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Potassium Tantalate-Niobate Mixed Crystal Thin Films for Applications in Nonlinear Integrated Optics

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Abstract. Potassium tantalate-niobate mixed crystal (KTN) thin films are promising candidates to meet the needs of integrated nonlinear optical devices for electro-optic and frequency-conversion applications. In this contribution we report on pulsed-laser-deposition growth of ferroelectric KTN films on MgO substrates. It is shown that highly-oriented KTN films are epitaxially grown as revealed by X-ray diffraction analysis. Moreover, the thermal annealing treatment can be further optimized to obtain optically smooth KTN films with RMS surface roughness as low as 1 nm.

1. Introduction

Perovskite potassium tantalate-niobate mixed crystals ($KTa_{1-x}Nb_xO_3$ with $0 \leq x \leq 1$, KTN) undergo a phase transition from a paraelectric cubic to a ferroelectric tetragonal structure with decreasing temperature [1]. By adjusting the Ta/Nb content ratio one can tune the phase-transition temperature of KTN and thus also its main properties at a given temperature. For example, temperature-dependent dielectric and electro-optic (EO) constants have been demonstrated with abrupt changes appearing around the Curie point of KTN, displaying the largest-known EO coefficients among all Kerr media [2]. Benefiting from this, this extremely promising material is showing potentials to overtake the well-known $LiNbO_3$ for EO applications such as beam scanners or modulators [2-4]. However, due to the fact that the crystals grown have compositions being different from those of the molten ingredients, high-quality and homogeneous single-crystalline KTN is difficult to produce, which is limiting the wide-spread use of this material [2]. Direct thin-film preparation, such as provided by pulsed laser deposition (PLD), appears to be an efficient way to solve the bulk-homogeneity problem. In this contribution, we report on the fabrication of smooth ferroelectric KTN thin films on MgO substrates by employing PLD and subsequent thermal annealing treatment and polishing process.

2. Experimental

The films were grown by employing a KrF excimer laser (λ = 248 nm, pulse duration of 20 ns) operating at a repetition rate of 1 Hz with a fluence of 2 J/cm^2 focused on a rotating K-enriched KTN ceramic target. The composition $x = 0.5$ is chosen because its corresponding Curie temperature ($T_c \approx$ 100 °C) ensures the crystalline structure of KTN stays in the ferroelectric phase at room temperature.

During the deposition, the substrate temperature was heated up to 700 ˚C, the target-substrate distance was set at 6 cm, and the oxygen atmosphere was around 0.15 mbar. Following the deposition procedure, in situ thermal post-annealing treatment at 700 ˚C with durations ranging from 0 to 4 hours and a subsequent chemo-mechanical polishing operation were conducted to further smoothen the KTN films. Specifically, a Logitech PM3 polishing machine and alkaline polishing suspension with 60 nm and 20 nm silicon dioxide particles (MasterMet1, MasterMet2) were used for the fine polishing process.

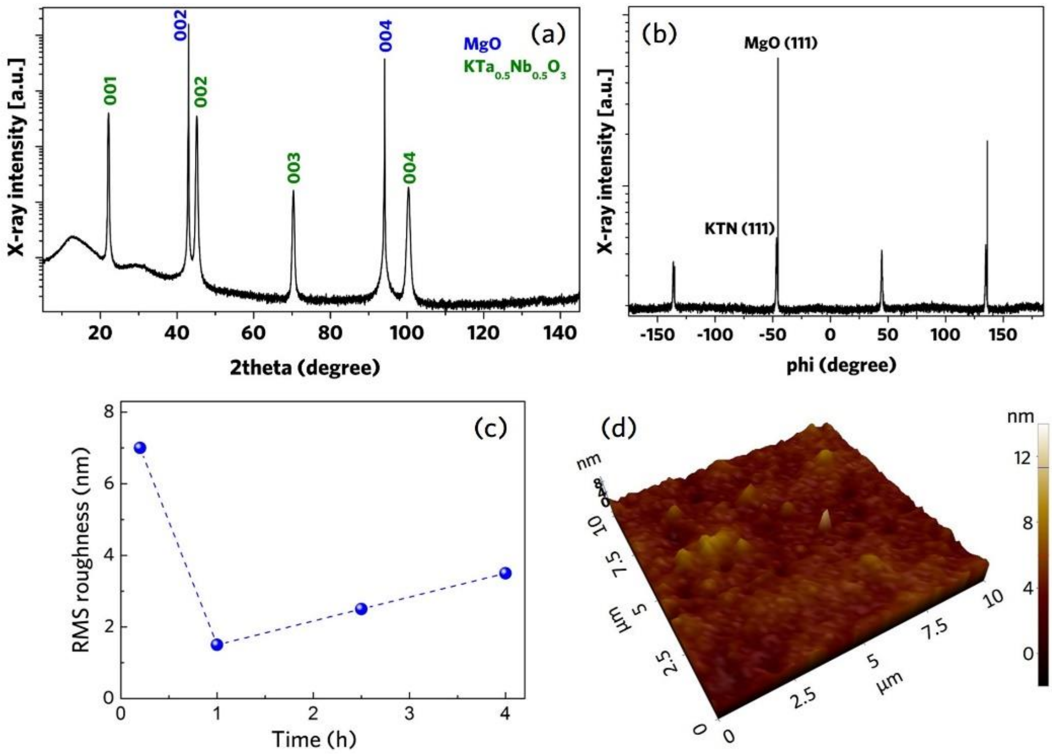

Figure 1. X-ray diffraction (a) θ-2θ and (b) φ-scan patterns of a KTN thin film deposited on (001)MgO. (c) RMS surface roughness of the polished KTN films after thermal annealing treatment with different durations. (d) AFM surface topographies of the KTN films with roughness of 1 nm.

3. Results and discussion

X-ray diffraction (XRD) analysis in θ-2θ and φ-scan modes were employed to investigate the crystalline structures of the KTN films. It was found that the as-deposited KTN films are completely (001)-oriented as shown by the θ-2θ XRD patterns in Fig. 1a, which exhibits only 00l peaks of KTN on (001)MgO, referring to the pseudocubic subcell. Rotationally-ordered growth of the films could be confirmed by the φ-scan XRD patterns (Fig. 1b) performed on the 111 reflection of KTN. Based on the XRD analysis, the lattice constants of the crystalline KTN thin films are determined to be $a = b =$ 3.9901 Å and c = 4.0121 Å, which are very close to those of KTN bulk crystals with tetragonal structures [5]. Moreover, energy dispersive X-ray (EDX) analysis performed on films showed that the K/(Nb+Ta) ratio is 0.95 ± 0.05 and that the Nb/Ta ratio in the films is the same as that in the starting targets, indicating good stoichiometry transfer between the target and the films. In order to obtain the surface topographies of the polished KTN films, atomic force microscopy (AFM) in contact mode was used. The change in roughness values as a function of annealing time (Fig. 1c) indicates that the duration of thermal annealing could be optimized to further reduce the RMS surface roughness to about 1 nm (Fig. 1d), which is the lowest reported value so far for PLD-grown KTN thin films. Furthermore, the waveguiding properties of the ferroelectric KTN films were studied by using the prism coupling method (Metricon® Model 2010 prism coupler). Based on the effective refractive indices of excited TE and TM guiding modes, the refractive indices n_o = 2.258 and n_e = 2.214 of the KTN films at 632.8-nm wavelength could be determined, respectively.

The data presented in this contribution shows that PLD-grown $KTa_{0.5}Nb_{0.5}O_3$/MgO films may reach a quality satisfactory for integrated-optical applications. Our future work will focus on the fabrication and functionalization of KTN waveguide-based modulators, frequency converters and on-chip micro-resonators.

References

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