Heisenberg-Kitaev Models on Hyperhoneycomb and Stripyhoneycomb Lattices: 3D–2D Equivalence of Ordered States and Phase Diagrams

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We discuss magnetically ordered states, arising in Heisenberg-Kitaev and related spin models, on three-dimensional (3D) harmonic honeycomb lattices. For large classes of ordered states, we show that they can be mapped onto two-dimensional (2D) counterparts on the honeycomb lattice, with the classical energetics being identical in the 2D and 3D cases. As an example, we determine the phase diagram of the classical nearest-neighbor Heisenberg-Kitaev model on the hyperhoneycomb lattice in a magnetic field: This displays rich and complex behavior akin to its 2D counterpart, with most phases and phase boundaries coinciding exactly. To make contact with the physics of the hyperhoneycomb iridate \( \beta \)-Li\(_2\)IrO\(_3\), we also include a symmetric off-diagonal \( \Gamma \) interaction, discuss its 3D–2D mapping, and determine the relevant phase diagrams. We also discover phases of the 3D models which evade the mapping, i.e., are of genuine 3D character. Our results pave the way to a systematic understanding of 2D and 3D Kitaev materials.

In studies of quantum magnetism, materials with strong spin-orbit coupling have moved to center stage, as they promise to realize novel phases beyond those known for spin-symmetric Heisenberg models [1–3]. A paradigmatic example for non-trivial effects of spin-anisotropic interactions is Kitaev’s celebrated honeycomb-lattice spin model, being exactly solvable and realizing a \( Z_2 \) spin liquid of Majorana fermions [4]. In the search for materials realizations, compounds with 4\( d \) and 5\( d \) transition-metal ions arranged on layered honeycomb lattices have been proposed [1], such as Na\(_2\)IrO\(_3\), \( \alpha \)-Li\(_2\)IrO\(_3\), and \( \alpha \)-RuCl\(_3\) [5–9]. In these materials, the combined effect of spin-orbit coupling, Coulomb interaction, and exchange geometry generates \( J_{eff} = 1/2 \) moments subject to a combination of exchange interactions, the most important ones being Kitaev, Heisenberg, and symmetric off-diagonal [10–12]. These two-dimensional (2D) Kitaev materials have generated tremendous interest, and the relevant extended Kitaev models have been shown to host both spin-liquid and symmetry-broken phases [13, 14].

Beyond two dimensions, it has been shown that the Kitaev model can also be exactly solved on particular three-dimensional (3D) lattices with three-fold coordination [15, 16], and the materials \( \beta \)-Li\(_2\)IrO\(_3\) [17, 18] and \( \gamma \)-Li\(_2\)IrO\(_3\) [19, 20] were found to realize two of these lattices, the hyperhoneycomb and stripyhoneycomb lattices. In fact, these two are part of an infinite family of 3D lattices whose limiting case is the 2D honeycomb lattice—the so-called honeycomb series [15, 19, 21]. While Na\(_2\)IrO\(_3\) and \( \alpha \)-RuCl\(_3\) display collinear zigzag order [6, 7, 22] at low temperatures, the magnetic states of the Li\(_2\)IrO\(_3\) polytypes involve non-collinear spin spirals. However, similarities in an applied magnetic field have been reported. In \( \beta \)-Li\(_2\)IrO\(_3\), for instance, a small field rapidly suppresses the spiral order and induces a zigzag order analogous to those of the planar honeycomb materials [23, 24]. While a number of concrete studies of either 2D or 3D models have appeared, a common systematic understanding is lacking.

In this Letter, we consider magnetically ordered states on the 3D hyperhoneycomb and other harmonic honeycomb lattices and establish a remarkable correspondence to states on the 2D honeycomb lattice: For large classes of states, we demonstrate a precise mapping between 3D and 2D, with the ground-state and magnon-mode energies being identical in the semiclassical limit. As an example, we determine the phase diagram of the classical nearest-neighbor Heisenberg-Kitaev model on the hyperhoneycomb lattice in a magnetic field: We show that this is almost identical to that of the same model on the honeycomb lattice [25], with the exception of a single field-induced phase that is of genuine 3D character and thus escapes the 3D–2D mapping. In order to model \( \beta \)-Li\(_2\)IrO\(_3\), we include symmetric off-diagonal \( \Gamma \) interactions on the hyperhoneycomb lattice, which can induce incommensurate spiral phases, and we discuss their exact 3D–2D mapping. Finally, we consider effects beyond the classical limit and compare quantum corrections to the magnetization between the 3D and 2D cases.

Mapping from 3D to 2D: General. The hyperhoneycomb lattice is a tricoordinated 3D lattice that can be classified as face-centered orthorhombic with a four-site atomic unit cell [26, 27]. In Fig. 1(a), we show the spin configuration of a so-called skew-zigzag antiferromagnetic state on this lattice, while Fig. 1(b) displays a corresponding zigzag state on the 2D honeycomb lattice. These two states are equivalent in the following sense: Projecting the hyperhoneycomb lattice onto its \( ac \) plane results in an elongated honeycomb lattice, Fig. 1(c), and this projection transforms the skew-zigzag state from panel (a) into the zigzag state of panel (b). Importantly, their classical-limit energies are identical, because in both cases each site faces neighbors with identical alignment. This projective equivalence, which is the central result of this Letter, applies to all 3D ordered states where sites separated by the lattice vector \( b \) [dashed line in Fig. 1(a)] are magnetically equivalent. As we will see below, this includes large classes of 3D magnetic states, which we will dub “quasi-2D”.

This 3D–2D equivalence holds despite the fact that the symmetry groups on the two lattices are obviously different. In particular, the number of sites in the primitive unit cell on the hyperhoneycomb lattice is four, and thus twice its value on the...
HK Hamiltonian [11, 13] in a uniform magnetic field $\mathbf{h}$,

$$H_{HK} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + K \sum_{\langle ij \rangle} \mathbf{S}_i \gamma \mathbf{S}_j - \mathbf{h} \cdot \sum_i \mathbf{S}_i,$$

(1)

where $\gamma \in \{x, y, z\}$ labels the three different types of bonds on the lattice. The couplings are conventionally parameterized as $J = A \cos \varphi$ and $K = 2A \sin \varphi$, where $A > 0$ is an overall energy scale [32]. The 2D model on the honeycomb lattice has been studied intensely, see Refs. 3, 33–35 for reviews. For non-zero field, the classical phase diagram (i.e., for $S \to \infty$) of this and related models has been determined [25, 36–38], and the $S = 1/2$ case has also been studied [39–48]. The 3D model on the hyperhoneycomb lattice has been considered in Refs. 26 and 49.

Here we have determined the phase diagram of the classical HK model in a magnetic field using a combination of high-field spin-wave theory and classical energy minimization. On the hyperhoneycomb lattice, the number of possible geometries of the magnetic unit cell drastically increases with its size. For reasons of numerical feasibility and consistency, we have restricted the numerical energy minimization in both the 2D and 3D cases to states with up to 12 sites in the magnetic unit cell, but have included all possible unit-cell geometries; for details see the SM [28]. Our findings on the honeycomb lattice are consistent with the previous analysis [25, 50].

The result, comparing the hyperhoneycomb and honeycomb cases for a field along the [111] $\propto (\hat{x} + \hat{y} + \hat{z})/\sqrt{3}$ direction, is displayed in Fig. 2. The various phases are characterized in the SM [28]. Remarkably, both phase diagrams agree quantitatively, with the exception of the 3D spiral phase, which appears only in the 3D case of panel (a). Inspecting the individual phases, we see that all phases except the 3D spiral have ordering wavevectors located in the $ac$ plane, such that 3D–2D mapping applies, whereas the 3D spiral phase has $\mathbf{Q} = \frac{2}{3} \mathbf{Y}$, evading the mapping. The latter is hence a genuine 3D phase, with a 12-site magnetic unit cell, and we have illustrated its spin configuration in Fig. 2(c).

We have also determined the phase diagram of the hyperhoneycomb model in a field along the [001] $\propto \hat{z}$ direction, for which all in-field ordered states are simply canted versions of the zero-field orders, without any field-induced intermediate phases, in complete quantitative equivalence with the 2D honeycomb-lattice result [25].

We note that early work on the hyperhoneycomb-HK model in a magnetic field [49] missed the nontrivial field-induced phases found here. We have explicitly checked that our novel intermediate phases have lower energies than the canted skew-zigzag and skew-stripy states suggested in Ref. 49.

$\Gamma$ and other interactions. For actual Kitaev materials, it has been shown that, in addition to nearest-neighbor Kitaev and Heisenberg interactions, also symmetric off-diagonal interactions, commonly dubbed $\Gamma$ interactions, are important [12, 37, 51–55]. To model $\beta$-Li$_2$IrO$_3$, we hence consider the

Heisenberg-Kitaev model in a magnetic field. To illustrate the power of the advertised mapping, we consider the spin-S

honeycomb lattice. It is thus possible to construct states (e.g., skew-zigzag states) that do not break the translation symmetry on the hyperhoneycomb lattice, although their 2D projections break the honeycomb translation symmetry.

Further insight is gained in reciprocal space. All high-symmetry points displayed in Fig. 1(d) have a vanishing component along the direction of the reciprocal lattice vector $\mathbf{b}^*$ (up to reciprocal-lattice translations). States with ordering wavevectors at these high-symmetry points thus exhibit no modulation along the $\mathbf{b}$ axis in real space and are quasi-2D: Their projection onto the $ac$ plane yields states on the honeycomb lattice with the exact same classical energy. This applies to all ordered phases in the nearest-neighbor Heisenberg-Kitaev (HK) model in zero field. Upon the inclusion of other symmetry-allowed interactions, as well as in a magnetic field, Kitaev systems also stabilize multi-$\mathbf{Q}$ and incommensurate states. We will show that even such more exotic states, including the counterrotating spiral states that are realized in the different Li$_2$IrO$_3$ polytypes, are quasi-2D. Moreover, the 3D–2D mapping discussed here for the hyperhoneycomb lattice can be extended to the full harmonic honeycomb series, for details see the supplemental material (SM) [28].

Figure 1. (a) Hyperhoneycomb lattice with magnetic skew-zigzag state and (b) honeycomb lattice with zigzag state; both are equivalent as explained in the text. (c) Hyperhoneycomb lattice viewed from the crystallographic $\hat{b}$ direction, illustrating its projection onto an elongated honeycomb lattice. Colored balls indicate spin directions. (d) Brillouin zones of the hyperhoneycomb lattice (black) and the elongated honeycomb lattice (red dashed). 3D states with ordering wavevectors in the $ac$ plane can be transformed into equivalent 2D states on the honeycomb lattice. The quarters of the front hexagon (such as the green quadrangle) can be shifted with reciprocally lattice vectors to the $ac$ plane and form, together with the $ac$-plane cut of the first Brillouin zone, a rectangle (black dashed). The latter becomes the 2D Brillouin zone of the elongated honeycomb lattice if a four-site unit cell is chosen. All high-symmetry points shown are quasi-2D.
Figure 2. Phase diagram of the classical HK model in a magnetic field $h$ along the $[111]$ direction for $T \to 0$, with $J = A \cos \varphi$, $K = 2A \sin \varphi$ [32], and the radial direction representing the field strength $h$, with $h/(A S) = 1, 2, 3, 4, 5$ from outer to inner gray circles, on the (a) hyperhoneycomb lattice and (b) honeycomb lattice. The latter agrees with the previous analysis [25, 36], with the exception of small regions for which the true ground state has incommensurate ordering wavevectors. Thick (thin) black lines denote first-order (second-order) phase transitions. Except for the 3D spiral phase, for which a representative spin configuration is shown in (c), all phases of the hyperhoneycomb lattice have a 2D analogue with exactly the same ground-state energy. (d) Order parameters in N´eel and zigzag phases from linear spin-wave theory for $S = 1/2$ and zero field, comparing the hyperhoneycomb (solid) and honeycomb (dashed) lattices.

Hamiltonian

$$
\mathcal{H}_{\text{HFK}} = \sum_{\langle ij \rangle} \left[ J S_i \cdot S_j + K S_i^+ S_j^+ \pm \Gamma \left( S_i^x S_j^x + S_i^y S_j^y \right) \right],
$$

where $\alpha$ and $\beta$ label the two remaining directions on a $\gamma$ bond. On the hyperhoneycomb lattice, there are two inequivalent types of pairwise parallel $x$ ($y$) bonds, denoted as $x$ and $x'$ ($y$ and $y'$), respectively, while all $z$ bonds are equivalent; see Fig. 1(a). We take the upper (lower) sign in front of the $\Gamma$ interaction on $x$, $y$, and $z$ ($x'$ and $y'$) bonds; this choice can be justified microscopically [52]. Applying the 3D–2D mapping to this model, which we dub HK±$\Gamma$ model, we see that it corresponds to an unusual 2D model. Compared to the Heisenberg-Kitaev-\(\Gamma\) (HFK) model typically considered for 2D Kitaev materials, this has a supermodulation in the $\Gamma$ interaction. Importantly, in the presence of sizable $\Gamma$ interactions, incommensurate states appear [12, 52].

Despite these complications, the concept of the 3D–2D mapping continues to apply. We illustrate this in Fig. 3, where we show the classical phase diagrams of the HK±$\Gamma$ model at zero field for the hyperhoneycomb and honeycomb lattices. These have been obtained via a combination of a Luttinger-Tisza analysis and a single-$Q$ ansatz (as the finite-cluster minimization does not capture incommensurate states); see the SM for details [28]. Our result on the hyperhoneycomb lattice agrees with the previous work [52], except for a small region around the $\alpha F_{\text{abc}}$ phase, for which the true ground state may be a multi-$Q$ state that is beyond our ansatz. Again, the 3D and 2D phase diagrams agree quantitatively, with the exception of the $\alpha F_{\text{bp}}$ and $\alpha F_{\text{abc}}$ phases, which appear only in panel (a) and are thus genuinely 3D. This result is particularly striking for the counterrotating spiral $\alpha F_{\text{a}}$ phases, for which the ground state is incommensurate, but with an ordering wavevector $Q || a^\prime$, i.e., within the $ac$ plane; cf. Fig. 3(c,d).

This phase includes the ground state realized in $\beta$-Li$_2$IrO$_3$ [17, 52–55], such that 3D–2D mapping directly applies to this material.

For completeness, we note that the 3D–2D mapping can be extended to interactions beyond nearest neighbors. For instance, second-neighbor interactions on the hyperhoneycomb lattice are mapped to a combination of second- and fourth-neighbor interactions on the honeycomb lattice, third-neighbor interactions map to third- and sixth-neighbor interactions; for details see the SM [28].

Beyond the classical limit. While the advertised qualitative 3D–2D mapping of ordered states is very general, their quantitative energetic equivalence only applies to the classical limit, $S \to \infty$. We have therefore studied quantum effects in a systematic $1/S$ expansion using spin-wave theory. A first remarkable insight is that the leading-order magnon spectra also follow the 3D–2D mapping, i.e., the magnon energies in both cases are identical for 3D wavevectors belonging to the $ac$ plane; this is demonstrated explicitly in the SM [28].

In Fig. 2(d), we display the order parameters (i.e., staggered magnetizations) evaluated for $S = 1/2$ in the N´eel and zigzag phases of the HK model at zero field, comparing the hyperhoneycomb and honeycomb lattices. In the hyperhoneycomb case, the quantum corrections to the classical value $M_{\text{stag}} = 1/2$ are smaller, but the overall shape of the order parameter as function of $\varphi = \text{arg}(2J + iK)$ is similar to those of the honeycomb lattice. In particular, in linear spin-wave theory, the critical values of $\varphi$ at which the order parameters van-
Figure 3. Phase diagram of the classical HK±Γ model for $T \to 0$, with $J = A \sin \theta \cos \varphi$, $K = A \sin \theta \sin \varphi$, and $\Gamma = A \cos \theta \leq 0$ [32] on the (a) hyperhoneycomb lattice [52] and (b) honeycomb lattice. Hatched regions in (a) denote genuine 3D phases with ordering wavevectors outside the $ac$ plane. The white dashed line indicates the regions in which the Luttinger-Tisza approach fails to satisfy the local length constraint and a single-$Q$ ansatz has been employed instead; see SM [28]. The 3D–2D equivalence holds also for the incommensurate spiral phase $\tilde{S}P_x$, relevant for $\beta$-Li$_2$IrO$_3$, for which a representative spin configuration on the hyperhoneycomb lattice is plotted in (c), together with its projection onto the honeycomb lattice in (d).

ish, indicating the transition to the Kitaev spin liquid phase, roughly agree. This suggests that the Kitaev spin liquid on the hyperhoneycomb lattice [56] covers a parameter range that is only slightly smaller than those of its honeycomb-lattice counterpart. In the SM [28], we also show the quantum corrections to the uniform magnetization in the asymptotic high-field phase.

These results illustrate that the different phase space renders quantum fluctuations stronger in 2D compared to 3D. As a result, phase boundaries will shift and spoil the exact 3D–2D equivalence for $S < \infty$. This also means that classical phases that are destroyed by quantum fluctuations in 2D possibly survive in the 3D case.

Summary. In this paper, we have established an exact correspondence between magnetically ordered spin states on the 3D harmonic honeycomb lattices and the 2D planar honeycomb lattice. This correspondence is quantitative in the classical limit and applies to large classes of ordered states. The condition is that the respective 3D ordering wavevector(s) lie(s) in the $ac$ plane (up to reciprocal-lattice translations), which pertains to all high-symmetry points in the Brillouin zone. We have demonstrated this 3D–2D mapping for the hyperhoneycomb-lattice Heisenberg-Kitaev model in a magnetic field, where we found exact agreement with the 2D case, with the exception of one intermediate phase which is of genuine 3D character.

The hyperhoneycomb material $\beta$-Li$_2$IrO$_3$ orders in an incommensurate spiral ground state at low temperatures [17, 18]. For this state, our 3D–2D mapping also applies. On the classical level, the physics of the incommensurate spiral state can thus be fully understood within a suitable 2D model. We have demonstrated this explicitly within a HK±Γ model; it should be emphasized that the quasi-2D nature of the experimentally observed state is independent of the choice of microscopic model. Our result establishes the equivalence of the experimentally observed spiral states in $\alpha$-, $\beta$-, and $\gamma$-Li$_2$IrO$_3$: Under the 3D–2D mapping, these states can be adiabatically transformed into each other. $\beta$-Li$_2$IrO$_3$ exhibits a nontrivial behavior in a finite magnetic field [23, 24, 54]. The 3D–2D equivalence suggests that similarly interesting in-field effects may occur also in $\alpha$- and $\gamma$-Li$_2$IrO$_3$ [57, 58]. This represents an excellent direction for future theoretical and experimental work. Together, our work paves the way to a unified understanding of the magnetism in 3D and 2D Kitaev materials and opens novel perspectives for dimensional diversification.

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[28] See Supplemental Material, which also contains Refs. 29–31, for a generalization of the 3D–2D mapping on the harmonic honeycomb lattice series including interactions beyond the HKΓ models, details of the different methods used, characterizations of the various phases found, and a discussion of quantum corrections in linear spin-wave theory.
Supplemental Material for:
Heisenberg-Kitaev Models on Hyperhoneycomb and Stripyhoneycomb Lattices:
3D–2D Equivalence of Ordered States and Phase Diagrams

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I. 3D–2D MAPPING: GENERALIZATIONS

In the main paper, the mapping of magnetically ordered states from 3D honeycomb-lattice variants to the planar honeycomb lattice has been explicitly shown for the hyperhoneycomb lattice; the mapping of Hamiltonian terms was restricted to nearest-neighbor interactions. Here we show that the mapping applies in fact more generally.

A. Harmonic honeycomb lattice series

A family of so-called harmonic honeycomb lattices has been introduced in Refs. 1 and 2. They consist of an arrangement of $N$ complete planar honeycomb rows along the $c$ axis, followed by a row of $c$-axis bonds which rotate between honeycomb planes, see Fig. S1. For $N = 0$ this yields the planar honeycomb lattice, while any finite $N$ corresponds to a 3D lattice. The case $N = 0$ is commonly called hyperhoneycomb; the corresponding lattice basis vectors can be written as $\mathbf{a}_1 = (2, 4, 0)$, $\mathbf{a}_2 = (3, 3, 2)$, $\mathbf{a}_3 = (-1, 1, 2)$, yielding the reciprocal basis vectors $\mathbf{b}_1 = (\pi/3, -2\pi/3, \pi/2)$, $\mathbf{b}_2 = (-2\pi/3, \pi/3, -\pi/2)$, $\mathbf{b}_3 = (2\pi/3, -\pi/3, -\pi/2)$. The case $N = 1$ is dubbed stripyhoneycomb, with basis vectors $\mathbf{a}_1 = (6, 6, 0)$, $\mathbf{a}_2 = (-2, 2, 0)$, $\mathbf{a}_3 = (-1, 1, 2)$. Parenthetically, we remark that Ref. 2 also discussed more general structures with more than one row of rotated bonds interspersed between planar bonds; these will not be considered here.

The 3D lattices displayed in Fig. S1 are orthorhombic. In all cases, projecting the structure onto the $ac$ plane yields a (distorted) honeycomb lattice; this operation projects sites separated by (multiples of) the lattice vector $\mathbf{b}$ onto a single site. Consequently, the mapping of ordered states advertised in the main paper applies unchanged. In momentum space, the family of mappable (i.e., quasi-2D) states is characterized by ordering wavevectors with vanishing component along $\mathbf{b}$.

B. Symmetries and supermodulations

The 3D–2D mapping of states is paralleled by a mapping of the underlying Hamiltonians, as shown in the main paper. Here symmetry considerations are crucial.

The 3D lattices of the harmonic honeycomb series have a crystallographic unit cell of $4 + 4N$ sites, to be contrasted with the two-site unit cell of the 2D honeycomb lattices. Moreover, the point-group symmetry of the (projected) 3D lattice is lower than that of the honeycomb lattice. For instance, the three types of bonds are no longer related by simple $Z_3$ rotations. Together, this implies a larger number of symmetry-allowed distinct interaction terms in 3D as compared to 2D.

The 3D–2D mapping thus connects, e.g., ordered states of the hyperhoneycomb lattice, the corresponding lattice basis vectors can be written as $\mathbf{a}_1 = (2, 4, 0)$, $\mathbf{a}_2 = (3, 3, 2)$, $\mathbf{a}_3 = (-1, 1, 2)$, yielding the reciprocal basis vectors $\mathbf{b}_1 = (\pi/3, -2\pi/3, \pi/2)$, $\mathbf{b}_2 = (-2\pi/3, \pi/3, -\pi/2)$, $\mathbf{b}_3 = (2\pi/3, -\pi/3, -\pi/2)$. The case $N = 1$ is dubbed stripyhoneycomb, with basis vectors $\mathbf{a}_1 = (6, 6, 0)$, $\mathbf{a}_2 = (-2, 2, 0)$, $\mathbf{a}_3 = (-1, 1, 2)$. Parenthetically, we remark that Ref. 2 also discussed more general structures with more than one row of rotated bonds interspersed between planar bonds; these will not be considered here.

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C. Interactions beyond nearest neighbors

The 3D–2D mapping of Hamiltonian terms can be extended beyond the simplest nearest-neighbor exchange interactions. However, the lower symmetry of the 3D lattices complicates the discussion of further-neighbor interactions: pairs of sites with the same Euclidean distance may be symmetry-inequivalent.

For instance, on the hyperhoneycomb lattice, there are two types of “second-neighbor bonds”: one where the two sites are connected by two first-neighbor bonds, and one where this connection is missing due to the twisted row of bonds. Upon projecting to the $ac$ plane, the former become second-neighbor bonds on the 2D honeycomb lattice, while the lat-
Due to the different nature of the expected phases we have used different methods in the two cases.

Figure S1. Harmonic honeycomb lattice series $\mathcal{H}(N)$, where $N$ refers to the number of complete honeycomb rows along the $c$ axis. (a) $N = 0$ (hyperhoneycomb), (b) $N = 1$ (stripyhoneycomb), (c) $N \to \infty$ (honeycomb), (d-f) building blocks for general $N$. $x, y,$ and $z$ bonds are marked in red, green, and blue. The labels “±” refer to the signs of the $\Gamma$ interaction on the different bonds.

Figure S2. Signs of the $\Gamma$ interaction on the different bonds in the projected hyperhoneycomb-lattice (a) and stripyhoneycomb-lattice (b) models. $x, y,$ and $z$ bonds are marked in red, green, and blue. Dashed rectangles indicate the four-site (a) and eight-site (b) primitive unit cells, respectively. The black and orange sites in (a) refer to the unitary transformation that removes the sign structure as explained in the text.

Figure S3. Projection of nearest-, second-nearest-, and third-nearest-neighbor interactions of an arbitrary central site on the hyperhoneycomb lattice. Under the projection, nearest neighbors remain nearest neighbors, while second-nearest (third-nearest) neighbors map to second- and fourth-nearest (third- and sixth-) nearest neighbors on the honeycomb lattice. “×2” indicates cases in which two hyperhoneycomb sites map to the same honeycomb sites.

is specific to each particular compound and requires ab-initio consideration that are beyond the scope of this paper.

II. CLASSICAL PHASES AND PHASE DIAGRAMS

Here we describe the methods used to determine the classical phase diagrams shown in Figs. 2 and 3 of the main paper. Due to the different nature of the expected phases we have used different methods in the two cases.
For the HK model in an applied field, a large fraction of the phase diagram is covered by commensurate phases, a number of them showing multiple modulation wavevectors (multi-\(Q\) states).\(^4,5\) Hence, it is efficient to assume a finite magnetic unit cell, parameterize all spins in this unit cell by two angles each, and minimize the classical energy as function of these angles. In both 2D and 3D, we have included all possible magnetic unit cells with up to 12 sites. In 2D and 3D, this leads to 8 and 21 different unit-cell geometries, respectively (plus symmetry-equivalent ones). Among the phases known for the 2D HK model in a field, this approach captures all phases apart from the “zigzag-star” and “diluted star” phases of Ref. 4, which occur at elevated [111] fields above the zigzag and stripy phases, respectively. In Ref. 4, these phases were found to have a 36-site (zigzag star) and 18-site (diluted star) unit cell, but it is likely that their true structures are incommensurate.\(^5\)

In the phase diagram of the HK\(\pm \Gamma\) model, a prominent role is taken by incommensurate spiral phases.\(^3\) Hence, an energetic minimization assuming a finite unit cell is inefficient. Instead, we use a combination of the Luttinger-Tisza approach\(^6,7\) and a single-\(Q\) ansatz. Here, the Luttinger-Tisza approach captures both commensurate and incommensurate magnetic states, but cannot discriminate between single-\(Q\) and multi-\(Q\) cases. Moreover, it is an approximation for situations with crystallographic unit cells larger than one site. Single-\(Q\) spiral-type states are most efficiently captured with the ansatz

\[
S_a(r) = \left[\hat{e}_x^a \cos(Q \cdot r) + \hat{e}_y^a \sin(Q \cdot r)\right] \sin \eta_a + \hat{e}_z^a \cos \eta_a, \tag{S1}
\]

where \(a = 1, \ldots, 4\) labels the sublattice and \(r\) the position of the spin. The ordering wavevector \(Q\), the basis vectors \(\{\hat{e}_x^a, \hat{e}_y^a, \hat{e}_z^a\}\) and the canting parameters \(\eta_a\) are variational parameters.

For the HK\(\pm \Gamma\) model, both approaches delivered identical results in most of the phase diagram, with the exception of a region (marked by white lines in Fig. 3 of the main paper) where the Luttinger-Tisza solution violated the spin-length constraint. Consequently, we have used the results of the single-\(Q\) ansatz in these cases.

The various phases that have been obtained in the HK model in a [111] magnetic field and in the HK\(\Gamma\) model at zero field are characterized in Table I.

### III. MAGNON SPECTRA AND QUANTUM CORRECTIONS

Here we discuss corrections to the classical picture, which formally corresponds to the limit of infinite spin magnitude, \(S \to \infty\). At \(T = 0\), quantum corrections from \(S < \infty\) are different in 3D and 2D and will hence invalidate the exact 3D–2D mapping. Nevertheless, the mapping continues to apply on a qualitative level for most regions of the phase diagrams.

Spin-wave theory is the canonical method to determine excitation spectra and quantum corrections for magnetically ordered states in spin models. Spin-wave theory can be organized in a systematic expansion in \(1/S\). Here we determine the leading-order spin-wave spectra which then can be used to calculate thermodynamic observables to next-to-leading order; most importantly the order parameter and the uniform magnetization.

We employ the Holstein-Primakoff representation with distinct boson operators on the four (two) sublattices on the hyperhoneycomb (honeycomb) lattice. The resulting Bogoliubov problem is diagonalized numerically. As the methodology is standard, we refer the reader to the review in Ref. 8 for details.

#### A. 3D–2D mapping of spectra

Inspecting the leading-order spin-wave spectra reveals a remarkable fact: For 3D momenta that are "quasi-2D", i.e., that have a vanishing component along \(\mathbf{b}^*\), the mode energies in the 3D case are exactly equal to the mode energies of the corresponding 2D model at the respective projected momenta. This is illustrated for the HK model in Fig. S4 for both the zero-field zigzag phase and the high-field phase; to facilitate the comparison, a four-site unit cell has been chosen in the 2D case. At zero field, pseudo-Goldstone modes are seen at \(Y\) and \(Y'\) wavevectors, which are shifted from the \(z\) skew-zigzag ordering wavevector \(Q = \Gamma\). In full agreement with the situation on the honeycomb lattice.\(^9\) In the high-field phase, all

| Phase        | \(Q\) | Comments |
|--------------|------|---------|
| skew-zigzag (SZ) | \(\Gamma\) or \(Y\) | \(S_i\parallel S_j\parallel \hat{y}\) (degenerate) |
| AF star      | \(\Gamma\) and \(Y\) | multi-Q |
| AF vortex    | \(E\) |         |
| 3D spiral    | (0, \(\frac{1}{2}\), 0) | true-3D |
| SZ\(_{a/b}\) | \(Y\) | zigzag in \(S_i\cdot \hat{x}\) or \(S_i\cdot \hat{y}\) (degenerate) |
| SZ\(_{b}\)  | \(\Gamma\) | zigzag in \(S_i\cdot \hat{b}\) |
| skew-stripy (SS) | \(\Gamma\) or \(Y\) | \(S_i\parallel S_j\parallel \hat{y}\) (degenerate) |
| FM star      | \(\Gamma\) and \(Y\) | multi-Q |
| vortex       | \(E\) |         |
| SS\(_{a/b}\) | \(Y\) | stripy in \(S_i\cdot \hat{x}\) or \(S_i\cdot \hat{y}\) (degenerate) |
| Néel         | \(\Gamma\) |         |
| AF\(_{a}\)   | \(\Gamma\) | Néel in \(S_i\cdot \hat{a}\), stripy in \(S_i\cdot \hat{b}\) |
| AF\(_{a/b}\) | \((\frac{1}{2}, \frac{1}{2}, \frac{1}{2})\) | true-3D |
| ferromagnet (FM) | \(\Gamma\) |         |
| polarized    | \(\Gamma\) | \(S_i\parallel \hat{h}\) |
| FM\(_{a}\)   | \(\Gamma\) | \(S_i\parallel \hat{e}\) |
| FM-SZ\(_{a}\) | \(\Gamma\) | FM in \(S_i\cdot \hat{b}\), zigzag in \(S_i\cdot \hat{a}\) |
| \(\mathbf{SP}_{a}\) | \((h, 0, 0)\) | incommensurate spiral, quasi-2D |
| \(\mathbf{SP}_{b}\) | \((0, k, 0)\) | incommensurate spiral, true-3D |
modes are gapped.

The 3D–2D mapping of the magnon spectra can be understood as follows: Given the equivalence of the lattice structures (and interactions) after projection on the ac plane, the Fourier-transformed interactions appearing in the magnon Hamiltonian are equivalent after projection as well, i.e., they are equal for quasi-2D momenta. It is these (bare) interactions that determine the magnon spectra at leading order in 1/S, hence the 3D–2D mapping is generically valid.

The equivalence breaks down at higher orders in 1/S because quantum corrections arising from magnon-magnon interactions involve momentum integrals running over the full Brillouin zone in either 3D or 2D and hence are different in both cases. The same applies to quantum corrections to thermodynamic quantities, as shown below.

B. Quantum corrections to magnetization

The spin-wave results can be used to determine zero-temperature quantum corrections to local expectation values. We employ this to calculate the magnetization in the high-field phase of the HK model. Here, the magnetization points in field direction and is saturated in the classical limit. Using the Holstein-Primakoff representation, the magnetization \( M \)

![Figure S5. Magnetization in the field-polarized phase for \( \varphi = 0.62\pi \) of the HK model as a function of the magnetic field applied in [111] direction, obtained from linear spin-wave theory for \( S = 1/2 \), comparing the hyperhoneycomb (solid) and honeycomb (dashed) lattices.](image)

along the field direction including quantum corrections is

\[
M = S - \frac{1}{N} \sum_q \left( a_q^\dagger a_q + b_q^\dagger b_q + c_q^\dagger c_q + d_q^\dagger d_q \right) \quad (S2)
\]

where \( a_q, b_q, c_q, \) and \( d_q \) are the magnon operators on the four sublattices, and \( N \) is the number of sites. In next-to-leading order in 1/S, this can be expressed in terms of the Bogoliubov
coefficients obtained from linear spin-wave theory, for details see Refs. 4 and 8. The numerical result, evaluated for $S = 1/2$, is shown in Fig. S5 for a particular value of $\varphi$, parameterizing the ratio between Heisenberg and Kitaev couplings according to $J = A \cos \varphi$ and $K = 2A \sin \varphi$. The comparison between the 2D honeycomb and 3D hyperhoneycomb results confirms that quantum effects are stronger in lower dimensions, as a result of the larger fraction of low-energy phase space.

We have also calculated the quantum corrections to the order parameter of selected zero-field phases: For collinear states this computation involves only amplitude corrections and is similar to that leading to the uniform magnetization in Eq. (S2); for non-collinear states angle corrections would need to be considered as well. Numerical results, again evaluated for $S = 1/2$, are shown for the Néel and zigzag phases of the HK model in Fig. 2(d) of the main paper. Also here, quantum effects are stronger in lower dimensions.

These results illustrate that quantum corrections invalidate the 3D–2D mapping for $S < \infty$. However, thermodynamic observables, such as magnetization curves, continue to agree in 3D and 2D on a qualitative level in large regions of the phase space, in particular deep inside magnetically ordered and classical paramagnetic phases.

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