Alternating-spin $S = \frac{3}{2}$ and $\sigma = \frac{1}{2}$ Heisenberg chain with three-body exchange interactions

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Abstract. The promotion of collinear classical spin configurations as well as the enhanced tendency towards nearest-neighbor clustering of the quantum spins are typical features of the frustrating isotropic three-body exchange interactions in Heisenberg spin systems. Based on numerical density-matrix renormalization group calculations, we demonstrate that these extra interactions in the Heisenberg chain constructed from alternating spin $S = 3/2$ and $\sigma = 1/2$ site spins can generate numerous specific quantum spin states, including some partially-polarized ferrimagnetic states as well as a doubly-degenerate non-magnetic gapped phase. In the non-magnetic region of the phase diagram, the model describes a crossover between the spin-1 and spin-2 Haldane-type states.

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1 Introduction

The biquadratic spin-spin interactions $(S_i \cdot S_j)^2$ and the three-body exchange couplings $(S_i \cdot S_j)(S_k \cdot S_l) + h.c.$ ($|S_i| > \frac{1}{2}$, $i \neq j, k; j \neq k$) naturally appear in the fourth order of the strong-coupling expansion of the two-orbital Hubbard model [1]. Since in this case both types of couplings are controlled by one and the same model parameter – which is about two orders of magnitude weaker than the principal Heisenberg coupling – it might be a challenge to identify experimentally accessible systems where the effects of higher-order interactions can be definitely isolated. Unlike the biquadratic exchange couplings [2], so far there is no clear evidence for effects in real systems related to three-body exchange interactions, although possible three-body exchange effects in some magnetic molecules [3][4] and in the spin-$\frac{3}{2}$ Heisenberg chain CsMn$_2$Mg$_{1-x}$Br$_3$ [5] have been discussed.

On the theoretical side, only recently some specific features of the three-body exchange interaction in Heisenberg spin models in space dimensions $D=1$ [6][7][8] and $D=2$ [9][10][11][12] have been discussed in the literature. In particular, two of us (N.B.I and J.S) recently analyzed the full quantum phase diagram of the alternating-spin Heisenberg chain [7] defined by the Hamiltonian

$$H = J_1 \sum_{n=1}^{L} (S_{2n} \cdot (\sigma_{2n-1} + \sigma_{2n+1})) + J_2 \sum_{n=1}^{L} [(S_{2n} \cdot \sigma_{2n-1})(S_{2n} \cdot \sigma_{2n+1}) + h.c.]$$

in the extremely quantum case of on-site spins $S = 1$ and $\sigma = \frac{1}{2}$. Here $J_1 = \cos \theta$, $J_2 = \sin \theta$ ($0 \leq \theta < 2\pi$), and $L$ denotes the number of unit cells containing two different spins ($S > \sigma$). The model provides a simple, but realistic, example of a Heisenberg system with three-body exchange interactions. For the class of models with $\sigma = \frac{1}{2}$ the biquadratic terms $(\sigma_i \cdot S_i)^2$ reduce to bilinear Heisenberg spin-spin interactions, so that Eq. (1) represents already the general form of the alternating-spin Heisenberg chain with higher-order isotropic exchange interactions.

In this article, we analyze the quantum phase diagram of the above model for the pair of local spins $S = \frac{3}{2}$ and $\sigma = \frac{1}{2}$. Our motivation for this work follows from a previously established tendency towards formation of composite spins from the local $S$ and $\sigma$ spins in the unit cell as an effect of the three-body exchange interactions in the region $\frac{\pi}{2} < \theta < \frac{3\pi}{4}$ of the classical phase diagram, which is characterized by a macroscopic $(2L)$ degeneracy of the ground state (GS) [7]. Therefore, one may expect completely different phase diagrams for systems with integer
and half-integer total spin \((S + \sigma)\) in the unit cell, especially in the highly degenerate classical region. Based on density-matrix renormalization group (DMRG) simulations, in the next Section we analyze the quantum phase diagram of the model \((1)\) with \(S = \frac{1}{2}\) and \(\sigma = \frac{1}{2}\) and discuss different properties of the phases appearing in the interval \(0 < \theta < \pi\). The last Section contains a summary of the results.

2 Quantum phase diagram

![Figure 1](image-url)

**Fig. 1.** (Color online) Quantum phase diagram of the model \((1)\) for \(S = \frac{1}{2}\) and \(\sigma = \frac{1}{2}\) in the interval \(0 < \theta < \pi\). The regions \(\theta < \theta_1\) and \(\theta > \theta_F\) are occupied, respectively, by the Néel ferrimagnetic (FiM) and ferromagnetic (FM) phases, whereas the intervals \(\theta_1 < \theta < \theta_2\) and \(\theta_2 < \theta < \theta_F\) are occupied by different types of partially-polarized magnetic states. A large parameter region, \(\theta_2 < \theta < \theta_1\), is occupied by a non-magnetic doubly-degenerate gapped phase (SL). The FM point \(\theta_F = \pi - \arctan(\frac{1}{2}) = 153.43^\circ\) is an exact boundary of the FM state, \(\theta_1 = 20.1^\circ\), \(\theta_2 = 25.5^\circ\), and \(\theta_3 = 132^\circ\).

The general structure of the phase diagram, as well as the accepted abbreviations for the phases, are presented in Figure 1. Most of the results in this section are obtained through DMRG simulations by performing seven sweeps and keeping up to 500 states in the last sweep [10,11,12]. This ensures a good convergence with a discarded weight of the order of \(10^{-8}\) or better. The numerical DMRG analysis of the lowest energy eigenvalues \(E(M)\) in sectors with a fixed \(z\) component of the total spin \(M\) imply (i) a doubly-degenerate non-magnetic gapped ground state (GS) in the interval \(\theta_2 < \theta < \theta_1\) and (ii) a number of specific partially-polarized magnetic states in the intervals \(\theta_1 < \theta < \theta_2\) and \(\theta_2 < \theta < \theta_F\). Many features of the phase diagram in Figure 1 are also encoded in the behavior of the short-range correlations (SRC) for open boundary conditions (OBC) (see Figure 2). In particular, most of the phase boundary points in Figure 1 can be associated with pronounced rearrangements of the SRC. As in the previously studied extreme quantum case of Eq. (1) with \(S = 1\) and \(\sigma = \frac{1}{2}\) [14], the basic rearrangements concern the SRC between the larger \(S\) spins, whereas - apart from the region close to the FM point \(\theta_F\) - the SRC between the \(\sigma = \frac{1}{2}\) spins remain almost constant [2]. The tendency towards spin clustering is revealed by different values of the spin-spin correlators \(C_L \equiv \langle \sigma_{2n-1} \cdot \sigma_{2n} \rangle\) and \(C_R \equiv \langle S_{2n} \cdot \sigma_{2n+1} \rangle\) in the SL state (see Figure 2).

1 The equation for the exact FM boundary \(\theta_F\) for arbitrary spins \(S\) and \(\sigma\) reads \(\cos \theta_F + \sigma(2S + 1) \sin \theta_F = 0\) [7].

2.1 Partially-polarized magnetic states

The established partially-polarized magnetic states in the intervals \(\theta_1 < \theta < \theta_2\) and \(\theta_2 < \theta < \theta_F\) do not appear in the classical phase diagram. The critical FiM phase in the first interval is identical to the partially-polarized phase discussed for the extreme quantum case \((S, \sigma) = (1, \frac{1}{2})\) [7]: It is characterized by a monotonically decreasing magnetization from \(m_0 = (S - \sigma) = 1\) at \(\theta = \theta_1\) down to \(m_0 = 0\) at the phase boundary \(\theta_2\) with the non-magnetic phase. At the phase boundary \(\theta_1\) the gap of the AFM branch of excitations \(\Delta_A = E(M_0 + 1) - E(M_0)\) vanishes and the system becomes critical. Here \(M_0 = (S - \sigma)L\) corresponds to the GS of the Lieb-Mattis FiM. Unlike the extreme quantum case, where the phase boundary \(\theta_2\) marks the transition to a gapless critical phase, here \(\theta_2\) is related with the vanishing of the triplet gap \(\Delta_T\) of the non-magnetic phase SL. Skipping further discussions on this interesting FiM critical state, we only mention that similar partially-polarized (non-Lieb-Mattis-type) FiM phases have been identified and studied in other spin models, as well [15,16,17].

Now, let us turn to the magnetic states stabilized in the interval \(\theta_2 < \theta < \theta_F\) close to the FM point \(\theta_F\). The exact phase boundary \(\theta_F\) coincides with one of the instability points of the one-magnon FM excitations and is characterized by a complete softening of the dispersion function in the whole Brillouin zone. As a result, one observes a strong reconstruction of the FM state for smaller values of \(\theta\). As a matter of fact, for \(\theta < \theta_F\) we observe a behavior of the SRC which is similar to one in the extreme quantum system (see Figure 4b in Ref. [7]). For this reason, we shall restrict our discussion mainly to the region which is extremely close to the FM point \(\theta_F\), as it is natural to expect that the formation of specific plateau states depends on the values of the local spins: According to the general rule, the number of unit cells in the periodic structure \(q\)
and the magnetic moment per unit cell \( m_0 \) of the plateau states fulfill the equation \( q(S + \sigma - m_0) = \text{integer} \) [18].

In Figure 3 we show DMRG results for some local magnetic moments related to the \( S \) spins at \( \theta = 153.4^\circ \), i.e., extremely close to the exact FM boundary \( \theta_F \). The results clearly indicate a periodic magnetic structure with a period of three unit cells. As required for a plateau state, the established magnetization at this point, \( m_0 = \frac{5}{7} \), fulfills the mentioned general rule with \( q = 3 \). The DMRG results for \( \Delta_A \) at \( \theta = 153.4^\circ \) shown in Figure 4 give further support for the suggested plateau state since the gap is very small but definitely non-zero. Unfortunately, due to strong finite-size effects, it is difficult to decide if the indicated state is realized only at \( \theta = \theta_F \), or in a small interval close to this point. Further, as in the extreme quantum case, the nearest-neighbor spin-spin correlator \( C_2 \) remains positive and signals a FM ordering of the spin-\( S \) subsystem in the entire interval \( \theta_1 < \theta < \theta_F \). The transition to a non-magnetic state is accompanied by an abrupt change of the sign of the correlator \( C_S \). Approaching the transition point \( \theta_3 \), the boundary effects in open chains become stronger, so that by using DMRG simulations it is difficult to study the vicinity of \( \theta_3 \) and to fix more precisely its position.

### 2.2 The non-magnetic SL phase

The numerical results presented in Figure 3 show that for OBC the non-magnetic phase (SL) occupying the interval \( \theta_2 < \theta < \theta_3 \) is characterized by different nearest-neighbor spin-spin correlations, \( C_L \neq C_R \). Excluding some vicinity of the phase boundary \( \theta_3 \), the numerical estimates for \( C_L \) are located near the eigenvalue \(-\frac{1}{4} \) of the operator \( \sigma_{2n-1} \).

Fig. 3. (Color online) On-site magnetizations \( M_k = \langle S_k^z \rangle \) \((k = 2n, 2n+2, 2n+4)\) and \( M_n^{\text{total}} = (M_{2n} + M_{2n+2} + M_{2n+4})/3 \) as functions of the cell index \( n \) (DMRG, \( \theta = 153.4^\circ \), \( L = 144 \), OBC). The results indicate a periodic three-cell \((q = 3)\) magnetic structure close to the FM transition point \( \theta_F \). The inset shows the magnetic supercell containing six spins \((i.e., \text{three unit cells})\). The total magnetization \( M_n^{\text{total}} \) in the supercell is constant.

Fig. 4. (Color online) Finite-size scaling of the AFM gap \( \Delta_A = E(M_0 + 1) - E(M_0) \) above the plateau state with magnetization \( m_0 = M_0/L = \frac{5}{7} \) (DMRG, OBC).

Here \( |\alpha_n\rangle \) \((\alpha_n = 0, \pm)\) are the canonical basis states of the composite-spin operator \( S_{2n} + \sigma_{2n-1} \) in the spin-1 subspace. In terms of the Ising states \( |S_{2n}^z, \sigma_{2n-1}^z\rangle \) the basis states \( |\alpha_n\rangle \) read

\[
|0\rangle_n = \frac{1}{\sqrt{2}} \left( |\frac{1}{2}, -\frac{1}{2}\rangle - |\frac{1}{2}, \frac{1}{2}\rangle \right)
|\pm\rangle_n = \frac{1}{\sqrt{2}} \left( |\frac{1}{2}, \frac{3}{2}\rangle \pm |\frac{1}{2}, -\frac{3}{2}\rangle \pm |\frac{1}{2}, \frac{1}{2}\rangle \pm |\frac{1}{2}, -\frac{1}{2}\rangle \right),
\]

where for simplicity we have omitted the cell index \( n \) on the right-hand side of the equations.

Calculating the matrix elements of the operators \( S_{2n} \) and \( \sigma_{2n-1} \) in the basis (2), one obtains

\[
Q_n^{S_{2n}}Q_n = \frac{5}{4}S_n^z, \quad Q_n^{\sigma_{2n-1}}Q_n = -\frac{1}{4}S_n^z,
\]

where the effective spin-1 operators \( S' \) are defined as follows: \( S'^z = |+\rangle\langle +| - |\cdot\rangle\langle \cdot| - |\cdot\rangle\langle +| + |+\rangle\langle \cdot| \), and \( S'^- = \left(S'^+\right)^\dagger \) for each unit cell. Finally, a substitution of Eqs. 3 in the expression for \( \mathcal{H}_{\text{eff}} \) leads to the following effective Hamiltonian

\[
\mathcal{H}_{\text{eff}} = \frac{5}{4}J_1 L + J_{\text{eff}} \sum_{n=1}^L S_n^z \cdot S_{n+1}^z,
\]
The dimerization effect of the three-body interaction in the whole interval $\theta_2 < \theta < \theta_1$ can be approximately studied by a simple decoupling of the three-body terms in the original Hamiltonian (1):

$$(S_{2n} \cdot \sigma_{2n-1})(S_{2n} \cdot \sigma_{2n+1}) + \text{h.c.}$$

$= 2C_L (S_{2n} \cdot \sigma_{2n+1}) + 2C_R (S_{2n} \cdot \sigma_{2n-1}) - 2C_L C_R.$

Substituting the above expression in Eq. (1), we obtain the following "mean-field" spin Hamiltonian with alternating FM-AFM exchange bonds

$$\mathcal{H}_{\text{MF}} = \sum_{n=1}^{L} [J_{AF}(S_{2n} \cdot \sigma_{2n+1}) + J_F(S_{2n} \cdot \sigma_{2n+1})] - E_0,$$  

where $J_{AF} = J_1 + 2C_R J_2$, $J_F = J_1 + 2C_L J_2$, and $E_0 = 2L J_2 C_L C_R$. Note that the decoupling procedure violates the translational symmetry of the original Hamiltonian (1). Since the unit cell in Eq. (5) is doubled, there is a pair of such Hamiltonians (connected by the symmetry transformation $J_F \leftrightarrow J_{AF}$) related to both types of dimerization functions ($\Psi_{L,R}$) introduced above. The decoupling procedure can be roughly justified by noting that almost in the whole non-magnetic interval the values of $C_L$ are close to the eigenvalue $-2/3$ of the operator $S_{2n} \cdot \sigma_{2n-1}$ (see Figure 2). This approximately implies spin-1 states in the unit cells for each $n = 1, \cdots, L$. In the spin-1 subspace, the matrix elements of the thee-body term in Eq. (1) coincide with the matrix elements of the Heisenberg term $-\frac{J}{3} J_2 \sum_{n=1}^{L} S_{2n} \cdot \sigma_{2n+1}$, so that the basic operator structure of Eq. (5) can be reproduced.

In approaching the phase boundary $\theta_2$, the coupling constant $J_F$ goes to zero (see Figure 6), so that in this case the decoupled-dimer limit becomes a valid approximation. Up to first order in $|J_F|/J_{AF}$, the Hamiltonian $\mathcal{H}_{\text{MF}}$ is equivalent to the projected spin-1 Hamiltonian (1) with $J_{eff} = \frac{3}{4} J_F (J_{eff} > 0)$. The obtained phase boundary (now defined as $J_F = 0$) surprisingly well reproduces the numerical estimate $\theta_2 = 25.5^\circ$. As far as the parameter $|J_F|$ increases with $\theta$, it seems relevant to evaluate the effect of the second-order perturbation in $|J_F|/J_{AF}$, as well. However, such a perturbation does not lead to any qualitative changes of the GS because its effect is restricted to a small renormalization of $J_{eff}$ and to appearance of an irrelevant (FM) next-nearest-neighbor Heisenberg term in Eq. (5). Actually, for larger $|J_F|$ it is instructive to analyze the other decoupled-dimer limit of Eq. (6) based on non-interacting (FM) spin-2 dimers and using the small parameter $J_{AF}/|J_F| \ll 1$. Up to first order in $J_{AF}/|J_F|$, this gives Eq. (6), but now with the AFM coupling $J_{eff} = 3J_{AF}$ and the effective spin-2 operators $\tilde{S}_n$. In terms of VBS states, the formation of local spin-2 states corresponds to an additional symmetrization of the cell spins, as shown in Figure 5(c,d), without any abrupt change in the topological structure of the singlet bonds. Therefore, it may be speculated that the transition between both dimer limits is realized through a smooth crossover between both Haldane-type gapped states.

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2 The alternating-bond FM-AFM Heisenberg chain (5), describing a smooth transition between the spin-1 and spin-2
In Figure 7 we present numerical results for the triplet energy gap $\Delta_T$ in the discussed parameter region. The growth of the gap approximately up to $\theta \approx 45^\circ$ can be related with the established increase of the effective exchange constant $J_{\text{eff}}$ in Eq. (4). In accord with the suggested VBS state in Figure 5(a), for OBC one observes the expected structure of the lowest excited states including a singlet GS, which is degenerate with the Kennedy edge triplet in the thermodynamic limit [19]. The first bulk excitation, related to the Haldane gap, appears as a spin-2 (quintet) state resulting from the combination of the bulk and Kennedy’s edge triplets. On the other hand, for $\theta > 45^\circ$ the structure of the lowest excited states becomes very complicated due to the presence of many parasitic edge excitations. Namely, as demonstrated in Figure 5(b,c,d), the number of free edge spins and their values depend on (i) the type of established VBS states ($|\Psi_L\rangle$ or $|\Psi_R\rangle$) and (ii) the increased tendency (with $\theta$) towards formation of local spin-2 states. For this reason, for larger $\theta$ the gap $\Delta_T$ is presented for periodic chains. Since the increase of $|J_{\text{f}}|$ is restricted to $|J_{\text{f}}| \lesssim 2$, the true spin-2 dimer limit is not reached. Nevertheless, as far as the pure spin-2 phase is characterized by an extremely small energy gap $\Delta = 0.085(5)J$ according to the DMRG result in Ref. [20], it is reasonable to admit that the established decrease of $\Delta_T$ for $\theta \gtrsim 45^\circ$ is connected with a smooth crossover between the spin-1 and the spin-2 Haldane-type non-magnetic states. Finally, due to the extremely small gap $\Delta_T$ and the large number of low-lying energy states, it is difficult to give a precise DMRG estimate for the other phase boundary $\theta_3$ and the properties of the GS close to this boundary.

Haldane phases, deserves a special detailed analysis going beyond the scope of the present study.

3 Summary

We have established the general structure of the quantum phase diagram of the alternating-spin $S = \frac{3}{2}$ and $\sigma = \frac{1}{2}$ Heisenberg chain with extra isotropic three-body exchange interactions. To some extent the established partially-polarized FiM phases resemble the magnetic phases of the extreme quantum chain with alternating spins $S = 1$ and $\sigma = \frac{1}{2}$ [15], apart from the vicinity of the FM point $\theta_F$ where both systems support different plateau states. On the other hand, due to the clustering effect of the three-body interactions, both models support completely different quantum phases in the non-magnetic region of the phase diagram: the critical phase in the $(1,\frac{1}{2})$ model is replaced by a specific doubly-degenerate phase, which can be described as a Haldane-type gapped state predominantly composed of effective cell spins with quantum spin numbers 1 or 2. It may be expected that most of the predicted effects and phases persist in higher space dimensions.

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