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# Evaluation of Ultrasonic Attenuation in Oxide Thin Films by Brillouin Oscillation Using Strong Wavelength Dependence of Refractive Index in Si

# Si 誘電率の強い波長依存性を利用したピコ秒超音波による酸化物薄膜の減衰評価

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## 1. Introduction

Amorphous oxide thin films are promising materials for ultrahigh-frequency resonators used in bulk acoustic wave filters[1]. Because they are free from the grain-scattering loss, they show low attenuation and then achieving high-$Q$ resonators. Also they are homogenously deposited by a reactive sputtering method, and their thickness can be precisely controlled. However, their acoustic properties such as sound velocity and attenuation are highly dependent on the deposition condition and cannot be measured with conventional ultrasonic methods. Recently, the sound velocity has been determined with 1% error using Brillouin oscillation excited by picosecond ultrasound[2]. However, it has been difficult to characterize attenuation. Emery and Devos[3] proposed a method for evaluating attenuation using the intensity of Brillouin oscillation in Si substrate. However, several specimens with different film thicknesses are necessary in this method, involving problems with using different materials. Furthermore, it is theoretically shown that the attenuation value cannot be accurately determined from thin films with different thicknesses because the intensity of Brillouin oscillation in Si is significantly affected by the film thickness[2].

In this study, we propose a new method for evaluating attenuation of oxide thin films from a single specimen using Brillouin oscillation in Si substrate by changing the wavelength of the probe light pulse. The successful determination relies on the high sensitivity of the refractive index of light in Si, allowing a wide-frequency-range measurement for attenuation. We evaluate the attenuation coefficient using the phonon-phonon interaction mechanism.

## 2. Brillouin oscillations

The pump-probe picosecond ultrasound method has been developed by Thomsen *et al.*[4,5]. Irradiation of the specimen with the ultrashort light pulse causes a coherent acoustic pulse, which propagates in the film thickness direction. Then, the delayed probe light pulse detects the acoustic waves through the reflectivity change. For transparent and translucent materials, the probe light pulse is diffracted backward by the acoustic wave in the film and substrate, and its reflectivity is oscillated as the propagation of the acoustic pulse. This is called Brillion oscillation, and its oscillation frequency $f$ is approximately given by $f=2nv/\lambda$, which is equal to the detectable sound-wave frequency. Where $n$, $\lambda$ and $v$ denote the refractive index, the wavelength of the probe light in vacuum, and the sound velocity, respectively.

We construct the multiple reflection model of light in a multilayer (ref. **Fig. 1**), and calculate the reflectivity change. The detectable reflectivity change $\Delta R$ is finally given by

$$\Delta R = \left| R_N + \sum_{l=0}^{N} \Delta R_l \right|^2 - \left| R_N \right|^2 , \qquad (1)$$

where $N$ is the number of layers. $R_N$ and $\Delta R_l$ are the total reflectivity including all layers without ultrasound and the reflectivity change caused by the acoustic field in the $l$ layer, respectively.

In amorphous materials, the ultrasound attenuation is principally caused by the absorption

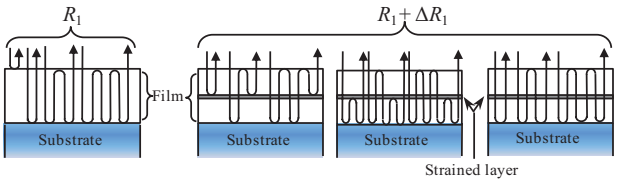


Fig. 1 The model of the multiple reflection of the light pulse in the single layer ($N$=1) without ultrasound (left) and that modified by the ultrasound strained layer (right). The gray strip indicates the strained layer where the refractive index is slightly modified by the acoustic strain, causing backscattering of the light pulses. The arrow lines show the possible light paths, considering only one reflection at the modified thin layer. The total reflectivity can be obtained by integrating the contributions of the backscattered light pulse along the thickness direction.

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loss due to phonon-phonon interactions. When the acoustic wavelength is much longer than the lattice parameter, the attenuation coefficient α can be of the form[6]

$$\alpha(\omega)=\beta\omega^2 . \qquad (2)$$

Here, ω and β indicate the angular frequency and the attenuation parameter for the absorption loss caused by the phonon-phonon relaxation process (Akhieser's relationship). It is important to determine the β value for the high frequency resonator.

The effective method to evaluate the β value is to measure the frequency dependence of attenuation. We then focus the high sensitivity of the dielectric constant of Si to the wavelength. *n* is sensitive to the wavelength of the probe light in Si. When $\lambda$ changes between 375 and 400 nm, *n* varies between 6.7 and 5.5 in Si, corresponding to the acoustic frequency change between 300 and 235 GHz. Thus, by changing the wavelength of the probe light, we can evaluate the frequency dependence of attenuation and then the β value in a single oxide thin film.

## 3. Experiments

We deposited silica thin films on the (001) surface of the Si substrate using a DC reactive sputtering method. The target was Si, and the gas was a mixture of Ar and $O_2$. The specimen was covered with a 10 nm Al thin film as a transducer material of the acoustic pulse.

The optics we developed is shown in **Fig. 2**. We used two mode-locking titanium-sapphire pulse lasers with 100 fs pulse width and 80 MHz repetition frequency. One was used for the pump light and focused on the Al film to generate the acoustic pulse through thermal expansion of Al. The other was wavelength-tunable (750-800 nm wavelength) and frequency doubled (375-400 nm wavelength), and irradiated the specimen as the probe light to detect Brillouin oscillations. We synchronized these two pulse lasers within a jittering smaller than 50 fs by the synchronization system. Thus, this optics allows to change only the probe-pulse wavelength, remaining all properties of the pump-pulse unchanged. We can therefore detect the Brillouin oscillations with various wavelength for the same sound-wave filed.

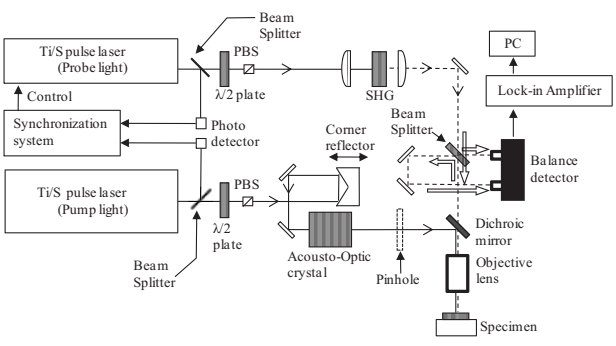


Fig. 2 Schematic of the optics. Solid lines show the pump beam (λ = 800 nm), and dashed lines show the probe beam (wavelength λ = 375-400 nm).

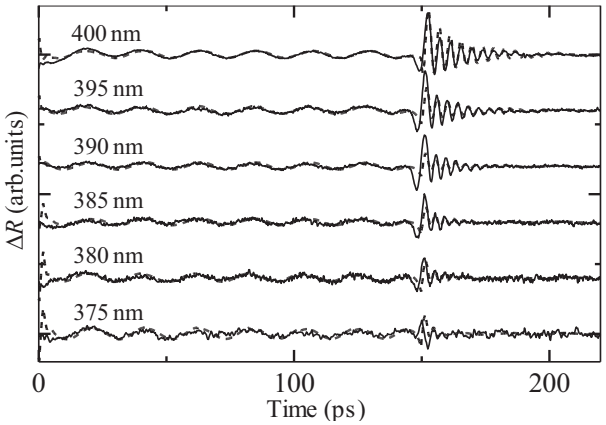


Fig. 3 Measured (solid lines) and calculated (broken lines) Brillouin oscillations of the $SiO_2$/Si specimen obtained by different probe light wavelengths.

We performed the theoretical calculation based on the model in Fig. 1 and fitted the calculated reflectivity changes to measurements with the variable parameter of β.

## 4. Results and Discussion

The solid lines in **Fig. 3** show the measured reflectivity responses, showing low-frequency and high-frequency oscillations, corresponding to Brillouin oscillations in the $SiO_2$ film and Si, respectively.

We constructed the numerical simulation of the reflectivity change including the β value as the fitting parameter. The other parameters were determined by fitting the simulation to the measurement at the wavelength of 400 nm. The results are shown in **Fig. 3** by broken lines, showing good agreements with the measurements. The determined β value was $1.1\times10^{-3}$ nm $^{-1}$ THz $^{-2}$, which was smaller than the previously reported value[3] by about 20%.

## 5. Conclusion

The newly proposed method is useful for determining the attenuation parameter of oxide thin films.

## References


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