Electrical and magnetic properties of RCu_3Al_2 (R = rare earth ions) compounds

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Abstract

We have investigated the crystal structure, electrical, and magnetic properties of the RCu_3Al_2 (R = rare earth ions) compounds by means of the powder X-ray diffraction, AC electrical resistivity ρ , and DC magnetic susceptibility χ measurements. Powder X-ray diffraction patterns reveal that all the samples studied are of the hexagonal CaCu_5 -type structure with space group $P6/mmm$ 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_3Al_2 and DyCu_3Al_2 with transition temperature T_N of 6 and 5 K, respectively. For SmCu_3Al_2 and GdCu_3Al_2 , ferromagnetic orderings are observed with $T_c = 12$ and 20 K, respectively. Except for SmCu_3Al_2 , the $\chi(T)$ curves for all samples follow the Curie–Weiss behavior for $T > 100$ K, with the obtained values of μ_{eff} close to the moment of the corresponding R^{3+} ions. The $\rho(T)$ curve for CeCu_3Al_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 $\beta = 71.92 \mu\Omega\text{cm/K}$.

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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_4Al 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_4Al and CeCu_3Al_2 has been reported [7,8]. In addition, superconductivity was observed in LaCu_3Al_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_3Al_2 (R = rare earth ions) compounds.

2. Experimental details

The polycrystalline samples of RCu_3Al_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 $\text{Cu K}\alpha$ 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.

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Fig. 1. (a) X-ray diffraction pattern of LaCu_3Al_2 with the Miller indices of the peaks labeled. (b) Lattice parameters a and c , and the ratio c/a for RCu_3Al_2 compounds.

This is shown in Fig. 1(a), where the X-ray diffraction pattern and the Miller indices of the diffraction peaks for LaCu_3Al_2 are plotted. The obtained values of the lattice parameters are $a = 0.52309(9)$ nm and $c = 0.41862(5)$ nm for LaCu_3Al_2 . In Fig. 1(b), the lattice parameters a and c , and the ratio of c/a for the RCu_3Al_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_3Al_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_3Al_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_3Al_2 ($R = \text{La, Ce, Gd, and Tb}$) compounds are plotted in Fig. 2 for $0\text{ K} \leq T \leq 300\text{ K}$. 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 ~ 5 and 19 K for TbCu_3Al_2 and GdCu_3Al_2 , respectively, indicating the occur-

rence of magnetic ordering in these compounds (see inset). The $R(T)/R(300\text{ K})$ curve for CeCu_3Al_2 reveals different characteristics and increases with decreasing T below room temperature. In fact, the $\rho(T)$ curve for CeCu_3Al_2 reveals characteristics of a Kondo system which can be fitted using

$$\rho(T) = \rho_0 + \rho_{\text{ph}} + \rho_{\text{K}} = \rho_0 + \alpha T - \beta \ln T, \quad (1)$$

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 ρ_{K} .

Fig. 3(a) shows the inverse susceptibility χ^{-1} as a function of temperature T for RCu_3Al_2 ($R = \text{Ce, Nd, Gd, and Tb}$) measured in an applied field of 5 kOe for $0\text{ K} \leq T \leq 300\text{ K}$. The magnetic susceptibility data reveal that both CeCu_3Al_2 and NdCu_3Al_2 are nonmagnetic down to 2 K. For $T > 50\text{ K}$, χ follows a Curie–Weiss behavior $\chi = C/(T - \Theta_{\text{p}})$, with $\Theta_{\text{p}} = -39.6$ and -11.7 K for CeCu_3Al_2 and NdCu_3Al_2 , respectively. The effective moment obtained from the Curie constant $C = N\mu_{\text{eff}}^2/3k_{\text{B}}$ yield the values of $\mu_{\text{eff}} = 2.49\ \mu_{\text{B}}/\text{Ce ion}$ and $\mu_{\text{eff}} = 3.55\ \mu_{\text{B}}/\text{Nd ion}$. The obtained values are close to that of the Ce^{3+} ($\mu_{\text{eff}} = 2.54\ \mu_{\text{B}}$) and Nd^{3+} ($\mu_{\text{eff}} = 3.62\ \mu_{\text{B}}$) ions.

Shown in Fig. 3(b) are the $\chi(T)$ curves for SmCu_3Al_2 , GdCu_3Al_2 and TbCu_3Al_2 below 50 K. The $\chi(T)$ curve for GdCu_3Al_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_3Al_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. 2. $R(T)/R(300\text{ K})$ vs. T curves for RCu_3Al_2 ($R = \text{La, Ce, Gd, and Tb}$). The inset depicts the $R(T)/R(300\text{ K})$ curves for GdCu_3Al_2 and TbCu_3Al_2 for $T < 25\text{ K}$.

Fig. 3. Inverse susceptibility χ^{-1} as a function of temperature T for RCu_3Al_2 (R = Ce, Nd, Gd, and Tb). The inset are the $\chi(T)$ curves of SmCu_3Al_2 , GdCu_3Al_2 and TbCu_3Al_2 for $T < 50$ K.

compounds can also be described with the Curie–Weiss law for $T > 100$ K and the obtained values of the effective moment are $\mu_{\text{eff}} = 7.99 \mu_{\text{B}}/\text{Gd ion}$ and $\mu_{\text{eff}} = 9.74 \mu_{\text{B}}/\text{Tb ion}$, respectively. For SmCu_3Al_2 , ferromagnetic transition was observed at ~ 12 K.

Except for SmCu_3Al_2 , the $\chi(T)$ curves for the other RCu_3Al_2 samples are also found to follow the Curie–Weiss behavior for $T > 100$ K, with the obtained values of μ_{eff} close to the moment of the corresponding R^{3+} ions.

In summary, we have studied the crystal structure, electrical, and magnetic properties of the RCu_3Al_2 (R = rare earth ions) compounds. We found that all the samples studied are of the hexagonal CaCu_5 -type structure with the lattice constants a and c decreasing monotonically with increasing atomic number of R. CeCu_3Al_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 TbCu_3Al_2 and DyCu_3Al_2 with transition temperature T_{N} of 6 and 5 K, respectively. For SmCu_3Al_2 and GdCu_3Al_2 , ferromagnetic ordering are observed with $T_{\text{C}} = 12$ and 20 K, respectively.

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