Anisotropic electrical resistivity of the Kondo insulator $CeRu_4Sn_6$

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Anisotropic electrical resistivity of the Kondo insulator $CeRu_4Sn_6$

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Abstract. The intermetallic compound $CeRu_4Sn_6$ has tentatively been classified as anisotropic Kondo insulator. Here we present electrical resistivity measurements on $CeRu_4Sn_6$ single crystals, both along the main directions a and c of the tetragonal crystal structure, and along the diagonal c' of the a-a plane. This direction was selected because $c' = \sqrt{2}a$ differs from c by only 0.2 % , suggesting that the coumpound might alternatively be regarded as quasi-cubic. Amazingly, strong anisotropy is observed not only between a and c but also between c and c'. We analyse the temperature dependent resistivities both with a simple semiconductor model and within a Kondo insulator picture.

1. Introduction

In Kondo insulators, frequently also referred to as heavy fermion semiconductors, a narrow gap (or pseudo gap) opens at low temperatures in the electronic density of states (DOS) at the Fermi level [1]. This is thought to be caused by the hybridization between localized (f-shell) and conduction (p, d-shell) electrons [2].

Most Kondo insulators investigated so far exhibit a cubic crystal structure (e.g., YbB_{12}, SmB_6, FeSi, $Ce_3Bi_4Pt_3$). Only a few materials (e.g., CeNiSn, CeRhSb) are orthorhombic. The anisotropic behaviour of physical properties observed in the orthorhombic compounds was discussed in terms of nodes of the energy gap along certain directions in k-space [3, 4].

The intermetallic compound $CeRu_4Sn_6$ was first synthesized by Das and Sampathkumaran [5]. It crystallizes in a tetragonal crystal structure (space group: $I\bar{4}2m$) with the lattice constants a=6.8810 Å and c=9.7520 Å. Since the diagonal of the tetragonal plane $c'=\sqrt{2}a$ differs from c by less than 0.2 %, the structure may alternatively be approximated by a quasi-cubic cell.

2. Experimental

The polycrystalline starting material for crystal growth was synthesized by melting first Ce and Ru, and subsequently the resulting Ce/Ru alloy with Sn in a horizontal water cooled copper boat using high-frequency heating. The purity of the starting materials was 99.99 % for Ce (Ames Lab) and Ru, and 99.9999 % for Sn. The single crystal growth was performed by the floating zone melting technique using optical heating in a four-mirror furnace of Crystal Corporation. All steps were performed under Ar 99.9999 % protective atmosphere after several purgings with Ar. Since $CeRu_4Sn_6$ melts incongruently we used a self-flux technique [6]. X-ray diffraction

and SEM/EDX investigations revealed that the crystals are single phased and have the correct stoichiometric composition.

Electrical resistivity measurements were done by a standard 4-point technique, with alternating dc current in a physical property measurement system (PPMS) from Quantum Design between 350 K and 2 K, and with low-frequency ac current in a ^{3}He/^{4}He dilution refrigerator from Oxford Instruments at temperatures between 2 K and 50 mK.

3. Electrical resistivity

Figure 1 shows the electrical resistivity ρ along the principal directions of the tetragonal cell (a, c) and along the diagonal of the tetragonal plane (c') from 350 K down to 50 mK on a semi-logarithmic scale. Each curve shows a remarkable increase of ρ with decreasing temperature down to approximately 11 K (9.5 K) for the directions a and c' (c). With further decreasing temperature ρ decreases slightly until another increase sets in, at about 2.5 K (1.9 K) for the a- and c'- (c-)direction. The c-direction curve shows additionally a small hump at about 150 K, which is more clearly seen in Fig. 2.

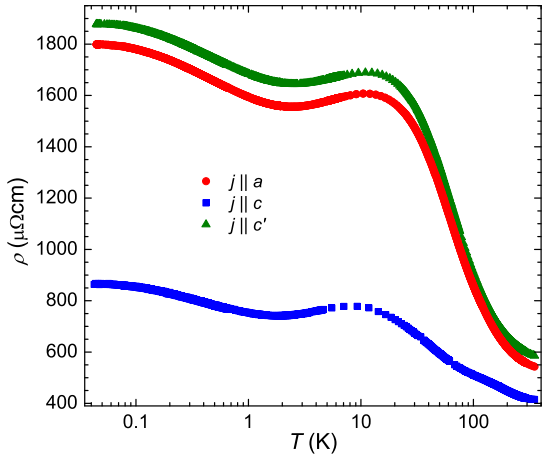

Figure 1. Temperature dependence of the electrical resistivity, $\rho(T)$, along the three directions a, c and c'. Pronounced anisotropy between the tetragonal plane directions a and c', and the c-direction is seen over the whole temperature range. The two different in-plane directions (a, c') show similar behaviour.

At first we try to model the behaviour of $\rho(T)$ by a two-gap structure in the DOS which is frequently found in simple semiconductors. In this picture, the high-temperature behaviour can be explained with an intrinsic gap. The second upturn at low temperatures is caused by a smaller extrinsic gap between impurity states and the intrinsic electron bands of the DOS. We fitted $\rho(T)$ at high temperatures with $\rho = \rho_1\ e^{(\Delta/2k_BT)}$ where ρ_1 is a constant, Δ is the energy gap, and k_B the Boltzmann constant. For each direction fits were made in two different temperature ranges. The resulting curves are shown in Fig. 2 and the fit parameters are listed in Tab. 1. A good description of the data is only possible in relatively narrow temperature ranges. A similar fit at low temperatures (between about 0.5 K and 2 K) yields gap values that are much smaller than the temperatures of the fitted range. Thus these fits are considered unphysical and are not shown.

An alternative explanation for the measured $\rho(T)$ curves is based on the interpretation of $CeRu_4Sn_6$ as Kondo insulator. In these materials a temperature-dependent hybridization gap is expected to develop below a characteristic temperature. In the absence of quantitative predictions for this behaviour fits to the data cannot be made. However, models for Kondo metals can be of some use. An important contribution to the temperature dependence of the resistivity comes from Kondo scattering. In the regime of incoherent scattering this results, for a metal, in the well-known -ln T behaviour. To analyse the data with this relation we have to first subtract the phonon contribution to the electrical resistivity, $\rho_{ph}(T)$, which we approximate by the electrical resistivity of the non-f reference compound $LaRu_4Sn_6$ [7]. The

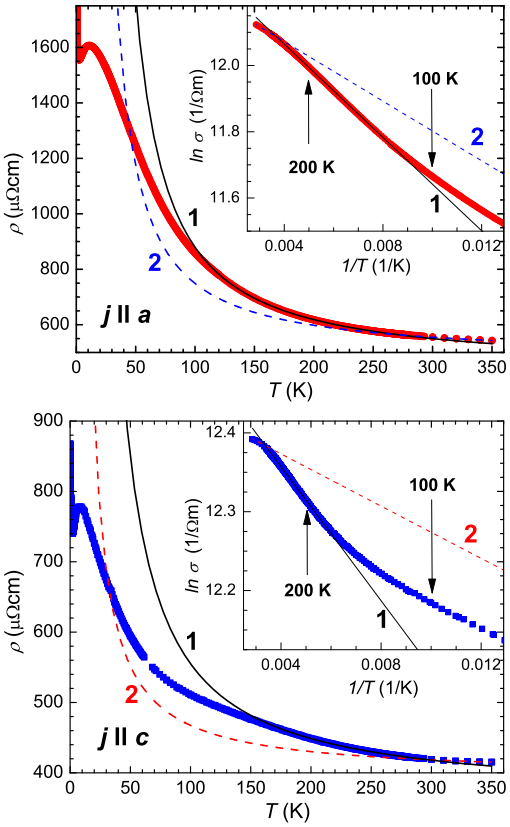

Figure 2. Electrical resitivity ρ versus temperature T with the electrical current j along the three investigated directions a, c and c'. The insets show an Arrhenius plot of the electrical conductivity σ. The lines represent fits to the data (see text).

Table 1. Parameters obtained by fitting $\rho = \rho_1\, e^{(\Delta/2k_BT)}$ to the $\rho(T)$ data for the indicated current directions and the indicated temperature ranges (see Fig. 2).

j along	fitnumber	fitted temperature range (K)	ρ_1 ($\mu\Omega$cm)	Δ/k_B (K)
a	1; 2	120 - 220; 300 - 350	434.7; 477.6	140.8; 89.9
c'	1; 2	120 - 220; 300 - 350	476.3; 519.7	132.6; 83.1
c	1; 2	190 - 270; 300 - 350	362.7; 395.9	85.0; 33.1

resulting $\Delta\rho = \rho - \rho_{ph}$ data are shown in Fig. 3. For each investigated direction three temperature regions can be identified where $\Delta\rho \sim$ -ln T is observed. Fits in these three ranges (1 - 3) are shown as straight lines in Fig. 3. A noteable observation is that the slopes of these straight lines decrease from range 1 to range 3 for the c-direction. For the directions within the tetragonal plane (a, c'), however, the slope in range 2 is larger than the one in range 1.

Some hints to understand this behaviour come from the Coqblin-Schrieffer model [8, 9]. While this is a single impurity model it has successfully explained some of the electrical resistivity features of Kondo lattice systems such as $CeAl_3$ or $CeAl_2$. One result of the Coqblin-Schrieffer model is that, for a constant DOS, different regions of -ln T behaviour can be resolved if the crystalline electric field-split multiplets of the magnetic impurity are energetically well enough separated. Since the prefactor of the -ln T-term decrease as the excited levels get thermally depopulated, the absolute value of the slope is expected to decrease, as qualitatively observed for the c-direction $\Delta\rho(T)$ of $CeRu_4Sn_6$. The three linear regions would then correspond to the Kondo effect in the full $J = 5/2$ multiplet (range 1), the two lowest-lying doublets (range 2),

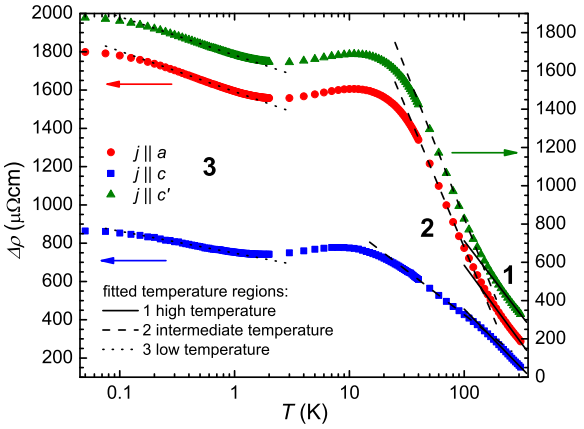

Figure 3. Magnetic contribution to the electrical resitivity $\Delta\rho$ versus temperature T, with the electrical current j along the three investigated directions a, c and c'. The lines indicate -ln T behaviour in three different temperature ranges (1 - 3).

and the ground-state doublet (range 3).

However, another effect should not be ignored. This is the change of the slope with the DOS at the Fermi level. At otherwise identical model parameters the absolute value of the slope of the -ln T-term decreases with decreasing DOS. Thus, the $\Delta\rho(T)$ behaviour for the c-direction could alternatively be interpreted as being due to incoherent Kondo scattering in a metal in which the DOS at the Fermi level decreases with temperature. The fact that within the tetragonal plane no such decrease of the absolute value of the slope is observed between range 1 and range 2 may then indicate that the gap opening is less relevant for the tetragonal plane, possibly due to the formation of nodes in the hybridization. Clearly, more realistic calculations are required to further sharpen our understanding of the electrical resistivity in this and other Kondo insulators.

4. Conclusion

Our electrical resistivity measurements on single crystalline $CeRu_4Sn_6$ revealed a remarkable anisotropy between the c-direction and directions within the tetragonal plane (a and c' directions) perpendicular to it. This confirms our previous observations from specific heat and magnetization measurements [6]. We have analysed the data qualitatively in terms of two different models, a simple semiconductor model with a two-gap structure and the Coqblin-Schrieffer model with temperature dependent electronic density of states. While these analyses will certainly be a guide for future experiments a more realistic theoretical description, possibly involving treatments with band structure calculations combined with dynamical mean field theory, is needed for a more complete understanding.

Acknowledgments

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