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Magnetic Properties of magnetron sputtered $La_{0.7}Sr_{0.3}MnO_3$ thin films

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The results of studying the influence of the thickness of $La_{0.7}Sr_{0.3}MnO_3$ films obtained by magnetron sputtering on (110) $NdGaO_3$ substrates on the magnetic and crystallographic properties using ferromagnetic resonance and X-ray spectroscopy are presented. The dependences of the uniaxial and cubic anisotropy fields on the sample thickness are established. Furthermore, it is shown that the magnetic and crystallographic properties of a film obtained by magnetron sputtering strongly depend on the target region from which it is made. The results obtained will be useful for interpreting the experimental data and creating a series of samples.

Keywords: ferromagnet, magnetic anisotropy, ferromagnetic resonance.

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Manganites attract a great deal of research attention owing to their unique properties: strong spin polarization [1,2], colossal magnetoresistance [3], and high Curie temperature [4]. Several studies focused on the examination of magnetic properties of manganites and their variation with sample thickness have already been published [5–7]. A significant increase in the anisotropy of samples at small thicknesses was shown [6]. Researching of the dependence of magnetic anisotropy on the film thickness may provide additional data on the thickness values needed to establish bulk properties of thin $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) films. The position and shape of ferromagnetic resonance lines may provide information regarding the magnetic state, local anisotropy, and defects [8–10]. The dependence of magnetic characteristics of LSMO on the strain induced by a substrate has been examined in [11,12], and their dependence on thickness and oxygen vacancies has been studied in [13]. However, the properties of LSMO films produced by magnetron sputtering from different parts of a target have not been characterized yet. The aim of the present study is to examine the influence of the sputtering region from which sputtering was carried out on the magnetic and crystallographic characteristics of LSMO films.

The structure of the target is presented in Fig. 1. The target was fabricated by solid-phase synthesis. Manganese (Mn_2O_3) and lanthanum (La_2O_3) oxides and strontium carbonate ($SrCO_3$) were used as the initial components for fabrication. These components were taken in a stoichiometric ratio and ground in an agate mortar for 30 min. The obtained homogeneous mixture was compacted using a metallic mold and a hydraulic press. The mold diameter was 25 mm. A pellet obtained after pressing was positioned on a platinum sheet and sintered in a tube furnace at a temperature of 1350°C for 5 h.

The phase composition of the prepared target was determined by X-ray diffraction. X-ray diffraction analysis revealed that the target features one crystalline phase corresponding to $La_{0.7}Sr_{0.3}MnO_3$. The results of X-ray spectrum analysis confirmed that the composition of the target matches the pre-defined elemental composition.

Thin LSMO films were deposited onto single-crystal polished planes of (110) $NdGaO_3$ (NGO) substrates 5×5 mm

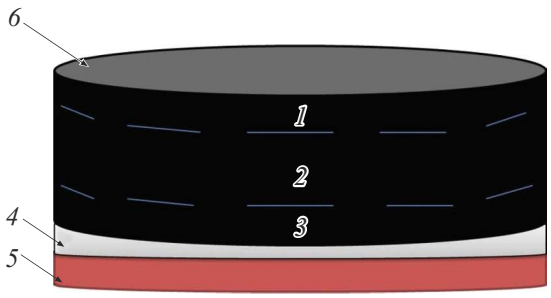

Figure 1. Diagram of the target. The target is split tentatively into three regions *1* — „top“, *2* — „middle“, and *3* — „bottom“. LSMO (*6*) is soldered with indium *4* to copper substrate *5*.

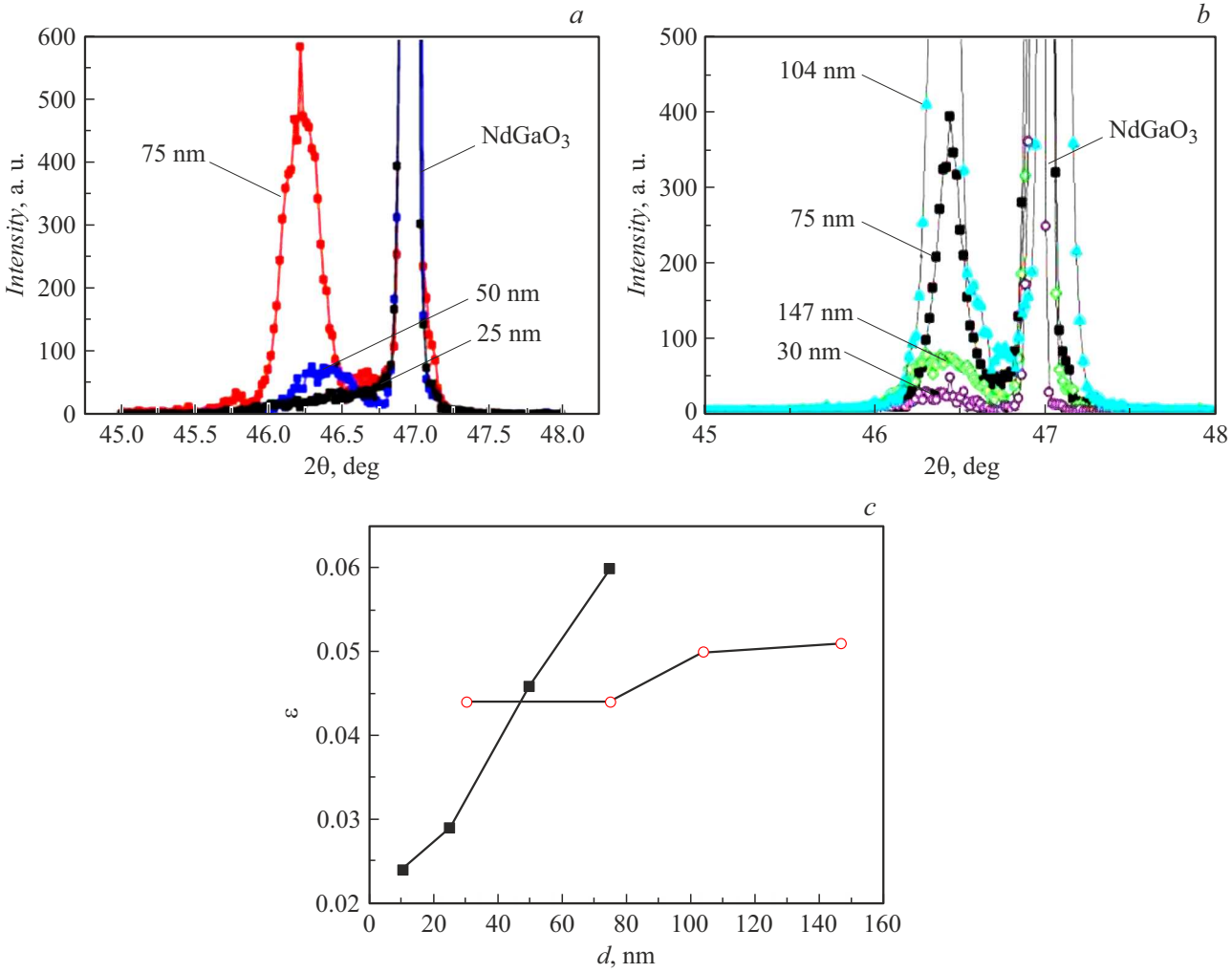

Figure 2. X-ray diffraction patterns of $La_{0.7}Sr_{0.3}MnO_3$ films grown on an NGO substrate with different film thicknesses (*a* — the first series, *b* — the second series). *c* — Dependence of the lattice mismatch between $La_{0.7}Sr_{0.3}MnO_3$ and the substrate in the perpendicular direction on the film thickness for samples of the first (squares) and the second (circles) series.

in size and 0.5 mm in thickness. Substrates were subjected to preparatory treatment: cleaned with an organic solvent (acetone) in an ultrasonic bath, rinsed with distilled deionized water, and dried in a flow of compressed air. Epitaxial growth of strontium and lanthanum manganite films was performed at a substrate temperature of 800°C in a mixture of Ar and O_2 (3:2) gases under a pressure of 0.5 mbar with a power of 50 W of a high-frequency generator and a magnetron gun. The process of epitaxial growth is understood here as the growth of a film oriented by a single-crystal substrate onto the surface of which the material is deposited. Epitaxy proceeds in such a way that the overall boundary energy with contributions from the substrate–crystal, crystal–environment, and substrate–environment regions is kept at a minimum. Figure 2 suggests that the crystal structure of all the films considered in the present study is single-phase and their parameters are governed by the crystal structure of the NGO substrate.

The growth rate was 0.25 nm/min at the indicated parameters. Following the deposition of a film of the needed thickness, oxygen was pumped into the chamber at a flow pressure of 1 atm with a gradual reduction of temperature to 500°C and subsequent free cooling to room temperature.

Two series of samples deposited from different parts of the target were prepared in order to examine the magnetic parameters of LSMO films of different thickness. The first series was sputtered from the „bottom“ part of the target, and the second series was fabricated from the „middle“ part. Figures 2, *a*, *b* present the patterns of Bragg reflection $2\theta/\omega$ for two series of thin LSMO films deposited on the NGO substrate. The cell structure and parameters were determined by examining the positions of peaks. Characteristics were measured at room temperature. The obtained results suggest that the LSMO film structure is „pseudocubic“ [14].

Figure 2, *c* presents the results of X-ray diffraction measurements of lattice constants for two series of LSMO

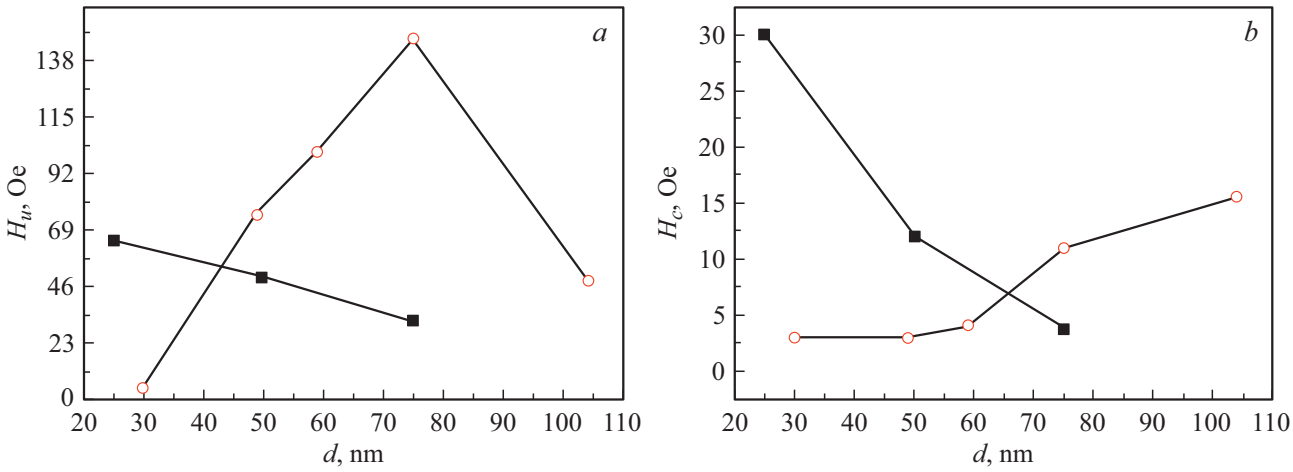

Figure 3. Dependences of uniaxial magnetic anisotropy H_u (a) and cubic magnetic anisotropy H_c (b) fields on the film thickness for samples of the first (squares) and the second (circles) series.

films. The sample thickness is plotted on the abscissa, and difference $\varepsilon = \frac{a_{\mathrm{LSMO}} - a_{\mathrm{NGO}}}{a_{\mathrm{NGO}}}$ (a is the out-of-plane lattice constant) between lattice constants of LSMO and the substrate in the perpendicular direction in plotted on the ordinate axis. The lattice structure and parameters were determined by examining the positions of peaks. At a film thickness of 75 nm, the out-of-plane lattice constant of the LSMO layer is maximized in the first series of samples, which was sputtered from the „bottom“ part of the target. Note that the thickness dependence of ε for films of the second series differs in being nonlinear from the dependence for the first series.

Figure 3 shows the thickness dependences of magnetic LSMO parameters: uniaxial (a) and cubic (b) magnetic anisotropy fields. Measurements were performed using a standard Bruker ER 200 spectrometer at a frequency of 9.2 GHz and room temperature [15]. The parameters of magnetic anisotropy were determined by processing the angular dependences of resonance fields of ferromagnetic resonance spectra. A solution of the Landau–Lifshitz equation for the evolution of magnetization M in an external constant magnetic field H under the influence of the magnetic component of a radio-frequency field was used. This solution yields an analytical relation for resonance field H_0 and frequency ω for a system with a single ferromagnetic layer [16]. A dip is seen clearly in all dependences for a 75-nm-thick film from the first series, which was produced from the „bottom“ part of the target, while the second series has a peak at 75 nm (although for uniaxial anisotropy only). A similar dependence was reported in [6] for film thicknesses of 11–22 nm. Strong enhancement of magnetic anisotropy in LSMO films with a thickness of 11 nm was associated with anisotropic strain. In LSMO with increasing thickness, the formation of satellite domains in films with a unidirectional lattice modulation should provide an explanation for the marked suppression of in-plane magnetic anisotropy. This explanation appears inapplicable to the present case, since the uniaxial anisotropy peak in the second series of samples, which is believed to correspond to „proper“ $La_{0.7}Sr_{0.3}MnO_3$, is found at a thickness of 75 nm (i.e., the thickness at which a film should be relieved of stress). We suspect that, in contrast to [6], satellite peaks form at thicknesses greater than 75 nm in the sputtering mode set in our experiments. An enhancement of cubic anisotropy attributable to the film crystallography explains the marked suppression of in-plane magnetic anisotropy, since growth satellite domains, which cause suppression of anisotropic stress as the LSMO film thickness increases.

It should be noted that films were grown under the same conditions (pressure, temperature). This excludes the possibility of emergence of oxygen vacancies that may affect the anisotropy parameter. In addition, samples of the entire series were grown on (110) NGO substrates; therefore, differences in stress induced in epitaxial films [12] are also out of the question. LSMO-type oxides themselves are regarded as mixed-valence compounds, which are solid solutions between $LaMnO_3$ and $SrMnO_3$. We believe that the difference in magnetic properties of thin LSMO films formed by magnetron sputtering of different parts of a target is attributable to the fact that the stoichiometry of the target material varies throughout its thickness. In other words, although the phase composition of the prepared target was monitored by X-ray diffraction, the stoichiometry may vary due to ion-electron emission under the influence of ion bombardment in the course of sputtering. Thus, manganite of an undetermined phase was produced in the first series, which makes it difficult to interpret the results for this series. In our view, the suppression of uniaxial anisotropy is related to the fact that a film with a thickness on the order of 75 nm becomes similar in terms of its crystallographic parameters to a crystal and is thus relieved of the stress induced by an NGO substrate.

The obtained data suggest that the uniaxial anisotropy field of epitaxial $La_{0.7}Sr_{0.3}MnO_3$ films grown on (110)

$NdGaO_3$ substrates intensifies as the film thickness grows to 75 nm, but decreases sharply in magnitude at greater thicknesses due to the formation of growth satellite domains. It is also likely that the sputtering regime has an effect on the uniaxial anisotropy maximum. Note that consistent characteristics of films formed by magnetron sputtering of a target may be achieved only if a series of samples is produced from one and the same part of a target. The reason for this is that the stoichiometric composition of targets varies over their thickness.

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Conflict of interest

The authors declare that they have no conflict of interest.

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