Magnons, fractional excitations, and field-induced transitions in $\alpha$-RuCl$_3$

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(Dated: March 15, 2019)

An external magnetic field can induce a transition in $\alpha$-RuCl$_3$ to a disordered state that is possibly related to the Kitaev quantum spin liquid. Here we present new field dependent inelastic neutron scattering data showing three distinct regimes of magnetic response. In the low field ordered state the response shows magnon peaks; the intermediate field quantum disordered state shows only continuum scattering, and a high field regime shows sharp magnon peaks at the lower bound of a strong continuum. Measurable dispersion of magnon modes along the (0,0,$L$) direction implies non-negligible inter-plane interactions. Magnetocaloric effect measurements yield thermodynamic evidence for three field driven phase transitions in the measured range. Combining these with other data we construct a $T − B$ phase diagram. The results constrain the range where one might expect to observe QSL behavior in $\alpha$-RuCl$_3$.

The fractional Majorana fermion excitations of a Kitaev quantum spin liquid (QSL) [1] have been proposed as a route to topologically protected qubits [2]. The suggestion that this physics is exhibited in certain honeycomb magnets with $J_{\text{eff}} = 1/2$ ground states [3] has led to an enormous amount of research on iridate materials [4–8], and more recently, an intense interest in $\alpha$-RuCl$_3$ [9–12]. In the absence of an external magnetic field $\alpha$-RuCl$_3$ orders below $T_N \approx 7$ K in a 3D stacked antiferromagnetic zigzag ground state [13, 14], however an in-plane field perpendicular to a Ru-Ru bond of $B_c \approx 7.5$ T results in a quantum disordered phase proposed to be a QSL [15]. The magnetic excitations of $\alpha$-RuCl$_3$ have been studied using a wide range of techniques including Raman scattering [16], ESR, THz, and microwave spectroscopy [17–21], and inelastic neutron scattering (INS) [22–25]. Spectroscopic evidence for fractionalization is seen in the form of unusual continuum scattering around the 2D $\Gamma$ point [16, 23, 25]. Above $B_c$ the spin waves associated with zigzag order disappear and the continuum is enhanced [26]. The intriguing possibility that the continuum excitations are related to chiral Majorana edge modes was raised by the observation of a quantized thermal Hall plateau with an onset at $B_u$ [27]. The disappearance of the plateau at a higher field, $B_u$, was attributed to a possible topological transition to a field polarized phase. Various theoretical proposals have been advanced to explain these observations [28–33], nevertheless, many important experimental issues remain unresolved. Here we report new field dependent INS and magnetocaloric effect (MCE) measurements showing that there is a transition at $B_u$ accompanied by a qualitative change in the magnetic excitation spectrum. Important aspects of the $T − B$ phase diagram are clarified.

Neutron scattering data was collected on a 2 g single crystal grown using vapor-transport techniques as described elsewhere [23]. The INS measurements utilized the FLEXX triple-axis spectrometer at the Helmholtz-Zentrum Berlin [34]. The spectrometer was configured with open collimation, double focusing PG monochromator, and horizontally focusing PG analyzer with a fixed final energy of 5 meV, yielding an energy resolution of 0.34 meV FWHM at the elastic position. The crystal was mounted initially with the $(H,0,L)$ scattering plane horizontal (using trigonal notation). In an applied field there was a slight 6° irreversible rotation about the $L$ axis. Given the known dependence of $B_c$ on the precise in-plane field direction [35] this has no significant effect on the results shown below.

Figure 1 shows constant wavevector scans for two values of $(0,0,L)$ corresponding to the 2D $\Gamma$-point. At energies below 1 meV incoherent elastic scattering dominates the signal. In the accessible inelastic range the data shows three distinct field regimes corresponding to (I) $B < B_c$, (II) $B_c < B < B_u$, and (III) $B > B_u$. In region I, including zero field (figure 1(a)), well-defined magnon peaks are visible arising from the zigzag order. The peak energies depend on $L$ as expected for 3D zigzag magnetic order. Above $B_c$ in region II the magnon peaks disappear as reported previously [26]. In region III, above $B_u$, a sharp gapped magnon mode reappears. Additional modes may be present at energies above those measurable in the current experiment. The continuum scattering reported previously is present in regions I and III at energies above the magnon peaks as well as in region II. The statistical significance of the scattering level can
FIG. 1. Field dependence of the inelastic neutron scattering at the 2D Γ-point for two values of the out-of-plane wavevector transfer. Data obtained at 1.5 K on a 2 g single crystal of α-RuCl₃ using the FLEXX triple-axis spectrometer. (a) Zero-field data. A field of (b) 8 T and (c) 13.5 T was applied in the honeycomb plane perpendicular to a Ru-Ru bond (see inset of (b)). The solid lines are fits and the dashed lines show the model free background for \( L = 3.3 \) as described in the text. Error bars represent one standard deviation assuming Poisson statistics.

TABLE I. Average counts per 3 minutes. The uncertainties represent one standard deviation of the last digit assuming Poisson statistics.

| Range [meV] | Counts |
|-------------|--------|
| \( L = 3.3 \) | \( L = 4.3 \) |
| Background  | 1 – 3.5 | 1 – 2 | 5.0(5) | 4.2(7) |
| 0 T         | 3 – 6  | 2.5 – 6 | 7.4(5) | 6.4(5) |
| 8 T         | 1 – 5  | 1 – 5  | 9.7(8) | 13(1)  |
| 13.5 T      | 4.5 – 6 | 4 – 6  | 15(1)  | 8.3(7)  |

be assessed in a model free fashion using the average count levels over selected ranges of energy transfer. The effective background level is determined from the low energy scattering measured at 13.5 T as this represents the cleanest signal. The levels for the scans depicted in figure 1 are shown in table I. Note that all time-of-flight (ToF) measurements reported previously [23, 25, 26] for the 2D Γ points in regions I and II represent integrations over a large range of \( L \). The ToF procedure captures more of the continuum scattering, and the measurements at \( B = 8 \) T were interpreted as showing a gap [26]. In the present experiment the statistics measured at single wavevectors are insufficient to confirm the value of any possible gap in region II. The continuum intensity, at least in regions I and II, does not show any significant dependence on \( L \), consistent with the 2D nature of the continuum scattering reported previously [23].

To extract values of the magnon peak positions the data in regions I and III was fitted to an empirical function consisting of the sum of: a Lorentzian representing the peak, a constant background, and a rounded hyperbolic tangent function with the origin at the peak position to model the continuum scattering on the high energy side. The data in region II was fitted to a peak centered at zero energy plus a constant background. The fitted peak positions are used to plot the dispersion of the magnons along \( (0, 0, L) \) as shown in figure 2(a) for zero field (open circles) and \( B = 13.5 \) T (closed circles). The spectrum at 13.5 T is shifted upwards, presumably by the Zeeman energy, and has a larger bandwidth than that in zero field, but the periodicity of the dispersion is unchanged. The dispersion is clear evidence of out-of-plane magnetic interactions, consistent with the 3D stacked zigzag order in zero field. The excitations in a fully polarized ferromagnetic state are expected to be sharp magnons with the dispersion determined by the underlying interactions [36]. The presence of the high energy continuum at \( B = 13.5 \) T is consistent with partial ferromagnetic polarization.

Empirically, the dispersion of the mode along \( L \) can be modeled by the simple function

\[
E = A + B \cos\left(\frac{2\pi L}{3}\right)
\]

(1)

where \( A \) represents an average magnon energy along \( (0, 0, L) \) including the Zeeman term, and the bandwidth \( B \) reflects interlayer coupling. From considerations of the 3D magnetic order the effective interlayer coupling is expected to be antiferromagnetic [13, 14]. A spin wave calculation can, in principle, lead to the observed periodicity along \( (0, 0, L) \) but the full 3D Hamiltonian for α-RuCl₃ remains unresolved and there is insufficient information to present a meaningful 3D model here. The fact that the in-plane interactions in particular are expected to be heavily frustrated makes it difficult to estimate the relative magnitude of the in-plane and inter-plane couplings. A rough upper bound can be obtained by comparing the in-plane bandwidth of the lowest magnon mode with that of the dispersion along \( L \). For example, in a simple model with nearest-neighbor Heisenberg couplings, \( J \), the leading order term of the out-of-plane dispersion is propor-
tional to $\sqrt{J_{\perp}}$ [37] where $J_{\perp}$ is the out-of-plane coupling. The zero field out-of-plane magnon bandwidth for $\alpha$-RuCl$_3$ is 0.6 meV. Experimental estimates of the in-plane bandwidth of the lowest spin wave mode vary from 1.3 meV [26] to 5.5 meV [24], corresponding in a simple model to ratios of $J_{\perp}/J = 1 - 20\%$. Frustration is expected to lead to a narrower in-plane bandwidth, and accounting for this we expect the inter-plane coupling in $\alpha$-RuCl$_3$ to be at most a few percent of the in-plane coupling. An accurate determination of the full Hamiltonian requires at minimum a measurement of the in-plane magnon dispersion in the high field limit, a significant undertaking beyond the scope of this Letter.

It is interesting to note that the point $(0,0,0)$ sampled by ESR, THz spectroscopy, and other optical techniques is a local maximum of the dispersion along $L$ and the excitations there do not represent an overall energy gap. The $L$ dispersion might explain the difference in the gap energy at 13.5 T inferred by thermal conductivity (2.8 meV [38]) vs. that seen in the strongest ESR (3.9 meV [17]) and THz (4.5 meV [18]) absorption lines since the former is presumably related to the global excitation energy minimum and agrees with the INS results at $(0,0,4.5)$.

The wavevectors ($L = 3.3$ and $L = 4.3$) plotted in figure 1 are near the maximum ($L=3$) and minimum ($L = 4.5$) of the $L$-dispersion. The peak positions as a function of magnetic field are plotted in figure 2(b). Inelastic peaks below 0.7 meV (hatched region) were undetectable. In region I the spin wave energies diminish with increasing field, disappearing completely at $B_c$, consistent with the ToF measurements [26]. No peaks could be discerned in region II and the scattering is dominated by the continuum, consistent with expectations for a QSL [39]. At 9 T and above (region III) sharp peaks reappear with a strong continuum on the high energy side. The sudden change in the spectrum is consistent with a value for $B_u$ between 8.5 T and 9 T. The peak energy increases roughly linearly with field in region III, and in the overlap region agrees with the gap energy extracted from thermal conductivity measurements [38]. As seen in figure 2(b), in region III the bandwidth increases with increasing field. The narrower bandwidth just above $B_u$ is possible an effect of quantum fluctuations and implies that as the QSL is approached the layers decouple, underscoring the 2D nature of the physics in region II.

The $T-B$ phase diagram was investigated further by MCE measurements spanning the region $0.8 - 7$ K and $5 - 11$ T. These used a 6 mg single crystal, with a sharp zero-field phase transition near 7 K as determined by heat capacity measurements. The magnetic field applied in the $ab$ plane was swept both upward and downward at a rate of 0.3 T/min. By comparison with the known angular dependence of the transition temperature at $B_c$ [35], the direction of the field was identified as 10° off from the direction perpendicular to a Ru-Ru bond. The field sweep produces a temperature difference $\Delta T$ between the sample and the thermal reservoir that depends on $\frac{\partial M}{\partial T}$ [40] where $M$ is the magnetization. Reversing the field-sweep direction reverses the sign of the temperature difference and phase transitions are identified by anomalies in the difference between the curves.

The main panel of figure 3(a) shows MCE difference data at 2.3 K, revealing three anomalies in the measured field range. Anomaly 1 (6.5 T at 2.3 K) appears as a weak maximum at a transition seen previously in AC susceptibility [35] corresponding to a change between two zigzag ordered states with different interlayer stacking [41]. The sharp maximum at anomaly 2 (7.35 T at 2.3 K) is the transition at $B_c$. These two transitions are also evident in the intensity of magnetic Bragg peaks seen
in neutron diffraction (ND, see arrows in the inset of figure 3(b)) as the intensity changes slope at 5.9 T and disappears at 7.3 T. Anomaly 3 appears as a kink in the MCE difference and defines $B_u$.

A $T - B$ phase diagram assembled from the available data is shown in Figure 3(b) utilizing the new MCE and previously published susceptibility data [26]. Additional points are obtained from the INS, the zero-field heat capacity, and ND. The phase diagram shows four phases: two ordered states labeled zz1 and zz2 comprising region I, the potential QSL state (II), and a state that is at least partially field polarized (III). The transition points derived from the different measurements are mutually consistent except for a minor detail: the zz2 phase at 2.2 K seen in ND has a lower onset than that inferred from MCE. This is likely because the ND was taken with the field precisely perpendicular to a Ru-Ru bond direction, and the width of the zz2 phase is known to depend on the in-plane field direction [35]. Most importantly a transition at $B_u$ is evident in both the INS and MCE data. This agrees with the conjecture of a topological transition inferred from thermal Hall measurements [27], and implies the addition of a high field state to existing phase diagrams for α-RuCl$_3$ [26, 42–45]. It is likely that there are additional transitions at fields above $B_u$, eventually leading to a fully polarized state. This would be indicated by a crossing of the two MCE curves shown in the inset of figure 3(a) since $(\frac{\partial M}{\partial T})_H$ will be negative in the fully polarized state.

The difference between $B_c$ and $B_u$ evidently decreases with temperature between 4 K and 1 K. Whether or not the transition lines converge at $T = 0$ remains unresolved experimentally. If in fact the lines converge at $T = 0$ the fractional excitations seen at higher temperatures may be a signature of a quantum critical point (QCP) rather than a finite field region with a $T = 0$ QSL ground state. Exact diagonalization calculations for finite systems using a Hamiltonian proposed to describe α-RuCl$_3$ [46] have been interpreted as implying that the quantum disordered state is smoothly connected to the field polarized state. This might be consistent with such a QCP, and lower temperature measurements are called for to help resolve this issue.

Many theoretical works have pointed to off-diagonal exchange (so called $\Gamma$ and $\Gamma'$ terms) in the Hamiltonian playing an important role in the possible field-induced (or field-revealed) QSL behavior of α-RuCl$_3$, see e.g. [15, 31–33, 47]. It has been argued recently [33] that in the presence of an external field such a term leads to a mass gap in the Majorana fermion spectrum, a necessary condition for observing quantization in the thermal Hall effect. Most of these calculations consider a field applied in the $<111>$ direction in spin space, corresponding to a field perpendicular to the honeycomb plane. New density matrix renormalization group calculations [32] exploring the effect of field direction show that the region of QSL behavior between the zigzag ordered and fully polarized phase is largest for an external field in this direction, but even with the field applied solely in-plane a narrow intermediate QSL region is possible. The field required to destroy the zigzag order quickly becomes very large when rotated out of the honeycomb plane.

In summary, the measurements reported here show
clear evidence for field induced transitions in $\alpha$-RuCl$_3$, including that from a quantum disordered phase to a partially polarized phase in agreement with the topological transition inferred from thermal Hall effect measurements [27]. INS measurements of magnetic excitations at wavevectors corresponding to the 2D $\Gamma$ point show markedly different dynamic behavior in three distinct regions of field. Looking to the future, an unambiguous determination of the effective spin Hamiltonian for $\alpha$-RuCl$_3$ will need to include inter-plane interactions. The present results for the $T-B$ phase diagram provide important constraints for such a theory.

We thank Mark Meisel and Steven Kivelson for valuable discussions. P.L.K. and D.M. were supported by the Gordon and Betty Moore Foundation. We acknowledge support from the U.S. Department of Energy (U.S.-DOE), Office of Science - Basic Energy Sciences (BES), Materials Sciences and Engineering Division. Work at the Oak Ridge National Laboratory Spallation Neutron Source was supported by U.S.-DOE, Office of Science - BES, Scientific User Facilities Division. We thank the Helmholtz-Zentrum Berlin for the allocation of neutron beamtime. A portion of this work was performed at the National High Magnetic Field Laboratory, which is supported by the National Science Foundation Cooperative Agreement No. DMR-1644779 and the State of Florida.

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