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Evidence for a metamagnetic transition in the heavy Fermion system CeTiGe

M. Deppe[1], N. Caroca - Canales[1], J. G. Sereni[2], C. Geibel[1]

[1] Max Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany
[2] Lab. Bajas Temperaturas, CNEA, 8400 S.C. de Bariloche, Argentina

E-mail: Micha.Deppe@cpfs.mpg.de

Abstract. A recent study of CeTiGe identified this compound as a paramagnetic heavy Fermion system where the full $J = 5/2$ multiplet is involved in the formation of the ground state. Here we present a preliminary investigation of the dc-magnetization $M_{\rm dc}(H)$ and of the magnetoresistance $\rho(H)$ of polycrystalline CeTiGe samples in applied magnetic fields up to $\mu_0 H = 14$ T. The results reveal a pronounced metamagnetic transition at a critical field $\mu_0 H_c \approx 13.5$ T at low temperatures, with a step like increase in $M_{\rm dc}(H)$ of at least 0.6 $\mu_{\rm B}$/Ce. The metamagnetic transition leads to a strong decrease in $\rho(H)$. A clear hysteresis in $M_{\rm dc}(H)$ and $\rho(H)$ indicate that in CeTiGe these metamagnetic features correspond to a true thermodynamic, first order type transition in contrast to the critical behavior observed in the canonical system $CeRu_2Si_2$. Measurements at higher temperatures showed a continuous suppression of the metamagnetic transition with increasing T, which vanishes at $T \sim 30$ K.

1. Introduction

Systems which are located close to a ferromagnetic or antiferromagnetic quantum critical point are of high interest, because the low lying quantum fluctuations can lead to new/exotic physical phenomena like e. g. unconventional (non-Fermi liquid) metallic or unconventional superconducting states [1]. On the other hand, the reduction of the spatial dimensionality also results in an increase of quantum fluctuations and favors the development of unconventional superconductivity [2, 3]. Therefore Kondo lattices with a quasi-two-dimensional structure are of high interest. In this context CeTiGe is an attractive compound: It crystallizes in the CeFeSi structure type which presents a stacking of Ce-Si-Fe2-Si-Ce layers leading to a clear 2D character [4]. Furthermore preliminary investigations performed by R. Welter et al. [5] indicated the presence of a trivalent Ce^{3+} state without magnetic order down to 4.2 K. Our recent detailed study of the physical properties down to 0.4 K established CeTiGe as a new paramagnetic heavy Fermion system with a Sommerfeld coefficient $\gamma \simeq 0.3\ J/(K^2 \cdot mol)$ [6], being thus located on the non-magnetic side of the (quantum) critical point within a Doniach type phase diagram. Maxima in $\chi(T)$ at 25 K and in C_{4f}/T at 16 K indicate a larger degeneracy of the involved local moment. Both $\chi(T)$ and C_{4f}/T could be well fitted with the prediction for the Coqblin-Schrieffer (CS) model for $J = 5/2$ with $T_0 = 82$ K, corresponding to a Kondo scale $T_{\rm K} = 55$ K [6]. For $J = 5/2$ one expect within the CS model a weak S-shaped curve in the magnetization $M(H)$ [7]. We therefore performed preliminary magnetization measurements in a commercial equipment. To our surprise, instead of the expected weak S-shaped $M(H)$

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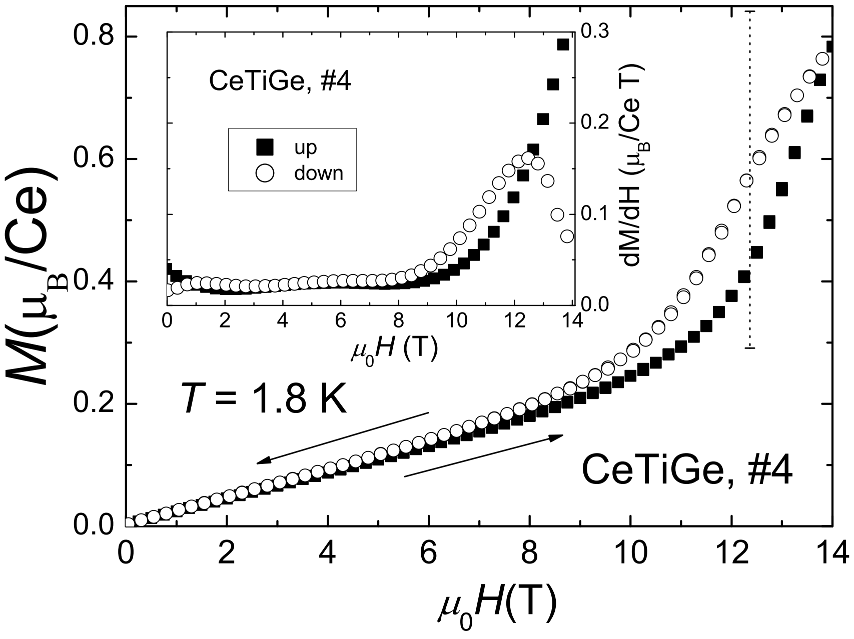

Figure 1. DC-magnetization of CeTiGe sample ♯4 measured in fields up to $\mu_0 H = 14$ T at $T = 1.8$ K. The arrows indicate the field-runs with increasing and decreasing field. A steplike increase of $M_{\rm dc}(H)$ sets in at $\mu_0 H \sim 12$ T upon increasing field. The hysteresis evidences a first order type transition. The inset shows the derivative $\mathrm{d}M/\mathrm{d}H$ vs. $\mu_0 H$ up to 14 T. A clear peak at 12.5 T marks the metamagnetic transition in the downwards measurement, while with increasing field a slight field shift to higher field values is observed.

we observed evidence for a pronounced metamagnetic transition (MMT), which bears some similarity with that observed in the archetypical Kondo lattice system $CeRu_2Si_2$ [8].

2. Materials and experimental techniques

Several polycrystalline CeTiGe samples were prepared by first melting the constituent elements (Ce, Ti, Ge) in an arc-furnace under argon atmosphere. Every pellet was homogenized by flipping and remelting several times. Because of a large amount of foreign phases in the as-cast ingots due to a strong peritectic reaction [6], the different batches (♯1, ♯4, ♯5) were then annealed for four days at 1273 K, 1373 K and 1423 K, respectively. The results of powder X-ray diffraction confirmed the tetragonal CeFeSi-type structure (space group P4/nmm) with lattice parameters close to the literature values [5, 6]. Microprobe analyses indicate a stochiometric composition of the main CeTiGe phase for all batches. For the measurements of the magnetoresistance $\rho(H)$ and the dc-magnetization $M_{\rm dc}(H)$ bars were cut from the different batches. Samples denoted as ♯1 are from the same batch as in [6], while ♯4 and ♯5 are from a new batch series. Despite the different annealing conditions, all batches presented very similar properties. $\rho(H)$ was measured with the conventional four-probe ac technique of the PPMS (Quantum Design), while $M_{\rm dc}(H)$ was measured with the AC/DC magnetometry insert using the extraction technique.

3. DC - magnetization and magnetoresistance of CeTiGe

Fig. 1. shows the result of the $M_{\rm dc}$ vs. H measurement at $T = 1.8$ K for the sample ♯4 with $m =$ 11 mg. The magnetization increases linearly until $\mu_0 H = 9$ T, where a pronounced upturn sets in, much more pronounced than expected within the CS-model. This almost step-like increase in the magnetization extends up to the maximum field of 14 T available in this equipment. The large slope at 14 T indicates that the saturation magnetization has not been reached. Therefore we can only give a lower bound $\Delta M > 0.6\ \mu_{\rm B}$/Ce for the magnetization step connected with the MMT. The measurement with decreasing field reveals a similar behavior, but starts with a smaller slope which result in the opening of a hysteresis with a shift in the field of $\Delta\mu_0 H \sim$ -1.5 T. The inset of Fig. 1. shows the derivative dM/dH for both increasing and decreasing field. In the later case we get a peak in dM/dH at $\mu_0 H \sim 12.5$ T, while in the former case the maximum is above the limit of the measurements. This suggests a critical field $\mu_0 H \sim 13.5$ T $\pm$ 1T, which shall certainly depend on the field orientation respective to the crystal axes. The hysteresis loop with a width of $\Delta\mu_0 H \sim$ -1.5 T is a first indication that the MMT in CeTiGe is of first order type. However, at this step one cannot exclude this hysteresis to origin from a canting of the sample during measurements. In Fig. 2a. we show similar $M(H)$ data of sample ♯1 with $m = 52$ mg, but now at different temperatures to study the T dependence of the MMT. While the onset of the MMT seems to be independent of T, the slope of the step-like increase in $M(T)$ becomes weaker resulting in a smaller $M(H)$ value at 14 T. This suggests that the MMT becomes weaker and/or shifts to higher temperatures. At 30 K the slope $M(H)$ is constant up to 14 T suggesting that at this temperature the MMT is completely suppressed. In order to study the MMT and the first order nature in a simple measurement where the sample can be fixed very tightly, we determined the magnetoresistance $\rho(H)$. Fig. 2b. shows $\rho(H)$ of sample ♯5 for different temperatures $T = 1.8$, 5 and 10 K. For sake of clarity $\rho(H)$ was normalized to the $\rho(T)$ value in zero field to compensate for the strong increase in $\rho(T)$ between 1.8 K and 10 K [6]. At $T = 1.8$ K we observe a large positive magnetoresistance, which first increases $\propto H^2$ and then evolves to a nearly linear field dependence in the range 4 T $< \mu_0 H <$ 11 T. At $\mu_0 H = 12.5$ T a sharp decrease sets in, marking the onset of the MMT. The large value of the positive magnetoresistance (+65 %) just before the onset of the MMT is an indication for the high quality of our CeTiGe samples. The large negative slope at 14 T confirms that the transition is not completed at this field. The curve for decreasing field starts with a positive slope, which results in a large hysteresis which closes at $\mu_0 H = 8$ T. Thus the magnetoresistance data confirm the first order nature of the MMT. Increasing the temperatures lead to reduction of the relative magnetoresistance for field below the MMT transition, as expected for standard mechanisms, while the drop associated with the MMT is even more pronounced at 5 K than at 1.8 K. Increasing T to 10 K is necessary to significantly decrease the size of both the anomalies and of the hysteresis associated with the MMT.

4. Conclusions

Our preliminary measurements of the magnetization and magnetoresistance reveal a pronounced metamagnetic transition in CeTiGe at $\mu_0 H \simeq 13.5$ T at low temperatures, with a magnetization step of at least $\Delta M > 0.6\ \mu_{\rm B}$/Ce. A well developed hysteresis in both type of measurements indicate a first order type transition. The broadness of the transition in our data is very likely related to the anisotropy of the critical field for the different crystallographic directions. The excellent reproducibility for different samples as well as the nice correspondence between the magnetization and magnetoresistance data proves the intrinsic nature of this MMT. Increasing the temperature lead to the suppression of the MMT in the T range between 10 K and 30 K. While in the prototype system $CeRu_2Si_2$ the metamagnetic transition at $\mu_0 H \simeq 7.8$ T is discussed as a crossover transition [9], the hysteresis we observed in CeTiGe indicate that in this system the MMT is a real thermodynamic phase transition. It is also different from the MMT

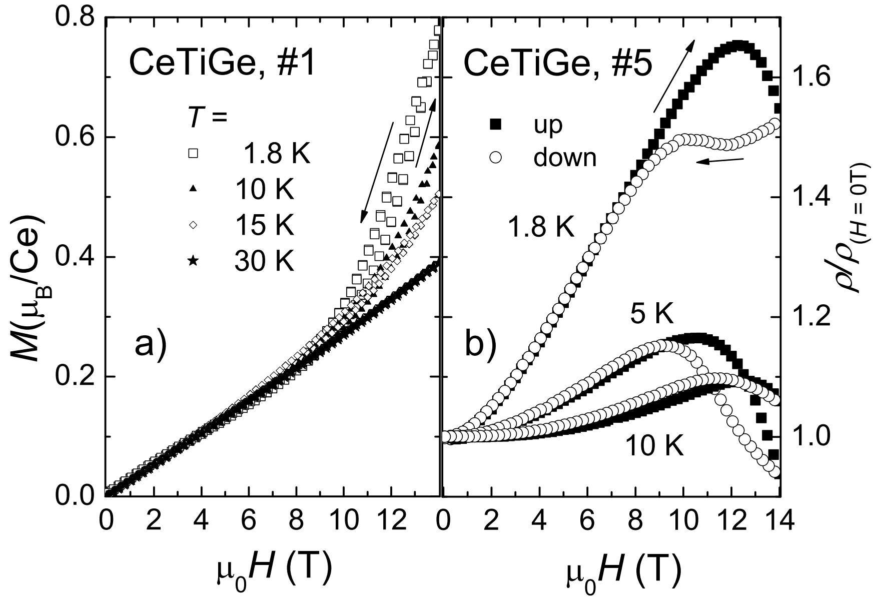

Figure 2. a) DC-magnetization of CeTiGe sample ♯1 measured at $T = 1.8$, 10, 15 and 30 K in field up to $\mu_0 H = 14$ T. The arrows indicate increasing and decreasing field. At 30 K the MMT disappears. Fig. 2b) $\rho(H)$ measured on sample ♯5 at $T = 1.8$ K, 5 K and 10 K with increasing and decreasing field. The MMT is associated with a strong decrease in $\rho(H)$.

observed in the isostructural CeGoGe which corresponds to a classical spin-flip transition from an antiferromagnetic ordered to a saturated ferromagnetic state [10]. Thus CeTiGe presents a likely new type of metamagnetic transition, which further on occurs in a quite accessible field range < 20 T and therefore provides a new excellent opportunity to study this kind of unusual transitions in paramagnetic heavy Fermion systems. We are now extending our investigations to higher fields and further physical properties.

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