

9-15-2017

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Abstract

Experimental and theoretical investigations on itinerant ferromagnetic systems under pressure have shown that ferromagnetic quantum criticality is avoided either by a change of the transition order, becoming of the first order at a tricritical point, or by the appearance of modulated magnetic phases. In the first case, the application of a magnetic field reveals a wing-structure phase diagram as seen in itinerant ferromagnets such as ZrZn₂ and UGe₂. In the second case, no tricritical wings have been observed so far. Here, we report on the discovery of wing-structure as well as the appearance of modulated magnetic phases in the temperature-pressure-magnetic field phase diagram of LaCrGe₃. Our investigation of LaCrGe₃ reveals a double-wing structure indicating strong similarities with ZrZn₂ and UGe₂. But, unlike these simpler systems, LaCrGe₃ also shows modulated magnetic phases similar to CeRuPO. This finding provides an example of an additional possibility for the phase diagram of metallic quantum ferromagnets.

Disciplines

Condensed Matter Physics

Comments

This article is published as Kaluarachchi, Udharma S., Sergey L. Bud'ko, Paul C. Canfield, and Valentin Taufour. "Tricritical wings and modulated magnetic phases in LaCrGe₃ under pressure." *Nature Communications* 8 (2017): 546. DOI: [10.1038/s41467-017-00699-x](https://doi.org/10.1038/s41467-017-00699-x). Posted with permission.

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ARTICLE

DOI: 10.1038/s41467-017-00699-x

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Tricritical wings and modulated magnetic phases in LaCrGe_3 under pressure

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Experimental and theoretical investigations on itinerant ferromagnetic systems under pressure have shown that ferromagnetic quantum criticality is avoided either by a change of the transition order, becoming of the first order at a tricritical point, or by the appearance of modulated magnetic phases. In the first case, the application of a magnetic field reveals a wing-structure phase diagram as seen in itinerant ferromagnets such as ZrZn_2 and UGe_2 . In the second case, no tricritical wings have been observed so far. Here, we report on the discovery of wing-structure as well as the appearance of modulated magnetic phases in the temperature-pressure-magnetic field phase diagram of LaCrGe_3 . Our investigation of LaCrGe_3 reveals a double-wing structure indicating strong similarities with ZrZn_2 and UGe_2 . But, unlike these simpler systems, LaCrGe_3 also shows modulated magnetic phases similar to CeRuPO . This finding provides an example of an additional possibility for the phase diagram of metallic quantum ferromagnets.

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Suppressing a second-order, magnetic phase transition to zero temperature with a tuning parameter (pressure, chemical substitutions, magnetic field) has been a very fruitful way to discover many fascinating phenomena in condensed matter physics. In the region near the putative quantum critical point (QCP), superconductivity has been observed in antiferromagnetic¹ as well as ferromagnetic (FM) systems^{2–4}. One peculiarity of the clean FM systems studied so far is that the nature of the paramagnetic-ferromagnetic (PM-FM) phase transition always changes before being suppressed to zero temperature⁵: in most cases, the transition becomes of the first order^{6–11}. Recently, another possibility, where a modulated magnetic phase (AFM_Q) appears (spin-density wave, antiferromagnetic order), has been observed in CeRuPO^{12, 13}, MnP^{14, 15}, and LaCrGe₃¹⁶.

Several theories have been developed to explain those possibilities^{17–26}. When a FM transition becomes of the first order at a tricritical point (TCP) in the temperature T pressure p plane, the application of a magnetic field H along the magnetization axis reveals a wing structure phase diagram in the T - p - H space^{20, 27}. This is seen in UGe₂^{28, 29} and ZrZn₂³⁰ and is schematically represented in Fig. 1a. This phase diagram shows the possibility of a different kind of quantum criticality at the quantum wing critical point (QWCP). In contrast with the conventional QCP, symmetry is already broken by the magnetic

field at a QWCP. In the more recently considered case where the transition changes to a AFM_Q phase, no wing structure phase diagram has been reported, but it is found that the AFM_Q is suppressed by moderate magnetic field^{12, 13}. This second possible T - p - H phase diagram has been schematically presented in a recent review⁵ and reproduced in Fig. 1b.

Here, we report electrical resistivity measurements on LaCrGe₃ under pressure and magnetic field. We determine the T - p - H phase diagram and find that it corresponds to a third possibility where tricritical wings emerge in addition to the AFM_Q phase. This type of phase diagram is illustrated in Fig. 1c: it includes both the tricritical wings and the AFM_Q phase. In addition, the phase diagram of LaCrGe₃ shows a double wing structure similar to what is observed in the itinerant ferromagnets UGe₂³¹ and ZrZn₂³², but with the additional AFM_Q phase. LaCrGe₃ is the first example showing such a phase diagram.

Results

T - p phase diagram. Recently, we reported on the T - p phase diagram of LaCrGe₃¹⁶, which is reproduced in Fig. 1d. At ambient pressure, LaCrGe₃ orders ferromagnetically at $T_C = 86$ K. Under applied pressure, T_C decreases and disappears at 2.1 GPa. Near 1.3 GPa, there is a Lifshitz point³³ at which a new transition line appears. The transition corresponds to the appearance of a modulated magnetic phase (AFM_Q) and can be tracked up to 5.2 GPa. Muon-spin rotation (μ SR) measurements show that the AFM_Q phase has a similar magnetic moment as the FM phase but without net macroscopic magnetization¹⁶. In addition, band structure calculations suggest that the AFM_Q phase is characterized by a small wave-vector Q and that several small Q phases are nearly degenerate. Below the PM-AFM_Q transition line, several anomalies marked as gray cross in Fig. 1d can be detected in $\rho(T)$ ¹⁶. These other anomalies within the AFM_Q phase are compatible with the near degeneracy of different Q -states (shown as AFM_Q and AFM_{Q'}) with temperature and pressure driven transitions between states with differing wavevectors.

In this article, we determine the three dimensional T - p - H phase diagram of LaCrGe₃ by measuring the electrical resistivity of single crystals of LaCrGe₃ under pressure and magnetic field.

FM1 and FM2 phases. Whereas most of the features in Fig. 1d were well understood in ref. 16, we also indicate the pressure dependence of T_x ($d\rho/dT_{\max}$) at which a broad maximum is observed in $d\rho/dT$ below T_C and shown as orange triangles in Fig. 1d. At ambient pressure, $T_x \approx 71$ K. No corresponding anomaly can be observed in magnetization¹⁶, internal field¹⁶ or specific heat³⁴. Under applied pressure, T_x decreases and cannot be distinguished from T_C ($d\rho/dT_{\text{mid}}$) above 1.6 GPa. As will be shown, application of magnetic field allows for a much clearer appreciation and understanding of this feature.

Figure 2a shows the anomalies at T_x and T_C observed in the electrical resistivity and its temperature derivative at 1.14 GPa. For comparison, Fig. 2b shows ambient pressure data for UGe₂²⁸ where a similar anomaly at T_x can be observed. In UGe₂, this anomaly was studied intensively^{35–37}. It corresponds to a crossover between two ferromagnetic phases FM1 and FM2 with different values of the saturated magnetic moment^{35, 36}. Under pressure, there is a critical point at which the crossover becomes a first-order transition, which eventually vanishes where a maximum in superconducting-transition temperature is observed². In the case of LaCrGe₃, we cannot locate where the crossover becomes a first order transition, since the anomaly merges with the Curie temperature anomaly near 1.6 GPa, very close to the TCP. However, as we will show below, the two transitions can be separated again with applied magnetic field

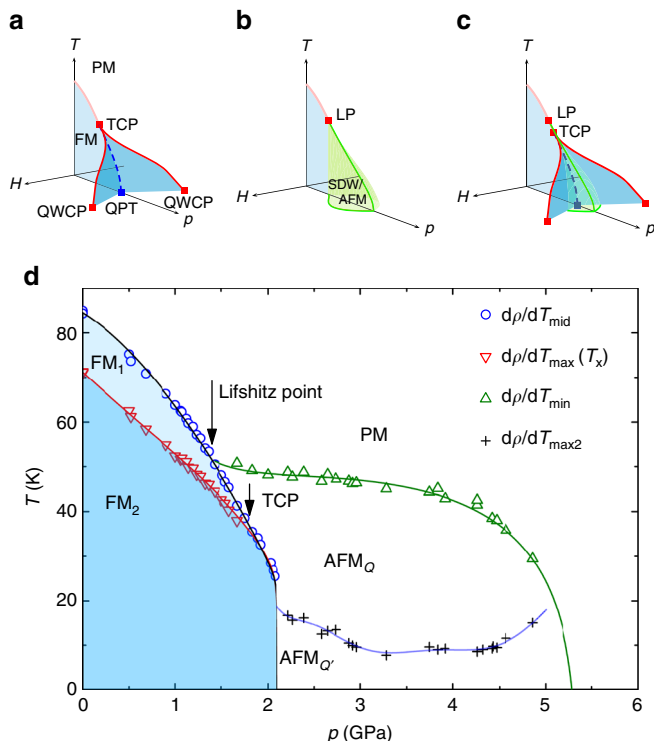


Fig. 1 Temperature–pressure phase diagram. **a** Schematic T - p - H phase diagram of a quantum ferromagnet: the paramagnetic-ferromagnetic (PM-FM) transition becomes of the first order at a tricritical point (TCP) after which there is a quantum phase transition (QPT) at 0 K. Tricritical wings emerge from the TCP under magnetic field and terminate at quantum wing critical points (QWCP). **b** Schematic T - p - H phase diagram of a quantum ferromagnet when a modulated magnetic phase (SDW/AFM) emerges from the Lifshitz point (LP). **c** New possible schematic T - p - H phase diagram for which tricritical wings as well as a new magnetic phase are observed. **d** T - p phase diagram of LaCrGe₃ from electrical resistivity measurements¹⁶ showing two FM regions (FM1 and FM2) separated by a crossover. The solid lines are guides to the eye

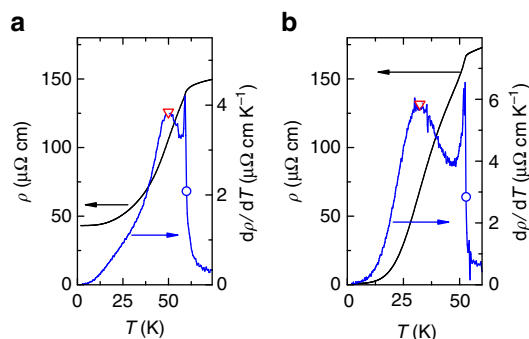


Fig. 2 Comparison of $\rho(T)$ and its $d\rho(T)/dT$ between LaCrGe_3 and UGe_2 . Temperature dependence of the resistivity (black line, left axis) and its derivative (blue line, right axis) of **a** LaCrGe_3 at 1.14 GPa and **b** UGe_2 at 0 GPa from ref. ²⁸. The crossover between the two ferromagnetic phases (FM1 and FM2) is inferred from the maximum in $d\rho/dT$ (T_x) and marked by a red triangle, whereas the paramagnetic-ferromagnetic transition is inferred from the middle point of the sharp increase in $d\rho/dT$ (T_C) and indicated by a blue circle

above 2.1 GPa. This is similar to what is observed in UGe_2 where the PM-FM1 and FM1-FM2 transition lines separate more and more as the pressure and the magnetic field are increased. Because of such similarities with UGe_2 , we label the two phases FM1 and FM2 and assume that the anomaly at T_x corresponds to a FM1-FM2 crossover. A similar crossover was also observed in ZrZn_2 ³². In refs ^{18, 25}, a Stoner model with two peaks in the density of states near the Fermi level was proposed to account for the two phases FM1 and FM2, reinforcing the idea of the itinerant nature of the magnetism in LaCrGe_3 .

Field-dependent resistivity measurement under pressure. In zero field, for applied pressures above 2.1 GPa, both FM1 and FM2 phases are suppressed. Upon applying a magnetic field along the c -axis, two sharp drops of the electrical resistivity can be observed (Fig. 3a) with two corresponding minima in the field derivatives (Fig. 3b). At 2 K, clear hysteresis of $\Delta H \sim 0.7$ T can be observed for both anomalies indicating the first order nature of the transitions. The emergence of field-induced first-order transitions starting from 2.1 GPa and moving to higher field as the pressure is increased (Supplementary Note 1) is characteristic of the FM quantum phase transition: when the PM-FM transition becomes of the first order, a magnetic field applied along the magnetization axis can induce the transition resulting in a wing structure phase diagram such as the one illustrated in Fig. 1a. In the case of LaCrGe_3 , evidence for a first order transition was already pointed out because of the very steep pressure dependence of T_C near 2.1 GPa and the abrupt doubling of the residual ($T = 2$ K) electrical resistivity¹⁶. In UGe_2 or ZrZn_2 , the successive metamagnetic transitions correspond to the PM-FM1 and FM1-FM2 transitions. In LaCrGe_3 , at 2 K, due to the presence of the AFM_Q phase at zero field, the transitions correspond to AFM_Q-FM1 and FM1-FM2.

Determination of the wing structure phase diagram. As the temperature is increased, the hysteresis decreases for both transitions, as can be seen in Fig. 3c, d and disappears at a wing critical point (WCP). Also, the transition width is small and weakly temperature dependent below the WCP and it broadens when entering in the crossover regime. Similar behavior has been observed in UGe_2 ²⁹. At 2.39 GPa for example, we locate the WCP of the first-order FM1 transition around 13.5 K and the one of the first-order FM2 transition around 12 K. At this temperature and

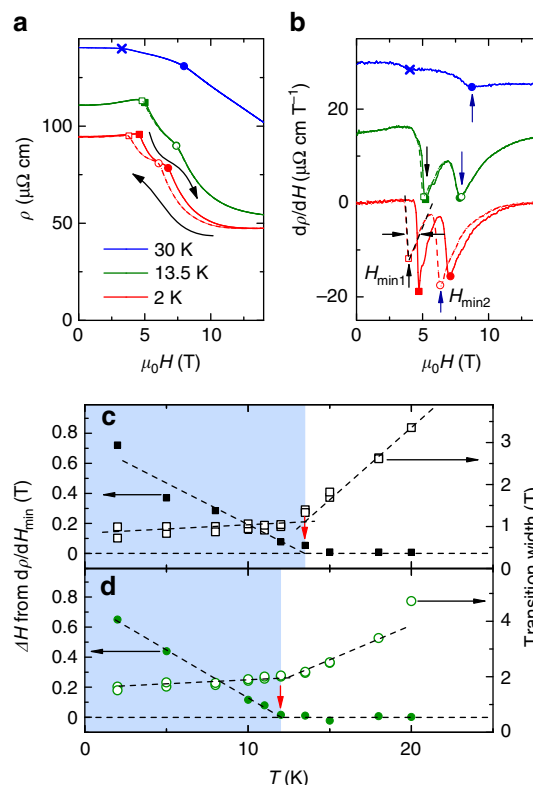


Fig. 3 Determination of wing critical point at 2.39 GPa. **a** Field dependence of the electrical resistivity at 2, 13.5, and 30 K at 2.39 GPa with applied field along the c -axis. Continuous and dashed lines represent the field increasing and decreasing, respectively. When the continuous and dashed lines do not overlap, there is indication for hysteresis. **b** Corresponding field derivatives ($d\rho/dH$). The curves are shifted by $15 \mu\Omega \text{ cm T}^{-1}$ for clarity. Vertical arrows represent the minima. The transition width is determined by the full width at half minimum as represented by horizontal arrows. Solid and open symbols in **a, b** represent the transition fields for field increasing and decreasing (Squares for AFM_Q-FM1 and circles for FM1-FM2). The blue cross symbols in **a, b** represent the AFM_Q-PM transition at this pressure. The temperature dependence of the hysteresis width of $H_{\text{min}1}$ and $H_{\text{min}2}$ (solid symbols) are shown in **c, d** (left axes). The hysteresis width gradually decreases with increasing temperature and disappears at T_w corresponding to the wing critical point (vertical red arrows). The right axes show the temperature dependence of the transition widths (open symbols). Dashed lines are guides to the eye. The width is small for the first-order transition and becomes broad in the crossover region. The blue-color shaded area represents the first order transition region, whereas the white color area represents the crossover region. These allow for the determination of the wing critical point of the FM1 transition at 13.5 ± 1.5 K, 2.39 GPa, and 5.1 T and the one for the FM2 transition at 12 ± 1 K, 2.39 GPa and 7.7 T

pressure, the transitions occur at 5.1 and 7.7 T, respectively. This allows for the tracking of the wing boundaries in the T - p - H space up to our field limit of 14 T. At low field, near the TCP, the wing boundaries are more conveniently determined as the location of the largest peak in $d\rho/dT$ (Supplementary Note 2).

The projections of the wings lines $T_w(p, H)$ in the T - H , T - p , and H - p planes are shown in Fig. 4a–c, respectively. The metamagnetic transitions to FM1 and FM2 start from 2.1 GPa and separate in the high field region as the pressure is further increased. For the FM1 wing, the slope dT_w/dH_w is very steep near $H = 0$ (Fig. 4a), whereas dH_w/dp_w is very small (Fig. 4c). This is in agreement with a recent theoretical analysis based on the Landau expansion of the free energy, which shows that dT_w/dH_w and dp_w/dH_w are infinite at the TCP²⁷. This fact was overlooked

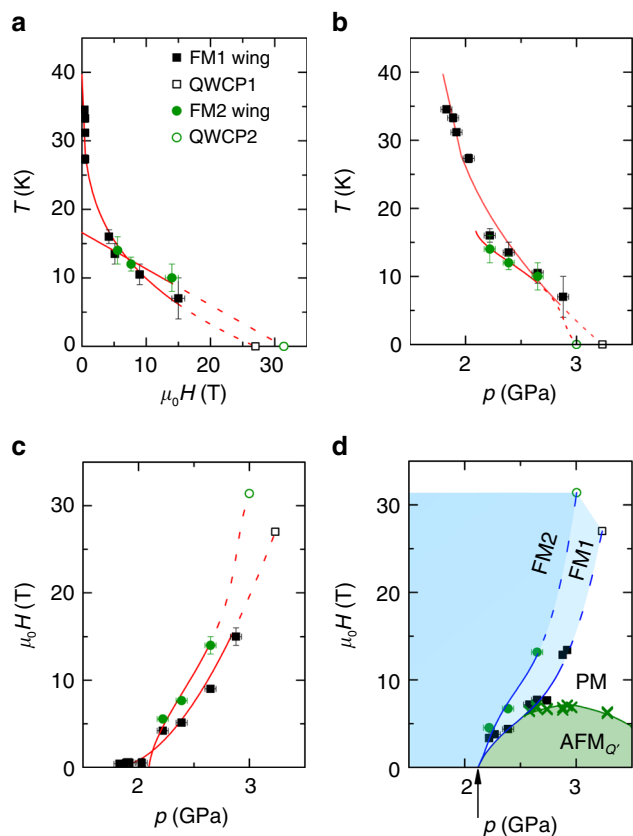


Fig. 4 Shape of the tricritical wing lines (T_w , p_w , H_w). Projection of the wings in **a** T - H , **b** T - p and **c** H - p planes. Black solid squares and green solid circles represent the FM1-wing and FM2-wing, respectively. Red lines (represented in the T - p - H space in Fig. 5) are guides to the eyes and open symbols represent the extrapolated quantum wing critical point (QWCP). **d** H - p phase diagram at 2 K. The arrow represents the pressure $p_c = 2.1$ GPa. The green color cross symbols represent the AFM_Q phase boundary. Dark blue, light blue, and green color shaded areas represent the FM2, FM1, and AFM_Q phases, respectively. The error bars in pressure for **b**-**d** are determined by the superconducting transition width of the Pb manometer. For **a**-**d**, the error bars in temperature and magnetic field are determined as half the data spacing

in the previous experimental determinations of the wing structure phase diagram in UGe_2 ^{28,29} and $ZrZn_2$ ³⁰, but appears very clearly in the case of $LaCrGe_3$. In the low field region, there are no data for the FM2 wing since the transition is not well separated from the FM1 wing, but there is no evidence for an infinite slope near $H = 0$. The wing lines can be extrapolated to QWCs at 0 K in high magnetic fields of the order of ~ 30 T (Fig. 4a) and pressures around ~ 3 GPa (Fig. 4b).

Figure 4d shows the H - p phase diagram at low temperature ($T = 2$ K). The magnetic field at the transition to the FM1 phase increases rapidly, whereas the field suppressing the AFM_Q phase does not exceed 7 T. Above 2.5 GPa, the AFM_Q and FM1 phases are separated by a region corresponding to the polarized PM phase. We note that a similar diagram where the wings extend beyond the AFM phase was recently obtained theoretically in the case of strong quantum fluctuations effects³⁸. The similarity of the H - p phase diagram at 2 K (Fig. 4d) and the projection of the wings in the H - p plane (Fig. 4c) reveals the near vertical nature of the wings.

T - p - H phase diagram of $LaCrGe_3$. The resulting three-dimensional T - p - H phase diagram of $LaCrGe_3$ is shown in Fig. 5,

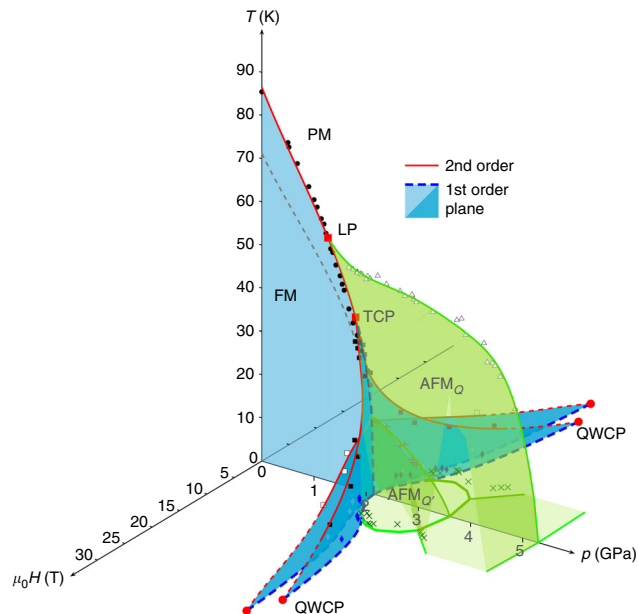


Fig. 5 T - p - H phase diagram of $LaCrGe_3$. Red solid lines are the second order phase transition and blue color planes are planes of first order transitions. Green color areas represent the AFM_Q and AFM_Q' phases. For clarity, only data points at 2 K and along the transitions lines are shown. Black circles indicate the paramagnetic (PM)-ferromagnetic (FM) transition. Black squares indicate the wing critical line for FM1 and empty squares indicate the wing critical line for FM2. Blue diamonds indicate the first order transition to the FM1 state at 2 K and empty diamonds, the first order transition to FM2 at 2 K. Empty triangles indicate the PM-modulated magnetic phase (AFM_Q) and “+” indicate the AFM_Q' boundary. “x” indicate the AFM_Q or AFM_Q' boundary at 2 K. The full set of data points can be found in Supplementary Note 3. For clarity, the green surfaces representing the boundaries for the AFM_Q or AFM_Q' phases are shown only in the region of positive magnetic field

which summarizes our results (Several of the constituent T - H phase diagrams, at various pressures, are given in Supplementary Fig. 3). The double wing structure is observed in addition to the AFM_Q phase. This is the first time that such a phase diagram is reported. Other materials suggested that there is either a wing structure without any additional magnetic phase^{28–30}, or a magnetic phase without a wing structure^{12,13}. The present study illustrates a third possibility where all such features are observed. The phase diagram of $LaCrGe_3$ and the existence of wings clearly establish that the quantum phase transition from FM to AFM_Q is of the first order. It is plausible that the reason is the same as for the FM to PM transition, but no theory is available for this case, as pointed out in a recent review⁵. Another interesting aspect is the existence of the two metamagnetic transitions (to FM1 and FM2) which suggests that this might be a generic feature of itinerant ferromagnetism. Indeed, it is observed in $ZrZn_2$, UGe_2 , and $LaCrGe_3$, although these are very different materials with different electronic orbitals giving rise to the magnetic states. We note that a wing structure has also been determined in the PM compounds $UCoAl$ ^{39–41} and $Sr_3Ru_2O_7$ ⁴², implying that a FM state probably exists at negative pressures in these materials. Strikingly, two anomalies could be detected upon crossing the wings in $UCoAl$ (two kinks of a plateau in electrical resistivity³⁹, two peaks in the ac susceptibility⁴¹), as well as in $Sr_3Ru_2O_7$ (two peaks in the ac susceptibility⁴²). These double features could also correspond to a double wing structure.

Conclusions. To conclude, the T - p - H phase diagram of $LaCrGe_3$ provides a distinct example of possible outcomes of FM quantum

criticality. At zero field, quantum criticality is avoided by the appearance of a new modulated magnetic phase, but the application of magnetic field shows the existence of a wing structure phase diagram leading towards QWCP at high field. These experimental findings reveal insights into the possible phase diagrams of FM systems. The emergence of the wings reveals a theoretically predicted tangent slope²⁷ near the TCP, a fact that was overlooked in previous experimental determination of phase diagrams of other compounds because of the lack of data density in that region. In addition, the double nature of the wings appears to be a generic feature of itinerant ferromagnetism, as it is observed in several, a priori, unrelated materials. This result deserves further theoretical investigations and unification.

Methods

Sample preparation. Single crystals of LaCrGe₃ were grown using a high-temperature solution growth technique^{43,44}. A mixture of La, Cr, and Ge with a molar ratio of La:Cr:Ge = 13:13:74 was premixed by arc melting. The material was then placed in a 2-mL alumina crucible and sealed in a silica ampoule under partial pressure of high purity argon gas. The sealed ampoule was heated to 1100 °C over 3 h and held for 5 h. After that, it was cooled to 825 °C and the remaining liquid was decanted using a centrifuge. Details about the crystal growth procedure and sample characterization at ambient pressure is described in ref. ³⁴.

The resistivity measurements under pressure. The samples for the pressure study were selected after ambient pressure characterization by the magnetization and resistivity measurements. Temperature and field dependent resistance measurements were carried out using a Quantum Design Physical Property Measurement System from 1.8 to 300 K. The resistivity was measured by the standard four-probe method with the current in the *ab* plane. Four Au wires with diameter of 12.5 μm were spot welded to the sample. A magnetic field, up to 9 or 14 T, was applied along the *c*-axis, which corresponds to the magnetization easy axis^{34,45}.

Two types of pressure cells were used for this experiment. A Be-Cu/Ni-Cr-Al hybrid piston-cylinder cell, similar to the one described in ref. ⁴⁶, was used for pressures up to 2.1 GPa. A mixture of 4:6 light mineral oil: *n*-pentane⁴⁶ used as a pressure medium, which solidifies at ~3–4 GPa at room temperature⁴⁷. For higher pressures, a modified Bridgman cell⁴⁸ was used to generate pressure up to 6 GPa. A 1:1 mixture of *n*-pentane:iso-pentane was used as a pressure medium. The solidification of this medium occurs around ~6–7 GPa at room temperature^{47,49}. For both cells, the pressure at low temperature was determined by the superconducting transition temperature of Pb⁵⁰ measured by the resistivity.

The resistivity measurement under pressure at zero field is described in ref. ¹⁶.

Data availability. The data that support the findings of this study are available on request from the corresponding authors.

Received: 4 November 2016 Accepted: 23 June 2017

Published online: 15 September 2017

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Acknowledgements

We would like to thank S.K. Kim, X. Lin, V.G. Kogan, D.K. Finnemore, E.D. Mun, H. Kim, Y. Furukawa, R. Khasanov for useful discussions. This work was carried out at the Iowa State University and the Ames Laboratory, US DOE, under Contract No. DE-AC02-07CH11358. This work was supported by the Materials Sciences Division of the Office of Basic Energy Sciences of the U.S. Department of Energy. V.T. was partly supported by Ames Laboratory's laboratory-directed research and development (LDRD) funding.

Author contributions

V.T. and P.C. initiated this study. U.K., V.T., and P.C. prepared the single crystals. U.K., V.T., and S.B. performed the pressure measurements. U.K., V.T., S.B., and U.K. analyzed and interpreted the pressure data. U.K. and V.T. wrote the manuscript with the help of all authors.

Additional information

Supplementary Information accompanies this paper at doi:10.1038/s41467-017-00699-x.

Competing interests: The authors declare no competing financial interests.

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