

## Mixed-spin systems: coexistence of Haldane gap and antiferromagnetic long range order

Indrani Bose and Emily Chattopadhyay

Department of Physics

Bose Institute

93/1, Acharya Prafulla Chandra Road

Calcutta - 700009, India

### Abstract

Recent experiments on the quasi-1D antiferromagnets  $R_2BaNiO_5$  (  $R =$  rare earth ) have shown the existence of purely 1D Haldane gap excitations propagating on the Ni chains. Below an ordering temperature, the gap excitations survive and coexist with the conventional spin waves in the ordered phase. We construct a model mixed-spin system in 2D for which the ground state can be exactly specified. Using the Matrix Product Method, we show the existence of Haldane gap excitations in the ordered phase. We consider different cases of ordering to study the effect of ordering on the degeneracy of the Haldane gap excitations.

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# I. Introduction

Haldane<sup>1</sup> in 1983 predicted that a Heisenberg antiferromagnetic (AFM) chain of integer spins has a gap in the excitation spectrum. Later, Haldane's conjecture was verified in several theoretical as well as experimental studies.<sup>2-4</sup> In a quasi-1D magnetic system, the integer spin chains are coupled via weak exchange interactions. If the strength of the coupling is very small, there is no possibility of ordering and individual spin chains remain in the disordered Haldane gap phase at all temperatures. In materials like  $CsNiCl_3$ , the inter-chain coupling is sufficiently strong to give rise to an AFM ordered phase below the Néel temperature  $T_N$ .<sup>5-6</sup> For temperature  $T > T_N$ , the three Haldane gap modes can be observed in experiments but as  $T$  approaches  $T_N$ , the gap vanishes at the 3D magnetic zone centre. In the ordered phase, two of the three modes become conventional gapless spin waves, while the third 'longitudinal' mode develops a gap.

For quasi-1D Haldane gap systems, there now exists a third possibility. Recent inelastic neutron-scattering experiments<sup>7-8</sup> on the mixed-spin antiferromagnets  $R_2BaNiO_5$  ( $R = Pr, Nd$  or  $Nd_xY_{1-x}$ ), provide clear evidence that the Haldane gap excitations propagating on the Ni chains survive even in the ordered phase. Unlike in systems like  $CsNiCl_3$ , the Haldane gap does not become zero as  $T$  approaches  $T_N$ . In the ordered phase, the gapped Haldane modes coexist with the conventional spin waves characteristic of the ordered phase. The Haldane gap excitations maintain their 1D character in the entire phase diagram. In this paper, we construct a model mixed spin system in 2D for which the ground state can be exactly specified. AFM long-range order exists in the ground state with gapless spin waves as excitations. The system also consists of integer spin chains, the ground states of which are the Valence Bond Solid (VBS) states. Haldane gap excitations can be created in the chains in the ordered phase of the mixed-spin system. We also discuss how the gap changes as the nature of the ordering changes.

# II. Ground state and excitations

Our model mixed spin system has an underlying lattice structure shown in Fig. 1. It basically consists of a square lattice at each site of which a spin-1/2 (A-spin, solid circle) is sitting. The centre of each square plaquette is

occupied by a S=1 spin (B-spin, solid square). The B-spins along a chain in the horizontal direction interact via nearest-neighbour (n.n.) interactions but the individual B-spin chains do not interact with each other. The B-spins interact with the n.n. A-spins along the diagonal lines in Fig. 1. The A-spins interact with n.n. A-spins along the horizontal and vertical bonds of the square lattice. In  $R_2BaNiO_5$  also, the S=1 Ni chains are decoupled and the  $R_3^+$  sites with half-odd integer spins are positioned between the chains. The  $R_3^+$  spins form a network via Ni-O-R and R-O-R superexchange routes.

We now demand that the B-spin chains are in Valence Bond Solid (VBS) ground states. Affleck, Kenedy, Lieb and Tasaki (AKLT)<sup>4</sup> constructed a Hamiltonian for which the VBS state is the exact ground state of an integer spin chain. We follow the same principle to construct the Hamiltonian of our model. Fig. 2 shows two successive rows of A and B-spins. Consider the B-spins to be in a VBS state. Each spin-1 can be considered to be a symmetric combination of two spin-1/2's. In the VBS state, each spin-1/2 forms a singlet with a spin-1/2 at a neighbouring site. This process has to be symmetrized so that a spin-1 is restored at each site. In the ground state, a valence bond (VB) or singlet covers every link along the integer-spin chain so that the total spin of the state is zero. The Hamiltonian for which the VBS state is the exact ground state is given by

$$H_{AB}^i = \sum_{\Delta} P_{5/2}(\vec{S}_a + \vec{S}_b + \vec{S}_c) \quad (1)$$

$P_{5/2}$  is the projection operator onto spin 5/2 for the triangle of spins. The sum is over successive triangles of spins with bases along the integer spin chain. The spins at sites  $B_1$  and  $B_2$  have magnitude 1 and the A-spin has magnitude 1/2. In the VBS state, the sum of the three spins can never be 5/2 as a VB exists between the basal spins. Thus, the projection operator  $P_{5/2}$  operating on the triangle of spins in the VBS state gives zero. The VBS state is the exact ground state of  $H_{AB}^i$  with eigenvalue zero. It can be easily verified that the A-spins are free when the Hamiltonian in (1) operates on the VBS state of the integer spin chain. The Hamiltonian  $H_{AB}^i$ , on expanding, is given by

$$H_{AB}^i = \frac{1}{20} [2 + 5(\vec{S}_a \cdot \vec{S}_b + \vec{S}_b \cdot \vec{S}_c + \vec{S}_c \cdot \vec{S}_a) + 2(\vec{S}_a \cdot \vec{S}_b + \vec{S}_b \cdot \vec{S}_c + \vec{S}_c \cdot \vec{S}_a)^2] \quad (2)$$

When we consider the full 2D spin system, the Hamiltonian is given by

$$H_{AB} = \sum_i H_{AB}^i$$

The sum is over individual B-spin chains. We now consider the interactions between the A-spins. The Hamiltonian  $H_A$  is given by the usual Heisenberg AFM Hamiltonian defined on the square lattice:

$$H_A = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j \quad (3)$$

The spins have magnitude 1/2 and  $\langle ij \rangle$  are n.n.s. The ground state of (3), though not exactly known, has long range Néel type of order. The total Hamiltonian H is

$$H = H_A + H_{AB} \quad (4)$$

The ground state of H has the following structure: the B-spin chains are in VBS states. The square network of A-spins is in the state which is the ground state of the S=1/2, n.n. Heisenberg AFM Hamiltonian on a square lattice. The decoupling of the S=1 spin and the S=1/2 spin ground states is possible because the A-spins are free when  $H_{AB}$  operates on the VBS states.

We now consider the excitations in the system. Spin wave excitations created in the A network of spins have no effect on the VBS ground states of the chains of B-spins. The spin wave excitations are gapless. Consider now the case when the network of A-spins is in the ground state and an excitation is created in a B-spin chain. Again, for convenience, we consider the two-chain strip of Fig. 2. The A-spins in the top chain are in the Néel state  $|\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow \dots\rangle$  indicative of long range AFM order. We determine the excitation spectrum by the method of Matrix Products<sup>9-11</sup> The VBS ground state of the chain of B-spins can be written as

$$|\psi_G\rangle = Tr(g_1 \otimes g_2 \otimes \dots \otimes g_L) \quad (5)$$

where L is the number of sites (spins) in the chain. The  $2 \times 2$  matrix  $g_i$  is given by

$$g_i = \begin{pmatrix} -|0\rangle & -\sqrt{2}|1\rangle \\ \sqrt{2}|\bar{1}\rangle & |0\rangle \end{pmatrix} \quad (6)$$

The states  $|1\rangle, |0\rangle, |\bar{1}\rangle$  correspond to  $S^z = 1, 0$  and  $-1$  respectively. One can easily verify that  $H_{AB} |\psi_G\rangle = 0$ . An excited state of the chain of B-spins is constructed as [3]

$$|\psi_B^a(k)\rangle = \frac{1}{\sqrt{L}} \sum_{j=1}^L e^{-ikj} |\psi_{Bj}^a\rangle \quad (7)$$

$|\psi_{Bj}^a\rangle$  is obtained by replacing  $g_j$  in the matrix product of the VBS state by the matrix  $g_j^a$ , where  $a = 1, 0, -1$  are the values of the  $z$ -component of the total spin,  $S^z$ , of the excited state. The matrices  $g_j^a$  have the form

$$g^1 = \begin{pmatrix} \sqrt{2} |1\rangle & 0 \\ -|0\rangle & 0 \end{pmatrix}, g^0 = \begin{pmatrix} -|0\rangle & \sqrt{2} |1\rangle \\ \sqrt{2} |\bar{1}\rangle & -|0\rangle \end{pmatrix}, g^{-1} = \begin{pmatrix} 0 & -|0\rangle \\ 0 & \sqrt{2} |\bar{1}\rangle \end{pmatrix} \quad (8)$$

$$|\psi_{Bj}^a\rangle = Tr g_1 \otimes g_2 \otimes \cdots \otimes g_{j-1} \otimes g_j^a \otimes g_{j+1} \otimes \cdots \otimes g_L \quad (9)$$

$$\langle \psi_B^a(k) | \psi_B^a(k) \rangle = \frac{1}{L} \sum_{l=1}^L \sum_{p=1}^L e^{ik(l-p)} \langle \psi_{Bl}^a | \psi_{Bp}^a \rangle \quad (10)$$

We now consider the case  $a = 1$ .

$$\begin{aligned} \langle \psi_{Bl}^1 | \psi_{Bp}^1 \rangle &= \sum_{\{n_\alpha, m_\alpha\}} g_{n_1 n_2}^\dagger g_{n_2 n_3}^\dagger \cdots g_{n_l n_{l+1}}^{1\dagger} g_{n_{l+1} n_{l+2}}^\dagger \cdots g_{n_L n_1}^\dagger \\ &\quad g_{m_1 m_2} g_{m_2 m_3} \cdots g_{m_p m_{p+1}}^1 \cdots g_{m_L m_1} \end{aligned} \quad (11)$$

Define three  $4 \times 4$  matrices  $G$ ,  $G'$  and  $G''$  as

$$\begin{aligned} G_{\mu_1 \mu_2} &= G_{(n_1 m_1)(n_2 m_2)} = g_{n_1 n_2}^\dagger g_{m_1 m_2} \\ G'_{\mu_1 \mu_2} &= G'_{(n_l m_l)(n_{l+1} m_{l+1})} = g_{n_l n_{l+1}}^{1\dagger} g_{m_l m_{l+1}} \\ G''_{\mu_1 \mu_2} &= G''_{(n_p m_p)(n_{p+1} m_{p+1})} = g_{n_p n_{p+1}}^\dagger g_{m_p m_{p+1}}^1 \end{aligned} \quad (12)$$

The ordering of multi-indices is given by

$$\mu = 1, 2, 3, 4 \longleftrightarrow (11), (12), (21), (22) \quad (13)$$

$$\begin{aligned} \langle \psi_{Bl}^1(k) | \psi_{Bp}^1(k) \rangle &= \sum_{\{n_\alpha, m_\alpha\}} G_{(n_1 m_1)(n_2 m_2)} G_{(n_2 m_2)(n_3 m_3)} \cdots \\ &\cdots G'_{(n_l m_l)(n_{l+1} m_{l+1})} G_{(n_{l+1} m_{l+1})(n_{l+2} m_{l+2})} \cdots \\ &\cdots G''_{(n_p m_p)(n_{p+1} m_{p+1})} \cdots G_{(n_L m_L)(n_1 m_1)} \\ &= \text{Tr} G^{l-1} G' G^{p-l+1} G'' G^{L-p} \end{aligned} \quad (14)$$

The matrices  $G$ ,  $G'$  and  $G''$  are

$$\begin{aligned} G &= \begin{pmatrix} 1 & 0 & 0 & 2 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 2 & 0 & 0 & 1 \end{pmatrix} \\ G' &= \begin{pmatrix} 0 & -2 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \end{pmatrix} \\ G'' &= \begin{pmatrix} 0 & 0 & -2 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 \end{pmatrix} \end{aligned} \quad (15)$$

The eigenvalues and eigenvectors of  $G$  are :

$$\begin{aligned} \lambda_1 = 3, \quad \lambda_2 = \lambda_3 = \lambda_4 = -1 \\ |e_1\rangle = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ 0 \\ 0 \\ 1 \end{pmatrix} \quad |e_2\rangle = \frac{1}{\sqrt{2}} \begin{pmatrix} -1 \\ 0 \\ 0 \\ 1 \end{pmatrix} \\ |e_3\rangle = \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix} \quad |e_4\rangle = \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix} \end{aligned} \quad (16)$$

From (14) and in the thermodynamic limit  $L \rightarrow \infty$ ,

$$\begin{aligned} \langle \psi_{Bl}^1(k) | \psi_{Bp}^1 \rangle &= \sum_{n=1}^4 \lambda_1^{l-1} \langle e_1 | G' | n \rangle \lambda_n^{p-l+1} \langle n | G'' | e_1 \rangle \lambda_1^{L-p} \\ &= \frac{1}{2} 3^L \left(-\frac{1}{3}\right)^{p-l} \end{aligned}$$

Therefore,

$$\begin{aligned} \langle \psi_B^1(k) | \psi_B^1(k) \rangle &= \frac{1}{L} \sum_{l=1}^L \sum_{p=1}^L e^{ik(l-p)} \frac{1}{2} 3^L \left(-\frac{1}{3}\right)^{|p-l|} \\ &= 3^L \frac{2}{5 + 3 \cos k} \end{aligned} \quad (17)$$

The chain of A-spins is in the Néel state  $|\psi_{Néel}\rangle$ . The total wave function  $|\psi\rangle$  of the two-chain system is a product of  $|\psi_B^1(k)\rangle$  and  $|\psi_{Néel}\rangle$ . The norm  $\langle\psi|\psi\rangle$  is given by the same expression as in (17). The excitation energy measured w.r.t. the ground state energy is

$$\omega_1(k) = \frac{\langle\psi|H_{AB}|\psi\rangle}{\langle\psi|\psi\rangle} \quad (18)$$

The denominator has already been calculated. We now calculate the numerator.  $H_{AB}$  for single chain is given by Eq. (2). The two nearest-neighbour sites of the lower chain in Fig. 2 and the intermediate site in the upper chain form the vertices of a triangle. If one of the sites of the lower chain has the ‘defect’ matrix  $g^1$  associated with it, then there are four possible states of the triangle :

$$\begin{aligned} |\varphi_{1j}\rangle &\equiv | \begin{array}{c} \uparrow \\ g_j^1 \otimes g_{j+1} \end{array} \rangle, & |\varphi_{2j}\rangle &\equiv | \begin{array}{c} \uparrow \\ g_j \otimes g_{j+1}^1 \end{array} \rangle \\ |\varphi_{3j}\rangle &\equiv | \begin{array}{c} \downarrow \\ g_j^1 \otimes g_{j+1} \end{array} \rangle, & |\varphi_{4j}\rangle &\equiv | \begin{array}{c} \downarrow \\ g_j \otimes g_{j+1}^1 \end{array} \rangle \end{aligned} \quad (19)$$

One can verify that

$$\begin{aligned} H_{j,j+1} |\varphi_{2j}\rangle &= 0 \\ H_{j,j+1} |\varphi_{4j}\rangle &= 0 \end{aligned}$$

$$\begin{aligned}
H_{j,j+1} | \varphi_{1j} \rangle &= \uparrow \frac{1}{20} \begin{pmatrix} -8\sqrt{2}(|10\rangle + |01\rangle) & -40|11\rangle \\ 8|00\rangle + 4|\bar{1}\bar{1}\rangle + 4|1\bar{1}\rangle & 8\sqrt{2}(|01\rangle + |10\rangle) \end{pmatrix} \\
&\quad + \downarrow \frac{1}{20} \begin{pmatrix} -8|11\rangle & 0 \\ 4\sqrt{2}(|01\rangle + |10\rangle) & 8|11\rangle \end{pmatrix} \\
H_{j,j+1} | \varphi_{3j} \rangle &= \uparrow \frac{1}{20} \begin{pmatrix} -8|00\rangle - 4|1\bar{1}\rangle - 4|\bar{1}\bar{1}\rangle & -8\sqrt{2}(|10\rangle + |01\rangle) \\ 4\sqrt{2}(|\bar{1}0\rangle + |0\bar{1}\rangle) & 8|00\rangle + 4|1\bar{1}\rangle + 4|\bar{1}\bar{1}\rangle \end{pmatrix} \\
&\quad + \downarrow \frac{1}{20} \begin{pmatrix} -4\sqrt{2}(|10\rangle + |01\rangle) & -8|11\rangle \\ 8|00\rangle + 4|1\bar{1}\rangle + 4|\bar{1}\bar{1}\rangle & 4\sqrt{2}(|01\rangle + |10\rangle) \end{pmatrix} \quad (20)
\end{aligned}$$

The up and down spins outside the brackets represent the states of the A-spin.

We represent  $|\psi\rangle$  by the state

$$|\psi\rangle = \begin{vmatrix} \psi^{Néel} \\ \psi_B^1(k) \end{vmatrix} \quad (21)$$

in which the upper and lower rows contain the states of the upper and lower chains respectively. One can verify that

$$\begin{aligned}
&\left\langle \begin{vmatrix} \psi^{Néel} \\ \psi_{B_{j'}}^1(k) \end{vmatrix} \middle| H_{AB} \middle| \begin{vmatrix} \psi^{Néel} \\ \psi_{B_j}^1(k) \end{vmatrix} \right\rangle \\
&= \delta_{jj'} \left\langle \begin{vmatrix} \psi^{Néel} \\ \psi_{B_j}^1(k) \end{vmatrix} \middle| (H_{AB})_{jj+1} \middle| \begin{vmatrix} \psi^{Néel} \\ \psi_{B_j}^1(k) \end{vmatrix} \right\rangle \\
&= \delta_{jj'} \text{Tr} G^{j-1} Z(H) G^{L-j-1} \quad (22)
\end{aligned}$$

where the matrix G is given in (15) and

$$\begin{aligned}
Z(H)_{\mu_1\mu_2} &= Z(H)_{(n_1m_1)(n_2m_2)} \\
&= (\varphi_{1j}^\dagger)_{n_1n_2} [(H_{AB})_{jj+1} \varphi_{1j}]_{m_1m_2} \quad (j \text{ odd}) \\
&= (\varphi_{3j}^\dagger)_{n_1n_2} [(H_{AB})_{jj+1} \varphi_{3j}]_{m_1m_2} \quad (j \text{ even}) \quad (23)
\end{aligned}$$



From Eqns. (20)

$$\begin{aligned}
Z^{odd}(H) &= \frac{1}{20} \begin{pmatrix} 16 & 0 & 0 & 80 \\ 0 & -16 & 0 & 0 \\ 0 & 0 & -16 & 0 \\ 8 & 0 & 0 & 16 \end{pmatrix} \\
Z^{even}(H) &= \frac{1}{20} \begin{pmatrix} 8 & 0 & 0 & 16 \\ 0 & -8 & 0 & 0 \\ 0 & 0 & -8 & 0 \\ 8 & 0 & 0 & 8 \end{pmatrix}
\end{aligned} \tag{24}$$

In the limit  $L \rightarrow \infty$ , Eq. (22) reduces to

$$\begin{aligned}
&\left\langle \begin{array}{c} \psi_{Néel} \\ \psi_{B_j}^1(k) \end{array} \middle| (H_{AB})_{jj+1} \middle| \begin{array}{c} \psi_{Néel} \\ \psi_{B_j}^1(k) \end{array} \right\rangle \\
&= \langle e_1 | G^{L-2} Z(H) | e_1 \rangle \\
&= \lambda_1^{L-2} \langle e_1 | Z(H) | e_1 \rangle \\
&= \frac{1}{20} \times 3^{L-2} \times 60 \quad \text{for } j = \text{odd} \\
&= \frac{1}{20} \times 3^{L-2} \times 20 \quad \text{for } j = \text{even}
\end{aligned} \tag{25}$$

$\lambda_1$  and  $|e_1\rangle$  are given in Eq. (16). From (7),

$$\begin{aligned}
&\langle \psi | H_{AB} | \psi \rangle \\
&= \frac{1}{L} \sum_{j'} \sum_j e^{ik(j'-j)} \delta_{jj'} \left\langle \begin{array}{c} \psi_{Néel} \\ \psi_{B_j}^1(k) \end{array} \middle| (H_{AB})_{jj+1} \middle| \begin{array}{c} \psi_{Néel} \\ \psi_{B_j}^1(k) \end{array} \right\rangle \\
&= \frac{1}{20} \times \frac{3^{L-2}}{L} \left( \frac{L}{2} 60 + \frac{L}{2} 20 \right) \\
&= 3^{L-2} \times 2
\end{aligned} \tag{26}$$

From (18), (17) and (26), one gets

$$\omega_1(k) = \frac{1}{9} (5 + 3 \cos k) \tag{27}$$

The excitation energy (27) has been determined for a two-chain strip. For the 2D spin system, the chain of B-spins is connected to two chains of A-spins and the excitation energy is twice of that in (27). The excitation energy  $\omega_{-1}(k)$  for  $S^z = -1$  is the same as  $\omega_1(k)$ . The excitation energy  $\omega_0(k)$  for  $S^z = 0$  is  $2\omega_1(k)$ . Thus the gaps in the excitation spectrum for the 2D spin system are :

$$\Delta_{\pm 1} = \frac{4}{9} \quad , \quad \Delta_0 = \frac{8}{9} \quad (28)$$

We now consider the case when excitations are created in both the A and B-chains. The excitation in the A-chain travels in the network of A-spins as a 2D spin wave. The excitation spectrum of the spin wave is gapless. The Haldane gap excitation is confined to the B-spin chain. For a ferromagnetic ordering of A-spins, one can verify that  $\Delta_{+1} \neq \Delta_{-1}$ , i.e., the degeneracy of the triplet Haldane modes is fully lifted.

### III. Conclusion

We have constructed a model mixed spin system for which the ground state can be exactly specified. The coexistence of AFM long range order and Haldane gap excitations has been explicitly shown. In the ordered phase, the degeneracy of the three Haldane gap modes is partially lifted. The transverse modes ( $S^z = \pm 1$ ) are still degenerate whereas the longitudinal mode ( $S^z = 0$ ) has an excitation gap twice that of the transverse modes. These results are in agreement with the experimental observations on the  $R_2BaNiO_5$  systems.<sup>7-8</sup> In these systems, coexistence of gapless spin waves in an ordered phase and the Haldane gap excitations confined to Ni chains, has been observed. The three Haldane gap modes in an isolated chain of integer spins are degenerate. The lifting of the degeneracy seen in  $R_2BaNiO_5$  is due to the ordering of the rare-earth spins.<sup>8</sup> It has been suggested<sup>12</sup> that in  $R_2BaNiO_5$  the Ni-spins participate in the long range order. This is, however, not so in our model, as in the ground state there is a complete decoupling of the A and B-spins. An important feature of the  $R_2BaNiO_5$  systems is the increase in the gap energy as the temperature is lowered, i.e., as the system becomes more ordered<sup>12</sup>. Finite temperature calculations on our model are in progress and the results will be reported elsewhere.

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## Figure Captions

Fig. 1. Model mixed spin system consisting of a square lattice of  $S = 1/2$  spins (A-spins, solid circle). The centre of each square plaquette is occupied by a  $S = 1$  spin (B-spin, solid square) .

Fig. 2. A chain of A-spins (upper row) connected to a chain of B-spins (lower row).



