c-axis parallel oriented ZnO film depositions by variable frequency RF bias sputtering

周波数可変 RF バイアススパッタ法による

c 軸平行配向 ZnO 薄膜の形成

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1. Introduction

Hexagonal piezoelectric films where the crystallite c-axis is unidirectionally-aligned and parallel to the substrate plane [(1120) or ($10\overline{10}$) textured films] are good candidates for exciting a shear bulk acoustic wave and surface horizontal (SH) type SAW devices.^{1,2}

In previous studies, we found that $(11\overline{2}0)$ or $(10\overline{1}0)$ texture formation was induced by the ion bombardment during deposition.¹⁾ Ion bombardment suppresses the (0001)-oriented grain growth, resulting in the preferential development of $(11\overline{2}0)$ or $(10\overline{10})$ texture instead of the (0001) texture. This is because the most densely packed (0001) plane should incur more damage by ion bombardment than the (1120) and (1010) planes. For example, using oxygen ion-beam-assisted electron-beam evaporation of zinc, texture formation of ZnO can be controled by increasing ion energy irradiated to the substrate.¹⁾ However, there are problems, such as the difficulty of large area deposition and the expensive ion source. On the other hand, $(11\overline{2}0)$ textured films can be formed under low pressure (< 0.1 Pa) using RF magnetron sputtering.²⁾ However, there are also problems, such as the instability of discharge and the limitation of deposition conditions. Therefore, it is difficult to obtain (1120) or (1010) textured ZnO films easily.

In this study, we propose DC or RF substrate bias RF magnetron sputtering method to induce the ion bombardment to the substrate. We succeeded in the control of $(11\overline{2}0)$ or $(10\overline{1}0)$ textured ZnO films using this method.

2. Bias sputtering method

Figure 1 shows a deposition system. A bias supply is connected to the bias electrode on which a substrate is set, in an RF magnetron sputtering system (Ulvac RFS-200).

Table I shows expected conditions of ion irradiation to the substrate during film deposition.³⁾ Without bias, high-speed negative ions from the ZnO target should bombard the substrate. In the positive DC bias condition, the ion irradiation is similar to the case without bias. In the negative DC bias condition,



Fig.1 Bias sputtering apparatus.

Table I Expected conditions of ion irradiation to the substrate.³⁾

	Positive ion	Negative ion
Without	Small amount	Medium amount
bias	Low energy	High energy
Positive	Small amount	Medium amount
DC bias	Low energy	High energy
Negative	Small amount	Small amount
DC bias	High energy	Low energy
RF bias	Large amount High energy	Small amount Low energy

positively ionized gas should be accelerated toward the substrate. On the other hand, in the RF bias condition, large amounts of gas molecules should ionize. Therefore, large amounts of positive ion bombard the substrate by the self-bias.

3. ZnO film fabrication

The ZnO films were fabricated on Al electrode film/silica glass substrate structures. The total gas pressure was always 1.0 Pa, gas composition ratio of Ar/O₂ was 1/3 and deposition time was 1 hour. RF power of 200 W at 13.56 MHz was supplied to the target electrode. In these conditions, $(11\overline{2}0)$ or $(10\overline{1}0)$ texture formation cannot occur in the conventional system. The ZnO films were deposited (1) without bias, (2) with DC bias of -200, -100 or 100 V, (3) with 2 MHz RF bias of 5, 10, 15, 20 or 25 W, or (4) with 80 MHz RF bias of 50, 100, 150, 200 or 250 W.

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4. Crystallographic orientation of the films

The crystallographic orientations of the samples were determined by a $2\theta - \omega$ scan XRD pattern using an x-ray diffractometer (Philips, X-Pert Pro MRD). Figure 2 shows the XRD patterns of the samples at the center of bias electrode under the different bias conditions.

In wurtzite films, like ZnO, the (0001) plane has a lower surface energy density than all other planes, which results in the easy formation of the (0001) preferred orientation.⁴⁾ As shown in Fig. 2(c), an intense (0002) peak was observed in the sample deposited without bias. This is because the negative ion irradiation from ZnO target to the substrate is not strong. In the positive DC bias condition, the (0001) preferred orientation did not change to other orientation, because the condition of ion irradiation changed little. In the negative DC bias conditions, the (0001) preferred orientation only occurs. The amounts of positive ion irradiation to the substrate seem to be small, because only a part of positively ionized gas bombards the substrate.

In the RF bias conditions, as shown in Figs. 2(a) and 2(b), the (0001) preferred orientation changed into the $(11\overline{2}0)$ preferred orientation with RF bias at both frequencies of 2 and 80 MHz. The $(11\overline{2}0)$ preferred orientation then changed into the (1010) preferred orientation with higher RF bias power. One reason of these changes is the increase of positively ionized gas by the RF bias, and a large amount of positive ion was finally irradiated to the substrate. In addition, the appearances of (0001), $(11\overline{2}0)$, and $(10\overline{1}0)$ textures correspond with the order of the surface energy density of 9.9, 12.3 and 20.9 $eV/nm^{2.4}$ The higher the surface energy density is, the lower the surface atomic density becomes. This agrees with the mechanism of the $(11\overline{2}0)$ or $(10\overline{1}0)$ preferential development as described previously. Comparing Figs. 2(a) and 2(b), in the samples deposited at RF bias frequency of 2 MHz, the orientation changes occurred at smaller RF bias power. Therefore, the ion energy irradiated to the substrate seems to be higher at the bias frequency of 2 MHz.

5. Conclusion

We examined the control of $(11\overline{2}0)$ or $(10\overline{1}0)$ textured ZnO films using the DC or RF substrate bias RF magnetron sputtering method. We successfully fabricated these ZnO films at the conditions in which these textures cannot be formed in the conventional system. Further investigation of the quantitative ion energy evaluation and the amount of ion irradiation is expected.

References

- T. Yanagitani and M. Kiuchi: J. Appl. Phys. 102 (2007) 044115.
- T. Kawamoto, T. Yanagitani, M. Matsukawa and Y. Watanabe: Jpn. J. Appl. Phys. 46 (2007) 4660.
- S. Takayanagi, et. al.: Proc. Piezoelectric Materials & Devices Symp. 2010 p. 3.
- 4. N. Fujimura, et. al.: J.Crystal Growth 130 (1993) 269.



Fig. 2 $2\theta \cdot \omega$ scan XRD patterns of the samples deposited without bias, (a) with 80 MHz RF bias of 50, 100, 150, 200 or 250 W, (b) with 2 MHz RF bias of 5, 10, 15, 20 or 25 W, or (c) with DC bias of -200, -100 or 100 V.