Magnetoresistance investigation on single crystalline Ce$_3$Pd$_{20}$Si$_6$ across the temperature-magnetic field phase diagram

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Abstract. The heavy fermion cage compound Ce$_3$Pd$_{20}$Si$_6$ crystallizes in a cubic crystal structure with two inequivalent Ce sites. It undergoes two phase transitions at $T_Q = 0.5$ K and $T_N = 0.31$ K which are tentatively attributed, respectively, to antiferroquadrupolar and to antiferromagnetic order. Recent specific heat investigations on single crystals detected additional anomalies within the antiferroquadrupolar phase, for a magnetic field applied along the [100] direction, suggesting the presence of a tricritical point at around 2.3 T and 0.5 K. We performed isothermal magnetoresistivity investigations (down to 0.055 K) on single crystalline Ce$_3$Pd$_{20}$Si$_6$ across the $B - T$ phase diagram to gain further insight on this new feature.

1. Introduction

The heavy fermion cage compound Ce$_3$Pd$_{20}$Si$_6$ crystallizes in the cubic Cr$_{23}$C$_6$-type structure with space group $Fm\overline{3}m$ and 116 atoms per unit cell [1]. The Ce atoms occupy two distinct sites of cubic point symmetry. At the 4$a$ site ($O_h$ symmetry) the Ce atoms are placed in a cage formed by 12 Pd and 6 Si atoms, whereas at the 8$c$ site ($T_d$ symmetry) the Ce atoms are surrounded by 16 Pd atoms. Its cubic crystal structure places Ce$_3$Pd$_{20}$Si$_6$ in the barely-explored three-dimensional part [2] of the global phase diagram of quantum critical heavy fermion system [3, 4].

Polycrystalline samples were extensively investigated. Two successive ordering temperatures were detected at $T_Q = 0.5$ K and at $T_N = 0.31$ K and tentatively attributed to antiferroquadrupolar (AFQ) and to antiferromagnetic (AF) order, respectively [5]. $T_N$ is suppressed to zero at a critical field around 0.9 T. At this field-induced quantum critical point (QCP) transport features of Kondo destruction are observed [2].

Magnetization, ultrasound and specific heat investigations on single crystalline samples revealed a strongly anisotropic response with respect to an external magnetic field $B = \mu_0 H$ above 1 T applied along the [100], [110] or [111] crystallographic direction [6, 7, 8]. Below 1 T, the phase diagram is isotropic. In particular, $T_N$ is suppressed to zero isotropically for all three directions with $B_N^c \sim 0.8$ T, whereas a full suppression of $T_Q$ is only reached at 4 T and 10 T for magnetic fields applied along [100] and [110], respectively. Interestingly, only for fields along [100] the specific heat shows an anomaly at $T_x$ within the AFQ phase. In Ref. [8] $T_x$ was...
interpreted as a genuine phase transition, which thus was suggested to separate two ordered phases, phase II and II’, inside the AFQ phase [8] (see Fig. 3). The nature of the two phases II and II’ remains to be elucidated.

In this work, we present isothermal magnetoresistivity measurements (down to 0.055 K) on two single crystalline samples of Ce₃Pd₂₀Si₆ under an external magnetic field between 0 and 15 T, applied parallel to the [100] and to the [110] crystallographic direction. Our aim is to explore the $B - T$ phase diagram to advance the understanding of the new feature at $T_x(B)$ or equivalently at $B_x(T)$.

2. Experimental

The single crystalline Ce₃Pd₂₀Si₆ samples investigated in this work were synthesized by growth from a slightly off-stoichiometric melt [9, 10]. X-ray diffraction and SEM/EDX investigations show that the crystals are single phased and have the correct stoichiometric composition. The temperature-dependent specific heat $c_p(T)$ of these samples shows two successive transitions at $T_Q \approx 0.4$ K and $T_N \approx 0.2$ K [10], values which are close to those reported in previous studies on single crystalline Ce₃Pd₂₀Si₆ [6, 7, 8]. The two transition temperatures determined for these single crystals are slightly lower than in the best polycrystals, presumably due to a very slight off-stoichiometry of the single crystals [9].

The transverse magnetoresistivity $\rho(B)$ was investigated on two samples with current along [001], and with the magnetic field applied perpendicular to the transport direction and parallel to the [100] and [110] crystallographic direction. The dimensions of the two samples are $4.5 \times 2 \times 0.3$ mm$^3$ and $1.1 \times 0.5 \times 0.3$ mm$^3$, respectively. $\rho(B)$ was measured using a standard 4-point configuration with a low-frequency ac current. The samples were polished to guarantee good thermal contact between their surface and the sample-holder at low temperatures and the electrical contacts were realized by spot-welding to minimize the contact resistance. The sample holder was anchored to a cold finger, thermally connected to the mixing chamber of a dilution refrigerator (Kelvinox-400, Oxford Instrument), with a superconducting coil which can supply magnetic fields up to 15 T. A RuO$_2$ sensor was used to determine the temperature of the samples down to 0.05 K; it was calibrated under magnetic field and presents an accuracy better than 1%. The measurements were performed by using low-temperature transformers (LTT-m, Cambridge Magnetic Refrigeration), installed on the 1-K pot stage of the dilution refrigerator.

3. Results and discussion

Figure 1 shows isothermal resistivity curves with the external magnetic field applied along the [100] (Fig. 1 (a)) and the [110] (Fig. 1 (b)) crystallographic direction. For both directions the magnetoresistance $MR = \frac{\rho(B) - \rho(0)}{\rho(0)}$ is small and positive in the AF phase below $T_N(B)$ (see Fig. 3). Outside the AF phase, the MR becomes first negative and then changes again to positive values above a certain magnetic field that increases with temperature.

Interestingly, the isothermal curves in Fig. 1 show additional features for both crystallographic directions, outside the AF phase. Only when $B \parallel [100]$ and in the magnetic field range where the MR is negative, a small shoulder can be observed below 0.4 K at a magnetic field around 2 T (marked with arrows in Fig. 1 (a)). This feature is absent for $B \parallel [110]$. On the other hand, in the regime where the MR is positive and only for $B$ applied along [110], a shoulder can be seen for the $T = 0.7$ K isotherm at $B \approx 9$ T. This feature gets weaker and shifts to higher $B$ values as temperature decreases approaching $B \approx 10$ T for 0.055 K (see arrows in Fig. 1 (b)). In comparison with the $T - B$ phase diagram obtained by magnetization [6], ultrasound [7] and specific heat [8] measurements, the latter feature observed in Fig. 1 (b) can be associated with the AFQ ordering transition, which is expected to be suppressed at the critical magnetic field
Figure 1. (color online) Isothermal magnetoresistivity of single crystalline Ce$_3$Pd$_{20}$Si$_6$. (a) For magnetic field applied along [100], the arrows are placed at $B_k$ (see text). (b) For magnetic field applied along [110] the arrows indicate the magnetic field where the AFQ phase is suppressed. Solid lines in (a) and (b) show the metallic background curve estimated for the two directions (see text). $B_{Q} \sim 10$ T when $B \parallel [110]$ (Ref. [8]).

To study the feature around 2 T ($B_k$) with higher accuracy, the metallic background contribution $\rho_{bg}(B)$ was estimated using $\rho(B, T = 0.055$ K) as specified in the following. $\rho_{bg}(B)$ follows a $H^2$ dependence in the field range 4.6 - 6.4 T for $B \parallel [100]$, and in the field range 4.6 - 8 T for $B \parallel [110]$. This quadratic-in-$H$ behavior is frequently found to be the leading term of the magnetoresistivity of non-magnetic metals [11]. $\rho_{bg}(B)$ is determined as

$$\rho_{bg}(H) = \begin{cases} 
31.8 + 0.064(\mu_0 H)^2 \ [\mu\Omega\text{cm}] & \text{for } 0 \ T \leq H \parallel [100] \leq 6.4 \ T \\
27.6 + 0.065(\mu_0 H)^2 \ [\mu\Omega\text{cm}] & \text{for } 0 \ T \leq H \parallel [110] \leq 8 \ T
\end{cases}$$

where the coefficients were obtained by least-squares fitting of $\rho(B, 0.055$ K). The quadratic behavior was then extrapolated to $\mu_0 H = 0$ T. The resulting background curves $\rho_{bg}(B)$ are plotted in Fig. 1 (a) and (b) as solid lines.

The difference of the raw data and the background contribution $(\rho - \rho_{bg})(B)$ is plotted
Figure 2. (color online) Isothermal resistivity curves after the subtraction of the background contribution $\rho_{bg}$ for magnetic field applied along $\parallel [100]$ (left) and along $\parallel [110]$ (right). The arrows indicate the position of the kinks at $B_k$.

in Fig. 2 with $B$ applied along the [100] (Fig. 2 (a)) and the [110] (Fig. 2 (b)) direction for all investigated temperatures.

In Fig. 2 the evidence of the kink at $B_k$ defined above for the [100] direction can be clearly confirmed for the 0.055, 0.2 and 0.32 K isothermal curves (arrows in Fig. 2). Thus, our experiment supports the presence of the anomaly at $T_x(B)$ [8]. For higher temperatures this feature broadens.

In order to compare these results with published data we first rescale all the characteristic temperatures and fields of the $B - T$ phase diagram obtained by $c_p$ [8] as described in the following. The magnetic ($T_N$) and the quadrupolar ($T_Q, T_x$) ordering temperatures obtained by $c_p$ are multiplied by 0.75 and 0.9, respectively. These factors were chosen to scale the $T_N$ and $T_Q$ ordering temperatures of the samples investigated in Ref. [8] to the values measured for our samples at $B = 0$ T [10]. Consequently, also the field values of all points in the $B - T$ phase diagram were renormalized by the same factors (Fig. 3). The kink positions $T_k(B)$ determined here are shown as red open symbols. They closely follow the $T_x(B)$ line determined by the $c_p$ measurements [8].

4. Conclusion

We have investigated the magnetoresistivity on single crystalline Ce$_3$Pd$_{20}$Si$_6$, with external magnetic field applied parallel to the [100] and to the [110] direction.

Only for $B$ applied along the [100] direction, we detect a kink $B_k(T)$ around 2 T, for temperatures $T \leq 0.32$ K. The kink temperature $T_k$ decreases with decreasing field, extrapolating to zero at 1.8 T, in excellent agreement with the line $T_x(B)$ detected by specific heat measurements [8]. Thus, whatever the nature of the boundary $B_x$ separating the two putative phases $II$ and $II'$ is, it has marked influence on the charge carriers in Ce$_3$Pd$_{20}$Si$_6$. Thus, further investigations of electronic transport across the $T - B$ phase diagram with $B \parallel [100]$ are called
Figure 3. (color online) $B - T$ phase diagram. $T_N$, $T_Q$ and $T_x$, from Ref. [8], as well as their magnetic field values are scaled by factors 0.75, 0.9, and 0.9, respectively, according to the values of $T_N$ and $T_Q$ of our samples at $B = 0$ T. The red open circles represent the kinks observed in our magnetoresistivity investigation $B_k(T)$.

for. In particular, the full suppression of $T_k$ at around 1.8 T might lead to another quantum critical point in this phase diagram, an aspect that deserves further attention.

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References
[1] Gribanov A V, Seropegin Y D, and Bodak O J 1994 *J. Alloys Compd.* 204 L9.
[2] Custers J, Lorenzer K A, Müller M, Prokofiev A, Sidorenko A, Winkler H, Strydom A M, Shimura Y, Sakakibara T, Yu R, Si Q, and Paschen S 2012 *Nature Mater.* 11 189.
[3] Si Q 2010 *Phys. Status Solidi B* 247 476.
[4] Si Q and Paschen S 2012 *Phys. Status Solidi B* 250 425.
[5] Strydom A M, Pikul A, Steglich F, and Paschen S 2006 *J. Phys. Conf. Ser.* 51 239.
[6] Goto T, Watanabe T, Tsuduku S, Kobayashi H, Nemoto Y, Yanagisawa T, Akatsu M, Ano G, Suzuki O, Takeda N, Doenni A, and Kitazawa H 2009 *J. Phys. Soc. Jpn.* 78 024716.
[7] Mitamura H, Tayama T, Sakakibara T, Tsuduku S, Ano G, Ishii I, Akatsu M, Nemoto Y, Goto T, Kikkawa A, and Kitazawa H 2012 *J. Phys. Soc. Jpn.* 79 0747121.
[8] Ono H, Nakano T, Takeda N, Ano G, Akatsu M, Nemoto Y, Goto T, Doenni A, and Kitazawa H 2013 *J. Phys.: Condens. Matter* 25 126003.
[9] Prokofiev A, Custers J, Kriegisch M, Laumann S, Müller M, Sassik H, Svagera R, Waas M, Neumaier K, Strydom A M, Paschen S 2009 *Phys. Rev. B* 80 235107.
[10] Prokofiev A and Paschen S 2012 *Crystal Growth and Stoichiometry of Strongly Correlated Intermetallic Cerium Compounds* (Modern Aspects of Bulk Crystal and Thin Film Preparation) ed Kolesnikov N and Elena E (InTech) chapter 11 pp 263-284.
[11] Pippard A B 2009 *Magnetoresistance in metals* (Cambridge University Press).