Anomalous magnetoresistance of EuB$_{5.99}$C$_{0.01}$: Enhancement of magnetoresistance in systems with magnetic polarons

M. Baťková,¹ I. Baťko,¹ K. Flachbart,¹ K. Jurek,² E. S. Konovalova,³ J. Kováč,¹ M. Reiffers,¹ V. Sechovský,⁴ N. Shitsevalova,³ E. Šantavá,² and J. Šebek²

¹Institute of Experimental Physics, Slovak Academy of Sciences, Watsonova 47, 04001 Košice, Slovakia
²Institute of Physics, Academy of Sciences of the Czech Republic, Na Slovance 2, 182 21 Prague, Czech Republic
³Institute for Problems of Material Science, NASU, 252680 Kiev, Ukraine
⁴Faculty of Mathematics and Physics, Charles University, Ke Karlovu 5, 121 16 Praha 2, Czech Republic

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We present results of measurements of electrical, magnetic and thermal properties of EuB$_{5.99}$C$_{0.01}$. The observed anomalously large negative magnetoresistance as above, so below the Curie temperature of ferromagnetic ordering $T_C$ is attributed to fluctuations in carbon concentration. Below $T_C$ the carbon rich regions give rise to helimagnetic domains, which are responsible for an additional scattering term in the resistivity, which can be suppressed by a magnetic field. Above $T_C$ these regions prevent the process of percolation of magnetic polarons (MPs), acting as “spacers” between MPs. We propose that such “spacers”, being in fact volumes incompatible with existence of MPs, may be responsible for the decrease of the percolation temperature and for the additional (magneto)resistance increase in systems with MPs.

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EuB$_6$ is a rare example of a low carrier density hexaboride that orders ferromagnetically at low temperatures and undergoes a metal-insulator phase transition. The ferromagnetic order is established via two consecutive phase transitions at $T_M = 15.5$ K and $T_C = 12.6$ K, respectively. The high-temperature magnetoresistance is large and negative; the absolute value is increasing with decreasing temperature, and reaches the maximum value of about 100% at the magnetic ordering temperature (15.6 K). In the ferromagnetic regime, the magnetoresistance is positive and reaches a value up to 700% at 7 T and 1.7 K.

Physical properties of EuB$_6$ are thought to be governed by magnetic polarons (MPs), which are in fact carriers localized in ferromagnetic clusters embedded in a paramagnetic matrix. As suggested by Stillow et al., the magnetic phase transition at $T_M$ represents the emergence of the spontaneous magnetization accompanied by metalization. At this temperature the MPs begin to overlap and form a conducting, ferromagnetically ordered phase that acts as a percolating, low-resistance path across the otherwise poorly conducting sample. With decreasing temperature, the volume fraction of the conducting ferromagnetic phase expands, until the sample becomes a homogeneous conducting bulk ferromagnet at $T_C$. As indicated by Raman scattering measurements, the polarons appear in EuB$_6$ at about 30 K.

Because of the very low number of intrinsic charge carriers ($\sim 10^{20}$ cm$^{-3}$), even a slight change of the concentration of conduction electrons (e.g., due to a change of chemical composition or number of impurities) can drastically modify the electric and magnetic properties of EuB$_6$. Substitution of B by C enhances the charge carrier concentration in EuB$_6$. As shown by neutron diffraction studies, the predominant ferromagnetic ordering in the stoichiometric EuB$_6$ changes with increasing carbon content through a mixture of the ferromagnetic phase and helimagnetic domains into a purely antiferromagnetic state. The paramagnetic Curie temperature $\theta_p$ of EuB$_{6-x}$C$_x$ changes its sign for $x = 0.125$. The helimagnetic domains are associated with carbon richer regions (with higher carrier density) due to local fluctuations of the carbon concentration. Different impact of the RKKY interaction in carbon richer and carbon poorer regions yields to different types of magnetic order.

The unusual transport properties of carbon doped EuB$_6$ single crystal were reported more than a decade ago. The results have shown that the electrical resistance becomes strongly enhanced below 15 K and exhibits a maximum around 5 K. The residual resistivity is exceptionally high; it is even higher than the room temperature resistivity $\rho(300 \text{ K})$. Application of a magnetic field of 3 T at 4.2 K causes a dramatic reduction of the resistivity yielding $\rho(0 \text{ T})/\rho(3 \text{ T}) = 3.7$. The huge residual resistivity has been ascribed to the scattering of conduction electrons on boundaries between the ferromagnetic and helimagnetic regions.

In this paper we present an extended study of the electrical resistivity, magnetoresistance, susceptibility and heat capacity on a EuB$_{5.99}$C$_{0.01}$ single crystal. We bring further experimental results supporting the afore mentioned hypothesis of the dominant scattering process at temperatures below $T_C$ originating from the mixed magnetic structure. In addition, our results adverst that above $T_C$ the electrical transport is governed by MPs and can be well understood within a recently proposed scenario involving the “isolated”, “linked” and “merged” MPs. Moreover, we argue that regions of proper size and space...
distribution, incompatible with existence of MPs can be the clue for understanding the origin of the colossal magnetoresistance in systems with MPs.

Samples used for magnetization and resistivity measurements were cut from the crystal used in previous studies, which has been grown by means of the zone-floating. Recent micro-probe analysis of this crystal revealed the carbon content corresponding to the stoichiometric formula EuB$_{5.99}$C$_{0.01}$. The electrical resistance, magnetoresistance, heat capacity and ac-susceptibility were measured in the Quantum Design PPMS and MPMS. The direction of the applied magnetic field was perpendicular to electrical current in all magnetoresistance measurements.

The electrical resistivity of EuB$_{5.99}$C$_{0.01}$ decreases upon cooling from 300 K until it reaches a shallow minimum at about 40 K. Below 10 K it increases steeply, passes a maximum at $T_{RM}$ ~5 K, and subsequently falls off having tendency to saturate at lowest temperatures. The low-temperature part of its dependence is shown in Fig. 1 as curve a). The temperature derivative of the resistivity in zero magnetic field, depicted in the figure inset, shows a sharp maximum at $T_m$ = 4.1 K indicating a proximity of magnetic phase-transition. Since the optical reflectivity data of the studied system have not revealed any shift in the plasma frequency between 4.2 and 20 K, the charge carrier concentration can be regarded as constant in this temperature interval. Therefore, we tentatively associate the anomalous resistivity behavior with magnetism in this material.

In Fig. 2 we plot temperature dependences of magnetoresistance $MR = [\rho(B) - \rho(0)]/\rho(0)$ for selected values of the applied magnetic field between 50 mT and 12 T, derived from the data shown in Fig. 1. The absolute value of magnetoresistance reaches a maximum (of about 0.83 for 12 T) in the vicinity of $T_{RM}$. Upon further cooling $MR$ decreases continuously in absolute value, and in difference to EuB$_6$, only in the smallest magnetic fields up to 0.3 T, and at the lowest temperatures, it passes through zero and reaches positive values.

We suppose that below $T_C$, the scattering of conduction electrons originates from phase boundaries of the mixed magnetic structure consisting of helimagnetic domains, associated with carbon richer regions, in the ferromagnetic matrix. Sufficiently high magnetic field makes the helimagnetic domains energetically unfavorable and therefore reduces their volumes (and probably destroys them completely at highest fields), giving rise to negative magnetoresistance.

The magnetic field influence on the resistivity and magnetoresistance behavior between 2 and 20 K and the magnetic field dependences of resistivity depicted in Fig. 1 and respectively, reveal two different magnetoresistance regimes: (i) for temperatures lower than $T_{RM}$ - the resistivity is enhanced by small fields ($B \leq 0.3$ T) and reduced by higher fields ($B \geq 0.5$ T); (ii) above $T_{RM}$ - the resistivity monotonically decreases with increasing applied magnetic field.

The low-field magnetoresistance measured at 2 K is dependent on magnetic history and exhibits large hysteresis. Fig. 3 shows the hysteresis behavior of the resistivity, including the virgin curve taken at 2 K after cooling from 30 K to 2 K in zero magnetic field. As it is visible in the figure, the hysteresis is significant for $|B| \lesssim 0.3$ T. The hysteresis of magnetisation is very weak, but not negligible in the interval where the resistivity hysteresis is observed, suggesting that the positive magnetoresistance in low magnetic fields is due to the conduction-electron scattering on the domain walls within the ferromagnetic matrix.
With the aim to get more information on the magnetic properties and the phase transition(s), we measured the real part of the temperature dependence of the ac-susceptibility $\chi(T)$ and the specific heat $C(T)$ in the temperature range 2 - 86 K and 2 - 30 K, respectively. The $1/\chi(T)$ satisfies the Curie-Weiss law in the region above $\sim 29$ K and yields the paramagnetic Curie temperature $\theta_p = 7$ K. Fig. 2 shows the $\chi(T)$ and $C(T)$ data below 10 K. The $\chi(T)$ dependence indicates two distinct regimes, one above and other below $\sim 4$ K, such as it obeys almost linear behavior, in the intervals 2 - 3.6 K and 4.1 - 4.8 K, respectively, however with different slopes. The specific heat exhibits a broad peak at 5.7 K, which we tentatively associate with the magnetic ordering transition at $T_C \sim 5.7$ K. The position of the peak correlates well with the position of the inflexion point of the $\chi(T)$ dependence (5.5 K). There is also a side anomaly at 4.3 K in the $C(T)$ dependence, which almost coincides with the afore mentioned resistivity anomaly at $T_m = 4.1$ K and with the change of the regime of the $\chi(T)$ dependence. Detailed microscopic investigation (e.g. neutron diffraction) is desired to elucidate the relation between the specific-heat and resistivity anomalies with magnetic phenomena in the studied material.

The observed behavior of EuB$_{5.99}$C$_{0.01}$ can be consistently explained within the framework of results obtained by Yu and Min, who investigated the magnetic phase transitions in MP systems using the Monte Carlo method. They supposed three consecutive temperature scales: $T^*$, $T_C$ and $T_p$. Upon cooling from the high-temperature paramagnetic state the isolated MPs with random magnetization directions begin to form at $T^*$. At further cooling the MPs grow in size. Down to $T_C$ carriers are still confined to MPs, thus the metallic and magnetic regions are separated from the insulating and paramagnetic regions. The isolated MPs become linked at the bulk ferromagnetic transition temperature $T_C$. Eventually, the polaron percolation occurs expressing itself as a peak in the heat capacity at $T_p < T_C$. Below $T_p$ all carriers are fully delocalized and the concept of MPs becomes meaningless. The other issue, which should be mentioned, is that the impurities reduce both, $T_C$ and $T_p$, but the discrepancy ratio $(T_p/T_C = 7/9 \approx 0.77)$ between these two temperatures is retained.

According to the concept of Yu and Min we interpret the obtained experimental results as follows. Consistently with the temperature dependence of the magnetization, EuB$_{5.99}$C$_{0.01}$ is paramagnetic above $\sim 29$ K. We expect the formation of isolated MPs at lower temperatures. The magnetic phase transition temperature reflected in the broad maximum of the $C(T)$ dependence, we associate with the temperature of ferromagnetic ordering $T_C$. We suggest that the isolated MPs begin to link at $T_C$. The MPs become merged and percolation occurs at the temperature of the (side) specific-heat
the role of non-ferromagnetic “spacers” in the magnetoresistance of EuB$_{6-x}$C$_x$. W e suppose that these regions are responsible for the higher value of the resistivity maximum at correspondingly lower temperatures, and consequently, for the larger magnetoresistance. Finally, we emphasize that introducing such “spacers”, which prevent the percolation of MPs may strongly enhance the magnetoresistance of systems with transport governed by MPs. The “spacers” are in fact regions of appropriate size and scale distribution, which are not compatible with ferromagnetic ordering. This might show a route for future research efforts in relation with the colossal magnetoresistance effect.

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