Magnetic anisotropy of the alkali iridate Na$_2$IrO$_3$ at high magnetic fields: evidence for strong ferromagnetic Kitaev correlations

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The magnetic field response of the Mott-insulating honeycomb iridate Na$_2$IrO$_3$ is investigated using torque magnetometry measurements in magnetic fields up to 60 tesla. A peak-dip structure is observed in the torque response at magnetic fields corresponding to an energy scale close to the zigzag ordering ($\approx 15$ K) temperature. We show using exact diagonalization calculations that such a distinctive signature in the torque response enables us to constrain the effective spin models for this material to ones with dominant ferromagnetic Kitaev and subdominant antiferromagnetic Heisenberg exchange, accompanied by smaller competing interactions. In contrast, alternative models with dominant antiferromagnetic Kitaev interactions do not exhibit such a characteristic peak-dip structure in the transverse magnetization. We further show that at high magnetic fields, long range spin correlation functions decay rapidly, suggesting that above the peak-dip feature, the zigzag ordered phase transitions into a phase with characteristics of a quantum spin liquid. Many of our conclusions are expected to apply more broadly to materials governed by the physics of a honeycomb Kitaev model, such as α-RuCl$_3$.

The alkali iridates A$_2$IrO$_3$ (A=Na,Li) have attracted much theoretical [1–14] and experimental [15–25] attention as promising candidates for realizing the physics of the honeycomb Kitaev model [26,27]. Interactions between the effective $j_{\text{eff}} = \frac{3}{2}$ pseudospins on every site of the two-dimensional hexagonal lattice in these strongly spin-orbit coupled materials, have been described by a dominant Kitaev and other subdominant interactions such as Heisenberg [28] and symmetric off-diagonal exchange [29]. Notwithstanding the great progress made, the question of the sign of the dominant Kitaev interaction still remains open. The importance of the magnetic field response in determining the same has been emphasized in multiple studies recently [31,32]. Here, we address this question by a combination of high-field torque magnetometry measurements and exact diagonalization calculations, which capture quantum effects that might otherwise be missed by classical simulations. We find a distinctive peak-dip structure in the experimental magnetic torque response at high magnetic fields, which we show is uniquely captured by a model with a dominant ferromagnetic Kitaev exchange, but not one with an antiferromagnetic Kitaev counterpart.

Na$_2$IrO$_3$ is a layered Mott insulator with an energy gap $E_g \approx 340$ meV [20] and spin-orbit coupling $\lambda \approx 0.5$ eV [6]. The magnetic susceptibility follows a Curie-Weiss law at high temperatures with $\theta_C \approx -116$ K and an effective Ir moment $\mu_{\text{eff}} = 1.82 \mu_B$ [16–18]. The frustrating effects of strong Kitaev correlations cause the suppression of long range order in this material to a Néel temperature ($T_N \approx 15$ K) far below the Curie temperature [19]. Neutron and X-ray diffraction [16], inelastic neutron scattering (INS) [17] and resonant inelastic X-ray scattering (RIXS) [24] measurements reveal the low temperature ordered phase to be an antiferromagnetic zigzag phase with an ordered moment $\mu_{\text{ord}} \approx 0.2 \mu_B$ [16–18]. The parameter space for these couplings for Na$_2$IrO$_3$ has thus far been constrained using ab-initio computations [3,7,10,11], numerical techniques such as exact diagonalization [5,13,30,33] and classical Monte Carlo simulations [4,8], and degenerate perturbation theory [1,2,5,13,30], as well as experimental investigation [17]. Based on the above data, the simplest model arrived at is a nearest-neighbor model with a dominant antiferromagnetic Kitaev [12,30] and a smaller ferromagnetic Heisenberg exchange. While there is some phenomenological justification for considering such a regime of parameters, quantum chemistry [3] and other ab-initio calculations [10,11] suggest a different model with a dominant ferromagnetic Kitaev and smaller antiferromagnetic Heisenberg exchange. In order to stabilize a zigzag phase within such a model, further neighbor couplings [2,11,15] or additional anisotropic interactions [2] are included. Here we investigate the relevant models by experimental measurements of the finite magnetic-field response of Na$_2$IrO$_3$ and compare our results with exact diagonalization simulations.

A single crystal of Na$_2$IrO$_3$, of dimension $\approx 100 \mu$m on a side, with a much smaller thickness, was mounted on a piezoresistive cantilever and measured on an in-situ rotating stage in pulsed magnetic fields up to 60 T. The torque
response was measured as a function of the magnetic field at various fixed angles (0° ≤ θ ≤ 90°) of the crystalline axis normal to the honeycomb lattice, with respect to the magnetic field axis. A distinctive non-monotonic feature is observed in the magnetic torque response (Fig. 1). A peak in magnetic torque in the vicinity of 30-40 T is followed by a dip in the vicinity of 45-55 T. The peak and dip features are separated by as much as ≈15 T near θ ≈ 45° - 55°, but draw closer together at angles closer to θ ≈ 0° and θ ≈ 90°. In the vicinity of θ ≈ 0° and θ ≈ 90°, the peak and dip features are seen to merge into a single plateau-like feature. This evolution of the signature peak-dip feature as a function of field-angle and magnetic field is shown in Fig. 2 for two different azimuthal orientations (φ = 0°, 90°), where φ is the angle that the crystallographic a axis makes with the axis of rotation of the cantilever. Here θ = 0° corresponds to the alignment of the normal to the honeycomb structure and the magnetic field. For φ = 0° the a axis of the crystal coincides with the axis of rotation of the cantilever and the magnetic field, yielding an angular variation τ ∝ μ₀H²sin[2θ] for the measured torque. Accordingly, the measured magnetic torque vanishes in the vicinity of θ = 90° and reverses sign on either side. The high magnetic field torque response of Na₂IrO₃ was independently measured for two crystals, for three different azimuthal orientations (φ = 0°, 90° and 180°), at a temperature of 1.8 K. In each case, the results were found to be very similar. Data for the second sample is shown in the Supplementary Information. Meanwhile, the isotropic magnetization(magnetometer) measured using an extraction magnetometer in pulsed magnetic fields up to 60 T, and a force magnetometer in steady fields up to 30 T, is found to be largely featureless and to increase linearly with field up to 60 T (Supplementary Information).

We use theoretical modeling of the non-monotonic features we observe in the high field torque response to distinguish between potential microscopic models. Our starting point is the usual spin Hamiltonian [1, 13] with nearest-neighbor Kitaev and Heisenberg interactions:

\[ J_h \sum_{<ij>} \sigma_i^\gamma \sigma_j^\gamma + J_K \sum_{<ij>} \sigma_i^\gamma \sigma_j^\gamma \tag{1} \]

where γ = x, y, z labels an axis in spin space and a bond direction of the honeycomb lattice, and the Hamiltonian is expressed in terms of Pauli matrices \( \sigma_i^\gamma \). **Model A:** This is a nearest-neighbor model with \( J_h < 0 \) and \( J_K > 0 \). **Model B:** In this model, further neighbor antiferromagnetic Heisenberg couplings \( J_2 \) and \( J_3 \) are introduced up to the third nearest neighbor, with \( J_h > 0 \). **Model C:** In this model, bond-dependent nearest-neighbor symmetric off-diagonal terms \( H_{\text{cd}}(\gamma) = \Gamma \sum_{\alpha \neq \beta \neq \gamma} \sum_{(i,j)} (\sigma_i^\alpha \sigma_j^\beta + \sigma_i^\beta \sigma_j^\alpha) \) (where α and β are the two remaining directions apart from the Kitaev bond direction γ) [30] and \( H'_{\text{cd}} = \Gamma' \sum_{\alpha \neq \beta \neq \gamma} \sum_{(i,j)} (\sigma_i^\alpha \sigma_j^\beta + \sigma_i^\beta \sigma_j^\alpha + \sigma_i^\gamma \sigma_j^\gamma + \sigma_i^\gamma \sigma_j^\gamma) \) [30] accounting for trigonal distortions of the oxygen octahedra, are introduced. While variants of the above three models that involve combinations of the above parameters [7, 10] as well as further neighbor Kitaev interactions [4] have also been investigated, here we consider the minimum distinctive models.

Theoretical calculations were performed in the IrO₆ octahedral frame, and subsequently transformed into the laboratory frame using a basis transformation which involves cantilever and crystal frames at intermediate stages (see SI for details). The effect of the applied magnetic field \( H = H_x \) (in the lab frame) on the system is described by \( H_{\text{mag}} = (g) \sum_i \sum_\gamma h_\gamma \sigma_i^\gamma \), with \( g \approx 1.78 \) being the Lande g-factor, assumed to be a constant, and \( H = (h_x, h_y, h_z) \) being the field as expressed in the crystal octahedron frame. We use a hexagonal 24-site cluster [11, 13, 30] with periodic boundary conditions. Exact diagonalization calculations for the ground state energy and eigenvector are performed using a Modified Lanczos algorithm [35] (for details see SI). The code was benchmarked by reproducing the results in [13]. The chosen parameters are further verified to be consistent with the zigzag ground state of Na₂IrO₃ by calculating the structure factors \( S(Q) \) [30, 31] for zigzag, stripy, ferromagnetic and antiferromagnetic ground states (see SI).

The calculated torque responses for the different models are shown in Figures [3] and [4]. Of these three models, only model B and model C reproduce the peak-dip feature in the torque response, unlike model A, which displays a monotonic increase in \( \tau \) with magnetic field. We have performed

![Figure 1. Magnetic torque (\( \tau \)) as a function of magnetic field for different angular orientations (\( \theta \)) and (\( \phi \)) = 90°. A peak dip structure is observed in the magnetic torque, and is seen to evolve with \( \phi \). Individual torque curves have been offset for clarity. (Inset: a crystal on the cantilever with the various coordinate systems: \( XYZ \rightarrow \text{lab frame}, \text{xyz} \rightarrow \text{frame fixed to the cantilever, so that} \ X \text{and} \ x \text{coincide.} \ \theta \text{is the angle that the normal to the crystal makes with the magnetic field and} \tau_X \text{is the torque that is being measured.} \) ]
Figure 2. $\frac{d\tau}{dH}$ as a function of magnetic field and angle ($\theta$) for $\phi = 90^\circ$ (Top) and $\phi = 0^\circ$ (Bottom). The position of the maxima in the torque is indicated by the regular triangles while that of the subsequent minima is marked by the inverted triangles.

Figure 3. Torque as a function of magnetic field (in $\mu_B$ tesla per site) for model B (denoted by $\tau_B$) with parameters $J_6 = 3.6, J_K = -30.0, J_2 = 0.6, J_3 = 1.8$ (in meV) corresponding to antiferromagnetic Heisenberg and ferromagnetic Kitaev correlations along with further neighbor Heisenberg interactions, corresponding to the orientation $\theta = 42^\circ, \phi = 0^\circ$. For this choice of the signs of $J_K$ and $J_6$, the further neighbor interactions are necessary to stabilize a zigzag ground state. The experimental data for this orientation is plotted along with the torque response obtained for this model for comparison.

Figure 4. Torque as a function of magnetic field (in $\mu_B$ tesla per site) for model A (denoted by $\tau_A$) with parameters $J_6 = -4.0, J_K = 21.0$ (in meV) for ferromagnetic Heisenberg and antiferromagnetic Kitaev interactions, corresponding to the orientation $\theta = 36^\circ, \phi = 0^\circ$, and for model C (denoted by $\tau_C$) with parameters $J_6 = 4.0, J_K = -16.0, \Gamma = 2.4, \Gamma' = -3.2$ (in meV) for antiferromagnetic Heisenberg and ferromagnetic Kitaev exchange, corresponding to the same orientation. In model A, while a stable zigzag phase is stabilized with only nearest-neighbor interactions [3, 15], no peak-dip feature appears, unlike experimental observations. In contrast, in model C, where the zigzag phase is stabilized by the introduction of nearest-neighbor anisotropic terms $\Gamma$ and $\Gamma'$ [5], the magnetic field dependence of magnetic torque shows a peak-dip feature corresponding with experiment.

ED simulations for fields up to 300 T for model A (for the same parameters as in Fig. 4), and found a single peak in the torque response at a field slightly lower than 150 T, beyond which the torque decreases with increase in field strength and no further features are observed. We have also considered variants of model A with isotropic $J_2$ and $J_3$ as well as anisotropic $\Gamma$ and $\Gamma'$ terms, and have confirmed that this model does not give a peak-dip feature in its torque response, even with such additional terms present (please refer to Table I in the SI for a summary of the different variants considered, and the corresponding torque curves). This strongly indicates that Na$_2$IrO$_3$ is described by a model dominated by ferromagnetic Kitaev exchange. In model B, the peak-dip feature is observed over a large parameter range in comparison with model C, which is more finely tuned and in particular, requires the presence of a significant $\Gamma' < 0$ term to reproduce the peak-dip feature. In contrast, the presence of significant anisotropy terms in model B does not yield additional peak-dip features, and within this model, the peak-dip survives only for relatively small values of additional anisotropic interactions. The key difference between the models B and C is the latter’s requirement of a significant $\Gamma'$ term, which physically is associated with trigonal distortion in Na$_2$IrO$_3$. Models B and C can thus potentially be distinguished by high magnetic field torque magnetometry measurements on chemically doped Na$_2$IrO$_3$ with various extents of trigonal distortion, which should have an observable effect on the peak-dip feature.
Importantly, the parameter space for obtaining a peak-dip feature in the torque response within model B is much larger than that for obtaining a zigzag ordered ground state, although for Na$_2$IrO$_3$, one needs to be guided by the experimental constraint that the ground state is zigzag ordered. The distinctive peak-dip feature in the torque response thus provides an independent handle for constraining experimental data. We note that classical Monte Carlo simulations were unable to reproduce the peak-dip feature, underlining the importance of quantum effects in this material, as has also been emphasized in the recent literature\cite{32}.

Finally, to probe the nature of the high magnetic field phase into which the zigzag ordered phase transitions above the peak-dip feature, we have computed the evolution of the spin correlation functions with distance for increasing magnetic field values. The extent of decay of the correlation functions with distance tells us about the presence or absence of long range magnetic order in the high field regime. The correlation functions \( C_{ij} = \langle \sigma_i^\alpha \sigma_j^\beta \rangle = \langle \sigma_i^\alpha \sigma_j^\alpha \rangle \) are calculated for a chosen set of neighboring sites in the 24-site cluster, and plotted in Fig. 5 as a function of \( |i−j|/a \) (a being the distance between nearest neighbor sites) for different values of the applied magnetic field. We find that as the magnetic field is increased, the behavior of these correlation functions becomes increasingly similar to a ferromagnetic pure Kitaev model, i.e. the decay of the correlation functions \( C_{ij} \) as a function of \( |i−j|/a \) is much faster at relatively higher values of the applied field, and the amplitude of the oscillation of the correlation functions falls off rapidly with increasing fields, in particular above the zigzag ordering scale. Furthermore, structure factor calculations do not show a crossover from antiferromagnetic zigzag order to a different ordered state at the position of the metamagnetic transition manifested through the peak-dip in the transverse magnetization. The high magnetic field regime beyond the peak-dip feature is therefore suggested to manifest spin-liquid physics in Na$_2$IrO$_3$. Intriguingly, a similar peak-dip feature in the transverse magnetization as a function of magnetic field was observed in \( α−\text{RuCl}_3 \)\cite{37}, where excitations characteristic of a magnetic field-induced spin liquid phase\cite{31,38,41} have also been reported. Our results suggest similar underlying magnetic interactions in \( α−\text{RuCl}_3 \) and Na$_2$IrO$_3$, and the likely relevance of the microscopic models we calculate here to a broader class of spin-orbit coupled honeycomb Kitaev materials. Our results imply that these systems may be tuned to a spin-liquid regime by applying high enough magnetic fields.

After submitting our preprint, we came across a report of \( \text{CuIr}_2\text{O}_4 \)\cite{42} which support a ferromagnetic sign for the Kitaev interaction.

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A. EXPERIMENTAL DETAILS

Crystals of Na$_2$IrO$_3$ about 100 micron along a side were prepared using Na$_2$CO$_3$ slightly in excess [18]. The insulating nature of the sample was confirmed using four-probe resistivity measurements. Magnetization measurements were performed using a Quantum Design SQUID magnetometer up to a field of 5 tesla, and these showed the presence of an antiferromagnetic transition around $T_N \sim 15$ K, below which the system orders into a zigzag phase [16]. A linear Curie-Weiss fit to the high temperature inverse susceptibility data gives an effective moment $\mu_{eff} = 1.67\mu_B$ and Curie temperature $\theta_p = -116$ K. This gives a frustration index $g_N \sim 8$, which is in agreement with the results obtained from other groups.

The torque ($\tau = m \times B$) was measured using a PRC 120 piezoresistive cantilever at the pulsed field facility at the National High Magnetic Field Laboratory, Los Alamos. The crystal was mounted on the cantilever with vacuum grease. The cantilever assembly was made of G-10 and capable of rotation about the magnetic field of the crystal (which is the nominal c-axis) and the face of the crystal (which is the nominal c-axis) and the magnetic field of the crystal (which is the nominal c-axis).
Figure 6. Torque as a function of magnetic field ($H$) measured independently for a second crystal corresponding to two different in-plane orientations $\phi$ separated by 90$^\circ$, is shown in (a) and (b), and is again found to show nonmonotonous behavior for a range of orientations. The corresponding plots for $\frac{d\tau}{dH}$ as a function of $H$ are shown in (c) and (d). $\theta$ is the angle that the normal to the crystal makes with the magnetic field, and is defined in the main text.

Figure 7. Torque ($\tau$) as a function of magnetic field for different angular orientations ($\theta$) and $\phi = 0^\circ$ corresponding to the first crystal as mentioned in the main text.

magnetic field $H$. For each value of $\theta$ the torque response was measured for the increasing and decreasing cycles of the pulse which had a high degree of overlap ruling out significant magneto-caloric effects which might become evident in pulsed field measurements. Further measurements were performed for various in-planar orientations $\phi$ of the crystal. The torque response for a second crystal from an independently grown batch, mentioned in the main text, is shown in Fig. 7.

A2. NUMERICAL SETUP AND EXACT DIAGONALIZATION ALGORITHM

The N lattice sites were numbered 0,1,2...N-1 (for N=24), and specific pairs of these sites were identified as ‘bonds’ or ‘links’, of type $x$, $y$ or $z$. Every site has a spin with two possible states $|1\rangle$ or $|0\rangle$. The system then has $2^N$ configurations or underlying basis states, where each configuration is denoted by $|s_{N-1}s_{N-2}...s_0\rangle$ with $s_i = 0,1$. Corresponding to such a set of binary numbers, we have a decimal equivalent given by $|s_{N-1}2^{N-1} + s_{N-2}2^{N-2} + ...s_02^0\rangle$. The basis vectors were thus denoted as $|0\rangle$, $|1\rangle$,...$|2^{N-1}\rangle$. An arbitrary state vector $|\psi\rangle$ can be expanded in terms of these basis vectors as $|\psi\rangle = \sum_{i=0}^{2^{N-1}} a_i |i\rangle$. The ground state $|\Psi_0\rangle$ for this Hamiltonian $H_0$ was determined using the Modified Lanczos algorithm.

The Modified Lanczos algorithm [35] requires the initial selection of a trial vector $|\psi_0\rangle$ (constructed using a random number generator in our case) which should have a nonzero projection on the true ground state of the system in order for the algorithm to converge properly. A normalized state $|\psi_1\rangle$, orthogonal to $|\psi_0\rangle$, is defined as

$$|\psi_1\rangle = \frac{H_0|\psi_0\rangle - <H_0>|\psi_0\rangle}{\sqrt{<H_0^2> - <H_0>^2}}$$

(2)

In the basis $\{|\psi_0\rangle, |\psi_1\rangle\}$, $H_0$ has a 2x2 representation which is easily diagonalized. Its lowest eigenvalue and corresponding eigenvector are better approximations to the true ground state energy and wavefunction than the quantities $<H_0>$ and $|\psi_0\rangle$ considered initially. The improved energy and wavefunction are given by

$$\epsilon = <H_0> + b \alpha$$

(3)

and

$$|\psi_0\rangle = \frac{|\psi_0\rangle + \alpha |\psi_1\rangle}{\sqrt{1 + \alpha^2}}$$

(4)

where $b = \sqrt{<H_0^2> - <H_0>^2}$, $f = \frac{<H_0> - 3H_0^2 + H_0^3}{2b^2}$ and $\alpha = f - \sqrt{1 + f^2}$. The method can be iterated by considering $|\psi_0\rangle$ as a new trial vector and repeating the above steps. The Modified Lanczos method helps in obtaining a reasonably good approximation to the actual ground state of the system while storing only
three vectors, $\psi_0$, $H_0\psi_0$ and $H^2_0\psi_0$ rather than the entire Hamiltonian in the spin basis representation. This is especially advantageous as the number of basis vectors increases rapidly with the number of spin sites. In the regular Lanczos algorithm, the matrix is first reduced to a tridiagonal form before computing the ground state eigenvector. However, there can be issues with the convergence to the true ground state because of loss of orthogonality among the vectors. This is circumvented in this algorithm as orthogonality is enforced at each and every step of the iteration.

After the determination of the ground state $\Psi_0$ to a reasonable approximation, the magnetization $\vec{m}|\psi_0>$ was obtained in this state with components $(m_x, m_y, m_z)$, where $m_{\gamma} = \langle \psi | \sum_{i=0}^{N-1} \sigma^\gamma_i | \psi >$, and was transformed to the lab frame from the octahedral frame, the components in the lab frame being $(m_X, m_Y, m_Z)$. Finally, in the lab frame, torque $\Gamma_X = m_Y H$.

### A3. COORDINATE SYSTEM TRANSFORMATIONS

For our exact diagonalization calculations, we have transformed the external magnetic field from the laboratory frame to the IrO$_6$ octahedral frame by defining intermediate crystal and cantilever axes, and transformed the calculated magnetization back from this frame to the lab frame. We explain the transformations used in the following:

#### Notations:

**Laboratory axes:** $\hat{X}, \hat{Y}, \hat{Z}$
**Cantilever axes:** $\hat{x}, \hat{y}, \hat{z}$
**Crystal axes:** $\hat{a}, \hat{b}, \hat{c}$
**Octahedral axes:** $\hat{p}, \hat{q}, \hat{r}$

### Laboratory to cantilever axes:

The lab $\hat{X}$-axis and the cantilever $\hat{x}$-axis are always coincident. Let $\theta$ be the angle between the $\hat{Z}$ and $\hat{z}$ axes. We have

\[ |\hat{x}\hat{y}\hat{z} > = M_{\text{Lab to Cantilever}} |\hat{X}\hat{Y}\hat{Z} > \]

\[ |\hat{X}\hat{Y}\hat{Z} > = L_{\text{Cantilever to Lab}} |\hat{x}\hat{y}\hat{z} > \]

where

\[
M_{\text{Lab to Cantilever}} = \begin{pmatrix}
1 & 0 & 0 \\
0 & \cos \theta & -\sin \theta \\
0 & \sin \theta & \cos \theta 
\end{pmatrix}
\]

\[
L_{\text{Cantilever to Lab}} = \begin{pmatrix}
1 & 0 & 0 \\
0 & \cos \theta & \sin \theta \\
0 & -\sin \theta & \cos \theta 
\end{pmatrix}
\]

#### Cantilever to crystal axes:

The honeycomb layer formed by the Ir atoms resides on the crystallographic $ab$ plane. Let the $\hat{a}$-axis of the crystal make an angle $\phi$ with the $\hat{x}$-axis of the cantilever. Then,

\[ |\hat{a}\hat{b}\hat{c} > = M_{\text{Cantilever to Crystal}} |\hat{x}\hat{y}\hat{z} > \]

\[ |\hat{x}\hat{y}\hat{z} > = L_{\text{Crystal to Cantilever}} |\hat{a}\hat{b}\hat{c} > \]

\[
M_{\text{Cantilever to Crystal}} = \begin{pmatrix}
\cos \phi & \sin \phi & 0 \\
-\sin \phi & \cos \phi & 0 \\
0 & 0 & 1
\end{pmatrix}
\]

\[
L_{\text{Crystal to Cantilever}} = \begin{pmatrix}
\cos \phi & -\sin \phi & 0 \\
\sin \phi & \cos \phi & 0 \\
0 & 0 & 1
\end{pmatrix}
\]

#### Crystal to octahedral axes:

Since the $[111]$ direction in the octahedral frame is perpendicular to the honeycomb lattice, the unit vectors are related as follows:

\[ \hat{e} = \frac{\hat{p} + \hat{q} + \hat{r}}{\sqrt{3}} \]

\[ \hat{b} = -\frac{\hat{p} + \hat{q}}{\sqrt{2}} \]

\[ \hat{a} = \frac{\hat{p} + \hat{q} - 2\hat{r}}{\sqrt{6}} \]

Then,

\[ |\hat{p}\hat{q}\hat{r} > = M_{\text{Crystal to Octahedra}} |\hat{a}\hat{b}\hat{c} > \]

\[ |\hat{a}\hat{b}\hat{c} > = L_{\text{Octahedral to Crystal}} |\hat{p}\hat{q}\hat{r} > \]

\[
M_{\text{Crystal to Octahedral}} = \begin{pmatrix}
\frac{1}{\sqrt{6}} & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{3}} \\
\frac{1}{\sqrt{6}} & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{3}} \\
-\frac{1}{\sqrt{3}} & 0 & \frac{1}{\sqrt{3}}
\end{pmatrix}
\]

\[
L_{\text{Octahedra to Crystal}} = \begin{pmatrix}
\frac{1}{\sqrt{3}} & \frac{1}{\sqrt{6}} & -\frac{1}{\sqrt{3}} \\
\frac{1}{\sqrt{3}} & \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{3}} \\
\frac{1}{\sqrt{3}} & \frac{1}{\sqrt{6}} & \frac{1}{\sqrt{3}}
\end{pmatrix}
\]
Lab to octahedral and octahedral to lab frame:

Let the components of the magnetic field be \((0, 0, H)\) in the lab frame and \((h_p, h_q, h_r)\) in the octahedral frame. Then

\[
|hpqHr| = M_{\text{Crystal}\to\text{Octa}}M_{\text{Canti}\to\text{Crystal}}M_{\text{Lab}\to\text{Canti}}|00H|
\]

which finally gives us

\[
h_p = (-\frac{1}{\sqrt{6}} \sin \theta \sin \phi + \frac{1}{\sqrt{2}} \sin \theta \cos \phi + \frac{1}{\sqrt{3}} \cos \theta)H
\]

\[
h_q = (-\frac{1}{\sqrt{6}} \sin \theta \sin \phi - \frac{1}{\sqrt{2}} \sin \theta \cos \phi + \frac{1}{\sqrt{3}} \cos \theta)H
\]

\[
h_r = (\sqrt{\frac{2}{3}} \sin \theta \sin \phi + \frac{1}{\sqrt{3}} \cos \theta)H
\]

Let the components of the magnetization vector \(\vec{m}\) be \((m_X, m_Y, m_Z)\) in the lab frame and \((m_p, m_q, m_r)\) in the octahedral frame. Then,

\[
|m_Xm_Ym_Z| = L_{\text{Canti}\to\text{Lab}}L_{\text{Crystal}\to\text{Canti}}L_{\text{Octa}\to\text{Crystal}}|mpqmrmr|
\]

from where we find

\[
m_Z = (-\frac{m_p}{\sqrt{6}} - \frac{m_q}{\sqrt{6}} + m_r \sqrt{\frac{2}{3}}) \sin \theta \sin \phi + \frac{m_p - m_q}{\sqrt{2}} \sin \theta \cos \phi + \frac{(m_p + m_q + m_r)}{\sqrt{3}} \cos \theta
\]

and

\[
m_Y = \frac{m_p}{\sqrt{6}} + \frac{m_q}{\sqrt{6}} - m_r \sqrt{\frac{2}{3}} \cos \theta \sin \phi + \frac{m_p - m_q}{\sqrt{2}} \cos \theta \cos \phi + \frac{(m_p + m_q + m_r)}{\sqrt{3}} \sin \theta
\]

A4. STRUCTURE FACTOR CALCULATIONS

To determine different phases of the system in the presence of an applied magnetic field, one needs to calculate the adapted structure factors acting as order parameters \([5]\). The corresponding dominant order wavevectors \(\vec{Q} = \vec{Q}_{max}\) characterize the nature of the magnetic ordering in various field regimes. The static structure factors \(S(\vec{Q})\) for different spin configurations are given by

\[
S_{\text{zigzag}}^\gamma = \frac{1}{N^2} \sum_{r,r',\beta,\beta'} \exp[i\vec{Q}\gamma(\vec{r}' - \vec{r})] \nu_{\beta,\beta'}(\sigma_{r,\beta',\beta'}^\gamma - \sum_{\gamma} \sigma_{r,\beta',\beta'}^\gamma > \sigma_{r,\beta',\beta'}^\gamma) \quad (5)
\]

\[
S_{\text{Neel}} = \frac{1}{N^2} \sum_{r,r',\beta,\beta'} \nu_{\beta,\beta'}(\sigma_{r,\beta',\beta'}^\gamma - \sum_{\gamma} \sigma_{r,\beta',\beta'}^\gamma > \sigma_{r,\beta',\beta'}^\gamma) \quad (6)
\]

\[
S_{\text{FM}} = \frac{1}{N^2} \sum_{r,r',\beta,\beta'} (\sigma_{r,\beta',\beta'}^\gamma - \sum_{\gamma} \sigma_{r,\beta',\beta'}^\gamma > \sigma_{r,\beta',\beta'}^\gamma) \quad (7)
\]

\[
S_{\text{stripy}}^\gamma = \frac{1}{N^2} \sum_{r,r',\beta,\beta'} \exp[i\vec{Q}\gamma(\vec{r}' - \vec{r})](\sigma_{r,\beta',\beta'}^\gamma - \sum_{\gamma} \sigma_{r,\beta',\beta'}^\gamma > \sigma_{r,\beta',\beta'}^\gamma) \quad (8)
\]

where each site is labeled by an index \(i\) and a position in the unit cell \(\vec{r}'_i, \beta\) denotes the sublattice index (\(\beta = A, B\), and
Figure 9. Evolution of structure factors for different ordered phases as a function of the field for (a) $J_h = 3.2$, $J_K = -12$, $J_2 = 4$, $J_3 = 2$(in meV), (b) $J_h = 1.6$, $J_K = -16.0$, $J_2 = 1.2$ and $J_3 = 0.8$(in meV), and (c) $J_h = 4.0$, $J_K = -16.0$, $\Gamma = 2.4$ and $\Gamma' = -3.2$(in meV).

A5. MAGNETIZATION AS A FUNCTION OF FIELD

The isotropic magnetization($m_Z$) was measured using an extraction magnetometer in pulsed magnetic fields up to 60 T and calibrated to obtain $m_Z$ per site using force magnetometry measurements in steady magnetic fields, and magnetization measurements in a SQUID magnetometer. It is found to be largely featureless and increases linearly with field up to 60 T. We have determined the behavior of $m_Z$ per site numerically for different relevant models and the results, along with the experimental curves, are shown in Fig. 10.

Figure 10. (a) Isotropic magnetization measured using an extraction magnetometer in pulsed magnetic fields, and using a force magnetometer in DC fields, shows no features up to 60 T, calibration is performed using magnetization measurements on a pellet of sodium iridate in a SQUID magnetometer, (b) Isotropic magnetization $m_Z$ (in $\mu_B$ per atom) calculated as a function of field, for model B with $J_h = 2.4$, $J_K = -12.0$, $J_2 = 1.6$, $J_3 = 1.2$(in meV) for the orientation $\theta = 18^\circ$, $\phi = 90^\circ$, and for model C with $J_h = 4.0$, $J_K = -16.0$, $\Gamma = 2.4$ and $\Gamma' = -3.2$(in meV), for the orientation $\theta = 36^\circ$, $\phi = 0^\circ$. 

$\gamma = x, y$ or $z$. The contribution to the structure factors coming from the alignment of the spins with the field direction has explicitly been deducted in this definition. The structure factors for the four different phases are plotted as a function of field for different models in Fig. 9 which clearly shows that AFM zigzag is the dominant spin configuration in all cases.
A robust peak-dip feature is observed for a wide range of orientations and different polar and azimuthal angles as indicated in the figures. A robust peak-dip feature is observed for a wide range of orientations in model B (see also the contourplot below in Fig. 19).

Figure 11. Calculated values of torque for model B with parameters $J_h = 2.4$, $J_k = -12.0$, $J_2 = 1.6$, $J_3 = 1.2$ (in meV) for different polar and azimuthal angles as indicated in the figures. A robust peak-dip feature is observed for a wide range of orientations in model B (see also the contourplot below in Fig. 19).

Figure 12. Calculated values of torque for model C with parameters $J_h = 4.0$, $J_k = -16.0$, $\Gamma = 2.4$, $\Gamma' = -3.2$ (meV) for different polar and azimuthal angles as indicated in the figure. In (b) the peak-dip feature is present but is shallower than that observed in (a).

Figure 13. Calculated values of the torque for models with $J_h = -8.0$, $J_k = 40.0$ (meV), for the orientation $\theta = 36^\circ$, $\phi = 0^\circ$, with $\Gamma$ and $\Gamma'$ values as indicated in the figures. We observe that additional $\Gamma$ and $\Gamma'$ terms do not give rise to any peak-dip features in the torque response.

Figure 14. Here we demonstrate the calculated torque response for different models with a ferromagnetic Heisenberg and antiferromagnetic Kitaev interaction with further neighbour Heisenberg interactions. (a) and (b) correspond to $J_h = -4.0$, $J_k = 21.0$ (meV) for the orientation $\theta = 48^\circ$, $\phi = 90^\circ$, for $J_2$ and $J_3$ interactions as indicated in the figure. We observe that further neighbour interactions $J_2$ and $J_3$ do not give rise to any peak-dip features in the torque response.

A6. EXTENDED MODELLING

A. Observation of peak dip feature for some more orientations:

Here we consider models B and C of the main text and show the existence of the peak-dip feature in the torque response for different combinations of polar and azimuthal angles. This is illustrated in figures 11 and 12 for models B and C respectively.

B. General absence of a peak-dip feature in models with an antiferromagnetic Kitaev interaction ($J_K > 0$):

The purpose of this section is to show that models with an antiferromagnetic sign of the Kitaev coupling tuned to a zigzag ground state by a variety of subleading interactions are generally unable to produce the peak-dip feature in the torque that is observed in experiment. Figures 13 and 16 illustrate this for models with additional $\Gamma$ and $\Gamma'$ interactions, and figures 14 and 15 for models with various combinations of antiferromagnetic as well as ferromagnetic further neighbour interactions $J_2$ and $J_3$. The different combinations of parameters considered is summarized in Table I.

C. Peak-dip features in models with a ferromagnetic Kitaev interaction ($J_K < 0$):

Here we consider the torque response for models with a ferromagnetic Kitaev interaction where the nearest neighbour Heisenberg interaction is also ferromagnetic, with an additional anisotropic $\Gamma$ and/or isotropic $J_3$ interaction. Such models have been proposed in the literature for the related Kitaev material $\alpha$-RuCl$_3$ which also has a zigzag ground state. We did not see a peak-dip feature in such models; how-
Figure 16. Here we demonstrate the calculated torque response for different models with a ferromagnetic Heisenberg and antiferromagnetic Kitaev interaction with additional anisotropic parameters. (a) corresponds to $J_h = -1.84$, $J_K = 3.2$ (meV) for the orientation $\theta = 60^\circ$, $\phi = 90^\circ$ with an additional $\Gamma$ term taking values indicated in the figure. (b) shows the torque response for two sets of parameters with $J_h < 0$, $J_K > 0$ and $\Gamma > 0$ at different orientations of the field. Here Set 1 corresponds to $J_h = -1.84$, $J_K = 3.2$ and $\Gamma = 1.528$ (meV) for $\theta = 36^\circ$ and $\phi = 0^\circ$, and Set 2 corresponds to $J_h = -12.0$, $J_K = 17.0$, and $\Gamma = 12.0$ (meV) for $\theta = 48^\circ$ and $\phi = 90^\circ$.

Figure 17. The figure (a) shows the calculated torque response for models with both the Kitaev and Heisenberg interactions ferromagnetic. Here Set 1 corresponds to $J_h = -1.0$, $J_h = -8.0$ and $\Gamma = 4.0$ (in meV) for $\theta = 48^\circ$ and $\phi = 90^\circ$ while Set 2 corresponds to $J_h = -1.7$, $J_K = -6.6$, $J_3 = 2.7$ and $\Gamma = 6.6$ (in meV) for $\theta = 18^\circ$ and $\phi = 90^\circ$. The figure (b) shows the calculated torque response for models with $|\Gamma| > |J_K|$ with an antiferromagnetic Kitaev and ferromagnetic Heisenberg interaction. Here, Set 1 corresponds to $J_h = -0.98$, $J_K = 1.17$ and $\Gamma = 3.69$ (in meV) while Set 2 corresponds to $J_h = -1.99$, $J_K = 1.99$ and $\Gamma = 2.83$ (in meV), both for $\theta = 60^\circ$ and $\phi = 90^\circ$. Clearly, none of the parameter sets with an antiferromagnetic Kitaev interaction give rise to any peak-dip features in the torque response.

ever, there is a slight flattening of the torque response curve at intermediate fields. This is illustrated in Fig. 17(a).

D. Peak-dip features in the absence of zigzag order:

Here we demonstrate that models with a ferromagnetic Kitaev $J_K$, antiferromagnetic Heisenberg $J_h$ and additional antiferromagnetic further-neighbour interactions $J_2$ and $J_3$ can give rise to peak-dip features even in the absence of zigzag order in the ground state. The presence of the peak-dip features thus provides an independent handle which can distinguish the response of such models from those with an antiferromagnetic Kitaev interaction. This is illustrated in Fig. 18 where we find that the peak-dip feature is observed even for those combinations of parameters where either $J_2$ or $J_3$ vanishes, or $J_2$, $J_3$ are both small as compared to the nearest-neighbour interactions $J_h$ and $J_K$. Such combinations of parameters often do not give rise to a zigzag ordered ground state, and cannot be used to represent Na$_2$IrO$_3$, but they still do give rise to prominent peak-dip features in the torque. Table 1 summarizes the different models we have considered, with a ferromagnetic Kitaev interaction and additional sub-dominant terms.
Figure 20. The figure shows the torque response corresponding to steady field measurements for $\theta = -20^\circ$ and $\phi = 0^\circ$ at various temperatures in the range 5 K-17.5 K. A shallow peak-dip feature, close to around 20 T, is observed at low temperatures and no longer discernible at temperatures beyond about 12.5 K. This shows that the peak-dip feature is associated with a transition from the zigzag ordered ground state to a state with a significantly different torque response.

We find that for model B, a peak-dip feature is robustly observed for all orientations $\theta$ for a given value of the azimuthal angle $\phi$, and the position as well as the shape of the peak-dip evolves as a function of $\theta$, as expected from the experiment. Theoretically, the transverse magnetization (torque) response could well be negative, and in such cases, we plot $-\frac{d\tau}{dH}$ instead, as we are not interested in the absolute value of the torque obtained, but only in the position of the peak-dip, which is indicated by the regions where the first derivative of the torque changes sign. Fig.19 illustrates our results, and it is clear that although there is qualitative agreement with the experimental results, unlike the actual data, the distance between the peak and the dip, i.e. the width of the region of nonmonotonicity increases at extreme values of $\theta$.

| Model A variant | $\Gamma$ | $\Gamma'$ | $J_2$ | $J_3$ | Ref.(if any) |
|-----------------|---------|---------|------|------|-------------|
| 1. With $\Gamma$, $\Gamma'$ | + | - | × | × | [5] |
| 2. With $J_2$, $J_3$ | × | × | + | + | [5] |
| 3. With $\Gamma'$ | + | × | × | × | [32,30] |

Table I. For model A ($J_K < 0$, $J_K > 0$), introduction of various additional terms $\Gamma, \Gamma'$, $J_2$ and $J_3$, with a zigzag ground state, does not reveal any peak-dip features. The + and - indicate the sign of the coupling and × denotes the absence of the corresponding coupling. Thus, for a wide variety of parameters, antiferromagnetic Kitaev couplings do not produce the observed peak-dip feature.

E. Evolution of the peak-dip feature as a function of polar angle $\theta$ and field $H$:

Here we discuss the evolution of the torque response for model B as a function of the polar angle $\theta$ and field value $H$, by presenting a contourplot of the first derivative of the torque, $\frac{d\tau}{dH}$ (for the torque $\tau$ and field $H$), and compare our results with the experimental data in Fig.2 of the main text.

| $J_1$ | $J_2$ | $J_3$ | $\Gamma$ | $\Gamma'$ | Peak-dip(present/absent) |
|-------|------|------|-------|--------|------------------|
| Model B | + | + | + | × | × | Yes |
| + | + | × | × | × | Yes |
| + | × | + | × | × | Yes |
| Model C | + | × | × | + | - | Yes |
| Ref.[32] | - | × | + | + | × | No |
| - | × | + | + | × | No |

Table II. For models with $J_K < 0$ (model B, model C as well as other models with various anisotropic and Heisenberg interactions), with a zigzag ground state, the peak-dip feature may or may not be present. The + and - indicate the sign of the coupling and × denotes the absence of the corresponding coupling. The last two sets of parameters have been suggested in Ref. [32] and correspond to a ferromagnetic Heisenberg interaction. The remaining ones correspond to an antiferromagnetic Heisenberg interaction. Clearly, the presence of zigzag order in models with $J_K < 0$ does not necessarily produce the peak-dip feature. Thus, the peak-dip feature is an independent tool to constrain the parameter space of possible effective Hamiltonians.

A7: TEMPERATURE-DEPENDENCE OF THE TORQUE RESPONSE

The torque response was measured capacitively for different temperatures in the range 5 K-17.5 K, at steady fields up to 30 T for the orientation $\theta = -20^\circ$, $\phi = 0^\circ$. A peak-dip feature observed close to about 20 T at low temperatures becomes indiscernible beyond a temperature of about 12.5 K. This is illustrated in Fig.20. The exact detection of the temperature at which this feature disappears is limited by the resolution of the measurement, as well as temperature resolution very close to the zigzag ordering temperature. Our results do however establish that the peak-dip feature is present only below the zigzag ordering temperature and is therefore related to a transition from the long-range ordered ground state.