Kondo-like transport and its correlation with the spin-glass phase in perovskite manganites

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A Kondo-like transport was observed in a metal-semiconductor transition (MSC) at low temperatures in the ferromagnetic metallic phase of the perovskite manganites. Experimental data can be best fitted in the framework of Kondo scattering, electron-electron (e-e), and electron-phonon (e-p) interaction. The results show that this behavior depends strongly on the content of the spin-glass phase and can be tuned with an applied magnetic field, which can be explained by the spin disorder scattering of electrons and/or antiferromagnetic cluster on a nanoscale and/or microscale, the interaction and strong correlation between electrons, and antiferromagnetic background. For the undoped samples, the MSC transition means the existence of intrinsic spin disorder with magnetic inhomogeneity. It is important that the present results give a direct evidence of Kondo scattering in ferromagnetic metallic manganites and prove that Kondo anomaly appears not only in metals containing small amounts of magnetic impurities but also in ferromagnetic conducting compounds containing spin-disorder clusters. This could be a general characteristic of the strongly correlated electron systems.

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 $: 75.20$.Hr, $75.30 - m$, $71.30 + h$, $71.27 + a$

Perovskite-type manganites have been extensively researched because of their rich physics contents in their behaviors of electrical transport and magnetic properties. These would be very important in clarifying the physical mechanism of strongly correlated electron systems and colossal magnetoresistance (CMR) effect.^{1,2} Among these physical properties, the strong correlations among spin, charge, orbital, and lattice degrees of freedom play important roles. The Kondo effect is one kind of famous and important physical phenomenon, which was found in metals containing small amounts of magnetic impurities and explained successfully in 1964.³ Heretofore, the Kondo phenomenon was only found in dilute magnetic metals, quantum dots, and heavy electron compounds.4 In recent years, it again caught people's interest and recognition because the Kondo phenomenon was found in quantum dot systems by many researchers.^{5–9} At the same time, the resistance minima, one of the most direct evidences for the Kondo effect, have been found at higher temperatures in structurally disordered magnetic and nonmagnetic metallic glasses as well as in compositionally disordered concentrated crystalline metallic alloys like Fe-Ni, Cu-Mn, and $(La, Pd_3)_8Mn$ etc.^{10–16} They were explained in terms of weak localization, electron-electron interaction, inelastic scattering of electrons, and uncontrolled magnetic impurities in the initially used nitrates, and so on. I^{7-20} In fact, its mechanism is still a puzzle and is under debate. As a strongly correlated electron system, the CMR manganites are one of the most interesting and widely noticed objects in the present physical and materials science based on its strongly electron correlation, ordering, and correlation effect of spin/charge/orbital, including lattice degree of freedom.5 So far its physical mechanism has not been understood because the electrical and magnetic behaviors cannot yet be explained perfectly. In this work, we present the results of Kondo-like transport in manganites observed by electrical and magnetic measurements. It is proved that Kondo-like transport can be explained by the scattering of electron resulted from intrinsic spin-disorder and *e*-*e* interac-

tion. This kind of spin-disorder possibly reflects the existence of intrinsic magnetic inhomogeneity in these mixedvalent manganites. This also proves that the Kondo effect exists not only in metals containing small amounts of magnetic impurities but also in ferromagnetic conducting compounds containing small amounts of nonmagnetic impurities including the spin-glass clusters. At the same time, based on our observations, it is also shown that this Kondo-like behavior and/or the MSC transition at low temperatures would possibly be one of the general characteristics for the strongly correlated electron systems.

For the experimental samples, we considered the undoped La_{2/3}Ca_{1/3}MnO₃ and Y-doped $(La_{1-x}Y_x)_{2/3}Ca_{1/3}MnO_3$ (*x* $= 0.0, 0.1, 0.2,$ and 0.4) system, where Y has smaller ionic radius, 0.94 Å, than La, 1.15 Å, and there are evident effects on CMR properties due to Janh-Teller effect and spin-glass behavior caused by the phase separation. This is a typical example of spin-disorder system and is suitable for studying the physical mechanism and their relative changes.21 The above samples were prepared by a conventional solid-state reaction method. The quality of the materials were checked by x-ray diffraction (XRD) (18 kW D/max-2500 power diffractometer, Cu K_{α} radiation). The XRD patterns are shown in Fig. 1 for all the samples. It can be seen that these spectra can be well characterized by perovskite $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ phase structure, indicating all samples are in good single phase of orthorhombic structure. Electrical and magnetic measurements were carried out using the physical property measurement system (PPMS-9) and superconducting quantum interference device (SQUID) (Quantum Design). Electrical transport measurements were carried out using the conventional four-probe technique in the temperature range from 2.5–300 K under applied dc magnetic fields of 0 and 6.0 T. All experimental results are repeatable.

The temperature dependence curves of electrical resistivity for $(La_{1-x}Y_x)_{2/3}Ca_{1/3}MnO_3$ (*x*=0-0.4) system are shown in Fig. 2 in the temperature range of 2.5–300 K under zero and an applied magnetic field of 6.0 T, respectively. It can be

FIG. 1. The experimental results (I vs 2θ) of x-ray diffraction for $(La_{1-x}Y_x)_{2/3}Ca_{1/3}MnO_3$ samples.

seen that the resistivity has a peak for all the samples indicating a metal-insulator (MI) transition. As expected, the peak temperature T_p at which MI transition occurs sharply decreases with increasing Y-doping content. Under zero field, T_p decreases from about 276 K for $x=0$ to 83 K for $x=0.4$ as shown in the inset of Fig. $2(a)$. The corresponding magnetoresistivity (MR) are shown in Fig. 2(b), which are the general characteristics of the mixed-valent manganites where

FIG. 2. (Color online) Temperature dependence of the electrical transport properties of $(La_{1-x}Y_x)_{2/3}Ca_{1/3}MnO_3$ in the range of 2.5– 300 K at zero and an applied field of 6.0 T. The inset shows the peak temperatures T_p corresponding to the MI transition as a function of Y content *x*.

FIG. 3. (Color online) The electrical resistivity behavior at low temperatures on an enlarged scale from 2.5 K to 30 K, where a reduced resistivity is used and the solid lines correspond to the fit using Eq. (1). The insets show the electrical resistivity in the range of 2.5–60 K in zero field.

CMR effect appears. Figure 1 and Fig. 2 indicate the high reliability and typical behavior of the samples.²¹ However, on the whole, from Fig. 2 we cannot evidently see any change or abnormal in the electrical transport at low temperatures. But when we take a look in detail at the resistivity curve in the low temperature region, some interesting phenomenon were found. Figure 3 shows the electrical resistivity behavior at low temperature on an enlarged scale, from 2.5 K to 30 K. For all the samples, we observed the electrical resistivity minimum phenomenon appearing below 30 K. In order to see the effect of the applied magnetic field, a reduced resistivity is used by dividing the resistivity at the lowest experimental temperature of 2.5 K in Fig. 3. The insets of Fig. 3 show the electrical resistivity behavior by unreduced units in an enlarged scale from 2.5 K to 60 K for field free samples. From Fig. 3, it is found that, for all samples, the electrical resistivity exhibits a minimum in the range of 20–30 K under zero field and 10–20 K under an applied field of 6.0 T. The resistivity-temperature curve at 6.0 T is higher than that under zero field. It is believed that an applied magnetic field tunes the transport effect. As is well known, the minimum of electrical resistivity at low temperatures are the typical characteristics of the Kondo effect.³ According to the original observation and Kondo theory in dilute magnetic alloys, the Kondo behavior is explained as the interaction between localized spins of magnetic impurities and the conduction electrons in a metal. However, the current perovskite manganites are completely different from the dilute magnetic alloys. In the temperature region of *T* T_c , they are ferromagnetic and show metallic conducting properties. Thus, there may be a completely different mechanism from that of the dilute magnetic alloys, which is the reason why we call it "Kondo-like transport." According to Kondo's theory, in the Kondo region, the spin-dependent electrical resistivity can be expressed formally as $\rho = \rho_0$ $-\rho_s \ln T$. The parameter ρ_0 is the residual resistivity, which is contributed by elastic scattering, e.g, electron-impurity scattering and coulomb interaction (CI). The second term, $\rho_s \ln T$,

is the contribution from the interaction between localized magnetic movement and the conduction electrons. The parameter ρ_s is mainly decided by the intensity of the spin scattering. Considering the lattice contribution, i.e, *e*-*p* interaction, the form of T^5 should be added into the account by the phonon scattering item in the low temperature region. It is said that the low-temperature resistivity behavior should have the formal $\rho(T) = \rho_0 - \rho_s \ln T + \rho_p T^5$ form. But as was mentioned before, the strongly correlated manganites are different from the dilute magnetic alloys. We failed to fit all the curves in Fig. 3 by this formula since it can only describe the electrical resistivity behavior below 15 K. It is believed that the above formula is not enough for the present strongly correlated manganites. In fact, the temperature dependence of the resistivity minimum was also observed in the similar weakly disordered systems and amorphous alloys, where such behavior is attributed to the *e*-*e* interference scattering from static inhomogeneity in three dimensions. By considering more general situations, we should take into account the e - e interaction. According to the theory of Lee,¹⁴ the e - e interaction has a temperature dependence of $T^{1/2}$. Thus, in the end, we fit the present electrical resistivity data at low temperatures in the wide range of 2.5–30 K region by the equation of the form

$$
\rho(T) = \rho_0 - \rho_s \ln T + \rho_e T^{1/2} + \rho_p T^5. \tag{1}
$$

The fitting results are drawn as solid lines for 0 and 6.0 T in Fig. 3. It is seen that all the data are fitted very well.

In Eq. (1) , the various coefficients correspond to the residual resistivity, spin scattering, *e*-*e* interaction, and *e*-*p* interaction, respectively. To see these directly, we plot the absolute values of these parameters in Eq. (1) against the Y content *x* of $(La_{1-x}Y_x)_{2/3}Ca_{1/3}MnO_3$ in Fig. 4. It can be seen that all the parameters exhibit a strong dependence on Y content *x* and they can be tuned with an applied magnetic field. The residual resistivity ρ_0 increases with increasing Y content *x*, which shows that Y substitution induces some disorder and/or impurity and increases the scattering of electron by disorder and CI. From the field-dependent relation, the value of ρ_0 is higher at 0 T than those at 6.0 T. Hence, the applied magnetic field makes the electrical resistivity decrease for Y substitution. This is different from the general conductors. It is well known that, in good conductors, ρ_0 would not depend on temperature *T* and magnetic field *H*. In fact, this does reflect CMR effect and strong disorder characteristic for manganite system. From Fig. 4(d), it is found that the *e*-*p* interaction is also sensitive to Y doping and the applied field. But it has an opposite behavior with magnetic field, i.e, all the values of ρ are higher at 6.0 T than those at 0 T. The increased magnetic order at an applied field would enhance the coupling and interaction between the electrons and the lattice background. This will increase the localization of conduction electrons and make the ρ_p increase at higher field of 6.0 T. This suggests that there exist interactions of electron with antiferromagnetic background and the lattice field with spin-glass disorder in manganic matrix. For ρ_s and ρ_e , they show a similar behavior with Y content *x* and the applied magnetic field. They increase with increasing Y-doping content x and the curve at 0 T is above the one at

FIG. 4. (Color online) The dependence of the fitted parameters on Y content *x* at 0 and 6.0 T, respectively, for (La_{1-*x*}Y_{*x*})_{2/3}Ca_{1/3}MnO₃.

6.0 T, i.e., for the same *x*, their values at the zero field are larger than those at 6 T. This reflects directly the effect of magnetic field and the origin of magnetic scattering for the charge carriers and *e*-*e* interaction. This may be the reason why spin scattering has not been noticed previously. It should be noticed that the spin-disorder degree is related with *x* as being described below. Figure 4 and the fitted values of ρ_s reflect a consistency between the current experiments and Kondo's theory, i.e, the present electrical transport behavior show the presence of Kondo-like transport. It is well known that, recently, the *e*-*e* interaction is the most popular explanation to the resistivity minima in the manganites but the spin disorder scattering was hardly being noticed. And then, it should be mentioned that the positive ρ_e value should be unexpected. According to *e*-*e* interaction, it should have a negative value. In fact, before this result is obtained, we even tried to fit these curves in the framework of the single e - e interaction by removing $\rho_s \ln T$ item in Eq. (1). However, the fitting cannot be done well although a negative ρ_e coefficient was obtained for all samples. It is said that, when we consider the Kondo and the *e*-*e* interaction at the same time, a positive ρ_e appears. This means possibly an existence of new interaction by strong correlation coincidence characteristics between the electrons in manganites. It would be more important and proves a possible existence of a new mechanism other than a simple *e*-*e* interaction as a conventional conductor. And recently, the Kondo effect was also found by Pasupathy *et al.* in ferromagnetism materials.25

FIG. 5. (Color online) Experimental results for spin-glass disorder of the sample with $x=0.4$; (a) dc magnetization and (b) ac susceptibility for various reference frequencies. The peak moves to higher temperatures from 42.71 K at 21 Hz to 43.75 K at 6 kHz as shown in the inset.

In the present paper, we will only focus our attention to the role of spin-disorder scattering in the low-temperature electrical transport behavior.

In order to understand the interesting change of the spin scattering strength shown by the parameter ρ_s and the physical mechanism of Kondo-like transport in ferromagneticphase manganites, we studied the magnetic properties as a function of Y doping *x*. Temperature dependence of dc magnetization was measured and the results are shown in Fig. 5(a). It can be seen that, there were two small steps below 50 K, as shown by the arrow marks on the M-T curves for *x* $= 0$ and 0.1. For the samples with $x = 0.2$ and 0.4, peaks appear at lower temperatures, about 35 K. For the sample with $x=0.4$, we observe the detailed changes in ac susceptibility and dc magnetization for zero-field-cooled (ZFC) and fieldcooled (FC) conditions under an applied field of 1000 Oe. The results are shown in Fig. $5(b)$. From the inset of Fig. 5(b), it can be seen that the two dc magnetization curves under ZFC and FC conditions are bifurcated at low temperatures, which are the typical characteristics of the possible existence of a spin-glass or antiferromagnetic phase. Correspondingly, for samples with $x=0$ and 0.1, a small step in the dc magnetization possibly shows only the appearance of a little amount of spin-glass or antiferromagnetic phase. Especially, the existence of spin-glass or antiferromagnetic phase in undoped samples should reflect a kind of intrinsic magnetic inhomogeneity in the perovskite manganites. In dc magnetization curves, one can find that the strongly correlated manganites are a kind of dirty compounds with intrinsic inhomogeneity characterized by the coexistence of the magnetic multiphase. In order to distinguish the spin-glass from antiferromagnetism, the ac susceptibility measurement is meaningful by changing ac reference frequency. Figure $5(b)$ gives the ac susceptibility curves for various reference frequencies, from 21 Hz to 6 kHz for the sample with $x=0.4$. We find that the peaks of the ac susceptibility curves move a little to higher temperatures from 42.7 K at 21 Hz to 43.8 K at 6 kHz, which is plotted in the inset of Fig. $5(b)$. This is a typical characteristic for the spin-glass state and the peak temperature T_f is generally defined as the freezing temperature of the spin-glass phase. This phenomenon is consistent with the canonical spin-glass systems.²² This result indicates that a phase separation appears with the coexistence of the ferromagnetic and the spin-glass phases. The spin-glass phase is an aggregate of spin-disorder clusters with nanosize and/or microsize. The ac susceptibility above shows an increasing of spin-glass phase when Y doping *x* increases, indicating that the spin-disorder degree of freedom increases with increasing *x*. These are also consistent with the results reported by Zhang²⁶ and Teresa *et al.*²⁷ Thus *x* can be considered as a measure of the spin disorder. Combined with the change of the electrical resistivity, this proves that the spin scattering strength increases as *x* increases. This provides a kind of possible explanation to the physical mechanism underlying the Kondo-like transport in perovskite manganites. Based on the fact of the increase of spin-glass phase with Y doping *x*, Kondo-like transport should be the result of spin disorder scattering in ferromagnetic manganites at low temperatures, which gives a new evidence for the explanation of the Kondo-like transport in manganites.

In fact, three aspects should be emphasized. Firstly, we cannot exclude the possibility of the existence of an antiferromagnetic phase based on the ac susceptibility data. It is well known that mix-valence manganites have antiferromagnetic ground state. Therefore, various nonequilibrium fluctuations, including heat and stoichiometry, etc., will induce inhomogeneity. The antiferromagnetic phase could be the first possible origin of the intrinsic magnetic inhomogeneity. This is the reason for presuming the intrinsic coexistence of ferromagnetic, spin-glass, and/or antiferromagnetic clusters in undoped and Y-doped systems. Secondly, from the electrical resistivity data, it is also reasonable to presume that there exist spin-glass and/or antiferromagnetic clusters, especially in the undoped system. Here, the spin-glass peak is not evidently observed in the dc magnetization and ac susceptibility measurements, perhaps due to its small amount and the limit of experimental resolution. The high ferromagnetic/spinglass (or antiferromagnetic) ratio should also suppress the peak effect of the spin-glass phase. But the two small steps are possibly reflections of the spin-glass and/or antiferromagnetic phase in dc magnetization curves shown in Fig. $5(a)$ for $x=0$ and 0.1. Therefore, for the sample with $x=0$, this Kondo-like behavior should reflect the existence of a spin-glass and/or antiferromagnetic phase in the undoped sample, a kind of intrinsic magnetic inhomogeneity. This intrinsic inhomogeneity has the characteristic of a phase separation, which is similar to the charge-ordered phase separation, and intrinsic inhomogeneity of nanometer size.²³ Recently, Ahn *et al.* also found the coexistence of distinct metallic and insulating electronic phases within the same sample. This is considered to be due to the nanometer and micrometer scale inhomogeneity showing up in chemical disorder, multiscale coexistence, and multiphase coexistence.²⁴ In this framework, the intrinsic inhomogeneity consists of some spin-glass clusters in a size of nanometer scale with limited number of atoms. Thirdly, all of our measurements on many manganites, including undoped $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$, $\text{Sm}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$, and a series of various doped manganites, show that the Kondo-like behavior and M-SC transition are possibly a general characteristic of the strongly correlated manganites. For the physical mechanism, the conducting carriers are scattered by spin disorder and/or antiferromagnetic clusters and produce Kondo behavior similar to the classical Kondo effect in dilute magnetic alloys. However, the present Kondo-like transport appears in strongly correlated manganites with ferromagnetic phase and thus it is completely different from that of the dilute magnetic alloys. In the present experiments, a new kind of Kondo behavior is suggested in the ferromagnetic perovskite manganites. This also proves that the Kondo anomaly appears not only in metals containing small amounts of magnetic impurities but also in ferromagnetic conducting compounds containing small amounts of spin-glass and/or antiferromagnetic impurities. Here, the spin-glass and/or antiferromagnetic clusters imply the disorder of spins and the increase of spin-glass and/or antiferromagnetic cluster contents, showing the degree of spin disorder. Thus, this Kondolike transport is the result of the spin-disorder scattering in an intrinsic magnetic inhomogeneous system and the *e*-*e* interactions at low temperature. This kind of intrinsic magnetic inhomogeneity provides a new physical understanding for various experimental results.

In summary, the Kondo-like behavior is found in the perovskite manganites at low temperatures and they can be best fitted in the framework of Kondo scattering, *e*-*e* interactions, and *e*-*p* interactions. The present study shows that the Kondo-like behavior depends strongly on the amount of the spin-glass phase and can be tuned with applied magnetic field, resulting from the scattering of conduction electrons by spin-disorder clusters in nano scale and/or micro scale. These results reflect the intrinsic spin-disorder and magnetic inhomogeneity with the characteristics of a phase separation, which is similar to the charge-order phase separation. With increasing Y content *x*, the spin-disorder degree of freedom increases and causes strong scattering of the carriers. The applied field can decrease the spin-disorder. From the measurements on several manganite systems, including undoped $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$, $\text{Sm}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$, and a series of doped manganites, it is proved that this Kondo-like behavior and the M-SC transition is possibly a general characteristic for the strongly electron-correlated manganite systems. The present results show that the Kondo behavior appears not only in metals containing small amounts of magnetic impurities but also in ferromagnetic conducting compounds containing small amounts of spin-disorder clusters.

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