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SLOW RELAXATION OF RESISTANCE IN NANOSTRUCTURED $La_{0.83}Sr_{0.17}MnO_3$ FILMS INDUCED BY PULSED MAGNETIC FIELDS

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The results of colossal magnetoresistance (MR) relaxation investigations in nanostructured and epitaxial La-Sr-Mn-O films grown by the MOCVD technique are presented. The films were studied in a pulsed magnetic field ranged from 2 to 10 T at the temperature of 80 K. The slow relaxation of resistance which takes place during milliseconds when the magnetic field is switched off is analysed using the Kohlrausch-Williams-Watts model. It was found that this relaxation is typical of spin-glass materials and is related with properties of disordered grain boundaries of nanostructured films. The MR relaxation of epitaxial films was not observed. The influence of film preparation conditions on MR relaxation was analysed in order to develop high pulsed magnetic field sensors exhibiting small dynamic "memory" effect and operating at low temperatures.

Keywords: colossal magnetoresistance, manganites, thin films, resistance relaxation, spin-glass systems, magnetic field sensors

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1. Introduction

Recently, interest in polycrystalline manganite films has increased for the fundamental understanding of the colossal magnetoresistance (CMR) phenomenon as well as due to potential device applications [1]. It was shown that polycrystalline manganite films can be successfully used for the development of CMR-B-scalar sensors which have a unique property to measure locally the magnitude of the high pulsed magnetic field. The CMR-B-scalar sensors have been used at room temperatures to measure the magnetic diffusion processes in railguns [2] and the distribution of magnetic fields during coilgun operation [4]. However, for magnetic field measurement in non-destructive coils [5] or magnetic diffusion in superconductors, sensors operating at cryogenic temperatures are required. In these cases, it is important to minimize magnetic "memory" effects which limit the operation speed of the sensors. For this purpose, the investigation of magnetoresistance (MR) relaxation processes in manganite films is of great importance.

The magnetic relaxation upon removal of the external magnetic field is widely studied in magnetic materials [5–9]. This relaxation is a complex phenomenon and various theoretical models have been proposed to explain it. There are several expressions to characterize the time dependence of magnetic relaxation, such as logarithmic, power-law-like, exponential (Debye), and stretched exponential relaxation (SER) [5–9]. In manganites having spin glass properties, slow logarithmic MR and magnetization relaxation is usually found [7–9]. Detailed studies of the behaviour of magnetic relaxation may give information about mechanisms of magnetization change in these materials and may let to distinguish between spin-glass, clustered, and ferromagnetic systems. It was shown [7] that due to close relation between the magnetic and transport properties of ferromagnetic manganites, magnetic relaxation could be measured by investigating the MR effect.

It has to be noted that in the cases of strongly interacting materials and complex systems, the relaxation can be better described by the SER ($\sim\exp[-(t/\tau)^{\beta}]$, where parameters τ and β are characteristic time and shape fraction, respectively) rather than a logarithmic-type function [5, 10]. The SER could be explained by a particular distribution of relaxation times in glasses. However, in such case it is difficult to understand the microscopic meaning of the shape fraction β. Phillips et al. [12, 13] utilized an axiomatic topological model in which all glassy relaxation is supposed to take place in an exponentially restricted configuration space, and relaxation paths are obtained by diffusion to randomly distributed relaxation sinks or defects in a space of restricted dimensionality. According to this model, for field-free relaxation in the microscopically

homogeneous samples (ideal glasses) the parameter β is supposed to have two "magic" values: 3/5 indicating one relaxation channel by short-range forces, and 3/7 when diffusion takes place in two channels: short- and long-range. It was shown that in the presence of nanocrystallites in polycrystalline $La_{0.83}Sr_{0.17}MnO_3$ films, the β values are found in the range of 0.65–0.8 [10] indicating short-range interactions in the disordered grain boundary regions.

In this paper, the magnetoresistance relaxation after removal of the magnetic field pulse is studied in nanostructured $La_{0.83}Sr_{0.17}MnO_3$ films having grain boundaries with a different level of disorder. The obtained results are discussed in the frame of the Kohlrausch-Williams-Watts model describing the spin-glass-like materials.

2. Experimental set-up and samples

The $La_{0.83}Sr_{0.17}MnO_3$ (LSMO) films were deposited using the pulsed injection metalorganic chemical vapour deposition technique onto a polycrystalline lucalox (99.9% Al_2O_3 + 0.1% MgO) substrate. Due to the polycrystalline structure of the substrate, polycrystalline films were produced. The films having the thicknesses of 400 nm were grown at different deposition conditions (changing deposition temperature T_d in the range of 650–750 °C) in order to obtain films with different average dimensions of the crystallites. The morphology of the films investigated using atomic force microscopy demonstrated nanostructured behaviour of the films: crystallites with dimensions of 10–20 nm were cumulated in larger clusters having dimensions D in the range of 110–285 nm [4]. The samples for studies of MR relaxation have a coplanar shape with two square-shaped Ag electrodes deposited by thermal evaporation at 420 °C and separated by a distance of 50 μm. The resistivity ρ vs. temperature T dependences were measured using a low dc electric field in the temperature range of 5–300 K. The ρ vs. T dependences revealed a typical of manganites transition from metal-like to insulator-like state at temperature T_m. It was found that for lower deposition temperatures, higher resistivity maximum ρ_m and smaller dimensions of clusters were obtained. The T_m, ρ_m, and D of the investigated films were found to be as follows: (1) 245 K, 0.8 Ωcm, 285 nm (F-0.8), (2) 235 K, 1.8 Ωcm, 170 nm (F-1.8), (3) 220 K, 6.5 Ωcm, 140 nm (F-6.5), and (4) 160 K, 53 Ωcm, 110 nm (F-53). It was found that a film prepared at a low deposition temperature (650 °C) exhibited resistivity more than 50 times larger if compared with a film grown at 750 °C. In order to clear up the influence of grain boundary material on the relaxation process, investigations were also performed on epitaxial films of the same composition deposited on the $LaAlO_3$ substrate at the same T_d. For these films, T_m = (315–320) K and ρ_m = (20–30) mΩcm.

The MR relaxation measurements were performed at temperatures close to the ferromagnetic state (80 K for nanostructured, 250 K for epitaxial films) using a pulsed magnetic field generator based on capacitor bank discharge through a special multi-shot magnetic field coil. In order to avoid a "tail" of the magnetic field pulse after the current is switched off, a non-metallic outer casing made from polyamide material was fabricated (for more details see [10]). Such coil was able to produce 0.9 ms duration half sine waveform magnetic field pulses with amplitudes of up to 10 T (see Fig. 1, left scale).

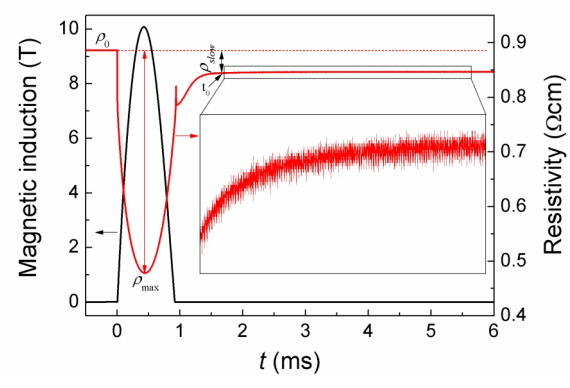

Fig. 1. Magnetic field pulse (left scale) and resistivity change (right scale) during this pulse.

3. Results and discussion

The typical resistivity dynamics for nanostructured maganite films after the application of a magnetic field pulse with an amplitude of 10 T is presented in Fig. 1 (right scale). The most part of resistivity change follows the magnetic field pulse. In this study, we focus on slow relaxation of resistivity (marked as ρ_{slow}) which takes place after the fast relaxation is finished and occurs in ms and longer time scales. The fast relaxation "tail" in nanostructured LSMO films was considered in more details in [10]. The slow relaxation was analysed using the well-known Kohlrausch-Williams-Watts model fitting experimental data to the 'stretched' exponential decay [5]:

$$\sigma(t) = \sigma_{0slow} + \sigma_{slow} \exp\left[-((t - t_0) / \tau_{slow})^{\beta}\right], \; 0 < \beta < 1, \quad (1)$$

where t_0 is the time instant at which the fast relaxation process is considered to be finished (in our case after at least 3 characteristic time constants), τ_{slow} is the time constant of the slow process, σ_{0slow} and σ_{slow} are conductivity

before pulse application and remanent conductivity amplitude, respectively. The corresponding resistivity $\rho_{slow} = 1/\sigma_{slow}$ is shown in Fig. 1.

The normalized time-varying resistivity changes in a slow relaxation time range for different samples are presented in Fig. 2. The resistivities are normalized to the $\rho_{slow} = 1/\sigma_{slow}$ at time instant t_0 for three samples. The starting point of the time-axis is chosen to be $t_0 = 0$. Due to high resistance (about 100 kΩ) of the sample F-53, the accuracy of its response signal was much lower and the filtered signal is presented separately in the inset. One can see that its relaxation takes place in much longer time scales than our measurement range. Figure 2 demonstrates that the normalized remanent resistivities are higher for films with higher maximal resistivities, indicating that spin-glass relaxation is more prominent in films with higher disorder of their grain boundaries. The dashed line marks the normalized initial resistivity before the action of the magnetic field pulse: ρ_{0slow}/ρ_{slow}. The normalized initial resistivity values for the films F-0.8, F-1.8, and F-6.5 were 1.043, 1.055, and 1.056, respectively. A similar tendency of increased remanent resistivity change with increased sample disorder was obtained by Kozlova et al. [8]. They found that slow logarithmic-type resistance relaxation in polycrystalline manganite films ($La_{0.7}Sr_{0.3}MnO_3$) is considerable at low temperatures, while in textured films of the same composition it is by one order smaller, and in epitaxial ones it is absent (at least by two orders smaller). We did not observe the magnetoresistance relaxation of investigated epitaxial films of the same thickness (400 nm) either. Sirena et al. [7] showed that large resistivity relaxation and magnetic viscosity ratios are measured when the thickness of the manganite film is decreased from 300 nm down to 10 nm and explained it by increase of structural disorder of thinner films due to lattice mismatch.

We obtained that the absolute amplitudes of remanant resistivities (ρ_{slow}) do not depend on magnetic field value in the range of 2–10 T. It may be explained by saturation of magnetic field flux pinned in various defects and dislocations at fields lower than 2 T. It has to be noted that for high magnetic field sensor applications the slow relaxation component relative to the maximal resistance change (magnetoresistance signal) is less than 10% for magnetic fields higher than 8 T and becomes insignificant in high magnetic fields [10]. For a sample having the smallest clusters and highest resistivity (F-53), this value is smaller (8% at 8 T), probably due to a much larger magnetoresistance value. The MR = 100% $[(\rho_B - \rho_0)/\rho_0]$, where ρ_B is resistivity in the magnetic field, in the magnetic field of 10 T was found to be –73% for film F-53, –50% for F-6.5, and –45% for F-1.8 and F-0.8.

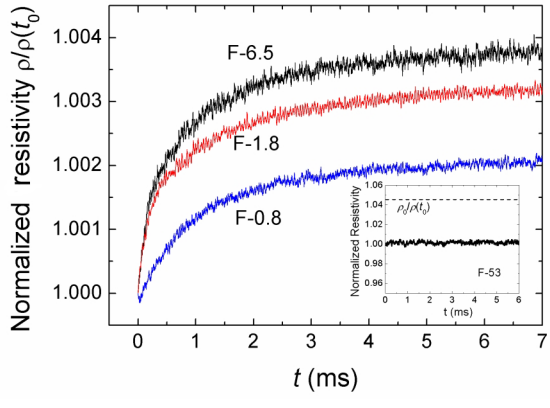

Fig. 2. Normalised remanant resistivity changes in time of 'slow' relaxation process for films with different ρ_m.

Figure 3 presents time constant τ_{slow} and exponent β dependences on the magnetic inductance of 'slow' conductivity relaxation fitted to the KWW model for four samples with different ρ_m. One can see that τ_{slow} for films F-0.8, F-1.8, and F-6.5 is in the range around 1ms, while for film F-53 it is around 10 ms. Such much longer relaxation could be explained by the fact that distribution of relaxation times in disordered material is much wider. We found that parameter τ slightly increases with the increase of the magnetic field. It could be explained by taking into account that the grain boundary region is structurally and magnetically disordered with decreased magnetization. It was shown that even up to 91 T the magnetoresistance saturation is not complete [4]. Therefore, at a higher magnetic field more magnetic centres in the grain boundary are magnetized and, therefore, involved in the relaxation process after the field is switched off. A comparison of parameter τ values obtained for different films is not correct without taking into account the values of parameter β which is shown in Fig. 3. It is obtained that for films with ρ_m around 1–2 Ωcm, β is in the range of 0.6–0.7 indicating that relaxation takes place due to short-range interactions in glassy material. It was demonstrated [12] that β values which differ from magic numbers could be used to test sample homogeneity and quality. Thus β for a film with more disordered grain boundary material (ρ_m around 6.5 Ωcm) demonstrates that at lower fields (2 T) the sample is less homogeneous like spin glass. The most disordered sample shows β close to 0.3 in the magnetic field of 10 T what indicates a very wide distribution of relaxation times and energy barriers of centres which take place in the relaxation process.

It was concluded that slow magnetoresistance relaxation in nanostructured $La_{0.83}Sr_{0.17}MnO_3$ films occurs in ms time scales. The relaxation strength (remanent amplitude) is proportional to the resistivity of the film and independent of the magnetic field in the range of 2–10 T. It was found that time dependences of

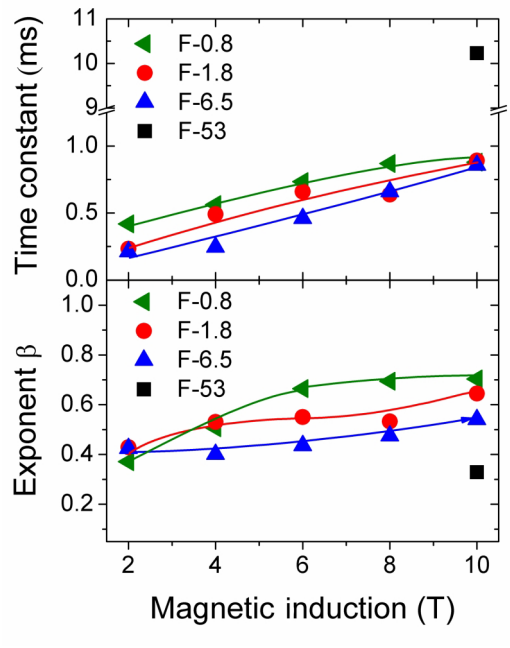

Fig. 3. Time constant τ_{slow} and exponent β dependences on the temperature of 'slow' conductivity relaxation fitted to the KWW model for films with different ρ_m. Curves are guides to the eye.

resistance relaxation could be well fitted by the stretched exponential function suggesting that the grain boundary material behaves as spin glass. For development of magnetic field sensors operating at low temperatures and measuring high pulsed magnetic fields, the films with a higher disorder of grain boundaries would be preferable.

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LĖTOJI VARŽOS RELAKSACIJA $La_{0,83}Sr_{0,17}MnO_3$ NANODARINIŲ SLUOKSNIUOSE IMPULSINIUOSE MAGNETINIUOSE LAUKUOSE

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Santrauka

Nanostruktūrizuotų manganitų $La_{0,83}Sr_{0,17}MnO_3$ sluoksnių, užaugintų PI MOCVD būdu ir pasižminčių skirtingu tarpkristalitinės medžiagos defektiškumo laipsniu, magnetovaržos relaksacija ištirta 80 K temperatūroje impulsinių magnetinių laukų ruože 2–8 T. Nustatyta, kad pasibaigus magnetinio lauko impulsui, lėtoji varžos relaksacija vyksta ms laikų skalėje ir gali būti aprašyta Kohlrausch–Williams–Watts (KWW) modeliu, įskaitančiu magnetinių momentų sąveiką netvarkiose tarpkristalitinėse srityse, būdingą stikliškosioms sistemoms. Epitaksiniuose sluoksniuose, užaugintuose tokiomis pačiomis sąlygomis, magnetovaržos relaksacija 250 K temperatūroje, artimoje feromagnetinei fazei, nebuvo stebėta.