Epitaxial growth of optical $Ba_2NaNb_5O_{15}$ waveguide film by pulsed laser deposition

J. M. Liu, F. Zhang, Z. G. Liu, and S. N. Zhu
National Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210093, China and Center for Advanced Studies in Science and Technology of Microstructures, Nanjing 210093, China

L. J. Shi
Department of Materials Science, Nanjing University of Science and Technology, Nanjing 210014, China

Z. C. Wu and N. B. Ming
National Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210093, China

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The waveguiding $Ba_2NaNb_5O_{15}$ films have been prepared on $KTiOPO_4$ substrates by pulsed laser deposition technique. Characterizations of the as-grown films by the x-ray θ–2θ scan and β scan and the scanning electron microscopy revealed that epitaxial (110)-oriented $Ba_2NaNb_5O_{15}$ films with small surface roughness were achieved on (100)-oriented $KTiOPO_4$ plates. The x-ray photoelectron spectrometer was employed to probe the chemical constitution of the as-grown films. Favorable optical waveguiding properties were demonstrated by *m*-line measurement and evaluation of propagation loss on the order of 1.0 dB/cm of TM and TE multimodes. © *1994 American Institute of Physics.*

Nonlinear optical materials attract particular interest both as objects of fundamentals in condensed matter, and also from the applicational point of view. One of the potential applications is fabrication of the new types of integrated-optic devices,[1] such as the blue or green sources used in optical storage, laser printers, etc., where generation of blue or green light is realized by frequency doubling of semiconductor diode lasers. Recently, particular attention has been paid to the fabrication of integrated-optic devices in which the nonlinear optical films show many advantages over the bulk crystals, and incorporation of the optical thin films into semiconductor integrated circuits.[2–7]

$Ba_2NaNb_5O_{15}$(BNN) is one of the most important materials for fabrication of electro-optic devices and exhibits advantages over $LiTaO_3$ and $LiNbO_3$ in the integrated optic applications. However, due to the difficulty in growth of bulk crystal with large size and easy formation of complicated twined substructures this material has not been emphasized in the last decade. In thin film form, it is expected that the twined substructure can be effectively prohibited. Therefore, fabrication of epitaxial BNN films on a suitable substrate becomes very attractive in opening a new application in the integrated optics.

In the last few years, a series of techniques for preparation of epitaxial ferroelectric films have been developed, including rf sputtering, molecular beam epitaxy, liquid phase epitaxy, and recently pulsed laser deposition (PLD).[2–7] Among these techniques, PLD shows some advantages over the others, especially the possibility of preparing epitaxial films and higher compositional consistency between the films and target. In this letter, preparation of BNN films on (100)-oriented $KTiOPO_4$(KTP) substrates by PLD will be reported.

BNN has a tetragonal-type structure with $a=1.7609$ nm and $c=0.7987$ nm with point group 2 mm. It will experience a ferroelastic transition at about 300 °C, which is responsible for the formation of twined substructures. KTP has an orthorhombic-type structure and its lattice constants are $a=1.058\,88$ nm, $b=1.281\,48$ nm, and $c=0.640\,36$ nm. The lattice differences between BNN and KTP are over 20% in both [100] and [010] axes. However, for the case of (110)BNN//(100)KTP, the lattice parameter of BNN in the [110] direction is almost two times that of KTP in the [010] direction, it is then possible for (110) BNN film to be grown epitaxially on (100) KTP with an in-plane alignment of [110]BNN//[010]KTP. And more, for a BNN/KTP combination, a theoretical modeling shows that in waveguiding second-harmonic generation applications, the phase-matching condition for second-harmonic generation can be satisfied for the TM_0 mode of the fundamental wave and the TE_1 mode of the second-harmonic wave over a wide range of film thickness if a laser beam of 1.02 μm is coupled into the film.[8] This relaxation in film thickness limitation makes the preparation of BNN film on KTP plates very attractive.

The PLD experiments were performed by using the KrF excimer pulsed irradiation of 248 nm in wavelength, 30 ns in pulse width, and 5 Hz in frequency. The apparatus used in this work has been described in our previous work.[9] The stoichiometric disc-shaped BNN single-crystal target was mounted at an angle of 45° to the laser beam and the substrate surface. The distance L between the substrate and target was 3.5 cm. During deposition the target and the substrate were driven to rotate at a speed of 0.1 and 0.2 rad/s, respectively. An optimum pressure $P=10$–20 Pa of oxygen which was leaked into the chamber as the reactive ambient was determined. To reduce the surface roughness and enhance the crystallinity of the films, the substrate temperature and energy fluence of the excimer irradiation were optimized at 700 °C and 1.2 J/cm^2, respectively. After deposition, the samples were kept in the chamber at 700 °C and 400 Torr O_2 ambient for 30 min to avoid oxygen deficiency, and then cooled to room temperature.

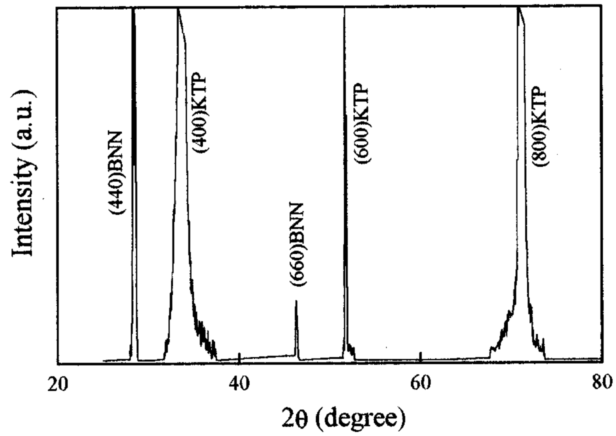

FIG. 1. The x-ray θ-2θ scan of the as-grown BNN film on (100)-oriented KTP substrate.

The x-ray θ–2θ scan of the as-grown film on (100)-oriented KTP plate is presented in Fig. 1. The presence of only (440) and (660) reflections of BNN film reveals that all the film is (110) oriented. Careful checks by focusing the x-ray on different regions of the film were performed and similar spectra were obtained. These results show that the formed films are either highly (110) textured or grown epitaxially on the substrates. The in-plane orientation and the alignment relation of the as-grown BNN films with the substrates were determined by the x-ray β scan. Figure 2(a) shows the x-ray β scan of the (820) reflections from the film, which are 58.82° off normal to the film surface. Only two peaks separated by a 180° azimuth angle were observed. Figure 2(b) displays the x-ray β scan of the (240) reflections of the substrate. A comparison of Figs. 2(a) and 2(b) reveals that (110)-oriented BNN films were grown epitaxially on (100)-oriented KTP plate with an in-plane alignment of [001]BNN//[001] KTP and [110]BNN//[010] KTP, as we argued above. Here, it should be pointed out that single crystallinity of the films is not necessary for optical and electro-optic applications, although single domain film is a good choice. For the present case, the in-plane alignment of [001]BNN//[001] KTP provides the possibility of utilizing d_{33}, because the current theory[10] predicts that as all the film has the same orientation, a single coefficient will be created.

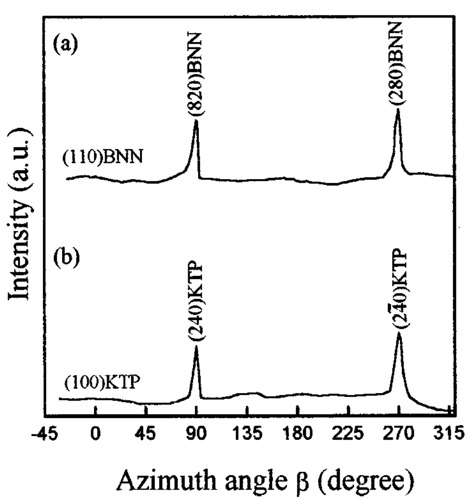

FIG. 2. The x-ray β scan of (a) the (820) and $(2\bar{8}0)$ reflections of the (110)-oriented BNN film and (b) the (240) and (240) reflections of the (100)-oriented KTP substrate on which the film was grown.

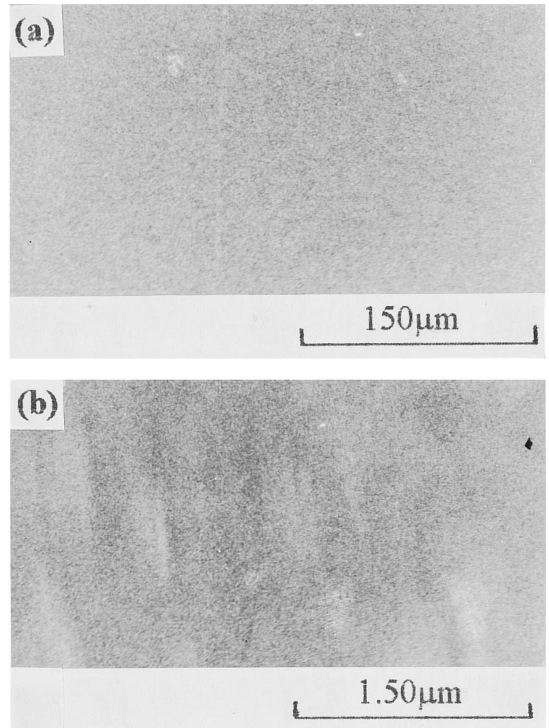

FIG. 3. The SEM pictures showing the surface morphology of the BNN film grown on a (100)-oriented KTP plate.

The surface morphology of the as-grown films were first observed by polarized optical microscopy and then by scanning electron microscopy (SEM). In Figs. 3(a) and 3(b) are presented SEM micrographs at two magnifications of the surface morphology of an as-grown film 1000 nm thick. From

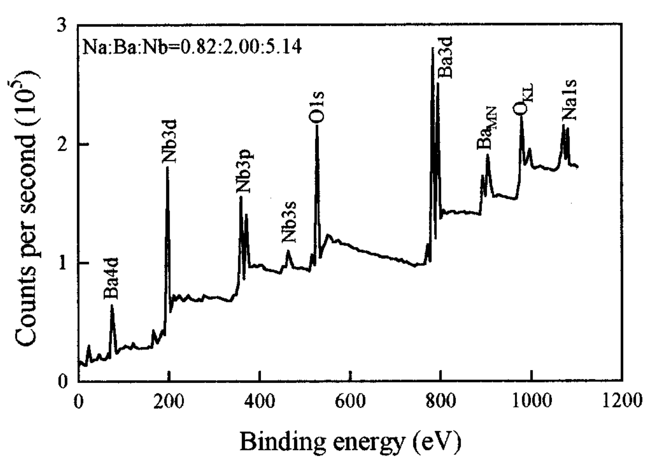

FIG. 4. The XPS spectrum of the as-grown BNN film on (100)-oriented KTP substrate, showing that the atomic ratio of Na:Ba:Nb is 0.82:2.00:5.14.

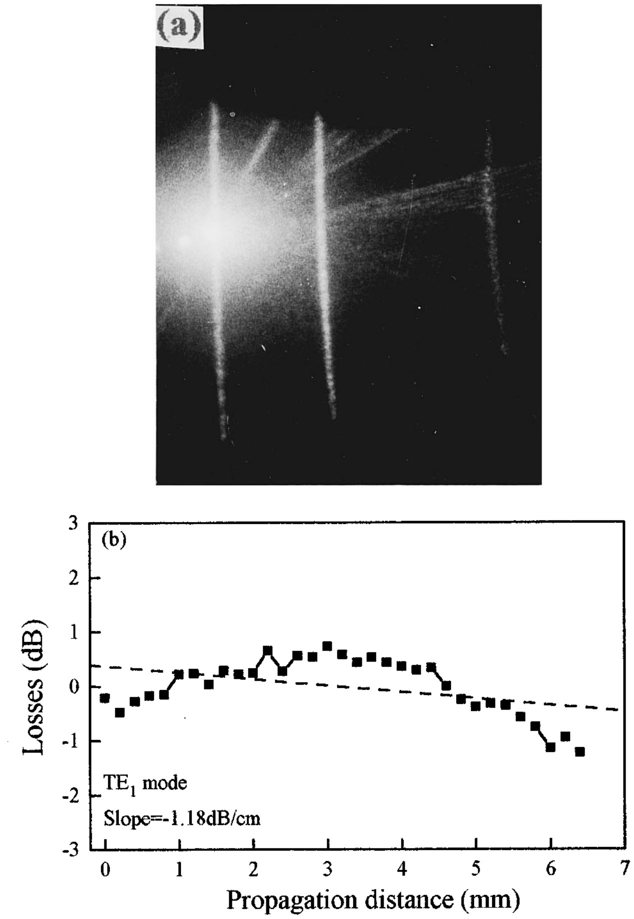

FIG. 5. The observed *m* lines of the TE multimodes of a (110)-oriented BNN film grown on (100)-oriented KTP plate (a) and the scattered light intensity out of the TE_1 mode as a function of the propagation distance. (b) The dashed line represents the best-linear fitting.

Fig. 3(a) it was revealed that all the film is single domained and no grain boundary can be identified. Figure 3(b) reveals that the surface roughness is small and no large particulate was observed on the surface. The stochiometry of the as-grown film was characterized by x-ray photoelectron spectrometry (XPS) technique. Before the XPS analysis, a thin surface layer of about 10 nm of the film was removed by the Ar^+ ion beam sputtering. The measured XPS spectrum is presented in Fig. 4, from which the atomic ratio of Na, Ba, and Nb is calculated to be 0.82:2.00:5.14. This value is shown to be roughly consistent with the nominal composition of BNN, although the element Na is lower than the nominal value possibly due to preferential evaporation during deposition and post-annealing.

The optical waveguiding property of the as-grown BNN films on a (100)-oriented KTP plate was measured. Figure 5(a) shows the observed *m* lines of the waveguiding TE multimodes for a BNN film of about 1000 nm in thickness, excited by a prism coupler through which a laser beam of 0.6328 μm was coupled into the film. The quite good quality of the film can be proved by the these sharp lines. Figure 5(b) presents the measured optical propagation loss. The data represent the intensity of the scattered light from the transmitting light beam in the TE_1 mode waveguide form, as a function of the propagation distance. It is revealed that there are large fluctuations of the measured data which are expected to be attributed to possible existence of some particulates in the interfacial layer between the film and substrate, because there exists a large lattice mismatch for the film and substrate along [001] axis. From the best fitting curve over these data a loss of about 1.18 dB/cm was obtained. For other guided modes, propagation loss on the order of 1.0 dB/cm was measured. By measuring the positions of these *m* lines, the refractive index *n* and thickness of the films can be calculated.[11] We have obtained the index $n=2.10$, slightly smaller than that of bulk BNN crystal (about 2.19). This difference may be attributed to compositional deviation and the lattice distortion of the film from the nominal value.

In summary, we have prepared (110)-oriented epitaxial BNN waveguiding films on (100)-oriented KTP substrate by PLD technique. The quality of the as-grown films is well by checking the x-ray diffraction techniques, second electron images of the film surface, and XPS. Favorable optical waveguiding properties have been demonstrated. To our knowledge this letter represents the first report on preparation of BNN film.

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