Fe-doping-induced evolution of charge-orbital ordering in a bicritical-state manganite

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Impurity effects on the stability of a ferromagnetic-metallic state in a bicritical-state manganite, (La0.7Pr0.3)0.65Ca0.35MnO3, on the verge of metal-insulator transition have been investigated by substituting a variety of transition-metal atoms for Mn ones. Among them, Fe doping exhibits the exceptional ability to dramatically decrease the ferromagnetic transition temperature. Systematic studies on the magnetotransport properties and x-ray diffraction for the Fe-doped crystals have revealed that charge-orbital ordering evolves down to low temperatures, which strongly suppresses the ferromagnetic-metallic state. The observed glassy magnetic and transport properties as well as diffuse phase transition can be attributed to the phase-separated state where short-range charge-orbital-ordered clusters are embedded in the ferromagnetic-metallic matrix. Such a behavior in the Fe-doped manganites form a marked contrast to the Cr-doping effects on charge-orbital-ordered manganites known as the impurity-induced collapse of charge-orbital ordering.

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I. INTRODUCTION

In hole-doped perovskite-type manganites, such as R1−xA1MnO3 (R being a trivalent rare-earth ion and A being a divalent alkaline-earth ion), a ferromagnetic-metallic (FM) and a charge-orbital-ordered antiferromagnetic insulating (CO/OO) phase coexist with each other. Complex states of matter and critical changes among them, such as colossal magnetoresistance and magnetic-field-induced insulator-metal transition,1,2 manifest themselves in a bicritical region, where such competing phases are almost degenerate in free energy and separated by a first-order phase transition line. Quenched disorder arising from local lattice distortions and/or dopant ions dramatically modifies the electronic structure near the bicritical point.3–8 One typical example of moderate disorder is the solid solution of the perovskite A site with R and A ions (of small and large ionic radii, respectively), which is a well-known and widely used technique to control the band filling (hole doping level) and the one-electron bandwidth of the system.5 Randomly distributed R and A ions result in the local lattice distortions and the random Coulomb potential, which can cause the suppression of long-range-ordered state of both FM and CO/OO.9,10 Consequently, a homogeneous spin-glass insulating phase dominates in the bicritical region when the mismatch of ionic radii between R and A ions is appreciable.12–14

As another typical example of strong disorder, effects of impurity doping onto Mn sites have been intensively studied so far. In particular, an impact of Cr doping in half-doped charge-orbital-ordered (CO/OO) manganites has attracted much attention for past years.15–19 Only a few percent substitution of Cr for Mn dramatically suppresses the long-range CO/OO state and makes the system (partly) ferromagnetic metallic. Since Cr3+ is quite stable in valence, it may serve as the immovable eferm charge-orbital deficiency in the CO/OO state and locally induce the competing FM state as the alternative, which leads to inhomogeneous phase separation with both CO/OO and FM clusters coexisting on various length scales.20 Other transition metals can also induce metallicity and ferromagnetism partially or completely in CO/OO manganites, depending on the kind of doping species.21–24 Ru, Ni, or Co doping, for example, easily suppresses the pristine CO/OO state and effectively stabilizes the FM state, as in the case of Cr doping. For Fe, Ga, or Al doping, on the other hand, no clear FM transition shows up in the absence of a magnetic field, but only a spin-glass-like (insulating) state is formed. It was suggested that the electronic configuration of impurity ions may cause such a difference in the behavior of destroying the CO/OO state.21,22

In this study, from the contrastive point of view, we have investigated effects of the disorder on the FM state by doping various transition-metal elements onto Mn sites. Although impurity effects on many kinds of FM manganites were studied previously,25–30 we focused here on the FM state of single-crystal (La0.7Pr0.3)0.65Ca0.35MnO3, which locates near the phase boundary to the CO/OO, to reveal the disorder effects on the FM phase near the bicritical point as complementary to those on the CO/OO phase. The adopted compound (La0.7Pr0.3)0.65Ca0.35MnO3 is close to the bicriticality but least affected by the disorder effect arising from the A-site solid solution and hence can provide the ideal arena to highlight the genuine B-site doping effect on the FM state. The stability of the FM state against impurity doping has been observed to strongly depend on the kind of dopants, as in the case of the CO/OO state. Among all the dopants, Fe dopants most effectively decrease the FM transition temperature, TC (by ~70% for 5% doping). In the Fe-doped manganites, we have observed the evolution of short-range charge-orbital ordering down to low temperatures, which strongly suppresses the FM state. Such a tendency, which was revealed in this purposely designed compound but should be generic for the FM state in the bicritical-state manganites, markedly contrasts with the Cr-doping effects on the CO/OO manganites.
II. EXPERIMENT

Single crystals of \((La_{1−x}Pr_x)_{0.65}Ca_{0.35}MnO_3\) \((0.2≤x≤0.8)\) and \((La_{0.7}Pr_{0.3})_{0.65}Ca_{0.35}Mn_{1−y}Fe_{y}O_3\) \((M=Fe, Cr, Ga, and Ru, 0≤y≤0.1)\) were grown by the floating zone method. Mixed powders of \(La_2O_3, Pr_6O_{11}, CaCO_3, Mn_3O_4, \alpha-Fe_2O_3, Cr_2O_3, Ga_2O_3, \) and \(RuO_2\) in stoichiometric proportions were first calcined at 1000–1050 °C for 10–20 h in air. The mixture was pulverized and again sintered at 1200–1250 °C for 30–40 h in air. The resulting powders were pulverized and then pressed into a rod with \(5 \text{~mm}\) in diameter and \(60 \text{~mm}\) in length. The rod was fired at 1350–1400 °C for 30–40 h in air. The crystal growths except for \(M=Ru\) were performed in an oxygen atmosphere with rotating the feed and seed rods in opposite directions at the rate of 15–20 rpm while the growth for \(M=Ru\) in air. The growth rate was set at 2–2.5 mm/h. Electron probe microanalysis revealed that the beginning part of the grown crystal rod has the composition variation along the growth direction, probably due to the chemical instability of the molten zone in an early process of the crystal growth. The middle and end parts, on the other hand, have a homogeneous composition, from which we have prepared the samples for all the measurements performed in this study.

Furthermore, inductively coupled plasma (ICP) spectroscopy on the obtained samples has shown that their composition is equal to the prescribed ratio with an accuracy of ±0.007 and ±0.003 for \(x\) and \(y\), respectively. For a Ru-doped compound, however, we had only a 2%-Ru-doped crystal of \((La_{0.7}Pr_{0.3})_{0.65}Ca_{0.35}Mn_{0.98}Ru_{0.02}O_3\) due to the high volatility of Ru oxides, although we prescribed the ratio for 5% doping in the mixed powders. The powder x-ray diffraction patterns showed that the obtained crystals are of single phase and that the crystal structure is orthorhombic at room temperature with \(a_0 ~ b_0 ~ c_0 / √2 ~ √2 a_p\), where \(a_p\) is the lattice parameter of the pseudocubic lattice. Because the orthorhombic distortion is small and the twin domains equally exist, we here employ cubic notation for simplicity. For several crystals, the synchrotron single-crystal x-ray diffraction was performed, using an imaging plate system on the beamline BL-1A of the Photon Factory, High-Energy Accelerator Research Organization (KEK), Japan. Magnetization was measured with a superconducting quantum interference device magnetometer. Resistivity was measured by a conventional four-probe method with electrodes formed by heat-treatment-type silver paint.

III. RESULTS AND DISCUSSION

A. Bicritical features in \((La_{1−x}Pr_x)_{0.65}Ca_{0.35}MnO_3\) crystals

We first show the bicriticality in \((La_{1−x}Pr_x)_{0.65}Ca_{0.35}MnO_3\), which is the mixed crystal system of FM \(La_{0.65}Ca_{0.35}MnO_3\) and CO/OOI \(Pr_{0.65}Ca_{0.35}MnO_3\). Figure 1 shows the temperature profiles of (a) resistivity at \(μ_0H=0\) T and (b) magnetization at \(μ_0H=0.5\) T for single crystals of \((La_{1−x}Pr_x)_{0.65}Ca_{0.35}MnO_3\) \((0.2≤x≤0.8)\). With increasing \(x\), the FM transition accompanying a sharp drop of resistivity systematically decreases in \(T_C\) and the CO/OOI state, which is manifested at around 200 K by a steep increase in resistivity or decrease in magnetization, shows up for \(x≥0.4\). The reentrant insulator (CO/OOI)–metal (FM) transition is observed with large temperature hysteresis for intermediate doping levels, \(x=0.4\) and 0.5. Finally, the CO/OOI phase dominates the region of \(x≥0.6\) and the charge-orbital ordering temperature, \(T_{C0D}\), gradually increases as \(x\) increases. For \(x=0.6\), the insulating low-temperature resistivity shows a small drop even at zero magnetic field, as is barely visible in a warming run. This stems from the fact that even a tiny disorder or inhomogeneity in the sample could induce a small fraction of the FM state in the long-range CO/OOI phase since the \(x=0.6\) crystal is just on their phase boundary at the ground state.

Figure 2 summarizes the electronic phase diagram for the \((La_{1−x}Pr_x)_{0.65}Ca_{0.35}MnO_3\) \((0≤x≤1)\) crystals. \(T_c\) is determined as the temperature where the resistivity curve in Fig. 1(a) shows a steep drop, while \(T_{CO}\) is where the \(d\ln ρ/dT\) curve shows a cusplike maximum. For clarity, only the transition temperatures in a cooling process are shown in Fig. 2. The FM phase is replaced with the CO/OOI phase with increasing \(x\). Note that the FM region stretches out in the CO/OOI region below ~150 K, reflecting the FM reentrant transition as observed for \(x=0.4\) and 0.5 in Fig. 1. The end point of the FM phase is around \(x=0.6\). As a result, the phase diagram for the \((La_{1−x}Pr_x)_{0.65}Ca_{0.35}MnO_3\) system exhibits an almost ideal bicritical feature arising from the competition between the long-range CO/OOI and FM, as previously reported for \(Pr_{0.65}(Ca_{1−x}Sr_{x})_{0.35}MnO_3\). Because the mismatch of the ionic size between trivalent rare earth (La and Pr) and divalent alkaline earth (Ca) is the smallest among a series of \(K_{0.65}A_{0.35}MnO_3\), the quenched disorder arising from the A-site solid solution can be kept minimal in magnitude to maintain either of the long-range orders in the vicinity of the bicritical point for this system.
We can now adopt \( (\La_{1-x} \Pr_x)_{0.65} \Ca_{0.35} \Mn O_3 \) as one of the most ideal and typical parent compounds for investigating the impurity doping effects on the bicritical regime. Hereinafter, to show the effects on the FM states close to the critical point, we restrict ourselves to the case of \( x=0.3 \) (FM), as denoted with the vertical arrow in Fig. 2. We have partially substituted the Mn sites with various transition-metal elements as a strong quenched disorder in this system. All the crystals investigated here are single crystals to exclude the effects from the extrinsic disorder such as grain boundaries and crystal lattice defects, which often dominates the properties in polycrystalline crystals.34

B. Stability of ferromagnetic-metallic state against a variety of impurity species

Figure 3 shows the temperature dependence of the resistivity for doped FM manganites, \( (\La_{0.7} \Pr_{0.3})_{0.65} \Ca_{0.35} \Mn_{0.99} \Mn_{0.01} O_3 \) (\( M = \Mn, \Fe, \Cr, \text{and} \Ga \)) and \( (\La_{0.7} \Pr_{0.3})_{0.65} \Ca_{0.35} \Mn_{0.98} \Ru_{0.02} O_3 \). The pristine \( (\La_{0.7} \Pr_{0.3})_{0.65} \Ca_{0.35} \Mn O_3 \) is FM at the ground state, as seen by the presence of a steep drop at \( T_C \sim 195 \text{ K} \) in the resistivity curve. In the Fe-doped crystal, even for only 5% doping, the FM state is strongly destabilized and \( T_C \) decreases to \(~66 \text{ K} \) where the resistivity drops by \(~6 \text{ orders of magnitude} \) for 5% doping of Cr and Ga, however, impurity effects are much smaller and \( T_C \) is slightly lowered to \(~172 \text{ K} \) and \(~187 \text{ K} \), respectively. Note that the changes in the carrier density due to doping are almost the same among the above 5% doped crystals since the trivalent states are stable for all the dopant species. The electronic (spin) configurations, however, are different, such as \( t_{2g}^2 e_g^2 \) (\( S=5/2 \)), \( t_{2g}^2 e_g^0 \) (\( S=3/2 \)), and \( t_{2g}^2 e_g^e \) (\( S=0 \)) for Fe\(^{3+} \), Cr\(^{3+} \), and Ga\(^{3+} \), respectively, which could play an important role in destabilizing the FM state. The Ru-doped crystal, on the other hand, exhibits even a slight increase in \( T_C \), as is often observed in other FM manganites.35–37

Inset in Fig. 3 summarizes shifts in \( T_C \) versus the doping level for the above crystals.

C. Transport and magnetic properties

We show in Fig. 4 the temperature profiles of (a) resistivity and (b) magnetization for the crystals of \( (\La_{0.7} \Pr_{0.3})_{0.65} \Ca_{0.35} \Mn_{1-y} \Fe_y O_3 \) (\( 0 \leq y \leq 0.1 \)). Measurements for the resistivity and magnetization were carried out at \( \mu_0 H=0 \text{ T} \) and 0.5 T, respectively, in both cooling and warming runs. The magnetization data measured in a warming run after cooling the samples at \( \mu_0 H=0 \text{ T} \) (zero-field cooling) are denoted with the open circles, while those measured after cooling them at \( \mu_0 H=0.5 \text{ T} \) (field cooling) are with the close circles. A few percent substitutes of Fe effectively suppress the FM state and drastically decrease \( T_C \). Hereinafter, we concentrate on systematic studies on Fe-doping effects by measurements of resistivity, magnetization, and x-ray diffraction.
FIG. 4. (Color online) Temperature profiles of (a) resistivity at $\mu_0H=0$ T and (b) magnetization at $\mu_0H=0.5$ T for crystals of $(La_{0.7}Pr_{0.3})_{1-y}Ca_{0.35}Mn_{1-y}Fe_2O_4$ ($0 \leq y \leq 0.1$). Measurements were performed in both cooling and warming runs. The magnetization data measured after the field-cooling (FC) and zero-field-cooling (ZFC) processes are denoted with the closed circles and the open ones, respectively.

\[ \sim 10^{-3} \text{\Omega cm}, \text{for homogeneous FM states. Therefore, this suggests that the observed insulator-metal transition is attributed to the percolation transition in the inhomogeneous system where the FM clusters randomly exist. In the crystals with } y \geq 0.07, \text{the FM transition completely disappears and the resistivity shows insulating down to the lowest temperature. The temperature profiles of magnetization for the } y \geq 0.07 \text{ crystals show the distinct history dependence between zero-field cooling and field cooling below } \sim 50 \text{ K, as shown in Fig. 4(b), which indicates that a spin-glass state becomes dominant at low temperatures for these crystals. Even in the crystals with } y=0.05 \text{ and 0.06, which show the FM transition, such a discrepancy between zero-field-cooled and field-cooled magnetizations is also observed below } \sim 50 \text{ K. This behavior reflects that the ground state for these crystals is not a homogeneous FM but an inhomogeneous mixture composed of the FM and CO/OO states, as evidenced by the diffraction study (vide infra).}

D. Single-crystal x-ray diffraction

The single-crystal x-ray diffraction measurements for the crystals with $y=0, 0.05,$ and 0.06 have revealed the close relation between the strong suppression of the FM state and the evolution of the CO/OO state. Figure 5(a) shows the x-ray diffraction profiles along the [110] direction around the (200) Bragg point at several temperatures, where we find a CO/OO-related superlattice reflection with a modulation vector of $\sim (1/4,1/4,0)$ in the cubic lattice setting. We also display in Fig. 5(b) the temperature dependence of the integrated intensity of the superlattice reflection for each crystal. For comparison, the corresponding resistivity curves are shown there with dashed lines. In the undoped crystal with $y=0$, the CO/OO-related superlattice peak shows up only just above $T_C$ and completely disappears below $T_C$. With the increase of $y$ from 0 to 0.05, the superlattice reflection intensity continues to increase to lower temperatures, but it suddenly decreases at $T_C$, which is in accord with the resistive transition. In the $y=0.06$ crystal, on the other hand, the superlattice peak is found to persist down to the lowest temperature, while the resistivity shows a steep drop around $T_C$, which suggests that both FM and CO/OO phases coexist at the ground state. Note that the correlation length of charge-orbital order as extracted from the superlattice peak width for $y=0.06$ is $\sim 20 \text{ Å}$, indicating that the CO/OO state survives with only a short-range correlation, such as in a form of clusters embedded in the FM matrix.

With the further increase of Fe-doping level ($y \geq 0.07$), the clear CO/OO superlattice reflection gradually disappears with the broadened peak width, while the diffuse scattering (Huang scattering) extending in the direction of [110] and [110] around the Bragg peak becomes dominant. Such diffuse scattering arises from the less-correlated polarons associated with the Jahn-Teller distortion due to the localization of carriers on Mn sites, which can be regarded as the remnant of the CO/OO superlattice spot. In fact, the tem-
The phase-separated (PS) state, or cluster glass (CG), where the ferromagnetic-metallic (FM) clusters and the short-range charge-orbital-ordered (SR CO/OO) clusters coexist, appears around $y \sim 0.06$. With further increasing $y$, it changes to the homogeneous spin-glass (SG) with atomic-scale correlation. The transition temperature to the FM, CO/OO, and SG (CG) phases is denoted with closed circles, squares, and triangles, respectively. The onset temperature of SR CO/OO, determined by the integrated intensity of x-ray superlattice reflection, is denoted with open squares.

E. Phase diagram for Fe-doped $(La_{1-x}Pr_x)_{0.65}Ca_{0.35}MnO_3$

Figure 6 shows the electronic phase diagram for $(La_{0.7}Pr_{0.3})_{0.65}Ca_{0.35}Mn_{1-y}Fe_yO_3$ $(0 \leq y \leq 0.1)$ combined with that for $(La_{1-x}Pr_x)_{0.65}Ca_{0.35}MnO_3$ $(0 \leq x \leq 1)$. The transition temperature to the spin glass (SG) is determined as the temperature below which the magnetization shows a history dependence between field cooling and zero-field cooling. The onset temperature of short-range (SR) CO/OO is estimated from Fig. 5. In the pristine crystal of $(La_{1-x}Pr_x)_{0.65}Ca_{0.35}MnO_3$, which locates near the phase boundary to the CO/OOI, the ground state is a homogeneous FM. The SR CO/OO manifests itself only in a narrow temperature range just above $T_C$ and suddenly vanishes below $T_C$. As $y$ increases, the FM state is suppressed with a steep decrease in $T_C$ and the SR CO/OO instead evolves down to lower temperatures. The onset temperature of SR CO/OO shows no significant change regardless of $y$ and hence may be interpreted as a Griffiths temperature. At around $y \sim 0.06$, the clear FM transition disappears and the ground state becomes an inhomogeneously phase-separated one, where the FM and CO/OO clusters coexist even at the lowest temperature, which can often be regarded as cluster glass (CG). With further increasing $y$, however, the correlation length of the CO/OO state as well as the FM state becomes shorter due to the strong randomness from highly concentrated Fe doping. Near $y \sim 0.1$, finally, the homogeneous spin-glass insulating phase with only an atomic-scale correlation prevails below $\sim 50$ K.

As shown above, Fe doping has a large impact on the FM state; it effectively weakens the FM state and instead develops the CO/OO state down to low temperatures. In the CO/OO state, however, Fe substitution seems to play only minor roles. Although a small fraction of the FM state could be induced at low temperatures for a tiny doping of Fe ($\sim 2\%-4\%$) in some cases, it is easily extinguished with further doping (up to as low as $\sim 4\%-7\%$). These suggest that Fe substitution selectively hinders the FM correlation rather than the CO/OO one. Such a tendency contrasts strikingly with that in the Cr-doping effects on the bicrystalline-state manganites. A few percent substitutes of Cr in the CO/OO manganite strongly destroy the long-range CO/OO and induce the FM state, as investigated intensively. Our study reveals the contrastive feature that Cr doping only has small effects on the FM state while slightly decreasing $T_C$.

In the following, we focus on the $y=0.07$ crystal with the phase-separated ground state in which the FM conducting matrix is barely disconnected by the short-range CO/OO clusters (see Figs. 4 and 6), that is, the volume fraction of the FM state will be just below the percolation threshold. We show in Fig. 7(a) the temperature profiles of resistivity for the $y=0.07$ crystal at various external magnetic fields. At zero field, the resistivity shows an insulating behavior down to the lowest temperature. The application of a magnetic field...
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of a few tesla, however, drastically reduces the resistivity at low temperatures, making the system metallic. More noteworthy is that the value of the residual resistivity varies in a very wide range depending on the magnitude of the magnetic field. We have further investigated such a multistable behavior by measuring the resistivity and magnetization at the lowest temperature of 2 K by changing the strength of the magnetic field, as shown in Figs. 7(b) and 7(c), respectively. For this measurement with the procedure of “magnetic-field annealing,”18,51 we first apply various magnetic fields \( H \) at room temperature and then cool the sample down to 2 K. Subsequently, we sweep the magnetic field between \( +H \) and \( -H \), as displayed in the inset of Fig. 7(a). The resistivity dramatically decreases by more than 5 orders of magnitude with increasing \( \mu_0H \) from 0 to 9 T, although those at \( \mu_0H \approx 2 \) T are too high to measure. Once after a temperature is lowered down to 2 K at \( H \), the values of the resistivity are little changed (on such a logarithmic scale) even after the magnetic field is removed, memorizing the history of the annealing process. In the corresponding magnetization curves, furthermore, the saturation moment steadily increases with the increase of \( H \). This systematic change in the resistivity and magnetization can be attributed to the change in the volume fraction of the CO/OO clusters embedded in the FM matrix. A magnetic-field annealing at \( \mu_0H \gg 7 \) T sufficiently suppresses the CO/OO clusters at the ground state, making the system almost homogeneous FM. In this way, we can easily control the volume fraction of the CO/OO clusters in the phase-separated state by changing the magnitude of \( H \).

Analogous glassy magnetotransport properties or relaxor ferromagnet phenomena have been reported for Cr-doped CO/OO manganites,18,51 where the FM phase is locally induced in the CO/OO matrix conversely to the present case. Thus, the phase-separated states, whose respective phase volume can be critically controlled by magnetic fields, are generated from the two very contrastive ordered states, FM and CO/OO, in the bicritical-state manganites.

**IV. CONCLUSION**

In conclusion, we have investigated the impurity doping effects on a ferromagnetic-metallic state in single crystals of \((\text{La}_{0.7}\text{Pr}_{0.3})_{0.85}\text{Ca}_{0.35}\text{Mn}_{1-y}\text{Fe}_y\text{O}_3\) (\( y = 0.07 \)) crystal. Magnetic-field dependence of \( \rho \) and \( M \) at 2 K after the magnetic-field annealing procedures for the \( y = 0.07 \) crystal. The measurements were performed once the magnetic fields were applied at room temperature and the crystal was cooled down to 2 K at the respective annealing fields, as depicted in the inset.

![Figure 7](image_url)

**FIG. 7.** (Color online) (a) Temperature profiles of resistivity at various magnetic fields for the \((\text{La}_{0.7}\text{Pr}_{0.3})_{0.85}\text{Ca}_{0.35}\text{Mn}_{1-y}\text{Fe}_y\text{O}_3\) \( (y = 0.07) \) crystal. Magnetic-field dependence of (b) resistivity and (c) magnetization at 2 K after the magnetic-field annealing procedures for the \( y = 0.07 \) crystal. The measurements were performed once the magnetic fields were applied at room temperature and the crystal was cooled down to 2 K at the respective annealing fields, as depicted in the inset.
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