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Magnetoresistance of $La_{0.7}Ca_{0.3}MnO_3$ single crystal and polycrystalline

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The electronic and magnetic properties of single crystal and polycrystalline $La_{0.7}Ca_{0.3}MnO_3$ have been studied in order to obtain the information of magnetoresistance. Both specimens exhibit a transition from a ferromagnetic-metal state to a paramagnetic-insulator state near 250K. The resistance $R(H)$ of single crystal specimen decrease gradually with increasing magnetic field (0 Oe $\leqq H \leqq$ 70 kOe). On the other hand, the resistance of polycrystalline specimen decreases drastically for the initial increment of magnetic field ($H \leqq$ 3 kOe), which is proportial to magnetization. Considering these results, we discuss the mechanism of magnetoresistance in single and polycrystalline compounds.

Key words:metal-insulator transition, colossal magnetoresistance, grain boundary, rare-earth metal ion

Perovskite manganites with 20-40% of trivalent rare-earth metal ions (R^{3+}) replaced by divalent alkaline-earth metal ions of (A^{2+}), $R_{1-x}A_xMnO_3$, have been studied for many years, and many interesting phenomena, such as metal-insulator transition and colossal magnetoresistance, were reported so far. These phenomena are mainly explained by double-exchange interaction; magnetic interaction between Mn^{3+} and Mn^{4+} that is caused by the hopping of e_g electrons between the two partially filled d shells with strong on-site Hund's coupling[1]. Hwang *el al.*[2] investigated the magnetoresistance (MR) of $La_{2/3}Sr_{1/3}MnO_3$ below Curie temperature T_C, and pointed out that there is similarity between the MR of polycrystalline specimen and that of granular nickel films[3]. Raychauhuri *et al.*[4] investigated the MR of polycrystalline specimens of $La_{0.7-x}Ho_xSr_{0.3}MnO_3$ (x = 0, 0.15) and proposed a tunneling model to explain the behavior of MR of polycrystalline specimens. However, the mechanism of this MR has not been clear yet. In order to confirm this model, we examine MR of $La_{0.7}Ca_{0.3}MnO_3$ maganite together with that of single crystal for comparison.

High purity La_2O_3, $CaCO_3$ and Mn_3O_4 were mixed for the compositions mentioned above and calcined in air at 1273K for 12h. Sintering was carried out in air at 1627K for 48h after intermediate grinding. Polycrystalline specimen was cut from sintered disks and its grain size was about 10μm. Single crystal specimen was prepared by a floating zone method. Both specimens were confirmed to be a single phase (pseudo cubic structure) by X-ray diffraction.

Electrical resistance R measurements for the single crystal specimen (0.8mm × 1.0mm × 2.8mm) and polycrystalline specimen (0.5mm × 2.5mm × 2.5mm) were made by a standard four-probe method. As a result, both specimens show a transition from ferromagnetic metal to paramagnetic insulator at about 250K.

Then, we measured magnetoresistance of single crystal and polycrystalline specimens by applying magnetic fields of up to 70 kOe below T_C. The results of single crystal and polycrystalline specimens are shown in Fig. 1(a) and (b), respectively, where rsistance $R(H)$ is normalized by the value of resistance at H = 0 ($R(H)/R(0)$). As known from Fig. 1(a), $R(H)/R(0)$ of single crystal specimen decreases gradually with increasing magnetic field. As for magnetoresisitance of single crystal, Searle and Wang derived a model, which is based on double-exchange interaction. Then, we calculated the MR of single crystal by using this model. The result is shown by dotted curves in Fig.1(a). It should be noted that experimental results are in good agreement with the calculated ones except for high magnetic field region. We also calculated temperature dependence of resistance by using this model. The calculated dependence is in good agreement with experimental one as shown in Fig.2

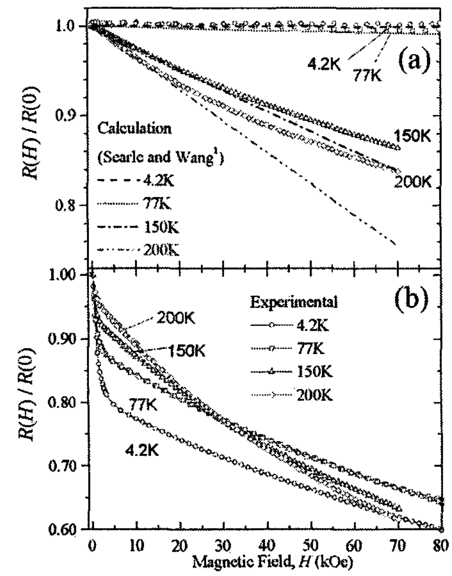

Fig.1 Magnetoresistance curves of $La_{0.7}Ca_{0.3}MnO_3$ single crystal and polycrystalline specimens.

Magnetoresistance of polycrystalline specimen is completely different from that of the single crystal specimen. As seen in Fig.1(b), $R(H)/R(0)$ drops by the initial increment of magnetic field (H< 3 kOe), as in the previous studies[2,4]. The drop becomes significant with decreasing temperature, and is about 20% at 4.2K. When magnetic field is higher than 3 kOe, $R(H)/R(0)$ of polycrystalline specimen decreases gradually with

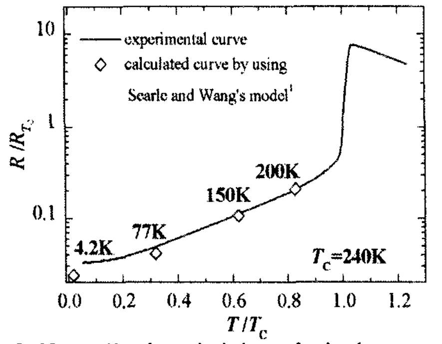

Fig.2 Normalized resistivity of single crystal specimen as a function of T/T_C.

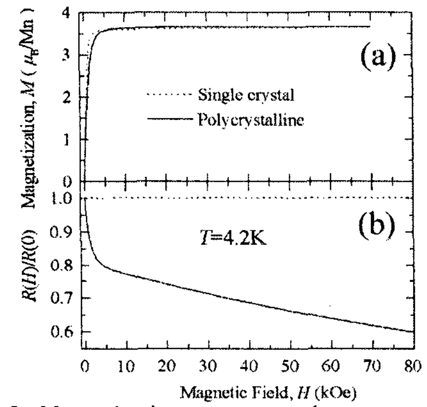

Fig.3 Magnetization curves and magnetoresistance curves of single crystal and polycrystalline specimens

increasing magnetic field up to 80 kOe

To obtain more information for MR of polycrystalline specimen, we measured the magnetization curve and compared it with $R(H)/R(0)$ as shown in Fig.3. In this figure, results of single crystal specimen are also shown for comparison. Magnetization processes are almost the same for polycrystalline and single crystal specimens. However, the magnetoresistance curve of polycrystalline is different from that of single crystal specimen, that is, while $R(H)/R(0)$ of polycrystalline specimen decreases with increasing magnetization, that of single crystal specimen does not. Raychaudhuri *et al.*[4] reported that the drastic decrease of $R(H)/R(0)$ in polycrystalline specimens at low magnetic fields can be explained by the tunneling conductivity through grain boundaries, and they derived the following relation:

$$\frac{\Delta R}{R(0)} = \frac{R(H)-R(0)}{R(0)}$$

$$= \frac{\left(\frac{M_S}{M_{S0K}}\right)^2 \langle \cos\theta(0)\rangle - \left(\frac{M_S}{M_{S0K}}\right)^2 \langle \cos\theta(H)\rangle}{1+\left(\frac{M_S}{M_{S0K}}\right)^2 \langle \cos\theta(H)\rangle} \quad (1)$$

where M_S and M_{S0K} are the spontaneous magnetization at T and 0K, respectively, $\theta(H)$ is the angle between magnetizations of two neighboring grains at a magnetic field H and $\langle\cos\theta(H)\rangle$ is the average of $\cos\theta(H)$. According to Eq.1, $\Delta R/R(0)$ depends on only M_S/M_{S0K} with a parameter $\langle\cos\theta(0)\rangle$ when the moment is saturated. Then we tried to fit $\Delta R/R(0)$ using Eq.(1) at H=3kOe, where magnetizations of grains are almost aligned with the magnetic field, but the fitting was not good. An example of the relation calculated from Eq.(1) with $\langle\cos\theta(0)\rangle = 0.717$ is shown in Fig.4 together with the experimental one. It is apparent that experimental results does not agree with Eq.(1). We also considered the expanded case in which each grain behaves like a cluster spin-glass. Then, it is well known that $\Delta R/R(0)$ is proportional to $(M/M_S)^2$ when $(M/M_S)^2$ is enough small[5]. However, we found that $\Delta R/R(0)$ of polycrystalline specimen is proportional to (M/M_S) rather than $(M/M_S)^2$. The relation between $\Delta R/R(0)$ and (M/M_S) is shown in Fig.5. This result suggests that we should consider a mechanism of which $\Delta R/R(0)$ of polycrystalline specimen is proportional to (M/M_S).

As we mentioned before, when magnetic field is higher than 3 kOe, $R(H)/R(0)$ of polycrystalline specimen decreases gradually with increasing magnetic field up to 80 kOe. The decrease is probably caused by the suppression of magnetic fluctuation around grain boundaries because magnetic moments in each grain is already aligned.

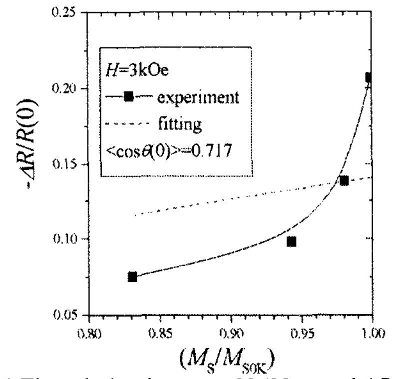

Fig.4 The relation between M_S/M_{S0K} and $\Delta R/R(0)$ of $La_{0.7}Ca_{0.3}MnO_3$ polycrystalline specimen.

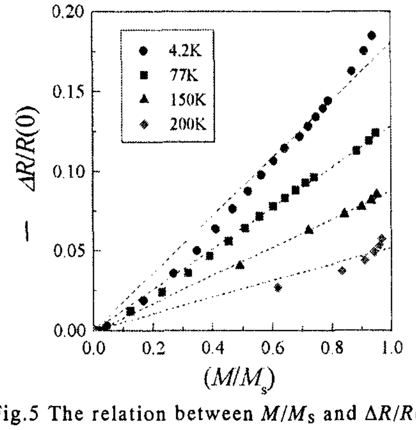

Fig.5 The relation between M/M_S and $\Delta R/R(0)$ of $La_{0.7}Ca_{0.3}MnO_3$ polycrystalline specimen.

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