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Magnetic and magnetoresistance properties of La_{0.825}Sr_{0.175}MnO₃ powder compact

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Abstract

The magnetic and magnetoresistance properties of $La_{0.825}Sr_{0.175}MnO_3$ powder compact prepared by the mechanical alloying method and high temperature– high pressure treatment have been investigated. Analysis by means of x-ray diffraction and scanning electron microscopy show that the $La_{0.825}Sr_{0.175}MnO_3$ powder has an average grain size of 100 nm and has been partly non-crystallized by mechanical alloying. Compared with the crystal and general nanogranular samples of the same material, the powder compact is considerably different in both magnetic and magnetoresistance properties.

1. Introduction

Observation of a very large negative magnetoresistance ratio (MR) near room temperature for doped Mn oxides ($A_{1-x}B_xMnO_3$, where A is a rare-earth element such as La, Nd, Pr, and B is a divalent alkaline-earth element such as Ca, Sr, Ba) has created renewed interest [1–3] both as regards theoretical understanding of the fundamental physics and device application as in magnetic field sensors and hard-disk-drive heads. For device application, the difficulty is that a comparatively large MR effect in $A_{1-x}B_xMnO_3$ appears only under a high magnetic field. This limits their application. In order to understand the origin of the fundamental physics of the colossal magnetoresistance and with a view to device application, wide and careful studies have been carried out. One of them investigated the granular manganese perovskite ($A_{1-x}B_xMnO_3$) [4–6].

For this system, Ning Zhang *et al* [7] present a model for calculating the resistance of granular perovskite based on taking the interface between neighbouring grains as a potential barrier. They think that the magnetic configuration in the grain surface is more chaotic than that in the core, from considering the facts that

(1) the lattice structure in the surface is amorphous, and the magnetic configuration of such material is very structure sensitive;

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Figure 1. The x-ray powder diffraction patterns of $La_{0.825}Sr_{0.175}MnO_3$.

Figure 2. Magnetization of $La_{0.825}Sr_{0.175}MnO_3$ at 5 K as a function of magnetic field.

(2) due to a large number of dangling bonds or non-coordination atoms existing in the surfaces, the coupling among the magnetic ions in the surfaces should be much weaker than that in the cores.

One could think that ionic spins are disordered in the surfaces. The double-exchange interaction could be weak in the surface. Then the interfaces between neighbouring grains together with the intergrain distance could play the role of the potential barrier as in the multi-layer. The ionic spins in the surface can be aligned by an external field, as in crystals. Therefore, the barrier height, and also the tunnel resistance, could be reduced.

Generally, granular samples are prepared by the sol–gel method and treated at various temperatures. For this study, we synthesized the material by the standard solid-state method, then made it granular by mechanical alloying. This method should enhance the amorphous nature of surfaces of particles. Increasing of the thickness of the amorphous region would influence the magnetoresistance of samples. We also put 2 wt% silver powder among the grains of La_{0.825}Sr_{0.175}MnO₃. This has two purposes as follows:

- (1) to improve the contact between the particles;
- (2) silver can also be a potential barrier.

In order to decrease the distance between the particles, we prepared the pellets under a pressure of 4 GPa and at a temperature of 400 °C.

2. Experimental procedure

The ceramic La_{0.825}Sr_{0.175}MnO₃ was synthesized using the standard solid-state method, then made into granules by mechanical alloying. The granules with about 100 nm particle size were compacted under a pressure of 4.0 GPa and at a temperature of 400 °C. The x-ray diffraction study of the granular product was carried out using Cu K α radiation. The microstructural observation of the granular sample was performed by a scanning electron microscope. Magnetic measurements were completed with a SQUID magnetometer. The resistance and magnetoresistance were measured using a four-probe dc method.

3. Results and discussion

Figure 1 presents the x-ray diffraction spectra of granular $La_{0.825}Sr_{0.175}MnO_3$. It is evident that the diffraction peaks of the sample are considerably broadened and a small amorphous



Figure 3. The temperature dependence of the magnetization in a field of 500 Oe for La_{0.825}Sr_{0.175}MnO₃.



200

Temperature (K)

300

100

peak has appeared. These are in good agreement with the SEM observation that indicates that the average grain size is 100 nm.

7000

6000

500

400 g

300

2000

1000

Figure 2 shows the magnetic field (H) dependence of magnetization at a temperature of 5 K. It can be noted that M continues to increase slowly up to the highest measuring field. The temperature dependence of M at 500 Oe is plotted in figure 3. It shows that M increases slowly with decreasing temperature in the range of 250–50 K. At lower temperature, there is a clear drop in M, which is called spin-glassy behaviour. Figure 4 presents the temperature dependence of the resistance at different magnetic fields. It is interesting to note that the resistance increases rapidly from the temperature of 400 K, then increases more slowly with decreasing temperature in the range of 250–50 K. The latter behaviour is like those of noncrystalline metals and semiconductors. This is not surprising, considering that the resistance is influenced by the presence of a grain boundary that is amorphous. In non-crystalline metal, the electron resistance is already very large. Scattering of electrons induced by thermal phonons could be neglected. Therefore, the change of electron resistance with decreasing temperature is small. And due to the kinetic energy of electrons going through the potential barrier decreasing, the electron resistance increases slowly with temperature decrease.

Figure 5 shows the temperature dependence of the magnetoresistance. It indicates that the MR is almost independent of the temperature in the range of 250-50 K.

Generally, the electric transport behaviour near T_c in the manganites has been attributed to a strong coupling between the conduction electrons and the local magnetic moment through the so-called double-exchange mechanism. With decreasing T, the resistance drops rapidly as the neighbouring spins become aligned and the magnetization approaches its saturation value. One would, therefore, expect the MR to approach zero at the lowest temperature. This is indeed observed for the general ceramic samples of manganite perovskite. But the powder compacts exhibit different MR effects. As Ning et al indicated, the differences are due to the lattice structure in the surface being amorphous, and the magnetic configurations of the material are affected by the structure. And, due to a large number of dangling bonds or non-coordination atoms existing in the surfaces, the coupling among the magnetic ions in the surfaces should be much weaker than that in the cores. The double-exchange interaction could be weaker in the surface than that in the cores. Therefore, the ionic spins in the surface align slowly with decreasing temperature; the barrier height as well as the tunnel resistance decreases slowly. Thus, the MR is almost a constant in the range of temperatures of 250–50 K.

ZERO

400

1T



Figure 5. The temperature dependence of the magnetoresistance.

Figure 6. The resistance versus magnetic field for the powder compact of $La_{0.825}Sr_{0.175}MnO_3$.

Another difference is in the magnetic dependence of the resistance. It is shown in figure 6. It is evidence that there is an anisotropic magnetoresistance [5]. This is due to an effect of the high pressure under which the sample was made. When the powder is put under a pressure, especially when it is both under high pressure and at high temperature, the particles of the powder like to align along some direction. Therefore, the sample exhibits somewhat crystalline behaviour.

4. Conclusions

We have examined the MR and magnetization of $La_{0.825}Sr_{0.175}MnO_3$ powder compact. Colossal MR is observed in a comparatively low magnetic field. And it is almost independent of temperature over a large range of temperatures. Also its magnetic properties are different from those of crystal and general nanogranular samples of the same material made by the sol–gel method. We speculate that this is due to the amorphous nature of the surfaces of the granules.

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