Magnetic properties of SrRu$_{0.9}$Fe$_{0.1}$O$_3$ thin films grown on different surfaces of SrTiO$_3$ substrates

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Kirstie Raquel Natalia Toreh, Min Young Lee, Octolia Togibasa Tambunan, Deok Hyeon Kim, Bo Wha Lee, Chang Uk Jung*

Department of Physics, Hankuk University of Foreign Studies, Yongin 449-791, Republic of Korea

ABSTRACT

We stabilized SrRu$_{0.9}$Fe$_{0.1}$O$_3$ single-crystalline films on SrTiO$_3$ (001) and SrTiO$_3$ (110) substrates using epitaxial strain during thin-film growth. X-ray diffraction (XRD) θ—2θ scans showed strong peaks demonstrating single-crystal quality. Fe doping in SrRuO$_3$ had negative effects on the ferromagnetic properties, such as decreasing the T_c and saturated magnetic moment, as well as weakening the ferromagnetism. The negative effects were reduced when a suitable surface of the cubic substrate was selected for thin-film SrRuO$_3$ growth. We found that the ferromagnetic properties, such as the T_c and saturated magnetic moment, differed depending on the substrate surface. The observed differences are discussed in terms of Ru—Ru nearest-neighbor distance.

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1. Introduction

Polycrystalline SrRuO$_3$ (SRO) has an orthorhombic perovskite structure. It has a Curie temperature (T_c) of ~160 K with a robust low-spin configuration, having a saturated magnetic moment of ~1.6 μ_B per Ru^{4+} ion [1]. SRO has been used widely as an electrode in oxide heterostructures due to its high conductivity and good lattice match with most popular single-crystalline perovskite oxide substrates such as SrTiO$_3$ (STO).

Doping in polycrystalline SRO has been used to control magnetic properties such as the T_c and magnetic coercive field. However, doping with Fe at the Ru site in SRO bulk crystals was not successful, resulting in non-singular phase [2]. To overcome this problem, codoping with (Sr$_{1-x}$La$_x$)(Ru$_{1-x}$Fe$_x$)O$_3$ and (Sr,Fe)$_{1+y}$(Ru,Fe)$_{1+y}$O$_{3+d}$ was used for polycrystals [3,4]. Moreover, Fe doping has not been studied in thin films.

In a previous report, we showed that Fe-doped SRO can be stabilized by using epitaxial strain during film growth, and we also reported studies on the structural, electrical, and basic magnetization properties [5]. The high-resolution x-ray diffraction (HRXRD) θ—2θ scan showed clear Sr(Ru$_{1-x}$Fe$_x$)O$_3$ film peaks with

fringes. The calculated out-of-plane lattice constant of the films grown on the STO (001) substrate increased from 3.955 Å to 3.963 Å as doping increased from $x = 0.1$ to 0.3. Reciprocal-space mappings of the films on a log scale showed that all films grew coherently, with the same in-plane lattice constant as those of the underlying STO substrate [5]. The surface image acquired by atomic force microscopy (AFM) after growth showed a nice step-and-terrace structure with a root-mean-square roughness of less than 0.3 nm. Systematic doping with Fe 10%, 20%, 30% was clearly evident in the transport measurements. As doping increased, conductivity decreased, especially at low temperatures. In addition to layer-by-layer growth with atomically flat surfaces, high conductivity even at room temperature demonstrates that Sr(Ru$_{1-x}$Fe$_x$)O$_3$ ($x \leq 0.2$) is a good oxide electrode, with in-situ thickness control.

A geometric factor, $t = (r_A + r_o)/\sqrt{2}(r_B + r_o)$, in most manganese and nickelate perovskite oxides was believed to be the dominant factor in determining the structural, magnetic, and electrical properties [4,6,7]. Having t smaller than 1.0 results in a more distorted structure having a smaller bond angle B—O—B [8]. This factor depends mostly on the optimal radius of a sphere within a cage consisting of eight octahedra. Recently, structural modification effects on magnetostriction in SrTi$_{1-x}$Fe$_x$O$_3$ thin films on STO (001) and (110) substrates were explained in terms of this geometric factor [9].

* Corresponding author. Fax: +82 31 330 4566.
   E-mail addresses: cu-jung@hufs.ac.kr, cujung@gmail.com (C.U. Jung).

In this paper, we discuss the physical properties of $SrRu_{0.9}Fe_{0.1}O_3$ films on top of $SrTiO_3$ (001) and (110) substrates. The physical properties were found to be closely related to the nearest-neighbor distance of the B-site ion. The ferromagnetic properties, such as T_c and the saturated magnetic moment, were quite different depending on the substrate orientation. We found that the differences in the magnetic properties can be interpreted in terms of different structural deformations of the $SrRu_{0.9}Fe_{0.1}O_3$ unit cell. We use the Ru—Ru nearest-neighbor (Ru_{nn}) distance to explain the strain effects in perovskite oxide thin films grown on different orientations of the $SrTiO_3$ substrate.

2. Experimental

$SrRu_{0.9}Fe_{0.1}O_3$ thin films were grown on top of STO (001) and STO (110) substrates using pulsed laser deposition with a KrF excimer laser and stoichiometric targets [5,8,10—12]. For simplicity, we will use "*SRFO$_{001}$ film*" and "*SRFO$_{110}$ film*", respectively. The oxygen partial pressure and substrate temperature during deposition were fixed around 50—100 mTorr and 760 °C, respectively. The optimum growth temperature covered a wide range for SRFO, similar to SRO. The thicknesses of the films were ~70 nm. The STO (001) substrate was initially prepared using etching and heat treatment to create the step-and-terrace structures. Due to non-separation of Sr^{2+} and Ti^{4+} ions between the two termination layers of the substrate, the STO (110) substrate was treated by in-situ annealing just before the thin-film growth without an etching process. The crystal structure of the grown films was identified using HRXRD having CuKα radiation, and the magnetic properties of the films were obtained using a superconducting quantum interference device (MPMSXL, Quantum Design).

3. Results and discussion

Fig. 1(a) shows the HRXRD results for $SrRu_{0.9}F_{0.1}O_3$ films grown on an STO (001) substrate, i.e., *SRFO$_{001}$ film*. In the log-scale pattern,
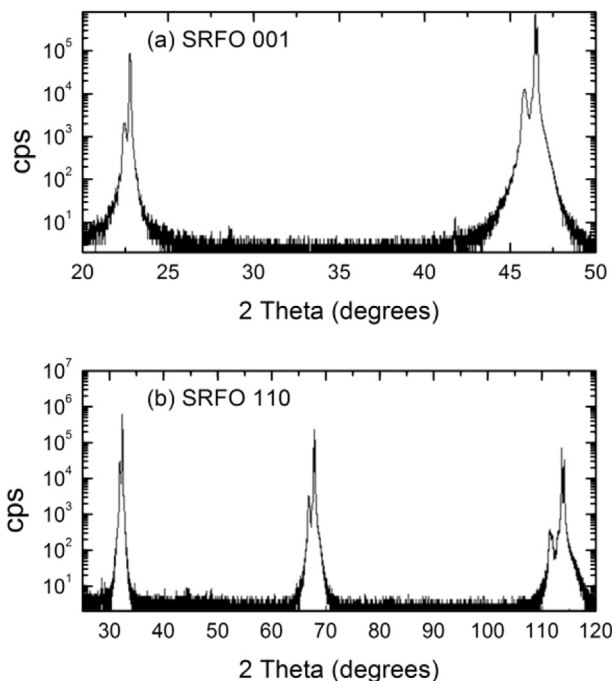

Fig. 1. X-ray $\theta-2\theta$ scan curves (a) for $SrRu_{1-x}Fe_xO_3$/STO (001) and (b) $SrRu_{0.9}Fe_{0.1}O_3$/STO (110).

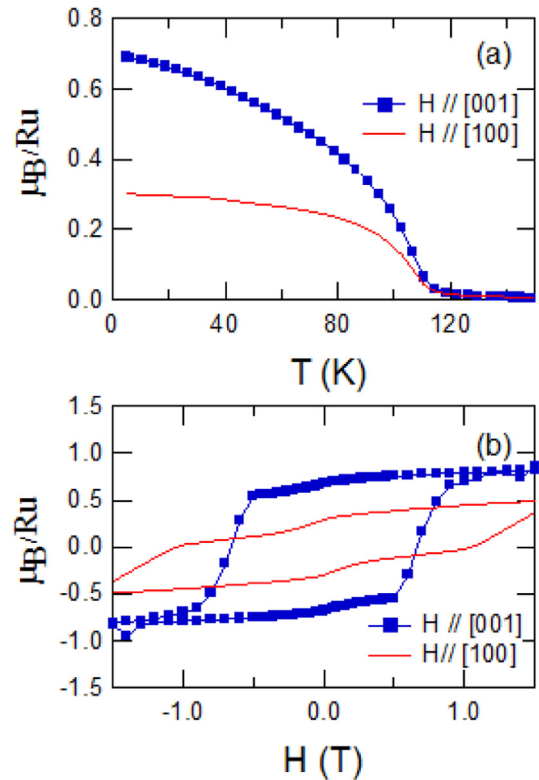**Fig. 2.** (a) Magnetization curves and (b) magnetic hysteresis curves for the $SrRu_{0.9}Fe_{0.1}O_3$/STO (001) substrate.

there was a strong SRO film peak with oscillation fringes showing uniform thickness on the left side of two large substrate peaks near $2\theta = 46.46°$ [the two strongest and well-separated substrate peaks corresponded to CuKα$_1$ and Kα$_2$ sources in the x-ray tube]. The calculated out-of-plane lattice constant was 3.955 Å. The x-ray $\theta-2\theta$ scan pattern in Fig. 1(b) shows that the $SrRu_{0.9}Fe_{0.1}O_3$ film grown on STO (110), i.e., *SRFO$_{110}$ film*, was of high quality. The separation of the SRFO film peak on the left side of the STO (330) peak is very clear. The out-of-plane lattice constant was $d_{220} = 1.398$ Å $= 3.954$ Å$/2\sqrt{2}$. Together with the coherent growth reported previously, the unit cell volume of both films are nearly the same [5].

We can estimate the oxygen vacancies using a simple linear calculation. The lattice constants of pseudocubic unit cells of SRO, $SrFeO_3$, and the oxygen-vacancy-ordered perovskite $SrFeO_{2.5}$ are 3.923 Å, 3.850 Å, and 3.940 Å, respectively [1,5,6]. So the pseudocubic unit cell volumes are 60.375, 57.067, and 61.163 Å^3, respectively. The linear sum of the unit cell volume of $(SRO)_{0.9}(SrFeO_3)_{0.036}(SrFeO_{2.5})_{0.064}$ gives 60.306 Å^3, which is very close to the observed unit cell volumes of $3.905 \times 3.905 \times 3.955$ Å^3 and $3.905 \times 3.905 \times 3.954$ Å^3. Thus, both films seemed to be slightly oxygen deficient, ~$SrRu_{0.9}Fe_{0.1}O_{2.97}$.

Fig. 2(a) shows the magnetization data of *SRFO$_{001}$ film* at a magnetic field of 500 Oe after high field-cooling. For consistency of comparison, the same specimen was used for the measurements by changing only the field direction with respect to the crystallographic axis for respective heterostructures. The data represented by solid rectangular symbols were acquired with the magnetic field applied along the surface normal direction, and data represented by solid lines were acquired with the field applied along the in-plane direction of each substrate. Compared to the magnetization of the undoped SRO film on the STO (001) substrate, the ferromagnetic transition temperature of *SRFO$_{001}$ film* decreased from ~150 K to