14th Czech and Slovak Conference on Magnetism, Košice, Slovakia, July 6–9, 2010

Anomalous Transport Properties of Carbon-Doped EuB₆

M. BATKOVA^{*a*}, I. BATKO^{*a*}, V. FILIPOV^{*b*}, K. FLACHBART^{*a*}, V. SECHOVSKÝ^{*c*}, N. SHITSEVALOVA^{*b*}, E. ŠANTAVÁ^{*d*} AND J. ŠEBEK^{*d*}

^aInstitute of Experimental Physics, Slovak Academy of Sciences, Watsonova 47, 040 01 Košice, Slovak Republic

^bInstitute for Problems of Material Science, NASU, 252680 Kiev, Ukraine

^cCharles University in Prague, Faculty of Mathematics and Physics, Ke Karlovu 5, 121 16 Prague, Czech Republic ^dInstitute of Physics, Academy of Sciences of the Czech Republic, Na Slovance 2, 182 21 Prague, Czech Republic

In the presented work we report electrical, magnetic and thermal properties of $\operatorname{EuB}_{6-x}C_x$ single crystals with an estimated value of $x \approx 0.07$. Our studies reveal an antiferromagnetic phase transition at $T_{\rm N} \approx 6.7$ K. Electrical resistivity at zero magnetic field shows a pronounced resistivity maximum at $T_{\rm M} \approx 7$ K, just above the antiferromagnetic phase transition temperature. With increasing applied magnetic field the maximum moves to lower temperature and becomes totally suppressed at the field of 9 T. Observed magnetoresistance is negative in the whole studied temperature range 2–20 K, yielding a ratio of $\rho(0 \text{ T}, 7 \text{ K})/\rho(9 \text{ T}, 7 \text{ K}) \approx 2.5$. The origin of such magnetoresistance is associated with formation of mixed magnetic structure in the system due to fluctuation of carbon concentration.

PACS numbers: 75.30.Kz, 72.15.Gd, 75.47.Gk

1. Introduction

 $\operatorname{EuB}_{6-x} C_x$ carbide borides behave as degenerate semiconductors, in which both carrier concentration and antiferromagnetic interaction increase with increasing carbon content [1]. While the pure EuB_6 behaves like a simple ferromagnet, $\operatorname{EuB}_{5.80}C_{0.20}$ has an incommensurate spiral structure [1, 2]. The magnetic structure of intermediate $\operatorname{EuB}_{5.95}C_{0.05}$ is described as a mixture of ferromagnetic and helimagnetic domains [1, 2], whereas the latter ones are a consequence of local increase of carbon concentration in the material.

Not long ago we dealt with the ferromagnetic $EuB_{5.99}C_{0.01}$ [3, 4] exhibiting anomalously large negative magnetoresistance. Our studies have revealed that the behaviour of this system can be attributed to the effect of fluctuations in carbon concentration. According to our observations, in the bulk ferromagnetic state, carbon--rich regions give rise to helimagnetic domains that are responsible for an additional scattering term in the electrical resistivity [3–5]. Above the temperature of the bulk ferromagnetic ordering, $T_{\rm C} = 4.3$ K, the carbon-rich regions act as spacers that prevent magnetic polarons to link, to form ferromagnetic clusters, and eventually to percolate. These spacers, being in fact volumes incompatible with the existence of magnetic polarons (and ferromagnetic state in general) are responsible for decrease in the percolation temperature and for additional (magneto)resistivity increase [4].

The main goal of this paper is to bring information on electrical, magnetic and thermal properties of carbon-doped EuB₆ from the antiferromagnetic region of the phase diagram. Namely, we studied EuB_{6-x}C_x single crystals prepared by zone-floating method with an estimated value of $x \approx 0.07$.

2. Experimental

Magnetic susceptibility of the sample was measured in the temperature range 2–300 K using a commercial SQUID magnetometer (MPMS from Quantum Design). As can be seen in the inset of Fig. 1, the real part of magnetic susceptibility, $\chi'(T)$, obeys the Curie–Weiss law in the temperature range 10–100 K. A fitted value of the paramagnetic Curie temperature, $\Theta_{\rm p} = -4$ K, confirms the antiferromagnetic ground state of the sample. Above 100 K there is a deviation from the behaviour described by temperature independent Curie constant which we associate with the mixed magnetic structure of the sample.

3. Results and discussion

Temperature dependence of the heat capacity, $C_p(T)$, in zero magnetic field was measured in the temperature range 2–40 K using a commercial Physical Property Measurement System (PPMS) from Quantum Design. The data are shown in Fig. 1. Taking into account the negative value of Θ_p , a pronounced λ -anomaly with a maximum at 6.7 K refers to the Néel temperature $T_{\rm N} = 6.7$ K.

Electrical resistivity measurements were performed in a commercial PPMS in the temperature range between 2 K and 20 K. Electric current was applied along the [110] direction of the sample and magnetic field was applied perpendicularly to the direction of the current flow. Obtained low temperature parts of $\rho(T)$ dependences for distinct magnetic fields between 0 T and 9 T are depicted in Fig. 2. As can be seen from the figure, below 20 K the electrical resistivity at zero magnetic field first increases steeply with decreasing temperature, then passes a maximum at $T_{\rm M} \approx 7$ K (nearby $T_{\rm N} = 6.7$ K),



Fig. 1. Temperature dependence of the heat capacity of $EuB_{5.93}C_{0.07}$ at zero magnetic field. Inset shows the real part of the magnetic susceptibility.



Fig. 2. Temperature dependences of electrical resistivity of $EuB_{5.93}C_{0.07}$ at distinct magnetic fields up to 9 T.

and subsequently decreases with further temperature decrease. The maximum of the $\rho(T)$ behaviour moves to lower temperature and becomes less pronounced with increasing magnetic field. Magnetic field of 9 T suppresses the maximum totally, indicating its magnetic origin. Magnetoresistance, MR = $[\rho(B) - \rho(0)]/\rho(0)$, of the system is negative in the whole studied temperature range, reaching the maximum absolute value of 0.59 for 9 T coinciding with the maximum in $\rho(T)$ at zero magnetic field. At the temperature of the resistivity maximum at zero magnetic field the data give the ratio of $\rho(0 \text{ T}, 7 \text{ K})/\rho(9 \text{ T}, 7 \text{ K}) \approx 2.5$. Although the magnetoresistance of the studied system is still relatively high. it is significantly less comparing to $EuB_{5.99}C_{0.01}$, where the analogous ratio $\rho(0 \text{ T}, 5 \text{ K})/\rho(9 \text{ T}, 5 \text{ K}) \approx 6$. We explain the observed properties as follows.

As mentioned above, the magnetic structure of $EuB_{5.95}C_{0.05}$ can be described as a mixture of ferromagnetic and helimagnetic domains [1, 2], and similar structure is expected in $EuB_{5.99}C_{0.01}$, too [4]. In systems with higher carbon content one can expect existence of a phase, in which helimagnetic domains (with less carbon content) coexist with antiferromagnetic ones (having higher content of carbon). We consider just such a state to be present in investigated here $EuB_{5,93}C_{0,07}$. The observed negative magnetoresistance can be associated with scattering of conduction electrons on the mixed magnetic structure. On the other hand, as it is well known for the stoichiometric EuB_6 , the magnetoresistance phenomena close above the temperature of ferromagnetic ordering are governed by magnetic polarons. Also in $EuB_{5.99}C_{0.01}$ magnetic polarons are believed to be present and have important impact on the enhancement of magnetoresistivity [4]. However, in the investigated here more doped system, both too high electron concentration and antiferromagnetism exclude a formation of magnetic polarons. The absence of magnetic polarons might be one of reasons for relatively less magnetoresistance of investigated here $EuB_{5.93}C_{0.07}$ in comparison with less doped $EuB_{5.99}C_{0.01}$. Nevertheless, for a solid confirmation of the proposed interpretation additional experimental studies and more detailed and complex analysis of experimental data are needed.

Acknowledgments

This work was supported by the Slovak Scientific Agency (project VEGA 2-0133/09), by the Slovak Research and Development Agency (APVV-0346-07) and by Acad. Sci. of Czech Republic (research project AVOZ10100520). Work of V.S. belongs to the research plan MSM0021620834 financed by the Ministry of Education of the Czech Republic. M.B. acknowledges the support of the European Science Foundation (COST-STSM-P16-03226).

References

- J. Etorneau, P. Hagenmuller, *Philos. Mag. B* 52, 589 (1985).
- [2] J.M. Tarascon, J.L. Soubeyroux, J. Etorneau, R. Georges, J.M. Coey, O. Massenet, *Solid State Commun.* 37, 133 (1981).
- [3] I. Batko, M. Batkova, K. Flachbart, D. Macko, E.S. Konovalova, Yu.B. Paderno, J. Magn. Magn. Mater. 140-144, 1177 (1995).
- [4] M. Batkova, I. Batko, K. Flachbart, Z. Janu, K. Jurek, J. Kováč, M. Reiffers, V. Sechovský, N. Shitsevalova, E. Šantavá, J. Šebek, *Phys. Rev. B* 78, 224414 (2008).
- [5] M. Batkova, I. Batko, E. Bauer, R.T. Khan, V.B. Filipov, E.S. Konovalova, *Solid State Commun.* 150, 652 (2010).