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Acoustic properties of fluorine-doped silica-glass thin films at low temperatures

低温域におけるフッ素添加石英ガラス薄膜の音響特性

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1. Introduction

Silica glass (SiO_2) shows many anomalous properties, including positive temperature dependences of the principal elastic constants (i.e. $dC_{ij}/dT < 0$): Many materials show negative temperature dependences. The elastic constants of silica glass decrease, take minimal values around 100 K, and increase as temperature decreases. Acoustic attenuations take maximum values around 100 K.[1)] These behaviors do not appear in the crystalline SiO_2 (α-quartz).

Because of its reversed temperature dependences of the elastic constants, silica glass has been used in many devices. For example, it is used to make the resonant frequencies of acoustic devices insensitive to temperature: The sound velocities of usual materials decrease as temperature increases, leading to the changes in transmitting frequencies of acoustic devices. To compensate for the temperature effects, silica-glass thin films are used with its reversed temperature dependences of the elastic constants. Thus, it is important to control the temperature dependence of sound velocity in silica glass. One candidate approach is adding other materials such as F, TiO_2, and so on.[2)]

It is, however, difficult to measure the acoustic properties of a deposited silica-glass thin film with conventional methods because the film thickness is less than sub-micron. Therefore, we use picosecond ultrasonic spectroscopy and measure the longitudinal-wave velocity and attenuation of silica-glass thin films between 20 and 300 K. Picosecond ultrasonic spectroscopy can excite and detect sub-terahertz ultrasound and accurately measure acoustic properties of a thin film.[3)] Moreover, we developed multi-incident-angle optics to measure the refractive index of the silica-glass thin films using picosecond ultrasonics spectroscopy.

2. Experiments

We deposited several type silica-glass thin films on Si substrates with various contents of fluorine between 0 and 8.8 atom%. Figure 1 shows the optics developed for low-temperature measurements with normal incidence. We used a cryostat as a specimen holder and applied the light pulses to the specimen through a glass window. The specimen was attached on a Cu heat exchanger cooled by liquid He, and we used a semiconductor thermometer attached to the heat exchanger to measure the specimen temperature.

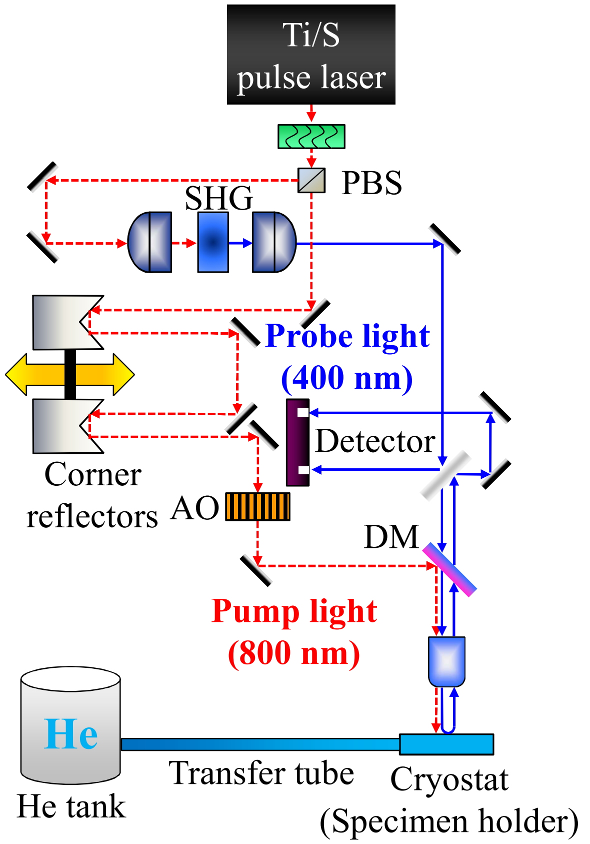

Fig. 1 Schematic of the optics we developed. Solid and dashed lines show 400-nm and 800-nm wavelength light, respectively.

We deposited a 10 nm Al thin film on each specimen surface, which absorbs the pump light energy for excitation of ultrasound. The time-delayed probe light pulse is reflected at the surface, but a part of the probe light is transmitted inside the specimen. The transmitted probe light is diffracted backwardly via the piezo-optic effect, which causes interference with the surface-reflected light, resulting in an oscillating signal in the

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reflected probe light as the ultrasound propagates. This oscillation signal is called the Brillouin oscillation, and its frequency f relates to the sound velocity via Bragg's condition

$$f = \frac{2v}{\lambda}\sqrt{n^2 - \sin^2\theta} \tag{1}$$

where λ is the wavelength of the probe light, v is the sound velocity, n is the refractive index, and θ is the incident angle from normal. We can then determine v and n through multi-angle measurements: Measurements of Brillouin-oscillation frequencies f_1 and f_2 at two different incident angles θ_1 and θ_2 yield n and v as follow:

$$n = \sqrt{\frac{f_2^2 \sin^2\theta_1 - f_1^2 \sin^2\theta_2}{f_2^2 - f_1^2}} \tag{2}$$

$$v = \frac{\lambda}{2}\sqrt{\frac{f_2^2 - f_1^2}{\sin^2\theta_1 - \sin^2\theta_2}} \tag{3}.$$

This multi-angle method enables us to measure n and v simultaneously without any other methods such as ellipsometry or thickness measurement.[4)]

3. Results and Discussion

Firstly, we measured the sound velocity and refractive index of a bulk silica-glass specimen at room temperature. Incident angles are 0 and 68.5 degree. Here, we calibrated the measurement angle using a reference specimen of a $SrTiO_3$ [100] single crystal, whose refractive index is accurately measured with ellipsometry. As shown in Fig. 2, we clearly observed different frequency Brillouin oscillations, and they lead to n=1.495 and v=5898 m/s.

Then, we measured temperature dependence of the longitudinal-wave velocity and refractive index of fluorinated silica-glass thin films. Figure 3 shows one of typical Brillouin oscillation at 19 K. The ultrasonic wave entered the Si substrate at 90

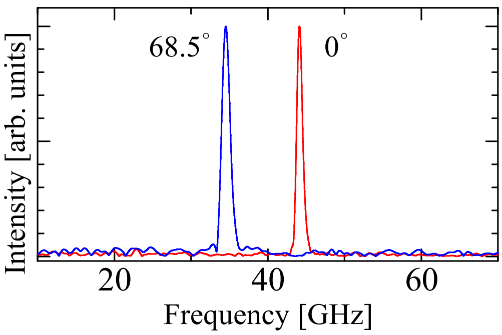

Fig. 2 Fast Fourier transformation spectra of a silica-glass substrate at incident angle 68.5(left) and 0(right) degree.

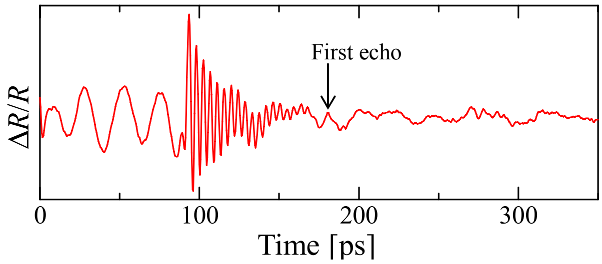

Fig. 3 Typical Brillouin oscillation of silica-glass thin film on Si substrate. The incident angle is 0 degree and temperature is 19 K.

ps, and the observed oscillation suddenly changed. The frequency of the former oscillation leads to the sound velocity and refractive index of silica glass, and the amplitude of Brillouin oscillation in Si substrate reflects the attenuation of higher-frequency components in the silica glass. We found that temperature coefficient of the longitudinal-wave elastic constant (dC_{11}/dT) of fluorinated silica-glass thin film increases with the fluorine fraction.

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