Low-temperature electrical resistivity of antiferromagnetic

\[ \text{Ce}_4\text{Pt}_{12}\text{Sn}_{25} \]

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Abstract. The compound \( \text{Ce}_4\text{Pt}_{12}\text{Sn}_{25} \) has a rather complex cubic crystal structure in which the Ce atoms are encapsulated in cages of Pt and Sn resulting in a large Ce-Ce distance. Recently, specific heat and magnetic susceptibility measurements revealed a phase transition from a paramagnetic to an antiferromagnetic (AFM) ground state with a low Néel temperature of \( T_N = 0.19 \) K. Application of a magnetic field was shown to weaken the AFM phase and ultimately suppress it at a critical field of 0.7 T. Here we investigate the observed antiferromagnetic phase transition by electrical resistivity measurements in applied magnetic fields in the vicinity of this field-tuned quantum critical point.

1. Introduction

Landau Fermi liquid (LFL) theory successfully describes the low-temperature behaviour of many metals, even of strongly correlated heavy fermion metals. However, at quantum critical points the LFL description has been shown to break down and non-Fermi liquid (NFL) behaviour arises \[1\].

\( \text{Ce}_4\text{Pt}_{12}\text{Sn}_{25} \) represents an interesting example of a strongly correlated cage compound. In the simple cubic crystal structure Ce atoms occupy a unique position inside a cage formed by Sn and Pt atoms (see upper inset of Fig. 1) \[2, 3\]. Despite the large Ce-Ce next-neighbour distance of 6.14 Å, that might be expected to favour the Kondo interaction over the RKKY interaction, AFM ordering is observed in \( \text{Ce}_4\text{Pt}_{12}\text{Sn}_{25} \) with a low Néel temperature \( T_N = 0.19 \) K \[4, 5, 6\]. Here we study the electrical resistivity of the material, with focus on the evolution of the AFM transition in magnetic fields.

2. Experimental

Single crystals of \( \text{Ce}_4\text{Pt}_{12}\text{Sn}_{25} \) were grown by the self-flux method. Polycrystalline samples of \( \text{Ce}_4\text{Pt}_{12}\text{Sn}_{25} \) and PtSn\(_4\), produced by repeated melting of elemental Ce (99.95 %), Pt (99.95 %), and Sn (99.9999 %) in a cold copper boat under argon atmosphere, were placed in a high-quality alumina crucible, which was subsequently sealed in a quartz ampoule. After short heat treatment at 900 °C the ampoule was cooled down to 650 °C. At this temperature, the ampoule was centrifuged to separate the single crystals of \( \text{Ce}_4\text{Pt}_{12}\text{Sn}_{25} \) from the PtSn\(_4\) flux. The phase and stoichiometry of the sample were confirmed by powder X-ray diffraction on a few ground single crystals and by SEM/EDAX analysis on bigger crystals. For the electrical resistivity measurements single crystals were aligned along the three principal crystallographic directions with the help of Laue back reflection patterns and cut/polished into small platelets of typical size 1.5 mm × 1.0 mm × 0.40 mm. Four gold wires were spot welded on the surface of each sample. One such sample was glued with GE Varnish to the sample platform of an Oxford MX400 dilution refrigerator, close to a field-calibrated ruthenium oxide temperature sensor. To improve
the resolution of the measurement, the voltage signal was amplified using low-temperature transformers before detecting it with a Linear Research LR700 resistance bridge. Magnetic field is provided by a 15 T superconducting magnet. For temperatures above 2 K this sample was measured with the AC transport option in a PPMS from Quantum Design. For all resistivity measurements the current $j$ was chosen to be parallel to the $[1 0 0]$ direction. For the magnetoresistance measurements the sample was thinned down to 0.18 $\mu$m to gain resolution. The magnetic field $H$ was applied perpendicular to the current $j \perp H$.

3. Zero-field electrical resistivity

Figure 1 shows the temperature dependence of the electrical resistivity, $\rho(T)$, of Ce$_4$Pt$_{12}$Sn$_{25}$ (0.04-295 K) in zero field. Metallic behaviour is observed, with a broad hump around 150 K. The hump might be due to two closely spaced crystal electric field (CEF) excitations that were revealed by specific heat measurements. Indeed the non-magnetic reference compound La$_4$Pt$_{12}$Sn$_{25}$ does not show this feature [4]. The lack of four-fold point symmetry of the surrounding cage at the Ce site gives rise to a splitting of the six-fold degeneracy into three Kramers doublets with a $\Gamma_7$ doublet as the ground state [6]. Between 1.9 K and 4.5 K, $\rho$ is essentially temperature independent. This indicates that both the phonon contribution $\rho_{ph}$ and the resistivity due to spin-disorder and spin-flip scattering $\rho_{mag}$ are negligibly small or independent of temperature. Below 1.9 K, a decrease of $\rho$ sets in, with increasing slope $\partial \rho/\partial T$ as $T$ is lowered. At $T_N = 0.19$ K, a sharp kink is seen in $\rho(T)$, signaling the transition into the AFM state (see lower insert of Fig. 1). Below 90 mK, LFL behaviour $\rho(T) = \rho_0 + AT^2$ is observed (Fig. 2 insert). In order to explain the data up to the Néel temperature, we add a term to account for the scattering of electrons on AFM magnons [7]

$$\rho_{SW}(T) = b\Delta^2 \sqrt{\frac{k_B T}{\Delta}} e^{-\Delta/k_B T} \left[ 1 + \frac{2}{3} \left( \frac{k_B T}{\Delta} \right) + \frac{2}{15} \left( \frac{k_B T}{\Delta} \right)^2 \right].$$

The coefficient $b$ is a material constant that depends on the spin wave stiffness $D$. The energy gap $\Delta$ of the spin wave dispersion is a result of anisotropy, either due to an intrinsic material anisotropy or caused by the presence of a symmetry-lowering external influence like an applied magnetic field [7]. The high symmetry of cubic Ce$_4$Pt$_{12}$Sn$_{25}$ in zero-field would a priori suggest a vanishing energy gap. However, Lee et al. [4] observed a LFL contribution with $A = 11 \mu\Omega cm K^{-2}$ plus a $T^5$ term. The large exponent of the latter clearly indicates the presence of spin waves with a moderate energy gap of the order of $k_B T_N$. Our fit parameters are listed in Tab. 1. The $A$ coefficient we find is slightly larger than the value given in Ref. [4], which is explained by a differently chosen spin wave term. Above $T_N$ the LFL contribution following a $T^2$ dependence is no longer observed. This behaviour is common for magnetically ordered heavy fermion systems near a quantum critical point, see e.g. Ref. [8]. The larger value of $\Delta$ compared to $T_N$ can be interpreted as the onset of spin gap opening at higher temperatures.

Figure 1. Temperature dependence of the electrical resistivity $\rho(T)$ of Ce$_4$Pt$_{12}$Sn$_{25}$. The lower insert displays a close-up of the low-temperature data with the AFM transition indicated by an arrow. The upper insert shows the atomic arrangement in the unit cell of Ce$_4$Pt$_{12}$Sn$_{25}$.
Table 1. Fit parameters for the zero-field resistivity below $T_N$ using the expression $\rho(T) = \rho_0 + AT^2 + \rho_{SW}(T)$ with the spin wave contribution according to Eqn. (1).

| $\rho_0$ ($\mu\Omega\text{cm}$) | $A$ ($\mu\Omega\text{cmK}^{-2}$) | $b$ ($\text{K}^{-2}$) | $\Delta$ ($\text{K}$) |
|-----------------------------|---------------------------------|-----------------|----------------|
| 12.79(1)                   | 14.4(5)                         | 35.7(9)         | 0.54(2)        |

This gives a natural explanation for the observed long tail in the specific heat [6] as well as the decrease of the resistivity below $T = 1.9 \text{ K}$. Alternatively, magnetic frustration and Kondo singlet formation were under discussion as possible origins for the latter observation [4].

4. Field tuning of the AFM transition

Application of magnetic fields along the [0 0 1] direction reduces $T_N$ continuously and finally suppresses it altogether at the critical field of $\mu_0H_N \approx 0.9 \text{ T}$ (Fig. 2). This defines a field-induced quantum critical point (QCP). The electrical resistivity at this field has the non-Fermi liquid form $\Delta\rho \sim T$. Besides the effect of suppression of the AFM state, a positive magnetoresistance is observed, seen in Fig. 2 as an upwards shift of the $\rho(T)$ curves with increasing field.

The field dependence of $T_N$ is best tracked by $\partial\rho/\partial T$, where $T_N$ manifests as a maximum (Fig. 3, left panel). With increasing fields the peak position shifts to lower temperatures and broadens. For fields above $H_N$ a much broader peak appears in $\partial\rho/\partial T$. We fitted the data with

$$
\rho_{CEF}(T) = \frac{3\pi N m}{\hbar e^2 \epsilon_F} J^2 (g_f - 1)^2 \sum_{i,f} \left< m_{s_i}, \Gamma_i \left| \vec{s} \cdot J \right| m_{s_f}, \Gamma_f \right>^2 \frac{2}{1 + e^{-\frac{E_i - \epsilon_{f}}{k_B T}}} e^{-\frac{E_f - \epsilon_{f}}{k_B T}}.
$$

a model that describes the influence of the crystal electric field (CEF) on the spin-disorder scattering of conduction electrons [9]. $N$ is the number of scattering centers, $m$ the effective mass of the charge carriers, $\epsilon_F$ the Fermi energy, $e$ the elementary charge, $J^2 (g_f - 1)^2$ the de Gennes factor, $|\Gamma_i|$ and $E_i$ the crystal field state and its eigenvalue, $|m_s\rangle$ the spin state of the conduction electrons and $k_B$ the Boltzmann constant. An external field lifts the ground state degeneracy by splitting the $\Gamma_7$ doublet into two singlets by the Zeeman energy $\pm g_J \mu_B \mu_0 (\Gamma_7 | J_z \Gamma_7 \rangle H$. Since the first excited CEF level is about 200 K we can neglect contributions from excited states. The fits give a fairly good description of the

Figure 2. Low-temperature electrical resistivity $\rho(T)$. In zero field the resistivity shows a sharp drop at $T = 0.19 \text{ K}$ which we associate with the Neél temperature $T_N$. The grey solid line is a fit to the model described in the text. The insert shows the zero-field resistivity plotted vs $T^2$ and the LFL contribution (dotted line) derived from the model fit. Magnetic fields continuously suppress $T_N$ to zero (black arrows). At the critical field $\mu_0H_N \approx 0.9 \text{ T}$ non-Fermi liquid behaviour, $\Delta\rho \sim T$ (red solid line) is observed. The upwards shift of the $\rho(T)$ curves with increasing field is due to a sizable positive magnetoresistance.
Figure 3. Temperature derivative of the resistivity, $\partial \rho / \partial T$, for the data shown in Fig. 2. At $T_N$, $\partial \rho / \partial T$ shows a pronounced peak (black arrows). The peak’s position is shifted to lower temperatures with increasing field $H$. Above the critical field $H_N$ a second broad feature can be discerned which is supposed to be due to a Zeeman-split CEF doublet. Solid lines are fits to a CEF model discussed in the text.

data (Fig. 3, right panel). For the lowest temperatures and for fields close to $H_N$, the CEF model does not reproduce $\partial \rho / \partial T$ accurately. This is not suprising since the model does not include effects due to critical fluctuations nor to a spin gap.

5. Conclusion
We have performed high-resolution low-temperature electrical resistivity measurements on Ce$_4$Pt$_{12}$Sn$_{25}$ in various magnetic fields to elucidate the evolution of its antiferromagnetic state with field. The zero-field resistivity reveals Landau Fermi liquid behaviour and a gapped spin wave contribution with $\Delta = 0.54$ K. Above $T_N$ a sublinear temperature dependence is observed up to 1.9 K which might be explained by short-range order fluctuations and/or the opening of the observed spin gap $\Delta$.

Field tuning continuously suppresses the transition temperature $T_N$ to zero at the critical field $\mu_0 H_N \approx 0.9$ T which defines a quantum critical point. At this critical field the electrical resistivity shows non-Fermi liquid behaviour with $\Delta \rho \sim T$. The resistivity data in fields above the critical field are described with a crystal electric field model with a Zeeman split ground state doublet. The agreement between data and model is good for magnetic fields sufficiently away from the quantum critical point.

Acknowledgments
We gratefully acknowledge financial support from the European Research Council/ERC Advanced Grant No 227378 and the TU Vienna Doctoral Program Functional Matter.

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