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A Comparative Study of the Magnetic and Electrical Properties of Perovskite Oxides and the Corresponding Two-Dimensional Oxides of K_2NiF_4 Structure*

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Electrical and magnetic properties of several oxide systems of K_2NiF_4 structure have been compared to those of the corresponding perovskites. Members of the $La_{1-x}Sr_{1+x}CoO_4$ system are all semiconductors with a high activation energy for conduction unlike $La_{1-x}Sr_xCoO_3$ ($x \geq 0.3$) which is metallic; the latter oxides are ferromagnetic. $La_{0.5}Sr_{1.5}CoO_4$ shows a magnetization of 0.5 μ_B at 0 K (compared to 1.5 μ_B of $La_{0.5}Sr_{0.5}CoO_3$), but the high-temperature susceptibilities of the two systems are comparable. In $SrO \cdot (La_{0.5}Sr_{0.5}MnO_3)_n$, both magnetization and electrical conductivity increase with the increase in n approaching the value of the perovskite $La_{0.5}Sr_{0.5}MnO_3$. $LaSrMn_{0.5}Ni_{0.5}(Co_{0.5})O_4$ shows no evidence of long-range ferromagnetic ordering unlike the perovskite $LaMn_{0.5}Ni_{0.5}(Co_{0.5})O_3$; high-temperature susceptibility behavior of these two insulating systems is, however, similar. $LaSr_{1-x}Ba_xNiO_4$ exhibits high electrical resistivity with the resistivity increasing proportionately with the magnetic susceptibility (note that $LaNiO_3$ is a Pauli-paramagnetic metal). High-temperature susceptibility of $LaSrNiO_4$ and $LaNiO_3$ are comparable. Susceptibility measurements show no evidence for long-range ordering in $LaSrFe_{1-x}Ni_xO_4$ unlike in $LaFe_{1-x}Ni_xO_3$ ($x \leq 0.35$) and the electrical resistivity of the former is considerably higher. Electrical resistivity of Sr_2RuO_4 is more than an order of magnitude higher than that of $SrRuO_3$. Some generalizations of the properties of two- and three-dimensional oxide systems have emerged from these experimental observations.

Introduction

Electrical and magnetic properties of perovskite oxides, ABO_3 (B = transition metal ion), have been investigated extensively (*1–6*). Quasi-two-dimensional oxides of the K_2NiF_4 structure, A_2BO_4, possessing ABO_3 layers in between rock-salt AO layers wherein B–O–B interaction occurs only in the ab plane have also been investigated in some detail (*7*). Electrical and magnetic properties of the A_2BO_4 oxides are considerably different from those of the ABO_3 perovskites. Thus, $LaNiO_3$ is a Pauli-paramagnetic metal while La_2NiO_4 exhibits two-dimensional canted antiferromagnetic ordering below 204 K and a semicon-

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* Contribution No. 413 is dedicated to Professor John B. Goodenough on the occasion of his 65th birthday.

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ductor–metal transition above 650 K (*8*, *9*). It is found that the evolution of metallic conductivity occurs as one goes from La_2NiO_4 to the higher members of the $LaO \cdot (LaNiO_3)_n$ or the $La_{n+1}Ni_nO_{3n+1}$ family (*10*). A strict comparison of the magnetic and electrical properties of the three-dimensional ABO_3 perovskites and two-dimensional oxides of the A_2BO_4 type would be possible only when the same oxidation state of the transition metal (B) ion is maintained. Such a comparison made with some of the available information (*7*, *10*, *11*) indicates fascinating differences between the two systems. For example, $LaSrNiO_4$ exhibits two orders of magnitude higher resistivity than $LaNiO_3$; the former also shows a higher susceptibility at low temperatures. $LaSr_3Co_2O_8$ is a semiconducting ferromagnet while $LaSrCo_2O_6$ is an itinerant-electron ferromagnet. We considered it most instructive to compare the magnetic and electrical properties of several two-dimensional oxide systems of K_2NiF_4 structure with those of the corresponding perovskites in order to arrive at possible generalizations. We have carried out investigations on several such systems: $La_{1-x}Sr_{1+x}CoO_4$ and $La_{1-x}Sr_xCoO_3$, $LaSrMn_{0.5}T_{0.5}O_4$ and $LaMn_{0.5}T_{0.5}O_3$ (T = Ni or Co), $LaBNiO_4$ (B = Sr or Ba) and *La*NiO_3, $SrO \cdot (La_{1-x}Sr_xMnO_3)_n$ and $La_{1-x}Sr_xMnO_3$, $LaSrFe_{1-x}Ni_xO_4$ and $LaFe_{1-x}Ni_xO_3$, and lastly Sr_2RuO_4 and $SrRuO_3$. In what follows, we briefly present the results of our studies on these systems and arrive at some important generalizations on the properties of these two- and three-dimensional oxides.

Experimental

$La_{1-x}Sr_{1+x}CoO_4$ compounds were prepared by the decomposition of coprecipitated carbonates obtained by adding an aqueous solution of the nitrates of the corresponding metal ions, in the required stoichiometry, to a solution of Na_2CO_3 taken slightly in excess. The resulting precipitates were washed thoroughly, dried, and decomposed at 1150 K, after which they were pelletized and heated at 1200–1400 K for 24–72 hr.

Compounds of the $LaFe_{1-x}Ni_xO_3$ and $LaSrFe_{1-x}Ni_xO_4$ system were prepared by adding the required proportions of the estimated solutions of $La(NO_3)_3$, $Sr(NO_3)_2$, $Al(NO_3)_3$, $FeCl_3$, and $NiCl_2$ to a concentrated solution of Na_2CO_3 under constant stirring. The precipitate was washed thoroughly with hot water to obtain alkali-free precipitate. The precipitates were decomposed at 1000 and 1170 K, respectively, for 27 hr. The powder obtained was ground, pelletized, and heated at 1100 K (24 hr) and 1570 K (48 hr), respectively, with intermittent grinding and pelletizing. LaSr (Ba)$MnTO_4$ (T = Ni, Co) and Sr_2RuO_4 were prepared by a ceramic method. Required amounts of La_2O_3 (dried), $SrCO_3$, $NiC_2O_4 \cdot 2H_2O$, $CoC_2O_4 \cdot 2H_2O$, $MnC_2O_4 \cdot 2H_2O$, and RuO_2 were weighed, mixed well, ground, and decomposed at 1270 K for 24 hr. The resulting powders were ground, pelletized and heated at 1470 K for 24 hr with intermittent grinding and pelletizing. Oxygen stoichiometry of all the compounds was determined by iodimetric titrations and the titrable oxygen content corresponded to the formal oxidation states expected in most of the cases.

The monophasic nature of all the compounds was confirmed using JEOL X-ray diffractometer. Magnetic susceptibility measurements were carried out by the Faraday method using a Cahn RG electrobalance. Magnetization measurements were carried out with a PAR vibrating sample magnetometer in conjunction with a closed cycle refrigeration (AIR products Displex) unit. Electrical resistivity studies were carried out on pressed, sintered pellets using the four-probe dc method.

Results and Discussion

$La_{1-x}Sr_{1+x}CoO_4$ and $La_{1-x}Sr_xCoO_3$

The inverse magnetic susceptibility, χ_M^{-1}–temperature ($T \geq 300$ K) plots of a few members of the $La_{1-x}Sr_{1+x}CoO_4$ ($0 \leq x \leq 0.5$) system are shown in Fig. 1. The μ_{eff} values calculated from the slopes of these plots in various temperature ranges are given in Table I along with the paramagnetic Curie temperatures. The χ_M^{-1}–T plot of the three-dimensional $La_{0.5}Sr_{0.5}CoO_3$ is well-documented in the literature (*12*). The important features of these results are as follows: (i) At high temperatures, there is an increase in μ_{eff}, the value being close to that expected for low-spin Co^{IV} ($t_{2g}^5\ e_g^0$) and high-spin Co^{3+} ($t_{2g}^4\ e_g^2$). (ii) The $x = 0.5$ samples in both the two- and three-dimensional systems exhibit nearly the same μ_{eff}, although the former is associated with a smaller value of θ, consistent with a smaller number of nearest-neighbor interactions in lower dimensions. (iii) There is a decrease in μ_{eff} with increasing x in the

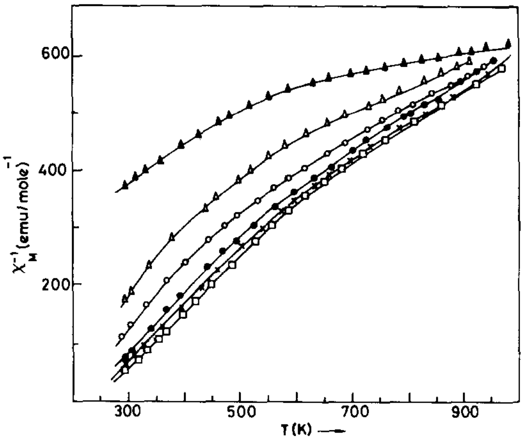

FIG. 1. χ_M^{-1} vs T(K) plots of $La_{1-x}Sr_{1+x}CoO_4$; $x = 0.0$ (closed triangles), $x = 0.1$ (open triangles), $x = 0.2$ (open circles), $x = 0.3$ (closed circles), $x = 0.4$ (crosses), and $x = 0.5$ (boxes).

TABLE I

MAGNETIC PROPERTIES OF A FEW OXIDES OF K_2NiF_4 STRUCTURE

Compound	Temperature range (T)	μ_{eff} (μ_B) observed	μ_{eff} (μ_B) calculated	θ (K)
$La_{1-x}Sr_{1+x}CoO_4$				
$x = 0.5$	>600	3.51	3.67	−15
	<500	2.95		
$x = 0.4$	>600	3.60	3.94	0
	<500	2.95		
$x = 0.3$	>600	3.72	4.20	+100
	<500	2.85		
$x = 0.2$	>600	4.14	4.43	+250
$x = 0.1$	>600	4.17	4.67	+450
$x = 0.0$	>600	6.27	4.90	+1400
$LaSrMn_{0.5}Ni_{0.5}O_4$	>600	5.37	5.19	+190
	<500	6.03	(4.79)	
$LaBaMn_{0.5}Ni_{0.5}O_4$	>600	5.246	5.19	+140
	<500	5.82	(4.79)	
$LaSrMn_{0.5}Co_{0.5}O_4$	>600	6.68	6.92	+175
	<500	7.41	(5.47)	
$LaSrFe_{1-x}Ni_xO_4$				
$x = 0.2$	>300	5.89	5.50	+1350
$x = 0.6$	>300	4.82	4.71	+1000
$x = 0.8$	>300	3.71	3.10	+600

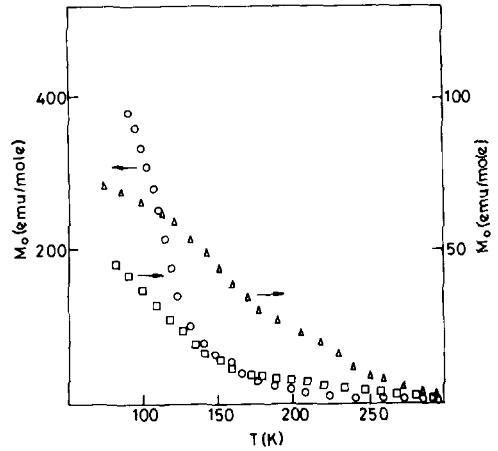

FIG. 2. Magnetization vs temperature of $La_2Sr_2MnCoO_8$ (circles), $La_2Sr_2MnNiO_8$ (boxes), and $La_{0.5}Sr_{1.5}CoO_4$ (triangles).

low-temperature region. Magnetization behavior of $La_{0.5}Sr_{1.5}CoO_4$ is shown in Fig. 2. The extrapolated value of magnetization at 0 K is only 0.5 μ_B, which is much too small compared to 1.5 μ_B shown by $La_{0.5}Sr_{0.5}CoO_3$ (*12, 13*). The similarity of the high-temperature magnetic susceptibility behavior of the two- and three-dimensional oxides with a distinctly different low-temperature magnetization behavior has been noticed by us in other systems as well.

The electrical resistivity data of $La_{0.5}Sr_{1.5}CoO_4$ is compared with that of $La_{0.5}Sr_{0.5}CoO_3$ in Fig. 3. All the compositions of the two-dimensional $La_{1-x}Sr_{1+x}CoO_4$ system are semiconducting with a negative temperature coefficient of resistance (TCR) and a fairly high activation energy. The three-dimensional oxides, on the other hand, behave like metals with a positive TCR. The value of resistivity in the two-dimensional oxides is several orders of magnitude higher than that of the three-dimensional ones. The sign and the magnitude of the Seebeck coefficient at room temperature are similar in the two- and three-dimensional systems. However, in two dimensions, there is a clear anomaly in the Seebeck coefficient near the magnetic ordering temperature (see inset of Fig. 3). Such an anomaly is not seen in the three-dimensional system (*14*).

In $LaCoO_3$, the ground state of cobalt is the low-spin Co^{III} ($t_{2g}^6\,e_g^0$) configuration (*3, 15*). In $LaSrCoO_4$, Demazeau *et al.* (*16*) argue that at low temperatures, there is an equal population of Co^{III} and Co^{3+} ($t_{2g}^4\,e_g^2$) states. In such a configuration the substitution of La^{3+} by Sr^{2+} should lead to the conversion of diamagnetic Co^{III} to paramagnetic low-spin Co^{IV} ($t_{2g}^5\,e_g^0$) causing an increase in μ_{eff}. Since we do not observe this behavior, the spin configuration of Co ions in $LaSrCoO_4$ is likely to be the intermediate-spin Co^{iii} ($t_{2g}^5\,e_g^1$). An examination of the Mössbauer, ferromagnetic resonance, and other physical properties of the $La_{1-x}Sr_xCoO_3$ system (*13, 14*) suggests that the properties are best explained by the existence of Co^{iii} and Co^{IV} ions. A ferromagnetic coupling of such spins would give a magnetization of 1.5 μ_B per cobalt as observed in $La_{0.5}Sr_{0.5}CoO_3$. Intermediate-spin Co^{iii} may be favored in $LaSrCoO_4$ (instead of Co^{III}) by the presence of elongated octahedra.

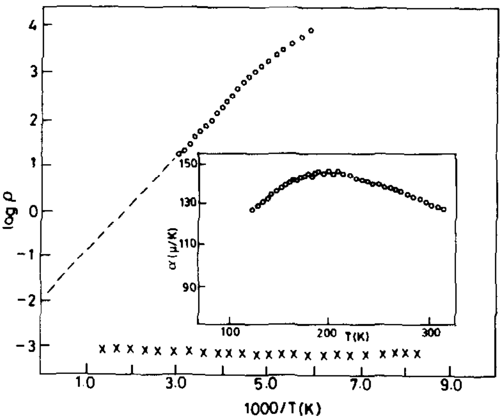

FIG. 3. log ρ vs 1000/*T* (K) plots of $La_{0.5}Sr_{1.5}CoO_4$ (circles) and $La_{0.5}Sr_{0.5}CoO_3$ (crosses). Inset shows the Seebeck coefficient vs temperature (K) plot of $La_{0.5}Sr_{1.5}CoO_4$.

SrO · $(La_{1-x}Sr_xMnO_3)_n$ and $La_{1-x}Sr_xMnO_3$

Oxides of the $La_{1-x}Sr_xMnO_3$ ($0.25 \leq x \leq 0.5$) family are well-known itinerant-electron ferromagnets (*17*). In the SrO · $(La_{1-x}Sr_xMnO_3)_n$ system, on the other hand, for $n = 1$, there is no spontaneous magnetization at low temperatures for any composition in the range $0.3 \leq x \leq 0.7$, although the paramagnetic Curie temperature is positive, indicative of ferromagnetic interactions. The paramagnetic Curie temperature is comparable to that of the corresponding three-dimensional oxides. When $n = 2$ or 3, there are two or three layers of adjacent perovskite layers so that with increasing n, the three-dimensional character increases. Magnetization and electrical resistivity data for the $x = 0.5$ composition are given in Figs. 4 and 5 for different values of n. We see that as the itinerant-electron behavior increases, the value of the magnetization increases. When $x = 0.3$, the low-temperature magnetization of the $n = 2$ composition matches that of the $n = \infty$ composition. Substitution of Sr by Ba or of La by Y does not bring about spontaneous long-range ferromagnetic ordering. Interestingly, Ba or Y substitution brings about an increase in the c/a ratio of the oxide accompanied by a decrease in the paramagnetic Curie temperature.

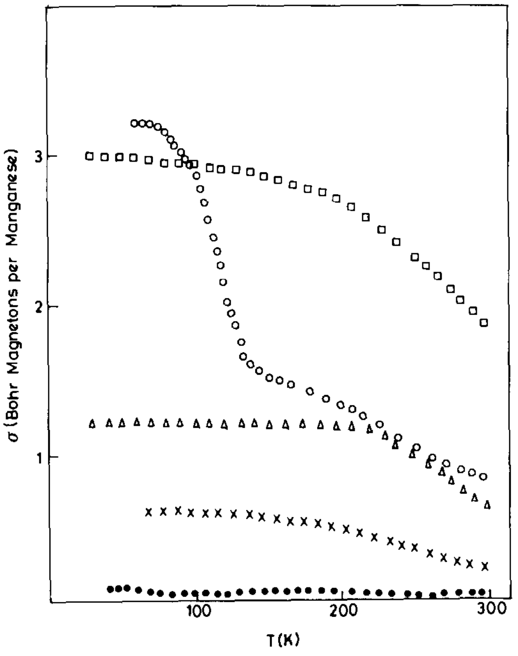

FIG. 4. Magnetization vs temperature plots of $La_{0.5}Sr_{1.5}Mn_3O_{10}$ (closed circles), $LaSr_2Mn_2O_7$ (crosses), $La_{1.5}Sr_{2.5}Mn_3O_{10}$ (triangles), $La_{0.5}Sr_{0.5}MnO_3$ (boxes), and $La_{1.4}Sr_{1.6}Mn_2O_7$ (open circles).

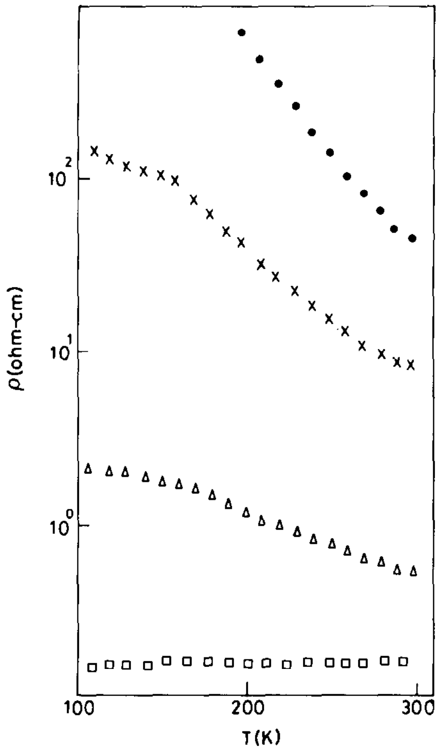

FIG. 5. ρ vs T (K) plots of $La_{0.5}Sr_{1.5}MnO_4$ (closed circles), $LaSr_2Mn_2O_7$ (crosses), $La_{1.5}Sr_{2.5}Mn_3O_{10}$ (triangles), and $La_{0.5}Sr_{0.5}MnO_3$ (boxes).

$LaSrMn_{0.5}T_{0.5}O_4$ and $LaMn_{0.5}T_{0.5}O_3$ (T = Ni or Co)

The $LaMn_{0.5}T_{0.5}O_3$ (T = Ni or Co) systems are the first examples of perovskites which are insulating and ferromagnetic (*18*). The magnetic susceptibility behavior

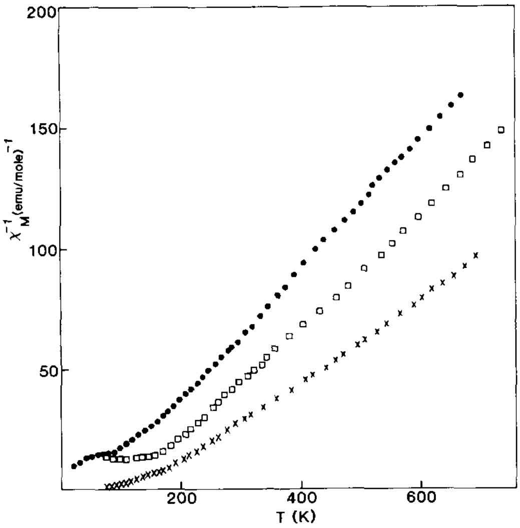

FIG. 6. χ_M^{-1} vs T(K) plots of $La_2Ba_2MnNiO_8$ (circles), $La_2Sr_2MnCoO_8$ (crosses), and $La_2Sr_2MnNiO_8$ (boxes).

of the corresponding two-dimensional compounds $LaSrMn_{0.5}T_{0.5}O_4$ are shown in Fig. 6. The slopes of the high-temperature χ_M^{-1}–T plots of these compounds are nearly identical to those of the corresponding three-dimensional compounds. The μ_{eff} values and the extrapolated paramagnetic Curie temperature are given in Table I. The decreases in θ in lower dimensions is consistent with the decrease in the number of nearest-neighbor magnetic ions. The magnetization behavior of these compounds as a function of temperature is given in Fig. 2. Despite a fairly high value of θ, these compounds do not show any significant magnetization at low temperatures. Since both the three- and two-dimensional oxides are insulating, the absence of long-range ferromagnetic ordering in two dimensions does not seem to be related to the change in the nature of electrical conductivity (metallic vs nonmetallic). The magnetic susceptibility behavior of $LaSrMn_{0.5}Ni_{0.5}O_4$ at low temperatures seems to be consistent with a ferromagnetically coupled $Mn^{3+}Ni^{3+}$ (S = 2) spin pair. In the case of $LaBaMn_{0.5}Ni_{0.5}O_4$, there appears to be a weak maximum around 80 K; *ac* susceptibility measurements are now being carried out on this compound.

$LaNiO_3$ and $LaBNiO_4$ (B = Sr or Ba)

The magnetic susceptibility of $LaNiO_3$ has been explained in terms of Pauli-paramagnetism because of the weak dependence of susceptibility at high temperatures (*19*). There is an enhancement in the susceptibility at low temperatures which can be accounted for in terms of the presence of a small amount of impurity. The metallic conductivity of $LaNiO_3$ is consistent with the itinerant-electron Pauli-paramagnetism picture. The electrical resistivity of LaSr(Ba)NiO_4 is much higher than that of $LaNiO_3$. Magnetic susceptibilities of $LaSrNiO_4$ and $LaBaNiO_4$ at high temperatures are comparable to the susceptibility of $LaNiO_3$ and there is no evidence for magnetic ordering in these oxides or in systems such as $LaSrAl_xNi_{1-x}O_4$. The slopes of the χ_M^{-1}–T plots of $LaBNiO_4$ at low temperatures increase as the electrical resistivity increases (*20*). In Fig. 7 we show the electrical resistivity behavior of the $LaSr_{1-x}Ba_xNiO_4$ system as a function of temperature. In the inset of Fig. 7, we show the log ρ vs log χ_M plots at different temperatures to illustrate that the magnetic susceptibility and electrical resistivity behavior are correlated. That is, the low-temperature increase in the susceptibility in these oxides is an intrinsic property of the system and not due to extrinsic impurities.

Demazeau *et al.* (*21*) suggest, based on ESR spectroscopic studies, that there is a spin equilibrium between the low- and the high-spin Ni^{3+} ions in $LaBaNiO_4$. We have not been able to obtain similar ESR results with any of our samples although the oxygen stoichiometry was good. Moreover, our susceptibility results do not show any evidence of a spin-state transition. In addition, the μ_{eff} values calculated from the

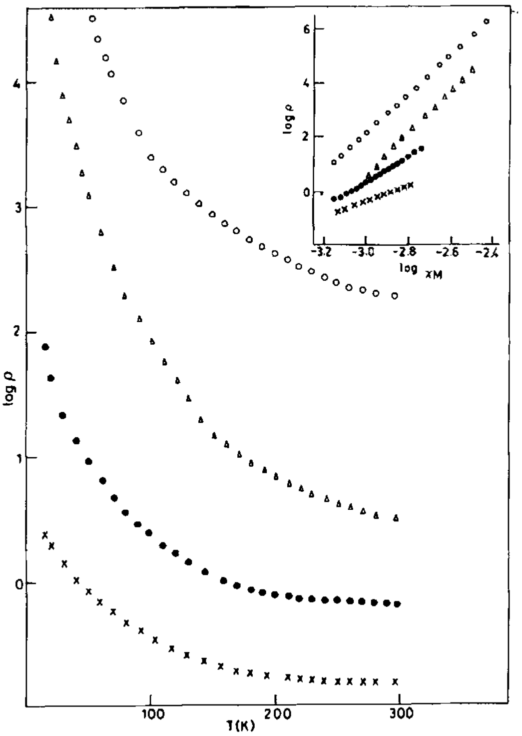

FIG. 7. log ρ vs T (K) plots of $LaSr_{1-x}Ba_xNiO_4$ system; $x = 1.0$ (circles), $x = 0.75$ (triangles), $x = 0.5$ (closed circles), and $x = 0.25$ (crosses). Inset shows the log ρ vs log χ_M plots for the same compounds. The values of ρ and χ_M were measured at the same temperature.

slopes of the χ_M^{-1} vs T plots of $LaSrMn_{0.5}Ni_{0.5}O_4$ and $LaBaMn_{0.5}Ni_{0.5}O_4$ are nearly identical (see Table I), suggesting that the Ni^{3+} ions remain in the low-spin state when Sr is substituted by Ba. We believe that the Ni^{3+} ions in $LaBaNiO_4$ are in the low-spin state at all temperatures. The change in resistivity in the $LaSr_{1-x}Ba_xNiO_4$ system is therefore not likely to be due to changes in the spin state of Ni^{3+} but rather because of an expansion of the lattice.

It has been noted by us that the high-temperature magnetic susceptibilities of $LaNiO_3$ and $LaSrNiO_4$ are similar. The electrical resistivity of the latter is, however, two orders of magnitude higher with a negative TCR. It seems that the increase in the resistivity in $LaSrNiO_4$ arises from increasing localization effects.

$LaSrFe_{1-x}Ni_xO_4$ and $LaFe_{1-x}Ni_xO_3$

The three-dimensional perovskites $LaFeO_3$, $LaCrO_3$, $LaVO_3$ are antiferromagnetic (*6*) with their well-defined ordering temperatures (750, 290, and 140 K, respectively). The corresponding two-dimensional compounds such as $LaSrFeO_4$ and $LaSrCrO_4$ are also antiferromagnetic, although with reduced antiferromagnetic ordering temperatures. The solid solutions of metallic $LaNiO_3$ with insulating $LaTO_3$ (T = Co, Mn, Fe, and Cr) have been studied

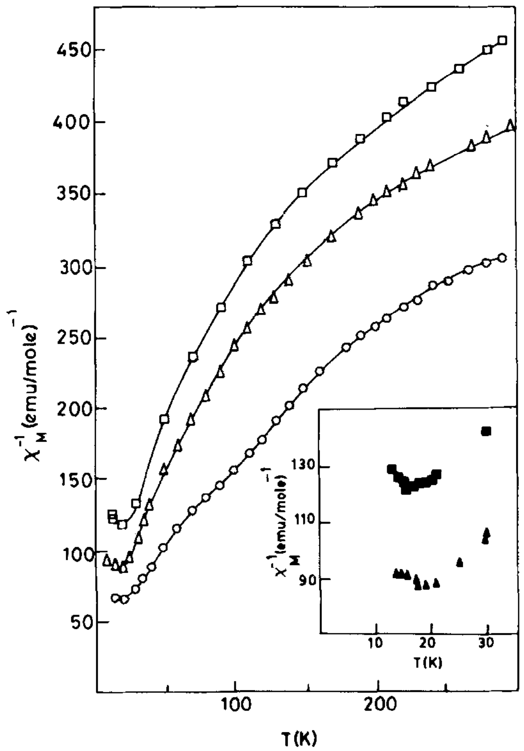

FIG. 8. χ_M^{-1} vs T (K) (T = 300 K) plots of $LaSrFe_{1-x}Ni_xO_4$ system; $x = 0.2$ (open circles), $x = 0.5$ (triangles), and $x = 0.8$ (boxes). Inset shows the χ_M^{-1} vs T (K) plots in the temperature range of 10–30 K of $LaSrFe_{1-x}Ni_xO_4$ ($x = 0.5$ (triangles), $x = 0.8$ (boxes)).

by us with respect to compositionally controlled metal–insulator transitions as well as changes in magnetic ordering (*19, 22, 23*). The $LaFe_{1-x}Ni_xO_3$ system shows a well-defined magnetic ordering temperature up to $x \leq 0.35$; beyond this value of x, the susceptibility shows a broad maximum at lower temperatures. For $x \geq 0.65$, the system becomes metallic with a positive TCR (*23*). The magnetic susceptibility and electrical resistivity data of the $LaSrFe_{1-x}Ni_xO_4$ system are presented in Figs. 8 and 9, respectively. The high-temperature values of μ_{eff} and the paramagnetic Curie temperatures, θ, are listed in Table I. These results show that the Ni^{3+} ions in these oxides are in the low-spin state while the Fe^{3+} ions are in the high-spin configuration; an increase in the Ni^{3+} content decreases the value of θ. There is no evidence for long-range antiferromagnetic ordering, but at low-temperatures there is a susceptibility maximum just as in the corresponding three-dimensional perovskites.

Electrical resistivities of all the members of the $LaSrFe_{1-x}Ni_xO_4$ family show negative TCR; the extrapolated high-temperature value of resistivity is generally $\sim 10^{-2}$ ohm cm. This value is about an order of magnitude less than that found in three dimensions (*23, 24*). Such behavior has been found in systems such as $La_{1-x}Sr_{1+x}CoO_4$ and $LaSrAl_{1-x}Ni_xO_4$ (*20*). The Seebeck coefficient of $LaSrFe_{1-x}Ni_xO_4$ compounds are negative for high values of x and positive for low values of x. The room temperature Seebeck coefficient data for these oxides are given in the inset of Fig. 9 along with those for $LaSrAl_{1-x}Ni_xO_4$, $LaFe_{1-x}Ni_xO_3$, and $LaCr_{1-x}Ni_xO_3$. We see that the lowering of dimensionality changes the composition at which the sign of the Seebeck coefficient changes. There seems to be a correlation between the critical value of the site-percolation threshold (~0.3 for three-dimensional cubic structures and 0.59 for two-dimensional square-planar structures) and the value of x at which the Seebeck coefficient changes sign. Evidence for such percolation effects has

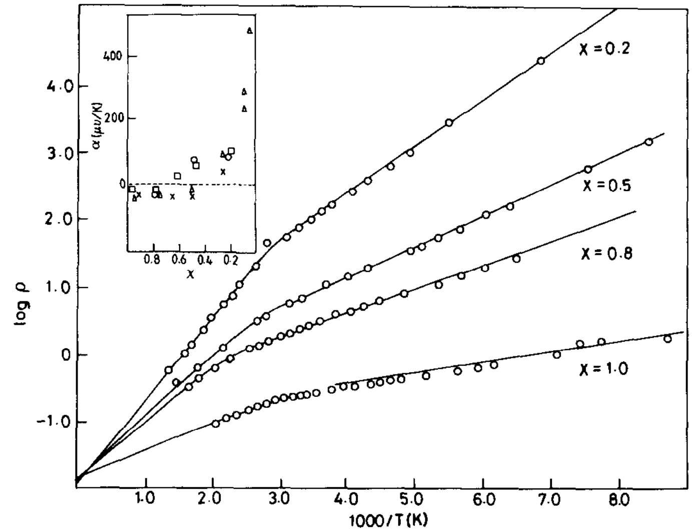

FIG. 9. log ρ vs $1000/T$ plots of $LaSrFe_{1-x}Ni_xO_4$. Inset shows the Seebeck coefficient (α) vs composition (x) plots of $LaFe_{1-x}Ni_xO_3$ (crosses), $LaSrFe_{1-x}Ni_xO_4$ (boxes), $LaCr_{1-x}Ni_xO_3$ (triangles), and $LaSrAl_{1-x}Ni_xO_4$ (open circles).

also been obtained by us in the magnetic susceptibility studies.

$SrRuO_3$ and Sr_2RuO_4

Magnetic susceptibility and electrical resistivity of $SrRuO_3$ and Sr_2RuO_4 are compared in Fig. 10. There is a rise in the suceptibility of Sr_2RuO_4 around 150 K due to the presence of ferromagnetic impurities of $SrRuO_3$; the steep rise persists after heating for long periods at high temperatures (even in the presence of excess Sr). The resistivity of Sr_2RuO_4 is more than an order of magnitude higher than that of $SrRuO_3$ and it has a negative TCR compared to the positive TCR of the latter. Since the magnetic and electrical properties of the ARuO$_3$ systems ($A = Ca^{2+}$, Sr^{2+}, and Ba^{2+}) are sensitive to the nature of the A ion, comparisons between the two- and three-dimensional systems would not be strictly valid. We note, however, that the increased resistivity and the absence of ferromagnetic ordering in $CaRuO_3$ is attributed to the more acidic nature of Ca ions. Since the A ion in Sr_2RuO_4 and $SrRuO_3$ is the same, the effect of decreasing the lattice parameters would be to decrease the resistivity as observed in the $LaSr_{1-x}Ba_xNiO_4$ system. It is noteworthy that while $BaPbO_3$ is metallic, Ba_2PbO_4 is an insulator with a resistivity greater than 10^8 at room temperature. It therefore seems to us that the higher resistivity of Sr_2RuO_4 is a real effect of dimensionality.

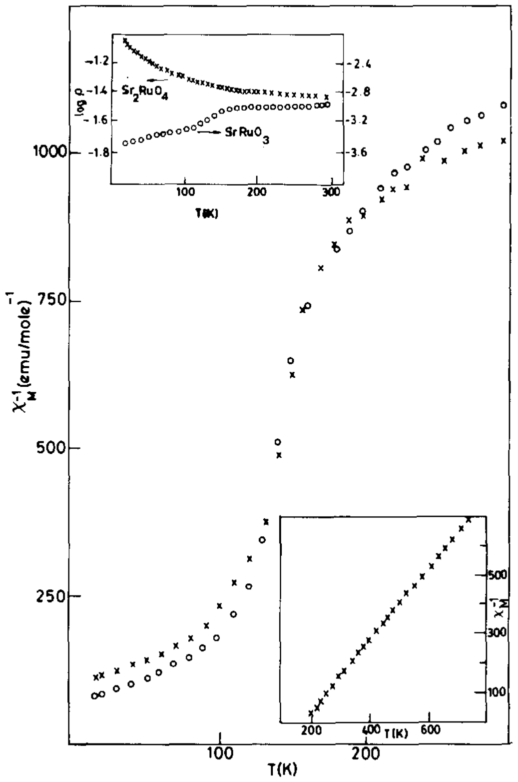

FIG. 10. χ_M^{-1} vs T (K) plots of Sr_2RuO_4 after heating for 24 hr (crosses) and after 7 days of heating (circles). Top inset shows log ρ vs T (K) plots of $SrRuO_3$ (circles) and Sr_2RuO_4 (crosses). Lower inset shows the χ_M^{-1} vs T (K) plot of $SrRuO_3$.

Concluding Remarks

The following general conclusions emerge from the present studies: (i) Perovskite oxides which are ferromagnetic in three dimensions do not show long-range ferromagnetic ordering in the corresponding two-dimensional oxides. This behavior is observed in both itinerant and insulating oxide systems. (ii) The magnetic susceptibility behavior of these oxides in the high-temperature region is not affected by changing the dimensionality. The observed changes in the paramagnetic Curie temperature is consistent with the reduction of the number of nearest neighbors. (iii) Oxides which are metallic in three dimensions with a resistivity less than that corresponding to the "minimum metallic conductivity" (2×10^{-3} ohm cm) show a negative TCR in two dimensions with a resistivity always higher than 10^{-2} ohm cm. (iv) We have not encountered an oxide with the K_2NiF_4 structure which is metallic over a wide range of temperatures. (v) High-temperature extrapolation of the electrical resistivity data indicates that 10^{-2} ohm cm may be the critical value for a change in the sign of TCR to occur in these oxides. So far, there is no solid known with the K_2NiF_4 structure which has a resistivity less than this value. What is somewhat curious in some of these oxide systems is that the magnetic properties reflect itinerant electron behavior while the electrical properties do not. (vi) In K_2CuF_4, Rb_2CrCl_4 (*25*), and such ferromagnetic halides of the K_2NiF_4 structure, the axis of elongation of the MX_6 octahedron and the easy axis of magnetization are in the *ab* plane. In the oxides of K_2NiF_4 structure, the easy axis of magnetization may be the axis of elongation of the BO_6 octahedra (*c* axis). If this indeed be the case, there would be no magnetization, a situation similar to that found in thin ferromagnetic disks with magnetization perpendicular to the plane of the disk. Elongated BO_6 octahedra could also give rise to strong polaronic effects leading to localization of charge carriers.

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