university of groningen / faculty of science and engineering

Spin-Isospin Model of a Triangular Antiferromagnet

Author:
Paul CASAMIAN
s4923294

Supervisor:
prof. dr. Maxim MOSTOVOY
Second examiner :
PhD Jagoda SLAWINSKA

Bachelor's Thesis
To fulfill the requirements for the degree of
Bachelor of Science in Physics
at the University of Groningen

July 14, 2024

Abstract

Spin $S = \frac{1}{2}$ triangular antiferromagnets are intriguing models that challenge our understanding of magnetic ordering. The variety of quantum phenomena they showcase, caused by their inherently frustrated geometry and quantum fluctuations, are of great interest due to their application in quantum computing, among others. Here, we discuss the triangular antiferromagnetic material $Ba_3TaFe_3Si_2O_{14}$, in which we replace the spin $s = \frac{5}{2}$ iron ions, by spin $s = \frac{1}{2}$ ions. This change results in the appearance of a new quantum degree of freedom, isospin. In the triangle chain, a one-dimensional model of triangles stacked along the c-axis, spins are in a disordered spin liquid state, whereas isospins show a short ranged helical spiral ordering. We then extend this model to three-dimensions by adding in-plane Heisenberg exchange interactions. It results in average spins to rotate along the c-axis, together with isospins. However, this rotation occurs in an arbitrary plane. The Dzyaloshinskii-Moriya interaction is responsible for the spins and isospins rotation axes to align, either parallel or anti-parallel. This thesis opens the door for investigations that go beyond the mean field approximation and account for quantum spin and isospin fluctuations in the ground state.

Contents

1 Introduction

Anti-ferromagnetism is the property of materials that favours anti-parallel alignment of neighbouring spins due to the Heisenberg exchange interaction. The order parameter of an antiferromagnet is the staggered magnetisation and it is defined as "the difference of the magnetisation on each sublattice" [1]. In an antiferromagnet, the critical temperature, called the Néel temperature, occurs when the thermal agitation overcomes the interaction effect and the spins of the magnetic ions become disordered [2]. Near this temperature, the staggered magnetisation is proportional to $(T_c - T)^\beta$, where T_c is the Néel temperature of the material and β is a critical exponent whose value depends on the material [3]. As the material reaches the critical temperature, the staggered magnetisation becomes zero and the ferromagnet turns into a paramagnet [2]. During this phase transition, an anomalous sharp increase in the heat capacity is observed, due to the long range magnetic ordering of the ions [4].
A similar behaviour is observed in ferromagnets, where the critical temperature is named the Curie temperature and the order parameter is the magnetisation [5]. However, in contrast to ferromagnetic materials, the ordered magnetic moment in antifferomagnets cancel each other, resulting in zero net magnetisation.

Antiferromagnetic spintronics has been an area of very active recent research due to interesting properties of antiferromagnets, such as high speed of magnons and domain walls and the absence of stray fields [6][7]. Among them, the langasite family is a group of crystal displaying unique non-linear optical properties [8][9]. Langasites containing magnetic iron ions show a complex anti-ferromagnetic ordering [10], as well as an electrical polarisation in the ordered state [11][12]. In particular, $Ba_3TaFe_3Si_2O_{14}$ (referred to as Fe-langasite) is a non-collinear antiferromagnet with a triangle based crystal lattice formed by Fe^{3+} ions with spin $s = \frac{5}{2}$. The triangles form a triangular lattice in the ab-plane and the antiferromagnetic interaction results in the 120° angle between spins in the triangles. These triangles are stacked on top of each other along the c-axis, and the spins form a helical structure, with a 51.4 deg rotation between spins in neighbouring layers [13]. The Néel temperature for this material is $T_N = 27.4$K [13]. It was predicted to exhibit unconventional magnetic topological defects, such as coreless vortex tubes and three-dimensional Skyrmions [14].

Another field of study worth noting is the two dimensional spin $\frac{1}{2}$ antiferromagnetic Heisenberg model, which was a focus of theoretical research due to, amongst other things, its use for high T_c superconductors [15] [16]. Multiples investigations have looked at the ground state and magnetic ordering of quantum antiferromagnets with lattices of various shapes [17][18]. Amongst these, square lattices have been studied and understood extensively, whereas triangular lattices still hold many uncertainties, especially regarding their spin ordering in two-dimensions and higher. Theoretical studies of the latter have suggested the existence of quantum spin liquids in this lattice [19]. This property could have applications in the growing field of topological quantum computation, as well as superconductors [20].
Another common area of research is the study of staggered magnetisation, the order parameter of (quantum) antiferromagnets [15][21][22]. This topic has applications in data storage, since the orientation of the order parameter can be used to store information in a dense and efficient way [23]. So, quantum antiferromagnets are theoretical materials that possess many interesting properties and their study helps us improve our understanding of all antiferromagnets.

This thesis lies in both of these fields, since it studies an antiferromagnetic material that possess the triangular structure of Fe-langasite, but in which the iron ions with spin $s = \frac{5}{2}$ have been replaced by spin $s = \frac{1}{2}$ ions. Since the small spins cannot be treated classically, a quantum treatment is necessary when investigating the properties of this theoretical model. The ground state of such material is explored by means of two different methods. First, to be able to describe all states of a triangle in the lattice, we need to introduce a new quantum degree of freedom, isospin. Then, we discuss a one dimensional model of the triangles stacked along the c-axis, which is referred to as a triangle spin chain. Using the Jordan-Wigner transformations, we find the rotation of isospins and disordered spins in this model.
The second model extends the previous one to three dimensions by including in-plane interactions. In this model, both spins and isospins order and rotate. However, while isospins rotate in the xy-plane, the spin rotation plane is arbitrary.
Using first order perturbation theory we discuss effects of internal excitations such as the Dzyaloshinskii-Moriya (DM) interaction and exchange anisotropy. The DM interaction forces result in the spin/isospin rotation axe to align parallel or anti-parallel, depending on the sign of the DM interaction.

2 Single Spin Triangle

Before looking at the triangle chain model, this section aims to introduce important concepts like the Heisenberg exchange interaction. Its purpose is also to present the tools that will be used in the later derivations. This is done by examining the simplest possible system: a triangle that does not interact with its neighbours.

2.1 Triangle Degeneracy

Consider a single triangle formed by three spins $s = \frac{1}{2}$ (see Fig.1). We first consider the so-called Heisenberg exchange interactions. In magnetic insulators, these exchange interactions result from the virtual hopping of electrons between magnetic ions, Coulomb repulsion and Fermi statistics of electrons. The spin Hamiltonian describing the Heisenberg exchange interactions is:

$$H_{ex} = \sum_{i,j} J_{i,j}\ \boldsymbol{S}_i \cdot \boldsymbol{S}_j\ , \tag{1}$$

where J is called the exchange constant, and $\boldsymbol{S}_i$ is the spin operator of the i-th ion. Since these interactions do not involve relativistic effects, the Hamiltonian (1) is invariant under rotation of all spins. Depending on the sign of J_{ij}, there are two different types of materials: those whose energy is the lowest for parallel spins are called ferromagnets, whereas in antiferromagnets neighbouring spins have opposite directions in the lowest-energy configuration.

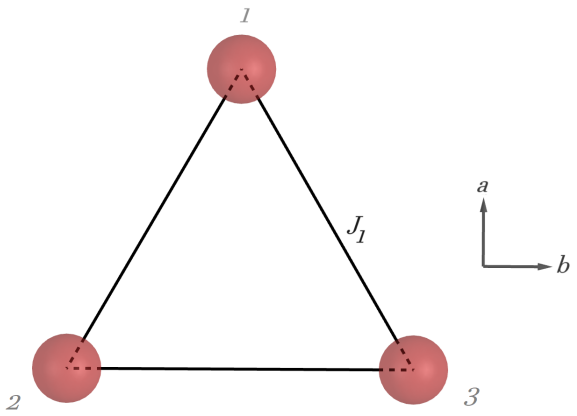

Figure 1: Triangle formed by three magnetic ions with spin $\frac{1}{2}$. $J_{12} = J_{13} = J_{23} = J_1$ due to the 120° rotational symmetry

The total spin of the triangle is $\boldsymbol{S} = \boldsymbol{S}_1 + \boldsymbol{S}_2 + \boldsymbol{S}_3$. Due to the rotational invariance of Heisenberg exchange interactions, the energy of the spin triangle only depends on the total spin S and is independent of the spin projection. There are in total eight states of the system of three spins $s = \frac{1}{2}$. Since there are four $S = \frac{3}{2}$ states ($S_z = \pm\frac{1}{2}, \pm\frac{3}{2}$) and two $S = \frac{1}{2}$ states ($S_z = \pm\frac{1}{2}$), there must be two different $S_z = \pm\frac{1}{2}$ states. Our attention now turns towards finding the spacing between the $S = \frac{3}{2}$ and $S = \frac{1}{2}$ energy levels by applying the Hamiltonian (1) to our system. The exchange constant of the interactions within the triangle is called J_1 and the Hamiltonian of the

triangle can be written as follows:

$$H_t = J_1[\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 + \boldsymbol{S}_2 \cdot \boldsymbol{S}_3 + \boldsymbol{S}_3 \cdot \boldsymbol{S}_1] = \frac{J_1}{2}\left[\boldsymbol{S}^2 - \boldsymbol{S}_1^2 - \boldsymbol{S}_2^2 - \boldsymbol{S}_3^2\right] \ , \tag{2}$$

where the relation $\boldsymbol{S}^2 = (\boldsymbol{S}_1+\boldsymbol{S}_2+\boldsymbol{S}_3)^2 = \boldsymbol{S}_1^2+\boldsymbol{S}_2^2+\boldsymbol{S}_3^2+2\left(\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 + \boldsymbol{S}_2 \cdot \boldsymbol{S}_3 + \boldsymbol{S}_3 \cdot \boldsymbol{S}_1\right)$ and the fact that $S(S+1)$ is the eigenvalue of $\hat{\boldsymbol{S}}$, were used to obtain the second inequality. Applying this operator to the states with total spin $S = \frac{3}{2}$ and $S = \frac{1}{2}$, the energy difference $\Delta E = E_{3/2} - E_{1/2}$ is:

$$\Delta E = \frac{J_1}{2}\left[\frac{3}{2}\cdot\frac{5}{2} - \frac{1}{2}\cdot\frac{3}{2}\right] = \frac{3}{2}J_1 \ .$$

In Fe-langasite, the interactions between the spins of the triangle are antiferromagnetic, corresponding to $J_1 > 0$ [24]. Therefore, the $S = \frac{1}{2}$ states have lower energy than the $S = \frac{3}{2}$ states. In what follows, only the manifold of $S = \frac{1}{2}$ states on each triangle is considered, assuming that J_1 is larger than all other interactions in the problem.

However, the degeneracy of the $S = \frac{1}{2}$ states raises an issue: $S = \frac{1}{2}$ implies that the quantum number of the z-component of spin can only take values: $\pm\frac{1}{2}$, which is not enough to describe all four states of the system. This necessitates the introduction of a new quantum number: isospin (or pseudo-spin) $T = \frac{1}{2}$, to label all states. Importantly, spin and isospin commute: $[\hat{S}_i, \hat{T}_j] = 0$. The four $S = \frac{1}{2}$ states are then described by the spin and isospin projections, $S_z = \pm\frac{1}{2}$ and $T_z = \pm\frac{1}{2}$. This new quantum degree of freedom is extremely important, as it is responsible for the differences between the classical model (Fe-langasite) and the quantum model discussed below.

2.2 The States of the System

Next, we construct the four degenerate $S = \frac{1}{2}$ states. They are partly specified by the z-component of the total spin and for $S_z = \frac{1}{2}$, they are a superposition of $|\downarrow\uparrow\uparrow\rangle$, $|\uparrow\downarrow\uparrow\rangle$ and $|\uparrow\uparrow\downarrow\rangle$, where $\uparrow = \left(\begin{smallmatrix}1\\0\end{smallmatrix}\right)$ denotes the $s_z = +\frac{1}{2}$ state and $\downarrow = \left(\begin{smallmatrix}0\\1\end{smallmatrix}\right)$ is the $s_z = -\frac{1}{2}$ state. To allow for all four states to be different, as explained above, they must depend on the new degree of freedom, isospin.
Since the equilateral spin triangle is invariant under $\frac{2\pi}{3}$ rotations around the z axis, the states should be eigenstates of the operator $D_z(\alpha) = \exp(-i\alpha\hat{\ell}_z)$, with $\alpha = \frac{2\pi}{3}$. Applying such an operator on any wavefunction $\psi(\varphi)$ results in $e^{-i\alpha\ell_z}|\varphi\rangle = |\varphi + \alpha\rangle$ and since in this case $\ell = -1, 0, 1$, the wavefunction should obey $\left|\varphi + \frac{2\pi}{3}\right\rangle = \lambda|\varphi\rangle$, where $\lambda = 1$, ω or $\bar{\omega}$; $\omega = \exp(i\frac{2\pi}{3})$. Taking all these properties into account, we construct the state $|S_z, T_z\rangle$:

$$\left|\, \frac{1}{2}\,,\, \frac{\tau}{2} \,\right\rangle = \frac{1}{\sqrt{3}}\left(|\downarrow\uparrow\uparrow\rangle + \bar{\omega}^\tau\,|\uparrow\downarrow\uparrow\rangle + \omega^\tau\,|\uparrow\uparrow\downarrow\rangle\right)\,, \tag{3}$$

where $t_z = \frac{\tau}{2}$ is the z-component of isospin. The other two $S = \frac{1}{2}$ states have z-component spin $S_z = -\frac{1}{2}$ and can be obtained by rotating the states given by equation (3) by π around the spin y-axis:

$$\left|\, -\frac{1}{2}\,,\, \frac{\tau}{2} \,\right\rangle = R_y(\pi)\left|\, \frac{1}{2}\,,\, \frac{\tau}{2} \,\right\rangle = \frac{1}{\sqrt{3}}\left(R_y(\pi)\,|\downarrow\uparrow\uparrow\rangle + \bar{\omega}^\tau R_y(\pi)\,|\uparrow\downarrow\uparrow\rangle + \omega^\tau R_y(\pi)\,|\uparrow\uparrow\downarrow\rangle\right)$$

$R_{\boldsymbol{n}}(\varphi)$ is the generator of the SU(2) group and is defined as:

$$R_{\boldsymbol{n}} = e^{i\varphi(\boldsymbol{n}\cdot\boldsymbol{S})} = e^{i\frac{\varphi}{2}(\boldsymbol{n}\cdot\boldsymbol{\sigma})} = \cos\tfrac{\varphi}{2}\mathbb{1} - i\sin\tfrac{\varphi}{2}\hat{\boldsymbol{n}}\cdot\boldsymbol{\sigma}\ .$$

For $\varphi = \pi$, $R_y = -i\sigma_y = \left(\begin{smallmatrix}0 & -1\\ 1 & 0\end{smallmatrix}\right)$, which when applied to $|\uparrow\rangle$ and $|\downarrow\rangle$ gives:

$$\left(\begin{smallmatrix}0 & -1\\ 1 & 0\end{smallmatrix}\right)\left(\begin{smallmatrix}1\\0\end{smallmatrix}\right) = \left(\begin{smallmatrix}0\\1\end{smallmatrix}\right) = \downarrow \quad \text{and} \quad \left(\begin{smallmatrix}0 & -1\\ 1 & 0\end{smallmatrix}\right)\left(\begin{smallmatrix}0\\1\end{smallmatrix}\right) = -\left(\begin{smallmatrix}1\\0\end{smallmatrix}\right) = -\uparrow\ .$$

So, the states with $S_z = -\frac{1}{2}$ read:

$$\left|\, -\frac{1}{2}\,,\, \frac{\tau}{2} \,\right\rangle = -\frac{1}{\sqrt{3}}\left(|\uparrow\downarrow\downarrow\rangle + \bar{\omega}^\tau\,|\downarrow\uparrow\downarrow\rangle + \omega^\tau\,|\downarrow\downarrow\uparrow\rangle\right)\ . \tag{4}$$

Since these four states with $S_z = \pm\frac{1}{2}$ and $T_z = \pm\frac{1}{2}$ are all orthogonal to each other, they span the entire Hilbert space of the spin $S = \frac{1}{2}$ state on a triangle.

2.3 Spins of ions in terms of spin and isospin operators of a triangle

Now that the wavefunctions have been constructed, spin operators for a single magnetic ion with spin $\frac{1}{2}$ can be written in terms of the total spin/isospin of the triangle. This is accomplished by studying the effects of s_γ, $\gamma = 1, 2, 3$ labels the ion, onto all possible four states and finding accordingly the matrix elements of $\langle S_z, t|\boldsymbol{S}_\gamma|S_z', t'\rangle$. In this way we obtain three 4x4 matrices, from which the following identities appear:

$$\left\langle S_z, T_z\middle|\boldsymbol{S}_\gamma\middle|S_z' T_z\right\rangle = \frac{1}{3}\left\langle S_z\middle|\boldsymbol{S}\middle|S_z'\right\rangle \ , \tag{5}$$

$$\left\langle S_z, -T_z\middle|\boldsymbol{S}_\gamma\middle|S_z', T_z\right\rangle = -\frac{2}{3}\omega^{(\gamma-1)\tau}\left\langle S_z\middle|\boldsymbol{S}\middle|S_z'\right\rangle \ . \tag{6}$$

Note that these results are obtained by projection on the four $S = \frac{1}{2}$ states and they do not hold for $S = \frac{3}{2}$ states. Equation (6) can be written as follows:

$$\left\langle S_z, -\tfrac{\tau}{2}\middle|\boldsymbol{S}_1 + \omega\boldsymbol{S}_2 + \bar{\omega}\boldsymbol{S}_3\middle|S_z', \tfrac{\tau}{2}\right\rangle = -\tfrac{2}{3}\left(1 + \omega^{-\tau+1} + \omega^{-2\tau+2}\right)\left\langle S_z\middle|\boldsymbol{S}\middle|S_z'\right\rangle \ .$$

For $\tau = 1$, $\left(1 + \omega^{-\tau+1} + \omega^{-2\tau+2}\right) = 3$, and for $\tau = -1$, $\left(1 + \omega^{-\tau+1} + \omega^{-2\tau+2}\right) = 0$. Hence

$$\left\langle S_z, -\tfrac{\tau}{2}\middle|\boldsymbol{S}_1 + \omega\boldsymbol{S}_2 + \bar{\omega}\boldsymbol{S}_3\middle|S_z', \tfrac{\tau}{2}\right\rangle = -2\delta_{\tau,1}\left\langle S_z\middle|\boldsymbol{S}\middle|S_z'\right\rangle \ .$$

The operator between the ket and bra states acts as the isospin lowering operator, T^- since the result is nonzero only for the isospin $T_z = +\frac{1}{2}$ ket, which after the operation, is transformed into the $T_z = -\frac{1}{2}$ state. Therefore,

$$\boldsymbol{S}_1 + \bar{\omega}\boldsymbol{S}_2 + \omega\boldsymbol{S}_3 = -2\ \boldsymbol{S}\cdot T^- \ . \tag{7}$$

Similarly, the raising isospin operator is given by,

$$\boldsymbol{S}_1 + \omega\boldsymbol{S}_2 + \bar{\omega}\boldsymbol{S}_3 = -2\ \boldsymbol{S}\cdot T^+ \ . \tag{8}$$

The raising and lowering isospin operators are defined in the usual manner: $T^\pm = T_x \pm iT_y$. The two derived identities are are equivalent to

$$\boldsymbol{S}_1 = \tfrac{1}{3}(1 - 4T^x)\boldsymbol{S} \qquad \boldsymbol{S}_2 = \tfrac{1}{3}(1 + 2T^x - 2\sqrt{2}T^y)\boldsymbol{S} \qquad \boldsymbol{S}_3 = \tfrac{1}{3}(1 + 2T^x + 2\sqrt{2}T^y)\boldsymbol{S} \ . \tag{9}$$

Equations (7) and (8) are intriguing, as they relate individual spin components to the total spin of the three ions system, and to the isospin. These results allow for the triangle systems to be treated as point-like objects, described by spin and isospin.

3 Triangle Chain

So far, only a single triangle formed by three magnetic ions with spin $\frac{1}{2}$ was considered. We now create a structure, referred to as a triangle chain, by stacking a multitude of these triangles on top of each other along the c-axis, as shown in Fig. 2. In this section, we study magnetic properties of this chain. Since any spin interaction involving individual atoms can be expressed in terms of the total spin/isospin operators using Eqs. (5-8), each triangle can be thought of as a site. As a result, the problem becomes one-dimensional, which greatly facilitates calculations.

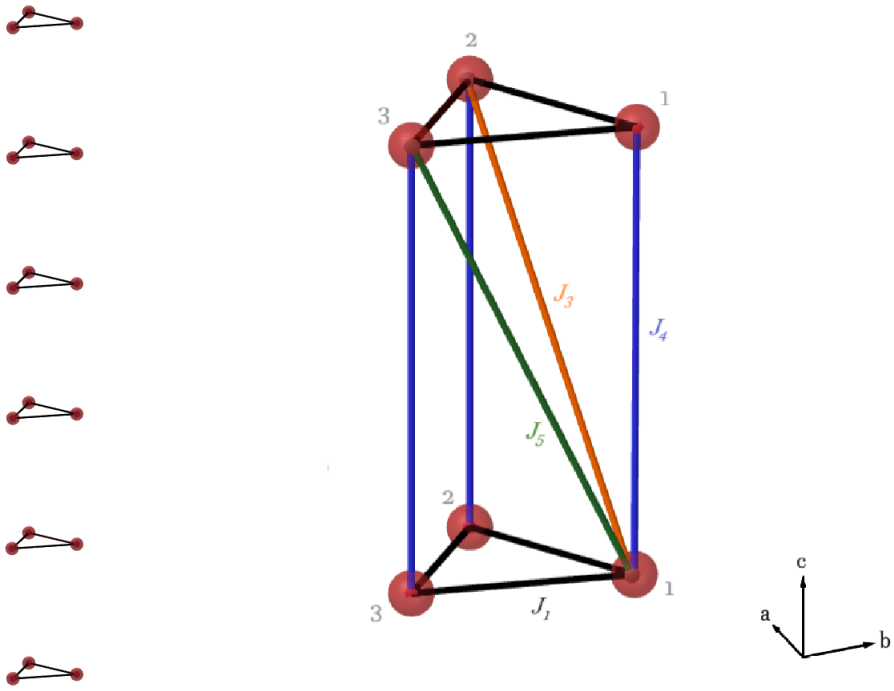

Figure 2: Left: Triangle chain. Right: Fragment of a triangular spin lattice of Fe-langasite

In chiral Fe-langasite, the intertriangle exchange constant J_3 is different from J_5, which leads to a rotation of the ordered magnetic moment of iron ions by 54.1° between layers, resulting in a helical structure [13]. The origin of the helical spiral is explained in literature by treating the large spins of Fe ions classically. In contrast, this section treats the triangle chain quantum-mechanically. We find its ground state, calculate its energy, as well as predict a new spin and isospin state replacing the classical spin helix.
In Fe-langasite (see Fig. 2), there are five distinct Heisenberg exchange interactions. The strongest, J_1, is the interaction within the triangles. J_3, J_4 and J_5 describe the interactions between two neighbouring triangles along the c-axis (see Fig. 2). The coupling J_2 describes interactions between triangles within the plane. However, this term is relatively small and does not appear in the triangle chain.

3.1 Triangle Chain Ground State

Before discussing the whole triangle chain, consider the simpler case of two adjacent triangles. In addition to the interactions within the triangle, described by the Hamiltonian (2), three extra terms appear due to intertriangle interactions depicted in Fig. 2:

$$\begin{aligned} H_{ex} = & J_3 \left(\boldsymbol{S}_{1,1}\cdot\boldsymbol{S}_{2,2} + \boldsymbol{S}_{2,1}\cdot\boldsymbol{S}_{3,2} + \boldsymbol{S}_{3,1}\cdot\boldsymbol{S}_{1,2}\right) \\ & + J_4 \left(\boldsymbol{S}_{1,1}\cdot\boldsymbol{S}_{1,2} + \boldsymbol{S}_{2,1}\cdot\boldsymbol{S}_{2,2} + \boldsymbol{S}_{3,1}\cdot\boldsymbol{S}_{3,2}\right) \\ & + J_5 \left(\boldsymbol{S}_{1,1}\cdot\boldsymbol{S}_{3,2} + \boldsymbol{S}_{2,1}\cdot\boldsymbol{S}_{1,2} + \boldsymbol{S}_{3,1}\cdot\boldsymbol{S}_{2,2}\right) \ , \end{aligned} \tag{10}$$

where the term J_1 was dropped, because only the manifold of $S = \frac{1}{2}$ states on each triangle is considered, which means that Eq.(2) is a constant. In the subscript (i, j), i labels ion in the triangle and j labels to the triangle. The experimental values for the exchange constants in $Ba_3NbFe_3Si_2O_{14}$ are: $J_1 = 1.6$ meV, $J_2 = 0.31$ meV, $J_3 = 0.13$ meV, $J_4 = 0.10$ meV and $J_5 = 0.33$ meV [24]. The constant J_1 describes the relatively strong antiferromagnetic interaction compared to the other terms; $J_1 \gg |J_3|, |J_4|, |J_5|$, which motivated us to consider only the manifold of low-energy $S = \frac{1}{2}$ states on each triangle.

As explained above, the aim is to derive a one-dimensional Hamiltonian written in terms of the spins and isospins operators that act on the whole triangle, rather than those of individual ions. To do so, consider the sum of the dot products $\sum_{n=1}^{3} \boldsymbol{S}_{n,1}\cdot\boldsymbol{S}_{n+j,1}$ expressed through Fourier transform of spins on triangles:

$$\sum_{n=1}^{3} \boldsymbol{S}_{n,1}\cdot\boldsymbol{S}_{n+j,2} = \frac{1}{\sqrt{3}}\sum_n\sum_k \boldsymbol{S}_{n,1}\cdot e^{i\frac{2\pi}{3}k(n+j)}\boldsymbol{S}_{n+j,2} = \sum_k^3 \omega^j \boldsymbol{S}_{-k,1}\cdot\boldsymbol{S}_{k,2} \ ,$$

which can be written as:

$$\begin{aligned} \sum_{n=1}^{3} \boldsymbol{S}_{n,1}\cdot\boldsymbol{S}_{n+k,2} = \frac{1}{3}\big[& (\boldsymbol{S}_{1,1}+\boldsymbol{S}_{2,1}+\boldsymbol{S}_{3,1})\cdot(\boldsymbol{S}_{1,2}+\boldsymbol{S}_{2,2}+\boldsymbol{S}_{3,2}) \\ & + \omega^k\left(\boldsymbol{S}_{1,1}+\omega\boldsymbol{S}_{2,1}+\bar{\omega}\boldsymbol{S}_{3,1}\right)\cdot\left(\boldsymbol{S}_{1,2}+\bar{\omega}\boldsymbol{S}_{2,2}+\omega\boldsymbol{S}_{3,2}\right) \\ & + \bar{\omega}^k\left(\boldsymbol{S}_{1,1}+\bar{\omega}\boldsymbol{S}_{2,1}+\omega\boldsymbol{S}_{3,1}\right)\cdot\left(\boldsymbol{S}_{1,2}+\omega\boldsymbol{S}_{2,2}+\bar{\omega}\boldsymbol{S}_{3,2}\right)\big] \ . \end{aligned}$$

The purpose of this transformation is clear: it allows for the substitution of the raising/lowering isospin operators (see Eqs.(8) and (7)):

$$\sum_{n=1}^{3} \boldsymbol{S}_{n,1}\cdot\boldsymbol{S}_{n+k,2} = \frac{1}{3}\boldsymbol{S}_1\cdot\boldsymbol{S}_2\left[1 + 4\omega^k\ T_1^+T_2^- + 4\bar{\omega}^k\ T_1^-T_2^+\right] \ .$$

The interaction described by the constant J_4 corresponds to $k = 0$, J_3 corresponds to $k = 1$ and J_5 corresponds to $k = 2$. The Hamiltonian of two adjacent triangles in the triangle chain is therefore:

$$\begin{aligned} H_{ex} = \frac{1}{3}\boldsymbol{S}_1\cdot\boldsymbol{S}_2\big[& J_4 + J_3 + J_5 + 2(2J_4 - J_3 - J_5)(T_1^+T_2^- + T_1^-T_2^+) \\ & + i2\sqrt{3}(J_3 - J_5)(T_1^+T_2^- - T_1^-T_2^+)\big] \ . \end{aligned} \tag{11}$$

We can now write down the spin-isospin Hamiltonian of the triangle chain,

$$\begin{aligned} H_{ex} = \frac{1}{3}\sum_n^N \boldsymbol{S}_n\cdot\boldsymbol{S}_{n+1}\big[& J_4 + J_3 + J_5 + 2(2J_4 - J_3 - J_5)(T_n^+T_{n+1}^- + T_n^-T_{n+1}^+) \\ & + i2\sqrt{3}(J_3 - J_5)(T_n^+T_{n+1}^- - T_n^-T_{n+1}^+)\big] \ , \end{aligned} \tag{12}$$

where $\boldsymbol{S}_n$ and $\boldsymbol{T}_n$ are the spin and isospin operators of the n-th triangle. At this point , the spin-isospin model involves a large number of interacting bodies, making it impossible to be solved exactly. Instead, one can simplify it using the mean field approximation: $H = H_A H_B \approx \langle H_A\rangle H_B + H_A\langle H_B\rangle - \langle H_A\rangle\langle H_B\rangle$, i.e $\langle H\rangle \approx \langle H_A\rangle\langle H_B\rangle$. This approximation holds only if the fluctuations of H_A and H_B are small. In our case, the Hamiltonian of the system is separated into spin and isospin components, which leads to the approximation: $\langle H\rangle = \langle H_s H_t\rangle \approx \langle H_s\rangle\langle H_t\rangle$, where $H_s = \frac{1}{3N}\sum_n^N \boldsymbol{S}_n \cdot \boldsymbol{S}_{n+1}$ and $H_t = (J_4 + J_3 + J_5) + 2(2J_4 - J_3 - J_5)(T^-_{n'} \cdot T^+_{n'+1} + T^+_{n'} \cdot T^-_{n'+1}) + i2\sqrt{3}(J_3 - J_5)(T^-_{n'} \cdot T^+_{n'+1} - T^+_{n'} \cdot T^-_{n'+1})$. As a result, the ground state energy per layer $E_0 = \langle H_s\rangle\langle H_t\rangle$ is:

$$\begin{aligned} E_0 = \frac{1}{3N}\sum_{n=1}^N \langle \boldsymbol{S}_n \cdot \boldsymbol{S}_{n+1}\rangle \cdot \Big[&(J_4 + J_3 + J_5) + 2(2J_4 - J_3 - J_5)\langle T^-_{n'} \cdot T^+_{n'+1} + T^+_{n'} \cdot T^-_{n'+1}\rangle \\ &+ i2\sqrt{3}(J_3 - J_5)\langle T^-_{n'} \cdot T^+_{n'+1} - T^+_{n'} \cdot T^-_{n'+1}\rangle\Big] \ . \end{aligned} \tag{13}$$

To find the ground state energy $E_0 = \langle H_s\rangle\langle H_t\rangle$, a systematic approach using the extrema of H_s and H_t is taken. We first calculate the largest and smallest possible expectation values for H_s and H_t, and then combine them. Out of the four possible combinations, the one with the lowest energy is the ground state.
In other words, $E_0 = \min(\langle H_s\rangle_{\min}\langle H_t\rangle_{\min}, \langle H_s\rangle_{\min}\langle H_t\rangle_{\max}, \langle H_s\rangle_{\max}\langle H_t\rangle_{\min}, \langle H_s\rangle_{\max}\langle H_t\rangle_{\max})$. We start by considering the isospin Hamiltonian H_t.

While spin $S = \frac{1}{2}$ systems can be extremely complicated to deal with, in one dimension they behave like fermions. Consequently, the triangle chain can be thought of as an interacting one-dimensional gas of fermions [25]. This is then solved by the introduction of Jordan-Wigner transformation, which consist in mapping all down states $|\downarrow\rangle$ and up state $|\uparrow\rangle$ of the isospin onto the empty state $|0\rangle$ and the state $|1\rangle$ ocuppied by a spinless fermion, respectively. To go from an empty to an occupied state, the creation operator $b^\dagger$ must be applied: $b^\dagger|0\rangle = |1\rangle$. Conversely, there exists an annihilation operator b, such that $b|1\rangle = |0\rangle$. Using commutation relations, the isospin operators can be expressed in terms of $b_n^\dagger$ and b_n as [25]:

$$\begin{aligned} T_n^+ &= (-1)^{\Sigma(1,n-1)} b_n^\dagger \ , \\ T_n^- &= (-1)^{\Sigma(1,n-1)} b_n \ , \\ T_n^z &= -\tfrac{1}{2} + (-1)^{\Sigma(1,n-1)} b_n^\dagger b_n \ , \end{aligned} \tag{14}$$

where $\Sigma(1, n-1) = \sum_{i=1}^{n-1} b_i^\dagger b_i$ represents the number of occupied fermionic states on the left side of the site n. The spinless fermion operators obey the anti-commutation relations: $\{b_n, b_m\} = \delta_{nm}$. Once substituted into the Isospin Hamiltonian, the Jordan Wigner Transformation yields:

$$H_t = (J_4 + J_3 + J_5) + 2(2J_4 - J_3 - J_5)\left(b_n^\dagger b_{n+1} + b_{n+1}^\dagger b_n\right) + i2\sqrt{3}(J_3 - J_5)\left(b_n^\dagger b_n - b_{n+1}^\dagger b_n\right) \ .$$

The Hamiltonian can be further simplified by means of Fourier transform:

$$\begin{cases} b_k &= \frac{1}{\sqrt{N}}\sum_n e^{-ikna} b_n \ , \\ b_n &= \frac{1}{\sqrt{N}}\sum_k e^{ikna} b_k \ , \end{cases}$$

where a is the lattice constant. Then $\langle b_n^\dagger b_{n+1}\rangle$ becomes $\frac{1}{\sqrt{N}}\sum_k\sum_{k'}\langle b_k^\dagger b_{k'}\rangle e^{i(k-k')X_n} e^{ika} = \sum_k e^{ika}\langle b_k^\dagger b_k\rangle$, since $\langle b_k^\dagger b_{k'}\rangle = \delta_{kk'}\langle b_k^\dagger b_{k'}\rangle$. Similarly, $\langle b_{n+1}^\dagger b_n\rangle = \sum_k e^{-ika}\langle b_k^\dagger b_k\rangle$. In this way,

we obtain:

$$\langle H_t\rangle = (J_4+J_3+J_5) + \frac{1}{N}\sum_k \left[2(2J_4-J_3-J_5)(e^{ika}+e^{-ika})\langle b_k^\dagger b_k\rangle + i2\sqrt{3}(J_3-J_5)(e^{ika}-e^{-ika})\langle b_k^\dagger b_k\rangle\right] .$$

Using the notations: $A=(2J_4-J_3-J_5)$, $B=\sqrt{3}(J_3-J_5)$, $\cos(k_o a)=\frac{A}{\sqrt{A^2+B^2}}$ and $\sin(k_o a)=\frac{B}{\sqrt{A^2+B^2}}$, we get:

$$\langle H_t\rangle = (J_4+J_3+J_5) + \frac{1}{N}\sum_k 4\sqrt{A^2+B^2}\ \cos((k+k_o)a)\langle b_k^\dagger b_k\rangle . \tag{15}$$

As explained above, we need to evaluate both the highest and lowest possible values of $\langle H_t\rangle$. The maximum value for $\langle H_t\rangle$ is reached when all the positive energy k-states are occupied, while the negative ones are left empty.

$$\langle H_t\rangle_{\max} = (J_4+J_3+J_5) + \frac{1}{N}\sum_k 4\sqrt{A^2+B^2}\ \cos((k+k_o)a)\langle b_k^\dagger b_k\rangle ,$$

where $-\frac{\pi}{2a}-k_o < k < \frac{\pi}{2a}-k_o$. Since $\Delta k=\frac{2\pi}{Na}\ll 1$, the Riemann Sum approximation can be applied:

$$\begin{aligned}\langle H_t\rangle_{\max} &\approx (J_4+J_3+J_5) + \frac{2a}{\pi}\sqrt{A^2+B^2}\int_{-\frac{\pi}{2a}-k_o}^{\frac{\pi}{2a}-k_o}\cos((k+k_o)a)\,dk \\ &\approx (J_4+J_3+J_5) + \frac{4}{\pi}\sqrt{A^2+B^2} .\end{aligned} \tag{16}$$

On the other hand, the lowest expectation value of H_t occurs when only the negative energy k-states are occupied. Using the same method, we find:

$$\langle H_t\rangle_{\min} \approx (J_4+J_3+J_5) - \frac{4}{\pi}\sqrt{A^2+B^2} .$$

In the spin term H_s, the spin of the triangles can interact in two different ways: antiferromagnetically or ferromagnetically. Depending on how the spins couple, the expectation value of H_s changes. The two possible value for the term H_s are [26]:

$$\frac{1}{3N}\sum_{n=1}^{N}\langle \boldsymbol{S}_n\cdot\boldsymbol{S}_{n+1}\rangle = \begin{cases}\frac{1}{12N}, & \text{ferromagnetic}\\ \frac{1}{3N}\left(\frac{1}{4}-\ln 2\right). & \text{antiferromagnetic}\end{cases} \tag{17}$$

Therefore, the ferromagnetic and antiferromagnetic couplings correspond to $\langle H_s\rangle_{\max}$ and $\langle H_s\rangle_{\min}$, respectively. The absolute value of $\langle H_s\rangle_{\max}$ is greater than that of $\langle H_s\rangle_{\min}$, i.e. $|\frac{1}{4}| < |\frac{1}{4}-\ln 2|$. In addition, the constants J_3, J_4 and J_5 are positive [24], so $|\langle H_t\rangle_{\min}| < |\langle H_t\rangle_{\max}|$. Therefore, the combination with the highest absolute value is $\langle H_s\rangle_{\min}\langle H_t\rangle_{\max}$, and as it turns out, this term is negative. This means that out of the four possible combinations, this is the one with the smallest value.

Hence, in the ground state of the triangle chain, spins interactions between triangles is antiferromagnetic, whereas isospin coupling is ferromagnetic. However, one-dimensional Heisenberg quantum antiferromagnets do not possess Néel long range ordering due to quantum fluctuations in the system [27]. Therefore, at temperature $T=0K$ (no thermal excitation), there is no long-range spin ordering in the triangle chain. Note that isospins are also disordered in the sense that $\langle \boldsymbol{T}_n\rangle$=0. The energy per triangle of the ground state is given by Eqs.(16) and (17):

$$E_0 = -(\ln 2-\frac{1}{4})\left[(J_4+J_3+J_5)+\frac{4}{\pi}\sqrt{(2J_4-J_3-J_5)^2+3(J_3-J_5)^2}\right] . \tag{18}$$

Using the values of the exchange constants given in Ref.[24], the ground state energy is of the order of magnitude $\sim -10^{-1}$meV. This energy is not affected by the angle $k_o a$ that appeared in the Hamiltonian $\langle H_t \rangle$ (see Eq.(15)). However, it does impact the ground state, as well as the structure of the triangle chain. This will be studied in the next section.

3.2 Isospin Rotation

While spins form a disordered spin liquid state, the isospin shows more structure. Fermi sea of spinless fermions is shifted by a wave-vector k_o, which although has no effect on the ground state energy, it impacts the isospin structure. To investigate this, consider the correlation functions $\langle T_n^i T_{n+1}^i \rangle$ and $\langle T_n^i T_{n+1}^j \rangle$. In particular,

$$\begin{aligned}\langle T_n^x T_{n+1}^x \rangle &= \tfrac{1}{4}\langle (T_n^+ + T_n^-)(T_{n+1}^+ + T_{n+1}^-) \rangle \ , \\ &= \tfrac{1}{4}\langle b_n^\dagger b_{n+1}^\dagger + b_n^\dagger b_{n+1} + b_{n+1}^\dagger b_n - b_n b_{n+1} \rangle \ .\end{aligned}$$

Recall that $\langle b_n^\dagger b_{n+1} + b_{n+1}^\dagger b_n \rangle = \frac{1}{N}\sum_k \left(e^{ika} + e^{-ika}\right) \langle b_k^\dagger b_k \rangle$. In the ground state, $\langle b_k^\dagger b_k \rangle = 1$, for $-\frac{\pi}{2a} - k_o < k < \frac{\pi}{2a} - k_o$, and zero otherwise. In addition, $\langle b_n^\dagger b_{n+1}^\dagger \rangle = \langle b_n b_{n+1} \rangle = 0$. Hence, the previous equation simplifies to:

$$\langle T_n^x T_{n+1}^x \rangle = -\tfrac{1}{4N}\sum_k \left(e^{ika} + e^{-ika}\right) \ , \qquad \text{where} -\tfrac{\pi}{2a} - k_o < k < \tfrac{\pi}{2a} - k_o \ ,$$

so that

$$\approx -\frac{a}{4\pi}\int_{-\frac{\pi}{2a}}^{\frac{\pi}{2a}} \cos((k-k_o)a)\, dk = -\frac{1}{2\pi}\cos(k_o a) \ . \tag{19}$$

Similarly:

$$\begin{aligned}\langle T_n^y T_{n+1}^y \rangle &= -\tfrac{1}{4}\langle (T_n^+ - T_n^-)(T_{n+1}^+ - T_{n+1}^-) \rangle \\ &= -\tfrac{1}{4}\langle b_n^\dagger b_{n+1}^\dagger - b_n^\dagger b_{n+1} - b_{n+1}^\dagger b_n - b_n b_{n+1} \rangle \\ &= \tfrac{1}{4}\langle b_n^\dagger b_{n+1} + b_{n+1}^\dagger b_n \rangle \\ &= \langle T_n^x T_{n+1}^x \rangle\end{aligned}$$

Substituting the definition of $\cos k_o a$ into Eq.(19), we obtain

$$\langle T_n^x T_{n+1}^x \rangle = \langle T_n^y T_{n+1}^y \rangle = -\frac{1}{2\pi}\frac{2J_4 - J_3 - J_5}{\sqrt{(2J_4 - J_3 - J_5)^2 + 3(J_3 - J_5)^2}} \ . \tag{20}$$

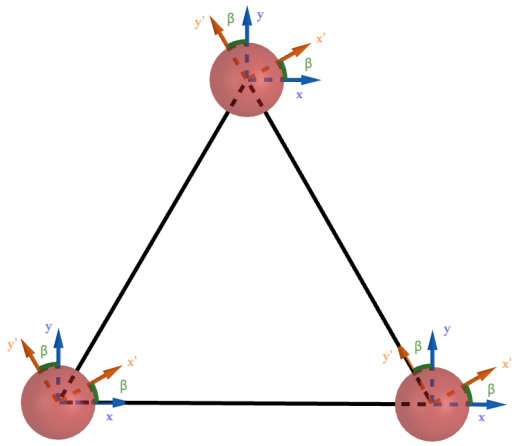

Figure 3: Isospin shift between two layers

In the same way, we obtain

$$\langle T_n^x T_{n+1}^y \rangle = -\frac{i}{4}\langle (T_n^+ + T_n^-)(T_{n+1}^+ - T_{n+1}^-)\rangle = \langle b_n^\dagger b_{n+1} - b_{n+1}^\dagger b_n \rangle$$
$$= -\frac{i}{4N}\sum_k \left(e^{ika} - e^{-ika}\right) \approx \frac{a}{4\pi}\int_{-\frac{\pi}{2a}}^{\frac{\pi}{2a}} \sin((k-k_o)a)\, dk = \frac{1}{2\pi}\sin(k_o a)) \ .$$

Concerning $\langle T_n^x T_{n+1}^y \rangle$, its expectation value is:

$$\langle T_n^y T_{n+1}^x \rangle = -\frac{i}{4}\langle (T_n^+ - T_n^-)(T_{n+1}^+ + T_{n+1}^-)\rangle = -\langle b_n^\dagger b_{n+1} - b_{n+1}^\dagger b_n \rangle = -\langle T_n^x T_{n+1}^y \rangle \ .$$

As a result, the two correlation functions are given by:

$$\langle T_n^y T_{n+1}^x \rangle = -\langle T_n^x T_{n+1}^y \rangle = -\frac{1}{2\pi}\frac{\sqrt{3}(J_3 - J_5)}{\sqrt{(2J_4 - J_3 - J_5)^2 + 3(J_3 - J_5)^2}} \ . \tag{21}$$

Equations (20) and (21) can be interpreted as follows: isospin of the triangle $n+1$ is rotated with respect to the isospin of the triangle n in the xy-plane by the angle β:

$$\tan\beta = \frac{\sqrt{3}(J_3 - J_5)}{2J_4 - J_3 - J_5} \ . \tag{22}$$

This result shows the (short ranged) helical spiral ordering of isospins in the ground state; φ. This isospin rotation is analogous to the spin rotation around the c-axis observed in the Fe-langasite. In fact, the rotation angle is equal in the classical and quantum models [24]. Still, the spin-isospin model is very different, since in addition to the isospin (and not the spin) rotating, there is no long range spin ordering.

The chirality of the material is also showcased, since the reflection accross the ac-plane or bc-plane of the triangle chain would interchange J_3 and J_5. The absence of mirrors in the symmetry group of the Fe-langasites is the source of the helical order. Chirality is also an important feature of langasites, which gives rise to interesting properties of this class of materials, such as topologically stable antiferromagnetic skyrmions [11].

4 Triangle Mean Field

Although the triangle chain model provides a great insight regarding isospin ordering and its rotation around the z-axis, it misses a key aspect of the material: in-plane interactions. So far, only the Heisenberg exchange between two vertically adjacent triangles was considered, however in the Fe-langasite crystal structure, spins also interact with their neighbours lying in the same ab-plane but in a different triangle. The exchange constant of this interaction is J_2. This section focus on finding the consequences of the in-plane intertriangle interaction for the spin ordering in the ground state.

4.1 Ground State

To extend the triangle chain model and approach a more realistic situation, we take into account the interactions between neighbouring spin triangles in the ab-plane. As for the previous sections, this crystal structure is inspired by the Fe-langasite, in which the spin triangles form a triangular lattice in each ab-layer (see Fig.4).

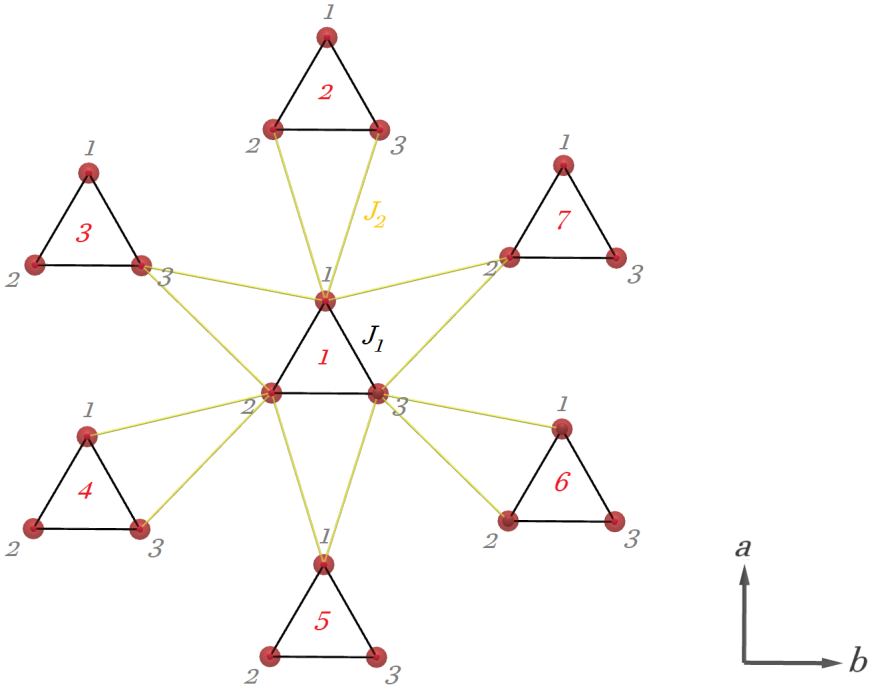

Figure 4: Interactions between spins of neighbouring triangles in the ab-plane

By adding a new interaction in the model, a new term must appear in the Hamiltonian. It describes the four J_2 Heisenberg exchange interactions experienced by each magnetic ion in the triangle and it is given by:

$$\begin{aligned} H_p = &- J_2[\boldsymbol{S}_{1,1} \cdot (\boldsymbol{S}_{2,2} + \boldsymbol{S}_{3,2} + \boldsymbol{S}_{3,3} + \boldsymbol{S}_{2,7}) + \boldsymbol{S}_{2,1} \cdot (\boldsymbol{S}_{1,4} + \boldsymbol{S}_{3,4} + \boldsymbol{S}_{3,3} + \boldsymbol{S}_{1,5}) \\ &+ \boldsymbol{S}_{3,1} \cdot (\boldsymbol{S}_{1,6} + \boldsymbol{S}_{2,6} + \boldsymbol{S}_{1,5} + \boldsymbol{S}_{2,7})] \ . \end{aligned}$$

Similarly to the triangle chain case, finding the exact ground states of this Hamiltonian is impossible and we have to resort to an approximation. Mean field theory is once again used to

simplify the Hamiltonian but now we assume that in the ground state spins are ordered. Instead of dividing the Hamiltonian into spin and isospin components, the mean field applied to the spins of a given triangle is created by spins from neighbouring triangles. The mean field Hamiltonian for spins in the triangle 1 has the form:

$$\begin{aligned} H_p = &- J_2[\boldsymbol{S}_{1,1} \cdot \langle \boldsymbol{S}_{2,2} + \boldsymbol{S}_{3,2} + \boldsymbol{S}_{3,3} + \boldsymbol{S}_{2,7} \rangle + \boldsymbol{S}_{2,1} \cdot \langle \boldsymbol{S}_{1,4} + \boldsymbol{S}_{3,4} + \boldsymbol{S}_{3,3} + \boldsymbol{S}_{1,5} \rangle \\ &+ \boldsymbol{S}_{3,1} \cdot \langle \boldsymbol{S}_{1,6} + \boldsymbol{S}_{2,6} + \boldsymbol{S}_{1,5} + \boldsymbol{S}_{2,7} \rangle] \ , \end{aligned}$$

where the brackets denote the average over the ground state that has to be calculated in a self-consistent way. The interactions between spin triangles in the same and different layers make the problem three-dimensional and result in a long range ordering or spins. In the Fe-langasite, the exchange constant is $J_2 > 0$, corresponding to an antiferromagnetic interaction [24] favouring antiparallel spins. However, it is impossible to have all neighbouring spins antiparallel in a triangle – geometric frustration. As a result, spins in the lowest-energy state form the Y-type structure with 120° between spins. Consequently, in the ground state of the three dimensional model, the average total spin of each triangle is zero: $\langle \boldsymbol{S}_1 \rangle + \langle \boldsymbol{S}_2 \rangle + \langle \boldsymbol{S}_3 \rangle = 0$.
Since the sum of the three average spin vectors is equal to zero, they must lie in the same plane. Let this plane be the XY-plane, then:

$$\langle \boldsymbol{S}_1 \rangle = S_o \hat{X} \ , \qquad \langle \boldsymbol{S}_2 \rangle = S_o \left(-\tfrac{1}{2} \hat{X} + \tfrac{\sqrt{3}}{2} \hat{Y} \right) \ , \qquad \langle \boldsymbol{S}_3 \rangle = S_o \left(-\tfrac{1}{2} \hat{X} - \tfrac{\sqrt{3}}{2} \hat{Y} \right) \ . \tag{23}$$

Where S_o is the length of the ordered spins. Note that the XYZ coordinates in the spin space are arbitrary and, in general, different from the xyz axes of the coordinate system spin used for isospin, and relation between the two has yet to be established. Importantly, the spin ordering is the same for all triangles: $\langle \boldsymbol{S}_{i,j} \rangle = \langle \boldsymbol{S}_{i,k} \rangle$, which makes it possible to simplify the in-plane interaction Hamiltonian:

$$\begin{aligned} H_p &= -2J_2 \textstyle\sum_n^3 \boldsymbol{S}_n \cdot \langle \boldsymbol{S}_n \rangle \\ &= -2J_2 S_o \left(S_1^X - \tfrac{1}{2}(S_2^X + S_3^X) + \tfrac{\sqrt{3}}{2}(S_2^Y - S_3^Y) \right) \\ &= 4J_2 S_o \left[T^x S^X + T^y S^Y \right] \ , \end{aligned}$$

where Eq.(9) was used to go from the second to the third line. The next step is to add the interactions between the triangles stacked along the c-axis, which we also treat in the mean field approximation. Along the c-axis, spins are coupled via the J_3, J_4 and J_5 interactions with spins in the triangle above and below. The mean field Hamiltonian describing these interactions is:

$$\begin{aligned} H_z = &\ J_4 \textstyle\sum_n \boldsymbol{S}_n \cdot (\langle \boldsymbol{S}_{n,+z} \rangle + \langle \boldsymbol{S}_{n,-z} \rangle) \\ &+ J_3 \textstyle\sum_n \boldsymbol{S}_n \cdot (\langle \boldsymbol{S}_{n+1,+z} \rangle + \langle \boldsymbol{S}_{n-1,-z} \rangle) \\ &+ J_5 \textstyle\sum_n \boldsymbol{S}_n \cdot (\langle \boldsymbol{S}_{n-1,+z} \rangle + \langle \boldsymbol{S}_{n+1,-z} \rangle) \ . \end{aligned} \tag{24}$$

An expression for $\langle S_{n,\pm z} \rangle$ is now needed to proceed further. Although the sum of the averaged spin in a triangle is zero no matter the layer, there is no reason for the individual expectation values to be the same in all layers. Since only the direction and not the magnitude of the average spin will change, let φ be the rotation angle of $\langle \boldsymbol{S}_i \rangle$ around the Z-axis when going up a layer. As a result, the vectors transform to:

$$\begin{aligned} \langle \boldsymbol{S}_{1,\pm z} \rangle &= S_o(\cos(\pm\varphi)\hat{x} + \sin(\pm\varphi)\hat{y}) \ , \\ \langle \boldsymbol{S}_{2,\pm z} \rangle &= S_o \left((-\tfrac{1}{2}\cos(\pm\varphi) - \tfrac{\sqrt{3}}{2}\sin(\pm\varphi))\hat{x} + (\tfrac{\sqrt{3}}{2}\cos(\pm\varphi) - \tfrac{1}{2}\sin(\pm\varphi))\hat{y} \right) \ , \\ \langle \boldsymbol{S}_{3,\pm z} \rangle &= S_o \left((-\tfrac{1}{2}\cos(\pm\varphi) + \tfrac{\sqrt{3}}{2}\sin(\pm\varphi))\hat{x} - (\tfrac{\sqrt{3}}{2}\cos(\pm\varphi) + \tfrac{1}{2}\sin(\pm\varphi))\hat{y} \right) \ . \end{aligned} \tag{25}$$

These values are then substituted into the Hamiltonian (24). Then, H_z and H_p are combined to obtain the full mean field Hamiltonian of the whole triangle mean field. The resulting expression is given by:

$$\begin{aligned} H &= 2S_o \left[2J_2 - 2\cos\varphi J_4 + (\cos\varphi + \sqrt{3}\sin\varphi)J_3 + (\cos\varphi - \sqrt{3}\sin\varphi)J_5\right](T^x S^X + T^y S^Y) \\ &= S_o \left[2J_2 - 2\cos\varphi J_4 + (\cos\varphi + \sqrt{3}\sin\varphi)J_3 + (\cos\varphi - \sqrt{3}\sin\varphi)J_5\right](T^+ S^- + T^- S^+) \ , \end{aligned}$$

where $S^{\pm}$ are the spin raising and lowering operator and are defined as $S^{\pm} = S_x \pm iS_y$. Note that since the spin and isospin operators act on different subset, it must also be the case for the lowering and raising operators. To simplify this expression, let $A = (-2J_4 + J_3 + J_5)$ and $B = \sqrt{3}(J_5 - J_3)$, along with $\cos(\theta) = \frac{A}{\sqrt{A^2+B^2}}$ and $\sin(\theta) = \frac{B}{\sqrt{A^2+B^2}}$. Then

$$H = S_o \left[2J_2 + \sqrt{A^2 + B^2}\cos(\theta - \varphi)\right](T^+ S^- + T^- S^+) \ .$$

We now have to find the ground state of the mean field Hamiltonian. Clearly, none of the states constructed in section 2.2 is an eigenstate of the operator $\hat{A} = T^+S^- + T^-S^+$. As a result, a new quantum number called the total angular momentum, J, is introduced. It is the sum of spin and isospin: $\boldsymbol{J} = \boldsymbol{S} + \boldsymbol{T}$. This allows for a change of basis where the four new states of the triangle are now expressed in the form $|J, J_z\rangle$, where J_z is the z-component of the total angular momentum. This gives rise to three triplet states: $|1\ 1\rangle = \uparrow\uparrow$, $|1\ 0\rangle = \frac{1}{\sqrt{2}}(\uparrow\downarrow + \downarrow\uparrow)$, $|1\ -1\rangle = \downarrow\downarrow$ and a singlet $|0\ 0\rangle = \frac{1}{\sqrt{2}}(\uparrow\downarrow - \downarrow\uparrow)$, where the first arrow denotes the Z-component of the total spin and the second one is the z-component of the total isospin. This new set of states spans the whole Hilbert space, since they are all orthogonal to each other. These four state are eigenstates of $\hat{A}$: $\hat{A}\,|1\ 0\rangle = |1\ 0\rangle$, $\hat{A}\,|1\ 1\rangle = \hat{A}\,|1\ -1\rangle = 0$ and $\hat{A}\,|0\ 0\rangle = -\,|0\ 0\rangle$.

Next we select the lowest-energy eigenstate. Immediately, the states $|1\ 1\rangle$ and $|1\ -1\rangle$ can be discarded, since their eigenvalue is zero and it is obvious that by using one of the other two states, a lower energy ground can be reached. For positive J_2, the lowest energy state is the state with the eigenvalue of the operator $\hat{A}$ $\lambda = -1$. In addition, the angle φ must be such that it minimises the energy, consequently it is chosen to make the term $\cos(\theta - \varphi)$ as large as possible. This occurs when $\cos(\theta - \varphi) = 1$, which corresponds to $\varphi = \theta$. Therefore, the ground state is the singlet state $|0\ 0\rangle$, the ground state energy is:

$$E_0 = -S_o[2J_2 + \sqrt{(2J_4 - J_3 - J_5)^2 + 3(J_3 - J_5)^2}] \ ,$$

and the angle φ is:

$$\varphi = \arctan\left(\frac{\sqrt{3}(J_3 - J_5)}{2J_4 - J_3 - J_5}\right) \ . \tag{26}$$

The magnitude of the average spin S_o is found from the self consistency conditions. This is done by calculating the expectation value of $\boldsymbol{S}_1$, $\boldsymbol{S}_2$ and $\boldsymbol{S}_3$ in the ground state $|0\ 0\rangle$. The result is:

$$\langle \boldsymbol{S}_1 \rangle = \frac{1}{3}\begin{pmatrix}1\\0\\0\end{pmatrix} \ , \qquad \langle \boldsymbol{S}_2 \rangle = \frac{1}{3}\begin{pmatrix}-1/2\\ \sqrt{3}/2\\0\end{pmatrix} \ , \qquad \langle \boldsymbol{S}_3 \rangle = \frac{1}{3}\begin{pmatrix}-1/2\\ -\sqrt{3}/2\\0\end{pmatrix} \ .$$

Using Eq.(23), we obtain $S_o = \frac{1}{3}$. Therefore, the ground state energy per triangle is:

$$E_0 = -\tfrac{1}{3}[2J_2 + \sqrt{(2J_4 - J_3 - J_5)^2 + 3(J_3 - J_5)^2}] \ . \tag{27}$$

This clarifies an important property of the spin ordering. In addition to the 120° spin ordering in each triangle, average spins also rotate from layer to layer. They rotate around the Z-axis and the angle between spins in two consecutive planes is given by Eq.(26). Intriguingly, the angle φ is equal to the angle of rotation of isospin β in the spin chain model. In three dimensional model spins and isospins rotate at the same rate. However, since the XYZ axes in spin space are arbitrary, spins and isospins do not necessarily rotate in the same plane.

5 Perturbations of the Ground State

Although the Heisenberg exchange is the governing interaction behind spin ordering in quantum antiferromagnets, other effects such as the Dzyaloshinskii-Moriya interaction or the exchange anisotropy, are known to have important effects on magnetic ordering. Still, those interactions are much weaker than the Heisenberg exchange in materials with $3d$ transition metal ions, which is why they could be initially ignored when the ground state is discussed. These relatively weak interactions can be treated using first-order perturbation theory, which is done in this section.

5.1 Dzyaloshinskii-Moriya Interaction

We first discuss the Dzyaloshinskii-Moriya (DM) interaction, which is an antisymmetric exchange interaction favouring spin canting, spirals and skyrmions [28]. This exchange coupling is the first-order correction of the Heisenberg exchange interaction due to the spin–orbit coupling [29]. Although the DM interaction is relatively weak, it is of great importance due to its tendency to favour twisted spin pairs, leading to the formation of various chiral topological magnetic structures [29]. We are interested in effects of the DM interaction on the helical structure formed by spins and isospins. The DM interactions between spins on sites i and j has the form

$$\hat{H}_{DM} = \textstyle\sum_{i,j} \boldsymbol{D_{ij}} \cdot (\boldsymbol{S}_i \times \boldsymbol{S}_j) \ . \tag{28}$$

Some components of this Hamiltonian may disappear due to symmetries of the (i,j)-bond [30]. For a quantum antiferromagnet with the structure of Fe-langasite, there exists a $\frac{2\pi}{3}$ discrete rotational symmetry around the z-axis, which results in the vanishing of the x and y components of the vector D_{ij} in spin triangles. Hence, the Dzyaloshinskii-Moriya in this triangular lattice is given by:

$$\begin{aligned} H_{DM} &= D_z \left([\boldsymbol{S}_1 \times \boldsymbol{S}_2]_z + [\boldsymbol{S}_2 \times \boldsymbol{S}_3]_z + [\boldsymbol{S}_3 \times \boldsymbol{S}_1]_z \right) \\ &= D_z \textstyle\sum_n^3 S_n^x S_{n+1}^y - S_n^y S_{n+1}^x \\ &= \tfrac{i}{2} D_z \textstyle\sum_n^3 S_n^+ S_{n+1}^- - S_n^- S_{n+1}^+ \ , \end{aligned}$$

where $S_4 = S_1$. To understand the effects of this operator, let us apply it to the spin/isospin states of the individual triangles (see Eqs.(3) and (4)):

$$\begin{aligned} H_{DM} \left| \ \frac{1}{2} \ , \ \frac{\tau}{2} \ \right\rangle &= \tfrac{i}{2\sqrt{3}} D_z \left(\textstyle\sum_n^3 S_n^+ S_{n+1}^- - S_n^- S_{n+1}^+ \right) \left(|{\downarrow}{\uparrow}{\uparrow}\rangle + \omega^\tau \, |{\uparrow}{\downarrow}{\uparrow}\rangle + \bar{\omega}^\tau \, |{\uparrow}{\uparrow}{\downarrow}\rangle \right) \\ &= \tfrac{i}{2\sqrt{3}} D_z \left((\bar{\omega}^\tau - \omega^\tau) \, |{\downarrow}{\uparrow}{\uparrow}\rangle + (1 - \bar{\omega}^\tau) \, |{\uparrow}{\downarrow}{\uparrow}\rangle + (\omega^\tau - 1) \, |{\uparrow}{\uparrow}{\downarrow}\rangle \right) \\ &= \tfrac{i}{2} D_z (\bar{\omega}^\tau - \omega^\tau) \left| \ \frac{1}{2} \ , \ \frac{\tau}{2} \ \right\rangle \\ &= \tau \frac{\sqrt{3}}{2} D_z \left| \ \frac{1}{2} \ , \ \frac{\tau}{2} \ \right\rangle \ , \\ H_{DM} \left| \ -\frac{1}{2} \ , \ \frac{\tau}{2} \ \right\rangle &= -\tfrac{i}{2\sqrt{3}} D_z \left((\omega^\tau - \bar{\omega}^\tau) \, |{\uparrow}{\downarrow}{\downarrow}\rangle + (\bar{\omega}^\tau - 1) \, |{\downarrow}{\uparrow}{\downarrow}\rangle + (1 - \omega^\tau) \, |{\downarrow}{\downarrow}{\uparrow}\rangle \right) \\ &= -\tau \frac{\sqrt{3}}{2} D_z \left| \ \frac{1}{2} \ , \ \frac{\tau}{2} \ \right\rangle \ . \end{aligned}$$

Thus in terms of the spin and isospin operators of the triangle, the Dzyaloshinskii-Moriya interaction is given by:

$$H_{DM} = \sqrt{3} D_z \; T^z S^z \; . \tag{29}$$

To find the change in energy of the triangle due to the DM interaction, we calculate the expectation value $\sqrt{3} D_z \, \langle 00 | T^z S^z | 00 \rangle$. However, the XY-plane is an arbitrary plane that differs from xy plane and $S^z \neq S^Z$. To account for this, $\hat{z}$ must be transformed into XYZ coordinates, using $\hat{z} = (\hat{z} \cdot \hat{X})\hat{X} + (\hat{z} \cdot \hat{Y})\hat{Y} + (\hat{z} \cdot \hat{Z})\hat{Z}$. In this manner, the operator $S^z = \boldsymbol{S} \cdot \hat{z}$ is expressed in the coordinate system XYZ as:

$$S_z = (\hat{z} \cdot \hat{X}) S^X + (\hat{z} \cdot \hat{Y}) S^Y + (\hat{z} \cdot \hat{Z}) S^Z \; .$$

When evaluating $\langle 00 | T^z S^z | 00 \rangle$, the terms $\langle 00 | T^z S^X | 00 \rangle$ and $\langle 00 | T^z S^Y | 00 \rangle$ vanish since $|00\rangle$ is not an eigenstate of these operators, and is instead transformed into a state orthogonal to itself. Therefore, the perturbation simplifies to:

$$\delta E_{DM} = \sqrt{3} D_z \, \langle 00 | T^z S^z | 00 \rangle \, \hat{z} \cdot \hat{Z} = -\frac{\sqrt{3}}{4} D_z \; \hat{z} \cdot \hat{Z} \; . \tag{30}$$

In the ground state, this term of the Hamiltonian should be minimised in order to reach the lowest energy configuration. However, since it contains $T^z S^z$, doing so implies that the isospin and spin z axes must align. Thus, we found that, depending on whether D_z is positive or negative, the z axes of the two coordinates systems are anti-parallel or parallel:

$$\begin{cases} \text{if } D_z < 0, \; \hat{z} \uparrow\uparrow \hat{Z} \\ \text{if } D_z > 0, \; \hat{z} \uparrow\downarrow \hat{Z} \end{cases} \; .$$

The previously unrelated z and Z axes, around which isospin and spin rotate, are now coupled by the DM interaction. Therefore, the DM interaction forces the average spins and isospins of the magnetic ions to lie in the same plane. If the constant D_z is positive, the z and Z axes align and hence the isospin and spin rotate in the same direction, whereas if the DM constant is negative, they will rotate in opposite directions.

5.2 Exchange Anisotropy

So far, it was assumed that the exchange constants in Eq.(1) for the Heisenberg exchange are the same for all spin directions. However, due to the low symmetry of the Fe-langasite, this is not the case. The exchange interaction between the two spins in triangles splits into two distinct parts: $J_{\parallel}$ and $J_{\perp}$:

$$\begin{aligned} H_{ex} &= J_{\perp}(S_1^x S_2^x + S_1^y S_2^y) + J_{\parallel} S_1^z S_2^z \\ &= J_{\perp}(S_1^x S_2^x + S_1^y S_2^y + S_1^z S_2^z) + (J_{\parallel} - J_{\perp}) S_1^z S_2^z \ . \end{aligned}$$

After substitution, $J_{\perp} = J$ and $J_{\parallel} - J_{\perp} = \Delta$, an extra term, called the exchange anisotropy, appears. Even though this splitting is present for all exchange constants J_i, since J_1 is the greatest term and this is already a relatively small perturbation, it is sufficient to consider the anisotropy only within spin triangles and ignore the less significant exchange interactions. The Hamiltonian of this perturbation is:

$$H_A = \Delta \sum_n^3 S_n^z S_{n+1}^z \ .$$

Applying H_A to the four states in the triangle, we obtain

$$\begin{aligned} H_A \left| \ \frac{1}{2} \ , \ \frac{\tau}{2} \ \right\rangle &= -\frac{1}{4}\Delta \left| \ \frac{1}{2} \ , \ \frac{\tau}{2} \ \right\rangle \ , \\ H_A \left| \ -\frac{1}{2} \ , \ \frac{\tau}{2} \ \right\rangle &= -\frac{1}{4}\Delta \left| \ -\frac{1}{2} \ , \ \frac{\tau}{2} \ \right\rangle \ . \end{aligned}$$

Clearly, the exchange anisotropy, when projected on $S = \frac{1}{2}$ states, is simply a constant, which does not depend on the state the operator is acting on. Hence, H_A simplifies to:

$$H_A = -\frac{\Delta}{4} \ . \tag{31}$$

We thus found that the exchange anisotropy does not affect the spin ordering in the quantum antiferromagnet.

6 Discussion

A new quantum degree of freedom appear, when the spins $s = \frac{5}{2}$ iron ions in Fe-langasite are replaced by spins $s = \frac{1}{2}$. This quantum number, called isospin, gives rise to new physics. The first noticeable difference occurs when looking at the ground state of the triangle chain: while spins form a disordered spin liquid state, isospin shows a short ranged helical spiral ordering. Between two ab layers, the average isospin rotates around the z-axis by an angle β, such that $\tan\beta = \frac{\sqrt{3}(J_3-J_5)}{2J_4-J_3-J_5}$. This helical order is the result of the lack of reflection symmetry across the ac-plane or bc-plane, which leads to $J_3 \neq J_5$. By contrast, in Fe-langasite there is no isospin. Instead, the classical spin rotates with the same angle β around the c-axis.

Adding in-plane interactions causes the configuration of average spins to change. The lowest energy configuration occurs when the average total spin of the triangle is zero and the three spins show a noncollinear 120° spin ordering. It corresponds to the state of the quantum spin-isospin model, in which the sum of spin and isospin in each triangle is zero. In contrast to the one-dimensional model, this ground state allows for spins to be ordered. In fact, just like for the helical isospin ordering, the average spins rotate along the c-axis. The angle of rotation φ equals β, which means that the average spin and isospin rotate together. However, the axis around which spins rotate, the Z-axis, is arbitrary. In other words, although they rotate with the same rate, the plane of rotation is not necessarily the same.

The DM interaction in this triangular crystal lattice structure favours the alignment of the the isospin z-axis and the spin Z-axis. Whether they are parallel or anti-parallel is dictated by the sign of the DM constant. As a result, the average spin rotates in the same xy-plane as isospin. Depending on the axes alignment, the spin rotation is clockwise or anti-clockwise. This is in agreement with the classical model, in which the chirality of the spin spiral also depends on D_z. The exchange anisotropy turned out not to affect the spin and isospin ordering.

These results were obtained using mean field approximation. It involves averaging all interactions acting on a system and it works well as long as the fluctuations of these interactions are small. However, quantum Heisenberg antiferromagnets are known to show significant quantum fluctuations in their ground state [31]. In low-dimensional quantum spin systems, these fluctuations can be strong enough to destroy magnetic ordering or give rise to new phases [32]. However, three dimensional quantum triangular spin models are often ordered in the ground state. At absolute zero, this ordering is only a locally stable minimum in the total energy, but it is not the absolute minimum, because long range ordering is disrupted by the quantum fluctuations [33]. Consequently, the use of mean field only allows for the prediction of spin-isospin short range ordering. Whether the long range orders discussed in this thesis survive spin fluctuations is an interesting question.

Another important aspect to consider is thermal fluctuations. In the derivations, we have assumed that the spin lattice was at temperature $T = 0$ K and, hence, there were no thermal excitations. Thermal fluctuations reduce the value of the ordered spins. However, even if the spin order is entirely suppressed, the isospin order may still persist. It would be interesting to consider this possibility.

This investigation opens the door for multiple topics of further research. One interesting direction would be to go beyond the mean field approximation. This approximation is rather crude, since it does not account for quantum spin and isospin fluctuations in the ground state. In the antiferromagnetic spintronics research, the use of various approximation methods is often needed to solve a many body problem. Among those, there are a few that capture quantum fluctuations, such as quantum Monte Carlo simulations [34][35], renormalisation group theory [36][37] and dynamical mean field theory [38][39].

Numerous applications of antiferromagnets involve making use of their relative insensitivity to external magnetic fields. Therefore, it is interesting to study behaviour of the quantum triangular antiferromagnet under an applied magnetic field. In this investigation, the anisotropy exchange was quickly discussed and put aside due to its limited influence on the structure. However, it plays an extremely important role when an external magnetic field is applied to the system. The effects of magnetic exchange anisotropies have to be studied in more detail.

The theoretical study of Fe-langasite has suggested existence of three-dimensional skyrmions and coreless vertex tubes in Fe-langasite [14]. It would be intresting to understand if these fascinating topological defects also exist in the the quantum spin-isospin model. The study of differences between topological defects in the classical and quantum models could yield interesting results.

7 Conclusion

In this thesis, we explored the effects of replacing the spins $s = \frac{1}{2}$ irons ions by spins $s = \frac{1}{2}$ ions in the Fe-langasite, a triangular antiferromagnet. This substitution resulted in the emergence of a new quantum degree of freedom, isospin. We first examined a structure that consists in triangles stacked on top of each other along the c-axis. In this one-dimensional model, called the triangle chain, we found that spins form a disordered spin liquid state, whereas isospins exhibit short ranged helical spiral ordering. Between two ab layers, the average isospin rotates around the z-axis by an angle β, such that $\tan\beta = \frac{\sqrt{3}(J_3-J_5)}{2J_4-J_3-J_5}$.

Then, the triangle chain was extended by taking into account the interactions between neighbouring spin triangles in the ab plane. This caused the average total spin of triangles to be equal to zero, and the ions to form a noncollinear 120° spin ordering. In the resulting ground state, spins rotate along the c-axis with an angle φ, where $\varphi = \beta$. However, the spins plane of rotation is arbitrary and is not necessarily the same as the isospins plane of rotation. To establish a relation between the two, the DM interaction must be introduced. Depending on the sign of the DM constant, the spins Z-axis and isospins z-axis will either be parallel or antiparallel. The anisotropy exchange was also examined, however it does not affect the spin ordering in the quantum antiferromagnet.

To obtain these results, a simplification was needed and we chose the mean field approximation. However, this approximation ignores quantum fluctuations, which are known for destroying magnetic ordering in antiferromagnets. Consequently, it is uncertain whether long range spin-isospin ordering survives against these fluctuations. Nonetheless, it is expected for the short-range orders discussed in this thesis to exist in the ground state.

Finally, we discussed potential topics for further research. These include going beyond the mean field approximation to account for possible quantum spins and isospins fluctuations using more powerful tools. Studying the effects of external magnetic fields on quantum triangular antiferromagnet could prove useful for potential applications. Additionally, investigating the potential existence of topological defects in the spin-isospin model would be of interest.

References

[1] S. Blundell and D. Thouless, "Magnetism in condensed matter," *American journal of physics*, vol. 71, no. 1, pp. 94–95, Dec. 2002.

[2] S. Palagummi and F.-g. Yuan, *Magnetic levitation and its application for low frequency vibration energy harvesting*. Jan. 2016, pp. 213–251.

[3] M. C. Guidi, G. Allodi, R. De Renzi, *et al.*, "Staggered magnetization, critical behavior, and weak ferromagnetic properties ofLaMnO3by muon spin rotation," *Physical review. B, Condensed matter*, vol. 64, no. 6, Jul. 2001.

[4] P. Khuntia and A. V. Mahajan, "Magnetic susceptibility and heat capacity of a novel antiferromagnet: LiNi2P3O10and the effect of doping," *Journal of physics. Condensed matter*, vol. 22, no. 29, p. 296 002, Jun. 2010.

[5] D. E. Laughlin, "Magnetic transformations and phase diagrams," *Metallurgical and materials transactions. A, Physical metallurgy and materials science*, vol. 50, no. 6, pp. 2555–2569, Apr. 2019.

[6] V. Baltz, A. Manchon, M. Tsoi, T. Moriyama, T. Ono, and Y. Tserkovnyak, "Antiferromagnetic spintronics," *Reviews of modern physics*, vol. 90, no. 1, Feb. 2018.

[7] T. Jungwirth, X. Marti, P. Wadley, and J. Wunderlich, "Antiferromagnetic spintronics," *Nature nanotechnology*, vol. 11, no. 3, pp. 231–241, Mar. 2016.

[8] T. G. Golovina, A. F. Konstantinova, A. P. Dudka, *et al.*, "Structure and Optical Properties of Langasite Family Crystals (La1–xNdx)3Ga5SiO14 (x = 0, 0.4, 0.6, 1)," *Crystallography reports*, vol. 68, no. 5, pp. 732–743, Oct. 2023.

[9] A. F. Konstantinova, T. G. Golovina, A. P. Dudka, I. O. Goryachuk, and V. I. Sokolov, "Measurement and Calculation of the Refractive Indices of Langasite-Family Crystals Sr3NbFe3Si2O14, Ba3NbFe3Si2O14, and Ba3TaFe3Si2O14 and the Relationship of Optical Activity with Peculiarities of Electron-Density Distribution," *Crystallography reports*, vol. 67, no. 6, pp. 951–957, Dec. 2022.

[10] K. Marty, P. Bordet, V. Simonet, *et al.*, "Magnetic and dielectric properties in the langasite-type compounds:A3BFe3D2O14," *Physical review. B, Condensed matter and materials physics*, vol. 81, no. 5, Feb. 2010.

[11] M. Ramakrishnan, E. Constable, A. Cano, *et al.*, "Field-induced double spin spiral in a frustrated chiral magnet," *npj quantum materials*, vol. 4, no. 1, Dec. 2019.

[12] H. Narita, Y. Tokunaga, A. Kikkawa, Y. Taguchi, Y. Tokura, and Y. Takahashi, "Observation of nonreciprocal directional dichroism via electromagnon resonance in a chiral-lattice helimagnetBa3NbFe3Si2," *Physical review. B./Physical review. B*, vol. 94, no. 9, Sep. 2016.

[13] I. S. Lyubutin, M. A. Chuev, S. S. Starchikov, K. O. Funtov, and I. Y. Starchikova, "Helical spin structure in the langasite family multiferroic Ba3TaFe3Si2O14, determined by Mössbauer spectroscopy," *Europhysics letters*, vol. 128, no. 6, p. 67 005, Feb. 2020.

[14] E. Barts and M. Mostovoy, "Magnetic particles and strings in iron langasite," *npj quantum materials*, vol. 6, no. 1, Dec. 2021.

[15] H. Q. Lin and W. Y. Chen, "Order parameter in the antiferromagnetic Heisenberg model," *Journal of the Physical Society of Japan*, vol. 68, no. 5, pp. 1699–1702, May 1999.

[16] C. M. Varma, *Critical overview of theories for high temperature superconductors*. Jan. 1988, pp. 13–25.

[17] D. A. Huse and V. Elser, "Simple variational wave functions for Two-Dimensional Heisenberg spin-1/2 antiferromagnets," *Physical review letters*, vol. 60, no. 24, pp. 2531–2534, Jun. 1988.

[18] X. Z. Liu, O. Prokhnenko, M. Bartkowiak, *et al.*, "Ground state of the S=12 triangular lattice Heisenberg-like antiferromagnet Ba3CoSb2," *Physical review. B./Physical review. B*, vol. 105, no. 21, Jun. 2022.

[19] Y.-F. Jiang and H.-C. Jiang, "Nature of quantum spin liquids of the S=12 Heisenberg antiferromagnet on the triangular lattice: A parallel DMRG study," *Physical review. B./Physical review. B*, vol. 107, no. 14, Apr. 2023.

[20] C. Broholm, R. J. Cava, S. A. Kivelson, D. G. Nocera, M. R. Norman, and T. Senthil, "Quantum spin liquids," *Science*, vol. 367, no. 6475, Jan. 2020.

[21] B. Bernu, P. Lecheminant, C. Lhuillier, and L. Pierre, "Exact spectra, spin susceptibilities, and order parameter of the quantum Heisenberg antiferromagnet on the triangular lattice," *Physical review. B, Condensed matter*, vol. 50, no. 14, pp. 10 048–10 062, Oct. 1994.

[22] S. R. White and A. L. Chernyshev, "Neél order in square and triangular lattice Heisenberg models," *Physical review letters*, vol. 99, no. 12, Sep. 2007.

[23] K. Bolsmann, A. Khudoyberdiev, and G. S. Uhrig, "Switching the magnetization in quantum antiferromagnets," *PRX quantum*, vol. 4, no. 3, Sep. 2023.

[24] C. Stock, L. C. Chapon, A. Schneidewind, *et al.*, "Helical spin waves, magnetic order, and fluctuations in the langasite compound Ba3NbFe3Si," *Physical review. B, Condensed matter and materials physics*, vol. 83, no. 10, Mar. 2011.

[25] P. Phillips, *Second quantization*. Mar. 2012, pp. 24–30.

[26] T. Giamarchi, *Quantum Physics in one dimension*. Oxford University Press, Jan. 2004.

[27] J. Richter, J. Schulenburg, and A. Honecker, *Quantum Magnetism*. Springer, Jan. 2004.

[28] R. E. Camley and K. L. Livesey, "Consequences of the Dzyaloshinskii-Moriya interaction," *Surface science reports*, vol. 78, no. 3, p. 100 605, Aug. 2023.

[29] H. Yang, J. Liang, and Q. Cui, "First-principles calculations for Dzyaloshinskii–Moriya interaction," *Nature reviews physics*, vol. 5, no. 1, pp. 43–61, Nov. 2022.

[30] T. Moriya, "Anisotropic superexchange interaction and weak ferromagnetism," *Physical review*, vol. 120, no. 1, pp. 91–98, Oct. 1960.

[31] K. W. Plumb, H. J. Changlani, A. Scheie, *et al.*, "Continuum of quantum fluctuations in a three-dimensional S = 1 Heisenberg magnet," *Nature physics*, vol. 15, no. 1, pp. 54–59, Oct. 2018.

[32] J. Merino and A. Ralko, "Role of quantum fluctuations on spin liquids and ordered phases in the Heisenberg model on the honeycomb lattice," *Physical review. B./Physical review. B*, vol. 97, no. 20, May 2018.

[33] P. Anderson, "Resonating valence bonds: A new kind of insulator?" *Materials research bulletin*, vol. 8, no. 2, pp. 153–160, Feb. 1973.

[34] K. J. Runge, "Quantum Monte Carlo calculation of the long-range order in the Heisenberg antiferromagnet," *Physical review. B, Condensed matter*, vol. 45, no. 13, pp. 7229–7236, Apr. 1992.

[35] J. D. Reger, J. A. Riera, and A. P. Young, "Monte Carlo simulations of the spin-1/2Heisenberg antiferromagnet in two dimensions," *Journal of physics. Condensed matter*, vol. 1, no. 10, pp. 1855–1865, Mar. 1989.

[36] G. Sierra and M. A. Martín-Delgado, "Real-space renormalization group approach to the 2D antiferromagnetic Heisenberg model," *Physics letters. B*, vol. 391, no. 3-4, pp. 381–387, Jan. 1997.

[37] F. Borges, A. Borissov, A. Singh, A. Schlief, and S.-S. Lee, "Field-theoretic functional renormalization group formalism for non-Fermi liquids and its application to the antiferromagnetic quantum critical metal in two dimensions," *Annals of physics*, vol. 450, p. 169 221, Mar. 2023.

[38] J. Otsuki and Y. Kuramoto, "Dynamical mean-field theory for quantum spin systems: Test of solutions for magnetically ordered states," *Physical review. B, Condensed matter and materials physics*, vol. 88, no. 2, Jul. 2013.

[39] A. Georges, R. Siddharthan, and S. Florens, "Dynamical Mean-Field Theory of Resonating-Valence-Bond antiferromagnets," *Physical review letters*, vol. 87, no. 27, Dec. 2001.