td>$1.38$ ps$^{-1}$</td>
</tr>
<tr>
<td>$\gamma_p$</td>
<td>$3.03$ ps$^{-1}$</td>
</tr>
<tr>
<td>$\Gamma_p$</td>
<td>$1.43$ ps$^{-1}$</td>
</tr>
</tbody>
</table>

Pulsed solutions For QD parameters (cf. Tab. 4.2) we find that an initial pulse evolves into a rectangular structure [DPE+12], cf. Fig. 4.6. The reason for that is a small dispersion such that a piecewise evolution to the homogeneous fixed point levels occurs, i.e. parts of the pulse with amplitudes between $E_+$ and $E_-$ are enhanced up to $E_+$, while amplitudes below $E_-$ are absorbed. The resulting structure is no pulse solution in that sense, but two fronts. Thus, in contrast to pulsed solutions there is no defined width of the localized structure, but it depends on the initial conditions.

For example an initially Gaussian pulse $E(\tau) = E_0 e^{-\tau^2}$ would result in a rectangular structure with a width of $\Delta \tau = 2T \sqrt{2 \ln(E_0/E_-)}$. In conclusion, a QD waveguide that is pumped such that the s shell exhibits gain and the p shell exhibits absorption, can act as a device that transforms optical pulses into a solitary rectangular shape. Of course, we have to note that the rectangular solution is only an approximate solution which cannot occur in its full beauty in reality since the steady state limit of the Bloch equations breaks down for the fast change of the electric field. The

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12 A front is the border between two homogeneously excited regions and of special interest in pattern formation. Many equations with pulsed solutions also support front solutions.
formation of stable rectangular pulses could be of importance in data transmission, where a here
presented amplifier would transform any pulse shape into a binary signal ideal for return-to-zero
on/off keying data transmission schemes, i.e. such a device would amplify the signal, but not the
noise.

4.3.4 Transition from dissipative to the conservative regime

In Chap. 3 the previously introduced non-linearities were derived from the Bloch equations with
and without losses leading to conservative and dissipative wave equations, respectively. Here, we
want to shed some light on this transition. Clearly, the dephasing rate $\gamma$ can be tuned to switch
between the conservative and dissipative regime.

A simple way to investigate this transition is to introduce a linear scaling factor $\lambda$ for the
dissipative parameters in Eq. (4.41).

$$\frac{i}{\xi} \frac{\partial \psi}{\partial \xi} - \frac{D}{2} \frac{\partial^2 \psi}{\partial \tau^2} = -i\tilde{\alpha} \lambda \psi - (1 + i\tilde{\epsilon} \lambda) |\psi|^2 \psi + (\tilde{b} - i\tilde{\mu} \lambda) |\psi|^4 \psi. \quad (4.49)$$

For $\lambda = 0$ an initial pulse, that is the solution to this NLSE is unchanged. Turning on the dissipation
parameter this initial condition evolves into a solution to the cGLE (cf. Fig. 4.7). However, in
a linear scaling approach the homogeneous solutions are not changed (cf. Eq. (4.44)), thus the
resulting solutions are similar in height and width.

![Figure 4.7: The evolution of an initial pulse for different values of $\lambda$ and thus in a conservative or more
dissipative regime. The pulse energy stays constant for the conservative case $\lambda = 0$. For the dissipative cases
the pulse energy is nearly the same, but the length scale on which the equilibrium is reach differs strongly.](image)
4.3. GENERALIZED COMPLEX GINZBURG-LANDAU TYPE EQUATIONS
Conclusion

In this work we described semiconductor QDs and their semiclassical optical properties. Chapter 2 showed the influence of phonons to optical spectra and to the occupation of QDs. The influence of the carrier wave functions, especially size and intrinsic dipole moment on various coupling mechanisms was discussed. Here, the importance of taking into account more realistic phonon dispersions for small QDs was stressed. Further, a phonon mediated coupling between QD states and a carrier reservoir was described via a rate equation approach. Therefore, a projection operator based formalism was used to derive an effective multiphonon Hamiltonian. With an equation of motion approach rate equations with temperature and reservoir carrier dependent rates were acquired.

The equations of motion for QDs in the rate equations limit were treated in Chap. 3 with the aim to derive analytical expressions. In a perturbative approach expressions for the polarization, that adiabatically follows the electric field were achieved for the three regimes of negligible losses, the presence of dephasing only, as well as for relaxation and dephasing.

Finally, in Chap. 4 the QDs’ optical response was used as a non-linearity in wave equations. Those wave equations are of the type of a generalized Ginzburg-Landau equation. The non-linear wave equations were investigated with respect to (dissipative) soliton solutions and the operating regime for pulse shaping semiconductor optical amplifiers was proposed.
Prefactors of the piezoelectric coupling

In Sec. 1.2.2 we introduce the piezoelectric coupling to acoustic phonons. Here, we want to show the angular dependent prefactor for Zincblende and Wurtzit systems.

The angle dependent Piezoelectric factors for Zincblende

\[ \mathcal{F}_Z(\vec{q}) = \frac{2e_{14}e}{q^2\varepsilon\varepsilon_0} (q_xq_y\varepsilon^z_{\kappa} + q_yq_z\varepsilon^x_{\kappa} + q_zq_x\varepsilon^y_{\kappa}), \]

or for Wurtzit systems (note that here the choice of the wave functions distance comes also into play. every thing shown here is valid for growth in 0001 direction)

\[ \mathcal{F}_W(\vec{q}) = \frac{e}{q^2\varepsilon\varepsilon_0} (e_{15}(q_x^2 + q_y^2)\varepsilon^z_{\kappa} + e_{33}q_z^2\varepsilon^z_{\kappa} + (e_{15} + e_{31})q_z(q_x\varepsilon^x_{\kappa} + q_y\varepsilon^y_{\kappa})). \]

In contrast to the deformation potential coupling both band have the same coupling strength, thus only the difference of the wave functions leads to a coupling. To calculate the angular average one has to insert \( \vec{q} = q\vec{e}_{LA} \), and the unit vectors in spherical coordinates.

Longitudinal mode \( \vec{e}_{LA} = \begin{pmatrix} \sin \theta \cos \phi \\ \sin \theta \sin \phi \\ \cos \theta \end{pmatrix} \), first transversal mode \( \vec{e}_{TA1} = \begin{pmatrix} \cos \theta \cos \phi \\ \cos \theta \sin \phi \\ -\sin \theta \end{pmatrix} \).
second transversal mode $\vec{e}_{TA2} = \begin{pmatrix} - \sin \phi \\ \cos \phi \\ 0 \end{pmatrix}$

$$F_{LA}^Z(\phi, \theta) = \frac{6e_{14}e}{\varepsilon_s \varepsilon_0} (\sin^2 \theta \cos \phi \sin \phi \cos \theta),$$

$$F_{TA1}^Z(\phi, \theta) = \frac{2e_{14}e}{\varepsilon_s \varepsilon_0} (-\sin^2 \theta + 2 \cos^2 \theta) \sin \theta \cos \phi \sin \phi,$$

$$F_{TA2}^Z(\phi, \theta) = \frac{2e_{14}e}{\varepsilon_s \varepsilon_0} (-\sin^2 \phi + \cos^2 \phi) \sin \theta \cos \phi,$$

averaged

$$\left(F_{LA}^Z\right)^2 = \frac{1}{4\pi} \frac{4e_{14}^2 e^2}{\varepsilon_s \varepsilon_0} \frac{12\pi}{35},$$

$$\left(F_{TA1}^Z\right)^2 = \frac{1}{4\pi} \frac{4e_{14}^2 e^2}{\varepsilon_s \varepsilon_0} \frac{23\pi}{210},$$

$$\left(F_{TA2}^Z\right)^2 = \frac{1}{4\pi} \frac{4e_{14}^2 e^2}{\varepsilon_s \varepsilon_0} \frac{\pi}{6}.$$

For Wurtzite

$$F_{LA}^W(\phi, \theta) = \frac{e}{\varepsilon_s \varepsilon_0} (e_{33} \cos^2 \theta + (2e_{15} + e_{31}) \sin^2 \theta) \cos \theta.$$

$$F_{TA1}^W(\phi, \theta) = \frac{e}{\varepsilon_s \varepsilon_0} (-e_{15} \sin^2 \theta + (e_{15} + e_{31} - e_{33}) \cos^2 \theta) \sin \theta.$$

$$F_{TA2}^W(\phi, \theta) = 0.$$

averaged

$$\left(F_{LA}^W\right)^2 = \frac{1}{4\pi} \frac{e^2}{\varepsilon_s \varepsilon_0} \left(\frac{4}{105} \left(8 (2e_{15} + e_{31})^2 + 12 (2e_{15} + e_{31}) e_{33} + 15e_{33}^2\right) \pi\right),$$

$$\left(F_{TA1}^W\right)^2 = \frac{1}{4\pi} \frac{e^2}{\varepsilon_s \varepsilon_0} \left(\frac{8}{105} \left(19e_{15}^2 + 3 (e_{31} - e_{33})^2 + 2e_{15} (-e_{31} + e_{33})\right) \pi\right),$$

$$\left(F_{TA2}^W\right)^2 = 0.$$

$$\left(F_{LA}^W\right)^2 = \frac{e^2}{\varepsilon_s \varepsilon_0} \frac{1}{105} \left(32e_{15}^2 + 8e_{31}^2 + 15e_{33}^2 + 32e_{15}e_{31} + 24e_{15}e_{33} + 12e_{31}e_{33}\right)$$

$$\left(F_{TA1}^W\right)^2 = \frac{e^2}{\varepsilon_s \varepsilon_0} \frac{1}{105} \left(38e_{15}^2 + 6e_{31}^2 + 6e_{33}^2 - 4e_{15}e_{31} + 4e_{15}e_{33} - 12e_{31}e_{33}\right)$$

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Induction proof of the Bloch equation’s perturbation treatment

In Sec. 3.1.2 an infinite series for the polarization and the inversion of the quantum dots is derived. Here is the proof via the mathematical induction method. For the proof we need Eqs. (3.7):

\[ \rho_{12}^{(n+1)}(t) = i \int_{-\infty}^{t} dt' e^{i\Delta\omega(t'-t)} \Omega(t') u_{3}^{(n)}(t'), \]  
\[ u_{3}^{(n+1)} = -4 \int_{-\infty}^{t} dt' \text{Im}(\Omega^*(t') \rho_{12}^{(n)}(t')). \] (B.1a)

Assume

\[ u_{3}^{(2n)} = (-1)^n \frac{4^n}{\Delta\omega^{2n}} \frac{\Gamma(n + 1/2)}{\sqrt{\pi n!}} |\Omega|^{2n} u_{3}^{(0)}, \] (B.2)

for \( n \geq 0 \) with the \( \Gamma \) function with the property \( x\Gamma(x) = \Gamma(x + 1) \) and \( \Gamma(1/2) = \sqrt{\pi} \). This definition is true for \( n = 0 \). Then, using Eq. (B.1a) the polarization of order \( 2n + 1 \) reads

\[ \rho_{12}^{(2n+1)} = (-1)^n \frac{4^n}{\Delta\omega^{2n+1}} \frac{\Gamma(n + 1/2)}{\sqrt{\pi n!}} u_{3}^{(0)} \left( \Omega^*(t)|\Omega|^{2n}(t) + i \frac{1}{\Delta\omega} \partial_t |\Omega|^{2n}(t) \Omega^*(t) \right). \] (B.3)

This yields the next order inversion via Eq. (B.1b)

\[ u_{3}^{(2n+1)}(t) = (-1)^{(n+1)} \frac{4^{n+1}}{\Delta\omega^{2n+2}} \frac{\Gamma(n + 1/2)}{\sqrt{\pi n!}} u_{3}^{(0)} \text{Im} \left( \frac{i}{\Delta\omega} \Omega(t) \partial_t |\Omega|^{2n}(t) \Omega^*(t) \right), \] (B.4)

where the derivative can be brought in front of the equation via

\[ \Omega \partial_t \Omega^* |\Omega|^{2n} = \frac{2n + 1}{2n + 2} \partial_t |\Omega|^{2n+2} - i |\Omega|^{2n+2} \partial_t \varphi; \quad \Omega = |\Omega| e^{i\varphi}. \] (B.5)
Thus the inversion of order $2(n+1)$ is

$$u_3^{(2(n+1))}(t) = (-1)^{(n+1)} \frac{4^{n+1}}{\Delta \omega^2(n+1)} \frac{\Gamma(n + 1 + 1/2)!}{\sqrt{\pi(n + 1)!}} u_3^{(0)} \Omega|^{2(n+1)}, \tag{B.6}$$

which is the proof of the general form assumed in Eq. (B.2). Using $\Gamma(n + 1/2) = \sqrt{\frac{(2n)!}{\pi^{n+1}}} [BSM+00]$ we can write:

$$u_3(t) = \left[ \sum_{n=0}^{\infty} (-1)^n \frac{1}{\Delta \omega^{2n}} \frac{(2n)!}{(n!)^2} |\Omega|^{2n} \right] u_3^{(0)}, \tag{B.7}$$

$$\rho_{12}(t) = \left[ \sum_{n=0}^{\infty} (-1)^n \frac{1}{\Delta \omega^{2n+1}} \frac{(2n)!}{(n!)^2} \left( \Omega^*(t)|\Omega|^{2n}(t) + i \frac{1}{\Delta \omega} \partial_{t'}|\Omega|^{2n}(t') \Omega^*(t') \right) \right] u_3^{(0)}. \tag{B.8}$$
Bibliography


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