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Direct measurement of surface displacement in picosecond laser ultrasonics

ピコ秒超音波法における表面変位の直接測定

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1. Introduction

Ultrashort light pulses can generate picosecond acoustic pulses in solids[1]. This pulse generation is related to various mechanisms, for example, to the ultrafast relaxation of excited electrons[2]. So the precise measurement of the shape of the generated pulses is useful for the investigation of this ultrafast relaxation. The pulse shape can be derived from the surface displacement of the sample caused by the acoustic pulses.

To measure the surface displacement of the sample, the optical pump and probe technique can be exploited. In this technique, termed picosecond laser ultrasonics, the propagation of acoustic pulses generated by the pump light pulses is monitored as optical transient reflectance changes using delayed probe light pulses. The reflectance change is, however, generally caused by the photoelastic effect as well as by the surface displacement, and it is difficult to distinguish these two contributions.

Here we propose a method to cancel out the photoelastic effect and to obtain a signal solely determined by the surface displacement. An incident probe beam at an oblique angle produces different reflectivity changes for s and p polarizations. We propose to eliminate the photoelastic effect by interfering the reflected light beams of the s and p polarizations with an appropriate relative phase difference. The resulting beam is phase-shifted by the surface displacement. The phase shift is detected by a specially designed interferometer setup.

2. Theory

The relative reflectance change of the probe light $\delta r/r$ is a complex quantity, and is expressed as

$$\frac{\delta r}{r} \equiv \rho + i\delta\varphi, \tag{1}$$

where ρ and $\delta\varphi$ are the real and imaginary parts of the reflectance change, respectively. The former is caused only by the photoelastic effect, whereas the latter caused by surface displacement. Though the use of interferometric probing allows both real and imaginary components of $\delta r/r$ to be measured independently[3], it is difficult to measure only the surface displacement because the photoelastic contribution is frequently has a comparable strength.

One method to solve this problem is to use two optical probe beams of different polarization at oblique incidence. In the case of an opaque semi-infinite sample, the reflectance change for these polarizations is expressed as[4]

$$\frac{\delta r_s}{r_s} = iA + \alpha B\,, \qquad \frac{\delta r_p}{r_p} = iA + \beta B, \tag{2}$$

where A is a real quantity and is proportional to the surface displacement, and B is a complex quantity related to the photoelastic effect. α and β are complex constants. The difference in the reflectance change between s and p polarizations arises only through the constants α from β. It has been shown that A can be deduced from the experimentally obtainable $\delta r_s/r_s$ and $\delta r_p/r_p$.

However, the accuracy of the above mentioned method is not sufficient in practice owing to the need to take two measurements, both subject to error. To avoid this difficulty, we propose a method for the direct measurement of the surface displacement. It uses the interference between the s and p polarized beams. The total electric field amplitude is proportional to

$$r_s(1 + iA + \alpha B) + \mu r_p(1 + iA + \beta B), \tag{3}$$

where the complex constant μ includes an amplitude scaling factor and a phase shift. To cancel out the photoelastic effect, we choose μ to satisfy $\mu r_p \beta = -r_s$. Then the interfering amplitude is given by

$$(r_s + \mu r_p)(1 + iA). \tag{4}$$

Since A is the real quantity, we need to use another interferometer to detect A. By mixing reference light with a quadrature phase retardation, the total amplitude becomes

$$(r_s + \mu r_p)(1 + i + iA), \tag{5}$$

and the detected intensity becomes proportional to

$$2\left|r_s + \mu r_p\right|^2(1 + A). \tag{6}$$

Now the total intensity is directly proportional to the surface displacement $u(t)$, and the photoelastic contribution is completely eliminated.

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3. Experimental Setup

To verify the above mentioned theory, we set up a newly designed interferometer and carry out the picosecond laser ultrasonics measurement. The sample is a thin film of tungsten of thickness 220 nm formed on a crown glass plate using RF sputtering. A mode-locked Ti:sapphire laser of central wavelength 830 nm, pulse duration ~200 fs, and repetition rate 82 MHz is used for a light source. The laser beam is divided into two beams: one for the probe light and another for the pump light. The pump light generates the acoustic pulses which propagate in the depth direction in the sample. Part of the propagating acoustic pulse is reflected at the film/substrate interface, and the returning acoustic pulse is detected at the sample surface by a delayed probe light pulse as a modulation in the optical reflectance.

To realize the scenario described around Eq. (3), a mixture of s and p polarized light is used for the probe with an oblique incident angle. Using appropriate optical components, the intensity ratio and the relative phase retardation can be adjusted so that the photoelastic contribution is eliminated. Then reference light is mixed with the probe beam reflected from the sample to obtain the intensity modulation due to the surface displacement, as described around Eq. (6).

4. Results of the experiments

Typical reflectivity changes using the s and p polarized probe beam are shown in **Fig. 1**. These reflectivity changes originate solely from the photoelastic effect, and we cannot detect the surface displacement in these curves. Then using a mixture of s and p polarized light and adjusting the amplitude ratio and relative phase retardation between them, we observed the elimination of the photoelastic effect as shown in **Fig. 2**. Finally, to measure the surface displacement, we turn on the reference light beam which interferes with the probe beam. As shown in **Fig. 3**, a peak again appears around 120 ps with a slightly different shape from the one in **Fig. 1**. This originates solely from the surface displacement variation.

5. Conclusion

We have demonstrated the direct measurement of the surface displacement in laser picosecond ultrasonics by using the pump and probe technique and a specially constructed interferometer. This should prove to be a useful method for the analysis of the strain pulse shape in this type of experiment.

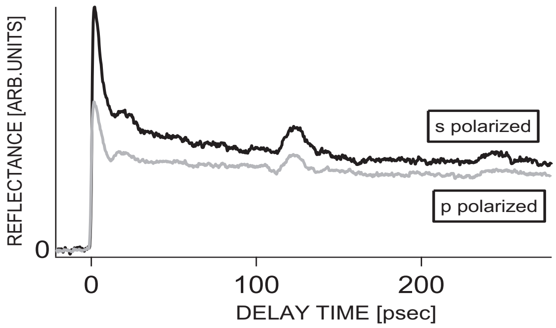

Fig. 1 The transient reflectivity change measured with s and p-polarized beam, as labeled.

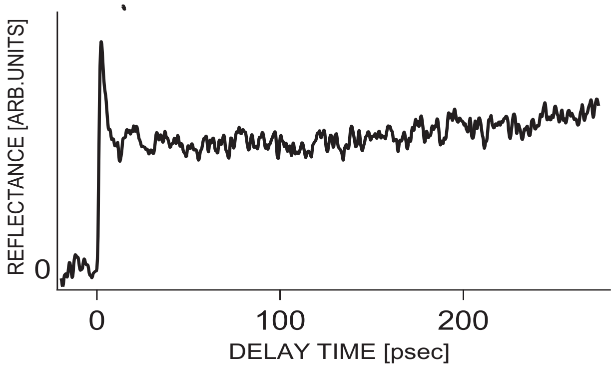

Fig. 2 By adjusting the amplitude ratio and relative phase difference of the s and p polarized light, the peak caused by the photoelastic effect is cancelled.

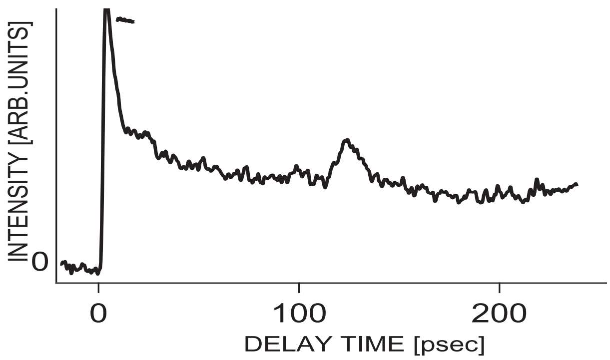

Fig. 3 Direct measurement of the surface displacement by mixing a reference beam to the probe beam used in Fig. 2. We can see a peak appears around 120 ps again.

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