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LETTER TO THE EDITOR

Strain enhancement of superconductivity in $CePd_2Si_2$ under pressure

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Abstract
We report resistivity and calorimetric measurements on two single crystals of $CePd_2Si_2$ pressurized up to 7.4 GPa. A weak uniaxial stress induced in the pressure cell demonstrates the sensitivity of the physics to anisotropy. Stress applied along the c-axis extends the whole phase diagram to higher pressures and enhances the superconducting phase emerging around the magnetic instability, with a 40% increase of the maximum superconducting temperature, T_c, and a doubled pressure range. Calorimetric measurements demonstrate the bulk nature of the superconductivity.

By varying an external control parameter, such as magnetic field, composition or pressure, many heavy-fermion systems may be pushed through a quantum critical point (QCP), where their magnetic ordering temperature goes to zero. In the immediate vicinity of this point, transport and thermodynamic measurements show striking deviations from standard Fermi-liquid behaviour [1–7]. In particular, the low-temperature resistivity, $\rho(T)$, exhibits a T^n-behaviour with $1 < n \leqslant 1.5$ over a wide temperature range. The nature of this non-Fermi-liquid (NFL) behaviour remains an open question [8]. Is the spin-fluctuation description [1] appropriate, with itinerant magnetism developing below a characteristic temperature such as the Kondo temperature, T_K? Alternatively, this characteristic temperature may collapse at the QCP, leading to localized magnetism down to the lowest temperatures [2, 8]. Particular attention has been paid to the case of stoichiometric compounds, whose weak disorder permits the observation of superconductivity around the magnetic instability. Due to the enhancement of low-lying magnetic excitations in this region, it is commonly believed that Cooper pairs are formed magnetically. In one of these systems, $CePd_2Si_2$, superconductivity was discovered in a window of about 1 GPa around the QCP at a critical pressure $P_c \simeq 2.8$ GPa. Simultaneously, NFL behaviour was found in resistivity measurements with a $T^{1.2-1.3}$-law over two decades in temperature [4–7]. As an exponent $d/2$ is predicted for a d-dimensional antiferromagnet by spin-fluctuation theory [9–11], it has been suggested that the magnetic excitation spectrum has an effective dimension close to 2. This assumption is supported by the quasi-linear pressure dependence of the Néel temperature, T_N, predicted to be $(P_c - P)^{2/d}$, by the tetragonal symmetry ($I4/mmm$) and by the magnetic structure containing a frustrated moment in the centre of the elementary cell [4].

The results quoted above were obtained in hydrostatic conditions using a 'liquid' pressure-transmitting medium. Another investigation [7] was carried out in a Bridgman anvil cell, using a soft solid (steatite) as a pressure-transmitting medium [12]. This showed a rather different phase diagram: around a higher critical pressure $P_c \simeq 3.6$ GPa, a strikingly expanded superconducting region was found, lying from 2 to 7 GPa with a maximum of T_c apparently disconnected from P_c, casting doubt upon spin fluctuations as the only mediation mechanism for superconductivity. As this pressure technique is suspected to provide higher pressure gradients, a residual stress along the cell axis could be at the origin of these differences. Our motivation was thus to demonstrate and understand the effect of uniaxial stress under pressure.

In this letter, we report resistivity and calorimetric measurements performed on two samples in a Bridgman anvil cell up to 7.4 GPa. The samples were set in the pressure cell with the force load direction perpendicular and parallel to the c-axis (see [7] for the latter). These measurements demonstrate the high sensitivity of the physics in $CePd_2Si_2$ to pressure conditions, and the crucial influence of anisotropy on the emerging superconductivity. The differences between the previously observed phase diagrams can be explained by the results from these two samples, with an enhancement of superconductivity when uniaxial stress is applied along the c-axis. Calorimetric measurements demonstrate the bulk nature of this superconductivity; a combination of the two types of measurement leads to further insight into the QCP and its associated energy scale.

The samples were extracted from the same single-crystalline platelet as was used in [5–7]. A parallelepiped sample ($510 \times 75 \times 60\ \mu m^3$), with a residual resistivity ratio RRR $\simeq 62$, was cut into two pieces of length 250 μm. These were polished to a small cross-section ($\sim 70 \times 20\ \mu m^2$), and spot-welded with 5 μm diameter gold wires, giving $\rho(293\ K) = 45\ \mu\Omega$ cm to within 10% for both samples and RRR values of 48 and 103. The corresponding residual resistivities, ρ_0, were respectively 1 and 0.48 $\mu\Omega$ cm. The samples will be referred to as $\parallel$ (higher ρ_0) and $\perp$ (lower ρ_0) in relation to the orientation of their c-axis with respect to the force load direction (and the additive uniaxial stress). Both samples were connected for four-point DC resistivity measurements, with sample $\perp$ having additional connections for a constantan resistive heater and a Au/Au-0.07 at.% thermocouple Fe suitable for AC calorimetric measurements [13]. The pressure was determined by the superconducting transition of a lead manometer.

Sample $\perp$ gave rise to a phase diagram similar to that obtained in hydrostatic conditions (figure 1). The superconductivity was limited to the range 2.14–3.25 GPa around $P_{c\perp} \simeq 2.7$ GPa, with $T_{c\perp}$ having a maximum of 375 mK (mid-point criterion). In contrast, the phase diagram of sample $\parallel$ seems to be stretched towards higher pressures. T_N collapses at $P_{c\parallel} = 3.9$ GPa with a critical behaviour $(P - P_c)^\alpha$, $\alpha = 0.60 \pm 0.05$, as distinct from the quasi-linear dependence in the hydrostatic case. Superconductivity occurred between 2.14 and 5.0 GPa (using a mid-point criterion). As in the previous investigation in a Bridgman cell [7], T_c reached a higher value, 520 mK in our case. This maximum of T_c coincides with P_c, suggesting that this extended superconductivity is still related to the QCP. The apparent discrepancy between P_c and the maximum of T_c in [7] can be explained by the criterion chosen (onset), sensitive to the large transition widths at extreme pressures.

Figure 2 shows $\rho(T)$ curves from sample $\parallel$ for selected pressures with a phononic linear contribution ($0.1\ T\ \mu\Omega\ cm^{-1}\ K^{-1}$) subtracted. The first pressure, 0.1 GPa, corresponds mainly to a small uniaxial stress along the force load direction. With increasing temperature, one can distinguish a clear kink at $T_N \simeq 11$ K, a maximum at T_{max} attributed to the Kondo effect and a 'shoulder' reflecting the influence of excited crystal-field (CF) levels. At high temperature, the

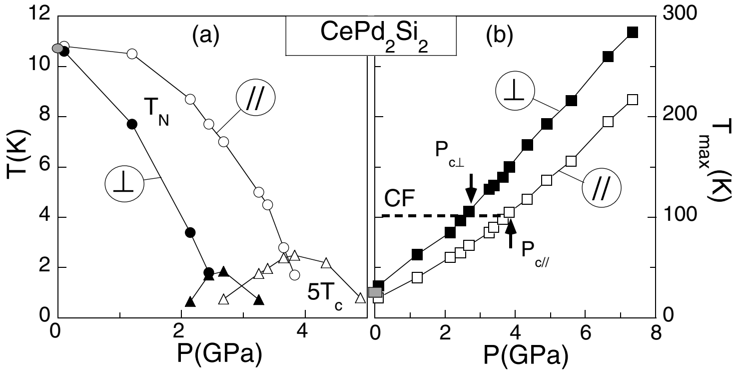

Figure 1. (a) The phase diagram of the two samples (filled and open symbols for samples $\perp$ and $\parallel$ respectively) pressurized as described in the text. (b) The pressure dependence of $T_{\max}$, the temperature of the maximum in the magnetic part of $\rho(T)$. The dashed line, which qualitatively indicates the position of the CF contribution, crosses $T_{\max}$ close to P_c for both samples. Grey symbols indicate values at $P = 0$ GPa.

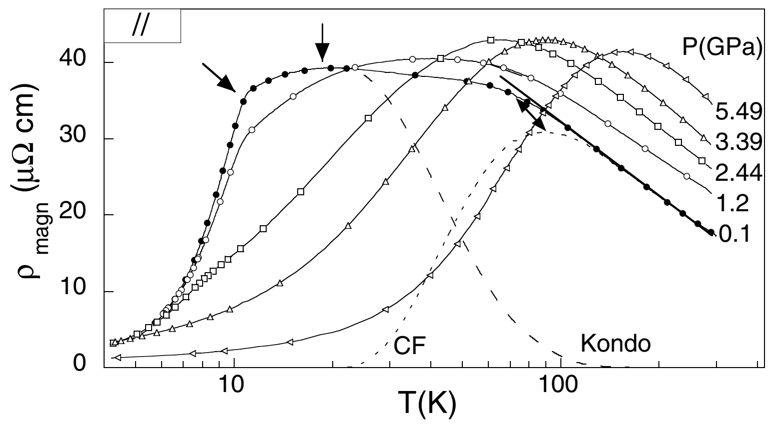

Figure 2. The magnetic contribution to $\rho(T)$ of sample $\parallel$ at selected pressures after subtraction of a linear term assumed for phonons. On the $P = 0.1$ GPa curve, the arrows indicate T_N, the Kondo peak at $T_{\max}$ and a shoulder attributed to the two excited CF levels. The straight line shows the $-\ln T$ Kondo dependence at high temperature. Dashed curves qualitatively represent the Kondo and CF contributions.

$-\ln T$ dependence is characteristic of Kondo scattering. As the pressure rises, $T_{\max}$ increases continuously whereas the excited CF anomaly is rather pressure independent. This latter progressively merges with the Kondo peak around 1.5 GPa and seems to collapse in amplitude at higher pressures where CF excitations cease to be well defined. $T_{\max}(P)$ shows no anomaly at P_c and identical values of $T_{\max}(P_c)$ for the two samples.

The resistivity was analysed at low temperature in terms of a power law $\rho(T) = \rho_0 + AT^n$. Such dependences are not stable over a wide temperature range except for pressures close to P_c, where these power laws extend up to 30 K. The fits were therefore limited to a window of 0.5–2 K, in order to compare data over the entire pressure range. Figure 3 shows the pressure dependence of the coefficient A and the exponent n (inset). A behaves as $(\mathrm{d}\rho/\mathrm{d}T^2)_{T\to 0}$ and may be interpreted as a Fermi-liquid contribution prefactor. As expected from the spin-

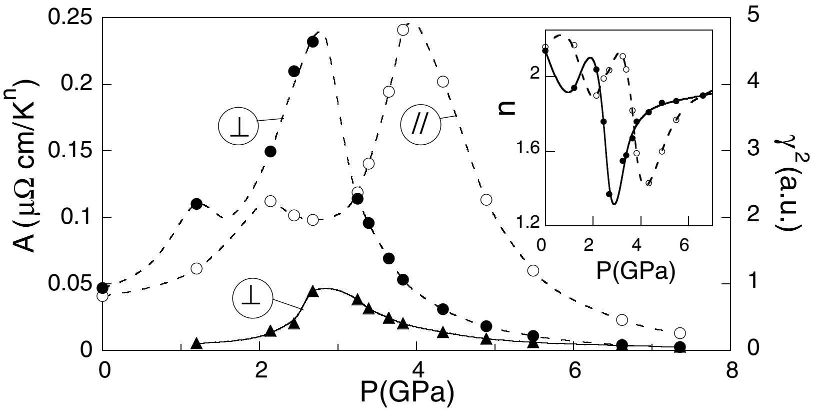

Figure 3. The A-coefficient (circles) and temperature exponent n (inset) of the low-temperature resistivity $\rho = \rho_0 + AT^n$ for both samples. Open and filled symbols indicate $\parallel$ and $\perp$ samples respectively. Filled triangles show γ^2, estimated for sample $\perp$ (see the text), normalized at 7.4 GPa to the value of A at the same pressure. Curves are guides for the eye.

fluctuation model [10], $A(P)$ shows a sharp maximum at P_c. This maximum is similar for the two samples with $A(P_c)/A(0) \simeq 5$. At 7.4 GPa, the A-coefficient of sample $\perp$ has fallen by a factor of 100 compared to its value at P_c. A small anomaly was found in $A(P)$ at about 1 GPa in sample $\perp$ and 2 GPa in sample $\parallel$, possibly corresponding to a pressure-induced magnetic phase transition. Such an anomaly seems to be present in the isoelectronic compound $CePd_2Ge_2$ at about 12 GPa just below $P_c \simeq 13.8$ GPa [14]. An additional curve in figure 3 shows the pressure dependence of $\gamma^2 = (C/T)^2_{T\to 0}$ in sample $\perp$; this also has a maximum at P_c. For each pressure, γ^2 was estimated at 100 mK by subtraction of $1/V^2$ taken at two frequencies (16 and 256 Hz) where V is the thermocouple voltage amplitude [13]. To obtain a reliable pressure dependence, the same working parameters were used for all pressures. Far from the instability, A and γ^2, both related to the square of the effective mass of quasi-particles, m^{*2}, are expected to follow the Kadowaki–Woods relation, $A \propto \gamma^2$ [15]. The peak in γ^2 at P_c is less pronounced than in $A(P)$ with $\gamma^2(P_c)/\gamma^2(7.4\ \text{GPa}) \simeq 18$, but γ may well include a contribution from the pressure-transmitting medium, reducing the relative size of the peak. For both samples, a sharp dip in the resistivity exponent $n(P)$ (inset of figure 3) is associated with the magnetic instability, reaching values lower than the 2 expected for Fermi-liquid behaviour. The minimum values obtained were 1.32 and 1.42 (± 0.03) for samples $\perp$ and $\parallel$ respectively. As in the $A(P)$ curves, a small anomaly appears around 1 and 2 GPa.

The superconducting transition appears in the calorimetric measurement only at 2.68 GPa, the pressure closest to P_c. Figure 4 shows a comparison between superconducting transitions in $\rho(T)$ and the calorimetric signal $1/V$ ($\sim C_p/T$). The onset of the calorimetric transition occurs at the temperature for which ρ reaches zero. In sample $\perp$ at 2.68 GPa $\simeq P_c$, we studied the effect of an external magnetic field on the superconductivity using the two types of measurement. The large initial slope of the upper critical magnetic field in the basal plane, $dH_c^a/dT \simeq -6$ T K^{-1}, indicates that heavy quasi-particles are involved in superconductivity. The size of the calorimetric anomaly collapses rapidly with increasing field and becomes undetectable above 0.5 T. If the calorimetric anomaly at $H = 0$ indicates a bulk transition, one cannot rule out the magnetic field revealing a non-homogeneous situation in the sample, as suggested by the large transition widths in $\rho(T)$ and disappearance of the anomaly in calorimetric measurement for pressures away from P_c. The inset in figure 4 shows the

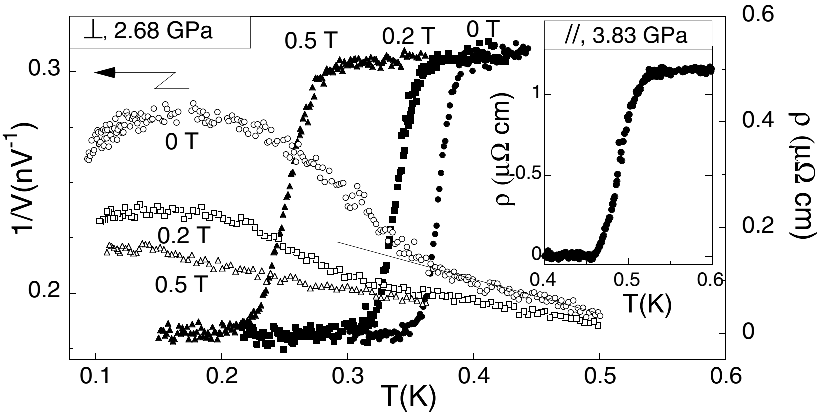

Figure 4. The effect of magnetic field on the resistive and calorimetric superconducting transitions (filled and open symbols) for sample ⊥ close to P_c. The inset shows the superconducting transition in $\rho(T)$ of sample ∥ close its P_c.

superconducting transition in $\rho(T)$ for sample ∥ close to its P_c. A striking point is that the highest value of T_c is obtained for the sample with the larger residual resistivity, showing that the superconductivity enhancement is not limited by the crystal purity.

The following discussion is supported by the unprecedented quality of our samples, with RRR values as high as 130 at P_c. Bearing this in mind, one should be aware that variations on a submillimetric scale exist even within a single crystal—the present samples and those of [5, 6] were cut from the same tiny single-crystalline platelet with RRR values varying by a factor of 3 at $P = 0$. Furthermore, we claim to have an accurate value for the resistivity, with a well-defined geometric factor enabling the determination of the absolute resistivity to within 10%. As suggested earlier, the temperature T_{max} of the maximum in the magnetic contribution to the resistivity should be related to the Kondo temperature, T_K. T_{max} takes the same value at P_c for the two samples, supporting the idea that T_{max} is a reliable characteristic energy for the QCP. Its pressure dependence allows us to take part in the heated debate on the nature of the QCP illustrated by another heavy-fermion system, $CeCu_{6-x}Au_x$. While neutron measurements on the substituted compound $CeCu_{5.9}Au_{0.1}$ seem to reveal localized magnetism at the lowest temperature [2], measurements under pressure on the stoichiometric compound $CeCu_5Au$ [17] showed no anomaly in T_K at P_c, suggesting that the magnetism remains itinerant around the QCP. As the latter behaviour is observed in our investigation, spin-fluctuation theory should also apply in the vicinity of the QCP of $CePd_2Si_2$. For both of our samples, the QCP occurred in a pressure domain where the characteristic Kondo energy $k_B T_K$ ($T_K \propto T_{max}$) typically reaches the CF splitting energy (see figure 1(b)). Furthermore, ln A is found to behave as $-\alpha \ln T_{max}$ with a slope $\alpha \simeq 4$, instead of the value of 2 expected for a normal heavy-fermion regime. This indicates entrance to an intermediate-valence regime, probably leading to deviations from the simple spin-fluctuation model. The fact that $\gamma^2(P)$ decreases more slowly than $A(P)$ above P_c might tempt us to invoke the predictions of [16], but our calorimetric measurement is not quantitative enough. It does allow us to demonstrate clearly a relationship between A and γ—showing in both a peak at P_c—but the γ-value extracted probably includes undefined addenda obscuring the physics.

As in previous measurements, NFL behaviour was observed in $\rho(T)$ at P_c in both samples over more than one decade in temperature. The stability of this behaviour with temperature has been proposed to result from a crossover between 'clean'- and 'dirty'-limit regimes for a specific amount of disorder [18]. However, this explanation disagrees with the systematic observation of power laws in $\rho(T)$ at P_c over a large temperature range for samples with residual resistivities spread over almost one decade [4–7]. The exponent in $\rho(T) = \rho_0 + AT^n$ in all cases reaches remarkably low values with $n \simeq 1.2$–1.3, a value generally attributed to a non-3D spin-fluctuation spectrum. However, let us recall that in other compounds such as $CeCu_2Ge_2$, a minimum of n close to 1 was found only for $P > P_c$, in a pressure domain where $k_B T_K$ reaches the CF splitting energy [19]. As this happens for $P \simeq P_c$ in $CePd_2Si_2$, one may wonder whether the low exponent observed is not a consequence of this change of regime.

Hydrostatic pressure reduces both lattice parameters a and c. At a given pressure, an additional uniaxial stress, σ, along one axis further reduces that lattice parameter while expanding the others. A description of the physical properties as a function only of the cell volume fails in this system, as shown by the various phase diagrams obtained for samples $\perp$ and $\|$. Whereas the situation remains mostly unchanged when σ is applied in the basal plane, the clear extension of the phase diagram for σ along the c-axis shows that the ratio c/a, reflecting the anisotropy of the system, is also a key parameter. Considering the spin-fluctuation prediction $T_N(P) \propto (P_c - P)^{2/d}$, the exponent 0.60 ± 0.05 obtained for sample $\|$ suggests that applying σ along the c-axis restores a 3D spin-fluctuation spectrum. With the same theoretical approach, the minimum values of the exponent in the $\rho(T)$ power law at P_c, predicted to be $T^{d/2}$, should be different for the two samples. However, the differences observed in $n(P_c)$ and in $A(P_c)$ (figure 3) are smaller than we might expect. The most striking effect revealed by this experiment is the apparent enhancement of superconductivity for σ applied along the c-axis with a 40% higher maximum value of T_c and a doubling of its pressure range. This enhancement is not related to a larger electronic mean free path due to reduced disorder, since the sample with a higher T_c also has the higher ρ_0. As spin fluctuations are thought to lie at the origin of the Cooper pairing, one may attribute the enhancement of superconductivity to different features of the spin-fluctuation spectrum. The values of the critical exponents in $T_N(P)$ suggest that 3D spin fluctuations would be more favourable for superconductivity, though many scenarios such as an increase in carrier density associated with a band modification under uniaxial stress remain possible.

Our measurements suggest a complex ground state in $CePd_2Si_2$ in the vicinity of its QCP at $P_c \simeq 2.7$–2.8 GPa where several energy scales such as the Kondo and excited CF energies interact. While the other archetypical system for a superconducting phase induced around its critical point, cubic $CeIn_3$, is insensitive to pressure conditions [4, 20], the physical properties of tetragonal $CePd_2Si_2$ are strongly affected by modification of anisotropy resulting from additional uniaxial strain along the c-axis. The quasi-2D behaviour evoked for spin fluctuations seems to be destroyed and superconductivity is enhanced around $P_c \sim 3.9$ GPa. As pure uniaxial stress experiments are extremely difficult to perform under pressure, the effect of the anisotropy on the superconductivity around a QCP should be checked for a compound close to its instability at ambient pressure. $CeNi_2Ge_2$, where traces of superconductivity as well as quasi-2D behaviour for spin fluctuations were found [3, 21], appears to be one of the best candidate compounds.

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