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Synthesis and Characterization of Highly Oriented Sol-Gel (Pb, La)TiO_3 Thin Film Optical Waveguides

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Abstract. The preparation and the optical characteristics of highly oriented PLT thin films are investigated. PLT films fabricated on MgO(100) and *c*-plane sapphire substrates have highly grown in (100) and (111) orientations, respectively. PLT films with high La content have a near cubic structure and weak anisotropy of refractive indices. The optical propagation losses of PLT films decrease as the La content of the films increases due to a complex interaction of surface roughness reduction and a reduction in the anisotropy refractive index. However, optical scattering in thicker sol-gel PLT thin film waveguides occurs by the internal scattering mechanism from the defects and the interfaces rather than by the surface scattering mechanism.

Keywords: sol-gel, lead lanthanum titanate, thin film, optical waveguide, optical propagation loss

1. Introduction

Much attention has been paid to lanthanum-modified lead zirconate titanate (PLZT) solid solution systems which are well-known ferroelectric materials for various applications such as high dielectric, pyroelectric, piezoelectric, and electro-optic devices, [1]. Since PLZT ceramics are transparent in the visible and near-infrared regions, and have superior electro-optic characteristics, thin films of this material are especially attractive for the application to integrated optical devices such as optical switches and optical waveguide modulators [1, 2].

Recently, sol-gel processing of PLZT thin films has been reported as giving advantages such as precise composition control and homogeneity, low temperature synthesis, large-area deposition, low cost, and short fabrication times [3–5].

In this paper, highly oriented PLT thin films are prepared by sol-gel processing and their optical waveguiding characteristics such as refractive index and optical propagation loss are investigated. The reason for studying compositions without zirconium is that PLT is known to be better for optical applications due to its finer grain size and higher transparency than PLZT [3].

2. Experimental Procedure

PLT films with the general chemical formula, $Pb_{1-x}La_xTi_{1-x/400}O_3$ (PLTx) where $x = 5$, 10, 15, 20 and 28 mol%, were prepared. A precise procedure for preparing the precursor solution is presented elsewhere [4, 5]. Lead acetate, lanthanum nitrate, and titanium isopropoxide were used as precursors. 5 mol% excess Pb was incorporated to compensate for PbO loss. 2-Methoxyethanol was used as a solvent. The substrates used in this study were MgO(100) and *c*-plane sapphire single crystals. The green films, which were spin-coated, were dried at 400°C on a hot plate for 10 min, yielding about 500 Å thickness per coating. The films were heat-treated with 5°C/min to 700°C holding for 30 min to crystallize them as a single perovskite phase. All the films were cooled at 2°C/min in air to prevent the microcracks from thermal shock during cooling.

The crystalline phase and the orientation of the PLT films were examined using X-ray diffraction. The microstructure and the RMS surface roughness of the films were observed using atomic force microscopy (Park Scientific Instruments, Autoprobe 5M). In order to measure the refractive indices and the optical propagation losses of the films, a prism coupler with a 632.8 nm wavelength was used [5].

3. Results and Discussion

3.1. Preparation of PLT Thin Films

XRD patterns of a PLT films (≈3500 Å) with a range of La contents, fabricated on MgO(100) single crystal substrates are shown in Fig. 1(a). All the films contain a single perovskite phase without any other noticeable phases, and are highly *a*-axis oriented with small extra peaks of (110) and (111). When the La content is low, both (100) and (001) peaks are observed. Only the (100) peak is detected when the La content increases to above 20 mol% due to the change of the perovskite structure from tetragonal structure to cubic structure [6]. XRD patterns of PLT films, with a range of La contents, fabricated on *c*-plane sapphire substrates are shown in Fig. 1(b). The films are highly oriented on the *c*-plane sapphire substrate, and the main (111) peak and small extra peaks are observed. Since the structure of the film changes from the tetragonal structure to the cubic structure as La content increases, the misfit between the film and the substrate decreases [7]. Therefore, the intensity of main (111) peak increases compared with other peaks, and the peak shifts to a higher 2θ angle and approaches the (0001) peak of the sapphire substrate as the La content increases.

3.2. Characteristics of PLT Thin Film Waveguides

The refractive indices n_o and n_e of the PLT films on MgO(100) substrates, measured using the prism coupler, are shown in Fig. 2. In the tetragonal perovskite PLT films (5 mol% La to 20 mol% La), the extraordinary refractive index, n_e, decreases significantly as La content increases, while the ordinary refractive index, n_o, remains at an almost constant level. However, the refractive indices n_o and n_e of PLT28 are almost the same since the film has a cubic structure. Thus, it is suggested that the anisotropy of the refractive indices decreas with increasing La content.

The optical propagation losses of the PLT films (≈2500 Å) on MgO and sapphire substrates are plotted in Fig. 3. The optical propagation losses of the films decrease with increasing La content for films on both the MgO and sapphire substrates.

Generally, optical propagation loss in the transparent thin film waveguides is considered to occur by scattering, and this scattering mechanism can be divided into surface scattering and non-surface scattering [8]. The non-surface scattering is affected by the defects

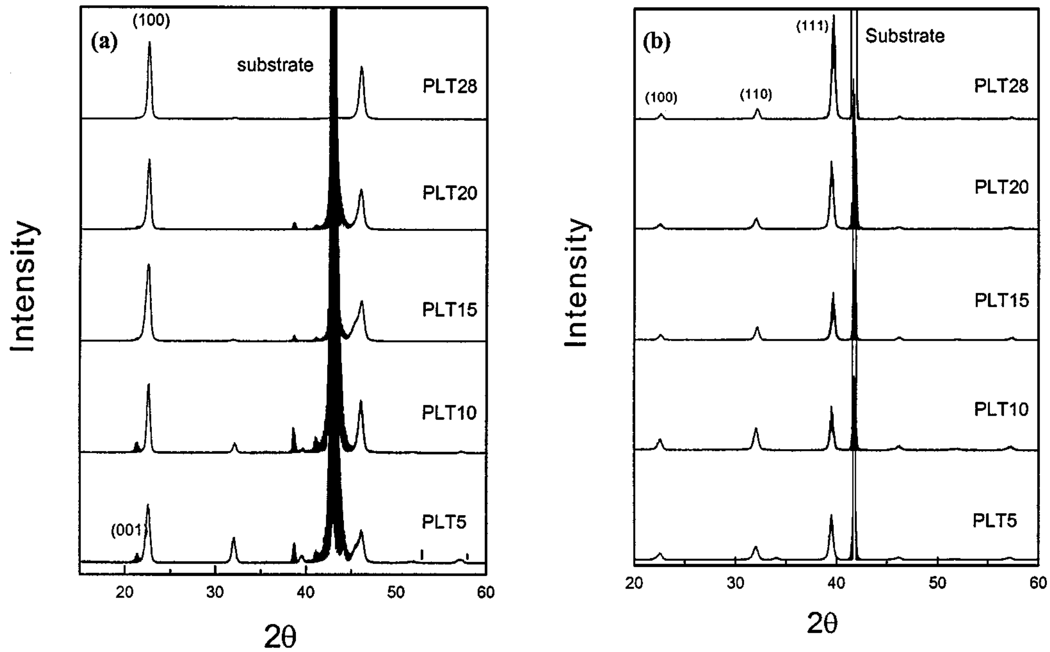

Figure 1. XRD patterns of PLT films on (a) MgO(100) substrates and (b) *c*-plane sapphire substrates as a function of La content.

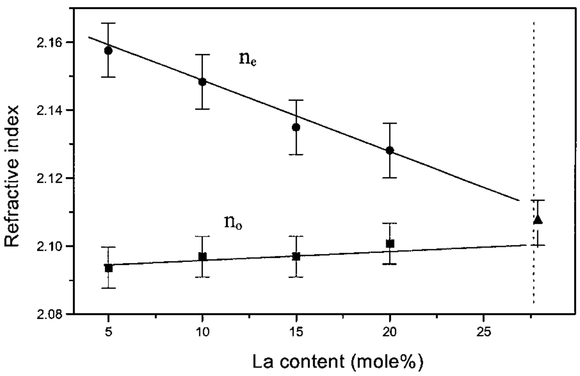

Figure 2. Refractive indices n_e and n_o of PLT films on MgO substrates as a function of La content.

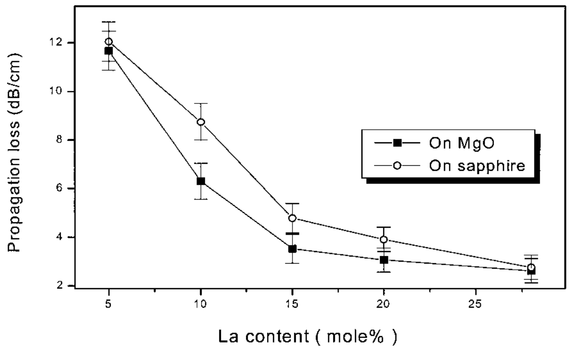

Figure 3. Propagation losses of PLT films fabricated on MgO and sapphire substrates as a function of La content.

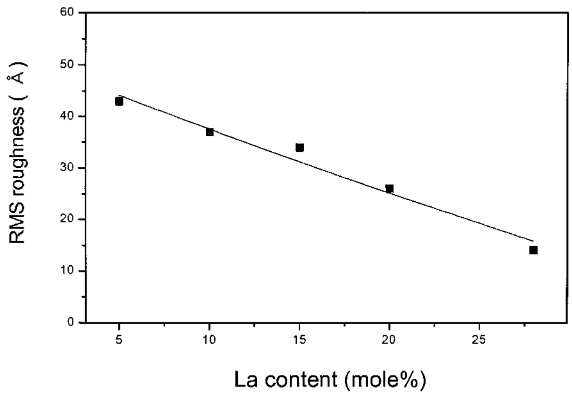

Figure 4. RMS surface roughness of PLT films fabricated on MgO substrates as a function of La content.

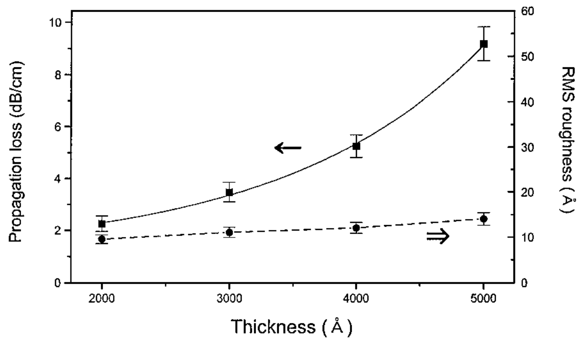

Figure 5. Propagation losses and RMS surface roughness of PLT28 films on MgO substrates as a function of film thickness.

or the anisotropy of refractive indices in the film. The main cause of the surface scattering is known to be surface roughness. AFM shows that the RMS surface roughness of the PLT films decreases linearly with increasing La content of the films as shown in Fig. 4. Since the grain size of the film becomes smaller as the La content increases, the surface roughness of the films is reduced. Therefore, the optical propagation loss decreases with the increase of La content due to a complex effect involving reduction of both non-surface scattering arising from the anisotropy of the refractive indices, and surface scattering due to surface roughness.

To ascertain the main scattering mechanism in the sol-gel PLT films, the optical propagation losses of the films were measured as a function of the film thickness (Fig. 5). The film composition of PLT28 was chosen since it has the lowest optical propagation loss because of its fine grain size and isotropic refractive index. The optical propagation losses increase exponentially with film thickness although the RMS surface roughness of the films is not greatly affected by the film thickness. Since thick sol-gel films are generally obtained by multiple coating, the presence of interfacial layers between coatings and/or the defects such as the pores organic and nitrate residues in the films enhances the optical scattering. Therefore, internal scattering by unavoidable defects and interfacial scattering in multiple coatings should be reduced to achieve lower optical propagation loss in the sol-gel PLT thin film waveguides with thicknesses greater than 4000 Å.

4. Conclusions

Highly oriented PLT thin films, which exhibit good optical properties, have been prepared on MgO and sapphire substrates by sol-gel processing. X-ray diffraction analyses indicate that the PLT films fabricated on MgO(100) and *c*-plane sapphire substrates grow preferentially in the (100) and (111) orientations, respectively. The refractive index and the optical propagation losses are affected by the film composition. The optical propagation losses of PLT films decreases with La content since the anisotropy of the refractive indices and the surface roughness of the films are reduced as the La content in the film increases. Thicker films obtained by multiple coatings have higher optical propagation loss regardless, although the film surface roughness is nearly constant. Thus, the main scattering mechanisms in thicker sol-gel PLT films are internal and interfacial scattering rather than surface scattering.

Acknowledgments

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