Vol. 34 (2003) *ACTA PHYSICA POLONICA B* No 2

SEARCH FOR A QUANTUM CRITICAL POINT IN $CePd_2Al_2Ga$ BY SPECIFIC HEAT MEASUREMENTS UNDER PRESSURE*

H. Neemann, A. Eichler

Institut für Technische Physik, Technische Universität Braunschweig
Mendelssohnstrasse 2, 38106 Braunschweig, Germany

S. Süllow

IMNF, Technische Universität Braunschweig
Mendelssohnstrasse 3, D-38106 Braunschweig, Germany

and J.A. Mydosh

Kamerlingh Onnes Laboratory, Leiden University, Leiden, The Netherlands

(Received July 10, 2002)

In $CePd_2Ga_3$, one of the few ferromagnetically ordered heavy fermion materials, the transition temperature $T_{\rm mag}$ ($\sim$ 6.4 K) is lowered by pressure. From a comparison of resistance data with DC susceptibility data there is evidence, however, that the FM magnetization disappears before $T_{\rm mag}$ is suppressed to zero. The isostructural compound $CePd_2Al_2Ga$ also shows FM order, but with a much lower $T_{\rm mag} \sim 1.8$ K. It could be expected, therefore, to reach $T_{\rm mag} = 0$ K within a pressure range where thermodynamic properties like χ and C_p can be determined. Earlier data of the ac susceptibility have shown that after an initial steep decline $T_{\rm mag}$ saturates with a tendency to increase again above 0.4 GPa. Here we present specific heat data extending up to 2.5 GPa which confirm this dependence as a bulk property of $CePd_2Al_2Ga$. Moreover, they show a maximum in $T_{\rm mag}$ around 1.8 GPa followed by a steep decline extrapolating to 0 K below 2.8 GPa. This decline is accompanied by a flattening of the transition anomaly above 1.9 GPa. Notwithstanding the hints on a change of the type of magnetic order from ac susceptibility results the magnetic structure under pressure is not known. So the interesting question if there exists a quantum critical point between a FM phase and a Kondo Fermi liquid is still open.

PACS numbers: 71.27.+a, 75.40.–s

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

A great number of antiferromagnetic (AFM) Kondo lattice materials can be described by the Doniach model in a very satisfactory way. According to this model an increase of the interaction parameter $g = N(E_F)J$ will drive the material from a magnetically ordered into a paramagnetic (PM) Fermi-liquid state. The increase in g can be attained by suitably alloying (x) or — in cerium alloys — by application of pressure (p). At critical values x_C or p_C, respectively, a so called Quantum Critical Point (QCP) is reached where $T_N = 0$. In the proximity of this QCP the physical properties are determined by quantum fluctuations. A description as a Fermi-liquid is not appropriate. For the temperature dependences of the specific heat, magnetic susceptibility and electrical resistance unusual power or logarithmic laws are found [1]. The study of QCPs under high pressure is especially useful since pressure does not change the chemical composition nor the atomic order.

There are only comparatively few examples of ferromagnetically (FM) ordering Kondo-lattices though to our knowledge there are no principial reasons against the occurrence of FM in materials showing Kondo-interactions. Rather often the magnetic phase diagrams of FM Kondo-lattices are more complicated; beyond the FM phase there exist different magnetic phases with the consequence that there is no direct transition from the FM to the PM state (as an example see $CeRu_2Ge_2$ [2]). Because of the limited choice of FM Kondo-lattices, the probability is rather low to find one with a p_C low enough that an extensive investigation of the bulk properties can be performed under pressure.

$CePd_2Ga_3$ is a FM cerium compound with clear signs of Kondo effect above the Curie temperature $T_C \sim 6.4$ K [3]. Its p_C appears to be around 5 GPa as was extrapolated from resistance data under pressure [4]. On the other hand DC susceptibility measurements under pressure have shown that the signs of FM abruptly disappear at 2.3 GPa [5]. Combined with the resistance data this finding suggests that the FM order changes to another type of magnetic order before the transition to the PM state is reached. To supplement the investigation of this phenomenon by measurements of ac susceptibility and specific heat is very difficult according to the high pressures needed. On the other hand, an approach to the QCP by continuously alloying into $CePd_2Ga_3$ is not possible [6]. There exists, however, a stoichiometric intermetallic compound $CePd_2Al_2Ga$ which also crystallizes in the hexagonal $PrNi_2Al_3$ structure and shows FM order below about 1.8 K [6]. Thus the pressure needed to suppress the FM order is expected to be much less than in $CePd_2Ga_3$. Indeed, ac susceptibility measurements under pressure have shown a steep incipient drop of T_C which stopped, however, near 0.07 GPa [7]. After staying constant T_{mag} started to rise again beyond 0.4 GPa, thus leaving the question of the possible existence of a QCP in this type of material open again.

Here we present results of specific heat measurements on $CePd_2Al_2Ga$ under high pressure which where performed to get additional informations on the state of the material in the bulk. The technique used was a relaxation method applied to the sample configuration in the interior of a piston cylinder cell. An overview on the results is shown in Fig. 1. The magnetic

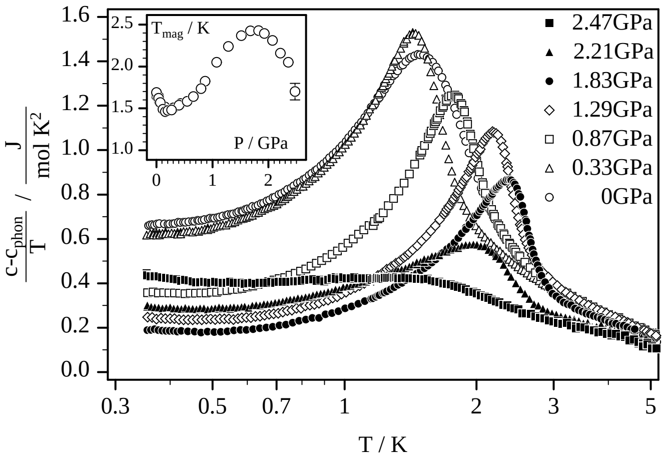

Fig. 1. Magnetic specific heat in a logarithmic scale. Insert: $T_{\rm mag}$ as defined by the position of the maximum in $C(T)$.

transition is clearly pronounced; there is no evidence for any other transition in the whole temperature range. The non monotonous dependence $T_{\rm mag}(p)$ as found in the susceptibility measurements [7] is confirmed in an excellent way and continued to much higher pressure. Around 1.8 GPa $T_{\rm mag}$ reaches its maximum value of about 2.4 K. At higher pressures $T_{\rm mag}$ falls again with increasing slope. Beyond 2.3 GPa, however, the determination of $T_{\rm mag}$ from the anomaly becomes more difficult. At $p = 0$ the transition anomaly already shows some intrinsic broadness, which diminishes a bit with increasing pressure. Only around 2.3 GPa it starts to increase distinctly accompanied by a pronounced decrease of the height of the anomaly. The critical state seems to be beyond the technical limit of our pressure cell and has not yet been reached. From extrapolation of the $T_{\rm mag}(p)$ data the limit $T_{\rm mag} = 0$ is expected to lie between $p = 2.6$ and $p = 2.8$ GPa. The magnetic entropy as determined by an integration of the $(C - C_{\rm phon})/T$ data is rather small and amounts to only $0.6\,R\ln 2$ at 6 K and $p = 0$. Under pressure it is further diminished.

There are no doubts on the existence of a magnetically ordered phase within the whole pressure range investigated. On the nature of the magnetic order, however, we still can only speculate more or less. At $p = 0$ $CePd_2Al_2Ga$ is definitely FM as shown by magnetization measurements [6].

At the same time the signal of the ac susceptibility is very strong as often found in FM materials. It decreases rapidly under pressure and becomes very weak beyond 0.3 GPa suggesting a change in the type of magnetic order [7]. Arguments in favor of a change of the type of magnetic order are based on the peculiarities of the $PrNi_2Al_3$ structure. The cerium moments are FM aligned within the basal planes of the hexagonal unit cell. Long range order is mediated by RKKY coupling across the intermediate layers composed of Al/Ga and can be FM ($CePd_2Ga_3$, $CePd_2Al_2Ga$) or AFM ($CePd_2Al_3$). The idea is that the magnetic LRO can be switched from FM to AFM by tuning the anisotropic RKKY interaction via the c-axis lattice parameter [6, 7]. If this is the main influence a common pressure phase diagram should hold for $CePd_2(Ga,Al)_3$. At least, the specific heat data on the shape of the transition anomaly seem to exclude the presence of a spin glass state. There are, however, no distinct indications for a change in the magnetic ordered state like for instance a change in the shape of the transition anomaly as was found in $CePb_3$, *e.g.* [8]. Measurements of the contribution of magnetic excitations to the electrical resistance at very low temperatures seem to indicate the disappearance of FM close to 0.4 GPa [9]. The best method for the determination of the magnetic order of a material is neutron scattering. Presently such investigations, however, are beyond any technical feasibility because of the high pressures and low temperatures needed, the small reduced magnetic moments, and the fact that single crystals are not available.

So the conclusion of our study is that there is a QCP in $CePd_2Al_2Ga$ close to $p = 2.8$ GPa. But the question is still open if this is a direct transition from the FM to the PM state or if there is an AFM phase in between.

The support of this work by the Deutsche Forschungsgemeinschaft is gratefully acknowledged.

REFERENCES

[1] H. v. Löhneysen, *J. Magn. Magn. Mater.* **200**, 532 (1999); G.R. Stewart, *Rev. Mod. Phys.* **73**, 797 (2001).
[2] S. Süllow *et al., Phys. Rev. Lett.* **82**, 2963 (1999).
[3] E. Bauer *et al., Z. Phys.* **B92**, 411 (1993).
[4] R. Hauser *et al., Physica* **B206 & 207**, 231 (1995).
[5] T. Burghardt, E. Hallmann, A. Eichler, *Physica* **B230–232**, 214 (1997).
[6] B. Ludoph *et al., Physica* **B223 & 224**, 351 (1996).
[7] T. Burghardt *et al., Physica* **B259–261**, 99 (1999).
[8] T. Kirsch, A. Eichler, P. Morin, U. Welp, *Z. Phys.* **B86**, 83 (1992).
[9] J.A. Larea J. *et al.,* this conference.