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Epitaxial growth of optical waveguiding $LiTaO_3$ films by excimer laser ablation

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Abstract. Waveguiding epitaxial $LiTaO_3$ films have been prepared on sapphire substrates by the excimer laser ablation technique. The as-grown films were characterized by Raman scattering, XRD and SEM techniques, which revealed that epitaxial (001)-oriented and (012)-oriented $LiTaO_3$ films with small surface roughness were achieved on sapphire (001) and (012) substrates, respectively. Optical waveguiding properties were demonstrated by *m*-line measurement of TM and TE multimodes.

$LiTaO_3$ (LT) is one of the most important materials for the fabrication of electro-optic devices and exhibits interesting pyroelectric, piezoelectric and non-linear optical properties. Recently, particular attention has been paid to the fabrication of integrated-optic devices in which non-linear optical films show many advantages over bulk crystals, and incorporation of the optical thin films into semiconductor integrated circuits [1]. At the same time, great advances have been made in using LT and $LiNbO_3$ (LN) thin films in waveguiding second-harmonic generation applications, because the large difference between the refractive indices of the thin films and the commonly used substrate, such as sapphire and MgO, will confine the propagating light in the films with a tight mode [2–7]. Therefore, preparation of epitaxial LT thin films becomes very attractive.

In the past few years, a series of techniques for the preparation of LT and LN thin films have been developed, including RF sputtering, ion plating, MBE, LPE and recently laser ablation [2–8]. Of these techniques, excimer laser ablation shows some advantages over the others; for example it is possible to prepare epitaxial films, a lower substrate temperature is required, a higher compositional consistency between the films and target is obtained, the process is highly efficient and there is no damage on the thermal source. It was reported recently that both LT and LN epitaxial films on *c*-oriented sapphire had been prepared by this technique [2, 8]. In this paper, the preparation of epitaxial LT films on both (012) and (001) *sapphire* substrates by laser ablation will be reported. The x-ray diffraction (XRD) technique and, for the first time, Raman spectroscopy will be applied to characterize the crystallinity of the as-grown films. Optical interferometry and scanning electron microscopy (SEM) will be applied to characterize the thickness uniformity and surface roughness of the as-grown films. The waveguiding TM and TE multimodes in the films will also be measured.

Laser ablation was performed using KrF excimer pulsed radiation of 248 nm wavelength, 30 ns pulse width and 5 Hz frequency. A schematic drawing of the system used in this work is given in figure 1. The laser beam was focused approximately on a rotating target by a quartz lens. The stoichiometric disc-shaped LT single-crystal target was mounted at an angle of 45° to the laser beam and the substrate surface. The distance l between the substrate

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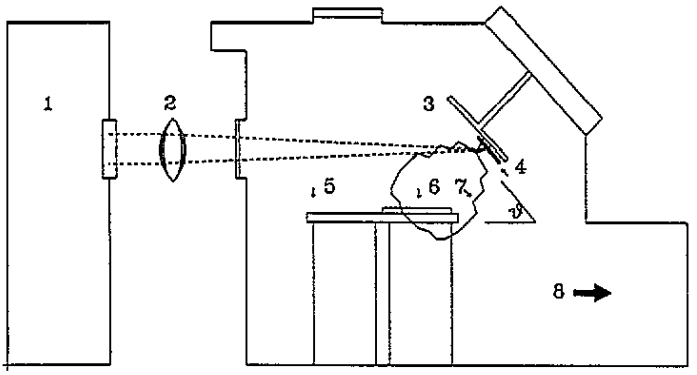

Figure 1. A schematic drawing of the laser ablation system used in this work. $\vartheta = 45°$. 1, LPX205i KrF excimer laser; 2, quartz focusing lens; 3, stage for mounting target; 4, target; 5, substrate stage; 6, substrate; 7, plume-like plasma spot; 8, to vacuum pump.

and target was 3.5 cm. The target was rotated at a speed of 0.1 rad s^{-1} in order to avoid long-time ablation of one spot of the target by the laser beam. In a reactive O_2 ambient the plume-like plasma spot can be observed while the laser beam interacts with the target. In the present case, an optimum O_2 ambient pressure $P_g = 13$ Pa was determined. As is well known, one of the major problems associated with the laser ablation preparation of films is the non-uniformity of the thickness and surface morphology of the films, which is critical for integrated-optic applications. Our measurements by optical interferometry revealed that only in the part of the substrate surface immersed in the plasma plume can uniform-thickness films be obtained; therefore, a rotatable substrate stage was designed. During deposition, the substrate was rotated at a speed of 0.2 rad s^{-1} in order to immerse each part of the substrate surface in the plasma plume with the same probability. To reduce the surface roughness of the films, the substrate temperature and energy fluence of the excimer radiation were optimized to 700 °C and 1.2 J cm^{-2}, respectively. The surface morphology of the films was characterized by SEM. With the present geometry, a uniform film of even larger than 2.0 cm^2 on sapphire (001) was successfully prepared, although most of the substrates used here are sapphire (012) and (001) of dimensions $5 \times 10 \times 0.5$ mm. After deposition, the samples were kept in the chamber at 700 °C and an O_2 ambient pressure of 400 Torr for 1 h to avoid oxygen deficiency.

The Raman spectra of the films were obtained with a Spex 1403 Raman spectrometer with back-scattering geometry. An Ar^+ laser with a wavelength of 488 nm and an output of 200 mW was used. The slit width is 200 μm. Details of similar Raman measurements can be found in [9 10].

The XRD spectra of the as-grown films on (001) and (012) substrates are shown in figures 2 and 3, respectively. In figure 2 the presence of only (006) and (00.12) lines of the LT film and sapphire substrate reveals that all the film is *c* oriented. Similarly, in figure 3, only (012), (024) and (036) lines of the LT film and sapphire substrate can be observed, showing that all the film is (012) oriented. Careful checks by focusing the x-rays on different regions of the films were performed and similar spectra were obtained. These results show that the films formed are either highly textured and have the same orientation of the substrates or have grown epitaxially on the substrates. A calculation shows that the difference between the lattice constant of the films and that of the bulk crystal of $LiTaO_3$ is only 0.13%.

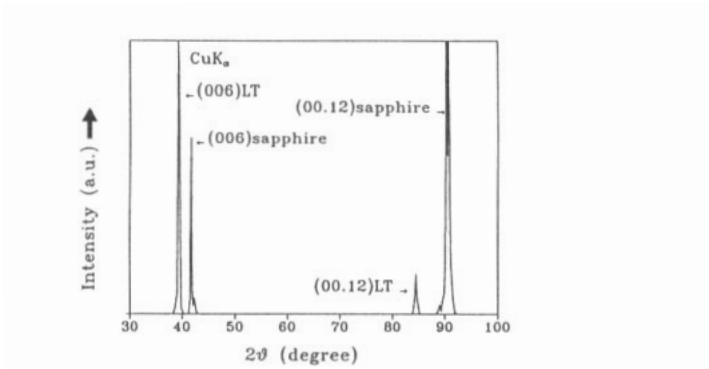

Figure 2. The XRD spectrum of the as-grown $LiTaO_3$ film on (001) sapphire substrate.

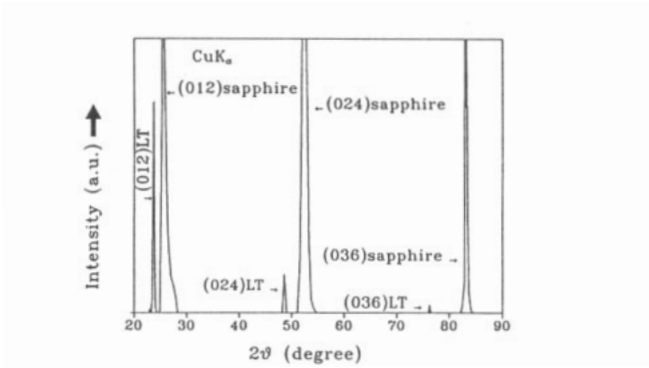

Figure 3. The XRD spectrum of the as-grown $LiTaO_3$ film on (012) sapphire substrate.

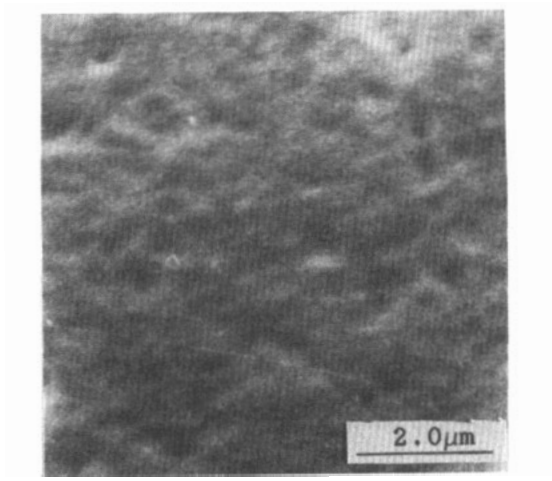

Figure 4. An SEM micrograph showing the surface morphology of a $LiTaO_3$ film on (001) sapphire substrate.

The surface morphologies of the as-grown films were first observed by polarized optical

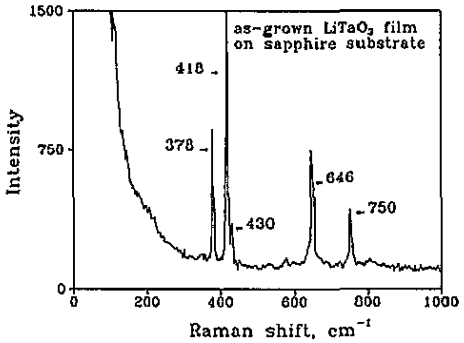

Figure 5. The Raman spectra of the as-grown $LiTaO_3$ film and (001) sapphire substrate at room temperature.

microscopy and then by the SEM technique. Before SEM observation, a thin layer of gold was deposited on the surface of the film which had been lightly etched in $HF:HNO_3$ agent. It was revealed that all the films are of a single-domain nature. The surface roughness is small and no large particle was observed on the surface. An SEM picture with a high magnification taken from a (001)-oriented film is shown in figure 4, which shows that the microstructure is very fine and the surface fluctuation is small compared with the film thickness (about 1.0 μm). It can be expected that this fluctuation will be further restrained as the film thickness is decreased. In fact, for the integrated-optic applications, films of a few hundreds nanometers thickness are normally needed.

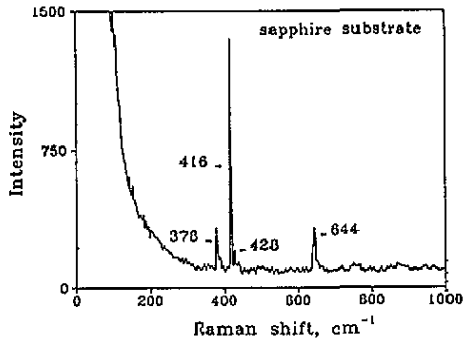

Figure 6. The Raman spectra of (001) sapphire substrate at room temperature.

Raman spectroscopy measurements were performed to check the quality of the as-grown films further. Raman spectra of the sapphire (001) substrate with and without the LT film are shown in figures 5 and 6, respectively. Considering that the Raman shift spectra are closely related to the phonon and electron structures of the sample, a polycrystalline film, even if it is highly textured, will lead to broadened Raman shift peaks. From the sharp shape of the Raman peaks shown in figure 5, it can be concluded that the LT film is a single crystal and grown epitaxially on the single-crystal sapphire substrate.

The back-scattering geometry $Z(X, M)\bar{Z}$ in our Raman measurements is used where Z and $\bar{Z}$ denote the directions of the incident and scattered light respectively, and X and M are the incident and scattered polarizations, respectively. For the latter case, M includes

x and y polarizations. The Raman tensor σ_{xx} in sapphire consists of the following Raman modes [9]: 378 (VW), 418 (VS), 432 (M), 451 (M), 580 (S), 645 (VW) and 756 (S) cm^{-1}, where VW is very weak, VS is very strong, M is medium and S is strong. It is obvious that the Raman peaks at 418 and 430 cm^{-1} in figure 5 are contributed purely from the transparent sapphire substrate. However, the 378, 644 and 748 cm^{-1} Raman peaks are from both the LT films and the substrate. The geometry $Z(X, M)\bar{Z}$ in the bulk crystal LT corresponds to the vibrational modes E(TO) and A_1(LO) [10]. Raman measurements showed that the E(TO) mode was at 382 and 383 cm^{-1} [11, 12]. Infrared measurements determined the E(TO) mode to be at 375 cm^{-1} [12]. Raman [11] and infrared [12] recorded the A_1(LO) modes at 660 and 750 cm^{-1} and at 625 and 760 (VW) cm^{-1}, respectively. On comparison of the results in figure 6 with those in figure 5, it is clearly shown that the Raman modes E(TO) at 378 cm^{-1}, and A_1(LO) at 646 and 750 cm^{-1} are mainly from the as-grown LT_3 films. The substrate sapphire made only a small contribution to them.

It should be pointed out that a single-crystal native of the films is not necessary for optical and electro-optic applications, although a single-domain film is a good choice. For integrated-optic applications, it is sufficient that c-oriented or a-oriented films be obtained because, for example for the c-orientation case, the current theory [2, 13] predicts that, as all the film is oriented totally along the c axis, a single coefficient d_{33} will be created. As reported [2], there may exist some microtwinned structures in the in-plane orientation, but this has no influence on d_{33}.

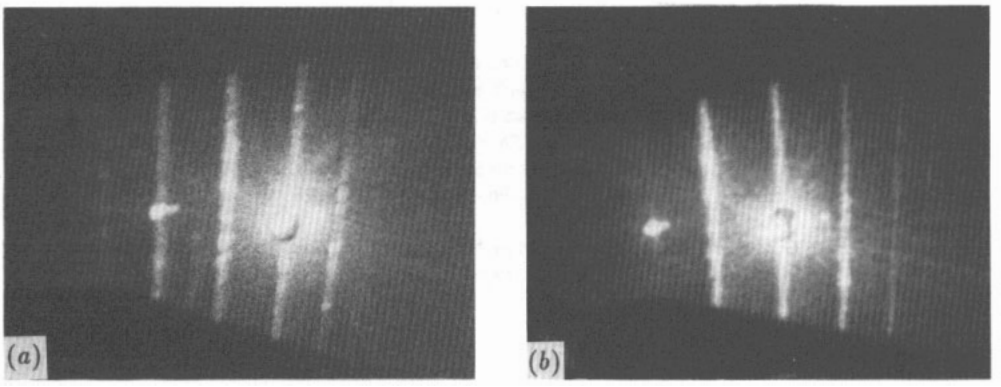

Figure 7. The observed m-lines of the waveguiding modes of the as-grown $LiTaO_3$ film on (001) sapphire, excited by a prism coupler through which a beam of 0.6328 μm laser was coupled into the film. (a) TM modes. (b) TE modes.

The optical waveguiding property of the LT films grown on both (001) and sapphire (012) was measured. Figure 7 shows the observed m lines of the waveguiding TM and TE multimodes of LT films excited by a prism coupler through which a laser beam of 0.6328 μm wavelength was coupled to the film. In the present case, six m lines for both TM and TE modes can be clearly identified, and these lines are sharp compared with the m lines observed by Haruna *et al* [8] in the epitaxial films of LN on sapphire. From these measurements, favourable waveguiding properties may be expected. By measuring the positions of these m lines, we can calculate the effective refractive indices of the films. Because the LT films were grown on sapphire substrates, we have *a priori* knowledge that the index profile across the interface between the film and the substrate has a step. The two refractive indices n_o

and n_e and the thickness of the film can be evaluated by a simple calculation. The details of the calculation have been described earlier [14]. We have obtained the indices $n_o = 2.161$ and $n_e = 2.171$, consistent with recently reported data [2, 8], and the calculated thickness of the film is 1.05 μm. For films with different thicknesses, fluctuations of only about 0.002 in the measured indices, compared with the values given above, have been found.

In summary, we have successfully prepared epitaxial LT waveguiding films on both (001) oriented and (012) oriented sapphire by excimer laser ablation. The quality of the as-grown films is quite good according to XRD, SEM and Raman scattering techniques. Optical waveguiding properties have been demonstrated.

Acknowledgments

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