Multi-magnon bound states in the frustrated ferromagnetic 1D chain

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We study a one-dimensional Heisenberg chain with competing ferromagnetic nearest-neighbor and antiferromagnetic next-nearest neighbor interactions in 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 multi-magnon states with a total momentum of \( \pi \) except for a small parameter range. We argue that Bose condensation of the bound \( n \) magnons leads to novel Tomonaga-Luttinger liquids with multi-polar 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,

\[
H = \sum_{l \in \mathbb{Z}} \left( J_1 \vec{S}_l \cdot \vec{S}_{l+1} + J_2 \vec{S}_l \cdot \vec{S}_{l+2} - h S^z_l \right),
\]

where \( \vec{S}_l = (S^x_l, S^y_l, S^z_l) \) is the spin-1/2 operator on the \( l \)th site, and external field \( h \) is applied in the \( z \) direction. The exchange interactions are frustrated as the NN interaction is antiferromagnetic (AF), \( J_2 > 0 \). Until recently most theoretical studies on 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, \( \text{Rb}_2\text{Cu}_2\text{Mo}_3\text{O}_{12} \) (Ref. \(^{[1]} \)) and \( \text{LiCuVO}_4 \) (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 \(^{[11,12]} \) 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. Interestingly, such a nematic ordered phase can also appear in a frustrated FM spin model on the two-dimensional square lattice. One can imagine further a Bose condensed phase of more-than-two-magnon bound states. Indeed it was shown recently that an octupolar-like 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 3-magnon bound states, and moreover 4-magnon bound states appear for a slightly stronger FM coupling regime. We then suggest exotic TL liquid phases with multipolar-like spin correlations to emerge from condensation of multi-magnon 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 \(^{[1]} \) 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 \( \phi = \phi_{l+1} - \phi_l = \pm \arccos (-J_1^2J_2) \) and a canting angle of \( \theta = \arccos [4hJ_2/(S(J_1 + 4J_2)^2/4J_2] \), 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. For the parameter range \(-4J_2 < J_1 < 0 \) of our main interest, Chubukov \(^{[4]} \) 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 \( \pi \) if \(-2.67 < J_1 < 0 \) \((2.67 \approx 1/0.38) \) 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 \( J_1/J_2 = -2.66908 \) \((= -1/0.374661) \). While earlier calculations of the ground-state magnetization process suggested metamagnetic transitions \(^{[11,13]} \) recent density-matrix renormalization group (DMRG) study \(^{[14]} \) 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.

To reveal how the fully polarized FM state collapses into new states with decreasing either 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...
the unique ground state for \(-4J_2 < J_1 < 0\), and all multi-magnon 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 \(k\),

\[
|k\rangle = \frac{1}{\sqrt{N}} \sum_{l=1}^{N} e^{ikl} S^+_l |\text{FM}\rangle ,
\]

on the fully polarized state \(|\text{FM}\rangle = |\uparrow\uparrow\uparrow\ldots\rangle\) has the excitation energy

\[
\epsilon_1(k) = J_1(\cos k - 1) + J_2[\cos(2k) - 1] + h ,
\]

which has a minimum of

\[
\epsilon_1(k_0) = -\frac{1}{8J_2}(J_1 + 4J_2)^2 + h
\]
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, 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 (CM) momentum \(k\) as a basis. For example, our basis for the two-magnon excitations has two \(\downarrow\) spins with total momentum \(k\) and a relative distance \(r = (1, 2, \ldots)\),

\[
|r; k\rangle = \frac{1}{\sqrt{N}} \sum_n e^{i(k(2l+r)/2)} S^+_l S^+_r |\text{FM}\rangle .
\]

In this basis the matrix elements of the Hamiltonian can be written as \(<r; k|H|r'; k'>\delta_{k,k'} H_{r,r'}\), where non-vanishing 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 .
\]

By separating off the CM 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 roots of a transcendental equation. The eigenvalue problem is greatly simplified at \(k = \pi\), where the excitation energy of the two-magnon bound state is

\[
\epsilon_2(\pi) = -J_1 - 3J_2 + \frac{J_2^2}{J_1 - J_2} + 2h .
\]

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 3-magnon basis is given by

\[
|r_1,r_2; k\rangle = \sum_n \frac{e^{i(k(3l+2r_1+r_2))/3}}{\sqrt{N}} S^+_l S^+_{l+r_1} S^+_{l+r_1+r_2} |\text{FM}\rangle ,
\]

with which the matrix elements \(<r_1,r_2; k|H|r_1',r_2'; k'>\) are easily found. The 4-magnon basis states \(|r_1,r_2,r_3; k\rangle\) are constructed similarly. To solve the 3- and 4-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 \(1 \leq r_1 \leq 27\) and \(|r_1,r_2,r_3; k\rangle\) with \(1 \leq r_1 \leq 9\), respectively. This seems to be sufficient to determine the lowest excitations.

The four panels of Fig. 1 show energy dispersions of multi-magnon 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 are 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. The lowest bound two-magnon excitation has momentum \(k = \pi\) for \(-2.67 < J_1/J_2 < 0\) and incommensurate momentum \(k < \pi\) for \(-4 < J_1/J_2 \leq -2.67\). 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 and those of bound states of up to \(n - 1\) magnons. For example, energies of 3-magnon scattering states are obtained by adding those of either three unbound magnons or of a 2-magnon bound state plus a free magnon, under the condition that the total momentum is \(k \ (\text{mod } 2\pi)\).

We see in Fig. 1 that for any \(J_1/J_2\) there is always a region in the \(k\) space where bound states lie well below the scattering continuum. The character of the lowest-lying bound states signalling the instability of the fully polarized state changes with \(J_1/J_2\). Unlike 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. \((1 - 2.72 < J_1/J_2 < -2.67)\), a three-magnon commensurate instability \((-3.52 < J_1/J_2 < -2.72)\), and a four-magnon commensurate instability \((-3.86 < J_1/J_2 < -3.52)\). 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 2-magnon sector is the bound state with \(k = \pi\). The 4-magnon scattering states of two such two-magnon bound states therefore have lowest energy (which vanish when \(h = h_a\)) at \(k = 0\). The fact that we do not have a mode of 4-magnon bound states near \(k = 0\) below the continuum indicates that the interaction between two 2-magnon bound states of \(k = \pi\) is repulsive, and therefore these 2-magnon bound states are stable. At \(J_1 = -3J_2\) we have the 3-magnon bound state with \(k = \pi\) as the lowest-energy excitation. The presence of stable 3- and 4-magnon bound states provides natural explanation for the \(\Delta S^z \geq 2\) 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 multi-magnon bound states. The calculated saturation field is in perfect agreement with the exact \(h_s\) estimated from Eq. (7) for \(-2.67 \leq J_1/J_2 < 0\). For \(-3.86 < J_1/J_2 < -2.72\) the saturation field is determined from the instability of 3- or more-magnon bound states.

In Fig. 3 we show the expectation value \((\sum_{i=1}^{n-1} r_i)\) 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 multi-magnon 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 multipolar-like 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 \(h \rightarrow h_s + 0\); \(n = 2\) 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 dilute gas of repulsively interacting bosons which represent the \(n\)-magnon bound states.\[16\] Here the boson creation
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 multi-magnon excitation.

FIG. 3: (Color online) Mean length $\langle \sum_i r_i \rangle$ of the bound multi-magnon 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 $\langle \sum_i r_i \rangle$ is larger) and commensurate ($k = \pi$).

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. It is our pleasure to acknowledge stimulating discussions with Philippe Sindzingre and Nic Shannon. This work was in part supported by Japan Society for Promotion of Science (Grant No. P06902) and by Grants-in-Aid for Scientific Research from MEXT (Grant No. 16GS0219 and No. 17071011).

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The value of $J_1 = -3.86J_2$ for the lower edge of the 4-magnon region is a lower bound. At $J_1 < -3.86J_2$ a loosely bound incommensurate 4-magnon state has lower energy than the commensurate bound state. It is quite possible that 5-magnon bound states with $k = \pi$ becomes lowest excitation at a slightly larger value, $J_1/J_2 \approx -3.8$. Assumption of 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. For the repulsive interaction the TL parameter $K$ is less than 1.

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

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