

Incommensurate Spin-Density Waves in a Frustrated Maple-Leaf Lattice Ferromagnet

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We study how ferromagnetism breaks down in the spin- $\frac{1}{2}$ nearest-neighbor Heisenberg model on the maple-leaf lattice with ferromagnetic J_t , J_d and antiferromagnetic J_h , motivated by the mixed ferro-antiferromagnetic interactions in $\text{Na}_2\text{Mn}_3\text{O}_7$. Exact diagonalization shows that the ferromagnetic boundary does not feature a zero-field spin-nematic phase on the clusters studied here, but an extended regime of incommensurate spin-density-wave correlations with continuously evolving ordering vector. The phase diagram also contains collinear Néel, canted 120° , and hexagonal-singlet regimes, separated by regions that remain difficult to classify from exact diagonalization alone. Variational tests of fully symmetric Gutzwiller-projected Abrikosov-fermion $U(1)$ and $\mathbb{Z}_2$ states find no competitive spin-liquid description of the interior unresolved regions. By contrast, on the ruby-lattice boundary we identify a point between the collinear Néel and hexagonal-singlet phases where a projected $\mathbb{Z}_2$ Ansatz reproduces the finite-size energy and spin correlations with good accuracy.

Introduction. Different inter-atomic distances and exchange mechanisms in crystals generically create both ferromagnetic (FM) and antiferromagnetic (AFM) interactions between local spin degrees of freedom. A common result of this interplay are frustrated ferromagnets whose phase transitions and instabilities are often highly non-trivial. A recurring theme in this context is that direct $\text{FM} \leftrightarrow \text{AFM}$ transitions occurring in a classical model can be accompanied by intermediate, e.g. spin-nematic or lattice-nematic, phases in the quantum case [1–6]. On the other hand, there are scenarios in which a classically existing intermediate phase vanishes in the presence of quantum many-body effects, leading either to a direct transition [7] or giving rise to one or more new putative quantum phases [7, 8]. In extreme cases of frustration, the mechanisms by which the ferromagnetic phase becomes unstable can be rather exotic. For instance, octupolar order was shown to arise on the triangular lattice due to three-magnon interactions [9] and a flat one-magnon band is known to exist above the FM state on the kagome lattice leading to so-called “magnon crystals” [10, 11].

The maple-leaf lattice (MLL) [12], which is illustrated in Fig. 1, is a site-depleted triangular lattice with coordination number five and no spatial reflection symmetry. It has recently gained significant theoretical and experimental attention in the context of exhibiting an exact (J_d) dimer eigenstate [14], coplanar orders [4, 15–27], magnetization plateaus [25, 28–30], anomalous thermodynamic signatures [26, 31–34] as well as potential quantum spin liquid (QSL) phases [13, 17, 18, 20–22, 35]. Indeed, several known MLL materials such as Bluebellite [29, 32, 36] or Spangolite [30, 37] approximately realize $S = 1/2$ ferro-antiferromagnetic nearest-neighbor

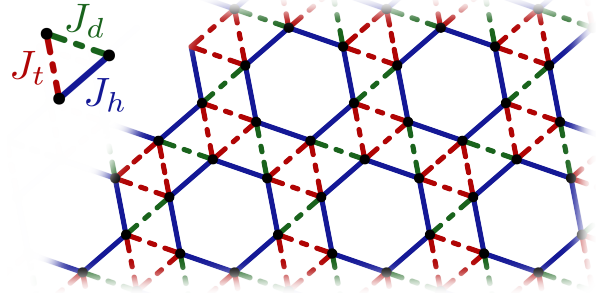


FIG. 1. Maple-leaf lattice with three nearest-neighbor bonds: J_t (red triangles), J_d (green “dimers”), and J_h (blue hexagons). In this work, dashed (solid) bonds correspond to ferromagnetic (antiferromagnetic) interactions.

Heisenberg models such as

$$H = J_t \sum_{\langle i,j \rangle \in t} \mathbf{S}_i \cdot \mathbf{S}_j + J_h \sum_{\langle i,j \rangle \in h} \mathbf{S}_i \cdot \mathbf{S}_j + J_d \sum_{\langle i,j \rangle \in d} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

with three (or more) independent nearest-neighbor couplings J_t, J_h, J_d as defined in Fig. 1. Spangolite is known to essentially host a J_d -dimer ground state that is dressed by ferromagnetic J_h and antiferromagnetic J_t interactions [30]. For Bluebellite it was argued that the strong ferro-antiferromagnetic interactions alternating around hexagons ($J_{h1} = -J_{h2}$) lead to an effective $S = 1$ kagome model with valence-bond-solid ground state [29]. Recently $\text{Na}_2\text{Mn}_3\text{O}_7$, realizing a $S = 3/2$ MLL system, was investigated experimentally [26, 31] and theoretically [34] with reports converging on strongly antiferromagnetic $0 \ll J_h$ and otherwise mostly ferromagnetic $J_t, J_d < 0$ interactions. The $J_t, J_d < 0 < J_h$ phase diagram of the Hamiltonian (1) is thus the natural test-bed for studying frustrated maple-leaf ferromagnets.

In this Letter, we discuss the $J_t, J_d \leq 0 \leq J_h$ phase diagram of the $S = 1/2$ Heisenberg model (1). We show that it exhibits FM, collinear Néel AFM, hexagonal sin-

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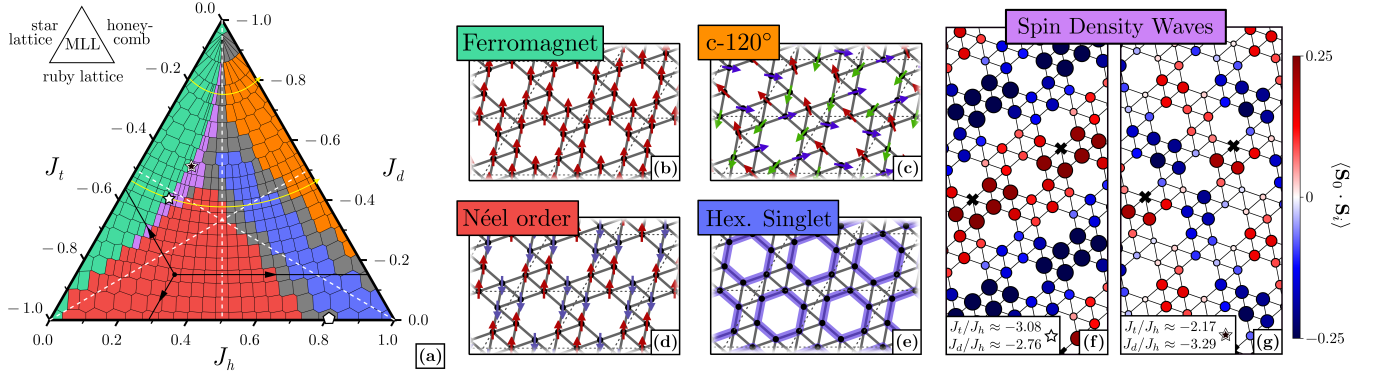


FIG. 2. (a): Color-coded ternary ($|J_t| + J_h + |J_d| = 1$) phase diagram of the Hamiltonian (1) on the MLL based on $N = 36$ ED data (shown as Voronoi tessellation), where the couplings $J_t, J_d \leq 0$ are ferromagnetic and $0 \leq J_h$ is antiferromagnetic. Data points which could not be conclusively attributed to a phase are drawn in gray. The three black lines with arrows pointing away from a common point illustrate how couplings should be read off. The paths of the phase diagram cuts shown in Fig. 3 are drawn as yellow arrows. As illustrated to the left of the diagram, the three external boundaries correspond to Heisenberg models on the ruby, star, and honeycomb lattices, respectively obtained by setting J_d, J_h or J_t to zero. Lines along which the magnitude of two couplings is equal are drawn in white, their intersection marking the “isotropic” point where $J_t = J_d = -J_h < 0$. The white pentagon in the bottom right of the phase diagram highlights the point at which the ED ground state data agrees well with the Gutzwiller-projected quantum spin liquid Ansatz Z1012 as classified in Ref. [13]. (b)-(e): Illustrations of the identified ferromagnetic, c-120°, collinear Néel, and hexagonal singlet phases. (f)-(g): Spin-density-wave regime exemplified by $N = 36$ spin-spin correlator data at the two points highlighted by stars inside the diagram (a).

glet (HS), and canted 120° (c-120°) phases, as well as a large spin-density-wave regime close to the FM boundary in which the ordering vector varies continuously and energy levels are compressed by orders of magnitude. This is to be contrasted to the previously hypothesized zero-field nematic phase [19], for which we do not find direct finite-size evidence. Furthermore, our data does not support the possibility of a dimerized hexagonal-singlet ground state [19, 35].

Methods. Our analysis is based on exact diagonalization (ED) using the XDiag library [38, 39] on size $N \in \{42, 36, 24, 18\}$ clusters. We resolve all irreducible representations (irreps) of their respective space groups as well as different values of the total $S_{\text{tot}}^z = \sum_i S_i^z$ magnetization, giving access to the $SU(2)$ quantum number S . Our identification of phases is based on the energy level and irrep structure of ED spectra, various ground state correlators, in particular structure factors, as well as tower-of-states (TOS) analysis. Furthermore, we perform targeted variational Monte Carlo studies of the candidate spin-liquid states classified in Ref. [13] on the same set of clusters, the latter being discussed in Sec. C of the Supplemental Material (SM) [40].

Phase Diagram. The resulting phase diagram of the $S = 1/2$ Hamiltonian (1) with $J_t, J_d \leq 0 \leq J_h$ is shown in the ternary plot of Fig. 2a. In agreement with Ref. [19] and the corner-sharing $0 \leq J_t, J_h, J_d$ phase diagram [22], we find a FM, a collinear Néel AFM, a c-120° order as well as the HS phase. At some of the scanned points, highlighted in gray in Fig. 2, we were unable to unambiguously classify the nature of the realized phase. For most of the collinear Néel $\leftrightarrow$ HS or the HS $\leftrightarrow$ c-120° phase boundaries we do not expect intermediate phases

although these regions are shown in gray. In particular, the dimerized hexagonal-singlet (d-HS) state (three dimers on each J_h -hexagon, breaking the point group from C_6 to C_3), conjectured to exist close to the boundary of the c-120° phases in Ref. [19] is not observed on $N \in \{36, 24, 18\}$ clusters. This not only follows from ground state correlators, but also from the fact that point-group-violating dimer states would be expected to produce a set of low-lying singlet states associated with the broken point-group symmetry, which are absent. As discussed in detail below, we see no indications for a spin-nematic phase near the FM boundary, as put forward in Ref. [19], but see clear signs of a spin-density wave instability in the purple region of Fig. 2a.

To illustrate how different phases can be inferred from the structure of low-lying eigenstates, two horizontal cuts, highlighted in yellow in Fig. 2a, are shown in Fig. 3. As expected, the ground state in the ferromagnetic phase belongs to the fully polarized $S = S_{\text{max}} = N/2$ sector with the first and second excited states being $S_{\text{max}} - 1$ and $S_{\text{max}} - 2$ levels respectively. In all other cases, an $S = 0$ (singlet) level is found to be the ground state and the excited states contain information about the realized phase. For instance, the collinear Néel (red background) region characteristically features a $\Gamma.C_2.B$ triplet ($S = 1$) and a $\Gamma.C_2.A$ quintuplet ($S = 2$), which matches the analytical TOS prediction (see Tab. 1 in Ref. [22]). The hexagonal singlet is characterized by a clear separation of energy levels according to S as well as a low-lying $\Gamma.C_2.B$ triplet level. Lastly, the TOS of the c-120° order can be analytically shown (see Ref. [22]) to be comprised of $\Gamma.C_6.A$, $\Gamma.C_6.B$, and $K.C_3.A$ levels in the thermodynamic limit, i.e., for each S on the $N = 36$ cluster we ex-

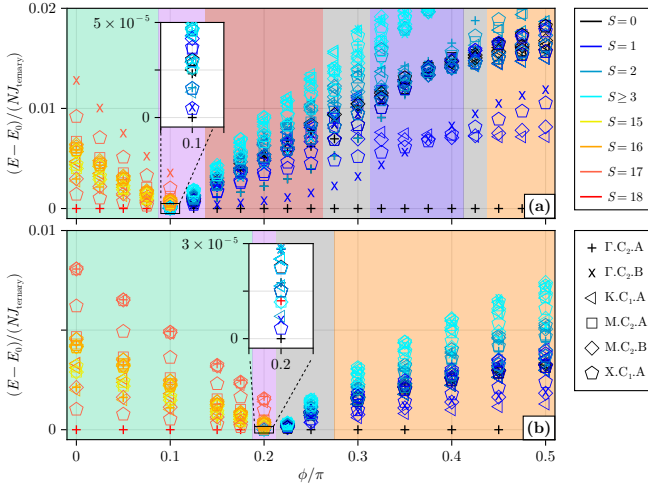


FIG. 3. Lowest energy levels along the two cuts illustrated in Fig. 2a as yellow arrows; (a) lower arrow at intermediate J_d ; (b) upper arrow at strong J_d . The angle ϕ originates from a spherical parametrization of the phase diagram in Fig. 2a; see Sec. D in the SM [40]. The energy scale is chosen such that $1 = J_{\text{ternary}} = |J_t| + J_h + |J_d|$. Background colors highlight the classification of phases in the color scheme of Fig. 2. Space-group irreps are fully resolved and SU(2) irreps labelled by S are shown for $S \leq 3$ outside and $S \geq 15$ inside the FM phase. Note that we cannot fully distinguish between $S = 3$ and $3 < S < 15$ levels without diagonalizing all $S_{\text{tot}}^z = 4$ representations, i.e., some of the $S \geq 3$ levels may actually belong to $3 < S < 15$ sectors. Note the strong compression of energy levels in the spin-density-wave region and the fact that an $S = 18$ level becomes one of the first excited states in the (b) inset (full $E_0(S)$ spectra in Fig. S2 in the SM [40]).

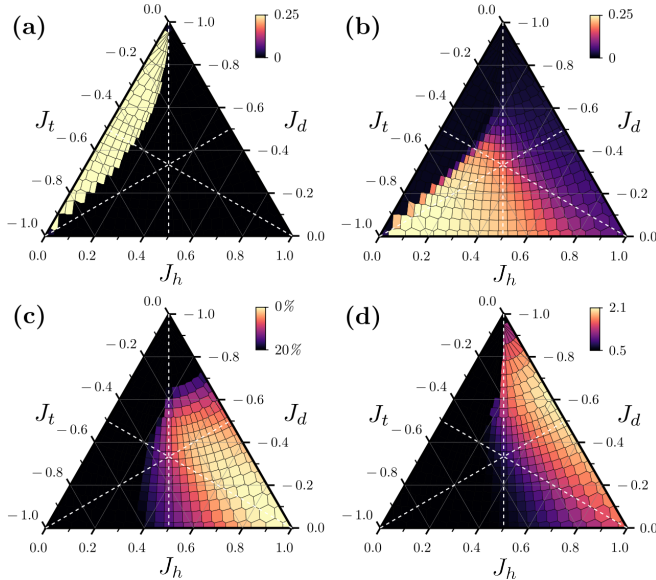


FIG. 4. Ground state correlators on the $N = 36$ MLL cluster. (a): Spin-spin correlator from Eq. (2); (b): Staggered spin-spin correlator from Eq. (3); (c): “Hexagonal singlet correlator” Δ_{HS} from Eq. (4); (d): Structure factor $\mathcal{S}(K_{\text{TRI}})$.

pect the lowest S level to be part of the Γ .C₂.A, Γ .C₂.B, or K.C₁.A irrep (as the finite cluster does not resolve the full little co-groups of each high symmetry point).

Further evidence from ground state correlators is shown in Fig. 4, respectively displaying the uniform (a) and staggered magnetization (b):

$$m = \frac{1}{N} \sum_{i=0}^{N-1} \langle \mathbf{S}_0 \cdot \mathbf{S}_i \rangle, \quad (2)$$

$$m_s = \frac{1}{N} \sum_{i=0}^{N-1} (-1)^i \langle \mathbf{S}_0 \cdot \mathbf{S}_i \rangle, \quad (3)$$

where $(-1)^i$ is used to denote the bipartition of the $J_t = 0$ (honeycomb) graph (see Fig. 2d). Here spin-spin correlators are needed since single-spin measurements are constrained by the fixed S_{tot}^z value. We find that the cleanest way of diagnosing the HS phase through ground state correlators is to compare the ED spin-spin correlations on J_h bonds to the value e_{HS} expected on isolated six-spin J_h rings

$$\Delta_{\text{HS}} = \frac{1}{N} \sum_{\langle i,j \rangle \in h} \frac{|\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle - e_{\text{HS}}|}{|e_{\text{HS}}|}, \quad (4)$$

which is shown in (c) of Fig. 4 and is near zero in the HS phase by construction. Although Δ_{HS} starts to deviate notably ($\approx 10\%$) from the pure HS spin-spin correlations north of the “isotropic point” we still attribute some of these data points to the HS phase as the ED spectra are still compatible with this scenario and spin-spin correlations are still dominated by antiferromagnetic intra-hexagon contributions and are near zero everywhere else. Lastly, it is well established through various numerical approaches [17–20, 22, 33] that the c-120° order features strong peaks of the equal-time spin-spins structure factor $\mathcal{S}(\mathbf{q})$ at the high-symmetry momentum K_{TRI} of the Brillouin zone of the underlying triangular lattice [41] which lies inside the extended Brillouin zone of the MLL (see Fig. 5). We thus show $\mathcal{S}(K_{\text{TRI}})$ in subplot (d), where the observed peak values are lower than in the purely antiferromagnetic Heisenberg model [22], however. This may hint at a stronger canting away from the uniform 120° order in the presence of FM interactions, which is supported by a very weak response of the “twist-twist” correlator [22] in that region, otherwise signaling a uniform 120° degree order.

Instabilities of the Zero-Field Ferromagnet The recent pf-FRG study [19] identified a region close to the FM boundary where the structure factor $\mathcal{S}(\mathbf{q})$ peaks along a continuous ring inside the MLL Brillouin zone. Combined with a strong response under an explicit breaking of the SU(2) spin symmetry down to U(1), this led to the hypothesis of a spin-nematic phase near the FM boundary [19]. It is well-established that quadrupolar (octupolar) spin-nematic phases are related to two-magnon (three-magnon) interactions and can hence be identified through low-lying $S = 2$ ($S = 3$) levels just outside the

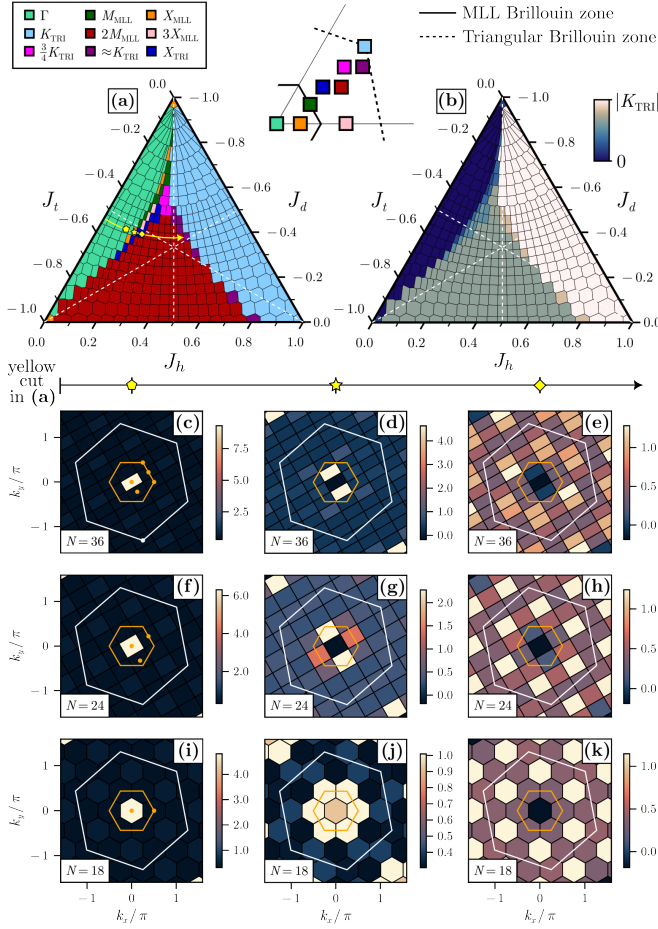


FIG. 5. (a): Map of reciprocal-lattice vectors $\mathbf{q}_{\text{max}}$ at which the structure factor $S(\mathbf{q})$ peaks on the $N = 36$ cluster, with an illustration of the extended Brillouin zone. (b): Distance from the $S(\mathbf{q})$ -maximizing reciprocal-lattice vector $\mathbf{q}_{\text{max}}$ on the $N = 36$ cluster to the Γ point, i.e., $|\mathbf{q}_{\text{max}}|$; (c)-(k): Structure factors along the yellow cut in (a) are shown in (c)-(e) for $N = 36$, (f)-(h) for $N = 24$, (i)-(k) for $N = 18$.

FM phase [2, 9, 42, 43]. Despite extensive high-resolution ED scans of the FM boundary on $N \in \{36, 24, 18\}$ clusters, we could not identify a single point at which such behavior is manifest. Moreover, on the $N = 36$ system we find the two-magnon interaction to be near zero or repulsive throughout the purple region in Fig. 2, and thus no evidence for a zero-field d -wave spin-nematic phase on the clusters studied here [2, 42] (data shown in Sec. A of the SM [40]). Instead, along the whole FM boundary we find ED spectra as in Fig. 3, where the low-energy spectrum collapses into a narrow window, with excitation energies reduced by more than an order of magnitude compared to neighboring phases, as the ground state flips from the $S = S_{\text{max}} = N/2$ to the $S = 0$ sector. Right after the transition, a regime of stripe orders is found where $S(\mathbf{q})$ peaks at different momenta $\mathbf{q}_{\text{max}}$ between the Γ point (FM phase) and the ordering vector of the respective proximate phase ($2M_{\text{MLL}}$ for the collinear Néel

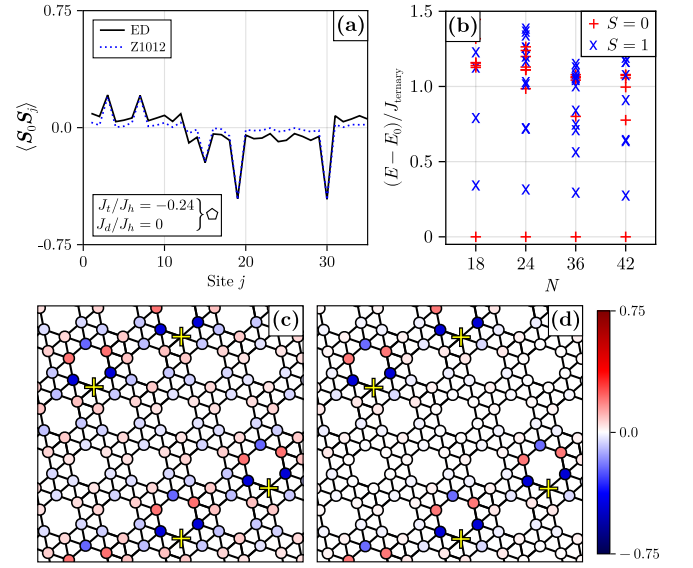


FIG. 6. (a) Spin-spin correlations in the ED ground state compared to the Gutzwiller-projected $\mathbb{Z}_2$ QSL Ansatz Z1012 on $N = 36$ sites at the point marked by a white pentagon in the phase diagram in Fig. 2. (b): Low-lying energy levels at the same point as in (a) but on different system sizes. The energy scale is chosen such that $1 = J_{\text{ternary}} = |J_t| + |J_h| + |J_d|$. (c) and (d): Spin-spin correlations from (a) drawn in real space with periodic boundary conditions for ED (c) and the Z1012 Ansatz (d).

AFM and K_{TRI} for the c -120° order). This continuous movement of the ordering vector away from Γ simultaneously occurs on clusters of different sizes as demonstrated in Fig. 5 along the cut shown in Fig. 3a for $N = 36$ in (c)-(e), for $N = 24$ in (f)-(h), and for $N = 18$ in (i)-(k). On the available clusters, this intermediate stripe regime is at least as prominent on the larger cluster as on the smaller ones, as exemplified by the fact that the structure factor in (e) has not yet reached the $2M_{\text{MLL}}$ point of the collinear Néel AFM phase, while (h) and (k) already show these characteristic Néel peaks. Subfigure (b) in Fig. 5 further shows the continuous nature of the structure factor peak moving away from Γ . Spin-spin correlations of two representative stripe patterns are shown in (f) and (g) of Fig. 2. The same regions are characterized by an unusually small TOS slope, indicating a large susceptibility to magnetic fields (see Fig. S2a in the SM [40]). At one point in the $J_t/J_h \lesssim 0$, $J_d/J_h \ll 0$ regime particularly close to the FM phase we even observe an arch-shaped behavior of the ground state energy $E_0(S)$ inside a fixed- S sector (see Fig. S2b in the SM [40]). If $E_0(S)$ retains this arch-shape with increasing system size, it signals a first-order transition at the upper regions of the FM boundary in Fig. 2 since the ground state then has to jump from the $S \approx 0$ sector into $S = S_{\text{max}}$. Thus, while the spectra show strong softening near the FM boundary, the finite-size ordering of spin sectors is closer to a soft incommensurate instability than to the standard

bound-magnon route to a zero-field quadrupolar phase.

Comparison to Fermionic Spin-Liquid Ansätze. Ref. [13] classified all fully symmetric Abrikosov-fermion $U(1)$ and $\mathbb{Z}_2$ QSL *Ansätze* on the MLL. We test these states by variational Monte Carlo (VMC), using Gutzwiller-projected wave functions to compare their energies and spin-spin correlations to the ED ground state. It is known that these variational descriptions work well for the $U(1)$ Dirac spin-liquids on the kagome and triangular lattices [3, 44, 45], reproducing the ED ground state with

$$\Delta_{\text{VMC}} = |(E_{\text{VMC}} - E_{\text{ED}})/E_{\text{ED}}| \quad (5)$$

of a few percent. Recently, we also reported on a promising regime in the nearest-neighbor maple-leaf fully antiferromagnetic model [22] where two Gutzwiller-projected $\mathbb{Z}_2$ *Ansätze* reach $\Delta_{\text{VMC}} \sim 10^{-3}$.

We repeat this analysis in the present ferro-antiferromagnetic setting, studying all twelve $U(1)$ and eight $\mathbb{Z}_2$ wave functions [13] at representative coupling values in the gray regions between the classified phases in Fig. 2a. At none of the interior points considered do these VMC wave functions reach competitive energies on the same $N = 36$ cluster. This distinguishes the gray unresolved regions from the fully symmetric fermionic spin-liquid manifold already classified for this lattice [13], rather than leaving it uncharacterized.

An exception is found on the $J_d = 0$ ruby (bounce) lattice boundary, at an intermediate point between the collinear Néel and HS phases marked by the white pentagon in Fig. 2a. At this point, the $\mathbb{Z}_2$ Ansatz Z1012, in the nomenclature of Ref. [13], yields a gapped wave function with an energy mismatch below 1%, i.e., $\Delta_{\text{VMC}} < 10^{-2}$. Moreover, the spin-spin correlations of the ED ground state are qualitatively well reproduced, as shown in Fig. 6, although the quantitative agreement is less precise than in the previously studied purely antiferromagnetic model [22]. The ED spectra at this point are displayed in Fig. 6b across different system sizes, showing that non-trivial low-lying singlet ($S = 0$) levels are present between the collinear Néel and HS phases on all clusters up to $N = 42$. It is thus possible that more singlet levels appear on larger clusters or that an extended model realizes a nearby QSL phase.

Discussion and Outlook. We investigated the phase diagram of the $J_t, J_d \leq 0 \leq J_h$ nearest-neighbor Heisenberg model on the maple-leaf lattice using symmetry-resolved exact diagonalization and tower-of-states analysis. While the ferromagnetic, collinear Néel antiferromagnetic, canted 120° , and hexagonal singlet phases were identified in agreement with another recent study [19], our data on $N \in \{36, 24, 18\}$ clusters do not support direct finite-size evidence for either of the hypothesized scenarios of a dimerized hexagonal singlet or a zero-field spin-nematic phase. The former is a dimer phase breaking the point group and must thus feature a low-lying singlet ($S = 0$) state, which is not observed in the respective regions of the phase diagram. On the

other hand, quadrupolar (octupolar) spin-nematic behavior is expected to be accompanied by low-lying $S = 2$ ($S = 3$) levels due to two-magnon (three-magnon) interactions [2, 9, 42], which are not observed anywhere along the boundary of the ferromagnetic phase and the two-magnon interaction appears to be repulsive. Instead, our data suggest that almost the entirety of the ferromagnetic phase in the $J_t, J_d \leq 0 \leq J_h$ Heisenberg model is destabilized by spin density waves whose ordering vectors continuously shift away from Γ to that of the neighboring magnetically ordered phases.

The high- S states of exact diagonalization spectra near the ferromagnetic boundary further display no anomalous behavior (such as an odd-even periodicity in the tower of states [2]; data shown in Fig. S2) which renders a spin-nematic phase induced through magnetic fields near saturation unlikely. Overall, the degree of frustration in the present $J_t, J_d \leq 0 \leq J_h$ model appears to be too weak to introduce the same type of multipolar or flat-band physics found on the triangular or kagome lattices [9–11]. The naturally emerging question is whether this is due to having only one antiferromagnetic coupling or whether the maple-leaf lattice itself lacks the sufficient degree of frustration.

Regarding materials, the presented $S = 1/2$ phase diagram is consistent with recent results on $\text{Na}_2\text{Mn}_3\text{O}_7$ which realizes a $S = 3/2$ maple-leaf geometry [26, 31, 34]. If the nine nearest-neighbor interactions obtained in Ref. [34] are averaged into three couplings values, one obtains (under ternary normalization) $J_h \approx 0.8$, $J_d \approx -0.12$, $J_t \approx -0.07$. $\text{Na}_2\text{Mn}_3\text{O}_7$ would thus be in the bottom right corner of Fig. 2a and inside the hexagonal singlet phase if it were a $S = 1/2$ material instead of $S = 3/2$. Our data thus match Ref. [34] reporting nearly isolated antiferromagnetic hexagons and rapidly decaying inter-hexagon correlations in the $S = 3/2$ case.

Using variational Monte Carlo we obtained Gutzwiller-projected wave functions of the fully symmetric fermionic quantum spin liquid *Ansätze* classified in Ref. [13]. We identify a point in the $J_d = 0$ ruby lattice limit at $J_t/J_h \approx -0.24$ between the collinear Néel and hexagonal singlet phases that is well described by a gapped $\mathbb{Z}_2$ Ansatz. This result could hint at a $\mathbb{Z}_2$ quantum spin liquid being realized at a nearby point in an extension of ferro-antiferromagnetic nearest-neighbor Heisenberg model on the ruby lattice – a geometry already realized in contemporary programmable Rydberg-array quantum-simulator platforms [46] and for which non-Heisenberg $\mathbb{Z}_2$ QSL phases are well-explored [47–49].

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Supplemental Material for

“Incommensurate Spin-Density Waves in a Frustrated Maple-Leaf Lattice Ferromagnet”

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Appendix A: Two-Magnon Interactions above Singlet Ground State

Spin-nematic phases are typically understood in the context of multi-magnon interactions, where the interaction has to be sufficiently strong for collective multi-magnon excitations to be preferred over forming one or more non-interacting magnons [2, 9]. Throughout the purple region in Fig. 2, which we like to probe for signs of spin-nematic behavior, the ground state is a singlet ($S = 0$) such that the n -magnon sector has $S = n$. If we define $E_0(S)$ as the lowest energy in the spin- S sector of the Hilbert space, we can give an estimate if, and to what degree, two-magnon interactions lower the excitation energy of the $S = 2$ sector. The energy associated with a single 1-magnon excitation is $E^{1,\text{free}} = E_0(1) - E_0(0)$ and forming a 2-magnon state corresponds to $E^{2,\text{interact.}} = E_0(2) - E_0(0)$. This means that

$$\Delta_2 = E^{2,\text{interact.}} - 2E^{1,\text{free}} = E_0(2) - 2E_0(1) + E_0(0), \quad (\text{S1})$$

becomes negative whenever two-magnon interactions lower the excitation energy of the $S = 2$ level compared to what is expected for two non-interacting magnons. Note that a more accurate computation of single or multi-magnon excitations would be concerned with momentum conservation constraints, which we neglect here. To the best of our knowledge, it is a necessary condition for d -wave spin-nematic phases to have $\Delta_2 < 0$ and similar condition exist for p -wave spin-nematic regimes where three-magnon interactions become important [9]. It is easy to see that $\Delta_2 < 0$ follows in clear-cut nematic phases where the $S = 2$ level becomes the first excited state and is thus lower in energy than the $S = 1$ level [42].

We show a heatmap of Δ_2 in Fig. S1 which is found to be zero or positive throughout the whole purple region near the ferromagnetic phase (note the purple and green lines respectively highlighting the extent of both phases). We therefore find no evidence for a d -wave spin-nematic phase in the purple region where spin-spin correlations display wave patterns.

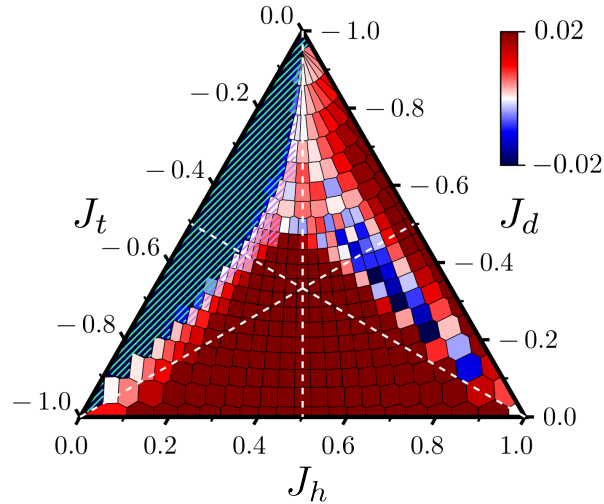


FIG. S1. Two-magnon interaction energy Δ_2 above a singlet ground state on the $N = 36$ cluster, as defined in Eq. (S1), where negative (blue) values indicate that two-magnon interactions lower the energy of the $S = 2$ level. Green (purple) lines are overlaid to show which data points are classified as the ferromagnetic (spin-density wave) phase in Fig. 2. Note that the blue color in the ferromagnetic region is meaningless since here the ground state is not a singlet and hence Δ_2 does not measure excitations above the ground state.

Appendix B: Ground State Energies as a Function of S near the Ferromagnetic Phase

As mentioned earlier, our ED data does not feature low-lying $S = 2$ or $S = 3$ levels that could lead to a zero-field spin-nematic phase. Moreover, no obvious anomalies are observed in the high- S regions of the spectrum which would warrant the expectation of a spin-nematic phase in non-zero magnetic fields (as e.g. in Ref. [2]). To illustrate this, we here show the full spectra at the two points corresponding to the insets in Fig. 3. The exact coordinates for subplot (a) are $J_t = -0.45$, $J_h = 0.146$, $J_d = -0.404$ or $\theta/\pi = 0.275$, $\phi/\pi = 0.1$ and for subplot (b) $J_t = -0.145$, $J_h = 0.106$, $J_d = -0.749$ or $\theta/\pi = 0.075$, $\phi/\pi = 0.2$, where the θ and ϕ angles refer to the spherical parametrization introduced in Sec. D below (see Eq. (S2)). Both plots only show the lowest-energy level for each space-group irrep on the $N = 36$ cluster in each spin- S sector.

The curve $E_0(S)$ (the lowest energy level with SU(2) quantum number S) that is observed throughout nearly all points in the spin density wave regime near the FM boundary is exemplified in (a). It is monotonically increasing (in particular no $S = 2$ or $S = 3$ level becomes the first excited state) and its low- S shape is generally convex (indicating e.g. a repulsive two-magnon interaction via Eq. (S1)). The high- S levels are observed to be only slightly concave and in most cases close to linear in S which implies that the prospects of observing a spin-nematic phase at non-zero fields in which these levels become the first excited states are low.

As briefly mentioned in the main text, our ED scans revealed a single point in the upper part of the phase diagram in Fig. 2a at which $E_0(S)$ follows an arch-type curve. The spectrum of this point, most likely astonishingly close to the FM phase transition, is shown in (b). Here the ground state is still in the non-polarized $S = 0$ sector while the fully-polarized $S = S_{\max}$ level becomes the sixth excited state (also see the inset in Fig. 3b). The system can thus be polarized by a small excitation and we generally expect $E_0(S)$ to maintain its shape moving closer towards the FM boundary, at which point the ground state jumps from $S = 0$ to $S = S_{\max}$. This, given that the shape of $E_0(S)$ is retained as system size is increased, presents a strong argument for a first-order transition in the $J_t/J_h \lesssim 0$, $J_d/J_h \ll 0$ regime of the FM phase boundary during which the magnetization jumps abruptly. We are currently unaware of another published ED spectrum so close to a FM phase boundary that $E_0(S)$ displays an arch-shape.

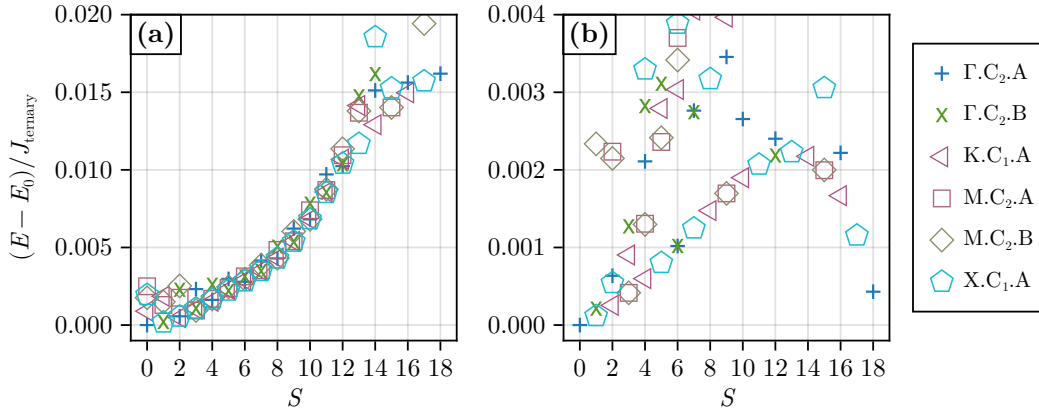


FIG. S2. Tower-of-states plot on the $N = 36$ cluster at the two exemplary spin-density-wave points shown as insets in Fig. 3 (a) and (b), respectively. The energy scale was chosen as to enforce $1 = J_{\text{ternary}} = |J_t| + J_h + |J_d|$. For each S we show the single lowest energy level in each space group irrep (see legend on the right). Note that only (a) displays the expected monotonous behavior while (b) shows an arch-shaped dependence of the ground state energy in each S sector.

Appendix C: Finite Size Clusters

In total, four different finite-size clusters of the MLL were used throughout this work, which are shown in Fig. S3. For each of the four system sizes $N \in \{42, 36, 24, 18\}$ we picked the most symmetric cluster of that size in the sense that important high-symmetry momenta are resolved (e.g. K_{MLL} , M_{MLL} , or X_{MLL} as defined in Fig. 5; the MLL subscript being dropped from now on).

Adopting the notation $\mathbf{k}.\hat{\mathcal{G}}(\mathbf{k})$ to denote that a cluster resolves momentum $\mathbf{k}$ and that the point group of the associated little group of $\mathbf{k}$ on that cluster is $\hat{\mathcal{G}}(\mathbf{k})$, the symmetries of the employed clusters can be summarized as follows: For $N = 18$ we resolve $\Gamma.C_6$ and $K.C_3$ (i.e. the full symmetries of the infinite MLL at both momenta). For

$N = 24$ we resolve $\Gamma.C_2$, $M.C_2$, and $Y.C_1$ where Y lies on the Γ -K line between X and K . For $N = 36$ we resolve $\Gamma.C_2$, $K.C_1$, $M.C_2$, and $X.C_1$, which makes it the most symmetric currently accessible cluster for phase diagram scans. For $N = 42$ we resolve $\Gamma.C_6$ and $O.C_1$, where O lies inside the Brillouin zone roughly between M and X .

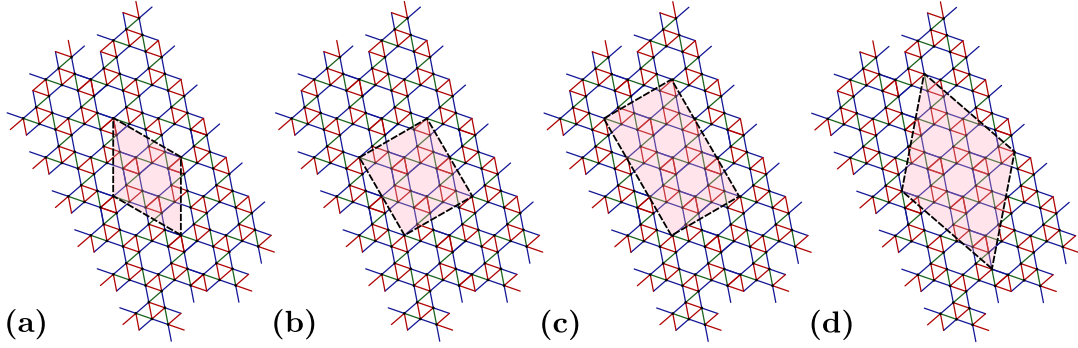


FIG. S3. Different finite size cluster used throughout this study: (a) $N = 18$, (b) $N = 24$, (c) $N = 36$, (d) $N = 42$.

Appendix D: Table of Classified Points and Couplings

The phase diagram of the Hamiltonian (1) with arbitrary $J_t, J_h, J_d \in \mathbb{R}$ can be mapped to the $J_t^2 + J_h^2 + J_d^2 = 1$ sphere by appropriately fixing the overall energy scale. Our scans of the $J_t, J_d \leq 0$ (ferromagnetic) $0 \leq J_h$ (antiferromagnetic) octant of this sphere are based on the spherical parametrization

$$J_t = -\cos \phi \sin \theta, \quad J_h = +\sin \phi \sin \theta, \quad J_d = -\cos \theta, \quad (S2)$$

under which the octant of interest is parametrized by $\phi, \theta \in [0, \pi/2]$. In the following, we show all coupling values for which at least the $S_{\text{tot}}^z = \sum_i S_i^z = 0, 1, 2$ sectors were scanned. At some data points we scanned considerably more sectors (see e.g. Fig. 3) and sometimes even every sector (see e.g. Fig. S2).

TAB. S1: List of coupling values inside the phase diagram scan of the Hamiltonian (1) with $J_t, J_d \leq 0 \leq J_h$. The angles $\theta, \phi \in [0, \pi/2]$ describe this model under the spherical parametrization from Eq. (S2) (i.e. under the $J_t^2 + J_h^2 + J_d^2 = 1$ constraint). The values given for J_t, J_h, J_d assume the ternary normalization $1 = |J_t| + J_h + |J_d|$ that we used throughout the main text and employed in the phase diagram in Fig. 2a. The ground state energies (per spin) also assume the ternary normalization and were obtained on the $N = 36$ cluster.

Index	θ/π	ϕ/π	J_t	J_h	J_d	classified phase	$E_0/N/J_{\text{ternary}}$
1	0	0	0	0	-1	isolated FM J_d - “dimers”	-0.125
2	0.025	0	-0.07296	0	-0.92704	FM	-0.13411996
3	0.025	0.05	-0.071312	0.011295	-0.91739	FM	-0.12967845
4	0.025	0.1	-0.068097	0.022126	-0.90978	FM	-0.12521485
5	0.025	0.15	-0.063411	0.03231	-0.90428	FM	-0.12081028
6	0.025	0.2	-0.057365	0.041678	-0.90096	FM	-0.11654135
7	0.025	0.225	-0.053868	0.046008	-0.90012	FM	-0.11448061
8	0.025	0.25	-0.050077	0.050077	-0.89985	inconclusive	-0.11411144
9	0.025	0.3	-0.041678	0.057365	-0.90096	inconclusive	-0.11723839
10	0.025	0.35	-0.03231	0.063411	-0.90428	inconclusive	-0.12061396
11	0.025	0.4	-0.022126	0.068097	-0.90978	inconclusive	-0.1241199
12	0.025	0.45	-0.011295	0.071312	-0.91739	inconclusive	-0.12767863
13	0.025	0.5	0	0.07296	-0.92704	inconclusive	-0.13121918
14	0.05	0	-0.13673	0	-0.86327	FM	-0.14209109
15	0.05	0.05	-0.13244	0.020976	-0.84659	FM	-0.13368855
16	0.05	0.1	-0.12557	0.040801	-0.83363	FM	-0.12539619
17	0.05	0.15	-0.11634	0.059277	-0.82438	FM	-0.11731328
18	0.05	0.2	-0.10492	0.076231	-0.81885	FM	-0.10952869
19	0.05	0.225	-0.098452	0.084086	-0.81746	spin density wave	-0.10640483
20	0.05	0.25	-0.0915	0.0915	-0.817	inconclusive	-0.10820926
21	0.05	0.3	-0.076231	0.10492	-0.81885	canted 120°	-0.11426367

Continued on next page

Table S1 – continued from previous page

Index	θ/π	ϕ/π	J_t	J_h	J_d	classified phase	$E_0/N/J_{\text{ternary}}$
22	0.05	0.35	-0.059277	0.11634	-0.82438	canted 120°	-0.12069881
23	0.05	0.4	-0.040801	0.12557	-0.83363	canted 120°	-0.12731359
24	0.05	0.45	-0.020976	0.13244	-0.84659	canted 120°	-0.13398745
25	0.05	0.5	0	0.13673	-0.86327	canted 120°	-0.14062242
26	0.075	0	-0.1936	0	-0.8064	FM	-0.14919995
27	0.075	0.05	-0.18603	0.029464	-0.78451	FM	-0.13720436
28	0.075	0.1	-0.1753	0.056958	-0.76774	FM	-0.12555308
29	0.075	0.15	-0.1617	0.082389	-0.75591	FM	-0.11431625
30	0.075	0.1625	-0.15788	0.088416	-0.75371	FM	-0.11157883
31	0.075	0.175	-0.15389	0.094306	-0.7518	FM	-0.10887186
32	0.075	0.2	-0.14545	0.10568	-0.74887	spin density wave	-0.1035646
33	0.075	0.225	-0.13639	0.11649	-0.74712	inconclusive	-0.10224623
34	0.075	0.25	-0.12673	0.12673	-0.74653	inconclusive	-0.10612757
35	0.075	0.3	-0.10568	0.14545	-0.74887	canted 120°	-0.11485434
36	0.075	0.35	-0.082389	0.1617	-0.75591	canted 120°	-0.12400565
37	0.075	0.4	-0.056958	0.1753	-0.76774	canted 120°	-0.13334582
38	0.075	0.45	-0.029464	0.18603	-0.78451	canted 120°	-0.14275356
39	0.075	0.5	0	0.1936	-0.8064	canted 120°	-0.15214618
40	0.1	0	-0.24524	0	-0.75476	FM	-0.15565466
41	0.1	0.05	-0.23395	0.037054	-0.729	FM	-0.14034843
42	0.1	0.1	-0.21925	0.071239	-0.70951	FM	-0.12569176
43	0.1	0.15	-0.20146	0.10265	-0.69589	FM	-0.11168893
44	0.1	0.1625	-0.19656	0.11008	-0.69336	FM	-0.10829031
45	0.1	0.175	-0.19148	0.11734	-0.69118	FM	-0.10493252
46	0.1	0.2	-0.18081	0.13136	-0.68783	spin density wave	-0.10032425
47	0.1	0.25	-0.15742	0.15742	-0.68516	inconclusive	-0.10699477
48	0.1	0.3	-0.13136	0.18081	-0.68783	canted 120°	-0.11803905
49	0.1	0.35	-0.10265	0.20146	-0.69589	canted 120°	-0.12951705
50	0.1	0.4	-0.071239	0.21925	-0.70951	canted 120°	-0.14120423
51	0.1	0.45	-0.037054	0.23395	-0.729	canted 120°	-0.15301534
52	0.1	0.5	0	0.24524	-0.75476	canted 120°	-0.16491625
53	0.125	0	-0.29289	0	-0.70711	FM	-0.16161165
54	0.125	0.05	-0.27757	0.043963	-0.67847	FM	-0.14321023
55	0.125	0.1	-0.25884	0.084103	-0.65706	FM	-0.12581667
56	0.125	0.15	-0.23702	0.12077	-0.64221	FM	-0.10933963
57	0.125	0.1625	-0.2311	0.12942	-0.63947	FM	-0.10535378
58	0.125	0.175	-0.22501	0.13789	-0.6371	spin density wave	-0.10180365
59	0.125	0.2	-0.21228	0.15423	-0.63348	spin density wave	-0.099883595
60	0.125	0.25	-0.1847	0.1847	-0.6306	inconclusive	-0.11017238
61	0.125	0.3	-0.15423	0.21228	-0.63348	inconclusive	-0.12311615
62	0.125	0.35	-0.12077	0.23702	-0.64221	canted 120°	-0.13652068
63	0.125	0.4	-0.084103	0.25884	-0.65706	canted 120°	-0.1502066
64	0.125	0.45	-0.043963	0.27757	-0.67847	canted 120°	-0.16414778
65	0.125	0.5	0	0.29289	-0.70711	canted 120°	-0.17838092
66	0.15	0	-0.33754	0	-0.66246	FM	-0.16719252
67	0.15	0.05	-0.31792	0.050353	-0.63173	FM	-0.14585732
68	0.15	0.1	-0.29511	0.095888	-0.609	FM	-0.12593112
69	0.15	0.15	-0.26938	0.13726	-0.59336	FM	-0.10720145
70	0.15	0.1625	-0.26251	0.14701	-0.59048	spin density wave	-0.10324734
71	0.15	0.175	-0.25545	0.15654	-0.588	spin density wave	-0.10173684
72	0.15	0.2	-0.24082	0.17497	-0.58421	inconclusive	-0.10235917
73	0.15	0.25	-0.2094	0.2094	-0.5812	inconclusive	-0.11522796
74	0.15	0.3	-0.17497	0.24082	-0.58421	inconclusive	-0.12961545
75	0.15	0.35	-0.13726	0.26938	-0.59336	inconclusive	-0.14456108
76	0.15	0.4	-0.095888	0.29511	-0.609	canted 120°	-0.15993983
77	0.15	0.45	-0.050353	0.31792	-0.63173	canted 120°	-0.17579256
78	0.15	0.5	0	0.33754	-0.66246	canted 120°	-0.1922403
79	0.175	0	-0.37996	0	-0.62004	FM	-0.17249508
80	0.175	0.05	-0.3558	0.056353	-0.58785	FM	-0.14834248
81	0.175	0.1	-0.32887	0.10686	-0.56428	FM	-0.12603761
82	0.175	0.125	-0.31441	0.13023	-0.55535	FM	-0.11546379

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Index	θ/π	ϕ/π	J_t	J_h	J_d	classified phase	$E_0/N/J_{\text{ternary}}$
83	0.175	0.1375	-0.30694	0.1415	-0.55155	FM	-0.1103043
84	0.175	0.15	-0.29931	0.15251	-0.54818	spin density wave	-0.10598752
85	0.175	0.2	-0.26712	0.19407	-0.5388	inconclusive	-0.10801606
86	0.175	0.25	-0.23214	0.23214	-0.53572	hexagonal singlet	-0.12184519
87	0.175	0.3	-0.19407	0.26712	-0.5388	hexagonal singlet	-0.13722382
88	0.175	0.35	-0.15251	0.29931	-0.54818	inconclusive	-0.15335999
89	0.175	0.4	-0.10686	0.32887	-0.56428	canted 120°	-0.17017244
90	0.175	0.45	-0.056353	0.3558	-0.58785	canted 120°	-0.18776706
91	0.175	0.5	0	0.37996	-0.62004	canted 120°	-0.20635884
92	0.2	0	-0.42081	0	-0.57919	FM	-0.17760097
93	0.2	0.025	-0.40661	0.032001	-0.56139	FM	-0.16382622
94	0.2	0.05	-0.39186	0.062065	-0.54607	FM	-0.15070839
95	0.2	0.075	-0.37657	0.090406	-0.53303	FM	-0.13816872
96	0.2	0.1	-0.36073	0.11721	-0.52206	FM	-0.12613815
97	0.2	0.125	-0.34435	0.14264	-0.51301	FM	-0.11455575
98	0.2	0.1375	-0.33595	0.15488	-0.50917	spin density wave	-0.10980965
99	0.2	0.15	-0.32741	0.16682	-0.50577	spin density wave	-0.10826538
100	0.2	0.175	-0.30988	0.18989	-0.50023	spin density wave	-0.10987365
101	0.2	0.2	-0.29173	0.21195	-0.49632	inconclusive	-0.11542369
102	0.2	0.225	-0.27292	0.23309	-0.49399	inconclusive	-0.12231738
103	0.2	0.25	-0.25339	0.25339	-0.49322	hexagonal singlet	-0.12976895
104	0.2	0.275	-0.23309	0.27292	-0.49399	hexagonal singlet	-0.13760019
105	0.2	0.3	-0.21195	0.29173	-0.49632	hexagonal singlet	-0.14573034
106	0.2	0.325	-0.18989	0.30988	-0.50023	hexagonal singlet	-0.15411987
107	0.2	0.35	-0.16682	0.32741	-0.50577	hexagonal singlet	-0.16275499
108	0.2	0.375	-0.14264	0.34435	-0.51301	inconclusive	-0.17163931
109	0.2	0.4	-0.11721	0.36073	-0.52206	canted 120°	-0.180789
110	0.2	0.425	-0.090406	0.37657	-0.53303	canted 120°	-0.19023032
111	0.2	0.45	-0.062065	0.39186	-0.54607	canted 120°	-0.19999863
112	0.2	0.475	-0.032001	0.40661	-0.56139	canted 120°	-0.21013833
113	0.2	0.5	0	0.42081	-0.57919	canted 120°	-0.22070351
114	0.225	0	-0.46065	0	-0.53935	FM	-0.18258114
115	0.225	0.025	-0.44382	0.034929	-0.52125	FM	-0.16737886
116	0.225	0.05	-0.42665	0.067575	-0.50577	FM	-0.15299088
117	0.225	0.075	-0.40913	0.098224	-0.49264	FM	-0.13930754
118	0.225	0.1	-0.39123	0.12712	-0.48165	FM	-0.12623438
119	0.225	0.1125	-0.38213	0.14098	-0.47689	FM	-0.11990054
120	0.225	0.125	-0.37292	0.15447	-0.47261	spin density wave	-0.11457716
121	0.225	0.1375	-0.3636	0.16762	-0.46878	spin density wave	-0.11301951
122	0.225	0.15	-0.35416	0.18045	-0.46539	spin density wave	-0.11358425
123	0.225	0.175	-0.3349	0.20522	-0.45988	spin density wave	-0.11809405
124	0.225	0.2	-0.31508	0.22892	-0.456	inconclusive	-0.12447076
125	0.225	0.225	-0.29465	0.25166	-0.45369	inconclusive	-0.13141215
126	0.225	0.25	-0.27354	0.27354	-0.45293	inconclusive	-0.13883877
127	0.225	0.275	-0.25166	0.29465	-0.45369	hexagonal singlet	-0.14672038
128	0.225	0.3	-0.22892	0.31508	-0.456	hexagonal singlet	-0.15500764
129	0.225	0.325	-0.20522	0.3349	-0.45988	hexagonal singlet	-0.16366106
130	0.225	0.35	-0.18045	0.35416	-0.46539	hexagonal singlet	-0.17266366
131	0.225	0.375	-0.15447	0.37292	-0.47261	inconclusive	-0.18201949
132	0.225	0.4	-0.12712	0.39123	-0.48165	inconclusive	-0.19174945
133	0.225	0.425	-0.098224	0.40913	-0.49264	canted 120°	-0.20188847
134	0.225	0.45	-0.067575	0.42665	-0.50577	canted 120°	-0.21248443
135	0.225	0.475	-0.034929	0.44382	-0.52125	canted 120°	-0.22359851
136	0.225	0.5	0	0.46065	-0.53935	canted 120°	-0.23530675
137	0.25	0	-0.5	0	-0.5	FM	-0.1875
138	0.25	0.025	-0.48035	0.037805	-0.48184	FM	-0.17086759
139	0.25	0.05	-0.46065	0.07296	-0.46639	FM	-0.15522127
140	0.25	0.075	-0.44082	0.10583	-0.45335	FM	-0.14041572
141	0.25	0.1	-0.42081	0.13673	-0.44246	FM	-0.1263277
142	0.25	0.1125	-0.41071	0.15152	-0.43777	spin density wave	-0.12024641
143	0.25	0.125	-0.40054	0.16591	-0.43355	spin density wave	-0.11868162

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Index	θ/π	ϕ/π	J_t	J_h	J_d	classified phase	$E_0/N/J_{\text{ternary}}$
144	0.25	0.1375	-0.3903	0.17993	-0.42977	spin density wave	-0.11983474
145	0.25	0.15	-0.37996	0.1936	-0.42644	inconclusive	-0.12214282
146	0.25	0.175	-0.35899	0.21999	-0.42103	collinear Néel AFM	-0.12833451
147	0.25	0.2	-0.33754	0.24524	-0.41722	collinear Néel AFM	-0.13487654
148	0.25	0.225	-0.31554	0.2695	-0.41496	collinear Néel AFM	-0.14171285
149	0.25	0.25	-0.29289	0.29289	-0.41421	inconclusive	-0.14896071
150	0.25	0.275	-0.2695	0.31554	-0.41496	inconclusive	-0.15671781
151	0.25	0.3	-0.24524	0.33754	-0.41722	hexagonal singlet	-0.16500441
152	0.25	0.325	-0.21999	0.35899	-0.42103	hexagonal singlet	-0.17379423
153	0.25	0.35	-0.1936	0.37996	-0.42644	hexagonal singlet	-0.18306413
154	0.25	0.375	-0.16591	0.40054	-0.43355	inconclusive	-0.19281368
155	0.25	0.4	-0.13673	0.42081	-0.44246	inconclusive	-0.20306579
156	0.25	0.425	-0.10583	0.44082	-0.45335	inconclusive	-0.21386356
157	0.25	0.45	-0.07296	0.46065	-0.46639	canted 120°	-0.2252687
158	0.25	0.475	-0.037805	0.48035	-0.48184	canted 120°	-0.23736207
159	0.25	0.5	0	0.5	-0.5	canted 120°	-0.25024623
160	0.275	0	-0.53935	0	-0.46065	FM	-0.19241886
161	0.275	0.025	-0.51668	0.040664	-0.44265	FM	-0.17433642
162	0.275	0.05	-0.49429	0.078288	-0.42742	FM	-0.15742819
163	0.275	0.075	-0.47205	0.11333	-0.41462	FM	-0.14150774
164	0.275	0.0875	-0.46095	0.13	-0.40905	FM	-0.13386827
165	0.275	0.1	-0.44985	0.14617	-0.40398	spin density wave	-0.12686931
166	0.275	0.1125	-0.43874	0.16186	-0.3994	spin density wave	-0.12532098
167	0.275	0.125	-0.42759	0.17712	-0.39529	spin density wave	-0.12695078
168	0.275	0.15	-0.40517	0.20645	-0.38838	collinear Néel AFM	-0.13309923
169	0.275	0.175	-0.38248	0.23439	-0.38313	collinear Néel AFM	-0.13960434
170	0.275	0.2	-0.35942	0.26114	-0.37944	collinear Néel AFM	-0.14619782
171	0.275	0.225	-0.33588	0.28687	-0.37726	collinear Néel AFM	-0.15295323
172	0.275	0.25	-0.31174	0.31174	-0.37653	collinear Néel AFM	-0.16001763
173	0.275	0.275	-0.28687	0.33588	-0.37726	inconclusive	-0.16756679
174	0.275	0.3	-0.26114	0.35942	-0.37944	inconclusive	-0.17572803
175	0.275	0.325	-0.23439	0.38248	-0.38313	hexagonal singlet	-0.18453771
176	0.275	0.35	-0.20645	0.40517	-0.38838	hexagonal singlet	-0.19398373
177	0.275	0.375	-0.17712	0.42759	-0.39529	hexagonal singlet	-0.20406042
178	0.275	0.4	-0.14617	0.44985	-0.40398	hexagonal singlet	-0.21478937
179	0.275	0.425	-0.11333	0.47205	-0.41462	inconclusive	-0.22622065
180	0.275	0.45	-0.078288	0.49429	-0.42742	canted 120°	-0.23843101
181	0.275	0.475	-0.040664	0.51668	-0.44265	canted 120°	-0.25152434
182	0.275	0.5	0	0.53935	-0.46065	canted 120°	-0.26563529
183	0.3	0	-0.57919	0	-0.42081	FM	-0.19739903
184	0.3	0.025	-0.55325	0.043542	-0.4032	FM	-0.17782841
185	0.3	0.05	-0.52799	0.083625	-0.38839	FM	-0.15963908
186	0.3	0.075	-0.5032	0.12081	-0.37599	FM	-0.14259727
187	0.3	0.0875	-0.49094	0.13846	-0.3706	spin density wave	-0.13466549
188	0.3	0.1	-0.47873	0.15555	-0.36572	spin density wave	-0.13283661
189	0.3	0.1125	-0.46656	0.17212	-0.36131	collinear Néel AFM	-0.13507543
190	0.3	0.125	-0.45442	0.18823	-0.35736	collinear Néel AFM	-0.13832926
191	0.3	0.15	-0.43012	0.21916	-0.35073	collinear Néel AFM	-0.14497304
192	0.3	0.175	-0.40569	0.24861	-0.3457	collinear Néel AFM	-0.15160197
193	0.3	0.2	-0.38101	0.27682	-0.34217	collinear Néel AFM	-0.15823158
194	0.3	0.225	-0.35593	0.30399	-0.34008	collinear Néel AFM	-0.16494577
195	0.3	0.25	-0.33031	0.33031	-0.33939	collinear Néel AFM	-0.17188426
196	0.3	0.275	-0.30399	0.35593	-0.34008	inconclusive	-0.17923984
197	0.3	0.3	-0.27682	0.38101	-0.34217	inconclusive	-0.18721726
198	0.3	0.325	-0.24861	0.40569	-0.3457	hexagonal singlet	-0.19595374
199	0.3	0.35	-0.21916	0.43012	-0.35073	hexagonal singlet	-0.20549186
200	0.3	0.375	-0.18823	0.45442	-0.35736	hexagonal singlet	-0.21583554
201	0.3	0.4	-0.15555	0.47873	-0.36572	hexagonal singlet	-0.22700514
202	0.3	0.425	-0.12081	0.5032	-0.37599	inconclusive	-0.23905624
203	0.3	0.45	-0.083625	0.52799	-0.38839	inconclusive	-0.25208157
204	0.3	0.475	-0.043542	0.55325	-0.4032	canted 120°	-0.26621181

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Index	θ/π	ϕ/π	J_t	J_h	J_d	classified phase	$E_0/N/J_{\text{ternary}}$
205	0.3	0.5	0	0.57919	-0.42081	canted 120°	-0.28162043
206	0.325	0	-0.62004	0	-0.37996	FM	-0.20250492
207	0.325	0.025	-0.59053	0.046476	-0.363	FM	-0.18138774
208	0.325	0.05	-0.56217	0.089039	-0.34879	FM	-0.16188158
209	0.325	0.075	-0.53468	0.12836	-0.33696	FM	-0.14369786
210	0.325	0.0875	-0.52118	0.14699	-0.33184	spin density wave	-0.14118839
211	0.325	0.1	-0.50781	0.165	-0.3272	collinear Néel AFM	-0.14398527
212	0.325	0.125	-0.48135	0.19938	-0.31927	collinear Néel AFM	-0.1508139
213	0.325	0.15	-0.45511	0.23189	-0.31301	collinear Néel AFM	-0.15759222
214	0.325	0.175	-0.42891	0.26283	-0.30826	collinear Néel AFM	-0.16428449
215	0.325	0.2	-0.40258	0.29249	-0.30494	collinear Néel AFM	-0.17093622
216	0.325	0.225	-0.37594	0.32109	-0.30297	collinear Néel AFM	-0.17763137
217	0.325	0.25	-0.34884	0.34884	-0.30232	collinear Néel AFM	-0.18449751
218	0.325	0.275	-0.32109	0.37594	-0.30297	collinear Néel AFM	-0.19171704
219	0.325	0.3	-0.29249	0.40258	-0.30494	inconclusive	-0.19952152
220	0.325	0.325	-0.26283	0.42891	-0.30826	inconclusive	-0.20813514
221	0.325	0.35	-0.23189	0.45511	-0.31301	hexagonal singlet	-0.21769514
222	0.325	0.375	-0.19938	0.48135	-0.31927	hexagonal singlet	-0.22825038
223	0.325	0.4	-0.165	0.50781	-0.3272	hexagonal singlet	-0.23983067
224	0.325	0.425	-0.12836	0.53468	-0.33696	hexagonal singlet	-0.25249784
225	0.325	0.45	-0.089039	0.56217	-0.34879	inconclusive	-0.26636156
226	0.325	0.475	-0.046476	0.59053	-0.363	canted 120°	-0.28158349
227	0.325	0.5	0	0.62004	-0.37996	canted 120°	-0.29838405
228	0.35	0	-0.66246	0	-0.33754	FM	-0.20780748
229	0.35	0.025	-0.62901	0.049504	-0.32149	FM	-0.18506208
230	0.35	0.05	-0.59728	0.0946	-0.30812	FM	-0.16418495
231	0.35	0.0625	-0.58193	0.11575	-0.30232	FM	-0.15433384
232	0.35	0.075	-0.56687	0.13609	-0.29704	spin density wave	-0.15050528
233	0.35	0.0875	-0.55205	0.15569	-0.29226	collinear Néel AFM	-0.15370321
234	0.35	0.1	-0.53744	0.17463	-0.28793	collinear Néel AFM	-0.15721364
235	0.35	0.125	-0.50872	0.21072	-0.28056	collinear Néel AFM	-0.16416878
236	0.35	0.15	-0.48045	0.2448	-0.27475	collinear Néel AFM	-0.17098969
237	0.35	0.175	-0.45241	0.27724	-0.27035	collinear Néel AFM	-0.17770064
238	0.35	0.2	-0.42439	0.30833	-0.26728	collinear Néel AFM	-0.18435739
239	0.35	0.225	-0.39617	0.33836	-0.26546	collinear Néel AFM	-0.1910428
240	0.35	0.25	-0.36757	0.36757	-0.26486	collinear Néel AFM	-0.19787432
241	0.35	0.275	-0.33836	0.39617	-0.26546	collinear Néel AFM	-0.2050183
242	0.35	0.3	-0.30833	0.42439	-0.26728	collinear Néel AFM	-0.21270208
243	0.35	0.325	-0.27724	0.45241	-0.27035	inconclusive	-0.22119527
244	0.35	0.35	-0.2448	0.48045	-0.27475	inconclusive	-0.23073397
245	0.35	0.375	-0.21072	0.50872	-0.28056	hexagonal singlet	-0.24145318
246	0.35	0.4	-0.17463	0.53744	-0.28793	hexagonal singlet	-0.25341926
247	0.35	0.425	-0.13609	0.56687	-0.29704	hexagonal singlet	-0.26670775
248	0.35	0.45	-0.0946	0.59728	-0.30812	inconclusive	-0.28144769
249	0.35	0.475	-0.049504	0.62901	-0.32149	canted 120°	-0.29783668
250	0.35	0.5	0	0.66246	-0.33754	canted 120°	-0.3161519
251	0.375	0	-0.70711	0	-0.29289	FM	-0.21338835
252	0.375	0.025	-0.66926	0.052672	-0.27807	FM	-0.18890517
253	0.375	0.05	-0.63381	0.10039	-0.2658	FM	-0.1665816
254	0.375	0.0625	-0.61681	0.12269	-0.2605	spin density wave	-0.16101557
255	0.375	0.075	-0.60022	0.1441	-0.25568	collinear Néel AFM	-0.16443912
256	0.375	0.1	-0.56804	0.18457	-0.2474	collinear Néel AFM	-0.17153429
257	0.375	0.125	-0.5369	0.22239	-0.24071	collinear Néel AFM	-0.1784772
258	0.375	0.15	-0.50648	0.25806	-0.23545	collinear Néel AFM	-0.18527213
259	0.375	0.175	-0.47651	0.292	-0.23149	collinear Néel AFM	-0.19195995
260	0.375	0.2	-0.44672	0.32456	-0.22872	collinear Néel AFM	-0.19860148
261	0.375	0.225	-0.41687	0.35604	-0.22708	collinear Néel AFM	-0.20527845
262	0.375	0.25	-0.38673	0.38673	-0.22654	collinear Néel AFM	-0.21210086
263	0.375	0.275	-0.35604	0.41687	-0.22708	collinear Néel AFM	-0.21922058
264	0.375	0.3	-0.32456	0.44672	-0.22872	collinear Néel AFM	-0.22684957
265	0.375	0.325	-0.292	0.47651	-0.23149	collinear Néel AFM	-0.23526742

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Table S1 – continued from previous page

Index	θ/π	ϕ/π	J_t	J_h	J_d	classified phase	$E_0/N/J_{\text{ternary}}$
266	0.375	0.35	-0.25806	0.50648	-0.23545	inconclusive	-0.24478084
267	0.375	0.375	-0.22239	0.5369	-0.24071	hexagonal singlet	-0.25563309
268	0.375	0.4	-0.18457	0.56804	-0.2474	hexagonal singlet	-0.26796668
269	0.375	0.425	-0.1441	0.60022	-0.25568	hexagonal singlet	-0.28189093
270	0.375	0.45	-0.10039	0.63381	-0.2658	inconclusive	-0.29756108
271	0.375	0.475	-0.052672	0.66926	-0.27807	inconclusive	-0.31521731
272	0.375	0.5	0	0.70711	-0.29289	canted 120°	-0.33520586
273	0.4	0	-0.75476	0	-0.24524	FM	-0.21934534
274	0.4	0.025	-0.71193	0.05603	-0.23204	FM	-0.19298027
275	0.4	0.0375	-0.69178	0.081878	-0.22634	FM	-0.18076845
276	0.4	0.05	-0.67233	0.10649	-0.22118	collinear Néel AFM	-0.1730302
277	0.4	0.0625	-0.65351	0.12999	-0.2165	collinear Néel AFM	-0.17652431
278	0.4	0.075	-0.63523	0.15251	-0.21226	collinear Néel AFM	-0.18007819
279	0.4	0.1	-0.60004	0.19496	-0.205	collinear Néel AFM	-0.18707018
280	0.4	0.125	-0.56628	0.23456	-0.19916	collinear Néel AFM	-0.19390279
281	0.4	0.15	-0.53356	0.27186	-0.19457	collinear Néel AFM	-0.20060559
282	0.4	0.175	-0.50154	0.30734	-0.19112	collinear Néel AFM	-0.20722659
283	0.4	0.2	-0.46989	0.34139	-0.18872	collinear Néel AFM	-0.21382901
284	0.4	0.225	-0.43833	0.37437	-0.1873	collinear Néel AFM	-0.2204934
285	0.4	0.25	-0.40659	0.40659	-0.18683	collinear Néel AFM	-0.22732423
286	0.4	0.275	-0.37437	0.43833	-0.1873	collinear Néel AFM	-0.23446208
287	0.4	0.3	-0.34139	0.46989	-0.18872	collinear Néel AFM	-0.24210241
288	0.4	0.325	-0.30734	0.50154	-0.19112	collinear Néel AFM	-0.25051576
289	0.4	0.35	-0.27186	0.53356	-0.19457	inconclusive	-0.26004291
290	0.4	0.375	-0.23456	0.56628	-0.19916	inconclusive	-0.27102665
291	0.4	0.4	-0.19496	0.60004	-0.205	hexagonal singlet	-0.2837221
292	0.4	0.425	-0.15251	0.63523	-0.21226	hexagonal singlet	-0.29830781
293	0.4	0.45	-0.10649	0.67233	-0.22118	hexagonal singlet	-0.31498145
294	0.4	0.475	-0.05603	0.71193	-0.23204	inconclusive	-0.33403625
295	0.4	0.5	0	0.75476	-0.24524	canted 120°	-0.35590354
296	0.425	0	-0.8064	0	-0.1936	FM	-0.22580005
297	0.425	0.025	-0.75785	0.059644	-0.18251	FM	-0.19736469
298	0.425	0.0375	-0.73523	0.087021	-0.17775	collinear Néel AFM	-0.18700696
299	0.425	0.05	-0.71354	0.11301	-0.17344	collinear Néel AFM	-0.19043632
300	0.425	0.0625	-0.69267	0.13778	-0.16955	collinear Néel AFM	-0.19389121
301	0.425	0.075	-0.6725	0.16145	-0.16604	collinear Néel AFM	-0.19731383
302	0.425	0.1	-0.63397	0.20599	-0.16004	collinear Néel AFM	-0.20405698
303	0.425	0.125	-0.59735	0.24743	-0.15523	collinear Néel AFM	-0.21067996
304	0.425	0.15	-0.56212	0.28642	-0.15146	collinear Néel AFM	-0.21721887
305	0.425	0.175	-0.52788	0.32348	-0.14864	collinear Néel AFM	-0.22372397
306	0.425	0.2	-0.49424	0.35909	-0.14667	collinear Néel AFM	-0.23025865
307	0.425	0.225	-0.46087	0.39362	-0.14551	collinear Néel AFM	-0.23690179
308	0.425	0.25	-0.42744	0.42744	-0.14512	collinear Néel AFM	-0.24375359
309	0.425	0.275	-0.39362	0.46087	-0.14551	collinear Néel AFM	-0.25094624
310	0.425	0.3	-0.35909	0.49424	-0.14667	collinear Néel AFM	-0.25866137
311	0.425	0.325	-0.32348	0.52788	-0.14864	collinear Néel AFM	-0.26715427
312	0.425	0.35	-0.28642	0.56212	-0.15146	collinear Néel AFM	-0.27677226
313	0.425	0.375	-0.24743	0.59735	-0.15523	inconclusive	-0.2879277
314	0.425	0.4	-0.20599	0.63397	-0.16004	hexagonal singlet	-0.30100377
315	0.425	0.425	-0.16145	0.6725	-0.16604	hexagonal singlet	-0.31629354
316	0.425	0.45	-0.11301	0.71354	-0.17344	hexagonal singlet	-0.33406868
317	0.425	0.475	-0.059644	0.75785	-0.18251	inconclusive	-0.35469399
318	0.425	0.5	0	0.8064	-0.1936	inconclusive	-0.37870809
319	0.45	0	-0.86327	0	-0.13673	FM	-0.23290891
320	0.45	0.0125	-0.83487	0.032802	-0.13233	FM	-0.21705753
321	0.45	0.025	-0.80803	0.063593	-0.12838	collinear Néel AFM	-0.20366216
322	0.45	0.0375	-0.78257	0.092623	-0.12481	collinear Néel AFM	-0.20686217
323	0.45	0.05	-0.7583	0.1201	-0.1216	collinear Néel AFM	-0.21008276
324	0.45	0.075	-0.71278	0.17112	-0.1161	collinear Néel AFM	-0.21648424
325	0.45	0.1	-0.67049	0.21785	-0.11166	collinear Néel AFM	-0.22282644
326	0.45	0.125	-0.63066	0.26123	-0.10812	collinear Néel AFM	-0.22912595

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Index	θ/π	ϕ/π	J_t	J_h	J_d	classified phase	$E_0/N/J_{\text{ternary}}$
327	0.45	0.15	-0.59267	0.30198	-0.10535	collinear Néel AFM	-0.23541717
328	0.45	0.175	-0.556	0.34072	-0.10328	collinear Néel AFM	-0.24174812
329	0.45	0.2	-0.52021	0.37795	-0.10184	collinear Néel AFM	-0.24817981
330	0.45	0.225	-0.48488	0.41413	-0.101	collinear Néel AFM	-0.25478842
331	0.45	0.25	-0.44964	0.44964	-0.10072	collinear Néel AFM	-0.26167049
332	0.45	0.275	-0.41413	0.48488	-0.101	collinear Néel AFM	-0.26895219
333	0.45	0.3	-0.37795	0.52021	-0.10184	collinear Néel AFM	-0.27680501
334	0.45	0.325	-0.34072	0.556	-0.10328	collinear Néel AFM	-0.28546984
335	0.45	0.35	-0.30198	0.59267	-0.10535	collinear Néel AFM	-0.29528656
336	0.45	0.375	-0.26123	0.63066	-0.10812	inconclusive	-0.30670387
337	0.45	0.4	-0.21785	0.67049	-0.11166	inconclusive	-0.32022119
338	0.45	0.425	-0.17112	0.71278	-0.1161	hexagonal singlet	-0.33628644
339	0.45	0.45	-0.1201	0.7583	-0.1216	hexagonal singlet	-0.35529539
340	0.45	0.475	-0.063593	0.80803	-0.12838	hexagonal singlet	-0.37771811
341	0.45	0.5	0	0.86327	-0.13673	inconclusive	-0.40423363
342	0.475	0	-0.92704	0	-0.07296	FM	-0.24088004
343	0.475	0.0125	-0.89441	0.035142	-0.070446	collinear Néel AFM	-0.22409087
344	0.475	0.025	-0.86382	0.067984	-0.068194	collinear Néel AFM	-0.22681317
345	0.475	0.05	-0.80771	0.12793	-0.064361	collinear Néel AFM	-0.23239245
346	0.475	0.075	-0.75699	0.18174	-0.061269	collinear Néel AFM	-0.23807351
347	0.475	0.1	-0.71039	0.23082	-0.058786	collinear Néel AFM	-0.24383014
348	0.475	0.125	-0.66693	0.27625	-0.056813	collinear Néel AFM	-0.24966635
349	0.475	0.15	-0.62584	0.31888	-0.05528	collinear Néel AFM	-0.25560651
350	0.475	0.175	-0.58647	0.35939	-0.054134	collinear Néel AFM	-0.26169095
351	0.475	0.2	-0.5483	0.39836	-0.053339	collinear Néel AFM	-0.2679749
352	0.475	0.225	-0.51083	0.43629	-0.052871	collinear Néel AFM	-0.27453017
353	0.475	0.25	-0.47364	0.47364	-0.052717	collinear Néel AFM	-0.28144959
354	0.475	0.275	-0.43629	0.51083	-0.052871	collinear Néel AFM	-0.28885519
355	0.475	0.3	-0.39836	0.5483	-0.053339	collinear Néel AFM	-0.29691208
356	0.475	0.325	-0.35939	0.58647	-0.054134	collinear Néel AFM	-0.30585101
357	0.475	0.35	-0.31888	0.62584	-0.05528	collinear Néel AFM	-0.31600138
358	0.475	0.375	-0.27625	0.66693	-0.056813	collinear Néel AFM	-0.32782475
359	0.475	0.4	-0.23082	0.71039	-0.058786	inconclusive	-0.34190708
360	0.475	0.425	-0.18174	0.75699	-0.061269	hexagonal singlet	-0.35886992
361	0.475	0.45	-0.12793	0.80771	-0.064361	hexagonal singlet	-0.3792968
362	0.475	0.475	-0.067984	0.86382	-0.068194	hexagonal singlet	-0.40382146
363	0.475	0.5	0	0.92704	-0.07296	hexagonal singlet	-0.43331629
364	0.5	0	-1	0	0	isolated FM J_t -triangles	-0.25
365	0.5	0.025	-0.92704	0.07296	0	collinear Néel AFM	-0.25381199
366	0.5	0.05	-0.86327	0.13673	0	collinear Néel AFM	-0.258107
367	0.5	0.075	-0.8064	0.1936	0	collinear Néel AFM	-0.26275356
368	0.5	0.1	-0.75476	0.24524	0	collinear Néel AFM	-0.26768546
369	0.5	0.125	-0.70711	0.29289	0	collinear Néel AFM	-0.27287761
370	0.5	0.15	-0.66246	0.33754	0	collinear Néel AFM	-0.27833326
371	0.5	0.175	-0.62004	0.37996	0	collinear Néel AFM	-0.28407755
372	0.5	0.2	-0.57919	0.42081	0	collinear Néel AFM	-0.29015512
373	0.5	0.225	-0.53935	0.46065	0	collinear Néel AFM	-0.29663074
374	0.5	0.25	-0.5	0.5	0	collinear Néel AFM	-0.30359287
375	0.5	0.275	-0.46065	0.53935	0	collinear Néel AFM	-0.3111608
376	0.5	0.3	-0.42081	0.57919	0	collinear Néel AFM	-0.31949705
377	0.5	0.325	-0.37996	0.62004	0	collinear Néel AFM	-0.32882804
378	0.5	0.35	-0.33754	0.66246	0	collinear Néel AFM	-0.33947702
379	0.5	0.375	-0.29289	0.70711	0	collinear Néel AFM	-0.35190908
380	0.5	0.4	-0.24524	0.75476	0	inconclusive	-0.36676629
381	0.5	0.425	-0.1936	0.8064	0	inconclusive	-0.38483518
382	0.5	0.45	-0.13673	0.86327	0	hexagonal singlet	-0.40694848
383	0.5	0.475	-0.07296	0.92704	0	hexagonal singlet	-0.43399488
384	0.5	0.5	0	1	0	isolated J_h -hexagons (=HS)	-0.46712927