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Dielectric, magnetic, and magnetotransport properties in Sr doped two-dimensional RE2CoO4 (RE=Pr,Eu) compounds

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Abstract

In this work, we report on the studies of dielectric, magnetic, and magnetotransport properties of Sr doped $RE_{2-x}Sr_xCoO_4$ (RE=Pr or Eu, x=0.25-1.5). These compounds were systematically studied in terms of structure, magnetic, magnetotransport, and dielectric constant measurements. Rietveld refinement indicated that these compounds crystallized in K_2NiF_4-type structure with space group *I*4/*mmm*. Lattice parameters increase with Sr doping level. The system changes from paramagnetic to ferromagnetic with increasing Sr doping level and finally becomes ferromagnetic with T_C of 230 K for $Pr_{0.75}Sr_{1.25}CoO_4$ and $EuSrCoO_4$, respectively. The temperature dependence of resistivity indicates that both systems change from semiconductive to metallic with Sr doping. The magnetoresistance (MR) value of 10% at 5 K and 8 T is found for the $EuSrCoO_4$ compound. Large dielectric constants with values of above 2000 were observed in low frequencies for samples with x around 1 for Pr based compounds.

Keywords

Dielectric, magnetic, magnetotransport, properties, doped, two, dimensional, RE2CoO4, compounds

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Dielectric, magnetic, and magnetotransport properties in Sr doped two-dimensional RE_2CoO_4 (RE=Pr, Eu) compounds

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In this work, we report on the studies of dielectric, magnetic, and magnetotransport properties of Sr doped $RE_{2-x}Sr_xCoO_4$ (RE=Pr or Eu, x=0.25–1.5). These compounds were systematically studied in terms of structure, magnetic, magnetotransport, and dielectric constant measurements. Rietveld refinement indicated that these compounds crystallized in K_2NiF_4-type structure with space group $I4/mmm$. Lattice parameters increase with Sr doping level. The system changes from paramagnetic to ferromagnetic with increasing Sr doping level and finally becomes ferromagnetic with T_C of 230 K for $Pr_{0.75}Sr_{1.25}CoO_4$ and $EuSrCoO_4$, respectively. The temperature dependence of resistivity indicates that both systems change from semiconductive to metallic with Sr doping. The magnetoresistance (MR) value of 10% at 5 K and 8 T is found for the $EuSrCoO_4$ compound. Large dielectric constants with values of above 2000 were observed in low frequencies for samples with x around 1 for Pr based compounds. © *2008 American Institute of Physics*.
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I. INTRODUCTION

Compounds with the K_2NiF_4-type structure are well known to exhibit various intriguing physical phenomena, such as high-temperature superconductivity in cuprates, spin-triplet superconductivity in ruthenates, and spin/charge stripes in nickelates and manganites. The discovery of superconductivity and metallic ferromagnetism in $Na_xCoO_2^*H_2O$ (Ref. 1) and Sr_2CoO_4 has generated a great interests in the study of the two-dimensional CoO_2 layer structured compounds. The K_2NiF_4-type Sr_2CoO_4 consists of CoO_2 planes separated by rocksalt-type SrO planes. It has been reported that the Sr_2CoO_4 single-crystalline thin films and polycrystalline samples are metallic ferromagnets with a fairly high Curie temperature (T_C) of 255 K, appreciable magnetic anisotropy, and quasi-two-dimensional transport properties.[2,3] It has been proposed that CoO_2 layers can act as a stage for both spintronic functionality as well as superconductivity.[3] The valence of the Co is 4+ Sr_2CoO_4 and makes the CoO_2 layer very conductive in the same way as that in $SrCoO_3$ three dimensional perovskite compounds. When the Sr is fully replaced by rare earth elements (namely, rare earth based RE_2CoO_4 system), the compound still remains as K_2NiF_4 with two-dimensional CoO_2 layers. In this case, the Co becomes 2+ and makes the system insulative or semiconductive and paramagnetic. This has been reflected in the studies of the Y doped $Sr_{2-y}Y_yCoO_4$ synthesized under high pressure.[2] The Y doping effect indicated that the T_C decreases from 255 K for y=0 to 150 K for y=0.5, and ferromagnetism was not observed for $y \geqslant 0.67$.[2] It would be interesting to see how the physical properties of the RE based CoO_2 layer structured K_2NiF_4-type compounds can be changed by the Sr doping. In this work, we report on the structures, magnetotransport, and dielectric and magnetic properties in $RE_{2-x}Sr_xCoO_4$ (RE=Pr or Eu) compounds.

II. EXPERIMENT

Polycrystalline samples $RE_{2-x}Sr_xCoO_4$ (RE=Pr or Eu, $0.25 \leqslant x \leqslant 1.75$) were synthesized by conventional solid-state reaction method. Highly pure powders of Pr_6O_{11}, Eu_2O_3, $SrCO_3$, and Co_3O_4 were mixed according to appropriate atomic ratios. Samples were sintered in air at 1000 °C for 12 h with several intermediate grindings. The powder x-ray diffraction measurements were carried out on a Phillips PW1730 model diffractometer using Cu $K\alpha$ radiation, and the Rietveld refinement method was applied using the RIETICA program.[4] Magnetic and transport properties were measured using a commercial Quantum design magnetic property measurement system (MPMS) and physical property measurement system (PPMS) between 5 and 330 K in magnetic fields up to 8 T. Dielectric constant and loss were measured using a HP 4194A impendence analyzer in the range from 200 up to 10 MHz.

III. RESULTS AND DISCUSSION

According to the XRD results, it is found that the Sr has higher solubility in the Pr_2CoO_4 system than in the Eu_2CoO_4 system. We have found that single 214 phase can be achieved for x=0.75–1.5 in $Pr_{2-x}Sr_xCoO_4$. However, single phase samples can only be formed for x=1–1.25 for $Eu_{2-x}Sr_xCoO_4$ system. Three typical XRD patterns for Pr and Eu based systems are shown in Fig. 1. It can be seen that all the diffraction peaks in the $Eu_{0.75}Sr_{1.25}CoO_4$ and $PrSrCoO_4$ belong

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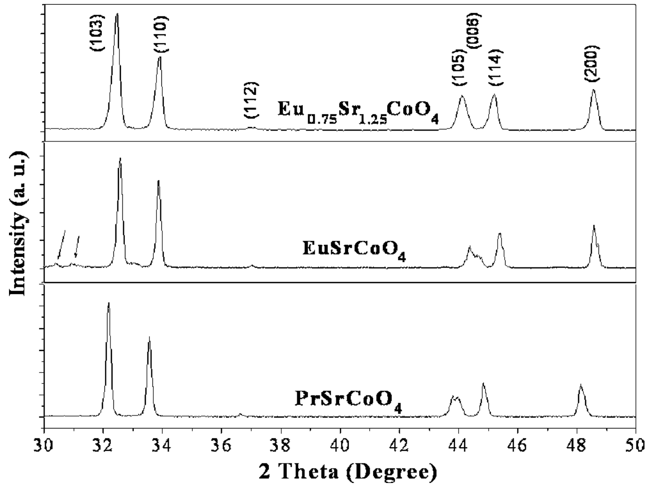

FIG. 1. X-ray diffraction patterns for $RE_{2-x}Sr_xCoO_4$.

to the 214 phase. Two additional unknown tiny reflections are seen in the $EuSrCoO_4$ compound at around 30.15° and 30.25°.

The lattice parameters were obtained from the Rietveld refinement using RIETICA program. It was found that the calculated c lattice parameters for both Pr and Eu systems increases with Sr doping level, while the a lattice parameters do not show significant changes. The increasing of the lattice parameters c with Sr doping level is in agreement with the fact that the sizes of Pr^{4+}, Pr^{3+}, or Eu^{3+} are smaller than that of Sr^{2+}.

The temperature dependence of the field cooled and zero field cooled dc magnetization for five samples measured at 0.2 T is shown in Fig. 2. Generally, the Sr doping changes both systems from paramagnetic to ferromagnetic and enhances the magnetization. Inset shows the inverse susceptibility (χ^{-1}) versus temperature for $Pr_{0.75}Sr_{1.25}CoO_4$. It reveals a ferromagnetic transition with a T_C of about 230 K for this sample (at the point where the concavity of the graph changes). The T_C drops with decreasing Sr content and eventually becomes paramagnetic for $x=0.5$ for the Pr based compounds.

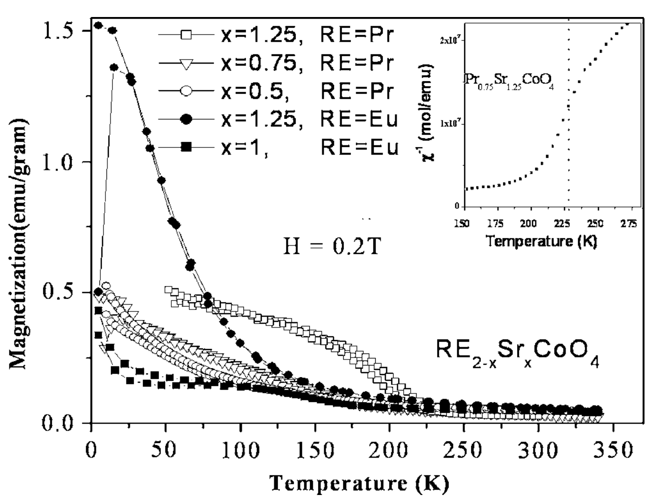

FIG. 2. Temperature dependence of the zero field cooled and field cooled magnetization. Inset shows the inverse susceptibility (χ^{-1}) vs temperature for $Pr_{0.75}Sr_{1.25}CoO_4$, it reveals a T_C of about 230 K for this sample.

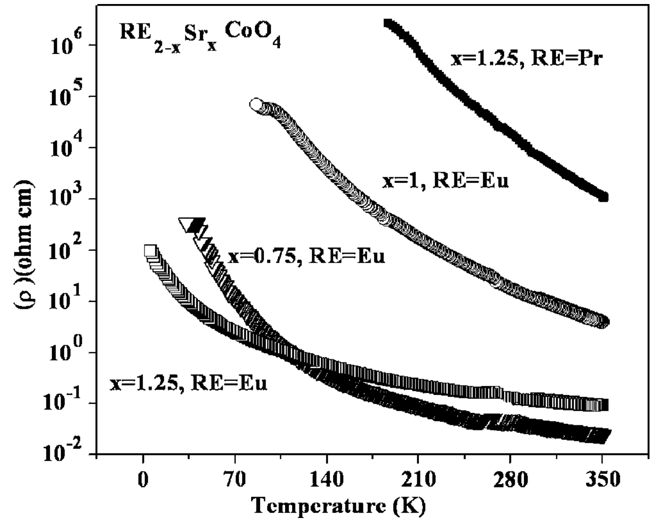

FIG. 3. Temperature dependence of the electrical resistivity.

The inverse molar susceptibility as a function of temperature in the range between 250 and 340 K for both systems was plotted (not shown here). We found that the data above 250 K can be well fitted to the Curie-Weiss law as indicated by the linear fitting. The linear fitting gives $\mu_{eff}=5.23\mu_B$ for Pr based system with $x>0.25$, and $\mu_{eff}=2.63\mu_B$ for Eu based system. For Pr based system with $x<0.25$, the rest of the μ_{eff} values lay between the above mentioned two values. Taking into account the contributions from both Eu and Pr irons, the spin states of the Co^{2+} and Co^{3+} are estimated to be intermediate or high spin states in both Eu and Pr based systems. A detailed study on the assessment of spin states in correlation with the crystal field will be published elsewhere.

The temperature dependences of the resistivity for both Pr and Eu based samples are shown in Fig. 3. The Sr doping reduces the resistivity of the Eu based compounds significantly as compared to the Pr system. The room temperature resistivity for $Pr_{0.75}Sr_{1.25}CoO_4$ is about 10 $k\Omega$ cm, while it is 10^{-1} and 10^{-2} Ω cm for the $Eu_{1.25}Sr_{0.75}CoO_4$ and

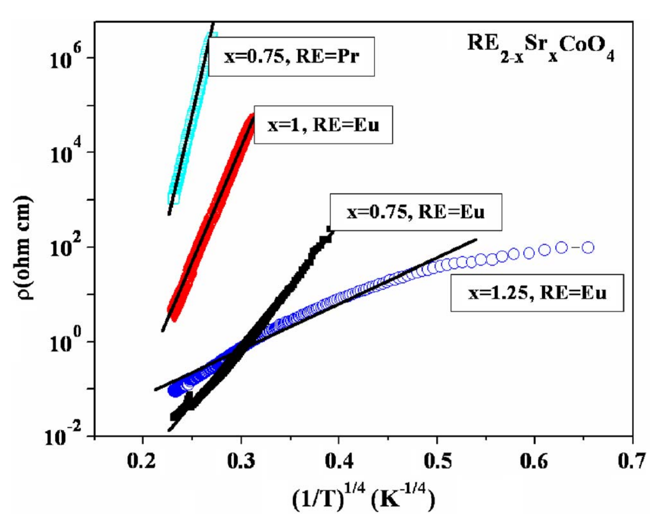

FIG. 4. (Color online) ρ vs $(1/T)^{1/4}$.

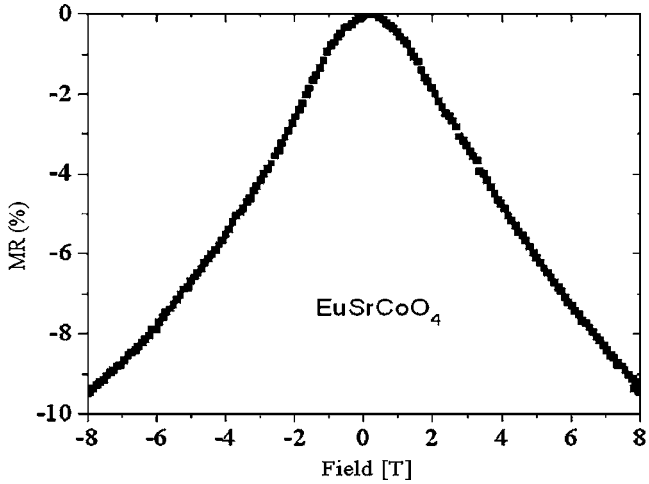

FIG. 5. The field dependence of magnetoresistance at 5 K.

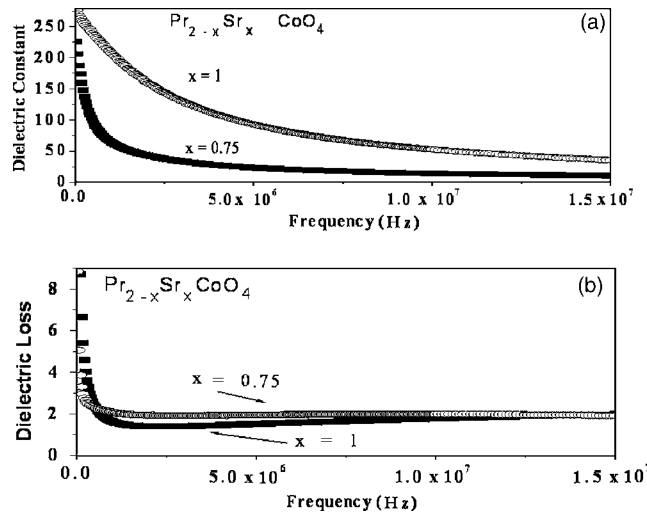

FIG. 6. Dielectric constant (a) and loss (b) of two Sr doped Pr_2CoO_4 samples.

$Eu_{0.75}Sr_{1.25}CoO_4$ samples, respectively. The Eu doped samples with $x<1$ have very low resistivity that is close to the resistivity of undoped Sr_2CoO_4.[2]

The resistivity above 200 K for these samples shown in Fig. 3 can be well fitted by an equation $\rho=\rho_\infty \exp(T_0/T)^{1/4}$, as shown in Fig. 4 for ($T>200$ K, $(1/T)^{1/4}<0.27$). This suggests that the three dimensional variable-range hopping (VRH) mechanism[5] which has been proposed by Mott and observed in manganites compound[6] can explain the electron conducting process in the Pr and Eu based compounds.

The field dependence of magnetoresistance of the $EuSrCoO_4$ sample measured at 5 K is shown in Fig. 5. The MR value reaches 10% at 8 T which is larger than that of the pure polycrystalline Sr_2CoO_4 compounds.[2] There is no clear hysteresis in the graph which is in agreement with the very small coercive field of this compound. This implies that the MR observed in this sample is due to the spin dependent scattering at grain boundaries.

The dielectric properties were studied for Pr based samples with $x=1$ and 0.75, as shown in Fig. 6. The values of the dielectric constant (ε) are over 2000 at low frequency less than 1 kHz (not shown here) and gradually decrease with increasing frequencies. The ε of the $x=1$ sample is greater than that of the $x=0.75$ sample, indicating that the charge induced capacitance in the $x=1$ sample is greater than that of the $x=0.75$ sample, this is in agreement with the trend of their resistivity measurements. The ε remains above 50 at frequency up to 10 MHz for the $x=1$ sample in contrast to about 10 at 10 MHz for the $x=0.75$ sample. The dielectric loss δ drops down to 2–3 at a frequency of 1 MHz and then increase very smoothly with increasing frequencies up to 10 MHz, indicating that extra conducting mechanism maybe responsible for such increase in δ.

IV. CONCLUSION

The $RE_{2-x}Sr_xCoO_4$ (RE=Pr or Eu) compounds crystallize in K_2NiF_4-type structures with space group $I4/mmm$. Lattice parameters increase with Sr doping level. The compounds changed from paramagnetic to ferromagnetic with increasing Sr content and finally become typical ferromagnetic with T_C of 230 K for the Pr based system with $x=1.25$. Both systems changed from semiconductive to metallic with Sr content. Large dielectric constants with values of above 2000 are observed in low frequencies for samples with x around 1 for Pr based compounds. The MR value of 10% is found in the $EuSrCoO_4$ compounds.

ACKNOWLEDGMENT

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