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Thermoelectric Properties of $CeCu_4Ag$ Compound

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The magnetic and electronic properties including electrical resistivity, thermal conductivity and thermopower are discussed for the heavy fermion $CeCu_4Ag$ compound. The electrical resistivity shows a Kondo-like logarithmic increase up to a maximum at $T = 75$ K. Thermopower is positive over the whole temperature range and below $T_{\max} = 25$ K falls rapidly. The measured thermal conductivity of the $CeCu_4Ag$ compound increases with increasing temperature.

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1. Introduction

The parent $CeCu_5$ compound was identified as a Kondo lattice compound exhibiting antiferromagnetism below 4 K [1]. The substitution of M (M = Ga, Al, In) into $CeCu_5$, which increases the average electron density, seems to be sufficient to screen all the localized $4f$ moments in $CeCu_4M$, which leads to a crossover from a magnetic ground state in $CeCu_5$ to a nonmagnetic ground state in $CeCu_4M$ [1–3].

Preparation of sample and experiment are the same as in the paper [3]. The X-ray diffraction (XRD) measurements have shown that $CeCu_4Ag$ crystallizes in the cubic $MgSnCu_4$-type structure (space group F-$43m$, a = 7.273 Å), however, a contribution of secondary phases has been detected. $CeCu_4Ag$ appeared to be paramagnetic with the effective magnetic moment $\mu_{\text{eff}} = 2.54\ \mu_{\text{B}}$ and the paramagnetic Curie temperature $\theta_{\text{p}} = -33$ K. We have observed a typical heavy fermion (HF) behaviour with γ value of about 0.5 J mol^{-1} K^{-2} (not shown here).

2. Results

The temperature variation of the electrical resistivity $\rho(T)$ of $CeCu_4Ag$ represents (Fig. 1) typical Kondo lattice behavior. The $\rho(T)$ shows a Kondo-like logarithmic increase up to a maximum at $T_{\max} = 75$ K. The sharp drop in the resistivity below $T_{\max}$ can be developed by the coherence onset in a Kondo lattice or by the crystalline electric field (CEF) split ground state alone. The temperature dependence of resistivity above the maximum can be described by the formula $\rho(T) = \rho_0 + \rho_0^{\infty} - c_{\text{K}} \ln T$, where the first term accounts for the scattering of the conduction electrons on the lattice defects and disordered magnetic moments and the second term represents the Kondo effect. This dependence is better visible on the logarithmic scale in the inset of Fig. 1. The fit parameters are $\rho_0 + \rho_0^{\infty} = 331.5\ \mu\Omega$ cm and $c_{\text{K}} = 57.9\ \mu\Omega$ cm, respectively.

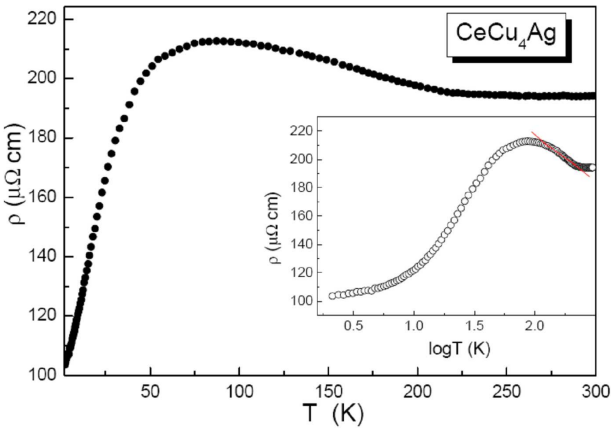

Fig. 1. Temperature dependence of the electrical resistivity for $CeCu_4Ag$. Inset: a low temperature part of $\rho(T)$ vs. $\log T$.

The temperature dependence of the thermopower for the sample of $CeCu_4Ag$ is displayed in Fig. 2. It is visible that as temperature decreases starting from the room temperature the value of S increases, reaching a relatively high value of 50 μV/K at $T_{\max} = 25$ K. The thermopower of $CeCu_4Ag$ is positive over the whole temperature range and below $T_{\max}$ falls rapidly. The positive value of $S(T)$ may indicate the possibility of holes as the dominant charge carriers in $CeCu_4Ag$. The peak at 25 K is probably due to the CEF effect, phonon drag effect and/or the inelastic scattering of charge carriers by acoustic phonons.

According to phenomenological resonance model [4] the dominant contribution to $S(T)$ is caused by scat-

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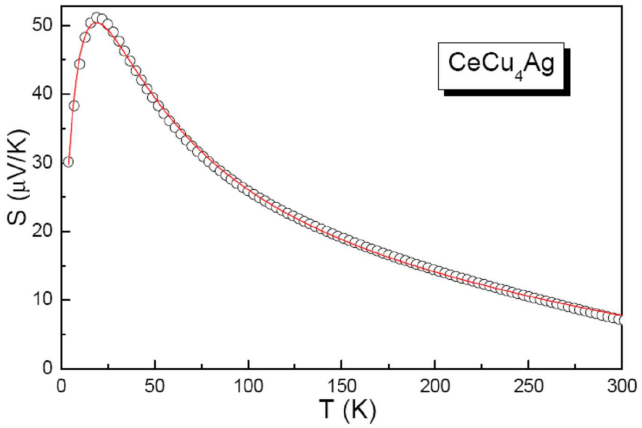

Fig. 2. Temperature dependence of the thermopower for $CeCu_4Ag$.

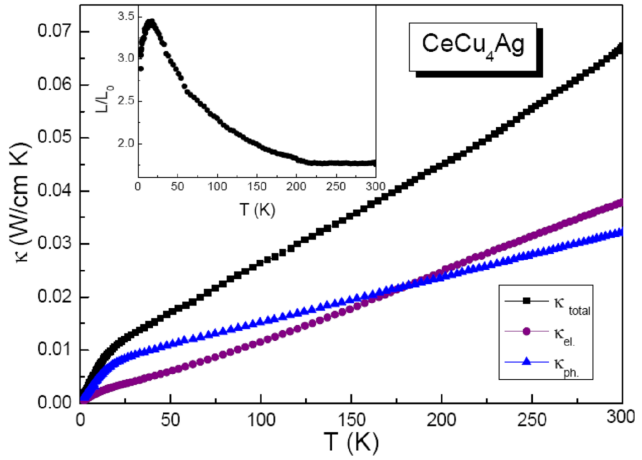

Fig. 3. Temperature dependence of the thermal conductivity for $CeCu_4Ag$. Inset: temperature dependence of the reduced Lorentz number L/L_0.

tering 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:

$$S(T) = \frac{2(\varepsilon_f - \varepsilon_F)T/|e|}{\frac{3[(\varepsilon_f-\varepsilon_F)^2+\Gamma^2]}{(\pi k_B)^2} + T^2} \,. \tag{1}$$

For the range 2–300 K the thermopower data can be described by an excitation $\varepsilon_f - \varepsilon_F = 0.7$ meV and the width $\Gamma = 7.8$ meV. The positive value for $CeCu_4Ag$ indicates that the density of states (DOS) peak is supposedly just above the Fermi level. The suspicion of an excitation close to the Fermi level supports the possibility of a resonance peak development, which is one of the essential features of the Kondo lattices.

The thermoelectric power factor $PF = S^2/\rho$ reaches the maximum value of 18 μW/(cm K^2) at $T = 25$ K. This PF value is smaller than for the conventional thermoelectric material Bi_2Te_3 ($PF = 40$ μW/(cm K^2) at $T = 300$ K) [4].

In nonmagnetic materials, the thermal conductivity is generally composed of two contributions, the electronic κ_e and the lattice contribution κ_{ph}, i.e., $\kappa = \kappa_e + \kappa_{ph}$. 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 $\kappa_e = L_0 T/\rho$ ($L_0 = 2.45 \times 10^{-5}$ W Ω K^{-2} is the Lorentz number).

The measured thermal conductivity of the $CeCu_4Ag$ compound increases with increasing temperature (Fig. 3). The lattice thermal conductivity κ_{ph}, obtained by subtracting κ_e from the observed κ is also plotted in Fig. 3. The electronic and lattice contributions are similar not only in their magnitudes but also in the overall temperature changes. From this figure it is clear that above about 180 K the electronic term dominates. Below this temperature it is suppressed. The scattering of electrons and phonons 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 inset of Fig. 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 $CeCu_4Ag$ increases rapidly on cooling and reaches a maximum value of 3.5 at 25 K. With further decrease of temperature L/L_0 becomes smaller. A large L/L_0 indicates that the dominant heat carries are phonons, and the spin scattering of charge carries does not play a significant role.

Acknowledgments

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