Field-induced quantum criticality in the Kitaev system α-RuCl3

DOI:
10.1103/PhysRevB.96.041405

Document status and date:
Published: 13/07/2017

Document Version:
Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

PHYSICAL REVIEW B **96**, 041405(R) (2017)

Field-induced quantum criticality in the Kitaev system α-$RuCl_3$

A. U. B. Wolter,[1,*] L. T. Corredor,[1] L. Janssen,[2] K. Nenkov,[1] S. Schönecker,[3] S.-H. Do,[4] K.-Y. Choi,[4] R. Albrecht,[5] J. Hunger,[5] T. Doert,[5] M. Vojta,[2] and B. Büchner[1,6]

[1]*Leibniz-Institut für Festkörper- und Werkstoffforschung (IFW) Dresden, 01171 Dresden, Germany*
[2]*Institut für Theoretische Physik, Technische Universität Dresden, 01062 Dresden, Germany*
[3]*Department of Materials Science and Engineering, KTH - Royal Institute of Technology, Stockholm 10044, Sweden*
[4]*Department of Physics, Chung-Ang University, Seoul 156-756, Republic of Korea*
[5]*Fachrichtung Chemie und Lebensmittelchemie, Technische Universität Dresden, 01062 Dresden, Germany*
[6]*Institut für Festkörperphysik, Technische Universität Dresden, 01062 Dresden, Germany*

(Received 11 April 2017; published 13 July 2017)

α-$RuCl_3$ has attracted enormous attention since it has been proposed as a prime candidate to study fractionalized magnetic excitations akin to Kitaev's honeycomb-lattice spin liquid. We have performed a detailed specific-heat investigation at temperatures down to 0.4 K in applied magnetic fields up to 9 T for fields parallel to the *ab* plane. We find a suppression of the zero-field antiferromagnetic order, together with an increase of the low-temperature specific heat, with increasing field up to $\mu_0 H_c \approx 6.9$ T. Above H_c, the magnetic contribution to the low-temperature specific heat is strongly suppressed, implying the opening of a spin-excitation gap. Our data point toward a field-induced quantum critical point at H_c; this is supported by universal scaling behavior near H_c. Remarkably, the data also reveal the existence of a small characteristic energy scale well below 1 meV, above which the excitation spectrum changes qualitatively. We relate the data to theoretical calculations based on a J_1-K_1-Γ_1-J_3 honeycomb model.

DOI: 10.1103/PhysRevB.96.041405

α-$RuCl_3$ is a $J_{\rm eff} = \frac{1}{2}$ Mott insulator with a layered structure of edge-sharing $RuCl_6$ octahedra arranged in a honeycomb lattice [1–8]. It has been suggested [9,10] that strongly spin-orbit-coupled Mott insulators with that lattice geometry realize bond-dependent magnetic "compass" interactions [11], which, if dominant, would lead to a quantum spin-liquid (QSL) ground state as discussed by Kitaev [12]. This exotic spin-disordered state displays an emergent Z_2 gauge field and fractionalized Majorana-fermion excitations relevant for topological quantum computation [12–15].

While α-$RuCl_3$ displays magnetic long-range order (LRO) of a so-called zigzag type, it has been proposed to be proximate to the Kitaev spin liquid based on its small ordering temperature and its unusual magnetic excitation spectrum [16–18]. The magnetic interactions between the Ru^{3+} magnetic moments are believed to be described by a variant of the Heisenberg-Kitaev model [10]: Electronic-structure calculations indicate that the Kitaev interaction in α-$RuCl_3$ is ferromagnetic and indeed defines the largest exchange energy scale [19,20]. However, the debate about the spin model most appropriate for α-$RuCl_3$—likely to include Heisenberg and off-diagonal exchange interactions, possibly also beyond nearest neighbors—has not been settled [19–28].

The physics of α-$RuCl_3$ in an external magnetic field promises to be particulary interesting: It has been reported that magnetic ordering disappears for 8 and 10 T for fields applied in the *ab* and c' (30° off the *c* axis) directions [29–31], respectively, while for fields perpendicular to the honeycomb plane, T_N does not shift up to 14 T [4]. Interestingly, NMR measurements with fields in the specific direction $H \parallel c'$ performed down to 4 K have indicated the formation of a sizable spin gap at high fields [30]. Additionally, numerical exact-diagonalization studies of an extended Heisenberg-Kitaev model found hints for a transition from zigzag magnetic ordering to a spin-liquid state when applying a magnetic field [19].

In this Rapid Communication, we report a careful heat-capacity study of α-$RuCl_3$ down to low temperature T of 0.4 K in in-plane fields up to 9 T. We confirm the field-induced suppression of LRO at a critical field of $\mu_0 H_c \approx 6.9$ T and provide a detailed account of the field evolution of the spin gap: This is small below H_c, closes at H_c, and progressively grows above H_c. The specific-heat data display universal scaling consistent with the existence of a quantum critical point (QCP) at H_c. The scaling analysis yields critical exponents $d/z = 2.1 \pm 0.1$ and $\nu z = 0.7 \pm 0.1$, where d is the space dimension and ν and z are the correlation-length and dynamic critical exponents, respectively. Based on explicit calculations for a J_1-K_1-Γ_1-J_3 spin model, we argue that the specific-heat behavior near H_c implies a mode softening at H_c that accompanies the disappearance of magnetic order. The observed violations of scaling for $T \geqslant 3$ K indicate the presence of an intrinsic sub-meV energy scale near the QCP which we interpret as a signature of Kitaev physics.

Experiment. High-quality single crystals of α-$RuCl_3$ were grown by a vacuum sublimation method. A commercial $RuCl_3$ powder (Alfa-Aesar) was thoroughly ground, and dehydrated in a quartz ampoule at 250 °C for 2 days. The ampoule was sealed in vacuum and placed in a temperature-gradient furnace. The temperature of the $RuCl_3$ powder was set at 1080 °C. After 5 h the furnace was cooled to 600 °C at a rate of −2 °C/h. The magnetic properties of the crystal were checked through measurements as a function of T and H using a vibrating sample magnetometer (Quantum Design) with superconducting quantum interference device detection (SQUID-VSM) (see the Supplemental Material [32] for the

*Corresponding author: a.wolter@ifw-dresden.de

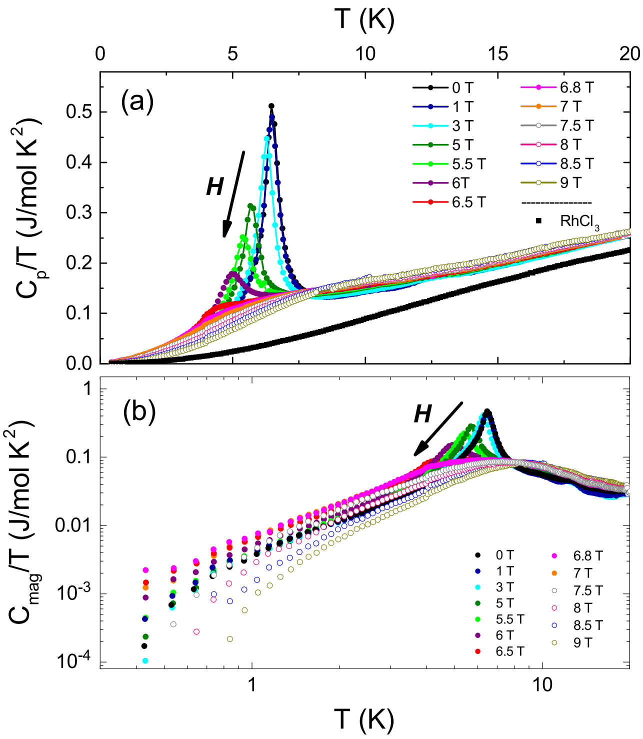

FIG. 1. (a) Temperature dependence of the specific heat, plotted as C_p/T, of α-$RuCl_3$ for different magnetic fields up to 9 T ∥ *ab*. (b) As before, but showing the magnetic contribution to the specific heat after phonon subtraction on a log-log scale; for details, see text.

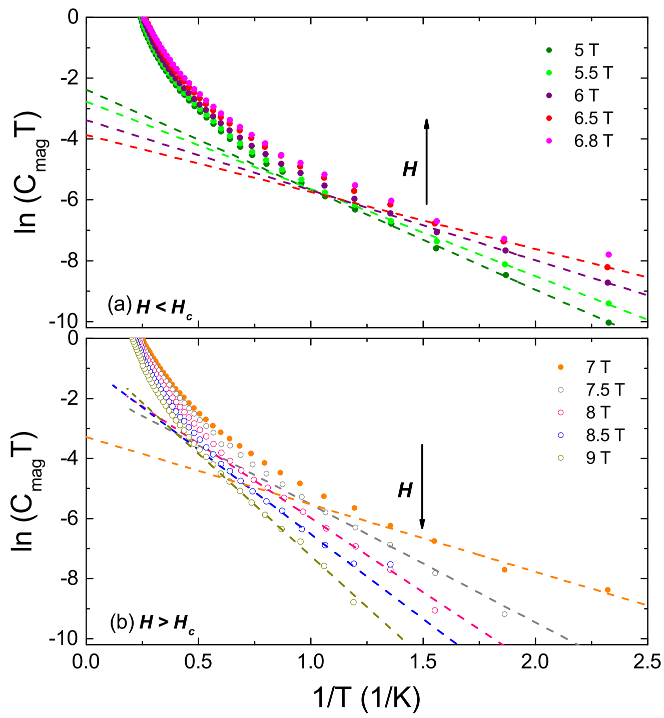

FIG. 2. Exponential fit of $C_{\text{mag}}T$ in order to extract the excitation gap for magnetic fields (a) $5\text{ T} \leqslant \mu_0 H \leqslant 6.8\text{ T}$ and (b) $7\text{ T} \leqslant \mu_0 H \leqslant 9\text{ T}$. The data at 6.8 T cannot be meaningfully fit by an exponential, i.e., the gap is too small.

magnetic characterization). Specific-heat measurements were performed on a single crystal ($m \sim 7$ mg) between 0.4 and 20 K using a heat-pulse relaxation method in a physical properties measurement system (PPMS, Quantum Design), in magnetic fields up to 9 T parallel to the *ab* plane. In order to obtain the specific heat of α-$RuCl_3$, the temperature- and field-dependent addenda were subtracted from the measured specific-heat values in the sample measurements.

Results. The low-T specific heat C_p/T as a function of temperature in different applied fields is shown in Fig. 1(a). The zero-field curve reveals the good quality of the sample, with a single magnetic transition at $T_N = 6.5$ K determined from the peak position. By applying a magnetic field, the peak becomes broader and the transition temperature is gradually suppressed. Finally, no thermal phase transition is detected for fields higher than 6.9 T, i.e., magnetic LRO disappears.

In order to extract the magnetic contribution to the low-T specific heat, the data were analyzed by subtracting the lattice contribution from the experimental $C_p(T)$ data by measuring the nonmagnetic structural analog compound $RhCl_3$ in pressed polycrystalline form. The difference of mass and volume between the Rh and Ru compounds was accounted for by scaling the experimental specific heat curve by the Lindemann factor [33], which was found to be 0.98. With the aim of ruling out possible errors due to nonperfect sample coupling during the measurements, the phononic contribution was also calculated for $RhCl_3$ by density-functional theory (see the Supplemental Material [32]). This approach confirmed that the phonon subtraction based on the experimental data is consistent with the theoretical calculations for $T \geqslant 1$ K.

The temperature dependence of the calculated magnetic contribution to the specific heat is shown in Fig. 1(b). In the lowest-T region, $T \leqslant 3$ K, an increase of C_{mag}/T with the applied field could be observed up to $\mu_0 H = 6.8$ T. Increasing the field even further, the opposite behavior is revealed: The magnetic contribution starts to decrease with field up to the highest field of 9 T. Hence, low-T entropy accumulates around 6.8–7 T. Remarkably, around 6.9 T, the magnetic specific heat displays an approximate power-law behavior between 0.4 and 2.5 K, with $C_{\text{mag}} \propto T^x$ with $x \approx 2.5$. Together, these observations imply the existence of a field-induced QCP [34,35] at $\mu_0 H_c \approx 6.9$ T.

Excitation gap. The lowest-temperature data away from the QCP, with a gradual suppression of $C_{\text{mag}}(T)$, indicate the opening of a magnetic excitation gap [Fig. 1(b)]. The simplest model of a bosonic mode with gap Δ and parabolic dispersion in $d = 2$ predicts that $C_{\text{mag}} \propto \exp[-\Delta/(k_B T)]/T$ (see the Supplemental Material [32]). According to this, the experimental $C_{\text{mag}}T$ data were fitted to a pure exponential behavior in order to extract the energy gap. The fits are shown in Fig. 2 and the results in Fig. 4.

Two key observations are apparent: First, the data below about 1.5 K indeed show an exponential suppression of C_{mag}, and the corresponding gap is minimal near the putative QCP at $\mu_0 H_c \approx 6.9$ T. It varies monotonically on both sides of the QCP, consistent with theoretical expectations [34,35]. (Note that a symmetry-broken phase below H_c should also display a gap, as no Goldstone modes are expected due to the presence of strong spin-orbit coupling.) Second, the data above ~1.5 K do *not* follow an exponential behavior (at least not in the field

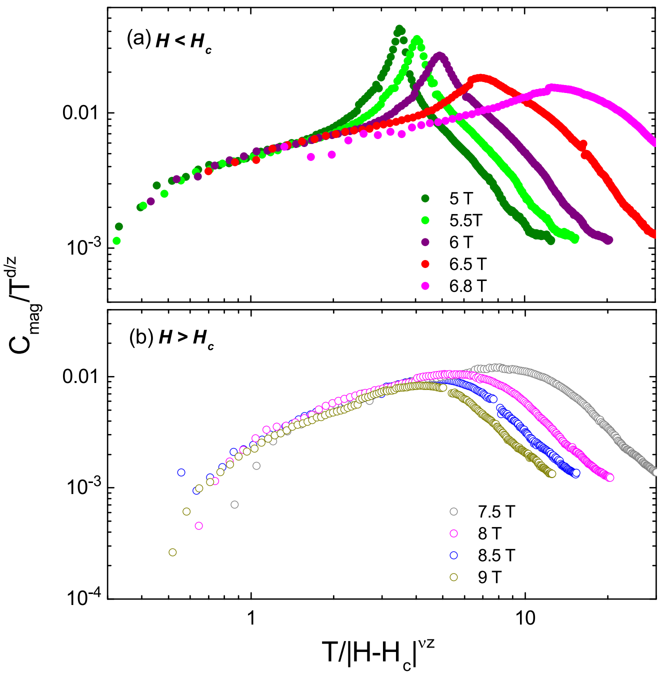

FIG. 3. Scaling plot of $C_{\mathrm{mag}}(T,H)$, showing $C_{\mathrm{mag}}/T^{d/z}$ vs $T/(H-H_{\mathrm{c}})^{\nu z}$, here for $\mu_0 H_{\mathrm{c}} = 7$ T, $d/z = 2.1$, and $\nu z = 0.7$. The two panels show fields (a) slightly below and (b) slightly above H_{c}. The universal piece in the upper (lower) panel corresponds to the scaling function f_- (f_+) in Eq. (1).

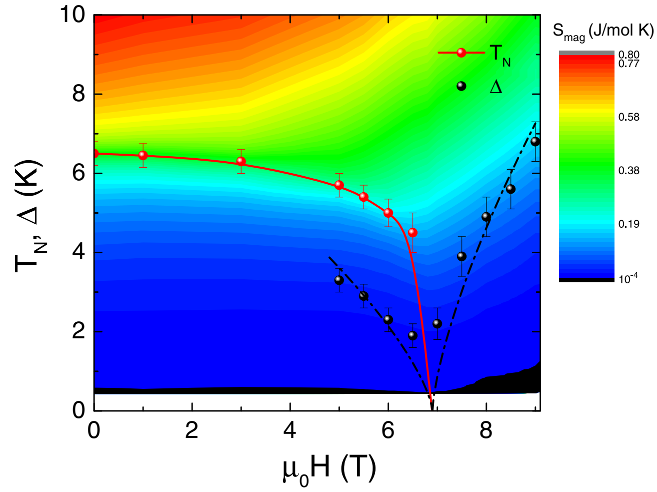

FIG. 4. T-H phase diagram for α-$RuCl_3$ magnetic ordering temperature and energy gap as a function of the applied magnetic field $\parallel ab$. The dashed line corresponds to the fit of the gap function to $\Delta \propto |H - H_{\mathrm{c}}|^{0.7}$. Additionally, the magnetic entropy $S_{\mathrm{mag}}(T,H)$ is shown in a color scale.

range studied here); in fact, C_{mag}/T between 1.5 and 5 K appears more consistent with a power law [Fig. 1(b)]. This indicates that the density of states of magnetic excitations changes its character at a small energy scale of a few tenths of a meV [32].

Scaling analysis. In order to further substantiate the QCP hypothesis, we have performed a scaling analysis of $C_{\mathrm{mag}}(T,H)$. Provided that hyperscaling holds, the critical contribution to the specific heat is expected [34,35] to scale as

$$C = T^{d/z} f_{\pm}(T/|H - H_{\mathrm{c}}|^{\nu z}), \qquad (1)$$

where $f_{\pm}$ are universal functions describing the scaling for $H > H_{\mathrm{c}}$ and $H < H_{\mathrm{c}}$, respectively, and the argument $T/|H - H_{\mathrm{c}}|^{\nu z}$ is made dimensionless by using suitable units. Plotting the specific heat as $C/T^{d/z}$ as a function of $T/|H - H_{\mathrm{c}}|^{\nu z}$, separately for $H \lesssim H_{\mathrm{c}}$ and $H \gtrsim H_{\mathrm{c}}$, we find an approximate data collapse for $d/z = 2.1 \pm 0.1$, $\nu z = 0.7 \pm 0.1$, and $\mu_0 H_{\mathrm{c}} = 6.9 \pm 0.1$ T; see Fig. 3 for an example. (Note that the data cannot be collapsed with $d/z = 2.5$.) For comparison, the Supplemental Fig. S5(c) shows the scaling collapse of specific-heat data obtained from a spin-wave-based model calculation for a field-driven QCP in a J_1-K_1-Γ_1-J_3 model; for details, see Ref. [32]. The agreement reinforces the notion of a field-induced QCP in α-$RuCl_3$.

It is instructive to analyze deviations from scaling in Fig. 3: (i) None of the data sets realizes the critical power law $C_{\mathrm{mag}} \propto T^{d/z}$, indicating that the critical point has not been reached precisely. The most likely reason is sample inhomogeneities, e.g., caused by crystallographic domains with different in-plane orientation. These would lead to a distribution of $|H - H_{\mathrm{c}}|$ values due to anisotropic g factors and hence to a smearing of the QCP. (ii) Only data below ~ 3 K follow the approximate scaling; this is particularly clear from Fig. 3(a), where the specific-heat peaks corresponding to T_N do not scale. This again implies the existence of a small energy scale, only below which standard quantum critical scaling applies.

Phase diagram. Our findings are summarized in the phase diagram (Fig. 4), which displays the Néel temperature (from the peak position in C_p/T as a function of T) and the gap values extracted as in Fig. 2. The loss of magnetic order at H_{c} is accompanied by the closing of the magnetic excitation gap Δ. Figure 4 also shows the magnetic entropy S_{mag}, obtained from integrating the specific-heat data from Fig. 1(b). Focusing on $S_{\mathrm{mag}}(H)$ at fixed T, the entropy accumulation near H_{c} is clearly visible, as is the gap formation at elevated fields.

According to standard scaling, the gap values should follow a power law $\Delta \propto |H - H_{\mathrm{c}}|^{\nu z}$. This is approximately obeyed by the experimental data with $\nu z = 0.7$, but deviations are visible very close to H_{c}. These deviations could in principle arise from the transition being weakly first order (in which case the gap would not vanish at H_{c}). We have checked this possibility by performing field sweeps at 1.8 K searching for hysteresis [32]. However, the detected hysteresis in $M(H)$ is tiny, presumably arising from defects, such that we can exclude intrinsic first-order behavior. Hence, the deviations from power laws likely originate from sample inhomogeneities, as discussed above. Alternatively, the formation of an additional narrow low-T phase near H_{c} appears possible, as theoretically predicted in Ref. [36] for the classical Heisenberg-Kitaev model; this requires more detailed low-T measurements as a function of continuous H.

Mode softening and nature of the high-field phase. We now return to the specific-heat data and discuss them in the context of theoretical scenarios for the quantum phase transition (QPT) at H_{c}. The data show that LRO is lost above H_{c}. If the QPT at H_{c} is continuous, then this should be accompanied with a soft mode, i.e., the high-field phase should display a gapped mode with gap $\Delta \to 0$ as $H \to H_c^+$, with this mode condensation establishing zigzag LRO below H_c. The specific-heat data above H_c are consistent with these considerations.

An exciting possibility is that the phase above H_c is a field-induced spin liquid, accompanied by topological order. Then, the mode which softens at H_c would presumably correspond to an excitation of the emergent gauge field (dubbed vison for a Z_2 spin liquid). The field-induced spin liquid cannot exist up to arbitrarily high fields, i.e., a second QPT at a higher field H_{c2} should exist where the spin liquid is destroyed in favor of the high-field phase; this has not been experimentally tested to date. While indications for a field-induced spin liquid in Heisenberg-Kitaev models were found in numerical simulations in Ref. [19], a full theory is not available.

Alternatively, the phase above H_c could be adiabatically connected to the high-field limit, and the soft mode would then correspond to a high-field magnon. We note that such a magnon condensation is rather different from that in an SU(2)-symmetric Heisenberg magnet due to spin-orbit coupling: First, the zero-temperature magnetization above H_c can be far below saturation. Second, due to the low symmetry, the QPT is not of BEC type ($z = 2$, $\nu = \frac{1}{2}$) but generically in the Ising universality class ($z = 1$, $\nu = 0.630$ in $d = 2$).

We have studied this type of magnon-condensation transition in the framework of an appropriate J_1-K_1-Γ_1-J_3 model [20] in some detail (see the Supplemental Material [32]). Within our semiclassical approach, the critical exponents of the transition are $\nu = \frac{1}{2}$ and $z = 1$. The results [32], including the value of H_c, appear in semiquantitative agreement with the experimental data. This lends further credit to the presence of a field-induced QCP in α-$RuCl_3$ but does not allow us to conclusively identify the nature of the high-field phase. We also note that the theoretical calculation shows the presence of an additional energy scale arising from strong van Hove singularities in the magnon band structure at high fields. This energy scale varies approximately linearly with field above H_c but does not vanish at H_c (see Fig. S6). Note the distinctly different nature of the *apparent* excitation gap likely induced by the van Hove singularity compared to the real thermodynamic gap at low temperatures $T \lesssim 7$ K extracted in this work. The *apparent* gap appears at higher temperatures and scales differently with H. Beyond the semiclassical limit these elevated-energy features are likely to lose their sharp-mode character, possibly due to fractionalization, as has been found in related models at zero field [37].

Summary. Via low-temperature specific-heat measurements, we have demonstrated that the frustrated magnet α-$RuCl_3$ displays field-induced quantum criticality at $\mu_0 H_c \approx 6.9$ T applied in the *ab* plane. The high-field phase is characterized by a field-induced gap to magnetic excitations, which is clearly visible below ~2 K. Our scaling analysis of the low-T specific-heat data yields estimates for the critical exponents $d/z = 2.1 \pm 0.1$ and $\nu z = 0.7 \pm 0.1$, consistent with Ising universality. While we cannot draw conclusions about the nature of the high-field phase, we believe that the hypothesis of a field-induced spin liquid deserves further studies.

Importantly, the data also reveal the existence of a sub-meV energy scale near the QCP, above which the nature of the excitation spectrum changes. It is conceivable that this scale corresponds to a crossover from more conventional dispersive modes at low energies to exotic fractionalized excitations driven by Kitaev interactions. Studying the evolution of these excitations at higher fields is an exciting task for the future.

Note added. Recently, we became aware of parallel works [38,39] documenting related studies of α-$RuCl_3$ in a magnetic field. While Ref. [38] reported gapped magnetic excitations at fields above H_c, the results of Ref. [39] were interpreted in terms of gapless excitations in this regime. Interestingly, Ref. [38] quotes the order-parameter exponent at H_c to be $\beta = 0.28$, in reasonable agreement with the Ising value 0.326, suggesting a conventional Ising transition. However, in both Refs. [38,39] the measurements were restricted to temperatures above 2 K. Our data show that lower temperatures are required to reach the asymptotic regime.

Acknowledgments. We acknowledge insightful discussions with C. Hess, A. Isaeva, R. Moessner, S. Nagler, F. Pollmann, S. Rachel, M. Richter, and J. van den Brink. The phonon simulations were performed on resources provided by the Swedish National Infrastructure for Computing (SNIC) at the supercomputer centers in Linköping and Stockholm. This research has been supported by the DFG via SFB 1143. A.U.B.W. acknowledges support by the DFG under Grant No. WO 1532/3-2. S.-H.D. and K.-Y.C. were supported by the Korea Research Foundation (KRF) grant funded by the Korea government (MEST) (Grant No. 2009-0093817).

L.T.C. and L.J. contributed equally to this work.

[1] K. W. Plumb, J. P. Clancy, L. J. Sandilands, V. V. Shankar, Y. F. Hu, K. S. Burch, H.-Y. Kee, and Y.-J. Kim, Phys. Rev. B **90**, 041112 (2014).

[2] J. A. Sears, M. Songvilay, K. W. Plumb, J. P. Clancy, Y. Qiu, Y. Zhao, D. Parshall, and Y.-J. Kim, Phys. Rev. B **91**, 144420 (2015).

[3] R. D. Johnson, S. C. Williams, A. A. Haghighirad, J. Singleton, V. Zapf, P. Manuel, I. I. Mazin, Y. Li, H. O. Jeschke, R. Valentí, and R. Coldea, Phys. Rev. B **92**, 235119 (2015).

[4] M. Majumder, M. Schmidt, H. Rosner, A. A. Tsirlin, H. Yasuoka, and M. Baenitz, Phys. Rev. B **91**, 180401 (2015).

[5] Y. Kubota, H. Tanaka, T. Ono, Y. Narumi, and K. Kindo, Phys. Rev. B **91**, 094422 (2015).

[6] S. Sinn, C. H. Kim, B. H. Kim, K. D. Lee, C. J. Won, J. S. Oh, M. Han, Y. J. Chang, N. Hur, H. Sato, B.-G. Park, C. Kim, H.-D. Kim, and T. W. Noh, Sci. Rep. **6**, 39544 (2016).

[7] M. Ziatdinov, A. Banerjee, A. Maksov, T. Berlijn, W. Zhou, H. B. Cao, J.-Q. Yan, C. A. Bridges, D. G. Mandrus, S. E. Nagler, A. P. Baddorf, and S. V. Kalinin, Nat. Commun. **7**, 13774 (2016).

[8] D. Weber, L. M. Schoop, V. Duppel, J. M. Lippmann, J. Nuss, and B. V. Lotsch, Nano Lett. **16**, 3578 (2016).

[9] G. Jackeli and G. Khaliullin, Phys. Rev. Lett. **102**, 017205 (2009).
[10] J. Chaloupka, G. Jackeli, and G. Khaliullin, Phys. Rev. Lett. **105**, 027204 (2010).
[11] Z. Nussinov and J. van den Brink, Rev. Mod. Phys. **87**, 1 (2015).
[12] A. Kitaev, Ann. Phys. **321**, 2 (2006).
[13] L. J. Sandilands, Y. Tian, K. W. Plumb, Y.-J. Kim, and K. S. Burch, Phys. Rev. Lett. **114**, 147201 (2015).
[14] J. Nasu, M. Udagawa, and Y. Motome, Phys. Rev. B **92**, 115122 (2015).
[15] S. Trebst, arXiv:1701.07056.
[16] Y. Singh, S. Manni, J. Reuther, T. Berlijn, R. Thomale, W. Ku, S. Trebst, and P. Gegenwart, Phys. Rev. Lett. **108**, 127203 (2012).
[17] A. Banerjee, C. A. Bridges, J.-Q. Yan, A. A. Aczel, L. Li, M. B. Stone, G. E. Granroth, M. D. Lumsden, Y. Yiu, J. Knolle, S. Bhattacharjee, D. L. Kovrizhin, R. Moessner, D. A. Tennant, D. G. Mandrus, and S. E. Nagler, Nat. Mater. **15**, 733 (2016).
[18] H. B. Cao, A. Banerjee, J.-Q. Yan, C. A. Bridges, M. D. Lumsden, D. G. Mandrus, D. A. Tennant, B. C. Chakoumakos, and S. E. Nagler, Phys. Rev. B **93**, 134423 (2016).
[19] R. Yadav, N. A. Bogdanov, V. M. Katukuri, S. Nishimoto, J. van den Brink, and L. Hozoi, Sci. Rep. **6**, 37925 (2016).
[20] S. M. Winter, Y. Li, H. O. Jeschke, and R. Valentí, Phys. Rev. B **93**, 214431 (2016).
[21] I. Kimchi and Y.-Z. You, Phys. Rev. B **84**, 180407 (2011).
[22] J. G. Rau, E. K.-H. Lee, and H.-Y. Kee, Phys. Rev. Lett. **112**, 077204 (2014).
[23] Y. Yamaji, Y. Nomura, M. Kurita, R. Arita, and M. Imada, Phys. Rev. Lett. **113**, 107201 (2014).
[24] J. Reuther, R. Thomale, and S. Rachel, Phys. Rev. B **90**, 100405 (2014).
[25] Y. Sizyuk, C. Price, P. Wölfle, and N. B. Perkins, Phys. Rev. B **90**, 155126 (2014).
[26] I. Rousochatzakis, J. Reuther, R. Thomale, S. Rachel, and N. B. Perkins, Phys. Rev. X **5**, 041035 (2015).
[27] S. M. Winter, K. Riedl, A. Honecker, and R. Valentí, arXiv:1702.08466.
[28] M. Laubach, J. Reuther, R. Thomale, and S. Rachel, arXiv:1701.04896.
[29] I. A. Leahy, C. A. Pocs, P. E. Siegfried, D. Graf, S.-H. Do, K.-Y. Choi, B. Normand, and M. Lee, Phys. Rev. Lett. **118**, 187203 (2017).
[30] S.-H. Baek, S.-H. Do, K.-Y. Choi, Y. S. Kwon, A. U. B. Wolter, S. Nishimoto, J. van den Brink, and B. Büchner, arXiv:1702.01671.
[31] Note that the deviations in critical field values for $H||ab$ in different works can be attributed to differences in sample quality as well as to some extent to differences in the in-plane orientation. While high-quality samples with one single transition around $T_N \sim 6.5$ K show a critical field $\mu_0 H_c \sim 7.0(0.5)$ T, samples with a non-negligible amount of stacking faults and two transitions reach $\mu_0 H_c \sim 8.0(0.5)$ T.
[32] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.96.041405 for a magnetic characterization of the sample, the phonon calculations for $RhCl_3$, and for a semiclassical analysis of a J_1-K_1-Γ_1-J_3 model in a magnetic field.
[33] F. A. Lindemann, Phys. Z **11**, 609 (1910).
[34] S. Sachdev, *Quantum Phase Transitions*, 2nd ed. (Cambridge University Press, Cambridge, UK, 2011).
[35] M. Vojta, Rep. Prog. Phys. **66**, 2069 (2003).
[36] L. Janssen, E. C. Andrade, and M. Vojta, Phys. Rev. Lett. **117**, 277202 (2016).
[37] M. Gohlke, R. Verresen, R. Moessner, and F. Pollmann, arXiv:1701.04678.
[38] J. A. Sears, Y. Zhao, Z. Xu, J. W. Lynn, and Y.-J. Kim, Phys. Rev. B **95**, 180411 (2017).
[39] J. Zheng, K. Ran, T. Li, J. Wang, P. Wang, B. Liu, Z. Liu, B. Normand, J. Wen, and W. Yu, arXiv:1703.08474.