Scale-invariance of a Spin Liquid in High Magnetic Fields

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(Dated: January 29, 2019)
Among strongly correlated states of matter, spin liquids stand as one of the best understood theoretically but the least understood experimentally. In RuCl$_3$, inelastic neutron scattering and Raman spectroscopy reveal a continuum of excitations that persist to high temperature, in accordance with Kitaev’s prediction for a spin liquid state on a honeycomb lattice. However, the effects of high magnetic fields—comparable to the spin exchange energy scale—have not been explored. Here we report measurements of the magnetotropic coefficient—the second derivative of the free energy with respect to magnetic field orientation—over a wide range of magnetic fields and temperatures. Unlike conventional spin systems, which polarize and become isotropic at high fields, the magnetization in RuCl$_3$ shows no signs of saturation up to 65 Tesla. Furthermore, temperature and magnetic field compete to determine the magnetic response in a way that is independent of the large exchange energy. This scale-invariant magnetic response is a new signature of the spin-liquid state that goes beyond current predictions of the Kitaev model.

I. INTRODUCTION

In the search for quantum spin liquids, Kitaev’s model$^1$ has directed extreme interest$^{2–11}$ towards honeycomb networks of transition-metal spins that interact via edge-shared octahedra$^{12,13}$. These systems exhibit the spin-anisotropic exchange interactions required by the Kitaev model$^{14,15}$, and consequently are expected to host a spin liquid ground state with a continuum of fractionalized excitations. The recent discovery of fractionalized excitations in RuCl$_3$ has provided compelling evidence for some aspects of the Kitaev spin liquid in real system$^{6–8}$. One critical observation is that this continuum of excitations persists up to a temperature scale that is comparable to the anisotropic exchange energy ($J_\alpha \sim 150$ K)$^{16}$. This is a direct consequence of the fact that the Kitaev model has only one energy scale—the anisotropic exchange $J_\alpha$. This implies that the spin liquid should also persist in a magnetic field that introduces a large Zeeman energy of order 150 Tesla, offering the opportunity to study the response of a spin liquid over a broad magnetic field range.
We use a newly-developed experimental technique – resonant torsion magnetometry – to measure the magnetic anisotropy of RuCl$_3$ as a function of field, field-orientation, and temperature$^{17}$. Our measurements up to 65 T reveal an unusual linear-in-field dependence of the magnetic anisotropy, accompanied by an anomalous angle dependence, that persists over the entire field-temperature phase diagram and rules out a conventional paramagnetic state$^{18,19}$. While the robustness of this behavior across the entire phase diagram attests to a large underlying $J_\alpha$, the measured magnetic anisotropy is entirely controlled by the thermal and magnetic energy scales. This emergent “scale-invariant” behavior – where $J_\alpha$ does not show up as crossover scale in temperature or magnetic field – indicates a new characteristic of the spin liquid state in RuCl$_3$.

All candidate materials (including (Na,Li)$_2$IrO$_3$ and RuCl$_3$)$^{14,20–22}$ deviate from the ideal Kitaev model in several critical ways: they have anisotropic $g$-factors; exchange interactions in channels other than the spin-anisotropic one; and inter-layer interactions. This motivates our complete angular study to identify the region over which the scale-invariance persists. Furthermore, RuCl$_3$ orders antiferromagnetically below $T_N \approx 7$ K$^{23}$, with a critical magnetic field of $B_c \approx 10$ T when applied in the honeycomb planes$^{24}$. This antiferromagnetic boundary in the temperature-field plane is highly anisotropic, reaching fields greater than 60 T when the magnetic field is perpendicular to the honeycomb planes$^{25}$. We fully characterize this phase boundary, and explore the field-temperature competition where $B_c$ is low and the field-range for accessing the spin liquid state is maximized.

II. RESULTS

We use resonant torsion magnetometry to study the magnetic anisotropy of RuCl$_3$$^{17}$. This technique measures the magnetotropic coefficient $k = \partial^2 F / \partial \theta^2$, where $F$ is the free energy and $\theta$ is the magnetic field orientation. The magnetotropic coefficient represents a material’s magnetic “rigidity” with respect to rotation in a magnetic field. It is detected as a shift in the natural frequency of a freely suspended cantilever onto which the sample is mounted$^{17}$. 
FIG. 1. A) Field scans of the magnetotropic coefficient $k$ at a base temperature of 1.3 K for several field orientations $\theta$, defined between the c-axis and the applied magnetic field. For field oriented near the honeycomb planes ($\theta \approx 90^\circ$), the magnetotropic coefficient monotonically increases up to 34.5 T. The maximum frequency shift (at all fields) occurs when magnetic field lies in the honeycomb plane, suggesting a magnetically easy axis (panel B). As field is rotated out of the plane, a sharp jump in the magnetotropic coefficient is observed as the second-order AFM phase boundary is traversed with increasing field. B) Angle scans of the magnetotropic coefficient, obtained by continuous rotation of the sample, in fixed magnetic fields ranging from 7.5 to 17.5 T. At 7.5 T, the magnetotropic coefficient (the angular derivative of torque) follows the $\cos 2\theta$ angle dependence expected in the linear response regime $M_i = \chi_{ij} H_j$. The linear regime allows us to define the field orientations with respect to the principal directions of the magnetic susceptibility tensor. Additionally, it allows calibration between the measured frequency shift (left axis in panel A) and the absolute value of the magnetotropic coefficient (right axis). This is important because the absolute frequency at zero field depends on the elastic properties and the geometry of the cantilever, as well as the position of the sample on the cantilever. At higher magnetic fields, entry into and out of the AFM phase is marked by the sharp jump. Together with A, these features outline the highly-anisotropic nature of the AFM phase (inset of C). C) The AFM transition field as a function of angle at base temperature, as determined by the data in A & B. The shift of the maximum and minimum in the transition field from 0 and 90\(^\circ\), respectively, is a result of the monoclinic crystal structure\textsuperscript{25}, which allows the principal magnetic axes to freely rotate with magnetic field and temperature (see SI).
Figure 1 shows the magnetic field and field orientation evolution of the magnetotropic coefficient of RuCl\textsubscript{3} at 1.3 K. A sharp jump in the magnetotropic coefficient outlines the anisotropic boundary of the AFM phase in the magnetic field, temperature, and field orientation phase space. This jump, both in magnetic field scans (panel A) and field rotation scans (panel B), is always down upon entry into the ordered state as required by Le Chatelier’s principle\textsuperscript{26}. Similar measurements allow a complete mapping of the AFM phase at higher temperatures (inset of Figure 1C and SI). The AFM transition field $B_c$ reaches a minimum of $\sim 10$ T when the field is applied in the honeycomb plane ($\theta = 90^\circ$ in Figure 1) and has a weak in-plane field orientation dependence (see SI). $B_c$ quickly increases beyond the field range of 35 T as magnetic field is rotated toward the direction perpendicular to the honeycomb planes (Figure 1C). The large sensitivity of this technique to magnetic anisotropy allows for the measurement of $\sim 10$ ng crystals of RuCl\textsubscript{3} (roughly $10^6$ times smaller than those used in other techniques). In previous work, multiple magnetic transitions were observed in the vicinity of the AFM phase boundary, which were shown to be due to additional domains of AB stacking or multiple monoclinic domains\textsuperscript{23,24,27}. Our measurements reveal a single magnetic phase transition (Figure 1) that divides the entire field-temperature phase diagram into only two regions: the low-field and low-temperature AFM state (inset of Figure 1c) and a spin-liquid state that sets in at the suppression of AFM and persists up to the highest magnetic fields.

For a wide range of field orientations near the honeycomb planes (near $90^\circ$ in Figure 1A), we observe a linear-in-field dependence of the magnetotropic coefficient above $B_c$. $B_c$ has a pronounced minimum for field orientations near the $ab$-plane. Therefore, we focus on a subset of angles near the $ab$-plane that allow us to view this anomalous behavior in the broadest field range (Figure 2). We note that unlike torque, the magnetotropic coefficient does not vanish near the crystallographic directions (see Ref. 17 and SI). Because $k$ is the second angular derivative of the free energy, the linear-in-field dependence of $k$ (Figures 2A & C) requires a linear-in-field dependence of the free energy itself\textsuperscript{17}. Put differently, the field dependence of $k$ captures the anisotropic part of the field-dependent free energy. The anomalous high-field behavior of the free energy in RuCl\textsubscript{3} can be seen most clearly in the saturation of the field-derivatives of $k$ at 1.3 K (Figure 2B). This saturation at high fields persists to at least 20 K – more than twice the Néel temperature (Figure 2D), indicating
FIG. 2. A) Angle dependence of the magnetotropic coefficient \( k \) as a function of magnetic field at a temperature within the AFM phase. Only some of the curves from Figure 1A are shown for clarity. B) The corresponding field-derivatives of the curves show saturation once the AFM state is suppressed with magnetic field. C) Similar to the data shown in A, but at a temperature of 20 K – well above the AFM state. D) The corresponding field-derivatives of the data shown in C.

that this behavior is not associated with AFM order. It is surprising that the crossover from quadratic-in-field to linear-in-field behavior of \( k \) occurs at a field well below \( J_\alpha \sim 150 \, \text{T} \). In a conventional spin system, one would expect this crossover to occur at a field-scale of order \( J_\alpha \).^{28}

Figures 1 and 2 indicate a peculiar competition between temperature and magnetic field. We performed a set of high-field measurements over a broader range of temperatures, with field applied in the honeycomb planes where the AFM transition is at a minimum (Figure 3). At low temperatures, the magnetotropic coefficient increases linearly with magnetic field over the entire field range once magnetic order is suppressed. As temperature is increased, the
FIG. 3. Field-temperature scaling of the magnetotropic coefficient in RuCl$_3$. A) Pulsed magnetic field sweeps up to 64 T with field applied in the honeycomb planes. At low fields (B $\leq$ 4 T), the magnetotropic coefficient is quadratic in field and proportional to $(\chi_a - \chi_c)B^2$, which was used to calibrate the absolute value of the magnetotropic coefficient (see SI). The magnetotropic coefficient is linear-in-field above the AFM transition up to the highest measured field. As temperature is increased, the onset field of the linear behavior increases approximately linearly with temperature (panel C). For temperatures above $\sim$70 K, the linear regime is not reached. The inset shows the field-derivative of the data (obtained by a running slope analysis), indicating saturation of the field-slope above the AFM transition and approximately linear behavior of the onset field $B^*$ with temperature. B) The temperature dependence of the zero-field intercept from the data in panel A. Each curve is obtained from the zero-field intercept of a linear fit in a 10 T field range around the field value indicated in the legend. In the intermediate temperature range, all curves collapse onto a straight line with a zero intercept, which indicates that the high-field offset of the linear-in-field behavior is itself linear in temperature (see SI). The saturation behavior at low temperature indicates the low-energy cutoff of the scale-invariance. The peeling away from the gray line at high temperature indicates the insufficient field range to reach the limiting high-field linear behavior. C) Temperature dependence of the crossover field $B^*(T)$ (depicted in the inset of panel A).
linear-in-field behavior of $k$ is cut off below a field $B^*(T)$ (Figure 3A). This cutoff field itself scales linearly with temperature (Figure 3C). Together, this behavior of $k$ in a broad field and temperature range indicates scale-invariant behavior of the form $k/B = \psi(B/T)$. This scaling allows for linear-in-temperature behavior at high fields, $\psi(x) = b + a/x, x \ll 1$ (see SI). Indeed, Figure 3B shows this scaling in the temperature dependence of the zero-field intercept as extrapolated from high fields. This intercept clearly shows a common slope with a zero-temperature intercept, representing $k_{\text{int}} \propto T$ behavior at high fields. The deviation of $k_{\text{int}}$ from linearity at high temperature results from an insufficient field range at these temperatures. At low temperature, the deviation represents a cutoff of the $B/T$ scaling, independent of magnetic field. This cutoff, approximately 10 K in energy units (Figure 3), may be simply related to the fact that this analysis cannot be performed below the AFM transition. Alternatively, it could indicate the upper bound of a small intrinsic energy scale that leads to a deviation from the Kitaev model and gives rise to magnetic ordering.

The angle dependence of RuCl$_3$ in high magnetic fields rules out the possibility that the observed $B/T$ scaling (Figure 3) results from isolated spins (or spin-clusters) with an anisotropic $g$-factor. Figure 4 shows the angle dependence of $k$ in magnetic fields up to 35 T and at 20 K – well above the AFM transition temperature. At low fields, the magnetotropic coefficient follows a $\cos 2\theta$ dependence as expected in the linear regime. At fields above 10 T, the magnetization becomes progressively more nonlinear: not only do higher angular harmonics become significant, but also an overall negative “weight” sets in. When magnetic field crosses the direction perpendicular to the honeycomb planes ($c^*$ in Figure 4), $k$ exhibits a “spike”, which becomes more prominent as magnetic field is increased. Both the spike and the negative weight point to a singularity in the angle dependence of the free energy (and its derivatives) as the magnetic field crosses the $c^*$-axis (see SI). This is independently confirmed by direct measurements of the torque, which show a jump when magnetic field crosses the direction perpendicular to the honeycomb planes (see SI). This singularity in the angle dependence of the free energy, and the high angular harmonics shown in Figure 4, indicates that it is the spin-anisotropic exchange interactions $J_\alpha$ that set the high field scale-invariant behavior, rather than $g$-factor anisotropy.$^{29}$
FIG. 4. A) The angle-dependent magnetotropic coefficient at a temperature of 20 K – well above the AFM state. With increasing magnetic field, a spike develops and sharpens along the $c^*$-axis. Note that the position of the spike shifts to angles below 180° with increasing magnetic field, which is due to the increasing importance of the exchange interactions within the honeycomb planes. The spike at $c^*$ and the vertical asymmetry provide evidence for a fixed magnetic moment in the honeycomb planes that flips as the magnetic field crosses the $c^*$-axis. The schematic inset shows magnetic field configurations with respect to $c^*$ that should lead to a symmetric response in the magnetotropic coefficient provided a mirror plane parallel to the honeycomb planes exists. These magnetic field configurations are marked on the data, showing that the magnetotropic is not symmetric about the $c^*$-axis.

III. DISCUSSION

The observed scale-invariant behavior is not foreign to spin systems. For example, Curie’s law describes the behavior of a gas of isolated spins for which the magnetization is only a function of field and temperature. This behavior is expected from the fact that there are no energy scales in the system other than those set by temperature and magnetic field. What is surprising is that the same behavior emerges in RuCl$_3$ in the presence of the large energy scale $J_\alpha$.

In condensed matter systems, the material properties are often controlled by a single large
underlying energy scale. For example, in Fermi liquids, where the large energy scale is $E_F$ despite strong correlations, all low-energy properties are perturbative with $E_F$ in the denominator (e.g., magnetization $\sim (B/E_F)$). Similarly, for most correlated magnets, the exchange energy provides such an energy scale and the response is perturbative in this energy scale. By contrast, in field theories like electromagnetism, physical observables at low energy are independent of any high-energy cutoff, even though the cutoff is essential to define the theory. Interestingly, the observed scale-invariant behavior in RuCl$_3$ suggests that the latter type may be applicable to condensed matter systems exhibiting spin liquid behavior.

IV. ACKNOWLEDGMENTS

The authors are grateful to Michael Baenitz, Ali Bangura, Radu Coldea, George Jackeli, Steve Kivelson, Steven Nagler, Roser Valenti, Chandra Varma, Stephen Winter, and Jan Zaanen for insightful discussions. Samples were grown at the Max Planck Institute for Chemical Physics of Solids. The DC-field measurements were done at the National High Magnetic Field Laboratory (NHMFL) in Tallahassee, FL. The pulsed-field measurements were done in the Pulsed Field Facility of the NHFML in Los Alamos, NM. X-ray analysis was done at Cornell University. Research conducted at the Cornell High Energy Synchrotron Source (CHESS) is supported by the National Science Foundation under award DMR-1332208. The DC- and pulsed-field work at the NHMFL is supported through the National Science Foundation Cooperative Agreement numbers DMR-1157490 and DMR-1644779, The United States Department of Energy, and the State of Florida. R.D.M. acknowledges support from LANL LDRD-DR 20160085 topology and strong correlations. Y.L. is supported by the US Department of Energy through the LANL/LDRD program and the G.T. Seaborg institute. J.C.P. is supported by a Gabilan Stanford Graduate Fellowship and a NSF Graduate Research Fellowship (grant DGE-114747)

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$J_\alpha$ is defined as the principal components of the exchange tensor on a Ru-Ru link, $J_{ij}S_i^1S_j^2$, where $S_i^1, S_j^2$ are spins on two neighboring Ru ions. For a given Ru-Ru link, the exchange tensor $J_{ij}$ is diagonalized in the orthogonal basis where $\alpha = \hat{x}$ is along the Ru-Ru link, $\alpha = \hat{y}$ is along the Cl-Cl link in the square Ru-Cl-Ru-Cl plaquette, and $\alpha = \hat{z}$ points perpendicular to the plaquette. The latter is the Kitaev spin-anisotropy term. The magnitude of $J_\alpha$ is the same on all links (for an undistorted honeycomb lattice), but the principle axes $\hat{\alpha}$ differ from link to link.

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Appendix S1: Experimental setup of resonant torsion in pulsed fields

In DC magnetic fields, the resonant frequency shift can be detected with a phase-locked loop (PLL), where the driving frequency is adjusted with negative feedback from the phase of the pickup\textsuperscript{30,31}. Our existing implementation of the PLL is not fast enough to reliably follow the resonant frequency in pulsed fields, where it shifts by 100’s of Hz over the duration of a $\sim$20 msec pulse.

To achieve the necessary resolution of the frequency shift, we analyze the free oscillation of the lever, somewhat similar to pulsed-echo measurements that detect the speed of sound\textsuperscript{32}. We ring-up the oscillation amplitude by driving the lever close to the resonant frequency and turn off the driving voltage right before the magnet pulses (Figure S5). The lever oscillates freely throughout the whole duration of the magnet pulse. In the adiabatic limit, the frequency shift reflects closely the static natural oscillation frequency at an instantaneous value of the magnetic field. As the pulse progresses, the energy dissipation in the lever, sample, and surrounding exchange gas reduce the amplitude of the lever oscillations.

The free evolution of the lever in the magnetic field is governed by

$$\frac{d^2 \Delta \theta}{dt^2} + \omega_0(t)^2 \Delta \theta = 0, \quad (S1)$$

where the time dependence of the frequency $\omega_0(t)^2 = (K + k(t))/I$ is a result of the field dependence of the magnetotropic coefficient $k(t) = k(B(t))$ during the pulse $B(t)$. The adiabaticity parameter $\alpha$ is defined as the relative change of frequency during the period of oscillation, $\alpha = d(\omega_0^{-1})/dt$. In our measurement (Figure 3 of the main text), the frequency $\omega_0(t)$ shifts by about 500 Hz around 50 kHz when the field is pulsed to 65 T over 50 msec (on the falling side of the pulse), therefore $\alpha \approx 5 \times 10^{-6} \ll 1$ and the measurements in Figure 3 of the main text are well within the adiabatic regime.

In the adiabatic limit $\alpha \ll 1$, the angle evolves according to

$$\Delta \theta(t) = A(t)e^{i\varphi(t)}, \quad \varphi(t) = \int_0^t \omega_0(t')dt', \quad \omega_0(t) = \frac{d\varphi(t)}{dt}, \quad (S2)$$
FIG. S5. The magnetotropic coefficient of RuCl$_3$ in pulsed fields. A) Piezo-induced voltage during free oscillation of the lever. The lever is driven close to the resonant frequency to achieve a large initial amplitude. For the Akiyama lever assembly, the piezo-voltage is about 1.5 mV at 100 mV drive and it corresponds to about 0.5° bending in the oscillations. The drive voltage is turned off at time $t_1 = 10$ msec (shortly after the start of digital recording). At $t_1$, the amplitude of the pickup voltage is increasing by about 20%. This is because when the drive is turned on, the pickup voltage is a superposition of the piezo voltage and the voltage induced in the wires leading to the lever. Their relative magnitude and phase depends on wiring details and differences in the drive frequency and the natural frequency of the lever. At $t > t_1$, the measured pickup voltage is entirely determined by the piezo voltage on the stressed (by the lever) transducer fork. The magnet pulses 20 msec later, at time $t_2 = 30$ msec. Peak field is reached approximately 8 msecs later at time $t_3 = 38$ msec. B) Zoom-in of the region indicated by the black line in panel A, highlighting the start of the magnet pulse. C) Frequency of the lever oscillations over the duration of the entire pulse. The frequency is obtained by analyzing the power spectrum in a 200 μsec window, about 10 periods. D) Time-evolution of the magnetic field in the multi-shot 65 T magnet system at the NHMFL-pulsed field facility in Los Alamos. E) Field-dependence of the frequency of oscillations obtained by plotting C vs D. The blue and purple curves show the raising and lowering side of the field pulse, respectively.

where the amplitude $A(t)$ is a slow function of time. Even when energy dissipation is absent, the energy $E(t)$ of the lever is changing during the pulse due to the work done by the time-varying magnetic field. We estimate the associated change in the amplitude $A(t)$ from the fact that the rate of change of the adiabatic invariant $J = E(t)/\omega_0(t)$ is proportional to the square of the rate of change of the frequency $d\omega_0/dt$, and therefore $J$ is approximately
constant during the pulse. The energy of the oscillator (Eq. S1) is 
\[ E = I_w^2 A^2 \]
and therefore 
\[ A(t) \propto 1/\sqrt{\omega_0(t)} \] or 
\[ \delta A(t)/A_0 = -(1/2)\delta \omega_0(t)/\omega_0 \]
for small changes in frequency. For the measurements in Figure 3 of the main text, this amounts to less than 1% changes in the amplitude \( A(t) \) (Figure S5), far less than the changes associated with direct energy dissipation effects, where \( A(t) \) changes by a factor of \( e \) over 100 msec at 5 K.

**Appendix S2: Calibration of the magnetotropic coefficient**

The absolute magnitude of the magnetotropic coefficient \( k \) of the sample can be determined in resonant torsion magnetometry from the measured frequency shift \( \Delta \omega \) from Eq. S2. The estimate of \( K \) in Eq. S24 is, in practice, only 10-20% accurate due to the complex shape of the Akiyama lever and the finite size of the sample (i.e., the sample is attached to the lever over a finite area rather than a single point as is assumed in the discussion in Section S8).

In principle, \( K \) can be calibrated experimentally either from measurements of the frequency shift in small fields on a sample for which the magnetic susceptibility is known independently, or from a “gravity” test similar to that used to calibrate conventional torque levers (Sec. S85). The current values of the gravitational frequency shift, <100 mHz, are within the complex systematic bias frequency shifts of the present lever setup: temperature fluctuations, effects of exchange gas convection, small conduction of the p-doped silicon lever, etc. are all <1 Hz. Each Akiyama lever must be calibrated separately because their oscillation frequency varies within \( \sim 15\% \) from lever to lever.

To obtain specific values for the magnetotropic coefficient (per unit volume, per mol, etc.), one must know the mass of the sample. In this work, the sample masses are all in the range of 10 - 80 ng (Figure S6D) for which we could not obtain a reliable mass measurement. Instead, we use the known magnetic susceptibility (per unit volume) of RuCl$_3$\(^4,7,9,24\). For small fields (in the linear regime), the magnetotropic coefficient is related to the magnetic susceptibility via 
\[ k = B^2(\chi_1 - \chi_2)\cos 2(\theta - \theta_0) \]
where \( \chi_{1,2} \) are the two components of the magnetic susceptibility tensor along the two principal directions in the plane of rotation.\(^{17} \)
FIG. S6. Frequency-shift measurements up to 63T for temperatures ranging from 1.5 to 150 K. A) The zero-field frequency shift is a result of small (≤ 1%) changes in the elastic stiffness of the lever in a broad temperature range. B) The absolute frequency shift with respect to zero-field frequency $\Delta f(B = 0) = 0$. The vertical axis on the right shows corresponding values of the magnetotropic coefficient (see Section S2). C) The low-field (linear regime) coefficient $a$ of frequency shift vs $B^2$, $\Delta f = aB^2$. For the (fixed) orientation of the lever in this measurement $\Delta f = (\chi_\parallel - \chi_\perp)B^2$ where $\chi_\parallel, \chi_\perp$ are the principal components of the magnetic susceptibility. The vertical axis on the right shows the corresponding values of the magnetic anisotropy $\chi_\parallel - \chi_\perp$ in per-mol units. D) Sample mounted on the lever for field aligned in the $ab$-plane.

(we assume that the plane of oscillations of the lever coincides with the plane of rotation of the stage). This way, identifying the measured frequency shift with its known dependence on field and angle at small fields allows a direct calibration.

The data in Figure 3 of the main text has been calibrated as follows. Figure S6C shows the coefficient of proportionality of the frequency shift vs $B^2$ in the low-field limit, which is pro-
portional to $\chi_{\parallel} - \chi_{\perp}$ for rotation out of the plane (when $\cos 2\theta = 1$). At temperatures below 5 K, the magnetic anisotropy $\chi_{\parallel} - \chi_{\perp}$ saturates at $21 \times 10^{-3}$ emu/mol = 0.21 J/T$^{24,7,9,24}$. The measured value of $df/d(B^2)$ is 0.48 Hz/T$^2$ and is equivalent to 0.21 J/mol or 1 Hz $\leftrightarrow$ 0.44 J/mol for the data in Figure 3 and Figure S6 (conversion shown on the right vertical axes in Figures S6B and C). This procedure was also used to calibrate the data in Figures 1, 2, and 4 of the main text. Here $df/dB^2$ is 0.46 Hz/T$^2$ at an angle of 100° at the lowest temperature, which yields 1 Hz $\leftrightarrow$ 0.43 J/mol/rad$^2$.

We can estimate the magnetotropic coefficient directly from Eq. (S2). At 1 T, the frequency shift is $\Delta f = 0.48$ Hz, or, using Eqs. (S2) and (S30), $k = 2K^{(0)}(\Delta f/f_0) = 3.6pJ/rad^2$ at 1 T. The sample size is roughly $50 \times 70 \times 2 \mu m^3$ which at density $3.1g/cm^3$ is 26 pmol (at 4 f.u. per unit cell). Thus $k = 4.7pJ/rad^2 / 26 pmol = 0.18 J/mol/rad^2$ at 1 T. At 1 T we are in the linear regime where $k = B^2(\chi_{\parallel} - \chi_{\perp}) \cos 2\theta$. Therefore, 0.18 J/mol/rad$^2$ obtained for specific $k$ here must be compared with the value of 0.21 J/mol/T$^2$ for magnetic susceptibility anisotropy $\chi_{\parallel} - \chi_{\perp}$ used above. The biggest uncertainty in this ”direct” calibration procedure is the thickness of the sample and the uncertainty in $K^{(0)}$ due to more complex shape of the lever (Eq. (S30) was derived for rectangular-crosssection lever).

Appendix S3: Thermodynamics of the AFM phase boundary.

Figure S7A shows the angle dependence of the magnetotropic coefficient at base temperature for a set of magnetic fields (the same data as in Figure 1B of the main text, but with the period doubled to highlight the asymmetry around $c^*$-axis, see Section S5 for details). Figure S7B shows a subset of the field scans from Figure 1A of the main text to emphasize the evolution of the jump magnitude $\Delta k(B_c, T_c, \theta_c)$ with the angle $\theta_c$ on the boundary (Figure S7C). The angle dependence of the jump $\Delta k$ indicates a stronger thermodynamic signature of the phase transition as the magnetic field approaches the $c$-axis, with the maximum occurring at an angle $\sim 10^\circ$ away from the $c$-axis. Jumps in the magnetotropic coefficient
FIG. S7. A) Angle scans of the magnetotropic coefficient $k$ at 1.3 K, obtained by continuous rotation of the sample in fixed magnetic fields ranging from 7.5 to 17.5 T. At 7.5 T, the angular dependence of magnetotropic coefficient is close to the \( \cos 2\theta \) as expected in the linear response regime where \( F = (1/2)\chi_{ij}H_iH_j \). Rotation of the crystal in magnetic fields \( \geq 10 \) T highlights transitions into and out of the AFM phase. Entry into the ordered state is observed as a sharp jump down in the magnetotropic coefficient\(^17\). Together with panel B, these features are used to map out the anisotropic AFM phase boundary (inset of C). B) Field scans of the magnetotropic coefficient at 1.3 K for several field orientations $\theta$, where $\theta$ is defined as the angle between the $c$-axis and the applied magnetic field. The magnitude of the jump $\Delta k$ across the AFM boundary is related to the discontinuity in other thermodynamic coefficients (such as heat capacity) via the Ehrenfest relations\(^17,26\). C) Angle dependence of the magnitude of the discontinuity at the AFM boundary in panel B. Note that the angular dependence is parametric: $\Delta k$ is a function on the surface of the AFM boundary (inset in panel C) $\Delta k(B_c, T_c, \theta)$, where (at fixed $T_c$) $B_c$ itself is a function of $\theta$. On general symmetry grounds, $\Delta k(B_c, T_c, \theta)$ must vanish along the symmetry directions in the lattice where the derivative ($\partial T_c/\partial \theta$)$_B$ on a fixed-field section of the AFM boundary vanishes. As magnetic field is rotated away from the $c$-axis toward the honeycomb plane at $\theta = 90^\circ$, the magnitude of the jump $\Delta k(B_c(\theta), T_c, \theta)$ increases and reaches a maximum at $\sim 10^\circ$.

$\Delta k$ and the heat capacity $\Delta C$ are related via the Ehrenfest relation

\[
\Delta k = -\Delta C \left( \frac{\partial T_c}{\partial \theta} \right)_B^2,
\]  

(S1)
where both $\Delta k$ and $\Delta C$ depend on the position on the AFM phase boundary $(B_c, T_c, \theta_c)$ (inset of Figure S7C)\textsuperscript{17}. By symmetry, $\left(\frac{\partial T_c}{\partial \theta}\right)_B$ must vanish near the crystallographic symmetry directions ($ab$-plane and the $c$-axis) as observed (Figure S7). Nernst’s theorem states that the jump in heat capacity $\Delta C$ must vanish at zero temperature\textsuperscript{26}, and therefore is small at temperatures well below $T_N$ ($1.3 \text{ K} \sim 0.2 \times T_N$ at $B = 0$). However, it is not clear if $\Delta C/T_c$ vanishes as well in the zero-temperature limit in RuCl\textsubscript{3}. Regardless, the magnitude of the jump in the magnetotropic coefficient $\Delta k$ appears to be finite in the zero-temperature limit indicating singular behavior of the derivative $\left(\frac{\partial T_c}{\partial \theta}\right)_B$ along the AFM phase boundary at low temperatures.

**Appendix S4: Evolution of the principal components of the magnetic susceptibility**

The monoclinic crystal structure of RuCl\textsubscript{3} does not require the principal components of the susceptibility tensor to coalign with the crystallographic axes or with the honeycomb planes. As such, we define $\theta$ using the low-field response of the magnetotropic coefficient, which is described by the linear magnetic susceptibility tensor $\chi_{ij}$. In this limit, the magnetotropic coefficient exhibits a $\cos 2\theta$ angle dependence (Figure S8A) that allows us to define three perpendicular principal components of the susceptibility tensor, for example as $a_m$, $b$, and $c_m$ shown schematically in Figure S8C. The monoclinic angle in RuCl\textsubscript{3} is perpendicular to the $b$-axis, which allows the other two principal components ($a_m$ and $c_m$) to freely rotate in the $ac$-plane with temperature. In other words, in a monoclinic system, not only are the principal components $\chi_i$ evolving with temperature, but so is their direction relative to the crystallographic axes. This is observed as a continuous phase shift in the $\cos 2\theta$ angle dependence (gray line in Figure S8A and right axis in Figure S8B). The magnetic axis $b_m$ is required by symmetry to coincide with the crystallographic $b$-axis at all temperatures.

A similar realignment of the magnetic response occurs as we increase the magnetic field. Because the magnetic response is nonlinear at high fields, the angle dependence no longer follows a $\cos 2\theta$ dependence. Instead, one can follow the angle of the maximum near the $ab$-plane in the magnetotropic coefficient, which shifts with increasing field (illustrated by
FIG. S8. A) The angle-dependent magnetotropic coefficient at 12 T for temperatures ranging from 5 to 200 K. At 12 T, the magnetotropic coefficient follows a $\cos 2\theta$ angle dependence at temperatures above the AFM phase. The amplitude of the $\cos 2\theta$ dependence is directly proportional to the magnetic anisotropy coefficient $\alpha_{ij} = \chi_i - \chi_j$ in the plane of the vibrating lever (left axis in panel B). B) The magnetic anisotropy monotonically increases as the AFM phase is approached at lower temperatures. This is accompanied by a shift in the phase of the $\cos 2\theta$ angle dependence (right axis), which is a direct consequence of the fact that the principal magnetic axes can rotate due to the monoclinic crystal structure. C) Schematic representation of the crystallographic and principal magnetic axes ($a_m$ and $c_m$) with in the monoclinic crystal structure of RuCl$_3$.

the gray line in Figure S9A). This behavior indicates that the magnetic response at high fields is determined entirely by the exchange interactions within the honeycomb planes (i.e., the maximum $k$ for each curve approaches the $ab$-plane as magnetic field increases). The
observed $\sim 10^\circ$ phase shift with increasing field indicates that the direction of the maximum in the magnetotrophic coefficient approaches the $ab$-plane. Simultaneously, the minimum in the magnetotrophic coefficient approaches the $c^*$-axis. The observed phase shift of $\sim 10^\circ$ is qualitatively consistent with the reported monoclinic angle $\beta = 108.8^\circ$ (i.e., the low field direction of $c_m$ is about halfway between the crystallographic $c$-axis and the direction $c^*$ perpendicular to the $ab$-plane.

One would expect both the amplitude of the magnetic anisotropy and the phase to depend on the $\phi$ plane of rotation, even in the linear response regime. This is confirmed in Figure S9B for the magnetotrophic coefficient normalized by magnetic field for two different planes of rotation. A change in amplitude and a slight shift in phase are observed as the principal magnetic axes map onto each plane of rotation.

FIG. S9. A) A close-up of the data shown in Figure 4 of the main text to highlight the increasing role of the exchange interactions within the honeycomb plane with increasing magnetic field. B) The angle dependence of the magnetotrophic coefficient normalized by magnetic field for rotation in the two azimuthal planes described in Figure S12. Both the amplitude and phase of the $\cos 2\theta$ dependence depend on the $\phi$ plane of rotation.
Appendix S5: Angle-dependence of the magnetic free energy

1. Symmetry discussion

We now discuss the asymmetry that develops around the $c^*$-axis in Figure 4 of the main text in the context of spin-anisotropic exchange interactions. We consider a single honeycomb plane, where each Ru$^{3+}$ ion is surrounded by perfect chlorine octahedra, which are oriented identically on all sites of the crystal lattice (Figure S10)\textsuperscript{25}. The exchange interactions of a particular Ru ion with its three neighbors on the honeycomb lattice are mediated via three mutually orthogonal “exchange plaquettes” (colored planes in Figure S10). This chemical environment breaks mirror reflection symmetry in the plane parallel to the honeycomb planes. Since the principal directions of the exchange tensor on each link are tied to the orientation of the exchange plaquettes\textsuperscript{12}, the spin Hamiltonian of RuCl$_3$ lacks this mirror plane symmetry as well, unless the exchange tensor $J_{ij}$ is isotropic on each Ru-Ru link.

This point is illustrated geometrically in Figure S10. For a given Ru ion, the perfect octahedra allows for two identical choices for the orientation of the triad of exchange planes\textsuperscript{14}. Each choice of triad transforms into the other under a mirror reflection in the honeycomb plane. To illustrate this point, one can think of each exchange triad as a three-winged propellor which rotates clockwise under water flowing from top to bottom (Figure S10). Under reflection, orientation of the triad would flip, changing the direction of rotation to counter-clockwise.

The lack of mirror reflection symmetry in the honeycomb plane can be seen directly in the angle-dependent data in Figures 1 and 4 in the main text and in Figures S7 and S12. The free energy (and the magnetotropic coefficient) for magnetic field oriented on opposite sides of the $c^*$-axis ($B$ and $B'$ in Figure S11) are related to each other via reflection in the honeycomb plane (magnetic field is an axial vector). The magnetotropic coefficient on two sides of the $c^*$-axis are not equal to each other $k(\theta) - k(-\theta) \neq 0$. This is true both below the Néel temperature (Figure 1 in the main text and Figure S7), as well as above
FIG. S10. RuCl$_3$ has a planar honeycomb structure, where each Ru ion (blue sphere) is surrounded by Cl octahedra. All Cl octahedra in the structure are oriented identically. Two nearest-neighbor Ru ions share two Cl ions, which delineate the common edge shared between the neighboring octahedra. The exchange interaction between the two Ru ions is therefore mediated by two equivalent Cl ions simultaneously – the interference between these two exchange paths is the microscopic basis for stronger spin-anisotropy of the exchange tensor $J_{ij}$ on each Ru-Ru bond$^{12}$. Geometrically, these two exchange paths define a rectangular Ru-Cl-Ru-Cl “exchange plaquette”. The orientation of the plaquette defines three mutually orthogonal axes: two in the plane of the plaquette and one perpendicular to it. These directions are the principal components of the exchange tensor $J_{ij}$ on each Ru-Ru link, with the one perpendicular to the exchange plaquette describing the Kitaev component. The exchange plaquettes connecting the three nearest neighbors of each Ru ion are mutually orthogonal (colored planes)$^{12}$. Although the Cl octahedron is inversion-symmetric around the center Ru ion, this is not true when considering the three exchange plaquettes around each Ru ion. In particular, under a mirror reflection parallel to the honeycomb planes, the exchange environment on neighboring Ru ions transform into each other. Overall, the exchange Hamiltonian describing the entire RuCl$_3$ plane does not have reflection symmetry with respect to the plane parallel to itself unless $J_{ij}$ is isotropic.

(Figure 4 of the main text and Figure S12). As discussed above, at temperatures above the Néel temperature, the asymmetry $k(\theta) - k(-\theta) \neq 0$ (or equivalently, the Free energy $F'(\theta) - F(-\theta) \neq 0$) is a consequence of an anisotropic exchange tensor $J_{i,j}$. Therefore, the measurements in Figures 1 and 4 in the main text and Figures S7 and S12 indicate that $J_{i,j}$ is not isotropic. Furthermore, one expects a strong $\varphi$-dependence in $F'(\theta) - F(-\theta)$ due to the three-fold rotation symmetry of the honeycomb lattice (Figure S11). Figures S12A and S12C demonstrate a large dependence of $k(\theta) - k(-\theta) \neq 0$ on the azimuthal angle $\varphi$ describing the plane of rotation.
2. Singularity in $F(\theta, \phi)$ across the $c^*$-axis

As discussed in the context of Figure 4 of the main text, the angle-dependent magnetotropic coefficient reveals an unusual "spike" around the $c^*$ direction and an overall negative "weight" (a negative value of the average over $180^\circ$) that both strengthen with magnetic field (Figure S12 and Figure S13). Both features are evidence for a thermodynamic singularity in the angle-dependent magnetic free energy of RuCl$_3$ for fields near the $c^*$ direction. This singularity, most apparent at high fields, occurs at temperatures both below and above the Néel temperature.

When the plane of rotation of the rotator stage $\theta$ is aligned with the plane of lever bending
FIG. S12. Angle scans of the magnetotropic coefficient in two azimuthal planes at 30° from each other. A) Angle scan in the $ac^*$-plane (i.e., rotation around the crystallographic $b$-axis). The inset shows the orientation of the crystal on the lever. Panel C shows the orientation of the crystal axes with respect to the crystal morphology. The data has been overlaid with a 180° shift to cover the full 360° rotation. 90° and 270° correspond to the field applied parallel to the $ab$-plane, 0° and 180° correspond to field along the $c^*$-direction. The segments near the $c^*$-direction were omitted due to poor resonance tracking because the width increases significantly and the resonance shifts too quickly. B) Similar to panel A, but for rotation between the $c^*$-axis and an in-plane direction 30° from the $a$-axis (parallel to $\phi_2$ in panel C). C) Schematic drawing of the morphology of the crystal used in these measurements and the orientation of the in-plane crystallographic directions (see Figure S14 for details).

$\Delta \theta$, the torque $\tau(\theta)$ and the magnetotropic coefficient $k(\theta)$ are related via $\tau(\theta) = \int_0^{\theta} d\theta' k(\theta')$. Because $\tau(\theta)$ is a periodic function of $\theta$ (with 180° periodicity required by time-reversal, Section S5.5), the area under the angle scans of the magnetotropic coefficient over 180° range, or simply the average $\langle k(\theta) \rangle$, must be equal to zero $\langle k(\theta) \rangle = (1/\pi) \int_{\theta}^{\theta + \pi} d\theta' k(\theta') = 0$. This is clearly not the case for the data in Figure 4 of the main text and Figure S12B, where the average $\langle k(\theta) \rangle_{\theta}$ over 180° is a negative value: $k(\theta)$ has a “negative weight”.

The spike at $c^*$ hints at the non-zero $\langle k(\theta) \rangle_{\theta}$: because the total area (including the area under the spike) under the angle-dependent magnetotropic coefficient curves must be equal
FIG. S13. A) The angle-dependent magnetotropic coefficient at a temperature of 20 K – well above the AFM state. With increasing magnetic field, a spike develops and sharpens along the $c^*$-axis. Note that the position of the spike shifts to angles below 180° with increasing magnetic field, which is due to the increasing importance of the exchange interactions within the honeycomb planes. The spike at $c^*$ and the vertical asymmetry provide evidence for a fixed magnetic moment in the honeycomb planes that flips as the magnetic field crosses the $c^*$-axis. The schematic inset shows magnetic field configurations with respect to $c^*$ that should lead to a symmetric response in the magnetotropic coefficient provided a mirror plane parallel to the honeycomb planes exists. These magnetic field configurations are marked on the data, showing that the magnetotropic is not symmetric about the $c^*$-axis. B) Torque divided by magnetic field $\tau/B$ as a function of field orientation angle at 4 K. At low fields $B \leq 10$ T, the expected $\sin 2\theta$ angle dependence of the linear response regime is observed. At magnetic fields above $B_c$, the angle dependence of $\tau/B$ is better described by a $\sin \theta$ component, accompanied by a sign($\cos \theta$) factor that captures a discontinuity at 180°. Note that when magnetic field is applied close to the $c^*$-axis, even with fields up to 34.5 T, the AFM state is not suppressed and as such, these data points (pale on plot) are excluded from the fits.

To zero, our data indicates that the measured area under the spike is not as large as expected and suggests a slight precession of the magnetic field about the singularity at $c^*$. Thus, the overall negative weight indicates a finite misalignment angle $\xi$ between the rotation and oscillation planes. The levers and samples are mounted by hand and we estimate $\xi = 5 - 10^\circ = 0.1 - 0.2$ rad $\ll 1$. For a smooth angle-dependent $k(\theta')$ over a broad angle range, a small misalignment $\xi \ll 1$ leads to $\propto \xi$ changes in the observed weight $\int_\theta^{\theta+\pi} d\theta' k(\theta')$. Such a small misalignment over a broad angle range cannot account for the large negative weight observed. Instead, it is a result of a sharp delta-function-like feature in $k(\theta)$ that is narrower than the misalignment in the measurement $\xi = 5 - 10^\circ$. In other words, we precess around an extremely sharp singularity in $k(\theta)$ and therefore miss a significant area in the experimentally observed $\langle k(\theta) \rangle$. Both the spike and the negative weight independently point to the same conclusion – a singularity in the free energy exists when field is applied along the $c^*$-direction.
Figures S12A and B directly indicate the location of this singularity: both show a spike in $k(\theta)$ near the $c^*$-axis. The spike gets stronger as we increase magnetic field, which is somewhat counterintuitive. However, the spike is associated with a singularity in the angle- rather than the field-dependent magnetotropic coefficient: the free energy increases monotonically with field, and with it, the magnitude of the spike increases. Due to the misalignment, the observed magnitude of the spike is much smaller than what is required to compensate for the missing area. We reemphasize that while the singularity across the $c^*$-axis exists within the broken symmetry state at low temperatures and low magnetic fields, it is a property of the spin-liquid state that exists at high magnetic fields and temperatures, and the spin-Hamiltonian.

A sketch of the free energy that is qualitatively consistent with the observed behavior and the symmetries of the crystal is shown in Figure S11. The delta-function-like spike in $k(\theta)$ (or step in $\tau(\theta)$, see below) across the $c^*$-direction requires that the free energy $F(\theta)$ has a cusp. The three-fold rotational symmetry of the honeycomb lattice requires that three such cusp lines at 120° from each other meet at each pole (at $\theta = 0, \pi$). The cusp lines “diffuse” away from the poles. Therefore, the magnitude of the cusp-like singularity across $c^*$ in $F(\theta)$ when $\theta$ rotates in an azimuthal plane at $\varphi = \varphi_{\text{fixed}}$ may vary strongly with $\varphi_{\text{fixed}}$. In this respect, the contrast between the angle scans in Figure S12A and Figure S12B is most telling. From the x-ray analysis, we know that the plane of $\theta$-rotation in Figure S12A coincides with crystallographic $ac^*$-plane (Figure S12C). In this rotation plane, the angle scan is nearly symmetric around the zero-field value, indicating a small magnitude of the missing area $\int_{\theta}^{\theta+\pi} d\theta' k(\theta')$. Rotation of the azimuthal plane by only 30° (Figure S12B) makes a dramatic change: at large fields, the negative weight $\langle k(\theta) \rangle_\theta = (1/\pi) \int_{\theta}^{\theta+\pi} k(\theta')d\theta'$ is large and comparable to the amplitude of variation of $k(\theta)$ itself. Another 30° rotation of the sample in $\varphi$ would restore the near-zero $\langle k(\theta) \rangle$ behavior of Figure S12A (see the discussion of the rotational properties of the free energy in Figure S11). Similarly, a large negative $\langle k(\theta) \rangle$ is observed for the sample in Figure 1 of the main text where only one azimuthal plane was explored (see Figures S7, S18, S19, and S20).

A spike observed in the magnetotropic coefficient (Figure S13A) is consistent with a jump in the magnetic torque (Figure S13B). In torque measurements though, the magnetic field
does not need to be well-aligned with the $c^*$-direction to infer the existence of a jump-like singularity: the jump-like behavior can be observed in a broad angle range $> 5 - 10^\circ$ from the $c^*$-axis because it is not affected by misalignment (Figure S13B). Similarly, in resonant torsion measurements, the “robust” indicator of a singularity is the non-zero value of the weight accumulated over a broad angle range $\int_0^{\theta + \pi} d\theta' k(\theta')$.

A discontinuous jump in the torque (or equivalently, a cusp-like singularity in the free energy) across the $c^*$-axis is reminiscent of the jump observed in the angle-dependent torque in the 3D honeycomb iridate $\gamma$-Li$_2$IrO$_3$ at high magnetic fields$^{34}$. In $\gamma$-Li$_2$IrO$_3$, there are two inequivalent honeycomb planes that share the crystallographic $c$-axis and are oriented $\sim 35^\circ$ from each side of the $bc$-plane. Here, the jump in torque $\tau(\theta)$ occurs when magnetic field crosses the crystallographic $c$-axis, which appears quite different from that occurring at $c^*$ in RuCl$_3$. In RuCl$_3$, the jump in the torque (or equivalently the spike in the magnetotropic coefficient) across the $c^*$-axis can be imagined as flipping of large spin “icebergs” between 3 equivalent orientations in the honeycomb plane. We emphasize that while such a “magnetic iceberg” picture is consistent with the observed linear-in-field free energy (and $k(B)$ and $\tau(B)$) at very high magnetic fields, it is not consistent with the unsaturated magnetization up to 60 T$^{25}$ or the $B/T$ scaling observed in this work (Figure 3 of the main text).

In $\gamma$-Li$_2$IrO$_3$, the singularity occurs when magnetic field is applied along the $c$-axis (i.e., when field is parallel to both intersecting honeycomb planes, rather than perpendicular to them as in RuCl$_3$). The exchange interactions along the Ir-Ir links connecting different honeycomb planes (the “vertical” links$^{14,34}$) may result in the spin-icebergs locking in two inequivalent honeycomb planes. The singularity along the $c$-axis in $\gamma$-Li$_2$IrO$_3$ can be understood if only two (out of six) of the spin-iceberg configurations remain degenerate. In each of these two configurations the spin icebergs still point parallel to their respective honeycomb planes – but are also restricted to their intersection with the $ab$-plane.
3. **X-ray analysis**

Synchrotron x-ray characterization was performed at the CHESS-A2 undulator beamline to determine the crystal orientations in Figures S12, S15, and S16. An incident photon energy of 11.217 keV was selected using a double-bounce diamond $<111>$ monochromator, with a bandwidth $dE/E \sim 1 \times 10^{-4}$. Higher harmonics were rejected using a Rh-coated collimating mirror. The few nanogram crystal (as mounted in Figure S15B) was bathed in the larger x-ray beam, and diffraction was measured over small volumes of reciprocal space using a diffractometer-mounted photon counting area detector. The crystal orientation was determined using low-order Bragg reflections within the hexagonal plane in trigonal notation (Space group P3112, #151, with $a = b = 5.975$ Å, $c = 17.332$ Å, $\alpha = \beta = 90$, $\gamma = 120$). We co-indexed the $[1 0 0]$, $[1 -1 0]$, and $[0 -1 0]$ peaks, which we found to be resolution-limited within the hexagonal plane, showing long-range order and a single domain (Figure S14A). However, these peaks displayed an anisotropic peak shape, with a finite correlation length in the out-of-plane direction, indicating some stacking disorder (Figure S14B).

The x-ray analysis was used to identify the $\phi_1$ and $\phi_2$ orientations in Figure S12, as well as the plane of rotation in Figure S15. For the measurements in Figure S15, we found that the $ab$-plane was tilted $17.1^\circ$ from the vibration plane of the lever – the plane that includes the long axis of the lever and the perpendicular to the lever surface. This precession about the $ab$-plane gives rise to the large two-fold component in Figure S15A.

4. **In-plane angle-dependence of the magnetotropic coefficient**

To test the rotation properties of the free energy *around* the $c^*$-axis, we measured the magnetotropic coefficient as magnetic field was rotated in the honeycomb planes (Figure S15). Here, we are probing the azimuthal curvature of the magnetic free energy – along the intersection of the red shape representing $F(\theta, \phi)$ (Figure S11) and the cyan hexagon representing the orientation of the honeycomb planes. Figure S15A shows a large 2-fold
FIG. S14. A) Bragg reflections within the hexagonal plane. The peaks are indexed using trigonal notation. The small bright spots at 60° from each other confirm a high degree of crystallinity within the honeycomb planes. B) Broad scattering along the L-direction indicates stacking disorder along the c-axis.

(180°-periodic) component that is associated with misalignment of the sample on the lever: rather then being perpendicular to the plane of the lever (Figure S15B), the ab-plane of the sample is $\xi = 17.1° \sim 0.3$ rad away from it S5 3. For a small misalignment angle, the magnitude of the two-fold component leaking into the ab-plane rotation signal is of order $\xi^2$.

The 50 Hz amplitude of the two-fold signal at 35 T in Figure S15A is therefore consistent with the 1 kHz frequency shift near the ab-plane (90° in Figure S12A): $1 \text{kHz} \times \xi^2 = 89$ Hz. Figures S12C and D show the angle scans with the two-fold component subtracted (as a best-fit at each field). It shows the clear 6-fold (60°-periodic) signal over the entire angle range. The ab-plane variation of $F(\theta = 90°, \phi)$ is 20 Hz at 35 T in Figure S15C, which is 30 times smaller than the 1 kHz signal in Figure S12A. Since these two measurements probe the curvature of the free energy $F(\theta, \phi)$ in two orthogonal directions (see Figure S11), the difference factor of 30 indicates that the realistic picture of $F(\theta, \phi)$ for RuCl$_3$ would look like a flat disk in Figure S11. Similar to the data in Figure 3 of the main text, there is not much change in the magnitude of the magnetotropic coefficient at 35 T when temperature changes from 20 K to 3.5 K (Figure S15D). This indicates that the field–temperature competition at high fields does not affect much the shape of (the properly normalized) $F(\theta, \phi)$.

The apparently-irregular behavior at low fields in Figure S15D is, in fact required by the symmetry as discussed in Section S5. Figure S16A shows the low-field angle scans in Figure
FIG. S15. The magnetotropic coefficient for field rotation in the $ab$-plane. A) Angle scan at 20 K for several fixed fields. The large two-fold component is due to a slight misalignment of the sample on the lever. B) Picture of the crystal (same crystal as in Figure S12, but propped on its side) for rotation/vibration in the honeycomb planes. C) Angle scan in panel A after the two-fold component has been subtracted (left) and the corresponding polar plot (right). D) Angle scan at 3.5 K for several fixed fields with the two-fold component subtracted (left), along with the corresponding polar plot (right). The low-field scans near the AFM boundary ($\leq 10$ T) are discussed in Figure S16.
FIG. S16. A) A close-up of the in-plane angle dependence in Figure S15D for fields near the AFM phase boundary (left), and a polar plot of the same data (right). The curves are offset for clarity. Similar to Figure S15D, the curves are overlaid with the same data plus a 180° shift to extend to the full 360° range. The magnetotropic coefficient shows clearly the 6-fold structure of the thermodynamic response in the $ab$-plane. B) A sketch of the $\theta$ and $\varphi$ dependence of $B_c$ consistent with the symmetries of the crystal lattice and the exchange environment of Ru in the honeycomb plane. C) The “trajectory” of the field in the $B_x, B_y$-plane (parallel to the honeycomb planes) for the angle scans in panel A (color-coded in the same way).

S15D in more detail. Here, the field is close to the value of the critical field $B_c$ at 3.5 K. The polar plot in Figure S16A shows clearly the 6-fold behavior at 6 T and 10 T, but there are 3 extra dips in the 8 T scan between each of the 6-fold dips. This structure can be explained by the 6-fold modulation of $B_c$ in the $ab$-plane (red line in panel B, black line in panel C). At 8 T, we observe features corresponding to the minima and maxima of $B_c(\phi)$, as well as features corresponding to the crossing of the AFM phase boundary (intersection of the green and black lines in Figure S16C).
As discussed in Section S5, the magnetic free energy $F(\theta, \phi)$ in a time-reversal-invariant system must be the same for opposite field orientations. This requires 180° periodicity of the angle scans in the plane perpendicular to the honeycomb plane and 60° periodicity for angle scans in the plane parallel to the honeycomb planes. All angle scans at high fields in the plane perpendicular to the honeycomb plane show 180° periodicity, indicating that the underlying fluctuations in the spin-liquid state are time-reversal invariant. Figure S17 shows a direct comparison of the frequency shift for two opposite field orientations in the $ab$-plane. Both at high and low fields, there is no experimental indication of a deviation from $B \rightarrow -B$ symmetry.

**FIG. S17.** Tests of time-reversal invariance of the magnetotropic coefficient for fields up to 64 T in the $ab$-plane. Data is taken for the same sample and orientation as Figure 3 of the main text and Figure S6. A) Full field scans at three different temperatures. The inset shows schematically the field orientation. The in-plane orientation of the magnetic field is unknown for the sample in Figure 3 of the main text. B) The field-reversed scan plotted versus the absolute value of magnetic field lays exactly over the positive field scans in panel A. C) Zoom-in of the low-field part of panel B.
Appendix S6: Additional data at 1, 20, and 40 K

Figures 1 and 2 in the main text indicate that the linear-B behavior of the magnetotropic coefficient at high fields is evident in a broad range of angles around the $ab$-plane. This is shown for angles where 35 T provides enough field range above the AFM transition to clearly demonstrate the linear response. Figures S18, S19, S20 show that the linear-B behavior above $B_c$ exists for a broad angle range at temperatures below (Figure S18) and well above (Figures S19 and S20) the Néel temperature.

FIG. S18. Magnetotropic coefficient of RuCl$_3$ as a function of field and angle at 1.3 K. A) Frequency versus magnetic field for fields applied at various angles between the $c$-axis (0) and the honeycomb planes (90). B) The field-derivative of the data in A showing that the magnetotropic coefficient is constant above the AFM transition. C) The slope of the data in A determined for narrow field ranges around 5 and 32 T. The behavior agrees with D) the angle dependence observed of the magnetotropic coefficient under continuous rotation of the sample in magnetic fields ranging from 2.5 to 34.5 T.
FIG. S19. Magnetotropic coefficient of RuCl$_3$ as a function of field and angle at 20 K. A) Frequency versus magnetic field for a wide range angles. B) The field-derivative of the data in A shows an approach to saturation at high fields. C) The slope of the data in A determined for narrow field ranges around 5 and 32 T. The behavior is consistent with D) the angle dependence of the magnetotropic coefficient as the sample is continuously rotated in magnetic fields ranging from 5 to 34.5 T.
FIG. S20. Magnetotropic coefficient of RuCl$_3$ as a function of field and angle at 40 K. A) Frequency versus magnetic field for various angles ranging from near the $c$-axis ($0^\circ$) and the honeycomb planes ($90^\circ$). B) The field-derivative of the data in A shows an approach to saturation at high fields when magnetic field is applied near the honeycomb planes. For magnetic field applied near the $c$-axis, saturation is pushed to an inaccessible field range. C) The slope of the data in A determined for narrow field ranges around 5 and 32 T. The behavior is consistent with D) the angle dependence of the magnetotropic coefficient as the sample is continuously rotated in magnetic fields of 5, 25, and 34.5 T.
FIG. S21. Magnetotropic coefficient of RuCl$_3$ as the sample is rotated in fixed magnetic fields. A) Frequency shift (proportional to the magnetotropic coefficient) as a function of field orientation angle. $\theta$ is defined from the $c$-axis (i.e., $\theta = 90^\circ$ corresponds to magnetic field applied in the honeycomb planes). At 5 T, the expected $\cos 2\theta$ dependence is observed with a reduced amplitude (proportional the anisotropy) upon increasing temperature. B) At 25 T and 5 K, entrance into (out of) the AFM phase occurs at $\theta = 162^\circ$ ($\theta = 182^\circ$). Signatures of the anisotropy that gives rise to the AFM phase are apparent at temperatures greater than 40 K.
FIG. S22. Magnetotropic coefficient of RuCl$_3$ at $\theta =$ 9.7° for several temperatures. At 1.3 K, the AFM phase boundary is traversed at roughly 18.5 T. At 20 K, the magnetotropic coefficient converges with the 1.3 K data above the AFM transition field. The high-field slope in the 40 K curve saturates to that of the lower temperature data, but with a larger field of approximately 30 T.
Figure 3 in the main text shows a scale-invariant free energy at temperatures and fields above a \( \sim 10 \) K energy scale, \( k = B \times f(B/T) \). Figure 3A indicates \( f(x) = b + \langle \text{smaller terms} \rangle \) at \( x \gg 1 \) where \( b \) is constant. Figure 3B shows that high-field scans at different temperatures are not asymptotically convergent to the same line. Instead, they have a linear-in-temperature offset, \( \delta K = aT \) where \( a \) is a negative constant. Such a high-field offset is consistent with simple \( B/T \) scaling, requiring only that \( f(x) \approx b+a/x+\langle \text{even smaller terms} \rangle \) at \( x \gg 1 \). Therefore, Figure 3 suggests that

\[
\begin{align*}
    f(x) &= b + a/x + \cdots, & x \gg 1, \\
    &= cx + \cdots, & x \ll 1.
\end{align*}
\]  

We note that for isolated spins, where \( f(x) \) is the Brillouin function, \( a = 0 \).
Appendix S8: Mechanics of the oscillating lever and associated experimental checks

Resonant measurement of the magnetotropic coefficient \( k = d^2 F / d\theta^2 \) of the sample relies on the lever being well-described by a simple harmonic oscillator with frequency \( \omega_0 \),

\[
E = I \left( \frac{d\Delta \theta}{dt} \right)^2 + K \Delta \theta^2, \quad \omega_0^2 = K/I. \tag{S1}
\]

Here, \( \Delta \theta(t) \) is the angle of rotation at the tip of the lever where the sample is attached. The finite magnetic anisotropy of the sample introduces a (small) additional field-dependent potential energy term \( (k/2) \Delta \theta^2 \), where \( k \) is the magnetotropic rigidity of the sample. The magnetotropic coefficient \( k \) of the sample is responsible for a small field- and angle-dependent frequency shift\(^{17}\),

\[
\frac{\Delta \omega}{\omega_0} = \frac{k}{2K}. \tag{S2}
\]

The bending shape of the lever near the resonance \( \omega_0 \) is not the same as the shape of a static lever under a fixed load at the tip, which would trace out a circular arc. The “effective bending stiffness” of the lever, \( K \), and the “effective moment of inertia” \( I \) depend not only on the density, shape, and elastic properties of the lever, but also on the shape of the resonance mode itself. In this section, we provide a self-contained discussion of the mechanics of the lever and associated experimental checks. In particular, we make direct estimates for the bending stiffness of the Akiyama lever used in this work, which allows us to independently validate this relatively new probe of the magnetotropic coefficient.

1. Thin-beam approximation

Throughout this discussion, we shall assume that the beam is thin, \( h \ll L \). The ratio of the thickness to the length of the lever \( h/L \) is about 1/80 for the Akiyama levers used in this work (Figure S23). This is sufficiently small to justify the “thin-beam” approximation\(^{35}\). In the thin-beam approximation, the shape of the lever as it bends is completely described by
a single function $\zeta(z)$ which gives the vertical ($x$-axis) displacement at a distance $z$ from the point of attachment. (Figure S23). The thin-beam approximation simplifies not only the description of the bending state of the lever, but also the elastic equations. As will be discussed below, the elastic energy depends on the local curvature of $d^2\zeta(z)/dz^2$ only.

The boundary conditions on a free surface of the lever is that the perpendicular component of force is zero. For a thin lever, these effectively require that all but one stress components are vanishingly small: the boundary conditions require that all $\sigma_{ij}(x, z)$ (except $\sigma_{zz}(x, z)$), must vanish on both sides of the lever at $x = \pm h/2$, therefore the Taylor expansion of their dependence on $x$ must start from a quadratic term $x$, and, therefore these are vanishingly small everywhere inside the thickness of the lever, $|x| < h/2$. In contrast, for $\sigma_{zz}$, the only component that is not constrained by the boundary conditions on the large surfaces of the lever, Taylor expansion is not constrained: $\sigma_{zz}(x, z) = a(z) + b(z)x$. The elastic equations will require that $\sigma_{zz}(x, z)$ vanishes at a particular depth $x_0(z)$ inside the lever which defines a “neutral plane”. For a lever of rectangular crosssection the neutral plane is the mid-plane of the crosssection of the lever, which we set at $x = 0$. Therefore, $\sigma_{zz}(x, z) = b(z)x$.

The elastic strain is found from the elastic equations. In a cubic crystal (silicon),

$$
\begin{align*}
\sigma_{xx} + \sigma_{yy} + \sigma_{zz} =& (c_{11} + 2c_{12})(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}) \\
\sigma_{xx} - \sigma_{yy} =& (c_{11} - c_{12})(\varepsilon_{xx} - \varepsilon_{yy}) \\
\sigma_{xx} + \sigma_{yy} - 2\sigma_{zz} =& (c_{11} - c_{12})(\varepsilon_{xx} + \varepsilon_{yy} - 2\varepsilon_{zz}) \\
\sigma_{i\neq j} =& c_{66}\varepsilon_{i\neq j},
\end{align*}
$$

where each line corresponds to one of the irreducible components of the stress/strain tensors.
The last equation sets all off-diagonal components of strain to zero because all off-diagonal components of stress belong to the same irreducible representation. These equations can be solved for the diagonal components of strain

\[ \varepsilon_{zz} = \frac{1}{E} \sigma_{zz} \]

\[ \varepsilon_{xx} = \varepsilon_{yy} = -\sigma = -\frac{\sigma}{E} \sigma_{zz} \]  \hspace{1cm} (S4)

which defines Poisson’s ratio \( \sigma \) and Young’s modulus \( E \). In particular,

\[ E = \frac{(c_{11} - c_{12})(c_{11} + 2c_{12})}{c_{11} + c_{12}} \]

\[ \sigma = \frac{c_{12}}{c_{11} + c_{12}} \]  \hspace{1cm} (S5)

The elastic energy (per unit volume) is \( dE/dV = (1/2) \varepsilon_{ij} \sigma_{ij} \), or

\[ dE/dV = \frac{1}{2} \varepsilon_{zz} \sigma_{zz} = \frac{1}{2E} \sigma_{zz}^2 \]  \hspace{1cm} (S6)

Since \( \sigma_{zz}(x) = bx \) is linear in \( x \), \( \varepsilon_{zz} \) is also linear in \( x \) (Eq. S4), \( \varepsilon_{zz} = (b/E)x \). Therefore, the bent shape of any short segment of the lever is an arc of a circle. The radius of curvature of the circle at a point \( z \) is determined by \( d^2 \zeta/dz^2 \) via \( \delta x = (\delta z)^2/2R \),

\[ \frac{1}{R} = \frac{d^2 \zeta(z)}{dz^2} \].  \hspace{1cm} (S7)

The strain is \( \varepsilon_{zz}(x, z) = x/R(z) \) and the elastic energy (per unit volume) is \( dE/dV = (E/2R^2)x^2 \). Finally, the elastic energy of a short segment of the lever is determined by the integral over the cross-section,

\[ \frac{dE}{dz} = \frac{E}{2R^2} m_y \]  \hspace{1cm} (S8)

where \( m_y \) is the moment of inertia of the cross-section \( m_y = \int x^2 dA \). For a rectangular cross-section, \( m_y = Ah^2/12 \). Adding the kinetic energy, we obtain the total energy of the lever

\[ E_{tot} = \frac{\rho A}{2} \int dz \left( \frac{\partial \zeta}{\partial t} \right)^2 + \frac{E m_y}{2} \int dz \left[ \Delta \zeta \right]^2 \].  \hspace{1cm} (S9)

This expression is used for a uniform lever: \( \rho, A, E, \) and \( m_y \) are constant along the lever. A non-uniform lever, such that the cross-sectional area \( A, E, \rho \) and \( m_y \) can vary along \( z \) is described by

\[ E_{tot} = \frac{1}{2} \int dz \rho(z) A(z) \left( \frac{\partial \zeta}{\partial t} \right)^2 + \frac{1}{2} \int dz E(z) m_y(z) \left[ \Delta \zeta \right]^2, \]  \hspace{1cm} (S10)
which can be used to estimate the resonant frequencies of levers with more complex shapes. For example, if the lever consists of two uniform segments \((A(z)\) and \(m_y(z)\) have a single sharp step), in addition to the continuity of \(\zeta(z)\) and \(d\zeta(z)/dz\), the equations of motion require continuity of \(Em_y \left( \frac{\partial}{\partial z} \right) [\Delta \zeta(z,t)]\) and \(Em_y \Delta \zeta(z,t)\) across the step point.

2. Resonating lever

Eq. S9 requires that \(\zeta(z,t)\) satisfies

\[
\frac{d^2 \zeta(z,t)}{dt^2} - \left( \frac{Em_y}{\rho A} \right) \frac{d^4 \zeta(z,t)}{dz^4} = 0. 
\] \hspace{1cm} (S11)

Near the resonance, the time-dependence is a simple periodic function of time

\[
\zeta(z,t) = \zeta(z) \cos(\omega t + \varphi), 
\] \hspace{1cm} (S12)

where \(\omega^2\) is an eigenvalue of the bilaplacian

\[
\omega^2 = \kappa^4 \left( \frac{Em_y}{\rho A} \right), \quad \kappa^4 \zeta(z) = \frac{d^4 \zeta(z)}{dz^4}. 
\] \hspace{1cm} (S13)

We have

\[
\zeta(z) = A \cos \kappa z + B \sin \kappa z + C \cosh \kappa z + D \sinh \kappa z 
\] \hspace{1cm} (S14)

The boundary conditions for a lever clamped at \(z = 0\) and free at the other end \(z = L\) are

\[
\frac{d^2 \zeta(L)}{dz^2} = 0, \quad \frac{d^3 \zeta(L)}{dz^3} = 0, \quad \frac{d \zeta(0)}{dz} = 0, \quad \zeta(0) = 0, 
\] \hspace{1cm} (S15)

which requires

\[
-A \cos \kappa L - B \sin \kappa L + C \cosh \kappa L + D \sinh \kappa L = 0 \newline
A \sin \kappa L - B \cos \kappa L + C \sinh \kappa L + D \cosh \kappa L = 0 \newline
B + D = 0 \newline
A + C = 0. 
\] \hspace{1cm} (S16)
This set of equations has a nontrivial solution only when the determinant of a matrix

$$\begin{vmatrix}
-\cos \kappa L & -\sin \kappa L & \cosh \kappa L & \sinh \kappa L \\
\sin \kappa L & -\cos \kappa L & \sinh \kappa L & \cosh \kappa L \\
0 & 1 & 0 & 1 \\
1 & 0 & 1 & 0
\end{vmatrix}$$

(S17)

vanishes,

$$1 + \cos \kappa L \cosh \kappa L = 0. \quad \text{(S18)}$$

This is the equation for a set of resonant frequencies via Eq. S13. The infinite set of $\kappa_n$ in Eq. S18 is given accurately by

$$\kappa_0 L = 0.597 \pi, \quad \kappa_n=1,2,\ldots, L \approx (n + 1/2) \pi. \quad \text{(S19)}$$

The lowest frequency mode is given

$$\omega_0 = \frac{\hat{\kappa}_0^2}{L^2} \sqrt{\frac{E m_y}{\rho A}}, \quad \omega_0^\square = 1.01 \frac{h}{L^2} \sqrt{\frac{E}{\rho}}, \quad \text{(S20)}$$

where $\hat{\kappa} = \kappa L$ is dimensionless value of $\kappa$. The second equation defines the frequency $\omega_0^\square$ of a lever with (uniform) rectangular crosssection (represented by superscript $\square$) of thickness $h$, for which $m_y/A = h^2/12$. The numeric coefficient in the second formula, $(0.597 \pi)^2/\sqrt{12} \approx 1.01$.

For the quartz fork leg (length= 2400 µm, width= 220 µm, thickness= 130 µm, $E \sim 70$ GPa, $\rho \sim 2.63$ g/cm³) one gets for the horizontal (“scissors”) mode $f_0 = 31.67$ kHz, ($f = \frac{1}{2\pi} \sqrt{\frac{220 \mu}{(2400 \mu)^2} \sqrt{\frac{70 \text{ GPa}}{2.63 \text{ g/cm}^3}}}$). The frequency of the series of higher modes starting at $\omega_0$ are given by $f_0/(0.597)^2 \times \{(0.597)^2, (1 + 1/2)^2, (2 + 1/2)^2, (3 + 1/2)^2, \ldots\}$. This estimate fits well with the frequency of the free quartz fork, designed to have $f_0 = 32768$ Hz = $2^{15}$ Hz in vacuum.

For the Akyama silicon lever ($h = 3.7 \mu$m, $L = 290 \mu$m, $E \sim 125$ GPa, $\rho \sim 2.33$ g/cm³), one gets $f_0 = 51.8$ kHz ($f = \frac{1}{2\pi} \sqrt{\frac{3.7 \mu}{(290 \mu)^2} \sqrt{\frac{125 \text{ GPa}}{2.33 \text{ g/cm}^3}}}$). Similarly, the higher frequency modes follow the sequence proportional to $f_0/(0.597)^2 \times \{(0.597)^2, (1+1/2)^2, (2+1/2)^2, (3+1/2)^2, \ldots\}$.
The shape of the silicon tip in the Akiyama probe is in fact not rectangular. The geometry can be better approximated with two segments of different width (but are otherwise identical in $h = 3.7 \mu m$, $E \sim 125$ GPa, $\rho \sim 2.33 \text{g/cm}^3$) stitched together along the length of the lever: $L_1 \approx 200 \mu m$, $w_1 \approx 60 \mu m$, and $L_2 \approx 90 \mu m$, $w_2 \approx 90 \mu m$. We can still use Eq. S13 with the boundary conditions discussed after Eq. S10. Here, $m_y$ and $A$ vary along the lever, keeping the ratio $m_y/A = h^2/12$ fixed. If we parameterize the shape of the two segments so that the width is a fraction $bw$ over a first part $aL$ of the lever’s length $L$ and it is $w$ over the rest $(1-a)L$, the eigenvalue equation takes the form

$$
(b^2 - 1) \cos(a\pi\kappa) \cosh(a\pi\kappa) + \cosh((a-1)\pi\kappa)\left[(b^2 - 1) \cos((a-1)\pi\kappa) \\
+ \cosh(a\pi\kappa)\left((b^2 + 1) \cos((a-1)\pi\kappa) \cos(a\pi\kappa) \\
+ 2b \sin((a-1)\pi\kappa) \sin(a\pi\kappa)\right)\right] + 2b \cos(a\pi\kappa) \sinh((1-a)\pi\kappa) \sinh(a\pi\kappa) + b^2 + 1 = 0,
$$

which reduces to $\cos(a\pi\kappa) \cosh(a\pi\kappa) + 1 = 0$ when $a = 0$ or $a = 1$. For $a = 0.7$ and $b = 0.66$ (Figure S23), we obtain $\kappa = 0.549$, somewhat smaller than $\kappa = 0.597$ for the uniform-cross-section lever. The frequency is given by the same expression as in the uniform cross-section case, $\omega_0 = ((0.549\pi)^2/\sqrt{12})(h/L^2)\sqrt{E/\rho}$. We obtain for the lowest frequency, $f_0 = (0.549/0.597)^251.8$ kHz = 44.2 kHz. This shows that the observed frequency of the silicon lever, 43-46 kHz, can be reasonable well understood using the thin-beam approximation and the “clamped-end” boundary conditions.

3. The effective bending stiffness and moment of inertia of the lever

When the driving frequency is close to a resonant mode frequency, the shape of the lever is approximated by $\zeta(z, t) = \zeta^{(n)}(z)e^{i\omega^{(n)}t}$, where $\omega^{(n)}$ were discussed above and $\zeta^{(n)}(z)$ are found from Eq. S16,

$$
A = -C = \cos \kappa L + \cosh \kappa L, \quad B = -D = \sin \kappa L - \sinh \kappa L
$$

or

$$
\zeta^{(n)}(z) = N_n \left[(\cosh \kappa_n z - \cos \kappa_n z) - \frac{\sinh \kappa_n L - \sin \kappa_n L}{\cos \kappa_n L + \cosh \kappa_n L} (\sin \kappa_n z - \sinh \kappa_n z)\right].
$$
The effective bending stiffness $K^{(n)}$ and the effective moment of inertia $I^{(n)}$ are defined by the representation of the lever’s energy as

$$ E = \frac{I^{(n)}}{2} \left( \frac{\partial \Delta \theta}{\partial t} \right)^2 + \frac{K^{(n)}}{2} \Delta \theta^2, \quad (S24) $$

where $\Delta \theta$ is the bending angle of the lever at the tip, $\Delta \theta(t) = (d\zeta(z,t)/dz)_{z=L}$. To calculate the values of $K^{(n)}$ and $I^{(n)}$ we need to evaluate the kinetic and potential energy of the lever, Eq (S9), where the normalization factor $N_n$ is chosen so that $[d\zeta^{(n)}(z)/dz]_{z=L} = \Delta \theta$:

$$ N_n = \Delta \theta \left[ 2\kappa_n \frac{\cosh \kappa_n L \sin \kappa_n L + \cos \kappa_n L \sinh \kappa_n L}{\cos \kappa_n L + \cosh \kappa_n L} \right]^{-1}, \quad (S25) $$

where expression in $[...]$ is equal to the derivative of the square bracket $d[...]/dz$ in Eq. (S23) for $z = L$. For the kinetic energy, we need to calculate

$$ \int_0^L dz \left( \frac{\partial \zeta^{(n)}(z)}{\partial t} \right)^2 = (a^{(n)}/\kappa_n^3) \left( \frac{\partial \Delta \theta}{\partial t} \right)^2, \quad (S26) $$

which defines numeric parameter $a^{(n=0,1,2,...)} = \{0.870, 1.13, 1.97, ...\}$ for each mode. We have

$$ I^{(n)} = a^{(n)} \rho AL^3/\tilde{\kappa}_n^3. \quad (S27) $$

For the elastic energy we need to calculate

$$ \int_0^L dz \left( \frac{\partial^2 \zeta^{(n)}(z)}{\partial z^2} \right)^2 = \kappa^4 \int_0^L dz \left( \zeta^{(n)}(z) \right)^2 = \kappa_n a^{(n)} \Delta \theta^2. \quad (S28) $$

Therefore,

$$ K^{(n)} = \tilde{\kappa}_n a^{(n)} E_{my}/L. \quad (S29) $$

We can check that the oscillation frequency given by Eq. S1, $\omega^{(n)} = \sqrt{K^{(n)}/I^{(n)}} = \tilde{\kappa}_n \sqrt{E_{my}/\rho AL^4}$ is identical to Eq. S20).

For the Akiyama silicon lever, $E_{my}/L = 125 GPa \times (60 \mu m \times (3.7 \mu m)^3/12)/290 \mu m = 109 nJ$. For the lowest mode ($a^{(0)} = 0.87$ and $\tilde{\kappa}_0 = 0.6\pi$), we get

$$ K^{(0)} = 180 nJ. \quad (S30) $$
4. Effects of the surrounding gas and liquid

All measurements in this work were performed either in liquid helium for temperatures at and below 4.2 K, or in helium exchange gas of ∼5-10 mbar for temperatures above 4.2 K. These have two main effects on the measurement – increasing the linewidth through viscous dissipation and shifting the frequency via the attached liquid/gas boundary layer. The viscous broadening of the resonance width also leads to a frequency shift via Kramers-Kronig relations.

The Reynold’s number of a vibrating lever is small ∼0.5 in air (1kg/m³ * 300μ * 1μ * 50kHz/2 × 10⁻⁵Pasec) and ∼5 in liquid He-4 at 2K. We can use Stoke’s formula to determine the viscous force. The resonance linewidth can be estimated via

\[
\Gamma = f \times \frac{\text{energy dissipated per cycle}}{\text{energy stored}} = f \times \frac{6\pi \eta R f u^2}{K(u/L)^2} = f \times \frac{6\pi \eta \sqrt{wL} f L^2}{K},
\]

which is about 30 Hz in air at room temperature and about 3 Hz in liquid He-4 at 2K. The commercial liquid/gas “sticky” layer thickness of the lever can be compared to the amplitude of oscillations of the lever at the tip of the lever—it is 5-10 μm at 1 V driving voltage (determined using laser beam reflection), whereas the typical drive voltage in the experiment is 10 mV. Therefore, we can use 0.2 μm as the thickness of the liquid/gas ”sticky” layer that is carried with the lever as it oscillates. The main effect here is to increase the effective moment of inertia \( I \) in Eq. S1. To estimate the additional moment of inertia of the liquid/gas, we note that a “sticky” layer of liquid He with thickness \( \delta \) is equivalent to an extra silicon thickness of \( (\rho_l/\rho) \times \delta \approx 0.01 \mu m \) (\( \rho_l \) and \( \rho \) are the He-4 and silicon densities, respectively). Because \( I \) is proportional to the thickness of the lever (Eq. S24), the addition of the sticky layer leads to a relative increase in \( I \) of \( \delta I/I = 2 \times 0.01\mu m/3.7\mu m \approx 0.005 \). Therefore, we expect that the frequency shifts down
as we go from surrounding He-4 gas to liquid by about 0.2%. The observed frequency shifts by about 1% after He4 condenses into liquid.

Finally, large changes in the resonance line-width can lead to shifts in frequency not directly associated with changes in the sample’s magnetotropic coefficient. These follow from the causal structure of the response measured by this probe (Kramers-Kronig relations). The frequency shift is comparable in magnitude to the change in the resonance line-width. For example, evacuating air around the lever at room temperature shifts the frequency up by about 20Hz, comparable to the associated decrease in line-width. These effects become more significant in He3-He4 mixtures at mK temperatures.

5. Effects of Earth’s gravity and a magnetic field gradient

As the lever oscillates near the \( n^{th} \) resonance frequency, each point moves vertically (perpendicular to the lever) according to \( \zeta_{\perp}(t) = A\zeta^n(z)\cos(\omega^{(n)}t) \). However, at the same time, there is also a horizontal motion along the lever by a small amount \( \zeta_{\parallel}(z,t) \propto (\zeta_{\perp}(z,t))^2 \),

\[
\zeta_{\parallel}(z,t) = \int_0^z dz' \sqrt{1 - \left( \frac{\partial \zeta_{\perp}(z')}{\partial z'} \right)^2} \approx -\frac{\zeta_{\perp}^2(z)}{2R_{\parallel}(z)},
\]

\[
\frac{1}{R_{\parallel}(z)} = \frac{1}{\zeta_{\perp}^2(z)} \int_0^z dz' \left( \frac{\partial \zeta_{\perp}(z')}{\partial z'} \right)^2,
\]

where \( \zeta_{\perp}(z) \) is itself an arc of a circle, \( \zeta_{\parallel}(z) \approx Az^2 \); for such shape the radius of curvature of the trajectory of the end-point is always \( R_{\parallel} = (3/4)L \).
Because \( \zeta_\parallel (z) \) is proportional to \( \zeta_\perp (L)^2 \) (and therefore \( \Delta \theta^2 \)), the bilinear-in-\( \Delta \theta \) term in the energy of the lever has Earth’s gravity component,

\[
\delta E_{G,\text{lever}} = \rho A g \cos \theta \int dz \zeta_\parallel (z) = \frac{1}{2} m (b^{(n)} L) \ g \cos \theta \Delta \theta^2 ,
\]

where the \( m = \rho A L \) is the mass of the lever, and \( b^{(n)} L \) is the distance to the “effective center of mass”, \( b^{(0)} \approx 0.21 \) for the zeroth mode of the rectangular-crosssection lever. Therefore, there is a weak frequency shift \( \Delta f \propto \cos \theta \) where \( \theta \) is the angle between the lever and the direction of gravitational force. For an Akiyama lever of mass \( m_A \approx 150 \ \text{ng} \) we have \( \Delta f / f \approx m_A g (b^{(0)} L) / 2K \approx 10^{-6} \) or about 50 mHz at \( f_0 \approx 50 \ \text{kHz} \). Similarly, one can estimate the angle-dependent part of the frequency shift due to a finite mass \( m_S \) of the sample,

\[
\delta E_{G,\text{sample}} = \frac{1}{2} m_S (c^{(n)} L) \ g \cos \theta \Delta \theta^2
\]

were \( c^{(0)} \approx 0.61 \).

Finally, if the lever is off-center in the magnet, there will be frequency shift associated with the magnetic-field-gradient, magnetic force = \( \nabla F(B) \). If we denote the projection of this force vector onto the direction of the lever as \( [\nabla F(B)]_\parallel \), then the magnetic-force term in the energy is obtained by replacing \( m_S g \cos \theta \) in Eq. (S34) with \( [\nabla F(B)]_\parallel \),

\[
\delta E_{\nabla B^2} = \frac{1}{2} [\nabla F(B)]_\parallel (c^{(0)} L) \Delta \theta^2
\]

the frequency shift due to the field gradient will be about \( L d / L_M^2 \) times smaller than the frequency shift due to the magnetotropic rigidity of the sample (\( L_M \) is the size of the magnet coil, \( d \) is the off-center distance). These effects are negligible for the magnetotropic coefficient measurements in this work.

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the Cl-Cl link in the square Ru-Cl-Ru-Cl plaquette, and $\alpha = \hat{z}$ points perpendicular to the plaquette. The latter is the Kitaev spin-anisotropy term. The magnitude of $J_\alpha$ is the same on all links (for an undistorted honeycomb lattice), but the principle axes $\hat{\alpha}$ differ from link to link.

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