Piezoelectric Characteristics of c-Axis Oriented CrAIN Films Grown by RF Magnetron Sputtering

RF マグネトロンスパッタ法による c 軸配向 CrAlN 膜の形成と 圧電特性評価

Yusei Takano[‡], Ryusei Hayakawa, Masashi Suzuki, and Shoji Kakio (Univ. of Yamanashi) 高野佑成[‡], 早川竜盛, 鈴木雅視, 垣尾省司 (山梨大)

1. Introduction

AlN film bulk acoustic wave resonators (FBARs) are used as a frequency filter in mobile communications because of its high frequency operation, high Q factor, and good temperature stability. However, the electromechanical coupling factor k^2 of approximately 6% in AlN FBARs is relatively small, which causes a narrow band width in frequency filter applications. In 2008, M. Akiyama et al reported that heavily doping Sc with c-axis oriented AlN films enhances piezoelectricity of the films [1]. The d_{33} of the Sc_{0.43}Al_{0.57}N films is approximately five times higher than that of a pure AlN film. The increase of piezoelectricity in ScAlN films were theoretically demonstrated by a density functional theory [2]. This enhancement of piezoelectricity in ScAlN films leads increase of electromechanical coupling factor of AlN FBAR. Therefore, several researchers have reported the characteristics of ScAlN FBARs [3-5]. The k_t^2 of c-axis oriented ScAlN films increased with increasing Sc concentration. The k_t^2 of 18.5% in c-axis oriented Sc_{0.4}Al_{0.6}N film is approximately 1.9 times higher than that of a pure AIN single crystal. However, rare earth Sc is very expensive. In addition, crystalline orientation is degraded by impurity ions irradiation from Sc target to ScAlN films during film growth. In 2014, the increase of electromechanical coupling factor in MgZrAlN FBAR was found by Taiyo Yuden Co., Ltd. [6]. Mg and Zr is cheaper than Sc, but Mg and Zr concentration control is more difficult than that of Sc. In 2018, S. Manna et al demonstrated that doping Cr with AlN films also enhanced piezoelectric constant d_{33} [7]. The cost of Cr doping is much cheaper than that of Sc doping. Moreover, we may obtained Cr doped AlN films in same film growth techniques as ScAlN film growth. However, piezoelectric characteristics in GHz range of CrAlN FBARs have not been reported yet.

In this study, c-axis oriented Cr doped AlN films were grown by Cr ingot RF magnetron sputtering. The crystalline orientation of the films were evaluated by an XRD analysis. The k_t^2 and longitudinal wave velocity V_L of CrAlN films were estimated from the conversion loss frequency characteristics of CrAlN film bulk acoustic wave resonators. From these results, we investigated the relationship k_t^2 or V_L and Cr concentration in CrAlN films.

2. CrAlN film growth

Cr doped AlN films were grown by Cr ingots RF magnetron sputtering shown in Fig. 1. Cr ingots (99.9 %, Kojundo chemical Lab. Co. Ltd.) set on Al target. Cr ingots and Al target are sputtered simultaneously, and Cr doped AlN film grows on the substrate. In order to control Cr concentrations of the films, the amount of Cr ingots is adjusted to be 0, 0.1, 0.25, 0.5, 1.0 g. (0001) oriented Ti bottom electrode films (400-700 nm) on silica glasses $(25 \times 25 \times 0.5 \text{ mm}^3, \text{Fujikagaku Inc.})$ were used as the substrates. Base pressure, gas pressure, N2:Ar gas ratio, RF power, distance form target to substrate, and substrate temperature were set to be $<10^{-3}$ Pa, 0.75 Pa, 1:2, 150-200 W, 40 mm, room temperature, respectively. The film thickness of the CrAlN films were 3-5 μ m. The Cr concentration in the Cr_xAl_{1-x}N films were determined to be x=0-0.16 by an Electron Probe Micro Analyzer (JXA-8200, JEOL).



Fig. 1 Schematic of Cr ingot RF magnetron sputtering deposition.

masashis@yamanashi.ac.jp

3. Crystalline orientation

Crystalline orientation of AlN and CrAlN films were estimated by an XRD analysis. **Figure 2** shows 2θ - θ XRD patterns of the samples. (0002) AlN peaks around 36°, indicating that c-axis is normal to substrate surface, were observed in all samples. Rocking curve FWHM (R.C. FWHM) of (0002) AlN peak in AlN, Cr_{0.01}Al_{0.99}N, and Cr_{0.04}Al_{0.96}N films were estimated to be 2.2°-2.8°. The crystalline orientation is good in Cr_xAl_{1-x}N film with Cr concentration x<0.04. On the other hand, R.C. FWHM in Cr concentration x>0.06 were more than 5°. We consider that peaks around 40° are due to (111) CrN.

4. Piezoelectric properties

We fabricated high overtone bulk acoustic wave resonators (HBARs) consisting of Cu (Au) top electrode film / Cr_{0<x<0.16}Al_{1-x}N film / Ti bottom glass electrode film silica substrate. / Electrotechnical coupling factor k_t^2 and longitudinal wave velocity $V_{\rm L}$ were estimated by comparing experimental conversion loss curves of the HBARs, measured by a network analyzer (Agilent Technologies, E5071B), with theoretical curves calculated by a Mason's equivalent circuit model. The density and dielectric constants of an AlN single crystal were used as that of CrAlN films in Mason's model. Figure 3 shows k_t^2 and V_L of CrAlN films as a function of Cr concentration. In good orientation samples (red circle plots, FWHM<3°), k_t^2 of Cr_{0.01}Ål_{0.99}N was approximately 1.3 times higher than that of the pure AlN film. These results demonstrated that k_t^2 is increased by low Cr concentration doping with AlN films. On the other hand, the k_t^2 decreased with increasing with Cr concentration form x=0.04. In addition, we did not observe the excitation of bulk wave in Cr_{0.16}Al_{0.84}N film HBAR. As shown in Fig. 2, (0002) peaks intensity of $Cr_{0.16}Al_{0.84}N$ film is much smaller than that of CrAlN films with low Cr concentration. From these results, the crystal phase of Cr_{0.16}Al_{0.84}N film may be changed from a piezoelectric wurtzite system to a non-piezoelectric cubic system. Longitudinal wave velocity $V_{\rm L}$ (blue plot in Fig. 3) decreased with increasing Cr concentration. This decrease of V_L by doping other elements with AlN films were observed in ScAlN films and MgZrAlN films.

5. Conclusion

We grew $Cr_{0\le x\le 0.16}Al_xN$ films by Cr ingot RF magnetron sputtering deposition. Electrotechnical coupling factor k_t^2 and longitudinal wave velocity V_L were estimated in GHz range. The increase of k_t^2 in low Cr concentration and the decrease of V_L with



Fig. 3 Relationship between k_t^2 or V_L and Cr concentration of $Cr_{0 < x < 0.16}Al_{1-x}N$ films.

increasing Cr concentration were observed in CrAlN films. The k_t^2 of 5.6% in Cr_{0.01}Al_{0.99}N film was approximately 1.3 times higher than that of 4.4% in the pure AlN film.

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