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To cite this version:
A. Germann, A. Nigam, J. Dutzi, A. Schröder, H. V. Löhneysen. MAGNETIC ORDERING IN THE HEAVY-FERMION ALLOYS CeCu6-xAux AND CeCu6-xAgx. Journal de Physique Colloques, 1988, 49 (C8), pp.C8-755-C8-756. <10.1051/jphyscol:19888340>. <jpa-00228517>

HAL Id: jpa-00228517
https://hal.archives-ouvertes.fr/jpa-00228517
Submitted on 1 Jan 1988

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MAGNETIC ORDERING IN THE HEAVY-FERMION ALLOYS CeCu$_{6-x}$Au$_x$ AND CeCu$_{6-x}$Ag$_x$

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Abstract. CeCu$_{6-x}$Au$_x$ and CeCu$_{6-x}$Ag$_x$ alloys order antiferromagnetically (e.g. $T_N = 0.95$ K for the Au alloy with $x = 0.5$) as seen in the specific heat, ac susceptibility, and electrical resistivity. The difference to CeCu$_6$, which shows neither magnetic order nor superconductivity, might be due to the negative lattice pressure of Au and Ag or to weakening of the Kondo-lattice ground state by breaking translational symmetry.

CeCu$_6$ is one of the few heavy-fermion metals that exhibits neither magnetic order nor superconductivity [1]. Upon replacing some of the Cu atoms by Au or Ag, a decrease of the Kondo temperature $T_K$ was inferred from susceptibility measurements above 1 K [2, 3]. In this paper, we report on specific-heat, ac susceptibility and resistivity measurements below 1 K which show that magnetic order occurs in CeCu$_{6-x}$Au$_x$ and CeCu$_{6-x}$Ag$_x$.

The samples were produced by arc melting under high-purity argon, they were remelted several times to ensure homogeneity and annealed at $\sim 700$ °C for two weeks in a high-vacuum furnace. X-ray diffraction showed that the samples were single-phase and retained the orthorhombic CeCu$_6$ structure.

The specific heat was measured in a dilution refrigerator between 0.05 K and 3 K and in a He$^4$ cryostat between 1.5 K and 10 K in magnetic fields up to 6 T. The standard heat-pulse method was employed. The ac susceptibility was measured with the mutual inductance method calibrated with a Foner magnetometer.

Figure 1 shows the specific heat of CeCu$_{5.5}$Au$_{0.5}$ (corresponding to a substitution of 8% of Cu by Au) above 1 K plotted as $C/T$ vs. $T^2$. The behaviour of the Au alloys is very similar to that of CeCu$_6$, which was also measured for comparison. Between 10 and 20 K, the specific heat can be described as $C = \gamma T + \beta T^3$, with $\gamma = 145$ mJ/mol K$^2$, $\beta = 1.32$ mJ/mol K$^4$ for $x = 0.5$ and $\gamma = 235$ mJ/mol K$^2$, $\beta = 1.14$ mJ/mol K$^4$ for CeCu$_6$ (cf. straight lines in Fig. 1). Here 1 mol refers to the formula unit. The latter values are in reasonable agreement with literature data [1, 4]. The slight change of $\beta$ corresponds to a change from $\theta_D = 229$ K (CeCu$_6$) to $\theta_D = 218$ K (CeCu$_{5.8}$Au$_{0.2}$) and can be attributed to the larger Au mass.

The rise of $C/T$ towards low $T$ is usually interpreted as the onset of "heavy-fermion" behaviour. It occurs at roughly the same temperature and is of the same magnitude for both samples. Thus the substitutional CeCu$_6$ alloys with Au replacing some Cu exhibit the same heavy-fermion behaviour as pure CeCu$_6$. In this context, a large change of $T_K$ [2, 3] is difficult to understand.

Figure 2 shows the specific heat of CeCu$_{5.5}$Au$_{0.5}$ in the low-$T$ region on a log-log plot in various applied magnetic fields. A rather sharp feature at $\sim 0.9$ K indi-
cates a phase transition. The inset of figure 2 shows the ac susceptibility $\chi$ which exhibits a maximum at the same temperature, thus suggesting a transition into an antiferromagnetic state at $T_N = 0.95$ K. (The rise of $\chi$ with decreasing $T$ observed below 0.5 K might be attributed to an impurity phase.) The interpretation in terms of an antiferromagnetic transition receives further support from the measurement of the electrical resistivity $\rho$ (not shown) which exhibits a sharp maximum at $T_N$: above $T_N$, $\rho$ increases with decreasing $T$ due to magnetic ordering, upon magnetic ordering the resistivity sharply decreases.

Figure 2 also displays the magnetic field dependence of $C$. The most important effect is that the maximum is shifted towards lower temperatures and becomes smeared out with increasing $B$. While the first feature is expected for an antiferromagnet, the second one could be due to an anisotropy of the $T_N(B)$ phase boundary, which is averaged out in our polycrystalline sample. Below $\sim 0.2$ K, the specific heat shows an upturn which increases with $B$. This contribution (upon subtraction of the extrapolated higher-temperature data) varies roughly as $C_n = b_n T^{-2}$.

Taking the value of $b_n$ expected for the Zeeman splitting of Cu$^{63}$ and Cu$^{65}$ nuclei ($b_n = 3.2 \times 10^{-6} B_{eff}^{\text{eff}}$ JK/mol Cu), we can describe the effective field reasonably well as $B_{eff} = B_0 + B$, where $B$ is the external field. However, the value $B_0 = 9.5$ T is unusually large for a transferred hyperfine field (originating from Ce) acting on the Cu sites. Of course, contributions from possible impurity phases cannot be ruled out. Clearly, this point deserves further study.

For completeness, we mention that we have found antiferromagnetic order also for a CeCu$_{5.8}$Au$_{0.2}$ sample ($T_N = 0.25$ K) and also upon replacing Cu by Ag, e.g. CeCu$_{5.6}$Ag$_{0.4}$ ($T_N = 0.55$ K), as evidenced by maxima in $C$ and $\chi$ at the temperatures $T_N$. The strong rise observed in $C/T$ for low $T$ in Ag substituted CeCu$_6$ [3] might hence be attributed to the onset of magnetic ordering, and is not necessarily due to a strong increase of $\gamma$ as compared to CeCu$_6$.

For the remainder of this article, we discuss possible reasons for the occurrence of magnetic order in CeCu$_6$ upon replacing Cu by iso electronic Ag or Au. The first possibility involves the negative lattice pressure exerted on the Ce ions by the large Au or Ag ions in comparison with the smaller Cu ions as evidenced by the increase in the lattice constant [2]. This could lead to a decrease of the hybridization between conduction-electron and 4f states and to a tendency towards magnetic ordering. On the other hand, breaking of translational symmetry could weaken the coherent Kondo ground state possibly again favouring magnetic order. An unambiguous check which mechanism is operative in establishing magnetic order, is the measurement of the pressure dependence of $T_N$ for our samples. If mainly the first mechanism is responsible, $T_N$ should decrease under pressure. Such measurements are presently under way.

Acknowledgments

We thank S. Ramakrishnan for the preparation of the samples and F. Stegelich for helpful discussions. One of us (A. K. N.) is indebted to the Minister für Wissenschaft and Kunst Baden-Württemberg for support during his stay at Karlsruhe.