The Heisenberg-Kitaev (HK) model on the triangular lattice is conceptually interesting for its interplay of geometric and exchange frustration. HK models are also thought to capture the essential physics of the spin-orbital entanglement in effective $j = 1/2$ Mott insulators studied in the context of various 5d transition metal oxides.

Here we argue that the recently synthesized Ba$_3$IrTi$_2$O$_9$ is a prime candidate for a microscopic realization of the triangular HK model. We establish that an infinitesimal Kitaev exchange destabilizes the 120° order of the quantum Heisenberg model and results in the formation of an extended $Z_2$-vortex crystal phase in the parameter regime most likely relevant to the real material. Using a combination of analytical and numerical techniques we map out the entire phase diagram of the model, which further includes various ordered phases as well as an extended nematic phase around the antiferromagnetic Kitaev point.
separate exchange paths as indicated in Fig. 1 b) leading to a destructive interference and subsequent suppression of the isotropic Heisenberg exchange. In comparison to the tricoordinated Iridates (Na,Li)_2IrO_3, which exhibit Ir-O-Ir exchange paths, the triangular Ba_2IrTi_2O_9 exhibits somewhat longer Ir-O-O-Ir exchange paths resulting in an overall lessening of the magnetic exchange strength. Second, the three principal bond directions of the triangular lattice structure cut through three different edges of the IrO_6 oxygen cages resulting in a distinct locking of the exchange easy axis along the three directions as illustrated Fig. 1 a) and ultimately giving rise to the three components of the Kitaev exchange. The description of the microscopic physics is thus given in terms of a Hamiltonian

\[
\mathcal{H}_{HK} = J_H \sum_{\langle i \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_K \sum_{\gamma \parallel \langle i \rangle} S^\gamma_i S^\gamma_j,
\]

where \(\mathbf{S}_i\) is a spin-operator located on site \(i\) of the triangular lattice spanned by the lattice vectors \(a_x = (1,0)^T\), \(a_y = (-1/2, \sqrt{3}/2)^T\), and \(a_z = -a_x - a_y\), see Fig. 2 a). Here and in the following, we measure lengths in units of the lattice constant \(a\). The first term is the standard Heisenberg coupling, \(J_H\), that describes an SU(2) invariant interaction between the spin-orbit entangled \(j = 1/2\) moments on nearest-neighbor lattice sites. The Kitaev interaction, \(J_K\), on the other hand, explicitly breaks spin-rotation invariance and acts only between single components, \(S^\gamma\), of adjacent spins. The precise component depends on the link between the lattice sites, see Fig. 2 a); for our particular choice here, the \(\gamma\)-component of spins interact via \(J_K\) if sites are connected by a lattice vector \(a_\gamma\), with \(\gamma = x, y, z\).

120° order and \(\mathbb{Z}_2\) defects.—The ground state of Hamiltonian (5) for antiferromagnetic \(J_H > 0\) and \(J_K = 0\) is characterised by 120° ordering of spins. At the classical level it is captured by a spin orientation \(\mathbf{S}_i = S\hat{\Omega}(r_i)\) with the unit vector \(\hat{\Omega}(r) = e_1 \cos (Q \cdot r) + e_2 \sin (Q \cdot r)\) where the consensual wavevector \(Q\) connects the center with a corner of the Brillouin zone, \(Q = \frac{\sqrt{3}}{2}(1,0)\). The orthonormal frame \(e_i\) with \(i = 1, 2, 3\) and \(e_3 = e_1 \times e_2\) constitutes an SO(3) order parameter for this phase. Crucially, the 120° ordering possesses \(\mathbb{Z}_2\) vortices as topologically stable point defects, which can be understood by considering the first homotopy group of its order parameter \(\Pi_1(\text{SO}(3)) = \mathbb{Z}_2\).

Instabilities of the 120° order.—Small fluctuations of the spin vector, \(\delta\hat{\Omega}(r) = \pi_1(r)[-e_1 \sin (Q \cdot r) + e_2 (\cos Q \cdot r)] + \pi_2(r)e_3\), can be parameterized by two real fields \(\pi_1(r)\) with \(a = 1, 2\), which become soft at momentum \(k = 0\) and \(k = \pm Q\), respectively. This identifies three Goldstone modes of the 120° Néel order with Fourier components \(\pi_1(k = 0)\) and \(\pi_2(\pm Q)\) describing long-wavelength rotation and tilting of the local orthonormal frame, respectively. Interestingly, it turns out that an infinitesimal Kitaev interaction \(J_K\) immediately results in negative eigenvalues for the \(\pi_2\) Goldstone modes implying a local tilting instability towards a different ground state, see the Supplementary Material for details. Indeed, allowing for a slowly spatially varying orthogonal frame \(e_i(r)\) one finds in the limit \(|J_K| \ll J_H\) the effective energy functional \(\mathcal{E} = \int d^2 r \mathcal{L}\) with

\[
\mathcal{L} = \frac{3J_H S^2}{4} \sum_{\gamma = x,y,z} e_\gamma^\gamma(r) \left[ - \nabla^2 - 2i \Lambda_K e_\gamma \cdot \nabla \right] e_\gamma^\gamma(r),
\]

where \(e^\pm = (e_1 \pm ie_2)/\sqrt{2}\). The Kitaev interaction induces a coupling \(\Lambda_K = 2 J_K/\sqrt{3} J_H\) to constant gauge fields given by the triangular lattice vectors \(a_\gamma\), that can be identified as Lifshitz invariants as previously mentioned in Ref. The magnetization can thus minimize its energy by allowing for a spatial modulation of the SO(3) order parameter on large length scales \(\propto 1/\Lambda_K \propto J_H/J_K\).

\(\mathbb{Z}_2\) vortex crystal.—The character of this modulated classical ground state can be obtained by minimizing the Hamiltonian treating the orthonormal constraint, \(e_1 \cdot e_j = \delta_{ij}\), or equivalently, \(\hat{\Omega}^2 = 1\), within an improved Luttinger-Tisza approximation. In the limit of large distances \(\Lambda_K |r| \gg 1\), we can restrict ourselves to the smallest momenta only and

Figure 2. (Color online) a) The triangular lattice with the three lattice vectors \(a_i\). Solid, dashed and dotted bonds carry the three distinct Kitaev interactions, see text. b) First Brillouin zone of the triangular lattice. The position and size of the coloured dots indicate the position and weight of Bragg peaks, respectively, expected in the static spin structure factor for the \(\mathbb{Z}_2\) vortex crystal. Each color corresponds to a different spin-component as listed in panel a).

Figure 3. (Color online) \(\mathbb{Z}_2\) vortex crystal stabilized for \(J_H > 0\) in the presence of a small but finite Kitaev interaction \(J_K\) revealed by the chirality vectors of Eq. (4) which were computed from the classical ground state (3). The colour code shows the length of the chirality vector, \(|\kappa(r)|\), normalized to one, that becomes minimal at the \(\mathbb{Z}_2\) vortex cores. The arrows in the close-up of the left panel correspond to projections of \(\kappa(r)\) onto the \(x-y\) plane.
obtain for the classical spin-configuration approximately \(^2\)
\[
S^\gamma(r) \approx \frac{4S}{3\sqrt{3}} \text{Re} \left\{ e^{i\phi} \times \right. \\
\left. \left( e^{i(Q-ta_\gamma)(r-r_0)} + \frac{1}{4} \sum_{\eta \neq \gamma} e^{i(Q-t(2a_\eta-a_\gamma))(r-r_0)} \right) \right\},
\]

where \(S^\gamma\) is the \(\gamma\)-component of the spin. The first term in Eq. (3) is the most important, primary Fourier component which also possesses the smallest deviation of momentum from the corner of the Brillouin zone, \(Q\); it is shifted by \(-ta_\gamma\) with \(t \propto \Lambda_K\) for \(\Lambda_K \ll 1\), see the Supplementary Material \(^2\) for details. The secondary Fourier components have a smaller weight and are shifted further away by \(-t(2a_\eta - a_\gamma)\) with \(\eta \neq \gamma\). The resulting Bragg peaks in the static structure factor are visualized in Fig. 2 b), which nicely agrees with previous numerical findings for the classical model \(^19\). The relative weight of secondary and primary Bragg peaks are predicted to be \(1/4^2 = 1/16\) within the above approximation. We find that the corresponding energy agrees well with our results from classical Monte Carlo simulations \(^2\), and, in particular, is independent of the choice of origin \(r_0 = (x_0,y_0)^T\) as well as the phase \(\phi\).

It turns out that the approximate classical ground state (3) corresponds to a triangular lattice of condensed \(\mathbb{Z}_2\) vortices, thus confirming the numerical results of Ref. 19. This is best seen by defining chirality vectors on upward pointing triangles of the lattice
\[
\kappa(r) = \frac{2}{3\sqrt{3}} \left( S_r \times S_{r+a_\gamma} + S_{r+a_\gamma} \times S_{r+a_\gamma} + S_{r+a_\gamma} \times S_{r+a_\gamma} \right).
\]

The length of \(\kappa(r)\) measures the rigidity of the local \(120^\circ\) ordering and it becomes minimal at the center of each \(\mathbb{Z}_2\) vortex \(^2\). The chirality vector profile, that derives from Eq. (3), is shown in Fig. 3 and clearly reveals the \(\mathbb{Z}_2\) vortex crystal.

Within our Luttinger-Tisza approximation we find three zero modes for the \(\mathbb{Z}_2\) vortex crystal represented by the phase \(\phi\) and the vector \(r_0\). The latter are expected as the vortex crystal spontaneously breaks translational symmetry so that a constant shift of the origin \(r_0\) does not cost any energy. The corresponding low-energy excitations are just the effective acoustic phonon excitations of the vortex crystal. If the coupling between the two-dimensional atomic triangular lattice planes of \(\text{Ba}_3\text{IrTi}_2\text{O}_8\) is sufficiently small, these low-energy modes will destroy true long-range order of the \(\mathbb{Z}_2\) vortex crystal at any finite temperature reflected in a characteristic broadening of the Bragg peaks in the structure factor of Fig. 2 b).

**Polarized neutron scattering.** – A characteristic of the spin-structure factor of the \(\mathbb{Z}_2\) vortex crystal are the correlations between the position of magnetic Bragg peaks and the corresponding spin-components, which are directly related to the form of the Kitaev interaction that explicitly breaks the \(\text{SU}(2)\) spin-rotation symmetry. This correlation might be resolved with the help of neutron polarimetry. For well-resolved primary Bragg peaks at \(Q = ta_\gamma\), one could probe their origin by choosing a longitudinal orientation \(\hat{e}\) of the polariser and analyser that is however transversal to the transferred momentum \(\hat{q}\), i.e., \(\hat{q} \cdot \hat{e} = 0\). Systematically varying \(\hat{e}\) in the orthogonal plane then, in principle, allows to relate the observed polarisation of the scattered neutrons due to magnetic Bragg scattering to the corresponding spin component, see \(^2\) for details.

**Phase diagram.** – We finally turn to a discussion of the entire phase diagram of the triangular HK model with our results summarized in Fig. 4, where we parametrized the interactions as \(J_{HJ}, J_{HK} = (\cos \alpha, \sin \alpha)\). The nature of most of the phases in the phase diagram can be readily deduced from the fact that the HK model – independent of the underlying lattice – exhibits a duality \(^24\) (also referred to as the Klein duality \(^13\)) relating a pair of interactions on the right-hand side of the circle to a pair of interactions on the left-hand side, i.e., \(J_H \to -J_H\) and \(J_K \to 2J_H + J_K\). In particular, this maps the ferromagnetic phase around the \(J_H < 0\) Heisenberg limit to a dual ferromagnetic phase for \(J_H > 0\) and \(J_K < 0\). The four-sublattice basis transformation relating the two states under this duality, see also Fig. 2 a), creates a state with alternating strips of up and down pointing spins, see the Supplementary Material for an illustration. Similarly, the \(120^\circ\) ordered state and its surrounding \(\mathbb{Z}_2\)-vortex crystal around the \(J_H > 0\) Heisenberg antiferromagnet map to a dual phase in the upper left quadrant with \(J_H < 0\) and \(J_K > 0\) with the respective orderings again illustrated in the Supplementary Material. In addition to these four ordered phases, there is an extended phase around the antiferromagnetic \(J_K > 0\) Kitaev limit, which can be characterized by an emergent nematic order. This can be seen by first considering the classical ground state in the Kitaev limit \((J_K > 0\) and \(J_H = 0\)), which spontaneously breaks the combined symmetry of the HK Hamiltonian of a \(C_n\) lattice rotation and a cyclic spin exchange and as such is referred to as nematic. Its magnetic order is characterized by the formation of independent, antiferromagnetically ordered Ising chains along a favored lattice direction resulting in a \(3 \times 2^L\)-fold sub-extensive degeneracy \(^19\). Considering the effects of quantum fluctuations on these classical states, our numerical analysis on small clusters using exact diagonalization and density matrix renormal-
Figure 5. (Color online) Histogram of the magnetization in O(3) spin space obtained for the O(3) ferromagnet for pure Heisenberg interactions and the magnetization of the Z/2 (stripy) ferromagnet pinned by an order-by-disorder mechanism. Shown are results for finite-temperature Monte Carlo simulations illustrating the effect of thermal fluctuations above the $T = 0$ state in the classical HK model.

The phase boundaries in Fig. 4 have been determined by calculating the ground state energy for clusters with $N = 6 \times 4 = 24$ lattice sites and periodic boundary conditions as well as clusters with 27 lattice sites keeping the original $C_3$ lattice symmetry – with both clusters preserving the SU(2) symmetry of the Heisenberg points under the Klein duality. Using exact diagonalization techniques we have determined the phase boundaries by identifying the points where the second derivative $-d^2E/\sigma^2$ appears to diverge (on these finite systems); see again the Supplementary Material for details. It should be noted that the overall form of this phase diagram for the quantum HK model closely mimics the one found for the classical HK model, which is due to the mainly classical nature of the various ordered phases.

Finally, we comment on the subtle effect of Kitaev interactions on the O(3) ferromagnetic order of the Heisenberg model. While they do not destroy the ferromagnetic order, a quantum order-by-disorder mechanism reduces the O(3) symmetry of the order parameter to a Z/2 symmetry with the order parameter pointing along one of the six equivalent (100) directions as illustrated in Fig. 5. A similar order-by-disorder mechanism has recently been discussed with regard to distortions in the hexagonal HK model.

**Conclusion.**—To summarize, we propose that a Z/2 vortex crystal phase might be observed in the recently synthesized $\text{Ba}_3\text{IrTi}_2\text{O}_9$. The latter forms a $j = 1/2$ Mott insulator, whose low-energy physics we argue to be captured by a triangular lattice Heisenberg-Kitaev model. We reemphasize that the Z/2 vortex crystal arises in the vicinity of the antiferromagnetic Heisenberg model, i.e. in the limit of small Kitaev interactions, and thus in the experimentally most relevant parameter regime – as revealed by numerous microscopic studies of the honeycomb Iridates indicating the presence of Kitaev-type interactions only in addition to a dominant Heisenberg exchange. Initial samples of $\text{Ba}_3\text{IrTi}_2\text{O}_9$ appear to suffer from significant Ir-Ti site inversion obscuring the formation of any ordered phase, but better samples should exhibit a distinct signature in polarized neutron scattering as we have discussed in detail. The physics of the triangular HK model is also relevant to the honeycomb Iridates, for which it has been argued that a next-nearest neighbor exchange (along the two triangular sublattices of the honeycomb lattice) is indeed present in the actual materials. Finally, we have left it to future research to explore whether the Z/2 vortex crystal also plays out in the bilayer triangular lattice material $\text{Ba}_3\text{Ti}_2\text{O}_9$, which is closely related to the $\text{Ba}_3\text{IrTi}_2\text{O}_9$ compound by replacing the role of Ir and Ti.

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I. PRELIMINARIES

This supplementary material supports our analytical and numerical study of the Heisenberg-Kitaev model on the triangular lattice given by the Hamiltonian
\[ \mathcal{H}_{\text{HK}} = J_H \sum_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + J_K \sum_{\gamma \parallel ij} S_\gamma^i S_\gamma^j. \]  

(5)

The triangular HK Hamiltonian (5) exhibits a rich phase diagram, which we will discuss in the following. We vary the relative strength of the Heisenberg and Kitaev couplings as a function of the ratio \( J_K/J_H \), which we parametrize on a circle as \( J_H = J \cos \alpha \) and \( J_K = J \sin \alpha \).

The outline of this supplemental material is the following: we first discuss in Sec. II the explicit form of the Klein duality transformation relating the right-hand and left-hand sides of the phase diagram parametrized in terms of an angle \( \tan \alpha = J_K/J_H \). In Sec. III we discuss the magnetically ordered phases in the triangular HK model. In particular, we will provide a detailed description of the expanded Luttinger-Tisza approximation, which we used to determine the formation and stability of the \( \mathbb{Z}_2 \)-vortex crystal phase in Sec. III A. In Sec. III B we subsequently turn to a discussion of the (quantum) order-by-disorder mechanism driven by the Kitaev interactions in the ferromagnetic phase. An extended discussion of the experimental signatures of the \( \mathbb{Z}_2 \)-vortex crystal in neutron polarimetry measurements is presented in Sec. IV. Finally, in Sec. V we turn to the physics of the pure Kitaev model.

II. DUALITY TRANSFORMATION

We start our discussion by reviewing the Klein duality relating couplings on the left and right-hand side of the circle phase diagram, see Fig. 6 b). Under this transformation, the Heisenberg-Kitaev Hamiltonian retains the same structure but the coupling parameters change as
\[ J_H \rightarrow -J_H, \quad J_K \rightarrow 2J_H + J_K. \]  

(6)

The transformation is performed by dividing the triangular lattice into four sublattices as illustrated in Fig. 6 a). Subsequently, each spin is subjected to a basis rotation, where the spins on the sublattice labeled “id” are not changed. For the three remaining sublattices each spin is rotated by \( \pi \) around the spin axis according to the sublattice labeling. Since a \( \pi \) rotation around one spin

Figure 6. (Color online) a) 24 site cluster with periodic boundary conditions containing all symmetries except for the rotational \( C_3 \) symmetry. The different symbols for the lattice sites indicate the four sublattices needed in the basis transformation underlying the Klein duality (6). b) Circle parametrization of the Heisenberg-Kitaev interactions \( J_H = J \cos \alpha \) and \( J_K = J \sin \alpha \) with the magenta lines indicating points on the left and right-hand side of the circle related by the Klein duality (6). The filled yellow and green circles indicate the points at which the Hamiltonian (5) is SU(2) symmetric.
axis effectively inverses the sign of the two other components, we can write the full transformation as

\begin{align}
\text{id} & : (S^x, S^y, S^z) \rightarrow (S^x, S^y, S^z) \\
x & : (S^x, S^y, S^z) \rightarrow (S^x, -S^y, -S^z) \\
y & : (S^x, S^y, S^z) \rightarrow (-S^x, S^y, -S^z) \\
z & : (S^x, S^y, S^z) \rightarrow (-S^x, -S^y, S^z).
\end{align}

Since this transformation is a simple local rotation of the spin basis, the original Hamiltonian and its counterpart after the transformation effectively describe the same physics, albeit for a resized unit cell. Interestingly, this transformation maps the SU(2) symmetric ferromagnetic and antiferromagnetic Hamiltonians at \( J_K = 0 \) and \( J_H = \pm 1 \) onto Heisenberg-Kitaev Hamiltonians with \( J_K = -2J_H \), revealing two more SU(2) symmetric points in the phase diagram. These points and their corresponding phases are termed the “stripy” (anti-)ferromagnets, due to the magnetic order after the basis rotation. The spin configurations at these points are illustrated in Fig. 9.

### III. PHASE DIAGRAM OF THE KITAEV-HEISENBERG MODEL ON THE TRIANGULAR LATTICE

In this section we will review in detail the analytical arguments revealing the various ordered phases of the triangular HK model (5). We start with the antiferromagnetically ordered states on the right-hand side of the circle phase diagram and subsequently the ferromagnetically ordered states on the left-hand side. The full phase diagram of the classical model is shown in Fig. 7 as well as the analytical estimates of the ground state energy in each of the ordered phases. The phase diagram of the quantum model, shown in Fig. 8, was obtained by exact diagonalization of small clusters. The phase boundaries are determined by identifying the values of \( \alpha \), where the second derivative \(-d^2E/d\alpha^2\) diverges, as indicated by the red line in Fig. 8.

#### A. Antiferromagnetic order

1. 120° antiferromagnet

For \( J_K = 0 \) and antiferromagnetic \( J_H > 0 \) the ground state is characterized by 120° ordering. Let us recapitulate the arguments revealing this order. To this end, we first consider the classically ordered states, for which we use the notation
\[ S_i = S\tilde{\Omega}(r_i) \] where \( r_i \) is the real-space vector pointing at lattice site \( i \) and \( \tilde{\Omega}(r) \) is a unit vector. The 120° state is then represented by a unit vector \( \tilde{\Omega}_{120°}(r) = e_1 \cos (Q \cdot r) + e_2 \sin (Q \cdot r) \) with e.g. \( Q = \frac{2\pi}{3}(1,0) \), and the orthonormal vectors \( e_i = (e^x_i, e^y_i, e^z_i)^T \), for which \( e_i \cdot e_j = \delta_{ij} \) with \( i,j = 1,2,3 \) and \( e_3 = e_1 \times e_2 \). The energy per site for this classical state is given by

\[
\varepsilon_{120°} = -S^2 \frac{1}{2}(3J_H + J_K).
\]  

However, for any finite \( J_K \) the 120° state becomes immediately unstable with respect to fluctuations, which we demonstrate in the following. We parametrize the fluctuations in the form

\[
\tilde{\Omega}(r) = \tilde{\Omega}_{120°}(r) \sqrt{1 - (\pi(r))^2} + \pi_1(r) (-e_1 \sin (Q \cdot r) + e_2 \cos (Q \cdot r)) + \pi_2(r)e_3,
\]

which ensures that \( \tilde{\Omega}^2(r) = 1 \) and \( \pi(r) = (\pi_1(r), \pi_2(r))^T \). Plugging this Ansatz in the Hamiltonian and expanding up to second order in the fluctuation fields one obtains for the energy \( E = N\varepsilon_{120°} + E^{(2)} \) with \( N \) denoting the number of lattice sites. The fluctuation part reads

\[
E^{(2)} = \sum_{\{ij\}} \left[ \left( -\varepsilon_{120°} \delta_{ij} - \frac{J_H S^2}{2} \right) \pi_{1i} \pi_{1j} + \left( -\varepsilon_{120°} \delta_{ij} + J_H S^2 \right) \pi_{2i} \pi_{2j} \right] \\
+ \sum_{\gamma \{ij\}} J_K S^2 \left[ \left( e^x_1 e^x_j \cos (Q \cdot r_i) \cos (Q \cdot r_j) + e^y_1 e^y_j \sin (Q \cdot r_i) \sin (Q \cdot r_j) \right) \pi_{1i} \pi_{1j} \\
- e^x_1 e^x_j \cos (Q \cdot r_i) \sin (Q \cdot r_j) \sin (Q \cdot r_i) \cos (Q \cdot r_j) \right) \pi_{1i} \pi_{1j} \\
+ e^y_3 e^y_3 \pi_{2i} \pi_{2j} + \left( -e^y_1 \sin (Q \cdot r_i) + e^y_2 \cos (Q \cdot r_i) \right) e^y_3 \pi_{1i} \pi_{1j} + (i \leftrightarrow j) \right],
\]

where \( \pi_{ai} = \pi_a(r_i) \) with \( a = 1,2 \). The fluctuation eigenmodes are determined with the help of the Fourier transform \( \pi_a(r_i) = \frac{1}{\sqrt{N}} \sum_{k \in \text{BZ}} e^{i k r_i} \pi_a(k) \). In the absence of the Kitaev interaction, \( J_K = 0 \), one obtains

\[
E^{(2)}|_{J_K=0} = \frac{1}{2} \sum_{k \in \text{BZ}} \left[ J_H S^2 \sum_{\gamma=x,y,z} (1 - \cos (k \cdot a_\gamma)) \pi^*_1(k) \pi_1(k) + J_H S^2 \sum_{\gamma=x,y,z} (1 + 2 \cos (k \cdot a_\gamma)) \pi^*_2(k) \pi_2(k) \right]
\]

with \( \pi^*_a(k) = \pi_a(-k) \). Whereas the \( \pi_1 \) mode becomes soft at the center of the Brillouin zone, i.e., at \( k = 0 \), the energy of the \( \pi_2 \) mode vanishes at its edge, e.g. for momenta \( k = \pm Q \). The zero modes \( \pi_1(k = 0) = \pi_2(\pm Q) \) identify three Goldstone modes that correspond to a long-wavelength rotation and tilting of the local orthogonal frame, respectively. In particular, the energy dispersion of the tilting mode \( \varepsilon_{\text{tilt}}^{\text{tilt}} |_{J_K=0} = J_H S^2 \sum_{\gamma=1,2,3} (1 + 2 \cos (k \cdot a_\gamma)) \) close to momentum \( Q \) possesses the form

\[
\varepsilon_{Q+k}^{\text{tilt}} |_{J_K=0} \approx J_H S^2 \frac{3}{8} k^2.
\]

A finite \( J_K \), however, immediately results in a negative energy eigenvalue and thus in an instability of the classical ground state. We can still diagonalize for the eigenenergies perturbatively in \( J_K \). In lowest order and in the long-wavelength limit the zero modes do not hybridize, and we obtain for the tilting mode a dispersion relation that is given in the long-wavelength limit, \( |k| \ll |Q| \), by

\[
\varepsilon_{Q+k}^{\text{tilt}} \approx J_H S^2 \frac{9}{4} k^2 - 2 J_K S^2 \sum_{\gamma=x,y,z} k \cdot a_\gamma \sin (Q \cdot a_\gamma) (\varepsilon^3_3)^2.
\]
It becomes maximally negative for a wavevector
\[
\mathbf{k}_{\text{inst}} = \frac{J_K}{J_H} \frac{4}{3} \sum_{\gamma=x,y,z} a_\gamma \sin(\mathbf{Q} \cdot \mathbf{e}_\gamma)(e^{2}_\gamma)^2 = \frac{J_K}{J_H} \left( \frac{1}{\sqrt{3}} \left[ (e^{y}_3)^2 + (e^{z}_3)^2 - 2(e^{y}_3)^2 \right] \right) \left( (e^{z}_3)^2 - (e^{y}_3)^2 \right),
\]
(13)
that can be expressed in terms of the normal \( \hat{e}_3 \). In the special case, where the spins of the 120° ordering are confined within the \( x\)-\( y \) plane and \( e_3 = 2 \), this wavevector is just given by \( \mathbf{k}_{\text{inst}} = J_K/\sqrt{3} (1, 0, 1)^T \). So it is the tilting Goldstone modes that trigger the instability of the 120° antiferromagnetic ordering in the presence of a finite Kitaev interaction \( J_K \).

2. Incommensurate antiferromagnet: \( \mathbb{Z}_2 \) vortex crystal

Having established the instability of the 120° antiferromagnetic ordering at \( J_H > 0 \) and infinitesimal Kitaev interaction \( J_K \), we now look for an alternative classical ground state. It turns out that an exact classical ground state is difficult to obtain analytically. However, using the Luttinger-Tisza approximation we can derive an approximate ground state that is a good finite secondary Fourier components.

Fourier components, \( \lambda \)

Having established the instability of the 120° antiferromagnetic ordering at \( J_H = 0 \) and infinitesimal Kitaev interaction \( J_K \), we now look for an alternative classical ground state. It turns out that an exact classical ground state is difficult to obtain analytically. However, using the Luttinger-Tisza approximation we can derive an approximate ground state that is a good approximation for large length scales, \( J_K |r|/J_H \gg 1 \), or, alternatively, for small momenta \( |\mathbf{q}| \ll J_K/J_H \). Note that this latter limit does not commute with \( J_K \to 0 \), and, as a consequence, does not smoothly connect with the Heisenberg point.

We start with the functional
\[
\mathcal{E} = J_K S^2 \sum_{\langle ij \rangle} \hat{\Omega}_i \cdot \hat{\Omega}_j + J_K S^2 \sum_{\gamma ||\langle ij \rangle\rangle} \hat{\Omega}_i^{\gamma \gamma} \hat{\Omega}_j^{\gamma \gamma} - \sum_i \lambda_i (\hat{\Omega}_i^2 - 1).
\]
(14)
The unit length of the vector \( \hat{\Omega}_i \) is locally imposed with the help of the Lagrange multiplier \( \lambda_i \). Upon spatial Fourier transformation the functional takes the form
\[
\mathcal{E}/N = \sum_{\mathbf{q}} \hat{\Omega}_{\mathbf{q}}^\alpha \mathcal{J}^{\alpha \beta}(\mathbf{q}) \hat{\Omega}_{\mathbf{q}}^\beta + \sum_{\mathbf{q}, \mathbf{p}} \lambda_{-\mathbf{q}+\mathbf{p}} \hat{\Omega}_{\mathbf{p}}^\alpha \hat{\Omega}_{\mathbf{q}}^\alpha - \lambda_0,
\]
(15)
where \( \lambda_0 = \lambda_{|\mathbf{q}|=0} \). The matrix \( \mathcal{J}^{\alpha \beta} \) possesses only diagonal entries with
\[
\mathcal{J}^{\alpha \alpha}(\mathbf{q}) = J_H S^2 \left( \cos(\mathbf{a}_x \cdot \mathbf{q}) + \cos(\mathbf{a}_y \cdot \mathbf{q}) + \cos(\mathbf{a}_z \cdot \mathbf{q}) \right) + J_K S^2 \cos(\mathbf{a}_\alpha \cdot \mathbf{q})
\]
(16)
and \( \mathcal{J}^{\alpha \beta}(\mathbf{q}) = 0 \) for \( \alpha \neq \beta \). At the Heisenberg point \( J_K = 0 \), the diagonal components of the matrix are minimal for momenta at the corner of the Brillouin zone, e.g. \( \mathbf{q} = \mathbf{Q} \), thus leading to 120° ordering. A finite \( J_K \), however, favours in general incommensurate order with wave vectors away from \( \mathbf{Q} \) as \( \mathcal{J}^{\alpha \alpha}(\mathbf{q}) \) become minimal for momenta of the form \( \mathbf{q}_\alpha^{(1)} = \mathbf{Q} - t \mathbf{a}_\alpha \) with \( t \in \mathbb{R} \). On the other hand, Fourier components, \( \hat{\Omega}_{\mathbf{q}_\alpha^{(1)}} \), of the spin with such incommensurate wave vectors induce finite Fourier components, \( \lambda_{\pm 2\mathbf{q}_\alpha^{(1)}}^{(1)} \) with \( \alpha = 1, 2, 3 \), of the Lagrange multiplier. Finite Lagrange multipliers \( \lambda_{\pm 2\mathbf{q}_\alpha^{(1)}} \), in turn induce two finite secondary Fourier components \( \hat{\Omega}_{\mathbf{q}_\alpha^{(2)}}^{(2)} \) with \( \mathbf{q}_\alpha^{(2)} = \mathbf{Q} - t (2\mathbf{a}_\beta - \mathbf{a}_\alpha) \) where \( \beta \neq \alpha \).

In the following, we discuss a Luttinger-Tisza approximation where we limit ourselves to the lowest finite Fourier components \( \hat{\Omega}_{\mathbf{q}_\alpha^{(1)}}^{(1)} \) and \( \hat{\Omega}_{\mathbf{q}_\alpha^{(2)}}^{(2)} \) for the spin and \( \lambda_0 \) and \( \lambda_{\pm 2\mathbf{q}_\alpha^{(1)}}^{(1)} \) for the Lagrange multiplier; all higher Fourier modes are neglected. In principle, this approximation can be systematically improved by including higher order modes. Minimizing the functional (15) we obtain for the energy per site
\[
\varepsilon_{\text{LT}}(t) = -\frac{S^2}{9} \left[ J_H \left( \cos \frac{\pi + 6t}{3} + 17 \sin \frac{\pi - 3t}{6} + 8 \sin \frac{\pi + 6t}{6} + \sin \frac{\pi + 15t}{6} \right) + J_K \left( \cos \frac{\pi + 6t}{3} + 8 \sin \frac{\pi + 6t}{6} \right) \right],
\]
(17)
which still depends on the parameter \( t \) that quantifies the distance of the primary Bragg peak from the corner of the Brillouin zone, \( \mathbf{q}_\alpha^{(1)} = \mathbf{Q} - t \mathbf{a}_\alpha \). The value of \( t_{\text{min}} \) identifying the position of the minimum of the function (17) finally determines the ground state energy \( \varepsilon_{\text{LT}}(t_{\text{min}}) \). It is shown as a green solid line in Fig. 7, and it is in excellent agreement with results from the classical Monte Carlo simulation. The energy \( \varepsilon_{\text{LT}}(t_{\text{min}}) \) for \( J_K \neq 0 \) is, in particular, smaller than the energy of the 120° state (8).

The corresponding state is given by
\[
\hat{\Omega}_i^{\alpha}(\mathbf{r}) = \frac{4}{3 \sqrt{3}} \Re \left[ e^{i\phi} \left(e^{i(\mathbf{Q} - t \mathbf{a}_\alpha) \cdot (\mathbf{r} - \mathbf{r}_0)} + \frac{1}{4} \sum_{\beta=x,y,z;\beta \neq \alpha} e^{i(\mathbf{Q} - t (2\mathbf{a}_\beta - \mathbf{a}_\alpha) \cdot (\mathbf{r} - \mathbf{r}_0))} \right) \right],
\]
(18)
Klein duality

\( \mathbb{Z}_2 \) vortex crystal
dual \( \mathbb{Z}_2 \) vortex crystal
(only one sublattice shown)

Figure 10. (Color online) Snapshots of spin configurations in the \( \mathbb{Z}_2 \)-vortex crystal (left) and the dual \( \mathbb{Z}_2 \)-vortex crystal (right). For clarity, only one of the three sublattices of the triangular lattice is shown. Yellow arrows point upwards out of the plane, while blue arrows point downwards out of the plane.

and the ground state is obtained by setting \( t = t_{\text{min}} \). In the Luttinger-Tisza approximation the length of the \( \hat{\Omega} \) vector is compromised to differ from unity, \( \sum_{\alpha=1}^{3}(\hat{\Omega}^\alpha(r))^2 \neq 1 \). Whereas the length \( |\hat{\Omega}(r)| \) varies in space it nevertheless remains always finite so that the orientation of \( \hat{\Omega}(r) \) is always well defined. Note that in the limit \( J_K \to 0 \) the distance \( t_{\text{min}} \to 0 \) and \( \varepsilon_{\text{LT}}(0) = -S^23J_H/2 \) recovers the exact ground state energy whereas the state itself, \( \hat{\Omega}_{r=0}(r) \), does not reproduce the 120° ordering as expected. According to Eq. (18), the relative weight between the primary and the secondary Bragg peaks as observed, e.g., in neutron scattering is predicted to be \( 1/4 = 1/16 \).

As discussed in the main text, the chirality evaluated with Eq. (18) reveals that this state corresponds to a \( \mathbb{Z}_2 \) vortex crystal. The orientation of this vortex crystal is fixed by the underlying triangular atomic crystal lattice. The ground state energy is found to be independent of the phase \( \phi \) and the choice of origin, i.e., the vector \( r_0 = (x_0, y_0)^T \).

3. Dual \( \mathbb{Z}_2 \) vortex crystal

From the duality transformation (6) follows that the antiferromagnetic Heisenberg point has a dual at \( J_H < 0 \) and \( J_K = -2J_H > 0 \). It follows immediately from the above consideration that the dual 120° ordering becomes unstable if \( J_K + 2J_H \neq 0 \) and a dual vortex crystal forms. The corresponding state and its energy are directly obtained from Eqs.(18) and (17) with the help of the duality transformation (6). The spin configuration of the \( \mathbb{Z}_2 \) vortex crystal and its dual partner are illustrated in Fig. 10.

B. Ferromagnetic order

1. \( \mathbb{Z}_6 \) Ferromagnet

We now address the case of a ferromagnetic Heisenberg interaction, \( J_H < 0 \). The classical ferromagnetic ground state is given by a constant, homogeneous spin configuration, \( \hat{\Omega}(r) \equiv \hat{\Omega} \) with \( \hat{\Omega}^2 = 1 \). The corresponding classical energy per site is independent of the orientation of \( \hat{\Omega} \) and reads

\[
\varepsilon_{\text{FM}} = S^2 \left( 3J_H + J_K \right)
\]  

(19)

For \( J_K = 0 \), this indeed corresponds to the exact ground state energy. Any finite \( J_K \), however, gives rise to fluctuation corrections to the ground state that also discriminate between the various orientations of \( \hat{\Omega} \). Performing a standard Holstein-Primakoff transformation, the spin-operator along the local \( z \)-axis, here defined by the classical vector \( \hat{\Omega} \), can be expressed as

\[
\hat{S}_i^z = S - a_i^\dagger a_i, \quad \hat{S}_i^z = a_i^\dagger \sqrt{2S - a_i^\dagger a_i}
\]

(20)
where \( \hat{S}^\pm_i = S_i^x \pm i S_i^y \). The spin-operator \( \hat{S} \) within the laboratory frame is related to \( \hat{S} \) by a rotation \( \mathbf{R} = R(0, 0, 1)^T \). Expanding the Hamiltonian in second order in the bosonic operators one obtains \( \mathcal{H} = N \varepsilon_{\text{FM}} + \mathcal{H}^{(2)} \) with

\[
\mathcal{H}^{(2)} = \frac{1}{2} \sum_{\mathbf{k} \in 1 \text{BZ}} (a_{\mathbf{k}}^\dagger a_{-\mathbf{k}}) h(\mathbf{k}) \left( a_{\mathbf{k}} \right) - \frac{S}{2} \sum_{\mathbf{k} \in 1 \text{BZ}} \left[ 2 J_H \sum_{\gamma=x,y,z} (\cos(\mathbf{k} \cdot \mathbf{a}_\gamma) - 1) + J_K \sum_{\gamma=x,y,z} (\cos(\mathbf{k} \cdot \mathbf{a}_\gamma) - 1)(1 - \hat{\Omega}^2_k) \right]
\]

where

\[
h(\mathbf{k}) = S \left[ 2 J_H \sum_{\gamma=x,y,z} (\cos(\mathbf{k} \cdot \mathbf{a}_\gamma) - 1) \mathbb{I} + 2 J_K \left\{ (\cos(\mathbf{k} \cdot \mathbf{a}_x) - 1) \left( e_x^+ e_x^- \mathbb{I} + (e_x^+)^2 \sigma^+ + (e_x^-)^2 \sigma^- \right) + (\cos(\mathbf{k} \cdot \mathbf{a}_y) - 1) \left( e_y^+ e_y^- \mathbb{I} + (e_y^+)^2 \sigma^+ + (e_y^-)^2 \sigma^- \right) + (\cos(\mathbf{k} \cdot \mathbf{a}_z) - 1) e_z^+ e_z^- (1 - \sigma^z) \right\} \right]
\]

with the Pauli matrices \( \sigma^x, \sigma^y, \) and \( \sigma^z \), and we used the abbreviations \( e^\pm = \frac{1}{\sqrt{2}} R(1, \pm i, 0)^T \) and \( \sigma^\pm = \frac{1}{2}(\sigma^x \pm i \sigma^y) \). With the help of a Bogoliubov transformation we can compute the correction to the classical ground state energy (19). In order to elucidate the analytical structure, we concentrate on the contribution to this correction only of lowest order in the Kitaev interaction,

\[
\delta \varepsilon_{\text{FM}} = -\frac{1}{4N} \sum_{\mathbf{k} \in 1 \text{BZ}} \frac{h_{21}(\mathbf{k}) h_{12}(\mathbf{k})}{h_{11}(\mathbf{k})} |_{J_K=0} \frac{1}{V_{1 \text{BZ}}} \int_{1 \text{BZ}} \frac{d\mathbf{k}}{V_{1 \text{BZ}}} \frac{(\cos(\mathbf{k} \cdot \mathbf{a}_\alpha) - 1) (\cos(\mathbf{k} \cdot \mathbf{a}_\beta) - 1)}{\sum_{\gamma=x,y,z} (1 - \cos(\mathbf{k} \cdot \mathbf{a}_\gamma))}
\]

To evaluate this expression we need the following integrals over the Brillouin zone

\[
\frac{1}{N} \sum_{\mathbf{k} \in 1 \text{BZ}} \frac{(\cos(\mathbf{k} \cdot \mathbf{a}_\alpha) - 1) (\cos(\mathbf{k} \cdot \mathbf{a}_\beta) - 1)}{\sum_{\gamma=x,y,z} (1 - \cos(\mathbf{k} \cdot \mathbf{a}_\gamma))} \xrightarrow{N \to \infty} \frac{1}{V_{1 \text{BZ}}} \int_{1 \text{BZ}} \frac{d\mathbf{k}}{V_{1 \text{BZ}}} \frac{(\cos(\mathbf{k} \cdot \mathbf{a}_\alpha) - 1) (\cos(\mathbf{k} \cdot \mathbf{a}_\beta) - 1)}{\sum_{\gamma=x,y,z} (1 - \cos(\mathbf{k} \cdot \mathbf{a}_\gamma))}
\]

\[
= \frac{6 \sqrt{3} - 2 \pi}{3 \pi} \delta_{\alpha \beta} + \frac{5 \pi - 6 \sqrt{3}}{6 \pi} (1 - \delta_{\alpha \beta}).
\]

Here, we evaluated the integrals in the thermodynamic limit where the volume of the first Brillouin zone is given by \( V_{1 \text{BZ}} = \frac{8 \pi^2}{\sqrt{3}} \) using the identities

\[
(e_x^+ e_x^-)^2 + (e_y^+ e_y^-)^2 + (e_z^+ e_z^-)^2 = 2
\]

\[
= - (e_x^+ e_y^-)^2 - (e_y^+ e_x^-)^2 + (e_x^+ e_z^-)^2 + (e_z^+ e_x^-)^2 + (e_y^+ e_y^+)^2 + (e_z^+ e_z^-)^2 = \frac{1}{4} \left( 1 + \hat{\Omega}_x^4 + \hat{\Omega}_y^4 + \hat{\Omega}_z^4 \right).
\]

The fluctuation correction to the energy in lowest order in the Kitaev interaction finally assumes the form

\[
\delta \varepsilon_{\text{FM}} = -\frac{S J_K^2}{2 |J_H|} \frac{3(2 \sqrt{3} - \pi)}{8 \pi} \left( 1 + \hat{\Omega}_x^4 + \hat{\Omega}_y^4 + \hat{\Omega}_z^4 \right),
\]

and is illustrated in Fig. 11. This correction favors the vector \( \hat{\Omega} \) to point along one of the six equivalent \((100)\) directions (as \( 2 \sqrt{3} - \pi > 0 \)). Whereas at the Heisenberg point, \( J_K = 0 \), the ferromagnetic ground state manifold is the 2-sphere, a finite Kitaev interaction reduces this manifold to only six points corresponding to a \( \mathbb{Z}_6 \) ferromagnetic order parameter.

2. Dual \( \mathbb{Z}_6 \) Ferromagnet

The dual point of the Heisenberg ferromagnet is located at \( J_H > 0 \) and \( 2 J_H + J_K = 0 \). The classical ground state energy of the dual ferromagnetic state is obtained from Eq. (19),

\[
\varepsilon_{\text{dFM}} = S^2 \left( - J_H + J_K \right).
\]

The dual ferromagnetic state is the exact ground state at \( 2 J_H + J_K = 0 \). Fluctuations arise for a finite \( 2 J_H + J_K \neq 0 \), which, following the arguments of the previous section, then favor a dual \( \mathbb{Z}_6 \) ferromagnetic ordering.
Figure 11. (Color online) a) Ground state energy of the quantum ferromagnet in an external Zeeman field as a function of the direction of the applied magnetic field $\mathbf{B}$, where we have subtracted the ground state energy for $\mathbf{B} = \hat{z}$. The Kitaev coupling strength is $J_K/J_H = \tan(11\pi/10) \approx 0.32$. The energy is minimal when the magnetization is pinned along one of the three axes, and maximal when pointing along the space diagonals. b) The same results shown for the cut along the yellow line in a). Each line corresponds to a different value of $J_K/J_H$. For $J_K/J_H = 0$ the ground state energy does not depend on the direction of the magnetization. Upon increasing $J_K/J_H$ up to $J_K/J_H = \tan(11\pi/10)$, the directional dependence becomes more and more pronounced. The dashed line is a fit of Eq. (29).

3. Numerical results

To corroborate our analytical results for the pinning of the magnetization when $J_K \neq 0$ around the ferromagnetic Heisenberg point, we performed exact diagonalization calculations on small systems. We implemented lattice clusters with periodic boundary conditions containing 12 sites, with a geometry that preserves the $C_6$ rotational symmetry of the triangular lattice. By applying a small magnetic field $\mathbf{B}$ to each spin, where

$$\mathbf{B} = B \begin{pmatrix} \cos(\phi) \sin(\theta) \\ \sin(\phi) \sin(\theta) \\ \cos(\theta) \end{pmatrix},$$

where $\phi \in [0, 2\pi)$ and $\theta \in [0, \pi]$, the magnetization was adjusted to point in different directions. Fig. 11a) shows results for the change in the ground state energy as a function of the orientation of $\mathbf{B}$, taken relative to the case when $\mathbf{B} \parallel \hat{z}$, for a small finite Kitaev coupling $J_K/J_H = \tan(11\pi/10) \approx 0.32$. We find that, in agreement with our analysis above, the ground state energy of the system is minimal when the magnetization points along one of the three spin axes, see Fig. 11a). Scanning the orientation of $\mathbf{B}$ along the yellow line shown in Fig. 11a), we compare the effect of different Kitaev couplings. In Fig. 11b) each line corresponds to a different value of $J_K$. For $J_K = 0$, the energy does not depend on the direction of $\mathbf{B}$. However, for any $J_K \neq 0$, minima in the energy occur immediately, becoming more pronounced as $J_K$ grows. The black dashed line in Fig. 11b) is a fit of Eq. (29), showing perfect agreement.
IV. NEUTRON POLARIMETRY ON THE MAGNETIC STRUCTURE OF THE $\mathbb{Z}_2$ VORTEX CRYSTAL

The structure factor of the $\mathbb{Z}_2$ vortex crystal possesses as a hallmark of the Kitaev interaction a characteristic correlation between the positions of the Bragg peaks and the associated spin-components, see Fig. 2(b) of the main text. We suggest that this correlation can be resolved with the help of neutron polarimetry.

The probability that an incoming neutron with spin $\sigma_{\text{in}}$ is scattered into a spin-state $\sigma_{\text{out}}$ is given by the energy-integrated scattering cross section $\sigma_{\sigma_{\text{out}}, \sigma_{\text{in}}}(\mathbf{q})$, where $\mathbf{q}$ is the transferred momentum. Consider a polarizer and analyzer with an orientation specified by the unit vectors $\mathbf{e}_{\text{in}}$ and $\mathbf{e}_{\text{out}}$, respectively. The total probability and the relative probability that a neutron is detected with spin $\pm \mathbf{e}_{\text{out}}$ is then given by

$$\sigma(\mathbf{q}, \mathbf{e}_{\text{out}}, \mathbf{e}_{\text{in}}) = \sum_{\tau_{\text{out}} = \pm 1} \sigma_{\tau_{\text{out}} \sigma_{\text{out}}, \sigma_{\text{in}}}(\mathbf{q}), \quad \text{and,} \quad \Delta \sigma(\mathbf{q}, \mathbf{e}_{\text{out}}, \mathbf{e}_{\text{in}}) = \sum_{\tau_{\text{out}} = \pm 1} \tau_{\text{out}} \sigma_{\tau_{\text{out}} \sigma_{\text{out}}, \sigma_{\text{in}}}(\mathbf{q}),$$

respectively. The polarization is then defined by the ratio $P(\mathbf{q}, \mathbf{e}_{\text{out}}, \mathbf{e}_{\text{in}}) = \Delta \sigma(\mathbf{q}, \mathbf{e}_{\text{out}}, \mathbf{e}_{\text{in}})/\sigma(\mathbf{q}, \mathbf{e}_{\text{out}}, \mathbf{e}_{\text{in}})$. In the following, we concentrate on the magnetically ordered phase when the scattering probabilities are dominated by magnetic Bragg scattering so that we can neglect all nuclear contributions. For the particular choice that the axis of polarizer and analyzer coincide, $\mathbf{e}_{\text{out}} = \mathbf{e}_{\text{in}} = \mathbf{e}$, but are orthogonal to the transferred momentum $\mathbf{e} \perp \mathbf{q}$, the polarization attributed to magnetic scattering then simplifies to

$$P_{\text{mag}}(\mathbf{q}, \mathbf{e}) |_{\mathbf{e} \perp \mathbf{q}} = 2 \frac{\epsilon_{ij} \chi_{ij}(\mathbf{q}) \epsilon_{j}}{\chi_{kl}(\mathbf{q})(\delta_{kl} - q_k q_l)} - 1,$$

where $\chi_{ij}(\mathbf{q}) = \chi_{ij}(\mathbf{q}, \omega = 0)$ is the magnetic susceptibility at zero frequency,

$$\chi_{ij}(\mathbf{q}, \omega) = 2i \int_{0}^{\infty} dt \ e^{i\omega t} \frac{1}{2} \langle [M_i(\mathbf{q}, t), M_j(-\mathbf{q}, 0)] \rangle.$$

The magnetic structure factor of the $\mathbb{Z}_2$ vortex crystal has only non-zero diagonal components, $\chi_{ii}$, which however differ from each other and, moreover, possess different Bragg peak positions. For example, for our choice of the Kitaev interaction the $\chi_{zz}$ component is expected to exhibit a primary Bragg peak at $\mathbf{q}^{(1)} = \mathbf{Q} - \mathbf{a}_z = \frac{1}{a} \left( \frac{1}{3} a (1, 0, 0) - t (\frac{1}{2}, -\frac{\sqrt{3}}{2}, 0) \right)$ where $a$ is the lattice constant and we assumed for simplicity that the two-dimensional triangular lattice lies in the $x$-$y$ plane. Measuring at this particular Bragg peak, one expects for $\mathbf{e} = \hat{z}$ the value $P_{\text{mag}} = 1$ in contrast to $P_{\text{mag}} = -1$ that is obtained for $\mathbf{e}$ in the direction perpendicular to $\hat{z}$ and $\mathbf{q}$. A systematic variation of the analyzer/polarizer orientation $\mathbf{e}$ should therefore allow, in principle, to resolve the correlation between the diagonal components $\chi_{ii}$ and their Bragg peak position.

V. KITAEV LIMIT

To calculate the energies of the ground state and the first few excited states at the Kitaev point we performed calculations using the density matrix renormalization group (DMRG)$^6$. Once the ground state was found, we targeted excited states by successively calculating states of lowest energy that are orthogonal to all previously found states. While the DMRG is highly successful for 1D systems, it can also be extended to systems with a small finite width, and we considered triangular lattice systems of width 3 and 4 and varying length with open boundary conditions. We ran calculations at bond dimensions $M = 600, 800, 1000$ making sure that the energies converged.

The geometry of the considered lattice clusters breaks the $C_6$ symmetry of the lattice and the spins order antiferromagnetically in the spin component corresponding to the interaction term along the longer direction. In Fig. 12 we show the energy differences between the lowest 8 excited states and the ground state, alongside spin-spin correlators. The first three excited states collapse exponentially onto the ground state energy as the length of the system increases. Likewise, the next four excited states collapse to the same energy, however growing linearly in system length. From the calculated spin-spin correlators we can identify this excitation to be given by a breaking of the antiferromagnetic ordering between next-nearest neighbor chains. Finally the 8th excited level corresponds to a local defect in a chain, which is indicated by the vanishing spin correlation in the center left corner of the lattice cluster. Fig. 13 shows the spin correlations in the ground states for systems of width 3 and 4 at the antiferromagnetic Kitaev point ($J_H = 0$). While nearest neighbor chains are uncorrelated, there is a clear antiferromagnetic correlation between next-nearest neighbor chains in the spin component given by the chain direction. This mechanism locks the spin alignment of next-nearest neighbor chains to each other and thus reduces the degeneracy of the ground state from $3 \times 2^L$ to $3 \times 2^2$. Other spin components show only very short-ranged correlations as shown in the lower two panels of Fig. 13.
Figure 12. (Color online) Energy gaps of a $3 \times L$ triangular lattice strip with open boundary conditions. All values are given in relation to the ground state energy $E_0$, i.e. $E_1 = E_1 - E_0$. The figures on the right show numerical results for $\langle S^x_{r_0} S^x_r \rangle$ spin correlations, where the black disk with the white dot indicates the position $r_0$, the diameter of the disks indicates the strength of the correlation and the color indicates the sign, with red corresponding to negative (antiferromagnetic) and black to positive (ferromagnetic) correlations. For details, see the main text.

Figure 13. (Color online) Spin spin correlations in the ground state of the antiferromagnetic Kitaev model on the triangular lattice. Black circles indicate a positive correlations, $\langle S^x_j S^x_i \rangle > 0$, whereas the red circles denote negative correlation. The small white dot indicates the position $r_0$. The geometry of the lattice clusters lifts the degeneracy of the lattice direction, favoring chains coupling antiferromagnetically in their $x$-component along the $x$-direction. Chains are not coupling to their neighbor chains, however they couple antiferromagnetically to their next-nearest neighbor chains. Along the $y$- and $z$-directions the correlations are suppressed.

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