Cr-doping effect on the orbital fluctuation of heavily doped Nd$_{1-x}$Sr$_x$MnO$_3$ \( (x \approx 0.625) \)

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We have investigated the Cr-doping effect of Nd$_{0.375}$Sr$_{0.625}$MnO$_3$ near the phase boundary between the \( x^2 - y^2 \) and \( 3z^2 - r^2 \) orbital ordered states, where a ferromagnetic correlation and concomitant large magnetoresistance are observed owing to orbital fluctuation. Cr-doping steeply suppresses the ferromagnetic correlation and magnetoresistance in Nd$_{0.375}$Sr$_{0.625}$Mn$_{1-y}$Cr$_y$O$_3$ with \( 0 \leq y \leq 0.05 \), while they reappear in \( 0.05 < y \leq 0.10 \). Such a reentrant behavior implies that a phase boundary is located at \( y = 0.05 \), or a phase crossover occurs across \( y = 0.05 \).

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A. Introduction

Mn oxides with a perovskite structure have attracted much attention because of the colossal magnetoresistance (CMR) effect\cite{1,2}. Since the magnetic and transport properties of the perovskite manganites are strongly affected by ordering patterns of \( x^2 - y^2 \) and/or \( 3z^2 - r^2 \) orbitals, a detailed investigation of the orbital-ordered (OO) states is significant for understanding the CMR effect. In heavily doped Nd$_{1-x}$Sr$_x$MnO$_3$ (NSMO), there exist two types of OO states, which exhibit highly anisotropic magnetic and transport properties\cite{3}. One is the \( x^2 - y^2 \) OO state (0.53 \( \leq x \leq 0.63 \)), accompanying the A-type antiferromagnetic (AF) order in which the \( x^2 - y^2 \) electrons are conducting within the ferromagnetic (F) plane\cite{4}. The other is the \( 3z^2 - r^2 \) OO state (0.63 \( \leq x \leq 0.80 \)), accompanying the C-type AF order. These two OO states compete with each other in a bicritical manner at \( x = 0.625 \)\cite{4}. Near the bicritical region, competition between the two OO states causes spatial orbital fluctuation on nanometer scale, which gives rise to the F correlation and concomitant large magnetoresistance (MR)\cite{5,6}.

It is well-known that the presence of quenched disorder in a bicritical (or multicritical) region where a ferromagnetic metallic (FM) and AF insulating states meet often causes phase separation phenomena, which are essential for the CMR. In Nd$_{0.5}$Ca$_{0.5}$MnO$_3$, for example, Cr-substitution on Mn-sites turns the charge- and orbital-ordered (CO/OO) state into the FM one\cite{7,8}.

Therefore, it can be expected that Cr-doping into NSMO near the bicritical region (\( x = 0.625 \)) induces phase separation and/or enhances the orbital fluctuation, which might lead to nontrivial phenomena such as the CMR. In this study, we have investigated the Cr-doping effect of Nd$_{0.375}$Sr$_{0.625}$Mn$_{1-y}$Cr$_y$O$_3$ (NSMCO) \( 0 \leq y \leq 0.10 \).

B. Experiment

NSMCO crystals with \( 0 \leq y \leq 0.10 \) were prepared using the floating zone method. We confirmed that all synthesized crystals are of single phase by the powder X-ray diffraction method. Magnetic and transport properties were measured using a Quantum Design physical property measurement system (PPMS). We randomly cut the synthesized crystals with the size larger than twin-domain size for measurements of magnetic and transport properties.

C. Results and discussion

First, we show in Figs. 1(a) and 1(b) the temperature \( (T) \) dependence of the magnetization \( (M) \) and MR(80 kOe) of NSMCO with \( 0 \leq y \leq 0.05 \), respectively. Here MR(80 kOe) is defined as MR(80 kOe) \( / \rho(0 \text{ Oe}) \). Note that the MR(80 kOe) are resistivities measured in \( H = 0 \text{ Oe} \) and \( 80 \text{ kOe} \), respectively. In \( y = 0 \), the F correlation is observed due to the orbital fluctuation below 65 K, and the MR(80 kOe) at 5 K is below 0.01: the resistivity drops more than two orders of magnitude by applying a magnetic field of \( H = 80 \text{ kOe} \). This result is consistent with our previous report\cite{6}. Cr-doping steeply suppresses the F correlation and concomitant MR, which are most suppressed at \( y = 0.05 \).

Then, we exhibit the \( T \) dependence of the \( M \) and MR(80 kOe) of NSMCO with \( 0.05 \leq y \leq 0.10 \) in Figs. 2(a) and 2(b). In \( y > 0.05 \), the F correlation and MR reappear and are evolving with an increase of \( y \) from 0.05 to 0.10. Note that the \( M \) and MR of \( y = 0.10 \) are quite similar to those of \( y = 0 \), as clearly seen from Figs. 1 and 2, indicating that the F correlation and MR of NSMCO show a reentrant behavior with a change of Cr-concentration \( y \).

We plot the \( M \) under \( H = 1 \text{ kOe} \) and MR(80 kOe) at 15K as a function of \( y \) in Figs. 3(a) and 3(b), respectively. These figures, as mentioned above, demonstrate that the F correlation and MR are most suppressed at \( y = 0.05 \).

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FIG. 1: (Color online) Temperature ($T$) dependence of (a) magnetization ($M$) and (b) magnetoresistance [MR($80$ kOe)] of Nd$_{0.375}$Sr$_{0.625}$Mn$_{1-y}$Cr$_y$O$_3$ (NSMCO) with $y = 0$, 0.03, and 0.05. ZFC represents zero field cooling process. MR($80$ kOe) is defined as $\rho(80$ kOe) / $\rho(0$ Oe).

and show the reentrant behavior, implying that a phase boundary is located at $y = 0.05$, or a phase crossover occurs across $y = 0.05$.

Let us discuss the origin of the reentrant behavior. In $0 \leq y \leq 0.05$, the F correlation and MR are systematically reduced with increasing $y$. This behavior is quite similar to that observed in NSMO with $x = 0.625$, the F correlation and MR of which are also suppressed with increasing hole-concentration $x$ from 0.625$^5$. Therefore, we interpret that Cr- and hole-doping have almost the same effect on the magnetic and transport properties of NSMCO with $x = 0.625$ in $0 \leq y \leq 0.05$. This is probably because Cr$^{3+}$ and Mn$^{4+}$ have the same electronic configuration of $t_{2g}^3e_g$. In $y > 0.05$, the F correlation and MR reappear; the F correlation is developing with further increasing $y$ from 0.05. This reminds us the fact that Cr-doping into CO/OO Nd$_{0.5}$Ca$_{0.5}$MnO$_3$ produces the FM clusters embedded in the CO/OO matrix$^7$. In NSMCO with $x = 0.625$ as well as Nd$_{0.5}$Ca$_{0.5}$MnO$_3$, the F correlation is probably induced around Cr$^{3+}$. However, the F correlation is not so strong compared with that of Cr-doped Nd$_{0.5}$Ca$_{0.5}$MnO$_3$, the reason for which might be explained by the fact that NSMO with $x = 0.625$ is apart from the FM state, which is often found in the low-doped ($x \leq 0.5$) perovskite manganites.

The reentrant behavior observed in Cr-doped NSMO with $x = 0.625$ is perhaps due to competition between the hole-doping effect and the FM cluster effect caused by Cr-doping. In
0 ≤ y ≤ 0.05, the number (or the size) of the FM clusters is so small that the hole-doping effect is dominant. With increasing y, the number (or the size) of the FM clusters is becoming large, and the FM cluster effect finally overcomes the hole-doping effect in y > 0.05. As a result, the F correlation is macroscopically observed again in the magnetic and transport properties of NSMCO with y > 0.05. The detailed mechanism of the reentrant behavior is now under investigation.

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