Magnetic and dielectric properties of o-LuFeO3/SrTiO3

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Abstract. Here we present the investigation of the orthorhombic o-LuFeO3 thin film grown on SrTiO3 (001) substrate. Temperature dependencies of magnetization measured in magnetic fields 1000 Oe and 2000 Oe were described in the assumption that ferromagnetic correlated regions are presented in samples together with the paramagnetic phase. Additional argument in favor of the ferromagnetic regions existence in o-LuFeO3 is the magnetic hysteresis curve measured at T=5 and 100K. The temperature dependences of dielectric response were measured and analyzed.

1. Introduction

The LuFeO3 can be crystalized in two modifications: hexagonal and orthorhombic. Thin films of the hexagonal LuFeO3 (h-LuFeO3) deposited on Al2O3 and YSZ are a multiferroic material that exhibits spontaneous electric and magnetic polarizations simultaneously [1, 2], which have great importance for future technologies. The orthorhombic crystallographic structure o-LuFeO3 is the thermodynamically stable with standard condition preparation [3], the hexagonal structure is metastable. The free energy of o-LuFeO3 is a few lower than that of the h-LuFeO3. In epitaxial thin films, the film-substrate interfacial energy may favor the h-LuFeO3 structure, if the symmetry of the substrate is triangular or hexagonal. This effect can stabilize the hexagonal structure in epitaxial thin films, to a certain critical thickness [1, 4] Therefore, when LuFeO3 is deposited onto substrate in the form of thin film it can coexist in the form of hexagonal and orthorhombic domains that was established in work [4] where two structural phases of LuFeO3 occurs on a length scale of micrometer, as visualized in real space using X-ray photoemission electron microscopy, and moreover, there is the structural transition at about 1000 °C, from the hexagonal to the orthorhombic phase of LuFeO3 [4] and then it results to interesting physical properties.

The crystal structure of the hexagonal h-LuFeO3 belongs to the space group P6_3cm with lattice parameters a = b = 5.993 Å, c = 11.634 Å and α=β=90°, γ=120°. The crystal structure of the orthorhombic o-LuFeO3 belongs to the space group Pnma with lattice parameters a=5.547 Å; b=7.565 Å; c=5.213 Å. The crystal structure of the substrate material SrTiO3 is Pm3m; a = b = c = 3.9046 Å, α
= \beta = \gamma = 90^\circ \) (T>105 K) and \( I4/mcm \). Magnetic and dielectric orders in orthorhombic and hexagonal structures LuFeO\(_3\) are observed in different temperatures. The strong magnetic order in \( o\)-LuFeO\(_3\) happens at \( T_N = 620 \) K and in \( h\)-LuFeO\(_3\) at \( T_N = 132 \) K [3]. Ferroelectric transition in \( h\)-LuFeO\(_3\) was observed below \( T_C = 1050 \) K connecting from a \( P6_3/mmc \rightarrow P6_3/cm \) structure distortion [3].

2. Samples preparation and experimental details

Single-phase polycrystalline LuFeO\(_3\) targets were obtained by solid-state synthesis using stoichiometric quantities of Lu\(_2\)O\(_3\) and Fe\(_2\)O\(_3\) powders as the starting materials. \( o\)-LuFeO\(_3\) thin films were deposited from the prepared targets by RF-magnetron sputtering with the “facing-target” scheme [5], we used the same technique for the deposition of manganite films in our previous work [6]. For the growth of orthorhombic \( o\)-LuFeO\(_3\) films \( (001) \) oriented SrTiO\(_3\) (STO) single-crystal substrates were used. A temperature of substrate during the deposition was 700 °C. A mixture of Ar and O\(_2\) with pressure 1–2 mTorr was used as the working atmosphere. The thickness of the obtained \( o\)-LuFeO\(_3\) films was 100 nm. The structure of the grown thin films was studied by X-ray diffraction (Bruker D8 DISCOVER, using Cu K\( \alpha \) radiation).

The magnetization was measured on the PPMS-9 device in the temperature range from 4 to 300 K and in magnetic field up to 2T. For simplicity presentation of the magnetization and magnetic susceptibility of \( o\)-LuFeO\(_3\) we subtracted a diamagnetic contribution of a substrate. In order to separate contributions of the thin film and substrate we measured the mass of a sample and defined the number of moles of SrTiO\(_3\) as \( v=m/\mu \), neglecting the mass of the thin film in comparison with the mass of the substrate. The diamagnetic susceptibility of substrate SrTiO\(_3\) is equal \( \chi_{\text{diamag}} = -6.0 \times 10^{-5} \) emu/mol and can be estimated using diamagnetic susceptibilities of single ions: \( \text{Sr}^{2+} -19; \text{Ti}^{4+} -5; \text{O}^{2-} -12 \) given in \( 10^{-6} \) emu/mol [7].

The dielectric response was studied at frequency range 0.1 Hz - 10 MHz and temperature region 100-450 K using ultra broadband dielectric spectrometer Novocontrol BDS80 with Quatro-cryosystem. Platinum IDE electrodes were deposited on the thin film by electron beam lithography method.

3. Experimental results

3.1. X-ray diffraction analysis

Figure 1 shows the \( \Theta-2\Theta \) scan of a LuFeO\(_3\)\( ||\)STO(001) sample. The peaks correspond to the (001) reflections of the substrate, as well as to the reflections from the (101) plane of the orthorhombic \( o\)-LuFeO\(_3\). This suggests that the films are textured, with the out-of-plane orientation \( o\)-LuFeO\(_3\)\( (101) ||\)STO(001). The out-of-plane \( d_{\text{(101)}} = 3.830 \) Å, somewhat larger than the value for bulk material, i.e. \( d_{\text{bulk\,(101)}} = 3.799 \) Å. The strain is \( \varepsilon_{\text{(101)}} = 0.8\% \).

3.2. Magnetization and magnetic susceptibility

Temperature dependence of magnetization in \( o\)-LuFeO\(_3\)/SrTiO\(_3\) was measured at 1000 Oe and 2000 Oe magnetic field in a wide temperature range. The temperature dependence of the magnetic susceptibility obtained as \( \chi = M/H \) is presented in Figure 2a for both experiments. It is clear visible the difference between absolute values of the magnetic susceptibility measured in different magnetic field, which suggests the existence of the ferromagnetic regions in the sample. In the case of coexistence of the ferromagnetic and paramagnetic phases the magnetic susceptibility can be calculated as:

\[
\chi = \chi_{\text{ferm}} + \chi_{\text{par}},
\]

where \( \chi_{\text{par}} \) is paramagnetic contribution, described by the Curie low \( \chi_{\text{par}} = C/T \); \( \chi_{\text{ferm}} = M_{\text{ferm}}/H \), where \( M_{\text{ferm}} \) is the temperature independent ferromagnetic magnetization. The fitting of the temperature dependence of the magnetic susceptibility of \( o\)-LuFeO\(_3\) thin film was performed using the equation (1) and fit parameters \( C = 4 \times 10^{-8} \) emu-K, \( M_{\text{ferm}}(1000 \) Oe) \( = 1.15 \times 10^{-5} \) emu-Oe; \( M_{\text{ferm}}(2000 \) Oe) \( = 1.9 \times 10^{-5} \) emu-Oe; fit curves are given in Figure 2a.
To get the additional argument of the ferromagnetic phase existence in o-LuFeO$_3$ we have performed the measurements of the field dependence of the magnetization at T=5 and 100K (see Figure 2b). The hysteresis loop observed in the magnetic field range from -0.6 kOe to 0.6 kOe for both temperatures 5K and 100 K. Therefore ones again we conclude that ferromagnetic regions are present in the sample together with the paramagnetic phase in o-LuFeO$_3$ thin films deposited on SrTiO$_3$. Previously weak ferromagnetism and hysteresis loops were observed in the bulk o-LuFeO$_3$ [8], assuming the preserving of the magnetic properties during the transition from bulk samples to thin films.

![Figure 1. $\theta$–2$\theta$ XRD scan of a o-LuFeO$_3$ film on the STO(001) substrate.](image)

![Figure 2. a) Temperature dependencies of M/H- $\chi_{\text{diam}}$ for LuFeO$_3$/SrTiO$_3$ in magnetic fields 1000 Oe and 2000 Oe; fitting was performed using the equation (1) (see the text); b) hysteresis curve measured at a temperature of 5 and 100K for LuFeO$_3$/SrTiO$_3$.](image)

3.3. Dielectric behavior

Temperature dependence of complex dielectric response of o-LuFeO$_3$/SrTiO$_3$ was measured at the frequency range 0.1 Hz - 10 MHz and at temperatures 100-450 K. In order to separate a contribution from the thin film and the substrate we measured dielectric properties of SrTiO$_3$ substrate with the
same electrodes and in the same conditions. Calculated temperature dependencies of dielectric permittivity $\varepsilon'$ of the thin film o-LuFeO$_3$ for several measuring frequencies are presented in Figure 3a. The schematic view and optic images of the sample with electrodes are given in Figure 3b and 3c, respectively.

The anomaly of the intrinsic dielectric response was observed in [3] near the magnetic transition point $T_N \approx 600$ K that clearly told about a coupling between dielectric and magnetic properties in o-LuFeO$_3$. In our case this anomaly has corresponded to the observed increase of $\varepsilon'$ with increasing temperature up to 450 K. We also observed a frequency dependent maximum of permittivity near 130 K. Similar low temperature anomaly, indicating possible charge ordering in the temperature range of 150-300 K, was observed in o-LuFeO$_3$ ceramics in [9], assuming the preserving of the dielectric properties during the transition from bulk samples to thin films how it was in the case of magnetic properties.

4. Conclusions
The magnetic and dielectric properties of the radio frequency magnetron sputtered orthorhombic LuFeO$_3$ thin films were investigated. Magnetic properties of o-LuFeO$_3$/SrTiO$_3$ thin film were described in the assumption that ferromagnetic correlated regions are presented in samples together with the paramagnetic phase. We also observed a frequency dependent maximum of permittivity near 130 K, indicating possible charge ordering in the temperature range of 150-300 K.
4. Acknowledgments
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