Spiral Magnetic Ordering
in the Double Exchange model
on the Triangular Lattice

Master Thesis

Author: Maria Azhar
MSc Nanoscience (2012-2014)

Supervisor: Prof. Dr Maxim Mostovoy

Referee: Prof. Dr. Thom Palstra

Period: September 2013-August 2014
## Contents

1. Introduction ................................................................. 1

2. The Double Exchange Model .............................................. 4  
   2.1 The Double Exchange Model .......................................... 4  
   2.2 Ferromagnetism in the Double Exchange Model due to a large Hunds Rule coupling ........................................ 7  
   2.3 Noncollinear, noncoplanar and chiral ordering in the Double Exchange Model .................................................. 8  
   2.4 Coulomb interactions between electrons .......................... 9  
   2.5 Spiral ordering in the presence of competing interactions ... 10  
   2.6 Phase separation in the Double Exchange Model .............. 12

3. Instability of the ferromagnetic state towards spiral ordering .... 14  
   3.1 Spiral ordering .......................................................... 15  
   3.2 Instability of the ferromagnetic state towards spiral ordering 15  
      3.2.1 … in a continuum double exchange model ................... 17  
      3.2.2 … on the triangular lattice ..................................... 19

4. The Double Exchange Model on the triangular lattice ................ 26  
   4.1 Itinerant electrons on the triangular lattice .................... 26  
   4.2 The Ferromagnetic state ............................................... 30  
   4.3 The “tetrahedral” state ............................................... 31  
      4.3.1 The stability of the tetrahedral state at ¼ and ¾ filling 33  
      4.3.2 Dispersion relation for the tetrahedral state ............. 34  
      4.3.3 Effective magnetic flux due to the tetrahedral ordering 36  
   4.4 Dispersion relation for the helical state ......................... 40  
      4.4.1 Continuity of the wavefunction ............................. 41  
   4.5 Other spin configurations ............................................ 44  
   4.6 The Phase Diagram .................................................... 45  
      4.6.1 Finite Size Effects .............................................. 49  
      4.6.2 Phase Separation ................................................ 50

5. Discussion and Conclusions ............................................... 52

Acknowledgements. ............................................................ 54

Appendix. ............................................................................. 55
References. .......................................................................... 57
Section 1

Introduction

Our understanding of magnetism has evolved rapidly starting from the unification of electricity, magnetism and optics in Maxwell’s equations. Magnetism can be described as a purely relativistic effect of the relative motion of an observer and electrical charges, and indeed this motivated Einstein’s theory of special relativity. At the same time magnetism in condensed matter systems is also a quantum mechanical phenomenon because according to the Bohr-van Leeuwen theorem, a classical system in thermal equilibrium has zero magnetization, even in the presence of an applied magnetic field. Magnetism has also been impossible to disentangle from our technological advancement, from compass needles to electric motors and communication systems.

Recently, helicoidal spiral, noncoplanar, and chiral magnetic structures have gained interest for spintronics applications [1-3]. Heterostructures consisting of ferromagnetic and spiral layers can be used for the manipulation of electronic spins. A relatively small magnitude of spin-polarized current flowing through such a device can allow the switching of the magnetization of the ferromagnetic layers through the spin transfer torque [4-5]. This holds potential for application in devices that use electric current for more efficient switching of magnetization with a smaller bit size, instead of the bulkier conventional magnetic storage that uses magnetic fields to read or write data.

In helical magnets, both the time and the space inversion symmetry is broken - in centrosymmetric crystals the space symmetry is broken spontaneously and in non-centrosymmetric crystals the symmetry breaking is enforced by the crystal lattice. In many multiferroic materials, a spiral ordering is induced by competing interactions, and the resulting inversion symmetry breaking switches on the Dzyaloshinskii-Moriya interaction, inducing an electric dipole moment [6-9] through a mechanism completely different from those working in conventional ferroelectrics. Multiferroics hold potential for technological applications with their ability for a 4-state logic (with the two polarization states and two
magnetization states), as magnetoelectric sensors, and for electric field control of magnetic memory. Noncoplanar magnetic orders with non-zero scalar chirality give rise to unusual transport and magnetoelectric properties and are attracting interest in the fields of multiferroics and topological insulators. [10-12] There is experimental evidence of the Topological Hall Effect arising from effective magnetic fields induced by chiral spin orders. [13-16]

In many materials an incommensurate spiral ordering can result from competing exchange interactions, e.g. nearest-neighbour ferromagnetic and next-nearest neighbor antiferromagnetic interactions or from oscillating RKKY interactions. Also Dzyaloshinskii-Moriya (DM) interaction can lead to spiral spin ordering or more complex noncoplanar orders, such as the skyrmion crystal that be viewed as a superposition of three spirals.

In this thesis we show that spiral ordering on a triangular lattice can originate also from the double exchange mechanism [17], which is commonly believed to stabilize the collinear ferromagnetic state. The double exchange model intrinsically embodies an interplay between charge and spin. It was introduced by Zener [17] to describe physics of the colossal magnetoresistance manganites, a class of materials in which electric transport is strongly coupled to the magnetic properties. Noncollinear spiral spin order arising as a result of the competition between the double exchange favoring ferromagnetism and superexchange favoring antiferromagnetic state was first studied by de Gennes [18].

More recently, a non-coplanar 4-sublattice “tetrahedral” chiral phase was found by Martin and Batiste to be stabilized by nesting at 3/4 filling in the double exchange model on the triangular lattice [19]. This structure had already been proposed to describe some triangular lattice systems [20,21] and could potentially be accessible in other physical systems such as liquid surfaces or nanostructured semiconductor heterostructures. The ground state phase diagram for the double exchange model was obtained numerically by Akagi and Motome [12] and this chiral phase was found to exist at the ¼ and ¾ fillings. The competition between the ferromagnetic exchange and the antiferromagnetic superexchange on the triangular lattice and its temperature dependence at the ½ filling was studied by Kumar and van den Brink [23] and at ¼ filling in Ref. [24]. None of these authors considered the possibility of incommensurate spiral magnetic orders in the Double Exchange Model on the triangular lattice. Here, we obtain analytically and numerically the range of parameters for which the spiral state is the ground state for the triangular lattice. The spin spiral state has been found analytically to be stable towards phase separation for an isotropic model for small fillings \( \leq 0.3 \) [25] and also for a square and cubic lattice at small values of the Hund’s Rule coupling [26].

In Chapter 2 the double exchange model and some of its limiting cases are presented. The spiral state in the presence of the competing ferromagnetic double exchange and antiferromagnetic exchange interaction and the instability of this state against phase
separation is studied for a one-dimensional chain. In Chapter 3 we give a general derivation of the conditions for the instability of the ferromagnetic state towards spiral ordering, which we then apply for the case of the triangular lattice. In Chapter 4 we discuss other possible magnetic orders on the triangular lattice, such as the ferromagnetic spin state and 4-sublattice tetrahedral chiral ordering. The dispersion relation for the spiral state on the triangular lattice is derived. Finally, we use numerical calculations of energies of the competing states to construct a phase diagram of the double exchange model on the triangular lattice and find that the spiral phase is the stable over a wide range of parameters.
Section 2

The Double Exchange Model

The Heisenberg exchange model,

\[ H = -J \sum_{ij} \mathbf{S}_i \cdot \mathbf{S}_j, \]  

provides an accurate description of spins in Mott insulators. It is, however, insufficient for magnetic conductors with itinerant (delocalized) electrons. A famous example is provided by colossal magnetoresistance manganites [27]. Parent compounds of these materials are typically antiferromagnetic Mott insulators, which upon a small amount doping become ferromagnetic conductors. The electrical transport properties of colossal magnetoresistance manganites are closely related to magnetism and *vice versa* and the need was felt to introduce models that clarified the role of conduction electrons in these materials.

### 2.1 The Double Exchange Model

The Double Exchange Model describes a system of localized spins interacting with the spins of itinerant (conduction) electrons [17,18,28,30]. The Hamiltonian of this model has the form

\[ H_{DE} = - \sum_{ij \alpha} t_{ij} \psi_{i \alpha}^{\dagger} \psi_{j \alpha} - J \sum_{i \alpha \beta} \psi_{i \alpha}^{\dagger} \sigma_{\alpha \beta} \psi_{i \beta} \cdot \mathbf{S}_i + J_S \sum_{ij} \mathbf{S}_i \cdot \mathbf{S}_j, \]  

where the first term is the kinetic energy of the conduction electrons according to the tight-binding model. The electrons are described by the annihilation (creation) operator \( \psi_{i \alpha} (\psi_{i \alpha}^{\dagger} \). \( \psi_{i \alpha} \) is the annihilation operator with spin index \( \alpha = \uparrow, \downarrow \) for the itinerant electron located at...
the site i. \( t_{ij} \) is the hopping amplitude for an electron to hop from site j to site i. In the second term, J is the magnitude of the Hund’s rule ferromagnetic coupling between the conduction electrons and the local spins, which tends to keep the electron spins parallel to the localised spins \( S_i \). \( \sigma = (\sigma^x, \sigma^y, \sigma^z) \) is a vector composed of the Pauli matrices. \( \frac{1}{2}\psi_{i\alpha}^+ \sigma_{\alpha\beta} \psi_{i\beta} \) is the spin operator for the conduction electrons on the site i and we can absorb the factor of \( \frac{1}{2} \) into J. The last term describes the Heisenberg exchange of magnitude \( J_S \) between the localised spins on lattice sites. An additional index can be introduced on the electron wavefunctions to label the orbital, but for simplicity in our calculations we shall consider the one orbital model.

This model was introduced by Zener [17] to explain the strong correlations between the movement of charge and the spin polarization of the magnetic lattice ions, in the perovskite manganites, e.g. \( R_{1-x}A_x\text{MnO}_3 \) or \( R_{2-2x}A_{1+2x}\text{Mn}_2\text{O}_7 \) where R and A are the rare earth and alkali earth metals, respectively. It has since been widely used to describe systems with colossal magnetoresistance such as \( \text{La}_{1-x}A_x\text{MnO}_3 \) (A=Ca, Sr). Unlike the parent compound \( \text{LaMnO}_3 \), the partially doped lanthanum manganites are spontaneously magnetized and show a dramatic change of other properties as the doping is changed [31]. Clearly the itinerant electrons had some role to play that had to be incorporated in the Hamiltonian, and indeed in the literature, the Hund’s Rule coupling is also referred to as the spin-charge coupling.

In a cubic environment with 6 nearest neighbouring O ions, the 3d level of the transition metal ion splits into two degenerate levels – the \( t_{2g} \) and the \( e_g \) levels respectively. In some transition metal compounds the \( t_{2g} \) electrons can be considered to be spins localized at atomic sites whereas the charge is carried by the \( e_g \) electrons, such as in metallic manganese and iron perovskites, \( \text{La}_{1-x}\text{Ca}_x\text{MnO}_3 \) and \( \text{SrFeO}_3 \). In other materials it may be possible to

---

**Figure 2.1** The Double Exchange mechanism, involving two electrons and an intermediate Oxygen atom
make a less clear-cut distinction between localized and itinerant $t_{2g}$ electrons, such as in CrO$_2$ and LiV$_2$O$_4$ [32,33]. The localized spins are the spins of the electrons occupying partially filled bands, and not the nuclear spin. The spin of the nucleus is too weak in most cases to be of sufficient consequence. Also, in a crystal field, quenching of the orbital angular momentum almost always occurs, so we do not consider the orbital contribution to the atomic moments.

The ferromagnetic Hund’s rule coupling term is valid when both the localized and the itinerant electrons belong to different subbands of the same partially filled d shell. This is the situation when considering ferromagnetism in metallic Fe or Ni, and in the manganites. However, when the localized and conduction electrons belong to different ions or when they have completely different character, the coupling is antiferromagnetic (of the Schrieffer-Wolf type) [34] e.g. f electrons of Ce and conduction electrons in materials like CeAl$_3$. In the Kondo Effect, this coupling is antiferromagnetic, whereby the spins of the conduction electrons shield the impurity’s magnetic moment in a nonmagnetic crystal lattice. For electrons belonging to the same band on the same ion, the Pauli exclusion principle tends to keep electrons having parallel spins apart in space, and hence the electron repulsion is minimized for a ferromagnetic Hund’s Rule coupling.

For most cases we can consider a simple model to describe double exchange systems [35] where the last term in Eq (1) is set to zero. In this case any coupling between the localized spins occurs only indirectly, through the conduction electrons.

The majority of theoretical work in the context of manganites ignores the presence of the oxygen, although historically the intermediate state involving oxygen and two electrons gave rise to the term “double” exchange (see Figure1.1). The oxygen degrees of freedom are not themselves crucial for double-exchange induced ferromagnetism, as has been shown in models that consider only Mn ions, as long as the Hund’s rule coupling is strong enough [36,37]. The electrons only need a background of magnetic spins that can be polarized to minimize the kinetic energy of the electrons.

The O atoms are more likely to have a role in the ferrates e.g. SrFeO$_3$ and CaFeO$_3$ which have an especially low charge transfer energy (the energy cost of transferring an electron from the oxygen p orbital to the d orbital of the metal). In an alternative d-p model to describe ferromagnetic coupling between the metal ions [38], the holes on O atoms couple antiferromagnetically with the localised spins on the Fe or Mn metal atoms, and the metal atoms in turn couple ferromagnetically to each other.
Ferromagnetism due to a large Hund’s Rule coupling

In actual materials, \( J/t \) is larger than 1. Typical values for the \( e_g \) conduction electrons in manganites are \( t \sim 0.2-0.5 \) eV and \( J \sim 1-2 \) eV [39]. The approximation \( J \gg t \) can be made to bring the Hamiltonian to a simpler form. For the case of \( J \gg t \) the directions of electron spins become parallel to the local spins. Consider the classical spin at the site \( i \),

\[
S_i = S(\sin \theta_i \cos \varphi_i, \sin \theta_i \sin \varphi_i, \cos \theta_i),
\]

(2.3)

where \( \theta_i \) and \( \varphi_i \) are the spherical angles describing the direction of the spin magnetic moment \( S_i \) at lattice site \( i \). In the strong \( J \) limit we can project out the spin parts of the electronic wavefunctions

\[
\psi_{i\alpha} = u_{i\alpha} c_i
\]

(2.4)

where \( u_i = \begin{pmatrix} \cos \frac{\theta_i}{2} \\ \sin \frac{\theta_i}{2} e^{i\varphi_i} \end{pmatrix} \) is the spin part of the electron wavefunction \( \psi_{i\alpha} \) and \( c_i \) is an operator describing electrons. This spin-polarised projection leads to an effective hopping amplitude depending on the orientations of the local spins as follows:

\[
\tilde{t}_{ij} = t_{ij} u_i^\dagger u_j = t_{ij} \cos \frac{\theta_{ij}}{2} e^{i\varphi_{ij}}
\]

(2.5)

where \( \theta_{ij} \) is the angle between spins \( S_i \) and \( S_j \):

\[
\cos \theta_{ij} = \cos \theta_i \cos \theta_j + \sin \theta_i \sin \theta_j \cos (\varphi_i - \varphi_j).
\]

(2.6)
In general, for noncoplanar spins the hopping amplitude also has a complex phase \( \phi_{ij} \) depending on the orientations of the two spins; for coplanar spin ordering this phase is zero (\( \phi_{ij} \) is the solid angle subtended by the unit vectors along the two spins \( S_i \) and \( S_j \), and the \( \hat{z} \) axis, on the unit sphere). Compare this phase to the Berry phase picked up by charge carriers in the presence of a magnetic field. A topologically nontrivial spin texture can affect the motion of charge carriers in the same manner as an applied magnetic field, and this is a mechanism proposed to understand the topological Hall Effect observed in the collosal magnetoresistance manganites.[40-42]

We obtain the following Hamiltonian to describe the electrons in the spin-up and spin-down bands (with spins parallel- and antiparallel- respectively to the localized spins) [28]:

\[
H_{DE}^{(\pm)} = - \sum_{ij} \tilde{t}_{ij} c_i^{\dagger} c_j \pm JS
\]  

(2.7)

The number of degrees of freedom have been reduced because the spin index is no longer needed, so the model becomes considerably simplified. Effectively, only those states of the system are being considered, that give a minimum of energy according to the Hund’s rules.

The kinetic energy of the itinerant electrons is minimised for ferromagnetic (FM) ordering, where the conduction bands are the widest (\( \theta_{ij}=0 \)). This is because, for the case of FM ordering, the itinerant electrons can hop from site to site, without changing the spin direction, hence the hopping amplitudes are maximal for FM spin ordering. This is the double exchange mechanism for ferromagnetism [17]. One can imagine that a conduction electron “pulls” the lattice spins into a ferromagnetic alignment in order to minimize its kinetic energy. For the case of antiparallel spins (\( \theta_{ij}=\pi \)) the hopping amplitude is zero, and the conduction electrons would be localized at lattice sites, giving an insulating state.

It would seem natural for the ferromagnetically ordered state to have the lowest total energy in the infinite-J limit. However it can be shown [43] that even in the infinite-J limit, a helicoidal spiral can have a lower energy than the ferromagnetic state.

### 2.3 Noncoplanar noncollinear and chiral ordering in the Double Exchange Model

The opposite limit of \( t \gg J \) is especially suitable for rare earth ions where the on-site coupling between the \( f \) electrons and the conduction electrons is weak. A perturbative treatment in \( J/t \) leads to an effective spin Hamiltonian with the oscillating long-range RKKY interaction between the localised \( f \)-electron spins [44].
\[ H_{RKKY} = \frac{1}{2} \sum_{i,j} J_{ij} \mathbf{s}_i \cdot \mathbf{s}_j \, . \] (2.8)

which in general favors noncollinear, incommensurate magnetic ordering of the lattice spins.

The competition between the ferromagnetic Double exchange and the antiferromagnetic exchange to give a noncollinear state was first studied by de Gennes \cite{18} as a compromise between the ferromagnetism of the “Double Exchange” (high doping) limit, and the antiferromagnetic superexchange in the limit of no doping in the manganites. However, even the simple Double Exchange model without the presence of antiferromagnetic exchange, can support helical ordering \cite{43}. Rare earth metals such as Ho and Er, which can be described by the model (Eq.(2.2)) show a helicoidal ordering \cite{45}. In spiral multiferroics, the breaking of inversion symmetry induces electric polarization by activating the Dzyaloshinski-Moriya interaction, leading to multiferroic properties of the system \cite{46}.

Depending on the value of JS/t and the filling fraction (number of electrons per lattice site), we may obtain different magnetic orders that can have the lowest energy, depending on the lattice: ferromagnetic, antiferromagnetic, noncollinear e.g. 120° ordered, (incommensurate) helicoidal, or noncoplanar e.g. states with a nonzero scalar chirality, in which case the hopping amplitudes are complex, and this can then lead to the interesting effects associated with noncollinear spin ordering, such as the topological Hall effect and multiferroicity.

### 2.4 Coulomb interactions between electrons

In this section we examine how the Double Exchange (DE) Model can account for Coulomb repulsion between electrons. For large values of JS/t, the splitting between the spin-up and spin-down bands is large and double occupancy of lattice sites is prevented, so for small filling fractions it would be reasonable to ignore on-site Coulomb repulsions. When on-site Coulomb interactions between electrons are included in the tight-binding model, we have the Hubbard model of the form

\[ H = -t \sum_{<i,j>,\sigma} \psi_{i\alpha}^\dagger \psi_{j\alpha} + U \sum_i n_{\uparrow i} n_{\downarrow i} \] (2.9)

where U is the energy penalty for the double occupancy of any lattice site. Unfortunately the addition of on-site Coulomb interactions makes the model too complex to be solved exactly using the techniques of many-body physics except in 1D. This model is solvable using the methods of perturbation theory only in the limits of U>>t and U<<t. In the limit U>>t the Hamiltonian of the Hubbard model (Eq. 2.10) effectively reduces to the form of the antiferromagnetic Heisenberg exchange
which can be taken into account through the antiferromagnetic exchange term in Eq (2.2). This tendency towards antiferromagnetism in the $U>>t$ limit of the Hubbard model can be explained simply. Consider a system of two sites, with one electron at each site. We can have three types of possibilities for the alignment of the electronic spins (see Figure 2.3). The state with antiferromagnetic alignment of spins (b) is lowest in energy. This is due to the contribution from the excited state (c) from second order perturbation theory, which always gives a negative contribution to the ground state energy. As the value of $U$ (the Coulomb interaction between electrons) is increased, a metal eventually undergoes a transition to an antiferromagnetic state and becomes a Mott insulator, similar to the metal-insulator transition observed in many transition metal oxides.

In general, when perturbative analysis is not valid, one has to apply nonperturbative methods of analysis, one of which is the Dynamical Mean Field Method to calculate behavior at a finite temperature. The DMF method has been applied to strongly correlated electron systems for the Hubbard model, [47,48] the t-J model, [49,50] and to the DE model [51].

2.5 Spiral ordering in the presence of competing interactions

Most systems with magnetic interactions are capable of exhibiting a helicoidal order. Indeed, the ferromagnetic and antiferromagnetic cases on any lattice are specific cases of a helicoidal spiral; for a one dimensional chain of spins with lattice constant $a$, such spirals would have wavevectors of magnitudes $Q=2\pi/a$ and $Q=\pi/a$ respectively. In general the helix is incommensurate with respect to the underlying lattice. Consider the simple example of the one-band double exchange model in 1D with the addition of an Antiferromagnetic exchange interaction,
Figure 1.4 Atomic spins on lattice sites in 1D. θ is the angle by which successive spins are rotated.

\[ H = -t \sum_{\langle ij \rangle} \psi_{ia}^\dagger \psi_{ja} - J \sum_i \psi_{ia}^\dagger \sigma_{a\beta} \psi_{i\beta} \cdot S_i + J_S \sum_n S_n \cdot S_{n+1}, \]  

(2.11)

where the summation in the third term is over the bonds between the neighbouring sites. Let θ be the “pitch” of the helix (see Fig 2.4). \( S_i \) is the spin at site number \( n_i \) along the chain, where \( n_i \) is an integer, and

\[ S_i = S(0, \sin \theta n_i, \cos \theta n_i). \]  

(2.12)

In the limit of very large \( J_H \) the electronic spins are parallel to the lattice spins. In this case we can perform the following substitution for nearest-neighbour hopping,

\[ t \rightarrow t \cos \frac{\theta}{2}, \]  

(2.13)

whereas the Hund’s rule coupling term becomes \( \pm J_S \) for the spin-up and spin-down bands, independent of θ. We can write the total energy of the system as a function of θ, N (the total number of lattice sites), and \( E_{\text{kin}}(0) \), the value of the kinetic energy term for the case of \( \theta = 0 \) (the ferromagnetic case),

\[ E = -|E_{\text{kin}}(0)| \cos \frac{\theta}{2} + J_S S^2 N \cos \theta. \]  

(2.14)

We can now find the value of θ that minimizes the total energy:

\[ \frac{\partial E}{\partial \theta} = \frac{|E_{\text{kin}}(0)|}{2} \sin \frac{\theta}{2} - J_S S^2 N \sin \theta = 0. \]  

(2.15)

if \( \theta \neq 0 \), the solution for this equation is

\[ \cos \frac{\theta}{2} = \frac{|E_{\text{kin}}(0)|}{4 J_S S^2 N}. \]  

(2.16)

For \( |E_{\text{kin}}(0)| < 4 J_S S^2 N \) we have a nonzero solution for θ which in general corresponds to an incommensurate helicoidal (spiral) state. For \( |E_{\text{kin}}(0)| \geq 4 J_S S^2 N \) the ferromagnetic state is lower in energy.
We have shown that a spiral state has a minimum energy when the “ferromagnetism” of the double exchange model competes with an antiferromagnetic exchange between spins on lattice sites. However, this state could be susceptible to phase separation, as quantitatively discussed in the next section.

2.6 Phase separation in the Double Exchange Model

Phase separation is a phenomenon inherent to strongly interacting electron systems. It is present in the Hubbard model, the t-J model, and also the Kondo lattice model. It has been experimentally documented in materials such as the manganites and cuprates. [39]

For a thermodynamically stable homogenous state, the chemical potential $\mu$ must fulfill the following requirement for the electronic inverse compressibility, $x$,

$$x = -V \frac{\partial P}{\partial V} = n^2 \frac{\partial \mu}{\partial n} = n^2 \frac{\partial^2 \varepsilon}{\partial n^2} > 0,$$  \hspace{1cm} (2.17)

where $n$ is the electron density – the number of electrons ($N_e$) per number of lattice sites ($N$). $P$ and $V$ are the thermodynamic quantities, pressure and volume respectively. In our case the volume corresponds to the total number of lattice sites. $\varepsilon = E/N$ is the energy per lattice site. If this requirement is not fulfilled, the system can reduce its energy by going to an inhomogeneous state with varying charge density e.g. there could exist small nanoscale pockets or large regions containing different spin configurations; the cases of “frustrated phase separation” and complete phase separation, respectively. [52]

We can calculate the compressibility for the one dimensional chain for the case of a very low electron concentration, $n$. We can approximate $|E_{kin}(0)| = n \frac{W}{2}$ where $W$ is the bandwidth, assuming that all electrons occupy the states close to the bottom of the band, with the energy $\sim -W/2$ as illustrated in Figure 2.5 (for the ferromagnetic case the bands are widest and the bandwidth is $t$).

Substituting the value for $|E_{kin}(0)|$ into Eq. 2.17, we obtain

$$\cos \frac{\theta}{2} = \frac{|E_{kin}(0)|}{4J_sS^2N} = \frac{nW}{8J_sS^2N},$$ \hspace{1cm} (2.18)

and this expression for $\cos \frac{\theta}{2}$ substituted in Eq. (2.15) yields

$$\varepsilon = -\frac{nW}{2N} \cos \frac{\theta}{2} + J_sS^2 \cos \theta = -\frac{n^2W^2}{16NJ_sS^2} - J_sS^2$$ \hspace{1cm} (2.19)
\[ \frac{\partial^2 e}{\partial n^2} < 0 \] so this state is unstable. The only case in which the criterion for thermodynamical stability can be fulfilled for this state is when \( W = 0 \Rightarrow cos \frac{\theta}{2} = 0 \Rightarrow \theta = \pi \), which is the antiferromagnetic state. We would therefore expect that in some part of the chain there will be a region of antiferromagnetic ordering with no mobile charge carriers present, while the remaining part of the chain will be ferromagnetic with high electron concentration. Such a phase separated state would have energy lower than that of a homogenous, periodically modulated spiral state [53,54].

If long-range Coulomb repulsion between electrons is included, electroneutrality would be favoured, and complete phase separation would be suppressed, with the possible formation of phase separated regions or microscopic domains of different charge density, with their size depending on the strength of the Coulomb interaction [52]. The long-range Coulomb repulsion between electrons (if included) would oppose the variation of \( n \) throughout the sample.

The condition (Eq. (2.18)) for compressibility following from thermodynamics always needs to be satisfied. In another scheme to determine the phase separation regions, at a fixed value of \( \mu \) we minimize the grand canonical potential \( \Omega \), where

\[ \Omega = E - \mu N_e . \] (2.20)

\( n(\mu) \) has a different value for each state. A jump in \( n(\mu) \) would signal a phase transition, and in the interval of those values of \( n \), a phase separated state between the phases at the ends of the interval would exist. Thus in this scenario there are certain values of the density \( n \) that can not be stabilised, whatever the value of \( \mu \).

In ferrates, the degrees of freedom for the oxygen ion can be considered, to yield a spiral magnetic ordering stable towards phase separation [43]. The manganites are well-known to phase separate [39] into metallic ferromagnetic and antiferromagnetic insulating domains. However the d-p model which takes into account the oxygen atoms, does allow a homogenous helical ordering stable towards phase separation in a wide range of parameters [38,43].

**Figure 2.5** Dispersion relation for the 1D chain with a bandwidth of \( W \).
Section 3

Instability of the ferromagnetic state towards spiral ordering

3.1 Spiral ordering

Spiral magnetic order gives rise to interesting effects in materials. A spiral helix does not have chiral (inversion) symmetry (a helix with a given wave vector can be either left-handed or right-handed; see Figure 3.1) and it breaks time reversal symmetry (like any other magnetic ordering).

Figure 3.1 Inequivalent “left-handed” and “right-handed” helices obtained from each other on space inversion

Spiral multiferroic materials are a class of multiferroic materials in which a spiral ordering can be induced e.g. due to competing interactions, and the resulting inversion symmetry breaking switches on the Dzyaloshinskii-Moriya interaction, causing the formation of an electric dipole moment [6-9] in a mechanism completely different from that in conventional ferroelectrics. Similarly in non-centrosymmetric superconductors, it has been predicted that a spiral modulation of the order parameter appears, which can produce magnetoelectric effects - the coupling of the supercurrent to the magnetization. [55-57] Recently, spiral helical structures have gained interest for spintronics applications [1-3]. A spin-polarized
current flowing through a heterostructure consisting of ferromagnetic and spiral layers can allow the switching of the magnetization of the ferromagnetic layers, by the mechanism of spin transfer torque [4,5].

In general, an incommensurate spiral ordering can result from frustrated competing interactions [58], or when the sign of the exchange coupling between spins is of an oscillatory nature, such as in the RKKY interaction. Spiral spin states can also originate from distortions of the ideal triangular lattice [59-61]. Dzyaloshinskii-Moriya (DM) interactions can also lead to spiral order or to other noncoplanar spin orders such as the skyrmion crystal.

3.2 Instability of the ferromagnetic state towards spiral ordering

In this section we show, following the approach of Ref. [43], that for some values of the filling fraction and JS/t, an incommensurate spiral ordering is favoured over the ferromagnetic ordering in the double exchange model. Interestingly, the spiral state can have lower energy than the uniform ferromagnetic state even in the limit of infinite JS/t and without the competing antiferromagnetic exchange. Our starting point is the Hamiltonian,

\[ H_{DE} = - \sum_{ij\alpha\beta} t_{ij} \tilde{\psi}_{i\alpha}^\dagger \psi_{j\alpha} - J \sum_{i\beta} \psi_{i\alpha}^\dagger \sigma_{\alpha\beta} \psi_{i\beta} \cdot \mathbf{S}_i. \] (3.1)

with \( J > 0 \), known as the ferromagnetic Kondo lattice model.

Consider a spiral state described by the wavevector \( \mathbf{Q} \) and the spin rotation axis \( \hat{z} \). The spin at \( \mathbf{x}_j = 0 \) is oriented along the \( \hat{z} \) axis and the spin \( \mathbf{S}_j \) at site \( j \) with location \( \mathbf{x}_j \) is rotated an angle \( \theta_j = -\mathbf{Q} \cdot \mathbf{x}_j \) around the spin rotation axis \( \hat{z} \).

\[ \mathbf{S}_j = S(0, \sin \mathbf{Q} \cdot \mathbf{x}_j , \cos \mathbf{Q} \cdot \mathbf{x}_j) \] (3.2)

We have the following form for the Hund’s rule coupling term,

\[ \mathbf{\sigma} \cdot \mathbf{S}_j = S(\sigma_x \cos \mathbf{Q} \cdot \mathbf{x}_j + \sigma_y \sin \mathbf{Q} \cdot \mathbf{x}_j) = S e^{i \sigma_z Q \cdot x_j} e^{-i \sigma_z Q \cdot x_j} \] (3.3)

where \( \mathbf{\sigma} \) is a vector composed of the Pauli spin matrices,

\[ \mathbf{\sigma} = (\sigma_x, \sigma_y, \sigma_z). \] (3.4)

The electron annihilation operators in the co-rotating spin frame, in which the \( \hat{z} \) axis at each lattice site is oriented along \( \mathbf{S}_j \), are given by

\[ \psi_{j\alpha} = e^{i \sigma_z Q \cdot x_j} \psi'_{j\alpha}, \] (3.5)
and we can write the second term in the Hamiltonian in the form that it has for the ferromagnetic state,

$$-JS \sum_{j\alpha} \psi^\dagger_{j\alpha} \sigma^z_{\alpha\beta} \psi^\dagger_{j\beta}.$$  

(3.6)

Equation (3.6) implies that the electron wave vector, $k$, is conserved in the co-rotating spin frame. We can, therefore, use Fourier transform to obtain the electron creation and annihilation operators in the momentum space:

$$c^+_{k\alpha} = \frac{1}{\sqrt{N}} \sum_k e^{ikx_i} \psi^\dagger_{i\alpha}$$

$$c_{k\alpha} = \frac{1}{\sqrt{N}} \sum_k e^{-ikx_i} \psi_{i\alpha},$$  

(3.7)

where $N$ is the number of lattice sites. Combining the Fourier transformation with the spin rotation, we obtain

$$\psi_{j\alpha} = \frac{1}{\sqrt{N}} \sum_k e^{i(k+\sigma_2 Q)x_j} c_{k\alpha},$$  

(3.8)

and the kinetic energy of electrons (the first term in Eq.(3.1)) can be written in the form,

$$T = \frac{1}{N} \sum_{k\alpha\beta} c^+_{k\alpha} e^{-i(k+\sigma_2 Q)x_j} t_{ij}^{\alpha\beta} c_{k\beta}$$

$$= \sum_{k\alpha\beta} c^+_{k\alpha} t^{\alpha\beta}_{k+\sigma_2 Q} c_{k\beta},$$  

(3.9)

where

$$t^{\alpha\beta}_{k+\sigma_2 Q} = \sum_j e^{-i(k+\sigma_2 Q)x} t^{\alpha\beta}_{j0}$$

is the hoping amplitude for the spin spiral state. Here, we have chosen $x_i = x_0$ as a reference site and the site index $j$ in Eq. (3.9) denotes nearest neighbours of the reference site. For small $Q$ we have

$$T \approx T^{(0)} + T^{(1)} + T^{(2)}$$

$$= \sum_{k\alpha} \left[ c^+_{k\alpha} t_k c_{k\alpha} + c^+_{k\alpha} \sigma_2^\alpha \partial_t c_{k\alpha} + \frac{1}{2} c^+_{k\alpha} \partial^2 t_k c_{k\alpha} \right],$$  

(3.10)
where $\vartheta = \frac{1}{2} Q \cdot \nabla_k$. To the second order in $Q$ the difference between the energies of the spiral and ferromagnetic (FM) states is

$$\delta E = - \sum_{\nu} \frac{|(v|T^{(1)}|0)|^2}{E_{\nu} - E_0} + \langle 0|T^{(2)}|0 \rangle. \quad (3.11)$$

$T^{(1)}$ contains $\sigma_x$, the spin-flip operator. Hence, the first term is the energy gained due to the mixing of spin-up and spin-down states, which lowers the energy of the occupied states (in quantum mechanics, the energy correction due to the second order perturbation theory is always negative). When the chemical potential is near the bottom of the spin-down band, the denominator of this term is smaller and hence this term can be larger. The second term represents the increase in the kinetic energy (reduction of the conduction band width) of the electrons in the spiral state. The bandwidth has a maximum value only for the ferromagnetic case.

Below, we numerically calculate the value of $\delta E$ for two models: the continuum model with the quadratic electron dispersion and the tight-binding model Eq.(3.1) on the triangular lattice. We show that the spiral state is more favourable than the ferromagnetic state for a wide range of filling fractions and values for $J_S/t$. It is remarkable that the Kondo Lattice Hamiltonian, which describes only the double exchange ferromagnetism, shows an instability of the ferromagnetic state towards spiral ordering even without the competing antiferromagnetic exchange. That does not necessarily mean that the spin spiral is the ground state of this model, as there can be other, more complex orders, such as multi-q states, that have a lower energy. In addition, there is a tendency for phase separation with different spin orders in different parts of the lattice. We discuss this possibility in Sec 4.6. In the two following sections we assume, for simplicity, that the rotation of spins (the pitch of the spiral) is uniform. This assumption can be justified by the fact that the long range Coulomb interactions between electrons (not included in our model) can suppress the instability towards phase separation.

### 3.2.1 Instability of the uniform ferromagnetic state towards spiral ordering in a continuum double exchange model

For a continuum double exchange model, the onset of the spiral instability was discussed earlier in Ref. [43]. For completeness, we reproduce this derivation here. The Hamiltonian of the model in the laboratory spin frame is
The Hamiltonian in the original (unrotated) frame is given by:

\[ H = \int d^D x \psi^\dagger [-\Delta - J(\mathbf{\sigma}.\mathbf{m})] \psi \]  

(3.12)

where \( \psi(x) \) is a two-component spinor (for simplicity, we omit the spin index on \( \psi \)) describing conduction electrons with the quadratic dispersion in \( D \)-dimensional space. The Hund’s rule coupling of strength \( J \) couples the electron spins to the classical field \( \mathbf{m}(x) \), describing local spins of unit length. Consider, furthermore, a spiral state with the wavevector \( \mathbf{Q} \) and the spin rotation axis \( \hat{y} \). The local spin at the point \( \mathbf{x} \) is rotated through the angle \( \theta(x) = \mathbf{Q}.\mathbf{x} \) around \( \hat{y} \):

\[ \mathbf{m}(x) = \hat{z} \cos \theta(x) + \hat{x} \sin \theta(x) \]  

(3.13)

The Hund’s rule coupling term has the form:

\[ \mathbf{\sigma}.\mathbf{m} = e^{-i\sigma_y \frac{\theta}{2}} \sigma_z e^{i\sigma_y \frac{\theta}{2}} \]  

(3.14)

We then perform a spin rotation such that the Hund’s Rule coupling term goes to the form that it has for the case of ferromagnetic spin alignment,

\[ \psi(x) = e^{-i\sigma_y \frac{\theta(x)}{2}} \psi'(x) \]  

(3.15)

In the rotated spin frame the Hamiltonian is of the form

\[ H = \int d^D x \psi'^\dagger \left[ \left( \mathbf{p} - A \sigma_y \right)^2 - J \sigma_z \right] \psi' \]  

(3.16)

where \( A = \nabla \theta \) is the vector potential generated by the rotation in spin space. For the spiral state with the wave vector \( \mathbf{Q} \), \( A = \mathbf{Q} \). We can assume that \( \mathbf{Q}=Q\hat{z} \) without loss of generality, because in the absence of spin-orbit coupling the axes in spin space are independent of the crystal axes, i.e. model has global spin rotational invariance [25]. The single-electron Hamiltonian in the momentum space becomes:

\[ \hat{h} = k^2 + \frac{Q^2}{4} - (J \sigma_z + Q k_z \sigma_x) \]  

(3.17)

Diagonalising this Hamiltonian, we obtain the electron spectrum with two bands with the electron spin parallel or antiparallel to the local spin:

\[ E^\pm_k = k^2 + \frac{Q^2}{4} \pm \sqrt{Q^2 k_z^2 + J^2} \]  

(3.18)

while for the ferromagnetic state, the electron dispersion is:
We see that $Q$ acts to increase the splitting between the spin-up and spin-down bands, which lowers the energy of the occupied states. This splitting is due to the admixture of the (excited) spin-down states to the spin-up states. The first term $Q^2/4$ raises the energies of all states and corresponds to the band narrowing (decrease in the hopping amplitude) as the system goes away from ferromagnetic spin alignment.

For a one-dimensional system, the instability towards the spiral ordering occurs at all values of the Fermi energy. In two dimensions it occurs when the Fermi surface just touches the bottom of the spin-down band (see Figure 3.2). In three dimensions this instability does not occur at all for the continuum model [25,43].

![Figure 3.2](image)

**Figure 3.2** Schematic of the band structure in two dimensions for ferromagnetic and spiral ordering in the isotropic model, (adapted from Ref. [25])

### 3.2.2 Instability of the ferromagnetic state towards spiral ordering on the triangular lattice

The conditions for a spiral state on the triangular lattice to have a lower energy than the ferromagnetic state can be derived using a similar approach. Consider a spiral state with the wavevector $Q$, in which the spin at site $j$ is rotated through the angle $\theta_j = -Q \cdot \hat{x}_j$ around the spin rotation axis $\hat{x}$. The Hamiltonian in the original (unrotated) frame is

$$E^\pm_k = k^2 \pm J$$

(3.19)
\[ H = T + H_f = - \sum_{\langle ij \rangle} t_{ij} \psi_{i\alpha}^\dagger \psi_{j\alpha} - J \sum_j S_j \psi_{j\alpha}^\dagger \sigma \psi_{j\alpha}. \] (3.20)

As before, we perform the transformation to a frame, in which the spin ordering is ferromagnetic: \( \psi_{j\alpha} = e^{i\sigma_j \theta_j} \psi_{j\alpha}' \) where \( \theta_j = -Q \cdot x_j \). We have then the following modification to the kinetic energy term for small \( Q \):

\[ T = -\sum_{ij} t_{ij} \psi_{i\alpha}^\dagger e^{i\sigma_x \frac{1}{2}(x_i - x_j)} \psi_{j\alpha}' \] (3.21)

\[ \approx -\sum_{ij} t_{ij} \psi_{i\alpha}^\dagger \left[ 1 + i \frac{\sigma_x}{2} Q \cdot (x_i - x_j) - \frac{1}{8} \left( Q \cdot (x_i - x_j) \right)^2 \right] \psi_{j\alpha}' \] (3.22)

\[ = -\sum_k t_k \psi_{k\alpha}^\dagger \psi_{k\alpha} + T_1 + T_2, \] (3.23)

where

\[ t_k = \frac{1}{N} \sum_{ij} e^{-ik(x_i - x_j)} t_{ij} \] (3.24)

\( N \) being the number of lattice sites, and \( T_1 \) and \( T_2 \) are the first-order and second-order corrections to \( T \), respectively. \( T_1 \) is given by

\[ T_1 = \sigma_x Q \cdot t_k^{(1)}, \] (3.25)

where we define \( t_k^{(1)} \) as

\[ t_k^{(1)} = \frac{i}{2N} \sum_{ji} e^{-ik(x_i - x_j)} t_{ji}(x_j - x_i). \] (3.26)

\((x_j - x_i)\) is an antisymmetric function inside the summation, so we need to keep only the antisymmetric part of the exponential, \(-i \sin k \cdot (x_j - x_i)\), and

\[ t_k^{(1)} = \frac{1}{2N} \sum_{ji} (\sin k \cdot (x_i - x_j)) t_{ji}(x_i - x_j). \] (3.27)

If we chose \( i = 0 \) with \( x_i = 0 \) as an arbitrary reference site on the lattice, then

\[ t_k^{(1)} = \frac{1}{2} \sum_{j(0)} t_{i(0)} (\sin k \cdot x_j) x_j \] (3.28)
where the summation is over the sites \( j \) that are the nearest neighbours of the site 0 and \( x_j \) are now the vectors joining the reference site to its (six) nearest neighbors. Applying this result to the triangular lattice, with \( \lambda = 1, 2 \) labeling the two crystallographic axes (see Figure 3.3), where

\[
e_1 = (1,0), \quad e_2 = \left( -\frac{1}{2}, \frac{\sqrt{3}}{2} \right), \quad e_3 = \left( -\frac{1}{2}, -\frac{\sqrt{3}}{2} \right),
\]

(3.29)

and \( x_j = \pm e_\lambda a \), the expressions for \( t_k^{(1)} \) and \( T_1 \) become

\[
t_k^{(1)} = ta \sum_\lambda e_\lambda (\sin k \cdot e_\lambda a)
\]

(3.30)

\[
T_1 = -\sigma_x ta \sum_\lambda (Q \cdot e_\lambda)(\sin k \cdot e_\lambda a)
\]

(3.31)

The second-order correction term,

\[
T_2 = \frac{1}{8} Q^\alpha Q^\beta \sum_{ij} d^+_i t_{ij} (x_i - x_j)^\alpha (x_i - x_j)^\beta d_j,
\]

(3.32)

is simplified in momentum space:

\[
T_2 = \frac{1}{8} Q^\alpha Q^\beta \sum_k t_k^{\alpha \beta} d_k^+ d_k,
\]

(3.33)

where

\[
t_k^{\alpha \beta} = \sum_j e^{-ikx_j} t_{j0} x_j^\alpha x_j^\beta.
\]

(3.34)

Using symmetry under \( x_j \rightarrow -x_j \), we can rewrite the last equation in the form,

\[
t_k^{\alpha \beta} = \sum_j (\cos k \cdot x_j) t_{j0} x_j^\alpha x_j^\beta
\]

(3.35)

\[
= 2ta^2 \sum_{\lambda=1}^3 \cos((k \cdot e_\lambda) a) e_\lambda^\alpha e_\lambda^\beta.
\]

(3.36)

The Hund’s rule coupling term after the rotation takes the form that it has for the ferromagnetic state. Using \( S_j = S n_j \) for the localized spins,

\[
H_j = -J \sum_j S_j c_j^+ \sigma c_j
\]
The Hamiltonian in the rotated frame takes the form

\[ H' = T + T_1 + T_2 + H_J \]

\[ = -\sum_k t_k d_k^+ d_k \]

\[ - \sigma_x t^a \sum_{\alpha=1}^3 (Q \cdot e_\alpha) (\sin k \cdot e_\alpha \alpha) + \frac{1}{8} Q^a Q^\beta \sum_{ij} d_i^+ t_{ij} (x_i - x_j)^\alpha (x_i - x_j)^\beta d_j \]

\[ - J \sum_j d_j^+ \sigma_x d_j. \]  

(3.37)

The first-order term \( T_1 \) gives the negative correction to energy proportional to \( Q^2 \) in the second order of perturbation theory (since this term contains the spin-flip operator, its contribution in the first order perturbation theory is zero). Another correction proportional to \( Q^2 \) comes from \( T_2 \) in the first order of perturbation theory, since this term has a nonzero expectation value in the ground state. Summing these two contributions, we obtain the following expression for the energy of the spiral state,

**Figure 3.3** The crystallographic axes (Eq. 3.29) for the triangular lattice
\[ E = - \sum_{k, \sigma} t_k n_{k \sigma} - J S \sum_{k, \sigma} \sigma n_{k \sigma} - \frac{(t a)^2}{2 J S} \sum_k n_{k 1} (1 - n_{k 1}) \left( \sum_{\lambda = 1}^{3} (Q \cdot e_{\lambda}) \sin(k \cdot e_{\lambda} a) \right)^2 + \frac{ta^2}{4} Q^\alpha Q^\beta \sum_{k, \sigma} n_{k \sigma} \sum_{\lambda = 1}^{3} \cos(k \cdot e_{\lambda} a) e_{\lambda}^\alpha e_{\lambda}^\beta. \]

\[ (3.39) \]

\( n_{k 1} \) is the number of electrons in the state in the lower (upper) band with wavevector \( k \), and can take values 0 or 1 when that state is unoccupied or occupied respectively. The factor \( n_{k 1} (1 - n_{k 1}) \) is nonzero only for those values of \( k \), for which the state in the lower band is occupied state and the state in the upper band is unoccupied. The spiral state appears when the difference in energies of the spiral and ferromagnetic states,

\[ E_{\text{spiral}} - E_{\text{FM}} = - \frac{(t a)^2}{J S} \sum_k n_{k 1} (1 - n_{k 1}) \left( \sum_{\lambda = 1}^{3} (Q \cdot e_{\lambda}) \sin(k \cdot e_{\lambda} a) \right)^2 + \frac{ta^2}{4} Q^\alpha Q^\beta \sum_{k, \sigma} n_{k \sigma} \sum_{\lambda = 1}^{3} \cos(k \cdot e_{\lambda} a) e_{\lambda}^\alpha e_{\lambda}^\beta, \]

\[ (3.40) \]

becomes negative.

We can already see that for large \( J S \), the factor \( n_{k 1} (1 - n_{k 1}) \) causes the first term in Eq (3.40) to be most negative at half filling (when the lower band is completely occupied), and zero for filling fraction 0 and 1. For \( a = 1, t = 1 \) and arbitrary fillings of the spin-up and spin-down bands, we numerically calculated the value of \( J S \), at which this instability occurs. The results and plotted in Figures 3.4 and 3.5. The lattice of 200x200 sites was used for the calculations. At half filling, the second term in Eq. (3.40) is zero and, therefore, the transition to a spiral state occurs at all values of \( J S \). At a general filling fraction, the spiral state is lower in energy than the ferromagnetic state, at sufficiently small \( J S \). In the limit of infinite \( J S \), the ferromagnetic state always has energy lower than the spiral, except at half filling.
Figure 3.4 The $JS$ versus filling fraction phase diagram showing the green region where a spiral state on the triangular lattice has lower energy than the ferromagnetic state.

Figure 3.5 Phase diagram for a smaller interval of $JS$ showing the green region where a spiral state has a lower energy than the ferromagnetic state.

To conclude, in this chapter we studied stability of the FM state against the transition into an incommensurate spiral state. We calculated the energy difference between the spiral and ferromagnetic states, given by Eq. (3.40), and found the region in the Hund’s rule coupling,
JS, versus the electron filling fraction, \( f = \frac{N_e}{2N} \), diagram where the spiral state has a lower energy.

The origin of the instability of the ferromagnetic state towards spiral ordering is the lowering of electron kinetic energy due to the mixing between the spin-up and spin-down states in the spiral state. This mechanism is similar to that discussed in section 2.4. The instability takes place when this energy decrease exceeds the increase due to the reduction of the hopping amplitude in the spiral state.

The expression for the energy difference Eq.(3.40), valid for small values of the helix wavevector \( \mathbf{Q} \), gives \( E_{\text{spiral-FM}} \) proportional to \( Q^2 \). It cannot be used to find the magnitude or the direction of \( \mathbf{Q} \) for which the energy has a minimum. Owing to the symmetry of the triangular lattice, one has to calculate \( E_{\text{spiral-FM}} \) up to the sixth order in \( \mathbf{Q} \) in order to determine the wave vector of the spiral. The optimal value of \( \mathbf{Q} \) will be found in the next chapter.
Section 4

Double Exchange model on the triangular lattice

The triangular lattice has been widely studied as the archetype for a system exhibiting geometric frustration [61], an example being the triangular Ising antiferromagnet. Geometric frustration can give rise to novel magnetic phases, such as the tetrahedral state and skyrmion crystal state, and interesting topological effects [62-66]. In this chapter we discuss free (itinerant) electrons on the triangular lattice and the ferromagnetic state. We discuss how nesting at 3/4 filling stabilizes the four-sublattice tetrahedral state [19] and the relation between the chiral nature of this state and the topological Hall Effect. The dispersion relation for the spiral magnetic ordering on the triangular lattice is derived and other possible types of magnetic order are also discussed. A phase diagram is constructed using numerical calculations of the energies of various competing phases.

4.1 Itinerant electrons on the triangular lattice

In this section we discuss free electrons on the triangular lattice. The basis vectors for the triangular lattice are, (see fig 4.1)

\[ a_1 = -\frac{a}{2}\hat{x} - \frac{\sqrt{3}}{2}\hat{y}, \]
The basis vectors for the triangular lattice

\[ \mathbf{a}_2 = a \hat{x}. \quad (4.1) \]

The reciprocal lattice vectors are then

\[ \mathbf{a}_1^* = -\frac{4\pi}{\sqrt{3}a} \hat{y} \quad \text{and} \]
\[ \mathbf{a}_2^* = \frac{4\pi}{\sqrt{3}a} \left( \frac{\sqrt{3}}{2} \hat{x} - \frac{\sqrt{3}}{2} \right). \quad (4.2) \]

We can consider \( a = 1 \) without loss of generality. In our calculations we consider only one orbital per lattice site available for the occupation by itinerant electrons. Therefore each lattice site shall contribute two states available for occupation by the electrons. The filling fraction \( f \) is defined as \( f = N_e / 2N \) where \( N_e \) is the total number of electrons and \( N \) is the total number of lattice sites. The Hamiltonian that describes this system contains only the kinetic energy term,

\[ H = -\sum_{ij\alpha} t_{ij} \psi_{i\alpha}^\dagger \psi_{j\alpha}. \quad (4.3) \]

We consider \( t_{ij} = t \) for nearest neighbour sites, and \( t_{ij} = 0 \) otherwise. The kinetic energy Hamiltonian in momentum representation has the form,

\[ H = -\frac{t}{N} \sum_{ij\alpha} \sum_{kk'} e^{-ikx_i} c_{k\alpha}^\dagger c_{k'\alpha} e^{ik'x_j}. \]

Summing over all nearest neighbours after taking \( x_j = 0 \), the Hamiltonian can be written in the following form,

\[ H = -2t \sum_{\alpha} \sum_{k} (\cos k \cdot \mathbf{a}_1 + \cos k \cdot \mathbf{a}_2 + \cos k \cdot \mathbf{a}_3) c_{k\alpha}^\dagger c_{k\alpha}. \quad (4.3) \]
The free-electron \((J=0)\) dispersion relation is thus

\[
E(k) = -2t(cos k \cdot a_1 + cos k \cdot a_2 + cos k \cdot a_3),
\]

with \(a_3 = a_1 + a_2\). The energy spectrum has double degeneracy; two electrons of opposite spin can occupy each \(k\) state. The band energy has a minimum value of \(-6t\) and a maximum value of \(+3t\). It is a metallic state at all fillings (doping levels). We set \(t = 1\) in our calculations (energy is measured in units of \(t\)) unless stated otherwise.

The contour plot of the energy dispersion in reciprocal space (see figure 4.2) shows that the Fermi surface is flat at \(f=0.75\) (for which the Fermi energy equals \(2t\)). This immediately

---

**Figure 4.2** The energy dispersion for free (itinerant) electrons on the triangular lattice (a) a 2D plot of \(E(k)\) and (b) a contour plot of equipotential surfaces, is plotted in cartesian coordinates in reciprocal space. In (a) the vertical axis depicts energy in units of \(t\).
suggests that the flat parts of the Fermi surface can be connected by nesting vectors leading to a transition from the ferromagnetic metallic state to the insulating Spin-Density Wave (SDW) state. Martin and Batista [19] showed that this mechanism stabilizes the tetrahedral “3Q state” with a 4-sublattice non-coplanar spin ordering, in which the angle between neighboring spins is 109.47°. We shall discuss this state in detail in the next section.

Mathematically, the requirement for a vector $Q$ to be a nesting vector at a given filling at a given wave vector $p$ is,

$$E(p + Q) - \mu = \mu - E(p),$$

where $\mu$ is the Fermi energy at T=0K. For the case of free electrons on the triangular lattice, the nesting vectors $Q$ that satisfy this property at 3/4 filling and $E_f=\pm 2t$ are,

$$Q_1 = \frac{2\pi}{a} (1,0) \quad Q_2 = -\frac{2\pi}{a} \left( \frac{1}{2}, \frac{\sqrt{3}}{2} \right) \quad Q_3 = \frac{2\pi}{a} \left( -\frac{1}{2}, \frac{\sqrt{3}}{2} \right).$$

A periodic magnetic ordering on the lattice with its reciprocal lattice vectors equal to $Q_1$, $Q_2$ or $Q_3$ would be stabilized by the nesting mechanism at $f=3/4$, therefore it would not be expected to be stable if $f$ is changed from $\frac{3}{4}$.

Another possibility for finding nesting vectors appears when the fermi energy has the value $-2t$ near $f=1/4$. At this value of the fermi energy we find vectors that nest the Fermi surface of itinerant electrons, albeit at very few points because the Fermi surface is almost circular at

![Figure 4.3 Examples of vectors (arrows) nesting the Fermi surface of itinerant electrons at $\frac{3}{4}$ filling. The dotted lines show the Fermi surface at $f=3/4$.](image_url)
this filling (see Fig 4.4). Nesting may not be sufficient to provide a mechanism to drive a transition to a state with this ordering at this filling fraction. Yet we shall discuss the significance of this observation in relation to the 3q-ordered state (the “tetrahedral” state) in section (4.3).

4.2 The Ferromagnetic state

For the ferromagnetic state, the Hund’s Rule coupling term becomes simply $\pm JS$ for the spin up (down) band, and the kinetic energy term has the form that it has for the free electron dispersion on the triangular lattice,

$$E(k)_{\pm} = -2t(cosk \cdot a_1 + cosk \cdot a_2 + cosk \cdot a_3) \mp JS. \quad (4.7)$$

There are two nondegenerate bands, the lower one for electronic states representing electrons with spins parallel to the local atomic spins, and the higher one for those with antiparallel spins. The overall energy splitting between the spin-up and spin-down bands is $2JS$ and it is an insulating state for $f=1/2$ and large JS ($JS>4.5$). For all values of $f$ (except for $f=1/2$, when a spiral has lower energy, as we saw in section 3.2.2) at sufficiently large JS, the ferromagnetic state has energy lower than that of any other magnetic order on the triangular lattice.
4.3 The 4-sublattice “tetrahedral” (3Q-ordered) state

The “tetrahedral” state had been proposed to describe the nuclear spin ground state of a two-dimensional $^3$He [60] and the magnetic moments of Mn monolayers on a Cu (111) plane substrate[20,21]. Martin and Batista [19] realized that this interesting noncoplanar chiral ordered state would be stable at $\frac{3}{4}$ filling because of the perfect nesting of the Fermi surface of the non-interacting conduction electrons by the ordering vectors (see Fig 4.7). Due to its nonzero scalar chirality, it exhibits the Topological Hall Effect [10,13]. Numerical simulations by Akagi and Motome showed that this state appeared in the phase diagram at $\frac{1}{4}$
filling, \( \frac{1}{4} \) filling, and near 0.57 filling. [12] These authors, however, did not consider incommensurate spiral states. We find that when the spiral states are taken into account, this state still exists in the phase diagram close to \( \frac{1}{4} \) and \( \frac{3}{4} \) fillings, as we shall discuss in section 4.6. The spin configuration for the four site unit cell is,

\[
S_1 = \frac{1}{\sqrt{3}}(1, 1, 1)
\]
\[
S_2 = \frac{1}{\sqrt{3}}(1, -1, -1)
\]
\[
S_3 = \frac{1}{\sqrt{3}}(-1, 1, -1)
\]
\[
S_4 = \frac{1}{\sqrt{3}}(-1, -1, 1).
\]

\[\text{(4.8)}\]

**Figure 4.6** The basis vectors for a 4-sublattice ordering on the triangular lattice

The unit cell has quadrupled in size as compared to that for the ferromagnetic case, and the basis vectors are

\[
a_1 = -a\hat{x} - \sqrt{3}\hat{y}
\]
\[
a_2 = 2a\hat{x}.
\]

\[\text{(4.9)}\]

The Brillouin zone of the tetrahedral state is four times smaller than the Brillouin Zone of the ferromagnetic state and accordingly contains 8 bands. These bands are doubly degenerate because the tetrahedral state on the triangular lattice has a symmetry - the spin structure remains invariant under a translation (along any of the crystal axes, e.g. along \( \mathbf{r}_{12} \)) combined with a rotation of 180 degrees. The tetrahedral state can be expressed as a 3q-noncoplanar state, with the order parameter

\[
S(r) = (S_a\cos Q_a \cdot r, S_b\cos Q_b \cdot r, S_c\cos Q_c \cdot r),
\]

\[\text{(4.10)}\]

where

\[
Q_a = -\frac{a_1}{2} = \frac{2\pi}{\sqrt{3}a} \hat{y},
\]
\[
Q_b = -\frac{a_2}{2} = \frac{2\pi}{\sqrt{3}a} \left( \frac{\sqrt{3}}{2}, -\frac{1}{2} \right),
\]
\[
Q_c = -\frac{a_3}{2} = \frac{2\pi}{\sqrt{3}a} \left( \frac{\sqrt{3}}{2}, \frac{1}{2} \right).
\]
\[ Q_c = -\frac{a_1 + a_2}{2} = \frac{2\pi}{\sqrt{3}a} \left( -\frac{\sqrt{3}}{2}, -\frac{1}{2} \right), \]  

such that the ordering vectors \( Q_a, Q_b, \) and \( Q_c \) satisfy \( Q_a + Q_b + Q_c = 0 \) and \( 2Q_\eta = G \) where \( G \) is a reciprocal lattice vector for free electrons on the triangular lattice. \( Q_a, Q_b, \) and \( Q_c \) are reciprocal lattice vectors for the triangular lattice with this 4-sublattice “tetrahedral” ordering.

The dispersion relation (Eq. 4.4) for itinerant electrons can be used to verify that \( Q_a - Q_b \), \( Q_b - Q_c \) and \( Q_c - Q_a \) as well as \( Q_a, Q_b, \) and \( Q_c \) are vectors that nest the Fermi surface of itinerant electrons at \( 3/4 \) filling and equally satisfy the mathematical condition for nesting:

\[
\begin{align*}
E(p + Q) - \mu &= E(p + Q_a - Q_b) - \mu \\
&= E(p - Q_c - 2Q_b) - \mu \\
&= E(p - Q_c) - \mu = E(p + Q_c) - \mu \\
&= \mu - E(p)
\end{align*}
\]

(4.12)

**Figure 4.7** The nesting vectors (Eq. 4.11) at \( 3/4 \) filling. Also shown are the reciprocal lattice vectors (Eq. 4.2) for itinerant electrons on the triangular lattice.
4.3.1 The stability of the tetrahedral state at $\frac{1}{4}$ and $\frac{3}{4}$ filling

As reported by Martin and Batista [19], the tetrahedral magnetic ordering is stabilized by perfect nesting of the Fermi Surface of the itinerant electrons at $\frac{3}{4}$ filling and as expected as a result of the nesting properties of the Fermi Surface, a gap exists between the upper two bands down to arbitrarily small $J_S/t$ at $\frac{3}{4}$ filling. This gap appears at the Fermi surface at $\frac{3}{4}$ filling, lowering the energies of the occupied states, letting the tetrahedral state have lower energy.

At a filling fraction $f \sim \frac{1}{4}$ filling, the nesting occurs again but only at a few points because the Fermi surface is almost circular at this filling (see Fig. 4.4). This, in general, may not be sufficient to drive the transition towards an SDW state. Nevertheless, we do observe the tetrahedral state from filling 0.226 to 0.25 in the phase diagram for small $J_S$. This connection of the ordering vectors to the Fermi surface near $\frac{1}{4}$ filling has been called a “generalized Kohn anomaly” and its origin explained as follows [67]. A perturbative expansion in $J$ results in a second order contribution that is degenerate among several spin configurations, due to the frustration intrinsic in the triangular lattice. Only at fourth order does an effective four-spin interaction stabilize the 4-sublattice tetrahedral order, unlike the usual Kohn anomaly which occurs at second order perturbation theory. A local gap is opened at several points on the Fermi surface which develops into a full gap as $J$ or $f$ is increased. This has been postulated to be a universal mechanism in spin-charge coupled systems, which can thus show novel magnetic textures. More comments about the stability of the $\frac{1}{4}$ and $\frac{3}{4}$ filled states shall be made in section 4.3.3.

The stability of this tetrahedral state has been studied at $\frac{1}{2}$ filling in the limit of infinite $J_S$ in the presence of competing antiferromagnetic exchange interactions and found to be the lowest energy state at low temperatures for the values $0.1 < J_S < 0.4$ of the antiferromagnetic exchange, $J_S$ [23]. It was found in Ref. [23] that at $J_S = 0.01$ the chiral phase becomes more stable near $\frac{1}{4}$ filling and $\frac{3}{4}$ filling in the presence of the competing antiferromagnetic exchange. Thus an antiferromagnetic exchange interaction is expected to increase the stability of this chiral state.

4.3.2 Dispersion relation for the tetrahedral state

To find the dispersion relation for electrons on the triangular lattice with the 4-sublattice “tetrahedral” ordering (the summation over the two spin degrees of freedom is implicit):

$$ H = -t \sum_{<ij>} \psi_i^\dagger \psi_j - J \sum_i \psi_i^\dagger \sigma \cdot S_i \cdot \psi_i $$

(4.13)

Taking the Fourier transform,
\[ H = -\frac{t}{N} \sum_{\alpha=1}^{4} \sum_{\langle q \rangle} \sum_{i,j} c_{q,\alpha}^{\dagger} \exp(-i\mathbf{q}.\mathbf{x}_i + i\mathbf{q}'\cdot\mathbf{x}_j) c_{q,\alpha} \]
\[ -\frac{J}{N} \sum_{\alpha=1}^{4} \sum_{i,qq'} c_{q,\alpha}^{\dagger} \sigma \cdot \mathbf{S}_\alpha c_{q',\alpha} \exp(-i\mathbf{q}\cdot\mathbf{x}_i + i\mathbf{q}'\cdot\mathbf{x}_i) \]  

(4.14)

where \( \alpha \) is an index labeling the spin within the 4-site unit cell and \( \mathbf{x}_{i\alpha} = \mathbf{R}_i + a_\alpha \) where the summation \( i \) runs over all unit cells. There are \( N \) lattice sites and \( N/4 \) unit cells.

\[ \begin{pmatrix} c_{1}^{\dagger} & c_{2}^{\dagger} & c_{3}^{\dagger} & c_{4}^{\dagger} \end{pmatrix} \begin{bmatrix} -\mathbf{j}\sigma \cdot \mathbf{S}_1 & -2t \cos \mathbf{q} \cdot \mathbf{r}_{12} & -2t \cos \mathbf{q} \cdot \mathbf{r}_{13} & -2t \cos \mathbf{q} \cdot \mathbf{r}_{14} \\ -2t \cos \mathbf{q} \cdot \mathbf{r}_{21} & -\mathbf{j}\sigma \cdot \mathbf{S}_2 & -2t \cos \mathbf{q} \cdot \mathbf{r}_{23} & -2t \cos \mathbf{q} \cdot \mathbf{r}_{24} \\ -2t \cos \mathbf{q} \cdot \mathbf{r}_{31} & -2t \cos \mathbf{q} \cdot \mathbf{r}_{32} & -\mathbf{j}\sigma \cdot \mathbf{S}_3 & -2t \cos \mathbf{q} \cdot \mathbf{r}_{34} \\ -2t \cos \mathbf{q} \cdot \mathbf{r}_{41} & -2t \cos \mathbf{q} \cdot \mathbf{r}_{42} & -2t \cos \mathbf{q} \cdot \mathbf{r}_{43} & -\mathbf{j}\sigma \cdot \mathbf{S}_4 \end{bmatrix} \begin{pmatrix} c_1 \\ c_2 \\ c_3 \\ c_4 \end{pmatrix} \]

\[ = \begin{pmatrix} c_{1}^{\dagger} & c_{2}^{\dagger} & c_{3}^{\dagger} & c_{4}^{\dagger} \end{pmatrix} \begin{bmatrix} -\mathbf{j}\sigma \cdot \mathbf{S}_1 & -2t \cos \mathbf{q} \cdot \mathbf{r}_{12} & -2t \cos \mathbf{q} \cdot \mathbf{r}_{13} & -2t \cos \mathbf{q} \cdot \mathbf{r}_{14} \\ -2t \cos \mathbf{q} \cdot \mathbf{r}_{21} & -\mathbf{j}\sigma \cdot \mathbf{S}_2 & -2t \cos \mathbf{q} \cdot \mathbf{r}_{23} & -2t \cos \mathbf{q} \cdot \mathbf{r}_{24} \\ -2t \cos \mathbf{q} \cdot \mathbf{r}_{31} & -2t \cos \mathbf{q} \cdot \mathbf{r}_{32} & -\mathbf{j}\sigma \cdot \mathbf{S}_3 & -2t \cos \mathbf{q} \cdot \mathbf{r}_{34} \\ -2t \cos \mathbf{q} \cdot \mathbf{r}_{41} & -2t \cos \mathbf{q} \cdot \mathbf{r}_{42} & -2t \cos \mathbf{q} \cdot \mathbf{r}_{43} & -\mathbf{j}\sigma \cdot \mathbf{S}_4 \end{bmatrix} \begin{pmatrix} c_1 \\ c_2 \\ c_3 \\ c_4 \end{pmatrix} \]

where \( \mathbf{S}_1, \mathbf{S}_2, \mathbf{S}_3 \) and \( \mathbf{S}_4 \) are the four spins (Eq. 4.8) and \( \mathbf{r}_{21} \) is the vector joining site 2 and site 1 as follows,

\[
\begin{align*}
\mathbf{r}_{12} &= \mathbf{r}_{34} = -\mathbf{a}_2 \\
\mathbf{r}_{14} &= \mathbf{r}_{32} = -\left(\mathbf{a}_1 + \mathbf{a}_2\right) \\
\mathbf{r}_{13} &= \mathbf{r}_{24} = \mathbf{a}_1 \\
\mathbf{r}_{13} + \mathbf{r}_{32} + \mathbf{r}_{21} &= 0
\end{align*}
\]

(4.15)

\( \mathbf{a}_1 \) and \( \mathbf{a}_2 \) are the unit vectors of the triangular lattice; see Figure 4.8. Diagonalisation of the \( 8 \times 8 \) matrix in the Hamiltonian yields the dispersion relations for eight bands, which can not be expressed analytically in a simple way. The spectrum has double degeneracy i.e. there are 4 doubly degenerate bands (see Fig 4.9). The tetrahedral ordering on the triangular lattice has a symmetry- the spin structure remains invariant under a translation (along any of the crystal axes, e.g. along \( \mathbf{r}_{12} \)) combined with a rotation of 180 degrees. Accordingly the spectrum is doubly degenerate. The total energy at any given filling fraction and value of JS can be found numerically and compared to that of the other spin configurations. This state is lower in energy than other magnetically ordered states at and near \( \frac{1}{2} \) filling and \( \frac{1}{4} \) filling, for small values of \( J \).
4.3.3. Effective magnetic flux due to the tetrahedral ordering

This state exhibits the Topological Hall Effect, because it has a noncoplanar spin configuration and the hopping amplitudes between the neighbouring sites have a complex phase factor, and a Berry phase can be picked up along certain closed paths in the lattice. The electrons effectively behave as if the closed paths enclosed a magnetic flux. In the infinite J limit we can quantify this effective magnetic flux.

The expression for each of the hopping amplitudes can be calculated using Eq 2.5,

\[ \tilde{t}_{ij} = t_{ij} \hat{u}_i \hat{u}_j \]  \hspace{1cm} (2.5)
We calculate only the spin-dependent part \((u_i^\dagger u_j)\) of the hopping amplitude. The spin-independent part of the hopping amplitude \((t_{ij}\) in Eq. (2.5)) for neighbouring sites is assumed to be equal to 1.

\[
\begin{align*}
t_{12} &= u_i^\dagger u_j \\
&= \cos \frac{\theta_1}{2} \cos \frac{\theta_2}{2} + \sin \frac{\theta_1}{2} \sin \frac{\theta_2}{2} e^{i(\phi_2 - \phi_1)} \\
&= \cos \frac{\theta_1 + \theta_2}{2} e^{i\theta_1} \\
&= \frac{1}{\sqrt{3}} e^{-\frac{\pi}{4}} \\
t_{13} &= \frac{1}{\sqrt{3}} e^{\frac{\pi}{4}} \\
t_{14} &= \frac{1}{\sqrt{3}} e^{0} \\
t_{23} &= \frac{1}{\sqrt{3}} e^{i\pi} \\
t_{24} &= \frac{1}{\sqrt{3}} e^{-\frac{\pi}{4}} \\
t_{34} &= \frac{1}{\sqrt{3}} e^{\frac{\pi}{4}} \\
t_{21} &= t_{12}^* \quad \text{and so on}
\end{align*}
\]

(4.16)

**Figure 4.10** The spin directions of the four spin sub-lattices, and the corresponding hopping amplitudes
These hopping amplitudes can be substituted into the Kondo Lattice Hamiltonian to find the dispersion relation for the tetrahedral ordering in the infinite J limit,

\[
H = -t \sum_{\langle i,j \rangle} (c_j^+ c_i + c_j c_i^+) + JS\sigma_z
\]

\[
= - \sum_{\langle i,j \rangle} t_{ij} c_j^+ c_i + JS\sigma_z
\]

\[
= - \sum_{q \alpha \alpha'} t_{\mathbf{R} \alpha \alpha'} e^{-i\mathbf{q} \cdot \mathbf{r}_{\alpha \alpha'}} c_{q \alpha}^+ c_{q \alpha'} + JS\sigma_z
\]

(4.17)

where \( \mathbf{r} = (\mathbf{R} + \mathbf{a}_\alpha) - (\mathbf{R} + \mathbf{a}_{\alpha'}) \). \( \mathbf{R} \) is the position of the unit cell (the basis vectors of the 4-sublattice state) and \( \mathbf{a}_\alpha \) is the position of the ion within the unit cell. The index \( \alpha \) runs from 1 to 4.

Since there are two \( r \)'s for e.g. \( 1 \rightarrow 4 \) hopping, and \( t_{ij} = t_{ji}^* \), we keep only the real part of the exponential factor,

\[
H = -\frac{2t}{\sqrt{3}} \sum_{\alpha \alpha'} e^{i\phi_{\alpha \alpha'}} \cos \mathbf{q} \cdot \mathbf{r}_{\alpha \alpha'} c_{\alpha}^+ c_{\alpha'} + JS\sigma_z
\]

\[
= -\frac{2t}{\sqrt{3}} (c_1^+ c_2^+ c_3^+ c_4^+) \begin{pmatrix}
0 & \cos \mathbf{q} \cdot \mathbf{r}_{12} e^{i\phi_{12}} & \cos \mathbf{q} \cdot \mathbf{r}_{13} e^{i\phi_{13}} & \cos \mathbf{q} \cdot \mathbf{r}_{14} e^{i\phi_{14}} \\
\cos \mathbf{q} \cdot \mathbf{r}_{21} e^{-i\phi_{21}} & 0 & \cos \mathbf{q} \cdot \mathbf{r}_{23} e^{i\phi_{23}} & \cos \mathbf{q} \cdot \mathbf{r}_{24} e^{i\phi_{24}} \\
\cos \mathbf{q} \cdot \mathbf{r}_{31} e^{-i\phi_{31}} & \cos \mathbf{q} \cdot \mathbf{r}_{32} e^{-i\phi_{32}} & 0 & \cos \mathbf{q} \cdot \mathbf{r}_{34} e^{i\phi_{34}} \\
\cos \mathbf{q} \cdot \mathbf{r}_{41} e^{-i\phi_{41}} & \cos \mathbf{q} \cdot \mathbf{r}_{42} e^{-i\phi_{42}} & \cos \mathbf{q} \cdot \mathbf{r}_{43} e^{-i\phi_{43}} & 0
\end{pmatrix} c_1 \otimes I_2
\]

(4.18)

The matrix is solved for its eigenvalues to yield the dispersion relation for the \( J \rightarrow \infty \) limit,

\[
E_q = \sigma \frac{2t}{\sqrt{3}} \sqrt{\cos^2 q \cdot \mathbf{a}_1 + \cos^2 q \cdot \mathbf{a}_2 + \cos^2 q \cdot \mathbf{a}_3 + \rho J},
\]

(4.19)

which describes eight doubly degenerate bands with an average energy splitting of \( 2JS \) and \( \sigma \) and \( \rho \) take the values +1 or -1 independently.

The effective flux through the plane of the lattice due to the magnetic ordering, can be calculated once the hopping amplitudes are known. Along a path in a magnetic field, electrons pick up a Berry phase related to the path integral of the magnetic vector potential \( \mathbf{A} \),
$\exp \left[ \frac{ie}{\hbar c} \int_{x_i}^{x_f} A \, dx \right]$. 

For a closed path this phase is related to the magnetic flux $\Phi$ through the loop

$$\exp \left[ \frac{ie}{\hbar c} \Phi \right] = \exp \left[ 2\pi i \frac{\Phi}{\Phi_o} \right],$$

where $\Phi_o = \hbar c/e$ is the unit flux. In our case, when the electrons hop around a triangular plaquette $1 \to 2 \to 3 \to 1$ or $1 \to 4 \to 2 \to 1$ (see fig 4.8), the hopping amplitude is given by

$$t_{21} t_{31} f_{13} = \frac{1}{3\sqrt{3}} e^{-i\pi/2}$$

so they acquire a phase of $e^{i\pi}$ or $e^{-i\pi}$ per unit cell of the triangular lattice, which contains two triangles. The magnetic unit cell (hopping around which contributes a phase factor of $e^{i2\pi}$) is twice the size of the unit cell for itinerant electrons on the triangular lattice. Eq (2.9) is also the energy dispersion of electrons on a triangular lattice in a uniform applied magnetic field [68].

The effect of the localized spins with “tetrahedral order” on the electrons is that of a uniform magnetic flux of magnitude $\Phi = \Phi_o/4$ per triangle (see Figure 4.11). Not only does this magnetic flux cause the splitting between the upper two and the lower two bands, this is the value of the flux for which the kinetic energy of the electrons is minimized at ¼ filling [68]. This observation has been called “a generalization of the Peierls instability to orbital magnetic instabilities in two dimensional lattices” in Ref [68]. Accordingly there could exist other robust noncoplanar chiral states at other fillings on the triangular lattice, that minimize the energy of the occupied states through the generation of an effective magnetic field via the

---

**Figure 4.11** The triangular lattice in a uniform applied magnetic field
complex phase of the hopping amplitudes, which opens a gap and lowers the energies of the occupied states. For the case of tetrahedral ordering, this lowering of energy is enough to compensate the increase in energy due to the reduction by a factor of \(1/\sqrt{3}\) of the magnitude of the hopping amplitude (Eq. 2.5) in comparison with its value for the ferromagnetic state.

### 4.4 Dispersion relation for the spiral state

A possibility for magnetic ordering in the Kondo Lattice Model on the triangular lattice is the spiral, and it has not been investigated for a triangular lattice in the literature. The spiral state has been found analytically to be stable towards phase separation for an isotropic model for small fillings \(\lesssim 0.3\) [25] and also for a square and cubic lattice at small values of the Hund’s rule coupling[26]. Studies for the double exchange model on the triangular lattice have focused on commensurate magnetic order [12]. In section 3.2.2 the region of the phase diagram where the long-wavelength spiral state is lower in energy than the ferromagnetic state, was identified using perturbation theory. In this section we analytically derive the dispersion relation for spiral ordering on the triangular lattice.

Consider the helicoidal state with the wavevector \(\mathbf{Q}\) and spin rotation axis \(\hat{y}\). The classical magnetic moment \(\mathbf{m}\) at the lattice site with position \(\mathbf{x}_i\) is given by,

\[
\mathbf{m}(\mathbf{x}_i) = 2 \cos \theta + \mathbf{x} \sin \theta \quad \text{where} \quad \theta(\mathbf{x}_i) = \mathbf{Q} \cdot \mathbf{x}_i = i \theta_i
\]

The second term in the Hamiltonian has the form,

\[
\mathbf{\sigma} \cdot \mathbf{m} = \sigma_z \cos \theta + \sigma_x \sin \theta
\]

\[
= e^{-i \frac{\sigma_y}{2} \theta} \sigma_z e^{i \frac{\sigma_y}{2} \theta}. \quad (4.21)
\]

If we perform the following spin rotation that aligns the spin \(\mathbf{m}\) at every site with the \(z\) axis,

\[
\psi(\mathbf{x}_i) = e^{-i \frac{\sigma_y}{2} \theta(\mathbf{x}_i)} \psi'(\mathbf{x}_i)
\]

(4.22)

the Hamiltonian is modified and the second term takes the form that it has for the ferromagnetic state. The spin indices are implicit.

\[
H = -t \sum_{\langle ij \rangle} \psi_i^\dagger \psi_j - J \sum_i \psi_i^\dagger \mathbf{\sigma} \psi_i \cdot \mathbf{S}_i
\]

\[
= -t \sum_{\langle ij \rangle} \psi_i^\dagger \exp \left(i \frac{\sigma_y}{2} \theta_i - i \frac{\sigma_y}{2} \theta_j \right) \psi_j - JS \sigma_z \sum_i \psi_i^\dagger \psi_i'. \quad (4.23)
\]

Taking the Fourier transform,
\[ \psi = e^{-i \frac{\sigma_z}{2} \theta(x_i)} \psi' \] which is required for continuity of the wavefunction to
be invariant under a translation along the basis vectors of the lattice e.g. along \( \mathbf{R} = N_1 \mathbf{a}_1 \).

This shall place quantisation conditions on the helix wavevector \( \mathbf{Q} \). Consider a unit cell such that there are an \( N_1 \) and \( N_2 \) number of k-points along the \( \mathbf{a}_1^* \) and \( \mathbf{a}_2^* \) directions – there are \( N_1 \times N_2 \) atoms per unit cell,

\[
\mathbf{Q} = \frac{m_1}{N_1} \mathbf{a}_1^* + \frac{m_2}{N_2} \mathbf{a}_2^*.
\]  

(4.25)

Consider spirals with \( \mathbf{Q} \) aligned along the Cartesian x-axis,

\[
\mathbf{Q} = Q \mathbf{x} = Q \mathbf{a}_2 = Q (a, 0).
\]  

(4.26)

Comparing Eq. (4.25) and (4.26),

\[
\mathbf{a}_1 \cdot \mathbf{Q} = 2\pi \frac{m_1}{N_1} = Qa_1 \cdot a_2 = -\frac{Q}{2} \implies Q = -\frac{4m_1\pi}{N_1},
\]

\[
\mathbf{a}_2 \cdot \mathbf{Q} = 2\pi \frac{m_2}{N_2} = Qa_2 \cdot a_2 = Q \implies Q = \frac{2m_2\pi}{N_2}
\]

Hence for \( N_1 = N_2 \) we have that \( m_2 = -2m_1 \) is the condition required for \( \mathbf{Q} \) to be along \( \mathbf{x} \)

\[
Q = \frac{m_1}{N_1} (\mathbf{a}_1^* - 2\mathbf{a}_2^*) = \frac{m_1}{N_1} (-\frac{4\pi}{\sqrt{3}} \mathbf{y} - 4\pi \mathbf{x} + \frac{4\pi}{\sqrt{3}} \mathbf{y}) = -\frac{4m_1\pi}{N_1} \mathbf{x}
\]

Now we consider the requirement on the spinors to be invariant under a translation along the basis vector \( \mathbf{R} = N_1 \mathbf{a}_1 \),

\[
\implies e^{-i \frac{\mathbf{a}_1}{2} \theta (N_1 \mathbf{a}_1)} = 1 \implies \frac{1}{2} N_1 \mathbf{a}_1 = 2n\pi \implies m_1 \text{ must be a multiple of 2}
\]

Consider, as an example, the 3-sublattice coplanar 120° ordered state on the triangular lattice, which is a helix along a crystallographic axis with wavevector \( Q = \frac{2\pi}{1.5} = \frac{4\pi}{3} \). The helix comes “full circle” after 1.5, or equivalently, 3 lattice periods (when \( \theta \) has changed by \( 4\pi \)). From the conditions found above (\( Q = -\frac{4m_1\pi}{N_1} \)) we find that \( N_1 \) must be a multiple of 3 to make the 120° ordered state possible as a specific case of spiral ordering. Similarly the Antiferromagnetic state with wavevector \( Q = 2\pi \) is only exhibited if \( N_1 \) is a multiple of 2.

From the \( D_6 \) symmetry of the triangular lattice, this discussion is valid for the helix wavevector \( \mathbf{Q} \) aligned along any of the crystallographic axes, i.e., along a line joining nearest neighbour atoms. Through trial it was found that helices along other directions lie higher in energy.
The total number of spiral configurations allowed is given by \((1+N_1/4)\). In the limit of a unit cell of an infinite size, the number of helices with different wavevectors allowed would be infinite, and the wavevector would change continuously as the parameters JS and n are varied.

\(\lambda = a\)

\(Q = 2\pi i\)

\(\lambda = \frac{3}{2}\)

\(Q = 4\pi\)

\(\lambda = 1.5a\)

\(Q = \frac{2\pi}{1.5} = \frac{4\pi}{3}\)

**Figure 4.12** Spin configurations for specific cases of the spin spiral with (a) \(Q=2\pi\) (2-sublattice antiferromagnetic ordering) (b) \(Q=4\pi\) (ferromagnetic ordering) and (c) \(Q=4\pi/3\) (3-sublattice coplanar state with the spins aligned at 120° angles as shown)
4.5 Other spin configurations

The other spin configurations that we consider for our calculations are detailed in Fig 4.13. In addition to these 13 states that were considered by [12] we consider also the spiral state. (1a), (2a) and (3a) are the spiral state with wave vectors $4\pi$, $2\pi$ and $4\pi/3$ respectively. Some states have an optimisable angle $\theta$. (4a) is the 3q-tetrahedral state, (2b) is the canted state first considered by de Gennes [18]. (3b) and (3c) are equivalent to (3a) for $\theta=\pi/2$ and $\theta=0$ respectively. (2b), (3b), (3c) and (4d) can at some angle be equivalent to the (1a) ferromagnetic state.

![Spin configurations on the triangle lattice considered for our calculations; adapted from [12]](image-url)
4.6 The Phase Diagram

For a given value of JS and n (where n is the filling fraction, \( n = \frac{N_e}{2N} \) where \( N_e \) is the total number of electrons and 2N is the total number of available electronic states; N is the total number of lattice sites) we can find the ground state energies of the 13 spin states in Fig 4.13, and the spiral state. The value of \( E/N \) (the energy per lattice site) is found numerically by using the dispersion relation in k-space and calculating the energy for each point in the Brillouin zone.

At each value of JS and n, the helix wavevector that minimizes the energy can be found (See Figure 4.14). In general an incommensurate spiral is favoured. The energies of all 14 phases can be compared as in Figure 4.15. A phase diagram for values of JS and n is plotted in Figures 4.16 and 4.17 using a lattice of size \( N_1 = 300 \) (see Appendix). 11 of these 14 phases were seen at some point in the phase diagram.

Figure 4.14 Value of \( E/N \) (energy per atom) for the spiral state plotted versus the magnitude of the helix wavevector at various fixed filling fractions n and values of JS (a) n=0.36, JS=1 (b) n=0.9, JS=1 (c) n=0.6, JS=2 (d) n=0.65, JS=0.2
Figure 4.15

$E/N$ (Energy per lattice site) of the tetrahedral, spiral, and 120° ordered states relative to the $E/N$ for the ferromagnetic state at $JS=1$, 10 and 0.2 and $N_z=300$. At each point the helix with the minimum $E/N$ is considered.
Figure 4.16 The T=0 phase diagram for the double exchange model on the triangular lattice.
The regions of phase separation were plotted at each point \( n_j \) where the inverse compressibility is negative. \( E''(n_j) < 0 \) was found according to the following approximation,

\[
E''(n_j) = \frac{E(n_{j+1}) + E(n_{j-1}) - 2E(n_j)}{\Delta^2},
\]

where \( \Delta = n_{j+1} - n_j = n_j - n_{j-1} \). When \( E''(n_j) \) is negative, the state is unstable towards phase separation or towards a transition to a more stable state. The overall profile of the ferromagnetic region in the diagram matches the one predicted from the calculations in section 3.2.2 for the transition from the ferromagnetic state to a spiral order with a small \( Q \) (see Figure 4.18).
Figure 4.18 Data points leading to Figure 3.5 (blue dots) superimposed on the phase diagram of Figure 4.16 to show a matching of the ferromagnetic ordering-spiral ordering transition line.

The tetrahedral state appears near ¾ filling and ¼ filling. At ¾ filling it is stabilized because of the nesting properties of the ordering vectors, which opens a gap and lowers the energies of the occupied states. At ¼ filling the Fermi surface is almost circular so the instability towards tetrahedral ordering at this filling fraction may not have a nesting origin. More comments on the stability of this state have been made in section 4.3. Earlier work on the zero temperature phase diagram had focused on commensurate magnetic order [12]. We have shown that the spiral state is stable for a wide range of parameters. In general the spiral shall be incommensurate. The wavevector Q was found to have a minimum value near the regions of ferromagnetic ordering, and a larger value away from them. At small n and large n, the ferromagnetic state is most stable. At large JS, spiral ordering is found near ½ filling and is expected to persist even in the limit of infinite JS, according to the calculations in section 3.2.2.

4.6.1 Finite Size Effects

Thermodynamic instability (negative compressibility) is seen in thin strips throughout the region of spiral ordering except at some locations except when it is equivalent to the 120° ordering or the (2a) antiferromagnetic state. The thermodynamically unstable regions occur whenever the helix wavevector changes in quantized jumps. This is a size effect due to the finite lattice size used for the calculations. For a larger lattice size, more values of the helix wavevectors would be allowed. In the limit of an infinite lattice the helix wavevector Q would change continuously on changing n, and we expect that there would be no such
instability. The spin spiral state has been found analytically to be stable towards phase separation for an isotropic model for small fillings \( \leq 0.3 \) \[25\] and also for a square and cubic lattice at small JS \[26\].

![Figure 4.19](image)

**Figure 4.19** Points having negative compressibility are shown in grey for a region in the diagram. Such points for the region of spiral ordering were omitted from Figure 4.18.

### 4.6.2 Phase separation

For any two phases to coexist, it is a necessary condition that their thermodynamic and chemical potentials be equal. We can find the state with the minimum grand canonical potential at fixed \( \mu \). Whenever there is a phase transition, \( n(\mu) \) can have a discontinuous jump, and for that range of values of \( n \), the two states on either side of the interval of \( n(\mu) \) can exist as phase-separated states with that value of \( \mu \) throughout the sample. The regions of phase separation according to this criterion are shown in figure 4.20. It is interesting to note that a phase separation exists between the region of ferromagnetic ordering and the spiral phase, when the spiral is equivalent to the (3a) phase. This means that if ferromagnetic domains should form on the triangular lattice, the domain walls are likely to have a spiral (3a) texture. In the limit of an infinite number of atoms in the unit cell, it is likely that phase separation between regions of different helix wavevectors would not occur. Such phase separated regions have not been included in Figure 4.20, because they are artefacts of the finite lattice size. A comparison with Figure 4.21, which contains all the 13 commensurate phases of Figure 4.13 shows that a spiral state is more stable than the tetrahedral phase appearing in Figure 4.21 near 0.56 filling. Also, the spiral state is lower in energy than ferromagnetic ordering, for \( n<0.15 \) and \( n>0.8 \) at small JS.
Our model neglects the significant effects of long range electron-electron interactions, which favour charge neutrality and would oppose the tendency to phase segregate. In the discussion leading to Figure 4.20 we have not considered the interface energies between separated phases, which often give significant contributions to the energy of a magnetically ordered system.

**Figure 4.20** Showing the phase separation regions separating regions that can coexist with the same chemical potential \( \mu \).

**Figure 4.21** Zero temperature phase diagram for the commensurate phases under the Double Exchange Model on the triangular lattice. adapted from [12].
Section 5
Discussion and conclusions

Previous studies of the phase diagram for the double exchange model on the triangular lattice have been focused on commensurate magnetic orders. A noncoplanar chiral state was first found at ¾ filling in both the double exchange and Hubbard models [19] and later at ¼ filling in the double exchange model at zero temperature [12]. It was shown that this unusual state is further stabilized by the competing antiferromagnetic exchange [12]. The temperature dependence of magnetic ordering has been studied at ½ filling [23] and at ¼ filling [24].

In none of these studies an incommensurate spin spiral ordering was taken into account. The results of our analytical and numerical calculations described in this work show that the spiral ordering actually dominates the phase diagram of the double exchange model on the triangular lattice and is the ground state of this model for a wide range of parameters. Our phase diagram looks quite different from the phase diagram obtained in Ref [12] For small values of the Hund’s rule coupling, the spin spiral is the ground state at all electron concentrations except for three narrow regions near special filling factors: 0.22, 0.3 and 0.75. Also for large $JS$, the spiral state occupies the large part of the phase diagram and at half filling it persists at all values of the Hund’s rule coupling.

This study can be extended further by including the antiferromagnetic exchange interaction between localized spins, which we think will make the region occupied by the spiral state even larger. Furthermore, one can consider more realistic double exchange models by considering several orbitals per lattice site and take into account effects of thermal spin fluctuations. There remains a possibility of discovering other unusual spin structures at special electron concentrations, such as the tetrahedral state stabilized by nesting at ¾ filling and by Kohn anomaly at ¼ filling [67].

Another interesting problem is the effect of an applied magnetic field on the spiral state. In the non-centrosymmetric helical magnets, such as MnSi, the magnetic field is known to stabilize the skyrmion crystal state, which can be viewed as a superposition of three helical spirals. Like the tetrahedral spin state, the skyrmion crystal has a non-zero average scalar chirality, which gives rise to the Topological Hall effect. The possibility to find skyrmions in materials with centrosymmetric lattice can be important for the development of “racetrack memory” [2] whereby information is encoded densely in the form of skyrmions in a magnetic nanoribbon moving under an applied current. Such memory would be more stable in the
presence of defects and more efficient and compact than a similar device employing the motion of domain walls.

The double exchange model intrinsically includes the interplay between charge and spin, and was first introduced to describe the strong coupling between electrical transport and magnetization in the colossal magnetoresistance manganites. [17] The rich phase diagram of this model containing many competing phases implies that magnetism in double exchange systems can be controlled with a relatively low-magnitude electrical current through the spin transfer torque [3-5]. Conversely, conductivity at the borderline between commensurate insulating states and metallic spiral states can be controlled with an applied magnetic field as in the colossal magnetoresistance systems. Moreover, non-collinear and non-coplanar magnetic spin structures appearing in this model can result in unusual transport phenomena, such as the Topological Hall effect.
Acknowledgements

I want to thank foremost my supervisor, Prof. Maxim Mostovoy, for the opportunity to learn from him and for his invaluable supervision and guidance throughout the Master thesis research. I thank Prof. Thom Palstra for refereeing my Master thesis. I acknowledge the support of the Zernike Institute for making my Master studies in Groningen possible. I thank the secretaries of the Theoretical Physics group and the SSME group for their help and the arrangements that they made.
Appendix

Notes on the numerical calculations

For numerical calculations of the total energy of a given spin configuration at temperature 0 K at different values of JS and the electron concentration n, a finite lattice size has to be chosen. The software MATLAB was used because it is a fast method when dealing with large arrays and matrices.

We consider a lattice of \( N_1 \) number of atoms along the crystallographic direction \( \mathbf{a}_1 \) and a \( N_2 \) number of atoms along the direction \( \mathbf{N}_2 \) with the total number of atoms \( N = N_1 \times N_2 \). The allowed k-values are:

\[
\mathbf{k} = x_1 \mathbf{a}_1^* + x_2 \mathbf{a}_2^* = \frac{m_1}{N_1} \mathbf{a}_1^* + \frac{m_2}{N_2} \mathbf{a}_2^* \quad \text{where} \quad m_i = 0, 1, ..., (N_i - 1)
\]

Note that in the dispersion relations, \( \mathbf{k} \) shall appear only as \( \mathbf{a}_1 \), \( \mathbf{k} \cdot \mathbf{a}_2 \) or \( \mathbf{k} \cdot \mathbf{a}_3 \)

\[
\mathbf{k} \cdot \mathbf{a}_1 = 2\pi x_1 \quad \mathbf{k} \cdot \mathbf{a}_2 = 2\pi x_2 \quad \mathbf{k} \cdot \mathbf{a}_3 = 2\pi(x_1 + x_2)
\]

Each atom contributes two electronic states to the spectrum, so the total number of electronic states is \( N_e = 2 \times N \).

The filling fraction \( n \) is defined as the fraction of electronic states \( N_e \) that are occupied, and ranges from 0 to 1.

In addition we have to enforce some boundary conditions. For m-sublattice structures, both \( N_1 \) and \( N_2 \) are required to be multiples of m. For the spiral states, these boundary conditions manifest as quantization conditions on the wavevector \( Q \).

We start with obtaining the analytical expression for the dispersion relation for the bands in k space, which is readily obtained for most phases.

A mesh (or lattice) of points is selected in the first Brillouin zone whereby \( \mathbf{k} \cdot \mathbf{a}_1 \) is allowed to range from 0 to \((N_1-1)/N_1\) and \( \mathbf{k} \cdot \mathbf{a}_2 \) is allowed to range from 0 to \((N_2-1)/N_2\)
For a given value of $JS$ the energies are calculated for each point on the grid. These energy eigenvalues are arranged in ascending order and for each filling fraction, the lowest energy states are considered to be occupied (the energies are calculated at $T=0K$). The sum of the energies of the occupied states is calculated for each of the phases and compared to find the phase that gives a minimum value of the energy per atom for a given filling fraction.

The lattice size used is 300x300 atoms which was sufficient to resolve the differences in energy between the different magnetic states, down to very small values of $JS/t$. Along $n$ the number of calculated values for the filling fraction was 10,000 for each value of $JS$. A total of 120 values of equally spaced values of $JS$ were considered between 0 and 8. Analytic expressions for the dispersion relation were used for the ferromagnetic and the spiral states. For all the other states, the eigenvalues of the Hamiltonian were calculated at each point in k-space only after substituting the numerical values of $JS$ and of the grid of points in k-space.

The 4-sublattice states have a quadrupled real-space unit cell, so 8 eigenvalues per point in k-space were calculated for points in the Brillouin Zone of a size quarter that of the (“original”) Brillouin Zone for the itinerant electrons / the ferromagnetic state. The 3-sublattice states have a tripled real-space unit cell and there are 6 eigenvalues per point in k-space. For these states the k-points were summed over the “original” Brillouin zone (so considering 3 times the number of k-points needed) but using a smaller lattice size of 150x150, in order to keep the calculation time reasonably low.

For the (2b), (3b), (3c), (3d) and (4d) states that have a variable, we consider 31 values of the angle over the ranges of angles 0 to $\pi$, 0 to $\pi/2$, $-\pi/3$ to $2\pi/3$, 0 to $\pi$ and 0 to $\pi$ respectively.
References

[17] Zener C 1951 Phys. Rev. 82 403
[34] Khomskii D I “Basic Aspects of the Quantum Theory of Solids” 2010
[38] Zhao 2000 Phys. Rev. B 62, 11639
[56] Edelstein V M, 1990 Solid State Commun. 73, 233
[61] Nakajima T et al 2008 Physical Review B 77 052401
[66] Svistov L E et al 2004 Journal of Experimental and Theoretical Physics Letters 80 204-7