Spin glass behavior in Sr2Mn0.7Fe0.3MoO6

Xianjie Wang, Yu Sui, Yao Li, Miao Xu, Wendong Wang, Pan Liu, and Jinke Tang

Articles you may be interested in

Observation of spin-glass behavior in antiperovskite compound Mn3Cu0.7Ga0.3N
Appl. Phys. Lett. **103**, 022405 (2013); 10.1063/1.4813412

Exchange bias and memory effect in double perovskite Sr2FeCoO6
Appl. Phys. Lett. **101**, 142401 (2012); 10.1063/1.4756792

Low temperature magnetically-inhomogeneous states of Sr2FeMoO6– compounds
Low Temp. Phys. **37**, 843 (2011); 10.1063/1.3670028

Structural, electrical, magnetic, and thermal studies of Cr-doped La 0.7 Ca 0.3 Mn 1 x Cr x O 3 (0 x 1) manganites
J. Appl. Phys. **107**, 083905 (2010); 10.1063/1.3342462

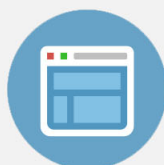Structural and magnetic properties of Co 1 x Fe x Sr 2 YCu 2 O 7 + compounds
J. Appl. Phys. **107**, 063905 (2010); 10.1063/1.3327452

Spin glass behavior in Sr$_2$Mn$_{0.7}$Fe$_{0.3}$MoO$_6$

Xianjie Wang,[1,2] Yu Sui,[1,a)] Yao Li,[2,b)] Miao Xu,[1] Wendong Wang,[3] Pan Liu,[4] and Jinke Tang[4]

[1]*Center for Condensed Matter Science and Technology (CCMST), Department of Physics, Harbin Institute of Technology, Harbin 150001, People's Republic of China*
[2]*Center for Composite Materials, Harbin Institute of Technology, Harbin 150001, People's Republic of China*
[3]*Institute of Microelectronics of Chinese Academy of Sciences, Beijing 100029, People's Republic of China*
[4]*Department of Physics & Astronomy, University of Wyoming, Laramie, Wyoming 82071, USA*

In this paper, we present a detailed study of the dc magnetization, ac susceptibility and aging effect in a polycrystalline Sr$_2$Mn$_{0.7}$Fe$_{0.3}$MoO$_6$ sample. Based on the tetragonal $I\,4/m$ space group, the XRD pattern is well refined, which shows that the sample is crystallized in a single phase double-perovskite structure without impurity phase. The dc magnetization, ac susceptibility, heat capacity, aging experiments and fitting results show the existence of the spin glass behavior in the sample. The spin glass behavior originates from the competition between the ferromagnetic and antiferromagnetic couplings due to the magnetic frustration at the interface of ferromagnetic and antiferromagnetic clusters in the phase separate sample. © *2011 American Institute of Physics*. [doi:10.1063/1.3562889]

I. INTRODUCTION

Recently, much more attention has been paid to the magnetotransport properties of ordered double perovskiten oxides,[1–3] especially Sr$_2$FeMoO$_6$, due to their promising potential for spintronic device applications. Sr$_2$FeMoO$_6$ is a ferromagnetic metal.[1] The magnetic structure of Sr$_2$FeMoO$_6$ was generally described as ferrimagnetic due to the antiferromagnetic coupling of localized spin-up electrons of Fe^{3+} ($3d^5$:t^3_{2g} e^2_g) and itinerant spin-down electrons of Mo^{5+} ($4d^1$:t^1_{2g}). Due to the similarity of electronic configurations between Fe and Mn ions, a lot of investigations have been done for Sr$_2$Fe$_{1-x}$Mn$_x$MoO$_6$, especially the magnetoresistance and magnetic properties of Fe-rich samples.[4–6] When the Fe ions are replaced by Mn, the Mo ions will lose the itinerant $4d^1$ electron, and the Sr$_2$MnMoO$_6$ is an antiferromagnetic insulator.[7,8] Complex magnetic behaviors are observed in the Sr$_2$Fe$_{1-x}$Mn$_x$MoO$_6$ system.

But less work has been done for Mn-rich samples, which should be more helpful for understanding the evolution of the magnetic properties from the ferromagnetic state to antiferromagnetic state. Recently, stronger magnetic frustration effect was observed in Mn-rich samples by Poddar and Mazumdar.[8] When the Fe concentration is about 20%, the isolated Fe cluster start to form and the cluster glass phenomenon is observed in Sr$_2$Mn$_{1-x}$Fe$_x$MoO$_6$. They also suggested that the ferromagnetic order is centered on substituted Fe atoms. Moritomo *et al*. also reported a ferromagnetic order for Fe content as low as 30% in Sr$_2$Mn$_{1-x}$Fe$_x$MoO$_6$ samples.[6] It is well known that the magnetic frustration at the interface of ferromagnetic and antiferromagnetic clusters will result in the spin glass behavior. Poddar *et al*. further proposed that the spin glass will form due to the cationic disorder in Sr$_2$Fe-

MoO$_6$ (Ref. 9). But the origin of the spin glass remains an open question in Sr$_2$Mn$_{1-x}$Fe$_x$MoO$_6$ system. It is interesting to explore the magnetic behavior in the complex magnetic system, and less work has been done in Sr$_2$Mn$_{1-x}$Fe$_x$MoO$_6$. Therefore, detailed investigation about Sr$_2$Mn$_{1-x}$Fe$_x$MoO$_6$ system is still necessary to understand these magnetic properties more thoroughly.

In this paper, we present a detailed study of the dc magnetization, ac susceptibility and aging effect in a polycrystalline Sr$_2$Fe$_{0.3}$Mn$_{0.7}$MoO$_6$ sample. Our results give strong evidence that the spin glass originates from the competition between the ferromagnetic and antiferromagnetic couplings as seen in the Fe-rich and Mn-rich samples, respectively.

II. EXPERIMENT

The Sr$_2$Mn$_{0.7}$Fe$_{0.3}$MoO$_6$ (SMFMO) polycrystalline samples were prepared by conventional solid-state reaction. The mixture of high purity SrCO$_3$, Fe$_2$O$_3$, MnO$_2$, and MoO$_3$ was pressed into pellets and calcined in air at 1173 K for 3 h. The pellets were then reground, pressed and reheated with sintering at 1473 K for 2 h in a stream of 1.5% H$_2$/N$_2$. X-ray diffraction (XRD) powder patterns were collected using a Bede D^1 XRD spectrometer with Ni-filtered CuKα radiation. The room-temperature XRD patterns were refined by the Rietveld method using the program FULLPROF.[10,11] The dc magnetization, ac susceptibility and heat capacity measurements were carried out using the physical properties measurement system (PPMS) of Quantum Design.

III. RESULTS AND DISCUSSION

Figure 1 shows the XRD patterns of Sr$_2$Mn$_{0.7}$Fe$_{0.3}$MoO$_6$ (SMFMO) polycrystalline samples. A good-quality solid solution was achieved and no impurity phase is observed. Based on the tetragonal $I\,4/m$ space group, the XRD pattern is well refined.[12] The cationic order of Fe(Mn) and Mo can
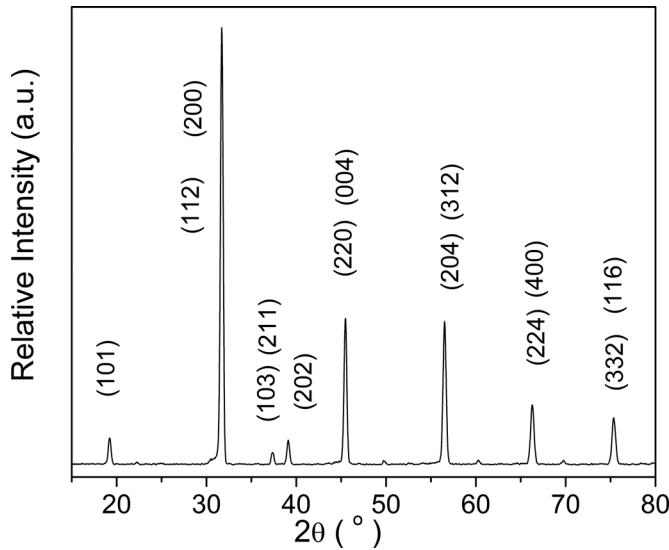
FIG. 1. Room temperature XRD pattern of $Sr_2Mn_{0.7}Fe_{0.3}MoO_6$ sample.

be calculated from the refined results, and its value is about 97%, which is consistent with the results of Feng *et al.*[4] The stronger diffraction peak of (101) also characterizes the high degree of cationic order and the good-quality of the double perovskite structure.[13,14]

It is well known that the Sr_2FeMoO_6 and Sr_2MnMoO_6 are ferromagnetic and antiferromagnetic, respectively. And the ferromagnetic and antiferromagnetic clusters will exist in the SMFMO sample. The interaction between the ferromagnetic and antiferromagnetic clusters will lead to a complex magnetic behavior. Figure 2 shows the dc magnetization as a function of temperature measured in a magnetic field of 100 Oe after zero-field-cooled (ZFC) and field-cooled (FC) processes for SMFMO. Both of the ZFC and FC curves show a peak at about 24 K, although the two deviate below the peak. The paramagnetic Curie temperature, calculated from the Curie−Weiss law ($1/M$ vs T curve, see Fig. 2, inset), is about
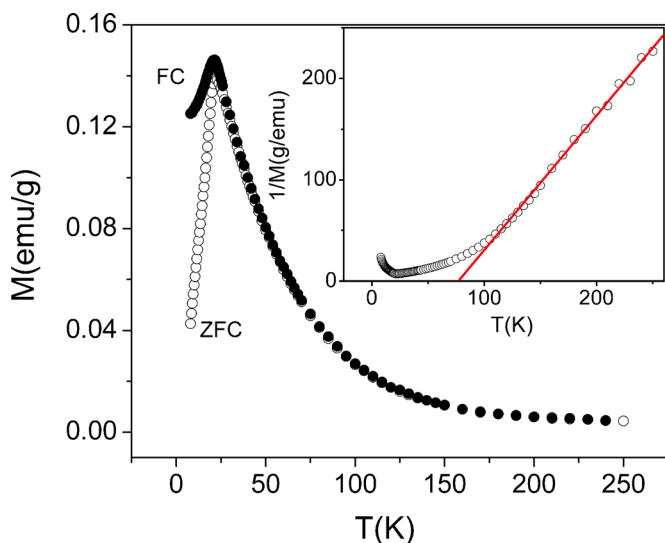

FIG. 2. (Color online) Temperature dependence of the dc magnetization after field cooled (closed symbols) and zero field cooled (open symbols). (b) Inset shows the temperature dependence of the $1/M$.
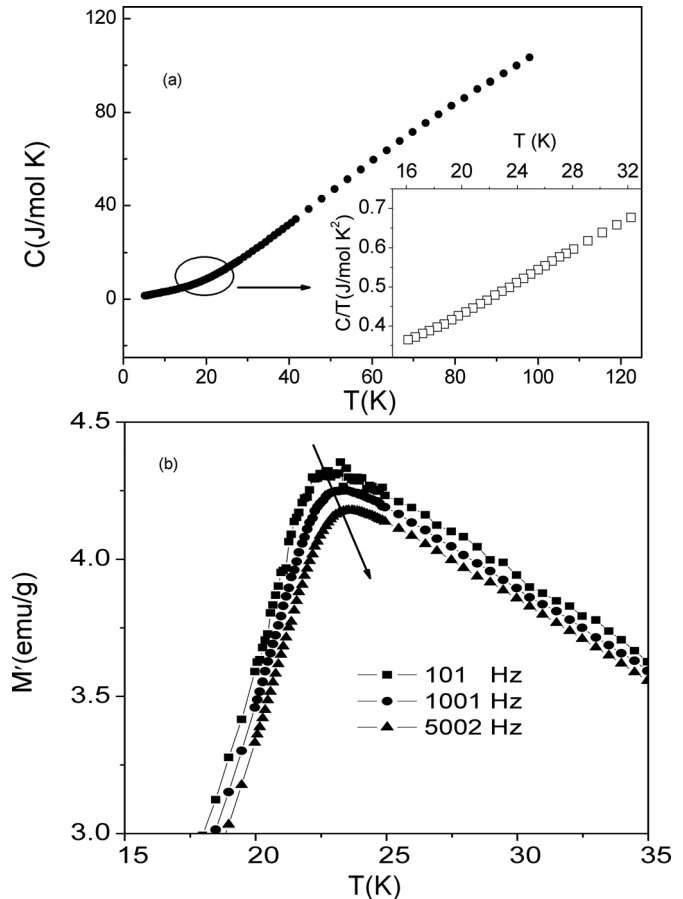

FIG. 3. (a) The heat capacity of $Sr_2Mn_{0.7}Fe_{0.3}MoO_6$ sample; (b) the temperature dependence of the ac susceptibility at different frequencies.

80 K, which implies ferromagnetic coupling. Similar result was also observed by Poddar and Mazumdar.[8] Figure 3(a) shows the specific heat capacity curve of SMFMO. There is no apparent peak near 24 K both in the specific capacity and C/T curves, as shown in Fig. 3(a) and its inset. This suggests what is observed at 24 K is a spin glass or superparamagnetism behavior instead of the antiferromagnetic behavior.

To explore the origin of the magnetic properties, we measure the ac susceptibility of SMFMO at different frequencies. Figure 3(b) shows the ac susceptibility as a function of temperature. The peaks of $M'(T)$ show frequency dependence and occur very close to the peak of ZFC curve, which is usually observed at the freezing temperature of a spin glass. As seen, the peak temperature increases with increasing frequency. It is well known that the frequency dependence in spin-glass system can be described as $\tau/\tau_0 = [(T - T_{SG})/T_{SG}]^{-z}$, where τ is the measured relaxation time, T_{SG} is the critical temperature of spin-glass ordering, z is the dynamical exponent, and τ_0 is the characteristic time scale of the spin glass.[15] This equation fits our experimental data excellently, and the fitting parameters of T_{SG}, z and τ_0 are 20.94 K, 4.28 and 2.70×10^{-12} s, respectively. These results suggest the existence of the spin glass behavior in the sample, which are consistent with the results of dc magnetization and special heat capacity.

To further confirm the nature of the spin glass behavior in SMFMO, we measured the magnetic relaxation and aging curve at 10 K. The sample was cooled from 300 to 10 K in
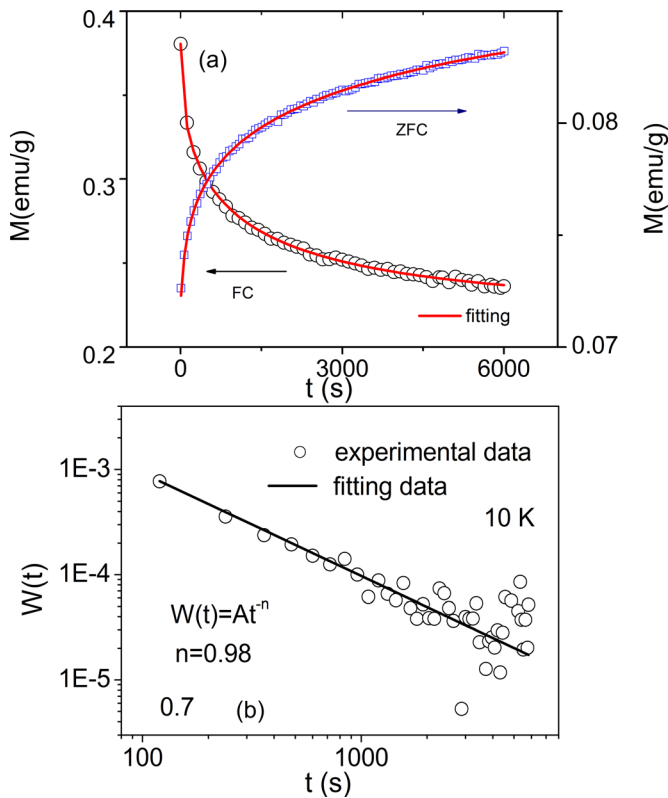
FIG. 4. (Color online) (a) Magnetic relaxation phenomena of ZFC and FC processes at 10 K. Solid line show fitted data. (b) the relaxation rate of FC process.

zero fields. After waiting 600 s, a dc magnetic field ($H = 1$ kOe) was applied and the magnetization M was recorded versus time t, as shown in Fig. 4(a). 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 $M(t) = M_0 - Mr \exp[-(t/\tau)^{1-n}]$,[16] where M_0 (M_0 is the magnetization at $t = 0$) and τ depend on temperature T and waited time t_w, while n is only a function of temperature T. Figure 4(a) shows that the experimental data are well represented by this equation, and the fitted parameters values are $\tau = 4.8 \times 10^3$ s and $n = 0.65$, respectively. We also estimated the aging effect for SMFMO, with the following procedure. The sample was cooled from 300 to 10 K in a magnetic field of 1 kOe. After waiting 600 s, the magnetic field was switched off and the magnetization M was recorded versus time t. The magnetic moment decreases with increasing time, and the FC $M(t)$ curve can be described by $M(t) = M_0 \exp[-(t/\tau)^{1-n}]$, as shown in Fig. 4(a), where M_0 (M_0 is the magnetization at $t = 0$) and τ depend on temperature T and waiting time t_w, while n is only a function of temperature T. The fitted parameter values are $\tau = 1.0 \times 10^3$ s and $n = 0.47$, respectively. These fitted results confirm that the SMFMO exhibits spin glass behavior, which is consistent with the ac susceptibility. The relaxation rate in interacting magnetic particles of FC process, $W(t) = -(d/dt)\ln M(t)$,

decays by a power law $W(t) = At^{-n}$ (Ref. 17). The decay behavior of $W(t)$ can be well described by this equation and $n = 0.98$, as shown Fig. 4(b). These results of relaxation in FC process suggest that the interaction between the ferromagnetic and antiferromagnetic clusters makes contribution to the spin glass behavior in SMFMO.

IV. CONCLUSIONS

In summary, we have presented the magnetic data of $Sr_2Mn_{0.7}Fe_{0.3}MoO_6$ in detail. The dc magnetization, ac susceptibility, aging experiments and fitting results show spin glass behavior in the sample. The spin glass behavior originates from the competition between the ferromagnetic and antiferromagnetic couplings due to the magnetic frustration at the interface of ferromagnetic and antiferromagnetic clusters in the phase separate sample.

[1]K.-I. Kobayashi, T. Kimura, H. Sawada, K. Terakura, and Y. Tokura, Nature (London) **395**, 677 (1998).
[2]R. I. Dass, J.-Q. Yan, and J. B. Goodenough, Phys. Rev. B **68**, 064415 (2003).
[3]X. J. Wang, Y. Sui, Y. Li, L. Li, X. Q. Zhang, Y. Wang, Z. G. Liu, W. H. Su, and J. K. Tang, Appl. Phys. Lett. **95**, 252502 (2009).
[4]X. M. Feng, G. H. Rao, G. Y. Liu, H. F. Yang, W. F. Liu, Z. W. Ouyang, L. T. Yang, Z. X. Liu, R. C. Yu, C. Q. Jin, and J. K. Liang, J. Phys.: Condens. Matter **14**, 12503 (2002).
[5]Z. Yang and L. Ye, J. Phys.: Condens. Matter **17**, 2035 (2005).
[6]Y. Moritomo, H. Kusuya, A. Machida, E. Nishibori, M. Takata, M. Sakata, and A. Nakamura, J. Phys. Soc. Jpn. **70**, 3182 (2001).
[7]E. N. Caspi, J. D. Jorgensen, M. V. Lobanov, and M. Greenblatt, Phys. Rev. B **67**, 134431 (2003).
[8]A. Poddar and C. Mazumdar, J. Appl. Phys. **106**, 093908 (2009).
[9]A. Poddar, R. N. Bhowmik, I. P. Muthuselvam, and N. Das, J. Appl. Phys. **106**, 073908 (2009).
[10]H. M. Rietveld, Acta Crystallogr. **22**, 151 (1967).
[11]J. Rodríguez-Carvajal, Physica B **192**, 55 (1993).
[12]D. Rubi, C. Frontera, J. Nogués, and J. Fontcuberta, J. Phys.: Condens. Matter **16**, 3173 (2004).
[13]Y. Sui, X. J. Wang, Z. N. Qian, J. G. Cheng, Z. G. Liu, J. P. Miao, Y. Li, W. H. Su, and C. K. Ong, Appl. Phys. Lett. **85**, 269 (2004).
[14]Ll. Balcells, J. Navarro, M. Bibes, A. Roig, B. Martínez, and J. Fontcuberta, Appl. Phys. Lett. **78**, 781 (2001).
[15]D. N. H. Nam, R. Mathieu, P. Nordblad, N. V. Khiem, and N. X. Phuc, Phys. Rev. B **62**, 8989 (2000).
[16]G. C. DeFotis, G. S. Coker, J. W. Jones, C. S. Branch, H. A. King, J. S. Bergman, S. Lee, and J. R. Goodey, Phys. Rev. B **58**, 12184 (1998).
[17]M. Ulrich, J. Garcia-Otero, J. Rivas, and A. Bunde, Phys. Rev. B **67**, 024416 (2003).