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MINIMIZING THE UNCERTAINTIES ASSOCIATED WITH THE MEASUREMENT OF THERMAL PROPERTIES BY THE TRANSIENT THERMO-REFLECTANCE METHOD

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ABSTRACT

An approach for optimizing the TTR measurement of thermal properties is presented. The influence of the most important parameters of the system on the accuracy of the TTR measurements is investigated. An overall performance criterion is defined based on the responsivity of a given system and the thermoreflectance coefficient of the sample under test. It is shown that in order to obtain the smallest measurement uncertainty one should use a metallic absorption layer with the highest possible thermoreflectance coefficient and then compute the optimum thickness of that layer by maximizing the responsivity of the TTR system.

1. INTRODUCTION

As electronic devices become smaller, their thermal design becomes more critical, making the knowledge of thermal properties essential to the overall design process. Additionally, new device structures and materials with superior properties are being developed, but many of these may have unknown thermo-physical properties. Moreover, recent studies have revealed that the thermal properties of thin-film materials used in electronics are dependent on the deposition process.

The transient thermoreflectance method (TTR) [1, 2] is favored among other experimental techniques [3] used to determine the thermal conductivity of thin-film and multi-layered materials. The main advantage of the TTR method is that it is a non-contacting and non-destructive optical approach, both for heating a sample under test and for probing the variations of its surface temperature [4, 5]. However, TTR measurements of the thermal conductivity can be hindered by less-than-desirable optical properties of the top layer material (i.e., low thermoreflectance coefficient, low reflectivity, high transparency, surface roughness, and oxidation), which degrade the measurement performance of a given system. To eliminate these difficulties, investigators have resorted to the use of a metal “absorption” layer on top of the material under test (e.g., Au in [2], Al in [6,7] and Mo in [7]). Metal films are used because they exhibit high absorptivity and their optical properties are usually well known.

To date, relatively few investigators have used the thermo-reflectance (TTR) method to measure the thermal conductivity of thin films, and to the best knowledge of the authors, none have looked at optimizing the method. The most common practice to date has been to build a TTR setup using the existing technology (lasers) and to use it for measurements of as many types of materials as the setup could handle. The only conclusion drawn so far by the scientific community has been that lasers with ultra-short pulse-widths (pico to femto-second range) are more appropriate for measuring metals, whereas lasers with pulse-widths in the range of hundreds of picoseconds and few nanoseconds are more suitable for measuring semiconductor and dielectric materials.

Maximizing the performance of TTR measurements can be approached from two different, but equally important, perspectives. Firstly, the *TTR setup* itself can be optimized by adjusting the various characteristics of the components of the TTR system. The most important parameters are the pulse-width of the heating light source and the wavelengths of both the heating and probing light sources. Evidently, the characteristics of all TTR components have to be considered and chosen properly to achieve optimal measurements. For example, when building a TTR system one should consider acquiring a *fast* photodetector*,* a *high resolution* oscilloscope, optical components with *low losses* (light directing, filtering and transporting components), *stable* and *low noise* lasers, heating laser with sufficiently high fluence, etc.

Secondly, the *TTR measurements* can be optimized through proper adjustments of the structure and geometry of the sample under test. As mentioned earlier, it is desirable to use a metallic coating on every sample tested (both on transparent and opaque layers). However, an optimal thickness of coating should be used in order to attain the best performance of the TTR setup. Nevertheless, one should be aware that if the thickness of the absorption layer is not properly chosen, the performance of the TTR measurements of a coated sample could be lower than the measurement of an uncoated sample.

In this work, the influence of the most important parameters of the TTR setup on the performance of the TTR method is investigated and the results are presented in the following sections, after a brief description of the TTR method. An overall performance criterion is introduced based on the responsivity of the TTR system and the thermoreflectance coefficient, which are completely independent parameters. The thermoreflectance coefficient is solely dependent on the parameters of the probing laser and the cover material used, while the responsivity is dependent on the parameters of the heating laser and the geometry of the sample. Thus, the responsivity and thermoreflectance coefficient results shown here could be used either partially or entirely, such that they match the parameters of the TTR system under consideration. The values of the overall performance coefficient are computed in this work for the TTR system at SMU.

The results presented here are useful in two important circumstances. First, when one is designing a new TTR system, this work can be used to choose the best possible probing and heating lasers. This is especially helpful when measuring samples of similar nature (e.g., measuring the thermal conductivity of materials embedded in a device using gold contacts as an absorption layer). Second, for existing TTR systems, one can use the results of this investigation in combination with our previous investigation [8] to fully design the TTR measurements (i.e., sample under test). Specifically, the present study will help one decide on the metal that needs to be used as an absorption layer. The appropriate thickness of the absorption layer can be then calculated using the results presented here an in our previous study [8]. Certainly, there is also a third inherently instructive value to gaining a fuller understanding of the overall efficiency of the thermo-reflectance measurement technique.

2. TTR MEASUREMENT METHODOLOGY

The source of energy in the TTR method is normally provided by a pulsed laser with a short pulse duration. During each pulse, a given volume below the sample surface heats up to a temperature level above ambient due to the laser light energy absorbed into the sample. The heating area is a function of the laser aperture and the optics of the system. The depth of the volumetric heating, on the other hand, is determined by the optical penetration depth, which is a function of laser wavelength and surface material properties. The heating energy distribution through the light penetration depth (δ_L) obeys an exponential decay law. After each laser pulse, the sample begins to cool down to its initial temperature. During this process, the probing CW laser light reflected from the sample surface at the heating spot center is collected on some sort of photodetector that reads the instantaneous surface reflectivity. The changes in surface reflectivity are linearly proportional to the changes in surface temperature, within a wide but finite temperature range ([9], [10]). The specific system in the Laboratory at SMU (engr.smu.edu/setsl) uses an Nd:YAG pulsed heating laser whose wavelength is 532 nm, pulse width is 8.6 ns, and maximum pulse energy is 0.5 mJ. Two CW probing lasers are available for use: one Argon-Ion (488 nm), and one He-Ne (633 nm).

Given that the TTR system consists of the heating subsystem delivering power to initiate a temperature response, and the probing subsystem tracking a change in reflectivity of the sample corresponding to the temperature response, the authors have introduced the overall performance coefficient, $\eta = Rs_K \times C_{TR}$, which can be used to characterize the quality of a thermal conductivity measurement system. The first parameter in the product, Rs_K, represents the responsivity of the TTR thermal conductivity measurements, which characterizes the sensitivity of the temperature response to heat generation and transfer within a sample (first subsystem). The second parameter, C_{TR}, is the thermoreflectance coefficient of the sample surface, which characterizes the signal-to-noise ratio in the reflectivity measurements (second subsystem). A detailed analysis of these parameters is the subject of the current investigation.

3. RESPONSIVITY OF THE TTR MEASUREMENTS

In a previous work [11], the authors suggested the use of the *responsivity, Rs_K,* of the thermal conductivity measurement defined as $Rs_K = K\,(d\Theta/dK)_{\max}$, where Θ is the normalized temperature response of the sample surface and K is the thermal conductivity of the material. The responsivity Rs_K is calculated at the non-dimensional time T where $d\Theta/dK$ is maximized. Indeed, Rs_K is directly connected with the accuracy of the method by the equation $\sigma_K = {Rs_K}^{-1}\sigma_\Theta$, where σ_K is the random measurement uncertainty of the thermal conductivity, K, and σ_Θ is the random apparatus uncertainty related to detecting the temperature response. While σ_Θ depends on the apparatus signal-to-noise ratio and can be considered as a conservative value for a particular setup, the latter equation shows that the measurement uncertainty, σ_K, of the TTR technique decreases with increases in the responsivity value, Rs_K. Hence, the responsivity, Rs_K, which depends on the properties and geometry of the materials making up a sample, and the TTR system parameters, can characterize the performance of the TTR method and be useful for optimizing a given experiment. By solving the heat equation for θ, it is possible to compute Rs_K and to bring out important aspects used to assess the performance of the TTR technique.

In another publication [8], the authors used the above-defined responsivity to demonstrate that by covering the sample of interest with a thin layer of gold (i.e., an absorption layer), the performance of TTR measurements could be significantly improved as compared with measurements made on uncovered but otherwise identical underlying samples. As an example, for a silicon sample covered by 5,700 Å of gold, the resulting improvement was found to be over 40%. As expected, if the layer of gold is too thick, the responsivity of the gold-covered silicon sample will be worse than the responsivity of an uncovered silicon sample, simply because a very thick layer of gold will essentially hide the influence of the thermal properties of the underlying silicon material. On the other hand, measuring samples that have extremely thin layers of the absorption layer one can expect to incur higher levels of uncertainty. In the same investigation [8], it was shown that the TTR measurement results could be dramatically improved by a proper choice of a metallic absorption layer, even for opaque materials.

The present work is an investigation that was carried out to find the best material and its optimum thickness to be used as an absorption layer. Only metal films are investigated because they exhibit high absorptivity and their optical properties are usually well known. The metals considered for this study were (from least to most thermally conductive): titanium (Ti), palladium (Pd), platinum (Pt), nickel (Ni), chromium (Cr), cobalt (Co), zinc (Zn), molybdenum (Mo), iridium (Ir), magnesium (Mg), tungsten (W), beryllium (Be), aluminum (Al), gold (Au), copper (Cu), and silver (Ag). The layers to be measured, and considered here as substrates, are made out of silicon and silicon oxide. Additionally, structures composed of a silicon substrate covered with thin layers of silicon dioxide are also investigated.

The responsivity of a bulk silicon sample covered with various layers of metals is shown in Fig. 1(a). The results indicate that Ag, Au, and Cu layers with thicknesses between 4,000Å and 8,000Å yield the highest responsivity for the TTR setup. However, 4,000Å thick layers of Mg, 3,300Å of Zn, or 2,700Å of Cr will also produce good results if the thickness is kept within 10% of these values. The responsivity of the uncovered Si sample is shown as a horizontal dashed line (Rs = 0.091). The responsivity of measuring metal-covered silicon samples is higher than the responsivity of the uncovered sample if the metallic coating thickness is between 2,500Å and 5,000Å.

The presence of a metallic layer of any thickness yields a gain in accuracy, as compared to a corresponding uncovered sample. Specifically, metal films in the wide thickness range of 500Å to 1 μm can be deposited on these materials and the performance of the TTR setup will be better than testing the samples uncovered.

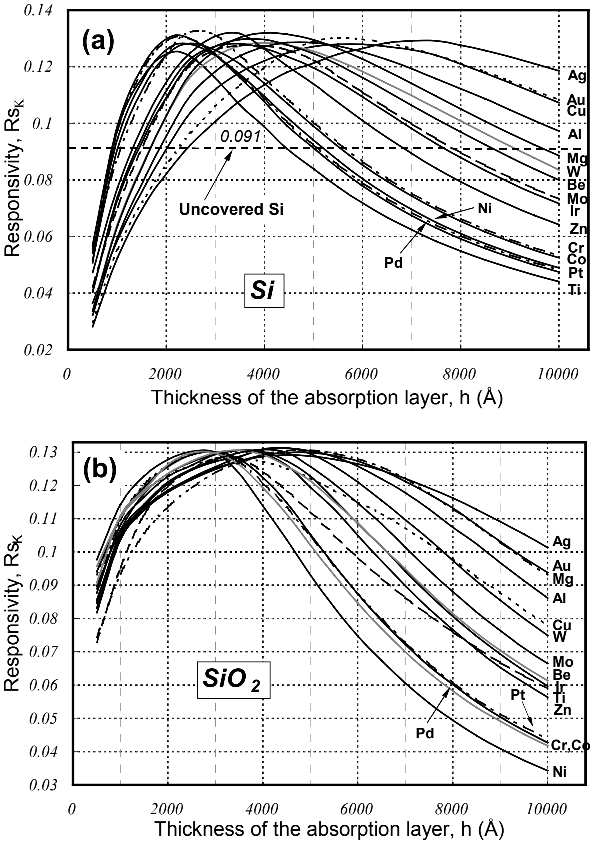

Fig. 1 Responsivity of TTR thermal conductivity measurements of bulk (a) Si and (b) SiO_2 covered with different metals

The responsivity of a metal-coated bulk silicon oxide sample is shown in Fig. 1(b). Again, the responsivity is maximum for particular thicknesses of coating metals. However, the maximum responsivity is now grouped in a much smaller thickness range of coating layers. Specifically, the maximum responsivity is observed for metallic absorption layers ranging from 2,800Å to 4,800Å. One should note that the responsivity of uncovered silicon dioxide is not provided, since TTR measurements for such (transparent) layers are not possible (using probing lasers with wavelengths in the visible spectrum).

Therefore, for all the materials presented above, one can find an optimal thickness of metal that can be deposited on top of the material, such that the responsivity will increase significantly. Nevertheless, there is not a single material, which if used as an absorption layer will produce a greater advantage in the responsivity, Rs_K, than other metals since the maximum value achievable is approximately the same for any metallic layer as long as an appropriate thickness is chosen.

The responsivity of measuring thin films of SiO_2 deposited on a Si substrate was also computed, and the results are shown in Fig. 2(a) for a 500Å film of $SiO_2$and in Fig. 2(b) for a 5,000Å film. The results corresponding to 10,000Å of $SiO_2$are identical to the results shown in Fig. 1(b) for bulk SiO_2. One may observe that the responsivity of measuring 500Å of SiO_2 (Fig. 2(a)) is twice as large as the responsivity of measuring bulk SiO_2. This corroborates the fact that the performance of the TTR method is much better for measuring thin SiO_2films on silicon substrate than for measuring bulk fused silica.

Although the present study was performed using the characteristics of the specific TTR system built at SMU, the results are applicable to other TTR systems as well. Furthermore, the present study may be used as a guide for investigating the possibility of using materials other than gold as an absorption layer.

4. THE THERMOREFLECTANCE COEFFICIENT

The sensitivity of the thermoreflectance technique is determined by the extent of the change in the reflectivity with changes in the temperature. The measure of this variation is called the thermoreflectance coefficient, C_{TR}, defined as the relative change in the reflectivity per unit change in temperature. For materials used in electronic devices, the values of the C_{TR} vary over several orders of magnitude above and below 10^{-4} K^{-1}. Also, the C_{TR} varies widely with the wavelength of the light source used for probing the change in the reflectivity of a sample.

Since the signal-to-noise ratio is directly proportional to C_{TR}, when looking into increasing the performance of the TTR measurements, one should choose the probing light source that will yield the maximum C_{TR}. As discussed later, finding the numerical values of C_{TR} for a certain material in a wide range of wavelengths is a difficult task since the data available in the literature for the C_{TR} coefficient for a certain wavelength of the probing laser irradiation is scarce or nonexistent. An alternate solution is to measure *in situ* the C_{TR} for a specific material at different wavelengths. However, this is difficult since it requires not only testing samples but also a system that needs to be developed specifically for this task.

For a given material, C_{TR} depends solely on the wavelength of the probing laser irradiation. Also, it has been observed (Figs. 1 and 2) that for most of the substrate materials investigated here, the maximum value of *Rs* is about the same for all of the investigated metals. Therefore, the probing laser (wavelength) can be selected independently by maximizing C_{TR}. The geometry of the sample can be subsequently designed such that the maximum value of responsivity is obtained.

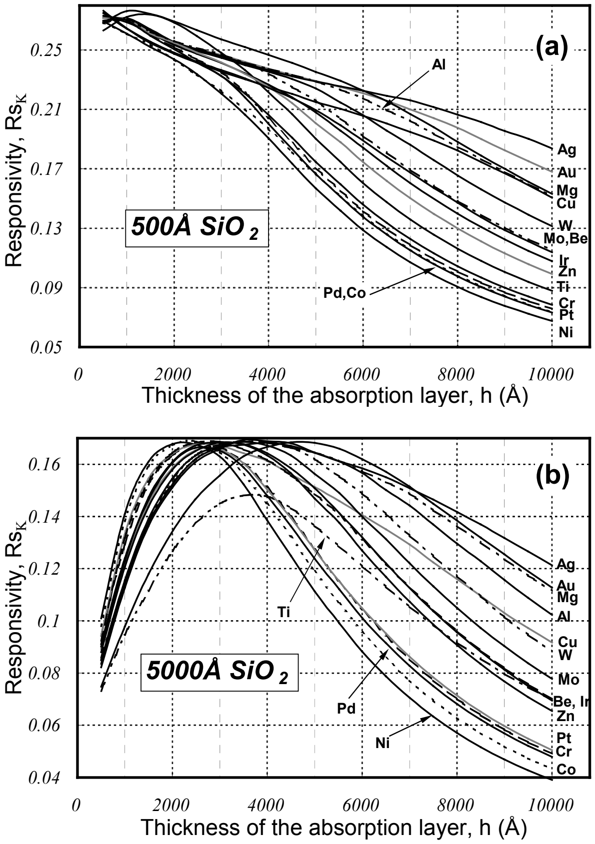

Fig. 2 Responsivity of TTR thermal conductivity measurements for (a) 500Å and (b) 5,000Å of SiO_2 covered with different metals

The values of C_{TR} are plotted in Figs. 3 and 4 for a select group of metals (Al, Ni, and Au) and wavelengths of the probing laser irradiation in the visible spectrum. The values were either taken from the open literature or measured by the authors in their laboratory at SMU. As depicted in figures, the change in C_{TR} with the wavelength does not exhibit a systematic behavior. Upon closer analysis of these data, one can draw two important conclusions about the qualitative behavior of the thermoreflectance coefficient. First, the C_{TR} coefficient is highly dependent on the wavelength of the probing irradiation, as expected. Perhaps less anticipated, however, is the fact that the magnitude of the variations is significant even for small changes in the wavelength. Second, practically for all metals, C_{TR} exhibits a change in sign for certain values of the wavelength. In other words, the reflectance of the sample could either increase or decrease when the temperature of the sample is increased, depend-

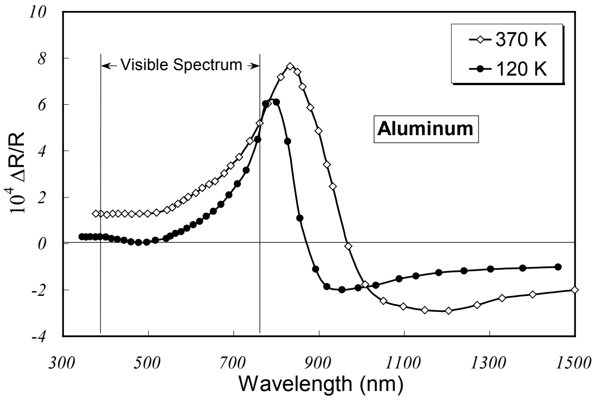

Fig. 3 Thermoreflectance spectra of aluminum [12]

ing on the wavelength of the light used for probing the change in the reflectivity of the sample.

Measuring C_{TR} in ideal conditions (good sample surface, vacuum, etc.) does not guarantee that the same value will be obtained for a "real" sample. Of special concern are the optical quality of the surface, the bulk and surface structure of the deposited layer, the surface and near surface wafer contamination, etc. The values of C_{TR} for Ni and Au are shown in Fig. 4 for a temperature of 120 K. The most prominent features of the C_{TR} curve for Au are the sine-like shape at around 2.5 eV (~500 nm). The width of this observed shape is around 0.1 eV and corresponds to a wavelength range of 470 to 520 nm. The qualitative behavior of the C_{TR} for Au is not expected to vary much at room temperature.

We measured the C_{TR} coefficient at room temperature (295 K) at a wavelength of 488 nm, and obtained a value of 2.9×10^{-4} K^{-1}, which is comparable to the values obtained by Scouler [14] at 120K (Fig. 4). The C_{TR} measurements were performed at SMU by the use of a measurement system similar to the one used by Abid et al. [15]. The system is composed of a light source (laser), a thermochuck, two identical photodiodes with corresponding multimeters, a CPU unit, and a couple of basic optical elements (lenses and mirrors). The samples are placed on the chuck and the reflectance of the sample is monitored and acquired by the CPU unit, while the temperature of the sample is incrementally increased using the precisely controlled (Temptronic) thermochuck. A two-diode differential approach is used to increase the signal-to-noise ratio. One diode (PD1) captures the light before it reaches the sample while the other diode (PD2) captures the light after it is reflected from the sample. The signal ratio PD2/PD1 is then plotted versus the temperature of the sample. The slope of the plotted curve represents the C_{TR}.

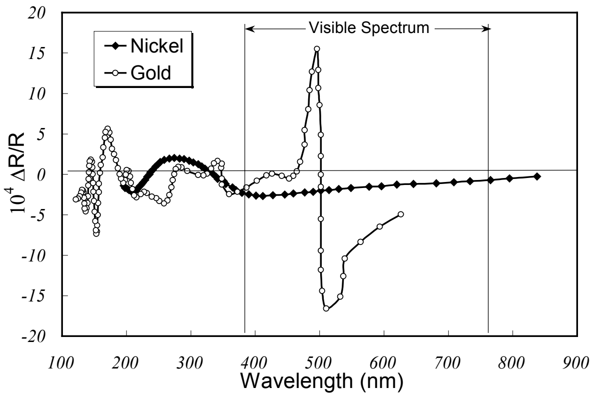

Fig. 4 Thermoreflectance spectra of Ni [13] and Au [14] at 120K

5. THE OVERALL PERFORMANCE COEFFICIENT OF THE TTR MEASUREMENTS

Since the thermoreflectance coefficient and the responsivity of the TTR measurements are the two parameters that can fully describe the performance of the TTR measurement, the following thickness optimizing parameter is proposed to characterize the performance of the TTR measurements, $\eta = Rs_K \times \left|C_{TR}\right|$. To minimize the uncertainty of the TTR measurements the above-defined coefficient must be maximized.

The value of the overall performance coefficient has been computed and is plotted in Fig. 5 for a bulk silicon sample covered with a gold, aluminum, or copper absorption layer at temperatures ranging from 120 K to 340 K. The coefficient η has been calculated using the characteristics of SMU's TTR system, namely for an Ar-Ion laser with a wavelength of 488 nm, and a YAG heating laser whose pulse width is 8.6 ns and wavelength is 532 nm. The computations were carried out using our measured data for the thermoreflectance coefficient of gold and the

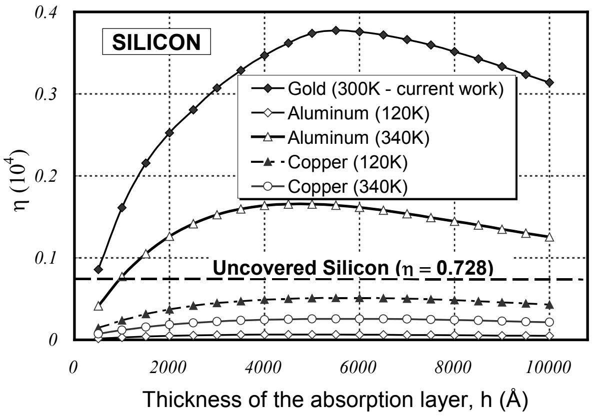

Fig. 5 Influence of a layer of Au, Al, and Cu on overall performance coefficient for a bulk silicon sample

data of Rosei and Lynch [12] for the C_{TR} of aluminum and copper. As shown in Fig. 5, a 5,500Å thick layer of gold will produce the most accurate TTR measurements. In contrast, for this wavelength of the probing laser, covering a bulk Si sample with a layer of copper will not improve the TTR measurements.

6. CONCLUSIONS

An approach for optimizing the measurement of thermal properties by the transient TTR method has been presented. The influence of the most important parameters of the TTR setup on the performance of the TTR method was investigated. The results show that the influence of the wavelength of the heating laser upon the performance of the TTR setup is not significant. However, a marked effect is observed when using heating laser with different pulse widths or probing laser with different wavelengths.

An overall performance criterion has been defined based on two independent parameters: the responsivity *Rs* of the TTR system, and the thermoreflectance coefficient C_{TR}. It was shown that the thermoreflectance coefficient is solely dependent on the parameters of the probing laser and the type of material used to cover the sample, while the responsivity is dependent on the parameters of the heating laser and the geometry of the sample. Thus, the *Rs* and C_{TR} results presented in this work can be combined to match the parameters of the TTR system under consideration and to compute the overall performance coefficient of a certain TTR setup. The values of the overall performance coefficient were computed in this work using the characteristics of the SMU TTR system.

Surprisingly, the results shown here indicate that in order to achieve the best responsivity of thermal conductivity measurement using a TTR system, any metal can be used to cover a semiconductor or amorphous dielectric sample, as long as the optimum thickness is deposited. This is an extremely important finding not only for scientific reasons but also for economical reasons. For example, one should use Cr instead of Au or Pt to cover a sample for TTR measurements (the price of a Cr sputtering target is 20 times less than that of Au or Pt). Therefore, in order to obtain the smallest uncertainty one should use an absorption layer that produces the highest C_{TR} and then compute the optimum thickness of that layer by maximizing the responsivity of the TTR system. Nonetheless, before a final recommendation can be made, future work needs to be performed to investigate the thermoreflectance coefficient of all existing metals at different wavelengths in the visible range.

11. REFERENCES

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