Magnetic phases of the mixed-spin $J_1 - J_2$ Heisenberg model on a square lattice

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We study the zero-temperature phase diagram and the low-energy excitations of a mixed-spin (\(S_1 > S_2\)) \(J_1 - J_2\) Heisenberg model defined on a square lattice by using a spin-wave analysis, the coupled cluster method, and the Lanczos exact-diagonalization technique. As a function of the frustration parameter \(J_2/J_1\) (> 0), the phase diagram exhibits a quantized ferrimagnetic phase, a canted spin phase, and a mixed-spin collinear phase. The presented results point towards a strong disordering effect of the frustration and quantum spin fluctuations in the vicinity of the classical spin-flop transition. In the extreme quantum system \((S_1, S_2) = (1, \frac{1}{2})\), we find indications of a new quantum spin state in the region \(0.46 < J_2/J_1 < 0.5\).

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I. INTRODUCTION

There has recently been an increasing interest in Heisenberg spin systems exhibiting quantum phases with an extensive magnetic moment. An intriguing example is the bilayer Hall system at filling factor \(\nu = 2\) which was shown to possess a canted spin state with spontaneously broken \(U(1)\) symmetry. A closely related phase diagram was studied in the framework of the bilayer quantum Heisenberg model subject to a perpendicular magnetic field. Some specific properties of the quantum phase transitions in systems with quantum ferromagnetic phases have been previously discussed by using a special class of lattice models with quantum-rotor degrees of freedom. In the present paper we study a \(J_1 - J_2\) Heisenberg model which is a natural (mixed-spin) extension of the well known antiferromagnetic \(J_1 - J_2\) model and exhibits most of the quantum magnetic phases found in the aforementioned rotor systems.

The model is defined by the Hamiltonian

\[
\mathcal{H} = J_1 \sum_{(r,r')} S_{1r} \cdot S_{1r'} + J_2 \sum_{[r,r']} (S_{1r} \cdot S_{1r'} + S_{2r} \cdot S_{2r'}) ,
\]

where \((r,r')\) and \([r,r']\) denote pairs of nearest and next-nearest (diagonal) sites of the square lattice. We choose a checkerboard arrangement for the spin operators \(S_{1r}\) and \(S_{2r}\) \((S_{1r}^2 = S_1(S_1 + 1), S_{2r}^2 = S_2(S_2 + 1), \text{and } S_1 > S_2)\) and introduce the parameters \(\alpha \equiv J_2/J_1\) \((J_1, J_2 \geq 0)\) and \(\sigma \equiv S_1/S_2\). With some minor modifications, e.g., introducing a spatial anisotropy in the nearest-neighbor exchange interaction, the Hamiltonian could describe a large class of real mixed-spin compounds such as the bimetallic molecular magnets.

In the classical limit the phase diagram of the mixed-spin system contains the ferrimagnetic state (F phase), the canted state (C phase), and the mixed-spin collinear state (N phase) shown in Fig. 1. The latter phases are stable, respectively, in the parameter regions \(\alpha < \alpha_c, \alpha_c < \alpha < 0.5, \text{and } \alpha > 0.5\). The classical F-C transition at \(\alpha_c = (2\sigma)^{-1}\) is continuous, whereas the C-N transition at \(\alpha = 0.5\) is connected with a flop of the \(S_1\) and \(S_2\) spins. The canted spin phase appears as a result of the magnetic frustration without explicit breaking of the \(SU(2)\) symmetry (in the mentioned bilayer systems the C phase is generated by a perpendicular magnetic field). Note also that the \(S_2\) spins remain collinear in the C phase. The angle \(\theta\) measuring the local orientation of the classical \(S_1\) spins in respect to the global magnetization axis \(z\) alternatively takes the values

\[
\theta = \pm \arccos \frac{1}{2\alpha\sigma} .
\]

Fig. 1. The classical ferrimagnetic (F), canted (C), and collinear (N) spin phases of the mixed-spin \(J_1 - J_2\) model on a square lattice.
II. PHASE DIAGRAM

A. Spin-wave analysis

To perform a SWT analysis we assume that the classical spins lie in the $x\hat{z}$ plane. Using as a quantization axis the local orientation of the classical spins (the $z'$ axis in Fig. 1), the leading spin-wave terms in the expansions for the spin operators $S_{1r}$ and $S_{2r}$ read

$$S_{1r}^z = \cos \theta_{ir}(S_i - a_{ir}^+ a_{ir}) - \sin \theta_{ir}\sqrt{\frac{S_i}{2}}(a_{ir}^+ + a_{ir}),$$

$$S_{1r}^x = \sin \theta_{ir}(S_i - a_{ir}^+ a_{ir}) + \cos \theta_{ir}\sqrt{\frac{S_i}{2}}(a_{ir}^+ - a_{ir}),$$

$$S_{1r}^y = i\sqrt{\frac{S_i}{2}}(a_{ir}^+ - a_{ir}) \quad (i = 1, 2),$$

where the angle $\theta_{ir}$ measures the local deviations of $z'$ from the magnetization axis $z$. A substitution of the latter expressions in Eq. (1) yields the following spin-wave expressions in Eq. (1) gives the following expression for the on-site magnetizations $m_1 \equiv \langle S_{1r}^z \rangle$ and $m_2 \equiv \langle S_{2r}^z \rangle$:

$$m_i = S_i - \frac{2}{N} \sum_k \left[ |v_{1k}^{(1)}|^2 + |v_{1k}^{(2)}|^2 \right] \quad (i = 1, 2),$$

where

$$v_{1k}^{(1)} = -\frac{\Delta_{12}(\omega_1)}{\sqrt{2\omega_1(\omega_1^2 - \omega_1^2)\Delta_{11}(\omega_1)}},$$

$$v_{1k}^{(2)} = -\frac{\Delta_{14}(\omega_1)}{\sqrt{2\omega_1(\omega_1^2 - \omega_1^2)\Delta_{11}(\omega_1)}},$$

$$v_{2k}^{(1)} = -\frac{\Delta_{13}(\omega_2)}{\sqrt{2\omega_2(\omega_2^2 - \omega_1^2)\Delta_{11}(\omega_2)}},$$

$$v_{2k}^{(2)} = -\frac{\Delta_{13}(\omega_2)}{\sqrt{2\omega_2(\omega_2^2 - \omega_1^2)\Delta_{11}(\omega_2)}}.$$  

In the above expressions, $\Delta_{ij}(\omega)$ is the minor of the $ij$ element of $\Delta(\omega)$ and $N$ is the number of sites of the lattice.

FIG. 2. The on-site magnetizations $m_1 = \langle S_{1r}^z \rangle$ and $m_2 = \langle S_{2r}^z \rangle$ plotted as a function of $J_2/J_1$: SWT (full lines), CCM (filled circles), ED for $N = 20$ spins and periodic boundary conditions (open circles).
B. Magnetic phases

The spin-wave results for the on-site magnetizations \( m_1 \) and \( m_2 \), Eq. (1), in the extreme quantum system \((1, \frac{1}{2})\) are presented in Fig. 3. We suppose that in the ferrimagnetic phase the sublattice magnetizations are oriented along the global \( z \) axis, i.e., \( m_1 = \langle S_1^z \rangle \) and \( m_2 = -\langle S_2^z \rangle \). The F phase is also characterized by the net ferromagnetic moment per cell \( M_0 = (S_1 - S_2) = \frac{1}{2} \) (see Fig. 3). In the general case it is straightforward to observe that \( M_0 \) takes only integer or half-integer values in the F phase. Following the authors of Ref. 3, this state may be refereed to as a quantized unsaturated ferromagnetic phase. The ED data from Figs. 2 and 3 show that the position of the continuous phase transition \( \alpha_{c1} \) is slightly changed by quantum fluctuations towards larger \( \alpha \). \( \alpha_{c1} \) is fixed as a point where the spin of the ground state changes from \( (S_1 - S_2)/2 \) to \( (S_1 - S_2)/2 - 1 \). Respectively, in the \( N = 20 \) system with \( (S_1, S_2) = (1, \frac{1}{2}) \) the magnetization per cell changes from \( M_0 = 0.5 \) to \( M_0 = 0.4 \) (Fig. 3).

![FIG. 3. Order parameters in the F and C phases as obtained from the SWT (thick lines) and the ED method for \( N = 20 \) with periodic boundary conditions (open circles). The thin broken line shows the ED data for the magnetization per cell \( M_0 \) (\( N = 20 \)). The classical values of the parameters are shown by dotted lines.](image)

More important changes take place in the C phase. The canted spin state is additionally characterized by the staggered transverse field \( \langle S_1^x \rangle \) which breaks the spin rotation symmetry \( U(1) \) in the \( xy \) plane. In Fig. 4 we present result for the transverse order parameter as obtained from the SWT and the ED method. The ED data are obtained from the ED results for \( \langle S_1^z \rangle \), by using the relation \( \langle S_1^x \rangle = \langle S_1^z \rangle \tan \theta \). The pitch angle \( \theta \) is extracted from the spin-spin correlation function (see Fig. 3).

\[
K_{S_1} = \frac{\langle S_{10} \cdot S_{1e1+e2} \rangle}{\langle S_{10} \cdot S_{12e2} \rangle}
\]  

(9)

which reduces to \( K_{S_1} = \cos 2\theta \) in the classical limit \( S_1 \to \infty \). The respective correlation function \( K_{S_2} \) for the \( S_2 \) subsystem can be defined in a similar way. Figs. 2 and 3 show that in the C phase the combined disordering effect of the magnetic frustration and quantum spin fluctuations is strongly enhanced as compared to the F phase. SWT predicts a complete magnetic disordering of the \( S_2 \) spins \( (m_2 = 0) \) starting at \( \alpha = \alpha^* \). The latter point exists in the SWT for arbitrary spins \( S_1 \) and \( S_2 \) and precedes the classical spin-flop transition point \( (\alpha = 0.5) \). The effect is strongly pronounced in the extreme quantum case \((1, \frac{1}{2})\) where both the SWT and the ED data predict \( \alpha^* \approx 0.46 \). The extrapolated CCM data seem to show the same tendency. The steps in the ED data are due to finite-size effects and reflect the change in the total spin \( S_t \) (a good quantum number) of the absolute ground state with the frustration parameter \( J_2/J_1 \). For instance, in the ED data \( \alpha^* \) appears as a point where \( S_t \) changes from \( S_t = 1 \) to \( S_t = 0 \) (respectively, \( M_0 \) changes from 0.1 to 0). Although SWT formally predicts \( m_1 > 0 \) even beyond \( \alpha^* \), the latter region is not accessible (for the standard SWT) as the phase with \( m_1 > 0 \) and \( m_2 = 0 \) does not appear in the classical phase diagram.[5] We suggest that the point \( \alpha^* \) is just the new position of the quantum spin-flop transition (see below). It is interesting to note that quantum fluctuations in the C phase do not change substantially the parameters \( M_0 \) and \( \theta \) (see Figs. 3 and 4).

Finally, the mixed-spin collinear state has properties which are similar to those of the collinear phase in the antiferromagnetic \( J_1 - J_2 \) model, i.e., it is a Néel-type magnetic state characterized by the on-site magnetizations \( m_1, m_2 \neq 0 \). Both the SWT and the CCM predict that the on-site magnetization \( m_2 \) vanishes at a point.
(αc2) which is very close to the classical spin-flop transition point.

\[ (S_1, S_2) = (1, 1) \]

FIG. 5. Spin-spin correlators vs. \( J_2/J_1 \) in the system with \( N = 20 \) spins (ED results). The presented functions are scaled by their absolute values at \( J_2 = 0 \).

The ED data for different spin-spin correlations (Fig. 5) give some additional evidence in favor of the suggested phase diagram. Here the phase transition point \( \alpha_{c2} \) can be indicated as a point where the mixed-spin correlations (being ferromagnetic in the N phase) change their sign. We observe that close to the point \( \alpha_{c2} \) the spin-spin correlations among the \( S_2 \) spins are small and also change their sign. On the other hand, the correlations among the \( S_1 \) spins remain relatively large (and antiferromagnetic, as in the N phase) down to the point \( \alpha^* \approx 0.46 \), where the singlet ground state disappears. Thus, there are some indications that the established quantum spin phase (\( \alpha^* < \alpha < \alpha_{c2} \)) has the symmetries of the canonical Néel state composed entirely of \( S_1 \) spins (i.e., \( m_1 \neq 0, m_2 = 0 \)). However, due to the finite-size effects in the ED data we can not exclude the spin-liquid state (\( m_1 = 0, m_2 = 0 \)) as a possible ground state in the above region. Further understanding of the proposed phase diagram may be achieved by studying the low-lying excitations in these magnetic phases.

C. Low-energy excitations

In the F phase \( B_{2k} = B_{2k} = D_k = 0 \) and the dispersion relations in Eq. (3) are simplified to

\[ \omega_1^{(F)} = \frac{\rho_s^{(F)}}{m_0} k^2 + O(k^4) , \]

where \( \rho_s^{(F)} = J_1 S_1 S_2 - J_2 (S_1^2 + S_2^2) \) is the spin-stiffness constant of the ferrimagnetic state and \( m_0 = (S_1 - S_2)/a^2 \) \((a = \sqrt{2})\) is the magnetization density. \( \rho_s^{(F)} \) remains finite at the F-C transition point \( \alpha_{c1} \). The F-C phase transition is connected with a softening of the acoustic branch \( \omega_1^{(F)} \) at the corners of the PBZ. For instance, close to \( k = (0, \pi) \) the magnon spectrum reads

\[ \omega_1^{(F)} = 8 J_1 S_1 (\alpha_{c1} - \alpha) + 4 J_1 S_1 [k_1^2 + (\pi - k_2)^2] . \]

On the other hand, the optical branch \( \omega_2^{(F)} \) remains stable at \( \alpha_{c1} \) and does not play important role in the quantum F-C phase transition.

FIG. 6. Spin-wave spectrum in the C phase along the \( e_2 \) direction of the PBZ \((k_1 = 0, J_1 \equiv 1)\).

The excitation spectrum in the C phase is described by the general expression (F), where \( A_{1k} = 4 J_1 S_2 t + 4 J_2 S_1 [1 - t^2 (2 - \nu_k)], A_{2k} = 4 J_1 S_1 t - 4 J_2 S_2 (1 - \nu_k), B_{1k} = -4 J_2 S_1 (1 - t^2) \nu_k, B_{2k} = 0, C_k = 2 J_1 \sqrt{S_1 S_2} (1 + t) \gamma_k, D_k = -2 J_1 \sqrt{S_1 S_2} (1 - t) \gamma_k, \) and \( t^{-1} = 2 \sigma / a \) (see Fig. 6). As may be expected, in addition to the quadratic spin-wave excitations, Eq. (6), close to the zone corners there appear linear Goldstone excitations connected to the spontaneously broken \( U(1) \) symmetry in the C phase. For example, in the vicinity of \( k = (0, \pi) \) the spectrum takes the form

\[ \omega_1^{(C)} = v \sqrt{k_1^2 + (\pi - k_2)^2} , \quad v = 4 J_1 S_1 \sqrt{\alpha^2 - \alpha_{c1}^2} . \]

Note that the spin-wave velocity \( v \) vanishes at the F-C transition point \( \alpha_{c1} \).
Finally, the spin-wave spectrum in the mixed-spin collinear phase is given by the general relation (5), where $A_{1k} = 4J_1S_1$, $A_{2k} = 4J_2S_2$, $B_{1k} = 4J_2S_1\nu k$, $B_{2k} = 4J_2S_2\nu k$, $C_k = 2J_1\sqrt{S_1S_2}\cos k$, and $D_k = 2J_1\sqrt{S_1S_2}\cos k_1$ (see Fig. 7). As compared to the antiferromagnetic $J_1 - J_2$ model, the excitation spectrum in the mixed-spin collinear phase shows some important peculiarities which specify the instability at the classical transition point $\alpha = 0.5$. First, in the mixed-spin system the degeneracy of the spin-wave branches $\omega^{(N)}_1$ and $\omega^{(N)}_2$ is completely removed. Second, the spin-wave excitations described by $\omega^{(N)}_2$ formally remain stable down to the point $\alpha = \sqrt{3}/(\sigma + 1) = \sqrt{2}/3 \approx 0.47$ for the $(1, 1/2)$ system. The phase transition at $\alpha = 0.5$ is entirely driven by instabilities in the lower $\omega^{(N)}_1$ excitation branch. At the classical spin-flop transition point ($\alpha = 0.5$), the branch $\omega^{(N)}_1$ exhibits two full lines of zero modes (i.e., $k_1 = 0$ and $k_2 = 0$) in the PBZ. The soft lines are connected with ferromagnetic fluctuations in the $e_1$ and $e_2$ directions on the square lattice, so that we can expect that quantum fluctuations do not change the position of the classical transition point (in accord with the data presented in Fig. 7).

III. CONCLUSIONS

In conclusion, we have studied the quantum spin phases in the mixed-spin $J_1 - J_2$ Heisenberg model defined on a square lattice. The reported results support a phase diagram with an additional quantum spin phase close to the classical spin-flop transition. In particular, in the extreme quantum case $(1, 1/2)$, we have found indications of a quantum spin phase (in the region $0.46 < \alpha < 0.5$) which is characterized by magnetically disordered $S_2$ spins and Néel long-range ordered $S_1$ spins. The latter suggestion is based (i) on the analysis of the low-lying excitations showing that the mixed-spin collinear state survives at most down to the classical transition point $\alpha = 0.5$ and (ii) on the SWT and ED results for the sublattice magnetization in the C phase $m_2$: both methods predict that $m_2$ vanishes at a point ($\alpha \approx 0.46$) preceding the classical transition point $\alpha = 0.5$. However, due to strong finite-size effects in the ED data and the qualitative character of the SWT analysis, we can not exclude the spin-liquid state as a possible ground state in the above region. Studies with other analytical and numerical methods would be necessary to characterize the true ground state in the above region.

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1. S. Das Sarma, S. Sachdev, and L. Zheng, Phys. Rev. B 58, 4672 (1998), and references therein.
2. Second, the spin-wave excitations described by $\omega^{(N)}_2$ formally remain stable down to the point $\alpha = \sqrt{3}/(\sigma + 1) = \sqrt{2}/3 \approx 0.47$ for the $(1, 1/2)$ system. The phase transition at $\alpha = 0.5$ is entirely driven by instabilities in the lower $\omega^{(N)}_1$ excitation branch. At the classical spin-flop transition point ($\alpha = 0.5$), the branch $\omega^{(N)}_1$ exhibits two full lines of zero modes (i.e., $k_1 = 0$ and $k_2 = 0$) in the PBZ. The soft lines are connected with ferromagnetic fluctuations in the $e_1$ and $e_2$ directions on the square lattice, so that we can expect that quantum fluctuations do not change the position of the classical transition point (in accord with the data presented in Fig. 7).

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1. S. Das Sarma, S. Sachdev, and L. Zheng, Phys. Rev. B 58, 4672 (1998), and references therein.
2. At filling factor $\nu = 2$ the system can be mapped onto the spin-$4$ easy-plane Heisenberg ferromagnetic model subject to a magnetic field along the $z$ direction: K. Yang, Phys. Rev. B 60, 15 578 (1999).
3. Y. Matsushita, M. Gelfand, and C. Ishii, J. Phys. Soc. Jpn. 66, 3648 (1997); M. Troyer and S. Sachdev, Phys. Rev. Lett. 81, 5418 (1998).
4. S. Sachdev, Z. Phys. B 94, 469 (1994); S. Sachdev and T. Seuthil, Ann. Phys. (NY) 251, 76 (1996).
5. L. Capriotti, F. Becca, A. Parola, and S. Sorella, Phys. Rev. Lett. 87, 097201 (2001); O.P. Sushkov, J. Oitman, and Z. Weinhold, Phys. Rev. B 63, 104420 (2001).
6. O. Kahn, Molecular magnetism (VCH, New York, 1993).
7. The classical N phase is degenerate in respect to global rotations of the $S_1$ (or $S_2$) subsystem. Quantum fluctuations choose one of the collinear configurations corresponding to ferromagnetic spin arrangements along, say, the $e_1$ or the $e_2$ directions on the square lattice.
8. Below we present extrapolated results obtained with the SBRm-m approximation scheme ($m = 2, 4$, and $6$ in the F and N phases; $m = 3, 4$, and $5$ in the C phase). For a recent elaborate description of the CCM method, see D.J.J. Farnell, K.A. Gernoth, and R.F. Bishop, Phys. Rev. B 64, 172409 (2001).
9. Such a method to obtain the pitch angle has previously been used by Ch. Waldtmann et al., Phys. Rev. B 62, 9472 (2000).
10. Since in the SWT for fixed $S_2 = 1/2$ the position of the point $\alpha^* < 0.5$ monotonically changes towards larger $\alpha$ for larger $S_1$, it is natural to expect that the instability at
$\alpha^* < 0.5$ exists in the extreme quantum case $S_1 = 1$ as well.

The region beyond $\alpha^*$ can be studied in the framework of the modified spin-wave theories: M. Takahashi, Phys. Rev. B 40, 2494 (1989); J.E Hirsch and S. Tang, Phys. Rev. B 40, 4769 (1989); Q.F Zhong and S. Sorella, Europhys. Lett. 21, 629 (1993).

Note that in the classical mixed-spin collinear phase the chain configurations in the $e_2$ direction are characterized with a finite ferromagnetic moment per cell. As a result, the soft line $k_1 = 0$ in the mixed-spin state is connected with ferromagnetic spin fluctuation. On the other hand, the above ferromagnetic moment is zero in the uniform-spin $(S_1 = S_2)$ collinear phase, so that the soft line $k_1 = 0$ is connected with antiferromagnetic fluctuations. This explains the change of the position of the $\alpha_{c2}$ point ($\alpha_{c2} \approx 0.6$) found in the spin-$\frac{1}{2}$ $J_1 - J_2$ Heisenberg model.