Electrical and magnetic properties of RCu$_3$Al$_2$ (R = rare earth ions) compounds

J.W. Chen$^a$, B.K. Wang$^{a,b}$

$^a$ Department of Physics, National Taiwan University, Taipei, Taiwan, ROC
$^b$ Center for Measurements Standard, ITRI, Taiwan, ROC

Received 15 May 2006; received in revised form 1 September 2006; accepted 18 September 2006
Available online 30 January 2007

Abstract

We have investigated the crystal structure, electrical, and magnetic properties of the RCu$_3$Al$_2$ (R = rare earth ions) compounds by means of the powder X-ray diffraction, AC electrical resistivity $\rho$, and DC magnetic susceptibility $\chi$ measurements. Powder X-ray diffraction patterns reveal that all the samples studied are of the hexagonal CaCu$_5$-type structure with space group $P6_3/mmm$ and the lattice constants $a$ and $c$ are observed to decrease monotonically with increasing atomic number of R. The occurrence of a peak in the $\chi(T)$ curve and a drop off in the $\rho(T)$ curve at the corresponding temperatures indicate that antiferromagnetic transition occurs in TbCu$_3$Al$_2$ and DyCu$_3$Al$_2$ with transition temperature $T_N$ of 6 and 5 K, respectively. For SmCu$_3$Al$_2$ and GdCu$_3$Al$_2$, ferromagnetic orderings are observed with $T_c$ = 12 and 20 K, respectively. Except for SmCu$_3$Al$_2$, the $\chi(T)$ curves for all samples follow the Curie–Weiss behavior for $T > 100$ K, with the obtained values of $\mu_{\text{eff}}$ close to the moment of the corresponding R$^3+$ ions. The $\rho(T)$ curve for CeCu$_3$Al$_2$ reveals characteristics of a Kondo system which can be described by $\rho(T) = \rho_0 + \alpha T - \beta \ln T$, with $\rho_0 = 622.43 \mu\Omega\text{cm}$, $\alpha = 0.2783 \mu\Omega\text{cm/K}$ and $\beta = 71.92 \mu\Omega\text{cm/K}$.

© 2007 Elsevier B.V. All rights reserved.

PACS: 71.27.+a; 71.20.LP; 75.25.Ha

Keywords: Rare earth alloys and compounds; Kondo effect; Magnetic properties; Electrical properties

1. Introduction

Among the different types of the intermetallic system AB$_5$ (A = alkaline earth, transition metal, and lanthanides; B = d- and/or p-element), compounds that adopt the hexagonal CaCu$_5$-type and its derived structures have been intensively studied for decades because of their interesting properties [1,2]. Previous studies reveal that RCu$_5$ (R = rare earth) compounds can crystallize in the hexagonal CaCu$_5$-type structure or in the cubic AuBe$_5$-type structure, or both structures depending on the size of the rare earth R [3–5]. It was found that the Al-substituted system RCu$_4$Al will adopt the CaCu$_5$-type structure only [1,6]. Recently, a crossover from the magnetic Kondo compound CeCu$_5$ to the heavy fermion compound in the Al-substituted systems CeCu$_4$Al and CeCu$_3$Al$_2$ has been reported [7,8]. In addition, superconductivity was observed in LaCu$_3$Al$_2$ with the superconducting transition temperature $T_c = 1.7$ K [9]. In this report, we present our results of the crystal structure, electrical, and magnetic properties of the RCu$_3$Al$_2$ (R = rare earth ions) compounds.

2. Experimental details

The polycrystalline samples of RCu$_3$Al$_2$ (R = rare earth ions) were prepared by arc-melting the high purity elements (R: 99.99%, Cu: 99.999%, Al: 99.995%) together in a water cooled copper hearth in a Zr-gettered argon atmosphere. The alloy buttons were flipped over and re-melted carefully several times. To improve the sample homogeneity, the as melted samples were subsequently wrapped in Ta foils, sealed in the evacuated quartz tubes and annealed at 700 °C for 1 week. The crystallographic data for all samples were obtained with powder X-ray diffractometer utilizing Cu K$_\alpha$ radiation. AC electrical resistivity of the bar-shaped samples has been measured between 4.2 and 300 K in a He$^4$ cryostat using a four probe technique. DC magnetic susceptibility measurements were performed in a commercial superconducting quantum interference device (SQUID) from 2 to 300 K in an applied magnetic of 5000 Oe.

3. Results and discussion

Powder X-ray diffraction patterns reveal that all the samples studied are of the hexagonal CaCu$_5$-type structure.
increasing atomic number $R$, both values of the $RCu_3Al_2$ are plotted with respect to the rare earth $R$. With $R=La$, $Ce$, $Gd$, and $Tb$, respectively, indicating the occurrence of magnetic ordering in these compounds (see inset). The $R(T)/R(300 \text{ K})$ curve for CeCu$_3$Al$_2$ reveals different characteristics and increases with decreasing $T$ below room temperature. In fact, the $\rho(T)$ curve for CeCu$_3$Al$_2$ reveals characteristics of a Kondo system which can be fitted using

$$\rho(T) = \rho_0 + \rho_{ph} + \rho_K = \rho_0 + \alpha T - \beta \ln T,$$

where $\rho_0 = 622.43 \mu\Omega\text{cm}$ is the residue resistivity, $\alpha = 0.2783 \mu\Omega\text{cm/K}$ the coefficient of the phonon contribution and $\beta = 71.92 \mu\Omega\text{cm/K}$ is the coefficient of $\ln T$ of the Kondo scattering part $\rho_K$.

**Fig. 3(a) shows** the inverse susceptibility $\chi^{-1}$ as a function of temperature $T$ for RCu$_3$Al$_2$ ($R=\text{Ce, Nd, Gd, and Tb}$) measured in an applied field of 5 kOe for $0 \leq T \leq 300 \text{ K}$. The magnetic susceptibility data reveal that both CeCu$_3$Al$_2$ and NdCu$_3$Al$_2$ are nonmagnetic down to 2 K. For $T > 50 \text{ K}$, $\chi$ follows a Curie–Weiss behavior $\chi = C/(T - \theta_p)$, with $\theta_p = -39.6$ and $-11.7 \text{ K}$ for CeCu$_3$Al$_2$ and NdCu$_3$Al$_2$, respectively. The effective moment obtained from the Curie constant $C = N\mu^2_{eff}/3k_B$ yield the values of $\mu_{eff} = 2.49 \mu_B/\text{Ce ion}$ and $\mu_{eff} = 3.55 \mu_B/\text{Nd ion}$. The obtained values are close to that of the Ce$^{3+}$ ($\mu_{eff} = 2.54 \mu_B$) and Nd$^{3+}$ ($\mu_{eff} = 3.62 \mu_B$) ions.

Shown in **Fig. 3(b)** are the $\chi(T)$ curves for SmCu$_3$Al$_2$, GdCu$_3$Al$_2$, and TbCu$_3$Al$_2$ below 50 K. The $\chi(T)$ curve for GdCu$_3$Al$_2$ reveals the occurrence of a ferromagnetic transition at 20 K, near the temperature at which a change of the slope in the $R(T)/R(300 \text{ K})$ curve was observed for this compound. The magnetic susceptibility curve for TbCu$_3$Al$_2$ exhibits a peak at $T = 6 \text{ K}$, which indicates the occurrence of antiferromagnetic ordering in this compound. The $\chi(T)$ curves for both

![Fig. 1. (a) X-ray diffraction pattern of LaCu$_3$Al$_2$ with the Miller indices of the peaks labeled. (b) Lattice parameters $a$ and $c$, and the ratio $c/a$ for RCu$_3$Al$_2$ compounds.](image-url)

This is shown in **Fig. 1(a)**, where the X-ray diffraction pattern and the Miller indices of the diffraction peaks for LaCu$_3$Al$_2$ are plotted. The obtained values of the lattice parameters are $a = 0.52309(9) \text{ nm}$ and $c = 0.41862(5) \text{ nm}$ for LaCu$_3$Al$_2$. In **Fig. 1(b)**, the lattice parameters $a$ and $c$, and the ratio of $c/a$ for the RCu$_3$Al$_2$ are plotted with respect to the rare earth $R$. With increasing atomic number $R$, both values of $a$ and $c$ are found to decrease while the ratio of $c/a$ is found to increase instead. The lanthanum contraction (monotonic decrease of $a$ and $c$) observed here indicates that the rare earth ions in the RCu$_3$Al$_2$ compounds are trivalent at room temperature.

There are two inequivalent sites for Cu atoms in the CaCu$_5$-type structure, the basal plane 2c site and the mid-plane 3g site [3]. Roughly speaking, the size of the atoms on 2c and 3g sites are directly related to the length of $a$ and $c$, respectively. The behavior of $c/a$ of the RCu$_3$Al$_2$ system indicates that the Al atoms substitute preferential the Cu atoms on the 3g site in the large-size rare earth compounds but randomly distributed over both 2c and 3g sites in the smaller-size rare earth compounds.

The normalized electrical resistance $R(T)/R(300 \text{ K})$ versus $T$ curves for the RCu$_3$Al$_2$ ($R=\text{La, Ce, Gd, and Tb}$) compounds are plotted in **Fig. 2** for $0 \leq T \leq 300 \text{ K}$. The $R(T)/R(300 \text{ K})$ curves for $R=\text{La, Gd, and Tb}$ exhibit typical characteristics of a common metal and decreases monotonically with decreasing temperature $T$. In addition, a change of the slope in the $R(T)/R(300 \text{ K})$ curve is observed at $\sim 5$ and $19 \text{ K}$ for TbCu$_3$Al$_2$ and GdCu$_3$Al$_2$, respectively, indicating the occurrence of magnetic ordering in these compounds. The inset depicts the $R(T)/R(300 \text{ K})$ curves for GdCu$_3$Al$_2$ and TbCu$_3$Al$_2$ for $T < 25 \text{ K}$. 

![Fig. 2. $R(T)/R(300 \text{ K})$ vs. $T$ curves for RCu$_3$Al$_2$ ($R=\text{La, Ce, Gd, and Tb}$). The inset depicts the $R(T)/R(300 \text{ K})$ curves for GdCu$_3$Al$_2$ and TbCu$_3$Al$_2$ for $T < 25 \text{ K}$](image-url)
compounds can also be described with the Curie–Weiss law for $T > 100 \text{K}$ and the obtained values of $\mu_{\text{eff}}$ close to the moment of the corresponding $\text{R}^{3+}$ ions.

In summary, we have studied the crystal structure, electrical, and magnetic properties of the $\text{RCu}_3\text{Al}_2$ ($\text{R} =$ rare earth ions) compounds. We found that all the samples studied are of the hexagonal $\text{CaCu}_5$-type structure with the lattice constants $a$ and $c$ decreasing monotonically with increasing atomic number of $\text{R}$. $\text{CeCu}_3\text{Al}_2$ is a Kondo system as inferred from the $\rho(T)$ data. Both $\chi(T)$ and $\rho(T)$ data indicate that antiferromagnetic transition occurs in $\text{TbCu}_3\text{Al}_2$ and $\text{DyCu}_3\text{Al}_2$ with transition temperature $T_N$ of 6 and 5 K, respectively. For $\text{SmCu}_3\text{Al}_2$ and $\text{GdCu}_3\text{Al}_2$, ferromagnetic ordering are observed with $T_c = 12$ and 20 K, respectively.

Acknowledgement

This work was supported by the ROC National Science Council under Grant No. NSC 94-2112-M-002-013.

References