ISSN: 1916-9639
E-ISSN: 1916-9647

Magnetization of $Nd_{1-x}Sr_xCoO_3$ ($0.1 \le x \le 0.5$)

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Special Scientific Research Program Funded by Ankang University(Program No.2007AKXY025)

Abstract

In this paper, the rare earth cobaltite $Nd_{1-x}Sr_xCoO_3$ ($0.1 \le x \le 0.5$) samples were prepared using conventional solid state reaction method. Dc magnetization, ac susceptibility**,** magnetic relaxation of polycrystalline $Nd_{1-x}Sr_xCoO_3$ is investigated in detail. The presence of the typical features of spin/cluster glass state in $x \le 0.2$ samples was well revealed by ac susceptibility. Therefore, our study confirms that the phase separation in $Nd_{1-x}Sr_xCoO_3$ originates from both the ferromagnetic clusters interaction and the spin glasslike phase at low temperature.

Keywords: Magnetization, Spin glass, The phase separation

1. Introduction

The doped cobaltite perovskite oxides, such as $La_{1-x}Sr_xCoO_3$ (LSCO), have received considerable attention because of the unusual phase competition between ferromagnetism (FM) and spin-glass (SG) or cluster-glass (CG) ground states(J. Wu, 2003, 174408, D. N. H. Nam, 1999, 4189 and S. Mukherjee, 1996, 9267). It is well known that the $LaCoO_3$ (LCO) is non-magnetic, while the doped Sr^{2+} in LCO will induce FM clusters due to the double exchange interaction between Co^{3+} and Co^{4+}. Thus, there are FM Sr-rich clusters and non-FM La-rich matrix in LSCO. Recently, Tang *et al.* further confirmed that the phase separation leads to the FM/SG interfaces in LSCO because the LSCO exhibits aging effect and abnormal exchange bias phenomenon (Yan-kun Tang, 2006, 174419). But the origin of the spin glasslike phases remains an open question. Thus, it is interesting to explore the magnetic behavior in the intrinsically phase-separated cobaltites. Stauffer and Leighton pointed out that the phase behavior in $Nd_{1-x}Sr_xCoO_3$ (NSCO) is remarkably similar to that of the LSCO system (Douglas D. Stauffer, 2004, 214414). However, less work has been done in NSCO. Therefore, further research about NSCO system is still necessary to understand these cobaltites more thoroughly.

In this paper, we present a detailed study of the DC magnetization, AC susceptibility, and aging effect in polycrystalline $Nd_{1-x}Sr_xCoO_3$ ($0.1 \le x \le 0.5$) samples. Our results give strong evidences for the existence of a strong tendency toward magnetic phase separation.

2. Experimentl details

The polycrystalline $Nd_{1-x}Sr_xCoO_3$ ($0.1 \le x \le 0.5$) samples were prepared by conventional solid-state reaction. First, the high purity Nd_2O_3 was precalcined in air at 800 ℃ for 4 h. Second, the mixture of Nd_2O_3, $SrCO_3$ and Co_2O_3 powders were thoroughly ground and fired in air at 1000℃ for 48 h and slowly cooled to room temperature. After that, the mixture was reground and pressed into pellets and sintered at 1100 ℃ and 1200 ℃ for 48 h, respectively. X-ray diffraction (XRD) powder pattern was collected using the Bede D^1 XRD spectrometer with Ni-filtered Cu Kα radiation. The room temperature XRD pattern (as shown in Fig.1) shows that the sample is single phase with orthorhombic structure. The DC magnetization and AC susceptibility measurements were carried out using the physical properties measurement system (PPMS) of Quantum Design.

3. Results and discussion

3.1 DC magnetization

Figure 2 shows the dc magnetization of four representative samples as a function of temperature in zero-field cooled (ZFC) and field cooled (FC) procedures. When the Nd^{3+} ion site is doped with Sr^{2+} ion, a proportional number of trivalent Co are converted into teravalent Co resulting in the system phase separation-hole rich ferromagnetic clusters and hole poor anti-ferromagnetic matrix. The double-exchange interaction between Co^{3+} and Co^{4+} gives rise to ferromagnetic clusters. In a wide range of Sr content, M_{FC} (T) increase in magnitude as a result of increasing size and number of ferromagnetic clusters. In the figure we can see that the sample also show a sharp increase in the M_{FC} (T) below temperature T_C. Such data has been report in La (Sr) compounds (J. Wu, 2003, 174408). With increasing Sr content, T_C increase sharply.

The x=0.1 sample present a considerably reduced magnetization and slowly increase in M_{FC} (T) on cooling. A maximum of M_{FC} (T) is present for x=0.1 at low temperature, but not visible anymore for else samples. This feature is similar previous observation in LSCO by Kriener and co-workers (M. Kriener, 2004, 094417). The magnetization of NSCO for x>0.1 exhibit a decrease in below 100 K, the ZFC and FC curves separate with each other. M_{FC} (T) curve increases with decreasing temperature above 30 K, but decreases when the temperature decreases further. Similar behavior was observed in $Nd_{1-x}Ba_xCoO_3$ system (A. P. Sazonov, 2005, 4181), and usually considered due to the magnetic contribution of Nd^{3+} ions at low temperature (below 60 K).

The x=0.1 and 0.15 samples show a peak in the ZFC magnetization at the some temperature T_{SG} .We assume that, in these samples, the peak in M_{ZFC} (T) is governed by SG dynamic transition and usually observed at the freezing temperature of spin glass(J. Wu, 2003, 174408 and D. N. H. Nam, 1999, 4189). In zero-field cooled procedure, magnetic moments of the spins may be frozen in direction energetically favored. The existence of a cusp in the $M_{ZFC}(T)$ for x≥0.2 samples at T_{SG} is then interpreted in term of a competition between the random orientation of the individual clusters and the external magnetic field. In fact, this behavior appears when the clusters freeze into random orientations and form the local anisotropy field in zero-field cooled. Simultaneously the M_{ZFC} (T) shows a cusp at the some temperature that increase with increasing Sr content.

Figure 3 shows the $\chi^{-1}(T)$ as a function of temperature for the sample with x=0.5. The χ (T) data can be fitted to a Curie-Weiss law only in a very narrow temperature interval above T_C. From the corresponding $\chi^{-1}(T)$ fitting, the effective moment per cobalt ion $\mu_{eff\text{-}Co}$ was calculated by subtracting the Nd^{3+} contribution ($\mu_{eff}(Nd^{3+})$ =3.68μ_B) from the total $\mu_{eff} = \sqrt{8c}$. The obtained value, $\mu_{eff\text{-}Co}$ =2.13μ_B, which is smaller than 2.53μ_B in $Nd_{0.5}Sr_{0.5}CoO_3$ previously described in the contributions (M. A. Señarís-Rodríguez, 1999, 281). From this result, we assume that most of the Co^{3+} ions are in the intermediate spin state and the Co^{4+} ions are in low spin. For other samples x=0.2~0.4, the values we obtained range from 1.3 to 1.83μ_B, indicating that in these samples most of the cobalt ions were the low spin configuration in the temperature where we investigated. On the basis of the fitting results our x=0.4,0.5 data exhibit no distinct changes in slope in the χ^{-1}(T) curves, suggesting that thermally driven spin-state transition do not take place in these samples. But x=0.2 and 0.3 samples have a strong positive deviation from that linearly appear below 200K, indicating that spin-state transition take place. On the other hand, the Weiss constant θ, increase with x in good agreement with the result reported by other authors (M. A. Señarís-Rodríguez, 1999, 281).

3.2 AC susceptibility

To explore the magnetic order further we performed the ac susceptibility as a function of temperature at difference frequencies. The presence of the typical features of spin-glass state in NSCO was well revealed by ac susceptibility for these samples. In Figs.4 (a), (b) and 4(c) the real part of the ac susceptibility are plotted for the samples of x=0.15, 0.2 and 0.4, respectively. The x=0.15 and x=0.2 sample are shown as examples as they are representative of the phase described as SG/CG. The x=0.15 sample shows a frequency-dependent peak which occurs very close to that corresponding to the maximum of M_{ZFC} (T). It is well known that such a behavior is the typical features of conventional spin-glass system. In the x=0.2 data also shows a frequency-dependent peak which occurs much higher by 25 K than that point at which the M_{ZFC} (T) reaches a maximum. This situation is similar to previous observation in $Gd_{1-x}Sr_xCoO_3$ (X. G. Luo, 2006, 1029). The x=0.2 sample has a small spontaneous magnetization and the weak frequency dependent peak so that we assume that the sample is cluster-glass.

In Fig.4(c) x=0.4 is a representative of the FM regime, the sample display a strong peak in $\chi'(T)$, follow by a monotonic decrease with decreasing of temperature. This feature has been investigated in $La_{1-x}Sr_xCoO_3$ (J. Wu, 2003, 174408). Therefore the measurements of ac susceptibility give us a detailed insight into the dynamics of freezing.

In order to illustrated more clearly in Fig.5 we provide a “close up” of this peak in $\chi'(T)$, measured with a temperature spacing of 0.2K, for the x=0.2 sample. We can observe clearly that T_f increase with increasing frequency. Moreover, by measuring the variation of T_f with frequencies we can decide if the data is well described by a fit to the conventional critical slowing down of the spin dynamics,

$$\frac{\tau}{\tau_0} \propto (\frac{T - T_{SG}}{T_{SG}})^{-Z\nu} \quad (1)$$

The best fit, for example x=0.2 sample, which is displayed in Fig.5 uses T_{SG} =72.06K, zν=4.02, τ_0 =6.06×10^{-11} s. This result gives strong evidences for the existence of a cluster glass phase. Therefore, ac susceptibility reveal presence of the typical features of spin/cluster glass state in x≤0.2 samples .

To further examine the nature of the SG behavior in $Nd_{1-x}Sr_xCoO_3$, we measured the aging effect of $Nd_{0.85}Sr_{0.15}CoO_3$both ZFC and FC processes. The sample was cooled from 300 K to 10 K in zero fields. After waiting 2000 s, a DC magnetic field (H=1 KOe) was applied and the magnetization M was recorded vs time t, as shown in Figure 6. Obviously, the magnetic moment increased with increasing measured time. This magnetic relaxation curves $M(t)$ in SG system can be described by the stretched exponential form

$$M(t) = M_0 - M_r \exp[-(\frac{t}{\tau_r})^{1-n}] \quad (2)$$

where M_0 (M_0 is the magnetization at t=0) and τ depend on T and t_w, while n is only a function of T. It can be seen from Fig. 6 that the experimental data can be well represented by the Eq. (2). These results of relaxation in ZFC and FC process suggest that the interaction between FM clusters makes contribution to the phase separation and SG behavior in $Nd_{1-x}Sr_xCoO_3$ (Yan-kun Tang, 2006, 012409).

3. Conclusions

In summary, we have presented the magnetization data of $Nd_{1-x}Sr_xCoO_3$ (0.1≤x≤0.5) samples in detail. $Nd_{1-x}Sr_xCoO_3$ samples are typical phase separation system, and shows SG behavior. The DC magnetization, AC susceptibility, aging experiments and fitted results show that the FM clusters interaction makes contribution to the SG behavior. Furthermore, we have observed magnetic relaxation of polycrystalline $Nd_{1-x}Sr_xCoO_3$, and this result gives strong evidences for the existence of a spin glasslike phase between FM and non-FM regions at low temperature.

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ISSN: 1916-9639
E-ISSN: 1916-9647

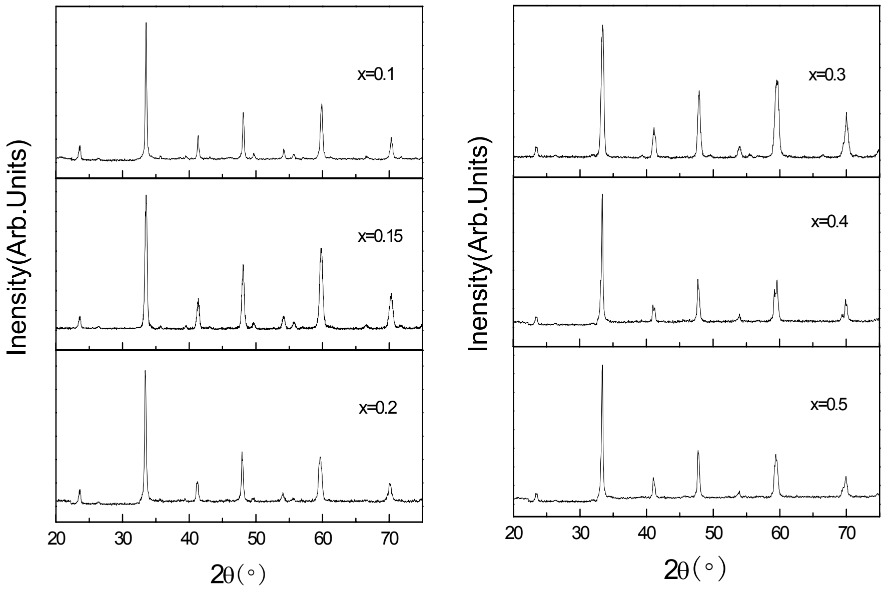

Figure 1.The room temperature X-ray diffraction pattern of $Nd_{1-x}Sr_xCoO_3$ ($0.1 \le x \le 0.5$)

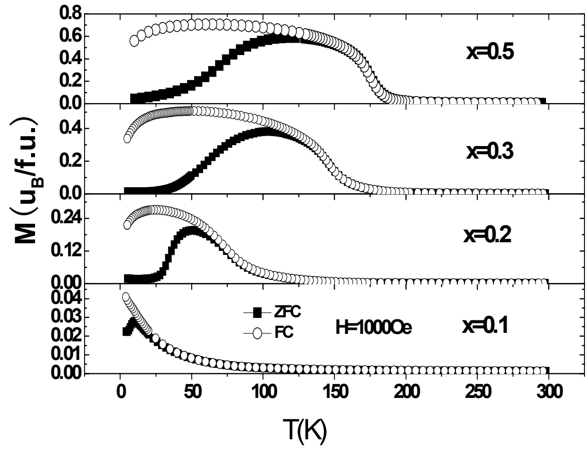

Figure 2. Field cooled (open symbols) and zero field cooled (closed symbols) magnetization of $Nd_{1-x}Sr_xCoO_3$ ($0.1 \le x \le 0.5$) as a function of temperature, measured in the field 1000Oe

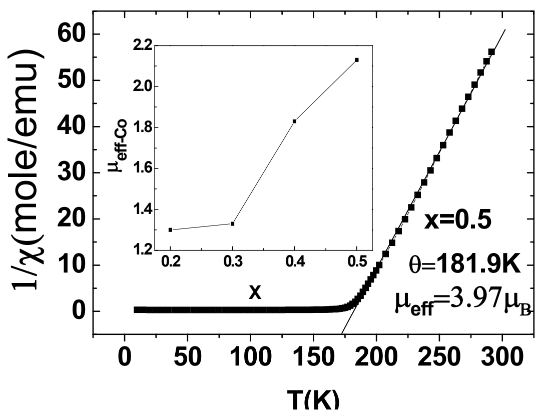

Figure 3. Inverse FC molar magnetic susceptibility as a function of temperature for the sample with x=0.5 .Inset shows the variation of effective magnetic moment with the doping of Sr

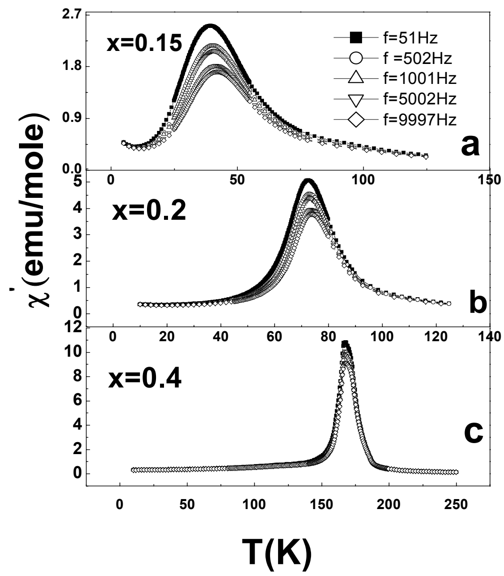

Figure 4. Temperature dependence of the real part of the ac susceptibility of x=0.15, 0.2 and 0.4 samples

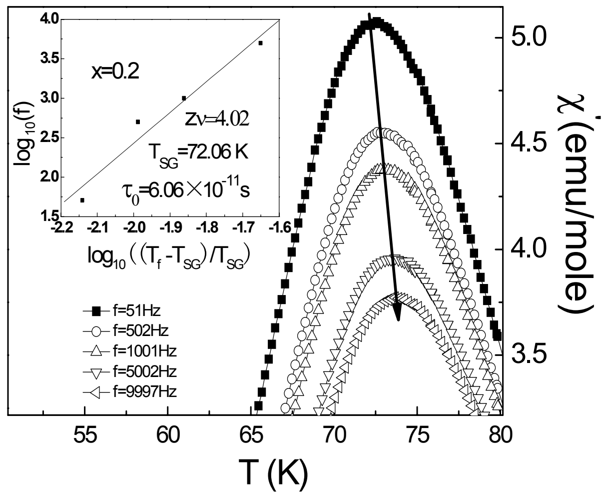

Figure 5. Close up of the temperature dependence of the AC susceptibility at different frequencies of $Nd_{0.8}Sr_{0.2}CoO_3$. Inset shows the fitted result of the experimental data of $\chi'(T)$

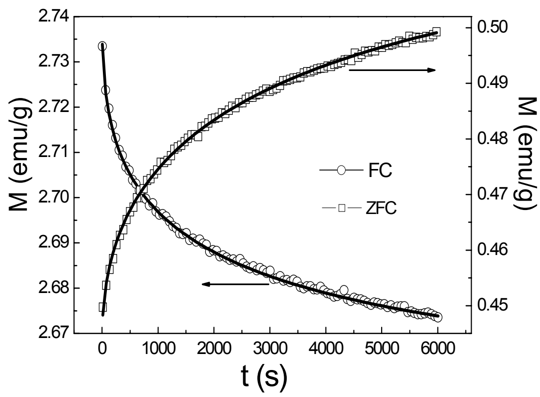

Figure 6. Aging effect of $Nd_{0.85}Sr_{0.15}CoO_3$ both ZFC/FC processes at 10 K