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Low-Temperature Physical Properties of Single-Crystalline $EuCoGe_3$ and $EuRhGe_3$

O. Bednarchuk and D. Kaczorowski*
Institute of Low Temperature and Structure Research, Polish Academy of Sciences,
P.O. Box 1410, 50-950 Wrocław, Poland

Since a few years europium-based intermetallics have been attracting more and more attention due to their intriguing physical properties with anomalous behaviours in magnetically ordered states. Here, we report on the formation and the bulk physical properties of two tetragonal compounds $EuCoGe_3$ and $EuRhGe_3$, studied on high-quality single-crystalline specimens. In both materials, the Eu ions are in their divalent state, which gives rise to an antiferromagnetic ordering below $T_N = 15.4$ K and $T_N = 11.3$ K, respectively. In addition, $EuCoGe_3$ exhibits a successive antiferromagnetic phase transition at $T_2 = 13.4$ K. Based on some characteristic features in the temperature variations of the magnetic susceptibility, specific heat and electrical resistivity, we suggest that in both germanides an amplitude modulated magnetic structure develops below the respective T_N, with the Eu magnetic moments directed along the crystallographic [001] axis in $EuCoGe_3$ and perpendicular to this direction in $EuRhGe_3$.

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1. Introduction

Rare-earth (R) intermetallics RTX_3 (T is a *d*-electron transition metal and X = Si, Ge) crystallizing with the tetragonal $BaNiSn_3$-type crystal structure continue to attract scientific attention due to their intriguing physical properties. It is enough to recall here a series of comprehensive studies on unconventional superconductivity in $CeTX_3$ compounds [1–6].

Recently, also Eu-bearing ternaries with the $BaNiSn_3$-type unit cells became a subject of investigations. The compound $EuPtSi_3$ was reported to undergo two successive antiferromagnetic (AFM) phase transitions at $T_{N1} = 17$ K and $T_{N2} = 16$ K, which were interpreted as the onset of an incommensurate amplitude modulated (AM) magnetic structure and its reconstruction into a commensurate equal moment (EM) structure, respectively [7]. In contrast, for the isostructural germanide $EuPtGe_3$ a sole phase transition was observed at $T_N = 11$ K, probably to an AFM state of EM character [8]. Similarly, just one AFM transition at $T_N = 12.4$ K was reported for $EuPdGe_3$, yet in this case the magnetic structure was tentatively assigned to be incommensurate [9]. Rather unclear situation occurs for $EuNiGe_3$. In the first literature report [10], based on polycrystalline data, the compound was described as a simple A-type collinear antiferromagnet with $T_N = 13.6$ K. However, in the most recent study on single crystals [11], this germanide was found fairly similar to $EuPtSi_3$, showing a sequence of two AFM phase transitions from an incommensurate AM-type to a commensurate EM-type spin arrangements, occurring at $T_{N1} = 13.2$ K and $T_{N2} = 10.5$ K, respectively.

In this contribution, we report on the low-temperature physical behaviours in the two novel representatives of the $EuTGe_3$ series, namely $EuCoGe_3$ and $EuRhGe_3$.

2. Experimental

High-quality single crystals of $EuCoGe_3$ and $EuRhGe_3$ were obtained by metal-flux method using liquid indium as solvent. The growth process, chemical characterisation and crystal structure determination were described in detail elsewhere [12]. Both compounds were found to crystallize in the body-centred tetragonal $BaNiSn_3$-type structure with space group $I4mm$ and the lattice parameters: $a = 4.3191(3)$ Å, $c = 9.8847(15)$ Å for $EuCoGe_3$, and $a = 4.4129(3)$ Å, $c = 10.0906(15)$ Å for $EuRhGe_3$. The difference in the unit cell dimensions correlates with the atomic radii of Co and Rh.

Magnetic measurements were carried out in the temperature range 1.75–400 K in magnetic fields up to 7 T using a Quantum Design MPMS SQUID magnetometer. The heat capacity and the electrical resistivity were studied over the temperature interval 0.35–25 K using a Quantum Design PPMS platform employing relaxation-time method and ac four-point technique, respectively. In the transport measurements, current and voltage leads were 50 μm thick silver wires attached to the parallelepiped-shaped specimens by means of tin soldering and spot welding, respectively.

3. Results and discussion

From the magnetic susceptibility data [12], $EuCoGe_3$ and $EuRhGe_3$ were found to order antiferromagnetically

*corresponding author; e-mail: D.Kaczorowski@int.pan.wroc.pl

below $T_N = 15.4$ K and $T_N = 11.3$ K, respectively. For $EuCoGe_3$, an extra feature was noticed at $T_2 = 13.4$ K, in the form of a kink in the magnetic susceptibility measured as a function of temperature with magnetic field oriented within the tetragonal *ab*-plane, while no corresponding anomaly in the magnetic susceptibility was observed along the *c*-axis. In the paramagnetic region, both materials exhibit the Curie–Weiss behavior with the effective magnetic moments close to that of divalent Eu ions (7.9 μ_B) and large negative paramagnetic Curie temperatures, consistent with the AFM orderings (for details see Ref. [12]).

Figure 1 displays the low-temperature variations of the specific heat over temperature ratio, C/T, of $EuCoGe_3$ and $EuRhGe_3$. For the former compound, two clear jumps in C/T are seen at $T_N = 15.4$ K and $T_2 = 13.4$ K, fully consistent with the magnetic data. Similarly, for $EuRhGe_3$ the rapid jump in $C/T(T)$ at $T_N = 11.3$ K corroborates the onset of AFM state. For both germanides, the observed change in the specific heat at the respective T_N amounts to about 12 J/(mol K) that is close to the value of 13.4 J/(mol K) theoretically predicted within the molecular field approach for AM antiferromagnets [13].

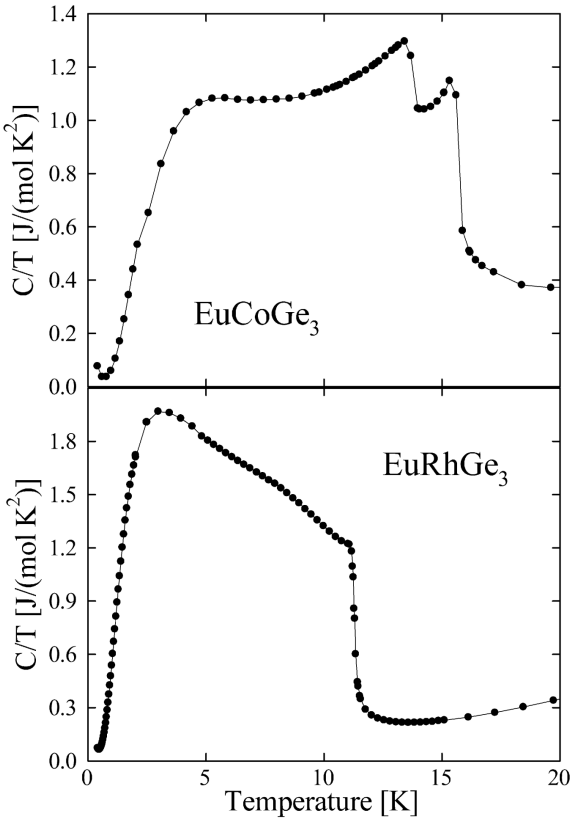

Fig. 1. Low-temperature dependences of the specific heat over temperature ratio of single-crystalline $EuCoGe_3$ and $EuRhGe_3$.

Deeply in the ordered state, roughly below 5 K, the $C/T(T)$ variations of $EuCoGe_3$ and $EuRhGe_3$ exhibit broad maxima, characteristic of Schottky-like contribution due to thermal population of the energy levels that stem from the $^8S_{7/2}$ multiplet of Eu^{2+} ions split in internal magnetic exchange field. Similar feature was observed also for $EuPtSi_3$ [6] and other $EuTGe_3$ phases [7–12], however it is worth noting that the magnitude of this anomaly in the specific heat data of $EuRhGe_3$ is unusually large. At the lowest temperature studied, a tiny upturn in $C/T(T)$ of both compounds is observed that likely arises due to nuclear contribution to the measured heat capacity.

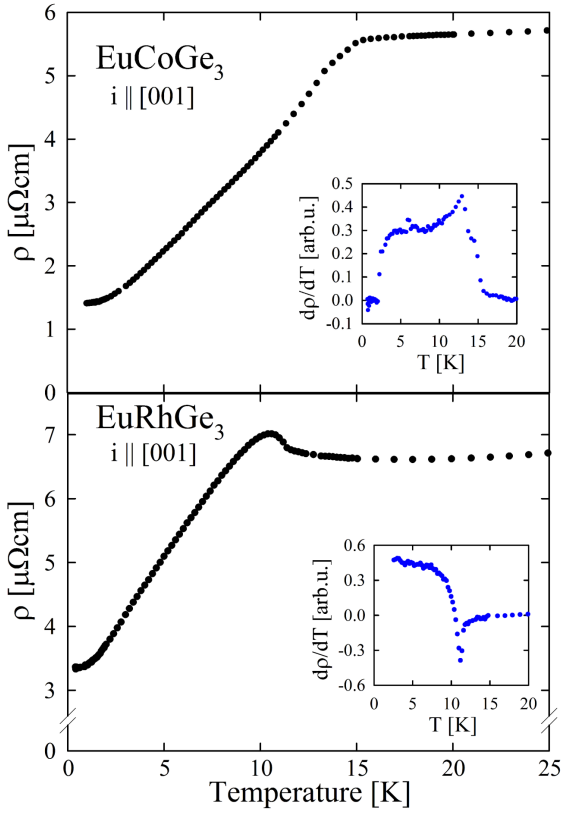

Fig. 2. Low-temperature variations of the electrical resistivity of $EuCoGe_3$ and $EuRhGe_3$ single crystals, measured with electrical current flowing along the crystallographic *c*-axis. The insets show the temperature derivatives of the *c*-axis electrical resistivity of both compounds.

The low-temperature dependencies of the electrical resistivity, ρ, of single-crystalline $EuCoGe_3$ and $EuRhGe_3$, measured with electric current flowing along the *c*-direction in the crystallographic unit cell, are shown in Fig. 2. Both materials exhibit metallic character with a fairly small residual resistivity that proves good quality of the single crystals investigated. The AFM phase transition in $EuCoGe_3$ manifests itself as a clear kink in $\rho(T)$, below which the resistivity rapidly diminishes with decreasing temperature because of gradual reduction in the spin-disorder scattering contribution. The spin reorientation at T_2 brings about a minor change in the slope of

$\rho(T)$ but it can be clearly discerned in the temperature derivative of the resistivity (see the inset).

As apparent from Fig. 2, close to the onset of the AFM state, the electrical resistivity of $EuRhGe_3$ behaves in a distinctly different manner from that of $EuCoGe_3$. Here, $\rho(T)$ forms a broad maximum below $T_N = 11.3$ K, and the $d\rho/dT$ derivative exhibits a sharp drop at T_N. These features signal the opening of an energy gap on the Fermi surface, induced by the AFM ordering. The magnetic unit cell of $EuRhGe_3$ is probably enlarged in relation to the chemical one in the crystallographic c-direction, which implies extra scattering of conduction electrons in the ordered state on "magnetic" Brillouin zone boundaries. This hypothesis must be verified by means of direct determination of the magnetic structure of $EuRhGe_3$ in neutron diffraction experiment.

4. Conclusions

The compounds $EuCoGe_3$ and $EuRhGe_3$ crystallize in the tetragonal body-centered $BaNiSn_3$-type structure. They order antiferromagnetically below $T_N = 15.4$ K and $T_N = 11.3$ K, respectively. In addition, $EuCoGe_3$ exhibits another magnetic phase transition at $T_2 = 13.4$ K that likely has a spin-reorientation character. Based on some characteristic features revealed in the temperature dependences of the magnetic susceptibility [12], specific heat and electrical resistivity it seems likely that both germanides order with an amplitude modulated magnetic structure. In $EuCoGe_3$ the magnetic moments are probably aligned mostly along the crystallographic c-axis, while in $EuRhGe_3$ they are almost confined within the tetragonal ab-plane. Neutron diffraction study is planned to verify these conjectures.

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