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VARIATION OF THE HEAVY FERMION GROUND STATE IN $CeCu_2Si_2$ CRYSTALS BY STOICHIOMETRY CONTROL

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To gain insight into the formation of the heavy Fermion state in $CeCu_2Si_2$ we have studied the relationship between the high temperature and low temperature physical properties. The single crystals are grown from metallic solvents, such as Sn, In or Ga. The low temperature susceptibility χ can be varied by factors of 5, and samples with smallest χ have the largest linear contribution γ to the specific heat. This "anticorrelation" of χ and γ is unexpected. The reduction of χ at low T is mainly due to a temperature independent antiferromagnetic molecular-field type interaction $\chi^{-1}(T) = \chi_0^{-1}(T) + \lambda$, and not due to interactions which become dominant below a characteristic temperature. Superconductivity occurs in small χ – large γ samples.

1. Introduction

After the discovery of superconductivity in $CeCu_2Si_2$ [1], heavy Fermion superconductivity rapidly became one of the most interesting fields in solid state physics. The heavy Fermion state forms at low temperatures in a lattice of cerium or uranium ions that carry their full magnetic moments at high temperature [2,3]. The details of the heavy Fermion state are not quite clearly understood, and we found it helpful to study the relation between high and low temperature properties in single crystals of $CeCu_2Si_2$. Controlled chemical substitutions strongly influence the susceptibility and the specific heat. The observed "anticorrelation" between χ and γ is discussed.

2. Crystal growth

Single crystals were grown from metallic solvents, such as Cu, Sn, In and Ga. As the nutrient-solvent ratio is varied, the amount of solvent atoms incorporated in the $CeCu_2Si_2$ lattice could be influenced. From steric considerations, the solvent atoms we used are most likely to occupy Cu sites. The most extensive studies involved crystals grown from Sn, and we found it necessary to introduce excess Cu to compensate for the formation of parasitically forming Cu–Sn compounds. In turn, the unit cell volume, the c/a ratio as well as the physical properties could be systematically varied as a function of the Sn concentration in $Ce(Cu, Sn)_2Si_2$. A more complete account with numerical data will be given elsewhere [4].

3. Magnetic susceptibility at high and low temperatures

From the large number of crystals we studied, three are selected here to demonstrate the relation between the various physical propertis. For brevity, the crystal grown from Cu solvent is labeled "A", the one from Sn with large Cu excess in the melt "B", and the one from In "C".

The magnetic susceptibility χ is anisotropic [5,6] as expected from the proposed CEF level scheme [7]. The differences among the various crystals is most pronounced at low temperatures where the susceptibilities vary by factors of 4–5. The origin of these differences, however, is not rooted in low temperature interactions. This is illustrated in fig. 1 where an overview of $\chi(T)$ is given for crystal A. The CEF-only susceptibility $\chi_0(T)$ is also shown [8]. The shape of the $\chi_0(T)$ and $\chi(T)$ curves are obviously very similar, and the vertical shift suggests a description in terms of a molecular field: $\chi^{-1}(T) = \chi_0^{-1}(T) + \lambda$. The molecular field parameter λ is larger in the a-direction than in the c-direction. We

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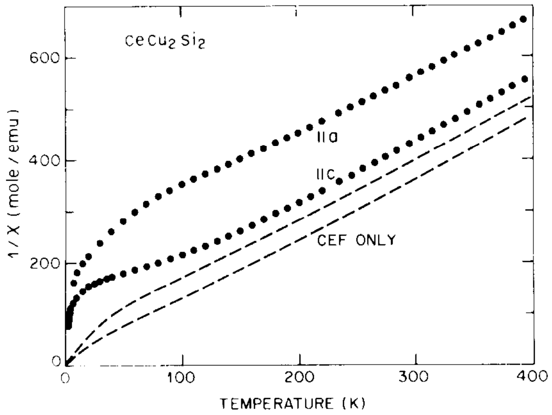

Fig. 1. Temperature dependence of the inverse susceptibility of a $CeCu_2Si_2$ crystal grown from Cu solvent. Also shown is the susceptibility according to the CEF level scheme of ref. [5].

note in passing that the layer of Ce-atoms is parallel to the a-direction. The effective moment at the highest temperatures is within experimental accuracy of the one of the free Ce^{3+} ion ($2.54\mu_B$).

Next we compare the three crystals in fig. 2. To simplify the graph, only χ_a is shown, and the striking difference among the crystals is the value of λ_a. In table 1 the molecular field parameters for the two crystallographic directions are compiled. It is interesting to note that $\lambda_a - \lambda_c$ is sample independent ((80 ± 2) mol/emu) despite the variation of λ_a by a factor of about two, and is larger than the CEF-only value of 35 mol/emu.

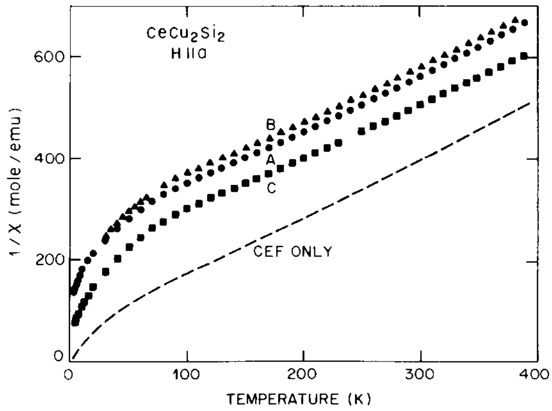

Fig. 2. Comparison of the susceptibility in the a-direction for three single crystals and the CEF level scheme. The chemical differences of the crystals are described in the text.

Table 1
Molecular field parameters λ describing the deviation of the measured susceptibilities from the CEF-only values. The two crystallographic directions are indicated by the subscripts a and c. The units are mol/emu. $\chi^{-1}(T) = \chi_0^{-1}(T) + \lambda$

Crystal	λ_a	λ_c	$\lambda_a - \lambda_c$
A	158	76	82
B	173	95	78
C	94	13	81
CEF only	–	–	35

At temperatures below 10 K, the susceptibility of A and B are very similar and small ($\chi_a = 6.2 \times 10^{-3}$ and 6.6×10^{-3} emu/mol, respectively at 5 K), whereas χ_a is twice as large for C. Both A and B exhibit only a very slight increase of χ on further cooling to 1.3 K, but χ of sample C goes through a maximum at ≈ 3.6 K. We never found it necessary to subtract an "impurity" contribution from the raw susceptibility data since there is no $1/T$ contribution present, and no saturation of magnetic impurities was found in fields up to 50 kOe. The slight increase of χ below 10 K therefore appears to be an intrinsic property of $CeCu_2Si_2$, and we will return to this point later.

4. Specific heat measurements

The specific heat c_p was measured from 0.3 K to 1.5 K for samples A and C, and to 12.5 K for sample B. The values of c_p at 1.5 K range between 635 mJ/mol K^2 for A to 890 mJ/mol K^2 for B. To emphasize the electronic contribution to c_p, it is customary to plot c_p/T vs. T. On such a plot, the temperature dependences are quite different for the three samples. For C the c_p/T value extrapolates to 400 mJ/mol K^2, whereas c_p/T increases on lowering T to ≈ 700 mJ/mol K^2 for A and to ≈ 1100 mJ/mol K^2 for B. The maximum of c_p/T for B at 0.8–0.9 K is noteworthy.

5. Discussion

The possibility to control the chemical composition of $CeCu_2Si_2$ single crystals offers an additional way to elucidate the origin of the heavy Fermion state. We find that an interaction of molecular-field type dominates the magnetic properties of $CeCu_2Si_2$ and the strength of this interaction is determining the low temperature susceptibility. The specific heat c_p/T is larger for the samples with small χ. It appears that χ and λ are "anticorrelated". The ratio χ/λ is ≈ 1 for A, ≈ 0.65

for B and 2.6 for C. To calculate this ratio, we used the values of $\frac{1}{3}(\chi_c + 2\chi_a)$ and c_p/T from an extrapolation $T \to 0$. It has been noted in the earliest work on $CeCu_2Si_2$ that for "good" samples the χ/λ ratio is < 1. We suggest this is due to the uncertainty at which temperature c_p/T should be taken. In the case of our sample B and also in a recent study [9] the value of c_p/T goes though a maximum below 1 K, the origin of which has been ascribed [9] to the fact that coherence is developing between the Ce sites.The new structure in the lowest temperature excitation spectrum makes any choice of a c_p/T value arbitrary. Therefore, the "enhancement" of c_p/T over χ in the "best" samples is not due to particular Fermi liquid parameters. Sample A shows diamagnetism in ac susceptibility measurements (0.5 K), but from the heat capacity data gapless superconductivity has to be inferred. It is noteworthy that $\chi(T)$ and c_p/T monotonously increase toward $T \to 0$ and the χ/λ ratio is close to 1. The occurrence of superconductivity is not necessarily tied to the formation of the coherent state below 1 K.

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