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LETTER TO THE EDITOR

Effect of Dimensionality on the Giant Magnetoresistance of the Manganates: A Study of the (La, Sr)$_{n+1}$Mn$_n$O$_{3n+1}$ Family

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An investigation of the $n = 1$, 2, 3 and ∞ members of the (La, Sr)$_{n+1}$Mn$_n$O$_{3n+1}$ or the (SrO)(La$_{1-x}$Sr$_x$MnO$_3$)$_n$ family has shown the dependence of the I–M transition temperature and the magnitude of magnetoresistance (MR) on dimensionality. MR decreases as dimensionality increases from 2 to 3.

The importance of dimensionality in determining the electron transport and magnetic properties of solids has long been recognized. Investigations of a few related series of transition metal oxides with varying dimensionality between 2 and 3 have indeed shown that the electron transport properties depend markedly on the dimensionality (1, 2). We therefore considered it important to investigate the effect of dimensionality on the giant magnetoresistance (GMR) and related properties of manganates by investigating the (La, A)$_{n+1}$Mn$_n$O$_{3n+1}$ (A = alkaline earth) system. Manganates of the formula La$_{1-x}A_x$MnO$_3$ (A = alkaline earth), which exhibit GMR (3–5), possess the three-dimensional perovskite structure. In the (La, A)$_{n+1}$Mn$_n$O$_{3n+1}$ system, the dimensionality, d, can be varied by varying the number of perovskite layers. In the present study we have synthesized members of the (La, Sr)$_{n+1}$Mn$_n$O$_{3n+1}$ system which can also be written as (SrO) (La$_{1-x}$Sr$_x$MnO$_3$)$_n$, where n is the number of perovskite layers. In Fig. 1 we show the schematic structures of the $n = 1$, 2, 3 and ∞ members, where the three-dimensional La$_{1-x}$Sr$_x$MnO$_3$ containing an infinite number of perovskite layers is the $n = \infty$ member. The $n = 1$ member has the two-dimensional K_2NiF_4-type structure ($d = 2$) while the $n = 2$ and 3 members would have dimensionality between 2 and 3. GMR and related properties of La$_{0.7}$Sr$_{0.3}$MnO$_3$, the $n = \infty$ member ($d = 3$), have been well established (3, 6). We compare the properties of the $n = 1$, 2, and 3 members with those of the $n = \infty$ member in this letter to illustrate the role of dimensionality on the GMR and other properties of manganates.

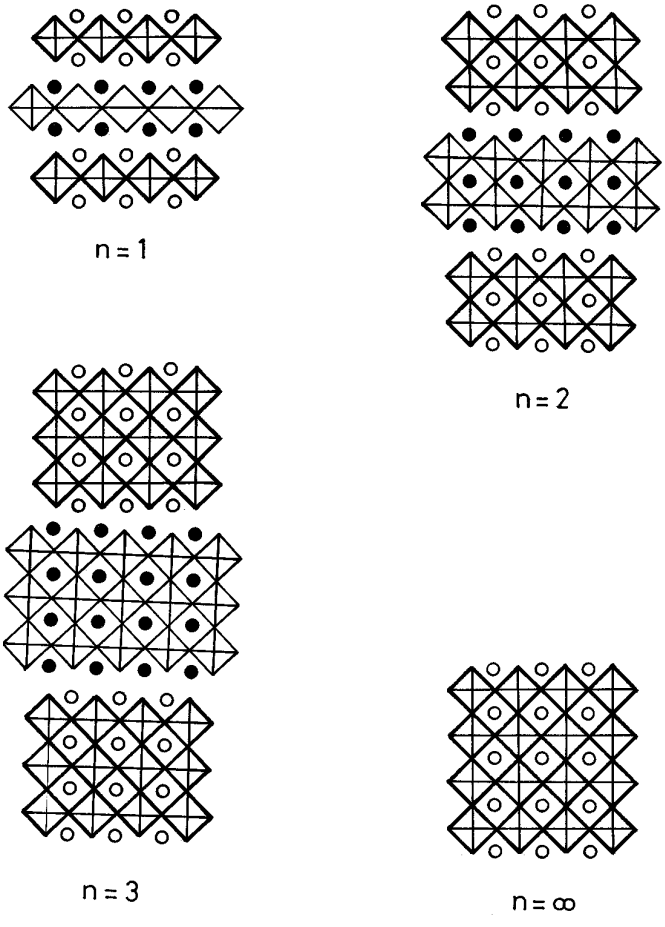

FIG. 1. Schematic representation of $n = 1$, 2, 3, and ∞ members of the (SrO) (La$_{1-x}$Sr$_x$MnO$_3$)$_n$ family. The $n = 1$ and ∞ members have dimensionality (d) of 2 and 3, respectively; (0, ●) and ⊕ represent La, Sr atoms and MnO$_6$ octahedra, respectively.

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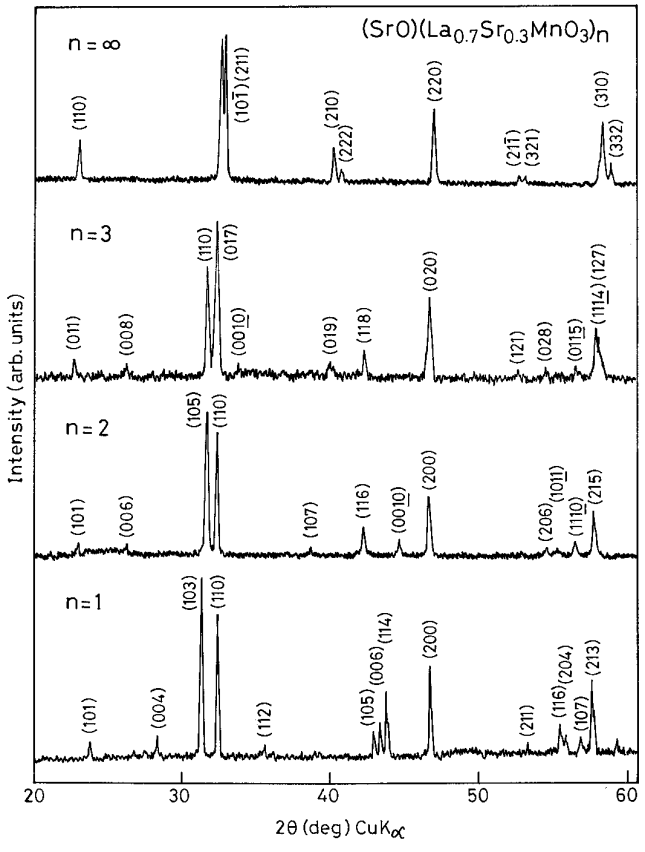

FIG. 2. Powder X-ray diffraction patterns of $n = 1$, 2, 3, and ∞ members of the (SrO) $(La_{0.7}Sr_{0.3}MnO_{3})_n$ family.

Manganates of the formula (SrO) $(La_{0.7}Sr_{0.3}MnO_3)_n$ with $n = 1$, 2, and 3 were prepared by the sol–gel technique by treating a nitrate solution containing La^{3+}, Sr^{2+}, and Mn^{2+} with citric acid and ethylenediamine. The gel so obtained after decomposition at 773 K was heated to 1273 K for 24 h in flowing oxygen. The powder thus obtained was pelletized and sintered at 1573 K for 72–120 h in flowing oxygen. Powder X-ray diffraction patterns were recorded to check the phase purity of various members. The Mn^{4+} content in the manganates was determined by redox titrations using potassium permanganate and ferrous sulfate. Electrical resistivity measurements were carried out on bar-shaped samples in the temperature range 4.2–400 K using the four-probe dc or ac (20 Hz) technique up to a maximum magnetic field of 6 T. The ac susceptibility was measured using a mutual inductance bridge operating at 100 Hz to find the ferromagnetic Curie temperature.

In Fig. 2 we show the powder X-ray diffraction patterns of $n = 1$, 2, 3, and ∞ members and list the unit cell parameters in Table 1. We see that the tetragonal c parameter varies between 12.512 Å for the $n = 1$ member and 28.148 Å for the $n = 3$ member, the a parameter being nearly constant. The progressive increase in c with n demonstrates that these oxides possess the structures shown in Fig. 1. Then $n = \infty$ member is rhombohedral with $a = 5.454$ Å and $\alpha = 60.14°$. Electrical resistivity measurements show that the $n = 2$ and 3 members exhibit resistivity maxima due to the insulator–metal (I–M) transitions just like the $n = \infty$ ($d = 3$) member, although at lower temperatures. The $n = 1$ member ($d = 2$), however, does not show a resistivity maximum corresponding to an I–M transition (Fig. 3); instead, it is an insulator down to low temperatures. The I–M transition temperature or the temperature corresponding to the resistivity peak, T_p, varies with the dimensionality or number of perovskite layers, n, the actual values being 110, 150, and 375 K for the $n = 2$, 3, and ∞ members, respectively. The ferromagnetic Curie temperature T_c also varies in the same direction.

Application of a magnetic field of 6 T results in a marked decrease in the resistivity of all the members of the (SrO) $(La_{0.7}Sr_{0.3}MnO_3)_n$ family, including the $n = 1$ ($d = 2$) member (Fig. 3). What is especially interesting is that the two-dimensional $n = 1$ member which is insulating also exhibits a GMR. The magnitude

TABLE 1
Structure and Properties of (SrO) $(La_{0.7}Sr_{0.3}MnO_3)_n$

	Lattice parameter[a]				MR (%) at		
n	a (Å)	c (Å)/α (°)	T_p(K)	T_c(K)	T_p	150 K	4.2 K
1 ($d = 2$)	3.892	12.512	—	—	—	60	—
2	3.895	20.277	110	110	80	60	60
3	3.898	28.148	150	—	30	40	46
∞ ($d = 3$)	5.454	60.14	375	395	40	30	38

[a] Mn^{4+} content in all the samples is in the range 33 ± 3%.

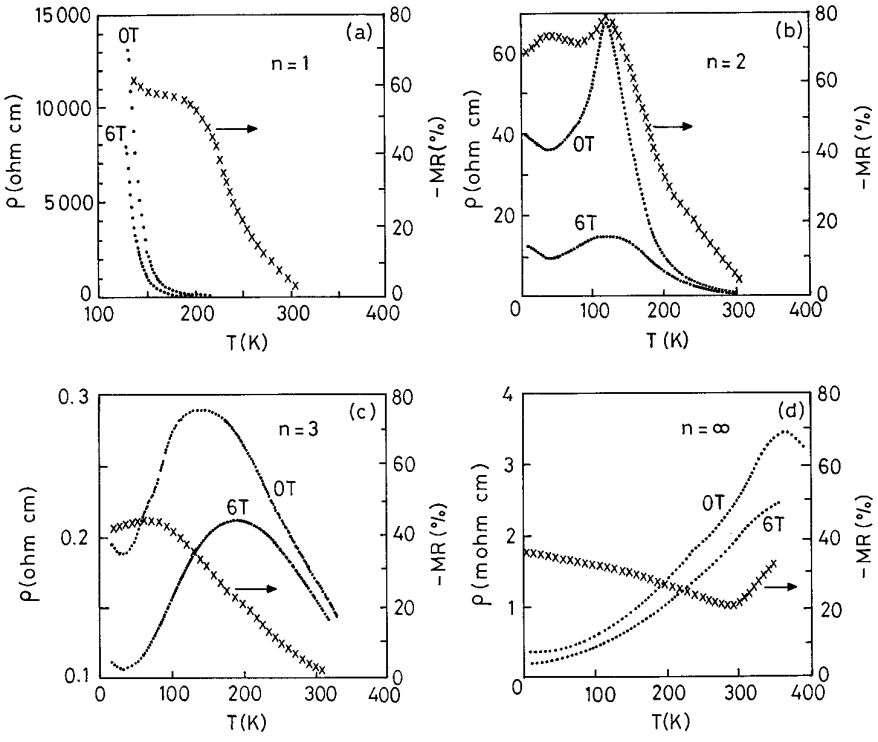

FIG. 3. Temperature variation of resistivity (at $H = 0$ and 6 T) and percent magnetoresistance (MR) of various members of the (SrO)$(La_{0.7}Sr_{0.3}MnO_3)_n$ family.

of the GMR, however, varies from member to member. In Table 1, we have listed the MR at T_p 150 K, and 4.2 K in the different members. We see that with an increase in the dimensionality (d) from 2 to 3, the magnitude of GMR decreases, independent of the temperature. This implies that a lower dimensionality is more favorable for GMR, a situation similar to that of high-temperature superconductivity in cuprates (7). Low T_c or T_p and high resistivity (around T_c or T_p) are known to favor high MR in manganates (6). Since T_c/T_p in (SrO)$(La_{0.7}Sr_{0.3}MnO_3)_n$ increases with n (or d), we would expect greater MR when the dimensionality is low ($d < 3$). In order to attain desirable MR characteristics, it therefore seems preferable to have a quasi-two-dimensional system with high resistivity, but with a reasonable value of T_c or T_p. Noting that the T_c or T_p in the $n = \infty$ ($d = 3$) member is 380 K and assuming that the value in the $n = 1$ ($d = 2$) member is appropriate, the dimensionality of the $n = 2$ and 3 member works out to be between 2 and 3, as expected.

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