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Magnetoresistance of $CeCu_2Si_2$: Differences and similarities to UBe_{13}

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The magnetoresistance of nonsuperconducting $CeCu_{1.9}Si_2$ and superconducting $CeCu_{2.2}Si_2$ was measured for temperatures between 1.4 and 25 K and magnetic fields to 14 T. For temperatures larger than 5 K, both sets of data satisfy a scaling relation derived for the conventional single-channel Kondo model. According to the magnetoresistance, the single ion characteristic temperature T_0 for $CeCu_{1.9}Si_2$ is about 30% smaller than that for $CeCu_{2.2}Si_2$. The T_0 values are in good agreement with the ones obtained from the low-temperature magnetic susceptibility but are larger than the characteristic temperatures derived from the low-temperature specific heat. This disagreement is especially large for the superconducting sample (by a factor of 2). The magnetoresistance results point to a fundamental difference between the low-temperature states in $CeCu_2Si_2$ and UBe_{13} and possibly between other Ce- and U-based heavy fermions.

Measurements of heavy fermion materials in a broad range of magnetic fields can offer new insights into the origin and nature of the heavy fermion phenomenon. There are at least two important reasons for this. (1) The heavy fermion state (or states) occurs in a close proximity to magnetic states, thus magnetic probes should be the prime tools in investigating this state. (2) Some of the proposed theoretical models have definite predictions for the magnetic field response of various physical properties.

In this paper we report on the magnetoresistance for the first discovered heavy fermion superconductor, $CeCu_2Si_2$.[1] The magnetoresistance for this compound has been investigated in the past,[2] and has been often cited in support of the Kondo lattice interpretation of the normal state of this system. This interpretation was based on two following observations: the magnetoresistance is negative at temperatures of order 10 K and changes its sign to positive somewhere below 4 K, depending on the sample. There are numerical approximate solutions for the magnetoresistance in the Coqblin-Schrieffer model[3] and in the periodic Anderson model.[4] However, direct application of these solutions to the case of $CeCu_2Si_2$ is difficult and not reliable for small absolute values of its magnetoresistance. In one of our previous publications[5] we have demonstrated that a scaling analysis can provide fundamental information about the physical system. We have shown that the field scaling dimension for UBe_{13}, another heavy fermion superconductor which also has negative magnetoresistance, strongly disagrees with the value expected for a conventional Kondo system. Our measured value, about 0.6, is in fact close to the one predicted for the magnetic two-channel Kondo effect (0.5). Furthermore, based on the scaling analysis, we have postulated presence of ferromagnetic-type correlations in UBe_{13}.

Low-temperature thermodynamic and transport properties of $CeCu_2Si_2$ are astonishingly similar to those of UBe_{13}.[6] The low-temperature enhancements of the low-temperature specific heat in both compounds are almost identical; the temperature dependence of the electronic specific heat and electrical resistivity in zero field are alike. These and several other observations provided justification for treating Ce- and U-based heavy fermion materials as belonging to the same category and the same theoretical approaches are normally applied to these two classes of physical systems. However, there are experimental data suggesting that, in fact, there might be fundamental discrepancies between Ce- and U-based heavy fermions. In our previous publications we have pointed out some systematic differences in response to magnetic fields or chemical alloying between materials belonging to these two groups.[7] There is a growing awareness of these differences as witnessed by some recent publications.[8] One of our major goals is to find out whether these systematic differences between U- and Ce-based systems result from a somewhat different range of parameters describing their low-temperature states, like the ones considered in the Kondo-necklace model, or whether completely different physical phenomena cause an appearance of large effective electronic masses in these two groups of materials at low temperatures.

Physical properties of $CeCu_2Si_2$, including superconductivity, critically depend on sample preparation conditions and especially on the Cu stoichiometry. In order to investigate possible correlations between the superconductivity and the normal state magnetoresistance, we have used in our study a superconducting $CeCu_{2.2}Si_2$ sample and a nonsuperconducting $CeCu_{1.9}Si_2$. The polycrystalline samples were prepared by an arc melting technique using the highest purity available Ce, from Ames Laboratory. Both samples were annealed at 900 °C for 1 week. X-ray-diffraction analysis indicated single phase, $ThCr_2Si_2$-type, materials. Superconductivity in both samples has been checked by ac-magnetic susceptibility down to 0.3 K.

Figure 1 shows the zero field electrical resistance between 1.4 and 25 K. The resistance values have been normalized to the corresponding lowest measured temperature (about 1.4 K) values. The Cu-deficient sample has a maximum at about 5 K. Such a peak is expected for a Kondo lattice defined as a translationally invariant assembly of Kondo impurities. The resistance for the Cu-superstoichiometric sample has a much less pronounced maximum near 20 K. A huge difference between the temperature position of these maxima has never been properly accounted for. It seems to suggest a significantly lower characteristic (single ion) temperature for the

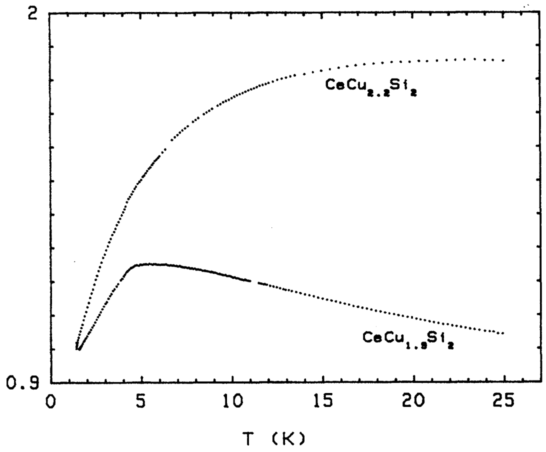

FIG. 1. Zero field resistance for $CeCu_{1.9}Si_2$ and $CeCu_{2.2}Si_2$.

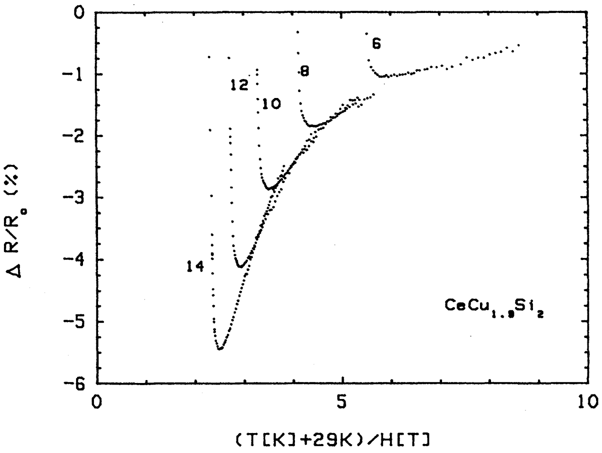

FIG. 3. $\Delta R/R_0$ versus $x=(T+T_0)/H$ for $CeCu_{1.9}Si_2$, where $T_0=29$ K; $\Delta R/R_0=(R(H)-R(H=0))/R(H=0)$.

$CeCu_{1.9}Si_2$ sample than for $CeCu_{2.2}Si_2$ or some additional disorder existing in the former sample.

The effect of magnetic fields, 10 and 14 T, on the resistance is illustrated in Fig. 2 for the Cu-deficient sample. The magnetoresistance is negative above 3.5 K and changes its sign below this temperature. The sign change of the magnetoresistance takes place at higher temperatures for lower fields. Qualitatively similar behavior was observed for $CeCu_{2.2}Si_2$, although the change of the sign of the magnetoresistance was systematically at lower temperatures, by about 1.5 K.

Further analysis of the magnetoresistance for the $CeCu_2Si_2$ samples is performed in the framework of the single ion Kondo model.[3] In this model the relative magnetoresistance $[=(R(H)-R(0))/R(0)]$ is only a function of $x=(T+T_0)/H$, where T_0 is a characteristic single impurity temperature. Therefore, magnetoresistance versus temperature data corresponding to different magnetic fields can be superimposed on a single curve by performing the $T\rightarrow x$ transformation.

Figure 3 shows six magnetoresistance curves for $CeCu_{1.9}Si_2$ corresponding to the fields of 6, 8, 10, 12, and 14 T plotted versus $x=(T+T_0)/H$, where $T_0=29$ K. For clarity, we do not include data for the 2 T field, also used in this study (the corresponding magnetoresistance was very small). All curves overlap for temperatures larger than 5 K. Interestingly, deviations from the behavior expected for the Kondo impurity system start at about 5 K independently on the value of the magnetic field. Similar scaling procedure performed for $CeCu_{2.2}Si_2$ (see Fig. 4) yields much larger T_0 of about 40 K. Again, clear deviations from the single impurity behavior begin at about 5 K for all applied fields.

This simple analysis leads to several conclusions. First of all, according to this analysis, the single ion characteristic temperature (T_0) for $CeCu_{2.2}Si_2$ is about 30% larger than that for $CeCu_{1.9}Si_2$. Although there is a significant uncertainty on our magnetoresistive T_0 (about 5 K) it cannot account for this large difference between the two samples. Other popular measures of the characteristic temperature, like those provided by the low-temperature specific heat, magnetic susceptibility, and magnetization are also usually consistent with

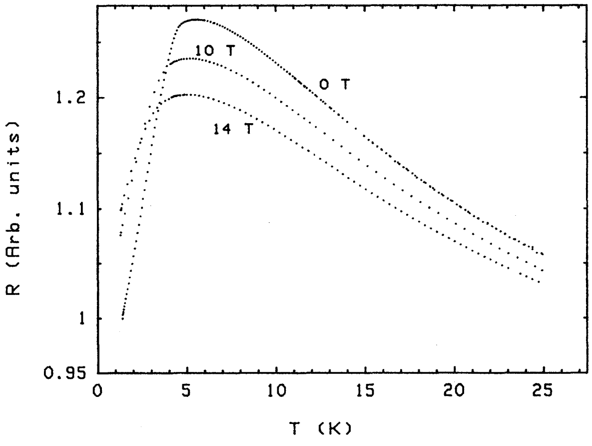

FIG. 2. Resistance versus temperature for $CeCu_{1.9}Si_2$ in $H=0$, 10, and 14 T.

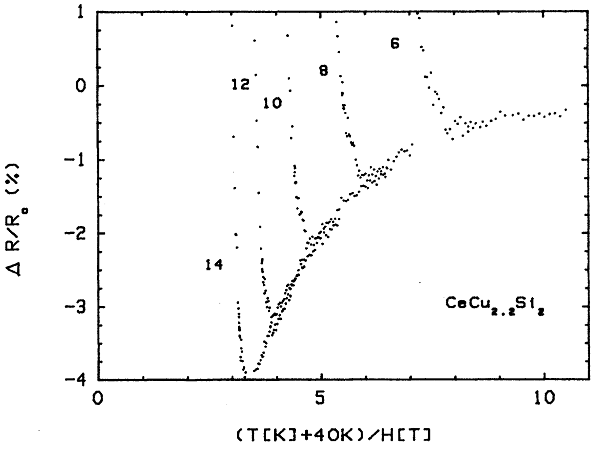

FIG. 4. $\Delta R/R_0$ versus $x=(T+T_0)/H$ for $CeCu_{2.2}Si_2$, where $T_0=40$ K.

the Cu-stoichiometric or superstoichiometric samples having a smaller characteristic temperature than the Cu-deficient samples. In our case, the 1.8 K susceptibility (the lowest temperature accessible by our magnetometer) for $CuCu_{2.2}Si_2$ was about 7.5 memu/Ce mol versus 12 memu/Ce mol for $CeCu_{1.9}Si_2$. In the single ion Kondo model T_0 and $\chi(0\ K)$ are related by the effective moment μ_{eff}, $T_0=\mu^2_{\mathrm{eff}}/3\chi(0)$. We employ this relation to calculate the effective moments using our magnetoresistive T_0 values and $\chi(1.8\ K)$; μ_{eff} is about $1.67\mu_B$ for $CeCu_{1.9}Si_2$ and $1.55\mu_B$ for $CeCu_{2.2}Si_2$. There are no reliable direct measurements of μ_{eff} for $T\rightarrow 0$ but we can compare our values with the hypothetical one, calculated for the lowest-lying crystal field level Γ_7 doublet. This hypothetical value, $1.65\mu_B$,[9] is in excellent agreement with our measured values. Otherwise, we conclude that the single ion characteristic temperatures derived from the magnetoresistance are in good agreement with the ones obtained from the low-temperature magnetic susceptibility. On the other hand, there is a substantial disagreement between T_0 and characteristic temperatures derived from the specific heat; $T_K=0.68R/\gamma$. Substituting C/T at 1.1 K for γ we arrive at $T_K=6.7$ K for $CeCu_{1.9}Si_2$ and 7.5 K for $CeCu_{2.2}Si_2$. T_0 and T_K are connected by the Wilson ratio R; $R=\pi^2k^2_B/\mu^2_{\mathrm{eff}}\chi(0)/\gamma$. For a spin-1/2 system, $R=2$ implying $T_0/T_K=2.42$. Thus, this disagreement, already noted[6] for the specific heat and susceptibility, is more than a factor of 2 for the superconducting sample and somewhat smaller for the Cu-deficient sample.

These results are consistent with several alloying studies[10] implying that only a fraction of the low-temperature specific heat (of order 50%) originates from the Kondo effect. The source of the remaining m^* enhancement is unknown. Some of the mechanisms considered include short range magnetic correlations, unusually strong electron-structural degrees of freedom coupling,[11] and Kondo lattice effects[12] which are not accounted for by the existing theories. It has been also argued[6] that a large difference between an energy scale derived from the thermodynamic measurements and a magnetic energy scale is characteristic to superconducting heavy fermion metals. Our results, i.e., a larger difference between these two energy scales for the superconducting sample than for a nonsuperconducting one, are also consistent with this assertion.

Interestingly, deviations from the single impurity behavior start at about the same temperature, 5 K, for both samples. Such deviations are expected below a characteristic Kondo lattice temperature, the coherence temperature, T_{coh}. Since T_0s for the two investigated samples are significantly different, our results suggest lack of any dependence between the coherence temperature and the single ion temperature. This last observation contradicts our understanding of a Kondo lattice which assumes T_{coh} to be proportional to T_0 ($T_{\mathrm{coh}}<T_0/N$, where N is an effective degeneracy of the single impurity).[12]

In summary, the magnetoresistance of $CeCu_2Si_2$ has a conventional Kondo character above 5 K. T_0s obtained from the magnetoresistance analysis are in agreement with the low-temperature magnetic susceptibility and significantly smaller than the characteristic temperatures derived from the specific heat. This disagreement is especially large for the superconducting sample and is somewhat smaller for the Cu-deficient sample.

We find a fundamental difference between the magnetoresistance of $CeCu_2Si_2$ and UBe_{13}. The $CeCu_2Si_2$ data are consistent with the magnetic field scaling dimension equal to 1 (conventional Kondo effect) while the magnetic field scaling dimension for UBe_{13} is close to 0.6, which argues for a non-Kondo origin of its magnetoresistance and m^*.

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