Quantum critical behavior in the heavy Fermion single crystal $Ce(Ni_{0.935}Pd_{0.065})_2Ge_2$

2011 J. Phys.: Conf. Ser. 273 012018

(http://iopscience.iop.org/1742-6596/273/1/012018)

Quantum critical behavior in the heavy Fermion single crystal $Ce(Ni_{0.935}Pd_{0.065})_2Ge_2$

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Abstract.
We have performed magnetic susceptibility, specific heat, resistivity, and inelastic neutron scattering measurements on a single crystal of the heavy Fermion compound $Ce(Ni_{0.935}Pd_{0.065})_2Ge_2$, which is believed to be close to a quantum critical point (QCP) at T = 0. At lowest temperature(1.8-3.5 K), the magnetic susceptibility behaves as $\chi(T) - \chi(0) \propto T^{-1/6}$ with $\chi(0) = 0.032 \times 10^{-6}$ m^3/mole (0.0025 emu/mole). For $T < 1$ K, the specific heat can be fit to the formula $\Delta C/T = \gamma_0 - T^{1/2}$ with γ_0 of order 700 mJ/mole-K^2. The resistivity behaves as $\rho = \rho_0 + AT^{3/2}$ for temperatures below 2 K. This low temperature behavior for $\gamma(T)$ and $\rho(T)$ is in accord with the SCR theory of Moriya and Takimoto[1]. The inelastic neutron scattering spectra show a broad peak near 1.5 meV that appears to be independent of Q; we interpret this as Kondo scattering with $T_K = 17$ K. In addition, the scattering is enhanced near Q=(1/2, 1/2, 0) with maximum scattering at $\Delta E = 0.45$ meV; we interpret this as scattering from antiferromagnetic fluctuations near the antiferromagnetic QCP.

In strongly correlated electron systems, a quantum critical point (QCP) separates an antiferromagnetic (AFM) or ferromagnetic(FM) state from a nonmagnetic Fermi liquid state at $T = 0$ K. This QCP can be tuned by adjusting a control parameter such as doping parameter x, external pressure P, or applied magnetic field H. In the vicinity of the QCP, the critical fluctuations are quantum in nature and induce unique behavior. The nature of this quantum ground state phase transition poses one of the most significant challenges in condensed matter physics. Heavy Fermion (HF) compounds are very good candidates for studying the QCP. These compounds behave as Fermi Liquids (FL), with large values for the specific heat coefficient $\gamma = C/T$ and susceptibility $\chi(0)$, and with the resistivity varying as $\Delta\rho \propto T^2$ at low temperature. When such systems are tuned close to a QCP, fluctuations of the nearby magnetically ordered state affect the thermodynamic behavior and lead to Non-Fermi Liquid (NFL) behavior such as $\Delta\rho \propto T^\alpha$ with $\alpha < 2$ and $C/T \propto ln(T_0/T)$ at low temperature[2].

The approach to a QCP has been attained in HF systems by doping in the tetragonal compounds $Ce(Ru_{1-x}Rh_x)_2Si_2$ (x=0.03)[3, 4] and $Ce_{1-x}La_xRu_2Si_2$ (x=0.075)[5, 6] as well as in the orthorhombic compound $CeCu_{6-x}Au_x$ (x=0.1)[7, 8, 9], by application of a magnetic field in $YbRh_2Si_2$[10], and with no additional control parameter in the compound β-$YbAlB_4$[11].

A conventional spin fluctuation theory has been established to explain the non-Fermi liquid behavior. This theory, implemented through a renormalization-group approach[12, 13] or through the self-consistent renormalization(SCR) method[1], has successfully explained various aspects of the non-Fermi liquid behavior. However, the experimental results for some systems do not follow this spin fluctuation theory. A locally critical phase transition has been invoked to explain the behavior of a few systems[14]. Hence, measurements in other systems are needed to understand the behavior near the QCP in HF materials.

Below 2 K, the tetragonal compound $CeNi_2Ge_2$ exhibits behavior for the resistivity and specific heat that obeys the predictions of the SCR theory for a 3D AFM QCP[1]: $\Delta\rho \propto T^{3/2}$ and $C/T \propto \gamma(0) - A\sqrt{T}$ [15, 16]. Inelastic neutron scattering shows two low energy features[17, 18, 19]. A broad peak at 4 meV that is only weakly **Q**-dependent corresponds to Kondo scattering with $T_K = 46$ K[20]. A peak at 0.7 meV that is highly **Q**-dependent and shows a maximum intensity at $\mathbf{Q}_N$= (1/2 1/2 0) corresponds to scattering from antiferromagnetic fluctuations. Although this compound is clearly close to a QCP, when T decreases below 0.4 K or B increases above 2 T, the system enters the FL state and C/T shows saturation[15, 16, 21]. The compound can be brought closer to the QCP by alloying with Pd. According to the phase diagram proposed by Knebel et al[22], the QCP occurs in $Ce(Ni_{2-x}Pd_x)_2Ge_2$ when x =0.065. Fukuhara et al found[23] that when $x = 0.10$, the ordering wavevector remains $\mathbf{Q}_N$= (1/2 1/2 0); for larger x it changes to $\mathbf{Q}_N$= (1/2 1/2 1/6).

Here we report measurements of the magnetic susceptibility, the specific heat for a series of magnetic fields, the resistivity, and the inelastic neutron scattering of a single crystal of $Ce(Ni_{0.935}Pd_{0.065})_2Ge_2$ grown with 58-Ni to reduce the incoherent scattering. For this alloy, we report the inelastic neutron spectra for the first time.

Single crystals were grown using the Czochralski method. The magnetization was measured in a commercial superconducting quantum interference device (SQUID) magnetometer. The specific heat measurements were performed in a commercial physical properties measurement system (PPMS). The electrical resistivity was also measured in the PPMS using the four wire method. The inelastic neutron experiment were performed on the MACS spectrometer[24] at the NIST center for neutron research (NCNR).

The magnetic susceptibility $\chi(T)$ is shown in figure 1(a). At high temperatures the data follow Curie-Weiss (CW) behavior $\chi(T) = C_{eff}/(T - \theta)$; in the range 150-300 K, the CW fit yields a moment 2.84 μ_B and θ = -118 K. Below 100 K, $\chi(T)$ is enhanced over the CW value but no long range magnetic order is observed down to 1.8 K. In the traditional spin fluctuation theory[1, 2, 12, 13] for a three dimensional system near a QCP, $\chi(T) \propto T^{-3/2}$ is expected for an AFM while $\chi(T) \propto T^{-4/3}$ is expected for a FM. In figure 1 (b), our data for $Ce(Ni_{0.935}Pd_{0.065})_2Ge_2$ follow the behavior of $(\chi(T) - \chi(0)) \propto T^{-1/6}$. Exponents for the susceptibility that are smaller than 1 are also observed for β-$YbAlB_4$[11] where $\chi(T) \propto T^{-1/3}$ when B=0.05 T and for $YbRh_2(Si_{0.95}Ge_{0.05})_2$ where $(\chi(T) - \chi(0)) \propto T^{-0.6}$[25].

In figure 2(a) we plot the specific heat C/T. The zero field data are very similar to that reported by Kuwai et al for $Ce(Ni_{0.936}Pd_{0.064})_2Ge_2$[26]. The data are logarithmic with temperature in the range 1 K to 5 K. When an external field is applied, the data deviate from the ln$T$ behavior and show saturation. As for $CeNi_2Ge_2$, this is because the magnetic field forces the system to enter the FL state. However, in figure 2(b), the low temperature (T <1 K) data can be seen to deviate slightly from the ln$T$ behavior; in this temperature range the data can be fit(figure 2(c)) to the form $\gamma_0 - aT^{1/2}$ (with γ_0 of order 695 mJ/mole-K^2), which as mentioned is the expected behavior in the SCR theory.

In figure 3(a) the resistivity $\rho(T)$ data are seen to be roughly linear in temperature over a wide range 0.4 K to 12 K, indicating NFL behavior. On the expanded scale of figure 3(b), $\rho(T)$

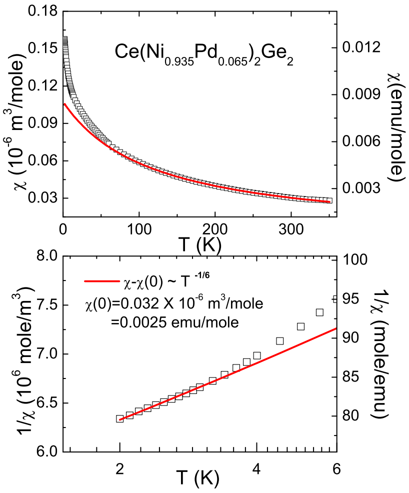

Figure 1. (a): Magnetic susceptibility of $Ce(Ni_{0.935}Pd_{0.065})_2Ge_2$. The solid line is the high temperature Curie-Weiss fit. (b): Low temperature inverse susceptibility. The solid line is $\chi(T) - \chi(0) \propto T^{-1/6}$.

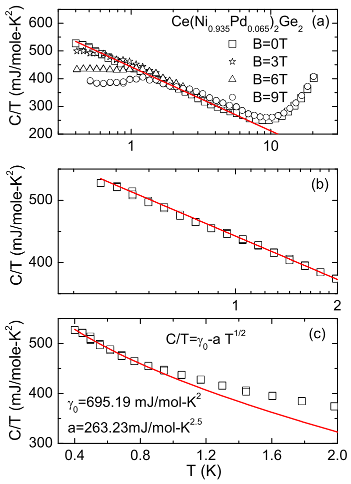

Figure 2. (a): C/T in applied fields of B=0, 3 T, 6 T and 9 T. (b): Low temperature zero field C/T curve in a logarithmic temperature scale. (c) Low temperature zero field C/T data. The solid line is $\gamma_0 - aT^{1/2}$.

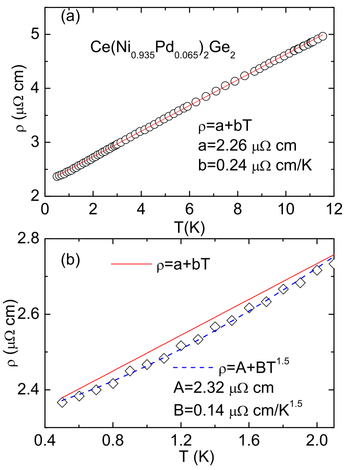

Figure 3. Resistivity for $Ce(Ni_{0.935}Pd_{0.065})_2Ge_2$. The solid line is the linear fit for the temperature range 0.4 K to 12 K while the dashed line represents $T^{3/2}$ dependence in the temperature range 0.4 K to 2 K.

deviates from T-linear, following a $T^{3/2}$ dependence. Again, this is the expected behavior in the SCR theory.

Hence, at temperatures higher than 2 K, this alloy exhibits typical NFL behavior, with the resistivity varying as $\rho(T) \propto T$, the specific heat varying as $C/T \propto \ln\frac{T_0}{T}$, and the susceptibility varying as $(\chi(T) - \chi(0)) \propto T^{-1/6}$. At temperatures below 1 K, however, $\rho(T) \propto T^{3/2}$ and $C/T \propto \gamma_0 - a\sqrt{T}$, thereby following the expected behavior of the SCR theory for a three dimensional AFM QCP spin fluctuation system.

In figure 4, we plot the INS spectra for $Ce(Ni_{0.936}Pd_{0.065})_2Ge_2$. A broad peak near 1.5 meV appears to be only weakly Q-dependent. As for the 4 meV peak in $CeNi_2Ge_2$, we identify this as representing Kondo scattering. Extra intensity is observed when $\Delta E < 1$ meV in the vicinity of $\mathbf{Q} = (1/2\ 1/2\ 0)$. This is the wavevector of the antiferromagnetic fluctuations seen for $CeNi_2Ge_2$ near 0.7 meV, hence we identify it as the critical wavevector $\mathbf{Q}_N$ for the QCP.

We first fit these spectra to the sum of a quasi-elastic Lorentzian (peak width Γ_0) and an inelastic Lorentzian (peak position E_1 and width Γ_1) (Fig. 4(a)). For $\mathbf{Q} = (1/2\ 1/2\ 0)$ we find E_1=1.51 meV, Γ_1=0.90 meV and Γ_0=0.35 meV; for $\mathbf{Q}$=(3/4 3/4 0), we find E_1=1.35 meV and Γ_1=0.88 meV. In figure 4(b), we use a second approach to obtain the AFM fluctuation spectra: we subtract the spectra for $\mathbf{Q} = (3/4\ 3/4\ 0)$ from that measured at (1/2 1/2 0). A fit of the difference to a quasielastic Lorentzian peak gives the AFM fluctuation energy scale as $\Gamma_0 = 0.44$ meV.

The value of E_1 is around 1.35-1.51 meV represents a Kondo temperature in the range 15.7-17.5 K. We note that this is smaller than the value 46 K seen in $CeNi_2Ge_2$, so that the approach to the QCP involves a reduction of T_K. We note in addition that T_K does not vanish at the QCP. A finite Kondo temperature is expected for a spin density wave system at a QCP.

The value of the AFM fluctuation energy Γ_0 is around 0.35-0.44 meV appears to be smaller

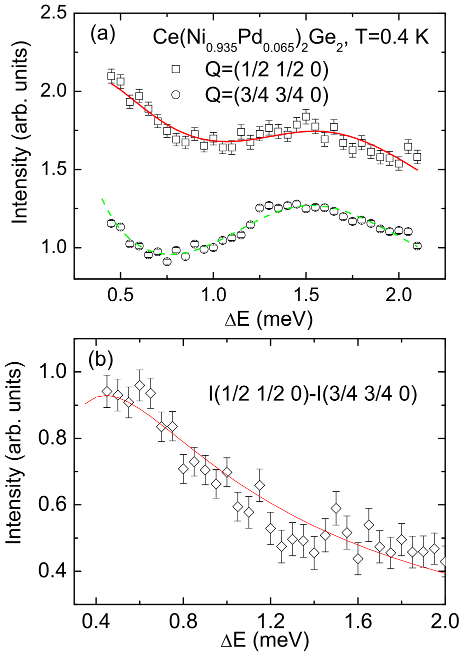

Figure 4. Inelastic neutron scattering spectra for $Ce(Ni_{0.935}Pd_{0.065})_2Ge_2$. The data were collected on MACS. In (a), the scattering at the critical wavevector $\mathbf{Q}_N = (1/2\ 1/2\ 0)$ is compared to the scattering at (3/4 3/4 0). The lines represent fits to the sum of a quasi-elastic and an inelastic Lorentzian peak, as described in the text. In (b) the difference between the data for **Q**= (1/2 1/2 0) and (3/4 3/4 0) is compared to a quasielastic Lorentzian (solid line).

than the fluctuation energy scale of pure $CeNi_2Ge_2$ where Γ=0.7 meV [17]. This represents "critical slowing down" of the AFM spin fluctutaions. However, despite the fact that this system is believed to sit at the QCP, the lifetime of the AFM spin fluctuations is finite, i.e. it does not diverge as expected at the critical point. Such a "saturation" of the spin fluctuation lifetime has been observed for other HF systems where the QCP is attained by alloying [4, 6] and may arise from the fact that the QCP occurs in a disordered environment.

Research at UC Irvine was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award DE-FG02-03ER46036. Work at Los Alamos National Laboratory was performed under the auspices of the U.S. DOE/Office of Science. Work at ORNL was sponsored by the Laboratory Directed Research and Development Program of ORNL, managed by UT-Battelle, LLC, for the U. S. DOE, and was supported by the Scientific User Facilities Division, Office of Basic Energy Sciences, DOE. We acknowledge the support of the National Institute of Standards and Technology, U. S. Department of Commerce, in providing the neutron research facilities used in this work. The identification of commercial products does not imply endorsement or recommendation by the National Institute of Standards and Technology.

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