Arbeit zur Erlangung des akademischen Grades Master of Science

# Dynamics of a Dimerized $S=1 / 2$ Chain <br> Continously Driven via a Coupled Phonon 

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#### Abstract

Being able to fully control a quantum system offers an enormous amount of possibilities in finding new effects in physics. Although lots of progress has been made over the past decades, this topic still holds great potential. To make a contribution to this interesting field of research we propose a simple model consisting, in its core, of a dimerized spin $1 / 2$ chain coupled to a phonon system. We model an excitation with a laser aiming at driving the spin system into a quasi steady state in order to investigate non-equilibrium dynamics. Employing the bond operator representation to describe the spin system and the Lindblad formalism to induce a damping to the phonon system we approximate the Hamiltonian to a bilinear level subsequently investigating the system's properties depending on the coupling strengths, and excitation and damping parameters.

We encounter non-physical effects of divergence of the spin system, which we identify as an effect of a resonance disaster, limiting the applicability of our model. For those regimes, where our assumptions are valid, we find non-trivial quasi steady states and further investigate properties of Green's functions in order to understand mechanisms behind the interactions between the spin and phonon section of the system.


## Kurzfassung

Die volle Kontrolle über ein Quantensystem zu haben bietet einen enormen Satz an Möglichkeiten, neue Effekte in der Physik zu finden. Wenngleich schon viele Fortschritte in den letzten Jahrzehnten gemacht wurden, bietet dieses Thema immer noch großes Potential. Um einen Beitrag zu diesem interessanten Forschungsgebiet zu leisten, entwickeln wir ein Modell, das im Kern aus einer dimerisierten Spin- $1 / 2$-Kette besteht, die an ein Phononsystem koppelt. Wir modellieren eine Anregung mit einem Laser mit dem Ziel, das Spinsystem in einen Quasigleichgewichtszustand zu treiben, um Nichtgleichgewichtsdynamiken untersuchen zu können. Wir verwenden die Bond-Operator-Darstellung für das Spinsystem und den LindbladFormalismus, um das Phononsystem zu dämpfen, und nähern den Hamilton-Operator auf eine bilineare Form, um in der Folge die Eigenschaften des Systems in Abhängigkeit von den Kopplungsstärken, der Anregungs-, sowie der Dämpfungsparameter zu untersuchen.

In unseren Untersuchungen finden wir nichtphysikalische Effekte in Form von Divergenzen des Spinsystems, die wir als die Folge einer Resonanzkatasdrophe ausmachen können, was die Anwendbarkeit unseres Modelles einschränkt. In denjenigen Bereichen, in denen unsere Annahmen Gültigkeit erlangen, beobachten wir nicht-triviale Quasigleichgewichtszustände und untersuchen weiterhin die Eigenschaften Green'scher Funktionen, um die Mechanismen hinter den Wechselwirkungen zwischen dem Spin- und dem Phononanteil des Systems zu verstehen.

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## 1 Introduction

Without the achievements of solid state physics our everyday life would differ entirely from what it is today. The investigation and classification of semiconductors led to the processing of bipolar junction of field-effect transistors, which form the basis of modern computers. There would be no touch-screens if it was not for the investigation of liquid crystals, and, most certainly, we would not have smartphones to connect us anywhere anytime.

In other fields, of which the applications do not affect our everyday life as strongly, progress has been made as well. The explanation of the effect of superconductivity serves as a prime example. Topological insulators could further revolutionise human technology and advances in soft matter physics could help us understand biological material better.

Non-equilibrium dynamics are a particularly active field in modern solid state physics. Here, we present a short overview on its current status outlining some concepts and open questions. The rest of this thesis is structured as follows: In chapter 2 we discuss methods we need to set up our model in chapter 3. We present our findings in chapter 4 reflecting on them in chapter 5

### 1.1 Motivation

With new technologies on the rise, we encounter effects that cannot be described by linear response theory [22]. Thus, we focus on non-equilibrium systems in hopes of gaining new insights especially on non-linear effects. Several problems have already been studied, e.g. non-adiabatic responses in BCS superconductors [23], quantum phase transitions [18] or spin waves [16]. For non-equilibrium states hardly occur in nature, particular focus is put on driven systems [25], e.g. on angle resolved photoelectron spectroscopy [34, 40]. Alongside the vast amount of experiments, there comes an equally large number of theoretical concepts trying to describe those effects or help solving associated problems. From a very problem specific approach via the representation as a Bose gas [28] to more general methods like continuous unitary transformations [7] and associated flow equations [20 theoretical physics offers a great set of tools when it comes to describing non-equilibrium processes. A very important concept is the introduction of new quasi particles called hardcore bosons 9 . Originally proposed by Sachdev and Bhatt [36] it now serves as a basis to many problems involving spin systems.

### 1.2 Goal

The mere observation and explanation of new effects alone may enhance our understanding of the physics behind them but of greater benefit would be their application in practice. Therefore, we are not only interested in describing new effects but also in manipulating the corresponding systems at will into states we can control and use for our purposes. In addition to some general work [19] rather special problems have been approached. The group of Mentink et al. managed to control the coupling strength in Mott insulators [27] whereas Lange at al. succeeded in inducting heat currents in their probes via pumping [24]. We will present a quite similar model of which the key purpose is to gain control over the state we pump the system into. Having driven the system into a quasi steady state we can further manipulate it which would present us with great potential to create an adjustable quantum system of which the number of applications is immense. Ideas reach from data carriers to gates to entire quantum computers. Hence, a detailed study of these aspects is of crucial interest, not only to physicists.

### 1.3 Possible Materials

A great amount of faith in finding appropriate candidates for an implementation of such systems is put into spin $1 / 2$ systems. Over the years of research several solids with different properties have been investigated. Thus, we shall only give a short overview and provide some examples. A comparably simple ansatz is given by $\mathrm{CuGeO}_{3}$ 45 which can be described by a spin chain, i.e. the model is only (quasi) one-dimensional. This inorganic spin-Peierls compound can as a first approximation be described as a frustrated Heisenberg antiferromagnet coupled to the lattice [8]. A more complex structure is given by (VO) ${ }_{2} \mathrm{P}_{2} \mathrm{O}_{7}(\mathrm{VOPO})$ [44] which consists of alternating chains. Such quantum magnets like $\mathrm{BiCu}_{2} \mathrm{PO}_{6}$ (BCPO) [32] with its orthorhombic crystal structure are described using two-dimensional models with BCPO being an example of a frustrated spin ladder. A similar compound is $\mathrm{SrCu}_{2}\left(\mathrm{BO}_{3}\right)_{2}$ alias SCBO [46]. It can be best described by the two-dimensional Shastry-Sutherland model. The strong-leg spin ladder system $\left(\mathrm{C}_{7} \mathrm{H}_{10} \mathrm{~N}\right) \mathrm{CuBr}_{4}$ also called DIMPY 38] features not only one but two ladder systems making it even more complex.

In this thesis, we will discuss a spin chain inspired by $\mathrm{CuGeO}_{3}$.

## 2 Methods

We present basic concepts to approach problems of the project. As most of those methods are textbook knowledge, we will provide some references the corresponding sections are primarily based on. Unless explicitly noted we take

$$
\begin{equation*}
c=\hbar=1 \tag{2.1}
\end{equation*}
$$

$c$ being the speed of light and $\hbar$ the reduced Planck constant.

### 2.1 Operators in Quantum Mechanics

The defining characteristic of a quantum system is its restriction to only absorb or emit energy in certain amounts called quanta. In order to describe such a system operators are introduced of which the most important properties are their commutation relations. These commutators provide a specific algebra that is essential when it comes to investigating their physical behaviour.

There are two basic classes of operators and, thus, (quasi) particles, bosons and fermions. If the relations

$$
\begin{align*}
\left\{\boldsymbol{f}_{M}, \boldsymbol{f}_{M^{\prime}}\right\} & =0  \tag{2.2a}\\
\left\{\boldsymbol{f}_{M}^{\dagger}, \boldsymbol{f}_{M^{\prime}}^{\dagger}\right\} & =0  \tag{2.2b}\\
\left\{\boldsymbol{f}_{M}, \boldsymbol{f}_{M^{\prime}}^{\dagger}\right\} & =\delta_{M, M^{\prime}}, \tag{2.2c}
\end{align*}
$$

where $\{\cdot, \cdot\}$ denotes the anticommutator, hold for two operators $\boldsymbol{f}$ and $\boldsymbol{f}^{\dagger}$ they are called fermionic operators. The indices $M$ and $M^{\prime}$ may represent any finite set of numbers, $f^{\dagger}$ denotes the hermitian conjugate of $\boldsymbol{f}$. If two operators $\boldsymbol{b}$ and $\boldsymbol{b}^{\dagger}$, however, satisfy

$$
\begin{align*}
{\left[\boldsymbol{b}_{M}, \boldsymbol{b}_{M^{\prime}}\right] } & =0  \tag{2.3a}\\
{\left[\boldsymbol{b}_{M}^{\dagger}, \boldsymbol{b}_{M^{\prime}}^{\dagger}\right] } & =0  \tag{2.3b}\\
{\left[\boldsymbol{b}_{M}, \boldsymbol{b}_{M^{\prime}}^{\dagger}\right] } & =\delta_{M, M^{\prime}} \tag{2.3c}
\end{align*}
$$

they are referred to as bosonic operators with $[\cdot, \cdot]$ being the commutator.
Spins are of particular interest in modern quantum physics. A spin operator

$$
\begin{equation*}
\overrightarrow{\boldsymbol{S}}=\left(\boldsymbol{S}_{x}, \boldsymbol{S}_{y}, \boldsymbol{S}_{z}\right)^{\top} \tag{2.4}
\end{equation*}
$$

is characterized by the commutation relation

$$
\begin{equation*}
\left[\boldsymbol{S}_{i}, \boldsymbol{S}_{j}\right]=\mathrm{i} \varepsilon_{i j k} \boldsymbol{S}_{k} \tag{2.5}
\end{equation*}
$$

where $\varepsilon_{i j k}$ is the Levi-Civita symbol and $i, j$ and $k$ may take values of $x, y$ and $z$. For spins $S=1 / 2$ the components are given by

$$
\begin{equation*}
\boldsymbol{S}_{i}=\frac{1}{2} \sigma_{i} \tag{2.6}
\end{equation*}
$$

with the Pauli matrices

$$
\begin{align*}
\sigma_{x} & =\left(\begin{array}{ll}
0 & 1 \\
1 & 0
\end{array}\right)  \tag{2.7a}\\
\sigma_{y} & =\left(\begin{array}{cc}
0 & -\mathrm{i} \\
\mathrm{i} & 0
\end{array}\right)  \tag{2.7b}\\
\sigma_{z} & =\left(\begin{array}{cc}
1 & 0 \\
0 & -1
\end{array}\right) . \tag{2.7c}
\end{align*}
$$

As is evident from equation (2.5) spin operators are neither of bosonic nor of fermionic type. There are, however, several methods to transform them into operators that are at least close to being bosonic or fermionic. We will discuss a useful represetation in the next chapter.

### 2.2 Bond Operator Representation

When describing dimers formed by two spins interacting with one another the bond operator representation introduced by Sachdev and Bhatt [36] comes in handy. Its wide application $[39,20,21,43,6,10]$ speaks for its usefulness when approaching strongly correlated systems. There are two slightly different ways to construct this representation. In what follows we shall outline the key ideas to both approaches.

### 2.2.1 Sachdev's Representation

We investigate a dimer consisting of two spins $\boldsymbol{S}=1 / 2$ as sketched in figure 2.1. The spin on


Figure 2.1: Schematic picture of a dimer; the spins $\overrightarrow{\boldsymbol{S}}_{i}^{1}$ and $\overrightarrow{\boldsymbol{S}}_{i}^{2}$ are depicted by red dots, whereas the black bar illustrates the coupling $J$
its left side we call $\overrightarrow{\boldsymbol{S}}_{i}^{1}$, and the one on the right $\overrightarrow{\boldsymbol{S}}_{i}^{2}$ respectively. The index $i$ denotes the number of the dimer considered derived from the site where the dimer is located.

A two spin system is described by a four dimensional Hilbert space which is thus the dimension of the Hamiltonian. Its four eigenstates which are given by a singlet and three triplet states serve as the basis to the bond operator representation. We envisage four operators $\tilde{\boldsymbol{s}}_{i}^{\dagger}, \tilde{\boldsymbol{t}}_{x, i}^{\dagger}, \tilde{\boldsymbol{t}}_{y, i}^{\dagger}$ and $\tilde{\boldsymbol{t}}_{z, i}^{\dagger}$ to create the singlet and triplet states $|s\rangle_{i},|x\rangle_{i},|y\rangle_{i}$ and $|z\rangle_{i}$ from a fictitious vacuum $|0\rangle$ as follows

$$
\begin{align*}
\tilde{\boldsymbol{s}}_{i}^{\dagger}|0\rangle & =|s\rangle_{i}
\end{aligned}=\frac{1}{\sqrt{2}}\left(|\uparrow \downarrow\rangle_{i}-|\downarrow \uparrow\rangle_{i}\right), \begin{aligned}
\tilde{\boldsymbol{t}}_{x, i}^{\dagger}|0\rangle & =|x\rangle_{i} \tag{2.8a}
\end{align*}=-\frac{1}{\sqrt{2}}\left(|\uparrow \uparrow\rangle_{i}-|\downarrow \downarrow\rangle_{i}\right) .
$$

Hence, the spin operators $\overrightarrow{\boldsymbol{S}}_{i}^{1}$ and $\overrightarrow{\boldsymbol{S}}_{i}^{2}$ are given by

$$
\begin{align*}
& \boldsymbol{S}_{\alpha, i}^{1}=\frac{1}{2}\left(\tilde{\boldsymbol{s}}_{i}^{\dagger} \tilde{\boldsymbol{t}}_{\alpha, i}+\tilde{\boldsymbol{t}}_{\alpha, i}^{\dagger} \tilde{\boldsymbol{s}}_{i}-\mathrm{i} \sum_{\beta \gamma} \varepsilon_{\alpha \beta \gamma} \tilde{\boldsymbol{t}}_{\beta, i}^{\dagger} \tilde{\boldsymbol{t}}_{\gamma, i}\right)  \tag{2.9a}\\
& \boldsymbol{S}_{\alpha, i}^{2}=-\frac{1}{2}\left(\tilde{\boldsymbol{s}}_{i}^{\dagger} \tilde{\boldsymbol{t}}_{\alpha, i}+\tilde{\boldsymbol{t}}_{\alpha, i}^{\dagger} \tilde{\boldsymbol{s}}_{i}+\mathrm{i} \sum_{\beta \gamma} \varepsilon_{\alpha \beta \gamma} \tilde{\boldsymbol{t}}_{\beta, i}^{\dagger} \tilde{\boldsymbol{t}}_{\gamma, i}\right) \tag{2.9b}
\end{align*}
$$

The Greek indices $\alpha, \beta$ and $\gamma$ may take the values of the flavours $x, y$ and $z$. In order to only create singlet or triplet states the hardcore constraint

$$
\begin{equation*}
\tilde{\boldsymbol{s}}_{i}^{\dagger} \tilde{\boldsymbol{s}}_{i}+\sum_{\alpha} \tilde{\boldsymbol{t}}_{\alpha, i}^{\dagger} \tilde{\boldsymbol{t}}_{\alpha, i}=1 \tag{2.10}
\end{equation*}
$$

has to hold. It is straight forward to deduce

$$
\begin{gather*}
\tilde{\boldsymbol{s}}_{i} \tilde{\boldsymbol{s}}_{i}=\tilde{\boldsymbol{t}}_{\alpha, i} \tilde{\boldsymbol{t}}_{\beta, i}=\tilde{\boldsymbol{s}}_{i}^{\dagger} \tilde{\boldsymbol{s}}_{i}^{\dagger}=\tilde{\boldsymbol{t}}_{\alpha, i}^{\dagger} \tilde{\boldsymbol{t}}_{\beta, i}^{\dagger}=0  \tag{2.11a}\\
\tilde{\boldsymbol{s}}_{i}^{\dagger} \tilde{\boldsymbol{s}}_{i}, \tilde{\boldsymbol{t}}_{\alpha, i}^{\dagger} \tilde{\boldsymbol{t}}_{\alpha, i} \in\{0,1\} \tag{2.11b}
\end{gather*}
$$

meaning there can only be one out of the four quasi particles at the same time at each dimer. The newly introduced bond operators $\tilde{\boldsymbol{s}}_{i}^{\dagger}, \tilde{\boldsymbol{t}}_{x, i}^{\dagger}, \tilde{\boldsymbol{t}}_{y, i}^{\dagger}$ and $\tilde{\boldsymbol{t}}_{z, i}^{\dagger}$ are of bosonic nature. Hence, the commutation relations read

$$
\begin{align*}
{\left[\tilde{\boldsymbol{s}}_{i}, \tilde{\boldsymbol{s}}_{j}^{\dagger}\right] } & =\delta_{i, j}  \tag{2.12a}\\
{\left[\tilde{\boldsymbol{t}}_{\alpha, i}, \tilde{\boldsymbol{t}}_{\beta, j}^{\dagger}\right] } & =\delta_{\alpha, \beta} \delta_{i, j}  \tag{2.12b}\\
{\left[\tilde{\boldsymbol{s}}_{i}, \tilde{\boldsymbol{s}}_{j}\right]=\left[\tilde{\boldsymbol{t}}_{\alpha, i}, \tilde{\boldsymbol{t}}_{\beta, j}\right]=\left[\tilde{\boldsymbol{s}}_{i}, \tilde{\boldsymbol{t}}_{\alpha, j}\right]=\left[\tilde{\boldsymbol{s}}_{i}^{\dagger}, \tilde{\boldsymbol{t}}_{\alpha, j}\right] } & =0  \tag{2.12c}\\
{\left[\tilde{\boldsymbol{s}}_{i}^{\dagger}, \tilde{\boldsymbol{s}}_{j}^{\dagger}\right]=\left[\tilde{\boldsymbol{t}}_{\alpha, i}^{\dagger}, \tilde{\boldsymbol{t}}_{\beta, j}^{\dagger}\right]=\left[\tilde{\boldsymbol{s}}_{i}, \tilde{\boldsymbol{t}}_{\alpha, j}^{\dagger}\right]=\left[\tilde{\boldsymbol{s}}_{i}^{\dagger}, \tilde{\boldsymbol{t}}_{\alpha, j}^{\dagger}\right] } & =0 . \tag{2.12d}
\end{align*}
$$

Note that additionally the hardcore constraint $(2.10)$ has to hold for any dimer $i$.

### 2.2.2 Triplon Operator Representation

In order to further substantiate the approach introduced in the previous section, we define the singlet state as the vacuum

$$
\begin{equation*}
|0\rangle:=|s\rangle_{i}=\frac{1}{\sqrt{2}}\left(|\uparrow \downarrow\rangle_{i}-|\downarrow \uparrow\rangle_{i}\right) . \tag{2.13}
\end{equation*}
$$

For the singlet state of a dimer is the one of the lowest energy this choice is convenient. Often times, this equals setting $\tilde{s}_{i}=1$ in equations of the previous section. Now, we introduce new operators

$$
\begin{align*}
\boldsymbol{s}_{i}^{\dagger} & =|s\rangle_{i}\left\langle\left. s\right|_{i}\right.  \tag{2.14a}\\
\boldsymbol{t}_{x, i}^{\dagger} & =|x\rangle_{i}\left\langle\left. s\right|_{i}\right.  \tag{2.14b}\\
\boldsymbol{t}_{y, i}^{\dagger} & =|y\rangle_{i}\left\langle\left. s\right|_{i}\right.  \tag{2.14c}\\
\boldsymbol{t}_{z, i}^{\dagger} & =|z\rangle_{i}\left\langle\left. s\right|_{i} .\right. \tag{2.14d}
\end{align*}
$$

Replacing the old bond operators in equation (2.9) with these new ones yields

$$
\begin{align*}
& \boldsymbol{S}_{\alpha, i}^{1}=\frac{1}{2}\left(\boldsymbol{t}_{\alpha, i}+\boldsymbol{t}_{\alpha, i}^{\dagger}-\mathrm{i} \sum_{\beta \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i}\right)  \tag{2.15a}\\
& \boldsymbol{S}_{\alpha, i}^{2}=-\frac{1}{2}\left(\boldsymbol{t}_{\alpha, i}+\boldsymbol{t}_{\alpha, i}^{\dagger}+\mathrm{i} \sum_{\beta \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i}\right) . \tag{2.15b}
\end{align*}
$$

Using the representation (2.14)

$$
\begin{equation*}
\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\beta, i}=\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\beta, i}^{\dagger}=0 \tag{2.16}
\end{equation*}
$$

obviously holds. The hardcore constraint (2.10) now reads

$$
\begin{equation*}
|s\rangle_{i}\left\langle\left. s\right|_{i}+\sum_{\alpha} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i}=1 \quad \Leftrightarrow \quad \mid s\right\rangle_{i}\left\langle\left. s\right|_{i}=1-\sum_{\alpha} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i} .\right. \tag{2.17}
\end{equation*}
$$

It is needed to calculate the only remaining non-trivial commutator

$$
\begin{align*}
{\left[\boldsymbol{t}_{\alpha, i}, \boldsymbol{t}_{\beta, i}^{\dagger}\right] } & =\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\beta, i}^{\dagger}-\boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\alpha, i}=|s\rangle_{i}\left\langle\left.\alpha\right|_{i} \mid \beta\right\rangle_{i}\left\langle\left. s\right|_{i}-\mid \beta\right\rangle_{i}\left\langle\left. s\right|_{i} \mid s\right\rangle_{i}\left\langle\left.\alpha\right|_{i}\right. \\
& =\delta_{\alpha, \beta}|s\rangle_{i}\left\langle\left. s\right|_{i}-\mid \beta\right\rangle_{i}\left\langle\left.\alpha\right|_{i} \stackrel{(2.17)}{=} \delta_{\alpha, \beta}\left(1-\sum_{\gamma} \boldsymbol{t}_{\gamma, i}^{\dagger} \boldsymbol{t}_{\gamma, i}\right)-\boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\alpha, i} .\right. \tag{2.18}
\end{align*}
$$

Because two dimers on different lattice sites are independent of one another, the final commutation relation is given by

$$
\begin{equation*}
\left[\boldsymbol{t}_{\alpha, i}, \boldsymbol{t}_{\beta, j}^{\dagger}\right]=\delta_{i, j}\left(\delta_{\alpha, \beta}\left(1-\sum_{\gamma} \boldsymbol{t}_{\gamma, i}^{\dagger} \boldsymbol{t}_{\gamma, i}\right)-\boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\alpha, i}\right) \tag{2.19}
\end{equation*}
$$

which is not bosonic but what is called hardcore bosonic. Thus, we obtain

- for $i \neq j$

$$
\begin{equation*}
\left[\boldsymbol{t}_{\alpha, i}, \boldsymbol{t}_{\beta, j}^{\dagger}\right]=0 \tag{2.20a}
\end{equation*}
$$

- for $i=j$ and $\alpha \neq \beta$

$$
\begin{equation*}
\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\beta, i}^{\dagger}=0 \tag{2.20b}
\end{equation*}
$$

- for $i=j$ and $\alpha=\beta$

$$
\begin{equation*}
\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i}^{\dagger}=1-\sum_{\gamma} \boldsymbol{t}_{\gamma, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \tag{2.20c}
\end{equation*}
$$

### 2.3 The Heisenberg Equation of Motion

The mathematics of quantum mechanics can be expressed in several ways. We shall, at first, present the Schrödinger picture where the time evolution of a state vector $|\psi(t)\rangle$ is governed by the Schrödinger equation

$$
\begin{equation*}
\mathrm{i} \frac{\mathrm{~d}}{\mathrm{~d} t}|\psi(t)\rangle=\boldsymbol{H}(t)|\psi(t)\rangle \tag{2.21}
\end{equation*}
$$

The Hamiltonian $\boldsymbol{H}$ may in the most general case be time dependent. A solution to (2.21) is obtained by introducing the time evolution operator $U\left(t, t_{0}\right)$ which transforms the state vector from its initial state $\left|\psi\left(t_{0}\right)\right\rangle$ at time $t_{0}$ into its current state $|\psi(t)\rangle$ at time $t$, i.e.

$$
\begin{equation*}
|\psi(t)\rangle=U\left(t, t_{0}\right)\left|\psi\left(t_{0}\right)\right\rangle . \tag{2.22}
\end{equation*}
$$

For a time independent Hamiltonian the time evolution operator reads

$$
\begin{equation*}
U\left(t, t_{0}\right)=\mathrm{e}^{-\mathrm{i} \boldsymbol{H} \cdot\left(t-t_{0}\right)} . \tag{2.23}
\end{equation*}
$$

The Heisenberg picture is characterised by transferring the time dependence of state vectors in the Schrödinger picture solely to the operators via

$$
\begin{equation*}
\boldsymbol{O}_{\mathrm{H}}(t)=U^{\dagger}\left(t, t_{0}\right) \boldsymbol{O}_{\mathrm{S}}(t) U\left(t, t_{0}\right) \tag{2.24}
\end{equation*}
$$

considering an arbitrary operator $\boldsymbol{O}$, denoting it as $\boldsymbol{O}_{\mathrm{H}}$ or $\boldsymbol{O}_{\mathrm{S}}$ if it is in the Heisenberg or the Schrödinger picture respectively. In order to calculate the time evolution, we make use of the Heisenberg equation of motion

$$
\begin{equation*}
\frac{\mathrm{d} \boldsymbol{O}_{\mathrm{H}}(t)}{\mathrm{d} t}=\mathrm{i}\left[\boldsymbol{H}_{\mathrm{H}}, \boldsymbol{O}_{\mathrm{H}}(t)\right]+\left(\partial_{t} \boldsymbol{O}_{\mathrm{S}}(t)\right)_{\mathrm{H}}, \tag{2.25}
\end{equation*}
$$

which we obtained from differentiating equation (2.24). Instead of focusing on the Heisenberg picture, it is also possible to investigate the expectation values yielding

$$
\begin{equation*}
\frac{\mathrm{d}\left\langle\boldsymbol{O}_{\mathrm{H}}\right\rangle(t)}{\mathrm{d} t}=\mathrm{i}\left\langle\left[\boldsymbol{H}_{\mathrm{H}}, \boldsymbol{O}_{\mathrm{H}}\right]\right\rangle+\left\langle\left(\partial_{t} \boldsymbol{O}_{\mathrm{S}}(t)\right)_{\mathrm{H}}\right\rangle \tag{2.26}
\end{equation*}
$$

which, when taking the independence of the picture into account, reduces to

$$
\begin{equation*}
\frac{\mathrm{d}\langle\boldsymbol{O}\rangle(t)}{\mathrm{d} t}=\mathrm{i}\langle[\boldsymbol{H}, \boldsymbol{O}(t)]\rangle+\left\langle\partial_{t} \boldsymbol{O}(t)\right\rangle \tag{2.27}
\end{equation*}
$$

For a time independent operator $\boldsymbol{O}$ in the Schrödinger picture, i.e.

$$
\begin{equation*}
\frac{\partial \boldsymbol{O}_{\mathrm{S}}(t)}{\partial t}=0 \tag{2.28}
\end{equation*}
$$

we obtain

$$
\begin{equation*}
\frac{\mathrm{d}\langle\boldsymbol{O}\rangle(t)}{\mathrm{d} t}=\mathrm{i}\langle[\boldsymbol{H}, \boldsymbol{O}(t)]\rangle . \tag{2.29}
\end{equation*}
$$

This derivation can be found in more detail in reference [41].

### 2.4 The Lindblad Formalism

Usage of the Heisenberg equation of motion enables us to deduce the complete time evolution of the whole system under investigation. For larger systems this procedure becomes more and more tedious or virtually impossible. Often, only a few quantities of a large system are of interest. Therefore, the exact behaviour of the remaining part can be neglected. The Lindblad formalism [26] provides a straight forward method to isolate a desired part from the rest of the system under consideration. This chapter largely follows the work of [3] where several examples are discussed as well.

Whereas previously only closed systems, of which the dynamics can be represented by a unitary time evolution, were examined, we now turn to open systems where this is, in general, not possible. Let $S+B$ be a total quantum system which we take to be closed. It consists of an open system $S$ in which we are interested and an environmental system $B$. Conveniently, the system $S$ is also referred to by the term "reduced system". The environmental system $B$ is called "bath" if it is in thermal equilibrium and has an infinite number of degrees of freedom. We neglect correlations between the environmental system $B$ and the reduced system $S$ meaning there is no memory to the environment which characterises a Markov process. By $\mathcal{H}$ we shall denote the total system's Hilbert space and by $\mathcal{H}_{\mathrm{S}}$ and $\mathcal{H}_{\mathrm{B}}$ those of the reduced and environmental system, yielding

$$
\begin{equation*}
\mathcal{H}=\mathcal{H}_{\mathrm{S}} \otimes \mathcal{H}_{\mathrm{B}} . \tag{2.30}
\end{equation*}
$$

Thus, the total Hamiltonian reads

$$
\begin{equation*}
\boldsymbol{H}=\boldsymbol{H}_{\mathrm{S}} \otimes \boldsymbol{I}_{\mathrm{B}}+\boldsymbol{I}_{\mathrm{S}} \otimes \boldsymbol{H}_{\mathrm{B}}+\boldsymbol{H}_{\mathrm{I}} \tag{2.31}
\end{equation*}
$$

with $\boldsymbol{H}_{\mathrm{S}}$ being the system's Hamiltonian, $\boldsymbol{H}_{\mathrm{B}}$ the Hamiltonian of the environment, $\boldsymbol{H}_{\mathrm{I}}$ the Hamiltonian describing the interaction between the open system $S$ and the environment $B$ and $\boldsymbol{I}$ the corresponding identities. Figure 2.2 sketches the investigated system. Having defined the system of interest $S$ an observable of interest reads $\boldsymbol{O} \otimes \boldsymbol{I}_{\mathrm{B}}$ for an operator $\boldsymbol{O}$ acting on the reduced system's Hilbert space $\mathcal{H}_{\mathrm{S}}$. Furthermore we are enabled to define the system's density matrix $\rho_{\mathrm{S}}$ by tracing the total system's density matrix $\rho$ over the Hilbert space $\mathcal{H}_{\mathrm{B}}$ describing the environmental system $B$

$$
\begin{equation*}
\rho_{\mathrm{S}}=\operatorname{Tr}_{\mathrm{B}}(\rho) . \tag{2.32}
\end{equation*}
$$



Figure 2.2: Schematic picture of an open quantum system, c.f. 3

Its time evolution is given by

$$
\begin{equation*}
\rho_{\mathrm{S}}(t)=\operatorname{Tr}_{\mathrm{B}}\left(\boldsymbol{U}\left(t, t_{0}\right) \rho\left(t_{0}\right) \boldsymbol{U}^{\dagger}\left(t, t_{0}\right)\right) \tag{2.33}
\end{equation*}
$$

with $\boldsymbol{U}\left(t, t_{0}\right)$ being the total system's time evolution operator. Analogously to the system $S$ its density matrix $\rho_{\mathrm{S}}$ goes by the name "reduced density matrix".

Assuming the dynamics of the total system to be described by a Markov process leads to the derivation of the Markovian quantum master equation. By making use of the semigroup property we introduce the dynamical map

$$
\begin{equation*}
V(t)=\exp (\mathcal{L} t), \tag{2.34}
\end{equation*}
$$

with the generator $\mathcal{L}$ to be further explained in equation (2.38), satisfying

$$
\begin{equation*}
\rho_{\mathrm{S}}(t)=V(t) \rho_{\mathrm{S}}(0) \equiv \operatorname{Tr}_{\mathrm{B}}\left(\boldsymbol{U}(t, 0)\left(\rho_{\mathrm{S}}(0) \otimes \rho_{\mathrm{B}}\right) \boldsymbol{U}^{\dagger}(t, 0)\right) \tag{2.35}
\end{equation*}
$$

where we have assumed that the total density matrix $\rho$ may initially be described by a product of system and environmental density matrix

$$
\begin{equation*}
\rho(0)=\rho_{\mathrm{S}}(0) \otimes \rho_{\mathrm{B}} \tag{2.36}
\end{equation*}
$$

with $\rho_{\mathrm{B}}$ as the density matrix of the environmental system. From equation (2.34) we obtain

$$
\begin{equation*}
\frac{\mathrm{d}}{\mathrm{~d} t} \rho_{\mathrm{S}}(t)=\mathcal{L} \rho_{\mathrm{S}}(t) \tag{2.37}
\end{equation*}
$$

the Markovian quantum master equation also called Lindblad equation. The quantity $\mathcal{L}$ is the generator of the quantum dynamical semigroup which can be constructed as

$$
\begin{equation*}
\mathcal{L} \rho_{\mathrm{S}}=-\mathrm{i}\left[\boldsymbol{H}, \rho_{\mathrm{S}}\right]+\sum_{k} \gamma_{k}\left(\boldsymbol{O}_{k} \rho_{\mathrm{S}} \boldsymbol{O}_{k}^{\dagger}-\frac{1}{2} \boldsymbol{O}_{k}^{\dagger} \boldsymbol{O}_{k} \rho_{\mathrm{S}}-\frac{1}{2} \rho_{\mathrm{S}} \boldsymbol{O}_{k}^{\dagger} \boldsymbol{O}_{k}\right) . \tag{2.38}
\end{equation*}
$$

Here, $\boldsymbol{H}$ describes the total system's Hamiltonian, the $\boldsymbol{O}_{k}$ are different operators in the reduced system's Liouville space and the $\gamma_{k}$ will take the role of damping parameters. The index $k$ counts from 1 to $\left(\operatorname{dim}\left(\mathcal{H}_{\mathrm{S}}\right)\right)^{2}-1$ if the Hilbert space $\mathcal{H}_{\mathrm{S}}$ is of finite dimension. We have also implied short correlation times in the environment. Introducing the dissipator

$$
\begin{equation*}
\mathcal{D}\left(\rho_{\mathrm{S}}\right) \equiv \sum_{k} \gamma_{k}\left(\boldsymbol{O}_{k} \rho_{\mathrm{S}} \boldsymbol{O}_{k}^{\dagger}-\frac{1}{2} \boldsymbol{O}_{k}^{\dagger} \boldsymbol{O}_{k} \rho_{\mathrm{S}}-\frac{1}{2} \rho_{\mathrm{S}} \boldsymbol{O}_{k}^{\dagger} \boldsymbol{O}_{k}\right) \tag{2.39}
\end{equation*}
$$

the Lindblad equation (2.37) takes the form

$$
\begin{equation*}
\frac{\mathrm{d}}{\mathrm{~d} t} \rho_{\mathrm{S}}(t)=-\mathrm{i}\left[\boldsymbol{H}, \rho_{\mathrm{S}}(t)\right]+\mathcal{D}\left(\rho_{\mathrm{S}}(t)\right) \tag{2.40}
\end{equation*}
$$

which resembles the Liouville equation

$$
\begin{equation*}
\frac{\mathrm{d}}{\mathrm{~d} t} \rho(t)=-\mathrm{i}[\boldsymbol{H}, \rho(t)] \tag{2.41}
\end{equation*}
$$

used to describe a closed system with Hamiltonian $\boldsymbol{H}$ and density operator $\rho$. Taking the Hermitian conjugate of equation (2.34) provides us with the adjoint propagator $V^{\dagger}\left(t, t_{0}\right)$ for which

$$
\begin{equation*}
\frac{\partial}{\partial t} V^{\dagger}\left(t, t_{0}\right)=V^{\dagger}\left(t, t_{0}\right) \mathcal{L}^{\dagger}=\mathcal{L}^{\dagger} V^{\dagger}\left(t, t_{0}\right) \tag{2.42}
\end{equation*}
$$

holds. Its key application is the transformation of observables from the Schrödinger picture into the Heisenberg picture via

$$
\begin{equation*}
\boldsymbol{O}_{\mathrm{H}}(t)=V^{\dagger}(t, 0) \boldsymbol{O}_{\mathrm{S}} \tag{2.43}
\end{equation*}
$$

Note that this does not contradict equation $(2.24)$ as we are focussing on an open system here. Differentiating (2.43) yields

$$
\begin{equation*}
\frac{\mathrm{d}}{\mathrm{~d} t} \boldsymbol{O}_{\mathrm{H}}(t)=V^{\dagger}(t, 0)\left(\mathcal{L}^{\dagger} \boldsymbol{O}_{\mathrm{S}}\right)=\mathcal{L}^{\dagger} V^{\dagger}(t, 0) \boldsymbol{O}_{\mathrm{S}}=\mathcal{L}^{\dagger} \boldsymbol{O}_{\mathrm{H}} \tag{2.44}
\end{equation*}
$$

with equation $(\overline{2.42})$. Here, we have used explicitly that the generator $\mathcal{L}^{\dagger}$ and the propagator $V^{\dagger}\left(t, t_{0}\right)$ commute which in general only applies if $\mathcal{L}$ is time independent, i.e.

$$
\begin{equation*}
\mathcal{L}(t)=\mathcal{L} \tag{2.45}
\end{equation*}
$$

A generalisation is possible, however not needed in this work and, hence, not presented. With the definition of the generator $\mathcal{L}$ in equation $(\overline{2.38)}$ we obtain from (2.44)

$$
\begin{align*}
\frac{\mathrm{d}}{\mathrm{~d} t} \boldsymbol{O}_{\mathrm{H}}(t)= & \mathrm{i}\left[\boldsymbol{H}, \boldsymbol{O}_{\mathrm{H}}(t)\right] \\
& +\sum_{k} \gamma_{k}\left(\boldsymbol{O}_{k}^{\dagger} \boldsymbol{O}_{\mathrm{H}}(t) \boldsymbol{O}_{k}-\frac{1}{2} \boldsymbol{O}_{\mathrm{H}}(t) \boldsymbol{O}_{k}^{\dagger} \boldsymbol{O}_{k}-\frac{1}{2} \boldsymbol{O}_{k}^{\dagger} \boldsymbol{O}_{k} \boldsymbol{O}_{\mathrm{H}}(t)\right) \tag{2.46}
\end{align*}
$$

which is called the adjoint quantum master equation.
An important special case of an open system in an environment is given by the damped harmonic oscillator. The reduced system $S$ is described by the ordinary harmonic oscillator, while the damping is obtained from the Lindblad formalism as introduced above. We take the Hamiltonian $\boldsymbol{H}$ to be

$$
\begin{equation*}
\boldsymbol{H} \propto \tilde{\omega} \boldsymbol{a}^{\dagger} \boldsymbol{a} \tag{2.47}
\end{equation*}
$$

with $\boldsymbol{a}^{\dagger}$ and $\boldsymbol{a}$ being bosonic creation and annihilation operators and $\tilde{\omega}$, the frequency. The quantum master equation then reads $[12,2,1,37]$

$$
\begin{align*}
\frac{\mathrm{d}}{\mathrm{~d} t} \rho_{\mathrm{S}}(t)= & -\mathrm{i}\left[\boldsymbol{H}, \rho_{\mathrm{S}}(t)\right] \\
& +\tilde{\gamma}\left(N_{\mathrm{Q}}+1\right)\left(\boldsymbol{a} \rho_{\mathrm{S}}(t) \boldsymbol{a}^{\dagger}-\frac{1}{2} \boldsymbol{a}^{\dagger} \boldsymbol{a} \rho_{\mathrm{S}}(t)-\frac{1}{2} \rho_{\mathrm{S}}(t) \boldsymbol{a}^{\dagger} \boldsymbol{a}\right)  \tag{2.48}\\
& +\tilde{\gamma} N_{\mathrm{Q}}\left(\boldsymbol{a}^{\dagger} \rho_{\mathrm{S}}(t) \boldsymbol{a}-\frac{1}{2} \boldsymbol{a} \boldsymbol{a}^{\dagger} \rho_{\mathrm{S}}(t)-\frac{1}{2} \rho_{\mathrm{S}}(t) \boldsymbol{a} \boldsymbol{a}^{\dagger}\right)
\end{align*}
$$

i.e. the operators classifying the dissipation are

$$
\begin{align*}
\boldsymbol{O}_{k} & =\boldsymbol{a}  \tag{2.49a}\\
\boldsymbol{O}_{k}^{\dagger} & =\boldsymbol{a}^{\dagger} \tag{2.49b}
\end{align*}
$$

A complete derivation can be found in 35 . There is only one parameter $\tilde{\gamma}$ left of which the size describes the oscillator's damping. The quantity

$$
\begin{equation*}
N_{\mathrm{Q}}=\left(\exp \left(\frac{\tilde{\omega}}{k_{\mathrm{B}} T}\right)-1\right)^{-1} \tag{2.50}
\end{equation*}
$$

representing the Bose-Einstein statistics is used to classify the thermal reservoir providing the mean number of energy quanta in the mode corresponding to the frequency $\tilde{\omega}$. As usual, $k_{\mathrm{B}}$ is the Boltzmann constant and $T$, the temperature. When comparing equation (2.37) with equation $(\overline{2.46})$ the form of the adjoint quantum master equation for the damped harmonic oscillator becomes evident

$$
\begin{align*}
\frac{\mathrm{d}}{\mathrm{~d} t} \boldsymbol{O}_{\mathrm{H}}(t)= & \mathrm{i}\left[\boldsymbol{H}, \boldsymbol{O}_{\mathrm{H}}(t)\right] \\
& +\tilde{\gamma}\left(N_{\mathrm{Q}}+1\right)\left(\boldsymbol{a}^{\dagger} \boldsymbol{O}_{\mathrm{H}}(t) \boldsymbol{a}-\frac{1}{2} \boldsymbol{a}^{\dagger} \boldsymbol{a} \boldsymbol{O}_{\mathrm{H}}(t)-\frac{1}{2} \boldsymbol{O}_{\mathrm{H}}(t) \boldsymbol{a}^{\dagger} \boldsymbol{a}\right)  \tag{2.51}\\
& +\tilde{\gamma} N_{\mathrm{Q}}\left(\boldsymbol{a} \boldsymbol{O}_{\mathrm{H}}(t) \boldsymbol{a}^{\dagger}-\frac{1}{2} \boldsymbol{a} \boldsymbol{a}^{\dagger} \boldsymbol{O}_{\mathrm{H}}(t)-\frac{1}{2} \boldsymbol{O}_{\mathrm{H}}(t) \boldsymbol{a} \boldsymbol{a}^{\dagger}\right)
\end{align*}
$$

which, taking the expectation value, yields

$$
\begin{align*}
\frac{\mathrm{d}}{\mathrm{~d} t}\left\langle\boldsymbol{O}_{\mathrm{H}}\right\rangle(t)= & \mathrm{i}\left\langle\left[\boldsymbol{H}, \boldsymbol{O}_{\mathrm{H}}(t)\right]\right\rangle \\
& +\tilde{\gamma}\left(N_{\mathrm{Q}}+1\right)\left\langle\boldsymbol{a}^{\dagger} \boldsymbol{O}_{\mathrm{H}}(t) \boldsymbol{a}-\frac{1}{2} \boldsymbol{a}^{\dagger} \boldsymbol{a} \boldsymbol{O}_{\mathrm{H}}(t)-\frac{1}{2} \boldsymbol{O}_{\mathrm{H}}(t) \boldsymbol{a}^{\dagger} \boldsymbol{a}\right\rangle  \tag{2.52}\\
& +\tilde{\gamma} N_{\mathrm{Q}}\left\langle\boldsymbol{a} \boldsymbol{O}_{\mathrm{H}}(t) \boldsymbol{a}^{\dagger}-\frac{1}{2} \boldsymbol{a} \boldsymbol{a}^{\dagger} \boldsymbol{O}_{\mathrm{H}}(t)-\frac{1}{2} \boldsymbol{O}_{\mathrm{H}}(t) \boldsymbol{a} \boldsymbol{a}^{\dagger}\right\rangle
\end{align*}
$$

The two master equations $(2.48)$ and $(2.51)$ are referred to as the (adjoint) quantum optical master equation as the interaction of matter with electromagnetic radiation is modelled this way.

### 2.5 Fermi's Golden Rule

One of the most important results in first order perturbation theory is Fermi's golden rule. In this chapter we shall sketch its derivation based on references [4, 5, 41, 42, 13] and discuss key results.

We investigate a Hamiltonian

$$
\begin{equation*}
\boldsymbol{H}(t)=\boldsymbol{H}_{0}+\tilde{\boldsymbol{V}}(t) \tag{2.53}
\end{equation*}
$$

where $\boldsymbol{H}_{0}$ describes the unperturbed system of which we know the eigenenergies $E_{n}$ and eigenvectors $\left|\phi_{n}\right\rangle$ satisfying

$$
\begin{equation*}
\boldsymbol{H}_{0}\left|\phi_{n}\right\rangle=E_{n}\left|\phi_{n}\right\rangle . \tag{2.54}
\end{equation*}
$$

The perturbation $\boldsymbol{V}(t)$ is given by

$$
\begin{equation*}
\tilde{\boldsymbol{V}}(t)=\lambda \Theta(t) \boldsymbol{V}(t) \tag{2.55}
\end{equation*}
$$

with $\Theta$ as the Heaviside function and a real parameter

$$
\begin{equation*}
0<\lambda \ll 1, \tag{2.56}
\end{equation*}
$$

i.e. there is only a slight perturbation which vanishes for negative times $t$. The Schrödinger equation for positive times then reads

$$
\begin{equation*}
\mathrm{i} \frac{\mathrm{~d}}{\mathrm{~d} t}|\psi(t)\rangle=\left(\boldsymbol{H}_{0}+\lambda \boldsymbol{V}(t)\right)|\psi(t)\rangle . \tag{2.57}
\end{equation*}
$$

Initially the system shall be in a stationary state $\left|\phi_{i}\right\rangle$ with eigenenergy $E_{i}$, thus

$$
\begin{equation*}
|\psi(t=0)\rangle=\left|\phi_{i}\right\rangle . \tag{2.58}
\end{equation*}
$$

We are interested in the probability $P_{i, f}$ describing the system reaching the final state $\left|\phi_{f}\right\rangle$ while starting in its initial state $\left|\phi_{i}\right\rangle$ which is calculated via

$$
\begin{equation*}
P_{i, f}=\left|\left\langle\phi_{f} \mid \psi(t)\right\rangle\right|^{2} . \tag{2.59}
\end{equation*}
$$

We first decompose the state vector $|\psi(t)\rangle$ into a sum of the eigenvectors $\left|\phi_{n}\right\rangle$. Having equation (2.56) in mind and realising that the eigenvectors $\left|\phi_{n}\right\rangle$ form a basis, a useful ansatz proves to be

$$
\begin{equation*}
|\psi(t)\rangle=\sum_{n} a_{n}(t) \mathrm{e}^{-\mathrm{i} E_{n} t}\left|\phi_{n}\right\rangle \tag{2.60}
\end{equation*}
$$

with complex coefficients $a_{n}(t)$ which, when substituted into the Schrödinger equation (2.57), yields

$$
\begin{equation*}
\mathrm{i} \frac{\mathrm{~d}}{\mathrm{~d} t} a_{n}(t)=\lambda \sum_{k} \mathrm{e}^{\mathrm{i} \omega_{n, k} t} \boldsymbol{V}_{n, k}(t) a_{k}(t) . \tag{2.61}
\end{equation*}
$$

We have introduced the Bohr frequency

$$
\begin{equation*}
\omega_{n, k}=E_{n}-E_{k} \tag{2.62}
\end{equation*}
$$

and the matrix elements of the perturbation operator

$$
\begin{equation*}
\boldsymbol{V}_{n, k}(t)=\left\langle\phi_{n}\right| \boldsymbol{V}(t)\left|\phi_{k}\right\rangle . \tag{2.63}
\end{equation*}
$$

In general equation $(2.61)$ cannot be solved analytically. Taking again the relation $(2.56)$ into account, we approximate the coefficients $a_{n}(t)$ by a power series

$$
\begin{equation*}
a_{n}(t)=\sum_{j \geq 0} \lambda^{j} a_{n}^{(j)}(t) \tag{2.64}
\end{equation*}
$$

in hopes of convergence. Substituting into equation (2.61) yields

$$
\begin{align*}
\mathrm{i} \frac{\mathrm{~d}}{\mathrm{~d} t} a_{n}^{(0)}(t) & =0  \tag{2.65a}\\
\mathrm{i} \frac{\mathrm{~d}}{\mathrm{~d} t} a_{n}^{(j>0)}(t) & =\sum_{k} \mathrm{e}^{\mathrm{i} \omega_{n, k} t} \boldsymbol{V}_{n, k}(t) a_{k}^{(j-1)} . \tag{2.65b}
\end{align*}
$$

Using the initial condition (2.58) we derive

$$
\begin{equation*}
a_{n}(t=0)=\delta_{n, i} \tag{2.66}
\end{equation*}
$$

which means for equation (2.64)

$$
\begin{align*}
a_{n}^{(0)}(t=0) & =\delta_{n, i}  \tag{2.67a}\\
a_{n}^{(j>0)}(t=0) & =0 . \tag{2.67b}
\end{align*}
$$

The first two coefficients are thus obtained from the recursive equation (2.65) as

$$
\begin{align*}
& a_{n}^{(0)}(t)=\delta_{n, i}  \tag{2.68a}\\
& a_{n}^{(1)}(t)=-\mathrm{i} \int_{0}^{t} \mathrm{e}^{\mathrm{i} \omega_{n, i} t^{\prime}} \boldsymbol{V}_{n, i}\left(t^{\prime}\right) \mathrm{d} t^{\prime} . \tag{2.68b}
\end{align*}
$$

Approximating the $a_{n}(t)$ only to the first order the transition probability is given by

$$
\begin{equation*}
P_{i, f}(t)=\lambda^{2}\left|a_{f}^{(1)}(t)\right|^{2}=\left|\int_{0}^{t} \mathrm{e}^{\mathrm{i} \omega_{f, i} t^{\prime}} V_{f, i}\left(t^{\prime}\right) \mathrm{d} t^{\prime}\right|^{2} \tag{2.69}
\end{equation*}
$$

where we have absorbed the parameter lambda into the perturbation $\boldsymbol{V}(t)$. If it is constant, i.e.

$$
\begin{equation*}
\boldsymbol{V}(t)=V, \tag{2.70}
\end{equation*}
$$

equation (2.69) takes the much easier form

$$
\begin{equation*}
P_{i, f}(t)=\left(\frac{\sin \left(\omega_{i, f} t / 2\right)}{\omega_{i, f} / 2}\right)^{2}\left|V_{i, f}\right|^{2} \tag{2.71}
\end{equation*}
$$

which for large times $t$ converges to

$$
\begin{equation*}
P_{i, f}(t)=t 2 \pi \delta\left(E_{f}-E_{i}\right)\left|V_{i, f}\right|^{2} . \tag{2.72}
\end{equation*}
$$

Its time derivative

$$
\begin{equation*}
\tilde{\Gamma}_{i, f}=2 \pi \delta\left(E_{f}-E_{i}\right)\left|V_{i, f}\right|^{2} \tag{2.73}
\end{equation*}
$$

describes the transition probability per unit of time. To obtain the complete transition probability per unit of time $\Gamma_{i, f}$ for a continuous system we have to calculate (2.73) for any energy $E$. Therefore, we make use of the density of states $\rho\left(E_{i}\right)$ which yields

$$
\begin{equation*}
\Gamma_{i, f}(t)=\int_{E \in \delta E_{f}} \tilde{\Gamma}_{i, f} \rho(E) \mathrm{d} E=2 \pi \rho\left(E_{i}\right)\left|V_{i, f}\right|^{2} \tag{2.74}
\end{equation*}
$$

where $\delta E_{f}$ describes a small interval around the energy $E_{f}$. Equation 2.74 goes by the name of Fermi's golden rule.

We note that the delta distribution in equation (2.73) provides the energy conservation. To a first order approximation we obtain the probability for a system to reach a final state $\left|\phi_{f}\right\rangle$ starting in the initial state $\left|\phi_{i}\right\rangle$. More importantly, if either the corresponding density of states $\rho\left(E_{i}\right)$ vanishes or the perturbation does not allow the particular transition, we do not expect any measurements in a corresponding experiment.

However, Fermi's golden rule $(\overline{2.74})$ is based on assumption $(\overline{2.70})$ which calls for a time independent perturbation. Clearly equation (2.70) cannot hold in general. For instance, it does not apply to any equilibrium. As the parameter $\lambda$ according to equation (2.56) is supposed to be very small Fermi's golden rule may still serve as a proper approximation.

### 2.6 Green's Functions

Green's functions are a powerful tool when it comes to describing properties of a quantum system. We will only focus on concepts necessary for this project thus presenting only a fraction of the complete theory. Further information can be found in reference [30].

Let $\boldsymbol{g}_{1}$ and $\boldsymbol{g}_{2}$ be bosonic operators. The conventional retarded Green's function is defined via

$$
\begin{equation*}
G_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}^{\text {ret }}\left(t_{\text {in }}, t_{\text {out }}\right)=-\mathrm{i}\left\langle\left[\boldsymbol{g}_{2}\left(t_{\text {out }}\right), \boldsymbol{g}_{1}\left(t_{\text {in }}\right)\right]\right\rangle \Theta\left(t_{\text {out }}-t_{\text {in }}\right), \tag{2.75}
\end{equation*}
$$

i.e. it provides a description of how the observables $\boldsymbol{g}_{2}$ at time $t_{\text {out }}$ is affected by a perturbation of the observable $\boldsymbol{g}_{1}$ at time $t_{\mathrm{in}}$. Of equal importance is the spectral density $S_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}$ which is defined by

$$
\begin{equation*}
G_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}^{\mathrm{ret}}\left(t_{\mathrm{in}}, \omega_{\text {out }}\right)=\lim _{\delta>0}\left(\int_{-\infty}^{+\infty} \frac{S_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}\left(t_{\text {in }}, \omega_{\text {out }}+\mathrm{i} \delta\right)}{\omega_{\text {out }}+\mathrm{i} \delta-x} \mathrm{~d} x\right) \tag{2.76}
\end{equation*}
$$

and for which

$$
\begin{equation*}
S_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}\left(t_{\text {in }}, \omega_{\text {out }}\right)=-\frac{1}{\pi} \operatorname{Im}\left(G_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}^{\mathrm{ret}}\left(t_{\text {in }}, \omega_{\text {out }}\right)\right) \tag{2.77}
\end{equation*}
$$

holds. Here, we have also introduced the Fourier transform of the Green's function (2.75) via

$$
\begin{equation*}
G_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}^{\text {ret }}\left(t_{\text {in }}, \omega_{\text {out }}\right)=\lim _{\delta \searrow 0} \frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} G_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}^{\text {ret }}\left(t_{\text {in }}, t_{\text {out }}\right) \mathrm{e}^{\mathrm{i}\left(\omega_{\text {out }}+\mathrm{i} \delta\right) t_{\text {out }} \mathrm{d} t_{\text {out }} .} \tag{2.78}
\end{equation*}
$$

To simplify the notation, the limit in the above equation is often omitted. Differentiating equation (2.75) we obtain the equation of motion for Green's functions

$$
\begin{equation*}
\frac{\mathrm{d}}{\mathrm{~d} t_{\text {out }}} G_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}^{\mathrm{ret}}\left(t_{\text {in }}, t_{\text {out }}\right)=\mathrm{i} G_{\left[\boldsymbol{H}, \boldsymbol{g}_{2}\right], \boldsymbol{g}_{1}}^{\mathrm{ret}}\left(t_{\text {in }}, t_{\text {out }}\right)-\mathrm{i}\left\langle\left[\boldsymbol{g}_{2}\left(t_{\text {in }}\right), \boldsymbol{g}_{1}\left(t_{\text {in }}\right)\right]\right\rangle \delta\left(t_{\text {out }}-t_{\text {in }}\right), \tag{2.79}
\end{equation*}
$$

where we have made use of the Heisenberg equation of motion (2.29). The time evolution for times

$$
\begin{equation*}
t_{\mathrm{out}}>t_{\mathrm{in}} \tag{2.80}
\end{equation*}
$$

is then given by

$$
\begin{equation*}
\frac{\mathrm{d}}{\mathrm{~d} t_{\text {out }}} G_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}^{\mathrm{ret}}\left(t_{\mathrm{in}}, t_{\text {out }}\right)=\mathrm{i} G_{\left[\boldsymbol{H}, \boldsymbol{g}_{2}\right], \boldsymbol{g}_{1}}^{\mathrm{ret}}\left(t_{\text {in }}, t_{\text {out }}\right) . \tag{2.81}
\end{equation*}
$$

Note that, because the expectation value in the definition of the Green's function in equation (2.75) is linear,

$$
\begin{equation*}
G_{\alpha \boldsymbol{g}_{2}+\beta \boldsymbol{g}_{3}, \boldsymbol{g}_{1}}^{\mathrm{ren}}\left(t_{\mathrm{in}}, t_{\mathrm{out}}\right)=\alpha G_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}^{\mathrm{ret}}\left(t_{\mathrm{in}}, t_{\mathrm{out}}\right)+\beta G_{\boldsymbol{g}_{3}, \boldsymbol{g}_{1}}^{\mathrm{ret}}\left(t_{\mathrm{in}}, t_{\mathrm{out}}\right) \tag{2.82}
\end{equation*}
$$

holds for operators $\boldsymbol{g}_{1}, \boldsymbol{g}_{2}$ and $\boldsymbol{g}_{3}$ and complex constants $\alpha$ and $\beta$. When investigating a system in equilibrium, $G_{g_{2}, g_{1}}^{\text {ret }}\left(t_{\text {in }}, t_{\text {out }}\right)$ should only depend on the time difference, such as

$$
\begin{equation*}
G_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}^{\text {ret }}\left(t_{\text {in }}, t_{\text {out }}\right)=G_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}^{\text {ret }}\left(0, t_{\text {out }}-t_{\text {in }}\right)=: G_{\boldsymbol{g}_{2}, \boldsymbol{g}_{1}}^{\text {ret }}\left(t_{\text {out }}-t_{\text {in }}\right) . \tag{2.83}
\end{equation*}
$$

However for non-equilibrium systems this relation does not generally apply leading to the definition of further Green's functions.

Conveniently, the retarded Green's function for triplons reads

$$
\begin{align*}
& G_{\alpha \alpha, k}^{\text {ret }}\left(t_{\text {in }}, t_{\text {out }}\right)=G_{\boldsymbol{t}_{\alpha, k}, t_{\alpha, k}^{\dagger}}^{\text {ret }} \\
&=-\mathrm{i}\left\langle\left[\boldsymbol{t}_{\alpha, k}\left(t_{\text {out }}\right), t_{\text {out }}\right)\right.  \tag{2.84}\\
&\left.\left.\boldsymbol{t}_{\alpha, k}^{\dagger}\left(t_{\text {in }}\right)\right]\right\rangle \Theta\left(t_{\text {out }}-t_{\text {in }}\right) .
\end{align*}
$$

Note that we are investigating triplons in momentum space here. We present the corresponding Fourier transform in section 3.2. The Fourier transform of the absolute retarded Green's function $G_{\alpha \alpha, k}^{\mathrm{ret}}\left(t_{\text {in }}, t_{\text {out }}\right)(2.84)$ is then computed via

$$
\begin{equation*}
G_{\alpha \alpha, k}^{\text {ret }}\left(t_{\text {in }}, \omega_{\text {out }}\right)=\frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} G_{\alpha \alpha, k}^{\text {ret }}\left(t_{\text {in }}, t_{\text {out }}\right) \mathrm{e}^{\mathrm{i} \omega_{\text {out }} t_{\text {out }} \mathrm{d} t_{\text {out }} .} \tag{2.85}
\end{equation*}
$$

We introduce the average time

$$
\begin{equation*}
\bar{t}=\frac{t_{\mathrm{in}}+t_{\mathrm{out}}}{2} \tag{2.86}
\end{equation*}
$$

and the time difference

$$
\begin{equation*}
t_{\text {diff }}=t_{\text {out }}-t_{\text {in }} \tag{2.87}
\end{equation*}
$$

which allows us to define the relative Green's function

$$
\begin{equation*}
G_{\alpha \alpha, k}^{\mathrm{rel}}\left(t_{\mathrm{in}}, t_{\mathrm{diff}}\right)=G_{\alpha \alpha, k}^{\mathrm{ret}}\left(t_{\mathrm{in}}, t_{\mathrm{in}}+t_{\mathrm{diff}}\right) \tag{2.88}
\end{equation*}
$$

and the time average Green's function

$$
\begin{equation*}
\tilde{G}_{\alpha \alpha, k}\left(\bar{t}, t_{\text {diff }}\right)=G_{\alpha \alpha, k}^{\mathrm{ret}}\left(\bar{t}-\frac{t_{\text {diff }}}{2}, \bar{t}+\frac{t_{\text {diff }}}{2}\right) . \tag{2.89}
\end{equation*}
$$

The respective Fourier transforms are given by

$$
\begin{align*}
G_{\alpha \alpha, k}^{\mathrm{rel}}\left(\omega_{\text {diff }}\right) & =\frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} G_{\alpha \alpha, k}^{\mathrm{rel}}\left(t_{\text {diff }}\right) \mathrm{e}^{\mathrm{i} \omega_{\text {diff }} t_{\text {diff }} \mathrm{d} t_{\text {diff }}}  \tag{2.90}\\
\tilde{G}_{\alpha \alpha, k}\left(\bar{t}, \omega_{\text {diff }}\right) & =\frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} \tilde{G}_{\alpha \alpha, k}\left(\bar{t}, t_{\text {diff }}\right) \mathrm{e}^{\mathrm{i} \omega_{\text {diff }} t_{\text {diff }}} \mathrm{d} t_{\text {diff }} \tag{2.91}
\end{align*}
$$

Both of the above defined Green's functions serve different purposes. The relative Green's function $(\overline{2.88)}$ omits the constant part of the retarded Green's function ( $\overline{2.84})$ making its Fourier transform easier to calculate and interpret. While this is a rather technical aspect, the meaning of the time average Green's function (2.89) goes somewhat deeper. Here we investigate whether the time difference is the sole parameter the Green's functions depend on as is the case in equilibrium systems. If so, there should be no difference in both Green's functions or their respective Fourier spectra, $(\overline{2.90})$ and $(\overline{2.91})$, whatsoever. Finally, we define the average Green's function

$$
\begin{equation*}
\bar{G}_{\alpha \alpha, k}\left(t_{\max }, t\right)=\frac{1}{t_{\max }-t} \int_{0}^{t_{\max }-t} G_{\alpha \alpha, k}^{\mathrm{ret}}\left(t^{\prime}, t^{\prime}+t\right) \mathrm{d} t^{\prime} \tag{2.92}
\end{equation*}
$$

and its Fourier transform

$$
\begin{equation*}
\bar{G}_{\alpha \alpha, k}\left(t_{\max }, \omega\right)=\frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} \bar{G}_{\alpha \alpha, k}\left(t_{\max }, t\right) \mathrm{e}^{\mathrm{i} \omega t} \mathrm{~d} t \tag{2.93}
\end{equation*}
$$

which provide a long range description of the response to a perturbation. We will list all the Green's functions of interest in section 4.3 where they will be discussed.

### 2.7 The Lindemann Criterion

In 1910 Frederick Lindemann presented a method to calculate the maximum displacement $x_{\text {max }}$ of atoms in solids at which the probe starts to melt [14]. He found

$$
\begin{equation*}
x_{\max } \gtrsim 0.2 \ldots 0.3 \cdot l \tag{2.94}
\end{equation*}
$$

where $l$ describes the lattice constant. Equation $(\overline{2.94})$ is reasonable hence a displacement of the order of a solid's lattice constant means that its previously translational invariant microscopic structure collapses. This particular property, however, characterizes liquids which in turn confirm Lindemann's statement.
Lindemann's criterion provides a simple mechanism to estimate the corresponding (quasi) particle density. We, to a first order approximation, investigate again the harmonic oscillator

$$
\begin{equation*}
\boldsymbol{H}=\hbar \tilde{\omega}\left(\boldsymbol{a}^{\dagger} \boldsymbol{a}+\frac{1}{2}\right) \tag{2.95}
\end{equation*}
$$

similar to equation (2.47) with $\boldsymbol{a}^{\dagger}$ and $\boldsymbol{a}$ as bosonic creation and annihilation operators respectively. They are given by

$$
\begin{align*}
\boldsymbol{a}^{\dagger} & =\sqrt{\frac{m \tilde{\omega}}{2 \hbar}}\left(\boldsymbol{x}-\mathrm{i} \frac{\boldsymbol{p}}{m \tilde{\omega}}\right)  \tag{2.96a}\\
\boldsymbol{a} & =\sqrt{\frac{m \tilde{\omega}}{2 \hbar}}\left(\boldsymbol{x}+\mathrm{i} \frac{\boldsymbol{p}}{m \tilde{\omega}}\right) . \tag{2.96b}
\end{align*}
$$

Here, $\boldsymbol{x}$ is the position operator, $\boldsymbol{p}$ the momentum operator, $m$ the atomic mass and $\tilde{\omega}$ the frequency. We observe the most trivial case of maximum displacement and zero momentum, i.e.

$$
\begin{align*}
& \boldsymbol{x}=x_{\text {max }}  \tag{2.97a}\\
& \boldsymbol{p}=0 \tag{2.97b}
\end{align*}
$$

which yields

$$
\begin{equation*}
\left\langle\boldsymbol{a}^{\dagger} \boldsymbol{a}\right\rangle=\frac{m \tilde{\boldsymbol{\omega}}}{2 \hbar} x_{\max }^{2} \tag{2.98}
\end{equation*}
$$

Approximating the frequency $\tilde{\omega}$ with the Debye frequency

$$
\begin{equation*}
\omega_{\mathrm{D}}=\frac{\Theta_{\mathrm{D}} k_{\mathrm{B}}}{\hbar}=\tilde{\omega} \tag{2.99}
\end{equation*}
$$

where $\Theta_{\mathrm{D}}$ is the Debye temperature and $k_{\mathrm{B}}$ the Boltzmann constant we obtain

$$
\begin{equation*}
\left\langle\boldsymbol{a}^{\dagger} \boldsymbol{a}\right\rangle=\frac{m \Theta_{\mathrm{D}} k_{\mathrm{B}}}{2 \hbar^{2}} x_{\max }^{2} . \tag{2.100}
\end{equation*}
$$

For common values of Debye temperature $\Theta_{\mathrm{D}}$, mass $m$ and lattice constant $l$

$$
\begin{align*}
\Theta_{\mathrm{D}} & \approx 100 \mathrm{~K}-1000 \mathrm{~K}  \tag{2.101a}\\
m & \approx 1 \mathrm{u}-100 \mathrm{u}  \tag{2.101b}\\
l & \approx 100 \mathrm{pm} \tag{2.101c}
\end{align*}
$$

and using (2.94) we receive

$$
\begin{equation*}
\left\langle\boldsymbol{a}^{\dagger} \boldsymbol{a}\right\rangle \lesssim 10 \tag{2.102}
\end{equation*}
$$

i.e. there is an upper limit to the (quasi) particle density.

### 2.8 The Damped Driven Harmonic Oscillator

The equation of motion describing the position $x$ of an ordinary classical harmonic oscillator reads

$$
\begin{equation*}
\ddot{x}(t)+\omega_{0}^{2} x(t)=0 \tag{2.103}
\end{equation*}
$$

where $\omega_{0}$ is the oscillation frequency. This system can be easily enhanced with a damping, yielding

$$
\begin{equation*}
\ddot{x}(t)+\gamma \dot{x}(t)+\omega_{0}^{2} x(t)=0 \tag{2.104}
\end{equation*}
$$

and, furthermore, a driving so we obtain [29]

$$
\begin{equation*}
\ddot{x}(t)+\gamma \dot{x}(t)+\omega_{0}^{2} x(t)=\frac{1}{m x_{0}} E(t) . \tag{2.105}
\end{equation*}
$$

Here $\gamma$ is the damping parameter of the system under consideration and $m$ the atomic. The displacement $x_{0}$ is as a first approximation determined by the maximum displacement of the undamped harmonic oscillator, hence

$$
\begin{equation*}
x_{0}=\sqrt{\frac{\hbar}{2 m \omega_{0}}} \tag{2.106}
\end{equation*}
$$

c.f. section 3.5. The only purpose of these parameters is to provide the proper dimension as $E(t)$ describes the energy obtained from driving with an external electric field $\tilde{E}(t)$. Equation (2.105) is the most general form of the equation of motion for a driven harmonic oscillator with linear damping.

A periodic driving like

$$
\begin{equation*}
E(t)=a \cos (\Omega t) \tag{2.107}
\end{equation*}
$$

makes for an important special case we will investigate further. The parameters $a$ and $\Omega$ describe the amplitude and the frequency of the driving. Equation (2.105) now reads

$$
\begin{equation*}
\ddot{x}(t)+\gamma \dot{x}(t)+\omega_{0}^{2} x(t)-\frac{1}{m x_{0}} a \cos (\Omega t)=0 . \tag{2.108}
\end{equation*}
$$

Introducing the complex quantity

$$
\begin{equation*}
z(t)=x(t)+\mathrm{i} y(t) \tag{2.109}
\end{equation*}
$$

we solve

$$
\begin{equation*}
\ddot{z}(t)+\gamma \dot{z}(t)+\omega_{0}^{2} z(t)=\frac{a}{m x_{0}} \mathrm{e}^{\mathrm{i} \Omega t}=\frac{a}{m x_{0}} \cos (\Omega t)+\mathrm{i} \frac{a}{m x_{0}} \sin (\Omega t) \tag{2.110}
\end{equation*}
$$

instead of equation (2.108) knowing only the real part of $z(t)$ will be of interest. We make use of the ansatz

$$
\begin{equation*}
z(t)=z_{\max } \mathrm{e}^{\mathrm{i} \Omega t} \tag{2.111}
\end{equation*}
$$

which yields when substituted into equation (2.110)

$$
\begin{equation*}
z_{\max }=-\frac{a}{m x_{0}} \frac{1}{\left(\Omega^{2}-\omega_{0}^{2}\right)-\mathrm{i} \gamma \Omega} . \tag{2.112}
\end{equation*}
$$

We define the phase $\varphi$, such as

$$
\begin{equation*}
z_{\max }=\left|z_{\max }\right| \mathrm{e}^{\mathrm{i} \varphi} \tag{2.113}
\end{equation*}
$$

so that we obtain from equation (2.111)

$$
\begin{equation*}
z(t)=\left|z_{\max }\right| \mathrm{e}^{\mathrm{i} \varphi} \mathrm{e}^{\mathrm{i} \Omega t} . \tag{2.114}
\end{equation*}
$$

From equation (2.109) we then derive

$$
\begin{equation*}
x(t)=\left|z_{\max }\right| \cos (\Omega t+\varphi) . \tag{2.115}
\end{equation*}
$$

Clearly, the maximum position is given by

$$
\begin{equation*}
x_{\max }=\left|z_{\max }\right|=\frac{a}{m x_{0}} \frac{1}{\sqrt{\left(\Omega^{2}-\omega_{0}^{2}\right)^{2}+\gamma^{2} \Omega^{2}}} . \tag{2.116}
\end{equation*}
$$

If the driving frequency $\Omega$ equals the oscillator's frequency $\omega_{0}$, i.e.

$$
\begin{equation*}
\Omega=\omega_{0} \tag{2.117}
\end{equation*}
$$

equation (2.116) reads

$$
\begin{equation*}
x_{\max }=\frac{a}{m x_{0} \gamma \omega_{0}} . \tag{2.118}
\end{equation*}
$$

In terms of quantization we learn by substituting this result in equation (2.98) from the previous section, where $\tilde{\omega}=\omega_{0}$, also using equation (2.106)

$$
\begin{equation*}
\left\langle\boldsymbol{a}^{\dagger} \boldsymbol{a}\right\rangle=\left(\frac{a}{\gamma}\right)^{2} \frac{1}{\hbar^{2}} \propto\left(\frac{a}{\gamma}\right)^{2}, \tag{2.119}
\end{equation*}
$$

i.e. the number or density of quasi particles in the quantum system is primarily determined by the square of the ratio of driving amplitude and damping parameter.

## 3 Model

As mentioned in the introduction we wish to create a system that can be driven into arbitrary quasi stationary states. Therefore, the corresponding model cannot be trivial but at the same time should also not be too complicated to simulate. We, thus, imagined the system as follows: A dimerized spin $1 / 2$ chain will serve as the principal model to derive new effects from. It is coupled to a phonon system of which the primary use is to be a mediator. The phonon system, on the one hand, is coupled to a laser which provides the necessary excitation to be transferred to the spin system. On the other hand, there exists a coupling to a bath in order to avoid overheating. A sketch of this system can be found in figure 3.1.


Figure 3.1: Schematic picture of the model under investigation

The rest of this chapter is structured as follows. First we shall set up a proper Hamiltonian describing the model in more detail. Next, as the sole model will be too complicated to solve, we explain the approximations made to obtain results. We will define observables and derive the corresponding equations of motion. Finally, we will discuss the parameters describing the model trying to deduce reasonable reasonable orders of magnitude for each one.

### 3.1 Hamiltonian

The total Hamiltonian $\boldsymbol{H}$ is composed of four different parts each describing a different subsystem or an interaction. The spin and phonon system are characterised by $\boldsymbol{H}_{\mathrm{S}}$ and $\boldsymbol{H}_{\mathrm{P}}$ respectively. The Hamiltonian $\boldsymbol{H}_{\mathrm{SP}}$ provides the coupling of those subsystems, while $\boldsymbol{H}_{\mathrm{L}}$ contains the coupling of phonon system and laser field. Hence, the total Hamiltonian is

$$
\begin{equation*}
\boldsymbol{H}=\boldsymbol{H}_{\mathrm{S}}+\boldsymbol{H}_{\mathrm{P}}+\boldsymbol{H}_{\mathrm{SP}}+\boldsymbol{H}_{\mathrm{L}} . \tag{3.1}
\end{equation*}
$$

The phonon system's coupling to a bath is not explicitly mentioned but will be introduced via the Lindblad formalism.

The Hamiltonian for the dimerized spin chain reads

$$
\begin{equation*}
\boldsymbol{H}_{\mathrm{S}}=\sum_{i} J \overrightarrow{\boldsymbol{S}}_{i}^{1} \cdot \overrightarrow{\boldsymbol{S}}_{i}^{2}+J^{\prime} \overrightarrow{\boldsymbol{S}}_{i}^{2} \cdot \overrightarrow{\boldsymbol{S}}_{i+1}^{1} \tag{3.2}
\end{equation*}
$$

with $J$ being the coupling strength of the two spins $\overrightarrow{\boldsymbol{S}}_{i}^{1}$ and $\overrightarrow{\boldsymbol{S}}_{i}^{2}$ forming the $i^{\text {th }}$ dimer and $J^{\prime}$ the inter dimer coupling strength. As we describe an antiferromagnetic system

$$
\begin{equation*}
0<J^{\prime}<J \tag{3.3}
\end{equation*}
$$

and the index $i$ takes values of

$$
\begin{equation*}
i \in\{1, \ldots, N\} \tag{3.4}
\end{equation*}
$$

where $N$ is the number of dimers. We also take periodic boundary conditions to simplify the model.

The phonon system is described by an ordinary harmonic oscillator, hence

$$
\begin{equation*}
\boldsymbol{H}_{\mathrm{P}}=\sum_{k \in \mathrm{BZ}} \omega_{\mathrm{B}}(k) \boldsymbol{b}_{k}^{\dagger} \boldsymbol{b}_{k} \tag{3.5}
\end{equation*}
$$

where "BZ" means the Brillouin zone, i.e. the interval from $-\pi$ to $\pi$. We have introduced the bosonic creation and annihilation operators $\boldsymbol{b}$ and $\boldsymbol{b}^{\dagger}$ for different modes

$$
\begin{equation*}
k_{i}=-\pi+2 \pi \frac{i}{N} . \tag{3.6}
\end{equation*}
$$

Most often the index $i$ will be omitted. The quantity $\omega_{\mathrm{B}}$ is the dispersion relation of this bosonic phonon system.

As for the spin-phonon coupling the phonon displacement $\left(\boldsymbol{b}_{i}+\boldsymbol{b}_{i}^{\dagger}\right)$ is to couple directly to the spins with coupling constant $g$, yielding

$$
\begin{equation*}
\boldsymbol{H}_{\mathrm{SP}}=\sum_{i} g\left(\boldsymbol{b}_{i}+\boldsymbol{b}_{i}^{\dagger}\right)\left(\overrightarrow{\boldsymbol{S}}_{i}^{1} \cdot \overrightarrow{\boldsymbol{S}}_{i}^{2}-\left\langle\overrightarrow{\boldsymbol{S}}_{i}^{1} \cdot \overrightarrow{\boldsymbol{S}}_{i}^{2}\right\rangle_{\mathrm{eq}}\right) . \tag{3.7}
\end{equation*}
$$

The equilibrium value $\left\langle\overrightarrow{\boldsymbol{S}}_{i}^{1} \cdot \overrightarrow{\boldsymbol{S}}_{i}^{2}\right\rangle_{\text {eq }}$ needs to be subtracted to have the ground state of spin and phonon system be the vacuum. Note that here the phonon operators are not given in momentum space but rather in real space.

Excitation is achieved by coupling an electric field $E(t)$ to the phonon displacement $\left(\boldsymbol{b}_{i}+\boldsymbol{b}_{i}^{\dagger}\right)$

$$
\begin{equation*}
\boldsymbol{H}_{\mathrm{L}}=\sum_{i} E(t)\left(\boldsymbol{b}_{i}+\boldsymbol{b}_{i}^{\dagger}\right), \tag{3.8}
\end{equation*}
$$

where the laser field $E(t)$ is never turned off and will also not be quantized. As the atoms carrying the phonons posses an electric dipole moment which couples to an electric field the physical foundation to this ansatz is provided.

### 3.2 Approximations

To obtain a system of ordinary differential equations the Hamiltonian needs to be further simplified. First, however, we will use the bond operator representation to describe the spin system. In particular, the triplon representation introduced in chapter 2.2 .2 is applied. Via Fourier transforming

$$
\begin{align*}
\boldsymbol{t}_{\alpha, i} & =\frac{1}{\sqrt{N}} \sum_{k} \boldsymbol{t}_{k, \alpha} \mathrm{e}^{\mathrm{i} k r_{i}}  \tag{3.9a}\\
\boldsymbol{t}_{\alpha, i}^{\dagger} & =\frac{1}{\sqrt{N}} \sum_{k} \boldsymbol{t}_{k, \alpha}^{\dagger} \mathrm{e}^{-\mathrm{i} k r_{i}}  \tag{3.9b}\\
\boldsymbol{b}_{i} & =\frac{1}{\sqrt{N}} \sum_{k} \boldsymbol{b}_{k} \mathrm{e}^{\mathrm{i} k r_{i}}  \tag{3.9c}\\
\boldsymbol{b}_{i}^{\dagger} & =\frac{1}{\sqrt{N}} \sum_{k} \boldsymbol{b}_{k}^{\dagger} \mathrm{e}^{-\mathrm{i} k r_{i}} \tag{3.9d}
\end{align*}
$$

the system will be completely transferred into momentum space.
As a first approximation we neglect any triplon operators of a level higher than bilinear, thus

$$
\begin{equation*}
\boldsymbol{H}_{\mathrm{S}}=J\left(\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}-\frac{\lambda}{4} \cos (k)\left(\boldsymbol{t}_{k, \alpha}^{\dagger}+\boldsymbol{t}_{-k, \alpha}\right)\left(\boldsymbol{t}_{-k, \alpha}^{\dagger}+\boldsymbol{t}_{k, \alpha}\right)\right), \tag{3.10}
\end{equation*}
$$

where we have introduced the rescaled inter dimer coupling strength

$$
\begin{equation*}
\lambda=\frac{J^{\prime}}{J} \tag{3.11}
\end{equation*}
$$

The second approximation drastically simplifies our model. We only take the $k=0$ phonon mode into account

$$
\begin{array}{ll}
\boldsymbol{b}_{k}=0 & \forall k \neq 0 \\
\boldsymbol{b}_{k}^{\dagger}=0 & \forall k \neq 0 \tag{3.12b}
\end{array}
$$

which yields in (3.9c) and (3.9d)

$$
\begin{align*}
\boldsymbol{b}_{i} & =\frac{1}{\sqrt{N}} \boldsymbol{b}_{0}  \tag{3.13a}\\
\boldsymbol{b}_{i}^{\dagger} & =\frac{1}{\sqrt{N}} \boldsymbol{b}_{0}^{\dagger} . \tag{3.13b}
\end{align*}
$$

The wavelengths of lasers are of some hundred nanometers whereas the atoms generating the phonons are of some ångström which makes for a difference of several orders of magnitude, i.e. the interaction is of long range order. Hence, it is reasonable to assume that the phonon frequency is dominated by its first order given by the applied frequency. Now, the phonon system Hamiltonian reads

$$
\begin{equation*}
\boldsymbol{H}_{\mathrm{P}}=\omega_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \tag{3.14}
\end{equation*}
$$

i.e.

$$
\begin{equation*}
\omega_{0}=\omega_{\mathrm{B}}(0), \tag{3.15}
\end{equation*}
$$

and the coupling to the laser field is

$$
\begin{equation*}
\boldsymbol{H}_{\mathrm{L}}=E(t) \sqrt{N}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right) \tag{3.16}
\end{equation*}
$$

We take the electric field $E(t)$ to be a cosine

$$
\begin{equation*}
E(t)=a \cos (\Omega t) \tag{3.17}
\end{equation*}
$$

with $a$ describing the amplitude and $\Omega$ the laser frequency.
In a first order approximation we use mean field theory for the remaining spin-phonon coupling to obtain a bilinear Hamiltonian, yielding

$$
\begin{align*}
\boldsymbol{H}_{\mathrm{SP}}= & g\left\langle\frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\right\rangle\left(\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}-\sum_{k, \alpha}\left\langle\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right\rangle_{\mathrm{eq}}\right) \\
& +g \frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\left\langle\left(\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}-\sum_{k, \alpha}\left\langle\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right\rangle_{\mathrm{eq}}\right)\right\rangle . \tag{3.18}
\end{align*}
$$

In summary we denote the full Hamiltonian $\boldsymbol{H}$ we hereinafter use as the basis for our calculations

$$
\begin{align*}
\boldsymbol{H}= & \boldsymbol{H}_{\mathrm{S}}+\boldsymbol{H}_{\mathrm{P}}+\boldsymbol{H}_{\mathrm{SP}}+\boldsymbol{H}_{\mathrm{L}}  \tag{3.19a}\\
\boldsymbol{H}_{\mathrm{S}}= & \left(J \sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}-\frac{\lambda}{4} \cos (k)\left(\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}+\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}+2 \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}+1\right)\right)  \tag{3.19b}\\
\boldsymbol{H}_{\mathrm{P}}= & \omega_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}  \tag{3.19c}\\
\boldsymbol{H}_{\mathrm{L}}= & a \cos (\Omega t) \sqrt{N}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)  \tag{3.19~d}\\
\boldsymbol{H}_{\mathrm{SP}}= & g\left\langle\frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\right\rangle\left(\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}-\sum_{k, \alpha}\left\langle\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right\rangle_{\mathrm{eq}}\right) \\
& +g \frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\left\langle\left(\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}-\sum_{k, \alpha}\left\langle\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right\rangle_{\mathrm{eq}}\right)\right\rangle . \tag{3.19e}
\end{align*}
$$

So far, the approximations made were to simplify the Hamiltonian $\boldsymbol{H}$. There is, however, another assumption that comes into play when the equations of motion are to be derived in section 3.4 involving the triplon operator representation used to describe the dimerized
spin chain. As we worked out in chapter 2.2 .2 , triplon operators are of hardcore bosonic nature rather than of bosonic nature, c.f. equation (2.19). However, we shall treat them as ordinary bosonic operators, thus using the commutation relations

$$
\begin{align*}
& {\left[\boldsymbol{t}_{\alpha, i}, \boldsymbol{t}_{\beta, j}^{\dagger}\right]=\delta_{\alpha \beta} \delta_{i j}}  \tag{3.20a}\\
& {\left[\boldsymbol{t}_{\alpha, i}, \boldsymbol{t}_{\beta, j}\right]=0}  \tag{3.20b}\\
& {\left[\boldsymbol{t}_{\alpha, i}^{\dagger}, \boldsymbol{t}_{\beta, j}^{\dagger}\right]=0 .} \tag{3.20c}
\end{align*}
$$

Because the triplon operators are by construction of hardcore bosonic type which was not touched when the Hamiltonian $\boldsymbol{H}$ was set up, the likelihood of two triplons of the exact same type showing up at the exact same lattice site is negligibly small justifying equation (3.20).

Furthermore, we shall restrict ourselves to investigating only the case of zero temperature

$$
\begin{equation*}
T=0 \tag{3.21}
\end{equation*}
$$

and leave the more general problem for further research to come.
More sophisticated calculations for the derivation presented here can be found in appendix A.

### 3.3 Observables

We begin by defining the following observables and expectation values for the phonon system

$$
\begin{align*}
& A(t)=\left\langle\frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\right\rangle(t)  \tag{3.22a}\\
& B(t)=\left\langle\frac{\mathrm{i}}{\sqrt{N}}\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right)\right\rangle(t)  \tag{3.22b}\\
& C(t)=\left\langle\frac{1}{N} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle(t), \tag{3.22c}
\end{align*}
$$

where $A(t)$ is the normalized phonon displacement, $B(t)$ the normalized phonon momentum and $C(t)$ the normalized phonon density.

For the triplon system we introduce

$$
\begin{align*}
U_{k}(t) & =\sum_{\alpha}\left\langle\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right\rangle(t)  \tag{3.23a}\\
V_{k}(t) & =\sum_{\alpha}\left\langle\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}+\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}\right\rangle(t)  \tag{3.23b}\\
W_{k}(t) & =\mathrm{i} \sum_{\alpha}\left\langle\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}-\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}\right\rangle(t) . \tag{3.23c}
\end{align*}
$$

Here, $U_{k}(t)$ describes the triplon density for a certain momentum $k$, whereas $V_{k}(t)$ and $W_{k}(t)$ do not have a descriptive meaning but will occur as expectation values of off diagonal
bilinear triplon operators when setting up the equations of motion. It will also prove useful to define the normalized triplon density

$$
\begin{equation*}
\mathcal{U}(t)=\frac{1}{N} \sum_{k} U_{k}(t) \tag{3.24}
\end{equation*}
$$

for which, due to the fact that triplons are hardcore bosons,

$$
\begin{equation*}
0 \leq \mathcal{U}(t) \leq 1 \tag{3.25}
\end{equation*}
$$

must hold for any time $t$.
An investigation of the average behaviour of those observables is achieved by

$$
\begin{equation*}
\bar{X}_{t_{0}}(t)=\frac{1}{t-t_{0}} \int_{t_{0}}^{t} X\left(t^{\prime}\right) \mathrm{d} t^{\prime}, \quad t>t_{0} \tag{3.26}
\end{equation*}
$$

where $X(t)$ may take the role of any of the expectation values introduced above. We will refer to $\bar{X}_{t_{0}}(t)$ as the time average of the observable $X(t)$ taken from $t_{0}$ on.

### 3.4 Equations of Motion

To take the damping of the phonon system into account, the equations of motion to its observables need to be calculated using the Lindblad formalism introduced in section 2.4 . In particular, we shall make use of equation (2.52) using the total Hamiltonian (3.1) in its approximated form derived in section 3.2 . Evidently, the corresponding creation and annihilation operators are $\boldsymbol{b}_{0}^{\dagger}$ and $\boldsymbol{b}_{0}$ as introduced in equation (3.13). We obtain

$$
\begin{align*}
& \frac{\mathrm{d}}{\mathrm{~d} t} A(t)=\omega_{0} B(t)-\frac{1}{2} \gamma A(t)  \tag{3.27a}\\
& \frac{\mathrm{d}}{\mathrm{~d} t} B(t)=-\omega_{0} A(t)-\frac{1}{2} \gamma B(t)-2\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right)  \tag{3.27b}\\
& \frac{\mathrm{d}}{\mathrm{~d} t} C(t)=-\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) B(t)-\gamma C(t) . \tag{3.27c}
\end{align*}
$$

The time evolution of the triplon system's observables is obtained from the Heisenberg equation of motion (2.29) as introduced in section 2.3

$$
\begin{align*}
\frac{\mathrm{d}}{\mathrm{~d} t} U_{k}(t) & =\frac{1}{2} J^{\prime} \cos (k) W_{k}(t)  \tag{3.27d}\\
\frac{\mathrm{d}}{\mathrm{~d} t} V_{k}(t) & =2\left(J-\frac{1}{2} J^{\prime} \cos (k)+g A(t)\right) W_{k}(t)  \tag{3.27e}\\
\frac{\mathrm{d}}{\mathrm{~d} t} W_{k}(t) & =-2\left(J-\frac{1}{2} J^{\prime} \cos (k)+g A(t)\right) V_{k}(t)+2 J^{\prime} \cos (k)\left(U_{k}(t)+\frac{1}{2}\right) \tag{3.27f}
\end{align*}
$$

The corresponding calculations are given in appendix $B$ in detail. Note that there are no correlations between different modes $k$ and that the system is totally symmetric in $k$ for it
only occurs as the argument of a cosine. Thus, the dimension of the ordinary differential equation system (3.27) can be reduced from $3 N+3$ to $3 N / 2+3$ given $N$ is even.

Initially the whole system should be in its ground state, i.e. in equilibrium, at zero temperature. Hence, the initial condition for the phonon system reads

$$
\begin{align*}
& A(0)=0  \tag{3.28a}\\
& B(0)=0  \tag{3.28b}\\
& C(0)=0 \tag{3.28c}
\end{align*}
$$

To achieve this effect for the triplon system, its Hamiltonian (3.10) needs to be diagonalized. We employ a Bogoliubov transform introducing new triplon operators via

$$
\begin{align*}
\boldsymbol{t}_{k, \alpha} & =\hat{\boldsymbol{t}}_{k, \alpha} \cosh (\theta)+\hat{\boldsymbol{t}}_{-k, \alpha}^{\dagger} \sinh (\theta)  \tag{3.29a}\\
\boldsymbol{t}_{k, \alpha}^{\dagger} & =\hat{\boldsymbol{t}}_{k, \alpha}^{\dagger} \cosh (\theta)+\hat{\boldsymbol{t}}_{-k, \alpha} \sinh (\theta) . \tag{3.29b}
\end{align*}
$$

It is straight forward but tedious to deduce

$$
\begin{equation*}
\boldsymbol{H}_{\mathrm{S}} \propto \sum_{k, \alpha} \hat{\boldsymbol{t}}_{k, \alpha}^{\dagger} \hat{\boldsymbol{t}}_{k, \alpha} \omega_{\mathrm{T}}(k) \tag{3.30}
\end{equation*}
$$

with the triplon dispersion relation

$$
\begin{equation*}
\omega_{\mathrm{T}}(k)=J \sqrt{1-\lambda \cos (k)}, \tag{3.31}
\end{equation*}
$$

c.f. appendix C. Rewriting the ground state condition of the diagonalized system with operators $\hat{\boldsymbol{t}}_{k, \alpha}^{\dagger}$ and $\hat{\boldsymbol{t}}_{k, \alpha}$ in terms of $\boldsymbol{t}_{k, \alpha}^{\dagger}$ and $\boldsymbol{t}_{k, \alpha}$ eventually yields

$$
\begin{align*}
U_{k}(0) & =\frac{\left(\omega_{\mathrm{T}}(k)-J\right)^{2}}{4 J \omega_{\mathrm{T}}(k)}  \tag{3.32a}\\
V_{k}(0) & =\frac{\left(J^{2}-\omega_{\mathrm{T}}^{2}(k)\right)}{2 J \omega_{\mathrm{T}}(k)}  \tag{3.32b}\\
W_{k}(0) & =0 . \tag{3.32c}
\end{align*}
$$

An important special case is obtained for $k=\pi / 2$. The triplon system will vanish completely as

$$
\begin{equation*}
\omega_{\mathrm{T}}(\pi / 2)=1 \tag{3.33a}
\end{equation*}
$$

so that according to equation (3.32)

$$
\begin{equation*}
U_{\pi / 2}(0)=V_{\pi / 2}(0)=W_{\pi / 2}(0)=0 \tag{3.33b}
\end{equation*}
$$

which then yields

$$
\begin{equation*}
U_{\pi / 2}(t)=V_{\pi / 2}(t)=W_{\pi / 2}(t)=0 . \tag{3.33c}
\end{equation*}
$$

### 3.5 Parameters of the Model

The system of ordinary differential equations $(3.27)$ set up in the previous section has the following degrees of freedom

$$
\begin{align*}
N & - \text { system size, number of dimers }  \tag{3.34a}\\
J & - \text { coupling between two spins of a dimer }  \tag{3.34b}\\
J^{\prime} & - \text { coupling between two dimers }  \tag{3.34c}\\
g & - \text { coupling between spin and phonon system }  \tag{3.34~d}\\
\omega_{0} & - \text { phonon frequency }  \tag{3.34e}\\
\Omega & - \text { frequency of the energy } E(t) \text { due to the laser field } \tilde{E}(t)  \tag{3.34f}\\
a & - \text { amplitude of the energy } E(t) \text { due to the laser field } \tilde{E}(t)  \tag{3.34~g}\\
\gamma & - \text { Lindblad damping parameter. } \tag{3.34h}
\end{align*}
$$

Before limiting certain parameters we shall at first simplify the system some more. The system size $N$ should not play any role in the time evolution of the observables introduced in equations (3.22) and (3.23), i.e. it should not effect the physics for it is a purely numerical parameter. Its only purpose is to be large enough so that finite size effects can be ruled out. Empirically we found

$$
\begin{equation*}
N=400 \tag{3.35a}
\end{equation*}
$$

to be sufficient. Furthermore, we wish to measure any other quantities in units of $J$, the coupling strength of a dimer, which means

$$
\begin{equation*}
J=1 \tag{3.35b}
\end{equation*}
$$

numerically. We choose

$$
\begin{equation*}
J^{\prime}=0.5 \Rightarrow \lambda=0.5 \tag{3.35c}
\end{equation*}
$$

at which the spin chain can still be regarded as dimerized, however, quantum effects become very important as there are mobile (quasi) particles in the system. To ensure that the laser has an effect on the phonon system we set

$$
\begin{equation*}
\omega_{0}=\Omega \tag{3.35~d}
\end{equation*}
$$

i.e. we set the laser in resonance with the phonon system. If both quantities were to far off from one another there would be no proper excitation, meaning the system would stay in its initial equilibrium state. Note that, as explained in section 2.8 , because of the damping the system does not diverge when pumped in resonance.

Thus, the four remaining quantities to be manipulated are the coupling strength between spin and phonon system $g$, the damping parameter $\gamma$ and the laser parameters, being the amplitude $a$ and the frequency $\Omega=\omega_{0}$. The phonon damping parameter $\gamma$ is the least intuitive quantity of this model. It should be stronger than the coupling between the phonon
and the spin system to make sure the probe does not overheat. If, however, it is too large, the excitation will not show any effect. Reasonable values include ${ }^{1}$

$$
\begin{equation*}
\gamma \approx 0.1 \mathrm{~J} \tag{3.36a}
\end{equation*}
$$

To ensure that the spin system is predominantly described by the dimerization the coupling to the phonon system should be of a lower order of magnitude, hence

$$
\begin{equation*}
g \approx 0.01 \mathrm{~J} \tag{3.36b}
\end{equation*}
$$

For the dispersion relations are of the order of $J$, the frequency $\Omega=\omega_{0}$ takes values of orders close to $J$, i.e.

$$
\begin{equation*}
\Omega=\omega_{0} \approx J \tag{3.36c}
\end{equation*}
$$

As there are, however, lasers operating on very different wavelengths, deviations are not non-physical. Estimating the value of the amplitude yields possible values around

$$
\begin{equation*}
a \approx 0.01 \mathrm{~J} . \tag{3.36d}
\end{equation*}
$$

The maximum energy corresponding to the excitation with the laser field, i.e. the amplitude, is given by

$$
\begin{equation*}
a=\tilde{E}_{0} e x_{0} \tag{3.37}
\end{equation*}
$$

where $e$ is the elementary charge, $x_{0}$ the maximum displacement and

$$
\begin{equation*}
\tilde{E}_{0}=\sqrt{\frac{I}{c \varepsilon_{0}}} \tag{3.38}
\end{equation*}
$$

the amplitude of the electric field strength $\tilde{E}(t)$. The parameters $c$ and $\varepsilon_{0}$ are the speed of light and the vacuum permittivity respectively. By $I$ we denote the intensity of the electric field $\tilde{E}(t)$. We take the ground state of an harmonic oscillator as a first approximation to calculate the maximum displacement as

$$
\begin{equation*}
x_{0}^{2}=\frac{\hbar}{m \omega} \tag{3.39}
\end{equation*}
$$

with the mass $m$ of the atoms of the system under consideration and the frequency $\omega$ which can be expressed by the wavelength $\lambda_{\mathrm{L}}$ as

$$
\begin{equation*}
\omega=2 \pi \frac{c}{\lambda_{\mathrm{L}}} . \tag{3.40}
\end{equation*}
$$

Substituting into equation (3.37) and dividing by the coupling strength $J$ yields

$$
\begin{equation*}
\frac{a}{J}=\frac{e}{J} \sqrt{\frac{\hbar I \lambda_{\mathrm{L}}}{2 \pi m c^{2} \varepsilon_{0}}} . \tag{3.41}
\end{equation*}
$$

For common values of intensity $\Gamma^{2}$, wavelength $\lambda_{\mathrm{L}}$, atomic mass $m$ and spin-phonon coupling constant $J$

$$
\begin{equation*}
I \approx 10^{9} \mathrm{~W} \mathrm{~m}^{-1} \tag{3.42a}
\end{equation*}
$$

[^0]\[

$$
\begin{align*}
\lambda_{\mathrm{L}} & \approx 100 \mathrm{~nm}-10^{5} \mathrm{~nm}  \tag{3.42b}\\
m & \approx 1 \mathrm{u}-0 \mathrm{u}  \tag{3.42c}\\
J & \approx 1 \mathrm{meV} \tag{3.42d}
\end{align*}
$$
\]

we obtain equation $(3.36 \mathrm{~d})$.

## 4 Results

We begin with some general remarks before presenting the results to our simulations. The system of ordinary differential equations (3.27) was implemented in C++. We computed solutions by making use of the GNU Scientific Library [11] where, unless noted otherwise, the Runge Kutta 4 algorithm with

$$
\begin{align*}
\varepsilon_{\text {rel }} & =0  \tag{4.1a}\\
\varepsilon_{\text {abs }} & =10^{-6}  \tag{4.1b}\\
h_{\text {start }} & =10^{-6}  \tag{4.1c}\\
N_{\text {data }} & =40000 \tag{4.1d}
\end{align*}
$$

was applied. The numerical parameters $\varepsilon_{\text {rel }}$ and $\varepsilon_{\text {abs }}$ are the relative and absolute maximum error for the algorithm to respect, $h_{\text {start }}$ gives the initial step size and $N_{\text {data }}$ provides the absolute number of data points generated. Fourier transforms as well as fit functions, however, were calculated with Python utilizing the Fast Fourier Transform implementation of Numpy 31 or the optimization routine of Scipy [17] respectively. Plots were created with Python's Matplotib 15.

As already mentioned in chapter 3.5 , the following quantities were set to the fixed values

$$
\begin{align*}
N & =400  \tag{4.2a}\\
J & =1  \tag{4.2b}\\
J^{\prime} & =0.5 \tag{4.2c}
\end{align*}
$$

and the system was driven in resonance, i.e.

$$
\begin{equation*}
\Omega=\omega_{0} \tag{4.2~d}
\end{equation*}
$$

In the following we shall only note those quantities down when there is a difference from equation (4.2).

Finally, we need to introduce the maximum time of simulation, $t_{\max }$, which in general is completely arbitrary. Unless noted otherwise we take

$$
\begin{equation*}
t_{\max }=1000 J^{-1} \tag{4.3}
\end{equation*}
$$

to be sufficient.

### 4.1 Primary Investigation of the Decoupled System

Before turning to the way more sophisticated problem of solving and interpreting equation (3.27) in general we first examine basic properties of the phonon system. Therefore, we decouple spin and phonon system, i.e. we set

$$
\begin{equation*}
g=0 \tag{4.4}
\end{equation*}
$$

As is evident from equation (3.27) the system now consists of $N+1$ independent subsystems. One the one hand, there is the phonon system composed of the phonon displacement $A(t)$, the phonon momentum $B(t)$ and the the phonon density $C(t)$ described by equations (3.27a) to $(\overline{3.27 \mathrm{c}})$. On the other hand, there are $N$ triplon systems, one for each momentum $k$, that are not dependent on one another as already pointed out in section 3.4. They are characterised by equations $(3.27 \mathrm{~d})$ to $(3.27 \mathrm{f})$. Figure 4.1 represents such a case. The phonon system makes for a prime example of the damped driven harmonic oscillator as discussed in section 2.8 . With the driving beginning at time $t=0$ the phonon density $C(t)$ establishes an oscillation around a specific value, i.e. after the transient oscillation the system reaches a quasi steady state which is the expected situation regarding equation $(2.119)$, c.f. figure 4.1 (a). close-ups of the initial and final times of the observed time period in figures 4.1 (b) and $4.1(\mathrm{c})$ confirm the oscillatory behaviour. The phonon displacement and momentum, $A(t)$ and $B(t)$, are printed in the following lines alongside a respective close-up for final times, c.f. $4.1(\mathrm{~d})$ and $4.1(\mathrm{e})$ or $4.1(\mathrm{f})$ and 4.1 (g) respectively. Both quantities oscillate around zero having gained a constant maximum amplitude after the transient event in the beginning. A comparison of them in figure 4.1 (h) illustrates their phase shift of $\pi / 2$ which corresponds with both the classical and the quantum description of the damped driven harmonic oscillator. For there is no direct excitation alongside the coupling to the phonon system, the spin system should not show any response. Hence, the normalised triplon density $\mathcal{U}(t)$ should keep its initial value at all times. Figure 4.1(i) matches our expectation thus proving the accuracy of the simulation.


Figure 4.1: Example of a decoupled system; values: $g=0, \omega_{0}=1.3 J, a=0.4 J, \gamma=0.1 J$

### 4.2 Investigation on the Occurrence of Quasi Steady States

As pointed out in the introduction, the ultimate goal of this project is to drive the system into a quasi steady state of which we are, to a certain degree, free to choose its parameters. Figure 4.2 depicts results of a simulation that exactly meet those requirements. We have introduced a finite spin-phonon coupling

$$
\begin{equation*}
g=0.03 \mathrm{~J} \tag{4.5}
\end{equation*}
$$

Here, both the phonon and the triplon density oscillate around a certain value that differs significantly from their initial state. The dynamics of the phonon density $C(t)$ can again be described by the damped driven harmonic oscillator. Having the results from section 4.1 and, thus, figure 4.1 in mind, we note that there is little difference to the behaviour of the phonon density $C(t)$ as compared to the decoupled system, c.f. figures 4.2 (a) to 4.2 (c). The same also applies to the phonon displacement and momentum, $A(t)$ and $B(t)$, which is why they are only printed macroscopically in figures $4.2(\mathrm{~g})$ and $4.2(\mathrm{~h})$. These properties come to no surprise when considering the system of ordinary differential equations (3.27). Introducing the spin-phonon coupling $g$ will only serve as an adjustment of the driving in $(3.27 \mathrm{~b})$ and $(3.27 \mathrm{c})$. As the quantity $g$, however, is of a much lower order of magnitude than the coupling strength $J$ between two spins of a dimer and the difference of the triplon density to its initial value must always be less than 1 due to relation $(3.25)$, the overall effect on the phonon system should be quite small. To the spin system in equation $(3.27 \mathrm{~d})$ to (3.27f) the spin-phonon coupling $g$ is the only connection to the excitation which calls for a strong impact. The plot of the triplon density $\mathcal{U}(t)$ in figure $4.2(\mathrm{~d})$ and its close-ups in figures 4.2 (e) and 4.2 (f) clearly depict the establishment of a quasi steady state for the triplon system as well.

We further investigate the system in terms of its accordance with equation (2.119). Therefore, we compare the phonon densities $C(t)$ of the decoupled system and the system with coupling (4.5). We take the time average as proposed in equation (3.26) over the final time period of $10 J^{-1}$, i.e. $\bar{C}_{990 J^{-1}}\left(1000 J^{-1}\right)$, plotting it against the amplitude per damping squared $(a / \gamma)^{2}$. Note that in the derivation of equation (2.119) we have made use of a specific maximum displacement $x_{\max }$ rather than a time averaged value. For both quantities are proportional to one another as an oscillatory process is described, our method is still valid. Figure 4.3 shows the results we obtain. A linear correlation is evident. Applying a linear fit

$$
\begin{equation*}
f(x)=\tilde{m} x+\tilde{n} \tag{4.6}
\end{equation*}
$$

the fit parameters read

$$
\begin{align*}
\tilde{m} & =0.99921552  \tag{4.7a}\\
\tilde{n} & =10^{-9} \tag{4.7b}
\end{align*}
$$

for the decoupled system and

$$
\begin{align*}
\tilde{m} & =0.99938  \tag{4.8a}\\
\tilde{n} & =10^{-5} \tag{4.8b}
\end{align*}
$$








Figure 4.2: Example of a desired time evolution; values: $g=0.03 J, \omega_{0}=1.3 J, a=0.4 J$, $\gamma=0.1 J$


Figure 4.3: Time averaged phonon density $\bar{C}_{990 J^{-1}}\left(t=1000 J^{-1}\right)$ vs. amplitude per damping squared $(a / \gamma)^{2}$; values: $\omega_{0}=1.3 J, \gamma=0.1 J$
for the coupled system. We determined the accuracy of these fit parameters by performing two fits for each parameter set using only every second datum for the second fit, i.e. we only present the parameters up to digits that were identical in both calculations. Both systems yield a number close to 1 as the proportionality factor $\tilde{m}$ and a number close to 0 as the offset $\tilde{n}$, with slightly better agreement for the decoupled system as the numbers show. Because the decoupled system should be easier to solve numerically, differences in the results can also be regarded as numerical artefacts. However, the results clearly show that equation $(2.119)$ holds either way for the deviations from the theoretical values are negligibly small. We have also proven that applying the coupling $g$ between phonon and spin system does have little to no effect on the time evolution of the phonon system.

### 4.2.1 Phenomena of Divergences

Instead of further analysing the time evolution of the triplon density in figure 4.1 we need to mention some other obstacles. Increasing the spin-phonon coupling to

$$
\begin{equation*}
g=0.04 J \tag{4.9}
\end{equation*}
$$

i.e. increasing the value by a fraction of $1 / 3$ compared to the previous instalment, we find the unexpected effects of divergences depicted in figure 4.4 .

In the beginning the spin system, as well as the phonon system show little difference to the previous setting in figure 4.2. However, at a certain time we find a collapse in the phonon density $C(t)$ followed by a completely non-linear time evolution. This effect is accompanied by a collapse of the phonon displacement and momentum, $A(t)$ and $B(t)$, which is then succeeded by what appears to be a transition. Those effects were preceded by a rapid increase of the phonon density $\mathcal{U}(t)$, c.f. figures $4.4(\mathrm{a})$ to $4.4(\mathrm{~d})$. This increase completely violates relation (3.25) which has to hold at all times for our model to be valid. Figure 4.4 (e) illustrates this violation by also plotting the actual upper limit of 1 . Another close-up of the oscillation can be taken from figure $4.4(\mathrm{f})$. As a result, we need to come to the conclusion that the approximations we made in section 3.2 when setting up the Hamiltonian $\boldsymbol{H}$ are not applicable to every set of parameters.
We can rule out numerical effects for we obtained those results using different solvers with different parameters. Although the time evolution after the first collapse might not have been equal for each solver used, the violation of relation (3.25) was still most evident and called for a further investigation.

The next section will be devoted to working out where our model describes the system under consideration well and where it needs to be modified.

### 4.2.2 Triplon Dispersion Relation and Higher Harmonics

As we pointed out in section 3.5, driving the system in resonance does not lead to a divergence because of the damping. However, we were only focussing on the phonon system. We also need to take the triplon system into account by regarding the triplon dispersion relation in equation (3.31). Inspecting the spin system's Hamiltonian $\boldsymbol{H}_{\mathrm{S}}$ in equation (3.10) we find that there are only combinations of triplon operators that either leave the triplon number constant or change it by two. We, therefore, introduce the one and two triplon band where the one triplon band is given by the triplon dispersion relation (3.31)

$$
\begin{equation*}
\omega_{1}(k)=\omega_{\mathrm{T}}(k)=J \sqrt{1-\lambda \cos (k)} \tag{4.10}
\end{equation*}
$$

and the two triplon band by the sum of the triplon dispersion relation (3.31) for two momenta $k$ and $k+p$, i.e.

$$
\begin{equation*}
\omega_{2}(k, p)=\omega_{\mathrm{T}}(k)+\omega_{\mathrm{T}}(k+p) . \tag{4.11}
\end{equation*}
$$

The maximum and minimum values of those bands will serve as their limits

$$
\begin{align*}
& \omega_{1, \min }=\min _{k}\left\{\omega_{1}(k)\right\}=J \sqrt{1-\lambda}  \tag{4.12a}\\
& \omega_{1, \max }=\max _{k}\left\{\omega_{1}(k)\right\}=J \sqrt{1+\lambda}  \tag{4.12b}\\
& \omega_{2, \min }=\min _{p}\left\{\min _{k}\left\{\omega_{2}(k, p)\right\}\right\}=2 J \sqrt{1-\lambda}=\omega_{\min }  \tag{4.12c}\\
& \omega_{2, \max }=\max _{p}\left\{\max _{k}\left\{\omega_{2}(k, p)\right\}\right\}=2 J \sqrt{1+\lambda}=\omega_{\max } . \tag{4.12d}
\end{align*}
$$



(c)

(d)

(e)



Figure 4.4: Example of diverging phenomena; values: $g=0.04 \mathrm{~J}, \omega_{0}=1.3 \mathrm{~J}, a=0.4 \mathrm{~J}$, $\gamma=0.1 J$

Details on the calculation can be found in appendix C. Here, we have introduced the limits $\omega_{\text {min }}$ and $\omega_{\text {max }}$ independently from the triplon band which lead to the definition of three regimes

$$
\begin{align*}
\omega_{0} & =\Omega<\omega_{\min }-\text { adiabatic / beneath band regime }  \tag{4.13a}\\
\omega_{\min }<\omega_{0} & =\Omega<\omega_{\max }-\text { in band regime }  \tag{4.13b}\\
\omega_{0} & =\Omega>\omega_{\max }-\text { anti-adiabatic / above band regime. } \tag{4.13c}
\end{align*}
$$

Based on the results we have already obtained, i.e. the behaviour of the phonon system is largely independent of the spin-phonon coupling $g$ and there are certain regions where the model is not applicable, we shall find a systematic approach to investigate the nature of those particular regions. Comparing the parameter sets used in section 4.2 and 4.2 .1 there appears to be a critical value to the spin-phonon coupling $g_{\text {crit }}$ at which the normalized triplon density $U(t)$ starts to take non-physical values. Thus, we calculate the time evolution of the system for several values of spin-phonon coupling $g$ depending on phonon and laser frequency $\omega_{0}=\Omega$ and the quantity $(a / \gamma)^{2}$ which is proportional to the phonon density $C(t)$ trying to find the critical value $g_{\text {crit }}$.

Figure 4.5 shows the corresponding results. We did neither compute results for $\omega_{0}=\Omega=0$ nor $(a / \gamma)^{2}=0$ as both cases describe non-dynamical systems which makes for an arbitrarily large value of the critical value $g_{\text {crit }}$. Calculations were done in steps of

$$
\begin{align*}
g_{\text {step }} & =0.01 \mathrm{~J}  \tag{4.14a}\\
\omega_{0, \text { step }} & =0.01 \mathrm{~J}  \tag{4.14b}\\
(a / \gamma)_{\text {step }}^{2} & =1 \tag{4.14c}
\end{align*}
$$

while the damping parameter was set to

$$
\begin{equation*}
\gamma=0.1 \mathrm{~J} . \tag{4.14d}
\end{equation*}
$$

As the maximum value of spin-phonon coupling we chose

$$
\begin{equation*}
g_{\max }=0.1 \mathrm{~J} \tag{4.14e}
\end{equation*}
$$

in order to meet the criterion (3.36b) ensuring that the coupling strength between spin system and phonon system $g$ is of a lower order of magnitude than the dimer coupling strength $J$. The time evolutions were computed until times of

$$
\begin{equation*}
t_{\max }=10000 J^{-1} \tag{4.14f}
\end{equation*}
$$

to avoid misjudging systems where a non-physical behaviour occurs later in time. The band limits $\omega_{\min }$ and $\omega_{\max }$ are also depicted in figure 4.5. Farther above the band there is no restriction to the spin-phonon coupling strength $g$. We note that another effect, when it comes to the establishment of the stable region above the band, is visible which we will not discuss in more detail. Instead, we label the regime significantly above the band as stable. By contrast, if we pump the system with an in band frequency $\Omega$, there will be an immediate divergence. We, thus, have to avoid laser frequencies $\Omega=\omega_{0}$ that are in resonance with the
triplon band frequencies $\omega_{\mathrm{T}}(k)$. A closer investigation of systems with above band frequencies showed that for reasonable parameters according to section 3.5 the triplon density will only oscillate around its initial value which complies with our intention. An example is presented in appendix D . Therefore, we need to focus on frequencies beneath the band where we have obtained a rather complex structure. Having introduced the resonance criterion the laser frequency $\Omega=\omega_{0}$ must also not take values of multiples of band frequencies for the system to stay stable. Figure 4.6 provides a close-up of the adiabatic regime where multiples of the band limits $\omega_{\min }$ and $\omega_{\max }$ are sketched in as well. Those limits isolate diverging from nonor less diverging regimes as the rapid increase or decrease along them shows. The more bands of multiples of the limits overlap and the lesser the multiple, i.e. the more likely their occurrence, the sooner the system diverges. This property becomes most apparent for frequencies

$$
\begin{equation*}
0.7 \lesssim \Omega=\omega_{0} \lesssim 0.8 \tag{4.15}
\end{equation*}
$$

where the bands of multiples two and three overlap. We appear to find two thin stable regions, one in between the one triplon band at

$$
\begin{equation*}
\omega_{0}=\Omega=1 J \tag{4.16}
\end{equation*}
$$

in both figures, 4.5 and 4.6, and another one in between the two triplon band in figure 4.5 at

$$
\begin{equation*}
\omega_{0}=\Omega=2 J \tag{4.17}
\end{equation*}
$$

Here, however, Fermi's golden rule comes into play. As already pointed out in section 3.4 the value of $\omega_{\mathrm{T}}=1$ goes with the momentum $k=\pi / 2$ where quantities of the triplet system are either constant or zero. Hence, the density of states $\rho(t)$ vanishes which results in an absence of transitions and, thus, a stable system. For this property is solely restricted to multiples of those frequencies $\omega_{\mathrm{T}}$ corresponding to $k=\pi / 2$, the system will show a divergent behaviour once the momentum $k$ is only slightly different. Had we calculated both plots with a higher resolution, the above discussed stable regions would have gotten even thinner.

### 4.2.3 Summary

We gained several insights on the properties of our model, especially is applicability. By construction we can separate the phonon and the spin system when it comes to investigating their respective behaviour. The phonon system can always be described by a damped driven harmonic oscillator where there are little to no signs of a perturbation. These results came to no surprise, in particular for the decoupled system, as they provide exactly the results obtained from an analytical calculation. The spin system, however, confronts us with problems. First, we learn that the model is restricted in terms of its physical sense. For certain parameter sets there are divergences going beyond the validity of the bond operator representation we made excessive use of. We managed to derive this effect as a result of a resonance disaster for there is no damping to the spin system but only an excitation via a coupling to the phonon system. Building on these findings, we introduced three regimes to the system concerning the phonon and laser frequency in relation to the triplon frequency. For those frequencies larger than the maximum resonant triplon frequency the model works but does not produce any of the desired quasi steady states in the triplon density. By
contrast, if the frequencies are in band with the maximum and minimum resonant triplon frequencies, resonance effects occur leading to completely non-physical results. Thus, the most interesting regime is obtained for frequencies beneath the above discussed band. Here, effects of divergences occur once the frequencies are a multiple of the band frequencies. Otherwise, there is a possibility of driving he spin system into a quasi steady state. Any other curious effects could not be explained. In general, the limited application of the model calls for an enhancement of the spin system although we proved that in principle there is a regime where quasi steady states exist.


Figure 4.5: Contour plot of phonon density $\propto(a / \gamma)^{2}$ vs. frequency $\omega_{0}=\Omega$ vs. critical value $g_{\text {crit }}$; values: $\gamma=0.1 J, \omega_{0, \text { step }}=0.01,(a / \gamma)_{\text {step }}^{2}=1$


Figure 4.6: Contour plot of phonon density $\propto(a / \gamma)^{2}$ vs. frequency $\omega_{0}=\Omega$ vs. critical value $g_{\text {crit }}$; close-up of the adiabatic regime; values: $\gamma=0.1 J, \omega_{0, \text { step }}=0.01,(a / \gamma)_{\text {step }}^{2}=1$

### 4.3 Investigation of Green's Functions

We are interested in the interaction between particles of the spin system. Thus, we need to introduce several Green's functions to gain further insights from. In this chapter we begin by deriving the equations of motion for the retarded Green's functions corresponding to equation (2.75). The actual analysis will then make use of the slightly modified Green's functions we proposed in chapter 2.6 and their respective Fourier transforms. For the sake of clarity we name them once more. We will investigate the relative Green's function

$$
\begin{align*}
G_{\alpha \alpha, k}^{\mathrm{rel}}\left(t_{\text {in }}, t_{\mathrm{diff}}\right) & =G_{\alpha \alpha, k}^{\mathrm{ret}}\left(t_{\text {in }}, t_{\text {in }}+t_{\mathrm{diff}}\right)  \tag{4.18a}\\
G_{\alpha \alpha, k}^{\mathrm{rel}}\left(t_{\mathrm{in}}, \omega_{\text {diff }}\right) & =\frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} G_{\alpha \alpha, k}^{\mathrm{rel}}\left(t_{\mathrm{in}}, t_{\text {diff }}\right) \mathrm{e}^{\mathrm{i} \omega_{\mathrm{diff}} t_{\text {diff }}} \mathrm{d} t_{\text {diff }}, \tag{4.18b}
\end{align*}
$$

the time average Green's function

$$
\begin{align*}
\tilde{G}_{\alpha \alpha, k}\left(\bar{t}, t_{\text {diff }}\right) & =G_{\alpha \alpha, k}^{\mathrm{ret}}\left(\bar{t}-\frac{t_{\text {diff }}}{2}, \bar{t}+\frac{t_{\text {diff }}}{2}\right)  \tag{4.19a}\\
\tilde{G}_{\alpha \alpha, k}\left(\bar{t}, \omega_{\text {diff }}\right) & =\frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} \tilde{G}_{\alpha \alpha, k}\left(\bar{t}, t_{\text {diff }}\right) \mathrm{e}^{\mathrm{i} \omega_{\text {diff }} t_{\text {diff }} \mathrm{d} t_{\text {diff }}} \tag{4.19b}
\end{align*}
$$

and the average Green's function

$$
\begin{align*}
\bar{G}_{\alpha \alpha, k}\left(t_{\max }, t\right) & =\frac{1}{t_{\max }-t} \int_{0}^{t_{\max }-t} G_{\alpha \alpha, k}^{\mathrm{ret}}\left(t^{\prime}, t^{\prime}+t\right) \mathrm{d} t^{\prime}  \tag{4.20a}\\
\bar{G}_{\alpha \alpha, k}\left(t_{\max }, \omega\right) & =\frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} \bar{G}_{\alpha \alpha, k}\left(t_{\max }, t\right) \mathrm{e}^{\mathrm{i} \omega t} \mathrm{~d} t \tag{4.20~b}
\end{align*}
$$

Naturally, only those systems will be analysed where there are no diverging effects. In this chapter we present results for an adiabatic regime. The results for an anti-adiabatic regime hardly differ from the ones we give here and can be found in appendix E.

### 4.3.1 Technical Details

Obviously, we cannot simulate for infinite times necessitating the introduction of the maximum simulation time $t_{\max }$, i.e.

$$
\begin{equation*}
t \in\left[0, t_{\max }\right] . \tag{4.21}
\end{equation*}
$$

Together with the number of generated data points $N_{\text {data }}$ we obtain the time resolution

$$
\begin{equation*}
t_{\mathrm{res}}=\frac{t_{\mathrm{max}}}{N_{\mathrm{data}}} \tag{4.22}
\end{equation*}
$$

In order investigate a broad frequency spectrum a long time evolution is needed so we choose again

$$
\begin{equation*}
t_{\max }=10000 J^{-1} \tag{4.23}
\end{equation*}
$$

which yields with equation (4.1d)

$$
\begin{equation*}
t_{\mathrm{res}}=0.25 \mathrm{~J}^{-1} \tag{4.24}
\end{equation*}
$$

Clearly,

$$
\begin{equation*}
t_{\mathrm{in}} \in\left[0, t_{\max }\right] \tag{4.25}
\end{equation*}
$$

holds for the time $t_{\text {in }}$ at which the perturbation of the system begins. As we are only interested in the actual time evolution of the Green's functions, the time $t_{\text {out }}$, when the effect of the perturbation is observed, needs to satisfy

$$
\begin{equation*}
t_{\text {out }} \in\left[t_{\mathrm{in}}, t_{\max }\right] . \tag{4.26}
\end{equation*}
$$

The time difference $t_{\text {diff }}$ introduced in equation (2.87) for the relative Green's function $G_{\alpha \alpha, k}^{\mathrm{rel}}\left(t_{\mathrm{in}}, t_{\mathrm{diff}}\right)$ then needs to be taken from the interval

$$
\begin{equation*}
t_{\mathrm{diff}} \in\left[0, t_{\max }-t_{\mathrm{in}}\right] \tag{4.27}
\end{equation*}
$$

For the relative Green's function $G_{\alpha \alpha, k}^{\mathrm{rel}}\left(t_{\mathrm{in}}, t_{\text {diff }}\right)$ is by definition only a shift in time of the retarded Green's function $G_{\alpha \alpha, k}^{\text {ret }}\left(t_{\text {in }}, t_{\text {out }}\right)$ the time resolution $t_{\text {res }}$ stays the same. This also applies to the time resolution of the average Green's function $\bar{G}_{\alpha \alpha, k}\left(t_{\text {max }}, t\right)$ because averaging is a linear operation. As for the time average Green's function $\tilde{G}_{\alpha \alpha, k}\left(\bar{t}, t_{\text {diff }}\right)$ the time resolution is increased by a factor of two. For each time step $t$ any Green's function is evaluated by

$$
\begin{equation*}
t=m \cdot t_{\mathrm{res}} \tag{4.28}
\end{equation*}
$$

where $m$ is a natural number. According to the definition of the time average Green's function $\tilde{G}_{\alpha \alpha, k}\left(\bar{t}, t_{\text {diff }}\right)$ in equation (4.19a) the arguments are divided by two which has to be taken into account when the computation is performed numerically. Thus, the time resolution needs to be adjusted by a factor of two as mentioned above. For this reason the Fourier spectrum of the time average Green's function $\tilde{G}_{\alpha \alpha, k}\left(\bar{t}, \omega_{\text {diff }}\right)$ only covers half of the Fourier spectrum of the other Green's functions $G_{\alpha \alpha, k}^{\mathrm{rel}}\left(t_{\mathrm{in}}, \omega_{\mathrm{diff}}\right)$ and $\bar{G}_{\alpha \alpha, k}\left(t_{\text {max }}, \omega\right)$. As the average time $\bar{t}$ is constant, the time difference $t_{\text {diff }}$ is taken from the interval

$$
\begin{equation*}
t_{\mathrm{diff}} \in\left[0, \min \left(2 \bar{t}, 2 t_{\max }-2 \bar{t}\right)\right] \tag{4.29}
\end{equation*}
$$

In order to resolve delta peaks in the Fourier transforms properly the Fourier transforms are not strictly calculated according to equations (4.18b), (4.19b) and (4.20b) but rather via

$$
\begin{equation*}
G(t, \omega)=\frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} G\left(t, t^{\prime}\right) \mathrm{e}^{\mathrm{i} \omega t^{\prime}} \cdot \mathrm{e}^{-\Lambda t^{\prime}} \mathrm{d} t^{\prime} \tag{4.30}
\end{equation*}
$$

We have not denoted any indices here to stress that this equation applies to any Fourier transform. The damping parameter $\Lambda$ was chosen to be

$$
\begin{equation*}
\Lambda=10^{-6} J \tag{4.31}
\end{equation*}
$$

Thus, delta peaks in the original Fourier transform will take the form of a Lorentzian

$$
\begin{equation*}
f(\chi)=\xi \frac{1}{\pi} \frac{\zeta}{\zeta^{2}+\left(\chi-\chi_{0}\right)^{2}} \tag{4.32}
\end{equation*}
$$

where $\chi_{0}$ is the mean value and $\zeta$ the scale parameter which multiplied by two provides the full width at half maximum. The quantity $\xi$ is a dilation parameter that for the actual Lorentzian is set to 1 . Because numerically we do not calculate the actual Fourier transform but rather the fast Fourier transform, we need to make one last adjustment to the bare data which goes by the name of zero padding. The signal will be enhanced by a string of zeros of equal length in order to avoid overlaps in the calculated Fourier spectrum [33].

As already mentioned, the Fourier transforms were calculated using Python's Numpy. All our obtained Fourier spectra as well as toy models entirely created with Python share a rather strong offset of up to $10^{-5}$. We wish to emphasize that we do not neglect any properties of our model here but are confronted with an obstacle specific to Python supposedly due to its accuracy. Therefore, we only take fit parameters as exact up to the fifth digit and present them up to the sixth digit where deviations start to occur in most cases. We confirmed this conclusion for selected parameter sets by calculating fits using only every second datum generated.

### 4.3.2 Definition and Equations of Motion

We define the following Green's functions in accordance with equations (2.75) and (2.84)

$$
\begin{align*}
& G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)=G_{\mathbf{t}_{k, \alpha} \text { ret } \mathbf{t}_{k, \alpha}^{\dagger}}^{\text {ret }}\left(t_{\text {in }}, t_{\text {out }}\right)=-\mathrm{i}\left\langle\left[\mathbf{t}_{k, \alpha}\left(t_{\text {out }}\right), \mathbf{t}_{k, \alpha}^{\dagger}\left(t_{\text {in }}\right)\right]\right\rangle \Theta\left(t_{\text {out }}-t_{\text {in }}\right)  \tag{4.33a}\\
& F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)=G_{\mathbf{t}_{-k, \alpha}^{\dagger}, \mathbf{t}_{k, \alpha}^{\dagger}}^{\text {ret }}\left(t_{\text {in }}, t_{\text {out }}\right)=-\mathrm{i}\left\langle\left[\mathbf{t}_{-k, \alpha}^{\dagger}\left(t_{\text {out }}\right), \mathbf{t}_{k, \alpha}^{\dagger}\left(t_{\text {in }}\right)\right]\right\rangle \Theta\left(t_{\text {out }}-t_{\text {in }}\right) \tag{4.33b}
\end{align*}
$$

Note that we have dropped the triplon flavour index $\alpha$ which the time evolution of the system does not depend on. The Green's function $G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)$ describes a process where a triplon of momentum $k$ and arbitrary flavour is inserted into the system at time $t_{\text {in }}$ and removed at time $t_{\text {out }}$ whereas for $F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)$ again a triplon of momentum $k$ is inserted at time $t_{\mathrm{in}}$. However, instead of removing that same triplon at time $t_{\text {out }}$, another triplon of the opposite momentum $-k$ is inserted to ensure the total momentum conservation. Introducing the quantities

$$
\begin{align*}
\varepsilon_{k}(t) & =J-\frac{J^{\prime}}{2} \cos (k)+g A(t)  \tag{4.34a}\\
\beta_{k} & =-\frac{J^{\prime}}{4} \cos (k) \tag{4.34b}
\end{align*}
$$

we obtain the following equations of motion from (2.81) and (2.82)

$$
\frac{\mathrm{d}}{\mathrm{~d} t_{\text {out }}}\binom{G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)}{F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)}=\left(\begin{array}{cc}
-\varepsilon_{k}\left(t_{\text {out }}\right) & -2 \beta_{k}  \tag{4.35a}\\
2 \beta_{k} & \varepsilon_{k}\left(t_{\text {out }}\right)
\end{array}\right)\binom{G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)}{F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)}
$$

with the initial condition

$$
\begin{align*}
G_{k}\left(t_{\mathrm{in}}, t_{\mathrm{in}}\right) & =-\mathrm{i}  \tag{4.35b}\\
F_{k}\left(t_{\mathrm{in}}, t_{\mathrm{in}}\right) & =0 \tag{4.35c}
\end{align*}
$$

derived from equation (4.33). Rewriting both Green's functions in terms of real and imaginary part yields

$$
\begin{align*}
& \frac{\mathrm{d}}{\mathrm{~d} t_{\text {out }}}\left(\begin{array}{l}
\operatorname{Re}\left(G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right) \\
\operatorname{Im}\left(G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right) \\
\operatorname{Re}\left(F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right) \\
\operatorname{Im}\left(F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)
\end{array}\right) \\
&=\left(\begin{array}{cccc}
0 & \varepsilon_{k}\left(t_{\text {out }}\right) & 0 & 2 \beta_{k} \\
-\varepsilon_{k}\left(t_{\text {out }}\right) & 0 & -2 \beta_{k} & 0 \\
0 & -2 \beta_{k} & 0 & -\varepsilon_{k}\left(t_{\text {out }}\right) \\
2 \beta_{k} & 0 & \varepsilon_{k}\left(t_{\text {out }}\right) & 0
\end{array}\right)\left(\begin{array}{l}
\operatorname{Re}\left(G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right) \\
\operatorname{Im}\left(G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right) \\
\operatorname{Re}\left(F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right) \\
\operatorname{Im}\left(F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)
\end{array}\right) \tag{4.36a}
\end{align*}
$$

with

$$
\begin{align*}
\operatorname{Re}\left(G_{k}\left(t_{\mathrm{in}}, t_{\mathrm{in}}\right)\right) & =0  \tag{4.36b}\\
\operatorname{Im}\left(G_{k}\left(t_{\mathrm{in}}, t_{\mathrm{in}}\right)\right) & =-1  \tag{4.36c}\\
\operatorname{Re}\left(F_{k}\left(t_{\mathrm{in}}, t_{\mathrm{in}}\right)\right) & =0  \tag{4.36d}\\
\operatorname{Im}\left(F_{k}\left(t_{\mathrm{in}}, t_{\mathrm{in}}\right)\right) & =0 \tag{4.36e}
\end{align*}
$$

as the initial condition. Instead of implementing (4.36a) directly we compute the time evolution of the triplon creation and annihilation operators, $\boldsymbol{t}_{k, \alpha}^{\dagger}(t)$ and $\boldsymbol{t}_{k, \alpha}(t)$ using the ansatz

$$
\begin{align*}
& \boldsymbol{t}_{k, \alpha}^{\dagger}(t)=\sigma_{k, \alpha}(t) \boldsymbol{t}_{k, \alpha}^{\dagger}+\lambda_{k, \alpha}(t) \boldsymbol{t}_{-k, \alpha}  \tag{4.37a}\\
& \boldsymbol{t}_{k, \alpha}(t)=\sigma_{k, \alpha}^{*}(t) \boldsymbol{t}_{k, \alpha}+\lambda_{k, \alpha}^{*}(t) \boldsymbol{t}_{-k, \alpha}^{\dagger} \tag{4.37b}
\end{align*}
$$

where $z^{*}$ indicates the complex conjugate of a complex number $z$. We decompose $\sigma_{k, \alpha}(t)$ and $\lambda_{k, \alpha}(t)$ into real and imaginary parts via

$$
\begin{align*}
& \sigma_{k, \alpha}(t)=\sigma_{k, \alpha}^{1}(t)+\mathrm{i} \sigma_{k, \alpha}^{2}(t)  \tag{4.38a}\\
& \lambda_{k, \alpha}(t)=\lambda_{k, \alpha}^{1}(t)+\mathrm{i} \lambda_{k, \alpha}^{2}(t) \tag{4.38b}
\end{align*}
$$

with real $\sigma_{k, \alpha}^{1}(t), \sigma_{k, \alpha}^{2}(t), \lambda_{k, \alpha}^{1}(t)$ and $\lambda_{k, \alpha}^{2}(t)$. The system of differential equations of motion then reads

$$
\begin{align*}
& \frac{\mathrm{d}}{\mathrm{~d} t} \sigma_{k, \alpha}^{1}(t)=-\varepsilon_{k}(t) \sigma_{k, \alpha}^{2}(t)+2 \beta_{k} \lambda_{k, \alpha}^{2}(t)  \tag{4.39a}\\
& \frac{\mathrm{d}}{\mathrm{~d} t} \sigma_{k, \alpha}^{2}(t)=\varepsilon_{k}(t) \sigma_{k, \alpha}^{1}(t)-2 \beta_{k} \lambda_{k, \alpha}^{1}(t)  \tag{4.39b}\\
& \frac{\mathrm{d}}{\mathrm{~d} t} \lambda_{k, \alpha}^{1}(t)=\varepsilon_{k}(t) \lambda_{k, \alpha}^{2}(t)-2 \beta_{k} \sigma_{k, \alpha}^{2}(t)  \tag{4.39c}\\
& \frac{\mathrm{d}}{\mathrm{~d} t} \lambda_{k, \alpha}^{2}(t)=-\varepsilon_{k}(t) \lambda_{k, \alpha}^{1}(t)+2 \beta_{k} \sigma_{k, \alpha}^{1}(t) . \tag{4.39d}
\end{align*}
$$

The initial condition is obtained from equation (4.37) as

$$
\begin{align*}
& \sigma_{k, \alpha}^{1}(0)=1  \tag{4.39e}\\
& \sigma_{k, \alpha}^{2}(0)=0  \tag{4.39f}\\
& \lambda_{k, \alpha}^{1}(0)=0  \tag{4.39~g}\\
& \lambda_{k, \alpha}^{2}(0)=0 . \tag{4.39h}
\end{align*}
$$

Substituting this representation back into equation (4.33), we are enabled to express the Green's functions in terms of the newly introduced quantities $\sigma_{k, \alpha}^{1}(t), \sigma_{k, \alpha}^{2}(t), \lambda_{k, \alpha}^{1}(t)$ and $\lambda_{k, \alpha}^{2}(t)$ via

$$
\begin{align*}
\operatorname{Re}\left(G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)= & \sigma_{k, \alpha}^{1}\left(t_{\text {out }}\right) \sigma_{k, \alpha}^{2}\left(t_{\text {in }}\right)+\lambda_{k, \alpha}^{1}\left(t_{\text {in }}\right) \lambda_{k, \alpha}^{2}\left(t_{\text {out }}\right) \\
& -\sigma_{k, \alpha}^{1}\left(t_{\text {in }}\right) \sigma_{k, \alpha}^{2}\left(t_{\text {out }}\right)-\lambda_{k, \alpha}^{1}\left(t_{\text {out }}\right) \lambda_{k, \alpha}^{2}\left(t_{\text {in }}\right)  \tag{4.40a}\\
\operatorname{Im}\left(G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)= & \lambda_{k, \alpha}^{1}\left(t_{\text {in }}\right) \lambda_{k, \alpha}^{1}\left(t_{\text {out }}\right)+\lambda_{k, \alpha}^{2}\left(t_{\text {in }}\right) \lambda_{k, \alpha}^{2}\left(t_{\text {out }}^{2}\right) \\
& -\sigma_{k, \alpha}^{1}\left(t_{\text {in }}\right) \sigma_{k, \alpha}^{1}\left(t_{\text {out }}\right)-\sigma_{k, \alpha}^{2}\left(t_{\text {in }}\right) \sigma_{k, \alpha}^{2}\left(t_{\text {out }}\right)  \tag{4.40b}\\
\operatorname{Re}\left(F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)= & \sigma_{k, \alpha}^{1}\left(t_{\text {in }}\right) \lambda_{k, \alpha}^{2}\left(t_{\text {out }}\right)+\sigma_{k, \alpha}^{2}\left(t_{\text {in }}\right) \lambda_{k, \alpha}^{1}\left(t_{\text {out }}\right) \\
& -\sigma_{k, \alpha}^{1}\left(t_{\text {out }}\right) \lambda_{k, \alpha}^{2}\left(t_{\text {in }}\right)-\sigma_{k, \alpha}^{2}\left(t_{\text {out }}\right) \lambda_{k, \alpha}^{1}\left(t_{\text {in }}\right)  \tag{4.40c}\\
\operatorname{Im}\left(F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)= & \sigma_{k, \alpha}^{2}\left(t_{\text {in }}\right) \lambda_{k, \alpha}^{2}\left(t_{\text {out }}\right)+\sigma_{k, \alpha}^{1}\left(t_{\text {out }}\right) \lambda_{k, \alpha}^{1}\left(t_{\text {in }}\right) \\
& 1\left(t_{\text {out }}\right)-\sigma_{k, \alpha}^{2}\left(t_{\text {out }}\right) \lambda_{k, \alpha}^{2}\left(t_{\text {in }}\right) . \tag{4.40d}
\end{align*}
$$

Looking at equation (4.35a), we immediately see that the time evolution of $G_{k}\left(t_{\mathrm{in}}, t_{\text {out }}\right)$ and $F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)$ decouple if $\beta_{k}$ vanishes, i.e. as is evident from equation (4.34b) $k=\pi / 2$ makes for a special case that is to be investigated separately. We shall restrict ourselves to discussing only properties of $G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)$.

### 4.3.3 Special Case $k=\pi / 2$

We directly obtain from equation (4.36a) that the equations of motion for real and imaginary part of both Green's functions, $G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)$ and $F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)$ resemble the ordinary harmonic oscillator but with a time dependent quantity $\varepsilon_{k}(t)$ (4.34a). The equations of motion read

$$
\begin{align*}
& \frac{\mathrm{d}}{\mathrm{~d} t_{\text {out }}}\binom{\operatorname{Re}\left(G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)}{\operatorname{Im}\left(G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)}=\left(\begin{array}{cc}
0 & \varepsilon_{k}\left(t_{\text {out }}\right) \\
-\varepsilon_{k}\left(t_{\text {out }}\right) & 0
\end{array}\right)\binom{\operatorname{Re}\left(G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)}{\operatorname{Im}\left(G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)}  \tag{4.41a}\\
& \frac{\mathrm{d}}{\mathrm{~d} t_{\text {out }}}\binom{\operatorname{Re}\left(F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)}{\operatorname{Im}\left(F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)}=\left(\begin{array}{cc}
0 & -\varepsilon_{k}\left(t_{\text {out }}\right) \\
\varepsilon_{k}\left(t_{\text {out }}\right) & 0
\end{array}\right)\binom{\operatorname{Re}\left(F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)}{\operatorname{Im}\left(F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)\right)} . \tag{4.41b}
\end{align*}
$$

Bearing in mind that the coupling between spin and phonon system $g$ is of a lower order of magnitude than the dimer coupling $J$, c.f. $(3.36 \mathrm{~b})$, its product with the phonon displacement $A(t)$, which is solely responsible for the time dependence of the quantity $\varepsilon_{k}$, can be regarded
as a small perturbation. Thus, to a zeroth order approximation the frequency of both harmonic oscillators is given by

$$
\begin{equation*}
\varepsilon_{\pi / 2}^{0}=J=1 \tag{4.42}
\end{equation*}
$$

Figure 4.7 depicts the Fourier spectrum of the relative, time average and average Green's function $G_{\pi / 2}^{\text {rel }}\left(0, \omega_{\text {diff }}\right), \tilde{G}_{\pi / 2}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and $\bar{G}_{\pi / 2}\left(10000 J^{-1}, \omega\right)$. We chose

$$
\begin{align*}
t_{\mathrm{in}} & =0 J^{-1}  \tag{4.43a}\\
\bar{t} & =5000 J^{-1} \tag{4.43b}
\end{align*}
$$

to receive the longest string of data possible for relative and time average Green's function. As there is no sense in neglecting any data when computing the average Green's function $\bar{G}_{\pi / 2}\left(10000 J^{-1}, \omega\right)$ we set its first argument to the maximum simulation time $t_{\max }$. In figure 4.7 (a) we find the Fourier spectra of the absolute values of the respected Green's function. Here the peak at the oscillation frequency $J=1$, which we expected from equation (4.42), is featured most prominently. Figure 4.7(b) provides the same plot but with a logarithmic ordinate in order to illustrate side peaks properly. For the relative Green's function $G_{\pi / 2}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right)$ we find those peaks at frequencies

$$
\begin{equation*}
\omega_{\mathrm{diff}}=\varepsilon_{\pi / 2}+n \omega_{0} \tag{4.44}
\end{equation*}
$$

where the parameter $n$ is an integer number. They also occur for the average Green's function $\bar{G}_{\pi / 2}\left(10000 J^{-1}, \omega\right)$ but only to first order, i.e. $n= \pm 1$. Any other side peaks vanished due to the averaging. Those side peaks proof triplon phonon interaction which is evident from the fact that the phonon frequency $\omega_{0}$ determines their position. Higher than first order side peaks are due to higher harmonics which also explains why they are not persistent over averaging. The time average Green's function shows a different behaviour as the side peaks occur at frequencies

$$
\begin{equation*}
\omega_{\mathrm{diff}}=\varepsilon_{\pi / 2}^{0}+n \frac{\omega_{0}}{2} \tag{4.45}
\end{equation*}
$$

which is due to the factor of two in the definition of the average time $\bar{t}$ in equation (2.86). All the observed Green's functions share the property of a highly suppressed interaction between phonon and spin system which is convincing given the small size of the spin-phonon coupling $g$. As for figure 4.7 (c) there is an extreme close-up of the main peak at $\varepsilon_{\pi / 2}^{0}$ for the negative imaginary part of the Green's functions. For each of them a fit according to equation $(4.32)$ is applied to the generated data points. The fit parameters for the relative Green's function $G_{\pi / 2}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right)$ read

$$
\begin{align*}
\xi & =0.000156  \tag{4.46a}\\
\chi_{0} & =0.999557 \mathrm{~J}  \tag{4.46~b}\\
\zeta & =0.000601 \mathrm{~J} \tag{4.46c}
\end{align*}
$$

which is close by the fit parameters of the time average Green's function $\tilde{G}_{\pi / 2}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$,

$$
\begin{align*}
\xi & =0.000157  \tag{4.47a}\\
\chi_{0} & =0.999556 J  \tag{4.47~b}\\
\zeta & =0.000601 J \tag{4.47c}
\end{align*}
$$



Figure 4.7: Fourier spectrum for momentum $k=\pi / 2$ of the relative, time average and average Green's function, $G_{\pi / 2}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right), \tilde{G}_{\pi / 2}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and $\bar{G}_{\pi / 2}\left(10000 J^{-1}, \omega\right)$; values: $g=0.03 J, \omega_{0}=1.3 J, a=0.4 J, \gamma=0.1 J$
and those of the average Green's $\bar{G}_{\pi / 2}\left(10000 J^{-1}, \omega\right)$ function,

$$
\begin{align*}
\xi & =0.000154  \tag{4.48a}\\
\chi_{0} & =0.999556 \mathrm{~J}  \tag{4.48b}\\
\zeta & =0.000600 \mathrm{~J} \tag{4.48c}
\end{align*}
$$

All these parameters clearly show that the Green's function are not exactly centered around $\varepsilon_{\pi / 2}^{0}$ but around a slightly smaller value which illustrates that equation (4.42) is only an approximation. The great agreement of these parameters supports the validity of the imaginary part of a Green's function as a measurement for the spectral density.

In figure 4.8 the real and imaginary part of the Fourier spectrum of all three Green's functions are depicted in their entirety. Of major importance are the imaginary parts as they are proportional to the spectral density. The real parts are simply depicted to ensure that they are not the origin of phenomena other than those also occurring in the imaginary parts. In conformity with the results obtained from figure 4.7, the side peaks are way more distinct in the relative Green's function $G_{\underline{\pi} / 2}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right)$ than in the time average or the average Green's function, $\tilde{G}_{\pi / 2}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ or $\bar{G}_{\pi / 2}\left(10000 J^{-1}, \omega\right)$. The shape of those side peaks is particularly interesting. While the deflections in both directions in case of the relative Green's function $G_{\pi / 2}^{\text {rel }}\left(0, \omega_{\text {diff }}\right)$, which are characteristic to a poorly resolved pole, the average Green's function $\bar{G}_{\pi / 2}\left(10000 J^{-1}, \omega\right)$ shows significantly smaller ordinary peaks which is most comprehensible. By contrast, the time average Green's function $\tilde{G}_{\pi / 2}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ shows two side peaks as well, however pointing in opposite directions.

In order to properly interpret results gained from the relative and time average Green's function, $G_{\pi / 2}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right)$ and $\tilde{G}_{\pi / 2}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$, we need to understand their dependence on the initial time $t_{\text {in }}$ or the average time $\bar{t}$ respectively. Therefore, figure 4.9 shows the Fourier spectra plotted with a logarithmic scale as well as a close-up of the dominant peak for the negative imaginary part for the relative and the time average Green's function, $G_{\pi / 2}^{\mathrm{rel}}\left(t_{\mathrm{in}}, \omega_{\text {diff }}\right)$ and $\tilde{G}_{\pi / 2}\left(\bar{t}, \omega_{\text {diff }}\right)$, for three different times $t_{\text {in }}$ or $\bar{t}$. The first two lines contain plots of the relative Green's function $G_{\pi / 2}^{\mathrm{rel}}\left(t_{\mathrm{in}}, \omega_{\text {diff }}\right)$ where, alongside the previously utilized initial time

$$
\begin{equation*}
t_{\mathrm{in}}=0 J^{-1} \tag{4.49a}
\end{equation*}
$$

in figures 4.7 and 4.8, functions for

$$
\begin{align*}
& t_{\mathrm{in}}=3000 J^{-1}  \tag{4.49b}\\
& t_{\mathrm{in}}=5000 J^{-1} \tag{4.49c}
\end{align*}
$$

are plotted as well. The Fourier spectrum in figure 4.9(a) clearly indicates that the offset increases with larger initial time $t_{\text {in }}$ which is reasonable given that the length of the signal to be processed in the Fourier transform has decreased as can be derived from equation (4.27). The peak's position is invariant under variation of the initial time $t_{\text {in }}$ according to our expectation. However, its shape changes slightly. Especially the side peaks are affected with the first peak on the right even changing its direction as the initial time $t_{\text {in }}$ is increased to $5000 \mathrm{~J}^{-1}$. For the explicit implementation of the fast Fourier transform in


Figure 4.8: Real and imaginary part of the Fourier spectra for momentum $k=\pi / 2$ of the relative, time average and average Green's function, $G_{\pi / 2}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right), \tilde{G}_{\pi / 2}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and $\bar{G}_{\pi / 2}\left(10000 J^{-1}, \omega\right)$; values: $g=0.03 J, \omega_{0}=1.3 J, a=0.4 J, \gamma=0.1 J$


Figure 4.9: Investigation of the dependence of relative Green's function $G_{\pi / 2}^{\mathrm{rel}}\left(t_{\mathrm{in}}, \omega_{\text {diff }}\right)$ on the initial time $t_{\text {in }}$ and of the time average Green's function $\tilde{G}_{\pi / 2}\left(\bar{t}, \omega_{\text {diff }}\right)$ on the average time $\bar{t}$ for momentum $k=\pi / 2$; values: $g=0.03 J, \omega_{0}=1.3 J, a=0.4 J, \gamma=0.1 J$

Python is unknown to us, we cannot rule out numerical effects here. Figure 4.9(b) provides fits according to equation $(4.32)$ to the generated data points for all three cases. The fit parameters for the Green's function $G_{\pi / 2}^{\text {rel }}\left(0, \omega_{\text {diff }}\right)$ can be obtained from equation (4.46). For the second Green's function under investigation $G_{\pi / 2}^{\mathrm{rel}}\left(3000 J^{-1}, \omega_{\text {diff }}\right)$ they read

$$
\begin{align*}
\xi & =0.000223  \tag{4.50a}\\
\chi_{0} & =0.999464 \mathrm{~J}  \tag{4.50b}\\
\zeta & =0.000858 \mathrm{~J} \tag{4.50c}
\end{align*}
$$

and for the third one $G_{\pi / 2}^{\mathrm{rel}}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ we receive

$$
\begin{align*}
\xi & =0.000309  \tag{4.51a}\\
\chi_{0} & =0.999463 \mathrm{~J}  \tag{4.51b}\\
\zeta & =0.001202 \mathrm{~J} . \tag{4.51c}
\end{align*}
$$

Comparing those values we note a slight shift of the peak position $\chi_{0}$ to smaller frequencies for large initial times $t_{\text {in }}$. However those changes take place in the fourth or sixth decimal place. Most evident from the plot and supported by the parameters is an increase in the half width $\zeta$ as the initial time $t_{\text {in }}$ increases. This observation becomes reasonable when we think of the quantity $\zeta$ as an inverse life time. For a shorter time of perturbation due to an increased initial time $t_{\text {in }}$ the life time naturally decreases thus leading to an increase of its inverse. These two effects also occur when the average time $\bar{t}$ of the time average Green's function $\tilde{G}_{\pi / 2}\left(\bar{t}, \omega_{\text {diff }}\right)$ is decreased but more intensely as can be seen in figures 4.9 (c) and 4.9(d). Additionally to

$$
\begin{equation*}
\bar{t}=5000 J^{-1} \tag{4.52a}
\end{equation*}
$$

we choose

$$
\begin{align*}
& \bar{t}=3000 J^{-1}  \tag{4.52b}\\
& \bar{t}=1000 J^{-1} . \tag{4.52c}
\end{align*}
$$

Figure 4.9 (c) provides again a comparison of the Fourier spectra. Besides the previously discussed increase of the offset we learn that the number of higher harmonics of spin-phonon interaction in the Fourier spectrum depends on the average time $\bar{t}$ chosen. However, there is no proportional dependence as it increases for $\tilde{G}_{\pi / 2}\left(3000 J^{-1}, \omega_{\text {diff }}\right)$ but then decreases for $\tilde{G}_{\pi / 2}\left(1000 J^{-1}, \omega_{\text {diff }}\right)$. Figure 4.9 (d) shows Lorentzian fits (4.32) applied to data generated for the negative imaginary parts of the time average Green's functions $\tilde{G}_{\pi / 2}\left(\bar{t}, \omega_{\text {diff }}\right)$. For $\tilde{G}_{\pi / 2}\left(3000 J^{-1}, \omega_{\text {diff }}\right)$ the fit parameters are given by

$$
\begin{align*}
\xi & =0.000254  \tag{4.53a}\\
\chi_{0} & =0.999346 \mathrm{~J}  \tag{4.53b}\\
\zeta & =0.001001 \mathrm{~J}, \tag{4.53c}
\end{align*}
$$

and for $\tilde{G}_{\pi / 2}\left(1000 J^{-1}, \omega_{\text {diff }}\right)$ by

$$
\begin{align*}
\xi & =0.000779  \tag{4.54a}\\
\chi_{0} & =0.998298 \mathrm{~J}  \tag{4.54b}\\
\zeta & =0.003009 \mathrm{~J} \tag{4.54c}
\end{align*}
$$

Comparing these results to equation (4.47), the numbers are consistent with the corresponding plot. Here, the increase of the half width $\zeta$ becomes most obvious. For the data strings of the time average Green's functions $\tilde{G}_{\pi / 2}\left(\bar{t}, \omega_{\text {diff }}\right)$ are only half as long as for the relative Green's functions $G_{\pi / 2}^{\mathrm{rel}}\left(t_{\text {in }}, \omega_{\text {diff }}\right)$ as worked out in chapter 4.3.1 this effect comes to no surprise.

### 4.3.4 Case $k \neq \pi / 2$

Having analysed the special case, we now turn to the more general condition where the system of ordinary differential equations (4.35a) does not decouple. We choose

$$
\begin{equation*}
k=\frac{\pi}{4} . \tag{4.55}
\end{equation*}
$$

Because

$$
\begin{equation*}
\beta_{k}<\varepsilon_{k} \tag{4.56}
\end{equation*}
$$

holds for any momentum $k$ as can be derived from equation (4.34), we may still take the zeroth order approximation of the quantity $\varepsilon_{\pi / 4}$ as a first estimation on where to expect the dominant peak in the Fourier spectrum. Equation (4.35a) then describes a linear system of differential equations with constant coefficients of which the solution can be determined as

$$
\begin{align*}
\frac{\mathrm{d}}{\mathrm{~d} t_{\text {out }}}\binom{G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)}{F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)}= & -\frac{\text { ie } \sqrt{\left(\varepsilon_{\pi / 4}^{0}\right)^{2}-4 \beta_{\pi / 4}^{2} t_{\text {out }}}}{2 \sqrt{\left(\varepsilon_{\pi / 4}^{0}\right)^{2}-4 \beta_{\pi / 4}^{2}}}\binom{-\varepsilon_{\pi / 4}^{0}+\sqrt{\left(\varepsilon_{\pi / 4}^{0}\right)^{2}-4 \beta_{\pi / 4}^{2}}}{2 \beta_{\pi / 4}} \\
& \left.+\frac{\text { ie } \begin{array}{l}
-\sqrt{\left(\varepsilon_{\pi / 4}^{0}\right)^{2}-4 \beta_{\pi / 4}^{2} t_{\text {out }}} \\
2 \sqrt{\left(\varepsilon_{\pi / 4}^{0}\right)^{2}-4 \beta_{\pi / 4}^{2}}
\end{array}\binom{-\varepsilon_{\pi / 4}^{0}-\sqrt{\left(\varepsilon_{\pi / 4}^{0}\right)^{2}-4 \beta_{\pi / 4}^{2}}}{2 \beta_{\pi / 4}}}{} .=\begin{array}{c}
\end{array}\right) \tag{4.57}
\end{align*}
$$

in a straight forward manner for we are dealing with a textbook case. Thus, the expression

$$
\begin{equation*}
\sqrt{\left(\varepsilon_{\pi / 4}^{0}\right)^{2}-4 \beta_{\pi / 4}^{2}} \approx 0.818 J \tag{4.58}
\end{equation*}
$$

with

$$
\begin{equation*}
\varepsilon_{\pi / 4}^{0}=J-\frac{J^{\prime}}{2^{\frac{3}{2}}} \tag{4.59}
\end{equation*}
$$

and

$$
\begin{equation*}
\beta_{\pi / 4}=-\frac{J^{\prime}}{2^{\frac{5}{2}}} \tag{4.60}
\end{equation*}
$$

describes the frequency of the oscillation of both Green's functions, $G_{k}\left(t_{\text {in }}, t_{\text {out }}\right)$ and $F_{k}\left(t_{\text {in }}, t_{\text {out }}\right)$.
The Fourier spectrum depicted in figure 4.10(a) confirms our assumption. Note that throughout this section we chose the same initial times $t_{\text {in }}$ and average times $\bar{t}$ as in the


Figure 4.10: Fourier spectrum for momentum $k=\pi / 4$ of the relative, time average and average Green's function, $G_{\pi / 4}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right), \tilde{G}_{\pi / 4}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and $\bar{G}_{\pi / 4}\left(10000 J^{-1}, \omega\right)$; values: $g=0.03 J, \omega_{0}=1.3 J, a=0.4 J, \gamma=0.1 J$
previous one, c.f. equations (4.43), (4.49) or (4.52) respectively. We also obtain side peaks at frequencies

$$
\begin{equation*}
\omega_{\text {diff }}=\varepsilon_{\pi / 4}+n \omega_{0} \tag{4.61}
\end{equation*}
$$

as for the decoupled case. The parameter $n$ is again taken to be an integer number. Inspecting the Fourier spectrum via a logarithmic display shown in figure 4.7 (b) we learn that there are also peaks occurring at frequencies

$$
\begin{equation*}
\omega_{\text {diff }}=-\varepsilon_{\pi / 4}+n \omega_{0} \tag{4.62}
\end{equation*}
$$

for an integer $n$ number in contrast to our previous findings. Those peaks, however, are highly suppressed compared to the common ones. Because of the negative sign of $\varepsilon_{\pi / 4}$ we take them as an indicator of antitriplon interaction which explains their suppression. In compliance with the results obtained for the decoupled case the frequencies are given by

$$
\begin{equation*}
\omega_{\text {diff }}= \pm \varepsilon_{\pi / 4}+n \frac{\omega_{0}}{2} \tag{4.63}
\end{equation*}
$$

for the time average Green's function $\tilde{G}_{\pi / 4}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$. Any other properties are also found for the decoupled case and, hence, discussed in the previous section. Figure 4.10(c) shows the Lorentzian fits according to (4.32) applied to the relative time average and average Green's function, $G_{\pi / 4}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right), \tilde{G}_{\pi / 4}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and $\bar{G}_{\pi / 4}\left(10000 J^{-1}, \omega\right)$. Trough the fit parameters

$$
\begin{align*}
\xi & =0.000158  \tag{4.64a}\\
\chi_{0} & =0.801202 \mathrm{~J}  \tag{4.64b}\\
\zeta & =0.000600 \mathrm{~J} \tag{4.64c}
\end{align*}
$$

for the reative Green's function $G_{\pi / 4}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right)$,

$$
\begin{align*}
\xi & =0.000160  \tag{4.65a}\\
\chi_{0} & =0.801158 J  \tag{4.65b}\\
\zeta & =0.000600 J \tag{4.65c}
\end{align*}
$$

for the time average Green's function $\tilde{G}_{\pi / 4}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and

$$
\begin{align*}
\xi & =0.000157  \tag{4.66a}\\
\chi_{0} & =0.801158 J  \tag{4.66b}\\
\zeta & =0.000600 J \tag{4.66c}
\end{align*}
$$

for the average Green's function $\bar{G}_{\pi / 4}\left(10000 J^{-1}, \omega\right)$ we see that the actual peak position is at a lower frequency than our estimation (4.58) predicted which clearly indicates that for a coupled case, such as this one, more effects play a larger role in its dynamics than for the decoupled case.
Figure 4.11 is, as in the previous chapter, to reassure that the absolute values of the Green's functions did not hide other effects that could only be observed in the real or imaginary


Figure 4.11: Real and imaginary part of the Fourier spectra for momentum $k=\pi / 4$ of the relative, time average and average Green's function, $G_{\pi / 4}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right), \tilde{G}_{\pi / 4}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and $\bar{G}_{\pi / 4}\left(10000 J^{-1}, \omega\right)$; values: $g=0.03 J, \omega_{0}=1.3 J, a=0.4 J, \gamma=0.1 J$
part. For we do not encounter any deviations from the anticipated position of the major peak, we take our hitherto existing results as confirmed. Aside from the one major peak the side peaks are strongly suppressed just like in the previous chapter which guides our attention to the final analysis.

In figure 4.12 we investigate the dependence of the relative Green's function $G_{\pi / 4}^{\mathrm{rel}}\left(t_{\text {in }}, \omega_{\text {diff }}\right)$ on the initial time $t_{\text {in }}$ and of the time average Green's function $\tilde{G}_{\pi / 4}\left(\bar{t}, \omega_{\text {diff }}\right)$ on the average time $\bar{t}$. In figure 4.12 (a) the Fourier spectrum on a logarithmic scale for the relative Green's function $G_{\pi / 4}^{\text {rel }}\left(t_{\text {in }}, \omega_{\text {diff }}\right)$ is shown for different initial times $t_{\text {in }}$. As it increases the offset does as well. There is no visible change in the peak positions but again the shapes vary for each function. Lorentzian fits (4.32) applied to data generated for the negative imaginary part are depicted in figure 4.12 (b). The fit parameters for $G_{\pi / 4}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right)$ can be found in equation (4.64) while those for $G_{\pi / 4}^{\mathrm{rel}}\left(3000 J^{-1}, \omega_{\text {diff }}\right)$ and $G_{\pi / 4}^{\mathrm{rel}}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ are determined by

$$
\begin{align*}
\xi & =0.000225  \tag{4.67a}\\
\chi_{0} & =0.801071 \mathrm{~J}  \tag{4.67b}\\
\zeta & =0.000858 \mathrm{~J} \tag{4.67c}
\end{align*}
$$

and

$$
\begin{align*}
\xi & =0.000315  \tag{4.68a}\\
\chi_{0} & =0.801064 J  \tag{4.68b}\\
\zeta & =0.001202 J \tag{4.68c}
\end{align*}
$$

respectively. For an increasing initial time $t_{\text {in }}$ we obtain the expectable result of an increasing half width $\zeta$. In figures 4.12 (c) and 4.12 (d) the time average Green's function $\tilde{G}_{\pi / 4}\left(\bar{t}, \omega_{\text {diff }}\right)$ is studied once more. Just like in the previous section the effects are more intense especially concerning the increase of the offset in the logarithmic Fourier spectrum given in figure 4.12 (c). Fits according to equation (4.32) for data of the negative imaginary part yield the parameters in equation (4.65a) for $\widetilde{G}_{\pi / 4}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$,

$$
\begin{align*}
\xi & =0.000257  \tag{4.69a}\\
\chi_{0} & =0.800948 \mathrm{~J}  \tag{4.69b}\\
\zeta & =0.001002 \mathrm{~J} \tag{4.69c}
\end{align*}
$$

for $\tilde{G}_{\pi / 4}\left(3000 J^{-1}, \omega_{\text {diff }}\right)$ and

$$
\begin{align*}
\xi & =0.000792  \tag{4.70a}\\
\chi_{0} & =0.799902 J  \tag{4.70b}\\
\zeta & =0.003008 J \tag{4.70c}
\end{align*}
$$

for $\tilde{G}_{\pi / 4}\left(1000 J^{-1}, \omega_{\text {diff }}\right)$. As before a slight shift to smaller frequencies $\chi_{0}$ and an increasing half width $\zeta$ are the key results of our findings.


Figure 4.12: Investigation of the dependence of relative Green's function $G_{\pi / 4}^{\mathrm{rel}}\left(t_{\text {in }}, \omega_{\text {diff }}\right)$ on the initial time $t_{\text {in }}$ and of the time average Green's function $\tilde{G}_{\pi / 4}\left(\bar{t}, \omega_{\text {diff }}\right)$ on the average time $\bar{t}$ for momentum $k=\pi / 4$; values: $g=0.03 J, \omega_{0}=1.3 J, a=0.4 J, \gamma=0.1 J$

### 4.3.5 Summary

We managed to derive the equations of motion for two kinds of Green's functions describing the insertion of a triplon into and its subsequent removal from the system or the insertion of a triplon and a subsequent insertion of another triplon of opposite momentum into the system. Analysing only the former we proposed two different cases by sole inspection of the equations of motion. For the decoupled case we found proof of interaction between triplons and phonons while for the general case also antitriplons as well as interaction were observed, however on a much lower order of magnitude. Those results were obtained for all three different Green's functions under consideration, the relative, the time average and the average Green's function. While the relative and average Green's functions only differed in the absence of higher harmonics of phonon modes in the latter, the time average Green's function showed its side peaks at intervals only half as large as those for the two other Green's function. We traced this effect back to a factor in the definition of the average time. A further investigation of the dependence of the relative Green's function on the initial time and of the time average Green's function on the average time showed that both an increase in the initial or a decrease of the average time lead to a larger offset in the Fourier spectrum as well as a shift to smaller frequencies and a broadening of the negative imaginary part proportional to the spectral density. The applied fits, however, proved that the shape is at all times given by a Lorentzian.

## 5 Conclusion

### 5.1 Summary

For this thesis we developed a simple model offering the possibility to investigate quasi steady states. We took existing solids like $\mathrm{CuGeO}_{3}$ as a basis, thus proposing a dimerized spin $1 / 2$ chain as the principle element with an underlying phonon system as its foundation and mediator of excitation. A laser served as origin of the driving. Resonance effects were supposed to be avoided by damping the phonon system using the Lindblad formalism. Applying the bond operator representation for dimers thus introducing the hardcore bosons triplons and singlets, we obtained in approximation a bilinear Hamiltonian from which we derived the equations of motion for the phonon displacement, the phonon momentum, the phonon density as well as the triplon density. Exciting the system in resonance with the phonon frequency was to drive the spin system into a quasi steady state, i.e. the triplon density was to oscillate around a value differing significantly from its initial value while obeying to the hardcore constraint stating there can only be one triplon at a time at each lattice site.

However, early on in the process we encountered effects of divergent behaviour that were unexpected and counterintuitive clearly violating the hardcore constraint with the triplon density taking values of several orders of magnitude larger than the theoretically maximum value of one. We identified this phenomenon as an effect of a resonance disaster not caused by the phonon system, where the damping worked well, as the description by a damped driven harmonic oscillator characterised the time evolution perfectly fine, but rather by the triplon system itself. Whenever the laser frequency was a multiple of the triplon band frequencies we found effects of divergence at a much earlier stage compared to other frequencies. We measured this stability of our system by the introduction of a critical value of the spin-phonon coupling which when being exceeded resulted in divergences. The quotient of laser amplitude and spin damping parameter squared and the laser and phonon frequency served as other parameters in our characterisation.

For the laser and phonon frequency we identified three regions depending on their relation to the triplon band, the in band, the beneath band or adiabatic and the above band or anti-adiabatic regime. In agreement with our assumption we found diverging systems for all in band frequencies and their multiples in the adiabatic regime whereas the anti-adiabatic regime proved to be comparably stable. Yet only in the adiabatic regime the triplon densities took average values reasonably larger than their initial value which was the desired state. For obvious reasons we needed to restrict the applicability of our systems to non-divergent regions.

In these regimes, however, the model provides convenient results. Not only, as we already mentioned, do we obtain the desired quasi steady states but also can we identify interactions between spin and phonon system. Therefore, we observed three different Green's functions, the relative, the time average and the average Green's function. Just like for the expectation values of the spin and phonon system we set up a system of ordinary differential equations for the Green's functions distinguishing between two types, one where a triplon is inserted at a certain time and removed later in the process and a second one describing the insertion of two triplons of opposite momentum after one another.

Investigating only the former we find a dominant peak in the Fourier spectra of the relative, the time average and the average Green's function at the corresponding triplon frequency. For the relative and average Green's function we identify side peaks at the triplon frequency plus or minus the phonon frequency which illustrates the interaction between both systems. As these effects are suppressed by several orders of magnitude the weakness of the coupling is further illustrated. Whereas higher harmonics indicated by peaks at the triplon frequency plus or minus a multiple of the phonon frequency vanish for the average Green's function, they occur for the relative Green's function but again being suppressed by at least one other order of magnitude. The time average Green's function shows the exact same behaviour but with side peaks occurring at the triplon frequency plus or minus multiples of half the phonon frequency which is due to a factor of two in the definition of the average time.

For the special case of momentum $k=\pi / 2$, where the equations of motion of the two types of Green's functions decouple, this is the whole spectrum we obtain. If, however, the momentum is $k \neq \pi / 2$, we find peaks at the negative triplon frequency plus or minus multiples of the phonon frequency for the relative and average or half the phonon frequency for the time average Green's function. Again these effects are suppressed by several orders of magnitude compared to the dominant peak. We take them as proof of the interaction of antitriplons with the phonon system. This discovery of antitriplons is indeed astonishing for they do not occur in the original Hamiltonian. A further investigation as part of further research on their behaviour could lead to new insights of properties of dimerized spin chains.

### 5.2 Discussion

The occurrence of diverging effects is of course unpleasant. We need to stress that we cannot omit any parts of our model as it already consists of only basic components. Still, the corresponding set of equations of motion is highly non-trivial and cannot be solved analytically. Therefore, any enhancements will most certainly further complicate the system making sources of problems harder to find. Even the model at its present state provides several obstacles that could not be explained entirely or found to be generic. Having always the resonance effects of the spin system as a possible source of those effects in mind, their validity as proof of new physics is hard to judge just like the applicability of our model. Of particular difficulty is the absence of a rule to when diverging effects occur. Certain systems showed these effects at very early times while others were found to be persistent over a long period of time but not infinitely. Therefore, we cannot guarantee that systems we identified as stable might not diverge later in time. The identification of those frequencies
where divergences occurred as multiples of triplon band frequencies worked well for in and beneath band cases. As there exist, however, critical values to the spin-phonon coupling above the band, fully understanding the model requires further investigation here.
Despite the model clearly failing for certain parameter sets it nevertheless shows that, in principle, the main goal is achievable. We indeed did succeed in driving the spin system into a non-trivial quasi steady state which differed significantly from its initial value. We found beneath band cases as an appropriate regime to obtain meaningful results. From a purely mathematical point of view it was even possible to create such quasi steady states in the above band regime but only for non-physical parameter sets. Applying reasonable parameters here led to a quasi steady state of which the average value was so little above the initial value that there was a significant overlap in the oscillations. In contrast to the adiabatic regime, here a decrease in damping could help improve the model.
Thus, a direct dependence of the damping on the frequency might be fruitful. This proposal leads to the introduction of temperature to the model for we have only considered the case of zero temperature so far. In summary, we are dealing with a model with indisputable impairments but still holding rich perspectives in the investigation of non-equilibrium dynamics. Further research may for example also be conducted on systems, where the laser's frequency differs slightly from the phonon frequency, as well as on the investigation of the second type of Green's function where two triplons of opposite momentum are inserted into the system.

### 5.3 Outlook

The most promising ansatz to improving the model involves the addition of a damping to the spin system in hopes of avoiding resonance phenomena that we identified as the source of the divergences. However, here we face some challenges as well for in theory the damping to each momentum could be chosen differently, i.e. the suppression of certain modes is possible. The simplest approach, of course, is to apply a constant momentum-independent damping. A complete analysis will be left for further investigations to come.

## A Derivation of the Hamiltonian

We provide some additional calculations to derive the full Hamiltonian in its final form (3.19). We focus on the spin system as calculations for the phonon system are drastically simplified by the approximations we outlined in section 3.2 or analogous to some of the ones we present here.

First, we use Sachdev's bond operator representation (2.9) to calculate

$$
\begin{align*}
\overrightarrow{\boldsymbol{S}}_{i}^{1} \cdot \overrightarrow{\boldsymbol{S}}_{i}^{2}= & \sum_{\alpha} \boldsymbol{S}_{\alpha, i}^{1} \boldsymbol{S}_{\alpha, i}^{2}  \tag{A.1a}\\
= & \frac{1}{4} \sum_{\alpha}\left(s_{i}^{\dagger} \boldsymbol{t}_{\alpha, i}+\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{s}_{i}-\mathrm{i} \sum_{\beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i}\right) \\
& \cdot\left(-s_{i}^{\dagger} \boldsymbol{t}_{\alpha, i}-\boldsymbol{t}_{\alpha, i}^{\dagger} s_{i}-\mathrm{i} \sum_{\beta^{\prime}, \gamma^{\prime}} \varepsilon_{\alpha \beta^{\prime} \gamma^{\prime}} \boldsymbol{t}_{\beta^{\prime}, i}^{\dagger} \boldsymbol{t}_{\gamma^{\prime}, i}\right)  \tag{A.1b}\\
= & \frac{1}{4} \sum_{\alpha}\left(-\boldsymbol{s}_{i}^{\dagger} \boldsymbol{t}_{\alpha, i} \boldsymbol{s}_{i}^{\dagger} \boldsymbol{t}_{\alpha, i}-\boldsymbol{s}_{i}^{\dagger} \boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{s}_{i}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{s}_{i} s_{i}^{\dagger} \boldsymbol{t}_{\alpha, i}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{s}_{i} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{s}_{i}\right) \\
& -\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} s_{i}^{\dagger} \boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i}-\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{s}_{i} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \\
& +\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{s}_{i}^{\dagger} \boldsymbol{t}_{\alpha, i}+\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{s}_{i} \\
& -\frac{1}{4} \sum_{\alpha, \beta, \gamma, \beta^{\prime}, \gamma^{\prime}} \varepsilon_{\alpha \beta \gamma} \varepsilon_{\alpha \beta^{\prime} \gamma^{\prime}} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\beta^{\prime}, i}^{\dagger} \boldsymbol{t}_{\gamma^{\prime}, i}  \tag{A.1c}\\
= & \frac{1}{4} \sum_{\alpha}\left(-\boldsymbol{s}_{i}^{\dagger} s_{i}^{\dagger} \boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i}-2 s_{i}^{\dagger} s_{i} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{s}_{i} s_{i}-\boldsymbol{s}_{i}^{\dagger} s_{i}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i}\right) \\
& +\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{s}_{i}^{\dagger}\left(\boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\alpha, i}-\boldsymbol{t}_{\alpha, i} i_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i}\right) \\
& +\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{s}_{i}\left(\boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} i_{\alpha, i}^{\dagger}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{i}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i}\right) \\
& -\frac{1}{4} \sum_{\beta, \gamma, \beta^{\prime}, \gamma^{\prime}}\left(\delta_{\beta, \beta^{\prime}} \delta_{\gamma, \gamma^{\prime}}-\delta_{\beta, \gamma^{\prime}} \delta_{\gamma, \beta^{\prime}}\right) \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\beta^{\prime}, i}^{\dagger} \boldsymbol{t}_{\gamma^{\prime}, i}  \tag{A.1d}\\
= & \frac{1}{4} \sum_{\alpha}\left(-\boldsymbol{s}_{i}^{\dagger} s_{i}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i}\right) \\
& +\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} s_{i}^{\dagger}\left(\boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\alpha, i}-\boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\alpha, i}-\delta_{\alpha, \beta} \boldsymbol{t}_{\gamma, i}\right) \\
& +\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} s_{i}\left(\boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} i_{\alpha, i}^{\dagger}-\boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\alpha, i}^{\dagger}-\delta_{\alpha, \gamma} \boldsymbol{t}_{\beta, i}^{\dagger}\right)
\end{align*}
$$

$$
\begin{align*}
& -\frac{1}{4} \sum_{\beta, \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i}+\frac{1}{4} \sum_{\beta, \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} t_{\gamma, i}^{\dagger} \boldsymbol{t}_{\beta, i}  \tag{A.1e}\\
= & -\frac{3}{4} s_{i}^{\dagger} s_{i}-\frac{1}{4} \sum_{\alpha} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i} \\
& -\frac{1}{4} \sum_{\beta, \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} t_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\gamma, i}-\frac{1}{4} \sum_{\beta, \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \delta_{\beta, \gamma}+\frac{1}{4} \sum_{\beta, \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{i}_{\beta, i}+\frac{1}{4} \sum_{\beta, \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\beta, i} \\
= & -\frac{3}{4} s_{i}^{\dagger} s_{i}-\frac{1}{4} \sum_{\alpha} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i}  \tag{A.1f}\\
& -\frac{1}{4} \sum_{\alpha} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i}+\frac{1}{4} \sum_{\beta, \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\beta, i} \boldsymbol{t}_{\gamma, i}^{\dagger} \boldsymbol{t}_{\gamma, i}-\frac{1}{4} \sum_{\beta, \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \delta_{\beta, \gamma}+\frac{3}{4} \sum_{\alpha} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i}  \tag{A.1g}\\
= & -\frac{3}{4} s_{i}^{\dagger} s_{i}-\frac{1}{4} \sum_{\alpha} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i} \\
= & -\frac{1}{4} \sum_{\alpha} \boldsymbol{s}_{i}^{\dagger} \boldsymbol{t}_{\alpha, i}^{\dagger}+\frac{1}{4} \boldsymbol{t}_{\alpha, i}+\frac{1}{4}\left(\sum_{\alpha} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i}\right.  \tag{A.1h}\\
= & -\frac{3}{4}\left(1-\sum_{\alpha, i} \boldsymbol{t}_{\alpha, i}^{\dagger}-\frac{1}{4} \sum_{\alpha} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i}+\frac{3}{4} \sum_{\alpha} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i}\right.  \tag{A.1i}\\
= & -\frac{3}{4}+\sum_{\alpha} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i} . \tag{A.1j}
\end{align*}
$$

Note that this result is exact.
Next, we calculate

$$
\begin{align*}
\overrightarrow{\boldsymbol{S}}_{i}^{2} \cdot \overrightarrow{\boldsymbol{S}}_{i+1}^{1}= & \sum_{\alpha} \boldsymbol{S}_{\alpha, i}^{2} \boldsymbol{S}_{\alpha, i+1}^{1}  \tag{A.2a}\\
= & \frac{1}{4} \sum_{\alpha}\left(-\boldsymbol{t}_{\alpha, i}-\boldsymbol{t}_{\alpha, i}^{\dagger}-\mathrm{i} \sum_{\beta^{\prime}, \gamma^{\prime}} \varepsilon_{\alpha \beta^{\prime} \gamma^{\prime}} \boldsymbol{t}_{\beta^{\prime}, i}^{\dagger} \boldsymbol{t}_{\gamma^{\prime}, i}\right) \\
& \cdot\left(\boldsymbol{t}_{\alpha, i+1}+\boldsymbol{t}_{\alpha, i+1}^{\dagger}-\mathrm{i} \sum_{\beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\beta, i+1}^{\dagger} \boldsymbol{t}_{\gamma, i+1}\right)  \tag{A.2b}\\
= & \frac{1}{4} \sum_{\alpha}\left(-\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i+1}-\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i+1}^{\dagger}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i+1}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{s}_{i} \boldsymbol{t}_{\alpha, i+1}^{\dagger} \boldsymbol{s}_{i+1}\right) \\
& -\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\alpha, i+1}-\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\alpha, i+1}^{\dagger} \\
& +\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\beta, i+1}^{\dagger} \boldsymbol{t}_{\gamma, i+1}+\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\beta, i+1}^{\dagger} \boldsymbol{t}_{\gamma, i+1} \\
& -\frac{1}{4} \sum_{\alpha, \beta, \gamma, \beta^{\prime}, \gamma^{\prime}} \varepsilon_{\alpha \beta \gamma} \varepsilon_{\alpha \beta^{\prime} \gamma^{\prime}} \boldsymbol{t}_{\beta^{\prime}, i}^{\dagger} \boldsymbol{t}_{\gamma^{\prime}, i} \boldsymbol{t}_{\beta, i+1}^{\dagger} \boldsymbol{t}_{\gamma, i+1}  \tag{A.2c}\\
= & \frac{1}{4} \sum_{\alpha}\left(-\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i+1}-\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i+1}^{\dagger}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i+1}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i+1}^{\dagger}\right)
\end{align*}
$$

$$
\begin{align*}
& -\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\alpha, i+1}-\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, \boldsymbol{i}} \boldsymbol{i}_{\alpha, i+1}^{\dagger} \\
& +\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\beta, i+1}^{\dagger} \boldsymbol{t}_{\gamma, i+1}+\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\beta, i+1}^{\dagger} \boldsymbol{t}_{\gamma, i+1} \\
& -\frac{1}{4} \sum_{\beta, \gamma, \beta^{\prime}, \gamma^{\prime}}\left(\delta_{\beta, \beta^{\prime}} \delta_{\gamma, \gamma^{\prime}}-\delta_{\beta, \gamma^{\prime}} \delta_{\gamma, \beta^{\prime}}\right) \boldsymbol{t}_{\beta^{\prime}, i}^{\dagger} \boldsymbol{t}_{\gamma^{\prime}, i} \boldsymbol{t}_{\beta, i+1}^{\dagger} \boldsymbol{t}_{\gamma, i+1}  \tag{A.2d}\\
= & \frac{1}{4} \sum_{\alpha}\left(-\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i+1}-\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i+1}^{\dagger}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i+1}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i+1}^{\dagger}\right) \\
& -\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma}{ }_{\gamma}^{\dagger} \boldsymbol{t}_{\beta, i} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\alpha, i+1}-\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, \boldsymbol{i}} \boldsymbol{t}_{\alpha, i+1}^{\dagger} \\
& +\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\beta, i+1}^{\dagger} \boldsymbol{t}_{\gamma, i+1}+\frac{1}{4} \mathrm{i} \sum_{\alpha, \beta, \gamma} \varepsilon_{\alpha \beta \gamma} \boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\beta, i+1}^{\dagger} \boldsymbol{t}_{\gamma, i+1} \\
& -\frac{1}{4} \sum_{\beta, \gamma} \boldsymbol{t}_{\beta, i}^{\dagger} \boldsymbol{t}_{\gamma, i} \boldsymbol{t}_{\beta, i+1}^{\dagger} \boldsymbol{t}_{\gamma, i+1}+\frac{1}{4} \sum_{\beta, \gamma} \boldsymbol{t}_{\gamma, i}^{\dagger} \boldsymbol{t}_{\beta, i} \boldsymbol{t}_{\beta, i+1}^{\dagger} \boldsymbol{t}_{\gamma, i+1} \tag{A.2e}
\end{align*}
$$

where we have made use of the triplon operator representation (2.15) for it is easier to include the local hard core constraint (2.10) as part of the commutation relation (2.17) with different sites being discussed. Taking only bilinear terms into account, we obtain

$$
\begin{equation*}
\overrightarrow{\boldsymbol{S}}_{i}^{2} \cdot \overrightarrow{\boldsymbol{S}}_{i+1}^{1}=\frac{1}{4} \sum_{\alpha}\left(-\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i+1}-\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i+1}^{\dagger}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i+1}-\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i+1}^{\dagger}\right) \tag{A.3}
\end{equation*}
$$

Fourier transforming both expressions into momentum space via equation (3.9) using

$$
\begin{equation*}
\delta_{k, l}=\frac{1}{N} \sum_{i} \mathrm{e}^{\mathrm{i}(k-l) r_{i}} \tag{A.4}
\end{equation*}
$$

yields

$$
\begin{equation*}
\sum_{i} \overrightarrow{\boldsymbol{S}}_{i}^{1} \cdot \overrightarrow{\boldsymbol{S}}_{i}^{2}=-\frac{3}{4} N+\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha} \tag{A.5}
\end{equation*}
$$

as well as

$$
\begin{align*}
\sum_{i} \overrightarrow{\boldsymbol{S}}_{i}^{2} \cdot \overrightarrow{\boldsymbol{S}}_{i+1}^{1}= & -\frac{1}{4} \sum_{i, \alpha}\left(\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i+1}+\boldsymbol{t}_{\alpha, i} \boldsymbol{t}_{\alpha, i+1}^{\dagger}+\boldsymbol{t}_{\alpha, i}^{\dagger} \boldsymbol{t}_{\alpha, i+1}+\boldsymbol{t}_{\alpha, 1}^{\dagger} \boldsymbol{t}_{\alpha, i+1}^{\dagger}\right)  \tag{A.6a}\\
= & -\frac{1}{4} \frac{1}{N} \sum_{i, k, k^{\prime}, \alpha}\left(\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{k^{\prime}, \alpha} \mathrm{e}^{\mathrm{i}\left(k+k^{\prime}\right)} \mathrm{e}^{\mathrm{i} k^{\prime}}+\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{k^{\prime}, \alpha}^{\dagger} \mathrm{e}^{\mathrm{i}\left(k-k^{\prime}\right)} \mathrm{e}^{-\mathrm{i} k^{\prime}}\right. \\
& \left.+\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha} \mathrm{e}^{\mathrm{i}\left(k^{\prime}-k\right)} \mathrm{e}^{\mathrm{i} k^{\prime}}+\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha}^{\dagger} \mathrm{e}^{\mathrm{i}\left(-k-k^{\prime}\right)} \mathrm{e}^{-\mathrm{i} k^{\prime}}\right)  \tag{A.6b}\\
= & -\frac{1}{4} \sum_{k, \alpha}\left(\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha} \mathrm{e}^{-\mathrm{i} k}+\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \mathrm{e}^{-\mathrm{i} k}+\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha} \mathrm{e}^{\mathrm{i} k}+\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger} \mathrm{e}^{\mathrm{i} k}\right)  \tag{A.6c}\\
= & -\frac{1}{4} \sum_{k, \alpha}\left(\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha} \mathrm{e}^{-\mathrm{i} k}+2 \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha} \mathrm{e}^{\mathrm{i} k}+\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger} \mathrm{e}^{\mathrm{i} k}+\mathrm{e}^{-\mathrm{i} k}\right)  \tag{A.6d}\\
= & -\frac{1}{8} \sum_{k, \alpha}\left(\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}\left(\mathrm{e}^{\mathrm{i} k}+\mathrm{e}^{-\mathrm{i} k}\right)+2 \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\left(\mathrm{e}^{\mathrm{i} k}+\mathrm{e}^{-\mathrm{i} k}\right)\right.
\end{align*}
$$

$$
\begin{align*}
& \left.+\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}\left(\mathrm{e}^{\mathrm{i} k}+\mathrm{e}^{-\mathrm{i} k}\right)\right)-\frac{1}{8} \sum_{k, \alpha}\left(\mathrm{e}^{\mathrm{i} k}+\mathrm{e}^{-\mathrm{i} k}\right)  \tag{A.6e}\\
= & -\frac{1}{4} \sum_{k, \alpha} \cos (k)\left(\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}+2 \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}+\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}\right)-\frac{3}{4} \sum_{k} \cos (k) . \tag{A.6f}
\end{align*}
$$

We have eventually replaced $k$ by $-k$ and multiplied by 1 .
Thus, we obtain

$$
\begin{align*}
\boldsymbol{H}_{\mathrm{S}}= & J \sum_{k, \alpha} \boldsymbol{t}_{\alpha, k}^{\dagger} \boldsymbol{t}_{\alpha, k}-\frac{1}{4} J^{\prime} \sum_{k, \alpha} \cos (k)\left(\boldsymbol{t}_{\alpha, k} \boldsymbol{t}_{\alpha,-k}+2 \boldsymbol{t}_{\alpha, k}^{\dagger} \boldsymbol{t}_{\alpha, k}+\boldsymbol{t}_{\alpha, k}^{\dagger} \boldsymbol{t}_{\alpha,-k}^{\dagger}\right)  \tag{A.7}\\
& -\frac{3}{4} J N-\frac{3}{4} J^{\prime} \sum_{k} \cos (k)
\end{align*}
$$

of which the constant part is of no interest concerning the dynamics.

## B Derivation of the Equations of Motion

Based on the Hamiltonian $\boldsymbol{H}$ in equation (3.19) we calculate the equations of motion for the observables defined in $(\sqrt[3.22]{ })$ and $(\sqrt[3.23]{ })$. We make use of the relations

$$
\begin{align*}
{\left[\boldsymbol{X}, \boldsymbol{X}^{\prime}\right] } & =-\left[\boldsymbol{X}^{\prime}, \boldsymbol{X}\right]  \tag{B.1}\\
{\left[z \boldsymbol{X}, \boldsymbol{X}^{\prime}\right] } & =z\left[\boldsymbol{X}, \boldsymbol{X}^{\prime}\right]  \tag{B.2}\\
{\left[\boldsymbol{X} \boldsymbol{X}^{\prime}, \boldsymbol{X}^{\prime \prime}\right] } & =\boldsymbol{X}\left[\boldsymbol{X}^{\prime}, \boldsymbol{X}^{\prime \prime}\right]+\left[\boldsymbol{X}, \boldsymbol{X}^{\prime \prime}\right] \boldsymbol{X}^{\prime} \tag{B.3}
\end{align*}
$$

where $\boldsymbol{X}, \boldsymbol{X}^{\prime}$ and $\boldsymbol{X}^{\prime \prime}$ are operators and $z$ is a complex number.
We begin with the spin sector, i.e. $U_{k}(t), V_{k}(t)$ and $W_{k}(t)$. Here, the Heisenberg equation of motion (2.29) is used. For $U_{k}(t)$ and $V_{k}(t)$ we need the commutator

$$
\begin{align*}
& {\left[\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}, \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}\right]=\sum_{k, \alpha}\left[\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}, \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger}\right] \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}} }  \tag{B.4a}\\
= & \sum_{k, \alpha} \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}} \delta_{-k, k^{\prime}} \delta_{\alpha, \alpha^{\prime}}+\sum_{k, \alpha} \boldsymbol{t}_{-k, \alpha} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}} \delta_{k, k^{\prime}} \delta_{\alpha, \alpha^{\prime}}=2 \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}} \tag{B.4b}
\end{align*}
$$

as well as

$$
\begin{align*}
& {\left[\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}, \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}\right]=\sum_{k, \alpha} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger}\left[\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}, \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}\right] }  \tag{B.5a}\\
= & -\sum_{k, \alpha} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k, \alpha}^{\dagger} \delta_{-k, k^{\prime}} \delta_{\alpha, \alpha^{\prime}}-\sum_{k, \alpha} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger} \delta_{k, k^{\prime}} \delta_{\alpha, \alpha^{\prime}}=-2 \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger} \tag{B.5b}
\end{align*}
$$

Thus, the Heisenberg equation of motion for $U_{k}(t)$ reads

$$
\begin{align*}
\frac{\mathrm{d}}{\mathrm{~d} t} U_{k}(t)= & \mathrm{i}\left\langle\left[\boldsymbol{H}, \sum_{\alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right]\right\rangle  \tag{B.6a}\\
= & \mathrm{i}\left\langle\left[-J \sum_{k^{\prime}, \alpha^{\prime}} \frac{\lambda}{4} \cos \left(k^{\prime}\right)\left(\boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger}+\boldsymbol{t}_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}\right), \sum_{\alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right]\right\rangle  \tag{B.6b}\\
= & -\mathrm{i} \sum_{\alpha}\left\langle\left[\sum_{k^{\prime}, \alpha^{\prime}} \frac{J^{\prime}}{4} \cos \left(k^{\prime}\right) \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right]\right\rangle \\
& -\mathrm{i} \sum_{\alpha}\left\langle\left[\sum_{k^{\prime}, \alpha^{\prime}} \frac{J^{\prime}}{4} \cos \left(k^{\prime}\right) \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}, \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right]\right\rangle  \tag{B.6c}\\
= & -\mathrm{i} \frac{1}{2} J^{\prime} \sum_{\alpha} \cos (k)\left\langle\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}-\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}\right\rangle  \tag{B.6d}\\
= & \frac{1}{2} J^{\prime} \cos (k) W_{k}(t) \tag{B.6e}
\end{align*}
$$

Likewise, for $V_{k}(t)$ we obtain

$$
\begin{align*}
\frac{\mathrm{d}}{\mathrm{~d} t} V_{k}(t)= & \mathrm{i}\left\langle\left[\boldsymbol{H}, \sum_{\alpha}\left(\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}+\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}\right)\right]\right\rangle  \tag{B.7a}\\
= & \mathrm{i} \sum_{\alpha}\left\langle\left[\sum_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}\left(J-\frac{J^{\prime}}{2} \cos \left(k^{\prime}\right)\right), \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}\right]\right\rangle \\
& +\mathrm{i} \sum_{\alpha}\left\langle\left[\sum_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}\left(J-\frac{J^{\prime}}{2} \cos \left(k^{\prime}\right)\right), \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}\right]\right\rangle \\
& +\mathrm{i} g\left\langle\frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\right\rangle \sum_{\alpha}\left\langle\left[\sum_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}, \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}\right]\right\rangle \\
& +\mathrm{i} g\left\langle\frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\right\rangle \sum_{\alpha}\left\langle\left[\sum_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}, \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}\right]\right\rangle  \tag{B.7b}\\
= & \left.\mathrm{i}\left(J-\frac{1}{2} J^{\prime} \cos (k)+g A(t)\right) 2\left\langle\sum_{\alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}-\sum_{\alpha} \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}\right\rangle\right\rangle  \tag{B.7c}\\
= & 2\left(J-\frac{1}{2} J^{\prime} \cos (k)+g A(t)\right) W_{k}(t) \tag{B.7d}
\end{align*}
$$

The quantity $W_{k}(t)$ requires some more thought. We need

$$
\begin{align*}
& {\left[\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}, \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger}\right]=\sum_{k, \alpha}\left[\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}, \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger}\right]}  \tag{B.8a}\\
& =\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}\left[\boldsymbol{t}_{-k, \alpha}, \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger}\right]+\sum_{k, \alpha}\left[\boldsymbol{t}_{k, \alpha}, \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger}\right] \boldsymbol{t}_{-k, \alpha}  \tag{B.8b}\\
& =\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger}\left[\boldsymbol{t}_{-k, \alpha}, \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger}\right]+\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}\left[\boldsymbol{t}_{-k, \alpha}, \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger}\right] \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger} \\
& +\sum_{k, \alpha} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger}\left[\boldsymbol{t}_{k, \alpha}, \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger}\right] \boldsymbol{t}_{-k, \alpha}+\sum_{k, \alpha}\left[\boldsymbol{t}_{k, \alpha}, \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger}\right] \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k, \alpha}  \tag{B.8c}\\
& =\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \delta_{k, k^{\prime}} \delta_{\alpha, \alpha^{\prime}}+\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger} \delta_{-k, k^{\prime}} \delta_{\alpha, \alpha^{\prime}} \\
& +\sum_{k, \alpha} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k, \alpha} \delta_{k,-k^{\prime}} \delta_{\alpha, \alpha^{\prime}}+\sum_{k, \alpha} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k, \alpha} \delta_{k, k^{\prime}} \delta_{\alpha, \alpha^{\prime}}  \tag{B.8d}\\
& =\boldsymbol{t}_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger}+\boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger}+\boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}+\boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}  \tag{B.8e}\\
& =2+2 \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}+2 \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}} \tag{B.8f}
\end{align*}
$$

and

$$
\begin{align*}
& {\left[\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}, \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}\right]=\left[\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}, \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger}\right]^{\dagger} }  \tag{B.9a}\\
= & 2+2 \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}+2 \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}} \tag{B.9b}
\end{align*}
$$

yielding

$$
\begin{equation*}
\frac{\mathrm{d}}{\mathrm{~d} t} W_{k}=\mathrm{i}\left\langle\left[\boldsymbol{H}, \mathrm{i} \sum_{\alpha}\left(\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}-\boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}\right)\right]\right\rangle \tag{B.10a}
\end{equation*}
$$

$$
\begin{align*}
= & -\sum_{\alpha}\left\langle\left[\boldsymbol{H}, \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}\right]\right\rangle+\sum_{\alpha}\left\langle\left[\boldsymbol{H}, \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}\right]\right\rangle  \tag{B.10b}\\
= & -\sum_{\alpha}\left\langle\left[\sum_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}\left(J-\frac{J^{\prime}}{2} \cos \left(k^{\prime}\right)\right), \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}\right]\right\rangle \\
& +\sum_{\alpha}\left\langle\left[\sum_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}\left(J-\frac{J^{\prime}}{2} \cos \left(k^{\prime}\right)\right), \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}\right]\right\rangle \\
& -g\left\langle\frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\right\rangle \sum_{\alpha}\left\langle\left[\sum_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime},}, \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}\right]\right\rangle \\
& +g\left\langle\frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\right\rangle \sum_{\alpha}\left\langle\left[\sum_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}, \boldsymbol{t}_{k, \alpha}} \boldsymbol{t}_{-k, \alpha}\right]\right\rangle \\
& \left.-\sum_{\alpha}\left\langle\left[\sum_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}} \frac{J^{\prime}}{4} \cos \left(k^{\prime}\right), \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}\right]\right\rangle\right\rangle \\
& \left.+\sum_{\alpha}\left\langle\left[\sum_{k^{\prime},,^{\prime}} \boldsymbol{t}_{k^{\prime}, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{-k^{\prime}, \alpha^{\prime}}^{\dagger} \frac{J^{\prime}}{4} \cos \left(k^{\prime}\right), \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{k, \alpha}\right]\right\rangle\right\rangle  \tag{B.10c}\\
= & -2\left\langle\left(\sum_{\alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}^{\dagger}+\sum_{\alpha} \boldsymbol{t}_{k, \alpha} \boldsymbol{t}_{-k, \alpha}\right)\left(J-\frac{1}{2} J^{\prime} \cos (k)+g A(t)\right)\right\rangle \\
& +\frac{1}{4} J^{\prime} \cos (k)\left\langle 4 \sum_{\alpha} \boldsymbol{t}_{k, \alpha^{\prime}}^{\dagger} \boldsymbol{t}_{k, \alpha}+4 \sum_{\alpha} \boldsymbol{t}_{-k, \alpha}^{\dagger} \boldsymbol{t}_{-k, \alpha}+4\right\rangle  \tag{B.10d}\\
= & -2\left(J-\frac{1}{2} J^{\prime} \cos (k)+g A(t)\right) V_{k}(t)+2 J^{\prime} \cos (k)\left(U_{k}(t)+\frac{1}{2}\right) . \tag{B.10e}
\end{align*}
$$

Here, we have interchanged $k \leftrightarrow-k$ and, thus, postulated that the system of ordinary differential equations is invariant under a change of sign in the momentum $k$.

As for the phonon sector characterised by $A(t), B(t)$ and $C(t)$ the Lindblad formalism introduced in section 2.4 needs to be employed to take the damping into account. In particular, we make use of equation (2.52). We begin with $A(t)$ by calculating

$$
\begin{align*}
& \left\langle\left[\boldsymbol{H}, \frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\right]\right\rangle=\left\langle\left[\omega_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}, \frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\right]\right\rangle  \tag{B.11a}\\
= & \frac{\omega_{0}}{\sqrt{N}}\left\langle\left[\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}, \boldsymbol{b}_{0}\right]+\left[\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}, \boldsymbol{b}_{0}^{\dagger}\right]\right\rangle=\frac{\omega_{0}}{\sqrt{N}}\left\langle-\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right\rangle=-\mathrm{i} \omega_{0} B(t) \tag{B.11b}
\end{align*}
$$

We obtain

$$
\begin{align*}
& \frac{\mathrm{d}}{\mathrm{~d} t} A(t) \\
= & \omega_{0} B(t) \\
& +\frac{\gamma}{\sqrt{N}}\left(N_{\mathrm{Q}}+1\right)\left\langle\boldsymbol{b}_{0}^{\dagger}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right) \boldsymbol{b}_{0}-\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)-\frac{1}{2}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right) \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle \\
& +\gamma \frac{N_{\mathrm{Q}}}{\sqrt{N}}\left\langle\boldsymbol{b}_{0}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right) \boldsymbol{b}_{0}^{\dagger}-\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)-\frac{1}{2}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right) \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}\right\rangle  \tag{B.12a}\\
= & \omega_{0} B(t)
\end{align*}
$$

$$
\begin{align*}
& +\frac{\gamma\left(N_{\mathrm{Q}}+1\right)}{\sqrt{N}}\left\langle\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}-\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}-\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle \\
& +\gamma \frac{N_{\mathrm{Q}}}{\sqrt{N}}\left\langle\boldsymbol{b}_{0} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}+\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger}-\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger}-\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}-\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}\right\rangle  \tag{B.12b}\\
= & \omega_{0} B(t) \\
& +\frac{\gamma}{\sqrt{N}}\left(N_{\mathrm{Q}}+1\right) \frac{1}{2}\left\langle\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle \\
& +\gamma \frac{N_{\mathrm{Q}}}{\sqrt{N}} \frac{1}{2}\left\langle\boldsymbol{b}_{0} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}+\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}\right\rangle  \tag{B.12c}\\
= & \omega_{0} B(t) \\
& +\frac{\gamma}{\sqrt{N}}\left(N_{\mathrm{Q}}+1\right) \frac{1}{2}\left\langle\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}-\boldsymbol{b}_{0}\right\rangle \\
& +\gamma \frac{N_{\mathrm{Q}}}{\sqrt{N}} \frac{1}{2}\left\langle\boldsymbol{b}_{0} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}+\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}+\boldsymbol{b}_{0}-\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger}+\boldsymbol{b}_{0}^{\dagger}\right\rangle  \tag{B.12d}\\
= & \omega_{0} B(t)-\frac{\gamma}{\sqrt{N}}\left(N_{\mathrm{Q}}+1\right) \frac{1}{2}\left\langle\boldsymbol{b}_{0}^{\dagger}+\boldsymbol{b}_{0}\right\rangle+\gamma \frac{N_{\mathrm{Q}}}{\sqrt{N}} \frac{1}{2}\left\langle\boldsymbol{b}_{0}^{\dagger}+\boldsymbol{b}_{0}\right\rangle  \tag{B.12e}\\
= & \omega_{0} B(t)-\frac{\gamma}{\sqrt{N}} \frac{1}{2}\left\langle\boldsymbol{b}_{0}^{\dagger}+\boldsymbol{b}_{0}\right\rangle  \tag{B.12f}\\
= & \omega_{0} B(t)-\frac{\gamma}{2} A(t) . \tag{B.12g}
\end{align*}
$$

To deduce the equation of motion for $B(t)$ we need

$$
\begin{align*}
& \left\langle\left[\boldsymbol{H}, \frac{\mathrm{i}}{\sqrt{N}}\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right)\right]\right\rangle \\
= & \left\langle\left[\omega_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}, \frac{\mathrm{i}}{\sqrt{N}}\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right)\right]\right\rangle+\left\langle\left[\sqrt{N} E(t)\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right), \frac{\mathrm{i}}{\sqrt{N}}\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right)\right]\right\rangle \\
& +\left\langle\left[g \frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\left\langle\left(\sum_{k, \alpha} \boldsymbol{t}_{k \alpha}^{\dagger} \boldsymbol{t}_{k \alpha}-\sum_{k, \alpha}\left\langle\boldsymbol{t}_{k \alpha}^{\dagger} \boldsymbol{t}_{k \alpha}\right\rangle_{\mathrm{eq}}\right)\right\rangle, \frac{\mathrm{i}}{\sqrt{N}}\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right)\right]\right\rangle  \tag{B.13a}\\
= & \frac{\mathrm{i} \omega_{0}}{\sqrt{N}}\left\langle\left[\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0},\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right)\right]\right\rangle+\frac{\mathrm{i} \sqrt{N} E(t)}{\sqrt{N}}\left\langle\left[\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right),\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right)\right]\right\rangle \\
& +\mathrm{i} g \frac{1}{N}\left\langle\left(\sum_{k, \alpha} \boldsymbol{t}_{k \alpha}^{\dagger} \boldsymbol{t}_{k \alpha}-\sum_{k, \alpha}\left\langle\boldsymbol{t}_{k \alpha}^{\dagger} \boldsymbol{t}_{k \alpha}\right\rangle_{\mathrm{eq}}\right)\right\rangle\left\langle\left[\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right),\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right)\right]\right\rangle  \tag{B.13b}\\
= & \frac{\mathrm{i} \omega_{0}}{\sqrt{N}}\left\langle\left[\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}, \boldsymbol{b}_{0}^{\dagger}\right]-\left[\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}, \boldsymbol{b}_{0}\right]\right\rangle+\mathrm{i} E(t)\left\langle\left[\boldsymbol{b}_{0}, \boldsymbol{b}_{0}^{\dagger}\right]-\left[\boldsymbol{b}_{0}^{\dagger}, \boldsymbol{b}_{0}\right]\right\rangle \\
& +\mathrm{i} g \frac{1}{N}\left(\sum_{k, \alpha}\left\langle\boldsymbol{t}_{k \alpha}^{\dagger} \boldsymbol{t}_{k \alpha}\right\rangle-\sum_{k, \alpha}\left\langle\boldsymbol{t}_{k \alpha}^{\dagger} \boldsymbol{t}_{k \alpha}\right\rangle_{\mathrm{eq}}\right)\left\langle\left[\boldsymbol{b}_{0}, \boldsymbol{b}_{0}^{\dagger}\right]-\left[\boldsymbol{b}_{0}^{\dagger}, \boldsymbol{b}_{0}\right]\right\rangle  \tag{B.13c}\\
= & \frac{\mathrm{i} \omega_{0}}{\sqrt{N}}\left\langle\boldsymbol{b}_{0}^{\dagger}\left[\boldsymbol{b}_{0}, \boldsymbol{b}_{0}^{\dagger}\right]-\left[\boldsymbol{b}_{0}^{\dagger}, \boldsymbol{b}_{0}\right] \boldsymbol{b}_{0}\right\rangle+\mathrm{i} E(t)\langle 1-(-1)\rangle
\end{align*}
$$

$$
\begin{align*}
& +\mathrm{i} g \frac{1}{N}\left(\sum_{k} U_{k}(t)-\sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\langle 1-(-1)\rangle  \tag{B.13d}\\
= & \frac{\mathrm{i} \omega_{0}}{\sqrt{N}}\left\langle\boldsymbol{b}_{0}^{\dagger}+\boldsymbol{b}_{0}\right\rangle+\mathrm{i} E(t)\langle 2\rangle+\mathrm{i} g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\langle 2\rangle  \tag{B.13e}\\
= & \mathrm{i} \omega_{0}\left\langle\frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}^{\dagger}+\boldsymbol{b}_{0}\right)\right\rangle+\mathrm{i} 2 E(t)+\mathrm{i} 2 g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)  \tag{B.13f}\\
= & \mathrm{i} \omega_{0} A(t)+\mathrm{i} 2 E(t)+\mathrm{i} 2 g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)  \tag{B.13g}\\
= & \mathrm{i}\left(\omega_{0} A(t)+2 E(t)+2 g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) . \tag{B.13h}
\end{align*}
$$

Thus,

$$
\begin{align*}
& \frac{\mathrm{d}}{\mathrm{~d} t} B(t) \\
= & -\omega_{0} A(t)-2\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) \\
& +\frac{\gamma \mathrm{i}}{\sqrt{N}}\left(N_{\mathrm{Q}}+1\right)\left\langle\boldsymbol{b}_{0}^{\dagger}\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right) \boldsymbol{b}_{0}-\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right)-\frac{1}{2}\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right) \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle \\
& +\gamma \frac{N_{\mathrm{Q}}^{\mathrm{i}}}{\sqrt{N}}\left\langle\boldsymbol{b}_{0}\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right) \boldsymbol{b}_{0}^{\dagger}-\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right)-\frac{1}{2}\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right) \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}\right\rangle  \tag{B.14a}\\
= & -\omega_{0} A(t)-2\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) \\
& +\frac{\gamma \mathrm{i}\left(N_{\mathrm{Q}}+1\right)}{\sqrt{N}}\left\langle\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}-\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}+\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}+\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle \\
& +\gamma \frac{N_{\mathrm{Q}}^{\mathrm{i}}}{\sqrt{N}}\left\langle\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger}-\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger}-\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}+\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}+\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}\right\rangle  \tag{B.14b}\\
= & -\omega_{0} A(t)-2\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) \\
& +\frac{\gamma \mathrm{i}}{\sqrt{N}}\left(N_{\mathrm{Q}}+1\right) \frac{1}{2}\left\langle\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}+\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle \\
& +\gamma \frac{N_{\mathrm{Q}}^{\mathrm{i}}}{\sqrt{N}} \frac{1}{2}\left\langle\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}+\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle  \tag{B.14c}\\
= & -\omega_{0} A(t)-2\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) \\
& +\frac{\gamma \mathrm{i}}{\sqrt{N}}\left(N_{\mathrm{Q}}+1\right) \frac{1}{2}\left\langle\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}+\boldsymbol{b}_{0}+\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle \\
& +\gamma \frac{N_{\mathrm{Q}}^{\mathrm{i}}}{\sqrt{N}} \frac{1}{2}\left\langle\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger}+\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\boldsymbol{b}_{0}+\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle \tag{B.14d}
\end{align*}
$$

$$
\begin{align*}
= & -\omega_{0} A(t)-2\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) \\
& -\frac{\gamma \mathrm{i}}{\sqrt{N}}\left(N_{\mathrm{Q}}+1\right) \frac{1}{2}\left\langle\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right\rangle+\gamma \frac{N_{\mathrm{Q}} \mathrm{i}}{\sqrt{N}} \frac{1}{2}\left\langle\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right\rangle  \tag{B.14e}\\
= & -\omega_{0} A(t)-2\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right)-\frac{\gamma \mathrm{i}}{\sqrt{N}} \frac{1}{2}\left\langle\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right\rangle  \tag{B.14f}\\
= & -\omega_{0} A(t)-2\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right)-\frac{\gamma}{2} B(t) . \tag{B.14g}
\end{align*}
$$

Finally, for $C(t)$ we calculate

$$
\begin{align*}
& \left\langle\left[\boldsymbol{H}, \frac{1}{N} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right]\right\rangle \\
= & \left\langle\left[E(t) \sqrt{N}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right), \frac{1}{N} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right]\right\rangle \\
& +\left\langle\left[g \frac{1}{\sqrt{N}}\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right)\left\langle\left(\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}-\sum_{k, \alpha}\left\langle\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right\rangle_{\mathrm{eq}}\right)\right\rangle, \frac{1}{N} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right]\right\rangle  \tag{B.15a}\\
= & E(t) \frac{\sqrt{N}}{N}\left\langle\left[\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right), \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right]\right\rangle \\
& +g \frac{1}{\sqrt{N} N}\left\langle\left(\sum_{k, \alpha} \boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}-\sum_{k, \alpha}\left\langle\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right\rangle_{\mathrm{eq}}\right)\right\rangle\left\langle\left[\left(\boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\right), \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right]\right\rangle  \tag{B.15b}\\
= & E(t) \frac{\sqrt{N}}{N}\left\langle\left[\boldsymbol{b}_{0}, \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right]+\left[\boldsymbol{b}_{0}^{\dagger}, \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right]\right\rangle \\
& +g \frac{1}{\sqrt{N} N}\left(\sum_{k, \alpha}\left\langle\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right\rangle-\sum_{k, \alpha}\left\langle\boldsymbol{t}_{k, \alpha}^{\dagger} \boldsymbol{t}_{k, \alpha}\right\rangle_{\mathrm{eq}}\right)\left\langle\left[\boldsymbol{b}_{0}, \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right]+\left[\boldsymbol{b}_{0}^{\dagger}, \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right]\right\rangle \\
= & E(t) \frac{\sqrt{N}}{N}\left\langle\left[\boldsymbol{b}_{0}, \boldsymbol{b}_{0}^{\dagger}\right] \boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\left[\boldsymbol{b}_{0}^{\dagger}, \boldsymbol{b}_{0}\right]\right\rangle \\
& +g \frac{1}{\sqrt{N} N}\left(\sum_{k} U_{k}(t)-\sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\left\langle\left[\boldsymbol{b}_{0}, \boldsymbol{b}_{0}^{\dagger}\right] \boldsymbol{b}_{0}+\boldsymbol{b}_{0}^{\dagger}\left[\boldsymbol{b}_{0}^{\dagger}, \boldsymbol{b}_{0}\right]\right\rangle  \tag{B.15c}\\
= & E(t) \frac{1}{\sqrt{N}}\left\langle\boldsymbol{b}_{0}-\boldsymbol{b}_{0}^{\dagger}\right\rangle+g \frac{1}{\sqrt{N}}\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\left\langle\boldsymbol{b}_{0}-\boldsymbol{b}_{0}^{\dagger}\right\rangle  \tag{B.15d}\\
= & \mathrm{i}\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right)\left\langle\frac{\mathrm{i}}{\sqrt{N}}\left(\boldsymbol{b}_{0}^{\dagger}-\boldsymbol{b}_{0}\right)\right\rangle  \tag{B.15e}\\
= & \mathrm{i}\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) B(t) \tag{B.15f}
\end{align*}
$$

which yields

$$
\frac{\mathrm{d}}{\mathrm{~d} t} C(t)
$$

$$
\begin{align*}
= & -\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) B(t) \\
& +\frac{\gamma}{N}\left(N_{\mathrm{Q}}+1\right)\left\langle\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}-\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle \\
& +\gamma \frac{N_{\mathrm{Q}}}{N}\left\langle\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}-\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\frac{1}{2} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}\right\rangle  \tag{B.16a}\\
= & -\left(E(t)+g\left(u(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) B(t) \\
& +\frac{\gamma}{N}\left(N_{\mathrm{Q}}+1\right)\left\langle\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}-\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle \\
& +\gamma \frac{N_{\mathrm{Q}}}{N}\left\langle\boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}-\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}+\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}-\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}+\frac{1}{2} \boldsymbol{b}_{0} \boldsymbol{b}_{0}^{\dagger}\right\rangle  \tag{B.16b}\\
= & -\left(E(t)+g\left(u(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) B(t)-\frac{\gamma}{N}\left(N_{\mathrm{Q}}+1\right)\left\langle\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle \\
& +\gamma \frac{N_{\mathrm{Q}}}{N}\left\langle\boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}+1\right\rangle  \tag{B.16c}\\
= & -\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) B(t)-\gamma\left\langle\frac{1}{N} \boldsymbol{b}_{0}^{\dagger} \boldsymbol{b}_{0}\right\rangle+\gamma \frac{N_{\mathrm{Q}}}{N}  \tag{B.16d}\\
= & -\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) B(t)-\gamma C(t)+\gamma \frac{N_{\mathrm{Q}}}{N}  \tag{B.16e}\\
= & -\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) B(t)-\gamma\left(C(t)-\frac{N_{\mathrm{Q}}}{N}\right) . \tag{B.16f}
\end{align*}
$$

Taking the restriction to zero temperature in equation $(\overline{3.21})$ into account yields, c.f. equation (2.50),

$$
\begin{equation*}
N_{\mathrm{Q}}=0 \tag{B.17}
\end{equation*}
$$

and, thus, the equation of motion reads

$$
\begin{equation*}
\frac{\mathrm{d}}{\mathrm{~d} t} C(t)=-\left(E(t)+g\left(\mathcal{U}(t)-\frac{1}{N} \sum_{k}\left\langle U_{k}\right\rangle_{\mathrm{eq}}\right)\right) B(t)-\gamma C(t) . \tag{B.18}
\end{equation*}
$$

We have presented the general calculation to show that the effect of the restriction to zero temperature $T$ only plays a role in this equation.

## C Triplon Dispersion Relation

We present the calculation to obtain the triplon dispersion relation $\omega_{\mathrm{T}}$.
We investigate the spin system Hamiltonian $\boldsymbol{H}_{\mathrm{S}}$ (3.10). Omitting the triplon flavour index $\alpha$ in the Bogoliubov transform (3.29) we obtain

$$
\begin{align*}
\boldsymbol{t}_{k}^{\dagger} \boldsymbol{t}_{k}= & \left(\hat{\boldsymbol{t}}_{k}^{\dagger} \cosh (\theta)+\hat{\boldsymbol{t}}_{-k} \sinh (\theta)\right) \cdot\left(\hat{\boldsymbol{t}}_{k} \cosh (\theta)+\hat{\boldsymbol{t}}_{-k}^{\dagger} \sinh (\theta)\right) \\
= & \hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta) \\
& +\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{-k}^{\dagger} \sinh ^{2}(\theta)  \tag{C.1a}\\
\boldsymbol{t}_{k}^{\dagger} \boldsymbol{t}_{-k}^{\dagger}= & \left(\hat{\boldsymbol{t}}_{k}^{\dagger} \cosh (\theta)+\hat{\boldsymbol{t}}_{-k} \sinh (\theta)\right) \cdot\left(\hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh (\theta)+\hat{\boldsymbol{t}}_{k} \sinh (\theta)\right) \\
= & \hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh (\theta) \sinh (\theta) \\
& +\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{k} \sinh ^{2}(\theta)  \tag{C.1b}\\
\boldsymbol{t}_{k} \boldsymbol{t}_{-k}= & \left(\hat{\boldsymbol{t}}_{k} \cosh (\theta)+\hat{\boldsymbol{t}}_{-k}^{\dagger} \sinh (\theta)\right) \cdot\left(\hat{\boldsymbol{t}}_{-k} \cosh (\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \sinh (\theta)\right) \\
= & \hat{\boldsymbol{t}}_{k} \hat{\boldsymbol{t}}_{-k} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k} \hat{\boldsymbol{t}}_{k}^{\dagger} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{-k}^{\dagger} \hat{t}_{-k} \cosh (\theta) \sinh (\theta) \\
& +\hat{\boldsymbol{t}}_{-k}^{\dagger} \hat{\boldsymbol{t}}_{k}^{\dagger} \sinh ^{2}(\theta) . \tag{C.1c}
\end{align*}
$$

Substitution in the Hamiltonian $\boldsymbol{H}_{\mathrm{S}}(3.10)$ then yields

$$
\begin{align*}
\boldsymbol{H}_{\mathrm{S}}=\sum_{k} & \left(J-\frac{1}{2} J^{\prime} \cos (k)\right)\left(\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh (\theta) \sinh (\theta)\right. \\
& \left.+\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{-k}^{\dagger} \sinh ^{2}(\theta)\right) \\
- & \frac{1}{4} J^{\prime} \cos (k)\left(\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh (\theta) \sinh (\theta)\right. \\
& \left.+\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{\boldsymbol{t}} \sinh ^{2}(\theta)\right) \\
- & \frac{1}{4} J^{\prime} \cos (k)\left(\hat{\boldsymbol{t}}_{k} \hat{\boldsymbol{t}}_{-k} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k} \hat{\boldsymbol{t}}_{k}^{\dagger} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{-k}^{\dagger} \hat{\boldsymbol{t}}_{-k} \cosh (\theta) \sinh (\theta)\right. \\
& \left.+\hat{\boldsymbol{t}}_{-k}^{\dagger} \hat{\boldsymbol{t}}_{\boldsymbol{t}}^{\dagger} \sinh ^{2}(\theta)\right)  \tag{C.2a}\\
=\sum_{k} & \left(J-\frac{1}{2} J^{\prime} \cos (k)\right)\left(\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh (\theta) \sinh (\theta)\right. \\
& \left.+\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{-k}^{\dagger} \hat{\boldsymbol{t}}_{-k} \sinh { }^{2}(\theta)\right) \\
- & \frac{1}{4} J^{\prime} \cos (k)\left(\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{-k}^{\dagger} \hat{\boldsymbol{t}}_{-k} \cosh (\theta) \sinh (\theta)\right. \\
& \left.+\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{k} \sinh { }^{2}(\theta)\right) \\
- & \frac{1}{4} J^{\prime} \cos (k)\left(\hat{\boldsymbol{t}}_{\boldsymbol{k}} \hat{\boldsymbol{t}}_{-k} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{-k}^{\dagger} \hat{\boldsymbol{t}}_{-k} \cosh (\theta) \sinh (\theta)\right.
\end{align*}
$$

$$
\begin{align*}
&\left.+\hat{\boldsymbol{t}}_{-k}^{\dagger} \hat{\boldsymbol{t}}_{k}^{\dagger} \sinh ^{2}(\theta)\right) \\
&+ {\left[\hat{\boldsymbol{t}}_{-k}, \hat{\boldsymbol{t}}_{-k}^{\dagger}\right]\left(J-\frac{1}{2} J^{\prime} \cos (k)\right) \sinh ^{2}(\theta) } \\
&- {\left[\hat{\boldsymbol{t}}_{-k}, \hat{\boldsymbol{t}}_{-k}^{\dagger}\right] \frac{1}{4} J^{\prime} \cos (k) \cosh (\theta) \sinh (\theta)-\left[\hat{\boldsymbol{t}}_{k}, \hat{\boldsymbol{t}}_{k}^{\dagger}\right] \frac{1}{4} J^{\prime} \cos (k) \cosh (\theta) \sinh (\theta) }  \tag{C.2b}\\
&=\sum_{k}\left(J-\frac{1}{2} J^{\prime} \cos (k)\right)\left(\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh (\theta) \sinh (\theta)\right. \\
&\left.+\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{-k}^{\dagger} \hat{\boldsymbol{t}}_{-k} \sinh ^{2}(\theta)\right) \\
&-\frac{1}{4} J^{\prime} \cos (k)\left(\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{-k}^{\dagger} \hat{\boldsymbol{t}}_{-k} \cosh (\theta) \sinh (\theta)\right. \\
&\left.+\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{k} \sinh ^{2}(\theta)\right) \\
&-\frac{1}{4} J^{\prime} \cos (k)\left(\hat{\boldsymbol{t}}_{k} \hat{\boldsymbol{t}}_{-k} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{-k}^{\dagger} \hat{\boldsymbol{t}}_{-k} \cosh (\theta) \sinh (\theta)\right. \\
&\left.+\hat{\boldsymbol{t}}_{-k}^{\dagger} \hat{\boldsymbol{t}}_{k}^{\dagger} \sinh ^{2}(\theta)\right)+\operatorname{const} . \tag{C.2c}
\end{align*}
$$

We exchange $k$ by $-k$ where necessary, which leaves the expression invariant due to

$$
\begin{equation*}
\cos (k)=\cos (-k) \tag{C.3}
\end{equation*}
$$

to obtain

$$
\begin{align*}
\boldsymbol{H}_{\mathrm{S}}=\sum_{k} & \left(J-\frac{1}{2} J^{\prime} \cos (k)\right)\left(\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh (\theta) \sinh (\theta)\right. \\
& \left.+\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \sinh ^{2}(\theta)\right) \\
- & \frac{1}{4} J^{\prime} \cos (k)\left(\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{-k}^{\dagger} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)\right. \\
& \left.+\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{k} \sinh ^{2}(\theta)\right) \\
- & \frac{1}{4} J^{\prime} \cos (k)\left(\hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{k} \cosh ^{2}(\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} \cosh (\theta) \sinh (\theta)\right. \\
& \left.+\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{-k}^{\dagger} \sinh ^{2}(\theta)\right)+ \text { const. } \tag{C.4}
\end{align*}
$$

Sorting by operators results in the expression

$$
\begin{align*}
\mathbf{H}_{\mathrm{S}}=\sum_{k} & \hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k}\left(J \cosh ^{2}(\theta)+J \sinh ^{2}(\theta)-\frac{1}{2} J^{\prime} \cos (k) \cosh ^{2}(\theta)-\frac{1}{2} J^{\prime} \cos (k) \sinh ^{2}(\theta)\right. \\
& \left.-\frac{1}{2} J^{\prime} \cos (k) 2 \cosh (\theta) \sinh (\theta)\right) \\
& +\hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{-k}^{\dagger}\left(J \cosh (\theta) \sinh (\theta)-\frac{1}{2} J^{\prime} \cos (k) \cosh (\theta) \sinh (\theta)-\frac{1}{4} J^{\prime} \cos (k) \cosh ^{2}(\theta)\right. \\
& \left.-\frac{1}{4} J^{\prime} \cos (k) \sinh ^{2}(\theta)\right) \\
+ & \hat{\boldsymbol{t}}_{-k} \hat{\boldsymbol{t}}_{k}\left(J \cosh (\theta) \sinh (\theta)-\frac{1}{2} J^{\prime} \cos (k) \cosh (\theta) \sinh (\theta)-\frac{1}{4} J^{\prime} \cos (k) \cosh ^{2}(\theta)\right. \\
& \left.-\frac{1}{4} J^{\prime} \cos (k) \sinh ^{2}(\theta)\right)+ \text { const } \tag{C.5a}
\end{align*}
$$

for which

$$
\left.\begin{array}{rl} 
& \left(J \cosh (\theta) \sinh (\theta)-\frac{1}{2} J^{\prime} \cos (k) \cosh (\theta) \sinh (\theta)-\frac{1}{4} J^{\prime} \cos (k) \cosh ^{2}(\theta)\right. \\
& \left.-\frac{1}{4} J^{\prime} \cos (k) \sinh ^{2}(\theta)\right) \\
\Leftrightarrow & \frac{1}{2} J^{\prime} \cos (k) \cosh (\theta) \sinh (\theta)=J \cosh (\theta) \sinh (\theta)-\frac{1}{4} J^{\prime} \cos (k) \cosh ^{2}(\theta) \\
& -\frac{1}{4} J^{\prime} \cos (k) \sinh ^{2}(\theta)
\end{array}\right] \begin{aligned}
\Leftrightarrow & \frac{1}{2} J^{\prime} \cos (k) 2 \cosh (\theta) \sinh (\theta)=2 J \cosh (\theta) \sinh (\theta)-\frac{1}{2} J^{\prime} \cos (k) \cosh ^{2}(\theta) \\
\Leftrightarrow & \cosh (\theta) \sinh (\theta)=\frac{1}{4} \frac{1}{2} J^{\prime} \cos (k) \sinh { }^{2}(\theta) \\
\Leftrightarrow & \left(\frac{1}{2} \mathrm{e}^{\theta}+\frac{1}{2} \mathrm{e}^{-\theta}\right) \cdot\left(\frac{1}{2} \mathrm{e}^{\theta}-\frac{1}{2} \mathrm{e}^{-\theta}\right)=\frac{1}{4} \frac{J^{\prime}}{J} \cos (k)\left(\frac{\cosh (\theta)+\sinh (\theta))^{2}}{2} \mathrm{e}^{\theta}+\frac{1}{2} \mathrm{e}^{-\theta}+\frac{1}{2} \mathrm{e}^{\theta}-\frac{1}{2} \mathrm{e}^{-\theta}\right)^{2} \\
\Leftrightarrow & \frac{1}{4} \mathrm{e}^{2 \theta}-\frac{1}{4} \mathrm{e}^{-2 \theta}=\frac{1}{4} \frac{J^{\prime}}{J} \cos (k) \mathrm{e}^{2 \theta} \\
\Leftrightarrow & 1-\mathrm{e}^{-4 \theta}=\frac{J^{\prime}}{J} \cos (k) \\
\Leftrightarrow & \mathrm{e}^{-4 \theta}=1-\frac{J^{\prime}}{J} \cos (k) \\
\Rightarrow & \mathrm{e}^{-2 \theta}=\sqrt{1-\lambda \cos (k)}
\end{aligned}
$$

has to hold true in order to be diagonal. We substitute equation (C.6a) and (C.6c) into equation (C.5a) receiving

$$
\begin{align*}
\boldsymbol{H}_{\mathrm{S}} & =\sum_{k} \hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} J(\cosh (\theta)-\sinh (\theta))^{2}+\text { const } \\
& =\sum_{k} \hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} J\left(\frac{1}{2} \mathrm{e}^{\theta}+\frac{1}{2} \mathrm{e}^{-\theta}-\frac{1}{2} \mathrm{e}^{\theta}+\frac{1}{2} \mathrm{e}^{-\theta}\right)^{2}+\text { const } \\
& =\sum_{k} \hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} J \mathrm{e}^{-2 \theta}+\text { const } \tag{C.7}
\end{align*}
$$

which reads making use of equation (C.6i)

$$
\begin{equation*}
\boldsymbol{H}_{\mathrm{S}}=\sum_{k} \hat{\boldsymbol{t}}_{k}^{\dagger} \hat{\boldsymbol{t}}_{k} J \sqrt{1-\lambda \cos (k)}+\text { const } \tag{C.8}
\end{equation*}
$$

Thus, the triplon dispersion relation is determined by

$$
\begin{equation*}
\omega_{\mathrm{T}}(k)=J \sqrt{1-\lambda \cos (k)} \tag{C.9}
\end{equation*}
$$

Now, we are enabled to set up equations characterising the two triplon band which according to equation (4.11) reads

$$
\begin{equation*}
\omega_{2}(p, k)=\sqrt{1-\lambda \cos (k+p)}+\sqrt{1-\lambda \cos (k)} . \tag{C.10}
\end{equation*}
$$

For an extreme value in $k$

$$
\begin{align*}
& 0=\frac{\mathrm{d}}{\mathrm{~d} k} \omega_{2}(p, k)= \frac{1}{2} \frac{J^{\prime}}{J} \sin (k+p)\left(1-\frac{J^{\prime}}{J} \cos (k+p)\right)^{-\frac{1}{2}} \\
&+\frac{1}{2} \frac{J^{\prime}}{J} \sin (k)\left(1-\frac{J^{\prime}}{J} \cos (k)\right)^{-\frac{1}{2}}  \tag{C.11a}\\
& \Leftrightarrow \sin (k+p)\left(1-\frac{J^{\prime}}{J} \cos (k+p)\right)^{-\frac{1}{2}}=-\sin (k)\left(1-\frac{J^{\prime}}{J} \cos (k)\right)^{-\frac{1}{2}} \tag{C.11b}
\end{align*}
$$

has to hold true. This equation is solved by

$$
\begin{equation*}
k=-\frac{p}{2} \Leftrightarrow p=-2 k . \tag{C.12}
\end{equation*}
$$

For the momenta $k$ and $p$ do only appear in the argument of a sine or a cosine the dispersion relations $\omega_{1}$ and $\omega_{2}$ are periodic in $k$ or antiperiodic in $k$ with period $2 \pi$. Hence,

$$
\begin{equation*}
k=-\frac{p}{2}+\pi \Leftrightarrow p=2(\pi-k) \tag{C.13}
\end{equation*}
$$

also solves equation (C.11b). Making again use of the cosine's symmetry (C.3) as well as the antiperiodicity of sine and cosine

$$
\begin{align*}
& \sin (x)=-\sin (x+\pi)  \tag{C.14a}\\
& \cos (x)=-\cos (x+\pi) \tag{C.14b}
\end{align*}
$$

we obtain

$$
\begin{align*}
\min _{k}\left\{\omega_{2}(k, p)\right\} & =\omega_{\mathrm{T}}\left(\frac{p}{2}\right)+\omega_{\mathrm{T}}\left(-\frac{p}{2}\right)=2 \omega_{\mathrm{T}}\left(\frac{p}{2}\right)=2 \sqrt{1-\lambda \cos \left(\frac{p}{2}\right)}  \tag{C.15a}\\
\max _{k}\left\{\omega_{2}(k, p)\right\} & =\omega_{\mathrm{T}}\left(\pi+\frac{p}{2}\right)+\omega_{\mathrm{T}}\left(\pi-\frac{p}{2}\right) \\
& =\sqrt{1-\lambda \cos \left(\pi+\frac{p}{2}\right)}+\sqrt{1-\lambda \cos \left(\pi-\frac{p}{2}\right)} \\
& =\sqrt{1+\lambda \cos \left(\frac{p}{2}\right)}+\sqrt{1+\lambda \cos \left(-\frac{p}{2}\right)}=2 \sqrt{1+\lambda \cos \left(\frac{p}{2}\right)} \tag{C.15b}
\end{align*}
$$

which yields

$$
\begin{align*}
\omega_{\min } & =2 \sqrt{1-\lambda}  \tag{C.16a}\\
\omega_{\max } & =2 \sqrt{1+\lambda} \tag{C.16b}
\end{align*}
$$

The band's mean $\omega_{\mathrm{m}}$ is then determined by

$$
\begin{equation*}
\omega_{\mathrm{m}}=\frac{\omega_{\max }+\omega_{\min }}{2}=\sqrt{1+\lambda}+\sqrt{1-\lambda} . \tag{C.17}
\end{equation*}
$$

With $\lambda=0.5$ we obtain

$$
\begin{align*}
\omega_{\min } & =\sqrt{2} \approx 1.414213562 \approx 1.5  \tag{C.18a}\\
\omega_{\max } & =\sqrt{6} \approx 2.449489743 \approx 2.5  \tag{C.18b}\\
\omega_{\mathrm{m}} & \approx 1.931851653 \approx 2 \tag{C.18c}
\end{align*}
$$

## D Above Band Quasi Steady States

We illustrate why a discussion of above band quasi steady states is not helpful as stated in section 4.2. Figure D.1 equals figure 4.2 where we have plotted a desired time evolution. Here, however, in figure D.1 (d) we find that the triplon density $\mathcal{U}(t)$ does not establish an oscillation around a value that differs significantly from its initial value, i.e. there is no non-linear response which we were aiming at. As a result we turn to beneath band cases.


Figure D.1: Example of an above band time evolution; values: $g=0.03 \mathrm{~J}, \omega_{0}=3.0 \mathrm{~J}$, $a=0.4 J, \gamma=0.1 J$

## E Above Band Green's Functions

In section 4.3 we have only presented results for beneath band Green's functions. Here we show plots for an above band case. All our findings discussed in section 4.3 are still valid here. We hereby illustrate that our findings are generic. In particular the equations describing the peak positions, $(4.44),(4.45),(4.61),(4.62)$ and $(4.63)$, hold. We shall just give the plots and corresponding fit parameters. For a detailed discussion of the phenomena c.f. section 4.3 .

## E. 1 Special Case $k=\pi / 2$

We obtain the following parameters according to equation (4.32) for the fits depicted in figure E.1(c),

$$
\begin{align*}
\xi & =0.000157  \tag{E.1a}\\
\chi_{0} & =0.999686 J  \tag{E.1b}\\
\zeta & =0.000601 J \tag{E.1c}
\end{align*}
$$

for the reative Green's function $G_{\pi / 2}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right)$,

$$
\begin{align*}
\xi & =0.000156  \tag{E.2a}\\
\chi_{0} & =0.999686 \mathrm{~J}  \tag{E.2b}\\
\zeta & =0.000600 \mathrm{~J} \tag{E.2c}
\end{align*}
$$

for the time average Green's function $\tilde{G}_{\pi / 2}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and

$$
\begin{align*}
\xi & =0.000157  \tag{E.3a}\\
\chi_{0} & =0.999686 \mathrm{~J}  \tag{E.3b}\\
\zeta & =0.000600 \mathrm{~J} \tag{E.3c}
\end{align*}
$$

for the average Green's function $\bar{G}_{\pi / 2}\left(10000 J^{-1}, \omega\right)$.
The fit parameters for the imaginary parts of $G_{\pi / 2}^{\mathrm{rel}}\left(3000 J^{-1}, \omega_{\text {diff }}\right)$ and $G_{\pi / 2}^{\mathrm{rel}}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ given in figure E.3(b) are determined by

$$
\begin{align*}
\xi & =0.000224  \tag{E.4a}\\
\chi_{0} & =0.999606 J  \tag{E.4b}\\
\zeta & =0.000857 J \tag{E.4c}
\end{align*}
$$



Figure E.1: Fourier spectrum for momentum $k=\pi / 2$ of the relative, time average and average Green's function, $G_{\pi / 2}^{\text {rel }}\left(0, \omega_{\text {diff }}\right), \tilde{G}_{\pi / 2}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and $\bar{G}_{\pi / 2}\left(10000 J^{-1}, \omega\right)$; values: $g=0.03 J, \omega_{0}=3 J, a=0.4 J, \gamma=0.1 J$


Figure E.2: Real and imaginary part of the Fourier spectra for momentum $k=\pi / 2$ of the relative, time average and average Green's function, $G_{\pi / 2}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right), \tilde{G}_{\pi / 2}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and $\bar{G}_{\pi / 2}\left(10000 J^{-1}, \omega\right)$; values: $g=0.03 J, \omega_{0}=3 J, a=0.4 J, \gamma=0.1 J$
and

$$
\begin{align*}
\xi & =0.000314  \tag{E.5a}\\
\chi_{0} & =0.999415 \mathrm{~J}  \tag{E.5b}\\
\zeta & =0.001201 \mathrm{~J} \tag{E.5c}
\end{align*}
$$

respectively while we obtain

$$
\begin{align*}
\xi & =0.000262  \tag{E.6a}\\
\chi_{0} & =0.999476 \mathrm{~J}  \tag{E.6b}\\
\zeta & =0.001002 \mathrm{~J} \tag{E.6c}
\end{align*}
$$

for $\tilde{G}_{\pi / 2}\left(3000 J^{-1}, \omega_{\text {diff }}\right)$ and

$$
\begin{align*}
\xi & =0.000787  \tag{E.7a}\\
\chi_{0} & =0.998428 J  \tag{E.7b}\\
\zeta & =0.003010 J \tag{E.7c}
\end{align*}
$$

for $\tilde{G}_{\pi / 2}\left(1000 J^{-1}, \omega_{\text {diff }}\right)$ which can be found in figure E.3(d).


Figure E.3: Investigation of the dependence of relative Green's function $G_{\pi / 2}^{\mathrm{rel}}\left(t_{\mathrm{in}}, \omega_{\text {diff }}\right)$ on the initial time $t_{\text {in }}$ and of the time average Green's function $\tilde{G}_{\pi / 2}\left(\bar{t}, \omega_{\text {diff }}\right)$ on the average time $\bar{t}$ for momentum $k=\pi / 2$; values: $g=0.03 J, \omega_{0}=3.0 J, a=0.4 J, \gamma=0.1 J$

## E. 2 Case $k \neq \pi / 2$

Here fits according to equation (4.32) shown in figure E.4(c) yield

$$
\begin{align*}
\xi & =0.000159  \tag{E.8a}\\
\chi_{0} & =0.804046 \mathrm{~J}  \tag{E.8b}\\
\zeta & =0.000600 \mathrm{~J} \tag{E.8c}
\end{align*}
$$

for the reative Green's function $G_{\pi / 4}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right)$,

$$
\begin{align*}
\xi & =0.000158  \tag{E.9a}\\
\chi_{0} & =0.804052 J  \tag{E.9b}\\
\zeta & =0.000600 J \tag{E.9c}
\end{align*}
$$

for the time average Green's function $\tilde{G}_{\pi / 4}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and

$$
\begin{align*}
\xi & =0.000158  \tag{E.10a}\\
\chi_{0} & =0.804052 J  \tag{E.10b}\\
\zeta & =0.000600 J \tag{E.10c}
\end{align*}
$$

for the average Green's function $\bar{G}_{\pi / 4}\left(10000 J^{-1}, \omega\right)$.
Fits applied to the imaginary parts of $G_{\pi / 4}^{\mathrm{rel}}\left(3000 J^{-1}, \omega_{\text {diff }}\right)$ and $G_{\pi / 4}^{\mathrm{rel}}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ to be found in figure E.3(b) are given by

$$
\begin{align*}
\xi & =0.000227  \tag{E.11a}\\
\chi_{0} & =0.803972 J  \tag{E.11b}\\
\zeta & =0.000857 J \tag{E.11c}
\end{align*}
$$

and

$$
\begin{align*}
\xi & =0.000318  \tag{E.12a}\\
\chi_{0} & =0.803784 J  \tag{E.12b}\\
\zeta & =0.001200 J \tag{E.12c}
\end{align*}
$$

respectively. For $\tilde{G}_{\pi / 4}\left(3000 J^{-1}, \omega_{\text {diff }}\right)$ we obtain

$$
\begin{align*}
\xi & =0.000265  \tag{E.13a}\\
\chi_{0} & =0.803843 J  \tag{E.13b}\\
\zeta & =0.001001 J \tag{E.13c}
\end{align*}
$$

and for $\tilde{G}_{\pi / 4}\left(1000 J^{-1}, \omega_{\text {diff }}\right)$

$$
\begin{align*}
\xi & =0.000797  \tag{E.14a}\\
\chi_{0} & =0.802797 J  \tag{E.14b}\\
\zeta & =0.003010 J \tag{E.14c}
\end{align*}
$$

The corresponding plots can be found in figure E.3(d).


Figure E.4: Fourier spectrum for momentum $k=\pi / 4$ of the relative, time average and average Green's function, $G_{\pi / 4}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right), \tilde{G}_{\pi / 4}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and $\bar{G}_{\pi / 4}\left(10000 J^{-1}, \omega\right)$; values: $g=0.03 J, \omega_{0}=3 J, a=0.4 J, \gamma=0.1 J$


Figure E.5: Real and imaginary part of the Fourier spectra for momentum $k=\pi / 4$ of the relative, time average and average Green's function, $G_{\pi / 4}^{\mathrm{rel}}\left(0, \omega_{\text {diff }}\right), \tilde{G}_{\pi / 4}\left(5000 J^{-1}, \omega_{\text {diff }}\right)$ and $\bar{G}_{\pi / 4}\left(10000 J^{-1}, \omega\right)$; values: $g=3 J, \omega_{0}=1.3 J, a=0.4 J, \gamma=0.1 J$


Figure E.6: Investigation of the dependence of relative Green's function $G_{\pi / 4}^{\mathrm{rel}}\left(t_{\mathrm{in}}, \omega_{\text {diff }}\right)$ on the initial time $t_{\text {in }}$ and of the time average Green's function $\tilde{G}_{\pi / 4}\left(\bar{t}, \omega_{\text {diff }}\right)$ on the average time $\bar{t}$ for momentum $k=\pi / 4$; values: $g=0.03 J, \omega_{0}=3 J, a=0.4 J, \gamma=0.1 J$

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[^0]:    ${ }^{1}$ private communication with Christian Rüegg, Paul Scherrer Institut
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