

MAGNETORESISTANCE OF Ce-BASED KONDO LATTICES: CeCu_2Si_2 AND CeAl_3

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The magnetoresistance $\Delta\rho(B)$ of CeCu_2Si_2 samples with different stoichiometry and of a CeAl_3 polycrystal has been measured at low temperatures and high magnetic fields. A change of sign in $\Delta\rho$ marks the formation of a coherent low-temperature state, which can be destroyed by a magnetic field or by disorder in the lattice.

Many properties of Kondo lattices displaying heavy-fermion behaviour [1] are very similar to those of dilute Kondo alloys like CuFe or CuCr [2], such as the susceptibility, the high-temperature resistivity and the electronic specific heat. Hence, an important question concerning the investigation of Kondo-lattices is, whether these systems behave merely as concentrated Kondo-alloys, or if the periodic arrangement of the scattering centers introduces new features characteristic for the lattice. Such a characteristic feature of the Kondo lattice not observed in dilute alloys is the low-temperature maximum in the electrical resistivity [3,4]. It is usually ascribed to the onset of coherent scattering. Besides this, Bredl et al [5], discovered a maximum in the specific heat coefficient γ near 0.4 K for both CeCu_2Si_2 and CeAl_3 . This result was interpreted as an indication for a coherent narrow 4f-band (quasiparticle band) at the Fermi level, that is formed in the Kondo lattice at low temperatures [6].

The transition between incoherent and coherent scattering is also reflected in the temperature dependence of the magnetoresistivity $\Delta\rho = \rho(B) - \rho(0)$. In dilute systems, $\Delta\rho$ is negative at all temperatures [7]. On the other hand, in CeAl_3 the negative $\Delta\rho$ that is observed above 1 K changes sign at very low temperatures [8,9]. This change of sign was explained by the formation of a 4f-derived conduction band. Similar behaviour of the magnetoresistance has later also been reported for CeCu_2Si_2 [10,11], but it was not clear,

whether the observed change of sign in $\Delta\rho$ was an intrinsic effect, or was caused by non-transformed Ce^{3+} ions ("magnetic impurities") [10].

Therefore, we have performed a systematic investigation of the magnetoresistivity $\Delta\rho$ of a number of CeCu_2Si_2 polycrystals with varying Cu-content, ranging from $\text{CeCu}_{1.8}\text{Si}_2$ to $\text{CeCu}_{2.2}\text{Si}_2$. In agreement with [12] the properties of the samples depend strongly on their Cu-content: the superconducting transition temperature varies smoothly from 250 mK ($\text{CeCu}_{1.8}\text{Si}_2$) to 740 mK ($\text{CeCu}_{2.2}\text{Si}_2$). The residual resistivity varies from $\rho_0 = 100 \mu\Omega \text{ cm}$ in $\text{CeCu}_{1.8}\text{Si}_2$ to $1.5 \mu\Omega \text{ cm}$ in an annealed sample of $\text{CeCu}_{2.2}\text{Si}_2$, which is to our knowledge the smallest ρ_0 -value ever measured in this compound. A single crystal which was grown from a melt with composition $\text{CeCu}_{2.2}\text{Si}_2$ was also studied ($T_c = 690 \text{ mK}$, $\rho_0 = 10 \mu\Omega \text{ cm}$). For comparison, a CeAl_3 polycrystal was studied as well.

The experiments were performed in a ^3He - ^4He dilution refrigerator in magnetic fields up to 8 T. The current through the sample was parallel to the field. AC-resistances were measured continuously, while the field was swept. Some samples were also studied up to fields of 35 T in a pulsed magnetic field at 1.5 and 4.2 K [14].

The results up to $B = 8 \text{ T}$ for four selected CeCu_2Si_2 -samples are displayed in figs 1 and 2. At 4.2 K, $\Delta\rho(B) = \rho(B) - \rho(0)$ is negative for all samples, in agreement with previous observations [10,11]. Typical values for $\Delta\rho/\rho(0)$ are 1–2% at $B = 8 \text{ T}$. The 4.2 K-results resemble those on

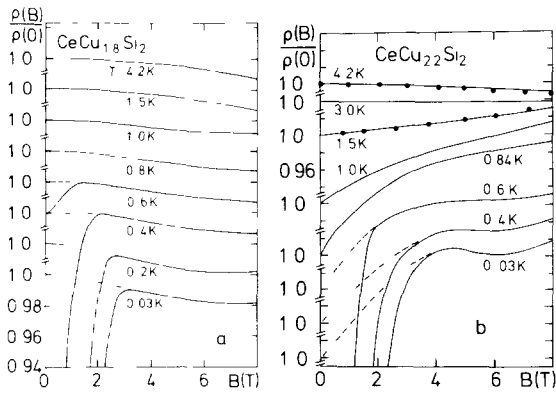


Fig 1 Resistivity in magnetic fields, $\rho(B)$, normalized to the zero-field value $\rho(0)$ for unannealed CeCu_{1.8}Si₂ (a) and CeCu_{2.2}Si₂ (b)-samples. Dashed lines are extrapolations to $B=0$, assuming that $\rho(T, B=0)$ follows a T^2 -law. Dots in (b) are results of high-field experiments on the same sample (see fig 3). Absolute values of $\rho(B=0)$ are $\rho_{4.2K} = 140 \mu\Omega \text{ cm}$, $\rho_0 = 100 \mu\Omega \text{ cm}$ in (a) and $\rho_{4.2K} = 80 \mu\Omega \text{ cm}$, $\rho_0 = 20 \mu\Omega \text{ cm}$ in (b).

dilute Kondo systems [7] and, hence, represent the regime where the Ce-ions behave as independent, incoherent Kondo ions. In the case of the unannealed CeCu_{1.8}Si₂-sample in fig 1a [15], this negative $\Delta\rho$ is found down to 30 mK, implying that no qualitative change in this incoherent scattering from the Kondo-ions in the Cu-deficit sample takes place.

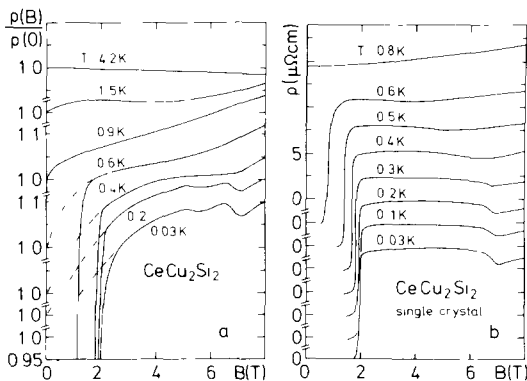


Fig 2 (a) $\rho(B)/\rho(0)$ for an annealed CeCu₂Si₂ sample. Dashed line: extrapolation to $B=0$, assuming that $\rho(T, B=0)$ follows a T^2 -dependence. $\rho_{4.2K} = 80 \mu\Omega \text{ cm}$, $\rho_0 = 12 \mu\Omega \text{ cm}$, (b) $\rho(B)$ for an unannealed CeCu₂Si₂ single crystal. The field was applied perpendicular to the tetragonal c -axis.

In all the other samples, the magnetoresistance changes sign and becomes positive between 4.2 and 1 K (figs 1b and 2). The temperature at which this happens increases with increasing T_c and decreasing ρ_0 , tracking the dependence of the position of the $\rho(T)$ -maximum at 10–25 K [3]. Apart from additional anomalies in $\Delta\rho$ around 6–7 T, to which we will return later, the $\rho(B)$ -dependence is especially strong at small fields, with a tendency to saturate at $B \approx 8$ T. Typical values for $\Delta\rho$ are a few $\mu\Omega \text{ cm}$ at $T \rightarrow 0$ and $B = 8$ T, corresponding to variations between a few % and 100%, depending on the residual resistivity. The fact that this positive $\Delta\rho$ can be destroyed by a Cu-deficit leads one to conclude that it is, indeed, a phenomenon that requires the periodicity of the lattice. We would, therefore, follow the explanation given in refs [8,9] for CeAl₃ that, at sufficiently low temperatures, a coherent 4f-band is formed which does then cause a usual metallic (positive) $\Delta\rho$.

Fig 3 shows the high-field results on the Cu-excess sample of fig 1b, and on a CeCu_{1.9}Si₂-sample. In agreement with the above results, $\Delta\rho$ of the Cu-deficit sample is negative at 1.5 and 4.2 K. For CeCu_{2.2}Si₂ on the other hand, $\Delta\rho$ at 1.5 K is

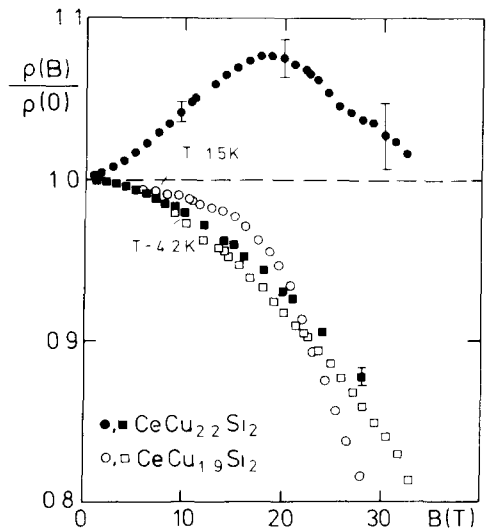


Fig 3 High-field results of $\rho(B)/\rho(0)$ for an unannealed CeCu_{1.9}Si₂ sample (open symbols) and for the CeCu_{2.2}Si₂ sample of fig 1b (full symbols).

positive, but passes through a maximum at $B = 20$ T. Thus, it appears that the low-temperature coherent state evidenced by the positive $\Delta\rho$ can be affected by a high magnetic field. Interestingly enough, the heavy-fermion compound UPt₃ shows a very similar behaviour [16,17], i.e. a change of sign in $\Delta\rho$ (near 14 K), and a maximum in the low- T magnetoresistivity near 20 T.

We will now return to the additional anomaly in the $\rho(B)$ -curves of high-quality CeCu₂Si₂ samples near 6–7 T. It shows up as a shallow dip in the results for the CeCu₂Si₂ polycrystal in fig. 1b, as a sharp minimum in the results of the single crystal in fig. 2b, and even as a double-structure in the case of the stoichiometric polycrystalline sample in fig. 2a. With increasing temperature, the feature broadens and shifts towards smaller magnetic fields, to become invisible at temperatures above 0.5 K. Since this characteristic temperature is close to that of the maximum in the specific heat coefficient $\gamma(T) = C(T)/T$ (0.4 K [5]) and of the change of sign in the thermopower $S(T)$ [18], we propose to explain the novel anomalies in $\rho(B)$ by the same type of reasoning employed to explain the $\gamma(T)$ and $S(T)$ anomalies, namely by assuming a coherence-derived pseudo-gap in the heavy-fermion density of states (DOS) near E_F , on an energy-scale $k_B \cdot 1$ K [5,13]. It is plausible to assume that, when the magnetic energy is comparable to the energy scale of the fine structure in the DOS, anomalies will show up in the resistivity on this magnetic field scale. Thus, the observed minimum in $\Delta\rho(B)$ is proposed to be another signature of coherence effects in the Kondo lattice CeCu₂Si₂. This conclusion is further supported by the fact that the $\Delta\rho(B)$ -minimum is most pronounced in high quality samples.

Because of the similarities that are found in the specific heats and thermopowers of CeAl₃ and CeCu₂Si₂ [18], we have also studied a CeAl₃-sample. Clearly, the results in fig. 4 show no anomaly like the one in fig. 2. In agreement with [8,9], $\Delta\rho$ is negative above 1 K. We do, however, not observe a monotonously positive $\Delta\rho$ at low-temperatures, but rather a positive peak near 2 T. Above $B \approx 3$ T, $\Delta\rho$ is negative at all temperatures. This is possibly related to the fact that we have measured the longitudinal magnetoresis-

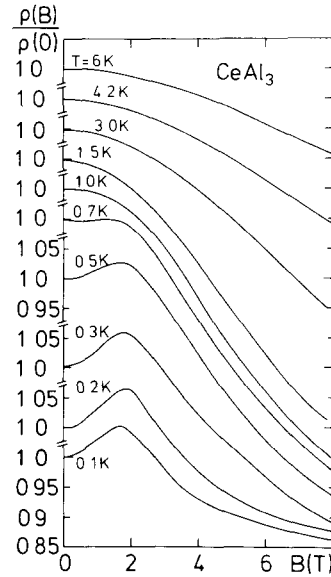


Fig. 4 $\rho(B)/\rho(0)$ for a CeAl₃ polycrystal, (sample no. 2 in ref [13]) $\rho_0 = 5 \mu\Omega \text{ cm}$

tance $I \parallel B$, while the results in ref [9] are obtained with $I \perp B$. At $T \geq 1.5$ K, a distinct anisotropy between the two configurations is reported in ref [9].

The peak in $\Delta\rho$ at 2 T has probably the same origin as the one found in CeCu₂Si₂ at 20 T, implying that the characteristic field scale in CeAl₃ is significantly smaller. This may be the reason, why nothing equivalent to the “7 T-anomaly” in CeCu₂Si₂ is found in CeAl₃.

To summarize, a change of sign of the magnetoresistance is observed in CeAl₃ and CeCu₂Si₂. In contrast to an earlier speculation [10], this feature is probably an intrinsic effect for both compounds and marks the formation of a coherent low-temperature state that can be destroyed by a sufficiently high magnetic field and by introducing disorder into the lattice. It is, therefore, another typical property of the Kondo lattice. An additional anomaly below 0.5 K in $\rho(B)$ of CeCu₂Si₂, which has no analogy in CeAl₃ may be considered another indication of a pseudo-gap structure in the heavy-fermion density of states that has been inferred from specific heat and thermopower data.

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References

- [1] G R Stewart, *Rev Mod Phys* 56 (1984) 755
- [2] B B Triplett and N E Phillips, *Phys Rev Lett* 27 (1971) 1001
- [3] C D Bredl, H Spille, U Rauchschalbe, W Lieke, F Steglich, G Cordier, W Assmus, M Herrmann and J Aarts, *J Magn Magn Mat* 31-34 (1983) 373
- [4] Y Onuki, Y Shimizu and T Komatsubara, *J Phys Soc Japan* 53 (1984) 1210. G R Stewart, Z Fisk and M S Wire, *Phys Rev B* 30 (1984) 482
- [5] C D Bredl, S Horn, F Steglich, B Luthi and R M Martin, *Phys Rev Lett* 52 (1984) 1982
- [6] N Grewe, *Solid State Commun* 50 (1984) 19
- [7] See e.g. K Samwer and K Winzer, *Z Phys B* 25 (1976) 269
- [8] A S Edelstein, R E Majewski and T H Blewitt, in *Valence Instabilities and Related Narrow Band Phenomena*, ed R D Parks (Plenum Press, New York, 1977) p 115
- [9] G Remeny, A Briggs, J Flouquet, O Laborde and F Lapiere, *J Magn Magn Mat* 31-34 (1983) 407
- [10] U Rauchschalbe, W Baus, S Horn, F Steglich, F R de Boer, J Aarts, W Assmus and M Herrmann, *J Magn Mat* 47&48 (1985) 33
- [11] Y Onuki, T Hirai, T Komatsubara, S Takayanagi, A Sumiyama, A Furukawa, Y Oda and H Nagano, *J Magn Magn Mat* 52 (1985) 338
- [12] H Spille, U Rauchschalbe and F Steglich, *Helv Phys Acta* 56 (1983) 165
- [13] C D Bredl, N Grewe, F Steglich and E Umlauf, *Proc LT17*, eds U Eckern, A Schmitt, W Weber and H Wuhl (Elsevier, Amsterdam, 1984) p 327
- [14] R Gersdorf, F R de Boer, J C Wolfrat, F A Muller and L W Roeland, in *High Field Magnetism*, ed M Date (North-Holland Amsterdam, 1983) p 277
- [15] The observed superconductivity with $T_c = 250$ mK in this sample is very probably not an intrinsic effect. The X-ray diagram of this unannealed sample showed spurious reflections of $CeSi_2$ -precipitations and, hence, regions of higher Cu-concentrations have to exist which may become superconducting and short-circuit the sample.
- [16] A de Visser, R Gersdorf, J J M Franse and A Menovsky, *J Magn Magn Mat* 54-57 (1986) 383
- [17] A de Visser, A Menovsky and J J M Franse, *J Magn Magn Mat* 63 & 64 (1987) 365
- [18] F Steglich, C D Bredl, W Lieke, U Rauchschalbe and G Sparn, *Physica* 126B (1984) 82