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Multi-Step Magnetic Phase Transition in $CeRh_3Si_2$ Studied by Specific Heat Measurements

A.P. Pikul and D. Kaczorowski

Institute of Low Temperature and Structure Research

Polish Academy of Sciences, P Nr 1410, 50-950 Wrocław 2, Poland

High-quality single crystal of a novel cerium silicide $CeRh_3Si_2$, crystallizing with the orthorhombic $ErRh_3Si_2$-type structure, was grown by the Czochralski pulling method and studied by means of specific heat measurements. The antiferromagnetic ordering of the compound manifests itself as a pronounced lambda-shaped anomaly at $T_N = 4.72(3)$ K, and is followed by a spike-shaped anomaly due to spin reorientation at $T_t = 4.48(2)$ K, in good agreement with the previously reported magnetic susceptibility data. Both transitions are very sensitive to applied magnetic field — they split into four separate anomalies, which independently shift to lower temperatures with rising field. Several scenarios are proposed as possible explanations of the multi-step character of the magnetic ordering in $CeRh_3Si_2$.

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1. Introduction

The ternary Ce–Rh–Si system is very rich in phases exhibiting wide spectrum of physical phenomena, including magnetic ordering, heavy-fermion behavior and unconventional superconductivity. In particular, $CeRhSi_2$ and $Ce_2Rh_3Si_5$ were found to exhibit properties characteristic of intermediate-valence systems [1, 2], $Ce_3Rh_3Si_2$ was classified in the literature as complex antiferromagnet [3], while $CeRh_2Si_2$ and non-centrosymmetric $CeRhSi_3$ were found to be antiferromagnetically ordered heavy-fermion compounds, which become unconventional superconductors upon applying hydrostatic pressure [4–6].

$CeRh_3Si_2$ is another ternary phase, existence of which was communicated for the first time in Ref. [7]. The authors briefly reported this novel intermetallic as a minor impurity phase in their polycrystalline samples of $CeRh_2Si_2$, crystallizing in an orthorhombic structure of the $ErRh_3Si_2$-type (space group *Imma*, No. 74) and being antiferromagnetically ordered below 5 K. Detailed magnetic properties investigations performed on a single-crystalline specimen of $CeRh_3Si_2$ [8] revealed that the compound orders antiferromagnetically at slightly lower temperature, i.e. $T_N = 4.5$ K, and undergoes another phase transition at $T_t = 4.2$ K. Moreover, the magnetic ordering in $CeRh_3Si_2$ was found to be very sensitive to applied magnetic field, which manifests itself in the occurrence of several successive metamagnetic transitions at fields below 1.5 T and a few field-dependent singularities in the magnetic susceptibility. All the magnetic properties studied exhibit a prominent easy-plane magnetocrystalline anisotropy with the a-axis being the hard magnetic direction.

The complex magnetic properties of single-crystalline $CeRh_3Si_2$ motivated us to undertake studies of the multi-step magnetic phase transition in $CeRh_3Si_2$ by means of other bulk properties measurements, i.e. specific heat and electrical resistivity. In the present paper we report on the results of our low-temperature calorimetric experiments and compare them with the obtained up-to-date magnetic data [8]. The electrical transport properties will be published elsewhere [9].

2. Experimental details

High-quality single crystal of $CeRh_3Si_2$ was grown by the Czochralski pulling method in a tetra-arc furnace under protective Ti-gettered argon atmosphere, from the following starting materials: 3N-Ce pieces (Ames Laboratory), 3N-Rh rod (Chempur) and 6N-Si chips (Chempur). The grown cylindrical ingot had a diameter of about 4 mm and a length of about 40 mm. It was subsequently wrapped in tantalum foil, sealed in an evacuated quartz tube and annealed at 900°C for two weeks.

The product quality was verified by means of X-ray powder diffraction (Stoe powder diffractometer with Cu K_α radiation) and microprobe analysis (Philips 515 scanning electron microscope equipped with an EDAX PV 9800 spectrometer). The crystal structure was examined on a four-circle diffractometer equipped with a CCD camera (Kuma Diffraction KM-4 with graphite-monochromatized Mo K_α radiation). The latter experiments corroborated that the $CeRh_3Si_2$ compound crystallizes with an orthorhombic unit cell of the $ErRh_3Si_2$-type structure (*Imma*, No. 74), with the lattice parameters $a = 7.1121(2)$ Å, $b = 9.6810(5)$ Å and

$c = 5.5828(2)$ Å, being close to those reported in the literature [7].

Heat capacity measurements were carried out using Quantum Design PPMS platform in the temperature range from 1.9 K up to 200 K and in applied magnetic fields up to 2 T, employing a thermal relaxation method [10].

3. Results and discussion

Figure 1 shows the molar heat capacity C of single-crystalline $CeRh_3Si_2$ as a function of temperature. As seen, in the paramagnetic region the $C(T)$ curve has a typical sigmoid (Debye-like) shape. At room temperature the specific heat achieves values close to the Dulong–Petit limit expected for 6 atoms of the formula unit, i.e. 150 J/(mol K). However, at 300 K, $C(T)$ does not saturate at the latter value but shows some tendency to go beyond it, suggesting the presence of some additional contribution to the specific heat, presumably the Schottky anomaly. This finding supports our previous conjecture on strong crystalline-electric-field (CEF) effect in $CeRh_3Si_2$, which was formulated on the basis of the magnetic data (cf. Ref. [8]).

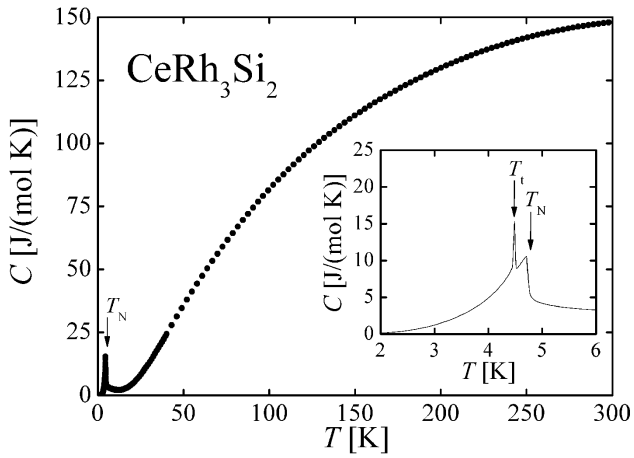

Fig. 1. Temperature variation of the specific heat of single-crystalline $CeRh_3Si_2$ measured in zero magnetic field. The inset shows low-temperature part of the curve. The arrows mark the transition temperatures.

The antiferromagnetic phase transition in $CeRh_3Si_2$ manifests itself in the zero-field temperature dependence of the specific heat as a distinct lambda-shaped anomaly located at $T_N = 4.72(3)$ K (see Fig. 1) that is a value close to the previously reported $T_N = 4.5$ K [8]. Another feature in $C(T)$, in the form of a spike-shaped singularity superimposed on the λ peak, is observed at $T_t = 4.48(2)$ K and clearly corresponds to the spin-reorientation transition revealed from the magnetic data [8].

As can be inferred from Fig. 2, the magnetic ordering in $CeRh_3Si_2$ is very sensitive to applied magnetic field. In 0.5 T both transitions split into as many as four separate anomalies, which independently shift towards lower temperatures with further increase in the magnetic field strength. In a field of 1.5 T all these features merge into a single peak in $C(T)$, which finally disappears above 2 T.

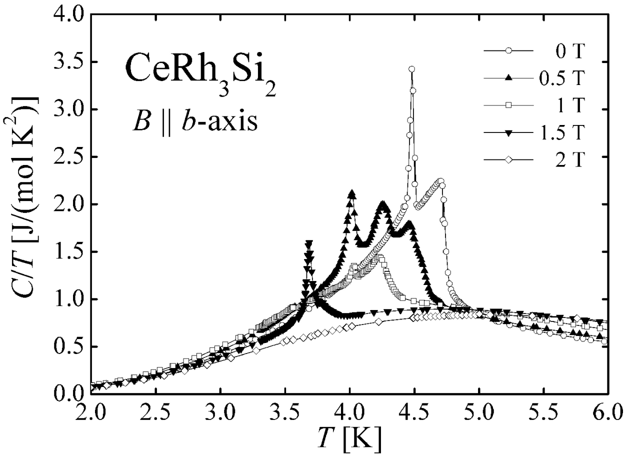

Fig. 2. Low-temperature specific heat of single-crystalline $CeRh_3Si_2$ measured as a function of temperature in several different magnetic fields applied parallel to the easy magnetization b-axis.

The observed complex behavior of the low-temperature specific heat is perfectly in line with those of the magnetic susceptibility and the magnetization, measured along the b-axis as a function of temperature and magnetic field, respectively [8]. Very similar properties have recently been reported for the isostructural compound $CeIr_3Si_2$, and tentatively interpreted in terms of the formation of uncompensated long-period antiphase magnetic structures with the magnetic moments locked into different intermediate directions while the magnetic field strength is varied [11, 12]. Such magnetization processes were found to be common in hexagonal rare-earth-based Laves phases with easy-plane magnetocrystalline anisotropy, like $PrGa_2$ and $NdGa_2$ [13]. Another possible origin of the multistep magnetic behavior of $CeRh_3Si_2$ is a competition between strong CEF effect and relatively weak Ruderman–Kittel–Kasuya–Yosida (RKKY) exchange interactions, associated with an interplay of CEF anisotropy and magnetic frustration (antiferro- vs. ferromagnetic order), as comprehensively discussed for similarly behaved intermetallics, such as $HoNi_2B_2C$ [14], TmAgGe [15] and TmB_4 [16]. Alternatively, one should consider interpretations based on the Ising models, e.g. a model that considers an Ising spin chain immersed in incommensurate exchange field, developed to interpret the unusual magnetic properties of a devil's staircase compound CeSb [17].

4. Summary

The calorimetric studies on $CeRh_3Si_2$ single crystals corroborated the previous magnetic data as regards the antiferromagnetic ordering at $T_N = 4.72(3)$ K and the order–order magnetic phase transition at $T_t = 4.48(2)$ K. Moreover, they confirmed the complex magnetic behavior in the ordered state, which is manifested by the formation of several distinct magnetic structures induced by external magnetic field. The possible interpretations of these findings involve several physical mechanisms, like interplay of crystalline electric field and RKKY interactions, competition between nearest-neighbors and next-nearest-neighbors exchange interactions, etc. In order to build up complete magnetic B–T phase diagrams along the principal crystallographic axes further measurements of single-crystalline $CeRh_3Si_2$ are presently under way. In parallel, some theoretical models are being developed, aimed at the quantitative description of the experimental data. The results of these efforts will be presented in our forthcoming paper [9].

Acknowledgments

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