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Magnetism and superconductivity in CeCu_2Si_2 single crystals

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Single crystals of CeCu_2Si_2 were grown from metal solvents and their magnetic properties were systematically varied by partial substitution of Cu by Sn or In. The susceptibility χ is anisotropic and at high temperatures it is dominated by the crystal field split $4f$ state. At low temperatures the magnetic moments are drastically reduced. This reduction, as well as the Curie-Weiss temperature, depend sensitively upon the chemical composition. The observed variations both in the magnetic and the resistance behavior point to a Kondo-like coupling between local moments and conduction electrons. The strength of this interaction varies as the Fermi level is shifted by chemical means.

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INTRODUCTION

CeCu_2Si_2 is the first compound in which "heavy fermion" superconductivity was established.¹ The valence of Ce is close to 3^+ and the magnetic properties at temperatures higher than 10 K are determined by well defined local $4f$ moments. Below this temperature a nonmagnetic state develops and the electrons at the Fermi level acquire huge effective mass ($> 200 m_e$).^{2,3} These heavy fermions then condense into a superconducting state at $T_c \approx 0.6$ K. Because all the original work was done on polycrystalline samples and because it was obvious from the very beginning that the low temperature properties depend sensitively on details of the preparation process¹⁻⁶ we started a systematic study of single crystals. The aim was to gain insight into the formation of the heavy fermion liquid by a controlled variation of the chemical composition. We find that the degree of "demagnetization" in the low temperature range is sharply reduced when Sn or In atoms substitute for Cu. We suggest that this is consistent with the picture of the heavy fermions owing their origin to a Kondo-like interaction between the local $4f$ moments and the conduction electrons.

EXPERIMENTAL RESULTS

The crystals were grown from a Sn, In, or Cu solvent. When stoichiometric amounts of the CeCu_2Si_2 is added to In or Sn, the resulting crystals contain some In or Sn. The presence of In or Sn is confirmed by x-ray fluorescence. By increasing the Cu concentration it is then possible to reduce the In or Sn content. We find this is a useful method to vary the physical properties, as we will describe below. The higher the Cu concentration, however, the more difficult it is to extract the crystals. A SQUID magnetometer (S.H.E. Model 905) was used to measure the magnetic properties in fields up to 50 kOe and between 1.5 and 400 K.

TEMPERATURE DEPENDENCE OF χ

A general overview of the temperature dependence of the susceptibility χ is given in Fig. 1. The inverse of χ for two crystals, as measured in a field of 1 kOe, is plotted versus the

temperature. The two crystallographic directions are denoted as $\parallel a$ and $\perp a$, where a is in the basal plane of the tetragonal unit cell. This is also the plane in which the Ce atoms lie. The two samples represent two extreme cases and we call them "Cu rich" (= grown from Cu flux; round symbols) and "Cu poor" (grown from In; triangles). The anisotropy of χ is obvious and its sign and temperature dependence is consistent with the proposed CF level scheme.³ At temperatures higher than 250 K and $H \perp a$ data fall on a straight line with a slope that corresponds within 0.2% to the effective moment of a free trivalent Ce ion ($\mu_{\text{eff}} = 2.536 \mu_B$). For $H \parallel a$, the free ion value of μ_{eff} is not yet reached below 400 K. The main difference between the two samples is the vertical shift of the χ^{-1} curves by 60 ± 2 mol/emu. This holds true for $\chi \parallel a$ at $T > 20$ K and for $\chi \perp a$ at $T > 100$ K. The Curie-Weiss temperatures θ deduced from the $\chi \perp a$ data are -50 ± 1 K for the Cu rich sample and 0 ± 2 K for the Cu-poor sample. Additional differences between the two samples are given in Fig. 2. Whereas the Cu-rich one exhibits no

FIG. 1. Inverse susceptibility for two crystals of CeCu_2Si_2 along the two principal crystallographic directions. The filled circles correspond to a crystal grown from Cu solvent, the open triangles to a crystal grown from In solvent.

FIG. 2. Low temperature part of the susceptibility for the same two crystals as in Fig. 1 (broken lines: from Cu; full line: from In).

structure in $\chi(T)$, in quantitative agreement with the properly prepared polycrystalline material,¹⁻³ the Cu-poor one shows a cusp in $\chi(T)$ around 3.5 K.

FIELD DEPENDENT MAGNETIZATION

Figure 3 illustrates the result which appears to be most relevant in understanding the formation of the heavy fermion liquid. First it must be pointed out that the magnetization at 50 kOe is of order $0.1 \mu_B$ per Ce and is much smaller

FIG. 3. Magnetization curves at $T = 6$ K for three CeCu_2Si_2 crystals ($H \parallel a$). The chemical composition of the solvent determines the amount of moment reduction, as described in the text. The full and broken lines refer to the same samples as in Fig. 2.

than anticipated for the CF ground state³: three doublets at 0–140 K and 364 K, respectively. The local $4f$ moments are partially compensated. From a systematic variation of the flux composition we were able to establish a relation between the Cu content and the magnetization. Apparently an increase of the Cu concentration results in a stronger moment compensation.

DISCUSSION

Because the main purpose of our study is to shed some light on the formation of the heavy fermion which ultimately can condense into a superconducting state, we concentrate the discussion on the main trends in our data. These trends, we suggest, are due to a variation of the strength of the interaction between the $4f$ moments and the conduction electrons. In particular we would like to argue that the chemical variations affect the position of the Fermi energy with respect to the $4f$ level of Ce. When the crystals are grown from Sn or In the Cu concentration in the flux determines how many Sn or In atoms can occupy the Cu sites in the crystal. Since both Sn or In supply more electrons than Cu does, the Fermi level is higher in the “Cu poor” samples. The dominant parameter in the given situation is presumably the same as in the single Kondo impurity case, namely, the position of the $4f$ level with respect to E_F . Thus even small shifts of E_F will have pronounced consequences of the kind observed here: decrease of the moment compensation as the Fermi level rises. Equivalently, one can describe the low temperature state of CeCu_2Si_2 in terms of a Fermi liquid. In this language the replacement of Cu by In or Sn causes a decrease of the characteristic temperature T_F .

Even though this paper deals mainly with the magnetic properties we should mention some other physical properties which we have studied. The electrical resistivity is anisotropic and is largest in the basal plane ($I \parallel a$). More important, however, is the difference in the temperature dependence, as shown in Fig. 4. Both $\rho_{\parallel}$ and $\rho_{\perp}$ go through a broad maximum around 100 K, and $\rho_{\parallel}$ continues to rise upon further cooling to a maximum of at $T_{\parallel}^* = 11.5$ K, whereas $\rho_{\perp}$ reaches a much lower maximum at $T_{\perp}^* = 15$ –16 K. At even lower temperatures, the resistivity drops precipitously, presumably due to the formation of the coherent Kondo-lattice ground state. We would like to note that the difference between $T_{\parallel}^*$ and $T_{\perp}^*$ makes the interpretation of T^* in polycrystalline samples somewhat ambiguous, particularly if T^* is used as a quantity related to the Kondo temperature. For our single crystals $T_{\parallel}^*$ varied from ~ 8 K for the “Cu-poor” sample to ~ 14 K for the “Cu-rich” one. This trend is in line with the ideas discussed earlier. At present we are studying the superconducting properties and a more detailed account will be given elsewhere. We only note that ac susceptibility and resistance measurements indicate superconductivity in the “Cu-rich” sample at $T_c = 0.5$ K. In addition we also study the specific heat properties as we vary the degree of moment compensation. First measurements of the longitudinal magnetoresistance up to 80 kOe revealed a quite complex behavior. Particularly striking is the magnitude ($\Delta R / R \sim -40\%$ for $I \parallel c$, $\Delta R / R \sim 10\%$ for $I \parallel a$) and the very pro-

FIG. 4. Temperature dependence of the normalized resistance along the two principal crystallographic directions.

nounced nonlinearity at $T \sim 1.5$ K. The magnetoresistance appears to be determined by an interplay between CF effects and the suppression of Kondo-type interactions.

CONCLUSION

We were able to grow single crystals of CeCu_2Si_2 and to influence their magnetic properties in a controlled way through chemical means. The magnetic susceptibility is anisotropic and its temperature dependence is qualitatively consistent with the proposed CF-level scheme. The most prominent difference among the various single crystals is found in the value for the Curie-Weiss temperature and the

degree of moment compensation of lowest temperatures. We argue that these observations are indicative of a variation of the local moment-conduction electron coupling strength. Resistivity data are supporting this view. It is quite interesting to observe the maximum in $\chi(T)$ at $T = 3.5$ K in the "Cu-poor" sample (Fig. 2). The transition from a nonmagnetic to an antiferromagnetic-type ground state is predicted to occur in a Kondo lattice as the strength of the antiferromagnetic f - d coupling T_{fd} increases.^{7,8} A significant variation of T_{fd} has to be inferred from the magnetization data in Fig. 3, and thus the present series of CeCu_2Si_2 based compounds may represent an example for the nonmagnetic-to-magnetic transition in a Kondo lattice. The data presented here help to understand the origin of the heavy fermions in CeCu_2Si_2 , and the additional work, which is in progress, allows to draw quantitative conclusions.

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