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Collapse of antiferromagnetism in $CeRh_2Si_2$: volume versus entropy

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Abstract
The thermal expansion of the heavy fermion compound $CeRh_2Si_2$ has been measured under pressure as a function of temperature using strain gages. A large anomaly associated with the Néel temperature has been detected even above the suspected critical pressure $P_c \sim 1.05$ GPa where no indication of antiferromagnetism has been observed in calorimetry experiments sensitive to the entropy change. The volume change at T_{N1} proves the first-order nature of the collapse of antiferromagnetism. An unexpected feature is the weak pressure dependence of the antiferromagnetic–paramagnetic transition in comparison to the fast pressure collapse predicted for a homogeneous first-order quantum phase transition with one single pressure singularity at P_c. A large pressure dependence is observed in the anisotropy of the thermal expansion measured parallel or perpendicular to the c-axis of this tetragonal crystal. This points out the collapse of the efficiency of the crystal field effects at P_v which indicates the entrance into the intermediate valence regime.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Some aspects of the disappearance of long-range magnetic ordering as a function of an external parameter like pressure or magnetic field at a critical point in strongly correlated electron systems (SCES) are still open questions [1]. Associated changes of the Fermi surface (FS) or the role of valence or charge instabilities close to the critical point are under debate. Even the nature (second-order or first-order) of the quantum phase transition at zero temperature from ferromagnetic or [2, 3] antiferromagnetic (AF) [4] to paramagnetic (PM) ground states is questioned.

Even in heavy fermion compounds (HFCs) which are suitable systems for such a study, very few measurements have been concerned with a careful study in the AF state on approaching the critical pressure P_c where AF order will switch to a PM ground state. Most discussions focus on non-Fermi liquid behavior observed in the PM state at finite temperature as P approaches P_c [1, 4]. This deficiency in the study of the collapse of the antiferromagnetism from the AF side is due to the fact that the signals related to the collapse of the order parameter (the sublattice magnetization $M_0(T)$) become very weak in neutron scattering as well as in calorimetry or transport measurements close to P_c [4].

In contrast to these probes, the relative volume (V) variation or its temperature derivative (the thermal expansion α) is expected to be more sensitive. For a second-order phase transition, the pressure variation of the Néel temperature of an antiferromagnet is expected to be large near P_c leading even to a divergence of its Grüneisen parameter ($\Omega_{T_{N1}} = \frac{-\partial \mathrm{Log} T_{N1}}{\partial \mathrm{Log} V}$) [5, 6] in the framework of the spin fluctuation theory [7]. For a first-order transition, a volume discontinuity can occur at P_c while the Nernst principle requires that the entropy reaches zero at $T \to 0$ K; according to the Clapeyron relation $\frac{dP}{dT} = \frac{\Delta S}{\Delta V}$, the P variation of T_{N1} starts with a vertical line. Two decades ago, it had already been pointed out that HFCs are characterized at very low temperature by a large electronic Grüneisen parameter $\Omega^* = -\frac{\partial \mathrm{Log} T^*}{\partial \mathrm{Log} V}$ where $k_B T^*$ stands for the dominant low-energy scale which can be the Kondo temperature, the spin fluctuation, or valence fluctuation temperature [8, 9].

Here we concentrate on the HFC $CeRh_2Si_2$. At ambient pressure, $CeRh_2Si_2$ presents two magnetic transitions: the first transition is second-order at ambient pressure, with $T_{N1} \sim$

35 K while the second one is first-order at ambient pressure, with $T_{N2} \sim 25$ K. $CeRh_2Si_2$ is an ideal system to study the quantum phase transition from AF order to PM as the Néel temperature $T_{N1} \sim 35$ K at ambient pressure is high, but the critical pressure $P_c \sim 1$ GPa is rather low. The first transition at T_{N1} is suspected to become first-order above 1.0 GPa since a pronounced reconstruction of the FS has been observed [10].

In this paper we present detailed measurements of the length variation of the heavy fermion compound $CeRh_2Si_2$ as a function of pressure. Previous experiments were reported in [11] on polycrystals and down to a temperature of 4.2 K. Some comparisons with ac calorimetry and inelastic neutron diffusion measurements are given, and the evolution of the nature of the magnetic transition at T_{N1} under pressure is discussed.

2. Experimental details

Single crystals of $CeRh_2Si_2$ were grown by the Czochralski pulling method. A sample of $2.5 \times 2.5 \times 3$ mm size was cut from a large crystal. To measure the relative elongation under pressure, we follow the procedure described in the pioneer's work on HFC [12]. A 2×2.5 mm Kyowa® strain gage was first glued along the c-axis direction of the sample using Kyowa® UC-27A glue and following a well established Kyowa procedure [13]. The ΔR change of the initial strain gage's resistance R is related to the relative elongation ε by (1):

$$\frac{\Delta R}{R} = K_s \varepsilon \tag{1}$$

R stands for the gage resistance at 300 K without any strain ($R \simeq 120\ \Omega$) and K_s is the gage factor, which is near 2 in general purpose strain gages. The temperature variation of K_s is known and does not exceed 2% between 2 and 300 K. A measurement along the a-axis direction was also performed: each relative elongation can either be measured separately with a CW reference sample, or with a differential method, by putting the gages on the two sides of the Wheatstone bridge.

A ^{4}He cryostat (1.6–300 K) enables us to perform low-temperature measurements. Since the resistance of the electric wires varies between the inside and the outside of the cryostat, we use a thermo-compensation system which consists of a strain gage glued on a tungsten carbide (CW) reference sample and a Wheatstone electrical bridge. The bridge is balanced at low temperature (30 K) by a precision adjustable potentiometer. Using a lock-in detector, a precision higher than 5×10^{-7} for the relative elongation of $CeRh_2Si_2$ is achieved.

Measurements under pressure up to 1.5 GPa can be performed inside a home-made CuBe pressure cell. The CW reference and the $CeRh_2Si_2$ sample are put inside a Teflon cartridge which is filled with Daphne® oil. This cartridge is put inside the pressure cell. A precise determination of the pressure value inside the cell is achieved at ambient temperature by a calibrated manganin sensor. At low temperature the pressure is determined by the superconducting transition of lead [14]. The precision on the value of the pressure value inside the cell is better than 0.005 GPa. Previously calorimetric measurements had been realized with the ac technique now extensively used in Grenoble [15].

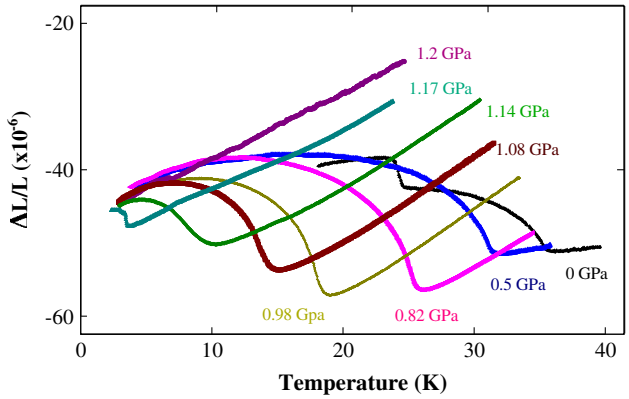

Figure 1. Relative variation $\Delta L/L$ along the c-axis as a function of temperature for different pressures.

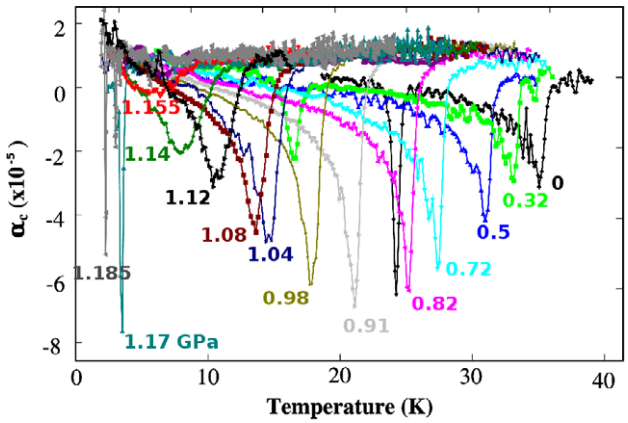

Figure 2. Thermal expansion coefficient α_c as a function of the temperature for different pressures in GPa.

3. Experimental results

Figure 1 shows the relative length variation $\Delta L/L$ along the c-axis for several pressures. At $P = 0$, on cooling the first AF transition occurs at $T_{N1} = 35$ K; a second transition appears in the AF state at $T_{N2} \sim 24$ K. This second transition disappears above $P \sim 0.5$ GPa [16]. In this paper, we focus on the disappearance of AF order, i.e. on the pressure variation of T_{N1} and related low-temperature properties.

It is worthwhile noticing that up to 1.0 GPa the signal does not change drastically. However, at 1.14 GPa it becomes weaker and surprisingly at 1.17 GPa a real discontinuity in $\Delta L/L$ occurs. The data at 15 different pressures is shown in figure 2 where the thermal expansion coefficient along the c-axis ($\alpha_c = \frac{\partial}{\partial T}(\Delta L/L)$) is represented. The measured thermal expansion takes into account the relative elongation of the CW reference sample (about 1.1×10^{-6} K^{-1}). In the following, the contribution of the CW reference sample has been subtracted to evaluate α_a and α_c, and also the thermodynamical functions (ΔS or ΔV, see figures 7 and 8). α_c obtained by dilatation measurements at $P = 0$ GPa is 10% lower than α_c reported in previous capacitive measurements [17].

One can see clearly the asymmetric signal observed at T_{N1} below 1.0 GPa and suddenly at 1.17 and 1.185 GPa a

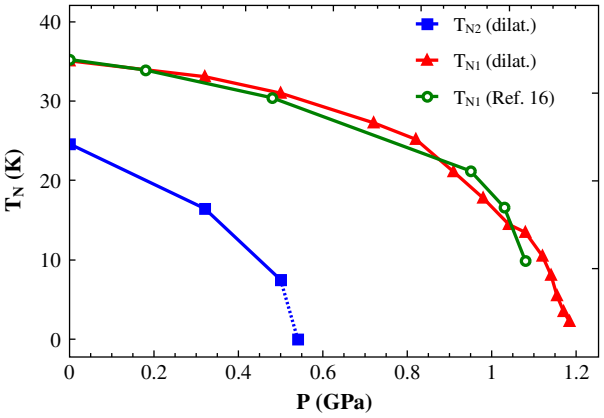

Figure 3. Phase diagram of $CeRh_2Si_2$ obtained by dilatation measurements and neutron scattering [16].

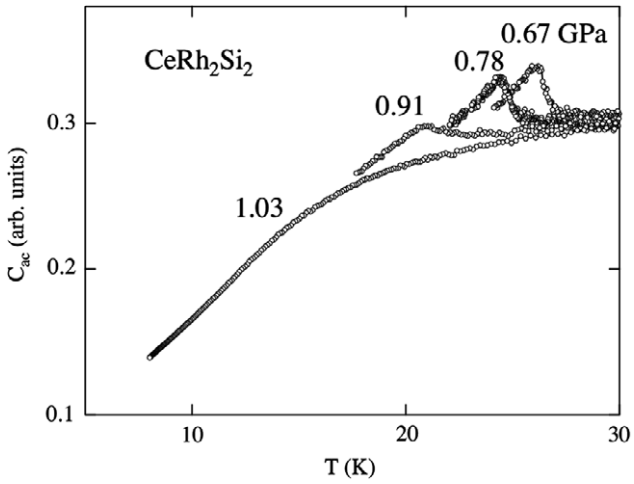

Figure 4. ac specific heat of $CeRh_2Si_2$ with the broadening and collapse of the AF anomaly on approaching the critical pressure P_c.

pronounced peak appears in $\Delta L/L$. At low pressure the change in α_c is associated with the P variation of the specific heat and the P increase in the slope of $\frac{\partial T_{N1}}{\partial P}$, according to the Ehrenfest relation for a second-order phase transition, $\frac{\partial P}{\partial T} = \frac{\Delta C}{T v \Delta \alpha}$, where C is the molar specific heat, T the temperature, v the molar volume and α the thermal expansion coefficient.

Figure 3 represents the pressure dependence of T_{N1} which is defined by the minimum of $\frac{\partial}{\partial T}(\Delta L/L)$. Rather good agreement is found between T_{N1} determined by different techniques at zero pressure. But also under pressure at least below 1 GPa when a signal associated to T_{N1} can be detected in resistivity $T_{N1}(P)$, ac calorimetry $T_{N1}(C)$, inelastic neutron scattering $T_{N1}(M_0)$, or dilatation experiments, a good agreement is found in the determination of T_{N1}. It is worthwhile noticing that a clear entrance in the AF ordering is observed at T_{N1} in this experiment above 1.04 GPa while no signal can be detected in resistivity experiments above 1.03 GPa [10, 18, 19] and also a very broadened signal can be seen in calorimetry experiments only up to $P \sim 1.03$ GPa as shown in figure 4 but also recently in [20].

Previous strain gage P results on $CeRh_2Si_2$ [11] are far more broadened than the one reported here. At 1.06 GPa, T_{N1} can be located at 15 K, very similar to the data at 1.04 GPa in figure 2, but the width of the transition (about 5 K) is roughly twice that measured here; the amplitude of α at T_{N1} in [11] is three time smaller than here. The drop of the specific heat signal at T_{N1} for $P > 1$ GPa by comparison to the large response in α cannot be expected from the Ehrenfest relation. When both signals (in C and α) have well defined anomalies at T_{N1}, an excellent agreement is found in the determination of T_{N1}.

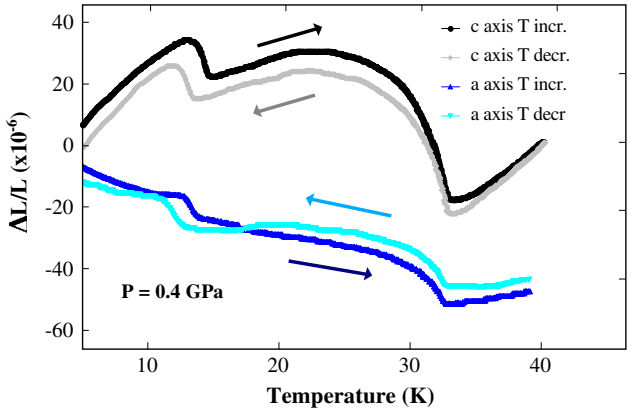

Figure 5. Relative variation of $\Delta L/L$ along the c-axis and the a-axis at 0.41 GPa.

The pressure variation of the ratio $\frac{\alpha_a}{\alpha_c}$ at T_{N1} has been also determined for a few pressures. A large pressure dependence of $\frac{\alpha_a}{\alpha_c}$ at T_{N1} is observed: $\frac{\alpha_a}{\alpha_c}$ at T_{N1} increases continuously from 0.23, 0.73, and 1 respectively for $P = 0.41$ GPa, 0.77, and 1.05 GPa; in [17], at $P = 0$, $\frac{\alpha_a}{\alpha_c} \sim 0.14$. Examples of the $\frac{\Delta L}{L}$ curves along a-and c-axes is given in figure 5 for $P = 0.41$ GPa and in figure 6 for $P = 1.05$ GPa. A very similar evolution of α_a and α_c is found for P higher than 1 GPa. An interesting point is the existence of a hysteresis in the transition associated with T_{N2} for $P = 0.41$ GPa. Such a behavior has already been reported by Ohashi *et al* [18] from resistivity measurements. This hysteresis is found only near T_{N2} for both c-and a-axes, confirming the first-order nature of the transition. No hysteresis is detected at T_{N1} in agreement with the assumption that below 1 GPa the transition is second-order (in excellent agreement with the Ehrenfest relation).

The volume change associated with the AF transition is represented in figure 7 by supposing an average P value of $\alpha_a = \frac{\alpha_c}{2}$. $\frac{\Delta V}{V}$ is calculated by withdrawing the CW reference contribution and by integrating the area below each curve represented in figure 2.

A clear change of regime appears for $P \sim 1.05$ GPa. Roughly the same characteristic pressure is detected in the broadening (ΔT) of α_c at T_{N1} where ΔT stands for the full width half-maximum of the α_c versus T curves (inset of figure 7).

The two surprising results are that

(i) contrary to both expected previsions of AF spin fluctuation ($T_{N1} \sim (P_c - P)^{2/3}$ [4]) and first-order transition, where the slope of T_{N1} versus P must be also infinite according to the Clapeyron relation and Nernst principle, T_{N1} appears

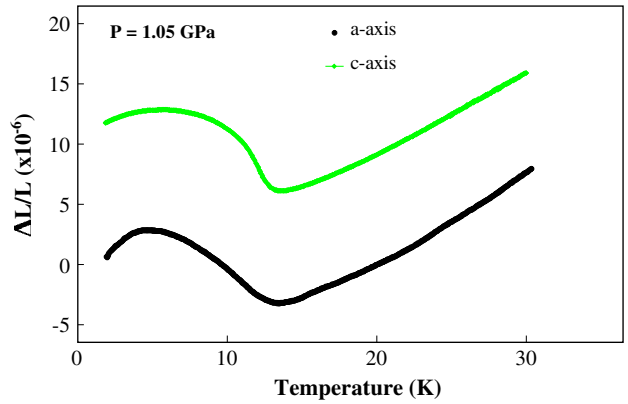

Figure 6. Comparison between the relative variation of $\Delta L/L$ along the c-axis and along the a-axis at 1.05 GPa.

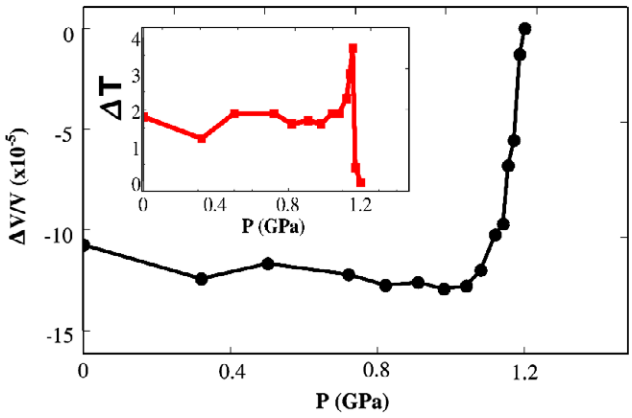

Figure 7. Relative change of volume at the AF transition extracted from figure 2. Inset: temperature broadening of T_{N1} (mean half-maximum calculated with the thermal expansion coefficient of figure 2).

to extrapolate linearly with P to zero down to $P_c \sim 1.2$ GPa and not with an infinite slope.

(ii) A real volume discontinuity is detected in the vicinity of P_c which points to a first-order transition.

Up to now only the regime close to T_{N1} was analyzed. Below T_{N1}, the low-temperature behavior is dominated by the P variation of the average effective mass m^* which is related to the pressure derivative of the entropy (S) according to the Maxwell relation (2):

$$\frac{\partial V}{\partial T} = -\frac{\partial S}{\partial P}. \tag{2}$$

The pressure derivative variation of the Sommerfeld coefficient γ of the linear temperature term of the specific heat is shown in figure 8. For $P > 1.08$ GPa, T_{N1} is too close to our low-temperature limit and the determination of $\frac{\partial \gamma}{\partial P}$ is not possible. The inset also shows the pressure variation of γ determined by previous calorimetric measurements [21] and now here by dilatation.

As indicated in figure 8, the direct specific heat measurements suggest a maximum of γ at P_c equal to 80 mJ mol^{-1} K^{-2} [21]. This value is not too far from our

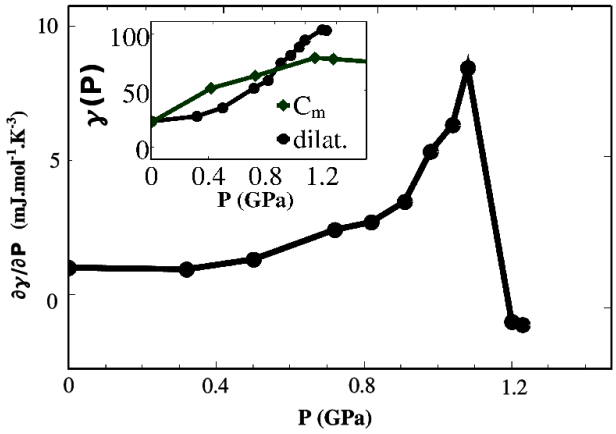

Figure 8. Evaluation of $\frac{\partial \gamma}{\partial P}(P)$. Inset: comparison of $\gamma(P)$ in mJ mol^{-1} K^{-2} determined by dilatation measurements and by specific heat [21].

extrapolation regarding the crudeness in the pressure variation of the $\frac{\alpha_a}{\alpha_c}$ ratio and also a different hydrostaticity between our pressure cell (with, at room temperature, a liquid transmitting medium) and the previous specific heat measurements (with a solid transmitting medium). The relative pressure variation of γ obtained by integration of $\frac{\partial \gamma}{\partial P}$ is in good agreement with effects observed in HFC [4] and the relative pressure variation of the A coefficient of the resistivity described below. The maximum of γ and of ΔT may not correspond to the collapse of T_{N1} which occurs for $P \sim 1.2$ GPa as is predicted for a second-order magnetic quantum critical point in spin fluctuation theory. We will analyze later the possible origin of such a shift.

4. Discussion

Strain gage probes are confirmed to be a unique tool to detect the magnetic ordering close to its collapse via a first-order transition. Only a few direct accurate measurements of the volume variation under pressures close to quantum criticality have been performed (see recently [11, 22, 23]). The accuracy of the relative volume change $\frac{\Delta V}{V}$ determined using strain gages is better than 5×10^{-7}, i.e. one order of magnitude higher in comparison to x-ray and two order of magnitude higher than neutron scattering. The power of strain gages was recently reemphasized by the discovery of the first-order quantum transition between the hidden order phase and the antiferromagnetic phase of URu_2Si_2 as a function of pressure [24].

A clear volume discontinuity is observed for $P \sim 1.170$ and 1.185 GPa. By comparison to a well known example such as the first-order transition or the melting curve of solid ^{3}He where $\frac{\Delta V}{V} \simeq 5\%$ but P_c is only 34 bars [4], in $CeRh_2Si_2$, $\frac{\Delta V}{V}$ is reduced roughly by three orders of magnitude but P_c is increased by roughly two order of magnitude. The mechanical work $P\Delta V$ at P_c in $CeRh_2Si_2$ is only 10% smaller than the one furnished in ^{3}He. Thus the first-order nature of the transition in $CeRh_2Si_2$ above 1.0 GPa is not weak. In terms of valence transition, if we take as reference the γ–α transition

of Ce metal where a 16% volume change [25] corresponds to a similar volume variation [26], the valence drop at P_c is weak. In $CeRh_2Si_2$, the P entrance in the intermediate valence domain is a transition at a very low temperature dominated by the discontinuity of the volume and not of the entropy, in strong contrast to the case of Ce metal [27].

Despite this weak variation of the valence, the tendency for an isotropic behavior in the thermal variation of ΔL_a and ΔL_c at low temperature above 1 GPa points out the quasi-coincidence between the magnetic quantum critical point (P_c) and the valence critical (or crossover) pressure (P_v) where the 4f electron will lose its sensitivity to the crystal field effect [4]. In contrast, for the $CeRu_2Si_2$ family, a strong decoupling occurs between $P_c \sim -0.3$ GPa and $P_v \sim 4$ GPa and a quasi constant value of $\frac{\alpha_a}{\alpha_c} \sim 0.3$ is observed on both sides of P_c for pressure sweeps near 1 GPa [28].

In $CeRh_2Si_2$, P_c and P_v are not discernible. Our guess will be that $\frac{\alpha_a}{\alpha_c} \to 1$ for $P > P_v$. Thus the pressure evolution of the ratio $\frac{\alpha_a}{\alpha_c}$ is a new interesting feature rarely considered under pressure. Here it was measured only for a few pressure points. The change in the ratio $\frac{\alpha_a}{\alpha_c}$ at T_{N1} is correlated with the P variation of the magnetic anisotropy [29]. The damping of the Ising type anisotropy under pressure is caused by the fast pressure increase of the Kondo temperature T_K. At $P = 0$, the estimation of T_K is between 30 and 100 K [30, 31], and the crystal field splitting C_{CF} is near 300 K [32]. Already near P_c, the Kondo effect energy reaches C_{CF}. As for $CeIn_3$ or $CePd_2Si_2$, the magnetic instability at P_c coincides or is very close to the pressure P_v where the Ce centres enter in their intermediate valence regime [4]. With a Kondo Grüneisen parameter $\Omega^*(T_K) \simeq 100$ (see [4, 8, 9, 28]), T_K will double at P_c while C_{CF} is suspected to be weakly pressure dependent, thus $k_B T_K$ will rapidly overcome C_{CF} above P_c.

To understand the slow P collapse of AF, an appealing possibility is that the collapse from AF to PM is not characterized by a unique pressure but a pressure slot [4]. This P interval may be an intrinsic property governed by quantum effects without hysteresis phenomena. It is worthwhile mentioning that different temperature cycles lead to the same signal at $P = 1.17$ GPa. The question of a pressure spreading between the onset of first-order transition at P_c^- and its disappearance at P_c^+ has been discussed for a structural transition but, as emphasized, spin matter offers a large diversity of situations [4]. Departures from the universality of second-order quantum phase transitions have already been pointed out, for example in MnSi [33–35], where a phase separation exists over an extended P range (1.2–1.6 GPa) as well as there being large departures from FL behavior at least up to 2 GPa and down to $T \sim 0.5$ K.

The complex critical pressure regime has already been pointed out in elastic neutron scattering experiments [16]. Up to 1.03 GPa, the normalized sublattice moment, M_0 is proportional to T_{N1}, while at 1.08 GPa, M_0 decreases far more strongly than T_{N1}. This particular behavior supports the image of a phase separation: in this frame, the fraction of AF phase decreases rapidly for P above 1.04 GPa since $T_{N1}(P)$ becomes weakly P dependent. In a conventional picture, T_{N1} will even be P invariant while the P change in the AF fraction governs the apparent P collapse of the sublattice magnetization. Of course the next step now will be to combine strain gage and neutron scattering experiments in order to clarify the problem of small surviving magnetic moments above 1.04 GPa.

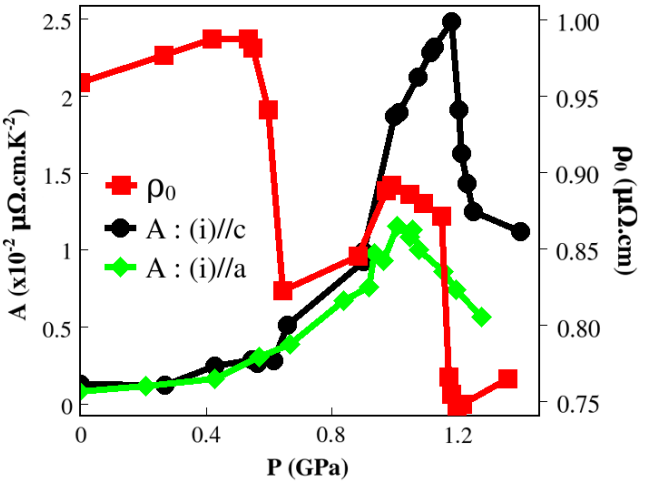

Figure 9. Pressure variation of the inelastic term A for the current (i) applied parallel to the a-axis [37] and parallel to the c-axis [10].

Evidence for a first-order transition is given by the observation of FS reconstruction near 1 GPa [36]. It is supported as described below by resistivity measurements, not only by the difficulty to observe non-Fermi liquid behavior close to P_c as predicted in the case of a second-order magnetic quantum phase transition [1, 4], but also by the observation of a pressure slot [37, 38] for the collapse of AF, in good agreement with the present dilatation data.

The resistivity anomaly associated to T_{N1} becomes small close to P_c. An indirect way to detect the enhancement of m^* and the mark of a complex behavior close to P_c is to look in detail at the low-temperature variation of the resistivity generally described by (3):

$$\rho(T, P) = \rho_0(P) + A_n(P)T^n. \tag{3}$$

In contrast to many HFCs close to P_c, in $CeRh_2Si_2$ a clear Fermi liquid regime is detected ($n = 2$) on both sides of P_c, and even close to P_c [10, 18, 19]. The persistence of this FL behavior even up to rather high temperature ($T \simeq 1$ K) is quite unique for Ce HFCs where usually close to P_c a non-FL behavior in resistivity ($n < 2$) persists well below 1 K in the vicinity of P_c. This singularity is caused by the strong first-order nature of the transition from AF to PM.

A recent careful analysis [37] of the electrical resistivity obtained with the current i applied in the basal plane has confirmed the existence of large P changes in residual resistivity and in the T^2 term [18] not only around a given maximum P_c but for a pressure range from 1.03 to 1.2 GPa, as shown in figure 9.

Careful studies of the resistivity around P_c were reported previously with the current along the c-axis [10]. No such singular behavior was found within a similar pressure interval. The pressure variation of A and ρ_0 near P_c is much stronger for the current applied along the a-axis than along

the c-axis. This difference must be related to the pressure variation of the magnetic and electronic anisotropy discussed previously [36, 29, 37].

We have not yet studied the superconducting properties of $CeRh_2Si_2$ which seem related to the AF–PM transition but very sensitive to the sample's quality [10, 39, 40]. Resistivity measurements with the current along the c-axis point out a full superconductivity drop only in a very narrow P window ($\Delta P = 0.05$ GPa at $P = 1.05$ GPa) [37]. The occurrence of a phase separation and associated change in spin and charge dynamics may lead to a quite furtive superconductivity. Simultaneous measurements of calorimetry and thermal expansion can certainly elucidate this complex quantum puzzle.

$CeRh_2Si_2$ is an excellent system to clarify the link between the Fermi surface (FS) evolution and heavy fermion properties under pressure and magnetic field. If the AF–PM phase transition is first order, a change in the localization of the 4f particle can be easily accepted. A key problem is the FS evolution first at $P = 0$ in the polarized paramagnetic state (PPM) state created above the metamagnetic field ($H_c \sim 23$ T) and then inside this PPM state under pressure, notably for $P > P_c$ [41]. By comparison to $YbRh_2Si_2$, where similar questions are under debate, $CeRh_2Si_2$ is a more realistic experimental case since FS determinations have already been achieved inside the AF and PM phases at low field [36]. Unfortunately, this situation is unlikely to happen in $YbRh_2Si_2$ due to the huge value of the effective masses and the low value of the magnetic critical field ($H \simeq 0.1$ T) [38, 42, 43]. By comparison to the $CeRu_2Si_2$ case where controversies still exist on the localization of the 4f electrons in the PPM state above H_c due to the lack of a complete determination of the FS [4], the favorable ingredient of $CeRh_2Si_2$ is the weak value of the effective masses (five times lower) at the metamagnetic field. That must give the opportunity for a full detection of the FS, results which are far from being achieved for $CeRu_2Si_2$.

5. Conclusion

Careful thermal expansion experiments on a $CeRh_2Si_2$ single crystal clarify the size of the volume discontinuity which is involved in its first-order AF collapse. The striking point is the occurrence of a P slot for the disappearance of AF. Such a window is in agreement with transport measurements. Thus this coincidence reveals an intrinsic property. At least our crystal responds to a pressure step of only 0.015 GPa, suggesting a weak P inhomogeneity. A favorable factor is the isotropic behavior of $CeRh_2Si_2$ close to P_c.

Acknowledgments

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