Oscillations of the thermal conductivity in the spin-liquid state of $\alpha$-RuCl$_3$

Peter Czajka$^{1,7}$, Tong Gao$^{1,7}$, Max Hirschberger$^{1,6}$, Paula Lampen-Kelley$^{2,3}$, Arnab Banerjee$^{4,5}$, Jiaqiang Yan$^{3}$, David G. Mandrus$^{2,3}$, Stephen E. Nagler$^{6}$ and N. P. Ong$^{1,3}$

In the class of materials called spin liquids$^{1-3}$, a magnetically ordered state cannot be attained even at millikelvin temperatures because of conflicting constraints on each spin; for example, from geometric or exchange frustration. The resulting quantum spin-liquid state is currently of intense interest because it exhibits unusual excitations as well as wave-function entanglement. The layered insulator $\alpha$-RuCl$_3$ orders as a zigzag antiferromagnet at low temperature in zero magnetic field$^1$. The zigzag order is destroyed when a magnetic field is applied parallel to the zigzag axis. At moderate magnetic field strength, there is growing evidence that a quantum spin-liquid state exists. Here we report the observation of oscillations in its thermal conductivity in that field range. The oscillations, whose amplitude is very large within this field range and strongly suppressed on either side, are periodic. This is analogous to quantum oscillations in metals, even though $\alpha$-RuCl$_3$ is an excellent insulator with a large gap. As the temperature is raised above 0.5 K, the oscillation amplitude decreases exponentially, anticorrelating with the gap. Above about 2 K, we recorded using the stepped-field method both the thermal resistivity $\lambda_\parallel$ and thermal Hall resistivity $\lambda_\perp$ as $H$ was slowly varied at fixed $T$ (Methods and Extended Data Fig. 1). As seen in Fig. 1b (for Sample 1 with $H \parallel a$), strong oscillations emerge in $\lambda_\parallel(H)$. Below 2 K, the data were recorded continuously, as well as with the stepped-field method (Extended Data Fig. 1d,e). The data, plotted as $\Delta\kappa/H$ in Fig. 1c, show that the oscillation amplitudes continue to grow as $T$ decreases until they comprise 30–60% of $\kappa_{\parallel}$ at 0.43 K. At around 11.5 T, $\kappa_{\parallel}$ displays a step increase to a flat plateau. In the high-field partially polarized state, where $\kappa_{\parallel}$ is dominated by the phonon conductivity $\kappa_{\parallel}\equiv\kappa_{\parallel}$, oscillations are rigorously absent. The plateau value of $\kappa_{\parallel}$ is a benchmark for sample quality (see below and Methods). Similar curves are observed in Sample 3.

The oscillation amplitudes are strongly peaked in the QSL state. To extract the amplitude, we first determined the smooth background curve $\kappa_{\parallel}(T, H)$ threading the midpoints between adjacent extrema (Extended Data Fig. 2d). The oscillatory component, defined as $\Delta\kappa := \kappa_{\parallel} - \kappa_{\parallel}$ (Extended Data Fig. 2c), allowed accurate determination of the amplitude $\Delta\kappa_{\parallel}$ which we plot in Fig. 1d for Sample 1. Above 6 T, $\Delta\kappa_{\parallel}$ rises steeply to peak at 9.6 T, followed by an abrupt collapse to zero above 11 T. Below 6 T, a weak remnant ‘tail’ survives to 4 T in a mixed state in which small QSL regions coexist with the zigzag state (we note that 4 T is roughly where the averaged zigzag Bragg intensity begins to weaken with $H$ (ref. 1)). By its profile, $\Delta\kappa_{\parallel}$ is largest within the field interval (7.3, 11.5) T of the QSL state. The profile in Sample 3 is similar (Extended Data Fig. 3a). A fourth sample did not exhibit oscillations with $H$ tilted at 45° relative to $a$ (Table 1).

Next, we show that the oscillations are periodic in $1/H$. Figure 2a displays plots of the integer increment $\Delta n$ versus $(\mu_0 H_a)^{-1}$, where $H_a$ are fields locating extrema of $d\kappa_{\parallel}/dH$ plotted in Fig. 2b. First, we focus on data shown as solid symbols. The data from Samples 1 and 3, measured with $H \parallel a$, fall on a curve composed of straight-line segments separated by a break in slope at around 7 T. The slopes $S_i$ of the straight segments are 41.4 T ($H > 7$ T) and 30.6 T ($H < 7$ T).

$^1$Department of Physics, Princeton University, Princeton, NJ, USA. $^2$Department of Materials Science and Engineering, University of Tennessee, Knoxville, TN, USA. $^3$Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN, USA. $^4$Department of Physics, Purdue University, West Lafayette, IN, USA. $^5$Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, TN, USA. $^6$Present address: Department of Applied Physics and Quantum-Phase Electronics Center, The University of Tokyo, Tokyo, Japan. $^7$These authors contributed equally: Peter Czajka, Tong Gao.
e-mail: npo@princeton.edu
Quantum oscillations in the QSL phase in α-RuCl₃ (Sample 1). a. The phase diagram showing the QSL phase (shaded red and orange) sandwiched between the zigzag (purple) and polarized states (navy blue) with H || a (axes a and b shown in inset). The ZZ2 phase that lies between critical fields $B_1$ and $B_2$ is outlined by the blue dashed curve. The yellow region is the paramagnetic state. The inset shows the zigzag order (red and blue arrows are local moments on Ru). b. The emergence of oscillations in $\kappa_{xx}(H)$ (solid circles) is strikingly prominent in the QSL state. Its profile (shaded orange) distinguishes the QSL from adjacent phases. A weak remnant tail extends below 7 T to 4 T in the zigzag state.

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**Table 1 | Dimensions of the four crystals investigated**

| Sample | Length $L_x$ (µm) | Width $w$ (µm) | Thickness $d$ (µm) | Separation $w_c$ (µm) | H  |
|--------|-------------------|----------------|-------------------|----------------------|----|
| 1      | 860               | 2,870          | 60                | 1,870                | $a, \theta = 39^\circ$, $55^\circ$ |
| 2      | 400               | 1,500          | 50                | 800                  | $b$            |
| 3      | 1,060             | 2,760          | 80                | –                    | $a$            |
| 4      | 870               | 1,700          | 30                | 700                  | $\theta = 45^\circ$ |

$L_x$ is the separation of longitudinal contact pads, $w$ is the crystal width and $w_c$ is the separation of the transverse (Hall) contact pads. In the last column, H is either $a$ or $b$ or at angle $\theta$ relative to $a$ in the a–c plane. Sample 4 is considerably thinner than the others.

When $H \parallel b$ (Sample 2, green circles), similar behaviour is obtained, with the low-field slope $S_l$ also at 30.6 T. However, the high-field slope is steeper with $S_h = 64.2$ T. As shown in Fig. 1b,c, the periods are independent of $T$ from 0.43 K to 4.5 K.

Taken together, the data shown in Fig. 2a–d provide strong evidence that the oscillations are intrinsic and reproducible across samples. The five datasets shown in Fig. 2a were derived from extrema of the derivative curves $d\kappa_{xx}/dB$ displayed in Fig. 2b. The profiles show the close agreement in both period and phase between Samples 1 and 3. The matching of the extrema is especially evident in Fig. 2c, which also shows that periodicity versus $H$ (as opposed to $H/H$) can be excluded. In Sample 2, the period and phase also agree with those of Samples 1 and 3 for $H < 7$ T (the period is shorter above 7 T, as noted already).

Oscillations observed with $H$ tilted in the a–c plane (at an angle $\theta$ relative to $a$) provide tests in an independent direction. Figure 2c shows curves of $\kappa_{xx}$ measured in Sample 1 with $\theta = 0^\circ$, $39^\circ$ and $55^\circ$ (curves of $\kappa_{yy}$ at various $T$ are in Extended Data Fig. 4a,b). By plotting the curves versus $H/H$, we find that the periods match quite well (with a possible phase shift for the curve at $55^\circ$).
The corresponding derivatives at $39^\circ$ and $55^\circ$ are plotted in Fig. 2b. We infer that, in tilted $\mathbf{H}$, the periods depend only on $H_x$. Moreover, the close agreement of the curves for $\theta = 0^\circ$ and $\theta = 39^\circ$ strongly supports an intrinsic origin.

In Methods, we discuss why the oscillations here are unrelated to those found in experiments on the narrow-gap correlated insulator SmB$_6$ (ref. 23). For the mechanism proposed to be at work in $\alpha$-RuCl$_3$, we would need $H$ in excess of 10,000 T. Also, we discuss in the Supplementary Information the evidence against artefactual origins, such as stacking faults produced by field-induced strain.$^{24,25}$.

The profile of $\Delta\kappa_{\text{amp}}$ versus $H$ actually imposes a tight constraint on possible mechanisms. Above 11.5 T in the polarized state, the oscillations vanish abruptly. Below $H_{\text{QSL}}$, the oscillations survive as a weak tail extending to 4 T in the zigzag state. The amplitude profile suggests that the oscillations are intrinsic to the QSL state. The 1/$H$ periodicity suggests an intriguing analogy with SdH oscillations, despite the absence of free carriers. We note that Landau-level oscillations in the QSL state of $\alpha$-RuCl$_3$ is widely anticipated.$^{30–32}$ Our finding that $S_z$ is determined by $H_y$ suggests either a fully three-dimensional QSL state or possibly a different mechanism. Regardless, quantization of a Fermi surface occupied by spin excitations is currently the most promising explanation of the oscillations.

**Fig. 2 | Periodicity and intrinsic nature of oscillations.** (a) The integer increment $\Delta n$ versus $1/H_y$ (or $1/H_x$), where $H_y$ are the fields identifying extrema of the derivative curves $d\kappa_{\text{amp}}/dB$ ($H_{\text{int}} = H_y \cos \theta$ for tilted $\mathbf{H}$). Solid symbols represent data taken with $\mathbf{H}$ strictly in-plane. The blue circles (Sample 1) and red stars (Sample 3) are plotted in Sample 2 with $\mathbf{H} \parallel a$. Open symbols are measurements in Sample 1 with $\mathbf{H}$ tilted in the $a$–$c$ plane at angles $\theta = 39^\circ$ (triangles) and $55^\circ$ (circles), relative to axis $a$. The datasets fall on the same segmented curve (comprised of line segments with slope $31\,\text{T}$ below $7\,\text{T}$ and $41\,\text{T}$ above $7\,\text{T}$ (solid black lines)). The exception is the high-field slope of $64\,\text{T}$ in Sample 2 with $\mathbf{H} \parallel a$. Arrows and vertical lines mark the values of $1/H_y$ and $1/H_x$, read off from the straight-line fits in (a) for integer increments $\Delta n$. (b) Curves of the derivative $d\kappa_{\text{amp}}/dB$ versus $1/H_y$ (or $1/H_x$) in arbitrary units (a.u.) for Samples 1, 2 and 3 ($H_y = H \cos \theta$). For Sample 1, we show $d\kappa_{\text{amp}}/dB$ measured with $\theta = 0^\circ$, $39^\circ$ and $55^\circ$. The extrema of $d\kappa_{\text{amp}}/dB$ are plotted in (a) for integer increments $\Delta n$. (c) Replot of integer increment $\Delta n$ versus $H_y$. (d) The effect of tilting $\mathbf{H}$ out of the plane by angle $\theta$ (relative to axis $a$) in Sample 1 at $T \approx 0.6\,\text{K}$. The curves are measured with $\theta = 0^\circ$ (blue), $39^\circ$ (purple) and $55^\circ$ (orange). When they are plotted versus $H_x$, the periods of the oscillations in $\kappa_{\text{amp}}$ match well for the three angles.
The value of $3$ correlates with a sixfold increase in the oscillation amplitude.

The dome-shaped profiles are similar to the planar thermal Hall effect reported in ref. 33, but in our experiment the values are not quantized.

The special role of $H_a$ (in determining the period) seems empirically related to the planar thermal Hall effect (PTHE), which appears only when $H \parallel a$. At a fixed $H$, the ratio $\Delta \kappa/\kappa_{xy}$ decays with $T$ at a rate that is consistent with an effective mass $m^* \approx 0.64 m_e$, where $m_e$ is the free electron mass (Fig. 3a). The decay in $\Delta \kappa/\kappa_{xy}$ is accompanied by a rapid growth in the PTHE observed with $H \parallel a$. Recently, ref. 35 reported that $\kappa_{xy}/T$ measured with $H \parallel a$ seems to be quantized, within a narrow interval in $T$ (3.8–6 K) and in $H$ ($10 T < \mu H < 11.2 T$). We have extended the PTHE experiment down to $T = 300$ mK to gain a broader perspective.

Below 4 K, it is necessary to use the method described in equations (2)–(6) in Methods to isolate the intrinsic thermal Hall signal $\delta_s$ (defined in equation (2)) from artefacts arising from hystereses in $\kappa_{xy}$, as shown in Fig. 3c, for example. For $H \parallel b$, the intrinsic thermal Hall signal is found to be zero for $0 T < H < 14 T$ and $0.3 K < T < 5 K$ (Fig. 3b, bottom panel). However, with $H \parallel a$, a finite $\delta_s$ emerges above around 2 K, as shown in the top panel of Fig. 3b. The antisymmetry of $\delta_s$ with respect to $H$ identifies it as a true PTHE. This is the thermal-conductivity analogue of the true planar anomalous Hall effect observed in ZrTe$_5$ (ref. 35).

Inverting the matrix $\lambda_s(H)$ to obtain $\kappa_s(H)$, we find that $\kappa_s$ displays a dome profile that grows with $T$ in the QSL phase (Fig. 3d).

![Image](https://example.com/image.png)

**Fig. 3 | The planar thermal Hall response.** a. The $T$ dependence of $\Delta \kappa/\kappa_{xy}$ at 8.4 T (blue circles and lines) and the planar thermal Hall conductivity $\kappa_{xy}$ at 9 T (red circles and lines). Arrows point to the relevant vertical axis for each parameter. The decrease of $\Delta \kappa/\kappa_{xy}$ with $T$ (consistent with an effective mass $m^*/m_e = 0.64$) is anticorrelated with the increase in $\kappa_{xy}$. b. Top: the emergence of the PTHE signal $\delta_s$ with $H \parallel a$. Red circles are data collected with $H$ increasing from left to right (red arrows); blue circles are collected with $H$ decreasing (blue arrows). Black circles are the average of the red and blue data. At $T = 4.03 K$, $\delta_s$ in Sample 1 (left axis) displays sharp peaks that are antisymmetric in $H$ for $H \parallel a$ (black circles). Corresponding values of $\lambda_s$ are on the right axis. Bottom: the null thermal Hall resistivity (expressed as the thermal Hall signal $\delta_s$) measured with $H \parallel a$ at 0.3 K, 2.6 K and 5 K in Sample 2. The total uncertainty in $\delta_s$ is 0.3 mK. c. The hysteresis in $\kappa_{xy}$ can contaminate $\kappa_{xy}$ if not properly subtracted. The right-going (purple) and left-going (red) scans have been antisymmetrized (AS) with respect to $H$. d. Values of $\kappa_s(H)$ derived from the measured tensor $\lambda_s$ are plotted for several $T$ from 3.4 K to 5.5 K. The dome-shaped profiles are similar to the planar thermal Hall effect reported in ref. 35, but in our experiment the values are not quantized.
Figure 3a,d provides a broad view of how the PTHE varies with $T$. Although the trends of our $\kappa_{xx}$ are consistent with those in ref. 33 (for example, the PTHE exists only with $H\parallel a$), we note that the strong $T$ dependence evident in Fig. 2d seems difficult to reconcile with a quantized value occurring in the interval 3.8–6 K. Where the two datasets overlap (4–5 K), our magnitudes are much smaller ($\kappa_{xx} \approx 0.3 \text{mW K}^{-2} \text{m}^{-1}$ versus 0.8 mW K$^{-2}$ m$^{-1}$ at $T = 5$ K).

In summary, we have observed quantum oscillations in $\kappa_{xx}$ in $\alpha$-RuCl$_3$ with $H$ in plane. The prominence of the amplitude in the interval ($7.3, 11$) T implies that the oscillations are specific to the QSL state. The amplitudes are largest when crystalline quality (measured by $\kappa_{xx}^\text{lat}/T$ at 1 K) is highest.

Online content
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Methods

Thermal conductivity matrix. In the initial step, we bond 5 mm (100 μm) Au wires to the crystal with Stycast 2850FT epoxy. Though Ag paint is convenient for attaching the Au wires to the sample, we found that it does not stick well to the smooth surfaces of the α-RuCl₃ crystals. Stycast tends to form large contact surfaces, which causes uncertainty in measures of the true dimensions, but the low thermal impedance of the contacts using Stycast is more important than the geometric uncertainty.

After the epoxy for the Au contacts has dried, one end of the crystal is thermally anchored to the brass bath by one of the Au wires using Stycast. Then, Lakeshore Cryotronics RX102A thermometers (1 kΩhm) are thermally attached to the remaining Au wires with Ag paint (Extended Data Fig. 1a). To regulate the bath temperature, we use the high-stability thermometer RX202, which has a small magnetoresistance. The resistances of all thermometers are read by contacting with 1-mm phosphor bronze wires, which have negligible thermal conductance. The power P applied to the warm edge of the crystal depended strongly on the bath temperature. Typical values of P were 225 mW (at a bath temperature of 0.36 K), 1.65 mW (at 0.78 K), 6.4 mW (at 1.31 K) and 22.5 mW (at 1.67 K).

Although the crystals are highly flexible, weak stresses on them can produce cracking faults that ruin their quality. We employ special care to minimize the applied stress during the mounting process. Essential support is provided by a Delrin post glued by stycast to the free end of the crystal (and anchored to the brass plate). The post prevents crystal bending from the torque τ = m × B in a strong field. The ultra-low thermal conductivity of Delrin and the large aspect ratio of the post minimize the heat current shunted by the post.

All parameters calibrated as a function of temperature and magnetic field using the polynomial-fitting procedure described in ref. 3. They are measured using Lakeshore temperature controllers. The low-noise lock-in models (LS370 and LS372) are employed to measure the transverse temperature difference ∆T ≡ T_x - T_y (the temperatures T_x and T_y are measured at the contacts shown in the sketch in Extended Data Fig. 1a. ∆T and ∆S denote temperature differences along the y and x axes, respectively). For the longitudinal temperature differences ∆T ≡ T_1 - T_0, the models LS340 and LS336 suffice.

Extended Data Fig. 1b shows a photo of the mounted Sample 2. The Delrin post and heater are visible in both panels. The 1 kΩ heater glued to the top of the crystal produces the thermal current density J. We define axes x̂∥[110] and ŷ∥[001] (normal to the a-b plane). The direction ŷ is the transverse 'Hall effect' axis. In analogy to the electrical case, the thermal resistivity matrix λ_ij relates the thermal gradient −∇T to J by

\[ \lambda_{ij} = \frac{J_i}{T_j - T_i} \]  

Ideally, the thermal resistivity matrix λ_ij is obtained by recording the two signals ∆T and ∆S at each value of H. Then, the thermal conductivity matrix is obtained as the reciprocal of the thermal resistivity matrix; that is, κ_ij = (x̄_T ̄x)_ij. At low T in α-RuCl₃, there are two important extraneous contributions to ∆T; namely, transient heating and cooling from magnetocaloric effects and large hysteretic effects. These two effects, which are 10–50 times larger than the intrinsic thermal Hall signal δ_T, have to be rigorously isolated and removed for reliable thermal Hall measurements.

Spin axes and bond axes. To clarify the relation between the spin axes S_x, S_y and S_z and the lattice axes a and b, we sketch in Extended Data Fig. 1c part of a unit cell isolating one of the honeycomb planes. Each Ru ion is enclosed in a Cl octahedron. The tilted planes shaded in blue and pink contain the X and Z Ru–Ru bonds, respectively. The spin axes are normal to their respective planes. The lower sketch in Extended Data Fig. 1c highlights one of the pink planes and the direction of the spin axis S_y normal to the plane.

Transients from magnetocaloric effect. We outline the protocol described in ref. 4 for high-resolution measurements of the thermal Hall quantities κ_xx and κ_yy. At cryogenic temperatures, changes in H or T engender very large entropy release or absorption by the spins. In addition, the step changes produce eddy current heating of the bath (brass). Both effects produce large thermoelectric transients, even when the applied step (H or T) is changed very gradually. We increment H step-wise (by ±125 mT) and hold it fixed (for example, for 420 s in Extended Data Fig. 1e), with the direct-current heater current continuously on. A step-increase in [H] causes a large amount of entropy to be released by the spins, by the magnetocaloric effect, which produces considerable transient warming of the phonons. Conversely, a step decrease in [H] leads to entropy absorption and cooling of the phonons. By contrast, eddy current heating (in the brass bath) always leads to warming of the phonons. The transients are visible in the recording in all thermostats T_1, ..., T_14 (Extended Data Fig. 1d and the expanded scale in Extended Data Fig. 1e). In both panels, both effects add to give large transient spikes in the latter half of the sweep (time > 26 000 s), whereas in the earlier half (t < 26 000 s), they partially cancel to leave transients that display both cooling and warming spikes. In both halves, the signs are consistent with the sign of d[H]/dt. After these transients have decayed, the temperatures T_1, ..., T_14 are recorded and averaged (in the interval shaded blue in Extended Data Fig. 1e) to determine Δ, T to a resolution of ±3 μK. The high resolution notwithstanding, the intrinsic thermal Hall signal δ_T (from which κ_xx is derived) is contaminated by the longitudinal Δ, T picked up by a slight misalignment of the Hall contacts (equation (3) below). The second term, which is hugely dominant and hysteretic in α-RuCl₃ below 4 K, greatly complicates thermal Hall experiments. Next, we describe how we eliminate the hysteretic term.

Hysteretic background. Owing to a slight misalignment of the Hall contacts, the observed signal in the transverse (Hall) channel includes not only the intrinsic thermal Hall signal δ_T, but also a fraction of the longitudinal signal ∆, T; that is

\[ \Delta, T(H) = \delta_T(H) + \Delta, T(H) \]  

where α < 1 measures the contamination caused by Hall contact misalignment. The second term is problematical if it is hysteretic; it cannot be removed because ∆, T(H) ≠ ∆, T(−H). Instead, field-antisymmetrization leads to a ‘butterfly’ loop.

To properly eliminate all hysteretic contributions, we combine sweep-up and sweep-down curves. We let ∆, T(H) denote the curve recorded when H is swept from −14 T to +14 T, and recorded in the reversed direction (+14 T to −14 T) will be called ∆, T(−H).

Even when the longitudinal signal ∆, T is strongly hysteretic, it has the following mirror symmetry:

\[ \Delta, T(H) = \Delta, T(−H) \]  

That is, ∆, T(−H) is identical to the mirror image of ∆, T(H) (reflection across the H=0 axis, which changes H → −H).

By contrast, the intrinsic Hall signal δ_T(H) is antisymmetric under the mirror reflection; that is

\[ \delta_T(H) = -\delta_T(−H) \]  

Operationally, when we sweep H from −14 T to +14 T, we record the trace expressed in equation (2). In the reverse sweep (+14 T to −14 T), we record

\[ \Delta, T(H) = \delta_T(−H) + \Delta, T(−H) \]

where we used equations (3) and (4) in the second step.

Forming the field-antisymmetrized combination of equations (2) and (3), we achieve isolation of the intrinsic thermal Hall signal δ_T(H), or explicitly

\[ \frac{1}{2} [\Delta, T(H) - \Delta, T(−H)] = \delta_T(H) \]  

For the thermal Hall experiments with H || b (armchair direction), the above procedure leads to a δ_T that is zero (within our overall uncertainty of ±200 μK) at all T (from 0.9 K to 5.0 K). The data are shown in the bottom panel of Fig. 3b. For experiments with H || a (zigzag axis), δ_T is also zero if H lies below around 2.5 K (for |H| < 14 T). However, above 2.5 K, we observe the emergence of peaks that are H-antisymmetric, as shown in the top panel of Fig. 3b. This is the PTHE.

Phonon conduction and lattice disorder. The observed κ_xx is the sum

\[ \kappa_{xx} = \kappa_e^{ph} + \kappa_e^{c} \]  

where κ_e^{ph} is the phonon conductivity and κ_e^{c} the thermal conductivity associated with spin excitations (usually magnons in the magnetically ordered state).

In the QSL state, strong spin–phonon coupling makes it difficult to disentangle the two terms. However, at fields above 11.5 T in the polarized state, κ_e^{c} is strongly suppressed by the opening and growth of the magnon gap. Scattering of the phonons by spin excitations is also strongly suppressed. The thermal conductivity κ_xx is then strongly dominated by e^{ph}. The flat field profiles above 12 T at T = 1.5 K are consistent with both the dominance of e^{ph} and the absence of magnetic scattering. We have used its values at the plateau (which we call κ_e^{ph}(T)) to provide a quantitative measure of lattice disorder.

At 1 K, κ_e^{ph}(T) measured at 13 T equals 2.2 W K⁻¹ m⁻¹ and 0.7 W K⁻¹ m⁻¹ in Samples 3 and 1, respectively, which suggests a threefold lower degree of disorder in Sample 3. This is correlated with the sixfold increase in the peak magnitude of the amplitude Δe_x in Sample 3 versus Sample 1. As mentioned in the text, the plateau value κ_e^{ph}(T) at 1 K, which varies widely between crystals, is an excellent yardstick for measuring the crystalline quality of the lattice.

The absolute magnitudes of κ_xx are subject to uncertainties associated with contact geometry and measurements of crystal dimensions. To avoid these errors, we may also form the ratio

\[ R_{e_x} = \frac{e_x^{ph}}{\kappa_{min}} \]  

where κ_{min} is the minimum value of the field profile (typically just above R_{e_x} ≈ 7.5 T). At 1.0 K, R_{e_x} ≈ 17 in Sample 3, compared with around 6 in Sample 1. The estimated relative lattice disorder is similar.
Yet another way to measure lattice disorder is the ratio of the peak value (near 5 K) of the thermal conductivity $\kappa(T)$ in zero field $H$ to its minimum value (near 7.5 K). Reference 22 have shown that the ratio steadily decreases from 1.91 to around 1.4 as the amount of stacking faults is increased. The ratio measured in our Sample 3 is 2.04 (inset in Extended Data Fig. 4d). Compared with $R_{xx}$, this ratio is not sensitive at the high end (the clean limit) because it tends towards saturation. Either of the quantities ($R_{xx}$ or $k_{xx}/T$ at 1 K) may be used to determine crystalline quality. They do the job of the residual-resistance ratio commonly used to benchmark metals.

**Samples.** We report measurements from four samples. As listed in Table 1, Samples 1 and 3 were measured with an in-plane $H \parallel a$ (zigzag axis). Sample 2 was also in an in-plane $H$, but aligned with $b$ (armchair). The thickness and physical separation of the contact pads are listed in Table 1. Sample 4 did not exhibit resolvable oscillations when measured with $H$ at angle $\theta = 45^\circ$ to $a$. The reason may be that the lifetime of the excitations in Sample 4 is considerably shorter than in Samples 1, 2 and 3. As in conventional SdH experiments in semimetals, a short lifetime dampens the SdH amplitudes. As discussed in Methods, resolving the oscillations demands a high degree of lattice perfection. It is possible that Sample 4 is too disordered.

The discussion in the text was largely based on results from Sample 1. Extended Data Fig. 3a displays curves of $\kappa_{xx}$ versus $H$ measured in Sample 3 with $H \parallel a$ at temperatures from 0.51 K to 1.57 K. As may be seen, the curves in Sample 3 show oscillations with periods that are closely similar to those in Sample 1 (Fig. 2a and Extended Data Fig. 4c). Moreover, the profile of the amplitude $\Delta \kappa_{xx}$ extracted from these curves is closely similar to that in Sample 1 (Fig. 4a), except that the scale of the magnitude is roughly five times larger at the peak. As shown in Extended Data Fig. 3b, the peaks in the derivatives $d\kappa_{xx}/dB$ in Sample 3 are also more prominent than those in Sample 1.

**Determining the oscillation amplitude.** The problem of accurately determining the magnitudes of the oscillations is complicated by the strong non-monotonic variation of the curve ($\kappa_{xx}(H)$) (without oscillations) over the entire field interval 0 → 14 T. We adopt the following procedure. First, we find the smooth ‘background’ curve $\kappa_{bg}$ threading the midpoints between successive extrema of $d\kappa_{xx}/dB$. In Extended Data Fig. 2d, we display the background $\kappa_{bg}$ as the red curve for $T = 0.43$ K in Sample 1. Assuming that $\kappa_{bg}$ provides a close approximation to ($\kappa_{xx}(H)$), we obtain the oscillatory component of $\kappa_{xx}$ as the difference $\Delta \kappa \equiv \kappa_{xx} - \kappa_{bg}$. Its amplitude is called $\Delta \kappa_{xxf}$.  

**Oscillations in narrow-gap semiconductors.** Rapid quantum oscillations in a range of frequencies (with period 300 T to 10,000 T) have been reported for the narrow-gap correlated insulator SmB$_6$ (ref. 23). In models advanced for the oscillations, the chemical potential lies within a small hybridization gap $\Delta_{hyb}$ that forms at the crossing either between a wide band and a flat band, or at the intersection of overlapping electron and hole bands. Both models show that Landau quantization of the original bands can cause field modulation of $\Delta_{hyb}$, leading to oscillations in the magnetization $M$ that are periodic in $1/H$. However, for the oscillations to be observable (under the most favourable conditions), $\Delta_{hyb}$ must be comparable in size to the Landau-level spacing $\hbar\omega_L$ ($\omega_L$ is the cyclotron frequency). For applied fields $H$ of 10 T, this constraint requires $\Delta_{hyb}$ to be in the range 1–3 meV. For this mechanism to apply to $\alpha$-RuCl$_3$ (for which the charge gap $\Delta$ equals 1.9 eV (ref. 23)), we would need $H$ to be around 10,000 T, which is well beyond the maximum stationary field (50 T) currently available. Hence, this scenario is inapplicable to our experiment, which lies in the opposite limit (large charge gap).  

**Data availability** The data in the plots in this paper are available via the Harvard DataVerse at https://doi.org/10.7910/DVN/CWLZCI.

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**Correspondence and requests for materials** should be addressed to N.P.O.

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**Extended Data Fig. 1 | Experimental details.** Panel (a) shows a schematic of the mounted crystal and the applied in-plane field. The temperatures $T_a$, $T_b$, and $T_c$ are read off RX102A thermometers as shown. Panel (b) shows a photo of Sample 2 contacted with thick Au wires (100 μm in diameter) to thermometers and bonded to the bath by stycast. The heater and Delrin post are visible at the top right and left, respectively. Panel (c) shows the CI octahedra enclosing Ru ions (adapted from 20). The pink and blue planes are normal to the spin axes $S_z$ and $S_x$, respectively. The lower sketch highlights the plane containing the ‘Z’ Ru-Ru bond and the spin axis $S_z$. Panel (d) displays time-traces of the temperatures $T_a$, $T_b$, and $T_{bath}$ in a $\lambda_{yx}$ measurement with the bath temperature fixed at 280 mK. The field is gradually increased from -13.5 to 13.5 T over 14 hours by a step-wise change of (for example) 125 mT at each step. After each step-increase, transients caused by heating (or cooling) of the spins via the magneto-caloric effect combined with eddy-current heating of the brass bath are seen in all channels. The total transverse signal $\Delta T$ is the difference between the red and black curves (as expressed in Eq. (2), $\Delta T$ is the sum of $\delta_y$ and the ‘pick-up’ of the longitudinal $\Delta x$ caused by contact misalignment). Panel (e) shows an expanded view of 5 transient pulses bracketing $H=0$ (vertical dashed line). For $t < 26,000$ s, the 2 effects partially cancel whereas for $t > 26,000$ s, they add to give large transients. Readings are recorded within the blue-shaded interval after all transients have decayed. The average over the readings gives $\Delta_y$ to a resolution of ± 3 μK. Because of systematic errors, however, the total uncertainty in measuring $\delta_y$ is ± 200 μK.
Extended Data Fig. 2 | Data analysis details. Panel (a): Contributions of hysteretic effects to the signal detected at the Hall contacts (Δ_T) and at the longitudinal contacts (Δ_x). Below 4 K, it is critically important to identify and separate these contributions from the intrinsic PTHE signal δ_y using the procedure described in Methods. Panel (b) shows traces of the derivative curves dκ_{xx}/dB above 2 K in Sample 1. The physical reality of the oscillations is apparent in the raw data. Panel (c): The oscillatory component Δk (divided by the background κ_{bg}) measured in Sample 1 at selected T. Panel (d) illustrates the procedure for determining κ_{bg} (red curve) from the mid-points between derivative extrema of the measured curve of κ_{xx} (black curve). The difference of the 2 curves gives Δk.
Extended Data Fig. 3 | Oscillations in Sample 3. Panel (a): Oscillations in $\kappa_{xx}$ observed in Sample 3 at selected $T$ from 0.51 to 1.57 K. The derivative curves $d\kappa_{xx}/dB$ are shown in Panel (b). Panel (c) compares the AC susceptibility $\chi_{ac}$ reported in ref. 13 with the oscillations in Sample 1 (adapted from Fig. 1c of main text). As shown by the two red dashed lines, sharp peaks in $\chi_{ac}$ occur close (but not exactly at) the minima in $\kappa_{xx}$ near 6 T and 7.2 T. However, no peaks are observed in $\chi_{ac}$ away from these $H$ values where multiple oscillations occur in $\kappa_{xx}$. 
Extended Data Fig. 4 | Oscillations in tilted field. Curves of $\kappa_{xx}/T$ vs. $H$ measured in Sample 1 at tilt angle $\theta = 39^\circ$ (Panel a) and $\theta = 55^\circ$ (Panel b) with $T$ fixed at the six values indicated. Panel (c) shows the effect of tilting $H$ out of the plane in Sample 1 at angle $\theta$ (relative to a). Curves of $d\kappa_{xx}/dB$ vs. $1/H$ are displayed for $\theta = 0$ (top panel), $39^\circ$ (middle) and $55^\circ$ (bottom panel). Panel (d) displays the magnetization $M$ in Sample 1 (expressed in emu) vs. $T$ measured with $H = 0.1$ T in the direction $\parallel a$ (blue circles) and $\parallel b$ (red). The smooth increase in $M$ for $H \parallel a$ as $T$ decreases to 7 K is direct evidence for absence of stacking faults. The presence of stacking faults leads to a distinctive flat-plateau feature extending from 7 to 14 K. The inset plots the curve of $\kappa$ (in zero $H$) vs. $T$ in Sample 1. The ratio of the peak value (at 5 K) to the minimum at 7.5 K is 2.04.