Magnetization of Pd$_{1-x}$Ni$_x$ near Quantum Criticality

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Abstract. Ni-doped Pd exhibits a paramagnetic to ferromagnetic quantum phase transition at $x_c \approx 2.5\%$. We report the magnetic field dependence of the magnetization of Pd$_{1-x}$Ni$_x$ for $x = 2.59\%$ at various temperatures. The nonlinear magnetization is well described by a conventional mean-field behavior at high temperatures and high magnetic fields. At low fields and low temperatures the magnetic field dependence deviates markedly from mean-field behavior suggesting a more complex magnetic state. Our study questions recent claims that Pd$_{1-x}$Ni$_x$ exhibits a ferromagnetic quantum criticality akin a pure itinerant ferromagnet.

1. Introduction

The non-linear dependence of an order parameter on its conjugate variable intimately reflects the interactions driving the emergence of the ordered state. The perhaps simplest example for illustrating this rather general statement is ferromagnetism, where the order parameter is the magnetization $M$ and the magnetic field dependence $M(B)$ represents a deep probe of the interactions that connect the magnetization with the susceptibility of the system.

In the simplest scenario the non-linear magnetization may be accounted for by a magnetic equation of state derived from a fourth order Ginzburg Landau free energy $F = aM^2 + bM^4 + \cdots$ (not writing explicitly the usual gradient term). The magnetic field $B$ which stabilizes the magnetization $M$ is then expected to vary with $M$ as follows

$$B = \frac{1}{V} \frac{\partial F}{\partial M} = aM + bM^3 + \cdots$$

where $a$ and $b$ are material specific phenomenological parameters. $a$ represents the inverse linear susceptibility, which limits for $T \to 0$ to the so-called inverse initial susceptibility $a_0 = a(T \to 0)$. The parameter $b$ represents the lowest approximation of the effects of mode-mode coupling.

In recent years so-called quantum phase transitions (QPT), representing a new class of phase transitions are attracting great interest. In contrast to entropy-driven thermal phase transitions QPT are driven by quantum fluctuations and result from a competition of dominant contributions in the internal energy. An important question concerns whether a given QPT is first or second order and whether the underlying dynamical properties are those of a pure or a disordered compound. For quantum critical transitions, a comprehensive theoretical framework has been developed. The effects of the quantum critical fluctuations in many cases may be
Figure 1. (A) Magnetization of Pd$_{1-x}$Ni$_x$ for $x = 2.59\%$ as a function of magnetic field up at various temperatures. (B) Arrott plots of the data shown in panel (A); for large fields mean field behavior is observed, i.e., to the right hand side of the arrows, where the non-linear magnetization is well-accounted for by a simple cubic term.

captured by an effective mean-field theory, when the sum of dynamical exponent $z$ and order parameter dimension $d$ exceeds the critical dimension $d_{eff} = d + z \geq 4$.

A prototypical example for a QPT and the perhaps best studied case concerns the border of itinerant ferromagnetism in three-dimensional systems. For clean systems $d = 3$ and $z = 3$ implies $d_{eff} = 6$, i.e., mean field behavior is expected [1–4]. At first sight one may expect a large number of systems to display ferromagnetic quantum criticality. However, upon closer inspection essentially all stoichiometric systems studied to date appear to display a first order ferromagnetic to paramagnetic QPT or the QCP is preempted by some other phase [5]. Examples include ZrZn$_2$ [6], Ni$_3$Al [7], CoS$_2$, UGe$_2$ [8] and NbFe$_2$ [9]. In essentially all of these systems the ferromagnetic QPT is reached through the application of hydrostatic pressure, making comprehensive studies encompassing many different experimental properties inherently difficult.

A system in which a ferromagnetic quantum phase transition may be reached as a function of doping is Pd$_{1-x}$Ni$_x$ [10]. As a function of Ni concentration ferromagnetic order emerges in Pd$_{1-x}$Ni$_x$ for $x > 2.5\%$. The temperature dependence of the specific heat, resistivity and susceptibility suggest the properties of a clean ferromagnetic quantum critical point. We have revisited this issue using complementary measurements of the magnetization as a function of magnetic field, thereby directly investigating the order parameter.

2. Experimental Technique

Our Pd$_{1-x}$Ni$_x$ sample was grown by a Czochralsky technique, starting from high purity elements with a nominal composition of $x = 2.59\%$ [11]. The ingot was characterized by Laue x-ray diffraction and on the single-crystal diffractometer RESI at FRM II, which both established a single-crystalline state. Neutron depolarization radiography established a range of Curie temperatures of several degree K across the ingot [12]. Several small specimens were cut from the ingot for measurements of the resistivity, specific heat and magnetization. The resistivity and specific heat were perfectly consistent with the literature. This strongly suggests that the behavior reported in the following is typical of Pd$_{1-x}$Ni$_x$ at low values of $x$. The magnetization was measured with an Oxford Instruments vibrating sample magnetometer, permitting studies at temperatures as low as 4 K and magnetic fields as high as 9 T.
Figure 2. Information inferred from the data shown in Fig. 1. (A) Inverse susceptibility inferred from the Arrott plots, where the red circles are taken in mean field regime, i.e., the high temperature/high field regime. Black data points are inferred from the low field behavior. (B) Ordered moment inferred from the intercept of the Arrott plots for $B \to 0$. A small ordered moment is observed, where the transition temperature corresponds with that inferred from panel (A). (C) Temperature dependence of the lowest level approximation of the mode-mode coupling parameter $B$. In the limit of the mean-field behavior (red data points) a weak decrease of $b$ with increasing temperature is observed. In the low-field limit (black data points) the mode coupling appears to be anomalously increased anomalously.

3. Results

Shown in Fig. 1(A) is the magnetization as a function of magnetic field up to 9 T at temperatures in the range 4 K to 60 K. With decreasing temperature a spontaneous ordered moment appears in the limit $B \to 0$ for temperatures below $\sim 10$ K. At all temperatures down to 4 K the magnetization increases as a function of magnetic field without any sign of saturation. Likewise a considerable non-linear field dependence may be seen at all temperatures. The tiny hysteresis between increasing and decreasing magnetic fields (not visible on the scales chosen here) may be traced to a small offset in the field measurement.

To better explore the nature of the non-linear field dependence we show in Fig. 1(B) the inverse DC susceptibility $B/M$ as a function of $M^2$ (the inverse plot is also known as Arrott plot; we refer to plots of $B/M$ vs $M^2$ as Arrott plots). At high temperatures a straight line is observed for all fields (and thus values of $M$). This is characteristic of a conventional mean field relationship between the susceptibility and the magnetization. As the temperature decreases the mean field behavior survives for sufficiently large values of $M$, notably to the right hand side of the arrows at high fields. The arrows hence mark the location of a cross-over, where we find for low values of $M$ and $T$ a behavior that is more complex. Notably the Arrott plots display negative curvature suggesting that the inverse susceptibility is lower than the corresponding static magnetization; equivalently the susceptibility for a given magnetization is larger than expected in a mean-field approximation. Correspondingly the curvature and thus the mode-mode coupling in leading order is stronger than predicted by the usual mean-field dependence.
4. Discussion and Conclusions

The anomalous behavior we observe in the Arrott plots for Pd$_{1-x}$Ni$_x$ near the quantum critical Ni concentration is summarized in Fig. 2. Shown in panel (A) is the inverse susceptibility as a function of temperature. The behavior at low fields and low temperatures is shown by black data points; this is the zero field extrapolation on the left hand side of the arrows in Fig. 1 (B). A pronounced Curie-Weiss dependence is observed with a large fluctuating moment of $\sim 1 \mu_B$. The susceptibility provides clear evidence of a ferromagnetic transition at $T_C \approx 11$ K. In contrast, when plotting the inverse susceptibility inferred from the Arrott plots in the regime of mean-field behavior, i.e., the susceptibility extrapolated for zero from high fields (left hand side of arrows in Fig. 1 (B)), a strong Curie Weiss dependence is only seen at high temperatures. At low temperatures these data would imply that the sample remains paramagnetic.

The properties suggested by the inverse susceptibility shown in Fig. 2 (A) are strongly supported by the ordered magnetic moment $M_s$ inferred from the Arrott plots as the value of $M$ for $B \to 0$ (Fig. 2 (B)). The ordered moment vanishes at a Curie temperature $T_C \approx 11$ K. As a final point to illustrate the increased curvature shown in Fig. 1 (B) we plot in Fig. 2 (C) an estimate of the initial slope of the Arrott plots for small fields. The mode-mode coupling parameter appears to be anomalously large when approaching low temperatures (black data points).

Our measurements of the magnetic field dependence of the magnetization clearly reveal a regime at low fields and low temperatures, where strong deviations emerge from the conventional mean field predictions of a ferromagnetic quantum critical point in the clean limit. However, quantum criticality of a clean system has been claimed based on the temperature dependence of the specific heat, resistivity and susceptibility [10]. Because our samples show temperature dependences of the specific heat and the resistivity consistent with the literature, we conclude that a description in terms of a clean ferromagnetic quantum critical point is not sufficient. Instead we expect that microscopic inhomogeneities and clustering of the Ni atoms represent aspects that have to be taken into account for a full description.

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