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# Colossal magnetoresistance and magnetic phase transitions in bulk $\text{Nd}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$

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Perovskite-type  $\text{Nd}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$  has been synthesized and its magnetic and magnetotransport properties studied in the temperature range from 5 K to 300 K. X-ray diffraction indicates that the crystal-symmetry is orthorhombic with  $a=0.5508(1)$  nm,  $b=0.5496(2)$  nm, and  $c=0.7764(3)$  nm. There exist two magnetic phase transitions. It first undergoes a paramagnetic-to-ferromagnetic phase transition at Curie temperature,  $T_C=115$  K, and then partial ordering of Nd magnetic moments develops at  $T_{\text{Nd}}=40$  K as temperature is further decreased. Resistivity measurements indicate corresponding insulator-to-metal transition near  $T_C$  and a reentrant metal-to-insulator transition near 40 K. Colossal magnetoresistance exists over a wide temperature range,  $MR = [R_{(H)} - R_{(0)}] / R_{(0)}$  is over 90% below 120 K. The maximum MR (98%) appears at 95 K, close to the paramagnetic-to-ferromagnetic phase transition. © 2000 American Institute of Physics. [S0021-8979(00)93308-5]

Rare-earth manganites  $\text{R}_{1-x}\text{T}_x\text{MnO}_3$  (R=rare earth, T=divalent cation) have recently attracted much consideration because of the colossal magnetoresistance (CMR) found in bulk and thin film samples of the compounds.<sup>1</sup> The partial substitution of the rare earths by the divalent cations results in  $\text{Mn}^{3+}$ - $\text{Mn}^{4+}$  mixed valence, leading to ferromagnetism and metallic conductivity at low temperatures. This behavior is usually explained with the double exchange theory<sup>2</sup> based on the exchange of electrons between  $\text{Mn}^{3+}$  and  $\text{Mn}^{4+}$  ions through an  $\text{O}^{2-}$  ion. The low temperature phase diagrams of doped rare-earth manganites are extremely rich and complex. Their ground states have been found to be either metallic or insulating and range from ferromagnetic, antiferromagnetic, charge ordered, canted antiferromagnetic, and spin glass states.<sup>1</sup> Most of the studies done on Ba-substituted compounds are lanthanum barium manganites.<sup>3-5</sup> We have recently synthesized a barium substituted neodymium compound  $\text{Nd}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$  and report in this article the magnetic and transport properties of a bulk  $\text{Nd}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$  sample.

The  $\text{Nd}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$  compound was synthesized by solid state reaction of stoichiometric mixture of dry  $\text{Nd}_2\text{O}_3$ ,  $\text{BaCO}_3$  (Aldrich), and  $\text{MnO}_2$  (Fluka) at 1200 °C for 50 h with several repeated grinding. The power was then pressed into a pellet of 12 mm diameter and 2 mm thickness. The pellet was sintered at 1350 °C for 12 h and then cooled to room temperature in air. The sintered pellet was later put into a tube furnace with flowing oxygen at 800 °C for 12 h and then cooled to room temperature. The powder x-ray diffraction data can be indexed with an orthorhombic or pseudotetragonal ( $a \approx b \geq c/2^{1/2}$ ) structure with  $a=0.5508(1)$  nm,  $b=0.5496(2)$  nm, and  $c=0.7764(3)$  nm.

We show in Fig. 1(a) the temperature dependence of

magnetization ( $M$ ) from 5 to 300 K measured with a Quantum Design (SQUID) in a field of  $H=1000$  Oe. It is clearly seen that there are two magnetic phase transitions, one of which is a paramagnetic-to-ferromagnetic transition at Curie temperature  $T_C=115$  K. The nature of the other at about 40 K is less clear. The decrease in  $M$  with decreasing temperature below 40 K is also evident in data taken with a higher field ( $H=5000$  Oe) in both zero-field-cooled and field-cooled runs. Ferromagnetic-to-antiferromagnetic transition is typically found in doped rare-earth manganites when the doping level is greater than  $x=0.5$  although such phase transition is also observed at lower doping levels, for example,  $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$ , (Ref. 6) and  $\text{Pr}_{0.7}(\text{Sr,Ca})_{0.3}\text{MnO}_3$ .<sup>7,8</sup> We believe, however, the low temperature phase is associated with the partial ordering of the Nd moments as will be discussed later. In Fig. 1(a),  $T_{\text{Nd}}$  denotes the onset temperature of the partial ordering of the Nd moments.

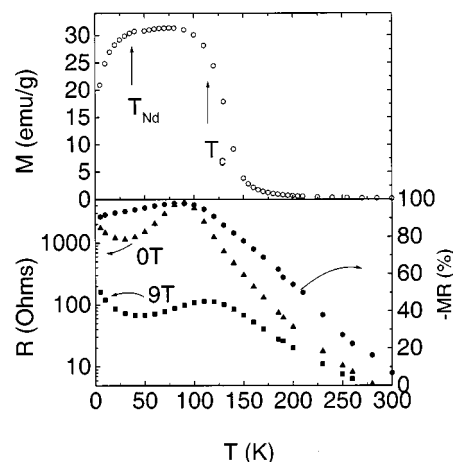


FIG. 1. Temperature dependence of (a) magnetization in a field of 0.1 T, (b) resistance in zero field and 9 T, and magnetoresistance (MR) from 5 to 300 K.

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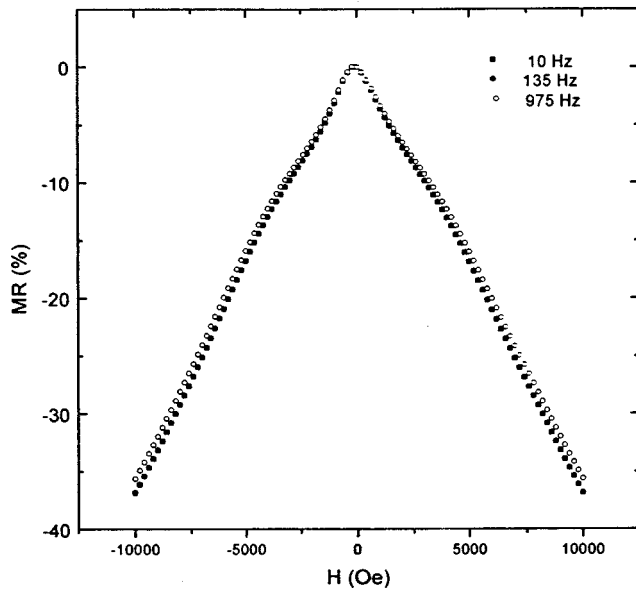


FIG. 2. ac magnetoresistance of  $\text{Nd}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$  at 95 K.

The temperature dependence of dc resistance  $R(H=0)$  is shown in Fig. 1(b) along with that measured in all applied field of  $H=9$  T. At zero field,  $R$  can be divided into three different regions between 5 and 300 K, which correlate well with the two magnetic phase transitions. With decreasing temperature, the resistance increases monotonically to 95 K, characteristic of an insulator/semiconductor ( $dR/dT < 0$ ).  $R$  reaches a peak at  $T=95$  K, then decreases rapidly as temperature is decreased to about 40 K. This metallic behavior is induced by the ferromagnetic ordering that drastically enhances the electron hopping rate from  $\text{Mn}^{3+}$  to  $\text{Mn}^{4+}$  which strongly depends on the relative orientation of the spins of the Mn ions.<sup>2</sup> From 40 to 5 K, the resistance increases again, showing a strong temperature dependence. This indicates that the system reenters an insulating state. The reentrant metal-to-insulator transition coincides with the magnetic phase transition at 40 K.

At  $H=9$  T, the resistance shows a very large reduction, especially at low temperatures. The peak in the  $R$  vs  $T$  curve is shifted to a higher temperature. This shift of peak position, which is due to field-induced insulator-to-metal transition, is mainly responsible for the colossal magnetoresistance observed in rare-earth manganites.<sup>1,9</sup> In Fig. 1(b), we also show magnetoresistance  $\text{MR} = [R_{(9T)} - R_{(0)}]/R_{(0)}$ , as a function of temperature. One can see clearly that large MR of 90%–98% persists over the entire temperature range from 5 to 120 K. The maximum MR (98%) appears at 95 K, close to the paramagnetic-to-ferromagnetic phase transition. That large MR extends down to the lowest temperature should be closely related to the existence of the low temperature phase below 40 K in  $\text{Nd}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ .

Figure 2 shows the ac MR of  $\text{Nd}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$  as a function of magnetic field measured at 95 K and up to 1 T. The magnetic field was applied perpendicular to the current direction. It reveals two distinct regions separated by a cusp at 2500 Oe. The two regions have different slopes in the MR versus field curves. The low field region, which has a fast response to the field, is probably related to the so-called extrinsic MR where the process is dominated by intergrain transport.<sup>10</sup> In the high field region, magnetotransport is that of intrinsic CMR as discussed earlier. The MR reaches about 35% at  $H=1$  T and 95 K. There is essentially no difference in MR when frequency is changed from 10, 135 to 975 Hz.

We now discuss the properties of the observed magnetic phases. Figure 3 shows the magnetization as a function of applied field measured at 4, 77, and 300 K, respectively. It is paramagnetic at 300 K. The curve at 77 K is that of a typical ferromagnet. The magnet moment seems to saturate at 72.4 emu/g, which is  $3.2\mu_B/\text{Mn}$ . This is slightly lower than the spin-only value of  $3.7\mu_B/\text{Mn}$  expected for the full alignment of the Mn moments at the given divalent ion concentration. There are several possible explanations for this reduced saturation moment including canted spin structure<sup>11</sup> and oxygen deficiency in the samples.<sup>4</sup>

At 4 K, the magnetization is again characteristic of a ferromagnet but increases beyond the expected maximum

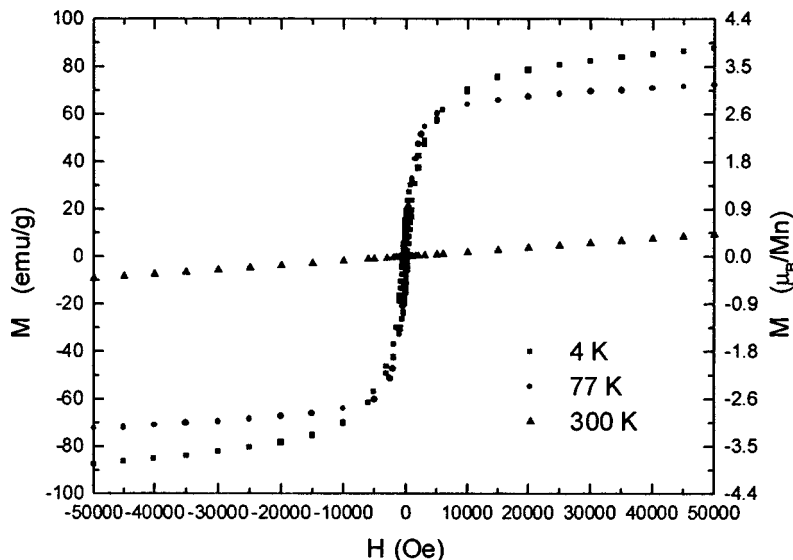


FIG. 3. Magnetization of  $\text{Nd}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$  as a function of applied field at 4, 77, and 300 K, respectively.

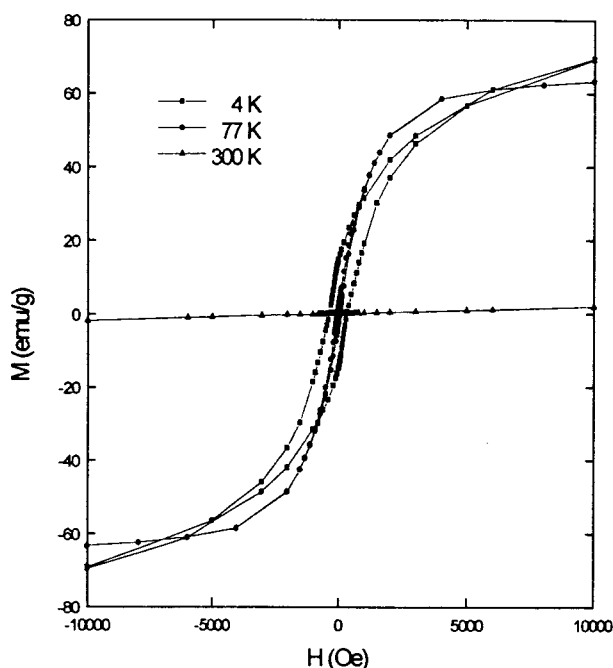


FIG. 4. Hysteresis curves in low field region.

$3.7\mu_B/\text{Mn}$  as the field increases and it shows no sign of saturation at 5 T. The magnetic moments is  $3.85\mu_B/\text{Mn}$  at 5 T and  $4.10\mu_B/\text{Mn}$  at 9 T (not shown). We interpret this as an indication of partial polarization of the Nd moments. The Nd moments may align parallel, or partially parallel, to the ferromagnetic Mn sublattice and contribute to the total magnetization. The reduced contribution from Nd ( $\sim 1\mu_B/\text{Nd}$ , see Fig. 3) compared to the expected value ( $3.27\mu_B/\text{Nd}$ ) may also arise from the crystal field effect, which splits the ground state  $^4I_{9/2}$  multiplet into five Kramers doublets.<sup>12</sup> The partial ordering of rare earth moments in the doped manganites has been suggested by high magnetic field studies.<sup>13,14</sup>

Figure 4 shows the hysteresis curves in the low field region. The high temperature ferromagnetic state at 77 K has a small coercivity  $H_c$  of about 60 Oe. Below  $T_{\text{Nd}}$ , the hysteresis curve shows a drastically increased coercivity,  $H_c \sim 350$  Oe at 4 K. A high coercivity is consistent with the involvement of the Nd moments, which has higher magnetic crystalline anisotropy, in the magnetic phase below  $T_{\text{Nd}}$ . The decrease in the magnetization below  $T_{\text{Nd}}$  shown in Fig. 1(a) is likely caused by the sudden increase in the magnetization

below  $T_{\text{Nd}}$  shown in Fig. 1(a) is likely caused by the sudden increase in the magnetic anisotropy constants. It should be pointed out that we could not rule out the possibility that the phase transition at 40 K is related to a charge ordering or some type of structural distortion in  $\text{Nd}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ . The exact nature of the low temperature ground state needs to be further studied.

In summary, we have synthesized a barium-substituted neodymium manganite  $\text{Nd}_{0.7}\text{Ba}_{0.3}\text{MnO}_3$ . The compound exhibits two magnetic phase transitions with Curie temperature  $T_C = 115$  K and  $T_{\text{Nd}} = 40$  K, where  $T_{\text{Nd}}$  is the onset temperature for the partial ordering of the Nd moments. We have shown experimental evidences that the low temperature magnetic phase involves the Nd moments and that the Nd moments align parallel, or partially parallel, to the ferromagnetically ordered Mn. Large magnetoresistance 90%–98% was observed over the whole temperature range  $T < 120$  K. The large magnetoresistance at very low temperature is probably associated with the reentrant metal-to-insulator transition that accompanies the magnetic phase transition at  $T_{\text{Nd}}$ . The exact mechanism responsible for this large magnetoresistance below  $T_{\text{Nd}}$  needs to be further studied.

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