Dominant Kitaev interactions in the honeycomb materials
$\text{Na}_3\text{Co}_2\text{SbO}_6$ and $\text{Na}_2\text{Co}_2\text{TeO}_6$

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Cobaltates with 3d based layered honeycomb structure were recently proposed as Kitaev magnets and putative candidates to host the long-sought Kitaev spin liquid. Here we present inelastic neutron scattering results down to 50 mK for powder samples of $\text{Na}_3\text{Co}_2\text{SbO}_6$ and $\text{Na}_2\text{Co}_2\text{TeO}_6$, with high resolution in regions of low momentum and energy transfers. We compare the experimental data below the antiferromagnetic zigzag ordering temperature with dynamical structure factors obtained within spin wave theory. We search the wide parameter range of a $K$-$J_1$-$\Gamma$-$\Gamma'$-$J_3$ spin 1/2 model and identify the best fits to constant momentum cuts of the inelastic neutron data. The powder average limits the selection of a unique parameter set for each material, but we see clear trends towards ferromagnetic Kitaev exchange in both compounds, and ratios $|K/J_1|$ could be as large as 5…25; in contrast, antiferromagnetic Kitaev exchange cannot be ruled out, but requires a fine-tuning of all involved spin exchange couplings with only moderate ratio $|K/J_1| \sim 1$. Our experimental data are incompatible with a purely isotropic Heisenberg model.

I. INTRODUCTION

The search for quantum spin liquids (QSLs) is one of the great challenges in the field of strongly correlated electrons [1–5]. A QSL state possesses short-ranged magnetic fluctuations but no long-range order. It is, hence, a featureless state which is mostly characterized by the absence of clear and easily-identifiable features. Despite many years of extensive research, few candidate spin liquid materials are known [6–8], and the list of theory proposals for experimental realisations is rather short. Mott insulators with geometric or spin-exchange frustration are the traditional places to search for QSLs [3]. As an additional complication, in two and three spatial dimensions models for QSLs can usually not be solved exactly and theoreticians rely on approximate numerical methods to identify and characterize such states.

Kitaev’s seminal work opened an alternative avenue, where he introduced an exactly soluble QSL Hamiltonian with bond-dependent Ising interactions on the honeycomb lattice [9]. The exact solution revealed that the QSL consists of free Majorana fermions coupled to a static $Z_2$ gauge field, nicely illustrating the fractionalization of the original spin degrees of freedom.

This purely theoretical development was further fueled by the influential work by Jackeli and Khaliullin [10] who proposed that Kitaev model might be realized in transition metal oxides with strong spin-orbit coupling. The idea is based on an observation that the exchange interactions between spin-orbital entangled moments are highly anisotropic and also depend on the bond directions [11]. For a pedagogical review about spin-orbit entangled states of matter see Ref. [12]. The first Kitaev candidate material was $\text{Na}_2\text{IrO}_3$ [13, 14] along with its sister compound $\alpha$-$\text{Li}_2\text{IrO}_3$ [15], which both turned out to be magnetically ordered at low temperatures. Despite this setback, subsequent work substantiated the claim that these iridates possess non-negligible Kitaev spin interactions along with other generic spin exchange [16–39].

The second wave of experiments focussed on $\alpha$-$\text{RuCl}_3$. While its low-temperature phase revealed magnetic long-range order of the zigzag type [40] just like $\text{Na}_2\text{IrO}_3$, its excitation spectrum measured with inelastic neutrons was interpreted as a combination of magnons stemming from the magnetic order and additional features due to the system’s proximity to a QSL [37, 41–44]. A milestone was the report of a quantized thermal conductance when a magnetic field is applied to $\alpha$-$\text{RuCl}_3$ [45, 46]; the measured conductance plateau is in agreement with a chiral Majorana mode as expected from Kitaev’s model with applied magnetic field (corresponding to a non-Abelian QSL) [9]. It is generally thought that the generic spin exchange spoiling the QSL state in zero field needs to be suppressed by a sufficiently strong magnetic field, until the remaining compass interactions can dominate and cause the non-Abelian QSL state to prevail.

A major challenge in obtaining a better theoretical understanding is to identify the details of the generic non-Kitaev interactions in the candidate materials. However, not even the strength and sign of the Kitaev compass interactions are known with certainty. Experimental determination of these coupling constants is necessarily indirect. Previous estimates have come from magnetic susceptibility measurements [44, 47, 48], Raman spectroscopy [49], X-ray scattering [50–52] and most commonly, inelastic neutron scattering [16, 41, 53–56]. Theoretical determination of these coupling constants is a very subtle task, ultimately relying on density functional

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theory [24, 30, 42, 57–63].

While most attention was given to the iridates and ruthenates of d⁵ ions Ir⁴⁺ and Ru³⁺, respectively, a recent proposal emphasized that cobaltates of d⁷ ions such as Co₂²⁺ might be another platform for Kitaev candidate materials with pseudospin-½ [63–65], despite some skepticism that spin-orbit coupling is insufficient to promote compass interactions. The first series of experimental results including Na₂Co₂TeO₆ and Na₃Co₃SbO₆ confirmed that their low-temperature phase is again antiferromagnetically zigzag-ordered [54–56, 66–69], but most works concluded that modeling involving Kitaev interactions is compatible with the results or even the likely scenario [60, 70].

Here we present inelastic neutron scattering results for Na₃Co₂SbO₆ and Na₂Co₂TeO₆. In particular, our data was measured at temperatures as low as 50 mK with high resolution in regions of low momentum and energy transfers. We model the dynamical structure factor within spin wave theory, and show that a purely isotropic J₁–J₂–J₃ model is incompatible with the experimental data. By fitting our model to our experimental data we show that best fits are obtained for extended Heisenberg–Kitaev models with either ferro- or antiferromagnetic Kitaev exchange K. We argue that the most likely scenario for both materials is a dominant ferromagnetic K < 0 with |K|/J₁ being in the range 5–25. Hence, our results indicate that perturbing the magnetically ordered ground-state by strain [71, 72], pressure [37] or an applied magnetic field [45, 57] would be an exciting attempt to push the material into the QSL phase.

The paper is organised as follows. In Sec. II we discuss the sample preparation of the materials used in Sec. III to perform inelastic neutron scattering experiments. In Sec. IV we perform linear spin wave theory and derive the scattering intensity which we fit to the experimental data in Sec. V. In the discussion in Sec. VI, we elaborate on a model duality between ferromagnetic and antiferromagnetic K in agreement with our best fits. Next, we argue that both excitation spectra are gapped, albeit with a much smaller gap for Na₂Co₂TeO₆ as previously reported. We discuss the classical ground states and the corresponding spin quantization axis, and elaborate on predictions for single crystal measurements. The paper ends with a conclusion in Sec. VII.

II. SAMPLE PREPARATION

The same polycrystalline sample of Na₃Co₂SbO₆ was used in this work as in Wong et al. [66]. A polycrystalline sample of Na₂Co₂TeO₆ was prepared from Na₂CO₃ (Merck, 99.9%), Co₂O₃ (Sigma-Aldrich, 99.99%), and TeO₂ (Sigma-Aldrich, 99.9995%). Following the method described by Berthelot et al. [73], the reagents were ground together at the correct stoichiometry, pressed into pellets, and calcined twice at 850°C in air for 12 hours with intermediate regrinding. Sample purity was confirmed by X-ray powder diffraction. Magnetic property measurements were also consistent with previous reports for Na₂Co₂TeO₆: zero-field cooled temperature-dependent susceptibility in a 0.1 T field showed a sharp antiferromagnetic transition at Tₐ = 27 K and a weaker feature at 16 K, which were suppressed under field-cooled conditions. A Curie-Weiss fit to the paramagnetic region yielded an effective magnetic moment of 4.02 μB/Co (within the range typically observed for Co₂²⁺) and a Weiss constant θCW = −1.4 K.

III. INELASTIC NEUTRON SCATTERING

Inelastic neutron scattering data were collected using the cold-neutron time-of-flight spectrometer Peli-can [74, 75] at the Australian Centre for Neutron Scattering. Approximately 5 g of each sample was held in an annular sample can fabricated from oxygen-free copper. This was cooled using a dilution insert inside a top-loading closed cycle cryostat. The instrument was aligned for 4.69 Å neutrons. The choppers were also rephased to allow the collection of data with λ/2 = 2.345 Å neutrons. Data were collected at 50 mK and 1.8 K and corrected for background by subtraction of an empty can and normalised to a standard vanadium sample. All raw detector data were processed using the freely available LAMP [76] software.

Magnetic Bragg reflections were observed at Q = 0.75 Å, confirming that the system was indeed magnetically ordered for the measured temperatures. Na₂Co₂TeO₆ at 50 mK clearly features a mode emanating from Q = 0.75 Å, corresponding to the M point in the Brillouin zone of the honeycomb layers. The dispersion relation close to this point appears to be linear. Our results are consistent with existing experiments [54–56, 77, 78], however, our ability to resolve features below 1 meV provides crucial detail at the gap point (i.e., where the magnon excitation gets closest to the elastic line). The single-Q cuts of the data in Fig. 1 (c), shown in Fig. 4 (a,c), exhibit a statistically significant excess of spectral weight down to the elastic line with an essentially linear intensity fall-off. This seemingly gapless spectrum disagrees somewhat with existing models, which are generally gapped by several meV. It it not obvious a priori how large of a modification to these models is necessary to fit our experiment.

Fig. 1 (e) shows the appearance of an unexpected ‘hourglass’ feature when the temperature is increased slightly from 50 mK. The excitation near zero energy is very broad in Q, and has a clearly observable waist that excludes the possibility that it is the result of magnon lifetime broadening. The Debye temperature of this material is known to be approximately 160 K [77], suggesting that this signal is unlikely to be due to phonon contamination. This feature appears well below the Neel temperature Tₐ, ruling out a critical phenomenon. This may be evidence of a nonlinear spin-wave mode, made possible
by the strong anisotropy in the system.

The spectrum for Na$_3$Co$_2$SbO$_6$ shows no evidence of any low-energy modes near the $M$ or $K$ points, the sole resolvable magnon excitation appears to emanate from $\Gamma$. The dispersion resembles a quadratic mode, which may be compatible with the magnon spectrum predicted by Ref. [64]. Unlike Na$_2$Co$_2$TeO$_6$, there is very little difference between the two temperatures in panels 1 (d) and (f).

IV. SPIN WAVE THEORY

We model the system using a six-parameter extended Heisenberg-Kitaev model as previously used in Refs. [54–56, 77]. The first order anisotropic interactions are the most general possible couplings that preserve $C_3$ symmetry, parameterised by $J_1, K, \Gamma, \Gamma'$ (explained below). These are supplemented by second and third-nearest neighbour Heisenberg couplings of respective strengths $J_2$ and $J_3$. This is motivated by ab initio calculations of realistic, distorted octahedral lattices [64] and the failure of simpler models to accurately reproduce analogous systems such as $\alpha$–RuCl$_3$ and A$_2$IrO$_3$ [79] (A=Na or Li). Following the original proposal for the realisation of bond-anisotropic couplings [10], we take the quantisation axes $x, y, z$ to be oriented as in Fig. 3, i.e., normal to the corresponding edge-sharing rectangles. The Hamiltonian may be expressed in the form

$$\mathcal{H} = \sum_{\langle ij \rangle} \left\{ J_1 S_i \cdot S_j + K S_i^\alpha S_j^\gamma + \Gamma \left( S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha \right) + \Gamma' \left( S_i^\sigma S_j^\alpha + S_i^\gamma S_j^\sigma + S_i^\sigma S_j^\gamma + S_i^\gamma S_j^\sigma \right) \right\} + \sum_{\langle \langle ij \rangle \rangle} J_2 S_i \cdot S_j + \sum_{\langle \langle \langle ij \rangle \rangle \rangle} J_3 S_i \cdot S_j \cdot$$

Here $J_i$ represents $i$th neighbor isotropic Heisenberg spin exchange, $K$ is nearest-neighbor Kitaev coupling while $\Gamma$ and $\Gamma'$ are symmetric off-diagonal couplings. Indices within single (double and triple) brackets $\langle \rangle$ denote nearest (2nd and 3rd nearest) neighbors lattice sites, and $\langle \langle \rangle \rangle$ is a bond with $\gamma = x, y, z$. On $z$ bonds we assume...
For both materials, our long-wavelength neutron experiments have access to the low energy $E < 4$ meV region of $Q - E$ space, at the cost of only seeing low-energy details. As we do not have access to the high-energy features that aided the qualitative fitting of models in Refs. [55] and [54], we require a quantitative goodness-of-fit measure to navigate the 5D parameter space. We therefore adopt the approach of Ref. [56]. We define

$$
\chi^2 = \sum_{Q,\omega} \frac{(I_{\text{exp}}(Q,\omega) - I_{\text{theory}}(Q,\omega))^2}{I_{\text{exp}}(Q,\omega)}
$$

where $I_{\text{exp}}$ is the experimental scattering intensity and $I_{\text{theory}}(Q,\omega) \propto F(Q)^2 S^\perp(Q,\omega)$. $F(Q)$ is the spherically symmetric atomic form factor of Co$^{2+}$ [80], and $S^\perp$ is as defined in (9). The theoretical intensity function is normalised to have the same total intensity as the experiment, when restricted to pixels within kinematic limits and at an energy above 4 meV. This energy cut was used to exclude the elastic line. All plots shown account for the form factor fall-off of the magnetic Co ions.

Due to the lack of clearly resolved high energy features in our 4.69 Å data on either substance [see Fig. 1 (a) and (b)], fine detail at low $Q$ provides the most useful information for model fitting. We therefore used the experimental intensity at 50 mK from Figs. 1 (c) and 1 (d) for $I_{\text{exp}}$, the outline of which is also shown superimposed on Fig. 5.

The powder averaging of the inelastic neutron data from the polycrystalline sample makes unique determination of best-fit parameters difficult. Previous reports on these compounds have consistently found weak $J_2$ when it is included in Na$_2$Co$_2$TeO$_6$ [54, 56, 77], and all previous fit attempts have neglected it when fitting Na$_3$Co$_2$SbO$_6$. Due to the small region of data available, we set $J_2 = 0$ to mitigate over-parameterisation.

Model parameters were optimised by the following procedure:

1. Suggest a starting guess for the six parameters.
2. Compute $\chi^2$ for a rectangle of side 2 meV centred on the guess in $\Gamma - \Gamma'$ space, $J_1 - K$ space and $J_1 - J_3$ space.
3. Compare the local minima of these three phase portraits, and update the guess to match.

The residual surface contained many shallow valleys, in which it was necessary to manually choose an updated parameter set. The parameters ultimately obtained should not be interpreted as rigorous global minima of the error term. Rather, they should be taken as estimates that reflect the general hierarchy of coupling strengths.

We chose the starting guesses for Na$_3$Co$_2$TeO$_6$ using the work of Ref. [56], which established regions of parameter space in agreement with their good quality high
FIG. 4. Best fit of LSWT models with $K > 0$ to the INS data of Na$_2$Co$_2$TeO$_6$ and Na$_3$Co$_2$SbO$_6$, showing both $K > 0$ and $K < 0$ fits.

energy data. We seek to further restrict the allowed parameter space and use these models to deduce qualitative features of the magnon spectrum, e.g., whether or not it has a gap in the excitation spectrum. No such study exists for Na$_3$Co$_2$SbO$_6$, so starting guesses were chosen centered on the sa and sb± best-fit parameters from Tab. I.

VI. DISCUSSION

A. Model Duality and sign of $K$

It is known that best fit parameters for the extended Heisenberg-Kitaev-$\Gamma$-$\Gamma'$ model tend to come in pairs. Looking at Na$_2$Co$_2$TeO$_6$, Na$_3$Co$_2$SbO$_6$ and the Na$_2$Co$_2$TeO$_6$-analogue Na$_2$Ni$_2$TeO$_6$, Refs. [55, 56] assigned two models to each compound – one with a dominant $K < 0$ interaction, and one with a more modest antiferromagnetic $K$ of similar magnitude to $J_1$, $\Gamma$ and $\Gamma'$. We will refer to these model classes as ‘ferromagnetic’ (FM, –) and ‘antiferromagnetic’ (AFM, +), respectively. Our work follows the same pattern: the top and bottom rows of Fig. 5 show two ostensibly unrelated models with extremely similar powder averages. This correspondence is due to the exact duality transformation identified by Chaloupka and Khalliulin [31]. The $T_1$ transformation as defined in their paper rotates spin space by $\pi$ about the honeycomb plane normal vector $e_{111}$, which in our $xyz$
\[
\begin{array}{|c|c|c|c|c|c|}
\hline
\text{Model} & J & K & \Gamma & \Gamma' & J_3 \\
\hline
\text{tx} & -0.20 & -7.00 & 0.50 & 0.15 & 1.40 \\
\tau_1(tx+) & 0.14 & -7.73 & 0.87 & 0.18 & 1.60 \\
\tau_2(tx-) & -3.50 & 3.20 & -3.00 & 2.00 & 1.60 \\
\tau_3(sx+) & -3.47 & 2.80 & -2.75 & 1.78 & 1.40 \\
\tau_4(sx-) & -1.40 & -10.00 & -0.30 & -0.60 & 0.60 \\
\hline
\end{array}
\]

TABLE II. Dual pairings of best fit parameters.

spin basis may be expressed as the rotation (5):

\[
S_i \mapsto Z S_i ,
\]

\[
Z = \begin{pmatrix}
-\frac{1}{\sqrt{3}} + \frac{2}{\sqrt{3}} & \frac{1}{\sqrt{3}} + \frac{2}{\sqrt{3}} \\
\frac{1}{\sqrt{3}} + \frac{2}{\sqrt{3}} & -\frac{1}{\sqrt{3}} + \frac{2}{\sqrt{3}} \\
\end{pmatrix}.
\]

Note that \(|Z| = 1, Z = Z^T = Z^{-1}\). By applying this transformation to the \(K_\alpha\) matrices defined in Eqs. (2) and (3), one may easily verify that there exist transformed parameters \(\tilde{J}, \tilde{K}, \tilde{\Gamma}, \tilde{\Gamma'}\) such that

\[
K_\alpha(\tilde{J}, \tilde{K}, \tilde{\Gamma}, \tilde{\Gamma'}) = Z K_\alpha(J, K, \Gamma, \Gamma') Z .
\]

This leads to a linear relationship between the original and transformed parameters. Following Ref. [31], we denote this parameter transformation \(\tau_1\). Its matrix representation is presented in Eq. (7).

\[
\begin{pmatrix}
\tilde{J} \\
\tilde{K} \\
\tilde{\Gamma} \\
\tilde{\Gamma'}
\end{pmatrix} =
\begin{pmatrix}
1 + \frac{4}{9} & -\frac{4}{9} & \frac{4}{9} & \frac{4}{9} \\
0 & -\frac{4}{9} & \frac{4}{9} & -\frac{4}{9} \\
0 & -\frac{4}{9} & \frac{4}{9} & -\frac{4}{9} \\
0 & -\frac{4}{9} & \frac{4}{9} & -\frac{4}{9}
\end{pmatrix}
\begin{pmatrix}
J \\
K \\
\Gamma \\
\Gamma'
\end{pmatrix} = \tau_1
\]

(7)

Similarly, \(Z^2 = 1\) implies that any purely isotropic couplings remain fixed under \(\tau_1\), i.e., \(J_i = J_i\) (\(i = 2, 3\)). In the absence of \(K, \Gamma, \Gamma'\), \(J_1\) would also be fixed.

This symmetry implies that two models possessing \(\tau_1\)-equivalent parameters will have indistinguishable magnon excitation spectra. Further, any choice of classical zigzag state is site-wise symmetric under a global \(\pi\) rotation about \(e_{111}\). This symmetry renders the neutron scattering cross section (8) invariant under \(S^{\alpha\beta} \mapsto Z^{\alpha\alpha'} Z^{\beta\beta'} S^{\alpha\beta'}\), implying that the two models are fundamentally indistinguishable by scattering experiments that couple only to the spins. The INS cross section is given by

\[
\frac{d^3\sigma}{d\Omega dE_f} \propto F(Q)^2 S_\perp(Q, \omega)
\]

with the dynamical structure factor

\[
S_\perp(Q, \omega) = \sum_{\alpha\beta} \left( \delta_{\alpha\beta} - \frac{Q^\alpha Q^\beta}{Q^2} \right) S^{\alpha\beta}(Q, \omega)
\]

and its matrix elements

\[
S^{\alpha\beta}(Q, \omega) = \sum_{ij} \int \frac{d\tau}{2\pi} e^{-i\omega \tau} \left\langle S_{-Qj}(0) S_{Qj}(\tau) \right\rangle .
\]

The four inequivalent sites in the zig-zag ground state's magnetic unit cell are indexed by \(i, j\), \(F(Q)\) is a spherically symmetric atomic scattering factor, and \(S_{Qj}(\tau)\) is a Fourier mode of a Heisenberg picture spin operator. We neglect the Debye-Waller factor due to the low temperature.

The symmetry pertains to the spin model, not the linearised spin wave approximation. The two parameter sets are therefore functionally indistinguishable — the antiferromagnetic (+) parameter sets, which would seem to have their weak \(K\) parameters suppressed by large \(J_{1,2,3}, \Gamma, \Gamma'\) terms, have equivalent excitation spectra to models with dominant \(K < 0\). Note that there are two axes in \(J_1, K, \Gamma, \Gamma'\) space corresponding to pure Kitaev models: the obvious \((0, K, 0, K)\) axis, and the axis generated by \(\tau_1(0, K, 0, 0) = K(\frac{3}{4}, -\frac{1}{4}, -\frac{1}{4}, -\frac{3}{4})\) for any \(K \neq 0\). In general, when off-diagonal contributions are sufficiently small a large \(K < 0\) parameter is mapped to a mixture of \(K, \Gamma, \Gamma'\) with \(K\) taking the opposite sign. Tab. II confirms that this is essentially the case with our model pairs, and indeed the visualisation in Fig. 7 confirms that the same is true of most other published models.

Arguably, this makes both parameter sets equally ‘close’ to a Kitaev spin liquid. All models presented in Tab. I (with the notable exceptions of \(t_d\) and \(sb^+\)) either have dominant \(K < 0\), or are \(\tau_1\)-equivalent to a model with dominant \(K\), i.e., close to the hidden Kitaev point \(K^-\) (Fig. 7) which is discussed further below. \(Na_3Co_2SbO_6\) and \(Na_2Co_2TeO_6\) are therefore good candidate materials for field-revealed or strain-revealed quantum spin liquids.

The Hubbard models studied in Refs. [63–65] indicate a robustly ferromagnetic Kitaev interaction over a wide range of parameters. The AFM models arguably all suffer from fine tuning in the \(\Gamma, \Gamma'\) parameters; a change of only a few percent in either parameter opens the gap to many meV. The FM models correspond to a small trigonal-field perturbation of an ideal edge-sharing geometry, and so would seem to be the more likely explanations of scattering in \(Na_2Co_2TeO_6\) and \(Na_3Co_2SbO_6\).

### B. Existence of a gap

Powder spectra from \(Na_2Co_2TeO_6\) [Fig. 1 (a), (c), (e)] exhibit a clear spin-wave mode emanating from \(Q = 0.75\ \text{Å}^{-1}\), corresponding to the \(M\) point of the in-plane Brillouin zone. Our data show a weak spectral weight extending all the way down to the elastic line, which may in principle be evidence of a gapless magnon excitation. However, our best-fit agree with other papers that the spectrum is gapped, though we revise the size of this gap down to \(\sim 1\ \text{meV}\).
Our model’s $M$ point gap is controlled primarily by the parameters $\Gamma$ and $\Gamma'$. As can be seen in Fig. 6(a) and (b), the goodness of fit has a W-shaped valley on either side of the gapless line. Crucially, the truly gapless models correspond to a local maximum while the best fit values of $\Gamma, \Gamma'$ for all models considered correspond to a gap of order $1$ meV. We take this to be strong evidence that Na$_2$Co$_2$TeO$_6$ has a gapped spin-wave spectrum. We attribute the weak excess of spectral weight below $1$ meV in the low temperature data [Fig. 1(c)] to energy broadening by magnon damping. The material's powder averaged magnon spectrum therefore bears a very close resemblance to that of $\alpha$-RuCl$_3$ at zero magnetic field[41, 81].

The 50mK powder spectrum of Na$_2$Co$_2$SbO$_6$ [Fig. 1(b), (d)] tells a different story. There is no evidence of a spin wave mode emanating from the $M$ point, all spectral weight is concentrated near a broad feature at the $\Gamma$ point. Both best-fit models concur that the mode is quadratic with a gap of order $\sim 1$ meV, as can be seen in the powder averaged theory predictions [Fig. 5(b), (d)].

**C. Classical ground state**

We make the ansatz that the material is in a collinear zig-zag phase, featuring ferromagnetic chains oriented normal to the $z$-bond direction. The classical magnetic ground state vector corresponds to the eigenvector of the matrix $M := K_x + K_y - K_z$ of smallest eigenvalue[55]. The Heisenberg interactions shift all eigenvalues uniformly, with no effect on the eigenvector: the eigenvalues and eigenvectors depend only on $K, \Gamma$, and $\Gamma'$, with the explicit forms

\[
\begin{align*}
\mathbf{e}_\perp & \propto (\Gamma, 1, 0), \\
\mathbf{e}_\pm & \propto (2x - 1 \pm \zeta, 2x - 1 \pm \zeta, 4) \quad (\Gamma \neq 0) \\
\delta E & = (2\Gamma - \Gamma - |\Delta\zeta|)/2.
\end{align*}
\]

$\mathbf{e}_\pm$ are defined by analytic continuation when $\Gamma = 0$, excepting the pure-Kitaev special cases. For convenience, we also define the basis vectors from Fig. 3,

\[
\begin{align*}
\mathbf{e}_\parallel &= \frac{1}{\sqrt{6}}(1, 1, -2), \\
\mathbf{e}_{111} &= \frac{1}{\sqrt{3}}(1, 1, 1).
\end{align*}
\]

We have set $\zeta = \text{sgn}(\Gamma)\sqrt{8 + (2x - 1)^2}$ and $x = (K + \Gamma)/\Gamma$. Note that $\lambda_+ > \lambda_-$. When $x = 1$, $\mathbf{e}_\pm$ are aligned with $\mathbf{e}_\parallel$ and $\mathbf{e}_{111}$. Changes in $x$ rotate this orthogonal eigenbasis about $\mathbf{e}_\perp$. The signed rotation angles for various $\Gamma, \Gamma', K$ are shown superimposed on Fig. 7.

Given that a minimal eigenvector remains a minimal eigenvector under the uniform rescaling $M \rightarrow \alpha M, \alpha \in \mathbb{R}_{>0}$, i.e., the zig-zag orientation is not sensitive to the overall energy scale, it is possible to represent all possible spin alignments of zig-zag ground states by looking at the unit sphere in $\Gamma - \Gamma - K$ space. The action of $T_1$ on the theory space can be visualised as mirroring about the plane $K + \Gamma' = \Gamma$. On this plane, the spin orientations must be one of $\mathbf{e}_{111}, \mathbf{e}_\parallel$ and $\mathbf{e}_\perp$, which are clearly eigenvectors of $Z$ with eigenvalues 1, −1 and −1 respectively.

Crucially, the two pure Kitaev points at the north and south poles are inequivalent. This is true even for a classical spin model: $K = -1$ is a degeneracy point between the $\mathbf{e}_\perp$ and $\mathbf{e}_\parallel$, $K = 1$ is deep in the $\mathbf{e}_\parallel$ region. Under $T_1$ they are mapped not to each other but to other points in parameter space, marked $K\pm$, which we call ‘hidden’ Kitaev points. The cluster of AFM Na$_2$Co$_2$TeO$_6$ models near $K^-$ is (approximately) mapped to the cluster near the $K = -1$ pole and vice versa, as are the $\mathbf{e}_\pm$ models (see Fig. 7).

Representation-theoretic analyses of X-ray and neutron diffraction data suggest that the spins in Na$_2$Co$_2$TeO$_6$ are aligned close to $\mathbf{e}_\parallel [67, 68]$, with angular uncertainty of order $\sim 30^\circ$. The Na$_2$Co$_2$TeO$_6$ models are only slightly beyond this uncertainty. It should be emphasised that since these models are found near the critical triple-degenerate Kitaev point at $(0,0,-1)$ in the $(\Gamma, \Gamma', K)$ basis, quantum fluctuations are expected to be large, and higher order magnon interactions may tune the effective zig-zag direction as seen by elastic scattering. For $\mathbf{e}_\perp$, the classical energy difference between the $\mathbf{e}_\perp$ and $\mathbf{e}_\parallel$ phase is of order $\delta E = 0.4$ meV. Working so close to a classical degeneracy, it is to be expected that the classical ground state calculation will be somewhat inaccurate. The established cluster of models $\mathbf{e}_\pm$, $\mathbf{e}_\parallel$.
FIG. 7. Ground-state zig-zag magnetic moment orientations over the sphere \( \Gamma^2 + (\Gamma')^2 + K^2 = 1 \) in \((\Gamma, \Gamma', K)\) space, assuming zigzag ground state stability. The sphere is parameterised using \( \Gamma = \sin(\vartheta) \cos(\varphi) \), \( \Gamma' = \sin(\vartheta) \sin(\varphi) \), \( K = \cos(\vartheta) \). In the \( e^- \) phase, meridians are labeled by the angle of \( e^- \) to \( e_{111} \). The fine black dotted line is the intersection of the \( T_1 \)-invariant plane \( K - \Gamma + \Gamma' = 0 \) with the unit sphere. \( K^+ \) and \( K^- \) indicate the nontrivial 'hidden' Kitaev points \([31]\) generated by \( K^\pm = T_1(0,0,\pm 1) \). Models are defined in Tab. I.

tc\(^\pm\) are therefore loosely compatible with the reported structure. We note in passing that the anomalous td model from Ref. [77] is a special case, occurring on the nodal \( T \) invariant line with a ground state along \( e_{111} \), which is in tension with the experiment.

This argument may go some way to explaining the hourglass feature at 1.7 K in Na\(_2\)Co\(_2\)TeO\(_6\) [Fig. 1(e)] – since \( \delta E/kT \approx 0.4 \), we may expect thermal fluctuations to be large enough for linear spin wave theory to break down. The 0.4 meV penalty to rotate a single (classical) spin from \( e^- \) to \( e_\perp \) is not completely frozen out at the higher temperature, suggesting that some kind of nonlinear spin wave mode could be excited in the vicinity of this energy.

D. Prediction of single crystal results

Fig. 8 shows a comparison between domain averages of the Na\(_2\)Co\(_2\)TeO\(_6\) models \( tx^+ \) and \( tx^- \) and the single crystal data of Ref. [78]. A single crystal will in general consist of a statistical population of three \( C_3 \) related domains corresponding to \( x, y \) and \( z \) zig-zags. An experiment will therefore detect an average over the three \( k \) space paths shown in Fig. 8(c). The paths \( \Gamma \rightarrow \Gamma_2 \) and \( \Gamma \rightarrow \Gamma_3 \) are equivalent under the \( D_2 \) symmetry of the Brillouin zone, implying that an experiment should always detect an even number of modes. However, this is not seen in the experimental data, leading the authors of Ref. [78] to suggest that Na\(_2\)Co\(_2\)TeO\(_6\) undergoes a spontaneous charge transfer that renders 1/4 Co sites spinless, leaving the remainder to follow a vortex-like triple-\( Q \) order. However, our calculations in Fig. 8(a) and (b) show that magnon modes arising from an ensemble of zig-zag orders are equally capable of reproducing the dispersion in Fig. 8(d). Each domain contributes two low-energy magnon modes, resulting in four inequivalent modes. Notably, the doubled modes are the sinusoid-like objects matching the experiment. The missing modes are suppressed by the spin orientation, leading to vanishing spectral weight. While the overall agreement between the single crystal data of Ref. [78] and our theory modeling fitted to our powder sample is surprisingly good, the theoretical prediction is not an exact match. The regions \( Q_\parallel < 0.25, Q_\parallel > 0.75 \) appear too weak in the theoretical prediction, while the crossover at \( Q_\parallel = 0.25 \) is absent in the experiment. The calculations here are done in the linear approximation, so slight deviations when far from the vacuum are expected. In particular, the modes near high density of states at the \( M \) point and the crossover would be expected to have stronger magnon damping,
VII. CONCLUSION

In this paper we have presented results from INS for Na$_3$Co$_2$SbO$_6$ and Na$_3$Co$_2$TeO$_6$ measured down to 50 mK. Analysis of the data and comparison with our theoretical modeling shows that spin waves arising from a zig-zag ground state of an extended Heisenberg–Kitaev model are capable of representing the INS behaviour of Na$_2$Co$_2$TeO$_6$ and Na$_3$Co$_2$SbO$_6$. The fine detail that our experiment provides of the $M$-point excitations of Na$_2$Co$_2$TeO$_6$ has allowed us to refine the existing cluster of parameters.

The failure of naive Heisenberg and XXZ models to reproduce the magnon dispersions of these materials [54] should be taken as a strong indication of the importance of anisotropic spin couplings. In particular, our Na$_3$Co$_2$SbO$_6$ data show a gapped spectrum with no evidence of linear modes emanating from the $M$ point, which Heisenberg-type couplings would require.

We have independently searched for antiferromagnetic and ferromagnetic best-fit models of Na$_2$Co$_2$TeO$_6$ and Na$_3$Co$_2$SbO$_6$, and demonstrated that they are mapped to each other under the exact duality transformation $T_1$ of Ref. [31]. This gives us confidence that our model fitting procedure has converged to a global minimum, or at least as close to it as experimental resolution allows.

Fine detail of the $M$ point of Na$_2$Co$_2$TeO$_6$ gives strong evidence for the existence of a small gap, which we conservatively estimate to be 1.0(5) meV. This gap is consistent with the dispersion measured by the single-crystal experiment of Ref. [78], and the parameters obtained are close to the existing cluster.

Our Na$_3$Co$_2$SbO$_6$ data show an energy minimum at the $M$ point, however practical constraints prevent the direct observation of the $Q = 0$ gap. Our use of 4.69 Å neutrons has given us fine detail as close as reasonably practicable to the $M$ point, and our best fit extrapolations remain consistent with a broad, quadratic mode. This implies substantial non-spontaneous breaking of SU(2) spin symmetry, however the lack of clearly resolvable features in the spectrum means that we cannot categorically exclude models other than the work presented here. The calculations in Fig. 8(e) and (f) show that the predicted magnon dispersion along the $M$ to $M'$ line is clearly observable in a single crystal INS experiment.

Our models of both substances emphasise the dominance of $K$ over other parameters, suggesting that these materials are very close to the Kitaev spin liquid phase. This would validate the $ab$ $initio$ arguments about the nature of 3d$^7$ materials as opposed to the d$^5$ metals Ir and Ru that have dominated searches in the past decade [16, 41, 57, 82], specifically that the tighter orbital confinement suppresses the direct Co-Co exchange responsible for Heisenberg interactions [63, 70, 79]. These models are consistent with $ab$ $initio$ calculations, which unambiguously show that cobaltate honeycombs have a ferromagnetic $K$. We consider the AFM models to be the less likely set of parameters – they have substantial off-
diagonal couplings, corresponding to large trigonal distortions of the honeycomb lattice, and are in some sense fine-tuned.

Current INS experimental data cannot distinguish between AFM and FM models linked by $T_1$ duality. This dichotomy has recently been conclusively resolved in α-RuCl$_3$ by the use of resonant elastic X-ray scattering on a high-quality single crystal [50]. In principle, INS with a symmetry breaking in-plane magnetic field might be an alternative strategy to distinguish the two.

Our best fit models of Na$_2$Co$_2$TeO$_6$ and Na$_3$Co$_2$SbO$_6$ seem to suggest that the substitution of tellurium by antimony only marginally affects the spin exchange physics. Recent experiments have observed that zig-zag order breaks down for Na$_2$Co$_2$TeO$_6$ at large magnetic fields, potentially entering a spin-liquid state [69]. To the authors’ best knowledge, Na$_3$Co$_2$SbO$_6$ has not been probed with a magnetic field to date. Our results suggest the cross-hexamer $J_3$ coupling, largely responsible for stabilising the zig-zag state, may be weaker in Na$_3$Co$_2$SbO$_6$, suggesting a possibly lower critical magnetic field. We therefore suggest that our work be read as further motivation for the study of Na$_2$Co$_2$SbO$_6$ as a field-revealed Kitaev QSL candidate.

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Appendix A: Details of Na$_2$Co$_2$TeO$_6$ broadening function determination

We determined the Lorentzian lifetime broadening phenomenologically, by performing a Voigt profile fit to the available single crystal data.

FIG. 9. Empirical fit of slices from single crystal data in [78] by Voigt functions, each with $\gamma = 0.25\text{ meV}$ and $\sigma = 0.135\text{ meV}$. Note that the $M$ point feature (notated here as $Q = (0, 0.5)$ reciprocal lattice units) shows the strongest broadening, while the higher energy features are sharper.