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Elastic constants c_{11} and c_{66} in $Sc_xAl_{1-x}N$ films determined by Brillouin scattering method

Brillouin 光散乱法による ScAlN 薄膜の弾性定数 c_{11} と c_{66} の Sc 濃度依存性

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1. Introduction

In recent years, significant enhancement of piezoelectricity was experimentally found in $Sc_{0.43}Al_{0.67}N$ film by Akiyama et al..[1)]

Large piezoelectricity in ScAlN films are attractive for the BAW and SAW resonators. Elastic properties of the films are important for BAW and SAW modeling. Therefore, the c_{33} and $(c_{33}^D/\rho)^{1/2}$ have been actually investigated using BAW resonators. Matloub et al. investigated the longitudinal wave velocity [$v = (c_{33}^D/\rho)^{1/2}$] in a $Sc_{0.12}Al_{0.88}N$ film bulk acoustic resonator (FBAR), and they show that the velocity (10300 m/s) was lower that of an AlN single crystal (11132 m/s).[2,3)] Moreira et al. investigated the elastic constants c_{33}^E in $Sc_xAl_{1-x}N$ FBARs ($0 \leqq x \leqq 0.15$), and indicated that the elastic constants decreased with increasing of Sc concentration.[4)] We also previously investigated the longitudinal wave velocities $v = (c_{33}^D/\rho)^{1/2}$ in the $Sc_xAl_{1-x}N$ high-overtone bulk acoustic resonators (HBARs) ($0 \leqq x \leqq 0.63$).[5)] In the investigations, the wave velocities increased with increasing of Sc concentrations of x >0.5. However, other elastic components such as c_{11} and c_{66} have not been investigated yet.

In this study, we investigated longitudinal and shear wave velocities [$v_L = (c_{11}/\rho)^{1/2}$ and $v_S = (c_{66}/\rho)^{1/2}$] in $Sc_xAl_{1-x}N$ films ($0 \leqq x \leqq 0.63$). These wave velocities were determined by Brillouin scattering method. In addition, we estimated the elastic constants c_{11} and c_{66} using the mass density of an AlN single crystal.

2. ScAlN film samples

$Sc_xAl_{1-x}N$ films were prepared using a conventional RF magnetron sputtering system. All (0001) ScAlN films (4 – 5 μm) were deposited on a (0001) Ti film (90 – 250 nm) on a silica glass substrate (25 × 50 × 0.5 mm^3). Sc/Al atomic concentration ratios were determined using an energy dispersion x-ray spectroscopy (JSM-7001FF, JEOL Ltd.). The crystal orientations were estimated using an x-ray diffraction analysis (X-pert Pro MRD, Philips).

3. Brillouin scattering measurement

The Brillouin scattering measurement system is shown in **Fig. 1**. The six-pass tandem Fabry-Pérot interferometer (JRS Scientific Instruments) and Ar ion laser (Innova-304, Coherent Inc., 514.5 nm) were used in this system. The laser power was 230 mW near the samples. The diameter of focused laser beam was approximately 50 μm on the samples. The temperature of samples increased to 33 – 34 ºC from 26 – 27 ºC (room temperature) by the laser beam irradiation. This temperature increase was measured by a thermocouple. The scattered light was detected using a photomultiplier (R464S, Hamamatsu Photonics). As shown in Fig. 1, the reflection-induced ΘA (RIΘA) scattering geometry was adopted to measure the longitudinal and shear wave velocities propagating in-plane direction simultaneously.[6)] The Ti bottom films were used as the optical reflector. The incident angle of the laser beam was set at 41º. The typical spectra observed for the films with Sc concentrations of 11, 41 and 63 % are shown in **Fig. 2**. From the frequency shifts of these Brillouin peaks, longitudinal or shear wave velocities $v^{\Theta A}_{(L,S)}$ are given by

$$v^{\Theta A}_{(L,S)} = f^{\Theta A}_{(L,S)} \frac{\lambda_i}{2\sin(\Theta'/2)}, \quad (1)$$

where $f^{\Theta A}_{(L,S)}$ is the shift frequency, λ_i is the wave length of the incident laser beam and Θ' is the scattering angle. We determined the shift frequencies by fitting with Voigt function. The scattering angles were calibrated by measuring the shift frequencies of the Brillouin peaks for standard a silica glass plate sample (5957 m/s at 23 ºC; ED-B, Tosho Corp.).[7)]

4. Result and discussion

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The measured longitudinal and shear wave velocities [$v_L = (c_{11}/\rho)^{1/2}$ and $v_S = (c_{66}/\rho)^{1/2}$] in $Sc_xAl_{1-x}N$ films are shown in **Fig. 3**. The both longitudinal and shear wave velocities decreased with Sc concentration in the Sc concentrations of x<0.5. On the other hand, the shear wave velocities in the Sc concentrations of x>0.5 seemed to increase. These results show similar tendencies with the longitudinal wave velocities $v = (c_{33}/\rho)^{1/2}$.[5)] However, as shown in Fig. 2 (c), we could not measure the longitudinal wave velocities in the Sc concentrations of x>0.47 owing to very weak Brillouin scattered light.

Next, the estimated elastic constants of $c_{11} = \rho v_L^2$ and $c_{66} = \rho v_S^2$ in $Sc_xAl_{1-x}N$ films are shown in **Fig. 4**. The theoretical values reported by Zhang et al. are also plotted.[8)] Assuming that the mass densities hardly change, the mass density (3260 kg/m^3) of an AlN single crystal was used for the estimations.[3)] As shown in Fig. 4, the estimated values were higher than the theoretical values. In addition, the decreasing rates of estimated values were similar to those of theoretical values.

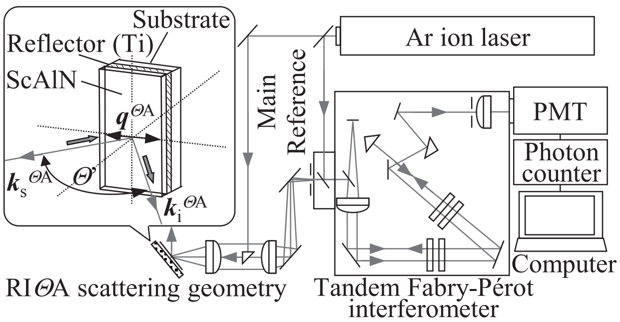

Fig. 1 Brillouin scatteriung measurement system and reflection-induced ΘA scattering geometry. $\boldsymbol{k}_i^{\Theta A}$ and $\boldsymbol{k}_s^{\Theta A}$ are the wave vectors of incident and scattered lights. $\boldsymbol{q}^{\Theta A}$ is the wave vector of the acoustic wave. Θ' is the scattering angle.

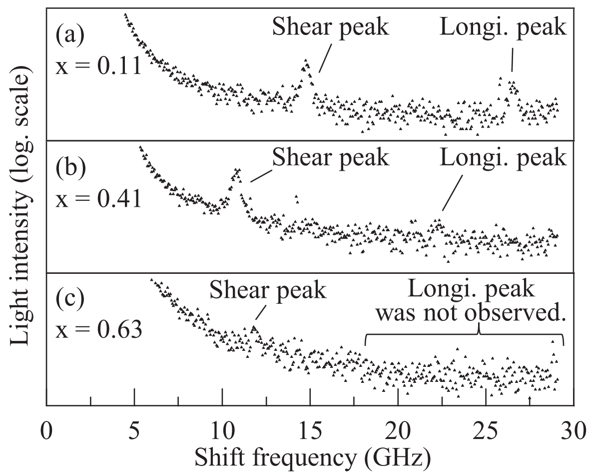

Fig. 2 The measured Brilllouin spectra from the $Sc_xAl_{1-x}N$ films with Sc concentrations x of (a) 0.11, (b) 0.41 and (c) 0.63. The Brillouin peak intensities decreased with Sc concentration.

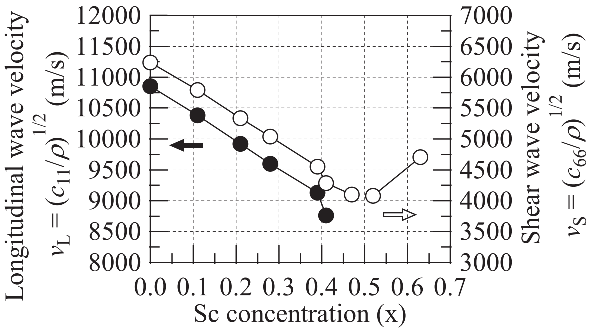

Fig. 3 The longitudinal and shear wave velocities [$v_L = (c_{11}/\rho)^{1/2}$ and $v_S = (c_{66}/\rho)^{1/2}$] as a function of Sc concentration x in the $Sc_xAl_{1-x}N$ films.

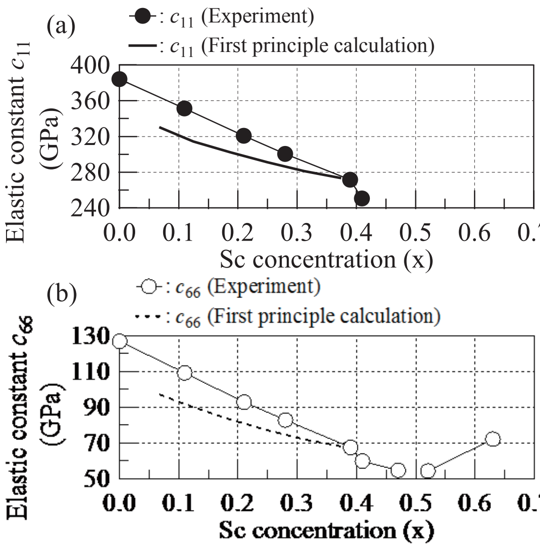

Fig. 4 The estimated elastic constants (a) c_{11} and (b) c_{66} as a function of Sc concentration x in the $Sc_xAl_{1-x}N$ films. The theoretical values reported by Zhang et al. are also plotted in this graph.[8)]

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