Proceedings of the European Conference Physics of Magnetism 2011 (PM'11), Poznań, June 27–July 1, 2011

Thermoelectric Power and Thermal Conductivity of Heavy Fermion $CeCu_4Al$

M. Falkowski and A. Kowalczyk*

Institute of Molecular Physics, Polish Academy of Sciences, M. Smoluchowskiego 17, 60-179 Poznań, Poland

The thermal conductivity and thermopower are discussed for the heavy fermion $CeCu_4Al$ compound. $CeCu_4Al$ is paramagnetic and follows the modified Curie–Weiss law with $\mu_{\rm eff} = 2.53\ \mu_{\rm B}$/f.u. and $\theta_{\rm P} = -10$ K indicating on the presence of well localized magnetic moments of Ce^{3+} ions. The determined electronic specific heat coefficient $\gamma = 2.2$ J mol^{-1}K^{-2} confirms the heavy fermion character of this compound. Thermopower is positive over the whole temperature range and below $T_{\rm max} = 25$ K falls rapidly. Based on a simple band model the position and width of the $4f$ peak nearest to the Fermi level have been estimated. The measured total thermal conductivity of the $CeCu_4Al$ compound increases almost linearly with increasing temperature.

PACS: 72.10.Fk, 72.15.Eb, 72.15.Jf, 72.15.Qm

1. Introduction

The thermoelectric power generation has gained increased attention during the last one and half decade due to its ability to generate electricity from waste heat. According to the thermoelectric figure of merit $ZT = S^2T/\rho\kappa$ (ρ is the electrical resistivity, S — the Seebeck coefficient and κ is the thermal conductivity) a thermoelectric material should possess high thermopower with low electrical resistivity and thermal conductivity. Due to the high S, strongly correlated electron systems are considered as potential candidates for cooling applications at cryogenic temperatures. Both the Seebeck coefficient and the conductivity strongly depend on the carriers concentration and the temperature. An increase in the carriers concentration leads to the decreases of the Seebeck coefficient and the increase of the conductivity.

$CeCu_4Al$ is paramagnetic down to 2 K and follows the Curie–Weiss law with $\mu_{\rm eff} = 2.53\ \mu_{\rm B}$/f.u. and $\theta_{\rm P} = -10$ K [1]. The experimental value of $\mu_{\rm eff}$ is close to the calculated one for a free Ce^{3+} ion (2.54 $\mu_{\rm B}$), thus indicating the presence of well localized magnetic moments carried by the stable Ce^{3+} ions. Below the Fermi energy the total density of states contained mainly d-states of Cu atoms which hybridized with the Ce f electronic states. The analyses of the Ce $3d$ and $4d$ XPS spectra confirmed the localized character of the Ce f-states. The determined electronic specific heat coefficient $\gamma = 2.2$ J mol^{-1}K^{-2} confirmed heavy fermion character of these compounds. The $\rho(T)$ for compound $CeCu_4Al$ shows a Kondo-like logarithmic increase down to a maximum at $T_{\rm max} = 2.9$ K and then a steep decrease towards low temperature [2]. The resistivity of $CeCu_4Al$ represents typical Kondo lattice behavior.

In this paper we describe our studies of the thermopower and thermal conductivity of the $CeCu_4Al$ compound.

2. Experimental

Preparation of sample and experiment are the same as in the paper [1]. The hexagonal $CeCu_5$-type structure was confirmed by the powder X-ray diffraction technique. All the thermoelectric measurements have been carried out using the PPMS commercial device (Quantum Design) in the temperature range 2–300 K using a direct heat-pulse technique.

3. Results and discussion

The thermoelectric power (S) of heavy fermion (HF) compounds below room temperature displays one or several peaks. In most cases these extrema are much larger than those found in the simple metals at the same temperature. A common feature in the thermopower of Ce-based heavy-fermion compounds is that there is almost always a large and broad maximum, whether the system is concentrated or dilute.

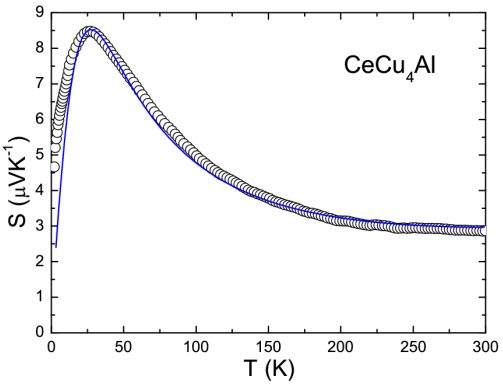

Fig. 1. Temperature dependence of the thermopower for $CeCu_4Al$.

* corresponding author; e-mail: ankow@ifmpan.poznan.pl

The temperature dependence of the thermopower for the sample of $CeCu_4Al$ is displayed in Fig. 1. It is visible that as temperature decreases starting from the room temperature the value of S increases, reaching a relatively high value of 8.5 μV/K at $T_{\max} = 27$ K i.e. at a higher temperature than that of the resistivity maximum ($T = 2.9$ K) [2]. The thermopower of $CeCu_4Al$ is positive over the whole temperature range and below $T_{\max}$ falls rapidly. Similar observations were presented in the previous studies [3]. The existing discrepancy between our and the reported data may be attributed to differences in the sample quality. The positive value of $S(T)$ may indicate the possibility of holes as the dominant charge carrier in $CeCu_4Al$.

The temperature dependence of the thermopower in the Kondo lattice systems arises from two contributions: the Kondo scattering at the ground state and incoherent scattering at excited crystal electric field levels. The former mechanism gives rise to a first maximum at low temperature T, being roughly equal to T_K [4], while the latter one appears as a broad high temperature anomaly at $T_{\max} \approx (1/3\text{–}1/6)\ \Delta_{CEF}$, with Δ_{CEF} being the overall CEF splitting [5]. Using this model Δ_{CEF} of $CeCu_4Al$ can be located between 80 to 160 K. Derived from specific heat measurements Δ_{CEF} and T_K are 120 K and 5 K, respectively [6]. As only one single feature is resolved in $S(T)$ in $CeCu_4Al$, it can be assumed that it originates from the combined Kondo scattering at the ground state doublet and at the excited doublets of the Ce^{3+} ion.

For many f-electron systems, one can apply the phenomenological resonance model [7] to describe their low-temperature thermopower data. According to this model the dominant contribution to $S(T)$ is caused by scattering between electrons of a broad s-band and a narrow f-band with the Lorentzian shape. There are two important parameters: the position of the f-electron band relative to the Fermi level, $\varepsilon_f - \varepsilon_F$ and the width of the resonance peak Γ. The temperature dependence of $S(T)$ of HF compounds can be written as follows:

$$S(T) = \frac{2}{3}\frac{k_B}{|e|}\pi^2 \frac{T\varepsilon_f}{(\pi^2/3)T^2 + \varepsilon_f^2 + \Gamma_f^2} + S_d, \qquad (1)$$

where $S_d = aT$ is Mott's diffusion term.

As a result we get a rough estimation of the parameters, $\Delta = 0.23$ meV, $\Gamma = 4.32$ meV and $a = 4.7 \times 10^{-3}$. The positive value for $CeCu_4Al$ indicates that the DOS peak is supposedly just above the Fermi level. This is consistent with theoretical calculations, which show that the Fermi level is situated on the left side of the prominent f-states peak [1].

The temperature dependence of the thermal conductivity $\kappa(T)$ for $CeCu_4Al$ is given in Fig. 2. The thermal conductivity κ of a metal has two components: an electronic component κ_e and the lattice component κ_L for which the phonons are the heat carriers. It follows:

$$\kappa(T) = \kappa_e(T) + \kappa_L(T). \qquad (2)$$

The electronic thermal conductivity κ_e depends both on the temperature and the carrier concentration. The electronic thermal conductivity is closely related to the conductivity of the material and in a simple model (a single band with a parabolic energy-wave number relation, $E \propto k^2$) it is described by the Wiedemann–Franz law $\kappa_e = L_0 T/\rho$ ($L_0 = 2.45 \times 10^{-8}$ W Ω K^{-2} is the Lorentz number). The $Ce(Cu_{1-x}Al_x)_5$ and $La(Cu_{1-x}Al_x)_5$ alloys exhibit a very different change of behavior in thermal conductivity by a change of the Al content [8]. For the boundary compounds $CeCu_5$ and $LaCu_5$ the lattice thermal conductivity is practically negligible. With the increase of Al concentration grows the lattice contribution to the total thermal conductivity.

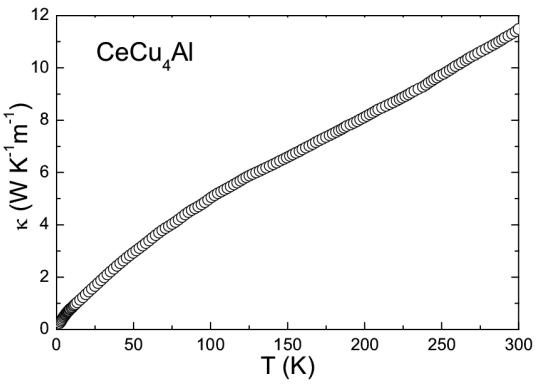

Fig. 2. Temperature dependence of the thermal conductivity for $CeCu_4Al$.

For $CeCu_4Al$, $\kappa(T)$ is proportional to T below 20 K. Since the linear $\kappa(T)$ is typical for scattering of electrons on lattice imperfections, we suggest that this mechanism plays essential role. For the highest temperature range ($T > 150$ K), the $\kappa(T)$ (Fig. 2) changes also linearly with temperature. The linear dependence $\kappa(T)$ is not predicted theoretically, but it had been observed in many rare earth compounds.

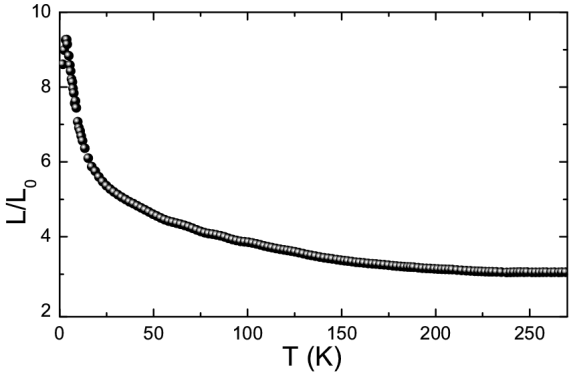

Fig. 3. Temperature dependence of the reduced Lorentz number L/L_0.

Figure 3 shows the temperature dependence of the reduced Lorentz number L/L_0. The Lorentz number, L

is defined by $\kappa\rho/T$. The L/L_0 of $CeCu_4Al$ increases rapidly on cooling and reaches a maximum value of 9 at 4 K. With further decrease of temperature L/L_0 becomes smaller. The values of L/L_0 for $CeCu_4Al$ are typical of heavy fermion compounds. A large L/L_0 indicates that the dominant heat carriers are phonons, and the spin scattering of charge carriers does not play a significant role.

From the application viewpoint, the efficiency of thermoelectric material is given by the dimensionless figure of merit $ZT = S^2T/\rho\kappa$. It is clear that the main problem to get a good thermoelectric material results from the fact that one has to minimize the thermal conductivity, while enhancing the electrical conductivity. For the present system, $ZT \ll 1$ at room temperature. The low ZT values of $CeCu_4Al$ are the consequence of too low electrical conductivity resulting from non-optimal concentration of current carriers in this material.

4. Conclusions

The thermopower of $CeCu_4Al$ is positive over the whole temperature range and increases with decreasing temperature until a maximum of 8.5 μV/K at $T = 27$ K. Based on a simple band model the position and width of the $4f$ peak nearest to the Fermi level have been estimated. The $4f$ peak is probably located just above E_F.

For $CeCu_4Al$ in the temperature range studied, the thermal conductivity increases almost linearly with increasing temperature. The scattering of electrons on the lattice imperfections is elastic and this mechanism is most important at low temperatures. In contrast, the phonon–electron and phonon–phonon interactions may have elastic as well as inelastic character and they are described by processes of the normal and the Umklapp type. The reduced Lorentz number L/L_0 of $CeCu_4Al$ increases with decreasing temperature and reaches a maximum value at low temperatures. A large L/L_0 at low temperatures indicates that the dominant heat carriers are phonons.

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