Non-Fermi-Liquid Behavior at a Ferromagnetic Quantum Critical Point in Ni_xPd_{1-x}

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Ni_xPd_{1-x} is investigated at the border of enhanced Pauli paramagnetism and itinerant ferromagnetism. We provide convincing experimental evidence for the occurrence of a ferromagnetic (FM) quantum critical point (QCP). At a nickel concentration $x = 0.025 \pm 0.002$, the concentration dependence of the Curie temperature, as well as the temperature dependencies of the electrical resistivity, the magnetic susceptibility, and of the linear term of the specific heat follow the theoretical predictions of a FM QCP within experimental uncertainties. [S0031-9007(99)09221-2]

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After the observation of striking departures from the predictions of Fermi-liquid theory in $U_xY_{1-x}Pd_3$ [1,2], during the last decade non-Fermi-liquid (NFL) behavior of highly correlated electron systems was in the focus of experimentalists and theoreticians [3]. In most cases reported so far, NFL behavior appears close to the phase boundary of magnetic order. A continuous increase of the linear term of the heat capacity down to the lowest temperatures and significant deviations from a T^2 dependence of the resistivity were classified as hallmarks of NFL behavior. The largest body of experimental evidence has been presented in heavy-fermion systems (HFS), in which competing RKKY and Kondo interactions offer the opportunity to tune the systems towards vanishing magnetic order. Alloying or pressure have been used to establish a $T = 0$ K magnetic phase transition.

To describe the experimental observations different theoretical concepts have been worked out. For Kondo systems two-channel [4] and multichannel Kondo models [5] have been developed. In diluted systems which reveal inherent disorder, theories taking a distribution of Kondo temperatures into account [6] or an interpretation in terms of a spin-glass-like Griffiths phase [7] have been proposed. Finally, theories were derived which exhibit a ferromagnetic (FM) or antiferromagnetic (AFM) quantum critical point (QCP) characterized by a $T = 0$ K phase-transition temperature. Using renormalization-group theories [8,9] it has been shown that quantum systems depend crucially on the spatial dimensions and on the dynamic exponent. Quantum phase transitions also were described in terms of phenomenological spin-fluctuation models with great success [10–12].

So far most of the investigations in the field of HFS deal with vanishing AFM phase transitions (for a review, see [13]). From the many systems that have been studied, $CeCu_{6-x}Au_x$ [14] and $CeNi_2Ge_2$ [15] are the most promising candidates for the observations of a QCP. In the former case, heat capacity and resistivity can be consistently explained taking two-dimensional spin fluctuations into account and neutron scattering results revealed that these fluctuations indeed may exist [16]. In $CeNi_2Ge_2$ [15] the heat capacity and the thermal expansion revealed an increase towards low temperatures as theoretically predicted for an AFM QCP. One of the rare examples of HFS which is close to ferromagnetic order is $Th_{1-x}U_xCu_2Si_2$ [17]. And indeed, in a limited temperature range and close to the critical concentration a logarithmic increase has been detected for the linear term of the specific heat.

The pure d-transition metal systems MnSi and $ZrZn_2$ have been investigated with respect to quantum critical behavior [11]. Here the resistivity as a function of pressure revealed significant deviations from Fermi-liquid behavior as ferromagnetism becomes suppressed. Following this line of research we wanted to study a FM QCP in a transition metal with marginal disorder only. Quite naturally Pd, which is a strongly enhanced Pauli paramagnet close to FM order, seems to be the ideal starting material to investigate a FM 0 K phase transition. The theoretical predictions for a FM QCP are a logarithmic increase of the coefficient of the linear term of the specific heat [12,18], a low-temperature resistivity that follows a $T^{5/3}$ dependence [12,19], a magnetic susceptibility that increases as $\chi_0 - \chi_1 T^{3/4}$ towards low temperatures [12], and a dependence on an external system parameter, which in our case will be the impurity concentration x, which follows $(x - x_c)^{3/4}$ [8,12]. x_c denotes the critical concentration where FM order is suppressed.

In this Letter we report on systematic heat capacity, electrical resistivity, and magnetization experiments in Ni_xPd_{1-x} alloys at ambient pressure. It has long been known that roughly 2.5% of nickel ions doped into palladium induce FM order [20]. Hence it seems an ideal system to study quantum critical behavior as the induced structural and magnetic disorder is rather small and disorder phenomena are believed to play only a minor role. Already 30 years ago, nickel doped Pd has been a paramount example to study the low-temperature heat capacity and resistivity. Within early spin-fluctuation (SF) theories [21] the occurrence of an additional $T^3 \ln(T/T_0)$ in

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the heat capacity has been predicted, a contribution that still can be described in the framework of Fermi-liquid (FL) theory. But no convincing experimental evidence has been provided for the existence of this SF term [22–25]. The electrical resistance at low temperatures in most cases was analyzed assuming a T^2 dependence [26], but one report reveals values of the exponent significantly below $n = 2$ [27].

Ni_xPd_{1-x} samples for concentrations $0 \leq x \leq 0.1$ were prepared from high purity (5N) starting materials by argon arc techniques. The samples were remelted many times and then annealed for five days at 1000 °C. X-ray diffraction and microprobe analysis revealed single-phase materials. Magnetization experiments have been performed using a quantum design SQUID magnetometer for temperatures $1.8 < T < 400$ K; the resistivity was measured in a He flow cryostat and in a $^3He/^4He$ dilution cryostat for temperatures $0.05 < T < 300$ K. The temperature dependence of the heat capacity was followed in a ^4He and in a ^3He cryostat for temperatures $0.3 < T < 30$ K.

At first we focus on the concentration dependence of the FM phase-transition temperature T_c close to the critical concentration. T_c does not show up in the temperature dependencies of the resistivity and heat capacity and has to be determined from the magnetization data [20]. Our results for nickel concentrations $x \leq 0.1$ are plotted in Fig. 1, together with all published data [20,28,29] (we only omitted Mössbauer data where additional Fe impurities have been used as probe ions [30]). We determined the T_c values from the paramagnetic (PM) Curie temperatures. But they are in good agreement with the data determined from low-field magnetization [20] and from small-angle neutron scattering [29]. The results of the FM phase-transition temperatures vs nickel concentration $x - x_c$ are shown on a double logarithmic plot. Close to $x = 0.1$ a clear change of slope can be observed. While T_c follows a square root dependence at high concentrations (see also inset of Fig. 1), it reveals a $(x - x_c)^{3/4}$ behavior for nickel concentrations close to the critical concentration x_c. Exactly this dependence has been predicted for a FM QCP [8,12]. From these experiments we determined $x_c = 0.026 \pm 0.002$.

Figure 2 shows the electrical resistance as a function of $T^{5/3}$ for concentrations close to the critical concentration. While the resistivity for $x = 0.026$ exactly follows the $T^{5/3}$ dependence, which is expected at a FM QCP [12], for more than two decades in temperature, samples with slightly higher and lower Ni concentrations clearly show significantly increased temperature exponents. To demonstrate that quantum critical behavior is limited to a narrow concentration range around the critical concentration we fitted the electrical resistivity using $\rho(T) = \rho_0 + A'T^n$. ρ_0 is the residual resistance, A' a generalized FL coefficient, and n a generalized temperature exponent. The results of these fits are shown as solid lines in Fig. 2 and the parameters are given in Fig. 3 with n (upper panel) and A' (lower panel) as a function of nickel concentrations x. n is close to 2 for low x, as expected for a pure FL and is significantly larger than 2 in the magnetically ordered phase due to the scattering of charge carriers by magnons. In a narrow concentration range n decreases and arrives exactly at a value of $n = 1.67$ close to x_c. Concomitantly A' reveals a sharp maximum at the critical concentration. The value $n = 5/3$ characteristic for a FM QCP holds for concentrations $0.022 \leq x \leq 0.026$ while the prefactor A' peaks at $x = 0.022$ [12]. Taking into account that the

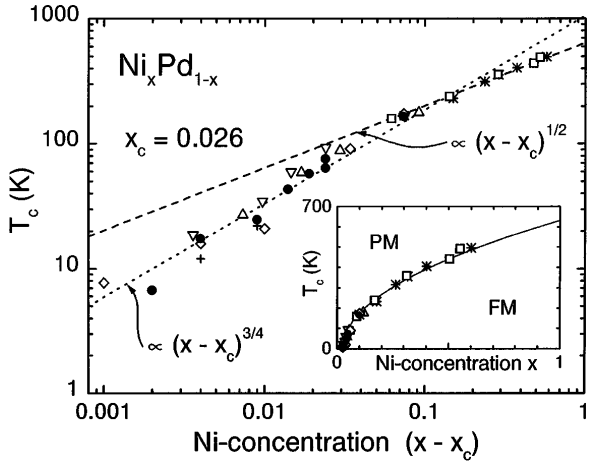

FIG. 1. Curie Temperature T_c as a function of $x - x_c$ in a double logarithmic plot: (□) Marian (1937); (△) Crangle and Scott (1965); (+) Murani *et al.* (1974); (∗) Fujiwara *et al.* (1976); (▽) Beille and Tournier (1976); (◇) S. K. Burke *et al.* (1982); (●) present data [20,28,29]. The dashed line indicates $(x - x_c)^{1/2}$, dotted line $(x - x_c)^{3/4}$. x_c denotes the critical concentration. The inset shows $T_c(x)$ on a linear scale following a square root behavior at high x (solid line).

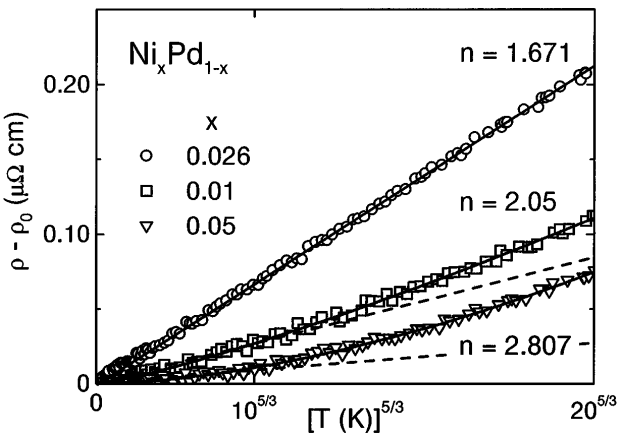

FIG. 2. $\rho - \rho_0$ is plotted vs $T^{5/3}$ for three different concentrations, $x = 0.01$ in the PM regime, $x = 0.026$ at the critical concentration, and $x = 0.05$ in the FM regime. The solid lines were calculated according to $\rho = \rho_0 + A'T^n$. In this representation the resistivity at a FM QCP shows a linear increase (dashed lines). For $x = 0.026$ solid and dashed lines coincide.

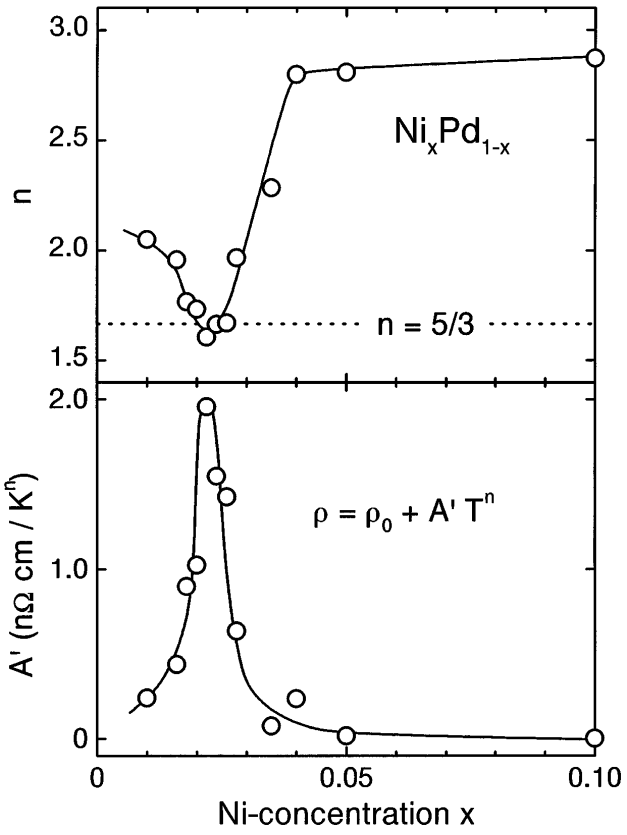

FIG. 3. A' and n determined by a fit to $\rho = \rho_0 + A'T^n$ vs Ni concentrations $0 \leq x \leq 0.1$. The solid lines are drawn to guide the eye. Upper panel: Generalized temperature exponent n vs x. Lower panel: Generalized FL coefficient A' vs x.

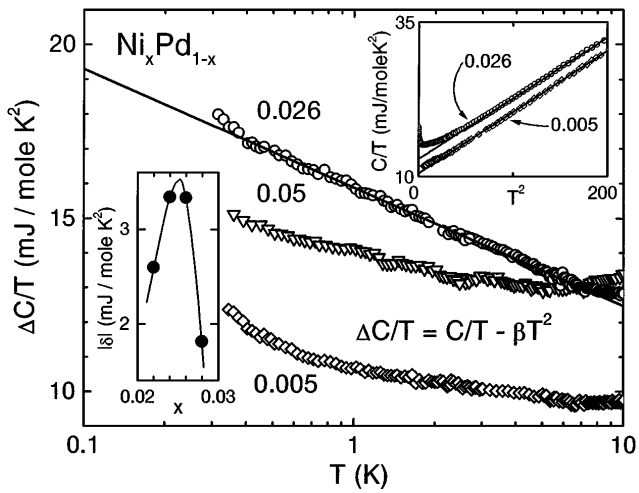

FIG. 4. $\Delta C/T$ vs the logarithm of the temperature for three concentrations $x = 0.005$, 0.026, and 0.05. The data for $x_c = 0.026$ were fitted by $\Delta C/T = \delta \ln(T/T_c)$ (solid line). The inset in the upper right corner shows the raw data plotted as C/T vs T^2. The solid lines are fits to determine the phonon contribution to the specific heat. The fitting parameter δ in the NFL regime is shown in the lower inset. The solid line is drawn to guide the eye.

temperature exponent is slightly lower at $x = 0.022$ and that the values of A' and n are strongly correlated, the best estimate of the critical concentration on the basis of the electrical resistance experiments is $x_c = 0.024 \pm 0.002$.

The results of the heat capacity experiments are given in Fig. 4. The upper inset of Fig. 4 shows the results plotted as C/T vs T^2. From these results, which reveal a constant slope roughly up to 15 K, we determined the phonon contributions for each concentration separately. This is important as it has been shown that also the T^3 phonon term strongly depends on the nickel concentration [23]. At low temperatures significant deviations from the normal metallic behavior show up and close to a FM QCP we expect $C/T = \delta \ln(T/T_0) + \beta T^2$ [12,18]. To elucidate this behavior in more detail, Fig. 4 shows the pure electronic heat capacity for three different samples. Here the phonon contributions, βT^2, as determined by the slopes of the solid lines in the inset of Fig. 4, were subtracted from the raw data. For low nickel concentrations ($x = 0.005$) we find an almost constant contribution which moderately increases towards low temperatures, possibly due to contributions from spin fluctuations. In the magnetically ordered sample ($x = 0.05$, $T_c = 75$ K) the linear term increases for $T > 8$ K, but becomes slightly enhanced for low temperatures. For a ferromagnet we expect an increase as $C_m \propto T^{3/2}$, due to magnon contributions. This, however, is not what we observe and we speculate that for $x = 0.05$ disorder effects contribute to the heat capacity at low temperatures. It is just at the critical concentration that C/T increases logarithmically towards low temperatures as predicted at a QCP. The logarithmic increase definitely holds for $0.4 < T < 8$ K. Similar results were observed for the concentrations $x = 0.022$, 0.024, and 0.028. The prefactor of the NFL contribution of these samples is indicated as solid circles in the lower panel of Fig. 4. Clearly we find a maximum of the NFL-like heat capacity at $x = 0.025 \pm 0.001$.

Finally Fig. 5 shows the temperature dependence of the dc-magnetization for a series of experiments for concentrations $0 < x < 0.05$. At a FM QCP, $\chi = \chi_0 - \chi_1 T^{3/4}$ is expected [12] and hence, we show the magnetic susceptibility vs temperature on a double logarithmic plot. Indeed, close to x_c we find an almost linear increase of the magnetic susceptibility with a slope of 3/4. But we have to agree that the magnetic susceptibility is the least convincing quantity of the data sets presented in this communication. However, the characteristic NFL behavior still can be detected over almost one decade. The deviations towards low temperatures can be explained by the fact that we performed dc-magnetization experiments at 0.5 T and any defect states will yield significant deviations especially at low temperatures. To get more reliable results ac-experiments have to be performed and these experiments, including low temperature measurements, are planned for the near future.

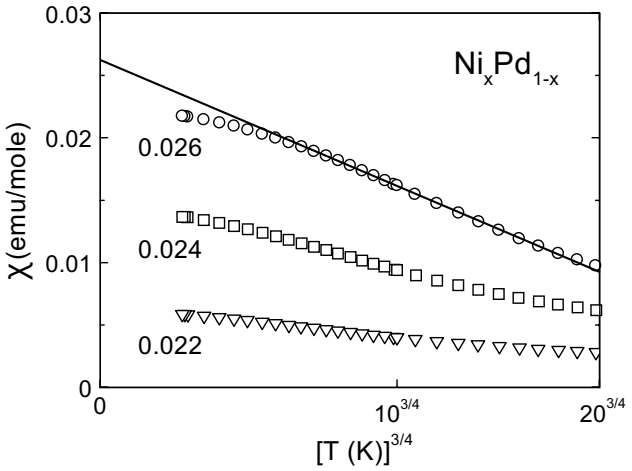

FIG. 5. The magnetic susceptibility χ vs $T^{3/4}$ for three concentrations, $x = 0.022$, 0.024, and 0.026, close to the critical concentration. The straight line indicates a fit using $\chi = \chi_0 - \chi_1 T^{3/4}$.

In conclusion, we provide convincing experimental evidence for the occurrence of a FM QCP in Ni_xPd_{1-x} at a critical concentration $x_c = 0.025$. This has been demonstrated for the concentration dependence of the Curie temperature and for the temperature dependencies of resistivity, heat capacity, and magnetic susceptibility. To our knowledge, this is the first time that the theoretical predictions of a QCP have been met in all four quantities, clearly demonstrating the validity of the model. It would be highly interesting to study the dynamical susceptibility in neutron scattering and in NMR experiments.

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