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Line-Graph Approach to Spiral Spin Liquids*

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Competition among exchange interactions is able to induce novel spin correlations on a bipartite lattice without geometrical frustration. A prototype example is the spiral spin liquid, which is a correlated paramagnetic state characterized by sub-dimensional degenerate propagation vectors. Here, using spectral graph theory, we show that spiral spin liquids on a bipartite lattice can be approximated by a further-neighbor model on the corresponding line-graph lattice that is non-bipartite, thus broadening the space of candidate materials that may support the spiral spin liquid phases. As illustrations, we examine neutron scattering experiments performed on two spinel compounds, ZnCr_2Se_4 and $\text{CuInCr}_4\text{Se}_8$, to demonstrate the feasibility of this new approach and expose its possible limitations in experimental realizations.

Introduction.— A spiral spin liquid (SSL) is an exotic correlated paramagnetic state of *sub-dimensional* degeneracy, meaning that the propagation vectors \mathbf{q} of the ground states form a continuous manifold, or spiral surface, in a dimension that is reduced from the original system [1–14]. Similar to geometrically frustrated magnets [15, 16], a SSL may host topological spin textures [17–19] and quantum spin liquid states [3–5, 20–22]. What differentiates a SSL from a conventional frustrated magnet is the sub-dimensional degeneracy, which induces highly distinctive dynamics since the spins are confined to fluctuate collectively as nonlocal spirals [23]. Recent calculations on a square lattice reveal that the low-energy fluctuations in a SSL may behave as topological vortices in momentum space [23], leading to an effective tensor gauge theory with unconventional fracton quadrupole excitations that are deeply connected to theories of quantum information, elasticity, and gravity [24–29].

To date, bipartite lattices have been the primary avenue through which SSLs are studied. This is because the ground state degeneracy on a bipartite lattice can be exact, so that all spin spirals with \mathbf{q} over the spiral surface have exactly the same energy [5]. Although this degeneracy stabilizes the SSL down to very low tem-

peratures [1, 3], it also imposes a strong constraint on real materials because most of the known bipartite-lattice compounds are dominated by the nearest-neighbor interactions J_1 [30–44]. Even for the established model compounds where the second-neighbor interactions J_2 are relatively strong [17, 45, 46], the degeneracy over the spiral surface is only approximate due to the existence of further perturbations [14]. This degeneracy lifting results in an approximate SSL state at elevated temperatures where thermal fluctuations overcome the slight energy difference among the spirals.

Inspired by recent density functional theory (DFT) calculations for the breathing pyrochlore lattice compounds [47], here we seek the realization of an approximate SSL, *i.e.* a SSL with an approximate degeneracy, beyond the bipartite lattices. According to the Luttinger-Tisza theory [48, 49], the degeneracy of a SSL model is encoded in the minimum manifold of the interaction matrix. Using graph theory [50, 51], we show that the J_1 - J_2 model on a bipartite lattice shares the same minimum manifold with a J_1 - J_3 model on the corresponding line-graph lattice, where J_3 denotes the third-neighbor interaction. Thus an approximate SSL state is achieved in the latter case when J_3 is sufficiently strong, which greatly expands the range of materials that may support a SSL state. This line-graph approach to SSL is vetted through neutron scattering experiments performed on two Cr-based chalcogenide spinels ZnCr_2Se_4 and $\text{CuInCr}_4\text{Se}_8$.

Line-graph approach.— Our starting point is a Heisenberg model on a l -regular lattice, where l counts the number of the nearest-neighbor (NN) sites. In the presence of a uniform NN exchange interaction J_1 , the coupled spins form a undirected graph $G = (V, D)$, with V denoting the set of vertices (*i.e.* the spin sites) and D denoting the set of edges (*i.e.* the NN bonds). Two vertices i and j are

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called *adjacent* if the graph contains an edge $e = \{i, j\}$, and the adjacency matrix is defined as $A(G)_{ij} = 1$ for $\{i, j\} \in D$ and 0 otherwise. Following this definition, the spin Hamiltonian, $\mathcal{H} = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$ with $\langle ij \rangle$ denoting the NN bonds, can be expressed through the adjacency matrix as $\mathcal{H} = \frac{1}{2} J_1 A(G)_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$. Therefore, according to the Luttinger-Tisza theory [1, 5], the classical ground state of \mathcal{H} can be determined from the eigensolution of $A(G)$, of which the eigenvalues are defined as the *spectrum* of the graph, denoted as $\sigma(G)$.

Algebraic graph theory defines that the related graphs should have related spectra [50, 51]. Of interest here is the line graph L_G [52, 53], whose vertices correspond to the edges of the root graph G and are adjacent if the original edges share a vertex: $L_G = (D, \{\{e, e'\} | e \cup e' \neq \emptyset, e \neq e'\})$. For a regular bipartite graph G , a convenient way to define its line graph is to select the vertices at the midpoints of the original edges. As illustrated in Fig. 1, for the honeycomb ($l = 3$) and diamond ($l = 4$) lattices that are the prototype hosts of the SSL, their line graphs form the kagome and pyrochlore lattices, respectively. According to graph theory [50, 51], the spectra of G and L_G are related by

$$\sigma(L_G) = (-2)^{m-n} \cup \sigma(G) + l - 2, \quad (1)$$

where m (n) is the total number of edges (vertices) of the root graph G . In reciprocal space, Eq. (1) indicates the existence of flat eigenbands on L_G with a degeneracy of $l-2$, which has been the focus of many recent studies [51, 54–56]. More importantly, it reveals that the non-flat eigenbands of $\sigma(L_G)$ and $\sigma(G)$ share the same dispersion up to a constant of $l-2$, and their eigenvectors are related by the incident matrix as discussed in Ref. [57].

Such a spectrum correspondence can be immediately verified for Heisenberg models. On a regular lattice of g sublattices and N primitive cells, the interaction matrix of a J_1 -only model

$$\mathcal{J}_1^{\alpha\beta}(\mathbf{q}) = \frac{J_1}{N} \sum_{\substack{i \in \alpha, j \in \beta \\ ij \in \langle ij \rangle}} \exp[-i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)] \quad (2)$$

is a $g \times g$ hollow matrix with zero diagonal elements. The eigenbands $\nu(\mathbf{q})$ for $J_1 < 0$ are shown as solid lines in Fig. 1(b) for the honeycomb and kagome lattices, and in Fig. 1(d) for the diamond and pyrochlore lattices. The minimum of the eigenbands ν_{\min} has been subtracted for comparison. Analytical expressions for the eigenbands are presented in the Supplemental Material [58], which includes additional Refs. [59–69]. Aside from the top flat bands in blue color, the two dispersive bands $\nu_{\pm}(\mathbf{q}) - \nu_{\min}$ on the kagome (pyrochlore) lattice overlap exactly with those on the honeycomb (diamond) lattice, which is a direct consequence of spectral graph theory.

This eigenband correspondence is maintained under the addition of certain further-neighbor interactions. For the J_1 - J_2 model on a bipartite lattice,

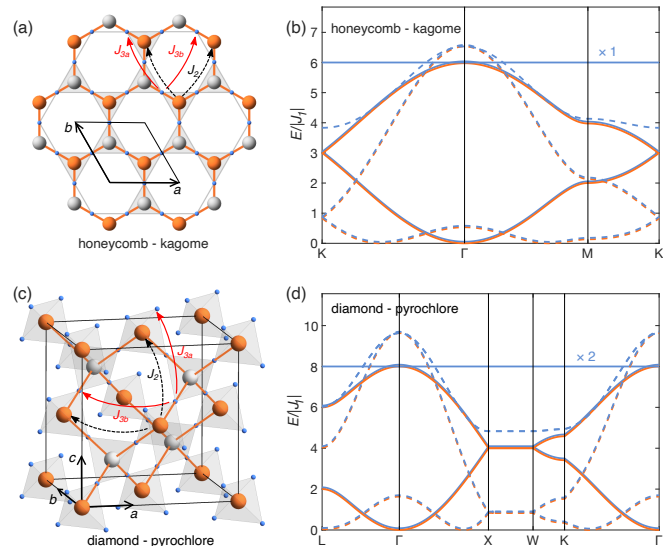


FIG. 1. (a) The line graph of a bipartite honeycomb lattice (large spheres linked by orange bonds) is a kagome lattice (small spheres linked by gray bonds). J_1 is the nearest-neighbor exchange coupling. Black dashed (red solid) arrows indicate the second-neighbor coupling J_2 (third-neighbor couplings J_{3a} and J_{3b}) over the honeycomb (kagome) lattice. (b) Eigenbands of the interaction matrix of Heisenberg models on the honeycomb and kagome lattices. Orange solid (dashed) lines are the two eigenbands for the J_1 model (J_1 - J_2 model with $J_2/J_1 = -0.25$) on the honeycomb lattice, which overlap with the two lower eigenbands of the J_1 model (J_1 - J_3 model with $J_3/J_1 = -0.25$) on the kagome lattice shown in blue solid (dashed) lines. (c-d) Similar correspondence exists between the diamond and pyrochlore lattices. The interaction matrices for the J_1 model (J_1 - J_2 model with $J_2/J_1 = -0.3$) on the diamond lattice and the J_1 model (J_1 - J_3 model with $J_3/J_1 = -0.3$) on the pyrochlore lattice share two same eigenbands shown by the overlapping orange and blue solid (dashed) lines.

J_2 couples the spins of the same sublattices. Therefore, its contribution to $\mathcal{J}(\mathbf{q})$ is a diagonal matrix $\mathcal{J}_2(\mathbf{q})$ that commutes with $\mathcal{J}_1(\mathbf{q})$, leading to a \mathbf{q} -dependent shift $\gamma_G(\mathbf{q})$ of the eigenbands with $\gamma_G(\mathbf{q}) = (J_2/Ng) \sum_{ij \in \langle\langle ij \rangle\rangle} \exp[-i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)]$, where $\langle\langle ij \rangle\rangle$ are the second-neighbor bonds [58]. Similar conclusions can be drawn for the J_1 - J_3 model on the line-graph lattices, as J_3 (including both J_{3a} and J_{3b}) also couples spins of the same sublattices [58]. Since $J_2(G)$ on the root-graph lattice and $J_3(L_G)$ on the line-graph lattice share the same exchange paths as compared in Fig. 1(a,c), we expect, in the case of $J_2(G) = J_3(L_G)$, the same dispersive eigenbands up to a constant on the root- and line-graph lattices. This correspondence is illustrated by the dashed lines in Fig. 1(b,d).

Approximate SSL.— An approximate SSL can be realized on the line-graph lattices through the eigenband correspondence. Following the results of the J_1 - J_2 model on the bipartite lattices [1, 5], it is clear that for the J_1 -

J_3 model on a line-graph lattice the eigenband minima of $\mathcal{J}(\mathbf{q})$ will form a degenerate manifold in reciprocal space at $|J_3/J_1| > 1/(2l)$, where l is the number of the NN sites on the root-graph lattice. Here J_1 and J_3 should be FM and AF, respectively, as the additional flat bands on line graphs remove the FM-AF duality of a bipartite lattice. Since the equal moment constraint over the eigenvectors of $\mathcal{J}(\mathbf{q})$ is not always satisfied for \mathbf{q} over the minimum manifold [58], the SSL realized through the line-graph approach is approximate and needs to be stabilized by thermal fluctuations. As discussed in the Supplemental Material [58], the degeneracy breaking over the spiral surface is relatively weak, which contrasts the strong modulation in the previously studied half-moon patterns [57, 70–72]. This weak degeneracy breaking on the line-graph lattices leads to a stable SSL state in a wide temperature regime that is comparable to that on the bipartite lattices.

Before making comparisons to the experimental data, we further generalize the SSL model by incorporating a breathing distortion on the line-graph lattice so that the NN interactions are modulated alternately as J_1 and J'_1 [47, 73, 74]. For the breathing pyrochlore lattice shown in Fig. 2(a), the eigenbands of $\mathcal{J}(\mathbf{q})$ can be solved as [58]:

$$\begin{aligned} \nu_{1,2} &= J_3 \kappa(\mathbf{q}) \pm \sqrt{4(J_1 - J'_1)^2 + J_1 J'_1 |\eta(\mathbf{q})|^2} + (J_1 + J'_1), \\ \nu_{3,4} &= J_3 \kappa(\mathbf{q}) - (J_1 + J'_1), \end{aligned}$$

where $\eta(\mathbf{q}) = \sum_{n=1}^4 \exp(-i\mathbf{q} \cdot \mathbf{d}_n)$ with \mathbf{d}_n denoting the 4 bonding vectors around each spin site [58] and $\kappa(\mathbf{q}) = |\eta(\mathbf{q})|^2 - 4$. Assuming $J_1 < J'_1 < 0$ and $J_3 > 0$, an approximate SSL state is realized for $\frac{1}{4} \frac{J_1 J'_1}{|J_1 + J'_1|} < J_3 < \frac{1}{4} \frac{J_1 J'_1}{|J_1 - J'_1|}$. The corresponding phase diagram is presented in Fig. 2(b). Representative spiral surfaces in the approximate SSL phase at $|J_3/J_1| = 0.2$ are shown in Fig. 2(c-e) for $J'_1/J_1 = 0.8, 0.6,$ and 0.5 , respectively. The surfaces are identical to those on the diamond lattice [1]. Similar conclusions on the breathing kagome lattice are presented in the Supplemental Material [58].

ZnCr₂Se₄ with a regular pyrochlore lattice.— The Cr-based chalcogenide spinels present nearly ideal model compounds to demonstrate the proposed line-graph approach to SSLs. In these systems, J_1 is FM due to the 90° superexchange path, while $J_2 \sim 0$ due to negligible orbital overlap [75, 76]. As the first example, we study the short-range spin correlations in ZnCr₂Se₄, where the Cr³⁺ ($S = 3/2$) ions form a regular pyrochlore lattice.

Single crystals of ZnCr₂Se₄ were grown using the chemical vapor transport method [58]. Figure 3 summarizes the diffuse neutron scattering results measured on CORELLI at the Spallation Neutron Source (SNS), Oak Ridge National Laboratory (ORNL) [58]. With the statistical chopper, the elastic channel of our CORELLI data has an average energy resolution of about 0.8 meV for an incident neutron energy range of 13 to 33 meV. At 20 K, below the Néel transition temperature $T_N \sim 22$ K,

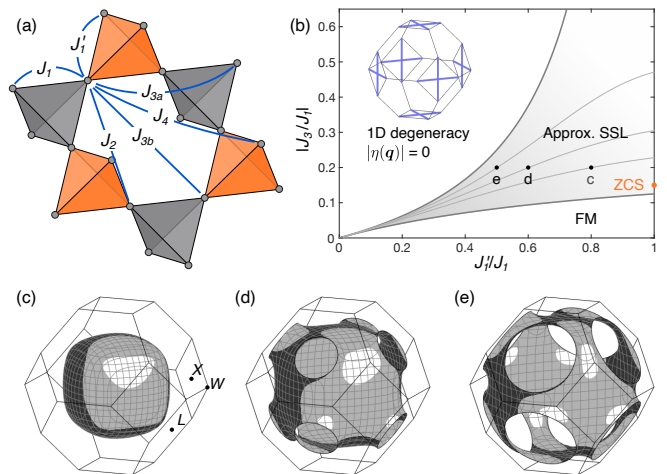


FIG. 2. (a) The breathing pyrochlore lattice is composed of corner-sharing tetrahedra of two different sizes, which results in alternating J_1 and J'_1 couplings over the gray and orange tetrahedra, respectively. The exchange paths of the J_2 , J_{3a} , J_{3b} , and J_4 interactions are also indicated. (b) Phase diagram on the breathing pyrochlore lattice with ferromagnetic J_1 and J'_1 interactions together with antiferromagnetic J_3 interactions assuming $J_{3a} = J_{3b}$. The shaded area indicates the region where an approximate SSL phase can be stabilized by thermal fluctuations. Contour lines for constant $|\eta(\mathbf{q})|$ are shown in the approximate SSL phase. Along these lines, the spiral surface stay the same. Location of ZnCr₂Se₄ (ZCS) with $J_1 = J'_1$ and $|J_3/J_1| = 0.15$ is indicated by the yellow dot. Inset shows the degenerate manifold (blue lines) for $J_3 > \frac{1}{4} \frac{J_1 J'_1}{|J_1 - J'_1|}$ out of the SSL regime. (c-e) Characteristic spiral surfaces with quasi-degenerate energies in the first Brillouin zone at $|J_3/J_1| = 0.2$ and $J'_1/J_1 = 0.8$ (c), 0.6 (d), and 0.5 (e). These points are indicated on the phase diagram in (b) using the panel labels.

magnetic Bragg peaks indexed by $\mathbf{q} = (0, 0, 0.47)$ are observed (see Fig. 3(a)). This is consistent with the helical ground state reported previously [77–79], with the weak ring-like scattering mainly arising from the low-energy magnon excitations [76, 80]. At 30 K, above T_N , magnetic Bragg peaks are replaced by broad diffuse scattering with a spherical shape (see Fig. 3(b-d)), evidencing the emergence of an approximate SSL state where gapped excitations are replaced by quasielastic fluctuations [58].

Assuming a Heisenberg model with exchange interactions up to the fourth neighbors (J_4), we fit the diffuse scattering data using the self consistent Gaussian approximation (SCGA) method [58]. The calculated slices in Fig. 3(b-d) reproduce the experimental data. The coupling strengths are fitted as $J_1 = -2.86(8)$, $J_2 = 0.00(1)$, $J_3 = 0.48(1)$, and $J_4 = -0.057(1)$ meV, which is very close to the values determined by inelastic neutron scattering (INS) [76, 80] and indicates marginal changes in the coupling strengths across the phase transition. The weak strengths of the J_2 and J_4 interactions allow a direct verification of the line-graph approach that is based on

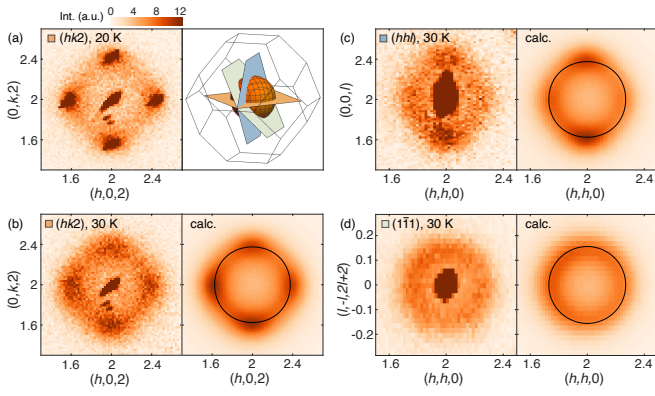


FIG. 3. Quasistatic spin correlations in ZnCr_2Se_4 measured on CORELLI at (a) 20 K and (b-d) 30 K. Slices are along the $(hk2)$ plane in panels (a,b), (hhl) plane in panel (c), and $(1\bar{1}1)$ plane in panel (d). The right sub-panel in (a) shows the complete spiral surface for the J_1 - J_3 model on a pyrochlore lattice with $J_3/J_1 = -0.15$. The flat planes correspond to the slice directions as indicated by the colored plaquette at the top left corner in each panel. The right sub-panels in (b-d) are the calculated diffuse scattering patterns for the fitted J_1 - J_2 - J_3 - J_4 model. Solid line is the spiral surface predicted by the J_1 - J_3 model with $J_3/J_1 = -0.15$. The same linear intensity scale is employed in all panels.

a J_1 - J_3 model. The solid circles on top of the calculated patterns in Fig. 3(b-d) are the contours of the spiral surface predicted by the J_1 - J_3 model with $J_3/J_1 = -0.15$. The contours capture the shapes of the diffuse scattering patterns. As compared in the Supplemental Material [58], the existence of the FM J_4 interaction slightly reduces the radius of the spiral surface while strongly modulates the scattering intensities.

CuInCr4Se8 with a breathing pyrochlore lattice.— Until now, we have assumed a uniform J_3 interaction. In real materials, the J_3 exchange paths on a line-graph lattice can be different as indicated in Fig. 1. This may lead to different coupling strengths and destabilize the SSL state. As the second example, we study the spin correlations in the breathing pyrochlore lattice compound $\text{CuInCr}_4\text{Se}_8$ [81], which has been proposed as a SSL candidate from DFT calculations [47].

A polycrystalline sample of $\text{CuInCr}_4\text{Se}_8$ was synthesized through the solid state reaction method [58]. INS experiments were performed on SEQUOIA at the SNS, ORNL. Neutron diffraction experiments were performed on HB-2A at the high flux isotope reactor (HFIR), ORNL. As is consistent with the previous report [81], magnetic susceptibility shown in Fig. 4(a) suggests a spin glass-like transition at $T_f \sim 15$ K with a clear frequency dependence. This is confirmed in the neutron diffraction results presented in Fig. 4(b), where only broad magnetic features are observed down to 0.25 K. The weak features at $Q \sim 0.41$ and 0.72 \AA^{-1} can be indexed by $\mathbf{q} = (0.48, 0.48, 0)$, while their intensities exhibit a his-

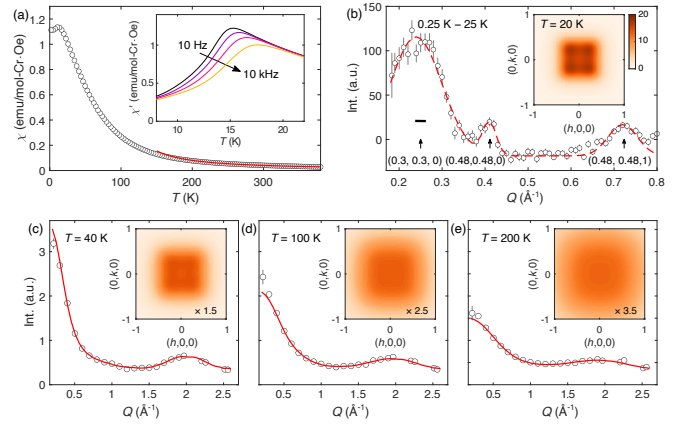


FIG. 4. (a) DC magnetic susceptibility of $\text{CuInCr}_4\text{Se}_8$ measured in a 1×10^4 Oe field (circles). Red line shows the Curie Weiss fit at temperatures between 200 and 380 K. The fitted magnetic moment and Weiss temperature are $\mu_{\text{eff}} = 4.6 \mu_B/\text{Cr}$ and 97(2) K, respectively. The inset shows the frequency dependence of the real part of the ac susceptibility measured in a 10 Oe field. Imaginary part of the ac susceptibility is presented in the Supplemental Material [58]. (b) Difference between the neutron diffraction intensity at 0.25 and 25 K measured on HB-2A. The dashed line is a guide to the eyes. Positions of the characteristic wave vectors are indicated by arrows. The full width at half maximum (FWHM) of the closest nuclear reflection is indicated by the horizontal bar. Inset shows the calculated neutron diffuse scattering pattern in the $(h, k, 0)$ plane at $T = 20$ K for $J_{3a} = 0.07$ meV (see text). (c-e) Circles show the Q -dependence of the equal-time spin correlations at $T = 40$ (c), 100 (d), and 200 K (e). Results of the SCGA fits using the J_1 - J'_1 - J_{3a} - J_{3b} model are shown by the red solid lines. Insets are the calculated neutron diffuse scattering patterns in the $(h, k, 0)$ plane for $J_{3a} = 0.07$ meV at the corresponding temperatures (see text).

tory dependence as expected for a spin glass state.

Figures 4(c-e) present the equal-time spin correlations obtained by integrating the INS spectra from $[-20, 20]$ meV [58]. Assuming a J_1 - J'_1 - J_{3a} - J_{3b} Heisenberg spin model, we fit the diffuse scattering data by the SCGA method [58, 82]. The fitted results are shown in Fig. 4(c-d) as solid lines. The fitted coupling strengths are $J_1 = -1.6(2)$, $J'_1 = -5.4(3)$, $J_{3a} = 0.1(1)$, and $J_{3b} = 0.6(1)$ meV, where the different strengths for J_{3a} and J_{3b} are necessary for a satisfactory fit. As shown in the inset of Fig. 4(b-e), the spin correlations in the $(hk0)$ plane does not follow a circular shape, implying the absence of a SSL state due to the unequal J_{3a} and J_{3b} interactions.

The fitted parameter set provides an explanation for the glassy ground state in $\text{CuInCr}_4\text{Se}_8$. J_{3a} , being the weakest interaction in the J_1 - J'_1 - J_{3a} - J_{3b} model, is however the most crucial parameter in determining the exact length of the LRO $\mathbf{q} = (q, q, 0)$. Within the standard deviation of J_{3a} , q varies from 0 at $J_{3a} = 0$ up to ~ 0.4 at $J_{3a} = 0.2$ meV. Therefore, the broad diffuse scattering at

$Q \sim 0.25 \text{ \AA}^{-1}$, which is tentatively indexed as $(0.3, 0.3, 0)$ in Fig. 4(b), may arise from a finite distribution in J_{3a} due to a tiny amount of structural defects. Weaker features indexed by $\mathbf{q} = (0.48, 0.48, 0)$ may be stabilized by local structural distortions considering its proximity to the commensurate $(\frac{1}{2}, \frac{1}{2}, 0)$ position [83, 84]. Studies of a single crystal sample will help verify the proposed mechanism.

Conclusion.— We have presented a line-graph approach to achieve approximate SSLs on non-bipartite lattices, which allows the experimental exploration of SSLs in a broader family of compounds. Besides ZnCr_2Se_4 studied in this work, chalcogenide spinels like ZnCr_2S_4 [78] and HgCr_2S_4 [85] are worth further investigations as an incommensurate helical ground state has been observed. The approximate SSLs on the line-graph lattices also provide a new platform to explore field-induced topological spin textures since the helicity of the single- \mathbf{q} component is maintained [18]. Such a mechanism may account for the magnetic skyrmions in the breathing kagome-lattice compound $\text{Gd}_3\text{Ru}_4\text{Al}_{12}$ [73]. On the theoretical side, it remains an open question whether the approximate SSLs may evolve into quantum spin liquids under sufficient quantum fluctuations. It is also interesting to explore whether the fracton physics predicted for the exact SSLs [23] may survive to some extent in the approximate SSLs.

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