Magnetization and spin gap in two-dimensional organic ferrimagnet BIPNNBNO

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Abstract

A magnetization process in the two-dimensional ferrimagnet BIPNNBNO is analyzed. The compound consists of ferrimagnetic \((S, s = 1/2)\) chains coupled by two sorts of antiferromagnetic interaction. Whereas the behavior of the magnetization curve in higher magnetic fields can be understood within a process for the separate ferrimagnetic chain, the appearance of the singlet plateau at lower fields is an example of non-Lieb–Mattis type ferrimagnetism. By using the exact diagonalization technique for finite clusters of size \(4 \times 6, 4 \times 8\) and \(4 \times 10\) we show that the interchain frustration coupling plays an essential role in stabilization of the singlet phase. These results are complemented by an analysis of four cylindrically coupled ferrimagnetic \((S, s = 1/2)\) chains via an Abelian bosonization technique and an effective theory based on the XXZ spin-1/2 Heisenberg model when the interchain interactions are sufficiently weak/strong, respectively.

1. Introduction

During the past fifteen years, two-dimensional (2D) quantum spin systems have attracted much attention both from theoretical and experimental physicists. A competition between conventional classically ordered phases and more exotic quantum ordered phases lies at the focus of the investigations. Magnetic systems with a finite correlation length at zero temperature and a finite spin gap above the singlet ground state, spin liquids, realize the Haldane prediction at the level of two space dimensions [1].

To date, one can distinguish two main routes in studies of 2D spin gap compounds. A formation of spin gap in spin dimer systems, for example \(\text{SrCu}_2(\text{BO}_3)_2\) [2] and \(\text{CaV}_4\text{O}_9\) [3], is explained by a modified exchange topology similar to the Shastry–Sutherland lattice [4]. Another way to increase quantum fluctuations and stabilize a spin liquid ground state is realized in kagome antiferromagnets [5]. Experimental candidates for 2D kagome antiferromagnets are currently available: herbertsmithite [6–8] and volborthite [9, 10]. Both these strategies deal with antiferromagnetic compounds. In view of this, an observation of a singlet ground state with a pronounced spin gap in 2D ferrimagnetic material BIPNNBNO seems exotic [11].

The crystal structure of BIPNNBNO is shown in figure 1. A magnetic unit of the spin system presents organic triradical BIPNNBNO. Each of the molecules includes three \(s = 1/2\) spins (see figure 2) with intramolecular ferromagnetic \(J_F\) and antiferromagnetic \(J_{AF}\) interactions. The magnitude of \(|J_F| \sim 300\) K is very large, and two spins coupled ferromagnetically behave as a \(S = 1/2\) moiety. Ferrimagnetic chains are stretched along the \(b\)-axis. There are two kinds of antiferromagnetic interchain interaction along the \(a\)-axis. One is between the \(S = 1/2\) spins, which connects the nearest neighboring chains. The other is between \(S = 1\) species, which connects the next-nearest neighboring chains and introduces spin frustration.

The puzzle is the following. It is well known that the low-energy physics of an isolated ferrimagnetic \((S, s)\) chain corresponds to a gapless \((S = s)\) ferromagnet [12]. Quite predictably one might expect the appearance of an ordered state with fluctuations in the form of spin waves near the classical state. However, measurements of magnetization show that an opening of a gap by analogy with the Haldane chain is the likely scenario. Such a behavior is a manifestation...
of non-Lieb–Mattis type ferrimagnetism [13]. Namely, the magnetization measured at 400 mK is nearly zero below 4.5 T, increases rapidly above 4.5 T, and exhibits a broad 1/3 plateau and a narrow 2/3 one at 7–23 T and around 26 T, respectively. Above 29 T, the magnetization is completely saturated [11].

The purpose of the paper is to investigate the magnetization process. The problem is complicated by a lack of reliable information about intra- and interchain exchange interactions. So, before studying the 2D ferrimagnetic system BIPNNBNO we develop a simple quantum mechanical approach that models a magnetization process of the ferrimagnetic chain (1, 1/2). The treatment agrees qualitatively with predictions of the theory for quantum spin chains [14] and provides reasonable estimations of the exchange intrachain couplings. In addition, it captures a peculiarity of the magnetization process in the prototype 2D material, i.e. the appearance of the intermediate 2/3 plateau. Given these estimations we examine the magnetization process in BIPNNBNO by analyzing exact diagonalization (ED) calculations for finite clusters of size $N = 24, 32$ and 40. A main conclusion to be drawn from these calculations is that the emergence of the anomalous singlet plateau is a consequence of the frustrating interchain interaction.

Two different mechanisms of formation of the plateau may be likely candidates: a generalization of Haldane’s conjecture to the weakly coupled ferrimagnetic chains, and a valence-bond (dimerized) type ground state in the strong-coupling limit. To determine which of these scenarios is relevant we develop low-energy effective theories for the 4-legs spin tube, which forms a minimal setup including the interchain couplings. In the regime of weakly coupled spin tube legs we apply an Abelian bosonization technique. The opposite limit of a strong ring interaction is analyzed in terms of an effective Heisenberg XXZ model, where the intrachain coupling is perturbatively taken into account. Our analytical treatment shows that only the first approach confirms an important role of frustration in stabilization of the singlet phase.

Note that a study of spin tubes is of interest by itself because both frustration and quantum fluctuation are strong [15]. Our model is directly related to the compound BIPNNBNO, but the main results are expected to apply to other frustrated spin tubes as well. Recently, it has been reported that the experimental candidate for the four-leg spin tube, Cu$_2$Cl$_2$-D$_3$C$_3$SO$_2$, is available [16].

The paper is organized as follows. In section 2 we consider the magnetization process in a ferrimagnetic chain (1, 1/2). In section 3 we discuss results of the magnetization process in a two-dimensional ferrimagnetic system obtained via the exact diagonalization method on a finite cluster.
In section 4 we derive the effective low-energy spin-1/2 Hamiltonian. A bosonization study of the spin tube is carried out in section 5. A discussion of these results is relegated to the conclusion section.

2. Magnetization of an isolated ferrimagnetic chain

The issue that we address below is whether a calculation for an isolated ferrimagnetic chain partially reproduces features of the magnetization curve observed in the BIPNNBNO crystal. We demonstrate that both the 1/3 plateau and the 2/3 plateau can be recovered within a simple quantum mechanical analysis of a magnetization process of an isolated quantum (1, 1/2) ferrimagnetic chain under an applied magnetic field.

For the ferrimagnetic spin chain \((S, s)\), a criterion of quantization [17] predicts an appearance of magnetization plateau with \(N\) magnetization. In the case \((S, s) = (1, 1/2)\) it explains both the ground state magnetization plateau with \(N/2\), which corresponds to the 1/3 plateau in the BIPNNBNO system, and the 3N/2 plateau with the saturated magnetization. Obviously, the 2/3 plateau requires a special treatment.

We start from the values of the critical fields derived from the energies \(E(M_{\text{tot}})\) of states with a given magnetization \(M_{\text{tot}}\) of the total chain [18]

\[
B_1 = \frac{\partial E_1}{\partial m} \bigg|_{m=1/3+0} \approx E(N/2 + 1) - E(N/2),
\]

\[
B_2 = \frac{\partial E_2}{\partial m} \bigg|_{m=2/3-0} \approx E(N) - E(N - 1),
\]

\[
B_3 = \frac{\partial E_3}{\partial m} \bigg|_{m=2/3+0} \approx E(N + 1) - E(N),
\]

\[
B_{\text{sat}} = \frac{\partial E}{\partial m} \bigg|_{m=1-0} \approx E(3N/2) - E(3N/2 - 1).
\]

Here \(E, m = M_{\text{tot}}/N\) are the energy and the magnetization per elementary cell of the \((1, 1/2)\) ferrimagnetic chain, \(N\) is a number of the elementary cells.

To estimate the energies in the right-hand side of equations (1), we use the Hamiltonian of the one-dimensional quantum (1, 1/2) ferrimagnet

\[
\hat{H}_c = J_\text{AF} \sum_{i=1}^{N} \hat{S}_1 \cdot \hat{S}_2 + J_1 \sum_{i=1}^{N} \hat{S}_2 \cdot \hat{S}_{i+1}
\]

\[(S_1 = 1, \quad s_2 = 1/2),\]

with the intrachain exchange parameters \(J_\text{AF}\) and \(J_1\), and construct the required quantum states \(|SM\rangle\) with the given quantum numbers of the total spin \(S\) and its \(z\)-projection \(M\).

The 1/3 magnetization plateau corresponds to the ground state of the \((1, 1/2)\) ferrimagnetic chain with \(S = N/2\). The wavefunction of the polarized state is given by

\[
|N/2, N/2\rangle = \prod_{i=1}^{N} \left| \left( \frac{1}{2} \right) \left( \frac{1}{2} \right) \right\rangle,
\]

and presents a direct product of the spin states of the magnetic elementary cells \((1, 1/2)\) (see figure 3). The energy eigenvalue equals

\[
E(N/2) = -J_\text{AF}N - \frac{1}{9}J_1N.
\]

With an increase of the magnetic field the ground state (3) is destroyed and the state with \(S = N/2 + 1\) stabilizes. A low-lying excitation may be qualitatively considered as the forming of one triplet bond. The corresponding wavefunction is

\[
|N/2 + 1, N/2 + 1\rangle = \frac{1}{\sqrt{N}} \times \sum_{k=1}^{N} \left[ \prod_{i(\neq k)=1}^{N} \left| \left( \frac{1}{2} \right) \left( \frac{1}{2} \right) \right\rangle \times \left( \frac{1}{2} \right) \left( \frac{3}{2} \right) \right] = \sum_{k=1}^{N} \alpha_k \Psi_k.
\]

It is composed from all arrangements of the excited block within the chain taken with equal weights \(\alpha_k\).

By introducing the state and calculating the matrix element

\[
\langle \Psi_k | \hat{H}_c | \Psi_k' \rangle = [E(N/2) + \frac{7}{18}J_\text{AF} + \frac{7}{18}J_1 - E(N/2 + 1)] \times \delta_{kk'} - \frac{1}{3}J_1 \delta_{kk'\pm1}
\]

one obtains the relationship for the coefficients \(\alpha_k\)

\[
\frac{1}{3}J_1 \alpha_k - 1 \left[ E(N/2) + \frac{1}{3}J_\text{AF} + \frac{7}{18}J_1 - E(N/2 + 1) \right] \times \alpha_k - \frac{1}{3}J_1 \alpha_{k+1} = 0,
\]

which is tantamount to

\[
E(N/2 + 1) = E(N/2) + \frac{7}{18}J_\text{AF} + \frac{7}{18}J_1 - \frac{1}{3}J_1 \left( \frac{\alpha_{k-1}}{\alpha_k} + \frac{\alpha_{k+1}}{\alpha_k} \right).
\]

This expression includes two independent variational parameters \(\alpha_{k-1}/\alpha_k\) and \(\alpha_{k+1}/\alpha_k\). The minimal value

\[
E_{\text{min}}(N/2 + 1) = E(N/2) + \frac{2}{3}J_\text{AF} - \frac{1}{3}J_1
\]

is reached provided \(\alpha_{k-1}/\alpha_k = \alpha_{k+1}/\alpha_k = 1\). This yields the critical magnetic field \(B_1\) destroying the 1/3 plateau

\[
B_1 = \frac{1}{2}J_\text{AF} - \frac{7}{18}J_1.
\]

Note that a calculation of elementary excitations of the Heisenberg ferrimagnetic chain (1, 1/2) based on a perturbation approach from the decoupled-dimer limit yields for the gapped antiferromagnetic branch (see equation (3.7) in [19])

\[
\omega_k^{\text{AFM}} = \frac{1}{2}J_\text{AF} + \frac{1}{3}J_1(7 - 12 \cos 2k) + O(n^2),
\]

where \(n = J_1/J_\text{AF}\). In the zero momentum limit, the result (10) for the critical field \(B_1 = \omega_k^{\text{AFM}}\) is recovered.

To find the critical fields \(B_2\) and \(B_3\) of the beginning and the end of the 2/3 plateau, respectively, we construct the trial
which are schematically shown in figure 3.

Figure 3. Schematic picture of states of the ferrimagnetic chain used in the construction of a magnetization curve. Excited blocks are marked by the gray shadow.

states

\[
|N, N\rangle = \prod_{i=1}^{N/2} \left(\frac{1}{2}\right)^{1/2} \left(\frac{1}{2}\right)^{1/2} \left(\frac{3}{2}\right)^{1/2} \left(\frac{3}{2}\right)^{1/2},
\]

(12)

\[
|N-1, N-1\rangle = \frac{1}{\sqrt{N}} \sum_{k=1}^{N/2} \prod_{i=1}^{k-1} \left(\frac{1}{2}\right)^{1/2} \left(\frac{1}{2}\right)^{1/2} \left(\frac{3}{2}\right)^{1/2} \left(\frac{3}{2}\right)^{1/2},
\]

(13)

\[
|N+1, N+1\rangle = \frac{1}{\sqrt{N}} \sum_{k=1}^{N/2} \prod_{i=1}^{k-1} \left(\frac{1}{2}\right)^{1/2} \left(\frac{1}{2}\right)^{1/2} \left(\frac{3}{2}\right)^{1/2} \left(\frac{3}{2}\right)^{1/2},
\]

(14)

which are schematically shown in figure 3.

In the same manner we obtain

\[
B_2 = \frac{5}{2} J_{AF} + \frac{7}{18} J_1, \quad B_3 = \frac{5}{2} J_{AF} + \frac{5}{9} J_1.
\]

(15)

The saturation field \(B_{sat}\) is determined with an aid of the wavefunction of the fully polarized state

\[
|3N/2, 3N/2\rangle = \prod_{i=1}^{N} \left(\frac{1}{2}\right)^{3/2} \left(\frac{3}{2}\right)^{3/2}.
\]

(16)

and the state

\[
|3N/2 - 1, 3N/2 - 1\rangle
\]

(17)

\[
= \frac{1}{\sqrt{N}} \sum_{k=1}^{N} \prod_{i=1}^{N} \left(\frac{1}{2}\right)^{3/2} \left(\frac{3}{2}\right)^{3/2} \left(\frac{1}{2}\right)^{3/2} \left(\frac{3}{2}\right)^{3/2},
\]

(13)

corresponding to one-particle excitation reducing magnetization. It yields the result

\[
B_{sat} = \frac{5}{2} J_{AF} + \frac{7}{18} J_1.
\]

(18)

The same value may be obtained via the linear spin-wave theory based on the fully polarized state (see appendix A).

Given the experimental estimations for the 2D BIPNNBNO system, \(B_1 \approx 31 \text{ K}, B_2 \approx 31 \text{ K} \approx 35 \text{ K}, \) and \(B_{sat} = 39 \text{ K},\) we obtain from equations (10) and (18) the values of the intrachain exchange couplings, \(J_{AF} \approx 21 \text{ K} \) and \(J_1 \approx 3.5 \text{ K}.\) By substituting them into equations (15) we get the critical fields of the 2/3 plateau, \(B_2 \approx 32.8 \text{ K} (24.4 \text{ T}) \) and \(B_3 \approx 34.4 \text{ K} (25.6 \text{ T}).\) A qualitative behavior of the ferrimagnetic chain magnetization curve built from these reference points is depicted in figure 4. We emphasize especially that the
emergence of the intermediate 2/3 plateau is not related to interchain frustration effects.

3. Magnetization: exact diagonalization

In order to understand the role of the interchain couplings, the magnetization process of the BIPNNBNO ferrimagnet was examined by a variant of a numerical diagonalization method with conservation of the total cluster spin [20, 21].

The model Hamiltonian is given by

\[ \hat{H}_{\text{clust}} = J_{\text{AF}} \sum_{ij} \vec{S}_i \cdot \vec{s}_j + J_1 \sum_{ij} \vec{S}_i \cdot \vec{s}_j + J_{\text{nn}} \sum_{ij} \vec{s}_i \cdot \vec{s}_j + J_{\text{nnn}} \sum_{ij} \vec{S}_i \cdot \vec{S}_j , \]

where \( \vec{S}_i \) (\( \vec{s}_i \)) denotes the spin-1 (spin-1/2) operator at site \( i \). The sublattices and the network of the antiferromagnetic interactions, \( J_{\text{AF}}, J_1, J_{\text{nn}} \) and \( J_{\text{nnn}} \), are shown in figure 5. We perform the calculation of the \( N \)-step magnetization curve for the \( N = 4 \times 8 = 32 \) cluster depicted in the same figure. The intrachain parameters, \( J_{\text{AF}} \) and \( J_1 \), have been estimated in section 2, whereas the interchain ones, \( J_{\text{nn}} \) and \( J_{\text{nnn}} \), are assumed to be less than \( J_1 \). Open boundary conditions are used for the numerical calculations.

The importance of the frustrating coupling is seen from comparison of the two magnetization curves displayed in figures 6 and 7. They correspond to the no frustration case and a pronounced frustrating coupling, respectively. The magnetization curves exhibit several interesting features. For instance, the magnetization behavior in higher magnetic fields (\( B > B_1 \)) is reproduced well within the model of non-interacting ferrimagnetic chains. Another remarkable

![Figure 4](image-url)  
Figure 4. Qualitative magnetization curve of the ferrimagnetic (1, 1/2) chain.

![Figure 5](image-url)  
Figure 5. Cluster of \( N = 4 \times 8 = 32 \) sites used in the exact diagonalization. The fixed exchange couplings are \( J_{\text{AF}} = 21 \) K, \( J_1 = 3.5 \) K, \( J_{\text{nn}} = 0.5J_1 \), and \( J_{\text{nnn}} = 0 \) (no frustration).

![Figure 6](image-url)  
Figure 6. Magnetization curve for the 32-site cluster. The exchange couplings are taken as \( J_{\text{AF}} = 21 \) K, \( J_1 = 3.5 \) K, \( J_{\text{nn}} = 0.5J_1 \), and \( J_{\text{nnn}} = 0 \) (no frustration). The dotted line marks a calculation via the model of non-interacting (1, 1/2) chains.

The magnetization process is compared with the results of the model of non-interacting (1, 1/2) ferrimagnetic chains. A standard way to build the magnetization curve at \( T = 0 \) is to define the lowest energy \( E(N, M) \) of the Hamiltonian (2) in the subspace where \( \sum_{j=1}^{N} (S^z_j + s^z_j) = M \) for a finite system of \( N \) elementary \((S, s)\) blocks. Applying a magnetic field \( B \) leads to a Zeeman splitting of the energy levels, and therefore level crossing occurs on increasing the field. These level crossing correspond to jumps in the magnetization until the fully polarized state is reached at a certain value of the magnetic field. The magnetization of four independent chains is then derived from

\[ m = 4M/N, \]

\[ M = \max[M|E(N, M + 1) - E(N, M) > B]|, \]

which gives a step curve.
feature revealed by figures 6 and 7 is that the singlet ground state plateau emerges at non-zero frustration interaction whereas the narrow 2/3 plateau appears regardless of the frustration. We numerically found that the width $\Delta S$ of the singlet plateau scales almost linearly with the $J_{\text{nnn}}$ value (figure 8). To check into the case of the dependence we repeat calculations on clusters of smaller, $N = 4 \times 6 = 24$, and larger size, $N = 4 \times 10 = 40$, with the same set of parameters, both of which support the finding. The observation indicates that the zero magnetization plateau has a quantum origin with a crucial role of frustration which destroys long-range order and drives the system into the singlet phase. Below, we address analytically the issue in the regimes of strong and weak interchain couplings.

4. Formation of the singlet plateau

In low-dimensional Heisenberg systems, frustrating couplings can drive transitions to gap-full quantum states, where local singlets form a ground state. These quantum gapped phases may have long-range singlet order (valence-bond state), or realize a resonating valence band spin liquid. In the latter case, the ground state is a coherent superposition of all lattice coverings by local singlets [22].

To recognize features of these phases in the ED results we undertake analytical treatments of the four-legs spin tube shown in figure 9. The new system is infinite along the $b$-axis, and periodic with the 4-site period along the $a$-axis. The tube forms a minimal setup including the interchain nearest- and next-nearest-neighbor couplings and contains the same number of ferrimagnetic chains parallel to the $b$-axis as the clusters in the ED study. As we demonstrate below, the simplified model elucidates the important role of frustration in stabilization of the singlet phase.

The singlet phase may arise in the limit of strong ring coupling $J_{\text{nn}}$, $J_{\text{nnn}} \gg J_1$. In this case, the problem can be analyzed in terms of a Heisenberg XXZ model similar to ladders in a magnetic field [23]. The opposite limit ($J_1 \gg J_{\text{nn}}, J_{\text{nnn}}$) results in a scenario of weakly interacting chains. Based on a block renormalization procedure, the original system is then mapped onto the model of a spin tube with four ferromagnetic spin-1/2 legs. We mention that the ground state properties of two-leg spin ladders with ferromagnetic intrachain coupling and antiferromagnetic interchain couplings have been discussed in [24, 25] in the absence of an external field. A magnetization process of those spin ladders with an even number of legs (2 and 4) has been studied in [26] in the regime of weak ferromagnetic coupling along the legs and strong antiferromagnetic coupling along the rungs.

The appearance of the singlet phase in the frustrated spin tube with four weakly coupled ferromagnetic spin-1/2 legs can be studied through the bosonization technique, which proves its effectiveness for quasi-one-dimensional spin-one-half systems. To the best of our knowledge, the system has never been previously reported, however our further analysis follows closely to that of given in [27], where the 4-legs spin tube with antiferromagnetic chains and a specific form of diagonal rung interactions (but with no frustration) has been treated. Note as well that spin ladders with ferromagnetic and ferrimagnetic legs are much less studied [28, 29] by the bosonization approach in comparison with ferromagnetic and ferrimagnetic legs. The main problem arising here is that the formalism is well defined only if there is an easy-plane exchange anisotropy. In this regard, we note that measurements of the angular dependence of the ESR linewidth for the BIPNNBNO system showed that the largest linewidth was observed for the field direction perpendicular to the $ab$ plane [30]. Due to the theoretical consideration by Oshikawa and Affleck [31] a critical regime of XY anisotropy is expected in the compound. In addition we point out that the ED algorithm invoked in the previous section enables one to treat clusters of sufficiently large sizes due to the use of the rotational $SU(2)$ symmetry. The latter is broken by the anisotropy, whose role in a singlet gap formation is a subject of future ED studies.

![Figure 7](image1.png)

**Figure 7.** Magnetization curve for the 32-site cluster. The exchange couplings are taken as $J_{ AF} = 21$ K, $J_1 = 3.5$ K, $J_{\text{int}} = 0.5J_1$, $J_{\text{nnn}} = 0.075J_1$. The dotted line marks a calculation via the model of non-interacting (1, 1/2) chains.

![Figure 8](image2.png)

**Figure 8.** Value of the singlet plateau $\Delta S$ as a function of the frustrating coupling $J_{\text{nnn}}$ obtained on a cluster of sizes $N = 4 \times 6 = 24$ (white circles), $N = 4 \times 8 = 32$ (black squares) and $N = 4 \times 10 = 40$ (white triangles).
To summarize, two different mechanisms of formation of the singlet plateau may be likely candidates: a valence-bond (dimerized) type ground state in the strong-coupling limit, and a generalization of Haldane’s conjecture to the weakly coupled ferrimagnetic chains. Below, we demonstrate that only the last approach confirms the important role of frustration in stabilization of the singlet phase and agrees with the ED calculations.

4.1. Spin tube: weakly interacting rings and a model of a single XXZ chain

We study the Hamiltonian of the spin tube (see figure 9)

$$\mathcal{H} = \sum_{n=1}^{N} H^{\text{ring}}_n + J_1 \sum_{n=1}^{N} (S_{n,1} S_{n+1,1} + S_{n,2} S_{n+1,2})$$

$$+ S_{n,3} S_{n+1,3} + S_{n,4} S_{n+1,4}) - B \sum_{n=1}^{N} \sum_{i=1}^{4} (S^z_{n,i} + S^z_{n,i+1}),$$

(21)

where the Hamiltonian of the separate ring is

$$H^{\text{ring}}_n = J_{nn} (S_{n,1} S_{n,2} + S_{n,2} S_{n,3} + S_{n,3} S_{n,4} + S_{n,4} S_{n,1})$$

$$+ J_{nnn} (S_{n,1} S_{n,3} + S_{n,2} S_{n,4}).$$

(22)

Here, $S = 1$ and $s = 1/2$, $n$ is the index of the ring, $N$ is the total number of rings, and the index $i$ marks the $(1, 1/2)$ blocks inside the rings. Periodic boundary conditions along the tube direction are imposed. In our model it is suggested that $J_{nn}, J_{nnn} \gg J_1$. Note that to construct an effective XXZ spin chain Hamiltonian only low-lying spin-1/2 states of the separate $(1, 1/2)$ block are retained whereas higher energy spin-3/2 states are dropped. This accounts for an absence of $J_{AF}$ in the ring Hamiltonian (22).

In the limit $J_1 = 0$ the system decouples into a collection of noninteracting rings. At zero magnetic field, the singlet and triplet states

$$|\psi_0\rangle = -\frac{\sqrt{3}}{2} \langle 00; 00 \rangle + \frac{1}{2} \langle 11; 00 \rangle,$$

$$|\psi_1\rangle = \frac{1}{\sqrt{2}} \langle 01; 11 \rangle + \frac{1}{\sqrt{2}} \langle 10; 11 \rangle$$

(23)

have the lowest energies $E_0 = -2J_{nn}/9 + 8J_{nnn}/9$ and $E_1 = -J_{nn}/9 + 8J_{nnn}/9$, respectively. The states of the ring $|S_{12} S_{34}; SM\rangle$ are obtained via the common rule of addition of moments, where $S_{12}$ ($S_{34}$) is the spin of a dimer composed of the spins of the 1 and 2 (3 and 4) blocks. The singlet and triplet states of the ring that enter into (23) are given in appendix B.

Upon increasing the magnetic field a transition between the singlet and triplet states occurs at $B = J_{nn}/9$ and the total magnetization jumps abruptly from zero to $M = N$.

At non-zero ring coupling the sharp transition is broadened and starts from a critical value $B_0$. To find the field we derive the XXZ spin chain Hamiltonian by using the standard approach, which is analogous to the study of a spin-1/2 ladder with strong rung exchange [23].

The Hamiltonian (21) is split into two parts

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1.$$

$$\mathcal{H}_0 = \sum_{n=1}^{N} H^{\text{ring}}_n - B_c \sum_{i=1}^{N} \sum_{n=1}^{4} \left( S^z_i + S^z_{i+1} \right),$$

$$\mathcal{H}_1 = J_1 \sum_{n=1}^{N} \left[ S_{n,1} S_{n+1,1} + S_{n,2} S_{n+1,2} + S_{n,3} S_{n+1,3} + S_{n,4} S_{n+1,4} \right] - (B - B_c) \sum_{i=1}^{N} \sum_{n=1}^{4} \left( S^z_i + S^z_{i+1} \right).$$
where $B_c = E_1 - E_0$. The $\mathcal{H}_1$ lifts the $2^N$-fold degeneracy of the ground state of the Hamiltonian $\mathcal{H}_0$. The latter can be in the state $|\psi_0\rangle$ or $|\psi_1\rangle$. By using standard many-body perturbation theory [32] the effective Hamiltonian can be derived as

$$\mathcal{H}_{\text{eff}} = J_{xy}^{\text{eff}} \sum_{n=1}^{N} \left( \tilde{S}_n^x \tilde{S}_{n+1}^x + \tilde{S}_n^y \tilde{S}_{n+1}^y \right) + J_{z}^{\text{eff}} \sum_{n=1}^{N} \tilde{S}_n^z,$$

(24)

where $J_{xy}^{\text{eff}} = -16J_1/27$, $J_{z}^{\text{eff}} = -J_1/9$ and $B_{\text{eff}} = J_1/9 + B - B_c$.

To get the expression the pseudo-spin $\tilde{S}_i$ is 1/2 operators in the restricted space are related by

$$\tilde{S}_n^+ |\psi_0\rangle_n = \frac{1}{2} |\psi_0\rangle_n, \quad \tilde{S}_n^+ |\psi_1\rangle_n = \frac{1}{2} |\psi_1\rangle_n,$$

$$\tilde{S}_n^- |\psi_0\rangle_n = |\psi_1\rangle_n, \quad \tilde{S}_n^- |\psi_1\rangle_n = 0.$$  

The starting spin-1 operators and the pseudo-spin operators in the restricted space are related by

$$S_{n}^z = \frac{1}{6} + \frac{1}{3} \tilde{S}_n^z, \quad S_{n}^+ = (1-i)^{-1} \frac{4}{3\sqrt{3}} \tilde{S}_n^+, \quad S_{n}^- = (1-i)^{-1} \frac{4}{3\sqrt{3}} \tilde{S}_n^-.$$  

(26)

The corresponding map for the spin-1/2 operators is

$$S_{n}^z = \frac{1}{2} + \frac{1}{2} \tilde{S}_n^z, \quad S_{n}^+ = \frac{1}{4} \tilde{S}_n^+, \quad S_{n}^- = \frac{1}{4} \tilde{S}_n^-.$$  

(27)

The Jordan–Wigner transformation maps the Hamiltonian [24] onto a system of interacting spinless fermions

$$\mathcal{H}_{\text{df}} = t \sum_{n=1}^{N} \left[ c_n^+ c_{n+1} + c_{n+1}^+ c_n + V \sum_{\lambda=1}^{N} n_{\lambda, n+1} - \mu \sum_{n=1}^{N} n_n \right],$$

(28)

where $t = J_{xy}^{\text{eff}}/2$, $V = J_{z}^{\text{eff}}$ and $\mu = J_{z}^{\text{eff}} + B_{\text{eff}}$.

The lowest critical field $B_0$ corresponds to that value of the chemical potential $\mu$ for which the band of spinless fermions starts to fill up. This yields the condition $\mu = -2t$ and leads to the result

$$B_0 = \frac{1}{9} J_{xy} + \frac{16}{27} J_1.$$  

(29)

The critical value involves no frustration parameter $J_{\text{ann}}$ that is clearly contrary to the ED results.

A similar analysis can be carried out for the saturation field. The details of the calculations are relegated to appendix C.

### 4.2. Spin tube: a model of weakly interacting ferromagnetic legs and Abelian bosonization

To apply the bosonization we should map the initial system, consisting of two sorts of spins, spin-1/2 and spin-1, to the spin-1/2 system by using the quantum renormalization group (QRG) in real space based on the block renormalization procedure [33]. To exploit the real-space QRG technique, one divides the spin lattice into small blocks, namely, the intrachain dimers (1, 1/2), and obtains the lowest energy states $|\alpha\rangle$ of each isolated block. The effect of inter-block interactions is then taken into account by constructing an effective Hamiltonian $\mathcal{H}_{\text{eff}}$, which now acts on a smaller Hilbert space embedded in the original one. In this new Hilbert space each of the former blocks is treated as a single site. The effective Hamiltonians $\mathcal{H}_{\text{eff}} = Q\mathcal{H}Q^\dagger$ is constructed via the projection operator $Q = \prod_{n=1}^{N} \mathcal{Q}_n$, with $Q_j = |\alpha\rangle \langle \alpha|$, where $m$ is the number of low-energy states that are kept and $N$ is a number of lattice cells.

We hold the lowest doublet $S = 1/2$ to find an effective low-energy Hamiltonian. The higher energy $S = 3/2$ states are neglected. One can check that the reduced matrix elements are ($S_1 = 1$, $s_2 = 1/2$)

$$\langle 11/2; 1/2|S_1|11/2; 1/2\rangle = 2\sqrt{\frac{5}{3}},$$

$$\langle 11/2; 1/2|s_2|1 1/2; 1/2\rangle = -\frac{1}{\sqrt{6}}.$$  

Therefore the effective spin-1/2 operators of the renormalized chain are

$$Q_1^\dagger S_1 Q_1 = \frac{4}{3} \tilde{S}_i, \quad Q_1^\dagger s_2 Q_1 = -\frac{1}{3} \tilde{S}_i \quad (S = 1/2).$$  

(30)

The renormalized Hamiltonian of the intrachain interactions corresponds to the ferromagnetic Heisenberg spin-1/2 model with the exchange coupling $J = -4J_1/9$ (figure 9). The interchain interactions between the nearest neighbors, spins $-1/2$, and next-nearest neighbors, spins $-1$, are renormalized as $J_{\perp} = J_{\text{in}}/9$ and $J_{\parallel} = 16J_{\text{in}}/9$, respectively.

Consider a four-legs spin tube consisting of spin-1/2 chains. The Hamiltonian of the system is

$$\hat{\mathcal{H}}_{\text{tube}} = \sum_{\lambda=1}^{4} \hat{\mathcal{H}}_{\lambda} + \hat{\mathcal{H}}_{12}^{\perp} + \hat{\mathcal{H}}_{23}^{\perp} + \hat{\mathcal{H}}_{34}^{\perp} + \hat{\mathcal{H}}_{14}^{\perp} + \hat{\mathcal{H}}_{13}^{\perp} + \hat{\mathcal{H}}_{24}^{\perp}.$$  

(31)

The spins along the chains are coupled ferromagnetically, the Hamiltonian for the separate $\lambda$th chain is

$$\hat{\mathcal{H}}_{\lambda} = -J^{\lambda} \sum_{i=1}^{N} \left( S_{\lambda,j}^\dagger S_{\lambda,j+1}^\sigma + S_{\lambda,j}^\sigma S_{\lambda,j+1}^\dagger \right) \left( S_{\lambda,j}^\dagger S_{\lambda,j+1}^\sigma + S_{\lambda,j}^\sigma S_{\lambda,j+1}^\dagger \right) + J^{\lambda} \sum_{j=1}^{N} \left( S_{\lambda,j}^\dagger S_{\lambda,k,j+1} + S_{\lambda,j}^\sigma S_{\lambda,k,j}^\dagger \right) \left( S_{\lambda,j}^\dagger S_{\lambda,k,j+1} + S_{\lambda,j}^\sigma S_{\lambda,k,j}^\dagger \right) + J^{\lambda} \sum_{j=1}^{N} S_{\lambda,j}^\dagger S_{\lambda,k,j}.$$

(32)
the spin Hamiltonian of the spin tube with model to an effective model of continuum field, we convert
the Luttinger model. The bosonic expressions for spin operators legs to a Hamiltonian of spinless fermions using the
the Hamiltonian with the antiferromagnetic legs.

Following the general procedure of transforming a spin model to an effective model of continuum field, we convert
the spin Hamiltonian of the spin tube with antiferromagnetic legs to a Hamiltonian of spinless fermions using the
Jordan–Wigner transformation, then map it to a modified Luttinger model. The bosonic expressions for spin operators
are
\[ S_{\lambda, j}^+ \rightarrow \frac{S_{\lambda, j}^+}{a}, \]
\[ S_{\lambda, j}^- \rightarrow \frac{S_{\lambda, j}^-}{a}, \]
\[ S_{\lambda, j}^z \rightarrow \frac{1}{\sqrt{\pi}} \partial_x \Phi_\lambda + \frac{1}{\pi a} \sin(\sqrt{4\pi} \Phi_\lambda), \]
where \( \Phi \) and \( \Theta \) are the bosonic dual fields, and \( x \) is defined on the lattice, \( x_j = ja \), \( a \) is a short-distance cutoff.

The bosonized form of the Hamiltonian of the non-interacting chains is
\[ \mathcal{H}_0 = \frac{\pi}{2} \int \frac{dx \left[ K \Pi_\lambda^2 + \frac{1}{K} (\partial_x \Phi_\lambda)^2 \right]}{2}, \]
where \( \Pi_\lambda(x) = \partial_x \Theta_\lambda \) is the canonically conjugate momentum to \( \Phi_\lambda \). The Luttinger liquid parameters are fixed from the Bethe ansatz solution [34]
\[ K = \frac{\pi}{2(\pi - \arccos \Delta)}, \quad u = J^{xy} \frac{\pi \sqrt{1 - \Delta^2}}{2 \arccos \Delta}. \]
The velocity \( u \) vanishes and \( K \) diverges for \( \Delta = 1 \). This corresponds to the ferromagnetic instability point of a single chain.

The interchain interactions (32) between the nearest neighbor chains reads as
\[ \mathcal{H}_{\perp, \lambda'}^{\perp} = J_{\perp, \lambda, \lambda'}^{xy} \sum_{j=1}^{N} \left( S_{\lambda, j}^x S_{\lambda', j}^x + S_{\lambda, j}^y S_{\lambda', j}^y \right) \]
\[ + J_{\perp, \lambda, \lambda'}^{z} \sum_{i=1}^{N} S_{\lambda, i}^z S_{\lambda', i}^z \]
and includes the nearest, \( J_{\perp} > 0 \), and the next-nearest, \( J'_{\perp} > 0 \), antiferromagnetic interleg couplings.

The unitary transformation keeping spin commutation relations
\[ S_{\lambda, j}^\pm \rightarrow (-1)^{j} S_{\lambda, j}^\pm, \quad S_{\lambda, j}^z \rightarrow S_{\lambda, j}^z \]
maps the Hamiltonian (31) to the Hamiltonian with antiferromagnetic legs. It changes \( J^{yz} \rightarrow -J^{yz} \) and \( J^z \rightarrow J^z \), and the ferromagnetic isotropic point is \( \Delta = J^z/J^{yz} = -1 \) in the Hamiltonian with the antiferromagnetic legs.

Following the general procedure of transforming a spin model to an effective model of continuum field, we convert
the spin Hamiltonian of the spin tube with antiferromagnetic legs to a Hamiltonian of spinless fermions using the
Jordan–Wigner transformation, then map it to a modified Luttinger model. The bosonic expressions for spin operators
are
\[ S_{\lambda, j}^+ \rightarrow \frac{S_{\lambda, j}^+}{a}, \]
\[ S_{\lambda, j}^- \rightarrow \frac{S_{\lambda, j}^-}{a}, \]
\[ S_{\lambda, j}^z \rightarrow \frac{1}{\sqrt{\pi}} \partial_x \Phi_\lambda + \frac{1}{\pi a} \sin(\sqrt{4\pi} \Phi_\lambda), \]
where \( \Phi \) and \( \Theta \) are the bosonic dual fields, and \( x \) is defined on the lattice, \( x_j = ja \), \( a \) is a short-distance cutoff.

The bosonized form of the Hamiltonian of the non-interacting chains is
\[ \mathcal{H}_0 = \frac{u}{2} \int \frac{dx \left[ K \Pi_\lambda^2 + \frac{1}{K} (\partial_x \Phi_\lambda)^2 \right]}{2}, \]
with
\[ \Pi_\lambda(x) = \partial_x \Theta_\lambda \]
\[ u = J^{yz} \pi \sqrt{1 - \Delta^2} \]
\[ \Delta = \frac{1}{2 \arccos \Delta}. \]

The relevant and marginally relevant terms of the interchain couplings are given by
\[ \mathcal{H}_{\perp} = 2g_1 \sum_{i=1}^{2} \int \frac{dx}{(2\pi a)^2} \cos(\sqrt{4\pi} \Phi_\lambda) \cos(\sqrt{4\pi} \Phi_\lambda) \]
\[ + 2g'_1 \int \frac{dx}{(2\pi a)^2} \cos(\sqrt{4\pi} \Phi_\lambda) \cos(\sqrt{4\pi} \Phi_\lambda) \]
The $g_5$ terms are irrelevant and are omitted.

The Hamiltonian (39) describes four independent gapless spin-1/2 chains coupled by the interchain interaction in the form of equation (41). It is expected that the interleg coupling results in the Haldane gap in the excitation spectrum. Note that in the vicinity of the single chain ferromagnetic instability, $\Delta = -1$, the effective bandwidth collapses, $u \rightarrow 0$, and the effect of the interleg couplings becomes crucial. To find the detailed behavior of the gap in the phase diagram with antiferromagnetic ($J_\perp$, $J > 0$) interleg coupling and ferromagnetic leg regime ($\Delta < 0$) we use the renormalization group analysis.

The RG equations are derived through the standard technique (see [36], for example). The result is

$$
\begin{align*}
\frac{dg_1}{dt} &= [2 - (K_s + K_{a1})]g_1, \\
\frac{dg_1'}{dt} &= [2 - (K_s + K_{a1})]g_1', \\
\frac{dg_2}{dt} &= [2 - (K_{a1} + K_{a2})]g_2, \\
\frac{dg_2'}{dt} &= [2 - 2K_{a1}]g_2', \\
\frac{dg_3}{dt} &= \left[ 2 - \frac{1}{3} \left( \frac{1}{K_{a1}} + \frac{1}{K_{a2}} \right) \right] g_3, \\
\frac{dg_3'}{dt} &= \left[ 2 - \frac{1}{3} \frac{1}{K_{a1}} \right] g_3', \\
\frac{dg_4}{dt} &= \left[ 2 - (K_s + K_{a1} + \frac{1}{4K_{a1}} + \frac{1}{4K_{a2}}) \right] g_4.
\end{align*}
$$

(41)

One sees that the $g_1$ terms are relevant for $K_s + K_{a1} < 2$; the $g_1'$ term is relevant for $K_s + K_{a1} < 2$; the $g_2$ terms are relevant for $K_{a1} + K_{a2} < 2$; the $g_2'$ term is relevant for $K_{a1} < 1$; the $g_3$ term is relevant for $K_{a1}^{-1} + K_{a2}^{-1} < 8$; the $g_3'$ term is relevant for $K_{a1} > 1/4$. Despite the $g_4$ and $g_4'$ terms being irrelevant they are the most relevant terms which couple the symmetric and antisymmetric modes [27].

Using the RG equations the behavior of the gap in the whole phase diagram can be established. Following the standard routine, we analyze the effect of the transverse ($J_{\perp}^y$) and longitudinal ($J_{\parallel}^x$) parts of the interleg coupling separately.

4.2.1. Transverse part of the interleg interactions. In this case $J_{\perp}^y$, $J_{\perp}^y \neq 0$ and $J_{\perp}^x$, $J_{\parallel}^x = 0$, the initial values of the coupling constants are given by $g_1(l = 0) = g'_1(l = 0) = 0$, $g_2(l = 0) = g'_2(l = 0) = 0$, $g_3(l = 0) = 2\pi J_{\perp}^y$, $g'_3(l = 0) = 2\pi J_{\parallel}^x$, $g_4(l = 0) = \pi J_{\perp}^y$ and $g'_4(l = 0) = \pi J_{\parallel}^y$. The bare Luttinger parameters are $u_s = u$, $u_{a1} = u_{a2} = u_{a3} = u$, and $K_s(l = 0) = K_s$, $K_{a1}(l = 0) = K_{a1}$, $K_{a2}(l = 0) = K_{a2}$, $K_{a3}(l = 0) = K_{a3}$. The term $g_3$ is relevant for $-1 \leq \Delta \leq 0$, while the $g_4$ term is irrelevant. It is easily checked numerically that $g_3$, $g_3'$ grow whereas $g_4$, $g_4'$ decrease under the RG. It means that $\Theta_1$, $\Theta_2$, $\Theta_3$ are locked in one of the vacuum states ($\Theta_1 = \Theta_2 = 0$, $\Theta_3 = \sqrt{\tau}$ or $\Theta_1 = \Theta_2 = \sqrt{\tau}$, $\Theta_3 = 0$ provided $J_{\perp}^y > J_{\parallel}^x$), fluctuations of the fields $\Theta_{a1}$ ($i = 1-3$) are completely suppressed. Therefore arbitrary $J_{\perp}^y > J_{\parallel}^x \neq 0$ do not generate a gap in the antisymmetric modes ($\Theta_1$ are pinned, disordered).

After the fluctuations of the fields $\Theta_i$ are stopped, the infrared behavior of the symmetric mode is governed by the
term of the coupling with the antisymmetric modes

$$\hat{H}_{\text{int}} = 2\tilde{g}_4 \sum_{i=1}^{2} \int \frac{dx}{(2\pi a)^2} \cos \left( \sqrt{4\pi} \Phi_s \right) \cos \left( \sqrt{4\pi} \Phi_a \right)$$

$$+ 2\tilde{g}_4' \int \frac{dx}{(2\pi a)^2} \cos \left( \sqrt{4\pi} \Phi_s \right) \cos \left( \sqrt{4\pi} \Phi_a \right),$$

where

$$\tilde{g}_4 = g_4 \cos \left( \sqrt{\pi} (\Theta_{a_1} + \Theta_{a_3}) \right)$$

$$\tilde{g}_4' = g_4' \cos \left( \sqrt{\pi} (\Theta_{a_2} + \Theta_{a_3}) \right),$$

and $\tilde{g}_4, \tilde{g}_4'$ are renormalized couplings provided $g_4, g_4' = O(1)$. Here, the invariance of $\hat{H}_{\text{int}}$ given by equation (41) under $\Theta_a \rightarrow -\Theta_a$ yields $\cos (\sqrt{\pi} (\Theta_{a_1} - \Theta_{a_2})) = (\cos(\sqrt{\pi} (\Theta_{a_1} + \Theta_{a_3})))$. Despite $e^{i\Phi_s}$ having exponentially decaying correlations due to the $\Theta_a$ being pinned, a scrupulous analysis [27] shows that the effective Hamiltonian for $\Phi_s$ presents a standard sine–Gordon Hamiltonian

$$\hat{H}_{\text{eff}} = \frac{u_s}{2} \int \frac{dx}{(2\pi a)^2} \cos \left( \sqrt{16\pi} \Phi_s \right),$$

where $K_s$ is a renormalized value of $K$, and $g$ is a new effective coupling constant. From the correlation function $\langle \exp[i\sqrt{16\pi} \Phi_s(x)] \exp[i\sqrt{16\pi} \Phi_s(y)] \rangle = (\alpha^2/|x-y|^2)^4 K_s$, it follows that the $g$ term has a scale dimension $4K_s$. Therefore, it is relevant for $K_s < 1/2$, when $\Phi_s$ is pinned, i.e. becomes massive [37].

To summarize, the transverse part of the interleg coupling supports gapped antisymmetric modes, the symmetric sector is gapped at $K_s < 1/2$, and remains gapless at $K_s > 1/2$. The condition $K_s = 1/2$ determines a boundary between the gapless Spin Liquid X1Y1 phase [38], and a generalization of the gapped Rung Singlets phase [39] for the four-leg spin tube

(figure 10). (Hereinafter, we retain the names of phases used in the theory of spin ladders with ferromagnetic legs.) In the last case, spins on the same rungs or along the shortest diagonals form singlet pairs in a dynamical way.

4.2.2. Longitudinal part of the interleg interactions. For the case of the longitudinal part of the interleg exchange, $J_{\perp,1}^y, J_{\perp,2}^y = 0$ and $J_{\perp,1}^z, J_{\perp,2}^z \neq 0$, the bare values of the coupling constants are given by $g_1(l = 0) = -2J_{\perp,1}^z, g_2(l = 0) = -2J_{\perp,2}^z, g_3(l = 0) = g_3'(l = 0) = 0$, and $g_4(l = 0) = g_4'(l = 0) = 0$.

The strong-coupling phase diagram in the vicinity of ferromagnetic instability point ($\Delta = -1$ and $J_{\perp,1}^z, J_{\perp,2}^z = 0$) obtained in the RG analysis is shown in figure 11. In the sector denoted as spin liquid II phase [39] the $g_{1,2}, g_{3,4}'$ terms are irrelevant. The symmetric and antisymmetric modes remain gapless. In the sector marked as a Haldane phase the terms $g_{1,2}, g_{3,4}'$ are relevant. Since all of the modes are coupled and locked together, both the symmetric and antisymmetric modes are gapped.

The phase of a ferromagnet with antiphase interchain order arises as a result of the ferromagnetic instability with increasing interleg antiferromagnetic coupling. The boundary of the transition into the phase is obtained by studying the velocity renormalization of the corresponding gapless excitations. We mark the transition at $u_{\omega} = 0$ ($i = 1–3$).

4.2.3. Isotropic interleg exchange. The initial values of the coupling constants are $g_1(l = 0) = -2J_{\perp,1}^z, g_2(l = 0) = -2J_{\perp,2}^z, g_3(l = 0) = g_3'(l = 0) = 2J_{\perp,1}^z, g_3(l = 0) = g_3'(l = 0) = 0, g_4(l = 0) = g_4'(l = 0) = 0$.

From the RG equations (42) it is seen that the most relevant operators are the $g_3, g_3'$ terms. Therefore, the antisymmetric sector is gapped, and $\Theta_a$ are locked in the disordered phase. As in the case of the transversal interleg interactions, an effective sine–Gordon Hamiltonian for the symmetric mode determines the phase boundary $K_s = 1/2$ between gapped and gapless phases. Numerical analysis shows that the ground state phase diagram consists of

![Figure 10.](image1.png) Figure 10. The ground state phase diagram in the vicinity $\Delta = -1$ of the four-leg tube with transverse coupling between legs.

![Figure 11.](image2.png) Figure 11. The ground state phase diagram in the vicinity $\Delta = -1$ of the four-leg tube with longitudinal coupling between legs.
We derive the critical field destroying the singlet plateau and it turns out that it does not depend on the frustration parameter $J_{nn}$. Another analytical strategy is realized via the Abelian bosonization formalism, which is relevant for the opposite limit $J_1 \gg J_{nn} \geq J_{nn}$. We demonstrate that the gap-full disordered Rung Singlets phase comes up when the XY exchange anisotropy may tilt the balance from the long-range order with an antiphase interchain arrangement of ferrimagnetic chains towards the spin liquid phase. A role of the anisotropy in a formation of the spin gap in the original two-dimensional system deserves further study.

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We wish to thank Professor N V Baranov for discussions.

Appendix A. The spin-wave calculation of the saturation field

We start from the fully polarized state with $M = 3/2N$, and define the bosonic operators for the spin deviation in each sublattice as

$$
S^+_{1i} = \sqrt{2S_1} a_i, \quad S^-_{1i} = \sqrt{2S_1} a_i^\dagger, \\
S^c_{1i} = S_1 - a_i a_i^\dagger, \quad S^z_{1i} = S_2 b_i, \quad S^z_{2i} = S_2 - b_i^\dagger b_i.
$$

(A.1)

By using the momentum representation of the bosonic operators

$$
a_i = \frac{1}{\sqrt{N}} \sum_k e^{ikR_i} a_k, \quad b_i = \frac{1}{\sqrt{N}} \sum_k e^{ikR_i} b_k,
$$

(A.2)

the initial Hamiltonian (2) with the added Zeeman interaction, $-B \sum_{i=1}^{N} e_i^z + s_2^z$, is converted into the quadratic form

$$
\mathcal{H} = \sum_k \left[ -s_2 (J_{AF} + J_1) + B |a_k|^2 a_k \\
+ |s_1 (J_{AF} + J_1) + B |b_k|^2 b_k \\
+ \sum_k \sqrt{S_1 S_2} [J_{AF} + J_1 e^{-ik}] a_k^\dagger b_k \\
+ \sum_k \sqrt{S_1 S_2} [J_{AF} + J_1 e^{ik}] b_k^\dagger a_k \right],
$$

(A.3)

where only the operator terms hold.

It results in the two-branched exact ferrimagnetic one-particle excitation [40]

$$
\omega_\pm(k) = B - \frac{1}{2} (S_1 + s_2) (J_{AF} + J_1) \\
\pm \frac{1}{2} \sqrt{(J_{AF} + J_1)^2 (S_1 + s_2)^2 - 8S_1 S_2 J_{AF} J_1 (1 - \cos k)}.
$$

(A.4)

Both excitation branches reduce magnetization. Below the critical field

$$
B_c = (S_1 + s_2) (J_{AF} + J_1) = \frac{1}{2} J_{AF} + \frac{1}{2} J_1
$$

(A.5)

one dispersion branch acquires negative energy, such that a breakdown of the fully polarized state occurs. The value $B_c$ coincides with the result (18).

5. Conclusion

We have studied the magnetization process in the two-dimensional compound BIPNNBNO, which exhibits ferromagnetism of non-Lieb–Mattis type. The investigation is complicated by a lack of reliable information about exchange interactions in the system. For a start, we proposed the naive model of non-interacting ferrimagnetic chains and showed that the appearance of both 1/3 and 2/3 plateaus can be explained within the model. This provides us with the intrachain exchange couplings $J_{AF}$ and $J_1$. By setting these parameters in the exact diagonalization routine the magnetization curve for the 32 and 40-sites clusters is numerically calculated. We demonstrate that a magnetization curve similar to that observed in the experiment is obtained in the regime of weak interchain coupling, $J_1 \gg J_{nn} > J_{nn}$. Another revealed phenomenon is that the width of the singlet plateau increases with the growth of the antiferromagnetic frustrating coupling between the next-to-nearest chains. Following these results, we apply on the tube lattice two low-energy theories which could explain an appearance of the singlet phase. The first one is based on the effective XXZ Heisenberg model in a longitudinal magnetic field in the limit where the interchain coupling dominates, $J_{nn} \gg J_{nn} \gg J_1$, the disordered Rung Singlet gap-full phase and the stripe ferromagnetic phase with dominating intraleg ferromagnetic ordering. The sector of the rung singlet phase increases at $J_1 \geq J_{nn} \gg J_{nn}$, which corresponds to the region of the disordered Rung Singlet phase close the point of the ferromagnetic instability, $\Delta = -1$. The ground state phase diagram in the vicinity $\Delta = -1$ of the four-leg tube with isotropic coupling between legs. The phase boundary between the disordered rung singlet gap-full phase and the stripe ferromagnetic phase is shown by the dotted and solid lines for $J_1/J_\perp = 0.2$ and $J_1/J_\perp = 1.0$, respectively.

$\Delta (J_1/J_\perp)$

Ferromagnet (antiphase interchain ordering)

Disordered Rung Singlets

Figure 12. The ground state phase diagram in the vicinity $\Delta = -1$ of the four-leg tube with isotropic coupling between legs. The phase boundary between the disordered rung singlet gap-full phase and the stripe ferromagnetic phase is shown by the dotted and solid lines for $J_1/J_\perp = 0.2$ and $J_1/J_\perp = 1.0$, respectively.
Appendix B. Wavefunctions of the Ring

The triplet states read as

\[ |\psi\rangle = -\frac{1}{\sqrt{3}} |\frac{1}{2} + \frac{1}{2} + \frac{1}{2} \rangle, \]

\[ \mp \frac{1}{\sqrt{3}} |10\rangle = \frac{1}{2} |\frac{1}{2} - \frac{1}{2} + \frac{1}{2} \rangle. \]

The basic functions of the singlet states are given by

\[ |00; 00\rangle = \frac{1}{2} |\frac{1}{2} + \frac{1}{2} - \frac{1}{2} \rangle - \frac{1}{2} |\frac{1}{2} + \frac{1}{2} + \frac{1}{2} \rangle, \]

\[ |11; 00\rangle = \frac{1}{\sqrt{3}} |\frac{1}{2} + \frac{1}{2} - \frac{1}{2} \rangle + \frac{1}{2} |\frac{1}{2} + \frac{1}{2} + \frac{1}{2} \rangle. \]

The triplet states read as

\[ |01; 11\rangle = \frac{1}{\sqrt{3}} |\frac{1}{2} + \frac{1}{2} + \frac{1}{2} \rangle \]

\[ |10; 11\rangle = \frac{1}{\sqrt{3}} |\frac{1}{2} - \frac{1}{2} - \frac{1}{2} \rangle. \]

Appendix C. Saturation field in the limit of the strong ring coupling

To get the saturation field the functions of the ring with the total spins \( S = 5 \) and 6

\[ |\psi_6\rangle = \frac{1}{2} |\frac{3}{2} + \frac{3}{2} + \frac{3}{2} \rangle, \]

\[ |\psi_5\rangle = -\frac{1}{2} |\frac{3}{2} + \frac{3}{2} + \frac{3}{2} \rangle. \]

with the energies \( E_5 = J_{XX}/2 - J_{nn}/3 + J_{mm}/2, E_6 = 2J_{AF} + J_{nn} + 2J_{mnn} \) are needed.

By introducing the pseudo-spin operators in the restricted space similar to equation (25), the original spin operators are presented as follows (\( S = 1, s = 1/2 \))

\[ S_n^+ = \frac{\sqrt{3}}{2} \mu_n, \quad S_n^- = \frac{\sqrt{3}}{2} \mu_n, \]

\[ S_n^{+\dagger} = (-1)^{1+s} \sqrt{\frac{2}{3}} S_n^+, \quad S_n^{-\dagger} = (-1)^{1+s} \sqrt{\frac{2}{3}} S_n^-, \quad (C.1) \]

The effective XXZ spin chain Hamiltonian has the form \( (24) \), with the parameters \( J_{eff}^y = -J_1/3, J_{eff}^z = J_1/18 \) and \( B_{eff}^z = -7J_1/9 + B - B_c \), where \( B_c = E_6 - E_5 \).

By performing a Jordan–Wigner transformation followed by a particle–hole transformation [23] the new Hamiltonian of spinless holes is

\[ H_{th} = -\mu \sum_{n=1}^{N} (d_n^+ d_{n+1} + d_{n+1}^+ d_n) + V \sum_{n=1}^{N} n_n^+ n_n^+ - \mu_h \sum_{n=1}^{N} n_n^+. \]

where \( t = J_{eff}^x/2, V = J_{eff}^z \) and \( \mu_h = J_{eff}^z - B_{eff}^z \).

The saturation field \( B_{sat} \) corresponds to the chemical potential where the hole band starts to fill up, \( \mu_h = -2r \). This yields \( B_{sat} = B_c + J_1/6 = 3J_{AF}/2 + 4J_{nn}/3 + 3J_{mnn}/2 + J_1/6 \).

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