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# Ferromagnetic quantum criticality: new aspects from the phase diagram of LaCrGe<sub>3</sub>

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## Abstract

Recent theoretical and experimental studies have shown that ferromagnetic quantum criticality is always avoided in clean systems. Two possibilities have been identified. In the first scenario, the ferromagnetic transition becomes of the first order at a tricritical point before being suppressed. A wing structure phase diagram is observed indicating the possibility of a new type of quantum critical point under magnetic field. In a second scenario, a transition to a modulated magnetic phase occurs. Our recent studies on the compound LaCrGe<sub>3</sub> illustrate a third scenario where not only a new magnetic phase occurs, but also a change of order of the transition at a tricritical point leading to a wing-structure phase diagram. Careful experimental study of the phase diagram near the tricritical point also illustrates new rules near this type of point.

*Keywords:* ferromagnet, quantum criticality, tricritical point, wing structure, Lifshitz point

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The studies of phase transitions have led to a wide range of physical concepts, industrial applications, and new properties and phenomena which challenge our understanding. Over the last forty years, part of the condensed matter scientific community has turned its attention to quantum phase transitions. Quantum phase transitions are a kind of transitions that occurs at low temperature as a function of a non-thermal parameter such as pressure, magnetic or electric field, chemical substitution. In this article, we address the case of the ferromagnetic transition for which a change from an ordered ferromagnetic state at low temperature to a disordered paramagnetic state at high temperature occurs at the Curie temperature. In the same way as pressure can initially decrease the freezing point of water, physicist have been able to use pressure to decrease the Curie temperature to absolute zero temperature, leading to a quantum phase transition. Surprisingly, a variety of unconventional properties have been unveiled near the ferromagnetic quantum phase transition [1], including superconductivity [2–5], non-Fermi liquid behavior [6], tri-criticality [5, 7–10], and complex magnetic structures [11–16].

Despite the diversity of properties and phenomena, a few features seem to emerge as generic for the phase diagram of ferromagnetic systems near a quantum phase transition. Indeed, it appears that whenever the ferromagnetic transition is suppressed toward a quantum phase transition using a clean parameter such as pressure, the nature of the transition changes. Two possibilities have been observed: the transition changes from second to first order

at a tricritical point, or a new magnetic phase appears at a Lifshitz point. When the transition becomes of the first order, magnetic-field induced transitions are observed leading to a “wing-structure” phase diagram in the temperature, pressure, magnetic field ( $T$ - $p$ - $H$ ) space. We recently determined the  $T$ - $p$ - $H$  phase diagram of LaCrGe<sub>3</sub> and found that it combines both possibilities: we observe a wing-structure phase diagram as well as a new magnetic phase. LaCrGe<sub>3</sub> provides another example of the unexpected outcome of the phase diagram of a ferromagnet, but at the same time we will discuss certain aspects that appears to be more universal. The myriad of fascinating phenomena associated with the phase diagram of ferromagnetic systems is illustrated in Fig.1, modified from Ref. [17].

LaCrGe<sub>3</sub> crystallizes in an hexagonal crystal structure (space group number 194:  $P6_3/mmc$ ) [18, 19]. It becomes ferromagnetic below 86 K [20] with the magnetic moment aligned along the  $c$ -axis of the unit cell. We show the crystallographic unit cell with the magnetic alignment in Fig. 2a. Arrott plots [21] shown in Fig. 2b. indicate a Curie temperature of  $T_C = 86.2$  K. The effective moment obtained from a fit of the magnetic susceptibility to the Curie-Weiss law is  $2.38 \mu_B/\text{Cr}$  (Fig. 2c), which is smaller than the value expected for  $\text{Cr}^{3+}$  ions ( $3.8 \mu_B$ ). In addition, the saturated moment is about  $1.25 \mu_B/\text{Cr}$  (Fig. 2d), leading to a Rhodes-Wohlfarth ratio of 1.26 [22], and indicating that there is some degree of delocalization of the magnetism in this compound. The magnetic anisotropy is quite large, with an anisotropy field of 4.5 T at 2 K, as shown in Fig. 2d.

We recently established the temperature-pressure phase

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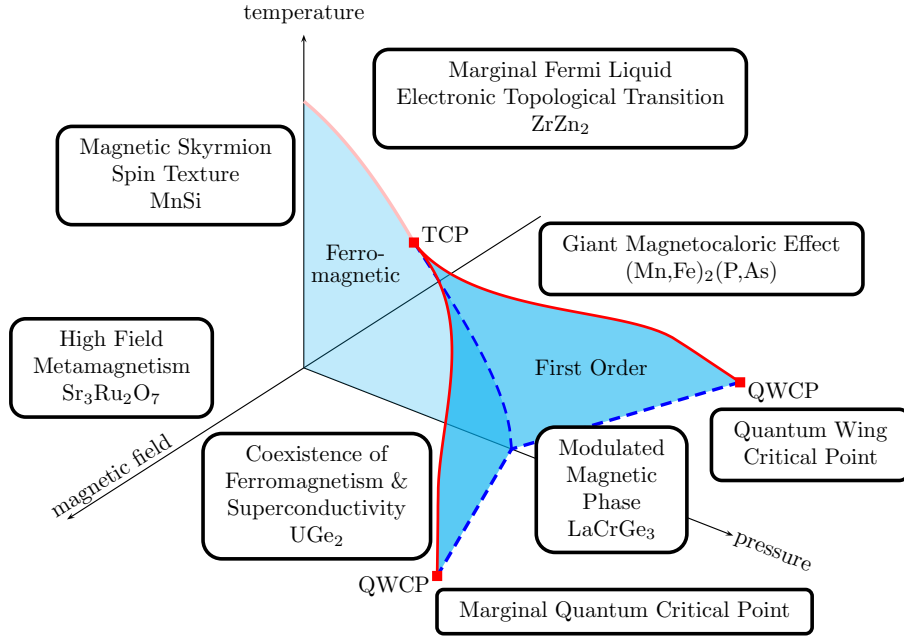


Figure 1: Typical temperature - pressure - magnetic field ( $T$ - $p$ - $H$ ) phase diagram near fragile ferromagnetism (modified from Ref. [17]). The wing-structure is shown as first-order transition planes which terminate at quantum wing critical points. Selected examples of phenomena observed on the border of ferromagnetism are also listed.

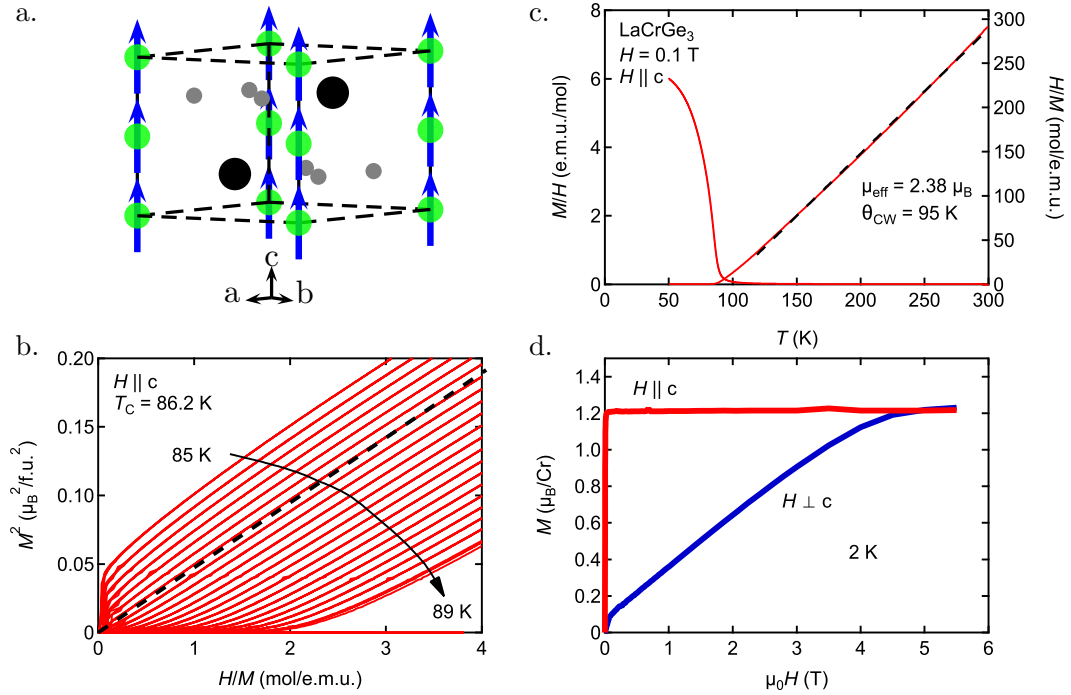


Figure 2: a. The crystallographic unit cell of  $\text{LaCrGe}_3$ . The arrows represent the Cr moments in the ferromagnetic state. b. Arrott plots measured with a magnetic field applied along the  $c$ -axis. The isotherms are measured from 85 K to 89 K in steps of 0.2 K. c. Temperature dependence of the magnetic susceptibility (left axis) and the inverse (right axis) with a field of 0.1 T along the  $c$ -axis. A fit to the Curie-Weiss law is shown as a dashed line. d. Field dependence of the magnetization at 2 K measured parallel and perpendicular to the  $c$ -axis of the hexagonal crystal structure.

diagram of  $\text{LaCrGe}_3$  from various measurements [16] (Fig. 3). Magnetization measurements showed that the ferromagnetic state is suppressed by 2.1 GPa. Resistivity measurements revealed another anomaly: a bump in the temperature dependence which could be tracked as a function of pressure as the green line (green triangles) on the phase diagram shown in Fig. 3. Muon spin-rotation showed that the internal field in this new phase is similar to the one in the ferromagnetic state. That demonstrates the magnetic nature of the new phase, therefore labeled as antiferromagnetic ( $\text{AFM}_Q$ ). The wavevector  $Q$  is unknown and neutron measurements under pressure are underway to determine the exact magnetic ordering. First-principle band structure calculations showed that several states with small  $Q$ -vectors are nearly degenerate under pressure and become more stable than the ferromagnetic state [16]. Small wavevectors such as  $Q < 1/4$  reciprocal lattice units ( $2\pi/c$ ) represent long-wavelength orders which would be consistent with a similar internal field as in the ferromagnetic state.

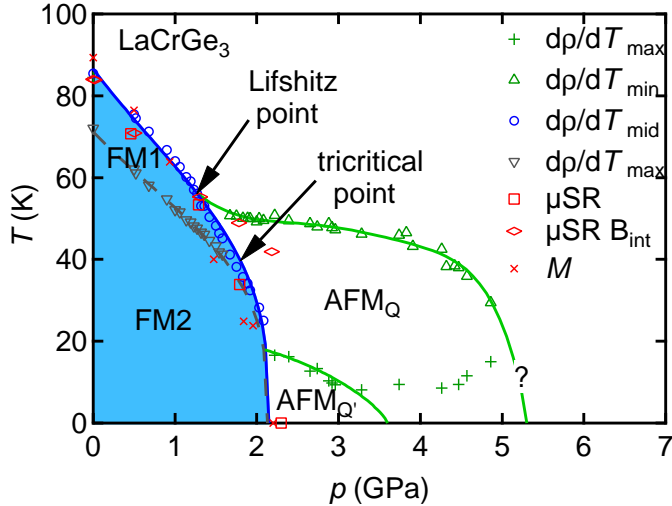


Figure 3: Temperature-pressure phase diagram of  $\text{LaCrGe}_3$  determined from magnetization, electrical resistivity and muon spin-rotation [16].

$\text{LaCrGe}_3$  is therefore an example of a simple  $3d$ -electrons ferromagnetic system for which a new magnetic phase appears as the Curie temperature is suppressed by pressure. Interestingly, this scenario has been considered recently by several theories [15, 23–29]. The idea is that particle-hole excitations corresponding to long wavelength correlations lead either to a change of order of the transition to the first order or to a new magnetic phase. As already mentioned,  $\text{LaCrGe}_3$  shows both cases: the transition between the ferromagnetic phase and  $\text{AFM}_Q$  becomes of the first order as well.

The first indication for a first order transition is the very steep slope of the pressure dependent FM- $\text{AFM}_Q$  transition line. The Clapeyron relation imposes that the slope is infinite at zero temperature for a first order tran-

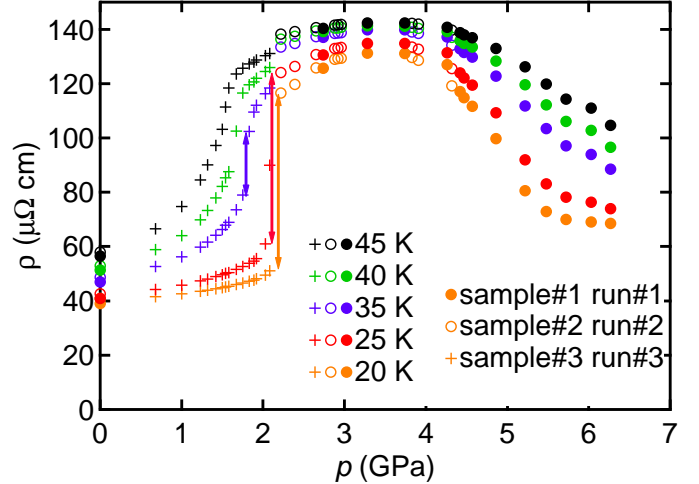


Figure 4: Pressure dependence of the resistivity at various temperature. Different symbols represent data of different samples from different pressure runs (run#1 and run#2 in modified Bridgman cells, run#3 in a clamp cell). The small disconnect near 2.2 GPa (most clearly seen in the  $T = 45$  or  $40$  K data sets) reflects a small sample and/or pressure cell dependence. The discontinuity characteristic of the first order FM- $\text{AFM}_Q$  transition is indicated by arrows [16].

sition. The second indication is the sharp discontinuity in the pressure dependence of the electrical resistivity [16]. The discontinuity is indicated by arrows in Fig. 4. It vanishes around 40 K which corresponds to the temperature of the so-called tricritical point: the point at which the transition changes between first and second order.

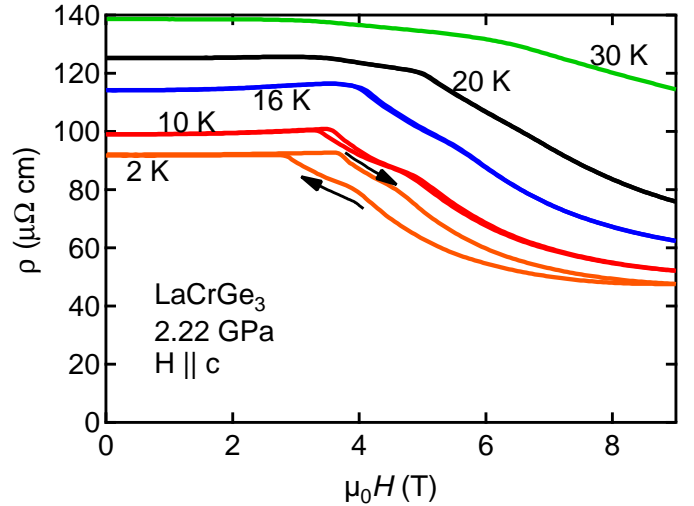


Figure 5: Magnetic field dependence of electrical resistivity at 2.22 GPa at various temperatures. Data for increasing and decreasing field reveal hysteresis behavior at low temperature. Data at 2.39, 2.65, and 2.88 GPa are presented in Ref. [10].

The stronger evidence for a first order transition comes from the observation of metamagnetic transitions for temperature below and pressure above the tricritical point

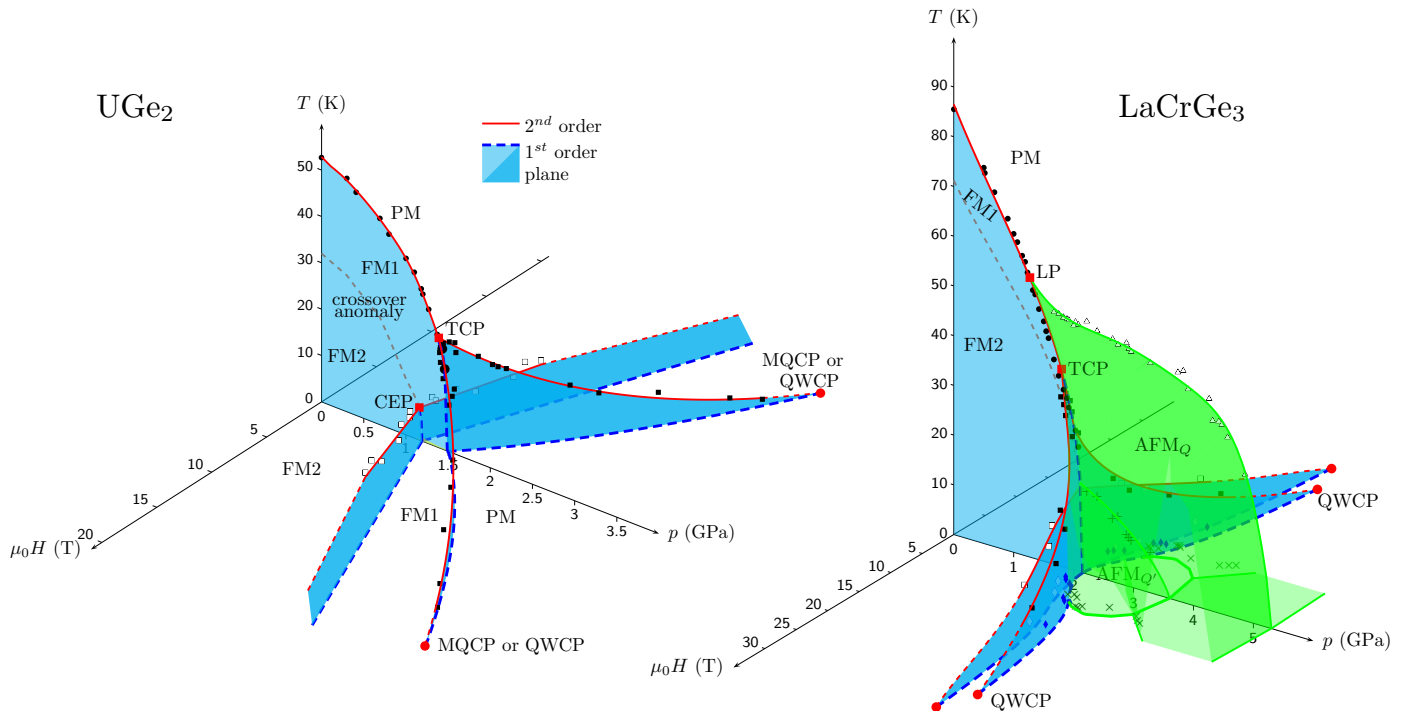


Figure 6: Temperature-pressure-magnetic field phase diagrams of  $\text{UGe}_2$  as determined from Refs. [8, 30, 31] and  $\text{LaCrGe}_3$  from Refs [10, 16].

when the field is applied along the easy axis of magnetization (c-axis) [10]. Indeed, this is a direct consequence of the transition being of the first order: as a first order ferromagnetic transition is suppressed, an applied magnetic field is able to re-induce the transition. Figure 5 shows the field induced transitions detected in resistivity measurements. Interestingly, there are two successive transitions, as in the case of  $\text{UGe}_2$  [32]. Another related similarity with  $\text{UGe}_2$  is a broad anomaly in the temperature dependence of resistivity at low pressures within the FM state [10]. This indicates that, similar to  $\text{UGe}_2$ ,  $\text{LaCrGe}_3$  has two ferromagnetic states (FM1 and FM2) which differ in magnetization value. At 2.22 GPa (Fig. 5), the magnetic field first induces the FM1 state with a smaller moment, followed by the FM2 state with a larger moment. The existence of multiple FM states is remarkable and calls for a re-consideration of other systems with ferromagnetic quantum phase transitions. Indeed, it seems that this could be a generic feature since it is observed in many systems. We note that in  $\text{UGe}_2$ , the crossover between the two states at ambient pressure is better revealed in thermal expansion measurements [33]. It will be interesting to perform similar studies on  $\text{LaCrGe}_3$ . Besides  $\text{UGe}_2$  and  $\text{LaCrGe}_3$ , two ferromagnetic states have also been reported in  $\text{ZrZn}_2$  [34]. In  $\text{UCoAl}$ , the anomaly corresponding to the field induced transition to the FM state shows two peaks in the ac susceptibility [35] and two kinks forming a plateau in electrical resistivity [36]. Two peaks in the ac susceptibility have also been reported in the metamagnetic transition of  $\text{Sr}_3\text{Ru}_2\text{O}_7$  [37]. Although the magnetism in these systems

originates from different electronic shells, it is remarkable that the multiple ferromagnetic states seems universal. In the case of  $\text{UGe}_2$ , a Stoner model with two peaks in the electronic density of states has been proposed to explain the origin of the FM1 and FM2 transitions [38, 39].

Another observation of the phase diagram of  $\text{LaCrGe}_3$  points toward a more general property of ferromagnetic quantum systems: the tangent merging of so-called “wings” at the tricritical point. As can be seen in Fig. 5, the hysteresis behavior of the metamagnetic transitions disappears at a critical point as the temperature is increased. The lines of critical points form a wing-structure phase diagram in the  $T$ - $p$ - $H$  space which has been determined recently in  $\text{LaCrGe}_3$  [10], and is shown in Fig. 6. Similar experimental studies have revealed other wing-structure phase diagrams in  $\text{UGe}_2$  [8, 30, 31] (also shown in Fig. 6 for comparison),  $\text{Sr}_3\text{Ru}_2\text{O}_7$  [37],  $\text{ZrZn}_2$  [9],  $\text{UCoAl}$  [35, 36, 40]. The behavior near the tricritical point could not be studied in  $\text{Sr}_3\text{Ru}_2\text{O}_7$  and  $\text{UCoAl}$  because those compounds are already in the paramagnetic regime at ambient pressure. Substitutions studies revealed the existence of a nearby tricritical point in  $\text{UCoAl}$  [40]. In  $\text{ZrZn}_2$ , the tricritical point is near 3 K, making the experimental investigations difficult. In  $\text{UGe}_2$  and  $\text{LaCrGe}_3$ , the tricritical point is near 24 and 40 K, respectively. During our recent investigation of the wing structure phase diagram of  $\text{LaCrGe}_3$ , a careful determination of the wing near the tricritical point revealed a near tangent merging of the transition lines [10]. In fact, simple considerations using Landau theory of phase transitions revealed that the tangent merging of the wing lines

at the tricritical point is general and should be observed in the other materials as well [41].

To summarize, in Fig. 6, we show the  $T$ - $p$ - $H$  phase diagrams of  $\text{UGe}_2$  and  $\text{LaCrGe}_3$ . These two compounds represent two outcomes of ferromagnetic quantum phase transition. The phase diagram of  $\text{LaCrGe}_3$  is an example of the case when a new magnetic phase appears as the ferromagnetic phase transition is suppressed. It also provides several insights into the phase diagram of other ferromagnetic quantum systems. In  $\text{LaCrGe}_3$  too, a wing structure is observed and, unexpectedly, it is double: the metamagnetic transition proceeds in two steps, which seems to be quite general. Because of the rather high temperature of the tricritical point, a careful investigation confirms certain constraints on the merging of the transition lines at the tricritical point in a wing-structure phase diagram.

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