Thermoelectric Power of Single-Crystalline Ce$_2$RhSi$_3$

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High-quality single crystal of Ce$_2$RhSi$_3$ was studied by means of thermoelectric power measurements carried out down to 2 K in external magnetic fields up to 13 T. The results obtained above 50 K were interpreted in terms of a modified two-band model that takes into account temperature variation of the width of $4f$-derived narrow band located near the Fermi level. At lower temperatures the thermopower exhibits more complex temperature dependences that likely involve interplays of magnetic exchange, Kondo and crystal-field interactions.

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1. Introduction

Cerium- and uranium-based ternary silicides with the composition R$_2$TSi$_3$, where R = Ce or U and T stands for a d-electron transition metal, usually crystallize with the hexagonal AlB$_2$-type structure or with one of its several derivatives. These compounds exhibit large variety in magnetic behavior, from the Pauli paramagnetism to long-range orderings. Frequently, spin-glass phenomena are observed at low temperatures that are related to atomic disorder and topological frustration in triangular magnetic sublattices [1].

The compound Ce$_2$RhSi$_3$ was reported to adopt a fully-ordered hexagonal unit cell of the Er$_2$RhSi$_3$-type, with both lattice parameters doubled with respect to the AlB$_2$-type [2, 3]. It was found to order antiferromagnetically below $T_N = 6$–7 K [2, 4, 5] with a simple collinear arrangement of the Ce magnetic moments along the b-axis of an orthorhombic magnetic structure [6]. The magnetic moment derived by neutron diffraction amounts to 1.3 μ_B at 1.3 K [6]. Most recent studies of the magnetic susceptibility and the electrical resistivity under hydrostatic pressure as well as ambient-pressure heat capacity measurements revealed that Ce$_2$RhSi$_3$ is a Kondo lattice located near the maximum T_N on the Doniach magnetic phase diagram [5].

All the previously reported studies of Ce$_2$RhSi$_3$ were made on polycrystalline samples. Recently we succeeded in growing high-quality single crystals of this material and undertook a systematic re-investigation of its physical behavior. The structural, magnetic, electrical and thermodynamic properties, measured along the principal directions in the hexagonal unit cell, have been described

in detail in [7], while here we present the thermoelectric characteristics of this interesting compound.

2. Experimental details

Single crystal of Ce$_2$RhSi$_3$ was grown by the Czochralski pulling method in a tetra-arc furnace under ultra-pure argon atmosphere and checked for its quality as described in [7]. Thermoelectric power measurements were performed from 2 to 300 K in magnetic fields up to 13 T employing an Oxford Instruments TESLATRON platform and using copper as a reference material. The Seebeck coefficient was determined with the temperature gradient ΔT, being linearly changed from 0.5 K at low temperatures up to 2 K near room temperature, which was applied along the a- and c-axes of the hexagonal unit cell, with the magnetic field B aligned parallel to ΔT.

3. Results and discussion

Figure 1 presents the temperature dependences of the thermoelectric power of single-crystalline Ce$_2$RhSi$_3$, taken in zero external magnetic field. At 300 K the value of S_a ($\Delta T \parallel a$) and S_c ($\Delta T \parallel c$) is about 9 μV/K. The positive sign of the thermopower at room temperature indicates that probably holes are the majority carriers in this compound. With decreasing temperature some Seebeck effect anisotropy becomes evident and both thermopower curves form broad maxima centered near 200 K for $S_a(T)$ and 150 K for $S_c(T)$. The anisotropy vanishes near 25 K, where both components change their sign to negative. At lower temperatures other sign changes are observed for S_c that is again positive in between 13 and 4 K, whereas S_a stays negative down to 2 K (the terminal temperature in the present study). Despite this difference the shapes of $S_a(T)$ and $S_c(T)$ are very similar to one another: they

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go first through a minimum (at 14 K and 18 K, respectively) and then form a sharp maximum (at 5 K and 6 K, respectively). For each component another negative minimum is expected to occur below 2 K in order to reach $S_a = 0$ and $S_c = 0$ at $T = 0$.
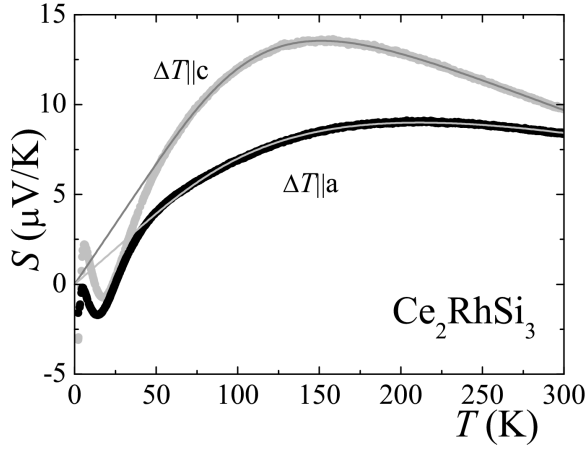

Fig. 1. Temperature variations of the thermoelectric power of Ce_2RhSi_3 measured along the a- and c-axes. The solid lines are least-squares fits to the experimental data of the function defined in Eq. (2).

In the paramagnetic region, the overall behavior of the thermopower of Ce_2RhSi_3 is similar to that reported for Ce-based Kondo lattices, e.g. $CeAl_3$, CePdSn, CePdGe, or CePtGe, and classified as "type (b)" being explainable in the framework of the Coqblin–Schrieffer model [8]. As shown in Fig. 1, above 50 K, $S_a(T)$ and $S_c(T)$ may also be properly described within the so-called two-band approach that originates from the Hirst model [9]. In this simple theory it is assumed that the conduction electrons are scattered by a $4f$-derived quasiparticle band of a Lorentzian form, located at ϵ_F below the Fermi level. The band-width Γ may generally be temperature dependent according to

$$\Gamma = T_f e^{-T_f/T}, \qquad (1)$$

where the parameter T_f is related to the width of a quasielastic line due to the Kondo effect that is measured by inelastic neutron scattering experiments [10]. Then, the thermoelectric power of a Kondo lattice can be expressed by the formula [10, 11]:

$$S(T) = C_1 T + C_2 \frac{T \epsilon_F}{\epsilon_F^2 + \Gamma(T)^2}, \qquad (2)$$

where C_1 and C_2 are temperature independent parameters, which determine the strength of the contributions arising from the nonmagnetic Mott-type and the magnetic scattering processes, respectively. Applying Eq. (2) to the thermopower data of Ce_2RhSi_3 one obtains a quite good approximation of the experimental curves above 50 K, and the so-derived values of the least-squares fitting parameters are as follows: $C_1 = -0.19 \ \mu V/K^2$, $C_2 = 51.0 \ \mu V/K$, $\epsilon_F = 185$ K, $T_f = 142$ K for $\Delta T \parallel a$ and $C_1 = -0.07 \ \mu V/K^2$, $C_2 = 20.6 \ \mu V/K$, $\epsilon_F = 101$ K,

$T_f = 194$ K for $\Delta T \parallel c$. The obtained parameters reflect the anisotropy of the thermoelectric power of the Ce-compound studied. They are similar to those derived within the same approach for some other $4f$- and $5f$-electron intermetallics with strong electronic correlations [10–13]. Apparently, the two-band model fails at lower temperatures where other effects, neglected in this simple theory, should be taken into account like phonon-drag, scattering on crystal-field levels, Kondo coherence and/or long-range magnetic ordering.
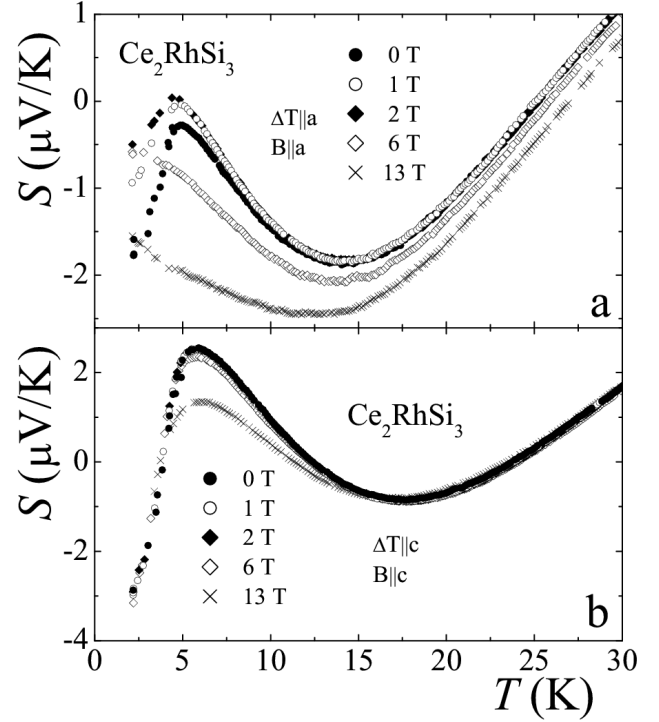

Fig. 2. Temperature dependences of the thermopower of Ce_2RhSi_3, measured in an external magnetic field of different strength, applied along the temperature gradient that was oriented along (a) the a-axis and (b) the c-axis of the hexagonal unit cell.

In order to get an insight into the role of the above-mentioned phenomena in the low-temperature thermoelectric power behavior of Ce_2RhSi_3, the Seebeck coefficient measurements were performed in finite magnetic fields applied in the same direction as the temperature gradient. The results are summarized in Fig. 2. For $\Delta T \parallel a$, one finds that rising the field strength up to 2 T causes a shift of the maximum in $S_a(T)$ towards lower temperatures, accompanied by some increase in its magnitude. In stronger fields the maximum vanishes. Such a behavior is characteristic of antiferromagnets that undergo a field-induced metamagnetic phase transition. In contrast, for $\Delta T \parallel c$, the thermopower is hardly affected by magnetic field up to as strong field as 6 T. Only in 13 T one observes some suppression of the maximum in $S_c(T)$ with the general shape of this curve being unal-

tered. Based on these findings one may conclude that the low-temperature maxima in the thermoelectric power of Ce_2RhSi_3 are related to the long-range antiferromagnetic ordering, which is modified by the Kondo interactions associated with a doublet crystal-field ground state. The observed anisotropic properties of the Seebeck coefficient are consistent with the magnetic structure of the compound where a- and c-axes are easy and hard magnetic directions, respectively [6].

4. Conclusion

The thermoelectric power of single-crystalline Ce_2RhSi_3 was measured in wide temperature and magnetic field ranges. At low temperatures it shows an anisotropic behavior related to the antiferromagnetic ordering. Most likely it is also influenced by the coherence in the Kondo screening interactions. In the paramagnetic region the temperature dependences of the Seebeck coefficient measured along the main crystallographic axes can be effectively described by a phenomenological model which takes into account scattering processes of the conduction electrons from a broad s–d band into a narrow $4f$ quasiparticle band. The overall shape of the temperature variations of the thermopower of Ce_2RhSi_3 is similar to that typical of dense Kondo systems, and hence it is tempting to apply to this compound a more sophisticated theoretical approach based on the Coqblin–Schrieffer model that takes into account crystal field interactions as well as spin and valence fluctuations [8, 14]. This issue will be addressed in our forthcoming paper.

Acknowledgments

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