In-plane Dielectric Properties of Epitaxial Orthorhombic HoMnO$_3$ Films
Grown on LaAlO$_3$ Substrates

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(Received 27 December 2012, in final form 6 August 2013)

Orthorhombic HoMnO$_3$ thin films were grown epitaxially on LaAlO$_3$ (001) substrates by using a pulsed laser deposition technique. The films showed perfectly orthorhombic crystallization and were well-aligned with the substrates. The in-plane dielectric constant and the dielectric loss of HoMnO$_3$ films were measured as functions of temperature (80 ∼ 300 K) and frequency (120 ∼ 100 kHz) by using interdigital surface electrodes. Two thermally-activated dielectric relaxations were found, and the respective peaks shifted to higher temperatures as the measuring frequency was increased. The in-plane dielectric properties of epitaxial orthorhombic HoMnO$_3$ films were considered to have a universal dielectric response behavior, and the dipolar effects and the hopping conductivity induced by the charge carriers were used to explain the results. The dielectric properties at low temperatures, which showed the multiferroicity, of the orthorhombic HoMnO$_3$ films are discussed.

PACS numbers: 81.15.Fg, 77.22.Gm, 77.55.+f

Keywords: Dielectric properties, Interdigital electrodes, HoMnO$_3$ thin films, Orthorhombic

DOI: 10.3938/jkps.63.1799

I. INTRODUCTION

The stoichiometric rare-earth manganites have attracted intensive theoretical and experimental studies for a long time because of their various exotic properties, including ferroelectric behaviors, ferromagnetism, colossal magnetoresistance effects and multiferroics [1–4]. Many multifunctional designed devices made of these materials have been reported to exhibit fascinating dielectric, magnetic, and electric tunable behaviors. Among these rare materials, HoMnO$_3$ (HMO) is a prototypical material and shows a unusual interplay of magnetic and electric properties. Most research has focused on the antiferromagnetic and ferroelectric properties of HMO bulk or single crystals at low temperatures, and large magneto-electric effects have been reported [5–7].

Generally, HMO forms a stable hexagonal crystal structure. At the same time, orthorhombic HMO (o-HMO) can also be prepared by means of special synthesis methods although it is not the thermodynamically stable phase under ambient conditions [8–10]. In fact, HMO thin films can be fabricated in hexagonal or orthorhombic structure expediently by choosing suitable substrates. Compared with bulk materials, HMO thin films exhibit more interesting behaviors during certain physical processes. Moreover, the preparation of high-quality films is an indispensable requirement for modern practical applications [11,12].

In this paper, we report the growth of epitaxial o-HMO films on LaAlO$_3$ (LAO) substrates by using a pulsed laser deposition technique. The in-plane complex dielectric properties were measured as functions of frequency (100 Hz ∼ 100 kHz) with interdigital electrodes (IDEs) to conduct a comprehensive study of dielectric properties of their surfaces. The universal dielectric response (UDR) models were used to explain the observed results. The dielectric properties at temperatures below the multiferroic transition temperature are provided to show the multiferroicity of the films.

II. EXPERIMENTAL DETAILS

The thin films were deposited using a KeF excimer laser (λ = 248 nm, τ = 17 ns, 3-Hz repetition rate) focused onto a high-purity ceramic target of HMO which had been prepared through a conventional solid-state reaction process using high-quality Ho$_2$O$_3$ and Mn$_2$O$_3$ raw materials. The X-ray diffraction (XRD) profile of the HMO ceramic target exhibited a well-crystallized hexagonal structure (a = 0.613 nm, c = 1.14 nm) and no impurity phases could be detected in the X-ray spectra. The typical laser energy density used was close to 2.5 J/cm$^2$, high enough to ablate the ceramic HMO. The target was mounted on a rotating holder, 40 mm from the pseudocubic LAO (001) substrates (a = 0.3792 nm, 0.5 mm in thickness), which were maintained at 700 °C under an oxygen pressure of 30 Pa during the deposition process.
After the deposition, the thin films were cooled in oxygen at atmospheric pressure at a cooling rate of 5 °C/minute. The thicknesses of the fabricated HMO thin films were about 120 nm.

The crystalline structures of the deposited HMO films were investigated by using XRD with Cu K$_\alpha$ radiation at 1.54 Å. The low-frequency (120 ~ 100 kHz) surface dielectric measurements were performed with interdigital electrodes (IDEs) by using a QuadTech1730 LCR Digibridge system in a cooling run from room temperature to 77 K, followed by a heating run to room temperature. The data of both runs were reproducible and reversible. The low-temperature dielectric properties and multiferroic effects were measured as functions of the frequency and the temperature by using a superconducting quantum interference device magnetometer.

**III. RESULTS AND DISCUSSION**

Figure 1 shows a typical θ-2θ XRD pattern of an as-prepared HMO film on a LAO (001) substrate. Besides the diffraction peaks from the substrate, only sharp (110) and (220) peaks for the HMO thin film are observed, indicating that the as-prepared HMO thin film has an orthorhombic structure and is preferably (110) oriented. To investigate the in-plane texture of the films, we performed X-ray ϕ scans around the o-HMO (112) reflections. As shown in the inset of Fig. 1, fourfold symmetry peaks are located at ϕ intervals of 45° and 90° for the films, suggesting that the o-HMO films are indeed epitaxial and well aligned with the substrate. At room temperature, the lattice parameters of the bulk o-HMO phase are $a = 5.255$, $b = 5.826$, and $c = 7.365$ Å [13], whereas two times of the lattice constants along the [010] and the [100] directions of the LAO substrate are 7.584 Å × 7.584 Å, which is quite suitable to accommodate the (110) plane of o-HMO. The in-plane epitaxial relationship between the o-HMO and the LAO (100) substrate is thought to be o-HMO [110] // LAO[010] and o-HMO [001] // LAO [100] or o-HMO [110] // LAO [100] and o-HMO [001] // LAO[010]. In either case, the mismatches along the [110] and the [001] directions of the HMO thin films are 3.4% and –2.8%, respectively.

The characterization of the dielectric properties of the HMO thin films is very important for a variety of potential electronic designs. However, for many applications, the in-plane electrical properties rather than the through-plane properties of the film need to be characterized [14–16]. For this purpose, the IDE structure is widely used to electrically achieve the in-plane dielectric properties [17–19]. An IDEs were deposited on the top surface of the HMO film by using a combination of magnetron sputtering and lift-off photolithography. Figure 2(a) presents a scanning electron microscopy (SEM) image of the fabricated IDE pattern, and a schematic of the measurement is shown in Fig. 2(b). The top Au electrodes were about 0.1-μm thick. The IDE patterns consisted of 50 fingers with identical finger widths (15 μm) and spacings (20 μm); the fingers were 0.7-mm long. The dielectric constant of a thin film with an IDE