AIP Journal of Applied Physics

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Citation: Journal of Applied Physics **99**, 08J113 (2006); doi: 10.1063/1.2177390
View online: http://dx.doi.org/10.1063/1.2177390
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JOURNAL OF APPLIED PHYSICS **99**, 08J113 (2006)

Enhanced low field magnetoresistance in Sr_2FeMoO_6-glass composites

Xianjie Wang, Xingquan Zhang, Yu Sui, Jinguang Cheng, Zhiguo Liu, Jipeng Miao, Xiqiang Huang, Zhe Lu, and Zhengnan Qian
Center for Condensed Matter Science and Technology (CCMST), Department of Applied Physics, Harbin Institute of Technology, Harbin 150001, People's Republic of China

Wenhui Su[a]
Center for Condensed Matter Science and Technology (CCMST), Department of Applied Physics, Harbin Institute of Technology, Harbin 150001, People's Republic of China and International Center for Materials Physics, Academia Sinica, Shenyang 110015, People's Republic of China

Jinke Tang
Department of Physics, University of New Orleans, New Orleans, Louisiana 70148

(Presented on 3 November 2005; published online 28 April 2006)

In this paper, we report the enhancement of the low field magnetoresistance in Sr_2FeMoO_6-glass composites with different wt % percents of glass. The crystal structure of Sr_2FeMoO_6 does not change by adding glass, and the glass is most likely located at the grain boundaries. The low field magnetoresistance up to 1 T of Sr_2FeMoO_6-glass composites at 10 K all shows obvious butterfly-shaped field dependence other than the pure Sr_2FeMoO_6 bulk sample. The magnetoresistance of Sr_2FeMoO_6-glass composites at 10 K is enhanced gradually with increasing the glass concentration and reaches 39% with a wt % of 50%. The enhancement of low field magnetoresistance in Sr_2FeMoO_6-glass composites can be well explained by the spin-dependent tunneling at the glass boundaries, and it also allowed us to conclude that the spin polarization of Sr_2FeMoO_6 is at least 80% at low temperature. © *2006 American Institute of Physics*.
[DOI: 10.1063/1.2177390]

I. INTRODUCTION

Intense research activities have been focused on the half-metallic double perovskite Sr_2FeMoO_6 (SFMO) in recent years, because the high value of spin polarization and the high Curie temperature of 420 K in this compound make it an ideal candidate for both technological applications and scientific investigation. Although a large magnetoresistance (MR) effect up to 10% at room temperature was found in the polycrystalline of this material,[1,2] there is nearly no low field MR (LFMR) values in single crystal and single crystalline thin films because the large LFMR of SFMO polycrystalline sample is due to the spin-dependent electron transfer across the grain boundaries.[3,4] So a large LFMR can be obtained by modulating the grain boundary in SFMO.

Various attempts have been made to enhance the LFMR of SFMO by modifying the grain boundaries, such as reducing the grain size,[5] using the grain boundary of a bicrystal,[4] and introducing the second phase of $SrMoO_4$ at the grain boundaries.[6,7] Gupta *et al.* reported on enhanced MR in $La_{0.7}Sr_{0.3}MnO_3$-glass composites, which was attributed to the presence of a glass layer at the grain boundaries in the composites.[8] Similar results have also been observed in other systems of ferromagnet-insulator composites, such as $La_{0.7}Sr_{0.3}MnO_3-Al_2O_3$,[9] $La_{2/3}Sr_{1/3}Mn_3O_3-CeO_2$,[10] $La_{0.67}Ca_{0.33}MnO_3-SrTiO_3$,[11] and $La_{1-x}Ca_xMnO_3-ZrO_2$.[12] However, there are few reports about SFMO using this strategy to enhance the LFMR.

In this paper, we present the magnetotransport properties of SFMO-glass composites. The (Pb–Si–B) glass with lower soft temperature can be dispersed easily into the grain boundaries at high temperature and become a good insulating layer that can improve the spin-dependent electron tunneling. The LFMR value of SFMO-glass composites can be enhanced after the glass addition and it increases with increasing content of glass.

II. EXPERIMENT

The SFMO-glass composite samples were prepared by the following procedure. First, pure SFMO bulk samples were synthesized by a conventional solid-state reaction.[13] Second, the samples were ground in a pure Ar (3 bars) for 1 h and then mixed with appropriate proportion (up to 50 wt %) of Pb–Si–B glass, whose soft temperature is about 400 °C, in pure Ar (3 bar) for 1 h. Finally, the mixed powders were compacted into thin pellets under 450 MPa pressure and then annealed at 430 °C for 20 min in 36% H_2/N_2.

X-ray diffraction (XRD) powder patterns were collected using the Bede D^1 XRD spectrometer with Ni-filtered Cu $K\alpha$ radiation. The surface morphology of the samples was obtained using a HITACHI S-4700 field emission scanning electron microscope. The resistivity measurement by standard dc four-probe method was carried out using a physical properties measurement system (PPMS) of Quantum Design.

III. RESULTS AND DISCUSSION

Figure 1 shows the XRD patterns of the glass added sample (20 wt %) and the pure SFMO sample annealed at

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[a] Author to whom correspondence should be addressed; electronic mail: suiyu@hit.edu.cn

0021-8979/2006/99(8)/08J113/3/$23.00

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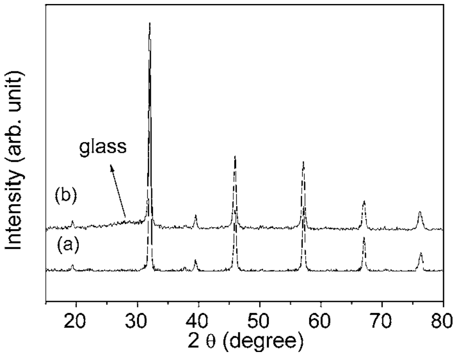

FIG. 1. Room-temperature XRD patterns of (a) pure SFMO and (b) 20 wt % glass added samples annealed at 430 °C in 36% H_2/N_2.

430 °C in 36% H_2/N_2. It is obvious that both samples are of a single double perovskite structure,[13] except for a broad peak of glass in the XRD pattern of the glass added sample. There are no any extra peaks arising from the crystallization of the glass after annealing. These data suggest that the structure of the SFMO grains does not change in the glass added composites. Scanning electron micrographs show that the SFMO grains are randomly distributed and the glass is most likely located at the grain boundaries.

The temperature dependence of the resistivity (ρ) at zero fields for pure SFMO and 10 wt % glass added samples are displayed in the inset of Fig. 2. The ρ of pure SFMO is much lower than that of typical polycrystalline ceramic samples[1] and shows a metallic behavior from 300 to the 60 K, which is similar to that reported in single crystal SFMO.[3] The metallic behavior indicates that the carrier resistance at the grain boundaries is weakened because much of the original interganular barriers in polycrystalline SFMO have been removed by annealing in high H_2 atmosphere. The resistivity of polycrystalline SFMO increases drastically after adding glass and exhibits a semiconducting behavior in the whole temperature range, showing that adding glass has a strong influence on the electrical transport in polycrystal SFMO. From Fig. 2, ρ of composites at room temperature increases drastically with increasing glass content. It indicates that a glass layer is most likely formed at the SFMO grain boundaries, which is consistent with the results of XRD and scanning electron microscopy (SEM) of SFMO-glass composites.

At low temperatures, the intergranular tunneling conductance as a function of temperature was given as:[14–17] $G = G_0(1+P^2m^2)\exp[-(\Delta/T)^{1/2}]$, where P is the spin polarization, and Δ is proportional to the Coulomb charging energy E_C and the barrier thickness. The value of Δ is determined from the slope of the linear part of $\ln G$ vs $T^{-1/2}$ at low temperature. Figure 3 shows the $\ln G$ vs $T^{-1/2}$ curves for different samples. Here, the conductance G is normalized to the value at 5 K. It is clear that $\ln G$ is linear to $T^{-1/2}$ at low temperatures, which means that the intergranular spin-dependent tunneling dominates the conductance of SFMO-glass composites at low temperature. The value of Δ is only 0.005 K for annealed SFMO, but it increases notably with increasing glass content, as shown in the inset of Fig. 3. This also demonstrates that the added glass is most likely located at the grain boundaries, which increases the thickness of the barrier for intergranular tunneling.

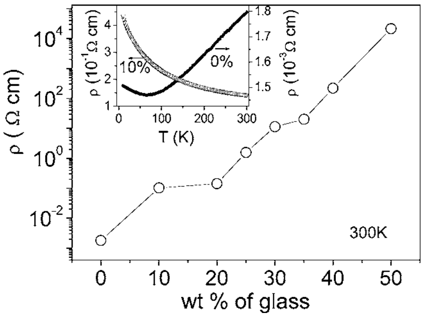

FIG. 2. The variation of the room-temperature resistivity vs glass concentration. The inset shows the temperature dependence of the resistivity at zero fields for annealed SFMO (right axis) and 10 wt % sample (left axis).

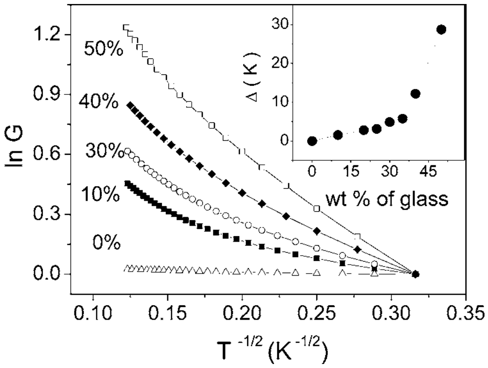

FIG. 3. Logarithmic conductance as a function of $T^{-1/2}$. The inset shows the variation of Δ which is determined from the slope of the linear part of $\ln G$ vs $T^{-1/2}$.

Figure 4 shows the MR of SFMO-glass samples as a function of magnetic field at 10 K. Here, MR is defined as $MR(\%)=[\rho_H-\rho_{\max}]/\rho_H$, where ρ_H and $\rho_{\max}$ are the resistivity of the sample at external field H and the highest resistivity value, respectively. It is well known that the large low field MR of polycrystalline SFMO originates from the spin-dependent tunneling at the grain boundaries.[1,13] The bulk SFMO annealed in high H_2 atmosphere exhibits a very weak MR similar to what was observed in single crystal SFMO.[3] This small MR in annealed SFMO is surely due to the weakening of tunneling barrier at the grain boundaries by the

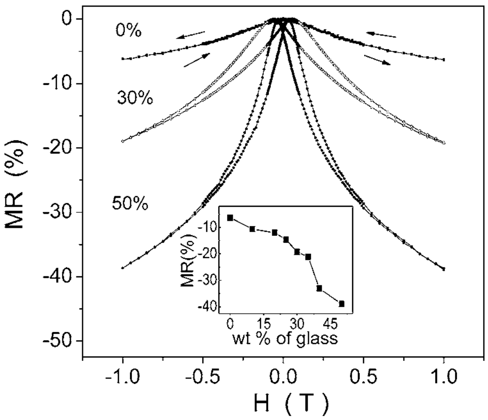

FIG. 4. The magnetic field dependence of MR at 10 K for SFMO-glass sample. The inset shows the MR data at 1 T and 10 K.

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annealing. On the other hand, the LFMR was remarkably enhanced after the glass addition, and the value of MR increases with increasing glass content, as shown in Fig. 4. As discussed earlier, the added glass is located at the grain boundaries and can improve the insulating barrier for the spin-dependent tunneling. This enhancement of MR is also found in some manganite-insulator systems.[9–12] The MR in Sr_2FeMoO_6-glass composites at 10 K shows obvious butterfly-shaped field dependence. The coercive field that was defined as the magnetic field corresponding to $\rho_{\max}$ increases first from near zero to around 600 Oe by adding glass (30 wt %), and then decreases gradually with increasing glass content because the glass layer locating at grain boundaries would weaken the magnetic interaction among the ferromagnetic grains in SFMO-glass composite, and so these grains could reorient more easily at external field.

Although the half-metallic nature of SFMO had been predicted by band calculation, there is little data about the spin polarization (P) of SFMO reported so far. Inoue and Maekawa pointed out that the intergranular tunneling MR in a random granular system was given by[15] $MR=100\% \times[\rho(0)-\rho(H)]/\rho(0)=P^2/(1+P^2)$. The spin polarization P of SFMO can be estimated from the MR data at 10 K and 5 T according to above equation. The value of P for SFMO-glass composite reached 80% at 50 wt % of glass added sample, which is close to that reported by Serrate *et al.* in $Ba_{1.6}Sr_{0.4}FeMoO_6$ under high field up to 50 T.[18] Our data suggest that the spin polarization of SFMO at low temperature is at least 80%, and these provide evidence for high polarization of SFMO.

IV. CONCLUSIONS

Adding glass in polycrystalline SFMO does not change its crystal structure but has a strong influence on the magnetotransport properties. The added insulating glass is most likely located at the grain boundaries of SFMO and improves the barrier for spin-dependent intergranular tunneling. As a result, the LFMR of SFMO-glass composite is greatly enhanced by the addition of glass. Adding nonmagnetic glass is an effective method for enhancing the LFMR of half-metallic materials. The large MR observed in our sample allowed us to determine a lowest limit on the spin polarization of SFMO at low temperature, which is about 80% at 10 K.

ACKNOWLEDGMENT

This work was supported by the National Natural Science Foundation of China (Grant No. 10304004).

[1] K.-I. Kobayashi, T. Kimura, H. Sawada, K. Terakura, and Y. Tokura, Nature (London) **395**, 677 (1998).

[2] M. García-Hernódez, J. L. Martinez, M. J. Martínez-Lope, M. T. Casais, and J. A. Alonso, Phys. Rev. Lett. **86**, 2443 (2001).

[3] Y. Tomioka, T. Okuda, Y. Okimoto, R. Kumai, K.-I. Kobayashi, and Y. Tokura, Phys. Rev. B **61**, 422 (2000).

[4] H. Q. Yin, J.-S. Zhou, J.-P. Zhou, R. Dass, J. T. McDevitt, and J. B. Goodenough, Appl. Phys. Lett. **75**, 2812 (1999).

[5] C. L. Yuan, S. G. Wang, W. H. Song, T. Yu, J. M. Dai, S. L. Ye, and Y. P. Sun, Appl. Phys. Lett. **75**, 3853 (1999).

[6] D. Niebieskikwiat, A. Caneiro, R. D. Sánchez, and J. Fontcuberta, Phys. Rev. B **64**, 180406 (2001).

[7] D. Niebieskikwiat, F. Prado, A. Caneiro, and R. D. Sánchez, Phys. Rev. B **70**, 132412 (2004).

[8] S. Gupa, R. Ranjit, C. Mitra, P. Raychaudhuri, and R. Pinto, Appl. Phys. Lett. **78**, 362 (2001).

[9] L. Yan, *et al.*, J. Appl. Phys. **96**, 1568 (2004).

[10] Ll. Balcells, A. E. Carrillo, B. Martinez, and J. Fontcuberta, Appl. Phys. Lett. **74**, 4014 (1999).

[11] D. K. Petrov, L. Krusin-Elbaun, J. Z. Sun, C. Field, and P. R. Duncombe, Appl. Phys. Lett. **75**, 995 (1999).

[12] D. Das, A. Saha, S. E. Russek, R. Raj, and D. Bahadur, J. Appl. Phys. **93**, 8301 (2003).

[13] Y. Sui, *et al.*, Appl. Phys. Lett. **85**, 269 (2004).

[14] T. Zhu and J. Wang, Phys. Rev. B **60**, 11918 (1999).

[15] J. Inoue and S. Maekawa, Phys. Rev. B **53**, R11927 (1996).

[16] J. Dai and J. Tang, Phys. Rev. B **63**, 064410 (2001).

[17] S. Mitani, S. Takahashi, K. Takanashi, K. Yakushiji, S. Maekawa, and H. Fujimori, Phys. Rev. Lett. **81**, 2799 (1998).

[18] D. Serrate, J. M. De Teresa, P. A. Algarabel, M. R. Ibarra, and J. Galibert, Phys. Rev. B **71**, 104409 (2005).

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