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Magnetic Properties of $SrRuO_3$ Epitaxial Thin Film Grown on $KTaO_3$ (001) Substrate

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We investigated the magnetic properties of $SrRuO_3$ epitaxial thin film synthesized by using pulsed laser deposition on $KTaO_3$ (001) substrate. X-ray reciprocal space mapping scans showed clear peaks demonstrating high quality, tensile strain with increased unitcell volume, and relaxed growth behavior. Magnetization data shows that the magnetization of the $SrRuO_3$ film with magnetic field along the in-plane direction was larger than one with magnetic field along surface-normal direction.

Keywords : epitaxial thin film, magnetic properties, $SrRuO_3$, $KTaO_3$

1. Introduction

Transition metal oxides with ABO_3 perovskite structure have been investigated extensively due to their various fascinating physical properties such as giant magneto-resistance behavior, resistance switching phenomena, and multiferroic functionality [1-3]. These oxides showed huge modification of its physical properties through strain engineering using epitaxial strain [4].

Bulk $SrRuO_3$ (SRO) is an orthorhombic perovskite oxide with a Curie temperature (*T*c) of ~160 K and a saturated magnetic moment of ~1.6 μ_B per Ru ion [5, 6]. Lattice constants of bulk SRO having orthorhombic structure are a = 5.567 (3.936×√2) Å, b = 5.5304 (3.9106 ×√2) Å, and c = 7.8446 (3.9223×2) Å. The effect of strain on the magnetic properties of SRO thin films have been widely studied on many substrates including $SrTiO_3$(STO), $DyScO_3$, and $GdScO_3$ substrates [6-11].

SRO is very interesting material due to its strong spin-orbit coupling and the *T*c vs strains has long been under intensive study with broad reader's attention. However, most studies focused on SRO on STO substrate where SRO film is under compress strain. One of the earliest reports on the effect of strain on the magnetic properties of SRO thin films reported the change of magnetization of SRO film grown on STO (001) substrate. By using life-off method, they showed that *T*c changed from 150 K to 160 K which is close to bulk value and concluded that the compressive strain results in the reduction of *T*c [7]. They also found that the magnetic easy axis returned from normal direction to in-plane direction after lift-off. C. U. Jung *et al*., found that the SRO film grown on STO (110) substrate has reduced *T*c ~150 K and surface normal magnetic easy axis [8].

The interest of SRO films under tensile strain comes from three categories. 1) The magnetization induced second-harmonic generation has proved to be a powerful tool for optically detecting the interface magnetism while it favors/requires the in-plane magnetic easy axis [12]. 2) Magnetic tunnel junction using SRO layer requires the in-plane magnetic easy axis. 3) SRO has served as a bottom electrode to study the dielectric properties of the ABO3 thin film and the effect of the tensile strain on the dielectric properties critically requires tensile strained SRO with enough thickness with tunable in -plane lattice constant. Half of the aim of this paper is highly related to this 3rd issue: SRO with enough thickness and tensile strain and its physical properties.

It is well known that the magnetic easy axis of SRO thin film can be along surface parallel direction when film is under tensile strain. Using novel buffer layer of $CaHfO_3$ and anisotropic in-plane of STO (110) substrate, CUJ showed that SRO can be given tensile strain along only one in-plane direction while the SRO along other in-

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plane direction and SRO along surface-normal direction is under compressive strain [8]. They demonstrated that 1) magnetic easy axis is along the tensile strained in-plane direction and 2) Tc is almost the same as that of bulk SRO. They also demonstrated similar trend using $CaHfO_3$ and miscut (001) STO substrate [9]. Here miscut of the substrate was necessary to remove twins of $CaHfO_3$ buffer layer due to its highly orthorhombic structure.

The lattice mismatch between SRO and $SrTiO_3$, $DyScO_3$, and $GdScO_3$ substrate is -0.64 %, 0.41 %, and 1.0 %, respectively [10, 13]. However, the study is not done extensively for SRO films under higher tensile strain. The lattice constant of $KTaO_3$ (KTO) substrate is as large as 3.989 Å and lattice mismatch between SRO and KTO is as large as 1.7 %. In this work we have successfully grown $SrRuO_3$ epitaxial thin film on $KTaO_3$ substrate and studied its structural and magnetic properties.

2. Experimental

$SrRuO_3$ epitaxial thin film on $KTaO_3$ (001) substrates was grown by using pulsed laser deposition technique using a KrF laser (35 mJ/cm^2, 4 Hz) with a wavelength λ = 248 nm. During the thin film growth, the substrate temperature was maintained at 750 ^{o}C with oxygen partial pressure ~50 mTorr. The film thickness was ~50 nm. Crystal structure was characterized by using a high-resolution X-ray diffractometer (HRXRD; Bruker D8). The magnetic properties were obtained using a superconducting quantum interference device (SQUID) magnetometer (MPMS, Quantum Design).

3. Results and Discussion

Figure 1 shows the XRD θ-2θ scan pattern of SRO epitaxial thin film on the KTO(001) substrate. While SRO (001) peak is marginally visible, the shoulder peak at the right side of KTO (002) peak assigned to be the SRO (002) film peak is more pronounced [We will use pseudo-cubic notation.]. But the reasonable estimation of the out-of-plane lattice constant of SRO film from this shoulder peak was not easy. So, we tried to measure x-ray reciprocal space mapping in order to get three lattice constants of the SRO film.

X-ray reciprocal space mapping has been widely used to study the lattice strain of the epitaxial thin films. The horizontal and vertical axis of the reciprocal space mapping is referred to the in-plane lattice constant and out of plane lattice constant, respectively. Figure 2 shows the reciprocal space mapping of SRO epitaxial thin film

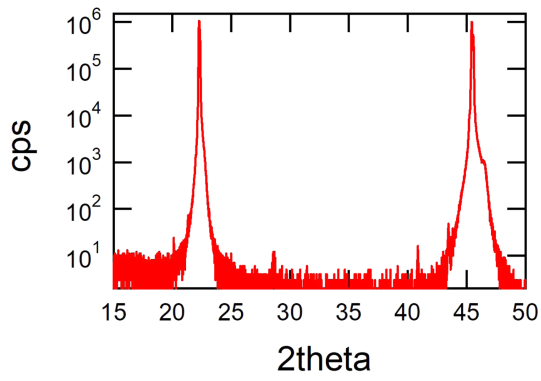

Fig. 1. (Color online) HRXRD θ-2θ scan patterns of epitaxial $SrRuO_3$ thin films on $KTaO_3$ (001) substrate.

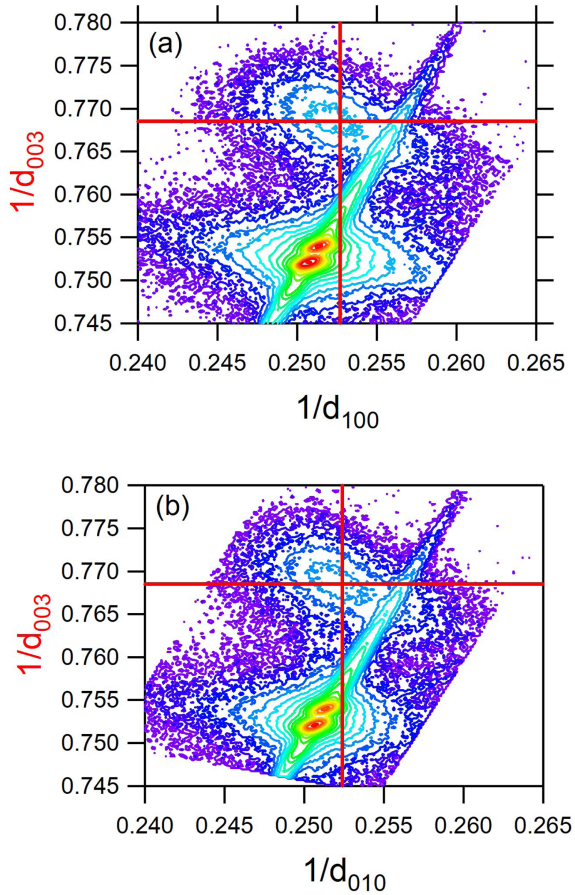

Fig. 2. (Color online) (a), (b) X-ray reciprocal space mapping around $KTaO_3$ (103) and (013) planes.

around (103) and (013) reflection of the KTO substrate. The clear separation of the very strong substrate peaks due to existence of two wavelength of x-ray source (Cu K-α_1 and K-α_2) demonstrates the very high accuracy of our measurement.

The measured reciprocal space mapping shows film

peak in upper right side relative to the substrate peaks. The horizontal position of the film peak is located at the right side of that of the substrate for both mapping data. So, the film showed relaxed growth behavior. This is due to quite large lattice mismatch (1.7 %) between the bulk SRO and the KTO substrate. 30 nm thick SRO film was reported to be grown coherently on top of $GdScO_3$ substrate where lattice mismatch is 1.0 % [7]. From the peaks in RSM data, we estimated the pseudo-cubic lattice constant of the SRO film: a = (3.965±4) Å, b = (3.968±4) Å, c = (3.902±4) Å and pseudo-cubic lattice constant is around (3.943±4) Å. Our mapping data showed that SRO lattice is elongated significantly along the surface parallel direction and unitcell volume of SRO was about 1.5 % larger than that of bulk SRO. Quite interestingly, the unitcell volume expansion of about 1.5 % for our 50 nm thick SRO film on the KTO substrate was almost the same as the reported vale of 1.8 % ($3.968\times3.966\times3.906$ $Å^3$ = 1.018×3.9231^3 $Å^3$) for 30 nm thick SRO film coherently grown on $GdScO_3$ substrate [10]. In the report, the oxidizing growth condition of SRO was the same for STO, $DyScO_3$, and $GdScO_3$ substrate and the unit-cell volume of SRO on STO was not lager than that of bulk SRO within 0.1 %; a = b = 3.905 Å and c = 3.964 Å. So, we can assume that volume expansion comes mostly from elastic strain not from oxygen vacancy.

Figure 3(a) shows the magnetization of the SRO film as a function of temperature over the range 5-200 K for two magnetic field directions. First, the ferromagnetic transition temperature of the SRO film, *T*c ~160 K was slightly larger than the reported values, *T*c ~150 K for SRO films on STO (001) substrates [6, 7]. The *T*c value of SRO film under tensile strain was mostly around 160 K which is the same as most results including our results [8, 9]. However, there is one contradictive report: the *T*c value of SRO film on $GdScO_3$ substrate was insisted to be around 144 K [13]. However, in the report, we need to look more carefully. First *T*c variation is not systematic between SRO/$DyScO_3$ and SRO/$GdScO_3$ in Fig. 4 of the report. The type of strain is the same: both tensile but the *T*c change is not systematic in Fig. 4 of the report. *T*c of SRO on STO substrate is more than 5 K smaller than most reported values of 150 K [6]. Finally, inset of Fig. 1(b) shows two step *T*c. The smaller portion of the STO/$GdScO_3$ sample has *T*c close to 160 K and the larger portion of the STO/$GdScO_3$ sample has *T*c close to ~144 K.

It is interesting that the SRO lattice deformation on the KTO substrate is rather similar to that on the $GdScO_3$ substrate; similar lattice expansion. More thorough study especially for SRO on $GdScO_3$ is required for the reasonable comparison since our data is consistent with the general trend of *T*c vs strain. It should be noted that that this trend is still valid for SRO on KTO substrate which has much larger lattice mismatch than $GdScO_3$ substrate.

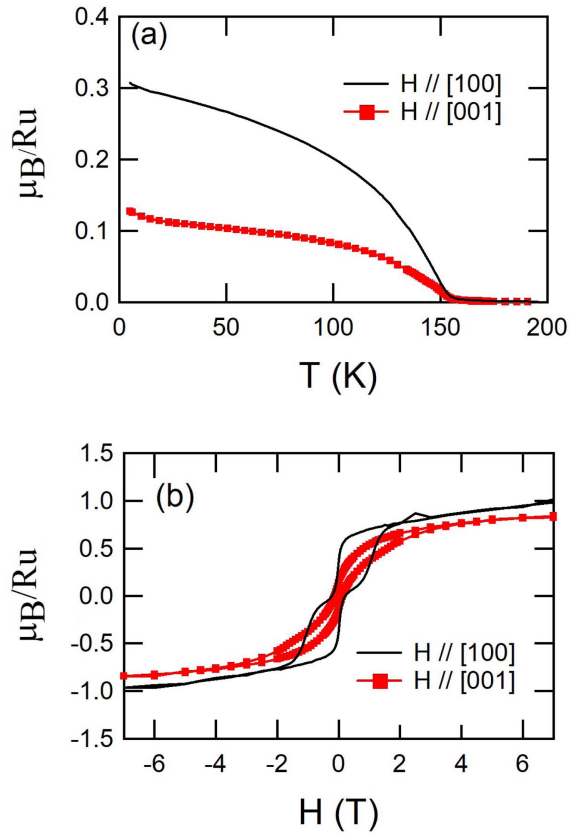

Fig. 3. (Color online) (a) Magnetization curve for $SrRuO_3$ thin films on $KTaO_3$ (001) substrate. (b) Magnetic hysteresis curve at T = 5 K for $SrRuO_3$ thin films on $KTaO_3$ (001) substrate. The films were cooled down to 5 K under the applied field of 9 T and the magnetization data were collected during warming after applying 500 Oe.

In Fig. 3(a), the magnetization for the SRO film with magnetic field along in-plane direction was found to be roughly 2.6 times larger than that with magnetic field along the surface normal direction. This is consistent with the general trend that magnetization along the elongated SRO lattice is higher than that along the compressed SRO lattice [8, 10].

Figure 3(b) shows magnetic hysteresis curve of the SRO film for two field directions. Here also, the magnetization for the SRO film with magnetic field along in-plane direction was found to be larger than that with magnetic field along the surface normal direction. Also, the coercive field value for the SRO film with magnetic field along in-plane direction was found to be smaller than that with magnetic field along the surface normal

direction. The existence of two steps, i.e. coercive field as shown in Fig. 3(b) has been observed for many ABO_3 single layer thin film including $SrRuO_3$ and $PrVO_3$ but its clear mechanism is not well known while surface layer was assumed to be one of the origins [14].

4. Conclusion

We investigated the magnetic properties of $SrRuO_3$ epitaxial thin film synthesized by using pulsed laser deposition on $KTaO_3$ (001) substrate. X-ray reciprocal space mapping scans demonstrated that film showed rather relaxed growth behavior, which is due to large lattice mismatch between the film and the substrate. The elongation of the lattice of the SRO film along the in-plane direction was consistent with the fact that the magnetization for the SRO film with magnetic field along in-plane direction was found to be larger than that with magnetic field along the surface normal direction.

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