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Magnetic Memory Effect in $La_{0.6}Sr_{0.4}MnO_3$ Perovskite Nanoparticles

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Abstract: Magnetic memory effect was newly observed in spin-glass magnetic $La_{0.6}Sr_{0.4}MnO_3$ perovskite nanoparticles prepared by sol-gel method. The applied magnetic field and the temperature were changed in defined protocols during the magnetic relaxation process and the magnetic memory effect occured at temperature below blocking temperature which is around room temperature. This magnetic memory effect can be explained by the energy distribution in comparison with strong interaction systems.

Keywords: Perovskite nanoparticles, magnetic memory effect**,** sol-gel, magnetic properties.

1. Introduction

One of the most interesting topics in condensed matter physics is slow dynamics such as non-exponential relaxation, ageing and memory effects. These phenomena are commonly observed in various systems, such as polymers [1], granular materials [2], high T_C superconductors [3] and especially in magnetic nanoparticles [4-8] due to their significance for technological application and fundamental magnetic properties [9]. The magnetic nanoparticles can be considered as individual magnetic moments and the macroscopic picture depends on the interaction between them. In non-interacting system, the dynamics is usually described by the Neel-Brown theory [10,11] with characteristic relaxation time $\tau(T) = \tau_0 \exp(KV/K_BT)$ where K_B, T, K and V are Botlzman constant, temperature, anisotropy constant and the volume of the particles, respectively. The relaxation time changes as temperature is changed and the magnetic moment is frozen or the flip of magnetic moment is blocked when KV/K_BT larger than 25. This blocking state is corresponding to the peak temperature T_B of zero-field-cooled (ZFC) curve. In weak interaction systems such spin-glass, the magnetic reversal process can be explained by a modification of the distribution energy barriers for

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superparamagnetic ones or can be understood by the hierarchical model [4]. In strongly interacting systems, such as hard magnetic ones, the magnetic memory effect in FePd hard magnetic nanoparticles and in exchange-spring magnet was newly reported [12,13], which can be described by a conventional magnetic reversal with an energy barrier distribution. The study of magnetic dynamics can provide some information related to the energy barrier distribution or particles size distribution, which seldom reported in manganites nanoparticles.

Among manganites nanoparticles, $La_{0.6}Sr_{0.4}MnO_3$ with Curie temperature T_C and blocking temperature T_B around room temperature has attracted much attention due to its good magnetic, electrical and catalytic properties and potential applications especially in magnetic storage and spintronics. In this study, we present magnetic memory effect observed in this material which may have important device application.

2. Experimental

$La_{0.6}Sr_{0.4}MnO_3$ nanoparticles were prepared by sol-gel method [14]. Stoichiometric amounts of the nitrate precursor reagents $La(NO_3)_3$, $Mn(NO_3)_2$ and $Sr(NO_3)_2$ were dissolved in distilled water. This solution was mixed with citric acid, forming a stable solution. The stable solution was then heated on a thermal plate under constant stirring at 80 °C for 3 h to eliminate the excess water and to obtain a viscous gel. The obtained gel was dried at 120 °C and then calcinated at 300 °C for 0.5 h. The second calcinated process was carried out at 1000°C for 2 h after an hour milling to obtain final powder products. The morphology and crystal structure of the powders were checked by Scanning Electron Microscope (SEM) and X-ray diffraction (XRD) pattern using CuK_α radiation source in the 2θ scan range from 20° to 70° and reported elsewhere [14]. The average particle sizes of the samples were estimated as 12 nm from the X-ray peak width at half maximum by using the Scherrer's formula. Hysteresis loop and magnetic reversal processes of samples were studied by using a Vibrating Sample Magnetometer (VSM, DMS 880).

3. Results and discussion

Figure 1 shows the field-cooled (FC) and zero-field-cooled (ZFC) magnetization curves of $La_{0.6}Sr_{0.4}MnO_3$ nanoparticles under the field of 100 Oe, which is reported in [14]. It can be seen that sample with Curie temperature of 360 K exhibits spin-glass-like behavior where ZFC curve have maximum at the blocking temperature of above room temperature and FC curve continuously increases as temperature decreases. According to $KV \sim 25\ K_BT_B$, the average anisotropy constant K ~ 1.1 x 10^6 erg/cm^3 can be estimated.

In order to study the dynamics of magnetization of sample, the magnetization relaxation itself and the influence of temperature as well as magnetic field on the relaxation behavior were measured. Figure 2 shows the magnetization relaxation measurements at 260 K and 300 K under various magnetic field change protocols. At 260 K, magnetic field was first applied to 2500 Oe (absolute

value) then the magnetization was started to be recorded for 200 s. After 200 s the magnetic field was changed to 2000 Oe for 100 s then changed back to 2500 Oe. It can be seen on the left-side figure that the magnetization relaxed under the field of 2500 Oe. When magnetic field changed to 2000 Oe, magnetization suddently changed to lower absolute value and relaxed under another relaxation process for 200 s. When magnetic field changed back to 2500 Oe, magnetization was increased in absolute value. The protocol was repeated and the magnetization showed same behavior. Especially, the magnetization at 2500 Oe and at 2000 Oe relaxed at different rate, but they kept almost same value before and after the field change as can be seen in the figure. At 300 K, the protocol was repeated with the field change from 4000 Oe to 3500 Oe and the similar behaviors were observed (right-hand figure). In addition, the magnetization change to higher absolute value and continue to relax under different relax rate when the magnetic field change to 4500 Oe at the end of the process. This behavior can be considered as magnetic memory effect of this material. This effect appears around room temperature providing the high possibility of application of sample in the field of smart materials.

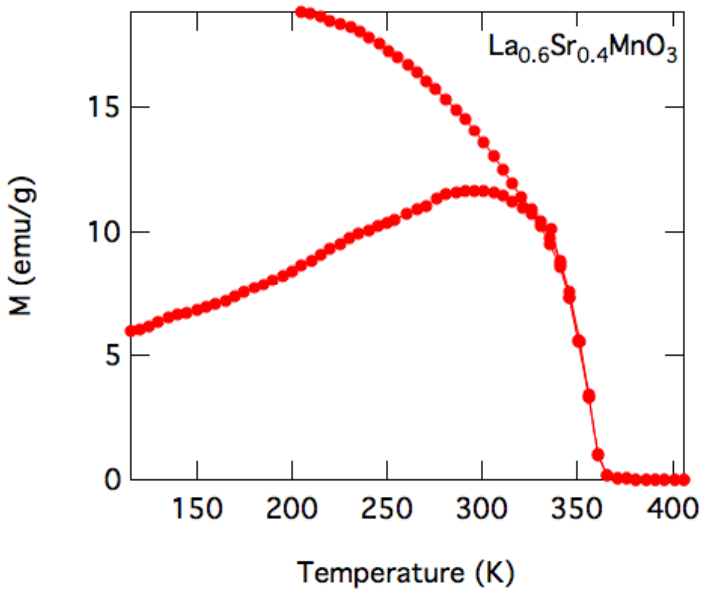

Fig. 1. Temperature dependence of the dc magnetization of $La_{1-x}Sr_xMnO_3$ nanoparticles in a 100 Oe field for FC and ZFC processes.

Figure 3 shows the ZFC relaxation of magnetization under constant field of 20 Oe but with changing temperature protocol. The temperature first was room temperature then changed to 280 K after a waiting time of 300 s then changed back to 300 K. The magnetization was started to be recorded after temperature was stabilized. However, during the recording, the temperature was not stable due to the measurement environment of VSM, so the manetization relaxation seems to be not so smooth. It can be seen in the figure that the magnetization at 300 K continued to relax at same relaxation rate when temperature changed back from 280 K. However, when the temperature changed to higher one of 310 K and changed back to 300 K, the magnetization did not keep the relaxation tendency.

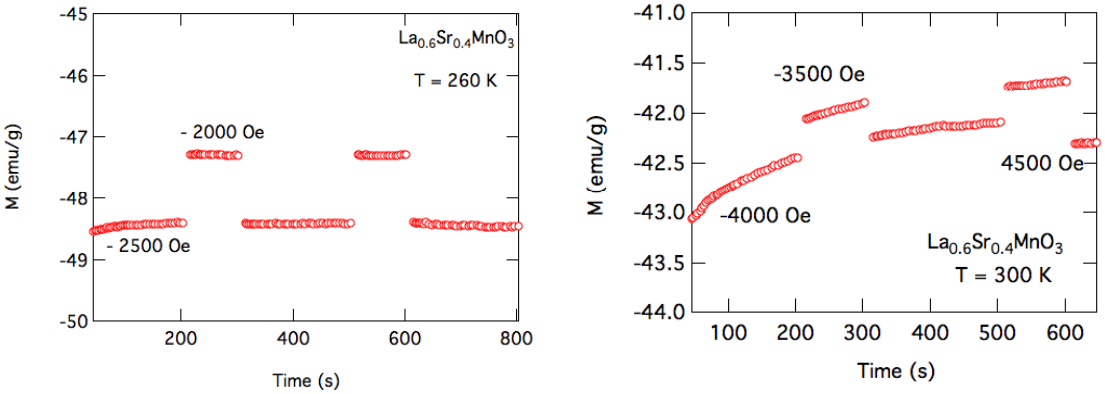

Fig. 2. Magnetization relaxation of $La_{0.6}Sr_{0.4}MnO_3$ nanoparticles at various protocols of changing temperature and magnetic field.

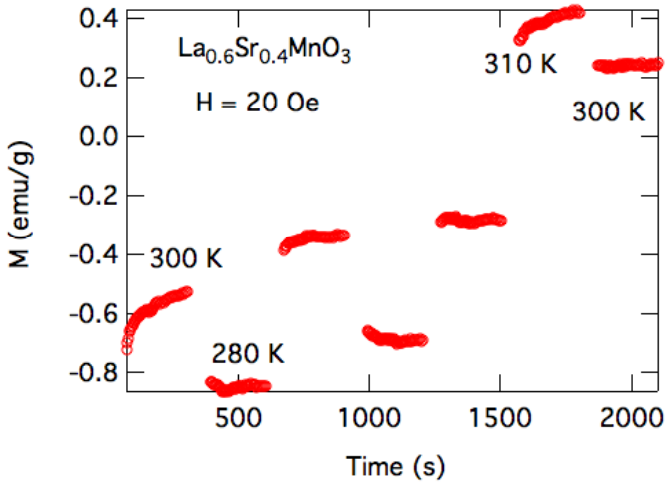

Fig. 3. ZFC magnetization relaxation of $La_{0.6}Sr_{0.4}MnO_3$ nanoparticles under the field of 20 Oe and temperature changes from 300 K to 280 K.

All above results clearly confirmed the memory effect in weak-interacting $La_{0.6}Sr_{0.4}MnO_3$ nanoparticles. The interesting memory effects in spin-glass materials have been discussed with a droplet [15] or hierarchical model [16,17]. In a hierarchical model, a multi-valley structure of energy landscape was formed on the free-energy surface at given temperature. The free energy valleys split into new subvalleys with decreasing temperature or magnetic field and merge with their increase. When the system is quenched from T to T - ΔT or H - ΔH, each free energy valley splits and develops a set of subvalleys because energy ΔE is a function of T and H or ΔT and ΔH. If ΔE are large, the energy barriers separating the main valleys become too high to be overcome during finite waiting time. The relaxation then occurs only within the subvalleys and the magnetization keeps almost constant at H - ΔH as seen in left-side figure 2. When temperature is higher, the same ΔH give smaller change in energy then the magnetization still relaxes at H - ΔH as seen in right-side figure 2. When temperature and magnetic field return to T or H, energy barriers merge back to the previous free energy landscape. The memory effect in figure 3 can be similarly explained using this model when

magnetic field was kept constant and the temperature was changed. Unlike the droplet model, which should be symmetrical with respect to heating and cooling [15], the hierarchical model predicts no memory effect would appear after a temporary heating in ZFC measurement as can be seen in figure 3 when the system was heated up to 310 K and then changed back to 300 K. Based on above discussion, the observed memory effect in this study can be understood by the hierarchical landscape which strongly related to the energy distribution. These results are also supported by the study in strongly interacting system [12,13] where magnetic memory effects were explained using the magnetic reversal due to the changing of energy barrier distribution function when magnetic field was changed. Those above arguments can be applied to explain the memory effect in all systems including non-interacting, weakly and strongly interacting.

4. Conclusion

In conclusion, magnetic memory effect of $La_{0.6}Sr_{0.4}MnO_3$ nanoparticles prepared by sol-gel method was observed due to various changing of magnetic field as well as temperature. This memory effect can be explained in the model of changing energy distribution in nanoparticles system, which was supported by the study in strongly interacting systems. This model provides some information about the energy barrier distribution function, which is related to the particles size distribution.

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