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Elastic Constants and Microstructure of Amorphous SiO_2 Thin Films Studied by Brillouin Oscillations

Hirotsugu OGI [1], Tomohiro SHAGAWA [1], Nobutomo NAKAMURA [1], Masahiko Hirao [1], Hidefumi ODAKA [2], and Naoto KIHARA [2]

[1] Graduate School of Engineering Science, Osaka University
Toyonaka Osaka 560-8531, Japan; e-mail:ogi@me.es.osaka-u.ac.jp
[2] Research Center, Asahi Glass Co., LTD
Yokohama, Kanagawa 221-8755, Japan

Abstract
The elastic constants of amorphous SiO_2 (*a*-SiO_2) thin films deposited on Si substrate were measured by the pump-probe picosecond ultrasounds with ellipsometry for refractive index, and their correlation with the microstructure is discussed. The pump pulse irradiated the aluminium ultrathin film on the oxide film for generating the longitudinal wave in the oxide film toward the Si substrate. The delayed probe pulse entered the specimen and it was diffracted by the ultrasonic wave in the film and substrate, causing oscillations (Brillouin oscillations) in the intensity of the reflected probe pulse. The oscillation frequency yielded the longitudinal-wave velocity and the elastic constant. The elastic constant shows a correlation with the microstructure of the film: it decreases as the thin film becomes rough. The effects of the defect volume fraction and aspect ratio on the elastic constant are analyzed by micromechanics modeling.
Keywords: Picosecond ultrasounds, Brillouin oscillation, SiO_2, Thin films, Elastic constant, Porosity

1. Introduction

Thomsen *et al.* [1, 2] have first detected high-frequency coherent acoustic phonons using ultrafast pump-probe light pulses. Following their work, significant efforts have been paid for establishing the picosecond ultrasounds techniques for the study of ultrahigh-frequency acoustic properties of thin films [3-11]. Brillouin oscillation is recently adopted for materials characterization of transparent or translucent thin films [5, 8], which arises from the interference between the light reflected at the specimen surface and the light refracted by the acoustic wave propagating in the thin film and substrate. Intensive studies have been made by Devos *et al.* [5, 8] for ultrahigh-frequency (~240 GHz) attenuation of transparent materials using the Brillouin oscillation in the Si substrate. On the other hand, the elastic constants and their correlation with the microstructure still remain unclear. Here, we measure the elastic constant of SiO_2 thin films deposited on Si substrates using Brillouin oscillations and present a methodology for microstructure characterization of transparent thin films.

2. Measurement

The ultrasonic wave excited by the pump light pulse propagates in the film thickness direction, and the delayed probe light pulse is diffracted by the ultrasonic wave because of the photoelastic effect. The diffracted light causes interference with the reflected light pulse at the film surface, which causes the Brillouin oscillation in the intensity of the reflected light pulse. The Brillouin-oscillation frequency f is approximately expressed by

$$f = \frac{2nv}{\lambda}, \tag{1}$$

when the probe pulse perpendicularly enters the specimen surface. n, v, and λ denote the refractive index, sound velocity, and the wavelength of the probe light pulse in vacuum, respectively. Thus, we can determine the sound velocity if we know n. We measured n by the ellipsometry for thin films assuming a homogeneous film/substrate model [12].

We deposited SiO_2 thin films on the (001) face of the Si substrate using a DC sputtering method. The target was Si and the gas was a mixture of Ar and O_2. The sputtering parameters, such as pressure, DC voltage, and substrate temperature, were varied to develop various microstructures. The total thickness was about 1 μm. We observed their cross-section microstructures by the field-emission electron microscopy (SEM), studied their crystallinity by the X-ray diffraction (XRD) measurement, and measured their refractive index by the ellipsometry. Furthermore, we added 10-nm Al films on the SiO_2 films for generating the ultrasonic waves by the pump light pulse through thermal expansion.

Figure 1 shows our optics [6, 7]. We used a mode-locking titanium-sapphire pulse laser with 100-fs pulse width, 0.7-W power, 80 MHz repetition frequency, and 800 nm wavelength. The light pulse was first split into two pulses by a polarization beam splitter (PBS). The straight-through output of the PBS was used for the pump pulse (800 nm): it was reflected by a corner reflector located on a microstage, which changed the light path of the pump pulse. Its was then modulated by an acoustic-optic (A/O) modulator with 100-kHz modulation frequency, and focused on the specimen surface by the objective lens. Its power at the specimen surface was about 20 mW. The pump pulse generated the longitudinal wave, which propagated in the thickness direction and penetrated into the Si substrate.

The other output of the PBS were frequency-doubled (400 nm) by a second-harmonic-generator (SHG) crystal, and it was further split into two beams by the beam splitter (BS): one entered the balanced photo detector to produce the reference signal and the other irradiated the specimen surface as the probe pulse. The reflected probe light

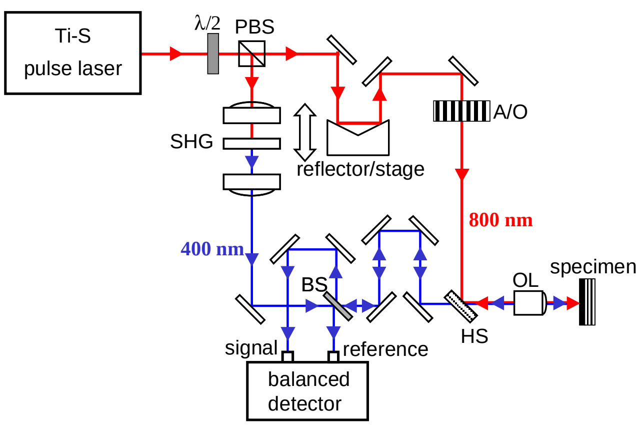

Figure 1. Optics for the pump-probe detection of Brillouin oscillations.

entered the other input of the photo detector. Their difference output was processed by a lock-in amplifier to extract the amplitude of the modulation frequency. By changing the path length of the pump light, the time delay between the pump and probe lights was changed. Thus, changing the microstage and acquiring the reflectivity change, we can detect Brillouin oscillations.

2. Results and Discussions

We prepared six specimens showing various microstructures and named them A to F; the microstructure was rough from A to F in order. Figure 2 shows cross-section microstructures of three of them. All specimens showed a broad halo peak without any crystallographic peaks except for the (004)Si peak in their XRD spectra, confirming that they show the amorphous phase. Figure 3 shows the reflectivity changes measured by the pump-probe method (black lines). The numerical simulation was also made following Thomsen *et al.* [2], which showed good agreement with the measurements (red lines). We thus calculated the reflectivity changes assuming various parameters (piezo-optic constants, film thickness, pulse width and so on), and determined the elastic constant using the FFT spectrum of the response from the film. This procedure confirmed that Eq. (1) was applicable to the determination of the elastic constants of the thin films with in 1% error when the film thickness was larger than 400 nm.

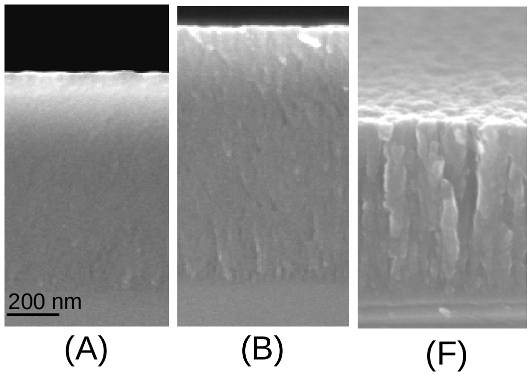

Figure 2. Cross-section microstructures of specimens A, B, and F.

We have observed the Brillouin oscillations from Si only for the smoothest specimen (A) as shown in Fig. 3, indicating high attenuation probably due to scattering by defects in the other specimens. The Brillouin-oscillation frequency in Si was 235 GHz, which

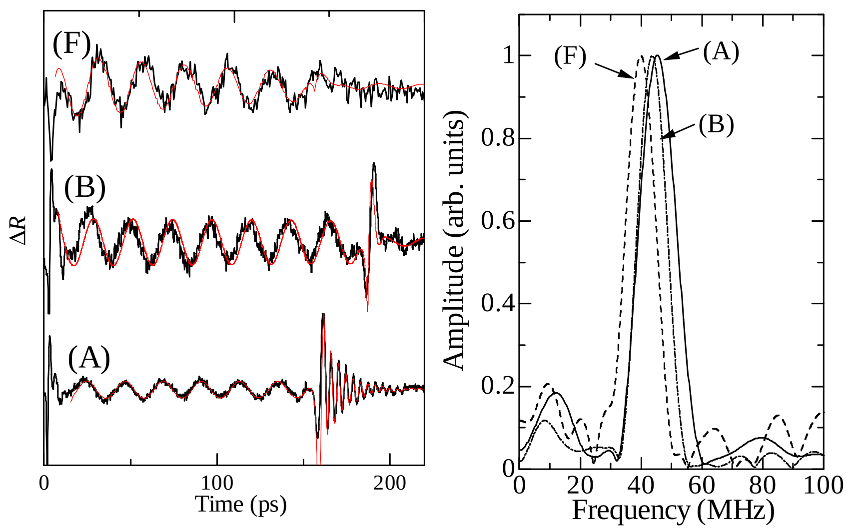

Figure 3. Measured (black lines) and calculated (red lines) reflectivity changes of specimens A, B, and F (left), and their corresponding Fourier spectra (right).

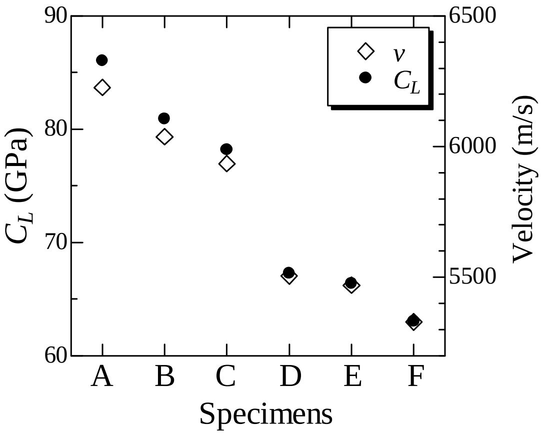

Figure 4. Elastic constant and sound velocity of *a*-SiO_2 thin films.

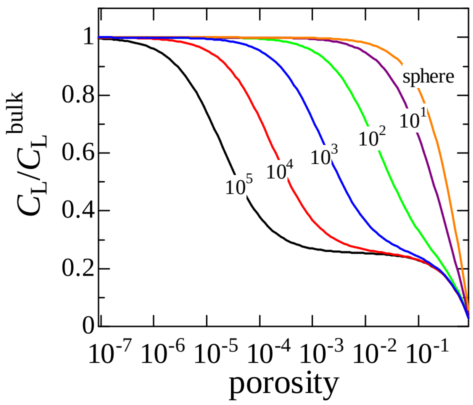

Figure 5. Change in the elastic constant C_L caused by defects. The numbers denote the aspect ratio of the defect.

well agreed with Eq. (1). Such a high-frequency component of the ultrasonic wave will be attenuated in the oxide films by scattering at the defects.

Figure 4 shows the longitudinal-wave elastic constant C_L of the *a*-SiO_2 thin films. As the specimen shows the rough microstructure, the elastic constant decreases. Thus, the elastic constant can be a measure for the reliability of the transparent and translucence thin films.

We estimated the effect of the thin defect on the macroscopic elastic constant using the micromechanics modeling, assuming that the defects distribute with random orientation in the matrix. The detailed calculation procedure appears elsewhere [9, 13]. Figure 5 shows the decrease of the elastic constant caused by the pancake-shape defect inclusions. When the aspect ratio of the defect is large, very small porosity causes significant decrease in the elastic constant. The defects in the *a*-SiO_2 thin films will appear near the boundaries of the columnar structures and *a*-SiO_2 particles, and their aspect ratio should be fairly small. Assuming a aspect ratio of 1000, for example, a 0.1% volume fraction of the defect can decrease the elastic constant by ~30%.

3. Conclusions

The amorphous SiO_2 thin films showed various microstructures depending on the sputtering condition, including rough microstructures. The elastic constant determined by the Brillouin oscillation highly correlates with the microstructure; it can be an important characteristic for evaluating the reliability of the oxide thin films. The micromechanics calculation was made to predict the effect of the defective region on the elastic constant, which showed that thin defect inclusion causes significant decrease of the elastic constant even when their volume fraction is very small.

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