The s-d exchange model
studied using DMFT and Dual Fermion methods

April 2012

Master thesis by:
Jonas Sweep
0513709

supervisors:
d. ф.-м. н. А.Н. Рубцов
prof. dr. M.I. Katsnelson

Department Theory of Condensed Matter
Institute for Molecules and Materials
Radboud University of Nijmegen
jsweep@science.ru.nl
Preface

Almost seven years ago I started to study physics because I wanted to understand the world around me. I chose the subject of condensed matter because I wanted to work on concrete materials. After my thesis work I realised that to meet the challenges the future poses to us, our world needs more than physics from me. My place in it is elsewhere, namely in the field of sustainable development. Where exactly, I am still figuring out.

I would like to express my sincere thanks to everyone who has made this master's thesis a success. Especially the following people:

Itsk e van Deursen for all her patience and understanding. It was not easy for her to miss me for half a year while I was working in Moscow. I am still glad I made the decision to go abroad but also for me there were some hard moments. All in all, I have seen and learned a lot, also outside of science. Therefore, I am very grateful and I feel very privileged to have worked in the most beautiful city of the world.

I thank Andrey Antipov and Igor Krivenko for all the useful discussions. Hartmut Hafermann for his kind permission to use the Dual Fermion code. Alexey Osipov I thank for guiding me through the wonderful world of Russian bureaucracy and for being a great friend abroad. Evgeni Shirshin and Stanislav Evlashin I thank for all the less scientific discussions and quality time. I also want to thank Inka Loch and Leendertjan Karssemeijer for reading the manuscript and helping me clarify many vaguenesses.

Natalia Nikolaevna from the international office of Moscow State University and the people from Nuffic and Neso Russia for making my internship possible. I want to thank Alexander Lichtenstein for his hospitality, providing me with the opportunity to join discussion in Hamburg.

Anthony Zee and Gerald Mahan I thank for their understandable writings in their excellent textbooks. They were the only physics writers that did not make me feel like an idiot when I was doing my literature research.

Last but definitely not least, I want to thank Alexey Rubtsov and Mikhail Katsnelson for continuous support, discussions, guidance and help with this intricate subject. I am again very grateful for the opportunity to go to Moscow. It has been an honour to work with you.

Jonas Sweep
Nijmegen, April 2012
## Contents

<table>
<thead>
<tr>
<th>Preface</th>
<th>iii</th>
</tr>
</thead>
<tbody>
<tr>
<td>Contents</td>
<td>iv</td>
</tr>
<tr>
<td>1 Introduction</td>
<td>1</td>
</tr>
<tr>
<td>2 Physical concepts</td>
<td>3</td>
</tr>
<tr>
<td>2.1 Introduction to Condensed Matter Theory</td>
<td>3</td>
</tr>
<tr>
<td>2.2 Approximations</td>
<td>4</td>
</tr>
<tr>
<td>2.2.1 Ab initio theories</td>
<td>4</td>
</tr>
<tr>
<td>2.2.2 Model Hamiltonians</td>
<td>4</td>
</tr>
<tr>
<td>2.2.3 A note on dimensions</td>
<td>5</td>
</tr>
<tr>
<td>2.3 Effects that require attention</td>
<td>6</td>
</tr>
<tr>
<td>2.3.1 Hidden order</td>
<td>6</td>
</tr>
<tr>
<td>2.3.2 Metal-insulator transitions</td>
<td>6</td>
</tr>
<tr>
<td>2.3.3 Kondo effect and its supposed competition with long-range ordering</td>
<td>8</td>
</tr>
<tr>
<td>2.3.4 High temperature superconductivity</td>
<td>8</td>
</tr>
<tr>
<td>2.3.5 Volume changes in actinides</td>
<td>9</td>
</tr>
<tr>
<td>2.4 Examples of Quantum Lattice Models</td>
<td>9</td>
</tr>
<tr>
<td>2.4.1 Anderson Impurity Model</td>
<td>10</td>
</tr>
<tr>
<td>2.4.2 s-d model</td>
<td>10</td>
</tr>
<tr>
<td>2.4.3 Hubbard model</td>
<td>10</td>
</tr>
<tr>
<td>2.4.4 Heisenberg model</td>
<td>11</td>
</tr>
<tr>
<td>2.4.5 Ising model</td>
<td>11</td>
</tr>
<tr>
<td>3 Theoretical methods</td>
<td>13</td>
</tr>
<tr>
<td>3.1 Introduction</td>
<td>13</td>
</tr>
<tr>
<td>3.2 Path integrals</td>
<td>13</td>
</tr>
<tr>
<td>3.2.1 Slits and screens revisited</td>
<td>14</td>
</tr>
<tr>
<td>3.2.2 Coherent state path integral and the partition function</td>
<td>15</td>
</tr>
<tr>
<td>3.3 Matsubara formalism</td>
<td>17</td>
</tr>
<tr>
<td>3.3.1 Introduction of Matsubara frequencies</td>
<td>17</td>
</tr>
<tr>
<td>3.3.2 Definitions</td>
<td>19</td>
</tr>
<tr>
<td>3.3.3 Non-interacting Matsubara Green's functions</td>
<td>20</td>
</tr>
<tr>
<td>3.3.4 Retarded and advanced Green's functions</td>
<td>20</td>
</tr>
<tr>
<td>3.3.5 Interacting theory: the Dyson equation</td>
<td>21</td>
</tr>
<tr>
<td>3.4 Dynamical Mean Field Theory</td>
<td>22</td>
</tr>
<tr>
<td>3.4.1 Introduction</td>
<td>22</td>
</tr>
<tr>
<td>3.4.2 Cavity method</td>
<td>23</td>
</tr>
</tbody>
</table>
Chapter 1

Introduction

In this thesis the reader finds a report of my Master’s research in the field of Condensed Matter Theory. I performed this research under the supervision of d. f.-m. n. Alexey N. Rubtsov and prof. dr. Mikhail I. Katsnelson at the department of Quantum Electronics of the physical faculty of Moscow State University.

The group of quantum electronics develops methods to study correlated systems: systems in which the electrons do not move independently of one another. This work studies electronic correlations in the s-d model using a full quantum-mechanical computational method called Continuous-Time Quantum Monte-Carlo (ct-qmc).

Analysis of the s-d model can help us to understand more involved physical problems. It is a very useful testing ground to develop new methods to study materials and effects we do not yet understand. Among these effects are ‘hidden order’ and volume changes in actinides, metal-insulator transitions in metal-oxides and high temperature superconductivity in cuprates.

Over the years, the computational and analytical methods to treat theoretical models of condensed matter have become more and more sophisticated. The increase in computer power has allowed us to consider areas previously unavailable to calculation. Still, the aforementioned effects (‘hidden-order’ etc.) continue to puzzle us. That is why at this moment there is a trend of including non-local correlations in the calculation. This trend was started by considering small clusters of multiple sites, after that through more general approaches. Another trend is to include the interaction between electrons situated in different bands. All this in order to paint a more complete picture of the physics at hand. The research in this thesis is part of these trends. Firstly, since it considers non-local correlations through the introduction of Dual Fermions. The results from this non-local method are compared to a mean-field (=local) treatment. Secondly, because it treats two different types of electrons (localized and conduction) that interact via their spins.

The goal of this research is to study the s-d model at different interaction strengths ($J$) to obtain its density of states (DoS). From the DoS important properties of the model can be inferred, for instance if the material is conducting or insulating. It is this metal-insulator transition in which we are particularly interested. We would like to compare the DoS’s of the s-d model obtained within two different theoretical frameworks: Dynamical Mean Field Theory and Dual Fermions.

The scientific novelty of this work is that it includes non-local correlations through the Dual Fermion formalism as opposed to previous local calculations [1][2][3]. We also compare our results to a quasi-classical treatment of the same regime of the s-d model in [4] and [5]. At this moment, only the Hubbard model has been studied non-locally using Dual Fermions in [6] and [7]. Mathematically the Hubbard model and the s-d model are very alike (cf. [8]). Physically however, they describe different interactions. Therefore it is natural to extend the research of Antipov and also study the s-d model using Dual Fermions.
This work is structured as follows.

**Chapter 1** is this introduction.

**Chapter 2** summarises the physical relevance of this work. It introduces the subject and specifically the s-d model itself, within the context of condensed matter theory and discusses its (necessary) approximations. It also explains why it is useful to study a toy model like the s-d model.

**Chapter 3** forms the necessary theoretical background to study the s-d model. It contains an explanation of the important concepts and an introduction into the formalism used. The tools of Dynamical Mean Field Theory (DMFT) and Dual Fermions are treated concisely.

**Chapter 4** is about the study of the s-d model using a Quantum Monte-Carlo program. Here, I will first define the system we are studying. I will briefly describe the computational method used, show the results we achieved and discuss them afterwards.

**Chapter 5** is the final chapter. It consists of a summary, conclusions and an outlook for future scientific work in this area.

**Appendices** are included to review Grassman calculus (appendix A) and to present a proof of a series expansion that is employed in the derivation of the formalism (appendix B).
Chapter 2

Physical concepts

2.1 Introduction to Condensed Matter Theory

How should we describe condensed matter? Essentially we are dealing with a quantum many-body problem because there are about $10^{23}$ particles involved that interact on a quantum-mechanical level.

We could finish all discussion by stating that we should simply solve the many-particle Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} \psi(q_1 \ldots q_n) = \hat{H} \psi(q_1 \ldots q_n),$$

(2.1)

considering nuclei (with charge $Z_l$) and electrons with the following Hamiltonian

$$\hat{H} = \sum_l \frac{\vec{P}_l^2}{2M_l} + \sum_i \frac{\vec{p}_i^2}{2m} + \frac{e^2}{2} \sum_{ij} \frac{1}{r_i - r_j} + \sum_{l\nu} \frac{Z_l Z_{l'}}{R_l - R_{l'}} + e^2 \sum_l \frac{Z_l}{R_l - r_i}.$$  

(2.2)

Here we even approximate the nuclei as point charges. Should we want to include relativistic effects we need to write the equations of quantum electrodynamics instead of the Schrödinger equation:

$$\gamma^\mu (i\delta_\mu - eA_\mu) \psi = m\psi$$

(2.3)

$$\delta_\nu F^{\mu\nu} = 4\pi j^\mu,$$

(2.4)

with

$$F^{\mu\nu} = \partial_\mu A_\nu - \partial_\nu A_\mu$$

(2.5)

$$j^\mu = e\bar{\psi}\gamma^\mu \psi,$$

(2.6)

where the fields $\psi$ and $A$ satisfy anti-commutation and commutation relations respectively:

$$\{\psi(x), \psi(x')\} = \delta(x - x')$$

(2.7)

$$[A(x), A(x')] = \delta(x - x').$$

(2.8)

This solves most problems one could encounter in principle, but unfortunately not for our purpose: it is very hard to say something useful about the solid state using just the above equations. For example, a theory of the solid state should tell us why iron is ferromagnetic and chromium is antiferromagnetic, why iridium is brittle and platinum is ductile and it should give us the different allotropes of carbon. Unfortunately, no-one has yet succeeded to explain these issues directly from the fundamental equations. For this reason we should find approximate solutions that contain some extra ingredients.

This chapter explains how one can arrive at these approximate solutions in section 2.2. It treats the need for approximate solutions through some examples of puzzling effects in solid state physics in section 2.3. After that, it covers some examples of approximate models of the solid state in section 2.4.
2.2 Approximations

The aforementioned extra ingredients are assumptions, constraints or sometimes theorems that lead us
to important approximations that make theories workable and their results meaningful. Approximations
are often divided into two classes: ab initio theories and model Hamiltonians. Ab initio theories, also
called theories from first principles, use the full Hamiltonian and make approximations later. A model
Hamiltonian or (quantum) lattice model is already an approximation in itself. Namely, an approximation
of the real Hamiltonian is used to obtain results that are as exact as possible within this framework. A
refreshing account of physical approximation schemes in general is given in chapter 1 of [9]. Here we will
briefly explain some ideas of ab initio theories and treat model Hamiltonians.

2.2.1 Ab initio theories

The most famous example of an ab initio theory is Density Functional Theory (DFT). The ingredients
that form the foundation of this theory are two theorems proven by Hohenberg and Kohn in 1964. The
first theorem states that all the ground state properties of the material can be inferred if the ground state
electron density is known. The second theorem provides a recipe to calculate this electron density.

Over the last years DFT has been very widely used in the description of many physical systems. It is
used in solid state physics for electronic structure calculations of weakly correlated systems and in
quantum chemistry for the calculation of molecular properties. Due to its success, DFT became a real
hype but despite its remarkable impact on contemporary physics DFT is not accurate enough to describe
materials that exhibit strong correlations. Some metal-oxides such as nickel oxide and manganese oxide
 evade description by DFT because their electrons cannot be described independently. These metal oxides
are predicted to be conducting by DFT when magnetic long-range order is absent but in fact they are
insulators. Neville Mott showed that these materials, later called Mott insulators, are better described
using the Hubbard model, which we will explain shortly. Furthermore, if we are interested in more than
the ground state properties, for instance if we want to include thermal excitations, we need a different
approach.

2.2.2 Model Hamiltonians

In model Hamiltonians, also called quantum lattice models, some form of the Hamiltonian is assumed
before calculating anything. Often, the lattice of the material is used as input. Before DFT was introduced,
this was the only possible framework of calculating material properties. The notion of phonons and
other quasi-particles and the harmonic approximation are described within this approximation scheme.
Although we often calculate the spectra of phonons using DFT.

We will now treat a generic way of how to obtain a quantum lattice model containing electrons and
an impurity, following Kondo (1969) as quoted in [10]. An impurity is a site in the lattice with different
properties than the rest of the sites. Impurity models were initially used for materials containing some
kind of defect but they turned out to be interesting in their own right on a purely theoretical level.

The impurity is located at the position \( \vec{R}_n \) and it is described by a localised electron orbital \( \phi_L(\vec{r}-\vec{R}_n) \) if
an electron occupies the impurity. Otherwise, the electron is in a continuum state \( \phi_\ell(\vec{r}) \) with energy \( \epsilon_\ell \). This can be a Bloch state of the crystal or a plane wave, depending on the medium. We write a
generalised state function as a summation over all possible states:

\[
\psi(\vec{r}) = \sum_{k_\sigma} \phi_k^\sigma(\vec{r}) X_\sigma C_{k_\sigma} + \sum_{\ell} \phi_\ell(\vec{r}-\vec{R}_n) X_\sigma C_n.
\]  

(2.9)

Here \( X_\sigma \) are the spin wavefunctions and the \( C^\dagger \)'s are annihilation operators. We take a generic Hamiltonian
of the following form:

\[ \hat{H} = \sum_i \left[ \frac{\hat{p}_i^2}{2m} + U(r_i) \right] + \frac{1}{2} \sum_{ij} \frac{e^2}{r_{ij}}, \]  

(2.10)

in which the first term is the kinetic energy operator, \( U \) is the potential for each electron (this may be the lattice potential and it may include the impurity potential) and the last term describes the inter-electron interactions. When we evaluate this Hamiltonian without the inter-electron interaction term, for our generalised state function we end up with a Hamiltonian corresponding to our quantum lattice model

\[
\int d^3r \psi^\dagger(\vec{r}) \left[ \frac{\hat{p}^2}{2m} + U(\vec{r}) \right] \psi(\vec{r})
= \sum_{\vec{k}k\sigma} \epsilon_{\vec{k}} C^\dagger_{\vec{k}k\sigma} C_{\vec{k}k\sigma} + \sum_{\vec{k}k'} U_{\vec{k}\vec{k}'} C^\dagger_{\vec{k}k\sigma} C_{\vec{k}'k'\sigma} + \sum_{\vec{k}} M_{\vec{k}} (C^\dagger_{\vec{k}k\sigma} C_{\vec{k}L\sigma} + C^\dagger_{L\sigma} C_{\vec{k}k\sigma}).
\]  

(2.11)

The first two terms on the right-hand side are the unperturbed parts of the Hamiltonian describing the continuum and impurity states respectively. The third term describes the scattering of continuum functions by the impurity. The last term mixes impurity states and continuum states.

What happens when we include the electron-electron interactions? Then two integrals over position need to be performed. The interaction term can be cast in this form:

\[ \hat{H}_{int} = \int d^3r_1 d^3r_2 \frac{e^2}{|r_1 - r_2|} \psi^\dagger(r_1) \psi(r_1) \psi^\dagger(r_2) \psi(r_2). \]  

(2.12)

This expression generates many pairings of creation and annihilation operators. We will consider one term that exchanges spin between the impurity state and a continuum state:

\[ \hat{H}_{ex} = - \sum_{\vec{k}\vec{k}'\sigma\sigma'} J_{\vec{k}\vec{k}'} C^\dagger_{\vec{k}k\sigma} C_{\vec{k}'k'\sigma'} C^\dagger_{LL\sigma} C_{LL\sigma}, \]  

(2.13)

where \( J_{\vec{k}\vec{k}'} \) is given by the following overlap integral between localised and continuum states:

\[ J_{\vec{k}\vec{k}'} = e^2 \int d^3r_1 \phi^*_{\vec{k}}(r_1) \phi_{\vec{k}'}(r_1 - \vec{R}) \int d^3r_2 \frac{e^2}{|r_1 - r_2|} \phi^*_{\vec{k}'}(r_2 - \vec{R}) \phi_{\vec{k}}(r_2). \]  

(2.14)

The numerical value of \( J \) lies somewhere between \(-2\) and \(2\) eV and it can be obtained experimentally as was done many times for different materials in the 1950’s and 60’s. We will be seeing this \( J \) again when we consider the s-d model in more detail. For the rest of this thesis we will quote dimensionless values for the interaction strengths, \( J, U \) etc. They are taken in electron volts, unless otherwise indicated.

We have shown a way to obtain a model Hamiltonian quasi-rigorously from the full Hamiltonian. In this model both localised and continuum (conduction) electrons occur. Interesting physics arises when there is a competition between the localised and continuum character of the same electrons. Materials in which this occurs are called correlated materials, since the electrons do not move independently of one another.

Often, lattice models are simply postulated to describe interactions in a form that is convenient for calculations. Lattice models are frequently treated in a mean field picture: summarising the interactions of the entire lattice with one site in particular in the form of a single valued function called the mean field.

### 2.2.3 A note on dimensions

We will treat a mean field theory in which the mean field is not constant in time in section 3.4. This Dynamical Mean-Field Theory (DMFT) is used very widely to obtain results in quantum lattice models.
2.3 Effects that require attention

It becomes exact in the limit of an infinite amount of nearest neighbours, also called coordination number \( z \). An infinite coordination number is equivalent to infinite dimensions. Deviations from the dynamical mean field result scale as \( \frac{1}{z} \). DMFT applied to a Hubbard Hamiltonian accurately reproduces the three-peak structure observed in the density of states (DoS) of materials with a large coordination number. This seems reasonable when we note that a three dimensional fcc lattice already has \( z = 12 \). We might remember that (static) mean field theory for the Ising model already becomes exact in four dimensions.

One of the great success stories of DMFT is its combination with the Local Density Approximation (LDA), an approximation that stems from Density Functional Theory. This hybrid theory between DFT and DMFT employs a mean field approach to calculate the material density for the DFT calculation. It is called LDA+DMFT, see [11] for a review.

Especially in smaller dimensions DMFT produces unreliable results, which is to be expected because, as already mentioned, the error in DMFT scales as \( \frac{1}{z} \). In infinite dimensions there cannot be any spatial correlations. Including these spatial correlations improves the results in infinite dimensions.

We will first elaborate the need for quantum lattice models in the next section (2.3) and see a few examples of quantum lattice models in finite dimensions in section 2.4.

2.3 Effects that require attention

Although a lot of progress has been made in the field of correlated systems there are some effects within this field we do not yet understand. While these effects are interesting in their own right for the scientific community, they can also have important consequences for society. They may for instance benefit the safe storage of radioactive isotopes or the accessibility of high temperature superconductivity.

In a large number of cases, studying a simplified model, also called a ‘toy-model’, of a system that needs deeper understanding yields insights that are applicable to the more complicated system as well. Furthermore, one particular toy-model may be used to describe a multitude of systems allowing us to derive general results. We will now display a few physical effects for which toy-models can play an important explicative role. These effects are all part of the frontier of condensed matter physics at this moment. A concise overview of some of these effects is also given in [12].

2.3.1 Hidden order

The actinide compound URu\(_2\)Si\(_2\) exhibits a peculiar second order phase transition at 17.5 K, releasing a large amount of entropy. The low-entropy state was called ‘hidden-order’. The exact properties of the transition cannot be detected experimentally, hence the aspect ‘hidden’, but the material was analysed within an LDA+DMFT approach in [13]. In that study, some important properties were unveiled and a workable order parameter was defined but still the details of this hidden-order state remain... hidden.

The journey towards uncovering these details can be started by studying a toy-model using a non-local approach, such as the one that will be developed in 3.5. This toy model should contain multiple bands of electrons since the \( f \) electrons of the uranium hybridise with the \( spd \) electrons and become delocalised.

Hidden order is also suspected to play a role in the behaviour of cuprates [14].

2.3.2 Metal-insulator transitions

Some metal-oxides exhibit a large change in resistivity when parameters such as the temperature or the pressure are varied. In certain regimes they are metallic, in other regimes insulating. As mentioned before, this behaviour was first explained by Neville Mott, using the Hubbard model. This is the reason these materials are also called Mott insulators.

The conduction behaviour of Mott insulators depends only the interaction energy \( U \), often called Hubbard-\( U \). We may calculate the DoS of Mott insulators using for instance DMFT. In terms of the DoS
the Mott transition looks as follows. For an interaction energy $U = 0$ the DoS consists of a single band, like the top picture of fig. 2.1. If we increase $U$ to at least twice the non-interacting bandwidth, $W$, of the material, we obtain an insulator. As can be seen on the bottom picture of fig. 2.1 there are no states at the Fermi energy. The non-interacting bandwidth is given by

$$W = 2\sqrt{z \times t}.$$  \hspace{1cm} (2.15)

Where $z$ is the amount of nearest-neighbours and $t$ is the overlap integral between the localised states on the lattice or alternatively the hopping matrix element: the probability amplitude of an electron for the hopping from one lattice site to another.

A clear review of results on the metal-insulator transition in the Hubbard model is presented in [15].

Figure 2.1: A stereotypical density of states (DoS) of the electrons in a Mott insulator. The pictures show how the DoS varies as a function of the ratio of the interaction energy $U$ to the bandwidth $W$. (a) On the top picture the non-interacting case is shown: $U = 0$. It is a conducting state as the Fermi level ($E = 0$) is populated. The exact shape of the curve is dependent on the lattice of the material. (b) For small $U/W$, also called the weakly correlated regime, visible in the second picture, the onset of the insulating bands is visible. (c) In the strongly correlated regime, visible on the third picture, the onset of the insulating bands continues and the result is a three-peak structure. (d) In the insulating state: $U > 2W$ on the bottom picture, the interaction is strong enough to fully localise the electrons on a given site. An insulator is the result. Picture taken from [12].

---

1The exact shape of the DoS is dependent on the lattice-type but we will not worry about that here.
Metal-insulator transitions induced by changing interaction strengths can also be present in other models than the Hubbard model. They can for instance be due to magnetic effects. In some materials large changes in resistivity occur when the ambient magnetic field is changed. This magneto-resistance effect occurs in different orders of magnitude. Ordinary magneto-resistance (OMR) changes the resistivity of materials by roughly 10%. It was discovered by Lord Kelvin and it can be explained classically. Colossal magneto-resistance (CMR) was first seen in the 1950’s and changes the resistivity by a few orders of magnitude. It has been explained by a mechanism called ‘double exchange’ combined with the Jahn-Teller effect ([15] among others), although some scientific debate is still continuing. Giant magneto-resistance (GMR) and tunnel magneto-resistance (TMR) are present in thin film magnetic structures and stem from magnetic domains influencing each other. They can change the resistivity by more than 80%. GMR is applied in computer hard drives and its discovery is considered the birth of spintronics. Albert Fert and Peter Grünberg were awarded with the 2007 Nobel Prize in physics because they discovered GMR experimentally. GMR and TMR are treated theoretically in a spin-wave picture.

### 2.3.3 Kondo effect and its supposed competition with long-range ordering

The dominant contribution to the electrical resistivity of metals is the scattering of conduction electrons by nuclei that vibrate about their equilibrium lattice positions. With temperature more lattice vibrations are excited and the resistivity increases. At low temperatures, when the lattice vibrations are frozen out, there should be a constant residual resistivity owing to the scattering by impurities, defects or vacancies. However, in 1934 De Haas, De Boer and Van den Berg observed a resistance minimum in gold. This would mean the scattering probability would increase with decreasing temperature in this low-temperature regime. The mechanism behind this effect remained unknown until Kondo [17] reproduced the resistance minimum theoretically in 1964. He considered a scattering process in which spin is exchanged between a conduction electron and an impurity. The appearance of this resistance minimum was named the Kondo effect.

Kondo’s result contained a logarithmic divergence for $T \to 0$ that makes the result unreliable below a certain temperature $T_K$, later called the Kondo temperature. The challenge to extend the description of the Kondo effect below the Kondo temperature became known as the Kondo problem. Wilson [18] solved this problem by introducing the Renormalisation Group in 1974.

The next hurdle was to consider a lattice of Kondo impurities: a collection of sites that interact via their spins. This exactly corresponds to the s-d model (see subsection 2.4.2), which is only solved in one spatial dimension. For this reason, the s-d model is often named the Kondo Lattice model. Such a lattice exhibits an interplay between magnetic ordering of the Kondo impurities and screening of these impurities by (the scattering of) the conduction electrons, inhibiting the ordering. The magnetic ordering is caused by the indirect RKKY-interaction, named after Rudermann, Kittel, Kanaya and Yosida. This interaction is mediated by the conduction electrons of the s-d model. The localised electrons (on the impurities) do not interact directly. The RKKY-interaction tends to align impurity spins.

A persistent notion was introduced by Doniach in 1977, namely that the above interplay would lead to mutual suppression of either effect. So there would be no magnetic ordering as well as no Kondo scattering. The opposite turns out to be true, there are magnetically ordered phases that display Kondo physics. This was proved theoretically by Irkhin and Katsnelson in 1997 [19], and independently by Moeller et al. in 1999 [20].

A good review of Kondo physics and related developments can be found in [21].

### 2.3.4 High temperature superconductivity

High-temperature superconductivity is just as old as the writer of this thesis. However, it is pure coincidence that my thesis subject is related to high-temperature superconductivity. In 1986 Bednorz and Müller published a paper stating the discovery of the first high-temperature ($\hbar T_c$) superconductor [22].
2.4 Examples of Quantum Lattice Models

The discovery of \( h_T \) superconductivity was a major step forward for science in particular and for our world in general: superconducting materials have the potential of enhancing the quality of life of a great amount of people. This major step forward is the reason Bednorz and Müller were awarded the Nobel prize in physics for their discovery, already in 1987.

From that moment on until 2001 an astonishing amount of roughly 100,000 papers was published on the subject. 25 years later the mechanism that leads to \( h_T \) superconductivity is still not well understood.

A large class of \( h_T \) superconductors are the so-called cuprates. These are materials in which the electrons move in two-dimensional CuO\(_2\) layers. Cuprates are made by doping Mott insulators that exhibit a long range anti-ferromagnetic ordering at low temperature. In his 2002 paper Anderson claims the cuprates to be understood and that debate is not necessary anymore [23]. He is convinced a one-component Hubbard model on a square lattice is sufficient to describe \( h_T \) superconductivity in the cuprates. Despite this statement debate has been continuing. The description of cuprates gives different results for a cluster-treatment or a one-site treatment, there is no consensus about which way to go from there. A review of what is known up to 2006 about \( h_T \) superconductivity is written by Leggett [24].

On the website www.superconductors.org the reader can monitor the progress on the frontier of superconductor physics. At the time of writing, the highest transition temperature is 28 degrees Celsius, at ambient pressure. Unfortunately the specific material that exhibits this large transition temperature is hard to synthesise in large concentrations.

2.3.5 Volume changes in actinides

Actinides such as plutonium and cerium come in a large number of different allotropes: plutonium even has six known phases. Often, these allotropes have different densities. Therefore large volume changes (on the order of 15-25%) occur with changing temperature or pressure while the material remains in the solid state. This effect poses a serious challenge for the safe processing and storage of radioactive isotopes because the transition temperatures/pressures are easily attained while processing these materials.

Both cerium and plutonium were analysed within DMFT [12, 25] and while these computational results agree with experiment to some extent, there is still room for improvement.

2.4 Examples of Quantum Lattice Models

Now that we have seen that quantum lattice models have such a rich field of application we will consider a few of these quantum lattice models, as promised earlier. The most important distinction between these models is twofold, namely:

1. The types of electrons between which the interaction occurs can be different. They may be localised or conducting, residing on the same lattice sites or on different sites.

2. The type of the interaction itself can be different. It can be proportional to the charge (like the Coulomb interaction) or it may for instance depend on spins and thus of magnetic type.

The different lattice models can be considered on different lattice types such as cubic, hexagonal, and also in different dimensions. Although the (name of the) lattice model is the same, considering it on a different lattice can change the results. Therefore, it is important to keep track of the lattice type.

Here we will give the Hamiltonian of several models and discuss their main features. Some models are more well-known than others. The well-known ones are included to make it easier to appreciate the differences between the models more fully. All the models treated in this section are more than 50 years old and they are still studied to this very day. They are not solved exactly, at least in most spatial dimensions, but because our computational tools become more advanced, the approximate solutions of these models improve.
2.4 Examples of Quantum Lattice Models

2.4.1 Anderson Impurity Model

First we treat the Anderson Impurity Model. We start with this model because it can be used as a theoretical tool within the development of DMFT to summarise the behaviour a lattice so the entire lattice can be treated as if it were a single site. Furthermore, the models treated in subsections 2.4.2 and 2.4.3 can both be considered as limiting cases of the Anderson model.

In 1961 Anderson proposed his model [26] to describe the localised magnetic moments of iron-group metals dissolved in non-magnetic materials. For this he used the following general Hamiltonian that contains both localised ($c_{l,σ}$) and conduction electron orbitals ($c_{c,σ}$). These orbitals have a spin $σ$ which can be either up or down. The model describes an impurity much like the way we saw in subsection 2.2.2.

$$H = \sum_{σ} ϵ_l n_{l,σ} + Un_{l,↑}n_{l,↓} + \sum_{k,σ} ϵ_k c_{kσ}^+ c_{kσ} + \sum_{k,σ} (V_{k,l} c_{kσ}^+ c_{l,σ} + V_{k,l}^* c_{lσ}^+ c_{kσ})$$  \hspace{1cm} (2.16)

The first term contains the energies of the localised electrons. The second term, the interaction term with $U$, describes a Coulomb interaction between localised electrons residing on the same site. It is also called a Hubbard-interaction term. The third term describes the kinetic behaviour of the conduction electrons, characterised by a dispersion $ϵ_k$. The interaction term with $V$ is general (magnetic or Coulomb) and it describes interactions between localised and conduction electrons. Because it couples two different kinds of electrons, this term is called the hybridisation.

2.4.2 s-d model

Vonsovskiy introduced the s-d model in 1946. It has been used to explain many effects in different magnetic systems over the years. Among others, the effects mentioned in the previous section 2.3. Although the model is celebrating its 66th birthday this year it is still far from retirement. In this thesis we will look at spatial correlations in the s-d model.

We can rigorously obtain the s-d Hamiltonian for one lattice site from the Anderson model as is shown in [8]. In short, this amounts to taking the limit $U \to \infty$ in the Anderson Hamiltonian. To obtain the model on a lattice, the same interaction for all the lattice sites. The result is:

$$H = -\sum_{i,j,σ} t_{ij} (c_{i,j,σ}^+ c_{i,σ}) + J \sum_{i,σ} \vec{S}_i \cdot \vec{s}_i.$$  \hspace{1cm} (2.17)

The first term represents conduction electrons ($d$-electrons in the original model) hopping from site $i$ to site $j$. The second term contains localised spin operators $\vec{S}_i$ (corresponding to the $s$-electrons in the original model) and spin operators $\vec{s}_i$ of a conduction electron that interacts with the localised electron at site $i$. The coupling constant $J$ can be both positive (anti-ferromagnetic interaction) and negative (ferromagnetic interaction).

The s-d model is only exactly solvable in one dimension using the Bethe ansatz [27], as Andrei and Wiegmann showed independently [28, 29]. The s-d model consists of a lattice of impurities that interact with conduction electrons. The interaction is proportional to the spin of the interacting particles, just like in the Kondo model. For this reason the s-d model is often referred to as the Kondo Lattice Model. We include a graphical representation of the s-d model in fig. 2.2.

2.4.3 Hubbard model

The Hubbard model is also a limiting case of the Anderson model. It is the case when the hybridisation is taken to zero. This model contains only localised electrons but now these are allowed to hop between

---

Wiegmann notes in passing that actually the entire Anderson model is solvable in one dimension using the Bethe ansatz.
neighbouring sites. The Hubbard Hamiltonian is:

\[ H_{\text{Hub}} = - \sum_{i,j,\sigma} t_{ij} (c_{i,\sigma}^\dagger c_{j,\sigma}) + U \sum_{i} n_{i,\uparrow} n_{i,\downarrow}. \]  

The interaction \( U \) is between localised electrons on the same site. Since it is energetically unfavourable to reside on the same site, the electrons will likely remain localised because of this term. The first term represents a hopping of the localised electrons from site to site, just like there is a hopping in the s-d model. In short, the terms in the Hubbard Hamiltonian result in a competition between localised and kinetic behaviour of the electrons. This competition is exactly the one we referred to in the section 2.2.2, which makes the Hubbard model the stereotype of a correlated system.

### 2.4.4 Heisenberg model

The Heisenberg model models the magnetic interaction between neighbouring sites. These sites can in principle have any spins whereas the previous models are mostly used for \( S = \frac{1}{2} \). The full spin-vector is taken into account. There are no different kinds of electrons in this model, only a nearest-neighbour interaction between fixed sites and an interaction of the sites with an optional external field \( h \). The energy is minimised when the neighbouring spins are aligned as much as possible for a positive value of \( J \), the ferromagnetic case, and when there is alignment with the external field as well. 

\[ H = - \sum_{<ij>} J_{ij} \sigma_i \cdot \sigma_j - h \sum_{i} \sigma_i \]  

For \( S = \frac{1}{2} \) and in three dimensions the \( \sigma \)'s in the above expression become the Pauli matrices.

### 2.4.5 Ising model

The Ising model (1920) contains sites that can have spin up or down, there is only one direction for the spins as opposed to the Heisenberg model. Again the nearest-neighbours interact and there are no
conduction electrons.

\[ H = -J \sum_{<ij>} \sigma_i \sigma_j - h \sum_i \sigma_i \]  

(2.20)

Although the Hamiltonian looks a lot like the Heisenberg model we stress once more that they are really different.

The one-dimensional Ising model was solved by Ising himself in 1925. The solution of the two-dimensional case took another 19 years. Onsager solved it in 1944. The Ising model can be generalised by making \( J \) dependent on the distance and sum over all pairs instead of only nearest-neighbours.

The Ising model is the stereotype model to apply the mean field idea to. Mean field theory focuses on one particular site, say \( i = 0 \) governed by an effective Hamiltonian:

\[ H_{\text{eff}} = - h_{\text{eff}} \sigma_0. \]  

(2.21)

The effective field contains all the other interactions with \( m_i = \langle \sigma_i \rangle \), the magnetisation at site \( i \).

\[ h_{\text{eff}} = h + \sum_i J_{0i} m_i = h + z J m. \]  

(2.22)

The sum only goes over the nearest neighbour sites, which causes the number of nearest neighbours \( z \) to appear. This yields the following equation for the total magnetisation \( m \):

\[ m = \tanh(\beta h + z \beta J m). \]  

(2.23)

Here we also see the temperature entering the expressions through \( \beta = 1/(k_B T) \).

Lumping the contributions of all the sites into one mean field works best in the limit where the number of nearest-neighbours, the lattice coordination \( z \), becomes large. In the case of the Ising model, the mean field approach is exact for a dimensionality \( d \geq 4 \). Intuitively this can be understood because if the amount of neighbours becomes large they might be treated as a uniform bath.

If we want to take the amount of neighbours to infinity we should rescale the coupling constant \( J \) as \( J = J^* / z \) to keep the entropy and the internal energy per site finite. In this particular mean field approach all time dependence is neglected. We generalise this mean field treatment in the following chapter in the form of Dynamical Mean Field Theory.
Chapter 3

Theoretical methods

3.1 Introduction

Now that we have seen some of the physics that requires our attention, we focus on the theoretical tools to deal with the problems at hand. We will need quite a heavy arsenal if we want to study strongly correlated systems. This is because we want to generalise the mean field picture to contain time-dependent correlations, and to look at spatial fluctuations and see what their contributions are to the physics of the s-d model.

So, what parts does this heavy arsenal contain? To begin with, we need the path integral to develop the temporal generalisation of mean field theory, \( \text{DMFT} \). As we have seen, the number of nearest neighbours, also called coordination number, is proportional to the dimensionality of the system. As the coordination number decreases spatial correlations become more important. Only in materials with a large number of nearest neighbours, \( z \), \( \text{DMFT} \) has a chance of succeeding. The theory of Dual Fermions is a spatial generalisation of \( \text{DMFT} \) and thus a good candidate to handle these spatial correlations. \( \text{DMFT} \) and Dual Fermions are formulated with heavy use of the Matsubara formalism: a technique particularly useful in (computational) condensed matter theory. It is useful because it simplifies computational work: quantities of interest (observables) can be read off in some cases, or their parsing/processing is straightforward. A central quantity in all this is the Green’s function or correlation function. It is used to obtain densities of states.

We cannot hope to give a detailed account of all the theoretical tools that are relevant for this study, since this beyond the scope of this thesis. We humbly refer to [10], [30], and [31] should the reader desire more context.

This chapter is devoted to the development of the mentioned arsenal. It starts with explaining path integrals in section 3.2. It applies them to fermionic fields in section 3.2.2 introducing the notion of coherent states. After that, the Matsubara formalism is introduced in section 3.3. This formalism is then put to use in the derivation of \( \text{DMFT} \) in section 3.4. The extension of \( \text{DMFT} \) describing spatial fluctuations, the Dual Fermion formalism, is shortly introduced in section 3.5.

3.2 Path integrals

Lagrangian mechanics are a useful way of describing reality. In particular, Lagrangian mechanics has helped us to define quantum field theory (QFT). The branch of QFT that we get by quantising Lagrangian mechanics is often referred to as the ‘path-integral’ branch.\(^1\) Although mathematically the usage of path integrals is controversial we will not bother with that since the method allows physicists around the world

---

\(^1\)As opposed to the ‘bracket’ branch, which comes from quantising Hamiltonian mechanics.
to make (accurate) predictions about nature to this very day and, according to Karl Popper, predictions are what science is all about, cf. [32], [33]. We will therefore say no more words on this matter.

In this section we will review very briefly what a path integral is. We will later use it to see how the action is treated in the system of interest. For this section I benefited greatly from the wisdom of Anthony Zee as displayed in his recent textbook on QFT [34]. A comprehensive treatment is also given in [35].

3.2.1 Slits and screens revisited

As we know from classical (Lagrangian) mechanics, the action (usually $S$) is a useful quantity. A quantum Lagrangian theory is obtained by using the so-called path integral. We will start from the path integral and the action and treat what we need from QFT in a nutshell from there. As a warm up, I will retell the story of how the path integral might have been discovered.

Imagine a quantum mechanics class treating the double slit experiment. We remember the superposition principle: the probability amplitude for detection is given by the sum of the amplitude for the particle to propagate from the source $Q$ to the first slit $A_1$ and then to the detector $O$ and the amplitude for the particle to propagate from $Q$ to the second slit $A_2$ and then to $O$.

Suddenly, a very bright student, let us call him Richard Feynman, asks: What if we drill three holes in the screen instead of two? Or even four, or five?

Well, obviously we have to sum over the amplitudes for passing through each slit separately. The professor answers.

Let us denote that as

$$A = \sum_i A(Q \rightarrow A_i \rightarrow O). \quad (3.1)$$

"OK, but what if we add another screen, say $B$, with a number of holes in it?" Feynman asked. "Then we need to take the amplitude from source $Q$ to the hole $A_i$ to the hole $B_j$ and from there to the detector at $O$, and sum over $i$ and $j$. But what if we add a third screen, and a fourth one. Or what if we drill an infinite number of holes, so the screen is not really there?"

This last observation leads us to the conclusion that for a particle to propagate through empty space we need to sum all the amplitudes for the particle to go through all the different points between the source and the detector. In other words, we need to sum over all the possible paths a particle can take from the source to the detector. If we let the number of points between the source and the detector go to infinity in this way, we actually perform an integral over all paths. Hence the name: "path integral".

Now let us make this notion of integrating over paths more precise. The probability $P_{i \rightarrow f}$ of a system in an initial state $q_i$ to end up in a final state $q_f$ after a time $t$ can be calculated using the time evolution operator as follows:

$$P_{i \rightarrow f} = \langle q_f | e^{iHt} | q_i \rangle, \quad (3.2)$$

where $H$ is the Hamiltonian. We will divide the time $t$ in $N$ equal steps $\delta t = t/N$, then

$$\langle q_f | e^{iHt} | q_i \rangle = \langle q_f | e^{iH\delta t} e^{iH\delta t} \ldots e^{iH\delta t} | q_i \rangle. \quad (3.3)$$

These states are normalised to a Dirac delta function: $\langle q' | q \rangle = \delta(q' - q)$. Next, we insert a completeness relation $\int dq |q\rangle \langle q| = \hat{1}$ between every exponential and we get:

$$\langle q_f | e^{iHt} | q_i \rangle = \prod_{j=1}^{N-1} \int dq_j \langle q_f | e^{iH\delta t} | q_{N-1} \rangle \langle q_{N-1} | e^{iH\delta t} | q_{N-2} \rangle \ldots \langle q_2 | e^{iH\delta t} | q_1 \rangle \langle q_1 | e^{iH\delta t} | q_i \rangle. \quad (3.4)$$

We let $p_i$ be the conjugate momentum to $q_i$. If we have a Hamiltonian that contains momenta $p$ as well as positions $q$ then we can show (cf. [35]) that if these $p$'s and $q$'s are Weyl ordered that the above
expression leads to

\[ \langle q_f | e^{iHt} | q_i \rangle = \int Dq(t') e^{i \int_0^t dt' L(q, \dot{q})}. \]  

Here we employ the notation using the measure \( D \), it signifies integrating all the paths \( q(t') \) that depend on time \( t' \), using the correct boundary conditions: \( q(0) = q_i \) and \( q(t) = q_f \).

The Hamiltonian here contains operators but as they are working on a complete set of eigenstates, we replace them with the corresponding eigenvalues (numbers!) in the Lagrangian. This is the real strength of this approach.

In the above story we only spoke about points \( q_j \) in between a source and a detector. In general we will consider states instead of only positions. The next step to obtain a multi-particle (or field-) theory is to consider the eigenvalues \( q \) as expectation values of a field called \( \phi(\vec{x}) \) that depends on space and time. Our action then becomes a Lagrangian density, \( \mathcal{L} \), integrated over space-time. To conclude, we remark that we can abandon the Hamilton-picture altogether if we use the Lagrangian density to define the temporal behaviour described by the Hamiltonian:

\[ \int D\phi e^{i \int d^4x \mathcal{L}} := \langle \phi_f(\vec{x}, t) | e^{iHt} | \phi_i(\vec{x}, 0) \rangle. \]  

### 3.2.2 Coherent state path integral and the partition function

To calculate and to work with partition functions corresponding to our problem, which contains electrons (fermions), more easily we apply the above ideas specifically to fermionic operators. This is needed to prove the necessary relations in DMFT and to develop the Dual Fermion formalism. This subsection might seem a repetition of the previous one but I include it to better get used to the ideas. Again, the approach relies heavily on the ability to work with numbers instead of operators.

This time states are expanded in terms of eigenstates of the annihilation operator, also called coherent states. Although fermionic coherent states have no underlying physical meaning (they do not correspond to physical Fock states, contrary to bosonic coherent states), they are useful in this derivation. The concise but clear treatment of this material in [7] was very helpful.

The fermion coherent state \(| \eta \rangle \) is defined as

\[ | \eta \rangle = e^{-\sum \eta_\alpha c_\alpha^\dagger} | 0 \rangle, \]  

where \( \alpha \) labels the single particle states (spin, momentum, etc.). Coherent states are eigenstates of the annihilation operator:

\[ c_\alpha | \eta \rangle = \eta_\alpha | \eta \rangle. \]  

As the fermionic operators anticommute, their eigenvalues should also anticommute: they are Grassmann numbers. For a review of Grassmann numbers and how to integrate them see appendix A.

The coherent states form an overcomplete set on which we will project our physical states. They satisfy

\[ \int d\eta_\alpha^* d\eta_\alpha e^{-\sum \eta_\alpha^* \eta_\alpha} | \eta \rangle \langle \eta | = \hat{1}. \]  

Let us insert the above completeness relation in the definition of the grand-canonical partition function, so we project the states \( | n \rangle \) on the coherent states \( | \eta \rangle \):

\[ Z = \text{Tr} e^{-\beta(H - \mu N)} = \sum_n \langle n | e^{-\beta(H - \mu N)} | n \rangle = \int d\eta_\alpha^* d\eta_\alpha e^{-\sum \eta_\alpha^* \eta_\alpha} \langle -\eta | e^{-\beta(H - \mu N)} | \eta \rangle. \]  

The minus sign in \( \langle -\eta \rangle \) is there because Grassmann numbers anticommute. We may look at the above expression as the trace over the imaginary time evolution operator. Imaginary time was introduced by
3.2 Path integrals

Matsubara \[22\] in 1955 by Wick rotating the real time axis in the complex plane, relating time to inverse temperature: $i\tau \rightarrow 1/T = \beta$. In short: $U = e^{-iHt} \rightarrow e^{-\beta(H - \mu N)}$. We will see more of this in section \[33\].

We can define a path integral over all imaginary time trajectories in much the same way as we did in the previous subsection \[3.2.1\]. We divide the imaginary time interval $[0, \beta]$ into $L$ slices of width $\epsilon = \beta/L$, insert completeness relation eq. (3.9) between every slice and take the continuum limit $L \rightarrow \infty$.

\[
\mathcal{Z} = \lim_{L \rightarrow \infty} \int \prod_{l=0}^{L} d\eta^*_l d\eta_l e^{-\sum_n \eta^*_l \eta_l \epsilon_l (\epsilon_l (H - \mu N))^{\eta_l}} \]

\[
= \lim_{L \rightarrow \infty} \int \prod_{l=0}^{L} d\eta^*_l d\eta_l e^{-\sum_n \eta^*_l \eta_l \epsilon_l e^{-\sum_{l=0}^{L} \sum_n \eta^*_l \eta_l \epsilon_l (\epsilon_l (H - \mu N))^{\eta_l}}} \times \langle e^{-\epsilon (H - \mu N)\eta_0} \rangle. \tag{3.12}
\]

This expression contains a total of $L$ matrix elements. We may now evaluate these if we take the operators they contain to be in normal ordered form. That means all terms have the creation operators on the left and the annihilation operators on the right. If the Hamiltonian is normal ordered, its exponential form above is normal ordered up to an error of order $\epsilon^2$, which we can neglect in the continuum limit. We thus get:

\[
\langle \eta|e^{\tau H_{\epsilon}}|\eta_{\beta-1}\rangle = e^{\sum_n \eta^*_l \eta_{l-1} - \epsilon H[\eta^*_l, \eta_{l-1}]} \epsilon^2 \]

(3.13)

Taking a trace is invariant under cyclic permutation, therefore and because of the usage of anticommutating Grassmann numbers we have antiperiodic boundary conditions: $\eta_f = -\eta_i$ for all trajectories. We also have $\eta^* = -\eta_f$ and $\eta_0 = \eta_i$. Finally this gives us for the action, in terms of numbers instead of operators:

\[
S[\eta^*, \eta] = \sum_{l=0}^{L} \left( \sum_n \eta^*_l \eta_{l-1} \frac{(\eta_{l-1} - \eta_{l-1})}{\epsilon} - \mu \eta_{l-1} \right) + H[\eta^*_l, \eta_{l-1}]. \tag{3.14}
\]

Having numbers instead of operators in the action is essential as it allows us to easily manipulate the expressions. When we take the continuum limit, the first term between the curly braces becomes a derivative and all the $\eta$'s become dependent on the imaginary time $\tau$, instead of on the fixed value $l$. We get the following partition function $\mathcal{Z}$ in its final form:

\[
\mathcal{Z} = \int_{\eta_i = -\eta_f} (D[\eta^*_l, \eta_l] e^{-\int_0^\beta d\tau \sum_n \eta^*_l(\partial_\tau - \mu)\eta_l(\partial_\tau) + H[\eta^*_l, \eta_l]}]. \tag{3.15}
\]

We again symbolically denote the product of different integration measures from eq. (3.12) with the large $D[\eta^*_l, \eta_l]$. The square brackets remind us that the integration variable $\eta$ is itself a function of $\tau$. We adopt the following shorthand notation, with $c$ and $c^*$ representing the Grassmann fields:

\[
\mathcal{Z} = \int D[c^*, c] e^{-S[c^*, c]} \tag{3.16}
\]

And the Green's function describing correlations between fermionic states with different frequency, momentum, orbital or spin indices (labeled by 1, 2), we write as follows:

\[
G_{12} := (c_1 c_2^*) = \int D[c^*, c] c_1 c_2 e^{-S[c^*, c]}. \tag{3.17}
\]

These forms are used in all texts about DMFT and Dual Fermions. They are shorthand and therefore allow us to prove the necessary relations in a concise way and do not make us lose track of the general story and what we are doing. We should however keep in mind that we are indeed working with expressions such as eq. (3.12).
3.3 Matsubara formalism

In this section we treat the concept of imaginary time. To understand what the aforementioned Wick rotation to the imaginary time axis really means we take a moment to develop a different approach to see where this rotation comes from.

The Matsubara formalism is an elegant way of doing quantum field theory at temperatures greater than zero. The need for such a treatment is understandable if we note that everything around us has a non-zero temperature. There are situations in which a $T=0$ approximation is acceptable but we would like to keep track of the thermal excitations. Also, in some regimes the magnitude of the thermal excitations is of the same order as the energy scales in the problem. Furthermore, seemingly neglectable low-energy excitations can play a key role in the behaviour of some materials: the interplay between low-energy spin-flip excitations and fermionic excitations is for instance believed to lead to high temperature superconductivity.

As it turns out the Matsubara formalism has quite simple expressions, which is why we use this formalism and not a different way to keep track of the temperature. It does however challenge our physical intuition.

This section is largely based on the intelligible treatment in the book written by Gerald D. Mahan [10]. It is organised as follows: First, Matsubara frequencies are introduced as poles from the Fermi or Bose distribution function. Then the definition of the Green’s function in terms of these Matsubara frequencies is covered. After this, we take a look at interacting Green’s functions with the help of Dyson’s equation.

3.3.1 Introduction of Matsubara frequencies

We will be working in the Hamilton formalism in this subsection, because it leaves more room to understand the concepts. The Hamilton and Lagrange approach are equivalent. Yet in practice, often the Hamiltonian is given to demonstrate the physics and when something needs to be calculated, we silently and swiftly switch to path integrals and the Lagrangian to prove the necessary relations.

As has often proved convenient we write the Hamiltonian as

$$\hat{H} = \hat{H}_0 + \hat{V}, \quad (3.18)$$

where $\hat{V}$ is a perturbation that is small compared to $\hat{H}_0$.

We are working in the interaction-representation. Therefore, we introduce the time-evolution of an operator $\hat{O}$ via the usual exponential factor:

$$\hat{O}_{\text{per}}(t) = e^{it\hat{H}} \hat{O}_{\text{per}} e^{-it\hat{H}}. \quad (3.19)$$

Note that exponentials with $\hat{V}$ will be cropping up here, as we are used to.

Since we are working at non-zero temperature we first assume there is something of non-zero temperature. That is, our particle of interest is in contact with some heat bath made up of other particles with an average energy determined by the temperature $T$. The exact state of this bath is not known as we allow for fluctuations between different configurations.

To calculate expectation values (of which a Green’s function is a nice example) we must trace over all possible states and incorporate the thermal excitations by the a Gibbs distribution $e^{-\beta \hat{H}}$. In the case of a Green’s function $G(\vec{p};t,t')$ describing the correlation between a state with momentum $\vec{p}$ at time $t$ and $t'$, we get an expression that looks like this:

$$iG(\vec{p};t,t') = \langle \hat{C}_{\vec{p} \sigma}(t) \hat{C}_{\vec{p} \sigma}^\dagger(t') \rangle = \frac{\text{Tr}[e^{-\beta \hat{H}} \hat{C}_{\vec{p} \sigma}(t) \hat{C}_{\vec{p} \sigma}^\dagger(t')]}{\text{Tr}[e^{-\beta \hat{H}}]}, \quad (3.20)$$
3.3 Matsubara formalism

with \( \hat{C}^\dagger \) and \( \hat{C} \) creation and annihilation operators of particles with momentum \( \vec{p} \) and spin \( \sigma \), \( \beta = 1/(k_BT) \) the inverse temperature, and as before

\[
\text{Tr}[\ldots] := \sum_n \langle n | \ldots | n \rangle. 
\]

(3.21)

Note again that the above expansion results in exponentiated factors of the interaction \( \hat{V} \).

One would expect these two exponentials (one corresponding to the time evolution and the other corresponding to the Gibbs distribution) of \( \hat{V} \) to result in two different power series. But Matsubara [25] in 1955 found a way to implement both exponentials using just one expansion, treating temperature and time as the real and imaginary parts of a complex variable. Furthermore, Matsubara’s method exploits the functional form of the Fermi and Bose occupation functions.

These occupation functions are a useful starting point. They may be expanded as a series using the deviation from the chemical potential as follows:

\[
n_{\text{Bose}}(\omega) = \frac{1}{e^{\beta \omega} - 1} = -\frac{1}{2} + \frac{1}{\beta} \sum_{n=-\infty}^{\infty} \frac{1}{2n\pi i/\beta - \omega}, 
\]

(3.22)

\[
n_{\text{Fermi}}(\epsilon) = \frac{1}{e^{\beta \epsilon} + 1} = \frac{1}{2} + \frac{1}{\beta} \sum_{n=-\infty}^{\infty} \frac{1}{(2n+1)\pi i/\beta - \epsilon}, 
\]

(3.23)

where \( \epsilon \) and \( \omega \) are the deviations from the chemical potential \( \mu \). These expressions are proven in appendix B by integrating a function that resembles the occupation function over a particular contour in the complex plane.

The boson occupation function has poles at \( \omega = 2n\pi i/\beta \), the fermion function at \( \epsilon = (2n+1)\pi i/\beta \), for integer \( n \). Since we will use the frequency values of these poles more often in the near future, let us name them Matsubara frequencies:

For bosons: \( \omega_n = \frac{2n\pi}{\beta} \)

For fermions: \( \omega_n = \frac{(2n+1)\pi}{\beta} \).

(3.24)

(3.25)

The preceding summations eqs. (3.22) and (3.23) may be written as:

\[
\sum_n \frac{1}{\omega_n - \omega} \quad \text{or} \quad \sum_n \frac{1}{\omega_n - \epsilon}. 
\]

(3.26)

The terms within the summation resemble the Green’s functions we know from electrodynamics and indeed they are the Green’s functions in the Matsubara method, as we shall see.

As was mentioned before, time becomes a complex quantity \( \tau = it \) of which the real part is related to the temperature and the imaginary part is the real time.

The Matsubara Green’s functions are functions of \( \tau \), with \( \tau \) taking values from \(-\beta\) till \( \beta \). A function \( f(\tau) \) defined on a finite interval, in our case \([-\beta, \beta]\), can be represented by the following Fourier series:

\[
f(\tau) = \frac{1}{2} a_0 + \sum_{n=1}^{\infty} \left[ a_n \cos \left( \frac{n\pi \tau}{\beta} \right) + b_n \sin \left( \frac{n\pi \tau}{\beta} \right) \right], 
\]

(3.27)

with, as usual

\[
a_n = \frac{1}{\beta} \int_{-\beta}^{\beta} d\tau f(\tau) \cos \left( \frac{n\pi \tau}{\beta} \right),
\]

(3.28)

\[
b_n = \frac{1}{\beta} \int_{-\beta}^{\beta} d\tau f(\tau) \sin \left( \frac{n\pi \tau}{\beta} \right).
\]

(3.29)
We define the different individual components \( f(i\omega) \) of the sum (3.27) as

\[
f(i\omega_n) = \frac{1}{2} \beta (a_n + ib_n).
\]

(3.30)

So that

\[
f(\tau) = \frac{1}{\beta} \sum_{n=-\infty}^{\infty} e^{-in\pi\tau/\beta} f(i\omega_n).
\]

(3.31)

Using this, we obtain the inverse transformation

\[
f(i\omega_n) = \frac{1}{2} \int_{-\beta}^{\beta} d\tau f(\tau) \alpha^{in\pi\tau/\beta}.
\]

(3.32)

For fermions, of particular interest to us, we can write this as:

\[
f(i\omega_n) = \int_{0}^{\beta} d\tau e^{i\omega_n \tau} f(\tau)
\]

(3.33)

\[
f(\tau) = \frac{1}{\beta} \sum_{n} e^{i\omega_n \tau} f(i\omega_n)
\]

(3.34)

\[
\omega_n = (2n + 1)\pi k_B T.
\]

(3.35)

With these tools in hand, we may finally define the Matsubara Green’s function. After a while we will notice that they are easy to use and that they directly relate to physical observables.

### 3.3.2 Definitions

The Matsubara Green’s function is defined like this:

\[
G(\vec{p}, \tau', \tau) = -\langle T_\tau \hat{C}_\sigma \vec{p} (\tau') \hat{C}_\sigma^\dagger \vec{p} (\tau) \rangle,
\]

(3.36)

where \( T_\tau \) is the time ordering operator, which ensures that the operators on the right have the smallest time, so their \( \tau \) is closest to \(-\beta\). \( \vec{p} \) represents the momentum of the particle created by \( C_\sigma^\dagger \), \( \sigma \) is the spin-index. As stated in subsection [3.3.1](#), the brackets amount to tracing over a complete set of states. At the same time we include the time-evolution via the usual exponential factor, but using complex time \( \tau = it \).

We work with the grand-canonical ensemble allowing a variable number of particles. \( \Omega \) is the thermodynamic grand potential, \( \mu \) the chemical potential and \( N \) is the particle number operator. To clean up the notation we will omit the hats from now on. When we introduce the time evolution for \( \tau \) just like in eq. (3.19) and include the Gibbs distribution, our Green’s function looks as follows:

\[
G(\vec{p}, \tau', \tau) = \langle e^{-\beta(H-\mu N-\Omega)}T_\tau e^{\tau(H-\mu N)}C_\sigma \vec{p} e^{-(\tau-\tau')(H-\mu N)}C_\sigma^\dagger \vec{p} e^{-\tau'(H-\mu N)} \rangle,
\]

(3.37)

where, for normalisation,

\[
e^{\beta\Omega} = \text{Tr}(e^{-\beta(H-\mu N)}).
\]

(3.38)

We did not time order eq. (3.20) because it contained not all (imaginary) time information. From eq. (3.37) we will see the real beauty of Matsubara’s approach: all the exponentials will later on be collected into one.

It can be proven that the function on the right hand side of equation (3.37) is only a function of the difference \( \tau - \tau' \). We will not do this here since it is not so instructive but with this knowledge we may drop the \( \tau' \) altogether, or equivalently put it to zero.
3.3 Matsubara formalism

3.3.3 Non-interacting Matsubara Green’s functions

To treat the special case of the non-interacting ($V = 0$) Green’s function, we proceed as follows. We take the following Hamiltonian and we write $K$ for the combination of $H$ and the chemical potential term $\mu N$:

$$H = H_0 = \sum_{\vec{p}\sigma} \epsilon_{\vec{p}} C_{\sigma}^{\dagger} C_{\sigma}$$  \hspace{1cm} (3.39)

$$K_0 = K \equiv H - \mu N = \sum_{\vec{p}\sigma} \epsilon_{\vec{p}} C_{\sigma}^{\dagger} C_{\sigma}$$  \hspace{1cm} (3.40)

$$\epsilon_{\vec{p}} = \epsilon_{\vec{p}} - \mu. \hspace{1cm} (3.41)$$

We introduce the time-evolution of the operators with the help of the Baker-Hausdorff theorem:

$$C_{\vec{p}\sigma}(\tau) = e^{K_0 \tau} C_{\vec{p}\sigma} e^{-K_0 \tau} = e^{-\epsilon_{\vec{p}} \tau} C_{\vec{p}\sigma}$$  \hspace{1cm} (3.42)

$$C_{\vec{p}\sigma}^{\dagger}(\tau) = e^{K_0 \tau} C_{\vec{p}\sigma}^{\dagger} e^{-K_0 \tau} = e^{\epsilon_{\vec{p}} \tau} C_{\vec{p}\sigma}^{\dagger}.$$  \hspace{1cm} (3.43)

The Green’s function looks a bit different for $\tau$ larger or smaller than zero. We can express this difference using a step function $\Theta(\tau)$. This enables us to write the time-evolution more easily.

$$G_0(\vec{p}, \tau) = -\Theta(\tau)e^{-\epsilon_{\vec{p}} \tau} \langle C_{\vec{p}\sigma} C_{\vec{p}\sigma}^{\dagger} \rangle + \Theta(-\tau)e^{-\epsilon_{\vec{p}} \tau} \langle C_{\vec{p}\sigma}^{\dagger} C_{\vec{p}\sigma} \rangle$$  \hspace{1cm} (3.44)

$$= e^{-\epsilon_{\vec{p}} \tau} \left[ \Theta(\tau) \{ 1 - n_F(\epsilon_{\vec{p}}) \} - \Theta(-\tau) n_F(\epsilon_{\vec{p}}) \right], \hspace{1cm} (3.45)$$

where

$$n_F = \langle C_{\vec{p}\sigma}^{\dagger} C_{\vec{p}\sigma} \rangle = \frac{1}{e^{\beta \epsilon_{\vec{p}}} + 1}. \hspace{1cm} (3.47)$$

With our previous definitions (3.33) we obtain the Matsubara Green’s function of frequency for fermions:

$$G_0(\vec{p}, i\omega_n) = \int_0^\beta d\tau e^{i\omega_n \tau} G_0(\vec{p}, \tau) = -(1 - n_F) \int_0^\beta d\tau e^{\tau(i\omega_n - \epsilon_{\vec{p}})}$$  \hspace{1cm} (3.48)

$$= \frac{(1 - n_F)(e^{\beta(i\omega_n - \epsilon_{\vec{p}})} - 1)}{i\omega_n - \epsilon_{\vec{p}}} \hspace{1cm} (3.49)$$

$$= \frac{(1 - n_F)(e^{-\beta\epsilon_{\vec{p}}} + 1)}{i\omega_n - \epsilon_{\vec{p}}} = \frac{1}{i\omega_n - \epsilon_{\vec{p}}}. \hspace{1cm} (3.50)$$

That is all, for the non-interacting case. The temperature information is contained in the frequencies through $\beta$.

3.3.4 Retarded and advanced Green’s functions

It can be shown, and in fact it is shown in [10], that the retarded Green’s function is obtained from the Matsubara one by analytic continuation. We may simply make the following substitution:

$$i\omega_n \rightarrow \omega + i\delta, \hspace{1cm} (3.51)$$

so that

$$G(\vec{p}, i\omega_n) = G_{ret}(\vec{p}, \omega). \hspace{1cm} (3.52)$$

The spectral function or density of states (DoS) is simply minus the imaginary part of the retarded Green’s function multiplied by 2. The advanced Green’s function can be obtained using a Kramers-Kronig relation. It can be derived that $\delta$ simply changes sign between retarded and advanced Green’s functions.
3.3.5 Interacting theory: the Dyson equation

In this subsection we will treat the case $V \neq 0$ and the Dyson equation for Matsubara Green’s functions.

We assume our Hamiltonian commutes with the particle number operator and that the non-interacting problem (the one containing $H_0$) can be solved.

Consider the following operator, which resembles the time-evolution operator but which incorporates only the perturbation part instead of the full Hamiltonian:

$$U(\tau) = e^{\tau K_0} e^{-\tau K}, \quad (3.53)$$

in which for brevity

$$K = K_0 + V = H_0 - \mu N + V. \quad (3.54)$$

We can derive and solve a differential equation for $U(\tau)$:

$$\frac{\partial U(\tau)}{\partial \tau} = e^{\tau K_0} (K_0 - K) e^{-\tau K} = -e^{\tau K_0} V e^{-\tau K}. \quad (3.55)$$

In the interaction representation we may write:

$$\frac{\partial U(\tau)}{\partial \tau} = -(e^{\tau K_0} V e^{-\tau K_0})(e^{\tau K_0} e^{-\tau K}) = -\tilde{V}(\tau) U(\tau), \quad (3.56)$$

where the tilde on $V$ signifies that it is written in the interaction representation. By repeated integrations we can solve for $U$, using $U(0) = 1$.

$$U(\tau) = 1 - \int_0^\tau d\tau_1 \tilde{V}(\tau_1) U(\tau_1) \quad (3.57)$$

$$= 1 - \int_0^\tau d\tau_1 \tilde{V}(\tau_1) + (-1)^2 \int_0^\tau d\tau_1 \int_0^{\tau_1} d\tau_2 \tilde{V}(\tau_1) \tilde{V}(\tau_2) U(\tau_2) \quad (3.58)$$

$$= \sum_{n=0}^\infty (-1)^n \int_0^\tau d\tau_1 ... \int_0^{\tau_{n-1}} \tilde{V}(\tau_1) ... \tilde{V}(\tau_n). \quad (3.59)$$

We may rewrite it as follows, introducing a time-ordering operator:

$$U(\tau) = \sum_{n=0}^\infty \frac{(-1)^n}{n!} \int_0^\tau d\tau_1 ... \int_0^{\tau_n} d\tau_n [T_\tau \tilde{V}(\tau_1) ... \tilde{V}(\tau_n)] \quad (3.60)$$

$$\equiv T_\tau \exp \left[ - \int_0^\tau d\tau_1 \tilde{V}(\tau_1) \right]. \quad (3.61)$$

This is the S-matrix expansion. We might remember it from QFT-class. The expansion is well suited for a Monte Carlo treatment as it is a large dimensional integral and this is exactly the way we will treat it later.

To continue to derive the interacting Green’s function, we define:

$$S(\tau_1, \tau_2) = T_\tau \exp \left[ - \int_{\tau_1}^{\tau_2} d\tau' \tilde{V}(\tau') \right], \quad (3.62)$$

so that

$$S(\tau) = U(\tau). \quad (3.63)$$

\footnote{Note that this $S$ is not the same as the action.}
Which can be rewritten as a trace over the non-interacting states, using the following notation:

\[ \text{Rearranging, and using that } S = \frac{\text{Tr}[e^{-\beta K_\alpha} T_\tau S(\beta) \hat{C}_{\tilde{p}_\alpha} (\tau) \hat{C}_{\tilde{p}_\alpha}^\dagger (0)]}{\text{Tr}[e^{-\beta K_\alpha} S(\beta)]}. \] (3.64)

Next, we rewrite the definition of the Green’s function (3.37) to

\[ G(\tilde{p}, \tau) = -\frac{\text{Tr}[e^{-\beta K_\alpha} T_\tau S(\beta) \hat{C}_{\tilde{p}_\alpha} (\tau) \hat{C}_{\tilde{p}_\alpha}^\dagger (0)]}{\text{Tr}[e^{-\beta K_\alpha} S(\beta)]}. \] (3.65)

Which can be rewritten as a trace over the non-interacting states, using the following notation: \( \text{Tr}[e^{-\beta K_\alpha \theta}] = 0 \) \( (\theta) \), where \( \theta \) is any operator:

\[ G(\tilde{p}, \tau) = -\frac{\langle T_\tau S(\beta) \hat{C}_{\tilde{p}_\alpha} (\tau) \hat{C}_{\tilde{p}_\alpha}^\dagger (0) \rangle}{\langle S(\beta) \rangle}. \] (3.66)

We can approximate this Green’s function by expanding the numerator in terms of the S-matrix:

\[ \langle T_\tau S(\beta) \hat{C}_{\tilde{p}_\alpha} (\tau) \hat{C}_{\tilde{p}_\alpha}^\dagger (0) \rangle = \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \int_0^\beta d\tau_1 ... \int_0^\beta d\tau_n \langle T_\tau \hat{C}_{\tilde{p}_\alpha} (\tau) \hat{V}(\tau_1) ... \hat{V}(\tau_n) \hat{C}_{\tilde{p}_\alpha}^\dagger (0) \rangle. \] (3.67)

This S-matrix expansion is usually calculated with the help of Feynman diagrams. But before we immerse ourselves into calculating this beast we will simplify matters a bit.

Feynman diagrams are made of lines representing Green’s functions and vertices representing interactions; they come in a number of shapes and forms. An important property of these diagrams is if they are connected or not. Also one can think of diagrams with no external lines: vacuum bubbles.

It can be shown (and it is in [10] but also in many other works on quantum field theory) that the disconnected diagrams we need in order to calculate the numerator of (3.66) exactly cancel against the vacuum polarization diagrams which make up the denominator. A refreshing treatment of this argument using diagrammatics, can be found in [27]. By this argument we may use the following, more practical, definition:

\[ G(\tilde{p}, \tau) = \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \int_0^\beta d\tau_1 ... \int_0^\beta d\tau_n \langle T_\tau \hat{C}_{\tilde{p}_\alpha} (\tau) \hat{V}(\tau_1) ... \hat{V}(\tau_n) \hat{C}_{\tilde{p}_\alpha}^\dagger (0) \rangle_{\text{connected}}. \] (3.68)

The above expansion also contains diagrams with internal loops, so-called self-energy diagrams. Lines with loops in them can be replaced with single lines if we call these lines dressed. The Dyson equation relates the dressed lines (interacting Green’s functions) to the bare ones.

The Dyson equation, eq. (3.62), is often proved by deriving a geometric series out of diagrammatic considerations. The result is the following:

\[ G^{-1} = G_{0}^{-1} - \Sigma. \] (3.69)

With \( \Sigma \) the self-energy, \( G_{0} \) is the non-interacting Green’s function and \( G \) is the full fledged interacting or, dressed, Green’s function.

In the sections 3.4 and 3.5 we will develop ways to approximate the self-energy. Actually, \( \Sigma \) will not be calculated directly. But we will use an equivalent quantity called the hybridisation, \( \Delta \).

### 3.4 Dynamical Mean Field Theory

#### 3.4.1 Introduction

We have already stated that Dynamical Mean Field Theory (DMFT) is a good way to describe time-dependent interactions in a wide range of materials. It is a local theory, as we shall see. Therefore,
3.4 Dynamical Mean Field Theory

Materials with properties largely attributable to non-local effects cannot be described by it. Nonetheless, it is a very useful starting point and the theory does explain several important effects [12, 25]. We will take DMFT as a basis for more detailed calculations. But to go beyond, we first need to understand DMFT as it is.

The mean field approach aims to freeze out all spatial fluctuations since we want to use a mean field instead of all the lattice sites separately in our expressions. DMFT is dynamical because correlations in (imaginary) time are taken into account.

We would like to calculate the Green’s function of some site interacting with the lattice: the lattice Green’s function. The calculation of this quantity by DMFT consists of two crucial steps:

1. **The mapping of the lattice problem to a single impurity problem.** This impurity problem is solvable because it consists of a single site with a certain action. This action contains a special part that carries the information about the lattice. This part, \( \Delta \), is called the hybridisation function and it acts as a mean field.

2. **The self-consistent calculation of the hybridisation function \( \Delta \).** The calculation of \( \Delta \) uses the lattice Green’s function as input. This Green’s function is then recalculated with the newly calculated \( \Delta \) and so on. This is continued until both quantities do not change anymore when carrying out this procedure. We say that self-consistency is reached when this is the case.

This derivation of the DMFT-equations carries the name ‘cavity method’ because it focuses on one site in particular and integrates out the rest. The contribution of the surrounding sites is gathered together in \( \Delta \), as we shall see. In this way DMFT really is a mean field theory.

### 3.4.2 Cavity method

In this subsection we treat a derivation of DMFT. We employ the following notation, using the fact that in the path integral contains only (Grassmann) numbers. A small \( c \) is the expectation value of the annihilation operator acting on a coherent state (these we have seen in subsection 3.2.2). \( c^* \) represents \( c \)'s Grassmannian conjugate.

In some cases the Hubbard model is a welcome stereotype to use in the derivation. We have encountered it in subsection 2.4.3 but we will repeat some of its properties here. The Hubbard model considers a lattice on which fermions can be created with spin up or down. Bringing two fermions on the same site costs an amount of Coulomb energy \( U \). Fermions on neighbouring sites do not interact, but there is a chance \( t_{ij} \) of the fermions hopping from site \( i \) to site \( j \). For simplicity we include only nearest-neighbour hopping, which is signified by the notation \( \langle ij \rangle \). The Hamiltonian of the Hubbard-model looks like this:

\[
H = - \sum_{\langle ij \rangle, \sigma} t_{ij} (c^*_{i\sigma} c_{j\sigma} + c^*_{j\sigma} c_{i\sigma}) + U \sum_i n_{i\uparrow} n_{i\downarrow}.
\] (3.70)

The easiest way to get from the Hamiltonian to the Green's functions we want, is by writing the partition function as a path integral over Grassmann variables in the Matsubara formalism. As we have seen in section 3.3, we can write the non-interacting Green’s function for a fermion like this:

\[
g_0(i\omega_n) = \frac{1}{i\omega_n - \epsilon_k},
\] (3.71)

where \( \epsilon_k \) is the dispersion relation of the fermion and \( \omega_n = \frac{(2n+1)\pi}{\beta} \) is the Matsubara frequency. We use a small letter \( g \) because we are describing just one particle. The subscript 0 is there because \( g \) is a non-interacting Green’s function.
For a collection of non-interacting atoms with fermions (conduction electrons) moving between them with dispersion $\epsilon_k$ and wavevector $k$ the action becomes:

$$S = \sum_{\omega_n,j} S_{at}[\omega_n,j, \omega_n,j] + \sum_{\omega_n,k} \epsilon_k c^*_\omega_n k c\omega_n k.$$  
(3.72)

$S_{at}[\epsilon_j, \epsilon^*_j]$ is the action corresponding to atom $j$. The second term represents the kinetic energy of the moving conduction electrons. We may Fourier transform either term in the above to obtain the Green’s function for this problem. The system is uncorrelated, or Gaussian, so the first term can be replaced by:

$$\sum_{\omega_n,j} \left( -g_{at}^{-1} \right)_{\omega_n,j} c^*_\omega_n j c_{\omega_n,j}.$$  
(3.73)

Now for the collection of sites we have the following Green’s function. We denote it with a capital letter $G$ because it describes a collection of sites:

$$G\omega_n k = \frac{1}{(g_{at})^{-1} - \epsilon_k}.$$  
(3.74)

$g_{at}$ is the Green’s function of the single atom, which we assume to be obtainable. We keep in mind that the Green’s functions are matrices with their rows and columns corresponding to the states between which correlations are described: $(c^*_\alpha c_\beta)$ describes correlations between states $\alpha$ and $\beta$.

We would like to find the Green’s function of a correlated lattice but we only know how to solve the uncorrelated lattice problem. What to do?

We proceed exactly as before. We solve a single-site correlated problem, in the literature this is called the impurity problem. The correlations in this case are included in the action through a yet undefined function $\Delta$. We then can suppose that the solution to this correlated single site problem is the same as the solution to the lattice problem, as long as we choose/calculate $\Delta$ in the right way.

In other words, we select one special site, the impurity, and treat it in general (obtaining its Green’s function from the action in which $\Delta$ is included). This is done by an impurity solver: a computer program that calculates the S-matrix expansion of the Green’s function. The next, crucial step for DMFT is to embed this impurity in an uncorrelated, Gaussian, lattice. This amounts to assigning the following local Green’s function to all of the other sites:

$$g_\omega = \frac{1}{i\omega_n - \epsilon_k}.$$  
(3.75)

To implement this, we rewrite the action, noting that the impurity is at $j = 0$:

$$S = S_{at}[\epsilon^*_j = 0, c_j = 0] + \sum_{j \neq 0} H[c^*_j c_j] + \sum_{j \neq 0} (\epsilon_{j0} c^*_j c_j + \epsilon_{j0} c^*_j c_{j=0}).$$  
(3.76)

The first term represents the action of the atom at $j = 0$ with all its possible energy levels. The second term contains all the interactions between the other lattice sites, not connected to $j = 0$. The last two terms represent the hopping of electrons from the rest of the lattice to the impurity at $j = 0$ and from the impurity to the rest of the lattice. We recognise the Anderson Impurity Model in the above action, see subsection 2.4.1.

Formally, we define the impurity action $S_{imp}$ as follows:

$$e^{-S_{imp}} = \int e^{-S[H(c^*_{j \neq 0} c_{j \neq 0})].}$$  
(3.77)

This can always be done when the right-hand side is a Gaussian integral of the form:

$$\int \mathcal{D}y e^{-S_{at}[x] + y \cdot \frac{1}{2} \hat{y} \times \hat{y}} = e^{-S_{at} + c \hat{a}^2}.$$  
(3.78)
We now sweep the entire lattice problem under the rug by requiring that all the information about the lattice is contained in the quantity $\Delta$, in the following way:

$$S_{\text{imp}} = S_{\text{lat}}[c_j^0c_j = 0] - \Delta c_j^0c_j = 0.$$  \hspace{1cm} (3.79)

This is what ‘integrating out all other sites’ really means. We see that by definition $\Delta$ is independent of the lattice, since the lattice is integrated out. So to calculate it we take the simplest lattice we can think of, an uncorrelated (Gaussian) one with the following Green’s function:

$$G_{\omega n} = \frac{1}{(g_{\text{at}})^{-1} - \omega_n - \varepsilon_k}.$$  \hspace{1cm} (3.80)

It seems paradoxical to summarise the contribution from the lattice into a quantity independent of the lattice. This Gaussian (uncorrelated) approximation is exactly the upshot of DMFT. In fact, this approximation amounts to taking the limit of infinite dimensions. It means that all spatial correlations are neglected.

Metzner and Vollhardt first explored this limit of infinite dimensions in 1989 [38]. They showed that it is a valid approximation because it gives a result for the correlation energy very close to that of the three-dimensional case. This situation resembles the mean field theory in classical statistical mechanics which is exact in four dimensions and forms a good starting point for the calculation of the three-dimensional Ising model. Metzner and Vollhardt’s article can be considered the birth of DMFT as it considered this infinite dimensional limit the first time and proved it was useful. Müller-Hartman in the same year elaborated the discussion of infinite dimensionalit y and proved some important relations needed for the later formulation of DMFT by Georges and Kotliar [39] and [40]. The mean-field approach is designed to freeze out spatial fluctuations, we use a mean field instead of all the lattice sites. The temporal fluctuations are still accounted for.

In this limit the non-interacting DoS of the lattice gets a particular simple form, namely a Gaussian in real space

$$D(\epsilon) = \frac{1}{\sqrt{2\pi}t} \exp\left(\frac{\epsilon^2}{2t^2}\right).$$ \hspace{1cm} (3.81)

Here $t$ is the hopping parameter that gives the probability of a conduction electron to hop to its nearest neighbour (mostly this hopping is part of the dispersion relation $\varepsilon_k$). $\epsilon$ is the energy measured from the Fermi energy. In Matsubara terms the above DoS corresponds to the Gaussian Green’s function (3.80).

We may obtain the local part of the Green’s function from a momentum-dependent lattice Green’s function by summing over all the momenta in the first Brillouin zone and dividing by the number of lattice sites:

$$G_{\omega n} = \frac{1}{N} \sum_k G_{\omega n k} = \frac{1}{N} \sum_k \frac{1}{(g_{\text{at}})^{-1} - \omega_n - \varepsilon_k}.$$ \hspace{1cm} (3.82)

We would like this expression to hold for all the sites if we equal $G_{\omega n}$ to the impurity Green’s function $g_{\text{imp}}$ obtained from the impurity action (3.79). This is called the self-consistency condition.

If we write all the terms in the impurity action eq. (3.79) in terms of Matsubara frequencies we can immediately read off the impurity Green’s function.

$$S_{\text{imp}} = \sum_{\omega_n} \left( - (g_{\text{at}})^{-1}c_{\omega n}^*c_{\omega n} + \Delta c_{\omega n}^*c_{\omega n} \right) + \Delta c_{\omega n}^*c_{\omega n}$$ \hspace{1cm} (3.83)

$$g_{\text{imp}} = \frac{1}{g_{\text{at}}^{-1} - \Delta}.$$ \hspace{1cm} (3.84)

\footnote{Simple when interactions are concerned, the geometry is unimportant for this argument.}
We now have two formulations of the impurity Green’s function, eq. (3.82) and eq. (3.84), so we can eliminate the atomic Green’s function from either of them:

\[
G_{\omega n} = \frac{1}{N} \sum_k \left( g_{\text{imp}} \right)_{\omega n k} = \frac{1}{N} \sum_k \left( g_{\text{at}} \right)_{\omega n k}^{-1} - \Delta_{\omega n} \tag{3.85}
\]

\[
G_{\omega n} = \frac{1}{N} \sum_k \left( g_{\text{at}} \right)_{\omega n k}^{-1} \frac{1}{\omega_n - \epsilon_k} \tag{3.86}
\]

\[
= \frac{1}{N} \sum_k \left( g_{\text{imp}} \right)_{\omega n k}^{-1} \frac{1}{\omega_n + \Delta_{\omega n} - \epsilon_k} \tag{3.87}
\]

Which concludes the derivation of DMFT.

We could have done roughly the same derivation using the self-energy \( \Sigma \). I have chosen to include only the one for \( \Delta \) since it is used more frequently in my calculations than \( \Sigma \). Both quantities are closely related. \( \Delta \) is computationally (more) straightforward. \( \Sigma \) has the advantage that it is conceptually more easy to understand.

Now that we have a closed form of the Green’s function, we turn our attention to how it can be obtained.

### 3.4.3 Self-consistency

The whole story of obtaining the right hybridisation and consequently the right lattice Green’s function through the impurity Green’s function hinges on self-consistency. This means we use the result from a previous iteration in the next one until it does not vary anymore between iterations. We say that the previous result is then consistent with the next one: self-consistency.

We may summarise the DMFT-method in three steps that form the so-called self-consistency loop. This loop is displayed in fig. 3.1.

1. Solve the one-site case directly: obtain the impurity Green’s function \( (g_{\text{imp}})_{\omega n} = \langle c^* c \rangle \) from the impurity action \( S_{\text{imp}} \).
2. Calculate the lattice Green’s function by eq. (3.87) using \( g_{\text{imp}} \).
3. Take a look at the difference between the lattice Green’s function and the impurity Green’s function. This difference is caused by \( \Delta \) not having a self-consistent value. Adjust \( \Delta \) according to eq. (3.88), see below.

After this step the impurity Green’s function is recalculated with the obtained \( \Delta \) after which it is used again as input for the hybridisation \( \Delta \). The loop is repeated until the final result (the one-site (impurity) Green’s function) converges: until it does not change between successive iterations. These steps form the ‘self-consistency loop’.

It can happen that some oscillation around a fixed point occurs. This can be repaired by not fully assigning a new \( \Delta \) but by mixing it with the old one, using the following linear mixing formula, where \( 0 < \xi < 1 \) is a constant:

\[
\Delta_{\text{new}} = \Delta_{\text{old}} + \xi \left( (g_{\text{imp}})_{\omega_n}^{-1} - \left( \sum_k \left( g_{\text{imp}} \right)_{\omega_n k}^{-1} \frac{1}{\Delta_{\text{old}} - \epsilon_k} \right)^{-1} \right) \tag{3.88}
\]

Self-consistency is reached if \( \Delta_{\text{old}} \rightarrow \Delta_{\text{new}} \) or equivalently, the lattice Green’s function indeed equals the impurity Green’s function. In the non-interacting case: \( U = 0 \), the loop already converges in a single step.

The DMFT method can be extended to models containing several fields by treating the Green’s function as higher dimensional matrices containing indices for the different fields as well as for the spin.
3.4 Dynamical Mean Field Theory

3.4.4 Extreme cases

Now that we have this DMFT-technique we would like to know how it relates to cases we already understand. We consider four extreme cases.

1. The interaction \( U \) in the Hubbard model, eq. (3.70) goes to zero. This is the non-interacting limit.

2. The hopping \( t \) goes to zero or equivalently \( \varepsilon_k = 0 \), so the electrons are bound to their atoms. We call this limit the 'atomic limit'.

3. The number of nearest neighbours \( z \) goes to infinity (this is equivalent to infinite dimensions).

4. We take a Bethe lattice/Cayley tree for the lattice (the self-consistency loop becomes very straightforward).

In the first case we immediately recover the free on-site Green’s function \( g_{\text{at}} \) as our full \( G(i\omega_n) \). The self-consistency loop will converge within a single iteration.

The second case can be considered as a collection of disconnected sites. Its Green’s function can be shown to have the following analytical form:

\[
G(i\omega_n)_{\text{at}} = \frac{1 - x/2}{i\omega_n + \mu} + \frac{x}{2} (i\omega_n + \mu - U),
\]

with

\[
x = \frac{e^{\beta \mu} + e^{\beta (2\mu - U)}}{1 + 2e^{\beta \mu} + e^{\beta (2\mu - U)}}.
\]

![Diagram](image-url)

**Figure 3.1**: The self-consistency loop in a DMFT-calculation. It displays the solution of the impurity problem by the impurity solver (top). The calculation of the lattice Green’s function, which should become equal to the impurity Green’s function (right). The calculation of \( \Delta \) which is fed again to the impurity solver (bottom). The left step is carried out by the impurity solver implicitly. Picture taken from [7].
In the third case, the limit of infinite coordination number $z$, there cannot be any spatial correlations and DMFT becomes exact. Therefore, the larger the coordination number of the material is, the better DMFT works as an approximation. For three dimensional materials an approximation with DMFT already leads to sensible results [11].

Although the Bethe lattice is unphysical, the fourth case has a particularly simple interaction, namely $\Delta = t^2 g_{\text{imp}}$. Therefore it is a welcome benchmark in a number of cases.

### 3.5 Dual Fermions

In this section we will very briefly sketch the ideas of Dual Fermions as an extension of DMFT. The treatment roughly follows [41], for a more elaborate discussion see [42].

#### 3.5.1 Demands of the extension

The general idea of DMFT was to focus on one site and to integrate out the interaction with all the other sites. If we use a Gaussian, uncorrelated Green’s function for the rest of the lattice, the theory becomes purely local. We can improve this treatment by retracing some of its steps. We will see that this improvement is a natural extension of DMFT.

Albeit a natural extension, we demand it to fulfill a number of conditions:

1. The basic conservation laws should hold, as in any approximation.
2. The theory should provide us with a series expansion around DMFT, at least in the non-interacting and atomic limits.
3. The series expansion around DMFT should be convergent. Moreover, we would like the first few terms already to capture most of the non-local physics.
4. The hybridisation should be chosen optimally.
5. The equations should be easy enough to perform practical calculations.

#### 3.5.2 Mimicking DMFT

Again we take the (two-dimensional) Hubbard model as a stereotype. Its lattice action is:

$$ S[c, c^*] = \sum_{\omega k \sigma} \left( \epsilon_k - \mu - i\omega \right) c_{\omega k \sigma}^* c_{\omega k \sigma} + U \sum_i \int_0^\beta n_{i\uparrow} n_{i\downarrow} d\tau. \quad (3.91) $$

In this expression we filled in the interaction term: $U(\ldots)$, while in section 3.4 we took a general form: $H[c_i \neq 0, c_i \neq 0]$

Also, like in DMFT, we consider each site $i$ as an impurity with the following action:

$$ S_{\text{imp}} = \sum_{\omega \sigma} (\Delta - \mu - i\omega) c_{i \sigma}^* c_{i \sigma} + U \int_0^\beta n_{i\uparrow} n_{i\downarrow} d\tau. \quad (3.92) $$

If we compare the lattice action (3.91) to the impurity action (3.93) we see that the former can be represented as follows:

$$ S[c, c^*] = \sum_i S_{\text{imp}}[c, c^*] + \sum_{\omega k \sigma} (\Delta_k - \epsilon_k) c_{\omega k \sigma}^* c_{\omega k \sigma}. \quad (3.93) $$

Basically this is just adding an subtracting an unspecified hybridisation $\Delta$, so this relation is still exact.
As mentioned, we would like to construct a perturbation expansion of which the DMFT result is the zeroth term. This is possible by a smart, non-linear change of variables called a Hubbard-Stratanovich transformation. Moreover, as is shown in [42] the convergence properties of the lattice Green’s function improve after this transformation.

This so-called dual transformation takes the Grassmann variables \( c, c^* \) to their dual counterparts \( f, f^* \), as follows

\[
e^{A^2\epsilon_{\omega,\sigma}c_{\omega,\sigma}c_{\omega,\sigma}^*} = B^{-2} \int e^{-BR(c_{\omega,\sigma}f_{\omega,\sigma} + f_{\omega,\sigma}^*c_{\omega,\sigma}) - B^2f_{\omega,\sigma}f_{\omega,\sigma}^*} df_{\omega,\sigma}^*df_{\omega,\sigma}.
\]  

(3.94)

This expression is valid for any complex numbers \( A \) and \( B \). We choose \( A^2 = (\Delta_\omega - \epsilon_k) \) and \( B^2 = g_{\omega}^{-2}(\Delta_\omega - \epsilon_k)^{-1} \) for each set of \( \omega, k \) and \( \sigma \).

We consider the partition function \( Z = \int e^{-S_Dc^*Dc} \), and rewrite it to

\[
Z = \prod_{\omega,k} g_{\omega}^2(\Delta_\omega - \epsilon_k) \int e^{-S[c, c^*, f, f^*]} df^*DfDc^*Dc, 
\]

(3.95)

with

\[
S[c, c^*, f, f^*] = \sum_i S_{\text{imp}}[c_i, c_i^*] + \sum_{\omega,k}\left[g_{\omega}^{-1}(f_{\omega,\sigma}c_{\omega,\sigma} + c_{\omega,\sigma}^*f_{\omega,\sigma}) + g_{\omega}^{-2}(\Delta_\omega - \epsilon_k)^{-1}f_{\omega,\sigma}^*f_{\omega,\sigma}\right].
\]

(3.96)

We now have two different expressions for the lattice action, eq. (3.93) and eq. (3.96). We can find the relation between the dual Green’s function \( G_{\text{dual}} = -(Tf^*) \) (\( T \) is the time ordering operator) and the Green’s function of the initial system \( G = -(Tc^*) \) as follows. We replace \( \epsilon_k \rightarrow \epsilon_k + \delta_{\omega k} \) and we differentiate the partition functions corresponding to the different actions with respect to \( \delta_{\omega k} \). This gives us the following relation between \( G \) and \( G_{\text{dual}} \):

\[
G_{\omega,n,k} = g_{\omega}^{-2}(\Delta_\omega - \epsilon_k)^{-2}G_{\omega,n,k}^{\text{dual}} + (\Delta_\omega - \epsilon_k)^{-1}. 
\]

(3.97)

To find an action depending only on the new variables \( f \) and \( f^* \) we first notice that it doesn’t matter whether we sum over momenta or sites: \( \sum_k (f_k^*c_k + c_k^*f_k) = \sum_i (f_i^*c_i + c_i^*f_i) \). Then we look at one given lattice site in particular and integrate over \( c_i^*, c_i \). We get:

\[
\int e^{-S_{\text{site}}Dc^*Dc} = Z_{\text{imp}}e^{-\sum_{\omega,\sigma} g_{\omega}^{-1}f_{\omega,\sigma}^*f_{\omega,\sigma} - V[f^*_i f_i]}. 
\]

(3.98)

\( Z_{\text{imp}} \) is the partition function of the original impurity problem: eq. (3.92). \( V[f^*_i f_i] \) is called the dual potential. It is a power series of which the first term equals \( \gamma_{1234}^{(4)}f_1^*f_2^*f_3^*f_4^* \). The indices represent combinations of \( \sigma \) and \( \omega \): \( f_1 \) means \( f_{\omega_1}^*c_{\sigma_1} \). \( \gamma_{1234}^{(4)} \) is the four point vertex of the local impurity problem, given by:

\[
\gamma_{1234}^{(4)} = g_{11}^{-1}g_{33}^{-1}[(c_1^*c_2^*c_3^*c_4^*) - (g_{14}g_{23} - g_{13}g_{24})]g_{22}^{-1}g_{44}^{-1}.
\]

(3.99)

The vertices \( \gamma \) were never needed in DMFT because all could be calculated in a Gaussian approximation.

We recall that the Green’s functions are matrices with their (spin-, orbital-, site- or equivalently momentum-) indices corresponding to the matrix rows and columns and their multiplication is a normal matrix multiplication.

The form of the higher order vertices is derived in appendix A.5 of [7]. It is important to note that the vertices are not momentum-dependent and thus local.

We take a moment to look at the dual self energy:

\[
\Sigma_{\omega,\sigma}^{\text{dual}} = -[(\Delta_\omega - \epsilon_k)^{-1}g_{\omega}^{-2} + g_{\omega}^{-1} + (G_{\omega,k}^{\text{dual}})^{-1}].
\]

(3.100)
3.5 Dual Fermions

To obtain a dual perturbation expansion for it we draw skeleton diagrams; in which the lines are renormalised, dual Green’s functions and the n-point vertices are $\gamma^{(n)}$. The normal rules for the construction of diagrams apply but now also higher-order vertices are present because they appear in $V_{\text{dual}}$.

It can be shown that this theory is conserving in the Baym-Kadanoff sense so that our first demand from the theory is met.

To be able to calculate real quantities of interest, we look at the hybridisation $\Delta$ for a moment. It is still arbitrary, but we can introduce a self-consistency condition for the dual Green’s function like this:

$$\sum_k G_{\text{dual}}^{\omega k} = 0.$$ (3.101)

In diagrammatic language this means that diagrams with simple closed loops, the Hartree-corrections, vanish. Other diagrams than the skeleton diagrams do not contribute to the self-energy. It also implies:

$$\sum_k g_{\omega} (\Delta_{\omega} - \epsilon_k)^{-1} = 0.$$ (3.102)

Which is exactly the same expression as the self-consistency condition for DMFT. So in the Dual Fermion formalism we have captured all the local physics already at the zeroth order of the expansion. DMFT really is the zeroth order term! The price we pay is that the interaction term contains skeleton diagrams with vertices of all seven orders.

The strength of this approach is that it uses the same self-consistent calculation procedure as DMFT: obtain an impurity Green’s function $g$ using a certain hybridisation $\Delta$ and use $g$ as input again for $\Delta$. Only the calculation of $\Delta$ itself is more involved in the Dual Fermion case.\(^4\)

3.5.3 Conclusion

By taking this highly non-linear Hubbard-Stratanovich transformation that seems weird at first sight, we may conclude that DMFT is the best local approximation: all local diagrams are taken into account. Our perturbation series for $\Sigma$, which at first was simple, now contains an infinite number of vertices. But, since the zeroth order approximation is already good because it contains all of DMFT, we hope that the correction terms are small so that summing only a few of them already produces satisfactory results.

In fact this seems to be the case in the weak-coupling, the strong coupling as well as the intermediate coupling regime, as is noted in [41]. If only the first diagram (b, as written in the same article) is included the results already improve dramatically.

The calculation procedure very much resembles the self-consistency loop from DMFT. Only an inner loop is added. The inner loop consists of the calculation of the dual diagram series for $\Sigma_{\text{dual}}$. With this $\Sigma_{\text{dual}}$ a new dual Green’s function is obtained through Dyson’s equation, this new $G_{\text{dual}}$ is again used to calculate a new $\Sigma_{\text{dual}}$.

This inner loop generally is an order of magnitude faster than the calculation of the impurity Green’s function itself because it does not involve the Monte Carlo measurement of the correlation functions but uses the previously obtained impurity Green’s function at different Matsubara frequencies.

When the dual Green’s function is obtained accurately enough, it is used to calculate a $\Delta$ that is used in the impurity solver again.

In short, we obtained a perturbation series around DMFT. We have argued (although not seen directly) that it contains most of the non-local physics already at low perturbation order. A proof of the optimality of the choice of the hybridisation is given in [42]: already at the Gaussian approximation for the Dual Green’s function, the results improve. Furthermore, the theory is conservative in the Baym-Kadanoff sense. The equations of the theory are workable so we conclude that Dual Fermions succeed in the task set out for them in terms of the demands at the beginning of this section.

\(^4\)This freedom of the choice of $\Delta$ allows for more room the experiment. The transformations of $\Delta$ form a group [42]. Exploring this notion made it possible to derive a new calculational scheme which we will see in section 5.2.
Figure 3.2: The self-consistency loop for the Dual Fermion procedure. The inner loop (in black) is used to calculate the dual self-energy or equivalently $\Delta$. The outer loop (in red) is the same as the self-consistency loop for DDMFT. The subscript $r=0$ in the dual lattice Green’s function (at the calculation of $\Delta$) means the local part is evaluated. Picture taken from [41].
Chapter 4

The study of the s-d model using CT-QMC

4.1 Introduction

As mentioned in the introduction, this thesis is a small step in the development and improvement of computational methods to study correlated systems. This development is needed because we keep finding materials that are not well explained by the existing models like Dynamical Mean Field Theory. Calculating their properties by hand instead of by computer yields less results because the interacting case can only be solved in one dimension. In materials where only momentum-independent (thus local) effects are important to describe the properties of the material DMFT has been a huge success. However, our limited understanding of the effects mentioned in section 2.3 stimulates us to look beyond a local approximation and develop non-local approaches. It is important to test such new methods and see what their features are, since we would like to know where they could be improved. To test one such method: Dual Fermions, we study a two dimensional s-d exchange model in the paramagnetic region, and compare the Dual Fermion results to their DMFT-counterparts.

It should be noted that the study of electrons on a quantummechanical level is computationally very challenging since fermions exhibit the famous sign problem [43]. The integrals that are needed to be approximated are oscillatory because by the Pauli principle: the interchange of two fermions yields a respective minus sign in their total wavefunction. Oscillatory integrals are very hard to approximate numerically. This is the reason why relatively simple models are still being considered. The computational tools to tackle them are being developed while you are reading this text.

The s-d model was proposed in 1946 by Vonsovskiy and it was introduced to explain the behaviour of localised electrons in partially filled shells coupling to conduction electrons. It contains an exchange coupling that is proportional to the spins of the two interacting particles. Such a coupling is found in nature if we consider the scattering of conduction electrons by impurities in a metal. This scattering mechanism leads to a resistance minimum at low temperatures: the Kondo effect. For this reason this type of lattice model is also referred to as the Kondo lattice model.

Two-dimensional materials are a welcome system to test non-local theories because treating these materials in a mean-field picture does not lead to satisfactory results. The reason for this is that the mean-field result is exact in dimensions larger than material under study. For DMFT the dimension for which the theory becomes exact is actually infinity. The Dual Fermion formalism, as developed in the previous chapter is a powerful tool to study non-local effects in correlated systems. We would like to know how the Dual Fermion results compare to the ones from a mean field (DMFT)-approach.

We expect that the s-d model exhibits a metal-insulator transition if the spin-spin interaction strength
(\(J\)) is varied. This transition is explicitly what we shall be looking for by examining the densities of states (DoS’s) constructed from the Green’s function. We will obtain the Green’s function using a Continuous-Time Quantum Monte Carlo, a calculational framework that will be explained in this chapter.

This work is a natural consequence of previous work done by Antipov [6] who calculated among other things the DoS’s for the Hubbard model, as opposed to the s-d model, within the Dual Fermion approach.

4.2 Definition of the system

4.2.1 Hamiltonian

To obtain the s-d exchange model we write the spin-spin-interaction in the Anderson Impurity model (as elaborated in subsection 2.4.1) explicitly. This interaction is a special case of the hybridisation (the interaction between the localised and the conduction electrons) that is already included in the Anderson model. We thus take the following interaction:

\[ H_{\text{int}} = J \vec{S} \cdot \vec{s}. \] (4.1)

Here \( \vec{S} \) denotes the spin vector of the localised electrons and \( \vec{s} \) corresponds to the spin vector of the conduction electrons. If we define the spin vectors in the usual way

\[ S_x = c_1^\dagger \sigma_x c \] (4.2)

with

\[ c^\dagger = (c_1^\dagger, c_\uparrow) \] (4.3)

we get for the spin-interaction Hamiltonian

\[ H_{\text{int}} = J(S_x s_x + S_y s_y + S_z s_z) = J(n_l \uparrow - n_l \downarrow)(n_c \uparrow - n_c \downarrow) + \frac{J}{2} c_l^\dagger c_l c_\uparrow^\dagger c_\uparrow + \frac{J}{2} c_l^\dagger c_l c_\downarrow^\dagger c_\downarrow \] (4.4)

The rest of the Hamiltonian looks as follows in equation (4.5). The first term is the hopping-term: the conduction electrons can hop between sites, their dispersion corresponds to the lattice under study, in our case a two-dimensional simple cubic lattice. The second term is called the Hubbard term: the localised electrons are dispersionless (they carry no kinetic energy) but they experience a repulsion with an energy equal to \( U \) if two of them are at the same site. This is called a Hubbard interaction. Because of this term, our case is a bit different from the original s-d model, \( U \) is included for calculational purposes as will be explained in subsection 4.4.1.

The third term is the spin-spin interaction term that we defined previously. The fourth term is the chemical potential term which allows for a variable number of particles.

\[ H = - \sum_{i,j,\sigma} t_{ij} (c_{i,j,\sigma}^\dagger c_{i,\sigma}) + \sum_i U n_{l,\uparrow} n_{l,\downarrow} + J \sum_{i,\sigma} \vec{S}_i \cdot \vec{s}_i + \mu N \] (4.5)

\( t_{ij} \) is the dispersion for the conduction electrons corresponding to the lattice involved. \( N \) is the total number of particles. In the two-dimensional simple cubic case, the dispersion is given by:

\[ t_{ij} = t(\cos(k_x x) + \cos(k_y y)) = t(\cos(\frac{2\pi i}{N_x}) + \cos(\frac{2\pi j}{N_y})) \] (4.6)
4.2.2 Parameters of the model and the calculation

It is common practice to quote dimensionless values for $U$, $J$, $\beta$ and $t$. If the first two are read in electron volts, $\beta$ is in inverse electron volts, so $\beta = 1$ corresponds to roughly 10000 K and room temperature corresponds to $\beta \approx 30$. $t$ is the probability for conduction electrons to hop from a site to its nearest neighbour, so it is indeed dimensionless. $N_x$ and $N_y$ are the number of lattice sites in the $x$- and $y$-direction, in our case $N_x = N_y = 24$ to keep the calculation tractable. Due to the difficulty of analysing such quantum systems computationally this is about as far as we can go nowadays.

In our analysis we study the effect of $J$-values between $-1$ and $1$. $U$ is taken large enough so that it does not interfere with our results for the conduction electrons: a value $U = 5$ is satisfactory. $\beta$ is taken just large enough to see an effect for varying $J$. $\beta = 10$ produces good results.

4.2.3 Expectations

The results of our model will still be comparable with other works when we look at the behaviour of the conduction electrons, which are not directly influenced by $U$. We are interested in the behaviour of the conduction electrons because they determine the state of the material: insulating or conducting.

A metal-insulator transition is expected to occur at some critical level of the spin-spin interaction strength $J = J_c$. In this work we try to find this $J_c$ and we want to know if the density of states (DoS) is different for positive and negative $J$-values. Most importantly, we investigate how these things change if we incorporate non-local corrections.

Our model does not distinguish between particle- or hole-excitations so the Hamiltonian should be particle-hole symmetric. If we perform this symmetry operation (creation operators $\rightarrow$ annihilation operators) we can evaluate the chemical potential term to be $\mu N = \frac{U}{2} N_l$. In other words, there is only a chemical potential for the localised electrons, the chemical potential of the conduction electrons equals zero.

For $J = 0$ there can be no magnetic ordering so here we have a paramagnetic state. We assume a paramagnetic state for the other $J$-values as well and study fluctuations. In principle we may impose any magnetic state we want because we consider an artificial model. The truth is, we do not know in which state a real material will be. It is very well conceivable that there exist materials in a paramagnetic state for the $J$-values we study.

As the conduction electrons are delocalised they give rise to an indirect Ruderman-Kittel-Kasuya-Yosida (RKKY)-interaction between the localised electrons. We call the interaction indirect because it is not included in the Hamiltonian but it follows from the other (Coulomb and exchange) interactions. The effect of such an interaction cannot be described in a mean field picture alone because it is inherently non-local. Naively, it is expected that such an interaction leads to long range magnetic ordering but a scaling analysis proves that the real picture is more complicated [19, 20]. We do not yet know what the effects of the RKKY-interaction are in the paramagnetic case.

4.3 Past approaches

Here we review two historical approaches to analysing the s-d model. Although these results can be improved we can still gain a crude understanding from them.

4.3.1 Quasi-classical analysis

When the s-d model is considered quasi-classically, as was done by Anokhin et al. [4], a metal-insulator transition is observed, as can be seen in fig. 4.1. Quasi-classical in this sense means that the Green's functions are calculated using the Heisenberg equation of motion: inherently quantum, but the spin $S$ of
the localised electrons is taken $S \to \infty$ with $J = 2 \cdot I \cdot S = \text{constant}$, a classical approximation. As this calculation contains a continuum of energy levels we expect it is insensitive to some quantum effects.

$$J = 2 \cdot I \cdot S = \text{constant},$$

As this calculation contains a continuum of energy levels we expect it is insensitive to some quantum effects.

In both the simplest self-consistent approximation and the Hubbard-III-approximation\footnote{referring to the third paper by Hubbard on this subject} there is a clear transition from a metal to an insulator for many lattice types as $J$ is increased. It is worth noting that the results are independent of the sign of $J$ in this approach.

### 4.3.2 Poor man’s analysis (Atomic limit)

To get a primitive picture of the DoS, we analyse our system in the atomic limit: when the hopping on the lattice is switched off ($t = 0$). In this way we can see how the DoS depends on $J$ if we would consider a lattice of independent atoms. It gives us some insight into the case $t \neq 0$. We use relation (3.120) in [10] for the DoS:

$$A(\omega) \propto \sum_{n,m} |\langle n|c_{\tau}^{\dagger}|m\rangle|^2 (e^{-\beta E_n} + e^{-\beta E_m}) \delta (\omega + E_n - E_m)$$

(4.7)

Here $A(\omega)$ is the DoS (also called spectral function) and $\omega$ is the energy. $\omega = 0$ corresponds to the Fermi level. $E_n$ is the energy of state $n$. We write down all the eigenstates $|\text{localised}\rangle|\text{conducting}\rangle$ of the Hamiltonian with $t = 0$ and their corresponding energy values. Since we are only interested in the DoS (spectral function) for the conduction electrons we use the corresponding annihilation operator in eq. (4.7). Since our (paramagnetic) case is spin symmetric it will not matter if we take spin up or down. There is a maximum of two electrons (spin up and down, according to the Pauli principle) of each kind, localised or conduction. The first ket corresponds to the localised electrons, the second ket to the conducting ones.

Filling in eq. (4.7) with all the possible states from table 4.1 we get:

$$A(\omega) \propto 6 \delta (\omega) + e^{\frac{\beta J}{2}} (1 + e^{\frac{\beta J}{2}}) \delta (\omega + \frac{J}{2}) + \delta (\omega - \frac{J}{2}) + (1 + e^{\frac{\beta J}{4}}) \delta (\omega + \frac{J}{4}) + \delta (\omega - \frac{J}{4})$$

(4.8)

When we apply the above formula to our problem: $U = 5$, $\beta = 10$, we obtain five Dirac-delta peaks. Namely at energies $\pm \frac{J}{2}$, $\pm \frac{J}{4}$ and 0 (the Fermi energy). The terms that yield Dirac-delta peaks at $\omega = |J/2|$
### 4.3 Past approaches

Possible states: $|\text{localised}\rangle |\text{conducting}\rangle$

<table>
<thead>
<tr>
<th>State</th>
<th>Energy value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$</td>
<td>0\rangle</td>
</tr>
<tr>
<td>$\frac{1}{\sqrt{2}}(</td>
<td>\uparrow\rangle</td>
</tr>
<tr>
<td>$</td>
<td>\uparrow\rangle</td>
</tr>
<tr>
<td>$</td>
<td>\uparrow\rangle</td>
</tr>
<tr>
<td>$</td>
<td>\uparrow\rangle</td>
</tr>
</tbody>
</table>

Table 4.1: Eigenstates and their corresponding eigenvalues of the Hamiltonian corresponding to our problem eq. (4.5) but without the interaction with the lattice ($t = 0$).

and $\omega = |J/4|$ are ten (!) orders of magnitude stronger than the peak at the Fermi level ($\omega = 0$), so we may neglect that last one. Approximately we have:

$$A(\omega) \propto (1 + e^{\beta J/4})(\delta(\omega + J/4) + \delta(\omega - J/4)) + (1 + e^{\beta J/2})(\delta(\omega + J/2) + \delta(\omega - J/2)). \quad (4.9)$$

The maximum mutual difference in strength of the $\omega \neq 0$ peaks is only a few percent for $J > 0$ but for $J < 0$ the maximum difference is a factor of about eleven. Therefore we might expect our interacting results to be a bit different for negative and positive $J$-values. In figures 4.2 and 4.3 one can see the relative difference in strength of the delta peaks.

![Atomic limit DoS in terms of δ-peak strengths for J=1](image-url)

Figure 4.2: The relative strength of the Dirac-delta peaks for $J = 1$ in the atomic limit.
4.4 Method

In this section we discuss how we go about the calculation of properties of the s-d model. We treat the Monte Carlo procedure itself, the implementation of the details of our model into it, the details of the calculation procedure and its limitations, the tests we performed to ensure the procedure was working and we conclude with how to obtain a DoS from Monte Carlo data.

The aforementioned quasi-classical approach clearly cannot capture quantum effects and the poorman’s approach has no notion of the lattice or the dimensionality of the problem. Therefore, we would like to extend go beyond the previous approaches. How do we do this?

As mentioned before we are dealing with a fermionic quantum system. Calculating it accurately with a full quantum-mechanical method is much harder than classical calculations. Consequently we have to settle for a small sample size: 24 x 24 atoms in our case. One such quantum-mechanical method is Quantum Monte Carlo.

There are alternatives such as Exact Diagonalisation but although very promising they are still in their initial stages. These methods were previously thought impossible but we now have access to larger computer power and memory sizes so the inversion of large (sparse) matrices can be done. Although Exact Diagonalisation produces less noise than Quantum Monte Carlo, at this point Exact Diagonalisation is not yet compatible with non-local calculations within the Dual Fermion formalism.

As we would like to know how non-local methods change the previously obtained results we stick to Quantum Monte Carlo for now.

Figure 4.3: The relative strength of the Dirac-delta peaks for $J = -1$ in the atomic limit.
4.4 Method

4.4.1 Continuous-Time Quantum Monte Carlo

In this subsection I will sketch the ideas of Continuous-Time Quantum Monte Carlo (ct-qmc). A thorough treatment of Quantum Monte Carlo in general and Continuous-Time Quantum Monte Carlo in particular can be found in the works of Gull and Hafermann [44, 7].

We recall the definition of the S-matrix expansion of the Green's function eq. (3.67). The interaction in our case is given by factors containing both $U$ and $J$ and fermionic operators. A quantum interaction containing these fermionic operators suffers from the famous sign problem [43] that can make computations exponentially hard. The sign problem is caused by the Pauli principle: interchanging two fermionic fields yields a minus-sign in the statistical weight introducing exponentially large errors and therefore exponentially large calculation times.

Hirsch and Fye circumvented the sign problem in a Monte Carlo scheme by decoupling the fermionic degrees of freedom using a time-discretisation which introduces auxiliary fields. If there is only one (auxiliary) field per time step there is nothing to interchange so also no sign problem. Their method became known as Hirsh-Fye Quantum Monte Carlo. This approach works well for small samples, as only a small number of auxiliary fields is needed, but for larger samples the computation times become a serious burden. Furthermore, the time-discretisation itself introduces a systematic error.

An approach that does not discretise $U$ and $J$ is not severely hampered by the sign problem was developed by Rubtsov et. al. [45]. It relies on introducing a tractable scalar quantity $\alpha$ in the interaction part of the action. It is a basically a shift for every pair of (equal spin) creation and annihilation operators of the form $c_\uparrow^\dagger c_\uparrow \rightarrow (c_\uparrow^\dagger c_\uparrow - \alpha \uparrow)$. The effect is a decoupling of the fermionic operators for the correct choice of $\alpha$. Mostly, a value of $\alpha \gtrsim 1$ produces good results. This approach, introducing $\alpha$-parameters, is generally referred to as Continuous-Time Quantum Monte Carlo (ct-qmc), because it circumvents time-discretisation. It is this approach that we will use for all the calculations. It should be noted that the choice of $\alpha$ is more difficult for spin-ip terms such as $c_\uparrow^\dagger c_\downarrow$. There we also need a more general $\alpha$, carrying two spin-indices, like $\alpha_{\uparrow\downarrow}$. The Monte Carlo procedure produces more noise in this case and needs more steps to reach convergence.

In practice, the S-matrix expansion and the Matsubara Green's function are calculated in ct-qmc using a solver, a calculational package that can take a Hamiltonian together with its parameters as input. This solver was also developed by Rubtsov et al. in C++. Rubtsov's code together with some documentation can be found under http://code.google.com/p/ct-qmc/.

This program uses the exact same interaction expansion as the definition of the Green's function, eq. (3.68) and is therefore often referred to as a ct-int program. Later, programs were developed that expand in the hybridisation (ct-hyb, also referred to as 'strong coupling expansion') or in an auxiliary field (ct-aux). This ct-aux method mimics the procedure developed by Hirsch and Fye but also avoids the time-discretisation. These hybridisation- and auxiliary field expansions have the advantage that low temperatures and/or high interaction strengths can be reached. Gull [44] treats the features of these different expansions in detail and compares them with each other.

4.4.2 Compatibility remarks

Expanding in the interaction (terms with $U$, $J$ and fermionic operators) is costly or even impossible for low temperatures and large coupling constants $U$ and $J$. Unfortunately, it is still the only option if we want to consider non-local correlations by Dual Fermions: there are no Dual Fermion packages available yet for other solvers. The Dual Fermion package we used was written by Hafermann and can be acquired on request.

Formally we cannot not take the Schrieffer-Wolff limit $U \rightarrow \infty$ because we need the interaction expansion for the Dual Fermion package to work. However, as already argued, the results we obtain will be comparable to earlier works because we focus purely on the behaviour of the conduction electrons that are not directly influenced by this finite $U$. If we observe a qualitatively different situation for varying $J$,
4.4 Method

it means that the influence of a finite \( U \) is small enough.

4.4.3 Implementation of the Hamiltonian

The solver we used is compatible with various Hamiltonians in terms of creation- and annihilation operators. If the Hamiltonian contains spin flip terms (which it does in our case) the localised and conduction electrons are represented by different orbitals for symmetry reasons. Defining the Hamiltonian is done by specifying all the different terms in the main source file \texttt{ct-qmc.cpp}. For reference the implementation of the Hamiltonian into this specific file can be found under \url{https://sites.google.com/site/jonassweepthesis/}. This is how the implementation is done:

- ‘r1’ represents the left two operators and ‘r2’ the right two in a term in the Hamiltonian.
- An underscore ‘_’ signifies that the operator is an annihilation-operator.
- The letter behind the ‘.’ stands for an operator index as specified below.

Any combination of operator indices can be formed as follows:

- \( r.t \) represents the time at which the operator works, in our case this was always equal to \( \tau \) since we didn’t have time-dependent interactions.
- \( r.z \) stands for the orbital or zone number: 0 for conduction, 1 for localised.
- \( r.i \) represents the spin of the operator: 0 for spin up, 1 for spin down. Our Hamiltonian is spin-symmetric.

As an example, the term \( c_{\downarrow}^\dagger(\tau)c_{\uparrow}(\tau)c_{\downarrow}^\dagger(\tau)c_{\uparrow}(\tau)c_{\downarrow}^\dagger(\tau)c_{\uparrow}(\tau)c_{\downarrow}(\tau)c_{\uparrow}(\tau) \) is represented by:

- \( r1.t=r1._t=r2.t=r2._t=\tau \),
- \( r1.z=r1._z=1 r2.z=r2._z=0 \),
- \( r1.i=1 r1._i=0 r2.i=0 r2._i=1 \).

The indirect \textit{RKKY}-interaction was implemented through the calculation of \( \gamma^{(4)} \). The vertex \( \gamma^{(4)} \) was namely taken non-zero only if it coupled with the conduction electrons. The higher order vertices were not calculated because including only the first diagram already captures a large part of the non-local physics, as argued before.

4.4.4 Implementation of the self-consistency loop

To obtain a lattice Green’s function the solver is responsible for the first part of the self-consistency loop as sketched in sections 3.4 and 3.6 in figures 3.1 and 3.2. The solver calculates the impurity Green’s function from the impurity action as defined in eq. (3.79). Recall that the interaction with the lattice is incorporated via the hybridisation \( \Delta \). If no \( \Delta \) is provided the solver assumes \( \Delta \) equals zero.

All the input parameters (\( U, J \), the amount of Monte Carlo steps, the mixing of the self-consistency loop (as defined in eq. (3.88) and the \( \alpha \)-parameter that is used to circumvent the sign problem) are stored in the file \_input.dat which is read by the solver before the calculation starts. Because our Hamiltonian contained spin-flip terms the flag \texttt{Use_Global_moves} should be switched off in \_input.dat. Also the value \texttt{Alpha_off_diagonal} should take a value \( 0.1 < \alpha_\sigma^\gamma < 1 \). What this means in Monte Carlo terms is described in \[15\]. In our case this increased the amount of noise, but only in the real part of the Matsubara Green’s function, which is not problematic as we consider a paramagnetic (disordered) case where the real part of the Matsubara Green’s function is zero anyway. An example of \_input.dat is included under
4.4 Method

https://sites.google.com/site/jonassweepthesis/. It contains all the parameters of a typical DMFT-calculation. For a Dual Fermion calculation both the calculate_Gamma4 and the DMFT_sigma flags should be changed.

After the impurity Green’s function is calculated, the interaction with the lattice is incorporated by calculating $\Delta$. The calculational difference between the DMFT-approach and the Dual Fermion-approach manifests itself in the different $\Delta$’s in DMFT and Dual Fermions. In DMFT, $\Delta$ is calculated by assuming all the sites to be uncorrelated, Gaussian. Within Dual Fermions, the interaction is included through the vertex $\gamma^{(4)}$ in the dual potential. These steps are carried out by Hafermann’s package, they correspond to the inner loop of figure 3.2.

With the new $\Delta$, a new impurity Green’s function is calculated, completing the self-consistency loop.

4.4.5 Calculation procedure

Calculations were run on a dual-core desktop-type machine. The final accurate calculations took about two weeks per $J$-value in the DMFT case and about 10% more in the Dual Fermion case. The amount of self-consistency loops was roughly 10-20 while the number of Monte Carlo steps was roughly doubled with each loop in the last 6 loops until it reached $8 \times 10^8$. The exact definition of a Monte Carlo step is different than in the standard Metropolis algorithm that is often used, because of the different decomposition scheme of the fermions. Details can be found in [45]. Self-consistency or convergence is said to be reached if the results do not change anymore between loops. In practice, this was hard to judge as some noise remained (likely because of the spin-flip terms in the Hamiltonian). Convergence was analysed using a convergence plot, comparing the results of successive loops with the naked eye. As we are looking for qualitative differences in the DoS’s and quote approximate values for the transition, this approach seems fair.

In the Dual Fermion calculations only the first relevant diagram (b) was included in the perturbation series. The small loop in figure 3.2 was then run multiple times. In effect some higher order diagrams are then taken into account. This is called a ladder approximation. Details of the ladder Dual Fermion calculation can be found [7] as well as a proof that such an approximation describes the entire Dual Fermion series reasonably well in a large number of cases.

The reader finds the run script used to carry out several self-consistency loops in a series of calculations also on the aforementioned website. In this way any calculation can be repeated, if need be. The parameter file, _input.dat, contains all relevant calculational parameters, they are changed in the script via the ‘change’ program, also delivered with the code. The calculational parameters are taken from an exhaustive list that includes a brief description of each parameter delivered with Rubtsov’s code, called input.dat.

4.4.6 Multi-processor support

At the moment of writing, the impurity solver that we used only supports multi-processor calculation of the impurity Green’s function through the message passing interface (MPI). The susceptibility and thus the vertex $\gamma^{(4)}$ cannot yet be calculated using MPI. It would have been worth the effort to introduce MPI-support for these steps within this master’s project because the Monte Carlo error decreases as the square root of the amount of cores within our solver: $MC_{error} \propto 1/\sqrt{N_{cores}}$. We deemed it unnecessary and postponed it every step of the way. Starting it at an early stage would have decreased our calculation times considerably.

4.4.7 Testing

To test the results from CT-QMC the following means were used:
4.4 Method

1. The results for the Green’s function on the Matsubara axis were compared to a Green’s function obtained by exact diagonalisation in the atomic limit. The exact diagonalisation was performed using the Pomerol-program written by Krivenko and Antipov and by using Mathematica 8. Pomerol including documentation can be obtained from http://code.google.com/p/pomerol/.

2. The non-interacting results \((J = 0)\) were compared to an analytically obtained lattice Green function to check if the ct-qmc procedure was working. The analytical Green’s function is defined in equation (3.86), with \(g_{\text{at}}\) Gaussian, like in (3.75).

3. The error originating from the ct-qmc calculation itself in the interacting case was estimated by comparing two successive iterations that were considered to be converged.

4. The error from the Dual Fermion-procedure was estimated by comparing local results (at \(J = 0\)) from the Dual Fermion-procedure with the local ones obtained using dmft.

5. The solver itself relies on the distribution of the Monte Carlo data to compute the accuracy and to determine which perturbation order is needed for each sampling.

The outcome of these tests ensured us that the results from ct-qmc and the Dual Fermion calculation could be trusted after 10-20 self-consistency loops, depending on the value of \(J\).

4.4.8 From Green’s function to density of states

As we would like to know if and where a metal-insulator transition takes place in the s-d model, we need to examine the DoS. If there are states at the Fermi level the material is conducting, if there are no states, it is an insulator. In theory, it is straightforward to obtain the DoS from the Green’s function that has been calculated. The calculated (retarded) Green’s function and the DoS differ only a factor of \(4\pi\) as is shown in by Landau in [46].

However, mostly we do not have a functional expression of the retarded Green’s function. We have to make do with Monte Carlo data: the values of the Matsubara Green’s function the at first \(=100\) discrete Matsubara frequencies. There are a few analytical methods, such as Padé approximants and Kubo formulas. But, just like in Landau’s case where we need the functional form of the Green’s function, the Padé approximants and Kubo formulas do not work with Monte Carlo data.

Fortunately a different approach has been developed by Jarell and Gubernatis [47] that goes under the name of ‘Maximum Entropy method’ or simply ‘Maxent’. Maxent tackles the following problem:

To relate the imaginary Green’s function to the real frequency spectrum \(A(\omega)\) we need to invert the following expression:

\[
G^\pm(\tau) = \int d\omega A(\omega)e^{-\tau\omega} \left( \frac{1 \pm e^{-\beta\omega}}{1 \pm e^{-\beta\omega}} \right). \tag{4.10}
\]

To do this, we need a linear operator \(\hat{F}\) with \(\hat{F}A(\omega) = A(i\omega)\) that is yet unknown. Because this inverse is not uniquely defined, we need an extra constraint. Maximising the following entropy \(S\) while changing the operator \(\hat{F}\) is such a constraint.

\[
S = ||\hat{F}A(\omega) - A(i\omega)||^2 - \int d\omega A(\omega)\ln(A(\omega)) \tag{4.11}
\]

This procedure has been put into a practical program by Sandvik [48]. For more than a decade the Maximum Entropy method has been used to analytically continue imaginary time results to the real axis. While the method is easy to use it has three major disadvantages.

\(^2\)At finite temperatures, this argument is not so strict.
4.5 Results

Firstly, Maxent introduces noise. While the qmc calculation and the Dual Fermion procedure are numerically exact, after the Maxent procedure we cannot be entirely sure whether the features of the curves are really there or that they are artefacts of the calculation.

Secondly, the Maxent-procedure itself is not transparent. The Laplace-transform, eq. (4.10), is exponentially sensitive to errors in the Monte Carlo data. Therefore we do not exactly know how our data gets transformed and with that the Maxent procedure becomes similar to a black box.

Thirdly, since no real alternatives exist it is impossible to compare the Maxent procedure to other ways of obtaining the DoS. We have no direct way of checking its validity other than feeding the procedure with known results and our physical intuition. A partial alternative to Maxent was formulated by Krivenko [49]. Unfortunately, it can only treat conducting states. On a brighter note: the error is far more tractable in Krivenko’s approach. Furthermore, Krivenko demonstrated a clear quantitative difference between the results of his calculation and the ones from Maxent. Either Maxent’s days are numbered or we might circumvent the analytic continuation altogether in the future.

4.5 Results

In this section the results of our ct-qmc-calculations are presented. We summarise the parameter regime of our calculation in the following box. In other works parameters are often given in different formats. They are divided by the hopping, or the bandwidth and sometimes the rescaled temperature \( T = 1/\beta \) is used instead.

\[
\begin{align*}
\beta &= 10 \\
T &= 0.1 \\
\text{Paramagnetic state} & \quad U = 5 \\
2D \text{simple cubic lattice} & \quad 24 \times 24 \text{ lattice sites} \\
\mu \text{ corresponds to half filling} & \quad U/t = 20 \\
J & \in [-0.7, 0.7] \\
J/t & \in [-2.8, 2.8]
\end{align*}
\]

Table 4.2: Parameter regime of our calculation.

We immediately state the three main findings of this study:

1. A metal-insulator takes place at some critical positive value of the coupling constant \( J = J_c \). This transition looks very Hubbard-like, although the interaction in our case is between spins and not of the Coulomb type.

2. The value \( J_c \) for which a metal-insulator takes place is different in different calculations. If non-local corrections are taken into account by a Dual Fermion calculation, the critical value \( J_c \) is much smaller than without these corrections.

3. The DoS’s are qualitatively different for positive (anti-ferromagnetic coupling) and negative (ferromagnetic coupling) \( J \)-values. This is contrary to the quasi-classical case found by the Ekaterinburg group in 1991 [4].

Now we will show how we arrived at these statements.

4.5.1 Raw data

As a warm up, to get a feeling for what the raw data from a ct-qmc-calculation looks like, we show the imaginary part of several calculated Matsubara Green functions for \( U=5 \), \( \beta=10 \) and varying \( J \) in figure 4.4. We also include an analytically obtained Green’s function on the Matsubara frequencies in the non-interacting case (\( J = 0 \)), from eq. (3.86).
4.5 Results

Matsubara Green’s functions for $U = 5$, $\beta = 10$, for various $J$ values

Figure 4.4: The imaginary part of several Green’s functions for the first seven Matsubara frequencies. The difference between the DMFT and Dual Fermion curves is clearly visible. The difference between positive and negative $J$ is more pronounced for larger absolute $J$-values. Note that an analytically obtained Green’s function on the Matsubara frequencies for $J = 0$, from eq. (3.86), is also plotted.

4.5.2 Critical $J$ for metal-insulator transition in different approaches

When we are studying the metal-insulator transition in the s-d model the following question comes to mind: at what (positive) value of $J$ does the metal-insulator transition occur in the model we studied?

Different approaches provide different answers to this question. The simplest quasi-classical self-consistent approximation yields $J_c = 0.816$ for the simple cubic lattice. An improved classical approximation that takes into account scattering corrections gives a value of about $J_c = 0.8$. Both of these results are from [3].

Let us compare these classical results to the results we obtained using ct-QMC. For this we need to analytically continue our raw data to the real axis. For this we use the ‘Maxent’-procedure described in subsection 4.4.8. Because the temperature in our calculations is quite high ($\beta = 10$) we expect that the transition will not be as pronounced as in the classical case. Thermal excitations might cause a DoS that appears to be conducting on paper to be insulating in real life, or vice versa. For this reason we will consider the state to be conducting if there is a local maximum at the Fermi level and insulating if there is a local minimum at the Fermi level.

Alternatively, we could define the phase to be insulating if the DoS at the Fermi level equals zero and conducting otherwise. We also could take a critical value for the energy gap equal to the magnitude of the thermal excitations to determine the transition. These different assumptions change the exact value of $J$ at which the transition takes place but the qualitative behaviour remains the same: $J_c$ is smaller than the Hubbard-III approximation.
4.5 Results

in a Dual Fermions calculation than within DMFT and there is a pronounced difference between positive and negative $J$.

Figure 4.5: The DoS around the critical values of $J$. The stars correspond to DMFT results, the lines are Dual Fermion calculations. We see the metal-insulator taking place as the local maximum (peak) at the Fermi level ($E = 0$) disappears for higher $J$-values. The values are chosen such that they are just around the transition for the two calculational schemes, DMFT and Dual Fermions. For DMFT the value $J_c \approx 0.65$, and for the Dual Fermion calculation it gets as low as $J_c \approx 0.3$.

On figure 4.5 we see the DoS’s just around the metal-insulator transition for the DMFT and Dual Fermion methods. The DMFT results are represented by stars and the Dual Fermion results by lines. We see the metal-insulator taking place in the DMFT-data just around $J \approx 0.65$. The purple stars represent the DoS for $J = 0.6$ with a small peak at the Fermi level, $E = 0$. This state will likely be conducting at this temperature. The pink stars correspond to the DoS for $J = 0.7$ that has a dip at the Fermi level: likely an insulating state at this temperature. The red line corresponds to the Dual Fermion data, just before the metal insulator transition: this state will likely be conducting. The green line is Dual Fermion data just after the transition: a clear dip, so likely an insulator at this temperature.

Away from the Fermi level, at $E \neq 0$, we see the shoulders of the curve. It is unclear if their exact shape, size and location tell us anything meaningful or if they are features of the calculation.

The value of $J$ where the metal-insulator transition takes place is significantly lower in our approaches than in a quasi-classical calculation. As our approximations improve, the critical value of $J$ decreases. The fact that the value of $J$ changes means that quantum effects and non-local correlations are significant. The fact that it decreases points to the improvement of the approximation as it is already sensitive to a smaller value of $J$.

The Dual Fermion calculation only incorporates non-local fermionic excitations. Including also bosonic excitations, for instance collective modes such as magnons, will likely improve the results. An
attempt has been made by Katanin et al. to include non-local correlations analytically but only on a
phenomenological level using a Bethe lattice. This makes it hard to compare their results with the
non-local ones we obtained. Katanin et al. acknowledge the need for a quantum calculation.

In table 4.3 we summarise the results for different calculational methods.

<table>
<thead>
<tr>
<th>Method</th>
<th>( J_c )</th>
<th>Features</th>
</tr>
</thead>
<tbody>
<tr>
<td>Simple self-consistent</td>
<td>0.816</td>
<td>Solution of Heisenberg equation of motion, classical limit: ( S \to \infty )</td>
</tr>
<tr>
<td>Hubbard-III</td>
<td>( \approx 0.8 )</td>
<td>Inclusion of scattering corrections</td>
</tr>
<tr>
<td>DMFT</td>
<td>( \approx 0.65 )</td>
<td>Quantum calculation, local correlations</td>
</tr>
<tr>
<td>Dual Fermions</td>
<td>( \approx 0.3 )</td>
<td>Quantum calculation, fermionic non-local corrections</td>
</tr>
</tbody>
</table>

Table 4.3: Critical values of \( J \) for the metal-insulator transition in different approaches.

4.5.3 Difference between anti-ferromagnetic and ferromagnetic couplings

Up to now, we have only looked at the behaviour of the s-d model with a positive \( J \). This corresponds
to anti-ferromagnetic coupling, although the state we consider is still paramagnetic. We can also look
at ferromagnetic coupling (\( J < 0 \)). Classical theory predicts the DoS’s to be the same for positive and
negative \( J \) because \( J \) only occurs to the second power in the expressions. In the atomic limit we
do have different results for positive and negative \( J \). In what way the difference will play out in the
quantum/interacting case is hard to foresee. In figure 4.6 we display the results for \( |J| = 0.4 \) calculated.

Figure 4.6: DoS’s for \( |J| = 0.4 \). There is a difference for positive and negative \( J \). For the local theory,
DMFT (stars), the difference is already visible but beyond this approximation, in the Dual Fermion
approach (lines) it is more pronounced.
within both dmft and Dual Fermions. The colours represent positive and negative \( J \)-values. Stars correspond once again to the dmft-calculation while the Dual Fermion result is drawn with lines. In this picture we look at the general shape of the curves. We see that they are not the same for positive and negative \( J \) and that incorporating non-local corrections through Dual Fermions changes the results: the difference is more pronounced in the Dual Fermion case.

![DoS's for \(|J|=0.7\)](image)

Figure 4.7: DoS’s for \(|J|=0.7\). In this regime the difference between positive and negative \( J \) is clearly visible in both the local (dmft, stars) and the non-local (Dual Fermion, lines) approaches.

At larger absolute \( J \)-values, displayed in figure 4.7 we clearly see the difference between positive and negative \( J \). Again, we should compare lines with lines (Dual Fermions) and stars with stars (dmft). Furthermore, we see once again that the dmft and Dual Fermion calculations are very different for larger absolute values of \( J \). In dmft there is no indication for a metal-insulator transition at any negative \( J \)-value, because the results do not seem to be changing if \( J \) is changed. Further calculations with larger absolute \( J \) should be carried out to prove this. It is unknown if the shoulders in the Dual Fermion DoS for \( J = -0.7 \) have any physical meaning.

As can be seen in 4.6 and 4.7 the Dual Fermion approach is more sensitive to the difference in the sign of \( J \). Moreover, the difference is more pronounced for a larger absolute value of \( J \).
4.6 Discussion of the results

In this section we discuss the meaning of the above results, along with possible consequences. We compare our results with what we expected at the beginning of this study and with earlier works. We also discuss how and where our study could be improved.

4.6.1 Comparison with expectations

As we have stated in the previous section, we actually do see what we expected: a metal-insulator transition at some critical positive $J$. Furthermore, the DoS’s change with respect to the sign of $J$. This is very different from the quasi-classical analysis performed by Anokhin et al. [4].

Improving the description by including non-local correlations through the Dual Fermions causes the critical value of $J$ to drop. It also causes a greater difference between the results for positive and negative $J$.

Because some curves were obtained close to a phase transition, the convergence of the results took a large number of Monte Carlo steps. The high temperature argument, stating that it is hard to say if a state is conducting or insulating at the temperature $\beta = 10$, also makes it harder to quote an exact critical value $J_c$ for the metal-insulator transition. Furthermore, the spin-flip terms in the Hamiltonian introduce an amount of noise that frustrates the convergence to some extent. Having said that, it is clear that the positive critical value of $J$ drops as our calculation improves from quasi-classical to Dynamical Mean Field Theory to Dual Fermions and that the s-d model can be treated within this CT-INT approach, at least qualitatively.

4.6.2 Comparison with other works

How do our results compare to those of other workers in the area?

The s-d model was first analysed using a qmc algorithm in 2001 by Capponi and Assaad [3]. They used an $8 \times 8$ lattice and retained spatial correlations. Their result for $\beta t = 3$ and $J/t = 0.8$ (the second curve from below in fig. 4.8) resembles the red curve in fig. 4.5. The locations of the shoulders of both peaks coincide if we take into account that the horizontal axis in Capponi and Assaad is rescaled with the hopping $t$.

In their work, anti-ferromagnetic fluctuations are strictly destroyed at $T > 0$ for positive $J$-values because all the spatial correlations are retained in the calculation. This would account for their low critical value of the coupling constant $J$ for the metal insulator transition. In our Dual Fermion calculation the anti-ferromagnetic fluctuations are also destroyed, at least partially. Although the lattice size of $8 \times 8$ that Capponi and Assaad use seems small, an increase to $24 \times 24$ in our calculation does not cause much difference to the results. Whereas Gull states that a continuous-time algorithm is orders of magnitude more efficient and therefore can treat larger sample sizes [44], earlier obtained results on smaller samples seem robust with respect to the size in this particular case.

We chose to calculate a $24 \times 24$ lattice because we wanted to approach a real system while keeping the calculation time manageable. Until now, we did not know that the results change so little with respect to the lattice size. We cannot be certain if this is the whole story because important effects might manifest themselves at even larger sample sizes. All this does however invite the question on what length scale the Dual Fermion formalism describes correlations. Furthermore, we should check what is the exact dependence on the sample size. Maybe the $8 \times 8$-results and the $24 \times 24$-results nearly coincide only by coincidence.

Otsuki et al. analysed the s-d model away from half filling within DMFT. Their density of conduction electrons per site $n_c = 0.9$ whereas we treated the half-filled case: $n_c = 1$. It is quite remarkable that they only needed $10^7$ Monte Carlo steps to reach accurate results within 10 iterations while also using the interaction expansion. We needed 10 times as many steps for reasonable data. A multi-processor
4.6 Discussion of the results

![Figure 4.8: Results for the DoS from Capponi and Assaad [3] on an 8 × 8 square lattice for $J/t = 0.8$. The horizontal axis is rescaled with the hopping $t$.](image)

calculation using our solver could be a step in the right direction. This does require building in multi-processor support for the calculation of the vertex $\gamma^{(4)}$ which is required for the Dual Fermion calculation.

Furthermore, the article of Otsuki et al. claims some interesting physics to take place at the Kondo temperature $T_K \approx 0.1 - 0.2$. Namely, the localised electrons are claimed to contribute to the Fermi volume. In the literature this is known as a Large Fermi Surface (LFS). Debate is still going on if the localised electrons really contribute to the Fermi volume. Next to this LFS problem, the charge susceptibility has a maximum at $T_K$ in both the one-dimensional case and in two dimensions within DMFT. $T_K$ is defined as

$$T_K = \sqrt{g}e^{-1/g}$$

and

$$g = 2JD^{-1}\sqrt{\frac{2}{\pi}},$$

in which $D$ is the bandwidth ($D = 1$ in our case). The famous Kondo resonance can be observed at temperatures much lower than $T_K$. It would be worthwhile to study the LFS- and the susceptibility maximum effects within our approach as well.

In 2006, Werner and Millis developed a strong coupling (hybridisation) expansion impurity solver [2]. In the same paper they analysed the s-d model at very low temperatures ($\beta \in [80, 800]$). The Kondo resonance is clearly visible in their study. However, their approach also limits itself to DMFT. Studying the s-d model at these temperatures including non-local correlations seems a prudent next step. Werner and Millis observe a metal-insulator transition for negative $J$ and they note that the absolute value of $J$ where the transition takes place is much larger than in a classical approach. $-8 < J_c/t < -6$ in the quantum case as opposed to $J/t = -4$ in the classical case. This is consistent with our findings because our absolute $J$-values were smaller so we did not yet observe the transition. Calculations for ferromagnetic (negative) $J$-values smaller than roughly $-0.7$ were too lengthy for this work.
4.6 Discussion of the results

Capponi and Assaad [3] and Werner and Millis [2] also analysed ordered states of the s-d model. As there are transitions between ordered and disordered states in the s-d model, extending our description to include those or even the transition between these states is another direction for future work in this area.

On a final note, we should remark again that the Maxent-procedure on which we and the other workers rely to obtain DoS’s from Monte Carlo data is far from perfect. For instance, we do not know the scientific value of (some of) the observed shoulders in the DoS’s. Compare in fig. 4.6 for instance the \( J = 0 \) curve (obtained with Maxent from an analytical calculation) which has shoulders to the DMFT-curve for \( J = 0.4 \), where the shoulders are absent. We would expect the onset of two distinct peaks to be visible for \( J = 0.4 \) in DMFT, as the transition to an insulator occurs gradually with increasing \( J \).
Chapter 5

Conclusions

5.1 Summary

We started this work wanting to study the s-d model at different interaction strengths within the frameworks of DMFT and Dual Fermions. We set out to obtain densities of states (DoS's) and to use these to analyse the metal-insulator transition that the s-d model exhibits.

Several pages later, we succeeded. We found the following positive critical values of the coupling constant for the metal-insulator transition in the s-d model on a two-dimensional simple cubic lattice in the paramagnetic state:

<table>
<thead>
<tr>
<th>Calculational Framework</th>
<th>$J_c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heisenberg equation of motion (quasi-classical) [4]</td>
<td>0.816</td>
</tr>
<tr>
<td>Hubbard-III (quasi-classical) [4]</td>
<td>$\approx 0.8$</td>
</tr>
<tr>
<td>DMFT [this work]</td>
<td>$\approx 0.65$</td>
</tr>
<tr>
<td>Dual Fermions [this work]</td>
<td>$\approx 0.3$</td>
</tr>
</tbody>
</table>

We clearly see that the critical value for the transition is smaller in a Dual Fermion calculation than within DMFT. And it is this critical value of $J_c \approx 0.3$ that likely approaches the real critical antiferromagnetic coupling constant of the s-d model for the metal-insulator transition. Firstly, because the DoS we obtained in Dual Fermions corresponds nicely to the result obtained in another work \[5\] by a different Quantum Monte Carlo method that also incorporates non-local corrections. Secondly, because Dual Fermions are the most advanced description of the s-d model to date, so this is the best result yet.

We also showed that there is a clear qualitative difference between the DoS’s for positive and negative values of the coupling constant. This difference is largest in our Dual Fermion calculation. Our results agree very well with previous works \[1\] \[2\] \[3\]. The results also leave room for future studies, by extracting more results from our calculational regime, by exploring new regimes and by developing better methods. Much has happened before we got these results. This summary serves to shed quick a light on this process.

We chose to analyse the s-d model because our greater objective is to develop and cultivate methods to study strongly correlated systems. Dual Fermions are one such method. We specifically chose this system because it is somewhat more involved than the previously analysed Hubbard model. We wanted to gauge the effectiveness of the theory of Dual Fermions. We conclude that they are very effective in the s-d model. Furthermore, results obtained on a toy model, such as the s-d model, can often be generalised to more complicated physical systems or more realistic models.

To say anything useful about the solid state, we argued that some approximations are necessary. Through these approximations we embedded the s-d model in a context of some other quantum lattice
models. We gave an account of some important physical phenomena, explaining the relevance of this work and its place in today's physics. Impurity models are a significant class of models of the solid state because they play a central role in the development of theories that have greatly improved our level of understanding of the world around us.

After this, we turned to the treatment of some modern theoretical approaches to models of the solid state. This we started by briefly treating path integrals as a useful instrument for the development of the aforementioned theoretical approaches. The following instrument we introduced was the Matsubara formalism which we used to derive Dynamical Mean Field Theory (DMFT) and the theory of Dual Fermions. These two formalisms (DMFT and Dual Fermions) allowed us to look at the significance of spatial correlations in the description of the s-d model. DMFT neglects spatial correlations and the theory of Dual Fermions includes them. We may conclude that these correlations are indeed important, especially in two-dimensional systems, because the results change significantly between the two descriptions.

DMFT consists of two crucial steps. Firstly, the mapping of the lattice problem to a single-site impurity problem. We suppose that the solution of the impurity problem coincides with the solution of the lattice problem if we take a special form of the impurity action. The action of this impurity problem contains a particular part that contains information about the lattice, the hybridisation. Secondly, the self-consistent calculation of the hybridisation. This means that the impurity problem is solved using some hybridisation as input after which the solution to the impurity problem is again used to obtain a more accurate hybridisation. This loop is repeated until self-consistency is reached.

The way in which the information about the lattice is encoded in the hybridisation is important for the results. In DMFT we carry out the above steps using a spatially uncorrelated, or Gaussian, lattice to obtain the hybridisation. In the Dual Fermion formalism we mimic these steps but we incorporate spatial correlations in the calculated quantities using a Hubbard-Stratanovich transformation.

The self-consistency loop in DMFT and Dual Fermions is carried out using a Continuous-Time Quantum Monte-Carlo (CT-QMC) solver for quantum impurity models. The CT-QMC procedure is a powerful way to circumvent the sign problem that often arises in fermionic calculations. We chose a solver that expands in the interaction because that is the only one compatible with the Dual Fermion package written by Hafermann.

Before performing the numerical calculations we analytically obtained some crude (poor man's) results in the atomic limit. After implementing the self-consistency loop and using the Maxent-procedure for the analytic continuation of the Monte-Carlo data to obtain DoS's, we obtained the main results of our work:

We first consider the case \( J > 0 \), anti-ferromagnetic coupling. For the metal-insulator transition in the s-d model, we found a positive critical value of the coupling constant \( J_c \approx 0.65 \) in DMFT and \( J \approx 0.3 \) in Dual Fermions). Note the difference between the Dual Fermions calculation and DMFT and their difference to a quasi-classical calculation \[1\] where \( J_c \approx 0.8 \). Our Dual Fermions result (on a 24×24 lattice) corresponds to that obtained using lattice Quantum Monte-Carlo on a smaller \( (8 \times 8) \) lattice \[3\]. Thus the result seems to be robust with respect to the sample size. Several other works \[1, 2\] also observe a metal-insulator transition when varying the coupling constant \( J \) and our results agree with theirs where they can be compared.

When we consider \( J < 0 \), ferromagnetic coupling, we find that the DoS's differ from the anti-ferromagnetic case while quasi-classically they should be equal \[1\]. In a Dual Fermions calculation this difference is larger than within DMFT. Our data does not show a metal-insulator transition yet, but our results are still consistent with \[2\]. They observed a metal-insulator transition at larger absolute \( J \)-values. These were inaccessible to us because we used an weak coupling (interaction) expansion whereas they used a strong coupling (hybridisation) expansion. We should mention that \[2\] is (only) a DMFT calculation. As spatial correlations are significant in the two dimensional s-d model, the critical value of the ferromagnetic coupling constant is likely different from its DMFT-value when considered within a more general approach such as Dual Fermions.

We conclude that the development of the theory of Dual Fermions is a major step forward in the
calculation of models of the solid state and of two-dimensional systems in particular. The Dual Fermion method allows us to include non-local correlations in a calculationally feasible way and thereby leads to significantly improved results. A description of models in terms of Dual Fermions is interesting in its own right but there is rich world beyond the Dual Fermion horizon that allows for a more accurate description of condensed matter. The application of Dual Fermions and their extensions offer great perspectives for the future of condensed matter physics as is explored in the next section.

5.2 Outlook

Although it is the case with all scientific work, the reader should be reminded that this work is part of a bigger picture that still partly needs to be painted. I am glad that I was able colour some patches, although a small number of them might need to be repainted afterwards. If this is indeed the case, we at least gained more insight in how we should use the Dual Fermion brush. In short, there is still a lot to do. Here we sketch some directions for future research. We distinguish between improvements in our current regime, the probing of other parameter regimes of the s-d model and the development of better methods in general.

5.2.1 Future development in our regime

There are three effects in our parameter regime that can be investigated further, a description including Dual Fermions might change results for these effects just like it did for the metal-insulator transition.

Firstly, the question if localised states contribute to the value of the DoS at the Fermi level has not yet been answered in full. This problem, explored in [1], is often referred to as the s-d model having a Large Fermi Surface or not.

Secondly, the charge susceptibility might have a maximum at the Kondo temperature as indicated in [1]. This calls for an investigation of the temperature behaviour of the s-d model around the Kondo temperature within the Dual Fermion formalism.

Thirdly, the magnitude of the insulating bandgap is expected to scale in some way with the magnetic coupling constant \( J \) as noted in [2]. We have not yet looked into this because we were primarily interested in the \( J \)-value of the metal-insulator transition itself.

Lastly, we only incorporated a nearest-neighbour interaction. This might be justified by the Kondo screening of the impurities. Of course, we cannot be sure of this until we have seen more advanced calculations or a proof that a nearest-neighbour treatment is enough.

5.2.2 Future studies in other regimes

Now that we know that a description using Dual Fermions really changes the outcome of the calculation, we should mimic the parameters of other studies to really make a comparison. In this study we only uncovered a part of the available parameter space, as we only allowed the exchange interaction to vary and only between \(-0.7\) and \(0.7\). Interesting effects might manifest themselves as a function of, for example, the temperature, the hopping parameter, the lattice type, the type of magnetic ordering, etc. Because Werner and Millis found a metal-insulator transition for large negative \(J\)-values as well it is worthwhile to extend our calculations to this regime as well and see what new results a Dual Fermion description would bring. The dependence on the lattice size is, of course, an artifact of the calculation. It seems wise to know up to which regime finite-size effects remain important. Treating lattice sizes between \(8 \times 8\) and \(24 \times 24\) could be a good starting point to study the calculational influence of these finite-size effects.

Studies of the s-d model at low temperatures might reveal new physics when treated in a Dual Fermion picture. If we want to study the Kondo resonance we need a much lower temperature than in our study. Keeping in mind that the Kondo resonance is the formation of a cloud of conduction electrons around
an impurity, we should include multiple conduction electrons if we want to study the Kondo lattice realistically. New physics might also arise if we investigate ordered states of the s-d model and states away from half-filling. We do not know how a Dual Fermion calculation would change the results in these cases.

To be able to study the low-temperature regime, the Dual Fermion package should be adapted to work with an impurity solver that is able to handle these temperatures, such as a strong coupling (CT-HYB) or auxiliary field (CT-AUX) solver. Ideally, a Dual Fermion package should be made to work together with ALPS, a very versatile and efficient open source software project for studies in strongly correlated systems. The ordered and away-from-half-filling regimes can still be treated with the existing CT-INT solver but the quantum phase transition from an ordered to a disordered state is only visible at $T = 0$ and would therefore also need a strong coupling treatment.

When it is more clear how the s-d model behaves in different regimes, the step to a detailed application of the model to one or more of the effects mentioned in section 2.3 could be taken. For instance, the phonon spectrum of plutonium, as calculated in [12] by DMFT differs from the experimental picture. A treatment using Dual Fermions could improve it. Also, a phase diagram of the s-d model could be obtained by varying at least two independent parameters and examining the state of the model.

5.2.3 New methods

Introducing Dual Fermion support in ALPS is only one example of new methods that have become available to us to study systems like the s-d model. Working with ALPS would allow us to apply Dual Fermion methods to other models more easily.

As we have seen, the results on the s-d model change when our description of the model improves. This would encourage us to improve the description even more and go beyond Dual Fermions, as Dual Fermions are unfortunately not perfect, although their results look very promising. A very natural extension of the formalism is the theory of Dual Bosons, as developed in a very recent article by Rubtsov, Lichtenstein and Katsnelson. This approach allows us to also include collective modes (magnons), such as the double exchange mechanism. These collective modes are vital in the description of systems exhibiting magnetoresistance effects. Likely, other systems also benefit from a more general approach.

The last direction we mention that can be taken from our study is the exploration of Exact Diagonalisation, ED, as a replacement of the Quantum Monte Carlo impurity solver. Hafermann claims that his ED procedure, jubilantly called the Superperturbation Solver, is much more efficient than contemporary QMC solvers. This is the case for small sample sizes, but as better computational resources become available, larger samples with several orbitals can also be treated in the ED-approach. Another strength of the ED-treatment is that the Green's functions can easily obtained on the real axis thus eliminating the need for intractable analytic continuation procedures such as Maxent. Furthermore, the results do not have a statistical error stemming from the Monte Carlo sampling. Pomerol, an ED-package developed by Krivenko and Antipov, is also a promising candidate for obtaining results in this approach. The s-d model can be a welcome test case to see how exact diagonalisation performs.

All this seems to be an enormous task for the coming years. I sincerely hope that my part in it has been useful for future scientific work.

---

1 Although the prefix Super- refers to the level where the perturbation takes place.
Appendix A

Grassmann numbers

If we want to consider a fermionic field theory, at some point we will need to work with Grassmann numbers. The reason for this is that the fermionic operators obey anticommutation relations. Their corresponding classical fields therefore also have to anticommutate and can be represented by anticommuting numbers: Grassmann numbers. They were first introduced in the context of quantum field theory by Berezin in 1966. We give a short introduction. The treatment here follows [35].

Any two Grassmann numbers $\theta$ and $\eta$ anticommute:

$$\theta\eta = -\eta\theta$$  \hspace{1cm} (A.1)

Therefore any square (or larger power) of one Grassmann number is zero:

$$\theta^2 = \theta\theta = -\theta\theta = 0$$  \hspace{1cm} (A.2)

The usual vector-space rules hold for adding to or multiplying with ordinary numbers. To define a functional integral we will need a definition of an improper Grassmann integral over all $\theta$. Here it is:

$$\int d\theta f(\theta) = \int d\theta (A + B\theta)$$  \hspace{1cm} (A.3)

Because the function $f$ can be expanded as a Taylor series which has at most a linear term. To assign a value to this integral we demand translational invariance. We shift $\theta$ by an amount $\eta$:

$$\int d\theta f(\theta + \eta) = \int d\theta (A + B\theta) + B\eta = \int d\theta (A + B\theta)$$  \hspace{1cm} (A.4)

The only linear function of $A$ and $B$ that has this property is a constant (we take to be unity) multiplied by $B$. So:

$$\int d\theta A + B\theta = B$$  \hspace{1cm} (A.5)

To be able to do quantum field theory with Grassmann numbers we need four more ingredients: integration over multiple Grassmann variables, complex Grassmann numbers, invariance of the Grassmann integral under unitary transformations and a notion of Grassmann fields. Because of a sign ambiguity we adopt the following convention for multiple integrals:

$$\int d\theta \int d\eta \theta = +1$$  \hspace{1cm} (A.6)
where the innermost integral is performed first. We may define complex Grassmann numbers in the same way as we usually do this with ordinary numbers. Only it convenient to reverse the order when complex conjugating a product of Grassmann numbers:

$$(\theta \eta)^* := \eta^* \theta^* = -\theta^* \eta^*$$  \hfill (A.7)

To be able to integrate over the real and imaginary parts of Grassmann numbers independently we define:

$$\theta = \theta_1 + i \theta_2 \sqrt{2}, \quad \theta^* = \theta_1 - i \theta_2 \sqrt{2}. \hfill (A.8)$$

With the convention $\int d\theta^* d\theta (\theta \theta^*) = 1$ we are ready to calculate the following Gaussian integral.

$$\int d\theta^* d\theta e^{-\theta^* b \theta} = \int d\theta^* d\theta (1 - \theta^* b \theta) - \int d\theta^* d\theta (1 + \theta^* b) = b \hfill (A.9)$$

An extra factor of $\theta \theta^*$ gives:

$$\int d\theta^* d\theta \theta \theta^* e^{-\theta^* b \theta} = 1 \hfill (A.10)$$

To prove invariance of a Grassmann integral under unitary transformations we need $n$ Grassmann numbers: $\theta_i$ and a unitary operator $U_{ij}$. We take $\theta'_i = U_{ij} \theta_j$. And we have the following general product:

$$\prod_i \theta'_i = \frac{1}{n!} e^{\sum_{i<j} \theta'_i \theta'_j} \hfill (A.11)$$

$$= \frac{1}{n!} e^{\sum_{i<j} U_{ik} \theta_k U_{lj} \theta_l \theta_j} \hfill (A.12)$$

$$= \frac{1}{n!} e^{\sum_{i<j} U_{ik} \theta_k U_{lj} \theta_l e^{\sum_{j'=j} \theta_j} (\prod_j \theta_j)} \hfill (A.13)$$

$$= (\det U) (\prod_i \theta_i). \hfill (A.14)$$

Now if we have a general integral

$$\left( \prod_i \int d\theta^*_i d\theta_i \right) f(\theta), \hfill (A.15)$$

only the term with exactly one factor of both $\theta_i$ and $\theta_i^*$ survives. This term is proportional to the product $(\prod_i \theta_i) (\prod_i \theta^*)$. If we perform a unitary transformation: replacing $\theta$ with $U \theta$, this term picks up a factor $(\det U)/(\det U)^* = 1$ so a general integral remains unchanged by a unitary transformation.

A general bilinear Gaussian integral involving $n$ Grassmann numbers and a Hermitian matrix $B$ with eigenvalues $b_i$ looks like:

$$\left( \prod_i \int d\theta^*_i d\theta_i \right) e^{-\theta^*_i B_{ij} \theta_j} = \left( \prod_i \int d\theta^*_i d\theta_i \right) e^{-\sum_i \theta_i^* b_i \theta_i} = \prod_i b_i = \det B. \hfill (A.16)$$

In a similar fashion:

$$\left( \prod_i \int d\theta^*_i d\theta_i \right) \theta_i \theta_i^* e^{-\theta^*_i B_{ij} \theta_j} = \left( \prod_i \int d\theta^*_i d\theta_i \right) e^{-\sum_i \theta_i^* b_i \theta_i} = \prod_i b_i = (\det B)(B^{-1})_{kl}. \hfill (A.17)$$

A Grassmann field $\psi(x)$ we may define as a summation over an ordinary set of orthonormal basis functions $\phi(x)$ in space-time with Grassmanian coefficients:

$$\psi(x) = \sum_i \psi_i \phi_i(x) \hfill (A.18)$$

This is all we need to evaluate the functional integrals that are for instance needed to develop DMFT.
Appendix B

Proof of expansion of Fermi function

In section 3.3 we encountered a series expansion for the Bose and Fermi occupation functions, eqs. (3.22) and (3.23). Here we shall prove these expansions.

We consider the integral \( I \) in the complex plane over the boundary of a square \( R \) with sides of length \( 2N + 1 \) symmetrically around the origin: the real axis is cut at the points \( \pm (N + \frac{1}{2}) \) where \( N \in \mathbb{N} \), between the poles at \( \pm N \) and \( \pm (N + 1) \).

\[ I = \int_{\square} \frac{\cos(\pi z)}{\sin(\pi z)} \frac{1}{z^2 - w^2} \, dz, \quad (B.1) \]

Figure B.1: The contour (dashed square) of integration and the poles (dots) of the function we are integrating.
where \( w \in \mathbb{C} \setminus \mathbb{Z} \), i.e., any complex number not coinciding with the zeroes of the sine in the first denominator. By the residue theorem we may evaluate \( \mathcal{I} \) by summing the residues at the poles of the integrand inside \( R \)

\[
\mathcal{I} = 2\pi i \sum_{\text{poles}} \text{Res} (\text{Res} \in R). \tag{B.2}
\]

There are first order poles at \( z = \pm w \) and for \( z \in \mathbb{Z} \).

\[
\text{Res}_w = \lim_{z \to w} (z - w) \frac{\cos(\pi z)}{\sin(\pi z)} z^2 - w^2 = \frac{\cos(\pi w)}{2w \sin(\pi w)}. \tag{B.3}
\]

The same value is found for the residue at \( z = -w \). The residue at the poles of the sine (\( z = k \)) with \( k \in \mathbb{Z} \) is:

\[
\text{Res}_{\text{sine}} = \lim_{z \to k} \frac{z - k}{\sin(\pi z) - \sin(\pi k)} \cos(\pi z) z^2 - w^2. \tag{B.4}
\]

We may add zero to the denominator in the form of \( \sin(\pi k) \)

\[
\text{Res}_{\text{sine}} = \lim_{z \to k} \frac{z - k}{\sin(\pi z) - \sin(\pi k)} \frac{\cos(\pi k)}{\pi \cos(\pi k) k^2 - w^2}. \tag{B.5}
\]

Should we take the limit we would end up with an expression of the form \( \frac{0}{0} \). We apply L'Hôpital's rule to the first factor and differentiate the first numerator and the denominator:

\[
\text{Res}_{\text{sine}} = \frac{1}{\pi \cos(\pi k) k^2 - w^2} \frac{\pi}{\pi k^2 - (\pi w)^2}. \tag{B.6}
\]

We now have for \( \mathcal{I} \),

\[
\mathcal{I} = 2\frac{1}{2w \sin(\pi w)} \frac{\cos(\pi w) + \sum_{k=-N}^{N} \frac{\pi}{(\pi k)^2 - (\pi w)^2}}{\sum_{k=-N}^{N} (\pi k)^2 - (\pi w)^2}. \tag{B.7}
\]

It would be convenient if this entire integral \( \mathcal{I} \) were equal to zero. Let us evaluate it to see if this is so. The curve has length \( 8N + 4 \), if the absolute value of the integrand is proportional to \( N^{-2} \) then \( \lim_{N \to \infty} \mathcal{I} = 0 \). Which is already a comforting thought. It is sufficient to show that \( A = \cos(\pi z) / \sin(\pi z) \) is bounded by a value independent of \( N \) since \( A(z^2 - w^2)^{-2} \) is bounded by \( A(N^2 + C)^{-1} \) on the contour.

We start with the term \( A \) at the part of the contour where \( y \geq 1 \); \( z = x + iy \) with \( x, y \in \mathbb{R} \).

\[
\cos(\pi z) = \frac{e^{i\pi z} + e^{-i\pi z}}{e^{i\pi z} - e^{-i\pi z}} \tag{B.8}
\]

\[
\frac{\cos(\pi z)}{\sin(\pi z)} = \frac{|e^{2i\pi z} + 1|}{|e^{2i\pi z} - 1|} \leq \frac{|e^{2i\pi z} + 1|}{|e^{2i\pi z} - 1|} \leq \frac{|e^{2i\pi z} + 1|}{|e^{2i\pi z} - 1|} \leq e^{2\pi y}. \tag{B.9}
\]

|\[ e^{2i\pi z} - 1 | = |e^{2i\pi x - 2\pi y}| = e^{-2\pi y}. \tag{B.10}
\]

Therefore

\[
\frac{\cos(\pi z)}{\sin(\pi z)} \leq e^{2\pi y} \tag{B.11}
\]

This function of \( y \) has a maximum at \( y = 1 \).

For \( y \leq 1 \) we get a similar result:

\[
\frac{\cos(\pi z)}{\sin(\pi z)} \leq e^{-2\pi y + 1} \tag{B.12}
\]
Now we are left with the line segments at $x = \pm (N + \frac{1}{2})$. Here $A$ is bounded because it is a periodic function: if it is bounded on one line segment it is bounded on all line segments. We conclude that $A$ is bounded on the entire contour $R$. Therefore we conclude:

$$
\frac{\cos(\pi w)}{\sin(\pi w)} = \sum_{k=-\infty}^{\infty} \frac{\pi w}{(\pi w)^2 - (\pi k)^2}.
$$

(B.13)

The restrictions on $w \neq 0$ and $w \neq k$ still seem reasonable in this expression. Next, we let $\pi w = z$, we introduce a minus sign in the denominator and we note that we may split the quadratic terms inside the sum, change the summation boundaries and gather all terms in one sum:

$$
\sum_{k=-\infty}^{\infty} \frac{\pi w}{(\pi k)^2 - (\pi w)^2} = \sum_{k=-\infty}^{\infty} \frac{\pi k - z}{\pi k - z} + \frac{\pi k + z}{\pi k + z} = 1 + 2 \sum_{k=1}^{\infty} \frac{\pi k - z}{\pi k - z} - \frac{1}{\pi k - z}.
$$

(B.14)

(B.15)

For the left hand side of equation (B.13) we write:

$$
\frac{\cos(z)}{\sin(z)} = i \left( \frac{e^{iz} + e^{-iz}}{e^{iz} - e^{-iz}} + 1 \right) = i \left( \frac{2e^{iz}}{e^{iz} - e^{-iz}} \right) - i = \frac{2i}{1 - e^{-2iz}} - i.
$$

(B.16)

We can now write, introducing a minus in front of $z$

$$
\frac{1}{e^{2iz} - 1} = -\frac{1}{2} + \frac{1}{2i} \sum_{k=-\infty}^{\infty} \frac{z}{(\pi k)^2 - z^2} = -\frac{1}{2} + \frac{1}{\beta} \sum_{k=-\infty}^{\infty} \frac{1}{2\pi ik/\beta - 2iz/\beta}.
$$

(B.17)

We rescale $z$ with the following factor: $z \rightarrow 2iz/\beta$, and we arrive at the desired result (at least for the boson-equation: (3.22)) if we identify $z$ with $\omega q$. So in the end we have:

$$
\frac{1}{e^{\omega q} - 1} = -\frac{1}{2} + \frac{1}{\beta} \sum_{k=-\infty}^{\infty} \frac{1}{2\pi ik/\beta - \omega q}.
$$

(B.18)

For the fermion-equation we write in much the same way

$$
\frac{\cos(z)}{\sin(z)} = i \left( \frac{e^{iz} + e^{-iz}}{e^{iz} - e^{-iz}} + 1 \right) = \frac{2i}{e^{2iz} - 1} + i = -\sum_{k=-\infty}^{\infty} \frac{1}{\pi k - z}.
$$

(B.19)

we introduce $\beta$ in the same way as before and make a shift $z \rightarrow z - \frac{\pi}{2}$ so that

$$
\frac{1}{e^{2iz} + 1} = \sum_{k=-\infty}^{\infty} \frac{1}{(2k + 1)\pi i - 2iz}.
$$

(B.20)

We again scale $z$ in the same way as before, which completes the proof of (3.23)

$$
\frac{1}{e^{\epsilon q} + 1} = \frac{1}{2} + \frac{1}{\beta} \sum_{k=-\infty}^{\infty} \frac{1}{(2k + 1)\pi i/\beta - \epsilon q}.
$$

(B.21)
Bibliography


