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2005 J. Phys.: Condens. Matter 17 S871

(http://iopscience.iop.org/0953-8984/17/11/016)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **17** (2005) S871–S876 doi:10.1088/0953-8984/17/11/016

# Successive phase transitions in $CePd_2Ga_3$ on the route from ferro- to paramagnetism

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Received 5 January 2005
Published 4 March 2005
Online at stacks.iop.org/JPhysCM/17/S871

**Abstract**
Several years ago $CePd_2Ga_3$ was characterized as a ferromagnetically ordered Kondo lattice system. Resistance measurements evidenced the disappearance of the ordered phase under high pressures around an extrapolated value of 5 GPa. Thus, this material appeared to offer the chance to study a quantum critical point between a ferromagnetic Kondo phase and the paramagnetic state which is a rather rare scenario. In DC susceptibility measurements under pressure, however, the magnetic phase transition was found to disappear already at much lower pressures (~2.4 GPa), which was interpreted as a change in the type of magnetic order.

Here we present new results from AC susceptibility and specific heat measurements closely investigating the pressure range around and above the assumed magnetic breakdown. While the specific heat data do not significantly show changes in the phase transition characteristics, the susceptibility data corroborate the former result of a magnetic change close to 2.3 GPa and, moreover, add another transition near 2.8 GPa. They also support the idea of antiferromagnetism in the phase next to the paramagnetic state.

## 1. Introduction

Heavy fermion systems have attracted new intensified attention in the past years because of the discovery of the phenomenon of quantum critical points (QCPs) in many of them. Since the hybridization between localized f-states and the conduction band states in these materials very sensitively depends on the electron distribution around the magnetic ions, i.e. the nature and distance of the nearest neighbours, the transition temperature between a magnetically ordered phase and a paramagnetic (PM) Fermi liquid phase can be easily tuned to $T = 0$ K by high pressure or chemical substitution. This peculiarity has been known for a long time, and is described by the well-known Doniach picture [1]. As a result a large number of Ce compounds and alloys (and to a lesser amount Yb and U systems) are reported close to a QCP where the properties are determined by quantum fluctuations leading to characteristic deviations from the power laws of the Fermi liquid state ('non-Fermi liquid (NFL) properties') [2]. Special excitement has been evoked by the discovery of superconductivity at rather low temperatures in close vicinity of the QCP, e.g. in $CeIn_3$ or $CePd_2Si_2$ [3].

In principle, there seem to be no limitations as far as the type of the magnetically ordered state disappearing at a QCP is concerned; it may be antiferromagnetic (AFM) as well as ferromagnetic (FM). The corresponding NFL properties, however, will be different (for a review see [2]). In practice only a small number of FM systems exist in contrast to many antiferromagnets. On the one hand, FM materials containing Ce are a minority on the whole. They seem to be restricted to a certain range of Ce–Ce distances [4]. Many Ce ferromagnets are well in the localized moment regime where they do not possess screened moments and large effective masses as is typical for heavy fermions. It is suspected that the Kondo effect tends to stabilize an AFM order [5]. On the other hand, the few FM systems which have been investigated for a QCP seem to avoid a direct transition into a PM phase via $T_C = 0$ K. For instance, in FM CeAg, which is taken as a prototype of a Kondo lattice described by the Doniach diagram, the magnetization abruptly disappears at a pressure of 3 GPa where the Curie temperature is still far from $T = 0$ K [6]. In $CeRu_2Ge_2$ the FM phase is completely imbedded in the regime of an AFM phase [7]. Recently, the Kondo lattice $CeAgSb_2$ has been shown first to switch to AFM at $\sim$3.5 GPa before becoming PM at around 5 GPa [8]. Only in CePt is a direct transition from FM to PM reported [9], but the critical pressure is as high as 12 GPa where analyses other than by electrical resistance are very difficult.

Moreover, it is worth mentioning that there is no *ferromagnetic* Ce system in which superconductivity has been detected, contrary to uranium compounds where superconductivity and FM have been found to coexist in UGe and URhGe [10]. Needless to say, it remains a great challenge to search for a QCP in FM Ce materials.

Here we present new detailed results on $CePd_2Ga_3$. This hexagonal compound was discovered in 1993 and shown to be FM below 6.2 K [11]. It can be characterized as a Kondo lattice with a low Kondo temperature $T_K \sim$ 4–5.5 K, an electronic specific heat $\gamma \sim 95$ mJ mol$^{-1}$ K$^{-2}$, and a PM moment of 2.72 $\mu_B$. The magnetic properties are influenced in part by a rather low lying first crystal field excitation at 45 K. From elastic neutron scattering it is known that the magnetic structure consists of ferromagnetically aligned moments of $\sim$1.5 $\mu_B$ lying in the basal planes which couple ferromagnetically with each other [12]. Resistivity measurements under pressures up to 5.3 GPa have revealed a decrease of the ordering temperature extrapolating to a QCP around 5 GPa. No peculiarities in the dependence of $T_C(p)$ had emerged [13].

A pressure of 5 GPa is still uncomfortably high for quantitative specific heat measurements. Since it is easier to perform magnetic investigations, the DC susceptibility (corresponding to the magnetization $M$ at low fields) was measured in a diamond anvil cell (DAC) using a SQUID detector. As a surprise it turned out that the signal proportional to $M$ rapidly decreased around 2.3 GPa; above 2.5 GPa there were no more traces of FM above 1.4 K [15]. Since the technique was only sensitive to FM a change into an AFM or modulated ordered state could not be ruled out and would also be in accord with the resistance data of Hauser *et al* [13].

## 2. Experimental details

To study the apparent breakdown of FM in $CePd_2Ga_3$ in closer detail we have applied additional techniques: AC susceptibility which is even sensitive to AFM transitions and specific heat measurements.

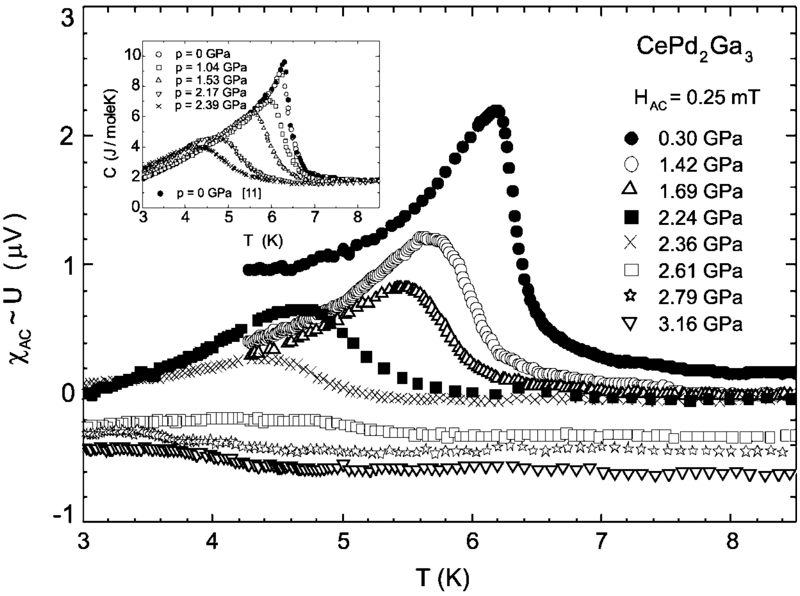


**Figure 1.** Pressure variation of the AC susceptibility of $CePd_2Ga_3$ measured with a driving field amplitude of 0.25 mT. Only part of the data is shown for the sake of clarity. Curves gained with different driving fields are not included, too. The inset shows the specific heat in the vicinity of the magnetic ordering transition under several different pressures.

The AC susceptibility is measured in a conventional diamond anvil cell where only a very small sample volume of about 0.01 $mm^3$ is available. The coil system is built up from two separate identical pairs of generator and pick-up coils, one enclosing the diamond anvils, the other mounted alongside, being empty. The filling factor of this configuration is about 1:5000. An additional winding on one of the coils can be used for fine compensation of the offset at any temperature wanted. The two generator coils connected in series are driven by an AC current source distinguished by a very high stability of the frequency (2000 Hz) and the amplitude ($\sim 10^{-4}$). A second phase-locked output which can be shifted in phase and varied in amplitude is used for the compensation winding, leading to a signal-to-noise ratio of better than 15. The generator current is kept below 100 mA (corresponding to an applied field amplitude of 3.5 mT) in order to avoid excessive heating in the coils.

Our technique to measure specific heat under pressure [16] has been improved during recent years by using a pressure cylinder made from high strength steel, thus extending the pressure range well beyond 2 GPa, and by employing relaxation calorimetry, leading to improved absolute accuracy as can be seen from a comparison with data from the literature [11].

The sample material is polycrystalline. Its thermal treatment and the check on structural data were described by Bauer *et al* [11]. It is taken from the same batch which was studied by DC susceptibility measurements [15].

## 3. Results

The specific heat was measured primarily with the aim to study the phase transition anomaly, the evolution of which up to 2.4 GPa is depicted in the inset of figure 1. We find a monotonic shift of the transition temperature $T_{\mathrm{Mag}}$ to lower values accompanied by a decrease of the transition height. The broadening of the transition partially originates from the pressure distribution

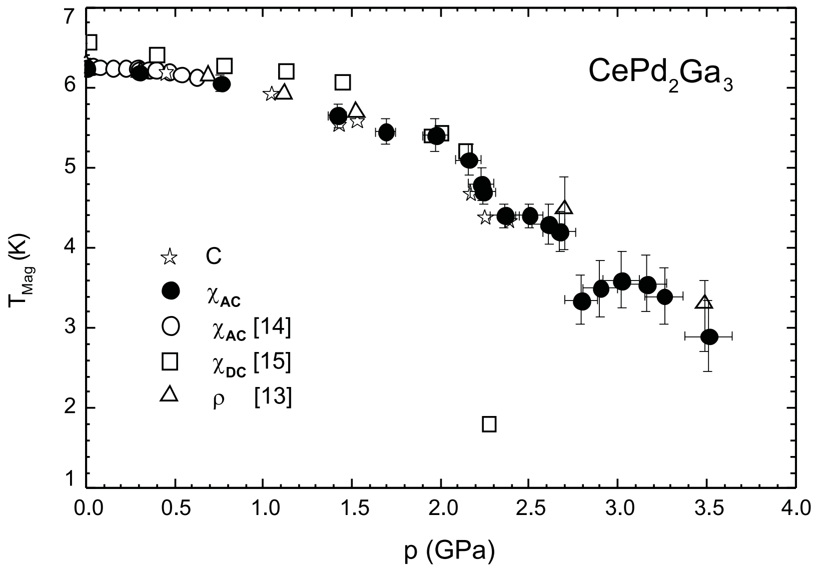


**Figure 2.** The magnetic ordering transition temperature of $CePd_2Ga_3$ as a function of pressure evaluated from different bulk properties (specific heat $C$, resistivity $\rho$, DC and AC magnetic susceptibility $\chi$).

estimated from the smearing of the superconducting transition of the indium embedding medium. At the highest pressures reached the region has just been entered, where in the early studies the disappearance of $M$ was observed. No signs of singular variations are seen. The transition is still clearly discernible and the anomaly does not show any change in its qualitative shape.

The data points between 1 and 9 K have been fitted by a mean field model including the Kondo effect [17], yielding both $T_{\mathrm{Mag}}$ (included in figure 2) and the Kondo temperature. The deviations of $T_{\mathrm{Mag}}$ determined this way from an entropy balance construction are less than 0.1 K; the fit uncertainty for $T_{\mathrm{K}}$ is about 7%. For the latter a continuous rise from $T_{\mathrm{K}} = 5.5$ K at $p = 0$ to $T_{\mathrm{K}} = 8$ K at $p = 2.4$ GPa results, the slope becoming steeper above $\sim$1.3 GPa. Even at 2.4 GPa signs of NFL behaviour do not show up in the covered temperature range down to 0.3 K.

In addition the material was studied by AC susceptibility measurements. Because of the use of a DAC there was in principle no limitation of the pressure range, not even by the comparably soft non-magnetic BeCu gasket and the somewhat larger than usual sample volume bore. So, it was possible to follow the transition up to 3.5 GPa in small pressure increments as well as in loading and releasing cycles. The maximum pressure is determined by the magnetic signal becoming too faint to be unanimously discerned from the background of the incipient superconducting transition of the indium manometer. This problem and the increasing self-heating of the coils finally prevents measurements in a $^3$He-cryostat to closely study the regime around the supposed QCP near 5 GPa.

The main part of figure 1 presents data of $\chi(T, p)$. The shift of the transition as well as the dropping height are evident. $T_{\mathrm{Mag}}$ is assigned to the maximum of the respective curves. The sequence of transition temperatures determined this way shows two steps at 2.3 and 2.8 GPa, respectively (figure 2). Additional information is drawn from variations of the driving field. Since an FM material below the Curie temperature responds to field changes by domain wall motions, an increase in the AC amplitude leads to an altered sampling of the hysteresis curve.

As a consequence the position of the maximum and the broadness of the anomaly are changed. At AFM transitions this effect is not present.

In figure 2 the data on the phase transition in $CePd_2Ga_3$ are collected from all experiments performed so far [13–15], demonstrating a most satisfactory agreement. The small deviations are within the combined errors and have mainly to be attributed to the different ways to extract $T_{Mag}$ from the data. Above all it holds true for the DC susceptibility where the Curie temperature was obtained from an extrapolation to zero of the temperature dependence of the magnetization $M(T)$.

As mentioned before, two steps of about 1 K height show up mainly based on the AC susceptibility data but not in contradiction to the other results. The first step occurs at the same pressure at which the measurements of the DC susceptibility recorded the dramatic breakdown of $M$. (The distinctly lower transition temperature then found may be traced back to the way of defining $T_C$ from $M$, see above.) Moreover, it is supported by the specific heat results, even if there is a lack of data at somewhat lower pressures. The second step at 2.8 GPa is only evidenced by the AC susceptibility data. An extension of the calorimetric measurements up to that pressure was not possible. The resistance data, on the other hand, are not sufficiently closely spaced to show the step. From different runs as well as from cycling the pressure around 2.8 GPa this feature is reproduced in $\chi_{AC}$. It is supported by the signal's dependence on the amplitude of the generator field. Domain effects, which are clearly seen between 0 and 2.3 GPa and, interestingly, persist between 2.3 and 2.8 GPa, are definitely lost beyond the second step at 2.8 GPa.

## 4. Discussion

The new results confirm our former supposition that $CePd_2Ga_3$ is another example of an FM Kondo system which first changes its type of magnetic order before reaching a QCP: the scenario is somewhat more sophisticated, however, insofar as there appear to be two transitions. The first one near 2.3 GPa comes along with a marked loss of magnetic moment as concluded from the DC susceptibility measurements [15]. Since the AC susceptibility shows a phase transition near 2.3 GPa with remaining domain effects some FM component seems still to be present, which could originate from a ferrimagnetic or a canted AFM order. Magnetic structure data are needed to answer this question. Beyond the next step in $T_{Mag}$ $(p)$ at 2.8 GPa the signal height has decreased again together with a complete disappearance of domain effects. We take that as strong evidence for the appearance of an AFM phase.

In our opinion the pressure effects in $CePd_2Ga_3$ cannot be discussed within just an isotropic frame like Doniach's diagram. Of course, the mean reduction of volume will cause an increase in hybridization leading to the rise of the Kondo temperature observed. Simultaneously the local moments are reduced. The RKKY interaction between the localized moments, however, cannot be treated on the same isotropic footing. It is expected to be anisotropic corresponding to the underlying lattice symmetry and, moreover, to react sensitively to anisotropic changes of the lattice parameters. Such an anisotropic behaviour of the lattice constants was observed: $c$ shrinks more rapidly under pressure than $a$ [18]. By substitution of Al for Ga the effect is even more pronounced: $a$ rises while $c$ and the cell volume decrease. This behaviour has led us to suggest a critical value of the $c$ parameter where the RKKY interaction switches from FM to AFM stacking of the planes perpendicular to $c$ [15, 19]. The pressure needed for that critical $c$-value in $CePd_2Ga_3$ was estimated as 1.8 GPa.

Further support for our assumptions is drawn from the behaviour of the stoichiometric compound $CePd_2Al_2Ga$ crystallizing in the same hexagonal structure [20]. $CePd_2Al_2Ga$ is FM at $p = 0$, too, and changes its magnetic properties under much lower pressure. From measurements of the PM susceptibility above the ordering temperature there is evidence for a change from FM to AFM order already near 0.3 GPa [19]. Beyond that pressure $T_{Mag}$ rises again, passes a maximum, and finally disappears close to 2.8 GPa [21]. It is remarkable that the NFL behaviour found in the specific heat corresponds to that of a 3d antiferromagnet [21]. If a qualitative picture of corresponding states in the similar compounds is applicable, this outcome gives a strong argument in favour of our assumption that $CePd_2Ga_3$ is an AFM beyond 2.8 GPa.

In summary, the search for a *ferromagnetic* QCP in $CePd_2Ga_3$ was not successful. Of course, there is still need for more experimental confirmation, e.g. by resistance measurements to fill the gap in the data between 1.5 and 2.7 GPa and further on to 3.5 GPa, or by specific heat measurements beyond 2.4 GPa. Great progress would be made if single crystals were available. Most important would be a determination of the magnetic structure under pressure by neutron scattering; this is a great challenge because of the combination of small magnetic moments, high pressure, and low temperatures. So far the assumption of [5] appears to be corroborated, that increasing hybridization tends to destabilize FM order in favour of an AFM one. As long as there exists no definite theoretical proof of this hypothesis, the search for other FM Kondo lattices close to a QCP should be continued.

## Acknowledgment

The support by the Deutsche Forschungsgemeinschaft is very gratefully acknowledged.

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