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2000 J. Phys.: Condens. Matter 12 L533

(http://iopscience.iop.org/0953-8984/12/32/101)

J. Phys.: Condens. Matter **12** (2000) L533–L540. Printed in the UK PII: S0953-8984(00)15614-0

LETTER TO THE EDITOR

Anomalous low temperature states in $CeNi_2Ge_2$ and $CePd_2Si_2$

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Received 18 July 2000

Abstract. High purity samples of the paramagnetic 4f-electron metal $CeNi_2Ge_2$ exhibit a non-Fermi-liquid form of the resistivity $\Delta\rho \sim T^x$ with $x < 1.5$ and decreasing towards 1 with increasing sample purity. Measurements of ρ versus T as a function of magnetic field and pressure show that this strange metallic phase is connected to the proximity of an antiferromagnetic quantum critical point as in the isoelectronic relative $CePd_2Si_2$ near 2.8 GPa. The anomalous power-law dependence is surprisingly stable over extended ranges in temperature and pressure and challenges current theory of magnetic quantum phase transitions.

1. Introduction

The heavy fermion compounds offer a fertile ground for the search for new states of interacting electron systems. Typically, one observes at high temperature a nearly constant and large resistivity consistent with scattering from thermally disordered local magnetic moments. Below an upper temperature scale, T_{sf}, the resistivity drops with decreasing temperature and in the absence of a phase transition the resistivity and other bulk properties follow the predictions of Fermi-liquid theory below a lower temperature scale, T_{FL}.

An increasing number of systems are coming to light, however, in which the Fermi-liquid regime is suppressed to very low temperatures or even masked by the onset of superconductivity (see e.g. [1]). Under these conditions, the range between T_{FL} and T_{sf}, sometimes described as a spin liquid regime, extends down to very low temperatures and becomes the dominant feature of the system. The unconventional normal states observed in these metals are usually examined in terms of phenomenological models for the fluctuations of the local order parameter, i.e. the local magnetization for ferromagnetism and the local staggered magnetization for antiferromagnetism [2]. When the magnetic ordering temperature is suppressed to absolute zero, these modes soften over large portions of reciprocal space at low temperatures, leading to a strong enhancement of the quasiparticle scattering rate and potentially to a breakdown of the Fermi-liquid description in its simplest form. This breakdown should be particularly apparent in the temperature dependence of the resistivity $\rho(T)$ that may deviate from the usual Fermi-liquid form $\rho(T) \sim T^2$ in pure samples at low temperatures.

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0953-8984/00/320533+08$30.00

The critical point, where the magnetic ordering temperature extrapolates to zero and a breakdown of the Fermi-liquid description is expected, can in a number of stoichiometric compounds be approached by tuning the lattice density under hydrostatic pressure. This route to unconventional metallic states is fundamentally distinct from the observations of non-Fermi-liquid behaviour in the more extensively studied doped and disordered uranium and cerium alloys [3, 4]. In the present investigation, the role of disorder for producing an unconventional normal state can be systematically studied and minimized by improving the sample quality. The systems that we have selected, namely the isostructural and isoelectronic relatives $CeNi_2Ge_2$ and $CePd_2Si_2$ are available with residual resistivities in the $\mu\Omega$ cm range and there now is strong evidence that in both cases the largest deviations from Fermi-liquid behaviour occur in the purest samples.

$CeNi_2Ge_2$ shows no magnetic order at low pressures and together with $CePd_2Si_2$, which has a slightly larger unit cell volume and orders antiferromagnetically at ambient pressure, conveniently spans an antiferromagnetic quantum critical point. The two systems have allowed the first detailed examination of a quantum phase transition in pure metals. In this letter we argue that the origin of strange metallic behaviour in ambient-pressure $CeNi_2Ge_2$ is unrelated to static disorder, but rather lies in the proximity to the threshold of magnetism. By examining the pressure-dependence of the low temperature power-law of the resistivity, which drifts with increasing lattice density towards the T^2-form associated with a conventional Fermi-liquid, we show that $CeNi_2Ge_2$ lies within a few tenths of a GPa to the high pressure side of the quantum critical point associated with the disappearance of magnetism in $CePd_2Si_2$. The re-emergence of Fermi-liquid behaviour with pressure is much slower in $CeNi_2Ge_2$ and $CePd_2Si_2$ than in the comparable reference materials $CeIn_3$, $CeRu_2Ge_2/Si_2$ or $CeRh_2Si_2$, indicating large correlation lengths over an extended region in pressure. This nearly critical normal state is boxed in between the magnetism studied in $CePd_2Si_2$ and another high pressure phase, which arises near 1.5 GPa in $CeNi_2Ge_2$.

$CeNi_2Ge_2$ and $CePd_2Si_2$ are isostructural to the heavy fermion superconductor $CeCu_2Si_2$ [5] and its larger volume relative $CeCu_2Ge_2$ [6] (both with the $ThCr_2Si_2$ structure), but differ from $CeCu_2Si_2$ in the number of d-electrons in the d-metal constituent, and hence in the character of the Fermi surface and in the magnetic properties. At ambient pressure, $CePd_2Si_2$ orders in an antiferromagnetic structure ($\boldsymbol{Q} = [\frac{1}{2}\frac{1}{2}0]$) with a comparatively small moment of 0.7 μ_B below a Néel temperature T_N of about 10 K [7, 8]. In a recent study [9], we have elucidated the phase diagram of $CePd_2Si_2$ up to hydrostatic pressures of about 3 GPa. The Néel temperature drops linearly with pressure above 1.5 GPa and extrapolates to zero at a critical pressure, $p_c \simeq 2.8$ GPa, while the shoulder in the resistivity, T_{sf}, shifts from 10 K at low pressure to about 100 K near p_c. Superconductivity appears below 430 mK in a limited pressure region of a few tenths of a GPa on either side of p_c. This behaviour, and perhaps that in the related systems $CeRh_2Si_2$ [10] and $CeIn_3$ [11], is consistent with anisotropic pairing arising from magnetic interactions [12, 13]. The electronically and structurally equivalent compound $CeNi_2Ge_2$ [14], which is of central interest here, has a slightly smaller lattice constant and its zero pressure behaviour may be expected to be similar to that of $CePd_2Si_2$ at a pressure close to but higher than p_c. Recently, this picture has been confirmed by partial substitution of Ni in $CeNi_2Ge_2$ with Pd [15] and by overdoping of $CeNi_2Ge_2$ with Ge [16], both of which produce ordered states at low temperature. In 4f-electron systems superconductivity is often linked to a magnetic quantum critical point. For the cleanest samples of $CeNi_2Ge_2$ we indeed find a short coherence length superconducting state at ambient pressure below about 0.2 K, similar to that of $CePd_2Si_2$ at p_c [17, 18]. The transition is very sensitive to hydrostatic pressure and above 0.4 GPa, no drop of the resistivity is observed. The resulting phase diagram is reminiscent of the behaviour of $CePd_2Si_2$ at high

pressure and is consistent with our assumption that $CeNi_2Ge_2$ is a smaller volume relative of $CePd_2Si_2$.

2. Results

In both $CeNi_2Ge_2$ and $CePd_2Si_2$, the temperature dependence of the resistivity is characterized over a wide range by a power-law $\rho = \rho_0 + AT^x$ with exponent x close to 1, and by rapid cross-overs to this form at high and low temperatures (figures 1 and 2). This power-law behaviour is a robust feature observed in all specimens of both materials. Experiments carried out on various samples and different sample orientations have revealed a variation of the exponents in the range $1.1 < x < 1.5$ and a general trend towards lower values for purer (lower ρ_0) specimens, indicating a possible limiting value close to 1—and thus the clearest departure from the Fermi-liquid form—for ideally pure samples (inset of figure 1).

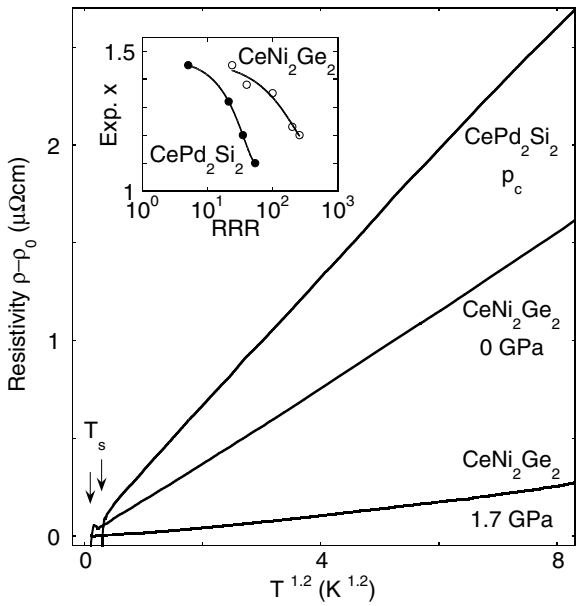

Figure 1. Low temperature resistivity plotted against $T^{1.2}$ (i) in $CeNi_2Ge_2$ at ambient pressure (denoted everywhere as 0 GPa) and at high pressure (1.7 GPa), and (ii) in $CePd_2Si_2$ near the critical pressure $p_c \simeq 2.8$ GPa, where $T_N \to 0$ K. For comparison, the curves are shifted by their respective $T = 0$ intercepts, i.e. their residual resistivities, ρ_0. For $CePd_2Si_2$, $\rho_0 \simeq 1.4\ \mu\Omega$ cm at 2.8 GPa, while for $CeNi_2Ge_2$, $\rho_0 \simeq 0.19\ \mu\Omega$ cm at 0 GPa and 0.16 $\mu\Omega$ cm at 1.7 GPa. Inset: the resistivity exponent x, defined by fitting $\rho(T) = \rho_0 + AT^x$ from 0.4 K to 4 K for a number of samples of $CePd_2Si_2$ and of $CeNi_2Ge_2$ of varying residual resistivity ratio, RRR $\equiv \rho(293\ \text{K})/\rho_0$. Details of sample preparation and experimental procedure are given in [17].

These properties of our two tetragonal metals contrast sharply with those of the cubic antiferromagnet $CeIn_3$ [11, 13]. In the latter, the resistivity deviates from the Fermi-liquid form only in a very narrow pressure range near the critical pressure p_c. At p_c and in low magnetic fields the resistivity exponent, or more precisely $\mathrm{d}\ln(\rho - \rho_0)/\mathrm{d}\ln T$, grows smoothly with decreasing temperature and tends towards a value of about $3/2$ near 1 K.

By contrast, the observed exponents in $CeNi_2Ge_2$ and $CePd_2Si_2$ are closer to 1 and nearly constant over almost two orders of magnitude in temperature. For the origin of this intriguing

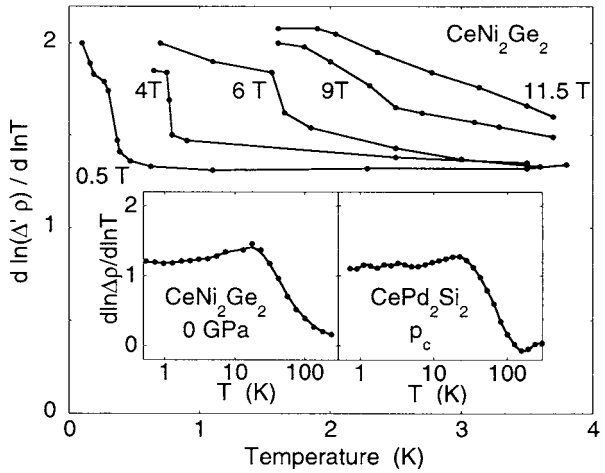

Figure 2. High sensitivity measurement of the logarithmic derivative ($\mathrm{d}\ln(\Delta'\rho \equiv T\mathrm{d}\rho/\mathrm{d}T)/\mathrm{d}\ln T$) of the resistivity $\rho(T)$ in $CeNi_2Ge_2$ ($\rho_0 \simeq 0.5\ \mu\Omega$ cm) at ambient pressure and in applied magnetic fields. The ordinate reduces to the resistivity exponent x when $\rho(T)$ can be expressed in the form $\rho(T) = \rho_0 + AT^x$. The resistivity was measured along the a-axis with the magnetic field applied along the c-axis. Left inset: $\mathrm{d}\ln(\Delta\rho \equiv \rho - \rho_0)/\mathrm{d}\ln T$ versus T for $CeNi_2Ge_2$ ($\rho_0 \simeq 0.19\ \mu\Omega$ cm) at ambient pressure. The ordinate again reduces to x under the condition given above. In contrast to $\Delta\rho$, $\Delta'\rho$ is independent of how ρ_0 is determined. In all cases presented in this paper analyses of $\mathrm{d}\ln\Delta\rho/\mathrm{d}\ln T$ and $\mathrm{d}\ln\Delta'\rho/\mathrm{d}\ln T$ yield qualitatively similar results. Right inset: $\mathrm{d}\ln(\Delta\rho \equiv \rho - \rho_0)/\mathrm{d}\ln T$ versus T for $CePd_2Si_2$ ($\rho_0 \simeq 1.4\ \mu\Omega$ cm) close to p_c.

difference, we may consider first the known magnetic structure of $CePd_2Si_2$, which suggests a frustrated spin coupling along the c-axis and hence a strongly anisotropic spin fluctuation spectrum [17, 13] that may be more nearly 2D than 3D as expected for $CeIn_3$. For a comparison with the predictions of the standard magnetic interaction model we consider first the limit in which regions about the 'hot spots' on the Fermi surface (those states whose self-energies can exhibit a non-Fermi-liquid form due to antiferromagnetic spin fluctuations) dominate the temperature dependence of the resistivity. This is expected to occur when a short circuiting via normal regions of the Fermi surface is blocked for example by isotropic potential scattering via weak residual impurities. At a continuous antiferromagnetic critical point one then expects in the low temperature limit $\Delta\rho \sim T^{d/z}$, where d is the effective dimension of the spin fluctuation spectrum and z is the dynamical exponent normally taken to be 2 in our problem [2]. For this simplest model the resistivity exponent is thus expected to be $3/2$ in 3D, in agreement with our observations in cubic $CeIn_3$, and unity for 2D, in closer agreement to the results in pure samples of the tetragonal $CePd_2Si_2$ and $CeNi_2Ge_2$. The precise degree of anisotropy can depend on details of the microscopic near-neighbour exchange constants, giving rise to the observed sample dependence of the power-law exponents.

Further evidence for this possibility is drawn from the linear pressure-dependence of T_N observed in $CePd_2Si_2$ close to p_c [13]. In the magnetic interaction model [2] this would indicate an effective dimension close to 2, in agreement with the simplest interpretation of the resistivity data. We note that by comparison $T_N(p)$ in $CeIn_3$ follows a power less than 1 close to p_c, consistent with $d = 3$ and with the observed low temperature resistivity $\Delta\rho \sim T^{1.5}$.

We stress that the dimensionality d refers to the magnetic fluctuation spectrum, not to the carriers, which remain unconstrained to move in 3D. Amongst the antiferromagnetic layered

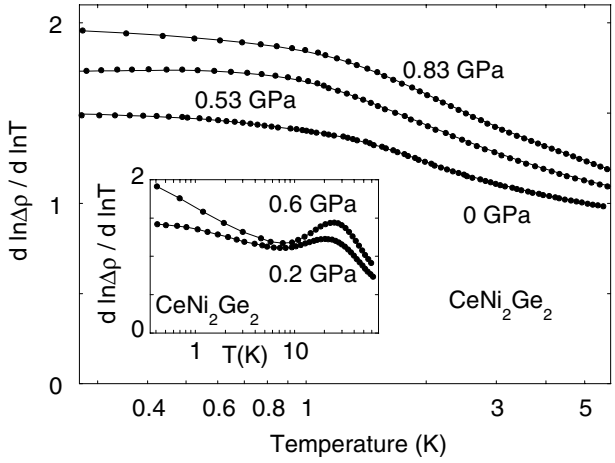

Figure 3. The logarithmic derivative $\mathrm{d}\ln(\Delta\rho \equiv \rho - \rho_0)/\mathrm{d}\ln T$ versus T in $CeNi_2Ge_2$ ($\rho_0 \simeq 3.8\ \mu\Omega$ cm) at zero magnetic field and in applied hydrostatic pressure. Inset: $\mathrm{d}\ln(\Delta\rho \equiv \rho - \rho_0)/\mathrm{d}\ln T$ versus T for a second sample of $CeNi_2Ge_2$ ($\rho_0 \simeq 1.8\ \mu\Omega$ cm) as a function of pressure.

insulators, quasi-2D magnetic behaviour has long been known to arise, for example, in the perovskite K_2NiF_4, in which the magnetic moments of Ni occupy bct positions and order with $\boldsymbol{Q} = [\frac{1}{2}\frac{1}{2}0]$ while its simple cubic counterpart $KNiF_3$ has $\boldsymbol{Q} = [\frac{1}{2}\frac{1}{2}\frac{1}{2}]$ and displays 3D magnetic behaviour [19].

Because of competing contributions to the electrical conductivity from hot and cold regions on the Fermi surface, the intermediate temperature regime is more complex. The standard treatment outlined above can be extended into this region [23]. Here, one finds an extended temperature range in which exponents less than $3/2$ can occur as a cross-over between high and low temperature forms of $\rho(T)$ even for a 3D spin fluctuation spectrum, but it is too early to tell whether or not it can account for all of the features we observe, both in tetragonal and cubic systems, in a consistent way. In particular, the power-law resistivity observed in $CePd_2Si_2$ at p_c extends to nearly 40 K, i.e. roughly 0.5 T_{sf}, and lies far outside the predictive range of the magnetic interaction model in its current form.

Further information about the nature of the strange metallic state can be obtained by tuning $CeNi_2Ge_2$ away from the quantum critical point by means of magnetic field or pressure, both of which are expected to give rise to a re-emergence of Fermi-liquid behaviour below a temperature scale $T_{FL} \ll T_{sf}$. Within the magnetic interaction model, T_{FL} is an indication of the low temperature magnetic correlation length, ξ (T_{FL} increases with decreasing ξ).

An increasing magnetic field produces a rapidly growing Fermi-liquid regime at low temperatures (figure 2). The cross-over into the low temperature T^2 form for $\rho(T)$ broadens at high magnetic fields, but it is surprisingly narrow at the lowest field of 0.5 T and parallels the rapid change to a nearly constant ρ at high temperature (insets of figure 2).

The effect of pressure is less dramatic than that of the magnetic field. We observe a rapid suppression of the absolute resistivity, but only a slow drift of the low temperature power-law exponent towards 2 (figure 3). In $CeNi_2Ge_2$, an exponent close to 2 is observed below $\simeq 1$ K near 1 GPa, while in $CeRu_2Ge_2$, for example, $T_{FL} \simeq 4.5$ K at $\rho - \rho_0 \simeq 1$ GPa [24]. The return to T^2 behaviour is even more rapid in $CeIn_3$ [11].

The slow recovery of a Fermi-liquid $\rho(T)$ with pressure indicates a nearly critical normal state in which unusually large correlation lengths persist over a region in the phase diagram.

This behaviour can be connected to an unexplained phase transition at higher pressures, near and above 1.5 GPa and about 1 K [17, 18, 21] (labelled T_x, in figure 4). $T_x(p)$ extends over a wide range in pressure, up to at least 3 GPa, and is accompanied by superconductivity below 0.4 K (figure 4).

The occurrence of further ordered phases on the high pressure side of the magnetic critical point near $p = 0$ in $CeNi_2Ge_2$ is a challenge to the conventional picture of heavy fermion systems based on the Doniach phase diagram. The speculation [20] that in $CeNi_2Ge_2$ the anomalous low pressure behaviour is a consequence of a critical point associated with the high pressure phase $T_x(p)$ would be inconsistent with our observation of a return to a T^2 resistivity with pressure. By contrast, we suggest that the variation of lattice density with pressure causes a changeover between two competing magnetic order parameters. Before entering the high pressure ordered state, the system is tuned through an intervening region, in which the correlation length can be anomalously large. Because this change is governed by details of the microscopic exchange parameters and of their pressure dependence, we may expect the width of the intervening region to be sample dependent. This situation is reminiscent of similar phase diagrams in alloyed systems, such as $CeRh_2Si_2$ doped with Ru or Pd [25]. $CeNi_2Ge_2$ is to our knowledge the first example of such behaviour in an ordered heavy fermion compound.

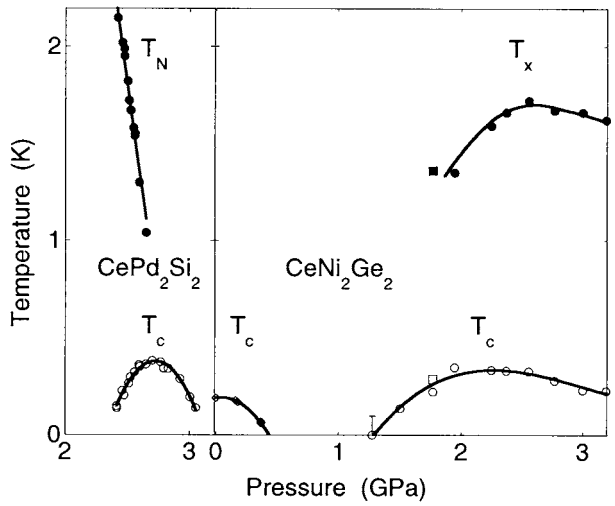

Figure 4. Ordered states in $CeNi_2Ge_2$ as a function of hydrostatic pressure. Transitions were identified from kinks in $\rho(T)$ in the case of T_N and T_x and from the 50% point of superconducting transitions for T_c. Different symbols refer to samples with different ρ_0. The high pressure phase diagram of $CePd_2Si_2$ is given for comparison.

The recovery of Fermi-liquid behaviour with pressure and magnetic field suggests that the unconventional normal state observed in $CeNi_2Ge_2$ and $CePd_2Si_2$ is connected to a magnetic quantum critical point. However, while the magnetic interaction model is in qualitative agreement with our findings on $CeNi_2Ge_2$ and $CePd_2Si_2$ at low temperatures, the striking power-law dependence of the resistivity over a wide range in temperature indicates that an important element in our theoretical picture may still be missing. An intuitive description of this strange metallic state could be based on a more extreme separation of the charge and spin degrees of freedom than is present in current approaches [26]. Convincing evidence is emerging that in at least one other Ce-based heavy fermion system, the $CeCu_{6-x}Au_x$, series, the quantum critical point is accompanied by unexpected local spin dynamics [27]. Within

the standard magnetic interaction model the imaginary part of the wavevector and frequency dependent susceptibility is of the form $\mathrm{Im}\chi(\boldsymbol{q},\omega) \sim g(\boldsymbol{q})\omega^{\alpha}$ at sufficiently low ω (> 0) and T, where g is some function of $\boldsymbol{q}$ and $\alpha = 1$. In the standard model this value of α holds even in the limit $T_N \to 0$ provided $d + z > 4$ [2]. However, in $CeCu_{6-x}Au_x$ it appears that the exponent α is anomalous and significantly less than unity near the antiferromagnetic quantum critical point. This constitutes a breakdown of Fermi-liquid theory of an entirely different order from that expected in any known model. Inelastic neutron scattering on the pure, stoichiometric compound $CeNi_2Ge_2$ could decide whether such behaviour can occur in the absence of disorder and whether it is, ultimately, more widespread in heavy fermion systems on the threshold of magnetism.

We thank, in particular, P Coleman, J Flouquet, P Gegenwart, C Geibel, I Gray, S Kambe, D Khmelnitskii, F Kromer, M Lang, A P Mackenzie, G J McMullan, A J Millis, P Monthoux, C Pfleiderer, A Rosch, A Schofield, G Sparn, F Steglich, S Süllow, O Trovarelli, A Tsvelik and I R Walker. The research has been supported partly by the Cambridge Research Centre in Superconductivity, headed by Y Liang, by the EPSRC of the UK, by the EU, and by the Cambridge Newton Trust.

Note added in proof. The scenario of nearly two-dimensional spin fluctuations discussed above has recently been applied to the new low-temperature antiferromagnet $YbRh_2Si_2$, which displays striking, nearly linear temperature-dependences of the resistivity over a wide range [28]. $YbRh_2Si_2$ appears to be a hole analogue to the tetragonal Ce-systems, but present samples differ from $CePd_2Si_2$ and $CeNi_2Ge_2$ in their lack of superconductivity, in the level of purity and in the impossibility of approaching the critical point by hydrostatic pressure. Taken together, the robust non-Fermi-liquid properties observed in all three stoichiometric compounds pose a serious challenge to our understanding of the metallic state.

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