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Elastic-Constant Measurement in Thin Films at Low Temperatures Using Picosecond Laser Ultrasound Spectroscopy
ピコ秒レーザー超音波法を用いた極低温領域における薄膜の弾性定数計測

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1. Introduction

The elastic constants of solids remain the central issue in condensed matter physics because they directly reflect the interatomic interaction. Especially, the elastic property of thin films deserves intensive study because of their unusual behaviors. We studied elasticity of thin films using picosecond laser ultrasound (PSLU) method[1-4] and theoretical analysis using *ab-initio* calculation[5,6].

In this study, we build the experimental equipments for the elastic-constant measurement of thin films at low temperatures using picosecond laser ultrasound spectroscopy. At liquid helium temperature (~4 K), essensial physical properties free from the thermal disturbance appear. In addition, the low temperature measurement is meaningful for the comparative discussion between mesurement and theoretical calculation, because *ab-initio* calculations provide the behavior of solids at the absolute zero temperature.

2. Method and Equipment

Thomsen and co-workers first detected high frequency coherent acoustic phonons using ultrafast pump-probe light pulses[7,8]. Following their work, the picosecond ultrasound techniques were developed for the study of ultrahigh frequency acoustic properties of solids. They are classified into three methods. First is the pulse-echo method[2,9,10], where the acoustic pulse generated by the pump light repeats reflections between the film surface and the film-substrate interface. The round-trip time and echo amplitudes were measured to determine the sound velocity and attenuation. Second is the phonon resonance spectroscopy[1,11,12], where the standing waves in nanostructures were measured to evaluate the elasticity through their resonance frequencies. Third is the Brillouin oscillation[3,4,8,13], which arises from interference between the light reflected at the specimen and the light refracted by the acoustic wave propagating in the transparent or translucent material. In the first-order approximation for strain, the oscillation frequency f relates with the sound velocity v via Bragg's condition when the probe light enters the film perpendicularly;

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

Here, n and λ denote the refractive index of the examined material and the wavelength of the probe light, respectively. Thus, the sound velocity is obtained by measuring the Brillouin oscillation frequency when the refractive index is known.

The pump-probe method determines the out-of-plane elastic constant from the velocity of the longitudinal wave propagating along the thickness direction. Generation and detection of the longitudinal waves are carried out by the pump-probe method with a femtosecond pulse laser. **Figure 1** shows optics we developed in this study. We irradiate a second-harmonic-generator (SHG) crystal with a titanium-sapphire pulse laser at 800 nm wavelength and 100 fs pulse width. SHG generates the frequency-doubled pulse, and then they are separated into the pump light (λ = 800 nm) and the probe light (λ = 400 nm) by a dichroic mirror. The former is focused on the specimen to generate an acoustic pulse through thermal expansion. The latter is used to detect generated acoustic pulse. The probe light is delayed by moving of the corner reflector, and the signals are detected as changes in the amplitude and phase of the reflected light. The specimen is held in the cryostat and cooled by liquid helium through the Cu heat exchanger down to 3 K.

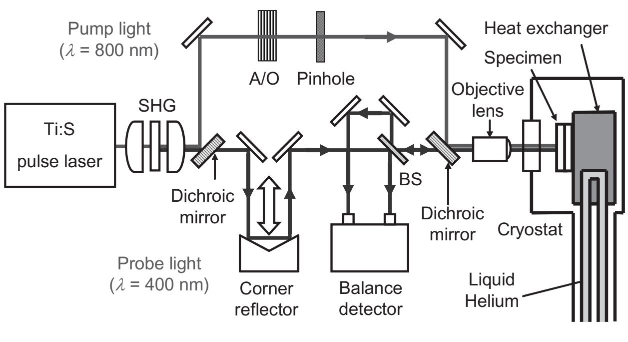

Fig.1 Developed optics for generation and detection of picosecond ultrasound at low temperatures (>~ 3K).

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3. Result and Discussion

We deposited Al thin film whose thickness is 10 nm on the (001) surface of the Si substrate. **Figure 2** shows observed reflectivity changes at room temperature and 3 K. We clearly obtained Brillouin oscillation from the Si substrate. Because the depth of penetration of the probe light influences the endurance of the Brillouin oscillation, such low damping in oscillations indicate the decrement of optical absorption of Si at low temperature.

Figure 3 shows Fourier spectra for observed Brillouin oscillations. The frequency at low temperature (broken line) is lower than at room temperature (solid line). However, it's noted that the elastic constants of solids generally increase at low temperature. The relationship between the out-of-plane elastic constant C_{33} and the frequency of the Brillouin oscillation f is given by;

$$C_{33} = \rho v^2 = \rho\left(\frac{f\lambda}{2n}\right). \qquad (2)$$

Here, ρ denote the mass density of the examined material. This decline in frequency and Eq. (2) indicate the decrease of the refractive index of Si at low temperature.

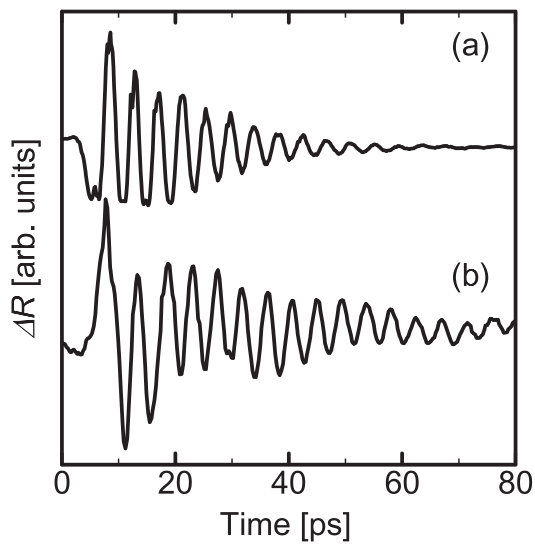

Fig.2 Reflectivity changes observed at room temperature (a) and 3 K (b).

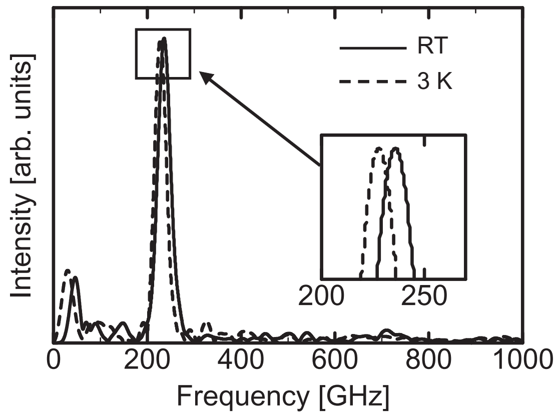

Fig.3 Fourier spectra for Brillouin oscillations observed at room temperature (solid line) and 3 K (broken line).

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