The frustrated ferromagnetic $S = 1/2$ Heisenberg chain in a magnetic field – How multipolar spin correlations emerge from magnetically ordered states

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Abstract. We present the phase diagram of the frustrated ferromagnetic $S = 1/2$ 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 order are found. Starting from classical considerations, we point out that various multipolar correlations are imprinted in a magnetic state and that they can survive the onset of frustration and quantum fluctuations which destroy the conventional magnetic order. Our results also shed new light on previously discovered spin multipolar phases in two-dimensional $S = 1/2$ quantum magnets in a magnetic field.

1. Introduction
Spiral or helical ground states are a well understood concept in classical magnetism [1]. Several materials are successfully described by theories based on spiral states. For low spin and dimensionality however, quantum fluctuations become important and might destabilize the spirals. 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 $\langle S_i \times S_j \rangle$ (vector chirality) [2]. It has been recognized that finite temperature [3] or quantum [4] fluctuations can disorder the spin moment $\langle S_i \rangle$ of the spiral, while the twist remains finite. Such a state is called $p$–type spin nematic [5]. In the case 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 Ref. [7]. The twist also gained attention in multiferroics, since it couples directly to the ferroelectricity [8].
Figure 1. Phase diagram of the frustrated ferromagnetic chain [Eq. (1)] as a function of $J_2$ and the magnetization $m$. The grey low-$m$ region exhibits vector chiral long range order. The colored regions denote spin multipolar Luttinger liquids of bound states with $p = 2, 3, 4$ spin flips. Close to saturation, the dominant correlations are multipolar (quadrupolar, octupolar, hexadecupolar), while below the dashed crossover lines, the dominant correlations are of SDW($p$)-type. The tiny cyan colored region possibly corresponds to an incommensurate $p = 2$ phase. The white region denotes a metamagnetic jump. The inset shows the same diagram in the $J_2$ vs. $h$ plane.

In this paper, we briefly review the main results of our recent preprint [9] in which we provide evidence for the existence of a novel instability of spiral states towards spin multipolar phases exemplified by the phase diagram of the frustrated ferromagnetic chain in a magnetic field

$$H = J_1 \sum_i S_i \cdot S_{i+1} + J_2 \sum_i S_i \cdot S_{i+2} - h \sum_i S_i^z .$$

Here $S_i$ are spin-1/2 operators at site $i$ and $h$ denotes the uniform magnetic field. In what follows, we set $J_1 = -1$ and consider $J_2 \geq 0$.

In Section 2 we discuss the numerical phase diagram reported by us in Ref. [9] and independently by Hikihara et al. in Ref. [10]. In Section 3, we start from the classical limit, and derive the chiral and multipolar correlators in a spiral state and confront these predictions with the quantum case by studying the spiral state in the uniformly distributed resonating valence bond (UDRVB) state. In Section 4, we provide evidence for the existence of a locking mechanism that pins different multipolar correlations to wave vector $\pi$ as $J_2$ is varied. Conclusions and open questions are presented in Section 5.

2. Phase diagram

In Fig. 1, we present the overall numerical phase diagram[9, 10] of the above Hamiltonian as a function of $J_2$ and the magnetization $m$. It was obtained by exact diagonalizations (ED) on systems with up to $L = 64$ sites, complemented by density-matrix-renormalization-group (DMRG) [11] simulations on open systems with maximally 384 sites, retaining up to 800 basis states. On these finite systems, the magnetization is defined as $m = \sum_i S_i^z / L$.

1 The concept applies actually to magnetically ordered states in general.
2.1. Vector chiral phase
For $m > 0$ we reveal a contiguous phase sustaining long range vector chiral order [16] (breaking discrete parity symmetry), similar to phases recently discovered for $J_1, J_2 > 0$ [17, 18]. The chiral correlator

$$\kappa^2(r, d) = \langle [S_0 \times S_d]^\pi [S_r \times S_{r+d}]^\pi \rangle$$  \hspace{1cm} (2)

at maximal distance ($r = L/2$) – which is an order parameter for this phase – is shown in the left panel of Fig. 2 for $d = 1, 2$. It provides direct evidence for the existence of vector chiral order in the low magnetization region.

2.2. Multipolar Luttinger liquid phases
An exciting property of Luttinger liquids of $p$ bound magnon states is that the transverse spin correlations decay exponentially as a function of distance due to binding, while multipolar spin correlations of order $p$ are critical with wave vector $\pi$, i.e.,

$$M_p(r) = \langle \prod_{n=0}^{p-1} S_{0+n}^+ \prod_{n=0}^{p-1} S_{r+n}^- \rangle \sim (-1)^r \left( \frac{1}{r} \right)^{1/K}.$$  \hspace{1cm} (3)

In the shown window, at least five different phases are present. The low magnetization region consists of a single vector chiral phase (grey) with broken parity symmetry, long range vector chiral order, and incommensurate spin correlations. 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 $J_2 \to \infty$ [14], and its lower border approaches $m = 0^+$ in that limit. All three multipolar liquids present a crossover as a function of $m$, 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 [15, 10], which we did not aim to localize in this 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 with $p \geq 5$. 

**Figure 2.** Left panel: chiral order parameter $\kappa^2(r = L/2, d)$ [Eq. (2)] obtained from DMRG calculations on a 192-sites system. Black and red symbols correspond to $d = 1, 2$, respectively. The three chosen values of $J_2$ reflect positions underneath each of the three spin multipolar Luttinger liquids shown in Fig. 1. Right panel: Luttinger parameters $K$ for several magnetizations. For comparison, the result from Ref. [14] for $m = 3/4$ and $J_2 = -1$ is also shown (black). At $K = 1$, along the turquoise line, multipolar correlations of order $p$ [Eq. (3)] and longitudinal spin correlations [Eq. (4)] decay with the same exponent.
Multipolar correlations with \( p' < p \) also decay exponentially [14, 15]. They can therefore be considered as one-dimensional analogues of spin multipolar ordered phases found in higher dimensions. Another important correlation function is the longitudinal spin correlator

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

(4)

We determined the Luttinger parameter \( K \) as a function of \( m \) and \( J_2 \) by fits to the local \( S^z \) profiles in DMRG simulations, see right panel of Fig. 2. An important information is contained in the crossover line \( K = 1 \), where multipolar correlations of order \( p \) 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 towards the tip of the lobes the longitudinal spin correlations decay more slowly, characterizing a spin density wave [SDW(p)] phase.

3. Multipolar correlations in a magnetic state

3.1. Classical spiral state

We consider a classical spiral with spins of length \( S \), parametrized by

\[
S_j = S \begin{pmatrix} \cos(qj) \\ \sin(qj) \\ \sin(\vartheta) \end{pmatrix},
\]

(5)

with propagation vector \( q \) and canting angle \( \vartheta \) due to the magnetic field. The ground state of the Hamiltonian [Eq. (1)] is a ferromagnet for \( J_2 < 1/4 \). Otherwise, the energy is minimized for

\[
q = \arccos \left( \frac{|J_1|}{4J_2} \right) \in [0, \pi/2] \quad \text{and} \quad \vartheta = \arcsin \left( \frac{4hJ_2}{S(4J_2 - |J_1|)^2} \right) \in [0, \pi/2] .
\]

(6)

Using the substitutions \( S_j^+ \rightarrow iS_j^y \) and \( S_j^- \rightarrow S_j^x - iS_j^y \), the classical spiral displays vector chiral correlations of the form

\[
\langle [S_0 \times S_d]^- [S_r \times S_{d+r}]^- \rangle \rightarrow S^4 \cos^4(\vartheta) \sin^2(qd)
\]

(7)

and multipolar correlators of order \( p = 1 \) (spin, dipolar), \( p = 2 \) (quadrupolar), \( p = 3 \) (octupolar), and \( p = 4 \) (hexadecupolar)

\[
\begin{align*}
&S_0^+ S_r^- + \text{h.c.} = 2\langle S_0^x S_r^- \rangle + 2\langle S_0^y S_r^- \rangle \rightarrow 2S^2 \cos^2(\vartheta) \cos(qr) \\
&S_0^+ S_1^+ S_r^- S_{r+1}^- + \text{h.c.} \rightarrow 2S^4 \cos^4(\vartheta) \cos(2qr) \\
&S_0^+ S_1^+ S_2^+ S_r^- S_{r+1}^- S_{r+2}^- + \text{h.c.} \rightarrow 2S^6 \cos^6(\vartheta) \cos(3qr) \\
&S_0^+ S_1^+ S_2^+ S_3^+ S_r^- S_{r+1}^- S_{r+2}^- S_{r+3}^- + \text{h.c.} \rightarrow 2S^8 \cos^8(\vartheta) \cos(4qr) .
\end{align*}
\]

(8)

Note that if the in-plane spin correlations have wave vector \( q \), the in-plane multipolar correlations of order \( p \) propagate with wave vector \( q_p = pq \).

3.2. Quantum spirals and the UDRVB state

Let us now consider the effect of quantum fluctuations. At the Lifshitz point \( J_2 = 1/4 \), the singlet ground state is exactly known [12] to be the uniformly distributed resonating valence bond (UDRVB) state given by

\[
|\text{UDRVB} \rangle = \frac{1}{N} \sum [i,j][k,l][m,n] \cdots
\]
Figure 3. Left plot: Vector chiral (upper panel) and spin multipolar correlators (lower panel) of the UDRVB state for $L = 30$ from ED (full symbols) compared to selected analytical results [Eqs. (7),(8)]. The multipolar correlator of order $p$ has a wave vector $q_p = pq$ (periods are given on the bottom), $q$ being the wave vector of the UDRVB spiral. Right plot: Comparison of the von Neumann entropy of a block of $1 \leq l \leq L/2$ consecutive spins for the unfrustrated antiferromagnetic Heisenberg chain and the UDRVB state.

where $[i,j]$ denotes two sites $i$ and $j$ that are paired up in a singlet state. The summation runs over all combinations of ordered pairs of spins $(i < j, k < l, m < n, \ldots)$.

In the limit of large $L$, the spin correlations of the UDRVB state have been obtained analytically [12]

$$
\lim_{L \to \infty} \langle \text{UDRVB}|S_i^+ S_{i+r}^-|\text{UDRVB}\rangle = \frac{1}{6} \cos \frac{2\pi}{L} r ,
$$

suggesting that this state should be considered as a quantum analog of a long wavelength spiral with an $L$-dependent propagation vector $q = 2\pi/L$. To study this analogy in more detail, we have numerically computed the vector chiral correlations $\kappa^2(r,d)$ [Eq. (2)] and the spin multipolar correlations $M_p(r)$ [Eq. (3)] up to order $p = 4$ in the UDRVB state. These results are presented on the left panel of Fig. 3. For comparison, the exact result of Eq. (9) for $p = 1$ is drawn as well. Our aim is not to describe the higher order correlation functions of the UDRVB state quantitatively 2, but to point out that the simple relations between the wave vectors of multipolar correlation functions in a classical spiral state are also obeyed in the quantum analog, i.e. in the UDRVB state.

We first note that vector chiral correlations (Fig. 3, left plot, upper panel) of the UDRVB state are independent of the distance $r$, which – as shown by Eq. (7) – is in complete agreement with a spiral state. Furthermore, the multipolar correlations (Fig. 3, left plot, lower panel) indeed follow the phenomenology of the classical spiral, where the $p$-multipolar correlations have a wave vector $pq$ for a spiral with wave vector $q$. A quantitative comparison would be more involved due to the sizable finite size effects at distance $\sim L/2$, and the fact that the UDRVB state is a singlet, which requires a contraction of all components of the $SU(2)$ multipolar operators of rank $p$.

We close this section by presenting the peculiar entanglement entropy of the UDRVB state in comparison to the well studied unfrustrated antiferromagnetic Heisenberg chain (right plot of Fig. 3) for periodic systems. First of all, the UDRVB state has a much larger entanglement entropy for large blocks than the Heisenberg chain for the same block length [13]. Second, the

$^2$ Although this would be possible, it is somewhat too technical to be detailed here.
finite size behavior of the entanglement entropy for a given block size is non monotonous, again in contrast to the pure Heisenberg chain. This might be due to the fact that the wave vector \( q = 2\pi/L \) depends on the system size, so that the local structure of the spiral changes as \( L \) is increased. It would be interesting to derive the finite size scaling form of the entanglement entropy analytically for comparison.

4. Interpretation

Let us first investigate how the longitudinal and transverse equal-time spin structure factors \( S^{zz}(q) \) and \( S^{xx}(q) \) evolve as a function of the magnetization \( m \). In the left plot of Fig. 4 we display the location of the maximum of \( S^{zz}(q) \) and \( S^{xx}(q) \) (disregarding the \( q = 0 \) peak in \( S^{zz}(q) \) due to the total magnetization) for three representative \( J_2 \). At \( m = 0 \), it is known that \( S(q) \) has a maximum at an incommensurate position \( q_{\text{max}}(J_2) \), which is strongly renormalized compared to the classical expectation \( q_{\text{class}}^{\text{max}}(J_2) = \arccos[1/(4J_2)] \), see also lower right panel of Fig. 4. 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 \( q_{\text{max}} \) of \( S^{zz}(q) \) locks onto a straight line with slope \( -\pi/p \). It seems that if \( q_{\text{max}}(J_2)/\pi > 1/3 \) at \( m = 0 \), the magnetization process enters the \( p = 2 \) multipolar phase at larger \( m \). If instead \( 1/3 > q_{\text{max}}(J_2)/\pi > 1/4 \) at \( m = 0 \), the system will enter the \( p = 3 \) phase. Based on an extended analysis including many \( J_2 \), we are lead to conjecture that if

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

\( q_{\text{max}} \) locks onto the line with slope \( -\pi/p \) at higher magnetizations. According to the behavior of the longitudinal spin correlations in multipolar Luttinger liquids (Eq. 4), the multipolar liquid of order \( p \) leads precisely to a slope of \( -\pi/p \). Eqs. (3) and (10) now show that while \( q_{\text{max}}(J_2) \)
approximately equals $\pi/p$, the spin multipolar correlations of order $p$ are accurately \textit{locked} to $\pi$. Note that while the above considerations devoted to the classical spiral showed that many multipolar correlations are finite without actually selecting a specific one, in the frustrated ferromagnetic chain in a field, it is the additional locking mechanism $q_{\text{max}}(J_2)p \sim \pi$, which is responsible for the selection of $p$.

The idea that spin multipolar phases can result from instabilities of spiral ordering is rather appealing, and not limited to one dimension. For instance, for the Heisenberg model on the square lattice with ferromagnetic nearest neighbor couplings and antiferromagnetic next-nearest neighbor couplings (and possibly ring exchange) in a magnetic field, it has been shown by Shannon \textit{et al.} \cite{19} that a bond quadrupolar phase exists close to the ferromagnetic state. The bond quadrupolar order is explained by a condensation of two-magnon bound states from the ferromagnetic side. Our results provide a complementary view as a destabilization of the neighboring antiferromagnetic striped collinear phase with momentum $(\pi, 0)$, see Fig. 5. A similar process occurs in the multiple-spin exchange model on the triangular lattice, where an octupolar phase is surrounded by both a ferromagnetic and a canted antiferromagnetic spiral phase \cite{20}.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5.png}
\caption{Left plot: Illustration of a $(0, \pi)$ magnetically ordered spiral state (collinear stripe) on the square lattice. Right plot: Illustration of the bond quadrupolar expectation values in the collinear stripe state. The black and red colors denote positive and negative expectation values (the orientation is chosen for the convenience of presentation). This pattern gives rise to the bond quadrupolar correlation pattern in the nematic phase reported in Fig. 5 of Ref. \cite{19}.
\end{figure}

5. Conclusion
We have established the phase diagram of the frustrated ferromagnetic $S = 1/2$ Heisenberg chain in a uniform magnetic field. A vector chirally ordered state, metamagnetic behavior and a sequence of spin multipolar Luttinger liquid phases up to hexadecupolar order have been identified. We have shown that the peculiar valence bond state at the Lifshitz point $J_2 = 1/4$ can be regarded as the quantum analogue of the classical spiral ground state, in which multipolar correlations naturally occur. We have argued that above a certain magnetic field, spiral ordering is destabilized by quantum fluctuations, giving rise to higher order multipolar spin correlations. This point of view is consistent with existing phase diagrams exhibiting such phases. It might therefore be used to predict multipolar phases in models with some form of spiral ordering. It is an open question whether these fluctuation driven multipolar phases and the locking mechanism also appear for $S > 1/2$. 
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