

between changes in reflectivity and changes in temperature has to include the range of transient temperatures experienced by the surface during the measurements. Otherwise, nonlinear effects could entirely distort the transient temperature response of a sample during analysis. Another difficulty connected with optical properties of the top layer is the changing of the optical properties with time due either to long-term oxidization at room temperature or to accelerated oxidization at the higher temperatures experienced during laser irradiation pulsing. These chemical modifications of the top layer add uncertainty to the measurement procedure since they are not easily quantifiable.

In order to eliminate these difficulties, investigators have resorted to the use of a so-called metal “absorption” layer on top of the material under test (for instance Au in [6], and Al in [7]). Metal films are used because they exhibit high absorptivity and their optical properties are usually well known. Although the use of other metals as an absorption layer has not been widely considered, gold seems to be a particularly attractive material for use as an absorption film because of good stability in its refractive indexes (n and k) in a normal laboratory environment, linear dependence between reflectivity and temperature changes in the range of up to 200 K, and sufficiently high thermorefectance coefficient.

Because of the transient nature of the laser heat source in the TTR method, the duration of a laser pulse and the wavelength of the laser light are essential parameters for analyzing the applicability of the TTR method to a particular situation. Although a number of publications have reported on the use of the TTR method for measuring different composite materials [8], a systematic investigation of the influence of the essential parameters in the TTR method on the uncertainty of the measured thermal conductivity of bulk semi-infinite layer samples has only recently been studied by the authors [9]. The present investigation represents the next step in the complexity spectrum of a sample under test, and is focused on the TTR measurements of samples that have at least two layers, one of which is the absorption layer. This work is taking into consideration the influence of the absorption layer on the performance of the thermal conductivity measurements by analyzing both the experimental results for the thick silicon dioxide layer covered by a Au/Al film and the numerical simulation results for the heat transfer through the layered sample. Special attention has been given to the influence of the absorption layer thickness on the temperature response of the sample and the performance of the TTR method.

2. Experimental procedure

The existing experimental TTR system at the SMU SETS Laboratory is depicted schematically in Fig. 1. The

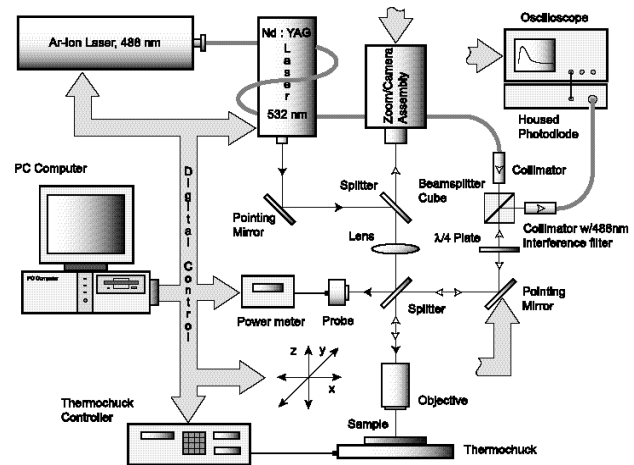


Fig. 1 Schematic of the experimental setup
(<http://www.engr.smu.edu/sets/>)

heating source is provided by an Nd:YAG pulsed laser whose wavelength is 532 nm, pulse width is 8.6 ns, and maximum pulse energy is 0.5 mJ. The laser power and output aperture are computer controlled, but the actual energy level delivered by each pulse is also measured by a power meter. The heating spot of the YAG was characterized by CCD imaging and fast photodiode detection, and was found to have good spatial uniformity and a Gaussian temporal distribution.

The probing light source is an Ar-Ion CW laser with a linearly polarized, single-mode irradiation beam at a wavelength of 488 nm. The beam is delivered to the microscope assembly via a polarization preserving, fiber optic cable with TEM_{00} mode. The microscope objective lens focuses the laser light on the sample surface concentrically with the heated spot. The probing beam reflects from the heated surface back along its optical path to the sensitive area of a pre-amplified silicon PIN photodetector (rise time 1 ns) through a fiber optic cable. The intensity of the reflected light depends on the reflectivity and temperature of the sample’s surface. The photodetector signal, representing the variations in surface reflectivity, is acquired with an 8-bit resolution via a digital oscilloscope at a rate of 2 Giga-samples per second. Several microscope objective lenses are available, but the one used here is 20X, providing heating and probing spots whose diameters are 226 μm and 2.4 μm , respectively, on the surface of a sample under test.

3. Results and discussion

Three length scales are sufficient to uniquely describe the heat transfer problem during pulsed laser heating in the TTR method; namely, the thickness of the absorption layer, h ; the optical penetration depth of the heating light, d_L ; and the heat penetration depth during the pulse width, d_H . However, as previous discovered [9], the ratio

between the two latter parameters entirely defines the transient temperature response of a bulk sample. Since the only absorption layer material considered in the present article is gold, it is reasonable to assume that the heat penetration depth is much bigger than the light penetration depth, i.e., $\tilde{a}_H \gg \tilde{a}_L$ (this assumption is also applicable to all metals subjected to nanosecond-pulse laser heating). It is equally reasonable to consider that the physical thickness of the absorption layer (e.g., gold) is bigger than the light penetration depth, i.e., $h > \tilde{a}_L$. Otherwise, the top layer would be inappropriate for light energy absorption within the scope of the TTR measurement approach. Therefore, if one wishes to analyze the heat transfer process in a gold-covered sample, only two length scales need be considered: h and d_H . After the introduction of the above reasonable limitations for the geometrical parameters of the problem, one can consider two ranges of variation for h with markedly differing temperature responses. The heat transfer process in the two above-defined thickness ranges of the top “absorption layer” is discussed next.

In categorizing the heat transfer process in a layer of material, a distinction is made between layers that are *thermally-thin* and layers that are *thermally-thick*. When the thickness of the absorption layer is larger than the heat penetration depth, the layer has sufficient internal thermal resistance to support temperature gradients, i.e., the gold layer behaves as a thick plate. Thus, such class of absorption layer is referred to as *thermally-thick* layer.

In related work, the authors [9] carried out an investigation in order to differentiate between “*semi-infinite layer*” and “*finite layer*” bulk samples. The fundamental premise was that, depending on the particular TTR setup used, even a sample made up of multiple layers of materials can be considered as being a *semi-infinite layer* sample if the depth of the top layer is sufficiently thick to account for all of the temperature gradients experienced as a result of a heating laser pulse. The investigation yielded a criteria for estimating the minimum thickness of the top layer, h^* , for which a sample can be classified as a *semi-infinite layer* sample.

For the case of gold, h_{Au}^* would be equal to $6.35 \mu\text{m}$.

In this work, a comparison is made between the temperature responses of two samples of thick SiO_2 , one covered with a (hypothetically) infinite thickness of gold and other with a layer of gold whose thickness is exactly $h^* = 6.35 \mu\text{m}$. The results of this comparison, obtained by numerical simulation, are shown as the lower two curves in Fig. 2. Some discrepancy can be observed between these two curves toward their tail ends, which is due to the influence of the SiO_2 on the heat transfer through the finite gold layer. Nevertheless, it is clear that the gold layer controls the majority of the temporal variations in temperature on the surface of the sample. Thus, the

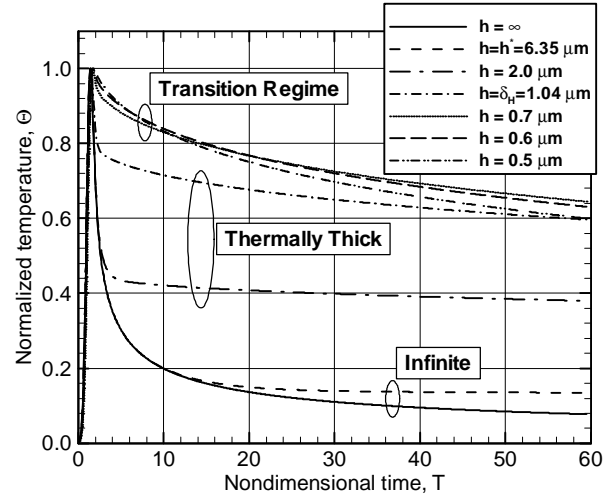


Fig. 2 Thermally-thick Layer: Temperature responses of an SiO_2 bulk sample covered with Au, whose thickness is larger than the heat penetration depth

thickness h^* defines the maximum possible thickness of gold to be used as an absorption layer; thicker layers of Au will hide the influence of the thermal properties of any underlying material on the surface temperature response in the TTR method.

Two typical normalized temperature responses for thermally-thick layers of gold ($h_{Au} = 2 \mu\text{m}$ and $1.04 \mu\text{m}$) are also shown in the middle part of Fig. 2. In both curves, the temperature decay exhibits a sharp change of shape at the time when the heat front reaches the less thermally conductive oxide layer. The cause for this marked shift is the high temperature gradients developed at the interface between the highly conductive gold layer and the more resistive silicon oxide layer. It is also worthwhile to note that more time is required for the heat front to reach the underlying oxide layer when the thickness of the absorption layer is increased. At the upper limit, when the thickness of the absorption layer is bigger than h^* , the response shows that there is very little, if any, heat flow in the silicon oxide layer, indicating that the bulk limit of the material has been reached.

In the discussion above, the thermal responses were analyzed as the thickness of the Au absorption layer was decreased from infinity to $1.04 \mu\text{m}$. While the curve for $h \rightarrow \infty$ represents the lower limit for the thermal response at the surface of the sample, the three upper curves in Fig. 2 ($h = 0.5, 0.6, \text{ and } 0.7 \mu\text{m}$) represent the upper range. Further reductions in the thickness of the gold absorption layer will produce curves that fall below this upper range, depicting the behavior of *thermally-thin* absorption layers. The *thermally-thin* and *transition* regimes are discussed later.

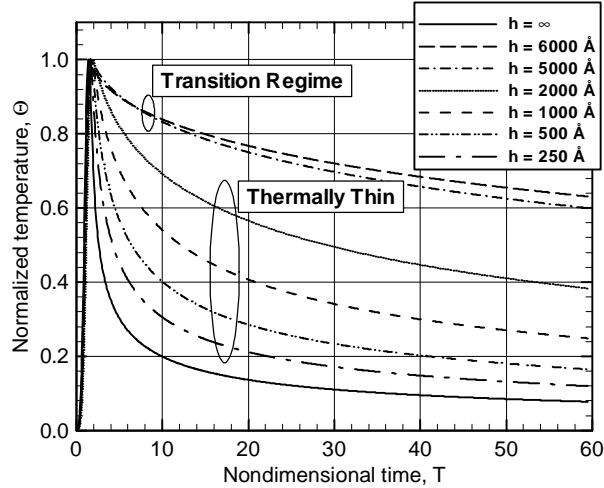


Fig. 3 Thermally-thin Films: Temperature responses of an SiO_2 bulk sample covered with Au, whose thickness is smaller than the heat penetration depth

When the thickness of the absorption layer is smaller than the heat penetration depth during the heating pulse, the layer has insufficient internal thermal resistance to support temperature gradients, and as a result, the instantaneous temperature field is relatively uniform throughout the material. Consequently, this type of absorption layer will be referred to as *thermally-thin* (also widely known as “lumped capacity”) layer.

Characteristic temperature responses for *thermally-thin* layers are shown in Fig. 3, bracketed from above by the curve for $h = 0.6 \mu\text{m}$ and from below by the curve for $h = \infty$. The latter curve is included for reference purposes and may not necessarily represent the absolute limit for all materials. The temperature decay is visibly faster as the thickness of the absorption layer decreases and all temperature responses lie above the response for bulk gold. The light penetration depth of the heating laser, d_L , is the lower limit for the thickness of an absorption layer. Therefore, layers thinner than d_L are not considered since they are not physically possible with the TTR method.

The discussion below can benefit from the use of the Fourier number defined as $Fo = at/h^2$ where a is the thermal diffusivity of gold and t is the pulse width of the heating laser. Taking the reciprocal of the square root of the Fo number (i.e., $Fo^{-1/2}$) represents the ratio between the thickness of the absorption layer, h , and the heat penetration depth during a single laser pulse width, d_H . The computed normalized temperature responses for gold-covered silicon oxide are plotted in Fig. 4, where a non-dimensional time based on the pulse width has been introduced, such that $T = t/t$. In these plots, the temperature responses from Figs. 2 and 3 are shown at specific time instances, beginning with a time equal to

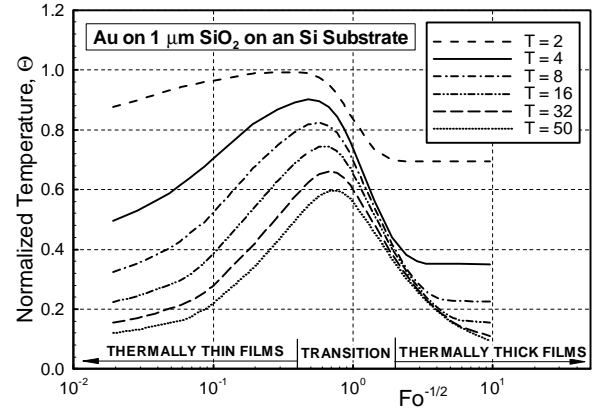


Fig. 4 Heat transfer regimes in gold covered silicon oxide samples

twice the pulse width, i.e., $T = 2$, and ending with $T = 50$. This view of the results makes it possible to more easily identify the three different regimes which are entirely defined by the nondimensional number $Fo^{-1/2}$. Behavior consistent with the *thermally-thick* regime appears for $Fo^{-1/2} \gtrsim 2$, while behavior consistent with the *thermally-thin* regime occurs for $Fo^{-1/2} \lesssim 0.4$. A *transition* regime between the *thermally-thin* and *thermally-thick* regimes is evident. For the specific materials considered here, this transition occurs in the range of $0.4 \lesssim Fo^{-1/2} \lesssim 2$.

To validate the results presented above, a one micron-thick silicon oxide layer was grown on five standard, 4-inch, silicon wafers and then covered with a metallic layer by a process of chemical vapor deposition. As previously discussed, the metal of choice for the top layer is gold since gold is chemically stable, has known optical and thermal properties, and has a high thermal reflection coefficient. However, very large thicknesses of gold are inherently difficult and expensive to deposit. Alternatively, a thinner layer of gold can be deposited on a thicker layer of aluminum, creating the desired metallic thickness while preserving the advantages of the use of gold as a surface layer. In order to assess the influence of the use of some aluminum in the metallic layer instead of using solely gold, numerical simulations were performed for the scenario of pure gold and a corresponding case of gold on top of aluminum. The simulation results showed that the differences between the temperature responses for pure gold and gold on aluminum were negligible since the two metals have similar thermal properties.

The experimental and corresponding numerical results are presented in Fig. 5. The numerical methodology consists of solving the heat transfer problem for each given finite layer configuration and varying the unknowns until the RMS error between the numerical and experimental curves is minimized. Typically, the numerical solution is sought to determine the unknown

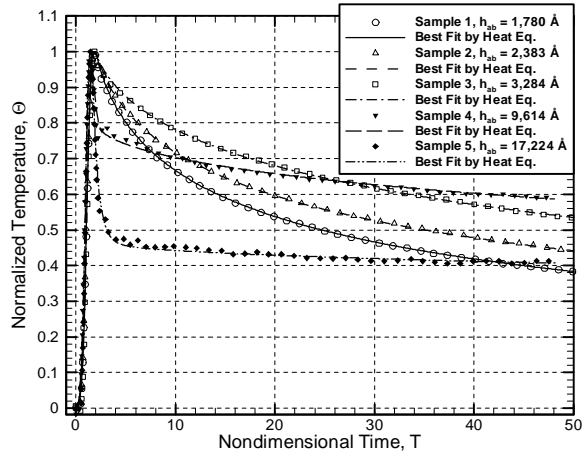


Fig. 5 Measured and computed temperature responses of an SiO₂ layer covered by different thicknesses of metal

thermal property of an underlying layer as well as a second variable, which can be either the thickness of the absorption layer or the thermal interface resistance between the cover layer and the underlying layer. Since in this investigation, the thermal properties of all the materials are known and the thermal interface resistance is negligible as compared to the resistance of the silicon dioxide layer, the only remaining variable to be determined by the optimization routine is the thickness of the absorption layer.

The behavior of the temperature responses of samples 1-3 is consistent with the previously discussed behavior of thermally-thin absorption layers (Fig. 5). In other words, the decay of the normalized temperature does not exhibit a sharp change of slope. Samples 4 and 5 exhibit a behavior consistent with the thermally-thick regime whereby the diffusion of heat through the underlying oxide layer is discernable by the presence of an abrupt change of slope at small values of T ($T \cong 2.5$ for sample 4 and $T \cong 3.5$ for sample 5).

To further assess the performance of the TTR method, it is useful to introduce a parameter whose value could directly characterize the accuracy of TTR measurements. We suggest the use of the *responsivity*, R_s , of the thermal conductivity measurement defined as $R_s = K d\Theta/dK$, where Θ is the normalized temperature response of the sample surface and K is the thermal conductivity of the material. Indeed, R_s is directly connected with the accuracy of the method by the equation $s_K = R_s^{-1} s_\Theta$, where s_K is the random measurement uncertainty of the thermal conductivity, K , and s_Θ is the random apparatus uncertainty of the TTR system. The latter equation shows that the measurement uncertainty of the TTR technique decreases with increases in the responsivity value, R_s .

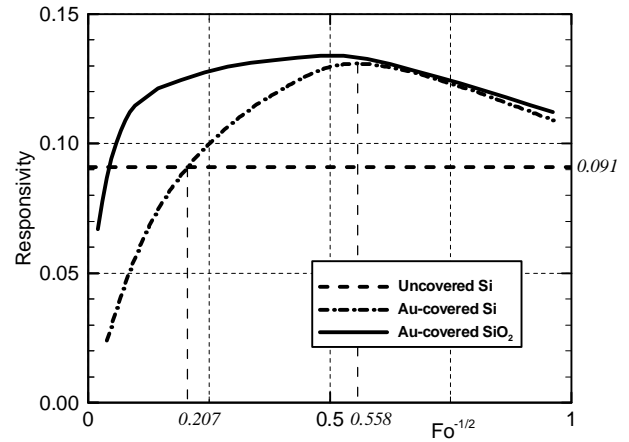


Fig. 6 Influence of thickness of Au absorption layer on the responsivity of thermal conductivity measurements for Si and SiO₂ samples

Hence, this definition of responsivity can characterize the performance of the TTR method. By numerically solving the heat equation for Θ , it is possible to compute R_s and to bring out an important issue that could be used to assess the performance of the TTR technique.

The influence of the thickness of the gold absorption layer on the responsivity of the thermal conductivity of two gold-covered samples, one silicon and the other silicon dioxide, is shown in Fig. 6. In addition, the responsivity value ($R_s = 0.091$) for an uncovered bulk silicon sample is shown as a dashed horizontal line for reference purposes. For thermally-thick absorption layers, it is expected that the responsivity of the gold-covered silicon sample will be worse than the responsivity of an uncovered silicon sample because a very thick layer of gold will essentially hide the influence of the thermal properties of the underlying silicon material. In the transition and thermally-thin regimes, the responsivity of the gold-covered silicon sample is significantly higher than that of the uncovered silicon sample, except when the thickness of the gold cover becomes smaller than $h_{Au} = 2,150 \text{ \AA}$ ($Fo^{-1/2} = 0.207$). The improvement is as high as 40% at a the specific optimal thickness of gold $h_{Au} = 5,700 \text{ \AA}$, which corresponds to $Fo^{-1/2} = 0.558$. Based on this investigation, one can conclude that it is not advisable to use absorption layers that are in the thermally-thin regime (i.e., $Fo^{-1/2} < 0.207$) since these layers result in much lower responsivity values. Furthermore, in attempting to measure lower thicknesses of the absorption layer one can expect to incur higher levels of uncertainty.

By increasing the thickness of the gold layer on an SiO₂ sample, the responsivity exhibits a similar behavior and remarkably high R_s values. However, unlike silicon, SiO₂ is a transparent material and so the heating laser would not have been able to heat the SiO₂ test samples, if it were

not for the presence of the metallic layer. Therefore, it is impossible to estimate the accuracy gain resulting from the use of a gold cover for an SiO₂ sample.

4. Conclusions

The influence of the thickness of a metallic absorption layer on the performance of the transient thermo-reflectance method has been investigated. A previous investigation [9] had identified the thickness h^* for which a sample can be classified as a *semi-infinite layer* sample in the context of the TTR method. In the present work, it has been determined that h^* also defines the maximum possible thickness of gold that needs to be used as an absorption layer since using a thicker layer would hide the influence of the thermal properties of any underlying material. Conversely, the lower limit for the thickness of an absorption layer is the light penetration depth of the heating laser, d_L , since layers thinner than d_L do not absorb enough irradiation energy in order to generate the heat source required in the TTR method.

For thicknesses of the absorption layer between the lower and upper limits (i.e., $d_L < h < h^*$), the numerically and experimentally obtained transient surface temperature responses differ according to the ratio between the absorption layer thickness, h , and the heat penetration depth, d_H , during a laser pulse. The two drastically different behaviors are referred to as *thermally-thick* and *thermally-thin*. The *thermally-thick* layer behavior is characterized by a sharp change in the shape of the transient temperature at the time when the heat front from the laser pulse reaches the less thermally conductive oxide layer. Also, in the *thermally-thick* regime, a decrease of the absorption layer thickness enhances the normalized temperature response and shortens the time during which the initial rapid temperature decay takes place. In contrast, the normalized temperature response on top of a *thermally-thin* layer exhibits the opposite behavior. Specifically, a decrease of the layer thickness leads to a lower normalized temperature response, but while lower, it remains above the temperature response for bulk gold, even at thicknesses close to d_L . Between the *thermally-thick* and *thermally-thin* layers there is a range of layer thicknesses where their influence on the temperature response is minimal. The temperature response behavior associated with this range of thicknesses has been referred to as a *transition regime*.

The analysis performed in the present work has led to the association of the Fourier number (Fo) with the different regimes of the normalized temperature response behavior. The *thermally-thin*, *thermally-thick*, and *transition* regimes have been associated with $Fo^{-1/2} \lesssim 0.4$, $Fo^{-1/2} \gtrsim 2$, and $0.4 \lesssim Fo^{-1/2} \lesssim 2$, respectively.

The responsivity of the TTR measurements characterizes the performance of the TTR method, and makes it possible to recommend optimal thicknesses for a metallic absorption layer. The numerical simulations carried out for the gold-covered SiO₂ and Si samples revealed that the responsivity values in the *thermally-thick* regime are too low for the measurements to be sufficiently accurate. The same holds true for lower values of $Fo^{-1/2}$ in the *thermally-thin* regime. However, for the range $0.1 \lesssim Fo^{-1/2} \lesssim 2$ that covers all of the transition regime and most of the *thermally-thin* layer regime, the responsivity of the TTR measurements for metallized silicon oxide and silicon samples is sufficiently high for measuring the thermal conductivity of the underlying material with acceptable uncertainty. Furthermore, the responsivity values for a metallized silicon sample exceed the responsivity value for an uncovered silicon sample over a wide range of the $Fo^{-1/2}$ parameter. And, the maximum performance of the TTR method is expected for gold covered Si and SiO₂ samples at the specific thickness of gold $h_{Au} = 5,700 \text{ \AA}$, which corresponds to $Fo^{-1/2} = 0.558$.

5. References

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