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Acoustic properties of co-doped AlN thin films at low temperatures studied by picosecond ultrasonics

ピコ秒超音波法を用いた 2 元素添加 AlN 薄膜の音響特性の極低温計測

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1. Introduction

AlN is an important piezoelectric material and used in film-bulk acoustic resonators (FBARs) in various wireless devices. Although AlN can operate at high temperatures (up to 1400 K), its low piezoelectric constants have been desired to be improved. Other group III nitrides and many transition-metal nitrides have been studied because of their outstanding physical properties; among them, ScN exhibits a cubic structure without piezoelectricity. Takeuchi however found that there is a metastable wurtzite structure of ScN, which has a large indirect band gap of ~3 eV, and suggested the possible fabrication of wurtzite Sc-IIIA-N nitrides using a first-principle calculation.1) In 2009, Akiyama *et al*. succeeded in synthesizing wurtzite $Sc_xAl_{1-x}N$ where $x < 0.5$ and found that the piezoelectric constant d_{33} was enhanced at least by 500% comparing to AlN.2) Its piezoelectricity enhancement was confirmed by first-principle calculations,3) and increment of electromechanical coupling constant with Sc was also confirmed in a FBAR.4) However, Sc is an expensive element and it is not suitable for mass production. Other doping elements have therefore been investigated; Yokoyama *et al*. proposed $(MgZr)_xAl_{1-x}N$, which shows approximately three times larger d_{33} comparing to AlN at x = 35 at.%, leading to a possible candidate of a high piezoelectricity material.5) However, its fundamental acoustic properties such as sound velocities, elastic constants, and their temperature coefficients remain unclear.

In this study, we directly measure the longitudinal-wave velocity v_{33} of $(MgZr)_xAl_{1-x}N$ and $(MgHf)_xAl_{1-x}N$ thin film between 10 and 300 K and deduce their temperature coefficients using picosecond ultrasound spectroscopy. The sound velocity governs resonant frequencies of FBARs and temperature coefficient of sound velocity (TCV) determines the sensitivity of the resonant frequency to temperature. Especially, out-of-plane elastic constant (C_{33}) is the most important elastic constant for FBARs because they use the through-thickness resonant mode. However, accurate measurements of C_{33} for a thin film are very difficult and never straightforward, and it is usually different from the bulk value. Therefore, it is important to directly measure v_{33} and TCV for a thin-film AlN based materials.

2. Experiment

We deposited a pure AlN thin film, two $(MgZr)_xAl_{1-x}N$ thin films, and three $(MgHf)_xAl_{1-x}N$ thin films on Si (100) using radio frequency reactive spattering method, whose film thicknesses are about 1 μm. The content ratio of MgZr or MgHf is between 5.8 and 12.6 at.%.

We developed the optics for the picosecond ultrasound spectroscopy measurements at cryogenic temperature and its detail appears elsewhere,6) and observed Brillouin oscillation, whose frequency f is well-approximately written by the Bragg's diffraction low: 7) $f = 2nv_{33}/\lambda$, where n and v_{33} are the refractive index and sound velocity of a specimen, and λ is the wavelength of probe light (400 nm). We measured n by ellipsometry method using an instrument V-VASE produced by J. A. Woollam Co. Therefore, we can determine v_{33} by measuring f.

4. Results and discussion

We show a typical Brillouin-oscillation signal for the pure AlN thin-film specimen in **Fig. 1 (a)**, where thermal-decay background was subtracted using a polynomial function. The initial small-amplitude and low-attenuation oscillation is the Brillouin oscillation of the AlN thin film and the subsequent high-frequency, larger-amplitude and higher-attenuation oscillation is that of Si substrate, reflecting higher refractive index, larger piezo-optic constant, and higher extinction coefficient of Si, respectively. We also succeeded in observing pulse-echo signals around 240 and 470 ps, but they

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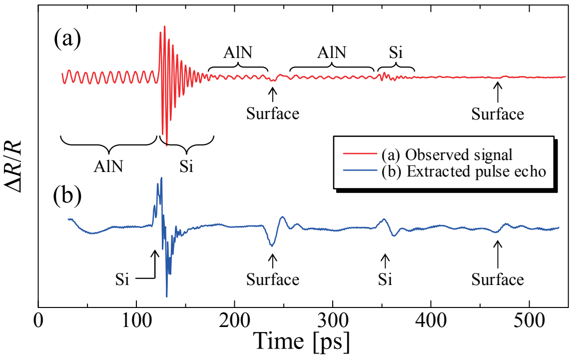

Fig. 1 (a) Observed signal for the pure specimen where thermal decay was subtracted using a polynomial function and (b) extracted pulse echo using equation (1).

are small and buried in the Brillouin oscillations. Therefore, we convert the raw data $S(t)$ into $S'(t)$ through the following equation:[8)]

$$S'(t) = \frac{1}{4}S\left(t+\frac{\tau}{2}\right)+\frac{1}{2}S(t)+\frac{1}{4}S\left(t-\frac{\tau}{2}\right) \qquad (1)$$

where τ is the period of a Brillouin oscillation. Using this equation, we can remove the oscillation component of period τ. To remove both Brillouin oscillations in AlN and Si, we applied equation (1) twice for τ_{AlN} and τ_{Si}, and obtained the Brillouin-oscillation-free signal as shown in Fig. 1 (b). This signal processing makes the pulse-echo signals clearly visible not only at the surface but also at the interface of AlN/Si, yielding the round-trip time of the strain pulse, through which film thickness or TCV is determined without refractive index. Thermo-optic coefficient dn/dT of AlN (38 ppm/K) is larger than thermal expansion coefficient (5.3 ppm/K) and would be more sensitive to the doped AlN.

All films have *c*-axis orientation, yielding measurable elastic constants C_{33}. We succeeded in measuring Brillouin-oscillation frequency and its standard deviations are less than 0.39%, and we show obtained C_{33} in **Fig. 2**. Reported C_{33} of pure AlN is 402 GPa,[9)] showing a good agreement with our measurement value 398.1 GPa. We found that C_{33} of co-doped AlN thin films largely decreased (down to 80%) and their stiffness decrease is more remarkable in the MgZr films than the MgHf films.

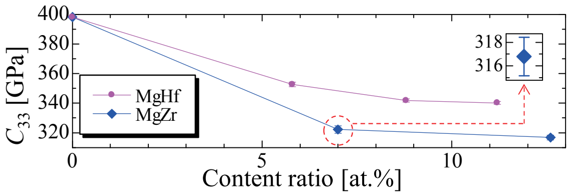

Fig. 2 Measured elastic constants from Brillouin oscillation at room temperature. Error bars are smaller than the symbols.

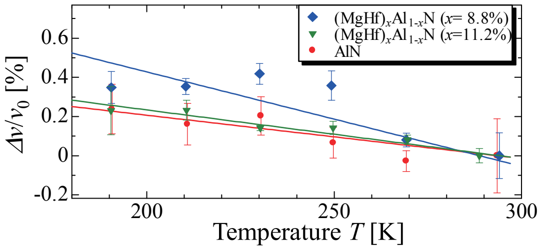

Fig. 3 Temperature dependence of v_{33} determined from pulse-echo signals.

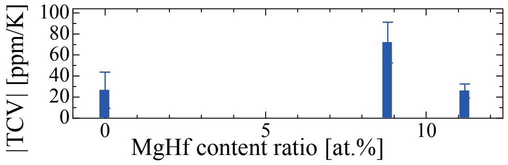

Fig. 4 The absolute values of TCV of (MgHf)$_x$Al$_{1-x}$N thin films determined from pulse-echo method.

Finally, we measured temperature dependence of v_{33} from pulse-echo signals as shown in **Fig. 3** and we show determined TCV of (MgHf)$_x$Al$_{1-x}$N in **Fig. 4**. We found that TCV of AlN based thin films is so small that measurements error prevented determining reliable TCV values of other 3 specimens. Temperature coefficient of Brillouin oscillation is even smaller than TCV from pulse echo, insisting that thermo-optic coefficient could not be neglected. If we can preciously measure the TCV from pulse echo using thinner specimens, for example, this method can determine the temperature dependence of refractive index at the same time.

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