Master 1 Internship by Pierre Adroguer

# A quantum spin wave theory for $AgNiO_2$

May - July 2008

*"Pensar, analizar, inventar no son actos anómalos, son la normal respiración de la inteligencia. Glorificar el ocasional cumplimiento de esa función, atesorar antiguos y ajenos pensamientos, recordar con incrédulo estupor que el doctor universalis pensó, es confesar nuestra languidez o nuestra barbarie. Todo hombre debe ser capaz de todas las ideas y entiendo que en el porvenir lo será."*

J.L. Borges, *Pierre Ménard, Autor del Quijote*

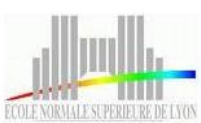

Supervisors:

Dr. N. Shannon, Theoretical Physics,
H. H. Wills Physics Laboratory, University of Bristol

Dr. R. Coldea, Correlated Electron Systems Group,
H. H. Wills Physics Laboratory, University of Bristol

University of Bristol - Max Planck Institut
ENS Lyon






# Contents

# Introduction : $AgNiO_2$

$AgNiO_2$ is a metallic magnetic oxide studied in the Physics department of the University of Bristol. The crystal structure comprises stacked triangular lattices planes ($Ni^{3+}$,$O^{2-}$ and $Ag^{+}$). A reconstruction of neighbouring $O$ ions divides the $Ni$ ions with valence 3+ into a triangular lattice of $Ni^{2+}$ ions and a honeycomb lattice of $Ni$ ions with average valence 3.5+. The $Ni^{2+}$ ions are believed to be well-formed spin S=1 local magnetic moments while electrons on the $Ni^{3.5+}$ ions are itinerant. The triangular lattice of $Ni^{2+}$ ions is magnetically ordered below a temperature $T_N = 19.7\ K$. Order is of a colinear Néel antiferromagnet type, with alternating stripe of up and down spins. The aim of this report is to develop a model of the spin dynamics of the ordered phase of $AgNiO_2$ which can be compared with the experimental data.

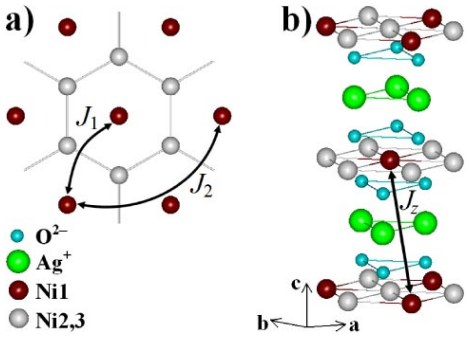



Figure 1: Structure of the $AgNiO_2$ crystal. Right : global structure presenting all the planes of ions. Left : Ni ions configuration in a plane. (Figure from [8]).

The first chapter of this report introduces spin wave theory as an approach to calculating the excitations of a quantum magnet including the leading corrections due to interactions. These techniques are then applied to the simplest model which can reproduce the form of magnetic order seen in a single plane of $AgNiO_2$. The second chapter is about the specific adaptation and application of this theory to a neutron scattering experiments, adding the third dimension to our calculations, and discussing the constraints on parameters set by experiments.

# Chapter 1

# Introduction to spin wave theory and its application to simple 2D models

## 1.1 Spin waves in lattices

### Heisenberg model

To describe the physics in a crystal lattice, we have to assume how the atoms in different sites interact one with each other. Heisenberg proposed the model of magnetic ions interacting with their neighbors by exchange interaction, the energy of a bond being $J\ \vec{S}_i \cdot \vec{S}_j$. The constant J is a characteristic of the intensity of the rate at which electrons are exchanged between magnetic ions. The sign of J affects strongly the interaction since $J < 0$ corresponds to ferromagnetic interaction (the Hamiltonian will tend to have all the spins pointing in the same direction) whereas $J > 0$ corresponds to antiferromagnetism.

The simplest Hamiltonian we can produce with Heisenberg model is the one where each atom only interacts with its nearest neighbors without any exterior magnetic field. We have (the sum runs over all nearest neighbour bonds, each counted once):

$$\mathcal{H} = J \sum_{\langle ij \rangle_1} \vec{S}_i \cdot \vec{S}_j \tag{1.1}$$

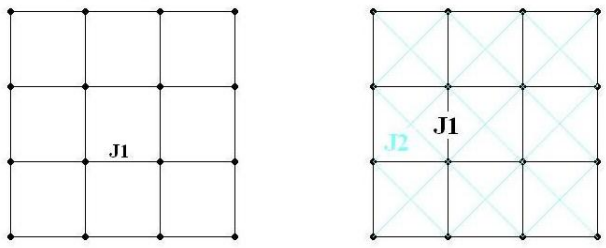



Figure 1.1: Representation of the two Heisenberg models for a square lattice

We can now add a second-range interaction between next-nearest neighbors to this one and we will obtain the $J_1$-$J_2$ Heisenberg model, frequently refered as the $J_1 - J_2$ model :

$$\mathcal{H} = J_1 \sum_{\langle ij \rangle_1} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{\langle ij \rangle_2} \vec{S}_i \cdot \vec{S}_j \tag{1.2}$$

### Spin waves

The classical ground state of the Heisenberg model is the configuration minimizing the energy $\mathcal{H}$ when spins are treated as clasical O(3) vectors. Spin wave theory gives us a systematic way of treating the quantum fluctuations about this state by mapping them onto a set of simple harmonic oscillators. For example, in a ferromagnet, with all the spins being in the same direction, let say $S_z$, these fluctuations will give :

$$\begin{aligned} S_x = 0 &\quad \rightsquigarrow \quad S_x \neq 0 \\ S_y = 0 &\quad \rightsquigarrow \quad S_y \neq 0 \\ S_z = S &\quad \rightsquigarrow \quad S_z = S - n \qquad n \in \mathbf{Z} \end{aligned}$$

This produces a gain in energy which is a quasi particle $a$ (a boson to be precise, the magnetic equivalent of a phonon, then called a *magnon*) with the relation $n = a^\dagger a$. The correct spin algebra is recovered if we write these changes as following, using the relations $S^+ = S_x + iS_y$ and $S^- = S_x - iS_y$[3] :

$$S_z = S - a^\dagger a \tag{1.3}$$

$$S^+ = \sqrt{2S - a^\dagger a}\ \ a \tag{1.4}$$

$$S^- = a^\dagger \sqrt{2S - a^\dagger a} \tag{1.5}$$

$$\vec{S}_i \cdot \vec{S}_j = S_i^z S_j^z + \frac{1}{2}\left(S_i^+ S_j^- + S_i^- S_j^+\right) \tag{1.6}$$

Formally, we treat 1/S as a small parameter (cf 1.3) and introduce a set of bosons $[a, a^\dagger] = 1$ which can be used to represent the spin algebra $[S^x, S^y] = iS^z$.

## 1.2 Simple applications of spin wave theory

### 1.2.1 Ferromagnetism in a square or cubic lattice

Expanding in 1/S order of magnitude around a ferromagnetic ground state the Hamiltonian gives :

$$\begin{aligned} \mathcal{H} &= -|J| \sum_{\langle ij \rangle_1} \vec{S}_i \cdot \vec{S}_j \qquad (1.7) \\ &= \underbrace{-\frac{zN}{2}|J|\,S^2}_{E_0} + |J|\,S \sum_{\langle ij \rangle_1} \left(a_i^\dagger a_i + a_j^\dagger a_j - a_i^\dagger a_j - a_j^\dagger a_i\right) + \mathcal{O}\left(\frac{E_0}{S^2}\right) \end{aligned}$$

where we have put z equal to the number of nearest-neighbors (z=4 for the square lattice, z=6 for the cubic) and N the number of atoms in the crystal.

We now use a simple Fourier transformation to obtain the Hamiltonian as a function of $a^{\dagger}_{\vec{q}} a_{\vec{q}}$, which represents a spin wave with wave vector $\vec{q}$. We neglect all the terms smaller than $E_0/S$ (cf 1.3).

$$a_i = \frac{1}{\sqrt{n}} \sum_{\vec{q}} a_{\vec{q}}\, e^{i\vec{q}\cdot\vec{r_i}} \tag{1.8}$$

$$\mathcal{H} = E_0 + \sum_{\vec{q}} \underbrace{|J|\, Sz\, (1-\gamma(\vec{q}))}_{\omega(\vec{q})}\, a^{\dagger}_{\vec{q}} a_{\vec{q}} \tag{1.9}$$

$$\text{where} \quad \gamma(\vec{q}) = \begin{cases} \frac{1}{2}(\cos q_x + \cos q_y) & \text{for the square lattice} \\ \frac{1}{3}(\cos q_x + \cos q_y + \cos q_z) & \text{for the cubic lattice} \end{cases}$$

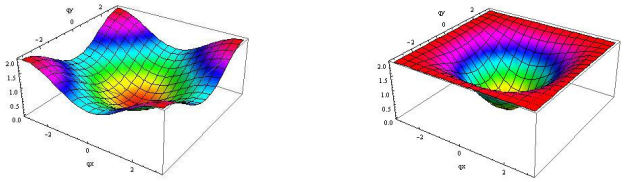


Figure 1.2: Dispersion spectrum for a square lattice ferromagnet, left with $J_1$ model, right with $J_1 - J_2$ model and a ratio $\frac{J_2}{J_1} = \frac{1}{2}$.

The profile of the dispersion in Fig. 1.2 show a parabolic profile at the origin, typical of the ferromagnets. Including an antifarromagnetic second range interaction $J_2$ does not lead to any qualitative changes in the spin wave dispersion for $J_2 \langle J_1/2$.

### 1.2.2 Antiferromagnetism in a square lattice

$AgNiO_2$ has a colinear antiferromagnet ground state. To develop a spin wave theory of this, we will need techniques suited to Néel order. We start here with the simplest model, a Heisenberg model on a square lattice with antiferromagnet nearest neighbours interactions, the colinear antiferromagnet (CAF) being studied in the appendix A.2. First of all, we have to specify what the Hamiltonian is :

$$\mathcal{H} = +|J| \sum_{\langle ij \rangle_1} \vec{S_i} \cdot \vec{S_j} \tag{1.10}$$

As we can see in Fig.1.3, we can consider this lattice as the union of two square sublattices with spins pointing respectively up (blue arrows, refered to as A below) or down (red , B). At each bond, the spins of the two atoms considered point in opposite directions. This will lead to some changes in our description of quantum fluctuations of the spin B (pointing downwards). We will introduce :

$$\begin{aligned} S^z_B &= -S + b^{\dagger} b \\ S^-_B &= \sqrt{2S - b^{\dagger} b}\quad b \\ S^+_B &= b^{\dagger} \sqrt{2S - b^{\dagger} b} \end{aligned}$$

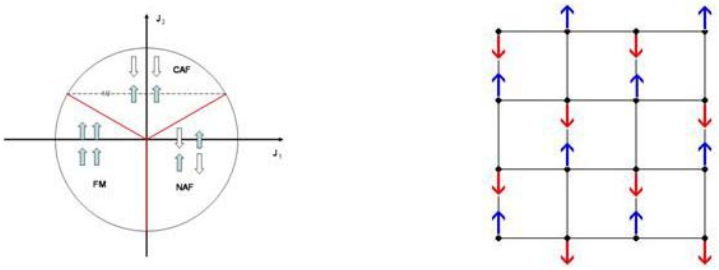



Figure 1.3: Left : Phase diagram of the classical magnetic ground state of the Heisenberg $J_1 - J_2$ model as a function of the values of $J_1$ and $J_2$. Right : Néel antiferromagnetic square lattice. The spins can be represented in the same plane as the lattice because of continous degeneracy of the magnetic ground state(cf. 2.1)

And this leads to a Hamiltonian :

$$\mathcal{H} = -\frac{NzJ}{2}S^2 + zJS\sum_{\vec{q}}\left[a^\dagger_{\vec{q}}a_{\vec{q}} + b^\dagger_{-\vec{q}}b_{-\vec{q}} + \gamma_{\vec{q}}\left(a_{\vec{q}}b_{-\vec{q}} + a^\dagger_{\vec{q}}b^\dagger_{-\vec{q}}\right)\right] \quad (1.11)$$

$$= E_0 + \sum_{\vec{q}}\left[\left(a^\dagger_{\vec{q}}, b_{-\vec{q}}\right)\begin{pmatrix} A(\vec{q}) & B(\vec{q}) \\ B(\vec{q}) & A(\vec{q}) \end{pmatrix}\begin{pmatrix} a_{\vec{q}} \\ b^\dagger_{-\vec{q}} \end{pmatrix} - A_{\vec{q}}\right] \quad (1.12)$$

$$\text{introducing} \quad A(\vec{q}) = zJS$$
$$\text{and} \quad B(\vec{q}) = zJS\gamma_{\vec{q}}$$

The problem now is how to deal with the term proportional to $a_{\vec{q}}b_{\vec{q}} + h.c.$ which represents the destruction (creation) of two spin waves at the same time. This arises from the difference in definition of spin operators between the A and B sublattices. In order to remove this term from the Hamiltonian, we use a Bogoliubov transformation (for detailed calculations refer to Appendix A) to map this problem into a nex set of non-interacting bosons (i.e. a new set of single harmonic oscillators). The (general) result we obtain after performing such a transformation is that :

$$\mathcal{H} = E_0 + \sum_{\vec{q}}\omega_{\vec{q}}\left(\alpha^\dagger_{\vec{q}}\alpha_{\vec{q}} + \beta^\dagger_{\vec{q}}\beta_{\vec{q}}\right) \quad (1.13)$$

$$\omega_{\vec{q}} = \sqrt{A(\vec{q})^2 - B(\vec{q})^2} \quad (1.14)$$

We can see these results in Fig. 1.4, the profile is now conic at the magnetic ordering vectors (here at the corners of the Brillouin zone). This is characteristic of the antiferromagnets.

## 1.3 $\mathcal{O}\left(\frac{1}{S^2}\right)$ and interactions between spin waves

So far, we have carried out calculations in a linear approximation, considering only terms quadratic in boson operators, which means that we have considered

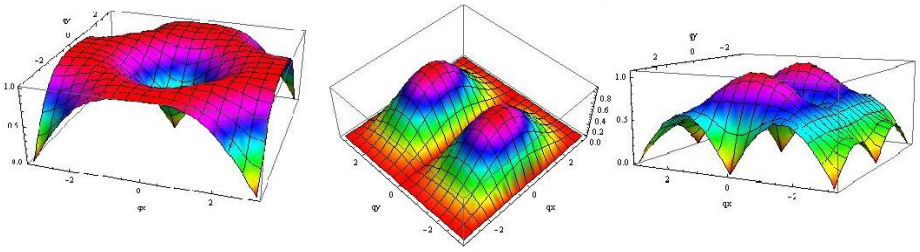


Figure 1.4: Dispersion spectra for antiferromagnetic lattices, from left to right : NAF, CAF ($J_2 = \frac{J_1}{2}$), CAF ($J_1 = J_2$).

the magnons as non-interacting particles. This approximation is a very good one for simple magnets with large spin S. However, $AgNiO_2$ is a more complex quantum magnet in which spin waves strongly interact with each other. We can treat these interactions by calculating the dispersion spectrum to higher order of magnitude, we will present two general ways of doing it with the ferromagnet phase (the same calculations for the NAF are done in appendix B).

## Ferromagnet

We remind what we have done for the calculations of the Hamiltonian of the ferromagnet on a square lattice :

$$\begin{aligned} \mathcal{H} &= -|J| \sum_{\langle ij \rangle_1} \vec{S}_i \cdot \vec{S}_j \qquad (1.15) \\ &= \underbrace{-\frac{zN}{2} |J| S^2}_{E_0} + \underbrace{|J| S \sum_{\langle ij \rangle_1} \left( a_i^\dagger a_i + a_j^\dagger a_j - a_i^\dagger a_j - a_j^\dagger a_i \right)}_{H_{LSW}} + \mathcal{O}\left( \frac{E_0}{S^2} \right) \end{aligned}$$

The LSW (linear spin-wave) term is the one we have calculated previously, it corresponds to the linear part of the Hamiltonian, similar to the harmonic oscillator. The aim of this section is to calculate the last term $\mathcal{O}\left(\frac{E_0}{S^2}\right)$ which is a 1/S correction to LSW theory, abbreviated to "1/S correction" below. This name must be taken carefully, actually, the expansion is formally in order of magnitude of $1/S$ but even when $1/S = \mathcal{O}(1)$ (as in $AgNiO_2$), convergence is guaranteed by the small order of magnitude of $a^\dagger a$. We can use two methods for this, one consists merely in expanding the square root in the formula of $S^+$ and $S^-$ one degree further. This gives:

$$\mathcal{H} = E_0 + H_{LSW} + \frac{1}{N} {\sum_{1,2,3,4}}' \quad \nu_{34}^{12} \quad a_1^\dagger a_2^\dagger a_3 a_4 \qquad (1.16)$$

$$\text{with} \quad \nu_{34}^{12} = \frac{J}{2} \left[ \gamma_1 + \gamma_2 + \gamma_3 + \gamma_4 - (\gamma_{3-1} + \gamma_{3-2} + \gamma_{4-1} + \gamma_{4-2}) \right] \qquad (1.17)$$

We have used the abbreviations $a_1 = a_{k_1}$ and $\gamma_{3-2} = \gamma(k_3 - k_2)$ and the prime means that the sum runs on every quadruplet of vectors of reciprocal space with

respect to the condition $k_1 + k_2 = k_3 + k_4 + G$ where G is a reciprocal lattice vector. Since the bosons are identical, the Hamiltonian should be symmetric by permutation of some indexes (here $1 \leftrightarrow 2$ or $3 \leftrightarrow 4$).

We can now calculate the 1/S correction in a mean-field approximation : once we have established the term with four bosons, we use the mean values of $\langle a^\dagger a \rangle$ to calculate the first correction to $\omega(\vec{q})$. Here, the calculation of this mean value gives (cf appendix B):

$$\left\langle a^\dagger_{\vec{q}} a_{\vec{q'}} \right\rangle = \delta_{q,q'} n_B(\omega_q) \quad \text{where} \quad n_B(x) = \frac{1}{e^{x/k_B T} - 1}$$

We can now treat the interactions between spin waves in a mean field approximation :

$$\mathcal{H} = E_0 + \mathcal{H}_{LSW} + \sum_{1,2} \nu_{21}^{12} a_1^\dagger a_1 \left\langle a_2^\dagger a_2 \right\rangle \tag{1.18}$$

$$\mathcal{H} = E_0 + \left(1 - \sum_{k'} n_B(\omega_{k'}) \frac{1 - \gamma_{k'}}{4NS}\right) H_{LSW} \tag{1.19}$$

Since $n_B(\omega_k) \xrightarrow[T \to 0]{} 0$, the linear spin wave theory is exact for zero temperature. We can use the same method to calculate 1/S correction in an antiferromagnet, but in this case, $\langle a^\dagger a \rangle$ is not the only non-zero average which can be formed of a four-bosons term :

$$\left\langle a_q^\dagger a_{q'} \right\rangle = \left\langle b_q^\dagger b_{q'} \right\rangle = \delta_{qq'} \left( \left(u_q^2 + v_q^2\right) n_B(\omega_q) + v_q^2 \right) \tag{1.20}$$

$$\left\langle a_q^\dagger b_{-q'}^\dagger \right\rangle = \left\langle a_q b_{-q'} \right\rangle = -\delta_{qq'} u_q v_q \left(2 n_B(\omega_q) + 1\right) \tag{1.21}$$

Following this program, we find :

$$\begin{aligned} \mathcal{H} \overset{T \to 0}{=} & \overbrace{JSz \sum_k (\epsilon_k - 1) + \sum_k \omega_k \left(\alpha_k^\dagger \alpha_k + \beta_k^\dagger \beta_k\right)}^{E_0 + LSW} \\ & + \frac{1}{2S} \underbrace{\frac{2}{N} \sum_k (1 - \epsilon_k)}_{A \simeq 0.158} \sum_k \omega_k \left(\alpha_k^\dagger \alpha_k + \beta_k^\dagger \beta_k\right) + \dots \end{aligned} \tag{1.22}$$

The details of the calculations are given in the Appendix B, such as another method to calculate the 1/S correction.

## 1.4 Colinear antiferromagnet

The inelastic neutron scattering experiments performed on $AgNiO_2$ show that the ions are placed on a triangular lattice, along one priviligied direction. In each plane, the spins are oriented along stripes as described in Fig1.5.

Considering colinear antiferromagnetism leads necessarily to a $J_1 - J_2$ model since the $J_1$ Heisenberg model minimizes its energy by having the spins non colinear but oriented along three axis, obtained one from the other by a 120° rotation. A study of this expression gives precise values of the range of the ratio

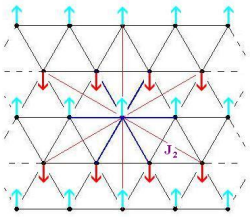



Figure 1.5: Colinear anti-ferromagnet in the triangular lattice configuration

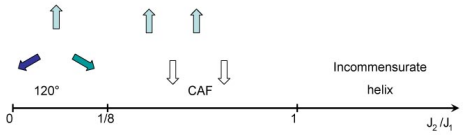



Figure 1.6: Phase diagram of the triangular lattice magnetic ground states in function of the ratio $J_2/J_1$

$J_2/J_1$, it should run from 1/8 to 1[2] (cf Fig. 1.6). This consideration leads to a Hamiltonian :

$$\mathcal{H} = J_1 \sum_{\langle ij \rangle_1} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{\langle ij \rangle_2} \vec{S}_i \cdot \vec{S}_j$$

with $J_1$ and $J_2$ being positive. We re-use the same technique we have seen for the square the square NAF and we obtain :

$$\mathcal{H} = \underbrace{N\left(J_1 + J_2\right) S^2}_{E_0} + \sum_{\vec{q}} \omega\left(\vec{q}\right) \left(\alpha^\dagger_{\vec{q}} \alpha_{\vec{q}} + \beta^\dagger_{\vec{q}} \beta_{\vec{q}}\right) \tag{1.23}$$

$$\omega\left(\vec{q}\right) = \sqrt{A^2 - B^2} \tag{1.24}$$

$$A = 2J_1 S \left(1 + \frac{J_2}{J_1} + \cos q_x + \frac{J_2}{J_1} \cos \sqrt{3} q_y\right) \tag{1.25}$$

$$B = 4J_1 S \cos \frac{\sqrt{3} q_y}{2} \left(\cos \frac{q_x}{2} + \frac{J_2}{J_1} \cos \frac{3 q_x}{2}\right) \tag{1.26}$$

For comparison with experiments, it is convenient to represent the dispersion by plotting its value on a path following symmetry directions of the Brillouin zone. Here we have chosen a path starting from an ordering vector (center of the second Brillouin zone) M $\left(0, \frac{2\pi}{\sqrt{3}}\right)$ then coming back to the center $\Gamma$ $(0,0)$ where the energy is zero. We then go to a corner of the Brillouin zone M' $\left(\pi, \frac{\pi}{\sqrt{3}}\right)$, and finally we follow the edge of the Brillouin zone until its middle $(\pi, 0)$ and go back to the center (cf Fig.1.7). This profile of the dispersion show that for the value $\frac{J_2}{J_1} = \frac{1}{4}$ there is a degeneracy of the ground state : the dispersion is zero at the ordering vectors as expected, but also at the soft point M'. This leads to an instability in the magnetic order within linear spin wave theory. The parabolic profile of the dispersion around M' show that this degeneracy is accidental and should not remain when taking into account the interactions between spin waves.

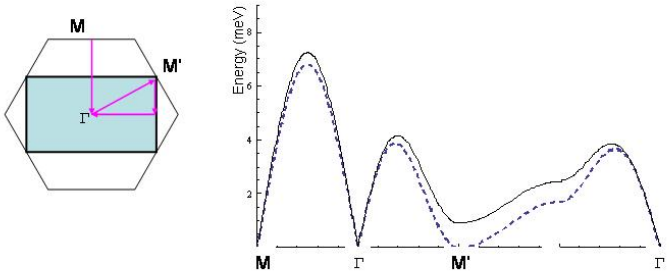



Figure 1.7: Brillouin zone for the ferromagnet (black line) and for the CAF (blue rectangle) and profile of the dispersion following the path drawn in purple for the LSW theory model (dashed line) and with the 1/S correction (solid line), $J_1 = 1.78$ meV, $J_2/J_1 = 0.25$

## 1.5 $1/S$ expansion in the case of the CAF

This apparent contradiction is resolved if we calculate the leading interaction corrections to spin wave in this model. The expansion of the $J_1 - J_2$ model gives, noting $a_n = a_{k_n}$, $\tilde{A} = \frac{A}{2J_1 S}$, $\tilde{B} = \frac{B}{2J_1 S}$ ,$\tilde{\omega} = \frac{\omega}{2J_1 S}$ and the prime on the sum standing for the condition $k_1 + k_2 = k_3 + k_4$ :

$$\begin{aligned}
\mathcal{H} &= E_0 + H_{LSW} + \qquad (1.27)\\
&+ \frac{J_1}{2N} \sum_{1234}{}' (-f_1 - f_2 - f_3 - f_4 + f_{3-1} + f_{3-2} + f_{4-1} + f_{4-2}) \left[ a_1^\dagger a_2^\dagger a_3 a_4 + b_1^\dagger b_2^\dagger b_3 b_4 \right] \\
&\quad - 4 \left( \tilde{B}_{3-2} + \tilde{B}_{4-1} \right) a_1^\dagger b_2^\dagger b_3 a_4 \\
&\quad - 2\tilde{B}_4 \, a_1^\dagger a_{-2} a_3 b_4 - 2\tilde{B}_{3+4-2} \, a_{-1} b_2^\dagger b_3 b_4 - 2\tilde{B}_4 \, a_1^\dagger a_2^\dagger a_3 b^\dagger - 4 - 2\tilde{B}_{4-3-2} \, a_1^\dagger b_2^\dagger b^\dagger_{-3} b_4
\end{aligned}$$

$$f_i = \cos(k_{ix}) + \frac{J_2}{J_1} \cos\left(\sqrt{3} k_{iy}\right) \qquad (1.28)$$

The sum over $k'$ is calculated (it runs over the first magnetic Brillouin zone) and with the $N/2$ prefactor it represents an average of the term summed; we

then obtain the correction to $A_k$ and $B_k$ :

$$\mathcal{H} = E_0 + H_{LSW} + \sum_k \left(a_k^\dagger, b_{-k}\right) \begin{pmatrix} \delta A_k & \delta B_k \\ \delta B_k & \delta A_k \end{pmatrix} \begin{pmatrix} a_k \\ b_{-k}^\dagger \end{pmatrix} \quad (1.29)$$

$$\delta A_k = 2J_1 \frac{2}{N} \sum_{k'} \frac{\tilde{A}_{k'} - \tilde{\omega}_{k'}}{2\tilde{\omega}_{k'}} F(k,k') + \frac{\tilde{B}_{k'}^2}{2\tilde{\omega}_{k'}} \quad (1.30)$$

$$F(k,k') = (1-\cos k_x)(1-\cos k'_x) + \frac{J_2}{J_1}\left(1-\cos\sqrt{3}k_y\right)\left(1-\cos\sqrt{3}k'_y\right) - 2\left(1+\frac{J_2}{J_1}\right)$$

$$\delta B_k = 2J_1 \frac{2}{N} \sum_{k'} \frac{\tilde{A}_{k'} - \tilde{\omega}_{k'}}{2\tilde{\omega}_{k'}} \tilde{B}_k - \frac{\tilde{B}_{k'}}{2\tilde{\omega}_{k'}} G(k,k') \quad (1.31)$$

$$G(k,k') = \cos\frac{\sqrt{3}k_y + k_x}{2}\cos\frac{\sqrt{3}k'_y + k'_x}{2} + \cos\frac{\sqrt{3}k_y - k_x}{2}\cos\frac{\sqrt{3}k'_y - k'_x}{2} + \frac{J_2}{J_1}\left(\cos\frac{\sqrt{3}k_y + 3k_x}{2}\cos\frac{\sqrt{3}k'_y + 3k'_x}{2} + \cos\frac{\sqrt{3}k_y - 3k_x}{2}\cos\frac{\sqrt{3}k'_y - 3k'_x}{2}\right)$$

This reproduces a result for this problem first obtained by A. Chubukov and T. Jolicoeur in [2]. A simple inspection of these results shows that the two corrections annihilates one each other on the ordering vector, but lift the degeneracy at the other point M' as $\delta A \neq 0$ and $\delta B = 0$, this is an "order-from-disorder" phenomenon [7], meaning that the disorder due to quantum fluctuations leads to a long-range stability. The resulting corrections to the spin wave dispersion are shown in Fig. 1.7.

# Chapter 2

# Applications to inelastic neutron scattering experiments on $AgNiO_2$

Researchers of the University of Bristol (Correletad Electron Systems Group) have carried out inelastic neutron scattering experiments on $AgNio_2$. A beam of neutrons is scattered from a powder sample of $AgNiO_2$. By looking at the changes in energy of a neutron for a given change in momentum, one can have a profile of the energy in function of the spin excitation created. A powder is used because of the difficulty to make large crystals. However, using a powder gives only averages of the values on all the directions so calculations of spin wave dispersion must ultimately be averaged over angle. These experiments motivate to improve our model : the fact that each plane containing a triangular lattice interacts with its neighbour is solved by adding an interlayer coupling ; the presence of a gap in the dispersion spectrum is explained by introducing easy-axis anisotropy.

## 2.1 Anisotropy

So far we have showed figures in which the spins were represented along the plane of the crystal. This was possible because of the symmetry of the Hamiltonian, one global rotation of the spin space does not affect the energy of the crystal. This gives a continuous degeneracy that can be lifted by applying a magnetic field to the crystal, or by introducing a term along an axis. We can have :

$$\mathcal{H}' = \mathcal{H} - \underbrace{h \sum_i S_i^z}_{\text{magnetic field}} - \underbrace{D \sum_i S_i^{z\,2}}_{\text{easy-axis anisotropy}} \tag{2.1}$$

The introduction of the easy-axis anisotropy term will change the value in the matrix and of the corrections as follow :

$$A_k = 2J_1 S\left(1+\frac{J_2}{J_1}+\cos k_x+\frac{J_2}{J_1}\cos\sqrt{3}k_y\right)+2DS \tag{2.2}$$

$$B_k = 4J_1 S\cos\frac{\sqrt{3}k_y}{2}\left(\cos\frac{k_x}{2}+\frac{J_2}{J_1}\cos\frac{3k_x}{2}\right) \tag{2.3}$$

$$\omega_k = \sqrt{A_k^2-B_k^2} \tag{2.4}$$

$$\delta A_k = 2J_1\frac{2}{N}\sum_{k'}\frac{\tilde{A}_{k'}-\tilde{\omega}_{k'}}{2\tilde{\omega}_{k'}}F'(k,k')+\frac{\tilde{B}_{k'}^2}{2\tilde{\omega}_{k'}} \tag{2.5}$$

$$\begin{aligned} F'(k,k') = {} & (1-\cos k_x)(1-\cos k'_x) + \frac{J_2}{J_1}\left(1-\cos\sqrt{3}k_y\right)\left(1-\cos\sqrt{3}k'_y\right) \\ & -2\left(1+\frac{J_2}{J_1}\right)-\frac{2D}{J_1} \end{aligned}$$

$$\delta B_k = 2J_1\frac{2}{N}\sum_{k'}\frac{\tilde{A}_{k'}-\tilde{\omega}_{k'}}{2\tilde{\omega}_{k'}}\tilde{B}_k-\frac{\tilde{B}_{k'}}{2\tilde{\omega}_{k'}}G(k,k') \tag{2.6}$$

$$\begin{aligned} G(k,k') = {} & \cos\frac{\sqrt{3}k_y+k_x}{2}\cos\frac{\sqrt{3}k'_y+k'_x}{2}+\cos\frac{\sqrt{3}k_y-k_x}{2}\cos\frac{\sqrt{3}k'_y-k'_x}{2} \\ & + \frac{J_2}{J_1}\left(\cos\frac{\sqrt{3}k_y+3k_x}{2}\cos\frac{\sqrt{3}k'_y+3k'_x}{2}\right. \\ & \left.+\cos\frac{\sqrt{3}k_y-3k_x}{2}\cos\frac{\sqrt{3}k'_y-3k'_x}{2}\right) \end{aligned}$$

We have just seen that the degeneracy of the ground state is lifted by the 1/S expansion, allowing an ordered phase. We are now interested in adding an anisotropic term in order to explain the presence of gap in the experimental spectrum. The spectra found experimentally show the impossibility to have a dispersion under the minimal value $\Delta = 1.78$ meV. This raises the question of knowing which one of the points between the soft point and the ordering vector is the lowest in energy when adding easy-axis anisotropy. If it is not the ordering vector, we would be in presence of a new phenomenon which was not expected. It is then interesting to plot the shift of the two points we are studying in function of the value of the $D$ term in the Hamiltonian. The Fig. 2.1 shows that the effects are different, for the soft point M' it acts linearly $\Delta = \delta A_{M'} + 2DS$ whereas it is in square root for the ordering vector point $\Delta = (A_M + B_M + \delta A_M + \delta B_M + 2DS)^{1/2}\sqrt{2DS}$. Moreover, we can see that for $D > 0.07$ meV the soft point M' becomes the lowest in energy, this being definitive since the asymptotic limit of both plots is the same : $\Delta \longrightarrow 2DS$.

We can now adjust the value of D to fit with experimental data and plot the dispersion with these parameters. This is shown in Fig. 2.2.

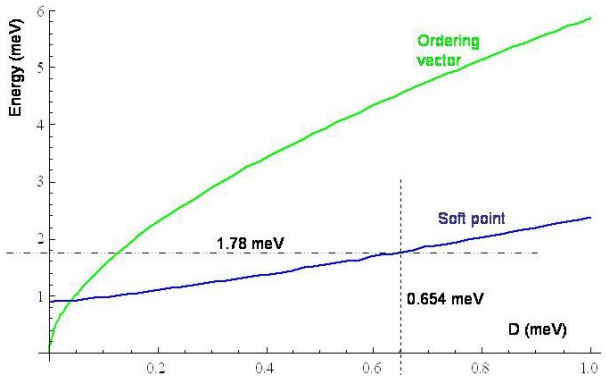



Figure 2.1: Evolution of the energy of a spin wave with wave vector being at the soft point M' (blue line) and at an ordering vector M (green line) in function of the value of $D$.

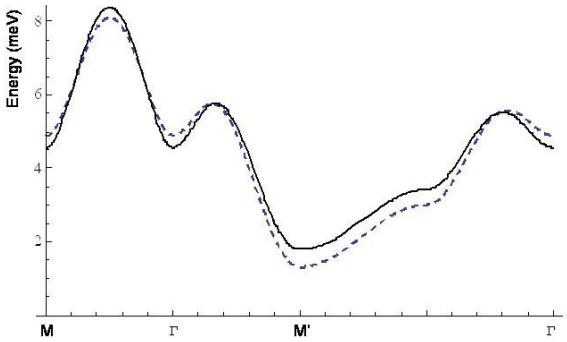



Figure 2.2: Profile of the dispersion with easy-axis anisotropy term for the LSW model (dashed line) and with the 1/S expansion (solid line) with $J_1 = 1.7$ meV, $J_2/J_1 = 0.25$, $D = 0.654$ meV so that the minimal gap is 1.78 meV for LSW+1/S model.

## 2.2 Interlayer coupling

Fine structures in the experimental spectra suggest the interlayer coupling to be not as important $J_1$ or $J_2$, but it must be taken into account. Now we have to consider a different unit cell, formed by four spins, one up and one down in each plane. We have named $a$, $b$, $\tilde{a}$, $\tilde{b}$ the spins on the sublattice 1, 2, 4, 3 (cf. fig. 2.3).

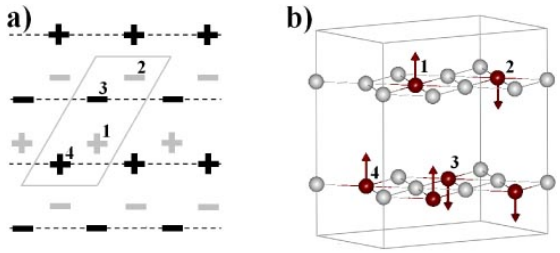



Figure 2.3: Representation of the four-spin magnetic cell in the $AgNiO_2$ crystal. Figure from [8]

In this case, the easiest thing to do is to introduce $X_k^\dagger = \left(a_k^\dagger, \tilde{a}_k^\dagger, b_{-k}, \tilde{b}_{-k}\right)$ and the hamiltonian is written as :

$$\mathcal{H} = E_0 + \sum_k X_k^\dagger \begin{pmatrix} A_k & B_k & C_k & D_k^* \\ B_k^* & A_k & D_k & C_k \\ C_k & D_k^* & A_k & B_k \\ D_k & C_k & B_k^* & A_k \end{pmatrix} X_k + \mathcal{O}(1/S) \quad (2.7)$$

$$A_k = 2S\left[J_1(1+\cos k_x) + J_2\left(1+\cos\sqrt{3}k_y\right) - J_z + D\right] \quad (2.8)$$

$$B_k = 4SJ_z \cos\left(\frac{k_x}{2}\right)\cos\left(\frac{k_z}{2}\right)\delta_k \quad (2.9)$$

$$C_k = 4S\cos\left(\frac{\sqrt{3}k_y}{2}\right)\left[J_1\cos\left(\frac{k_x}{2}\right) + J_2\cos\left(\frac{3k_x}{2}\right)\right] \quad (2.10)$$

$$D_k = 2SJ_z\cos\left(\frac{k_z}{2}\right)\delta_k^2 \quad (2.11)$$

$$\delta_k = e^{-ik_y/2\sqrt{3}} \quad (2.12)$$

To solve this we have to find another basis where this hamiltonian is diagonal. We can do so by using a matrix S transforming X into X', this matrix, explicited further, will give us the mean values of two bosonic operators and we can calculate the 1/S correction. At the first order, the dispersion relation is given by :

$$\left(\omega^\pm\right)^2 = A^2 + BB^* - C^2 - DD^* \pm \sqrt{4\left|AB - CD^*\right|^2 - \left|B^*D^* - BD\right|^2}$$

To plot the dispersion, we have changed the beginning of the plot, so that before plotting in the (M,M',Γ) plane, we first plot the dispersion along the z axis, from $\mathrm{N}\left(0, \frac{2\pi}{\sqrt{3}}, \pi\right)$ to $\mathrm{M}\left(0, \frac{2\pi}{\sqrt{3}}, 0\right)$, the plot is shown in Fig 2.4. The matrix

S turning the X basis into a basis X' where the Hamiltonian is diagonal is :

$$S = \begin{pmatrix} \bar{w}_1 & \bar{w}_2 & \bar{w}_3 & \bar{w}_4 \\ \bar{y}_1 & \bar{y}_2 & \bar{y}_3 & \bar{y}_4 \\ \bar{x}_1 & \bar{x}_2 & \bar{x}_3 & \bar{x}_4 \\ \bar{z}_1 & \bar{z}_2 & \bar{z}_3 & \bar{z}_4 \end{pmatrix}$$

$$\begin{aligned}
w_i &= -(A+\omega_i)\left(A^2+BB^*-C^2-DD^*-\omega_i^2\right)+2ABB^*-C\left(B^*D^*+BD\right) \\
y_i &= B^*\left((A+\omega_i)^2-BB^*+C^2\right)-2C\left(A+\omega_i\right)D+BD^2 \\
x_i &= C\left(A^2+BB^*-C^2+DD^*-\omega_i^2\right)-A\left(B^*D^*+BD\right)-\omega_i\left(B^*D^*-BD\right) \\
z_i &= D\left(A^2+C^2-DD^*-\omega_i^2\right)+B^{*2}D^*-2AB^*C \\
n_i &= |w_i w_i^* + y_i y_i^* - x_i x_i^* - z_i z_i^*| \\
\bar{x}_i &= \frac{x_i}{\sqrt{n_i}}
\end{aligned}$$

In these calculations, $\omega_1 = \omega^-$, $\omega_2 = \omega^+$, $\omega_3 = -\omega^-$ and $\omega_4 = -\omega^+$. The next step has been to check numerically the following relations between the mean fields. This is done by using the X' basis and using that in this basis, all the mean field are zero, so the remaining field is created by the commutation relation of the boson operators. We have :

$$m_1 = \left\langle a_k^\dagger a_k \right\rangle = \left\langle \tilde{a}_k^\dagger \tilde{a}_k \right\rangle = \left\langle b_{-k}^\dagger b_{-k} \right\rangle = \left\langle \tilde{b}_{-k}^\dagger \tilde{b}_{-k} \right\rangle = |\bar{w}_3|^2 + |\bar{w}_4|^2 \in \mathbf{R}$$

$$m_2 = \left\langle a_k^\dagger \tilde{a}_k \right\rangle = \left\langle \tilde{a}_k^\dagger a_k \right\rangle^* = \left\langle b_{-k}^\dagger \tilde{b}_{-k} \right\rangle = \left\langle \tilde{b}_{-k}^\dagger b_{-k} \right\rangle^* = \bar{w}_3^* \bar{y}_3 + \bar{w}_4^* \bar{y}_4 \in \mathbf{C}$$

$$m_3 = \left\langle a_k^\dagger b_{-k}^\dagger \right\rangle = \langle a_k b_{-k} \rangle = \left\langle \tilde{a}_k^\dagger \tilde{b}_{-k}^\dagger \right\rangle = \left\langle \tilde{a}_k \tilde{b}_{-k} \right\rangle = \bar{w}_1^* \bar{x}_1 + \bar{w}_2^* \bar{x}_2 \in \mathbf{R}$$

$$m_4 = \left\langle a_k^\dagger \tilde{b}_{-k}^\dagger \right\rangle = \langle \tilde{a}_k b_{-k} \rangle = \left\langle \tilde{a}_k^\dagger b_{-k}^\dagger \right\rangle^* = \left\langle a_k \tilde{b}_{-k} \right\rangle^* = \bar{y}_1^* \bar{x}_1 + \bar{y}_2^* \bar{x}_2 \in \mathbf{C}$$

We can then write the 1/S corrections to the terms of the matrix :

$$\begin{aligned}
\delta A_k &= \frac{4}{N}\sum_{k'} m_1(k')\left(F(k,k')+2J_z\right) \\
&- \left(m_2(k')m_2^*(k')\right)\frac{B_{k'}+B_{k'}^*}{2S} - \frac{m_4^*(k')D_{k'}^* + m_4(k')D_{k'}}{S} \\
F(k,k') &= 2J_1\left(1-\cos k_x\right)\left(1-\cos k_x'\right) + 2J_2\left(1-\cos\sqrt{3}k_y\right)\left(1-\cos\sqrt{3}k_y'\right) \\
&- 4\left(J_1+J_2\right) - 4D \\
\delta B_k &= \frac{J_z}{2}\frac{4}{N}\sum_{k'} m_2(k')\frac{B_{k-k'}+B_{k-k'}^*}{J_z S} - 2m_1(k')\frac{B_k+B_k^*}{J_z S}
\end{aligned}$$

$$
\begin{aligned}
\delta C_k &= 2J_1 \frac{4}{N} \sum_{k'} m_1(k') \frac{C_k}{2J_1 S} + m_3(k') G(k,k') \\
G(k,k') &= \cos\frac{\sqrt{3}k_y + k_x}{2} \cos\frac{\sqrt{3}k'_y + k'_x}{2} + \cos\frac{\sqrt{3}k_y - k_x}{2} \cos\frac{\sqrt{3}k'_y - k'_x}{2} \\
&\quad + \frac{J_2}{J_1} \left( \cos\frac{\sqrt{3}k_y + 3k_x}{2} \cos\frac{\sqrt{3}k'_y + 3k'_x}{2} \right. \\
&\quad \left. + \cos\frac{\sqrt{3}k_y - 3k_x}{2} \cos\frac{\sqrt{3}k'_y - 3k'_x}{2} \right) \\
\delta D_k &= \frac{J_z}{2} \frac{4}{N} \sum_{k'} m_4^*(k') \frac{D_{k-k'} + D^*_{k-k'}}{J_z S} - 4 m_1(k') \frac{D_k}{J_z S}
\end{aligned}
$$

We can finally plot the dispersion with all these parameters, the plot is shown in Fig. 2.4. We are interested to see how evolves the dispersion around its minimal

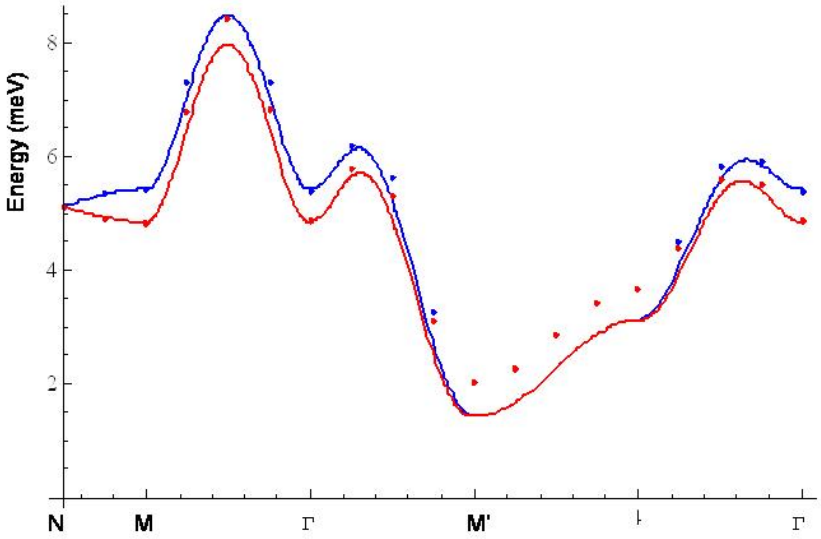



Figure 2.4: Plot of the dispersion with interlayer coupling and 1/S correction. Red is the $\omega^-$, blue the $\omega^+$, the solid lines show the LSW dispersion, the dots are the corrected terms.

value to compare with experimental data. The plot shows no dispersion around M' so we also plot the dispersion around this point along the z-direction, from M' to $N' = M' + 2\pi\vec{z}$ to see how the minimum of the energy evolves around this point (cf Fig 2.5). The Fig. 2.5 does not show any significative change around this point, the cause being that this change in the dispersion is in order of magnitude $(J_z/J_1)^2$.

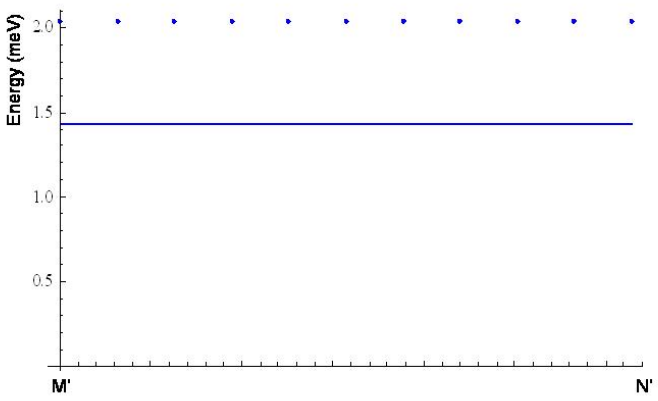



Figure 2.5: Profile of the dispersion $\omega^+$ around M' and on the z-axis. Solid line for the LSW model, dots for the 1/S correction

# Conclusion

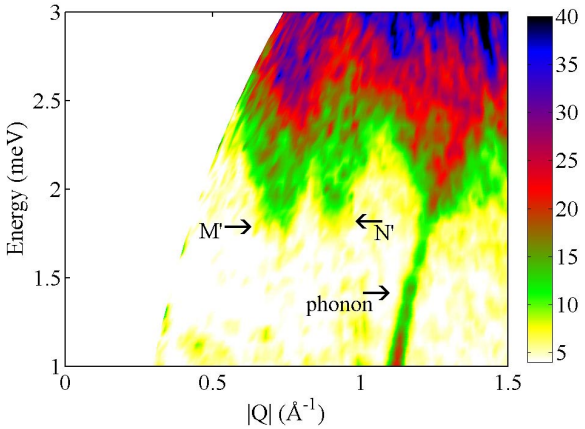



Figure 2.6: Experimental data of the energy of the scattered neutron in fonction of the modulus of the change in the momentum obtained by the Correlated Electron Systems Group.

The aim of this internship was to develop a model that could explain the experimental data Fig. 2.6. The M' arrow show the minimum of energy, the modulus corresponding to points as M'. The second minimum is obtained for N' since a larger distance in the real space means a smaller one in the reciprocal space, then N' appears before any other significative point. The D parameter is given by the minimum of energy (the phonon branch is not considered). The fact that the profile is not flat from M' to N' would indicate that the parameter $J_z$ should be larger. However, a larger $J_z$ leads to changes in the profile of the dispersion, shifting the minimum away from M'. The next work to do is fitting the experimental with this model to see if it is good enough to describe the $AgNiO_2$ crystal by playing with the value of the parameters, or if this model should be upgraded to a model also considering the itinerant electrons of the $Ni$ ions from the honeycomb lattice.

In this report, we have introduced a quantum spin wave theory in order to explain as simply as possible the spin excitations of a crystal. This consisted firstly in repeating calculations previously carried out by A. Chubukov and T. Jolicoeur in [2]. This gave a precise comprehension of the theory and allowed us to upgrade this approach to take into account both easy-axis anisotropy and interlayer coupling, which was performed by the end of the internship. However the adaptation of this model to experimental data arises new questions which should be answered within few weeks.

I acknowledge useful discussion with N. Shannon, R. Coldea, L. Seabra and A. Chubukov.

# Bibliography

[1] B.Schmidt, P. Thalmier, and N. Shannon. Magnetocaloric effect in the frustrated square lattice $J_1 - J_2$ model. *Physical review B*, 76, 2007.

[2] A. Chubukov and T. Jolicoeur. Order-from-disorder phenomena in Heisenberg antiferromagnets on a triangular lattice. *Physical review B*, 46, 1992.

[3] Fetter and Walecka. *Quantum Theory of Many-Particle Systems.* McGraw-Hill, 1971.

[4] Junishi Igarashi. 1/S expansion for thermodynamic quantities in a two-dimensional Heisenberg antiferromagnet at zero temperature. *Physical review B*, 46(17), 1992.

[5] C. Kittel. *Introduction to Solid State Physics.* Wiley.

[6] N. Shannon, B. Schmidt, K. Penc, and P. Thalmeier. Finite temperature properties and frustrated ferromagnetism in a square lattice Heisenberg model. *European Physical Journal B*, 2004.

[7] E.F. Shender and P.C.W. Holdsworth. Order by Didsorder and Topology in Frustrated Magnetic Systems. In *Fluctuations and Order.* Springer.

[8] E. Wawrzinska, R. Coldea, E.M. Wheeler, I.I. Mazin, M.D. Johannes, T. Sörgel, M. Jansen adn R.M. Ibberson, and P.G. Radaelli. Orbital degeneracy removed by charge order in triangular antiferromagnet $AgNiO_2$. *Physical Review Letters*, 2008.

## Software

*Mathematica 6.0*, Wolfram research.

# Appendix A

# Canonical transformation and application to CAF phase

This part will show how the dispersion is calculated in the case of antiferromagnetism and simplified by the $\omega^2 = A^2 - B^2$ formula. We will focus on the square (or cubic) lattice case and apply the same method to other lattices; consequently we start the calculations with :

$$\mathcal{H} = -\frac{NzJ}{2}S^2 + \underbrace{zJS\sum_{\vec{q}}\left[a^\dagger_{\vec{q}}a_{\vec{q}} + b^\dagger_{-\vec{q}}b_{-\vec{q}} + \gamma_{\vec{q}}\left(a_{\vec{q}}b_{-\vec{q}} + a^\dagger_{\vec{q}}b^\dagger_{-\vec{q}}\right)\right]}_{H_{LSW}}$$

We can introduce a change in the bosonic operators by introducing $\alpha$ and $\beta$ operators given by the equations [2]:

$$\begin{aligned}
a_{\vec{q}} &= u_q\alpha_{\vec{q}} - v_q\beta^\dagger_{-\vec{q}} \\
a^\dagger_{\vec{q}} &= u_q\alpha^\dagger_{\vec{q}} - v_q\beta_{-\vec{q}} \\
b_{\vec{q}} &= u_q\beta_{\vec{q}} - v_q\alpha^\dagger_{-\vec{q}} \\
b^\dagger_{\vec{q}} &= u_q\beta^\dagger_{\vec{q}} - v_q\alpha_{-\vec{q}}
\end{aligned}$$

$$\begin{aligned}
\text{with conditions} \quad \left[\alpha_i, \alpha^\dagger_j\right] &= \delta_{ij} \\
\left[\beta_i, \beta^\dagger_j\right] &= \delta_{ij} \\
u_q^2 - v_q^2 &= 1
\end{aligned}$$

This change of the bosonic operators leads to a writing of $H_{LSW}$ :

$$\begin{aligned}
H_{LSW} &= zJS\sum_{\vec{q}}\left[2\left(v_q^2 - \gamma_{\vec{q}}u_qv_q\right)\right. \\
&+ \left(u_q^2 + v_q^2 - 2\gamma_{\vec{q}}u_qv_q\right)\left(\alpha^\dagger_{\vec{q}}\alpha_{\vec{q}} + \beta^\dagger_{-\vec{q}}\beta_{-\vec{q}}\right) \\
&+ \left.\left(\gamma_{\vec{q}}\left(u_q^2 + v_q^2\right) - 2u_qv_q\right)\left(\alpha_{\vec{q}}\beta_{-\vec{q}} + \alpha^\dagger_{\vec{q}}\beta^\dagger_{-\vec{q}}\right)\right]
\end{aligned}$$

We want the last term to vanish, this gives a condition upon $u_q = \cosh\theta$ and $v_q = \sinh\theta$. We have :

$$\begin{aligned}
\tanh 2\theta &= \frac{B_q}{A_q} = \gamma_{\vec{q}} \\
u_q^2 &= \frac{1}{2}\left(1 + \frac{1}{\sqrt{1-\gamma_{\vec{q}}^2}}\right) \\
v_q^2 &= \frac{1}{2}\left(\frac{1}{\sqrt{1-\gamma_{\vec{q}}^2}} - 1\right) \\
H_{LSW} &= zJS\sum_{\vec{q}}\sqrt{1-\gamma_{\vec{q}}^2} - 1 + \sum_{\vec{q}}\underbrace{zJS\sqrt{1-\gamma_{\vec{q}}^2}}_{\omega(\vec{q})}\left(\alpha_{\vec{q}}^\dagger\alpha_{\vec{q}} + \beta_{-\vec{q}}^\dagger\beta_{-\vec{q}}\right)
\end{aligned}$$

We hence see that the formula $\omega^2 = A^2 - B^2$ is valid in this case, and since we have performed a very general change of the coordinates, this result does not depend on the values of A and B (except of course if $A^2 < B^2$).

## A.1 Adding magnetic field

In the section 2.1, one of the option to lift the continous degeneracy of the rotation of the spins is to add a magnetic field h, this leads to a change :

$$\omega \rightsquigarrow \omega^\pm = \omega \pm h$$

The calculations give :

$$\begin{aligned}
\mathcal{H}' &= \mathcal{H} - \underbrace{h\sum_i S_i^z}_{\text{magnetic field}} \\
&= E_0 + \sum_k \left(a_k^\dagger, b_k\right)\begin{pmatrix} A_k - h & B_k \\ B_k & A_k + h \end{pmatrix}\begin{pmatrix} a_k \\ b_k^\dagger \end{pmatrix}
\end{aligned}$$

We use the canonical transformation :

$$\begin{aligned}
a_{\vec{q}} &= u_q\alpha_{\vec{q}} - v_q\beta_{-\vec{q}}^\dagger \\
a_{\vec{q}}^\dagger &= u_q\alpha_{\vec{q}}^\dagger - v_q\beta_{-\vec{q}} \\
b_{\vec{q}} &= u_q\beta_{\vec{q}} - v_q\alpha_{-\vec{q}}^\dagger \\
b_{\vec{q}}^\dagger &= u_q\beta_{\vec{q}}^\dagger - v_q\alpha_{-\vec{q}} \\
u_q^2 &= \frac{1}{2}\left(1 + \frac{A_q}{\sqrt{A_q^2 - B_q^2}}\right) \\
v_q^2 &= \frac{1}{2}\left(\frac{A_q}{\sqrt{A_q^2 - B_q^2}} - 1\right)
\end{aligned}$$

The calculations then give :

$$\mathcal{H}' = E_0 + \sum_q (\alpha_q^\dagger, \beta_{-q}) \begin{pmatrix} \sqrt{A_q^2 - B_q^2} + h & 0 \\ 0 & \sqrt{A_q^2 - B_q^2} - h \end{pmatrix} \begin{pmatrix} \alpha_q \\ \beta_{-q}^\dagger \end{pmatrix}$$

## A.2 Colinear antiferromagnet on a square lattice

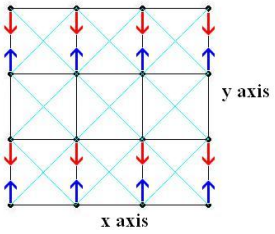



Figure A.1: Colinear antiferromagnetic lattice

This state is observed only with the $J_1 - J_2$ model and for certain values of the ratio of the two coupling contants, more precisely we should have :

$$J_2 > \frac{|J_1|}{2}$$

If this condition is realised we can write our Hamiltonian as in the previous section, introducing A and B. We obtain :

$$\begin{aligned} A &= 2S\left(2J_2 + J_1 \cos q_x\right) \\ B &= 2S \cos q_y \left(J_1 + J_2 \cos q_x\right) \\ \left(\frac{\omega_{\vec{q}}}{4S}\right)^2 &= J_2^2\left(1 - \cos^2 q_x \cos^2 q_y\right) + J_1 J_2 \cos q_x \left(1 - \cos^2 q_y\right) + \ldots \\ &+ \frac{J_1^2}{4}\left(\cos^2 q_x - \cos^2 q_y\right) \end{aligned}$$

# Appendix B

# Leading quantum corrections to the spin wave dispersion of a square lattice NAF - Comparison of methods

### First method

Expanding the calculation one degree further will show that the Hamiltonian is :

$$\mathcal{H} = E_0 + H_{LSW} - \underbrace{J\sum_{\langle ij\rangle_1} a_i^\dagger b_j^\dagger b_j a_i + \frac{1}{4}\left(a_i^\dagger a_i a_i b_j + b_j^\dagger b_j b_j a_i + h.c.\right)}_{H_1} + \dots$$

$$\begin{aligned} H_1 &= \frac{2zJ}{N}{\sum_{1234}}' \Big( a_1^\dagger b_2^\dagger b_3 a_4 \gamma_{3-2} + \\ &+ \frac{1}{4}\left[a_1^\dagger a_{-2} a_3 b_4 \gamma_4 + a_{-1} b_2^\dagger b_3 b_4 \gamma_{-2+3+4} + a_1^\dagger a_2^\dagger a_3 b_{-4}^\dagger \gamma_{-4} + a_1^\dagger b_2^\dagger b_{-3}^\dagger b_4 \gamma_{2+3-4}\right]\Big) \end{aligned}$$

We have used the abbreviations $a_1 = a_{k_1}$ and $\gamma_{3-2} = \gamma(k_3 - k_2)$ and the prime means that the sum runs on every quadruplet of vectors of reciprocal space with respect to the condition $k_1 + k_2 = k_3 + k_4 + G$ where G is a reciprocal lattice vector. We now have to perform the same canonical transformation we have made for the linear spin wave term to put the Hamiltonian in term of $\alpha^\dagger\alpha$ and $\beta^\dagger\beta$. We will obtain 16 terms for each four-bosons term, terms we have to write under the normal ordering form $\alpha^\dagger\beta^\dagger\beta\alpha$. This writing will lead to :

$$
\begin{aligned}
H_1 &= \sum_{k,k'} \left(a^\dagger_{\vec{k}}, b_{\vec{-k}}\right) \begin{pmatrix} A_{kk'} & B_{kk'} \\ B_{kk'} & A_{kk'} \end{pmatrix} \begin{pmatrix} a_{\vec{k}} \\ b^\dagger_{-\vec{k}} \end{pmatrix} \\
&+ \frac{1}{4} \sum_{1234}{}' \; u_1 u_2 u_3 u_4 \Big[ \alpha^\dagger_1 \alpha^\dagger_2 \alpha_3 \alpha_4 \; B^a + \beta^\dagger_{-3} \beta^\dagger_{-4} \beta_{-1} \beta_{-2} \; B^b + 4 \alpha^\dagger_1 \beta^\dagger_{-4} \beta_{-2} \alpha_3 \; B^c \\
&\qquad + 2 \left( 2 \alpha^\dagger_1 \beta_{-2} \alpha_3 \alpha_4 \; B^d + 2 \beta^\dagger_{-4} \beta_{-1} \beta_{-2} \alpha_3 \; B^e + \alpha^\dagger_1 \alpha^\dagger_2 \beta^\dagger_{-3} \beta^\dagger_{-4} \; B^f + h.c. \right) \Big]
\end{aligned}
$$

Where :

$$
\begin{aligned}
\sum_{k'} A_{kk'} &= \sum_{k'} (1 - \epsilon_{k'}) \, \epsilon_k \\
\sum_{k'} B_{kk'} &= 0 \\
B^a &= \gamma_{1-4} x_1 x_4 + \gamma_{1-3} x_1 x_3 + \gamma_{2-4} x_2 x_4 + \gamma_{2-3} x_2 x_3 \\
&\quad - \frac{1}{2} \left( \gamma_1 x_1 + \gamma_2 x_2 + \gamma_3 x_3 + \gamma_4 x_4 + \gamma_{2-3-4} x_2 x_3 x_4 + \right. \\
&\qquad \left. + \gamma_{1-3-4} x_1 x_3 x_4 + \gamma_{4-2-1} x_1 x_2 x_4 + \gamma_{3-2-1} x_1 x_2 x_3 \right) \\
B^b &= \gamma_{2-4} x_1 x_3 + \gamma_{1-4} x_2 x_3 + \gamma_{1-3} x_2 x_4 + \gamma_{2-3} x_1 x_4 \\
&\quad - \frac{1}{2} \left( \gamma_2 x_1 x_3 x_4 + \gamma_1 x_2 x_3 x_4 + \gamma_3 x_1 x_2 x_4 + \gamma_4 x_1 x_2 x_3 + \right. \\
&\qquad \left. + \gamma_{2-3-4} x_1 + \gamma_{1-3-4} x_2 + \gamma_{4-1-2} x_3 + \gamma_{3-1-2} x_4 \right) \\
B^c &= \gamma_{2-4} + \gamma_{1-3} x_1 x_2 x_3 x_4 + \gamma_{1-4} x_1 x_2 + \gamma_{2-3} x_3 x_4 \\
&\quad - \frac{1}{2} \left( \gamma_2 x_4 + \gamma_1 x_1 x_2 x_4 + \gamma_{2-3-4} x_3 + \gamma_{1-3-4} x_1 x_2 x_3 + \right. \\
&\qquad \left. + \gamma_4 x_2 + \gamma_3 x_2 x_3 x_4 + \gamma_{4-1-2} x_1 + \gamma_{3-2-1} x_1 x_3 x_4 \right)
\end{aligned}
$$

$$
\begin{aligned}
B^d &= -\gamma_{2-4}x_4 - \gamma_{1-4}x_1x_2x_4 - \gamma_{2-3}x_3 - \gamma_{1-3}x_1x_2x_3 \\
&\quad +\frac{1}{2}\left(\gamma_2 + \gamma_1x_1x_2 + \gamma_3x_2x_3 + \gamma_4x_2x_4 + \gamma_{2-3-4}x_3x_4\right. \\
&\quad \left. +\gamma_{1-3-4}x_1x_2x_3x_4 + \gamma_{3-2-1}x_1x_3 + \gamma_{4-2-1}x_1x_4\right) \\
B^e &= -\gamma_{2-4}x_1 - \gamma_{2-3}x_1x_3x_4 - \gamma_{1-4}x_2 - \gamma_{1-3}x_2x_3x_4 \\
&\quad +\frac{1}{2}\left(\gamma_2x_1x_4 + \gamma_1x_2x_4 + \gamma_4x_1x_2 + \gamma_3x_1x_2x_3x_4\right. \\
&\quad \left. +\gamma_{2-3-4}x_1x_3 + \gamma_{1-3-4}x_2x_3 + \gamma_{4-2-1} + \gamma_{3-2-1}x_3x_4\right) \\
B^f &= \gamma_{2-4}x_2x_3 + \gamma_{2-3}x_2x_4 + \gamma_{1-3}x_1x_4 + \gamma_{1-4}x_1x_3 \\
&\quad -\frac{1}{2}\left(\gamma_2x_2x_3x_4 + \gamma_{3-2-1}x_1x_2x_4 + \gamma_4x_3 + \gamma_1x_1x_3x_4\right. \\
&\quad \left. +\gamma_{2-3-4}x_2 + \gamma_{1-3-4}x_1 + \gamma_{4-1-2}x_1x_2x_3 + \gamma_3x_4\right)
\end{aligned}
$$

We have noted $x_k = \frac{v_k}{u_k}$, $\epsilon_k = \frac{\omega_k}{zJS}$ and symmetrized all the expressions in function of identical bosons. The sum over A and B are simplified using the relations between $u_k$, $v_k$ and $\gamma_k$. The conclusion of these calculations is that we now have :

$$
\begin{aligned}
\mathcal{H} &= \overbrace{JSz\sum_k(\epsilon_k - 1) \; + \; \sum_k \omega_k\left(\alpha_k^\dagger\alpha_k + \beta_k^\dagger\beta_k\right)}^{E_0+LSW} \\
&+ \quad \frac{1}{2S}\underbrace{\frac{2}{N}\sum_k(1-\epsilon_k)}_{A\simeq 0.158}\sum_k \omega_k\left(\alpha_k^\dagger\alpha_k + \beta_k^\dagger\beta_k\right) \\
&+ \quad \text{four-bosons term} + \ldots
\end{aligned}
$$

## Second method

What we have writen for the Hamiltonian in term of $a_1^\dagger, b_2^\dagger, a_3$ and $b_4$ is still true. Now the aim is to use a mean-field value of two bosonic operators to contract it so as to have two-bosons terms only.

### Mean values of two bosonic operators

In the case of ferromagnet one technique is to add a magnetic field term to the Hamiltonian, calculate the partition function then the free energy and finally the magnetization per site. This gives :

$$
\begin{aligned}
\mathcal{H} &= J\sum_{\langle ij\rangle_1}\vec{S_i}\cdot\vec{S_j}+h\sum_i S_i^z \\
&= -\frac{zN}{2}JS^2-NhS+\sum_q\left[\underbrace{JSz\left(1-\gamma_q\right)}_{\omega_q}+h\right]a_q^\dagger a_q \\
Z &= Tr\left(e^{\mathcal{H}/k_bT}\right)=e^{\frac{N\left(\frac{z}{2}JS+h\right)S}{k_BT}}\prod_q\underbrace{\sum_n e^{-\frac{n(\omega_q+h)}{k_BT}}}_{=\left(1-e^{(\omega_q+h)/k_BT}\right)^{-1}} \\
F &= -k_BT\log Z=-\frac{NzJS^2}{2}-NhS+k_BT\sum_q\log\left(1-e^{-(\omega_q+h)/k_BT}\right) \\
m &= -\frac{1}{N}\left.\frac{\partial F}{\partial h}\right|_{h=0}=S-\frac{1}{N}\sum_q n_B\left(\omega_q\right)\quad\text{where}\quad n_B\left(x\right)=\frac{1}{e^{x/k_BT}-1} \\
m &= S-\left\langle a_q^\dagger a_q\right\rangle
\end{aligned}
$$

$$
\text{Conclusion :}\qquad \left\langle a_q^\dagger a_{q'}\right\rangle=\delta_{qq'}n_B\left(\omega_q\right)\xrightarrow[T\to 0]{}0
$$

We can use a similar technique to calculate it for the antiferromagnet. The fact that the spins point in opposite direction is solved by considering an hypothetic field pointing upwards for A sites, downwards for B sites. The other difference is that the Hamiltonian is now diagonal in $\alpha$ and $\beta$ instead of $a$ and $b$ so the mean values are simplest for $\alpha$ and $\beta$ (in fact the same calculations can be reproduced so we have the same value, the bosonic occupation number, as expected). Consequently, we have :

$$
\begin{aligned}
\left\langle\alpha_q^\dagger\alpha_{q'}\right\rangle &= \left\langle\beta_q^\dagger\beta_{q'}\right\rangle = \delta_{qq'}n_B\left(\omega_q\right) \\
\left\langle\beta_q^\dagger\alpha_{q'}\right\rangle &= \left\langle\alpha_q^\dagger\beta_{q'}\right\rangle = 0
\end{aligned}
$$

$$
\begin{aligned}
\left\langle a_q^\dagger a_{q'}\right\rangle &= \left\langle b_q^\dagger b_{q'}\right\rangle = \delta_{qq'}\left(\left(u_q^2+v_q^2\right)n_B\left(\omega_q\right)+v_q^2\right)\overset{T\to 0}{=}\delta_{qq'}\frac{A_q-\omega_q}{2\omega_q} \\
\left\langle a_q^\dagger b_{-q'}^\dagger\right\rangle &= \left\langle a_q b_{-q'}\right\rangle = -\delta_{qq'}u_qv_q\left(2n_B\left(\omega_q\right)+1\right)\overset{T\to 0}{=}-\delta_{qq'}\frac{B_q}{2\omega_q}
\end{aligned}
$$

We now use these values of the means in the four-bosons terms of the Hamiltonian. For example :

$$
\begin{aligned}
\sum_{1234}a_1^\dagger b_2^\dagger b_3a_4\ \gamma_{3-2}\ \delta_{1+2,3+4} &\simeq \sum_{qq'}\left\langle b_{q'}^\dagger b_{q'}\right\rangle a_q^\dagger a_q+\left\langle a_{q'}^\dagger a_{q'}\right\rangle b_q^\dagger b_q \\
&+ \left\langle a_{q'}^\dagger b_{-q'}^\dagger\right\rangle b_{-q}a_q\gamma_{q-q'}+\left\langle a_qb_{-q'}\right\rangle a_q^\dagger b_{-q}^\dagger\gamma_{q-q'}
\end{aligned}
$$

Doing the same for all the four-bosons terms, and for zero temperature leads to :

$$\mathcal{H} \;=\; E_O \;+\; H_{LSW} \quad - \quad \frac{2zJ}{N}\sum_{q,q'}\left(a^{\dagger}_{\vec{q}}, b_{\vec{-q}}\right)\begin{pmatrix} A_{qq'} & B_{qq'} \\ B_{qq'} & A_{qq'}\end{pmatrix}\begin{pmatrix} a_{\vec{q}} \\ b^{\dagger}_{-\vec{q}}\end{pmatrix}$$

$$\text{where} \quad \sum_{q'} A_{qq'} \;=\; \sum_{q'} v^2_{q'} - u_{q'}v_{q'}\gamma_{q'} \;=\; \frac{-NA}{4}$$

$$\sum_{q'} B_{qq'} \;=\; \sum_{q'} \gamma_q v^2_{q'} - \gamma_{q-q'}u_{q'}v_{q'} \;=\; \frac{-NA}{4}\,\gamma_q$$

$$A \;=\; \frac{A}{N}\sum k\,(1-\epsilon_k)$$

We can now conclude and check that the two methods give the same result :

$$\mathcal{H} = E_0 \;+\; H_{LSW} \;+\; \frac{JSz}{2S}\,A\sum_{q}\left(a^{\dagger}_{\vec{q}}, b_{\vec{-q}}\right)\begin{pmatrix} 1 & \gamma_q \\ \gamma_q & 1\end{pmatrix}\begin{pmatrix} a_{\vec{q}} \\ b^{\dagger}_{-\vec{q}}\end{pmatrix}$$