HYBRID MONTE CARLO SCHEME FOR INTERACTING DOUBLE-EXCHANGE SYSTEMS

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Abstract

Many magnetic and electronic properties of diverse materials of technological interest such as diluted, magnetic semiconductors, manganites and hexaborides can be described using kinetic-exchange models in which itinerant carriers are coupled to local magnetic moments. Monte Carlo simulations of the magnetic properties of such models usually treat the local moment spins as classical and ignore electron-electron interactions due to the need to diagonalize a fermion problem at each spin flip. We have developed the first hybrid Monte Carlo scheme that allows the inclusion of electron-electron interactions at a mean field level. We have implemented the scheme for a model which includes on-site Coulomb repulsion. The inclusion of on-site repulsion tends to enhance the tendency towards magnetic order, the extent of which depends on the coupling between itinerant carriers and local moments.
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Chapter 1

Introduction

The phenomenon of magnetoresistance (MR) has been known since 1856 when William Thomson measured the resistance of iron in a magnetic field [1]. He found that the resistance increased when a magnetic field perpendicular to the current was applied. The physics behind such MR is that a magnetic field perpendicular to the current deflects the electrons in the direction perpendicular to both the magnetic field and the current, decreasing the conductivity. The change in resistivity, or the MR value defined as \( \frac{R(H = 0) - R(H)}{R(H)} \), is limited to a few percent in most metals with practical magnetic fields.

1988 marked the discovery of the giant magnetoresistance effect (GMR) [2, 3]. GMR is observed in layered magnetic thin film structures which have alternating layers of ferromagnetic (FM) and non-magnetic materials. Unlike ordinary MR, which can be explained at a basic level with classical arguments, GMR is a quantum mechanical effect. If the FM layers have their magnetic moments aligned, spin dependent scattering of conduction electrons is minimized. Conversely, if the FM layers have their moments anti-aligned, spin dependent scattering is maximized, and the resistance of the heterostructure is maximized. For GMR the MR value is defined as \( \frac{R_P - R_{AP}}{R_P} \), where \( R_P \) is the resistance across the layers with the magnetization of the layers being parallel, and \( R_{AP} \) the resistance when they are anti-parallel. GMR structures can be fabricated to produce significant changes in resistivity from relatively small fields at room temperature; modern structures show a MR between 200-400 % [4]. Less than ten years after their discovery, GMR materials were being used in commercial applications, and such materials are now widely used in read heads for magnetic hard disks [5].

In the late 1980s, studies of MR in other materials increased, especially in manganese
oxides. In early experiments, thin films of \( \text{Nd}_{1/2}\text{Pb}_{1/2}\text{MnO}_3 \) and \( \text{La}_{2/3}\text{Ba}_{1/3}\text{MnO}_x \) were found to have MR values comparable to the magnitude of GMR in layered structures, independent of the direction of the applied magnetic field [6, 7]. Interest in manganese-oxides grew further with the discovery of the \textit{colossal magnetoresistance effect} (CMR) in 1994, when Jin \textit{et al.} [8] measured MR ratios as high as 127,000% near temperatures of 77 K and 1300% near room temperature in thin films of \( \text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_x \) in a 6 T magnetic field. CMR materials have been heavily studied since their discovery due to their potential for use in applications that currently use multi-layered GMR materials [5].

The manganese-oxides belong to a class of materials whose magnetic behaviour arises from the interaction of itinerant electrons with magnetic ions present within the lattice. Other materials with similar physics are dilute magnetic semiconductors (DMS) and rare earth hexaborides. DMS are semiconductors such as GaAs that are doped with magnetic atoms such as Mn.\(^1\) Much of the interest in DMS, especially in the past ten years, has been driven by the hope that they may be useful materials for spintronics; electronics that utilize the spin as well as the charge of electrons [9, 10]. A unifying feature of manganites, DMS, and hexaborides is that a minimal theoretical description of their magnetism can be given using a double exchange type model, as discussed further in Sec. 1.4. In addition to technological applications, these materials present a challenging theoretical problem. They are systems which have spin, charge, and lattice degrees of freedom which are intimately interrelated, and whose behaviour is strongly influenced by the presence of disorder and electron correlations [11]. Study of the interplay of these degrees of freedom in such systems has the potential to deepen our understanding of complex materials.

In this Chapter we introduce the physical properties of manganese-oxides, DMS, and rare earth hexaborides and review the current theoretical picture of these materials emphasizing commonalities between the seemingly disparate materials. The model of magnetic properties is sufficiently difficult to solve that it requires numerical methods to accurately determine its properties. We will focus particularly on developments using Monte Carlo (MC) methods. This Chapter concludes with an outline of the thesis.

\(^1\)(Ga,Mn)As is probably the most intensively studied DMS, however DMS involving II-VI semiconductors and other III-V semiconductors have also been studied.
1.1 Manganites

The manganese-oxide compounds generally referred to as the manganites are compounds which have the chemical composition $T_{1-x}D_x\text{MnO}_3$. $T$ is a trivalent rare earth cation, and $D$ is a divalent alkaline cation. The oxygen in such compounds is in the $O^{2-}$ state, and thus the addition of divalent alkaline or cation hole-dopes the system. The manganites have a perovskite lattice structure as depicted in Fig. 1.1. The oxygen atoms form an octahedral sub-lattice, the manganese atoms form a 3 dimensional cubic sublattice. The $T$ and $D$ atoms substitute into their own 3 dimensional cubic sub-lattice [12].

In undoped manganites ($x = 0$), the $\text{Mn}^{3+}$ ions have four electrons in the $3d$ shell. The octahedron of $O^{2-}$ atoms surrounding a $\text{Mn}^{3+}$ ion break the full rotational invariance of the orbitals of the $\text{Mn}^{3+}$. This causes a split between the two $e_g$ and three $t_{2g}$ orbitals. Lattice distortions also tend to split the two $e_g$ orbitals. Strong Hund coupling favours the spin of all four electrons to be parallel. The $e_g$ electrons are mobile in the compound, and can use the $O^{2-}$ $p$-orbitals to hop between $\text{Mn}^{3+}$ sites. On the other hand, the $t_{2g}$ electrons are bound quite strongly to the $\text{Mn}^{3+}$ ions. Holes doped into the material will induce $\text{Mn}^{4+}$ sites with only three localized $t_{2g}$ electrons. Such sites have a vacancy for the $e_g$ electrons from other sites to hop to, or equivalently, the holes introduced by the dopant can hop freely about the cubic manganese sub-lattice [13].
In addition to the dependence on the hole doping level, the compounds used for T and D can also significantly alter the physical properties, since these atoms have a strong effect on the hopping amplitude between Mn sites. This leads to significantly different phase diagrams between the different families of manganites. They can be broken up into three classifications; large, intermediate, and low bandwidth depending on the magnitude of the hopping amplitude relative to the strength of the Hund coupling, which is roughly constant for the three classes [14].

Many of the early studies of the manganites focused on the compound La_{1-x}Sr_xMnO_3. It is categorized as a large bandwidth manganite as the hopping amplitude between Mn atoms is large relative to the majority of other manganites [15]. The phase diagram for La_{1-x}Sr_xMnO_3 as well as resistivity versus temperature plots for x = 0.175 are depicted in Fig. 1.2. For intermediate hole doping, transitions between a ferromagnetic metal (FM) and either a paramagnetic insulator (PI) or metal (PM) occur at temperatures close to room temperature. There is also a high temperature metal-insulator transition that occurs at x ≈ 0.26 as shown in Fig. 1.2. At low temperatures the system has an antiferromagnetic (AFM) ground state at high doping, and either a ferromagnetic insulator (FI) or spin-canted insulator (CI) ground state at low doping, with the FM phase at intermediate dopings shown in Fig. 1.2.

The phase diagram for the intermediate bandwidth manganite La_{1-x}Ca_xMnO_3 as well as resistivity versus temperature plots for x = 0.25 are depicted in Fig. 1.3. The phase diagram is similar to that of the large bandwidth La_{1-x}Sr_xMnO_3, but a charge/orbital-ordered phase (CO) forms at certain densities and temperatures. Pr_{1-x}Ca_xMnO_3, which has no FM phase, is an example of a low bandwidth manganite. It has a charge/orbital ordered insulating phase (COI), as well as a low temperature canted antiferromagnetic insulator phase (CAFI). The phase diagram as well as resistivity versus temperature plots for x = 0.3 are depicted in Fig. 1.4. It is interesting to note that the resistance reacts to an increase in pressure in the same way as when a magnetic field is applied.

By referring to the phase diagrams for the different bandwidth compounds, one can note that the transition temperature for the ferromagnetic transition increase along with the bandwidth. However, the CMR effect is significantly stronger in the lower bandwidth compounds. The large bandwidth materials are the most likely candidates for applications, as their CMR peaks occur around room temperatures for a wide range of doping. On the other hand, study of the low bandwidth compounds, where more exotic phases appear and
the CMR effect is maximized, may be a more suitable avenue for studying the origin of the effect.

For a particular doping, the MR ratio of the manganites tends to peak in the vicinity of phase transitions. This suggests that the dominant factor contributing to the CMR is not the low or high temperature phase of the system, but rather the competition between phases [14]. In fact, for $La_{1-x}Sr_xMnO_3$, studies by Tokura et al. have shown that the CMR effect is maximized in the doping region about $x = 0.175$, corresponding to the point in the phase diagram where there is a transition at low temperatures between a FM and FI ground state. Thus there can be three competing phases in the vicinity of the Curie temperature [16]. There is widespread belief that phase separation occurs in these systems close to transition boundaries in the phase diagram. Percolatory phase separation in thin films has also been directly verified by experiment [17, 18]. A qualitative description of CMR in terms of a random resistor network induced by phase separation was proposed by Burgy et al. [19].

A description of these complex materials which only includes the kinetic energy of the fermions and the Hund coupling interactions is known not to capture all of the relevant
Figure 1.3: a) Phase diagram of La$_{1-x}$Ca$_x$MnO$_3$. b) Resistivity versus temperature plots for La$_{0.75}$Ca$_{0.25}$MnO$_3$ in several magnetic fields. (Both panels taken from Ref. [14])

Figure 1.4: a) Phase diagram of Pr$_{1-x}$Ca$_x$MnO$_3$. b) Resistivity versus temperature plots for Pr$_{0.7}$Ca$_{0.3}$MnO$_3$ at several magnetic fields. c) Resistivity versus temperature plots for several pressures. (Panels taken from Ref. [14])
physics. The existence of phase separation and complex charge/orbital-ordered phases appears to arise from various combinations of strong electron-lattice coupling, electron-electron interactions, disorder, and multiple orbital effects.

1.2 Dilute Magnetic Semiconductors

Dilute magnetic semiconductors are semiconducting materials that have been doped with a magnetic atom, generally a transition metal such as Mn. Most of the early work in the field focused on Mn doped II-VI semiconductors such as Cd$_{1-x}$MnxTe [20], where one can achieve high concentrations, ($x > 0.75$), of the transition metal dopant. Manganese in II-VI semiconductors possesses the same valence as the II element and so the dopant atoms form magnetic moments in the compound but do not introduce any carriers. Early attempts to introduce Mn into GaAs saw slow progress due to the low solubility of Mn in GaAs ($x \lesssim 0.001$) [21]. This changed when Ohno et al. [22] demonstrated that (Ga,Mn)As could be grown with much higher Mn content, up to $x \sim 0.1$, using molecular beam epitaxy (MBE). This lead to the observation of relatively high Curie temperatures ($T_C$) of order 110 K. The current record is $T_C \sim 185$ K [23].

GaAs has a zinc blende structure, where the sub-lattice of the Ga atoms forms a face-centered cubic (FCC) lattice. When doped with Mn, the Mn impurities can substitute for Ga atoms or insert interstitially into the lattice. Figure 1.5 shows the lattice structure for Ga$_{1-x}$MnxAs (typically $x \in [0,0.1]$). We will focus on the substitutional Mn as it gives rise to the interesting magnetic properties of the material. Three $t_{2g}$ and two $e_g$ electrons are strongly bound to the Mn atom, and a large Hund coupling favors the alignment of their spins forming a magnetic moment. At the same time, the Mn atom acts as an acceptor when substituting for Ga, and therefore introduces itinerant holes into the material [24]. The interaction between the itinerant holes and the local moments, both introduced to the compound by the Mn, leads to ferromagnetism in which electronic and magnetic properties are intimately linked.

Several different III-V compounds have been characterized experimentally, and there is consensus that $T_C$ is strongly dependent on the particular group III and V elements used, the concentration on Mn, the concentration of holes, and the sample preparation procedure. The strong dependence of $T_C$ on the preparation procedure, due to various unintended defects, such as Mn interstitials, As antisite defects, and spontaneous formation.
Figure 1.5: Lattice structure of Ga$_{1-x}$Mn$_x$As showing the two types of Mn impurities in the lattice; substitutional (Mn$_{Ga}$), and the less common interstitial (Mn$_{I}$). (Figure taken from Ref. [25])

of MnAs clusters, initially made it difficult to determine the intrinsic behaviour of $T_C$, particularly for larger concentrations of Mn [23, 25]. Annealing procedures were developed to improve sample quality by removing Mn interstitials (especially at larger x) and these have led to higher maximum observed values of $T_C$; from 110 K at $x = 0.053$ in 2002 [24] to 185 K for $x = 0.12$ in 2008 [23]. The transport properties of (Ga,Mn)As are also improved by annealing, however there is intrinsic disorder in the material due to the random positions of substitutional Mn atoms. This results in an insulator to metal transition of the Anderson type as x is increases; typical values of x are 0.02-0.03 depending on the level of compensation. The magnetic properties do not appear to be greatly affected across this transition. Figure 1.6 summarizes recent $T_C$ measurements for Ga$_{1-x}$Mn$_x$As, and shows insulating and metallic regions.

### 1.3 Rare earth hexaborides

The rare earth hexaborides are compounds with the chemical composition $R_{1-x}A_xB_6$, where R is a rare earth magnetic ion and A is a alkaline-earth metal. The R and A atoms
form a simple cubic lattice, with boron octahedra residing in the center of the cubic lattice as shown in Fig. 1.7 [26]. Each divalent R ion has seven localized electrons which half fill the 4f shell. A Hund coupling favors the alignment of the spins of the 4f electrons, giving each ion a localized spin of $\frac{7}{2}$. A very small effective carrier density of order $10^{-3}$ per unit cell has been measured for these compounds, which arises from the presence of defects in the crystalline environment. In particular, it has been suggested that boron vacancies contribute excess electrons [27, 28]. Measurements have also shown a sizeable gap between the valence and conduction band in some of these compounds [28]. These factors suggest that the ferromagnetism in these compounds has a similar origin to that in DMS and the manganites, in which itinerant carriers interact with localized spins [29].

Of the rare earth hexaborides, the compound which has received the most attention is Eu$_{1-x}$Ca$_x$B$_6$. In the absence of Ca, EuB$_6$ has a transition to a ferromagnetic phase close to 12 K. The ferromagnetic transition temperature decreases with increasing Ca content, with no magnetism present in CaB$_6$, Ca being a non-magnetic element. Ca and Eu are isovalent, so carriers are not introduced by the Ca dopant which serves to dilute the magnetic system and conducting lattice [29]. Eu$_{1-x}$Ca$_x$B$_6$ exhibits CMR at low temperatures. Wigger et al. [30] observed a MR ratio of $\approx 10^5$ in a 10 T field for $x = 0.27$ at 0.35 K, and suggested

![Figure 1.6: Schematic phase diagram as a function of temperature and $x$ for Ga$_{1-x}$Mn$_x$As, combining results from Refs. [25] and [23]. FM refers to ferromagnetic metal, while FI refers to ferromagnetic insulator.](https://example.com/figure1_6.png)
that the large value can be attributed to magnetic domain percolation caused in part by the disorder associated with the positions of Ca ions [30]. Similarly to the manganites [14], it is expected that interactions between the carriers may enhance the tendency towards phase separation in these materials [31].

1.4 Theoretical work

The manganites were first investigated long before the discovery of CMR, when Jonker and Van Santen studied ferromagnetism experimentally in some manganite compounds in the 1950s [32]. Soon after their experiment, Zener proposed a theory that correctly predicted the low temperature ferromagnetic behaviour of the system, that of double exchange (DE) [33]. Similar DE type models have also been proposed to describe the ferromagnetism in DMS [34, 35] and hexaborides [29]. Although the theory is qualitatively straightforward, analytical solutions for the general case present an extremely difficult challenge. Theoretical studies of DE type systems therefore usually resort to simplifying approximations and computational techniques. We now give an introduction to the model and numerical approaches to its solution.
The Hamiltonian of the double exchange model (DEM) contains two terms: one associated with the kinetic energy of the electrons, the other associated with the exchange interaction between itinerant electrons and local moment spins. The Hamiltonian is

\[
H = -\sum_{i,j} t_{ij} \left[ c_{i\sigma}^\dagger c_{j\sigma} + \text{h.c.} \right] + \sum_{i,j} J_{ij} S_i \cdot \left( c_{j,\alpha}^\dagger \frac{1}{2} \sigma_{\alpha\beta} c_{j,\beta} \right),
\]  

(1.1)

where \( t_{ij} \) is the hopping amplitude between sites \( i \) and \( j \), \( J_{ij} \) is the strength of the Hund coupling between a localized spin at site \( i \) with spin \( S_i \) and an electron at site \( j \), \( \sigma_{\alpha\beta} \) is the vector of Pauli spin matrices, and \( c_{i\sigma}^\dagger \) and \( c_{i\sigma} \) are the creation and annihilation operators for electrons at site \( i \) with spin \( \sigma \) respectively. The local moments may be diluted (so that only some sites are occupied) or dense (every site is occupied). From here on, the \( J_{ij} \) term will be referred to as the exchange coupling, as it couples the local moments to the spin of the free electrons [14]. If one chooses to consider only nearest neighbour hopping and on-site exchange coupling, the position subscripts can be dropped from the hopping and exchange coupling parameters; \( t_{ij} \rightarrow t \) and \( J_{ij} \rightarrow J \).

Despite its simple appearance, the DE model contains physics associated with a rich variety of behaviours. If the number of carriers, \( N_e \), is much larger than the number of local moments, \( N_{lm} \), Kondo physics of magnetic impurity scattering dominates at low temperatures. For a larger concentration of local moments and \( J/t < 1 \), the physics can be viewed as local moments interacting via a Fermi sea, with an effective Heisenberg interaction given by the RKKY exchange interaction [36], which oscillates as a function of \( k_F r \), where \( k_F \sim (N_e/N_{lm})^{1/3} \) is the Fermi wavevector and \( r \sim (3/N_{lm})^{1/3} \) is the typical distance between local moments. In general there will be both ferromagnetic and antiferromagnetic exchanges between pairs of spins and the resulting magnetic state may be a spin glass.

For the manganites, DMS, and hexaborides, carrier concentrations are typically small; such that \( N_e/N_{lm} < 1 \). In the manganites, some hexaborides, and certain doping ranges of DMS, it can be appropriate to assume that local moments form a dense lattice. Hopping is then restricted to occur only between local moment sites in the models for these materials Eq. (1.1), and thus the maximum allowed value of \( N_e \) is \( 2N_{lm} \).

### 1.4.1 Qualitative behaviour of the DEM

Even without calculation it is possible to gain some appreciation for the properties of the DEM [Eq. (1.1)]. The hopping term favours delocalization of the electrons in order to
minimize kinetic energy. The exchange term has the effect of promoting the alignment of local moments through an effective interaction mediated by electrons (see Fig. 1.8). If $J$ is strong it can have the effect of spin polarizing electrons, and if the local moments are dilute, it can lead to carrier localization. The tendencies that win out depends in detail on the ratios $N_e/N_{lm}$ and $J/t$.

In the low carrier density limit, $N_e < N_{lm}$, and with finite $J/t$ we should expect a ferromagnetic ground state, as the kinetic energy term favours delocalized carriers, and the exchange energy is minimized by aligning all of the local moments and spin-polarizing the carriers. Regardless of the doping, at high temperatures entropy will lead to a paramagnetic (PM) phase, and we therefore expect a transition from a ordered phase to a PM configuration at some finite temperature (provided the dimensionality is large enough for such a transition to occur at non-zero temperature). The temperature at which the transition occurs evidently depends on the ratios $N_e < N_{lm}$ and $J/t$.

1.4.2 Monte Carlo simulations of the DEM

From our discussion above of the DEM, it is evident that the low temperature behaviour of the model is strongly dependent on the value of the model parameters, $J/t$, and the ratio $N_e/N_{lm}$. A more quantitative picture than the one given above requires calculation, and
particularly at finite temperature, Monte Carlo (MC) simulations are a useful technique.

Initial MC studies of the DEM were performed by Yunoki et al. [37] and Calderon et al. [38]. These authors made the following simplifications to the DEM as compared to Eq. (1.1): i) the local moments were treated as classical vector spins, ii) hopping was restricted to be of nearest neighbour form, and iii) long range Hund couplings were ignored. Since the DEM is quadratic in electron operators it can be diagonalized exactly for a given configuration of classical spins and the Metropolis algorithm can be used to update the local moments.\footnote{Technical details of MC simulations of quadratic DE type systems are left for discussion in Chapters 2 and 3} Figure 1.9 shows results obtained by Yunoki et al. [37]. Panel a) shows the phase diagram as a function of electron density versus $J/t$ for the model in two dimensions at a temperature $T = t/50$. The diagram was constructed based on data from several small system sizes. As expected, regions of the phase diagram with large $J/t$ or small $\langle n \rangle$, (here $\langle n \rangle$ is the average number of electrons per site), show FM ordering of the local moments. As the doping approaches $\langle n \rangle = 1.0$, the authors identify a region of phase separation (PS). The authors found that they could not equilibrate samples for particular dopings in that region. The second panel in Fig. 1.9 shows the spin-spin correlation function for a one dimensional chain with $J/t = 8.0$ for two different dopings. One can see a very clear FM correlation for $\langle n \rangle = 0.72$ and an AF correlation for $\langle n \rangle = 1.0$.

1.4.3 Beyond the DEM

Although the DEM qualitatively predicts the existence of a FM and AF phase, it neglects many of the other important features of DE type systems. The physics that should be included to obtain a more accurate model vary depending on the material. Other physics present in DE type materials include anti-ferromagnetic super-exchange interactions between the local moments, Coulomb interactions between the charge carriers, various forms of disorder in the systems, multi-band effects, and, particularly for the manganites, electron-phonon coupling due to Jahn-Teller lattice distortions [14]. We discuss these effects in turn, but our main interest will be on electron-electron interactions.

Anti-ferromagnetic super-exchange interactions between local moments in most DE type systems are generally small compared to the other energy scales in the materials [41, 42], but it is of great importance near $\langle n \rangle = 0, 1, 2$ doping. At $\langle n \rangle = 0, 2$ we have either no
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Figure 1.9: a) Phase diagram for the DEM in two dimensions as a function of doping and $J/t$ at a temperature $T = t/50$ using sample sizes $4 \times 4, 6 \times 6, 8 \times 8$. Along with the FM phase, the authors identified regions of incommensurate spin correlations (IC) and phase separation (PS). b) Spin-spin correlation as a function of distance ($l$) for a 20 site 1D chain and two different dopings at $T = t/75$ and $J/t = 8.0$. (Figures taken from Ref. [37])

electrons, or a full band, and the effective coupling between local moments from the double exchange mechanism goes to zero, and the small but finite super-exchange interaction is important in these doping limits. At $\langle n \rangle = 1$ systems have nearly localized electrons, and again the super-exchange becomes relevant. At all other intermediate dopings, the local moment exchange term dominates the super-exchange term and thus it can be excluded from more complex models without qualitatively changing the physics [43].

For the manganites, it is widely accepted that electron-phonon coupling due to Jahn-Teller lattice distortions can be important for some systems. Several studies have shown that Jahn-Teller effects are particularly important for obtaining quantitative results for the conduction properties and ferromagnetic transition temperature $T_C$ [11, 14, 44]. In fact, one of the important effects of the Jahn-Teller lattice distortions is to split the two-fold degenerate $e_g$ band [39]. These effects are beyond the scope of the present study, and therefore an in depth discussion is omitted. We summarize previous work on including interactions and disorder in DE type models below.
Coulomb interactions

In Eq. (1.1), the electrons are taken to be non-interacting, however in general we expect that Coulomb interactions between electrons will be present, and determining where Coulomb interactions can be ignored, and when they must be considered, is not always an easy task. For all three classes of materials discussed, the on-site Coulomb repulsion can be a large energy scale [14, 40]. Long range Coulomb interactions should also generically be present, and have been suggested to lead to phase separation.

Two early studies [46, 48] of the interacting DEM investigated models of the form:

\[
H = -t \sum_{(i,j)} \left[ c_{i\sigma}^\dagger c_{j\sigma} + h.c. \right] + J \sum_i \vec{S}_i \cdot \left( c_{j,\alpha}^\dagger \frac{1}{2} \vec{\sigma}_{\alpha\beta} c_{j,\beta} \right) - J_{AF} \sum_{(i,j)} \vec{S}_i \cdot \vec{S}_j \\
+ U \sum_i n_{i\uparrow} n_{i\downarrow} + V \sum_{\langle i,j \rangle, \sigma, \sigma'} n_{i\sigma} n_{j\sigma'},
\]

(1.2)

where \( U \) and \( V \) are the strength of the on-site and nearest neighbour Coulomb repulsion respectively, and \( J_{AF} \) is the strength of the anti-ferromagnetic super-exchange interactions between local moments.

Malvezzi et al. [48] studied the model with \( J_{AF} = 0 \) using the density matrix renormalization group on one dimensional systems with spin-1/2 local moments. They found phase separation at finite nearest neighbour interactions, observing islands of magnetic ordering about nearly localized electrons. This behaviour is what one would expect in one dimension regardless of the doping. As the repulsion is increased the electrons segregate on the lattice, and once the overlap of their wavefunctions becomes negligible, each electron will have exchange coupling to only a few local moments. Even without Coulomb interactions we would not expect long range magnetic ordering in one dimension with only short range interactions.

Mishra et al. [46], performed exact diagonalization of Eq. (1.2). They found that interactions had dramatic effects on the ground state. Four ground state phases were found, FM, FM charge ordered, AF, and AF charge ordered, depending on the doping and model parameters. They also treated their model at the mean field level for finite temperatures and observed that these phases persisted above \( T = 0 \) (see Fig. 1.10). Berciu and Bhatt [49] also investigated the effect of on-site Coulomb repulsion on a model for DMS, and found \( T_C \)

\(^3\)For example, one study found for the manganite \( \text{LaMnO}_3 \), \( J \approx 1.8 \text{ eV} \) and the on site Coulomb repulsion to be \( U \approx 10 \text{ eV} \) [45]. The nearest neighbour hopping amplitude is much smaller, \( t \approx 0.15 \text{ eV} \) [46].
Introduction

The phases in the diagram are paramagnetic charge non-ordered (PN), ferromagnetic charge non-ordered (FN), ferromagnetic charge ordered (FO), and antiferromagnetic charge ordered (AFO). Hatched areas are regions of two phase coexistence. (Figure taken from Ref. [46])

to increase with the strength of the interaction, as the on site repulsion tends to split the two spin bands.

Disordered systems

Disorder in DE type systems can arise in various ways. For III-V DMS materials, the Mn atoms substitute randomly in the lattice for the type III element leading to positional disorder in the local moment sites, which affects $t_{ij}$ and $J_{ij}$ if hopping is only between Mn sites [35]. Under certain growth conditions the disorder may not be completely random as Coulomb repulsion between the Mn ions can lead to correlated disorder [50, 51]. In manganites the electrons hop between the Mn ions which form an ordered lattice. The hopping amplitude of the electrons between two Mn sites is determined by the angle of the Mn-O-Mn bond of each site, which in turn is related to the fraction of T/D in the unit cell (see Fig. 1.1). Since the D atoms are randomly distributed, the hopping amplitude between neighboring sites will vary. This form of disorder can be modeled by a random on site disorder potential.

Berciu and Bhatt [34] investigated the effect of positional disorder on the DEM applied to DMS materials, mainly focusing on (Ga,Mn)As. They placed local moments randomly on
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Figure 1.11: Plot of the average Mn spin (upper lines) and average hole spin (lower lines) at several disorder strengths for Ga$_1-x$Mn$_x$As with $x = 0.00926$ and $N_e/N_{lm} = 0.1$ obtained from a DEM calculated using mean field theory. (Figure taken from Ref. [34])

an fcc lattice, and treated the magnetism at the mean field level by making the substitution $S_i s_j \rightarrow \langle S_i \rangle s_j + S_i \langle s_j \rangle - \langle S_i \rangle \langle s_j \rangle$, where $s_j = \frac{1}{2} e_{j\alpha}^{\dagger} \sigma_{\alpha\beta} e_{j\beta}$. Some of the magnetization curves they obtained for a hole doped system are shown in Fig. 1.11. The magnetization plots have a peculiar concave-upwards form. The origin of the concave up shape is both $N_e/N_{lm} \ll 1$ and disorder which make ferromagnetism more inhomogenous.

Kennett et al. [35] performed a similar study on the same disordered DEM as Berciu and Bhatt, but treated the classical local moments exactly using MC simulations. Due to the inclusion of disorder, there are several key differences between their study and the MC studies on the DE model discussed earlier [37, 38, 39]. In those earlier simulations the chemical potential was specified and the expectation value of the number of carriers was observed to determine the doping level of the sample. In the presence of disorder, one must average measurements over many realizations of disorder, which requires an averaging over many samples that have the same doping. For finite sized samples the chemical potential required to give an expectation value of a particular doping depends on the realization of disorder. Kennett et al. therefore had to develop a procedure to determine the appropriate
value for the chemical potential for each disordered sample. This work confirmed the concave shape of the magnetization (see Fig. 1.12). The $T_C$ values were lower, which can be attributed to the neglect of temporal fluctuations in mean field. Also shown in Fig. 1.12 is a plot of the joint distribution function of local magnetization and local charge density at a particular concentration and hole density below $T_c$. One can see a relatively strong correlation between the two observables, further strengthening the argument for the presence of an inhomogeneous ferromagnetic phase in the system. Another important feature verified in the study was the fact that disorder indeed raised $T_c$ relative to an ordered system for low doping.

1.5 Outline of work

The goal of the work here is to develop and implement a MC scheme for DE type models including electron-electron interactions. In order to obtain results on moderate number of local moments, interactions are treated at the mean field level. In this thesis we discuss the Monte Carlo scheme and implementation that we developed along with some results for a particular DE model. In Chapter 2 we discuss the theoretical background to MC simulations of interacting DE models. In particular we discuss the Hartree-Fock approximation to the model, and exact diagonalization results. In Chapter 3 we discuss the details of the actual MC simulation, with particular emphasis on issues of equilibration, and results for our interacting DE model are presented in Chapter 4. Conclusions are drawn in Chapter 5.
Figure 1.12: a) Plot of magnetization as a function of temperature. Here $x$ is the local moment concentration, and $p$ is the number of holes per local moment. b) Joint distribution function of local magnetization and charge density for $x = 0.01$ and $p = 0.1$. $T = 0.01\; J$ (Figures taken from Ref. [35])
Chapter 2

Hartree-Fock Monte Carlo treatment of the interacting double exchange model

In this Chapter we present the theoretical basis of our treatment of DE type models with Monte Carlo (MC) simulations that include electron-electron interactions at the mean field level. In Sec. 2.1 we discuss MC simulations of DE type models in which non-interacting fermions are coupled to classical spins. In Sec. 2.2 we introduce a generalization of the DE model which includes an interaction term, and discuss the Hartree-Fock (HF) treatment of this interaction term. The HF approximation allows for the reduction of a Hamiltonian which is quartic in fermion operators to one which is quadratic, so that the MC framework developed in Sec. 2.1 can then be applied. In Sec. 2.3 we present calculations which compare energies and densities calculated from the model using HF to those found by exact diagonalization of the many body Hamiltonian for a variety of local moment spin configurations. These results allow us to identify regions of parameter space where a MC simulation is likely to give an accurate account of the behaviour of the model.

2.1 Monte Carlo simulations

Before discussing MC simulations for interacting fermion systems, we briefly review MC simulations of the double exchange model.
The local moments in the DE model have a quantum spin $S$, hence the Hilbert Space of the local moments contains $(2S + 1)^N_{lm}$ states, where $N_{lm}$ is the number of local moments in the system. For a single band of non-interacting spin-$1/2$ fermions coupled to local moments, there are $2N_{lm}$ fermion states, leading to $2N_{lm}(2S + 1)^N_{lm}$ states in total. Evidently, if one wishes to treat the local moments exactly, system sizes must be kept very small. To investigate larger systems one approximation that can be made is to treat the local moments as classical spins. In many DE type systems, the spin $S$ is relatively large,\(^1\) and since one expects quantum corrections to scale as $1/2S$ \(^52\), the approximation should give qualitatively reasonable results. The classical spin approximation means that we can parametrize the local moments by their $z$ component and azimuthal angle:\(^2\)

$$
S_i = (S_i^z, \phi_i),
$$

$$
S_i^x = S \sqrt{1 - \left(\frac{S_i^z}{S}\right)^2} \cos \phi_i,
$$

$$
S_i^y = S \sqrt{1 - \left(\frac{S_i^z}{S}\right)^2} \sin \phi_i.
$$

In order to calculate the thermodynamic properties of the system, we can note that the partition function may be written in terms of the classical spins $\{S_i\} = \{(S_i^z, \phi_i)\}$ as:

$$
Z = \left[ \prod_{i=1}^{N_{lm}} \int_{-1}^{1} dS_i^z \int_{0}^{2\pi} d\phi_i \right] e^{-\beta \mathcal{E}(\{S_i\})},
$$

where $\beta$ is the inverse temperature, $\beta = 1/k_BT$.

In a system of classical spins not coupled to fermions, $\mathcal{E}(\{S_i^z, \phi\})$ is the internal energy of the configuration ($\{S_i\}$). With the inclusion of fermions we need to use the free energy and the grand canonical partition function. With the form of $Z$ in Eq. (2.2), each configuration of classical spins has a weight in the partition function given by

$$
w(\{S_i\}) = e^{-\beta \mathcal{E}(\{S_i\})}.
$$

One can sample a representative portion of phase space for the purpose of calculating thermodynamic averages. The construction of a Markov chain (i.e. a set of spin configurations

\(^1\)For example, in (Ga,Mn)As $S = 5/2$, and in EuB$_6$ $S = 7/2$, while the manganites have $S = 3/2$.

\(^2\)The discussion in this section follows closely to that from Kennett et al. \[35\].
that are connected via detailed balance) is a way to do this that guarantees sampling with equilibrium weights. One way to satisfy detailed balance for a transition between configurations, \( \{ S_i \}_1 \rightarrow \{ S_i \}_2 \), is to use the rule that the change be accepted with a probability given by:

\[
\Pr (\{ S_i \}_1 \rightarrow \{ S_i \}_2) = e^{-\beta (E(\{ S_i \}_2) - E(\{ S_i \}_1))}, \quad E(\{ S_i \}_2) > E(\{ S_i \}_1)
\]

\[
\Pr (\{ S_i \}_1 \rightarrow \{ S_i \}_2) = 1, \quad E(\{ S_i \}_2) < E(\{ S_i \}_1).
\] (2.4)

This choice of probabilities is utilized by the Metropolis algorithm.

Now, consider a DE type Hamiltonian which is quadratic in fermion operators, such as the DEM given by Eq. (1.1). One can diagonalize \( H \) to find the single electron eigenstates and energies for a given configuration of classical spins:

\[
H (\{ S_i, a_i^\dagger, a_n \}) = \sum_n E_n (\{ S^z_i, \phi_i \}) a_n^\dagger a_n,
\] (2.5)

where \( a_n^\dagger \) and \( a_n \) are the creation and annihilation operators for the \( n \)th eigenstate of \( H \),

\[
a_n^\dagger = \sum_{i\sigma} \psi_{n\sigma}(i)c_{i\sigma}^\dagger, \quad a_n = \sum_{i\sigma} \psi^*_{n\sigma}(i)c_{i\sigma}.
\] (2.6)

Here \( \psi_{n\sigma}(i) \) is the eigenfunction of the \( n \)th state with spin \( \sigma \) at site \( i \), and there are \( N_0 = 2N_{lm} \) such states \( n \) that form a diagonal basis for \( H \). The partition function in the grand-canonical ensemble can be written as:

\[
Z = \prod_{i=1}^{N_{lm}} \int_{-1}^{1} dS_i^z \int_0^{2\pi} d\phi_i \sum_{n_1=0,1} \cdots \sum_{n_2N_{lm}=0,1} e^{-\beta \sum_{k=1}^{2N_{lm}} (E_k(\{ S^z_i, \phi_i \}) - \mu)n_k},
\] (2.7)

where \( \mu \) is the chemical potential and \( n_k \) is the occupation number of level \( k \). One can readily sum over the fermion degrees of freedom to obtain:

\[
Z = \prod_{i=1}^{N_{lm}} \int_{-1}^{1} dS_i^z \int_0^{2\pi} d\phi_i \prod_{n=1}^{N_0} \left( 1 + e^{-\beta (E_n(\{ S^z_i, \phi_i \}) - \mu)} \right).
\] (2.8)

The carrier free energy, \( F_c(\{ S^z_i, \phi_i \}) \), for a given configuration of classical spins is:

\[
F_c(\{ S^z_i, \phi_i \}) = -\frac{1}{\beta} \ln Z_e
\] (2.9)

\[
= -\frac{1}{\beta} \sum_{n=1}^{N_0} \log \left( 1 + e^{-\beta (E_n(\{ S^z_i, \phi_i \}) - \mu)} \right),
\]
Rewriting the partition function using the carrier free energy it takes the form:

\[
Z = \prod_{i=1}^{N_{\text{lm}}} \int_{-1}^{1} dS_i^t \int_0^{2\pi} d\phi_i e^{-\beta \mathcal{F}_c(S_i)},
\]  

which is precisely the form of Eq. (2.2). We can thus use the probability given by Eq. (2.4), but with the carrier free energy rather than the internal energy:

\[
\Pr(S_1 \rightarrow S_2) = e^{-\beta (\mathcal{F}_c(S_2) - \mathcal{F}_c(S_1))}, \quad \mathcal{F}_c(S_2) > \mathcal{F}_c(S_1)
\]

or

\[
\Pr(S_1 \rightarrow S_2) = 1, \quad \mathcal{F}_c(S_2) < \mathcal{F}_c(S_1).
\]

2.1.1 Thermodynamic properties

With an equilibrated sample of classical spins, we can measure both global and local quantities from the classical spin distribution and the single electron eigenstates. The moments of the average magnetization per classical spin with spin \(S\) are given by

\[
M^q = \left( \frac{1}{SN_{\text{lm}}} \sqrt{\sum_i S_i^2} \right)^q,
\]

while the moments of the average magnetization per carrier are

\[
m^q = \left( \frac{1}{2N_e} \sqrt{\sum_{i,\alpha\beta} \langle \hat{S}_{\alpha\beta}^i \rangle^2} \right)^q,
\]

where \(N_e\) is the number of electrons in the system. Using Eq. (2.12) we can compute the dimensionless Binder cumulant (BC) [53],

\[
g(N,T) = \frac{1}{2} \left[ 5 - 3 \left( \frac{\langle M^4 \rangle}{\langle M^2 \rangle^2} \right) \right],
\]

which is useful for determining the transition temperature. In the limit \(T \rightarrow \infty\), the BC tends to zero in the paramagnetic phase, while in the ferromagnetic phase it tends to unity. The important feature of the BC is that provided finite size effects are small, the BC is independent of system size at the transition temperature \(T = T_C\). For large enough \(N_{\text{lm}}\) such that finite size corrections are not important, one can then find \(T_C\) from the temperature at which there is simultaneous crossing of \(g(N_{\text{lm}}, T)\) vs \(T\) curves for several different values of \(N_{\text{lm}}\). Another quantity that can signal the presence of a phase transition is the magnetic susceptibility which is given by

\[
\chi_{\text{lm}} = \beta \left( \langle M^2 \rangle - \langle M \rangle^2 \right).
\]
The susceptibility diverges at the transition temperature in the infinite system size limit. In finite size systems, peaks in the susceptibility are usually less reliable than the BC for determining $T_C$ [35]. We also measure the density of states discretely by binning eigenvalues,

$$DOS(\varepsilon_i) = \frac{1}{\Delta \varepsilon} \int_{\varepsilon_i - \Delta \varepsilon/2}^{\varepsilon_i + \Delta \varepsilon/2} d\varepsilon \sum_{n=1}^{N_0} \delta(\varepsilon - \varepsilon_n), \quad (2.16)$$

where $\Delta \varepsilon$ is the width of the bins, and $\varepsilon_i$ is the central energy in the $i^{th}$ bin.

### 2.2 Model

As discussed in Chapter 1, to model the electronic and magnetic properties of specific materials in which double exchange physics takes place, one often needs to include terms such as those associated with bandstructure, disorder, lattice interactions, and Coulomb interactions. Previous work [46, 48, 49] considered Coulomb interactions in the DE model using ground-state, DMRG, or mean field (where the local moments are also treated at a mean field level) calculations. These techniques are either impractical in dimensions greater than one, or do not include the effects of fluctuations of the local moment spins. The Monte Carlo approach used here has no limit on dimensionality, although being a mean field theory in electron interactions, will be more accurate in higher dimensions, and takes into account the fluctuations of the spins, at the price of replacing quantum spins with classical vectors.

We include electron interactions and treat the classical spins exactly in the DEM with a Hubbard $U$ term that takes into account on-site Coulomb repulsion. The Hamiltonian we consider is

$$H = -t \sum_{\langle i,j \rangle} \left[ c_{i\sigma}^\dagger c_{j\sigma} + h.c. \right] + J \sum_i \mathbf{S}_i \cdot \left( c_{i\alpha}^\dagger \frac{1}{2} \mathbf{\tilde{\sigma}}_{\alpha\beta} c_{i\beta} \right) + U \sum_i n_{i\uparrow} n_{i\downarrow}. \quad (2.17)$$

Here $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ is the electron number operator at site $i$ for spin $\sigma$, and $U$ is the strength of the on-site Coulomb repulsion. There will also generically be terms associated with the long-range tail of the Coulomb interaction, which we have ignored here: these can be introduced by a straightforward generalization of our approach. The Hamiltonian [Eq. (2.17)] is quartic in fermion operators, and its eigenstates are many body states of $N_e$ electrons, $\Psi_n(\vec{r}_1, \ldots, \vec{r}_{N_e})$. There are $(2N_{lm})!/(N_e!(2N_{lm} - N_e)!)$ such states. The energy of the $n^{th}$ state is given by the solution of...
\[ H \Psi_n(\vec{r}_1, \ldots, \vec{r}_{N_e}) = E_n \Psi_n(\vec{r}_1, \ldots, \vec{r}_{N_e}). \quad (2.18) \]

It is important to note that the eigenvalue energy \( E_n \) is the energy of the many body eigenstate \( \Psi_n(\vec{r}_1, \ldots, \vec{r}_{N_e}) \).

### 2.2.1 Hartree-Fock

The challenge associated with treating the model given by Eq. (2.17) is that the Hilbert space of the Hamiltonian grows rapidly with the number of local moments \( N_{lm} \). In matrix form Eq. (2.17) is a \( (2N_{lm})!/(N_e!(2N_{lm} - N_e)!) \) square matrix, which is enormous even for modest values of \( N_{lm} \).\(^3\) This entails a choice: either treat the problem exactly with very few local moments, or treat a larger number of local moments with some approximations that reduce the size of the Hilbert space. We choose the latter option and use the Hartree-Fock (HF) approximation to reduce the problem to be quadratic in fermion operators. A standard HF decomposition of the Hubbard \( U \) term gives

\[
H_{HF} = -t \sum_{\langle i,j \rangle} \left[ c^\dagger_{i\sigma} c_{j\sigma} + h.c. \right] + J \sum_i \mathbf{S}_i \cdot (c^\dagger_{i,\alpha} \frac{1}{2} \sigma_{\alpha\beta} c_{i\beta}) \]

\[ + U \sum_i \left[ \langle n_{i\uparrow} \rangle c^\dagger_{i\downarrow} c_{i\downarrow} + \langle n_{i\downarrow} \rangle c^\dagger_{i\uparrow} c_{i\uparrow} - \langle c^\dagger_{i\uparrow} c_{i\downarrow} \rangle \langle c^\dagger_{i\downarrow} c_{i\uparrow} \rangle - \langle n_{i\uparrow} \rangle \langle n_{i\downarrow} \rangle + \langle c^\dagger_{i\uparrow} c_{i\downarrow} \rangle \langle c^\dagger_{i\downarrow} c_{i\uparrow} \rangle \right], \]

\[ \equiv H_t + H_J + H_{ee} + U \sum_i \left[ -\langle n_{i\uparrow} \rangle \langle n_{i\downarrow} \rangle + \langle c^\dagger_{i\uparrow} c_{i\downarrow} \rangle \langle c^\dagger_{i\downarrow} c_{i\uparrow} \rangle \right]. \quad (2.19) \]

The Hamiltonian after the HF decomposition is now quadratic in fermion operators. At zero temperature the eigenstates of the quadratic Hamiltonian are the many body Slater determinants of one-electron states, which are Fock states.

\[
\Psi_n(\vec{r}_1 \ldots \vec{r}_{N_e}) = \prod_{\{\alpha\}} |\phi_\alpha\rangle. \quad (2.20) \]

---

\(^3\)For example a \( 3 \times 3 \times 3 \) system has \( N_{lm} = 27 \) local moments. The Hamiltonian Eq. (2.17) for 5 electrons is a \( 3162510 \times 3162510 \) matrix.
The product is over only those \( \alpha \) that satisfy \( n_{n,\alpha} = 1 \), where \( n_{n,\alpha} \) is the occupation of the one-electron states \( |\phi_\alpha\rangle \) which satisfy

\[
[H_t + H_f + H_{ee}] |\phi_\alpha\rangle = \varepsilon_\alpha |\phi_\alpha\rangle,
\]

where for the ground state,

\[
\begin{align*}
n_{0,\alpha} &= 1, \ \alpha \leq N_e \\
n_{0,\alpha} &= 0, \ \alpha > N_e.
\end{align*}
\]

The total energy of the many electron Fock state is given by the Hamiltonian [Eq. (2.19)]:

\[
H_{HF}\Psi_n = E_n^{HF}\Psi_n.
\]

To find the states and its energies, one needs to diagonalize \([H_t + H_f + H_{ee}]\) to find the one-electron states, then construct the Fock state \(\Psi_n\) and calculate the total energy \(E_n\) from [Eq. (2.23)]. The total energy is then given by

\[
E_n^{HF} = \sum_\alpha \varepsilon_\alpha n_{n,\alpha} - U \sum_i \left[ \langle n_{i\uparrow}\rangle \langle n_{i\downarrow}\rangle - \langle c_{i\uparrow}^\dagger c_{i\uparrow}\rangle \langle c_{i\downarrow}^\dagger c_{i\downarrow}\rangle \right].
\]

From this expression, one can see that the total energy is not merely a sum of the energies \(\varepsilon_\alpha\) found from Eq. (2.21). This is because one must subtract the Hartree-Fock potential (the constant terms in Eq. (2.19)) from the total energy [54]. This implies that the spectrum \(\{\varepsilon_1, \varepsilon_2, \ldots, \varepsilon_N\}\) is not actually the spectrum of the one-electron states \(|\phi_\alpha\rangle\). In order for us to use the MC framework outlined previously, the spectrum of the one-electron states must be determined in order to calculate the carrier free energy [Eq. (2.9)]. Equation (2.24) can be rewritten in terms of the Hartree-Fock potential [55].

\[\text{Evidently, this Hamiltonian depends on the one-electron states themselves through the expectation values so self consistent one-electron states must be found. The procedure to find the self consistent states is discussed in Chapter 3.}\]
\[ E_n^{HF} = \sum_\alpha \varepsilon_\alpha n_{n,\alpha} - \frac{1}{2} \sum_\alpha V_{n,\alpha\alpha}^{HF} n_{n,\alpha} \]
\[ = \sum_\alpha \tilde{\varepsilon}_\alpha n_{n,\alpha}, \]
\[ V_{n,\alpha\beta}^{HF} = \frac{U}{2} \sum_i \sum_\gamma \left( \langle n_i \uparrow \rangle \psi_{i\alpha \uparrow}^* \psi_{i\beta \uparrow} \psi_{i\gamma \downarrow} \psi_{i\gamma \uparrow} - \langle n_i \downarrow \rangle \psi_{i\alpha \downarrow}^* \psi_{i\beta \downarrow} \psi_{i\gamma \uparrow} \psi_{i\gamma \uparrow} \right) n_{n,\alpha}, \]

and where the \( \psi_{i\alpha\sigma} \) are defined by
\[ a_{\alpha}^\dagger = \sum_i \psi_{i\alpha\sigma} c_{i\sigma}^\dagger, \]

where \( a_{\alpha}^\dagger \) is the creation operator for the one-particle state \( |\phi_\alpha\rangle \). The \( \tilde{\varepsilon}_\alpha \) are the one-electron state energies we are looking for, and can be expressed as
\[ \tilde{\varepsilon}_\alpha = \varepsilon_\alpha - \frac{1}{2} V_{\alpha\alpha}^{HF}. \]

Evaluating \( V_{\alpha\alpha}^{HF} \) leads to the final expression,
\[ \tilde{\varepsilon}_\alpha = \varepsilon_\alpha - \frac{U}{2} \sum_i \left( \langle n_i \uparrow \rangle \psi_{i\alpha \uparrow}^* \psi_{i\alpha \uparrow} + \langle n_i \downarrow \rangle \psi_{i\alpha \downarrow}^* \psi_{i\alpha \downarrow} \right. \]
\[ \left. - \langle c_{i\uparrow}^\dagger c_{i\downarrow} \rangle \psi_{i\alpha \uparrow}^* \psi_{i\alpha \downarrow} - \langle c_{i\downarrow}^\dagger c_{i\uparrow} \rangle \psi_{i\alpha \downarrow}^* \psi_{i\alpha \uparrow} \right). \]

To determine the one-electron state spectrum one must thus find the self consistent eigenstates and spectrum \( \{\varepsilon_1, \varepsilon_2, \ldots, \varepsilon_N\} \) using the Hamiltonian in Eq. (2.21), then calculate the single electron state spectrum, \( \{\tilde{\varepsilon}_1, \tilde{\varepsilon}_2, \ldots, \tilde{\varepsilon}_N\} \), using Eq. (2.29). The term subtracted in Eq. (2.29) corrects for the double counting of the interaction potential energy in Eq. (2.21).

### 2.3 Exact diagonalization study

The HF approximation is uncontrolled, so while there is a well defined HF carrier free energy for every local moment spin configuration and a MC simulation will obey detailed
balance, there is no guarantee that the behaviour will be the same as the original interacting problem of interest. The strategy that we used to try to minimize this systematic error arising from the HF approximation was to use comparisons of exact diagonalization and HF on small systems at zero temperature to determine in which regions of parameter space the HF approximation gives accurate results. We review the exact diagonalization calculations and their comparison with the results of the HF approximation in this section.

To find the exact eigenvalues and eigenstates of Eq. (2.17) we construct a matrix representation of Eq. (2.17) using basis states for $N_e$ electrons. The basis states used are states where the electrons are localized on specific sites and have spin up ($\uparrow$) or down ($\downarrow$)

$$\langle \varphi_{\alpha} \rangle = | n_{1\uparrow}, n_{1\downarrow}, n_{2\uparrow}, n_{2\downarrow}, \ldots, n_{N_{lm}\uparrow}, n_{N_{lm}\downarrow} \rangle$$

where the occupation numbers $n_{i\sigma}^\alpha$ satisfy the conditions

$$n_{i\sigma}^\alpha = 0 \text{ or } 1; \quad \sum_{i, \sigma} n_{i\sigma}^\alpha = N_e.$$  \hspace{1cm} (2.32)

We treat the local moment spins as classical vectors since we will also do this in our MC simulation. For a given spin configuration the elements of the Hamiltonian matrix can be found from

$$H_{\alpha\beta} = \langle \varphi_{\alpha} | H | \varphi_{\beta} \rangle.$$  \hspace{1cm} (2.33)

Care must be taken in evaluating the contribution from the hopping term in the above expression, as the creation and annihilation operators satisfy the anti-commutation relations

$$\{c_i^\dagger, c_j^\dagger\} = \{c_i, c_j\} = 0 \quad , \quad \{c_i^\dagger, c_j\} = \delta_{ij}.$$  \hspace{1cm} (2.34)

Although the magnitude of the term will be $t$, the sign of the term will depend on the number of commutations involved in permuting the operators from the hopping term in the
Hamiltonian through the basis state $|\psi_\beta\rangle$.

The eigenstates of the Hamiltonian [Eq. (2.17)] are then linear combinations of the many body basis states

$$\Psi_n = \sum_\alpha \Phi_{n\alpha} |\varphi_\alpha\rangle,$$

with energies given by Eq. (2.18). The electron density at site $i$ with spin $\sigma$ for the $n^{\text{th}}$ eigenstate is then given by

$$\rho_{n,i\sigma} = \sum_\alpha |\Phi_{n\alpha}|^2 n_{i\sigma}^\alpha,$$

(2.35)

For the HF method, the single electron states and energies are found using Eq. (2.21) and Eq. (2.29). The total energy of the many body state is then calculated using Eq. (2.25). From the definition [Eq. (2.27)] of $\psi_{i\alpha\sigma}$, the electron density at site $i$ with spin $\sigma$ for the $n^{\text{th}}$ eigenstate is

$$\rho_{HF, n, i\sigma} = \sum_\alpha \psi_{i\alpha\sigma}^* \psi_{i\alpha\sigma} n_{n,\alpha},$$

(2.37)

where $n_{n,\alpha}$ was defined in Eq. (2.20).

To see how the HF method compares to exact diagonalization, we compute the relative error in the energies of the ground state, and first few excited states

$$\Delta E_n = \frac{|E_n - E_n^{HF}|}{|E_n|},$$

(2.38)

and the average relative error in the electron densities

$$\Delta \rho = \frac{1}{2N_{lm}} \sum_{i,\sigma} \frac{|\rho_{n,i\sigma} - \rho_{HF, n,i\sigma}|}{|\rho_{n,i\sigma}|}. $$

(2.39)

2.3.1 Results

To determine where our scheme should give the most accurate results in the parameter space of the model, we computed the relative errors (Eqs. (2.38) and (2.39)) for the first
three energy levels and ground state densities for various system sizes, classical spin configurations, doping, and values of $U$ and $J$. For all calculations we express $U$ and $J$ in units of $t$.

Figures 2.1, 2.2, and 2.3 show the relative error in the first three energy levels for an 8 site $2 \times 2 \times 2$ lattice with $N_e = 2, 3, 4$, and the local moments ferromagnetically, antiferromagnetically, and averaged over 25 random spin configurations. Figures 2.4 and 2.5 show the relative error in the first three energy levels and densities for a $3 \times 2$ lattice with $N_e = 2, 3, 4$, averaged over 50 random spin configurations. The general trend for all cases is the same: when $J/U$ is small there are significant errors in the energies, whereas for $U$ of order $J$ there is good agreement between the approaches. In Fig. 2.1, the abrupt changes in relative error at large $U/J$ arise from the flipping of the spin of one of the ground state electrons. It is interesting to note that in Figs. 2.4 and 2.5 even though the HF approximation gets the ground state densities correct for $J = 0$, there remains a discrepancy in the ground state energies.

In Fig. 2.6 we show the region in parameter space for which the relative error in all exact diagonalizations is less than 5%. The condition $U \leq 3J$ would thus appear to be reasonable to define values of $U$ for MC simulations. Within this region, when $J/U \gg 1$, the strong exchange coupling tends to spin polarize the fermions at low temperatures, and we would expect the interactions to have little impact. At very small $J/t$, HF provides accurate results for only small $U/t$. The region which appears the most interesting to study appears to be for moderate $J/t$ and $U/t \approx 3J/t$, where the exchange coupling has not completely spin polarized the fermions.
Figure 2.1: Contour plot of the relative error (given by Eq. (2.38)) between the ground state (GS), first excited state (ES) and second excited state (SES) energies calculated using HF and exact diagonalization for $N_e = 2, 3$ and 4 on a $2 \times 2 \times 2$ lattice with the local moments FM aligned.
Figure 2.2: Contour plot of the relative error (given by Eq. (2.38)) between the ground state (GS), first excited state (ES) and second excited state (SES) energies calculated using HF and exact diagonalization for $N_e = 2, 3$ and 4 on a $2 \times 2 \times 2$ lattice with the local moments AF aligned.
Figure 2.3: Contour plot of the relative error (given by Eq. (2.38)) between the ground state (GS), first excited state (ES) and second excited state (SES) energies calculated using HF and exact diagonalization for $N_e = 2, 3$ and $4$ on a $2 \times 2 \times 2$ lattice averaged over 25 random spin configurations.
Figure 2.4: Contour plot of the relative error (given by Eq. (2.38)) between the ground state (GS), first excited state (ES) and second excited state (SES) energies calculated using HF and exact diagonalization for $N_e = 2, 3$ and 4 on a $3 \times 2$ lattice averaged over 50 random spin configurations.
Figure 2.5: Contour plot of the relative error (given by Eq. (2.39)) between the ground state electron densities calculated using HF and exact diagonalization for $N_e = 2$, 3 and 4 on a $3 \times 2$ lattice averaged over 50 random spin configurations.
Figure 2.6: Plot showing the region in parameter space where we suggest one can have confidence in using HF within a MC simulation. The blue region indicates where the relative error in the ground state energy did not exceed 5% in any of our calculations with more than 2 electrons. All of the data presented in Chap. 4 lie in the hatched region, $U < 3J$. 
Chapter 3

Simulation Details

In Chapter 2 we discussed the formalism we use to perform a Monte Carlo simulation of the Hamiltonian [Eq. (2.17)], as well as the global and local quantities of interest for the study. The simulation procedure may seem straightforward at first glance, it is worthwhile to discuss the implementation of the algorithm. Given an equilibrated sample of the system and chemical potential (μ), one can simply sample a Markov chain constructed via the Metropolis algorithm and determine the equilibrium average of any quantity of interest. In practice, a time consuming part of MC simulation of a spin systems is establishing that a particular system has equilibrated. The coupling of fermions to the spins leads to an additional level of complexity as compared to regular classical MC simulations as the chemical potential of the fermions must be equilibrated along with the properties of the classical spins. In this Chapter we discuss the technical details of the simulation. The general structure of the code will first be presented, followed by a discussion of the equilibration procedure and the algorithm for calculating the free energy for a particular spin configuration using iterative HF. Some procedures used to optimize the code are discussed, and we conclude with a discussion of testing the algorithm and simulation on a toy model which admits an exact solution.

3.1 General Properties

We first discuss some general properties of the simulation and introduce terminology that is used in the remainder of this Chapter.

We define a sample as a dynamic spin configuration corresponding to a particular thermal
Simulation Details

<table>
<thead>
<tr>
<th>$N_x, N_y, N_z$</th>
<th>Dimensions of the cubic lattice, there is a local moment at each lattice site, so the whole system thus contains $N_{lm} = N_x N_y N_z$ local moments</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N_e$</td>
<td>Number of electrons in the system</td>
</tr>
<tr>
<td>$t \equiv 1$</td>
<td>Nearest neighbour hopping amplitude</td>
</tr>
<tr>
<td>$J$</td>
<td>On-site exchange coupling</td>
</tr>
<tr>
<td>$U$</td>
<td>On-site Hubbard U term</td>
</tr>
<tr>
<td>$T$</td>
<td>Temperature</td>
</tr>
</tbody>
</table>

Table 3.1: Sample simulation input parameters

history from which we take data for a given set of fixed input parameters. For one simulated sample the parameters summarized in Table (3.1) must be specified. In the simulation we set $k_B = 1$ and measure $J$, $U$, and $T$ in units of the nearest neighbour hopping amplitude $t$. Periodic boundary conditions are used for the hopping term, and we assume the classical vector spins to have unit amplitude, since any other amplitude can be absorbed into $J$.

With the parameters above, the simulation modifies the spin configuration of a sample according to the algorithm described in Sec. 2.1. In a spin flip, one spin is randomly selected from the lattice and the simulation attempts to randomly reorient its spin. the move is accepted or rejected according to the probability given by Eq. (2.11). We define a MC sweep as performing $N_{lm}$ such spin flips.

3.2 Calculating the Free Energy of a Particular Spin-Configuration

For each spin-flip performed in the simulation, one must calculate the free energy [Eq. (2.9)] of the both the initial and proposed spin configurations in order to make a decision on whether to accept or reject the new configuration. In order to calculate the free energy one must find the self consistent eigenfunctions of the Hamiltonian given by Eq. (2.21), and calculate the single electron energy spectrum using Eq. (2.29). We implement an iterative procedure in order to achieve this.

To begin the iterative method for a spin flip, we first need a guess for the fermion expectation values in the Hamiltonian [Eq. (2.19)]. For the first spin flip we use the densities produced by the Hamiltonian, for the given local moment configuration, with $U$ set to zero. For subsequent spin flips, the densities found from the previous spin configuration are used.
as the first guess. The Hartree-Fock Hamiltonian is constructed and diagonalized to obtain its eigenstates and spectrum. The following correlations can then be evaluated:

$$\langle c_{i\alpha}^\dagger c_{j\alpha'} \rangle = \sum_\alpha f(\tilde{\varepsilon}_\alpha) \psi_{i\alpha\sigma}\psi_{j\alpha'\sigma'},$$  \hspace{1cm} (3.1)

where $f(\tilde{\varepsilon}_\alpha)$ is Fermi occupation of the $\tilde{\varepsilon}_\alpha$ level. The Hamiltonian [Eq. (2.19)] is then constructed with the desired value of $U$ using the expectation values calculated from Eq. (3.1). This process continues until two successive iterations give equal densities; that is, the densities calculated from the spectrum and eigenstates of the Hamiltonian are equal to the densities that were used in constructing the Hamiltonian.

**Figure 3.1:** At finite temperatures, two nested iterative loops are used in the procedure to find the HF single electron energies.

From close examination of the expression for the correlations in Eq. (3.1) and the expression for the energy spectrum [Eq. (2.29)], one can see that the expectation values depend on the spectrum via the Fermi function, and the spectrum depends on the expectation values through the HF potential term. To speed up convergence another iterative loop in
the procedure is added which ensures that the spectrum calculated using the expectation values gives back the same expectation values when substituted into Eq. (3.1). Therefore the overall iterative HF procedure has two nested iterative loops, as shown in Fig. 3.1.

3.3 Equilibration Procedure

To simulate the sample in the Grand Canonical Ensemble we must have a suitable value for the chemical potential \( \mu \), and be confident that our sample is properly equilibrated. We utilize a equilibration procedure, originally developed by Kennett et al. [35], with which we can equilibrate a sample and determine an appropriate \( \mu \) simultaneously.

The equilibration procedure begins by constructing two initial replicas of the system. For the first replica all of the local moments are set to be random implying the replica has a very small average magnetization per spin (see Eq. (2.12)). For the second replica we set the local moment spins to be all aligned in the positive \( z \) direction. This second replica thus has an initial magnetization per local moment spin of unity. We evolve both the replicas independently until their global properties match to within predefined tolerances, at which point we deem the replicas have equilibrated. Since we do not know \( \mu \) initially, we set it for each replica according to a self-consistent solution of the equation

\[
N_e = \sum_n \frac{1}{e^{\beta(\tilde{\varepsilon}_n - \mu)} + 1},
\]

(3.2)

where \( \tilde{\varepsilon}_n \) are the energies calculated using Eq. (2.29) given the spin configuration of the replica.\(^1\) After every 4 MC sweeps, we use the condition Eq. (3.2) to calculate new estimates of \( \mu \) for each replica.

In a previous study [35], unbiased results were obtained by equilibrating until the running average of the global magnetization, global magnetization squared, and the chemical potential from both replicas were within a few percent (\( \sim 0.5\% \)) of each other. Using only the above criteria alone can lead to very long equilibration times, particularly at high temperatures, where there are large oscillations in the instantaneous global magnetization. Another convergence criteria known as the \( z \)-test [56] was added to the code in parallel to the criterion above, meaning that only one of the criteria must be met for the simulation to proceed to taking data. The \( z \)-test takes two different distributions (which may or may not

\(^1\)One can use a variation of the bisection search algorithm to calculate the chemical potential from the calculated spectrum.
have the same variance) and computes the probability that the two different distributions have the same expectation value in the limit of infinite measurements. If we have taken $N$ measurements of an observable $A$ for the two replicas the $z$-number for the observable $A$ is computed as:

$$z = \sqrt{N} \frac{\langle A \rangle_1 - \langle A \rangle_2}{\sqrt{\sigma^2_{\langle A \rangle_1} + \sigma^2_{\langle A \rangle_2}}}$$

(3.3)

Here $\langle A \rangle$ is the running average of the observable $A$, calculated every $4N$ MC sweeps. The double sided integral from $\pm(z, \infty)$ of the standard normal distribution,

$$P = \int_{-\infty}^{z} \frac{1}{\sqrt{2\pi}} e^{-x^2/2} + \int_{z}^{\infty} \frac{1}{\sqrt{2\pi}} e^{-x^2/2},$$

(3.4)

gives the probability that both distributions share the same expectation value. The convergence criteria we use is that the $z$-number for the running averages is less than 0.15, which corresponds to a 0.95 probability of exact convergence. Using more stringent convergence criteria for the $z$-number did not seem to improve the results. Once the replicas have converged, only one of the replicas is retained and used to take equilibrium measurements, with its $\mu$ fixed at the value of the running average calculated in the equilibration procedure.

The $z$-test is a more systematic approach, but as mentioned earlier, a previous study found the first criteria to be sufficient to establish equilibration. The benefit in including both is that each criterion has limitations in certain temperature ranges. At low temperatures the global magnetization has very small fluctuations which are not necessarily gaussian which can lead to a very large $z$-value. At low temperatures the first criteria in general will be satisfied much sooner than the $z$-test and yields very accurate results for an exactly solvable toy model, as discussed in Sec. 3.6. At high temperatures there are large fluctuations in the global magnetization which tend to be gaussian. The first criteria implies a very long time for equilibration as the variance of the global magnetization is very large, whereas the $z$-test is generally satisfied in a much more reasonable amount of time.

Figure 3.2 shows an example of how the magnetization and chemical potential vary in the two replicas at the start of the equilibration procedure. A point to note about the equilibration procedure is that we ignore initial transients and only begin to compute the running averages of global magnetization and $\mu$ once the magnetizations of the two replicas become comparable.
Figure 3.2: An example of the evolution of the average magnetization and chemical potential for the two replicas. The magnetization is modified with each spin flip whereas the chemical potential used for each replica is updated every 4 MC sweeps ($4N_{lm}$ spin flips). The region where we start to calculate the running averages of the magnetization and $\mu$ in the equilibration procedure is shaded. Data is from equilibration procedure for the model Eq (2.17) on a $4 \times 4 \times 4$ lattice with $N_e = 8$, $T/t = 0.02$, $J/t = 5$, and $U/t = 0$.

One can never completely guarantee that a particular sample is equilibrated long enough for the procedure to be successful. While taking data on an equilibrated sample, the code calculates the number of electrons after each accepted spin-flip using Eq. (3.2). If the simulation is run for long enough and we have the correct value of $\mu$, then $\langle N_e \rangle_s$ will take the value we used to determine $\mu$. If the calculated value of $\langle N_e \rangle_s$ deviates significantly from the specified value of $N_e$, this indicates that the sample was not fully equilibrated and this data is excluded in our averaging. This procedure was used by Kennett et al. [35] and was found to give excellent results when applied to the toy model of Sec. 3.6.
3.4 Statistical Analysis

For a distribution of measurements over an equilibrated sample, the expectation value of an observable A and the error associated with the expectation value are given by the mean of the distribution and the variance of the mean:

\[
\langle A \rangle = \frac{1}{N_M} \sum_{k=1}^{N_M} A_k
\]

\[
\sigma_{\langle A \rangle} = \sqrt{\frac{1}{N_M(N_M - 1)} \sum_{k=1}^{N_M} (A_k - \langle A \rangle)^2},
\]

where \(N_M\) is the number of data points taken from the sample, and the subscript \(k\) refers to the \(k^{th}\) data point, \(k = 1 : N_M\). Averages over one sample are denoted by angled brackets \(\langle ... \rangle\). The error associated with the Binder Cumulant [Eq. (2.14)] is given by standard propagation of errors:

\[
\sigma_G = \frac{3}{2} \sqrt{\left(\frac{\sigma_{\langle M^4 \rangle}}{\langle M^2 \rangle^2}\right)^2 + 4 \left(\frac{\langle M^4 \rangle \sigma_{\langle M^2 \rangle}}{\langle M^2 \rangle^3}\right)^2}.
\]

As discussed in Sec. 3.3, we equilibrate \(\mu\) in order for \(\langle N_e \rangle_s\) to be equal to the specified \(N_e\). If we run the data taking for long enough we would expect to achieve very small statistical errorbars for \(\langle N_e \rangle_s\). In practice the value of \(\langle N_e \rangle_s\) we calculate for a particular sample is never exactly equal to \(N_e\), and it is common to find deviations that vary from sample to sample. These deviations are not the statistical variances of the measurements of \(N_e\), which we expect in the grand canonical ensemble, but rather an error in the mean value, \(\langle N_e \rangle_s\) from not equilibrating for long enough. We observed that for some sets of input parameters (Table 3.1) the expectation values of measured global parameters can sometimes, particularly close to \(T_C\), be significantly affected by even slight deviations from the desired \(N_e\). We therefore simulate multiple samples for a particular set of inputs. If \(N_M\) measurements are taken from \(N_S\) samples, the average over samples, denoted by angled brackets with a bar \(\langle ... \rangle\), for an observable \(A\) is given by

\[
\langle A \rangle = \frac{1}{N_S} \sum_{j=1}^{N_S} \langle A \rangle_j,
\]

where \(j\) refers to the \(j^{th}\) sample, \(j = 1 : N_S\). The variance of the mean is then given by

\[
\sigma_{\langle A \rangle} = \sqrt{\frac{1}{N_S(N_S - 1)} \sum_{j=1}^{N_S} (\langle A \rangle_j - \langle A \rangle)^2}.
\]
As mentioned previously, if the deviations from \( N_e \) for a particular sample are large enough, we exclude the data from that sample in averages over samples.

### 3.5 Optimization

The simulation developed is very computationally intensive. A typical run of the simulation for a given set of parameters [Table 3.1] requires at least about ten thousand MC sweeps. Due to the iterative HF procedure (Sec. 3.2), each spin flip involves diagonalizing the Hamiltonian Eq. (2.21) to reach self consistency. The number of iterations required is usually between 5 and 10, although occasionally a large number of iterations (> 25) is required. For illustration purposes we assume an average of 10 iterations per self-consistency loop. A reasonable estimate for the number of times the full Hamiltonian must be diagonalized is then given by \( \sim 10^5 N_{lm} \). The matrix representation of the Hamiltonian has dimensions \( 2N_{lm} \) by \( 2N_{lm} \); the computational cost of diagonalizing the matrix then scales as \( \sim (2N_{lm})^2 \). Putting this all together, the overall cost of running a simulation goes as \( \sim 4 \times 10^5 N_{lm}^3 \), a remarkably large number for even modest system sizes, although still much less than exact diagonalization of the electron problem for each spin flip.

In order to reduce the time taken to simulate a sample so that larger systems sizes can be investigated, several algorithms were added to improve the speed of the code. Great care was taken to ensure that these additions did not affect the results of the simulation procedure.

To reduce the computational cost of diagonalizing the Hamiltonian Eq. (2.21), we truncate the Hilbert space, ignoring high energy states, and include only states which contribute significantly to the carrier free energy [Eq. (2.9)]. The procedure is as follows; at the beginning of each MC sweep of the sample the Hamiltonian is diagonalized to find all of the \( 2N_{lm} \) eigenvalues and eigenvectors. The Fermi occupation of each eigenvalue is then calculated to determine the integer \( k \) which satisfies \( f(\beta(\tilde{\epsilon}_k - \mu)) < 10^{-43} \). There are two reasons this particular tolerance was chosen; first it ensures a negligible contribution to the free energy, and second an \( f(\beta(\tilde{\epsilon}_k - \mu)) < 10^{-43} \) corresponds to a \( f(\beta(\tilde{\epsilon}_k - \mu)) = f(99) \). The largest exponential which can be evaluated on a 64 bit machine is roughly \( e^{99} \). For each subsequent spin flip in the MC sweep, only the first \( k \) eigenvalues and eigenvectors are calculated. The computational savings will depend on the temperature and \( N_e/N_{lm} \). \( k/2N_{lm} \) typically ranged from \( \sim 1/5 \) at low temperatures to \( \sim 1/2 \) at high temperatures for \( N_e/N_{lm} = 1/8 \).
The diagonalization procedure has two steps, first the matrix is converted into tridiagonal form, and second the eigenvalues and eigenvectors are found from the tridiagonal matrix. The computational cost of each step goes as $2N_{lm}$. With the truncation of the Hilbert space we save time on the second step, reducing the computational cost to $(k/2N_{lm})2N_{lm} = k$, and thus the overall cost of the diagonalization goes as $2N_{lm}k$. The fact that $k$ increases with temperature implies there is a greater saving in computation time for low temperature samples.

Figure 3.3: An example of restricting the the value of the randomly generated new spin vector to lie within an angle $\theta$ of its initial vector. Here the initial spin vector is given by $\mathbf{S} = (S_z, \phi) = (1/2, 0)$, and $\theta = \pi/2$. The red points are a distribution of randomly selected vectors from the unit sphere, whereas the blue points are the distribution of random vectors restricted to an angle of $\theta$ about the initial spin vector.

In a spin flip, a spin is randomly selected from the lattice and its spin is chosen at random. Rather than randomly selecting a new classical spin vector from the entire unit sphere, we found that the equilibration procedure can be sped up by restricting the new spin vector to lie on the surface of the unit sphere within an angle $\theta$ of the initial spin vector. The value of this angle is continuously modified in the equilibration procedure to attempt to obtain a 50% spin flip acceptance rate. The point of this procedure is so as not to spend most of the time rejecting spin flips at low temperature because of their high energy cost. Figure 3.3 shows an example of how the new value of the classical spin is randomly
generated. The random points for each distribution are evenly distributed over the specified regions, so that in the limit of a large number of MC sweeps $\vec{S}$ is allowed to wander over the entire surface of the unit sphere so that there is unbiased sampling over the surface of the sphere.

The majority of the data presented in Chapter 4 was taken on the high performance computing system of the Western Canada Research Grid (WestGrid). The processors used ran between 2.66 and 3.00 GHz with 2GB memory. With the optimizations included, between 6 to 24 hours were required to equilibrate and take 2000 data points for the model Eq. (2.17) on a $4 \times 4 \times 4$ lattice with $N_e = 8$ and non-zero $U$. The same data taken for a $6 \times 6 \times 6$ lattice required between 240 to 500 hours.

3.6 Testing on a Toy Model

To test the algorithm and simulation, we calculated the thermodynamic properties of a simple non-interacting model of classical spins coupled to fermions, introduced in Ref. [35], which admits an exact solution for a one-dimensional chain with periodic boundary conditions

$$H = - \sum_{i,j} t_{ij} \left[ c_{i\sigma}^\dagger c_{j\sigma} + h.c. \right] + \frac{J}{N_{lm}} \sum_i S_i \cdot \sum_{j,\alpha,\beta} c_{j,\alpha}^\dagger \frac{1}{2} \sigma_{\alpha\beta} c_{j,\beta}. \quad (3.9)$$

In this model the electrons are equally strongly coupled to all classical spins in the chain, and the exchange interaction scales as $1/N_{lm}$ to ensure that the energy is extensive. Details of the exact solution (also presented in Ref. [35]) are given in Appendix A. A comparison of the exact results to those found by the simulation for $M(T)$ are shown in Fig. 3.4.

One can see from Fig. 3.4 that the simulation provides very accurate results for the toy model, which gives us confidence to move forward and simulate the model of interest [Eq. (2.19)].
Figure 3.4: Comparison of the exact result (line) and MC results (points) for the magnetization of the toy model calculated for a 20 site chain with \( N_e = 3 \) and \( t = J = 1 \). Errorbars are on the order of the size of the points. Each point represents an average over 1000 data points for 1 sample.
Chapter 4

Results

In this Chapter we present the results obtained from our MC simulation of the model given by Eq. (2.17) using the scheme outlined in Chapters 2 and 3. We first took data with $U = 0$, presented in Sec. 4.1, to provide a baseline with which to compare $U \neq 0$ data. We present results for finite $U$ in Sec. 4.2. In Sec. 4.3 we give a discussion of our results and their interpretation. The data we show in this Chapter were all taken on $4 \times 4 \times 4$ and $6 \times 6 \times 6$ lattices. Larger system sizes were computationally inaccessible. Nevertheless our system sizes are as large as simulations performed on non-interacting systems [35, 37, 38, 58].

Each point on any particular figure presented in this Chapter represents an average over 5 to 10 samples with 2000 data points taken for each sample for the $4 \times 4 \times 4$ data, and an average over 800-2000 data points for one to two sample was used for the $6 \times 6 \times 6$ data. If not visible in the figures, errorbars for $4 \times 4 \times 4$ data are on the order of thickness of the lines, and on the order of thickness of the points for $6 \times 6 \times 6$ data.

4.1 DE model without interactions, $U = 0$

In this section we calculate the magnetization of the DEM for a variety of $J/t$ values and show how $T_C$ can be extracted via the Binder cumulant. Previous studies have shown that $T_C$ is enhanced by increasing $N_e/N_{lm}$ for $N_e \lesssim N_{lm}$ [58]. Figure 4.1 shows the magnetization as a function of temperature obtained for a $4 \times 4 \times 4$ lattice with $N_e = 8$ for values of $J/t$ ranging from 0.5 to 10.0, and Fig. 4.2 shows the magnetization as a function of temperature obtained for a $4 \times 4 \times 4$ lattice with $J/t = 1.0$ and $N_e = 8$ and $N_e = 29$. The results have very small errors, and show very smooth transitions from a low temperature FM state to a
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high temperature PM state. At high temperatures, the magnetization saturates at a finite positive value which is independent of the ratio $J/t$. This value should be given by

$$\left\langle \frac{1}{N_{lm}} \sqrt{\sum_i S_i^2} \right\rangle \approx \frac{1}{N_{lm}} \sqrt{N_{lm}} \approx \frac{1}{\sqrt{N_{lm}}}.$$ (4.1)

From the figures, there is a clear enhancement of the magnetization with increasing $J/t$ and increasing $N_e/N_{lm}$ for $N_e < N_{lm}$.

Figure 4.1: Local moment magnetization as a function of temperature for a $4 \times 4 \times 4$ lattice with varying $J$, $U = 0$ and $N_e = 8$.

We plot global magnetization and Binder cumulant for $4 \times 4 \times 4$ and $6 \times 6 \times 6$ lattices with $J = 8t$, $U = 0$ and $N_e = 8$ in Figs. 4.3 and 4.4. The magnetization plot, Fig. 4.3, shows that there is a noticeable sharpening of the transition for the larger lattice. This behaviour is also apparent in the Binder cumulant plot, where the larger lattice has a higher value at low temperatures, but experiences a sharper drop close to the transition. We use the crossing point of the Binder cumulant curves to estimate $T_C/t = 0.042 \pm 0.005$. 
Figure 4.2: Local moment magnetization as a function of temperature for a $4 \times 4 \times 4$ lattice with $J/t = 1$, $U/J = 0$ and $N_e = 8$ and $N_e = 29$.

Figure 4.3: Local moment magnetization as a function of temperature for $4 \times 4 \times 4$ and $6 \times 6 \times 6$ lattices with $J = 8t$, $U = 0$ and $N_e/N_{lm} = 1/8$. 
4.2 DE model with finite interactions, $U \neq 0$

Having established some of the properties of the DE model with $U = 0$, we now present our results for the model given by Eq. (2.17) with finite $U/t$ with $N_e/N_{lm} = 1/8$ and at three different values of $J/t$ corresponding to three different situations for energy of the states near the Fermi energy with $N_e/N_{lm} = 1/8$. In the first kinetic energy dominates $J/t = 1.0$ (Sec. 4.2.1), in the second the energy is split nearly evenly between exchange interactions and kinetic energy $J/t = 5.0$ (Sec. 4.2.2), and third where the exchange dominates $J/t = 10.0$ (Sec. 4.2.3). We discuss attempts to determine $T_C$ as a function of $U$ for $J/t = 5$ in Sec. 4.2.4.

4.2.1 $J = t$

Figure 4.5 shows the local moment magnetization as a function of temperature for $U/J$ ranging from 0 to 3 for $N_e = 8$. There is a clear enhancement of the magnetization with increasing $U/J$ at temperatures in the vicinity of $T_C$. This behaviour is even more evident
in the carrier magnetization plotted in Fig. 4.6. At high temperatures (∼T_C) U certainly enhances the carrier magnetization, but has little effect at low temperatures (∝ T_C), and the carrier magnetization saturates at a value which is independent of U/J. The low temperature saturation value of the carrier magnetization depends on J/t, and we expect will decrease (towards −0.5) with increasing J/t (this is confirmed in Secs. 4.2.2 and 4.2.3). Figure 4.7 shows the local moment magnetization versus U/J for several temperatures. This plot confirms that there is enhancement of the magnetization with increasing U/J for high temperatures, and little effect at low temperatures.

Figures 4.8 to 4.10 show the density of states (DOS) for J/t = 1 at several different temperatures and values of U/J. We determine the DOS by binning the eigenvalues found after each spin flip during the data taking; for the data presented here we averaged over 384000 sets of eigenvalues. The bins are then normalized by the width of the interval and the number of spin flips.

Figure 4.8 shows the DOS for U/J = 0 at several temperatures. We can infer the spin of the electrons occupying each peak by the shift in the position of the peak as the temperature is increased. We will call spins anti-parallel to the magnetisation of the local moments up-spins, and spins parallel to the magnetisation of the local moments down-spins. The two small peaks at low energy are clearly associated with the spin split ground state of the hopping term (with kinetic energy ≈ −6t in the absence of the exchange term), the lowest energy peak corresponding to the state with the up-spin electron (with energy ≈ −6t − J/2), while the other has a down-spin (with energy ≈ −6t + J/2). The two larger high energy peaks also correspond to states with opposite electron spins (corresponding to energies ≈ −4t ± J/2). A clear decrease of the gaps separating the peaks with equal kinetic energy but opposite spin and broadening of the peaks with increasing T is observed which is associated with the loss of alignment of the local moments.

Figures 4.9 and 4.10 show the density of states for 3 values of U/J for T = 0.001 and T = 0.020 respectively. As the interactions are increased, the states corresponding to electrons with down-spin are increased in energy to a greater extent than the up-spin states, and can thus push the down-spin states above the chemical potential to higher energies. This naturally explains the observed enhancement of the magnetization with increasing U and is consistent with the data for the carrier magnetization in Fig. 4.6 which shows enhancement of the carrier magnetization with increasing U.
Figure 4.5: Local moment magnetization as a function of temperature for a $4 \times 4 \times 4$ lattice with $J = t = 1$ and $N_e = 8$.

Figure 4.6: Carrier magnetization as a function of temperature for a $4 \times 4 \times 4$ lattice with $J = t = 1$ and $N_e = 8$. 
Figure 4.7: Local moment magnetization as a function of $U/J$ at several temperatures for a $4 \times 4 \times 4$ lattice with $J = t = 1$ and $N_e = 8$. 
Figure 4.8: Density of states at several temperatures for a $4 \times 4 \times 4$ lattice with $J = t = 1$, $U/J = 0$, and $N_e = 8$. The vertical colored line represents the average value of $\mu$ calculated for the samples used to make the figure.
Figure 4.9: Density of states at $T = 0.001$ for several values of $U/J$ for a $4 \times 4 \times 4$ lattice with $J = t = 1$ and $N_e = 8$. The vertical colored line represents the average value of $\mu$ calculated for the samples used to make the figure.
Figure 4.10: Density of states at $T = 0.020$ for several values of $U/J$ for a $4 \times 4 \times 4$ lattice with $J = t = 1$ and $N_e = 8$. The vertical colored line represents the average value of $\mu$ calculated for the samples used to make the figure.
In Fig. 4.11 we show the local moment magnetization as a function of temperature for $U/J$ ranging from 0 to 1.25 for $N_e = 29$. The magnetization appears to be affected by finite $U$ in a similar manner to $N_e = 8$, as is also illustrated in plots of the local moment magnetization at fixed temperature with varying $U/J$ in Fig. 4.12. Figure 4.12 also indicates that there is perhaps a very small suppression of ferromagnetism with increasing interaction strength at low temperatures.

Figure 4.11: Local moment magnetization as a function of temperature for a $4 \times 4 \times 4$ lattice with $J = t = 1$ and $N_e = 29$. Errorbars are on the order of thickness of the lines.
Figure 4.12: Local moment magnetization as a function of $U/J$ at several temperatures for a $4 \times 4 \times 4$ lattice with $J = t = 1$ and $N_e = 29$. 
4.2.2  $J = 5t$

In Figs. 4.13 and 4.14 we plot the local moment magnetization and carrier magnetization for a $4 \times 4 \times 4$ lattice for several values of $U/J$ and for $J/t = 5$. We observe some enhancement of the magnetization at low and high temperatures due to the interactions (More at low than high temperatures). The plot of the carrier magnetization [Fig. 4.14] shows a significant enhancement of the carrier magnetization with $U/J$, notably at low temperatures where the low temperature saturation point decreases significantly with $U/J$, in contrast to the saturation value for $J/t = 1$ which remained relatively unchanged by the interactions.

In Fig. 4.15 we plot the local moment magnetization versus $U/J$ for several temperatures. There is a 5-10% enhancement of the magnetization with interactions for all of the temperatures shown.

Figure 4.16 shows the DOS for $U/J = 0$ and at several temperatures while Figs. 4.17 and 4.18 show the density of states for 3 values of $U/J$ for $T = 0.003$ and $T = 0.020$ respectively. Although it is more difficult to make assignments of spin to the peaks in these plots, the two peaks labeled exhibit the same behaviour with increasing temperature and $U$. One notable observation is the location of the down-spin peak in Figs. 4.17 and 4.18 for $E \sim -5$. It has an energy which is just below the chemical potential while there is a larger up-spin peak with energy just above the chemical potential. As $U$ is increased, both peaks shift towards the chemical potential, increasing the Fermi occupation of the up-spin states and decreasing the Fermi occupation of the down-spin state. This is consistent with the data for the carrier magnetization in Fig. 4.14 which shows the carrier magnetization decreasing at low temperatures.
Figure 4.13: Local moment magnetization as a function of temperature for a $4 \times 4 \times 4$ lattice with $J = 5t$ and $N_e = 8$ and at several values of $U/J$.

Figure 4.14: Carrier magnetization as a function of temperature for a $4 \times 4 \times 4$ lattice with $J = 5t$ and $N_e = 8$ and at several values of $U/J$. 
Figure 4.15: Local moment magnetization as a function of $U/J$ at several temperatures for a $4 \times 4 \times 4$ lattice with $J/t = 5$ and $N_e = 8$. 
Figure 4.16: Density of states at several temperatures for a $4 \times 4 \times 4$ lattice with $J/t = 5$, $U/J = 0$, and $N_e = 8$. The vertical colored line represents the average value of $\mu$ calculated for the samples used to make the figure. In order to make the scales consistent for all four plots, two of the peaks for $T = 0.003$ have been cut off.
Figure 4.17: Density of states at $T = 0.003$ for several values of $U/J$ for a $4 \times 4 \times 4$ lattice with $J/t = 5$ and $N_e = 8$. The vertical colored line represents the average value of $\mu$ calculated for the samples used to make the figure.
Figure 4.18: Density of states at $T = 0.020$ for several values of $U/J$ for a $4 \times 4 \times 4$ lattice with $J/t = 5$ and $N_e = 8$. The vertical colored line represents the average value of $\mu$ calculated for the samples used to make the figure.
4.2.3 $J = 10t$

In Figs. 4.19 and 4.20 we plot the local moment magnetization and carrier magnetization for a $4 \times 4 \times 4$ lattice with $N_e = 8$ with $J/t = 10$ and $U/J$ ranging from 0 to 2.75. The interactions appear to enhance the magnetization the most at intermediate temperatures (where $\langle M \rangle$ 0.5). When we compare the carrier magnetization [Fig. 4.20] to the corresponding plot for $J/t = 1$ [Fig. 4.6], an important difference is that the low temperature value of $m$ has decreased to be close to $-0.5$ as compared to $-0.37$. This effect is due to $J$ being strong enough to spin polarize the electrons, and $U$ has little effect, unlike the situation at $J/t = 5$, where increasing $U$ decreased the value of the saturation magnetization. The enhancement of the local moment magnetization and carrier magnetization with increasing $U/J$ is weaker than for $J/t = 5$. This behaviour is in accord with the discussion in Sec. 2.3.1 that suggested moderate $J/t$ would be the most interesting region in parameter space to study the effects of interactions with our scheme.

![Figure 4.19](image)

Figure 4.19: Local moment magnetization as a function of temperature for a $4 \times 4 \times 4$ lattice with $J/t = 10$ and $N_e = 8$ for several values of $U/J$. 
Figure 4.20: Carrier magnetization as a function of temperature for a $4 \times 4 \times 4$ lattice with $J/t = 10$ and $N_e = 8$ for several values of $U/J$. 
4.2.4 Determining $T_C$

We now discuss measures that can be used to determine the Curie temperature $T_C$, namely the magnetic susceptibility and the Binder cumulant, and attempt to determine $T_C$ as a function of $U/J$ for $J/t = 5$.

The susceptibility for several values of $U/J$ is plotted in Fig. 4.22 for $J/t = 1$ and Fig. 4.21 for $J/t = 5$. For both values of $J/t$ there is a fairly well defined peak in the susceptibility for $U/J = 0$, but the peak in the plots for finite interactions becomes less defined as $U/J$ is increased. The lowering of the susceptibility in the vicinity of the peak for increasing $U/J$ suggests that interactions dampen the fluctuations in the magnetization about the peak, while the enhancement of susceptibility with $U/J$ at high temperatures suggest that interactions increase fluctuations at high temperatures. From the figures, it appears that the peak in the susceptibility is not a good measure for $T_c$, this is a known issue associated with small system sizes, and has been discussed by Kennett et al. [35].

Of the three values of $J/t$ that we presented results for in this Chapter, the effects of interactions were most pronounced for $J/t = 5$. Due to limited computational resources we focus on this value of $J/t$ for our Binder cumulant calculations. We first show the local moment magnetization for finite interactions and $J/t = 5$ on a $6 \times 6 \times 6$ lattice for $U/J$ ranging from 0 to 2 in Fig. 4.23. We then estimate $T_C$ by plotting the Binder Cumulant as a function of $T/t$ for the two lattice sizes and determine the crossing point. The Binder cumulant for both system sizes are plotted in Figs. 4.24 to 4.26 for $U/J = 0.0$, 0.5, and 1.0.
Figure 4.21: Local moment susceptibility as a function of temperature for a $4 \times 4 \times 4$ lattice with several values of $U/J$, $J/t = 5$ and $N_e = 8$.

Figure 4.22: Local moment susceptibility as a function of temperature for a $4 \times 4 \times 4$ lattice with several values of $U/J$, $J = t = 1$ and $N_e = 8$. 
Figure 4.23: Plot of the magnetization as a function of temperature for a $6 \times 6 \times 6$ lattice with $J/t = 5$, $U/J = 0.0, 0.5, 1.0, \text{and} 2.0$, and $N_e/N_{lm} = 1/8$, with a plot of the magnetization for a $4 \times 4 \times 4$ lattice with $U/J = 0$ included for reference.
Figure 4.24: Binder cumulant as a function of temperature for $4 \times 4 \times 4$ and $6 \times 6 \times 6$ lattices with $J/t = 5$, $U/J = 0$ and $N_e/N_{tm} = 1/8$. The inset shows the vicinity of the crossing temperature.
Figure 4.25: Binder cumulant as a function of temperature for $4 \times 4 \times 4$ and $6 \times 6 \times 6$ lattices with $J/t = 5$, $U/J = 0.5$ and $N_e/N_{lm} = 1/8$. The inset shows the vicinity of the crossing temperature.
Results

Figure 4.26: Binder cumulant as a function of temperature for $4 \times 4 \times 4$ and $6 \times 6 \times 6$ lattices with $J/t = 5$, $U/J = 1.0$ and $N_c/N_{lm} = 1/8$. The inset shows the vicinity of the crossing temperature.
Table 4.1 shows the values of $T_C$ as a function of $U/J$, calculated from the plots of the Binder cumulant. The data is also plotted in Fig. 4.27. Binder cumulant plots for values of $U/J$ not presented above but included in our results are plotted in Appendix B. The values of $T_C$ and their errors were determined by the bootstrap method [57] performed on a linear fit between the two points on either side of the crossing. Although this method provides a reliable estimate for both $T_C$ and its variance for the given data, there are systematic errors associated with the equilibration procedure and linear extrapolation which may still affect the errors. Nevertheless, there is certainly a clear trend in the crossing points that suggests $T_C$ is a monotonically increasing function of $U/J$ in the parameter range studied. This is in accord with the magnetization and DOS data presented in this Chapter.

Table 4.1: $T_C$ as determined from the Binder cumulant as a function of $U/J$ for $J/t = 5$

<table>
<thead>
<tr>
<th>$U/J$</th>
<th>$T_C/t$</th>
<th>error</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00</td>
<td>0.0220</td>
<td>±0.0003</td>
</tr>
<tr>
<td>0.25</td>
<td>0.0240</td>
<td>±0.0010</td>
</tr>
<tr>
<td>0.50</td>
<td>0.0248</td>
<td>±0.0003</td>
</tr>
<tr>
<td>0.75</td>
<td>0.0251</td>
<td>±0.0003</td>
</tr>
<tr>
<td>1.00</td>
<td>0.0256</td>
<td>±0.0004</td>
</tr>
<tr>
<td>1.25</td>
<td>0.0269</td>
<td>±0.0006</td>
</tr>
<tr>
<td>1.50</td>
<td>0.0293</td>
<td>±0.0014</td>
</tr>
<tr>
<td>1.75</td>
<td>0.0305</td>
<td>±0.0006</td>
</tr>
<tr>
<td>2.00</td>
<td>0.0319</td>
<td>±0.0068</td>
</tr>
</tbody>
</table>

4.3 Discussion

The results presented in this Chapter provide evidence that on-site Coulomb repulsion tends to enhance ferromagnetism in the double exchange model at moderate fillings ($N_e/N_{lm} \lesssim 1/2$). The enhancement is further illustrated by the $T_C$ values calculated for $J/t = 5$. Unfortunately, due to the very long simulation times required for $6 \times 6 \times 6$ samples, we were unable to simulate enough samples to account for errors associated with the equilibration procedure. Plots of the density of states and carrier magnetization imply that the enhancement is achieved by increasing the splitting in energy between the states in the vicinity of the chemical potential whose spin is parallel to the direction of local moment magnetization and the states which have spins antiparallel to the local moment magnetization as $U/J$ increases. The extent of the further splitting by the interactions depends
in part on the splitting caused by the exchange interaction. At $J/t = 10$ the exchange splitting is so large compared to the kinetic energy that the electrons are almost completely spin polarized at low temperatures and so we expect the on-site interaction to have an effect only at temperatures of order $T_C$. At small $J/t$ there is only a weak splitting caused by the interactions as there are a comparable number of parallel and anti-parallel spin states with energies below the chemical potential. We thus conclude that on-site Coulomb interactions enhance magnetism for moderate fillings and have the greatest effect at moderate values of $J/t$.

Beyond the restrictions we place on the regions of parameter space that we investigate, there are several limitations to our results, mostly stemming from accessible computational resources. The largest system sizes we were able to study were $6 \times 6 \times 6$, and although this is comparable to the system sizes treated in other studies which ignore Coulomb interactions [35, 58], our results still suffer from various finite size effects, particularly for the $4 \times 4 \times 4$ data.

A first limitation is that it is only practical to simulate small systems at very specific filling factors. The density of states plots point to the cause of this. In both $J/t = 1$ and...
$J/t = 5$ plots for $N_e = 8$, $\mu$ lies within a sizeable gap in the DOS between two peaks of states with opposite spin which are degenerate in their kinetic energy. As it is in a gap, a reasonable deviation in $\mu$ does not significantly affect $\langle N_e \rangle$. If one attempts to simulate for a filling where $N_e$ lies in a peak of a spin and energy degenerate state, very very long equilibration times are needed at low temperatures. For the $4 \times 4 \times 4$ system, $N_e = 8$ and 29 were the only fillings at which we could equilibrate in a reasonable amount of time and observe ferromagnetism. For larger system sizes the DOS starts to resemble a continuum, and although equilibration times increase, we would expect to be able to equilibrate for a larger set of fillings. Adding disorder to the problem would also help to further split the peaks and enable equilibration at other fillings. This was certainly the experience in Refs. [34, 35].

Another limitation in the results is the quantitative accuracy of $T_C$ computed in Sec. 4.2.4. Even with cleaner data for the Binder cumulant crossings, finite size effects are still likely to be non-negligible. In the study by Alonso et al. [39] for the DE model excluding interactions, they found the magnetization to be overestimated at mid-range temperatures by smaller system sizes, and the overestimation decreased with increasing system sizes. In particular, their results showed that only system sizes of $8 \times 8 \times 8$ and larger do not suffer significantly from overestimation at mid-range temperatures. Their study took the limit that $J = \infty$, but it is reasonable to assume that this same effect occurs for finite $J$. Taking this into account we can conclude that although one can make a robust argument as to the qualitative effect on $T_C$ due to interactions, there may still be finite size effects in our determination of $T_C$. 
Chapter 5

Conclusion

Manganites, dilute magnetic semiconductors, and rare-earth hexaborides have been studied intensively in recent years. Some of the magnetic properties of these seemingly disparate materials can be described by the same class of theoretical model; the double exchange model. Electron-electron interactions should be included in a full theoretical treatment for these systems although this has often not been the case. Until now, interactions have always been ignored in Monte Carlo simulations of these models. In this thesis we have shown that there is a region in parameter space where one can reasonably combine mean field theory for electron-electron interactions with classical spin Monte Carlo simulations for the interacting double exchange model.

We have developed and implemented a scheme to simulate a double exchange model which includes a on-site Hubbard $U$ term. With available computing resources, we were able to simulate lattice sizes of up to 216 sites, which is competitive with previous studies which ignored electron-electron interactions. Results obtained from our model suggest that the on-site interactions enhance magnetism by strengthening the tendency towards carrier spin polarization. Computational resources limited the system sizes that we were able to investigate and hence the quantitative results we were able to achieve. Increased processing power and possibly further optimization of the code appear to be the most realistic avenues for achieving higher quality results.

The main development in this work is the Monte Carlo scheme itself. This thesis exhibits the first successful attempt to include electron-electron interactions in a Monte Carlo simulation of the double exchange model. In addition to the on-site Coulomb interactions
studied here, additional physics of long range Coulomb interactions and disorder are believed to lead to physics such as phase separation and CMR in the manganites, and so the natural extension of the work here is to include these effects in the double exchange model. Generalization of the current computer code to include long range Coulomb interactions and disorder should require minimal additional effort, and will hopefully lead to improved understanding of the microscopic physics of materials where double exchange physics is important.
Bibliography


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Appendix A

Exact solution for the toy model

The derivation in this appendix is due to Mona Berciu and was presented in Ref. [35]. It is included for completeness, since the solution to this model was used in the testing of the Monte Carlo algorithm used for our work. Consider the Hamiltonian

\[ H = -t \sum_{\langle i,j \rangle} \left[ c^\dagger_i c_j + \text{h.c.} \right] + \frac{J}{N} \mathbf{S} \cdot \sum_{j=1}^{N} c^\dagger_{j} \frac{1}{2} \sigma_{\alpha \beta} c_{j\beta}, \]  

(A.1)

where

\[ \mathbf{S} = \sum_{i=1}^{N} \mathbf{S}_i, \]

is the total spin of the system.

Parametrize \( \mathbf{S} = S(\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta) \) and perform the canonical transformation

\[
\begin{align*}
d_{i\uparrow} &= \cos \frac{\theta}{2} c_i + \sin \frac{\theta}{2} e^{-i\phi} c_{i\downarrow}, \\
d_{i\downarrow} &= -\sin \frac{\theta}{2} e^{i\phi} c_{i\uparrow} + \cos \frac{\theta}{2} c_{i\downarrow}.
\end{align*}
\]

It is then straightforward to show that

\[
H = -t \sum_{\langle i,j \rangle} d^\dagger_{i\sigma} d_{j\sigma} + \frac{J S}{2N} \sum_{i=1}^{N} \left( d_{i\uparrow}^\dagger d_{i\uparrow} - d_{i\downarrow}^\dagger d_{i\downarrow} \right).
\]

As a result, the Hamiltonian can be diagonalized in \( \vec{k} \)-space

\[ H = \sum_k \epsilon_{k\sigma}(S) d^\dagger_{k\sigma} d_{k\sigma}, \]  

(A.2)
where
\[ E_{k\sigma}(S) = -2t \cos ka + \frac{1}{2N} J_0 S \sigma, \]  
(A.3)
with \( \sigma = \pm 1 \) and the cyclic boundary conditions imply that \( ka = 2\pi n/N, \) for \( n = 0, 1, \ldots, N - 1. \) There is a single lowest eigenvalue corresponding to \( k = 0, \) and then degenerate eigenvalues corresponding to left and right moving modes.

Thus the grand-canonical partition function is
\[ Z_N = \prod_{i=1}^{N} \left[ \int d\Omega_i \right] \prod_{k\sigma} \left( 1 + e^{-\beta (E_{k\sigma}(S) - \mu)} \right), \]  
(A.4)

where the simplified notation \( \int d\Omega_i = \int_0^\pi \sin \theta_i d\theta_i \int_0^{2\pi} d\phi_i \) is used for the integral over solid angle. One can avoid the \( 2N \) multiple integrals over individual spin angles, and replace them by an expression of the general form
\[ Z_N = \int_0^{2\pi} d\phi \int_0^\pi d\theta \int_0^N dS S^2 F_N(S, \theta, \phi) \prod_{k\sigma} (1 + e^{-\beta (E_{k\sigma}(S) - \mu)}), \]  
(A.5)

where \( \theta, \phi \) define the orientation of the total spin \( S, \) and it is noted that the magnitude of the total spin, \( S, \) varies only between 0 and \( N. \) Comparing Eqs. (A.4) and (A.5), the definition of the “weight” \( F_N(S) \) is
\[ F_N(S) = \prod_{i=1}^{N} \left[ \int d\Omega_i \right] \delta \left( S - \sum_{i=1}^{N} S_i \right), \]  
(A.6)

from which it is straightforward to derive the recurrence relation
\[ F_N(S) = \int d\Omega_N F_{N-1}(S - S_N) \theta(N - 1 - |S - S_N|), \]

where the Heaviside function \( \theta \) ensures that the argument of \( F_{N-1} \) cannot have a magnitude larger than \( N - 1. \) It is apparent that, in fact, \( F_N(S) = F_N(S). \) This can easily be seen from the definition Eq. (A.6) as well, since one can choose to define the angles \( \Omega_i \) with respect to the system of coordinates in which \( S = S \Omega = S \mathbf{z}, \) and the result cannot depend on the particular orientation \( \theta, \phi \) of the total spin \( S. \) Using the new variable \( y = \cos \theta_N, \)
and performing the trivial integral over \( \phi_N, \) the recurrence relation can be rewritten as:
\[ F_N(S) = 2\pi \int_{-1}^{1} dy F_{N-1}(\sqrt{1 + S^2 - 2Sy}) \theta(N - 1 - \sqrt{1 + S^2 - 2Sy}). \]  
(A.7)
Defining
\[ F_N(S) = (4\pi)^{N-1} f_N(S), \]  
(A.8)
one obtains the recurrence formula
\[ f_N(x) = \frac{1}{2x} \int_{|x-1|}^{\min(x+1,N-1)} dz f_{N-1}(z). \]  
(A.9)
This is supplemented by the “initial” condition
\[ f_2(x) = \frac{1}{2x}, \]  
(A.10)
which can be obtained through direct integration from Eq. (A.6).

From the recurrence relation Eq. (A.9), one can determine the general solution for the weight function \( f_N(x) \) for any integer \( N \). This is a piece-wise continuous function, given by
\[ f_N(x) = \frac{1}{2^{N-1}(N-2)!} x \sum_{k=0}^{m} (-1)^k C_N^k (N - 2k - x)^{N-2} \]
on the subinterval \( N - 2(m + 1) \leq x \leq N - 2m \) of the support interval \([0, N]\), with \( m \) an integer \( 0 \leq m < N/2 \) and where \( C_N^k = \frac{N!}{k!(N-k)!} \) is the appropriate binomial coefficient.

The partition function is thus
\[ Z_N = (4\pi)^N \int_0^N dS S^2 \prod_{k\sigma} \left( 1 + e^{-\beta(E_{k\sigma}(S)-\mu)} \right), \]
where the chemical potential \( \mu \) is fixed from the condition for the average number of fermions:
\[ \langle n_c \rangle = \frac{\int_0^N dS S^2 f_N(S) \sum_{\sigma,\alpha} n_{q\alpha} \prod_{k\sigma} \left( 1 + e^{-\beta(E_{k\sigma}(S)-\mu)} \right)}{Z_N}, \]
where \( n_{q\alpha} = [e^{\beta(E_{\alpha q}(S)-\mu)} + 1]^{-1} \) are the occupation numbers of the fermionic levels. Since an analytical expression is available for the weight function \( f_N(S) \), these integrals can be evaluated numerically. Once the chemical potential \( \mu \) is known, any other expectation value, such as the total spin magnitude
\[ \langle S \rangle = \frac{1}{Z_N} \int_0^N dS S^3 f_N(S) \prod_{k\sigma} \left( 1 + e^{-\beta(E_{k\sigma}-\mu)} \right) \]
or the total fermionic spin
\[ \langle |s| \rangle = \int_0^N dS S^2 f_N(S) \sum_{q\alpha} n_{q\alpha} \frac{\prod_{k\sigma} \left( 1 + e^{-\beta(E_{k\sigma}-\mu)} \right)}{Z_N} \]
can be computed for any given temperature \( T \).
Appendix B

Binder Cumulant plots

Here we present plots of the Binder cumulant, for $4 \times 4 \times 4$ and $6 \times 6 \times 6$ lattices with $J/t = 5$, $N_e/N_{lm} = 1/8$ and several values of $U/J$, used to determine the values of $T_C$ presented in Chapter 4. Each point on any particular figure presented in this Chapter represents an average over 5 to 10 samples with 2000 data points taken for each sample for the $4 \times 4 \times 4$ data, and an average over 500-2000 data points for one sample was used for the $6 \times 6 \times 6$ data. In all plots errorbars for $4 \times 4 \times 4$ data are on the order of thickness of the line.
Figure B.1: Binder cumulant as a function of temperature plots for $U/J = 0.25$.

Figure B.2: Binder cumulant as a function of temperature plots for $U/J = 0.75$. 
Figure B.3: Binder cumulant as a function of temperature plots for $U/J = 1.25$.

Figure B.4: Binder cumulant as a function of temperature plots for $U/J = 1.5$. 
APPENDIX B. BINDER CUMULANT PLOTS

Figure B.5: Binder cumulant as a function of temperature plots for $U/J = 1.75$.

Figure B.6: Binder cumulant as a function of temperature plots for $U/J = 2.0$. 