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Improvement in the long-term stability of parameters of encapsulated magnetic field sensors based on La—Sr—Mn—O thin films

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ABSTRACT

The effectiveness of different approaches used for improvement of stability of colossal magnetoresistance-B-scalar magnetic field sensors made from polycrystalline manganite $La_{0.83}Sr_{0.17}MnO_3$ thin films were considered. We show that the ageing process of sensors can be accelerated by annealing the films in Ar atmosphere at 75–125 °C and covering them by hot melt adhesive (polyethylene). Ageing kinetics of electrical resistance and magnetoresistance (*MR*) were investigated as well as the influence of hot melt adhesive. Despite of increased resistance only small changes of the *MR* and the temperature coefficient of resistance were observed. The obtained results are explained by the oxygen release, displacement or redistribution which takes place most probably in grain boundaries of polycrystalline manganites. Based on the obtained results optimal conditions for the stabilization of encapsulated sensors parameters were found resulting in the resistance drift less than 1.5% per year.

1. Introduction

A colossal magnetoresistance (CMR) effect in manganites with general formula of $R_{1-x}A_xMnO_{3-\delta}$ (where R and A are rare earth and alkaline elements) makes them perspective materials for the magnetic field sensors. Polycrystalline manganite films are of the greatest interest since they show high sensitivity in wide ranges of magnetic fields and temperature [1]. Moreover, it was shown that the magnetoresistance (*MR*) of the polycrystalline manganite films weakly depends on the magnetic field direction. This allowed development of the CMR-B-scalar magnetic field sensors, which were successfully used for the local measurements of the dynamic magnetic fields in complex magnetic systems [2,3]. Operation of such sensors is based on the change of the film resistivity depending on the applied magnetic field magnitude. However, both the initial resistance and the sensitivity to magnetic field depend on ambient temperature. This imposes a problem in applications for magnetic field measurements as temperature compensation mechanism is required. In case of pulsed magnetic fields it can be solved by measuring initial resistance of the sensor at zero magnetic field and using calibration curves obtained in advance [3]. For this reason, it is very important to know the exact value of the initial resistance at fixed ambient temperature. It was shown [4] that this resistance of the film exhibits drift in time (ageing) phenomenon. This ageing influences the life time of the sensor and requires its frequent recalibration. The possible reasons of this drift can be the oxygen migration at the grain boundaries [5–9] of polycrystalline manganites, their relaxation, and the interaction of the free surface of thin film with the environment.

It was found that in order to minimize these effects a protecting cover have to be used [4]. However, the influence of this cover and thermal treatment of manganite film, which accompanies the processing of this cover, was not investigated. Moreover, the treatment by additional accelerated ageing has to be applied in order to stabilize parameters of the sensors.

In this paper the influence of thermal treatment and encapsulation of LSMO films on stability of the CMR-B-scalar magnetic field sensor is investigated.

2. Sensor preparation and experimental procedures

CMR-B-scalar magnetic field sensors are based on the 400 nm thick $La_{0.83}Sr_{0.17}MnO_3$ (LSMO) films grown on the lucalox (polycrystalline Al_2O_3 + Mg) substrates using a metal organic chemical vapor deposition (MOCVD) technique. The detailed description of the growth and properties of such sensors is available elsewhere

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[1]. X-ray and RHEED diffraction patterns show polycrystalline structure of the films containing a single crystalline phase. The traditional photolithography and mesa etching was used to form rectangular shaped $0.4 \times 0.25\,mm^2$ LSMO films. The substrate was cut into pieces of $0.5 \times 1\,mm^2$ and two $0.45 \times 0.45\,mm^2$ large and 0.5 μm thick Ag electrodes were deposited with Cr underlayer in 10^{-5} bar vacuum. The distance between the electrodes was 50 μm. Thus, the part of electrodes was deposited over the LSMO film (0.1 mm) and another part was over the substrate. Therefore, the active area of $50 \times 400\,\mu m^2$ was formed. The Ag electrodes were deposited throughout a metallic stencil on the sample heated to 200 °C. Thermal treatment at different temperature in Ar atmosphere was made to ensure both strong metal/ceramic interface and a good electrical contact. Annealing of the electrodes was performed at 450 °C for 60 min followed by fast (50 deg/min) cooling to room temperature. Bifilar twisted wires used for elimination of "loop effect" [10] (induced electromotive force) which appears in pulsed magnetic field were soldered to the electrodes of the sample. The cross-section and top view of a ready magnetic field sensor is shown in Fig. 1. The wires were soldered to the parts of the electrodes above the substrate free of the manganite film. Thus, tension induced by the solder is not transferred to the interface between the electrodes and the film. Finally, some samples were covered with the polyethylene hot-melt adhesive. A more thorough description of such sensors is presented in [2].

Thermal treatment of the electrodes may cause silver atoms diffuse not only to manganite layer depth, but also towards the terminal. The last phenomenon can be a source of in-time instability of the electrical parameters of the sensors. The silver distribution across the surface of the manganite film was measured by using a Scanning Electron Microscope with attached Energy Dispersive X-Ray Spectrometer (SEM/EDS) HITACHI TM3000. Line scan profiles done before and after annealing of the electrodes are shown in Fig. 2. It can be seen that the Ag concentration profile did not change after the annealing within the spatial resolution of the method ($1\,\mu m^2$). This means that no appreciable diffusion takes place at such temperatures (450 °C).

Ageing of prepared samples (or resistance drift) without application of magnetic field was measured as a relative change of resistance $(R - R_0)/R_0$, where R_0 is the initial resistance measured at (20 ± 0.02) °C before annealing and R is the resistance value measured at 20 °C after annealing the sensors for different periods of time. Resistance measurements were taken using a Fluke 8846A multimeter. Temperature of the sample was stabilized using Lauda ECO Re420 thermostat.

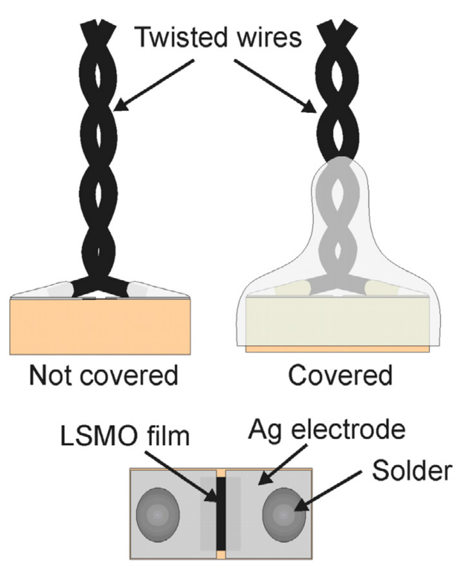

Fig. 1. Top view and cross-section of covered and not covered sensor.

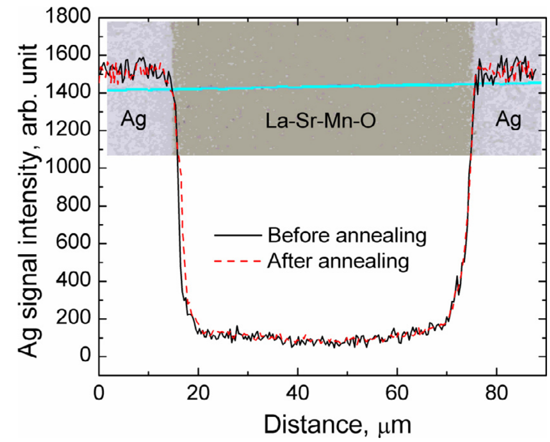

Fig. 2. EDS line scan of Ag profile across the surface of the manganites film between the two electrodes. Inset shows a SEM image of the sample surface and the scan line.

All resistance measurements were performed using two contact methods (see Fig. 1). Our previous results [10] showed that the contact resistance in this design of the samples is ~1000 times lower than of the active area of the manganite film and thus cannot have significant impact on the resistance drift.

Magnetoresistance (*MR*) was measured at temperatures of 0–40 °C and magnetic fields of 0–2.3 T created by a DC electromagnet. The *MR* is defined as $MR = 100 \times [R(B) - R(B=0)]/R(B=0)$, where $R(B)$ is the sample resistance at magnetic induction B and $R(B=0)$ – sample resistance at "zero" magnetic induction.

3. Results

3.1. Annealing influence on the resistance

It was observed, that resistance of the uncovered manganites samples prepared according to the earlier described technology increases during the long-term operation. The rapid (up to 5% during 14 days) change was observed during the first two weeks followed by the slower though significant drift of up to 40% of the initial value during two years. All sensors were kept at room temperature.

In order to study the possibilities of acceleration of the ageing process, the samples were treated at different temperatures in the Ar atmosphere at three different temperatures and time periods. Each time the samples were cooled down to room temperature to measure the resistance. Fig. 3 presents the relative resistance changes at each step. It can be seen that in all cases fast resistance increase at the beginning of the treatment followed by saturation. Saturation occurs faster at higher temperatures; however, in each of the three cases different final values were reached.

3.2. Annealing influence on TCR and magnetoresistance of the samples

As mentioned above, the principle of operation of the CMR-B-scalar sensors is based on measurements of the change of resistance caused by magnetic field. Sensitivity of the sensors depends on ambient temperature, therefore, after manufacturing they are calibrated at different temperatures. Calibration data obtained for each temperature are then used for conversion of the measured signal (voltage) into values of magnetic field [2]. Thus the long term stability of the sensor's resistance is very important to ensure accurate measurements without the need of frequent recalibration.