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Study of ultrafast electron relaxation by the direct measurement of picosecond surface displacement

ピコ秒表面変位の直接測定による超高速電子拡散の研究

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1. Introduction

Picosecond laser ultrasonics makes use of optical pump and probe pulses with sub-picosecond temporal width to generate picosecond acoustic pulses in materials. The acoustic propagation is monitored by transient optical reflectance or phase changes caused by the photoelastic effect and surface displacements of the sample [1-5].

This technique is a useful method for studying ultrafast electron dynamics in metals, [2] in which the ultrashort light pulses are absorbed by the electrons near the sample surface within the optical absorption depth ζ. These nonequilibrium electrons diffuse into the sample. During this picosecond diffusion process the electrons transfer their excess energy to the lattice. This ultrafast electron relaxation raises the lattice temperature of the sample, and the resulting thermal stress generates the acoustic pulses. The spatiotemporal evolution of the lattice temperature rise thus governs the shape of the acoustic strain pulse generated. Precise measurement of the shape of the strain pulses is therefore useful for the investigation of ultrafast electron relaxation [2,3]. The pulse shape can be derived from the surface displacement by interferometrically measuring transient optical phase changes. However, the detected phase changes are also affected by the photoelastic effect in addition to the surface displacement, and it is difficult to distinguish these two contributions.

We previously proposed a method to cancel out the photoelastic effect and to obtain a signal solely determined by the surface displacement [7]. The contribution from the photoelastic effect is eliminated by interfering *s* and *p* polarization components of the probe light beam with an appropriate relative phase difference.

In this paper, we further develop this method and apply it to measurements on a metal film. The experimental results should be compared with a theory including electron diffusion. Such a theory is the two-temperature model (TTM) in which the electrons and lattice vibrations thermalize almost independently, and interact weakly through electron-phonon coupling [2-4].

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2. Theory

Two contributions to the reflectance can be distinguished by the use of optical probe beams of *s* and *p* polarization at oblique incidence [6,7]. In the case of an opaque semi-infinite sample, the complex reflectance changes for these polarizations can be expressed as

$$\frac{\delta r_s}{r_s} = iA + \alpha B\,, \qquad \frac{\delta r_p}{r_p} = iA + \beta B, \qquad (1)$$

where A is a real quantity and is proportional to the surface displacement, B is a complex quantity related to the photoelastic effect, and α and β are complex constants. The difference between the complex reflectance change of *s* and *p* polarizations arises only through the constants α and β. It has been shown that A can be deduced from the experimentally obtainable dr_s/r_s and dr_p/r_p [6].

However, the accuracy of this method is not sufficient in practice owing to the need to take two independent measurements for *s* and *p* polarizations: to avoid this difficulty, we proposed a method for the direct measurement of the surface displacement [7]. The total detected optical probe intensity can be arranged to be directly proportional to the surface displacement, and the photoelastic contribution can be completely eliminated .

The shape of the optically generated acoustic pulse can be described by the TTM [2-4]. In this model the electron temperature T_e and the lattice temperature T_i are described by two coupled partial differential equations

$$C_e \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial z}\left(\kappa \frac{\partial T_e}{\partial z}\right) - g(T_e - T_i) + \frac{1}{\zeta} S(t) e^{-z/\zeta} \qquad (2)$$

$$C_i \frac{\partial T_i}{\partial t} = g(T_e - T_i) \qquad (3)$$

where C_e is the electron heat capacity per unit volume, κ is the thermal conductivity. g is the electron-phonon coupling constant, and $S(t)$ is the normalized temporal variation of the power deposited by the pump light pulse. The thermal stress is given by $-C_i \gamma \Delta T_i$, where C_i is the lattice heat capacity per unit volume and γ is the Grüneisen parameter. By combining the acoustic wave equation with the generated stress distribution, the spatiotemporal strain is obtained as follows:

$$\eta(z,t) = -\frac{\gamma C_i}{2\rho v^2}\int_{-\infty}^{t} \mathrm{sgn}\left[z - v\left(t - t'\right)\right] \times \left.\frac{\partial T_i}{\partial t}\right|_{(|z-v(t-t')|,t')} dt'. \quad (4)$$

3. Experimental Setup

The sample is a thin film of tungsten of thickness 220 nm deposited on a crown glass plate using radio-frequency sputtering. We use a mode-locked Ti:sapphire laser of central wavelength 830 nm, pulse duration ~200 fs, and repetition rate 82 MHz as a light source to generate the ultrasonic pulses by illumination from the top surface .

A mixture of *s* and *p* polarized light components at 415 nm is used for the probe beam at an oblique incident angle. We adjust the intensity ratio and relative phase retardation between the *s* and *p* components to cancel out the photoelastic effect. Finally, a reference light beam is mixed with the probe beam to obtain the intensity due to the surface displacement only.

4. Results of the experiments

Typical reflectivity changes measured using an *s* polarized probe beam are shown in **Fig. 1 (**top curve). This reflectivity change originates solely from the photoelastic effect, and we cannot detect the surface displacement in this curve. Then, as we adjust the intensity ratio between *s* and *p* probe light beam components and their relative phase retardation, the photoelastic effect can be cancelled out. This is shown in the curve labeled as $\Delta\phi = 0^{\circ}$.

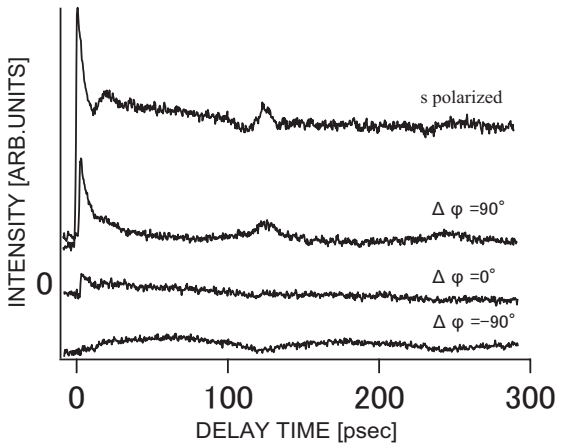

Fig.1 Transient reflectivity changes measured with an *s* polarized beam (top curve), and direct measurement of the surface displacement (see $\Delta\phi = -90^{\circ}$ and $\Delta\phi = 90^{\circ}$) by mixing with a reference beam.

Next, in order to measure the surface displacement, we turn on the reference light beam which interferes with the probe beam. As shown in **Fig. 1**, a peak again appears around 120 ps with a slightly different shape from that in the curve labeled as *s* polarized. This originates solely from the surface displacement variation. By varying the probe-reference phase retardation from $\Delta\phi = -90^{\circ}$ to $\Delta\phi = 90^{\circ}$, we observe a sign flip in the obtained intensity variation as expected. This strain pulse shape agrees reasonably well with theoretical simulations based on the TTM.

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