Signal analysis and characterization of experimental setup for the transient thermoreflectance technique

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The transient thermoreflectance (TTR) technique is a powerful optical pump-probe technique often used to measure thermal properties and monitor ultrafast processes. The technique has been used to measure a range of properties including the thermal conductivities of thin films and electron-phonon coupling factors to mention a few. TTR measurements are sensitive to residual heating and misalignment, which can lead to erroneous analysis of TTR data. To minimize these errors, we have developed a simple phase correction technique to reduce errors associated with residual heating and other background noise. Besides its simple implementation, the technique also requires no previous knowledge of the transient reflectance response. The technique is verified with simulated experimental data. The importance of proper alignment of the pump and probe beams over the entire range of pump-probe time delays of interest is discussed, along with a means of quantifying error associated with misalignment. © 2006 American Institute of Physics. [DOI: 10.1063/1.2336187]

I. INTRODUCTION

The time dependent transient thermoreflectance (TTR) technique has been used to study a range of thermal processes. The technique uses an ultrashort pulsed laser to excite the material of interest and a weaker probe beam to detect the transient reflectance. Paddock and Eesley were the first to use the technique to measure thermal diffusion in thin metal films.1 Since then, TTR has been used to make a range of material property measurements including thermal diffusivity of metal films,2,3 sound velocity,2,4 electron-phonon coupling factors of metal films,5-7 electron-phonon coupling within nanoparticles,8,9 thermal boundary resistances between metal films and dielectric substrates,10-12 and thermal property imaging.13 For a detailed description of the TTR technique and its applications see paper by Norris et al.14

For many of the above studies, the pump beam is modulated at a frequency of ~1 MHz, which results in a period of time when the probe reflectance is affected by the pump followed by a reference period when the pump beam is blocked. Therefore, a modulation occurs within the probe reflectance. The magnitude and phase of this signal are both significant and combine to yield the change in reflectance of the surface as a function of time. The change in reflectance is then related to change in temperature using thermoreflectance models.15 Once the change in temperature of the sample surface resulting from the heating event has been obtained, accurate determination of the thermophysical properties is possible.

Proper interpretation of TTR signals is critical in measuring thermal and other ultrafast processes, especially for the case when there are transients that are longer than the time between pump pulses. Capinski and Maris proposed a correction technique for slowly decaying responses.16 The Capinski and Maris technique requires that the functional form of the change in reflectivity is known and analytic. Although the functional form of the process of interest might be well known, longer and more complex decay processes such as three-dimensional substrate diffusion can affect the signal and lead to errors or require several free parameters. Bonello et al. developed a technique to measure relaxation processes with time scales on the order of the modulation time by varying the modulation frequency.17 The technique of Bonello et al. appears to be well suited for examining thermal diffusion in semi-infinite samples such as bulk materials or thick materials with low thermal diffusion coefficients. Taketoshi et al. proposed using the signal phase rather than the signal amplitude to measure thermal processes.18 The model of Taketoshi et al. was developed based on the condition that the overall residual temperature rise is much larger than the temperature rise from a single pump excitation. This is restricted to relatively thin metal films (~100 nm) on low thermal effusivity substrates (glass) and only after the electrons and lattice have equilibrated. Cahill et al. proposed a technique that uses the ratio of the in-phase to the out-of-phase lock-in signal by starting with the frequency domain solution.19 The technique reduces the errors associated with drift in the pump and probe as well as misalignment. Again, the full functional form for the system being investigated must be well known.

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In this study, we present a simple technique that uses the lock-in signal and phase from TTR experiments to resolve the change in reflectance for a wide range of thermal processes. The technique does not require the analytical functional form of the decay processes. Application of the technique for measuring thermal processes for a range of time scales $1$–$1000$ ps is demonstrated. The model is then compared to simulated data to determine the appropriate circumstances for which it is applicable. Finally, a technique is discussed to check and minimize errors associated with misalignment of the pump and probe beams during long TTR scans.

II. SAMPLE DATA

The TTR method uses an intense ultrashort laser pulse to generate a transient thermal response, and a weaker probe pulse to monitor the reflectivity response of the surface. Changes in reflectivity can be related to the change in temperature for most metals. A schematic of the experimental setup is shown in Fig. 1. The pulses from a $76$ MHz Ti:sapphire laser with a full width at half maximum (FWHM) pulse width of $200$ fs are separated into two beams with an intensity ratio of $9:1$ by a nonpolarizing beam splitter. The intense “pump” beam is used to heat the film while the low power “probe” beam is used to monitor the reflectivity. The pump beam passes through an acousto-optic modulator, which creates a pulse train at a frequency of $1$ MHz. The pump beam is focused to $80$ μm at an incident angle of $30^\circ$ with an estimated fluence of $\sim 2 \text{ J/m}^2$. The probe beam passes through a dovetail prism mounted on a variable delay stage that is used to increase the optical path length of the probe beam and hence the time delay between the pump and probe pulses. A half-wave plate rotates the probe beam’s polarization parallel to the plane of incidence. The probe, which is centered in the heated area, is focused to $\sim 15$ μm at near normal incidence to minimize the illuminated area. A polarizer, oriented such that only the probe light passes and not the pump, is positioned before a silicon photodiode, which monitors the probe beam’s reflection off the sample. Fluctuations in the probe beam are detected by a high speed photodetector connected to a lock-in amplifier. The lock-in amplifier displays both the magnitude and the phase of the voltage signal received from the photodetector.

Figure 2 shows the information obtained from the lock-in amplifier as a function of time for a $23$ nm Au film. On this particular scan the incident power was low, such that the nontransient signal occurring at $1$ MHz was on the same order of magnitude as the transient signal to be determined. This type of scan is used to study the electron-phonon coupling that occurs during the first few picoseconds. Assuming that the Au film exhibits a generally linear response to the incident fluence, increasing the incident power should influence the magnitude but not the overall shape of the response. Figure 3 shows the magnitude and phase of a scan taken on the same sample with twice the incident power as that in Fig. 2.
III. PHASE CORRECTION MODEL

As can be seen in Figs. 2 and 3, the magnitude and phase of the signal are each comprised of a transient and a nontransient component. In the initial part of the scan, only the nontransient portion of the signal is present. When the time delay between the pump and probe is zero \((t=0)\) the transient signal increases and then as the delay increases the signal reaches a maximum, and then decays on a picosecond time scale.

The nontransient portion of the signal comes from several sources, such as rf noise, fluctuation in the probe intensity, pump scatter, and residual heating. There is a certain amount of rf noise at 1 MHz, which is detected by the lock-in amplifier; this signal is on the order of a few microvolts for the scan shown. The Ti:sapphire laser used in this setup is very stable once mode locked; however, there is still a small amount of fluctuation in the intensity that results in a signal of approximately 1 \(\mu\)V. Any scatter of the pump beam that reaches the detector results in a 1 MHz signal, since the pump is modulated at this frequency. Another source of the nontransient signal comes from residual heating. Although the model assumes that all the energy is dissipated prior to the arrival of the next pulse, the average power of the pump beam does heat the sample. The pump beam is modulated at 1 MHz, therefore when the pump beam is blocked the sample surface will cool slightly. This results in a temperature modulation of the reflected probe beam at 1 MHz. The magnitude of residual heating and scattered pump signals vary from sample to sample. The nontransient signal due to residual heating is greater for thin films on insulating substrates versus highly conductive substrates.

The magnitude and phase of the response obtained from the lock-in amplifier are both functions of time. The amplitude of the transient response that results from the pump pulse is a function of time; however, the phase of this signal should not change with time. Using the above assumptions, the following expression can be formulated:

\[
A_n(t)e^{i(\omega t+\phi_n(t))} = A_m(t)e^{i(\omega t+\phi_m)} + A_n e^{i(\omega t+\phi_n)},
\]

(1)

where \(t\) is the time delay, \(\omega\) is the modulation angular frequency, \(A_m(t)\) and \(\phi_m\) are the magnitude and phase of the measured signal as a function of delay time, \(A_n\) and \(\phi_n\) are the magnitude and phase of the transient response to be quantified, and \(A_n\) and \(\phi_n\) are the magnitude and phase of the nontransient signal. Solving for the magnitude of the transient response that arises due to the heating event yields the following expression:

\[
A_n(t) = A_m(t)\cos(\phi_n(t) - \phi_m) - A_n\cos(\phi_n - \phi_n).
\]

(2)

The magnitude and phase of the signal are again obtained from the lock-in amplifier. The amplitude and phase of the nontransient signal can be obtained for the measured signal prior to the arrival of the pump pulse. The only missing piece of information is the phase of the transient response \(\phi_n\). The transient response of the film generated by the pump pulse is assumed to be in phase with the modulated pump beam. This is a reasonable assumption since the modulation time (1 \(\mu\)s) is much greater than the time of the event being measured (<1.5 ns). Therefore, a transient response occurring in the probe reflectance that is due to the heating from the pump pulse will be in phase with the modulated pump beam.

The phase correction model was used to calculate the magnitude of the transient response of the scans shown in Figs. 2 and 3. The results are shown in Fig. 4 where both scans have been normalized at the peak. Considering only the magnitude of the signal, these two scans seem very different, while in fact the two scans are almost identical in shape with only a slight nonlinearity that occurs at higher fluences.\(^{20}\)

The magnitude and phase of the signal can also be important on long scans. Figure 5 shows a scan taken on a 29 nm Pt film deposited on silicon. This type of scan is used to determine the thermal boundary resistance at the interface between the film and the substrate. In this scan, the signal
phase varies slowly by approximately 30° on a 1 ns delay. This slow change in the signal phase has a significant influence on the calculated magnitude of the transient signal. Using the uncorrected signal data, the thermal boundary conductance is calculated to be $1.02 \times 10^8$ W/m$^2$ K and displays a poor fit to the transient interface thermal model described by Stevens et al.\textsuperscript{11} Using the corrected data the thermal boundary conductance is calculated to be $1.48 \times 10^8$ W/m$^2$ K with excellent agreement between the data and the thermal model. Not accounting for both the magnitude and phase of the signal creates errors in the measured value of the thermal boundary conductance.

**IV. SIMULATED DATA AND PHASE CORRECTION**

As demonstrated in the previous section, accounting for the changes in the signal phase for both short and long scans can be critical. Although the previous examples provide strong evidence to support the phase correction technique, more justification for the technique would be valuable. To this end, the proposed phase correction technique is applied to simulated pump-probe data that account for residual signals, pump noise, and other electronic noise. Because the data are simulated using a known decay function, the phase corrected data can be compared to the known decay function to verify the technique’s validity.

To simulate pump-probe data, an approach and notation similar to the ones used by Capinski and Maris are used\textsuperscript{16} and depicted in Fig. 6. Assuming that pump pulses are square wave modulated at an angular frequency of $\omega$, the pump pulses arrive at the sample at

$$\left(l\tau + \tau_0\right)H[\sin(\omega(l\tau + \tau_0))],$$

where $\tau$ is the time between pulses, $\tau_0$ is time between modulation and the first pump pulse, and $l$ is an integer representing all pump pulses before and after $\tau_0$. $H[\cdot\cdot\cdot]$ is the Heaviside function. Note that a square modulation function is used rather than a cosine function, which is also the case for the experimental work presented earlier. A nonsquare modulation can lead to errors when testing materials with nonlinear thermoreflectance relationships, since pump fluences will vary during each modulation.

If the change in reflectivity of the sample due to a single pump pulse is given by $\Delta R(t)$, the overall reflectance of the sample with respect to time for square wave modulated pump pulses as depicted in Fig. 6 is

$$R(t) = R_0 + \sum_{l=0}^{\infty} H[\sin(\omega(l\tau + \tau_0))]\Delta R(l\tau - l\tau - \tau_0),$$

where $R_0$ is the equilibrium reflectivity coefficient.

If the delay between the pump and probe pulses is $\tau_{pp}$, then the reflected probe energy is proportional to

$$R_m = \left(R_0 + \sum_{n=-\infty}^{\infty} H[\sin(\omega(n\tau + \tau_0))]\right) \times \Delta R(\tau_{pp} + (m-n)\tau) E_{\text{probe}}$$

for each of the $m$ pulses at times

$$t_m = \tau_0 + \tau_{pp} + m\tau$$

for $m = 0, \pm 1, \pm 2, \pm 3, \ldots$, where $E_{\text{probe}}$ is the energy of each probe pulse. At all other times the reflected probe is zero.
Although much effort is often taken to reduce reflected pump pulses using polarizers and pinholes, a small amount of the scattered pump pulses can be observed at the detector, which occurs at the times defined by Eq. (3). Assuming the detector has a typical exponential decay with a time constant \( \tau_{\text{det}} \), the overall input signal to the lock-in is

\[
V_{11} = \frac{dV_{\text{det}}}{dP_{\text{det}}} \sum_{m} \frac{R_{m}}{\tau_{\text{det}}} \exp \left[ \frac{-(t-t_m)}{\tau_{\text{det}}} \right] H(t-t_m)
\]

\[
+ \frac{dV_{\text{det}}}{dP_{\text{det}}} \sum_{m} \frac{R_{\text{pump}}}{\tau_{\text{det}}} \exp \left[ \frac{-(t-t_n-\tau_0)}{\tau_{\text{det}}} \right]
\]

\[
\times H(t-n\tau-\tau_0)H[\sin(\omega(n\tau+\tau_0))],
\]

(7)

where \( R_{\text{pump}} \) is the amount of reflected energy of each pump pulse that is incident on the detector and \( dV_{\text{det}}/dP_{\text{det}} \) is the voltage output of the detector per incidence power.

The lock-in determines the in-phase component \( X \) by multiplying the input signal with a demodulation function at the reference frequency \( \omega \) and then passes the multiplied signal through a low pass filter. Often lock-ins do not use a pure sine wave, but either a square wave or a more complex function such as a Walsh function approximation of a sinusoid. To obtain the quadrature component \( Y \), the input signal is multiplied by a demodulation function at the reference frequency \( \omega \) with 90° phase shift and then passed through a low pass filter. See paper by Horowitz and Hill for details on lock-in theory.\(^{21}\) The signal amplitude and phase are then

\[
V_{\text{sig}} = \sqrt{X^2 + Y^2}, \quad \theta = \tan^{-1}(Y/X).
\]

(8)

The above lock-in procedure can be applied to the input signal defined by Eq. (7), where the key input parameters are \( \omega \), \( \tau \), and \( R_{\text{pump}} \) along with the optical reflectivity \( \Delta R(t) \). Up until now, no assumptions have been made regarding the form of \( \Delta R(t) \). To test the proposed phase correction technique we consider an optical response after the initial heating from a pump pulse of the form

\[
\Delta R(t) = \Delta R_1 \exp(-t/\tau_1) + \Delta R_2 \exp(-t/\tau_2) \quad \text{for} \quad t > 0,
\]

(9)

where \( \Delta R_1 \) and \( \Delta R_2 \) are the initial reflectivity associated with different decay processes with time constants \( \tau_1 \) and \( \tau_2 \), respectively. The first decay process might be associated with thermal boundary resistance and have a time constant of approximately 1–2 ns and for typical metals \( \Delta R_1 \) can be \( \sim 10^{-4}–10^{-3} \), while the second decay term might be associated with heat conduction in the substrate and be on the order of 10–1000 ns with \( \Delta R_2 \approx 0.1 \) to 0.01 \( \Delta R_1 \).

Figure 7 shows the simulated signal amplitude and phase for a reflectance response defined by Eq. (9) using \( \Delta R_1 = 10^{-3}, \tau_1 = 1 \) ns, \( \Delta R_2 = 10^{-5} \), and \( \tau_2 = 500 \) ns for a pump-probe experiment with a probe pulse frequency of 80 MHz and modulation frequency of 1 MHz. It is clear that the actual signal and simulated measured signal differ. Using the phase correction technique described in the previous section by Eq. (1) and a sine function as the demodulation function, the simulated signal can be phase corrected to obtain the actual signal, which is also shown in Fig. 7. For this particular case, if the measured signal was not corrected, the error in determining \( \tau_1 \) would be greater than 10%. If the functional form of the decay process is not known, the classical approach is to subtract the signal immediately before the arrival of the pump pulse from all the data and normalize the offset data. For this particular case the classical approach error in determining \( \tau_1 \) would be greater than 9%. This offset correction is also shown in Fig. 7.

To determine the regime for which the signal correction technique works a series of simulations was run for a range of fast and slow time constants and background noise conditions. For each simulation conducted, the signal was corrected and then was fitted to Eq. (9) where the time constants were free parameters. The error for the technique was obtained by comparing the fitted fast time constant with the actual fast time constant. The technique was extremely effective in removing both pump noise and any added rf noise for time decay processes that have time constants of interest which are less than 35% of the time between pulses, 0.35\( \tau \). The solid curve in Fig. 8, case 1, shows the percentage error in the corrected data versus the time constant of interest, \( \tau_1 \). There are no other noise or decay processes (\( \Delta R_2 = 0 \)). The time constant is nondimensionalized by dividing the time constant, \( \tau_1 \), by the time between pulses, \( \tau \). Below a time constant ratio of 0.35, the error is less than 5%. The same results for the phase corrected signal are obtained for a simulation where the pump noise is equal to the negative of the signal of interest (\( R_{\text{pump}} = -\Delta R_1 \)), case 2. The solid curve with the “+” in Fig. 8 is for the same case but with no phase correction.

The addition of a slow decay that falls between 0.35\( \tau \) and \( \sim 10/\omega \) can also cause errors if the data are not phase corrected. For slow processes that do fall in this range, the error is minimal as long as the change in reflectance magnitude associated with this decay process is small compared to that of the process of interest (\( \Delta R_2/\Delta R_1 < 0.01 \)). The dotted curves in Fig. 8, case 3, are the errors for the corrected, uncorrected, and classical offset corrected signals for
shown in Fig. 9. Again for decay processes where decay that is dependent on the fast decay process such as the phase correction technique. Consider a slow decay process, the model should work for a magnitude for the decay process of interest, then the error in the signal due to pump noise is less than 10% of the peak magnitude of the residual heating should be small compared to that of the heating of interest. Based on multiple simulations, if the lock-in signal when the probe is incident on the sample immediately before the pump pulse (t=0) minus the signal due to pump noise is less than 10% of the peak magnitude for the decay process of interest, then the error in determining the decay constant using the proposed phase correction method will be less than 5%.

Other decay processes were simulated to further validate the phase correction technique. Consider a slow decay process that is dependent on the fast decay process such as

$$\Delta R_2/\Delta R_1=0.01$$ and \(\tau_2\omega=\pi\) with no other signal noise. For \(\Delta R_2/\Delta R_1=0.001\), errors are indistinguishable from the case when \(\Delta R_2=0\).

Based on the simulations using exponential decay processes, the phase correction technique for pump-probe data is effective at reducing pump noise, rf noise, and noise associated with residual heating when the time constant of interest is less than \(\sim 0.35\) of the time between pulses. In addition, the magnitude of the residual heating should be small compared to that of the heating of interest. Based on multiple simulations, if the lock-in signal when the probe is incident on the sample immediately before the pump pulse (t=0) minus the signal due to pump noise is less than 10% of the peak magnitude for the decay process of interest, then the error in determining the decay constant using the proposed phase correction method will be less than 5%.

Other decay processes were simulated to further validate the phase correction technique. Consider a slow decay process that is dependent on the fast decay process such as

$$\Delta R(t) = \Delta R_1 \exp(-t/\tau_1) + \Delta R_2\left[1 - \exp(-t/\tau_2)\right]$$

This reflectance response results in nearly identical results and errors as the reflectance response defined by Eq. (9) and is restricted to the same regime. Similar results are obtained when simulating a Gaussian decay given by

$$\Delta R(t) = \Delta R_1 \exp[-(t/\tau_1)^2] + \Delta R_2 \exp[-(t/\tau_2)^2]$$

for \(t > 0\).

The errors in determining \(\tau_1\) for the response defined by Eq. (11) using uncorrected, phase corrected, and classical offset corrected signals for \(\Delta R_2/\Delta R_1=0.01\) and \(\tau_2\omega=2\pi\) are shown in Fig. 9. Again for decay processes where \(\tau_1 < 0.35\tau\), the phase correction technique works well with minimal error. Although the simulations were conducted for a variety of decay functions, the model should work for a range of unknown and arbitrary decay processes as long as the time scale associated with the decay of interest is less than 0.35\(\tau\).

V. MISALIGNMENT ERROR

Misalignment of the probe beam with the translation stage, a poorly collimated probe beam, or a warped delay stage can all result in misalignment of the pump and probe beams. These errors could lead to incorrect recording of the decay process of interest. The phase correction described above will not be able to remove errors associated with pump-probe misalignment, so other techniques should be used to quantify and minimize errors. One way to effectively avoid many of these errors is to incorporate a single-mode optical fiber, which is well suited for some applications. An alternative is to collimate the probe beam, carefully align the probe beam with the delay stage, and expand the pump beam spot size relative to the probe beam spot size. Enlarging the pump beam does reduce the signal to noise ratio as pointed out by Capinski and Maris. By setting the pump and probe beam spot diameters approximately equal, one must assume that the optical response is linear, which is not always the case. To avoid having to make this assumption, we often use a pump-probe radius ratio of \(\sim 5:1\).

To determine the error due to misalignment of the pump and probe spots, the signal will be proportionate to the correlation of the probe and pump responses:

$$\Delta R \propto \int_0^\infty f(r') g(r) \, dr,$$

where \(f(r')\) is the probe intensity function, \(g(r)\) is the optical response of the pump function, and \(R_{\text{error}}\) is the vector between the center of the pump and probe spot. Assuming a Gaussian profile for both probe and pump functions and a simple linear response with pump intensity.
\( f(r') \propto \exp\left(-\frac{2r'^2}{\omega_{\text{probe}}^2}\right) \),
\( g(r) \propto \exp\left(-\frac{2r^2}{\omega_{\text{pump}}^2}\right) \),

where \( \omega_{\text{probe}} \) and \( \omega_{\text{pump}} \) are the probe and pump radii at the sample. Changing to the Cartesian coordinate system and solving Eq. (12) leads to a signal proportional to

\[ \Delta R \propto \exp\left(-\frac{2R_{\text{error}}^2}{\omega_{\text{probe}}^2 + \omega_{\text{pump}}^2}\right) \].

For the case when the pump to probe radius ratio is 5:1, an error due to misalignment of the pump probe of less than 3% can be obtained by ensuring that the magnitude of \( R_{\text{error}} \) is less than 1/8 of the pump radius.

To measure the radius and location of the probe beam, a knife edge can be placed in the sample holder. The sample holder can be moved in both axes perpendicular to the probe beam. By sweeping the knife edge across the path of the focused probe beam while measuring the transmitted probe power, the probe spot radius and center can be determined. This technique is described in greater detail by Suzuki and Tachibana.\(^{22}\) The transmitted laser power for a knife-edge sweeping across a Gaussian spot is given by \(^{22}\)

\[ P(x) = P_0 \sqrt{\frac{2}{\pi}} \int_{-\infty}^{\infty} \exp\left[-\frac{2x^2}{\omega^2}\right] dx = P_0 \frac{\text{erfc}(\beta)}{2}, \]  

where \( P_0 \) is the total laser power, \( x_0 \) is the location of the center of the beam, and \( \beta = \sqrt{2(x-x_0)}/\omega \).  

The spot radius and center can be determined by sweeping a knife edge across the spot and measuring the transmitted laser power. The measured power is directly related to \( \beta \), Eq. (17), which in turn can be plotted versus \( x \) to determine \( x_0 \) and \( \omega \) using Eq. (18).

VI. SUMMARY

A straightforward technique for removing both residual heating and pump scatter noise for transient thermoreflectance measurements has been developed. The correction technique has been applied in measuring electron-phonon coupling rates,\(^{20}\) thermal diffusivity in thin metal films,\(^{23}\) thermal boundary resistances,\(^{11}\) and ultrafast carrier dynamics in semiconductors.\(^{24}\) The technique does not require knowing the functional form of the residual heating, unlike previous methods. The technique was verified using simulated experimental data and shown to work well for decay processes with time constants that are less than 35% of the time between pump pulses. The impact of pump-probe misalignment on the TTR signal was quantified, and the impact of optical misalignment errors over the entire range of pump-probe delay times was demonstrated.

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