Evidence of the Berezinskii-Kosterlitz-Thouless phase in a frustrated magnet

Ze Hu1,13, Zhen Ma2,3,13, Yuan-Da Liao4,5,13, Han Li6,13, Chunsheng Ma1, Yi Cui1, Yanyan Shangguan2, Zhentao Huang2, Yang Qi7,8,9, Wei Li6,10,13, Zi Yang Meng4,11,12, Jinsheng Wen2,9 & Weiqiang Yu1,13

The Berezinskii-Kosterlitz-Thouless (BKT) mechanism, building upon proliferation of topological defects in 2D systems, is the first example of phase transition beyond the Landau-Ginzburg paradigm of symmetry breaking. Such a topological phase transition has long been sought yet undiscovered directly in magnetic materials. Here, we pin down two transitions that bound a BKT phase in an ideal 2D frustrated magnet TmMgGaO₄, via nuclear magnetic resonance under in-plane magnetic fields, which do not disturb the low-energy electronic states and allow BKT fluctuations to be detected sensitively. Moreover, by applying out-of-plane fields, we find a critical scaling behavior of the magnetic susceptibility expected for the BKT transition. The experimental findings can be explained by quantum Monte Carlo simulations applied on an accurate triangular-lattice Ising model of the compound which hosts a BKT phase. These results provide a concrete example for the BKT phase and offer an ideal platform for future investigations on the BKT physics in magnetic materials.
Topology plays an increasingly important role in understanding different phases and phase transitions in correlated quantum matters and materials. One prominent example is the Berezinskii–Kosterlitz–Thouless (BKT) mechanism in two-dimensional (2D) systems\textsuperscript{1,2,3}, which is associated with the binding and unbinding of topological defects. The BKT transition cannot be characterized by conventional order parameters and constitutes the earliest example of phase transition beyond the Landau–Ginzburg paradigm of spontaneous symmetry breaking. Historically, the BKT mechanism was introduced in the XY spin model and long predicted to occur in magnetic thin films\textsuperscript{4–13}. In experiments, signatures of the BKT transition have been observed in superfluid helium films\textsuperscript{6}, as well as in 2D superconducting films\textsuperscript{7,8} and arrays\textsuperscript{9}. Regarding the original proposal in layered XY-type magnets, despite intensive efforts\textsuperscript{10–15}, direct and unambiguous observation of the BKT transition is still lacking. One major obstacle is the three-dimensional (3D) couplings in the magnets, although weak, will inevitably enhance the configurational energy probe, to detect the BKT phase. We applied a moderate in-plane field $H = 3\, \text{T}$, which is adequate to collect the $^{69}$Ga NMR spectra (see Fig. 2c for details). The BKT phase between $T_U$ and $T_L$ is illustrated by the solid vertical line, while the AFM regime is indicated by the arrow. The contour background depicts the magnetic field in between the PM, BKT, and AFM phases. The $T^*$ denotes the temperature at a specific field where a peak is found in the differential susceptibility $dM/dH$, shown in Fig. 3b. The Curie–Weiss temperature $\theta_{\text{CW}}$ is obtained from the $1/T_1T$ (see Supplementary Fig. 1). Remarkably, $T^*$, $T^*_U$, $T^*_L$, and $\theta_{\text{CW}}$ all collapse to the same phase boundary between the BKT-like regime and AFM phase. A magnetically ordered phase is supposed to lie below the dome-like boundary. Errors represent 1 SD throughout the paper.

**Results**

**NMR probe of the BKT phase.** The obtained NMR spectra with an in-plane magnetic field $\mu_0H = 3\, \text{T}$ are shown in Fig. 2a at representative temperatures. To better resolve the magnetic transition, the hyperfine shifts $^{69}K_0$ of the spectra were analyzed and plotted in Fig. 2b as a function of temperature. Upon cooling, $^{69}K_0$ peaks at about 0.8–0.95 K and then starts to drop at lower temperatures. Therefore, the ordering temperature is determined to be $T_L \approx 0.9\, \text{K}$, consistent with neutron scattering experiments\textsuperscript{16,18}. In addition, both the second moments (width of the spectra) $C_m/T$ and the third moments (asymmetry of the spectra) $\theta_{\text{CW}}$ all collapse to the same phase boundary between the BKT-like regime and AFM phase. A magnetically ordered phase is supposed to lie below the dome-like boundary. Errors represent 1 SD throughout the paper.

As shown in Fig. 1, from our NMR measurements of the spin-lattice relaxation rate $1/T_1$, we identify $T_U \approx 1.9\, \text{K}$ and $T_L \approx 0.9\, \text{K}$, which represent the upper- and lower-BKT transitions, where a critical BKT phase resides at zero magnetic field in between the high-$T$ paramagnetic and low-$T$ antiferromagnetic phases. This finding is further substantiated by our scaling analysis of the measured susceptibility data near $T_U$, as well as the simulated NMR and susceptibility data using large-scale quantum Monte Carlo (QMC) calculations.

**Fig. 1 Phase diagram of TmMgGaO$_4$ under out-of-plane magnetic fields.** Under zero field, there are paramagnetic (PM), BKT, and antiferromagnetic (AFM) phases. The $T_U$ ($T_L$) is the upper (lower) BKT transition temperature, determined from the plateau structure in the NMR spin-lattice relaxation rate $1/T_1T$ (see Fig. 2c for details). The BKT phase between $T_U$ and $T_L$ is illustrated by the solid vertical line, while the AFM regime is indicated by the arrow. The contour background depicts the magnetic field in between the PM, BKT, and AFM phases. The $T^*$ denotes the temperature at a specific field where a peak is found in the differential susceptibility $dM/dH$, shown in Fig. 3b. The Curie–Weiss temperature $\theta_{\text{CW}}$ is obtained from the $1/T_1T$ (see Supplementary Fig. 1). Remarkably, $T^*$, $T^*_U$, $T^*_L$, and $\theta_{\text{CW}}$ all collapse to the same phase boundary between the BKT-like regime and AFM phase. A magnetically ordered phase is supposed to lie below the dome-like boundary. Errors represent 1 SD throughout the paper.
transition. In Fig. 2c, we show the $1/69T_1$ obtained under in-plane fields $\mu_0 H = 3$ T and 1 T, which reflects intrinsic spin fluctuations with zero out-of-plane field. At 3 T, $1/69T_1$ first decreases upon cooling from 10 K then suddenly increases below $T_{U} \approx 1.9$ K, indicating the onset of strong low-energy spin fluctuations. The data at 1 T show similar behaviors. Below $T_{1} \approx 0.9$ K, $1/69T_1$ drops sharply, consistent with the onset of the magnetic ordering as also inferred from the hyperfine shift. Here, $1/69T_1$ is dominated by the gapped long wavelength excitations in the ordered state. At the magnetic phase transition, a peaked feature in $1/T_1$ develops, caused by the gapless low-energy spin fluctuations with diverging correlation length. Remarkably, at temperatures between $T_{U} \approx 1.9$ K and $T_{1} \approx 0.9$ K, $1/69T_1$ exhibits a plateau-like structure, indicating the emergence of a highly fluctuating phase with diverging spin correlations yet no true long-range order, which is the hallmark of a BKT phase. Therefore, it is for the first time that such a phase is unambiguously observed in a magnetic crystalline material.

Our unbiased QMC simulations on the TLI model of the material (see “Methods”), with accurate coupling parameters determined in ref. 19, quantitatively justifies the existence of the BKT phase between $T_{L}$ and $T_{U}$. We computed $1/T_1$ and compared with the experiment below. Figure 2d shows the calculated $1/T_1$ by including fluctuations from all momentum points in the Brillouin zone (cf. Supplementary Fig. 2) and from $K'$ point in the vicinity of the K point (right scale), through large-scale QMC simulations (see “Methods”).
Fig. 3 Uniform magnetic susceptibility of TmMgGaO₄ and scaling analysis. a dc susceptibility χ as functions of temperatures under small out-of-plane (H//c) and in-plane (H//ab) fields. The latter is multiplied by a factor of 20 for visualizing purpose. The deviation of data below 2 K indicates the entry to the BKT phase and the field-suppression of magnetic correlations. b The differential susceptibility dM/dH under out-of-plane fields at different temperatures. The kinks at low fields, as denoted by the down arrows, suggest the transition from the BKT-like phase to the ordered phase (under the dome in Fig. 1) with increasing fields. The peaked features at high fields suggests a quantum phase transition to the polarized phase. c Fits of dM/dH to the power-law scaling function dM/dH ∼ H⁻α with α = 2/3 for the 0.4 and 0.8 K data, and α = 0.123 for 2.1 K data in the log-log scale. The 3 K data follow the α = 0 line in the paramagnetic phase. d dM/dH by the QMC calculations in the same field and temperature range as in c, and fits to the power-law function with exponents α, which give consistent results as experiments.

doublet, the interlayer couplings not included in our model calculations, and the lack of knowledge on the precise local hyperfine coupling constant, etc., may explain the difference remaining between Fig. 2c, d.

Universal magnetic susceptibility scaling. Magnetic susceptibility measurements were also performed to strengthen the finding of the BKT phase. In Fig. 3a, we show the overall temperature dependence of χ at different out-of-plane fields. For T < 2 K, χ increases monotonically upon cooling and barely changes with fields. However, for T > 2 K, approximately the upper BKT transition T_U as obtained from the 1/69T_J measurements, χ increases as the field decreases, suggesting the onset of peculiar magnetic correlations. With further cooling, the susceptibility gets flattened with temperature. The magnetization M(H) was further measured at selected temperatures (data shown in Supplementary Fig. 6), and for the sake of clarity, the differential susceptibility dM/dH is plotted in Fig. 3b. At around 2.5 T, a pronounced peak can be observed at low temperature, indicating the existence of a quantum phase transition, and the phase at lower fields should be a magnetically ordered phase, although its precise nature remains to be uncovered. Besides the high-field feature, for temperatures at 0.8 K and above, a kinked feature is clearly resolved on each dM/dH curve at low fields, whereas at 0.4 K, the low-field kink disappears, which poses a question of whether there is a quantum transition or merely a crossover from the zero-field AFM phase to the finite-field ordered phase under the dome in Fig. 1. The temperature and field values indicated by the down arrows in Fig. 3 are denoted as T' and T'' in the phase diagram (Fig. 1).

Field-theoretical analysis of the TLI model has predicted that upon applying a small out-of-plane field, the differential susceptibility dM/dH shall exhibit a divergent power-law behavior as dM/dH ∼ H⁻α in proximity to the BKT phase. At T_L, α has the value of 2/3, which corresponds to a critical exponent η = 1/9 at the lower-BKT transition and is originated from the sixfold symmetry breaking. The exponent α gradually decreases as temperature increases, and above an intermediate temperature between T_L and T_U, α = 0 due to non-universal contributions. This is exactly what we observe in Fig. 3c. We fit the dM/dH with the power-law function at different temperatures, with the fitting regime chosen between 0.6–0.9 T. At 0.8 K, α is very close to the expected value of 2/3 (and thus η ≈ 1/9) at T_L, which constitutes a remarkable fingerprint evidence for the BKT transition. At lower temperatures such as 0.4 K, the exponent is also close to 2/3, because the susceptibility saturates with temperature, as shown in Fig. 3a. At high temperatures, α drops rapidly to a small value 0.12 at 2.1 K and becomes effectively zero at 3.0 K.

Therefore, the susceptibility scaling gives the lower-BKT transition at about 0.8 K and upper transition probably between 2.1 and 3 K, in good agreement with the T_L and T_U estimated from NMR. These results are also fully consistent with our QMC calculations on the susceptibility shown in Fig. 3d. At T_L or lower, α is 2/3, then decreases to a very small exponent 0.086 at 2.67 K, and above 3 K, becomes zero within numerical uncertainty. Such a power-law behavior in dM/dH again signifies the finite-temperature window of the BKT phase with diverging magnetic correlations, which gives rise to the universal power-law scaling of magnetic susceptibility.

Discussion
We believe the findings in this work are of various fundamental values. Since the original proposal of a BKT phase in magnetic films, which also triggered the currently thriving research field of topology in quantum materials, tremendous efforts have been devoted to finding the BKT phase in magnetic crystalline materials, yet hindered by the obstacle outlined in the Introduction. Here, benefiting from NMR as a sensitive low-energy probe, and the nearly zero planar gyromagnetic factor in a TLI magnet TmMgGaO₄, we are able to reveal two BKT transitions and a critical BKT phase with an emergent XY symmetry. Together with the power-law behavior of the susceptibility and excellent agreement between our QMC simulation and experiment data, we unambiguously identify the long-sought BKT phase in a magnetic crystalline material.

Many intriguing questions are stimulated, based on the phase diagram in Fig. 1 obtained here. First, what is the nature of the ordered phase under finite fields, are there further exotic phases and transitions in the phase diagram, and will there be a field-induced quantum phase transition at the high-field side of the dome—these are all of great interests to be addressed in future studies. Second, it should be noted that the dynamical properties obtained by QMC calculations in Fig. 2 are computed on a large, while finite-size, 36 × 36 lattice, which already produces 1/Tₙ data in very good agreement with the experimental measurements.
Such a great agreement is surprising, given the possible existence of randomness from Ga/Ge site mixing in the material TmMgGaO₄, and also the lattice disorder revealed by the large high-temperature second moment of the NMR spectra (Supplementary Note 4). Although the random distribution in intrinsic transverse fields and spin couplings does not seem to alter the low-temperature spin-ordered phase and the sharp spin excitation line shapes, its intriguing effects on the finite-temperature phase diagram of TLI and also the compound TmMgGaO₄ call for further studies.

Third, in the study of BKT transition in superfluid systems, it has been observed experimentally and understood theoretically that additional dissipations also appear above the transition temperature due to fluctuations of vortices. Hence, the plateau of 1/Γₜ at 1.2 K, we did not find any change of 1/Γ₂, with two different radio frequency excitation levels (14 mT and 24 mT), and with different frequencies across the NMR line, within the error bar.

\[
\beta_{\text{Kosterlitz-Thouless}} = \frac{1}{\Lambda} \ln \left( \frac{\Lambda}{\sqrt{\pi}} \right)
\]

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Data availability

All numerical codes in this paper are available upon request to the corresponding authors.

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Author contributions

W.Q.Y. and J.S.W. designed the experiments, with proposals from Y.Q., W.L., and Z.Y.M. Z.M. grew and characterized the single crystals and performed susceptibility measurements and analysis, with help from Y.Y.S.G., Z.T.H., Z.H., and H.L. Z.H., C.S.M., and T.C. performed NMR measurements and analysis. Y.D.L. and H.L. carried out the large-scale quantum many-body calculations, with the guidance from Y.Q., W.L., and Z.Y.M. W.Q.Y., J.S.W., W.L., Z.Y.M., and Y.Q. wrote the manuscript with comments from all coauthors.

Competing interests

The authors declare no competing interests.

Additional information

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Correspondence and requests for materials should be addressed to Y.Q., W.L., Z.Y.M., J.W. or W.Y.

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