
Supplementary information

Oscillations of the thermal conductivity in the spin-liquid state of α -RuCl₃

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Supplementary Information: Oscillations of the thermal conductivity in the spin-liquid state of α -RuCl₃

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(Dated: March 28, 2021)

Stacking faults and multiple transitions

We discuss the hypothesis that the oscillations, rather than being intrinsic, are an artefact of multiple transitions caused by stacking faults induced by strain during a field-sweep at low $T < 4$ K.

In early experiments on α -RuCl₃, notably Kubota *et al.* [26], multiple transitions were observed (between 7 and 14 K) in the curve of the magnetic susceptibility M/B vs. T measured in a weak $H \simeq 0.1$ T (M is the magnetization). In addition, Kubota *et al.* performed the orthogonal sweep in which the differential susceptibility $\chi_{ac} = dM/dH$ is measured vs. H with T fixed at 1.3 K, and also observed multiple features (five) in the field profile of χ_{ac} . In Ref. [26] Kubota *et al.* marked the 5 features with arrows (see Fig. 6b of Ref. [26]) and called them collectively “multiple transitions.”

Subsequently, Cao *et al.* [27] showed explicitly that the multiple transitions seen in curves of M/B vs. T (in weak H) by Kubota *et al.* were caused by stacking faults. When stacking faults appear in mishandled crystals, their curves of M/B vs. T are distinguished by a sharp break-in-slope at 14 K followed by flat-top plateau extending from 14 K to 7 K (Fig. 4a in Ref. [26]). By contrast, the flat-top plateau below 14 K is absent in pristine crystals – they display just one transition, that to the zig-zag state at $T_C \simeq 7$ K [16, 27]. The crystals used in our experiment were checked to be free of stacking faults. As shown in Extended Data Fig. S4d, the curve of $\chi = M/B$ vs. T in our Sample 1 displays just one transition (at 7 K). There is no evidence of the flat-top plateau.

Unfortunately, the “multiple transition” scenario that Kubota *et al.* extended to include features in the sweep of the differential susceptibility χ_{ac} vs. H at 1.3 K has led to considerable confusion. Kubota *et al.* identified the 5 features with arrows, to mark a series of field-induced transitions. It is easy to conflate the stacking-fault finding by Cao *et al.* with Kubota’s “5-arrow” χ_{ac} profile to arrive at the notion that field scans at low T produce

“multiple transitions” caused by stacking faults. To our knowledge, there is *no experimental evidence* that a field sweep at low T (< 4 K) on pristine crystals can create stacking faults that may lead to multiple transitions.

We summarize the χ_{ac} profile (for details see Lampen-Kelley[16]). Of the 5 features that Kubota marked with arrows (Fig. 6b in Ref. [26]), 4 are actually intrinsic features. First, the two most prominent peaks are intrinsic to the phase diagram rather than artefacts of stacking faults. We illustrate these peaks in Extended Data Fig. S3c showing χ_{ac} measured by the ORNL group in a pristine crystal [13]. The 2 sharp features at $B_{c1} = 6.1$ and $B_{c2} = 7.3$ T define the boundaries of the ZZ2 phase [17]. Likewise, the weak shoulder below 0.5 T (also intrinsic) reflects rotation/motion of domain walls in the zigzag phase. The only feature attributable to stacking faults is a weak shoulder at 9.5 T which is emphatically not a transition (note that it is absent in Extended Data Fig. S3c). Hence Kubota’s proposal of multiple transitions induced by sweeping H at low T is now known to be incorrect.

The confusion has led to the proposal that the oscillations in κ_{xx} may arise from field-induced stacking faults. In this section we review the evidence against this proposal. The stacking-fault mechanism is inherently stochastic and history dependent. Because the nucleation of stacking faults cannot be controlled experimentally, the parameters of oscillations (if caused by stack faults) should differ randomly between samples, and between separate runs on the same sample.

The strongest argument against the stochastic stacking-fault mechanism is the reproducibility of the periodic pattern across Samples 1, 2 and 3 in the index plots (main text Fig. 2a). The 5 derivative curves in main text Fig. 2b emphasize the reproducibility of both the periodicity in $1/H$ and phasing of the oscillations across samples as well as across different field-tilt angles. The left-most peaks (at $H = 11.5$ T) correspond to the step-

increase in κ_{xx} as the sample exits the QSL state. They are not part of the SdH series and are not plotted in Fig. 2a (of the main text).

The agreement of extrema in Samples 1 and 3, both measured with $\mathbf{H} \parallel \mathbf{a}$, is especially important. With current technology, it is impossible to control and tune the strain fields in each crystal to the desired precision. The challenge is compounded by the different mounting configurations in each experimental run. We used large sty-cast contact pads on Samples 1 and 2, but much smaller Ag epoxy pads on 3. A related challenge is the long time interval separating some of the runs on the same crystal (6-12 months). In spite of the long interval, the oscillations remain reproducible in period and phase. Hence the close agreement of the oscillations shown in Figs. 2a, b and c (main text) is strongly incompatible with a stochastic strain-induced mechanism.

For Sample 2 (green circles) measured with $\mathbf{H} \parallel \mathbf{b}$, the data also fall on the same low-field segment. Above 7 T, the slope is larger, reflecting an intrinsic anisotropy in S_f with respect to in-plane rotation of $\mathbf{H}$.

Tilting of $\mathbf{H}$ out of the a - b plane in measurements performed on the same sample (Sample 1) provides another stringent test of reproducibility in an independent “orthogonal” direction. Extended Data Figs. S4a and ??b display curves of κ_{xx} vs. H measured with $\mathbf{H}$ tilted in the a - c plane at angles θ (relative to $\mathbf{a}$) = 39° and 55° , respectively. Curves were taken at the 6 temperatures indicated. Although, at casual glance, no simple pattern is apparent across the 3 angles (including $\theta = 0$), we find that the same periods emerge if we replot the curves vs. the in-plane component H_a (as shown in main text Fig. 2c). In the index plot in Fig. 2a (main text), the integer increments Δn vs. $1/H_a$ fall on the same segmented curve (open circles and triangles).

To us, the two orthogonal tests of reproducibility in the 5 data sets in Figs. 2a, b and c (main text) provide compelling evidence supporting the intrinsic nature of the oscillations. It is unlikely that the stochastic process of nucleating stacking faults can mimic oscillations closely similar in frequency and phase in the 5 sets of data.

A very different argument against stacking faults is provided by the phonon conductivity as estimated by κ_{xx}^{plat}/T in the polarized state at 1 K (Methods). We showed that the amplitudes of the oscillations are highly sensitive to lattice disorder. The presence of stacking faults should strongly suppress both κ_{xx}^{plat}/T and the oscillations altogether.

Quantum oscillations from neutral Fermi Sea

We briefly sketch how quantum oscillations may arise from neutral fermions in the QSL state following Ref. [29]. In the widely adopted slave-particle approach, the electron operator $c_{i\alpha}$ is decomposed into a charged spinless boson φ_i and a neutral spin- $\frac{1}{2}$ fermion $\psi_{i\alpha}$, i.e.

$$c_{i,\alpha} = \varphi_i \psi_{i\alpha}, \quad (1)$$

where i is the site index and α denotes the spin polarization.

To describe the insulating QSL state, the charged bosons (called holons) are assumed to be gapped whereas the neutral fermions (spinons) remain itinerant. However, the fermions are strongly correlated. They interact via an internal gauge field $a_\mu = (a_0, \mathbf{a})$, which we motivate as follows.

By Eq. 1 the electron operator is invariant under the local gauge transformation

$$\varphi_i \rightarrow e^{i\theta_i} \varphi_i, \quad \psi_{i\alpha} \rightarrow \psi_{i\alpha} e^{-i\theta_i}. \quad (2)$$

This gauge freedom is implemented by an internal $U(1)$ gauge field $\mathbf{a}$ that couples to φ_i and $\psi_{i\alpha}$ with *opposite* gauge charges ± 1 .

In addition, the decomposition Eq. 1 demands the local constraint

$$\sum_\alpha \psi_{i\alpha}^\dagger \psi_{i\alpha} = 1, \quad (3)$$

which is imposed by the Lagrange multiplier a_0 . Combining a_0 with $\mathbf{a}$, we have the internal gauge field $a_\mu = (a_0, \mathbf{a})$. The internal field is distinguished from the physical gauge field $\mathbf{A}$ corresponding to the physical magnetic field $\mathbf{B} = \nabla \times \mathbf{A}$.

In the starting Lagrangian $\mathcal{L}$, we describe the fermions (of finite mass m_ψ) as minimally coupled to the internal gauge field $a_\mu = (a_0, \mathbf{a})$. The bosons are also minimally coupled to a_μ but with the opposite sign. In addition, the bosons couple minimally to the external gauge field $\mathbf{A}$. To allow a transition between the gapped phase ($\langle \varphi \rangle = 0$) and the condensed phase ($\langle \varphi \rangle \neq 0$) which is a metal, we expose the bosons to a field potential described by a Ginzburg-Landau expansion (terms involving $|\varphi|^2$ and $|\varphi|^4$).

Hence the Lagrangian has the form

$$\begin{aligned} \mathcal{L} = & \psi^\dagger \left(i\partial_t - a_0 - \frac{(\mathbf{p} - \mathbf{a})^2}{2m_\psi} \right) \psi + |(i\partial_\mu + a_\mu - A_\mu)\varphi|^2 \\ & - w|\varphi|^2 - \frac{1}{2}g|\varphi|^4 + \dots \end{aligned} \quad (4)$$

It bears repeating that the bosons couple to the *difference* field $A_\mu - a_\mu$, whereas the neutral fermions couple only to a_μ .

As low-lying charge excitations are absent, the bosons are gapped ($\langle \varphi \rangle = 0$), and we may integrate them out. The final result is a mean-field expression for the energy density u given by

$$u(b, B) = n\epsilon_\psi(n, b) + \frac{1}{2}\chi_\varphi(B - b)^2 + \frac{1}{2}\chi B^2. \quad (5)$$

where ϵ_ψ is the fermion energy, n the fermion density, and $\mathbf{b} = \nabla \times \mathbf{a}$ is the internal field. As noted, the energy of the gapped bosons increases with applied field $\mathbf{B}$, but in the form $\frac{1}{2}\chi_\varphi(B - b)^2$ (with χ_ϕ the susceptibility). By way of comparison, other (spectator) filled bands indifferent to

the decomposition of $c_{i\alpha}$ contribute as $\frac{1}{2}\chi B^2$ (the third term in Eq. 5).

In analogy with 2D electrons (with quadratic dispersion), quantization in the internal field $\mathbf{b}$ leads to Landau levels (LLs) of degeneracy $g = b/2\pi$ and uniform spacing ω_c (we set $\hbar$ and e to 1). Writing the filling factor $\nu = n/g$ as $\nu = \nu_i + \nu_f$ where ν_i and ν_f are its integer and fractional part, respectively, we readily find the total energy of the fermions to be

$$E_\psi(n, b) = \frac{1}{2}g\omega_c\nu_i^2 + g(\nu_i + \frac{1}{2})\omega_c\nu_f. \quad (6)$$

Here the first term is the energy of ν_i filled LLs and the second is the energy of the highest, partially-filled level.

We then obtain for $\epsilon_\psi(n, b) = E_\psi(n, b)/n$

$$\epsilon_\psi(n, b) = \frac{\pi n}{m_\psi} \left(1 + \frac{\nu_f(1 - \nu_f)}{\nu^2} \right). \quad (7)$$

As a function of b , $\epsilon_\psi(n, b)$ displays a scalloped profile with deep, cuspy minima occurring whenever all occupied LLs are fully filled ($\nu = \nu_i$).

At a fixed $\mathbf{B}$, we minimize $u(b, B)$ by allowing b to vary. From Eq. 5, b selects the nearest value $b_p = 2\pi n/p$ at which exactly p levels are filled ($p \in \mathbb{Z}$). The cuspy profile of the minima in ϵ_ψ ensures that b remains pinned to b_p in the face of slight field increments ΔB . However, for large enough ΔB , the term $\frac{1}{2}\chi_\varphi(B - b)^2$ tips the global minimum to the next cusp, causing b to jump to b_{p-1} . Hence, sweeping B continuously leads to a hysteretic, staircase profile for b , which produces sharp oscillations in observable quantities. The staircase behavior of the internal field was first found by Motrunich [28].

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