Spin structure factor and quantum phases of frustrated spin-1/2 chains

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Abstract – The static structure factor $S(q)$ of frustrated spin-1/2 chains with isotropic exchange and a singlet ground state (GS) diverges at wave vector $q_m$ when the GS has quasi-long-range order (QLRO) with periodicity $2\pi/q_m$ but $S(q_m)$ is finite in bond-order-wave (BOW) phases with finite-range spin correlations. Exact diagonalization and density matrix renormalization group (DMRG) calculations of $S(q)$ indicate a decoupled phase with QLRO and $q = \pi/2$ in chains with large antiferromagnetic exchange between second neighbors. $S(q_m)$ identifies quantum phase transitions based on GS spin correlations.

Received: 29 April 2010; Accepted: 15 May 2010

PACS 75.10.Jm – Quantized spin model including frustrations
PACS 64.70.Tg – Quantum phase transition
PACS 73.22.Gk - Broken symmetry phases

1. Introduction

The $J_1J_2$ model with isotropic exchange $J_1J_2 > 0$ between first and second neighbors is the prototypical frustrated spin-1/2 chain with a bond-order-wave (BOW) phase. 1–15 The Hamiltonian with periodic boundary conditions (PBC) and frustration $g = J_2/J_1 > 0$ is

$$H(g) = \sum_p (\vec{s}_p \cdot \vec{s}_{p+1} + g \vec{s}_p \cdot \vec{s}_{p+2})$$

$$= g (H_A + H_B) + \sum_p \vec{s}_p \cdot \vec{s}_{p+1}$$

(1)

$H_A$ and $H_B$ are linear Heisenberg antiferromagnets (HAFs) with PBC on sublattices of odd and even-numbered sites, and $H(0)$ is also an HAF. The ground state (GS) of Eq. 1 is a singlet, $S=0$. The infinite chain has nondegenerate GS at small $g$ that becomes doubly degenerate 16 at $g^*= 0.2411$, the boundary of the BOW phase with broken inversion symmetry at sites and a finite energy gap 17 $E_{\text{ed}}(g)$ to the lowest triplet state. The exact GS at the Majumdar-Ghosh point, 1 $g = 1/2$, are the Kekulé diagrams [K1] and [K2] of organic chemistry that correspond to singlet-paired spins on adjacent sites. Recent studies 16–20 have focused on ferromagnetic $J_1 < 0$ as the starting point for modeling oxides with chains of $s = 1/2$ Cu(II) ions.

Bursill et al. 10 studied the static spin structure factor $S(q;g)$ of the $J_1J_2$ model and took the $S(q_m)$ peak as the effective periodicity $2\pi/q_m$. They compared $q_m$ to chains of classical spins, for which the GS energy of Eq. 1 goes as $\cos \chi + g \cos^2 \chi$ where $\chi$ is the pitch angle between successive spins. Minimization gives $\cos \chi = -1/4g$, or $\chi = \pi$ for $g < 1/4$ and a continuous decrease to $\chi = \pi/2$ as $g \to \infty$. Quantum effects 21–22 are pronounced at small $g$, where $q_m = \pi$ persists to $g = 1/2$. The BOW phase extends to arbitrarily large $g$ according to Bursill et al. 10 and the field theories of White and Affleck 11 and Itoi and Qin 13. We find instead that the BOW phase terminates at $1/g^{**} \sim 0.40$ at the start of a gapless decoupled phase 21,22 with nondegenerate GS. We return in the Discussion to reasons for reexamining the quantum phase diagram at large $g$.

In this paper, the magnitude of $S(q_{m;g})$ is applied to the quantum phase diagram of frustrated spin chains. $S(q_{m;g})$ diverges when the GS has quasi-long-range order (QLRO) at wave vector $q_m$. The HAF at $g = 0$ has QLRO($\pi$) while the BOW phase has finite $S(q_{m;g})$ and spin correlations that are just to nearest neighbors at $g = 1/2$. The quantum transition between the QLRO($\pi$) and BOW phases is the largest $g$ at which $S(\pi;g)$ diverges; as shown in Section 3, this agrees with $g^{*} = 0.2411$ based 17 on the degeneracy, $E_m = E_{\text{ed}}$, of the triplet and lowest singlet excitations. HAFs on sublattices at $1/g = 0$ have QLRO($\pi/2$) and divergent $S(\pi/2;\infty)$. The largest $1/g$ at which $S(\pi/2;g)$ diverges marks the transition from the decoupled to the BOW phase. In our analysis, the frustrated BOW phase with finite $S(q;g)$ is intermediate between phases with dominant QLRO($\pi$) at small $g$ and QLRO($\pi/2$) at small $1/g$.

We obtain $S(q;g)$ using exact diagonalization (ED) of finite $J_1J_2$ models, density matrix

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renormalization group (DMRG) calculations and extrapolation to the infinite chain. The procedure is general for spin chains. Sections 2 and 3 present \( S(q; g) \) results and the size dependence of \( S(q; g) \), respectively. In Section 4 we briefly discuss the gapless decoupled phase and specific challenges of solving \( H(g) \) at large \( g \).

2. Static structure factor \( S(q) \)

The static structure factor \( S(q) \) of 1D systems with one spin per unit cell is the GS expectation value

\[
S(q) = \frac{1}{N} \sum_{p,r} \left< \hat{S}_p \cdot \hat{S}_{1+p} \right> e^{iq(p-r)} = \sum_p \left< \hat{s}_1 \cdot \hat{s}_{1+p} \right> e^{iqp}
\]

(2)

The wave vectors in the first Brillouin zone are \( q = \frac{2m\pi}{N} \) with \( m = 0, \pm1, \ldots, N/2 \). We define spin correlation functions \( C(p,g) = \left< s_1 s_{1+p} \right> \) at frustration \( g \) in Eq. 1 and consider \( S(q;g,4n) \) with \( N = 4n \) spins that ensure integer total spin \( S \leq n \) and sublattice spin \( S_n \leq n \). The peak at \( q = 0 \) component satisfies \( S(0; g) = \frac{S^2}{4n} \) when the GS is a singlet; the sum of \( C(p,g) \) over \( p \) is zero; summing over \( q \) in the Brillouin zone and taking the limit \( n \to \infty \) leads to

\[
\frac{1}{4n} \sum_q S(q; g, 4n) = \frac{3}{4}
\]

(3)

since \( C(0,g) = 3/4 \) for \( s = 1/2 \).

If the \( C(p,g) \) have finite range, \( S(q;g) \) is finite and the sum in Eq. 2 becomes constant once the system size exceeds the correlation length. For even \( N \) in Eq. 2, the exact GS at \( g = 1/2 \) gives

\[
S(q; 1/2) = 3(1 - \cos q)/4
\]

(4)

The size dependence is entirely in the discrete \( q \) values. A finite energy gap \( E_m(g) \) in the BOW phase indicates a localized GS and finite-range spin correlations. \( S(q;g) \) is readily found directly for some \( g \) in the BOW phase. The lower panel of Fig. 1 contrasts \( S(q;g) \) at \( g = 0 \) and 3 with \( g = 1/2 \). Open symbols are exact \( S(q;g,24) \) at \( g = 0 \) and 3; the dashed and solid lines are DMRG results for 48 and 100 spins, respectively. Quite generally, we have \( S(q;0,4n) = S(q/2,3/4,8n) \) since both \( g = 0 \) and 1/2 correspond to 4n-spin HAFs. The \( q_n = \pi \) peak for 24 spins at \( g = 0 \) is almost exactly equal to the \( q_n = \pi/2 \) peak for 48 spins at \( g = 3 \). Fig. 1 already suggests that the BOW phase does not extend to \( g = 3 \). As shown in Section 3, the lowest-order changes go as

\[
S(\pi; g, 4n) = S(\pi; 0,4n) - A_n g
\]

(5)

\[
S(\pi/2; 1/ g, 8n) = S(\pi/2; 0,8n) - B_n / g^2
\]

with \( A_n, B_n > 0 \). Since the peaks are equal at \( g = 0 = 1/g \), the \( \pi/2 \) peak in finite systems is less sensitive to frustration \( 1/g << 1 \) than the \( \pi \) peak is to \( g << 1 \).

The wave vector \( q_n \) is shown in Fig. 2 as a function of \( g/(1 + g) \). Open circles are exact for 24 spins. The peak remains at \( \pi \) up to \( g = 1/2 \) and then decreases to...
Fig. 2. Wave vector $q_m$ of the structure factor peak $S(q_m)$ of the J$_1$J$_2$ model with $N = 24$ spins as function of frustration $g/(1 + g)$. The chain of classical spins has pitch angel $\chi = \pi$ for $g \leq 1/4$ and $\chi = \cos^{-1}(-1/4g)$ for $g > 1/4$.

$q_m = \pi/2$. Classical spins have pitch angle $q_m$ with $\cos q_m = -1/4g$ for $g > 1/4$, doubly degenerate GS with long-range Néel order up to $g = 1/4$, and a spiral GS with LRO($q_m$) for $g > 1/4$. We find strong quantum effects at large $g$ that compress the BOW phase and lock in $q_m = \pi/2$.

3. Phase transitions

The static structure factor identifies the three quantum phases of the J$_1$J$_2$ model. The location of phase transitions is more demanding. Finite $N$ in Eq. 2 clearly gives finite $S(q)$. We must infer whether $S(\pi;g,4n)$ or $S(\pi/2;g,4n)$ diverges with increasing system size rather than merely becoming large. The numerical problem is to compute all spin correlations $C(p,g)$ in systems of $N = 4n$ spins. We use ED up to 24 spins and a finite DMRG algorithm for larger systems with four spins added per step$^{15}$ and cyclic boundary conditions.$^{24}$ The algorithm is more accurate than conventional DMRG because adding four spins per step ensures that the sublattices always have $S_A = S_B = 0$ at $1/g = 0$ rather than $S_A = S_B = 1/2$ at every other step. Truncation errors in the sum of the eigenvalues of the density matrix are less than $10^{-10}$ in the worst case when $m = 200$ eigenvalues are kept. Finite size effects increase at large $g$, DMRG returns $C(p,g)$ whose accuracy can be tested rigorously by comparison to the exact result, $S(0;g,4n) = 0$. We find $S(0;g,100) < 10^{-3}$ in the QLRO($\pi$) phase up to $4n = 100$ and comparable accuracy to $4n = 64$ in the QLRO($\pi/2$) phase.

We also rely on HAF spin correlation functions$^{23}$ that establish the divergence of $S(\pi,0)$ or $S(\pi/2,\infty)$. The $q = \pi$ term of Eq. 2 for $4n$ spins is

$$S(\pi; g, 4n) = \frac{3}{4} + C(2n, g) + 2 \sum_{p=1}^{2n-1} C(p, g)(-1)^p$$

(6)

Since $C(p,0)$ goes as $(-1)^p$, the sum is over $|C(p,0)|$. As shown in the inset to Fig. 3, $S(\pi;g,4n)$ is a linear function at small $g$ with slope $-A_n$ and $A_n = 1.6$ for 24 spins. Finite $g > 0$ is frustrating while $g < 0$ enhances short-range $q = \pi$ order.

Incremental increases of $S(\pi;g,4n)$ from $4n$ to $4n + 4$ spins are shown in Fig. 3 as a function of $100/N$ with $N = 4n + 2$, followed by linear extrapolation to the infinite chain. $S(\pi;0.40,4n)$ converges rapidly as noted in Fig. 1. Within our accuracy, $S(\pi;g)$ diverges at $g = 0.20$ and converges at $g = 0.25$. The estimated $g^*$ between 0.20 and 0.25 based on the structure factor is consistent with, but much less precise than $g^* = 0.2411$ based$^8$ on $E_m = E_n$. The two methods are independent since the GS determines $S(\pi;g)$ but does not enter in the excited-state degeneracy.

Only spin correlations within one sublattice contribute to $S(\pi/2)$

$$S(\pi/2; g, 4n) = \frac{3}{4} + C(2n, g)(-1)^n$$

(7)

$$+ 2 \sum_{p=1}^{n-1} C(p, g) \cos(\pi p/2)$$

Fig. 3. Incremental increase of the structure factor peak $S(\pi,4n)$ from $n$ to $n + 1$ as a function of $1/N$ with $N = 4n + 2$ using ED up to 24 spins. DMRG to 100 spins and linear extrapolation to the infinite chain; $S(\pi;g)$ diverges at $g = 0.20$, converges at $g = 0.25$. Inset: linear dependence of $S(\pi;g)$ on $g$ near the origin for 16, 20 and 24 spins.
The $\pi/2$ peak for 8n spins reduces as expected to Eq. 6. In contrast to $S(\pi; g)$ at small $g$, however, there is no linear contribution in $1/g$ because $J_z > 0$ is frustrating for either sign of $J_1$. The first-order correction $|\psi\rangle$ in $1/g$ is given by

$$
(H_A + H_B - 2E_0)|\psi\rangle = -\frac{1}{g} \sum_p \hat{s}_p \cdot \hat{s}_{p+1} |G_A\rangle |G_B\rangle
$$

(8)

$H_A$ and $H_B$ are HAFs on sublattices whose singlet GS and energy are $|G\rangle$ and $E_0$. Adjacent spins generate a singlet linear combination of triplets on each sublattice; $|\psi\rangle$ is a linear combination of such product states. Without explicitly solving Eq. 8, we obtain the general result for $N$

$$
\langle \phi \big| \hat{s}_1 \cdot \hat{s}_{1+2p} |G_A\rangle |G_B\rangle = 0
$$

(9)

When both spins are in the same sublattice, the matrix element is zero since the triplet and GS of the other sublattice are orthogonal. It follows that $C(2p; g)$ and hence $S(\pi/2; g, 8n)$ initially decreases as $1/g^2$.

Figure 4 shows incremental increases of $S(\pi/2, 8n)$ from 8n to 8n + 8 spins as a function of $100/N$ with $N = 8n + 4$, followed by linear extrapolation to the infinite chain. The $1/g = 0$ points to 200 spins are $g = 0$ results to 100 spins. As noted above, shorter chains of 64 spins meet the requirement of $S(0; g) < 10^{-3}$ at large $g$. $S(\pi/2, g)$ converges and is clearly finite at $g = 2.0$ in the BOW phase. The estimated transition $g^{**}$ between the BOW and decoupled phases is around $1/g^{**} \approx 0.40$. As shown in the inset, $S(\pi/2, g, 8n)$ initially goes as $-B/dg^2$ with $B_d = 0.17$ for 24 spins and is almost constant.

4. Discussion

We have related the structure factor peak, $S(q_m; g)$, to the quantum phases of the $J_1J_2$ model, Eq. 1. $S(q_m; g)$ diverges at $q_m = \pi$ up to $g^*$ in the spin liquid phase with QLRO($\pi$), is finite in the frustrated BOW phase between $g^*$ and $g^{**}$, and diverges for $g > g^{**}$ in the gapless decoupled phase with QLRO($\pi/2$). We now address conflicting results that extend the BOW phase to $1/g = 0$.

To start with, theoretical and numerical works\textsuperscript{13} have focused mainly on the quantum phase transition at $g^*$ to the BOW phase and recent studies\textsuperscript{16-20} of Eq. 1 also deal with other sectors than large $g$. Interesting and exotic GS are generated by an external magnetic field, by anisotropic or antisymmetric rather than isotropic exchange, by changing the sign of $J_1$, or by increasing the range of exchange interactions. The magnetic properties of organic and inorganic crystals that contain spin chains provide other applications.

There are several reasons for a closer look at the $1/g \ll 1$ regime. First, the initial DMRG calculations\textsuperscript{13} were limited to $1/g > 0.5$, far from the limit. Second, Okamoto and Namura\textsuperscript{a} used ED in finite systems to obtain $g^*$ from the degeneracy $E_m = E_0$, the same degeneracy at $1/g^{**}$ was not pointed out until later.\textsuperscript{14} As a matter of consistency, ED in finite systems cannot decide for locating the phase transition at $g^*$ but irrelevant at $g^{**}$. Third, exact HAF states describe both limits. ED of Eq. 1 with 4n spins yields n points $g_n$ with doubly degenerate GS and broken inversion symmetry, starting with $g_1 = 1/2$. The degenerate GS at the largest $g_n$ are closely related\textsuperscript{21} to the product of sublattice ground states, $|G_1\rangle |G_0\rangle$, and the singlet linear combination of the lowest triplets, $|T_0\rangle$. In view of the insets to Figs. 3 and 4, it would be remarkable have a nondegenerate GS with divergent $S(\pi; g)$ up to $g^*$ while strictly limiting nondegenerate GS and divergent $S(\pi/2; g)$ to $1/g = 0$.    

![Fig. 4. Incremental increase of the structure factor peak S(π/2, 8n) from n to n + 1 as a function of 1/N with N = 4n + 4 using ED up to 24 spins, DMRG to 64 spins and linear extrapolation to the infinite chain: S(π/2, g) diverges at 1/g = 0.33, converges at 1/g = 0.50. Inset: quadratic dependence of S(π/2, g) on 1/g near the origin for 16, 20 and 24 spins; the maxima are at 1/g = 0.]

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The large-g sector of Eq. 1 is particularly challenging, a point that may be relevant to spin chains as many-body problems. Field theories\textsuperscript{11,13} starting with an HAF at \( g = 0 \) lead to different expressions for \( E_m \) and rely on the same limited\textsuperscript{11} DMRG for numerical support. Allen and Senechal\textsuperscript{12} start with two HAFs at \( 1/g = 0 \) and discuss three different continuum descriptions of Eq. 1 along with various approximations. Turning to DMRG, we note that open boundary conditions (OBC) are typically used for an even number of spins. Quite aside from strong end effects,\textsuperscript{15} inversion symmetry at sites is lost for even \( N \). We find doubly degenerate GS and broken inversion boundary conditions (OBC) are typically used for an even number of spins. Quite aside from strong end effects,\textsuperscript{15} inversion symmetry at sites is lost for even \( N \).

The magnitude of \( S(q;g) \) bears directly on the quantum phases of frustrated spin chains. ED partly compensates for finite-size limitations by returning exact correlation functions \( C(p,g) \). \( S(q;g) \) is found directly for short-range correlation. Extrapolation to infinite chains is guided by the known HAF divergences of \( S(\pi,0) \) or \( S(\pi/2,\infty) \). But extrapolation entails approximations. Numerical methods and field theory are in agreement for the quantum transition of the J\(_1\)J\(_2\) model from the QLRO (\( \pi \)) to BOW phase at \( g^* = 0.2411 \), but disagree at present at large \( g \). The peak \( S(q_m;g) \) is finite in the BOW phase, diverges at \( q_m = \pi \) for \( g < g^* \) in the spin liquid phase with QLRO (\( \pi \)) and at \( q_m = \pi/2 \) for \( 1/g > 1/g^* \) in the decoupled phase. Frustrated spin chains whose GS has LRO (\( \pi \)) at \( g = 0 \) undergo a first order quantum transition\textsuperscript{21} with increasing \( g \) directly to the decoupled phase. The transition occurs at \( g_c = 1/4\ln 2 \) in an analytical model\textsuperscript{21} with equal \( J = 2/(4n - 1) \) between spins in opposite sublattices and \( -J \) between spins in the same sublattice.

Acknowledgments: We thank D. Sen, A.W. Sandvik and S. Ramasesha for instructive discussions of BOW phase systems and the NSF for partial support of this work through the Princeton MRSEC (DMR-0819860). MK thanks DST for a Ramanujan Fellowship and support for thematic unit of excellence on computational material science.

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