

Research Article **Magnetic and Electric Properties of Sr**_{2-x} Y_x **CoO**₄, (0.50 $\leq x \leq 0.80$) Layered Perovskites

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The electric and magnetic properties of $Sr_{2-x}Y_xCO_4$ layered perovskites have been investigated systematically over the doping range $0.50 \le x \le 0.80$. It was found that both $Sr_{1.5}Y_{0.5}CoO_4$ and $Sr_{1.4}Y_{0.6}CoO_4$ undergo ferromagnetic (FM) transition around 145 K and 120 K, respectively. On the other hand, $Sr_{1.3}Y_{0.7}CoO_4$ and $Sr_{1.2}Y_{0.8}CoO_4$ compounds showed paramagnetic behavior over a wide range of temperatures. In addition, spin-glass transition (T_{SG}) was observed at 10 K for $Sr_{1.3}Y_{0.7}CoO_4$. All investigated samples are semiconducting-like within the temperature range of 10–300 K. The temperature dependence of the electrical resistivity, $\rho(T)$, was described by two-dimensional variable range hopping (2D-VRH) model at 50 K < $T \le 300$ K. Comparison with other layered perovskites was discussed in this work.

1. Introduction

Cobalt oxides have particular interest, not only because of the unique features of Co ions, but also due to their technological applications, such as solid oxide fuel cells and membranes for gas separation [1–5]. Double-exchange interaction between Co⁺³-O-Co⁺⁴ is known to be ferromagnetism (FM) while superexchange (SE) interaction between Co ions with the same oxidation of states is antiferromagnetism (AFM) [6-10]. The spin states of Co ions exhibit several possible spin states: low-spin $t_{2g}^6 e_g^0$ (S = 0), intermediate-spin $t_{2g}^5 e_g^1$ (S = 1), or high-spin $t_{2g}^4 e_g^2$ (S = 2) for Co⁺³ ions and $t_{2g}^5 e_g^0$ (S = 1/2), $t_{2g}^4 e_g^1$ (S = 3/2), or $t_{2g}^3 e_g^2$ (S = 5/2) for Co⁺⁴ ions. All spin states of Co⁺³ or Co⁺⁴ ions are possible because the crystalfield splitting energy of *d*-state electrons (E_{cf}) and Hund energy (E_{ex}) are comparable for perovskite cobaltates. This implies that the energy gap between the t_{2g} and e_g states is small and the electrons in t_{2q} can be thermally exited into the e_a state. As a result, it is difficult to determine the spin states of these ions in cobalt oxides. In other words, complex

magnetic properties arise not only in the original perovskite compounds $R_{1-x}Sr_xCoO_3$ (R is the rare earth ions) which exhibit a rather isotropic 3D arrangement of magnetic ions, but also in layered perovskite systems, $R_{2-x}Sr_xCoO_4$, R = La, Nd or Pr.

The layered-type cobaltates A_2BO_4 with K_2NiO_4 structure are characterized by two-dimensional confinement of the B-O-B network that significantly reduces the e_a electron bandwidth. In turn, electron correlations are strong and can alter the interplay between the different microscopic degrees of freedom (lattice charge, orbital, and spin degrees of freedom). For instance, the magnetic state of the compounds $La_{2-x}Sr_{x}CoO_{4}$ transforms from AFM to FM upon doping for $x \ge 0.6$ [11]. On the other hand, the zero field susceptibility of $Nd_{1-x}Sr_{1+x}CoO_4$ (0.25 $\leq x \leq 0.6$) shows different magnetic transitions [12]. The first is due to spin-glass (SG) transition at 18 K as reported for layered manganites [13]. The magnetic frustration necessary for the formation of a spin-glass state is attributed to the competition between double-exchange $(Co^{4+}-Co^{3+})$ and superexchange $(Co^{3+}-Co^{3+} \text{ or } Co^{4+}-Co^{4+})$ interactions. The second transition is ascribed to a Griffiths

singularity around 210 K, that is, the formation of short-range FM clusters with large spins in the paramagnetic matrix.

Magnetic and transport properties of the layered perovskite system $Sr_{2-x}Y_xCoO_4$, $0 \le x \le 1.0$, were studied by Wang and Takayama-Muromachi [14]. It was found that Sr_2CoO_4 is FM with a transition temperature, T_C = 255 K [14, 15]. Substituting Sr by Y ions, T_C decreases to 150 K for x = 0.50 and the ferromagnetism does not exist when the doping level x exceeds 0.67. All samples are semiconducting-like. Compared to other layered perovskites such as $Nd_{1-x}Sr_{1+x}CoO_4$, a semiconducting behavior was also dominant and the resistivity ($\rho)$ follows an Arrhenius law for x = 0.25, 0.33, and 0.60. This electrical behavior is due to hopping of small lattice polarons, as in the case of manganites. Doping-dependent charge and spin superstructures in $Pr_{2-x}Ca_{x}CoO_{4}$ (0.39 $\leq x \leq 0.73$) and $La_{2-x}Sr_{x}CoO_{4}$ (x =0.40, 0.61) were reported [16]. It was noticed that the charge carriers in these doped cobaltates are strongly localized and so that thermal activated behavior was observed. A systematic increase of the resistivity with decreasing A-site rare earth ionic radius r^{3+} in cobaltates was suggested [17] implying that upon narrowing the bandwidth, the mobility of e_q electrons is decreased.

The number of reports of two-dimensional layered cobaltates (A₂CoO₄) is still relatively small compared with the isotropic three-dimensional perovskite cobaltate (ACoO₃) although they are quite interesting. Therefore, an attempt has been made in this work to throw light on the electrical properties of $Sr_{2-x}Y_xCoO_4$, $0.50 \le x \le 0.80$, and to examine the gradual disappearance of the ferromagnetic property in this range of doping. In addition, a comparison of the investigated system with similar layered perovskites was given. The systematic study of the physical properties of $Sr_{2-x}Y_xCoO_4$ layered perovskites as a function of doping gives an interplay between spin, charge, and lattice/orbital degrees of freedom at the Y^{3+} ion-rich side of the phase diagram.

2. Experimental

Polycrystalline $Sr_{2-x}Y_xCoO_4$ samples $(0.50 \le x \le 0.80)$ were prepared by the solid state method. Stoichiometric mixtures of Y_2O_3 (4N, MTI), SrCO₃ (5N, Cerac), and Co₃O₄ (4N, Aldrich) were well ground and palletized. Then the specimens were calcined at 1100°C and sintered in oxygen at 1050~1100°C for 48 h with intermediate grindings. The sintered samples were annealed in oxygen at 1150°C for 48 h. A polycrystalline $Sr_{1.4}Pr_{0.6}CoO_4$ sample was prepared as in [18]. All investigated samples were confirmed to be single phase by x-ray diffraction (XRD) using CuK α radiation (Bruker D8).

DC magnetization measurements were performed in a SQUID magnetometer (Quantum Design MPMS-5S). Temperature-dependent magnetization (*M*-*T*) curves were measured in field cooled (FC) mode between 5 and 300 K with an applied magnetic field (*H*) of 0.20 kOe. Zerofield cooled (ZEC) magnetization of $Sr_{1.4}Y_{0.6}CoO_4$ and $Sr_{1.4}Pr_{0.6}CoO_4$ were also performed at 0.2 kOe applied field.



FIGURE 1: The main frame represents the field cooled (FC) susceptibility $\chi(T)$ of Sr_{1-x}Y_xCoO₄ (x = 0.50 and 0.60) at H = 0.20 kOe. The inset shows the FC- $\chi(T)$ of the samples of x = 0.70 and 0.80 at H = 0.20 kOe.

The field-dependent magnetization M(H) was measured within $H = \pm 60$ kOe at 5 K. The temperature dependence of the electrical resistivity $\rho(T)$ was measured both on cooling and on heating (from 10 K to 300 K) by the standard dc fourprobe method using a CCR type refrigerator.

3. Results and Discussion

3.1. Magnetic Properties. The temperature dependence of the field cooled (FC) magnetic susceptibility, $\chi(T)$, of $Sr_{2-x}Y_{x}CoO_{4}, 0.50 \le x \le 0.80$, at 0.20 kOe is shown in Figure 1. $\chi(T)$ of both Sr_{1.5}Y_{0.5}CoO₄ and Sr_{1.4}Y_{0.6}CoO₄ (the main frame of Figure 1) show ferromagnetic transitions, $T_C \approx 145$ and 120 K, respectively. Increasing of $x~(\geq 0.70),$ the compounds exhibited PM behavior in a wide range of temperatures as seen in the inset of Figure 1. In addition to the PM behavior of Sr_{1.3}Y_{0.7}CoO₄, spin-glass (SG) transition was observed at 10 K in agreement with other cobaltates [19, 20] and manganites [13]. No traces of a Griffith phase were identified within this range of doping compared with that in similar layered perovskites [12]. It can be suggested that the substitution of Sr by Y ions weakens the ferromagnetic exchange because no long range ordered FM phase was found for the compounds of $x \ge 0.70$. The disappearance of FM in $Sr_{2-x}Y_xCoO_4$ by increasing $x (\ge 0.70)$ can be attributed to two major parts compensated by a decrease of both the magnetic moments in the Co sublattices and the tolerance factor (t < 1). It was suggested previously [14] that both Co³⁺ and Co^{4+} are in intermediate spin states when $x \le 0.67$.

The verification of Curie-Weiss (CW) law was examined for the investigated samples. For x = 0.50 and 0.60 compounds, CW was verified at T > 100 K as shown in the inset and the main frame of Figure 2, respectively. Further increasing x (= 0.70), CW behavior was observed over a wide range of temperatures (see the inset of Figure 2).



FIGURE 2: The temperature dependence of inverse of susceptibility (χ^{-1}) for Sr_{1.4}Y_{0.6}CoO₄ (the main frame) and Sr_{2-x}Y_xCoO₄, x = 0.50 and 0.70 (the inset). The red solid lines represent a fit according to Curie-Weiss law.

A small deviation from CW was found at low temperature for this compound due to SG transition. In contrast, CW of the sample of x = 0.80 (not shown) was verified over the entire range of temperatures. Based on the CW law, Curie-Weiss temperatures, T_{θ} , and the effective paramagnetic moments, $\mu_{\rm eff}$, were calculated and listed in Table 1. It was noted that T_{θ} alters from positive to negative values for the compounds of x > 0.60 [14]. Similar behavior was also observed with the change of x in both $Pr_{1-x}Sr_{1+x}CoO_4$ [18] and $La_{1-x}Sr_{1+x}CoO_4$ [21]. The change of the sign of T_{θ} with x showed that the substitution of Sr^{2+} by Y^{3+} ions changes the magnetic transition from predominantly FM ($x \le 0.60$) to Curie-Weiss PM (x > 0.70). The effective paramagnetic moment μ_{eff} shows moderate changes upon increasing the content of Y ions. Because the value of $\mu_{\rm eff}$ depends on the temperature range over which we estimated it, there is a difference between $\mu_{\rm eff}$ and that in previous report [14].

To give more evidence about fully developed FM phase, the magnetic hysteresis of all samples at T = 5 K is displayed in Figures 3(a) and 3(b). As seen from Figure 3(a), the samples of x = 0.50 and 0.60 are FM and the area of the hysteresis loop of the first compound is larger than that of the second one. A paramagnetic behavior of the samples of x = 0.80 is evidenced by negligible hysteresis although M(H)shows slight deviations from a strict linear behavior. A very narrow hysteresis with no tendency of M to saturate was found for Sr_{1.3}Y_{0.7}CoO₄ due to SG phase (see Figure 3(b)). Remnant magnetization (M_r) , maximum magnetization at 5 K, $(M_{\text{max},5\text{ K}})$, and coercive field (H_{co}) for the investigated samples were tabulated in Table 1. The values of M_{max} and M_r of Sr_{1.5}Y_{0.5}CoO₄ are consistent with previous report [14] and $M_{\text{max},5\text{ K}}$ is comparable with that of similar perovskites [20].

It is better to compare ZFC- $\chi(T)$ of Sr_{1.4}Y_{0.6}CoO₄ with that of Sr_{1.4}Pr_{0.6}CoO₄ at 0.20 kOe as shown in Figure 4. One noticed that the ZFC- $\chi(T)$ of Sr_{1.4}Pr_{0.6}CoO₄ is larger

than that of Sr_{1.4}Y_{0.6}CoO₄ because of the large magnetic moment of Pr³⁺ ions. More than one transition can be seen in Sr_{1.4}Pr_{0.6}CoO₄. One transition is due to Griffiths phase (T_G) around 190 K, where there are magnetic clusters with large spins in the paramagnetic matrix. The second transition was observed around 160 K that can be speculated to short-range FM. Similar transitions were also observed in La_{1-x}Sr_{1+x}CoO₄ [21]. The interplay between these different magnetic transitions provides the basis of the complex magnetic behaviors in layered perovskites. The third transition was identified around 70 K for both Sr_{1.4}Pr_{0.6}CoO₄ and Sr_{1.4}Y_{0.6}CoO₄ and probably is due to SG transition, T_{SG} . This means that T_{SG} transition has the same origin in both systems and is probably due to the transition states of Co ions.

3.2. Electrical Properties. The normalized resistivity values of some selected samples of $Sr_{2-x}Y_xCoO_4$ system are presented in Figure 5. Due to microcracks in the ceramic samples, the absolute values of resistivities may not be very reliable and the room-temperature normalization was used to show the different temperature dependencies of the resistivity of x = 0.60, 0.70, and 0.80. Since the resistivity of the sample of x = 0.50 is close to that of x = 0.60, the resistivity of the first compound was not shown here. The temperature dependence of the resistivity $\rho/\rho_{300 \text{ K}}$ exhibits semiconducting characteristics ($d\rho/dT < 0$), consistent with previous report [14] and the results of similar perovskites [18, 19]. The values of the room temperature resistivity ($\rho_{300 \text{ K}}$) with increasing Y content were given in Table 1. One notes that there is no systematic change of $\rho/\rho_{300 \text{ K}}$ with increasing х.

To describe the resistivity of these compounds, different models were checked. One of them is the Mott's twodimensional variable range hopping (2D-VRH) model [19, 22]:

$$\rho(T) = \rho_0 \exp\left(\frac{T_0}{T}\right)^{1/3},\tag{1}$$

where ρ_0 is constant and T_0 is the variable-range hopping parameter described by Mott and Davis [23]. As shown in Figure 6(a), the temperature range over which 2D-VRH model was verified depends on *x*. Within the temperature range of 50 K $\leq T \leq 300$ K, $\rho(T)$ for the compounds of x = 0.50, 0.6, and 0.80 obeys well 2D-VRH model. The range of the fitting $\ln \rho$ versus $T^{-1/3}$ of Sr_{1.3}Y_{0.7}CoO₄ is narrow (150 K $\leq T \leq 300$ K) compared with that of other studied compounds. The values of the fitting parameters were listed in Table 1.

It was found that the temperature-dependent resistivity $\rho(T)$ of the investigated samples is thermally activated within the range 10 K < $T \leq 50$ K; that is, Arrhenius law was verified:

$$\rho(T) = \rho_{0t} \exp\left(\frac{E}{k_B T}\right),\tag{2}$$

where ρ_{0t} , *E*, and k_B are a material constant, the activation energy, and Boltzmann's constant, respectively. The variation of $\ln(\rho)$ as a function of 1/T is shown for



FIGURE 3: (a) Magnetization hysteresis, M, of the Sr_{2-x}Y_xCoO₄, samples of x = 0.50, 0.60 and 0.80, versus applied field (H) at T = 5 K. (b) M(H) of Sr_{1.3}Y_{0.7}CoO₄ at 5 K.

TABLE 1: Magnetic and electrical parameters of $Sr_{2-x}Y_xCoO_4$, $0.50 \le x \le 0.80$. Listed are the Curie-Weiss temperature, T_{θ} , the effective paramagnetic moment, μ_{eff} , remnant magnetization, M_r , coercive field, H_{co} , maximum magnetization at 5 K, $M_{max,5K}$, the room temperature resistivity, $\rho_{300 \text{ K}}$, the fitting parameters according to 2D-VRH, and the activation energy, E, of the thermally activated part.

<i>x</i> =	0.50	0.60	0.70	0.80
T_{θ} (K)	139.73 ± 0.99	134.12 ± 1.1	-14.32 ± 1.2	-61.91 ± 0.59
$\mu_{\rm eff} \left(\mu_B / {\rm ion} \right)$	4.15 ± 0.29	3.29 ± 0.16	4.24 ± 0.29	4.81 ± 0.25
$M_r(\mu_B)$	1.0	0.33	0.06	0.01
$H_{\rm co}$ (T)	1.6	0.8	0.4	0.1
$M_{\rm max,5K}~(\mu_B)$	1.24	0.87	0.61	0.53
$\rho_{300\mathrm{K}}$ ($\Omega\mathrm{cm}$)	1.03	1.47	14.32	7.18
$\rho_0 (\Omega \mathrm{cm})$	0.02	0.14	1.38	0.33
$T_0 \times 10^3$ (K)	1.0	1.4	73.8	2.6
<i>E</i> (eV)	0.45 ± 0.03	0.53 ± 0.04	1.46 ± 0.09	0.61 ± 0.035



FIGURE 4: Zero-field cooled (ZFC) susceptibility $\chi(T)$ of Sr_{1.4}M_{0.6}CoO₄, (M = Y and Pr) at *H* = 0.20 kOe. The corresponding magnetic transitions indicated by arrows.



FIGURE 5: The normalized resistivity $(\rho/\rho_{300 \text{ K}})$ as a function of *T* for Sr_{2-x}Y_xCoO₄, x = 0.60, 0.70 and 0.80.



FIGURE 6: (a) The variation of $\ln(\rho)$ versus $T^{-1/3}$ for $\operatorname{Sr}_{2-x} \operatorname{Y}_x \operatorname{COO}_4$, x = 0.60, 0.70 and 0.80. The solid red lines represent the fitting of the resistivity at 50 K < $T \le 300$ K according to 2D-VRH model. The top layer shows shows the corresponding T range to $T^{-1/3}$. (b) The variation of $\ln(\rho)$ against 1/T for the same selected samples of $\operatorname{Sr}_{2-x} \operatorname{Y}_x \operatorname{COO}_4$. The solid red lines display the fitting according to Arrhenius law and the top layer shows the corresponding T range to $T^{-1/3}$.

x = 0.60, 0.70, and 0.80 in Figure 6(b). The calculated values of *E* were given in Table 1. One noticed from Table 1 that the values of *E* increase with increasing *x* and Sr_{1.3}Y_{0.7}CoO₄ still exhibited higher *E*, $\rho_{300 \text{ K}}$, ρ_0 , and T_0 compared to that of other investigated samples because this content of Y is the boundary of vanishing ferromagnetism in the studied system.

4. Conclusions

Both magnetic and electrical properties of Sr_{2-x}Y_xCoO₄ system have been investigated as a function of doping x. The FM double-exchange disappears with increasing Y content (x > 0.60) maybe due to the mismatch between Sr and Y ions and the decrease of the oxidation of Co⁴⁺ to Co³⁺ ions. A spin-glass ground state was observed for $Sr_{1.3}Y_{0.7}CoO_4$ followed by a PM phase at high temperatures. The compounds of $x \ge 0.80$ show PM behavior in the entire range of temperatures. With the dilution of Sr^{2+} by Y^{3+} ions in $Sr_{2-x}Y_{x}CoO_{4}$ compound, the Curie-Weiss temperature alters from positive to negative while the effective paramagnetic magnetic moment increases. The electrical conductivities of all studied samples are semiconducting-like. Based on the fitting of $\rho(T)$ data, a change from thermally activated conduction to 2D-VRH one was identified with increasing temperature. Finally, preparation of single crystalline samples with performing an additional experiment was suggested to emphasize the gradual disappearance of FM phase with x in this system.

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