Field dependence of the thermopower of CeNiSn

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Abstract. Previously measured thermopower data of CeNiSn exhibit a significant sample dependence and non-monotonous behavior in magnetic fields. In this paper we demonstrate that the measured thermopower $S(T)$ may contain a contribution from the huge Nernst coefficient of the compound, even in moderate fields of 2 T. A correction for this effect allows to determine the intrinsic field dependence of $S(T)$. The observed thermopower behavior can be understood from Zeeman splitting of a V-shaped pseudogap in magnetic fields.

Introduction: The orthorhombic system CeNiSn has been classified as a Kondo semimetal, in which an anisotropic pseudogap opens in the density of states (DOS) below approximately 10 K [1]. Various experimental probes confirm the presence of a finite quasiparticle DOS at the Fermi level, such as the metal-like resistivities of samples with high purity [1] and the linear-in-$T$ dependence of the thermal conductivity below 0.3 K [2]. Magnetic fields of the order of 10 T along the easy magnetic $a$ axis suppress the gap formation significantly, while fields along $b$ and $c$ are less effective [2, 3]. The thermopower $S$ of CeNiSn is highly anisotropic and exhibits a significant sample dependence [2, 4, 5, 6]. Below 10 K, the absolute values of $S$ are enhanced, which has been attributed to the gap formation in this temperature range [1, 5, 6]. Consequently, application of a magnetic field of 8 T along the easy $a$ axis has been found to induce a significant lowering of the thermopower along $a$, $S_a$, at low $T$ [1, 2]. However, the experimental data for $S_b$ and $S_c$ with $B \parallel a$ are inconsistent. First measurements at 4.2 K and 1.3 K showed a lowering of $S_c$ in magnetic fields [5], while investigations on samples of higher purity revealed an increasing $S_c$ upon increasing field [2]. Likewise, for $S_b$ at 1.3 K either a monotonous decrease up to 10 T [5] or an increase in 4 T with a subsequent decrease in 8 T [2] was found.

For correlated semimetals as CeNiSn a large Nernst coefficient has been predicted [7]. The corresponding transverse thermal voltage can become comparable in magnitude to the longitudinal one even in moderate magnetic fields. In such a case, small deviations from the ideal contact geometry may give rise to a non-negligible Nernst contribution to the measured thermopower, an effect which has not been considered previously [5]. It is expected to be most relevant for $S_b$, which exhibits significantly lower absolute values at low temperatures than $S_a$ and $S_c$. In this work we discuss the intrinsic behavior of $S_b(T)$ for $B \parallel a$ and $c$ determined from measurements in positive and negative fields up to 7 T.

Experiments: The investigated samples originate from the single crystal #5, which was grown by the Czochralski technique and subsequently purified by the solid-state electrotransport (SSE)
Exemplary error bars indicate the uncertainty of the data at low $T$ and high $B$. Above 4 K it is typically less than 1 $\mu$V/K. The inset compares the zero-field data obtained on two samples of CeNiSn. (b) The in-field antisymmetric contribution of the measured thermopower $\frac{1}{2} \Delta S_{\text{meas}}$ in comparison to the Nernst signal $N$ in 2 T. $\frac{1}{2} \Delta S_{\text{meas}}$ can be scaled to $N$ by a factor of 8.

**Results:** The zero-field thermopower of CeNiSn measured along $b$, $S_b(T)$, is shown in the inset of Fig. 1. The data sets obtained on two different samples agree relatively well in the whole investigated temperature range. Above 10 K, the thermopower exhibits a similar $T$ dependence as reported for a high-purity single crystal without SSE treatment [6]: $S_b(T)$ is positive with maxima at 20 K and 100 K. The two maxima are attributed to Kondo scattering from the ground-state doublet and thermally populated CEF levels. A contribution from paramagnon drag was also suggested as a possible origin for the maximum at 20 K [2]. Toward lower $T$, $S_b(T)$ changes sign at 8 K and goes through a large negative minimum of $-30$ $\mu$V/K at around 3.5 K. This value represents the largest negative $S$ ever observed for a CeNiSn sample. Below 1.7 K the thermopower assumes again positive values. A similar temperature dependence has been observed in samples grown by a Czochralski technique without SSE treatment [5]. These samples exhibit a negative $S_b$ between 2.5 and 7 K, however, with significantly smaller absolute values of at most -6 $\mu$V/K. By contrast, investigations on single crystals of similar quality as those presented here yielded a thermopower $S_b(T)$ with opposite sign and a maximum value of 8 $\mu$V/K at 3.5 K [2].

The effect of a magnetic field along $a$ on the low-$T$ thermopower $S_b$ is shown in the main plot of Fig. 1. With increasing $B$ the minimum at $T_{\text{min}} = 3.5$ K shifts to lower $T$ and the absolute values at the minimum $|S_{\text{min}}|$ are enhanced for $B \leq 4$ T. In 7 T a weak lowering of $|S_{\text{min}}|$ is observed, which, however, is of the order of the uncertainty in the data. Application of a magnetic field along $c$ gives rise to a similar evolution of $S(T)$ (not shown). Compared to the configuration $B \parallel a$, the shift of the minimum is less pronounced and no lowering of $|S_{\text{min}}|$ is observed around
Figure 2. Field dependence of the minimum temperature in $S_b(T)$, $T_{\text{min}}$ (a), and the thermopower value at $T_{\text{min}}$, $S_{\text{min}}$ (b), for different orientations of $B$. The dashed lines are meant guides to the eye. The solid line in the left plot is a linear fit to the data for $B \parallel a$. The error bars given for $S_{\text{min}}$ represent the scattering of the data around $T_{\text{min}}$.

7 T, (cf. Fig. 2). Fig. 2a shows the field dependence of $T_{\text{min}}$ for $B \parallel a$ and $B \parallel c$. It clearly reveals that the shift of the minimum is stronger for $B \parallel a$. A linear extrapolation of $T_{\text{min}}(B \parallel a)$ to zero temperature yields a critical field of 14 T. This value is comparable to the energy-gap quenching field along $a$ of 18 T determined from resistivity measurements [8]. Therefore, it is supposed that the shift of the minimum is related to the closing of the gap in field. Fig. 2b shows the evolution of the thermopower value at the minimum, $S_{\text{min}}$ for $B \parallel a$ and $B \parallel c$. While $|S_{\text{min}}|$ first increases with increasing field, a saturation and subsequent reduction is observed at higher $B$. Again, the effect of a field along $a$ is more pronounced than that of $B \parallel c$. It is suspected that $|S_{\text{min}}|$ for both orientations is further reduced in higher $B$ as the minimum shifts to lower $T$.

Discussion: The strong sample dependence of $S(T)$ of CeNiSn reported in literature has been related to the differing purity of the investigated crystals [2]. However, two other effects have to be taken into account. Firstly, in view of the sensitive direction dependence of $S(T)$ it cannot be excluded that tiny misorientations are responsible for at least part of the reported variations. The large negative $S_b$ observed around 3 K in the present investigation could be easily diminished by a small contribution from the huge positive $S_a$ and $S_c$ of up to 70 $\mu$V/K expected in the same temperature range [2, 4, 5, 6]. In this context, the small discrepancy of about 20 % between $S_{\text{min}}(B = 0)$ of sample 1 and 2 might be attributed to this effect. Secondly, the large Nernst signal expected for CeNiSn can contribute to the thermopower voltage for a non-ideal contact geometry. In the present investigation, this effect was already appreciable in 2 T as demonstrated in Fig. 1b. Below 8 K the measured thermopower curves $S_{\text{meas}}(T)$ for positive and negative fields differ significantly. The in-field antisymmetric contribution $\frac{1}{2}\Delta S_{\text{meas}} = \frac{1}{2}[S_{\text{meas}}(+2T) - S_{\text{meas}}(-2T)]$ exhibits a temperature dependence similar to that of the Nernst signal $N = E_y/\nabla_x T$ in 2 T and can be scaled to it by a factor of 8. This corresponds to a misorientation of the contacts by only 7°. For the presented data this effect has been corrected for by averaging between $S_{\text{meas}}(+B)$ and $S_{\text{meas}}(-B)$. However, it has generally not been accounted for in previous investigations [5]. In particular, the drastic and non-monotonous change in $S_b$ around 2 K reported for crystals of similar quality as those investigated here [2] might be to some extent influenced by the huge Nernst signal in this $T$ range. Therefore, the current study represents the first investigation with the intrinsic temperature dependence of $S_b$ in fields up to 7 T.
Application of magnetic fields $B \parallel a, c$ is found to induce a systematic shift of the thermopower minimum at 3.5 K to lower temperatures (cf. Fig. 2b). The estimated critical field along $a$ of 14 T as well as the stronger effect for $B \parallel a$ compared to $B \parallel c$ confirms that the minimum is related to the gap formation in CeNiSn. In this context, the enhancement of $|S_{\text{min}}|$ in parallel with the closing of the gap in field (Fig. 2b) is surprising. Apparently, application of a magnetic field does not only suppress the gap but also influences the residual DOS inside the gap. A similar effect has been found from investigations of the specific-heat $c_p$ of CeNiSn [9], which revealed an enhancement of $c_p$ at low $T$ in applied magnetic fields $B \parallel a$. This observation had been interpreted within a simple model assuming Zeeman splitting of a modified V-shaped DOS [9]. An illustration, how the same mechanism can give rise to an enhancement of $|S|$ at low $B$ is depicted in Fig. 3. Application of a magnetic field induces a shift of the sub-bands for the spin-up and spin-down states to different energetic directions, as shown for an arbitrary field in Fig. 3a. Thus, the resulting total DOS around the gap structure depends sensitively on the field magnitude (Fig. 3b). If the Fermi level is situated slightly off the symmetry line of the gap structure, the slope of the DOS at $E_F$ becomes field dependent (Fig. 3c). Within this simplified picture, the absolute values of the thermopower $S \propto T(\partial \ln N(\epsilon)/\partial \epsilon)|_{E_F}$ [2] increase in small magnetic fields and decreases for higher $B$. It is admitted, that the sketched behavior is much too simple to explain in detail the behavior of $S_b(T,B)$ observed for CeNiSn. Nevertheless, the presented picture demonstrates that the increase in $|S|$ is not fundamentally contradictory to the suppression of the gap in magnetic fields. Zeeman splitting appears a possible and simple mechanism to understand the observed behavior. It seems likely that a realistic band structure and the allowance for thermal broadening enables a more sophisticated description of the data. In conclusion, the minimum in $S_b(T)$ of CeNiSn at 3 K is attributed to the gap formation and the observed field dependence is related to the suppression of the gap in magnetic fields.

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