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Structure and Magnetism of LSMO/BTO/MgO/LSMO Multilayers

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A multiferroic tunnel junction (MFTJ) is a promising device for future memory systems with discrete and different logic states which are controlled by a combination of electric and magnetic fields. The goal of ongoing research is to present ferroelectric and ferromagnetic properties, especially at room temperature (RT), represented as high values of tunnel electroresistance (TER) and tunnel magnetoresistance (TMR). A key aspect is the appropriate preparation of a sample allowing epitaxial growth. The thin layers were prepared by pulsed laser deposition on atomically smooth monocrystalline $SrTiO_3$ (STO) substrates. The ferromagnetic metal layers $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) are separated by a layer of a ferroelectric insulator - $BaTiO_3$ (BTO). The same structure of LSMO, BTO and STO (perovskite) and a similar lattice constant make it possible to obtain high-quality heterostructures. Magnetic measurements confirm differences in the magnetic coercivity of the top and bottom LSMO layer, which allows to obtain their parallel and antiparallel magnetization orientation. A modification of the interfaces of BTO by thin MgO layer enables an increase in the value of the TER effect.

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1. Introduction

Nowadays, Si-based electronics reaches its physical limits (the mobility of electric charge carriers, the speed and size of a gate in a transistor of about a few atoms predicted to be achieved in 2020, the density of information storage, etc.). Therefore, many efforts are focused on the ongoing research towards next-generation computation and data storage based on new kinds of physical phenomena. One of the most promising approaches is spintronics, which utilizes not only the charge of the electron, but also its spin. Two milestones of this research, which affect everyday life, are: (1) the giant magnetoresistance (GMR) [1] and the tunnel magnetoresistance (TMR) [2] effect (currently, they are applied in hard disks) and (2) spin transfer torque (STT) [3], which is successfully used for magnetization switching in magnetic random access memory (MRAM) cells [4]. Nowadays, research is focused, among many others, on multiferroic tunnel junctions (MFTJ), which combine ferroelectric tunnel junctions (FTJ) with ferromagnetic tunnel junctions (MTJ). This kind of device shows four different resistance values for different polarizations of the ferroelectric tunneling barrier (FE) and for different magnetization orientations of the ferromagnetic electrodes (FM). The existence of four logic states distinguishable at room temperature is a practical requirement for MRAM applications. The parameters describing the difference in resistance for different electric polarizations and different magnetization orientations are tunnel electroresistance (TER) and TMR, respectively. A TER value of the order of 10000% is obtained at low temperature in case of LSMO/BTO/Co junction [5]. However, high values of TER usually occur in combination with a small TMR effect and with no upper electrode, which is replaced by a tip in scanning tunneling microscopy (STM) experiments. The use of half-metals for electrodes (100% spin polarization at the Fermi level) allows obtain a reasonable TMR [6].

One of the best candidates for the FM electrodes of the MFTJ, used in our experiment, is $La_{0.67}Sr_{0.33}MnO_3$(LSMO), which is ferromagnetic at RT and has nearly 100% spin polarization at the Fermi level [6]. In turn, $BaTiO_3$ (BTO) is used as the FE barrier in our MFTJs because of its structural properties which are complementary to LSMO. Hence, an MFTJ can be built as an LSMO/BTO/LSMO trilayer structure. However, using the same material for both the FM electrodes, and separating them by the FE barrier only, results in some disadvantages. In particular, the reversed electric polarization does not sufficiently affect the TER, because of the same screening length in both the FM/FE interfaces. An idea of enhancement of the TER effect proposed by Zhuravlev et al. [7], relates to a dielectric layer placed at one of the FM/FE interfaces: the dielectric layer changes potential across the barrier and creates two different resistance values for the two different electric

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polarizations of the FE barrier. A theoretical calculation predicts an increase of the TER by using STO or MgO in the composite barrier [7]. This prediction is experimentally confirmed for STO by Ruan et al. [8]. However, the use of an MgO-FE composite barrier should result in a higher TER. The effect of MgO has not been proved experimentally as yet, probably because of the lattice mismatch ($a_{\mathrm{STO}} = 3.905$, $a_{\mathrm{LSMO}} = 3.88$, $a_{\mathrm{BTO}} = 4.033$, $a_{\mathrm{MgO}} = 4.216$). In this paper, we report on structural and magnetic measurements of a high-quality multilayer with the insulating barrier of MgO in upper of the FM/FE interfaces (adjacent in bottom interface could affect BTO growth and therefore its ferroelectric properties), i.e. of the STO/LSMO/BTO/MgO/LSMO structure.

2. Experimental

Thin films of LSMO, BTO and MgO were grown by means of the pulsed laser deposition (PLD) technique onto the single-crystal STO (100), using the 248 nm excimer laser system (Coherent COMPexPro 110F) operated at an energy density of ~ 2 J/cm^2, a pulse width of 20 ns, and a repetition rate of 10 Hz. The targets were stoichiometric LSMO, MgO and BTO. Laser deposition was performed under 200 mTorr partial oxygen pressure and a substrate temperature of 750 °C for all thin films building up the LSMO/BTO/MgO/LSMO multilayer. After deposition the samples were cooled down with oxygen pressure increasing up to the value of 50 Torr.

The structural quality and morphology were tested by transmission electron microscopy (TEM). A thin foil of the STO/LSMO/BTO/MgO/LSMO sample was measured in HR mode.

The samples were measured in a vibrating sample magnetometer (VSM). Firstly, we explored the temperature dependence of magnetization for a single layer of LSMO grown on STO in the temperature range from 80 K to 350 K at 1000 Oe. The magnetic hysteresis loops of the LSMO/BTO/MgO/LSMO sample were measured at 80 K in an external magnetic field applied in the sample plane along the [100] crystallographic direction in both cases.

3. Results and discussion

3.1 Structure and morphology

A single-crystalline STO substrate and a lack of structural defects ensure epitaxial growth. The first step towards the production of an MFTJ is to properly prepare an atomically smooth substrate. Connell et al. [9] showed a simple method for such a preparation, based on annealing in air at 1000 °C for at least one hour and deionized water treatment. We applied a similar method to obtain the TiO_2-terminated STO substrate, which is preferred for LSMO deposition.

The results of such a preparation procedure were verified via atomic force microscopy (AFM). The obtained images of the surface of the TiO_2-terminated STO substrate confirm the existence of regular terraces about 1 u.c (~ 0.4 nm) high and about 100 nm wide (Fig. 1a). They result from polishing the STO crystal not precisely along the (001) surface (the miscut effect). The LSMO layer grown on the STO substrate of a bit larger miscut angle and a different miscut orientation is shown in the right image (Fig. 1b). Terraces on the STO substrate surface may cause stress in the LSMO layer, which introduces the so called step-anisotropy contributing to the total magnetic anisotropy energy.

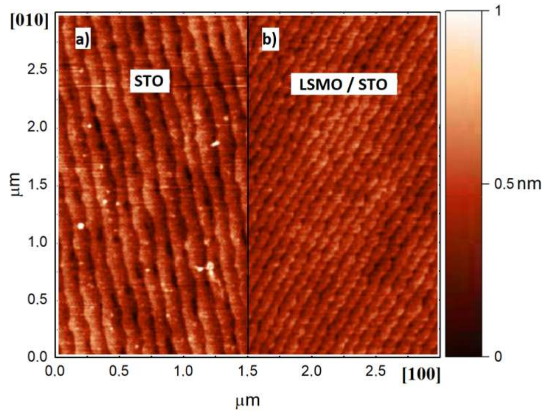

Fig. 1. AFM image of an atomically smooth surface: a) the STO substrate and b) an LSMO/STO sample.

Actually, it is still challenging to engineer high-quality and atomically sharp interfaces of the LSMO/BTO/MgO/LSMO multilayer grown on TiO_2-terminated STO substrates. Figure 2 shows a cross-sectional bright field TEM image of such a multilayer structure, which confirms high quality of our samples: the epitaxial growth of LSMO, MgO and BTO thin films along the [001] direction were obtained. Moreover, the TEM images allow to count the atomic planes, i.e. to measure precisely the thickness of each thin film building up the final structure. In a real junction the barrier thickness will be reduced down to 4 nm.

3.2 Magnetic properties

The electrical and magnetic properties determine the utility of the STO/LSMO/BTO/MgO/LSMO structure for MFTJ applications. Even a single-crystalline structure does not guarantee the expected stoichiometry and the expected values of conductance and saturation magnetization.

The results of the magnetic measurements performed on our samples are in perfect agreement with the values reported so far in the literature [10,11]. The measured Curie temperature of a single LSMO-layer is about 25 K above RT. The magnetization at 80 K is close to the value for bulk LSMO, which is 3.7 μ_{B}/Mn. The saturation magnetization at 1000 Oe measured RT is still nearly

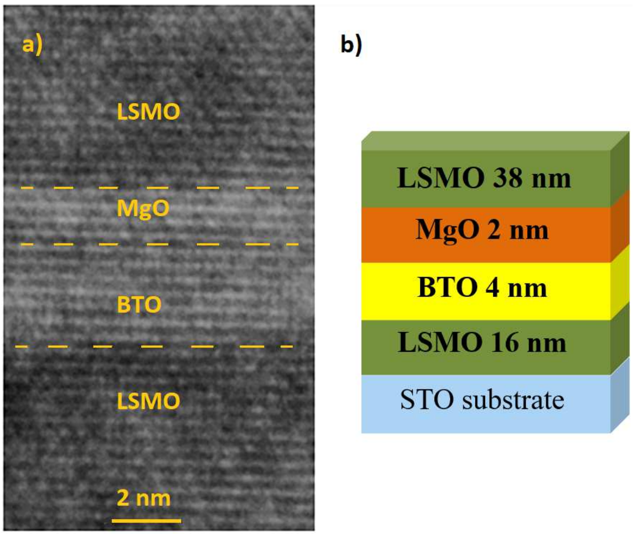

Fig. 2. a) HR-TEM image of the cross-section of the epitaxial multilayers of the tunnel barrier. b) The structure of the sample is STO/LSMO/BTO/MgO/LSMO.

1/3 of the value obtained at low temperature. It gives a hope for constructing a MFTJ operational at RT.

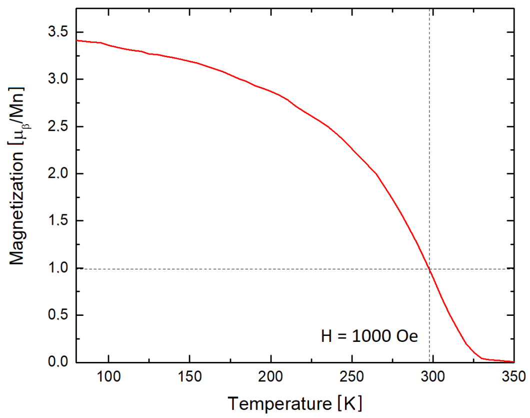

Fig. 3. Temperature dependence of the magnetic moment of the LSMO/STO sample measured in the 1 kOe in plane magnetic field.

The next step was to measure the magnetic hysteresis loops for the STO/LSMO/BTO/MgO/LSMO samples. The magnetically decoupled upper and bottom LSMO electrodes of different coercivity allow to switch the magnetization in each layer independently and set them in a parallel or antiparallel magnetization orientation.

The magnetic hysteresis loop measured at 80 K along the easy magnetization axis (i.e. along the terraces steps) (Fig. 4). The FM electrodes show different coercivity and magnetization proportional to their thickness, therefore there is a field range of antiparallel alignment. In real junction one of the electrodes will be doped with ruthenium in order to enlarge coercivity [12]. From the TEM

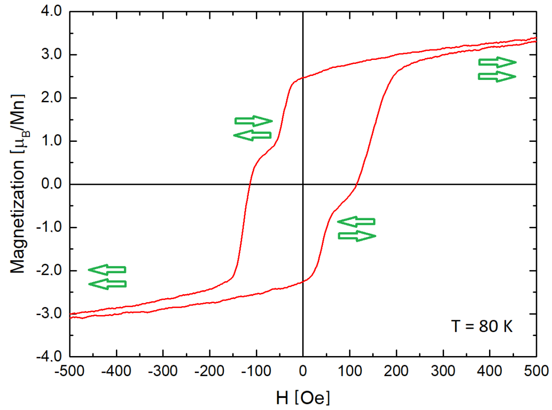

Fig. 4. Magnetic hysteresis loop of the LSMO/BTO/MgO/LSMO multilayer structure. Two different coercivities for the upper and bottom electrode allow us to independently switch each of them.

images we know that the upper layer is 38 nm and the bottom one 16 nm thick (Fig. 2). Thus, the signal from the upper layer should be much larger than from the bottom one, as confirmed by Fig. 4 (the magnetization of thinner layer switches first showing that coercivity is proportional to the thickness). We gain the same shape of the hysteresis at higher temperatures up to RT with the expected decrease in magnetization and coercivity.

The first results of magnetic measurements are promising for growing an operational MFTJ at RT. We also expect a reasonable TER due to the insertion of an MgO layer at the FE/FM interface.

4. Summary

We have grown a high-quality multilayer of STO/LSMO/BTO/MgO/LSMO as a base for an MFTJ. MgO was inserted in one of the LSMO/BTO interfaces in order to increase TER. However, such junctions have not been produced and characterized yet. We have demonstrated that the growth of such a multilayer on an atomically smooth surface of the TiO_2-terminated STO substrate is possible. Epitaxial growth and sharp interfaces were confirmed using AFM and TEM imaging. The magnetic measurements showed the expected saturation magnetization and the value of the Curie temperature above RT. The magnetic hysteresis loops showed the existence of two different coercivities for the magnetically decoupled upper and bottom electrodes, which allowed to switch their magnetization independently (and set their parallel and antiparallel orientations).

References

[1] M.N. Baibich et al., *Phys. Rev. Lett.* **61**, 2472 (1988).
[2] J.S. Moodera et al., *Phys. Rev. Lett.* **74**, 3273 (1995).
[3] M.D. Stiles et al., *Phys. Rev. B* **66**, 014407 (2002).

[4] M. Frankowski et al., *J. Appl. Phys.* **117**, 223908 (2015).

[5] H. J. Mao et al., *J. Appl. Phys.* **116**, 053703 (2014).

[6] M. Bowen et al., *Appl. Phys. Lett.* **82**, 233 (2003).

[7] M. Ye. Zhuravlev et al., *Appl. Phys. Lett.* **95**, 052902 (2009).

[8] J. Ruan et al., *Appl. Phys. Lett.* **107**, 232902 (2015).

[9] J.G. Connell et al., *Appl. Phys. Lett.* **101**, 251607 (2012).

[10] Z. Liao et al., *Phys. Rev B* **92**, 125123 (2015).

[11] Ĺ. Monsen et al., *J. Magn. Magn. Mater.* **369**, 197-204 (2014).

[12] H. Yamada et al., *Appl. Phys. Lett.* **86**, 192505 (2005).