Multi-Node Quantum Spin Liquids on the Honeycomb Lattice

Jiucai Wang,† Xiaoqun Wang, and Zheng-Xin Liu

1Department of Physics, Renmin University of China, Beijing 100872, China
2Key Laboratory of Artificial Structures and Quantum Control of MOE, Shenyang National Laboratory for Materials Science, Shenyang 110016, China
3School of Physics and Astronomy, Tsung-Dao Lee Institute, Shanghai Jiao Tong University, Shanghai 200240, China

(Dated: March 25, 2020)

Recently it was realized that the zigzag magnetic order in Kitaev materials can be stabilized by small negative off-diagonal interactions called $\Gamma'$-terms. To fully understand the effect of the $\Gamma'$-interactions, we investigate the quantum $K$-$\Gamma$-$\Gamma'$ model on the honeycomb lattice using variational Monte Carlo method. Interestingly, besides the Kitaev spin liquid, two additional gapless $Z_2$ quantum spin liquids (QSLs) are found when $\Gamma' > 0$. These two QSLs, which are proximate to the Kitaev spin liquid, contain 14 and 8 Majorana cones in their spinon excitation spectrum and are named PKSL14 and PKSL8 respectively. The physical properties of these nodal QSLs are studied by applying out-of-plane and in-plane magnetic fields, and the results are dependent on the number of cones. We show that the QSLs in our phase diagram are belonging to a family of multi-node quantum phases which share the same projective symmetry group but contain different number $(6n + 2, n \in \mathbb{Z})$ of Majorana cones in their excitation spectra. Our results may provide guidance for potential experimental realization of non-Kitaev gapless QSLs in relevant materials.

Introduction — Quantum spin liquids (QSLs) are exotic phases of matter exhibiting no conventional long-range order down to the lowest temperatures. However, it is challenging to construct lattice models to support spin-liquid ground states and then realize them in candidate materials. In 2006, Kitaev proposed a honeycomb lattice model which has an exactly solvable QSL ground state and a gapless or gapped excitation spectrum. In a general magnetic field, the gapless Kitaev spin liquid (KSL) can be turned into a gapped chiral spin liquid (CSL) that supports non-Abelian anyonic excitations. It has been proposed that spin-orbit entangled materials, such as Na2IrO3 and α-RuCl3, contain Kitaev interactions $$(S^\alpha_i S^\beta_j)$$ and are candidates to realize the KSL. However, these materials manifest magnetic long-range order at low temperatures, indicating that the non-Kitaev interactions such as the off-diagonal symmetric $\Gamma$ interactions $$(S^\alpha_i S^\beta_j + S^\beta_i S^\alpha_j)$$ are not negligible. Although many lattice models have been proposed as the effective interactions of the Kitaev materials, none of them can explain all of the experimental data. A third-neighbour Heisenberg interaction was proposed to interpret the zigzag order, but the question is that long-range interactions are usually too small to stabilize the order. Recently, another nearest neighbour off-diagonal interaction $$(S^\alpha_i S^\beta_j + S^\beta_i S^\alpha_j + S^\gamma_i S^\gamma_j)$$ called the $\Gamma'$-term attracts some attention. Density matrix renormalization group and infinite tensor network studies have shown that a very small $\Gamma' < 0$ can support a zigzag-ordered ground state. The physical origin of the $\Gamma'$ interactions may be owing to the trigonal distortion of the Kitaev materials. Since the parameters of the effective interactions in different materials are generally different, it is possible that in some compound the $\Gamma'$-interaction may switch its sign. The physical consequence of the interactions in such a parameter regime still needs to be revealed.

In the present work, the quantum $K$-$\Gamma$-$\Gamma'$ honeycomb model is studied using variational Monte Carlo (VMC) method and the global phase diagram is obtained. Besides the well known KSL phase, we find two more QSLs at $\Gamma' > 0$ which share the same projective symmetry group (PSG) with the KSL and are thus called the proximate Kitaev spin liquids (PKSL). These two phases contain 14 and 8 Majorana cones in their excitation spectrum and are labeled as PKSL14 and PKSL8 respectively. The physical properties of the PKSL phases depend on the number of cones. For example, in a weak magnetic field oriented along the $\hat{c} \equiv \hat{x} + \hat{y}$, the number of cones reduces to 6 and 4 respectively. With the presence of $\Gamma'$ interactions, even the zigzag ordered phase can be tuned into QSLs by magnetic fields. For instance, under a field along the $\hat{x} - \hat{y}$ direction, an 8-cone QSL and a 4-cone QSL are induced with proper field strength. It is shown that these two field-induced gapless QSLs originate from a parent 20-cone zero-field KSL state. Therefore, our study reveals a family of nodal $Z_2$ QSLs which share the same projective symmetry group but contain different number $(6n + 2, n \in \mathbb{Z})$ of Majorana cones in their excitation spectrum. This result is instructive for experimental search of gapless QSLs in related materials.

The Model and the method — The model we consider is

$$H = \sum_{\langle i,j \rangle \gamma} (K S^\gamma_i S^\gamma_j + \Gamma (S^\alpha_i S^\beta_j + S^\beta_i S^\alpha_j)$$

$$+ \Gamma' (S^\alpha_i S^\beta_j + S^\beta_i S^\alpha_j + S^\gamma_i S^\gamma_j + S^\gamma_i S^\gamma_j),$$

(1)

where $\langle i,j \rangle$ denotes nearest-neighbor sites, $\gamma$ labels the type of the bond $\langle i,j \rangle$ on the honeycomb lattice, and $\alpha, \beta, \gamma$ stand for the spin index. In most Kitaev materials the Kitaev terms have negative sign $K < 0$. In the present work, we adopt the parameters of the interactions such that $K < 0$, $\Gamma > 0$ and $\Gamma'$ is either positive or negative. Due to spin-orbit coupling, the symmetry of the model is described by the finite magnetic
point group $D_{3d} \times Z_2^T$ besides lattice translation symmetries, where $Z_2^T = \{E, T\}$ is the time reversal group.

Our VMC method is based on spinon representation, where the spin operators are written in quadratic forms of fermionic spinons $S_\alpha^\gamma = \frac{1}{2} c_i^\alpha \sigma^\gamma c_i$, where $c_i = (c_i^\uparrow, c_i^\downarrow)$, $m = x, y, z$, and $\sigma^m$ are Pauli matrices. The particle-number constraint, $\hat{N}_i = c_i^\dagger c_i + c_i c_i^\dagger = 1$, should be imposed at every site such that the size of the fermion Hilbert space is the same as that of the original spin. The spin interactions in Eq. (1) are rewritten in terms of interacting fermionic operators and are decoupled into a non-interacting mean-field Hamiltonian. Since the symmetry group of the system is discrete, the mean field Hamiltonian of a spin liquid should contain spin-orbit coupling and the most general form reads\textsuperscript{22,26,27}

$$H_{\text{mf}}^{\text{SL}} = \sum_{(i,j) \in \beta} \text{Tr}[U_{ji}^{(0)} \rho_i^\sigma \rho_j^\sigma] + \text{Tr}[U_{ji}^{(1)} (i R_{\alpha \beta}) \rho_i^\sigma \rho_j^\sigma]$$

$$+ \text{Tr}[U_{ji}^{(2)} \rho_i^\tau \rho_j^\tau] + \text{Tr}[U_{ji}^{(3)} \rho_i^\tau \rho_j^\tau]$$

where $\rho_i = (C_i \bar{C}_i^\dagger), \bar{C}_i = (c_i^\uparrow, -c_i^\downarrow)^T$, $R_{\alpha \beta} = -\frac{i}{\sqrt{2}}(\sigma^\alpha + \sigma^\beta)$ is a rotation matrix, and $U_{ji}^{(0,1,2,3)}$ are mean-field parameters.

A QSL ground state preserves the whole space group symmetry whose point group is $D_{3d} \times Z_2^T$. However, the symmetry group of a spin liquid mean-field Hamiltonian is the projective symmetry group (PSG)\textsuperscript{20,21} whose group elements are space group operations followed by SU(2) gauge transformations. Using PSG, the number of independent parameters in a QSL phase is considerably reduced. The coefficients $U_{ji}^{(n)}$ in Eq. (2) are constrained to the forms, $U_{ji}^{(0)} = i\eta_0 + i(\rho_a + \rho_c)$, $U_{ji}^{(1)} = i(\rho_a - \rho_c + \rho_d + 2\rho_f)(\tau^\alpha + \tau^\beta) + i\eta_3(\tau^x + \tau^y + \tau^z)$, $U_{ji}^{(2)} = i(\rho_a + \rho_c)\tau^\gamma + i(\rho_f(\tau^\alpha + \tau^\beta) + i\eta_3(\tau^x + \tau^y + \tau^z)$, and $U_{ji}^{(3)} = i(\rho_a - \rho_c - \rho_d)(\tau^\alpha - \tau^\beta)$. On the other hand, to include the competing magnetically ordered states we should also consider the term $H_{\text{mf}}^\text{total} = -\frac{1}{2} \sum_i (M_i \cdot C_i \sigma \sigma C_i + \text{h.c.})$.

The essence of the VMC approach is that the local constraint is enforced by Gutzwiller projection. The Gutzwiller projected mean-field ground states provide a series of trial wave functions $|\Psi(x)\rangle = P_G |\Psi(x)\rangle$, where $x$ denotes the variational parameters, $\{\rho_a, \rho_c, \rho_d, \rho_f, \eta_0, \eta_3, \eta_5, M, \phi\}$. The energy of the trial state $E(x) = \langle \Psi(x) | H | \Psi(x) \rangle$ is computed using Monte Carlo sampling, and the variational parameters $x$ are determined by minimizing the energy $E(x)$. Our calculations are performed on a torus of $8 \times 8$ unit cells, i.e. of 128 lattice sites.

The phase diagram – Different classes of ansatz have been adopted as trial wave functions in the VMC calculations. The state with the lowest variational energy is considered as the ground state. Fig 1 shows the VMC phase diagram of the $K$-$\Gamma$-$\Gamma'$ model in which three QSL phases are obtained. The phase containing the exactly solvable point is called the generic KSL (GKSL) phase whose spinon excitation spectrum contains two Majorana cones in the first Brillouin zone. The GKSL is bounded approximately by $|\Gamma'/|K| = 0.1$ at $\Gamma = 0$ and $|\Gamma'/|K| = 0.15$ in Fig 1(a). The other two gapless QSLs share the same PSG as the GKSL phase and are called the proximate KSL (PKSL) phases\textsuperscript{22}. The one locating at $\Gamma'/|K| > 0.2$ and $\Gamma'/|K| > -0.02$ has 14 Majorana cones in its spinon excitation spectrum (the positions of the cones are shown in Fig 2(a)) and is named PKSL14. With the increasing of $\Gamma'$, the PKSL8 phase shows up which contains 8 Majorana cones (see Fig 2(b)). The conic excitations in the PKSL phases are robust against Gutzwiller projection or local perturbations since they are protected by the combination of spatial-inversion ($P$) and time-reversal ($T$), namely, the $PT$ symmetry\textsuperscript{29,30}. Therefore, if neither $PT$ symmetry breaking nor phase transition takes place,
FIG. 3. Phase diagrams when a magnetic field is applied with \( B \parallel (\hat{x} + \hat{y} + \hat{z}) \). (a) At \( \Gamma/|K| = 1, \Gamma'/|K| = 0.05 \), the PKSL14 evolves into two non-Abelian CSLs (\( \nu = 5, -1 \)) and one Abelian CSL (\( \nu = 4 \)). (b) At \( \Gamma/|K| = 0.9, \Gamma'/|K| = 0.3 \), the PKSL8 is turned into three gapped Abelian CSLs (\( \nu = 2, 4, -2 \)). (c) At \( \Gamma/|K| = 0.2, \Gamma'/|K| = 0.1 \), a direct first-order phase transition occurs from the FM phase to the trivial phase. (d) At \( \Gamma/|K| = 1.4, \Gamma'/|K| = -0.05 \), a direct first-order phase transition occurs from the zigzag phase to the trivial phase. The red solid (blue hollow) points represent first-order (continuous) phase transitions.

The number of cones in the low-energy excitation spectrum of the PKSLs cannot be changed.

Fig. 1(a) contains three magnetically ordered phases, namely, the zigzag phase, the ferromagnetic (FM) phase and the incommensurate spiral (IS) phase. Consistent with Refs. 18 and 19, we find that the zigzag ordered state is robust for \( \Gamma'/|K| < 0 \) and \( \Gamma/|K| > 0.15 \). The FM phase is bounded approximately by \( \Gamma/|K| < 0.6 \) and \( \Gamma'/|K| > 0 \), while the IS phase is sandwiched by the PKSL14 and the zigzag phase, and its region is very small. Fig. 1(b) shows the special case with \( \Gamma = 0 \), where the system falls in the AFM phase (\( \Gamma' \rightarrow +\infty \)) or the FM' phase (\( \Gamma' \rightarrow -\infty \)) in the large \( |\Gamma'| \) limit.

All of the phase transitions between the magnetically ordered phases are of first order. A continuous transition between the PKSL14 state and the PKSL8 state is compatible with symmetry and would take place via merging 6 of the cones (the blue dots in Fig. 2(a)) at the \( \mathbf{k} = 0 \) point. However, our VMC calculation indicates that the transition between the two PKSL phases is accompanied by discontinuous changes of certain variational parameters and is thus first-order (see Appendix A).

**The effect of magnetic fields** – In this part, we consider the consequences of adding an external magnetic field to the gapless QSLs. We first consider the case \( B \parallel (\hat{x} + \hat{y} + \hat{z}) \). It is known that in such a field the KSL opens a gap and becomes a non-Abelian field with Chern number \( \nu = 1 \), where the non-Abelian statistics arise due to unpaired Majorana zero modes associated with the vortices. With the increasing strength of the field, the system undergoes a continuous phase transition (with a gap closing at \( \mathbf{k} = 0 \)) to a trivial polarized phase. In the following we analyze what happens to the two PKSL phases when they are put into magnetic fields.

The 14 cones in the PKSL14 state can be divided into three groups which are marked by different colors in Fig. 2(a). The cones within each group are symmetry-related (namely, they can be transformed into each other via symmetry operations), while the ones in different groups are independent. When magnetic field \( B \parallel (\hat{x} + \hat{y} + \hat{z}) \) is applied, all of the cones are gapped out and the ones in each group contribute the same amount (namely, either \( \frac{1}{2} \) or \( -\frac{1}{2} \)) to the total Chern number when the magnetic field is weak enough to be treated as a perturbation. Therefore, the total Chern number should be \( \nu = 3 \chi_1 + 3 \chi_2 + \chi_3 \), where \( \chi_{1,2,3} = \pm 1 \) denote the chiralities of the three groups of cones. Our numerical calculation indicates that \( \chi_1 = \chi_2 = 1, \chi_3 = -1 \) [see Fig. 2(a)], where the hollow (solid) dots stand for the cones with negative (positive) chirality and thus the total Chern number is \( \nu = 5 \). With increasing \( B \), the system undergoes a continuous phase transition from the \( \nu = 5 \) CSL to \( \nu = -1 \) CSL phase. At the transition point the excitation spectrum closes its gap at 6 Majorana points (similar to the symmetry-constraint to the total Chern number, symmetry also constrains the change of \( \nu \) in a continuous phase transition, for details see Appendix A). With further increasing of \( B \), there are two successive, weakly first-order phase transitions, to a \( \nu = 4 \) Abelian CSL and then to the trivial phase (it is trivial because it is connected to the fully polarized state).

We note that the physical properties of the PKSL14 phase for \( \Gamma' > 0 \) is similar to that of the PKSL phase observed in Ref. 22. The main difference is that the intermediate \( \nu = -1 \) non-Abelian CSL is missing when \( \Gamma = 0 \). Therefore, the PKSL phase is a special line in the PKSL14 phase.

Similarly, a small \( B \) turns the PKSL8 into an Abelian CSL phase with \( \nu = 2 \), as shown in Fig. 3(b). A first-order phase transition from the \( \nu = 2 \) CSL to the \( \nu = 4 \) CSL takes place at the first critical field \( g\mu_B B/|K| = 1.47 \). Interestingly, a continuous phase transition is observed from the
For out-of-plane field with since the region of the size of the IS phase is very small. We only consider the FM and the zigzag orders are suppressed by the field via first-order phase transitions after which the system enters the trivial phase [see Fig.3(c) and (d)]. Notice that the critical field of the zigzag phase in Fig.3(d) is large, no intermediate CSL phase is found (we indeed obtain an intermediate state whose mean-field Chern number is \( v = 1 \), but this state becomes trivial after Gutzwiller projection because its ground state degeneracy on a torus is 1 which indicates \( Z_2 \) confinement).

Then we apply in-plane magnetic field \( \mathbf{B} \parallel (\hat{x} - \hat{y}) \) to the ordered phases. Again, the FM phase and the trivial polarized phase are separated with a direct first-order transition [see Fig.4(c)]. In contrast, in some region of the zigzag phase, after the magnetic order being suppressed by the field, an 8-cone phase and a 4-cone phase emerge in sequence with increasing \(|\mathbf{B}|\), as shown in Fig.4(d) and Fig.5 (b)-(d). The phase transition from the 8-cone phase to the 4-cone phase and the transition from the 4-cone phase to the trivial phase are both of second order, which are characterized by smooth changing of the variational parameters and the pairwise merging and disappearance of the Majorana cones (see Fig.5(c)).

Notice that the two field-induced 4-cone QSLs [Fig.4(b) VS. Fig.4(d)] have different physical properties because the chirality distributions of the cones are different, as shown in Fig.2(b) and Fig.5(d) respectively. The difference between above two 4-cone states can also be seen if we restore the \( D_{3d} \times Z_T^2 \) symmetry by removing the magnetic field manually while keeping all the other variational parameters unchanged, in which case the former [i.e. the one in Fig.4(b)] becomes a PKSL8 state while the latter [i.e. the one in Fig.4(d)], as well as the field-induced 8-cone state] becomes a 20-cone PKSL (PKSL20) state. In other words, the two field-induced QSLs in Fig.4(d) are descending from a PKSL20 phase. Actually, at zero field the PKSL20 state is competing in energy with the zigzag state for \( \Gamma < 0 \) (the energy difference is of magnitude \( 10^{-3} |K| \sim 10^{-2} |K| \) per site). This helps to understand that a proper magnetic field can switch the ground state from the 8-cone phase or the 4-cone state.

We note that no field-induced gapless QSLs are found between the zigzag phase and the trivial phase when \( \Gamma \) is small. Therefore both \( \Gamma \) and \( \Gamma' \) interactions are important for the appearance and the robustness of the intermediate field-induced gapless QSLs.

A family of multi-node \( Z_2 \) QSLs – We have seen that for a given PSG there exist more than one gapless QSLs which are distinguished by the number of cones in their excitation spectrum. Actually, the number of cones is restricted by symmetry. Notice that a general momentum point \( k \) is invariant under the little co-group \( \{ E, PT \} \) whose coset containing \( 12/2 = 6 \) elements. Generally, the representation in each coset transforms a cone into a new one. Therefore, the cones locating at general \( k \) points appear in multiple of 6. The \( K \) and \( K' \) points are special since they are invariant under the little co-group \( D_3 \times \{ E, PT \} \) and are transformed into each other by time-reversal \( T \). Therefore, if there is a cone at \( K \)(or \( K' \)) then there must be a pair of them. Finally, the zone center, \( k = 0 \), which respects the fully \( D_{3d} \times Z_T^2 \) symmetry usually does not support a cone. Hence, a general gapless spin liquid contains 6\( n + 2 \) Majorana cones, where \( n \geq 0 \) is an integer. This defines a family of nodal \( Z_2 \) QSLs which share the same PSG. The QSLs with different \( n \) have different physical properties and can be distinguished by applying magnetic fields. In the present work we realized three of them, with \( n = 0, 1, 2 \), respectively. By adjusting interactions, more phases in this family might be realized. This
may be helpful for further theoretical and experimental studies.

**Conclusion** – We have studied the quantum K-Γ-Γ’ model on the honeycomb lattice using variational Monte Carlo method. We find that as \( K < 0 \) the non-Kitaev interactions \( \Gamma > 0 \), \( \Gamma’ > 0 \) give rise to two proximate Kitaev spin liquid phases which contain 14 Majorana cones (PKSL14) and 8 Majorana cones (PKSL8) respectively. The PKSL14 is similar to the PKSL phase in our previous work while the PKSL8 is new. As the magnetic field \( \mathbf{B} \parallel (\hat{x} + \hat{y} + \hat{z}) \) is added to the PKSL8 state, three gapped Abelian CSLs (with Chern number \( \nu = 2, 4, -2 \)) are realized in sequence with increasing \( |\mathbf{B}| \) before the system entering a trivial gapped phase. As an in-plane weak magnetic field is applied (with \( \mathbf{B} \parallel \hat{x} - \hat{y} \)), a 6-cone (4-cone) field-induced gapless QSL phase is obtained from the PKSL14 (PKSL8) phase. Interestingly, when the zigzag order is suppressed by magnetic field \( \mathbf{B} \parallel (\hat{x} - \hat{y}) \), a gapless \( Z_2 \) QSL phase with 8 or 4 Majorana cones can be induced at intermediate field strength. This provides an alternative interpretation of the nuclear magnetic resonance experiment of \( \alpha \)-RuCl\(_3\)\(^{26,31}\).

Especially, the GKSL, PKSL14 and PKSL8 in our phase diagram have the same PSG and are belonging to a big family of multi-node \( Z_2 \) QSLs whose spinon excitation spectra contain \((6n + 2)\)-Majorana cones. Our study opens a door to seek different QSL phases in quantum magnets with strong spin-orbit couplings. We note that other families of nodal \( Z_2 \) QSLs with different PSGs are still missing in literature and deserve future study. We trust that our theoretical exploration also shed light on realization of various QSLs in experiments.

**Acknowledgement** – We thank B. Normand for valuable discussions and comments. JW and ZXL are supported by the Ministry of Science and Technology of China (Grant No. 2016YFA0300504), the NSF of China (Grants No. 11574392 and No. 11974421), and the Fundamental Research Funds for the Central Universities and the Research Funds of Renmin University of China (No. 19XNLG11). XQW is supported by MOST: 2016YFA0300501 and NSFC: 11974244 and additionally from a Shanghai talent program.

**Appendix A: First-order VS. continuous phase transitions**

**1. Zero magnetic field**

Although a continuous transition between PKSL14 and PKSL8 in the phase diagram of K-Γ-Γ’ model is allowed by symmetry, the numerical results turn out to be weak first-order. This can be seen from the level crossing in the ground-state energy [Fig. 6(a)] or through the sudden change of variational parameters (see Tab. I).

The phase transitions between the QSLs (namely the GKL, the PKSL14, and the PKSL8) and the magnetically ordered phases are sharply first-order, which are characterized by a sudden change of the variational order parameter \( M \). For example, at \( \Gamma'/|K| = 1 \) and \( \Gamma’''/|K| = 0.04 \) (the PKSL14 phase), we obtain \( M = 8.9 \times 10^{-3} \) which is nearly zero, but at \( \Gamma'/|K| = 1 \) and \( \Gamma’''/|K| = 0.02 \) (the zigzag phase), we get \( M = 0.3932 \) which is a finite number.

The phase transitions between magnetically ordered phases must be of first order because continuous phase transitions between different symmetry breaking orders are forbidden in the Landau paradigm.

| States   | \( \rho_a \) | \( \rho_c \) | \( \rho_d \) | \( \rho_f \) | \( \eta_0 \) | \( \eta_1 \) | \( \eta_5 \) |
|----------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|
| PKSL14   | 0.5457       | -0.5630      | 0.5497       | 0.0081       | -0.0363      | -0.5993      | -0.0220      |
| PKSL8    | -0.0235      | -0.6213      | 0.4226       | -0.0059      | 0.2788       | 0.4800       | 0.1262       |

**TABLE I.** Variational parameters of two spin-liquid states, namely, the PKSL14 (for \( \Gamma'/|K| = 0.05 \)) and the PKSL8 (for \( \Gamma'/|K| = 0.1 \)) at fixed \( \Gamma'/|K| = 0.6 \).

**2. In a magnetic field \( \mathbf{B} \parallel (\hat{x} + \hat{y} + \hat{z}) \)**

In this case, the time reversal symmetry (\( T \)) is broken by the field and the remaining symmetry group \(^{32} \) is \((\mathcal{C}_2 \rtimes \{E, TC_2\}) \times Z_2^p \) where \( C_2 \) is a 2-fold rotation and \( Z_2^p = \{E, P\} \) is the spatial inversion group. The gapless QSLs are fully gapped

![ FIG. 6. (a) The energy curves of two variational states (i.e. the PKSL14 and the PKSL8) at fixed \( \Gamma'/|K| = 0.6 \) where the level-crossing indicates a weak first-order phase transition; (b) Spinon dispersion at the critical point between the \( \nu = 4 \) CSL and the \( \nu = -2 \) CSL in Fig. 3(b), where the 6 red dots stand for the gapless points.](image-url)
out and become CSLs. In the following we only consider the transitions between different CSLs.

If a phase transition between two CSLs is a continuous one, at the critical point the spinon spectrum must close its gap and form cone-like dispersions. Since no symmetry is broken at the transition point, the number of ‘cones’ are restricted by symmetry. In the following we consider three possibilities:

(A) there is only one ‘cone’ locating at $\hat{K} = 0$. Since one cone contribute either $1/2$ or $-1/2$ to the Chern number when it is gapped out, the Chern numbers at the two sides of the critical point differ by either 1 or $-1$;

(B) there are two ‘cones’ locating at $K$ and $K'$. Since these two cones are related by inversion symmetry, they have the same contribution to the total Chern number. Therefore, at the transition point the Chern number may change by either 2 or $-2$;

(C) there are six symmetry-related ‘cones’ at general momentum points. At the transition point, the Chern number may change by either 6 or $-6$.

The transition from the $\nu = -1$ CSL to the $\nu = 4$ CSL in Fig. 3(a) is consistent with none of the above cases and is first-order. This is verified in our VMC calculation where a variational parameter $\eta$ changes its sign at the transition.

The transition from the $\nu = 5$ CSL to the $\nu = -3$ CSL in Fig. 3(a) and the one from the $\nu = 4$ CSL to the $\nu = -2$ CSL in Fig. 3(b) is consistent with case (C) and is indeed a continuous transition. The spinon dispersion at the critical point is illustrated in Fig. 6(b).

The transition from $\nu = 2$ CSL to $\nu = 4$ CSL in Fig. 3(b) is consistent with case (B) and might be a continuous one. However, it turns out to be first-order since the variational parameter $\rho_a$ changes its sign at the transition point.

3. In a magnetic field $B \parallel (\hat{x} - \hat{y})$

In this case, $C_3$ and $T$ are broken and the symmetry group reduces to $\{E, C_2T\} \times Z_2^\nu$ where the $C_2$ axis is perpendicular to $B$. Some of the cones in the original gapless QSLs are gapped out by the magnetic field and the rest are locating on the horizontal line, i.e. the symmetric line of $C_2T$. As stated in the main text, continuous transitions are characterized by pairwise merging and disappearance of the cones. We only list the first-order transitions below.

In Fig. 4(a), the transition from the 6-cone state to the trivial phase is first-order, where the variational parameters $\rho_a$, $\eta_0$, $\eta_3$ and $\eta_5$ reverse their sign (shown in Tab. II). The transitions from the FM to the trivial phase in Fig. 4(c) and from the zigzag phase to the 8-cone state in Fig. 4(d) are first-order, since the order parameter $M$ has a jump (not shown).

* xiaojunwang@sjtu.edu.cn
† liuxyp@ruc.edu.cn

1 L. Balents, Nature (London) 464, 199 (2010).
2 Y. Zhou, K. Kanoda, and T.-K. Ng, Rev. Mod. Phys. 89, 025003 (2017).
3 A. Kitaev, Ann. Phys. 321, 2 (2006).
4 G. Jackeli and G. Khaliullin, Phys. Rev. Lett. 102, 017205 (2009).
5 J. Chaloupka, G. Jackeli, and G. Khaliullin, Phys. Rev. Lett. 105, 027204 (2010).
6 J. A. Sears, M. Songvilay, D. P. Clancy, Y. Qiu, Y. Zhao, D. Parshall, and Y.-J. Kim, Phys. Rev. B 91, 144420 (2015).
7 R. D. Johnson, S. C. Williams, A. A. Haghighirad, J. Singleton, V. Zapf, P. Manuel, I. I. Mazin, Y. Li, H. O. Jeschke, R. Valentí, and R. Coldea, Phys. Rev. B 92, 235119 (2015).
8 H.-B. Cao, A. Banerjee, J.-Q. Yan, C. A. Bridges, M. D. Lumsden, D. G. Mandrus, D. A. Tennant, B. C. Chakoumakos, and S. E. Nagler, Phys. Rev. B 93, 134423 (2016).
9 F. Ye, S.-X. Chi, H.-B. Cao, B. C. Chakoumakos, J. A. Fernandez-Baca, R. Custelcean, T.-F. Qi, O. B. Korneta, and G. Cao, Phys. Rev. B 85, 180403(R) (2012).
10 S. K. Choi, R. Coldea, A. N. Kolmogorov, T. Lancaster, G. I. Mazin, S. J. Blundell, P. G. Radaelli, Y. Singh, P. Gegenwart, K. R. Choi, S.-W. Cheong, P. J. Baker, C. Stock, and J. Taylor, Phys. Rev. Lett. 108, 127204 (2012).
11 S. C. Williams, R. D. Johnson, F. Freund, S. Choi, A. Jesche, I. Kimchi, S. Manni, A. Bombardi, P. Manuel, P. Gegenwart, and R. Coldea, Phys. Rev. B 93, 195158 (2016).
12 L. Janssen, E. C. Andrade, and M. Vojta, Phys. Rev. B 96, 064430 (2017).
13 K. Ran, J. Wang, W. Wang, Z.-Y. Dong, X. Ren, S. Bao, S. Li, Z. Ma, Y. Gan, Y. Zhang, J. T. Park, G. Deng, S. Danilkin, S.-L. Yu, J.-X. Li, and J. Wen, Phys. Rev. Lett. 118, 107203 (2017).
14 P. Laurell and S. Okamoto, npj Quantum Mater. 5, 2 (2020).
15 S. M. Winter, K. Riedl, P. A. Maksimov, A. L. Chernyshov, A. Honecker, and R. Valentí, Nat Commun 8, 1152 (2017).
16 S. M. Winter, Y. Li, H. O. Jeschke, and R. Valentí, Phys. Rev. B 93, 214431 (2016).
17 J. G. Rau and H.-Y. Kee, arXiv:1408.4811.
18 J. S. Gordon, A. Catuneanu, E. S. Sørensen, and H.-Y. Kee, Nat Commun 10, 2470 (2019).
19 H.-Y. Lee, R. Kaneko, L.-E. Chern, T. Okubo, Y. Yamaji, N. Kawashima, and Y.-B. Kim, arXiv:1908.07671.
20 X.-G. Wen, Phys. Rev. B 65, 165113 (2002).
21 Y.-Z. You, I. Kimchi, and A. Vishwanath, Phys. Rev. B 86, 085145 (2012).
22 J. Wang, B. Normand, and Z.-X. Liu, Phys. Rev. Lett. 123, 197201 (2019).
23 A. Banerjee, J. Q. Yan, J. Knolle, C. A. Bridges, M. B. Stone, M. D. Lumsden, D. G. Mandrus, D. A. Tennant, R. Moessner, and S. E. Nagler, Science 356, 1055 (2017).
24 W. Wang, Z.-Y. Dong, S.-L. Yu, and J.-X. Li, Phys. Rev. B 96, 115103 (2017).
25 J. Cookmeyer and J. E. Moore, Phys. Rev. B 98, 060412(R) (2018).
26 Z.-X. Liu and B. Normand, Phys. Rev. Lett. 120, 187201 (2018).
27 J. Wang and Z.-X. Liu, arXiv:1912.06167.
28 J. G. Rau, E. K.-H. Lee, and H.-Y. Kee, Phys. Rev. Lett. 112,
A. Kitaev, AIP Conf. Proc. 1134, 22 (2009).

X.-G. Wen, Phys. Rev. B 85, 085103 (2012).

J. Zheng, K. Ran, T. Li, J. Wang, P.-S. Wang, B. Liu, Z.-X. Liu, B. Normand, J. Wen, and W. Yu, Phys. Rev. Lett. 119, 227208 (2017).

H.-C. Jiang, C.-Y. Wang, B. Huang, and Y.-M. Lu, arXiv:1809.08247.