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Oblique incidence ultrasonic reflectometry device based on c-axis tilted ScAlN films for evaluating viscoelastic properties of liquids above 100 MHz

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ABSTRACT

Ultrasound-based evaluation of fluid properties allows for real-time measurement of small amounts of liquid samples. The ultrasonic reflection method is used to obtain the complex reflection coefficient, which can be used to evaluate the viscoelastic properties of liquids. However, this method has not been used with shear waves at frequencies above 100 MHz because shear-mode piezoelectric films are difficult to obtain at such frequencies. We propose using the oblique incidence reflectometry with quasi-shear waves excited by c-axis tilted scandium aluminum nitride (ScAlN) thin films to realize high-sensitivity evaluation of the viscoelastic properties of liquids in the above 100 MHz. In experiments, the shear elasticity and shear viscosity of glycerin solutions were estimated from their complex reflection coefficients.

1. Introduction

Ultrasound-based evaluation of fluid properties allows for the real-time measurement of small liquid samples. This technique has attracted attention in the medical field for applications such as blood tests, where the fluid viscoelasticity may be used to detect diseases. The ultrasonic reflection method [1], [2] is used to obtain the complex reflection coefficient of a viscoelastic liquid, which can be used to evaluate viscoelastic properties such as the

complex acoustic impedance, shear modulus, and shear viscosity. Although many studies have used this method [3]-[6], shear waves at frequencies above 100 MHz have not been considered because shear-mode piezoelectric films are difficult to obtain in this frequency range. In this study, we used the oblique incidence reflectometry with quasi-shear mode c-axis tilted scandium aluminum nitride (ScAlN) thin films to realize the high-sensitivity evaluation of the viscoelastic properties of liquids at frequencies above 100 MHz. Experiments were performed with glycerin solutions to demonstrate the performance of the proposed method.

2. c-Axis tilted ScAlN

Static piezoelectric constant d_{33} in AlN films is increased by Sc doping [7]. We have previously reported, for the first-time, the experimental demonstration of electromechanical coupling coefficient of thickness-extensional mode k_t^2 ($k_t^2=15\%$) in the c-axis oriented ScAlN film bulk acoustic wave resonator (FBAR), and that of thickness quasi-shear mode k'_{35^2} ($k'_{35^2}=10\%$) in the c-axis tilted ScAlN FBAR [8]. The theoretical electromechanical coupling coefficient of quasi thickness shear mode (k'_{35^2}) and that of quasi thickness extensional mode (k'_{33^2}) in ScAlN and AlN were calculated using Caro's constants [9] as a function of c-axis tilt angle. The maximum k'_{35^2} for $\text{Sc}_{0.45}\text{Al}_{0.55}\text{N}$ is much larger than that of AlN. k'_{35} reaches maximum at c-axis tilt angle of 35° .

3. Measurement principle

The principle of the proposed method can be described as follows. The complex reflection coefficient Γ^* at the interface between a substrate and liquid is given by

$$\Gamma^* = \frac{Z_S^* \cos \phi - Z_L^* \cos \psi}{Z_S^* \cos \phi + Z_L^* \cos \psi} \quad (1)$$

where Z_S^* , Z_L^* , ϕ , and ψ are the complex acoustic impedances of the substrate and liquid and the angles of incidence

and refraction, respectively, as shown in **Fig. 1**. If $\Gamma^* = \Gamma e^{-j\theta}$, then Z_L^* is given by

$$Z_L^* = \frac{\cos \phi}{\cos \psi} \frac{1 - \Gamma^2 + j2\Gamma \sin \theta}{1 + \Gamma^2 + 2\Gamma \cos \theta} Z_S^* \quad (2)$$

If $|Z_S^*| \gg |Z_L^*|$, then $\cos \psi = 1$. In this study, because the incident angle of the trapezoidal silica glass prism

was 60° ($\cos \phi = 0.5$), the measurement was twice as sensitive as that based on normal incidence ($\cos \phi = 1$). The

theoretical Z_L^* is given by

$$Z_L^* = R_L + jX_L = \sqrt{\rho_L (c_{44L} + j\omega\eta_L)} \quad (3)$$

where ω , ρ , c_{44L} , and η are the angular frequency and the density, shear modulus, and viscosity of the fluid,

respectively. c_{44L} and η are given by

$$c_{44L} = \frac{R_L^2 - X_L^2}{\rho_L} \quad (4)$$

and

$$\eta_L = \frac{2R_L X_L}{\omega \rho_L} \quad (5)$$

Fig. 1 Schematic of the trapezoidal silica glass prism. Also shown is the SH wave incidence at the interface between the substrate and fluid.

4. Methods

4.1 Fabrication of the trapezoidal silica glass prism

A trapezoidal silica glass prism was used to realize oblique incidence of the SH wave. **Figure 1** shows the schematic of the prism (top electrode: Au, piezoelectric film: c-axis tilted ScAlN, bottom electrode: Al, substrate: silica glass). The c-axis was tilted in the direction parallel to the interface to minimize mode conversion. The c-axis tilted ScAlN thin film was grown by glancing angle RF magnetron sputtering [10]. c-Axis tilt angle of 33° was confirmed by XRD pole figure analysis.

4.2 Analysis with a network analyzer

The experimental system used in this study is shown in **Fig. 1**. The n^{th} echo S_{nL} , which reflects at the interface between the substrate and liquid, was obtained from multiple-reflection data with a network analyzer (Agilent Technologies, E5071C). The n^{th} echo S_{nA} at the interface between the substrate and air was also obtained. The complex reflection coefficient Γ^* is given by

$$\Gamma^* = \left(\frac{S_{nL}^*}{S_{nA}^*} \right)^{\frac{1}{n}}. \quad (6)$$

The complex acoustic impedance of the liquid Z_L^* can be determined by using Γ^* in Eq. (6) and the known Z_S^* . In this study, six glycerin solutions with different concentrations (0, 20, 40, 60, 80, and 97 wt.%) were prepared as the liquid samples. The experiment was performed at 25 °C in a thermostatic chamber. The temperature of the liquid was monitored with a platinum thermometer.

5. Results and discussion

The properties of silica glass [11] and the glycerin solutions [12] were used to calculate Z_S^* , c_{44} and η , where the substrate was assumed to be a purely elastic material ($\eta = 0$). **Figure 2** shows the time domain response of the trapezoidal silica glass prism. The SH wave from the piezoelectric film was observed to propagate along the route abcb (88 mm) in **Fig. 1**. The peaks every $23.4 \mu\text{s}$ in **Fig. 2** were those of the SH wave because the shear wave velocity of silica glass is 3768.9 m/s [11]. **Fig. 3** shows the frequency domain response of the insertion loss obtained by gating and Fourier transforming the first echo SH_1 . The efficiency of the SH wave excitation was highest at approximately 143 MHz.

The shear wave conversion loss [13] is measured to investigate $k'_{35}{}^2$ as shown in **Fig. 4**. The $k'_{35}{}^2$ of c-axis tilted ScAlN film is determined to be 15.8% by comparing the experimental and theoretical conversion loss curves simulated by the electromechanical transmission line model (Mason's equivalent circuit model).

Fig. 2 Time response characteristics of the trapezoidal silica glass prism.

Fig. 3 Frequency characteristics for the insertion loss of the trapezoidal silica glass prism obtained by gating and Fourier transforming the first echo SH_1 .

Fig. 4 Experimental shear wave conversion loss curves of c-axis tilted ScAlN films (red plot), which are in good agreement with the theoretical shear wave conversion loss curves (blue line) simulated by the electromechanical transmission line model (Mason's equivalent circuit model).

Figure 5 shows the measurement results for the shear elasticity c_{44} and shear viscosity η with the oblique incidence reflectometry. **Figure 5 (a)** shows that the amplitude of Γ^* decreased monotonically with increasing glycerin concentration. However, **Fig. 5 (b)** shows a considerable variation in the phase of Γ^* , particularly for the 20 and 40 wt.% glycerin solutions. This irregularity affected the calculations of the shear modulus and shear viscosity, as shown in **Fig. 5 (c)** and **(d)**. The trapezoidal prism was so large that the propagation distance increased, and the temperature control of the entire prism was difficult, which caused errors in the result.

Next, we prepared smaller prism device to avoid these errors. The smaller prism is 6.4% volume of the previous

one. As shown in **Fig. 5**, variation in the phase of Γ^* were improved when smaller-type prism was used. This result in more precise measurement of the c_{44} and η .

Fig. 5 Experimental results of the (a) amplitude and (b) phase of the complex reflection coefficient measured for the glycerin solutions. Calculated (c) shear elasticity and (d) shear viscosity of glycerin solutions based on the complex reflection coefficients.

6. Conclusion

We used the complex reflection coefficient obtained with the oblique incidence reflectometry to measure the viscoelastic properties of a liquid. Measurements based on SH waves at frequencies above 100 MHz were achieved with c-axis tilted ScAlN films.

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Data availability

The datasets in the current study are available from the corresponding author on reasonable request.

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Author Contributions

S. Kinoshita performed the data analysis, T. Yanagitani supervised the work. S. Kinoshita and T. Yanagitani prepared the manuscript.

Competing Interests

The authors declare no competing interests.

Additional information

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Figures

Figure 1

Schematic of the trapezoidal silica glass prism. Also shown is the SH wave incidence at the interface between the substrate and fluid.

Figure 2

Time response characteristics of the trapezoidal silica glass prism.

Figure 3

Frequency characteristics for the insertion loss of the trapezoidal silica glass prism obtained by gating and Fourier transforming the first echo SH1.

Figure 4

Experimental shear wave conversion loss curves of c-axis tilted ScAlN films (red plot), which are in good agreement with the theoretical shear wave conversion loss curves (blue line) simulated by the electromechanical transmission line model (Mason's equivalent circuit model).

Figure 5

Experimental results of the (a) amplitude and (b) phase of the complex reflection coefficient measured for the glycerin solutions. Calculated (c) shear elasticity and (d) shear viscosity of glycerin solutions based

on the complex reflection coefficients.

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