Size-Induced Exchange Bias Effect and Griffiths Phase in Electron Doped $Sm_{0.09}Ca_{0.91}MnO_3$ manganites

S K Giri$^{1,2}$ and T K Nath$^1$

$^1$Department of Physics and Meteorology, Indian Institute of Technology, Kharagpur, West Bengal 721302, India

Email: samir@phy.iitkgp.ernet.in

Abstract. We report here the magnetic properties of electron-doped $Sm_{1-x}Ca_xMnO_3$ manganites with the doping level of $x=0.91$. Exchange bias effect has been observed in $Sm_{0.09}Ca_{0.91}MnO_3$ nanomagnitites system and can be tuned by the strength of cooling magnetic field ($H_{cool}$). The values of exchange bias parameter i.e. exchange bias fields ($H_E$), coercivity ($H_C$), remanence asymmetry ($M_E$) and magnetic coercivity ($M_C$) are found to strongly depend on $H_{cool}$. The larger effective magnetic moments and deviation of inverse susceptibility ($\chi^{-1}$) from Curie-Weiss law indicate the possible existence of Griffiths phase (GP). The effect of size reduction on exchange bias effect and GP is addressed here. The enhancement of exchange bias effect and GP has been argued to be due to the modification of the phase separated state on size reduction.

1. Introduction

The effect of size reduction on the magnetic properties of electron doped manganites is a topic of considerable interest which exhibits various fundamental phenomena such as colossal magnetoresistance (CMR), exchange bias, Griffiths phase and first-order ferromagnetic transition etc. [1-4]. The main identification of the presence of exchange bias (EB) effect is (1) horizontal shift of the field-cooled (FC) magnetic hysteresis loop (2) enhancement of magnetic coercivity as compared to the zero-field-cooled (ZFC) case (3) the existence of the magnetic training effect (TE). One of another interesting behavior near and above the ferromagnetic (FM) transition temperature $T_C$ focuses on the performance of ferromagnetic cluster [1]. This often leads to Griffiths phase (GP) [4] which is

---

2 To whom any correspondence should be addressed.
originally proposed for randomly diluted Ising ferromagnet. The Griffiths phase means the existence of short range ferromagnetic (FM) clusters in paramagnetic regime in the temperature range $T_C \leq T \leq T_G$. Where $T_G$ is Griffiths temperature at which the FM clusters start to nucleate [5] and $T_C$ is the ferromagnetic Curie temperature.

Here we report the magnetic properties of Sm$_{0.09}$Ca$_{0.91}$MnO$_3$ electron doped manganites showing the exchange bias effect below $T_N$ in a field cooled magnetic state and the FM interaction dominates the inhomogeneous phase above $T_C$ i.e. the Griffiths singularity in the PM regime. Here we have reported only two particle sizes; one is S750 (nano) and other one is S1150 (bulk) sample. Our results suggest that the enhancement of exchange bias effect and GP has been argued to be due to the modification of the phase separated state on size reduction.

2. Results and Discussion

2.1. Dc Magnetization

Temperature dependent zero field cooled (ZFC) and field cooled (FC) magnetization curve of S750 sample are shown in the figure 1 (a). The Curie temperature ($T_C$) of the sample is estimated from the $dM/dT$ vs. $T$ plot (not shown here). The interesting behavior is that the bifurcation between the curves (ZFC and FC) starts much above $T_C$ i.e. from 113 K (inset of figure 1 (a)). But for a typical ferromagnet, the bifurcation starts from $T_C$. This feature clearly demonstrates the onset of short range ferromagnetic ordering above $T_C$. The dc magnetization plot of S1150 sample is shown in the figure 1 (b). It is found that the $T_C$ (~ 107 K) value of S1150 sample is lower than that of S750 (~110 K) sample and no anomaly between the ZFC and FC curves of S1150 sample is observed. So, we conclude that $T_C$ of Sm$_{0.09}$Ca$_{0.91}$MnO$_3$ manganites increases with decreasing the particle size.

2.2. Griffiths phase singularity

The downturn nature of inverse magnetic susceptibility ($\\chi^{-1}$) as a function of temperature above $T_C$ is considered as a hallmark of Griffiths phase (GP) singularity [5]. The temperature dependence inverse dc susceptibility of S750 sample at 50 Oe dc magnetic field has been plotted in figure 2 (a). At higher temperature, $\\chi^{-1}$ varies almost linearly with $T$ following the Curie-Weiss law. But with decreasing the temperature, a sharp downturn is observed below a characteristic temperature. The onset of this downturn is denoted as Griffiths temperature ($T_G$) i.e. the temperature where $\\chi^{-1}$ deviates from Curie–Weiss law and below this temperature FM clusters emerge in the PM matrix as described in Griffiths phase system [5]. Generally, GP is univocally characterized by the $\\chi^{-1}(T)$ exponent ($\lambda$) lower than unity, as obtained from the following relation: [6]

$$\\chi^{-1} = (T - T_C^{\text{Rand}})^{-\lambda}, \quad 0 \leq \lambda \leq 1$$

(1)

where $T_C^{\text{Rand}}$ is the critical temperature of the random ferromagnet where susceptibility tend to diverge. In figure 2 (a), the red line shows the fitted data with equation (1). The fitted results shows $\lambda = 0.72$ and $T_C^{\text{Rand}} = 115$ K. The value of $T_C^{\text{Rand}}$ is close to the value of $T_C$. In the paramagnetic regime, the value of $\lambda$ is expected to be zero. Here $\lambda = 0.72$, this concludes that there is a finite size ferromagnetic clusters in the paramagnetic regime above $T_C$ where susceptibility
deviates from the CW behavior. For S1150 sample, the inverse magnetic susceptibility \(\chi^{-1}\) vs. temperature plot at 50 Oe dc magnetic field is shown in the figure 2(b). In this case \(\chi^{-1}\) varies linearly with T following the Curie-Weiss law from \(T_C\) to higher temperature and GP like properties are not observed.

2.3. Exchange bias effect

We have observed the shift in magnetic hysteresis loop when the S750 sample is cooled in a static dc magnetic field. The shift is almost absent when the cooling magnetic field is almost zero. A typical magnetic hysteresis loop at 5 K after cooling the sample with 10 kOe magnetic field is shown in the inset of figure 3 (a). We have calculated the Exchange Bias (EB) parameter by using the standard definition [7]. The value of \(H_E\) and \(H_C\) is consistent with the value of reported other electron doped nanomanganites [8]. We have studied the magnetic cooling field \(H_{cool}\) dependence of exchange bias parameter. Figure 3 (a) shows the variation of exchange bias parameter \(H_E\) with different cooling field at 5 K and hysteresis is measured with \(\pm 5T\) magnetic field. At first, \(H_E\) increases with \(H_{cool}\) up to the cooling field = 10 kOe. Again it shows the decreasing trend with further increasing of \(H_{cool}\). The value of \(M_E\) also increases up to 10 kOe magnetic field and then decreases with further increase of \(H_{cool}\) as shown in the figure 3 (b). It is
found that the value of $H_E$ decreases up to $\sim 11\%$ for the increase of cooling field from 10 to 70 kOe. This suggests that the growth of FM cluster at the high cooling field could contribute partly to the decay of $H_E$. Thus we conclude that at the high cooling field, not only the alignment of degree of FM cluster increases but the size of FM cluster also increases. Again for S1150 sample, we have also studied the magnetic hysteresis loop after the field cooled with 10 kOe magnetic field at 5 K (inset of figure 3 (b)). But there is no loop shift with the cooling field. So we conclude that the EB effect enhances in the nanomanganites S750 sample.

Figure 3. (a) $H_E$ vs. $H_{cool}$ of S750 sample. Inset shows the M-H loop of S750 sample after FC with 10 kOe magnetic field (b) $M_E$ vs. $H_{cool}$ of S750 sample. Inset shows M-H loop of S1150 sample after FC with $H_{cool} = 0$ and 10 kOe.

3. Conclusions

In summary, a systematic investigation on the magnetic properties of the electron-doped Sm$_{0.09}$Ca$_{0.91}$MnO$_3$ manganites has been investigated. A distinct signature of Griffiths phase was found in inverse magnetic susceptibility above $T_C$ only for S750 sample. We have also observed the intrinsic interface EB effect in nanomanganite system with disordered glassy interfaces. Thus for Sm$_{0.09}$Ca$_{0.91}$MnO$_3$ manganites, the Griffiths phase and EB effect gradually enhances with decreasing the particle size and we have explained it on the basis of modification of phase separated state on size reduction.

References

[1] Dagotto E, 2002 Nanoscale Phase Separation and Colossal Magnetoresistance, Springer, Berlin
[2] Tokura Y 2006 Rep. Prog. Phys. 69 797
[3] Meiklejohn W H and Bean C P 1956, Phys. Rev. 102 1413
[4] Griffiths R B 1969 Phys. Rev. Lett. 23 17
[5] Tong P, Kim Bongiu, Daeyoung Kwon, Qian T, Lee Sung-Ik, Cheong S-W, and Kim Bog G 2008 Phys. Rev. B 77 184432
[6] Castro Neto A H, Castilla G, and Jones B A 1998 Phys. Rev. Lett. 81 3531.
[7] Giri S K and Nath T K 2011 AIP Advances 1 032110
[8] Huang X H, Ding J F, Zhang G Q, Hou Y, Yao Y P, and Li X G 2008 Phys. Rev. B 78 224408