Proceedings of the CSMAG'07 Conference, Košice, July 9-12, 2007

Magnetotransport Properties of $CaTi_x Ru_{1-x}O_3$ (x = 0, 0.07)

A. ZORKOVSKÁ, A. BARAN, A. FEHER

Centre of Low Temperature Physics of the P.J. Šafárik University and the Slovak Academy of Science, Park Angelinum 9, 04154 Košice, Slovakia

J. Šebek, E. Šantavá

Institute of Physics, AS CR, Na Slovance 2, 18221 Prague, Czech Republic

and I. Bradarić

The "Vinča" Institute of Nuclear Sciences, P.O. Box 522, 11001 Belgrade, Serbia

Electrical resistance, transversal magnetoresistance and the Hall effect were studied on polycrystalline $\operatorname{CaTi}_x \operatorname{Ru}_{1-x} O_3$ (x = 0, 0.07) samples using a conventional Quantum Design PPMS-9 equipment in the temperature range 2–300 K and magnetic field up to 9 T. Substantial differences were found between the two samples: (i) opposite to the metallic character of CaRuO₃, the substituted sample has insulating-like electrical resistance; (ii) the magnetoresistance of the substituted sample changes the sign from negative to positive values with increasing temperature. The magnetoresistance of CaRuO₃ is negative, the sign reversal is induced by magnetic field and only at temperatures below 15 K, such a behaviour is predicted for clustered systems; (iii) the Hall voltage in pure CaRuO₃ also changes sign from negative to positive values above 35 K. This temperature coincides with the observed magnetic transition temperature, indicating that the magnetic state and the carrier character interrelate.

PACS numbers: 74.70.Pq, 73.43.Qt, 75.47.-m

1. Introduction

Metallic perovskites, specifically, the ruthenates of the Ruddlesden–Popper series $(Sr,Ca)_{n+1}Ru_nO_{3n+1}$ are in the focus of interest because of the wide range of electronic and magnetic properties they exhibit: from the Mott metal–insulator transition to unconventional anisotropic superconductivity, from paramagnetism through complicated metamagnetism to itinerant electron magnetism. Many of still open questions concern the CaRuO₃ system: the magnetic ground state of

(351)

A. Zorkovská et al.

this compound has not been resolved yet, it is supposed to be close to ferromagnetic instability, with unconventional electronic properties due to strong correlation effects [1]. Substitution of $\operatorname{Ru}^{4+}(4d^4)$ by nonmagnetic $\operatorname{Ti}^{4+}(3d^0)$ was found to induce ferromagnetism in this system with transition temperature independent of Ti concentration [2]. Despite the clear signs of ferromagnetism in magnetic responses no features indicating long-range ordering have been observed in specific heat [3]. If the system possesses short-range order only, this will affect the magnetotransport and may reveal more information about the magnetism in this system. Here we present our results on magnetoresistance and the Hall effect investigations.

2. Experimental results and discussion

Polycrystalline samples of CaRuO₃ and CaTi_{0.07}Ru_{0.93}O₃ have been prepared by mixing CaCO₃, RuO₂ and TiO₂, afterwards pressed into pellets that had been sintered at 1100–1200°C for 72 h in air. Powder X-ray diffraction measurements confirmed the purity of the samples. Electrical resistivity in the temperature range of 2–300 K has been measured, transversal magnetoresistance and the Hall voltage have been taken within a full magnetization curve in the range of 9 T at selected temperatures (2, 5, 10, 15, 30, 33, 35, 40, 50, and 100 K) using the conventional Quantum Design PPMS-9 device.



Fig. 1. Resistivity vs. temperature of $CaRuO_3$ and $CaTi_{0.07}Ru_{0.93}O_3$. The solid and dotted lines represent the power-law fits. Inset: the Hall coefficient of $CaRuO_3$.

Resistivity. The temperature dependence in Fig. 1 shows that while the CaRuO₃ sample is metallic, Ti substitution changes the character of transport to semiconducting type. This is not unexpected, since the CaTiO₃ is an insulator and the CaRuO₃ itself is a narrow band conductor. The analysis of the dependence reveals non-Fermi liquid features with two types of anomalous T^n behaviour for the CaRuO₃ sample, in accordance with [4]: below 40 K n = 1.5 was found, which is typical of a 3D system near the antiferromagnetic quantum-critical point [5] and



Fig. 2. Transversal magnetoresistance of (a) CaTi_{0.07}Ru_{0.93}O₃ and (b) CaRuO₃.

is in contrast with the ferromagnetic features of the sample; between 45 and 100 K the dependence can be fitted by n = 0.38.

Magnetoresistance (MR). We found a sign reversal between 10 and 15 K for the substituted sample (at low temperatures interestingly negative, increasing temperature brings about the change to positive values). The MR of $CaRuO_3$ is more complicated, the sign reversal is induced by magnetic field, and only at temperatures below 15 K the MR turns from negative to positive at certain critical field (Fig. 2). All field dependences are unhysteretic, only for the substituted sample there was found a small hysteresis at 2 K. The sign change and the field dependence of MR in these systems is very similar to the prediction of the model proposed by [6] for non-magnetic granular materials in which electron transport is dominated by hopping between neighbouring clusters; the sign reversal is related to the value of cluster separation. If we apply this model to our system (that is a strongly correlated, close to metal-insulator transition, in which the conductivity can be dominated by hopping), it should be a clustered system, in which the cluster separation increases with temperature increase. The irreversibility of zero field and field cooling magnetisation curves [3] supports this conjection, indicating that the nanoclusters found in manganites, cuprates, and cobaltates might be characteristic of other oxides as well. As the sign reversal occurs at temperatures slightly below the temperature of the magnetisation onset, we may suppose that magnetic clusters can be concerned.

The inset in Fig. 1 shows how the Hall coefficient in CaRuO₃ changes with temperature: there is again a sign reversal from electron-like negative values to positive ones at 35 K. This sign reversal might indicate multiband contributions to conductivity, on the other hand there is a question how it coud be related to the clustered character of the system. The fact that it occurs near the magnetic transition temperature $T_c = 34$ K suggests that the magnetic state and the carrier character interrelate.

3. Conclusion

Electrical resistivity, transversal magnetoresistance and the Hall voltage measurements on $CaRuO_3$ and $CaTi_{0.07}Ru_{0.93}O_3$ perovskites have revealed peculiarities that might be connected to the change of the magnetic state and to the clustered character of the system. Specifically, temperature and field dependent sign reversals of magnetotransport characteristics have been observed. Morover, the resistivity of $CaRuO_3$ indicates non-Fermi liquid features in two different temperature regions.

Acknowledgments

This work was supported by the Grant Agency VEGA, grant No. 1/3027/06, by the Science and Technology Assistance Agency, contract No. 20-005204, Research Project AVZO 10100520, and by the Czech–Slovak bilateral project SK-CZ-09206. I. Bradarić was supported by the Serbian Ministry of Science, Technology and Development, grant No. 141014/2006. Material support of U.S. Steel is acknowledged.

References

- K. Yoshimura, T. Imai, T. Kiyama, K.R. Thurber, A.W. Hunt, K. Kosuge, *Phys. Rev. Lett.* 83, 4397 (1999).
- [2] T. He, R.J. Cava, *Phys. Rev. B* **63**, 172403 (2001).
- [3] A. Zorkovská, A. Baran, I. Bradarić, I. Savić, J. Šebek, E. Šantavá, P. Svoboda, D. Marinčev, S. Kohout, H. Keller, A. Feher, J. Magn. Magn. Mater. 316, e699 (2007).
- [4] L. Klein, L. Antognazza, T.H. Geballe, M.R. Beasley, A. Kapitulnik, *Phys. Rev. B* 60, 1448 (1999).
- [5] F. Steglich, B. Buschinger, P. Gegenwart, M. Lohmann, R. Helfrich, C. Langhammer, P. Hellmann, L. Donnevert, S. Thomas, A. Link, C. Geibel, M. Lang, G. Sparn, W. Assmus, *J. Phys., Condens. Matter* 8, 9909 (1996).
- [6] X.R. Wang, S.C. Ma, X.C. Xie, Europhys. Lett. 45, 368 (1999).