Journal of Physics: Conference Series **592** (2015) 012002 doi:10.1088/1742-6596/592/1/012002

Phase diagram of $CeRh_2Si_2$ under pressure studied by thermopower measurements

A Palacio-Morales[1,2], A Pourret[1,2], G Knebel[1,2], D Aoki[1,2,3], D Braithwaite[1,2] and J Flouquet[1,2]

[1] Univ. Grenoble Alpes, INAC-SPSMS, F-38000 Grenoble, France
[2] CEA, INAC-SPSMS, F-38000 Grenoble, France
[3] IMR-Tohoku University, Oarai, Japan

E-mail: alexandra.palaciomorales@cea.fr

Abstract.

We report the evolution of thermoelectric power under hydrostatic pressure up to 17 kbar and at low temperature in the heavy fermion compound $CeRh_2Si_2$. These measurements were performed using a thermoelectric setup specially designed for piston cylinder pressure cells. The suppression of the antiferromagnetic order (AF) into a paramagnetic order (PM) state was studied and the (T, P) phase diagram was precisely obtained. The different magnetic transitions at low temperature as a function of pressure, AF_1-AF_2 transition at P_c' and AF_1-PM transition at P_c, show significant changes in the thermoelectric signal. This support reconstructions of the Fermi surface in agreement with previous de Haas van Alphen experiments.

1. Introduction

The disappearance of the long range magnetic ordering as a function of an external parameter like pressure or magnetic field at a quantum critical point (QCP) in strongly correlated electron systems is still an open question [1]. The driving mechanism of the quantum phase transition at zero temperature from antiferromagnetic (AF) to paramagnetic (PM) ground states is questioned: what is the role of valence versus spin instabilities and is the disappearance coupled with Fermi surface (FS) changes? In heavy fermion compounds, which are well suited systems for such a study, very few measurements concern a careful investigation of FS instabilities in the AF state on approaching the critical pressure P_c or the critical field H_c where the AF order will switch to a PM ground state or to a field induced polarized paramagnetic state (PPM), respectively. A related open question is the resemblance between the AF order suppression by pressure and magnetic field mechanism. However, in some heavy fermion compounds FS reconstruction has been directly shown by quantum oscillations experiments. Prominent examples are the AF compounds $CeRhIn_5$ [2] and $CeRh_2Si_2$ [3] where the evolution of the FS has been studied as a function of pressure through their QCP. DHvA measurements confirmed a FS reconstruction at the QCP and led to the development of unconventional models such as the breakdown of the Kondo effect [4]. The relevant picture is that a variation from a small FS to a large FS through the critical pressure P_c in AF systems can be observed near a QCP [5].

Here, we focus the study on the heavy fermion compound $CeRh_2Si_2$. $CeRh_2Si_2$ is an ideal system to study the quantum phase transition from AF to PM states as the Néel temperature

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.
Published under licence by IOP Publishing Ltd

T_{N1} at ambiant pressure is high and the critical pressure $P_c \simeq 10.3$ kbar is rather low [6–8]. At ambiant pressure, $CeRh_2Si_2$ presents two magnetic transitions defining two AF domains, AF_1 and AF_2. The first transition, AF_1-PM with $T_{N1} \simeq 36$ K, is second order at ambient pressure, whereas the second one, AF_1-AF_2 with $T_{N2} \simeq 25$ K, is first order. These two AF states present distinct AF wave vectors deduced from neutron scattering experiments [9]. AF_1 presents the AF vector $q_1 = (1/2, 1/2, 0)$ and AF_2 is characterized by two AF vectors: q_1 and $q_2 = (1/2, 1/2, 1/2)$. The appearance of this second AF vector in the AF_2 phase is related to the folding of the magnetic structure [9]. T_{N1} and the T_{N_2} vanish at critical pressures $P'_c \simeq 5$ kbar and $P_c \simeq 10.3$ kbar, respectively [3, 7]. The quantum phase transition at P_c is characterized by the apparition of superconductivity with a maximum superconducting temperature T_{sc} of 400 mK at P_c [3, 10] suggesting magnetic fluctuations as origin of the superconducting pairing [11]. These spin fluctuations near the QCP produce significant increases in various thermodynamic and transports probes such as specific heat [12, 13] and resistivity through the A coefficient of the T^2 term [3, 14].

The FS of $CeRh_2Si_2$ at ambient [15] and at high pressure [6] are well known. Indeed, $CeRh_2Si_2$ presents a multiple connected Fermi surface with a localized behavior in the AF_2 state. Under pressure, modifications of the Fermi surface appear at P'_c and P_c suggesting a possible localized to itinerant FS transition. To clarify this point, an unambiguous proof of a FS change is necessary requiring a concomitance of various macroscopic and microscopic measurements. Among them, thermoelectric power is a very powerful probe as it is directly linked to the energy derivative of the density of states. In this paper, we present the development of a new setup to measure the different thermoelectric coefficients under pressure. Using this new device the pressure dependence of the thermoelectric power in $CeRh_2Si_2$ has been observed and the phase diagram (T, P) has been obtained. The strong increase of the the thermoelectric power near P_c confirms a FS reconstruction due the enhancement of spin/valence fluctuations near the QCP.

2. Experimental setup

Single crystals of $CeRh_2Si_2$ were grown by the Czochralski pulling method in a tetra-arc furnace. The sample was cut by spark cutter and oriented by x-ray Laue diffractometer. Thermopower measurements have been performed on a long-bar shape sample ($2.25 \times 0.70 \times 0.33$ mm) with a thermal gradient along the a-axis. The residual resistivity ratio RRR (ρ_{RT}/ρ_0) for the studied sample is ~ 10. We used a double wall piston cylinder pressure cell with an external cell body made of CuBe, reinforcing an inner cylinder of non-magnetic Ni-Cr-Al alloy. The transmitting medium was daphne oil 7373 which is a hydrostatic oil at room temperature and for pressures up to 22 kbar [16].

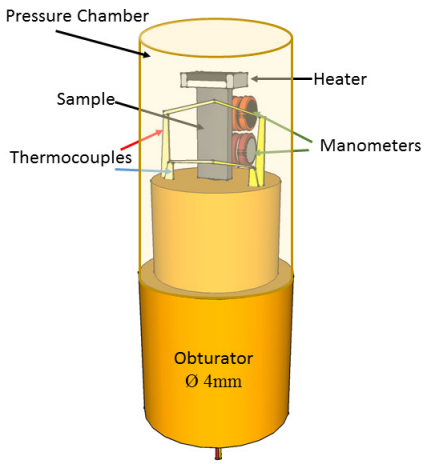


Figure 1. Schematic thermopower setup for pressure measurements. The reduced volume of the pressure chamber implies the use of thermocouples.

Similarly to various experimental techniques dedicated to measure thermoelectric coefficients under pressure [17–19], we have chosen to perform thermopower measurement using thermocouples instead of resistances as used in the well known "One heater-Two thermometers" vacuum setup due to the extremely reduced volume of the pressure chamber. The use of a pressure transmitting medium induces power-leaks inside the pressure chamber. Thermocouples measure directly the temperature gradient on the sample eliminating errors due to the thermal leak. AuFe(0.07% Fe)-Au thermocouples were used in

the temperature range $[0.5 - 50]$K [20] due to their quite good sensitivity. A 50 μm diameter wire of AuFe(0.07%Fe) is flattened to a 15 μm thickness and cut in 50-70 μm widths. A 15 μm diameter Au wire is used to complete the thermocouple. The thermocouples are spot welded on the hot/cold sides of the sample. Both thermocouples have a similar configuration inside the pressure cell in order to get an identical thermal path for each one. Moreover, the thermal coupling between the heater and the sample was optimized by applying silver paste on the electrical contact. A schematic representation of the pressure cell setup is shown in Fig.1. Two manometers (Mn-coil and Pb-coil) are also present inside the pressure chamber in order to follow the applied pressure. The coil made of resistive manganin wire allows to follow the pressure variations at room temperature. The Pb-coil is a piece of lead around which a copper wire is wound and it determines the pressure through the Pb superconducting transition at low temperature. The Mn coil can also be used as a heater.

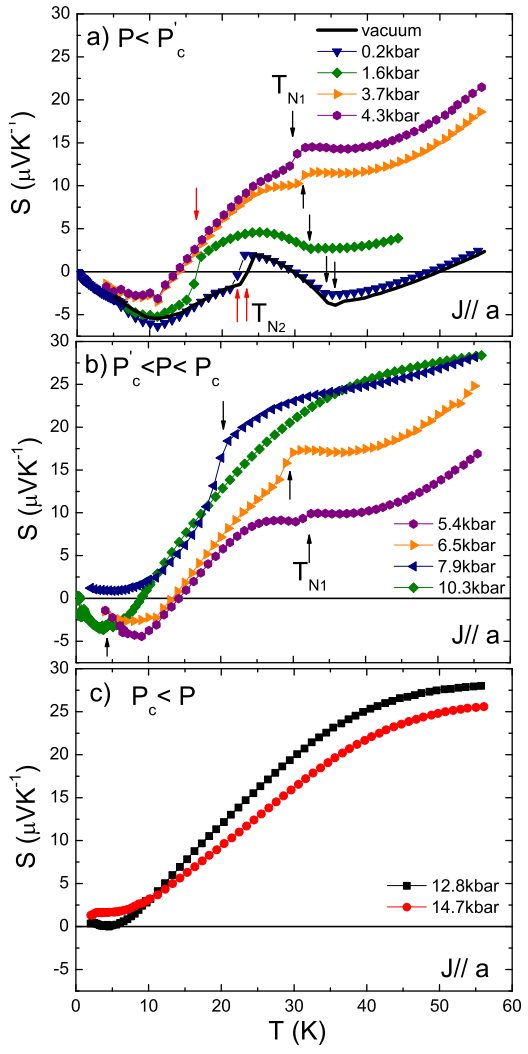


Figure 2. (Color online) Evolution of $S(T)$ measurements under pressure for a) $P < P'$, b) $P' < P < P_c$ and c) $P_c < P$. The verticals arrows indicate the different magnetic transitions T_{N_1} and T_{N_2}.

A precise comparison between vacuum and pressure setups reveals that the generated thermal gradient in the pressure setup was $5 - 10$ times smaller than in the vacuum setup for the same applied Joule heating in despite of all the efforts to reduce the leakage of power. The high sensitivity of the thermocouples and their extremely short thermalization time allows us to perform measurement with extremely small thermal gradient down to $\Delta T/T \simeq 0.4\%$ in the present device compare to $\Delta T/T \simeq 3\%$ in a conventional vacuum device. The good agreement between $S(T)$ measurements in vacuum and at low pressures ($P \sim 0.2$ kbar) shown in Fig.2a) validate the new device to measure thermoelectric coefficients under pressure.

3. Results

The temperature dependence of thermopower $S(T)$ in $CeRh_2Si_2$ performed for the different pressures regimes are shown on Fig.2 a), b) and c). $S(T)$ shows a very complex structure with several changes of sign due to different bands contribution to the thermoelectric power in this multiband compound.

At low pressure (Fig.2a)), the suppression of AF_2 at T_{N_2} and AF_1 at T_{N_1} correspond respectively to a drop and a kink in the thermoelectric power as indicated by vertical arrows. As pressure increases, the drop of $S(T)$ at T_{N2} becomes smaller and T_{N_2} decreases in temperature. Above 4 kbar, the signature of the AF_1-AF_2 transition cannot be identified in $S(T)$ in the temperature range of the measurements. The AF_1-PM transition at T_{N1} decreases in temperature under pressure and becomes a step like transition for $P > 3$ kbar. We can notice

that in this low pressure range the thermoelectric power is always negative at low temperature.

Close to $P_c' \simeq 5$ kbar, $S(T)$ becomes positive at low temperature and on approaching $P_c \simeq 10.3$ kbar, $S(T)$ changes sign again becoming negative at low temperature (Fig.2b)). These different changes of sign indicate changes of the dominant heat carriers in this multiband system near the critical pressures.

Finally, at high pressure above P_c (Fig.2c)), $S(T)$ becomes positive in all the temperature range. Consequently, the usual $S(T)$ behaviour for the Kondo Ce-family, i.e a positive $S(T)$ value at low temperature [21], is observed in the PM phase.

The different anomalies in the thermoelectric power previously mentioned are reported on the (T, P) phase diagram of $CeRh_2Si_2$ shown in Fig.3a). The blue symbols represent T_{N1} and the green ones represent T_{N2}. The associated error bars of the different transitions obtained by the first derivative of $S(T)$ with temperature are also reported. The phase diagram obtained from previous magnetoresistance (T_{N_1}, T_{sc}) [3] and neutron (T_{N_2}) [9] experiments is superimposed (grey symbols). Our thermoelectric power anomalies are in good agreement with previous results. From the T_{N_1} line obtained by thermoelectric power measurement, the value of P_c can be estimated around 10.8 kbar. Similarly, the value of P_c' was estimated from the T_{N_2} by comparison with the pressure dependance obtained by neutron measurements. The quality of our sample was too low to present the superconducting domain.

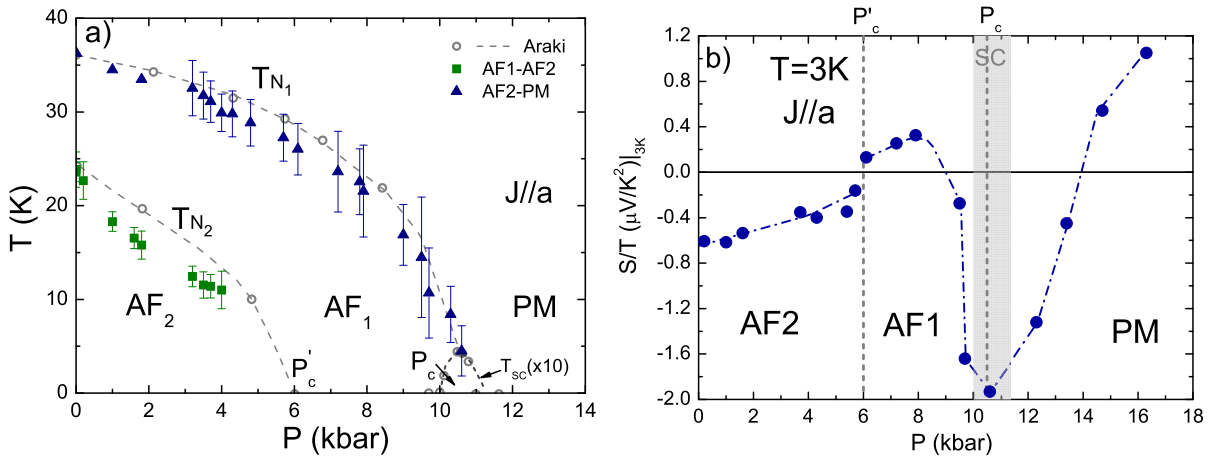


Figure 3. (Color online) a) (T, P) phase diagram of $CeRh_2Si_2$ obtained from $S(T)$ measurements with thermal current along the a-axis and compare to previous magnetoresistance (T_{N_1}, T_{sc})[3] and neutron (T_{N_2})[9] measurements. b) S/T values at 3 K as a function of pressure through the different magnetic domains. The critical pressures P_c' and P_c, as well as the superconducting domain are represented.

The evolution of S/T as a function of pressure at $T = 3$ K is shown in Fig.3b). Despite the fact that $T = 3$K is a rather high temperature, the constant value of S/T on the broad range of temperature validates this analysis. In the AF_2 state, S/T is negative and increases slowly with pressure. As already mentioned, a sign change occurs at the AF_2-AF_1 transition suggesting a possible reconstruction of the FS. This is supported by dHvA measurements [7] that show the suppression of the low frequencies electron-like branches (κ, ζ and O) at P_c' and the appearance of new hole-like branches q, p and r in the AF_1 domain. Around P_c, a sudden change of sign of S/T is observed with a strong decrease of S/T suggesting an important FS reconstruction as expected from dHvA measurements [7]. The dHvA frequencies clearly change at 10.3 kbar, suggesting

a first-order like phase transition. This indicates a discontinuous change of the Fermi surface at P_c. This FS reconstruction is coupled to spin/valence fluctuations [22] as shown the strong variation of the A coefficient of the T^2 resistivity law as well as of the Sommerfield coefficient of the specific heat. In the PM phase, S/T of $CeRh_2Si_2$ becomes positive and increases with pressure. All the frequencies of the AF phase disappear at P_c and 3 new high frequencies (A, B and C) appear in the PM state which total contribution is hole-like [7]. In many correlated metals, the absolute value of the dimensionless ratio $q = \frac{S}{T}\frac{N_{Av}e}{\gamma}$ (where e is the elementary charge and N_{Av} the Avogadro number) is close to unity [23]. In a single band model the number of carriers is inversely proportional to q. The value of S/T at 3 K and the corresponding γ [13] yield $q \sim 2$ at ambient pressure and $q \sim 2.5$ close to P_c suggesting no significant change of carriers number. At P_c, spin/valence fluctuations and inherent FS reconstruction move away the system from Fermi liquid behavior. Therefore, such a simple estimation of the number of charge carriers from the thermoelectric power is not possible in this multiband compound specially at P_c. The presence of the low energy fluctuations associated to FS changes seem to be a general feature of correlated materials at low temperature close to a QCP.

4. Conclusion

We have clarified a drastic change of the FS for the antiferromagnet $CeRh_2Si_2$ compound near the critical pressure P_c corresponding to the collapse of the AF order. The FS reconstruction at P_c is associated to the enhancement of valence/spin fluctuations close to P_c. The thermoelectric anomaly is spread in a large pressure range and masks the nature of the Fermi surface reconstruction at P_c according to previous dHvA measurements. The present result is the first observation of thermoelectric power through a FS instability where both FS reconstruction and drastic change of the low energy fluctuations are associated.

Acknowledgments

We acknowledge the financial support of the French ANR (within the program PRINCESS) and the ERC starting grant (NewHeavyFermion).

References

[1] Löhneysen H, Rosch A, Vojta M and Wölfle P 2007 *Rev. Mod. Phys.* **79** 1015
[2] Shishido H, Settai R, Harima H and Ōnuki Y 2005 *J. Phys. Soc. Jpn.* **74** 1103
[3] Araki S, Nakashima M and Settai R 2002 *J. Phys. Cond. Mat.* **14** L377
[4] Si Q, Rabello S, Ingersent K and Smith J L 2001 *Nature* **413** 804
[5] Gegenwart P, Si Q and Steglich F 2008 *Nat. Phys.* **4** 186
[6] Araki S, Settai R, Kobayashi T, Harima H and Ōnuki Y 2001 *Phys. Rev. B* **64** 224417
[7] Settai R, Araki S, Shishido H, Inada Y, Haga Y, Yamamoto E, Kobayashi T C, Tateiwa N and Ōnuki Y 2003 *J. Magn. Magn. Mater.* **262** 399
[8] Boursier R, Villaume A, Lapertot G, Aoki D, Knebel G and Flouquet J 2008 *Phys. B: Cond. Mat.* **403** 726
[9] Kawarazaki S, Sato M, Miyako Y, Chigusa N, Watanabe K, Metoki N, Koike Y and Nishi M 2000 *Phys. Rev. B* **61** 4167
[10] Araki S, Nakashima M and Settai R 2003 *Act. Phys. Pol. B* **43** 439
[11] Movshovich R, Graf T, Mandrus D, Thompson J, Smith J and Fisk Z 1996 *Phys. Rev. B* **53** 8241
[12] Graf T, Hundley M, Modler R, Movshovich R, Thompson J, Mandrus D, Fisher R and Phillips N 1998 *Phys. Rev. B* **57** 7442
[13] Graf T, Thompson J D, Hundley M F, Movshovich R, Fisk Z, Mandrus D, Fisher R A and Phillips N E 1997 *Phys. Rev. Lett.* **78** 3769
[14] Villaume A, Aoki D, Haga Y, Knebel G, Boursier R and Flouquet J 2008 *J. Phys.: Cond. Mat.* **20** 015203
[15] Abe H, Kitazawa H and Aoki H 1998 *J. Phys. Soc. Jpn.* **67** 1852
[16] Yokogawa K, Murata K, Yoshino H and Aoyama S 2007 *Jpn. J. Appl. Phys.* **46** 3636
[17] Link P, Jaccard D and Lejay P 1996 *Physica B* **225** 207
[18] Zhou J and Goodenough J 1996 *Phys. Rev. B* 393

[19] Hedo M, Nakamura D, Takaesu Y, Fujiwara T, Uchima K, Yagasaki K and Nakama T 2010 *J. Phys.: Conf. Series* **215** 012186
[20] Chaussy J, Guessous A and Mazuer J 1981 *Rev. Sci. Inst.* **52** 1721
[21] Amato A and Sierro J 1985 *J. Magn. Magn. Mater.* **4748** 526
[22] Miyake K and Ikeda H 2006 *J. Phys. Soc. Jpn.* **75** 033704
[23] Behnia K, Jaccard D and Flouquet J 2004 *J. Phys.: Cond. Mat.* **16** 5187