Quantum Griffiths Phase Inside the Ferromagnetic Phase of $Ni_{1-x}V_x$

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We study by means of bulk and local probes the d-metal alloy $Ni_{1-x}V_x$ close to the quantum critical concentration, $x_c \approx 11.6\%$, where the ferromagnetic transition temperature vanishes. The magnetization-field curve in the ferromagnetic phase takes an anomalous power-law form with a nonuniversal exponent that is strongly x dependent and mirrors the behavior in the paramagnetic phase. Muon spin rotation experiments demonstrate inhomogeneous magnetic order and indicate the presence of dynamic fluctuating magnetic clusters. These results provide strong evidence for a quantum Griffiths phase on the ferromagnetic side of the quantum phase transition.

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Quantum phase transitions (QPTs) [1] continue to be a central topic in condensed-matter physics because they are responsible for a variety of unconventional low-temperature phenomena. For example, the spin fluctuations associated with QPTs between magnetic and nonmagnetic ground states can lead to non-Fermi liquid behavior or even induce novel phases of matter [2].

Real materials always contain some disorder in the form of vacancies, impurities, and other defects. In particular, disorder is unavoidable if the QPT is tuned by varying the composition x in a random alloy such as $Ni_{1-x}Pd_x$, $CePd_{1-x}Rh_x$, or $Sr_{1-x}Ca_xRuO_3$. Research has shown that disorder can dramatically change a QPT and induce a quantum Griffiths phase, a parameter region close to the transition point that is characterized by anomalous thermodynamic behavior. This was established for model Hamiltonians [3,4] and later predicted to occur in itinerant magnets [5,6], superconductors [7,8], and other systems (for reviews see, e.g., Refs. [9]).

Signatures of a magnetic quantum Griffiths phase have been observed, e.g., in diluted Ce compounds [10] and, perhaps most convincingly, in the paramagnetic phase of the d-metal alloy $Ni_{1-x}V_x$ [11,12]. They consist in anomalous nonuniversal power-law dependencies of the magnetization, susceptibility, and other thermodynamic quantities on temperature and magnetic field for concentrations x close to but above the quantum critical concentration x_c (where the ferromagnetic transition temperature is suppressed to zero). These quantum Griffiths singularities can be attributed to rare magnetic regions embedded in the paramagnetic bulk, as predicted in the infinite-randomness scenario for disordered itinerant Heisenberg magnets [6,7].

Do such Griffiths singularities also exist inside the long-range ordered, ferromagnetic phase? Theoretical arguments [13,14] suggest that rare isolated magnetic clusters produce anomalous thermodynamic behavior on the ferromagnetic side of the QPT as well as on the paramagnetic side. However, the resulting quantum Griffiths singularities are less universal; depending on the details of the underlying disorder, they range from being stronger than the paramagnetic ones to being much weaker. So far, clear-cut experimental observations of a quantum Griffiths phase inside the long-range ordered phase have been missing [15] (see Ref. [18] for a comprehensive review of QPTs in metallic ferromagnets).

In this Letter, we report the results of magnetic measurements and muon spin rotation (μSR) experiments in $Ni_{1-x}V_x$ across the ferromagnetic QPT. Close to the critical concentration $x_c \approx 11.6\%$, the dependence of the low-temperature magnetization M on the magnetic field H is well described by anomalous power laws on both sides of the transition. On the paramagnetic side, $M \sim H^\alpha$ as in earlier work [11,12]. On the ferromagnetic side, we observe $M - M_0 \sim H^\alpha$ where M_0 is the spontaneous magnetization. The exponent α is strongly x dependent (i.e., nonuniversal) and decreases towards zero at x_c. Strikingly, its x dependence is almost symmetric in $x - x_c$. μSR measures the local magnetic fields inside the sample and reveals the microscopic origins of this anomalous behavior. In the ferromagnetic phase we find a broad distribution of local magnetic fields signifying inhomogeneous magnetic order. μSR data for samples close to x_c also indicate that fluctuating magnetic clusters coexist with the long-range ordered bulk. These results provide strong evidence for a quantum Griffiths phase on the ferromagnetic side of the QPT in $Ni_{1-x}V_x$.

Polycrystalline spherical samples of $Ni_{1-x}V_x$ with $x = 0\%$ to 15% were prepared and characterized as described in Refs. [11,19]. A pair-distribution function

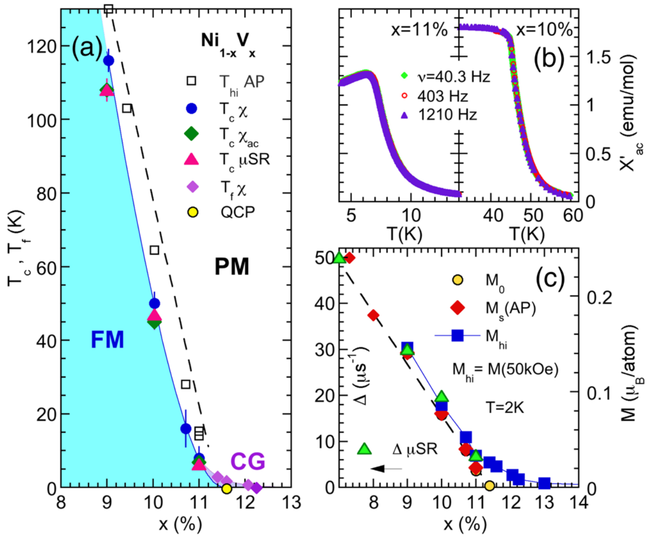

FIG. 1. (a) Phase diagram of $Ni_{1-x}V_x$ showing paramagnetic (PM), ferromagnetic (FM), and cluster glass (CG) phases. The ferromagnetic transition temperature T_c is found using three different methods (see text), leading to a quantum critical point (QCP) at $x_c = 11.6\%$. The high-field (Arrott plot) estimate $T_{\rm hi}$ of the transition shows a linear x dependence (dashed line). (b) The ac susceptibility $\chi'_{\rm ac}$ vs. temperature T (absolute scale estimated by dc-M). T_c is marked by a cusp independent of frequency ν. (c) Zero-field moment M_s (from Arrott plots), M_0 [from $M(H)$ power law]. and μSR field distribution width Δ show linear x dependencies (dashed line). $M_{\rm hi}$ is the magnetization in a field of $H = 50$ kOe. Data of $T_{\rm hi}$ and M_s from [33,34] are included.

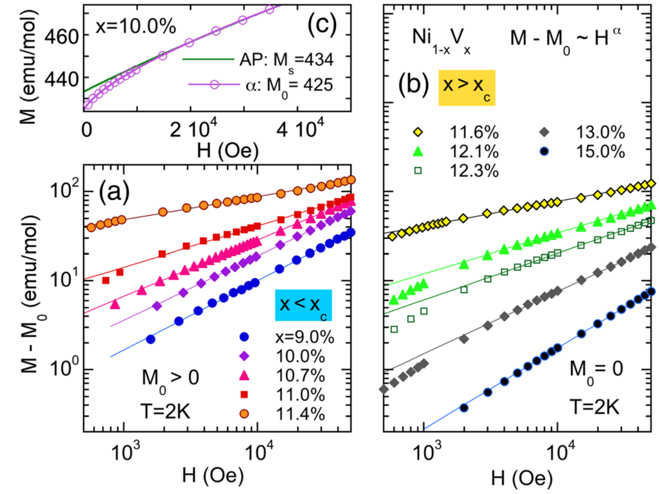

FIG. 2. Magnetization M vs (internal) magnetic field H for several compositions x at the lowest $T = 2$ K. An offset $M_0(x) > 0$ has been subtracted for $x < x_c$ in (a); $M_0 = 0$ for $x \geq x_c$ in (b). Solid lines represent fits to $M(H) = M_0 + d_\alpha H^\alpha$. (c) M vs H for $x = 10\%$ with power-law fit defining M_0 and Arrott plot (AP) fit defining M_s.

analysis supports the random distribution of the V atoms. Details of the sample preparation, the characterization with neutron scattering, and the magnetization and μSR measurements (performed at the Paul Scherrer Institut and the ISIS Neutron and Muon Source) are summarized in the Supplemental Material [20].

At first glance, $Ni_{1-x}V_x$ features a simple phase diagram: The ferromagnetic ordering temperature T_c and the spontaneous magnetization M_s are linearly suppressed with increasing x and vanish between $x = 11\%$ and 12%, as shown in Figs. 1(a) and 1(c). This critical concentration is much smaller than the corresponding $x_c = 97.5\%$ for $Ni_{1-x}Pd_x$ [29] because the V atoms, with 5 fewer d electrons than Ni, also suppress the spins of their Ni neighbors and thus create large defects [30,31]. The inhomogeneous suppression of magnetic order causes deviations from the linear x dependence of T_c close to the critical concentration. We determined T_c from the maximum of the susceptibility $dM/dH(T, H \to 0)$ [12], the cusp in the ac susceptibility $\chi'_{\rm ac}(T, H = 0)$ [32] [see Fig. 1(b)], and the onset of the zero-field μSR amplitude $A_{\rm FM}(T)$ [19] (see Fig. 4 below). All estimates agree well with each other. The resulting $T_c(x)$ curve develops a tail and follows the prediction [7] of the infinite-randomness scenario, giving a critical concentration $x_c = 11.6\%$. (In contrast, the tail is absent when an ordering temperature $T_{\rm hi}$ is estimated via extrapolation from high fields, e.g., via standard Arrott plots of H/M vs M^2.)

The actual quantum critical point at $T = 0$ and $x = x_c$ is masked by a cluster glass phase that appears for $x \gtrsim 11.4\%$ below a freezing temperature $T_f \leq 3$ K, see Fig. 1(a) [11,32]. It is rapidly suppressed by small dc fields and does not affect the physics considered in this Letter.

We now analyze the field dependence of the magnetization M at low T. Figure 2 shows M vs H at $T = 2$ K for V concentrations x on both sides of the QPT. For paramagnetic samples ($x \geq x_c = 11.6\%$), the magnetization follows the anomalous power law $M(H) = d_\alpha H^\alpha$ over an extended field range from about 2 kOe to the highest available field of 50 kOe. Interestingly, the field dependence of the magnetization in the long-range ordered ferromagnetic phase ($x < x_c$) is also well described by a power-law form, viz., $M(H) = M_0 + d_\alpha H^\alpha$, where M_0 represents the nonzero spontaneous magnetization. As in the paramagnetic phase, these power laws hold in a wide field range from about 1 or 2 kOe to 50 kOe [while the conventional Arrott plot description breaks down below about 10 kOe, see Fig. 2(c)].

The exponent α is nonuniversal, i.e., strongly x dependent. It has a minimum close to the critical concentration x_c and increases monotonically towards the linear-response value $\alpha = 1$ with increasing distance from x_c, as shown in Fig. 3(b). Strikingly, the $\alpha(x)$ curve is nearly symmetric in $x - x_c$. It can be fitted with a power law, $\alpha(x) \sim |x - x_c|^{\nu\psi}$ with exponent $\nu\psi \approx 0.34 \pm 0.08$ [35], confirming $x_c = 11.6\% \pm 0.1\%$.

What is the origin of these unusual magnetization-field curves? In the paramagnetic phase, they can be attributed to magnetic clusters that are embedded in the paramagnetic bulk [11,12]. These clusters exist on rare Ni-rich regions in the sample. Their slow independent fluctuations lead to

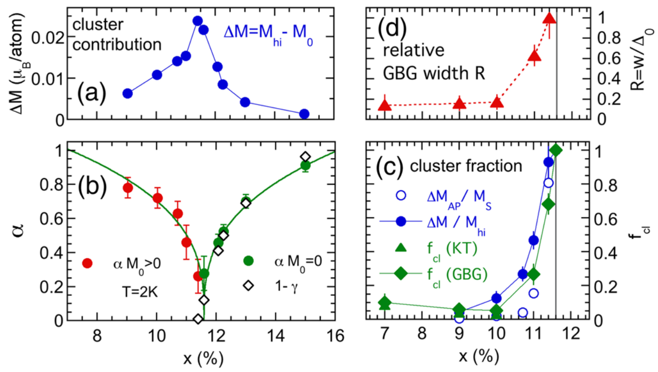

FIG. 3. (a) Cluster contribution $\Delta M = M_{\rm hi} - M_0$ with $M_{\rm hi} = M(50\ {\rm kOe})$ vs concentration x in $Ni_{1-x}V_x$. (b) Nonuniversal exponent α vs x, and susceptibility exponents γ from Ref. [11]. Lines are universal power-law fits $\alpha(x) \sim |x - x_c|^{\nu\psi}$. (c) Cluster fraction $f_{\rm cl}$ vs x from different methods. (d) Relative width $R = w/\Delta_c$ vs x of the Gaussian-broadened Gaussian (GBG) used in the μSR analysis. Data evaluated at lowest temperature (1.5–2 K for $x > 10\%$).

anomalous power laws, the Griffiths singularities, in the temperature and field dependencies of various thermodynamic quantities [9]. Deviations at the lowest fields and temperatures stem from weak interactions between the rare regions that freeze their dynamics [11,36,37]. Our observation of anomalous magnetization-field curves below the critical concentration x_c indicates that disconnected magnetic clusters that fluctuate independently from the long-range ordered bulk also play a crucial role inside the ferromagnetic phase.

To analyze the importance of these clusters quantitatively, we estimate their contribution to the magnetization. A conservative estimate can be obtained by comparing the spontaneous magnetization M_0 with the zero-field magnetization M_s obtained via Arrott plot extrapolation from high fields [see Fig. 1(c)]. As the clusters are disconnected from the bulk, they do not contribute to M_0. In high fields they are fully polarized, however, and thus included in M_s. Consequently, $\Delta M_{\rm AP} = M_s - M_0$ measures the cluster contribution to M. Alternatively, one could simply evaluate $\Delta M = M_{\rm hi} - M_0$ with $M_{\rm hi} = M(H = 50\ {\rm kOe})$ and define the cluster fraction as $\Delta M / M_{\rm hi}$ [39]. The x dependence of ΔM is shown in Fig. 3(a). ΔM has a maximum close to x_c and decreases for $x > x_c$ because the total number of magnetic Ni atoms decreases. ΔM also decreases for $x < x_c$ because it becomes less likely that a magnetic cluster remains disconnected from the bulk. By comparing ΔM with the typical cluster moment of 12 μ_B [11,12], we estimate a cluster density at x_c of about 1 cluster per 500 Ni atoms. Figure 3(c) presents the cluster fractions $\Delta M_{\rm AP}/M_s$ and $\Delta M / M_{\rm hi}$ as functions of x. The measures track each other and indicate that clusters become relevant for $x > 10\%$.

To gain microscopic insight into these clusters and their dynamics, we employ μSR experiments (see, e.g., Ref. [40] for an introduction and Ref. [41] for a technical review). In this technique, spin-polarized positive muons are implanted in the sample. Their spins then precess in the local magnetic field at the stopping site until the muon decays, with a positron emitted preferentially in the direction of the muon spin. Analyzing the asymmetry $A(t)$ of the positron emission as a function of time thus gives direct access to the distribution of local magnetic fields in the sample. μSR played an important role in characterizing unconventional magnetism, e.g., in heavy-fermion compounds [42], spin glasses [43], and disordered, non-Fermi liquid metals [44]. As μSR experiments are sensitive towards small magnetic moments, spatial inhomogeneities, and slow fluctuations, they are well suited to identify and study magnetic clusters.

Data for the muon asymmetry $A(t)$ in zero magnetic field for several samples from $x = 0\%$ to 12.3% are presented in the Supplemental Material [20], together with further details of the analysis. For pure Ni ($x = 0$), $A(t)$ features a single (nearly undamped) precession frequency confirming a uniform local magnetic field and thus uniform ferromagnetic order. In contrast, the $x = 12.3\%$ sample on the paramagnetic side of the QPT shows a very weak depolarization. It can be described by a simple exponential decay, $A(t) = A_{\rm PM} P_{\rm PM}(t) = A_{\rm PM} \exp(-\lambda t)$, caused by quasistatic diluted V nuclear spins as well as by fluctuating Ni clusters in the extreme motional narrowing limit.

Here, we focus on two samples ($x = 10\%$ and 11%) that are close to the QPT but on its ferromagnetic side. At low temperatures, $A(t)$ of the $x = 10\%$ sample [shown in Fig. 4(a)] features a single dip but no further oscillations. Analogous behavior is observed for $7\% \leq x \leq 10\%$ [19]. It can be described by a Gaussian distribution of local magnetic fields of width Δ, leading to $A(t) = A_{\rm FM} P_{\rm FM}(t)$ with $P_{\rm FM}(t) = P_{\rm KT}(t; \Delta)$, where $P_{\rm KT}(t; \Delta)$ is the well-known Kubo-Toyabe depolarization function [45]. At temperatures below about $0.5 T_c$, the data follow the static KT form, signifying a moderately inhomogeneous, long-range ordered state.

Over the entire temperature range, $A(t)$ can be modeled by two components (with temperature-dependent amplitudes) and a small constant background term,

$$A(t) = A_{\rm PM}(T) P_{\rm PM}(t) + A_{\rm FM}(T) P_{\rm FM}(t) + A_{\rm BG}. \quad (1)$$

The temperature dependence of the relative amplitude $f_{\rm FM} = A_{\rm FM}/(A_{\rm FM} + A_{\rm PM})$, which represents the FM fraction of the sample, is presented in Fig. 4(c). It rapidly increases as the temperature is lowered below T_c and reaches values close to unity for $T \leq 0.7 T_c$. The width Δ of the local magnetic field distribution increases with decreasing T; below about $0.7 T_c$, $\Delta \propto M_0$ as shown in Fig. 4(d).

For $x = 11\%$, the KT form fails to describe $A(t)$ [shown in Fig. 4(d)] as the typical dip is missing; data taken in longitudinal fields also exclude a dynamic KT form [19]. A nearly static broader-than-Gaussian field distribution can

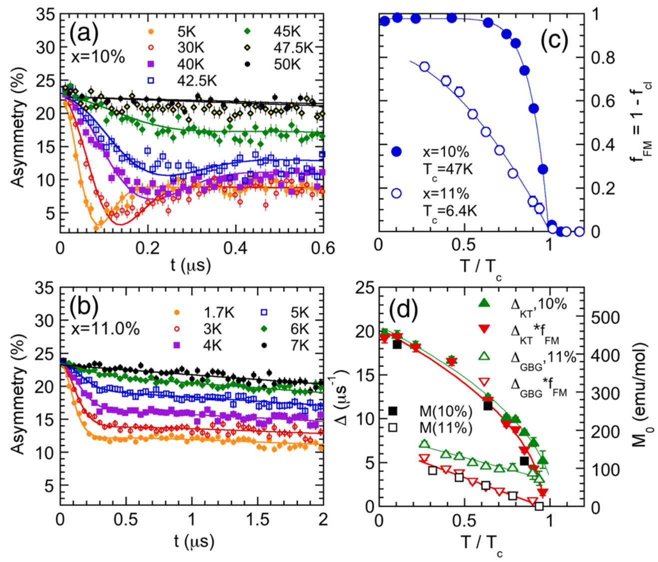

FIG. 4. (a),(b) μSR asymmetry A vs time t for different concentrations x and temperatures T (collected at the DOLLY instrument). Lines represent fits to Eq. (1) using different $P_{\mathrm{FM}}(t)$: (a) Kubo-Toyabe (KT) form for $x = 10\%$, (b) GBG form for $x = 11\%$ (for details see text). (c) Ferromagnetic fraction (amplitude ratio) f_{FM} vs temperature T. (d) Field distribution width $\Delta = \Delta_{\mathrm{KT}}$ for $x = 10\%$ and $\Delta = \Delta_{\mathrm{GBG}}$ for $x = 11\%$ in frequency units $\Delta = \gamma_\mu \langle B_{\mathrm{loc}}^2 \rangle^{1/2}$ (with $\gamma_\mu = 2\pi \times 135.5$ MHz/T). The magnetization M_0 and Δ are proportional to each other (with $M_0/\Delta \approx 23$ emu/mol MHz), but only if Δ is scaled by f_{FM}.

account for the main, fast time dependence of $A(t)$. In fact, $A(t)$ can be fitted well using Eq. (1) with $P_{\mathrm{FM}} = P_{\mathrm{GBG}}(t; \Delta_0, w)$ where P_{GBG} is the static Gaussian-broadened Gaussian (GBG) depolarization function suggested in Ref. [46], and Δ_0 and w are the average and width of the Gaussian of Gaussians. The temperature dependencies of the effective distribution width $\Delta_{\mathrm{GBG}} = (\Delta_0^2 + w^2)^{1/2}$ and of the relative amplitude f_{FM} are shown in Figs. 4(d) and 4(c). The need for a broad field distribution to describe the ferromagnetic component indicates strongly inhomogeneous order. Moreover, the ferromagnetic ratio f_{FM} increases only slowly below T_c, and a sizable paramagnetic contribution representing about 20% of the sample volume remains even at the lowest T. This paramagnetic contribution stems from the fluctuating moments of Ni-rich clusters that are disconnected from the long-range ordered bulk.

The cluster fraction $f_{\mathrm{cl}} = 1 - f_{\mathrm{FM}}$ can be obtained for all x using KT and GBG fits of $A(t)$ at the lowest T. As shown in Fig. 3(c), these μSR-based cluster fractions agree well with the estimates from the magnetization data and indicate that clusters are relevant for $x > 10\%$. Accordingly, the relative width $R = w/\Delta_0$ of the Gaussian of Gaussians [46] in the field distribution starts increasing for $x > 10\%$, as shown in Fig. 3(d).

In summary, we studied the d metal alloy $\mathrm{Ni}_{1-x}\mathrm{V}_x$ close to its quantum-critical concentration x_c, focusing on the ferromagnetic side of the QPT. We found that the low-temperature magnetization-field curve in the ferromagnetic phase follows the power law $M(H) = M_0 + d_\alpha H^\alpha$ in analogy to the power-law Griffiths singularity $M(H) \sim H^\alpha$ on the paramagnetic side. This anomalous behavior can be attributed to magnetic clusters existing on disconnected rare Ni-rich regions of the sample. Further evidence for such clusters comes from μSR experiments that reveal strongly inhomogeneous magnetic order and the presence of paramagnetic, fluctuating moments inside the long-range ordered ferromagnet (for samples sufficiently close to x_c). These results provide evidence for a quantum Griffiths phase inside the ferromagnetic phase and demonstrate that QPTs in strongly disordered systems are qualitatively different not just from their clean counterparts but also from disordered classical phase transitions. Disorder at a classical transition may change its universality class or turn a first-order transition continuous. In contrast, we observed much stronger effects. Thermodynamic and other properties of $\mathrm{Ni}_{1-x}\mathrm{V}_x$ close to its QPT are dominated by rare events, resulting, for example, in a diverging magnetic susceptibility not just at x_c but over a range of x close to x_c.

In theoretical studies of model Hamiltonians [13,14], quantum Griffiths phases on the magnetic side of the QPT are much less universal than those on the paramagnetic side. This stems from the fact that the probability of finding a magnetic cluster that is disconnected from the long-range ordered bulk of the system depends on the details of the disorder. Specifically, in a percolation scenario, a magnetic cluster can be isolated by a surface (shell) of nonmagnetic sites (or broken bonds). Such events have a comparatively high probability; the resulting Griffiths singularities on the ferromagnetic side are thus expected to be stronger than power laws, i.e., stronger than their paramagnetic analogs [13]. For weak disorder, in contrast, a cluster has be far away from the long-range ordered bulk to be isolated. This reduces the cluster probability and leads to ferromagnetic Griffiths singularities that are weaker than the power laws on the paramagnetic side [14]. The disorder in $\mathrm{Ni}_{1-x}\mathrm{V}_x$ is not purely percolational because the material is a metal, but it is rather strong because each V atom creates a large local defect. The strength of the quantum Griffiths singularities is therefore expected to be between the above limiting cases, in agreement with our observations. However, the existing theories cannot explain the striking symmetry in $x - x_c$ of the Griffiths singularities found here [47]. This remains a challenge for future work.

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