VILNIUS GEDIMINAS TECHNICAL UNIVERSITY

Sonata TOLVAIŠIENĖ

TRANSPORT OF CHARGE CARRIERS IN ULTRATHIN FILMS OF MANGANESE OXIDES

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Condensed Matter: electronic structure;
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VILNIAUS GEDIMINO TECHNIKOS UNIVERSITETAS

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General Characteristic of the Dissertation

Topicality and problem of the work

Lantanium manganites are materials having the general formula $La_{1-x}M_xMnO_3$ (here M = Ca^{2+}, Ba^{2+}, Sr^{2+}). They exhibit a phase transition from paramagnetic to ferromagnetic state and colossal negative magnetoresistance phenomenon. Since 1993, manganites were studied intensively in order to develop magnetic field sensors and high-density magnetic storage systems. During the past several years, investigation of the physical properties of the manganites increased due to their possible application in power electronics, electrical engineering and spintronic devices. The effects induced by strong electrical currents and high pulsed magnetic fields were studied. It was obtained that high electric currents can cause irreversible changes in the electrical conductivity of the manganites, non-linear voltage-current characteristic and strengthens the colossal magnetoresistance phenomenon. Also, it was shown that thin manganite films can be used to design high-speed protectors against electromagnetic pulses and B-scalar magnetic field sensors operating up to 50 T and measuring the magnitude of the magnetic field inductance.

The aim of the work

The objective of this effort was to investigate the electrical conductivity of ultrathin (4–140 nm thickness) $La_{0.83}Sr_{0.17}MnO_3$ films deposited by MOCVD technique on $NdGaO_3$ substrate in magnetic and strong electric fields.

Tasks of the work

1. To investigate how the anisotropy of magnetoresistance in epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films at low (20–600 mT) magnetic field depends on thickness of these films.
2. To perform analysis of the magnetoresistance anisotropy in epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films and to suggest models explaining features of this phenomenon.
3. To create an experimental setup for high-power ns duration electrical pulse effects studies on electrical conductivity of $La_{0.83}Sr_{0.17}MnO_3$ films.
4. To perform investigations of the conductivity of epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films when these films were affected by strong pulsed electric field.
5. To create a setup that enables one to study the electrical conductivity of films simultaneously affected by ns duration electrical pulses and high magnetic field pulses.

6. To investigate possibility of modulating the amplitude of high frequency electrical signals by means of triggering epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films by high magnetic field pulses.

Scientific novelty

Novelties of this effort include the following results in the area of solid state electronics:

1. It was obtained that the value and sign of the magnetoresistance anisotropy of $La_{0.83}Sr_{0.17}MnO_3$ films deposited on $NdGaO_3$ substrates with (100) orientation depends on the thickness of the film. This property appears due to anisotropic magnetoresistance and magnetocrystalline anisotropy and can be used for the design of magnetic field direction sensors.
2. Reversible thermoelectrical instability in epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films appearing as the result of strong electric field action on these films was discovered and investigated. An explanation of damaging process of the films due to action of nanosecond duration strong electric field pulses was suggested.
3. A new way for nanosecond duration electrical pulse amplitude modulation using magnetoresistance phenomenon in epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films was suggested and experimentally approved.

Practical value

It was obtained that nm thick $La_{0.83}Sr_{0.17}MnO_3$ films grown on $NdGaO_3$ substrate with (100) orientation exhibit structural anisotropy in the substrate plane. When electric current was directed along the (010) axis, the anisotropy of magnetoresistance in the substrate plane did not exceed 2%, while in the direction perpendicular to the substrate plane it was about 30%. This could be used for the design of low (up to 0.5 T) magnetic field direction sensors. Experimentally it was shown how epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films can be used for modulating the amplitude of high frequency electrical signals by an external magnetic field and determined the maximal value of the pulse amplitude, which can be modulated by this way. The fast thermoelectrical instability can be used for formation of high-power pulse waveforms.

Approval of the results

The main results of this dissertation were published in 9 scientific papers, 2 of them in journals included in Thomson ISI Web of Science and 1 in Thomson ISI Proceedings and presented at several international and national conferences including 5th Int. Conf., Smolenice, Slovakia, 2005; ERK-2006, Portorož, Slovenia, 2006; UFPS-13, Vilnius, Lithuania, 2007; „Tenth Annual Directed Energy Symposium" 2007, Huntsville, USA; The 8-th International

Conference – School „Advanced materials and technologies”, Palanga, Lithuania, 2006, and four conferences of Lithuanian Young Scientists: Vilnius, Lithuania 2005, 2006, 2007, 2008.

Defended propositions

1. The value of the magnetoresistance and the anisotropy of ultra-thin $La_{0.83}Sr_{0.17}MnO_3$ films deposited on (100) orientation $NdGaO_3$ substrate in the plane of this substrate, as well as this value dependence on temperature and film thickness at low magnetic fields, is determined by two-phase structure of the films.
2. The peculiarities of magnetoresistance and its anisotropy in epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films can be well explained by using a model based on the mean field approach and by assuming that the ratio between volumes of structural phases depends on thickness of the film.
3. Strong (> 30 kV/cm) pulsed electric fields induce in $La_{0.83}Sr_{0.17}MnO_3$ films fast reversible thermoelectrical instability, which can be well described in terms of the homogeneous adiabatic heating model. When the electric field strength exceeds 50-70 kV/cm, during a few nanoseconds switching from the high resistance state to the low resistance state having electronic nature appears. This switching is accompanied by formation of a current channel and damage of the electrodes material.
4. Epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films can be successfully used for development of magnetically triggered subnanosecond duration electrical pulse amplitude modulators and magnetic field direction sensors.

Structure of the dissertation

The dissertation (in Lithuanian) consists of abstract, introduction, 6 chapters, and main results and conclusions, references, list of publications.

The content of the dissertation

The introduction contains relevance of the dissertation, main objectives, tasks of the work, novelty and propositions to be defended.

Chapter 1 presents a review of the common physical properties of manganites at low magnetic field and strong electrical current effects. The description of these common properties contains crystallographic structure, electrical conductivity, metal-insulator phase transition, paramagnetic and ferromagnetic phase separation. The main behaviour (structure, magnetic and electric properties) of thin La-Sr-MnO films are also presented in this chapter. Magnetoresistance phenomenon and its anisotropy at low (up to $\approx$ 0.5 T) magnetic fields were analyzed. The last section of this chapter is intended for high current effects. It is demonstrated that the application of dc currents to manganites has limitation due to large heating effects and short (ns duration) pulses have to be used for these studies.

Chapter 2 consist of two parts. The first part contains a description of thin $La_{0.83}Sr_{0.17}MnO_3$ (LSMO) films prepared by a pulsed injection MOCVD method, properties of substrates, Ag electrodes deposition and two-terminal sample manufacture technologies. The $La_{0.83}Sr_{0.17}MnO_3$ films were prepared on $LaAlO_3$ (LAO) and $NdGaO_3$ (NGO) substrates at 825 °C from a liquid source solution having the composition $La_{0.78}Sr_{0.22}Mn_{0.733}$. The thickness of the films was changed from 4 nm to 400 nm. Samples had a co-planar shape with the electrodes spaced relative each to the other at *d* = 50 µm distance. The total length of the sample was 1 mm, the width 0.5 mm. Thin Ag film electrodes were deposited by thermal evaporation at 200 °C, and then annealed in an argon gas atmosphere at 400 °C.

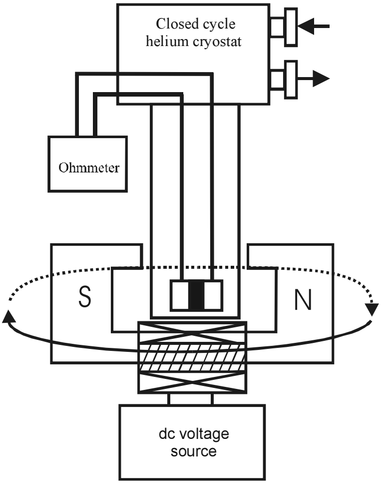


Fig. 1. Schematic diagram of experimental setup for electrical conductivity measurement at low electric and magnetic fields

The second part presents the experimental setups used to investigate the electric and magnetic properties of LSMO films. Fig. 1 shows the schematic diagram of the experimental setup used for investigating the electric and magnetic properties of thin LSMO films at low dc current and in permanent magnetic field. A closed cycle helium cryostat was used for measurement in the temperature range from 4.2 K to 300 K. The setup enables one to perform investigations at different orientations of permanent magnetic fields that ranged from 0.2 to 0.8 T.

For electric field induced resistance change measurements, the samples were connected in series to a 50 Ω impedance 18 GHz frequency transmission line and pulsed by 10–18 ns duration and 0.5 ns rise time, rectangular shaped single electrical pulses with amplitudes up to 1 kV. The schematic diagram of the experimental set-up is presented in Fig. 2. The surface morphology of damaged films was studied by optical and scanning electron microscopes.

For short electrical pulse modulation measurements, the thin film coplanar two-terminal device, was connected in parallel to a 50 Ω impedance, 12 GHz bandwidth transmission line and mounted inside the bore of a pulsed magnetic field coil. A single 7.5 V amplitude electrical pulse, having a pulse length of 10 ns and a rise time of 250 ps was transmitted through the line and was synchronized with a half-sinus shape magnetic pulse having pulse length of 0.6 ms at the moment in time that it reached its maximal value (13 T) (see Fig. 3). The magnetic field pulses were generated by discharging a capacitor bank (4 kV) through a coil having a length of 50 mm and an inside bore diameter of 12 mm. The electric pulse wave-form and amplitude were measured by using a 0–5 GHz bandwidth real time oscilloscope. Measurements were performed over temperatures ranging from 200 K to 300 K.

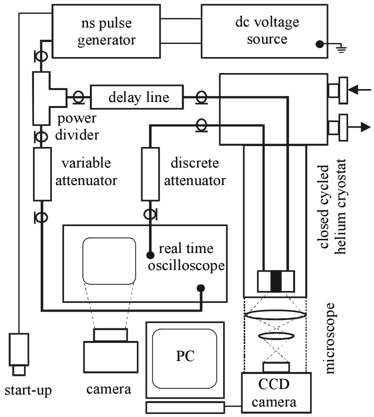


Fig. 2. Schematic diagram of experimental set-up used for manganite films thermoelectrical instabilities and damaging dynamics measurements

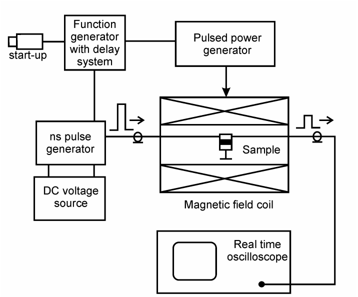


Fig. 3. Experimental setup used for modulation of short electrical pulse measurements

Chapter 3 is intended for studies of electrical conductivity of ultrathin epitaxial LSMO films at low electric and magnetic fields. The influence of Mn amount in chemical composition in the film on electric conductivity was demonstrated and that can be explained by using a mean-field approach, assuming that the decrease of Mn amount changes the angle between Mn-O-Mn bonds. The calculations using this model are in good agreement with experimental results, showing the strong influence of Mn deficit and relative small influence of its excess on the T_m of the films.

This chapter also contains the results of magnetoresistance (MR) and its anisotropy (MRA) studies in thin epitaxial $La_{0.7}Sr_{0.3}MnO_3$ films, which were grown on $NdGaO_3$ (001) substrate. MR is defined by the formula:

$$MR = \frac{R(B) - R(0)}{R(0)} \cdot 100\%; \qquad (1)$$

where $R(B)$ and $R(0)$ are the resistances of the film for magnetic fields of B and $B = 0$, respectively.

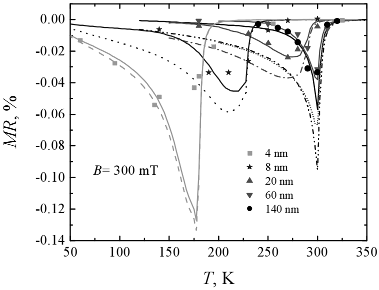


Fig. 4. *MR* as a function of temperature for different thicknesses of the LSMO film. Experiments – symbols, calculations – lines

It was obtained that the resistivity and MR strongly depends on the thickness of the films, when this thickness varied from 4 nm to 140 nm. Figure 4 shows the results of experi–mental investigations (symbols) of MR as a function of temperature for different thicknesses of the film. The dashed and solid lines represent results of calculations performed using a mean-field approach and assuming that the film is a mixture of two phases having different lattice parameters. Dashed lines show calculations made when only the magnetically sensitive part of the resistivity was taken into consideration; solid lines are obtained when both sensitive and non-sensitive parts are accounted for. For calculations, the film was simulated as a multi-plane system in which each plane had different ratio between concentrations of the phases. The first phase which is closer to the substrate is a result of large strain, while the second phase, which dominates closer to the free surface of the film, is less effected by strain and therefore its magnetic properties are closer to those of the bulk system. The concentration x_{1i} of the first phase in each plane (i = 1 ... N) has to decrease while the concentration of the second phase $x_{2i} = 1 – x_{1i}$ has to increase with increase of the number of planes or film thickness $d = Na$, where a is lattice constant in the direction perpendicular to film plane. This dependence was simulated by the empirical formula $x_{2i} = 1/\{1+\exp[–k(d-d_0)]\}$, where d_0 is the thickness at which the concentrations of phases are equal ($x_{1i}(d_0) = x_{2i}(d_0) = 0.5$) and k is the parameter, characterizing the phase separation conditions.

Investigation of the resistivity these films as a function of the magnitude of the magnetic field directed at different angles between magnetic field, electrical current (*J*) direction and the plane of the substrate demonstrated the following features of the *MR* anisotropy (MRA). 1) All films exhibited MRA when the direction of the magnetic field was changed from perpendicular to the plane of the substrate. This was attributed to a demagnetization phenomenon of the film (shape anisotropy). 2) For B < 0.4 T, changes in the angle (*Θ*) between the current and the magnetic field in the plane of the film showed that the typical resistance *R vs. Θ* dependence for films, with thickness $d > 60$ nm can be well described by the following formula: $R(\Theta)/R(0) = 1+\delta\sin 2\Theta$, where $R(0)$ corresponds to the resistance of the film at $B//J$ and when the ratio $R(\Theta)/R(0)$ was larger than one. Such dependence is typical for anisotropic magnetoresistance (AMR) in manganites. 3) For films with $d \ll 20$ nm, the same behaviour of *R vs. Θ* dependence was observed, however in this case the ratio $R(\Theta)/R(0)$ was less than one if the current was directed parallel to the [100] direction of the substrate and it was larger than one if the current was parallel to the [010] direction (see Fig. 5). In this case, the *R vs. Θ* dependence did not satisfy the $(1+\delta\sin 2\Theta)$ dependence at $B < 0.2$ T. It was concluded that it has to be associated with the existence of magnetocrystalline anisotropy (MCA) in these films. At medium thicknesses (from 8 nm to 60 nm) and when the current was directed in the [100] direction, the MRA was a combination of both the AMR and MCA effects.

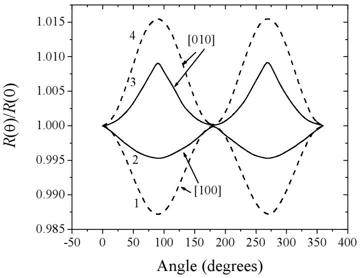


Fig. 5. $R(\Theta)/R(0)$ vs. Θ dependence for 4 nm thickness film at T= 140 K and B = 100 mT (solid), and B = 230 mT (dashed). Curent direction [010] (curves 3, 4) and [100] (curves 1, 2)

Figure 6 shows the temperature dependence of MRA for $La_{0.83}Sr_{0.17}MnO_3$ films having different thicknesses. These results were explained assuming that changes in thickness caused the change in ratio between structural phases of the film and that MRA at low magnetic fields is directly proportional to the MR. Using an empirical formula that shows the relationship between thickness and phase content, which was used for MR calculations, it was obtained that:

$$MRA = \left[\frac{A_{c0} + A_{s0}}{1 + e^{-k(d-d_0)}} - A_{s0} \right] \times MR(d,T) \,. \qquad (2)$$

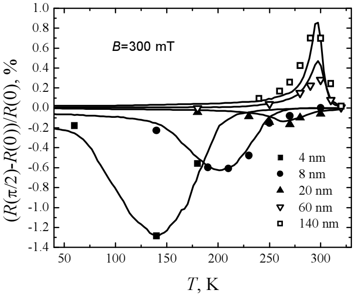


Fig. 6. The temperature dependence of MRA for $La_{0.83}Sr_{0.17}MnO_3$ films with different thicknesses

The coefficient A_{c0} is related to the MCA and can be obtained from MRA measurements of films with $d > 60$ nm, while A_{s0} is related to AMR and can be obtained from 4 nm thickness films. The solid lines in Fig. 6 represent calculation using formula (1). It was noted that at $d \approx 30$ nm the MRA in the film plane direction is very low, while perpendicular to this plane it is of order of 30%. This can be used for the design of magnetic field direction sensors.

Chapter 4 is intended for the investigations of effects of high power nanosecond electrical pulses on the electrical conductivity of epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films.

Figure 7 shows the resistivity ρ (left scale) of the epitaxial film as a function of temperature *T*, which was measured using low dc currents (full squares). The grey curve (right scale) presents the temperature coefficient of the resistance, TCR = (*d*R/*d*T)/R·100%, versus temperature. The open circles represent the resistivity measured using nanosecond pulses, where the temperature was calculated by assuming that the film was uniformly and adiabatically heated. The amount of heat generated, $Q = \int_0^\tau I(t) \times V(t) dt$ was calculated by using the current *I* and the voltage *V* measured at successive instances in time τ. The temperature *T* of the film at time τ, due to electric pulse action, was calculated by using: $T = T_0 + Q/c \cdot m$, where *c* is the specific heat capacity, *m* is the mass of the film in the inter-electrode space, and T_0 is the initial temperature of the film. The factor $c \cdot m$ was kept constant over the entire temperature range and calculated at $T = T_m$, assuming that *Q* is the energy necessary to heat the film up to T_m. The ρ *vs.* *T* dependence (see open circles in Fig. 7) obtained by this method at $T_0 = 70$ K is very close to the same dependence measured with low dc fields (full squares). This observation demonstrates that strong electric fields (up to 120 kV/cm) only cause heating of the epitaxial film and pure electronic processes play no role in determining the film electrical conductivity.

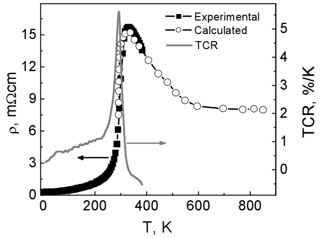


Fig. 7. Resistivity ρ and temperature coefficient of resistance TCR of epitaxial $La_{0.83}Sr_{0.17}MnO_3$ film as a function of temperature

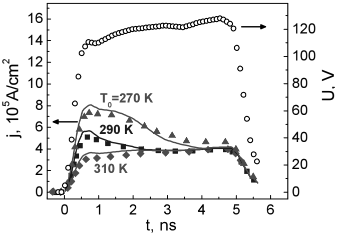


Fig. 8. Dynamics of current density (full squares, left scale) through epitaxial film at three different initial temperatures T_0. Open circles mark incident electrical pulse (right scale)

Fig. 8 shows the experimentally measured current density $j = I/S$, where I is the total current, S is the cross-sectional area of the film, through epitaxial film at three different initial temperatures T_0 (full squares). The shape of the incident electrical pulse is marked by open circles (right scale). When $T_0 << T_m$, the current density dynamics consists of two phases: first, the current decreases within a few ns after the pulse achieves its rise time and second, it is relatively stable or even slowly increases at the end of the pulse. When T_0 is close to T_m (see curve at $T_0 = 310$ K) or higher, only current density increases in time are observed. The solid lines demonstrate the results of computer simulations of the switching dynamics using ρ *vs*. *T* data from Fig. 7 (full squares) and assuming that heating is adiabatic and uniform. The change in current density during the pulse is faster when the amplitude of the incident pulse is higher and when T_0 is closer to the region at which there is an abrupt growth in resistance with temperature (highest values of TCR).

Chapter 5 is intended for investigation of fast electrical switching from high resistance off-state to low resistance on-state in thin epitaxial $La_{0.7}Sr_{0.3}MnO_3$ films appearing as a result of nanosecond duration strong electric field pulse action.

Typical current-voltage characteristics of epitaxial films measured at different time instants (τ_i) at temperature $T = 295$ K are shown in Fig. 9a. They are nonlinear in the positive differential resistance region up to the threshold voltage (V_{th}) at which switching from high resistance (off-state) to low resistance (on-state) occurred. The fast electrical switching was obtained at current densities higher than 10^6 A/cm^2. The shape of *I-V* curves at $V < V_{th}$ corresponding to different τ_i is a result of the Joule heating and resistivity *vs.* temperature dependence of epitaxial film exhibiting phase transition from ferromagnetic to paramagnetic state (see Fig. 7). At τ_i = 2 ns, the heating is relatively small and *I-V* curve up to 175 V demonstrates an increase in the film resistance, which means that the temperature of the film is less than resistivity maximum temperature T_m. For higher voltages the temperature growth of the film is accompanied with resistance decrease as in this case $T > T_m$. At times longer than τ_i = 5 ns, heating even at 50 V is sufficient to transform the film from the ferromagnetic to the paramagnetic state. For this reason at $V > 50$ V the temperature of the film after 5 ns is always higher than T_m and the nonlinear *I-V* curve shows a decrease in the resistance. The same reasons determine the behavior of *I-V* curve measured at τ_i = 10 ns.

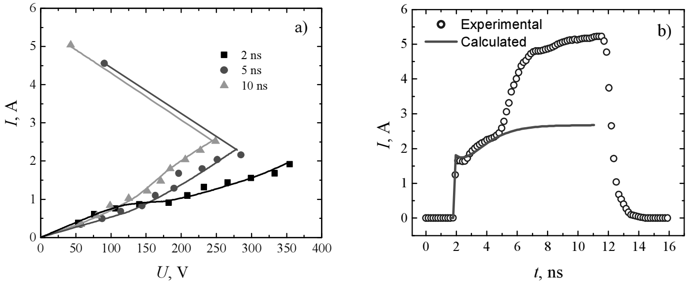
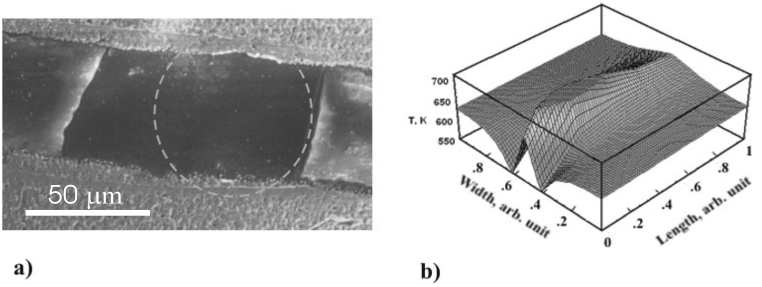


Fig. 9. (a) Current-voltage characteristics of $La_{0.83}Sr_{0.17}MnO_3$ film measured at different time instants. T = 295 K. (b) Switching dynamics at applied pulsed voltage across the sample with amplitude V = 450 V

The investigation of wave forms of the current through the sample has demonstrated that switching process exhibits three phases (see Fig. 9b). The first phase (delay time) appearing immediately after the pulse rise time is a period during which relatively slow (few ns) increase in current takes place, the second phase (switching) is associated with abrupt (less than ns) jump, about ten times, in current and the last third phase (few ns) is the on-state formation period which is necessary to create time-independent low resistive state. The on-state could be maintained only if the voltage across the film is higher than a certain value V_{on}. The increase of the overvoltage (V-V_{th}) decreases both delay and switching time.

The investigations show that after on-state formation, the current in the interelectrode area is distributed non-uniformly. This can be seen form SEM pictures, demonstrating damage regions at the current electrodes (see Fig. 10a) The damage was concentrated in the narrow parts of the electrodes and manifested itself as melting of electrodes material in the interconnection region between the Ag electrode and the manganite film. The area of the damage region was always larger at the anode and narrowed at the cathode. Typical ratios between such “two-dimensional” channel width at the anode and cathode varied from 4 to 8. Moreover, it has to be noted that high current in on-state damaged only electrode material, meanwhile the manganite film was not destroyed.

The obtained phenomenon was explained by the following model. During the first phase (delay time) high current induces fast heating of the manganite film and charge carriers are injected from the cathode at a local region associated with imperfections in the Ag thin film electrode. These imperfections cover about 0.1 of the total interelectrode distance (see Fig. 10a). This led to the appearance of highly localized low resistance regions at the cathode. Further heating causes current crowding and increased power dissipation at certain places in the interelectrode area. As a result, a high-current density channel is formed between electrodes.



a)

b)

Fig. 10. SEM picture demonstrating damage regions of $La_{0.83}Sr_{0.17}MnO_3$ film at the current electrodes (a); Simulated temperature distribution in the film along its length after 5 ns (b)

The manganite film was simulated as a two–dimensional resistor ladder network with four series-parallel resistors of the same value in each element of the ladder. Using typical manganite films resistance vs. temperature dependence it was obtained that peculiarities of current dynamics during the delay time period can be well understood on the basis of an electro-thermal model (see Fig. 9b, solid curve). The imperfection was introduced as a resistance located at the cathode and having three times smaller value compared to the rest of the resistances in the network. This model also enabled us to predict the asymmetrical shape of the current channel and showed that such a shape is a result of non-homogeneous heating during the delay time (Fig. 9b). However, the thermal model was unable to explain the second phase, i.e. fast switching period. It was suggested that this phase has to be associated with electronic processes such as avalanche of the charge carriers in solid state material, which was necessary to reach critical power in the interelectrode space. Calculations of the distribution of power dissipation in this area demonstrated that at the end of the delay time, the highest power is concentrated close to the anode. Asymmetrical damage of the current electrodes could be an additional argument that electronic switching starts at the anode and then propagates to the cathode.

It was demonstrated high current is able to induce irreversible damage of the sample mainly in the region of the interconnection between the film and the electrodes (see Fig. 5). The experiments and calculations showed that at high overvoltages (> 700 V which corresponds to 140 kV/cm in our case) the temperature increase ΔT of the film is always high enough (~ 500 K after 5 ns) to induce irreversible damage. Moreover, the damage of the electrodes demonstrates that the temperature in the on-state reaches the Ag melting temperature (about 10^3 K).

It was concluded that the fast switching appearing in thin manganite films as a result of nanosecond duration strong electric field pulse action is of electronic nature. Meanwhile, the thermal processes occurring in high–current regime are responsible for the origin of the delay time and the asymmetrical shape of the current channel in the on–state.

Chapter 6 is intended to show RF electrical pulse modulation using magnetically triggered thin manganite films**.**

Modulation of the amplitude of radio frequency (RF) electrical pulses was realized by applying external magnetic fields to thin epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films grown by the MOCVD method. Detailed investigations of the dc electrical resistance and magnetoresistance as a function of temperature for films having thicknesses 4–140 nm showed that the largest *MR* value occurring at room temperature is obtained for films having thickness of 20 nm. For this reason, the samples made from 20 nm thick films were chosen to perform RF pulse modulation. As it was mentioned in Chapter 2, the sample was prepared as a coplanar two-terminal device and connected in parallel to a 50 Ω impedance, 12 GHz bandwidth transmission line, and mounted inside the bore of a pulsed magnetic field coil. As it is shown in Fig. 11, a single 7.5 V amplitude electrical pulse, having a pulse length of 10 ns and a rise time of 250 ps was synchronized with a half–sinus shape magnetic pulse having a pulse length of 0.6 ms at the moment in time that it reached its maximal value (13 T).

Figure 12 shows the waveform of the beginning of an electrical pulse at various temperatures modulated by an external half-sinus shaped magnetic field pulse with amplitude 13 T. As a result of modulation, the amplitude of the electrical pulse at room temperature was decreased about 30%. This decrease was not accompanied by any changes in the shape or rise time of the modulated pulse. It was demonstrated that this method can be used for RF signal attenuation by external magnetic fields and that no galvanic coupling is required between the RF circuit and the circuit which generates the external action.

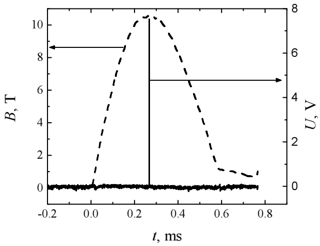


Fig. 11. Synchronization of electric and magnetic pulses

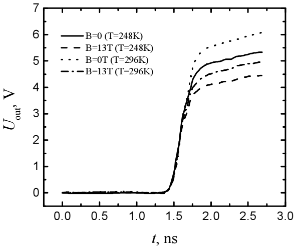


Fig. 12. Waveforms of modulated pulse

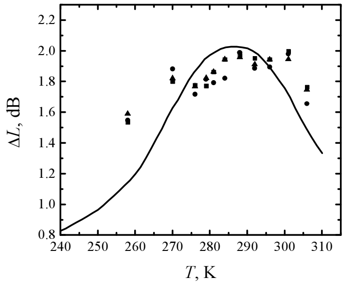


Fig. 13. Attenuation vs. temperature dependence. Symbols show pulse experiment, line shows calculations

It was shown that the attenuation (L_B), which appeared in the Z = 50 Ω impedance transmission lines if it is shunted by thin film with resistance at zero magnetic field $R_0(T)$, can be calculated by using the following formula:

$$L_B = 20\log\left\{1 + \frac{50 \cdot Z}{R_0(T)\left[100 - MR(B,T)\right]}\right\}; \tag{3}$$

where the magnetoresistance $MR(B,T)$ is in percent. The attenuation $\Delta L(T) = L_B - L_0$ as a function of temperature T at 13 T magnetic field achieved from kinetics measurements (square dots) is presented in Fig. 13. It shows that the highest $\Delta L \approx 2$ dB achieved was at temperatures around room temperature. The solid line shows the calculated results obtained by using Eqs. (3) and the experimental data of dc resistance and *MR* measurements.

It was concluded that thin manganite films can be used to modulate RF electrical signals having a bandwidth of several GHz by applying a pulsed magnetic field with an inductance of about 10 T. By this way, it is possible to attenuate RF signals up to several dB. The attenuation can be increased by choosing the optimal values of the magnetoresistance and of the dc resistance of the films.

General Conclusions

1. The magnetoresistance anisotropy (MRA) of nanometre thick thin $La_{0.83}Sr_{0.17}MnO_3$ films was investigated at low (20–600 mT) magnetic fields. It was obtained that the value and sign of MRA in films grown on (100) orientation $NdGaO_3$ substrate depends of thickness of these films. It was demonstrated that this property of the film is due to anisotropic magnetoresistance (AMR) and magnetocrystalline anisotropy (MCA) phenomenon. The relative impact of which on total MRA changes with the thickness of the film.
2. Using a mean-field approach and taking into consideration that the structure of the film changes with the thickness, the analysis of the MRA phenomenon in ultra-thin $La_{0.83}Sr_{0.17}MnO_3$ films was performed. On the basis of this analysis, models explaining the main features of this phenomenon were suggested.
3. It was shown that 20 nm thickness epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films grown by MOCVD method on (100) orientation $NdGaO_3$ substrates, at low magnetic fields (up to 0.5 T) exhibit very small values of MRA in the film plane (about 2%). While in a direction perpendicular to the film plane, the MRA is about 30%. This can be used for magnetic field direction sensors design.
4. The kinetics of electrical conductivity changes in epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films induced by strong pulsed electric fields was investigated. Reversible thermo-electrical switching was discovered and investigated in these films. It was suggested to use this phenomenon for ns duration high-power electrical pulse waveform shaping. The reasons and behaviour of irreversible damaging of the films induced by short high power current pulses were determined.

5. The possibility of using epitaxial $La_{0.83}Sr_{0.17}MnO_3$ films to modulate the amplitude of high frequency electrical signals by external magnetic fields was demonstrated and investigated.

List of Published Works on the Topic of the Dissertation

In the scientific periodical publications included in Thomson ISI Web of Science

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7. BALEVIČIUS, S.; ŽURAUSKIENĖ, N.; STANKEVIČ, V.; TOLVAIŠIENĖ, S.; KERŠULIS, S. 2007. Spartus grįžtamasis termoelektrinis perjungimas plonuosiuose manganitų sluoksniuose, *10-osios*

jaunųjų mokslininkų konferencijos leidinyje, „Mokslas –Lietuvos ateitis“, 133–138.

8. BALEVIČIUS, S.; ŽURAUSKIENĖ, N.; STANKEVIČ, V.; ČESNYS , A.; TOLVAIŠIENĖ, S.; ABRUTIS, A.; PLAUŠINAITIENĖ, V.; CIMMPERMAN, P. 2006. Plonųjų $La_{0,83}Sr_{0,17}Mn_xO_3$ sluoksnių, užaugintų metalų organinių junginių cheminio nusodinimo iš garų fazės būdu ant Al_2O_3 padėklo, elektrinių ir magnetinių savybių tyrimas, *9-osios jaunųjų mokslininkų konferencijos leidinyje, „Mokslas –Lietuvos ateitis“*, 127–130.
9. BALEVIČIUS, S.; ŽURAUSKIENĖ, N.; STANKEVIČ, V.; TOLVAIŠIENĖ, S.; PLAUŠINAITIENĖ, V.; CIMMPERMAN, P. 2005. Mn kiekio įtaka La-Sr-MnO_3 plonų sluoksnių elektriniam laidumui, *8–osios Lietuvos jaunųjų mokslininkų konferencijos „Lietuva be mokslo – Lietuva be ateities“ pranešimų rinkinys*, 151–154.

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Sonata Tolvaišienė was born in Virbalis on April 21, 1976. Bachelor degree obtained in Physics and astronomy in Faculty of Physics and Technolgy at Vilnius Pedagogical University, in 2000. In 2002 – Master of Science in Faculty of Physics and Technolgy at Vilnius Pedagogical University. In 2004–2008 studied in Fundamental Sciences Faculty PhD programme at Vilnius Gediminas Technical University.

KRŪVININKŲ PERNAŠA ULTRAPLONUOSIUOSE MANGANO OKSIDŲ SLUOKSNIUOSE

Tiriamoji problema ir darbo aktualumas. Disertacijoje nagrinėjami lantano manganitai, pasižymintys faziniu virsmu iš paramagnetinės į feromagnetinę būseną bei milžiniškos neigiamos magnetovaržos efektu. Tiriami magnetovaržos ir jos anizotropijos efektai silpnuose (iki ≈ 0,5 T) magnetiniuose laukuose bei stiprių impulsinių srovių ir magnetinių laukų sukelti efektai plonuose epitaksiniuose manganitų sluoksniuose. Pateikiami pasiūlymai tyrimo rezultatus panaudoti kuriant magnetinio lauko jutiklius, sparčiųjų elektrinių impulsų formuotuvus bei amplitudės moduliatorius.

Tiriant silpnų magnetinių laukų poveikį ultraplonųjų La-Sr-MnO_3 sluoksnių elektriniam laidumui, buvo nustatyta, kad magnetovaržos anizotropijos ženklas ir vertė šiuose laukuose priklauso nuo sluoksnio storio. Pateiktas modelis, paaiškinantis eksperimentinius rezultatus, paremtas vidutinio lauko artiniu ir įskaito sluoksnio struktūros kitimą kintant jo storiui. Aptiktas ir ištirtas grįžtamasis termoelektrinis perjungimas, išaiškintos šio reiškinio atsiradimo priežastys. Pasiūlytas ir eksperimentiškai realizuotas naujas ns trukmės elektrinių impulsų amplitudės moduliavimo išoriniu magnetiniu lauku būdas, naudojant epitaksinius $La_{0,87}Sr_{0,17}MnO_3$ sluoksnius.

Darbo tikslas ir uždaviniai

Šio darbo tikslas – ištirti ultraplonųjų (4–140 nm storio) epitaksinių $La_{0,83}Sr_{0,17}MnO_3$ sluoksnių, užaugintų metalo organinių junginių cheminio nusodinimo iš garų fazės metodu ant $NdGaO_3$ padėklų, elektrinį laidumą silpname (iki 0,6 T) magnetiniame ir stipriame (iki 100 kV/cm) elektriniame laukuose.

Tikslui pasiekti darbe buvo numatyta išspręsti šiuos uždavinius:

1. ištirti, kaip $La_{0,83}Sr_{0,17}MnO_3$ sluoksnių magnetovarža silpnuose (20–600 mT) magnetiniuose laukuose priklauso nuo sluoksnio storio;
2. atlikti magnetovaržos anizotropijos epitaksiniuose $La_{0,83}Sr_{0,17}MnO_3$ sluoksniuose analizę ir sukurti modelius, paaiškinančius šio reiškinio ypatumus;
3. sukurti metodiką didelės galios nanosekundinės trukmės elektrinių impulsų poveikio $La_{0,83}Sr_{0,17}MnO_3$ sluoksnių elektriniam laidumui tirti;
4. atlikti epitaksinių $La_{0,83}Sr_{0,17}MnO_3$ sluoksnių elektrinio laidumo kinetikos tyrimus, veikiant šiuos sluoksnius stipriu impulsiniu elektriniu lauku;
5. sukurti metodiką, leidžiančią sluoksnius vienu metu paveikti stipriu impulsiniu magnetiniu lauku ir ns trukmės elektrinio lauko impulsu;
6. ištirti kaip, naudojant epitaksinius $La_{0,83}Sr_{0,17}MnO_3$ sluoksnius, moduliuoti aukšto dažnio elektrinių impulsų amplitudę, keičiant išorinio magnetinio lauko indukciją.

Mokslinis naujumas

Atlikus tyrimus buvo gauti tokie kietojo kūno elektronikos moksle nauji rezultatai:

1. nustatyta, jog $La_{0,83}Sr_{0,17}MnO_3$ sluoksnių, užaugintų ant (100) orientacijos $NdGaO_3$ padėklų magnetovaržos anizotropijos ženklas ir dydis priklauso nuo sluoksnio storio. Parodyta, jog ši sluoksnių savybė atsiranda dėl anizotropinės magnetovaržos bei magnetokristalinės anizotropijos reiškinių ir gali būti panaudota, kuriant magnetinio lauko krypties matuoklius;
2. aptiktas ir ištirtas grįžtamasis termoelektrinis nestabilumas bei spartusis elektroninis perjungimas iš didelės varžos būsenos į mažos varžos būseną, atsirandantis epitaksiniuose $La_{0,83}Sr_{0,17}MnO_3$ sluoksniuose, veikiant juos stipriu elektriniu lauku. Išaiškintos priežastys, iššaukiančios šių sluoksnių suardymą nanosekundinės trukmės didelės galios impulsais;

3. pasiūlytas ir eksperimentiškai realizuotas naujas nanosekundinės trukmės elektrinių impulsų amplitudės moduliavimo išoriniu magnetiniu lauku, naudojant magnetovaržos reiškinį epitaksiniuose $La_{0,83}Sr_{0,17}MnO_3$ sluoksniuose, būdas.

Praktinė vertė. Aptikta, jog nanometrų storio $La_{0,83}Sr_{0,17}MnO_3$ sluoksniai, užauginti ant (100) orientacijos $NdGaO_3$ padėklų, pasižymi struktūros anizotropija padėklo plokštumoje. Kai elektros srovė sluoksnyje teka išilgai (010) krypties, magnetovaržos anizotropija kambario temperatūroje padėklo plokštumoje neviršija 2 %, o plokštumoje statmenoje padėklui siekia 30 %. Tai gali būti panaudota, kuriant silpnų (iki 0,5 T) magnetinių laukų krypties matuoklius. Eksperimentiškai parodyta, kaip naudojant epitaksinius $La_{0,83}Sr_{0,17}MnO_3$ sluoksnius išoriniu magnetiniu lauku galima moduliuoti aukšto dažnio elektrinių signalų amplitudę ir nustatyta, kokios maksimalios galios signalai gali būti moduliuojami tokiu būdu. Aptiktas termoelektrinio nestabilumo reiškinys gali būti panaudotas didelės galios ns trukmės impulsų formuotuvuose.

Ginamieji teiginiai

1. Ultraplonųjų $La_{0,83}Sr_{0,17}MnO_3$ sluoksnių, užaugintų ant (100) orientacijos $NdGaO_3$ padėklų, magnetovaržos ir jos anizotropijos padėklo plokštumoje dydį ir priklausomybę nuo temperatūros bei sluoksnio storio silpname magnetiniame lauke nulemia dvifazė šių sluoksnių struktūra.
2. Magnetovaržos ir jos anizotropijos ypatumus epitaksiniuose $La_{0,83}Sr_{0,17}MnO_3$ sluoksniuose gerai aprašo modelis, kuriame sluoksnio įmagnetėjimas apskaičiuojamas naudojant vidutinio magnetinio lauko artinį ir laikant, kad struktūrinių fazių tūrių santykis priklauso nuo sluoksnio storio.
3. Stiprus, viršijantis 30 kV/cm, impulsinis elektrinis laukas $La_{0,83}Sr_{0,17}MnO_3$ sluoksniuose iššaukia spartųjį grįžtamąjį termoelektrinį nestabilumą, kurio pagrindinius ypatumus gerai aprašo vienalyčio adiabatinio kaitimo modelis. Kai elektrinio lauko stipris viršija 50 – 70 kV/cm, per kelias nanosekundes išsivysto elektroninės prigimties sluoksnio perjungimas iš didelės varžos į mažos varžos būseną, kurioje srovė, tekėdama siaura gija, iššaukia elektrodų medžiagos suardymą.
4. Epitaksiniai $La_{0,83}Sr_{0,17}MnO_3$ sluoksniai gali būti panaudoti kuriant magnetiniu lauku valdomus subnanosekundinės trukmės elektrinių impulsų amplitudės moduliatorius bei magnetinio lauko krypties matuoklius.

Rezultatų aprobavimas. Disertacijos tema paskelbti du straipsniai žurnaluose, įtrauktuose į Thomson ISI Web of Science sąrašą, vienas – konferencijų medžiagoje, referuotoje Thomson ISI Proceedings duomenų bazėje, keturi – respublikinių konferencijų medžiagose, perskaityti 7 pranešimai Lietuvos bei užsienio konferencijose.

Darbo apimtis ir struktūra. Disertaciją sudaro reziumė lietuvių ir anglų kalbomis, įvadas, šeši skyriai, pagrindiniai rezultatai ir išvados, literatūros sąrašas, publikacijų disertacijos tema sąrašas.

Įvadiniame skyriuje nagrinėjamas problemos aktualumas, formuluojamas darbo tikslas bei uždaviniai, aprašomas mokslinis darbo naujumas, pristatomi autoriaus pranešimai ir publikacijos, disertacijos struktūra.

Pirmasis skyrius skirtas literatūros apžvalgai. Jame pateikta literatūros analizė apie pagrindines manganitų savybes ir atliktus tyrimus, plonuosius La-Sr-MnO_3 sluoksnius veikiant magnetiniu lauku bei stipria elektros srove.

Antrajame skyriuje pateikiami eksperimentinės įrangos schemos ir bandinių gamybos technologijos aprašymai.

Trečiajame skyriuje aprašomi nanometrų storio $La_{0,87}Sr_{0,17}MnO_3$ sluoksnių magnetovaržos bei jos anizotropijos tyrimai silpname magnetiniame lauke. Aptariami būdai ir galimybės šį reiškinį panaudoti kuriant magnetinio lauko krypties matuoklius.

Ketvirtasis skyrius skirtas reiškiniams, atsirandantiems paveikus manganitų sluoksnius stipriu impulsiniu ns trukmės elektriniu lauku.

Penktasis ir šeštasis skyriai skirti stiprios impulsinės srovės nulemto perjungimo reiškinio manganitų sluoksniuose tyrimui bei nanosekundinių impulsų amplitudės moduliavimui, naudojant La-Sr-MnO_3 sluoksnių magnetovaržą.

Pagrindiniai darbo rezultatai ir išvados

1. Eksperimentiškai ištirta nanometrų storio $La_{0,83}Sr_{0,17}MnO_3$ sluoksnių magnetovaržos anizotropija silpnuose (20–600 mT) magnetiniuose laukuose. Nustatyta, jog sluoksnių, užaugintų ant (100) orientacijos $NdGaO_3$ padėklų, magnetovaržos anizotropijos ženklas ir dydis priklauso nuo sluoksnio storio. Parodyta, jog ši sluoksnių savybė atsiranda dėl anizotropinės magnetovaržos bei magnetokristalinės anizotropijos reiškinių, kurių santykinis indėlis į bendrą magnetovaržos vertę kinta priklausomai nuo sluoksnio storio.
2. Naudojant vidutinio lauko artinį ir įskaitant nanometrų storio sluoksnių struktūros kitimą, kintant storiui, atlikta magnetovaržos anizotropijos reiškinio analizė ir naudojant vidutinio lauko artinio modelį paaiškinti šio reiškinio ypatumai.

3. Parodyta, jog 20 nm storio epitaksiniai $La_{0,83}Sr_{0,17}MnO_3$ sluoksniai, užauginti ant (100) orientacijos $NdGaO_3$ padėklų, pasižymintys struktūros anizotropija padėklo plokštumoje, gali būti panaudoti, kuriant silpnų (iki 0,5 T) magnetinių laukų krypties matuoklius. Tokių matuoklių magnetovaržos anizotropija kambario temperatūroje padėklo plokštumoje neviršytų 2 %, tuo tarpu plokštumoje statmenoje padėklui siektų 30 %.
4. Sukurta eksperimentinė metodika didelės galios nanosekundžių trukmės elektrinių impulsų poveikio mangano oksido sluoksnių elektriniam laidumui tirti. Atlikti epitaksinių $La_{0,83}Sr_{0,17}MnO_3$ sluoksnių elektrinio laidumo kinetikos tyrimai, veikiant šiuos sluoksnius stipriu impulsiniu elektriniu lauku. Aptiktas ir ištirtas grįžtamasis termoelektrinis nestabilumas, bei spartus perjungimas iš didelės varžos į mažos varžoas būseną, nustatytas sluoksnio suardymo mechanizmas, vykstant šiam perjungimui. Parodyta, jog jį sukelia elektroninės prigimties stipraus elektrinio lauko efektai.
5. Sukurta metodika leidžia tuo pačiu metu mangano oksido sluoksnius paveikti stipriu impulsiniu magnetiniu lauku ir ns trukmės elektrinio lauko impulsu. Ištirtos galimybės, naudojant epitaksinius $La_{0,83}Sr_{0,17}MnO_3$ sluoksnius, moduliuoti aukšto dažnio elektrinių impulsų amplitudę, keičiant magnetinio lauko indukciją.

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TRANSPORT OF CHARGE CARRIERS IN ULTRATHIN FILMS OF MANGANESE OXIDES

Summary of Doctoral Dissertation
Physical Sciences, Physics (02P), Condensed Matter: Electronic Structure, Electrical, Magnetic and Optical Properties, Superconductors, Magnetic Resonance, Relaxation, Spectroscopy (P260)

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KRŪVININKŲ PERNAŠA ULTRAPLONUOSIUOSE MANGANO OKSIDŲ SLUOKSNIUOSE

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