Proc. Symp. Ultrason. Electron., Vol. 27, (2006) pp. 287-288
15-17 November, 2006

Fabrication and Evaluation of Potassium Niobate Thin-Film by RF Sputtering

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1. Introduction

Potassium niobate $KNbO_3$ is a promising material for realizing excellent functional devices due to large constants such as the piezoelectric constant and nonlinear optical constant. However, the growth of a $KNbO_3$ single crystal of a large size is difficult due to the occurrence of several phase transitions. Therefore, the deposition of high -quality $KNbO_3$ thin films[1,2] is required.

Odagawa *et al.* proposed the deposition of $KNbO_3$ thin film by rf-sputtering with dual targets of $KNbO_3$ and K_2CO_3 to prevent the lack of K in the deposited film.[3] By using this deposition method, the authors deposited an oriented $KNbO_3$ thin film onto a MgO(100) substrate.[4] Although a $KNbO_3$ thin film sputtered with a single target of $KNbO_3$ was oriented, the composition ratio of K/Nb was small, around 0.13. On the other hand, the film sputtered with dual targets of K_2CO_3 and $KNbO_3$ showed a large K/Nb ratio of 0.65, but the orientation was deteriorated.

In this study, to clarify the sputtering conditions necessary for obtaining a high-quality $KNbO_3$ thin film, the relationships among the degree of orientation, the K/Nb ratio, and the X-ray diffraction angle were evaluated for $KNbO_3$ film sputtered using several kinds of targets.

2. Fabrication of $KNbO_3$ thin film

2.1 Sputtering conditions

Figure 1 shows the configuration of the rf-sputtering system used for the deposition of the $KNbO_3$ film onto the MgO(100) substrate. The common sputtering parameters are as follows. The rf power applied to the cathode and the O_2-radical source was 50 W. The atmosphere gas flow rate Ar:Ar:O_2 for two cathodes with/without the target and radical source was 4:4:2 ccm. The deposition time was 10 hours. The deposition rate was in the range from 35 to 60 nm/h. The K/Nb ratio of the thin film was determined from the analytical results of X-ray photoelectron spectroscopy (XPS).

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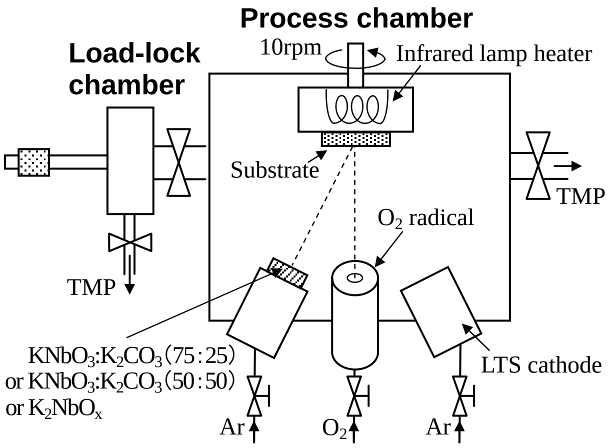
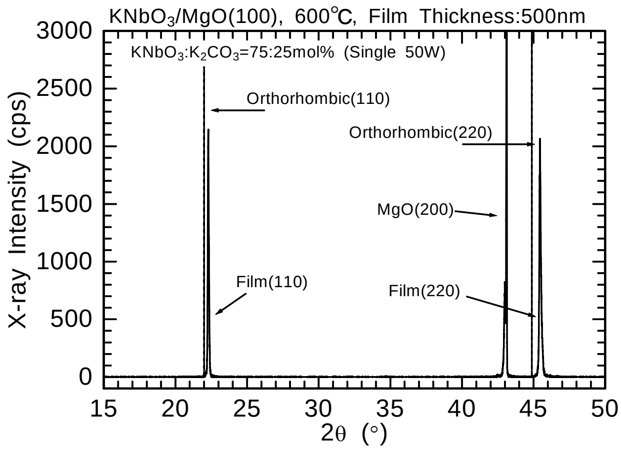
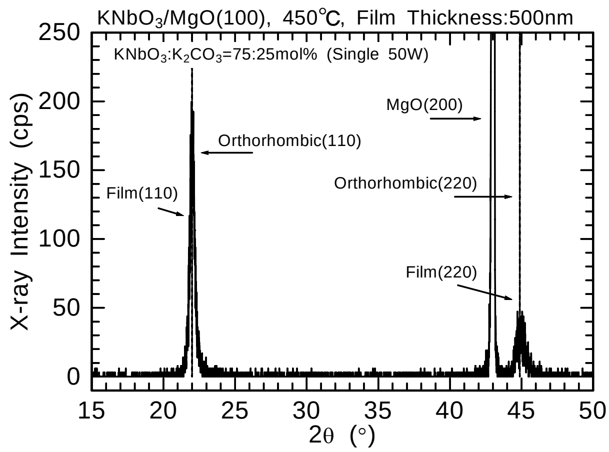


Fig. 1 Configuration of RF sputtering system.



(a) substrate temperature of 600℃



(b) substrate temperature of 450℃

Fig. 2 X-ray diffraction pattern of $KNbO_3$ film sputtered onto MgO(100) substrate using mixture target of $KNbO_3$:K_2CO_3 with 75:25 mol%.

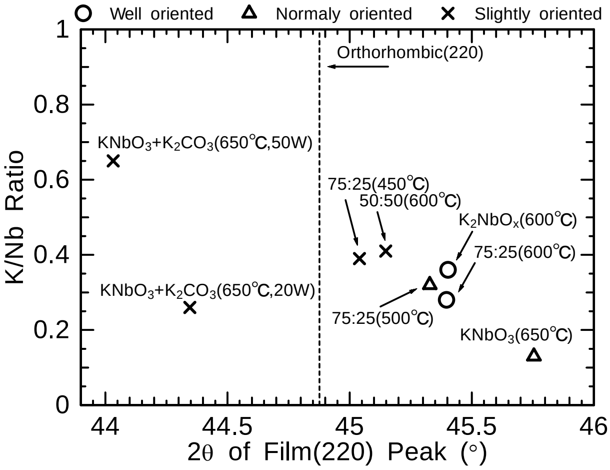


Fig. 3 Relationship between measured K/Nb ratio and X-ray diffraction angle (220)-film. Symbols are plots of degree of orientation.

2.2 Sputtering using mixture target of $KNbO_3$:K_2CO_3

First, the mixture target of $KNbO_3$:K_2CO_3 with the mixture ratio of 75:25 mol% was used. **Figure 2** (a) shows the X-ray diffraction pattern of $KNbO_3$ film sputtered at a substrate temperature of 600℃. A better orientation and larger K/Nb ratio of 0.28 were obtained compared with the film sputtered with the single $KNbO_3$ target. To prevent the reevaporation of K from the deposited film during sputtering, the substrate temperature was decreased to 450℃. As a result, the K/Nb ratio was increased to 0.39, however, the orientation was markedly deteriorated, as shown in Fig. 2 (b).

Second, to obtain a higher K/Nb ratio, another mixture target of $KNbO_3$:K_2CO_3 with the mixture ratio of 50:50 mol% was adopted. The K/Nb ratio of the thin film deposited at a substrate temperature of 600℃ was improved to 0.41 compared with that in the case of the mixture target with 75:25 mol%, but the orientation deteriorated rapidly.

Figure 3 shows the relationship between the measured K/Nb ratio and the X-ray diffraction angle corresponding to the (220)-plane for the $KNbO_3$ thin films deposited using the above-mentioned targets. The symbols in the figure indicate the degrees of orientation of the thin film, as determined from the X-ray diffraction patterns. The ratios indicated in the figure are the mixture ratios of the targets used.

In dual sputtering with the K_2CO_3 target, the X-ray diffraction angle deviated from that for the (220)-plane of orthorhombic bulk $KNbO_3$ with increasing K/Nb ratio. There is a possibility that the crystal structure of the thin film prepared using the K_2CO_3 target is different from that of orthorhombic bulk $KNbO_3$.

On the other hand, in single sputtering with the mixture target of $KNbO_3$:K_2CO_3, the X-ray diffraction angle approaches that of orthorhombic bulk $KNbO_3$ with increasing K/Nb ratio. Therefore, it is considered that this group has a crystal structure close to that of orthorhombic bulk $KNbO_3$. Furthermore, the orientation of this group deteriorated with increasing K/Nb ratio. This may be caused by an increase in the content of CO_2 in the mixture target.

2.3 Sputtering using K_2NbO_x target without CO_2

On the basis of the above results, a K-rich target, K_2NbO_x, without any CO_2 content was adopted. The target was prepared by burning K_2CO_3 and Nb_2O_5 in air at 700℃ to evaporate CO_2, and followed by sintering at 700℃. By using this K_2NbO_x target, good orientation and a K/Nb ratio of 0.36 were obtained simultaneously, as shown in Fig. 3.

As a result, by using a K-rich target without CO_2, the well-oriented $KNbO_3$ thin film with a high K/Nb ratio and a crystal structure close to that of orthorhombic bulk $KNbO_3$ can be expected.

3. Conclusions

To clarify the sputtering conditions necessary for obtaining a high-quality $KNbO_3$ thin film, the relationships among the degree of the orientation, the K/Nb ratio, and the X-ray diffraction angle were evaluated. By using a K-rich target without CO_2, the well-oriented $KNbO_3$ thin film with a high K/Nb ratio and a crystal structure close to that of orthorhombic bulk $KNbO_3$ can be expected.

Acknowledgment

This work was supported by the Ministry of Internal Affairs and Communications, Strategic Information and Communications R&D Promotion Programme, and the Industrial Technology Research Grant Program in 2006 from NEDO of Japan.

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