Journal of Magnetics 17(3), 168-171 (2012) http://dx.doi.org/10.4283/JMAG.2012.17.3.168

Irreversibility and Thermoremanent Magnetization in $Y_{0.8}Sr_{0.2}MnO_3$

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(Received 23 February 2012, Received in final form 5 June 2012, Accepted 11 June 2012)

Irreversible magnetization between the zero-field-cooled (ZFC) and field-cooled (FC) states in $Y_{1-x}Sr_xMnO_3$ (x=0 and 0.2) was investigated. $YMnO_3$ and $Y_{0.8}Sr_{0.2}MnO_3$ have a hexagonal structure and the lattice parameter *a* decreases from 7.4408 Å to 7.4327 Å while *c* increases from 12.2244 Å to 12.2287 Å for $YMnO_3$ and $Y_{0.8}Sr_{0.2}MnO_3$, respectively. An anomaly is observed at around 74 K in ZFC and FC magnetization measurements for $YMnO_3$, whereas in $Y_{0.8}Sr_{0.2}MnO_3$ the σ_{ZFC} and σ_{FC} are split at low temperature, indicating glass-like behavior.

Keywords : anisotropy field, glass behavior, irreversibility magnetization, thermoremanent magnetization

1. Introduction

The phase of rare earth manganites, $RMnO_3$, depends on the ionic size of the rare earth ion it contains, we see an orthorhombic phase for ions with a big ionic radius *R* = La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, or Dy; and a hexagonal phase for ions with a small ionic radius *R* = Ho, Er, Tm, Yb, Lu, or Y. Magnetic ordering is observed in both hexagonal and orthorhombic phases, but ferroelectric ordering occurs only in the hexagonal phase. Among multiferroic hexagonal $RMnO_3$ manganites, the $YMnO_3$ compound has been proposed as a new candidate for nonvolatile memory devices due to the single polarization that exhibits little domain-wall motion in thin film, which is important in this type of device [1]. As a multiferroic material, the origin of magnetic ordering in $YMnO_3$ (T_N = 80 K) comes from an indirect exchange interaction between the Mn ions through the O ions in which the angle Mn-O-Mn is close to 180°, while the ferroelectric ordering (T_c = 914 K) results from the buckling of the MnO_5 polyhedral followed by displacements of the Y ions which produce a net electric polarization [2]. However, both the mechanism for driving ferroelectricity and its coupling with magnetic order are not yet well understood.

The exciting properties in this substance emerge upon hole doping in order to attain magnetic ordering of Mn^{3+} and Mn^{4+}. Hole doping is performed by the partial substitution of Y^{3+} with Sr^{2+}. Since the ionic size of Sr^{2+} is similar to that of Y^{3+}, the substitution does not cause significant structural changes. Hole doping weakens the Mn^{3+}-Mn^{3+} antiferromagnetic superexchange interactions and enhances the Mn^{3+}-Mn^{4+} ferromagnetic double exchange interactions. Since this substance has great potential, this work is undertaken in order to investigate the irreversible magnetization between zero-field-cooled (ZFC) and field-cooled (FC) states, which is correlated to the existence of glass behavior in hole doped $Y_{1-x}Sr_xMnO_3$.

2. Experiments

Stoichiometrical polycrystalline samples of $Y_{1-x}Sr_xMnO_3$ (x=0 and 0.2) were prepared by a solid-state reaction method. The starting materials, Y_2O_3, $SrCO_3$ and Mn_2O_3 (purity of 99.99%) were mixed, ground, calcined at 900 °C and reground before pressing into pellets. The pellets were sintered at 1200 °C and 1350 °C in air with intermediate grindings and cooled to room temperature. Powder X-ray diffraction (XRD) was carried out using Rigaku diffractometer Miniflex with Cu-Kα radiation. The lattice constants were calculated using the least square method. The temperature dependence magnetizations σ(T) were presented for ZFC, FC and thermoremanent magnetization (TRM). The ZFC and FC modes were evaluated during the heating process in a field after cooling to 10 K in a zero field and in an applied field, respectively. TRM mode was performed by field cooling the sample from room temperature down to 10 K in a constant field while

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reducing the field to zero, measurements were performed in the heating process.

3. Results and Discussion

The XRD results of the $YMnO_3$ (YMO) and $Y_{0.8}Sr_{0.2}MnO_3$ (YSMO) polycrystalline samples are displayed in Fig. 1. The hexagonal structure peaks are well defined with no secondary phase within experimental error. The lattice parameter *a* decreases from 7.4408 Å to 7.4327 Å and *c* increases from 12.2244 Å to 12.2287 Å for YMO and YSMO, respectively.

The temperature dependence of the ZFC and FC magnetization (referred as σ_{ZFC} and σ_{FC}, respectively) of YMO was presented in Fig. 2. Previous reports showed no anomalies in the magnetic behavior of YMO [2]; however, an anomaly at around 74 K is clearly observed in both the σ_{ZFC} and σ_{FC}, which is consistent with recent report [3]. The σ_{ZFC} and σ_{FC} coincide, except around the anomaly region. Furthermore, the investigation of this anomaly is still underway, through this we hope to elucidate any relationship to the spin fluctuations.

The σ_{ZFC} and σ_{FC} curves of the doped YSMO sample are revealed in Fig. 3. Magnetic transition can be clearly found in the data. Both σ_{ZFC} and σ_{FC} curves reveal a magnetic transition at $T_c \sim 45$ K as obtained by the derivative. Unlike the magnetic behavior of YMO, YSMO exhibits a split of the σ_{ZFC} and σ_{FC}, which is one of the characteristic features of spin glass systems [4]. The irreversibility is related to the magnitude and the temperature variation of the coercivity, which is a measure of the magnetic anisotropy [5]. The spin response to the external field depends on competition between the anisotropy and applied field [6]. When the sample is at $T > T_c$ in zero field, the spin moments are totally random. While cooling the sample through the T_c in ZFC, the spins are locked in random directions for a polycrystalline specimen. If a field is applied to the sample at $T << T_c$, the spins turn in the field direction, and the sample shows magnetization. However, the magnetic anisotropy aligning the spins in a preferred direction resists the rotation of spins to the field direction. During the ZFC process, the magnitude of magnetization depends on the anisotropy of the system. At low applied fields, all spins are not aligned along the direction of field for highly anisotropic systems. When the sample is cooled in a non-zero field (FC process), the spins are locked in a particular direction as soon as the

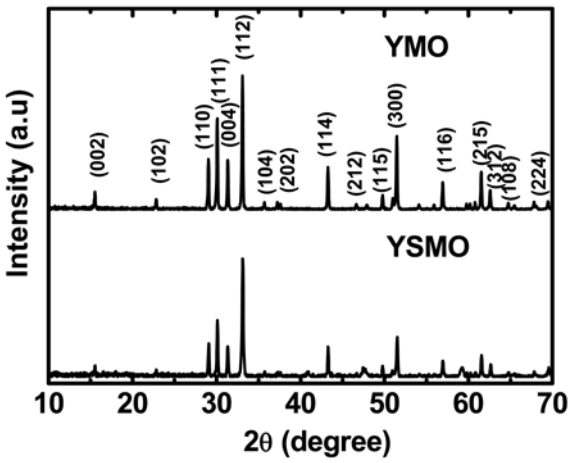

Fig. 1. Powder x-ray diffraction (XRD) patterns for $YMnO_3$ (YMO) and $Y_{0.8}Sr_{0.2}MnO_3$ (YSMO) at room temperature.

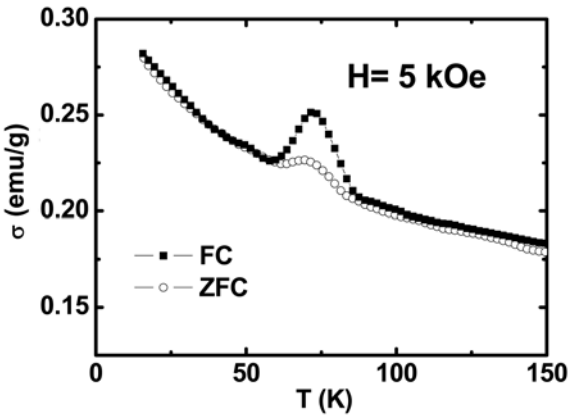

Fig. 2. Temperature dependence of the ZFC and FC magnetization for $YMnO_3$ in an applied field of 5 kOe.

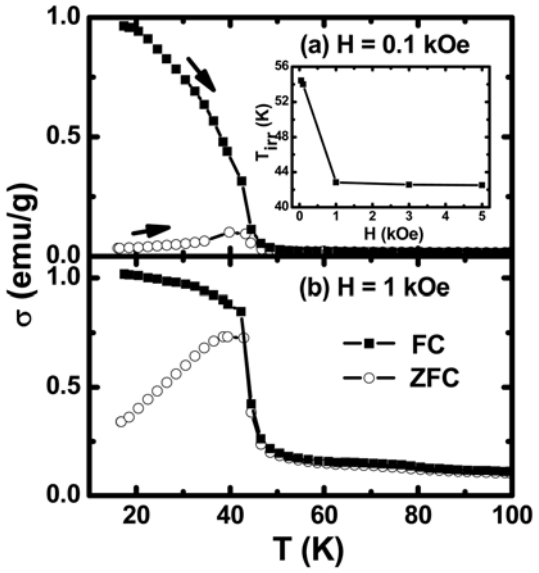

Fig. 3. Temperature dependence of the ZFC and FC magnetization for $Y_{0.8}Sr_{0.2}MnO_3$ with (a) H = 0.1 kOe and (b) H = 1 kOe. Inset shows the field dependence of the irreversibility temperature (T_{irr}).

sample is cooled below T_c. Therefore the spins are frozen in directions energetically favored by their local anisotropy or by the external field when the sample is cooled down in a zero or nonzero field, respectively, leading to a differences between the ZFC and FC magnetizations [7]. Fig. 3 shows that the irreversibility is still observed at low temperature even though the applied field is as high as 5 kOe, which indicates that the anisotropy field, responsible for freezing the spin, is rather large. It was found that the peak in σ_{ZFC} becomes broader at higher fields. The irreversibility temperature (T_{irr}) of σ_{ZFC} and σ_{FC} is shown in the inset of Fig. 3; it shows that T_{irr} shifts to a lower temperature with increasing field.

Magnetic hysteresis measurements were performed at various temperatures. The large coercivity value of 2.9 kOe at 12 K is shown in Fig. 4. As the temperature increases, the hysteresis loop becomes small, and no hysteresis is observed above 50 K. The observed coercivity is shown in the inset of Fig. 4. It can be seen that the coercivity, which is a measure of the magnetic anisotropy [5], decreases rapidly above freezing temperature T_f (temperature of the peak in ZFC). This coercivity can be explained by the freezing of spins below T_f.

Since the σ_{ZFC} and σ_{FC} in YSMO do not coincide each other below ~50 K, it is expected the thermoremanent state is below ~50 K. The thermoremanent state is obtained by cooling a sample in a magnetic field from a temperature at which all spins fluctuate, to one at which some of the fluctuations are quenched. After reducing the field to zero at low temperature during the TRM measurements, the spins try to relax to an initial state. In Fig. 5, the TRM measurements were performed in various applied fields. The field dependence of magnetic behavior and magnetic transition temperatures of σ_{TRM} are in good agreement with those of σ_{ZFC} and σ_{FC}. With increasing applied magnetic field, the value of TRM increases as shown in Fig. 5. Higher applied fields increase the energy of spins to enable them to change their orientations. Consequently, a faster increase of TRM can be observed in higher applied fields. In addition, the TRM versus temperature results depend on the applied field. In low applied fields, the TRM increases slowly with decreasing temperature. However, in high applied fields, the TRM increases faster.

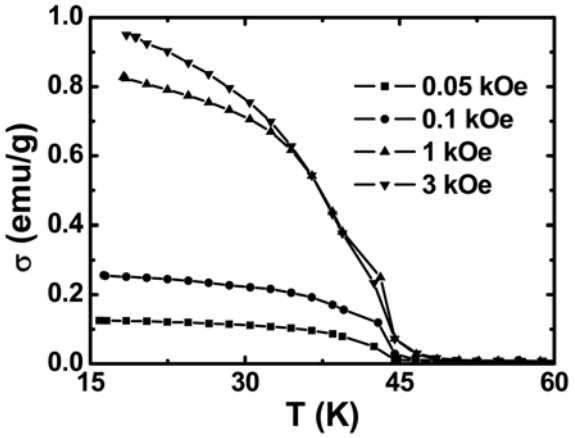

Fig. 5. Temperature dependence of TRM magnetization σ(T) for $Y_{0.8}Sr_{0.2}MnO_3$ with various external applied fields.

From the temperature dependent magnetization data, the isothermal magnetization values at 20 K are selected (Fig. 6). The σ_{ZFC} increases rapidly with increasing field due to the dependence of σ_{ZFC} on the anisotropy field, whereas the σ_{FC} increases linearly with applied field. The comparison between σ_{TRM} and $\sigma_{FC} - \sigma_{ZFC}$ obtained by subtracting experimental curves at 20 K is revealed in the inset of Fig. 6. It shows clearly that σ_{TRM} increases proportionally with the applied field and then saturates for

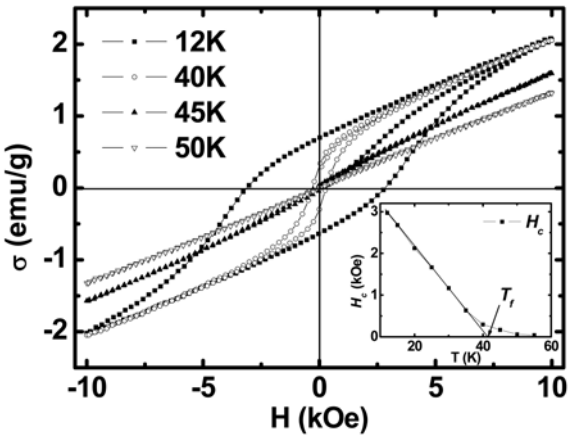

Fig. 4. Magnetic hysteresis of $Y_{0.8}Sr_{0.2}MnO_3$ measured at various temperatures. Inset shows the coercivity (H_c).

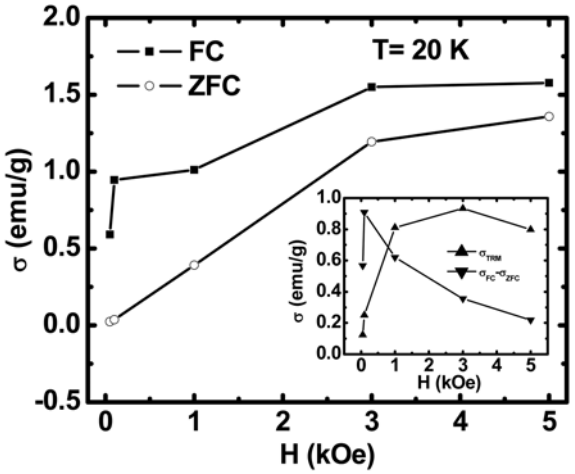

Fig. 6. Isothermal σ_{ZFC} and σ_{FC} of $Y_{0.8}Sr_{0.2}MnO_3$ at 20 K. Inset shows isothermal σ_{TRM} and $\sigma_{FC} - \sigma_{ZFC}$.

H > 1 kOe. The difference of $\sigma_{FC} - \sigma_{ZFC}$ magnetization has a higher value than the σ_{TRM} at low field and decreases with increasing field. Although a coincidence of functions $\sigma_{FC} - \sigma_{ZFC}$ and σ_{TRM} may be expected in spin glass systems, in cluster glass systems a deviation from this behavior may arise due to anisotropy of the clusters possibly associated with their shape and orientation [8].

4. Conclusion

$YMnO_3$ and $Y_{0.8}Sr_{0.2}MnO_3$ polycrystalline samples have a hexagonal structure. An anomaly is observed at ~74 K in temperature dependent magnetization of $YMnO_3$, whereas in $Y_{0.8}Sr_{0.2}MnO_3$ the σ_{ZFC} and σ_{FC} are split at low temperature, indicating glass like behavior. The irreversibility temperature is found to be dependent on the applied field.

Acknowledgment

This work was supported by supported by Hankuk University of Foreign Studies Research Fund of 2011.

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