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2001 J. Phys.: Condens. Matter 13 9335

(http://iopscience.iop.org/0953-8984/13/41/320)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **13** (2001) 9335–9347 PII: S0953-8984(01)23782-5

Further pressure studies around the magnetic instability of $CePd_2Si_2$

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Received 9 April 2001, in final form 6 July 2001
Published 28 September 2001
Online at stacks.iop.org/JPhysCM/13/9335

Abstract
Using helium as a pressure-transmitting medium in a diamond anvil cell enabled a high-quality sample of the antiferromagnet $CePd_2Si_2$ to be studied in a highly hydrostatic environment. With the aid of a low-temperature force-modulation device, ac calorimetric measurements were made up to 25 kbar and resistivity measurements up to 33 kbar, on both sides of P_C, found between 28 and 29.5 kbar. The narrow superconducting domain found around P_C indicates the connection between the magnetic instability and the formation of Cooper pairs. We assert that there is an influence of the residual resistivity both on the emergence of superconductivity, and on the corresponding transition width. Close to P_C, the temperature dependence of the resistivity in the normal state qualitatively agrees with the spin-fluctuation model. While standard Fermi liquid behaviour was found to break down in the immediate vicinity of P_C, it reappeared below 700 mK at 33 kbar, less than 5 kbar above.

1. Introduction

Most Ce-based intermetallic compounds are close to a magnetic instability. By varying an external parameter, such as pressure or chemical composition (chemical pressure), the interaction between the 4f and conduction electrons may be changed and the system tuned through its instability. In the vicinity of the quantum critical point (QCP), the conventional Fermi liquid behaviour tends to disappear, while a superconducting phase is found to emerge [1–6]. Though the mechanism of the Cooper pairing remains unknown, it is commonly believed to be related to magnetic fluctuations [7].

Experimental investigations of the QCP are extremely sensitive to all types of inhomogeneity. In particular, reaching the magnetic instability by doping inevitably induces defects, damaging for the superconductivity. As the physical properties are expected to vary rapidly in the proximity of the QCP, any inhomogeneity will lead to their drastic averaging.

By comparison with alloying, application of pressure on stoichiometric compounds is a much better way to probe their QCP from the magnetic ordered ground state. However, the existence of non-hydrostatic effects such as pressure gradients, residual uniaxial stresses and even stress gradients makes the quantitative analysis considerably more difficult. These effects may be further magnified by the compound's anisotropy.

$CePd_2Si_2$ crystallizes in the $ThCr_2Si_2$ structure (space group $I4/mmm$). At ambient pressure, it orders below the Néel temperature $T_N \simeq 10$ K [8] in a simple antiferromagnetic structure, described as alternating ferromagnetic planes along the $(1, 1, 0)$ direction [9, 10]. The ordered moment of about 0.65 μ_B is smaller than that predicted by crystal-field calculations [11] indicating partial Kondo screening ($T_K \sim 10$ K). High-pressure studies performed on samples with residual resistivities, ρ_0, higher than 10 $\mu\Omega$ cm [12, 13] did not find superconductivity. However, the Cambridge group succeeded [4] in growing high-quality single crystals with ρ_0 down to 1 $\mu\Omega$ cm and a superconducting phase was found close to the critical pressure $P_C \sim 28$ kbar. This aroused new interest in the compound. Later, the Geneva group also produced [5] good samples and confirmed the existence of superconductivity under pressure. The striking difference between the experimental results is as regards the pressure interval in which superconductivity exists. In the former investigation [4], performed up to 32 kbar in the nearly hydrostatic conditions of a piston–cylinder-type cell, superconductivity was observed in the narrow range 22–32 kbar. The latter experiment was performed in a Bridgman anvil cell. This provided less hydrostatic conditions due to the use of a soft solid as the pressure-transmitting medium, but higher pressures could be applied. In the two samples measured in this cell, superconductivity survived over a much broader pressure interval (20–70 kbar) for an estimated P_C of about 35 kbar.

The motivation for this work was to investigate a sample of $CePd_2Si_2$ from Geneva, expected to be similar to that of reference [5], but in the most hydrostatic conditions possible. This was achieved using a diamond anvil cell with solid helium as the pressure-transmitting medium to perform electrical resistivity and AC calorimetric measurements. Thanks to these good experimental conditions, we were able to carefully analyse the antiferromagnetic transition and its evolution under pressure. Furthermore, we extend the previous Cambridge results on the temperature dependence of resistivity in the vicinity of P_C. Finally, being able to reach pressures higher than P_C, we can shed some light on the question of the pressure range in which superconductivity is found.

2. Experimental details

Samples were grown in Geneva using a simple technique of platelet germination from a large melted ingot. The single-crystalline platelets obtained have a length of about 1 mm and the c-axis normal to their surface. After optimized annealing (1200 °C, 48 h), they had a rather low residual resistivity, down to 1 $\mu\Omega$ cm, corresponding to a residual resistivity ratio RRR $\sim$ 47. Further details of the preparation and characterization are given elsewhere [5]. The samples used for the measurements by the Geneva group [5] were cut from one of them (RRR $\sim$ 17). Two more samples, separated by 200 μm, were extracted from another platelet. One of them, as reported elsewhere [6], showed a very low residual resistivity $\rho_0 \sim 1$ $\mu\Omega$ cm at ambient pressure. Pressurized up to 27 kbar in a piston–cylinder cell suitable for high magnetic fields, it provided interesting results that we will refer to in the following. The sample measured in the present work has $\rho_0 \simeq 2$ $\mu\Omega$ cm, suggesting the existence of inhomogeneity on a submillimetre scale within the platelet. The sample dimensions ($200 \times 40 \times 40$ μm^3) were chosen for resistivity measurements in the small pressure chamber of the diamond anvil cell.

The technique of using helium as a pressure-transmitting medium has been recently adapted for transport measurements [14, 15], overcoming the difficulties of introducing wires into the pressure chamber. Helium is known to provide the best possible hydrostatic conditions, with pressure gradients remaining lower than 0.2 kbar even at 200 kbar. Experimentally, this was still found to be the case up to at least 30 kbar, even when the pressure is increased at low temperature, when helium is solid [16]. The pressure was measured by the shift of the fluorescence signal from tiny ruby chips spread in the pressure chamber. The excitation was provided by an argon laser (50 mW) through an optical fibre. The pressure can be varied quasi-continuously *in situ*, owing to a recently developed force-generating device using bellows pressurized by liquid helium [17]. The reduced thermal contact between the bellows and the top of the cryostat allows this device to work at very low temperature ($\sim$100 mK) and to generate forces up to 20 kN, high enough for our experimental pressure range.

The electrical resistivity was measured down to 80 mK by a standard four-point AC technique with lock-in detection at a frequency of 11 Hz. The four 10 μm gold wires, directly spot welded on the sample, ensure good electrical contacts. Though the technique for introducing wires is now reliable, the set-up remains extremely fragile. In spite of the ability of the cell to operate up to 200 kbar, our investigation was limited to 33 kbar because of a wire rupture. Depending on the temperature range, a current of 10 or 100 μA (0.6 or 6 A cm^{-2}) was passed along the *a*-axis. For the measurements in the dilution cryostat, we used a low-temperature transformer able to measure 1 mΩ with a peak–peak noise ratio of 10^{-3}.

Thanks to the possibility of introducing wires into the pressure chamber, we have very recently adapted the AC calorimetric technique to our diamond anvil cell. A detailed description of this technique is given elsewhere [18]. It is based on the measurement of temperature oscillations induced in the sample by alternating the power (using a modulated argon laser) at relatively high frequency ($\sim$1 kHz), by a Au/Au:Fe (0.03%) thermocouple, directly spot welded to the sample to ensure a good thermal contact. Though this technique is currently only qualitative, it is still a powerful tool for exploring the specific heat anomalies associated with phase transitions. In particular, the phase of the thermocouple signal is capable of revealing irreversible phenomena that could be an indication of a first-order transition [19, 20]. Currently, this type of measurement is performed at temperatures above 1.7 K. The method was applied to the same sample as was used for resistivity measurements, providing complementary information. As the sample was initially shaped for resistivity measurements, its volume was not quite sufficient for this calorimetric experiment. Therefore the sample contribution to the overall signal was rather small and only the phase of the signal, more sensitive to the sample heat capacity, yielded a clear anomaly associated with the magnetic transition. Above 23 kbar, we were already unable to detect the whole transition.

3. Results and discussion

Figure 1 shows the $CePd_2Si_2$ pressure–temperature phase diagram obtained by both resistivity and calorimetric measurements. In this figure, we also present all the previously reported results where superconductivity has been observed. It should be remembered that our results and those of reference [5, 6] were obtained on samples of similar origin, different from that of reference [4]. On the other hand, the measurements reported in reference [5] were carried out under less hydrostatic conditions than those of the other studies. In figure 1, one can clearly distinguish two essentially different behaviours of both the magnetic and superconducting parts of the phase diagram. Apart from the lower maximum value of T_C (290 mK and 400 mK in reference [4]), our results are in qualitative agreement with references [4, 6]. The decrease of T_N is rather similar and T_C seems to pass through a maximum at P_C. In contrast, those obtained

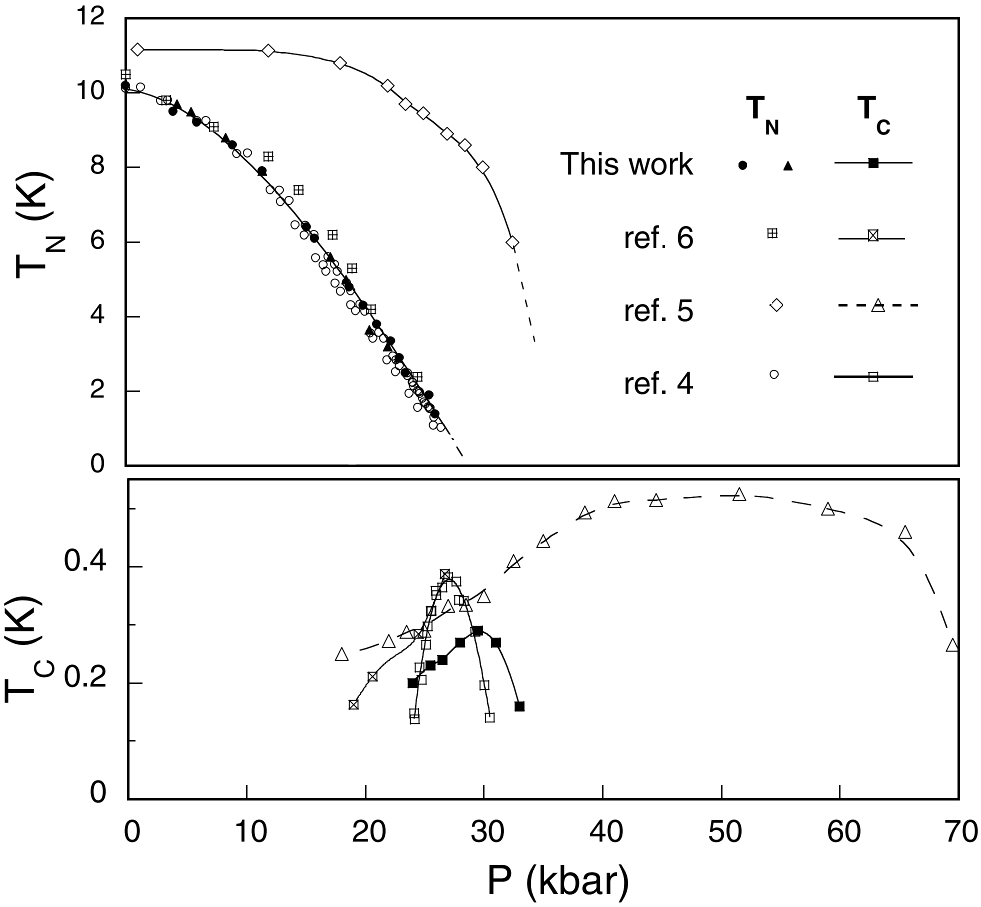

Figure 1. The pressure–temperature phase diagram of $CePd_2Si_2$. The pressure dependence of the magnetic ordering temperature $T_N(P)$ is plotted as • from resistivity measurements and ▲ from calorimetry. The dependence of the superconducting transition temperature $T_C(P)$ is given by ■. The results of reference [6] (⊞ and ⊠), reference [5] (◇ and △) reference [4] (○ and □) for T_N and T_C respectively are shown for comparison. Lines are guides for the eye given in order to help in identifying the antiferromagnetic and superconducting phases in different experiments.

in reference [5] showed phase boundaries markedly skewed towards higher pressures. In order to understand this striking discrepancy, we will give a detailed comparison of our results with the others. In particular, we will concentrate on the antiferromagnetic and superconducting transitions as well as the particularities of the normal state.

3.1. Antiferromagnetic transition

Figures 2(a) and 2(b) show the temperature dependence of the resistivity, $\rho(T)$, and the phase of the calorimetric signal, $\phi(T)$, for selected pressures on both sides of the antiferromagnetic transition. The transition appears as a clear kink in $\rho(T)$. The magnetic ordering temperature T_N is determined by the onset criterion on $d\rho/dT$ versus T curves (figure 3). In calorimetric measurements, T_N is taken at the minimum of $\phi(T)$, which normally coincides with the maximum of the sample specific heat. The values of T_N obtained by the two techniques

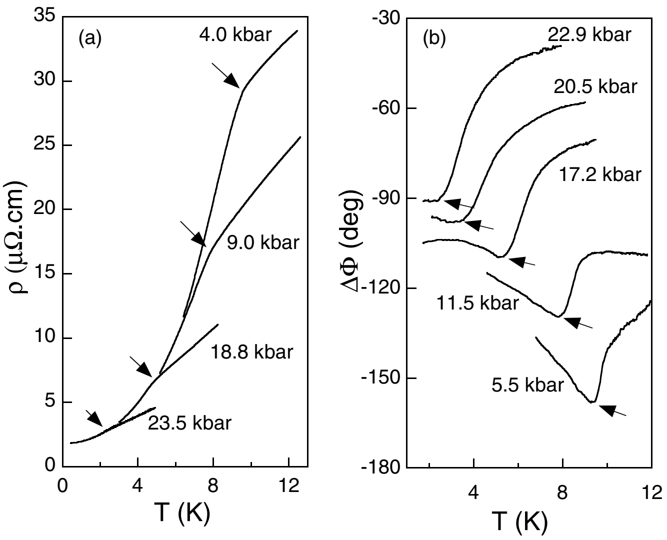

Figure 2. The anomaly associated with the antiferromagnetic transition in the resistivity (a) and the phase of the calorimetric signal (b) for selected pressures. Phase curves are arbitrarily shifted along the vertical axis for clarity. Arrows indicate the position of the magnetic ordering temperature T_N.

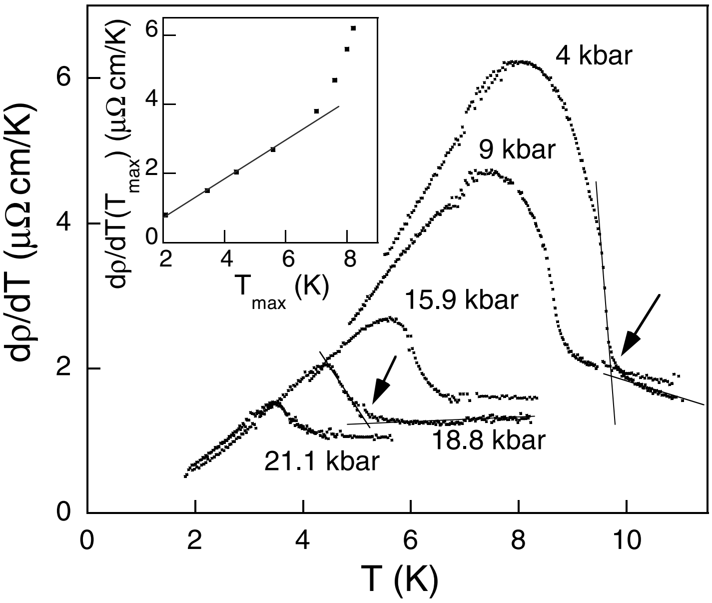

Figure 3. The temperature dependence of $\mathrm{d}\rho/\mathrm{d}T$ for various pressures. The magnetic ordering temperature is determined by an onset method. With increasing pressure, one observes the reduction of the anomaly associated with the magnetic transition as well as its broadening. The inset shows the temperature maximum of $\mathrm{d}\rho/\mathrm{d}T$ (expected to be related to the specific heat) plotted versus T_{max}, the corresponding temperature.

agree well and follow the general behaviour found by the Cambridge group [4] and for the sample of reference [6]. As the pressure is increased, T_N decreases monotonically and finally vanishes at $P_C = 28 \pm 1$ kbar. As previously suggested [4], $T_N(P)$ seems to demonstrate a quasi-linear behaviour above 15 kbar. Spin-fluctuation models describing the vicinity of a QCP [21–24] predict a dependence as $(P_C - P)^{2/d}$ where d is the dimensionality of the magnetic excitation spectrum. The antiferromagnetic structure of $CePd_2Si_2$ is stabilized by the in-plane interactions only [25], regardless of the strength of the magnetic interactions along the c-direction, $J_\perp$. Indeed, the $J_\perp$-contribution to the mean field experienced by any Ce ion cancels out in this structure. This leads to a natural tendency towards two-dimensional behaviour as assumed by Mathur *et al* [4]. Restricting our data to a pressure range close to P_C (15–28 kbar), the best fit is obtained with a critical exponent of 0.9. However, our error bars prevent us from distinguishing an exponent of 1 (2D) from 2/3 (3D). Extending the pressure range to lower pressures reduces the critical exponent determined. This apparent change of regime around 15 kbar might be related to the evolution of the dimensionality or rather to a change from an itinerant magnetism close to P_C to a more localized one at lower pressure. The assumption of a low dimensionality of spin fluctuations will be discussed later.

Both our results and those of reference [6], obtained on samples from Geneva, differ from those obtained earlier in a Bridgman-type pressure cell [5]. This supports not an intrinsic origin for the observed extension in pressure of the magnetic order domain, but rather the influence of pressure conditions. As pointed out by the authors of [5], from thermal expansion measurements [10], the Ehrenfest relation predicts a pronounced anisotropy of $T_N(\sigma)$ with opposite initial slopes for uniaxial stresses σ along the a- and c-axes respectively. As a result, uniaxial stress appearing along the cell axis might be responsible for this particular behaviour of $T_N(P)$.

The negative dip in $\phi(T)$ associated with the jump of the specific heat at the transition indicates the absence of irreversible phenomena and therefore suggests that the antiferromagnetic transition remains of second order up to P_C. Unfortunately, our calorimetric measurements are not quantitative enough for analysing the size of the phase transition anomaly, which is related to the square of the sublattice magnetization. Another way to get information about the specific heat C is via $\mathrm{d}\rho/\mathrm{d}T$ as has been proposed for a mean-field transition [26, 27]. Approaching the QCP, it has been shown that the critical fluctuations develop in a reduced temperature region around the magnetic transition with the result that a mean-field description of the transition is expected to be increasingly relevant [21, 28]. Continentino [24] describes the evolution of the specific heat approaching the instability. He predicts a crossover from a Gaussian to a classical mean-field behaviour. Reflecting this change of regime, $C(T_N)$ is expected to pass from a $T_N^{3/2}$-behaviour (for 3D) at low pressure (high T_N) to a linear dependence when approaching the QCP from the ordered antiferromagnetic phase. The inset of figure 3 shows the evolution of the maximum of $\mathrm{d}\rho/\mathrm{d}T$ at the temperature T_{max} versus T_{max} itself, which is roughly proportional to T_N. The quasi-linear dependence at low T_{max} shows that the mean-field regime extends up to $T_N = 6$ K, corresponding to $P = 15$ kbar. This behaviour is also found in the pressure dependence of the anomaly's size $\Delta(\mathrm{d}\rho/\mathrm{d}T)$. Zülicke and Millis [28] predict a mean-field anomaly of the specific heat $\Delta C \propto T_N^{(d-1)}$. The linear dependence of $\Delta(\mathrm{d}\rho/\mathrm{d}T)$ versus T_{max} supports the idea of a dimension close to 2.

Another striking effect apparently associated with proximity to the QCP is the smearing of the magnetic transition, visible in both resistivity and calorimetric measurements. At low pressure, the sharp increase in $\mathrm{d}\rho/\mathrm{d}T$ at T_N is quite distinct from the maximum corresponding to the inflection point of $\rho(T)$ (see figure 3). On approaching the QCP, the transition becomes broader and finally appears as a smooth increase from the transition onset up to the maximum. A similar broadening of the anomaly of $\phi(T)$ is observed (figure 2(b)). Usually, such transition smearing is attributed to the presence of pressure gradients. In our particular case, the gradients in solid helium over this pressure range are negligible and do not account for the smearing

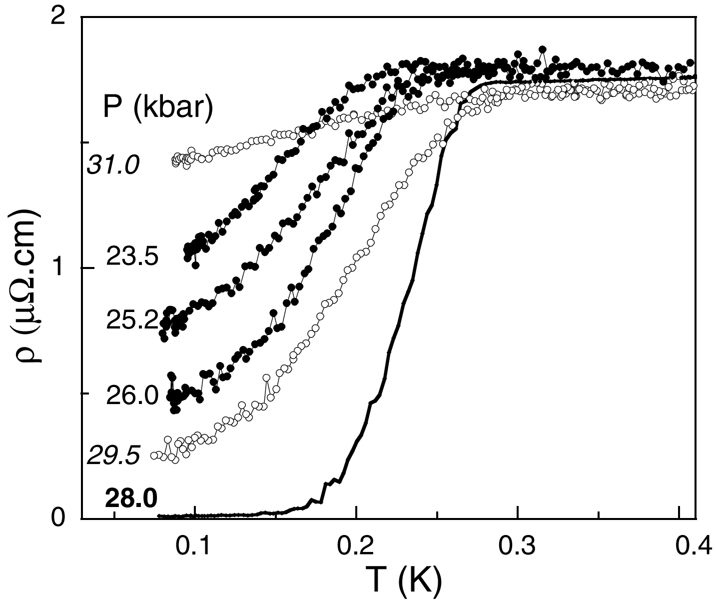

Figure 4. The superconducting transition observed in the resistivity measurements. The superconducting temperature T_C is taken at the transition onset. Full and open circles indicate pressures below and above P_C respectively.

of 0.5 K around 20 kbar. We believe rather that it is a disorder effect amplified by an increased magnetic coherence length approaching the QCP [32]. Understanding of the precise mechanism involved in the magnetic transition broadening requires further information from microscopic probes.

3.2. Superconducting phase

Superconductivity reported by the Cambridge group appeared between 22 and 32 kbar with T_c^{onset} reaching about 400 mK at 28 kbar, which roughly corresponds to the critical pressure [4]. The transition width was found to vary with pressure, and the resistivity actually vanished only close to P_C. Later, the investigation performed in a Bridgman-type pressure cell revealed that superconductivity extended from 18 to 70 kbar with a relatively high and broad maximum of T_C close to 500 mK [5]. In that case, transitions were not only broadened but always partial. For the pressure range corresponding to the highest T_C, the resistivity never fell to zero but instead saturated at a finite value. Experiments carried out on the sample cut, in the immediate vicinity of ours, from the same platelet showed a continuous development of superconductivity from 19 kbar to 27 kbar, the experimental pressure limit [6]. At 19 kbar, the transition was rather broad and incomplete even at 50 mK. The critical temperature was found to increase with pressure, reaching 390 mK at 27 kbar, while the transition became sharper, with a width of 40 mK.

One of the main features of our work was the ability to investigate both sides of P_C. In figure 4, we present the temperature dependence of the resistivity over the pressure range where superconductivity occurs. Most of the transitions that we have observed were incomplete at 80 mK and sensitive to the current density. The resistivity at the lowest temperature (80 mK) dropped by 50% when reducing the current density from 6 to 0.6 A cm^{-2}. Below this value, the noise level became too high for reliable measurements. Our study revealed the emergence of superconductivity at 23.5 kbar where the resistivity dropped by 35% between 200 mK and 80 mK. As the pressure increases, the superconductivity is reinforced, achieving higher T_C and a steeper drop in resistivity. Close to P_C, at 28 kbar, the transition is complete, with an onset of 270 mK and a width of 100 mK. Above this pressure, the transition becomes broad again while T_C seems to pass through a maximum of 290 mK at 29.5 kbar and to vanish at higher pressure. The superconductivity seems to disappear above 33 kbar, where the resistivity fell by only 5% (not shown in figure 4 for clarity).

This behaviour is in apparent contradiction with the Geneva results where superconductivity was found to survive at pressures far above the extrapolated P_C. Pressure gradients alone do not seem sufficient to produce such an extension of the superconducting domain away from the QCP. An explanation could be the existence, at higher pressures, of another mechanism of Cooper pair formation, not directly related to the magnetic QCP. The large domain of superconductivity observed in reference [5] would result from the union of two distinct phases induced by the non-hydrostatic conditions of the Bridgman pressure cell. This scenario of two mechanisms for the superconductivity has already been proposed to explain the large superconducting domain in $CeCu_2Si_2$ and $CeCu_2Ge_2$ [32]. Another possibility is that the QCP has been completely washed out under non-hydrostatic conditions, leading to the large domain of superconductivity. Further measurements at higher and still hydrostatic pressure are required to confirm this hypothesis.

Comparison between our work and reference [6] reveals an apparent correlation between the residual resistivity, ρ_0, and superconductivity, as already pointed out in reference [6]. The temperature dependence of resistivity at ambient pressure, as well as close to P_C (figure 7, later), suggests that the difference between $\rho(T)$ in these two studies is essentially given by an additive term (Matthiessen type) included in ρ_0. Taking into account the T_C-values and transition widths, we are tempted to make a link between the superconducting coherence length, ξ_s, and the electronic mean free path, ℓ_e. The exact origin of the superconducting phase, and its symmetry, are currently unknown. As it is commonly believed that the superconductivity in most heavy-fermion systems is non-conventional, the dirty limit ($\xi_s > \ell_e$) in that case could be destructive for superconductivity.

Close to the critical pressure, T_C reaches its highest value, suggesting that ξ_s is at a minimum. The upper-critical-field measurements reported in reference [6] lead to an estimate of ξ_s of 100 Å at P_C. One would not expect an emergence of superconductivity when the mean free path is lower than this value, which corresponds to a residual resistivity of about 10 $\mu\Omega$ cm. With a mean free path of 500 Å in our case, the clean limit is reached at P_C and thus the superconductivity is fully developed. As the pressure is varied away from P_C, ξ_s increases and gets dangerously close to ℓ_e. At this moment, inhomogeneities or internal stresses due to defects may easily result in a coexistence of normal and superconducting domains, which could explain the strong smearing of the transition and even finite resistivity below it. In order to confirm such a scenario, microscopic measurements sensitive to the whole volume of the sample (e.g. NMR, x-ray and neutron diffraction, specific heat) around the QCP should be developed.

3.3. Normal-state resistivity behaviour at low temperature

At low pressure, where the ground state is still magnetically ordered, a rough temperature dependence of resistivity well below T_N is described by $\rho = \rho_0 + AT^2 + \rho_{sw}$. Here, the second term is due to scattering between the heavy quasi-particles, and ρ_{sw} is the contribution of spin waves, which depends essentially on their excitation spectrum. Usually, this last term

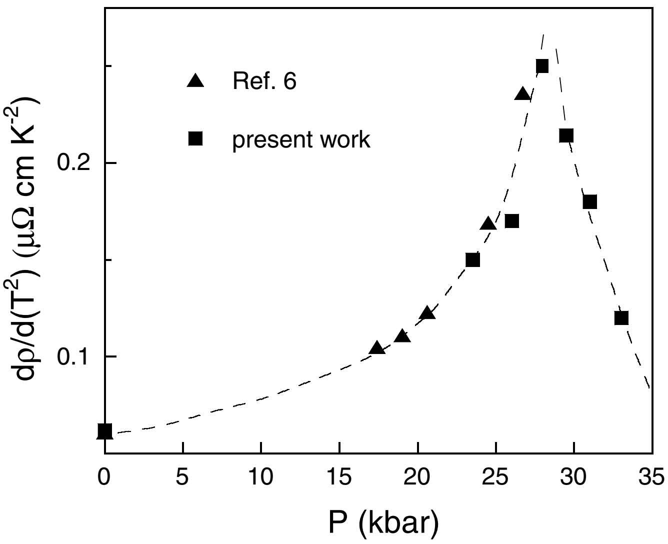

Figure 5. Evolution of the Fermi liquid T^2-term of the resistivity illustrated by the quantity $d\rho/d(T^2)$ evaluated in the low-temperature range 300–700 mK just above the superconducting temperature.

leads to a temperature dependence stronger than the common Fermi liquid behaviour, T^2. In previous works [5, 6], a dependence $\rho_{sw} = BT(1 + 2T/\Delta)\exp(-\Delta/T)$ corresponding to a gapped antiferromagnetic spectrum has been proposed. We decided to analyse our data in two easier ways. First of all, the pressure dependence of the A-term can be estimated by $d\rho/d(T^2)$ taken over a fixed temperature range 300–700 mK well below $\Delta \simeq 14$ K. The lower boundary corresponds to the highest observed T_C and the interval width is sufficiently large for performing a reliable fit. Secondly, approaching the QCP, the local temperature dependence of resistivity is often described by an unusual power law, $\rho = \rho_0 + BT^n$. When considered over a large temperature range, both ρ_0 and n are weakly temperature dependent. It might therefore be useful to extract the temperature dependence of the effective exponent $\eta = d\ln(\rho - \rho_0^*)/d\ln T$, where ρ_0^* is extrapolated from a power-law fit just above the superconducting transition.

Figure 5 illustrates the evolution with pressure of the Fermi liquid (FL) coefficient, A, including measurements from [6]. Its ambient-pressure value of 0.06 $\mu\Omega$ cm K^{-2} is compatible with previous measurements and agrees with the linear term of the specific heat $C/T \sim 100$ mJ mol^{-1} K^{-2} [11, 33]. When approaching the QCP, spin fluctuations contribute to quasi-particle scattering and thus enhance A. In the limit $P \to P_C$, spin-fluctuation theory [22] predicts the divergence of this quasi-particle coefficient. As, close to P_C, this Fermi liquid regime only exists at very low temperature, the quantity $d\rho/d(T^2)$, calculated over a finite temperature range distant from zero, deviates from A. Nevertheless, it clearly reveals the underlying tendency. A sharp increase of A by a factor of four compared to its ambient-pressure value confirms the critical pressure to be around 28 kbar. A small pressure discrepancy of 1 kbar with reference [6] might be related to calibration problems between the spectrometer used for the ruby fluorescence and the superconducting manometer in that experiment.

In figure 6(a), the temperature dependence of $\eta = d\ln(\rho - \rho_0^*)/d\ln T$, the effective exponent, is shown in the vicinity of P_C while figure 6(b) shows the evolution of η just above T_C and at $T = 2$ K. For $P < P_C$ (23.5 and 26 kbar) and low temperature, the presence of

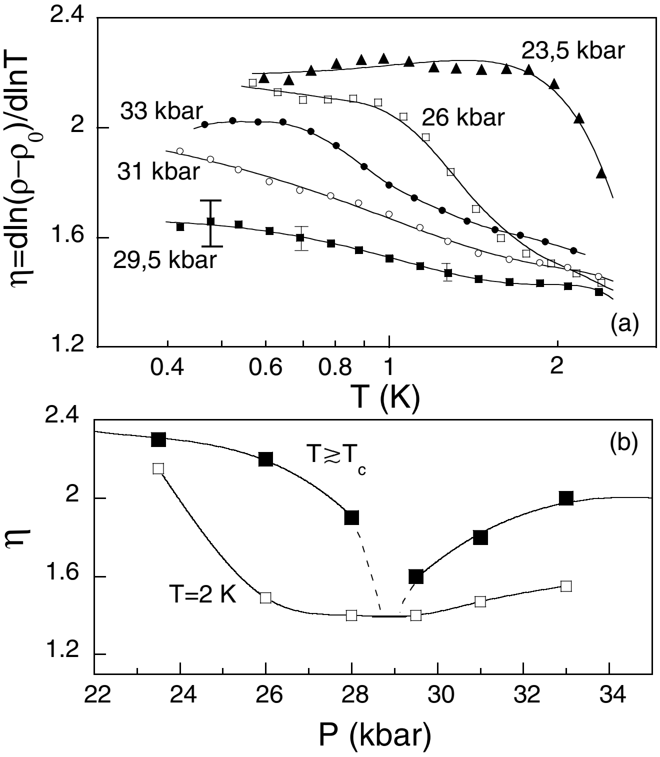

Figure 6. (a) The temperature dependence of the effective exponent $\eta = \mathrm{d}\ln(\rho - \rho_0^*)/\mathrm{d}\ln T$ at pressures close to P_C. ρ_0^* is determined by the extrapolation of a power-law fit in the low-temperature range 300–700 mK. The error bars (similar for all the curves) included errors on ρ_0 and in calculation of η. The curve obtained at 28 kbar is similar to that obtained at 31 kbar (not shown here for clarity). (b) The pressure dependence of η at low temperature (just above T_C) and at $T = 2$ K. Lines are guides for the eye reflecting the probable evolution.

spin waves below T_N gives rise to $\eta > 2$. When approaching P_C at 28 kbar (not shown in figure 6(a) for clarity) and 29.5 kbar, as the temperature decreases, the exponent rises smoothly from a value close to 1.3. As shown in more detail in figure 7, the quasi-linear dependence of ρ on $T^{1.3}$ at high temperature (restricted here to 4 K) may suggest that the $T^{1.3}$-law is extended down to T_C at P_C situated between 28 and 29.5 kbar; this seems to be the case in the Cambridge results ($\eta = 1.2$ is found instead of 1.3) [4] as well as in the Geneva and Grenoble results [5, 6]. Our speculation is that a deep minimum in η will occur at P_C, a pressure at which this exponent is very robust in temperature. At higher pressure, the low-temperature exponent increases, again indicating a tendency of the quasi-particles to reappear. At 33 kbar, our highest pressure investigated, the Fermi liquid behaviour is stabilized below 700 mK. The general evolution of η illustrated in figure 6(b) emphasizes its strong pressure dependence and therefore the importance of working under hydrostatic conditions. The pressure representation of η at higher temperature (i.e. 2 K) is quite different with no clear minimum and basically two plateaus of $\eta > 2$ at low pressure (magnetically ordered) and $\eta \simeq 1.3$ on the paramagnetic side. The depth of the exponent dip at P_C will be determined more precisely in future. Let us stress the main difference between a very low-temperature scan with the observation of a deep minimum of η at P_C and the smearing out of the magnetic anomalies as $T_N \to 0$.

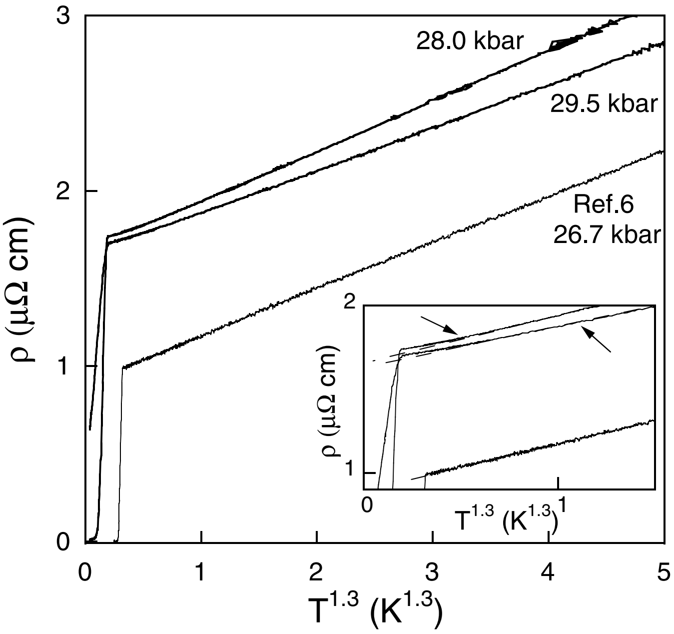

Figure 7. Temperature dependencies of the resistivity for pressures enclosing P_C. The inset magnifies the low-temperature region. Dashed lines illustrate the $T^{1.3}$-dependence observed over most of our experimental temperature range and its deviation in the limit of low temperature.

So-called Non-Fermic liquid (NFL) behaviour has stimulated many theoretical and experimental investigations. Usually spin-fluctuation theory [22] predicts a dependence such as $\rho \propto T^{d/2}$, where d is the dimensionality of the spin-fluctuation spectrum for a nearly itinerant antiferromagnet. This dependence arises from the scattering of carriers located on the Fermi surface regions separated by the antiferromagnetic ordering wave vector. Thus one would expect to observe the exponent $3/2$ for a common 3D magnetic system. However, this NFL temperature dependence should only appear at low temperature, where the short-circuit by the rest of the Fermi surface is suppressed by impurities, as suggested by Rosch [36]. A temperature crossover was predicted, where the exponent could reach a value close to 1 before reaching this NFL regime. Taking into account a small amount of isotropic disorder, a relatively steady exponent of 1.2 over almost two decades in temperature, observed in reference [4], was reproduced numerically. However, this model of the NFL behaviour seems inconsistent with two results: (i) three samples from Geneva having different residual resistivities show the same exponent and (ii) the sample from Cambridge shows a different exponent for a similar ρ_0.

Another way of explaining a low NFL exponent is again to evoke a pronounced anisotropy of the spin-fluctuation spectrum, which would lead to an effective dimensionality close to 2. There are arguments both for and against this idea. Previous neutron scattering measurements performed at ambient pressure [10] show a weak anisotropy of the magnetic excitation spectrum. The ratio of the exchange interactions along the main diagonal of the unit cell and in the basal plane is calculated to be close to $1/3$. In comparison, this ratio reaches 10^{-3}–10^{-4} in well known quasi-2D magnetic systems [38]. The collapse of this ratio with increasing pressure in $CePd_2Si_2$ is rather surprising. However, the apparent quasi-linear pressure dependence of T_N in the vicinity of P_C along with the particular NFL behaviour of the resistivity are sufficient to make this possibility worth seriously considering. Experimental limitations related to the pressure technique currently prevent us from studying the anisotropy of the spin-fluctuation spectrum. The isoelectronic compound $CeNi_2Ge_2$, with a unit-cell volume smaller than that of $CePd_2Si_2$, can be considered as its equivalent at P_C. At ambient pressure, this system already shows a superconducting phase followed by NFL behaviour of resistivity at higher temperature. Recent inelastic neutron scattering measurements [39] revealed that the magnetic correlations have a quasi-2D character. These results, obtained on a very similar system, imply that the existence of such features is possible in $CePd_2Si_2$.

The upper temperature limit of our experiment prevented us from checking the persistence of NFL temperature power laws of resistivity observed up to 40 K [4–6], a temperature which seems significantly high compared to the expected characteristic energy T_K.

4. Conclusions

The suppression of the antiferromagnetic phase by hydrostatic pressure at $P_C \simeq 28$ kbar was carefully investigated by electrical resistivity and calorimetric measurements. We have observed the emergence of superconductivity over a pressure range of 10 kbar centred at P_C, which could be an indication that superconductivity and magnetic instability are correlated phenomena. Both the superconducting critical temperature and the transition width were found to be strongly pressure dependent. The temperature dependence of the resistivity in the normal state was found to be in agreement with spin-fluctuation theory. At 26 kbar, i.e. less than 3 kbar below P_C, the low temperature $\rho(T)$ is correctly described by a power law with an exponent higher than 2 resulting from a combination of quasi-particle and spin-wave scattering. At 33 kbar, i.e. less than 5 kbar above P_C, Fermi liquid behaviour reappears below 700 mK. At 28 and 29.5 kbar, a $T^{1.3}$-power law was found to develop down to below 1 K. However, at lower temperature, an increase in the T-exponent related to the proximity of the Fermi liquid was detected. This indicates a strong pressure dependence of the normal-state resistivity when the pressure is varied from P_C, located between 28.0 and 29.5 kbar. Because solid helium used as the pressure-transmitting medium provides the most hydrostatic conditions possible, we assume that the mechanisms responsible for the broadenings of magnetic and superconducting transitions apparently related to the proximity to the QCP are correlated with the amount of disorder in the sample, i.e. the residual resistivity ρ_0. This assumption is supported by comparison with the results obtained on a sample cut from the same single crystal with half the ρ_0 [6]. The observation of a wide pressure domain of superconductivity on samples cut from another single crystal from the same initial ingot remain currently unexplained. While the first reaction is to attribute this odd result to non-ideal pressure conditions, our results show that disorder and impurity of any kind may have an unexpected influence. In particular, one cannot exclude a tiny dispersion of the chemical composition in the different single crystals.

Owing to a previous metallurgical effort to reduce the residual resistivity, $CePd_2Si_2$ is a suitable system in which to study the correlation between magnetism and superconductivity. Our results, obtained under the ideal pressure conditions of solid helium, show clearly that sample quality remains the limiting factor for the emergence of superconductivity. A deep and systematic study of the chemical composition is necessary to fully understand the physics around the critical point. The combination of a quasi-hydrostatic pressure with the ability to change it quasi-continuously *in situ* will allow us to perform a more detailed investigation of both the superconducting domain and non-Fermi liquid behaviour of the resistivity in the future. Finally, because the diamond anvil cell allows hydrostatic conditions to be combined with high pressures, the existence of superconductivity should be checked over a larger pressure range, at least up to 100 kbar.

Acknowledgments

We thank K Miyake, M A Continentino and I Fomin for helpful discussions and F M Grosche for allowing us to include his results.

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