Quantum Critical Scaling in the Disordered Itinerant Ferromagnet $UCo_{1-x}Fe_xGe$

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The Belitz-Kirkpatrick-Vojta (BKV) theory shows in excellent agreement with experiment that ferromagnetic quantum phase transitions (QPTs) in clean metals are generally first order due to the coupling of the magnetization to electronic soft modes, in contrast to the classical analogue that is an archetypical second-order phase transition. For disordered metals the BKV theory predicts that the second-order nature of the QPT is restored because the electronic soft modes change their nature from ballistic to diffusive. Our low-temperature magnetization study identifies the ferromagnetic QPT in the disordered metal $UCo_{1-x}Fe_xGe$ as the first clear example that exhibits the associated critical exponents predicted by the BKV theory.

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Quantum phase transitions (QPTs) have been a topic of intense research efforts for several decades [1–4]. Here, the earliest theory of a QPT was provided by Stoner in 1938 for itinerant ferromagnets [5]. Because of the exotic behavior frequently observed in the vicinity of ferromagnetic QPTs in metals, such as unconventional spin-triplet superconductivity [6], partial magnetic order [7], and topological non-Fermi liquid behavior [8], their theoretical understanding is at the origin of modern solid state physics. In their seminal work, Hertz and later Millis predicted that ferromagnetic QPTs are continuous, or second order, and calculated the associated critical exponents [9,10]. However, at the turn of the century, an extension of the Hertz-Millis theory by Belitz, Kirkpatrick, and Votja demonstrated in remarkable agreement with experiments that a QPT in two and three dimensions from a paramagnetic to a homogeneous ferromagnetic state is generically discontinuous (or first order) provided that the underlying metal is sufficiently clean [11]. The responsible mechanism is the coupling of the magnetization to electronic soft modes that universally exist in metals, which in turn leads to a fluctuation-induced first-order transition. We note that fluctuation-induced first-order transitions are broadly important in solid-state physics and even beyond [12].

The Belitz-Kirkpatrick-Vojta (BKV) theory further reveals the existence of a tricritical point that separates a line of first-order transitions at low temperature from a line of second-order transitions at higher temperatures when the nonthermal control parameter x that provides access to the QPT is varied. Including the effects of an external magnetic field H in the BKV calculations generates tricritical wings that emerge from the tricritical point as a function of magnetic field [13]. The resulting unique temperature T vs x and H phase diagram has been observed in experiments on many clean ferromagnetic metals, making the BKV theory one of the most successful theories of QPTs [4].

Because in a large number of itinerant ferromagnets the QPT may be accessed via chemical substitution, and some materials show incipient disorder, considering the effect of disorder on the nature of a ferromagnetic QPT is crucial. If the disorder is sufficiently strong, the nature of the electronic soft modes changes from ballistic to diffusive. This slowing down of the itinerant electrons promotes ferromagnetism. According to the BKV theory, this results in the suppression of the tricritical point to zero temperature and the resulting QPT is second order [14–16], with critical exponents that suggest the transition is even more continuous than in the Hertz-Millis theory [4,17]. Although most disordered ferromagnetic metals exhibit second-order QPTs, the critical exponents predicted by the BKV theory (reviewed below) have never been observed consistently.

In this Letter, we demonstrate that the critical behavior observed at a ferromagnetic QPT in UCoGe that is accessed via chemical substitution of Co with Fe is in excellent agreement with the BKV theory. UCoGe orders ferromagnetically below a Curie temperature $T_C = 3$ K and coexists with unconventional superconductivity below $T_S = 0.8$ K [18]. Superconductivity in $UCo_{1-x}Fe_xGe$ is only observed for Fe concentrations $x \leq 0.025$ [19]. In contrast, T_C first increases to the maximum $T_C \approx 9$ K at $x = 0.075$–0.1, and then smoothly decreases to zero temperature at $x_{cr} = 0.23$ consistent with a second-order QPT at x_{cr} [19]. The observed increased values of the residual resistivity $[\rho_0(x_{cr}) \approx 420\ \mu\Omega\,\mathrm{cm}]$ [19] suggest a significant amount of disorder making $UCo_{1-x}Fe_xGe$ an ideal candidate to look for the critical exponents predicted by the BKV theory for disordered metals. In our previous study, the magnetization near the QPT was found to scale as $M(T = 2\ \mathrm{K}, H) \propto H^{1/\delta}$ as a function of H. Here, the corresponding critical exponent was determined to be $\delta \approx 3/2$ in agreement with the BKV theory for a disordered QPT. Because strictly speaking the exponent δ takes on the value associated with

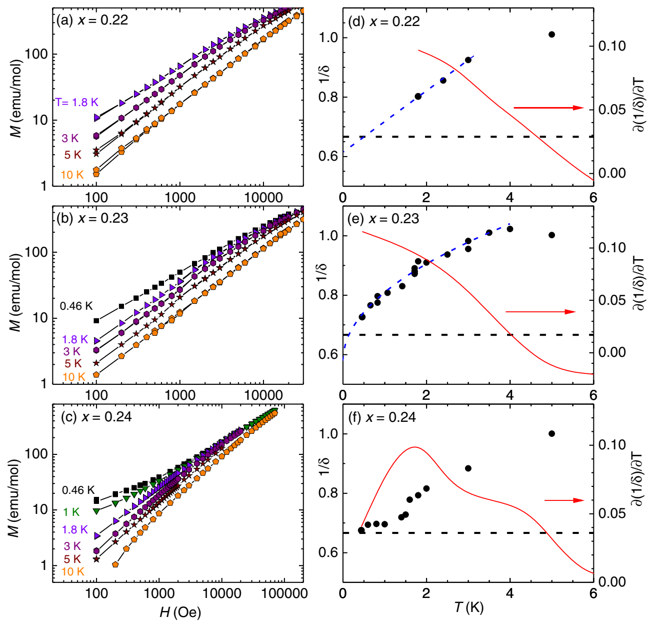

FIG. 1. Isotherms of the magnetization $M(T,H)$ of $UCo_{1-x}Fe_xGe$ as a function of magnetic field H for Fe concentrations $x = 0.22$ (a), 0.23 (b), and 0.24 (c) displayed in a log-log plot. Because $M(T,H) \propto H^{1/\delta}$ the slope of each curves describes $1/\delta$. In panels (d)–(f), the temperature dependence of $1/\delta$ is shown for each concentration. Because for $H \leqq 0.1$ T scaling is not expected due to domain effects, the corresponding data were omitted for the determination of $1/\delta$ [22]. The blue dashed lines in panels (d) and (e) are guides to the eye. The horizontal dashed black line denotes $\delta = 3/2$. The red solid curves are the partial derivative $\partial(1/\delta)/\partial T$ with respect to T.

the QPT only for $T = 0$, this has motivated our present study of $M(T,H)$ down to much lower temperatures. Our results show that near the disordered ferromagnetic QPT in $UCo_{1-x}Fe_xGe$, all critical exponents may be accurately determined from M and agree quantitatively with the BKV theory.

Samples of $UCo_{1-x}Fe_xGe$ were synthesized using a custom built single-arc furnace using a water-cooled copper hearth in an argon atmosphere with a zirconium getter. The starting materials of U (99.9%), Co pieces (99.99%), Fe pieces, and Ge pieces (99.9999 + %) were weighed stoichiometrically, arc melted, flipped over, and remelted five times to ensure chemical homogeneity. Chemical analysis of all samples was carried out using a commercial scanning electron microscope (FEI Inspect F) equipped with an energy dispersive spectroscopy microprobe. The energy dispersive spectroscopy analysis (see the Supplemental Material [20]) shows that they are indeed chemically homogeneous, where in particular the nominal Fe concentration x agrees with the Fe concentration x_{meas} within the error bar. Therefore, we use the nominal Fe concentration x throughout the Letter. We note that polycrystalline samples were chosen purposefully to obtain the most reliable data. Specifically, the magnetic properties of $UCo_{1-x}Fe_xGe$ near the QPT are extremely sensitive to the Fe concentration as shown below. Single crystals of $UCo_{1-x}Fe_xGe$ are grown via the Czochralski method, which typically leads to concentration gradients that would be detrimental for the determination of critical exponents. Finally, it has been demonstrated that the use of polycrystalline samples does not affect the ability to reliably determine scaling exponents of phase transitions [21,22].

All magnetization $M(T,H)$ data presented here were obtained in a Quantum Design magnetic property measurement system with a ^{3}He insert, reaching temperatures T from 300 K down to 460 mK in fields up to 7 T. Figure 1 shows isotherms of the magnetization for various T and $x = 0.22$ (a), 0.23 (b), and 0.24 (c). The data are displayed in a log-log plot so that the slope of each curve corresponds to $1/\delta$. We note that for the determination of $1/\delta$, the data for $H \leqq 0.1$ T, where scaling is not expected because of domain effects, were omitted [22]. The resulting temperature dependence of $1/\delta$ for each of the three concentrations is shown in Figs. 1(d)–1(f). Inspecting Fig. 1(f) for $x = 0.24$, it is clear that $1/\delta$ saturates at $2/3$ for $T \to 0$, in excellent agreement with theory. For $x = 0.22$ and 0.23 no saturation is observed and the value of $1/\delta$ for $T \to 0$ is more challenging to estimate.

Before continuing the discussion of our results, it is useful to recall the critical exponents that can be determined from magnetization data and their values as calculated via the BKV theory for a three-dimensional, ferromagnetic QPT in a metal with significant disorder [4,17]. They are summarized in Table I. Two regimes have to be considered: (a) the so-called asymptotic regime that should only exist in a narrow region near the QPT, and (b) the preasymptotic region that describes the critical exponents further away from the QPT [23]. For δ the asymptotic and preasymptotic values are $3/2$ and $11/6$, respectively. This suggest that $x = 0.24$ is directly in the vicinity of the QPT and thus shows asymptotic behavior. In contrast, for $x = 0.22$ and 0.23 preasymptotic scaling is expected, and indeed provides an excellent description of our data as shown in detail

TABLE I. The critical exponents for a ferromagnetic second-order quantum phase transition for a "dirty" itinerant ferromagnet in three dimensions according to the BKV theory [4,17] are provided for both the (a) asymptotic and (b) preasymptotic regimes. Column (c) denotes the corresponding values for an unstable Hertz type fixed point in three dimensions in the dirty limit.

Critical exponent	(a) Asymptotic	(b) Preasymptotic	(c) Hertz (dirty)
δ	$3/2$	$11/6$	3
β_T	1	$3/4$	$5/8$
γ_T	$1/2$	$5/8$	$5/4$
ν	1	$3/5$	$1/2$
z_m	2	$8/3$	$8/5$

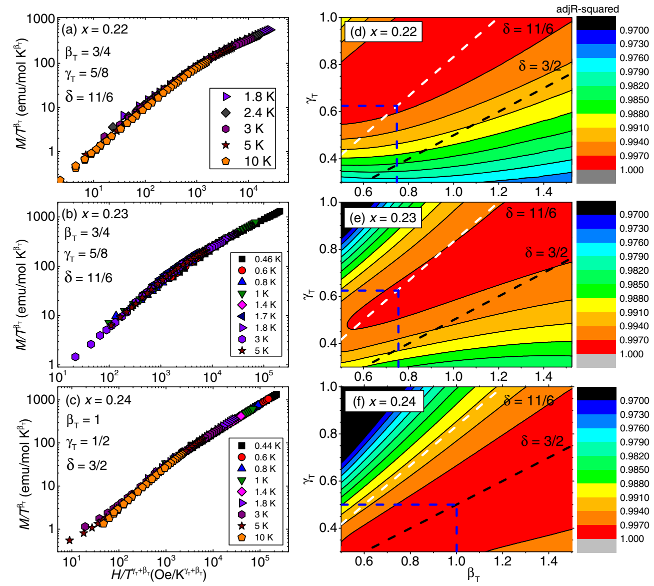

FIG. 2. Scaling of the magnetization M of $UCo_{1-x}Fe_xGe$ as function of magnetic field H and temperature T. (a)–(c) M/T^{β_T} vs $H/T^{\gamma_T+\beta_T}$ for the Fe concentrations $x = 0.22$, 0.23, and 0.24, respectively. The respective critical exponents β_T, γ_T, and δ are denoted in each plot (see the main text). (d)–(f) The corresponding adjusted R^2 value that describes the goodness of fit for the scaling of M in panels (a)–(c) ($R^2 = 1$ is the best agreement between the data and fit) for a wide range of combinations of the critical exponents (β_T, γ_T) for each x. The details of how to calculate the adjusted R^2 value are provided in the Supplemental Material [20]. The black and white dashed lines denote the Widom relationship $\gamma_T = \beta_T(\delta - 1)$ that relates the critical exponents β_T and γ_T with the exponent δ determined from Fig. 1. Here, the black and white lines use the asymptotic and preasymptotic values of δ, respectively (see the text and Table I). The blue dashed lines denote the values of (β_T, γ_T) used for the modified scaling plots of M in panels (a)–(c).

below. The absence of a low-temperature saturation of $1/\delta$ for $x = 0.22$ and 0.23 is explained by the fact that critical scaling is typically only observed to higher temperatures close to the QPT [1]. Therefore, measurements to lower temperatures than accessible in our experiments are required to determine δ unambiguously. Nevertheless, inspection of the partial derivative $\partial(1/\delta)/\partial T$ [Figs. 1(d)–1(f)] demonstrates that the slope of $1/\delta$ is finite for $T \to 0$ [cf. for $x = 0.24$, $\partial(1/\delta)/\partial T \to 0$], suggesting that $1/\delta < 2/3$ in the preasymptotic regime in agreement with the BKV theory.

Because the exponent δ takes on the value associated with the quantum critical point only for $T = 0$, it is crucial to inspect the critical behavior as a function of temperature. Here, the critical exponents β_T and γ_T describe the temperature scaling of $M(T, H)$, where isotherms converge onto a single curve when displayed as $M(T, H)/T^{\beta_T}$ vs $H/T^{\gamma_T+\beta_T}$. Moreover, β_T and γ_T are related to δ via the so-called Widom relationship $\gamma_T = \beta_T(\delta - 1)$ [17].

To confirm our findings for δ we plot the measured isotherms of the magnetization from Figs. 1(a)–1(c) via the scaling relation $M(T, H)/T^{\beta_T}$ vs $H/T^{\gamma_T+\beta_T}$ using the values for β_T and γ_T predicted by the BKV theory for a metal with significant disorder (cf. Table I). Here, according to our analysis for δ, we have used the preasymptotic values for β_T and γ_T for $x = 0.22$ and 0.23, and the asymptotic values for $x = 0.24$. As shown in Fig. 2, the scaling works remarkably well for all three concentrations, especially at low temperatures, thus supporting our results for δ, and highlighting the excellent agreement with the BKV theory.

We note, however, that detailed analysis of the scaling of $M(T, H)$ reveals that the theoretical BKV values of β_T and γ_T are not the only combination that produces good scaling. This is illustrated in Figs. 2(d)–2(f), where we plot the adjusted R^2 value that describes the goodness of fit ($R^2 = 1$ is the best agreement) for a wide range of combinations (β_T, γ_T) (see the Supplemental Material [20] for details). From the small differences in R^2 for the various combinations (β_T, γ_T), it is clear that the magnetization data are not sensitive enough to pick a single set of values. This is also confirmed by visual inspection of the scaling plots of M, where the scaling looks identical for all combinations (β_T, γ_T) that correspond to the red areas in Figs. 2(d)–2(f) (see the Supplemental Material [20]). However, a consistent set of values is obtained when the Widom relationship that also takes into account δ is considered. For reference, we plot the Widom relationship for each concentration in Figs. 2(d)–2(f), where the dashed black and white lines use the asymptotic and preasymptotic value of δ determined from the BKV theory (cf. Table I). From inspection of Figs. 2(d) and 2(e), it is clear that combinations (β_T, γ_T) that lead to the best scaling for $x = 0.22$ and 0.23 generally agree better with the preasymptotic value of δ (i.e., the white dashed line runs through the red area with $R^2 \approx 1$). In contrast, for $x = 0.24$ the asymptotic value of δ gives better agreement as shown in Fig. 2(f) (the black dashed line runs through the red area with $R^2 \approx 1$) in agreement with our results from Fig. 1.

Figure 2(f) also demonstrates that the asymptotic values ($\beta_T = 1$, $\gamma_T = 1/2$) (cf. blue dashed lines) lie well in the center of combinations (β_T, γ_T) that lead to the excellent scaling of $M(T, H)$. This further highlights how well the data for $x = 0.24$ agree with the asymptotic scaling predicted by the BKV theory, and suggest that $x = 0.24$ is the critical Fe concentration, or is at least very near to it. Similarly, as shown in Figs. 2(d) and 2(e) for $x = 0.22$ and 0.23, the preasymptotic values ($\beta_T = 3/4$, $\gamma_T = 5/8$) (cf. blue dashed lines) belong to the combinations of (β_T, γ_T) that converge all isotherms of M onto a single curve.

The product of two additional critical exponents ν and z_m can be determined from M. Here, z_m is the relevant dynamical exponent [17]. Together, they describe how T_C is suppressed as a function of the tuning parameter x via $T_C = (x - x_{\mathrm{cr}})^{z_m \nu}$. The theoretical asymptotic and

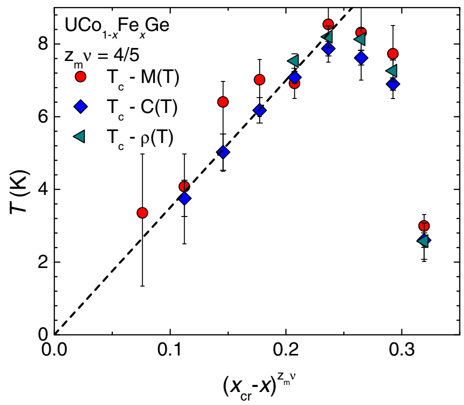

FIG. 3. The Curie temperature T_C of $UCo_{1-x}Fe_xGe$ as a function of $(x - x_{cr})$. Here, x is the Fe concentration and x_{cr} denotes the concentration at which T_C is suppressed to zero temperature. The black line is a fit to $T_C = (x - x_{cr})^{z_m \nu}$ with $z_m \nu = 4/5$ for $x_{cr} = 0.24$. The red circles, blue diamonds, and green triangles denote T_C determined from the magnetization $M(T)$, specific heat $C(T)$, and electrical resistivity $\rho(T)$ from Ref. [19].

preasymptotic values for $z_m \nu$ are 2 and 8/5, respectively (Table I). According to the BKV theory, and also in agreement with our data that only show asymptotic scaling for $x = 0.24$, the asymptotic regime typically only exists in a very narrow region around the QPT. Further, as demonstrated above for $x \leq 0.23$, the magnetization exhibits preasymptotic scaling, suggesting that $T_C(x)$ should scale with $z_m \nu = 8/5$. In contrast, we find that $T_C(x)$ as determined in Ref. [19] scales well with $z_m \nu = 4/5$, as we show in Fig. 3. $z_m \nu = 4/5$ is consistent with a Hertz type ferromagnetic QPT in the dirty limit (cf. Table I). Despite the fact that a Hertz fixed point is unstable, it is expected from the BKV theory that it will determine the observable behavior over sizable regions of the phase diagram in many disordered materials [17]. Only in a close vicinity of the QPT is it expected to observe the (pre) asymptotic behavior. However, because the phase transition rapidly becomes extremely broad for $x \to x_{cr}$, the scaling of $T_C(x)$ near x_{cr} cannot be precisely determined.

To summarize, the salient findings of our study are as follows: (i) the critical exponent δ defined via $M(T \to 0, H) \propto H^{1/\delta}$ is equal to 3/2 for the critical Fe concentration $x_{cr} = 0.24$ and is significantly smaller than 3/2 for $x = 0.22$ and 0.23 slightly away from the QPT; (ii) our data are in excellent agreement with the critical exponents β_T and γ_T that describe the temperature scaling of $M(T, H)$ being 1 and 1/2 for $x = 0.24$, and 3/4 and 5/8 for $x = 0.22$ and 0.23, respectively; (iii) our results are consistent with the QPT being at or in the very near vicinity of $x_{cr} = 0.24$; (iv) the asymptotic and preasymptotic regions are situated at the Fe concentration $x > 0.23$ and $x \leq 0.23$, respectively; (v) T_C is found to scale as $T_C = (x - x_{cr})^{z_m \nu}$ for a wide range of x with $z_m \nu = 4/5$ consistent with a Hertz fixed point and in agreement with the BKV theory. This establishes that $UCo_{1-x}Fe_xGe$ is the first metal that exhibits critical behavior near a ferromagnetic QPT accessed by chemical substitution that is in complete agreement with the predictions of the BKV theory for a metal with significant disorder.

For completeness, we note that a critical exponent $\delta = 3/2$ has already been observed in $URu_{2-x}Re_xSi_2$ [24] and $Sr_{1-x}A_xRuO_3$ [25] at $T = 1.8$ K and $T = 5$ K, respectively. However, these results were obtained at finite temperature, whereas the result of the BKV theory is only valid for $T \to 0$ and in the immediate vicinity of the QPT. Notably, for $URu_{2-x}Re_xSi_2$, the QPT is situated at $x_c = 0.15$–0.2 but $\delta = 3/2$ was obtained for $x = 0.3$ [24,26]. Finally, all other exponents β_T, γ_T, and $z_m \nu$ either do not agree with the BKV theory [24,26] or were not determined.

In conclusion, our results demonstrate that metals with significant disorder exhibit second-order ferromagnetic QPTs that are more continuous than in the Hertz-Millis theory. This establishes for the first time that the BKV theory not only describes clean materials extremely well, but is also able to calculate the critical exponents for disordered metals. Our work also identifies several reasons why no other disordered materials have been reported to show BKV critical exponents. Most notably, it is difficult to achieve the right amount of disorder to observe such behavior; for too small disorder, the tricritical point remains at nonzero temperatures and no quantum critical behavior occurs. In contrast, for too strong disorder, quantum Griffiths effects that compete with the critical behavior are expected [4]. Moreover, there is currently no clear method of determining disorder quantitatively to allow for comparison with theory, and choosing a suitable material is challenging. For example, the often used residual resistivity ratio is only useful in comparing the disorder for different specimens of the same material, and is not meaningful for distinct materials. Further, as shown in detail here, special diligence is required to obtain meaningful critical exponents, as typical magnetization measurements are too insensitive to choose a single set of critical exponents (see the Supplemental Material [20]). Finally, for $UCo_{1-x}Fe_xGe$, we have shown that the asymptotic scaling is only observed in a tiny region around the QPT [23], which implies that great care is required to identify this region. However, our study may provide a recipe for identifying further disordered metals that exhibit BKV critical exponents near a ferromagnetic QPT.

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