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<td>Author(s)</td>
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<tr>
<td>Citation</td>
<td>Journal of the Optical Society of America B - Optical Physics, 30(7), 1911-1921</td>
</tr>
<tr>
<td>Issue Date</td>
<td>2013-07</td>
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<tr>
<td>Doc URL</td>
<td><a href="http://hdl.handle.net/2115/53078">http://hdl.handle.net/2115/53078</a></td>
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<td>Rights</td>
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<td>Type</td>
<td>article</td>
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<tr>
<td>File Information</td>
<td>josab-30-7-1911.pdf</td>
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Ultrafast ellipsometric interferometry for direct detection of coherent phonon strain pulse profiles

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Received March 18, 2013; revised April 27, 2013; accepted April 30, 2013; posted May 8, 2013 (Doc. ID 187284); published June 19, 2013

We describe an ultrafast optical technique to quantitatively detect picosecond ultrasonic displacements of solid surfaces, thus giving access to the longitudinal strain pulse shape. Transient optical reflectance changes recorded at oblique optical incidence with a common-path interferometric configuration based on ultrafast ellipsometry monitor gigahertz coherent phonon pulses. We demonstrate for a tungsten film the quantitative extraction of the strain pulse shape free of distortions arising from the photoelastic effect, and analyze the results with the two-temperature model to obtain the value $g \approx 3 \times 10^7 \text{Wm}^{-2}\text{K}^{-1}$ for the electron–phonon coupling constant. Analysis of the data also reveals a thermo-optic contribution. © 2013 Optical Society of America

OCIS codes: (120.3180) Interferometry; (120.2130) Ellipsometry and polarimetry; (320.0120) Instrumentation, measurement, and metrology.

http://dx.doi.org/10.1364/JOSAB.30.001911

1. INTRODUCTION

Picosecond pump light pulses incident on opaque solid or liquid media can generate coherent phonon pulses in the gigahertz to terahertz range. Their propagation can be monitored with delayed probe light pulses as transient optical reflectance changes. This pulse-echo technique is referred to as picosecond laser ultrasonics, and has been used to study material properties and the vibrations of nanostructures [1–28]. Picosecond laser ultrasonics has also proved useful in elucidating the ultrashort dynamics of excited electrons [4,5,7,12,13,17,22]. For such quantitative studies, it is crucial to know the precise shape of the propagating acoustic strain pulses.

It is difficult in general, however, to measure the strain pulse shape because of the photoelastic interaction between the probe light and the sample, that is, the coupling of the optical indices to the strain. The probe light penetrates into the solid and monitors the strain over a finite region. This leads to a smoothing effect that normally prevents the exact strain pulse shape from being measured. With noninterferometric optical probing, only in certain limiting cases, for example, when the strain pulse spatial extent is much larger than the optical penetration depth, can the strain pulse shape be accurately measured [22].

Some steps toward the goal of measuring the strain pulse shape in laser picosecond ultrasonics were made when the detection technique was generalized to include optical beam deflection or distortion caused by surface displacements [3,5,29]. For longitudinal coherent phonon pulses, the surface displacement temporal variation can be used to extract the strain pulse profile [3]. However, although the surface displacement can be approximately monitored by these deflection or distortion methods, the radial variation of the probe reflectance over the optical pump spot caused by the photoelastic effect contributes to the recorded signals, thus preventing an accurate measurement of strain pulse profiles. A similar problem arises in the use of conventional (normal-incidence) interferometric detection [6,8,9]. In such detection setups, the surface displacement modulates the phase of the reflected probe light. For measurements at normal optical incidence, the phase modulation by the surface displacement is, again, always contaminated by the photoelastic effect.

Following these advances, a theoretical method for the uncontaminated extraction of ultrashort-timescale surface displacements with s- and p-polarized probe light at oblique incidence was proposed [30]. This method, however, involves two separate (i.e., s and p polarization) measurements and numerical analysis requiring their subtraction, thus compromising the method’s potential accuracy. To avoid this problem an interferometric method was later proposed that is functionally equivalent to the use of s- and p-polarized light measurements, but only requiring a single measurement with obliquely incident light [31]. In this paper, we describe in detail and experimentally demonstrate for the first time this ellipsometry-based [32–37] interferometric method by monitoring gigahertz longitudinal coherent-phonon generation and propagation in a tungsten film. We interpret the echo shapes obtained by implementing a two-temperature model of the electron relaxation in tungsten.

In Section 2 we outline the required light-scattering theory and the principle of the separation of the surface displacement and photoelastic contributions. Section 3 concerns the common-path interferometer setup, Section 4 the
experimental results, and Section 5 their analysis. A brief summary is given in Section 6.

2. THEORY

In a typical measurement in picosecond laser ultrasonics, the pump light is focused onto the sample surface to a spot with a radius ~10 μm, much larger than the depth usually important for acoustic wave propagation (typically <1 μm). The spatial divergence of the generated acoustic waves can therefore be neglected and, in homogeneous materials, plane wave acoustic propagation along the depth direction can be assumed. In addition, in isotropic materials, the only significant contribution to the generated acoustic waves comes from longitudinal polarization.

The propagating acoustic waves displace the surface position and modulate the permittivity of the sample through the photoelastic effect. Probe light, focused to the same region as the pump, can detect these permittivity changes through amplitude and phase modulations of the reflected probe light, and also detect the surface displacement through phase modulation. This latter modulation can also be effectively regarded as a permittivity modulation [38]. For depths smaller than the probe spot radius, the focused probe light can be considered to be a plane wave in the sample.

We further assume that the probe light is monochromatic with angular frequency ω or wavenumber k = ω/c. Since the acoustic frequency, up to terahertz-order in general, is much lower than the optical frequency, the modulation can be regarded as quasi-static, that is, the optical frequency of the reflected probe light is unchanged. The modulation of the reflected probe light electric field \( E(r, t) \) exp(-iωt) is then obtained by solving the quasi-static wave equation

\[
\{\nabla^2 - \text{grad} \ \text{div} + k^2 \varepsilon (r, t)\} E(r, t) = 0,
\]

where \( \varepsilon \) is a permittivity tensor including all perturbations caused by the acoustic wave propagation.

Suppose that a semi-infinite opaque isotropic homogeneous medium occupies \( z > 0 \) and a vacuum occupies \( z < 0 \). The permittivity modulation is assumed to be a function of coordinate \( z \) because of the one-dimensional nature of the problem we are describing.

Our oblique probe incidence exploits the polarization dependence of the probing process. The \( x \)-axis is taken along the sample surface and within the plane of optical incidence, so that the \( x \)-component of the incident probe light wave vector \( k \) is positive (\( k_x > 0 \) and \( k_y = 0 \)). Because of the lateral homogeneity of the system in the \( x-y \) plane, the electric field of the probe light takes the form

\[ E(r, t) = E(z, t) \exp(i k_x x), \]

that is, \( k_x \) is conserved for the incident, reflected, and transmitted probe light. The wave equation, Eq. (1), is then simplified to

\[ \{L(k_x) + k^2 \varepsilon (z, t)\} E(z, t) = 0, \]

where the operator \( L(k_x) \) is defined as

\[
L(k_x) \equiv \begin{pmatrix}
\frac{\varepsilon}{\varepsilon_0} & 0 & -ik_x \frac{\varepsilon}{\varepsilon_0} \\
0 & \frac{\varepsilon}{\varepsilon_0} - k_x^2 & 0 \\
-ik_x \frac{\varepsilon}{\varepsilon_0} & 0 & -k_x^2
\end{pmatrix}.
\]

The permittivity \( \varepsilon(z, t) \) can be expressed as the sum of a homogeneous part \( \varepsilon_h(z) \) and an inhomogeneous perturbation \( \Delta \varepsilon(z, t) \):

\[
\varepsilon(z, t) = \varepsilon_h(z) + \Delta \varepsilon(z, t).
\]

\( \varepsilon_h(z) \) is equal to the permittivity of the vacuum, i.e., unity, for \( z < 0 \), and to the permittivity of the medium, \( \varepsilon_1 \), for \( z > 0 \). \( \Delta \varepsilon(z, t) \) includes all other spatiotemporally varying perturbations, including the photoelastic effect and the surface displacement.

In the case \( \Delta \varepsilon(z, t) = 0 \) (i.e., the nonperturbed system), Equations (3)–(5) give the total electric field of the incident, reflected, and transmitted probe light for \( s \)-polarized incident light of unit amplitude

\[
E_{0s} = \begin{cases} \exp(i k' z) e_s + r_s \exp(-i k' z) e_s & (z < 0) \\ t_s \exp(i k'' z) e_s & (z > 0) \end{cases}
\]

where

\[
e_s = \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}
\]

is the normalized polarization vector,

\[
r_s = \frac{k' - k''}{k' + k''}, \quad t_s = \frac{2k'}{k' + k''}
\]

are the amplitude reflectance and transmittance, respectively, and

\[
k' = (k^2 - k_0^2)^{1/2}, \quad k'' = (\varepsilon_1 k^2 - k_0^2)^{1/2}
\]

are the \( z \)-components of the wave vectors in vacuum and in the medium, respectively. Similarly, the total electric field for \( p \)-polarized incident light of unit amplitude is given by

\[
E_{0p} = \begin{cases} \exp(i k' z) e_{ip} + r_p \exp(-i k' z) e_{ip} & (z < 0) \\ t_p \exp(i k'' z) e_{ip} & (z > 0) \end{cases}
\]

where

\[
e_{ip} = \frac{1}{k} \begin{pmatrix} k' & 0 \\ -k_x & k_x \end{pmatrix}, \quad e_{rp} = \frac{1}{k} \begin{pmatrix} -k' & 0 \\ -k_x & k_x \end{pmatrix}, \quad e_{tp} = \frac{1}{\varepsilon_1^{1/2} k} \begin{pmatrix} k' & 0 \\ 0 & -k_x \end{pmatrix}
\]

are the normalized polarization vectors for the incident, reflected, and transmitted light, and

\[
r_p = \frac{\varepsilon_1 k' - k''}{\varepsilon_1 k' + k''}, \quad t_p = \frac{2\varepsilon_1^{1/2} k'}{\varepsilon_1 k' + k''}
\]

are the amplitude reflectance and transmittance, respectively.
With a small, finite perturbation $\Delta \varepsilon(z, t) \neq 0$, the total electric field becomes

$$
E(z, t) = E_0(z) + \int_{-\infty}^{\infty} k^2 G(z, z') \Delta \varepsilon(z', t) E(z', t) \, dz'
$$

$$
\approx E_0(z) + \int_{-\infty}^{\infty} k^2 G(z, z') \Delta \varepsilon(z', t) E_0(z') \, dz' \tag{9}
$$
to first-order. The vector $E_0$, corresponding to the polarization of the incident light, is a linear combination of Eqs. (5) and (8). $G(z, z')$ is a $3 \times 3$ Green’s function matrix which satisfies

$$
\{L(k_z) + k^2 \eta_{0}(z)\} G(z, z') = -\delta(z - z') I, \tag{10}
$$
where $I$ is the $3 \times 3$ identity matrix. For the system in question, the nonzero components of $G(z, z')$ are given by

$$
G_{xx}(z, z') = \frac{ik' k''}{k^2 (k_1' k'' + k')^2} \exp\{i(k' z' - k z)\},
$$

$$
G_{zz}(z, z') = \frac{ik' k''}{k^2 (k_1' k'' + k')^2} \exp\{i(k' z' - k z)\},
$$

$$
G_{yy}(z, z') = \frac{k}{k' + k''} \exp\{i(k' z' - k z)\},
$$

$$
G_{yx}(z, z') = \frac{ik' k''}{k^2 (k_1' k'' + k')^2} \exp\{i(k' z' - k z)\},
$$

$$
G_{yz}(z, z') = \frac{ik' k''}{k^2 (k_1' k'' + k')^2} \exp\{i(k' z' - k z)\}, \tag{11}
$$
for the region of $z < 0$ and $z' > 0$ [21, 30, 39]. $\Delta \varepsilon(z, t)$ is, for the time being, assumed to be composed of a sum of the photoelastic contribution $\varepsilon_{pe}$ and the surface displacement contribution $\varepsilon_{sd}$. Other contributions will be considered later. For an isotropic medium and $z$-propagating longitudinal acoustic waves, the photoelastic contribution is given by

$$
\varepsilon_{pe}(z, t) = \begin{pmatrix} P_{12} & 0 & 0 \\ 0 & P_{12} & 0 \\ 0 & 0 & P_{11} \end{pmatrix} \eta_{zz}(z, t), \tag{12}
$$
where $\eta_{zz}$ is the $zz$ strain component, and $P_{11}$ and $P_{12}$ are photoelastic tensor components in simplified notation, corresponding to $P_{1111}$ and $P_{1122}$, respectively, in full tensor notation. The surface displacement contribution is given by

$$
\varepsilon_{sd}(z, t) = \begin{cases} (\epsilon_f - 1) I & (u_0(t) < z < 0) \\ (1 - \epsilon_f) I & (0 < z < u_0(t)) \\ 0 & \text{otherwise} \end{cases}, \tag{13}
$$
where $u_0(t)$ is the $z$ component of the displacement at the surface ($z = 0$).

When $|u_0(t)|$ is much smaller than the optical wavelength, Eq. (9) is simplified to

$$
E(z, t) \approx E_0(z) + \int_{-\infty}^{\infty} k^2 G(z, z') \varepsilon_{pe}(z', t) E_0(z') \, dz' + k^2 u_0(t) G(z, +0)(1 - \epsilon_f) E_0(-0). \tag{14}
$$

Because of the discontinuity of $E_0$ and $G$ at $z = 0$, these values for the left ($-0$) and right ($+0$) limits toward $z = 0$ should be properly distinguished. Substituting Eqs. (6), (8), (11), and (12) into Eq. (14), the relative reflectance changes, $\delta r_s/r_s$ and $\delta r_p/r_p$, can be obtained. The resulting total electric field in the region $z < 0$ for $s$-polarized incident light is given by

$$
E = (\exp(ik' z) + r_s + \delta r_s) \exp(-ik' z)) e_s,
$$
where

$$
\frac{\delta r_s}{r_s} = 2ik u_0(t) + \frac{2ik^2 P_{12}}{1 - \epsilon_1} \int_0^\infty \eta_{zz}(z', t) \exp(2ik' z') \, dz'. \tag{15}
$$

Similarly, that for $p$-polarized incident light is given by

$$
\frac{\delta r_p}{r_p} = 2ik u_0(t) + \frac{2ik^2 P_{12}^2 - P_{11}^2 k^2}{k^2 - \epsilon_2 k^2} \int_0^\infty \eta_{zz}(z', t) \exp(2ik' z') \, dz'. \tag{16}
$$

Experimentally, the real and imaginary parts of the reflectance change $\delta r$ are independently obtainable by the use of an interferometer [6, 8, 9]. However, with measurement with a single polarization configuration, $\delta r_p/r_p$ or $\delta r_s/r_s$, for example, is not sufficient to separate the surface displacement contribution from the photoelastic contribution, since the imaginary part of the reflectance change contains both contributions.

This difficulty can be circumvented by the measurement of reflectance changes corresponding to two different polarization configurations together: $\delta r_p/r_p$ and $\delta r_s/r_s$. Equations (15) and (16) can be rewritten in condensed form as follows:

$$
\frac{\delta r_s}{r_s} = iA + c_s B, \tag{17}
$$

$$
\frac{\delta r_p}{r_p} = iA + c_p B, \tag{17}
$$
where $A$ is the real quantity $2k' u_0(t)$, $B$ is the integral appearing in Eqs. (15) and (16), and $c_s, c_p$ are the factors in front of these integrals, i.e.,

$$
c_s = \frac{2ik^2 P_{12}}{1 - \epsilon_1}, \quad c_p = \frac{2ik^2 (P_{12}^2 - P_{11}^2 k^2)}{k^2 - \epsilon_2 k^2}.
$$

Using Eq. (17), it is possible to cancel the quantity $B$ and retrieve $A$ from the independently observed variations $\delta r_s/r_s$ and $\delta r_p/r_p$ [31]. However, as previously mentioned, the subtraction of these quantities obtained from two different measurements to derive the surface displacement compromises accuracy.

Direct measurement of the surface displacement can be achieved by the procedure schematically outlined in Fig. 1. Consider the use of linearly polarized probe light with electric field amplitudes $E_s$ and $E_p$ for the $s$- and $p$-polarized components, respectively. As described later, it is possible to make an optical setup which interferes $s$- and $p$-polarized probe-light reflection components with an arbitrary phase retardation imposed between them. The resulting electric field amplitude is

$$
E_{in} = E_s r_s (1 + iA + c_s B) + \mu E_p r_p (1 + iA + c_p B), \tag{18}
$$
where \( \mu = \exp(i\phi) \) is an adjustable phase factor. If \( E_s, E_p \), and \( \mu \) are chosen to satisfy
\[
\mu E_p r_p c_p + E_s r_s c_s = 0
\]
the amplitude becomes
\[
E_{\text{int}} = (E_s r_s + \mu E_p r_p)(1 + iA),
\]
in which the photoelastic contribution is canceled out. Since \( A \) is a real quantity, the intensity of the reflected probe light is given by
\[
I_1 \propto |(E_s r_s + \mu E_p r_p)|^2,
\]
independent of \( A \), where a small second-order term proportional to \( A^2 \) is omitted. It is, however, possible to retrieve an intensity modulation proportional to \( A \) by use of a second interference with reference light exhibiting a \( \pi/2 \) phase difference with respect to the unperturbed probe light electric field \( E_s r_s + \mu E_p r_p \), yielding an intensity of detected probe light given by
\[
I_2 \propto |(E_s r_s + \mu E_p r_p)(1 + i + iA)|^2
\]
\[
= 2|E_s r_s + \mu E_p r_p|^2(1 + A).
\]

A linear term in \( A \) now appears in the intensity, allowing the temporal variation of the surface displacement to be followed. We implement this method in the present paper.

To carry out the above procedure, the condition \( c_p \neq c_s \) should hold; otherwise the reflected probe light disappears when the cancellation condition is fulfilled. This is not problematic in general because \( c_p \) and \( c_s \) depend on \( k_s \), i.e., on the probe-light incident angle, in different ways. It is therefore always possible to realize the condition \( c_p \neq c_s \) by choosing this angle appropriately.

3. EXPERIMENTAL SETUP

To demonstrate the above method, we shall apply an appropriate interferometric setup to the measurement of a tungsten film sample. Tungsten is chosen because it is acoustically and optically isotropic in the presence of texture, even in the case of elongated or aligned crystalline grains [40]. It is also advantageous for comparison purposes that several previous reports involving picosecond laser ultrasonics on tungsten films are available [7,8,11,17,29,41]. The film is deposited on a crown-glass substrate (of 1 mm thickness) by radio frequency sputtering. The nominal film thickness is 300 nm, and the root-mean-square surface roughness is measured by atomic-force microscopy to be \( \sim 9 \text{ nm} \) [42]. The ultrasonic measurements involve the use of a mode-locked Ti:sapphire laser with a repetition frequency of 80 MHz, a pulse duration of 200 fs, and a central wavelength of 830 nm. The fundamental beam (of wavelength of 830 nm) is focused onto the sample surface (the film side) with a spot diameter of 20 \( \mu \text{m} \) at full width at half-maximum (FWHM). This pump light, of fluence 25 \( \mu \text{J cm}^{-2} \) per pulse, thermoelastically generates the longitudinal coherent phonon pulses in the sample. Frequency-doubled probe light (of wavelength of 415 nm) generated by a \( \beta \)-BaB\(_2\)O\(_4\) crystal, of fluence 50 \( \mu \text{J cm}^{-2} \) per pulse, is focused onto the sample surface with a FWHM spot diameter of 8 \( \mu \text{m} \). The probe light detects transient optical reflectance changes using the interferometer setup described in this paper. The weaker pump light fluence compared to that of the probe does not pose any specific problems, because the interferometer output was found to be linear in both pump and probe fluences. The arrival of the probe light pulses at the sample is varied over the range 0–400 ps using an optical delay line.

Figure 2 shows the details of the interferometer. The probe light, after passing through the delay line, traverses a polarizer (POL1) and then a half-wave plate (HWP1) to make it linearly polarized at 45° to the plane of the optical bench (the horizontal plane). A polarizing beam splitter (PBS) then divides the light into horizontally and vertically polarized components, which propagate counterclockwise and clockwise, respectively, in the loop defined by the PBS, the sample, and the two mirrors. The former light pulses (the reference pulses) reach the sample about 1 ns before the latter (the probe pulses). The pump light pulses reach the sample between the arrival of the reference and probe pulses. The ratio between \( E_s \) and \( E_p \) for the probe light is adjusted by a half-wave...
plate (HWP2). The probe light reflected at the sample passes through a quarter-wave plate (QWP1), a polarizer (POL2), and a half-wave plate (HWP3). The optical axes of QWP1 are set at 45° to the horizontal plane so that the s- and p-polarized reflected probe light components interfere with one another with an arbitrary phase retardation determined by the rotation angle of POL2 (see Appendix A). HWP3 is set to rotate the polarization plane of the probe light exiting POL2 to be horizontal, so that the probe light traverses the PBS efficiently. The counterclockwise propagating reference light passes HWP3, POL2, QWP1, sample, and HWP2, and then its vertically polarized component is reflected at the PBS. The clockwise-propagating (horizontally polarized) probe light pulses and counterclockwise-propagating (vertically polarized) reference light pulses are unifed at the PBS, are reflected off a nonpolarizing beam splitter (NPBS), are interfered at a quarter-wave plate (QWP2) and a polarizer (POL3), and are finally fed to the photodetector. Because of the common-path configuration, the counterpropagating probe pulses are automatically temporally overlapped at the photodetector, and the optical system is insensitive to mechanical vibrations. The optical axes of QWP2 are set at 45° to the horizontal plane so that an arbitrary phase retardation between the probe and reference light is introduced by rotating POL3. A phase retardation of \( \pm \pi/2 \), achieved by setting POL3 to pass the vertical or horizontal polarization component, provides the optimum interference condition.

The intensity variation \( \Delta I \) of the probe light caused by the \( \approx \) pm surface displacement is typically very small, \( \Delta I/I \approx 10^{-6} \). To observe this variation with sufficient signal-to-noise ratio, the pump light pulses are modulated at 1 MHz with an acousto-optic modulator, and the modulation in the photodetector output at this frequency is detected by a lock-in amplifier. The required bandwidth for the photodetector is thus in the MHz range (about 2 MHz in the present case).

The setup can be easily converted for oblique-incidence interferometric measurements [59, 43]. For this purpose, QWP1 and POL2 are removed. The polarization of the probe light is selected to be either \( s \) or \( p \) by the use of HWP2 and HWP3. According to the position of POL3, the reflected probe light intensity is chosen in turn by the use of HWP2 and HWP3. The required optical components in these different situations are summarized in Table 1.

### Table 1. Required Optical Components for Different Interferometric Measurements

<table>
<thead>
<tr>
<th>Functionality</th>
<th>QWP1</th>
<th>POL2</th>
<th>QWP2</th>
<th>POL3</th>
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<tbody>
<tr>
<td>Cancellation of photoelastic contribution</td>
<td>*</td>
<td></td>
<td></td>
<td>H</td>
</tr>
<tr>
<td>Direct displacement measurement</td>
<td></td>
<td>*</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Oblique incidence interferometer</td>
<td></td>
<td></td>
<td>*</td>
<td></td>
</tr>
<tr>
<td>Simple reflectivity measurement</td>
<td></td>
<td></td>
<td></td>
<td>H</td>
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</tbody>
</table>

*—, not used; *, used; H, used and set to a horizontal orientation.

4. RESULTS

The curves in Figs. 3(a)–3(d) show the results of oblique-incidence interferometric measurements. The curves (a) and (b) correspond to the real \( (\rho_j) \) and imaginary \( (\delta \varphi_j) \) parts of the relative reflectance change for s-polarized probe light, whereas (c) and (d) correspond to those for p-polarized probe light \( (\rho_p \text{ and } \delta \varphi_p) \).

Consider first Figs. 3(a) and 3(c), corresponding to \( \rho_j \) and \( \rho_p \), respectively. A sharp peak is observed at zero delay time \( (t = 0) \). This result from the electron temperature rise caused by the absorption of the pump photon energy and from the subsequent energy transfer to the lattice (see Section 5). After this sharp peak, a gradual decrease is observed, related to the cooling of the sample. Upon this slowly decaying background, an echo is observed at 120 ps. This is caused by a longitudinal coherent phonon pulse returning after reflection from the film-substrate interface. The echo consists of a central peak and two small dips on either side. This is a typical shape for echoes arising from the photoelastic effect in opaque samples. Half of this shape is also observed after zero delay time owing to the departure of the coherent phonon pulse from the film surface after thermoelastic generation.

A second, smaller echo is also resolved at 240 ps.

In Figs. 3(b) and 3(d), corresponding to \( \delta \varphi_j \) and \( \delta \varphi_p \), respectively, 3 echoes are clearly observed at times determined by the sound velocity \( v_l = 5160 \text{ ms}^{-1} \) [44] and thickness (estimated to be \( d = 313 \text{ nm} \)) of the tungsten film. As shown in Eqs. (15) and (16), the terms contributing from the photoelastic effect are similar to a spatial Fourier transform of the strain field, and exhibit a high sensitivity to the wavenumber components of the strain field \( \eta_{zz}(z, t) \) near \( |k_{\text{strain}}| \approx 2k \) (where \( k_{\text{strain}} \) is the acoustic wavenumber). This selectivity is essentially the same as that involved in the detection of Brillouin oscillations in transparent media. In the curves (a) and (c) for \( \rho_j \) and \( \rho_p \), such frequency components are involved in the first echo, but not much in higher-order echoes because of frequency-dependent ultrasonic attenuation. This also implies that the second and third echoes in curves (b) and (d) for \( \delta \varphi_j \) and \( \delta \varphi_p \) are less affected by the photoelastic contribution than the first peak is, and that the decrease of the peak height in the curves (b) and (d) approximately follows the decrease of the overall strain pulse height.

The data of Figs. 3(a)–3(d) are in overall agreement with previous laser-picosecond-ultrasonics based investigations on tungsten films [7,8,11,17,29,41]; the contribution to the echo shape from the photoelastic effect depends strongly on the probe wavelength [11] and probe incident angle [45]. The latter dependence is evident in Eqs. (15) and (16).

Our direct displacement measurement consists of three steps. The first step is an oblique-incidence reflectivity change measurement with s- or p-polarized light corresponding to the
or a phase shift compared to \(0.0133\) for comparison purposes. In tungsten, it turns out that the phase retardation in this setup converts phase modulations from \(\sim 20\) to \(\sim 0.0133\) by means of the photoelastic effect. The phase retardation can be expressed as \(\phi_{PS} = \frac{\pi}{2} \xi / v t\), and is much broader than the central peak of the echo observed in Fig. 4(a) by means of the photoelastic effect.

Unlike the photoelastic contribution, which has a nonuniform frequency response up to terahertz acoustic frequencies, this direct displacement measurement shows that the curves (b) and (d) are still affected by the photoelastic contribution. This will be shown in detail in the next section.

5. DISCUSSION

In this section, we first explain a model that can account for the strain pulse shape and at the same time can retrieve the strain pulse shape. The corresponding experimental curve is shown in Fig. 4(e). The strain pulse width (at FWHM of \(\partial u_z / \partial t\)) is \(\sim 40\) ps, and is much broader than the central peak of the echo observed in Fig. 4(a) by means of the photoelastic effect.

We reproduce the results of the direct displacement measurement [corresponding to the upper curve in Fig. 4(c)] in Fig. 3(e) for comparison purposes. In tungsten, it turns out that the photoelastic contribution to the probe optical phase variation is, for \(s\)- and \(p\)-polarizations, \(\sim 7\%\) and \(\sim 20\%\), respectively, of the surface displacement contribution, and so the curves in Figs. 3(b), 3(d), and 3(e) are similar. However, the top of the first echo in Fig. 3(e) is somewhat sharper than the corresponding features in the curves (b) and (d). This shows that the curves (b) and (d) are still affected by the photoelastic contribution. This will be shown in detail in the next section.

\[
\frac{\partial u_z}{\partial t} = 20 f(v t).
\]

In other words, by monitoring the temporal derivative of the surface displacement, one can extract the coherent phonon strain pulse shape. The corresponding experimental curve is shown in Fig. 4(e). The strain pulse width (at FWHM of \(\partial u_z / \partial t\)) is \(\sim 40\) ps, and is much broader than the central peak of the echo observed in Fig. 4(a) by means of the photoelastic effect.

\[
u_z(0, t) = \int_{-\infty}^{\infty} f(z' - vt) + f(z' + vt) \, dz'.
\]

By taking the temporal partial derivative of Eq. (23), the strain waveform \(f(\xi)\) is given by [3]

\[
\frac{\partial u_z}{\partial t} \bigg|_{\xi = 0} = 2f(v t).
\]
physical properties of the tungsten film. We include the ultra-
fast dynamics of the photoexcited electrons, the generation
and propagation of acoustic waves, and the modulation of
the reflected probe light through the photoelastic effect,
the surface displacement, and the thermo-optic effect.

For the present experimental geometry, the thermoelastic
generation and acoustic propagation can be regarded as a
one-dimensional problem involving the coordinate \( z \). For
thermoelastic expansion in an isotropic medium, only longi-
tudinal acoustic waves are directly generated; in our one-
dimensional treatment, mode conversion to shear waves is
also precluded. The acoustic wave equation is then given by

\[
\frac{\partial^2 u_z}{\partial z^2} = \frac{\sigma_{zz}}{\rho} = c_{11} \frac{\partial^2 u_z}{\partial z^2} - (c_{11} + 2c_{12})\beta \frac{\Delta T}{\partial z},
\]

(25)

where \( \rho \) is the mass density, \( \sigma_{ij} \) are stress tensor components,
\( c_{ij} \) are elastic stiffness tensor components, \( \beta \) is the linear ther-
mal expansion coefficient, and \( \Delta T \) is the temperature rise.

The spatial extent of the generated strain pulse, as shown in
curve (c) in Fig. 4, is significantly larger, \( \sim 200 \) nm (equivalent to
\( \sim 40 \) ps), than the pump optical penetration depth (\( \sim 30 \) nm)
because of ultrafast electron diffusion during generation. This
process can be described by the two-temperature model, in
which the electron and lattice systems are considered to be
in quasi-independent thermal equilibrium characterized by the
temperatures \( T_e \) and \( T_l \), respectively, but interacting in a non-
linear fashion through the electron–phonon interaction [47]:

\[
\begin{align*}
C_e \frac{\partial T_e}{\partial t} & = \frac{\partial}{\partial z} \left( \kappa \frac{\partial T_e}{\partial z} \right) - g(T_e - T_l) + S(z, t), \\
C_l \frac{\partial T_l}{\partial t} & = g(T_e - T_l),
\end{align*}
\]

(26)

where \( C_e = \gamma T_e \) and \( C_l \) are the volume specific heats of the
electron and lattice systems, respectively, \( g \) is the electron–
phonon coupling constant, and \( \kappa = \kappa_0 T_e / T_l \) is the thermal
conductivity, where \( \kappa_0 \) is the value of \( \kappa \) when the electron
and lattice systems are in equilibrium [48, 49]. The lattice heat
diffusion is much smaller than the electron heat diffusion in
metals, and is ignored here. \( S(z, t) \) is the deposited energy per
unit time per unit volume by the absorption of a pump light
pulse. At the film surface and the film/substrate interface,
there should be no electron flow. This corresponds to the
boundary condition \( \kappa \partial T_e / \partial z = 0 \). Equation (26) is solved nu-
erically by a finite difference time domain method to obtain the
spatiotemporal evolution of \( T_e \) and \( T_l \). Immediately after the
excitation by a light pulse with a temporal width shorter
than 1 ps, \( T_l \) becomes in our case higher by several tens of K,
whereas \( T_l \) stays near room temperature. The electron energy
is then transferred to the lattice. Since the electrons diffuse
during this thermalization process, the depth of the region
with raised \( T_l \) is generally larger than the optical penetration
depth. In general, the spatial distributions of \( T_e \) and \( T_l \) at a particular
time (e.g., several ps) after pump pulse absorption are broader for metals with smaller \( g \) or larger \( \kappa \). The spatio-
temporal evolution of the strain field \( \eta_{zz}(z, t) \) can be calculated from
Eq. (25) with a knowledge of \( \Delta T \), defined here as the variation of \( T_l \) from its initial value. In this theoretical
simulation, the maximum transient lattice temperature rise is
about 1 K [50].

In Fig. 3 the ratio between the heights of the first and
second echoes for \( \rho_s \) and \( \rho_p \) is about 0.3, whereas that for \( \delta \rho_s \) and \( \delta \rho_p \) is about 0.6. As discussed in Section 4, \( \rho_s \) and \( \rho_p \) arise from the photoelastic effect, and have a sensitivity
characterized by a nonuniform acoustic frequency spectrum,
whereas \( \delta \rho_s \) and \( \delta \rho_p \) are mostly governed by the surface dis-
placement, and have a nearly frequency-independent sensitiv-
ty. The observed difference in the echo height ratio points to
the presence of a frequency-dependent ultrasonic absorption
coefficient \( a \). We assume a variation in the empirical form

\[
a = a + bf^2,
\]

(27)

where \( f = \omega/2\pi \), and \( a \) and \( b \) are constant coefficients. The
quadratic frequency absorption coefficient \( b \) may contain contribu-
tions from viscoelasticity [51, 52], whereas the frequency-independent absorption coefficient \( a \) can arise from deviations of the acoustic reflection coefficient at the
W/crown-glass interface from the assumed literature value.
The values of \( a \) and \( b \) can be obtained by comparison of the Fourier amplitude of the first, second, and third echoes [53].
This attenuation is also included in the simulation of the
propagation of the coherent phonon pulses. The detailed
procedure is described elsewhere [54].

Once \( \eta_{zz}(z, t) \) is obtained, Eqs. (15) and (16) can be used to
calculate the optical transient reflectance change. But, as
shown in Fig. 4(b), there is a residual negative reflectivity step
even when the photoelastic contribution is effectively can-
celled. To explain the presence of this step, we consider a per-
mittivity modulation by the temperature variation (i.e., the
thermo-optic effect) in addition to that by the photoelastic
effect. The delay time in question is greater than a few picosec-
onds, at a time when the electrons can be regarded as
thermalized with the lattice. This thermo-optic effect is in-
cluded by replacing \( \varepsilon_{pe} \) in Eq. (14) with [17, 55]

\[
\Delta \varepsilon_{ij} = P_{io} \delta_{ij} \Delta T + P_{ijkl} \eta_{ijkl}.
\]

(28)

Here, \( P_{io} \) and \( P_{ijkl} \) are complex thermo-optic coefficients and
photoelastic tensor components, respectively. The second
term in this equation is the photoelastic contribution, \( \varepsilon_{pe} \),
obtained from Eq. (12) for pure longitudinal waves (\( \eta_{ij} = 0 \)
except \( \eta_{zz} \)), and is not an isotropic second-rank tensor,
whereas the first term, the thermo-optic contribution, is.
Because of the mixed isotropic/anisotropic nature of these
terms, the reflectance change arising from the thermo-optic
contribution does not in general cancel when that from the
photoelastic contribution does.

In Fig. 5 the experimental intensity variation obtained from
curve (c) in Fig. 4 is fitted in the region from 80 to 400 ps using
the parameters shown in Table 2. The temporal range from 0
to 80 ps is not well fitted partly because of the excited electron
contribution, which is not included here. For the permittivity
at the pump light wavelength (830 nm), the value obtained by
ellipsometry measurement on our sample is adopted rather
than the literature value [14] of 4.1 + 19.4i [50]. The
thermo-optic contribution and the surface displacement con-
tribution are also shown separately in Figs. 5(c) and 5(d).
The slow background decay in curve (a) can be attributed to
the (equilibrated electron and lattice) temperature variation.
The surface displacement contribution gives the echoes as well
as the flat (in fact, step-like) negative background which is
caused by the (quasi-) static strain distribution arising from thermal expansion. The numerical value of the surface displacement corresponding to the first echo is ~0.6 pm, in reasonable agreement with the predictions of our model (0.4 pm).

In particular, the fitting yields a value for the electron–phonon coupling constant $g$, as shown with other parameters in Table 2. We obtain a possible range of $g$ values from $1 \times 10^{17}$ to $1 \times 10^{18}$ Wm$^{-3}$ K$^{-1}$, with a least-squares optimal value at $g = 3 \times 10^{17}$ Wm$^{-3}$ K$^{-1}$. This value is in good agreement with the values in [61] ($g \approx 5 \times 10^{17}$ to $1 \times 10^{18}$ Wm$^{-3}$ K$^{-1}$ obtained from transient reflectivity changes), [7,62] [$g \approx (2.6 \pm 0.39) \times 10^{17}$ Wm$^{-3}$ K$^{-1}$ from the transient reflectivity changes], and [17] ($g = 1.0 \times 10^{17}$ Wm$^{-3}$ K$^{-1}$ from optical interferometric measurements of acoustic pulse shapes). These various values for $g$ might arise because of differences in sample purity or microstructure. Further studies, for example with varying film thickness or preparation method, would help elucidate this point.

As a check on our theoretical approach, we now attempt to reproduce all the experimental data for the first echoes by the use of Eqs. (15) and (16). Figures 6(a)–6(d) show the results for amplitude and phase for the first echo, with thermo-optic background subtracted, from experiment (solid lines) and from theory (dotted lines), which exhibit very good agreement overall. In (b) and (d) for $\delta \phi_p$ and $\delta \phi_{p\rho}$, respectively, the photoelastic (pe) and surface displacement (disp.) contributions are shown below their sum. Table 3 shows the optical and photoelastic parameters obtained by least-squares fitting [63]. In (e) and (f), the surface displacement with thermo-optic background subtracted and its temporal derivative (i.e., the shape of the strain pulse propagating inside the film) are also, respectively, shown, and exhibit reasonable agreement between experiment and theory within the noise level.

Figure 7 shows the first-echo frequency spectrum obtained experimentally (solid line) and theoretically (dashed line) from the displacement variation [64]. Good agreement is obtained, with frequency components significant up to ~50 GHz (limited by electron diffusion).

Table 2. Parameters Used for the Theoretical Simulation of the Tungsten Film

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value (unit)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal conductivity</td>
<td>$1.78 \times 10^2$ Wm$^{-3}$ K$^{-1}$</td>
</tr>
<tr>
<td>Lattice specific heat</td>
<td>$2.53 \times 10^6$ Jm$^{-3}$ K$^{-1}$</td>
</tr>
<tr>
<td>Constant for electron specific heat</td>
<td>$1.37 \times 10^2$ Wm$^{-3}$ K$^{-1}$</td>
</tr>
<tr>
<td>$g$ (Wm$^{-3}$ K$^{-1}$)</td>
<td>$3 \times 10^{17}$</td>
</tr>
<tr>
<td>Mass density, $\rho$ (kgm$^{-3}$)</td>
<td></td>
</tr>
<tr>
<td>for W [44]</td>
<td>$1.93 \times 10^4$</td>
</tr>
<tr>
<td>for crown glass [50]</td>
<td>$2.24 \times 10^3$</td>
</tr>
<tr>
<td>Electrical permittivity, $\varepsilon_1$</td>
<td>$4.6 + 5.5i$</td>
</tr>
<tr>
<td>Thermal expansion coefficient [44], $\beta$ (K$^{-1}$)</td>
<td>$4.5 \times 10^{-6}$</td>
</tr>
<tr>
<td>Elastic stiffness (Pa)</td>
<td></td>
</tr>
<tr>
<td>$c_{11}$ for polycrystalline W [44]</td>
<td>$5.13 \times 10^{11}$</td>
</tr>
<tr>
<td>$c_{44}$</td>
<td>$1.59 \times 10^{10}$</td>
</tr>
<tr>
<td>$c_{11}$ for crown glass [50]</td>
<td>$5.82 \times 10^{10}$</td>
</tr>
<tr>
<td>$c_{44}$</td>
<td>$1.81 \times 10^{10}$</td>
</tr>
<tr>
<td>Film thickness, $d$ (nm)</td>
<td>$3.13 \times 10^2$</td>
</tr>
<tr>
<td>Ultrasonic absorption coefficient</td>
<td>$3 \times 10^6$</td>
</tr>
<tr>
<td>$b$ (m$^{-1}$)</td>
<td>$4 \times 10^2$</td>
</tr>
<tr>
<td>Thermo-optic coefficient, $P_{10}$ (K$^{-1}$)</td>
<td>$(-2.0 - 0.4i) \times 10^{-3}$</td>
</tr>
</tbody>
</table>

*Values $g$, $d$, $a$, $b$, and $P_{10}$ are obtained by fitting (marked by the * symbol). The permittivity for 830 nm is obtained from ellipsometry measurements. All other parameters are taken from the literature.

Table 3. Parameters Used for the Theoretical Simulations for Oblique-Incidence Interferometric Measurements

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value (unit)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electrical permittivity, $\varepsilon_1$</td>
<td></td>
</tr>
<tr>
<td>for W at 415 nm</td>
<td>$3.8 + 4.4i$</td>
</tr>
<tr>
<td>Photoelectric constants for W at 415 nm</td>
<td></td>
</tr>
<tr>
<td>$P_{11}$</td>
<td>$-5.1 + 8.5i$</td>
</tr>
<tr>
<td>$P_{12}$</td>
<td>$-5.1 - 1.5i$</td>
</tr>
</tbody>
</table>

*Values are obtained by the fitting.
A polarizer with its polarization plane rotated by $\pi/4 + \phi$ from the $X$ axis is placed behind the QWP. Then the amplitude of the electric field along the polarization plane at the point A is given by

$$E_A = \left( \cos \left( \frac{\pi}{4} + \phi \right) \sin \left( \frac{\pi}{4} + \phi \right) \right)_{\text{QWP}} E$$

$$= \frac{i}{\sqrt{2}} (E_X \exp(i\phi) + E_Y \exp(-i\phi)).$$

One can interpret this by thinking of $E_X$ as retarded by $2\phi$ and then added to $E_Y$. In this way, it is possible to interfere two orthogonal polarization components with arbitrary phase retardation by a single adjustment of the polarizer angle $\phi$.

### ACKNOWLEDGMENTS

This work is partially supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Science, Sports, and Culture of Japan.

### REFERENCES AND NOTES


40. W is optically isotropic because it crystallizes in the cubic (bcc) phase. In addition, the elastic constants for tungsten single crystal are $c_{11} = 502$, $c_{12} = 246$, and $c_{44} = 199$ GPa [59], and coincidentally obey the relation $c_{11} - 2c_{12} = 2c_{44}$, so that (independent of the film microstructure) the film is effectively cia stropic.


42. The surface roughness contributes to the frequency-dependent ultrasonic absorption (see [22]), but we estimate its effect to be small over the frequency spectrum of the ultrasonic pulse associated with the first echo. The roughness of the W/crown-glass interface is ~5 nm, and its effect can also be neglected.


46. Some residual photothermal effect that was not cancelled is, however, still visible near $t = 0$. This level of small remnant should not significantly affect the shape of the final extracted surface displacement variations.

60. As discussed in relation to Fig. 6, the permittivity at the probe light wavelength can be retrieved from the interferometric data. This fitted value for the probe light lies close to that of corresponding ellipsometry data, so we also adopted the ellipsometry data value for the pump light.


63. We used fitted values for the permittivity of the W film at the probe wavelength rather than from the literature, 5.3 + 16.2i (see [44]), or from ellipsometry measurement, 3.1 + 4.1i. These differences in permittivity may be attributable to impurities in the W film.

64. The spectrum in strain, obtained by multiplication of that of the displacement by iω, shows significant noise above ~30 GHz.

