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Incommensurate spiral order is a common occurrence in frustrated magnetic insulators. Typically, all magnetic moments rotate uniformly, through the same wavevector. However the honeycomb iridates family Li_2IrO_3 shows an incommensurate order where spirals on neighboring sublattices are counterrotating, giving each moment a different local environment. Theoretically describing its spin dynamics has remained a challenge: The Kitaev interactions proposed to stabilize this state, which arise from strong spin-orbit effects, induce magnon umklapp scattering processes in spin-wave theory. Here we propose an approach via a (Klein) duality transformation into a conventional spiral of a frustrated Heisenberg model, allowing a direct derivation of the dynamical structure factor. We analyze both Kitaev and Dzyaloshinskii-Moriya based models, both of which can stabilize counterrotating spirals, but with different spin dynamics, and we propose experimental tests to identify the origin of counterrotation.

Introduction. Quantum spin liquid phases [1] have enjoyed renewed attention in recent years, driven by candidate material platforms. Possible experimental settings in magnetic insulators [2] include the layered kagome systems, the nearly-metallic organics, as well as iridates including the recently explored family of honeycomb iridates, $(Na/Li)_2IrO_3$ and the related α-$RuCl_3$, distinguished by their significant spin-orbit coupling. Here Ir^{4+} (Ru^{3+}) hosts an effective $S=1/2$, observed to order magnetically at low temperature. While Na_2IrO_3 and α-$RuCl_3$ show collinear zigzag antiferromagnetism [3–14], the three structural polytypes of the lithium iridate, α,β,γ-Li_2IrO_3, all order into an unconventional incommensurate magnetic phase, involving counterrotating spirals [15–19].

Recent experiments on β-Li_2IrO_3 under high pressures [16] as well as hydrogenated α-Li_2IrO_3 [20] under ambient pressure found no evidence for magnetic long-range order at base temperatures, raising the interesting possibility of a transition into a long-sought Kitaev quantum spin liquid. Robustly identifying the properties of such a phase is experimentally rather challenging as the defining long-range entanglement cannot be directly measured in a solid, and the expected emergent fractionalized excitations are predicted to produce only broad spectral features [21–26]. A possible route to quantify proximity to spin-liquid physics is through a knowledge of the appropriate Hamiltonian in the magnetically-ordered phase, whose properties could in principle be more directly accessible experimentally. This requires detailed predictions for characteristic signatures in the spin dynamics for various Hamiltonians to be able to distinguish between competing models.

The counterrotating spiral orders in α,β,γ-Li_2IrO_3 offer a promising avenue for such an approach. However, theoretically computing the spin dynamics has proven to be a nontrivial task. As we show below, the barrier consists of strong magnon umklapp scattering, associated both with the nonuniform spin environment of counterrotation as well as with the lack of any continuous spin rotation symmetry in the Hamiltonian. A similar issue was recently discussed for β-$CaCr_2O_4$ [27,28]. Easy-axis and easy-plane anisotropy, as well as antisymmetric Dzyaloshinskii-Moriya (DM) exchange, which are expected to arise from spin-orbit coupling, can preserve a continuous SO(2) symmetry subgroup; in contrast, the "Kitaev" exchange of Kitaev's honeycomb spin liquid [29], proposed to arise in the honeycomb iridates [30–35], breaks it down to a discrete subgroup. Such a reduced symmetry in a minimal Hamiltonian implies a remarkable spin-orbit coupling effect.

In this Rapid Communication we theoretically analyze the spin dynamics of a minimal 1D model on a zigzag chain with coplanar xy spiral order with counterrotation on top/bottom sites as shown in Fig. 1(a). This captures the unifying common feature of the magnetic structures in all three Li_2IrO_3 structural polytypes; the actual structures differ in the value of the spin rotation angle, the magnitude of the tilt of the rotation plane away from the xy plane, and the pattern of those tilts between adjacent chains, and we consider the tilts to be secondary features left for future work. We describe the spin rotation along the zigzag chain via a magnetic ordering wave vector q in units of $2\pi/a_1$, where $a_1 = 5.16$ Å is the repeat distance along the zigzag chain, see Fig. 1(c). In this description [17–19] $q = 0.32$ for α and 0.28 for β and γ-Li_2IrO_3. It is important to note [36] that while for maximum generality and simplicity we focus here on the parent 1D model, ultimately we want properties that are relevant for 3D systems. Hence we are not interested in the true quantum excitations of an isolated 1D chain [37], which are usual 1D spinons. Instead, using the spin-wave method we expose precisely those features which are common to the 2D and 3D ordered materials. Our goal is to capture the "semiclassical" quantum fluctuations, appropriate for the real materials, within a unified transparent setting.

The Hamiltonians we study are constructed as the Klein duals of the known parent Hamiltonians for conventional spirals. The Klein duality, a four-sublattice spin transformation whose site-dependent π rotations connect to the Kitaev exchange via the multiplication rules of the Klein four group, was previously used to expose a fluctuation-free point in a stripy antiferromagnet [31] among other contexts [31,35,38–43]. Here we find that it transforms a co-rotating spiral in a frustrated J_1-J_2 model into a counterrotating spiral in a Kitaev-based model, with additional J_2 xy anisotropy appropriate for the xy-coplanar spiral mode. We compare this mechanism
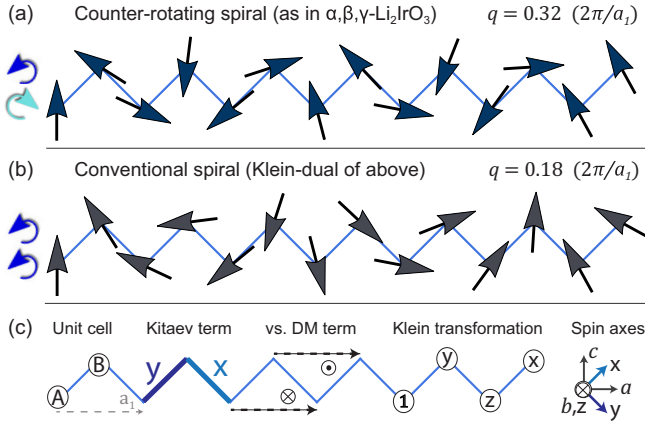
FIG. 1. Counterrotating spiral order of α,β,γ-Li$_2$IrO$_3$ as the Klein dual of a conventional (co-rotating) spiral. (a): The counterrotating spiral on a zigzag chain is the unifying common feature of the magnetic structures of all three α,β,γ-Li$_2$IrO$_3$ honeycomb iridates. The bottom sublattice rotates clockwise, while the top rotates counterclockwise. (b): The co-rotating spiral, of a conventional Heisenberg J_1-J_2 model, transforms by Klein duality into a counterrotating spiral with a Kitaev-J_1-J_2 model and xy anisotropy. (c): Competing models to stabilize counterrotation: Kitaev exchange (x,y) or second-neighbor Dzyaloshinskii-Moriya (DM) exchange (out/in for top/bottom bonds). The Klein transformation $\mu \in \{1,x,y,z\}$ acts as identity 1 or by π rotation around a spin's x,y,z axis. This exact duality for the counterrotating spiral shows its stability and circumvents the magnetic umklapp of its Kitaev exchange for computing its dynamical structure factor.

against a model of antisymmetric DM couplings, here required to be purely intrasublattice and with a sublattice-dependent orientation [44]. We compute the dynamical spin structure factor for various models of both classes, through a rotating frame exposed by the duality transformation. The dynamics in the Kitaev-based model are found to be quite unusual but can be interpreted via the duality to the J_1-J_2 model's well-understood dynamics.

The general Hamiltonian consists of the following:

$$H = \sum_{\langle ij \rangle} \left[K S_i^{\gamma_{ij}} S_j^{\gamma_{ij}} + J_1^{xy} \left(S_i^x S_j^x + S_i^y S_j^y \right) + J_1^z S_i^z S_j^z \right]$$
$$+ \sum_{\langle\langle ij \rangle\rangle} \left[J_2^{xy} \left(S_i^x S_j^x + S_i^y S_j^y \right) + J_2^z S_i^z S_j^z \pm D_2 \, \hat{z} \cdot \vec{S}_i \times \vec{S}_j \right],$$

(1)

where $\langle ij \rangle$ and $\langle\langle ij \rangle\rangle$ refer to first and second neighbor bonds, respectively, $\gamma_{ij} \in \{x,y\}$ is the Kitaev bond type, and the Dzyaloshinskii-Moriya coupling D_2 is oriented as in Fig. 1(c), with $j > i$ and $\pm$ sign for the A/B sublattice.

Classical ground states: mechanism for stability of the counterrotating spiral. First let us consider how to stabilize the corotating and counterrotating spirals as ground states for various terms in this Hamiltonian. There are two known mechanisms for stabilizing conventional (corotating) spiral orders: (A) Frustration from competing exchanges, such as ferromagnetic nearest-neighbor and antiferromagnetic second-neighbor exchanges; and (B) DM couplings. As an example of mechanism (A), we take a J_1-J_2 ($K = 0$) model with $J_1 < 0$

and $J_2 > 0$; its classical ground state is a spiral order with a rotation angle between consecutive sites $\arccos(-J_1/4J_2)$ for $J_2 > |J_1|/4$. For mechanism (B), the rotation angle is $\arctan(D/J_1)$ for the usual nearest-neighbor DM model. When the zigzag chain separates into two decoupled A/B chains with DM interaction of opposite sign the angle of rotation for each chain is $\theta_{A,B} = \pm \arctan(D_2/J_2)$.

The Klein duality, which maps a conventional co-rotating spiral to a counterrotating spiral, transforms these conventional spiral Hamiltonians to produce Hamiltonians for the counterrotating spiral. It is easy to see (Fig. 1) how the classical conventional spiral order is transformed, by the rules of the Klein transformation, into the counterrotating spiral order, with $q \to \pi/a_1 - q$. Let us then consider how the transformation acts on the Hamiltonians for mechanisms (A) and (B) above, or relatedly on the Hamiltonian Eq. (1) at $K = 0$ and uniform orientation of the DM term ($-D_2$ rather than $\pm D_2$). It is easy to show the following action for the Klein transformation:

$$\left(J_1^{xy}, J_2^{xy}, J_1^z, J_2^z \right) \leftrightarrow \left(-J_1^{xy}, -J_2^{xy}, -J_1^z, +J_2^z \right) \quad (2)$$

$$-D_2 \leftrightarrow \pm D_2 \quad (3)$$

$$(K = 0) \leftrightarrow \left(K = -2J_1^{xy} \right). \quad (4)$$

A Kitaev term is produced, with twice the magnitude and opposite sign relative to the J_1^{xy} term. This transformation is a duality, i.e., it maps Eq. (1) to itself with a different set of parameters.

A known Hamiltonian for a conventional spiral thus produces a Hamiltonian for the counterrotating spiral, via the mapping above. The dual of mechanism (B) is obvious—one can force counterrotation between sublattices by giving opposite signs to pure-second-neighbor (intrasublattice) DM terms, as in Eq. (1). The dual of mechanism (A) however produces a Kitaev-based model, with additional first and second neighbor Heisenberg-type terms, whose classical ground state is the counterrotating spiral. We note that the Klein duality necessarily introduces easy-plane anisotropy via the differing transformation of J_2^z. Since the J_1^z, J_2^z couplings do not change the nature of the spiral order when the spin rotation plane is xy, i.e., for sufficient easy-plane xy anisotropy, a minimal description is afforded by setting $J_1^z = J_2^z = 0$. The result (Fig. 2) is a Kitaev-J_1^{xy}-J_2^{xy} model whose classical ground state is the counterrotating spiral.

Spin dynamics and magnetic umklapp from spin-orbit coupling. To compute the dynamical structure factor via spin wave theory, one transforms the Hamiltonian Eq. (1) into a "rotating" (or "moving") frame, i.e., a site-varying coordinate system which is locally aligned with the spin orientation in the ordered spiral configuration. In the following we find it convenient to use the orthorhombic axes (a,b,c) instead of the Kitaev (x,y,z) axes for the spin components, with the relation [45] $\hat{x} = (\hat{a} + \hat{c})/\sqrt{2}$, $\hat{y} = (\hat{a} - \hat{c})/\sqrt{2}$ and $\hat{z} = \hat{b}$ shown in Fig. 1(c), where $\hat{x}$ indicates a unit vector along x and so on. Let $R[\theta]$ be a rotation by angle θ around the spin $z \equiv b$ axis. The local spin orientation in the wave-vector-q spiral is expressed by $e^3 \equiv R[-\eta_s qr] \cdot \hat{c}$. Here the sublattice sign η_s is $\eta_s = \mp$ on the A/B sublattice for the counterrotating
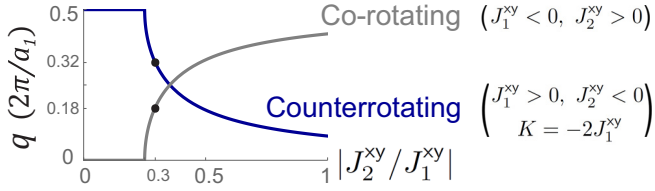
FIG. 2. Duality of classical spirals. The classical Heisenberg J_1-J_2 model with ferromagnetic $J_1 < 0$ and frustrating second-neighbor $J_2 > 0$ has a (co-rotating) spiral ground state with nonzero wavevector q for $|J_2/J_1| > 0.25$ (gray curve). With easy-plane xy anisotropy, the resulting xy-plane spiral is independent of J_1^z, J_2^z and depends only on J_2^{xy}/J_1^{xy}. The Klein transformation produces a counterrotating spiral with $q \to \pi/a_1 - q$ (blue curve) while flipping the signs of $J_1^{xy}, J_2^{xy}, J_1^z$, preserving J_2^z and creating a Kitaev exchange $K = -2J_1^{xy}$. The resulting model has the counterrotating spiral shown in Fig. 1(a) as its classical ground state.

spiral, or is uniformly $\eta_s = +$ for the co-rotating spiral. The local coordinate system $e^\pm \equiv R[-\eta_s qr] \cdot (\hat{a} \pm i\hat{b})$ can then be used to write the spin operator as $\vec{S} = e^3 S^3 + (e^- S^+ + e^+ S^-)/2$. In the $1/S$ spin wave expansion, $S^3 \to 1/2 - b^\dagger b$ and $S^\pm \to b, b^\dagger \equiv a^\pm$. The spin wave Hamiltonian is then $H_{\mathrm{SW}} = \sum_{ij} [\tilde{J}_{i,j}^{\mu\rho} \sigma_{\rho\nu}^1/8 - \delta_{\mu\nu}\delta_{ij} E_{\mathrm{cl}}/2](a_i^\mu)^\dagger a_j^\nu$ with repeated indices summed. The important ingredient is the interaction matrix in the rotating frame, $\tilde{J}_{i,j}^{\mu,\nu} \equiv e_i^\mu \cdot J_{i,j} \cdot e_j^\nu$, where $J_{i,j}$ is the spin interaction matrix between spins i, j associated with Eq. (1).

We thus turn to evaluate the interactions in the rotating frame, $\tilde{J}_{i,j}^{\mu\nu}$. The rotation around $z \equiv b$ leaves $\hat{b}$ invariant, $e^\pm = (R \cdot \hat{a}) \pm i\hat{b}$, so its effects are contained in the $R \cdot \hat{a}$ component; for concreteness, we can isolate it by setting $J_1^z = J_2^z = 0$, in which case $\tilde{J} \to \hat{a} \cdot R^T \cdot J \cdot R \cdot \hat{a}$. Evaluating this term on nearest-neighbor bonds (i, j), which connect opposite sublattices, we find [36]

$$\tilde{J}_{i,j} = \hat{a} \cdot R^T[-\eta_s qr] \cdot J_{i,j} \cdot R[\eta_s q(r + a_1/2)] \cdot \hat{a}$$
$$= -\frac{K}{2}\sin\left(\frac{qa_1}{2}\right) + \left[J_1^{xy} + \frac{K}{2}\right]\cos\left(\frac{qa_1}{2} + 2qr\right),$$
$$(5)$$

where $\eta_s = \mp$ is defined by the A/B sublattice of site i, at position r. The explicit dependence on coordinate r in the last term—the rotated Hamiltonian is not translationally invariant—changes the spin wave physics drastically. This is exposed by Fourier transform, where the expression above produces magnetic umklapp terms such as $b_k^\dagger b_{k+2q}$. The magnons experience magnetic umklapp scattering that changes their wave vector by multiples of q. Even if q is taken to be approximately commensurate, the wave-vector quantum number k is lost outside of a highly-folded magnetic Brillouin zone; for incommensurate q, the magnon wave vector k becomes ill defined.

One might generally expect to lose the wave-vector quantum number k when translation symmetry is fully broken by an incommensurate order; this is masked in conventional spirals through a rotating frame, which relies on continuous SO(2) rotation symmetry in the model Hamiltonian. The SO(2)-symmetric J_2^{xy}-D_2 second-neighbor model of the counterro-

tating spiral can similarly preserve the magnon wave vector k. However the counterrotation configuration means that each spin has a different local (nearest-neighbor) environment, giving rise to magnetic umklapp processes even through the SO(2)-symmetric J_1^{xy} term, as well as through the discrete symmetry K terms. The loss of k as a good quantum number is fully apparent.

Here we circumvent the magnetic umklapp scattering by tuning parameters to the duality with the co-rotating spiral. Recall from Eq. (4) that the counterrotating spiral Hamiltonian with $K = -2J_1^{xy}$ is dual to a J_1-J_2 XY model. The continuous SO(2) symmetry group of the XY model is preserved in an altered form by the duality, allowing the Hamiltonian at $K = -2J_1^{xy}$ to preserve the magnon quantum numbers. Indeed, Eq. (5) shows that the translation symmetry in the rotated frame is restored when $K + 2J_1^{xy} = 0$. We proceed by analyzing this case. Perturbations away from this parameter point will generically open gaps in the spin wave dispersions via Bragg reflections through multiples of the spiral wave vector q, such as at wave vectors $k = \pm q$, as well as mix the S^a, S^c spin polarizations.

Using the counterrotating spiral model produced by the duality, the dynamical structure factor can be computed straightforwardly by diagonalizing the spin wave Hamiltonian. The results are shown in Fig. 3 for various polarizations as well as for a spherical average relevant to powder samples. The Kitaev-based model shows unusual features, which are nevertheless transparently related, via the Klein duality, to the usual features from the conventional spiral. The duality shifts magnon wave vectors by $\pm\pi/a_1$ for the S^a and S^c spin components, respectively, and by $2\pi/a_1$ (corresponding to Néel correlations) for the S^b spin component. The Bragg peaks and intensity pattern are thus found by appropriately shifting the known structure factor of the J_1-J_2 conventional spiral. Observe that the counterrotating spiral can be considered as a sum of two distinct S^a, S^c spin density waves $\pi/2$ out-of-phase. The two sublattices have in-phase S^c but π-out-of-phase ($2\pi/a_1$-modulated) S^a, producing S^c-polarized Bragg peaks at $k = 0 \pm q$, but S^a-polarized Bragg peaks at $k = 2\pi/a_1 \pm q$. Universal, linearly-dispersing Goldstone modes with the same polarization as the Bragg peaks emerge from q and $2\pi/a_1 \pm q$ positions. The S^{bb} dynamical correlations (out-of-plane fluctuations) contain a mode with maximum energy and strong intensity at the zone center ($k = 0$), as in other Kitaev-based models [41]. We expect these generic features survive when the 1D chains are coupled together [45] as in the actual 2D and 3D honeycomb iridates and to help distinguish between Kitaev or other exchange models.

Conclusion. We have identified a transparent theoretical mechanism for the key feature in the unconventional magnetic orders recently observed in three honeycomb iridates. These materials host different crystal structures, but nonetheless their magnetism shares the unifying feature of counterrotating spirals, with opposite handedness in neighboring sublattices. This magnetic configuration, as well as a Kitaev-based parent Hamiltonian, are constructed by acting with the Klein duality on the well-understood frustrated J_1-J_2 model of a spiral order. This connection also enables us to solve for the spin dynamics in this system and to interpret them transparently. We have
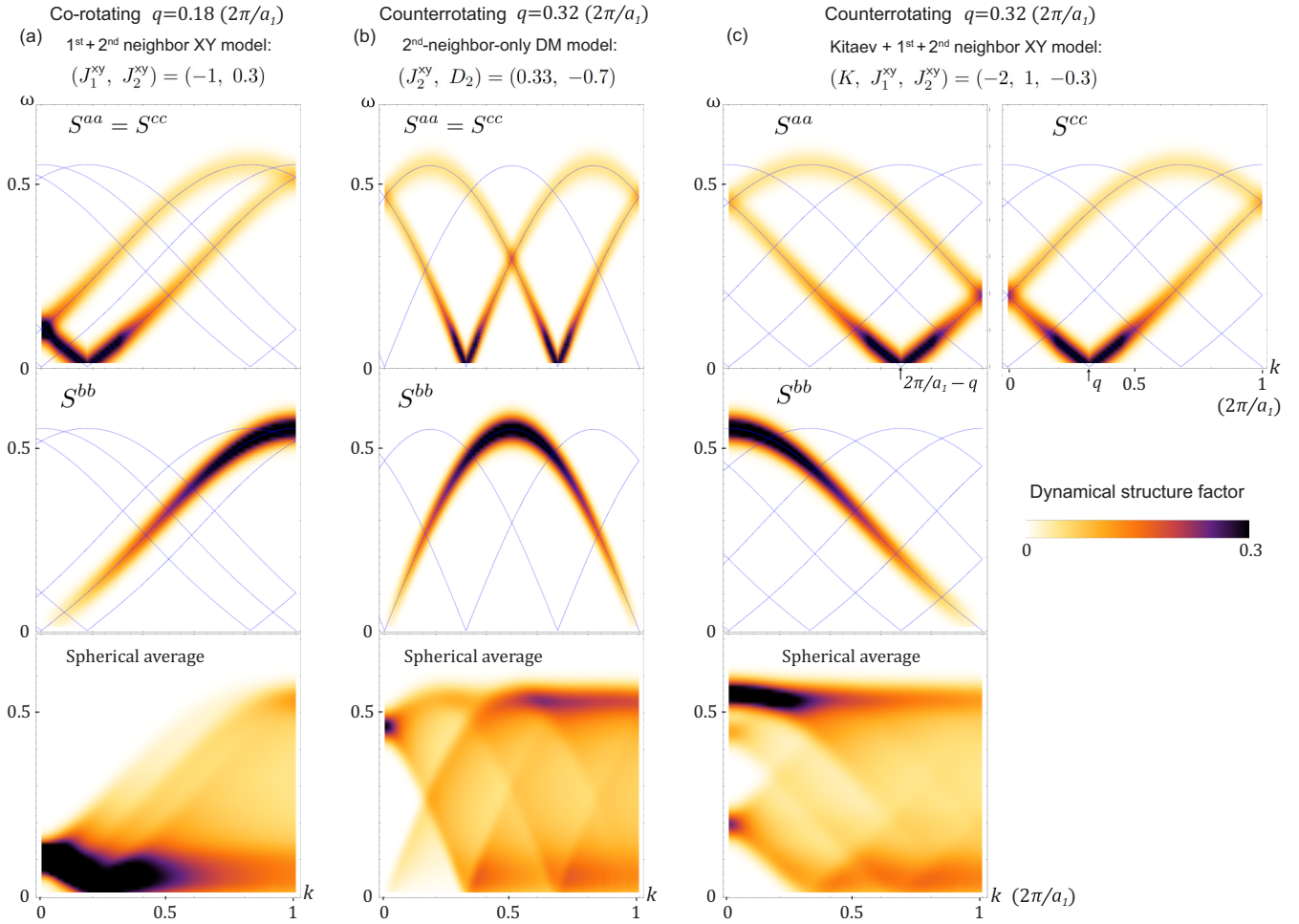
FIG. 3. Dynamical structure factor signature of Kitaev exchange. The dynamical correlations of various spin polarizations [a,b,c axes defined in Fig. 1(c)] are computed via spin wave theory for two possible models of the counterrotating spiral: decoupled sublattices with pure-second-neighbor DM exchanges of opposite signs (column b), and nearest-neighbor Kitaev exchange together with smaller easy-plane J_1-J_2 (column c). The plots shown were computed for the minimal models with $J_1^z, J_2^z \rightarrow 0$. (Color is the dynamical spin structure factor, convolved with a $\sigma = 0.025$ energy Gaussian; thin blue lines are underlying spin wave dispersions.) Magnetic umklapp scattering, which usually breaks down spin waves of the Kitaev exchange, was avoided by tuning to the duality with the conventional co-rotating spiral of a J_1-J_2 XY model (panel a). The Klein duality between panels (a) and (c) shifts wave vectors by $\pm\pi/a_1$ for S^a, S^c and by $2\pi/a_1$ for S^b, producing distinctive signatures for the Kitaev exchange; for example, the shifted S^{bb} is evident in the spherical average, via the strong signal at high energy and low momentum.

identified key features in the dynamical structure factor that could be tested via polarized and unpolarized inelastic neutron scattering or resonant inelastic x-ray scattering experiments. Our work helps build towards a full understanding of the lattice-scale model Hamiltonians for these systems, which would shed further light on the unusually similar features across these disparate materials, as well as enable a controlled

identification and understanding of possible proximity to a spin liquid state.

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