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Anisotropic Heisenberg quantum spin chains: Excitations and neutron scattering cross sections

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The thesis is submitted to University College Dublin in fulfilment of the requirements for the degree of Doctor of Philosophy

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Abstract

The research presented here focuses on the theoretical investigation of spinons in quasi one-dimensional quantum spin chains from the perspective of neutron scattering, with emphasis placed on the calculation of dynamic spin structure factors for both unpolarised and polarised incident neutrons. Two anisotropic versions of the Heisenberg spin chain are considered for such an examination: the spin-1/2 antiferromagnetic $XXZ$ model and the spin-1/2 ferromagnetic $XYZ$ model.

The $XYZ$ model supports spinon scattering (modelled at finite temperature) and is related to the so-called Villain mode. The introduction of a perturbation in the form of an external transverse magnetic field or Dzyaloshinskii-Moriya interaction (DMI) leads to the emergence of incommensurability in the system; such a result is a signature of fractional excitations. The presence of these interactions also give rise to a chiral response associated with the spinons in the polarised term of the scattering cross section. The inelastic scattering cross section is computed for both unpolarised and polarised neutrons.

In relation to the antiferromagnetic $XXZ$ model, emphasis is on spinon pair creation (modelled at $T = 0$ K). Motivated by the presence of a chiral response in the case of the $XYZ$ model mentioned above, a new technique is developed in order to incorporate DMI and the transverse magnetic field so that one can determine if there is a chiral property associated with the two-spinon system. This technique is numerical in nature and is based on Green function methods. Using the results produced by the Green function approach, the inelastic cross section in the presence of an external transverse magnetic field is computed for the first time. The result is compared with experimental neutron scattering data; good agreement is demonstrated between theory and experiments on CsCoBr$_3$.

Finally, the Green function approach is generalised to account for the interactions between spin chains in magnetic compounds described by a staggered field. Various dynamic structure factors are computed for this scenario with results compared to experiment; good agreement between the theory and experiments on RbCoCl$_3$ is shown.
Statement of Original Authorship

I, Leonard Patrick English, hereby certify that the submitted work is my own work, was completed while registered as a candidate for the degree of Doctor of Philosophy, and I have not obtained a degree elsewhere on the basis of the research presented in this submitted work.
Collaborations

In my work I have been fortunate to collaborate with the following researchers:

Jiri Kulda (ILL, France) - provided details of experimental equipment and experimental data for inelastic scattering cross section related to CsCoBr$_3$ in the presence of a transverse magnetic field. Experimental results presented in Chapter 6.

Matt Mena (PSI, Switzerland and UCL, England) - provided experimental data in Chapter 7 for interchain interaction present in RbCoCl$_3$. 

Dedicated to my parents.
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Chapter 1

Introduction

Low dimensional magnetism has proved to be a rich area of research for physicists since the development of the field’s first classical model in 1925 by Ising [1]. At the time, investigation in this direction was very appealing to theorists as it allowed for the computation of analytical results while avoiding the complexities of the three-dimensional models. The development of quantum mechanics lead to further progress, with a more fundamental microscopic theory of magnetism arising in the late 1920s from the work of Heisenberg [2] and Dirac [3] on the concept of exchange interaction. In this thesis, we focus on quantum aspects of magnetism, in particular the low-lying magnetic excitations, known as spinons, present in quasi one-dimensional quantum spin chains. Spinons are modelled as completely delocalised entities and, in the context of this research, may be introduced into a spin chain either by thermal fluctuations or by probing neutrons in a scattering experiment. Unlike magnons, which are spin-1 excitations, spinons possess spin-$1/2$ and are therefore referred to as fractional excitations. The theoretical investigation that is discussed in subsequent chapters is performed in the context of inelastic neutron scattering. Specifically, interest lies in calculating dynamic spin structure factors which detail the dynamics of the excitations in question.

From a fundamental perspective, low dimensional spin systems represent a paradigm for exploring truly quantum phenomena: quantum phase transitions [4], quantum entanglement [5, 6], and frustrated spin configurations [7] are just some of the interesting and novel properties that these systems can exhibit. By incorporating the experimental method of polarised neutron scattering into this work, one can obtain insight relating to another fascinating property of spinons, namely chirality; detecting chiral responses associated with structure factors is a key component of this work. Furthermore, following from Braun and Loss [8], it appears that chirality plays a key role in 2D antiferromagnetic (AFM) systems. The significance of this lies in the fact that the high-$T_c$ superconductor class of undoped cuprates can be mapped to a generic 2D AFM system. Hence, the chiral nature of magnetic excitations may hold the key to explaining the elusive mechanism
behind high-$T_c$ superconductivity.

One of the significant outcomes of the research presented in this thesis is the generalisation of a numerical technique, based on Green functions, which permits the computation of dynamic spin structure factors. It specifically pertains to the case of spinon pair creation and allows one, for the first time, to incorporate the effects of an external magnetic field and Dzyaloshinskii-Moriya interaction in the spin systems of interest; the corresponding algorithm is discussed in detail in Chapter 4. Motivation for including such interactions stems from the desire to gain greater insight about the chiral properties of spinons. This powerful technique is further generalised in Chapter 7 to account for interactions arising between spin chains.

Chapter 2 introduces the one-dimensional anisotropic or “Ising-like” quantum spin models central to this work; these are the spin-$1/2$ antiferromagnetic (AFM) $XXZ$ models and spin-$1/2$ ferromagnetic (FM) $XYZ$ models. The low-lying excitations occurring in spin chains are also introduced. Specifically, the cases of interest are spin chains catering for one and two spinons; the corresponding dispersion relations are determined. The concept of chirality is also explained.

Chapter 3 details the investigation of a spin-$1/2$ FM $XYZ$ system at finite temperature accommodating a single spinon. This theoretical examination pertains to neutron scattering, specifically to the computation of dynamic spin structure factors for the low energy (quasielastic) response at finite temperatures. The process associated with this part of the research can be viewed as spinon scattering within a single band and provides the FM analogue of the Villain mode. In addition to this, the structure factors associated with this scattering process are calculated when the system experiences a transverse magnetic field or Dzyaloshinskii-Moriya interaction. Motivation for including such interactions stems from the desire to gain greater insight about the chiral properties of spinons in bulk crystals and artificially produced spin chains on substrates.

In Chapter 4, the analytical approach involving Green functions originally used to compute dynamic spin structure factors for the case of two spinon excitations in an AFM background is introduced. This provides the inspiration to apply a numerical recursion technique to obtain the same quantities, as well as generalising it to include an external transverse magnetic field and Dzyaloshinskii-Moriya interaction.

Chapter 5 presents the results of spinon-pair creation in a spin chain described by an anisotropic spin-$1/2$ AFM $XXZ$ model. Specifically, the dynamic structure factors are calculated within the Green function framework. For the first time, the dynamic structure factors are computed when the spin chain is in the presence of an external transverse magnetic field and/or possesses the Dzyaloshinskii-Moriya interaction.

Chapter 6 details the theoretical result derived by the Green function technique for the inelastic neutron scattering cross section in the presence of a transverse magnetic field, the first time that
such a result that has been produced. This is compared to experimental neutron scattering data of CsCoBr$_3$ for the case of both unpolarised and polarised incident neutrons.

In Chapter 7, this Green function approach is further progressed to take into account the effects of the interaction which arises between spin chains in various magnetic compounds. Such a staggered field produces bound states; the theoretical results for this aspect of the research are compared with experimental data pertaining to neutron scattering on RbCoCl$_3$.

Chapter 8 contains summary of the work presented in this dissertation and discusses potential future work.
Chapter 2

Spin-1/2 models

2.1 Introduction and motivation

This chapter describes the two anisotropic spin-1/2 models for one-dimensional spin chains that are at the heart of this research; these are the AFM $XXZ$ model and the FM $XYZ$ model. Specifically, one is interested in the low-lying excitations concerning one and two spinons in the chain; the corresponding eigenstates are determined and dispersion relations are derived. While the AFM $XXZ$ model has been studied extensively, not much theoretical work has been produced for the FM $XYZ$ case. Finally, chirality associated with the elementary excitations is also discussed.

2.2 Heisenberg model

As discussed in the previous section, Heisenberg [2] and Dirac [3] derived the first model that encapsulated the purely quantum effect of exchange. The corresponding Hamiltonian was originally presented in terms of permutation operators, but the typical form which it assumes (henceforth to be referred to as the Heisenberg Hamiltonian) is given, for an open chain with $N$ sites with nearest-neighbour interaction:

$$H = \sum_{\langle i,j \rangle} J S_i \cdot S_j,$$

(2.1)

where $S_k$ represents the spin operator at site $k$ and $J$ is the exchange constant; $\langle i,j \rangle$ denotes nearest neighbour interaction. In general, $J$ may be positive or negative, favouring antiferromagnetic or ferromagnetic coupling, respectively. Even though the original model was based on the assumption of orthogonal states and localised electrons [2], the Heisenberg Hamiltonian has been surprisingly successful in describing a plethora of magnetic systems, with early emphasis be-
ing placed on the one-dimensional case with nearest-neighbour interactions, \textit{i.e.}, Equation (2.1) above. For $S = 1/2$, the earliest and most striking result is that of Bethe’s work [9]. While this tour de force is much lauded and produced the ground state wavefunction for the 1D AFM linear spin chain, it was Hulthén [10] who extended this endeavour to make the formalities associated with the Bethe Ansatz more accessible and, in doing so, calculated the system’s ground state energy. These ground-breaking accomplishments marked the beginning of what would become one of the most intensely active areas of research in condensed matter physics: one-dimensional quantum spin systems. Such systems have acted as a test bed for a gamut of theoretical approaches, which in turn has led to a spawning of techniques that have far outgrown their original purposes. The Heisenberg model itself has served as a prototype of quantum integrable models [11], with the mathematical structure of Bethe Ansatz motivating the development of the Yang-Baxter framework [12], followed by the formulation of the more exotic quantum groups [13]. Continuing in the vein of exactly solvable models, analytical results of the 1D spin-1/2 $XY$ model can be achieved by mapping this system, using a Jordan-Wigner transformation, to that of spinless fermions described by a tight-binding model, a perspective that led to bosonisation techniques. In terms of numerical approaches, a prominent example is that of the density matrix renormalisation group (DMRG), which was first applied to a 1D quantum spin system [14] but is now implemented in a vast array of low dimensional strongly-correlated systems [15, 16]. In fact, DMRG was originally applied to the spin-1/2 Heisenberg model in order to obtain the ground state energy [14].

A more inventive approach to solving problems of low-dimensional magnetism has arisen from the use of quantum field theory techniques: ever since Landau’s interpretation of the ground state of a many-body system as being equivalent to a vacuum in field theory, and elementary excitations associated with such a system as being like quanta originating from the field, condensed matter physics and quantum field theory have borrowed concepts from one another at increasing frequency; models and techniques from quantum field theory used in the realm of condensed matter include bosonisation, conformal field theory and the non-linear sigma model [17].

With the ground state intensely studied, the natural progression was towards investigating the excited states; the elementary excitations of the Heisenberg model are referred to as spinons. The first examination of these particular states was performed by des Cloiseaux and Pearson [18], and subsequently Yamada [19]. The result is the now-familiar excitation continuum depicted in Figure 2.1; one of the first observations of the theoretically predicted continuum was by Tennant et al. [20]. The theoretical investigation of spinons progressed with the salient work of Karbach et al. [21] providing an exact expression for the longitudinal dynamic spin structure factor for the case of a two-spinon system described by the 1D AFM nearest-neighbour spin-1/2 Heisenberg model. As will be discussed in subsequent chapters, it is dynamic structure factors which form the vital connections between theory and experiment. These endeavours are representative of the highly collaborative research surrounding low dimensional magnetic systems; with the advent of the number of high-flux neutron sources (for example, Institut Laue-Langevin (ILL), France;
Figure 2.1: Two spinon continuum (blue region) associated with the isotropic spin-1/2 AFM Heisenberg model. The continuum is bounded by the curves $\omega_1$ and $\omega_2$.

Paul Scherrer Institute (PSI), Switzerland; Oak Ridge National Laboratory (ORNL), USA), there has been an intense interaction between theory and experiment, all of which has been mediated by the development of sophisticated crystal-growth techniques. With that, the work presented here revolves around the calculation of dynamic spin structure factors associated with low-lying excitations governed by two different models which are variations of the nearest-neighbour spin-1/2 Heisenberg model of Equation (2.1); these are the $XXZ$ model with AFM coupling and the $XYZ$ model with ferromagnetic (FM) coupling. In keeping with the spirit of collaboration between theory and experiment, the theoretical results calculated as part of this research are compared to neutron scattering data collected at ILL and PSI in Chapters 6 and 7, respectively. In order to further discuss both the $XXZ$ and $XYZ$ models, it is instructive to write the Heisenberg Hamiltonian with general exchange constants

\[
H = \sum_i J_x S_i^x S_{i+1}^x + J_y S_i^y S_{i+1}^y + J_z S_i^z S_{i+1}^z.
\] (2.2)

Equation (2.1) is recovered for $J = J_x = J_y = J_z$. In order to realise either the $XXZ$ model or $XYZ$ model, the exchange energy associated with the different spin components in Equation (2.2) must be weighted differently: for the $XXZ$ model, $J_x = J_y < J_z$, while for the $XYZ$ model $J_x \neq J_y \neq J_z$. In particular for this work, we focus on the strongly anisotropic (“Ising”) cases
with $J_z \gg J_t$ for the $XXZ$ model and $J_z \gg J_x, J_y$ for the $XYZ$ model. This altering of the exchange energies corresponds to a transition from an isotropic gapless system (Heisenberg model) to one that is anisotropic ($XXZ$ model or $XYZ$ model) and which exhibits a gap for two-spinon excitations. A physical mechanism that is able to induce anisotropy is spin-orbit coupling. As well as providing physics which is rich both from a theoretical and experimental point of view, the anisotropic nature of these models allows one to develop a more intuitive picture of the excitations that occur in one-dimensional quantum spin systems: in subsequent discussions it will be noted that the dominant Ising exchange interaction naturally gives rise to the elementary excitations in the form of delocalised domain walls.

### 2.3 The Spin-1/2 AFM $XXZ$ Model

In the case of the $XXZ$ model, inspired by compounds such as CsCoBr$_3$ and CsCoCl$_3$, the focus is on the case $J_z \gg J_t > 0$. With that, equation (2.2) now becomes

$$H_{XXZ} = \sum_i J_z S_z^i S_z^{i+1} + J_t (S_x^i S_x^{i+1} + S_y^i S_y^{i+1}),$$

(2.3)

This particular nearest-neighbour model has a rich history of investigation. As in the case of the Heisenberg model, the $XXZ$ model is exactly solvable by implementation of the Bethe Ansatz [22], or via the framework developed by Baxter with the six-vertex model [23]; one can also deduce properties associated with the system’s ground state, as well as thermodynamic quantities [24]. Realisations of this anisotropic model have come in the form of compounds such as TlCoCl$_3$ [25], CsCoCl$_3$ [26] and CsCoBr$_3$ [27]; in CsCoBr$_3$, for example, $J_t/J_z \approx 0.1$ [17]. Examining Equation (2.3) in more detail, the transverse term in the $XXZ$ Hamiltonian can be treated as a perturbation and also encompasses the quantum fluctuations. These fluctuations become apparent when the transverse spin operators are expressed in terms of raising and lowering operators:

$$H_{XXZ} = \sum_{i} J_z S_z^i S_z^{i+1} + \sum_{i} J_t \left( S_x^i S_x^{i+1} + S_y^i S_y^{i+1} \right),$$

(2.4)

Given the dominant longitudinal exchange interaction, the natural choice of basis is the Ising basis, which serves as the starting point for investigating this anisotropic system. Due to the AFM
ordering, there are two degenerate ground states of $\mathcal{H}_I$, the Néel states (Figure 2.2(a)):

\begin{align}
|A\rangle &= |\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\cdots\uparrow\downarrow\rangle, \\
|B\rangle &= |\downarrow\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\cdots\downarrow\uparrow\rangle.
\end{align}

Note that the lattice spacing, $a$, between each lattice site is here set to unity throughout this dissertation, unless stated otherwise.

### 2.3.1 One spinon states

For the case of a pure Ising system, the lowest-lying static excitation that can be introduced is a single domain wall. Following the notation of [8], the corresponding kets are given as:

\begin{align}
|m, Q = +1\rangle &= |\downarrow\uparrow\cdots\downarrow\uparrow\downarrow\cdots\downarrow\uparrow\rangle, \ m \ odd, \\
|m, Q = +1\rangle &= |\downarrow\uparrow\cdots\uparrow\downarrow\downarrow\cdots\downarrow\uparrow\rangle, \ m \ even, \\
|m, Q = -1\rangle &= |\uparrow\downarrow\cdots\uparrow\downarrow\uparrow\cdots\uparrow\downarrow\rangle, \ m \ odd, \\
|m, Q = -1\rangle &= |\uparrow\downarrow\cdots\uparrow\downarrow\uparrow\cdots\uparrow\downarrow\rangle, \ m \ even.
\end{align}

$Q$ represents the so-called topological charge [8], and indicates whether the domain wall alters from Néel state $|A\rangle$ to $|B\rangle$ or from $|B\rangle$ to $|A\rangle$. It is $m$ which marks the site after which this interpolation occurs, i.e., indicates the position of the domain wall. Because of the translational invariance of the system, the states in Equations (2.6) are $2N$-fold degenerate, with $N$ being the number of sites in the chain; $N$ is odd to enforce the existence of one domain wall in the system.
under periodic boundary conditions. These excited states are eigenstates of $H_I$:

$$H_I|m, Q\rangle = \frac{J_z}{2}|m, Q\rangle,$$  \hspace{1cm} (2.7)

and therefore an energy cost of $J_z/2$ is required to create one domain wall from a Néel state. However, when the transverse term, $H_{XY}$, acts upon these states (2.6), either new domain wall pairs are introduced into the system or the original domain wall is mobilised. The creation or translation of domain walls depends on which site the $H_{XY}$ acts upon: if $H_{XY}$ operates on the two lattice sites which are respectively adjacent and one lattice spacing away from the initial nucleation site, denoted by $m$ in Equations 2.6, then the domain wall is moved by two lattice spacings ‘backwards’ or two lattice spacings ‘forward’; this is presented in Equation (2.8) for sites 4 and 5, and 6 and 7.

$$H_{XY}|\downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \rangle_{123456789} = \begin{cases} \left| \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \rangle_{123456789} \right| & \text{act on 4 and 5,} \\ \left| \downarrow \uparrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \rangle_{123456789} \right| & \text{act on 6 and 7,} \\ \left| \downarrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \rangle_{123456789} \right| & \text{act on 2 and 3.} \\ 0 & \text{act on 5 and 6} \end{cases}$$  \hspace{1cm} (2.8)

If $H_{XY}$ acts on the two sites either side of the initial domain wall, an eigenvalue of zero will result, as in Equation (2.8) for sites 5 and 6; any other site will lead to the creation of new domain wall pairs (sites 2 and 3 in the same equation). However, the calculations performed here are in a low energy regime ($\approx J_z$) and the Villain [28] approximation is implemented. This is whereby the creation of new domain walls in the spin chain is ignored due to an additional energy cost of $O(J_z)$ and focus is given solely to the dynamics of the single domain walls represented in Equations (2.6). The overall physical interpretation of this is that the existence of quantum fluctuations in the 1D Ising-type AFM chain, quantified by $H_{XY}$, are responsible for the motion of the domain walls; explicitly

$$H_{XY}|m, Q\rangle = \frac{J_z}{2}|m + 2, Q\rangle + \frac{J_t}{2}|m - 2, Q\rangle.$$  

Overall one has

$$H_{XXZ}|m, Q\rangle = \frac{J_z}{2}|m, Q\rangle + \frac{J_t}{2}|m + 2, Q\rangle + \frac{J_t}{2}|m - 2, Q\rangle.$$  \hspace{1cm} (2.9)

It is clear that $|m, Q\rangle$ are not eigenstates of $H_{XXZ}$; to the leading order in $J_t/J_z$, the eigenstates in fact have the Bloch form [8]:
Figure 2.3: Dispersion relation of one spinon state in the first Brillouin zone. For CsCoBr$_3$, $J_z = 13.8$ meV, $J_t = \epsilon J_z = 1.9$ meV, $\epsilon \equiv J_t/J_z = 0.137$. [17]. $\epsilon_k$ is measured units of meV while $k$ is expressed in units of inverse lattice constant.

$$|k\rangle = \frac{1}{\sqrt{2N}} \sum_{m=1}^{2N} e^{ikm} |m, 1\rangle. \quad (2.10)$$

Here one defines $|m + N, Q\rangle = |m, -Q\rangle$, implying that only after two circulations around the ring-like chain does the domain wall arrive back at the original state. The most pertinent remark about the above superposition is that the domain walls are completely delocalised, and it is this superposition which one refers to as a spinon. The corresponding dispersion relation, as illustrated in Figure 2.3, is [28]:

$$\epsilon_k = \frac{J_z}{2} + J_t \cos 2k. \quad (2.11)$$

### 2.3.2 Two spinon states

The second class of low-lying excitations that are investigated is that of the two spinon states; instead of having a single delocalised domain wall, as in the above, there are now two. With calculations associated with these entities pertaining to a temperature regime of $T = 0$ K, a realistic assumption to make is that these excitations are dilute. With that, one can assume that the two spinons can be treated independently so that the total energy is obtained by adding the individual energies, given by Equation (2.11),
Figure 2.4: Two-spinon continuum for spin-1/2 AFM XXZ model for $J_z = 13.8$ meV, $J_t = \epsilon J_z = 1.9$ meV, $\epsilon \equiv J_t/J_z = 0.137$. [17].

$$\omega(k) = \epsilon_{k_1} + \epsilon_{k_2} = J_z + 2J_t \cos k \cos 2q,$$  \hspace{1cm} (2.12)

where $k = k_1 + k_2$ is the centre-of-mass momentum and $q = (k_1 - k_2)/2$ is the relative momentum of the low-lying excitations. By varying $k$ for a fixed value of the relative momentum, $q$, one can produce the familiar gapped two spinon continuum, as presented in Figure 2.4. Comparing this to the isotropic case of Figure 2.1, one sees that the anisotropy of the model induces the excitation spectrum to become gapped; this will also be the case for the XYZ model discussed in the next section.

Before further discussing the two-spinon states within this dilute approximation, it is worthwhile to mention the work undertaken to produce the exact solution for the XXZ model: using the Bethe Ansatz, the first theoretical analysis was performed by Johnson and McCoy [29] to provide an exact solution of the model. The corresponding excitation spectrum is shown in Figure 2.5. The continuum is bounded by the following expression [30]

$$\epsilon_L(k) = A_c(1 + m) \sin k,$$  \hspace{1cm} (2.13a)

$$\epsilon_U(k) = 2A_c \sqrt{\sin^2 \frac{k}{2} + m^2 \cos^2 \frac{k}{2}},$$  \hspace{1cm} (2.13b)

$$\bar{\epsilon}_U(k) = 2A_c \sqrt{m^2 \sin^2 \frac{k}{2} + \cos^2 \frac{k}{2}},$$  \hspace{1cm} (2.13c)
where $m$ is the parameter which dictates whether the system is in the Ising limit ($m \rightarrow 1$) or the $XY$ limit ($m \rightarrow 0$). The amplitude $A_c$ is detailed in the following set of equations [30]:

\begin{align}
\frac{\pi K(m)}{K'(m)} &= \lambda, \\
\lambda &= 2\zeta, \\
\text{sech } 2\zeta &= \alpha, \\
A_c &= \frac{J_z \sqrt{1 - \alpha^2} K'(m)}{\pi}, \\
\cos k_c &= \frac{(1 - m)}{(1 + m)},
\end{align}

where $k_c$ indicates the domain of $\varepsilon_L(k)$; $K$ is the elliptic integral of the first kind and $K'(m) = K(\sqrt{1 - m^2})$ (Abramowitz and Stegun notation). One can see that differences exist between the excitation spectrum for a given value of $m$ of the exact solution (Figure 2.5) and that of Figure 2.4. The most obvious difference occurs around $k = \pi/2$, where there is a finite width in the exact excitation continuum, whereas the approximation investigated here has essentially a “pinch point”. This broadening can arise from the interaction between the two spinons. In their seminal work, Ishimura and Shiba [32] emphasised that such continuum of Figure 2.4 arises by interpreting the system’s excitations as roaming domain walls, i.e., spinons. Their work focused on the calculation of dynamic spin structure factors associated with these excitations. As previously
mentioned, such structure factors are of immediate relevance to neutron scattering experiments; they are direct means to test a theoretical model and the assumptions that underpin any such a model. This is strong motivation to compute such quantities, with many theoretical techniques being employed to do so: the utilisation of quantum groups [33], as well as the Bethe Ansatz [34], have furnished exact results, while perturbation techniques have been employed to generate approximate but insightful outcomes [32]. At this juncture, it should be stated that some of the methods of investigation that are utilised here have been inspired by the approach implemented by Ishimura and Shiba [32]. As will be seen in Chapter 4, this involves first order non-degenerate perturbation theory along with recasting the dynamic structure factors in terms of Green functions. This chapter also details how such an approach is generalised to include new interactions, such as an external magnetic field applied transverse to the Ising direction and the Dzyaloshinskii-Moriya interaction; these interactions are discussed in more detail in Chapter 3. From an experimental perspective, there has been much investigation of the two spin-soliton states in compounds which are described by the $XXZ$ model; some prominent examples are: CsCoCl$_3$ [35] CsCoBr$_3$ [36] and TlCoCl$_3$ [25].

2.4 The Spin-1/2 FM $XYZ$ Model

The second model that we shall investigate in detail is the fully anisotropic $XYZ$ model with FM coupling; the corresponding nearest-neighbour Hamiltonian is given by

$$
\mathcal{H}_{XYZ} = -\sum_{i=1}^{N}\left\{ J_x S_i^x S_{i+1}^x + J_y S_i^y S_{i+1}^y + J_z S_i^z S_{i+1}^z \right\}. \tag{2.15}
$$

As in the case of the $XXZ$ model, $J_x$ and $J_y$ are small compared to the Ising coupling and these transverse exchange interactions give rise to quantum fluctuations in the system, with $J_x \leq J_y \leq -|J_z|$. Following on from his work on the six-vertex model and the $XXZ$ model, Baxter [37] utilised the eight-vertex model to produce the ground state energy per site for the $XYZ$ model. In terms of boundary conditions, it is important to note that, while the AFM case utilises an odd number of spin sites with periodic boundary conditions to enforce the presence of one spinon, the FM configuration of the current system dictates that twisted boundary conditions are implemented in order to realise a system catering for just a single excitation [8]: $S_{N+i}^x = S_i^x$, $S_{N+i}^y = -S_i^y$, $S_{N+i}^z = -S_i^z$. 


2.4.1 One spinon states

Following an analogous line of approach as in the previous section, one begins with the two-fold degenerate ground states associated with a pure Ising part

\[ |A\rangle = |↑↑↑↑↑↑↑↑ \cdots ↑↑⟩, \tag{2.16a} \]
\[ |B\rangle = |↓↓↓↓↓↓↓↓ \cdots ↓↓⟩. \tag{2.16b} \]

the dispersion relation associated with a single spinon roaming in a system governed by Equation (2.15) is obtained from the static domain walls with the following kets; \( m \) and \( Q \) detail the same information as before:

\[ |m, Q = +1\rangle = |↓↓↓ \cdots ↓↓ \uparrow \uparrow \uparrow \cdots \uparrow \uparrow⟩, \tag{2.17a} \]
\[ |m, Q = -1\rangle = |↑↑↑ \cdots ↑↑ \downarrow \downarrow \cdots \downarrow \downarrow⟩. \tag{2.17b} \]

Keeping within the Villain approximation [28], the Bloch states that diagonalise \( H_{XYZ} \) have the same form as those expressed in Equation (2.10) (the states \( |m, 1\rangle \) are of course different)

\[ |k\rangle = \frac{1}{\sqrt{2N}} \sum_{m=1}^{2N} e^{ikm} |m, 1\rangle, \tag{2.18} \]

with corresponding dispersion relation [8]

\[ \varepsilon_k = \frac{J_z}{2} + \frac{J_y - J_x}{2} \cos 2k. \tag{2.19} \]

Comparing this with Equation (2.11), one notices a striking feature: the one spinon dispersion exhibits a \( \cos 2k \) dependence for both AFM and FM cases. Such a property is highlighted because, if one would treat the AFM case in isolation, one would be inclined to assume that \( \cos 2k \) term arises from the doubling of the unit cell. However, because the FM case also exhibits this feature, such a claim can be refuted. In fact, it can be argued [38] that this halving of the Brillouin zone is related to the spin-soliton obtaining a Berry phase [39] as it propagates through the lattice; the concept of Berry phase is discussed in Section 2.5. This newly-acquired phase develops in such a way that nearest-neighbour transitions are suppressed through destructive interference, while amplifying next-nearest neighbour transitions. All of this can be related to the inherent chiral nature of these spinon [38]; the investigation of such an inherent property associated with the low lying excitations of interest will be a feature in subsequent sections.
2.4.2 Two spinon states

Turning now to the two spinon case, much work has already been performed on this subspace of excitations for a system governed by $H_{XYZ}$; the excitation spectrum was first detailed by Johnson et al. [40] who demonstrated that this subspace produces an excitation continuum, but also, interestingly, a set of bound states in a certain range of exchange couplings. The dispersion associated with the full continuum for the $XYZ$ model is given as [40]:

$$\varepsilon(k, q) = A_c \left[ \sqrt{\sin^2(q + \frac{k}{2}) + m^2 \cos^2(q + \frac{k}{2})} + \sqrt{\sin^2(q - \frac{k}{2}) + m^2 \cos^2(q - \frac{k}{2})} \right], \quad (2.20)$$

where $k = k_1 + k_2$ is the centre-of-mass momentum, $q = \frac{1}{2}(k_1 - k_2)$ is the relative momentum and the amplitude is $A_c = \frac{1}{2}|J_x| \text{sn}(2\zeta, l) K'(m)/K'(l)$, with $\text{sn}(u, m)$ being a Jacobi elliptic function. The bound states are described by the following [30]:

$$\varepsilon_n(k) = A_n \sqrt{\sin^2(k/2) + a_n^2 \cos^2(k/2)} \sqrt{\sin^2(k/2) + b_n^2 \cos^2(k/2)} \quad (2.21)$$

with $n = \mu/(\pi - \mu)$ indexing the bound states and $\mu = \pi \zeta/K(l)$; the amplitude is given as

$$A_n = |J_x| \frac{K'(m)}{K'(l)} \frac{\text{sn}(2K(l) - 2\zeta, l)}{\text{sn}(y_n, m)}, \quad (2.22)$$
where $a_n = msn(y_n, m)$, and $b_n = sn(y_n, m)$, $y_n = n\lambda'K'(m)/\pi$ and $\lambda' = \tau - \lambda$. An example of this excitation spectrum is given in Figure 2.6. In contrast to the XXZ model with AFM background, the spin-1/2 XYZ FM model has only been realised in a few chemical compounds, such as CoCl$_2$·2H$_2$O [41].

Now that the pertinent models and excitations have been presented, the natural question that arises is what new insight can one add to this area since it has undergone such an exhaustive investigation in the past decades. This leads to the motivation of my work: as hinted previously, the approach by Ishimura and Shiba [32] is generalised in this work to compute the dynamic structure factors in the presence of an external magnetic field applied transverse to the Ising direction, the Dzyaloshinskii-Moriya Interaction (DMI), and the interaction that arises when the presence of neighbouring spin chains is taken into account. Furthermore, the dynamic structure factors for polarised neutrons is computed when these interactions are present. In the case of the XXZ model, such results are compared to experimental polarised neutron scattering data produced at ILL. As one shall see, this particular technique allows one to probe the chiral nature of the excitations of interest; details of of chirality in the context of quantum spin chains are outlined in the following section.

### 2.5 Berry phase

To explain how Berry phase arises and how it aligns with the research presented in this dissertation, one retraces steps back to the original work of Michael Berry [39]. Here, one of the key elements of the existence of such a phase lies with the so-called adiabatic theorem. The essence of such a theorem relates to the states of the system not undergoing any transitions as the system itself evolves in time: if the system is initially in the $m$th eigenstate, it will be found at a later stage in the $m$th eigenstate belonging to the time-evolved Hamiltonian, $\mathcal{H}(t)$.

With this key concept in place, the following content demonstrates how an additional term arises in the phase of the wavefunction associated with the time-evolution of a state when the system is assumed to evolve adiabatically; this additional term is a geometric phase known as Berry phase. Following the arguments presented in [42], we first take a general case where the Hamiltonian depends on a set of parameters denoted by the vector $\mathbf{R} = (R_1, R_2, \ldots)$. These terms are time-dependent, $R_i = R_i(t)$, meaning that the Hamiltonian is also time-dependent and, in the case of a 3D example, the parameters can represent the direction and strength of a magnetic field. Since one can find an instantaneous eigenbasis, $\{|n(\mathbf{R})\rangle\}$, with corresponding eigenvalues, $E_n(\mathbf{R})$ with $\mathbf{R} = \mathbf{R}(t)$, one has

$$H(\mathbf{R})|n(\mathbf{R})\rangle = E_n(\mathbf{R})|n(\mathbf{R})\rangle.$$  \hspace{1cm} (2.23)

Also, according to the adiabatic approximation, the initial eigenstate $|n(\mathbf{R}(0))\rangle$ will evolve com-
pletely into the eigenstate $|n(R(t))\rangle$. It is possible, however, that the state acquires a phase factor; it is this phase that is of interest to us and whose existence was first highlighted by Berry [39]. With this in mind, one can therefore write the most general state as:

$$|\psi(t)\rangle = e^{i\theta(t)} |n(R(t))\rangle.$$  \hspace{1cm} (2.24)

In order to determine this phase, we begin with the time-dependent Schrödinger equation:

$$H(R(t))|\psi(t)\rangle = i\hbar \frac{d}{dt} |\psi(t)\rangle.$$  \hspace{1cm} (2.25)

Using $|\psi(t)\rangle = e^{i\theta(t)} |n(R(t))\rangle$ and the fact that the states $|n\rangle$ are mutual orthonormal, we have

$$E_n(R(t)) - i\hbar \langle n(R(t))|\frac{d}{dt}|n(R(t))\rangle = \hbar \frac{d}{dt} \theta(t).$$  \hspace{1cm} (2.26)

Therefore,

$$\theta(t) = \frac{1}{\hbar} \int_0^t E_n(R(t')) dt' - i \int_0^t \langle n(R(t'))|\frac{d}{dt'}|n(R(t'))\rangle dt'.$$  \hspace{1cm} (2.27)

In the above, the first term on the right-hand side is the usual dynamical phase. However, the second term on the right-hand side is the Berry phase and is a result of the fact that the states $|n(t)\rangle$ and $|n(t + dt)\rangle$, at times $t$ and $t + dt$ are not identical. Explicitly, Berry phase is

$$\gamma_n = i \int_0^t \langle n(R(t'))|\frac{d}{dt'}|n(R(t'))\rangle dt'.$$  \hspace{1cm} (2.28)

To see the geometric aspect of Berry phase, we note that the time dependence need not be explicitly involved:

$$\gamma_n = i \int_0^t \langle n(R(t'))|\nabla_R|n(R(t'))\rangle \frac{dR}{dt'} dt' = i \int \langle n(R)|\nabla_R|n(R)\rangle dR.$$  \hspace{1cm} (2.29)

An important point to note at this stage is that this phase is observable when the parameter vector $R(t)$ is brought back to its starting point, that is, $R(t_f) = R(t_i)$. This means that we have a closed path which $R$ follows in parameter space and that the states $|n(R(t_f))\rangle$ and $|n(R(t_i))\rangle$ interfere with each other. The fact that the Berry phase is observable can be further motivated by reformulating Equation (7.7a) to see the explicit gauge property:

$$\gamma_n = \oint_C A_n(R) \cdot dR,$$  \hspace{1cm} (2.30)
where $C$ is the contour determined by $R(t)$, and

$$A_n(R) = i \langle n(R) | \nabla_R | n(R) \rangle. \quad (2.31)$$

One can therefore interpret $A_n$ as a type of “vector potential” analogous to electron transport in an electromagnetic field; in this sense, Berry phase can be described as an Aharonov-Bohm phase of an artificial electromagnetic field.

Since normalisation of $|n\rangle$ ensures that $\langle n(R) | \nabla_R | n(R) \rangle$ is purely imaginary, one uses this, along with Stokes’ theorem to further manipulate Equation (2.30):

$$\gamma_n = - \text{Im} \int_{\Xi} \nabla_R \cdot \langle n(R) | \nabla_R | n(R) \rangle \cdot d\Sigma,$$

$$= - \text{Im} \int_{\Xi} \langle \nabla_R n(R) | \nabla_R n(R) \rangle \cdot d\Sigma,$$

$$= - \text{Im} \int_{\Xi} \sum_{m \neq n} \langle \nabla_R n(R) | m \rangle \cdot \langle m | \nabla_R n(R) \rangle \cdot d\Sigma, \quad (2.32)$$

where the domain of integration is the surface $\Xi$. Also, with $\mathcal{H}(R) | n(R) \rangle = E_n(R) | n(R) \rangle$, one has

$$E_n \langle m | \nabla_R n \rangle = \langle m | \nabla_R (\mathcal{H} n) \rangle = \langle m | (\nabla_R n) \rangle + E_m \langle m | \nabla_R n \rangle \quad (2.33)$$

for the case of $m \neq n$ and $\langle m | n \rangle = 0$. From the above, one can deduce that

$$\langle m | \nabla_R n \rangle = \frac{\langle m | (\nabla_R \mathcal{H}) | n \rangle}{E_n - E_m}. \quad (2.34)$$

A similar equation arises for $\langle \nabla_R n | m \rangle$. Equation (2.30) can now be written as

$$\gamma_n = - \int_{\Xi} V_n \cdot d\Sigma, \quad (2.35)$$

where

$$V_n = \text{Im} \sum_{m \neq n} \frac{\langle n(R) | \nabla_R \mathcal{H}(R) | m(R) \rangle \cdot \langle m(R) | \nabla_R \mathcal{H}(R) | n(R) \rangle}{(E_m(R) - E_n(R))^2}. \quad (2.36)$$

This above is exploited in the next part of discussion, where the well-known example of computing the Berry phase of a spin-$S$ particle in a time-evolving magnetic field is presented; the Hamiltonian associated with such a system is

$$\mathcal{H}(B) = \mathbf{B} \cdot \mathbf{S}, \quad E_n(B) = Bn, \quad n = -S, -S+1, \ldots, S-1, S. \quad (2.37)$$
where \( B \) is the magnetic field and \( S \) is the vector spin operator. Computing Equation (2.36), one has

\[
V_n = \text{Im} \sum_{m \neq n} \frac{\langle n, S(B)|S|m, S(B)\rangle \wedge \langle m, S(B)|S|n, S(B)\rangle}{B^2(m-n)^2}
\]  

(2.38)

Choosing the instantaneous \( z \)-axis to be parallel to the magnetic field, \( B \), one has the usual relations:

\[
S_z|n, S\rangle = n|n, S\rangle,
\]  

(2.39a)

\[
S^+|n, S\rangle = \sqrt{s(s+1) - n(n+1)}|n+1, S\rangle,
\]  

(2.39b)

\[
S^-|n, S\rangle = \sqrt{s(s+1) - n(n-1)}|n-1, S\rangle
\]  

(2.39c)

Using these relations in Equation (2.38), one finds that

\[
V_n = n \frac{B}{B^2},
\]  

(2.40)

and so

\[
\gamma_n = -\int_{\Xi} n \frac{B}{B^2} \hat{n} \cdot d\Xi.
\]  

(2.41)

One now sees that the Berry phase can be interpreted as the flux through the area denoted by \( \Xi \) of a monopole of strength \( -n \). One can also interpret it as \( n \) times the solid angle that a closed contour \( C \) (bounding the area \( \Xi \)) subtends at \( B = 0 \). The corresponding vector potential of a magnetic monopole is given as (for a unit sphere)

\[
A = \frac{1 - \cos \theta}{\sin \theta} \hat{\phi}
\]  

(2.42)

This particular vector potential is central to investigating the semi-classical system of a ferromagnetic spin chain with easy-plane configuration (in the \( xy \)-plane), which is itself the the main motivation for examining the spin-1/2 ferromagnetic \( XYZ \) model. In the context of Berry phase, the propagation of domain walls in such a system, as shown in Figure 2.7, was first explored by Braun and Loss [8]; they did so by generalising the quantum action to include space dependent spin configurations,

\[
S = is \int_0^\beta d\tau \int \frac{dx}{a} \hat{n} \cdot \mathbf{A} + \int_0^\beta d\tau \mathcal{H},
\]  

(2.43)

Here \( s \) is the spin, \( \beta = 1/k_B T \), \( \tau \) is the imaginary time, \( a \) is the lattice constant, \( \mathbf{n} \) is parameterised as \( \mathbf{n} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta) \) and \( \mathbf{n} \cdot \mathbf{A} = \dot{\phi}(1 - \cos \theta) \). After inserting a domain wall
\( \phi_{DW}(x - X\tau) \) at position \( X(\tau) \), the Berry phase term becomes
\[
is \int_0^\beta d\tau \int dx \frac{1}{a} (1 - \cos \theta) \dot{\phi} = -i \pi s C \int_0^\beta d\tau \dot{X}.
\]
(2.44)
where \( \phi \) is the azimuthal angle and \( C \) is the “chirality”
\[
C = \frac{1}{\pi} \int_{-\infty}^{\infty} dx \, \partial_x \phi.
\]
By noting that
\[
\Delta X = \int_0^\beta d\tau \dot{X},
\]
(2.45)
Equation (2.44) now reads
\[
- i \pi s C \Delta X / a.
\]
(2.46)
In terms of excitations, and conforming with the terminology used in the literature [8], these moving domain walls are referred to as solitons. In previous sections, and in the rest of this dissertation, we use the term spinon; in the context of the discussion of this particular section, soliton, spinon and domain wall may all be used interchangeably. Examples of such solitons are shown in Figure 2.7. In this easy-plane configuration, solitons come in quadruples [43] (Figure 2.8)
\[
\phi_{DW}(x) = -QC\pi/2 + 2 \arctan e^{Cx/\delta}, \quad \theta = \pi/2,
\]
(2.47)
where \( \delta \) is the domain wall width, \( C \) is the chirality and \( Q \) is the “charge”,
\[
Q = \frac{1}{2} \int_{-\infty}^{\infty} dx \, \partial_x (\sin x).
\]
For the case of this easy plane system, \( Q = \pm 1; \) \( C = \pm 1 \) with \( C = +1 \) corresponding to right-handed soliton and \( C = -1 \) corresponding to left-handed soliton, as indicated in Figure 2.7. The chirality is related to the so-called winding number; if we take a mapping \( f \) such that \( f : M \to N \), where \( M \) and \( N \) are manifolds, then the winding number of the mapping counts how many times \( M \) is wrapped around \( N \) under the map \( f \) [44]. By definition, the relationship between the chirality and the winding number is \( C = 2w \), where \( w \) denotes the winding number. In terms of the easy-plane configuration, the four possible realisations of soliton in this easy-plane configuration (looking along the \( z \)-axis) are shown in Figure 2.8, along with the values of the chirality and the winding number.
To reconcile the above discussion with the \( XYZ \) spin-1/2 model, one takes this semi-classical
system to the quantum limit, i.e., the spin is spin-1/2 and the displacement is a single lattice site, \( \Delta X = a \); the phase of Equation (2.46) is now

\[
- \frac{i \pi}{2} C
\]  

(2.48)

This means that as the soliton moves through the lattice, it acquires a Berry phase and this phase can assume a value associated with right-handed or left-handed sense of soliton propagation, depending on whether \( C = +1 \) or \( C = -1 \), respectively. This is effectively what is occurring in the system described by the ferromagnetic \( XYZ \) spin-1/2 that is under investigation in this chapter. More explicitly, the Berry phases for the solitons or spinons at the quantum level are given as, for nearest-neighbour transitions,

\[
e^{+i\pi/2} = +i, \quad \text{right-handed chirality,} \tag{2.49a}
\]
\[
e^{-i\pi/2} = -i, \quad \text{left-handed chirality.} \tag{2.49b}
\]

Graphically, this is depicted in Figure 2.9 where the spin “up” state has two paths to transition to a spin “down” state.

### 2.6 Spinon chirality

One aspect of this research is the investigation of the chiral nature associated with spinons in systems accommodating either one- or two-spinons. As one will see, this property is not readily detectable in spin systems in the context of neutron scattering. Therefore, the challenge is to find some innovative means to unveil this property; the solution involves a combination of polarised neutron scattering and implementing a perturbation into the system, such as an external magnetic field applied transverse to the Ising direction, or the Dzyaloshinskii-Moriya interaction (DMI)
Figure 2.8: Possible realisations of a $\pi$ domain wall in easy plane configuration with the corresponding values of winding number and chirality, denoted by $w$ and $C$, respectively.
The latter is an anisotropic form of the exchange interaction and will be discussed in detail in Chapter 3. The concept of chirality has a classical origin and quite intuitive: a left- and a right-handed structure cannot be superimposed. What is remarkable about this property is its apparent ubiquity: in chemistry, one has chiral molecules or enantiomers, yet chirality can even spontaneously emerge in some 1D quantum spin chains, as has been recently demonstrated experimentally [38]. The existence of chirality in the context of spin chains reflects the fact that the propagation of a spinon can occur either in a right-handed or left-handed fashion. In order to further understand this property, it is illuminating to take a topological perspective [44]: chirality may be defined through the so-called winding number; in mapping from a circle to a circle, this quantity is defined as the number of times the image of the map is wrapped around the circle [47] and is represented by a positive or negative integer. Chirality, in the continuum, may be defined by the following [8]

\[ C = \frac{1}{\pi} \int_{-\infty}^{\infty} dx (\mathbf{n}_s \wedge \partial_x \mathbf{n}_s), \tag{2.50} \]

where the integration variable \( x \) denotes the axis of the chain and \( \mathbf{n}_s(x) \) is the unit vector field along the classical spin direction. Note that (2.50) is twice the winding number of the mapping from a circle to a circle. In the case of the discrete lattice, in the quantum regime, one defines
Figure 2.10: Band minima of dispersion associated with spin-1/2 AFM XXZ model, Equation (2.11), correspond to maximal chirality, with one of the minima associated with left-handed sense of spin soliton propagation, while the other with a right-handed sense; analogous are obtained for spin-1/2 FM XYZ model.

the chiral operator with nearest-neighbour interaction as [8]

\[ C = \sum_{i=1}^{N} S_i \wedge S_{i+1}, \] (2.51)

where periodic boundary conditions are assumed. Specifically for this research, one is interested in the chirality associated with the \(x\)-direction of the spin chain:

\[ C_x = \sum_{i=1}^{N} (S_i \wedge S_{i+1})_x. \] (2.52)

What is quite interesting about Equation (2.51) is that it essentially has the same form as the Hamiltonian associated with the unstaggered DMI; in its nearest-neighbour form, this is given as [45, 46]

\[ \mathcal{H}_D = \sum_i D_{i,i+1} \cdot (S_i \wedge S_{i+1}), \] (2.53)

where \(D_{i,i+1}\) is the so-called Dzyaloshinskii-Moriya vector; assuming this to be constant and directed along the \(x\), the above is
\[ \mathcal{H}_{D_x} = D_x \sum_i^{N} (S_i \wedge S_{i+1})_x, \]

which has the same form as the \( x \)-component of the chiral operator in Equation (2.52). Therefore, one sees the intimate connection between chirality and the Dzyaloshinskii-Moriya interaction; further details are provided in Chapter 3. As has been performed in the previous section, the natural procedure is to uncover the diagonal representation of \( C_x \), or equivalently \( \mathcal{H}_{D_x} \). Remarkably, the eigenbasis turns out to be the set of Bloch states given by Equation (2.10), and so one has

\[
C_x |k\rangle = \sin k |k\rangle, \tag{2.55a}
\]
\[
\mathcal{H}_{D_x} |k\rangle = D_x \sin k |k\rangle. \tag{2.55b}
\]

In terms of expectation values, the maximal chirality occurs at \( k = \pm \pi/2 \), for then \( \langle C_x \rangle = \pm 1 \); the sign dictates the sense of chirality which can be interpreted as a left- or right-handedness of the spinon. This is indicated in Figure 2.10. Plotting the expectation value of the chiral operator, \( \langle k| C_x |k\rangle = \sin k \), as in Figure 2.11, one is led to a significant observation which cannot be understated: within the first Brillouin zone, characterising each momentum state by \( \langle k| C_x |k\rangle \), one can clearly see that a given \( k \) state has a distinct chiral state associated with it. This implies that, even though the dispersions (2.11) and (2.19) possess \( \pi \)-periodicity, one cannot reduce the Brillouin zone, as had been asserted in earlier work on such quantum spin systems [48].
In the next chapter, the discussion concerns that of the a one-spinon system described by the spin-1/2 FM $XYZ$, specifically calculating the dynamic spin structure factors so that one can relate the results to neutron scattering experiments.
Chapter 3

**Spin-1/2 FM XYZ model: spinon scattering and Villain mode**

3.1 Introduction and motivation

Inspired by the work of Braun et al. [38], this chapter focuses on computing, for the first time, the dynamic spin structure factors for the case of spinon scattering in a chain described by the spin-1/2 FM XYZ model. In addition to this, an external magnetic field transverse to the Ising direction is introduced. Such an interaction introduces incommensurable gapless modes and also leads to a finite signal from the off-diagonal structure factor, indicating a chiral response. Similar results arise when the Dzyaloshinskii-Moriya interaction is present. It should be noted that the discussion of dynamic spin structure factors presented in the opening section is not restricted to the FM XYZ case, in which it is embedded here.

3.2 Dynamic Spin Structure Factors

The dynamic spin structure factor is the temporal and spatial Fourier transform of the two-point dynamic spin correlation function, which is itself a way of quantifying the influence of the alteration of the local magnetisation at two different positions at two different times. Therefore, it is highly applicable for investigating dynamic excitations in quantum spin systems. For a lattice with \( N \) sites, the dynamic spin structure factor is given as

\[
S^{\alpha\beta}(Q, \omega) = \frac{1}{2\pi\hbar} \int dt \ e^{i\omega t} \frac{1}{N} \sum_{i,j} \langle S_i^{\alpha}(t)S_j^{\beta}(0) \rangle e^{iQ \cdot (R_i - R_j)},
\]

(3.1)

with \( \alpha, \beta = x, y, z \), and \( R_n \) denoting the position vector of the \( n^{th} \) magnetic site within the lattice, assumed to be without a basis. In the context of neutron scattering, \( Q \) represents the
momentum transfer that the probing neutron imparts on the system, while $\omega$ is the energy transfer. For convenience, the dynamic spin structure factor is now referred to as simply the dynamic structure factor (DSF). What makes the DSF highly relevant to neutron scattering experiments is its direct relationship with the partial differential scattering cross section. This quantity is defined as the ratio of the scattered neutron flux in the energy range $E'$ and $E' + dE'$ scattered into a solid angle $d\Omega$ to that of incident neutron flux [49]. Therefore, it represents the probability that such a scattering event takes place and, experimentally, this probability corresponds to the scattered neutron intensity measured by a detector or a bank of detectors. For more details on the partial differential scattering cross section, one should refer to Appendix B where the derivation of Equation (3.2) below is also presented. For purely magnetic scattering, the relationship between the partial differential cross section and the dynamic spin structure factor is, for $T = 0$ K,

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{\kappa_f}{\kappa_i} \left( \frac{2\pi \hbar^2 r_0}{m_n} \right)^2 \frac{1}{2} \left| g F(Q) \right|^2 \sum_{\alpha,\beta} (\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta) S^{\alpha\beta}(Q, \omega), \quad (3.2)$$

where $S^{\alpha\beta}(Q, \omega)$ is given in Equation (3.1) above; $r_0 = \gamma e^2/m_e c^2 = -0.54 \times 10^{-12}$ cm, with $\gamma = -1.91$ being the gyromagnetic ratio of the neutron; $\hat{Q}_\alpha = Q_\alpha/||Q||$; the mass of the neutron is denoted by $m_n$; $\kappa_i$ and $\kappa_f$ are the initial wavevector and final wavevector of the neutron, respectively; $F(Q)$ is the form factor which is the Fourier transform of the magnetisation of an electron distribution associated with a given magnetic ion (details of this quantity are expanded in Appendix C); finally, $(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta)$ is referred to as the polarisation factor and encapsulates an important aspect of neutron scattering events: the probing neutrons can only couple to the system’s spin fluctuations perpendicular to $Q$.

In addition to this, Equation (3.2) clearly demonstrates the link between the theoretically computed DSF on the right and the experimentally measured cross section on the left. As a further note, the calculation of DSFs, represented by Equation (3.1), involves quantities which pertain solely to the unperturbed spin chain, while the actual scattering experiment, by its very nature, pushes the system out of equilibrium. This is the essence of the Fluctuation-Dissipation Theorem [50]: the non-equilibrium perturbation of a system (here, the scattering by the neutron) is essentially determined by the fluctuations of the system in the equilibrium state (due to the presence of magnetic excitations). It should be noted at this stage that while Equation (3.2) deals specifically with unpolarised neutron scattering, which is the most common type of probe utilised for the investigation of magnetic structures and magnetic excitations, it is *polarised* inelastic neutron scattering which is of interest in the context of this research: polarised neutrons couple to non-collinear magnetic configurations [51] and, therefore, performing such a technique on a magnetic system not only details the dispersion relation of the excitations under investigation, but also informs about the chiral nature of these entities. For example, this technique has been used in recent years to indeed unveil spinon chirality in Ising-like one-dimensional spin chains [38]. One
challenge associated with the use of polarised neutrons, however, is that a lower incident flux is produced and thus, from the experimental point of view, one needs longer counting time in order to achieve statistics comparable to that of unpolarised neutrons. Further information on polarised neutron scattering is provided in Appendix B, where one will also find the derivation of the form of the partial differential cross section most relevant to the discussion presented here; this is given by

\[ \frac{d^2\sigma}{d\Omega dE'} \propto \int_{-\infty}^{\infty} dt \ e^{-i\omega t} \{ (S_-^\perp \cdot S_\perp^\perp(t)) \pm i\mathbf{P} \cdot \langle S_-^\perp \wedge S_\perp^\perp(t) \rangle \}, \]  

(3.3)

with \( \mathbf{P} \) being the polarisation vector; the polarisation orientation that will be of interest throughout this dissertation is along the \( x \)-direction, i.e., \( \mathbf{P} = \pm \hat{x} \). Also, \( S_\perp^\perp(t) = \hat{Q} \wedge (S_Q(t) \wedge \hat{Q}) \) and is present to emphasise the sensitivity of the neutron to spin fluctuations perpendicular to \( \hat{Q} \).

Now that the importance of the dynamical spin structure factors has been emphasised, particular attention is given to the following DSFs: \( S_{xx}(Q, \omega) \), \( S_{yy}(Q, \omega) \), \( S_{zz}(Q, \omega) \), \( S_{yz}(Q, \omega) \)-\( S_{zy}(Q, \omega) \). The diagonal DSF \( S_{xx}(Q, \omega) \) is known as the magnon or spin-wave response and informs one about the propensity for spin flips to occur in the chain; \( S_{yy}(Q, \omega) \) issues similar information; \( S_{zz}(Q, \omega) \) is commonly referred to as the longitudinal response and pertains to how the length of the spin varies along the chain, and in the these Ising-like systems of interest, it indicates how the magnetic ordering in the system fluctuates in the presence of magnetic excitations; \( S_{yz}(Q, \omega) \)-\( S_{zy}(Q, \omega) \) is intimately connected with the polarised term in Equation (3.3) and furnishes information regarding the chiral nature of the spinons; this explicit connection is outlined in the Appendix B. With this framework now established, the subsequent discussion is broken into two different categories: the first category deals with spinon scattering, while the second concerns spinon pair creation; spinon scattering is discussed in the next section.

### 3.3 Spinon Scattering and Villain mode

In the context of neutron scattering, spinon scattering pertains to the investigation of a single roaming domain wall described by the kets presented in Chapter 2

\[ |k, Q\rangle = \frac{1}{\sqrt{N}} \sum_m e^{ikm} |m, Q\rangle \]  

(3.4)

with \( k \) being the momentum of the spinon and \( m \) labelling the point in the lattice after which the domain wall is introduced; summation is over all lattice sites. A salient point to note for this case is that the one spinon dispersion is not directly accessible by neutron scattering techniques: a transition from the system’s ground state is not permitted as this would entail a simultaneous
\( \pi \)-rotation of a large number of spins. In addition to this, from a general point of view, an incident neutron cannot exchange spin-1/2 with the system as this would violate spin conservation. Again, this implies that the one-spinon dispersion cannot be probed directly. From an experimental perspective, cold neutrons are used to induce spinon scattering; this corresponds to the energy, \( E_i \), of the incident neutrons being in the range \( 0 < E_i < (J_y - J_x) \), i.e., the neutrons can at most impart an energy equal to that of the dispersion bandwidth. Therefore, it is only through transitions within this band, described by the dispersion relation given in Chapter 2,

\[
\varepsilon_k = \frac{J_z}{2} + J_t \cos 2k, \quad \text{for spin-1/2 AFM XXZ model,} \tag{3.5}
\]

or

\[
\varepsilon_k = \frac{J_z}{2} + \frac{J_y - J_x}{2} \cos 2k, \quad \text{for spin-1/2 FM XYZ model,} \tag{3.6}
\]

that one can hope to gain insight about these excitations; the neutron’s transfer of energy and momentum effectively scatters the spinon within the band, hence the term spinon scattering; Figure 3.1 illustrates this. Since the probing neutrons cannot introduce a single spinon into the system, it is through the presence of thermal fluctuations that the one-spinon band is populated: the coupling of the system to a heat bath induces a spontaneous flipping of a spin cluster, leading to the introduction of two domain walls which are subsequently delocalised by the transverse exchange interaction. From the experimental perspective, the thermal regime is such that the number of spinons introduced via this process is dilute, leading to a one-spinon model as a good approximation to what is actually occurring in the magnetic compound. Since one is dealing
with transitions, it is not the actual dispersion relation that is of interest, but rather the difference between the initial one-spinon energy, with momentum \( k_i = k \) and the final energy with momentum \( k_f = k - q \), with \( q \) being the momentum transfer along the chain within the first Brillouin zone:

\[
\omega(k) = \varepsilon_{k-q} - \varepsilon_k = 2A \sin q \sin(2k - q),
\]

(3.7)

where \( A = J_t \) or \( A = (J_y - J_x)/2 \), depending on whether the system is governed by \( XXZ \) AFM model or \( XYZ \) FM model, respectively. Since transitions for a given momentum transfer are degenerate, there is correspondingly a continuous range of energies; the result is that a continuum exists within a range of the \( q\omega \)-plane depicted in Figure 3.2. The extreme values of Equation (3.7) occur at \( \pm \Omega_q \) [28], with

\[
\Omega_q = 2A |\sin q|,
\]

(3.8)

and represent boundaries of the continuum. As one will see, the DSFs possess a characteristic square root singularity for this scattering process at such boundaries. This singularity in scattering intensity is commonly referred to as the “Villain mode”, with Villain [28] being the first to highlight the existence of thermally activated excitations in a system governed by the spin-1/2 \( XXZ \) AFM model. In an actual neutron scattering experiment, this singularity is a sharp peak with finite intensity due to the finite lifetime of the excitations under investigation.
With these preliminary details now introduced the calculations and results of the dynamic spin structure factors for the one-spinon case in a system described by the spin-1/2 FM XYZ model are presented.

### 3.4 Calculation of dynamic spin structure factors for spin-1/2 FM XYZ model for spinon scattering

As will be highlighted, much work has been performed on the AFM case of spinon scattering, both theoretically and experimentally. However, the theoretical examination of ferromagnetic quantum spin chains has been largely untouched in the context of neutron scattering; from the experimental perspective, few investigations have taken place [41], with recent endeavours indicating the technological promise of exploiting such systems at the microscopic scale [52, 53]. Therefore, understanding the fundamental aspects of these particular magnetic systems at the quantum level is a worthwhile aim. As before, the spin-1/2 FM XYZ Hamiltonian reads

\[
\mathcal{H}_{XYZ} = -\sum_{i=1}^{N} \{ J_x S^x_i S^x_{i+1} + J_y S^y_i S^y_{i+1} + J_z S^z_i S^z_{i+1} \},
\]  

(3.9)

In order to investigate the spinon scattering in spin chains governed by such interactions, one utilises the key components for theoretically examining such processes, the dynamic spin structure factors. For this part of the discussion, Equation (3.1) is recast to explicitly take into account thermal fluctuations, with \( Q \) being replaced by \( q \) to emphasise that interest is on the first Brillouin zone,

\[
S^{\alpha\beta}(q, \omega) = \sum_{k, k'} e^{-\epsilon_{k}/k_B T} Z^{-1} \langle k, Q | S^\alpha_{-q} | k', Q' \rangle \langle k', Q' | S^\beta_q | k, Q \rangle \delta(\omega - \epsilon_k - q + \epsilon_k),
\]  

(3.10)

where \( Z \) is the partition function; \( k_B \) being the Boltzmann constant; \( \epsilon_k \) is the dispersion relation (3.6); \( |k, Q\rangle \) are the Bloch states which approximately diagonalise \( \mathcal{H}_{XYZ} \) in the Ising limit, with \( k \) being the initial momentum and \( k' \) being the final momentum; \( S^\alpha_q \) being the Fourier transform of the spin operators defined as

\[
S^\alpha_q = \frac{1}{\sqrt{N}} \sum_{n=1}^{N} e^{-i q n} S^\alpha_n.
\]  

(3.11)
To compute the various DSFs of interest, the natural starting point for this investigation lies with the kets introduced in the previous chapter:

\[ |m, Q = +1⟩ = |↓↓↓ \cdots ↓↓⟩_m | \uparrow \uparrow \uparrow \cdots \uparrow⟩, \tag{3.12a} \]

\[ |m, Q = -1⟩ = |↑↑↑ \cdots ↑↑⟩_m | ↓↓↓ \cdots ↓↓⟩. \tag{3.12b} \]

The application of the various spin operators on these states produces the following results:

\[ S_n^x |m, Q⟩ = \frac{1}{2} \left[ \delta_{Q,+1} \left\{ \delta_{n,m} |m - 1, Q⟩ + \delta_{n,m+1} |m + 1, Q⟩ \right\} + \delta_{Q,-1} \left\{ \delta_{n,m+1} |m + 1, Q⟩ + \delta_{n,m} |m - 1, Q⟩ \right\} \right], \tag{3.13a} \]

\[ S_n^y |m, Q⟩ = \frac{1}{2i} \left[ \delta_{Q,+1} \left\{ \delta_{n,m} |m - 1, Q⟩ - \delta_{n,m+1} |m + 1, Q⟩ \right\} + \delta_{Q,-1} \left\{ \delta_{n,m+1} |m + 1, Q⟩ - \delta_{n,m} |m - 1, Q⟩ \right\} \right], \tag{3.13b} \]

\[ S_n^z |m, Q⟩ = (\delta_{Q,+1} - \delta_{Q,-1}) \left[ \frac{1}{2} - \Theta(m - n + \varepsilon) \right] |m, Q⟩, \tag{3.13c} \]

where \(0 < \varepsilon < 1\). As expected, \(S_n^x\) and \(S_n^y\), mobilise the domain walls, while \(S_n^z\) returns an eigenvalue. Using the above, in conjunction with Equation (3.11), the relevant matrix elements required to construct the DSF in Equation (3.10) are

\[ \langle k', Q'|S_q^x |k, Q⟩ = \frac{1}{2\sqrt{N}} (e^{+ik'} + e^{-ik'}) \frac{1}{N} \sum_{m=1}^{N} e^{i(k-k'-q)m} \tag{3.14a} \]

\[ \langle k', Q'|S_q^y |k, Q⟩ = \frac{1}{2i\sqrt{N}} (\delta_{Q,+1} - \delta_{Q,-1}) (e^{+ik'} - e^{-ik'}) \frac{1}{N} \sum_{m=1}^{N} e^{i(k-k'-q)m} \tag{3.14b} \]

\[ \langle k', Q'|S_q^z |k, Q⟩ = \frac{1}{\sqrt{N}} (\delta_{Q,-1} - \delta_{Q,+1}) \frac{1}{N} \sum_{m=1}^{N} e^{i(k-k')m} \left( \sum_{n=1}^{m} e^{-iqn} - \frac{1}{2} \sum_{n=1}^{N} e^{-iqn} \right) \tag{3.14c} \]
where \( |k, Q\rangle \) is the initial spinons state and \( |k', Q\rangle \) is the final state. Utilising appropriate combinations of the above in Equation (3.10), one obtains

\[
S^{xx}(q, \omega) = \frac{1}{N} \sum_k e^{-\beta \varepsilon_k} \cos^2\left(k - \frac{q}{2}\right) \delta(\omega - \varepsilon_{k-q} + \varepsilon_k), \tag{3.15a}
\]

\[
S^{yy}(q, \omega) = \frac{1}{N} \sum_k e^{-\beta \varepsilon_k} \sin^2\left(k - \frac{q}{2}\right) \delta(\omega - \varepsilon_{k-q} + \varepsilon_k), \tag{3.15b}
\]

\[
S^{zz}(q, \omega) = \frac{1}{N} \sum_k e^{-\beta \varepsilon_k} \frac{1}{4 \sin^2 q/2} \delta(\omega - \varepsilon_{k-q} + \varepsilon_k), \tag{3.15c}
\]

\[
S^{yz}(q, \omega) - S^{zy}(q, \omega) = -i \frac{1}{N} \sum_k e^{-\beta \varepsilon_k} \frac{\sin(k - q/2)}{\sin q/2} \delta(\omega - \varepsilon_{k-q} + \varepsilon_k), \tag{3.15d}
\]

now with \( \beta = 1/k_B T \). In the thermodynamic limit \( \frac{1}{N} \sum_k \rightarrow \int_{-\pi}^{\pi} dk \), i.e., the domain of integration is the first Brillouin zone, the above equations become

\[
S^{xx}(q, \omega) = \int_{-\pi}^{\pi} dk \frac{e^{-\beta \varepsilon_k}}{z} \cos^2\left(k - \frac{q}{2}\right) \delta(\omega - \varepsilon_{k-q} + \varepsilon_k), \tag{3.16a}
\]

\[
S^{yy}(q, \omega) = \int_{-\pi}^{\pi} dk \frac{e^{-\beta \varepsilon_k}}{z} \sin^2\left(k - \frac{q}{2}\right) \delta(\omega - \varepsilon_{k-q} + \varepsilon_k), \tag{3.16b}
\]

\[
S^{zz}(q, \omega) = \int_{-\pi}^{\pi} dk \frac{e^{-\beta \varepsilon_k}}{z} \frac{1}{4 \sin^2 q/2} \delta(\omega - \varepsilon_{k-q} + \varepsilon_k), \tag{3.16c}
\]

\[
S^{yz}(q, \omega) - S^{zy}(q, \omega) = -i \int_{-\pi}^{\pi} dk \frac{e^{-\beta \varepsilon_k}}{z} \frac{\sin(k - q/2)}{\sin q/2} \delta(\omega - \varepsilon_{k-q} + \varepsilon_k). \tag{3.16d}
\]

As can be seen from Equations (3.16), the only contributions to the integrals occur at \( \omega = \varepsilon_{k-q} - \varepsilon_k \), which coincides with Equation (3.7). In order to evaluate Equations (3.16) fully, one must change the variable of the delta function to \( k \); this is achieved by employing the following identity:

\[
\delta(\omega - f(k)) = \sum_i \frac{1}{|f'(k_i)|} \delta(k - k_i), \tag{3.17}
\]

where \( f(k) = (J_y - J_x) \sin q \sin(2k - q) \), and \( k_i \) are such that \( f(k_i) = \omega \). Setting \( \Lambda_q \equiv (J_y - J_x) \sin q \), the possible solutions of

\[
\sin(2k - q) = \frac{\omega}{\Lambda_q} \tag{3.18}
\]
are

\[ k_1 = \frac{1}{2} \arcsin\left( \frac{\omega}{\Lambda q} \right) + \nu \pi + \frac{1}{2} q, \quad (3.19a) \]
\[ k_2 = -\frac{1}{2} \arcsin\left( \frac{\omega}{\Lambda q} \right) + \pi (\nu + \frac{1}{2}) + \frac{1}{2} q, \quad (3.19b) \]

with \( \nu \in \mathbb{Z} \). Then, using the above in conjunction with Equation (3.17) in Equations (3.16), one obtains for the diagonal dynamic structure factors

\[
S_{xx}(q, \omega) = \frac{1}{2z} e^{-\beta \frac{1}{2} \frac{1}{2} \cos q} \cosh \left( \frac{\beta}{2} \sqrt{\Lambda^2_q - \omega^2} \right) \sinh \left( \frac{\beta}{2} \sqrt{\Lambda^2_q - \omega^2 \cot q} \right) \left| \Lambda q \right|^{-1} \]  

(3.20a)

\[
S_{yy}(q, \omega) = \frac{1}{2z} e^{-\beta \frac{1}{2} \frac{1}{2} \cos q} \sinh \left( \frac{\beta}{2} \sqrt{\Lambda^2_q - \omega^2} \right) \sin \left( \frac{\beta}{2} \sqrt{\Lambda^2_q - \omega^2 \cot q} \right) \left| \Lambda q \right|^{-1} \]  

(3.20b)

\[
S_{zz}(q, \omega) = \frac{1}{2z} e^{-\beta \frac{1}{2} \frac{1}{2} \cos q} \sin \left( \frac{\beta}{2} \sqrt{\Lambda^2_q - \omega^2} \right) \]  

(3.20c)

As is evident from above, the DSFs possess a square singularity at \( \omega = \pm \Lambda_q \), which is a characteristic feature associated with these particular spin systems; there is also a singularity occurring at \( |\Lambda_q| \). These singularities coincide with Equation (3.8), for \( A = (J_y - J_x) / 2 \). However, in order to take into account the finite lifetime of the excitations, Equations (3.21) are convolved with a Lorentzian; for convenience, it is the equations below which are used to produce the results that follow. The new equations are

\[
S_{xx}(q, \omega) = \frac{1}{N} \sum_k e^{-\beta \epsilon_k} \cos^2(k - q/2) \frac{1}{\pi} \frac{\gamma}{(\omega + \epsilon_k - \epsilon_{k-q})^2 + \gamma^2}, \quad (3.21a) \]
\[
S_{yy}(q, \omega) = \frac{1}{N} \sum_k e^{-\beta \epsilon_k} \sin^2(k - q/2) \frac{1}{\pi} \frac{\gamma}{(\omega + \epsilon_k - \epsilon_{k-q})^2 + \gamma^2}, \quad (3.21b) \]
\[
S_{zz}(q, \omega) = \frac{1}{N} \sum_k e^{-\beta \epsilon_k} \frac{1}{4 \sin^2 q/2} \frac{1}{\pi} \frac{\gamma}{(\omega + \epsilon_k - \epsilon_{k-q})^2 + \gamma^2}, \quad (3.21c) \]
\[
S_{yz}(q, \omega) - S_{zy}(q, \omega) = \frac{1}{N} \sum_k e^{-\beta \epsilon_k} \frac{1}{i \sin q/2} \frac{1}{\pi} \frac{\gamma}{(\omega + \epsilon_k - \epsilon_{k-q})^2 + \gamma^2}, \quad (3.21d) \]

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where $\gamma$ is the parameter dictating the value for the half-width at half-maximum (HWHM). Turning now to the graphical representation of the results, we will first present the results with Ising and transverse exchange only. This implies that $\varepsilon_k$ in the above equations takes the form given in Equation (3.6); all results shown are produced for $T = 40$ K, which is a typical temperature at which experiments are performed.

Figure 3.3: Results for $S^{xx}(q,\omega)$ for $q = \pi/2$ for spinon scattering 1D spin system described by the spin-1/2 FM $XYZ$ model; $\gamma = 0.1$ meV, $J_z = 13.3$ meV, $J_x = 1.0$ meV, $J_y = 3.0$ meV. The unequal intensity of the peaks is a consequence of the principle of detailed balance. The intensity is given in absolute units of meV$^{-1}$ per formula unit.

The plot shown in Figure 3.3 is produced using Equation (3.21a) for $q = \pi/2$. The most noticeable features are the two peaks which occur at $\omega = \pm \Lambda_q$, representing the Villain mode. In physical terms, for $\omega > 0$, the neutron loses energy in the scattering process and the spinon in the chain transitions to a higher energy state within the band, as indicated by the red arrow in Figure 3.1. For the case of $\omega < 0$, the probing neutron gains energy and so the elementary excitation is demoted from a high energy state to a lower one, corresponding to the orange arrow in Figure 3.1. However, because the excitation in question is thermally nucleated, it is more likely to occupy the low energy states around the band minima, implying that a promotion to higher energy due to the neutron is more probable than that of a demotion. This higher probability of occurrence is reflected in the higher intensity of $\omega > 0$ compared to that of $\omega < 0$. Formally, this is dictated by the Boltzmann factor in (3.21a) and is a manifestation of the principle of detailed balance [51].

In general, the dynamic spin structure factor of interest is $S^{xx}(q,\omega)$ as this represents the spin-wave response and provides information regarding the propagation of a spin-flip process occurring in the chain. While this is often the most dominant response, this is different for the scattering of thermally activated spinons considered here: it is in fact $S^{zz}(q,\omega)$ that is the dominant term, as illustrated in Figure 3.4. This particular dynamic structure factor provides the longitudinal response and reflects how the magnetisation varies along the chain. In this model containing a
single spinon, it is this single excitation that is responsible for fluctuations in the $z$-component of the spin as it propagates through the chain.

![Figure 3.4: Comparison of $S^{zz}(q,\omega)$ (solid blue line) and $S^{xx}(q,\omega)$ (dashed black line) in the presence of only exchange interactions, with $q = \pi/2$, $\gamma = 0.1$ meV, $J_z = 13.3$ meV, $J_x = 1.0$ meV, $J_y = 3.0$ meV. This demonstrates the dominance of the longitudinal response compared to that of the transverse. The intensity is given in absolute units of meV$^{-1}$ per formula unit.](image)

The three-dimensional plots of the diagonal structure factors are shown in Figure 3.5. All of the plots possess the same support as highlighted by the corresponding density plots, mimicking somewhat trivial features of the cross section, namely the elastic peaks; the peaks at $q = 0$ and $q = \pi$ correspond to FM and AFM (short-range) ordering, respectively. The fact that these are the dominant peaks is a reflection that the elementary excitations in question are modelled as being dilute. Note that the peak at $q = 0$ in $S^{zz}(q,\omega)$ is truncated so as to amplify the rest of the features in the plot. The peak itself is a manifestation of the singularity in Equation (3.21c), but is finite due to the convolution with a Lorentzian. Since $S^{zz}(q,\omega)$ reflects the fluctuations of the magnetic ordering in this case of Ising-like spin chains, it is understandable that such a dominant peak is present: the dilute nature of the excitations permits the FM ordering to still prevail. While few neutron scattering experiments have been performed for FM systems, it should be noted that these quasielastic magnetic responses at $q = 0$ are experimentally difficult to detect because of the presence of the more dominant nuclear Bragg peaks. Furthermore, while the FM ordering is induced by the dominant Ising exchange interaction in Equation (3.9), the weaker transverse terms are responsible for introducing quantum fluctuations which manifest themselves in the form of spin flips; it is this spontaneous spin flipping which introduces local AFM ordering giving the weaker peak at $q = \pi$. The most interesting features of these plots, however, are those which occur away from $q = 0$ and $q = \pi$: the boundary of the continuum coincides with the most likely transitions, reflected in the presence of peaks as a function of energy for fixed $q$: the peak
associated with positive energy transfer corresponds to the transition of the spinon from a low energy state within the one-spinon band to one of higher energy; this peak is specifically referred to as the Villain mode. On the other hand, the peak with negative energy transfer is associated with transitions from high energy to one of lower energy.

Progressing the discussion towards the antisymmetric linear combination of dynamic structure factors of Equation (3.21d), $S^{yz}(q, \omega) - S^{zy}(q, \omega)$, the result, as presented in Figure 3.14(a), is a rather uninteresting zero signal. Although there is clearly a singularity at $q = 0$ in Equation 3.21d, one can demonstrate, using L’Hôpital’s rule [54], that $\lim_{q \to 0} S^{yz}(q, \omega) - S^{zy}(q, \omega) = 0$. Superficially, this result seems unimportant, yet it is still a worthwhile venture to uncover why one obtains this zero signal. From Appendix B, one is aware that $S^{yz}(q, \omega) - S^{zy}(q, \omega)$ is intimately connected to the polarised term in Equation (3.3). In an actual polarised neutron scattering experiment, the quantity that is measured is the difference between the two possible orientations of the incident polarised neutrons. With this in mind, one utilises Equation (B.42) in Equation (B.41) of Appendix B to obtain

$$I_+ - I_- \propto \frac{2(1 - \hat{Q}_z^2)}{N \sin q/2} \sum_k e^{-\beta \varepsilon_k} \sin \left( \frac{k + k'}{2} \right) \delta(\omega - \varepsilon_{k'} + \varepsilon_k), \quad (3.22)$$

where we defined $I_\pm = \frac{\partial^2 \sigma}{\partial \Omega \partial E_\pm}$, $\hat{Q}_z$ is the component of the wavevector transfer unit-vector along the spin chain, and $k' = k - q$. The sine term in the above, as one shall see, is the pertinent term for explaining why such a featureless plot in Figure 3.14(a) arises. Figure 3.7 represents an example that encapsulates what is occurring with the various scattering processes taking place within the band. As indicated in this diagram, it is helpful to fix the parameters of momentum transfer, $q$, and energy transfer, $\omega$, as is typically performed in a scattering experiment. In Figure 3.7, there are two scattering processes to take note of: the transition from orange state 1 to orange to state 2, and from purple state 1 to purple state 2. Of course these processes can happen in reverse, but then the energy and momentum transfers are negative. Because of symmetry, the same argument can be applied to the pair of states $A$ and $B$, and $C$ and $D$. Therefore, there are a total of eight processes occurring in this example. Focusing attention on those of just 1 and 2 (both orange and purple), and noting that one sums over all $k$ states permissible for fixed $q$ and $\omega$ values in Equation (3.22), one sees that due to the $\pi$-periodic nature of the band, and that $\sin(x + \pi) = -\sin x$, Equation (3.22) will produce summands that will cancel each other, resulting in a vanishing signal. From the physical point of view, since spinon chirality is intimately related to $I_+ - I_-$, this result indicates that chirality is hidden. However, as is demonstrated in the next section, this is not the case when even an external transverse magnetic field is present.

An interesting side remark to conclude this section is that as $k' \to k$, the sine term in Equation (3.22) approaches $\sin k$, the eigenvalue of the chiral operator in $x$-direction, given by Equation...
Figure 3.5: Diagonal dynamic spin structure factors for spin-1/2 FM $XYZ$ model for spinon scattering with only exchange interactions present. $J_x = 13.3 \text{ meV}$, $J_y = 3.0 \text{ meV}$, $J_z = 1.0 \text{ meV}$. The intensity is given in absolute units of $\text{meV}^{-1}$ per formula unit.
Figure 3.6: (a) $S^{yz}(q,\omega) - S^{zy}(q,\omega)$ produces zero signal when only the Ising and transverse interactions are present. $J_z = 13.3$ meV, $J_y = 3.0$ meV, $J_x = 1.0$ meV. (b) Non-zero response in $S^{yz}(q,\omega) - S^{zy}(q,\omega)$ when $B_x = 3$ T, otherwise same parameters as in (a). The intensity is given in absolute units of meV$^{-1}$ per formula unit.

(2.55a) of Chapter 2.

Figure 3.7: One spinon band: in the absence of Dzyaloshinskii-Moriya interaction and transverse magnetic field. The $\pi$-periodicity of the band induces a cancellation of scattering processes for fixed $q$ and $\omega$.

3.5 One-dimensional FM Ising-like spin chain in the presence of a transverse field

As mentioned previously, additional interactions are examined for the spin-$1/2$ FM $XYZ$ catering for a single spinon, one of which is that of an external magnetic field applied transverse to the Ising direction. In this section the effects of such an interaction are delineated. The actual motivation for applying a transverse magnetic field stems directly from the work of Braun et al. [38], where
the introduction of this external perturbation aided in the detection of spinon chirality in a system described by the spin-1/2 AFM $XXZ$ model. Here, for the first time, the theoretical results of the dynamic spin structure factors, for the spin-1/2 FM $XYZ$ model with external field, are presented. Unlike the case in the previous section, where the only interactions in the system were the Ising and transverse exchange interactions, the presence of a transverse magnetic field is responsible for producing a finite response in $S^{yz}(q,\omega) - S^{yz}(q,\omega)$; this is a signature of the chiral nature of spinons, as highlighted by the comparison of zero and non-zero transverse field in Figure 3.6. A starting point to quantify the discussion lies with the Hamiltonian describing the magnetic applied field transverse to the Ising direction,

$$\mathcal{H}_{B_x} = g\mu_B B_x \sum_{i=1}^{N} S^x_i,$$

(3.23)

where $\mu_B$ is the Bohr magneton, and $g$ is the electron spin $g$ factor, and $B_x$ is the magnitude of the external transverse magnetic field. Applying this to the states (3.12), one obtains

$$\mathcal{H}_{B_x}|m, Q\rangle = \frac{g\mu_B B_x}{2}(|m + 1, Q\rangle + |m - 1, Q\rangle)$$

(3.24)

Therefore, one immediately sees that the additional quantum fluctuations induced by the presence of a transverse magnetic field delocalises the static domain walls. Furthermore, the presence of this transverse magnetic field leads to the fact that $S_{tot}^z$ is no longer a conserved quantity. To obtain the eigenenergies associated with this interaction, the Bloch states (3.4) are employed to
diagonalise $\mathcal{H}_{B_x}$:

$$\mathcal{H}_{B_x} |k, Q\rangle = g\mu_B B_x \cos k |k, Q\rangle.$$ (3.25)

The full Hamiltonian now reads $\mathcal{H} = \mathcal{H}_{XYZ} + \mathcal{H}_{B_x}$, with corresponding dispersion relation

$$\varepsilon_k = \frac{J_z}{2} + \frac{J_y - J_x}{2} \cos 2k + g\mu_B B_x \cos k.$$ (3.26)

Also, in terms of the one spinon band, the effect of this field is to deform the band, as in Figure 3.8. However, its influence is felt most prominently in terms of removing the $\pi$-periodicity that is present when only the Ising and transverse exchange interactions are involved; the dispersion relation above now simply becomes $2\pi$-periodic. Keeping Equation (3.22) in mind, and with $B_x \neq 0$, the scattering processes in Figure 3.9 with the same $q$ and $\omega$ values involve $k$-values which, unlike the case for $B_x = 0$ T, are not related by $n\pi$, with $n$ odd. With $q$ and $\omega$ fixed, this means that the sine term in Equation (3.22) does not, in general, induce a cancellation among the summands and so $I_+ - I_-$ produces a finite response; this indicates that there is indeed a chiral property associated with the spinons and provides the main motivation for considering the effects of the transverse magnetic field. The presence of the transverse magnetic field is also responsible for the introduction of two additional peaks in the diagonal dynamic spin structure factors, $S^{\alpha\alpha}(q, \omega)$, $\alpha = x, y, z$, as illustrated in Figure 3.11. This is due to deformation of the band, induced by the transverse magnetic field, allowing for two different transition energies for a given momentum transfer, $q$; such a scenario is depicted in Figure 3.10. It should be emphasised that this is not a
Zeeman splitting since these additional peaks arise out of the band being deformed, not split. A simple Zeeman splitting in the context of magnetic excitations would correspond to the “splitting” of the original band into new bands; this has been demonstrated, for example, for a dimerized antiferromagnet [55]: here, the magnetic excitations are referred to triplons and are bosonic in nature. With non-zero magnetic field, the dispersion relation transforms into three bands since triplons possess $S = 1$, analogous to a Zeeman splitting of a $S = 1$ particle in an external magnetic field. To further embellish the notion that the additional peaks that one observes are not a result of Zeeman splitting, one examines past investigations for the case of the spin-$1/2$ AFM $XXZ$ model, in the particular the work of Devreux and Boucher [48]; they claim that the Brillouin zone can be reduced by assuming that the momentum states characterised by $k$ and $k - \pi$ are equivalent when the applied transverse magnetic field is zero. However, in light of the discussion surrounding chirality in Chapter 2, the momentum states with $k \in (-\pi, \pi]$ are unique in terms of the chiral expectation value,

$$\langle C_x \rangle = \sin k.$$

(3.27)

Therefore, the assumption that the Brillouin zone can be reduced is a false one, and subsequently one cannot interpret the additional peaks as a manifestation of Zeeman splitting, akin to that observed in reference [55]; this argument can be transferred to the spin-$1/2$ FM $XYZ$ model dealt with here.

Figure 3.13 and Figure 3.14 display, for non-zero transverse field, the 3D and density plots of the diagonal and off-diagonal dynamic spin structure factors, respectively. Once again, the plots of $S^{zz}(q, \omega)$ are truncated to ensure that all features are noticeable. For the diagonal DSFs in Figure 3.13, the most noticeable feature is the existence of incommensurate points, i.e., the existence...
of peaks that occur in the $q\omega$-plane where $\omega = 0$ for values of $q$ different from $q = 0$ and $\pi$; these are most noticeable for $S^{yy}(q, \omega)$ plots (Figure 3.13(c) and 3.13(d)). The significance of these points is related to the fact that they are signatures of spinons since spin waves do not produce such points; in fact, the first experimental evidence for incommensurate spin excitations in an antiferromagnetic quantum spin chains ($S = 1/2$) came from the work of Dender et al. [56]. For $S^{xz}(q, \omega)$ plot in Figure 3.13(a), FM ordering corresponds to $(0, 0)$ in the $q\omega$-plane; it is clear that incommensurability is represented by a peak occurring at $q = \pi - \epsilon$ ($\epsilon > 0$), and thus ferromagnetic correlations in the $x$-direction become incommensurate. Incommensurability for $S^{yy}(q, \omega)$ and $S^{zz}(q, \omega)$ is also observed at $q = \pi - \epsilon$. To quantify the incommensurate points in the $q\omega$-plane, it is instructive to find the extrema of

$$\omega(q) = \varepsilon_{k-q} - \varepsilon_k = (J_y - J_x) \sin q \sin(2k - q) + 2g \mu_B B_x \sin(q/2) \sin(k - q/2)$$

(3.28)

with respect to the initial momentum of the spinon, $k$. The result is a set of four values

$$k_{\eta\eta'} = \frac{q}{2} + \eta \arccos \left[ \frac{-D(q) + \eta' \sqrt{D(q)^2 + 32C(q)^2}}{8C(q)} \right] + 2\pi \nu, \quad \nu \in \mathbb{Z}; \quad \eta, \eta' = \pm 1,$$

(3.29)

where $C(q) = (J_y - J_x) \sin q$ and $D(q) = 2g \mu_B B_x \sin q/2$. These $k$-values are placed back into (3.28) and compared with the density plot of $S^{yy}(q, \omega)$, as shown in Figure 3.12. As can be seen, $k_{++}$ and $k_{--}$ generate the boundary of the “inner” continuum, while $k_{--}$ and $k_{+-}$ bound the
“outer” continuum. Furthermore, the broadening of the continuum at \( q = \pi \) has width

\[
\Delta \omega = 4g\mu_B B_x.
\]  

(3.30)

Figure 3.12: Density plot of \( S^{yy}(q, \omega) \) for spinon scattering associated with the Villain mode and described by the spin-1/2 FM XYZ model. \( \omega(k_{++}) \) is given by white curve; \( \omega(k_{+-}) \) is represented by cyan curve; \( \omega(k_{-+}) \) corresponds to pink curve; \( \omega(k_{--}) \) is depicted by the grey curve. Plot is produced with HWHM = 0.025 meV to emphasise features; \( B_x = 3 \) T, \( J_x = 13.3 \) meV, \( J_y = 3.0 \) meV, \( J_z = 1.0 \) meV. The colour bar gives intensity in absolute units of meV\(^{-1}\) per formula unit.

An easy way to understand incommensurability is to interpret the incommensurable points for the spin-1/2 AFM Heisenberg model is to map the spins to interacting spinless fermions [57]. When the external magnetic field is absent, one has a half-filling of the fermion band. However, the presence of a field alters the Fermi level and results in incommensurate excitations [56].
Figure 3.13: Diagonal dynamic spin structure factors for spin-1/2 FM $XYZ$ model for spinon scattering with exchange interaction and an external transverse magnetic field present. $J_z = 13.3$ meV, $J_y = 3.0$ meV, $J_x = 1.0$ meV, $B_x = 3$ T. The intensity is given in absolute units of meV$^{-1}$ per formula unit.
Figure 3.14: (a) 3D and density plots of $S^{yz}(q,\omega) - S^{zy}(q,\omega)$ for the FM $XYZ$ chain in presence of transverse field. $J_z = 13.3$ meV, $J_y = 3.0$ meV, $J_x = 1.0$ meV, $B_x = 3$ T. The intensity is given in absolute units of meV$^{-1}$ per formula unit.

3.6 Dzyaloshinskii-Moriya Interaction

In 1958, Dzyaloshinskii [45] proposed, from symmetry considerations, a new type of anisotropic antisymmetric exchange interaction in order to explain the observation of so-called weak ferromagnetism occurring in antiferromagnets, i.e., the presence of a net magnetic moment in a system which should have none (due to strict AFM ordering). While this interaction seemed to account for experimental observations, its microscopic origins were unclear. It was then in 1960 when Moriya [46] put this matter on firmer ground by deriving the interaction based on perturbation theory, specifically taking into account spin-orbit coupling, which is essentially a relativistic correction term, in the theory of superexchange; the magnitude of this anisotropic interaction is then much smaller than that associated with the superexchange interaction. The interaction, which bears the name of both of these investigators, takes the untaggered form

$$\mathcal{H}_{D} = \sum_{i,j} D_{ij} \cdot (S_i \wedge S_j), \quad (3.31)$$

where $D_{ij}$ is the so-called Dzyaloshinskii-Moriya vector. The necessary requirement for Equation (3.31) to be non-zero is that the inversion symmetry between sites containing spin $i$ and spin $j$ must be broken. Low dimensional spin systems, as well as those containing an interface, are candidates for exhibiting DMI as they generally lack inversion symmetry, whereas in bulk materials, the interaction is typically absent. However, the notable exception to this is the class of B20 materials, of which MnSi is an example; here the presence of bulk DMI facilitates the existence of skyrmions [58, 59]. To add a more physical picture to this discussion, one takes the example of a crystal exhibiting one-dimensional AFM ordering: one is aware that the usual exchange interaction is responsible for creating an anti-parallel spin configuration; however, the presence of DMI causes
the spins to cant as in Figure 3.15 in order to minimise the system’s energy. Hence, a net magnetic moment results.

Figure 3.15: Canting in 1D AFM spin chain due to the presence of DMI; the result is a net magnetic moment producing weak ferromagnetism.

While DMI was proposed over half a century ago, much debate is still generated about this interaction [60], and only recently have experimental techniques been developed [61] to successfully probe and determine the direction of $D_{ij}$ in Equation (3.31). As well as this enhancement of fundamentally understanding DMI, the practical implications of harnessing this interaction are of great importance, particularly in the area of multiferroics [62] and spintronics where the ultimate aims are to realise high-density data storage technologies and low-energy-cost logic devices. Focusing on spintronics, which is relevant to the research presented here, the specific role that is envisaged for DMI is to exploit its intimate relationship with chiral spin configurations in order to enhance data storage technology [63]. Remarkably, it has been recently demonstrated [53, 52] that domain walls in ultrathin ferromagnetic films possessing a chiral property can, under the influence of an electrical current, propagate through the nanowire at surprisingly high velocities due to the propensity of the domain walls to assume one chiral state over the other. Other experimental work involves placing a single layer of manganese atoms on a tungsten substrate [64]; the explicit breaking of inversion symmetry occurring at the interface results in DMI being a non-negligible interaction. Overall, understanding and manipulating spin chirality via DMI opens up the avenue for controlling spin currents, which are themselves of interest in this dissertation. Furthermore, it has already been emphatically demonstrated that chirality plays a central role in the propagation of spinon occurring in 1D AFM quantum spin chains [38].

With all of this, there is a strong motivation to investigate the effects of DMI in the anisotropic spin chains discussed so far, specifically in the context of neutron scattering. The aim in what follows is to demonstrate the effect that this interaction has on the one-spinon dispersion. Then, for the first time, we investigate how the presence of the Dzyaloshinskii-Moriya interaction affects the various dynamic spin structure factors. Following this, the unique signatures of DMI are identified through the use of polarised neutron scattering, the primary tool for the investigation of such systems [65]; the results will demonstrate that DMI and the external transverse magnetic field, discussed in the previous section, have a similar effect on the diagonal dynamic spin structure factors ($S^{xx}(q, \omega)$, $S^{yy}(q, \omega)$, $S^{zz}(q, \omega)$), while it is only through the use of polarised neutrons that one can unambiguously distinguish between the influence of DMI and that of an external field; this is connected to the linear combination of dynamic spin structure factors directly related to the polarisation of the neutrons, $S^{yz}(q, \omega) - S^{zy}(q, \omega)$. 
Once again, the spin-1/2 FM XYZ model is investigated, with this system catering for a single spinon. The specific form of the (unstaggered) Dzyaloshinskii-Moriya interaction that is of interest is

\[ \mathcal{H}_{D_x} = D_x \sum_i (S_i \wedge S_{i+1})_x \]  

(3.32)

Following a similar procedure employed for the external transverse magnetic field, one applies \( \mathcal{H}_{D_x} \) to the FM kets given in Equation (3.12):

\[ \mathcal{H}_{D_x} |m, Q\rangle = \frac{D_x}{2t} (|m-1, Q\rangle - |m+1, Q\rangle), \]  

(3.33)

and diagonalises the Hamiltonian in question with usual the Bloch states, given by Equation (3.4),

\[ \mathcal{H}_{D_x} |k, Q\rangle = D_x \sin k |k, Q\rangle. \]  

(3.34)

Figure 3.16: Deformation of one-spinon band due to DMI; the chiral state at \( k = -\pi/2 \) is now favoured over that at \( k = \pi/2 \).

For the spin-1/2 FM XYZ model, and with Ising exchange, transverse exchange and Dzyaloshinskii-Moriya interactions all included, the dispersion relation now takes the form

\[ \epsilon_k = \frac{J_z}{2} + \frac{(J_y - J_x)}{2} \cos 2k + D_x \sin k. \]  

(3.35)

The corresponding plot is shown in Figure 3.16 and highlights an extremely salient feature: in contrast to the deformation of the one-spinon band induced by the transverse magnetic field in Figure 3.8, the effect of the Dzyaloshinskii-Moriya interaction is to lift the degeneracy associated
with the band minima, with one being lowered in energy, while the other is raised. This behaviour is related to the presence of $\sin k$ in the above dispersion relation, instead of $\cos k$ in the case of the external transverse field (Equation (3.26)). On more physical grounds the band minima, as alluded to in Chapter 2, correspond to the one-spinon states of maximal chirality, and the lowering of one compared to the other indicates that DMI is introducing a bias into the system with one chiral state being preferred over the other. In fact, for these states of maximal chirality, this scenario is reminiscent of Zeeman splitting induced by a magnetic field: the presence of the magnetic field lifts the degeneracy associated with the electron’s spin; in a similar fashion, the Dzyaloshinskii-Moriya interaction, with infinitesimal strength, can break the chiral degeneracy of the spinon; this is illustrated in Figure (3.17). From Equation (3.35), for small $D_x$, the energy difference between two the spinon states with maximal chirality (occurring at $k = -\pi/2$ and $k = +\pi/2$) is

$$\varepsilon_{k=+\pi/2} - \varepsilon_{k=-\pi/2} = 2D_x$$  \hspace{1cm} (3.36)

Figure 3.17: Cartoon illustrating the analogy between degeneracy-lifting of (a) electron spin, due to the presence of a magnetic field, (b) and that of spinon chirality due to the Dzyaloshinskii-Moriya interaction ($\hbar = 1$).

Turning now to the results, Figure 3.18, in a similar fashion to before compares the $S^{yy}(q,\omega)$, with $S^{zz}(q,\omega)$ once again possessing the dominant response. There are also new peaks observed, just as in the case of the transverse field; the explanation of their origin runs a parallel line to that presented in the previous section: DMI deforms the band in such a manner as to remove degeneracies.
present, leading to new observable transitions which manifest themselves in the form of peaks. Figure 3.19 displays the 3D plots for the various diagonal dynamic spin structure factors, along with corresponding density plots. Analogous to the case of the transverse field, incommensurate points arise close to $q = \pi$ when the Dzyaloshinskii-Moriya is non-zero. Furthermore, Figure 3.20 presents the results associated with $S^{yz}(q, \omega) - S^{zy}(q, \omega)$ for finite $D_x$. Interestingly a distinct asymmetry arises. Finally, the DSF results associated with the transverse field are compared to those of DMI in Figures 3.21 and 3.22. One can clearly see the close similarity between the two sets of results for the diagonal dynamic structure factors. However, the most striking aspect of this comparison is that, through the investigation of $S^{yz}(q, \omega) - S^{zy}(q, \omega)$, one can distinguish between the polarised response of that due to the transverse field and that due to DMI. In addition to this, since DMI is an inherent property of many magnetic compounds, polarised neutron scattering can be used as a means to detect it.

![Graph](image)

Figure 3.18: Comparison between $S^{zz}(q, \omega)$ (continuous black line) and $S^{xx}(q, \omega)$ (dashed blue line) for $D_x = (J_y - J_x)/7$ at $q = \pi/2$. New peaks appear due to the presence of DMI; the $S^{zz}(q, \omega)$ produces the strongest signal. The intensity is given in absolute units of meV$^{-1}$ per formula unit.
Figure 3.19: Diagonal dynamic spin structure factors for spin-1/2 FM $XYZ$ model for spinon scattering with exchange interaction and Dzyaloshinskii-Moriya interaction present. $J_z = 13.3$ meV, $J_y = 3.0$ meV, $J_x = 1.0$ meV, $D_x = (J_y - J_x)/7$. The intensity is given in absolute units of meV$^{-1}$ per formula unit.
Figure 3.20: $S^{yx}(q, \omega) - S^{xy}(q, \omega)$ produces a finite signal the Ising when $D_x$ is non-zero; $D_x = (J_y - J_x)/T$ and $J_z = 13.3$ meV, $J_y = 3.0$ meV, $J_x = 1.0$ meV. Because the DMI induces an antisymmetry in $S^{yx}(q, \omega) - S^{xy}(q, \omega)$ with respect to $\omega$, the intensity falls to zero along the axis $\omega = 0$. The intensity is given in absolute units of meV$^{-1}$ per formula unit.
Figure 3.21: Comparison of diagonal dynamic spin structure factors for spin-1/2 FM \(XYZ\) model for spinon scattering with exchange interaction in the presence of a transverse magnetic field \[(a), (c), (e)\] Dzyaloshinskii-Moriya interaction \[(b), (d), (f)\] present. 
\[J_z = 13.3 \text{ meV}, \quad J_y = 3.0 \text{ meV}, \quad J_x = 1.0 \text{ meV}, \quad D_x = (J_y - J_x)/7.\]
The intensity is given in absolute units of meV\(^{-1}\) per formula unit.
(a) $S^y_z(q, \omega) - S^y_z(q, \omega); B_x = 3 \, \text{T}$.  
(b) $S^y_z(q, \omega) - S^y_z(q, \omega); D_x = (J_y - J_x)/7$.

Figure 3.22: Comparison of $S^y_z(q, \omega) - S^y_z(q, \omega)$ for $B_x \neq 0$ and $D_x \neq 0$.  
(a) $B_x = 3 \, \text{T}$.  
(b) $D_x = (J_y - J_x)/7$; 
$J_x = 13.3 \, \text{meV}$, $J_y = 3 \, \text{meV}$, $J_z = 1 \, \text{meV}$.  
The intensity is given in absolute units of $\text{meV}^{-1}$ per formula unit.
Chapter 4

Green function technique applied to one-dimensional spin-$\frac{1}{2}$ systems

4.1 Introduction and motivation

The subject of this chapter is to describe the numerical technique utilised in calculating the dynamic spin structure factors for 1D spin chains accommodating two spinons at $T = 0$ K. Motivation to develop such a method stems from earlier work by Ishimura and Shiba [32], who themselves exploited the close relationship between Green functions and dynamic spin structure factors; their approach was analytical in nature. However, as one will see, this method is limited in terms of the interactions which it can accommodate. The numerical technique involving Green functions developed here allows one to incorporate new interactions, specifically an external magnetic field applied transverse to the Ising direction and the Dzyaloshinskii-Moriya interaction; such interactions are not realisable in the analytical approach.

4.2 Spinon pair creation

In the preceding chapter, the excitations discussed are thermally nucleated within the spin chains. However, one now turns to a regime in which quantum fluctuations are totally dominant, namely the limit $T \to 0$ K. Since thermal fluctuations are completely suppressed, the process by which excitations can be introduced into the system is through the transfer of energy and momentum from probing neutrons. As well as the absence of thermal energy, this marks another difference to that of the processes described before: instead of scattering the excitations within the one-spinon band, one creates a pair of spinons out of the ground state at, as illustrated in Figure 4.1; it is the quantum fluctuations in the system which mobilise these excitations once they are introduced. In an actual scattering experiment, if a neutron “flips” a spin, the neutron transfers $\Delta S_z = 1$ to the magnetic compound under investigation. If the sample in question exhibits 3D magnetic
ordering, then this scattering process generates a single magnon, a spin-1 excitation. Therefore, for a given momentum transfer, there is a fixed energy transfer, resulting in a narrow peak in the energy distribution corresponding to an excitation (a magnon); an experimental example is Collins et al. [66]. However, if the elementary excitation is a fractional excitation, as in the case of a spinon which possesses spin-1/2, then transferring spin-1 into the system by a neutron introduces a pair of spinons, the energy of which can be distributed in a number of ways depending on their relative momentum. Hence, for a given momentum transfer, there is a range of energies that the excitations can possess. Therefore, in this zero temperature regime, the existence of an excitation continuum is a signature of spinons being the independent elementary excitations.

In order to quantify the investigation of the two-spinon system, one is again interested in calculating the dynamic spin structure factors and, in the regime $T = 0$ K, the most general form of a dynamic spin structure factor is

$$S^{\alpha\beta}(Q, \omega) = \int \frac{dt}{2\pi \hbar} e^{-i\omega t} \langle \lambda | S^{\alpha\beta}_Q(0) S^{\alpha\beta}_Q(t) | \lambda \rangle,$$

where all the symbols have been outlined in the previous chapter. In terms of the spin-1/2 AFM $XXZ$ model, Ishimura and Shiba (IS) [32] were the first to theoretically investigate this type of creation process in the context of neutron scattering. Specifically, they calculated $S^{xx}(Q, \omega)$ and $S^{zz}(Q, \omega)$ using a combination of non-degenerate perturbation theory and a Green function approach, which itself was inspired by the work of Kalkstein and Soven [67]. While this approach has proven very fruitful, it is nonetheless quite restrictive: IS limited themselves to the starting point of Néel state $|A\rangle = |\uparrow\downarrow\uparrow\downarrow\cdots\uparrow\downarrow\rangle$ with the transverse exchange interaction being the
only perturbation which was accommodated for within the theoretical framework put forth. In the research presented here, however, both Néel states, $\ket{A}$ and $\ket{B}$, are taken into account for completeness; more importantly, the approach of IS is generalised to include additional interactions: the presence of an external magnetic field transverse to the Ising direction, Dzyaloshinskii-Moriya interaction and the existence of a so-called staggered field, due to presence of neighbouring spin chains, are all incorporated simultaneously. Furthermore, not only are $S^{xx}(Q,\omega)$ and $S^{zz}(Q,\omega)$ calculated with these new interactions present, but also $S^{yy}(Q,\omega)$ and $S^{yz}(Q,\omega) - S^{zy}(Q,\omega)$. As one will see, this set of DSFs will ultimately lead to the computation of the inelastic neutron scattering cross section for the two-spinon system governed by the spin-1/2 AFM XXZ model; this is presented in Chapter 6, while the case of interchain interaction is discussed in Chapter 7. In order to develop this new generalised approach to compute the structure factors given by Equation (4.1), it is illuminating to first follow a path parallel to that originally taken by Ishimura and Shiba.

### 4.3 Perturbed ground state and dynamic spin structure factors

As mentioned above, the work of Ishimura and Shiba started with first order perturbation theory concerning the Néel ground state $\ket{A}$. Here, however, both Néel states, $\ket{A}$ and $\ket{B}$, are examined with the perturbed ground state now taking the form

$$|GS\rangle_{\chi} \approx |\chi\rangle + \frac{1}{E_{GS} - \mathcal{H}_I} \mathcal{H}' |\chi\rangle,$$

(4.2)
with $|\chi\rangle = |A\rangle$ or $|B\rangle$; $\mathcal{H}_I$ is the Ising Hamiltonian and $\mathcal{H}' = \mathcal{H}_{XY} + \mathcal{H}_{B_x} + \mathcal{H}_{D_x}$. Explicitly,

$$\mathcal{H}_I = \sum_i N J_z S_i^z S_{i+1}^z$$

(4.3a)

$$\mathcal{H}_{XY} = \sum_i N J_t \frac{1}{2} \left( S_i^+ S_{i+1}^- + S_i^- S_{i+1}^+ \right),$$

(4.3b)

$$\mathcal{H}_{B_x} = g\mu_B B_x \sum_{i=1}^N S_i^x,$$

(4.3c)

$$\mathcal{H}_{D_x} = D_x \sum_{i=1}^N (S_i^x \wedge S_{i+1}^x).$$

(4.3d)

$\mathcal{H}_I$ is the dominant term with all other interactions above treated as perturbations. It should be emphasised that IS did not consider $\mathcal{H}_{B_x}$ or $\mathcal{H}_{D_x}$. In a more explicit form, Equation (4.2) reads

$$|GS\rangle_{\chi} \approx |\chi\rangle - \frac{1}{2J_z} \left( \mathcal{H}_{XY} |GS\rangle_{\chi} + \mathcal{H}_{B_x} |GS\rangle_{\chi} \right),$$

(4.4)

where

$$|j, j+\rho\rangle_A = \uparrow \downarrow \uparrow \downarrow \uparrow \uparrow \cdots j \uparrow \downarrow \cdots \uparrow \downarrow \uparrow \downarrow \cdots j \uparrow \downarrow \cdots \uparrow \downarrow \downarrow \uparrow \downarrow \rightarrow, \text{ for } |A\rangle,$$

(4.5a)

$$|j, j+\rho\rangle_B = \downarrow \uparrow \downarrow \uparrow \uparrow \cdots j \uparrow \downarrow \cdots \uparrow \downarrow \downarrow \uparrow \downarrow \cdots j \uparrow \downarrow \cdots \uparrow \downarrow \uparrow \uparrow \rightarrow, \text{ for } |B\rangle.$$ 

(4.5b)

denote clusters of $\rho$ reversed spins after site $j$. Therefore, the transverse exchange is responsible for flipping two adjacent spins, while the quantum fluctuations induced by the transverse magnetic field produce just a single spin flip. Notably, DMI is absent in Equation (4.4). This is due to this particular interaction being closely linked to chiral magnetic correlations, as mentioned in Chapter 2, and that pure Néel states do not possess such a property. Thus it is not surprising that DMI does not provide a contribution to the above perturbed ground state.

The other essential ingredient in computing the DSFs, as mentioned previously and forming the central theme of this chapter, is Green functions. Before providing details about these entities, it is best to first see how they naturally arise from the dynamic spin structure given by Equation (4.1). Using the resolution of identity, $\sum_{\lambda'} |\lambda\rangle \langle \lambda'| = \mathbb{I}$, and taking the explicit form of the Heisenberg representation to obtain the Dirac $\delta$-function (representing energy conservation), one has ($\hbar = 1$)

$$S_{\alpha\beta}^\lambda (Q, \omega) = \delta(\omega - E_{\lambda'} + E_\lambda) \langle \lambda | S_{\alpha Q}^\lambda | \lambda' \rangle \langle \lambda' | S_{\beta Q}^\lambda | \lambda \rangle,$$

(4.6)
In the context of spinon-pair creation, $|\lambda\rangle$ is the perturbed ground state $|GS\rangle$ in Equation (4.2), which includes the presence of quantum fluctuations in the system; $|\lambda'\rangle$ represents the final states of interest, i.e., the two-spinon states, which will be labelled by $|f\rangle$. While IS only considered $S^{xx}(Q, \omega)$ and $S^{zz}(Q, \omega)$ for Ising and transverse exchange interactions, here, as in the case of spinon scattering, the following DSFs are computed for the transverse field, DMI and interchain interaction, as well as the Ising and transverse exchange: $S^{xx}(Q, \omega)$, $S^{yy}(Q, \omega)$, $S^{zz}(Q, \omega)$, $S^{yz}(Q, \omega) - S^{zy}(Q, \omega)$. In terms of the diagonal structure factors, these take the form of Fermi’s Golden Rule [68]:

$$S^{\alpha\alpha}(Q, \omega) = \sum_f \delta(\omega - E_f + E_{GS}) \langle f|S^{\alpha}_{Q}|GS\rangle^2, \quad (4.7)$$

$\alpha = x, y, z$. Using Sokhotsky’s formula,

$$\lim_{\epsilon \to 0} \frac{1}{x + i\epsilon} = -i\pi \delta(x) + P \left( \frac{1}{x} \right), \quad (4.8)$$

the $\delta$-function takes the new form

$$\delta(\omega - E_f + E_{GS}) = -\frac{1}{\pi} \lim_{\epsilon \to 0} \text{Im} \left[ \frac{1}{\omega - E_f + E_{GS} + i\epsilon} \right]. \quad (4.9)$$

Implementing this in Equation (4.7), one has

$$S^{\alpha\alpha}(Q, \omega) = -\frac{1}{\pi} \sum_f \lim_{\epsilon \to 0} \text{Im} \left[ \langle GS|S^{\alpha}_{Q} \frac{|f\rangle\langle f|}{\omega - E_f + E_{GS} + i\epsilon} S^{\alpha}_{Q}|GS\rangle \right]. \quad (4.10)$$

Defining the Green function spanned by Equations (4.5)

$$G = \sum_{f} \lim_{\epsilon \to 0} \frac{|f\rangle\langle f|}{\omega - E_f + E_{GS} + i\epsilon}, \quad (4.11)$$

where $|f\rangle$ is a two-spinon state, one obtains the final form of the diagonal structure factors:

$$S^{\alpha\alpha}(Q, \omega) = -\frac{1}{\pi} \text{Im} \left[ \langle GS|S^{\alpha}_{Q} G S^{\alpha}_{Q}|GS\rangle \right]. \quad (4.12)$$

The other quantity of interest is $S^{\alpha\beta}(Q, \omega) - S^{\beta\alpha}(Q, \omega)$, and using Equation (4.6) above, one can express this linear combination as

$$S^{\alpha\beta}(Q, \omega) - S^{\beta\alpha}(Q, \omega) = \frac{i}{\pi} \text{Im} \left[ i\langle GS|S^{\alpha}_{Q} G S^{\beta}_{Q}|GS\rangle - i\langle GS|S^{\beta}_{Q} G S^{\alpha}_{Q}|GS\rangle \right]. \quad (4.13)$$
In order to fully evaluate the above dynamic structure factors, one follows IS and computes the basic constituents in both Equations (4.12) and (4.13):

\[ S_\alpha^Q |GS\rangle , \]

with \(|\chi\rangle = |A\rangle\) or \(|B\rangle\) and \(\alpha = x, y, z\); to emphasise that momentum transfer is along the spin chain, \(Q \rightarrow Q\). Given that \(S_\alpha^Q = \frac{1}{\sqrt{N}} \sum_n e^{iQn} S_\alpha^n\), \(S_\alpha^n\) acting on the \(|GS\rangle\) in Equation (4.2) either undoes one of the flipped spins created by \(H_{XY}\) or, keeping within the two-spinon subspace, produces an additional flipped spin adjacent to the two initially flipped spins. In the case of the single flipped spin introduced by the transverse magnetic field, a similar outcome arises. However, one can return back to the ground state when the single flipped spin is reversed. \(S_y^n\) produces an analogous outcome, while \(S_z^n\) returns the form of that given in Equation (4.2), given that Equations (4.5) are eigenstates of \(S_z^n\). For the case of the unperturbed Néel ground state \(|A\rangle\) for the spin-1/2 AFM XXZ model, these results are inherent in the following results pertaining to Equation (4.14).

\[
S_x^Q |GS\rangle_A \approx \frac{1}{2} \left[ e^{iQ} \left(1 - \frac{J_t}{J_z} \cos Q\right) |1\rangle_A - e^{iQ} \frac{J_t}{2J_z} \left(1 + e^{i2Q}\right) |3\rangle_A - e^{iQ} \frac{g\mu_B B_x}{2J_z} \left(1 + e^{iQ}\right) |2\rangle_A 
- e^{iQ} \frac{g\mu_B B}{2J_z} \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{iQj} |A\rangle \right],
\]

\[
S_y^Q |GS\rangle_A \approx \frac{i}{2} \left[ e^{iQ} \left(1 - \frac{J_t}{J_z} \cos Q\right) |1\rangle_A - e^{iQ} \frac{J_t}{2J_z} \left(1 + e^{i2Q}\right) |3\rangle_A + \frac{g\mu_B B_x}{2J_z} e^{iQ} \left(e^{iQ} - 1\right) |2\rangle_A 
- e^{iQ} \frac{g\mu_B B}{2J_z} \frac{1}{\sqrt{N}} \sum_{j=1}^{N} (-1)^j e^{iQ(j+1)} |A\rangle \right],
\]

\[
S_z^Q |GS\rangle_A \approx \frac{J_t}{2J_z} (1 - e^{iQ}) e^{iQ} |2\rangle^\prime_A + \frac{g\mu_B B}{2J_z} e^{iQ} |1\rangle^\prime_A,
\]

where

\[
|\zeta\rangle_A = \frac{1}{\sqrt{N}} \sum_j e^{iQj} |j, j + \zeta\rangle_A,
\]

\[
|\rho\rangle_A = \frac{1}{\sqrt{N}} \sum_j e^{i(Q+j)} |j, j + \rho\rangle_A,
\]

and \(\zeta, \rho \in \{1, 2, 3, 4, \ldots\}\), i.e., \(\zeta\) and \(\rho\) dictate the number of flipped adjacent spins. One can
also establish the following relationship between the two different Néel states, \(|A\rangle\) and \(|B\rangle\), in the context of these operators acting on the perturbed ground state:

\[
S^x_Q|GS\rangle_B \rightarrow S^x_Q|GS\rangle_A \quad (4.17a)
\]
\[
S^y_Q|GS\rangle_B \rightarrow -S^y_Q|GS\rangle_A \quad (4.17b)
\]
\[
S^z_Q|GS\rangle_B \rightarrow -S^z_Q|GS\rangle_A \quad (4.17c)
\]

Because \(S^x = 1/2(S^+ + S^-)\), application of \(S^x\) on either \(|GS\rangle_A\) or \(|GS\rangle_B\) produces the same result, hence there is no sign change. On the other hand, with \(S^y = 1/2i(S^+ - S^-)\), there is a minus sign arising from \(S^-\) when acting on \(|GS\rangle_A\) and \(|GS\rangle_B\). In the calculations that will follow, the states \(|A\rangle\) and \(|B\rangle\) will be omitted from Equations (4.17) since one is only interested in the two-spinon subspace. While the states \(|f\rangle\) in Equation (4.11) are not explicitly known, this does not impede the full computation of all the DSFs; the following illustrative calculation of \(S^{xx}(Q, \omega)\) demonstrates this, and also serves as way of showing how all other DSFs of interest are calculated within the two-spinon subspace. From Equation (4.15a)

\[
S^x_Q|GS\rangle_A \approx \frac{1}{2} [\Gamma|1\rangle_A - \Delta|3\rangle_A - \Lambda|2\rangle_A], \quad (4.18)
\]

where

\[
\Gamma = e^{iQ} (1 - \frac{J_t}{J_z} \cos Q), \quad (4.19a)
\]
\[
\Delta = e^{iQ} \frac{J_t}{2J_z} (1 + e^{2iQ}), \quad (4.19b)
\]
\[
\Lambda = e^{iQ} \frac{g \mu_B B_x}{2J_z} (1 + e^{iQ}). \quad (4.19c)
\]

Utilising Equation (4.12), one recasts the calculation in terms of explicit Green function matrix elements:

\[
S^{xx}(Q, \omega) \approx -\frac{1}{4\pi} \text{Im} \left[ |\Gamma|^2 \mathcal{G}_{11} - \Gamma^* \Delta \mathcal{G}_{13} - \Gamma^* \Lambda \mathcal{G}_{12} - \Gamma \Delta^* \mathcal{G}_{31} + |\Delta|^2 \mathcal{G}_{33} + \Delta^* \Lambda \mathcal{G}_{32} - \Gamma \Lambda^* \mathcal{G}_{21} + \Lambda^* \Delta \mathcal{G}_{23} + |\Lambda|^2 \mathcal{G}_{22} \right], \quad (4.20)
\]

where \(\Gamma, \Delta\) and \(\Lambda\) are functions of \(Q\); \(\mathcal{G}_{\zeta \rho} = \langle \zeta | \mathcal{G} | \rho \rangle\), defined by Equation (4.11), are the matrix elements of \(\mathcal{G}\), with the states given by Equation (4.16a). Therefore, the problem of calculating the DSF has been reduced to determining the matrix elements of the Green function, since \(\Gamma, \Delta\) and \(\Lambda\) are parameters that can be fixed. In the next section, an analytical approach (used by...
Ishimura and Shiba) and the numerical technique at the heart of our research are presented so that these matrix elements can be determined.

4.4 Green Function

To commence the discussion on Green functions, a representation for this entity is

\[ G(E) = \lim_{\eta \to 0} \frac{1}{E - \mathcal{H} + i\eta}. \] (4.21)

This is derived in Appendix A (Equation (A.15)), where a general overview of Green functions is provided. One can mould the above into two new incarnations; one spawning an analytical approach and the other, a numerical technique. The analytical approach, to be presented in Section 4.4.1, was employed by IS; the numerical technique is discussed in Section 4.4.3, with the starting point for the derivation of the algorithm associated with it being

\[ (E - \mathcal{H})G = I. \] (4.22)

To the best of the author’s knowledge, this is the first time that such a numerical approach is applied to a spin system.

In what follows, it is beneficial to illustrate how this numerical technique and analytical approach are applied to a simple system, which, in this case, is chosen to be the semi-infinite 1D tight-binding model. While serving as a prototype, this system is also directly applicable to this research since, as it will be delineated, one maps the spin systems of interest to the tight binding model. From such a vantage point, one can utilise all the Green function framework developed for the semi-infinite tight-binding model to ultimately compute the dynamic spin structure factors associated with two-spinon system governed by spin-1/2 AFM XXZ model.

4.4.1 Semi-infinite tight-binding system: analytical Green function technique

In this section the analytical approach is developed for extracting information regarding the Green function, specifically its matrix elements. This is all constructed around the framework of the one-dimensional semi-infinite tight-binding system with nearest-neighbour interaction, the Hamiltonian of which is given by

\[ \mathcal{H}_{TB} = \sum_{n=0}^{\infty} \varepsilon |n\rangle \langle n| + t |n\rangle \langle n + 1| + t^* |n + 1\rangle \langle n|, \] (4.23)

where \( \varepsilon \) is the on-site energy and \( t \) is referred to as the hopping term, representing the nearest-neighbour interaction; this system is depicted in Figure 4.3, with \( n = 0 \) referring to the surface site. What is described here also leads to the strategy implemented by IS to calculate \( S^{xx}(Q, \omega) \)
and $S^{zz}(Q, \omega)$, as they mapped the relevant excitations of the spin-1/2 AFM $XXZ$ model to this tight-binding model. The focus, in what follows, will be on calculating a particular type of Green function, namely the surface Green function. The usefulness of this quantity will become apparent when we return to the discussion of actually computing the dynamical spin structure factors. In order to obtain this surface Green function, the tight binding system itself is arranged to be in either an unperturbed or perturbed state. The unperturbed part consists of the surface site being “disconnected” from the rest of the lattice, as in Figure 4.4(a). Therefore, $\mathcal{H}_{TB}$ can be decomposed into an unperturbed Hamiltonian ($\mathcal{H}_0$) and a perturbation operator ($V$):

$$\mathcal{H}_{TB} = \sum_{n=1}^{\infty} \varepsilon |n\rangle \langle n| + \varepsilon |n\rangle \langle n| + t |n+1\rangle \langle n+1| + t^* |n+1\rangle \langle n| + t |1\rangle \langle 1| + t^* |1\rangle \langle 0| + V \langle 0| \langle 0| \langle 1| \langle 1| - \varepsilon |0\rangle \langle 0| + \varepsilon |0\rangle \langle 0| + t |1\rangle \langle 0| + t^* |0\rangle \langle 1|$$  \hspace{1cm} (4.24)

The perturbed state corresponds to the the surface site being connected to the rest of the lattice, as illustrated in Figure 4.4(b).

The analytical approach for calculating the surface Green function begins with slightly modified form of Equation (4.21),

$$G(z) = \frac{1}{z - \mathcal{H}_{TB}}$$  \hspace{1cm} (4.25)

where $z = E + i\eta$ with the limit $\eta \to 0$ implicit. Using Equation (4.24), one obtains

$$G = \frac{1}{z - \mathcal{H}_0 - V}.$$  \hspace{1cm} (4.26)
It is also convenient to introduce the unperturbed Green function:

\[ G^0 = \frac{1}{z - \mathcal{H}_0}, \quad (4.27) \]

Next, noting the identity

\[ \frac{1}{A + B} = \frac{1}{A} - \frac{1}{A} \frac{B}{A + B}, \quad (4.28) \]

and setting \( A = z - \mathcal{H}_0 \) and \( B = -V \), one obtains the Dyson equation [69]

\[ G = G^0 + G^0 V G \quad (4.29) \]

Since one wishes to calculate the surface Green function, namely \( G_{00} = \langle 0 | G | 0 \rangle \), it is natural to interpret Equation (4.29) as a matrix equation in the basis of localised states \( |n\rangle \). This matrix representation of the Green function, \( G \), associated with the perturbed system, depicted in Figure 4.4(b), is given as

\[
G = \begin{pmatrix}
G_{00} & G_{01} & G_{02} & G_{03} & G_{04} & \cdots \\
G_{10} & G_{11} & G_{12} & G_{13} & G_{14} & \cdots \\
G_{20} & G_{21} & G_{22} & G_{23} & G_{24} & \cdots \\
G_{30} & G_{31} & G_{32} & G_{33} & G_{34} & \cdots \\
G_{40} & G_{41} & G_{42} & G_{43} & G_{44} & \cdots \\
\vdots & \vdots & \vdots & \vdots & \vdots & \ddots
\end{pmatrix},
\]

(4.30)

while \( V \) from Equation (4.24), due to the configuration of the perturbation, has the corresponding matrix representation

\[
V = \begin{pmatrix}
0 & V_{01} & 0 & 0 & 0 & \cdots \\
0 & 0 & V_{10} & 0 & 0 & \cdots \\
0 & 0 & 0 & 0 & 0 & \cdots \\
0 & 0 & 0 & 0 & 0 & \cdots \\
0 & 0 & 0 & 0 & 0 & \cdots \\
\vdots & \vdots & \vdots & \vdots & \vdots & \ddots
\end{pmatrix},
\]

(4.31)

It is also clear from Equation (4.24) that

\[ V_{01} = t \quad (4.32a) \]
\[ V_{10} = t^* \quad (4.32b) \]

However, there is a further condition on the matrix elements of the unperturbed Green function,
\( G^0 \), which is due to the semi-infinite nature of the 1D chain. Generally, the matrix elements of \( G^0 \) are given by

\[
G^0 = \begin{pmatrix}
G_{00}^0 & G_{01}^0 & G_{02}^0 & G_{03}^0 & G_{04}^0 & \cdots \\
G_{10}^0 & G_{11}^0 & G_{12}^0 & G_{13}^0 & G_{14}^0 & \cdots \\
G_{20}^0 & G_{21}^0 & G_{22}^0 & G_{23}^0 & G_{24}^0 & \cdots \\
G_{30}^0 & G_{31}^0 & G_{32}^0 & G_{33}^0 & G_{34}^0 & \cdots \\
G_{40}^0 & G_{41}^0 & G_{42}^0 & G_{43}^0 & G_{44}^0 & \cdots \\
\vdots & \vdots & \vdots & \vdots & \vdots & \ddots
\end{pmatrix}.
\] (4.33)

Since the surface site is disconnected from the rest of the lattice in the unperturbed configuration of Figure 4.4(a), one can conclude that

\[
G_{0j}^0 = 0; \quad G_{j0}^0 = 0 \quad j \in \{1, 2, 3, \ldots \}
\] (4.34)

Therefore, the matrix representation of \( G^0 \) now takes the form

\[
G^0 = \begin{pmatrix}
G_{00}^0 & 0 & 0 & 0 & 0 & \cdots \\
0 & G_{11}^0 & G_{12}^0 & G_{13}^0 & G_{14}^0 & \cdots \\
0 & G_{21}^0 & G_{22}^0 & G_{23}^0 & G_{24}^0 & \cdots \\
0 & G_{31}^0 & G_{32}^0 & G_{33}^0 & G_{34}^0 & \cdots \\
0 & G_{41}^0 & G_{42}^0 & G_{43}^0 & G_{44}^0 & \cdots \\
\vdots & \vdots & \vdots & \vdots & \vdots & \ddots
\end{pmatrix}.
\]

The above can be further manipulated by exploiting the semi-infinite nature of the chain: since the chain which is decoupled from the surface site is itself semi-infinite, its existence simply mimics the perturbed system that is under investigation. Therefore, elements of the submatrix (inside the dashed red line in the matrix above) may be replaced with those of the full Green function associated with the perturbed system:

\[
G^0 = \begin{pmatrix}
G_{00}^0 & 0 & 0 & 0 & 0 & \cdots \\
0 & G_{01}^0 & G_{02}^0 & G_{03}^0 & \cdots \\
0 & G_{10}^0 & G_{11}^0 & G_{12}^0 & \cdots \\
0 & G_{20}^0 & G_{21}^0 & G_{22}^0 & \cdots \\
0 & G_{30}^0 & G_{31}^0 & G_{32}^0 & \cdots \\
\vdots & \vdots & \vdots & \vdots & \ddots
\end{pmatrix}.
\] (4.35)

To compute the surface Green function, \( G_{00} \), the resolution of identity \( \sum_{m=0}^{\infty} \langle m | m \rangle = I \) is used
in conjunction with Equation (4.29)
\[ G_{00} = \langle 0| G^0 |0 \rangle + \sum_{n,m} \langle 0| G^0 |n \rangle \langle n| V|m \rangle \langle m| G|0 \rangle \]  \hspace{1cm} (4.36)

Because the perturbation operator \( V \) concerns only site 0 and 1 in Equation (4.31) and Figure 4.4(b), the above simplifies to
\[ G_{00} = G_{00}^0 + G_{00}^0 V_{01} G_{10} + G_{01}^0 V_{01} G_{00}. \] \hspace{1cm} (4.37)

In order to produce an explicit form for \( G_{00} \), one notes that \( G_{01}^0 = 0 \) from Equation (4.34), and that
\[ G_{10} = G_{10}^0 + G_{10}^0 V_{01} G_{01} + G_{11}^0 V_{10} G_{00}. \] \hspace{1cm} (4.38)

Using matrix (4.35) and Equations (4.32), one then obtains a quadratic equation in \( G_{00} \)
\[ G_{00} = G_{00}^0 + G_{00}^0 |t|^2 (G_{00})^2. \] \hspace{1cm} (4.39)

The solution to this equation is
\[ G_{00}(z) = \frac{(z - \varepsilon)}{2|t|^2} \pm \frac{\sqrt{(z - \varepsilon)^2 - 4|t|^2}}{2|t|^2}, \] \hspace{1cm} (4.40)
given that
\[ G_{00}^0 = \langle 0| \frac{1}{z - H_0} |0 \rangle = \frac{1}{z - \varepsilon}. \] \hspace{1cm} (4.41)

In order to unravel Equation (4.40) further, one notes the condition of Equation (A.23) in Appendix A, namely \( \text{Im} \ G(E)_{aa} \leq 0 \); this implies that
\[ \text{Im} \ G_{00}(z) = \begin{cases} -\frac{\sqrt{4|t|^2 - (z - \varepsilon)^2}}{2|t|^2} & \text{for } |z - \varepsilon| < 2|t|, \\ 0 & \text{for } |z - \varepsilon| \geq 2|t|. \end{cases} \] \hspace{1cm} (4.42)

In a similar fashion, one can determine \( G_{01}, G_{10} \) and \( G_{11} \); Figure 4.5 displays the imaginary part of \( G_{00}(z) \).
4.4.2 Application of analytical Green function technique

With this analytical framework now in place, one can relate it back to the work of Ishimura and Shiba [32] and demonstrate how the dynamic spin structure factors can be calculated. Also, as highlighted previously, IS examined the scenario of having the transverse exchange interaction as the only perturbation in the system; Dzyaloshinskii-Moriya interaction, transverse magnetic field and interchain interaction were all absent from their model. They also imposed the condition that the total \( \mathbf{z} \)-component of spin, \( S^z_{\text{tot}} \), be a conserved quantity, with \( S^z_{\text{tot}} = +1 \) subspace being the only subspace that was investigated. This had the subsequent effect that the basis of states was composed of only an odd number of flipped spins, i.e.,

\[
|\zeta\rangle = \frac{1}{\sqrt{N}} \sum_j e^{iQj} |j, j + \zeta\rangle.
\]  

(4.43)

Correspondingly they considered \( \zeta \in \{1, 3, 5, \ldots \} \), which dictates the number of adjacent flipped spins. Since the transverse exchange induces a spin-flipping of two adjacent spins, this interaction connects states \(|1\rangle\) to \(|3\rangle\), \(|3\rangle\) to \(|5\rangle\), \(|5\rangle\) to \(|7\rangle\), etc.. Such a line of investigation allowed IS to map the spin system directly to the semi-infinite tight binding system; to explicitly see this, \( S^{xx}(Q, \omega) \) of Equation (4.20) is once again presented, but in compliance with IS, we choose \( D_x = 0 \) meV and \( B_x = 0 \) T (or equivalently \( \Lambda = 0 \)). Thus, one has the following

\[
S^{xx}(Q, \omega) \approx -\frac{1}{4\pi} \text{Im} \left[ |\Gamma| G_{11} - \Gamma^\ast \Delta G_{13} - \Gamma \Delta^\ast G_{31} + |\Delta|^2 G_{33} \right],
\]  

(4.44)

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Referring to Figure 4.6 below and keeping in mind that the Green function approach aids in evaluating Equation (4.44) in so far as extracting the various matrix elements present, one begins to see why one maps the spin system to that of a tight-binding model: sites of the tight-binding system can be mapped to the "sites" of the spin-1/2 AFM XXZ model.

$$\begin{align*}
TB & \xrightarrow{t} \varepsilon \\
XXZ & \xrightarrow{\gamma} J_z
\end{align*}$$

Figure 4.6: Mapping of XXZ spin system to semi-infinite tight-binding (TB) system

Each site in the XXZ model represents the state

$$\zeta \equiv |\zeta\rangle = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{iQj} |j, j + \zeta\rangle$$

More explicitly,

$$\zeta \equiv \begin{cases} 
\frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{iQj} |\uparrow \downarrow \uparrow \downarrow \cdots \uparrow | \uparrow \downarrow \cdots \uparrow \downarrow \downarrow \uparrow \downarrow \downarrow \rangle & \text{for} |A\rangle, \\
\frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{iQj} |\downarrow \uparrow \downarrow \uparrow \cdots \uparrow | \uparrow \downarrow \cdots \downarrow \uparrow \uparrow \downarrow \uparrow \uparrow \rangle & \text{for} |B\rangle.
\end{cases}$$

The interaction denoted by $\gamma$ in Figure 4.6 corresponds to the matrix elements

$$\gamma = \langle 2\nu - 1 |H_{XY}| 2\nu + 1 \rangle = \frac{J_t}{2} (1 + e^{-i2Q}), \quad (4.45)$$

where $\nu \in \{1, 2, 3, \ldots \}$. Therefore, $\gamma$ is associated with the energy of a transition from one state in Equation (4.43), to an "adjacent" state induced by the transverse exchange interaction, $H_{XY}$. Therefore, $\gamma$ can be mapped to the hopping term $t$ in the tight binding model. While $J_z$ is the energy cost of nucleating a pair of spinons, it can also be interpreted as the on-site energy $\varepsilon$ within
the framework of the semi-infinite tight-binding system. With Figure 4.6 in mind, one can also map the Green function matrix elements of the tight-binding model to that of the spin-1/2 XXZ system, as shown in Table 4.1.

<table>
<thead>
<tr>
<th>TB</th>
<th>XXZ</th>
</tr>
</thead>
<tbody>
<tr>
<td>$G_{00}$ ↔ $G_{11}$</td>
<td></td>
</tr>
<tr>
<td>$G_{01}$ ↔ $G_{13}$</td>
<td></td>
</tr>
<tr>
<td>$G_{10}$ ↔ $G_{31}$</td>
<td></td>
</tr>
<tr>
<td>$G_{11}$ ↔ $G_{33}$</td>
<td></td>
</tr>
</tbody>
</table>

Table 4.1: Mapping of Green function matrix elements from semi-infinite tight binding (TB) system to XXZ system.

Utilising Equations (4.42) and (4.38) as a starting point to obtain all other pertinent Green function matrix elements in Table 4.1, which from Equation (4.29) have the form

$$G_{01} = G_{01}^0 + G_{00}^0 V_{01} G_{11} + G_{01}^0 V_{10} G_{01},$$  \hspace{1cm} (4.46)

$$G_{11} = G_{11}^0 + G_{10}^0 V_{01} G_{11} + G_{11}^0 V_{10} G_{01},$$  \hspace{1cm} (4.47)

one can fully evaluate $S^{xx}(Q, \omega)$ in Equation (4.44).

### 4.4.3 Limitations of analytical Green function technique motivation of numerical approach.

Even though the above discussion illustrates the strength of this analytical approach involving Green functions, there are still challenges involved. For example, when an external magnetic field (with magnitude $B_x$) transverse to the Ising direction or the Dzyaloshinskii-Moriya interaction (magnitude $D_x$), is introduced, one can no longer focus on the subspace characterised by $S_{tot}^z = +1$. 

Figure 4.7: The presence of the transverse magnetic field and/or DMI connects the different subspaces characterised by the total $z$-component of the spin; consequently, $S_{tot}^z$ is no longer a conserved quantity, in contrast to the assumption underlying the work by Ishimura and Shiba [32].
+1, which was the case for IS: these new interactions connect the $S_{\text{tot}}^{z} = +1$ subspace to the $S_{\text{tot}}^{z} = 0$, and also the $S_{\text{tot}}^{z} = 0$ subspace to the $S_{\text{tot}}^{z} = -1$ subspace, as illustrated in Figure 4.7. Therefore, the conservation of $S_{\text{tot}}^{z}$ is removed. A more serious ramification of the presence of these interactions is that the lattice of spin states for the XXZ system, depicted in Figure 4.6, is no longer restricted to odd sites: $B_x$ and $D_x$ connect both odd and even sites, as indicated in Figure 4.8, and act as nearest-neighbour interactions. The original transverse exchange can now be interpreted as a next-nearest-neighbour interaction. Again, the various symbols in Figure 4.8 represent the non-zero matrix elements associated with transitions induced by the transverse exchange interaction $H_{XY}$ (Equation (4.3b)), transverse magnetic field, $H_{B_x}$ (Equation (4.3c)), and Dzyaloshinskii-Moriya interaction (Equation (4.3d)), $H_{D_x}$:

\[
\gamma \equiv \langle \zeta | H_{XY} | \zeta + 2 \rangle = \frac{J_t}{2} (1 + e^{-i2Q}), \tag{4.48a}
\]

\[
\tau \equiv \langle \zeta | H_{B_x} | \zeta + 1 \rangle = \frac{g \mu B x}{2} (1 + e^{-iQ}), \tag{4.48b}
\]

\[
\lambda \equiv \langle \zeta | H_{D_x} | \zeta + 1 \rangle = \frac{i D_x}{2} (1 - e^{-iQ}), \tag{4.48c}
\]

with

\[
|\zeta\rangle = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{iQj} |j, j + \zeta\rangle \tag{4.49}
\]

where now $\zeta \in \{1, 2, 3, \ldots \}$. This presence of effective nearest- and next-nearest-neighbour interactions has the negative effect that if one would attempt to generate Green function matrix elements using the analytical approach outlined above for the system depicted at the bottom of Figure 4.8, one would conclude that the approach breaks down precisely because both nearest- and next-nearest neighbour interactions are present. Therefore one is forced to find a new avenue which allows the extraction of all relevant Green function matrix elements.
4.4.4 Numerical Green function approach: decimation technique

This new approach, known as a decimation technique, which is completely independent of the method outlined above, allows one the flexibility to incorporate a transverse magnetic field, DMI, and interchain interactions into the spin system of interest; it is based on the work of Sancho et al. [70]. This technique begins with a simple one-dimensional lattice with nearest-neighbour interactions. Through an iterative process, the lattice constant is doubled with each step and simultaneously the nearest-neighbour interactions are replaced by residual interactions which are weaker than those associated with the initial lattice. The procedure is repeated until these residual interactions are vanishingly small, at which point the effective sites become decoupled. The technique has been implemented in a wide range of investigations, including topological insulators [71], electronic transport in carbon nanotubes [72], and calculations concerning giant magnetoresistance [73]. Once again, the aim is to obtain the surface Green function and in order to outline the algorithm, one refers back to the semi-infinite tight-binding model. The actual derivation of the algorithm begins with Equation (4.22),

\[(E - \mathcal{H})G = \mathbb{I} \tag{4.50}\]

In order to explore the result of producing $G_{00}$ from the equation above, one has, assuming an orthonormal basis,
\[ \langle 0 | (E - \mathcal{H}) \mathcal{G} | 0 \rangle = \mathbb{I}; \quad (4.51) \]

Employing the resolution of identity \( \sum_{m=0}^{\infty} |m\rangle \langle m| = \mathbb{I} \) and noting that \( \mathcal{H} \) in the above equation contains only on-site and nearest-neighbour interactions,

\[ (E - \mathcal{H}_{00}) \mathcal{G}_{00} = \mathbb{I} + \mathcal{H}_{01} \mathcal{G}_{10}. \quad (4.52) \]

Assuming all on-site energies are the same, as is the case for the semi-infinite tight-binding system, one can use the the notation of the semi-infinite tight-binding model (4.24)

\[
\begin{align*}
\mathcal{H}_{ij} &= t, \\
\mathcal{H}_{ji} &= t^\dagger, \\
\mathcal{H}_{ii} &= \varepsilon,
\end{align*}
\]

with \( i < j \); \( t^\dagger \) is in place instead of \( t^* \) in anticipation that matrices will be used. Equation (4.52) above now becomes

\[ (E - \varepsilon) \mathcal{G}_{00} = \mathbb{I} + t \mathcal{G}_{10}, \quad (4.54) \]

Taking the off-diagonal (10) matrix element of Equation (4.50), \( \mathcal{G}_{10} \) can be framed as follows

\[ (E - \varepsilon) \mathcal{G}_{10} = t^\dagger \mathcal{G}_{00} + t \mathcal{G}_{20}. \quad (4.55) \]

In order to retrieve \( \mathcal{G}_{00} \), one must obtain an expression for \( \mathcal{G}_{20} \); once again, using Equation (4.50):

\[ (E - \varepsilon) \mathcal{G}_{20} = t^\dagger \mathcal{G}_{10} + t \mathcal{G}_{30} \quad (4.56) \]

Due to the restriction to nearest-neighbour interaction, one can infer the general relation:

\[ (E - \varepsilon) \mathcal{G}_{n0} = t^\dagger \mathcal{G}_{n-1,0} + t \mathcal{G}_{n+1,0} \quad \text{for } n \geq 1 \quad (4.57) \]

At the end of this initial step, this recursive relation implies that

\[
\begin{align*}
(E - \varepsilon) \mathcal{G}_{00} &= \mathbb{I} + t \mathcal{G}_{10}, \\
(E - \varepsilon) \mathcal{G}_{n0} &= t^\dagger \mathcal{G}_{n-1,0} + t \mathcal{G}_{n+1,0} \quad \text{for } n \geq 1. 
\end{align*}
\]
Continuing in this fashion to determine the surface Green function \( G_{00} \), and placing Equation (4.58b) in Equation (4.58a), one obtains

\[
(E - [\epsilon + t(E - \epsilon)^{-1}t^\dagger])G_{00} = I + t(E - \epsilon)^{-1}tG_{20}.
\]  

(4.59)

From Equation (4.58b), new expressions for \( G_{n-1,0} \) and \( G_{n+1,0} \) can be deduced:

\[
G_{n-1,0} = (E - \epsilon)^{-1}t^\dagger G_{n-2,0} + (E - \epsilon)^{-1}tG_{n0}
\]  

(4.60a)

\[
G_{n+1,0} = (E - \epsilon)^{-1}t^\dagger G_{n0} + (E - \epsilon)^{-1}tG_{n+2,0}
\]  

(4.60b)

These terms are now fed back into Equation (4.58b):

\[
(E - \epsilon)G_{n0} = t^\dagger(E - \epsilon)^{-1}t^\dagger G_{n-2,0} + t^\dagger(E - \epsilon)^{-1}tG_{n0} + t(E - \epsilon)^{-1}t^\dagger G_{n0} + t(E - \epsilon)^{-1}tG_{n+2,0}
\]  

(4.61)

After this iteration, one now has

\[
(E - \epsilon_1^s)G_{00} = I + \alpha_1 G_{20},
\]  

(4.62a)

\[
(E - \epsilon_1)G_{n0} = \beta_1 G_{n-2,0} + \alpha_1 G_{n+2,0} \quad \text{for } n \geq 2,
\]  

(4.62b)

where

\[
\alpha_1 = t(E - \epsilon)^{-1}t,
\]  

(4.63a)

\[
\beta_1 = t^\dagger(E - \epsilon)^{-1}t^\dagger,
\]  

(4.63b)

\[
\epsilon_1^s = \epsilon + t(E - \epsilon)^{-1}t^\dagger,
\]  

(4.63c)

\[
\epsilon_1 = \epsilon + t(E - \epsilon)^{-1}t^\dagger + t^\dagger(E - \epsilon)^{-1}t.
\]  

(4.63d)

With the above, one can conclude that \( \alpha_1 \) and \( \beta_1 \) effectively encode the nearest-neighbour interactions, with \( \epsilon_1 \) representing an effective on-site energy within the bulk, and \( \epsilon_1^s \) representing the same for the effective surface site. Repeating the procedure, with Equations (4.62) as the starting point, one obtains

\[
(E - \epsilon_2^s)G_{00} = I + \alpha_2 G_{40},
\]  

(4.64a)

\[
(E - \epsilon_2)G_{n0} = \beta_2 G_{n-4,0} + \alpha_2 G_{n+4,0}, \quad \text{for } n \geq 2^2
\]  

(4.64b)
where

\[ \alpha_2 = \alpha_1 (E - \epsilon_1)^{-1} \alpha_1, \] (4.65a)
\[ \beta_2 = \beta_1 (E - \epsilon_1)^{-1} \beta_1, \] (4.65b)
\[ \epsilon_2^s = \epsilon_1^s + \alpha_1 (E - \epsilon_1)^{-1} \beta_1, \] (4.65c)
\[ \epsilon_2 = \epsilon_1 + \beta_1 (E - \epsilon_1)^{-1} \alpha_1 + \alpha_1 (E - \epsilon_1)^{-1} \beta_1. \] (4.65d)

Continuing in this process, one develops the recursion relationship, for \( n \geq 1 \), with recursion parameter \( k \)

\[ (E - \epsilon_k^s)G_{00} = I + \alpha_k G_{2^k,0}, \] (4.66a)
\[ (E - \epsilon_k)G_{2^k,n,0} = \beta_k G_{2^k(n-1),0} + \alpha_k G_{2^k(n+1),0}, \] (4.66b)

where

\[ \alpha_k = \alpha_{k-1} (E - \epsilon_{k-1})^{-1} \alpha_{k-1}, \] (4.67a)
\[ \beta_k = \beta_{k-1} (E - \epsilon_{k-1})^{-1} \beta_{k-1}, \] (4.67b)
\[ \epsilon_k^s = \epsilon_{k-1}^s + \alpha_{k-1} (E - \epsilon_{k-1})^{-1} \beta_{k-1}, \] (4.67c)
\[ \epsilon_k = \epsilon_{k-1} + \beta_{k-1} (E - \epsilon_{k-1})^{-1} \alpha_{k-1} + \alpha_{k-1} (E - \epsilon_{k-1})^{-1} \beta_{k-1}. \] (4.67d)

The initial conditions for the above are

\[ \alpha_0 = t, \] (4.68a)
\[ \beta_0 = t^\dagger, \] (4.68b)
\[ \epsilon_0^s = \epsilon, \] (4.68c)
\[ \epsilon_0 = \epsilon. \] (4.68d)

with \( t, t^\dagger \) and \( \epsilon \) given in Equation (4.53). In order to compute the surface Green function, \( G_{00} \), from Equation (4.66a), the iterative scheme represented by Equations (4.66) and (4.67) is repeated until \( \alpha_k \) and \( \beta_k \) are sufficiently small; this leads to the following central result from Equation (4.66a) that will be used to calculate the dynamical spin structure factors

\[ G_{00} \approx (E - \epsilon_k^s)^{-1} \] (4.69)

To cast the Green function as causal, one must introduce \(+i\eta\) in Equation (4.69). Applying this algorithm to the case of the 1D semi-infinite tight-binding model, governed by the Hamiltonian (4.23), one obtains the plot in Figure 4.9, which specifically displays the imaginary part of the surface Green function (4.69). This data is compared to that of the analytical approach in Figure
4.10; due to rapid convergence, one obtains excellent agreement.

Figure 4.9: Numerical results for the imaginary part of the surface Green function associated with the 1D semi-infinite nearest-neighbour tight binding model; \( \varepsilon = 2.0 \text{ meV}, \ t = 0.5 \text{ meV} \). Results produced using algorithm described in main text (Equations (4.66) and (4.67)), with \( \eta \to 0 \); output from algorithm terminating at \( k = 17 \).

Figure 4.10: Comparison between analytical result of Equation (4.42) (continuous blue line) presented separately in Figure 4.5 and that of the iterative approach of Equations (4.66) and (4.67) (red dots); \( \varepsilon = 2.0 \text{ meV}, \ t = 0.5 \text{ meV} \). Iterative data show excellent agreement with analytical results.
4.5 Green function technique applied dynamic spin structure factors

Even though a new framework for computing the surface Green function has now been put in place, it still needs to be demonstrated that this method allows one to compute all matrix elements required to fully compute $S_{xx}(Q, \omega)$ and all other dynamic spin structure factors of interest. In particular, it is desirable to include the transverse exchange interaction, transverse magnetic field, DMI, and interchain interaction; from Equation (4.20), $S_{xx}(Q, \omega)$ is given as

$$S_{xx}(Q, \omega) \approx -\frac{1}{4\pi} \text{Im} \left[ |\Gamma|^2 G_{11} - \Gamma^* \Delta G_{13} - \Gamma^* \Lambda G_{12} - \Gamma \Delta^* G_{31} + |\Delta|^2 G_{33} \right.$$ 

$$+ \Delta^* \Lambda G_{32} - \Gamma \Lambda^* G_{21} + \Lambda^* \Delta G_{23} + |\Lambda|^2 G_{22} \right], \quad (4.70)$$

In its current form, the result of the algorithm described in the previous section is $G_{00}$ (Equation (4.69)) and, recalling Table 4.1, the mapping between tight-binding model and XXZ model is $G_{00} \leftrightarrow G_{11}$. This means that only one of the Green function matrix elements in Equation (4.70) can be computed. With that, one would be inclined to believe that this decimation technique only furnishes information about state $|1\rangle$, or equivalently, site 1 in Figure 4.6. In order to circumvent this issue, one must take an innovative approach which involves a rearrangement of the lattice in Figure 4.8 so that the surface Green function encapsulates all the relevant quantities for the full computation of $S_{xx}(Q, \omega)$; an analogous approach is put in place to compute $S_{yy}(Q, \omega)$, $S_{zz}(Q, \omega)$, and $S_{yz}(Q, \omega) - S_{zy}(Q, \omega)$. This reconfiguration of the system is depicted in Figure 4.11; one refers to the dashed boxes as super sites, with each super site containing four individual sites, and each individual site representing a particular two-spinon state dictated by Equation (4.49). In this conversion process, and with additional information now encoded, Equations (4.66) and (4.67) are converted from scalar equations to matrix equations in the basis $|n\rangle$; to signify this, all elements are indicated with a hat. Explicitly,

$$\begin{align*}
(EI - \hat{\epsilon}_k^*) \hat{G}_{00} &= \mathbb{I} + \hat{\alpha}_k \hat{G}_{2k,n,0}, \\
(EI - \hat{\epsilon}_k) \hat{G}_{2k,n,0} &= \hat{\beta}_k \hat{G}_{2k(n-1),0} + \hat{\alpha}_k \hat{G}_{2k(n+1),0}, \quad (4.71a, 4.71b)
\end{align*}$$

where

$$\begin{align*}
\hat{\alpha}_k &= \hat{\alpha}_{k-1} (EI - \hat{\epsilon}_{k-1})^{-1} \hat{\alpha}_{k-1}, \\
\hat{\beta}_k &= \hat{\beta}_{k-1} (EI - \hat{\epsilon}_{k-1})^{-1} \hat{\beta}_{k-1}, \\
\hat{\epsilon}_k^* &= \hat{\epsilon}_{k-1}^* + \hat{\alpha}_{k-1} (EI - \hat{\epsilon}_{k-1})^{-1} \hat{\beta}_{k-1}, \\
\hat{\epsilon}_k &= \hat{\epsilon}_{k-1} + \hat{\beta}_{k-1} (EI - \hat{\epsilon}_{k-1})^{-1} \hat{\alpha}_{k-1} + \hat{\alpha}_{k-1} (E - \hat{\epsilon}_{k-1})^{-1} \hat{\beta}_{k-1}. \quad (4.72a, 4.72b, 4.72c, 4.72d)
\end{align*}$$
The initial conditions for the above are

\[ \hat{\alpha}_0 = t, \quad (4.73a) \]
\[ \hat{\beta}_0 = t^\dagger, \quad (4.73b) \]
\[ \hat{\epsilon}_s^0 = \varepsilon, \quad (4.73c) \]
\[ \hat{\epsilon}_0 = \varepsilon. \quad (4.73d) \]

Due to the reconfiguration illustrated in Figure 4.11, \( \varepsilon \) and \( t \) are matrices respectively representing the on-site energy and nearest-neighbour interaction for super sites. In the basis given by Equation (4.49), the matrix representations of the on-site energy \( \varepsilon \) and the energy associated with nearest-neighbour interaction, \( t \), between super sites are given as (associated with spin-1/2 XXZ model):

\[
\varepsilon = \begin{pmatrix}
J_z & \tau + \lambda & \gamma & 0 \\
\tau^* + \lambda^* & J_z & \tau + \lambda & \gamma \\
\gamma^* & \tau^* + \lambda^* & J_z & \tau + \lambda \\
0 & \gamma^* & \tau^* + \lambda^* & J_z
\end{pmatrix},
\quad (4.74a)
\]

\[
t = \begin{pmatrix}
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
\gamma^* & \tau^* + \lambda^* & 0 & 0 \\
0 & \gamma^* & 0 & 0
\end{pmatrix},
\quad (4.74b)
\]

where, as in Equation (4.48),

\[
\gamma \equiv \langle \zeta | \mathcal{H}_{XY} | \zeta + 2 \rangle = \frac{J_z}{2} (1 + e^{-iQ}),
\quad (4.75a)
\]
\[
\tau \equiv \langle \zeta | \mathcal{H}_{Bx} | \zeta + 1 \rangle = \frac{g\mu_B B_x}{2} (1 + e^{-iQ}),
\quad (4.75b)
\]
\[
\lambda \equiv \langle \zeta | \mathcal{H}_{Dx} | \zeta + 1 \rangle = \frac{iD_x}{2} (1 - e^{-iQ}),
\quad (4.75c)
\]

with DMI being an implicit interaction since it does not appear explicitly in the perturbed ground state, Equation (4.4). Returning to the surface Green function, and relating to the Green function matrix elements in Equation (4.70), one explicitly has

\[
\tilde{G}_{00} = \begin{pmatrix}
G_{11} & G_{12} & G_{13} & G_{14} \\
G_{21} & G_{22} & G_{23} & G_{24} \\
G_{31} & G_{32} & G_{33} & G_{34} \\
G_{41} & G_{42} & G_{43} & G_{44}
\end{pmatrix} = \begin{pmatrix}
G_{11} & G_{12} & G_{13} & 0 \\
G_{21} & G_{22} & G_{23} & G_{24} \\
G_{31} & G_{32} & G_{33} & G_{34} \\
0 & G_{42} & G_{43} & G_{44}
\end{pmatrix},
\quad (4.76)
\]

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Matrix elements $G_{00}^{41}$ and $G_{00}^{14}$ are zero since there is no interaction present to create a direct transition between site 1 and site 4, as indicated in Figure 4.11(b). It should also be noted that, comparing Equation (4.76) with Equation (4.70), the following matrix elements are not relevant: $G_{24}, G_{34}, G_{43}, G_{44}, G_{42}$. Due to degeneracy, all super sites have the same on-site energy, $\varepsilon$; due to the translational invariance of the transverse exchange, transverse magnetic field and Dzyaloshinskii-Moriya interactions, the effective nearest-neighbour interaction $t$ is the same between any two adjacent super sites. As described above, these two matrices are the initial matrices used in Equations (4.63) to output the surface Green function associated with super site 0 (dashed box) in Figure 4.11(b):

$$\tilde{G}_{00} \approx (E - \varepsilon_{k}^{s})^{-1}. \quad (4.77)$$

According to Equation (4.76), $\tilde{G}_{00}$ contains all the matrix elements required to compute $S^{yx}(Q, \omega)$ in Equation (4.20). Most notably, this computation does not only include the transverse exchange interaction that IS dealt with, but also the presence of a transverse magnetic field and Dzyaloshinskii-Moriya interaction. In a similar fashion, one can compute $S^{yy}(Q, \omega), S^{zz}(Q, \omega)$ and $S^{yz}(Q, \omega) - S^{zy}(Q, \omega)$. The results for all of these structure factors are presented in the next chapter for the case of the spin-1/2 XXZ model.
Figure 4.11: (a): Original lattice with transverse exchange interaction ($\gamma$), DMI ($\lambda$) and $B_x$ ($\tau$) present. (b) Reconfiguration of lattice in (a) to an arrangement that is conducive to extract all matrix elements required to compute the various DSFs, using the numerical approach delineated in main text. (c) Mapping the lattice in to an effective 1D nearest-neighbour lattice.
Chapter 5

Dynamic structure factors for spinon pair creation in spin-$^{1/2}$ antiferromagnetic $XXZ$ Heisenberg model

5.1 Introduction and motivation

Following on from the work developed in the previous chapter, the dynamic spin structure factors of interest are computed using the Green function framework. The cases of Ising and transverse exchange interactions, application of an external magnetic field transverse to the Ising direction, and the presence of the Dzyaloshinskii-Moriya interaction are all considered; for the case of transverse field and DMI, this is the first time such results have been produced, with motivation to include such interactions once again stemming from the chiral nature of spinons. It should also be emphasised that the computation of dynamic spin structure factors via quantum groups does not permit the inclusion of a transverse magnetic field.

5.2 Calculation of dynamic spin structure factors

Having established the framework for the Green function technique, the results pertaining to the inelastic neutron scattering cross section of the spin-$^{1/2}$ AFM $XXZ$ system catering for two spinons are presented in this chapter. Using first order perturbation theory outlined in chapter 4, the explicit forms of dynamic spin structure factors of interest are given below in terms of Green function matrix elements; all coefficients presented are implicit functions of the momentum transfer along the chain, $Q$, with all other symbols explained in previous sections.
As mentioned in Chapter 4, the dynamic structure factor $S_{xx}(Q,\omega)$, is the spin wave response and describes spin flip scattering:

$$S_{xx}(Q,\omega) \approx -\frac{1}{4\pi} \text{Im} \left[ \Gamma^2 G_{11} - \Gamma^* \Delta G_{13} - \Gamma^* \Lambda G_{12} - \Gamma \Delta^* G_{31} + |\Delta|^2 G_{33} \\
+ \Delta^* \Lambda G_{32} - \Lambda \Delta^* G_{21} + \Lambda^* \Delta G_{23} + |\Lambda|^2 G_{22} \right],$$

(5.1a)

$$\Gamma = e^{iQ} \left( 1 - \frac{J_t}{J_z} \cos Q \right),$$

(5.1b)

$$\Delta = e^{iQ} \frac{J_t}{2J_z} \left( 1 + e^{i2Q} \right),$$

(5.1c)

$$\Lambda = e^{iQ} \frac{g\mu_B B}{2J_z} \left( 1 + e^{iQ} \right).$$

(5.1d)

$S_{yy}(Q,\omega)$ provides similar information to that of $S_{xx}(Q,\omega)$ above:

$$S_{yy}(Q,\omega) \approx -\frac{1}{\pi} \text{Im} \left[ \Pi^* \Pi G_{22} + \Pi^* \Theta G_{21} + \Theta^* \Pi G_{12} + \Theta^* \Theta G_{11} + \Xi^* \Phi G_{31} + \Xi^* \Xi G_{33} + \Xi G_{32} + \Phi^* \Pi G_{21} + \Phi^* \Phi G_{22} \right],$$

(5.2a)

$$\Pi = -\frac{i}{2} e^{iQ} + e^{iQ} \frac{iJ_t}{4J_z} (e^{-iQ} + e^{iQ}),$$

(5.2b)

$$\Xi = e^{iQ} \frac{iJ_t}{4J_z} (1 + e^{i2Q}),$$

(5.2c)

$$\Phi = e^{iQ} \frac{g\mu_B B}{4J_z} (1 - e^{iQ}).$$

(5.2d)

$S_{zz}(Q,\omega)$ represents the longitudinal magnetic response and, in the limit $\omega \to 0$, this particular dynamic structure factor (DSF) relates to the magnetic ordering of the system:

$$S_{zz}(Q,\omega) \approx -\frac{1}{\pi} \text{Im} \left[ \Omega^* \Omega G_{22} + \Omega^* \Theta G_{21} + \Theta^* \Omega G_{12} + \Theta^* \Theta G_{11} \right],$$

(5.3a)
\[ \Omega = \frac{J_t}{2J_z} (1 - e^{iQ}) e^{iQ}, \quad (5.3b) \]
\[ \Theta = \frac{g\mu_B B_x}{2J_z} e^{iQ}, \quad (5.3c) \]

- As outlined in Appendix B, \( S^{yz}(Q, \omega) - S^{zy}(Q, \omega) \) can be measured by scattering of polarised neutrons and is the quantity which assists in detecting signatures of chirality associated with spinons:

\[
S^{yz}(Q, \omega) - S^{zy}(Q, \omega) \approx \frac{i}{\pi} \text{Im} \left[ i \{ \chi^* \phi G_{12} + \chi^* \theta G_{11} + \xi^* \phi G_{32} + \xi^* \phi G_{31} + \zeta^* \phi G_{22} + \zeta^* \theta G_{21} \} 
- i \{ \chi \phi^* G_{21} + \chi \theta^* G_{11} + \xi \phi^* G_{23} + \xi \phi^* G_{13} + \zeta \phi^* G_{22} + \zeta \theta^* G_{12} \} \right],
\]
\[
(5.4a)
\]

\[
\chi = \frac{i}{2} e^{iQ} \left( 1 - \frac{J_t}{2J_z} (e^{iQ} + e^{-iQ}) \right),
\]
\[
(5.4b)
\]
\[
\xi = -\Xi,
\]
\[
(5.4c)
\]
\[
\zeta = -\Phi,
\]
\[
(5.4d)
\]
\[
\phi = -\frac{J_t}{2J_z} (1 - e^{iQ}) e^{iQ},
\]
\[
(5.4e)
\]
\[
\theta = -\Theta
\]
\[
(5.4f)
\]

The results which follow are grouped into three sections: the first deals with only Ising and transverse exchange interactions present in the system; the second part concerns Ising exchange, transverse exchange and an external field applied transverse to the Ising direction; the final section concerns the inclusion of the Dzyaloshinskii-Moriya interaction. In each of these sections, the results are discussed and physically interpreted.

### 5.3 System with Ising and transverse exchange interactions

The \( XXZ \)-Hamiltonian including only Ising and transverse exchange interactions is given by

\[
\mathcal{H}_{XXZ} = \sum_{i=1}^{N} J_z S_i^z S_{i+1}^z + \sum_{i=1}^{N} \frac{J_t}{2} (S_i^+ S_{i+1}^- + S_i^- S_{i+1}^+).
\]
\[
(5.5)
\]
Figure 5.1: Response of $S^{yz}(Q,\omega) - S^{zy}(Q,\omega)$ in the absence and presence of a transverse magnetic field. With $J_z = 13.8$ meV, $J_t = \epsilon J_z = 1.9$ meV, $\epsilon \equiv J_t/J_z = 0.137$. The intensity is given in absolute units of meV$^{-1}$ per formula unit.

The results for the dynamic structure factors following from this Hamiltonian are shown in Figure 5.2; as expected, $S^{xx}(Q,\omega)$ and $S^{yy}(Q,\omega)$ have similar intensities, as both issue information about the spin wave response and reflect the spin-flip process occurring in the system. On the other hand, the longitudinal response, $S^{zz}(Q,\omega)$, is weaker. Qualitatively, this is the opposite of what is observed for the Villain mode in Chapter 3, where $S^{zz}(Q,\omega)$ possessed the most dominant signal. For the present case of no transverse magnetic field and zero DMI, $S^{yz}(Q,\omega) - S^{zy}(Q,\omega)$ has no signal and is displayed in Figure 5.1(a).
Figure 5.2: Three-dimensional plots and density plots of the components of the diagonal dynamic structure factors for spinon pair creation governed by spin-1/2 AFM $XXZ$; With $J_z = 13.8$ meV, $J_t = \epsilon J_z = 1.9$ meV, $\epsilon \equiv J_t/J_z = 0.137$. The 3D plots possess a white background instead of black for ease of viewing. The intensity is given in absolute units of meV$^{-1}$ per formula unit.
In order to explain how the various features of the plots in Figure 5.2 arise, it is useful to recall the actual technique which produces these particular results. Specifically, the analogy with the semi-infinite tight-binding model serves not just as a framework to progress the calculations but also allows for physical interpretation. In terms of the lattice diagrams used in the previous section, Figure 5.3(b) illustrates the configuration for the scenario of only Ising and transverse exchange being present: the two chains are decoupled with the top chain connecting all odd sites and the bottom chain containing all even sites. Each tight-binding site corresponds to a state (from here on, referred to as a *cluster state*), represented by
\[ \zeta = |Q; \zeta \rangle = \begin{cases} \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{iQj} |\uparrow \downarrow \uparrow \cdots \uparrow_{\frac{1}{2}} \uparrow_{\frac{3}{2}} \cdots \downarrow_{\frac{1}{2}} \downarrow_{\frac{3}{2}} \cdots \uparrow \rangle, & \text{for } |A\rangle, \\ \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{iQj} |\downarrow \uparrow \downarrow \cdots \downarrow_{\frac{1}{2}} \uparrow_{\frac{3}{2}} \cdots \downarrow_{\frac{1}{2}} \downarrow_{\frac{3}{2}} \cdots \uparrow \rangle, & \text{for } |B\rangle, \end{cases} \]

where \(|A\rangle\) and \(|B\rangle\) are the two Néel states; in a more compact form,

\[ |Q; \zeta \rangle = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{iQj} |j, j + \zeta \rangle, \quad \zeta \in \{1, 2, 3, \ldots\}. \tag{5.6} \]

In the work of Ishimura and Shiba (IS) [32], the focus was exclusively on the \(S_{\text{tot}}^z = +1\) subspace with only the transverse exchange interaction present. In terms of Figure 5.3(b), this corresponds to the top chain of odd sites with only one Néel background considered. While IS restricted themselves to the \(S_{\text{tot}}^z = +1\) subspace, one can readily reproduce their results by equivalently investigating the two spinon system characterised by \(S_{\text{tot}}^z = 0\), i.e., focusing only on the bottom chain of even sites. As noted before, due to the presence of a transverse magnetic field or Dzyaloshinskii-Moriya interaction, all subspaces, represented by \(S_{\text{tot}}^z = +1, S_{\text{tot}}^z = -1,\) and \(S_{\text{tot}}^z = 0\), are taken into account in this research; this is depicted in Figure 5.3(a). In order to put matters on a more quantitative footing, it is helpful to recall the matrix elements related to the transverse exchange interaction:

\[ \gamma \equiv \langle Q; \zeta | \hat{H}_{XY} | Q; \zeta + 2 \rangle = \frac{J_t}{2} (1 + e^{-iQ}). \tag{5.7} \]

Because one is momentarily distinguishing between the top and bottom chains, \(\zeta \in \{1, 3, 5, \ldots\}\) (top) or \(\zeta \in \{2, 4, 6, \ldots\}\) (bottom). Within the framework of the tight-binding model, \(\gamma\) is the analogue of the hopping term and so is related to the bandwidth of the two-spinon dispersion. To see this explicitly, it is illuminating to investigate the magnitude of \(\gamma\), which represents the strength of the nearest-neighbour interaction of the tight-binding model (Figure 5.3(b)):

\[ |\gamma| = \left| \frac{J_t}{2} (1 + e^{-iQ}) \right| = |J_t \cos Q|. \tag{5.8} \]

A plot of \(|\gamma|\) as a function of \(Q\) is presented in Figure 5.4; \(\gamma\) has maximal strength at \(Q = 0\) and \(Q = \pi\). This is reflected in Figure 5.2 where the spread in the excitation continuum is at its largest for these momentum transfer values; \(\gamma\) has minimal strength at \(Q = \pi/2\), where the bandwidth of the continuum essentially goes to zero at a “pinch point”, as in Figure 5.2. Any momentum transfer value in the range \([0, \pi] \setminus \{0, \pi, \pi/2\}\), corresponds to an intermediate strength of \(\gamma\).

To understand in particular the distribution of intensities produced in the various plots, it is revealing to first discuss the extreme case of \(\gamma = 0\) at \(Q = \pi/2\); the transition from non-zero \(\gamma\)
to zero $\gamma$ is shown in Figure 5.5. In terms of the tight-binding model, this scenario is equivalent to hopping becoming completely suppressed.

It is then natural to ask on which site in Figure 5.3(b) does the system find itself, or equivalently what cluster state does the spin chain possess with this interaction switched off. The probability of a spin cluster containing $\rho$ flipped spins in Equation (5.6), or equivalently, being on a particular site $l$ in the the tight binding system, is

$$P_{\rho} = |\langle l; \rho | Q; \zeta \rangle|^2 = \left| \frac{1}{\sqrt{N}} e^{iQl} \right|^2 = \frac{1}{N},$$

where $|Q, \zeta\rangle$ is given by Equation (5.6) and

$$|l, \rho\rangle \equiv |l, l + \rho\rangle = |\uparrow \downarrow \cdots \uparrow_{l} | \uparrow \downarrow \cdots \uparrow_{l+\rho}, \quad \text{for } |A\rangle,$$

$$|l, \rho\rangle \equiv |l, l + \rho\rangle = |\downarrow \uparrow \cdots \downarrow_{l} | \uparrow \downarrow \cdots \uparrow_{l+\rho}, \quad \text{for } |B\rangle.$$

Therefore, it is equally likely that a given site is occupied in Figure 5.3(b). Tantamount to this, occupying a state with a fixed number of flipped adjacent spins, as given by Equation (5.6) with $\zeta$ fixed. Figure 5.5 shows an example of site 3 being occupied when $\gamma$ vanishes; this corresponds to exactly three flipped adjacent spins roaming around the spin chain as a cluster state.
Figure 5.5: Transition from non-zero effective nearest neighbour interaction ($\gamma$) to isolated sites for $Q = \pi/2$. When the transverse exchange interaction is switched off, it is equally probable that a given site is “occupied”, according to Equation (5.9). In the example shown here, the system is “trapped” on site 3 (highlighted in yellow), which corresponds to three flipped adjacent spins, according to Equation (5.6).

Further to this, one recalls the one-spinon dispersion for the case of only transverse and Ising exchange in the spin-1/2 AFM $XXZ$ model [28],

$$\varepsilon_k = \frac{J_z}{2} + J_t \cos 2k. \quad (5.11)$$

Under the assumption that the excitations are non-interacting, the dispersion relation which arises for the two-spinon system, as presented in Chapter 2, is

$$\omega(Q) = \varepsilon_{k_1} + \varepsilon_{k_2} = J_z + 2J_t \cos Q \cos 2\kappa, \quad (5.12)$$

where $Q = k_1 + k_2$ is the centre-of-mass momentum and $\kappa = (k_1 - k_2)/2$ is the relative momentum. In the case of $Q = \pi/2$, where the influence of the transverse exchange is effectively suppressed, the system is equivalent to a pure Ising system. At $Q = \pi/2$ and energy $J_z$, the continuum
Figure 5.6: Comparison between two independent spinon picture ans that of cluster states. The boundary curves are produced for fixed relative momentum, $\kappa = \pi/2$ (yellow) and $\kappa = \pi$ (brown); the continuum and intensities are produced from a basis of cluster states. With $J_z = 13.8$ meV, $J_t = \epsilon J_z = 1.9$ meV, $\epsilon \equiv J_t/J_z = 0.137$. The colour bar intensity is given in absolute units of meV$^{-1}$ per formula unit.

The colour bar intensity is given in absolute units of meV$^{-1}$ per formula unit.

collapses to a point and all states of different relative momentum are degenerate with each other. Consequently, there is a high intensity at this point. Another interesting observation to note is that when $\kappa$ is fixed to be $\pi/2$ or $\pi$, one obtains the bounding curves to the continuum, as illustrated in Figure 5.6. As one varies the relative momentum $\kappa$ from 0 to $\pi$, the curves in (5.12) sweep across the continuum; such a description corresponds to the two independent spinon picture. Note, however, that the intensities shown in Figure 5.6 are produced via the Green function technique which utilises the cluster states of Equation (5.6). Therefore, one sees the two extreme pictures of cluster states and independent elementary excitations merging; the agreement between the bounding curves and the continuum in Figure 5.6 is an indication of this.

If one now goes to the other special cases of $Q = 0$ and $Q = \pi$, the effective tight-binding amplitude is at its strongest. Even though the basis of states is a set of cluster states (Equation (5.6)), it is this quantum tunnelling induced by the transverse exchange interaction that leads to the system accommodating excitations which mimic those behaving independently of each other, i.e., a system comprising of two non-interacting spinons. However, because of the strong transitioning between states (5.6) at $Q = 0$ and $Q = \pi$, the energy spread is maximal.

With all of this in mind, the quantity denoted by $\gamma$ can now be interpreted as that which determines the width of the excitation continuum; $Q$-values between 0 and $\pi/2$ and $\pi/2$ and $\pi$ represent intermediate strengths of $\gamma$. 90
(a) Ising exchange interaction and transverse exchange interaction ($\gamma$) present, along with a transverse magnetic field ($\tau$).

(b) At $Q = \pi/2$, the transverse exchange vanishes while the transverse magnetic field is still non-zero.

Figure 5.7: Lattice of super sites. (a) All interactions are present. (b) Only transverse exchange interaction is non-zero.

### 5.4 System in the presence of transverse magnetic field

Now that the case of the Ising and transverse exchange interactions has been discussed, this section deals with the presence of an external magnetic field applied transverse to the Ising direction. The Hamiltonian describing such a system is

$$
\mathcal{H}_{XXZ} + \mathcal{H}_{B_x} = \sum_i J_z S_i^z S_{i+1}^z + J_t (S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) + g\mu_B B_x \sum_{i=1}^{N} S_i^x
$$

As before, it is helpful to present the matrix elements associated with the transverse field:

$$
\tau \equiv \langle Q; \zeta | \mathcal{H}_{B_x} | Q; \zeta + 1 \rangle = \frac{g\mu_B B_x}{2} (1 + e^{iQ})
$$
with $\zeta \in \{1, 2, 3, \ldots\}$, and, specifically its magnitude,

$$|\tau| = \left| g\mu_B B_x \cos \left( \frac{Q}{2} \right) \right|$$ (5.15)

with the corresponding plot shown in Figure 5.8. Unlike the effective hopping term $\gamma$ associated with the transverse exchange interaction, $|\tau|$ monotonically decreases in the interval $[0, \pi]$, with maximal strength occurring at the momentum transfer value of $Q = 0$. This interaction vanishes for $Q = \pi$ where the system behaves as if only the transverse exchange interaction is responsible for transitioning between different cluster states. With the transverse field non-zero, the quantum tunnelling which arises due to its presence can be interpreted as a nearest-neighbour hopping in the tight-binding framework, and the transverse exchange interaction is now recast as a next-nearest-neighbour interaction; this is depicted in Figure 5.7(a). In a similar vein as that discussed previously, $\tau$ can be interpreted as being related to the bandwidth and so dictates the spread of the excitation continuum. This can be seen in Figure 5.9 where the additional spread is largest at $Q = 0$ with no contribution at $Q = \pi$, conforming with the information presented in Figure 5.8. Compared to the transverse exchange, the pinch point which occurred at $Q = \pi/2$ is now broadened due to the presence of the external magnetic field. Interestingly, when the momentum transfer value is $\pi/2$, the transverse exchange is suppressed and one is then left with an Ising spin chain with a transverse field applied [74, 4]. In this regime, the dynamics of the excitations are purely related to the quantum fluctuations induced by the field; in terms of tight binding model, this is illustrated in Figure 5.7(b).

Following the line of argument in the previous section, now with the term associated with the magnetic field introduced, the one spinon dispersion reads

$$\varepsilon_k = \frac{J_z}{2} + J_t \cos 2k + g\mu_B B_x \cos k.$$ (5.16)
Figure 5.9: Three-dimensional plots and density plots of the components of the diagonal dynamic structure factors for a two-spinon system governed by spin-1/2 AFM $XXZ$ model; $J_z = 13.8$ meV, $J_t = cJ_z = 1.9$ meV, $c \equiv J_t/J_z = 0.137$. The transverse magnetic field is non-zero with $B_x = 3$ T. The intensity is given in absolute units of meV$^{-1}$ per formula unit.
The corresponding two-spinon dispersion relation given as

\[ \omega(Q) = \varepsilon_{k_1} + \varepsilon_{k_2} = J_z + 2J_t \cos Q \cos 2\kappa + 2g\mu_B B_x \cos Q / 2 \cos \kappa, \] (5.17)

where once again \( q \) is the centre-of-mass momentum and \( \kappa \) is the relative momentum. The bounding curves associated with the continuum when the transverse field is non-zero are generated by fixing \( \kappa \in \{0, \pi, \pi/2\} \). The comparison between the continuum, produced from a cluster state picture, and the bounding curves produced from the two non-interacting spinon picture is shown in Figure 5.10; the excellent agreement between the two indicates that a superposition of cluster states of Equation 5.6 do in fact mimic the two free spinons. The interesting feature which arises from this comparison is that the peaks in the Figure 5.10 occur exactly at the intersection points of these curves. To quantify this, it is illuminating to use Equation (5.17) for the relative momenta of interest:

\[ \omega(Q)|_{\kappa=0} = J_z + 2J_t \cos Q + 2g\mu_B B_x \cos Q / 2, \] (5.18a)
\[ \omega(Q)|_{\kappa=\pi/2} = J_z - 2J_t \cos Q, \] (5.18b)
\[ \omega(Q)|_{\kappa=\pi} = J_z + 2J_t \cos Q - 2g\mu_B B_x \cos Q / 2. \] (5.18c)
Figure 5.11: Bounding curves of continuum when external transverse field is non-zero. The intersection points of the curves mark the most intense signals in the dynamic spin structure factors. The energy difference $\Delta E$ is related to the transverse field (Equations (5.27) and (5.28)). The curves are generated from Equation (5.17) with $\kappa = \pi/2$ (yellow), $\kappa = \pi$ (white), $\kappa = 0$ (brown); $B_x = 3$ T; $J_z = 13.8$ meV, $J_t = \epsilon J_z = 1.9$ meV, $\epsilon \equiv J_t/J_z = 0.137$.

Then, solving for $Q$ in the equation

$$\omega(Q)_{|\kappa=\pi/2} = \omega(Q)_{|\kappa=0},$$

(5.19)

one has

$$4J_t \cos Q + 2g\mu_B B_x \cos Q/2 = 0.$$  \hspace{1cm} (5.20)

This has solutions

$$k_1 = 2 \arccos \left[ \frac{1}{2} \left( -\frac{C}{2A} + \sqrt{\frac{C^2}{4A^2} + 2} \right) \right], \hspace{1cm} (5.21)$$

$$k_2 = 2 \arccos \left[ \frac{1}{2} \left( -\frac{C}{2A} - \sqrt{\frac{C^2}{4A^2} + 2} \right) \right], \hspace{1cm} (5.22)$$

where $A = 4J_t$, $C = 2g\mu_B B_x$. In a similar fashion, solving for $Q$ in

$$\omega(Q)_{|\kappa=\pi/2} = \omega_{|\kappa=\pi}(Q),$$

(5.23)
or more explicitly,

\[ 4J_t \cos Q - 2g \mu_B B_x \cos Q/2 = 0, \quad (5.24) \]

produces

\[ k_3 = 2 \arccos \left[ \frac{1}{2} \left( \frac{C}{2A} + \sqrt{\frac{C^2}{4A^2} + 2} \right) \right], \quad (5.25) \]

\[ k_4 = 2 \arccos \left[ \frac{1}{2} \left( \frac{C}{2A} - \sqrt{\frac{C^2}{4A^2} + 2} \right) \right]. \quad (5.26) \]

These particular momentum transfer values are highlighted in Figure 5.11. The next natural step is to determine the energy difference between the two intersection points:

\[ \Delta E = \omega(k_3)_{\kappa=\pi} - \omega(k_1)_{\kappa=0} = -2g \mu_B B_x (\cos k_1/2 + \cos k_3/2) + 2J_t (\cos k_1 - \cos k_3), \quad (5.27) \]

or, equivalently,

\[ \Delta E = \omega(k_2)_{\kappa=0} - \omega(k_4)_{\kappa=\pi} = -2g \mu_B B_x (\cos k_2/2 + \cos k_4/2) + 2J_t (\cos k_2 - \cos k_4). \quad (5.28) \]

\[ \Delta E > 0 \] since \( J_t > 0 \) and the magnetic field is applied in the negative \( x \)-direction; this spread is illustrated in Figure 5.11. Coincidentally, if one looks at a purely Ising system \( J_t = 0 \) with an external transverse field, the dispersion reads

\[ \Omega(q) = J_z + 2g \mu_B B_x \cos Q/2 \cos \kappa. \quad (5.29) \]

The bounding curves of the continuum associated with the above are \( g > 0 \) and external field applied in negative \( x \)-direction

\[ \Omega_{\max}(Q) = J_z - 2g \mu_B B_x \cos Q/2, \quad (5.30) \]

\[ \Omega_{\min}(Q) = J_z + 2g \mu_B B_x \cos Q/2. \quad (5.31) \]

When one looks at the energy difference of the intersection points displayed in Figures 5.10 and 5.11, the result is

\[ \Omega_{\max}(k_3) - \Omega_{\max}(k_1) = -2g \mu_B B_x (\cos k_1/2 + \cos k_3/2). \quad (5.32) \]

This is similar to Equation (5.27), except that the presence of second term in this expression arises from the transverse exchange interaction. However, from a physical perspective, the intersection points occur around \( Q = \pi/2 \) and \( Q = 3\pi/2 \) which correspond to the transverse exchange being
“switched off”, so the term corresponding to this interaction is quite weak. Therefore, one is indeed close to realising a purely Ising system with a transverse field at the point where these intersection points occur. Another interesting remark concerns the momentum transfer values of \( k_1, k_2, k_3 \) and \( k_4 \), which correspond to the intersection points and coincide with the points of strongest intensities: from Equation (5.20) (related to \( k_3 \) and \( k_4 \))

\[
2J_t \cos Q = -g\mu_B B_x \cos Q/2, \tag{5.33}
\]

and from Equation (5.24) (related to \( k_1 \) and \( k_2 \))

\[
2J_t \cos Q = g\mu_B B_x \cos Q/2. \tag{5.34}
\]

Then, noting that

\[
|\gamma| = |J_t \cos Q|, \tag{5.35a}
\]

\[
|\tau| = |g\mu_B B_x \cos Q/2|. \tag{5.35b}
\]

Therefore, \( |\tau(Q_\alpha)| = 2|\gamma(Q_\alpha)| \), where \( Q_\alpha \in \{k_1, k_2, k_3, k_4\} \). In terms of the tight-binding picture (Figure 5.7(a)), this implies that it is more likely that quantum tunnelling occurs between the top and bottom chains than along a chain. Furthermore, it should be emphasised that one set of equations arises naturally from the Green function technique using cluster states (5.6) (Equations (5.35)), while the other originates from treating the two spinons as independent excitations (Equations 5.33 and 5.34).

In relation to \( S^{yz}(Q, \omega) - S^{zy}(Q, \omega) \), it is interesting in its own right that a finite response appears when \( B_x \neq 0 \), as highlighted in Figure 5.1. However, as indicated already, this combination of DSFs is connected to the chirality of the spinons and the existence of a non-zero signal is a fingerprint of such a property. The occurrence of a finite signal in the presence of a transverse field can be explained in an analogous manner to that of the one-spinon case described by the spin-1/2 FM XYZ model in chapter 3: the magnetic field creates a deformation of the one-spinon band so that degeneracy associated with the chiral states is lifted.

Even with these interesting results, one must remember that the ground state involved in the calculation of Green function matrix elements is based on first order perturbation theory. Therefore, it must be that the energy associated with the external transverse magnetic field is considerably less than that of the Ising exchange energy, \( J_z \). In order to illustrate the failure of the Green function approach, Figure 5.13 depicts the two-spinon continuum for various values of \( B_x \). As an interesting side note, the quantum fluctuations introduced by the external magnetic field can drive the system to be quantum critical. The notion of quantum criticality is naturally embedded in the theory of quantum phase transitions [4]: these are phase transitions that occur at zero temperature and are therefore not driven by thermal fluctuations, but instead quantum fluctuations. For quantum
spin systems, this is manifested in the Heisenberg uncertainty principle for spin observables; the quantum critical point is one at which a many-body system undergoes a transition from one ground state to another. In the context of the antiferromagnetic spin-1/2 XXZ model, this corresponds to the gapped excitation continuum gradually becoming gapless as the strength of the magnetic field is increased, as depicted in Figure 5.13.

### 5.5 System with Dzyaloshinskii-Moriya interaction

In this final section of the results pertaining to the two-spinon continuum governed by the spin-1/2 AFM XXZ model, the Dzyaloshinskii-Moriya interaction is introduced into the system; the corresponding Hamiltonian reads

\[
\mathcal{H}_{XXZ} + \mathcal{H}_{Dx} = \sum_i J_z S_i^z S_{i+1}^z + J_t (S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) + D_x \sum_i (S_i^x S_{i+1}^x)
\]

As can be seen from Figure 5.14, the Dzyaloshinskii-Moriya interaction induces the same transitions as that of the transverse field. For this reason, one can follow a similar line of argument for the diagonal dynamic spin structure factors as discussed in the previous section. However, for the off-diagonal term, \( S^{yz}(Q, \omega) - S^{zy}(Q, \omega) \), there is a subtlety related to a finite response and this will be explained in what follows.

As before, the starting point concerns the non-zero matrix elements associated with the basis of states (5.6). Now interest lies with the Dzyaloshinskii-Moriya interaction with each matrix element
Figure 5.13: Density plots of $S^{xx}(Q,\omega)$ for various values of the transverse magnetic field. As the field strength increases, the continuum becomes gapless and the system goes quantum critical. The colour bar intensity is given in absolute units of meV$^{-1}$ per formula unit.

represented by $\lambda$ in Figure 5.14:

$$\lambda \equiv \langle Q; \zeta |\mathcal{H}_D| Q; \zeta + 1 \rangle = \frac{iD_x}{2}(1 - e^{iQ})$$

(5.37)

with $\zeta \in \{1, 2, 3, \ldots\}$; the corresponding magnitude is

$$|\lambda| = \left| \frac{iD_x}{2}(1 - e^{-iQ}) \right| = \left| D_x \sin \frac{Q}{2} \right|.$$ 

(5.38)

The corresponding plot of $|\lambda|$ is shown in Figure 5.15. Compared with the plot of $|\tau|$, one now observes a monotonic increase from $Q = 0$ to $Q = \pi$, instead of a decrease. Because of this, the largest spread in the excitation spectrum occurs at $Q = \pi$, as observed in Figure 5.16, with no additional spread occurring at $Q = 0$. As in the previous section, it is illuminating to look at the
bounding curves of the continuum that are themselves generated from the two non-interacting
spinon picture; the one-spinon dispersion for the spin-1/2 AFM XXZ model with DMI present is
given as

\[ \epsilon_k = \frac{J_z}{2} + J_t \cos 2k + D_x \sin k, \]  

(5.39)

with corresponding two-spinon dispersion

\[ \omega(Q) = \epsilon_{k_1} + \epsilon_{k_2} = J_z + 2J_t \cos Q \cos 2\kappa + 2D_x \sin Q / 2 \cos \kappa. \]  

(5.40)

The bounding curves of the continuum occur at relative momentum values of \( \kappa \in \{0, \pi/2, \pi\} \), as shown in Figure 5.17. As before, the strongest intensities occur at the intersection of these peaks. In terms of the energy difference between the intersection points, one has

\[ \Delta E = \omega(k_3')_{\kappa=0} - \omega(k_4')_{\kappa=\pi} = 2D_x (\sin k_3'/2 + \sin k_4'/2) + 2J_t (\cos k_3' - \cos k_4'), \]  

(5.41)

or, tantamount to the above,

\[ \Delta E = \omega(k_3')_{\kappa=0} - \omega(k_4')_{\kappa=\pi} = 2D_x (\sin k_2'/2 - \sin k_4'/2) + 2J_t (\cos k_2' - \cos k_4'), \]  

(5.42)

where \( k_1', k_2', k_3' \) and \( k_4' \) are, analogous to the case of the transverse field, the momentum transfer values at which the intersection points occur in Figure 5.17. Again, since DMI originates from spin-orbit coupling, the magnitude of \( D_x \) in actual systems is expected to be small compared with the transverse exchange energies. With that, the broadening due to its presence is not particularly large and consequently \( k_1', k_2', k_3' \) and \( k_4' \) are all close to \( Q = \pi/2 \) or \( Q = 3\pi/2 \). Furthermore, the
combination of the pure Ising case with DMI has the corresponding dispersion relation

\[ \Omega'(Q) = J_z + 2D_x \sin Q/2 \cos \kappa, \]  

(5.43)

with the bounding curves of the continuum associated with the above given by

\[ \Omega'_{\text{max}}(Q) = J_z + 2D_x \cos Q/2, \]  

(5.44)

\[ \Omega'_{\text{min}}(Q) = J_z - 2D_x \cos Q/2. \]  

(5.45)

Figure 5.15: Variation of the magnitude of the matrix elements \( \lambda \) associated with DMI with momentum transfer, \( Q; D_x = J_t/7; J_t = 1.9 \text{ meV}, J_z = 13.8 \text{ meV}. \)
Figure 5.16: Three-dimensional plots and density plots of the components of the diagonal dynamic structure factors for a two-spinon system governed by spin-1/2 AFM XXZ model. With $J_z = 13.8$ meV, $J_t = \epsilon J_z = 1.9$ meV, $\epsilon \equiv J_t/J_z = 0.137$. The Dzyaloshinskii-Moriya interaction is now present with $D_x = J_t/7$. The intensity is given in absolute units of meV$^{-1}$ per formula unit.
Figure 5.17: White is $\kappa = 0$; Yellow is $\kappa = \pi/2$; Brown is $\kappa = \pi$. $D_x = J_z/7$; with $J_z = 13.8$ meV, $J_t = \epsilon J_z = 1.9$ meV, $\epsilon \equiv J_t/J_z = 0.137$. The colour bar intensity is given in absolute units of meV$^{-1}$ per formula unit.

The energy difference associated with the momentum transfer values of $k_1'$ and $k_2'$ is

$$\Omega'_{\text{max}}(k_3') - \Omega'_{\text{max}}(k_1') = 2D_x (\sin k_3'/2 + \sin k_1'/2). \quad (5.46)$$

a similar result arises for $k_2'$ and $k_4'$. One therefore sees the similarity between Equations (5.41) and (5.46) and how a pure Ising system begins to emerge around $Q = \nu \pi/2$ for $\nu$ odd, where the effects of the transverse exchange die out. Furthermore, by a similar approach to that presented in the previous section, it can be demonstrated that $2|\gamma(Q_\beta)| = |\lambda(Q_\beta)|$, with $Q_\beta \in \{k_1', k_2', k_3', k_4'\}$. This again indicates that it is more likely to transition between the top and bottom chains than along a chain in Figure 5.14 when DMI is present in the system.

Turning attention to the off-diagonal dynamic spin structure factors, one of the most interesting features to arise out of the investigation of 1D spin chains with the Dzyaloshinskii-Moriya interaction is the fact that, with only DMI, Ising and transverse exchange present, there is a zero response from $S^{yz}(Q, \omega) - S^{zy}(Q, \omega)$, as shown in Figure 5.18(a). This may be due to destructive interference, indicating that the system possesses an accidental degeneracy in terms of chirality. However, when the transverse field is switched on, a finite signal is produced since the accidental degeneracy is now lifted. For very small fields, the support of the plot takes the expected form when compared to that of the diagonal DSFs in Figure 5.16. The presence of a larger transverse
Figure 5.18: Lifting of degeneracy for $S^{yz}(Q, \omega) - S^{zy}(Q, \omega)$: the presence of a transverse field unveils response associated with DMI. For small transverse fields, the intensity increases with increasing field. With $J_z = 13.8$ meV, $J_t = \epsilon J_z = 1.9$ meV, $\epsilon \equiv J_t/J_z = 0.137$. The intensity is given in absolute units of meV$^{-1}$ per formula unit.

field only amplifies the intensity; it does not distort the support. This concludes the presentation and results for the two-spinon continuum associated with AFM spin chains. In the next chapter the interchain interaction is taken into account.
Chapter 6

Inelastic scattering cross section for spin-$1/2$ AFM $XXZ$ chain: a comparison with experiment

6.1 Introduction and motivation

Up to this point, the focus has been exclusively on the dependence of the dynamic structure factor on a wavevector along the spin chains. Now, with substantial theoretical foundations laid in the previous chapters, these efforts can be fully exploited to compute the cross section as a function of momentum transfer parallel and perpendicular to the crystallographic $c$-axis for both unpolarised and polarised incident neutrons. Specifically, the interest revolves around the case of spinon pair creation (in the limit of $T = 0$ K) in a system described by spin-$1/2$ AFM $XXZ$ model; these results are compared to experimental data collected at Institute Laue-Langevin (ILL) with a Flatcone multidetector. While 1D correlations are of interest in this section, Chapter 7 delineates the effects of the interaction between neighbouring chains when the system is cooled into the 3D phase.

6.2 Quasi one-dimensional magnetic compound: CsCoBr$_3$

The magnetic compound chosen for the experimental investigation of the spin-$1/2$ excitations of interest is CsCoBr$_3$; it has already been well established [17, 36, 75, 35] that this compound, along with its isomorphs CsCoCl$_3$ and RbCoCl$_3$, exhibit spinon dynamics. More importantly for this research, this antiferromagnetic insulator is an excellent realisation of the spin-$1/2$ AFM $XXZ$ model, with the magnetic ion Co$^{++}$ carrying an effective spin-$1/2$; $J_z = 13.8$ meV and
Figure 6.1: Antiferromagnetic arrangement of spins in a segment of CsCoBr₃ along crystallographic c axis. Co⁺⁺ ions are represented by the green (spin "up") and purple (spin "down") arrows; Br atoms are depicted by cyan spheres; the large yellow spheres correspond to Cs atoms.

\[ J_t = 1.9 \text{ meV} \] [36] are the parameter values in the Hamiltonian

\[
\mathcal{H}_{XXZ} = \sum_{i=1}^{N} J_z S_i^z S_{i+1}^z + J_t (S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) \quad (6.1)
\]

There are two formula units per unit cell in CsCoBr₃ with lattice constants \( c = 6.261 \text{ Å} \) and \( a = 7.445 \text{ Å} \) [36]; the compound possesses a hexagonal configuration in the \( ab \)-plane with space group \( P6_3/mmc \), as shown in Figure 6.2. Co⁺⁺ is the magnetic ion, while Cs ions act as a buffer between spin chains to reduce the interchain interaction and therefore enhance the one-dimensional nature of the compound’s magnetic configuration; the presence of Br accommodates a superexchange pathway for the Co⁺⁺ ions to produce AFM ordering. The three-dimensional ordering temperature is \( T = 28.3 \text{ K} \) [27]; above this, the system enters into the 1D regime. Furthermore, the crystal structure is such that a given Co⁺⁺ ion is surrounded by an octahedron of Br ions, which, as in Figure 6.1, is trigonally distorted. This distortion and non-negligible spin-orbit coupling are treated as perturbations [76]; the result is a ground state consisting of Kramers doublets with effective spin-1/2; the interested reader should refer to Appendix C.
6.3 Inelastic neutron scattering experiment

With this suitable magnetic compound chosen, the scattering experiment itself was performed at Institute-Laue Langevin for the duration of nine days. This facility provides the highest flux of polarized neutrons presently available in the world. At ILL there are cold, thermal and hot neutron sources; IN20 is on a thermal beam tube. This specific instrument utilised is a triple-axis spectrometer (TAS), with Flatcone multianalyser attached. A standard triple axis arrangement allows energy transfer to be measured as a function of momentum transfer [77]: using a monochromator, a particular initial momentum can be selected, with final momentum being chosen using an analyser; this approach also constrains the energy transfer. A schematic of a standard TAS is shown in Figure 6.3, while that of Flatcone setup is shown in Figure 6.4. The advantage of using Flatcone lies in the fact that one can collect many data points simultaneously. Therefore, large areas of reciprocal space can be mapped without the need to increase beam time. Flatcone, in this case, possesses a bank of thirty-one detectors, as shown in Figure 6.4(a). As shown below, due to the arrangement of these detectors, the plots associated with the experimental data will have a peculiar shape consisting of intersecting arcs. The sample of CsCoBr$_3$ was placed in a horizontal cryomagnet where the temperature was lowered and maintained at $T = 40$ K, which is well above 3D ordering temperature of $T = 28.3$ K. The transverse field was generated by a superconducting magnet, with all experimental data produced for $B_x = 3$ T. Finally, Figure B.2 illustrates how a neutron is spin-polarised: the monochromator is magnetised and the magnetisation direction sets the orientation of the incident neutrons. When the spin flipper is active, the polarisation of the incident neutrons is reversed. This permits the comparison of the scattering intensities associated
with each spin orientation of the incident beam; further details are given in Appendix B.

6.4 Inelastic scattering cross section: unpolarised neutrons

Now that the experimental facets of this collaboration have been outlined, theoretical considerations are brought back into focus with attention firstly placed on unpolarised neutrons. In Chapter 4, it is demonstrated how the dynamic spin structure factors can be computed via the Green function framework. However, in order to compare theory and experiment, it is necessary to produce the total inelastic scattering cross section, of which DSFs are just one constituent. More explicitly, one should recall the scattering cross section for unpolarised incident neutrons, derived in Appendix B:

\[
\frac{d^2\sigma}{d\Omega dE} = \kappa_f \left( \frac{2\pi \hbar^2 r_0}{m_n} \right)^2 \left| \frac{1}{2} g F(Q) \right|^2 \sum_{\alpha,\beta} (\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta) S^{\alpha\beta}(Q,\omega),
\]

(6.2)

where the polarisation factor \((\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta)\), as mentioned previously, arises from the fact that the neutron’s spin couples only to the system’s spin fluctuations perpendicular to the scattering vector \(Q\); all other symbols have been defined in Chapter 3. Expanding out the right-hand side of the above, one has
Figure 6.4: Flatcone multianalyser. (a) Top view: flatcone possesses a bank of 31 detectors over an angular range of 75°. (b) Side view: each detector can measure either cold neutrons \( (k_f = 1.4 \text{ Å}^{-1}) \) or thermal \( (k_f = 3 \text{ Å}^{-1}) \). Courtesy of J. Kulda.

\[
\frac{d^2\sigma}{d\Omega dE'} = \frac{\kappa_f}{\kappa_i} \left( \frac{2\pi h^2 r_0}{m_n} \right)^2 \frac{1}{2} g F(Q)|^2 \left[ (1 - \tilde{Q}_x^2) S^{zz}(Q_z, \omega) + S^{yy}(Q_z, \omega) + (1 - \tilde{Q}_z^2) S^{zz}(Q_z, \omega) \right],
\]

where \( Q \rightarrow Q_z \) is momentum transfer parallel to the spin chain and \( Q_x \) is the momentum transfer perpendicular to the chain. Hence, both components are required to compute the inelastic scattering cross section; note that \( \tilde{Q}_y = 0 \) since \( xx \)-scattering only takes place within the scattering plane. It is also necessary to compute the form factor \( F(Q) \), the details of which are outlined in Appendix C. Since the diagonal DSFs have already been computed, one does not require too much more computation to evaluate Equation (6.3). The notable difference now is that, instead of the energy transfer and momentum transfer (along the chain) being the two independent variables, the energy transfer is fixed in the experiment and the independent variables are now \( Q_z \) and \( Q_x \).

### 6.4.1 Comparison of theory and experiment: unpolarised neutrons

Now that all the necessary elements are in place, the theoretical results of the inelastic scattering cross section for unpolarised neutrons are presented below for various energy transfer values, along with experimental data for comparison to test the theoretical basis of this research; all plots shown are displayed in reciprocal lattice units. It should also be strongly emphasised that this is the
first computation of the total inelastic neutron scattering cross section for spin-1/2 AFM $XXZ$ system in presence of a transverse field. Due of the presence of the spin flipper (Figure B.2), the experimental data for the unpolarised cross section are obtained from the polarised neutrons by simply adding the two spin channels; details are given in Appendix B. To provide context to these plots comparing the experimental data for the scattering cross section with that of the Green function results in the case of non-zero transverse field, it is instructive to first look at the case when the transverse field is in fact zero. To achieve this, one relates the scattering cross section back to the two-spinon continuum; Figure 6.6(a) shows a density plot of $S^{xx}(Q, \omega)$ for $B_x = 0$ T. While the scattering cross section includes projection factors and form factors, $S^{xx}(Q, \omega)$ is chosen as it dominates the experimental cross section. The horizontal white line in the plot corresponds to fixing the energy transfer at 15 meV; as can be seen, incommensurable signals occur around $Q_z \in \{0.5, 1.5, 2.5, 3.5\}$ for this energy transfer, with $Q_z$ being the momentum transfer along the chain. When the scattering cross section is then computed for $\omega = 15$ meV, which is displayed in Figure 6.6(b) ($Q_x$ is the momentum transfer perpendicular to the chain), one does indeed observe this incommensurability arising. The intensity diminishes in the cross section the further one goes from the origin, but this is simply related to the presence of the form factor.

Returning now to the central theme of this chapter when the transverse field is non-zero, Figure 6.7 presents the first comparison between theory and experiment for energy transfer $\omega = 13.5$ meV, while Figure 6.8 contains those associated with $\omega = 15.0$ meV. The most prominent feature arising out of both sets of plots, for this case of unpolarised neutrons, is the presence of stripes in the intensity. These correspond to a modulation in momentum transfer along the chain, $Q_z$, but not in momentum transfer perpendicular to the chain, $Q_x$. From the theoretical perspective, this is simply a manifestation that the spin excitations are modelled to occur along the crystallographic $c$-axis; the experimental results corroborate this assumption, with good agreement being displayed between theory and experiment in Figures 6.7(c) and 6.7(d), and Figures 6.8(c) and 6.8(c). This is the first explicit demonstration of the one-dimensional nature of the spin excitations in CsCoBr$_3$. Another salient point to make is that the experimental plots appear to possess virtually no intensity.
for $Q_z \in [3, 4]$ (in reciprocal lattice units). This is due to physical presence of the magnet blocking any neutron flux from being measured. As a further comment, in both theoretical and experimental results, one observes an overall decay in $||Q||$ due to the form factor $F(Q)$, which itself reflects the non-point like distribution of electrons around the nucleus. With these results, best agreement between theory and experiment is obtained for experimental data produced for an energy transfer of $\omega = 13.5$ meV and theoretical results produced at $\omega = 13$ meV, as displayed in 6.7(d); in the case of the experiment plot in Figure 6.8(d) for $\omega = 15.0$ meV, the theoretical inelastic scattering cross section computed for $\omega = 14.25$ meV provides best agreement. A potential reason for this discrepancy may be due to the theoretical framework being based on first order perturbation theory, and that the shift in energy may be associated with second order effects.
(a) Theoretical result: density plot of total inelastic scattering for unpolarised neutrons; $\omega = 14.25 \text{ meV}$ $B_x = 3 \text{ T}$.

(b) Theoretical result: 3D plot of total inelastic scattering for unpolarised neutrons; $\omega = 14.25 \text{ meV}$ $B_x = 3 \text{ T}$.

(c) Theoretical result: density plot above has been cropped for easier comparison with experimental data.

(d) Experimental result: data of total inelastic scattering cross section for unpolarised incident neutrons produced at IN20 at ILL; $\omega = 15 \text{ meV}$ $B_x = 3 \text{ T}$. Experiment performed for $T = 40 \text{ K}$. Courtesy of J. Kulda and H. B. Braun.

Figure 6.8: Comparison between theory and experiment for the total inelastic scattering cross section for unpolarised incident neutrons for fixed energy transfer of $\omega = 15.0 \text{ meV}$. With $J_z = 13.8 \text{ meV}$, $J_t = \epsilon J_z = 1.9 \text{ meV}$, $\epsilon \equiv J_t/J_z = 0.137$. The intensity is given in absolute units of mbarn sr$^{-1}$ meV$^{-1}$ per formula unit.
(a) Theoretical result: density plot of total inelastic scattering cross section for unpolarised neutrons; $\omega = 13.0 \text{ meV}$ $B_x = 3 \text{ T}$.

(b) Theoretical result: 3D plot of total inelastic scattering for unpolarised neutrons; $\omega = 13.0 \text{ meV}$ $B_x = 3 \text{ T}$.

(c) Theoretical result: density plot above has been cropped for easier comparison with experimental data.

(d) Experimental result: data of total inelastic scattering cross section for unpolarised incident neutrons produced at IN20 at ILL; $\omega = 13.5 \text{ meV}$ $B_x = 3 \text{ T}$. Experiment performed for $T = 40 \text{ K}$. Courtesy of J. Kulda and H. B. Braun.

Figure 6.7: Comparison between theory and experiment for the total inelastic scattering cross section for unpolarised incident neutrons for fixed energy transfer of $\omega = 13.5 \text{ meV}$. With $J_z = 13.8 \text{ meV}$, $J_t = \epsilon J_z = 1.9 \text{ meV}$, $\epsilon \equiv J_t/J_z = 0.137$. The intensity is given in absolute units of mbarn sr$^{-1}$ meV$^{-1}$ per formula unit.
6.5 Inelastic scattering cross section: polarised neutrons

Now that unpolarised scattering has been discussed, this section centres on the polarised case which is the primary focus of this work, with results from both the theoretical and experimental perspectives being presented. As explained in Chapter 3 and detailed in Appendix B, the scattering cross section which explicitly takes into account the polarisation of incident neutrons is given by

\[
d\frac{d^2\sigma}{d\Omega dE'} P_x = \frac{\kappa_f}{\kappa_i} \left(\frac{2\pi\hbar^2 \tau_0}{m_n}\right)^2 \frac{1}{2} gF(Q)^2 \left[ \sum_{\alpha,\beta} \left(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta\right) S_{\alpha\beta}^\alpha(Q_x, \omega) \right]
\]

\[+ i P_x (1 - \hat{Q}_z^2) (S_{yz}^y(Q_z, \omega) - S_{zy}^y(Q_z, \omega)) + i P_x \hat{Q}_x \hat{Q}_z (S_{xy}^y(Q_z, \omega) - S_{yx}^y(Q_z, \omega)) \right].
\]

(6.4)

with \( P_x = \pm 1 \) representing the polarisation along the \( x \)-direction. If one averages over this polarisation, one recovers the unpolarised cross section of Equation (6.2). What is measured in the experiment is in fact each spin channel separately. The interesting case arises for scattering intensities that are different for the two spin directions, or equivalently, the following non-zero result

\[
I_+ - I_- \propto \frac{1}{2} gF(Q)^2 \left[ i P_x (1 - \hat{Q}_z^2) (S_{yz}^y(Q_z, \omega) - S_{zy}^y(Q_z, \omega)) + i P_x \hat{Q}_x \hat{Q}_z (S_{xy}^y(Q_z, \omega) - S_{yx}^y(Q_z, \omega)) \right],
\]

(6.5)

with \( I_{\pm} = \frac{d^2\sigma}{d\Omega dE'} \pm \). For the case of \( \text{CsCoBr}_3 \), \( S_{yz}^y(Q, \omega) - S_{yx}^y(Q, \omega) \) is now zero in the perturbation underlying the method of Ishimura and Shiba. Its contribution vanishes since, from Chapter 4, specifically Equations 4.17, one sees that first order perturbation theory yields a change in sign for \( S_{yn}^n \) acting on one Néel state compared to the other, while \( S_{xn}^x \) has no change of sign; this leads to an overall cancellation when \( S_{xy}^y(Q, \omega) - S_{yx}^y(Q, \omega) \) is computed. The focus is therefore solely on \( S_{yz}^y(Q, \omega) - S_{zy}^y(Q, \omega) \) for this case of polarised neutrons and, as highlighted in previous sections, \( S_{yz}^z(Q, \omega) - S_{zy}^z(Q, \omega) \) only produces a non-zero signal when the transverse field is itself non-zero; as in the previous section, the experimental applied field is \( B_x = 3 \) T.

6.5.1 Comparison of theory and experiment: polarised neutrons

For the polarised neutron results, Figure 6.10 displays that both theory and experiment exhibit a finite response for the left-hand side of Equation (6.5). While such a signal is interesting in its own right, it is the connection between \( S_{yz}^y(Q, \omega) - S_{zy}^y(Q, \omega) \) and chirality that is of interest in the context of this research. This is primarily the reason why the magnetic field is applied: analogous
to that of the one-spinon case, the transverse field is present to unveil a “chiral” cross section associated with the two-spinon continuum. The fact that there is a non-zero signal present in both theoretical and experimental results of Figures 6.10(c) and 6.10(d), respectively, indicates the existence of a chiral property associated with the low-lying excitations under investigation. Also, as in the unpolarised case, there is a discrepancy between the energy chosen for the theory (\(\omega = 14.45 \text{ meV}\)) and that of the experiment (\(\omega = 15.75 \text{ meV}\)). Since the technique employed here is based on that of Ishimura and Shiba (IS) [32], and considering that this approach has been demonstrated [79] to not wholly correspond to that of exact results [33, 80] due to it relying heavily on the approximation that \(J_t/J_z \ll 1\), this may well explain the discrepancy that exists between theory and experiment illustrated in Figure 6.10. In addition to this, the presence of stripes in the plots once again advocate the 1D nature of the spin chains in CsCoBr\(_3\). Referring to the experimental data of Figure 6.10(d), one observes a “shadow” for higher values of Q\(_x\) due to the actual design of the transverse magnet which involves “blind spots” for the beam. Also, as a consistency check for the decimation technique involving Green functions, one expects the intensity for the polarised part of the scattering cross section to change sign when the magnetic field direction is reversed. As shown in Figure 6.9 for the case of \(B_x = 3 \text{ T}\) and \(B_x = -3 \text{ T}\), this antisymmetry does indeed arise. Finally, it should also be noted that, while the transverse field produces a finite response in the off-diagonal dynamic spin structure factors, it has also been demonstrated in Chapter 4 that the Dzyaloshinskii-Moriya interaction, if present, has a part to play in producing a non-zero response in \(S^{yz}(Q,\omega) - S^{zy}(Q,\omega)\). However, Given that DMI is not present in many 1D spin chain compounds, the significance of its role in this particular part of the investigation is not a prominent one.

Figure 6.9: Signal for polarised part of scattering cross section for (a) \(B_x = 3 \text{ T}\) and (b) \(B_x = -3 \text{ T}\). The intensity is given in absolute units of mbarn sr\(^{-1}\) meV\(^{-1}\) per formula unit.
(a) Theoretical result: density plot of total inelastic scattering for polarised neutrons; $\omega = 14.45$ meV $B_x = 3$ T.

(b) Theoretical result: 3D plot of total inelastic scattering for $\omega = 14.45$ meV $B_x = 3$ T.

(c) Theoretical result: density plot above cropped for easier comparison with experimental data.

(d) Experimental result: data of total inelastic scattering cross section for polarised incident neutrons produced at IN20 at ILL; $\omega = 15.75$ meV $B_x = 3$ T. Experiment performed for $T = 40$ K. Courtesy of J. Kulda and H. B. Braun.

Figure 6.10: Comparison between theory and experiment for the total inelastic scattering cross section for polarised incident neutrons for fixed energy transfer of $\omega = 15.75$ meV. With $J_z = 13.8$ meV, $J_t = \epsilon J_z = 1.9$ meV, $\epsilon \equiv J_t/J_z = 0.137$. The intensity is given in absolute units of mbarn sr$^{-1}$ meV$^{-1}$ per formula unit.
Chapter 7

Interchain Interaction

7.1 Introduction and motivation

Thus far the theoretical investigation has been in relation to a single spin chain. In actual magnetic compounds exhibiting magnetic configurations with one-dimensional behaviour, such as the perovskites CsCoCl$_3$, CsCoBr$_3$ and RbCoCl$_3$ (all with AFM ordering), there are obviously a myriad of these chains. To experimentally realise this 1D phase, one must be in a temperature regime which is above the 3D ordering temperature so that interchain interactions are so weak as to be negligible. However, then the natural question arises: what happens to the two-spinon continuum, which has been investigated thus far, when the temperature is lowered below the 3D ordering temperature? The answer to this question is the subject of this chapter.

7.2 Interchain Interaction

In CsCoBr$_3$, for example, the 3D ordering temperature occurs at $T_N = 28.3$ K [27]. From a theoretical perspective, Shiba [81] was the first to tackle this problem by building on the work surrounding the excitation continuum; a new term was included in the Hamiltonian to model nearest-neighbour chains using molecular field theory and it was deduced that the new effective interaction was encapsulated by the following

$$H_{II} = h \sum_j (-1)^j S^z_j, \quad (7.1)$$

with $h \geq 0$ and $h \ll J_z$. In order to provide a geometric perspective of how this interchain interaction may arise, Figure 7.1(a) shows the three-dimensional magnetic ordering in the crystallographic $ab$-plane for CsCoBr$_3$ and its isomorphs CsCoBr$_3$ and RbCoCl$_3$. For clarity, only the cobalt atoms are displayed; these are arranged in a hexagonal pattern. The actual spin con-
Figure 7.1: Magnetic 3D ordering for CsCoCl$_3$, CsCoBr$_3$ and RbCoCl$_3$ [82]. (a) For convenience, in the crystallographic $ab$-plane, only the cobalt atoms are shown. The colouring scheme indicates the spin orientation of the atom: spin up is depicted with green, while spin down is shown in magenta. Nearest neighbours of sites $A$ and $B$ have different spin configurations: surrounding site $A$ are spins of opposite orientation (red hexagon); surrounding site $B$ are spins of alternating spin orientation (blue hexagon). (b) The AFM spin chains are directed out of the page in (a) and are shown along the crystallographic $c$ direction; the small cyan spheres represent the Br or Cl ions.

Figuration should also be noted: site $A$ in Figure 7.1(a), for example, has nearest neighbours which all possess opposite spin orientation, while site $B$ is surrounded by nearest neighbours with orientations equally distributed between up and down. Shiba [81] argued that these two spin environments lead to, for site $B$, a cancellation of the magnetic interactions between the chains, so one recovers the continuum, i.e., as if the $B$-site spin chain is completely isolated; site $A$, on the other hand, experiences a non-zero interchain interaction in the nearest-neighbour approximation. The non-trivial result arising from this new interaction is that the continuum is quantised into a set of bound states, which Shiba referred to as a Zeeman ladder. Figure 7.5 displays these states for the structure factor $S^{xx}(Q,\omega)$ for various values of $h$. In order to explain why these states arise, it is convenient to set the basis to be composed of the states employed thus far

$$|\zeta\rangle = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{iQj}|j,j + \zeta\rangle,$$

(7.2)

where $\zeta \in \{1, 2, 3, \ldots\}$, corresponding to the number of flipped spins lying between the two domain walls. Since $\mathcal{H}_{II}$ only contains $S^z$ operators, one immediately realises that the states above are eigenstates of this Hamiltonian:

$$\mathcal{H}_{II}|\zeta\rangle = h\zeta|\zeta\rangle = E_{\zeta}|\zeta\rangle.$$

(7.3)

Therefore, in the presence of this interchain interaction, the energy eigenvalue is dependent on
the number of flipped spins, with the energy cost growing linearly with each new adjacent spin being flipped. It is this dependence which induces the quantization of the excitation continuum into a set of bound states in Figure 7.5. In order to furnish a more physical argument as to why bound states are produced, the interchain interaction can be interpreted as a mechanism which counteracts the delocalising effect of the transverse exchange interaction present in a given chain and so confines the spin cluster into a bound configuration, as indicated in Figure 7.2(a).

In addition to this, another prominent feature that these bound states exhibit is that, in terms of intensities produced by calculating the various dynamic spin structures, there is a monotonic decrease in going from bound states of low energy to those of higher energy. This is illustrated in Figure 7.3 for energy scans of $S_{xx}(Q, \omega)$ at fixed momentum transfer value of $Q = \pi$ for various values of $h$. Such results are in accordance with that of Shiba’s [81] who also demonstrated a monotonic decrease. As an interesting side remark, this particular system can be mapped to one in which two free quarks are confined by a linear potential, as in Figure 7.2(b), leading to the formation of bound states. The two spinons in the context of this research undertake the role of quarks and the bound states represent a type of spinon confinement. Recent experimental work [83] has already been carried out on CoNb$_2$O$_6$, a good realisation of a 1D Ising ferromagnet, with this theme in mind. Furthermore, with this analogy, one can determine the spacing of energies for the observed bound states when the interchain interaction is non-zero for this two-spinon subspace, by taking the continuum approximation so that the discreteness of the lattice is ignored and one can now think of the pair of spinons possessing a reduced mass, $\mu$. Within this approximation and taking into account that the interchain interaction generates a linear potential, the corresponding
Schrödinger equation reads

\[ -\frac{\hbar^2}{\mu} \frac{d^2}{dx^2} \psi(x) + \lambda |x| \psi(x) = (E - 2E_0) \psi(x). \]

(7.4)

The solutions of the above involves Airy functions, leading to the following spectrum \[E_n = 2E_0 + z_n \lambda^{2/3} \left( \frac{\hbar}{\mu} \right)^{1/3}, \quad n = 1, 2, 3, \ldots\] (7.5)

where \(E_n\) is the bound state energy, \(E_0\) is the energy to break the nearest-neighbour exchange interaction, \(\lambda\) is proportional to the longitudinal staggered magnetic field \((\lambda = h/2)\), and the \(z_n\)'s are the negative zeros of the Airy function of the first kind. From Equation (7.5), it is apparent that the spacing of the bound state energies is highly non-trivial. To see how well this model and the concept of “spinon confinement” compares with the results produced by the Green function technique, the example of \(S^{xx}(Q, \omega)\) for \(Q = \pi, J_z = 13.8\) meV, \(J_t = \epsilon J_z, \epsilon = J_t/J_z = 0.137\).
is used; the bound states (labelled 1 - 5) arising from the interchain interaction are shown in Figure 7.4(a). The comparison between the results of Equation (7.5) and that of the Green function technique are displayed in Figure 7.4(b). The points associated with spinon confinement represented by Equation (7.5) are produced for $2E_0 = 9.63 \text{ meV}$ and $\lambda^{2/3} \left( \frac{\hbar}{\mu} \right)^{1/3} = 1.20 \text{ meV}$. In an ideal situation of no transverse exchange, one would expect that $2E_0 = J_z = 13.8 \text{ meV}$. The fact that $2E_0 < J_z$ arises is due to the presence of a finite transverse exchange interaction.

In addition to the above, one notes the presence of forbidden regions where no dynamics occur for the bound states; this is exemplified by the density plot of $S_{xx}(Q, \omega)$ in Figure 7.5(e) and represented by the white lines which mark the boundaries to such regions. The existence of these regions will be explained in Section 7.3. For now it is beneficial to explain the numbering of each excitation branch in Figure 7.5(e). Since an external transverse magnetic field and the Dzyaloshinskii-Moriya interaction are not of concern at this juncture, the various subspaces characterised by the total $z$-component of spin, $S^z_{\text{tot}}$, are disconnected. This implies that one is free to choose one of these subspaces for the investigation at hand; here, within our framework of the generalised Green function technique, the Shiba [81] choice of $S^z_{\text{tot}} = +1$ is employed for convenience. As a consequence, one is only dealing with an odd number of flipped adjacent spins: branch 1 represents a roaming bound state consisting of a single flipped spin, branch 3 consists of three flipped adjacent spins, etc.. Although, since the Green Function technique (the generalised version which includes the interchain interaction is explained below) takes into account all $S^z_{\text{tot}}$ subspaces, the branches labelled by 1, 3, 5 and 7 can be equally interpreted as 2, 4, 6 and 8, respectively; this would correspond to $S^z_{\text{tot}} = 0$, but the Shiba choice is chosen for convenience when only considering transverse and Ising exchange interactions, i.e., when the Hamiltonian is invariant with respect to rotation around the $z$-axis.

It should be noted at this stage that Shiba [81] only computed the structure factor $S^{xx}(Q, \omega)$ case for non-zero interchain interaction with Ising and transverse exchange interactions present along the chain. The work presented here goes far beyond this: building on the Green function technique presented in Chapter 4, this is generalised to include the interchain interaction so that one can fully compute not only $S^{xx}(Q, \omega)$, but $S^{yy}(Q, \omega)$, $S^{zz}(Q, \omega)$ and $S^{yz}(Q, \omega) - S^{zy}(Q, \omega)$, the latter relevant for polarised neutron scattering. In addition to this, the Green function approach, as alluded to, possesses the advantage over the Ishimura and Shiba approach, that one can introduce a transverse magnetic field and/or Dzyaloshinskii-Moriya interaction; these will be discussed in subsequent sections. It is through the use of this algorithm that the plots of Figure 7.5 are produced; an outline of how one develops this new algorithm is presented Section 7.3.

### 7.3 Green function technique for interchain interaction

In order to actually compute the results shown above, the Green function technique explained in detail in Chapter 4 is generalised. However, due to the presence of the interchain interaction,
Figure 7.4: (a) Five bound states arising in spin-1/2 described by antiferromagnetic XXZ model when interchain interaction is present. This is for $S^{xx}(Q, \omega)$ when $J_z = 13.8$ meV, $h = 2\%$ of $J_z$, $Q = \pi$. The intensity is given in absolute units of meV$^{-1}$ per formula unit. (b) Comparing the bound state energies of the data produced by the Green function technique (blue triangles) in Figure 7.4(a)) and that of spinon confinement (orange circles) encapsulated by Equation (7.5).
Figure 7.5: $S^{xx}(Q,\omega)$ density plots for two-spinon system governed by spin-1/2 AFM $XXZ$ model ($H_{XXZ}$) along with interchain interaction, $H_{II}$, of Equation (7.1) for various values of $h$; With $J_z = 13.8$ meV, $J_t = \epsilon J_z = 1.9$ meV, $\epsilon \equiv J_t/J_z = 0.137$. The intensity is given in absolute units of meV$^{-1}$ per formula unit.
Figure 7.6: Mapping of spin system to tight binding system: transverse exchange interaction, signified by $\gamma$, represents next-nearest-neighbour interactions in the tight-binding model. The presence of a transverse magnetic field ($\tau$) or Dzyaloshinskii-Moriya interaction ($\lambda$) maps to nearest-neighbour interaction; interchain interaction ($\nu$) corresponds to a transition back to the initial site, as indicated by the closed loops.

the algorithm itself must be modified so that this new interaction is taken into account. Quantitatively one recognises that, following from Equation (7.3), the transition matrix element associated with the interchain interaction is only non-zero when the “hopping” leads back to the original site:

$$\nu \equiv \langle \zeta | H_{II} | \zeta \rangle = E_\zeta,$$

(7.6)

where $\zeta \in \{1, 2, 3, \ldots\}$. As discussed before, the result indicates that the energy associated with this transition is a function of the number of flipped adjacent spins, $\zeta$, in the spin system, or in the tight-binding picture, which site is occupied; Figure 7.6 illustrates this, with each site in the diagram representing the spin states already introduced:

$$\zeta \equiv |Q; \zeta\rangle = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{iQj} |j, j + \zeta\rangle.$$

As in the previously discussed case where the interchain interaction was absent, the first step in exploiting the powerful Green function technique is to reconfigure the lattice representing the spin system in Figure 7.6 to one which is one-dimensional in nature with effective nearest-neighbour interactions. More importantly, the rearrangement should be such that the surface super site 0 (dashed box) contains all relevant Green function matrix elements; this is all illustrated in Figure 7.3. From Equations (4.48) in Chapter 4, the matrix elements respectively associated with the transitions induced by transverse exchange interaction ($\gamma$), external transverse magnetic field
(τ) and DMI (λ) are given as
\[
\begin{align*}
\gamma & \equiv \langle \zeta | H_{XY} | \zeta + 2 \rangle = \frac{J_1}{2} (1 + e^{-iQ}), & (7.7a) \\
\tau & \equiv \langle \zeta | H_{Bz} | \zeta + 1 \rangle = \frac{g_H B_z}{2} (1 + e^{-iQ}), & (7.7b) \\
\lambda & \equiv \langle \zeta | H_{Dz} | \zeta + 1 \rangle = \frac{i D_z}{2} (1 - e^{-iQ}), & (7.7c)
\end{align*}
\]

As before, the algorithm outputting the Green function begins with the following form
\[
(E - \mathcal{H}) \hat{\mathcal{G}} = \mathbb{I}.
(7.8)
\]

Keeping in mind Equation (7.6), which quantifies the site-dependent energy, one can develop an algorithm for computing the surface Green function by proceeding in a similar fashion to that of chapter 4. However, because of this site-dependent energy, the recursion relations are more complicated than that of Equations (4.66) and Equations (4.67) in Chapter 4. The analogues of Equations (4.66) are
\[
[E^\parallel - \hat{e} s_k(0)] \hat{\mathcal{G}}_{00} = \mathbb{I} + \hat{a}_k(0) \hat{\mathcal{G}}_{2^{k+1},0},
(7.9a)
\]
\[
[E^\parallel - \hat{e} s_k(n)] \hat{\mathcal{G}}_{2^k n,0} = \hat{a}_k(n) \hat{\mathcal{G}}_{n+2^{k+1},0} + \hat{b}_k(n) \hat{\mathcal{G}}_{n-2^{k+1},0},
(7.9b)
\]

and those of Equations (4.67) are
\[
\begin{align*}
\hat{a}_k(n) & = \hat{a}_{k-1}(n)(E^\parallel - \hat{e}_{k-1}(n + 2^k))^{-1} \hat{a}_{k-1}(n + 2^k), & (7.10a) \\
\hat{b}_k(n) & = \hat{b}_{k-1}(n)(E^\parallel - \hat{e}_{k-1}(n - 2^k))^{-1} \hat{b}_{k-1}(n - 2^k), & (7.10b) \\
\hat{e}_k(n) & = \hat{e}_{k-1}(n) + \hat{a}_{k-1}(n)(E^\parallel - \hat{e}_{k-1}(n + 2^k))^{-1} \hat{b}_{k-1}(n + 2^k) & (7.10c) \\
& \quad + \hat{b}_{k-1}(n)(E^\parallel - \hat{e}_{k-1}(n - 2^k))^{-1} \hat{a}_{k-1}(n - 2^k), \\
\hat{s}_k(n) & = \hat{e}_{k-1}(n) + \hat{a}_{k-1}(n)(E^\parallel - \hat{e}_{k-1}(2^k))^{-1} \hat{b}_{k-1}(2^k), & (7.10d)
\end{align*}
\]

with initial conditions
\[
\begin{align*}
\hat{a}_0(n) & = \hat{u}(E^\parallel - \hat{H}_{n+1n+1})^{-1} \hat{u}, & (7.11a) \\
\hat{b}_0(n) & = \hat{u}^\dagger(E^\parallel - \hat{H}_{n-1n-1})^{-1} \hat{u}^\dagger, & (7.11b) \\
\hat{e}_0(n) & = \hat{H}_{nn} + \hat{u}(E^\parallel - \hat{H}_{n+1n+1})^{-1} \hat{u}^\dagger + \hat{u}^\dagger(E^\parallel - \hat{H}_{n-1n-1})^{-1} \hat{u}, & (7.11c) \\
\hat{s}_0(n) & = \hat{H}_{nn} + \hat{u}(E^\parallel - \hat{H}_{n+1n+1})^{-1} \hat{u}^\dagger. & (7.11d)
\end{align*}
\]
In the above, \( n \) refers to a super site (dashed box): \( n = 0 \) is the surface super site; \( n = 1 \) is super site 1, etc., \( k \in \{0, 1, 2, 3, \ldots \} \) and refers to what stage the iterative process is at. Also, \( \mathcal{H}_{nn} \) replaces the on-site super site energy, \( \varepsilon \), and explicitly contains the energy terms due to the interchain interaction:

\[
\widehat{H}_{nn} = \begin{pmatrix}
J_z + h(1 + 4n) & \tau + \lambda & \gamma & 0 \\
\tau^* + \lambda^* & J_z + h(2 + 4n) & \tau + \lambda & \gamma \\
\gamma^* & \tau^* + \lambda^* & J_z + h(3 + 4n) & \tau + \lambda \\
0 & \gamma^* & \tau^* + \lambda^* & J_z + h(4 + 4n)
\end{pmatrix}.
\] (7.12)

Similar to the case of zero interchain interaction,

\[
u = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ \gamma^* & \tau^* + \lambda^* & 0 & 0 \\ 0 & \gamma^* & 0 & 0 \end{pmatrix},
\] (7.13)

From Equation (7.9a), the aim is to obtain the surface Green function matrix, \( \widehat{G}_{00} \) associated with the surface super site 0, or equivalently to have \( a_k(0) \) converge to the zero matrix, with \( k \) being finite. One then obtains

\[
\widehat{G}_{00} \approx [E_{II} - \epsilon s_k(0)]^{-1}.
\] (7.14)

Using the expressions at the start of chapter 5 and keeping in mind that the above Green function has the matrix representation

\[
\widehat{G}_{00} = \begin{pmatrix} G_{11} & G_{12} & G_{13} & 0 \\ G_{21} & G_{22} & G_{23} & G_{24} \\ G_{31} & G_{32} & G_{33} & G_{34} \\ 0 & G_{42} & G_{43} & G_{44} \end{pmatrix},
\] (7.15)

one can fully compute all dynamic spin structure factors of interest.

Returning to the forbidden regions in Figure 7.5(e) and how they arise, one recalls that the interchain interaction counteracts the system’s tendency to accommodate two free elementary excitations and produces bound states. From Equation 7.3, the interchain interaction is directly proportional to the number of flipped spins, \( \zeta \), in a state. In terms of Figure 7.5(e), this means that branches 5 and 7 experience a stronger interchain interaction compared to that of branches of 1 and 3. The transverse exchange, on the other hand, is responsible for the propagation of these spin excitations and therefore attempts to overcome any formation of bound states. Hence, these two interactions are in competition with each other. Furthermore, in relation to Figure 7.3, the
Figure 7.7: Steps in converting spin system into one-dimensional model with effective nearest-neighbour interactions. It is this interpretation of the spin system that permits one to employ the decimation technique involving Green functions. The presence of interchain interaction now permits a transition from a given spin state (or a site in the tight-binding framework) back to itself, as represented by $\nu$. 

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transverse exchange is related to the hopping term, $\gamma$, through Equation (7.7a) and, as mentioned in Chapter 5, this mechanism is completely suppressed for $Q = n\pi/2$, with $n$ odd; from Figure 7.5(e), one sees that the forbidden regions emerge out of these momentum transfer values. In Figure 7.3, when the momentum transfer value approaches $n\pi/2$, the probability of transitioning from site 1 to site 3 weakens (no hopping can occur between site 1 and site 2 since $\tau = \lambda = 0$ for now) and so the probability of a state consisting of three flipped spins existing is diminished. To compound this, the presence of the interchain interaction acts against a transition from site 1 to site 3 as its presence restricts the growth of a spin cluster, as illustrated in Figure 7.2(a). This effectively means that the hopping mechanism associated with $\gamma$ is not only suppressed at the aforementioned momentum transfer values, but now for a range of values surrounding $n\pi/2$; $\gamma$ is weakened to such an extent by the interchain interaction at these values that no transitions can occur between adjacent sites. In addition to this, since the interchain interaction is dependent on the number of flipped spins, or equivalently the site number in Figure 7.3, the momentum transfer range in which transitions are not permitted increases the further one goes up in the excitation branches in Figure 7.5(e), or the further one goes along the top chain of odd sites in Figure 7.3. Since the interchain interaction grows linearly with each flipped spin (Equation 7.3), the forbidden region is bounded by straight lines. With all of this, forbidden regions are therefore a consequence of the states not being realised/sites not being occupied for particular momentum transfer values due to the dominance of the interchain interaction; tantamount to this is that one cannot hop from site 1, in Figure , to another site. This then helps to explain why the lowest-lying excitation branch does not exhibit forbidden regions. This excitation branch corresponds to one flipped spin roaming around the spin chain and is thus related to site 1 in Figure 7.3. With the theoretical framework presented here dealing only with the two-spinon subspace, the system must occupy one of the sites in Figure 7.3; with site 1 costing the least amount of energy in the presence of an interchain interaction, this site/state is the most likely to occur. When $\gamma$ is diminished by the interchain interaction, the system effectively becomes trapped on site 1. As mentioned above, the forbidden regions for each excitation branch are a manifestation that the site cannot be occupied at particular momentum transfer values. However, site 1 can be occupied for all $Q$-values since it does not rely on a transition from a state within the two-spinon subspace of lower energy to be realised; therefore, there are no forbidden regions for this excitation branch.

7.4 Comparison with experiment: RbCoCl$_3$

In this section, experimental data for RbCoCl$_3$, an isomorph of CsCoBr$_3$ and CsCoCl$_3$, are presented along with a comparison to theoretical results produced by the generalised Green function technique just discussed. The experiment itself was performed at the Paul Scherrer Institute (PSI), Villigen, Switzerland, using a spallation neutron source; the magnetic compound was placed in a cryostat maintained at $T = 4$ K. Experimental results for $S^{xx}(Q, \omega)$ are presented in Figure 7.8(a),
for constant momentum transfer value of $Q = \pi$, and in Figure 7.8(b) for constant momentum transfer value of $Q = \pi/2$. Following the argument of Shiba [81], and referring back to site $B$ in Figure 7.1(a) and its nearest neighbours, the signal occurring around 11.2 meV (peak 1) in Figure 7.8(a) and that present around 13.6 meV (peak 5) in Figure 7.8(b) correspond to the continuum of an independent chain. The reason for this is that the interchain interaction is cancelled out due to the spin configuration and the system returns to the case of a 1D spin chain experiencing no interaction from neighbouring chains. Therefore, these particular responses coincide with a continuum arising. One should also be aware that in Figure 7.8(b), the apparent signal produced at approximately 18.5 meV (signal 7) is simply unwanted noise due to the arrangement of the instrumentation. Now focusing on the other peaks in Figures 7.8(a) (2, 3 and 4) and 7.8(b) (peak 6), these do indeed arise from interchain interaction and are related to the spin configuration associated with site $A$ in Figure 7.1(a) where the net effect of the neighbouring chains is non-zero and so bound states emerge.

In order to explain these results from a theoretical perspective, one notes that since the experiment is performed at a very low temperature, it is quite feasible to include next-nearest neighbour interactions along the chain: these interactions are, in general, weak and only become prominent when thermal fluctuations are greatly suppressed. Originally, the inclusion of such an interaction was motivated by gaining better agreement between theory and experiment. This was pointed out by Matsubara et al. [85], who also investigated CsCoCl$_3$ and CsCoBr$_3$ in the low temperature regime using Raman scattering. With all of this, the explicit form of the Hamiltonian is now

$$
\mathcal{H} = \sum_i^N J_z S_i^z S_{i+1}^z + J_t (S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) - J'_z (S_i^z S_{i+2}^z - J'_t (S_i^x S_{i+2}^x + S_i^y S_{i+2}^y)) + h(-1)^i S_i^z,
$$

(7.16)

with $nn$ the nearest-neighbour Hamiltonian; $nnn$ the next-nearest-neighbour Hamiltonian and $II$ is the interchain interaction Hamiltonian; $J'_z > 0$ and $J'_t > 0$ are next-nearest neighbour exchange interactions, with the minus signs being present to reflect the ferromagnetic coupling between next-nearest neighbour magnetic sites.

In order to compare theory with experiment, one employs the Green function technique of Section 7.3 with next-nearest neighbour interaction along the chain now included. This entails additional energy contributions to each site representing a spin state in Figure 7.3; specifically, these are

$$
J'_z - J'_t \cos 2Q \quad \text{for site 1},
$$

(7.17a)

$$
2J'_z - J'_t \cos Q \quad \text{for site 2},
$$

(7.17b)

$$
2J'_z \quad \text{for all other sites}.
$$

(7.17c)
The parameter values used to achieve best agreement between theory and experiment, as illustrated in Figures 7.9 and 7.10, are given in Tables 7.1 and 7.2, respectively. One notes that there are some minor deviations between the set of parameter values associated with each fit. As highlighted by Matsubara et al. [85], these values are not precisely known, with earlier experimental work [86, 36] showing notable differences in these particular quantities. Furthermore, one recalls that the Green function technique is underpinned by first order perturbation theory. With that, the deviations may arise from higher order effects occurring in the system. Referring to Figure 7.9, the plot of best fit does not encompass the experimental signal at approximately 17 meV. This outcome may occur due to the reasons outlined above or it may be related to a possible second interchain interaction originating from next-nearest neighbour chains within the ab-plane in Figure 7.8. Furthermore, in relation to the scaling factor of Tables 7.1 and 7.2 and considering the spin configuration of the cobalt spins displayed in Figure 7.1(a), one would naively think that the ratio should be be 1 : 3 [81]. However, this is an idealisation and the presence of minute thermal fluctuations can disturb the ordering so that the ratio is not 1 : 3. As to why it is 1 : 1.4, this is not entirely clear. Again, this may well go back to the first order perturbation which the Green function technique is based on.
(a) RbCoCl$_3$: energy scan for $Q = \pi$ or, in reciprocal lattice units, $Q = (0, 0, -1)$.

(b) RbCoCl$_3$: energy scan for $Q = \pi/2$ or, in reciprocal lattice units, $Q = (0, 0, -1.5)$.

Figure 7.8: Experimental neutron scattering data for RbCoCl$_3$; performed at PSI, Villigen. Courtesy of M. Mena.
Figure 7.9: RbCoCl\textsubscript{3}: comparison between theory produced by generalised Green function technique with experimental data produced at PSI in Figure 7.8(a). The fit with red line corresponds to that modelled for the case of the continuum; the black line relates to the presence of an interchain interaction. For this energy scan $Q = \pi$. The intensity is given in absolute units of meV\textsuperscript{-1} per formula unit.

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Table 7.1: Comparison between two theoretical fits in Figure 7.9 for energy scan with $Q = \pi$. 
Figure 7.10: RbCoCl$_3$: comparison between theory produced by generalised Green function technique with experimental data produced at PSI in Figure 7.8(a). The fit with red line corresponds to that modelled for the case of the continuum; the black line relates to the presence of an interchain interaction. For this energy scan $Q = \pi/2$. The intensity is given in absolute units of meV$^{-1}$ per formula unit.

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<th>Magenta (interchain interaction) / meV</th>
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Table 7.2: Comparison between two theoretical fits in Figure 7.9 for energy scan with $Q = \pi/2$. 
7.5 Interchain interaction and transverse magnetic field

As mentioned in the opening section of this chapter, Shiba’s work on the formation of bound states out of the continuum was limited to having only the transverse exchange and interchain interactions as perturbations. Here, for the first time, a transverse magnetic field is introduced, similar to the case when the excitations in question formed a continuum. As before, this interaction is represented by $\tau$ in Figure 7.3 and the quantum fluctuations that it induces are responsible for causing, within the two-spinon regime, a single spin to be flipped. In Figure 7.11 below, the results of $S^{\nu\nu}(Q, \omega)$ are presented for various values of $B_x$. The most notable feature of these plots is the emergence of new excitation branches which become more pronounced as the field strength is increased. Furthermore, the gap between branches, which is notable in Figure 7.5 in the absence of a transverse field at higher energy transfer values, is bridged for $B_x \neq 0$ T. This confluence is related to the magnetic field permitting transitions around the momentum transfer of $Q = n\pi/2, n$ odd. In order to explain why these new excitation branches appear, one should recall that the transverse field connects the various subspaces characterised by $S^z_{\text{tot}}$, something which Shiba’s framework fails to cater for. With the presence of the interchain interaction giving a different “on-site” energy for each site in Figure 7.3 (or equivalently, different spin states), governed by Equation (7.3), the transverse field lifts the degeneracy between the top chain of odd sites and bottom chain of even sites, producing new branches that are present in the plots below. Another interesting feature which arises in these plots is the presence of localised regions or “islands” of relatively high intensity around the momentum transfer value of $\pi$ in Figure 7.11. These become more pronounced with increasing magnitude of the transverse field. To explain how these islands originate, we retrace back to the initial case of the two spinon continuum when both the transverse magnetic field and interchain interaction are zero. For this, the dispersion is that of Equation (2.12):

$$\omega(k) = J_z + 2J_t \cos Q \cos 2\kappa,$$

(7.18)

where $Q$ is the total momentum transfer and $\kappa$ is the relative momentum of the two spinons. By varying the value of the relative momentum, one can reproduce the familiar two-spinon continuum; Figure 7.12(a) shows various relative momentum states represented by white lines. Note that around $Q = \pi$ the relative momentum states do not overlap. However, at $Q = n\pi/2$, with $n$ odd, there are indeed “crossing” points; it is at these points where degeneracies are introduced into the continuum and consequentially possess the greatest spectral weight. When the transverse field is then introduced, the dispersion reads

$$\omega(k) = \varepsilon_{k_1} + \varepsilon_{k_2} = J_z + 2J_t \cos Q \cos 2\kappa + 2g\mu_B B_x \cos Q/2 \cos \kappa.$$

(7.19)
The additional term on the right-hand side, that is proportional to $B_x$, is responsible for inducing an interesting property of the relative momentum states: around $Q = \pi$ there are now new crossing points (a whole family in fact) introduced and, similar to the case of $B_x = 0$ T, degeneracies in the system arise at these points. As the transverse field is increased in magnitude, an increasing number of crossing points around $Q = \pi$ arise; this is all encapsulated in Figure 7.12. To fully explain how these islands arise, one must also investigate the effect of the interchain interaction. From Figure 7.5, one observes that the presence of the interchain interaction effectively separates these different relative momentum states. Therefore, when both the transverse field and interchain interaction are present, these crossing points become “visible” and manifest themselves as localised points of high intensity in reciprocal space; in real space and time these points would correspond to standing wave patterns which change with time. As the transverse field is increased, these features become more pronounced and this is apparent in the density plot of $S^{yy}(Q,\omega)$ for $B_x = 15$ T (Figure 7.12).

Furthermore, the results of $S^{yz}(Q,\omega) - S^{zy}(Q,\omega)$ are displayed in Figure 7.14. As in the case when the interchain interaction is absent, this combination of DSFs produces no signal for $B_x \neq 0$ T; this is indicated in Figure 7.14(a). However, when the magnitude of the transverse field is finite, one now observes new excitation branches, just as for $S^{yy}(Q,\omega)$. However, now an asymmetry arises with both positive and negative responses being produced.

Finally, from a general point of view, one may argue that spin wave theory could be applied to assess the accuracy of the Green function technique in the case of bound states arising from the presence of the interchain interaction. However, this approach is only valid for the case of the spin chain possessing one flipped spin which is delocalised, i.e., a magnon. In terms of modes, as in Figure 7.5(e) for example, this corresponds to the lowest lying excitation branch. As one goes up the Zeeman ladder to higher energy modes, the corresponding excitations do not contain a single flipped spin but a cluster of flipped spins. They are therefore not strictly magnons and the spin wave approach breaks down at this very juncture. In fact this is why a novel approach, such as the Green function technique presented here, is required to tackle the issue of several complicated interactions being simultaneously present in the spin chains.
Figure 7.11: $S^{yy}(Q, \omega)$ density plots for two-spin soliton system governed by spin-1/2 AFM $XXZ$ when interchain interaction is considered in the presence of a transverse magnetic field; With $J_z = 13.8$ meV, $J_t = \epsilon J_z = 1.9$ meV, $\epsilon \equiv J_t/J_z = 0.137$. $h = 2\%$ of $J_z$. The colour bar intensity is given in absolute units of meV$^{-1}$ per formula unit.
Figure 7.12: Density plots of $S^{xx}(Q, \omega)$ for various values of the transverse magnetic field. White lines indicate certain relative momentum states from Equation 7.19. The colour bar intensity is given in absolute units of meV$^{-1}$ per formula unit.
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7.6 Interchain interaction and Dzyaloshinskii-Moriya interaction

Similar to the case of the transverse field just discussed, this section delineates for the first time the presence of the Dzyaloshinskii-Moriya interaction in a quasi one-dimensional spin chain experiencing interchain interactions. As highlighted in chapter 3, DMI has, in recent years, come to the fore, in particular in relation to interfacial effects in low-dimensional quantum spin systems; the investigation presented here is therefore appropriate and will hopefully produce new insight about this interaction. In terms of results, Figure 7.15 displays the output for $S^{zz}(Q, \omega)$ and $S^{yz}(Q, \omega) - S^{zy}(Q, \omega)$ for a range of $D_x$ values. Again, as in the case of the transverse magnetic field, one observes new excitation branches. Now, however, one actually sees completely separate branches appearing; this is in contrast to the previous section, where new features arose out of existing branches. Since DMI induces the same transitions as the transverse field in the tight-binding picture, these new responses are related to the various $S_{tot}$ subspaces being connected and how the top and bottom chains in Figure 7.3 are coupled. The difference in how these new features appear may well be related to how the two different interactions originate: DMI is inherent in the system while the transverse field is an external perturbation. In addition to this, DMI is intimately linked with chiral nature of the excitations. For these reasons it would be beneficial to identify systems where DMI plays a role and to perform experiments. It should be noted that CsCoBr$_3$ does not possess DMI since inversion symmetry is not broken. However, DMI can be generated in, for example, spin chains produced on surfaces [63].

In relation to the results of $S^{yz}(Q, \omega) - S^{zy}(Q, \omega)$ depicted in Figure 7.15, it is interesting to observe that a finite signal is produced when DMI is present without the assistance of a transverse field, which was the case when the excitation continuum was being dealt with in Chapter 5. As a final note, since $S^{yz}(Q, \omega) - S^{zy}(Q, \omega)$ is directly linked with polarised scattering, as outlined in Appendix B, the unique intensity signatures in the plots below demonstrate that polarised neutron scattering represents a novel way to detect the presence and strength of the Dzyaloshinskii-Moriya interaction in a given magnetic compound.
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Chapter 8

Conclusions and outlook

The work of this thesis focuses on two main processes occurring in one-dimensional quantum spin chains: spinon scattering and spinon pair creation. These processes are theoretically examined in the context of neutron scattering with the two main aims of this work being (i) the computation of dynamic structure factors in the presence of an external magnetic field applied transverse to the Ising direction and the Dzyaloshinskii-Moriya interaction, and (ii) the exploitation of the influence of these interactions on the various spin systems to gain a deeper understanding of the chiral nature of the excitations in question. In the case of spinon scattering, the dynamic structure factors are obtained analytically within the Villain approximation \cite{28}; while for spinon pair creation, a numerical approach involving a new Green function technique is implemented. By generalising this numerical approach for spinon pair creation, one is able to incorporate the aforementioned interactions into the system; this furnishes information about the chiral property of spinon pair creation. In what follows, the main results of this research are summarised as well as a discussion on potential future work.

Firstly, in Chapter 3, the investigation of spinon scattering is presented for a 1D spin chain described by the spin-\(1/2\) FM XYZ model, the first of its kind; both diagonal dynamic structures and the off-diagonal DSFs, \(S^{yz}(Q,\omega) - S^{zy}(Q,\omega)\), are computed. The introduction of a transverse magnetic field (or Dzyaloshinskii-Moriya interaction) in such a system is motivated by earlier work on spin-\(1/2\) AFM XXZ model \cite{38}, where the presence of a transverse field was used to uncover the chirality associated with spinons in CsCoBr\(_3\). These interactions also lead to the emergence of incommensurable modes, which are themselves signatures of fractional excitations.

Secondly, in terms of spinon pair creation, Chapter 4 describes the development of the novel technique that lies at heart of the computation of much of the results presented throughout this dissertation. It is inspired by the analytical approach employed by Ishimura and Shiba \cite{32} and built upon the work of Sancho et al. \cite{70}. A generalisation of this approach is derived in Chapter 7 so that interchain interactions can be taken into account.

Thirdly, as a means to test the validity of this novel technique, Chapter 6 presents the comparison
between the total scattering cross section in the presence of a transverse field produced by this numerical approach (the first time such a computation has been performed) and that of experimental data of CsCoBr$_3$; the results show good agreement. The results of this chapter also confirm the 1D correlations of spin excitations in CsCoBr$_3$. Furthermore, in Chapter 7, the comparison between experimental scattering data of RbCoCl$_3$ for the case of non-zero interchain interaction and that of the results related to the Green function technique also demonstrate reasonable agreement. Finally, both the results of spinon scattering and spinon pair creation pertaining to polarised incident neutrons CsCoBr$_3$, in the presence of a perturbation (be it a transverse field or DMI), demonstrate a finite response; an indication that a chiral property of these excitations does indeed exist.

In terms of potential future work, it would be desirable to find an improved approximation to the ground state, rather than a perturbed one presented in Chapter 4. This would be incorporated into the Green function approach to evaluate more accurate dynamic structure factors. A possible route to achieve an improved ground state would be to use a Bethe Ansatz treatment. In addition to this, to corroborate the findings of the Green function approach presented in this dissertation, a technique such as DMRG would a suitable choice. This well-known numerical technique can also be extended to investigate systems at finite temperature and therefore would a candidate for investigating spinon scattering. From the experimental perspective, it would be illuminating to
realise spin-1/2 FM $XYZ$ compound so as to investigate the spinon scattering process via neutron scattering. In addition to this, since its upgrade, ILL now possesses a more optimised source of polarised neutrons in the form of IN14 (ThALES); this will aid in producing better statistics for experimental data associated with a spin system’s chiral response.

Finally, it would be interesting to pursue the investigation of the spinon pair creation process in 1D spin system described by the spin-1/2 FM $XYZ$ model. As can be seen in Figure 8.1, within the Green function framework, a 1D spin chain described by the spin-1/2 FM $XYZ$ model accommodates a single bound state as well as the two spinon continuum that was observed for the spin-1/2 AFM $XXZ$ case. There is qualitative agreement between this result and that of the exact one presented in Figure 8.2. It would desirable to take such investigation further and attempt to uncover better agreement and extend the number of interactions present, such as a transverse field and DMI. It would also be appropriate to compute the other diagonal structure factors, as well as $S^{yz}(Q,\omega) - S^{zy}(Q,\omega)$. As another line of inquiry, analogous to the “staggered field” present to produce bound states for spin-1/2 AFM $XXZ$ model, a uniform field in the Ising direction quantises the two-spinon continuum in the FM case; it should be permissible to include this in the Green function framework.

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Appendix A

Green functions

To provide an overview of Green functions, one begins with the general case of a time-dependent Hamiltonian, the Schrödinger equation,

\[ i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = \mathcal{H}(t) |\psi(t)\rangle, \]

(A.1)

which has the formal solution:

\[ |\psi(t_2)\rangle = U(t_2, t_1) |\psi(t_1)\rangle. \]

(A.2)

\(U(t_2, t_1)\) is the time evolution operator and is explicitly given as [88]

\[ U(t_2, t_1) = \sum_{n=0}^{\infty} \frac{1}{n!} \left(-\frac{i}{\hbar} \right)^n \int_{t_1}^{t_2} d\tau_1 \int_{t_1}^{t_2} d\tau_2 \cdots \int_{t_1}^{t_2} d\tau_n \ T \mathcal{H}(\tau_1)\mathcal{H}(\tau_2)\cdots \mathcal{H}(\tau_n) = Te^{-\frac{i}{\hbar} \int_{t_1}^{t_2} d\tau \mathcal{H}(\tau)}, \]

(A.3)

where \(T\) is the time ordering operator. The presence of this operator implies that the time evolution is causal. Yet one is also free to employ a time evolution operator which propagates backwards in time; this is defined as

\[ U(t_1, t_2) = U(t_1, t_2)^{-1} \]

(A.4)

When the Hamiltonian is time-dependent, as in the above, it is standard to refer to the operator (A.3) as the propagator [89] and, from a physical perspective, one is interested in the causal case; causality can be incorporated by introducing a Heaviside function:

\[ K(t_2, t_1) \equiv \theta(t_2 - t_1) \ U(t_2, t_1). \]

(A.5)
For the special case of a time-independent Hamiltonian, which is true for both the spin-1/2 AFM $XXZ$ and spin-1/2 FM $XYZ$ Hamiltonians of interest, Equation (A.3) becomes

$$U(t_2, t_1) = e^{-\frac{i}{\hbar} \mathcal{H}(t_2 - t_1)}, \quad (A.6)$$

In this time-independent case, the propagator is now referred to as the Green function [89], and the causal case has the following form

$$G(t_2, t_1) = \theta(t_2 - t_1) \ e^{-\frac{i}{\hbar} \mathcal{H}(t_2 - t_1)}. \quad (A.7)$$

The basic interpretation of the Green function can be viewed in terms of matrix elements: $\langle r' \vert G(t_2, t_1) \vert r \rangle$ furnishes the probability amplitude of transitioning from a state $\vert r \rangle$ at time $t_1$ to state $\vert r' \rangle$ at time $t_2$. For convenience, one sets $t_1 = 0$, and so

$$G(t) = \theta(t) \ e^{-\frac{i}{\hbar} \mathcal{H} t}. \quad (A.8)$$

Because of the time independence, the total energy associated with the system is conserved. With that, it is informative to transform from the temporal domain to the frequency domain. In order to complete this transformation, it is convenient to work in the eigenbasis associated with the Hamiltonian

$$G(t) = \theta(t) \ \sum_m e^{-i\omega_m t} \vert m \rangle \langle m \vert, \quad (A.9)$$

with $E_m = \hbar \omega_m$. Taking the Fourier transform of the above equation

$$G(\omega) = \int_{-\infty}^{\infty} e^{i\omega t} \ G(t) \ dt = \sum_m \int_{-\infty}^{\infty} dt \ e^{i\omega t} \ \theta(t) \ e^{-i\omega_m t} \ |m\rangle \langle m|, \quad (A.10)$$

and using the well-known integral representation of the Heaviside function [68],

$$\theta(t) = \lim_{\eta \to 0} \frac{i}{2\pi} \int_{-\infty}^{+\infty} \frac{e^{-i\omega t}}{\omega + i\eta} \ d\omega, \quad (A.11)$$
with the pole occurring in the lower-half plane at \(-i\eta\) related to the causality of \(\theta(t)\), one has

\[
G(\omega) = \frac{i}{2\pi} \sum_m \int_{-\infty}^{\infty} d\omega' \left\{ \int \frac{1}{\omega' + i\eta} e^{i(\omega - \omega')t} |m\rangle \langle m| \right\}
\]

\[
= i \sum_m \int_{-\infty}^{\infty} d\omega' \lim_{\eta \to 0} \frac{1}{\omega' + i\eta} \delta(\omega - \omega') |m\rangle \langle m|
\]

\[
= i \lim_{\eta \to 0} \sum_m |m\rangle \langle m| \omega - \omega_m + i\eta.
\]

Using \(E_m = \hbar \omega_m\) and \(H = \sum_m E_m |m\rangle \langle m|\),

\[
G(E) = i\hbar \lim_{\eta \to 0} \frac{1}{E - H + i\eta}(A.13)
\]

It is convenient to redefine \(G(E)\) above as

\[
\mathcal{G}(E) = -\frac{i}{\hbar} G(E)
\]

(A.14)

Setting \(\hbar = 1\), the incarnation of the Green function that will be referred to in the rest of the discussion is

\[
\mathcal{G}(E) = \lim_{\eta \to 0} \frac{1}{E - \mathcal{H} + i\eta}(A.15)
\]

Taking the limit in \(\eta\), one clearly sees that the poles of \(\mathcal{G}(E)\) correspond to eigenvalues of \(\mathcal{H}\). It is this feature of \(\mathcal{G}(E)\) that makes it a meromorphic function [90], and a subset of a larger class of operators known as resolvents (Find Reference), which have the generic form

\[
\mathcal{R}(z) = \frac{1}{z - A},(A.16)
\]

where \(z \in \mathbb{C}\) and \(A\) is a self-adjoint operator. From a physical perspective, the poles correspond to the presence/creation of quasi particles. To unveil a very useful property of the Green function, it is instructive to decompose Equation (A.15) in to its real and imaginary parts:

\[
\mathcal{G}(E) = \sum_m \lim_{\eta \to 0} \frac{(E - E_m)}{(E - E_m)^2 + (\eta)^2} |m\rangle \langle m| - \frac{i\eta}{(E - E_m)^2 + (\eta)^2} |m\rangle \langle m|.
\]

(A.17)

Taking the limit in both terms,

\[
\mathcal{G}(E) = i\hbar \sum_m \frac{1}{(E - E_m)} |m\rangle \langle m| - i\pi \delta(E - E_m) |m\rangle \langle m|,
\]

(A.18)
one is lead to the relationship

\[-\frac{1}{\pi} \text{Im} G(E) = \sum_m \delta(E - E_m) |m\rangle\langle m| \]  

(A.19)

For a basis of states, \{\langle a \rangle\}, not necessarily the eigenbasis, one has

\[-\frac{1}{\pi} \text{Im} \langle a | G(E) | a \rangle = \sum_m \delta(E - E_m) |\langle a | m \rangle|^2 \]  

(A.20)

This is the local density of states, \textit{i.e.}, a weighted density of states. Therefore, when one calculates a diagonal element of the Green function matrix, one is calculating the local density of states associated with that element. The density of states itself, for a discrete spectrum, is given as

\[D(E) = \sum_m \delta(E - E_m), \]  

(A.21)

and so one can immediately deduce, from Equation (A.20), that

\[D(E) = -\frac{1}{\pi} \text{Tr}[\text{Im} \ G(E)], \]  

(A.22)

where \text{Tr} represents the trace operation. Another salient point to note from Equation (A.20) is that

\[\text{Im} \ G(E)_{aa} \leq 0 \]  

(A.23)
Appendix B

Neutron scattering cross section

As has been demonstrated in the main text, neutron scattering is at the heart of the research presented. In this appendix the main constituents of neutron scattering are discussed as well as a delineation of how one arrives at the primary expressions used throughout this dissertation. It should be noted that this is by no means a complete exposition of neutron scattering as this topic is dealt with exhaustively in many textbooks [49, 77, 51].

In such scattering experiments, neutrons are used as a means to furnish information about the sample under investigation, specifically by measuring the scattered flux as a function of the momentum transfer and energy transfer that arises during the scattering events. To quantify this process, the partial differential scattering cross-section is introduced. This quantity represents the number of neutrons scattered into a solid angle element $d\Omega$ with a final energy lying in the range $E'$ and $E' + dE'$ (initial energy is denoted by $E$), and normalised to the incident neutron flux. Therefore, it details the probability that a given incoming neutron is scattered in a certain direction with an energy lying in a particular range; the scattering process is illustrated in Figure B.1. In order to explicitly calculate the partial differential scattering cross section in some tractable way, one must invoke the so-called Born approximation: here, it is assumed that the interaction between the probing neutron and scattering centres is of such small magnitude that the neutron only experiences one scattering event within the sample; this is essentially first order perturbation theory. Furthermore, scattered neutrons can be assumed to take the form of plane waves far enough away from the scattering centres; this is represented by

$$|\kappa_i\sigma_i\rangle = \frac{1}{\sqrt{L^3}} e^{i\kappa_i \cdot r_n} |\sigma_i\rangle, \quad (B.1)$$

where $L$ is the length of the system with periodic boundary conditions understood; $\sigma_i$ is the initial spin state of the neutron; $r_n$ is the position vector of the neutron; $\kappa_i$ is, as already mentioned, the initial momentum of the neutron; the state corresponding to the scattered neutron, $|\kappa_f\sigma_f\rangle$, takes a similar form to that of the above. The assumption of that the neutrons take the form of plane
waves is tantamount to saying that they behave as free particles far from the sample, and this particular feature of the scattering process implies that energy conservation takes the following simple form

\[ \hbar \omega = \frac{\hbar^2 \kappa_i^2}{2m_n} - \frac{\hbar^2 \kappa_f^2}{2m_n}, \]  

(B.2)

where \( m_n \) is the mass of the neutron; \( \omega \) represents the energy transfer to or from the system, depending on whether \( \omega > 0 \) or \( \omega < 0 \). In terms of momentum transfer, the conservation is quantified by

\[ Q = \kappa_i - \kappa_f, \]  

(B.3)

where \( Q \) is referred to as the scattering vector, with \( \kappa_i \) and \( \kappa_f \) representing the initial and final wavevectors of the neutron, respectively; it is \( \kappa_i \) and \( \kappa_f \) which define the so-called scattering plane.

Since one is interested in how the quantum states associated with the sample transition due to the change in quantum states of the probing neutrons, and since one is dealing with first order interactions, the natural starting point is Fermi’s golden rule \[91\]. If one denotes the initial state of the sample by \(| \lambda_i \rangle \), and the final state by \(| \lambda_f \rangle \), then probability per unit time of the scattered neutron transitioning from state \(| k_i \sigma_i \rangle \) to state \(| k_f \sigma_f \rangle \), and the sample going from state \(| \lambda_i \rangle \) to \(| \lambda_f \rangle \) is

\[ W = \frac{2\pi}{\hbar} \sum_{i,f} p_{\lambda_i} |\langle \kappa_i \sigma_i; \lambda_i | V | \kappa_f \sigma_f; \lambda_f \rangle|^2 \delta(h\omega + \varepsilon_{\lambda_i} - \varepsilon_{\lambda_f}), \]  

(B.4)

where \( V \) is the interaction between the neutron and the scattering centres in the sample, with the sum including all possible scattering processes; \( p_{\lambda_i} \) dictates the distribution of initial sample
states when, for example, thermal fluctuations are present; \( \varepsilon_{\lambda_i} \) and \( \varepsilon_{\lambda_f} \) denote the initial and final energies of the sample, respectively. Equation (B.4) relates to the case where only the scattered neutrons propagating with precisely the energy \( E' \) contribute. However, detectors in any experiment are limited by finite precision, with the scattered neutrons with energies in the range \( E' \) and \( E' + dE' \) being measured. Given the dispersion relation inherent in Equation (B.2), the energy interval is

\[ dE' = \frac{\hbar^2 \kappa_f}{m_n} d\kappa_f, \]  

and the direction of the scattered neutron lies in the solid angle element \( d\Omega = \sin \theta d\theta d\phi \). The number of neutrons in this small energy range associated with the state \( |k_f\sigma_f\rangle \) is then

\[ \Delta N = \frac{L^3}{(2\pi)^3} (\kappa^2_f) d\kappa_f d\Omega = \frac{L^3 m_n \kappa_f}{\hbar^2} dE' d\Omega. \]  

Therefore, the probability per unit time of a neutron undergoing a transition from an initial state into a small range of final states is given by \( \Delta N \times W \). The final quantity to compute in order to obtain an expression for the partial differential cross section is the incident neutron flux; this is given by the probability current:

\[ j(\kappa_i) = \frac{\hbar \kappa_i}{m_n}. \]  

The partial differential scattering cross section then follows from \( ||j(\kappa_i)||^{-1} \times \Delta N \times W \):

\[ \frac{d^2\sigma}{d\Omega dE'} = \frac{\kappa_f}{\kappa_i} \left( \frac{m_n}{2\pi \hbar^2} \right)^2 \sum_{i,f} p_{\lambda_i} |\langle \kappa_i\sigma_i; \lambda_i|V|\kappa_f\sigma_f; \lambda_f \rangle|^2 \delta(h\omega + \varepsilon_{\lambda_i} - \varepsilon_{\lambda_f}) \]  

(B.8)

Since the momentum states of the neutrons are assumed to be plane wave states, one can modify the above so that

\[ \frac{d^2\sigma}{d\Omega dE'} = \frac{\kappa_f}{\kappa_i} \left( \frac{m_n}{2\pi \hbar^2} \right)^2 \sum_{i,f} p_{\lambda_i} |\langle \sigma_i; \lambda_i|V(Q)|\sigma_f; \lambda_f \rangle|^2 \delta(h\omega + \varepsilon_{\lambda_i} - \varepsilon_{\lambda_f}), \]  

(B.9)

where \( V(Q) \) is the Fourier transform of the interaction between the neutron and a scattering centre in the sample.

### B.1 Magnetic scattering

With this general framework in place, attention is now brought to the specific type of scattering concerning this research, magnetic scattering. Following from Lovesey [49], and the starting point
to produce an explicit expression for Equation (B.9), the interaction between the magnetic moment of a neutron,

$$\mu = \gamma \mu_N \sigma$$  \hspace{1cm} (B.10)

with a magnetic field $\mathbf{H}$ produced by the sample, is given by

$$V = -\mu \cdot \mathbf{H} = -\gamma \mu_N \sigma \cdot \mathbf{H}.$$  \hspace{1cm} (B.11)

In the above, the gyromagnetic ratio is given as $\gamma = -1.91$, and $\mu_N$ is the nuclear Bohr magneton. According to [92], the magnetic field associated with an electron moving with velocity $\mathbf{v}$ is:

$$\mathbf{H} = \nabla \wedge \left( \frac{\mu_e \wedge \mathbf{r}_e}{|\mathbf{r}_e|^3} \right) + \left( -\frac{e}{c} \frac{\mathbf{v} \wedge \mathbf{r}_e}{|\mathbf{r}_e|^3} \right),$$  \hspace{1cm} (B.12)

where $\mathbf{r}_e$ is the position vector of where the magnetic field is measured, with the position of the electron serving as the origin. Using Equations (B.12) and (B.11) in the expression for the cross section, (B.9), the cross section which arises due to magnetic scattering is [49]

$$\frac{d^2\sigma}{d\Omega dE'} = \left( \frac{m_e}{2\pi h^2} \right)^2 (2\gamma \mu_N \mu_B)^2 (4\pi)^{\frac{K_f}{K_i}} \sum_{\sigma,\sigma_f} \sum_{\lambda_i,\lambda_f} p_{\lambda_i} p_{\sigma_i} (\lambda_i \sigma_i | (\sigma \cdot \mathbf{Q}_\perp)^\dagger | \lambda_f \sigma_f)$$

$$\times (\lambda_f \sigma_f | \sigma \cdot \mathbf{Q}_\perp | \lambda_i \sigma_i) \delta(h\omega + \epsilon_{\lambda_i} - \epsilon_{\lambda_f}),$$  \hspace{1cm} (B.13)

where $\sigma$ are the Pauli spin operators (vector) and Fourier transform of the electron spin component perpendicular to the scattering vector, $\mathbf{Q}$:

$$\mathbf{Q}_\perp = \sum_i e^{i\mathbf{Q} \cdot \mathbf{r}_i} \left( \mathbf{Q}_i \wedge \mathbf{s}_i \wedge \mathbf{Q} - \frac{i}{\hbar|\mathbf{Q}|} \mathbf{Q} \wedge \mathbf{p}_i \right),$$  \hspace{1cm} (B.14)

where $\mathbf{Q} = \mathbf{Q}/||\mathbf{Q}||$; $\mathbf{s}_i$ is the spin operator associated with the $i^{th}$ electron in the sample, while $\mathbf{p}_i$ is the momentum operator. In the above equation, the first term relates purely to the spin property of the electron, while the second is pertinent to the orbital motion of the electron. For spin-only scattering, the focus is only on the first term above; Equation (B.13) then transforms to

$$\frac{d^2\sigma}{d\Omega dE'} = r_0^2 \frac{K_f}{K_i} \sum_{\alpha,\beta} \sum_{\lambda,\lambda'} (\delta_{\alpha\beta} - 
abla(\mathbf{Q} - Q_\alpha \mathbf{Q}_\beta)) \sum_{l,d} p_{\lambda_i} \sum_{l',d'} F_d^*(\mathbf{Q}) F_d'(\mathbf{Q}) e^{i\mathbf{Q} \cdot (R_{l'd'} - R_{ld})} (|\lambda| S_{l'd'}^\alpha |\lambda| \delta(h\omega + \epsilon_{\lambda_i} - \epsilon_{\lambda_f})$$  \hspace{1cm} (B.15)

where $S_{l'd'}^\alpha$ is the component of the total spin associated with a magnetic ion and $\alpha,\beta \in \{x,y,z\}$; $l$ labels the lattice vector going from one unit cell to another and $d$ refers to a magnetic ion within a unit cell; $r_0 = \gamma e^2/m_e c^2 = -0.54 \times 10^{-12}$ cm, with $\gamma = -1.91$ being the gyromagnetic ratio of
the neutron; \( k_i \) and \( k_f \) are the initial and final momentum/momenta of the neutron, respectively; \( p_{\lambda i} \) represents the statistical distribution of the initial states of the system and is present to take into account the more general case of \( T \neq 0 \) K; \( \mathbf{R}_{ld} \) denoting the position vector of the \( d^{th} \) magnetic ion within a unit cell; \( F_d(Q) \), which is discussed in Appendix C is the form factor which is the Fourier transform of the magnetisation of an electron distribution associated with a given ion and arises as one does not deal with the assumed point-like structure of a nucleus but instead an electronic/ spin “cloud” surrounding the nucleus; finally, \( (\delta_{\alpha \beta} - \hat{Q}_\alpha \hat{Q}_\beta) \) is referred to as the polarisation factor and encapsulates an important aspect of neutron scattering events: the probing neutrons can only couple to the system’s spin fluctuations perpendicular to \( Q \). Introducing \( S_{ld}^\perp(t) = \hat{Q} \wedge (S_{ld}(t) \wedge \hat{Q}) \), the above can be recast as

\[
\frac{d^2\sigma}{d\Omega dE'} = \kappa_f \kappa_i \sum_{l,d} \sum_{l',d'} e^{iQ \cdot (\mathbf{R}_{l'd'} - \mathbf{R}_{ld})} \frac{1}{2} g_d F_d^*(Q) \frac{1}{2} g_{d'} F_{d'}(Q) \frac{1}{2\pi \hbar} \int_{-\infty}^{\infty} dt \ e^{-i\omega t} \langle S_{ld}^\perp(0) \cdot S_{l'd'}^\perp(t) \rangle.
\]

(B.16)

In addition to this, if one assumes the form factors to be the same for each site, one is left with

\[
\frac{d^2\sigma}{d\Omega dE'} = \frac{1}{2} g F(Q) \frac{1}{2\pi \hbar} \int_{-\infty}^{\infty} dt \ e^{-i\omega t} \langle S_{ld}^\perp(0) \cdot S_{ld}^\perp(t) \rangle.
\]

(B.17)

Finally, recalling that a dynamic spin structure factor may be written as

\[
S^{\alpha\beta}(Q,\omega) = \int \frac{dt}{2\pi \hbar} e^{-i\omega t} \langle S_{ld}^\alpha(0) S_{ld}^\beta(t) \rangle,
\]

(B.18)

and that

\[
\langle S_{ld}^\perp(0) \cdot S_{ld}^\perp(t) \rangle = \sum_{\alpha,\beta} (\delta_{\alpha \beta} - \hat{Q}_\alpha \hat{Q}_\beta) \langle S_{ld}^\alpha(0) S_{ld}^\beta(t) \rangle,
\]

(B.19)

Equation (B.17) takes the form which makes the explicit connection between the partial differential cross section and dynamic spin structure factors, or in a broader context, the connection between experiment and theory:

\[
\frac{d^2\sigma}{d\Omega dE'} = \frac{\kappa_f}{\kappa_i} \left( \frac{2\pi h^2 r_0}{m_n} \right)^2 |g F(Q)|^2 \sum_{\alpha,\beta} (\delta_{\alpha \beta} - \hat{Q}_\alpha \hat{Q}_\beta) S^{\alpha\beta}(Q,\omega)
\]

(B.20)

This is the case for unpolarised incident neutrons; in the next section the case of polarised neutrons is discussed.
B.2 Polarised scattering

So far, the interest has been only on two degrees of freedom associated with the neutrons probing the sample; these are energy and momentum of the neutron. However, there is also another degree of freedom which has yet to be explicitly incorporated into the framework of the partial differential cross section; this is of course the spin degree of freedom of the neutron, the property responsible for interacting with the magnetic scattering centres of the sample. First implementation of neutron polarization analysis was performed by Moon, Riste, and Koehler [93]. Polarising the incident neutron beam can furnish additional information about the sample. In the case of this research, this additional information surrounds the detection of non-collinear magnetic configurations within a sample [51]; for example, Nakotte et al. observed canting of magnetic moments in UNiGe by employing polarised neutron scattering. The general theoretical formulation of polarised neutron scattering was put in place by Blume [94] and Maleyev; later, Maleyev [95] proposed the investigation of spin chirality using polarised neutron scattering, and was later part of a team that observed [43] its existence in the triangular lattice antiferromagnet, CsMnBr$_3$.

Polarised neutron scattering has also been employed to uncover the spin chirality associated with the elementary excitations in quasi-one dimensional spin chains [38]. To offer detail on how a polarised beam of neutrons is actually produced, we, for simplicity, ignore incoherent scattering. In a generic neutron scattering experimental arrangement, one has a monochromator to select incident neutrons of a certain energy. In the case of polarised neutron scattering, one not only uses the monochromator for this purpose, but also uses it to polarise the neutron beam. The first aspect of this process is applying a magnetic field strong enough so that the internal magnetic moments of the monochromating crystal, which is typically a Heusler alloy, are themselves polarised so that the crystal is essentially a single ferromagnetic domain; this magnetisation of the monochromator is depicted in Figure B.2 and we take the polarisation direction to be along the $z$-direction. The next aspect to consider is when an unpolarised beam of neutrons is passed through this magnetised crystal, Bragg diffraction occurs. However, due to the polarisation of monochromator, the incident neutrons scatter differently, depending on their spin orientation. For the case of elastic scattering, this is quantified in the following expressions for the cross section [77]:

\[
\frac{d\sigma}{d\Omega} \propto (S_N(Q) + S_M(Q))^2, \quad (B.21a)
\]

\[
\frac{d\sigma}{d\Omega} \propto (S_N(Q) - S_M(Q))^2, \quad (B.21b)
\]

where $+$ refers to the neutron spin “up” state in the sense that it is parallel to the applied magnetic field acting on the monochromating crystal; $-$ is the neutron spin “down” state, which is antiparallel to the applied field. Also, $Q$ is the total momentum transfer, and $S_N(Q)$ and $S_M(Q)$ are
Figure B.2: Triple-axis spectrometer with polarisation analysis. The monochromator is magnetised to polarise the incident neutron beam. The spin flipper is in place to reverse the polarisation of the neutron beam, if required. Reference Highlights in ILL Research. Image taken from reference [78].

the \textit{static} nuclear and magnetic structure factors, respectively. Explicitly,

\begin{equation}
S_N(Q) = \sum_m b_{nuc} e^{iQ \cdot r_m},
\end{equation}

where \( r_m \) is the position vector for \( m^{th} \) atomic site and \( b_{nuc} \) is the nuclear scattering length (assumed to be the same for each site). Also, \[51\]

\begin{equation}
S_M(Q) = \sum_m \frac{1}{2} \gamma r_0 F(Q) \langle S \rangle e^{iQ \cdot r_m},
\end{equation}

where \( F(Q) \) is the magnetic form factor, \( \gamma \) is the gyromagnetic ratio, \( r_0 = \gamma e^2/m_e c^2 \) (\( \gamma = -1.91 \) is the gyromagnetic ratio, \( m_e \) is the electron mass), \( S \) is the spin at each ion site (assumed to be the same for all sites). Therefore, if one can locate a reflection for which \(|S_N(Q)| = |S_M(Q)|\), which essentially equates to nuclear and magnetic scattering lengths being of comparable magnitude, then the magnetic and nuclear scattering will cancel and Equation (B.21b) becomes zero while Equation (B.21a) is amplified. Therefore, an unpolarised beam undergoing Bragg diffraction from those planes in the magnetised monochromator will emerge fully polarised. In Figure B.2 the spin flipper, when switched on, reverses the polarisation of the incident neutron beam and its presence allows one to compare the intensities of the two different neutron spin orientations of the incident beam. The disadvantage of using polarised neutron beams is that the intensity is quite low compared to an unpolarised beam, and so the resolution becomes poor. In fact, using a Heusler analyser, the scattered beam can also be polarised but this will only further diminish the intensity, leading to even lower resolution. In order to progress the discussion about polarised inelastic neutron scattering cross section in a theoretical framework, it is useful to continue along path set out by Blume [94]: naturally generalising the expectation value of the neutron spin states
to a density matrix formulation, equation (B.13) can be expressed as

$$\frac{d^2 \sigma}{d\Omega dE'} = \frac{\kappa f}{\kappa_i} \sum_{\lambda, \lambda'} p_{\lambda} \text{Tr} \rho (\lambda | V^\dagger(Q) | \lambda') (\lambda' | V(Q) | \lambda) \delta (\hbar \omega + \varepsilon_{\lambda_i} - \varepsilon_{\lambda_f}), \quad (B.24)$$

where \( \text{Tr} \) represents the trace operation with regard to the neutron states and the density matrix, \( \rho \), concerns the neutron spin. An explicit form of the \( \rho \) can be developed by noting that an arbitrary spin state can be represented by

$$|\chi\rangle = \alpha |\uparrow\rangle + \beta |\downarrow\rangle, \quad (B.25)$$

where the quantisation axis is chosen to be the \( z \)-axis; the coefficients, \( \alpha \) and \( \beta \), satisfy \(|\alpha|^2 + |\beta|^2 = 1\). The corresponding density matrix is \([68]\)

$$\rho = |\chi\rangle \langle \chi| = \begin{pmatrix} |\alpha|^2 & \alpha \beta^* \\ \beta \alpha^* & |\beta|^2 \end{pmatrix}, \quad (B.26)$$

with the normalisation of the spinor ensuring that \( \text{Tr} \rho = 1 \). To elaborate on this and form a more explicit connection with the neutron spin, as is the purpose of introducing the density matrix, one recalls that the Pauli matrices, given by

$$\sigma^1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma^2 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma^3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad (B.27)$$

and the identity matrix, \( I \), form an orthogonal basis for the Hilbert space of Hermitian operators. Therefore, one can express the density matrix as

$$\rho = aI + \sum_i b_i \sigma^i, \quad (B.28)$$

where \( a = \frac{1}{2} \text{Tr} \rho \) and \( b_i = \frac{1}{2} \text{Tr} \sigma^i \rho \). Utilising Equation (B.27) and the fact that the trace of the density matrix returns the identity, one has

$$a = \frac{1}{2} \text{Tr} \rho = \frac{1}{2}, \quad (B.29a)$$

$$b_1 = \frac{1}{2} \text{Tr} \sigma^1 \rho = \text{Re}\{\alpha^* \beta\}, \quad (B.29b)$$

$$b_2 = \frac{1}{2} \text{Tr} \sigma^2 \rho = \text{Im}\{\alpha^* \beta\}, \quad (B.29c)$$

$$b_3 = \frac{1}{2} \text{Tr} \sigma^3 \rho = \frac{1}{2} (|\alpha|^2 - |\beta|^2). \quad (B.29d)$$
Equation (B.28) now takes the form

$$\rho = \frac{1}{2}(\mathbb{I} + \mathbf{P} \cdot \mathbf{\sigma}), \quad \text{(B.30)}$$

with

$$P_1 = 2\text{Re}\{\alpha^* \beta\}, \quad \text{(B.31a)}$$

$$P_2 = 2\text{Im}\{\alpha^* \beta\}, \quad \text{(B.31b)}$$

$$P_3 = |\alpha|^2 - |\beta|^2. \quad \text{(B.31c)}$$

Since, in general, the expectation value of an operator can be cast in the following form [68]:

$$\langle A \rangle = \text{Tr} \ A \rho, \quad \text{(B.32)}$$

where $A$ is a Hermitian operator, it is clear in the above that the coefficients denoted by $b_i$ are related to the expectation values of the Pauli matrices of Equation (B.27). This subsequently implies, with Equations (B.31) in mind, that

$$\mathbf{P} = \langle \mathbf{\sigma} \rangle. \quad \text{(B.33)}$$

$\mathbf{P}$ is referred to as the polarisation vector. If $\|\mathbf{P}\| = 1$ then the beam is completely polarised with all the neutron spins aligned parallel to each other, i.e., a neutron is in a pure quantum state; if $\|\mathbf{P}\| = 0$, the beam is unpolarised, while $0 < \|\mathbf{P}\| < 1$ is referred to as a partially polarised beam.

Following Lovesey’s [49] derivation for inelastic magnetic scattering, Equation (B.24), in conjunction with Equation (B.30), becomes

$$\frac{d^2 \sigma}{d\Omega dE'}_{\mathbf{P}} = \frac{\kappa_f}{\kappa_i} \sum_{l,d} \sum_{l',d'} e^{iQ \cdot (\mathbf{R}_{l'd'} - \mathbf{R}_{ld})} \frac{1}{2} g_d F_d^* (Q) \frac{1}{2} g_{d'} F_{d'} (Q) \frac{1}{2\pi \hbar} \times \int_{-\infty}^{\infty} dt \ e^{-i\omega t} \{ \langle \mathbf{S}_{ld} \cdot \mathbf{S}_{l'd'} (t) \rangle + i \mathbf{P} \cdot \langle \mathbf{S}_{ld} \wedge \mathbf{S}_{l'd'} (t) \rangle \}. \quad \text{(B.34)}$$

The equation above is similar to that of Equation (B.20), with all symbols explained above. Now, however, one has an additional term which explicitly depends on the polarisation vector, $\mathbf{P}$; this couples to the cross product of spin operators perpendicular to the scattering vector. If one again assumes that the form factor is the same for each site, then

$$\frac{d^2 \sigma}{d\Omega dE'}_{\mathbf{P}} = \frac{\kappa_f}{\kappa_i} \left( \frac{2\pi \hbar^2 r_0}{m_n} \right)^2 \frac{1}{2} g F (Q) \sum_{\lambda} \int dt \ e^{-i\omega t} p_{\lambda} \{ \langle \mathbf{S}_{-Q} \cdot \mathbf{S}_{Q} (t) \rangle + i \mathbf{P} \cdot \langle \mathbf{S}_{-Q} \wedge \mathbf{S}_{Q} (t) \rangle \}, \quad \text{(B.35)}$$

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where the Fourier transform of $S_{ld}(t)$, denoted by $S_Q(t)$, is implicitly taken. For the case of $T = 0$ K, as for the spinon-pair creation, thermal fluctuations are absent and so the term $p_\lambda$ is omitted:

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{\kappa_f}{\kappa_i} \left( \frac{2\pi \hbar^2 r_0}{m_n} \right)^2 \left| \frac{1}{2} \mu F(Q) \right|^2 \int dt \frac{2\pi}{2\pi} e^{-i\omega t} \left\{ \langle S_{-Q}(0) \cdot S_Q(t) \rangle + i P \cdot \langle S_{-Q}(0) \wedge S_Q(t) \rangle \right\}$$

(B.36)

### B.3 Inelastic scattering cross section

In a typical neutron scattering experiment, the sample is orientated such that one of the main crystallographic planes coincides with the scattering plane; with that, the momentum transfer vector, $Q$, only has two components, with $Q_y$ chosen by convention to be zero. Furthermore, for technical reasons, the polarisation of the neutrons is along the direction of the applied magnetic field ($x$-direction): $P = P_x \hat{x}$, and $P_x = \pm 1$. Therefore, the polarisation term in Equation (B.36) above maps to

$$P \cdot \langle S_{-Q}(0) \wedge S_Q(t) \rangle \rightarrow P_x \cdot \langle (S_{-Q}(0) \wedge S_Q(t))_x \rangle$$

(B.37)

Expanding on this

$$P_x \cdot \langle (S_{-Q}(0) \wedge S_Q(t))_x \rangle = P_x (1 - \hat{Q}_y^2) \langle (S_{-Q} \wedge S_Q(t))_x \rangle + P_x \hat{Q}_y \hat{Q}_z \langle (S_{-Q} \wedge S_Q(t))_z \rangle.$$  

(B.38)

It should be highlighted at this point that $Q_z$ is the momentum transfer along the chain, while $Q_x$ represents the momentum transfer perpendicular to the chain. Furthermore, using Equation (B.38), one has

$$\int \frac{dt}{2\pi \hbar} e^{-i\omega t} \langle P \cdot (S_{-Q} \wedge S_Q(t)) \rangle = P_x (1 - \hat{Q}_y^2) (S_{yz}(Q,\omega) - S_{zy}(Q,\omega)) + P_x \hat{Q}_x \hat{Q}_z (S_{z}(Q,\omega) - S_{z}(Q,\omega))$$

(B.39)

Finally, noting that

$$\int \frac{dt}{2\pi \hbar} e^{-i\omega t} \langle S_{-Q}(0) : S_Q(t) \rangle = \sum_{\alpha,\beta} \langle \delta_{\alpha,\beta} - \hat{Q}_i \hat{Q}_j \rangle S^{\alpha,\beta}(Q,\omega),$$

(B.40)
Equation (B.36) can be rewritten as

\[
\frac{d^2\sigma}{d\Omega dE_p} = \frac{\kappa_f}{\kappa_i} \left( \frac{2\pi \hbar^2 r_0}{m_n} \right)^2 \frac{1}{2} |gF(Q)|^2 \left[ \sum_{\alpha,\beta} (\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta) S^{\alpha\beta}(Q,\omega) \right.
\]
\[+ i P_x (1 - \hat{Q}_z^2) (S^{yz}(Q,\omega) - S^{zy}(Q,\omega)) + i P_x \hat{Q}_z \hat{Q}_z (S^{xz}(Q,\omega) - S^{zx}(Q,\omega)) \]
\[\left. \right] \right). \tag{B.41}
\]

For the cases of the spinon scattering and spinon pair creation, the term \(S^{yz}(Q,\omega) - S^{zy}(Q,\omega)\) produces such a weak intensity that it is ignored. Therefore, since the polarised term is connected with unveiling helical magnetic configurations, with a pitch along the \(x\)-direction, it is \(S^{yz}(Q,\omega) - S^{zy}(Q,\omega)\) which aids in the detection of the chiral nature of the excitations in question.

### B.4 Derivation of Equation (3.22) in Chapter 3

In order to derive Equation (3.22) in section 3.4, one recalls for the case of spinon scattering that

\[
S^{yz}(q,\omega) - S^{zy}(q,\omega) = -i \frac{1}{N} \sum_k \frac{e^{-\beta\epsilon_k}}{z} \sin\left(\frac{k - q/2}{\sin q/2} \delta(\omega - \epsilon_{k'-q} + \epsilon_k)\right), \tag{B.42}
\]

with \(q\) being the momentum transfer along the chain in the first Brillouin zone. As alluded to in the main text, in a polarised neutron scattering experiment, it is the difference between the two spin orientations of the incident polarised neutrons that is measured. Using Equation (B.41), along with the equation above, the unpolarised (diagonal) term vanishes and one is left with

\[
I_+ - I_- \propto \frac{2(1 - \hat{Q}_z^2)}{N \sin q/2} \sum_k \frac{e^{-\beta\epsilon_k}}{z} \sin \left(\frac{k + k'}{2}\right) \delta(\omega - \epsilon_{k'} + \epsilon_k), \tag{B.43}
\]

where \(I_\pm = \frac{d^2\sigma}{d\Omega dE_p} \pm\) and \(\hat{Q}_z\) is the component of the wavevector transfer unit-vector along the spin chain.
Appendix C

Magnetic form factor

The magnetic form factor is the Fourier transform of the magnetisation of an electron distribution associated with a given ion and arises as one does not deal with the assumed point-like structure of a nucleus but instead an electronic “cloud” surrounding the nucleus; the larger this cloud is, the more rapidly the form factor decrease in reciprocal space. From Squires [51], the magnetic form factor, denoted by \( F(Q) \),

\[
F(Q) = \int e^{iQ \cdot r} \rho(r) \, dr
\]

where \( Q \) is the scattering vector, and \( \rho(r) \) is the normalised spin density at position \( r \) of the unpaired electrons in a given magnetic ion. When the magnitude of the scattering vector, \( Q \), is much smaller than the radius of the radial wavefunction associated with the unpaired electrons, which are responsible for magnetic scattering of neutrons, the magnetic form factor assumes the following simplified form [51]

\[
\frac{1}{2} g F(Q) = \frac{1}{2} g_s \langle j_0(Q) \rangle + \frac{1}{2} g_L \left( \langle j_0(Q) \rangle + \langle j_2(Q) \rangle \right),
\]

where \( j_0 \) and \( j_2 \) are spherical Bessel functions of the 0\(^{th}\) and 2\(^{nd}\) order, respectively; \( g_s \) and \( g_L \) are given as

\[
g_s = \frac{J(J + 1) - L(L + 1) + S(S + 1)}{J(J + 1)}, \tag{C.3a}
\]
\[
g_L = \frac{J(J + 1) + L(L + 1) - S(S + 1)}{2J(J + 1)}, \tag{C.3b}
\]

with \( g = g_L + g_s \). Note that the form factor in Equation (C.2) now depends solely on the magnitude of the scattering vector, \( i.e., \) it is now an isotropic function. This scenario encapsulates the dipole approximation [51], which is approximation employed for calculations pertaining to this research. To calculate the expectation values of the spherical Bessel functions in Equation (C.2),
one follows the work of Brown [87] who deduced the following forms from the work of Forsyth
and Wells [96], and Lisher and Forsyth [97]:

$$\langle j_0(s) \rangle = A e^{-as^2} + B e^{-bs^2} + C e^{-cs^2} + D, \quad (C.4a)$$

$$\langle j_2(s) \rangle = (A e^{-as^2} + B e^{-bs^2} + C e^{-cs^2} + D) s^2, \quad (C.4b)$$

where $s = Q/4\pi$ and $Q = ||Q||$; the parameters $A$, $a$, $B$, $b$, $C$, $c$ have been tabulated Brown and
are ion-specific. In the case of this research, $\text{Co}^{2+}$ is of interest. Table C.1 below contains all
relevant parameter values to compute Equation (C.2).

The values of $S$, $L$, and $J$ in Equation C.2 have been determined by Lines [76] for the compound
$\text{CoCl}_2$, which is structurally very similar to $\text{CsCoBr}_3$. In his work, the crystal field splitting
and spin-orbit coupling were introduced as perturbations. The calculations lead to ground state
consisting of a Kramer’s doublet

$$|m_J = +\frac{1}{2} \rangle = c_1|1,\frac{3}{2} \rangle + c_2|0,\frac{1}{2} \rangle + c_3|1, -\frac{1}{2} \rangle, \quad (C.5)$$

$$|m_J = -\frac{1}{2} \rangle = c_1|1, -\frac{3}{2} \rangle + c_2|0, -\frac{1}{2} \rangle + c_3|1, \frac{1}{2} \rangle. \quad (C.6)$$

It is clear from the above that $S = 3/2$, $L = 1$ and $J = 5/2$. Therefore the system possesses
an effective spin-$1/2$. The coefficients, $c_1$, $c_2$ and $c_3$ have been determined by Tellenbach [98] for
$\text{CsCoCl}_3$, an isomorph of $\text{CsCoBr}_3$. The necessary ingredients to compute Equation (C.2) are
now all available.

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</table>

Table C.1: Parameter values of $\text{Co}^{2+}$ [87].
Bibliography


[81] H. Shiba, Progress of Theoretical Physics 64, 466 (1980).


[91] W. Pauli, Hirzel 4, 60 (1928).


