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Reentrant Metal–Insulator Transition in $Ca_{1-x}Eu_xB_6$

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Resistivity, Hall and Seebeck effects have been studied on single crystals of $Ca_{1-x}Eu_xB_6$ ($0 \le x \le 1$) at temperatures 2–300 K and in magnetic fields up to 8 T. An insulating ground state is found to be limited by narrow range of Eu doping $0.6 \le x \le 0.8$. This region is characterized by an enhanced colossal magnetoresistance (CMR), which reaches values of $\rho(0)/\rho(7\ T) > 10^6$ for $x = 0.63$ at $T < 10$ K. Decreasing of Eu content in $Ca_{1-x}Eu_xB_6$ below $x^* \approx 0.6$ restores the metallic ground state with moderate resistivity ($\rho \sim 1 \div 5$ mΩ·cm) and CMR amplitude ($\rho(0)/\rho(7\ \mathrm{T}) < 7$). The second metal-insulator transition (MIT) in $Ca_{1-x}Eu_xB_6$ is observed beyond the whole conductivity region found earlier in the narrow range of Eu doping ($0.7 \le x \le 0.8$). The correlation between the enhanced CMR amplitude, the onset of positive diffusive thermopower and the elevation of anomalous Hall effect, determined for Eu content $0.6 \le x \le 0.85$, favors the idea that a smooth change of band structure is the main factor governing the reentrant MIT in $Ca_{1-x}Eu_xB_6$.

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1. Introduction

The onset of insulating ground state, appearing due to strong interaction between itinerant electrons and localized magnetic moments in $Ca_{1-x}Eu_xB_6$ [1–3], provides an interesting opportunity for identifying the microscopic mechanisms of colossal magnetoresistance (CMR) in these rare earth hexaborides. The scenario of magnetic polaron percolation, which is used to explain the temperature driven metal-insulator transition (MIT) in EuB_6 [4] and the exponential decrease of resistivity under applied magnetic field in $Ca_{0.4}Eu_{0.6}B_6$ [5], fails to describe the onset of hole-like conductivity and huge CMR enhancement under transition into the insulating ground state in Eu-rich solid solutions ($x < x_{MIT} \approx 0.8$) [3]. In this respect the study of charge transport in $Ca_{1-x}Eu_xB_6$ ($x < x_{MIT}$) is of great importance for understanding the nature of CMR in this strongly correlated electron system.

2. Results and discussion

To shed light on the origin of CMR charge transport in $Ca_{1-x}Eu_xB_6$ ($0 \le x \le 1$) single crystals were studied at temperatures 2–300 K in magnetic fields up to 8 T. The samples have been grown by the crucible-less inductive zone melting in argon atmosphere. SEM analysis showed that these solid solutions are homogeneous within ±0.5 at.% of Eu ($\Delta x \sim 0.005$). The experimental setups for measurement of the transport parameters have been earlier described in [3].

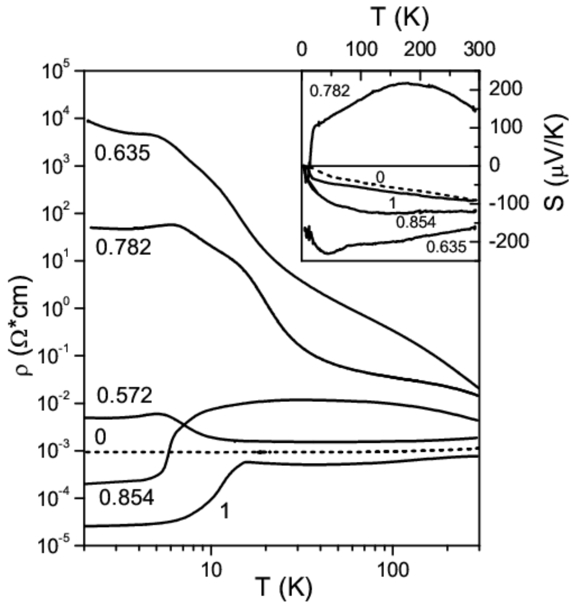

Fig. 1. Temperature dependences of resistivity (main panel) and Seebeck coefficient (inset) for $Ca_{1-x}Eu_xB_6$ ($0 \le x \le 1$). The data for CaB_6 are shown by dash lines.

Temperature dependences of resistivity in $Ca_{1-x}Eu_xB_6$ (Fig. 1) show that at low temperatures ρ increases under Eu doping, resulting in an onset of ground state with high resistivity ($\rho > 10\ \Omega$·cm, Fig. 1). This insulating state, appearing in the limited range of Eu content ($0.6 \le x \le 0.8$), is very

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sensitive to applied magnetic field, which induces the huge decrease of resistivity. The maximal values of $\rho(0)/\rho(7\ \mathrm{T}) \approx 2.4 \times 10^7$, measured for $x = 0.635$ at $T = 2$ K, seem to be the record ones for this class of materials. The increase of Ca content restores the metallic ground state with moderate values of resistivity ($\rho \sim 1 \div 5$ mΩ·cm) and CMR effect ($\rho(0)/\rho(7\ \mathrm{T}) < 7$) comparable with that of $\rho(0)/\rho(7\ \mathrm{T}) \sim 11$ found for EuB_6.

The data of Hall resistivity show that positive Hall effect is detected only in the paramagnetic phase in a narrow range of Eu doping ($0.7 < x < 0.8$), changing to the negative one in moderate magnetic field [3]. A standard procedure was applied to extract normal (R_H) and anomalous (R_H^A) Hall coefficients using the magnetic data of Ref. 7. For $x > 0.85$ and $x < 0.6$ the normal contribution to Hall effect dominates over the anomalous one and becomes 3 times as large, when moving to the Ca-rich region (Fig. 2c). On the contrary, the absolute values of R_H^A rise up to $|R_H^A(x = 0.635)| \approx 0.2$ cm^3/C upon entering the insulating region of concentration driven MIT. Hall mobility of charge carriers in the ferromagnetic phase doesn't depend on the doping level in the range $0.6 < x < 0.95$: $\mu_H = -(210 \pm 40)$ cm^2/(V·s).

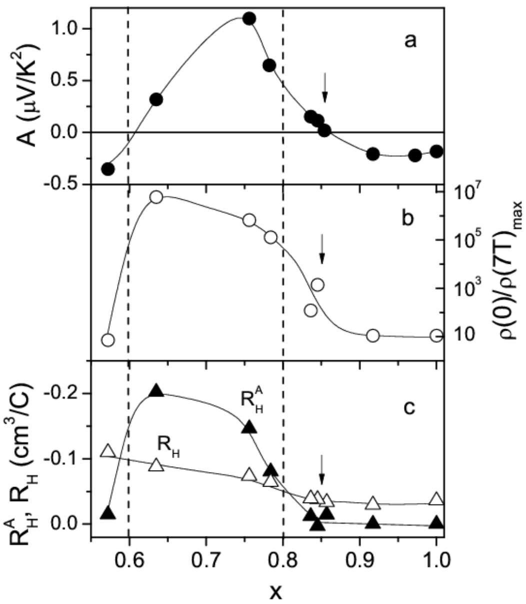

Fig. 2. Eu concentration dependence of the coefficient of diffusive thermopower A (a), the maximal value of CMR amplitude $\rho(0)/\rho(7\ \mathrm{T})$ (b) and the normal Hall coefficient R_H compared to anomalous one R_H^A at 4.2 K (c) for Eu-rich $Ca_{1-x}Eu_xB_6$ compounds. Solid lines are guides-to-eyes.

To understand the nature of anomalous galvanomagnetic properties (Fig. 2b, c) we analyzed the data of Seebeck effect, which revealed a qualitative change of thermoelectric properties at Eu content $x > 0.6$ (inset in Fig. 1). Actually, Seebeck coefficient for $x = 0.635$ compound falls down to $S = -(160 \div 220)$ μV/K and changes the sign to positive values $S > 0$ for $0.7 < x < 0.8$ (inset in Fig. 1). At the same time, a positive slope of $S(T) \sim AT$, corresponding to diffusive thermopower in $Ca_{1-x}Eu_xB_6$ [6], is observed in a wider range of concentrations $0.6 < x < 0.85$ (Fig. 2a). In our opinion, the different signs of Hall resistivity and diffusive thermopower point to a nontrivial fine structure of the density of states at the Fermi level on insulating sides of concentration driven MIT in $Ca_{1-x}Eu_xB_6$, while the correlated behavior of galvanomagnetic properties and diffusive thermopower found at $0.6 < x < 0.85$ favors a common nature of charge transport anomalies related to a smooth change of the $Ca_{1-x}Eu_xB_6$ band structure under Eu doping.

3. Conclusion

A combined study of resistivity, Hall and Seebeck effects, carried out on $Ca_{1-x}Eu_xB_6$ single crystals in magnetic fields up to 8 T, allows to identify the range of Eu doping ($0.6 \le x \le 0.8$) corresponding to the onset of insulating ground state with a large amplitude of CMR effect ($\rho(0)/\rho(7\ \mathrm{T}) > 10^5$). The established correlation between the enhancement of CMR effect, the onset of positive diffusive thermopower and the elevation of anomalous Hall effect in the range $0.6 \le x \le 0.85$ favors idea that a smooth change of band structure is a main factor governing this reentrant MIT in $Ca_{1-x}Eu_xB_6$.

Acknowledgments

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