

Al-substitution effect on charge transport in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$: Incoherent metallic state in a double-exchange ferromagnet

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Resistivity $\rho(T)$ was measured on $(\text{La}_{1-x}\text{Sr}_x)(\text{Mn}_{1-z}\text{Al}_z)\text{O}_3$ single crystals over a temperature T range 4–1000 K. The doped Al decreases Curie temperature T_C and increases $\rho(T)$. At low temperatures, the resistivity shows the upturn when the residual resistivity reaches the critical value estimated about $300 \mu\Omega \text{ cm}$. This upturn is most likely to originate from the carrier localization and therefore mean free path l reaches Fermi wavelength λ_F at this critical resistivity. This suggests that the basic assumption of an ordinary metal, $l > \lambda_F$, is violated when $\rho(T)$ exceeds this critical value even though $\rho(T)$ is metallic ($d\rho/dT > 0$). This ‘‘coherent-incoherent’’ crossover is also suggested by the recent optical studies. The metallic $\rho(T)$ above T_C , on the other hand, becomes insulating by only a few percent Al. The conduction above T_C is dominated by a short-range correlation between the t_{2g} spins.

I. INTRODUCTION

The charge transport has been widely recognized to be a key for understanding colossal magnetoresistance (CMR) in the double-exchange (DE) ferromagnetic manganites as well as for designing these materials for industrial applications.¹ However, the transport mechanism is far from comprehensively understood: (i) Although the dc resistivity $\rho(T)$ shows metallic behavior ($d\rho/dT > 0$), its magnitude seems unusually large.^{2–4} (ii) There is a lot of variation within the transport and optical properties; for example, some materials do not show Drude-like response in the optical conductivity $\sigma(\omega)$ spectrum even though $\rho(T)$ is metallic below Curie temperature T_C .^{5–12} These features suggest a conduction mechanism beyond the previous framework consisting of the Bloch-Boltzmann theory¹³ and the DE mechanism.^{14–17}

We have measured the dc resistivity of Al-substituted $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ (LSMO) single crystals over a wide temperature range (4–1000 K). Aluminum doped at the $B(\text{Mn})$ site has the closed-shell configuration—with no d electrons and no magnetic moments—and has the same valence with the host manganese (+3). Therefore Al substitution induces only the following two types of randomness: (i) random potential (electrical randomness) and (ii) local cutoff of magnetic interaction between the spins of the t_{2g} electrons (magnetic randomness). For the case of substitution with transitional metals such as Cr or Co,^{18–20} the doped element may change the doping level and/or may cause other secondary effects due to excess d electrons and/or magnetic moments, which make the study complicated and less transparent.

The main claim of the present study is that the ferromagnetic-metallic phase of the manganites is classified into two phases—low- T coherent and high- T incoherent—phases according to the magnitude of mean free path. We discuss the role of the additional features, especially of randomness, in appearance of the incoherent metallic state which is relevant for CMR.

II. EXPERIMENT

Single crystals of $(\text{La}_{1-x}\text{Sr}_x)(\text{Mn}_{1-z}\text{Al}_z)\text{O}_3$ and $(\text{Nd}_{0.70}\text{Sr}_{0.30})(\text{Mn}_{1-z}\text{Al}_z)\text{O}_3$ were grown by a floating zone method.² The chemical composition of the crystal was checked by inductively coupled plasma (ICP) spectroscopy and was confirmed to be the same with the starting compositional ratio. Magnetization $M(T)$ was measured with a superconducting quantum interference device magnetometer under a field of $\mu_0 H = 0.5$ T after cooling down to 5 K in zero field. The measurements of $\rho(T)$ were performed using a conventional four-probe method. The crystal was cut into a rectangular shape, with typical dimension of $4 \times 1 \times 0.5 \text{ mm}^3$. After electrodes were formed with heat-treatment-type gold paste, the crystals were annealed under flowing O_2 gas for 20 h at 600–450 °C. The high-temperature measurements above 300 K were conducted in an atmosphere in O_2 . We confirmed that there is negligible small difference between the $\rho(300 \text{ K})$ values measured before and after heating up to 1000 K. In addition, for representative compositions we also measured $\rho(T)$ below 300 K on the samples quenched from 700 °C to liquid nitrogen after annealing under oxygen gas for 20 h. No difference was observed between these quenched and those annealed samples. These facts guarantee that the oxygen content was not changed during the $\rho(T)$ measurement at high temperatures. The oxygen content was determined by iodometric titration and was found to be within 2.980–2.988 for all of the crystals. We measured $\rho(T)$ on several crystals with the same composition to confirm that the scattering of the data is within the dimensional errors ($\pm 4\%$).

III. RESULTS

Figure 1 shows the results of the Al-substitution effect on the dc resistivity of LSMO for various Sr compositions ($x = 0.175–0.30$). As the substitution proceeds, T_C decreases and $\rho(T)$ increases (Fig. 2). $\rho(T)$ shows upturn at low temperatures for the Al content $z \geq 0.02$ ($x = 0.175$), ≥ 0.043 (0.20), and ≥ 0.0675 (0.30) (insets of Fig. 1). The

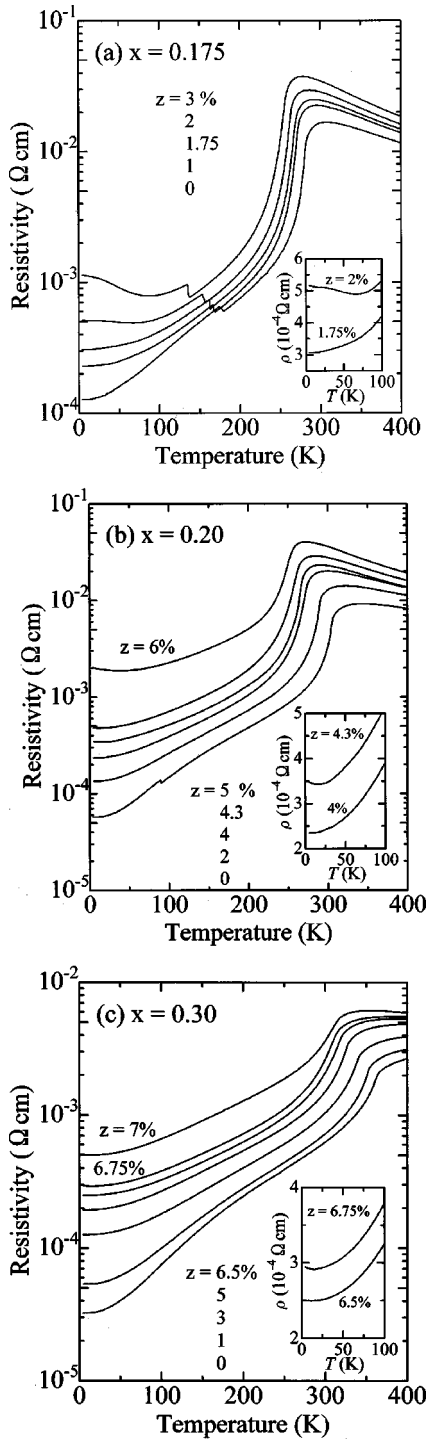


FIG. 1. Temperature dependence of the dc resistivity measured on $(\text{La}_{1-x}\text{Sr}_x)(\text{Mn}_{1-z}\text{Al}_z)\text{O}_3$ single crystals for (a) $x=0.175$, (b) $x=0.20$, and (c) $x=0.30$. Insets show magnified dc resistivity in a low-temperature region (0–100 K).

increase of $\rho(T)$ can be ascribed mainly to the elastic scattering by the Al-induced random potential and therefore the upturn is probably to reflect localization of the carriers. The reason of this interpretation is described below.

As is shown later (insets of Fig. 4), Al substitution little changes the saturated magnetization and hence it does not alter essentially the magnetic structure; the decrease of T_C , which shows z -linear behavior, originates from suppression of long-range order of the t_{2g} spins by local cutoff of the

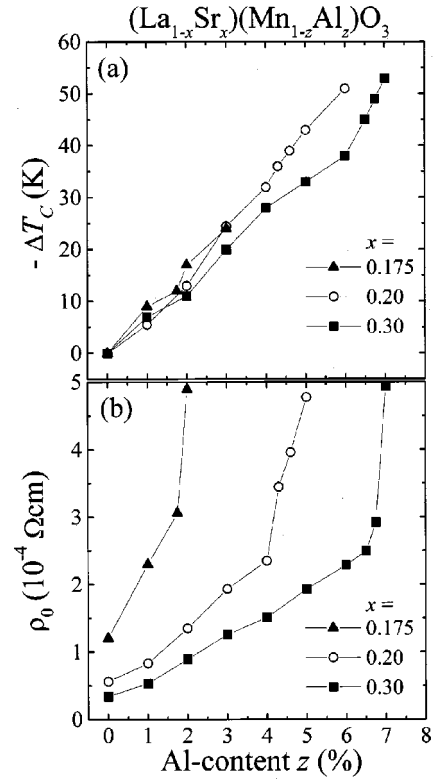


FIG. 2. Variation of (a) T_C and (b) ρ_0 with Al content z . ρ_0 represents the minimum value of $\rho(T)$, which corresponds to the residual resistivity for the itinerant compositions.

magnetic interaction. Therefore, at low temperatures where the magnetic degrees of freedom are frozen out because of the complete spin polarization, the doped Al is expected to act only as a potential scatterer with negligible small change in the doping level.²¹ The anomaly observed at about 150 K for $x=0.175$, shown in Fig. 1(a), is due to the structural transition from the high- T rhombohedral to low- T orthorhombic form. Because the anomaly temperature T_s is sensitive to the distortion,² rather weak z dependence of T_s indicates that the Al substitution has only a little effect on the crystal structure. In addition, T_s decreases with increasing z , which shows that the distortion becomes weaker because the ionic radius of aluminum is larger than that of manganese. Because the perovskite manganite generally becomes more conductive as the distortion becomes weaker, the increase of $\rho(T)$ cannot be ascribed to the structural change.

In Fig. 2(b), ρ_0 represents the minimum $\rho(T)$ value. ρ_0 shows z -linear behavior for the small- z compositions for which the upturn is absent. However, it deviates from the z -linear behavior and rapidly increases with z for the large- z compositions for which $\rho(T)$ shows the upturn. This change is similar to the typical behavior of the residual resistivity at the neighborhood of the Anderson localization.²² Here, we define σ^* ($=1/\rho^*$) as the critical dc conductivity value which separates itinerant conduction from localized one at absolute zero. The upper limit of ρ^* is defined as an extrapolated value to the minimum Al content at which $\rho(T)$ shows the upturn, assuming the z linearity of ρ_0 for the itinerant compositions. The lower limit of ρ^* is, on the other hand, defined as the value of ρ_0 for the maximum Al content in the itinerant region. The values of σ^* estimated by

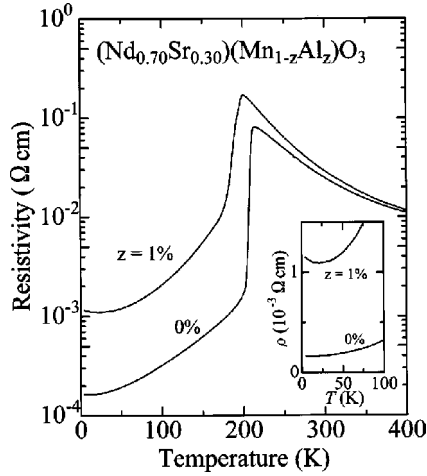


FIG. 3. Temperature dependence of the dc resistivity measured on $(\text{Nd}_{0.70}\text{Sr}_{0.30})(\text{Mn}_{1-z}\text{Al}_z)\text{O}_3$ single crystals. Inset shows magnified dc resistivity in a low-temperature region (0–100 K).

the above procedure are $2800\text{--}3400 \Omega^{-1} \text{cm}^{-1}$ ($x=0.175$), $3500\text{--}4100 \Omega^{-1} \text{cm}^{-1}$ (0.20), and $3700\text{--}4000 \Omega^{-1} \text{cm}^{-1}$ (0.30). It is noteworthy that σ^* shows no remarkable x dependence.

The slope of residual resistivity vs density of impurity, $\Delta\rho/n_{\text{imp}}$, is $\sim 106 \mu\Omega \text{cm/at. \% Al}$ ($x=0.175$), $\sim 46 \mu\Omega \text{cm/at. \% Al}$ (0.20), and $\sim 32 \mu\Omega \text{cm/at. \% Al}$ (0.30). These values are much larger than that of a typical transitional-metal alloy AlCr , $\sim 8 \mu\Omega \text{cm/at. \% Cr}$,²³ and, for $x=0.175$, the slope is comparable with that of $\text{YBa}_2(\text{Cu}_{1-z}\text{Zn}_z)_3\text{O}_{6.68}$, $\sim 125 \mu\Omega \text{cm/at. \% Zn}$.²⁴ For the case of $\text{YBa}_2(\text{Cu}_{1-z}\text{Zn}_z)_3\text{O}_{6.68}$, the doped Zn acts as a strong potential scatterer in the unitarity limit. Effective scattering cross section per one impurity, Q_{eff} , is estimated to be $\sim 420 \text{ \AA}^2$ ($x=0.175$), $\sim 160 \text{ \AA}^2$ (0.20), and $\sim 90 \text{ \AA}^2$ (0.30), using the following formula:

$$Q_{\text{eff}} = \frac{ne^2}{\hbar k_F} \cdot \frac{\Delta\rho}{n_{\text{imp}}} \quad (1)$$

(n : carrier density; k_F : Fermi wave number) and the estimation of n from the Hall-effect study,²⁵ $\sim 2.5 \times 10^{22} \text{ cm}^{-3}$ ($x=0.175$), $\sim 2.1 \times 10^{22} \text{ cm}^{-3}$ (0.20), $\sim 1.5 \times 10^{22} \text{ cm}^{-3}$ (0.30). These Q_{eff} values are much larger than that of Cr in Al, $\sim 33 \text{ \AA}^2$. The metallic state of the manganites is unstable against randomness. This may be a characteristic of a half metal and/or the DE system. Otherwise, this may originate from other features, for example, dynamical Jahn-Teller instability,²⁶ charge ordering instability,²⁷ orbital degree of freedom,^{28,29} and so on.

The rapid increase of $\Delta\rho/n_{\text{imp}}$, or of Q_{eff} , with decreasing x correlates with narrowing of the bandwidth with decreasing x . We also confirmed that the metallic state of $\text{Nd}_{0.70}\text{Sr}_{0.30}\text{MnO}_3$, the bandwidth of which is narrower than that of LSMO system, is destroyed by the slight (less than 1%) amount of Al (Fig. 3). This is probably because the above additional features emerges with decreasing bandwidth.¹¹

For LSMO system, σ^* is not only the critical value for the carrier localization caused by impurities but also a critical value around which the electronic state undergoes a cer-

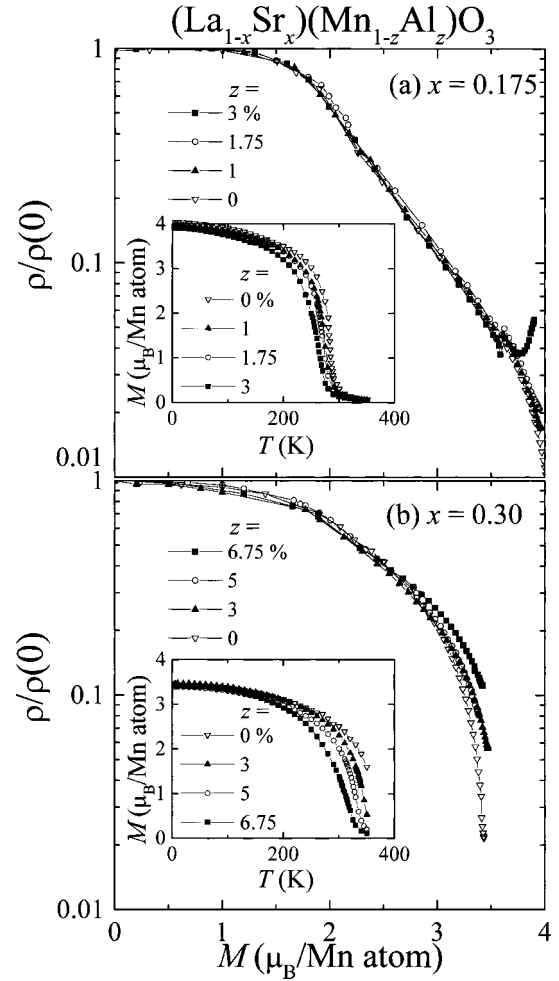


FIG. 4. The dc resistivity is replotted as a function of magnetization M ; (a) $x=0.175$, (b) $x=0.30$. Here, $\rho(0)$ is a constant of resistivity at $M=0$. Insets: temperature dependence of magnetization measured under a field of $\mu_0 H=0.5$ T.

tain essential change. In Fig. 4, the dc resistivity is replotted as a function of magnetization M for $x=0.175$ in (a) and $x=0.30$ in (b). Here, $\rho(0)$ is a constant of resistivity at $M=0$. Insets show $M(T)$ measured under a field of $\mu_0 H=0.5$ T. For both compositions, response of the charge transport to Al substitution changes around σ^* . At high temperatures, where $\rho(T)$ is, roughly speaking, larger than ρ^* , the curves of the renormalized resistivity merge into a single one, which indicates that $\rho(T)$ is governed by $M(T)$ alone and the Al substitution affects the charge transport only through $M(T)$. At low temperatures, on the other hand, the nature of the doped Al as a potential scatterer becomes conspicuous; the doped Al induces residual resistivity. $\rho(T)$ is no longer governed by $M(T)$ alone and the curves of the renormalized resistivity separate from the single one. The recent optical reflectivity studies also shows the coherent-incoherent crossover in $\sigma(\omega)$ at about $\sigma_{\text{dc}} (=1/\rho) \sim 2000 \Omega^{-1} \text{cm}^{-1}$.^{11,12} These results suggest that there is an essential change in the electronic state, roughly speaking, around $\sigma_{\text{dc}} \sim 2000\text{--}4000 \Omega^{-1} \text{cm}^{-1}$. Because it is originally difficult to estimate a distinct critical value by this procedure, and also because this change is continuous and indistinct, this uncertainty of the critical value is not significant.

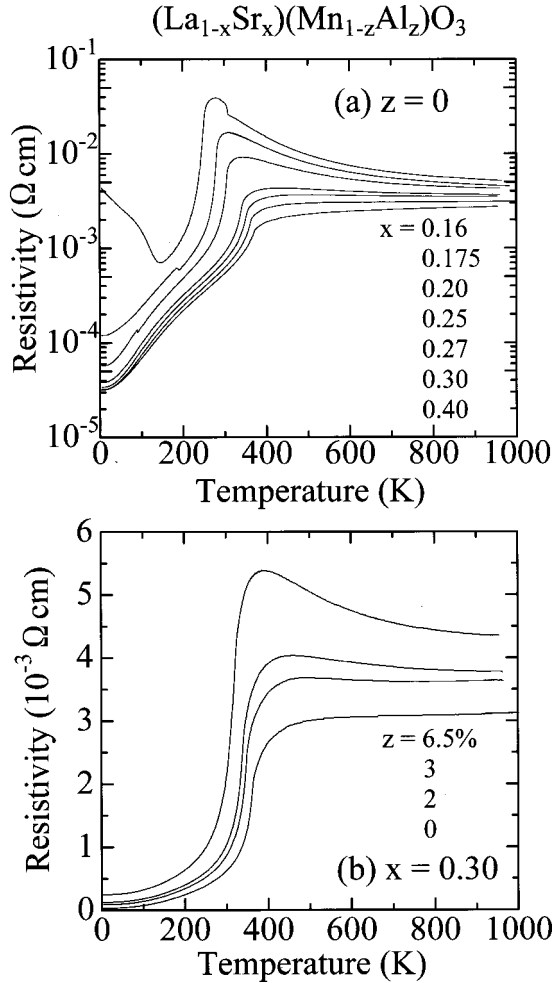


FIG. 5. (a) Temperature dependence of the dc resistivity measured on Al-free $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ single crystals for various Sr compositions ($0.16 \leq x \leq 0.40$) up to 1000 K. (b) Al-substitution effect on the dc resistivity at high temperatures for $x=0.30$.

In Fig. 5(a), we show $\rho(T)$ of Al-free LSMO up to 1000 K for various Sr compositions ($x=0.16-0.40$). In the paramagnetic phase above T_C , $\rho(T)$ changes gradually from insulating to metallic behavior with increasing x . For the heavily doped region $x \geq 0.30$, $\rho(T)$ is metallic above T_C . Though $\rho(T)$ shows weak T dependence, it does not show a tendency of saturation over a T range covered here. However, the metallic phase above T_C is quite unstable against Al substitution; for $x=0.30$, $\rho(T)$ becomes insulating by only 2% Al [Fig. 5(b)]. The insulating behavior above T_C , which is not predicted by the simple DE model,¹⁶ seems to originate from randomness.

IV. DISCUSSIONS

The upturn of $\rho(T)$ induced by Al probably originates from the localization. Therefore, around σ^* , mean free path (MFP) l is close to Fermi wavelength $\lambda_F = 2\pi/k_F$.³⁰ Therefore, because the doped Al act only as a potential scatterer at low temperatures, also for the pure system, l is close to λ_F when σ_{dc} reaches σ^* .^{31,32} Therefore, for the manganites, the condition $l > \lambda_F$, which is assumed in an ordinary metal, is violated at the higher T region of the metallic phase where

σ_{dc} is smaller than σ^* ; around σ^* the ferromagnetic-metallic phase is divided into two phases, that is, high-temperature incoherent metallic (HIM) and low-temperature coherent metallic (LCM) phases. The word ‘‘incoherent’’ means the violation of the assumption $l > \lambda_F$.

The LCM phase is realized below a certain temperature T^* at which σ_{dc} exceeds σ^* . In this phase, l is longer than λ_F and hence the concept of an ordinary metal seems valid. However, this metallic phase is so unstable against randomness that it disappears by slight Al substitution, especially for the narrow-band system.

The charge carriers gradually lose coherency with increasing temperature. HIM phase is realized over a temperature range $T^* \leq T \leq T_C$. In this phase, σ_{dc} is less than σ^* , in other words, l is less than λ_F in appearance, though $\rho(T)$ is metallic ($d\rho/dT > 0$). Therefore the concept of an ordinary metal is violated; the electrical conduction may be caused by some hopping mechanism. Actually, in this T region, a Drude-like term is absent in $\sigma(\omega)$ and, instead, $\sigma(\omega)$ is characterized by an incoherent broad peak at mid-to-near infrared region. The T dependence of ρ is determined mainly by the effective carrier density n/m^* (m^* : effective mass) because the spectral weight in $\sigma(\omega)$ at low frequencies shows a radical T dependence.¹²

One should note the large resistivity in HIM phase. For example, ρ exceeds 10 m Ω cm just below T_C for $x=0.175$, which corresponds to unphysically small MFP, $l \sim 0.15 \text{ \AA}$. In an ordinary metal, l cannot be less than lattice constant a , no matter how strongly the carriers are scattered.³⁰ Therefore some metals show ‘‘resistivity saturation’’ at high temperatures.³³ Such a large ρ of LSMO means that not only the condition $l > \lambda_F$ but also $l > a$ are violated in HIM phase;³⁴ the ρ seems to increase without saturation still after l reaches a .

As is similar to HIM phase, in the paramagnetic phase the electrical conduction is incoherent and hence random potential probably does not directly affect the charge transport. In addition, because the long-range order is absent above T_C , the remaining magnetic effect is expected to be ‘‘local.’’ Therefore the electrical conduction above T_C is determined by short-range correlation between the t_{2g} spins.¹⁷

CMR is a characteristic of HIM phase. One of the main issues is whether HIM phase is realized by the DE mechanism alone. The answer suggested here is ‘‘no.’’ This is based on the following facts: (i) The dc resistivity of CrO_2 ($T_C=392 \text{ K}$), which is one of the ideal DE materials free from the additional features,³⁵ is rather small ($\sim 6 \times 10^{-4} \text{ \Omega cm}$ at 500 K) and it seems to be in a ‘‘coherent’’ metallic state over a whole temperature range.³⁶ (ii) The stable HIM phase of the narrow-band (or high-resistivity) system is usually accompanied by the insulating phase above T_C which cannot be explained by the simple DE mechanism. (iii) An incoherent broad peak which characterizes the $\sigma(\omega)$ spectrum in HIM phase¹² also cannot be explained by the simple DE mechanism.

If there is no t_{2g} -spin misalignment, resistivity is determined by the effective carrier density and by the residual imperfections—the above-mentioned additional features, randomness, phonons, and so on. ‘‘Intrinsic’’ resistivity $\rho_i(T)$ and MFP $l_i(T)$ are defined as the resistivity and MFP

in this ideal situation, respectively. $l_i(T)$ is determined by these residual imperfections. The above three facts suggest the following: When l_i is long, the carriers keep coherency even though the misalignment of the t_{2g} spins exists; the realized metallic state is not so much different from an ordinary one; and some mechanisms which shorten l_i are indispensable for HIM phase. CMR is a process that the coherency of the carriers is restored to some extent by magnetic field. It is the incoherency of the charge dynamics itself that is essential for CMR. One should calculate how $\sigma(\omega, T)$, $M(T)$, T_C , and so on, vary with l_i (or “intrinsic” scattering time τ_i), by taking l_i (τ_i) into consideration in a phenomenological manner. [The simple DE model corresponds to the limit of $l_i(\tau_i) \rightarrow \infty$.]

The changes in the physical properties of the real DE materials are due to variation in l_i . One example may be x dependence. The universal value of σ^* indicates that n does not show remarkable x dependence because σ^* is irrelevant to carrier scattering and hence is determined primarily by carrier density n . This is consistent with the Hall-effect study.²⁵ The variation in the electronic state with Sr composition is considered to originate from the amount and/or effectiveness of the additional features. Among them, randomness seems dominant. The present study shows that there is rather large residual resistivity, corresponding to $z \sim 1\%$, also for pure LSMO [Fig. 2(b)]. It is contrary to the case of high- T_C cuprates in which the residual resistivity is negligibly small for the optimally and highly doped regions.³⁷ For the manganites, a magnetic domain wall may be an origin of the persistent residual resistivity.³⁸

There are various experimental results suggesting that randomness plays a central role in determining the electronic state of the DE materials: (i) absence of LCM phase in two-dimensional^{3,6} and slightly doped [Fig. 4(a), Ref. 2] systems, (ii) drastic changes caused by the grain boundaries of polycrystalline samples³⁹ and by the defects in the surfaces,⁹ (iii) insulating behavior of $\rho(T)$ above T_C in Al-substituted LSMO ($x \geq 0.30$) [Fig. 5(b)], (iv) large sample dependence of $\rho(T)$ for the cases of, for example, CrO_2 .^{36,40} In the DE system, charge (e_g) and spin (t_{2g}) intimately interact. Con-

sequently, σ and M are self-consistently modulated by the additional imperfections.⁴¹ In this case, effects of randomness may be much more drastic than that expected for the case of an ordinary metal; for example, the present result indicates that Matthiessen’s rule is not applicable except for a very low-temperature region at which the t_{2g} -spin alignment is almost complete. The rather large residual resistivity for highly doped, Al-free LSMO and the large Q_{eff} may be another example. Among various candidates for controlling l_i , randomness is one of the most useful one for the manganites because we can easily and systematically alter it via B -site substitution.

V. SUMMARY

The ferromagnetic-metallic phase of the doped manganites is divided into two—LCM and HIM—phases at a certain critical dc conductivity, which is indicated not only by the present study of Al-substitution effect on dc resistivity but also by the recent optical reflectivity studies. The present result shows that this essential change in the charge dynamics, that is, the LCM-HIM crossover, is determined by the magnitude of mean free path. Even for the DE system, MFP l is one of the significant indexes in discussing the electronic state. The present result also shows that the metallic $\rho(T)$ above T_C becomes insulating by slight amount of Al. The conduction above T_C is determined by the short-range correlation between the t_{2g} spins. The present study suggests that randomness is one of the key factors for controlling the physical properties of the manganites and we can design these materials via B -site substitution.

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- ³²Assuming a spherical Fermi surface, k_F is $(3\pi^2n)^{1/3}$. Therefore, using a Drude formula $\sigma_{dc} = ne^2\tau/m^*$, l is represented as $l = \hbar k_F \sigma_{dc} / ne^2$. For the case of $x=0.30$, for example, n is estimated to be $1.5 \times 10^{22} \text{ cm}^{-3}$ (0.87 per Mn atom) from the Hall-effect study (Ref. 25). This yields $k_F \sim 7.6 \times 10^7 \text{ cm}^{-1}$. If we substitute σ_{dc} with σ^* estimated here, l is estimated to be $8-8.5 \text{ \AA}$, which roughly coincides with $\lambda_F \sim 8.3 \text{ \AA}$.
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