

# BACHELOR THESIS:

# Multidimensional Density of States for Many-Body Calculations

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# Contents

Ał	stract		1
1	Introduction1.1Many-Body Calculations:1.2Hubbard model1.3Limit of infinite dimensions:DMFT1.4Density of States (DOS)1.5General remarks	· · · ·	<b>2</b> 2 3 6 6 7
2	Density of States         2.1       free DOS in d dimensions         2.1.1       Calculating the DOS by using Green's functions         2.2       Tight-binding (TB) method         2.3       TB DOS in finite dimensions         2.4       TB DOS in infinite dimensions         2.4.1       Effect of infinite Dimensions on the Green's function	· · · ·	<b>9</b> 9 13 19 25 27
3	Implementation in code and technical details         3.1       Noninteracting DOS         3.2       DMFT Self-Consistency-Cycle:         3.3       Spectral function         3.4       Optimization for calculating the non-interacting DOS         3.4.1       Variational k-subdivision         3.4.2       Trapezoid- or Simpson-summation         3.5       Symmetries	· · · · · · · ·	<ol> <li>29</li> <li>32</li> <li>35</li> <li>36</li> <li>36</li> <li>39</li> <li>40</li> </ol>
4	Results         4.1       Two dimensions         4.2       Three dimensions         4.3       Infinite dimensions         4.4       'Under-cooling' the Mott-Hubbard-transition	· · · ·	<b>42</b> 43 52 58 63
5	Conclusion and Outlook		66

#### Abstract

Some of the most intriguing phenomena in condensed matter physics are found in *strongly correlated materials*. However, these interacting (or correlated) systems represent, by their nature, a many-body-problem which makes the theoretical (as well as the experimental) studies extremely challenging. In the last two decades, a new theoretical framework was developed that makes it possible to calculate the electronic structure of those materials. This is the so called *Dynamical Mean Field Theory* (DMFT). This theory exploits the possibility of mapping, in the limit of large spatial dimensions, a lattice type model like the *Hubbard Model* onto a single site *Anderson Impurity Model* in a self-consistency cycle. Concerning purely model studies, which are of crucial importance for the further development of the theory, most calculations take into account only the movement of the electrons between two lattice sites *directly* neighbouring each other. While this is indeed almost always the dominant 'hopping term', in many situation the disregard of an overlap of sites further apart will not be justified.

It is the main goal of this work to implement in an existing DMFT code for singleband calculations electronic dispersions with different hopping terms beyond nearest neighbour (NN) hopping, and identify their main effect on the DOS and the Fermi surface of the model. While the DMFT approach is exact only in infinite dimensions, most of the materials of interest are two or three-dimensional. Thus, we will look especially at these cases and analyse the effects of further hopping terms in two and three dimensional materials, and, eventually, we will look at the more academic, infinite dimensional case.

# 1 Introduction

## 1.1 Many-Body Calculations:

After decades of research we appear to come close to answer Faust's question

## '... That I may understand whatever Binds the world's innermost core together,...

Physicists managed to make remarkably progress in answering this question (e.g., by the standard model). But by doing so, the theories became more and more complex. This is why what sometimes is believed to become the 'Theory of Everything' (if it exists) might have rather little impact on our understanding of physics at low energy scales,[1] In solid state theory, to which we are interested here, due to the energy scales considered, any observable could be calculated, if we were able to solve the 'simple' Schrödinger eigenvalue problem:

$$\hat{H} \left| \psi \right\rangle = E \left| \psi \right\rangle \tag{1}$$

If we neglect relativistic effects for the moment, our Hamiltonian of a set of atomic nuclei  $\{\alpha\}$  with atomic numbers  $\{Z_{\alpha}\}$  and nuclear masses<sup>1</sup>  $\{M_{\alpha}\}$  and electrons  $\{i\}$  has the form:

$$\hat{H} = -\frac{1}{2}\sum_{i}\nabla_{i}^{2} - \sum_{\alpha}\frac{1}{2M_{\alpha}}\nabla_{\alpha}^{2} - \sum_{\alpha,i}\frac{Z_{\alpha}}{|\mathbf{r}_{i} - \mathbf{R}_{\alpha}|} + \frac{1}{2}\sum_{i,i'}\frac{1}{|\mathbf{r}_{i} - \mathbf{r}_{i}'|} + \frac{1}{2}\sum_{\alpha,\alpha'}\frac{Z_{\alpha}Z_{\alpha'}}{|\mathbf{R}_{\alpha} - \mathbf{R}_{\alpha'}|}$$

Our innocent eigenvalue problem is not that innocent any more. Not only because the sums might have an Avogadro number as an upper limit, but also for of the *correlation* of the whole system. Each element of the system (protons, electrons) interacts electro statically with every other element of the system, which is a vast number of involved particles. In some cases this interaction is rather weak (e.g. due to screening effects) and a standard mean field approach can be applied. Other cases, especially materials with open 3d and 4f shells, can not be treated this way. We can understand this if we take a look at the radial-part of the corresponding wave function. Whenever the main quantum-number of the orbital differs from the angular momentum quantum number by exactly one there are no nodes in the wave function. Thus the electrons residing in these orbitals have on average a higher probability of being close in space, which is exactly the reason why they effectively 'interact', with one another, rather strongly. The strong correlation combined with the electron's internal degrees of freedom (spin, charge, angular momentum), can lead to a whole zoo of exotic ordering phenomena at low temperatures [2]. Hence, though possible in principle, it is practically impossible to solve the many-body problem without any approximation. Moreover, the reductionist's approach to derive universal laws and build up the cosmos by deduction, reaches its limit quickly as the Nobel laureate P. A. Anderson remarked: [3]

<sup>&</sup>lt;sup>1</sup>in atomic units

The ability to reduce everything to simple fundamental laws does not imply the ability to start from these laws and reconstruct the universe.[...]

The behaviour of large and complex aggregates of elementary particles, it turns out, is not to be understood in terms of simple extrapolation of the properties of a few particles.

Therefore as a first step we clearly need to reduce our model to a simpler one. This is by all means not trivial. The simplified model needs to be simple enough, to be worked with, but it also still needs to contain the physics we are interested in. This physics will be extracted by calculating, for the reduced model, the following quantities:

- eigenvalues of observables
- expectation values of observables  $\langle \hat{A}(t) \rangle, \langle \hat{B}(t') \rangle$
- correlation functions of observables  $\langle \hat{A}(t)\hat{B}(t') \rangle$

Because of the large number of spectroscopic transport data available in experiments, we are especially interested in the third kind. Hence one must choose a model which, at least in principle, allows for a non-trivial result with respect to these properties. A drastically simplified model was introduced by J. Hubbard and it is still one of the most studied in non relativistic quantum many body theory.

#### 1.2 Hubbard model

The Hubbard model reduces the vast set of bound and continuum electron-levels to a single localized orbital level. In its simplest form, it is a single band model. Each lattice position has one electron orbital with four possible electron configurations: Empty, singly occupied (with either spin up or down), or doubly occupied. The Coulomb interaction between the electrons is considered to be purely local. If a site is doubly occupied the system has to 'pay' the energy U. (This may seem as if this was a very problematic assumption. We know that the electrons and the ions will have some sort of a Coulomb interaction, which is of long range type and therefore by no means localized. However, the screening of the electrons leads to an effective interaction in a much smaller range. This can already be motivated by the Thomas-Fermi screening theory or its extensions.) We can also derive the Hubbard Hamiltonian directly from the many-body-problem by performing a basis-transformation to localized functions (e.g. tight-binding, Nth order muffin tin orbital or a Wannier basis), and then only retain the major part of the Coulomb-interaction. <sup>2</sup> An illustration of the Hubbard model can be seen in figure 1. Eventually the Hamiltonian (in its simplest version) contains only two terms:

(a) a term diagonal in real space, that is just the positive constant energy U times the number of doubly occupied sites. We could also introduce a contribution which

<sup>&</sup>lt;sup>2</sup>For a detailed derivation of the Hubbard model see [4].

results from each singly occupied site, but this would only lead to an unimportant additional correction of the chemical potential, and therefore can be neglected. U is the *interaction/ potential energy*.

(b) an off-diagonal term with constant non-zero matrix t elements between 'neighbouring' sites in real space. The matrix-element t is therefore called *hopping term*. Neighbouring sites can mean, in the simplest case, that the difference in position space is exactly the lattice constant a. Of course, one can also allow other contributions: In the most general case we write  $t(\mathbf{i}, \mathbf{j})$  where as  $\mathbf{i}$  and  $\mathbf{j}$  are the positions of the respective sites.

In the absence of the first term the Hubbard-model reduces to the *tight binding* model and we can diagonalize the remaining Hamiltonian in k-space, which will be done in the next section. This contribution represents the *kinetic energy* of the system.

The Hubbard Hamiltonian in second quantization reads

$$H = H_{I} + H_{II}$$
$$\hat{H}_{I} = \sum_{\mathbf{R}_{i}, \mathbf{R}_{j}} \sum_{\sigma} t_{(i,j)} \hat{c}_{\sigma,i}^{\dagger} \hat{c}_{\sigma,j} = \sum_{\mathbf{k}, \sigma} \epsilon_{\mathbf{k}} \hat{n}_{\mathbf{k}, \sigma}$$
$$\hat{H}_{II} = U \sum_{\mathbf{R}_{i}} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$

where  $\hat{c}_{\sigma,i}^{\dagger}(\hat{c}_{\sigma,i})$  is the creation (annihilation) operator for an electron at site  $\mathbf{R}_i$  with spin  $\sigma$ . So the number-operator  $\hat{n}_{i,\sigma} = \hat{c}_{\sigma,i}^{\dagger}\hat{c}_{\sigma,i}$  in real space ( $\hat{n}_{k,\sigma} = \hat{c}_{\sigma,k}^{\dagger}\hat{c}_{\sigma,k}$  in k-space) gives the number of electrons at a certain lattice site (k-state) with spin  $\sigma$ . If only one term is present, the system favours localized magnetic momenta for  $H_{II}$ , whereas  $H_I$  leads to a conventional band structure where each electron is spread throughout the entire crystal. However, when both terms are present and of similar magnitude the situation becomes quite complicated. In fact, despite the apparent simplicity of its operational expression, the Hubbard model leads to a complicated many-body-problem. An analytic solution for this problem has not been found vet.<sup>3</sup> However, as we discuss

in the next section, its properties can be investigated accurately in some limiting cases, e.g. in infinite dimensions, where the theory is capable of explaining a rich variety of interesting physical phenomena, like the *Mott-Hubbard-metal-isolator-transition* (MIT).

<sup>&</sup>lt;sup>3</sup>Except for the one-dimensional Hubbard model by means of an algebraic Bethe ansatz.



Fig. 1 Illustration of the interacting electrons in the Hubbard-model. The ions form a static grid of lattice sites. The electrons can move between 'neighbours' by the corresponding matrix elements (t, t', ...) as long as the Pauli-exclusion-principle is fulfilled. Whenever a site is doubly occupied the system 'pays' the interaction energy U.

Now that we have found a suitable model for the physical phenomena we are interested in, it is time to think about methods for solving it. We recall that a well-established method for obtaining the electronic structure, from the general solid state Hamiltonian Eq. (1), is the density functional theory (DFT), which treats (for example in the local density approximation LDA) correlation effects on the level of the non-interacting electron gas. The energy of the system can, then, be regarded as a functional of the electron density, what is a tremendous simplification (e.g. there are no longer three to the power of the particle-number degrees of freedom to consider, but merely three). This approach is surprisingly accurate for a vast class of materials, but because of its single electron nature it is restricted to materials where the picture of non-correlated electrons is adequate. However, the most intriguing physics is exhibited by those materials where this assumption is no longer valid. We are, of course, especially interested in those 'hot topics' of modern solid state physics, like high temperature superconductors in cuprates, large thermopower in cobaltates, or unconventional superconductivity in iron based compounds. All of these phenomena have in common that they occur in so called *strongly* correlated materials where the standard methods for calculating electronic structures break completely down.

A simple mean field theory will clearly not do the job, since it averages over space as well as over time, and therefore cannot capture the correlations. <sup>4</sup> There exists, however, a 'quantum' generalisation of the standard classical mean field theory (MFT).

<sup>&</sup>lt;sup>4</sup>For most of those hot topics the Hubbard Hamiltonian, in its single band form, would be an oversimplification. The DMFT approach, however, is quite general and leads to valuable insights also for more realistic multi-orbital cases [5], where the Hamiltonian parameter can be estimated 'ab-initio'.

#### 1.3 Limit of infinite dimensions: DMFT

Just as MFT, the dynamical mean field theory becomes exact in infinite dimensions. (So each lattice site is imagined to have infinitely many neighbours.) To still obtain non trivial results the one particle part  $H_I$  Hamiltonian has to be rescaled by the (square root of the) number of dimensions [6]. In this scaling process the competition between kinetic energy and the major (local) part of the Coulomb interaction is retained. This leads to tremendous simplifications compared to finite dimensions. With these assumptions, by taking the limit of infinite dimensions, the self-energy becomes purely local (which is equivalent to a k independent Fourier-transform). By making use of this fact we can solve a model of the Hubbard-type by mapping the model self-consistently onto a quantum impurity model (e.g. the Anderson impurity model). This is illustrated in figure 2: In the limit of high spatial dimensions the d-dimensional lattice model can be effectively described by the dynamics of correlated fermions on a single site embedded in a bath of other particles [7].

The complete set of self-consistency equations of DMFT are given in Chapter 3, where this self-consistency cycle is reviewed in more detail.

However, the results can be seen as a good approximation for three- and maybe even two-dimensional systems. The main difference to MFT is that we only take the spatial average, but not the temporal average [8]. So the temporal, but local, fluctuations are completely retained in DMFT.



Fig. 2 Illustration of the Dynamical Mean Field Theory. The complicated lattice problem is approximated by a mapping onto a single impurity interacting with a dynamical bath.

In the single band case (which is the only case that shall be discussed in this thesis) as well as in the degenerated multiorbital one, due to the locality of the DMFT, the **only** way how the structure of our lattice can enter the calculations is through the *Density of States* (DOS) of the non-interacting electrons.

## 1.4 Density of States (DOS)

The density of states is a widely used concept in condensed matter theory and statistical mechanic, well beyond the DMFT treatments of simple problems.

To explain the role of the DOS, we recall that calculations of thermodynamic quantities, response functions and Feynman diagrams in QFT of condensed matter systems often require the evaluation of integrals or sums over all momenta  $\mathbf{k}$  (typically over the first Brillouin Zone). An important simplification of these  $\mathbf{k}$ -summations is possible, however, when the integrand  $\mathcal{F}$  is depending on the energy  $\epsilon(\mathbf{k})$  only. In this case the integration/sum is best performed by using the energy  $\epsilon$  as a variable. In the case of a cubic lattice of volume  $L^d$  in d dimensions we have a given observable F: [9]

$$F = \frac{1}{L^d} \sum_{\mathbf{k}} \mathcal{F}(\epsilon_{\mathbf{k}}) = \frac{1}{(2\pi)^d} \frac{(2\pi)^d}{L^d} \sum_{\mathbf{k}} \mathcal{F}(\epsilon_{\mathbf{k}}) \approx \frac{1}{(2\pi)^d} \int \mathrm{d}^d k \, \mathcal{F}(\epsilon_{\mathbf{k}}) = \int \mathrm{d}\epsilon \, \mathcal{N}(\epsilon) \, \mathcal{F}(\epsilon)$$

This yields the Density of States (DOS) as:

$$\mathcal{N}(\epsilon) = \frac{1}{L^d} \sum_{\mathbf{k}} \delta(\epsilon - \epsilon_{\mathbf{k}})$$

And for the continuous case:

$$\mathcal{N}(\epsilon) = \frac{1}{(2\pi)^d} \int \mathrm{d}^{\mathrm{d}}k \,\delta(\epsilon - \epsilon_{\mathbf{k}})$$

Hence the DOS can be used to transform a d-dimensional k-integral into an onedimensional energy integral by counting the available states in a certain energy interval. The number of available states changes with energy and the density of states captures this variation.

The calculation of the DOS for different dimensions and dispersion relations, as well as their algorithmic implementing into the DMFT cycle, is the central part of this bachelorthesis.

## 1.5 General remarks

For the sake of simplicity, in this thesis we only consider cubic crystal where all lattice constants are equal to a (simple cubic crystal). The specific 'Bravais lattices' (sc, bcc & fcc) enter through the corresponding hopping terms (see figure 4). Within the programmed subroutines, in the program 'ver\_tprime', the lattice constant a is equal to 1. We also set  $\hbar$  to 1, as well as the NN hopping term t, throughput the whole thesis, as well as in the program. <sup>5</sup>

Since we are only interested in bulk properties, we use periodic boundary conditions for crystal structures. Moreover, phonons and other quasi-particles involving the lattice dynamics are not considered in this thesis. Thus, we assume the lattice to be rigid. Improper Riemann integrals with different forms (and other non-trivial mathematical objects like the Dirac delta distribution) occur within this thesis quite often and are not explicitly and properly mentioned at each point of use. Unless specified, it is simply assumed that by using the 'right measure' convergence is guaranteed. In the same

<sup>&</sup>lt;sup>5</sup>In the spirit of Univ. Prof. Dr. Arno Rauschenbeutel: 'Wir setzen alles zu 1, was nicht schnell genug auf dem Baum ist ...'(orally transmitted during an AKT I lecture in March 2014)

tenor also uniform convergence is assumed and not dealt with explicitly (e.g. to ensure the integral commutativity) and shorthand notation is usually adopted for the Fourier transformation.<sup>6</sup>

<sup>&</sup>lt;sup>6</sup>Or in other words: The usual 'physical' approach to these calculations is applied.

## 2 Density of States

## 2.1 free DOS in d dimensions

Before starting with the tight binding dispersions we will consider for the implementation in the DMFT code, we can familiarize ourselves with the density of states by taking a look at the case of free electrons. For free electrons the dispersion relation reads as follows  $\epsilon_k = \frac{1}{2} \sum_{r=1}^d k_i^2$ . If we transform to spherical *d*-dimensional coordinates  $\mathbf{x} = r\mathbf{e}_r$  and use

the linearity of the functional determinant  $F(r) = r^{d-1}F(1)$  we get:

$$\frac{1}{(2\pi)^d} \int_{\mathbb{R}^d} \mathrm{d}^d k \, \delta(\epsilon_{\mathbf{k}} - \epsilon) = \frac{1}{(2\pi)^d} \tilde{F}(1) \int_0^\infty \mathrm{d}r \, r^{d-1} \delta\left(r^2/2 - \epsilon\right) = \frac{1}{(2\pi)^d} \tilde{F}(1) r^{d-2} \bigg|_{\sqrt{2\epsilon}} = \frac{\tilde{F}(1)}{2^{d/2+1} \pi^d} \epsilon^{d/2-1} \delta\left(r^2/2 - \epsilon\right) = \frac{1}{(2\pi)^d} \left(r^2/2 - \epsilon\right) = \frac{1}{(2\pi)$$

where  $\tilde{F}(1) = \int_{B(1)} \mathrm{d}\Omega F(1)$  is the surface of an *d*-dimensional ball of radius 1.

Moreover, now we can write a general relation between the Fermi-energy  $\epsilon_F$  and the electron-density n = N/V by using:

$$\int_{-\infty}^{\epsilon_F} \mathrm{d}\epsilon \,\mathcal{N}(\epsilon) = n$$

hence one finds that  $\epsilon_F \propto n^{2/d}$ , and for the Fermi-radius  $k_F \propto n^{1/d}$ .

## 2.1.1 Calculating the DOS by using Green's functions

This subsection is intended to get us familiar with Green's functions, and their connection to the DOS. Notice, that we only look at **non-interacting** systems from the very beginning<sup>7</sup>. In order to solve the time-dependent Schrödinger equation<sup>8</sup>, Green's functions turn out to be useful. There are also quite a few other applications of Green's functions, especially regarding many-body-theory.<sup>9</sup>

The Green's function(s) for the differential equation  $\mathcal{L}_x\psi(x) = f(x)$  in the mathematical sense is defined by:  $\mathcal{L}_x G(x, x') = \delta(x - x')$  Where  $\mathcal{L}_x$  is the linear operator of our linear differential equation, and f(x) is the inhomogeneity of our differential equation. Thus, for the time-dependent Schrödinger equation, one finds

$$(i\partial_t - H(r))G(r, r', t, t') = \delta(r - r')\delta(t - t')$$

Because of the translational invariance of the linear operator regarding  $t^{10}$ , the Green's function only depends on the difference  $t - t' = \tau$  and a Fourier-transformation can be

<sup>&</sup>lt;sup>7</sup>For a interacting system we would have to deal with a whole hierarchy of equations determining the Green's functions.

<sup>&</sup>lt;sup>8</sup>Or any other linear differential equation with an inhomogeneity for that matter.

<sup>&</sup>lt;sup>9</sup>Whereas the definition of these 'Green's functions' are only in very few cases (only two point-Green functions of non interacting systems) the same as in the mathematical sense.

<sup>&</sup>lt;sup>10</sup>t is the time in this subsection and may not be confused with the hopping term.

applied:

$$G(r, r', \tau) = \frac{1}{2\pi} \int G(r, r', E) \exp(-iE\tau) dE$$

G(r, r', E) can be evaluated easily if a complete set of eigenfunctions  $\psi_j(r, t) = \psi_j(r)e^{\frac{iEt}{\hbar}}$ is known:  $G(t, t') = \sum_{i=1}^{n} \frac{\psi_i(t)\overline{\psi_i(t')}}{\lambda_i}$  (This approach is called finding Green's function by 'spectral decomposition' or 'Lehmann representation') assuming  $\lambda_i$  are the eigenvalues <sup>11</sup> (for the Schrödinger equation:  $E - E_n$ ) Thus

$$G(r, r', E) = \frac{1}{2\pi} \int \sum_{i=1}^{n} \frac{\psi_i(r)\overline{\psi_i(r')}}{E - E_i} \exp(-iE\tau) dE$$

Since this integral has a singularity on the real axis the residue theorem cannot be applied in this form. If, however, the singularity is shifted by  $\pm i\delta$  it can. Because of its related properties on the time-domain, for  $+i\delta^{-12}$  the function is called 'retarded Green's-function'  $G^R$ , and in the case of  $-i\delta$  'advanced Green's function'  $G^A$ .

The Sokhotsky formula<sup>13</sup>

$$\frac{1}{x+i0^{\pm}}[\phi] = \lim_{\alpha \to 0^+} \frac{1}{x \pm i\alpha}[\phi] = \mathcal{P}\left(\frac{1}{x}\right)[\phi] \mp i\pi\delta[\phi] \tag{2}$$

can be applied on the (retarded or advanced) Green's function. The Cauchy principal value (denoted by  $\mathcal{P}$ ) of a real distribution is again real, moreover the second term is purely imaginary, and can therefore easily be separated from the first one by taking only the imaginary part.

$$\lim_{\delta \to 0^+} \operatorname{Im} \left( \int_{-\infty}^{\infty} G^R(r, r', E) dE \right) = \lim_{\delta \to 0^+} \operatorname{Im} \int_{-\infty}^{\infty} \left( \sum_n \frac{\psi_n(r) \overline{\psi_n(r')}}{E - E_n + i\delta} dE \right) = \\ = \lim_{\delta \to 0^+} \operatorname{Im} \int_{-\infty}^{\infty} \left( \sum_n \frac{\psi_n(r) \overline{\psi_n(r')}}{\underbrace{(E - E_n)}_x + i\delta} \underbrace{d(E - E_n)}_{dx} \right) = \\ = -\pi \int_{-\infty}^{\infty} \underbrace{\sum_n \psi_n(r) \overline{\psi_n(r')} \delta(E - E_n)}_{\rho(r, r', E)} dE$$

where the integrand in the last term is the definition of the density matrix [10]. The desired density of states can be extracted by letting r' go towards r and taking the spatial

<sup>&</sup>lt;sup>11</sup>This proves to be correct since:  $\mathcal{L}_x G(x - x') = \sum_{i=1}^n \frac{\mathcal{L}_x \psi_i(x) \overline{\psi_i(x')}}{\lambda_i} = \sum_{i=1}^n \psi_i(x) \overline{\psi_i(x')} = \delta(x - x')$ where we used that the set of wave functions are complete

 $<sup>^{12}\</sup>delta$  is assumed > 0

<sup>&</sup>lt;sup>13</sup>Also known as Plemelj-, Weierstass-, Sokhatsky-Weierstrass-, or the Plemelj-Sokhotsky-formula. In this notation:  $f(x)[g] = \int_{-\infty}^{\infty} f(x)g(x)dx$  in the sense of a principal value distribution

average:

$$\frac{1}{L^d} \int_{L^d} \mathrm{d}^d r \lim_{r' \to r} \rho(r, r', E) \approx \frac{1}{L^d} \frac{L^d}{(2\pi)^d} \int_{-\pi}^{\pi} \mathrm{d}^d k \,\delta(E - \epsilon_{\mathbf{k}}) = \mathcal{N}(\epsilon)$$

Summarizing the results we get<sup>14</sup>:

$$\frac{1}{L^d} \frac{\mp 1}{\pi} \int_{L^d} \mathrm{d}^d r \lim_{r' \to r} \lim_{\delta \to 0^+} Im\left(G^{R/A}(r, r', E)\right) = \mathcal{N}(\epsilon) \tag{3}$$

So this would be a further (though completely equivalent) way of calculating density of states. Let's take a look at the already discussed free-electron-gas.

#### Free electron Green's-Function in 1D

The set of eigenfunctions and the corresponding eigenvalues are:

$$\psi_k(r) = \frac{1}{\sqrt{L}} e^{ikx} \quad , \quad E_k = \frac{k^2}{2}$$

Starting from the retarded Green's function in spectral representation we get

$$G^{R}(x, x', E) = \sum_{n} \frac{\psi_{n}(r)\overline{\psi_{n}(x')}}{E - E_{n} + i\delta} \approx \frac{L}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}k \frac{\frac{1}{\sqrt{L}}e^{ikx}\frac{1}{\sqrt{L}}e^{-ikx'}}{E - \frac{k^{2}}{2} + i\delta} = \frac{2}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}k \frac{e^{ik(x-x')}}{(\underbrace{\kappa}{\sqrt{(E+i\delta)2}} - k)(\kappa + k)}$$

for x - x' > 0 disappears the upper integration contour, for x - x' < 0 the lower one.<sup>15</sup> For x - x' < 0 the relevant pole is at  $k = -\kappa$ ,<sup>16</sup> thus by using the residue theorem

<sup>&</sup>lt;sup>14</sup>One might remark that this Density of States needs to be understood in the distributive sense, since we left the integration away. This is a further reminder that the Density of States does only make sense in combination with an energy summation. (Just like the delta-distributions is not defined point by point, but only by its 'effect').

<sup>&</sup>lt;sup>15</sup>The integration contour is chosen to be a halfcircle of radius  $r \to \infty$ . A sufficient (though not necessary) condition for its disappearance is that the integrand times r goes towards 0 as r goes to infinity. Since in  $re^{ik\alpha} = re^{ire^{i\phi}\alpha} = re^{ir\cos(\phi)\alpha}e^{-r\sin(\phi)\alpha}$ ,  $\sin(\phi) > 0$  for  $\phi \in (0, \pi)$  and < 0 for  $\phi \in (0, -\pi)$ , and moreover an exponential decay always suppresses any polynom, so that the allegation holds. Notice, however, that the sign of the parameter  $\alpha$  changes the Green's function (which is not unique).

We assume x - x' < 0.

<sup>&</sup>lt;sup>16</sup>The square-root of any complex number is sign-conservative regarding the imaginary part. Thus  $\text{Im}(\kappa) > 0$  and Im(k) < 0. So only  $(\kappa + k)$  has a relevant zero point

$$\begin{aligned} G^{R}(x,x',E) &= -2\pi i \lim_{k \to -\kappa} \left( \frac{1}{\pi} \frac{e^{ik(x-x')}}{(\kappa-k)(\kappa+k)} (\kappa+k) \right) &= \\ &= -2\pi i \frac{1}{\pi} \frac{e^{-i\kappa(x-x')}}{2\kappa} &= \\ &= -i \frac{e^{-i\sqrt{(E+i\delta)2}(x-x')}}{\sqrt{(E+i\delta)2}} \end{aligned}$$

The Density of States follows:

$$\mathcal{N}(E) = \frac{1}{\pi\sqrt{2}} \frac{1}{\sqrt{E}}$$

## Free electron Green's-Function in 3D

The set of eigenfunctions and the corresponding eigenvalues are:

$$\psi_{\mathbf{k}}(r) = \frac{1}{\sqrt{L^3}} e^{i\mathbf{k}\mathbf{r}}, \quad E_{\mathbf{k}} = \frac{\mathbf{k}^2}{2} \quad \text{with} \quad \mathbf{k} = \begin{pmatrix} k_x \\ k_y \\ k_z \end{pmatrix} \quad \text{and} \quad \mathbf{r} = \begin{pmatrix} x \\ y \\ z \end{pmatrix}$$

Inserting these into the Green's function in spectral representation, and by choosing the system of reference to fulfil  $\mathbf{e}_z$  parallel to  $\mathbf{r} - \mathbf{r}'$ , one gets

$$\begin{aligned} G^{R}(x,x',E) &= \sum_{n} \frac{\psi_{n}(r)\overline{\psi_{n}(x')}}{E - E_{n} + i\delta} \approx \frac{L^{3}}{(2\pi)^{3}} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \mathrm{d}^{3}k \frac{\frac{1}{\sqrt{L^{3}}} e^{i\mathbf{k}\mathbf{r}} \frac{1}{\sqrt{L^{3}}} e^{-i\mathbf{k}\mathbf{r}'}}{E - \frac{\mathbf{k}^{2}}{2} + i\delta} = \\ &= \frac{1}{(2\pi)^{3}} \int_{0}^{2\pi} \mathrm{d}\phi \int_{0}^{\infty} \mathrm{d}k \underbrace{\int_{0}^{\pi} \sin(\theta) e^{ik} \underbrace{|\mathbf{r} - \mathbf{r}'|}_{ika} \cos(\theta) \mathrm{d}\theta}_{\frac{1}{2}} k^{2} \frac{1}{\underbrace{E - \frac{k^{2}}{2} + i\delta}_{\frac{1}{2}(\underbrace{\kappa}_{\sqrt{2(E + i\delta)}} - k)(\kappa + k)}} \end{aligned}$$

In order to solve the remaining k-integral by using the residue theorem one must change the borders from  $(0, \infty)$  to  $(-\infty, \infty)$ . This can be done the following way:

$$\int_0^\infty \frac{\mathrm{d}x \, xe^{ix}}{f(x^2)} - \int_0^\infty \frac{\mathrm{d}x \, xe^{-ix}}{f(x^2)} = \int_0^\infty \frac{\mathrm{d}x \, xe^{ix}}{f(x^2)} - \int_0^{-\infty} \frac{\mathrm{d}x \, xe^{ix}}{f(x^2)} = \int_0^\infty \frac{\mathrm{d}x \, xe^{ix}}{f(x^2)} + \int_{-\infty}^0 \frac{\mathrm{d}x \, xe^{ix}}{f(x^2)} = \int_{-\infty}^\infty \frac{\mathrm{d}x \, xe^{ix}}{f(x^2)} = \int_$$

Thus, one gets

$$G^{R}(x, x', E) = -\frac{1}{2\pi} \frac{e^{i\sqrt{2(E+i\delta)}|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|}$$

Which leads directly <sup>17</sup> to the density of states for a free electron

$$\mathcal{N}(E) = \frac{1}{\sqrt{2}\pi^2} \sqrt{E}.$$

## 2.2 Tight-binding (TB) method

Before going into detail of the implementation of the implementation of different dispersion relations into the DMFT code, we want to elaborate briefly on their general physical meaning. Since we are considering a solid material we demand discrete (lattice) periodicity of our potential  $\hat{V}(\mathbf{r}) = \hat{V}(\mathbf{r} + \mathbf{R})$ . Where **R** is a lattice vector. If electron interaction can be neglected, this also corresponds to a very useful property of our wave function:  $\psi_{\mathbf{k}}(\mathbf{r} + \mathbf{R}) = e^{i\mathbf{k}\mathbf{R}}\psi_{\mathbf{k}}(\mathbf{r})$  which is known as Bloch's theorem.

While these statements are quite general we will focus now on the tight binding assumption. If we think about our material as a grid of atoms far apart (we may think of a lattice constant of several centimetres), it becomes clear that the electrons are all closely bound to their respective atoms. This ('atomic') means that for such a material, an ideal choice of a basis would be a set of localised electronic orbitals. The energy-levels would simply be those of the isolated atoms. When we now shrink this huge interatomic spacing to a value comparable to the spacial extent of its wave function we expect our energy levels to be altered. These corrections to the isolated case, which come from the overlap of the wave functions are dealt with by the tight binding method.

**Eigenvalue problem** We have to solve the Schrödinger eigenvalue problem:

$$\hat{H}\psi = (\hat{T} + \hat{V})\psi = \epsilon\psi$$

In order to solve this equation we need to choose a basis. In principle we could choose any basis and we could work out the exact solution. However, the wave function lives in an infinite dimensional Hilbert space and this means we would have to consider an infinite linear combination of our basis elements. Since this is in practice not possible, we have to cut of our basis somewhere (at least when dealing with the eigenvalue problem numerically). This shows how essential the choice of an appropriate basis is. We want to have a basis which approximates the wave function already sufficiently for a small number of basis elements. We expect (for the tight binding assumption) the wave function to be similar to the case of the atomic limit. This would suggest to take the localised atomic orbitals directly as basis elements  $\tilde{\Phi}^c(\mathbf{r} - \mathbf{R})^{-18}$ . The problem with this simple choice would be, however, that the orbitals of different lattice sites are not orthogonal to each other.

<sup>&</sup>lt;sup>17</sup>after performing exactly the same steps as in one dimension and furthermore applying de L'Hôspital's rule.

 $<sup>^{18}</sup>$  where the index c gives the quantum numbers of the *core* orbitals (e.g. 1s, 2s, 2p, ...) which also gives us the band index.

Therefore we choose basis elements of the following form:

$$|\mathbf{k},c\rangle = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}} e^{i\mathbf{k}\mathbf{R}} \Phi_c$$

Where  $\Phi_c$  are the Wannier functions, which are in the tight binding case similar to the atomic wave functions  $\tilde{\Phi}^{c \ 19}$  (so we can still speak of localized functions), already satisfy Bloch's theorem and are moreover orthogonal to each other.

$$\langle \Phi^c(\mathbf{r}-\mathbf{R}) | \Phi^{c'}(\mathbf{r}-\mathbf{R'}) \rangle = \delta_{\mathbf{R},\mathbf{R'}} \delta_{c,c'}$$

Having chosen an appropriate basis, we can now formally expand our wave function with some unknown coefficients  $[a_c^{(k)}]$ :

$$\psi_{\mathbf{k}} = \sum_{c} a_{c}^{(k)} \left| \mathbf{k}, c \right\rangle$$

And we can rewrite our eigenvalue problem for our non interacting electrons in the following way:

$$\hat{H} |\psi_{\mathbf{k}}\rangle = \left(-\frac{\nabla^2}{2} + \hat{V}^{at} + \underbrace{\hat{V} - \hat{V}^{at}}_{\hat{W}}\right) |\psi_{\mathbf{k}}\rangle = \epsilon |\psi_{\mathbf{k}}\rangle \tag{4}$$

Hence, we split our potential into two parts:  $\hat{V}^{at}$ , which is simply the potential of a single atom at  $\mathbf{R}_0$  (the argument is applicable to any  $\mathbf{R}_0$  due to the discrete translational invariance of the system), and the whole rest  $\hat{W}$ . We can also see the term  $\hat{W}$  as a perturbation to the atomic limit. When we now project equation (4) onto  $\langle \Phi^c(\mathbf{r} - \mathbf{R}_0) | e^{-i\mathbf{k}\mathbf{R}_0}$  we get:

$$\begin{aligned} \epsilon(\mathbf{k})a_c &= E_c a_c + \sum_{c'} \left\langle \Phi_c(\mathbf{r} - \mathbf{R}_0) \right| \hat{W} \left| \Phi_{c'}(\mathbf{r} - \mathbf{R}_0) \right\rangle a_{c'} + \\ &+ \sum_{\mathbf{R} \neq \mathbf{R}_0, c'} e^{i\mathbf{k}(\mathbf{R} - \mathbf{R}_0)} \left\langle \Phi_c(\mathbf{r} - \mathbf{R}_0) \right| \hat{W} \left| \Phi_{c'}(\mathbf{r} - \mathbf{R}) \right\rangle a_{c'} \end{aligned}$$

Where  $E_c$  gives the energy of a electron state in the atomic limit. The second term only shifts the energy level due to the potential of the surrounding lattice sites. The third term takes into account that the wave functions overlap and that, therefore, an electron can 'hop' (or tunnel) from one lattice site to another. Hitherto we didn't use any approximation, but in order to get the solution, for an arbitrary one-particle external potential  $\hat{W}$ , we would still have to solve an infinite set of coupled equations to get the coefficients of our wave function. However, due to our educated basis choice we can hope that already a small number of basis elements already give a sufficient approximation.

<sup>&</sup>lt;sup>19</sup>They can be constructed from the core orbitals:  $\Phi^c(\mathbf{r} - \mathbf{R}) = \frac{1}{v_0} \int d\mathbf{k} e^{-i\mathbf{R}\mathbf{k}} \tilde{\Phi}^c_{\mathbf{k}}(r)$  where  $v_0$  is the volume of the first Brillouin zone. (see [11])

The simplest case is the single band approximation, where we assume (as the name already suggests) that there is only one band (e.g. that there is only one kind of atomic wave function to consider). In this case we don't need to determine the wave function coefficients and one simply gets as an energy:

$$\epsilon_c(\mathbf{k}) = E_c + \bar{W} - \sum_{\mathbf{R} \neq \mathbf{R}_0} t_{(\mathbf{R}, \mathbf{R}_0)} e^{i\mathbf{k}(\mathbf{R} - \mathbf{R}_0)}$$
(5)

Where  $\overline{W} = \langle \Phi_c(\mathbf{r} - \mathbf{R}_0) | \hat{W} | \Phi_{c'}(\mathbf{r} - \mathbf{R}_0) \rangle$  can be viewed as the energy correction in fist order perturbation theory in  $\hat{W}$ . The second term gives a contribution due to the overlap of the Wannier functions, considering the potential correction  $\hat{W}$  in which the matrix hopping elements appear:

$$t_{(\mathbf{R},\mathbf{R}_0)} = \langle \Phi_c(\mathbf{r} - \mathbf{R}_0) | \hat{W} | \Phi_{c'}(\mathbf{r} - \mathbf{R}) \rangle \tag{6}$$

This is the physical origin of all the specific tight binding dispersions, which will be considered in the rest of the thesis.

**Diagonalizing the tight binding Hamiltonian in second quantization:** In the limit of no interaction, the single band Hubbard Hamiltonian, for which our DMFT calculations will be performed, coincides with the tight-binding single band Hamiltonian:

$$\hat{H} = \sum_{\mathbf{R}_i, \mathbf{R}_j} \sum_{\sigma} t_{(i,j)} \hat{c}^{\dagger}_{\sigma,i} \hat{c}_{\sigma,j} = \sum_{\mathbf{k},\sigma} \epsilon_{\mathbf{k}} \hat{n}_{\mathbf{k},\sigma}$$
(7)

The creation (annihilation) operators create (or destroy) electrons in states given by localised Wannier functions. Comparing eq. (5) and eq. (7) we see the similarity. (The only difference being the zero offset in eq.(7))

We can diagonalize our problem and obtain its eigenvalues, which results in the *dispersion relation* ( $\epsilon_{\mathbf{k}}$ ) either by directly Fourier-transforming the creation and annihilation operators, or completely without second quantization as shown before. These two ways of diagonalizing the Hamiltonian are equivalent. In the previous calculation the wave-function is expanded into Wannier functions, which is the same set of functions chosen for a basis to map the many-body-problem on. This needs to be done when properly deriving the Hubbard-Hamiltonian (see [4]).

We shall quickly sketch the approach starting from the Hamiltonian in second quantization. The Fourier-transformation of the operators can be seen as an 'ansatz', which works because we are only dealing with the kinetic term of the Hubbard-model:

$$c_{\mathbf{k},\sigma} = \frac{1}{\prod_{i=1,\dots,d}\sqrt{N_i}} \sum_{\mathbf{R}} e^{i\mathbf{k}\mathbf{R}} c_{\mathbf{R},\sigma}$$
$$c_{\mathbf{k},\sigma}^{\dagger} = \frac{1}{\prod_{i=1,\dots,d}\sqrt{N_i}} \sum_{\mathbf{R}} e^{-i\mathbf{k}\mathbf{R}} c_{\mathbf{R},\sigma}^{\dagger}$$

Where  $N_i$  is the lattice size (or number of atoms) in the *i*th direction the cubical lattice. We use the fact that  $\frac{1}{N_i} \sum_{R_i=0}^{N_i-1} e^{R_i(k_i-k'_i)} = \delta_{k_i,k'_i}$  as long as all wave-numbers are  $k_i = 2\pi/N * n$  with  $n \in \mathbb{Z}$ .<sup>20</sup> After specifying and inserting the hopping terms we get the dispersion relation given in table 3. We shall use the following convention for the hopping terms  $t(\mathbf{i}, \mathbf{j})$ :

variable	name	mathematical description
$t_{t'}$	NN <sup>21</sup> direct hopping term	$t = t(\mathbf{i}, \mathbf{i} \pm \hat{e_j})$ $t' = t(\mathbf{i}, \mathbf{i} \pm \hat{e_i} \pm \hat{e_i}) \text{ with } i \neq k$
t''	$NNN^{22}$ diagonal hopping term	$t'' = t(\mathbf{i}, \mathbf{i} \pm \hat{e_j} \pm \hat{e_k}) \text{ with } j \neq k$ $t'' = t(\mathbf{i}, \mathbf{i} \pm \hat{e_j} \pm \hat{e_k} \pm \hat{e_l}) \text{ with } j \neq k \neq l \neq j$
$t^{\prime\prime\prime}$	NNN direct hopping term	$t = t(\mathbf{i}, \mathbf{i} \pm 2\hat{e_j})$

Where  $\hat{e_j}$  is the unit vector in the *j*th direction, and all Roman indices can go from 1 to d.

For a visualization of the hopping terms consider figure 4.

Now we can derive the energy dispersion for various hopping terms from equation 8 which is the same as equation 5, except for that the offset energy was set to zero.

$$\epsilon(\mathbf{k}) = -\sum_{\mathbf{R}\neq\mathbf{R}_0} t_{(\mathbf{R}_0,\mathbf{R})} e^{i\mathbf{k}(\mathbf{R}-\mathbf{R}_0)}$$
(8)

Assuming orbital<sup>23</sup> and crystal symmetry the sum can be split into terms with one leading constant each (e.g. in four terms with t,t',t'',t''' for the three dimensional case, as can be seen in Fig. 3).

<sup>&</sup>lt;sup>20</sup> in fact  $k_i = 2\pi/(N*a)*n$  with the lattice constant a which was set to 1

<sup>&</sup>lt;sup>21</sup>where NN stands for nearest neighbour

 $<sup>^{22}{\</sup>rm where}$  NNN stands for next nearest neighbour

<sup>&</sup>lt;sup>23</sup>Only one type of orbital with the same symmetry as the crystal (e.g. s-level) was considered.

variable name	energy dispersion
$\epsilon_{1D}$	$-2t\cos(ka) - 2t'''(\cos(2ka))$
$\epsilon_{2Dt,t',t'''}$	$-2t(\cos(k_x a) + \cos(k_y a)) - 4t'\cos(k_x a)\cos(k_y a) - 2t'''(\cos(2k_x a) + \cos(2k_y a))$
$\epsilon_{3Dt,t',t'',t'''}$	$-2t(\cos(k_xa) + \cos(k_ya) + \cos(k_za) -$
	$-4t'(\cos(k_xa)\cos(k_ya) + \cos(k_ya)\cos(k_za) + \cos(k_xa)\cos(k_za)) -$
	$-8t''\cos(k_xa)\cos(k_ya)\cos(k_za) -$
	$-2t'''(\cos(2k_xa) + \cos(2k_ya) + \cos(2k_za))$

Fig. 3 Summary of energy dispersions derived from the tight binding model  $\mathbf{F}$ 



Fig. 4 Illustration of the different possible hopping terms. The same nomenclature was also used in the two-dimensional case were of course only t, t' and t''' can be unequal to 0.

Now that we have calculated the dispersion relations, we can also immediately derive the Fermi surfaces corresponding to the various cases. The Fermi surface can formally be seen as the abstract boundary in reciprocal space, separating the occupied k-states of the system from the unoccupied (at the limit of zero temperature). The defining equation of the Fermi surface for a non-interacting system is:

$$\epsilon(\mathbf{k}) - \mu = 0$$

Where  $\mu$  is the chemical potential. The specific shape of the Fermi surface is extremely useful in predicting thermal, electrical, magnetic and optical properties of a material. We see that the above equation is the same as the argument of the delta distribution occurring in the calculation of the DOS, for the energy  $\mu$ . Thus, we can conclude that a non zero value of the DOS at the chemical potential is equivalent to the existence of a Fermi surface. The existence of a Fermi surface tells us that the concerning material is a conductor, rather than an band insulator. In Figure 5 the Fermi surface is shown in the case of a two-dimensional system. In Figure 6 for a three-dimensional system. One may note that the chemical potential  $\mu$  can be calculated from the filling of the system n by finding a solution to:  $n = \int d\epsilon \mathcal{N}(\epsilon) \frac{1}{e^{\beta(\epsilon-\mu)}+1}$ .<sup>24</sup> So the chemical potential is usually determined from a fixed filling condition (number of electrons per unit volume) of the system. In the following plots, however, the filling is not fixed to a certain value, but the chemical potential is set to zero (for the left plot), or to t (for the right plot).



Fig. 5 The Fermi-surfaces are defined by the condition  $\epsilon_{\mathbf{k}} = \mu$ . Here they are plotted for different maximal energies  $(=\mu)$  and hopping ratios in the two-dimensional case. The left figure shows that with t' > 0 half filling is no longer realized at  $E_{max} = \mu = 0$ . Notice, furthermore, that the collared area gives the filling.

We find from the Fermi surface plots that in some cases there are vectors  $\mathbf{Q}$  that fulfil  $\epsilon_{\mathbf{k}} = \epsilon_{\mathbf{k}+\mathbf{Q}}$  for large parallel parts of the Fermi surface. These  $\mathbf{Q}$  vectors are called *nesting vectors* and are especially interesting if terms of the form  $g(\mathbf{k}, \mathbf{Q})\hat{c}^{\dagger}_{\mathbf{k}+\mathbf{Q}}\hat{c}_{\mathbf{k}}$  appear

<sup>&</sup>lt;sup>24</sup>Where  $\beta$  is the inverse temperature.

in the Hamiltonian.<sup>25</sup> For  $\mu = t' = 0$  we find  $(\pi, \pi)^T$  to be a nesting vector, whereas for the other cases shown on figure 5 this no longer holds to be true. This means that the introduction of a further hopping term destroyed the nesting property.



Fig. 6 Three dimensional Fermi surfaces for different and hopping ratios. The chemical potential  $\mu$  is set to zero for all plots in this figure.

In three dimensions we find for  $\mu = t' = t'' = t''' = 0$  the nesting vector  $\mathbf{Q} = (\pi, \pi, \pi)^T$ (which corresponds to the upper left plot of Fig. 6) Since adding  $\pi$  in the argument of each cosine term only flips the sign, which doesn't matter because  $\epsilon_{\mathbf{k}_f+\mathbf{Q}} = -\epsilon_{\mathbf{k}_f} = -0 =$  $0 = \epsilon_{\mathbf{k}_f}$ . We also find that for an additional NN-diagonal or a NNN-hopping term (t' or t'''), unequal to zero,  $(\pi, \pi, \pi)^T$  is no longer a nesting vector, whereas the introduction of NNN-diagonal term (t'') does not destroy the nesting properties of  $(\pi, \pi, \pi)^T$ .

## 2.3 TB DOS in finite dimensions

Since the DMFT single band calculations include the effect finite dimensional lattice only in term of its corresponding DOS, here we outline in more details DOS features of TB dispersions. For the tight binding model it is in general not possible to derive explicit

<sup>&</sup>lt;sup>25</sup>As an example we may recall that the denominator of the Lindhard function (in the limit of  $\omega \to 0$  disappears for the nesting vectors.)

analytic expressions for the DOS. The details how we dealt with this numerically are given in chapter 3.

After having calculated the DOS one can try to understand some of its parameter dependent characteristics. To derive those, a vast amount of plots were made, which shall not be given here. We merely want to list the found features. In particular, we will look where *Van Hove singularities* appear since those give hints where phase transitions and interesting low-temperature phenomena might occur.

We recall that in the literature by the term Van Hove singularities either actual singularities of  $\mathcal{N}(\epsilon)$  are meant or singularities of the slope of the  $\mathcal{N}(\epsilon)$ . The later are visible as cusps in the DOS. Both kinds are of course integrable in energy, (since the integral over the DOS must yield a constant).

From now on we measure energies in units of the direct NN hopping term t which will be set to one (which is equivalent to rescaling the energy and additional parameters e.g. further hopping terms regarding to t).

Two-dimensional DOS with only nearest neighbour direct hopping (only t): As we see in figure 7 the DOS is symmetric around E = 0, reflecting the perfect particle-hole-symmetry of the problem, and that there is one (real) Van Hove Singularity at E = 0. There are non-zero values only from -4 to 4 which are also the extremal values of the dispersion relation  $\epsilon_{\mathbf{k}} = -2t[\cos(k_x) + \cos(k_y)]$ .



Fig. 7 DOS for the two dimensional tight binding model with only nearest neighbour direct hopping. The small spikes should not occur and can be eliminated by 'smoothing' the function. (This is done automatically when calculating the spectral function)

Two-dimensional DOS with nearest neighbour direct and diagonal hopping (t and t'): If t' > 0 is varied one can see that:

• t' moves the Van Hove singularity (for increasing t' from left to right, see figure 8)

• There is a symmetry regrading the sign of t':  $\mathcal{N}(\epsilon, t') = \mathcal{N}(-\epsilon, -t')^{26}$  (which also predicts the axis symmetry for  $\mathcal{N}$  if t' = 0)

• for big t' (approximately starting at t' > 0.5t) a Van Hove singularity of the slope of the DOS also appears. (see figure 8)

We recognise that:

• t"'>0 leads to an additional real singularity at negative energies (see figure 9)

• t"'>0 can also lead to a further discontinuity of  $\mathcal{N}(\epsilon)$  's first derivative which for higher t"' tends to be localized at smaller energies compared to smaller t"' values. (see figure 9)



Fig. 8 DOS for the two-dimensional tight binding Fig. 9 DOS for the two dimensional tight binding model with NN direct hopping as well as NN diagonal hopping. We find that the main Van Hove singularity is located at higher energies compared to the case with t' = 0 and that there is a further singularity of the slope of the DOS.

model with NN as well as NNN direct hopping. We find a further real Van Hove singularity and also two singularities of the slope of the DOS.

Three-dimensional case with nearest neighbour direct hopping only: For more than two dimensions true singularities in  $\mathcal{N}(\epsilon)$  can no longer appear in the DOS, but one can still find singularities of the slope of the DOS. The shape of the DOS for the tight binding model in three dimensions with only nearest neighbour direct hopping (so t' = t'' = t''0) can be seen in figure 10.

<sup>&</sup>lt;sup>26</sup> showing this relation is not trivial since an analytical expression for  $\mathcal{N}(\epsilon, t')$  can not be found



Fig. 10 DOS for the three dimensional tight binding model with only nearest neighbour direct hopping.

Three-dimensional case with various hopping terms: After a variation of t' one can see that:

- For small values of t' the DOS gets slightly deformed as seen in figure 11.
- For larger values a further Van Hove singularity of the slope of the DOS appears.

If only t'' is changed we find:

- The DOS remains symmetric.
- Two further Van Hove Singularities in the slope of  $\mathcal{N}(\epsilon)$  appear as seen in figure 12.



model with NN direct hopping as well as NN diagonal hopping. We find that the shape of the DOS is deformed in an asymmetric way.



Fig. 11 DOS for the three dimensional tight binding Fig. 12 DOS for the three dimensional tight binding model with NN as well as NNN diagonal hopping (t''). The DOS stays symmetric, but we find further Van Hove cusps.

If we change only t''' the DOS shows various Van Hove singularities as show in figure 13.



Fig. 13 DOS for the three dimensional tight binding model with NN (t) as well as NNN direct hopping (t'''). There appears a variety of further VH cusps.

**Remarks on the Bandwidth** For a simple dispersion relation containing only terms with direct NN hopping t the (non-interacting) DOS is symmetric with respect to E = 0(which is the Fermi energy in our case). In this case the bandwidth can be easily calculated as a simple sum over terms of the form  $\cos(k_i)$  which give +1 as a maximum, and -1 as a minimum.

When other terms also enter the energy minimum is still trivial to calculate (since  $(+1)^n = +1$  with  $n \in \mathbb{N}$  the minimum is at  $\mathbf{k} = (0, 0, ..)^T$ ), but the maximum is no necessarily still at  $(\pi, \pi, ..)$ . Up to a certain value of t' the changes cancel each other and the bandwidth remains unchanged, but starting at a critical ratio the maximum stops changing with the same rate as the minimum and the bandwidth increases. These characteristic ratios are written down in figure 14.

Moreover, also for t''' (NNN-hopping) this leads to a similar characteristic. The  $-8t'' \cos(k_x)\cos(k_y)\cos(k_z)$  term (again for 3D) is easier to deal with, since  $(-1)^3 = -1$ . This contribution leads to a linear broadening.

$\operatorname{nomenclature}$	relevant variable	critical value 2D	critical value 3D
NN diagonal	t'	0.5	0.25
NNN direct	t" '	0.25	0.25

Fig. 14 Separated critical hopping values where the energy maximum stops being at BZ corners

Summarizing, in general, with increasing hopping parameter, the bandwidth gets bigger, except for the NN diagonal (t') hopping and the NNN direct hopping (t''') contribution with the corresponding parameters smaller than a certain critical value, where the bandwidth stays constant, before it starts increasing. (Which can be seen in figure 15 for a variation of t' in three dimensions.) This means that starting at some critical ratio the energetic maximum is no longer at  $(\pm \pi, \pm \pi, ..)^T$  but at some other point within the first BZ.

#### 27



Fig. 15 Bandwidth and energy-borders for a 3D dispersion relation with only t and t'

The bandwidth is an important result which can be used for evaluating renormalization effects, comparing energy scales and many further applications. Therefore it is calculated in two dedicated subroutines Bandwidth2(minb,maxb,En) for 2D or Bandwidth(minb,maxb,En) for 3D.

<sup>&</sup>lt;sup>27</sup>The 'critical values' only remains correct for some specific contributions (all hopping terms are 0 except t and one further). When dealing with all hopping terms at once the critical values change, but the mentioned general behaviour remains unchanged.

#### 2.4 TB DOS in infinite dimensions

Since DMFT becomes exact in the limit of infinite dimensions, it is sometimes useful to perform calculations directly in this limit, to compare with results of the finite dimensional case, where DMFT works only as an approximation. Hence in this sections, we will provide a short derivation of the TB-dispersion and the corresponding terminology in  $d \to \infty$ . Following Ref. [7, 1] we have, first of all, to define our  $d \to \infty$  approach. Our goal is to rescale the Hamiltonian in such a way that neither the kinetic part nor the potential part disappears or diverges. Otherwise we would only get trivial results. We can define a rescaled hopping parameter  $t^*$  by:

$$t := \frac{t^*}{\sqrt{2d}}$$

 $t^*$  turns out to be constant. If we consider a hyper-cubic lattice, our unit cell basis vectors are of the form:  $\mathbf{e}_n = (0, .., 0, 1, 0, .., 0)^T$  We define the hopping amplitudes as

$$t_{ij} = t(\mathbf{R}_i - \mathbf{R}_j) = \begin{cases} t \text{ if } \mathbf{R}_i - \mathbf{R}_j = \pm \mathbf{e}_n \\ 0 \text{ otherwise} \end{cases}$$

and get the dispersion relation:  $\epsilon_{\mathbf{k}} = -2t \sum_{i=1}^{d} \cos(k_i)$ . Now we can derive the Fourier transformed DOS:

$$\phi(s) = \int_{-\infty}^{\infty} d\epsilon \, e^{is\epsilon} \mathcal{N}(\epsilon) = \int d\epsilon e^{is\epsilon} \int_{-\pi}^{\pi} \frac{d^d k}{(2\pi)^d} \delta(\epsilon - \epsilon_{\mathbf{k}}) = \int \frac{d^d k}{(2\pi)^d} e^{is\epsilon_{\mathbf{k}}} = \int \frac{d^d k}{(2\pi)^d} e^{-i2st(\cos(k_1) + \cos(k_2) + \dots + \cos(k_d))} = \left(\int \frac{dk}{(2\pi)} e^{-i2st\cos(k)}\right)^d = \int (J_0(2st))^d$$

In the last step we used the Bessel-function of the first kind[12]:  $I_0(x) := J_0(ix) = 1/\pi \int_0^{\pi} \exp[x \cos(\theta)] d\theta$ .

By using the power-series expansion  $I_0(x) = \sum_{m=0}^{\infty} \frac{1}{(m!)^2} \left(\frac{x}{2}\right)^{2m}$  we get:

$$\phi(s) = \left[1 - \frac{t^{*2}s^2}{2d} + O(d^{-2})\right]^d = \exp\left[-\frac{t^{*2}s^2}{2d} + O(d^{-1})\right]$$

After letting the number of dimensions go to infinity and applying the inverse Fouriertransformation one can finally write the DOS as:

$$\mathcal{N}(\epsilon) = \tilde{\phi} = \frac{1}{\sqrt{2\pi}|t^*|} e^{-\frac{\epsilon^2}{2t^{*2}}}$$

Notice that the whole derivation looks very much like the central limit theorem (CLT) proof. We could have applied the CLT directly and derived the same result with less effort [7, 1].

In figure 16 the DOS for various dimensions with merely nearest neighbour direct hopping is shown. Moreover, the limit of infinite dimensions (Gaussian) is also given for comparison. We see that for small dimensions (e.g one and two) there is a rather big discrepancy, but for three dimensions there already appears a clear similarity to the infinite dimensional case. This suggests that an infinite dimensional DOS can be a reasonable approximation to a three dimensional case. Looking at higher dimensions we find that the convergence towards the Gaussian happens fast, (e.g. for six dimensions there is hardly any difference visible any more).



Fig. 16 Convergence of the DOS for higher dimensions towards a Gauss distribution. (finite dimension with renormalized t ... blue;  $\mathcal{N}_{\infty}$ ... purple). See also appendix of [1].

#### 2.4.1 Effect of infinite Dimensions on the Green's function

In order to find the Green's function's behaviour at infinite dimensions, one should start considering the number of 'neighbours' of a given site. The one-particle-density-matrix element squared  $|g_{ij,\sigma}^0|^2$  is proportional to the 'hopping' probability from site i to j, and the sum of these probabilities over all 'neighbours' must yield a constant to avoid ending in a trivial uncorrelated situation in  $d \to \infty$ . Moreover, the dimensional scaling is preserved when calculating the one-particle Green's function and also when applying the Fourier-transformation in order to get  $G_{ij,\sigma}^0(\omega)$ .

Therefore if  $n_b(d)$  is the number of neighbours then the  $G^0_{ij,\sigma}(\omega)$  needs to be proportional to  $1/\sqrt{n_b(d)}$ .

If we try to find the number of relevant neighbours of one lattice site which can be reached by s steps of the lattice constant in d dimensions, it seems rather straightforward to move away from the geometrical view towards an abstract vector notation. However, in order to understand what s steps in d dimensions means we have presented a specific case in figure 17 where d=3 and s=3.



Fig. 17 One neighbour for d=3 and s=3

Here the number of steps is three (equals the number of red arrows), therefore this neighbouring site can be reached viva the vector:

$$\begin{pmatrix} -1\\1\\1 \end{pmatrix},$$

where all lattice constants are set to 1.

This formalism can be generalized: If we want to know the number of neighbours in d dimensions which can be reached by exactly s steps, each one can be written as a vector with s entries unequal to 0 and d-s equal to 0. If we would write each of them down, we would get :  $(\pm 1, \pm 1, ..., \pm 1, 0, ..., 0)^T$ ,  $(\pm 1, ..., \pm 1, 0, \pm 1, 0, ..., 0)^T$ , ... Therefore there can be  $\binom{d}{s}$  different vectors by ordering the entries differently. Moreover, since each sign can be changed separately we get a total number of neighbours of:

$$n_b(d,s) = 2^s \binom{d}{s}$$

In figure 18 we see that the number of neighbours for d=3 and s=3 is  $2^{3} \binom{3}{3} = 8$  which is correct for the example.



Fig. 18 All neighbours for d=3 and s=3

This result can be used to derive, more rigorously, a general formula for the dimensional scaling of the one particle Green's-function:

$$n_b(d,s) = 2^s \binom{d}{s} = 2^s \frac{d!}{s!(d-s)!} \approx \mathcal{O}(d^s)$$

And therefore:

$$G^0_{ij,\sigma} \approx \mathcal{O}\left(\frac{1}{d^{s/2}}\right)$$

Which is equivalent to Eq. (21) in Dieter Vollhard's chapter in 'LDA+DMFT in strongly correlated materials' [1, 1.9] where also all lattice constants are 1 and the applied metric is the Manhattan metric:

$$G_{ij,\sigma}^{0} \approx \mathcal{O}\left(\frac{1}{d^{||\mathbf{R}_{i}-\mathbf{R}_{j}||/2}}\right)$$
(9)

Equation 9 was one of the key insights into DMFT since, by applying the standard Feynman diagramatic rules, it implies that the self-energy becomes local in the limit of infinite dimensions.

## 3 Implementation in code and technical details

Within this thesis three major cases are treated: two, three and infinite-dimensional lattices. The lattice always was assumed to be cubic. The basic approach always stayed the same:

- Calculating the non-interacting DOS  $\mathcal{N}(\omega)$  from the dispersion relation  $\epsilon(\mathbf{k})$ .
- Determining the local Green's function  $G_{loc}(i\omega_n)$  as well as the self-energy  $\Sigma(i\omega_n)$  by the DMFT self-consistency cycle.
- After convergence: Using the imaginary part of the Green's function as well as the non-interacting DOS to calculate the spectral function  $A(\omega)$ .<sup>28</sup>

## 3.1 Noninteracting DOS

There are several ways to calculate the DOS. One is to define a delta sequence e.g. a Lorentz curve  $\frac{1}{\pi} \frac{\epsilon}{x^2 + \epsilon^2}$ , but since the limit of  $\epsilon \to 0$  can not be implemented exactly in FORTRAN a small discretization- dependent value of  $\epsilon$  needs to be chosen. (e.g. 4 'energy-steps'). One obtains the following DOS in two dimensions for t' = t''' = 0 as can be seen in figure 19.

<sup>&</sup>lt;sup>28</sup>The spectral function is also refereed to as the 'interacting density of states'



Fig. 19 Density of states calculated numerically with a Lorentz curve in 2D with only NN hopping.

The DOS in figure 19 looks smeared out because of the finite  $\epsilon$  choice. For the same reason, the Van Hove singularity is not logarithmically diverging (as it should be), but has a finite value. Aimed to get more accurate results, one can solve a part of the integral 'by hand'. For the two and the three dimensional cases integrals over a Dirac-delta-'function' have to be solved. Specifically, the innermost integrations can be dealt with analytically, if the roots of the deltafunction's argument are known.<sup>29</sup>

In two dimensions: The following equation should be solved:

$$\epsilon(\mathbf{k}) = -2t(\cos(k_x) + \cos(k_y)) - 4t'\cos(k_x)\cos(k_y) - 2t'''(\cos(2k_x) + \cos(2k_y)) = \epsilon$$

Because of the  $\cos(2k_i)$  terms it is a quadratic equation. However, if there is no NNN-hopping (t''' = 0), the solution is simple:

$$k_x = \pm \cos^{-1} \left( -\epsilon/2 - \frac{t \cos(k_y)}{2t' \cos(k_y) + t} \right)$$

For a  $t''' \neq 0$ , we can follow exactly the same procedure. If we use  $\cos(2k_x) = 2\cos^2(k_x) - 1$ , we get a quadratic equation for  $\cos(k_x)$  which we can solve easily.

$$k_x = \pm \cos^{-1} \left( \frac{-t - 2t' \cos(k_y)}{4t'''} \pm \frac{\sqrt{(t + 2t' \cos(k_y))^2 - 4t'''(\epsilon + 2t \cos(k_y) - 2t''')}}{4t'''} \right)$$

 $<sup>^{29}</sup>$ Without any restriction the innermost integration was chosen to be over  $k_x$ 

Note the division by t''' in the result, which could be eliminated for  $t''' \to 0$  either by using the de l'Hospital rule or by distinguishing different cases like we did. Since  $\cos(-k_x) = \cos(k_x)$  we get in total four solutions.

Now we can rewrite the innermost integral  $\int_{-\pi}^{\pi} dk_x \, \delta(\epsilon - \epsilon_{\mathbf{k}})$ : <sup>30</sup>

$$\sum_{i} \frac{\theta(k_x + \pi)\theta(\pi - k_x)}{|4t'\sin(k_x)\cos(k_y) + 2t\sin(k_x) + 4t'''\sin(2k_x)|} \Big|_{k_{x_i}}$$

Where the sum runs over all roots of  $\epsilon_{\mathbf{k}} - \epsilon$ .

#### Algorithmic details:

The roots are calculated in the subroutine NstDos2, the absolute value of the partial derivative is calculated in the function AAble2,

the full density of states is calculated in the subroutine dos2C which uses the subroutine Den02D (which calculates the DOS only for a specific energy).

In three dimensions: Now we have to solve:

$$\epsilon(\mathbf{k}) = - 2t(\cos(k_x) + \cos(k_y) + \cos(k_z)) - 4t'(\cos(k_x)\cos(k_y) + \cos(k_y)\cos(k_z) + \cos(k_x)\cos(k_z)) - \\ - 8t''\cos(k_x)\cos(k_y)\cos(k_z) - 2t'''(\cos(2k_x) + \cos(2k_y)) = \epsilon$$

The simplest way to implement the result in the program is again to distinguish the two cases. For t''' = 0 we get:

$$k_x = \pm \cos^{-1} \left( \frac{-\epsilon - 4t' \cos(k_y) \cos(k_z) - 2t \cos(k_y) - 2t \cos(k_z)}{2(4t'' \cos(k_y) \cos(k_z) + 2t' \cos(k_y) + 2t' \cos(k_z) + t)} \right)$$

And for  $t''' \neq 0$  we again can bring the equation to the form  $a \cos^2(k_x) + b \cos(k_x) + c = 0$ and solve it by using the quadratic solution formula. With:

$$a = 4t'''$$
  

$$b = 4t'(\cos(k_y) + \cos(k_z)) + 8t''\cos(k_y)\cos(k_z) + 2t$$
  

$$c = \epsilon + 2t(\cos(k_y) + \cos(k_z)) + 4t'\cos(k_y)\cos(k_z) + 2t'''(\cos(2k_y) + \cos(2k_z) - 1)$$

So we get again four solutions:

$$k_x = \pm \cos^{-1}\left(\frac{-b \pm \sqrt{b^2 - 4ac}}{2a}\right)$$

Now we can eliminate the innermost integral as:

$$\sum_{i} \frac{\theta(k_x + \pi)\theta(\pi - k_x)}{|4t'(\sin(k_x)\cos(k_y) - \sin(k_x)\cos(k_z)) + 8t''\sin(k_x)\cos(k_y)\cos(k_z) + 2t\sin(k_x) + 4t'''\sin(2k_x)|} \Big|_{k_{x_i}}$$

<sup>&</sup>lt;sup>30</sup>Of course one can insert the roots and evaluate the trigonometric function e.g. by using the identity:  $\sin(\cos^{-1}(x)) = \sqrt{1 - \cos^2(\cos^{-1}(x))} = \sqrt{1 - x^2}$ , but the result still wouldn't not look very nice.

The remaining integral can not be solved analytically in general. Therefore the remaining integrations were performed numerically by using the Simpson algorithm.

However, for only NN hopping we can calculate the DOS quite simply by Fourier-transformation. In d dimensions we get:

$$\mathcal{N}_d(\epsilon) = \frac{1}{2\pi} \int e^{is\epsilon} \left( J_0(2ts) \right)^d \mathrm{d}s$$

Algorithmic details:

The roots are calculated in the subroutine NstDos,

the absolute value of the partial derivative is calculated in the function AAble, the full density of states is calculated in the subroutine dos3C, which uses the subroutine DenO3D (calculates the DOS only for a specific energy)

In one and two dimensions this yields a rather compact expression:

$$\mathcal{N}_1(\epsilon) = \frac{\sqrt{\frac{2}{\pi}}(\theta(-\epsilon-2)-1)(\theta(\epsilon-2)-1)}{\sqrt{2\pi}\sqrt{4-\epsilon^2}}$$
(10)

$$\mathcal{N}_2(\epsilon) = \frac{K\left(1 - \frac{\epsilon^2}{16}\right)\left(\theta(-\epsilon - 4) - 1\right)\left(\theta(\epsilon - 4) - 1\right)}{\left(\sqrt{2}\pi^{3/2}\right)\sqrt{2\pi}}$$
(11)

Where t = 1 and K is the 'complete elliptic integral' of the first kind[12].

In infinite dimensions: For infinite dimensions, following Ref. [7], the DOS does not need to be calculated numerically, since by using the central limit theorem, and by rescaling the hopping term  $t \to t^*/\sqrt{2d}$  (with  $t^*$  an arbitrary constant which defines the energy-scale, and d the dimensions) we can write it analytically as:

$$\mathcal{N}_{\infty}(\omega) = \frac{1}{\sqrt{2\pi}t^*} e^{-\frac{1}{2}\left(\frac{\omega}{t^*}\right)^2}$$

Algorithmic details:

The noninteracting DOS for the infinite dimensional case is calculated in the subroutine dosInf.

## 3.2 DMFT Self-Consistency-Cycle:

For the sake of clarity, let's briefly revisit the, already in the introduction mentioned, self-consistency cycle. (See Fig. 20)

The central quantity within DMFT is the local one-particle Green's function given by:

$$G_{ij}(\tau) = -\langle T_{\tau}c_i(\tau)c_j^{\dagger}(0)\rangle$$
(12)

where as  $T_{\tau}$  is the time-ordering operator. It is especially useful for numerical purposes to define the Matsubara Green's function  $G_{\alpha\beta} = (i\omega_n)$ :

$$G_{ij}(i\omega_n) = \int_0^\beta \mathrm{d}\tau \, G_{ij}(\tau) \, e^{i\omega_n \tau}$$

where  $\beta$  is the inverse temperature, and  $\omega_n = \pi/\beta(2n+1)$  are the Matsubara frequencies for fermions.

One of the key properties of DMFT was that in infinite spatial dimensions the electronic self-energy becomes local. [6] Hence, one can use this fact to map, for high spatial dimension, our lattice model onto a single impurity site embedded in a bath of surrounding non-interacting particles. [7] Hence, one can map the original lattice problem onto the single impurity Anderson model (for which there are quite a few efficient numerical solutions available like the Hirsch-Fye continuous time Quantum Monte-Carlo Method, Exact Diagonalization<sup>31</sup>, ...). We only have to determine the 'appropriate' Anderson impurity model. This is done by imposing the equality of the local DMFT Green's function of our lattice  $G_{loc}(\omega)$  and the AIM Green's function  $G_{AIM}(\omega)^{-32}$ . So our self-consistency condition is:

$$G_{AIM}(\omega) = G_{loc}(\omega)$$

<sup>&</sup>lt;sup>31</sup>For all the calculations the exact diagonalization (ED) was used within this thesis.

<sup>&</sup>lt;sup>32</sup>from a practical point of view one can also check for convergence by comparing the selfenergies  $\Sigma(\omega)$ 



Fig. 20 Illustration of the self-consistency cycle of the dynamical mean field theory.. The bottleneck of the algorithm is the method of solving the Anderson impurity model. This is done, in this thesis, by Exact Diagonalisation (ED) (reproduced from [8])

We briefly recall here the definitions of the local Green's-function  $G_{\sigma}(i\omega_n)$  the dynamical mean field  $\mathcal{G}_{\sigma}(i\omega_n)$  and the local selfenergy  $\Sigma_{\sigma}(i\omega_n)$ : (e.g. from [1])

$$G_{\sigma}(i\omega_n) = \langle c_{\sigma}(i\omega_n)c^{\dagger}_{\sigma}(i\omega_n) \rangle_{\mathcal{A}[\mathcal{G}]}$$
(13)

$$\Sigma_{\sigma}(i\omega_n) = \mathcal{G}_{\sigma}(i\omega_n)^{-1} - G_{\sigma}(i\omega_n)^{-1}$$
(14)

$$G_{\sigma}(i\omega_n) = \int \mathrm{d}\epsilon \frac{\mathcal{N}(\epsilon)}{i\omega_n + \mu - \Sigma_{\sigma}(i\omega_n) - \epsilon}$$
(15)

In the explicit case of an ED-impurity solver, such as that used in this thesis, the self-consistency is imposed in the following way:

Fist we need an initial dynamical mean field  $\mathcal{G}_0$  (or Weiss function), which can be calculated by using equation (16),

$$\mathcal{G}_0(i\omega_n)^{-1} = i\omega_n + \mu - \int \mathrm{d}\omega' \frac{\Delta(\omega')}{i\omega_n - \omega'} \tag{16}$$

where the conduction bath density of states  $\Delta(\omega)$  is yet unknown, but can be approxi-

mated by a discrete version (17).

$$\mathcal{G}_0^{n_s}(i\omega_n)^{-1} = i\omega_n + \mu - \sum_{p=2}^{n_s} \frac{V_p^2}{i\omega_n - \tilde{\epsilon}_p}$$
(17)

there  $V_p$  and  $\tilde{\epsilon}_p$  (corresponding respectively to **tpar** and **eps** as program variables) are the Anderson parameters of the discretized bath of the ED algorithm. Equation (17) shows the meaning of the Anderson parameters in the algorithm. They approximate the bath DOS by  $n_s - 1$  delta distributions with the partial spectral weight  $V_p^2$  at the energies  $\tilde{\epsilon}_p$ .

After having approximated the Weiss function, with a discrete electronic bath, one can *diagonalize* the  $n_s$  orbital AIM Hamiltonian (see for instance [13]) *exactly*, and calculate the Green's function (see equation 13).

Then by using the Dyson equation (14), we calculate the self energy from the Anderson impurity Green's function and the old dynamical mean field. Finally, we use the selfconsistency equation (15) to calculate the 'new' Green's function and by applying the Dyson equation (14) again be get the new dynamical mean field. This new Weiss function  $\mathcal{G}_0$  is then approximated by a function  $\mathcal{G}_0^{n_s}$  with a new set of Anderson parameters  $V_p, \tilde{\epsilon}_p$ .

The procedure repeats until convergence is reached.

In many cases the iterative procedure has only one stable solution and converges independently of the initial conditions towards this solution. In some cases, there exists more than one solution and the final result depends on our choice of the initial Anderson parameter. This will happen, for instance, in the so called coexistence region, close to the Mott-Hubbard metal-isolator phase transition. This does not represent a numerical artefact, but it is rather an indication of the Mott-Hubbard MIT at low temperatures [14].

#### Algorithmic details:

The self-consistency cycle (except for the ED part) was implemented in the program for the two-dimensional case in the subroutine selfconst2C\_direct, for the three dimensions case in selfconst3\_dir\_sym and for the infinite dimensional case in selfconstInf.

#### 3.3 Spectral function

After the selfenergy is transformed back into real frequencies rather than Matsubara frequencies we can calculate the spectral function:

$$A(\omega) = -\frac{1}{\pi} \operatorname{Im} \left( \int \frac{\mathcal{N}(\epsilon)}{\omega - \epsilon + \mu - \Sigma(\omega)} \, \mathrm{d}\epsilon \right)$$

where  $\mathcal{N}(\epsilon)$  is the non-interacting DOS.

The spectral function tells us a lot about the physical properties. For instance, if there is a finite spectral weight at the Fermi energy, (e.g. in the case of half-filling) one can easily excite electrons into the conduction band. Therefore we would get a finite conductivity.

Algorithmic details:

The spectral function was implemented in the program for the two-dimensional case in the subroutine intdos2C, for the three dimensions case in intdos3C and for the infinite dimensional case in intdosinf.

## 3.4 Optimization for calculating the non-interacting DOS

Although the computation power needed to calculate the non-interacting DOS is rather small in comparison with the ED-impurity solver, we may still want to optimize this part of the algorithm as well. This might be especially important if one works close to Van Hove singularities and/or regions of rapidly varying density of states away from the particle-hole symmetric case.

The calculation of the tight-binding DOS in finite dimensions reduces to the evaluation of a (multidimensional) integral. As already discussed, we can always eliminate one of the integrals by evaluating the delta distribution at its arguments roots, but the remaining integrations have to be dealt with numerically. This means that we have to discretize the remaining Brillouin zone into a finite number of elements. In order to determine the precision of the numerical procedure, we first have to introduce some sort of an error estimate.

**Errorscore:** In order to compare the integration error we define the following *errorscore*:

$$ES = \sum_{i=0}^{Emax-1} \frac{|x_i(feinh, ..) - x_i(feinh \to \infty)|}{Emax}$$

where feinh gives the number of subdivisions in the k summation from  $-\pi$  to  $-\pi$  separately for each dimension. So the total resulting number of subdivisions is  $feinh^d$  where d is the dimension of numerical integration.<sup>33</sup>

The limit  $feinh \to \infty$  cannot be calculated. We chose a value of 10000 for the calculations, which later on proved to be sufficiently large since the integral had already converged, and therefore the error score hardly changed with a further variation of feinh. Evidently, the error must be compared to the available computing power. The CPU-time needed to calculate the non-interacting DOS is proportional to  $feinh^d$  in first approximation (also if the subdivision parameter changes by using 'factor' times feinh in specific areas as reflected by the '...'-notation (e.g. areas of higher DOS).

#### 3.4.1 Variational k-subdivision

When screening the first resulting DOS it seemed that in some energy regions the integrals behaved 'nicely' and converged rather fast. Others, however, did not and had a rather

<sup>&</sup>lt;sup>33</sup>Note, however, that the necessary dimensions for the numerical integration were already reduced by one by dealing with the innermost analytically.

highly oscillating behaviour as we can see e.g. in the following plot where t'/t=0.3, t''/t=0.2, t''/t=0.2 (see figure 21).



Fig. 21 Comparing DOS for different k-subdivisions

We can see, from Fig. 21, that for low values of the DOS , the result behaves 'nicely' and there is hardly any difference visible between approximating the integration by a sum over 'ten thousand' or a 'hundred million' points, whereas in regions of higher DOS significant differences are observed.

This behaviour can be ascribe could be due to the integrable singularities of the integrand, for two reasons. First, if the integrand has an integrable singularity it makes a big difference if points particularly close to  $\tilde{x}$  are used or not. Second, if we look at the error approximation dependence on the number of k-subdivisions (see Figure 22) we find repetitive behaviour for the number of subdivisions being 210\*n with  $n \in \mathbb{N}$ . There we suspect the high error to result from single points which we seem to strike for special k-subdivisions.

Aiming at preserving the accuracy of the results, but also in acceptable computing time, it seems reasonable to vary the number of k-subdivisions dependently on the relative DOS value. For this reason (and because it is interesting for further investigations) we evaluated the bandwidth as a reference parameter.

We divided the DOS into regions of high and low DOS values w. r. t. the bandwidth (region 1 and 2 in Fig. 21). In region 1 we divided each k-direction of the fist Brillouin zone into *feinh* points, whereas in region 2 we used *feinh* times *factor*. (This is schematically sketched in Fig. 21 where *feinh* = 3 and *factor* = 2.)<sup>34</sup>



Fig. 22 Error-CPU time relation for trapezoid algorithm without variational subdivision

In order to find the right values for the parameters (*feinh* and the various *factors*) we tried (systematically) different combinations. We looked at the errorscore dependece compared to the CPU time, and made our final choice by minimizing the product of 'CPU time' and 'ES'. The four cases shown in figure 23 shall merely illustrate the process of minimizing the error as well as the calculation time. We found that the parameters chosen for the upper left set of calculations in figure 23 resulted in the fastest convergence. Therefore we chose those parameters for the final version of our program in two dimensions.<sup>35</sup>

<sup>&</sup>lt;sup>34</sup>This shell only be seen as a schematic explanation. For the final program we chose three different DOS regions with corresponding *factors* to define the BZ discrtization.

<sup>&</sup>lt;sup>35</sup>Similar tests were also made for the three dimensional case. Where we chose different values for the



Fig. 23 Errorscore dependent on various different k-scalings (corresponding parameters are listed at the bottom of each panel). We found that parameters corresponding to the upper left graph lead to the smallest error. Therefore, this 'optimal' parameter-set has been used for the final program. All of the calculations shown were done for a sample DOS in three dimensions with t'/t=0.3, t"/t=0.2, t"'/t=0.2

#### 3.4.2 Trapezoid- or Simpson-summation

We had to decide whether to do the k-sum by using a standard trapeze algorithm, or the slightly more complicated extended Simpson algorithm. (See Ref. [16]). Therefore we tested both versions. Interestingly, both showed almost equal convergence behaviour,

parameters (feinh and the factors).

although the usual error approximation should go as  $\mathcal{O}(1/n^4)$  for the Simpson algorithm, but only with  $\mathcal{O}(1/n^2)$  for a simple trapeze summation (where *n* is the number of nodes). The reasons for this may be that the necessary conditions, which are usually responsible for the faster convergence of the extended Simpson-algorithm compared to the trapezoidsummation, are in not **not** fulfilled in this case.

This error approximation (as given above) relies on the <u>fundamental theorem of calculus</u>. It requires the integrand to be continuously differentiable as many times as necessary. The extended Simpson algorithm requires therefore the existence and continuity of 4 and the Trapezoid algorithm of 2 derivatives, which is, regarding the calculation of the DOS, in general not obeyed anywhere within the integration borders.

We do not know if an extended Simpson algorithm would lead to more accurate results than a simple trapezoid summation, however, there also is no reason to believe that it should be worse. For the integrations occurring in the calculation of the noninteracting DOS an extended Simpson algorithm according to 'Numerical Recipes in FORTRAN' [16, p. 128] was chosen.

## 3.5 Symmetries

Whenever symmetries arise in a calculation, it would be wise to explore them. They usually make the readability of the code slightly more complicated, but the effort often pays off in computational time. If we look at any of the derived dispersion relations derived in the previous sections, from the tight binding model, one immediately recognises the following *symmetries*:

• translational symmetry

For all dispersions it holds to be true that  $\epsilon(\mathbf{k}) = \epsilon(\mathbf{k} + \hat{i}2\pi n + \hat{j}2\pi m + ..)$  with  $m, n, ... \in \mathbb{N}$  and  $\hat{i}, \hat{j}, ...$  Cartesian unit vectors.

- mirror symmetry There are three in the three dimensional case (e.g.  $\epsilon^{3D}(kx, ky, kz) = \epsilon^{3D}(-kx, ky, kz) = \epsilon^{3D}(kx, -ky, kz) = \epsilon^{3D}(kx, ky, -kz)$ ) and two in the two-dimensional case. (e.g.  $\epsilon^{2D}(kx, ky) = \epsilon^{2D}(-kx, ky) = \epsilon^{2D}(kx, -ky)$ )
- substitution symmetry There there is only one in two dimensions and four in three dimensions (e.g.  $\epsilon(kx, ky, kz) = \epsilon(ky, kx, kz)$ ). (Of course one can argue that these are only a special kind of mirror symmetries (e.g. the kx - ky = 0 plane))

The first one has already been used to restrict all calculations to the first Brillouin-zone, the second simplifies all integrations further by only considering positive values for  $k_x, k_y$ (and  $k_z$ , if existing). The third one again leads to a further reduction of the calculation boundaries which is rather straightforward to derive in the two-dimensional case (see figure 24).



Fig. 24 Two-dimensional energy dispersion for t'=0, t"'=0.7t within the first BZ and maximal possible reduction

In 3D, this can be visualized e.g. by looking at the Fermi surface (Fig. 25). In three dimensions one could restrict the calculation to 1/48 of the full first BZ. In the DMFT program, however, the 'substitution' symmetries were not used and therefore (partially redundant) calculations were performed on 1/8 of the first BZ. This might be improved in further versions of the program package.



Fig. 25 3D Fermi surface  $\mu = 0.5t$ , t' = 0.45t, t'' = 0.45t, t''' = 0; full 1 BZ (left) and maximaly possible reduced zone (right)

If the above mentioned symmetries apply to f(x, y, z) (assuming the translational symmetries were already used to restrict ourselves to the first BZ) then

$$\int_{-\pi}^{\pi} \mathrm{d}x \int_{-\pi}^{\pi} \mathrm{d}y \int_{-\pi}^{\pi} \mathrm{d}z f(x, y, z) = 48 \int_{0}^{\pi} \mathrm{d}x \int_{0}^{x} \mathrm{d}y \int_{0}^{y} \mathrm{d}z f(x, y, z)$$

## 4 Results

In order to test the new k-sum implementation in the program, simple calculations were made and compared to the expected results obtained, in limiting cases by an old version of the code. For the finite dimensional case we only looked at different cases of the hopping term t':

**Non-interacting system:** We set the interaction term U of the Hubbard model to zero which means that the electrons are completely uncorrelated. The *self energy*, which describes the difference between the propagation of an interacting and the non-interacting electron, is here identical to zero. Hence, the spectral function A(E) must be the same as the tight binding density of states, for this case.

Weakly interacting system: As as second step the interaction term U was set to a finite, but small, value w. r. t. the bandwidth. Under the assumption that the choice for U was sufficiently small, we expect little changes in the spectral function and that there will still be some spectral weight at the Fermi-level (usually close to E = 0). This also means that the imaginary part of the Green's function continued towards  $i\omega_n \to 0$  needs to have a sizeable finite value. The self-energy should now no longer be zero, however, its continuation towards  $\omega_n = 0$  should be small and Taylor-expandable. Im $[\Sigma(i\omega_n \to 0)]$  can be see as an inverse quasi particle lifetime  $\Gamma$ , and since we expect for the case of small interaction our system to still be in the conducting phase, the lifetime of an electronic excitation has to be large.

Strongly interacting system: In a third calculation we shall look at the case where the interaction is large, compared to the hopping terms. Now the spectral weight, should split into a lower and an upper Hubbard band. We can understand this by thinking about the limiting case of no hopping (atomic limit). The allowed energy levels are E = 0 (one electron per site) and E = U (two electrons at same lattice site). The chemical potential only redefines the energy origin symmetrically for a choice of  $\mu = U/2$ . Thus, we expect two delta peaks at  $E = \pm U/2$ . Introducing a small hopping term then leads to a 'smearing' of the delta functions similar in shape to the non-interacting density of states. The analytic continuation of  $\text{Im}[\Sigma(\omega_n \to 0)]$  should now diverge, since the quasi particle life time is expected to be zero (or at least very small) in an insulating material. Because of the divergence of  $\text{Im}\Sigma(i\omega_n \to 0)$  we also expect the continuation of  $\text{Im}[G(i\omega_n \to 0)]$  to go to zero.

All of the following calculations (unless stated otherwise) were done at a fixed chemical potential. We chose  $\mu$  to be halve of the interaction parameter U.

#### 4.1 Two dimensions

#### Non-interacting (U=0)

In the non iterating case (U=0) we find that the spectral function (see Fig. 26) is the same as the tight binding DOS. (For comparison see plots in chapter 2.) The imaginary part of the self energy is identical to 0, as it should, for all Matsubara frequencies as can be verified from figure 27. In order to check the new program against the old one, we compared the calculations in the case of t' = 0. Notice that for t' = 0 there are two data sets plotted in figures 26, 27 and 28, but only one line shows up: The reason for this is that both program versions give exactly the same results (for t'=t"'=0) and therefore the two plotted curves are on top of each other.



Fig. 26 Spectral function for U=0 with various diagonal nearest neighbour (t') hopping terms.





Fig. 27 Imaginary part of the self energy. We find it to be identical to zero, as expected.

Fig. 28 Imaginary part of the Green's function.

	$\mid$ t'=0 old	t'=0	t'=0.1	t'=0.5
$\epsilon_1(k)$	-1.49285375403088	-1.49285375403088	-1.50538128438227	-1.49526108533045
$t_{par1}$	0.244270722801538	0.244270722801538	0.164716272111184	0.295362331300039
$\epsilon_2(k)$	-0.296270941782202	-0.296270941782202	-0.554275260359337	-0.357956840177598
$t_{par2}$	0.296431656680485	0.296431656680485	0.389299421710294	0.431649567585519
$\epsilon_3(k)$	1.49285375403088	1.49285375403088	1.50825989060177	1.50014411651222
$t_{par3}$	0.244270722801538	0.244270722801538	0.154448965868814	0.206484629137688
$\epsilon_4(k)$	0.296270941782202	0.296270941782202	0.246193112063909	0.424993641000846
$t_{par4}$	0.296431656680485	0.296431656680485	0.311323527044965	0.243157299906638
$\int A(\omega) \mathrm{d}\omega$	-	0.999341895839648	0.999103053699569	$0.976324561018963$ $^{36}$
$\langle n_{\uparrow}n_{\downarrow} angle$	0.24999999999999999999999999999999999999	0.249999999999999999	0.212772909102385	0.153726975475679
$\langle n \rangle$	0.9999999999999981	0.9999999999999981	0.922556017310932	0.784175477502728

**Tab. 1** Data resulting after convergence of the self consistency cycle, in addition to figure 26, 27 and 28 (non-interacting case U=0). All calculations were done at the same inverse temperature  $\beta = 10$ .

Looking at the data in table 1 we notice that column 1 and 2 are identical. This means that the old program version gives exactly the same results as the new one for this case.<sup>37</sup> The average double occupancy per site is 1/4 in the case of no interaction and t' = 0. This is expected, since for a non-interacting system there is no correlation. Therefore we can decouple the expectation value of the product of two operators into the product oft the two expectation values of these operators:  $\langle n_{\uparrow}n_{\downarrow}\rangle = \langle n_{\uparrow}\rangle \langle n_{\downarrow}\rangle$ . Moreover, neither of the spin orientations are distinguished  $\langle n_{\uparrow}n_{\downarrow}\rangle = \langle n/2\rangle \langle n/2\rangle$ . Which is 0.25 for the particle-hole-symmetric case.

<sup>&</sup>lt;sup>36</sup>The rather big error of 2.5 % is due to the big van Hove singularity for this case, but can be reduced by using more data points for the non interacting DOS. This holds to be true for all the 2D cases with t' = 0.5t.

<sup>&</sup>lt;sup>37</sup>Both program versions had the same initial values for the Anderson parameters.

When leaving half filling (e.g column 3 and 4) we are still dealing with an uncorrelated system, but there are fewer electrons in the system <sup>38</sup> and this leads to an average double occupancy less than 1/4 (e.g. we get for  $t' = 0.5t \langle n_{\uparrow}n_{\downarrow} \rangle = 0.784175^2/4 = 0.15373$ )

## weak coupling



Fig. 29 Spectral function for U = 0.3t and  $\mu = 0.15t$  with various diagonal nearest neighbour (t') hopping terms.

When introducing a small interaction term of U = 0.3t, and change the chemical potential to  $\mu = 0.15$  (in order to stay at least for the case of t' = 0 at half filling) we find our spectral function to change slightly. In particular, we find that the spectral function is changed significantly at some specific energies. This is not a band splitting, but only an effect of the discretization of ED, visible through  $\Sigma(i\omega_n)$  which is now non-zero. We find that there is still some spectral weight at the Fermi-energy (around E = 0), which indicates that our system still is in the conducting phase This can also be seen from the self energy  $\Sigma(i\omega_n \to 0)$  shown in figure 30 and 32.

<sup>&</sup>lt;sup>38</sup>unless the chemical potential is changed accordingly which it was not for the calculations in table 1





Fig. 30 Imaginary part of the self energy.

Fig. 31 Imaginary part of the Green's function.

More specifically, we can fit a polynomial function of a low degree through a number of data points and than calculate a reasonable approximation to the continuation (value as well as slope) of the self energy at  $\omega = 0$ . This is shown in figure 32. From the tangent parameters we can than derive the inverse quasiparticle lifetime  $\Gamma = -\text{Im}[\Sigma(i\omega_n \to 0)]$  and the quasiparticle weight  $Z = \left(1 - \frac{\partial Im[\Sigma(\omega_n)]}{\partial \omega_n}\Big|_{i\omega \to 0}\right)^{-1} = \frac{m}{m^*}$ .



Fig. 32 Imaginary part of the self energy as well as a polynomial function fitted through 6 data points in order to calculate  $Im[\Sigma(i\omega_n \to 0)]$  and  $\frac{\partial Im[\Sigma(\omega_n)]}{\partial \omega_n}\Big|_{\omega_n \to 0}$ 

	t'=0 old	t'=0	t'=0.1	t'=0.5
$\epsilon_1(k)$	-1.50506725562377	-1.50506725562377	-1.49797163655166	-1.48785226796032
$t_{par1}$	0.183873016567092	0.183873016567092	0.249278205740854	0.369559742710354
$\epsilon_2(k)$	-0.321392519277843	-0.321392519277843	-0.367175679770873	-0.352068979465285
$t_{par2}$	0.311295199660366	0.311295199660366	0.360782858540734	0.405017502080542
$\epsilon_3(k)$	1.50506725562377	1.50506725562377	1.49608000343959	1.50165480982262
$t_{par3}$	0.183873016567092	0.183873016567092	0.259472198405778	0.174732094019413
$\epsilon_4(k)$	0.321392519277843	0.321392519277843	0.370431695014154	0.368582769393250
$t_{par4}$	0.311295199660366	0.311295199660366	0.300099040680088	0.237427265244786
$\int A(\omega) \mathrm{d}\omega$	-	0.999341838635642	0.999102993808134	0.976324499389737
$\langle n \rangle$	0.999999999999999990	0.99999999999999990	0.951490865909534	0.810035306042746
$\langle n_{\uparrow} n_{\downarrow} \rangle$	0.218232625161249	0.218232625161249	0.196563213358202	0.141676725006597
Γ	-	7.3960037 E-03	7.8342743 E-03	3.5352062 E-03
$\frac{m^*}{m}$	-	1.013278	1.009090	1.013520

**Tab. 2** Data resulting after convergence of the self consistency cycle, in addition to figure 29, 30 and 31 (non-interacting case). All calculations were done at the same inverse temperature  $\beta = 10$ .

For the data in table 2 we find that the old and the new program give the same results for the case t'=0. Moreover, we see that now the average double occupancy is no longer 1/4 for t'=0, but smaller. The local Coulomb interaction  $U \neq 0$  induces correlation effects and, hence,  $\langle n_{\uparrow}n_{\downarrow} \rangle \neq \langle n_{\uparrow} \rangle \langle n_{\downarrow} \rangle$  the energy degeneration of all possible states does no longer exist. Thus, the system favours states where there is only one electron per site. Notice, that since the interaction term is still small the average double occupation values are not altered that much from their previous values, where there was no interaction. Figure 30 suggests that the phase transition from a conducting to an insulating material occurs at bigger interaction values if t' is larger, but since we no longer have particle hole symmetry this can not be merely deduced from the imaginary part of the selfenergy.

#### strong coupling

Now we set the interaction term U to 4.0t and change the chemical potential accordingly to  $\mu = 2.0t$ . (half filling condition for t' = 0) We find in figure 33 a broad spectral gap centred around the Fermi energy (rescaled to E = 0). From figure 34 we can see that the quasi particle life time will go to zero matching our spectral gap observation as well as the fact that now  $\text{Im}[G_{i\omega_n \to 0}]$  goes to zero.



Fig. 33 Spectral function for U=4 and  $\mu = 2$  with various diagonal nearest neighbour (t') hopping terms.





Fig. 34 Imaginary part of the self energy. We find analytic continuation of the self energy in  $\omega_n \rightarrow 0$  display a diverging behaviour.

analytic continuation of the self energy in Fig. 35 Imaginary part of the Green's function.

From figure 33 we notice that there is not only the expected band gap at the Fermi energy, rather other smaller gaps appear at energies  $\approx \pm U/2$ . When comparing the

Anderson parameters from table 3 we find, that the major contributions in the spectral function to be at the Anderson energies. In order to check weather this is only effect of the discretization of the bath in the exact diagonalisation (ED) method, we redid the calculation for a different number of bath sites. Instead of 4 we used 6 to calculate the spectral function shown in figure 36. Since the position of the peaks changes, we find that the multi band splitting was actually only a numerical effect due to ED method, and conclude that the spectral contribution for E < 0 (E > 0) should form only one lower (upper) Hubbard band.



Fig. 36 Spectral function for U=4,  $\mu = 2$  and t' = 0. Comparing the two results (6 bath sites for the red curve, and 4 bath sites for the green curve) we find the multiband splitting to be an intrinsic effect of the ED method.

	$\mid { m t'=0  old}$	t'=0	t'=0.1	t'=0.5
$\epsilon_1(k)$	-2.44943089739828	-2.44943089739828	-2.39963450737626	-2.50225456948725
$t_{par1}$	0.340017133184689	0.340017133184689	0.336695319871839	0.403327307979448
$\epsilon_2(k)$	-1.00154956389231	-1.00154956389231	-1.05300771591365	-1.07748880739932
$t_{par2}$	0.147480461006942	0.147480461006942	0.151021472978515	0.145341244425999
$\epsilon_3(k)$	2.44943089739828	2.44943089739828	2.49882679043840	2.69716456808324
$t_{par3}$	0.340017133184689	0.340017133184689	0.300428542724779	0.244163250222396
$\epsilon_4(k)$	1.00154956389231	1.00154956389231	1.13697556295320	1.19510593675307
$t_{par4}$	0.147480461006942	0.147480461006942	0.193142005665908	0.274983487473534
$\int A(\omega) \mathrm{d}\omega$	-	0.999331165002388	0.999092327986973	0.976314051989087
$\langle n \rangle$	1.0000000000003	1.0000000000003	1.00002376967195	0.999966711463752
$\langle n_{\uparrow}n_{\downarrow} angle$	0.008271635665681509	0.008271635665681509	0.00831451433599330	0.01021313032211823

**Tab. 3** Data resulting after convergence of the self consistency cycle, in addition to figure 33,34 and 35 (non-interacting case). All calculations were done at the same inverse temperature  $\beta = 10$ .

Furthermore, we verify that the old version and the new version of the program give again precisely the same results for the same initial conditions, as can be seen from table 3 by comparing column two and three. We also notice from figure 34 and 35 that for the case of large interaction there is hardly any difference in the Green's function and the self energy for different values of t'. This suggests that further hopping terms have little effect on the systems properties in the case of strong interaction.

## 4.2 Three dimensions

In three dimensions we here again looked at the three representative cases of different values of interaction, and for each case at three different cases of t'.

## Non-interacting (U=0)



Fig. 37 Spectral function for U=0 with various diagonal nearest neighbour (t') hopping terms.

Without interaction the system is uncorrelated, and we find the spectral function A(E) (shown in figure 37) to be the same as the tight-binding DOS (see chapter 2).





Fig. 38 Imaginary part of the self energy. Here we find it to be identical to zero, as expected.

Fig. 39 Imaginary part of the Green's function.

	$\mid$ t'=0 old	t'=0	t'=0.1	t'=0.5
$\epsilon_1(k)$	-1.49280699589518	-1.49280699589518	-1.50598552839273	-1.50250026759181
$t_{par1}$	0.244478120929311	0.244478120929311	0.151581125258302	0.206936650408214
$\epsilon_2(k)$	-0.293171702931790	-0.293171702931790	-0.563545149681878	-0.276922814812577
$t_{par2}$	0.302365516236382	0.302365516236382	0.328079230574687	0.186396456836253
$\epsilon_3(k)$	1.49280699589518	1.49280699589518	1.50780593186320	1.59607170597931
$t_{par3}$	0.244478120929311	0.244478120929311	0.165446422045693	0.303918595001502
$\epsilon_4(k)$	0.293171702931790	0.293171702931790	0.259777199647849	0.393925425546625
$t_{par4}$	0.302365516236382	0.302365516236382	0.354014872189399	0.396926081931107
$\int A(\omega) \mathrm{d}\omega$	-	0.999171230948973	0.998300538676773	1.00247992760463
$\langle n \rangle$	0.99999999999999999	0.999999999999999990	1.04579249460398	1.25094863522518
$\langle n_{\uparrow}n_{\downarrow} \rangle$	0.250000000000004	0.250000000000004	0.273420123529000	0.391218156824892

**Tab. 4** Data resulting after convergence of the self consistency cycle, in addition to figure 37, 38 and 39 (non-interacting case U=0,  $\mu = 0$ ). All calculations were done at the same inverse temperature  $\beta = 10$ .

Table 4 shows that also for the three dimensional case (and no interaction) the old and the new version of the program give the same results (by comparing column two and three). We further see that if t'>0 the particle-hole symmetry breaks and this time we get a higher average double occupancy. The reason for this is simply that we have more electrons in the system. However, the electrons are still uncorrelated ( as we can check by calculating  $\langle n_{\uparrow}n_{\downarrow}\rangle = \langle n_{\uparrow}\rangle \langle n_{\downarrow}\rangle = \frac{\langle n\rangle^2}{4}$ ).

#### weak coupling

We chose a value of U = 0.3t and  $\mu = 0.15t$  for the case of small interaction. In figure 40 we see that the spectral function at U = 0.3t is similar to the non-interacting case, but we also find that a band gap starts to form around E = 0. We see in figure 41 that  $\text{Im}[\Sigma(i\omega_n \to 0)]$  is unequal to zero. In contrast to this the imaginary part of our Green's

function still becomes very large as  $i\omega_n$  goes to zero. This suggests that  $\text{Im}[\Sigma(i\omega_n \to 0)]$  would in fact still tend to go to zero, but that the choice of temperature prevents us from seeing this directly from figure 41. We find this to be true from figure 43. where the inverse temperature was set to 70 rather that to 10 and therefore the fist Matsubara frequency was at  $\frac{\pi}{70}$  rather than at  $\frac{\pi}{10}$ .



Fig. 40 Spectral function for U = 0.3t with various diagonal nearest neighbour (t') hopping terms.



Fig. 41 Imaginary part of the self energy for  $\beta = 10$ . Fig. 42 Imaginary part of the Green's function for  $\beta = 10$ .



Fig. 43 Imaginary part of the self energy for  $\beta = 70$ .

	$\mid { m t'=0  old}$	t'=0	t'=0.1	t'=0.5
$\epsilon_1(k)$	-1.79784483931142	-1.79784483931142	-1.79880542776856	-1.40145259441054
$t_{par1}$	0.267418096228720	0.267418096228720	0.246798435901758	0.190058261626624
$\epsilon_2(k)$	-0.453354476524438	-0.453354476524438	1.89640030150082	-0.432106812068145
$t_{par2}$	0.388016915985949	0.388016915985949	0.381523686725595	0.307664339425553
$\epsilon_3(k)$	1.79784483931142	1.79784483931142	-0.438220980989463	2.19462731028271
$t_{par3}$	0.267418096228720	0.267418096228720	0.291813947304787	0.330092527429619
$\epsilon_4(k)$	0.453354476524438	0.453354476524438	0.547293267732479	0.719433085211548
$t_{par4}$	0.388016915985949	0.388016915985949	0.438265918890771	0.539691122000824
$\int A(\omega) \mathrm{d}\omega$	-	0.999171168070949	0.998300465606832	1.00247984603449
$\langle n \rangle$	0.99999999999999980	0.99999999999999980	1.05261018939097	1.21916863463198
$\langle n_{\uparrow}n_{\downarrow}  angle$	0.221847097014169	0.221847097014169	0.249675864354864	0.348519571673607

**Tab. 5** Data resulting after convergence of the self consistency cycle, in addition to figure 40,41 and 42 (case of small interaction U=0.3,  $\mu = 0.15$ ). All calculations were done at the same inverse temperature  $\beta = 10$ .

We notice from table 5 that the average double occupancy is slightly smaller than in the non-interacting case. Moreover, we again find that the old and the new program-versions agree with each other.

#### strong coupling

To illustrates the case of larger interaction, we chose a vale of U = 4t and  $\mu = 2t$ . We find the expected spectral gap around E = 0 for all cases of t' (see figure 44). Again  $\operatorname{Im}[\Sigma(i\omega_n \to 0)]$  confirms that we have an insulating material.



Fig. 44 Spectral function for U = 4t with various diagonal nearest neighbour (t') hopping terms.



Fig. 45 Imaginary part of the self energy.

Fig. 46 Imaginary part of the Green's function.

For the scope of this thesis, it is important to stress that also for the case of a 3D system at strong coupling the old and the new program yield exactly the same results, if starting the calculations from the same initial conditions as can be seen from the data in the second and third column of table 6. This proves the correctness of the new implemented algorithm for the case t' = 0 (at all couplings).

	$\mid { m t'=0  old}$	t'=0	t'=0.1	t'=0.5
$\epsilon_1(k)$	-2.31480051283108	-2.31480051283108	-2.18936177534964	-2.14476120306561
$t_{par1}$	0.295011519236473	0.295011519236473	0.183266994134386	-0.155711771538620
$\epsilon_2(k)$	-1.28699318493289	-1.28699318493289	-1.56155953509879	-1.07096877382946
$t_{par2}$	0.194007105849740	0.194007105849740	0.286216594242213	0.306867379467773
$\epsilon_3(k)$	2.31480051283108	2.31480051283108	2.30225119873961	2.28479733634864
$t_{par3}$	0.295011519236473	0.295011519236473	0.263979925226117	0.345468138667884
$\epsilon_4(k)$	1.28699318493289	1.28699318493289	1.70260743077203	1.74505156295318
$t_{par4}$	0.194007105849740	0.194007105849740	0.251820747667892	0.278788631133834
$\int A(\omega) \mathrm{d}\omega$	-	0.999160510193872	0.998289819799703	1.00246910996737
$\langle n \rangle$	1.0000000000003	1.0000000000003	1.00003307768512	0.999775697847226
$\langle n_{\uparrow} n_{\downarrow} \rangle$	0.008179544051584896	0.008179544051584896	0.008413951471101360	0.01172656926434320

**Tab. 6** Data resulting after convergence of the self consistency cycle, in addition to figure 44, 45 and 46 (case of 'big' interaction U=4.0,  $\mu = 2.0$ ). All calculations were done at the same inverse temperature  $\beta = 10$ .

## 4.3 Infinite dimensions

The hopping term t is (after rescaling by the square root of the dimensions, see Ref. [1] or chapter 2) now redefined as the constant  $t^*$ , which set to 1.

#### Non-interacting (U=0)

In infinite dimensions the non-interacting (tight binding) DOS is a Gaussian (Ref. [1]) as we also have derived in chapter 2 and it can be seen in figure 47. For U=0  $\Sigma(i\omega_n) = 0$  (Fig. 48) Of course there is no bandgap in the noninteracting case and a large spectral weight at E=0 (Fig. 49). The data at figure 47, 48 and 49 is shown in table 7. We also chose  $\mu = 0$  which corresponds to particle hole symmetry.



Fig. 47 Spectral function for a non-interacting infinite dimensional hyper-cubic lattice



Fig. 48 Imaginary part of the self energy.

Fig. 49 Imaginary part of the Green's function.

### weak coupling

Now we set the interaction parameter U to a small value and the chemical potential to  $\mu = U/2$  respectively (to retain particle hole symmetry). For the red lines in figure 50, 51 and 52 we set U to  $0.25t^*$  for the green one to  $0.50t^*$ . We find the spectral function

to still to the one in the non-interacting case. Also the average double occupancy has not changed much from the previous value of 0.25 (see table 7). The imaginary part of the self energy, which still goes, with a negative slope, to a very small value as  $i\omega_n \to 0$ , showing the existence of the quasiparticles (electronic excitations) having a long lifetime. This means that we are dealing with a conducting system. The data regrading figure 50, 51 and 52 is shown in table 7.



Fig. 50 Spectral function for a system with weak coupling (small U).



Fig. 51 Imaginary part of the self energy.



Fig. 52 Imaginary part of the Green's function.

#### strong coupling

When we now set the interaction term to a 'big' value if  $U = 6t^*$  (and the chemical potential to  $\mu = 3t^*$  respectively) the most characteristic feature in the spectral function (shown in figure 53) is the bandgap around E=0. This means that we are dealing with an insulating material. Thus, the quasiparticle lifetime goes to zero, which can be seen from the fact that  $\text{Im}[\Sigma(i\omega_n \to 0)]$  becomes very large. As we have already concluded from the two dimensional case, the three-peak splitting of each Hubbard band is only a bath dicretization artefact of the ED method.



Fig. 53 Spectral function for U=6.

0

-0.02

-0.04

-0.06

-0.08

-0.1

-0.12

-0.14

-0.16

0

20

Im[G(w<sub>n</sub>)]



Fig. 54 Imaginary part of the self energy.

Fig. 55 Imaginary part of the Green's function.

ω<sub>n</sub>

40

60

U=7.0

80

100

•	U=0	$U{=}0.25$	$U{=}0.5$	U=6
$\epsilon_{1/3}(k)$	-2.08359419996468	-2.08344614584238	-2.08341828024892	-2.90183502260538
$t_{par1/3}$	0.575192685140926	0.575750779581209	-2.08341828024892	0.707191470565289
$\epsilon_{2/4}(k)$	-0.340999421776861	-0.338856533904027	-0.335441280828860	-0.378631894971853
$t_{par2/4}$	0.470508423435922	0.468639273165546	0.465393260783864	0.0859763296543600
$\int A(\omega) d\omega$	0.999562734552412	0.999562691002686	0.999562560647117	0.999536640009741
$\langle n \rangle$	0.99999999999999981	1.000000000000000000000000000000000000	0.9999999999999993	0.999999999999958
$\langle n_{\uparrow}n_{\downarrow} angle$	0.250000000000002	0.237679281878751	0.225307126997107	0.01508091216748104

**Tab. 7** Data resulting after convergence of the self consistency cycle for the infinite-dimensional cases. Since the Anderson parameters are symmetric around  $\epsilon_k = 0$  in the particle-hole symmetric case considered here, only two of the four are listed above. All calculations were done at the same inverse temperature  $\beta = 10$ .

### 4.4 'Under-cooling' the Mott-Hubbard-transition

Finally, we want to test our program by looking a little closer into the physical properties of the Mott-Hubbard metal insulator phase transition of DMFT. Notice, that for the purpose of illustration we only considered the case of an infinite dimensional cubic lattice. Specifically, we looked at the average double occupancy depending on the interaction parameter U. As it is known (Ref. [14, 17]) there are regions in the U-T-phase diagram, where there is only one stable solution, but there are also regions where the solution depends on the choice of the initial electronic bath, i.e. in the initial Anderson parameters. We wanted to check this feature of the DMFT solution by starting in a region where the solution is unique (e.g. starting with no interaction (U=0), or starting from strong interaction  $(U = 6t^*)$ ). Then for any following calculations we used the resulting Anderson parameters form the previous program-run as starting values, but changed the interaction term slightly by  $\delta U$ . We want to emphasize the analogy to freezing water. When water is cooled down carefully and slowly it can remain a liquid even when the temperature is below zero (degree Celsius). In the same sense, within the numerical treatment of DMFT, we can 'undercool' our Mott-Hubbard insulator (only that now the temperature stays constant and the varying parameter is the interaction U) for suitable conditions (e.g. a temperature below the critical temperature).



Fig. 56 There is no phase transition occurring at  $\beta =$  Fig. 57 For an inverse temperature of  $\beta = 70$  we find a 10. This also means there is only one stable solution.

discontinuity in the average double occupancy. There is more than one stable solution in a certain region of interaction.

For an inverse temperature of  $\beta = 10$  (see figure 56) no phase transition was visible, which means that the temperature was above the critical temperature: Here we are in the crossover region. In the contrary, at lower temperature ( $\beta = 70$ , see Fig. 57), we find that there is a *discontinuity* in the average double occupancy as a function of U, which means that we observe, numerically, a first order phase transition. Moreover, at this temperature the solution is not unique: For decreasing U (red line 'big U to small') the system remained in an insulating state, whereas for the set of calculations of increasing U (green line 'small U to big') the system remained in the conducting phase, up to a larger value  $U_{cr}$ . This can also be seen form figure 58 where we looked at the spectral function at  $\beta = 70$  around E = 0 at the intermediate interaction term  $U = 4.8t^*$  and found for the decreasing U case that the system had no spectral weight at E=0, whereas for the increasing case there still was a considerable spectral contribution.



Fig. 58 Spectral function around E=0 in the coexistence region ( $U = 4.8t^*$  with  $\beta = 70$ ). The red line illustrates that for an increasing interaction parameter ('small U to big') the system is still in the conducting phase, whereas the green line shows that for initial Anderson parameters corresponding to a higher interaction parameter ('big U to small') the system remains in the insulating phase.

# 5 Conclusion and Outlook

The implementation, and the successful benchmark of our new DMFT(ED) code, which allows to treat cubic lattices with hopping more complicated than the NN-one, will be exploited for interesting physical studies. The next interesting task would be to explore a phase diagram for the Mott-Hubbard metal insulator transition in analogy to figure 59, where we find the phase state dependence on temperature T and interaction parameter U. Specifically, the consideration of further hopping terms and their influences on the Mott-Hubbard MIT would be of interest.



Fig. 59 Phase diagram of the Mott- Hubbard MIT for an infinite dimensional Bethe lattice (from [17]). This represents one of the most famous results of DMFT.

However, the Mott-Hubbard metal-insulator phase transition would be here only the first step. We may also want to look at other phase transitions (e.g. magnetic ones). It is expected that further hopping terms have strong effects on magnetic ordering phenomena. To emphasise this we could take a look at the extended zone scheme for a two dimensional system at half filling with the hopping term t' = 1.0 in comparison to t' = 0 (figure 60).



Fig. 60 Extended Brillouin zone scheme for t' = 0 (left) in comparison to t' = 1 (right) at half filling. For t' = 0 we find as a reciprocal lattice vector  $Q = (\pi, \pi)^T$  that maps many points from the Fermi surface again onto the Fermi surface (nesting vector), corresponding to an instability towards anti-ferromagnetism. For t' = 1 we can identify vectors q and q', which indicates that different magnetic ordering becomes possible.

We find indications that the maximum in the magnetic susceptibility may be at another point in reciprocal space than  $Q = (\pi, \pi)^T$ . This suggests analogue tendencies in the magnetic ordering phenomena.

Eventually, since all second order phase transitions, like the magnetic ones, are associated with strong non-local correlations at low temperatures, the DMFT (ED) studies which are possible with the new program could be now used for performing calculations of spin (magnetic), charge (density) or superconducting (pairing) non-local fluctuations. e.g. with diagrammatic extensions of the DMFT, such as the Dynamical Vertex Approximation (D\GammaA, Ref. [18])

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