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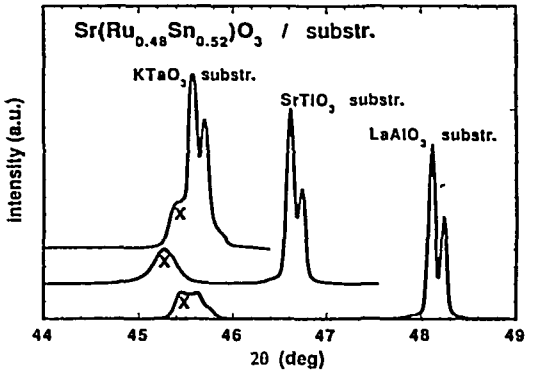

Figure 1. X-ray θ–2θ scans for 200 nm thick $Sr(Ru_{0.48}Sn_{0.52})O_3$ films on various substrates. $CuK\alpha_1$ and $K\alpha_2$ lines are visible. The substrate (001) reflections are labeled; those of $Sr(Ru_{0.48}Sn_{0.52})O_3$ are indicated by (x). The curves are normalized with respect to the substrate reflections.

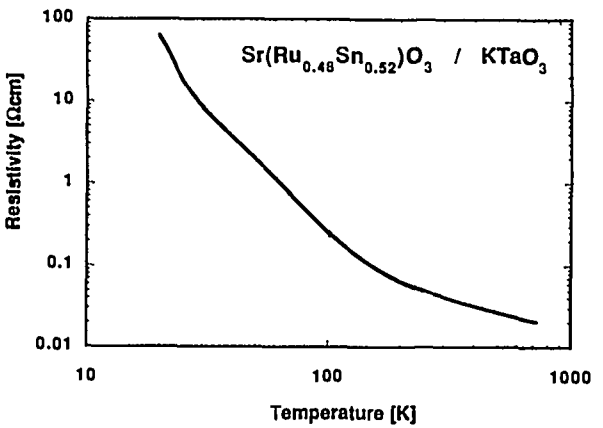

Figure 2. Resistivity as a function of temperature for a 400 nm thick $Sr(Ru_{0.48}Sn_{0.52})O_3$ film grown on (001)$KTaO_3$.

therefore, expected that a film grown on $KTaO_3$ will be fully "clamped" to the substrate. The in-plane lattice constant will thus be reduced to that of $KTaO_3$, forcing the out-of-plane spacing to expand. This is clearly seen in the data obtained for $Sr(Ru_{0.48}Sn_{0.52})O_3/KTaO_3$: the $Sr(Ru_{0.48}Sn_{0.52})O_3$ peak, largely overlapped by the $KTaO_3$. substrate reflection, is shifted to lower angles - corresponding to a larger lattice spacing. The present interpretation is further supported by the data for a film grown onto $SrTiO_3$. Here, the in-plane lattice constant is even smaller, forcing the out-of-plane spacing to expand even further.

Rutherford backscattering (RBS)/ion channeling techniques were used to analyze the $Sr(Ru_{0.48}Sn_{0.52})O_3$ films grown on $KTaO_3$. Within experimental accuracy, the data show that the composition of the film is identical to that of the target. However, the observed 15% channeling yield indicates that the film does not exhibit perfect crystallinity.

The resistivity of a 400 nm thick film was measured using a conventional four-probe technique in the temperature range from 20 to 700 K. The result is shown in Fig. 2. Contrary to the case of $SrRuO_3$, $dp/dT<0$ for $Sr(Ru_{0.48}Sn_{0.52})O_3$. The resistivity at room temperature (50 mΩ cm) is significantly increased from the value for $SrRuO_3$ (0.2 mΩ cm [4]), but it is sufficiently low for performing capacitance measurements on ferroelectric films. For a 1 nF capacitor, for example, the room-temperature resistivity of $Sr(Ru_{0.48}Sn_{0.52})O_3$ leads to a cut-off frequency of 1 MHz in a typical sample geometry. This cut-off is outside of the frequency window allowed by most sample holders, and, therefore, does not affect the accessible range.

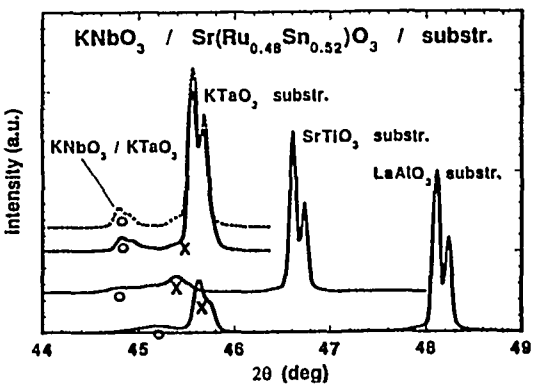

Figure 3. X-ray θ–2θ scans for $KNbO_3$ (150 nm) on $Sr(Ru_{0.48}Sn_{0.52})O_3$ (200 nm) bilayers on various substrates. $CuK\alpha_1$ and $K\alpha_2$ lines are visible, and the intensities are normalized with respect to the substrate peaks. $KNbO_3$ reflections are indicated by (o); $Sr(Ru_{0.48}Sn_{0.52})O_3$ peaks by (x).

Figure 3 shows X-ray θ–2θ scans for $KNbO_3/Sr(Ru_{0.48}Sn_{0.52})O_3$ bilayers grown on the same series of substrates. Also shown is the data for a $KNbO_3$-film grown on $KTaO_3$. As the comparison shows, the conducting "bottom" electrode does not force the $KNbO_3$ film to grow in a structure that is different from the one obtained in the case of the $KNbO_3/KTaO_3$ samples.

CONCLUSIONS

The present work shows that $Sr(Ru_xSn_{1-x})O_3$ can be used as a new conducting oxide in epitaxial multilayer-structures. $Sr(Ru_{0.48}Sn_{0.52})O_3$ exhibits an excellent lattice match with $KTaO_3$, and by varying the Ru:Sn ratio, the lattice constant can easily be tailored to match a wide range of materials with d-spacings larger than that of $SrRuO_3$.

DISCLAIMER

CONF-951155--68

PULSED LASER DEPOSITION OF EPITAXIAL $Sr(Ru_xSn_{1-x})O_3$ THIN FILM ELECTRODES AND $KNbO_3/Sr(Ru_xSn_{1-x})O_3$ BILAYERS

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ABSTRACT

$Sr(Ru_xSn_{1-x})O_3$ is proposed as a new conducting oxide for use in epitaxial multilayer structures. The $Sr(Ru_{0.48}Sn_{0.52})O_3$ composition exhibits an excellent lattice match with (100)-oriented $KTaO_3$, and films of this composition grown by pulsed laser deposition on $KTaO_3$, $SrTiO_3$, and $LaAlO_3$ substrates have been analyzed by X-ray diffraction, Rutherford backscattering/ion channeling, and resistivity measurements. Epitaxial $KNbO_3/Sr(Ru_{0.48}Sn_{0.52})O_3$ bilayers have been successfully grown.

INTRODUCTION

Due to their excellent electro-optic properties and high dielectric polarizabilites, epitaxially grown perovskite thin films are increasingly studied for applications in optical data transmission and in memory cells. These devices consist of an insulating layer sandwiched between two electrodes, of which the "bottom" electrode also acts as a substrate in the film-growth process. Typically, the bottom electrode is formed by either by a doped, semiconducting crystal or an epitaxial metal film. Niobium-doped $SrTiO_3$ has been used as an example of the former, but exposure of the substrate surface to an oxygen ambient, as required in the film growth of many materials, leads to the formation of an insulating layer at the substrate/film interface [1]. Platinum films have been used as an alternative, but ferroelectrics grown on these layers suffer from increased fatigue and high current leakage [2]. More recently, epitaxial films of conducting oxides such as RuO_2 [3] and $SrRuO_3$ [4], grown by pulsed laser deposition, have been used successfully as electrodes for $(Ba,Sr)TiO_3$ and PZT.
The lattice parameter of $SrRuO_3$ (c = 3.93 Å) is well below that of $KTaO_3$ (3.99 Å), $KNbO_3$ (c = 3.97 Å), and $KTa_{1-x}Nb_xO_3$. The latter two materials are currently being studied due to their excellent electro-optical properties [5]. Renewed interest in $KTaO_3$ has recently arisen due to the use of this material as a substrate for high-Tc films [6]. Most recently, it has become possible to grow superlattices consisting of alternating paraelectric $KTaO_3$ and ferroelectric $KNbO_3$ layers on (100)-oriented $KTaO_3$ single crystals [7]. These structures are expected to yield information about size-effects in ferroelectrics and - by comparison to the solid solution $KTa_{1-x}Nb_xO_3$ - about the difference between the random and layered introduction of ferroelectric "units" ($KNbO_3$) in a paraelectric medium ($KTaO_3$). It has been shown [7] that thin $KNbO_3$ films grown on $KTaO_3$ exhibit a different type of orthorhombic structure than that of the bulk. The influence of the substrate is thus evident, and $KNbO_3$ films grown on $SrRuO_3$ may behave differently from those grown on $KTaO_3$.

In order to investigate the ferroelectric polarization in $KTaO_3/KNbO_3$ superlattices, the structures need to be sandwiched between lattice-matched conducting films. In the present work we have developed a conducting oxide to be used in this situation by appropriately doping $SrRuO_3$. Substitution of Sr by the relatively smaller Ca ion results in a perovskite with a smaller lattice parameter. An analogous substitution by the larger Ba, however, doesn't lead to an expanded unit cell; instead, our results show clear signs of phase separation that is likely due to the rigidity of the RuO_6 octahedra. This problem can be solved by ionic substitution on the Ru-site instead of the Sr-site. $SrSnO_3$ has a double perovskite structure with a volume per unit-cell of (4.03 Å)^3 which is larger than that of $KTaO_3$. It is, therefore, to be expected that a solid solution can be found that is lattice matched to $KTaO_3$.

GROWTH OF $Sr(Ru_xSn_{1-x})O_3$ FILMS

For the preparation of $Sr(Ru_xSn_{1-x})O_3$ targets, $SrCO_3$ and SnO powders were first wet-milled, dried, and mixed with 20-mesh Ru sponge. The hand-ground powder was then calcined for 12 hours at 1200°C, and wet-milled to obtain a grain size below 1 µm. To obtain the desired laser-ablation targets, pellets were pressed at 60 MPa and sintered in air at 1350°C for 15 hours. A Ru:Sn concentration ratio of 48:52 was chosen - based on the simplified assumption that the unit cell volume of the solid solution $Sr(Ru_xSn_{1-x})O_3$ varies linearly with x, and in order to obtain a close lattice match with $KTaO_3$.

Film growth was carried out using a KrF eximer laser (248 nm, 38 nm FWHM pulse duration) with a focused energy density of 1.5 to 3 J/cm^2. The pulse repetition rate was 3.3 Hz, and the films were grown in 100 mTorr of oxygen.
The $KTaO_3$ substrates used in this study were cut along the cubic (100) axes of crystals grown by spontaneous nucleation from a flux containing an excess of K_2O. A KOH-buffered solution of colloidal silica (pH=11) was used to polish the 1 mm thick wafers. For comparison purposes, commercially available $SrTiO_3$ and $LaAlO_3$ substrates were also used.

RESULTS AND DISCUSSION

The $Sr(Ru_xSn_{1-x})O_3$ targets were first analyzed by X-ray powder diffraction. Contrary to the two cubic end-members, $SrRuO_3$ and $SrSnO_3$, the solid solution $Sr(Ru_{0.48}Sn_{0.52})O_3$ exhibits a non-cubic distorted perovskite structure. This is observed as a 1.5% splitting of the peak corresponding to the pseudo-cubic (011) reflection. The $KTaO_3$ (011) peak falls between these two pseudo-cubic lines. Figure 1 shows X-ray θ–2θ scans for 200 nm thick $Sr(Ru_{0.48}Sn_{0.52})O_3$ films on various substrates. Note that in this low-resolution scan, both Cu-$K\alpha_1$ and $K\alpha_2$ lines are visible; a double peak therefore corresponds to a single set of lattice planes.

The data in Fig. 1 are interpreted in the following way: Due to the relatively large lattice mismatch between $Sr(Ru_{0.48}Sn_{0.52})O_3$ and $LaAlO_3$, $Sr(Ru_{0.48}Sn_{0.52})O_3$ grows in a fully relaxed structure on this substrate. The film peak around 45.6° is clearly split, indicating that $Sr(Ru_{0.48}Sn_{0.52})O_3$ does not grow with a single orientation. The "average" lattice constant is seen to be only slightly larger than that of the $KTaO_3$ substrate. It is,