Phase diagram of a frustrated mixed-spin ladder with diagonal exchange bonds

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Using exact numerical diagonalization and the conformal field theory approach, we study the effect of magnetic frustrations due to diagonal exchange bonds in a system of two coupled mixed-spin (1, 3/2) Heisenberg chains. It is established that relatively moderate frustrations are able to destroy the ferrimagnetic state and to stabilize the critical spin-liquid phase typical for half-integer-spin antiferromagnetic Heisenberg chains. Both phases are separated by a narrow but finite region occupied by a critical partially-polarized ferromagnetic phase.

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I. INTRODUCTION

Many experiments on bimetallic quasi-one-dimensional (1D) molecular magnets imply that the magnetic properties of these compounds are basically described by the Heisenberg spin model with antiferromagnetic exchange couplings. In the last decade, there has been an increasing experimental and theoretical interest in these mixed-spin systems exhibiting intriguing quantum spin properties of mixed-spin ladders.

In this paper we study the ground-state phase diagram of a ferrimagnetic two-leg ladder containing frustrating antiferromagnetic diagonal exchange bonds (see Fig. 1). The model is defined by the Hamiltonian

\[ \mathcal{H} = \sum_{n=1}^{L} \left[ J_1 (s_{1n} \cdot s_{2n+1} + s_{2n} \cdot s_{1n+1}) + J_\perp s_{1n} \cdot s_{2n} \right] + J_2 \sum_{n=1}^{L} (s_{1n} \cdot s_{1n+1} + s_{2n} \cdot s_{2n+1}) , \]

where \( L \) is the number of rungs and the spin operators \( s_{1n} \) and \( s_{2n} \) are defined on the rung with index \( n \): \( (s_{1n})^2 = s_1(s_1+1), (s_{2n})^2 = s_2(s_2+1), s_1 > s_2, \) and \( J_1, J_2, J_\perp > 0. \)

We introduce the frustration parameter \( \alpha = J_2/J_1 \), the spin ratio \( \sigma = s_1/s_2 \), and set the energy and length scales by \( J_1 \equiv 1 \) and \( a_0 = 1 \), where \( a_0 \) is the spacing between neighboring rungs. In the remainder of this paper, if not especially noted, we will consider the case \( J_\perp = J_1 \).

As a function of the frustration parameter \( \alpha \), the classical phase diagram of Fig. 1 exhibits three phases described by the angles \( (\theta, \phi) \) fixing the directions of the classical spins \( s_{1n} \) and \( s_{2n} \) in respect to the classical ferrimagnetic configuration with spins \( s_{1n} \) and \( s_{2n} \) oriented along the axes \( z_1 \) and \( z_2 \), respectively (Fig. 1). The classical canting state (C) shown in Fig. 1 is stable in the interval \( \alpha_{c1} < \alpha < \alpha_{c2} \) where \( \alpha_{c1} = 3[(\sigma^2 + 1) - \sqrt{(\sigma^2 + 1)^2 - 32\sigma^2/9}] / 8\sigma \) and \( \alpha_{c2} = \left[-(\sigma^2 + 1) + \sqrt{(\sigma^2 + 1)^2 - 32\sigma^2}/8\sigma \right] \). The following analysis of the quantum phase diagram is performed by using exact numerical diagonalization (ED) of small periodic systems, finite-size analysis of the ED data, and analytical spin-wave calculations. The emphasis is on the properties of the quantum paramagnetic phase.

FIG. 1: Sketch of the classical canted state for \( J_\perp = J_1 \) described by the angles \( (\pm \theta, \pm \phi) \) for every magnetic cell composed of two neighboring rungs. \( \theta \) and \( \phi \) measure the deviations of the classical spins from the ferrimagnetic configuration: \( s_{1n} \parallel z_1 \) and \( s_{2n} \parallel z_2 \).
II. MAGNETIC PHASES

As may be expected, the positions of the classical phase transition points \( \alpha_{c1} \) and \( \alpha_{c2} \) are changed by quantum spin fluctuations. Using ED and a simple finite-size scaling, it is possible to find precise estimates for the quantum transition points. The latter are connected with the following changes in the total spin of the ground state, \( S_T := (s_1 - s_2)L \) for \( 0 < \alpha < \alpha_{c1} \) (F phase), \( 0 < S_T < (s_1 - s_2)L \) for \( \alpha_{c1} < \alpha < \alpha_{c2} \) (C phase), and \( S_T = 0 \) for \( \alpha > \alpha_{c2} \) (quantum paramagnetic phase).

The extrapolated data for \( L = 8, 10, 12 \) and 14 give the results \( \alpha_{c1} = 0.341 \) and \( \alpha_{c2} = 0.399 \) showing that the region occupied by the quantum C phase is narrowed but definitely finite. Figure 2 provides a summary of the reported results in terms of the net ferromagnetic moment per rung \( M_0 \) for \( L = 12 \).

![Figure 2: ED results for the ferromagnetic moment per rung \( M_0 \) vs \( J_2/J_1 \) (dashed line). The step-like form of \( M_0(\alpha) \) is connected to finite-size effects. The midpoints of the steps close to \( \alpha_{c2} \) are well approximated by the ansatz \( M_0 = m_1(\alpha_{c2} - \alpha)^q + m_2(\alpha - \alpha_{c2}), \) where \( m_1 = 1.65, m_2 = 0.56, \) and \( \beta = 0.5 \) (solid line).](image)

The ferromagnetic phase has already been studied for the model (1) without frustration. As \( M_0 > 0 \), both magnetic phases (F and C) are characterized by quadratic spin-wave excitations

\[
E(k) = \frac{\rho_s k^2}{M_0} + O(k^4),
\]

where \( \rho_s \) is the ferromagnetic spin-stiffness constant. In approaching \( \alpha_{c1} \) from the ferromagnetic phase, the linear spin-wave theory predicts that the lower spin wave branch softens in the vicinity of \( k = \pi \) and the gap at this point vanishes for \( \alpha > \alpha_{c1} \). Thus the linear Goldstone mode, characteristic of the classical C phase, seems to survive quantum fluctuations although on general grounds it may be expected that the spin rotation symmetry \( U(1) \) in the \( xy \) plane is restored in one space dimension, i.e., \( \langle s_1^{x} \rangle = \langle s_2^{x} \rangle = 0 \). This scenario is supported by the renormalization-group analysis of similar phases in quantum rotor models, implying that the true transverse long-range magnetic order in the classical C phase is transformed to a quasi-long-range \( xy \) order in the quantum system. On the other hand, the spin-stiffness constant \( \rho_s \) remains finite in both magnetic phases as well as at the transition point \( \alpha_{c1} \). Following the terminology of Ref. [9], the quantum C phase may be called partially polarized ferromagnetic phase, as the ferromagnetic moment \( M_0 \) is less than the maximal value \( s_1 - s_2 \) in the ferromagnetic phase (see Fig. 2). This quantum state may also be classified as a kind of ferromagnetic Luttinger liquid. We postpone the detail analysis of this quantum phase for future studies.

III. QUANTUM PARAMAGNETIC PHASE

Now, let us turn to the region \( \alpha > \alpha_{c2} \) of the phase diagram characterized by \( S_T = 0 \). It is instructive to rewrite (1) in the following form

\[
\mathcal{H} = \sum_{n=1}^{L} (J_1 s_n \cdot s_{n+1} + J_L s_{n+1} \cdot s_{2n})
\]

\[
+ (J_2 - J_1) \sum_{n=1}^{L} (s_{n+1} \cdot s_{n+1} + s_{2n} \cdot s_{2n+1}).
\]

The operator \( 2s_{n=1} \cdot s_{2n} = (S_1^2 - s_1)_{s_1 = 1} - s_2_{s_2 = 1} \) is a conserved quantity for \( J_2 = J_1 \) and at this special point the low-energy physics of (1) is described by the antiferromagnetic spin-1 Heisenberg chain: \( (S_1^2 - s_1)_{s_1 = 1} = \frac{1}{2} \). In the case of special interest \( (s_1, s_2) = (1, \frac{1}{2}) \), and for relatively small interchain couplings \( J_1 \leq 1.5 J_1 \) we have numerically found that the lowest excited states are characterized by \( S = \frac{3}{2} \) antiferromagnetic Heisenberg chain.

The above statements concern the special point \( J_2 = J_1 \) where \( S \) is a good quantum number. As an example, in Fig. 3 we show the energies of the lowest excited states \( (L = 12 \text{ and } 14) \) for \( \alpha = 0.4 \) and 1. Apart from the \( k = 0 \) state, which is characterized by the total spin 2, the lowest excited states are triplets above the singlet ground state. This structure of the low-energy spectrum is valid in the whole region \( \alpha > \alpha_{c2} \) up to the limit \( \alpha = \infty \) where the system is composed of two independent spins \( s_1 = 1 \) and \( s_2 = \frac{1}{2} \) antiferromagnetic Heisenberg chains. In accord with the generalized Lieb-Schultz-Mattis (LSM) theorem, it is natural to suppose that the gapless linear structure of the spectrum around \( k = \pi \) survives away from the point \( J_2 = J_1 \).

To study the properties of the quantum paramagnetic phase in the whole region \( \alpha > \alpha_{c2} \), we may compare the finite-size scaling properties of the ground state and the lowest excited states with those based on the SU(2) Wess-Zumino-Witten (WZW) nonlinear \( \sigma \) model. This model with the topological coupling \( k_0 = 1 \) is believed to describe the antiferromagnetic Heisenberg chains with half-integer site spins. In the following we restrict our
an analysis to the \((s_1, s_2) = (1, \frac{1}{2})\) system. According to the conformal field theory, the ground state energy \(E_0(L)\) of a periodic system with length \(L\) is given by the following expression:

\[
E_0 = \sum_{n=1}^{L-1} \left[ \frac{3}{8} g^2 + O(g^4) \right] + \frac{a_L}{L}.
\]

Here \(\varepsilon_0\) is the ground state energy per rung in the thermodynamic limit, \(v_s\) is the spin-wave velocity, and \(g = g(L)\) is the effective coupling constant of the marginally irrelevant operator \(-2\pi g J_L \cdot J_R\) at the length scale \(L\). \(J_L\) and \(J_R\) are the conserved current operators for the left and right movers in the WZW theory. The \(L^{-4}\) contribution comes from irrelevant operators. The coupling \(g\) is defined by the renormalization-group (RG) equation

\[
\frac{1}{g} + \frac{1}{2} \ln g = \frac{L}{L_c},
\]

where \(L_c\) is a non-universal effective length scale depending on the microscopic model. An iterative solution of Eq. (5) yields the following expansion for \(g\)

\[
g = \frac{1}{\ln(L/L_c)} - \frac{\ln \ln(L/L_c)}{2 \ln(2)(L/L_c)} + O \left( \frac{1}{\ln^3(L/L_c)} \right).
\]

so that the marginally irrelevant operator introduces logarithmic corrections in Eq. (4).

The energy of the lowest triplet excitation \(E_t(L)\) with momentum \(k = \pi\) can be expressed in the form

\[
E_t - E_0 = \frac{2\pi v_s}{L^2} \left[ \frac{1}{2} - \frac{g}{4} + b_1 g^2 + O(g^3) \right] + b_2 \frac{L}{L^3}.
\]

At moderate length scales \((L = 8, 10, 12, 14)\), the coupling constants \(g(L)\) in (4) and (7) may have different values, so that instead of \(L_c\) we introduce the effective length scales \(L_0\) and \(L_t\) for the ground state energy and the energy of triplet excitations.

FIG. 3: ED results for the lowest excited states of the system \((s_1, s_2) = (1, \frac{1}{2})\) for two different frustration parameters \((\alpha = 1\) and \(0.4); L = 12\) (open circles), \(L = 14\) (filled circles). Curves represent the two branches of spin-wave modes in the paramagnetic Brillouin zone, as obtained from the linear spin-wave theory. The spin-wave energies are multiplied by the normalization factor \(v_s/\nu_{sw} = 1.29\) where \(v_s = 3.87\) and \(\nu_{sw} = 2\pi = 3\) are the spin-wave velocities, as obtained by the density matrix renormalization group method \(\alpha\) and the linear spin-wave theory. Note that the classical transition point at \(\alpha = 0.4606\) appears as an instability point for the lower spin-wave branch.

FIG. 4: Reduced energy gap between the lowest excited state at \(\pi = 2\pi/L\) and the ground state. The interpolation of ED data is performed by the ansatz \(E(2\pi/L)/2\pi L = \sum_{n=1}^{L-2n} c_n L^{-2n}\). For \(\alpha = 0.45, 0.5, 1\) this yields, respectively, the spin-wave velocities \(v_s = 1.19, 1.78, 3.81\). Note that the interpolation function for \(\alpha = 0.4\) has a positive curvature.

The parameter \(v_s\) in Eqs. (10) and (11) can be independently determined from the scaling of the reduced gap \(E(2\pi/L)/2\pi L\), where \(E(2\pi/L)\) is the energy gap between the lowest excited state with momentum \(\pi = 2\pi/L\) and the ground state. Using the interpolation ansatz \(E(2\pi/L)/2\pi L = \sum_{n=1}^{L-2n} c_n L^{-2n}\), we find, in particular, the estimate \(v_s = c_1 = 3.81\) at \(\alpha = 1\). This is close to the density matrix RG result \(3.87 \pm 0.02\) for the antiferromagnetic \(S = \frac{1}{2}\) Heisenberg chain \(\alpha\). In Fig. 4 we present the interpolation curves for different values of the frustration parameter \(\alpha\). Excluding the point \(\alpha = 0.4\), our estimates for \(v_s\) can be well interpolated by the ansatz

\[
v_s = v_1(\alpha - \alpha_2^*)^\gamma + v_2(\alpha - \alpha_2) + O[(\alpha - \alpha_2^*)^2]
\]

up to \(\alpha = 1\), provided that \(\gamma \approx \frac{1}{2}\). The linear spin-wave theory gives the exponent \(\gamma = \frac{1}{2}\). Note that the spin-wave ansatz \(\alpha\) assumes that the velocity \(v_s\) vanishes at the critical point \(\alpha_2\). Of course, the above interpolation of ED data can not definitely confirm such an assumption, although the apparent change in the curvature of \(E(2\pi/L)/2\pi L\) vs \(1/L^2\) close to \(\alpha = 0.4\) gives some indication in favor of \(v_s(\alpha_2) = 0\).

Having the parameter \(v_s\) for different \(\alpha\), now we can interpolate the ED data for \(E_0(L)\) and \(E_t(L)\) by using the scaling expressions (4) and (7). The fitting parameters
in Eqs. (4) and (5). As an example, in Fig. 5 we present the interpolation curves $E_0(L)/L$ vs $1/L^2$ for frustration parameters $\alpha = 0.45$ and 0.5. Using the RG equation (5) for $g(L)$, the best fit at $\alpha = 1$ is obtained for $\varepsilon_0 = -2.3290$, $L_0 = 0.94$ and $a_1 = -20.5$. This is in accord with the density matrix RG result $\varepsilon_0 = -2.3283$. The parameter $L_0 = 0.94$ corresponds to an effective coupling constant $g(10) = 0.35$. Performing the fits down to $\alpha = 0.4$, we observe that the characteristic length $L_0$ remains almost unchanged (excluding the point $\alpha = 0.4$ where formally $L_0 \to \infty$). An interpolation procedure using only the leading term in the logarithmic expansion (5) produces similar results, although with a slightly larger effective length $L_0 = 1.08$.

In conclusion, we have examined the impact of magnetic frustration on the ground-state phase diagram of two coupled mixed-spin $(s_1, s_2) = (1, 1/2)$ ferrimagnetic Heisenberg chains. The analysis of ED data implies an interesting phase diagram containing the ferrimagnetic phase and a singlet paramagnetic phase exhibiting the characteristics of the critical spin-liquid phase in half-integer-spin antiferromagnetic Heisenberg chains. Both phases are separated by a tiny but finite region occupied by a critical partially-polarized ferromagnetic phase. It is natural to expect similar phase diagrams for the whole class of frustrated $(s_1, s_2)$ two-leg ladders with half-integer rung spins.

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For brevity, we do not present the ED data revealing the anisotropy in the spin-spin correlations in both magnetic phases.

9 See, e.g., L. Bartosch, M. Kollar, and P. Kopietz, Phys. Rev. B 67, 092403 (2003), and references therein.

10 I. Affleck, D. Gepner, H.J. Schultz, and T. Ziman, J. Phys. A: Math. Gen. 22, 511 (1989).

11 Note that the $n$-th unit cell of the model contains only the rung spins $s_{1n}$ and $s_{2n}$ so that for $(s_1, s_2) = (1, \frac{1}{2})$ we can apply the generalized LSM theorem: see, Affleck and E. Lieb, Lett. Math. Phys. 12, 57 (1986).

12 K. Hallberg, X.Q.G. Wang, P. Horsch, and A. Moreo, Phys. Rev. Lett. 76, 4955 (1996).

13 S. Lukyanov, Nucl. Phys. B 522, 533 (1998).

14 J. Solyom, Adv. Phys. 28, 201 (1979).

15 The assumption $v_s(\alpha c^2) = 0$ has also been considered as a general criterion for ferromagnetic instabilities in the critical paramagnetic phase: see, D.C. Cabra and J.E. Drut, J. Phys.: Condens. Matter 15, 1445 (2003), and references therein.

16 T. Ziman and H.J. Schulz, Phys. Rev. Lett. 59, 140 (1987).

17 Respectively, the effective coupling constant $g(L)$ increases from $g(10) = 0.54$ at $\alpha = 1$ up to $g(10) = 0.57$ at $\alpha = 0.42$. 