Testing topological phase transitions in Kitaev materials under in-plane magnetic fields: Application to α -RuCl₃

Jacob S. Gordon¹ and Hae-Young Kee^{1,2,*}

¹Department of Physics, University of Toronto, Ontario, Canada M5S 1A7

²Canadian Institute for Advanced Research, CIFAR Program in Quantum Materials, Toronto, ON, Canada M5G 1M1

(Received 20 April 2020; revised 7 October 2020; accepted 8 December 2020; published 24 February 2021)

Material realization of the non-Abelian Kitaev spin liquid phase—an example of Ising topological order (ITO)—has been the subject of intense research in recent years. The 4d honeycomb Mott insulator α -RuCl₃ has emerged as a leading candidate, as it enters a field-induced magnetically disordered state where a half-integer quantized thermal Hall conductivity κ_{xy} was reported. Further, a recent report of a sign change in the quantized κ_{xy} across a certain crystallographic direction is strong evidence for a topological phase transition between two ITOs with opposite Chern numbers. Although this is a fascinating result, independent verification remains elusive, and one may ask if there is a thermodynamic quantity sensitive to the phase transition. Here we propose that the magnetotropic coefficient k would serve such a purpose. We report a singular feature in k that indicates a topological phase transition across the $\hat{b}$ -axis where ITO is prohibited by a C_2 symmetry. If the transition in α -RuCl₃ is indeed a direct transition between ITOs, or is a more broad realization of a topological phase transition, then this feature in k should be observable. Implications of C_2 symmetry on other experimental quantities are also discussed.

DOI: [10.1103/PhysRevResearch.3.013179](https://doi.org/10.1103/PhysRevResearch.3.013179)

I. INTRODUCTION

The Kitaev honeycomb model [1] is an important example of an exactly solvable spin- $\frac{1}{2}$ system displaying topological order. Spins in the Kitaev model fractionalize into itinerant Majorana fermions (MFs) on a static $\mathbb{Z}_2$ gauge field, which have a Dirac dispersion when the bond-dependent coupling constants are nearly isotropic. A perturbative magnetic field retains the exact solvability of the model, and serves to gap out the MFs and impart them with a nontrivial Chern number. The resulting gapped phase exhibits *Ising topological order* (ITO), which hosts non-Abelian anyon excitations and a chiral edge MF that manifests in a half-integer quantized thermal Hall conductivity κ_{xy} .

Remarkable properties of this model have incited a flurry of research into its material realization. One mechanism to generate the bond-dependent Kitaev interactions between $j_{\text{eff}} = \frac{1}{2}$ states was proposed by Jackeli and Khaliullin [2], and it found relevance in honeycomb d^5 Mott insulators with strong spin-orbit coupling. Later, Rau *et al.* derived the generic nearest-neighbor interactions in such a system, and found additional off-diagonal symmetric interactions dubbed Γ and Γ' [3,4]. In parallel, promising material candidates such as the iridates A₂IrO₃ [2,5–7] ($A = \text{Li, Na}$) and α -RuCl₃ [8–12]

were identified and shown to exhibit magnetic ordering at low temperatures [13–20]. It was found that an applied in-plane magnetic field can destroy the underlying magnetic order [17], and shortly thereafter excitement was built around the prospect of an intermediate spin-liquid phase [21,21–26].

Most recently, a half-integer quantized thermal Hall conductivity was reported by Kasahara *et al.* [27] in α -RuCl₃ under a small range of magnetic fields (~ 7 –10 T) where the magnetic order is suppressed—a smoking-gun signature of a chiral edge MF associated with the bulk ITO. A follow-up study [28] reported a varied sign structure in the quantized κ_{xy} when the field is rotated in the honeycomb plane, suggesting a topological phase transition between ITOs with opposite Chern number. While this is an exciting development, the thermal transport measurements have been notoriously hard to reproduce, and other interesting probes [29] may be out of reach for some time. In addition, the fact that $\kappa_{xy} \simeq 0$ for fields along the $\hat{b}$ -axis does not necessarily implicate ITO, as it is enforced by a twofold symmetry. Furthermore, a growing κ_{xy} with temperature could be interpreted as thermally excited magnons in a band with finite Berry curvature [30], which, in the polarized phase, must also vanish along the $\hat{b}$ -axis. One may wonder, then, if there is a thermodynamic quantity that is sensitive to such topological phase transitions (TPTs) wherein the Chern number changes. Here we propose that the magnetotropic coefficient k [31,32] as a function of the in-plane magnetic field orientation would serve this purpose. If the transition in α -RuCl₃ between two different Chern number phases, including ITOs, is indeed present in a range of in-plane magnetic field strengths, then a singular feature in k should be observed as the field angle φ sweeps across the $\hat{b}$ axis as shown in Fig. 1(a). We also discuss transitions between

*hykee@physics.utoronto.ca

Published by the American Physical Society under the terms of the Creative Commons Attribution 4.0 International license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

FIG. 1. (a) In-plane magnetic field angle ϕ with respect to the honeycomb $\hat{a}$ - and $\hat{b}$ -axis, with the bond types indicated. (b) The $\hat{c}^*$ component of the torque τ_{c^*} and (c) magnetotropic coefficient k as a function of ϕ at different field strengths in the isotropic Kitaev model—a particular realization of ITO. The inset in (c) contains $\partial_\phi k$, which displays a jump discontinuity. While the torque is not clearly singular, the magnetotropic coefficient k is sensitive to the transition and develops a cusp across the C_2 -preserving $\hat{b}$ -axis. See the main text for details.

nontrivial topological magnon phases at high fields across the $\hat{b}$ axis, which lack such a feature.

Other physical quantities should also reflect the fact that ITO is forbidden by a C_2 symmetry, i.e., the intermediate phase (if any) when the in-plane field is aligned with a C_2 direction will have different properties from those with a generic field direction. This statement is valid so long as the reported intermediate phase is actually a realization of ITO, independent of the microscopic realization.

II. SYMMETRY CONSIDERATIONS

A. Ising topological order

A partial characterization of ITO is through its anyonic excitations; one is a complex fermion ψ and the other is the Ising anyon σ . These excitations obey the following fusion rules:

$$\psi \otimes \psi = \mathbb{1}, \quad \sigma \otimes \sigma = \mathbb{1} \oplus \psi, \quad \psi \otimes \sigma = \sigma, \quad (1)$$

as well as certain associative and braiding relations. In particular, σ is the non-Abelian anyon as their fusion has more than one outcome. ITO can emerge in a variety of systems, including the Kitaev honeycomb model when a small magnetic field is applied to the gapless phase. The conical dispersion of the MFs is gapped by the applied field, and they acquire a nonzero Chern number ν that governs the edge thermal transport. If ν is odd, then the excitations of the $\mathbb{Z}_2$ gauge field carry an unpaired MF that is responsible for their non-Abelian statistics and ultimate realization of ITO [1]. No unpaired MF accompanies the gauge field excitations for even ν , and the resulting phase is an Abelian topological order.

Symmetry places important constraints on ITO, which can be easily understood through a precise analogy. ITO can be

TABLE I. An external in-plane magnetic field $\hat{h} = \cos(\phi)\hat{a} + \sin(\phi)\hat{b}$ breaks TRS Θ , but it maintains a C_2 symmetry for special directions. D_{3d} symmetry is assumed so that there are three distinct C_2 operations. The $\checkmark/\times$ in the first two columns refer to the presence/absence of the indicated symmetry, and likewise in the last column for the presence/absence of ITO.

ϕ	Θ	C_2	ITO
$\frac{\pi}{6}(2n+1)$	$\times$	$\checkmark$	$\times$
else	$\times$	$\times$	$\checkmark$

regarded as a spinless $p_x + ip_y$ superconductor after gauging the $\mathbb{Z}_2$ fermionic parity of the Bogoliubov quasiparticles. Time-reversal symmetry (TRS, Θ) is clearly broken, and under Θ the gap function maps to $p_x - ip_y$ and the chirality of the MF edge mode is reversed. If the system possesses a C_2 symmetry, say, about $\hat{y}$, then the gap function maps to $-p_x + ip_y$ and again the chirality of the edge mode is reversed. When both TRS and such a twofold symmetry are present, ITO is forbidden unless the latter is spontaneously broken [33]. This may occur if TRS is broken by an external magnetic field whose direction is chosen such that the twofold symmetry is maintained. If this field is swept across the symmetric direction, then, barring any spontaneous symmetry breaking, there are two possibilities. One is a direct transition between ITOs with opposite Chern numbers (i.e., time-reversal partners), accompanied by a sign change in κ_{xy} . Another is an intermediate phase surrounding the symmetric direction with an even Chern number. In either case, there must be at least one phase transition around these special directions, and they are therefore prime locations to seek them out.

α -RuCl₃ and the iridate candidates consist of quasi-two-dimensional (quasi-2D) honeycomb layers, with a likely space-group symmetry of $C2/m$ [13,16,18,19,34,35]. We restrict our attention to a single honeycomb layer, which has D_{3d} symmetry in the ideal case and is reduced to a C_2 subgroup with anisotropy. Without loss of generality, we will consider the $C_2(\hat{b})$ rotation common to both groups about the crystallographic $\hat{b}$ direction $[\bar{1}10]$. Under $C_2(\hat{b}) = e^{-i\pi\hat{b}\cdot\mathbf{S}}$ the pseudospin- $\frac{1}{2}$ operators transform as

$$\begin{aligned} C_2(\hat{b})S_j^xC_2^\dagger(\hat{b}) &= -S_{C_2(\hat{b})j}^y, \\ C_2(\hat{b})S_j^yC_2^\dagger(\hat{b}) &= -S_{C_2(\hat{b})j}^x, \\ C_2(\hat{b})S_j^zC_2^\dagger(\hat{b}) &= -S_{C_2(\hat{b})j}^z, \end{aligned} \quad (2)$$

from which we can see that a Zeeman term $-\mathbf{h} \cdot \mathbf{S}$ preserves $C_2(\hat{b})$ for $\hat{h} \parallel \hat{b}$. The ideal honeycomb layer contains two additional C_2 operations related to $C_2(\hat{b})$ through $C_3(\hat{c}^*)$. For fields in the $\hat{a}\hat{b}$ plane shown in Fig. 1(a) and D_{3d} symmetry, a C_2 symmetry is preserved for $\phi = \frac{\pi}{6}(2n+1)$, while it is preserved with C_2 symmetry only for $\phi = \pm\frac{\pi}{2}$ ($\hat{h} = \pm\hat{b}$). A summary of these symmetries can be found in Table I.

Due to the C_2 symmetry about the $\hat{b}$ -axis, there is no ITO when the field is along this direction, and a direct transition between two ITOs with opposite ν is possible as the field sweeps across the $\hat{b}$ -axis. While the sign change of the

quantized κ_{xy} reported in α -RuCl₃ suggests such a transition, one may ask if there are other experimental quantities that may corroborate this finding. Below we show that the magnetotropic coefficient, the second derivative of the free energy with respect to the in-plane angle φ , is a way to independently observe this transition.

III. MAGNETOTROPIC COEFFICIENT

We begin with a brief review of the magnetotropic coefficient k , then we apply this idea to a low-energy theory of the transition between different ITOs, the exactly solvable Kitaev model, and to a linear spin-wave theory description of the polarized phase. A cusp singularity in k across the $\hat{b}$ axis will be demonstrated in the first two cases, as seen in Fig. 1(c) for a specific instance of ITO, and traced to the behavior of the bulk fermion gap.

Familiar thermodynamic quantities such as the magnetization $\mathbf{m}$ and torque $\boldsymbol{\tau}$ can be defined as the first derivative of the free energy $\mathcal{F}$ with respect to the magnetic field strength and orientation, respectively, and they are widely used as indicators of phase transitions. In particular, derivatives of $\mathcal{F}$ with respect to the field strength and angle yield the magnetization along the field direction and the torque perpendicular to the plane of rotation, respectively. However, susceptibilities involving second derivatives of $\mathcal{F}$ can offer more fundamental insights into the nature of phase transitions. In the same spirit as the magnetic susceptibility, the magnetotropic coefficient k is the second derivative of $\mathcal{F}$ with respect to the field angle—or equivalently, the first derivative of the perpendicular component of the torque. Since torque must vanish along the crystallographic axes, the magnetotropic coefficient has a distinct advantage when studying transitions across these directions. Early work in this direction was done using conventional torque magnetometry on Na₂LiO₃ [36] and α -RuCl₃ [37].

Resonant torque magnetometry [31] was developed as a tool to directly measure k through a shift in the natural frequency of a cantilever, and it has allowed for unprecedented accuracy in measurements of magnetic anisotropy. In addition, the small size of the samples used has been seen to mitigate effects due to crystallographic domains [32]. This technique was recently applied to α -RuCl₃ to determine the phase boundary between long-range zigzag order and the intermediate nonmagnetic phase in the $\hat{a}\hat{c}^*$ plane [31,32], and it was studied theoretically in the context of β -Li₂IrO₃ [38] and α -RuCl₃ [39]. While the torque only displays a deviation from the linear-response regime of $\propto h^2$, the magnetotropic coefficient signals this transition with a sharp jump, demonstrating its sensitivity to the phase boundary.

A. Continuum Dirac fermion

We will first consider a simple continuum model of a topological phase transition in which the Chern number changes—a single continuum Dirac fermion whose mass changes sign at the transition point. This, for instance, could describe the emergent ψ fermion associated with ITO. Higher Chern number transitions can be described with several species of Dirac fermion, so this result is not limited to $\nu = \pm 1$. The free energy at $T = 0$ is obtained by integrating

over the negative energy states of $\varepsilon_{\pm}(\mathbf{q}) = \pm\sqrt{(v\mathbf{q})^2 + m^2}$. Restoring the UV cutoff, we find

$$\begin{aligned}\mathcal{F} &\simeq \frac{1}{2} \frac{A}{(2\pi)^2} \int_{vq \leq \Lambda} d\mathbf{q} \varepsilon_{-}(\mathbf{q}) \\ &= \frac{A}{12\pi v^2} [|m|^3 - (\Lambda^2 + m^2)^{3/2}] \sim |m|^3,\end{aligned}$$

where A is the area of the honeycomb unit cell, and we have identified $|m|^3$ as the singular contribution. Close to the angle φ_* where the magnetic field is tuned along a C_2 direction, the mass can be expanded as $m \propto \delta\varphi = \varphi - \varphi_*$ so that $\partial_m \propto \partial_{\delta\varphi}$. Keeping only the singular contributions, we easily find the torque and magnetotropic coefficient

$$\tau_{c^*} \sim m|m|, \quad k \sim |m|, \quad \partial_{\varphi} k \sim \text{sgn}(m). \quad (3)$$

Therefore, a cusp singularity appears in the magnetotropic coefficient as a direct consequence of the bulk fermion gap closing and reopening as a function of the field angle. We can also see that the magnitude of this contribution is more pronounced at larger values of the magnetic field due to the enhanced mass on either side of the transition. Further derivatives only magnify this singularity; for instance, the derivative of k with respect to φ exhibits a jump discontinuity across the C_2 -symmetric direction, reflecting the change in Chern number.

At finite temperatures, the positive part of the band begins to be occupied and roughly contributes $-|m|^3$ to the free energy. This contribution partially cancels the existing $+|m|^3$ part, and it causes a smearing of the sharp cusp. Thus, the regime of low temperatures and high fields is desirable to study such topological phase transitions.

B. Kitaev model in a perturbative magnetic field

We now turn our attention to the exactly solvable Kitaev model with a perturbative magnetic field to demonstrate this physics. The Kitaev model is defined through the bond-dependent Ising interactions $\mathcal{H}_K = \sum_{\langle j,k \rangle_{\mu}} K_{\mu} S_j^{\mu} S_k^{\mu}$, where the nearest-neighbor bond $\langle j,k \rangle_{\mu}$ is of the type $\mu \in \{x, y, z\}$. For simplicity, we will focus on the isotropic limit with $|K_{\mu}| \equiv 1$. We introduce a Zeeman field through $\mathcal{H}_Z = -h \sum_j \hat{\mathbf{h}} \cdot \hat{\mathbf{g}} \cdot \mathbf{S}_j$, where $h = g\mu_B H$ is the effective field strength, and $\hat{\mathbf{g}}$ encodes the g -factor anisotropy. Focusing on in-plane magnetic fields, we can neglect g -factor anisotropy because $g_a = g_b$. To retain the exact solvability of the model, the Zeeman term can be captured perturbatively in the zero-flux sector as

$$\mathcal{H}_Z^{\text{eff}} = -\frac{h_x h_y h_z}{\Delta_F^2} \sum_{(j,k,l)} S_j^x S_k^y S_l^z, \quad (4)$$

where $\Delta_F \simeq 0.065|K|$ is the two-flux gap [1,40], and $\langle\langle j, l \rangle\rangle$ forms a second-nearest-neighbor bond.

The in-plane magnetic field direction is parametrized as $\hat{\mathbf{h}} = \cos(\varphi)\hat{\mathbf{a}} + \sin(\varphi)\hat{\mathbf{b}}$, and the $\hat{\mathbf{c}}^*$ component of the torque and the corresponding magnetotropic coefficient k are obtained by differentiating the free energy $\mathcal{F}$ with respect to φ ,

$$\tau_{c^*} = \boldsymbol{\tau} \cdot \hat{\mathbf{c}}^* = \frac{\partial \mathcal{F}}{\partial \varphi}, \quad k = \frac{\partial \tau_{c^*}}{\partial \varphi} = \frac{\partial^2 \mathcal{F}}{\partial \varphi^2}. \quad (5)$$

FIG. 2. Thermodynamic properties calculated with the Kitaev model in a perturbative magnetic field. (a) Free energy $\mathcal{F}$ displaying maxima along three C_2 -symmetric directions for an ideal honeycomb layer. (b) The $\hat{c}^*$ component of the torque, which changes sign across the crystallographic axes as well as the C_2 -symmetric lines. (c) The magnetotropic coefficient k shows most clearly the transitions between ITOs with opposite Chern number. The midpoints of the ITO phases display a broad, positive peak in k while the ITO transition points are marked by a sharp, negative cusp indicating an instability with respect to a rotation of the field.

The fermionic Hamiltonian is

$$\mathcal{H}_\psi = \sum_q \varepsilon_q \left(g_q^\dagger g_q - \frac{\mathbb{1}}{2} \right),$$

where the dispersion $\varepsilon_q \geq 0$ involves certain well-known functions of $\mathbf{q}$ given in Appendix A. The free energy density at $T = 0$ is therefore given by $\mathcal{F} = -\frac{1}{2} \int_{\mathbb{T}^2} d\mathbf{q} \varepsilon_q$. Results of this calculation are shown in Figs. 1(b), 1(c) and 2 as a function of φ for different values of h assuming an ideal honeycomb layer.

The free energy displays deep minima at the center of the ITO phases, and it peaks along the C_2 -symmetric directions where the MF mass term vanishes. Inside the ITO, the MFs acquire a gap $\propto h^3$ and a nonzero Chern number $\nu = \text{sgn}(h_x h_y h_z) = \pm 1$ for a generic field as shown by the coloring in Fig. 3(b). Along the $\hat{b}$ axis ($[\bar{1}10]$) of C_2 symmetry (and equivalent directions under C_3) the gap vanishes, consistent with the symmetry constraints on ITO. Due to these extremal points, the $\hat{c}^*$ component of the torque vanishes along these directions, as well as along the crystallographic axes. As we can see from the contour lines in Fig. 2(b) and the slices in Fig. 1(c), τ_{c^*} changes much more rapidly around the C_2 -symmetric directions, resulting in a sawtoothlike profile. Of the three quantities in Fig. 2, the magnetotropic coefficient k signals the ITO transition lines most clearly. Midpoints of the ITO phases are marked by a broad, positive maxima in k while the transition points show a sharp, negative cusp. We can expand the Kitaev Hamiltonian about the $\mathbf{K}$ point of the Brillouin zone to obtain an effective Dirac Hamiltonian whose mass $m \propto -h^3 \cos(3\varphi)$ changes across the C_2 -symmetric directions. Therefore, the singular feature in k is directly linked to the sign change of the Dirac mass term.

Although we can only pinpoint the location of the transition points in the $\hat{a}\hat{b}$ plane, the transition lines will generically extend into the $\hat{c}^*$ direction, where they may merge or cross.

The precise angular phase diagram depends on the specific model parameters—for instance, the Kitaev model boundaries are shown in Fig. 3(b). However, even if there is out-of-plane sample misalignment, the ITO transition will still be visible so long as the angular path sweeps across the $\hat{b}$ -axis. Angular cuts away from the $\hat{a}\hat{b}$ plane are not guaranteed to cross ITO transitions, but if they do then a singular cusp in k should be observable. In fact, since the critical polarizing field grows rapidly as the direction is tuned toward $\hat{c}^*$ in α -RuCl₃, the mass gap will be enhanced along with the cusp singularity. For the same reason, a sweep that crosses the phase boundary at an angle will effectively have a smaller mass on either side of the

FIG. 3. (a) Orientation of $\hat{x}, \hat{y}, \hat{z}$ spin axes with respect to the crystallographic $\hat{a}, \hat{b}, \hat{c}^*$ axes. The spin axes all make the same angle with the out-of-plane $\hat{c}^*$ axis. (b) Angular phase diagram for the Kitaev model in a perturbative magnetic field. Red and blue indicate $\nu = \text{sgn}(h_x h_y h_z) = \pm 1$, respectively. The green line indicates the phase boundary ℓ_B (given in the main text) that intersects the $\hat{b}$ axis at an angle of $\sim 54.74^\circ$ to the $\hat{a}\hat{b}$ plane. An out-of-plane line $\ell_\perp$ passing through the $\hat{b}$ axis is shown in purple, which intersects the phase boundary ℓ_B at a right angle. The cusp in the magnetotropic coefficient k will be maximal along $\ell_\perp$ and reduced along $\ell_{\hat{a}\hat{b}}$.

FIG. 4. (a) Path along which the linear spin-wave spectrum is plotted in (b) is indicated by the purple line in the Brillouin zone. (b) Spin-wave spectrum for field angles sweeping across the $\hat{b}$ axis at $h/S|K| = 0.75$. The parameters used are $K = -1.00$, $J = -0.10|K|$, $\Gamma = 0.50|K|$, $\Gamma' = -0.05|K|$, and they are taken to be isotropic across the bond types so that the classical contribution to the energy is constant. When the field is along the $\hat{b}$ axis, we see a band crossing near the $K_{1,2}$ points driven by the field angle dependence. (c) Torque calculated from the angular dependence of the zero-point magnon energy in the polarized phase for a few values of the magnetic field, and (d) corresponding magnetotropic coefficient. At high fields we recover a $\cos(2\varphi)$ dependence of the free energy, leading to a $\sin(2\varphi)$ dependence of the torque and a $\cos(2\varphi)$ dependence of the magnetotropic coefficient.

transition, which diminishes the singularity. As an example, in the Kitaev model with a perturbative magnetic field, the phase boundary that intersects the $\hat{b}$ -axis lies in the $\hat{x}\hat{y}$ plane and is parametrized by

$$\ell_B(\phi) = \cos(\phi) \left(\frac{-\hat{x} + \hat{y}}{\sqrt{2}} \right) + \sin(\phi) \left(\frac{\hat{x} + \hat{y}}{\sqrt{2}} \right),$$

as shown by the green line in Fig. 3(b). With a parametrization of the $\hat{a}\hat{b}$ plane $\ell_{\hat{a}\hat{b}}(\phi) = \cos(\phi)\hat{b} + \sin(\phi)\hat{a}$, one can show that this intersects the phase boundary at an angle of $\sim 54.74^\circ$. If instead one crosses the $\hat{b}$ axis along the line

$$\ell_{\perp}(\phi) = \cos(\phi) \left(\frac{-\hat{x} + \hat{y}}{\sqrt{2}} \right) + \sin(\phi)\hat{z},$$

as shown by the purple line in Fig. 3(b), the phase boundary is intersected at a right angle. It is for this line that the cusp in k is maximal. This dependence on the angular path, i.e., the two independent angular directions on the sphere, provides additional information about the phase boundary.

C. Magnons in the polarized phase

To compare with the ITO transitions, we consider the magnetotropic coefficient deep into the field-polarized phase by using linear spin-wave theory. In Kitaev magnets, the magnons in the high-field phase are generically topological [30,41]. By performing the calculation for a field direction $\hat{h} = \cos(\varphi)\hat{a} + \sin(\varphi)\hat{b}$, we can use the resulting dependence on φ to calculate the torque and magnetotropic coefficient. Details of the calculation are given in Appendix B. Within linear spin-wave theory, there are two contributions to the ground-state energy at $T = 0$ —one is the classical energy of

the spins, and the other is the quantum zero-point energy of the magnon modes. Adopting the generic nearest-neighbor spin model for α -RuCl₃, involving J, K, Γ, Γ' , we impose the $C2/m$ symmetry by setting $J_z = J$, $J_x = J_y = J + \delta J$, and the same for the other exchange parameters. The classical contribution to the torque and magnetotropic coefficient is found to be

$$\begin{aligned} \hat{c}^* \cdot \tau_{cl} &= \frac{1}{2}[\delta K + 2\delta\Gamma - 2\delta\Gamma'] \sin(2\varphi), \\ k_{cl} &= [\delta K + 2\delta\Gamma - 2\delta\Gamma'] \cos(2\varphi), \end{aligned} \quad (6)$$

whose amplitude only depends on a certain combination of the non-Heisenberg exchange anisotropy.

After a bosonic Bogoliubov transformation, the linear spin-wave Hamiltonian can be brought into canonical form,

$$\mathcal{H}_{\text{LSWT}} = S \sum_{q,\alpha} \omega_{q,\alpha} \left(\gamma_{q,\alpha}^\dagger \gamma_{q,\alpha} + \frac{1}{2} \right),$$

where we recognize the quantum zero-point energy density of the magnons as $\frac{S}{2} \sum_{\alpha} \int_{\mathbb{T}^2} d\mathbf{q} \omega_{q,\alpha}$. Let us consider a situation illustrated in Fig. 4(b) for a reasonable set of the exchange constants appropriate for α -RuCl₃. For a field along the $\hat{b}$ direction, there are Dirac-like band crossings in the vicinity of the $K_{1,2}$ points of the Brillouin zone. As the field is tuned away from the $\hat{b}$ direction, these Dirac points acquire a mass and the system is driven into a topological magnon phase with opposite Chern numbers on either side of the transition (enforced by the $C_2(\hat{b})$ symmetry).

There are a few differences from the fermionic case that deserve discussion. First, since we are at high fields away from the phase boundary, the lower magnon branch is gapped: $\omega_{q,-} > 0$. Second, the dispersion (and the magnitude of the

Dirac mass) is set by the exchange constants and the field angle, rather than the field strength, so it does not grow with h . Despite the fact that the mass of the Dirac points changes across the $\hat{b}$ axis, there is no associated cusp singularity in k , shown in Fig. 4(d). This fact can be understood easily by adopting a continuum model around the Dirac point $\omega_{q,\pm} = \omega_0 \pm \sqrt{(vq)^2 + m^2}$, which only contributes a constant to the free energy,

$$\mathcal{F} \simeq \frac{S}{2} \frac{A}{(2\pi)^2} \sum_{\pm} \int_{vq \leq \Lambda} d\mathbf{q} \, \omega_{q,\pm} = \frac{AS}{4\pi v^2} \omega_0 \Lambda^2.$$

One can see that this is due to cancellation between the positive and negative branches of the Dirac dispersion, much like the fermionic case at elevated temperatures. Thus, the topological phase transition associated with fermions (including ITO) displays a singular feature in k at low temperatures and high fields, a signature that differs from that of topological magnons at high fields.

IV. SUMMARY AND OPEN ISSUES

The observed sign change in κ_{xy} across the $\hat{b}$ -axis by Yokoi *et al.* [28] in α -RuCl₃ suggests a direct transition between ITOs with opposite Chern numbers. This is an important development, but we are currently lacking evidence of the phase transition through any thermodynamic quantities in the intermediate phase. Here we propose the magnetotropic coefficient k as a *sensitive* probe of a direct transition between ITOs, which we expect to appear under in-plane magnetic fields in α -RuCl₃ between the zigzag and polarized phases. Based on a continuum theory of the low-energy ITO degrees of freedom, we have demonstrated a singular cusp in k (i.e., a discontinuous jump in its derivative) at a direct transition between ITOs with opposite Chern numbers. The origin of the cusp was shown to be directly linked to the closing of the bulk MF mass gap, and its enhancement tied to the magnitude of this gap. Using a prototypical example of ITO (Kitaev's model), we have illustrated this physics. We have also studied the magnetotropic coefficient in the higher-field polarized phase with topological magnons using linear spin-wave theory. Despite a topological phase transition across the $\hat{b}$ axis, we see no such singular cusp in k which we traced back to a cancellation between the two branches of the magnon dispersion. Thus, the fermionic statistics are important to be able to see this nonanalytic behavior.

Remarkably, extensive measurements of k in α -RuCl₃ have been amassed by Modic *et al.* [32] for a wide range of magnetic field strengths and orientations. While the existing in-plane magnetotropic coefficient data in the intermediate phase are suggestive, a more detailed study around the $\hat{b}$ axis is necessary to test our proposal. In particular, this will require a careful analysis with respect to the field strengths and temperatures. The mass term is enhanced at larger field strengths, as is the magnitude of the singularity. Nonzero temperatures (but smaller than the flux gap) will smear this singularity, and will therefore become sharper as the temperature is lowered. A comparison with the high- and low-field magnetotropic coefficients can be used to distinguish the ITO transitions from features related to the magnetic order. As we mentioned

earlier, the torque is required to vanish along the crystallographic directions. This presents a practical challenge, as a small signal is more susceptible to noise. We therefore advocate for the use of resonant torque magnetometry [31] to directly measure the magnetotropic coefficient.

The limitations of our result in relation to a microscopic model deserve some discussion. Our result is limited to a direct transition between ITOs with different ν . The Kitaev model displays such transitions, so if a Kitaev-dominant model is appropriate for α -RuCl₃, then a sign change of κ_{xy} and our proposed singularity in k across $\hat{b}$ should be observed. However, these experimental results do not necessarily imply that the underlying spin model is Kitaev-dominant. In other words, they are necessary but not sufficient; they are consequences of the Kitaev model, but they do not rule out the possibility of other microscopic models with ITO. Note that ITO is prohibited for fields along the $\hat{b}$ -axis for any generic model preserving the C_2 symmetry. Thus the experimental signatures discussed here are present if ITO exists in a generic model. While there is no microscopic model that leads to a Kitaev spin liquid under intermediate *in-plane* magnetic fields, if such a route exists the model would inevitably exhibit a transition—either between two ITOs or to an intermediate phase, unless the C_2 symmetry is spontaneously broken. In the absence of a microscopic understanding of the putative intermediate ITO, one cannot rule out the possibility of other significant interactions leading to an alternative mechanism, and such a microscopic model remains to be found. It is our hope that a comprehensive mapping of the intermediate phase boundaries with respect to the field angle can shed light on the puzzle of α -RuCl₃.

Note added. After the completion of this work, a numerical study [42] showing that the Γ exchange tends to push the ITO boundaries toward the $\hat{a}\hat{b}$ -plane, and measurements of anisotropic specific heat of α -RuCl₃ under in-plane fields [43], were reported.

ACKNOWLEDGMENTS

We would like to thank S. R. Julian and Y.-J. Kim for useful discussion. This work was supported by NSERC Discovery Grant No. 06089-2016, CIFAR, and the Canada Research Chairs Program.

APPENDIX A: EXACT SOLUTION OF THE KITAEV MODEL

In this Appendix, we review the well-known solution [1] of the Kitaev model,

$$\mathcal{H}_K = \sum_{(j,k)_\mu} K_\mu S_j^\mu S_k^\mu, \quad (\text{A1})$$

and we establish the equations referenced in the main text. Introducing four Majorana fermions γ_j^μ per site satisfying $\{\gamma_j^\mu, \gamma_k^\nu\} = 2\delta_{jk}\delta^{\mu\nu}\mathbb{1}$, the spin operators can be faithfully represented as

$$S_j^\mu = \frac{i}{2} \gamma_j^0 \gamma_j^\mu, \quad (\text{A2})$$

by restricting ourselves to the physical subspace defined through $\mathcal{D}_j \equiv 1 \forall j$ with $\mathcal{D}_j = \frac{1}{i} \sigma_j^x \sigma_j^y \sigma_j^z = \gamma_j^0 \gamma_j^x \gamma_j^y \gamma_j^z$. In this language, the Hamiltonian takes the form

$$\mathcal{H}_K = \frac{i}{4} \sum_{(j,k)_\mu} \gamma_j^0 (K_\mu \hat{u}_{jk}^\mu) \gamma_k^0, \quad (\text{A3})$$

with the bond operators $\hat{u}_{jk}^\mu = -i \gamma_j^\mu \gamma_k^\mu$ that (i) commute with each other, (ii) commute with the Hamiltonian, and (iii) $(\hat{u}_{jk})^2 = \mathbb{1}$, furnishing an extensive set of conserved quantities. These bond variables serve as an emergent static $\mathbb{Z}_2$ gauge field coupled to the γ_j^0 fermions, and they are related to the conserved plaquette operators in the original spin model via

$$W_p = \sigma_j^x \sigma_k^y \sigma_l^z \sigma_m^x \sigma_n^y \sigma_o^z = \prod_{(j,k) \in p} \hat{u}_{jk}. \quad (\text{A4})$$

The ground state lies in the flux-free sector with $W_p \equiv +1 \forall p$ realized with the gauge $\hat{u}_{jk}^\mu \equiv +1$ on all bonds—thus reducing the Hamiltonian to a quadratic form in the γ_j^0 fermions,

$$\mathcal{H}_K = \frac{i}{4} \sum_{(j,k)_\mu} \gamma_{j,A}^0 (K_\mu) \gamma_{k,B}^0. \quad (\text{A5})$$

Here we take $\hat{u}_{j,k}^\mu = +1$ with the convention that j belongs to sublattice A and k belongs to sublattice B . Defining the column vector $\boldsymbol{\gamma}_j = (\gamma_{j,A}^0, \gamma_{j,B}^0)^T$, we exploit translational invariance via the Fourier transform

$$\boldsymbol{\gamma}_j = \frac{1}{\sqrt{N_c}} \sum_{\mathbf{q}} e^{i\mathbf{q} \cdot \mathbf{x}_j} \boldsymbol{\gamma}_{\mathbf{q}}, \quad \boldsymbol{\gamma}_{\mathbf{q}} = \frac{1}{\sqrt{N_c}} \sum_j e^{-i\mathbf{q} \cdot \mathbf{x}_j} \boldsymbol{\gamma}_j, \quad (\text{A6})$$

where N_c is the number of unit cells. The anticommutation relations $\{\boldsymbol{\gamma}_j, \boldsymbol{\gamma}_k\} = 2\delta_{j,k} \mathbb{1}$ become $\{\boldsymbol{\gamma}_{\mathbf{q}}, \boldsymbol{\gamma}_{\mathbf{p}}^\dagger\} = 2\delta_{\mathbf{q},\mathbf{p}} \mathbb{1}$. Note that $\boldsymbol{\gamma}_{\mathbf{q}}^\dagger = \boldsymbol{\gamma}_{-\mathbf{q}}^T$ due to the reality of the $\boldsymbol{\gamma}_j$ fermions, so the degrees of freedom at $\pm \mathbf{q}$ are not independent. In momentum space, the Hamiltonian in the flux-free sector becomes

$$\mathcal{H}_K = \frac{1}{2} \sum_{\mathbf{q}} \boldsymbol{\gamma}_{\mathbf{q}}^\dagger \mathcal{H}_{\mathbf{q}} \boldsymbol{\gamma}_{\mathbf{q}}, \quad (\text{A7})$$

with

$$\mathcal{H}_{\mathbf{q}} = \begin{pmatrix} 0 & \alpha_{\mathbf{q}} \\ \alpha_{\mathbf{q}}^* & 0 \end{pmatrix}, \quad (\text{A8})$$

$$\alpha_{\mathbf{q}} = \frac{i}{4} (K_x e^{2\pi i q_1} + K_y e^{2\pi i q_2} + K_z),$$

expressed in the reciprocal-lattice basis $\mathbf{q} = q_1 \mathbf{b}_1 + q_2 \mathbf{b}_2 = (q_1, q_2)$ for $q_j \in [0, 1)$.

The effect of a Zeeman field in the zero-flux sector can be incorporated at third order in perturbation theory via

$$\mathcal{H}_Z^{\text{eff}} = -\frac{h_x h_y h_z}{\Delta_F^2} \sum_{(j,k,l)} S_j^x S_k^y S_l^z. \quad (\text{A9})$$

In terms of the Majorana fermions, the three-spin interaction can be written as

$$S_j^x S_k^y S_l^z = \frac{i}{2^3} \hat{u}_{jk}^\mu \mathcal{D}_k \hat{u}_{lk}^\nu \gamma_j^0 \gamma_l^0 = \pm \frac{i}{2^3} \gamma_j^0 \gamma_l^0, \quad (\text{A10})$$

which contributes a second nearest neighbor akin to the Haldane term. Hopping along the arrow direction in the above figure contributes $+i\kappa \gamma_j^0 \gamma_k^0$ (with opposite sign going against this direction), where the amplitude is

$$\kappa = \frac{h_x h_y h_z}{2\Delta_F^2}. \quad (\text{A11})$$

The Hamiltonian then acquires a sublattice-dependent hopping term

$$\mathcal{H}_{\mathbf{q}} = \begin{pmatrix} \beta_{\mathbf{q}} & \alpha_{\mathbf{q}} \\ \alpha_{\mathbf{q}}^* & -\beta_{\mathbf{q}} \end{pmatrix}, \quad (\text{A12})$$

with

$$\beta_{\mathbf{q}} = \frac{1}{2} \kappa \{ \sin[2\pi q_1] - \sin[2\pi q_2] - \sin[2\pi(q_1 - q_2)] \}.$$

To obtain the ground-state energy, we now combine the Majorana fermions into a spinless complex fermion $f_{\mathbf{q}} = \frac{1}{2}(\gamma_{\mathbf{q},A}^0 + i\gamma_{\mathbf{q},B}^0)$, $f_{-\mathbf{q}}^\dagger = \frac{1}{2}(\gamma_{\mathbf{q},A}^0 - i\gamma_{\mathbf{q},B}^0)$ satisfying $\{f_{\mathbf{q}}, f_{\mathbf{p}}^\dagger\} = \delta_{\mathbf{q},\mathbf{p}} \mathbb{1}$ and $\{f_{\mathbf{q}}, f_{\mathbf{p}}\} = 0$. With the Nambu spinor,

$$\boldsymbol{\psi}_{\mathbf{q}} = \begin{pmatrix} f_{\mathbf{q}} \\ f_{-\mathbf{q}}^\dagger \end{pmatrix} = \frac{1}{2} \begin{pmatrix} 1 & +i \\ 1 & -i \end{pmatrix} \begin{pmatrix} \gamma_{\mathbf{q},A}^0 \\ \gamma_{\mathbf{q},B}^0 \end{pmatrix} = \frac{1}{2} \begin{pmatrix} 1 & +i \\ 1 & -i \end{pmatrix} \boldsymbol{\gamma}_{\mathbf{q}}, \quad (\text{A13})$$

the Hamiltonian is transformed into a Bogoliubov–de Gennes form,

$$\begin{aligned} \mathcal{H} &= \frac{1}{2} \sum_{\mathbf{q}} \begin{pmatrix} \gamma_{-\mathbf{q},A}^0 & \gamma_{-\mathbf{q},B}^0 \end{pmatrix} \begin{pmatrix} \beta_{\mathbf{q}} & \alpha_{\mathbf{q}} \\ \alpha_{\mathbf{q}}^* & -\beta_{\mathbf{q}} \end{pmatrix} \begin{pmatrix} \gamma_{\mathbf{q},A}^0 \\ \gamma_{\mathbf{q},B}^0 \end{pmatrix} \\ &= \frac{1}{2} \sum_{\mathbf{q}} \begin{pmatrix} f_{\mathbf{q}}^\dagger & f_{-\mathbf{q}} \end{pmatrix} \begin{pmatrix} h_{\mathbf{q}} & \Delta_{\mathbf{q}} \\ \Delta_{\mathbf{q}}^* & -h_{-\mathbf{q}} \end{pmatrix} \begin{pmatrix} f_{\mathbf{q}} \\ f_{-\mathbf{q}}^\dagger \end{pmatrix} \\ &= \sum_{\mathbf{q}} f_{\mathbf{q}}^\dagger h_{\mathbf{q}} f_{\mathbf{q}} + \frac{1}{2} \sum_{\mathbf{q}} (f_{\mathbf{q}}^\dagger \Delta_{\mathbf{q}} f_{-\mathbf{q}}^\dagger + f_{-\mathbf{q}} \Delta_{\mathbf{q}}^* f_{\mathbf{q}}), \end{aligned}$$

where $h_{\mathbf{q}} = i(\alpha_{\mathbf{q}}^* - \alpha_{\mathbf{q}}) = h_{-\mathbf{q}}$ and $\Delta_{\mathbf{q}} = 2\beta_{\mathbf{q}} + i(\alpha_{\mathbf{q}}^* + \alpha_{\mathbf{q}}) = -\Delta_{-\mathbf{q}}$. After a Bogoliubov transformation, we bring the Hamiltonian into canonical form,

$$\begin{aligned} \mathcal{H} &= \frac{1}{2} \sum_{\mathbf{q}} \begin{pmatrix} g_{\mathbf{q}}^\dagger & g_{-\mathbf{q}} \end{pmatrix} \begin{pmatrix} \varepsilon_{\mathbf{q}} & 0 \\ 0 & -\varepsilon_{-\mathbf{q}} \end{pmatrix} \begin{pmatrix} g_{\mathbf{q}} \\ g_{-\mathbf{q}}^\dagger \end{pmatrix} \\ &= \sum_{\mathbf{q}} \varepsilon_{\mathbf{q}} \left(g_{\mathbf{q}}^\dagger g_{\mathbf{q}} - \frac{\mathbb{1}}{2} \right), \end{aligned}$$

with $\varepsilon_q = \sqrt{|h_q|^2 + |\Delta_q|^2}$, and the ground-state energy per unit cell,

$$\begin{aligned} \frac{E_0}{N} &= -\frac{1}{2N} \sum_q \varepsilon_q \\ &\rightarrow -\frac{1}{2} \int_{\mathbb{T}^2} d\mathbf{q} \varepsilon_q \simeq -0.393\,649\,309\,6|K|. \end{aligned} \quad (\text{A14})$$

The numerical value is quoted in the isotropic limit at zero field in the thermodynamic limit. Therefore, the free energy at $T = 0$ is $\mathcal{F} = -\frac{1}{2} \int_{\mathbb{T}^2} d\mathbf{q} \varepsilon_q$, which we differentiate to calculate thermodynamic quantities.

We now consider $K_x = K_y \leq K_z$, consistent with $C2/m$ symmetry, but sufficiently close to the isotropic limit ($K_x \geq \frac{1}{2}K_z$) so that the Majorana spectrum remains gapless for $h = 0$. The position of the nodes at $h = 0$ is easily found to be at

$$\mathbf{Q}_\pm = \pm(1 - q_*, q_*), \quad q_* = \frac{1}{2\pi} \arccos\left(-\frac{1}{2} \frac{K_z}{K_x}\right), \quad (\text{A15})$$

written in the reciprocal-lattice basis. In the isotropic limit we have $q_* = \frac{1}{3}$, so that $\mathbf{Q}_\pm$ reduce to the $\mathbf{K}_{1,2}$ points of the Brillouin zone, respectively. Away from this limit, the nodal points move symmetrically toward the $\mathbf{M}_3$ point until the critical anisotropy $K_x = \frac{1}{2}K_z$ beyond which the fermionic spectrum is gapped.

Expanding around the $\mathbf{Q}_\pm$ points, we can obtain an effective continuum Dirac theory,

$$\mathcal{H}_\pm(\mathbf{q}) \simeq \mathcal{H}_{\mathbf{q}+\mathbf{Q}_\pm} - \mathcal{H}_{\mathbf{Q}_\pm} \simeq v_F[\mp\eta q_x \tau_y + q_y \tau_x] \pm m\tau_z, \quad (\text{A16})$$

valid up to some UV cutoff Λ , with

$$\begin{aligned} v_F &= \frac{\sqrt{3}}{8} K_z, \quad \eta = \left(\frac{4r^2 - 1}{3}\right)^{\frac{1}{2}}, \quad r = \frac{K_x}{K_z}, \\ m &= \left(-\frac{3\sqrt{3}}{4}\kappa\right) \left(\frac{2r+1}{3r^2}\right) \left(\frac{4r^2-1}{3}\right)^{\frac{1}{2}}. \end{aligned}$$

The functions of $r = K_x/K_z$ capture the anisotropic couplings and are chosen such that they are unity in the isotropic limit. Note that we have neglected the linear terms in the expansion of β_q which also modify the velocity in the q_x direction. Anisotropic velocities do not affect our main result, so we opt to work in the isotropic limit for simplicity. The mass $m \sim \kappa \sim h_x h_y h_z \sim \cos(3\varphi)$ is easily seen to vanish for fields along the C_2 -symmetric directions with in-plane fields of the form $\hat{\mathbf{h}} = \cos(\varphi)\hat{\mathbf{a}} + \sin(\varphi)\hat{\mathbf{b}}$.

APPENDIX B: LINEAR SPIN-WAVE THEORY AT HIGH MAGNETIC FIELDS

To analyze the magnetotropic coefficient at high magnetic fields, we perform a linear spin-wave expansion about the polarized state. We first establish the formalism, largely following the notation of [44]. A general translationally invariant spin exchange Hamiltonian can be defined through the matri-

cies $\mathbf{J}_{\delta,\alpha,\beta}$, along with a Zeeman field $\mathbf{h}$,

$$\mathcal{H} = \frac{1}{2} \sum_{\mathbf{x},\delta} \sum_{\alpha,\beta} \mathbf{S}_{\mathbf{x}+\delta,\alpha} \cdot \mathbf{J}_{\delta,\alpha,\beta} \cdot \mathbf{S}_{\mathbf{x},\beta} - h \sum_{\mathbf{x},\alpha} \hat{\mathbf{h}} \cdot \mathbf{S}_{\mathbf{x},\alpha}, \quad (\text{B1})$$

where $\mathbf{x}$ labels the N_c magnetic unit cells, α, β label the N_s magnetic sublattices, and δ labels the bond direction. The total number of spinful sites is $N = N_c N_s$. We can characterize the classical magnetic order by the direction $\hat{\mathbf{z}}_\alpha$ on sublattice α , which we write as part of a local frame $\{\hat{\mathbf{x}}_\alpha, \hat{\mathbf{y}}_\alpha, \hat{\mathbf{z}}_\alpha\}$. It is convenient to use $\{\hat{\mathbf{e}}_{0,\alpha}, \hat{\mathbf{e}}_{+,\alpha}, \hat{\mathbf{e}}_{-,\alpha}\}$ defined by

$$\hat{\mathbf{e}}_{0,\alpha} = \hat{\mathbf{z}}_\alpha, \quad \hat{\mathbf{e}}_{\pm,\alpha} = \frac{1}{\sqrt{2}}(\hat{\mathbf{x}}_\alpha \pm i\hat{\mathbf{y}}_\alpha),$$

so that the spin operator becomes

$$\begin{aligned} \mathbf{S}_{\mathbf{x},\alpha} &= S_{\mathbf{x},\alpha}^x \hat{\mathbf{x}}_\alpha + S_{\mathbf{x},\alpha}^y \hat{\mathbf{y}}_\alpha + S_{\mathbf{x},\alpha}^z \hat{\mathbf{z}}_\alpha \\ &= \frac{1}{\sqrt{2}}(S_{\mathbf{x},\alpha}^- \hat{\mathbf{e}}_{+,\alpha} + S_{\mathbf{x},\alpha}^+ \hat{\mathbf{e}}_{-,\alpha}) + S_{\mathbf{x},\alpha}^z \hat{\mathbf{e}}_{0,\alpha}. \end{aligned}$$

We will also rewrite the exchange matrices $\mathbf{J}_{\delta,\alpha,\beta}$ and the Zeeman field $\hat{\mathbf{h}}$ in the local frame as

$$\mathcal{J}_{\delta,\alpha,\beta}^{\mu,\nu} = \hat{\mathbf{e}}_{\mu,\alpha} \cdot \mathbf{J}_{\delta,\alpha,\beta} \cdot \hat{\mathbf{e}}_{\nu,\beta}, \quad h^\mu = \hat{\mathbf{e}}_{\mu,\alpha} \cdot \hat{\mathbf{h}}, \quad (\text{B2})$$

with $\mu, \nu \in \{0, +, -\}$. The Holstein-Primakoff bosons satisfy

$$[a_{\mathbf{x},\alpha}, a_{\mathbf{y},\beta}^\dagger] = \delta_{\mathbf{x},\mathbf{y}} \delta_{\alpha,\beta} \mathbb{1}, \quad [a_{\mathbf{x},\alpha}, a_{\mathbf{y},\beta}] = 0,$$

and they are related to the spin operators via

$$\begin{aligned} S_{\mathbf{x},\alpha}^- &= \sqrt{2S} a_{\mathbf{x},\alpha}^\dagger \left(\mathbb{1} - \frac{\hat{n}_{\mathbf{x},\alpha}}{2S} \right)^{\frac{1}{2}} \simeq \sqrt{2S} a_{\mathbf{x},\alpha}^\dagger + O(S^{-\frac{1}{2}}), \\ S_{\mathbf{x},\alpha}^+ &= \sqrt{2S} \left(\mathbb{1} - \frac{\hat{n}_{\mathbf{x},\alpha}}{2S} \right)^{\frac{1}{2}} a_{\mathbf{x},\alpha} \simeq \sqrt{2S} a_{\mathbf{x},\alpha} + O(S^{-\frac{1}{2}}), \\ S_{\mathbf{x},\alpha}^z &= S - \hat{n}_{\mathbf{x},\alpha}, \end{aligned} \quad (\text{B3})$$

where the boson density on each site is $\hat{n}_{\mathbf{x},\alpha} = a_{\mathbf{x},\alpha}^\dagger a_{\mathbf{x},\alpha}$. The approximation on the right is valid in the limit $\langle \hat{n}_{\mathbf{x},\alpha} \rangle \ll 2S$, giving us

$$\mathbf{S}_{\mathbf{x},\alpha} \simeq \sqrt{S}(a_{\mathbf{x},\alpha}^\dagger \hat{\mathbf{e}}_{+,\alpha} + a_{\mathbf{x},\alpha} \hat{\mathbf{e}}_{-,\alpha}) + (S - \hat{n}_{\mathbf{x},\alpha}) \hat{\mathbf{e}}_{0,\alpha}.$$

Using the Holstein-Primakoff transformation in the spin Hamiltonian, and looking no further than quadratic terms, we have two contributions. First we have the classical contribution to the energy

$$\begin{aligned} E_{\text{cl}} &= N_c [S(S+1)\varepsilon_{\text{xc}} - hN_s S], \\ \varepsilon_{\text{xc}} &= \frac{1}{2} \sum_{\alpha,\beta} \sum_{\delta} \mathcal{J}_{\delta,\alpha,\beta}^{00}, \end{aligned} \quad (\text{B4})$$

with ε_{ex} being the exchange energy per magnetic unit cell. The $N_c S^2 \varepsilon_{\text{ex}}$ and $-N_h S$ pieces appear directly, and the $N_c S \varepsilon_{\text{ex}}$ is an extra piece absorbed from the quadratic boson Hamiltonian when putting it in Bogoliubov-de Gennes form. Each $\mathcal{J}^{\pm 0}, \mathcal{J}^{0\pm}$ part of the exchange (at order $S^{3/2}$) and $h^\pm$ parts of the magnetic field (at order $S^{1/2}$) constitute the linear terms in the bosons. We will assume that we are at sufficiently high magnetic fields so that the polarized state $\hat{\mathbf{z}}_\alpha = \hat{\mathbf{h}}$ is the global minimum of $\mathcal{H}^{(0)}$, which means that $\mathcal{H}^{(1)}$ vanishes. The next terms are those quadratic in the boson operators, which are most easily expressed in terms of the momentum space

quantities

$$a_{q,\alpha} = \frac{1}{\sqrt{N_c}} \sum_x e^{-iq \cdot x} a_{x,\alpha}, \quad \mathcal{J}_{q,\alpha,\beta}^{\mu,\nu} = \sum_{\delta} e^{-iq \cdot \delta} \mathcal{J}_{\delta,\alpha,\beta}^{\mu,\nu}, \quad (\text{B5})$$

and the Nambu spinors

$$A_q = \begin{pmatrix} a_q \\ a_{-q}^\dagger \end{pmatrix}, \quad [A_q, A_p^\dagger] = \delta_{p,q} \delta_{x,y} \begin{pmatrix} +\mathbb{1} & \\ & -\mathbb{1} \end{pmatrix} = \delta_{p,q} \tau_3. \quad (\text{B6})$$

We find

$$\begin{aligned} \mathcal{H}^{(2)} &= \frac{S}{2} \sum_q \sum_{\alpha,\beta} [A_{q,\alpha,\beta} a_{q,\alpha}^\dagger a_{q,\beta} + A_{-q,\beta,\alpha} a_{-q,\alpha}^\dagger a_{-q,\beta} + B_{q,\alpha,\beta} a_{q,\alpha}^\dagger a_{-q,\beta}^\dagger + B_{q,\beta,\alpha}^* a_{-q,\alpha} a_{q,\beta}] \\ &= \frac{S}{2} \sum_q A_q^\dagger \begin{pmatrix} \mathcal{A}_q & \mathcal{B}_q \\ \mathcal{B}_q^\dagger & \mathcal{A}_{-q}^T \end{pmatrix} A_q, \end{aligned} \quad (\text{B7})$$

with the $N_s \times N_s$ matrices

$$\begin{aligned} \mathcal{A}_{q,\alpha,\beta} &= \mathcal{J}_{q,\alpha,\beta}^{+-} + \delta_{\alpha\beta} \left(\frac{h}{S} - \sum_{\gamma} \mathcal{J}_{q=0,\alpha,\gamma}^{00} \right), \\ \mathcal{B}_{q,\alpha,\beta} &= \mathcal{J}_{q,\alpha,\beta}^{++}, \end{aligned} \quad (\text{B8})$$

describing the boson hopping and pairing, respectively. After a bosonic Bogoliubov transformation, $\mathcal{H}^{(2)}$ can be brought into canonical form,

$$\begin{aligned} \mathcal{H}^{(2)} &= \frac{S}{2} \sum_q (\gamma_q^\dagger)^T (\gamma_{-q})^T \begin{pmatrix} \Omega_q & 0 \\ 0 & \Omega_{-q} \end{pmatrix} \begin{pmatrix} \gamma_q \\ \gamma_{-q}^\dagger \end{pmatrix} \\ &= \frac{S}{2} \sum_{q,\alpha} (\omega_{q,\alpha} \gamma_{q,\alpha}^\dagger \gamma_{q,\alpha} + \omega_{-q,\alpha} \gamma_{-q,\alpha}^\dagger \gamma_{-q,\alpha}) \\ &= S \sum_{q,\alpha} \omega_{q,\alpha} \left(\gamma_{q,\alpha}^\dagger \gamma_{q,\alpha} + \frac{1}{2} \right). \end{aligned} \quad (\text{B9})$$

From this we identify the quantum zero-point energy as

$$E_{\text{qu}} = \frac{S}{2} \sum_{q,\alpha} \omega_{q,\alpha}.$$

The ground-state energy per magnetic unit cell within linear spin-wave theory is

$$\frac{E_{\text{cl}} + E_{\text{qu}}}{N_c} = S(S+1)\varepsilon_{\text{xc}} - hN_s S + \frac{S}{2N_c} \sum_{q,\alpha} \omega_{q,\alpha} \rightarrow \frac{1}{2} S(S+1) \sum_{\alpha,\beta} \sum_{\delta} \hat{z}_\alpha \cdot \mathbf{J}_{\delta,\alpha,\beta} \cdot \hat{z}_\beta - hS \sum_{\alpha} \hat{\mathbf{h}} \cdot \hat{z}_\alpha + \frac{1}{2} S \sum_{\alpha} \int_{\mathbb{T}^2} d\mathbf{q} \omega_{q,\alpha}, \quad (\text{B10})$$

where the last line holds in the thermodynamic limit.

In the polarized phase, the magnetic unit cell coincides with the honeycomb unit cell with $N_s = 2$, and the local moments are aligned with the field direction $\hat{\mathbf{h}}(\varphi)$. We will use this field angle dependence of the corresponding free energy to calculate the torque and magnetotropic coefficient. Our choice of local axes describing the magnetic order is

$$\begin{aligned} \hat{x}_\alpha &= -\sin(\varphi) \hat{\mathbf{a}} + \cos(\varphi) \hat{\mathbf{b}} = \partial_\varphi \hat{\mathbf{h}}, \\ \hat{y}_\alpha &= \hat{\mathbf{c}}^* = \hat{\mathbf{h}} \times \partial_\varphi \hat{\mathbf{h}}, \\ \hat{z}_\alpha &= \cos(\varphi) \hat{\mathbf{a}} + \sin(\varphi) \hat{\mathbf{b}} = \hat{\mathbf{h}}. \end{aligned} \quad (\text{B11})$$

Note that we are free to globally rotate our local frame by an arbitrary angle θ about $\hat{z}_\alpha = \hat{\mathbf{h}}$, under which $\hat{\mathbf{e}}_{\pm,\alpha} \mapsto e^{\pm i\theta} \hat{\mathbf{e}}_{\pm,\alpha}$. Under such a rotation, the classical energy and hopping part of the boson Hamiltonian are unchanged, while the pairing part

of the boson Hamiltonian picks up an overall phase of $e^{2i\theta}$. This extra phase can be removed by transforming $a \mapsto e^{+i\theta}a$ and $a^\dagger \mapsto e^{-i\theta}a^\dagger$, so the resulting Hamiltonians are equivalent.

We begin with the Kitaev-Heisenberg-Gamma interactions with potentially anisotropic couplings ($x = y \neq z$ consistent with the space-group symmetry $C2/m$), with, for instance,

$$\mathbf{J}_{\delta_z, A, B} = \begin{pmatrix} J_z & \Gamma_z & \Gamma'_z \\ \Gamma_z & J_z & \Gamma'_z \\ \Gamma'_z & \Gamma'_z & J_z + K_z \end{pmatrix}. \quad (\text{B12})$$

Let us consider the classical part of the energy first. By setting $J_z = J$ and $J_x = J_y = J + \delta J$ (as well as for K, Γ, Γ'), we find

$$\begin{aligned} e_{\text{cl},0} &= \frac{1}{2}S(S+1) \sum_{\alpha,\beta} \sum_{\delta} \hat{\mathbf{e}}_{0,\alpha} \cdot \mathbf{J}_{\delta,\alpha,\beta} \cdot \hat{\mathbf{e}}_{0,\beta} - hS \sum_{\alpha} \hat{\mathbf{h}} \cdot \hat{\mathbf{e}}_{0,\alpha} \\ &= S(S+1) \left[3 \left(J + \frac{2}{3}\delta J \right) + \left(K + \frac{2}{3}\delta K \right) - \left(\Gamma + \frac{2}{3}\delta\Gamma \right) - 2 \left(\Gamma' + \frac{2}{3}\delta\Gamma' \right) \right] \\ &\quad - 2hS - \frac{1}{3}S(S+1)[\delta K + 2\delta\Gamma - 2\delta\Gamma'] \cos(2\varphi). \end{aligned} \quad (\text{B13})$$

In the absence of non-Heisenberg anisotropy ($\delta K = \delta\Gamma = \delta\Gamma' = 0$), the classical part of the energy is constant. With anisotropy there is an extra $\cos(2\varphi)$ dependence that only couples to the anisotropy in the non-Heisenberg exchange, $\delta K + 2\delta\Gamma - 2\delta\Gamma'$. Therefore, the classical part contributes $\sin(2\varphi)$ to the $\hat{\mathbf{c}}^*$ component of the torque and $\cos(2\varphi)$ to the corresponding magnetotropic coefficient

$$\begin{aligned} \hat{\mathbf{c}}^* \cdot \boldsymbol{\tau}_{\text{cl}} &= \frac{2}{3}S(S+1)[\delta K + 2\delta\Gamma - 2\delta\Gamma'] \sin(2\varphi), \\ k_{\text{cl}} &= \frac{4}{3}S(S+1)[\delta K + 2\delta\Gamma - 2\delta\Gamma'] \cos(2\varphi). \end{aligned} \quad (\text{B14})$$

We now consider the linear spin-wave correction to the free energy, which involves diagonalizing the quadratic boson Hamiltonian. Writing the terms in a way that makes the dependence on anisotropy and field angle explicit, we find

$$\begin{aligned} \mathcal{A}_{q,A,A} = \mathcal{A}_{q,B,B} &= \frac{h}{S} - \left[3 \left(J + \frac{2}{3}\delta J \right) + \left(K + \frac{2}{3}\delta K \right) - \left(\Gamma + \frac{2}{3}\delta\Gamma \right) - 2 \left(\Gamma' + \frac{2}{3}\delta\Gamma' \right) \right] + \frac{1}{3}[\delta K + 2\delta\Gamma - 2\delta\Gamma'] \cos(2\varphi), \\ \mathcal{A}_{q,A,B} = \mathcal{A}_{-q,B,A} &= \frac{1}{6}[(6J + 2K + \Gamma + 2\Gamma')(e^{2\pi i q_1} + e^{2\pi i q_2} + 1) + (6\delta J + 2\delta K + \delta\Gamma + 2\delta\Gamma')(e^{2\pi i q_1} + e^{2\pi i q_2})] \\ &\quad + \frac{1}{12}[(K + 2\Gamma - 2\Gamma')(e^{2\pi i q_1} + e^{2\pi i q_2} - 2) + (\delta K + 2\delta\Gamma - 2\delta\Gamma')(e^{2\pi i q_1} + e^{2\pi i q_2})] \cos(2\varphi) \\ &\quad + \frac{1}{4\sqrt{3}}[(K + 2\Gamma - 2\Gamma') + (\delta K + 2\delta\Gamma - 2\delta\Gamma')](e^{2\pi i q_1} - e^{2\pi i q_2}) \sin(2\varphi) \end{aligned} \quad (\text{B15})$$

for the hopping matrix, and

$$\begin{aligned} \mathcal{B}_{q,A,B} = \mathcal{B}_{-q,B,A} &= -\frac{1}{2}[(\Gamma + 2\Gamma')(e^{2\pi i q_1} + e^{2\pi i q_2} + 1) + (\delta\Gamma + 2\delta\Gamma')(e^{2\pi i q_1} + e^{2\pi i q_2})] \\ &\quad - \frac{i}{\sqrt{6}}[(K - \Gamma + \Gamma') + (\delta K - \delta\Gamma + \delta\Gamma')](e^{2\pi i q_1} - e^{2\pi i q_2}) \cos(\varphi) \\ &\quad - \frac{i}{3\sqrt{2}}[(K - \Gamma + \Gamma')(e^{2\pi i q_1} + e^{2\pi i q_2} - 2) + (\delta K - \delta\Gamma + \delta\Gamma')(e^{2\pi i q_1} + e^{2\pi i q_2})] \sin(\varphi) \\ &\quad + \frac{1}{12}[(K + 2\Gamma - 2\Gamma')(e^{2\pi i q_1} + e^{2\pi i q_2} - 2) + (\delta K + 2\delta\Gamma - 2\delta\Gamma')(e^{2\pi i q_1} + e^{2\pi i q_2})] \cos(2\varphi) \\ &\quad + \frac{1}{4\sqrt{3}}[(K + 2\Gamma - 2\Gamma') + (\delta K + 2\delta\Gamma - 2\delta\Gamma')](e^{2\pi i q_1} - e^{2\pi i q_2}) \sin(2\varphi) \end{aligned} \quad (\text{B16})$$

for the pairing matrix. The spectrum of the linear spin-wave Hamiltonian can be found by diagonalizing the (generally non-Hermitian) matrix

$$\mathcal{M}_q = \tau_3 \mathcal{H}_q^{(2)} = \begin{pmatrix} \mathcal{A}_q & \mathcal{B}_q \\ -\mathcal{B}_q^\dagger & -\mathcal{A}_{-q}^\dagger \end{pmatrix},$$

which gives us the eigenvalues in $\pm$ pairs $\{\omega_{q,\alpha}, -\omega_{-q,\alpha}\}$ ($\mathcal{H}_q^{(2)}$ can be arranged to be positive-definite with a sufficiently high magnetic field).

- [1] A. Kitaev, Anyons in an exactly solved model and beyond, *Ann. Phys.* **321**, 2 (2006).
- [2] G. Jackeli and G. Khaliullin, Mott Insulators in the Strong Spin-Orbit Coupling Limit: From Heisenberg to a Quantum Compass and Kitaev Models, *Phys. Rev. Lett.* **102**, 017205 (2009).
- [3] J. G. Rau, E. Kin-Ho Lee, and H.-Y. Kee, Generic Spin Model for the Honeycomb Iridates Beyond the Kitaev Limit, *Phys. Rev. Lett.* **112**, 077204 (2014).
- [4] J. G. Rau and H.-Y. Kee, Trigonal distortion in the honeycomb iridates: Proximity of zigzag and spiral phases in Na_2IrO_3 , [arXiv:1408.4811](#) (2014).
- [5] J. Chaloupka, G. Jackeli, and G. Khaliullin, Kitaev-Heisenberg Model on a Honeycomb Lattice: Possible Exotic Phases in Iridium Oxides A_2IrO_3 , *Phys. Rev. Lett.* **105**, 027204 (2010).
- [6] Y. Singh, S. Manni, J. Reuther, T. Berlijn, R. Thomale, W. Ku, S. Trebst, and P. Gegenwart, Relevance of the Heisenberg-Kitaev Model for the Honeycomb Lattice Iridates A_2IrO_3 , *Phys. Rev. Lett.* **108**, 127203 (2012).
- [7] K. A. Modic, T. E. Smidt, I. Kimchi, N. P. Breznay, A. Biffin, S. Choi, R. D. Johnson, R. Coldea, P. Watkins-Curry, G. T. McCandless, J. Y. Chan, F. Gandara, Z. Islam, A. Vishwanath, A. Shekhter, R. D. McDonald, and J. G. Analytis, Realization of a three-dimensional spin-anisotropic harmonic honeycomb iridate, *Nat. Commun.* **5**, 4203 (2014).
- [8] K. W. Plumb, J. P. Clancy, L. J. Sandilands, V. V. Shankar, Y. F. Hu, K. S. Burch, H.-Y. Kee, and Y.-J. Kim, $\alpha\text{-RuCl}_3$: A spin-orbit assisted Mott insulator on a honeycomb lattice, *Phys. Rev. B* **90**, 041112(R) (2014).
- [9] L. J. Sandilands, Y. Tian, K. W. Plumb, Y.-J. Kim, and K. S. Burch, Scattering Continuum and Possible Fractionalized Excitations in $\alpha\text{-RuCl}_3$, *Phys. Rev. Lett.* **114**, 147201 (2015).
- [10] H.-S. Kim, V. S. Venkataraman, A. Catuneanu, and H.-Y. Kee, Kitaev magnetism in honeycomb $\alpha\text{-RuCl}_3$ with intermediate spin-orbit coupling, *Phys. Rev. B* **91**, 241110(R) (2015).
- [11] A. Banerjee, C. A. Bridges, J.-Q. Yan, A. A. Aczel, L. Li, M. B. Stone, G. E. Granroth, M. D. Lumsden, Y. Yiu, J. Knolle, S. Bhattacharjee, D. L. Kovrizhin, R. Moessner, D. A. Tennant, D. G. Mandrus, and S. E. Nagler, Proximate Kitaev quantum spin liquid behavior in a honeycomb magnet, *Nat. Mater.* **15**, 733 (2016).
- [12] L. J. Sandilands, Y. Tian, A. A. Reijnders, H.-S. Kim, K. W. Plumb, Y.-J. Kim, H.-Y. Kee, and K. S. Burch, Spin-orbit excitations and electronic structure of the putative Kitaev magnet $\alpha\text{-RuCl}_3$, *Phys. Rev. B* **93**, 075144 (2016).
- [13] J. M. Fletcher, W. E. Gardner, A. C. Fox, and G. Topping, X-ray, infrared, and magnetic studies of α - and β -ruthenium trichloride, *J. Chem. Soc. A* **1038** (1967).
- [14] S. K. Choi, R. Coldea, A. N. Kolmogorov, T. Lancaster, I. I. Mazin, S. J. Blundell, P. G. Radaelli, Y. Singh, P. Gegenwart, K. R. Choi, S.-W. Cheong, P. J. Baker, C. Stock, and J. Taylor, Spin Waves and Revised Crystal Structure of Honeycomb Iridate Na_2IrO_3 , *Phys. Rev. Lett.* **108**, 127204 (2012).
- [15] J. Chaloupka, G. Jackeli, and G. Khaliullin, Zigzag Magnetic Order in the Iridium Oxide Na_2IrO_3 , *Phys. Rev. Lett.* **110**, 097204 (2013).
- [16] J. A. Sears, M. Songvilay, K. W. Plumb, J. P. Clancy, Y. Qiu, Y. Zhao, D. Parshall, and Y.-J. Kim, Magnetic order in $\alpha\text{-RuCl}_3$: A honeycomb-lattice quantum magnet with strong spin-orbit coupling, *Phys. Rev. B* **91**, 144420 (2015).
- [17] R. D. Johnson, S. C. Williams, A. A. Haghighirad, J. Singleton, V. Zapf, P. Manuel, I. I. Mazin, Y. Li, H. O. Jeschke, R. Valentí, and R. Coldea, Monoclinic crystal structure of $\alpha\text{-RuCl}_3$ and the zigzag antiferromagnetic ground state, *Phys. Rev. B* **92**, 235119 (2015).
- [18] H. B. Cao, A. Banerjee, J.-Q. Yan, C. A. Bridges, M. D. Lumsden, D. G. Mandrus, D. A. Tennant, B. C. Chakoumakos, and S. E. Nagler, Low-temperature crystal and magnetic structure of $\alpha\text{-RuCl}_3$, *Phys. Rev. B* **93**, 134423 (2016).
- [19] H.-S. Kim and H.-Y. Kee, Crystal structure and magnetism in $\alpha\text{-RuCl}_3$: An ab initio study, *Phys. Rev. B* **93**, 155143 (2016).
- [20] L. Janssen, E. C. Andrade, and M. Vojta, Magnetization processes of zigzag states on the honeycomb lattice: Identifying spin models for $\alpha\text{-RuCl}_3$ and Na_2IrO_3 , *Phys. Rev. B* **96**, 064430 (2017).
- [21] Z.-X. Liu and B. Normand, Dirac and Chiral Quantum Spin Liquids on the Honeycomb Lattice in a Magnetic Field, *Phys. Rev. Lett.* **120**, 187201 (2018).
- [22] P. Lampen-Kelley, L. Janssen, E. C. Andrade, S. Rachel, J. Q. Yan, C. Balz, D. G. Mandrus, S. E. Nagler, and M. Vojta, Field-induced intermediate phase in $\alpha\text{-RuCl}_3$: Non-coplanar order, phase diagram, and proximate spin liquid, [arXiv:1807.06192](#) (2018).
- [23] S.-H. Baek, S.-H. Do, K.-Y. Choi, Y. S. Kwon, A. U. B. Wolter, S. Nishimoto, J. van den Brink, and B. Büchner, Evidence for a Field-Induced Quantum Spin Liquid in $\alpha\text{-RuCl}_3$, *Phys. Rev. Lett.* **119**, 037201 (2017).
- [24] A. U. B. Wolter, L. T. Corredor, L. Janssen, K. Nenkov, S. Schönecker, S.-H. Do, K.-Y. Choi, R. Albrecht, J. Hunger, T. Doert, M. Vojta, and B. Büchner, Field-induced quantum criticality in the Kitaev system $\alpha\text{-RuCl}_3$, *Phys. Rev. B* **96**, 041405(R) (2017).
- [25] J. Zheng, K. Ran, T. Li, J. Wang, P. Wang, B. Liu, Z.-X. Liu, B. Normand, J. Wen, and W. Yu, Gapless Spin Excitations in the Field-Induced Quantum Spin Liquid Phase of $\alpha\text{-RuCl}_3$, *Phys. Rev. Lett.* **119**, 227208 (2017).
- [26] N. Janša, A. Zorko, M. Gomilšek, M. Pregelj, K. W. Krämer, D. Biner, A. Biffin, C. Rüegg, and M. Klanjšek, Observation of two types of fractional excitation in the Kitaev honeycomb magnet, *Nat. Phys.* **14**, 786 (2018).
- [27] Y. Kasahara, T. Ohnishi, Y. Mizukami, O. Tanaka, S. Ma, K. Sugii, N. Kurita, H. Tanaka, J. Nasu, Y. Motome, T. Shibauchi, and Y. Matsuda, Majorana quantization and half-integer thermal quantum Hall effect in a Kitaev spin liquid, *Nature (London)* **559**, 227 (2018).
- [28] T. Yokoi, S. Ma, Y. Kasahara, S. Kasahara, T. Shibauchi, N. Kurita, H. Tanaka, J. Nasu, Y. Motome, C. Hickey, S. Trebst, and Y. Matsuda, Half-integer quantized anomalous thermal Hall effect in the Kitaev material $\alpha\text{-RuCl}_3$, [arXiv:2001.01899](#) (2020).
- [29] D. Aasen, Roger S. K. Mong, B. M. Hunt, D. Mandrus, and J. Alicea, Electrical Probes of the Non-Abelian Spin Liquid in Kitaev Materials, *Phys. Rev. X* **10**, 031014 (2020).
- [30] P. A. McClarty, X.-Y. Dong, M. Gohlke, J. G. Rau, F. Pollmann, R. Moessner, and K. Penc, Topological magnons in Kitaev magnets at high fields, *Phys. Rev. B* **98**, 060404(R) (2018).
- [31] K. A. Modic, M. D. Bachmann, B. J. Ramshaw, F. Arnold, K. R. Shirer, A. Estry, J. B. Betts, N. J. Ghimire, E. D. Bauer,

- M. Schmidt, M. Baenitz, E. Svanidze, R. D. McDonald, A. Shekhter, and P. J. W. Moll, Resonant torsion magnetometry in anisotropic quantum materials, *Nat. Commun.* **9**, 3975 (2018).
- [32] K. A. Modic, R. D. McDonald, J. P. C. Ruff, M. D. Bachmann, Y. Lai, J. C. Palmstrom, D. Graf, M. Chan, F. F. Balakirev, J. B. Betts, G. S. Boebinger, M. Schmidt, D. A. Sokolov, P. J. W. Moll, B. J. Ramshaw, and A. Shekhter, Scale-invariance of a spin liquid in high magnetic fields, *Nat. Phys.* (2020), doi:[10.1038/s41567-020-1028-0](https://doi.org/10.1038/s41567-020-1028-0).
- [33] L. Zou and Y.-C. He, Field-induced QCD₃-Chern-Simons quantum criticalities in Kitaev materials, *Phys. Rev. Res.* **2**, 013072 (2020).
- [34] S. Y. Park, S. H. Do, K. Y. Choi, D. Jang, T. H. Jang, J. Schefer, C. M. Wu, J. S. Gardner, J. M. S. Park, J. H. Park, and S. Ji, Emergence of the isotropic Kitaev honeycomb lattice with two-dimensional Ising universality in α -RuCl₃, [arXiv:1609.05690](https://arxiv.org/abs/1609.05690) (2016).
- [35] L. Janssen, S. Koch, and M. Vojta, Magnon dispersion and dynamic spin response in three-dimensional spin models for α -RuCl₃, *Phys. Rev. B* **101**, 174444 (2020).
- [36] S. D. Das, S. Kundu, Z. Zhu, E. Mun, R. D. McDonald, G. Li, L. Balicas, A. McCollam, G. Cao, J. G. Rau, H.-Y. Kee, V. Tripathi, and S. E. Sebastian, Magnetic anisotropy of the alkali iridate Na₂IrO₃ at high magnetic fields: Evidence for strong ferromagnetic Kitaev correlations, *Phys. Rev. B* **99**, 081101(R) (2019).
- [37] I. A. Leahy, C. A. Pocs, P. E. Siegfried, D. Graf, S.-H. Do, K.-Y. Choi, B. Normand, and M. Lee, Anomalous Thermal Conductivity and Magnetic Torque Response in the Honeycomb Magnet α -RuCl₃, *Phys. Rev. Lett.* **118**, 187203 (2017).
- [38] M. Li, I. Rousochatzakis, and N. B. Perkins, Reentrant incommensurate order and anomalous magnetic torque in the Kitaev magnet β -Li₂IrO₃, *Phys. Rev. Research* **2**, 033328 (2020).
- [39] K. Riedl, Y. Li, S. M. Winter, and R. Valentí, Sawtooth Torque in Anisotropic $j_{\text{eff}} = 1/2$ Magnets: Application to α -RuCl₃, *Phys. Rev. Lett.* **122**, 197202 (2019).
- [40] J. Knolle, D. L. Kovrizhin, J. T. Chalker, and R. Moessner, Dynamics of a Two-Dimensional Quantum Spin Liquid: Signatures of Emergent Majorana Fermions and Fluxes, *Phys. Rev. Lett.* **112**, 207203 (2014).
- [41] L. E. Chern, E. Z. Zhang, and Y. B. Kim, Sign structure of thermal Hall conductivity for in-plane-field polarized Kitaev magnets, [arxiv:2008.12788](https://arxiv.org/abs/2008.12788) (2020).
- [42] A. Go, K. Hwang, J. H. Seong, T. Shibauchi, and E.-G. Moon, Identification of a Kitaev spin liquid by magnetic field angle dependence, [arXiv:2004.06119](https://arxiv.org/abs/2004.06119) (2020).
- [43] O. Tanaka, Y. Mizukami, R. Harasawa, K. Hashimoto, N. Kurita, H. Tanaka, S. Fujimoto, Y. Matsuda, E.-G. Moon, and T. Shibauchi, Thermodynamic evidence for field-angle dependent Majorana gap in a Kitaev spin liquid, [arXiv:2007.06757](https://arxiv.org/abs/2007.06757) (2020).
- [44] J. G. Rau, P. A. McClarty, and R. Moessner, Pseudo-Goldstone Gaps and Order-by-Quantum Disorder in Frustrated Magnets, *Phys. Rev. Lett.* **121**, 237201 (2018).