Thermal-transport properties of CeNiSn

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Abstract

We present thermopower $S$ and thermal-conductivity $\kappa$ measurements on high-quality single crystalline CeNiSn along the three crystallographic axes $a$, $b$, and $c$ in the temperature range between $100 \text{ mK}$ and $7 \text{ K}$ and in magnetic fields up to $8 \text{ T}$, applied along the $a$ axis. Both $\kappa$ and $S$ are highly anisotropic. However, below $10 \text{ K}$, characteristic features that may be attributed to the opening of a pseudogap in the charge-carrier density of states (DOS) at the Fermi energy $\eta$ are seen for all three crystallographic directions. These features are strongly suppressed by a magnetic field of $8 \text{ T}$. At the lowest temperatures we have evidence for the presence of a residual charge-carrier density, again for all the three directions. © 2001 Elsevier Science B.V. All rights reserved.

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The claim [1] that CeNiSn is one of the rare examples of valence-fluctuating Cerium compounds with an energy pseudogap in the electronic density of states (DOS) at $\eta$ triggered strong interest in this compound. Our main concern in the present work is a potential residual DOS within the pseudogap. Thermal-transport measurements at low temperatures are a valuable tool for extracting information on such a residual DOS. In particular, measurements along the three crystallographic axes provide information on the anisotropy of the residual charge-carrier density. Our single-crystalline CeNiSn samples were grown by a Czochralski method and purified by the technique of solid-state electro-transport (SSE), which is crucial for improving the sample quality [2]. The $S$ and $\kappa$ measurements to be presented below are the first on SSE-treated CeNiSn. A comparison with low-temperature data on less pure CeNiSn [3] shows that $S$ is more sensitive to impurities than $\kappa$.

$S(T)$ is highly anisotropic and shows several well-defined structures (Fig. 1). We interpret these $S(T)$ data in terms of a diffusion thermopower, since possible phonon-drag and paramagnon-drag contributions are expected to be small in the temperature range investigated here. In systems like CeNiSn containing both almost localized $4f$ and delocalized conduction electrons, the thermopower may be ascribed [4] to the conduction electrons, being scattered from the $4f$ electrons. In a first approximation, $S$ is proportional to $T(\partial \ln N_e/\partial \epsilon_f - \partial \ln N_f/\partial \epsilon_h)_\eta$ where $N_e$ and $N_f$ are the conduction- and $4f$-electron DOS, respectively. $(\partial \ln N_f/\partial \epsilon_h)_\eta$ may reach very large values compared to the corresponding expression in d-band metals. Also $(\partial \ln N_e/\partial \epsilon_h)_\eta$ may contribute non-negligibly to $S$, because the charge-carrier concentration is known to decrease steeply upon cooling below $5-10 \text{ K}$ [5]. The anomalies (maxima along $a$ and $b$, minimum along $c$-axis) between 2 and $7 \text{ K}$ reflect extreme values in the energy derivative of $N_e$ and/or $N_f$, which we relate to the opening of the pseudogap. They are strongly suppressed by a magnetic field of $8 \text{ T}$ (Fig. 1). In the Kondo-lattice model, a destruction of the coherence gap is expected at low magnetic fields if the RKKY and the Kondo interactions are of similar strength [6]. The features in $S(T)$ at even lower temperatures are most probably the signs of a temperature-dependent DOS structure at the Fermi level within the gap. Their unusual anisotropic magnetic-field dependence indicates that the magnetic field leads to a charge redistribution in $k$-space.

Similar to $S(T)$, $\kappa(T)$ is highly anisotropic (Fig. 2). However, anomalous enhancements of $\kappa$ are observed for...
Fig. 1. Temperature dependence of the thermopower $S(T)$ of CeNiSn at 0, 4 and 8 T along the $a$-, $b$-, and $c$-axis in panel (a), (b), and (c), respectively.

Fig. 2. Temperature dependence of the thermal conductivity $\kappa(T)$ of CeNiSn at 0, 4 and 8 T along the $a$-, $b$-, $c$-axis in panel (a), (b), and (c), respectively. The solid curve in panel (a), $\kappa_{\text{el}}^W(T)$, represents the electronic thermal conductivity along the $a$-axis obtained from the Wiedemann–Franz law. The insets show close-ups at low temperatures. The lines are best linear fits to the data.

all three directions: A pronounced maximum for $\kappa_a(T)$ at 3.8 K, a very shallow maximum for $\kappa_b(T)$ at 5 K, and a plateau for $\kappa_c(T)$ at 4.5 K. Application of a magnetic field leads to a reduction of all these enhancements. The electronic contribution to the thermal conductivity along the $a$-axis, calculated from the electrical resistivity along the $a$-axis under the assumption that the Wiedemann–Franz law is valid, is relevant only below 1 K (Fig. 2). Below 300 mK, $\kappa$ is a linear function of $T$ for all three directions (insets of Fig. 2) suggesting that in this temperature range the heat is predominantly carried by charge carriers. This is strong evidence for a residual charge-carrier density for all the three crystallographic directions. The anisotropic magnetic-field dependence of the linear term again points to a field-induced charge-carrier redistribution in $k$-space. Between 600 mK and 2 K, $\kappa \propto T^2$, a temperature dependence commonly attributed to a phononic thermal conductivity with predominant phonon–electron scattering. Making the reasonable assumption that phonon–electron scattering remains dominant up to at least 7 K, the above-mentioned enhancements in $\kappa(T)$ can be explained as follows: When the gap starts to open upon cooling below 10 K, electrons start to freeze out which reduces the phonon–electron scattering rate and leads to the observed enhancements in $\kappa(T)$. This mechanism was already proposed in Ref. [7]. Our analysis provides strong evidence for this scenario. Finally, we would like to note that the reduced Lorenz number $L/L_0$ measured along the $a$ axis saturates below 200 mK to the value 1.5, which is distinctly enhanced over 1. This is a striking observation which deserves further investigation. A more detailed analysis of our data will be given elsewhere [8].

References