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Transport and thermodynamic properties of $CeCu_xAg_{1-x}Al_3$

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Abstract

Measurements of the specific heat, resistivity, and magnetoresistance of $CeCu_xAg_{1-x}Al_3$ classify this series of alloys as Kondo lattices with a magnetically ordered ground state. Though isoelectronic substitutions are performed, significant changes of the electronic density of states occur, causing a strong concentration-dependent variation of the antiferromagnetic ordering temperature. © 1999 Elsevier Science B.V. All rights reserved.

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Ce compounds with the $BaAl_4$ structure or its ordered variants attracted much interest because of superconductivity at ambient condition like in $CeCu_2Si_2$ [1] or at hydrostatic pressure like in $CePd_2Si_2$ or $CeNi_2Ge_2$ [2]. Previous investigations [3,4] on the $BaAl_4$-type compounds $CeCuAl_3$ and $CeCuGa_3$ revealed both Kondo interaction and antiferromagnetic order below $T_N = 2.8$ and 1.9 K, respectively, as well as strong crystal field splitting. An isoelectronic Cu/Ag substitution preserves the $BaAl_4$ structure in a wide concentration range but leads to an increase of the unit cell volume. The resulting negative chemical pressure is expected to stabilize the magnetic order due to a more local state of the cerium ion. In this paper we report about crystallographic, resistivity, magnetoresistance, and specific heat results of $CeCu_xAg_{1-x}Al_3$ which indicate that even isoelectronic substitutions cause large changes of the electronic structure.

A number of alloys of $CeCu_xAg_{1-x}Al_3$ were prepared by high-frequency melting. X-ray powder diffraction proved phase purity and revealed for both lattice parameters a and c, a continuous increase with decreasing x; the lattice parameter a ranges from 4.26 Å ($x = 1$) to 4.30 Å ($x = 0.35$), and c from 10.66 Å to 10.87 Å. For $x < 0.35$, diffractometry suggests a change of the crystal structure.

The electrical resistivity of this series is characterized by minima in $\rho(T)$ and a steep decrease below the respective ordering temperatures. A strong curvature in the paramagnetic state hints at crystal field effects.

Specific heat measurements of this series are presented in Fig. 1. λ-like anomalies trace the transition into the antiferromagnetic ground state. The transition temperatures initially decrease with rising Ag content, but beyond $x = 0.5$, T_N starts to rise again attaining eventually $T_N = 4$ K for $x = 0.1$. The jump of the heat capacity Δc_{mag} in the vicinity of T_N appears to be reduced with respect to an unperturbed crystal field ground state doublet ($\Delta c_{mag} \approx 12.5$ J/molK). We attribute this observation, in part, to the Kondo reduction of the ordered magnetic moments. The relatively broad peak featured for compounds with $x \geqslant 0.35$ indicates a less sharp phase transition due to short-range order effects or material inhomogeneities. An empirical ansatz, which has been discussed in Ref. [5–7], in the scope of the resonance level model of Schotte and Schotte [8], as well as the mean field theory, allows to calculate $c_{mag}(T)$ as a function of the Kondo temperature T_K and the exchange constant J. By fitting $c_{mag}(T)$ of the present alloys, reasonable values for J and T_K were obtained, e.g. $T_K = 8.0$ K and $J = 12.8$ K for $CeCuAl_3$. Values of T_K thus obtained are indicated in Fig. 3, revealing a slight increase with rising

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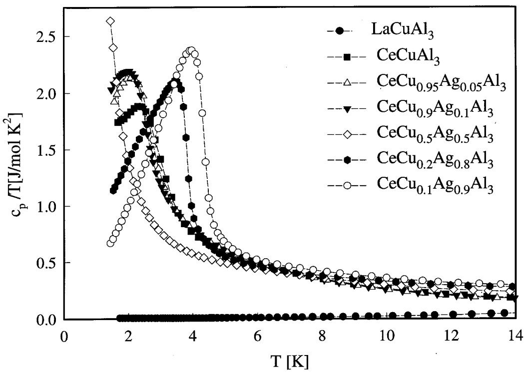

Fig. 1. Temperature-dependent specific heat c_p of various alloys of $CeCu_xAg_{1-x}Al_3$ plotted as $c_p(T)/T$ versus T.

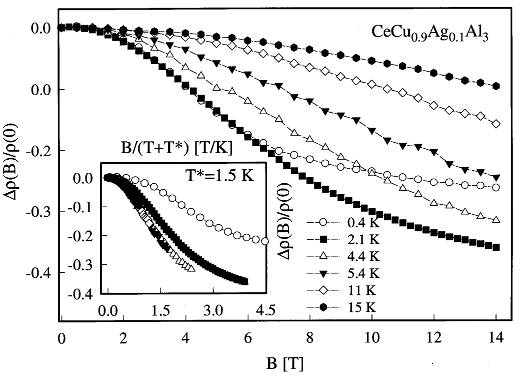

Fig. 2. Magnetoresistance $\Delta\rho/\rho$ of $CeCu_{0.9}Ag_{0.1}Al_3$ at various temperatures. The inset demonstrates scaling behaviour of $\Delta\rho/\rho$ for the paramagnetic temperature range.

values of x. The magnetic entropy observed from a comparison of the data with the appropriate La-based alloys reveals a lifting of the ground state degeneracy into three doublets with the first level roughly 30 K and the uppermost level about 100 K above the ground state doublet, in agreement with previous results [9].

Magnetoresistance measurements were performed in a temperature range down to 300 mK and in fields up to 14 T. Large negative magnetoresistance values were deduced for the whole series. As an example, $\Delta\rho/\rho$ is shown in Fig. 2 for $CeCu_{0.9}Ag_{0.1}Al_3$. In the paramagnetic temperature range the overall shape of $\Delta\rho/\rho(B)$ is characterized by a typical Kondo-like behaviour. The decrease of $\Delta\rho/\rho$ as the field rises is therefore a consequence of a field-induced suppression of the Kondo effect. Below the respective ordering temperatures, $\Delta\rho/\rho$ behaves different: initially it decreases strongly, but levels off at fields above 6 T. Usually, this behaviour is a typical sign of a magnetically ordered ground state in presence of the

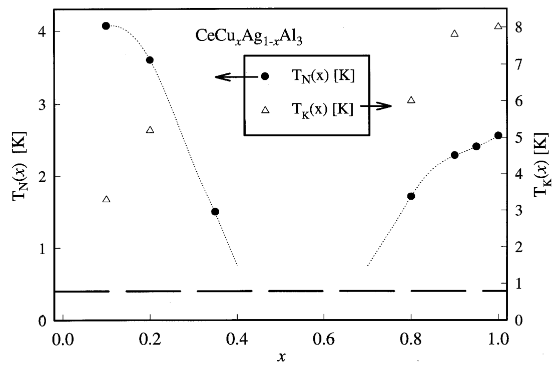

Fig. 3. Concentration dependence of the Néel temperature T_N (circles) and the Kondo temperature T_K (triangles) as derived from specific heat.

Kondo effect. In order to demonstrate that the physical behaviour is dominated by a single energy scale, i.e. $k_B T_K$, the field ramps were rescaled by a method proposed in Ref. [10]. The result for $CeCu_{0.9}Ag_{0.1}Al_3$ displayed as an inset in Fig. 2 shows that the curves in the paramagnetic regime coincide, whereas the curves within the ordered state deviate significantly. Fig. 3 shows a phase diagram of $CeCu_xAg_{1-x}Al_3$ deduced from specific heat and $\Delta\rho/\rho$ measurements. The circles give the Néel temperatures resulting from the specific heat measurements which are limited to 1.5 K. The dotted line serves as a guide for the eyes whereas the dashed line indicates that according to $\Delta\rho/\rho$ measurements all alloys adopt a magnetically ordered state above 0.5 K. The triangles are the respective Kondo temperatures T_K, derived also from specific heat data.

In conclusion, $CeCu_xAg_{1-x}Al_3$ represent a typical series of magnetically ordered Kondo alloys. The various properties observed match well with predictions of standard models of this subject.

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