Laser Repetition Rate in Time-domain Thermoreflectance Techniques

L.S. Larkin ^{*,a}, J.L. Smoyer ^{*,b}, P.M. Norris ^{** ,a}

^a*Department of Mechanical and Aerospace Engineering, 122 Engineer's Way,
Charlottesville, VA, 29903*

^b*Department of Physical Sciences, Kutztown University, Kutztown, PA 19530*

^{*}Corresponding author

^{**}Principal corresponding author

Email addresses: `ls19hd@virginia.edu` (L.S. Larkin ^{*,a}), `jsmoyer@kutztown.edu`
(J.L. Smoyer ^{*,b})

Preprint submitted to Elsevier

February 8, 2017

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^c*Department of Mechanical and Aerospace Engineering, 122 Engineer's Way,
Charlottesville, VA, 29903*

^d*Department of Physical Sciences, Kutztown University, Kutztown, PA 19530*

Abstract

Pump-probe thermoreflectance techniques have been utilized extensively to measure the thermal properties; such as, thermal conductivity and interfacial conductances of thin film systems. While there have been many implementations of transient measurements, the most commonly operated systems utilize a near-IR femtosecond pulsed system as the fundamental laser source. The repetition rate of these femtosecond pulsed systems can vary from kHz to MHz. Thus, we perform an in-depth comparison of the implementation and analysis of low-repetition rate and high-repetition rate time-domain thermoreflectance (TDTR), demonstrating that despite varying the repetition-rate, the measured thermal properties are consistent. To illustrate the universality of these systems, we selected a range of materials to study on both TDTR set-ups, including the thermal conductivity of a SiC film, a SiO₂ film, and the interfacial conductances of Al/SiO₂ and Al/SiC systems.

Keywords: Time-domain Thermoreflectance, Thin film thermal properties

1. Introduction

Thermoreflective spectroscopy first appeared in the literature in the 1970s, where modulated continuous wavelength (CW) lasers were used to measure the photoacoustic spectra of solids and the thickness of opaque films [1, 2].

^{*}Corresponding author

^{**}Principal corresponding author

Email addresses: ls19hd@virginia.edu (L.S. Larkin ^{*,a}), jsmooyer@kutztown.edu (J.L. Smoyer ^{*,b})

Over the next decade, these non-contact techniques began to incorporate pico- and femtosecond pulsed lasers to investigate a range of phenomena, such as non-equilibrium dynamics [3, 4], optical constants [5], coherent acoustic phonons [6, 7], and heat diffusion in thin films [8, 9, 5]. Today, transient thermoreflective (TTR), or as it has come to be called, time-domain thermoreflectance (TDTR), is one of the most powerful and versatile techniques to extract thermal properties of multi-layered thin film systems [10, 11, 12].

Most current TDTR systems use a high-power, high-repetition rate, Ti:sapphire oscillator as the primary pulsed laser source [10, 12, 11]. These systems further modulate the pump beam on the order of 100s of kHz to several MHz, which permits several successive heating events, with insufficient cooling time between events to return the system to equilibrium, during the “ON” portion of the modulation window. In this work, this type of system will be referred to as a high-repetition rate, or “high-rep”, TDTR system. In order to better study non-equilibrium thermal phenomena, such as electron-phonon coupling, the basic high-repetition rate laser oscillator is often coupled with a regenerative amplifier to increase the pulse energy at the expense of the repetition rate [13, 14, 15]. These low-repetition rate systems typically operate at frequencies on the order of several 10s to several 100 kHz. In this scheme, only a single heating event occurs within the modulation window and the system is allowed to return to thermal equilibrium before the arrival of the next pump pulse. In the following work this one-shot-on/one-shot-off system is classified as a low-repetition rate, or “low-rep” TDTR system. In addition to non-equilibrium dynamics, recent work has shown low repetition systems are ideal for probing semiconductor surfaces directly [16].

Several prior studies have shown that the beam diameter and modulation frequency can impact the measured thermophysical properties in TDTR experiments, and care must be taken to ensure an accurate analysis is done [17, 18, 19, 20]. Despite this collection of work, there has never been a work which has analyzed the overall repetition rate of the fundamental laser system. With an increasing number of facilities using TDTR, it is critical to establish the universality of the technique regardless of the repetition rate of the fundamental laser, a defining characteristic of a pulsed laser system. Therefore, we used two laser systems, varying only in repetition rate, one of high repetition rate (80 MHz) and one of low repetition rate (250 kHz), to measure the thermal conductivity, κ , two materials (an oxide layer and a conductive substrate), and the thermal boundary conductance, h_{BD} , at two interfaces (a conductive metal/semiconductor, and a metal/oxide/semiconductor system).

2. Time-domain ThermoReflectance

A basic schematic of a TDTR system, regardless of laser repetition rate, follows the layout in Fig. 1. A pulsed laser source is split into the high energy pump beam and the lower-energy probe beam. The pump beam is further modulated, through an electro-optical modulator (EOM) or equivalent equipment, to a frequency distinct from the fundamental laser frequency, and then directed onto the sample to induce a small heating event on the order of 1 K. The probe is delayed temporally with respect to the pump via a mechanical delay stage, and then directed onto the sample. The reflected probe beam from the sample surface is then collected via a photodetector and the resultant signal processed by a lock-in amplifier detection scheme. The reflectivity of a material is, in general, correlated to the temperature of the material through the thermorefectance coefficient. Thus, the intensity of the reflected probe, shown in Fig. 2, after the thermal excitation of the pump beam can be used to deduce the temperature profile of the system. The data is then compared to a thermal model, to be discussed in the following sections, and unknown variables are left as free parameters and varied until the sum of the least-squares differences between the data and model have been minimized.

Figure 1: The basic schematic of a TDTR experimental set-up.

2.1. Low-Repetition Rate

The low-repetition rate set-up used in this study is centered around a high-repetition rate (76 MHz) Coherent MIRA 900 Ti:sapphire oscillator, the output of which is seeded into a Coherent RegA 9000 Ti:sapphire regenerative amplifier. After amplification, the emerging pulses have increased in energy from 9 nJ/pulse to around 4 μ J/pulse, at a wavelength of 800 nm, a pulse duration of less than 200 fs, and at a repetition rate of 250 kHz. The pump beam is further modulated to 125 kHz by a Conoptics EOM. In this system, the pump and the probe are spatially separated from one another in order to remove backscattered pump from the measured signal.

In order to infer the material's thermal properties, the temperature (reflectivity) on the sample surface must be related to a temperature gradient throughout the material. The thermal model used to analyze the data differs depending on the temporal regime of the TDTR scan. In the non-equilibrium regimes, thermalized electrons begin to cool by exchanging energy with the cold lattice system via electron-phonon scattering. The cooling of the electrons can be described by the two-temperature model (TTM), which couples the electron and lattice temperature [14]. Under the TTM, the interaction between electrons and phonons is diffusive, but ballistically traveling electrons can influence the resulting temperatures [21]. However, as we are interested in steady-state thermal properties, such as κ and h_{BD} , we will focus on the transient regime of the TDTR scan, *i.e.*, times greater than ~ 0.5 ns. At this time scale, the phonons and electrons (to a lesser degree in semiconductors) dominate the thermal transport through diffusive scattering processes [22, 12]. Therefore, the heat diffusion equation can be used to relate the measured surface temperature to the thin film's thermal properties [23]. The heat diffusion equation is solved through a Crank-Nicolson (CN) method. CN is a finite difference method which is numerically stable only when the value $\alpha(\delta t/\delta x^2)$ is large, where α is the thermal diffusivity, δt is the discrete time step, and δx is the discrete spatial step [24, 25]. Thus, small spatial and temporal steps must be used for an accurate analysis, and as with any finite element analysis model, the computational power required scales with the number of discrete steps needed. In order to greatly increase the speed of our analysis, a one-dimensional heat diffusion equation was utilized. At low-frequencies, a one-dimensional analysis can only be applied when the pump's diameter is significantly larger than the probe. In this experiment, we maintain a 5:1 ratio, which is sufficient to assume one-dimensional heat transfer [17]. In the following sections, the 1-D heat equation was solved

using CN in the steady-state regime. The unknown thermal properties, such as the thermal conductivity of a substrate or the thermal boundary conductance, are left as free parameters. Using a non-linear least-squares regression analysis, we vary the unknown properties until the residuals are minimized.

2.2. High-Repetition Rate

The high-repetition system utilized in this work is primarily the same as described above, without the amplified cavity. The femtosecond pulsed laser used is a Spectra Physics Tsunami, which operates at a repetition rate of 80 MHz, a pulse width less than 200 fs, and an energy/pulse around 30 nJ. After emanating from the cavity, the pump beam is further modulated to 1 MHz. The primary difference between high and low-rep rate systems is a phase, or imaginary component, of the measured data. In low-rep systems, the sample returns to thermal equilibrium, following a pump pulse, before the arrival of a subsequent pump pulse. Consequently, the signal arriving at the detector is from one heating event, and therefore has a constant phase. In high-rep systems, the sample does not have adequate time to cool down completely before the next heating event is created and there is a resulting thermal accumulation on the sample surface, producing a changing phase signal. The phase signal has been used to provide additional sensitivity to thermal properties, such as radial thermal conductivity in anisotropic systems [26].

A similar analysis as described above for the low-repetition system is used in high-repetition systems, where a thermal model is compared to the data and unknown properties are varied until the difference between the model and data is minimized. The thermal accumulation on the sample surface complicates the analysis so that we cannot just discretely solve the 1D heating equation. Instead, we must account for the sinusoidal nature of the heating event, and can use a Feldman analysis for heat flow [27]. Again, the modulation frequency must be considered when determining the relative pump and probe sizes to insure cross-plane heating. In this study, a $1/e^2$ diameter of 48 μm and 19 μm were used for the pump and probe, respectively.

3. Results and Discussions

3.1. Case Study: SiC and Oxide Layer

Silicon carbide (SiC) is a wide-bandgap semiconductor with a large thermal conductivity and breakdown field. These properties make SiC desirable for high-power electronic devices operating in high-temperature environments [28, 29]. While SiC possessing a native oxide, SiO₂ makes it a great candidate in MOS-devices, it also produces the potential to substantially impact the resistance of the interface, which could lead to a build-up of energy carriers at an interface [30]. The presence of an oxide at the Al/Si interface has been shown to reduce the thermal boundary conductance by a factor of six at room temperature [31, 32]. Here, we report on the thermal boundary conductance, h_{BD} , at room temperature ($T=300$ K) at an Al/SiC interface and an Al/SiO₂/SiC interface.

N-doped 4H-SiC was grown epitaxially on a substrate of SiC. The films of SiC are 11 μm thick with doping concentrations of 10^{15} cm^{-3} . The SiC was thermally oxidized, and then annealed in an ambient NO environment at 1175°C. Annealing in an NO process has been shown to remove excess carbon at the SiC/SiO₂ interface, which produces a large density of interface states that can result in charge trapping and Coulomb scattering [33, 34]. The surface of the SiC substrates were then etched with buffered oxide etch (BOE) 7:1. The sample was then cleaved into several pieces. Part of the sample was immediately put in an electron evaporation chamber to deposit a nominal thickness of 100 nm of polycrystalline Al. On the other sample, a nominal thickness of 40 nm of SiO₂ was grown through plasma-enhanced chemical vapor deposition (PECVD). N₂O and 2% silane (SiH₄) were maintained at a 2:1 flow rate, the pressure of the chamber was kept at 250 mTorr, and the growth temperature was 270°C. With these growth parameters, the growth rate was ~ 30 nm/min. The thickness of the PECVD SiO₂ film was measured with ellipsometry and determined to be 40 ± 1 nm, and the index of refraction was measured to be around 1.43-1.45, just below the bulk SiO₂, indicating a slightly less dense film. The films were then metallized with 100 nm of Al grown via electron beam evaporation. The samples were then thermally characterized with both the high-rep and low-rep laser systems. Six scans were collected on each of the samples at room temperature and the values of thermal boundary conductance and thermal conductivity represent the average of those six individual fit values. The error bars on the data points

represent the standard deviation of those six values, plus the effect of an additional 2 nm uncertainty in the thickness of the Al transducer.

Picosecond acoustics were collected during TDTR analysis, and used to determine the local thickness of the Al films [35, 36]. In the thermal model, bulk specific heat values for the Al and SiC were assumed and a reduced thermal conductivity was assumed for Al (due to finite size and boundary scattering), and the values used can be found in Refs. [37, 38, 39]. We also left the thermal conductivity, κ_{SiC} as a free parameter for the 2 layer system, Al/etched SiC, and it was determined to be 323 ± 2.0 W/mK in the low-rep system, and 322 ± 9.6 W/mK using the high-rep system. These results are within 10% of the measured value by Ref. [37], which is reasonable for a doped sample. The deviation between individual measurements was much larger for the high-rep system, and we believe this is due to differences in the experimental sensitivity of the two systems. The experimental sensitivity, S , of a particular parameter, α , is defined as,

$$S_{\alpha} = \frac{d(\ln(V))}{d(\ln(\alpha))} \quad (1)$$

where V is the signal from TDTR [40]. In the low-rep system, we are very sensitive to both thermal conductivity of the substrate, κ_{SiC} , and the thermal boundary conductance, $h_{BD,Al/SiC}$, but in the high-rep system, the sensitivity to κ_{SiC} is overshadowed by the sensitivity to $h_{BD,Al/SiC}$, see the inset of Fig. 3. This is likely due to a much larger thermal penetration depth in the low-rep system, since modulation frequency is inversely proportional to thermal penetration depth [18]. In Fig. 3, $h_{BD,Al/SiC}$ is shown for both laser systems, the high-rep and the low-rep.

In the three layer system, Al/PECVD SiO₂/SiC, the thermal conductivity and specific heat of the PECVD film must also be determined since the transport properties of SiO₂ have been shown to be sensitivities to processing parameters [41]. Again, the differing experimental sensitivity of the two systems is of importance here. In the high-rep system, the experimental sensitivity is dominated by the SiO₂ layer, and we are not sensitive to the SiC substrate, but in the low-rep system, we are sensitive to all three layers due to an enhanced penetration depth. Therefore, in the high-rep system, we fit the specific heat of SiO₂, the thermal conductivity of the SiO₂, and the thermal boundary conductance of Al/SiO₂. In the low-repetition system, we treat the interfaces, as well as the SiO₂ layer as a series of resistors in order to lump all the unknowns into a single unknown. We also leave the

thermal conductivity of the SiC layer as an open parameter in the low-rep system as a method for checking the accuracy of our fits. The thermal conductivity of the SiO₂, $\kappa_{SiO_2} = 1.26 \pm 0.1$ W/mK and the specific heat of the SiO₂, $C = 2.05 \times 10^6$ Jm⁻³K⁻¹, as determined from the high-repetition fits. These values are very close to other studies of thermal properties in PECVD SiO₂ [41]. In the low-repetition system, the thermal conductivity of the SiC substrate was determined to be $\kappa_{SiC} = 324 \pm 5$ W/mK. For the three-layer system in low-rep analysis, we are significantly more sensitive to the net resistance of the Al/SiO₂ relative to the SiC system, so the error bars on this measurement are larger than the two layer, low-rep, measurement of κ_{SiC} , but the value is within the error bars of our 2-layer measurement, and therefore, the measurement of the net resistance is justified. In Fig. 3, the net conductances of the low-rep and high-rep system are shown. In the three layer system, the total conductance is given by Eqn. 2.

$$h_{total}^{-1} = h_{BD,Al/SiO_2}^{-1} + h_{SiO_2}^{-1} \quad (2)$$

The conductance of the SiO₂ layer is given by κ_{SiO_2}/d_{SiO_2} . The two fit values are almost identical, but the error bars for the high-rep system are larger due to more parameters being left as free parameters. These results demonstrate that the two major classification of TDTR systems, a single shot (low-rep) and a high-rep modulated response yield the same thermophysical properties, and only impact the sensitivity of the results. If the user is careful to ensure the beam parameters have been set-up properly and the correct analysis is being used, the thermophysical properties should not vary between a low-rep and high-rep system. As long as there is sufficient sensitivity, compared to the noise of the system, there is no reason to anticipate the thermal properties being dependent on the repetition rate of the system. As seen in Fig. 3, the presence of the PECVD oxide lowers the conductance by almost exactly 1 order of magnitude. This reduction agrees with literature that the presence of an oxide layer, particularly one with minimal intermixing at the interface, greatly reduces the interfacial conductance [31, 42, 43].

4. Conclusion

In this note, we establish the robustness of transient reflectance measurements using two femtosecond pulsed laser systems with different repetition rates, one high in the MHz range and one low, in the kHz range. Both

systems were used to measure the thermal boundary conductance of a resistive metal/oxide and a metal/semiconductor, the thermal conductivity of a highly conductive substrate, SiC, and the thermal conductivity of a resistive oxide, SiO₂.

The same properties were measured on both systems, but with different error bars associated with the measurements. We attribute the deviation in error bars to the differences in signal filtering. These results establish the versatility and universality of TDTR measurements.

Acknowledgment

The authors would like to thank J.L. Hostletler for the SiC samples. The authors would also like to acknowledge B.M. Foley and P.E. Hopkins for providing us with a high-repetition laser system to use. L.S. Larkin is appreciative of funding through the National Science Foundation's Graduate Research Fellowship Program. Both authors would like to acknowledge the financial support of the Air Force Office of Scientific Research under Grant No. FA9550-14-1-0395

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Figure 2: A sample time-domain thermoreflectance, TDTR, scan with the primary temporal regimes labeled. The first regime, the hot electron thermalization and electron-phonon coupling regime, occurs immediately after the initial excitation of the material system by the laser. Data from this regime can be used to determine a film's electron-phonon coupling constant, as well as its optical constants. In the second regime, the picosecond acoustic regime, the data is influenced by the effects of thermal expansion in the thin metallic film following the rapid thermal excitation. These picosecond acoustic response can be used to determine the thickness within the metal film, if the speed of sound in the film is known. In the third region, heat transfer is dominated by diffusive transport, and the steady state thermal properties, such as film thermal conductivity, diffusivity, effusivity, and thermal boundary conductance can be determined. The solid blue line shows the resulting thermal model for this data. In the inset of this figure, the real component of the reflectance signal collected from the high-rep and low-rep systems are shown in black and red, respectively.

Figure 3: The thermal boundary conductance, $h_{BD,Al/SiC}$, and the total conductance, given by Eq. 2, of an Al/PECVD SiO_2/SiC system. The measurements are shown as a function of oxide thickness, and the etched sample (Al/SiC) has a 0 nm oxide thickness, and the three-layer Al/PECVD SiO_2/SiC system has a 40 nm oxide thickness. The measurements were performed on both a low-repetition system, described in Sec. 2.1, marked 'Low-rep', and shown in a solid-black square, and a high-repetition system, where the set-up of this system is described in Sec 2.2, and shown here in the figure as open diamonds. The oxide layer lowers the conductance by an order of magnitude. Despite the differing experimental sensitivity of these systems, both the high-rep and low-rep system measurements are within the error bars of one another, demonstrating the robustness of TDTR. In the inset of this figure, the sensitivity for selected parameters of the 2-layer Al/(etched)SiC are shown for both the high-rep and low-rep system, labeled h.r. and l.r., respectively. The substantial impact of frequency on experimental sensitivity is quite pronounced. In the high-rep system, the sensitivity to $h_{BD,Al/SiC}$ greatly overshadows the sensitivity to κ_{SiC} . On the other hand, the sensitivity to $h_{BD,Al/SiC}$ and κ_{SiC} are approximately the same for early parts of the scan, but the sensitivity to κ_{SiC} begins to surpass at later times in the scan, when the laser has begun penetrating the SiC substrate.