

Lattice distortion and uniaxial magnetic anisotropy in single domain epitaxial (110) films of SrRuO₃

Q. Gan, R. A. Rao, and C. B. Eom^{a)}

Department of Mechanical Engineering and Materials Science, Duke University, Durham, North Carolina 27708

L. Wu and F. Tsui

Department of Physics and Astronomy, University of North Carolina, Chapel Hill, North Carolina 27599

Single domain epitaxial (110) films of SrRuO₃ exhibit uniaxial magnetic anisotropy instead of the biaxial anisotropy observed in the bulk material. The magnetic easy axis for the film is along the orthorhombic [010] direction below T_C , and it rotates toward the [110] perpendicular direction as temperature decreases. The [100] direction, which is also magnetically ‘‘easy’’ in the bulk, becomes ‘‘hard’’ in the film. X-ray diffraction experiments show that this unique transformation of magnetic anisotropy is related to a distortion from the bulk orthorhombic lattice into a triclinic structure in the epitaxial film, such that the lattice along the [010] direction expands while its [100] counterpart contracts. The distortion appears to arise from rotation and tilt of RuO₆ octahedra. The finding indicates that the magnetic anisotropy in epitaxial SrRuO₃ films is rooted in the crystalline anisotropy influenced by strong spin-orbit interactions. © 1999 American Institute of Physics. [S0021-8979(99)75608-2]

I. INTRODUCTION

The colossal magnetoresistance (CMR) manganites and other ferromagnetic perovskites are potential materials for next generation magnetic devices, such as recording heads, memories, and sensors.¹ These applications require the synthesis of epitaxial thin films or multilayers. However, lattice mismatch and differential thermal expansion between the film and substrate can generate considerable strain in the film, and as a result the properties of thin film perovskites are quite different from those of the bulk.^{2,3} In epitaxial SrRuO₃ films, earlier studies have shown that strain can suppress the saturation magnetic moment by ~20% and T_C by a similar fraction.⁴ Therefore, the effects of strain on magnetic properties of epitaxial films, particularly magnetization, T_C , and anisotropy, must be thoroughly studied before device applications can be implemented. In this article, we report studies of magnetic anisotropy in single domain epitaxial (110) films of SrRuO₃ and its relationship with lattice distortions due to epitaxy.

SrRuO₃ is the only known itinerant ferromagnetic perovskite with a bulk T_C of ~160 K.⁵ It has an orthorhombic, GdFeO₃ type structure with lattice parameters $a_0=5.53$ Å, $b_0=5.57$ Å, and $c_0=7.85$ Å, and a slightly distorted pseudocubic perovskite unit cell of $a_0^p=b_0^p=c_0^p=3.93$ Å, $\alpha=\beta=90^\circ$ and $\gamma=89.6^\circ$. In this article Miller indices based on the orthorhombic unit cell are used. The crystallographic relationship between the orthorhombic and the pseudocubic structures is shown schematically in Fig. 1(a). The orthorhombic distortion arises from the tilting and rotating of the corner-sharing RuO₆ octahedra zig-zag chains, while RuO₆ octahedra are undistorted.⁶

Magnetic anisotropy in SrRuO₃ has been attributed to strong spin-orbital coupling. Studies of bulk single crystals have shown, that there are two magnetic easy axis along either the face diagonals of the pseudocubic unit cell,⁷ or orthorhombic [100] and [010] directions.^{7,8} Recently, Klein *et al.*⁹ reported that for single domain epitaxial (110) SrRuO₃ films, there is only a *single* easy axis that varies with temperature from T_C to 4.2 K by about 15°. However, they did not determine whether the easy axis is along the [100] or the [010] direction, and they offered no explanations on their assertion that the uniaxial anisotropy arises from strain effects. The focus of our study reported here is to resolve these issues.

II. EXPERIMENT

Epitaxial SrRuO₃ (110) films used in this study were grown on 2° miscut (001) SrTiO₃ substrates using 90° off-axis sputtering techniques. The sputtering conditions are described elsewhere.¹⁰ The thicknesses of the films are about 1000 Å. Scanning tunneling microscopy studies have shown that the surfaces of the SrRuO₃ films consist of atomically smooth terraces with nearly periodic ledges along the miscut, resulting from coherent single domain growth.¹¹ The small lattice mismatch with the substrate (~0.67% compressive) allows the coherent growth of SrRuO₃.¹⁰ Previous x-ray diffraction experiments¹⁰ indicate that the three respective symmetry directions are aligned with those of the substrate, i.e. perpendicular, and in-plane along and normal to the miscut direction, as shown in Fig. 1(b). For example, the [110] of the film is aligned with the [001] of the substrate, and the [$\bar{1}$ 10] aligned with [010], respectively. In this study x-ray diffraction experiments were carried out, in order to determine the detailed lattice distortions. A variety of scans were

^{a)}Electronic mail: eom@acpub.duke.edu

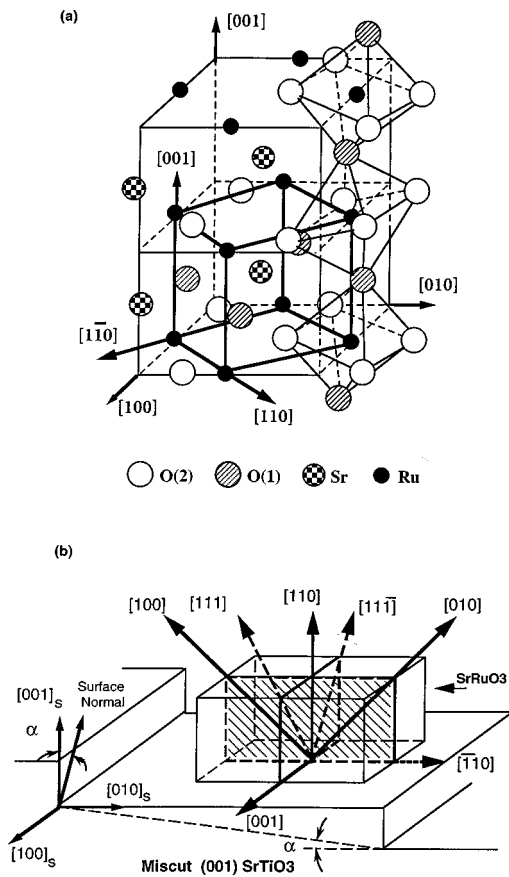


FIG. 1. (a) Schematic diagram of SrRuO₃ crystal structure in orthorhombic unit cell. The inner cube constructed by thick solid lines is the pseudocubic unit cell. (b) The epitaxial arrangement of single domain epitaxial SrRuO₃ (110) films on miscut (001) SrTiO₃ substrates. The arrows indicate the directions along which the magnetic measurements were performed.

performed using a four-circle diffractometer, including normal θ - 2θ scans, grazing incidence diffraction (GID) scans, and off-axis scans.

Magnetic properties of the SrRuO₃ (110) films were measured using a Quantum Design SQUID magnetometer. Special sample holders were made to position the sample in various orientations with respect to the applied field. Thus, orientation dependent magnetization measurements were made for the SrRuO₃ films, particularly along the seven symmetry directions, i.e. the three orthorhombic principle axes [100], [010], and [001], and the four pseudocubic face diagonals [110], $[\bar{1}10]$, [111], and $[\bar{1}\bar{1}\bar{1}]$, as indicated in Fig. 1(b).

III. RESULTS AND DISCUSSION

The epitaxial SrRuO₃ (110) films develop ferromagnetism below ~ 150 K, where they exhibit a *single* large uniaxial magnetic anisotropy. This is demonstrated by the field-cooled magnetizations along symmetry directions, as shown in Fig. 2. The large anisotropy, much larger than the shape anisotropy (or demagnetization factor), renders the measured low field magnetization essentially the zero field projection of the moment along the measuring axis. According to Fig. 2 with help from the diagram in Fig. 1(b), the moment is along the [010] direction just below T_C , indicated

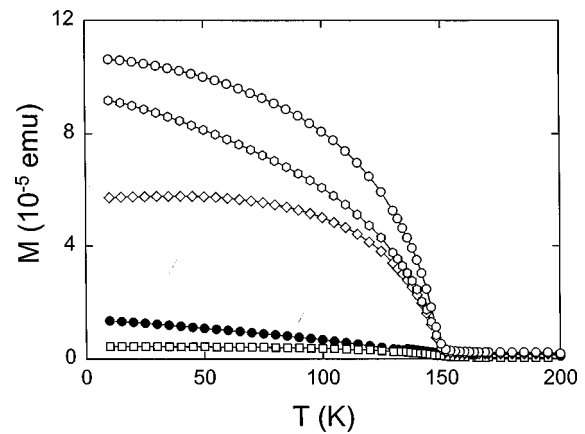


FIG. 2. Field-cooled magnetization of a single domain epitaxial (110) SrRuO₃ film for a 100 G field applied along [010] (open circles), [110] (hexagons), $[\bar{1}10]$ (diamonds), [100] (closed circles), and [001] (squares) directions.

by the equal projections along [110] and $[\bar{1}10]$, and small or no projections along [100] and [001]. At low temperatures, the moment rotates about 15° towards the [110] perpendicular direction. This is indicated by the increased projections along [110] and [100] and the corresponding decreased values along [010] and $[\bar{1}10]$, while the behavior along [001] remains relatively unchanged. However, for an ideally distorted SrRuO₃ along the [110] growth direction, as shown in Fig. 1, one would expect that [010] and [100] are still equivalent leading to two easy axes similar to the bulk behavior.⁸ Therefore, the observed single uniaxial anisotropy is quite unexpected.

In order to understand the observed magnetic anisotropy, it is critical to find out the detailed lattice distortions in the film. To enhance resolution, high index x-ray reflections were scanned, including (303), (600), (060), (111), (113), and (221) reflections. Three-dimensional lattice parameters and angles between symmetry directions were measured using normal, off-axis, and GID scans. From these measurements, crystal structure of the film is reconstructed. The resulting structure is pseudocubic and highly distorted, or more accurately triclinic. The measured lattice parameter along [010] is 5.585 ± 0.005 Å, while its [100] counterpart is 5.52 ± 0.01 Å. This result confirms unequivocally that the symmetry between the two axes is broken, and the observed uniaxial anisotropy is linked with the lattice distortion. Before discussing the effects on magnetic anisotropy further, the measured lattice parameters and bond angles are summarized as follows (also in Fig. 3). The out-of-plane $d_{(110)}$ is 3.953 ± 0.005 Å, and the in-plane $d_{(1\bar{1}0)}$ and $d_{(001)}$ are 3.90 ± 0.01 Å. The angle between $[\bar{1}\bar{1}0]$ and [001] is 90°, and that between [100] and [010] is 89.3°, and that between [110] and $[\bar{1}10]$ is 89.4°–89.5°.

The broken symmetry between the [100] and [010] directions appears to be the result of epitaxial growth. During coherent growth, the substrate compresses the lattice of the film along $[\bar{1}\bar{1}0]$ and [001] and hence expands along its [110] direction. This leads to a further distortion of the already distorted bulk lattice by tilting and rotating the RuO₆

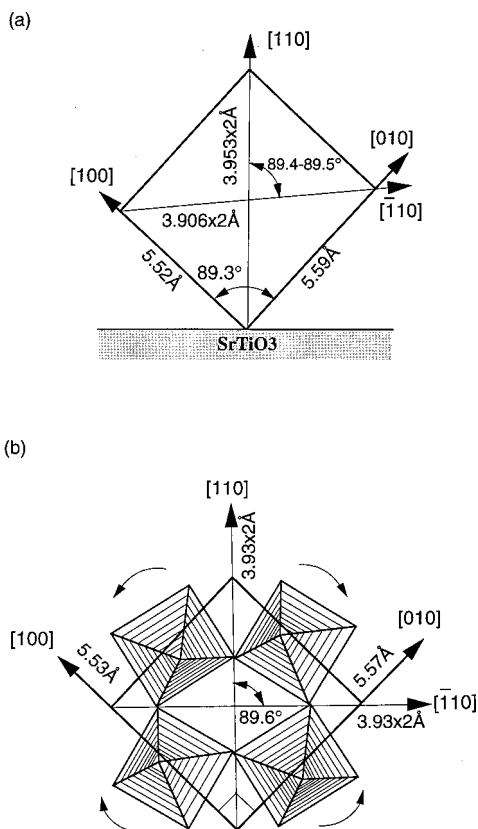


FIG. 3. Schematic diagrams of the orthorhombic unit cell in the (001) plane for (a) epitaxial SrRuO_3 (110) films and (b) bulk SrRuO_3 , in which the projections of the RuO_6 octahedra are also shown. The curved arrows in (b) indicate the rotating directions of the octahedra under epitaxial strain, i.e., tensile strain in [110] direction and biaxial compressive strain in [001] and $[\bar{1}\bar{1}0]$ directions.

octahedra, as it is schematically shown in Fig. 3(b), resulting in an elongated [010] and a shortened [100]. The presence of strong spin-orbit interactions coupled with a “uniaxial” crystalline anisotropy can evidently give rise to the observed uniaxial magnetic anisotropy.

IV. SUMMARY

We have studied the effects of epitaxial strain on magnetic anisotropy, particularly the observed uniaxial anisotropy, of single domain epitaxial SrRuO_3 (110) films grown on (001) SrTiO_3 substrates. We show that the unique magnetic easy axis is along or near the [010] direction. Our structural analysis of the film indicates that the epitaxial strain in the film has further distorted the lattice structure of SrRuO_3 from orthorhombic to triclinic. This leads to a shortening of lattice along [100] and lengthening along [010]. The observed strong crystalline anisotropy in SrRuO_3 appears to be responsible for the observed uniaxial magnetic anisotropy.

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