Origin of Colossal Magnetoresistance in $Nd_{1-x}Ca_xMnO_3$ *

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Abstract

Structural, transport and magnetic properties of $Nd_{1-x}Ca_xMnO$ ($0 \leqslant x \leqslant 1$) have been studied to probe the consequence of strong lattice distortion and reduced double exchange. Charge ordering has been observed over a large composition range of $0.30 \leqslant x \leqslant 0.80$. For $0.33 \leqslant x \leqslant 0.40$, at low temperatures, a magnetic field induces a first-order AF semiconductor to FM metal transition, and reduces the resistance by several orders of magnitude. These results illustrate the competition between double exchange and mechanisms that promote charge localization.

Recently, the colossal magnetoresistance (*CMR*) effect seen in La-based perovskite-type oxides has been the focus of intense research interest[1]. The *CMR* effect has been often examined within the framework of double exchange (DE[2]). However, it has been suggested[3] that some other mechanisms, such as the strong electron-phonon coupling, may be of crucial importance to the physics of *CMP*. To study the relative significance of DE and other mechanisms, we have selected the $Nd_{1-x}Ca_xMnO_3$ system. The replacement of La by the smaller Nd introduces a larger lattice distortion, thereby weakening the DE. Therefore the effects of electronphonon coupling, Coulomb interaction and antiferromagnetic (*AF*) superexchange can be highlighted.

A series of samples of $Nd_{1-x}Ca_xMnO_3$ ($0 \leqslant x \leqslant 1$) were prepared through conventional solid state reaction method. X-ray diffraction shows that the samples are of single-phase. As Ca doping increases, the crystal structure changes gradually from orthorhombic ($0 \leqslant x \leqslant 0.60$) to tetragonal ($0.60 < x < 1$).

The temperature dependence of resistivity (ρ) and magnetization M for representative samples with $x = 0.20$, 0.35 and 0.80 are shown in Fig. 1. For samples with $0 \leqslant x \leqslant 0.25$, only semiconducting behavior with activation characteristics has been observed at all temperatures, with some *MR* effect at low temperatures. Magnetically, these samples exhibit a paramagnetic (*PM*) to ferromagnetic (*FM*) transition. For samples with $x > 0.8$, a *PM* to *AF* transition has been observed, while the samples also remain semiconducting with no appreciable magnetoresistance.

The samples with $0.30 \leqslant x \leqslant 0.80$ show a rich variety of phenomena. Although these samples are again semiconductors in zero magnetic field, there is a noticeable increase in ρ below a temperature of T_{CO} at which the magnetization correspondingly shows a peak. Electron diffraction measurements show that T_{CO} is the charge ordering temperature. At still low temperatures, the samples undergo a *PM* to *AF* transition at a temperature $T_N < T_{CO}$ in zero field.

In the following, we concentrate only on the sample with $x = 0.35$. At low temperatures, *AF* behavior and spin canting have been observed. The zero-field resistivity $\rho(0)$ increases monotonically with decreasing temperature and reaches about $10^3 \Omega$-m at 50K, below which $\rho(0)$ is so large that it has exceeded our measuring limit. Under an external field of $5T$, the resistivity

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decreases by more than seven order of magnitude to $\rho(5T)\approx10^{-4}\Omega$-m at low temperature, exhibiting one of the largest magnitudes of *CMR*. The sharp drop in ρ i. e. *CMR*, is accompanied by a field-induced magnetic transition from *AF* to *FM*, as indicated by the zero-field-cooled (ZFC) and field-cooled (FC) magnetization curves in Fig. 1(b). This magnetic transition is first-order in nature, signified by pronounced temperature hysteretic effects.

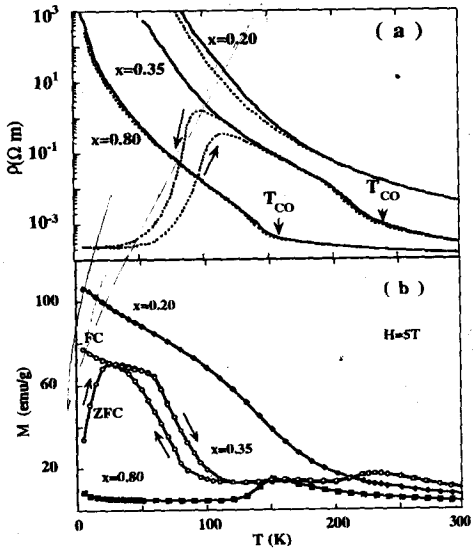

Fig. 1 Representative results of temperature dependence of (a) resistivity ρ in zero field (solid line) and 5T (dashed line), and (b) magnetization M in 5T field for x = 0.20, 0.35, and 0.80.

The interesting transport and unusual magnetic behaviors exhibited in the present system originate from a competition between mechanisms that promote electron itineracy and those that favor localization. Double exchange facilitates electron itineracy and ferromagnetism. In competition with DE, a few mechanisms favor electron localization. The Jahn-Teller distortion of the Mn^{3+} ($3d^4$) state lowers the energy of the e_g electron and impedes it from hopping to the undistorted e_g state of Mn^{4+}, leading to the formation of polarons. The mutual repulsion between charge carriers accommodates charge ordering at low temperature. The *AF* superexchange gives rise to antiparallel core spin alignment which prevents hopping of spin-carrying e_g electrons. These effects lead to charge localization, ultimately charge ordering, and antiferromagnetism.

In systems like $La_{1-x}Ca_xMnO_3(0.20\leqslant x\leqslant0.45)$[4], DE dominates the competition below T_c, metallic conduction and *FM* therefore prevail. Above T_c, electron localization takes place, resulting is semiconducting and *PM* behavior. In the present system, the replacement of La by the smaller Nd introduces a larger lattice distortion, reduces the Mn-O-Mn bond angle in particular, thereby weakens the DE[5]. The competition now favors charge localization in the whole temperature range in zero field. Consequently, only semicomducting behavior can be observed in all compositions $0\leqslant x\leqslant1$. The resistivity values of the Nd-based materials are also larger than those in the La-or Pr-based systems[4,5]. Furthermore, charge ordering transition has been observed in a broad composition range $0.30\leqslant x\leqslant0.80$ of the Nd-based materials. Finally, *AF*, instead of *FM*, occurs at low temperatures in the same composition range. These results illustrate well the close competition between double exchange and the mechanisms of electron-phonon, Coulomb interaction and antiferromagnetic superexchange.

References

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