dc and ac resistivity of amorphous $UCu_{4+x}Al_{8-x}$ thin films

P. Lunkenheimer, M. Kramer, R. Viana, C. Geibel, and A. Loidl
Institut für Festkörperphysik, Technische Hochschule Darmstadt, D-64289 Darmstadt, Germany

W. Suski
Polish Academy of Sciences, W. Trzebiatowski Institute of Low Temperature and Structure Research, P.O. Box 937, 50-950 Wroclaw 2, Poland

H. Ratajczak
Polish Academy of Sciences, Institute of Molecular Physics, 60-179 Pozna, Poland

I. Gociaska and T. Luciski
Institute of Physics, A. Mickiewicz University, 60-780 Pozna, Poland
(Received 10 September 1993; revised manuscript received 24 March 1994)

Thin films of the heavy-fermion system $UCu_{4+x}Al_{8-x}$ have been prepared and characterized by x-ray diffraction and by microprobe analysis. The films were found to be in an amorphous state and were subsequently investigated by ac-resistivity techniques for frequencies $20 \le \nu \le 10^9$ Hz and temperatures $0.1 \le T \le 300$ K. From these measurements it was concluded that compared to the results in the crystalline bulk material, the onset of magnetic order is suppressed at low Cu concentrations x, while the onset of a coherent heavy-fermion state is suppressed at high x. All samples investigated reveal a single-ion Kondo behavior down to the lowest temperatures. Significant deviations from the behavior of dipolar Kondo systems were detected. Up to the highest frequencies no indications of a narrow Drude peak could be found.

I. INTRODUCTION

Alloys in the ternary system $UCu_{4+x}Al_{8-x}$ reveal an alloying-induced transition from a magnetic ground state for low Cu concentrations ($x \le 1.25$) to a pure heavy-fermion (HF) behavior for $x \ge 1.5$.[1,2] Below $T_N = 37$ K, $UCu_{4.25}Al_{7.75}$ exhibits a simple collinear antiferromagnetic (AF) *I*-type magnetic structure, with the magnetic moments aligned along the c axis and alternating ferromagnetic order within the a b planes. At $T = 1.6$ K the ordered moment μ_s amounts to $1.6\,\mu_B$.[3] With increasing x, the magnetic ordering temperature T_N and the ordered moment at the uranium site are continuously diminished.[3] Finally, for $x = 1.25$, $T_N = 10$ K and the ordered moment is smaller than $0.23\mu_B$ as determined by elastic neutron scattering studies. In the heavy-fermion regime, for $x \ge 1.5$ the absence of formation of a fully coherent HF state was attributed to the Cu-Al disorder in this system.[1,2] From the analysis of the neutron results it has been concluded that for concentrations $0.25 < x < 1.5$ the excess Cu is substituted on the Al(j) sites only, while for $x = 1.9$ the Cu atoms are substituted randomly on the two inequivalent Al sites.[3] The aim of this paper is to examine how additional disorder, here especially introduced by the loss of long-range translational order on the U sites, influences the Kondo-lattice state and the magnetically ordered phase.

Here we report on the preparation and the characterization of amorphous thin films of $UCu_{4+x}Al_{8-x}$. The samples were characterized by x-ray investigations and microprobe analysis. In addition, measurements of the ac resistance and reactance at frequencies 20 Hz $\le \nu < 1$ GHz and for temperatures 100 mK $\le T \le 300$ K are presented. The low-frequency resistance of the three concentrations investigated shows a temperature dependence which is typical for single-ion Kondo systems. In most cases the temperature dependence of the resistivity reveals characteristic fingerprints if magnetic order is established. The absence of anomalies in $R(T)$ we interpret as the absence of long-range magnetic order in the thin films under consideration. We attribute this observation to the amorphous character of the samples, by which both the magnetic order and the formation of a coherent Fermi-liquid state are suppressed, leaving single-ion Kondo scattering as the dominant process down to the lowest temperatures.

II. EXPERIMENTAL DETAILS

The samples have been prepared by flash evaporation techniques in ultrahigh vacuum of 2×10^{-7} Pa on glass substrates. The thickness of the films was typically 100 nm. Here we report results for films of the compositions $UCu_4Al_{8.3}$, $UCu_{4.5}Al_{7.8}$, and $UCu_5Al_{7.3}$, which differ slightly from the stoichiometry of ideal bulk samples. The actual compositions of the films were determined using an energy-dispersive analysis with x rays. The structure of the samples was investigated utilizing surface-sensitive x-ray measurements in the Bragg-Brentano geometry. The scattered x-ray patterns showed no Bragg peaks but were characterized by a broad structureless intensity with a maximum at low angles characteristic of the amorphous state. Microprobe analysis was per-

0163-1829/94/50(13)/9581(4)/$06.00

formed in order to control the quality of the samples. It provided clear evidence that the films are highly homogeneous with no indications of phase separation.

The ac resistance and reactance of the films have been recorded using different experimental setups. At low frequencies ($\nu \leq 10$ MHz) autobalance bridges were used (HP4284 and HP4192 impedance analyzers). In addition, all samples were investigated at 113 Hz using a standard four-probe technique and lock-in amplifiers. These experiments were performed in home-manufactured variable-flow ^{4}He cryostats ($2 \leq T \leq 300$ K). For temperatures below 2 K the samples were attached to the mixing chamber of a ^{3}He-^{4}He dilution refrigerator. The high-frequency data (1 MHz $\leq \nu \leq$ 1 GHz) were measured using an HP4191 impedance analyzer connected to the sample in a ^{4}He cryostat by a coaxial line with Teflon as dielectric spacer material. Due to the temperature dependence of the dielectric properties of Teflon, calibration errors at low temperatures and high frequencies ($\nu > 100$ MHz) had to be taken into account.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the resistance R measured at 27.5 kHz for the three concentrations investigated. All data were normalized at 300 K. A reliable determination of the absolute values of the specific resistivity is hampered by a 50% uncertainty of the film thickness. Since no frequency dependence was detectable from 20 Hz up to 10 MHz these curves resemble the dc resistance of the samples as verified by additional lock-in measurements at 113 Hz. For all samples, a well-pronounced minimum in $R(T)$ shows up at temperatures below 100 K. For $T \rightarrow 0$ the resistance approaches a saturation value. At temperatures $T > 100$ K, R increases almost linearly with T.

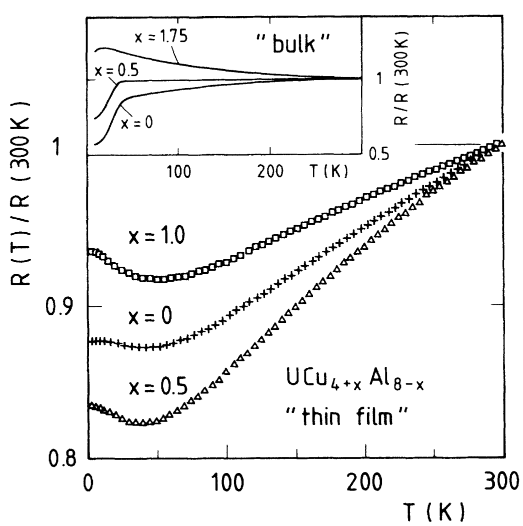

FIG. 1. Temperature dependence of the normalized resistivity in $UCu_{4+x}Al_{8-x}$ thin amorphous films for three concentrations, $x=0(+)$, $x=0.5(\Delta)$, and $x=1(\square)$. The inset shows the results of the normalized resistivity in polycrystalline bulk samples (Ref. 1) for comparison.

These observations are in clear disagreement with the results of resistivity measurements on bulk samples. Some typical results obtained on crystalline materials are shown in the inset of Fig. 1.[1] At low Cu contents ($x \leq 1.25$) a transition into an antiferromagnetic ground state leads to a drop in $R(T)$ (see the two lower curves in the inset of Fig. 1). Increasing x shifts the transition towards lower temperatures and finally leads to a suppression of the antiferromagnetic phase transition. For $x > 1.25$ a broad maximum in $R(T)$ develops, the position T_{max} of which shifts to higher temperatures with increasing Cu content (see, e.g., the upper curve in the inset of Fig. 1). The negative temperature coefficient of $R(T)$ for $T > T_{max}$ has been interpreted as being due to single-ion Kondo scattering.[1,2,4] At high Cu concentrations it becomes dominant and has its origin in an enhancement of the hybridization of the $5f$ electrons with the neighboring non-f-electron atoms.[3] The decrease of $R(T)$ for $T < T_{max}$ can be ascribed to the development of coherent scattering by the periodic U sublattice and indicates the formation of a heavy-fermion ground state. However, due to the Cu-Al disorder the $R(T)$ maximum is less pronounced than in other dense Kondo lattices.[1,2]

From the Cu to Al ratio of our samples, an antiferromagnetic ground state should be expected. However, the observed minimum in $R(T)$ indicates single-ion Kondo scattering with no sign either of a magnetic phase transition or of a coherent heavy-fermion state at low temperatures. To examine the behavior of $R(T)$ in more detail, we tried to fit the observed temperature dependence with the well-known Kondo behavior, i.e.,

$$R \sim \ln(1/T) \quad \text{for } T > T_K , \tag{1}$$

and

$$R = R_0(1-\alpha T^n) \quad \text{for } T < T_K . \tag{2}$$

For the dipolar Kondo effect n should be 2. In Fig. 2 typ-

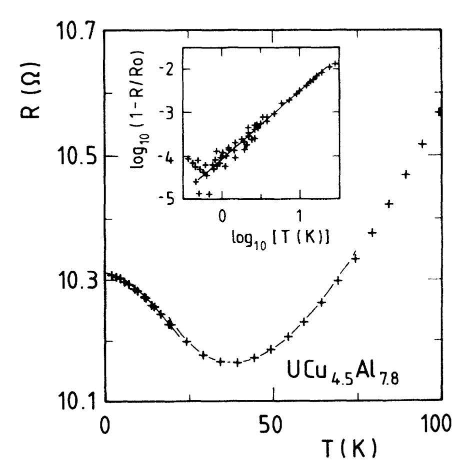

FIG. 2. Temperature dependence of the resistivity in $UCu_{4.5}Al_{7.8}$ ($x=0.5$). The solid lines are fits to the data as described in the text. The inset shows the power-law behavior $1-R/R_0 \sim T^n$ with $n=1.5$ (solid line).

ical results of these fits for $UCu_{4.5}Al_{7.8}$ are shown as solid lines. For higher temperatures a behavior $R \sim T^2$ has been assumed, which is typical for amorphous metals with a positive temperature coefficient of the resistance.[5] The fit gives a good description of the experimental data for temperatures down to 20 K including the minimum (solid line for $T > 20$ K in Fig. 2). The data for $T < 20$ K can be well reproduced using Eq. (2) which is demonstrated in the inset of Fig. 2. Here $1 - R/R_0$ is plotted double logarithmically vs T. The exponent $n \approx 1.5 \pm 0.1$ is smaller than expected for the conventional Kondo effect. A similar result was observed for $UCu_5Al_{7.3}$, with an exponent $n \approx 1.4 \pm 0.1$. However, single-ion behavior with $n \approx 2 \pm 0.1$, characteristic for the dipolar Kondo effect, has been found in $UCu_4Al_{8.3}$.

A reasonable explanation for the observed single-ion behavior and the absence of long-range magnetic order in our films is that the amorphous structure of our samples leads to a suppression of the magnetic transition. This can be easily understood considering the high sensitivity of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction to variations in the U-U distances. However, the occurrence of a spin-glass state as has been found in amorphous U-(Pt,Pd)-Si alloys[6] cannot be ruled out by our experiments. In the absence of a long-range-ordered magnetic ground state, Kondo scattering is the dominant process even at the relatively low Cu concentrations of the samples investigated. Due to the disorder of the uranium sublattice, coherent scattering does not occur and single-ion Kondo scattering remains observable down to the lowest temperatures.

The origin of the power-law behavior of $R(T \to 0\text{ K})$ with a significant decrease of the exponent n with increasing Cu concentration is less obvious. At first glance one would expect the behavior of a conventional dipolar Kondo system with $n = 2$. Deviations from this behavior with an exponent $n = 1$ have been proposed for the quadrupolar Kondo effect,[7] and, indeed, a power-law behavior with $n = 1.13$ has been found in $Y_{0.8}U_{0.2}Pd_3$ and has been interpreted in terms of a two-channel quadrupolar Kondo effect.[8] In the thin films under investigation, heat capacity and magnetic susceptibility measurements can hardly be performed and so far any conclusions concerning a "marginal-Fermi-liquid" behavior remain speculative.

Finally, we discuss the frequency dependence of the real part of the conductivity in these systems. In heavy-fermion materials the dynamical conductivity shows a narrow Drude peak at relatively low frequencies, which scales with the effective mass of the heavy quasiparticles. This behavior has been predicted theoretically[9,10] and was experimentally confirmed by Beyermann *et al.*[11] and Awashi *et al.*[12] Figure 3 shows the conductance $G(\nu)$ over eight decades of frequency as measured for $UCu_4Al_{8.3}$ at three characteristic temperatures. Similar results were obtained for the other samples. At all temperatures G is frequency independent up to frequencies of 10 MHz. However, above 10 MHz a strong decrease of $G(\nu)$ can be observed in Fig. 3. For the high effective masses which characterize the charge carriers in $UCu_{4+x}Al_{8-x}$ a drop in the conductivity (the Drude peak) is expected at relatively low frequencies, indeed. The decrease of the conductivity should scale with the mass renormalization of the heavy quasiparticles and should be visible only well below T_K. As can be seen in Fig. 3 a similar frequency dependence was, however, detected at 2 K and at 296 K.

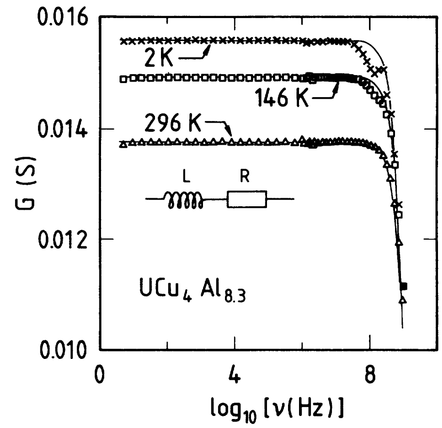

FIG. 3. Frequency dependence of the conductivity G in $UCu_4Al_{8.3}$ at three temperatures. The solid lines were calculated using the equivalent circuit indicated in the figure.

We interpret our high-frequency results using the simple equivalent circuit indicated in Fig. 3 which takes into account the inductance L of the sample. The lines in Fig. 3 are calculated using this equivalent circuit. At high temperatures a good fit of the data could be achieved. At low temperatures measurement errors due to the calibration problems mentioned above lead to minor deviations from the fits but the overall behavior can still be well described. The resulting inductance of ≈ 6 nH seems to be of reasonable magnitude for a metallic film. The absence of a Drude-like peak can be understood within the framework of the theory of Cox and Grewe.[10] They show that for HF systems revealing single-ion Kondo behavior, as is observed in our thin films, the Drude peak is almost completely smeared out.[10]

IV. CONCLUSIONS

In this paper we have presented dc- and ac-resistivity results on $UCu_{4+x}Al_{8-x}$ amorphous thin films and we compared these results to those obtained in bulk crystalline samples. In the thin films long-range magnetic order is suppressed and the measured resistivities closely follow a single-ion Kondo behavior. For $R(T \to 0\text{ K})$ significant deviations from a conventional dipolar Kondo effect were observed. The ac conductivity was measured up to GHz frequencies. The data provided no evidence for a low-frequency Drude-like peak. Our results are in accord with current theories of the conductivity in Kondo-impurity systems.[10]

ACKNOWLEDGMENTS

This work was partly supported by the Sonderforschungsbereich 252 (Darmstadt/Frankfurt/Mainz). One of us (R.V.) acknowledges support from CNPq (Conselho Nacional de Desenvolvimento Cientifico e Tecnologico do Brasil).

[1]C. Geibel *et al.*, Physica B **163**, 194 (1990).
[2]F. Steglich *et al.*, J. Magn. Magn. Mater. **90–91**, 383 (1990).
[3]A. Krimmel *et al.*, J. Magn. Magn. Mater. **73**, 103 (1992).
[4]R. Köhler *et al.*, Physica B **165–166**, 429 (1990).
[5]See, e.g., P. J. Cote and L. V. Meisel, in *Glassy Metals I,* edited by H. J. Güntherodt and H. Beck (Springer, Berlin, 1981).
[6]V. Wehrle *et al.*, Z. Phys. B **89**, 161 (1992).
[7]D. L. Cox, Phys. Rev. Lett. **59**, 1240 (1987).
[8]C. L. Seaman *et al.*, Phys. Rev. Lett. **67**, 2882 (1991).
[9]A. J. Millis and P. A. Lee, Phys. Rev. B **35**, 3394 (1987); R. Freytag and J. Keller, Z. Phys. B **80**, 241 (1990).
[10]D. L. Cox and N. Grewe, Z. Phys. B **71**, 321 (1988).
[11]W. P. Beyermann *et al.*, Phys. Rev. Lett. **60**, 216 (1988); J. Magn. Magn. Mater. **76–77**, 207 (1988).
[12]A. M. Awashti *et al.*, Phys. Rev. B **39**, 2377 (1989).