phys. stat. sol. (a) **189**, No. 3, 673–676 (2002)

Absence of Polaron Conductivity in $La_{0.8}Ba_{0.2}MnO_3$

A. A. Arsenov (a), N. G. Bebenin (b), V. S. Gaviko (b), V. V. Mashkautsan (b), Ya. M. Mukovskii[1]) (a), D. A. Shulyatev (a), V. V. Ustinov (b), R. I. Zainullina (b), C. P. Adams (c, d), and J. W. Lynn (c, d)

(a) Moscow State Steel & Alloys Institute, Leninskii prosp. 4, Moscow 117936, Russia

(b) Institute of Metal Physics, Ural Division of RAS, Kovalevskaya Str. 18, Ekaterinburg 620219, Russia

(c) NIST Center for Neutron Research, Gaithersburg, MD 20899-8562, USA

(d) Department of Physics, University of Maryland, College Park, MD 20742, USA

(Received May 1, 2001; accepted September 30, 2001)

Subject classification: 72.20.Ee; 75.50.Pp; S10.15

Hall effect and inelastic neutron scattering measurements have been carried out to study the properties of a $La_{0.8}Ba_{0.2}MnO_3$ single crystal grown by the floating zone method. The Hall effect measurements give evidence for a mobility edge that dominates conduction, which implies that the localized e_g electrons play an insignificant role. This result is in agreement with the neutron data which shows no central peak in the magnetic fluctuation spectrum below the Curie temperature, a feature associated with polaron formation in $La_{1-x}Ca_xMnO_3$ and $La_{1-x}Sr_xMnO_3$ manganites. In the vicinity of the Curie temperature, the change in the resistivity and in the normal and anomalous Hall coefficients of $La_{0.8}Ba_{0.2}MnO_3$ is due to the change in the concentration of holes activated by the mobility edge.

Introduction Colossal magnetoresistance (CMR) is observed in the lanthanum manganites $La_{1-x}D_xMnO_3$ where D = Ca, Sr, Ba [1–4] and $0.15 < x < 0.5$. Far below the Curie temperature T_C, a CMR manganite can be a metal with a rather complicated Fermi surface [5, 6] or an insulator, depending on x. Near T_C, the resistivity ϱ usually exhibits a peak; application of a magnetic field suppresses the peak and shifts it to a higher temperature giving rise to the CMR effect. The origin of the CMR as well as the nature of the insulating states above and below T_C still remain unclear although many models have been proposed. The published experimental data seem to be insufficient to distinguish between different scenarios because the data refer mainly to resistivity and magnetoresistance, which depend on the mechanism of charge transport in a too complicated manner. In the present article, we report on neutron scattering and Hall effect measurements in a $La_{0.8}Ba_{0.2}MnO_3$ single crystal. This crystal was selected for study because there is comparatively little information on the manganites of $La_{1-x}Ba_xMnO_3$ family. We believe, however, that our results will be useful for understanding the transport properties of all the manganites regardless of the type of divalent ion.

Experimental The single crystal of $La_{0.8}Ba_{0.2}MnO_3$ was grown by the floating zone method with radiation heating as described in Ref. [7]. The Hall resistivity ϱ_H was mea-

[1]) Corresponding author; Phone: 095-2304689; Fax: 095-2365495;
e-mail: mukovskii@synt.misa.ac.ru

 0031-8965/02/18902-0673 $ 17.50+.50/0

sured by a potentiometric method for two directions of a magnetic field and electric current. The inelastic neutron scattering experiments were carried out on the BT-2 triple-axis spectrometer at the NIST research reactor.

Results and Discussion At low temperatures, the crystal is in the orthorhombic Pnma phase; the transition to rhombohedral R3c phase occurs at $\approx$190 K. The peculiarities connected with the structural phase transition have been published elsewhere [8]; here we consider the properties in the vicinity of the Curie point.

The Curie temperature determined through Arrott-Belov curves is 251 K, which is close to the value of 248 K that was extracted from the temperature dependence of the ferromagnetic Bragg peak. This small difference between the Curie temperature values determined by different methods is typical of the CMR manganites [9].

The neutron scattering measurements revealed that in the long wavelength regime, the magnetic excitations are spin waves at low temperatures with a quadratic dispersion relation $E = Dq^2$. Figure 1 shows a magnetic inelastic spectrum for a wave vector $q = 0.16$ Å^{-1} along the (001) direction in the $La_{0.8}Ba_{0.2}MnO_3$ single crystal near the (001) peak. The measurement was taken at 240 K, only 8–10 K below the Curie temperature. The fit shown is a resolution corrected fit to the data. The data are well described by damped spin waves whose spin wave stiffness D is 40(2) meV Å^2 at this temperature. At the transition D renormalizes to zero as would be expected for a conventional ferromagnetic transition. A temperature-independent flat background (2 counts) and incoherent background (20 counts at $E = 0$ meV) have been subtracted from the data.

In $La_{1-x}Ca_xMnO_3$ and $La_{1-x}Sr_xMnO_3$ manganites a quasielastic component comparable in intensity to the spin waves appears in the ferromagnetic phase on approaching T_C [10–13]. As we can see in Fig. 1 this is not the case in $La_{0.8}Ba_{0.2}MnO_3$. It has been suggested that this central peak is associated with polarons or the localization of the e_g electrons on the Mn^{3+}/Mn^{4+} lattice. As was shown in Ref. [13] in the case of $La_{0.7}Ca_{0.3}MnO_3$ the behavior of the temperature dependence of the intensity of the satellite peak is very similar to the temperature dependence of resistivity. The absence of the central peak in $La_{0.8}Ba_{0.2}MnO_3$ thus indicates that the concentration of the localized states in $La_{0.8}Ba_{0.2}MnO_3$ is markedly less than in $La_{1-x}Ca_xMnO_3$ or $La_{1-x}Sr_xMnO_3$ and hence charge carriers in extended states dominate the transport properties, at least near T_C.

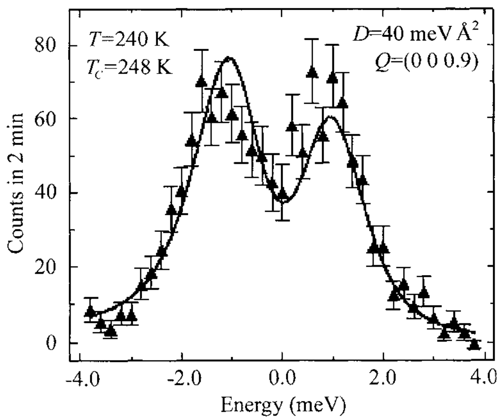

Fig. 1. Constant-Q scan showing spin waves along the (001) direction in $La_{0.8}Ba_{0.2}MnO_3$ single crystal. The measurement was taken at 240 K, only 8–10 K below T_C. The fit shown is a resolution-corrected fit to the data. The data are well described by damped spin waves with no quasielastic central peak

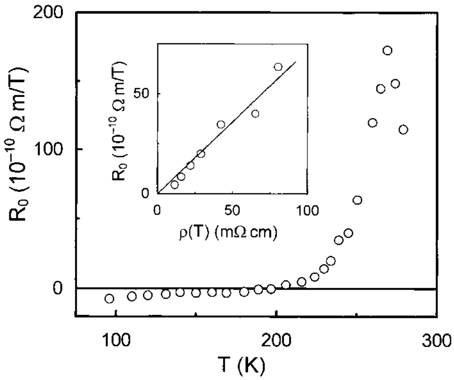

Fig. 2. Temperature dependence of the normal Hall coefficient R_o in a $La_{0.8}Ba_{0.2}MnO_3$ single crystal. Inset: R_o versus resistivity ϱ

Let us turn to the Hall effect data. In a ferromagnet:

$$\varrho_H = R_o B + R_s M, \tag{1}$$

where R_o and R_s are normal and anomalous (spontaneous) Hall coefficients, respectively, B is the magnetic induction, which can be taken to be equal to H since the demagnetizing factor is ordinarily close to one. The data for R_o and R_s are usually interpreted in terms of the relations derived for a metal with the band carriers of one type:

$$R_o = \pm \frac{1}{enc}\,, \tag{2}$$

$$R_s = a\varrho + b\varrho^2, \tag{3}$$

where the upper (lower) sign in (2) refers to holes (electrons), n is the concentration of the carriers and ϱ is the resistivity. The first term in (3) is associated with skew scattering and the second is due to the side-jump process. Figure 2 shows the temperature dependence of R_o. Above 200 K, R_o is positive, increases rapidly with T and reaches a maximum at ≈260 K. In the inset, we plot R_o against ϱ in the range 210 to 250 K. It is evident that $R_o \propto \varrho$ with the Hall mobility μ_H = 0.07 $cm^2/(Vs)$. In accordance with Ref. [14] we infer that for T > 210 K the transport is determined by the holes activated over the mobility edge. The hopping conductivity is therefore of minor importance, which agrees with our neutron scattering data that implies no localized e_g electrons. It follows also that Eq. (2) does not hold near and above T_C and hence an attempt to use that equation for determining the change in the charge carriers' concentration in the vicinity of T_C would be incorrect.

The measurements of the Hall effect in $La_{1-x}Sr_xMnO_3$ with $x \geq 0.18$ [15] have revealed that even at low temperatures, where the conductivity is metallic, the normal Hall coefficient estimated in accordance with (2) is much less than the experimental one. This perhaps indicates that the hops between

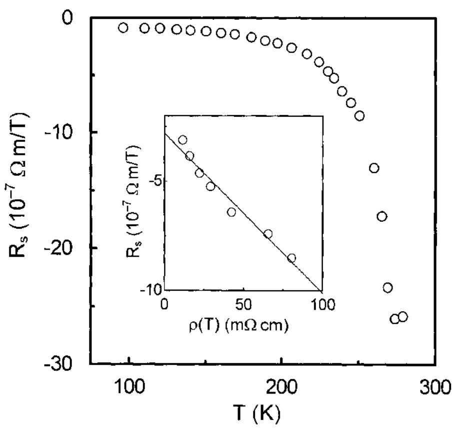

Fig. 3. The temperature dependence of the anomalous Hall coefficient R_s in a $La_{0.8}Ba_{0.2}MnO_3$ single crystal. Inset: R_s versus ϱ

localized states, whose contribution to R_o is negative, give a significant contribution to R_o, which agrees with the strong central peak in $La_{1-x}Sr_xMnO_3$ manganites arising from these localized states.

Unlike R_o, the behavior of R_s is similar in all CMR materials. Figure 3 shows R_s for our $La_{0.8}Ba_{0.2}MnO_3$ crystal. The anomalous Hall coefficient is negative at all temperatures, and in the ferromagnetic phase it is a monotonic function of T. Obviously, in the whole ferromagnetic region, Eq. (3) is not fulfilled except over the range 210 to 250 K. R_s is approximately proportional to ϱ as is apparent from the inset in Fig. 3. This is not, however, evidence for skew scattering. Indeed since both R_o and R_s are proportional to ϱ, the changes in ϱ and the normal and anomalous Hall coefficients are due to one and the same cause, i.e. change in the concentration of holes activated to the mobility edge.

In summary, we performed neutron scattering and Hall effect measurements in a $La_{0.8}Ba_{0.2}MnO_3$ single crystal and found that near the Curie temperature the charge transport is determined by holes activated to the mobility edge, which implies that localized e_g electrons do not play a significant role. The anomalous Hall coefficient is proportional to the resistivity in the vicinity of T_C. This is not, however, evidence for skew scattering since the temperature dependence of ϱ, R_o, and R_s is caused by the change in the concentration of holes in delocalized (extended) states.

Acknowledgements The work was supported by RFBR grants #00-02-17544 and 00-15-96745. Work at the University of Maryland was supported by the National Science Foundation, DMR 97-01339.

References

[1] E. L. NAGAEV, Phys. – Uspekhi **39**, 781 (1996).
[2] A. P. RAMIREZ, J. Phys.: Condens. Matter **9**, 8171 (1997).
[3] Y. TOKURA and Y. TOMIOKA, J. Magn. Magn. Mater. **200**, 1 (1999).
[4] J. M. D. COEY, M. VIRET, and S. VON MOLNAR, Adv. Phys. **48**, 167 (1999).
[5] W. E. PICKETT and D. J. SINGH, J. Magn. Magn. Mater. **172**, 237 (1997).
[6] E. A. LIVESAY, R. N. WEST, S. B. DUGDALE, G. SANTI, and T. JARLBORG, J. Phys.: Condens. Matter **11**, L279 (1999).
[7] D. SHULYATEV, S. KARABASHEV, A. ARSENOV, and YA. MUKOVSKII, J. Cryst. Growth **198/199**, 511 (1999).
[8] V. E. ARKHIPOV, N. G. BEBENIN, V. P. DYAKINA et al., Phys. Rev. B **61**, 11229 (2000).
[9] S. E. LOFLAND, S. M. BHAGAT, K. GHOSH et al., Phys. Rev. B **56**, 13705 (1997).
[10] J. W. LYNN, R. W. ERWIN, J. A. BORCHERS et al., Phys. Rev. Lett. **76**, 4046 (1996).
[11] L. VASILIU-DOLOC, J. W. LYNN, Y. M. MUKOVSKII et al., J. Appl. Phys. **83**, 7342 (1998).
[12] L. VASILIU-DOLOC, J. W. LYNN, A. H. MOUDDEN et al., Phys. Rev. B **58**, 14913 (1998).
[13] C. P. ADAMS, J. W. LYNN, Y. M. MUKOVSKII, A. A. ARSENOV, and D. A. SHULYATEV, Phys. Rev. Lett. **85**, 3954 (2000).
[14] N. F. MOTT and E. A. DAVIS, Electronic Processes in Non-Crystalline Materials, 2nd ed., Clarendon Press, Oxford 1979.
[15] A. ASAMITSU and Y. TOKURA, Phys. Rev. B **58**, 47 (1998).