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Emergent multipolar spin correlations in a fluctuating spiral: The frustrated ferromagnetic spin-$\frac{1}{2}$ Heisenberg chain in a magnetic field

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We present the phase diagram of the frustrated ferromagnetic $S=\frac{1}{2}$ Heisenberg J_1-J_2 chain in a magnetic field, obtained by large scale exact diagonalizations and density matrix renormalization group simulations. A vector chirally ordered state, metamagnetic behavior and a sequence of spin-multipolar Luttinger liquid phases up to hexadecupolar kind are found. We provide numerical evidence for a locking mechanism, which can drive spiral states toward spin-multipolar phases, such as quadrupolar or octupolar phases. Our results also shed light on previously discovered spin-multipolar phases in two-dimensional $S=\frac{1}{2}$ quantum magnets in a magnetic field.

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Spiral or helical ground states are an old and well-understood concept in classical magnetism,[1] and several materials are successfully described based on theories of spiral states. For low spin and dimensionality however quantum fluctuations become important and might destabilize the spiral states. Given that spiral states generally arise due to competing interactions, fluctuations are expected to be particularly strong.

A prominent instability of spiral states is their intrinsic twist $\langle \mathbf{S}_i \times \mathbf{S}_j \rangle$ (vector chirality).[2] It has been recognized that finite temperature[3] or quantum[4] fluctuations can disorder the spin moment $\langle \mathbf{S}_i \rangle$ of the spiral, while the twist remains finite. Such a state is called p-type spin nematic.[5] In the context of quantum fluctuations such a scenario has been confirmed recently in a ring-exchange model,[6] while possible experimental evidence for the thermal scenario has been presented in.[7] The twist also gained attention in multiferroics, since it couples directly to the ferroelectricity.[8]

In this Rapid Communication we provide evidence for the existence of yet a different instability of spiral states *toward spin-multipolar phases*. The basic idea is that many spin-multipolar order parameters are finite in the magnetically ordered spiral state, but that under a suitable amount of fluctuations the primary spin order is lost, while a spin-multipolar order parameter survives. We demonstrate this mechanism based on the magnetic field phase diagram of a prototypical model, the frustrated $S=\frac{1}{2}$ Heisenberg chain with ferromagnetic nearest-neighbor and antiferromagnetic next nearest-neighbor interactions. Furthermore we show that this instability provides a natural and unified understanding of previously discovered two-dimensional spin-multipolar phases.[9,10]

To be specific, we determine numerically the phase diagram of the following Hamiltonian:

$$H = J_1 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} + J_2 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+2} - h \sum_i S_i^z, \tag{1}$$

and we set $J_1=-1$, $J_2 \geq 0$ in the following. $\mathbf{S}_i$ are spin-1/2 operators at site i, while h denotes the uniform magnetic field. The magnetization is defined as $m := 1/L \Sigma_i S_i^z$. We employ exact diagonalizations (EDs) on systems sizes up to $L=64$ sites complemented by density matrix renormalization group (DMRG) (Ref. 11) simulations on open systems of maximal length $L=384$, retaining up to 800 basis states.

The classical ground state of Hamiltonian (1) is ferromagnetic for $J_2 < 1/4$ and a spiral with pitch angle $\varphi = \arccos(1/4J_2) \in [0, \pi/2]$ otherwise. The Lifshitz point is located at $J_2=1/4$. In a magnetic field the spins develop a uniform component along the field, while the pitch angle in the plane transverse to the field axis is unaltered by the field.

The zero field quantum mechanical phase diagram for $S=\frac{1}{2}$ is still unsettled. Field theoretical work predicts a finite, but tiny gap accompanied by dimerization[12,13] for $J_2 > 1/4$, which present numerical approaches are unable to resolve. The classical Lifshitz point $J_2=1/4$ is not renormalized for $S=\frac{1}{2}$, and the transition point manifests itself on finite systems as a level crossing between the ferromagnetic multiplet and an exactly known singlet state.[14] The theoretical phase diagram at finite field has recently received considerable attention,[15–17] triggered by experiments on quasi one-dimensional cuprate helimagnets.[18–20] One of the most peculiar features of the finite size magnetization process is the appearance of elementary magnetization steps of $\Delta S^z = 2, 3, 4$ in certain J_2 and m regions. This has been attributed to the formation of bound states of spin flips, leading to dominant spin-multipolar correlations close to saturation. A detailed phase diagram is however still lacking.

We present our numerical phase diagram in the $J_2/|J_1|$ vs. m/m_{sat} plane in Fig. 1. At least five different phases are present. The low magnetization region consists of a single vector chiral phase (gray). Below the saturation magnetization we confirm the presence of three different multipolar Luttinger liquid phases (red, green, and blue). The red phase extends up to $J_2 \to \infty$,[16] and its lower border approaches $m=0^+$ in that limit. All three multipolar liquids present a crossover as a function of m/m_{sat}, where the dominant correlations change from spin-multipolar close to saturation to spin-density wave (SDW) character at lower magnetization. One also expects a tiny incommensurate $p=2$ phase close to the $p=3$ phase,[17] which we did not aim to localize in this

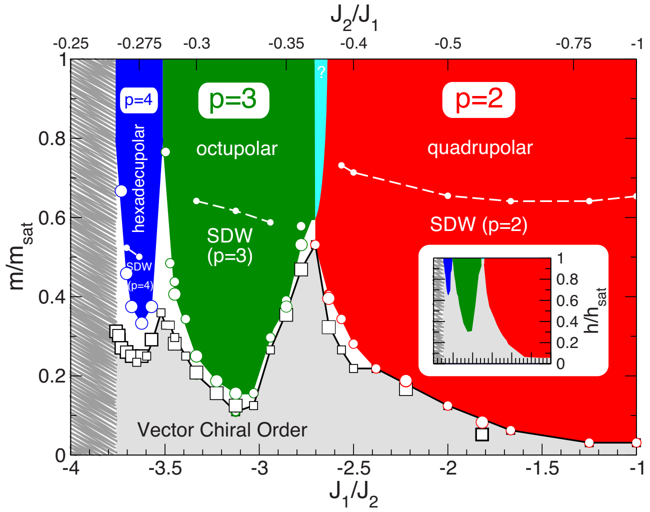

FIG. 1. (Color online) Phase diagram of the frustrated ferromagnetic chain (1) in the J_1/J_2 vs $m/m_{\rm sat}$ plane. The gray low-m region exhibits vector chiral long range order. The colored regions denote spin-multipolar Luttinger liquids of bound states of $p=2,3,4$ spin flips. Close to saturation the dominant correlations are multipolar, while below the dashed crossover lines, the dominant correlations are of SDW(p) type. The tiny cyan colored region corresponds to an incommensurate $p=2$ phase. The white region denotes a metamagnetic jump. Finally the scribbled region close to the transition $J_1/J_2 \to -4$ has not been studied here, but consists most likely of a low field vector chiral phase, followed by a metamagnetic region extending up to saturation magnetization. The inset shows the same diagram in the J_1/J_2 vs $h/h_{\rm sat}$ plane.

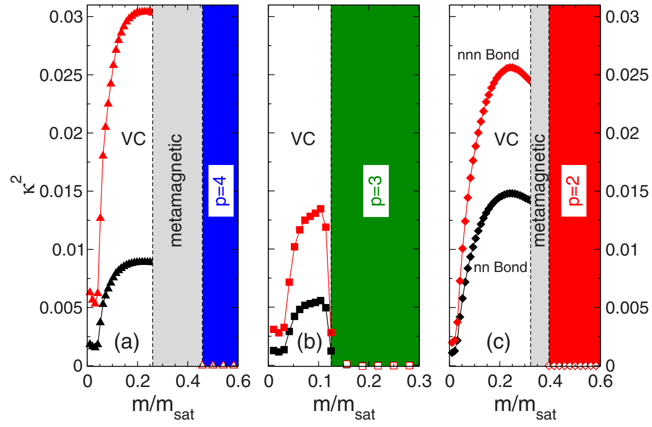

FIG. 2. (Color online) Squared vector chirality order parameter κ^2 [Eq. (2)] in the low magnetization phase for different values of $J_2/|J_1|$ as a function of $m/m_{\rm sat}$: (a) $J_2/|J_1|=0.27$, (b) $J_2/|J_1|=0.32$, and (c) $J_2/|J_1|=0.38$. The order parameter vanishes in the multipolar Luttinger liquids.

study. Finally the multipolar Luttinger liquids are separated from the vector chiral phase by a metamagnetic transition, which occupies a larger and larger fraction of m as $J_2 \to 1/4^+$, leading to an absence of multipolar liquids composed of five or more spin flips. In the following we will characterize these phases in more detail and put forward an explanation for the occurrence and locations of the spin-multipolar phases.

For $m>0$ we reveal a contiguous phase sustaining long range vector chiral order[21] (breaking discrete parity symmetry), similar to phases recently discovered for $J_1, J_2>0$.[22,23] Direct evidence for the presence of this phase is obtained from measurements of the squared vector chiral order parameter,

$$\kappa^2(r,d) \coloneqq \langle [\mathbf{S}_0 \times \mathbf{S}_d]^z [\mathbf{S}_r \times \mathbf{S}_{r+d}]^z \rangle. \tag{2}$$

In Fig. 2 we display DMRG results for long distance correlations between J_1 bonds ($d=1$, black symbols) and J_2 bonds ($d=2$, red symbols) obtained on a $L=192$ system. The three chosen values of J_2 reflect positions underneath each of the three spin-multipolar Luttinger liquids shown in Fig. 1. The non-monotonic behavior of the correlations at very small m is probably a finite size artifact or convergence issue. Beyond the long range order in the vector chirality, the system behaves as a single channel Luttinger liquid (with central charge $c=1$, confirmed by our DMRG based entanglement entropy analysis[24]) with critical incommensurate transverse spin correlation functions.[22] The transition to the spin-multipolar phases at larger m seems to occur generically via metamagnetic behavior (cf. left and right panels of Fig. 2). For the parameter set in the middle panel we expect the same behavior, but it can't be resolved based on the system sizes used.

Hamiltonian (1) presents unusual elementary step sizes $\Delta S^z>1$ in some extended $J_2/|J_1|$ and m domains, where ΔS^z is independent of the system size.[15] This phenomenon has been explained based on the formation of bound states of $p=\Delta S^z$ magnons in the completely saturated state, and at finite $m/m_{\rm sat}$ a description in terms of a single component Luttinger liquid of bound states has been put forward.[16,17] We have determined the extension of the $\Delta S^z=2,3,4$ regions in Fig. 1, based on exact diagonalizations on systems sizes up to 32 sites and DMRG simulations on systems up to 192 sites. The boundaries are in very good agreement with previous results[15] where available. The $\Delta S^z=3$ and 4 domains form lobes which are widest at $m_{\rm sat}$ and whose tips do not extend down to zero magnetization. The higher lobes are successively narrower in the J_2 direction. We have also searched for $\Delta S^z=5$ and higher regions, but found them to be unstable against a direct metamagnetic transition from the vector chiral phase to full saturation. Individual bound states of $p\geq 5$ magnons do exist (see below), but they experience a too strong mutual attraction to be thermodynamically stable.

An exciting property of the Luttinger liquids of p bound magnon states[17] is that the transverse spin correlations are exponentially decaying as a function of distance due to the binding, while p-multipolar spin correlations

$$\left\langle \prod_{n=0}^{p-1} S^+_{0+n} \prod_{n=0}^{p-1} S^-_{r+n} \right\rangle \sim (-1)^r \left(\frac{1}{r}\right)^{1/K} \tag{3}$$

are critical with wave vector π (multipolar correlations with $p'<p$ also decay exponentially). $p=2,3,4$ correspond to quadrupolar, octupolar, and hexadecupolar correlations, respectively. Therefore they can be considered as one-dimensional analogs of spin multipolar ordered phases found

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in higher dimensions. Another important correlation function is the longitudinal spin correlator, which is also critical,[17]

$$\langle S_0^z S_r^z \rangle - m^2 \sim \cos\left[\frac{(1-m/m_{\text{sat}})\pi r}{p}\right]\left(\frac{1}{r}\right)^K. \tag{4}$$

We verified numerically $c=1$ and determined the Luttinger parameter K as a function of m and J_2 by fits to the local S^z profile in DMRG simulations. An important information is contained in the crossover line $K=1$ where p-multipolar and longitudinal spin correlations decay with the same exponent. This crossover line is shown for the three lobes in Fig. 1. Close to saturation the spin-multipolar correlations dominate while toward the tip of the lobes the longitudinal spin correlations decay more slowly. The crossover line is rather flat in the J_2 direction, but drops to lower m values when going from $p=2$ to 3 and 4. This can be understood from the analogy with a Luttinger liquid of hardcore bosons of bound states which effectively becomes more dilute when increasing p at constant m.

After having established and characterized the phase diagram, we are lead to the intriguing question whether there is a deeper relation between the spin multipolar phases which would enable us to understand their presence and possibly predict related phenomena in other systems.

Let us first investigate how the longitudinal and transverse equal-time spin structure factors $\mathcal{S}^{zz}(q)$ and $\mathcal{S}^{xx}(q)$ evolve as a function of m. In Fig. 3 we display the location of the maximum of $\mathcal{S}^{zz}(q)$ and $\mathcal{S}^{xx}(q)$ [disregarding the $q=0$ peak in $\mathcal{S}^{zz}(q)$ due to the total magnetization] for three representative J_2 values. At $m=0$ it is known that $\mathcal{S}(q)$ has a maximum at an incommensurate position $q_{\max}(J_2)$, which is strongly renormalized compared to the classical expectation $q_{\max}^{\text{class}}(J_2)=\arccos(1/4J_2)$.[25] In the low magnetization region, corresponding to the vector chiral phase, the location of the maxima of both structure factors are only weakly dependent on m, and in a first approximation remain the same as for $m=0$. However as m is increased, the $q_{\max}$ of $\mathcal{S}^{zz}(q)$ locks onto a straight line with slope $-\pi/p$. It seems that if $q_{\max}(J_2)/\pi > 1/3$ at $m=0$ then the magnetization process will enter the $p=2$ multipolar phase at larger m. If instead $1/3 > q_{\max}(J_2)/\pi > 1/4$ at $m=0$ then the system will enter the $p=3$ phase. Based on an extended analysis including several more J_2/J_1 values we are thus lead to conjecture that if

$$1/p > q_{\max}(J_2)/\pi > 1/(p+1) \quad \text{at} \quad m=0, \tag{5}$$

then the higher m region will lock onto the line with slope $-\pi/p$. According to the behavior of the longitudinal spin correlations in the multipolar Luttinger liquids [Eq. (4)], the multipolar liquid of order p leads precisely to a slope of $-\pi/p$.

A complementary point of view is provided by the consideration of a simple *classical* spiral. The corresponding magnetic structure factor has a peak at the propagation vector q of the spiral. It is straightforward to show that in such a state the in-plane multipolar correlations of order p [analog to Eq. (3)] have a propagation vector $q_p=p\cdot q$. Of course in a magnetically ordered state these multipolar correlations are not primary order parameters, but strong quantum fluctuations can wipe out the spin order, while a particular multipolar order parameter survives. Applying this observation to the structure factor results, we can understand the rule (5) as a locking of the multipolar-p correlations to π, which precisely corresponds to the staggered nature of the multipolar-p correlations in Eq. (3).

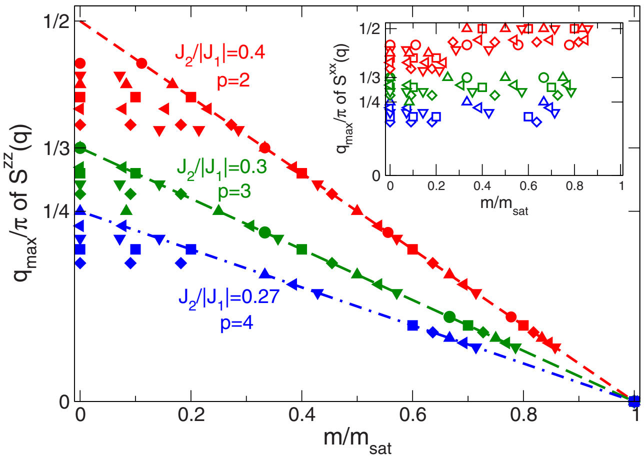

FIG. 3. (Color online) Wave vector q maximizing the longitudinal (inset: transverse) spin structure factor as a function of the magnetization m/m_{sat} for selected $J_2/|J_1|$ parameters, each one belonging to a different p sector at high m. The straight lines are guides to the eyes and show $\pi(1-m/m_{\text{sat}})/p$. The different symbols denotes ED results on system sizes ranging from 18 to 28 sites.

Despite the fact that there are no stable multipolar Luttinger liquids with $p>4$, it is possible to examine the process of individual bound state formation by pushing previous calculations from $p\leq 4$ in Ref. 17 to $p=8$. We performed ED without truncation on system sizes up to $L=64$, calculate the binding energy of p flipped spins [expressed as $h_{\text{sat}}(p)/h_{\text{sat}}(p=1)$] and display it in the upper panel of Fig. 4. We obtain a surprising series of stable bound states up to $p=8$, which are successively smaller in their J_2 extent, suggesting that $J_2=1/4$ is an accumulation point of $p\to\infty$ bound states. We now proceed to a comparison of the stability domain of the bound states to those predicted by locking rule (5) applied to the *zero field* structure factor. We fit a power law $q_{\max}(J_2)\sim(J_2-1/4)^\gamma$ (with $\gamma\approx 0.29$) to the finite size transition points, as shown by the bold black line in the lower panel of Fig. 4. This fitted line is used to construct the boundaries, giving rise to the lower color bar. The agreement between the transition boundaries in the upper and lower panels is excellent (apart from the $p=2$ to $p=3$ transition). It is striking that we are able to reproduce the domain of stability of bound states of magnons at the saturation magnetization, based solely on the spin structure factor obtained at zero field. This constitutes strong evidence for an approximate validity of locking rule Eq. (5) and the presence of a locking mechanism which pins the multipolar correlations to π at larger m as opposed to incommensurate transverse spin correlations for lower m. The transition from $p=2$ to $p=3$ is somewhat shifted compared to the prediction, and we attribute this discrepancy to the formation of a peculiar incommensurate $p=2$ bound state,[17,26] leading to a collapse of the kinetic energy of the $p=2$ bound state.

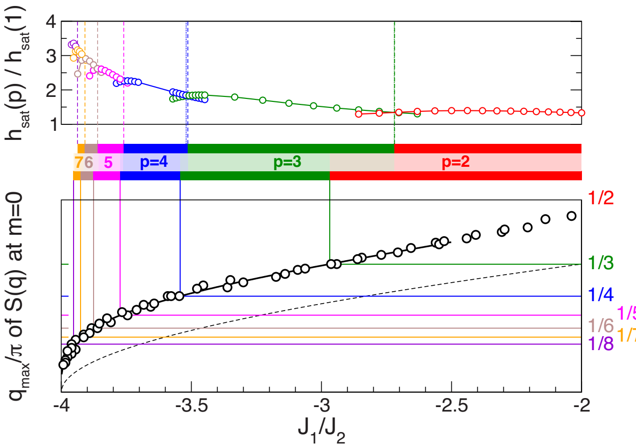

FIG. 4. (Color online) Upper panel: ED results for the domain of stability of bound states of p spin flips above the saturated ferromagnetic state. Lower panel: Location $q_{\max}$ of the maximum of the zero field spin structure factor $\mathcal{S}(q)$ as a function of J_2. The colored lines indicate the construction of the domain boundaries, based on the locking rule Eq. (5). The thin dashed line displays $q_{\max}^{\text{class}}$ for the classical model.

Our considerations presented above are not restricted to one dimension. Indeed the recently reported quadrupolar[9] and octupolar phases[10] in frustrated $S=\frac{1}{2}$ ferromagnets in a field also allow an interpretation as fluctuation destabilized magnetic states. In the J_1-J_2 model on the square lattice there is a magnetically ordered striped phase $[Q=(\pi,0)]$ in the neighborhood of the "bond nematic" phase. The nearest neighbor bond quadrupolar correlation functions in such a classical striped state have momentum (0,0) and are odd under a $\pi/2$ rotation of one bond. Interestingly this is exactly the signature of the correlation function reported in the magnetically disordered, "bond nematic" phase. Similarly for the "triatic" or octupolar phase, there is a canted magnetic three-sublattice state $[Q=(4\pi/3,0)]$ neighboring the "triatic" phase at high fields.[10] The derivation of the octupolar correlations on a triangle in the classical state again yields octupolar correlations matching the symmetry of the bound states reported in the "triatic" phase. Based on the success of our considerations we speculate that it could be possible to stabilize a uniform hexadecupolar phase in the square lattice J_1-J_3 model with ferromagnetic nearest neighbor interactions.

The different phases we found should be detectable experimentally in the quasi-1D compounds studied in Refs. 18–20. In neutron scattering experiments these phases are expected to manifest as follows: (i) a Bragg peak at low magnetizations corresponding to spiral order in the plane transverse to the magnetic field, (ii) a Bragg peak at intermediate magnetizations at wave vector $\pi(1-m/m_{\text{sat}})/p$ corresponding to SDW(p) magnetic order modulated along the field, and (iii) the absence of nontrivial magnetic Bragg peaks at high fields in the spin multipolar ordered phases.

We have established the phase diagram of the frustrated ferromagnetic $S=\frac{1}{2}$ Heisenberg chain in a uniform magnetic field. We provided an explanation for the appearance of a large number of bound states upon approaching the Lifshitz point at $J_2=1/4$, based on a locking mechanism leading to spin multipolar phases. It is an open problem whether these fluctuation driven multipolar phases and the locking mechanism also appear for $S>\frac{1}{2}$. In a recent paper by Hikihara *et al.*[27] a phase diagram very similar to ours has been obtained.

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