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Multimagnon bound states in the frustrated ferromagnetic one-dimensional chain

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We study a one-dimensional Heisenberg chain with competing ferromagnetic nearest-neighbor and antiferromagnetic next-nearest-neighbor interactions in a magnetic field. Starting from the fully polarized high-field state, we calculate the dispersions of the lowest-lying n-magnon excitations and the saturation field ($n=2,3,4$). We show that the lowest-lying excitations are always bound multimagnon states with a total momentum of π except for a small parameter range. We argue that Bose condensation of the bound n magnons leads to novel Tomonaga-Luttinger liquids with multipolar correlations; nematic- and triatic-ordered liquids correspond to $n=2$ and $n=3$.

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Frustrated spin chains are simple models that still show surprisingly rich physics. A prototype of these is the spin-$\frac{1}{2}$ Heisenberg chain with nearest-neighbor (NN) J_1 and next-nearest-neighbor (NNN) J_2 couplings,

$$H=\sum_{l\in\mathbb{Z}}(J_1\vec{S}_l\cdot\vec{S}_{l+1}+J_2\vec{S}_l\cdot\vec{S}_{l+2}-hS_l^z), \tag{1}$$

where $\vec{S}_l=(S_l^x,S_l^y,S_l^z)$ is the spin-1/2 operator on the lth site and the external field h is applied in the z direction. The exchange interactions are frustrated as the NNN interaction is antiferromagnetic (AF), $J_2>0$. Until recently most theoretical studies of the J_1-J_2 model (1) considered the case where the NN coupling J_1 is also AF. However, interest is now growing rapidly in the ferromagnetic (FM) case ($J_1<0$) as well, which is triggered by experimental reports on thermodynamic properties of various quasi-one-dimensional frustrated FM spin chains (for a list of frustrated quasi-one-dimensional materials, see Table 1 in Ref. 1 and Fig. 5 in Ref. 2). For example, $Rb_2Cu_2Mo_3O_{12}$ (Ref. 1) and $LiCuVO_4$ (Ref. 3) are considered to be described by the J_1-J_2 model with $J_1\approx-3J_2$ and $J_1\approx-0.3J_2$, respectively.

Recent theoretical studies[4,5] have shown that, in a magnetic field, the FM J_1-J_2 chain ($J_1<0,J_2>0$) becomes a Tomonaga-Luttinger (TL) liquid having nematic quasi-long-range order as dominant correlation for some range of J_1/J_2. This nematic state can be thought of as arising from condensation of two-magnon bound states.[6] Interestingly, such a nematic-ordered phase can also appear in a frustrated FM spin model of the two-dimensional square lattice.[7] One can imagine further a Bose-condensed phase of more-than-two-magnon bound states. Indeed it was shown[8] recently that an octupolarlike triatic-ordered phase can result from condensation of three-magnon bound states in a frustrated ferromagnet on the triangular lattice. These results motivated us to examine the possibility of such many-magnon bound states in the FM J_1-J_2 model (1). In this paper we show, by explicitly constructing bound-state wave functions, that for $-3.52\lesssim J_1/J_2\lesssim-2.72$ the lowest-lying excitations from the fully polarized FM state are three-magnon bound states, and moreover four-magnon bound states appear for a slightly stronger FM coupling regime. We then suggest exotic TL liquid phases with multipolarlike spin correlations to emerge from condensation of multimagnon bound states below the saturation field.

Before presenting our calculations, let us briefly review known results that are relevant to our study. First, in the classical limit, we may regard $\vec{S}_l$ as a c-number vector of length S. The ground state of Eq. (1) in this limit has a (right- or left-winding) helical spin structure, $\vec{S}_l/S=(\sin\theta\cos\phi_l,\sin\theta\sin\phi_l,\cos\theta)$, with a pitch angle of $\phi=\phi_{l+1}-\phi_l=\pm\arccos(-J_1/4J_2)$ and a canting angle of $\theta=\arccos[4hJ_2/S(J_1+4J_2)^2]$, so that spins are fully polarized at a saturation field of $h_s=S(J_1+4J_2)^2/4J_2$ if $-4J_2<J_1<0$ or at zero field if $J_1<-4J_2$. The helical order does not survive quantum fluctuations.

In the quantum spin-$\frac{1}{2}$ case, the ground state is fully polarized without magnetic field if $J_1<-4J_2$. At the boundary $J_1=-4J_2$ the zero-field ground state is highly degenerate such that states from vanishing magnetization to full polarization share the same energy.[9] For the parameter range $-4J_2<J_1<0$ of our main interest, Chubukov[6] suggested that the ground state just below the saturation field should be a nematic state made up of bound magnon pairs with a commensurate total momentum $k=\pi$ if $-2.67J_2<J_1<0$ ($2.67\approx1/0.38$) and with an incommensurate momentum $k<\pi$ otherwise, which was partly verified by mean-field theory,[10] numerical study,[11] Green's function analysis which fixed the commensurate-incommensurate transition point to $J_1/J_2=-2.669\,08(=-1/0.374\,661)$,[12] and weak-coupling bosonization analysis.[4,11,13] While earlier calculations of the ground-state magnetization process suggested metamagnetic transitions,[10,14] a recent density-matrix renormalization group (DMRG) study[11] finds that the total magnetization of finite-size chains changes by $\Delta S^z=2$ at $J_1=-J_2$, $\Delta S^z=3$ at $J_1=-3J_2$, and $\Delta S^z=4$ at $J_1=-3.75J_2$ below saturation, implying that the magnetization curve is continuous in the thermodynamic limit.[13]

To reveal how the fully polarized FM state collapses into new states with decreasing either the magnetic field or the coupling ratio $|J_1|/J_2$, we analyze magnon instability in the fully polarized state. We apply a large enough magnetic field h such that the fully polarized state is a unique ground state for $-4J_2<J_1<0$ and all multimagnon excitations have positive excitation energies which decrease as h is reduced. The saturation field h_s is then defined as the field at which the lowest excitation energy vanishes. We consider bound states of up to four magnons in a finite chain of N spins.

To explain our computational scheme, we begin with one- and two-magnon states. The one-magnon excited state with momentum k,

$$|k\rangle = \frac{1}{\sqrt{N}}\sum_{l=1}^{N} e^{ikl}S_l^-|\mathrm{FM}\rangle, \tag{2}$$

on the fully polarized state $|\mathrm{FM}\rangle=|\uparrow\uparrow\uparrow\cdots\uparrow\rangle$ has the excitation energy

$$\epsilon_1(k) = J_1(\cos k - 1) + J_2[\cos(2k) - 1] + h, \tag{3}$$

which has a minimum of

$$\epsilon_1(k_0) = -\frac{1}{8J_2}(J_1 + 4J_2)^2 + h \tag{4}$$

at $\cos k_0 = -J_1/4J_2$. Obviously the one-magnon instability just reproduces the classical helical spin state in the applied field below the saturation field. However, as shown by earlier studies,[4,6,10–12] this is not the true instability for the quantum case in the whole parameter region $-4J_2 < J_1 < 0$.

To calculate bound n-magnon excitations, we take n-magnon states with a center-of-mass (c.m.) momentum k as a basis. For example, our basis for the two-magnon excitations has two $\downarrow$ spins with a total momentum k and a relative distance r $(=1,2,\ldots)$,

$$|r;k\rangle = \frac{1}{\sqrt{N}}\sum_{l} e^{ik(2l+r)/2}S_l^-S_{l+r}^-|\mathrm{FM}\rangle. \tag{5}$$

In this basis the matrix elements of the Hamiltonian can be written as $\langle r;k|H|r';k'\rangle = \delta_{k,k'}H_{r,r'}$, where nonvanishing entries of $H_{r,r'}$ are

$$H_{r,r} = J_1(\delta_{r,1} - 2) + J_2(\delta_{r,1}\cos k + \delta_{r,2} - 2) + 2h,$$

$$H_{r,r+1} = H_{r+1,r} = J_1 \cos(k/2),$$

$$H_{r,r+2} = H_{r+2,r} = J_2 \cos k. \tag{6}$$

By separating off the c.m. motion we have reduced the two-magnon problem to a one-particle one which in principle can be solved exactly. For general k this involves finding the roots of a transcendental equation.[12] The eigenvalue problem (6) is greatly simplified at $k=\pi$, where the excitation energy of the two-magnon bound state is[6,12]

$$\epsilon_2(\pi) = -J_1 - 3J_2 + \frac{J_2^2}{J_1 - J_2} + 2h. \tag{7}$$

To calculate the energy dispersion $\epsilon_2(k)$ of the two-magnon bound states for general $k \in [0,\pi]$, we numerically diagonalize the matrix $H_{r,r'}$ by restricting r and r' up to 1000. As long as the maximum value of r is sufficiently larger than the size of bound states, finite-size corrections should be exponentially small.

The bound states of more than two magnons can be calculated in a similar manner. For the n-magnon sector, we take, as a basis set, states with total momentum k in which n down spins are separated by distance $r_1,\ldots,r_{n-1}$. For example, the three-magnon basis is given by

$$|r_1,r_2;k\rangle = \sum_{l} \frac{e^{ik(3l+2r_1+r_2)/3}}{\sqrt{N}} S_l^- S_{l+r_1}^- S_{l+r_1+r_2}^- |\mathrm{FM}\rangle, \tag{8}$$

with which the matrix elements $\langle r_1,r_2;k|H|r_1',r_2';k\rangle$ are easily found. The four-magnon basis states $|r_1,r_2,r_3;k\rangle$ are constructed similarly. To solve the three- and four-magnon bound states, we numerically diagonalized the Hamiltonian matrix expressed in terms of the finite number of basis states $|r_1,r_2;k\rangle$ with $1\leq r_i \leq 27$ and $|r_1,r_2,r_3;k\rangle$ with $1 \leq r_i \leq 9$, respectively. This seems to be sufficient to determine the lowest excitations.

The four panels of Fig. 1 show energy dispersions of multimagnon excitations at $J_1/J_2 = -1.0$, -2.7, -3.0, and -3.8. For each value of J_1/J_2 the magnetic field is set equal to the saturation field where the lowest-lying excited state is gapless. The dispersion of bound $n=2,3,4$ magnons is obtained by diagonalizing the Hamiltonian in the finite basis, as we described above. Our numerical calculation reproduces the known results for the two-magnon sector:[6,10,12] the lowest bound two-magnon excitation has momentum $k=\pi$ for $-2.67 \lesssim J_1/J_2 < 0$ and incommensurate momentum $k<\pi$ for $-4 < J_1/J_2 \lesssim -2.67$. Figure 1 also shows the lower edges (thin lines) of the continuum spectra of scattering states made up of n magnons, which are calculated from the dispersion of a single magnon (3) and those of bound states of up to $n-1$ magnons. For example, the energies of three-magnon scattering states are obtained by adding those of either three unbound magnons or of a two-magnon bound state plus a free magnon, under the condition that the total momentum be k (mod 2π).

We see in Fig. 1 that for any J_1/J_2 there is always a region in k space where bound states lie well below the scattering continuum. The character of the lowest-lying bound states signaling the instability of the fully polarized state changes with J_1/J_2. Unlike what was previously thought, the system shows rather different regimes as a function of J_1/J_2: a two-magnon commensurate ($k=\pi$) instability ($-2.67 < J_1/J_2 < 0$), a two-magnon incommensurate instability as predicted in Ref. 6 ($-2.72 < J_1/J_2 < -2.67$), a three-magnon commensurate instability ($-3.52 < J_1/J_2 < -2.72$), and a four-magnon commensurate instability ($-3.86 < J_1/J_2 < -3.52$).[15] As J_1/J_2 approaches further towards -4, we expect to have more-than-four-magnon commensurate phases, but these are outside the scope of our numerics. We note that, except in the incommensurate regime ($-2.72 < J_1/J_2 < -2.67$), the bottom of the lowest-lying mode is at $k=\pi$. For example, at $J_1/J_2 = -1.0$ the lowest-energy state in the two-magnon sector is the bound state with $k=\pi$. The four-magnon scattering states of two such two-magnon bound states therefore have the lowest energy (which becomes gapless when $h=h_s$) at $k=0$. The fact that we do not have a mode of four-magnon bound states near $k=0$ below the continuum indicates that the interaction between 2 two-magnon bound states of $k=\pi$ is repulsive, and therefore these two-magnon bound states are stable. At

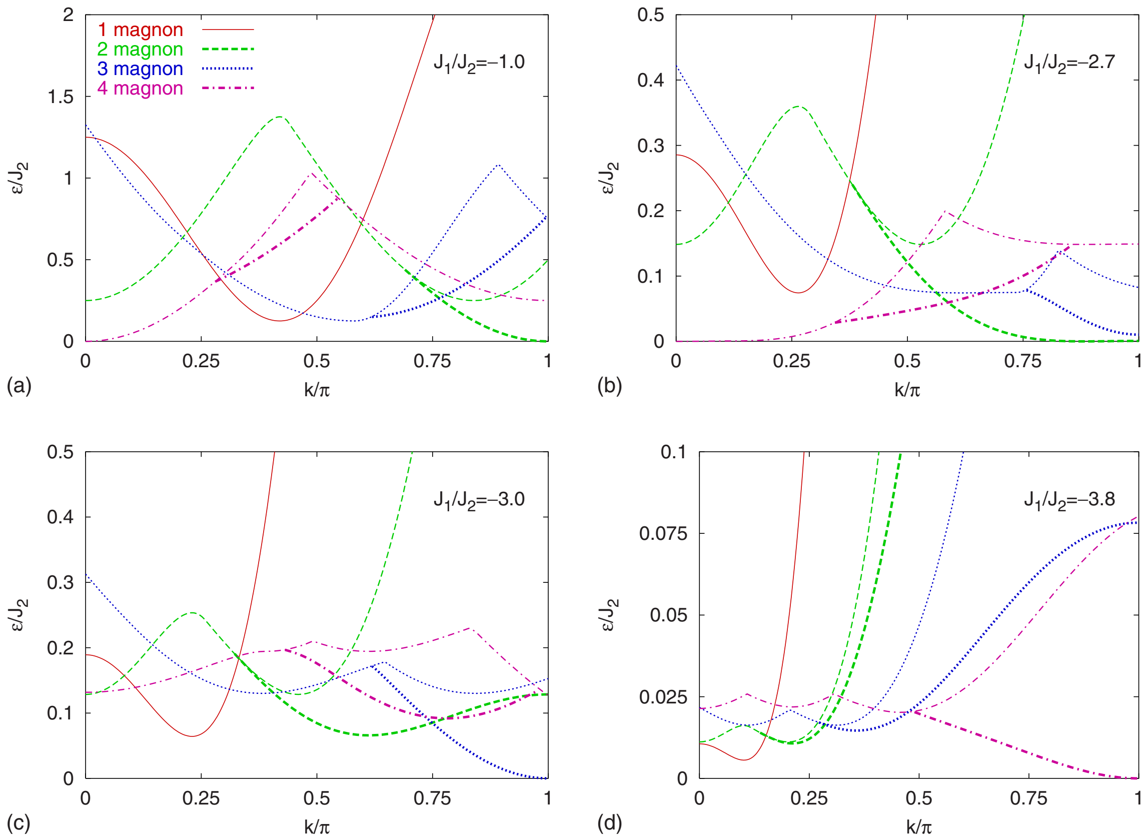

FIG. 1. (Color online) Energy dispersions for the multimagnon bands at the saturation field. Thin lines denote the onset of the scattering continuum, and thick lines show n-magnon bound states where relevant. With decreasing J_1/J_2, the lowest excitations change from commensurate ($k=\pi$) two-magnon bound states, incommensurate ($k<\pi$) two-magnon bound states, commensurate three-magnon bound states, to commensurate four-magnon bound states.

$J_1=-3J_2$ we have the three-magnon bound state with $k=\pi$ as the lowest-energy excitation. The presence of stable three- and four-magnon bound states provides a natural explanation for the $\Delta S^z>2$ jumps found in the DMRG study of Ref. 11.

Figure 2 shows the saturation field h_s determined by the instability from the softening of the lowest excitations. The saturation field estimated from a single-magnon instability is always smaller than the true saturation field which is determined by multimagnon bound states. The calculated saturation field is in perfect agreement with the exact h_s estimated from Eq. (7) for $-2.67\lesssim J_1/J_2<0$. For $-3.86<J_1/J_2<-2.72$ the saturation field is determined from the instability of three- or more-magnon bound states.

In Fig. 3 we show the expectation value $\langle\sum_{i=1}^{n-1}r_i\rangle$ characterizing the size of the n-magnon bound states, where the average is taken for the lowest-energy bound state of n magnons for a given J_1/J_2; the minimum value of the average is by definition $n-1$ for the n-magnon bound state. We confirm that the magnons are tightly bound when they are the lowest-lying excitations in the energy spectra. This justifies our use of basis states with finite r_i for calculating the low-lying bound states.

We now discuss the implications of the multimagnon instabilities that lead to condensation of bound magnons below the saturation field. We argue that the bound n-magnon condensation gives rise to a phase with multipolarlike quasi-long-range order but without spin (dipole) ordering in the XY direction. To be specific, let us consider the case where the n-magnon bound states with momentum $k=\pi$ become gapless as $h\to h_s+0$: $n=2$ for $-2.67<J_1/J_2<0$, $n=3$ for $-3.52<J_1/J_2<-2.72$, and $n=4$ for $-3.86<J_1/J_2<-3.52$. Just below the saturation field, the system can be viewed as a

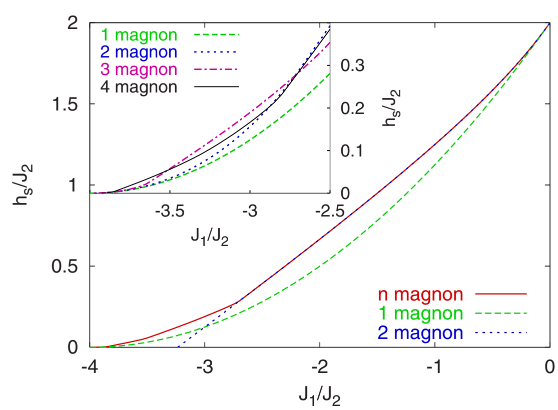

FIG. 2. (Color online) Saturation field versus J_1/J_2. The solid curve "n magnon" shows the saturation field h_s obtained from the numerical n-magnon solutions ($n=2,3,4$), while the curves "1 magnon" and "2 magnon" denote h_s calculated from Eqs. (4) and (7), respectively. The inset shows the region where our saturation field deviates from the two-magnon solution and the saturation fields for each multimagnon excitation.

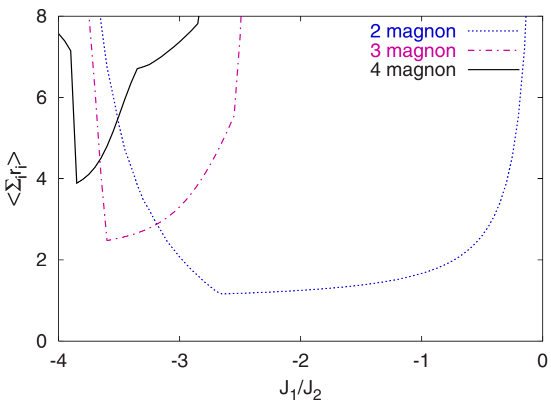

FIG. 3. (Color online) Mean length $\langle \Sigma_i r_i \rangle$ of the bound multimagnon states versus J_1/J_2. The kinks in the curves appear when the total momentum of the n-magnon bound state changes between incommensurate (where $\langle \Sigma_i r_i \rangle$ is larger) and commensurate ($k=\pi$).

dilute gas of repulsively interacting bosons which represent the n-magnon bound states.[16] Here the boson creation operator $b_i^\dagger$ is identified with $(-1)^i S_i^- S_{i+1}^- \cdots S_{i+n-1}^-$ in crude approximation. These bosons condense to form a TL liquid[16] in which various correlation functions of the boson fields decay algebraically. The most slowly decaying two-point correlation function will be the propagator of the bosons $\langle b_0 b_r^\dagger \rangle$, which decays as $(-1)^r r^{-1/\eta}$ with[17] $\eta \to 2$ as $h \to h_s - 0$. However, the spin operators that cannot be simply represented with bosons b_i, such as S_i^- and its products $\Pi_{j=i}^{i+p} S_j^-$ with $p=0,1,\ldots,n-2$, should have only short-range correlations. This is because exciting one magnon costs binding energy. Since n down spins form a tightly bound state, we may use the approximation $\frac{1}{2} - S_i^z = n b_i^\dagger b_i$. This allows us to calculate the correlation function of S_i^z from the density correlation of bosons, which has the asymptotic form $\langle b_0^\dagger b_0 b_r^\dagger b_r \rangle \sim \rho^2 + A r^{-\eta} \cos(2\pi\rho r) - \eta(2\pi r)^{-2}$, where ρ is the boson density $\left(n\rho = \frac{1}{2} - \langle S^z \rangle\right)$ and A is a constant. For the case $n=2$, the above theory indicates that the TL liquid has nematic quasi-long-range order,[6] which is indeed found by the recent DMRG calculation at $J_1 = -J_2$.[4,17] The theory also predicts that the TL liquid with larger n $(n=3,4)$ should exist for larger $|J_1|/J_2$ in the phase diagram of the FM J_1-J_2 model near the saturation field. For $n=3$ this is a TL liquid with antiferrotriatic quasi-long-range order, which is a one-dimensional analog of triatic order found in Ref. 8. Our numerics suggests that, as J_1/J_2 approaches -4, instability from bound states of more magnons appears. We do not know how far these new phases extend to lower fields.

In summary we have numerically calculated many-magnon bound states and determined their energy dispersions. We have found that the fully polarized FM state has instabilities to Bose condensation of these many-magnon bound states, which lead to TL liquids with multipolar magnetic correlations below the saturation field.

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[1] M. Hase, H. Kuroe, K. Ozawa, O. Suzuki, H. Kitazawa, G. Kido, and T. Sekine, Phys. Rev. B **70**, 104426 (2004).

[2] S.-L. Drechsler, O. Volkova, A. N. Vasiliev, N. Tristan, J. Richter, M. Schmitt, H. Rosner, J. Málek, R. Klingeler, A. A. Zvyagin, and B. Büchner, Phys. Rev. Lett. **98**, 077202 (2007).

[3] M. Enderle, C. Mukherjee, B. Fåk, R. K. Kremer, J.-M. Broto, H. Rosner, S.-L. Drechsler, J. Richter, J. Malek, A. Prokofiev, W. Assmus, S. Pujol, J.-L. Raggazzoni, H. Rakoto, M. Rheinstädter, and H. M. Rønnow, Europhys. Lett. **70**, 237 (2005).

[4] T. Vekua, A. Honecker, H.-J. Mikeska, and F. Heidrich-Meisner, arXiv:0704.0764.

[5] See also A. Kolezhuk and T. Vekua, Phys. Rev. B **72**, 094424 (2005).

[6] A. V. Chubukov, Phys. Rev. B **44**, 4693 (1991).

[7] N. Shannon, T. Momoi, and P. Sindzingre, Phys. Rev. Lett. **96**, 027213 (2006).

[8] T. Momoi, P. Sindzingre, and N. Shannon, Phys. Rev. Lett. **97**, 257204 (2006).

[9] T. Hamada, J. Kane, S. Nakagawa, and Y. Natsume, J. Phys. Soc. Jpn. **57**, 1891 (1988).

[10] D. V. Dmitriev and V. Ya. Krivnov, Phys. Rev. B **73**, 024402 (2006).

[11] F. Heidrich-Meisner, A. Honecker, and T. Vekua, Phys. Rev. B **74**, 020403(R) (2006).

[12] R. O. Kuzian and S. L. Drechsler, Phys. Rev. B **75**, 024401 (2007).

[13] D. C. Cabra, A. Honecker, and P. Pujol, Eur. Phys. J. B **13**, 55 (2000).

[14] A. A. Aligia, Phys. Rev. B **63**, 014402 (2000).

[15] The value of $J_1 = -3.86 J_2$ for the lower edge of the four-magnon region is a lower bound. At $J_1 < -3.86 J_2$ a loosely bound incommensurate four-magnon state has lower energy than the commensurate bound state. It is quite possible that five-magnon bound states with $k=\pi$ become the lowest excitation at a slightly larger value, $J_1/J_2 \approx -3.8$.

[16] The assumption of a mutual repulsive interaction is justified by the absence of a metamagnetic transition below the saturation field and by the $\Delta S = n$ jumps in the magnetization curve (Refs. 11 and 13). For the repulsive interaction the TL parameter K is less than 1.

[17] The exponent η is related to K as $\eta = 2K$. Reference 4 obtained $\eta = 1.1$ for $J_1 = -J_2$ $(n=2)$ and $\langle S^z \rangle = 3/8$.