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Phys. Rev. B **106**, 094420 — Published 16 September 2022

DOI: [10.1103/PhysRevB.106.094420](https://doi.org/10.1103/PhysRevB.106.094420)

Quantum Spin Liquid with Emergent Chiral Order in the Triangular-lattice Hubbard Model

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(Dated: June 30, 2022)

The interplay between spin frustration and charge fluctuation gives rise to an exotic quantum state in the intermediate-interaction regime of the half-filled triangular-lattice Hubbard model, while the nature of the state is under debate. Using the density matrix renormalization group with $SU(2)_{\text{spin}} \otimes U(1)_{\text{charge}}$ symmetries implemented, we study the triangular-lattice Hubbard model defined on the long cylinder geometry up to circumference $W = 6$. A gapped quantum spin liquid, with on-site interaction $9 \lesssim U/t \lesssim 10.75$, is identified between the metallic and the antiferromagnetic Mott insulating phases. In particular, we find that this spin liquid develops a robust long-range spin scalar-chiral correlation as the system length L increases, which unambiguously unveils the spontaneous time-reversal symmetry breaking. In addition, the degeneracy of the entanglement spectrum supports symmetry fractionalization and spinon edge modes in the obtained ground state. The possible origin of chiral order in this intermediate spin liquid and its relation to the rotonlike excitations have also been discussed.

I. Introduction

Since Anderson's seminal work of the resonating valence bond (RVB) state in quantum antiferromagnets [1, 2], searching for spin liquid states and the consequent superconductivity after doping, constitutes an exciting topic in condensed matter physics [3]. While it has been widely accepted that spin frustration plays the key role for the emergence of spin liquid in Mott insulators [4–6], it has also been noticed that the strong charge fluctuations near the Mott transition may add an additional active ingredient to the system [7, 8]. Although stable spin liquid states in the half-filled bipartite-lattice Hubbard models have not been established [9], the frustrated triangular-lattice Hubbard (TLU) model, harboring stronger spin and charge fluctuations at intermediate Hubbard interaction U , has raised great interests in the possible intermediate spin liquid state [10–16].

Meanwhile, the experimental progresses on the triangular-lattice organic-salt compounds κ -(BEDT-TTF)₂Cu₂(CN)₃ [17–22] and EtMe₃Sb[Pd(dmit)₂]₂ [23, 24] also shed light on the spin-liquid states near the Mott transition. The absence of spin ordering down to the lowest experimental temperature and the linear- T dependence of low-temperature specific heat suggest a possible gapless spin liquid in these compounds [19, 24]. However, recent thermal conductivity measurements indicate the absence of mobile gapless excitations [25, 26]. The experimental identification of the spin liquid and the pursuit of its nature have further stimulated intensive theoretical studies.

To include the charge fluctuation effects, one can consider the higher-order ring-exchange coupling in the effective spin model [7, 27–30] or simulate the Hubbard model directly. Indeed, numerical simulations on the ring-exchange spin model have identified a gapless spin liquid state with the emergent spinon Fermi surface [7, 28, 31], which can partly explain the experimental findings. On the other hand, large-scale density matrix renormalization group (DMRG) simulations on the TLU itself have uncovered a spin liquid phase near the Mott transition [14, 15]. However, the two different studies lead to drastically distinct conclusions on the nature of this spin liquid. While the finite-DMRG calculation [14] suggests a Dirac-like gapless spin liquid preserving time reversal symmetry (TRS), the more extensive infinite-DMRG study [15] finds a gapped chiral spin liquid (CSL) with finite chiral order [32–35]. Moreover, the spinon Fermi-surface state is not found in these DMRG simulations, in contrast to the previous understanding based on the effective spin model.

In this work, we further determine the precise nature of the spin liquid phase in the TLU. After introducing the model and method in Sec. II, we perform extensive DMRG calculations on finite-size cylinders for fixed width $W = 4$ (YC4; cf. Fig. 1) in Sec. III where we gradually increase the system length up to $L = 64$. This goes far beyond the previous finite-size DMRG [14] and thus significantly reduces finite-size effects, which is followed by an analysis of width $W = 6$ cylinders (YC6) in Sec. III E, with strong evidence also for a chiral phase there, albeit with slightly altered phase boundaries. Throughout, we emphasize the necessity to exploit the $SU(2)_{\text{spin}} \otimes U(1)_{\text{charge}}$ symmetries in our DMRG simulation, as this permits us to reliably reach large-scale systems. We identify an intermediate non-magnetic phase with ultrashort single-particle and spin correlation lengths on the order of one lattice spacing, having $(U_{c1} \approx 9t) \lesssim U \lesssim (U_{c2} \approx 10.75t)$ for $W = 4$. On short cylinders we find exponentially decaying chiral correla-

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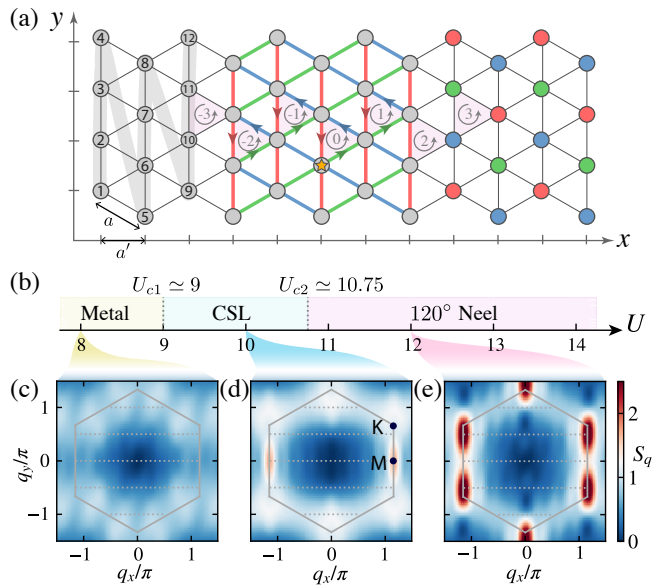


FIG. 1. Model and phase diagram. (a) Triangular-lattice cylinder with open (periodic) boundary conditions along the x (y) direction and lattice spacing $a = 1$. Having a straight open edge, i.e., YC4 geometry, the columnar spacing is $a' = \sqrt{3}/2$. (For later reference, an XC geometry has straight horizontal chains at distance a' with a zigzag open boundary left and right). The DMRG simulations employ a 1D “zigzag” mapping as indicated by the grey-shaded path (with part of the site ordering also indicated). The yellow star denotes the central reference site to which the spin-spin correlation is calculated, while the pink triangles for evaluating the chiral correlations are ordered symmetrically away from the center. The arrows on the bonds denote the current directions of the chiral order, having three colors for the three different directions. (b) Phase diagram of the TLU model consists of a metallic, a fully gapped CSL, and a 120° spin-ordered phases, with the three colors of the sites denoting the 3-sublattice structure. (c-e) Typical static spin structure factors S_q for $U = 8, 10$, and 12 in the three phases.

tion in agreement with Ref. [14], but the result changes fundamentally with increasing system length, showing very robust long-range chiral correlation characterizing spontaneous TRS breaking. We also find a large degeneracy in the entanglement spectrum, which agrees with symmetry fractionalization and the existence of edge spinon in the obtained ground state. Therefore, we conclude that the low-energy physics of the TLU model at the intermediate- U is governed by a gapped CSL. Our results show the importance of a sufficiently large system length to overcome finite-size effects for identifying TRS breaking in this system. In the outlook in Sec. IV we point out a possible link of the present results to the chiral nature of excitations found in the triangular lattice Heisenberg (TLH) model at finite temperature [36].

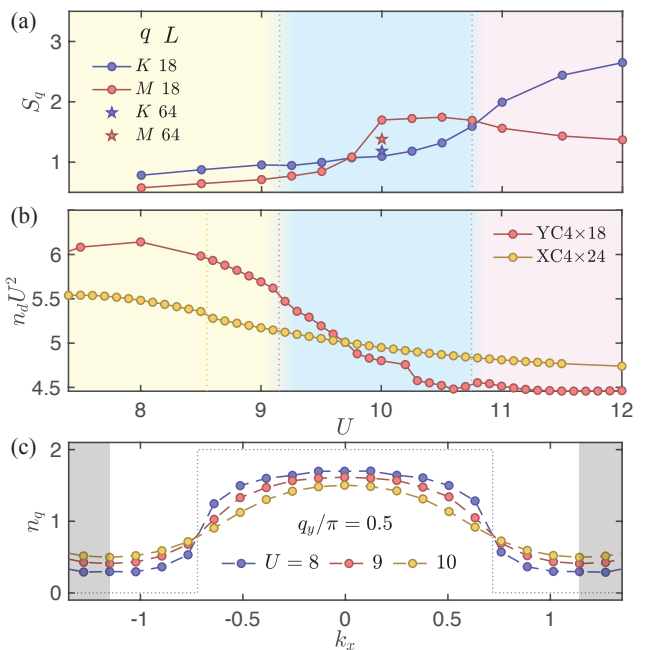


FIG. 2. Hubbard U dependence of different quantities on the YC4 \times L cylinders. (a) Spin structure factors S_q at the K and M points. The $L = 64$ data are also shown at $U = 10$. The asterisks show data points for the significantly longer system that exhibits long-range chiral correlations as analyzed in Fig. 4. (b) Double occupancy shown as $n_d U^2$. This also includes XC4 data which exhibits only one discontinuity around the metal-insulator transition. (c) Distribution of n_q vs. q_x in the momentum space along $q_y = \pi/2$. The dashed line plots n_q for $U = 0$ on the torus system with the same size 4×18 . The grey region marks the momenta outside the Brillouin zone.

II. Model and Method

The TLU model is defined as

$$\hat{\mathcal{H}} = -t \sum_{\langle i,j \rangle, \sigma} (\hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma} + \text{H.c.}) + \frac{U}{2} (\hat{n}_i - 1)^2, \quad (1)$$

where $\langle \cdot, \cdot \rangle$ represents the summation over the nearest-neighbor (NN) couplings, $\hat{c}_{i\sigma}$ ($\hat{c}_{i\sigma}^\dagger$) denotes the fermionic annihilation (creation) operator with spin $\sigma \in \{\uparrow, \downarrow\}$ on site i , and $\hat{n}_i = \sum_{\sigma} \hat{n}_{i\sigma}$ with $\hat{n}_{i\sigma} \equiv \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma}$ the particle number operator. We set $t := 1$ as the unit of energy, and the unit of distance via the lattice spacing $a := 1$, throughout. In our DMRG calculations [37], we mainly focus on YC4 cylinders as shown in Fig. 1(a), but also extend to XC4 and YC6 cylinders. Throughout, we implement the $SU(2)_{\text{spin}} \otimes U(1)_{\text{charge}}$ symmetries based on the QSpace tensor library [38, 39] which enables us to retain up to $D^* = 8192$ multiplets [equivalent to about $D \sim 24000$ $U(1)$ states] and ensures full convergence with a truncation error $\leq 1 \times 10^{-6}$.

III. Width 4 Cylinders (YC4)

A. Ground-state phase diagram

We summarize our DMRG phase diagram in Fig. 1(b), with a metallic phase for $U < U_{c1} (\approx 9)$, a 120° magnetically coplanar-ordered phase [40–42] for $U > U_{c2} (\approx 10.75)$, and an intermediate CSL phase with spontaneous TRS breaking. Fig. 1(c-e) show the representative snapshots of the static spin structure factors $S_q \equiv \sum_j e^{iq \cdot R_{0j}} \langle \hat{S}_j \cdot \hat{S}_0 \rangle$ in the different phases where site 0 refers to a fixed site in the center of the system [c.f., the asterisk site in Fig. 1(a)]. In the metallic phase (small U) S_q is found to be featureless. In the CSL phase [Fig. 1(d)] a peak emerges at the M point which is related to short-ranged stripe correlation. Further increasing U , sharp peaks emerge around the K points [Fig. 1(e)], consistent with a semiclassical 120° spin order. Even though the present YC4 geometry is not fully compatible with the 120° order, the feature of the dominant K point peak can still be observed. For very large $U \gtrsim 20$, eventually, the ground state on the particular YC4 cylinder switches to an RVB ring-like state [43] which is outside the window of interest here.

The phase boundaries of the CSL phase are estimated in Fig. 2. By contrasting the U dependence of S_q at $q = K$ with M in Fig. 2(a) [44], strongly enhanced magnetic correlations at the M points appears in the intermediate regime, up to $L = 64$. For $L = 18$, this leads to two crossing points of S_M with S_K . We use the upper crossing to estimate the phase boundary towards the 120° order, resulting in $U_{c2} \approx 10.75$. The lower phase boundary towards the metallic phase represents a metal-insulator transition, that is more naturally characterized by an analysis of the double occupancy $n_d \equiv \langle \psi | \hat{n}_{0\uparrow} \hat{n}_{0\downarrow} | \psi \rangle$, which is related to local charge or energy fluctuations of a single site (computed at the central site $i = 0$ here). Starting from half-filling, this expectation value acquires a finite value for $U \gg t$ via a second order process. In the infinite- U limit one can thus estimate for the magnetically completely uncorrelated case, $n_d \approx \frac{z^2}{2U^2}$ with z the coordination number. Antiferromagnetic correlations tend to increase this value. Also as U is lowered, charge fluctuations generally increase via higher-order processes. Hence in the present case with $z = 6$ and $t = 1$, we can consider $n_d U^2 \gtrsim 3$ a lower bound. Also from a numerical perspective with $U \sim 10$, we have for the double occupancy $n_d \ll 1$, such that accurate numerical simulations are important. Now when plotting $n_d U^2$ vs. U for the YC4 system [Fig. 2(b)] we observe the onset of a kink around $U \approx 9$ which we thus interpret as the lower phase boundary U_{c1} . Note, however, that this value for U_{c1} does not coincide with the lower crossing of S_M vs. S_K in Fig. 2(a) which may be owing to the finite-size effects, considering that the lower phase boundary is significantly more demanding numerically given that both, spin and charge gaps close there. For YC4, the double occupancy $n_d U^2$ shows another discontinuity around the magnetic transition U_{c2} , and stays rather constant thereafter. For comparison, we also include data for the XC4 geometry in Fig. 2(b). In contrast to YC4, this only exhibits one discontinuity around U_{c1} . Overall, it is smoother, varies significantly less vs. U , and remains

higher for the largest U . Together with the fact that 120° order is commensurate with XC4, we take the above finding as an indication that the intermediate chiral phase is absent for the XC4 geometry. As such, it exhibits a lesser degree of frustration and favors the large- U magnetic correlations already at smaller U . As an aside, we note that the interpretation that frustration is less pronounced for XC4 is also supported by [42] where no dimerization was observed in the Heisenberg limit.

The metal-insulator transition also manifests itself in the change of the Fermi surface with increasing U . For this, we analyze the one-particle charge density in momentum space $n_q \equiv \sum_{\sigma} \sum_j e^{iq \cdot R_{0j}} \langle \hat{c}_{j\sigma}^\dagger \hat{c}_{0\sigma} \rangle$ in Fig. 2(c). At $U = 0$, n_q is a step-like function (dashed line). With increasing U , the drop of n_q is gradually smoothed until the Fermi surface disappears [15]. To be specific, within the resolution for $L = 18$, n_q at $U = 8$ still exhibits an appreciable “jump” at $q_x \approx \pm 3\pi/4$ with fixed $q_y = \pi/2$. However, for $U \gtrsim 9$, n_q changes smoothly and the Fermi surface appears to be absent (cf. App. D), indicating a metal-insulator transition. Our estimates of the phase boundaries are roughly consistent with those reported in previous studies [13–15, 29, 45]. Minor quantitative deviation of the lower boundary U_{c1} is likely due to the different geometry and system size.

B. Fully gapped spin liquid

Next we focus on the charge and spin excitations in the spin liquid phase. In Fig. 3(a), the single-particle Green’s function $\langle \hat{c}_i^\dagger \hat{c}_j \rangle$, averaged amongst all site pairs with the same distance d_{ij} , decays exponentially versus d_{ij} with a short correlation length $\xi_c \approx 0.81$. This is consistent with a sizable charge (single-particle excitation) gap $\Delta_c \approx 0.88$ extrapolated for $1/L \rightarrow 0$ (see App. G) and verifies that the spin liquid resides in the Mott insulator phase.

In Fig. 3(b), we show the spin correlation $\langle \hat{S}_i \cdot \hat{S}_j \rangle$ versus d_{ij} , which is well converged and clearly decays exponentially, with a short correlation length $\xi_s \approx 1.66$. Such a short correlation length implies gapped spin excitations, which do not support a spinon Fermi surface state with algebraically decaying spin correlation [7] but could be consistent with either a gapped spin liquid or a Dirac-like gapless spin liquid that is gapped due to finite size on narrow-width cylinders. As shown in Fig. 3(c), this conclusion is further supported by the saturated bipartite entanglement entropy S_E vs. subblock size l_x , following an area law [46].

By analyzing the convergence of the DMRG simulation with increasing D^* (see App. B and G for details), we find that the chiral correlations become well-established only once the accuracy of the energy per site reaches a resolution of $\Delta e_\chi \equiv e_g - e_g^0 \approx 10^{-3}$ for YC4 \times 64 [e.g., see Fig. A2(h), where the kink in the convergence of the ground state energy around $1/D^* \sim 5 \cdot 10^{-4}$ with $\delta e_g \approx 10^{-3}$ relates to a significant build-up of entanglement entropy in Fig. A2(g) still, before it converges to a plateau]. Based on this, one may estimate a sizable bulk gap $\Delta_\chi \approx N \cdot \Delta e_\chi \sim 0.26$ with the chiral phase, with $N \equiv LW$ the total number of sites. Nevertheless, since

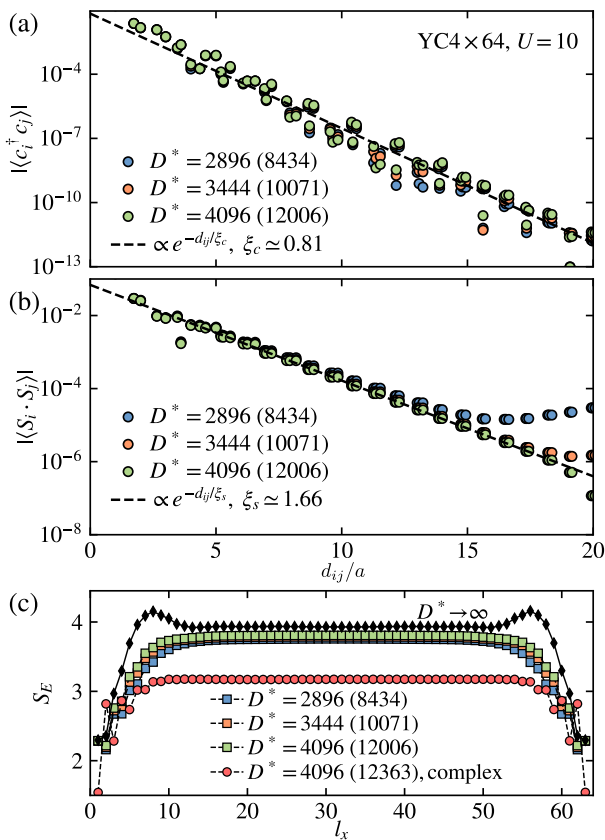


FIG. 3. Single-particle Green's function, spin correlation, and entanglement entropy obtained on the YC4×64 cylinder at $U = 10$. Log-linear plot of (a) single-particle Green's function $|\langle c_i^\dagger c_j \rangle|$, and (b) spin correlation $|\langle S_i \cdot S_j \rangle|$ as a function of distance d_{ij} , which show exponential decay with short decay lengths $\xi_c \approx 0.81$ and $\xi_s \approx 1.66$, respectively. (c) Bipartite entanglement entropy S_E vs. block size l_x . This shows a well-developed plateau for $12 \leq l_x \leq 52$, and hence obeys the area law in the bulk. We include a linear extrapolation $1/D^* \rightarrow 0$ (black symbols). S_E is also obtained from a complex wave-function (red symbols; all other data for a real wave function) with the value of the plateau in the center reduced by $\log(2 \pm 0.1)$.

large system sizes are required to capture the chiral phase, the bulk gap's *relative* effect on the ground state energy is very small, thus requiring an energy accuracy of at least 0.2% for the case of $YC4 \times 64$. Therefore sufficiently accurate simulations on large systems are important. In the present case this is made possible by fully exploiting the $SU(2)$ spin symmetry. The sizeable bulk gap Δ_χ is consistent with the extremely short spin and charge correlation lengths on the order of the lattice spacings itself as seen in Fig. 3. Importantly, these correlation lengths are already also much shorter than the width of the YC4 cylinder analyzed here. In this sense, it appears plausible that the chiral phase persists to wider systems in the low-energy regime. And, indeed, as we will demonstrate further below, we see a consistent picture including chiral long-range correlations also for the YC6 cylinder.

C. Spontaneous time-reversal symmetry breaking

One key debate in the previous DMRG studies is whether there exists a spontaneous TRS breaking [15] or not [14]. Here we resolve this issue by calculating the spin chiral correlation $\langle \hat{\chi}_i \hat{\chi}_j \rangle$ (see appendix for more details) between two three-spin triangles Δ_i and Δ_j symmetrically separated from the system center, i.e., having $i=-j$ or $i=1-j$, as shown in Fig. 1(a). The involved scalar chirality operator is $\hat{\chi}_i = (\hat{\sigma}_\alpha \times \hat{\sigma}_\beta) \cdot \hat{\sigma}_\gamma$ with $\alpha, \beta, \gamma \in \Delta_i$ in counter-clockwise order for the Pauli operators. Given that the hermitian operator $\hat{\chi}_i$ has purely imaginary matrix elements, using real-valued DMRG with the real-valued Hamiltonian in Eq. (1) will always yield $\langle \hat{\chi}_i \rangle = 0$. Evidently, a real-valued wave function cannot (spontaneously) break TRS. Hence we compute static chiral correlations $\langle \hat{\chi}_i \hat{\chi}_j \rangle$. Eventually, however, we do repeat precisely the same DMRG calculations but using complex arithmetic, which then permits a plain non-zero expectation value of the order parameter $\langle \hat{\chi}_i \rangle$.

We first check the system-length dependence of the chiral correlation at $U = 10$. As shown in Fig. 4(a), for small length $L = 16$ and 18, the chiral correlations decay exponentially in the same sign, in agreement to the previous study [14]. Interestingly, by further increasing system length, a very robust chiral correlation establishes over long distances, with $\sqrt{\langle \hat{\chi}_i \hat{\chi}_j \rangle} \approx 0.36$ for $d_{ij} = 30$ already well-converged over distance for $D^* \gtrsim 3444$ [c.f. the orange horizontal guide in Fig. 4(a)]. We also perform a complex-valued DMRG simulation in YC4×64 system, to directly estimate the chiral order parameter $\langle \hat{\chi} \rangle \approx 0.35$ in the bulk, in excellent agreement with previous infinite-DMRG value $\chi_{\text{DMRG}}(U=10) \approx 0.34$ [15]. Our results indicate that the system length is crucial for identifying the spontaneous TRS breaking in the DMRG calculations, which reconciles the different observations in previous studies. In Fig. 4(b) we also compute the chiral correlations in the neighboring phases. In either case, ($U=9$) $\lesssim U_{c1}$ and ($U=11$) $> U_{c2}$, chiral correlations decay exponentially, consistent with the preserved TRS in the two phases.

Besides, we also studied the XC cylinder, i.e., with one of the bond directions along the x -axis, with circumference up to $W = 6$. There, however, even for large L , we find no strong signature of long-range chiral correlation (see appendix for more details). Such different behaviors of chiral correlation on different geometries have also been observed in the DMRG study of a triangular spin model with further-neighbor interactions [35], where the chiral order on the XC cylinder emerges only at large circumference. We suspect that long-range chiral correlation ultimately also can be found on wider XC cylinder in future studies.

D. Degeneracy in the entanglement spectrum

Besides the long-range chiral correlation, we observe systematic large degeneracy in the entanglement spectra (ES) defined by $\mathcal{E}_i \equiv -\log(\lambda_i)$ with λ_i the Schmidt spectrum of the reduced density matrix of half the system. As shown in Fig. 4(c), the spectrum levels are grouped versus the charge

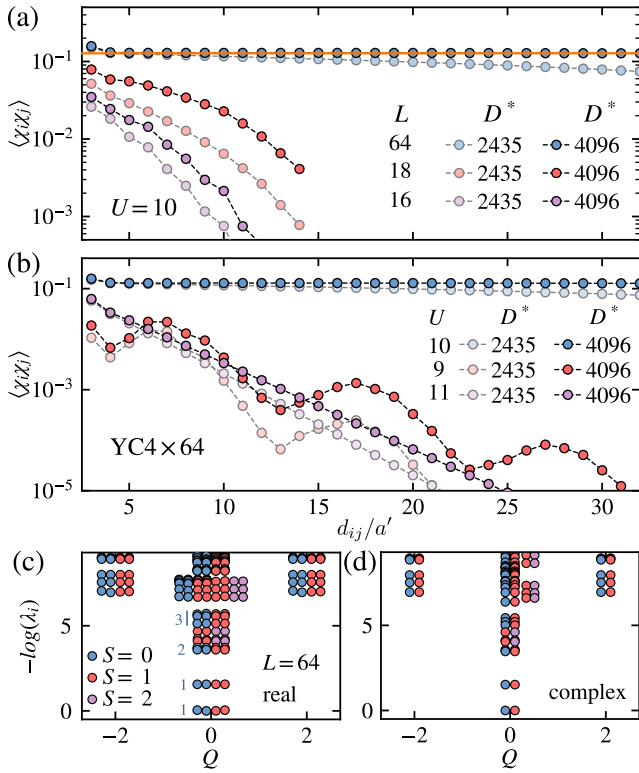


FIG. 4. Chiral correlation and entanglement spectrum of the chiral spin liquid state. (a) Chiral correlations $\langle \chi_i \chi_j \rangle$ for $U = 10$ on the YC4 cylinders with different system lengths $L = 16, 18, 64$. The orange horizontal line indicates the value of $\langle \chi_i \chi_j \rangle \approx 0.128$ for $d_{ij} = 30$. (b) Chiral correlations for $U = 9, 10, 11$ on the YC4 \times 64 cylinder obtained by keeping the bond dimensions up to $D^* = 4096$ multiplets. Entanglement spectra of the YC4 \times 64 for both (c) real and (d) complex wavefunctions, grouped by charge sectors (for even Q), with spin labels color coded as specified in the legend. The bars and respective numbers with the $Q = 0$ column in (c) indicate group degeneracy.

quantum number Q (relative to half-filling) of the subblock, and color coded based on the spin quantum number S . The levels are symmetric for $Q \rightarrow -Q$ because the ES are obtained in the system center. There due to the mirror symmetry removing (adding) a particle from the left subblock necessarily adds (removes) it from the right one.

In the infinite-DMRG calculation [15] the ground state on the YC4 system is found in the semion sector of gapped CSL, where a degeneracy of 2 is observed due to the free spin- $1/2$ edge mode. Our finite-size DMRG simulations ultimately also lead to the same conclusion based on the ES structures in Fig. 4(c-d) in the chiral regime at $U = 10$. However, by comparison to [15], we find even larger ES degeneracies. In Fig. 4(c) the levels show at least a 8-fold degeneracy. For example, there is systematic grouping of two singlets with two triplets (two blue and red dots, respectively). A factor two of this degeneracy is owing to our wavefunction being real while the system is spontaneously TRS broken. This yields the systematic doubling of any spin multiplet in Fig. 4(c), which can be precisely reduced by conducting the same simulation with

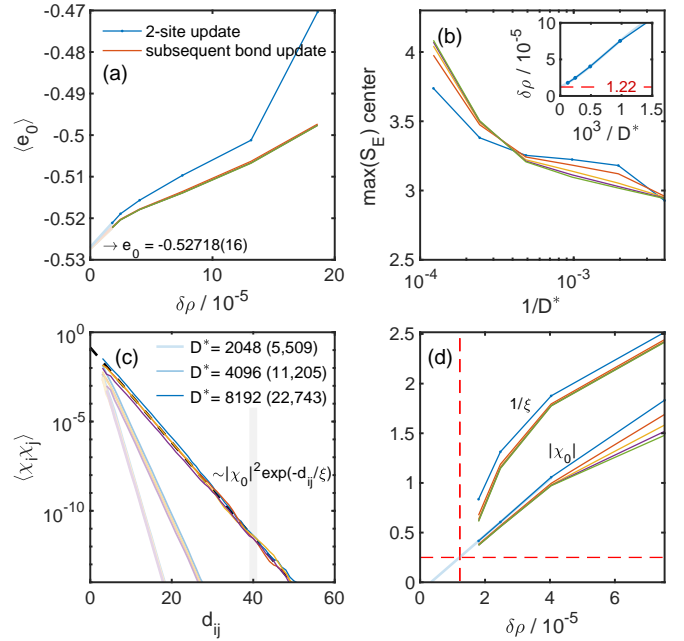


FIG. 5. DMRG simulations (real valued) on YC6 \times 64 at $U = 9$. (a) Convergence of ground state energy vs. discarded weight $\delta\rho$. The lowest energies are extrapolated in a linear fashion towards $\delta\rho \rightarrow 0$, with the resulting ground state energy per site as shown. (b) Maximum entanglement entropy around the system center vs. $1/D^*$, with D^* the number of multiplets kept in the simulation. Inset shows the discarded weight vs. $1/D^*$. The lines in panels (a-b) combine data from equivalent sweeps, such as the 2-site update (line with symbol) when increasing D^* , or the subsequent four bond updates [all other lines; see legend to (a)]. (c) Chiral correlations vs. distance. While this data shows exponential decay, the correlation length is strongly dependent on D^* still. Here the various lines derive from different combinations of up and down triangles vs. distance. Different levels of color intensity refer to different D^* (vs. distance). (d) Analysis of the parameters from the exponential fits in (c) vs. discarded weight $\delta\rho$.

complex arithmetic in Fig. 4(d). Correspondingly, an approximately $\log(2)$ reduction of S_E is also seen in Fig. 3(c).

Besides this two-fold degeneracy, our remaining 4-fold degeneracy between $S=0$ and 1 can be understood as a consequence of the $SU(2)$ DMRG simulation on the state with $S=1/2$ edge spinons, similar to the Haldane phase with $S=1/2$ edge modes in the open spin-1 chain [47]. When computing the ES, we cut the system into two halves such that additional fictitious edge $S=1/2$ degrees of freedom appears at the subblock boundary. This leads to a direct product of the $S=1/2$ edge spinon with the fictitious $S=1/2$, and thus gives rise to the sum of a singlet and a triplet, i.e. $\frac{1}{2} \otimes \frac{1}{2} \equiv 0 \oplus 1$. Similarly, with a boundary $S=3/2$ excitation, one arises at $\frac{3}{2} \otimes \frac{1}{2} \equiv 1 \oplus 2$ [red and purple dots in Fig. 4(c-d)]. The degeneracy of the ES levels agrees with the obtained ground state in the semion sector [48]. Importantly, we find that the low-lying $0 \oplus 1$ levels satisfy the $(1, 1, 2, 3, \dots)$ near-degenerate counting, which is consistent with the $SU(2)_1$ chiral conformal field theory [49] and thus provides further strong support for the gapped CSL [50].

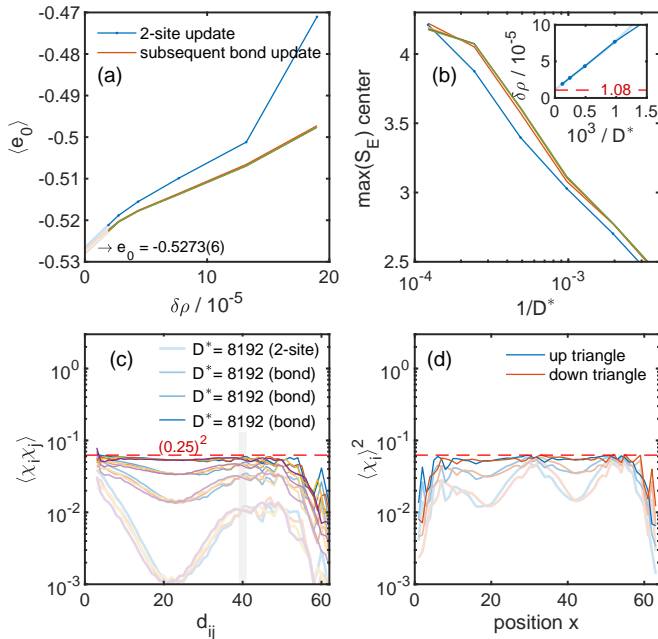


FIG. 6. DMRG simulation (complex valued) on $YC6 \times 64$ at $U = 9$. (a-c) Identical analysis as in Fig. 5. (d) Having complex arithmetic, this permits a non-zero expectation value for the chiral order parameter $\langle \chi_i \rangle$, showing the square for direct comparison with (c). We note that $\langle \chi_i \rangle$ has the same sign for all triangles and, consistent with (c), converges towards $|\langle \chi \rangle| \approx 0.25$ (horizontal red dashed line). The data in (d) corresponds to the last three entries in the legend in (c).

E. Width 6 Cylinders (YC6)

In this section we proceed to YC6 cylinders. A major incentive to look at YC6 is the fact that the 120° phase for large U fits naturally into YC6, but *not* into YC4 (for this reason YC4 switches into an RVB-like phase for very large $U \gtrsim 20$, as already pointed out earlier [43]). As we will see, the chiral intermediate phase also persists in YC6. We take this as strong support for the existence of the intermediate CSL potentially also in the 2D thermodynamic limit.

While the YC6 simulations are considerably more challenging, ultimately, we encounter a rather similar and thus consistent overall picture as for YC4 [15]. Once the cylinders just become long enough, we observe clear long-range chiral correlations. Let us recall: the YC4 cylinders established long-range chiral correlations when (i) the cylinder was sufficiently long [which turned out to be much longer than the circumference, $L > 18$ for $W = 4$ in Fig. 4(a)], and at the same time (ii) the relative energy accuracy was clearly below 1%. Assuming that the bulk gap remains about the same, we have to aim at an even better relative energy accuracy given the increased number of sites here for $YC6 \times 64$, before one can start expecting to see long-range chiral correlations. This makes the YC6 calculations much more challenging.

Yet as we show below, we succeed to demonstrate the buildup and full establishment of long-range chiral correlations also for YC6. This is in full agreement with Szasz *et al.* [15] which based on iDMRG concluded that there is also a

chiral intermediate phase for YC6. However, our results are in stark contrast, e.g., to the more recent variational Monte Carlo simulations Tocchio *et al.* [51]. While they confirmed a chiral intermediate phase for $W = 4$, they concluded the chiral phase to be absent in the low-energy regime of $W = 6$ and thus also in the 2D limit. Similarly, the thermal simulations of Witek *et al.* [52] argued in favor of a gapless stripy intermediate phase (or a gap too small to be detected within their DMRG-based approach of minimally entangled thermal states on YC4 cylinders). We speculate that such conclusions are related to the challenges in the DMRG simulations, as also clearly encountered here based on the requirement of large system sizes. Once under control, however, the spin bulk gap in the chiral intermediate phase is estimated to be large, i.e., of order one or similarly of order $J_{\text{eff}} \approx \frac{4t^2}{U} \approx 0.44$. For reference, a finite spin-gap in the TLU at intermediate Coulomb interaction has also been reported recently in experimental studies on κ -(BEDT-TTF) $_2$ Cu $_2$ (CN) $_3$ [22].

The phase boundaries of the intermediate chiral phase can be expected to be weakly shifted for YC6 as compared to YC4. Indeed, we do not find evidence for long-range chiral correlation for $U = 10$, as used for YC4 in Fig. 4(a). This is consistent with the analysis in [15] which also showed for $W = 6$ that the upper phase boundary for the chiral intermediate phase moves towards slightly lower values, having $U_{c2}^{YC6} \lesssim 10$ just below $U = 10$. Hence we focus on $U = 9$ for the YC6 system. In Fig. 5 we present a DMRG simulation on the $YC6 \times 64$ system using real-valued arithmetic. This is complemented in Fig. 6 by an identical simulation, except that it used complex-valued arithmetic which thus permits spontaneous TRS breaking. In these simulations, for the sake of efficiency, the number D^* of kept multiplets was ramped up quickly by a factor of 2 in a 2-site update, followed by four sweeps with a plain bond-update at the same D^* .

For the real-valued DMRG simulations on YC6 in Fig. 5, we plot the ground state energy vs. the discarded weight $\delta\rho$ in Fig. 5(a). The data converges uniformly, except that it starts to show an onset towards a stronger decrease of the ground state energy for the very smallest $\delta\rho \lesssim 2 \cdot 10^{-5}$. This relates to the fact that for the real-valued DMRG simulations on YC6 we cannot converge the chiral long-range correlations [Fig. 5(c)], despite keeping up to $D^* = 8192$ multiplets (corresponding to $D = 22\,743$ states). To be precise, while the chiral correlations in Fig. 5(c) appear to decay perfectly exponentially over long distances, the correlation length ξ is not converged, in that it keeps increasing with increasing D^* , i.e., chiral correlations become stronger.

The maximum entanglement entropy in the system center vs. $1/D^*$ is tracked in Fig. 5(b), where its inset relates D^* to the respective discarded weight in the DMRG simulation. Since D^* is ramped up very quickly according to the DMRG sweeping protocol specified above, it looks as if $\delta\rho$ extrapolates to a finite value $\delta\rho_0 \approx 1.22 \times 10^{-5}$ for $1/D^* \rightarrow 0$ (horizontal red dashed line). This is artificial, of course, owing to the sweeping protocol, and is attributed to the overall strong truncation still given that the system barely started to move into the low-energy chiral regime. Nevertheless, from a practical point of view, based on that inset in Fig. 5(b), computed

quantities may thus be extrapolated to $\delta\rho \rightarrow \delta\rho_0 > 0$ for quantitative estimates, rather than $\delta\rho \rightarrow 0$.

The exponentially decaying chiral correlations in Fig. 5(c) were fitted by $|\chi_0|^2 e^{-d_{ij}/\xi}$. The resulting fitting parameters $|\chi_0|$ and inverse correlation length $1/\xi$ (slope) are summarized in Fig. 5(d) vs. $\delta\rho$. This suggests that at the same time as $1/\xi$ extrapolates to zero at $\delta\rho \gtrsim \delta\rho_0$ [vertical red dashed line, identical with red dashed line in inset to Fig. 5(b)], the long-range chiral correlation assume the finite value $|\chi| \approx 0.25$ (horizontal red dashed line). Therefore despite the fact that the real DMRG simulation cannot be converged to explicitly show long-range chiral correlations, a careful extrapolation of the data vs. $\delta\rho \rightarrow 0$ (or rather $\delta\rho \rightarrow \delta\rho_0$) does support the conclusion that the YC6 \times 64 cylinder based on real-valued DMRG simulations is chiral for $U = 9$.

This can be significantly more substantiated still by repeating precisely the same DMRG simulation, yet with complex arithmetic, with the results presented in Fig. 6. While the convergence of the ground state energy in Fig. 6(a) looks nearly identical to Fig. 5(a), the maximal entanglement entropy S_E around the system center in Fig. 6(b) already starts to level off and converge for the smallest $1/D^*$ (largest $D^* = 8192$). In particular, the entanglement entropy from the 2-site update (blue) remains already the same for the smallest $1/D^*$ when compared to the subsequent bond updates still (other solid lines). In Fig. 6(c), now one can explicitly observe how the long-range chiral correlations build up for over the last DMRG sweeps, resulting in long-range chiral correlations of approximately $|\chi| \approx 0.25$ which is in agreement with the earlier extrapolation in Fig. 5(d). For the complex DMRG, we can also compute the chiral order parameter $|\chi|$ directly for a line of triangles along the cylinder as shown in Fig. 6(d). The data agrees well in magnitude, and thus is consistent with the chiral correlations shown in Fig. 6(c).

The non-extrapolated DMRG ground state energy of the complex simulation ($e_0 \approx -0.52270$) also agrees well with the real-valued calculation ($e_0 \approx -0.52229$). The former is just slightly lower as it can explicitly make use of the TRS breaking. After simple linear extrapolation of the ground state energies vs. $\delta\rho \rightarrow 0$, one obtains $e_0 \approx -0.5273(1)$. On a more conservative level, extrapolating $\rho \rightarrow \rho_0$ yields $e_0 \approx -0.5245(6)$ which lowers the ground state energy still by about 0.34%. Now given the presence of a downward kink in the convergence of the ground state energy at the smallest energies reached, as already also seen for YC4, the subsequent extrapolated energy gain may thus again be attributed to the presence of a chiral bulk gap. Here for YC6, the estimate yields $\Delta_\chi \cong N \cdot (0.0034 e_0) \sim 0.69$ with $N = LW$ the total number of sites. The estimate for Δ_χ here for YC6 is about a factor of 2.7 larger as previously obtained in the same manner for YC4. The difference may be attributed to the slower sweeping protocol used in the DMRG for YC4 which thus underestimated the actual gap there. Importantly, in the present case the gap estimate compares well to the charge gap $\Delta_c \approx 0.88$ explicitly evaluated for YC4 in App. E [cf. Fig. A5].

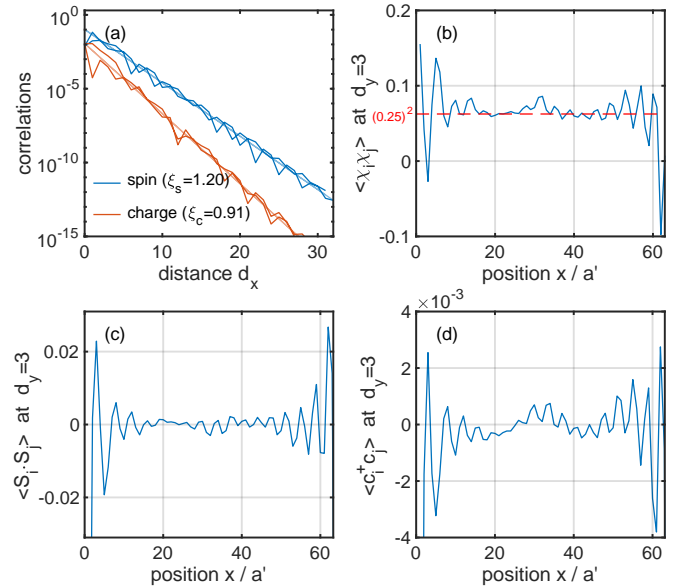


FIG. 7. Correlations in the YC6 \times 64 system after the last sweep in the complex DMRG simulation at $D^*=8192$ in Fig. 6 – (a) Spin $\langle S_i^- S_j^+ \rangle$ and charge correlations $\langle c_i^+ c_j^- \rangle$ (where the dot product sums over spin) relative to the system center. The exponential fit $\propto e^{-x/\xi}$ (straight line in matched light color) yields the corresponding correlation length as shown with the legend. The correlations are computed along a tilted straight path of length $L - 1$ in units of lattice spacing from left to right YC boundary [cf. Fig. 1(a)]. (b) Chiral correlations, (c) Spin correlations, (d) Charge correlations, all at constant vertical distance $d_y=3$ ($j=2, 5$) vs. position x along the cylinder. Here x is the distance from the boundary of the cylinder converted to column index $x/a' \in [1, L]$ [cf. Fig. 1(a)].

F. On the need for long cylinders

All of the systems above required an a priori surprisingly large degree of asymmetry in the aspect ratio of the cylinder geometry in order to realize the intermediate phase with long-range chiral correlations. Specifically, this required the cylinders to be much longer (i.e., in the direction of the open boundary) as compared to their circumference (periodic boundary). With the notion in mind that for a topological system, the bulk gap needs to close towards the boundary, the finite open boundary of the cylinders studied, in principle, can affect the cylinder considerably into the bulk itself. Naively one may have speculated that the effects of the open edges diminish quickly like with the bulk correlation lengths for spin or charge which, based on the data in Fig. 3 for YC4, are ultra-short on the order of a single lattice spacing. But this ignores the fact, that the bulk gap actually needs to physically close towards the open boundary. The length scale over which this occurs is, a priori, far from clear.

In order to gain further insight into the effect of the open boundary, we look more closely into correlations also with reference to the boundary for YC6 \times 64. The results are summarized in Fig. 7 where we take the ground state from the complex-valued DMRG simulation as in Fig. 6 after the last sweep. While $D^* = 8192$ already ensures visible con-

vergence and established long-range chiral correlations, the variations in the data in Fig. 7(b-d) around the system center $x \sim L/2 = 32$ need to be taken with a grain of salt, bearing in mind the residual variations in the data of the last sweep in Fig. 6(d) itself.

To start with, the bulk spin and charge correlations for YC6 remain extremely short ranged, as seen Fig. 7(a). The correlation lengths are of about one lattice spacing, consistent with the YC4 data in Fig. 3. Given the slightly smaller $U = 9$ here, the correlation length for charge transfer is slightly increased, ($\xi_c = 0.81 \rightarrow 0.91$), yet the spin-spin correlation length is actually reduced ($\xi_s = 1.66 \rightarrow 1.20$), bringing the two correlation lengths closer to each other.

Now in order to analyze effects vs. distance from the boundary, we compute chiral, spin, and charge correlations at fixed vertical distance $d_y = 3$ of site i relative to site j (i.e. taken halfway around the cylinder) vs. position $x/a' = 1, \dots, L$ along the cylinder, with the results shown in Fig. 7(b-d), respectively. While superficially, the data behaves similarly, the magnitudes vary considerably, with the scale of values decreasing from chiral to spin to charge correlations, in agreement with the expected respective relevance in the low-energy regime. Furthermore, one notices that all data shows pronounced oscillatory behavior close the boundary with a period of around four columns in YC6, i.e., $\lambda \simeq 4a'$. More importantly, the enveloping amplitude decays rather slowly into the bulk, taking about $\Delta x \sim 5\lambda$, i.e., 20 columns from the open boundary of the YC6 cylinder to diminish. Together with the right boundary, this suggests that in order for the effects of the open boundary to have significantly decayed to actually see bulk behavior in the system center, one needs a rather long cylinder with $L \gtrsim 10\lambda \sim 40$ columns. For shorter systems, the two boundaries can thus be expected to interfere with each other which can be detrimental to the development of long-range chiral correlations, as observed, for example, for the shorter YC4 systems in Fig. 4.

IV. Summary and Conclusions

We show clear numerical evidence for an intermediate chiral spin-liquid phase on long yet finite-size YC4 and YC6 cylinders of the half-filled triangular lattice Hubbard model for sizeable $U \in [U_{c1}, U_{c2}]$ based on exact large-scale DMRG simulations. This phase is surrounded by a metallic phase for $U < U_{c1}$ (or possibly a Luther-Emery liquid [16, 53]), and a 120° magnetic insulating phase for $U > U_{c2}$. For YC4 we find $U_{c1} \simeq 9$ and $U_{c2} \simeq 10.75$, whereas for YC6, $U_{c2} \lesssim 10$. The intermediate spin-liquid phase has been debated intensely in recent literature, with contradicting conclusions on whether it represents a chiral spin liquid or not. Our results demonstrate that finite-size effects can drastically alter the conclusions. Here, the system length in DMRG simulation constitutes a key factor to identify the spontaneous TRS breaking in the CSL state.

In the effective spin model derived from the Hubbard model [7, 28–30], the ring-exchange couplings have order t^4/U^3 . Previous study estimated the metal-insulator transition to occur

at $U/t \simeq 5$ [10], leading to important ring-exchange couplings that can drive a spinon Fermi surface state [7]. However, DMRG studies find the Mott transition at $U/t \simeq 9$, which indicates the much weaker ring-exchange couplings and may explain why the spinon Fermi surface state is not found. Properly accounting for spin couplings in an effective spin model allows one to understand the emergence of the gapped CSL of Kalmeyer-Laughlin type [30].

A handwaving argument on a possible origin of the CSL phase at the intermediate U may be taken from the phase diagram a finite temperature [52]. In the large U limit, TLU reduces to an effective Heisenberg spin model. Besides the long-wavelength soft modes at the K point corresponding to the 120° order, there exists additional rotonlike modes near the M point at higher energy [36]. The softening of the M -point rotonlike excitations, either by quantum fluctuations [54] or thermal fluctuations [36, 43], seems to be accompanied with an emergent liquid-like phase with anomalously enhanced chiral fluctuations. Here the charge fluctuations also lead to a spin liquid phase with long-range chiral order and enhanced spin fluctuations at the M point, naturally implying the softening of the rotonlike excitations and spinon deconfinement in the transition with decreasing U . Therefore it would be interesting and important to explore the spin dynamics of this Hubbard model in future study.

Acknowledgments

We acknowledge the stimulating discussions with Olexei I. Motrunich and Hong-Hao Tu. W.L., Z.C. and S.-S.G. were supported by the National Natural Science Foundation of China grants Nos. 11974036, 11834014, 11874078, 12074024, 11774018, and the Fundamental Research Funds for the Central Universities. D.N.S. was supported by National Science Foundation Grant PREM DMR-1828019. B.B.C. was also supported by the German Research foundation, DFG WE4819/3-1. B.B.C. and W.L. thank the High-performance Computing Center at ITP-CAS for the technical support and generous allocation of CPU time. A.W. was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division.

A. Symmetric Construction of Chiral Operator

With spin rotation $SU(2)$ symmetry implemented, the spin operators need to be reorganized into an irreducible operator (irop), i.e., the spinor also schematically depicted in Fig. A1(a) [38],

$$\hat{S}^{q=1,m} \equiv \begin{pmatrix} \frac{-1}{\sqrt{2}} \hat{S}^+ \\ \hat{S}^z \\ \frac{1}{\sqrt{2}} \hat{S}^- \end{pmatrix}, \quad (\text{A1})$$

with the ladder operators $S^+ = (S^x + iS^y)$, $S^- = (S^x - iS^y)$. Here the components $\hat{S}^{1,+1}$, $\hat{S}^{1,0}$, and $\hat{S}^{1,-1}$, transform like an

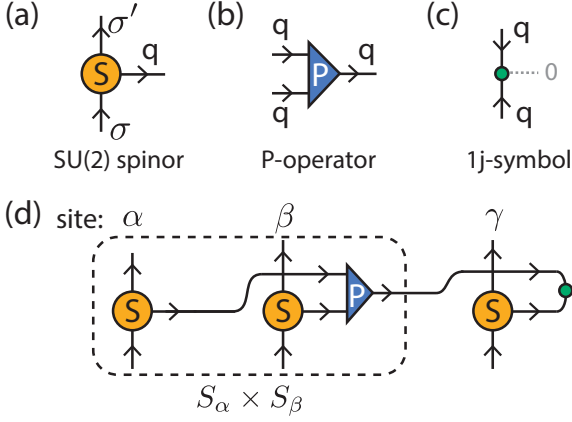


FIG. A1. Symmetric tensor representation of (a) SU(2) spinor S^q [Eq. (A1)], (b) P -operator, (c) the 1j-symbol, and (d) the scalar chirality operator order χ [c.f. Eq. (A2b)].

irreducible representation (irrep) $|S; S_z\rangle$ with $S_z = +1, 0, -1$, respectively. For this, the relative sign on the first component is important. The spin operator always corresponds to the adjoint representation, i.e., $S = 1$ for SU(2). The normalization of the irrep in Eq. (A1) is chosen such that $\hat{S}_i^\dagger \cdot \hat{S}_j$ corresponds to the standard Heisenberg interaction $\hat{S}_i \cdot \hat{S}_j$ where, nevertheless, when having Eq. (A1), the dagger on \hat{S}_i becomes important.

Next we can build the scalar chirality, which we expand as

$$\begin{aligned} \hat{\chi}_{ijk} &= (\hat{\sigma}_i \times \hat{\sigma}_j) \cdot \hat{\sigma}_k \equiv 8 (\hat{S}_i \times \hat{S}_j) \cdot \hat{S}_k \\ &= 4i (\hat{S}_i^+ \hat{S}_j^- \hat{S}_k^z + \hat{S}_i^z \hat{S}_j^+ \hat{S}_k^- + \hat{S}_i^- \hat{S}_j^z \hat{S}_k^+ - \text{H.c.}). \end{aligned} \quad (\text{A2a})$$

Now while this may look somewhat tedious, in practice, there is a simple transparent procedural way for dealing with it from a tensor network perspective with SU(2) spin symmetry enabled. As required for an observable, the chiral operator is a scalar operator with nonzero eigenvalues $\pm\sqrt{12}$. It combines three spin operators \hat{S} into a scalar operator $\hat{\chi}$. As depicted in Fig. A1(d), one needs to “tie together” the three $S = 1$ irrep indices (horizontal leg for each S). This can be simply achieved by fusing two $S = 1$ multiplets (ingoing) into $S = 1$ (outgoing). From a symmetry perspective, this is also the only possible combination here. The result corresponds to a tensor \hat{P} that is proportional to the Clebsch-Gordan coefficient tensor (CGT) $C_{1,1}^1 \equiv (1, 1|1) \propto P$ [Fig. A1(b)]. With this, the chiral term can be compactly written as the nested contractions (denoted by $*$)

$$\hat{\chi}_{ijk} = (\hat{S}_i * (\hat{S}_j * \hat{P})) * \hat{S}_k^\dagger, \quad (\text{A3})$$

with the precise order of pairwise contractions by the brackets irrelevant. Optionally, this expression can be further symmetrized. The direction of an arrow (leg) can be reversed based on a so-called ‘1j’ symbol [39]. In the present case this corresponds to the CGT $(1, 1|0) = \frac{1}{\sqrt{3}}U$ that fuses two $S = 1$ multiplets into a singlet. The index with the outgoing $S = 0$ is a singleton dimension and hence can be skipped.

After proper normalization, this reduces to a unitary operator U [Fig. A1(c)]. Thus inserting $U^\dagger * U = 1$, having $U = U^\dagger$ here, and contracting one U onto the outgoing index of P , denoted as $P_A \equiv P * U$ [green dot contracted onto blue triangle in Fig. A1(d)], and the other U onto S_k^\dagger (which effectively removes the dagger), the chiral term becomes [Fig. A1(d)]

$$\hat{\chi}_{ijk} = (\hat{S}_i * (\hat{S}_j * \hat{P}_A)) * \hat{S}_k, \quad (\text{A4})$$

now with all three spin operators \hat{S} on an equal symmetric footing. Here P_A is a completely antisymmetric tensor for its three indices, all of which are incoming now. As such, it corresponds to a Wigner 3j symbol, that in the present case precisely corresponds to the Levi-Civita tensor as it appears in the original definition of the chiral operator in Eq. (A2a), up to an overall purely imaginary normalization factor.

B. Convergence of the DMRG calculation

In this section, we show the computed entanglement entropy S_E and ground-state energy per site e_g vs. bond dimension $1/D^*$. As can be seen below, the results shown in the main text for the intermediate chiral spin liquid (CSL) and large- U regimes are well-converged vs. D^* . In practice, to ensure convergence of the data, we ramp up the bond dimension D^* for sweep n in the uniform exponential manner, as described by $D_n^* = D_0^* \cdot a^n$. Here a is a parameter that controls the speed of increase of D^* from one sweep to the next, until the final bond dimension $D_{n_{\max}}^*$ is reached. We start with an initial bond dimension D_0^* during a random initialization in the global symmetry sector $(S, Q) = (0, 0)$ where S and Q denote the spin and charge quantum numbers, respectively. In this work, we use $D_0^* = 512$, $a = 2^{1/5}$, $D_{n_{\max}}^* = 8192$ and thus $n_{\max} = 20$ sweeps. This leads to well converged results in most cases. In addition, we may slow down the ramping of D^* by actually performing up to 5 sweeps for a given ‘stage’ n , before moving on to the next stage $n + 1$ with increased bond dimension D_{n+1}^* , e.g., seen as vertical stacking of data points in Fig. A2.

In the left panels of Fig. A2, we show particular specifics of the block-entanglement entropy simulations as we ramp up the number of multiplets D^* in our DMRG simulations. We show both the maximum values of entanglement entropy S_E (labeled by ‘Max’) and the ones cutting at the center bond of the system (labeled by ‘Mid’). In Fig. A2(a), for $U = 8$, we can see the fast growth of entanglement entropy S_E , expected in metallic state that is extremely challenging for the DMRG calculation, even though overall convergence, e.g. of the ground state energy, already appears systematic. For the CSL phase at $U = 10$ [panel (c,g)] and the magnetically ordered phase at $U = 12$ [panel (e)], S_E results are well converged vs. $1/D^*$.

In the right panels of Fig. A2 we check the convergence of the ground-state energy e_g . We linearly extrapolate it towards $1/D^* \rightarrow 0$ based on the last three data points (red line in insets). We use the extrapolated value $e_g^0 \equiv \lim_{1/D^* \rightarrow 0} e_g(D^*)$, to estimate the ‘error’ of finite-bond-dimension energy as the

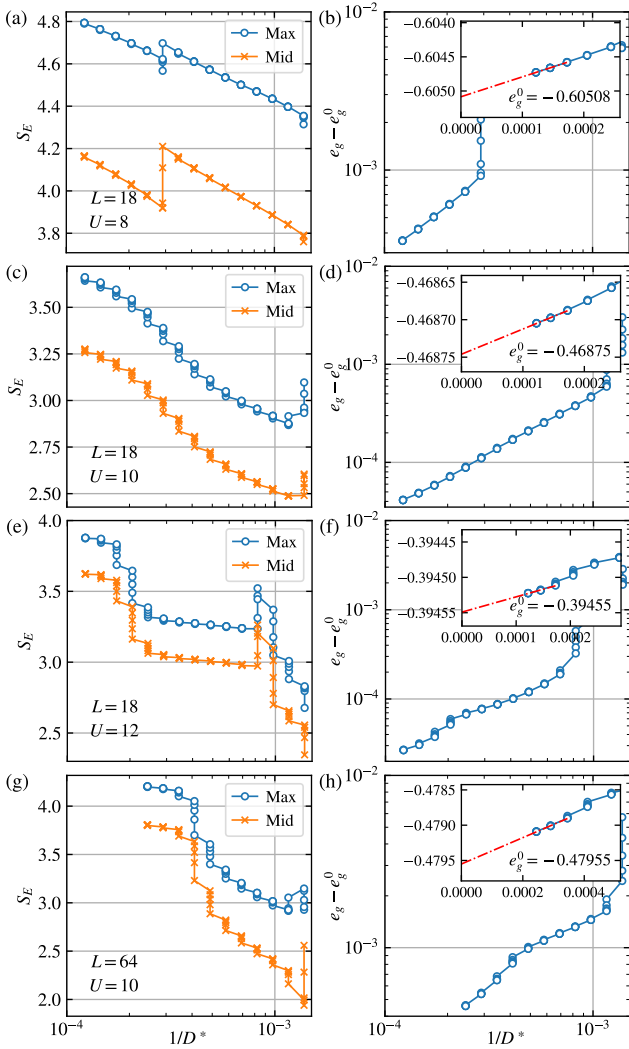


FIG. A2. Entanglement entropy S_E (left panels) and ground-state energy (right panels) vs. $1/D^*$ for YC4 for (a, b) $U = 8$; (c, d) $U = 10$; (e, f) $U = 12$ at $L = 18$; and for (g, h) $U = 10$ at $L = 64$. Entanglement entropy S_E labeled 'Max' are the maximal entanglement amongst all bonds throughout the system; while the 'Mid' ones are values measured at the center of the systems. The linear extrapolations with $1/D^*$ are shown in the inset of all right panels (b,d,f,h). Although the calculations are very challenging in the metallic phase with $U = 8$, the data have reached very good convergence for $U = 10$ and 12.

difference $\epsilon_e \equiv e_g(D^*) - e_g^0$, as shown in the main right panels in Fig. A2. From these we see that, within their respective system size, at $L = 18$ the energies are converged to $\epsilon_e \sim 10^{-4}$ for $U = 8$ and to $\epsilon_e \sim 10^{-5}$ for $U = 10, 12$; and at $L = 64$, the energies are converged to $\epsilon_e \sim 10^{-4}$ for $U = 10$.

Overall, convergence is not always smooth with increasing bond dimension D^* . For example, there may be excitations in the system due to the arbitrary initialization of the wave function for small D^* which, nevertheless, get ironed out early on. Certain low-energy excitation as well as edge modes can be dealt with over longer distances only once a sufficient accuracy, i.e., sufficiently large D^* , has been reached which then

may lead to a rather sharp drop or increase in the maximal block entanglement across the entire system, as well as a rapid drop of energy, as observed for intermediate and also larger D^* .

On a physical level, a certain choice of D^* permits a certain energy resolution, as evident from the analysis of the right panels of Fig. A2. This gives insights into the energy scales of the system under consideration. Consider, for example, the bottom panels of Fig. A2 for the chiral intermediated state at $U = 10$ for $L = 64$. There the chirality of the ground state only emerges for $D^* \gtrsim 2500$ ($1/D^* \lesssim 4 \times 10^{-4}$) which leads to a sharp rise in the entanglement entropy [Fig. A2(g)]. This by itself already suggests a significant change in the underlying DMRG wave function, e.g., as also seen in the convergence of the chiral long range correlation in Fig. 4(a) of main text. Indeed, the systematic degeneracies in the entanglement spectra also only emerge once the sufficiently large $D^* \gtrsim 2500$ is reached in the present case. Simultaneous with strong rise in the entanglement entropy in Fig. A2(g), one also observes a kink in the convergence of the ground state energy [Fig. A2(h)].

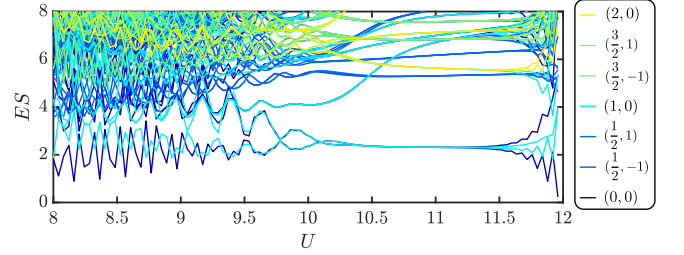


FIG. A3. Entanglement spectrum (ES) along the system and in between full columns of a DMRG scan along a YC4x100 cylinder. For this we tune $U \in [8, 12]$ linearly along the cylinder. We keep the bond dimensions up to $D^* = 5793$ ($D \lesssim 17053$). The lines are color coded according to their symmetry sector (S, Q) as indicated.

This suggests that for the chiral state to be seen in the DMRG simulations, one needs an energy resolution as found at the kink, that is $\Delta e_\chi \cong e_g - e_g^0 \simeq 10^{-3}$. If one were to interpret this to reflect an actual energy gap below which the DMRG convergence is accelerated, the corresponding estimate would be $\Delta_\chi \cong LW \cdot \Delta e_\chi \simeq 0.25$ and thus sizeable. Increasing the ground state energy of the $L = 64$ system by adding Δe_χ , it still has a significantly lower energy than the $L = 18$ system, given that $\Delta e_g^0 = e_g^0(L = 18) - e_g^0(L = 64) \sim 0.011$. Conversely then, a non-chiral state may be seen if the energy of a given (eigen) state is higher by Δ_χ above the ground state for given width $W = 4$ system, either due to insufficient D^* or due to finite-length effects. The latter is demonstrated by comparison to the same $U = 10$ Hamiltonian, yet for the significantly shorter $L = 18$ in Fig. A2(cd). From the above estimates, the finite size correction for this smaller system size is significantly larger than Δ_χ , and so we do not yet see a chiral signatures in its converged ground state [see also Fig. A6(a)]. Therefore also the entanglement profile in Fig. A2(c) evolves much more smoothly as compared to Fig. A2(g), and the convergence of the ground state energy in

Fig. A2(d) also shows no kink.

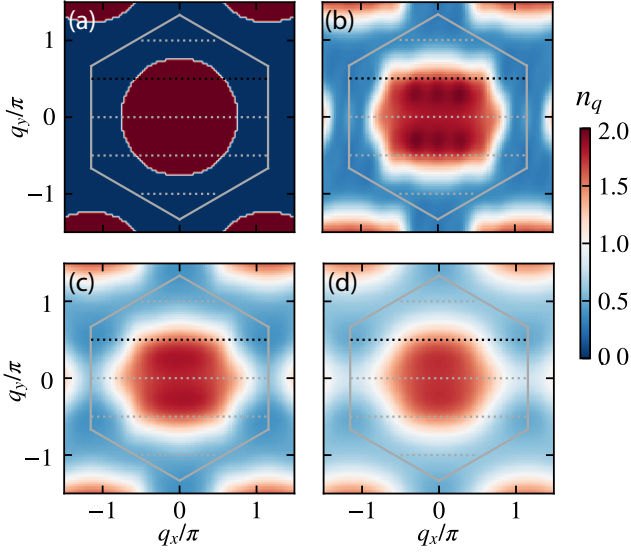


FIG. A4. Electron density in the momentum space n_q on the YC4 \times 18 systems with (a) $U=0$ (b) $U=8$ (c) $U=9$ (d) $U=10$. Grey hexagons represent the boundary of first Brillouin zone, and the horizontal dotted lines represent the allowed momenta q_y for the YC4 cylinder.

C. DMRG scan of the cylinder with smoothly changing U

To study the phase diagram of the model, we also performed a linear DMRG scan [55] for a YC4 \times 100 system with the varying U for different columns i as $U_i = U_1 + \frac{i-1}{L-1}(U_L - U_1)$. Here, we set $U_1 = 8$ for the first column and $U_L = 12$ for the last one. The results are shown in Fig. A3, where we compute and collect the entanglement spectra for each cut between the columns i and $i+1$. Since each column i corresponds to a unique U_i , the ES ‘flows’ as U changes along the cylinder.

In the present YC4 system, three possible phases can be discerned in the entanglement spectrum, separated by the two critical points at $U_{c1} \simeq 9.5$ and $U_{c2} \simeq 10.5$. The ES is non-degenerate in the small- U ($U < 9.5$) phase, while it approaches 4- and 8-fold degeneracy in the intermediate- U ($9.5 < U < 10.5$) and large- U ($U > 10.5$) regime (2- and 4-multiplet degeneracy, respectively). When approaching the open right boundary of the cylinder at $U = 12$, the degeneracy starts to split again. This is consistent with the splitting in the ES between the $S = 0$ and $S = 1$ multiplets already discussed with Fig. 4(c-d) in main text.

D. Destruction of the Fermi surface

In Fig. A4, we show the electron density in momentum space n_q on the YC4 \times 18 system. The distribution n_q of the free fermion system at $U = 0$ in the thermodynamic limit is also included in Fig. A4(a) as a reference, which shows a perfect Fermi surface. In Fig. A4(b-d), the calculated n_q exhibits

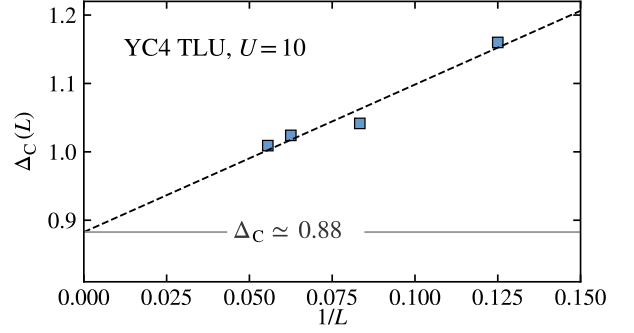


FIG. A5. Charge gap Δ_C calculated on the YC4 cylinders with different lengths $L = 8, 12, 16, 18$. The linearly extrapolated value with $L \rightarrow \infty$ is $\Delta_C \simeq 0.88$.

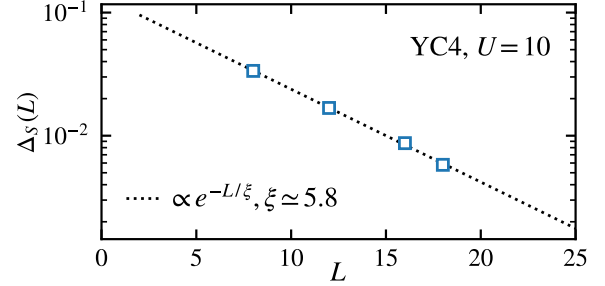


FIG. A6. Energy difference Δ_S between the total spin-0 and total spin-1 sectors. We calculate the YC4 cylinders with system length $L = 8, 12, 16, 18$. This shows an exponential decay with $\xi \simeq 5.8$ with increasing L as indicated.

a recognizable Fermi surface at $U = 8$. With further growing U , the Fermi surface gets significantly blurred at $U = 9$ and 10 , characterizing a metal-insulator transition with destructed Fermi surface.

E. Charge gap

In this section, we directly calculate the charge (single-particle excitation) gap,

$$\Delta_C = \frac{1}{2}[E(\frac{1}{2}, +1) + E(\frac{1}{2}, -1) - 2E(0, 0)],$$

where $E(S, Q)$ denotes the lowest eigenenergy with total spin S , and charge Q taken as the number of particles relative to the half-filling. For the ground state we have $S = Q = 0$. As shown in Fig. A5 for $U = 10$, Δ_C decreases with system length L , and the linear extrapolation over $1/L$ results in a large nonzero gap $\Delta_C \simeq 0.88$. This confirms that the spin liquid resides in the Mott insulating phase.

F. Identification of edge spinons from spin excitation

In the CSL phase, we have found the obtained ground state (on the YC4 cylinder) in the semion topological sector, with a

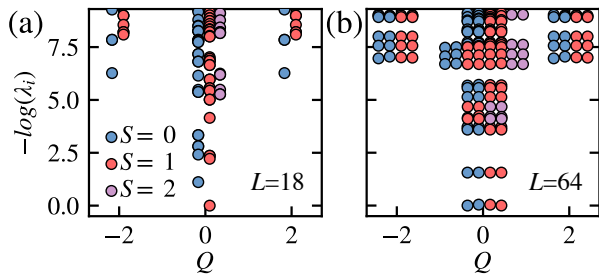


FIG. A7. Entanglement spectrum calculated at the center of YC4 systems with (a) $L = 18$, and (b) $L = 64$ [cf. Fig. 4(c) in main text], grouped by charge sectors (showing even Q only), with spin labels color coded as specified in the legend.

spin-1/2 spinon on each open boundary. Therefore we expect a 4-fold ground state degeneracy ($S = 0 \oplus 1$) for sufficiently long cylinders. This may be seen analogous to the Haldane phase of the open $S = 1$ spin chain, where also spin-1/2 edge modes are weakly coupled. Thus, the energy difference between the total spin-0 and spin-1 sectors is expected to decay exponentially with growing system size [56].

Here, we use the similar strategy to identify the edge spinons in the semion sector of the CSL state. We show the energy difference Δ_S between the total spin-0 and spin-1 sectors with growing system length in Fig. A6. Clearly, Δ_S is very small and indeed decays exponentially with L . Since the spin triplet excitation is gapped in the bulk (c.f. Fig. 3 in main text), this vanishing energy difference must be ascribed to the edge spinon modes in the semion sector of the CSL.

G. Finite-size effect of TRS breaking detection

To further emphasize the absence long-range chiral correlation on short systems, we also contrast the ES for $L = 18$ to $L = 64$ in the main paper. As seen in Fig. A7, $L = 18$ is still clearly qualitatively different. An obvious difference from the ES for $L = 64$ [Fig. A7(b)] is the complete absence of degeneracies which, in particular, demonstrates that the two low-lying real wave functions that respect TRS are still split by a relatively large gap due to finite-size effects. Therefore simulations on such short systems may lead to the premature conclusion of no chiral order. However, with growing system length L , this gap decreases, and eventually becomes negligible. This then allows DMRG calculation to obtain the minimal entangled state with spontaneous TRS breaking [57].

H. Chiral correlation on XC4 system

In this section, we show the chiral correlations $\langle \chi_i \chi_j \rangle$ between two triangles labeled i and j with distance $d = |i - j|$, in XC4×64 systems for the case of $U = 9.5$. As shown in Fig. A8, with a bond dimension up to $D^* = 8192$ SU(2) multiplets (corresponding to $D > 22000$ individual states), chiral correlations are strongly suppressed. We further perform a linear $1/D^* \rightarrow 0$ extrapolation from the correlation data of the largest three bond dimensions $D^* = 4096, 5793, 8192$, and still see no sign of long-range chiral correlation. Hence XC4 behaves very differently from YC4 or YC6 where for cylinders of the same length already robust long-range chiral correlations were observed, having $\chi^2 = 0.128$ for YC4 at $D^* = 4096$ [Fig. 4], or $\chi^2 \approx 0.25$ for YC6 at $D^* \gtrsim 8192$ [Fig. 6]. Here for XC4, the chiral correlations drop rapidly over short distances $d_{ij} < 10$. However, they appear to gain weak support for $d_{ij} > 10$ around the much smaller value $\chi^2 \sim 10^{-4}$. While the long-distance correlations still gain strength with increasing D^* , nevertheless, the chiral correlations do show (weak) decay with distance, even within the numerically converged range $d_{ij} \lesssim 25$. In this sense, we see no clear support for long-range chiral correlation in XC4 cylinders.

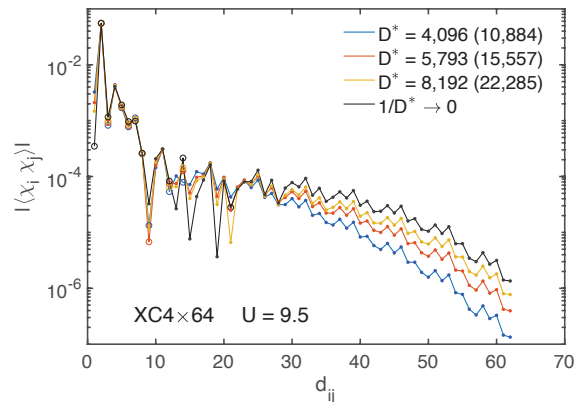


FIG. A8. Chiral correlations calculated in a $U = 9.5$ XC4×64 system with bond dimensions $D^* = 4096, 5793, 8192$, show no long-range correlations. The black line depicts the results of linear extrapolation $1/D^* \rightarrow 0$ from the above bond dimensions. Here, the filled symbol indicated the positive sign of chiral correlation, and otherwise the negative.

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