

Nematic Order in Square Lattice Frustrated Ferromagnets

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We present a new scenario for the breakdown of ferromagnetic order in a two-dimensional quantum magnet with competing ferromagnetic and antiferromagnetic interactions. In this, dynamical effects lead to the formation of two-magnon bound states, which undergo Bose-Einstein condensation, giving rise to bond-centered nematic order. This scenario is explored in some detail for an extended Heisenberg model on a square lattice. In particular, we present numerical evidence confirming the existence of a state with d -wave nematic correlations but no long-range magnetic order, lying between the saturated ferromagnetic and collinear antiferromagnetic phases of the ferromagnetic J_1 - J_2 model. We argue by continuity of spectra that this phase is also present in a model with 4-spin cyclic exchange.

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The search for a true quantum “spin liquid”—a quantum magnet which remains disordered at the very lowest temperatures—in dimensions greater than one has been central to research on quantum magnets for more than three decades. Following Anderson [1], most models of quantum spin liquids proposed to date have been based on frustrated antiferromagnetic (AF) interactions. The resulting spin-liquid states involve strong singlet bonds between spins, which give rise to a gap in the spin excitation spectrum (for a review, see, e.g., [2]).

Nonetheless, the best characterized experimental realization of a quantum spin liquid is believed to occur in two-dimensional films of solid ^3He , where the interactions between spins are predominantly ferromagnetic (FM), and the resulting state is gapless [3,4]. This raises the interesting question of whether the breakdown of long-ranged FM order offers a new route to a spin-liquid ground state.

The purpose of this Letter is to demonstrate that a gapless spin liquid can indeed occur in a simple two-dimensional model of quantum spins on a square lattice with predominantly FM interactions. To this end, we present a set of analytic and numerical results which show how a new type of “bond-nematic” state emerges from the ruins of FM order in an extended Heisenberg model on a square lattice. Our work is inspired by a number of new FM square lattice compounds [5,6] and has close parallels in earlier work on spin chains [7]. The special case of the multiple spin exchange model on a triangular lattice—which is believed to describe the magnetism of two-dimensional solid ^3He —will be dealt with elsewhere [8].

While these results are rather general, for concreteness in this Letter we consider a square lattice $S = 1/2$ frustrated ferromagnet defined by the Hamiltonian

$$\mathcal{H} = 2J_1 \sum_{\langle ij \rangle_1} \mathbf{S}_i \cdot \mathbf{S}_j + 2J_2 \sum_{\langle ij \rangle_2} \mathbf{S}_i \cdot \mathbf{S}_j + K \sum_{\langle 1234 \rangle} (P_{1234} + P_{1234}^{-1}) - h \sum_i S_i^z, \quad (1)$$

where $\langle ij \rangle_1$ counts nearest neighbor bonds, $\langle ij \rangle_2$ counts next-nearest neighbor bonds, and P_{1234} performs a cyclic exchange of spins on a single square plaquette. We restrict ourselves to the case where $J_1 < 0$ is FM and the other two interactions $J_2 \geq 0$ and $K \geq 0$ are AF.

The essence of our results, summarized in Figs. 1 and 2, is as follows—the competition between FM and AF interactions leads to a large “accidental” degeneracy in the one-magnon spectrum of the FM phase, which then becomes highly susceptible to the formation of two-magnon

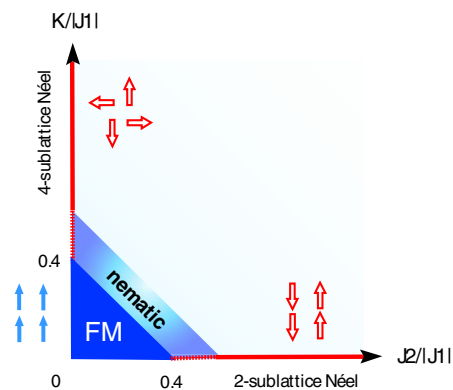


FIG. 1 (color online). Phase diagram for the spin-1/2 J_1 - J_2 - K Heisenberg model on a square lattice, in the parameter range where $J_1 < 0$ is FM and both $J_2 \geq 0$ and $K \geq 0$ are AF. Solid lines on the $J_2/|J_1|$ and $K/|J_1|$ axes denote known Néel phases. A single nematic phase interpolates between the FM occurring for large FM J_1 and these two, different, AF ordered states.

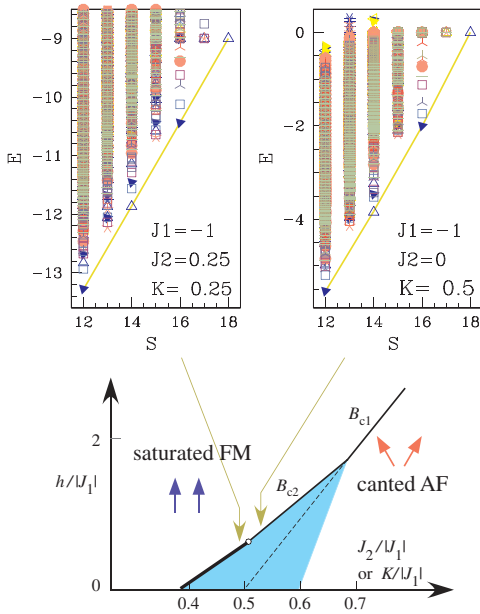


FIG. 2 (color). Schematic phase diagram of the FM J_1 - J_2 - K extended Heisenberg model in applied field, showing how the two-magnon instability crosses over from 1st to 2nd order in applied field and is eventually succeeded by a conventional one-magnon instability. The inset shows representative spectra for 1st and 2nd order transitions into d -wave nematic state (high-spin states only).

bound states. These condense and give rise to a new singlet phase lying between the classical FM and known Néel phases of the model. This phase has a well defined Goldstone mode but no long-range magnetic order in the conventional sense. We introduce an appropriate order parameter for this phase and identify it as a d -wave n -type “bond-nematic” state, distinct from conventional “ n -type” and “ p -type” nematic states known to occur in other closely related models [9,10]. From numerical exact diagonalization calculations, we are able both to confirm that this phase exists and to establish that it interpolates between the two well-studied limits of Eq. (1), the square lattice J_1 - J_2 model for $K \rightarrow 0$ and the square lattice J_1 - K multiple spin exchange model for $J_2 \rightarrow 0$.

We now turn to the analysis. For large FM J_1 , the ground state of Eq. (1) is a uniform saturated FM. In the FM J_1 - J_2 model, for $K = 0$ this gives way to 2-sublattice collinear Néel AF at large J_2 [11]. In the FM J_1 - K model, for $J_2 = 0$, the corresponding Néel state is an orthogonal 4-sublattice structure [12]. Our analysis begins with a very simple observation—at a classical level, the transition between FM and Néel states is 2nd order and takes place where $|J_1| = 2(K + J_2)$. However, exactly at this transition, the one-magnon spectrum of the FM phase possesses entire lines of zero modes [13]. This accidental degeneracy has profound consequences for magnetism.

First, it leads to large zero point fluctuations which shift the first instability of the FM to somewhat weaker AF

coupling. We have performed numerical exact diagonalization (ED) of Eq. (1) for clusters of 16, 20, 32, and 36 spins [14]. The evolution of the ground-state energy indicates that a 1st order transition from saturated FM to singlet ground states occurs for $|J_1| \approx 2.5(K + J_2)$.

Second, these fluctuations also destabilize the neighboring Néel phase. Semiclassical estimates for the square lattice J_1 - J_2 model suggest that the sublattice magnetization of its collinear Néel phase vanishes for $|J_1| \approx 1.97J_2$ [13]. These calculations almost certainly underestimate quantum effects.

These arguments provide a good *a priori* reason to believe that the extended Heisenberg model Eq. (1) supports a new spin-liquid phase for a finite range of parameters between its FM and Néel phases. However, they give little insight into what this phase might be.

In order to better answer this question, we introduce magnetic field h and examine the nature of the first instability of the fully saturated paramagnet as the magnetic field is reduced. The situation is summarized in Fig. 2. For parameters well within the classical Néel phases of Eq. (1), the first instability of the saturated state occurs at $h = B_{c1}$ and is controlled by the lowest one-magnon excitation in an applied field. This instability is against a conventional 2-sublattice canted AF state.

In the proposed spin-liquid region, however, the one-magnon spectrum is highly degenerate. In the extreme case $J_2 = 0$, $K = |J_1|/2$, the one-magnon dispersion vanishes altogether, so there is no net energy gain in flipping one spin at any \mathbf{q} and the single flipped spins are always localized. On the other hand, *pairs* of flipped spins on neighboring sites *can* propagate coherently under the action of the cyclic exchange operator P_{1234} and so gain kinetic energy. It is therefore reasonable to ask whether the transition out of the FM phase is controlled by such two-magnon bound states.

Both the one-body problem of a single flipped spin-1/2 in a saturated FM background $|FM\rangle$ and the two-body problem of two interacting flipped spins can be solved exactly. We have calculated the energy of both states in the proposed spin-liquid region and find that, at a critical value of field $h = B_{c2}$, two-magnon bound states of the form $|\phi\rangle = \sum_{ij} \phi_{ij} S_i^- S_j^- |FM\rangle$ with d -wave symmetry are gapless and give the first instability, while one-magnon states have higher positive energy—see Fig. 2. The binding energy of the two-magnon state is of purely kinetic origin. In zero field, the two-magnon bound states first become stable in the J_1 - J_2 model for $J_2/|J_1| = 0.408$ (cf. Ref. [15]) and in the J_1 - K model for $K/|J_1| = 0.364$. These values should be compared with the numerically determined boundary of the FM phase at $(J_2 + K)/|J_1| \approx 0.4$. In the special case $J_2 = 0$, $K = |J_1|/2$, the two-magnon bound state has the compact form $\phi_{ij} = (1/\sqrt{2}) \times [\delta_{i,j-e_1} - \delta_{i,j-e_2}]$, where $\mathbf{e}_1 = (1, 0)$ and $\mathbf{e}_2 = (0, 1)$ connect neighboring sites on a square lattice.

What then happens below B_{c2} ? In the case of the FM J_1 - K model for $J_2 = 0$, the answer is quite simple. Two-magnon pairs undergo Bose-Einstein condensation. For this set of parameters, there is a weak, repulsive interaction between two-magnon pairs, so the transition is 2nd order. This is evident from ED spectra—the locus of the lowest lying state in the high-spin sector has a *convex* curvature as a function of S , so the magnetization of the system must evolve continuously at B_{c2} —see the inset in Fig. 2. Even spin sector states appear only in the lowest states in magnetic field. This period two is the evidence of condensation of two-magnon pairs.

In the FM J_1 - J_2 model for $K = 0$, the situation is a little more complicated. The net interaction between two-magnon pairs is attractive, and the transition is 1st order. Again this is evident in ED from the *concave* curvature of the high-spin states, which signals a jump in magnetization at saturation field. A similar 1st order transition appears in the J_1 - K model for $J_2 = 0$ well close to the FM phase boundary. However, in all of these cases the structure of the low lying states is essentially the same—even spin sector states always have lower energy than odd spin sector states and show an alternation between the s -wave A_1 and d -wave B_1 irreps of the square lattice space group C_{4v} . We return these points below.

Where bound pairs of magnons condense, they give rise to a new form of bond-nematic order. This is to say that, while the orientation of individual spins remains undetermined, the traceless rank two tensor

$$\mathcal{O}^{\alpha\beta}(\mathbf{r}_i, \mathbf{r}_j) = \frac{1}{2}(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) - \frac{1}{3}\delta^{\alpha\beta}\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle \quad (2)$$

does exhibit long-range order [16]. The matrix elements of $\mathcal{O}^{\alpha\beta}(\mathbf{r}_i, \mathbf{r}_j)$ are connected to the magnon pairing operator through the relation $S_i^- S_j^- = \mathcal{O}^{xx} - \mathcal{O}^{yy} - 2i\mathcal{O}^{xy}$. Physically, one can think of this tensor operator as revealing the order “hidden” in the spin-1/2 quantum spin liquid by projecting into a symmetrized spin-1 Hilbert space of bond variables with long-range correlations.

The order parameter Eq. (2) is distinct from the site-centered n -type nematic order seen in certain models with $S = 1$ [9] and from the p -type chiral order found in the multiple spin exchange model with AF interactions [10]. However, a very similar order parameter was previously introduced in the context of one-dimensional frustrated spin chains [7].

Having established the unconventional nature of the state near saturation, we now turn to the nature of the ground state in the absence of applied field. Proof that this is a spin liquid follows from the finite-size scaling properties of ED spectra. In Fig. 3, we present the spectrum of a cluster of $N = 36$ spins for $J_1 = -1$, $J_2 = 0.4$, and $K = 0$, very close to the boundary with the saturated FM phase. The signature of long-range Néel order in such a spectrum would be the existence of a set of “quasidegenerate joint states” (QDJS) which form the $N = \infty$ ground

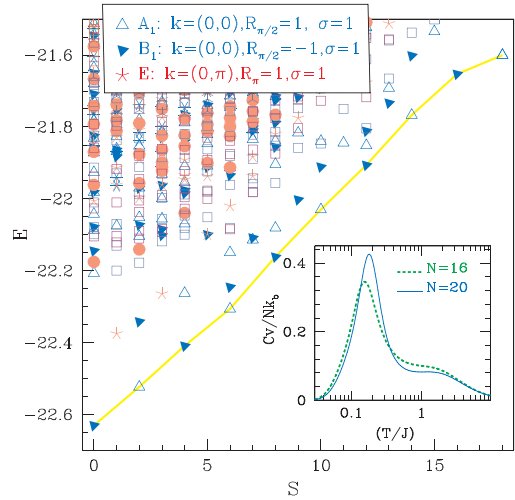


FIG. 3 (color). Spectrum of extended Heisenberg model for $N = 36$ spin cluster with $J_1 = -1$, $J_2 = 0.4$, $K = 0$, showing the marked alternation in energies of the lowest lying states in even and odd spin sectors. The inset shows the double peak structure of the heat capacity $c_V(T)$ for clusters of $N = 16$ and $N = 20$ spins and the same set of parameters.

state, present in every spin sector (up to $S \sim \sqrt{N}$), with energies scaling as $E_{\text{QDJS}} \sim \frac{S(S+1)}{N}$ and are well separated (at finite N) from the lowest excitations of the continuum (magnon excitations scaling as $\sim 1/\sqrt{N}$). The QDJS specific to $(\pi, 0)$ collinear AF order comprise one A_1 and one B_1 state for even S and a (twofold degenerate) E level for S odd [17]. The gaps to the low lying states in each spin sector should evolve as $\Delta \sim \frac{1}{N}[\alpha - (\beta/\sqrt{N})]$, with coefficients α and β determined by the spin stiffness and spin wave velocity of the Néel state, and thus display a negative curvature when plotted against $1/N$ [18].

It is immediately clear from the spectrum in Fig. 3 that the energies of the odd spin states, which include the E symmetry states, are well separated from those of the even spin states. Thus, the spectrum is not compatible with the expected Néel order. The unconventional nature of the ground state is also reflected in the finite-size scaling of gaps, shown in Fig. 4. The gaps in the even spin sectors scale to zero in the expected manner. But the gaps of E symmetry states in odd spin sectors do not—in fact, they appear to scale to a finite value $\Delta_{\text{odd}} \sim 0.4$.

This is conclusive proof that the FM J_1 - J_2 model *does not* exhibit Néel order in the immediate neighborhood of its FM phase. But we can draw a still stronger conclusion. The spin even-odd oscillation found in high-spin states persists down to low-spin states, and the A_1 and B_1 states found in even spin sectors are precisely the same states which emerge from the two-magnon instability of the saturated FM in applied magnetic field. The existence of this set of QDJS is the signature of nematic order with d -wave symmetry. Thus, the nematic order found near to saturation persists down to zero magnetic field.

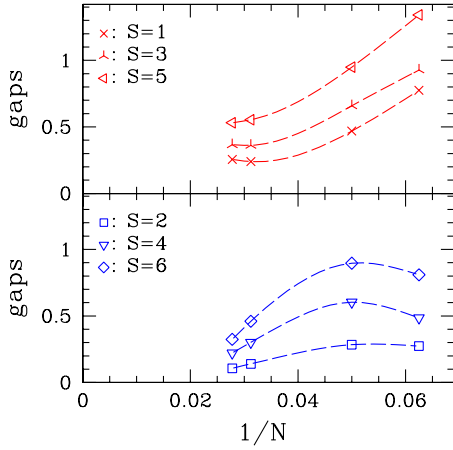


FIG. 4 (color online). Finite-size scaling of the energy gaps of extended Heisenberg model with $J_1 = -1$, $J_2 = 0.4$, $K = 0$, for A_1 and B_1 symmetry states (lower) and E ones (upper) in the even and odd spin sectors, respectively.

Such nematic order has a degenerate ground state composed of even spin states. It has gapless Goldstone modes analogous to the lowest magnons states of the Néel state, present in the $S = 1$ sector, scaling as $1/\sqrt{N}$. Results for a prototypical nematic correlation function $\sum_{\alpha,\beta} \langle O^{\alpha\beta}(\mathbf{0}, \mathbf{e}_1) O^{\alpha\beta}(\mathbf{r}, \mathbf{r} + \mathbf{e}_a) \rangle$ ($a = 1, 2$) in the nematic ground state are presented in Fig. 5. Nematic correlations exhibit a “stripey” character and exist in the whole system. Because of the underlying d -wave symmetry, correlations on parallel bonds have positive sign, while correlations on perpendicular bonds are negative.

The finite temperature properties of the FM J_1 - J_2 model provides further insight into the bond-nematic state. In an inset in Fig. 3, we show the heat capacity calculated for the same exchange parameters. It exhibits two clear energy

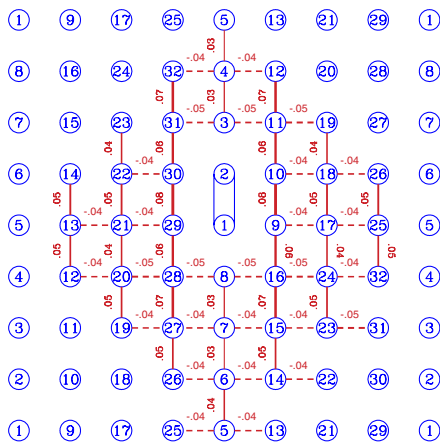


FIG. 5 (color online). Bond-nematic correlations of an extended Heisenberg model with $J_1 = -1$, $J_2 = 0.4$, $K = 0$, for a cluster of $N = 32$ spins, showing the d -wave character of the nematic phase. Correlations are measured relative to the reference bond 1–2.

scales—a broad structure extending to $T \sim |J_1|$, which reflects the formation of $S = 1$ objects from the paramagnetic “soup,” and a sharp peak at $T \sim 0.1|J_1|$, where these $S = 1$ objects start to order nematically.

Although it is hard to put a precise boundary on the stability of the $(\pi, 0)$ collinear AF state, our ED studies suggest that Néel order begins to break down in the FM J_1 - J_2 model for $J_2 \sim 0.6$ – $0.7|J_1|$. We also have performed ED calculations for parameter sets interpolating between the FM J_1 - J_2 model for $K = 0$ and the FM multiple spin exchange model for $J_2 = 0$. These confirm that the nature of the state bordering the FM phase does not change. Thus, a single nematic phase interpolates between the two different Néel order parameters in these two limits and the FM phase for large $|J_1|$, as shown in Fig. 1.

To conclude—in the frustrated square lattice FM’s considered in this Letter, the saturated FM ground state undergoes a first order transition into a gapless spin-liquid state with bond-nematic order. In applied magnetic field, the transition becomes second order and can be related directly to the condensation of two-magnon bound states.

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