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## Electrical-field-induced order transitions in Sr-doped manganite perovskites

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#### Abstract

Spin-dependent varistor-like electrical transport was observed in Sr-doped  $Nd_{2/3}Sr_{1/3}MnO_{3-\delta}$  perovskites for  $\delta > 0.05$ , in which magnetic grain or phase boundaries is likely to be responsible. In addition to the varistor-like phenomena below a characteristic temperature, we also present evidence for electrical-field-induced magnetic order–order precursor transition at even lower temperature in regions close to highly insulating boundaries where larger electrical fields can be placed. This electrical-field-induced metamagnetic transition was understood by the competition among double exchange, superexchange interactions and external electrical or magnetic field. © 2008 Elsevier B.V. All rights reserved.

Keywords: Boundary resistance; Spin-dependent electrical transport; Manganite; Colossal magnetoresistance

#### 1. Introduction

Recently, a current dependent resistance has been reported for ceramic Nd<sub>2/3</sub>Sr<sub>1/3</sub>MnO<sub>z</sub> with  $z \le 2.90$  below a characteristic temperature  $T_{\rm B}$  which was defined as a transition from linear (ohmic) I(V) characteristic to a curved (varistor-like) one: for example  $T_{\rm B} = 150$  K for z = 2.85 [1] and Fig. 1. In the varistorlike regime an abrupt step is observed at  $T_{\rm K} < T_{\rm B}$  which shifts higher when the constant load current  $I_0$  or the constant applied voltage  $V_0$  is enhanced. As a spin-dependent varistor-like transport through a magnetic boundary would be continuous [2–4], this suggests a convolution of electrical transport through the (grain or phase) boundaries and a change of the magnetic state in the domains which border the boundary and which would act on the boundary resistance through the orientation of the magnetic quantization axes (magnetization M). The direction of Mwould change with the magnetic state in the domains and this state would well undergo a phase transition under global external forces like temperature T or magnetic field H. In this contribu-

\* Corresponding author. *E-mail address:* cpyang@hubu.edu.cn (C.P. Yang). tion we will try to connect the current induced shift of  $T_{\rm K}$  with an electrical-field-induced magnetic order–order transition. However, magnetic phase transitions induced by an electrical-field E would be confined to a region where a large E-field can be applied, i.e. inside of or near the boundary.

#### 2. Experimental details

Ceramic Nd<sub>2/3</sub>Sr<sub>1/3</sub>MnO<sub>z</sub> samples have been prepared using conventional ceramic technology as described in detail elsewhere [1]. X-ray analysis show that all the compounds have an orthohombically distorted perovskite structure (space group *Pbnm*) without any traces of foreign phases. The unit cell volume gradually increases as the nominal oxygen vacancy content increases, which is usually associated with the conversion of Mn<sup>4+</sup> ions into Mn<sup>3+</sup> ions with a much larger ionic radius. For the measurement of the magnetization the Faraday method was used and for the resistivity measurement the standard four-point method, the sample being under constant current or voltage load. The temperature range of measurement was limited by temperature of liquid nitrogen.

### 3. Results

#### 3.1. Magnetic states inside of the domains

As in manganites the magnetic and transport properties are usually strongly correlated, we first try to find out which

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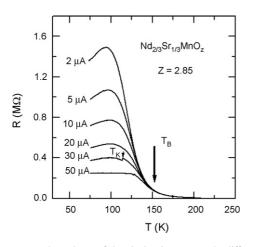


Fig. 1. Temperature dependence of electrical resistance *R* under different current load of a sample  $Nd_{2/3}Sr_{1/3}MnO_z$  with z=2.85.  $T_K$ : step in R(T) occurring at a critical current  $I_C$  and an electrical-field-induced order–order transition point.  $T_B$ : a transition point from non-linear to ohmic behavior of *R*.

magnetic states are involved (inside the grains). For that we have measured the magnetization versus temperature curves. As examples Fig. 2 shows the (bulk) magnetization M(T) (at 0.5 T) and the reciprocal susceptibility  $\chi^{-1}(T)$  of the compounds with z = 3.0 and 2.90. M(T) of the base compound Nd<sub>2/3</sub>Sr<sub>1/3</sub>MnO<sub>3</sub> compares well with the literature [5] and the paramagnetic and ferromagnetic Curie temperatures as well as the metal to insulator transition temperature coincide,  $\Theta_{\rm f} = T_{\rm C} = T_{\rm MI}$ . For z = 2.90, the situation is more complex:

(1) By fitting the lower part of the reciprocal susceptibility  $\chi^{-1}(T)$  to a ferromagnetic (FM) Curie-Weiss law, one obtains a paramagnetic Curie temperature  $\Theta_f = 89$  K, which is similar to that of the base compound. Although the magnetically polarized state is not reached in this experiment,

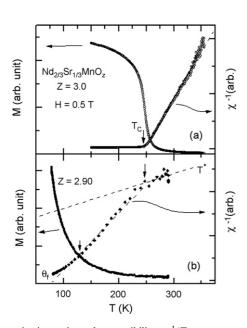


Fig. 2. Magnetization reciprocal susceptibility  $\chi^{-1}(T)$  vs. temperature and for Nd<sub>2/3</sub>Sr<sub>1/3</sub>MnO<sub>z</sub> samples with nominal z=3.0 (a) and 2.90 (b).  $T_{\rm C}$ : ferromagnetic Curie temperature;  $\Theta_{\rm f}$ : paramagnetic Curie temperature;  $T^*$ : Néel point.

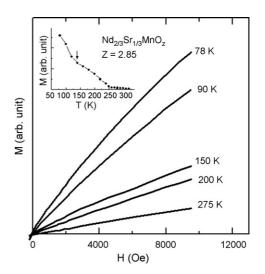


Fig. 3. Magnetization and thermomagnetic (insert) curves at temperatures from 300 to 78 K of a sample  $Nd_{2/3}Sr_{1/3}MnO_z$  with z=2.85. Magnetization vs. field showing a transition from linear to non-linear around T=150 K and a similar abrupt step also observed in the thermomagnetic curve at around 150 K (insert).

we expect a FM or canted metallic state below  $T_{\text{MI}} = \Theta_{\text{f}}$  (see also Fig. 4).

- (2) For z=2.9, χ<sup>-1</sup>(T) goes below a FM Curie–Weiss law at a higher temperature T\* = 250 K, suggesting mixed couplings. A similar behavior and antiferromagnetic structure (AFM) was observed for a compound characterized as Nd<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>2.90</sub>, in which a small peak in the specific heat capacity c<sub>p</sub>(T) was observed at 240 K where there was no spontaneous magnetization *M*, while a steep rise in *M* (at 0.01 T) appeared only at 150 K. [6] Moreover, χ<sup>-1</sup>(T) for the case that AFM order is followed by a canted state is present in Ref. [7], and the result is very similar to what found here.
- (3) For z = 2.85 the magnetization versus field curves M(H) at different temperatures (Fig. 3) also suggest the existence of a AFM canted state below  $T_1 > \Theta_f = 50$  K as the slope of M(H) is linear at high temperatures and gets curved only below

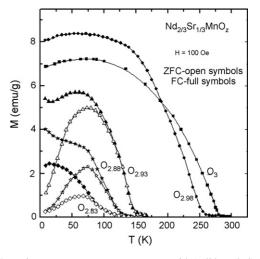


Fig. 4. Magnetic moment vs. temperature curves with (solid symbol) and without cooling field (open symbol) for z = 2.93, etc. within a temperature range from 300 to 4.2 K.

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