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Phys. Rev. B **92**, 140403 — Published 2 October 2015

DOI: 10.1103/PhysRevB.92.140403

Competing Spin Liquid States in the Spin-$1/2$ Heisenberg Model On Triangular Lattice

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We study the spin-$1/2$ Heisenberg model on the triangular lattice with the antiferromagnetic first (J_1) and second (J_2) nearest-neighbor interactions using density matrix renormalization group. By studying the spin correlation function, we find a 120° magnetic order phase for $J_2 \lesssim 0.07J_1$ and a stripe antiferromagnetic phase for $J_2 \gtrsim 0.15J_1$. Between these two phases, we identify a spin liquid region characterized by the exponential decaying spin and dimer correlations, as well as the large spin singlet and triplet excitation gaps on finite-size systems. We find two near degenerating ground states with distinct properties in two sectors, which indicates more than one spin liquid candidates in this region. While the sector with spinon is found to respect the time reversal symmetry, the even sector without a spinon breaks such a symmetry for finite-size systems. Furthermore, we detect the signature of the fractionalization by following the evolution of different ground states with inserting spin flux into the cylinder system. Moreover, by tuning the anisotropic bond coupling, we explore the nature of the spin liquid phase and find the optimal parameter region for the gapped Z_2 spin liquid.

PACS numbers: 73.43.Nq, 75.10.Jm, 75.10.Kt

Quantum spin liquids (SLs) are long-range entangled states with remarkable properties of fundamental importance [1]. The SL physics has been considered to be essential to understand strongly correlated systems and unconventional superconductivity [2, 3]. The simplest and perhaps most striking SLs are the gapped topological SLs, which develop a topological order [4–6] with the emergent fractionalized quasiparticles [7–9]. Although SLs have been studied intensively for two decades and demonstrated in contrived models [10–20], the microscopic condition for the emergence of SLs in frustrated magnetic systems is not well understood.

At the experimental side, possible SLs have been discovered in various materials. Among these materials, the most promising systems are the kagome antiferromangets including the Herbertsmithite and Kapellasite [21–25], as well as the organic Mott insulators with a triangular lattice structure such as κ-$(ET)_2Cu_2(CN)_3$ [26–29] and $EtMe_3Sb[Pd(dmit)_2]_2$ [30, 31]. In all these materials, no magnetic order is observed at the temperature much lower than the interaction energy scale. These experimental findings have inspired intensive theoretical studies on the frustrated magnetic systems.

Theoretically, the kagome Heisenberg model appears to possess a robust SL. Density matrix renormalization group (DMRG) studies suggest a gapped Z_2 SL [32–35]. Variational studies based on the projected fermionic parton wave functions however favor a gapless Dirac SL [36–38]. Interestingly, by introducing the second and third neighbor couplings [39–41] or the chiral interactions [42], DMRG [40–42] studies recently discovered another topological SL — chiral spin liquid (CSL) [43, 44], which breaks time reversal symmetry (TRS) spontaneously and is identified as the $\nu = 1/2$ bosonic fractional quantum Hall state. On the other hand, the non-magnetic phases in the frustrated honeycomb and square J_1-J_2 models appear to be conventional valence-bond solid state [45–48].

The spin-$1/2$ triangular nearest-neighbor antiferromagnetic (AF) Heisenberg model was the first candidate proposed to realize a SL [2], although it turns out to exhibit a 120° AF order [49–53]. To understand the triangular weak Mott insulator materials, combined theoretical and numerical studies [54–56] on a spin model with four-site ring-exchange couplings [57] find a gapless *spin bose metal*. To enhance frustration [58–68], one way is to include the second-neighbor coupling J_2, where a stripe ordered state is found with larger J_2 coupling [58, 59], and an intermediate non-magnetic region may emerge [60–63]. The variational Monte Carlo simulations find a nodal d-wave SL [61] and a gapless SL [62] as the candidates for this intermediate phase. Very recently, a DMRG work [69] found the indication of a gapped SL with TRS in the non-magnetic phase. However, the nature of this intermediate phase remains far from clear.

In this Letter, we study the spin-$1/2$ triangular model with the AF first and second nearest-neighbor $J_1(J_1')$-J_2 couplings using DMRG. The Hamiltonian is given as

$$H = J_1 \sum_{\langle i,j\rangle_{\rm vertical}} \vec{S}_i\cdot\vec{S}_j + J_1' \sum_{\langle i,j\rangle_{\rm zigzag}} \vec{S}_i\cdot\vec{S}_j + J_2 \sum_{\langle\langle i,j\rangle\rangle} \vec{S}_i\cdot\vec{S}_j,$$

where the sums $\langle i,j\rangle$ and $\langle\langle i,j\rangle\rangle$ run over all the first- and second-neighbor bonds, respectively. The first-neighbor couplings J_1 and J_1' are for the vertical and zigzag bonds as shown in Fig. 1(a). We study most systems with $J_1' = J_1$ unless we specify otherwise. We set $J_1 = 1$ as energy scale. By studying spin correlations, we find a non-magnetic region sandwiched by a 120° AF phase with three sublattices for $J_2 \lesssim 0.07$ and a stripe AF phase for $J_2 \gtrsim 0.15$ as shown in Fig. 1. In this non-magnetic region, we identify two ground states with distinct properties in two sectors, indicating two competing candidates for SL phases. The spin and dimer correlations decay exponentially with small correlation lengths. Interestingly, the chiral correlations decay exponentially fast in the odd sector with an edge spinon, while it develops the long-range correlations in the even sector (with no spinon) for finite-size systems, consistent with the level crossing between two SLs for the systems with different boundaries. The fractionalized spinon is detected through adiabatically inserting spin flux. While the state in the odd sector agrees with a TRS

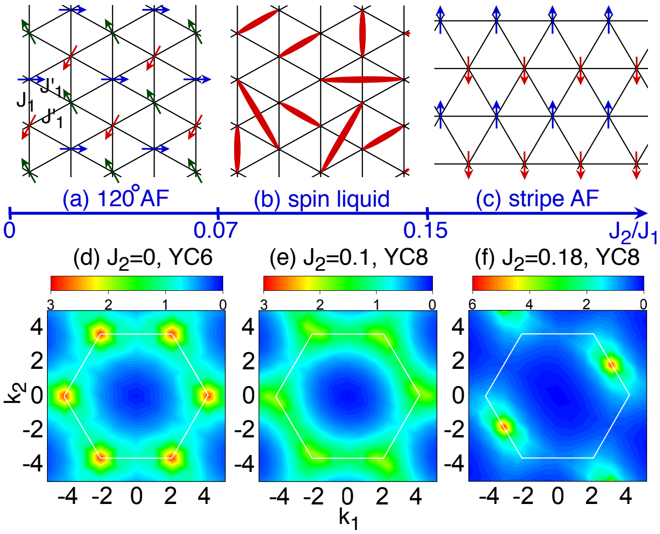

FIG. 1: (color online) Quantum phase diagram of the isotropic spin-$1/2$ J_1-J_2 Heisenberg model on triangular lattice ($J_1 = J_1'$). With growing J_2, the system has a 120° AF phase for $J_2 \lesssim 0.07$, a stripe AF phase for $J_2 \gtrsim 0.15$, and a SL phase in between. The schematic figures of the different phases also show the YC (a,b) and XC (c) cylinder geometries. (d)-(f) are the contour plots of spin structure factor for each phase.

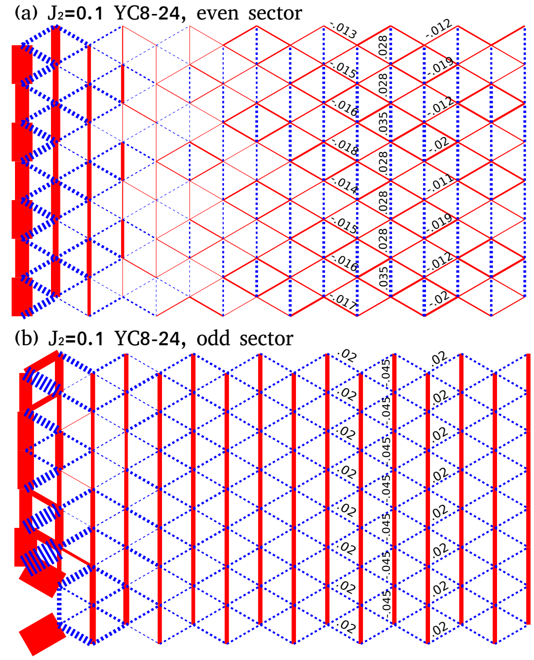

FIG. 2: (color online) The NN bond energy $\langle S_i \cdot S_j \rangle$ for $J_2 = 0.1$ on the YC8-24 cylinder in (a) the even and (b) the odd sector. The left 16 columns are shown here. The odd sector is obtained by removing one site in each boundary of cylinder. In both figures, all the bond energy have subtracted the average value -0.18. The red solid and blue dashed bonds denote the negative and positive bond energies after subtraction (with some numbers shown for clarity).

preserving SL, the TRS breaking SL (e.g., CSL) may be a competing or nearby state in more extended parameter space. Moreover, the strong bond anisotropy is observed for some finite-size systems, which may imply a nematic Z_2 SL [70]. This possible Z_2 SL is observed to be stabilized by a small bond coupling anisotropy ($J_1' \gtrsim J_1$), which suppresses chiral order in both sectors.

We study the cylinder systems using accurate $SU(2)$ DMRG [71, 72] for most of calculations and $U(1)$ DMRG [71] for inserting flux [40]. Two cylinder geometries known as XC and YC are studied, which have one lattice direction parallel to the x or y axis as shown in Fig. 1. We denote them as XCL_y-L_x (YCL_y-L_x), where L_y and L_x are the number of sites along the y and x directions, respectively. We study the cylinder systems with L_y up to 10 lattice spacings by keeping up to 20000 $U(1)$-equivalent states in $SU(2)$ DMRG and 5000 states for inserting flux. The truncation errors are less than 10^{-5} in all calculations, which leads to accurate results.

Even and odd topological sectors.— Based on the resonating valence-bond picture, the ground states of SL on cylinder can be either in the even or odd sector according to the parity of the number of bonds cut by a vertical line along the enclosed direction. Usually, the odd sector can be obtained by removing or adding one site on each open edges, which has been used successfully to find different sectors of the gapped SLs in kagome systems [33, 73, 74].

By tuning boundary condition, we always find two different sectors on YC cylinders ($L_y = 6, 8, 10$), which are shown in Fig. 2 for YC8 cylinder as an example (See Supplemental Material for YC10 cylinder) [75]. We find that the two sectors have the different bond energy distributions in the bulk of cylinder. While the vertical bonds are weaker in the even sector, they become the stronger ones in the odd sector. The nematic order, which is defined as the difference between the strong and weak bonds exhibits the distinct behaviors for the two states on studied systems. While the nematic order grows with increased cylinder width in the odd sector, it decreases in the even sector [75].

Characteristic properties of different SL states.— Next, we further characterize the two states by studying correlation functions. In Fig. 3, we show the spin correlations and dimer correlations $D_{(ij),(kl)} = \langle (S_i \cdot S_j)(S_k \cdot S_l) \rangle - \langle S_i \cdot S_j \rangle \langle S_k \cdot S_l \rangle$ for $J_2 = 0.1$, which all decay faster with growing cylinder width in both states, indicating the vanishing orders in both states. Interestingly, the states in the odd sector have very short correlation lengths almost independent of the system width. But in the even sector, the correlation lengths are longer than those in the odd sector for the smaller $L_y = 6$ and 8, which decrease with growing system width.

To study possible TRS breaking, we calculate the scalar chiral correlation function $\langle \chi_i \chi_j \rangle$ ($\chi_i = (S_{i,1} \times S_{i,2}) \cdot S_{i,3}$). In both sectors, the chiral order in the up and down triangles has the same direction. As shown in Fig. 3(c), in the odd sector, the chiral correlations decay quite fast to vanish. However, the chiral order and spontaneous TRS breaking are robust for the $L_y = 6, 8$ systems in the even sector at the intermediate

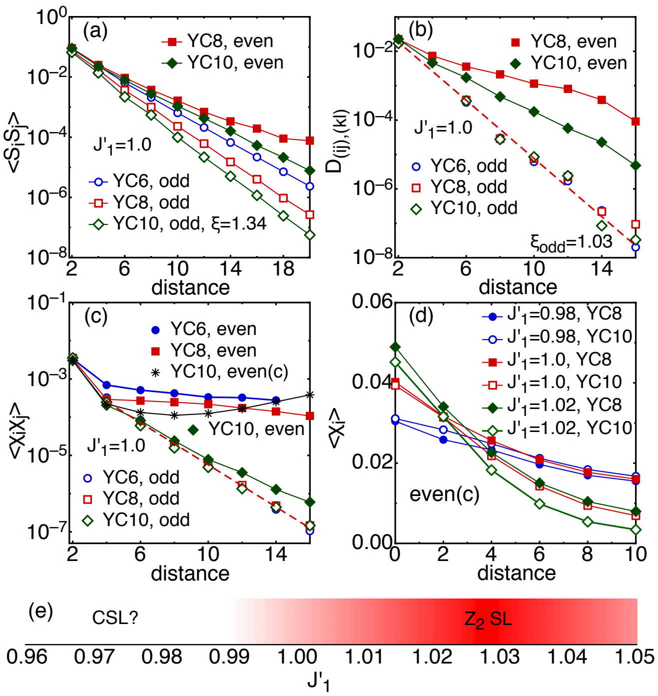

FIG. 3: (color online) (a) and (b) are the spin and dimer correlation functions on different YC cylinders for $J_2 = 0.1$. ξ denotes the corresponding correlation lengths on YC10 cylinder in the odd sector. (c) Chiral correlations for $J_2 = 0.1$ on different cylinders. All the data are obtained from real number DMRG calculations except "YC10, even(c)", which denotes the data obtained from complex wave function DMRG calculations on YC10 cylinder in the even sector. (d) Chiral order from the boundary to the bulk for the bond anisotropic system in the even sector, which are obtained from the complex DMRG. (e) Schematic phase diagram for the bond anisotropic system at $J_2 = 0.1$.

J_2,YCL_y	$e_{\rm even}$	$e_{\rm odd}$	Δe	$\Delta_{\rm T}$	$\Delta_{\rm S}$
0.1,YC6	−0.5155	−0.5210	0.0055	0.365	0.30
0.1,YC8	−0.5171	−0.5195	0.0024	0.335	0.26
0.1,YC10	−0.5181(2)	−0.5177	−0.0004(2)	0.30(1)	0.18
0.125,YC6	−0.5104	−0.5145	0.0041	0.389	0.33
0.125,YC8	−0.5115	−0.5133	0.0018	0.343	0.22
0.125,YC10	−0.5119(2)	−0.5120	0.0001(2)	0.30(1)	0.15

TABLE I: The bulk energy per site in the even ($e_{\rm even}$) and odd ($e_{\rm odd}$) sectors, the energy difference $\Delta e = e_{\rm even} - e_{\rm odd}$, the spin triplet ($\Delta_{\rm T}$) and singlet ($\Delta_{\rm S}$) gaps in the odd sector for $J_2 = 0.1$ and 0.125 on the YCL_y ($L_y = 6, 8, 10$) cylinders. We use the fully converged results for $L_y = 6$ and 8, and the extrapolated results for $L_y = 10$ as shown in Fig. 4.

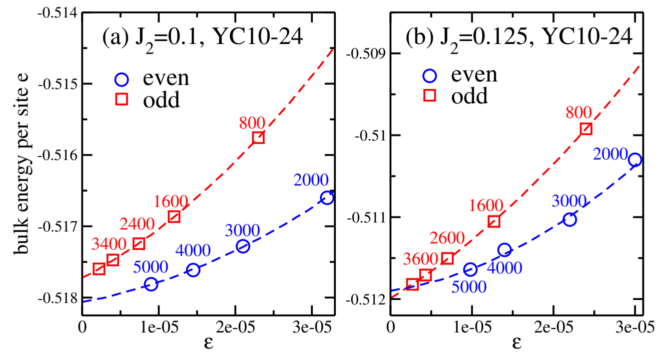

FIG. 4: (color online) Bulk energy per site e versus DMRG truncation error ε for the ground states in the even and odd sectors for (a) $J_2 = 0.1$ and (b) $J_2 = 0.125$ on the YC10-24 cylinder. The numbers denote the kept $SU(2)$ states. The energy data are fitted using the formula $e(\varepsilon) = e(0) + a\varepsilon + b\varepsilon^2$.

phase. As we increase system width to $L_y = 10$, the chiral correlation becomes less robust, where different results are obtained depending on if we use complex or real initial wave function in DMRG simulation. The chiral correlation remains to be long-ranged in the complex wave function. However, if we use real number wave function, the DMRG will find a state with short-range chiral correlations (the real state has near identical bulk energy as the complex state but higher energy near the edge).

To further clarify the chiral order, we consider a bond anisotropy perturbation by tuning the nearest-neighbor zigzag bond as J_1' (see Fig. 1(a)). For $J_2 = 0.1$, we find the SL persisting for $0.95 < J_1' < 1.05$. In the odd sector, all the properties are consistent with $J_1' = 1.0$. In the even sector, the chiral order appears stronger for $0.95 < J_1' \lesssim 0.99$, where it grows a bit from YC8 to YC10 cylinder (see Fig. 3(d)). For $J_1' \gtrsim 1.0$, the chiral order decays pretty fast from the boundary to the bulk especially for the larger YC10 cylinder, which indicates a possible vanishing of the chiral order in the thermodynamic limit. At $J_1' = 1.0$, the chiral correlations are strong and show long-range behavior, but the chiral order also decays with increasing system width. Thus, the two states may recover TRS at large system limit for $1.0 \lesssim J_1' < 1.05$, which is the most possible region for stabilizing a Z_2 SL. On the other hand, for $0.95 < J_1' \lesssim 0.99$, chiral order becomes stable in the even sector, while the fate of such a phase remains unclear depending on if the chiral order would develop in the spinon sector in the thermodynamic limit. We illustrate our finding in the phase diagram Fig. 3(e).

We calculate the bulk ground-state energy and gaps in both sectors, which are shown in Table I for $J_2 = 0.1, 0.125$. For the smaller system widths ($L_y = 6$ and 8), the odd sectors have the lower energy than the even sectors, leading to a positive energy splitting $\Delta e = e_{\rm even} - e_{\rm odd}$. However, this splitting drops fast with increasing L_y, which is tiny for $L_y = 10$, indicating the close energy in both sectors. We also compute the singlet Δ_s and triplet Δ_T gaps by obtaining the ground state first and then sweeping the two low-lying states simultaneously or the $S = 1$ sector in the bulk of cylinder [67].

Inserting flux and the nature of different sectors.— Inserting flux is an effective way to identify topological properties, which has been used in DMRG to study topological SLs [40, 73, 74]. To introduce a flux, we impose the twist boundary condition in the y direction by replacing terms $S_i^+ S_j^- + h.c. \rightarrow S_i^+ S_j^- e^{i\theta} + h.c.$ for all neighboring bonds

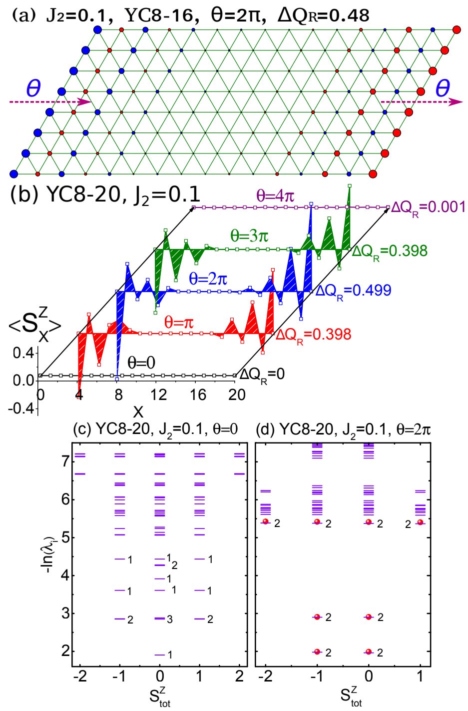

FIG. 5: (color online) (a) Real-space configuration of spin magnetization $\langle S^z_i \rangle$ after adiabatically inserting a flux $\theta = 2\pi$. The red and blue circles denote the positive and negative $\langle S^z_i \rangle$ with the area of circle proportional to the amplitude of $\langle S^z_i \rangle$. $\Delta Q_{\rm R}$ is the net spin accumulation on the right edge $\Delta Q_{\rm R} = \sum_i \langle S^z_i \rangle$, where i denotes the site on the right edge of the cylinder. (b) Real-space configuration of the accumulated spin magnetization in each column with increasing flux. Low-lying ES at the flux (c) $\theta = 0$ and (d) $\theta = 2\pi$. The numbers in ES denote the near-degenerate eigenvalues in the large weight levels. At $\theta = 2\pi$ in (d), all the eigenvalues are double degenerate as explicitly shown by the circles for the low-lying levels.

crossing y-boundary. We start from the even sector by adiabatically increasing θ and measuring the evolution of the spin-z local magnetization $\langle S^z_i \rangle$. With increasing θ, a net spin-z accumulates on the open edges as shown in Fig. 5(a), indicating that the quasiparticle responding to the flux here must carry spin, such as the spinon in CSL [40] and the fermionic spinon (spinon bonded with vison) in Z_2 SL [73, 76, 77].

With the flux $\theta = 0 \to 2\pi$, the net spin grows continuously from 0 to 0.5 on the edges as shown in Fig. 5(b). At $\theta = 2\pi$, an $S^z = \pm 1/2$ spinon develops on each boundary, and the ground state evolves to a new sector. By further increasing $\theta = 2\pi \to 4\pi$, the net spin dissipates continuously to zero, and the system evolves back to the even sector. In Figs. 5(c) and 5(d), we demonstrate the entanglement spectrum (ES) with inserting flux. At $\theta = 0$ in the even sector, the whole ES is symmetric about $S^z = 0$. At $\theta = 2\pi$, the ES becomes symmetric about $S^z = 1/2$, consistent with the fractionalized spin-carrying quasiparticles on boundaries. Interestingly, each eigenvalue of the ES at $\theta = 2\pi$ is doubly degenerate. By comparing the odd sector obtained by removing sites and the sector with flux $\theta = 2\pi$ here, we find that these two states have the same bulk energy and ES, indicating that the two sectors obtained by different methods are exactly the same. This odd sector might be consistent with the fermionic spinon sector of the Z_2 SL [76–78].

Summary and Discussions.— By means of DMRG calculations on wide cylinder systems of the spin-1/2 J_1-J_2 Heisenberg model on the triangular lattice, we find a SL region bordered by a 120° Néel AF phase for $J_2 \lesssim 0.07$ and a stripe AF phase for $J_2 \gtrsim 0.15$. The spin and dimer correlations all decay fast for wider systems with small correlation lengths comparable to lattice constant with large spin and singlet excitation gaps. The ES in the odd sector could be consistent with the fermionic spinon in Z_2 SL. However, the long-range chiral correlation is observed in the even sector for a space isotropic model. By tuning the anisotropic bond coupling, we find that the possible gapped Z_2 SL is stabilized by some weak anisotropy ($J_1' \sim 1.02$). The chiral correlations are enhanced in the even sector by the opposite anisotropy ($J_1' \sim 0.98$), which may be stabilized in both sectors by TRS breaking terms and we leave this open issue for future studies.

We acknowledge the stimulating discussions with T. Senthil, X. G. Wen, Z. Y. Zhu, S. R. White, F. Wang, and Y. Qi. This research is supported by the National Science Foundation through grants PREM DMR-1205734 (W.J.Hu) and DMR-1408560 (S.S.G., D.N.S.), and the U.S. Department of Energy, Office of Basic Energy Sciences under grant No. DE-FG02-06ER46305 (W.Z.).

Note added.—While completing this work, we became aware of some related papers [69, 79]. We reached the similar conclusion on gapped SL with Ref. [69].

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