

Limitations and Advances in Optical Thermal Transport Measurements: Extremes in Properties, Length Scales, and Temperature

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Keywords

thermoreflectance, pyrometry, thermal transport, thermal measurements, high temperature, ultrahigh temperature, challenges, thermal conductivity, radiative transport, thermal boundary resistance

Abstract

Conductive and radiative thermal transport play a critical role in the design, development, and performance of a wide array of technologies and applications. In this review, we focus on the challenges associated with nano- and microscale thermal measurements and the strategies developed thus far to overcome them. For measurements below $\sim 1,000^\circ\text{C}$, numerous thermoreflectance techniques are already in wide use; however, uncertainty and measurement error may limit the measurement of samples in certain regimes. These regimes include materials of high thermal conductivity ($\gtrsim 2,000\text{ W/m}\cdot\text{K}$), thin films ($\lesssim 100\text{ nm}$), or interfaces located well below the sample surface. A rigorous treatment of uncertainty and

error is thus required for measuring these samples and for the development of future metrology tools. At higher temperatures, pyrometry techniques are being developed; however, several physical and experimental limitations exist. Some methods rely on a known emissivity for the measurement of temperature, and significant radiative transport can introduce error in modeling. Both of these mean that knowledge of spectrally dependent and temperature-dependent emissivity properties may be required.

1. INTRODUCTION

Heat dissipation via conductive and surface/interface transport dictates functionality and efficiency across a wide array of engineering applications. These range from the ultrasmall (nano) scales of interconnects and technology nodes in integrated circuits to the macroscales associated with buildings and airborne vehicles. The importance of heat dissipation for functionality and efficiency increases as technologies and applications push to their respective extreme limits.

For example, with technology nodes approaching the low-single-digit nanometer scale with hierarchical 3D geometries, thermal dissipation away from the joule-heat-driven hot spots becomes a primary concern. Thermal dissipation relies on not only the thermal conductivity (k) of the metal interconnects and interlayer dielectrics but also the thermal boundary resistance (TBR) (also termed Kapitza resistance, R_K) across the solid/solid interfaces. The high densities of interfaces (on the order of nm^{-1}) in electronic devices and associated thermal resistances can inhibit the dissipation of the extreme power densities, as is required to ensure continued operation. Similar, if not higher, power densities are achieved in wide-bandgap devices that form the basis of communications, radar, and charging stations. In these applications, thermal transport across films and interfaces with characteristic length scales from nano- to micrometers dictates the temperature of operation and ability to limit thermal runaway. Clearly, the ability to accurately measure the thermal resistance mechanisms (k and R_K) across these relevant nano-to-micro length scales is paramount to ensure continued improvements in performance in solid-state electronics.

While measurements of thermal properties of materials and interfaces at nano-to-micro length scales continue to be important to ameliorate extreme power densities in electronic devices, different challenges are found in extreme environments. For example, the heat fluxes and resulting temperatures of materials in extreme environment applications can also lead to thermal-, thermomechanical- and thermochemical-driven catastrophic failures at extreme temperatures. Engine coatings that experience temperatures upward of $2,000^\circ\text{C}$ are required to both thermally and chemically protect turbine blades from the high heat fluxes and particulates in the combustion environment. At these temperatures, the thermal conductivity of the coating may be only part of the problem in understanding the heat transport mechanisms at play, as the radiosity of the coating can also play a significant role. More specifically, exitance from hot walls of the combustion chamber or radiation flux from species in the hot gas environment can transmit through the coating and directly heat the turbine blade. Recent advances in turbine blade coatings have thus included designing for high-absorptance and high-emittance radiation barrier coatings to protect the turbine blade. This means the ability to measure the thermal conductivity of coatings in addition to their radiant exitance and emittance (M) and/or concomitant emissivities (ε) becomes a primary challenge in the thermal design of materials in extreme environments. The role of k and M in materials and coatings is even more critical when considering the extreme temperatures and heat fluxes experienced during hypersonic flight, when subjected to nuclear fuels, and when impinging on plasma-facing walls of fusion reactors. With these extreme heat fluxes leading

to material ablation and melting, the need to quantify k and ε (and related spectral quantities) at the relevant temperatures (i.e., up through their melting points) is becoming a major challenge.

Whether measuring k and R_k at extremely small length scales or k and ε at extremely high temperatures, various optical thermometry platforms have proven to be viable tools for measurements over a range of temperature and length scales. The flash diffusivity method (1) [frequently termed laser flash analysis (LFA)] was developed in the 1960s as one of the earliest measurement techniques involving noncontact heating (e.g., via lasers or flash lamps) and noncontact measurement (via pyrometry). This technique has been widely used to measure the thermal diffusivity of solids over a range of temperatures, including elevated temperature regimes pertinent to the extreme environments of jet engines, nuclear power, and hypersonics. Other techniques based on laser heating and temperature detection via infrared thermometry or thermography have been developed, forming the basis of modulated photothermal radiometry (2, 3). Typically, these techniques operate using modulated laser heating and lock-in detection to monitor the temperature magnitude and/or phase to extract the thermal diffusivity of a material. More recently, a steady-state approach was developed called thermo-optical plane source (4), which simplifies the measurement down to an application of Fourier's law to directly measure thermal conductivity. Despite the progress in such methods, infrared detection is fundamentally limited in resolution to approximately tens of micrometers at best. In order to overcome this limit, the exploitation of visible light is necessary. To this end, laser-based pump/probe thermorefectance techniques were developed starting in the 1980s, which gave rise to new directions for thermal conductivity measurements. These measurements are based on monitoring changes in reflectivity using a probe laser beam, allowing much better spatial resolution than a laser flash and offering new opportunities for measuring thermal properties that were previously not accessible. In 2004, Cahill (5) standardized a short-pulse-based thermorefectance method—time-domain thermorefectance (TDTR)—that gave rise to numerous related techniques such as frequency-domain thermorefectance (FDTR) and steady-state thermorefectance (SSTR). The thermal conductivity and spatial regimes are highlighted in **Figure 1** for the optical techniques reviewed in this article.

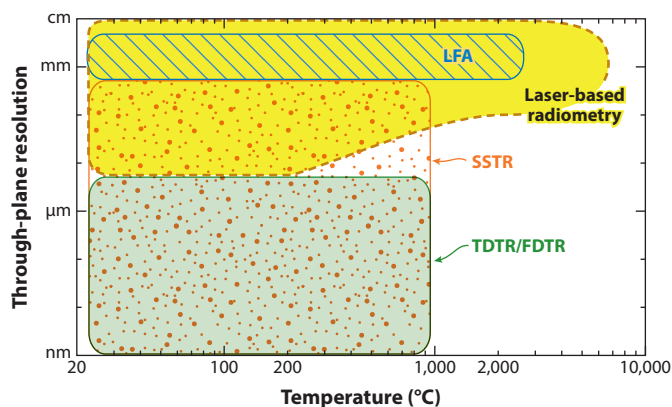


Figure 1

A regime map of noncontact thermal transport measurements, comparing the length scales measurable and the temperatures that can be reached. Thermorefectance techniques excel in the measurement of micro- and nanoscale properties such as the thermal resistances of interfaces but are limited to lower temperatures as a result of issues relating to the use of a transducer. Abbreviations: FDTR, frequency-domain thermorefectance; LFA, laser flash analysis; SSTR, steady-state thermorefectance; TDTR, time-domain thermorefectance.

In general, thermorefectance techniques are limited to temperatures less than $\sim 1,000^{\circ}\text{C}$ [limited by the transducer material selected (6)], while traditional LFA techniques can be employed up to $\sim 2,000^{\circ}\text{C}$ (limited by the furnaces used to heat the sample volume). Recent advances have taken advantage of laser-based heating combined with measurement of modulated or steady-state temperature perturbations, enabling measurements at ultrahigh temperatures (exceeding $2,000^{\circ}\text{C}$). One example of this is steady-state temperature differential radiometry (SSTD) (7), with which thermal conductivity measurements have been made on solid and molten tungsten, thus highlighting the ultrahigh-temperature potential for these thermometry platforms. A key to these high-temperature measurement techniques is the quantification of radiative losses from the sample (i.e., M and ε), which ultimately control the accuracy of these high-temperature techniques in determining k .

In this review, we outline and provide examples for several of the limitations faced by these various techniques and discuss methods developed to date aimed at overcoming them. In several areas, advances are still required in order to accurately measure thermal properties, including both common cases (e.g., thin films) and extremes (ultrahigh conductivity or measurements at ultrahigh temperatures). These advances might include careful consideration of transducer and substrate sample stacks, novel sample geometries to enhance sensitivity to the thermal properties of interest, combinations of experiments under multiple conditions, or even new metrology techniques.

2. REVIEW OF TRADITIONAL OPTICAL MEASUREMENTS

Below we outline several existing measurement techniques for determining micro- and nanoscale thermal properties optically. The advantages and disadvantages of each are discussed generally here, with specific examples provided in the following section. These techniques are traditionally performed at moderate temperatures ($\lesssim 1,000^{\circ}\text{C}$); however, alternate methodologies for measurement at extreme temperatures are provided in Section 4.

2.1. Laser Flash Analysis

The first technique (in chronological order of development) is the laser flash method (1). In LFA, a flash of light is used to heat one side of a thin sample, and pyrometry or a high-speed IR camera is used to track the time-dependent temperature rise and decay on either the same or opposite face of the sample. Thus, the time required for heat to dissipate through is captured, which is a measure of the net thermal diffusivity of the sample. Amorphous carbon coatings are typically applied to the sample in order to ensure a finite optical absorption depth and a surface-level, wavelength-independent emissivity reading. The heated area is typically uniform over the surface, meaning thermal transport is primarily 1D. Note that the diffusion time is related to the length scales involved and the thermal properties of the sample. This means fast detectors are required for millimeter- or submillimeter-scale measurements, and one should not expect to accurately resolve films with a thickness below $10\text{ }\mu\text{m}$. Additional details and discussion on LFA are also included in Section 4, as LFA is one of the few traditional techniques that has been shown to be easily extended to ultrahigh temperatures.

2.2. Thermorefectance

While LFA uses infrared detection to track the temperature changes on the surface of the sample, other experiments take advantage of the thermorefectance effect, wherein the reflectivity of a material changes slightly depending on temperature (8, 9). In thermorefectance experiments, one laser beam is used to heat the sample (pump), while a second monitors pump-induced changes in reflectivity (probe). Both the pump and probe beams are typically directed to the same face of the

Table 1 Overview of traditional thermoreflectance techniques

Measurement technique	Length scales (μm)	Advantages	Disadvantages
TDTR (5)	0.1–2 (sample dependent)	Pulsed laser enables measurement of transducer thickness via picosecond laser ultrasonics. It is able to measure multiple unknowns in a single experiment.	Mechanical delay stage adds complexity. Pulsed laser is costly compared with CW laser.
FDTR (12)	0.1–2	Pulsed FDTR uses same apparatus as TDTR and CW-FDTR uses same apparatus as SSSTR. CW lasers are inexpensive compared with pulsed lasers. Frequency sweep is automatically sensitive to a range of depths.	Correction of systematic phase in CW case can be difficult.
SSSTR (13)	1–10 (pump/probe spot-size dependent)	Measurements are extremely fast (enables mapping). CW apparatus is simple and relatively inexpensive. Experiment is insensitive to heat capacity.	Technique requires the use of a reference sample. Single measurement solves for single unknown (multiple measurements may be required).
ns-TTR (14)	0.1–2 (sample dependent)	Temporal data are acquired without the use of a mechanical delay stage.	Magnitude-based data acquisition requires normalization, which may result in higher uncertainty.

All techniques have a measurement uncertainty of 10% or higher, depending on the sample and experimental configuration(s). Thermal conductivities ranging from 0.1 to 2,000 W/m-K are measurable. Abbreviations: CW, continuous wave; FDTR, frequency-domain thermoreflectance; ns-TTR, nanosecond transient thermoreflectance; SSSTR, steady-state thermoreflectance; TDTR, time-domain thermoreflectance.

sample, and both are focused to diameters on the order of a single digit to tens of micrometers. In nearly all cases, a thin metal film (termed the transducer) is deposited on the surface of the sample, which isolates heating to the sample surface and provides an optically reflective surface for the probe. The changes in reflectivity for few-degree temperature changes are extremely small (8, 9) (1×10^{-4}), so the pump beam is modulated, and a lock-in amplifier is used to select the AC component of the probe signal at the modulated frequency. The most common thermoreflectance techniques are discussed below, and an overview of these techniques can be found in **Table 1**.

Early transient thermoreflectance (TTR) experiments (10) made use of a single pulsed laser for both the pump and probe, with a mechanical delay stage used to add a time delay between the arrival of pump and probe pulses at the sample (**Figure 2a**). By adjusting the position of the delay stage, the path length of one beam is changed, and the time delay between the arrival of pump and probe pulses is controlled. A thermal decay can thus be reconstructed and subsequently analyzed (**Figure 2c**). In 2004, Cahill (5) developed a rigorous analytical model for this experimental configuration, and the technique known as TDTR has been in wide use since. Note that the modulation of the pump beam is separate from the pulsing of the laser, meaning the true heating signal can be thought of as trains of pulses, all of which are captured by the aforementioned thermal model. In contrast to previous TTR experiments, the ratio of in-phase and out-of-phase components of the locked-in signal is typically analyzed in TDTR, eliminating the need to normalize the data. Note that the measurement depth for TDTR is typically on the order of hundreds of nanometers to a few micrometers [depending on the sample properties and modulation frequency used (11)], making it well-suited for the measurement of TBRs or thermal resistances of thin films.

A variant on TDTR uses a continuous-wave (CW) laser as the probe (14) (**Figure 2b**), which allows collection of the temporal signal over hundreds of nanoseconds (limited by the pulse repetition rate of the laser as opposed to the length of the delay stage). This has been termed nanosecond TDTR (15) (or erroneously just TDTR) or nanosecond TTR (ns-TTR) (16) (or erroneously TTR); however, it should not be confused with the aforementioned TTR

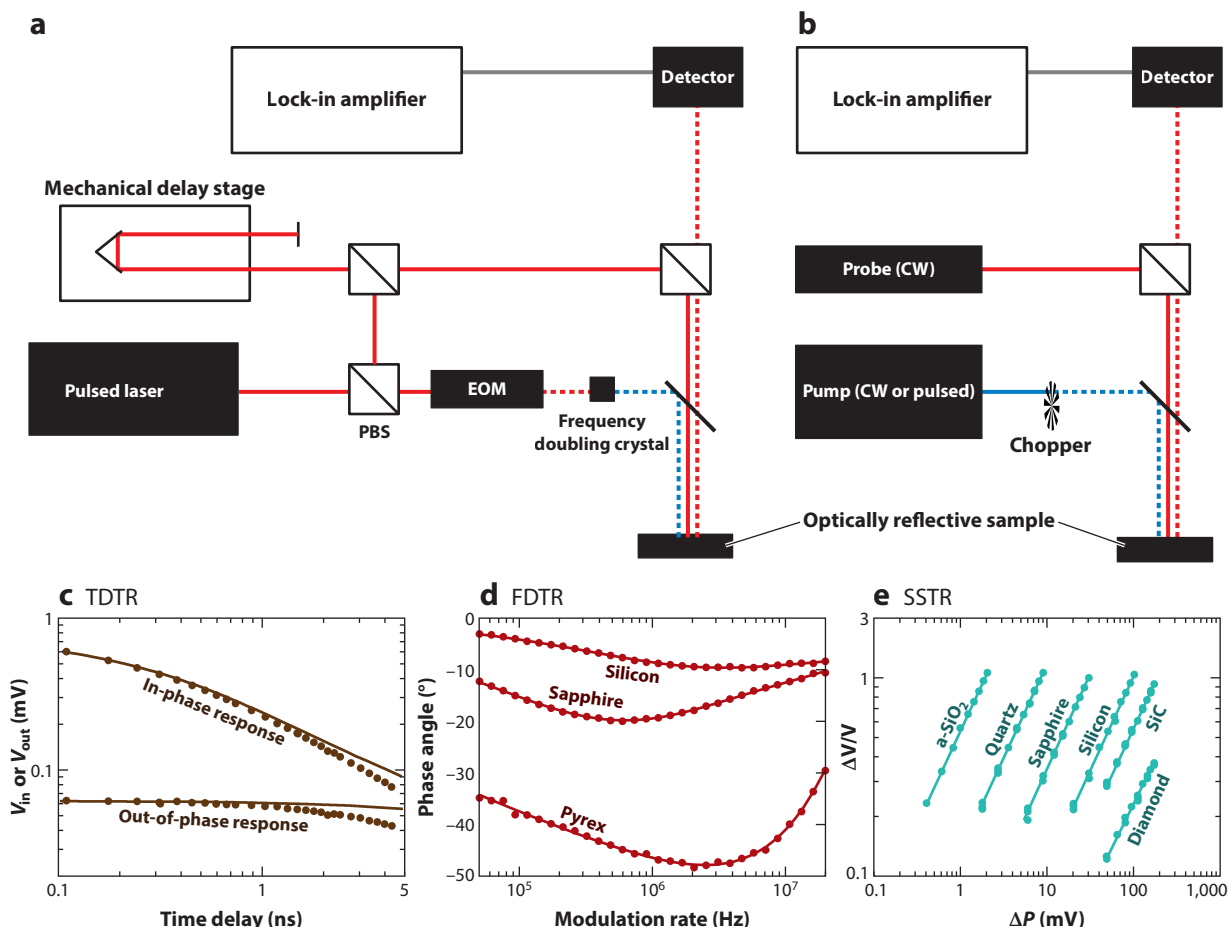


Figure 2

(a) A schematic for the pulsed-pump/pulsed-probe thermoreflectance experiments is shown, as used for TDTR, FDTR, and early TTR experiments. (b) A CW probe can be used instead, eliminating the need for a mechanical delay stage. A pulsed pump is used for nanosecond TTR, and a CW pump is used for FDTR and SSTR. (c–e) Data from founding works for (c) TDTR (5), (d) FDTR (12), and (e) SSTR (13). Panel c adapted with permission from Reference 5, rights managed by AIP Publishing. Panel d adapted with permission from Reference 12, rights managed by AIP Publishing. Panel e adapted from Reference 13 (CC BY 4.0). Abbreviations: CW, continuous wave; EOM, electro-optic modulator; FDTR, frequency-domain thermoreflectance; PBS, polarizing beam splitter; SSTR, steady-state thermoreflectance; TDTR, time-domain thermoreflectance; TTR, transient thermoreflectance.

or TDTR measurements. As shown below, the sensitivity of these pulsed-pump/CW-probe experiments (ns-TTR) differs substantially from that of the original pulsed-pump/pulsed-probe experiments (TTR and TDTR), as has been alluded to by the pioneering authors of ns-TTR (15). Since the pulsed pump is no longer modulated (a periodic waveform analyzer instead samples over the duration of time between laser pulses), only the magnitude of the temporal response is collected, which means normalization of the data is necessary. The response rate of the photodetector is also a limiting factor, and the temporal resolution will not match that of the pulsed-pump/pulsed-probe TDTR or TTR experiments.

Thermoreflectance data can also be collected in the frequency domain, using either the same pulsed-pump/pulsed-probe apparatus as TDTR (Figure 2a) or a CW-pump/CW-probe system

(**Figure 2b**). This is termed FDTR (12), and the angle of the in-phase versus out-of-phase signal (i.e., the phase lag between the cyclic heating and cyclic thermal response) is collected as a function of the modulation frequency (**Figure 2d**). The CW version of FDTR has the advantage of being a simpler and cheaper apparatus (eliminating the need for the pulsed laser or mechanical delay stage). Care must be taken, however, to eliminate effects of systematic errors [e.g., an additional frequency-dependent phase introduced to the signal by the electronics or optics involved (17)]. In the thermoreflectance techniques discussed thus far, the use of surface heating yields a thermal decay through the depth of the sample, and surface probing means there is diminishing sensitivity to parameters through the depth as well. This is often discussed in terms of the thermal penetration depth (TPD), which is estimated as $d = \sqrt{k \times \pi^{-1} \times C^{-1} \times f_{\text{mod}}^{-1}}$ (11) based on the thermal conductivity k , heat capacity C , and modulation rate f_{mod} . Given that FDTR sweeps through modulation frequencies (and that the measurement depth depends in part on the modulation frequency), FDTR is naturally sensitive to varying depths of the sample at different frequencies, making it well-suited for the measurement of complex sample geometries. Generally speaking, a measurement over a wider frequency range results in sensitivity to a greater number of parameters (18–20). FDTR has also been demonstrated to detect subsurface interfaces and defects at depths much larger than the estimated TPD (21–25). As the TPD is not an abrupt cutoff, even with a reduced level of sensitivity to thermal resistance changes at depths larger than the TPD, detection of structural changes and interfaces deep within the sample can be achieved. The key to these measurements is a larger contrast in thermal properties (e.g., interface delamination with extreme changes in TBR). As these realizations in FDTR are relatively recent and, at present, do not encompass the most commonly applied configurations and scenarios in typical FDTR measurements, we do not include these depth resolution limits in our regime map in **Figure 1**.

The magnitude of the temperature rise can also be considered, as in the case of SSTR (13). SSTR operates in the low-frequency regime (<1 kHz), where the measurement is naturally unaffected by the heat capacity of the sample, and the temperature rise is roughly proportional to the net thermal resistivity within the measured volume. Note that the previous expression for TPD is no longer applicable (26). Instead, steady-state heating yields an approximately hemispherical heated volume, with a measurement depth on the order of the pump and probe beam radii. In practice, this means SSTR measurements typically have a higher TPD compared with TDTR or FTDR, increasing sensitivity to interfaces and layers well below the surface (termed buried interfaces or layers) (13, 26, 27). The experimental configuration for SSTR is the same as that of CW-FDTR (**Figure 2b**); however, the CW pump is modulated at a single frequency, while the magnitude of the response from the probe is recorded at various pump powers (**Figure 2e**). In other techniques where the magnitude of the signal is considered, the signal is normalized prior to analysis, since numerous sample-dependent and system-dependent properties will contribute to the scaling of the signal (e.g., the thermoreflectance coefficient, laser transmission through optical components, or electronic effects). In SSTR, however, the normalization factor (γ) is measured using a known reference sample with an identical transducer (e.g., an Al/a-SiO₂ material system, where the transducer and substrate have been well-characterized separately). Since the slope of the signal magnitude versus pump power dataset is the only meaningful feature, only a single unknown can be determined from a single measurement. Multiple measurements (e.g., using multiple spot sizes or modulation frequencies) may allow fitting of additional parameters, however. In cases where heat capacity is not known, the insensitivity of SSTR to heat capacity is an advantage over other techniques that measure thermal effusivity or diffusivity (LFA, TDTR, and FDTR). Similarly, the simplicity of the experiment (CW beams, no mechanical delay stage, no need to account for the systematic phase) makes SSTR measurements comparably fast, even allowing for high-resolution thermal property mapping across samples with inhomogeneity (28).

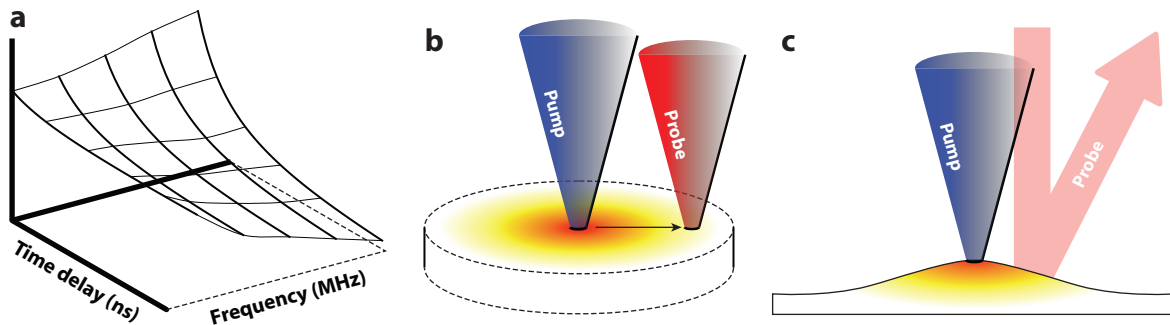


Figure 3

Several methods have been developed to improve experimental sensitivity, either reducing error or enabling fitting of additional parameters. (a) Given that time-domain thermoreflectance (TDTR) and frequency-domain thermoreflectance (FDTR) experiments are fundamentally similar (measuring the temporal and frequency response of the system, respectively), a surface-fitting approach combining the two has been developed. This involves collecting TDTR at multiple frequencies (or FDTR at multiple time delays) and fitting datasets simultaneously. (b) By spatially offsetting the pump/probe beams, the lateral spreading of heat is sensed, enhancing sensitivity to in-plane transport. (c) A beam deflection technique has recently been developed, taking advantage of the thermally induced mechanical strain of the sample.

2.3. Variants and Combined Techniques

Numerous variations of these techniques have been developed to address specific problems or to better measure specific properties. By intentionally offsetting the pump/probe spots on the sample (29, 30), increased sensitivity to in-plane thermal conductivity can be obtained (**Figure 3b**). Others have achieved similar results by using intentionally elliptical beam shapes (31, 32). Similarly, a diffraction grating can be used (as in a technique termed transient grating spectroscopy) to set up a pattern of sinusoidal heating on the sample surface. Relaxation of this temperature profile can then be related to the thermal transport phenomena within the sample (33–35).

Multiple measurements can also be combined to achieve greater sensitivity or fit for additional parameters. For example, TDTR (36, 37) and SSTR (13) data have been collected using multiple spot sizes, enabling the fitting of additional parameters. Similarly, numerous authors have combined TDTR and FDTR. By performing TDTR at multiple modulation frequencies (or FDTR at multiple time delays), additional unknown parameters can be extracted (37–46). This has also been referred to as surface fitting, where the thermoreflectance data are collected across time and frequency domains, as depicted in **Figure 3a**. Similar strategies involve simultaneous fitting of independent TDTR and FDTR datasets (47), analyzing both ratio and magnitude data from TDTR (43, 48, 49), or combined fitting of other datasets (e.g., TDTR and SSTR) (50, 51).

Additional means of collecting and analyzing thermoreflectance data have also been considered. Recently, a technique titled square pulse thermoreflectance (52) was introduced, wherein a square modulation is applied to the CW pump laser, and the waveform of the probe response is collected with a periodic waveform analyzer. This is similar in principle to ns-TTR, which analyzes the post-pulse temporal decay. In both cases, normalization of the data is required, which may result in increased uncertainty and may limit the simultaneous extraction of multiple thermal parameters. The time ranges of data analyzed may also have a significant effect on the sensitivity of the experiment to a given thermal parameter. For example, a sample with high thermal conductivity may reach the steady state within microseconds (depending on the spot sizes used), meaning collection of the entire periodic waveform at lower modulation rates may not be of any benefit. For this reason, experimental conditions should be selected carefully based on the samples under consideration. Similarly, mechanical chopping of a beam will yield a rise or fall time that is dependent on the diameter of the beam passing through the chopper, which may effectively wash out the

meaningful time regimes within the signal. Electronic modulation (e.g., with an electro-optic modulator) offers an alternative with fast response times but is generally limited by allowable incident power, which in turn may limit the ability to measure high thermal conductivity substrates.

Beyond thermoreflectance and pyrometry, other physical phenomena have been used for the optical detection of pump-induced temperature changes. While the reflectivity of a sample changes with temperature, the polarization of the reflected light may also change via the magneto-optic Kerr effect. This effect occurs only with magnetic materials but has enabled the use of ultrathin transducers to achieve better sensitivity to certain thermal properties (53, 54). Beginning in the 1980s, multiple authors have used the changing refractive index of the air adjacent to a solid surface to deflect the path of a probe laser (55, 56). A rigorous mathematical treatment of this experiment enables the use of this phenomenon to measure thermal diffusivity (57). Beam deflection due to thermally induced mechanical strain within the sample itself (**Figure 3c**) has also been used as a thermometer (38) and more recently to extract thermal properties (58, 59). While the pump-induced beam deflection is only tens of microradians, extending the path length can amplify the physical displacement of the beam on the detector, meaning signals of higher magnitude can be obtained as compared with thermoreflectance. Similarly, propagation of error from beam spot sizes is also reduced (due to the use of offset beams and the underlying thermomechanical physics at play). The damping of an acoustic wave echoing off an interface has also been used as an indicator of TBR (60–63). This is measured via picosecond laser ultrasonics (PLU), in which the echo of an acoustic wave generated by a pump pulse is detected by the probe. Confounding effects prevent PLU from being used quantitatively, however (64). Finally, Raman spectroscopy has been used to measure thermal properties, with both peak shifts and amplitudes being dependent on temperature. This eliminates the need for the metal film transducer, meaning the sample material of interest is directly measured, mitigating some of the transducer-related issues (65).

3. CHALLENGES: UNCERTAINTY AND MEASUREMENT ERROR

The reliability of a measurement comes down to the assumptions required by the analysis (measurement uncertainty), the capabilities and calibration of the experiment itself (systematic error), and randomness (random error). Within the field of nanoscale thermal measurements, these are occasionally considered but almost never rigorously incorporated in full.

3.1. Propagation of Uncertainty

To understand measurement uncertainty, it is necessary to understand the interdependence of multiple thermal parameters. For example, consider a measurement of the net thermal resistivity (such as the slope of an SSTR dataset) or total thermal diffusivity (as is the case in an LFA measurement). The net resistivity measurement will be unable to differentiate between a multilayer sample with high thermal conductivity and high TBR and one with low thermal conductivity and low TBR, and, similarly, the thermal diffusivity measurement can not differentiate between a sample with high k and high C or low k and low C . If one of the interdependent parameters is being assumed in the analysis, then the assumption used for this value will affect the fitted values for the other parameter (termed measurement uncertainty).

This phenomenon is common when films are involved, as changing the thickness of a layer in the sample stack will almost proportionally affect the final fitted conductivity. Similarly, the resistance associated with an interface is often directly interchangeable with the resistance of a film. It is useful to consider the Kapitza lengths associated with thermal interfaces ($l_K = R_K \times k$), which describe the thickness for which a film would have an equivalent thermal resistance to an interface. For example, a TBR of $5 \text{ m}^2 \cdot \text{K}/\text{GW}$ is equivalent to $0.2 \text{ } \mu\text{m}$ of sapphire ($35 \text{ W}/\text{m} \cdot \text{K}$) or 7 nm of an amorphous oxide (a-SiO_2 , $1.4 \text{ W}/\text{m} \cdot \text{K}$), meaning even slight uncertainty in the sample

thickness and film conductivities can yield a larger uncertainty associated with the measurement of the TBR and vice versa. This has been a noted challenge for LFA when measuring thin films (66) and is an issue for thin high-conductivity films for thermoreflectance experiments as well.

There are two ways this effect is typically quantified, and both begin with reasonable assumptions (or measurements) of uncertainty for all input parameters (e.g., layer thicknesses, heat capacities, assumed thermal conductivities, laser spot sizes). In Monte Carlo analysis, combinations of input parameters are generated on the basis of a normal distribution of each parameter. The fitting is then repeated for each combination, and the standard deviation of the fits is taken as the uncertainty. A simpler (and more computationally efficient) approach is to simply perturb each input parameter individually and refit. While this does not capture the combinations across the normal distributions of each parameter, it tends to yield similar results and is typically sufficient. Alternatively, the sensitivity of the fitted parameters to each input parameter can be calculated and incorporated analytically (67). Note that these various methods are all fundamentally probing the same phenomenon, which is the propagation of uncertainty from input parameters (examples and a mathematical comparison are available in the **Supplemental Appendix**).

When analyzing uncertainty in either manner, it is important to consider independent versus dependent parameters. In SSSTR, the γ term is found by fitting from a known reference sample (known thermal properties, identical transducer), meaning the uncertainty in γ is dependent on the uncertainty in other parameters. It is also common to use four-point probe electrical resistivity measurements to infer the transducer thermal conductivity via the Wiedemann–Franz equation, meaning uncertainty in the transducer thermal conductivity may be directly dependent on the uncertainty in the transducer thickness. In some situations, treating these as independent parameters may overestimate the uncertainty (examples are available in the **Supplemental Appendix**).

3.2. Relationship Between Multiple Fitted Parameters

When multiple parameters are fitted simultaneously, the effects of their interdependence must also be considered (termed multicollinearity in the field of regression analysis, this is a form of systematic error). In the most basic example, we consider a single SSSTR dataset, where the slope is the only meaningful piece of information. For two fitting parameters, infinitely many combinations of values will achieve an identical fit to the data. Interestingly, the use of machine learning algorithms in place of traditional least-squares curve fitting does not alleviate this problem, as the measurement itself fundamentally might not be able to discern between different resistances. This phenomenon exists in all thermoreflectance experiments; however, the interdependence of the fitting parameters (and thus the ability of the experiment to measure multiple parameters) is often not obvious. For this reason, we recommend that contour analysis be used to explore this effect.

In contour analysis, many permutations of the fitted parameters are tested against the data (model curve generated, residual between data calculated). For two fitting parameters, the residual as a function of both parameters can be visualized as a contour plot, with pairs of values providing a similar quality of fit, represented as a contour. All combinations of parameters that yield a satisfactory fit (residual below a chosen threshold) are candidate solutions to the dataset, meaning the extent of the given contour indicates the measurement error. For three or more fitting parameters, probing the entire 3D+ parameter space may be computationally inefficient. Instead, a 2D contour plot can be generated by sweeping through pairs of values for two parameters (or in one dimension, sweeping through a range of values for one parameter), fitting for the remaining additional parameter(s) at each location and recording the residual of the best fit. An example contour plot is shown in **Figure 4a**, with the inset showing the 3D parameter space and the outset showing the collapsed 2D contour. Selection of the appropriate value for the contour threshold is discussed in the following section.

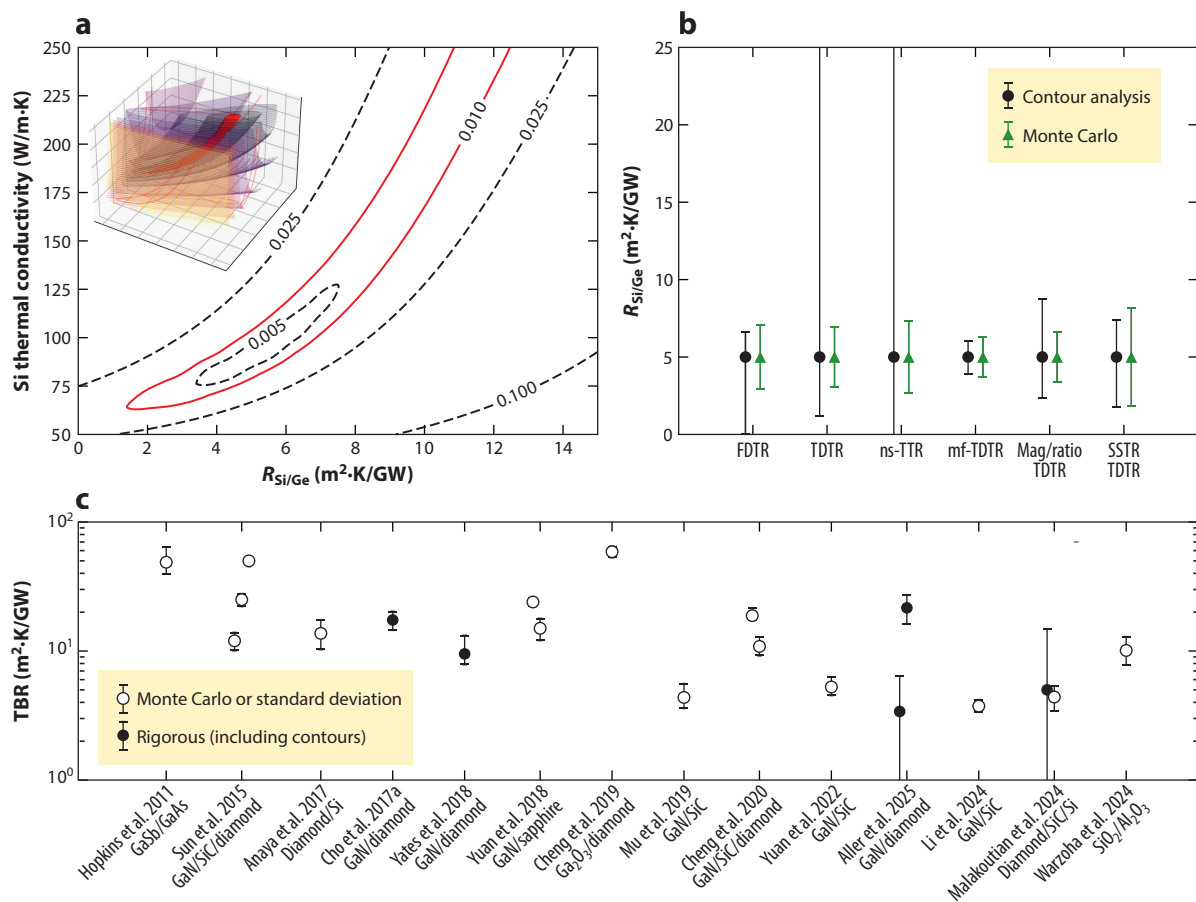


Figure 4

(a) Considering the hypothetical measurement of the silicon/germanium interface ($R_{Si/Ge}$) within an aluminum/silicon/germanium stack, three unknown parameters (thermal conductivity k_{Si} and TBRs $R_{Al/Si}$ and $R_{Si/Ge}$) mean contour analysis must be performed over a 3D parameter space. This can be visualized in three dimensions (*inset*) or collapsed along a given axis (*outset*). For a given quality-of-fit threshold, the bounds on the corresponding contour region denote the combinations of parameters yielding an acceptable fit between the model and data. This is a measure of the error due to multicollinearity and systematic issues or misalignment. (b) We performed contour and Monte Carlo analysis on synthetic data for multiple measurement types and variants. In almost all cases, Monte Carlo analysis underestimates the total error, and in some cases the $R_{Si/Ge}$ contour uncertainty is unbounded (i.e., an arbitrary value will yield a good fit if k_{Si} and $R_{Al/Si}$ are adjusted accordingly). (c) We present data from the literature for buried interfaces for various material systems (43, 44, 50, 68–78). Open circles denote uncertainty calculated via Monte Carlo analysis or the standard deviation across measurements, while closed circles denote measurements with more rigorous uncertainty, capturing multicollinearity (e.g., contour analysis). Abbreviations: FDTR, frequency-domain thermoreflectance; Mag, magnitude; mf-TDTR, multifrequency TDTR; ns-TTR, nanosecond transient thermoreflectance; SSTR, steady-state thermoreflectance; TBR, thermal boundary resistance; TDTR, time-domain thermoreflectance.

3.3. Apparatus Misalignment

It is also important to recognize the role of systematic errors originating from the measurement apparatus, as poor system alignment will yield datasets with incorrect trends, producing inaccurate fits. For example, misalignment of the pump/probe spots by a fraction of a micrometer can easily change the slope of an SSTR measurement by more than 1% (calculation shown in the **Supplemental Appendix**). Similarly, a misalignment down a TDTR delay stage could cause a

Supplemental Material >

shifting of pump or probe beams or a change in focused radii (5), yielding a decay curve with an incorrect slope. This misalignment-induced offset applied to the data will then yield incorrect results when fitting is performed. Note that the sensitivity to the fitted parameter(s) loosely determines the influence of these systematic errors on the fitted result. For example, a parameter that minimally affects the final curve may need to be adjusted quite far in order to correct the systematic deviation in the data originating from experimental apparatus misalignment. This would mean the fitted value could be quite far from the material property's true value. Conversely, for the hypothetical perfect measurement system (which does not exist), there would be no reason to accept anything but a perfect fit, despite potentially low sensitivity to a given parameter.

One's confidence in the alignment of their measurement system thus has a bearing on their confidence in the result, and the rigor with which the system was calibrated determines the confidence in the alignment. We can thus establish guidelines on the selection of a contour threshold on the basis of considerations of the calibration procedure used. It is common practice to validate a measurement system by measuring a (series of) known calibration sample(s) and confirming that the correct value(s) is found. This means we can use the range of acceptable calibration values and corresponding changes in the model's curve to guide selection of the contour threshold. For an SSTTR measurement on an Al/a-SiO₂ system, changing the thermal conductivity of the SiO₂ by 4% will yield a 1% residual. This means if one accepts a fitted SiO₂ conductivity of 1.35–1.45 W/m·K during calibrations (from nominally 1.4 W/m·K), then a 1% residual may suffice. In contrast, for a TDTR measurement on Al/sapphire, a 4% change in the sapphire conductivity yields a 1.5% residual. Note that the level of noise in a measurement (or the lack thereof) may have little to do with the alignment and calibration of a system, so a low threshold should not be used based solely on the level of noise in the experiment.

Since contour analysis is intended as a measurement of the interdependence of fitted parameters and the capabilities of a given measurement system, a minor modification is the use of synthetic data in the analysis (as opposed to using real data resulting from a measurement). A noisy dataset (with a higher residual despite being an otherwise accurate fit) should not serve to reduce the claimed error associated with a measurement. Instead, if synthetic data with zero noise are used for contour analysis, then a more realistic determination of the capabilities of a given measurement will be found (system noise aside).

The relationship between the size of a given parameter's contours and the parameter's sensitivity also minimizes the need for sensitivity analysis. If a measurement is insensitive to a parameter, a large range of values of that parameter may yield a satisfactory fit to the data, and the contours will be large. Sensitivity analysis is typically performed by perturbing a parameter in the model and comparing the resultant curve to the unperturbed model; however, it may be difficult to infer from this whether additional parameters can be fitted. Conversely, generating contour plots for a hypothetical scenario means the experimental error can be predicted directly. Furthermore, the benefit (or lack thereof) of alternative analysis schemes (e.g., multifrequency TDTR or ratio and magnitude fitting of TDTR data) becomes apparent. By overlapping multiple contours (e.g., contours for TDTR at multiple frequencies), it should become immediately clear whether the additional measurements help in reducing the contour bounds.

3.4. Comparing Propagated Uncertainty to Systematic Error

It is critical to note that contour analysis is testing a fundamentally different source of error compared with Monte Carlo analysis. Monte Carlo captures uncertainty in the end result originating from uncertainty associated with the input values. This is in contrast to contour analysis, which considers the interdependence of the fitted parameters with regard to one another (multicollinearity) and the ability or inability of the measurement to discern multiple parameters simultaneously.

To demonstrate this concept, we evaluate the hypothetical measurement of a buried interface (Al/Si/Ge; tabulated values are available in the **Supplemental Appendix**), as semiconductor–semiconductor TBRs have great relevance in semiconductor device design and manufacturing. Due to surface roughness and cleanliness effects as well as variations in transducer deposition, the transducer/film TBR ($R_{\text{Al/Si}}$) should not be assumed. Similarly, size and crystallinity effects may yield unexpected film thermal conductivities (k_{Si}), so this parameter should also be fitted. Given the final film/substrate TBR ($R_{\text{Si/Ge}}$) as our parameter of interest, we are thus left with three fitting parameters ($R_{\text{Al/Si}}$, k_{Si} , and $R_{\text{Si/Ge}}$).

Three unknown parameters mean contour analysis must be performed over a 3D parameter space. This can be visualized in three dimensions (inset in **Figure 4a**) or collapsed along a given axis (outset in **Figure 4a**). The bounds on the contour region for a chosen threshold denote the combinations of parameters that yield the corresponding quality of fit, which yield the error due to multicollinearity and systematic issues or misalignment. Practically, as we are interested only in $R_{\text{Si/Ge}}$, we sweep through values of $R_{\text{Si/Ge}}$ and fit for $R_{\text{Al/Si}}$ and k_{Si} at each point to establish bounds for $R_{\text{Si/Ge}}$ for subsequent analysis (**Figure 4b**). Note that an additional example comparing Monte Carlo and contour analysis is available in the **Supplemental Appendix**. The code used in the analysis is also freely available at https://github.com/ExSiTE-Lab/TDTR_fitting.

We also performed contour and Monte Carlo analysis on synthetic data for multiple measurement types and variants (**Figure 4b**). In all cases, Monte Carlo analysis underestimates the total error. In some cases, the contour analysis suggests $R_{\text{Si/Ge}}$ cannot be measured, as an arbitrary value for this parameter will yield a good fit if appropriate corresponding values for k_{Si} and $R_{\text{Al/Si}}$ are used. Interestingly, each experiment also has differing contributions from contour analysis and Monte Carlo. This means that one's ability to characterize all input parameters (contributing to Monte Carlo) or the level to which they calibrate their apparatus (contributing to contours) can be used to guide selection of the experiment.

We also present data from the literature (**Figure 4c**) for buried interfaces for various material systems (43, 44, 50, 68–77). In many cases, the uncertainty reported is likely artificially low due to incomplete uncertainty quantification. In other cases, the authors took great care to capture error from various sources and found they needed to develop new measurement techniques or modify existing ones in order to adequately characterize the interface. This includes measurement at multiple modulation frequencies or with multiple spot sizes (43, 44, 68, 70) or fitting of multiple data types (50), development of transducerless measurements (71, 75), and the use of unique sample geometries (44) or experimental configurations (77). In some cases, high TBR values are simply easier to measure (as compared with the aforementioned Al/Si/Ge example), as the experiments tend to be most sensitive to the highest resistance within the system.

3.5. Influence of Sample Variation and Noise

Another common method of quantifying uncertainty is to simply take the standard deviation across multiple measurements. It is important to recognize that this approach captures neither of the above sources of uncertainty or error and instead simply captures the random variation across the sample, multiple samples, or the noise within a given experimental apparatus. This method of calculating uncertainty is important because it is an indication of the repeatability of the experiment, as some samples may have a high degree of variability across them, and some growth or sample preparation techniques may produce highly variable samples. That said, neither propagated uncertainty (analyzed via Monte Carlo) nor the relationship between multiple fitted parameters (multicollinearity) or apparatus systematic error (as seen via contour analysis) should be neglected. Each method should be used (propagated uncertainty, contour analysis, and random

variation), with the total error found via the root mean sum (ensuring that a low error from one source does not serve to reduce the overall error).

3.6. Examples

We now briefly consider two additional examples to highlight the regimes where challenges persist. In the case above, we considered the measurement of the TBR for a buried interface, highlighting the importance (and nonequivalency) of the various sources of error (measurement uncertainty, explored via Monte Carlo analysis, and systematic error, explored via contour analysis). For the hypothetical example of Al/Si/Ge, we showed that none of the standard measurement techniques were able to establish both upper and lower bounds on $R_{\text{Si/Ge}}$, and only the hybrid approaches worked (hybrid FDTR plus TDTR and combined fitting of TDTR and SSSTR data). Note that these findings are by no means generalized; a specific sample's properties (e.g., substrate conductivity or film thickness) may yield different results, and this analysis should be performed for any new sample under consideration. In fact, Aller et al. (50) found that even SSSTR plus TDTR fitting was able to establish only an upper bound on their sample's $R_{\text{diamond/AlGaN}}$, likely due to the low AlGaN conductivity, low diamond conductivity, and high diamond thickness.

Another challenging regime includes samples with extremely high thermal conductivity. For example, the development of high-purity single-crystal materials (e.g., diamond) is of interest for thermal management of traditional electronics and for quantum devices where operations may occur at cryogenic temperatures. These materials are also fundamentally likely to have higher TBR (79), meaning the quantity of interest (k) will have only a minor contribution to the overall measurement. In **Figure 5a**, we consider the case of an aluminum transducer on diamond at cryogenic temperatures, where the diamond thermal conductivity is predicted to be 10 kW/m·K. None of the traditional measurements, nor most variants, do a suitable job of determining the diamond conductivity. Where multiple spot sizes are used for SSSTR, focused laser diameters as high as 100 μm are required, which means extremely high laser powers may be required to obtain an adequate signal. This does, however, highlight one benefit of SSSTR, where the direct dependence of TPD on the laser spot size can be taken advantage of relatively easily. By comparison, when the thermal conductivity is in the hundreds or even thousands, the traditional methods do well (as is the case with an Al/AlN material system, shown in **Figure 5b**). Reviewing measurements of diamond found in the literature (**Figure 5d**), a clear trend of increasing uncertainty can be seen with increasing diamond thermal conductivity [a trend that has been observed previously as well (80)].

We next consider the thin-film regime. Atomic layer deposition films can be on the order of nanometers, and thin metal films on the order of tens of nanometers are of interest as contacts for devices. For films of this size, it may be difficult or impossible to separate the thermal resistances of the interfaces from the thermal resistance of the film itself. Most measurements are most sensitive to the highest thermal resistance within the measured volume, meaning thin films with higher thermal conductivities will be especially difficult to measure. In **Figure 5c**, we consider the case of a hypothetical 40-nm film with a thermal conductivity of 5 W/m·K on an a-SiO₂ substrate. While this is not an exceptionally thin film or exceptionally high conductivity, none of the techniques do a satisfactory job of resolving the film's conductivity. Considering measurements from the literature (**Figure 5e**), there is a general trend in which the thinnest films measurable have the lowest thermal conductivities, and only thicker films are measurable for materials with high thermal conductivities. In some scenarios, separation of the boundary resistances from the resistance of the film can be achieved through creative experimental design. Measurement over a series of film thicknesses can enable extraction of thermal conductivities and TBRs for ultrathin films (81–83) (under the assumption that thickness and crystallinity effects do not impact the film

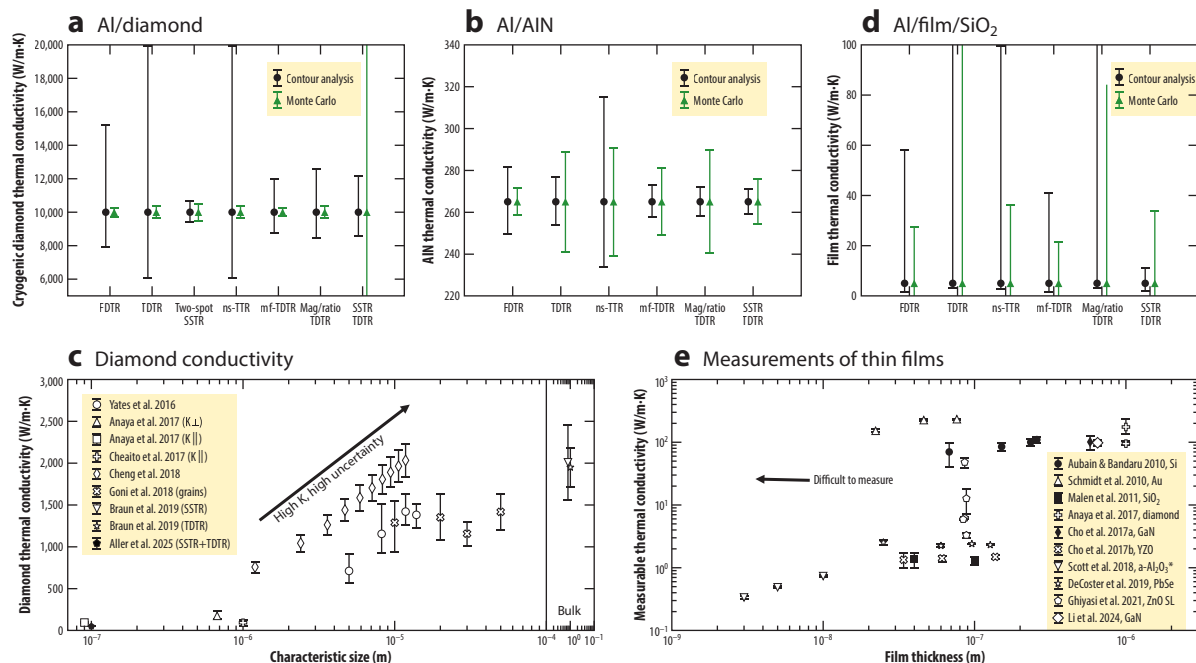


Figure 5

(a) We consider the hypothetical measurement of diamond at cryogenic temperatures. A high thermal conductivity means the thermal boundary resistance is dominant, making it extremely difficult to measure the diamond conductivity with a high degree of certainty. In the case where multiple spot sizes are used with SSTR, laser diameters as high as 100 μm are required to achieve reasonable uncertainty. (b) By comparison, a wide-bandgap semiconductor (AlN) with more moderate thermal conductivity is easily measurable. (c) This phenomenon is observable in published data on diamond, where the uncertainty tends to increase as the diamond conductivity increases (13, 37, 44, 50, 85–87). (d) In the measurement of thin films (40 nm, 5 W/m in this example), it is extremely challenging to differentiate between the resistance of the film and the resistances of the transducer/film and film/substrate interfaces. (e) This appears in the literature as well, where thin films with high conductivities are extremely difficult to measure with certainty (43, 44, 78, 88–94). Abbreviations: FDTR, frequency-domain thermoreflectance; Mag, magnitude; mf-TDTR, multifrequency TDTR; ns-TTR, nanosecond transient thermoreflectance; SSTR, steady-state thermoreflectance; TDTR, time-domain thermoreflectance.

thermal conductivity and that TBR is constant). Similarly, the use of a highly resistive substrate can encourage in-plane heat spreading within the film, while suppression of in-plane spreading in the transducer can enhance sensitivity to the film (84). This latter approach may not work for all measurement techniques, as only a measurement sensitive to in-plane properties will benefit.

4. HIGH- AND ULTRAHIGH-TEMPERATURE MEASUREMENTS

Above, we discuss measurements of conductive transport within materials (thermal conductivity, k) and across boundaries (TBR, R_K); however, in high-temperature environments (e.g., nuclear reactors or aerospace applications), radiative transport can also play a significant role. An understanding and ability to characterize coupled radiative and conductive processes at high temperatures are critical to enhance thermal management of rocket nozzles, gas turbine blades, and hypersonic leading edges, for example. Meanwhile, thermal measurements in these temperature regimes can be quite difficult, and a number of fundamental challenges arise when attempting to apply traditional measurement techniques in this area. This has led to the development of a number of metrologies to determine the conductive and radiative properties of materials at high temperatures (1,000–2,500°C) and ultrahigh temperatures (>2,500°C).

4.1. Thermoreflectance Limitations at High Temperatures

As temperatures exceed 1,000°C, significant experimental limitations on traditional thermoreflectance techniques become evident. The major limiting factor that restricts the effective use of these techniques at high temperatures is the requirement for a thin metal transducer. This transducer traditionally isolates pump absorption to the sample surface while providing an optically reflective surface for the probe. As measurements are pushed to higher temperatures, new transducer materials may be required (6, 95–97), as traditional transducer materials are limited by oxidation, melting (8), or thermomechanical failure (98) due to thermal expansion mismatch between the transducer and substrate. Thermoreflectance techniques are also limited by the need for the material's surface to be optically smooth (root mean square roughness on the order of tens of nanometers). For multiphase materials, composites, or highly porous materials for high-temperature applications (where high hardness and modulus are key), achieving a smooth-enough surface may not be possible.

At temperatures well above ambient, radiative heat transfer may become significant (e.g., up to 30% for measurements at 3,000°C; example calculations are available in the **Supplemental Appendix**). None of the thermal models used in the aforementioned measurement techniques capture radiative heat transfer. Quantification of emissivity (temperature dependent and wavelength dependent) and total radiative energy flux is critical in many applications, however, and measurements at high temperatures must be able to capture these effects.

4.2. Overview of Existing High-Temperature Measurements

While thermoreflectance techniques are limited in use at high temperatures, several optical techniques have been developed to study the thermal conductivity of materials at temperatures >1,000°C. These can be split into three categories: steady-state, transient, and indirect techniques. These techniques are discussed below and outlined in **Table 2**. For both steady-state and transient methods, the material system is heated and the temperature of the sample is inferred (e.g., via pyrometry as opposed to thermoreflectance), allowing determination of thermal transport properties (thermal conductivity and emissivity). For steady-state techniques, the temperature of the sample is largely held constant, such that the influence of heat capacity on the measurement is negated. In transient techniques, periodic or instantaneous heating (as in LFA) means knowledge of the heat capacity is required, or else the measurement is of thermal diffusivity as opposed to thermal conductivity. In both cases, radiative heat flux may be significant, meaning appropriate modeling is required to capture radiative losses. If total radiative emission is not known exactly, this will affect the extracted thermal conductivity (in much the same way uncertainty in unknown thermal properties affects traditional thermoreflectance experiments). Similarly, there may be difficulty in the measurement of true temperature via optical pyrometry, as this depends on the emissivity of the sample. High-temperature pyrometry-based techniques need to either avoid sensitivity to emissivity coefficients altogether (similar to thermoreflectance techniques, where dR/dT is not assumed) or accurately characterize these emissive properties. For indirect techniques, measurement of the sample's temperature is not required for the determination of thermal conductivity. Instead, the thermal conductivity is inferred via some other property (e.g., electrical resistivity, via the Wiedemann–Franz relation, or emissivity via the Hagen-Rubens or Drude relations). Indirect techniques might not always properly predict thermal conductivity, as they only have the ability to determine the electrical contribution to the thermal conductivity. Indirect techniques are insensitive to the radiative losses (which affect both steady-state and transient measurements), however, meaning they are typically much simpler to implement.

We review several techniques below, focusing on those that currently reach temperatures in the upper end of the high-temperature regime and those that can reach ultrahigh temperatures.

Table 2 Comparison of high-temperature thermal conductivity measurement techniques

Measurement technique	Temperature (°C)	Thermal conductivity (W/m·K)	Uncertainty (%)	Advantages	Disadvantages
Steady state					
Axial and radial heat flow method (99–108)	25–3,000	0.020–400	3–25	Reaches ultrahigh temperatures Can evaluate porous insulation materials and powders Direct k measurement	Large sample sizes and extended measurement times Large contact resistances at boundaries limit high conductivity measurement Cannot reach ultrahigh temperatures for electrical insulators Limited sample geometry Need to account for radiative losses Limited to solids at high temperatures.
Steady-state temperature differential radiometry (7)	25–4,000+	1–2,500+ ^a	5–20 ^a	Reaches ultrahigh temperatures Direct k measurement Containerless, optical, and noncontact Fast measurement times Can measure solids and liquids Front-side probing can measure thin coatings on substrates Accommodates a variety of sample dimensions	Complex and expensive experimental setup Known spectral emissivity or hyperspectral pyrometry needed to determine true temperature Total emissivity required for the thermal model at high temperatures Need to know laser absorptivity New technique; sensitivity analysis needed to reduce experimental uncertainty
Transient techniques					
Commercial and custom laser flash (109–113)	Commercial: 25–2,000 Custom: 25–4,000+	Diffusivity: 0.01–1,000 (mm ² /s)	Commercial: 3–10 Custom: 4–18	Reaches high temperatures (custom apparatuses can reach ultrahigh temperatures) Diffusivity measurement Simple to implement Large conductivity measurement range Simultaneous measurement of heat capacity and conductivity possible	Commercial systems only reach high temperatures For ultrahigh temperatures, complex experimental setup required Diffusivity measurement and knowledge of heat capacity required Total emissivity required for the thermal model at high temperatures Limited to solids
Indirect techniques					
Electrical resistivity (99, 114–118)	25–12,000+	No inherent limitation	4–15	Can reach ultrahigh temperatures Simple experimental setup Small sample sizes Fast measurement times Only captures electrical contribution to k Can evaluate solids and liquids Thermal model not required	Potential error from unknown variation in Lorentz number Cannot capture phonon conductivity Material must be electrically conductive

(Continued)

Table 2 (Continued)

Measurement technique	Temperature (°C)	Thermal conductivity (W/m·K)	Uncertainty (%)	Advantages	Disadvantages
Emissivity relation (119)	No inherent limitation ^b	No inherent limitation	3–10 ^b	Can reach ultrahigh temperatures Simple experimental setup Only captures electrical contribution to k Can evaluate solids and liquids Thermal model not required	Potential error from unknown variation in Lorentz number Cannot capture phonon conductivity Material must be electrically conductive Limited by ability to measure spectral emissivity

^aNew technique. Additional optimization and sensitivity analysis needed to reduce uncertainty. Conductivity range taken from steady-state thermoreflectance.
^bTemperature range and uncertainty limited by the ability to measure the spectral emissivity in the Hagen–Rubens or Drude wavelengths.

Each technique discussed and example datasets are shown in **Figure 6**. It is important to note that other lower-temperature techniques may be reconfigurable for use at high temperatures through adjustments to the experimental implementation.

While the experimental apparatus used for traditional thermoreflectance techniques is not viable for use at high temperatures, some of the procedures and concepts implemented may be beneficial when incorporated into high-temperature experiments. A few examples include the following: analysis of the transience of the temperature response and the quasi-steady-state temperature distribution simultaneously, consideration of the phase response for systems under cyclic heating so as to eliminate the need for normalization or a known emissivity, and the application of multiple modulated heater frequencies or multiple spot sizes to determine multiple parameters simultaneously (thermal conductivity, heat capacity, directional dependent thermal conductivity, TBR, etc.) (12, 13, 121).

4.2.1. Emissivity, emittance, and true temperature measurement. Many of the techniques we discuss below rely on an accurate measure of spectral emissivity (which is often temperature, angle, and wavelength dependent) or emittance (which may depend on surface texture, defects, impurities, etc.) and total hemispherical emissivity and emittance (emissivity or emittance summed over all angles and wavelengths). When a single-color pyrometer is used to monitor the temperature of the sample, the temperature reading is directly dependent on the spectral emissivity at the monitored wavelength. Similarly, if there is a significant amount of radiative heat flux, the total hemispherical emissivity and emittance are required to model the heat flow. We thus consider measurement of these properties to be of extreme importance for high- and ultrahigh-temperature applications and thermal property measurements, and we discuss briefly how this can be done.

4.2.2. True temperature measurement. Single-color pyrometry is the most basic and common form of optical pyrometry. Radiance is collected at a single discrete wavelength, and a radiance or brightness temperature T_{rad} (temperature assuming emissivity $\varepsilon = 1$) is determined from a derivation of Planck’s law or Wien’s law. This is shown in Equation 1,

$$T_{\text{rad}} = \frac{C_2}{\lambda \ln \frac{K}{S} + 1}, \tag{1}$$

where S is the voltage output from the pyrometer, K is the calibration factor (determined from a blackbody source or known radiance standard), C_2 is the second radiation constant, and λ is the center measurement wavelength of the pyrometer with narrow bandwidth. The true surface

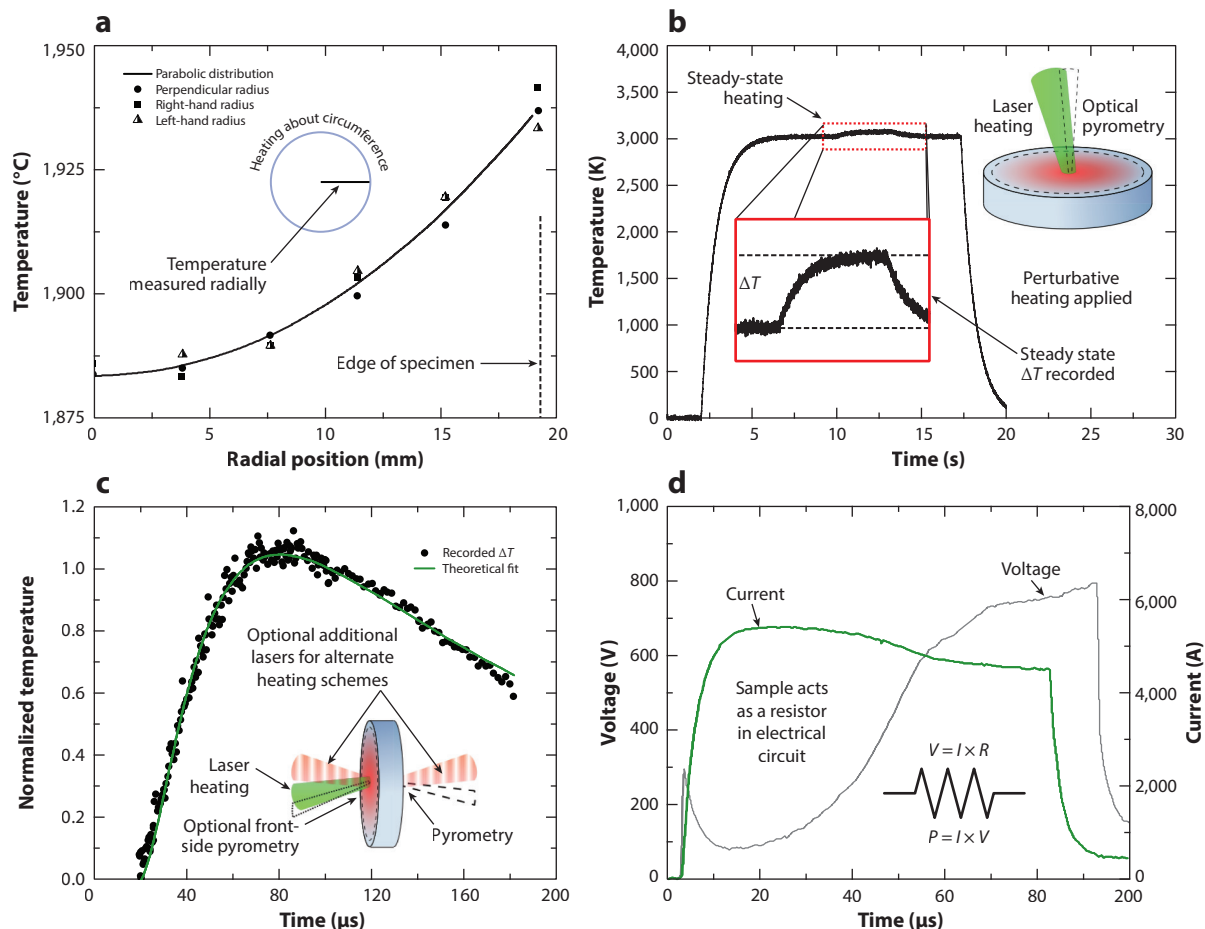


Figure 6

(a) Radial flow data from Israel et al. (120) are shown, with a measurement schematic shown in the inset. Thermal conductivity can be extracted by measuring the steady-state temperature along the radius of a specimen. Panel adapted from Reference 120 (public domain). (b) The steady-state temperature differential radiometry technique uses laser heating, and perturbative measurements are taken once a quasi-steady-state condition is reached. (c) An advanced laser flash apparatus [see, e.g., Vlahovic et al. (113)] may include front-side and back-side heating and temperature sensing, and radial heat flow modeling is typically required at high temperatures. Plots in panel c reproduced with permission from Reference 113 (CC BY-NC-ND 4.0); copyright 2017 The Authors. (d) Indirect measurement techniques use electrical conductivity as a proxy to infer thermal conductivity (99). In the example dataset from Kaschnitz et al. (118), the electrical current and voltage are used to determine electrical resistivity and, thus, thermal conductivity. Panel adapted with permission from Reference 118; copyright 1969 Plenum Publishing Corporation.

temperature (T_{true} or T) is then expressed via Planck's or Wien's law (122) as seen in Equation 2,

$$T_{\text{true}} = \frac{C_2}{\lambda} \frac{1}{\ln [\varepsilon_\lambda (e^{\frac{C_2}{\lambda T_{\text{rad}}}} - 1) + 1]}, \quad 2.$$

where ε_λ is the emissivity or emittance defined by Equation 3,

$$\varepsilon_\lambda(\lambda, T, \theta, \phi) = \frac{B(\lambda, T, \theta, \phi)}{B_b(\lambda, T, \theta, \phi)}, \quad 3.$$

where B is the spectral radiance of the material of interest at temperature T , and B_b is the spectral radiance of a blackbody (described by Planck's law at the same temperature). Unknown terms are sometimes calculated on the basis of an alternate measure of temperature for calibration (e.g., contact measurements, or using features in the data such as observation of a phase change at a known melting temperature). Care must be taken, however, to ensure the assumptions involved are applicable for the regimes at which measurements are performed. In this article, as well as in a majority of the outstanding literature, emissivity refers to the intrinsic radiative capacity of a material and emittance refers to the extrinsic radiative capacity of an opaque surface (123). Both of these are a function of temperature, angle, and wavelength, but emittance is also heavily dependent on the surface condition (e.g., defects and roughness). For proper determination of the true surface temperature using single-color pyrometry, knowledge of ε_λ must be known with high precision; otherwise, large measurement errors can occur. As ε_λ is highly variable, this parameter must often be estimated in situ to obtain results with low-enough uncertainty (124), especially for complex systems (materials other than pure elements).

This requirement has led to the development of pyrometric techniques that can either negate the need for the knowledge of ε_λ or allow for simultaneous determination of the true temperature and ε_λ , the most common of which is dual-color pyrometry. Radiance at two discrete wavelengths is measured, and the ratio of the radiated intensities can then be used to determine the true temperature. The key assumption in this analysis is that emissivity at the two wavelengths is gray (equal to one another). For this reason, two similar wavelengths are typically selected so as to minimize the spectral dependence from the material. If variation from graybody behavior is observed, then large measurement errors can occur (125). Many-wavelength pyrometers (number of $\lambda \geq 3$, sometimes hundreds of λ) have been developed to mitigate this, allowing fitting for the assumed spectral variance of emissivity or determination of the true temperature and emissivity without any previous knowledge or assumptions required (126–128). The latter developments would allow for thermal property measurements to be conducted at high and ultrahigh temperatures on a wider variety of materials. Notable multiwavelength pyrometric developments that do not require assumptions about the spectral dependence for the determination of the true temperature include the works of Felice and colleagues (127, 129–131), Ni et al. (132), and Taunay & Choueiri (128). The work of Araújo (126) is an excellent resource and review of advances and limitations of some of the multispectral pyrometric approaches conducted in the literature.

In addition to the application of multispectral pyrometry, other methods have been used to reduce the need for knowledge of ε_λ . The application of graphite coatings on the sample of interest is common, allowing the use of the blackbody assumption with only small measurement errors. This method is limited in practice to temperatures of $\sim 2,000^\circ\text{C}$ due to chemical reactions between the carbon, sample, and environment. Alternatively, reflectivity measurements can be used to characterize the sample's absorptivity in situ, which is then related to the emissivity via Kirchhoff's law (133). Pyrometry data can also be collected at the Christensen wavelength, which is the wavelength for heteropolar dielectric materials where the ε_λ can be estimated as unity and is temperature independent (134).

4.2.3. Total hemispherical emissivity and emittance. Characterizing the total hemispherical emissivity or emittance ε_t is of extreme importance to the measurement of thermal properties in cases where modeling of the energy balance of heat transfer is required. Considering the power balance between $Q_{\text{radiative}}$ and $Q_{\text{conductive}}$, the uncertainty in one will proportionally affect the uncertainty in the other. ε_t is defined in Equation 4,

$$\varepsilon_t(T) = \frac{\int_0^\infty \int_0^{2\pi} \int_0^{\frac{\pi}{2}} B(\lambda, T, \theta, \phi) d\theta d\phi d\lambda}{\int_0^\infty \int_0^{2\pi} \int_0^{\frac{\pi}{2}} B_b(\lambda, T, \theta, \phi) d\theta d\phi d\lambda}, \quad 4.$$

where $B(\lambda, \theta, \phi)$ emitted from the surface is integrated over all λ (wavelength), θ (angle relative to the surface, termed the glancing angle), and ϕ (angle about the surface normal vector) and compared with the blackbody radiance B_b of the same temperature. Note that the emissivity often does not change substantially based on the angle of radiance about the surface normal (ϕ), and this is often neglected. ε_t is often evaluated by measuring a percentage of wavelengths (ideally $\geq 99\%$), as not all wavelengths can be evaluated in practice. Additionally, as higher temperatures are reached, a smaller range of wavelengths need to be evaluated due to a shifting and tightening of the spectral radiance. There are a vast number of techniques and methodologies that can determine the ε_t (123, 135), but review of these techniques is largely beyond the scope of this article.

For many of the techniques we discuss below, ε_t is critical for the modeling of the energy balance of the input flux, and any error in ε_t will propagate to the measured parameters. The radiant flux can be modeled via Stefan–Boltzmann’s law where the total power radiated is defined by $Q_{\text{rad}} = A\varepsilon_t\sigma T^4$ (where σ is Stefan–Boltzmann’s constant and A is the surface area) (7).

4.3. Steady-State Measurements

Steady-state techniques are characterized by the measurement of heat flow or temperature gradients through a sample after a heat flux is applied once steady-state conditions have been reached (136). For some experiments, heat flow is isolated to one dimension, enabling the use of Fourier’s law to extract the thermal conductivity, and many steady-state experiments take advantage of this to simplify analysis. Other research may use more complicated geometries and require the use of the full transient or steady-state solution to the heat equation, as in the work of Milich et al. (7) and Konôpková et al. (137).

Depending on the particular apparatus implemented, high and ultrahigh temperatures can be reached. Traditional methods of heating include high-temperature furnaces and ohmic heating, which can reach temperatures of $\sim 3,000^\circ\text{C}$ with very specialized equipment (99–107). In the case of contactless, laser-based measurements, ultrahigh temperatures can readily be achieved using lasers in the tens of watts to the kilowatt power regime (depending on the sample properties and focused laser spot sizes) (7, 137).

A major limiting challenge to both steady-state and transient techniques is the requirement for accurate thermal modeling. In both cases, the heat flow away from the point of applied heat flux must be considered, with an energy balance between conduction and radiation included at high temperatures. Modeling of the heat transfer and knowledge of the total hemispherical emissivity and emittance can lead to larger uncertainties when compared with other metrologies (119). Measurement of materials with highly varying surfaces and unknown optical properties can add greater uncertainties to the fitted conductivity or will require a more complex apparatus to determine the radiative properties *in situ*.

4.3.1. Axial and radial heat flow method. The axial and radial heat flow methods are some of the longest-standing measurement techniques used at high temperatures. These have been used extensively since the 1960s to characterize a wide variety of materials, from fully dense systems to low-density porous insulators (101, 105, 106, 108). There have been a number of unique axial and radial heat flow methodologies that have been applied in the literature, but each operates off of the same fundamental principle. An axial or radial heat flux is applied to a rod, and the apparatus is designed in order to restrict heat flow to one dimension, as depicted in **Figure 6a**. For radial heat flow, this heat flux can be applied on either the central axis or the outer edge of the cylinder depending on the experimental apparatus (120, 136). The temperature of the rod can then be probed by either contact measurement or optical pyrometry and analyzed via the 1D Fourier’s law or a steady-state solution to the heat equation. The most common method of heating

for these experiments is through the use of either furnaces (high-temperature regime) or direct electrical heating (able to reach ultrahigh temperatures in some cases) (99–108). The use of radial heat flow is more commonly used at high temperatures when compared with axial heat flow as there is a reduced need for thermally insulating materials to be in contact with the specimen for long periods of time (102). In these cases, the temperature as a function of position on the sample can be monitored (see the inset in **Figure 6a**) and thermal properties extracted.

Thermal barriers typically needed in these techniques can make measurements on higher thermal conductivity materials a challenge, as contact resistances and properties of the insulation materials must then be considered in the thermal modeling (100). Although higher thermal conductivities can be reached if the contact resistances are measured in situ, maximum thermal conductivity measurements in the literature have been limited to around 400 W/m·K (100). Similarly, sample mounting can be a challenge, as heat flow through the sample holder may prevent reaching ultrahigh temperatures. These approaches also require larger sample sizes, in some cases requiring bars of materials 200 mm long and 10 mm in diameter to conduct a measurement with low-enough uncertainty (100). Long measurement times are also associated with the large geometries; in some cases, measurements can take multiple hours, with the materials being subject to the high-temperature conditions for the entire duration (100).

The techniques are capable of measuring porous insulators, however, and conductivities down to 0.02 W/m·K have been experimentally measured (102). Uncertainties ranging from 3% to 25% are found in the literature for axial and radial heat flow depending on the particular apparatus, measurement temperature, and thermal conductivity of the material evaluated (102, 107, 120). Despite the challenges mentioned, these techniques are often simple to implement when compared with some of the other methodologies we discuss below and are often able to reliably measure the thermal conductivity of porous thermal insulators at high temperatures (101).

4.3.2. Steady-state temperature differential radiometry. SSTDR is a new experimental metrology that we developed in a previous paper (7). This technique has the ability to measure the thermal conductivity of materials up to ultrahigh temperatures in both the solid and liquid states. For example, we measured the thermal conductivity of pure elemental tungsten above $\sim 1,800^\circ\text{C}$ up to and across the transition in the molten state at temperatures of $\sim 4,000^\circ\text{C}$. This was the first experimental measurement of the molten thermal conductivity of tungsten not conducted using the Wiedemann–Franz relation from electrical resistivity data.

SSTDR was inspired by the fundamentals of SSTR and the work of Braun et al. (13) and Konôpková et al. (137), as discussed above. These techniques function on the basis of modulated surface heating, where a pump laser is modulated (or the power is perturbed) at low frequencies, resulting in quasi-steady-state heating (**Figure 6b**). While SSTR uses a second laser (probe) to monitor the pump-induced changes in reflectance, SSTDR uses optical pyrometry. A solution to the 2D axisymmetric steady-state heat equation can then be used to model the experimental data and extract the thermal conductivity. Since a quasi-steady-state condition is reached, the measurement is insensitive to heat capacity.

Since SSTDR heats the sample via a localized high-power laser, and since the temperature is probed via pyrometry (noncontact), no insulation materials are needed, and the measurement times are on the order of tens of seconds. Several authors have reached temperatures exceeding $4,000^\circ\text{C}$ using a similar apparatus (124, 138–141). Similarly to SSTR, multiple beam sizes can also be used to gain further sensitivity to unknown properties. By nature of being a front-side pumped and front-side probed experiment, measurement of coating materials deposited on substrates is also possible. In the case of the work by Milich et al. (7), samples 10 mm in diameter and 2 mm in thickness were evaluated, which are relatively small compared with those in some of the other techniques discussed in this review.

The main limitations with the SSTDR technique (and others) are the modeling complexity and the requirement to characterize parameters such as absorptivity (relating to pump absorption), normal spectral emissivity (relating to the measured temperature), and total hemispherical emissivity at the temperatures of interest (needed for the modeling of the experimental results). As the technique is in its infancy, rigorous sensitivity and uncertainty analysis also needs to be conducted in order to determine the optimal experimental geometries (e.g., sample size compared with laser spot size) needed to reduce the experimental uncertainty. Initial estimates for the experimental uncertainty lie between 5% and 20% depending on the measurement temperature, experimental geometry, and knowledge of modeling parameters. The technique could also benefit from a rigorous analytical thermal model (rather than the finite difference models currently used). Modeling should capture the nonlinear radiative boundary conditions for finite (nonnegligible back-side radiative transfer) or semi-infinite samples (sample thickness exceeding laser spot size, adiabatic back-side boundary condition). Similarly, using a high-power laser for bulk heating with a secondary focused lower-power laser for the modulated pump (142) would mean semi-infinite conditions could be preserved in both the radial and axial directions (121). This would likely reduce the sensitivity to the thermal radiative losses in the high- and ultrahigh-temperature regimes, reducing the uncertainty in the thermal conductivity measurement.

While SSTDR is still in the early stages of development, it demonstrates that concepts from traditional thermoreflectance techniques (SSTR in this case) can be applied to pyrometry in the high- and ultrahigh-temperature regimes, and we expect other concepts from the traditional techniques can similarly be applied.

4.4. Transient Measurement

Transient measurement techniques monitor heat flow or a temperature gradient through a sample as a function of time, following the application of a heat flux. These methods are a measurement of thermal diffusivity or effusivity, and heat flux is typically applied through periodic or pulsed heating (1, 136). The thermal conductivity can be extracted from the diffusivity measurement if the volumetric heat capacity at the temperature of interest is known (1).

Similarly to steady-state techniques, high-power lasers (7, 124, 138–140) or furnaces and inductive heaters (109, 143) can be used to heat the sample before the application of the transient pulse. At elevated temperatures, the sample is typically probed via optical pyrometry on the surface opposite of the laser heater (144). At moderate temperatures and with proper experimental implementation, 1D solutions to the heat equation can be used to model experimental results (111). At higher temperatures, multidimensional solutions to the heat equation (including radiative boundary conditions) are required to properly model the experiment (110, 111, 145, 146).

Depending on the sample and experimental configuration, it may take multiple tens of seconds for the system to reach the steady state. This is one area where transient techniques excel, as multiple repeated measurements can be acquired without waiting for the system to equilibrate. Sensitivity to heat capacity can also be considered an advantage in cases where thermal conductivity is known and heat capacity is the parameter of interest (e.g., when paired with indirect measurement schemes that separately measure thermal conductivity). With proper implementation, some transient techniques can also simultaneously fit for thermal conductivity and heat capacity of materials at high temperatures, as demonstrated by Ronchi et al. (147).

Meanwhile, if heat capacity is not known precisely, uncertainty in heat capacity can result in large propagated uncertainty in the measurement of thermal conductivity (7, 137, 147). Measurements are also mostly limited to solid materials due to reactions between the liquid and sample holder, and transient flows on the molten material can affect experimental results (99). As is the case with steady-state techniques, transient methods also require knowledge of total hemispherical

emissivity and emittance (for modeling of the radiative boundary condition) and spectral emissivity and emittance (at the pyrometer measurement wavelength) (110, 111, 145).

Commercial LFA is the only widely used, commercially available measurement that can measure thermal diffusivity at high temperatures. Typical commercial systems are limited to measurement temperatures of around 2,000°C, and the samples are heated with a furnace. Some commercial systems can reach temperatures up to 2,800°C, but measurements up to these temperatures are uncommon. Commercial LFA systems conduct measurements on cylindrical samples where either a flash lamp or pulsed laser applies an instantaneous heat flux to the front side of the surface of the specimen. A high-speed pyrometer then monitors the time-dependent temperature rise of the back surface of the sample.

Numerous other high-temperature transient experimental apparatuses have also been constructed, many of which are based on the laser flash methodology. By using high-power CW laser heating, temperatures well above 3,000°C can be reasonably achieved. Others make use of furnaces or inductive heating, but these are mostly limited to the high-temperature regime (109). A secondary pulsed laser is then used to apply the transient heating, and the changing temperature on the back side is monitored.

At low temperatures, a 1D assumption can be made, and the analytical solution to the heat equation (Equation 5) can be used for analysis,

$$\alpha = 0.1388 \times \frac{d^2}{t_{50}}, \quad 5.$$

where d is the sample thickness, t_{50} is the time for half the maximum temperature rise to be reached, and α is the thermal diffusivity k/C (1). This equation is only valid for the following assumptions: The pulse energy is uniformly applied to the surface, the pulse duration is much shorter than the time it takes for the maximum temperature to be reached, the heat flow across the specimen is 1D, and the peripheral heat loss is negligible (148). A number of previous papers extended this methodology for cases where one or more of these requirements were not met, as is often the case for high-temperature measurements (110, 111, 145, 146). Once the thermal diffusivity is determined, the thermal conductivity can be found, assuming the volumetric heat capacity is known.

In some cases, the front surface temperature is also monitored, and often as a function of the radius (110, 112) (**Figure 6c**), which then requires the use of a multidimensional solution to the heat equation to fit the experimental data (110, 111, 113) (**Figure 6c**). Where laser heating is used, the use of insulation around the sample may not be required, and a wide variety of sample sizes can be considered. Some experiments have also included the use of aerodynamic and electromagnetic levitation to further restrict conductive losses through a sample holder. These experiments also minimize chemical reactions and oxidation, allowing for measurements to be conducted in the liquid state (145).

Given the long history of laser flash, commercial systems can achieve uncertainties in the diffusivity measurement ranging from 3% to 10%, while custom apparatuses have shown uncertainties ranging from 4% to 18%, depending on the apparatus used, sample of interest, and measurement temperature (109, 110). Commercial systems also typically benefit from ease of use, while the custom systems are better able to reach ultrahigh temperatures despite the added complexity. Analysis may also be complicated when consideration of spectral or total hemispherical emissivity is required.

4.5. Indirect Measurement

Indirect measurement techniques are characterized by the measurement of a separate material property, where that property is then correlated to the thermal conductivity via some physical

relation. Most notable examples of this at high temperatures are through electrical resistivity and emissivity measurements, which can then be used to infer the thermal conductivity via the Wiedemann–Franz and the Hagen–Rubens and Drude relations, respectively (114, 115, 118, 119).

These methodologies have the advantage of reduced experimental complexity compared with the aforementioned techniques since the sample temperature is no longer measured. This also eliminates the challenges associated with thermal modeling (capturing conductive and radiative heat transport) (119), which thus yields higher measurement precision and lower resulting uncertainty. Additionally, due to the simpler experimental configurations and fast measurement times, ultrahigh temperatures can easily be reached. Temperatures much greater than 12,000°C have been reached using high electrical discharge (resistive pulse heating) (114, 115, 118), and high-power lasers or high-temperature furnaces have been used to reach temperatures >3,000°C.

The major disadvantage of these methods is the requirement that the materials be electrically conductive. Similarly, since the thermal conductivity is derived through relations to electronic conductivity (e.g., the Wiedemann–Franz relation), only the electronic contribution to the thermal conductivity can be determined. If a phonon contribution to the thermal conductivity is significant, the total thermal conductivity can not be determined. As such, the relation being used and its applicability to the material system at hand must be considered. For electrical resistivity measurements, variation in the Lorentz number can also occur, complicating the determination of thermal conductivity (104).

4.5.1. Electrical resistivity relation. Electrical resistivity-based measurement techniques are the most widely used indirect techniques for determining thermal conductivity at high and ultrahigh temperatures. These are based on measurement of the electrical resistivity, which is then related to the thermal conductivity via the Wiedemann–Franz relation:

$$k(T) = \frac{L_0 T}{\rho_{\text{el}}(T)}, \quad 6.$$

where ρ_{el} is the electrical resistivity, T is the temperature, and L_0 is the Lorentz number (typically $2.44 \times 10^{-8} \text{ V}^2 \text{K}^{-2}$ for metals at high temperatures) (122).

The most notable experimental implementation of this methodology at ultrahigh temperatures is the submicrosecond ohmic pulse heating technique developed by Gallob et al. (114) and later improved on by Kaschnitz et al. (118) and Pottlacher et al. (115). These authors demonstrated the measurement of numerous materials in the solid and liquid states between 2,000°C and 12,000°C. In this technique, a capacitor bank (charged between 4 and 15 kV) is discharged, producing fast ohmic heating (50–100 μs) in a small cylindrical wire specimen and achieving heating rates of more than 10^9 K/s . The voltage drop and current across the specimen are monitored, and the temperature is measured via optical pyrometry. An example schematic and voltage and current data are shown in **Figure 6d**. Further description of this technique can be found in the works of Pottlacher et al. (116) and Cagran (99).

This technique allows the measurement of thermal conductivity of electrically conducting materials in the solid and liquid states, across the phase transition, and up to their boiling points (117). The extremely fast measurement times also allow for evaluation of the material at ultrahigh temperatures before any chemical change of the system can occur. Additionally, no thermal modeling is required, negating uncertainty from emissivity as discussed above.

4.5.2. Emissivity relation. A relatively new methodology was recently introduced by Fisenko & Lemberg (119) involving the relation of a material's emissivity to its thermal conductivity. This approach uses both the Hagen–Rubens and Drude relations to tie the spectral or total emissivity to electrical conductivity and subsequently the Wiedemann–Franz relation to tie the electrical conductivity to thermal conductivity.

For this approach, the total normal emissivity (spectrally averaged) from 1 to 10 μm is measured, which goes into the Hagen–Rubens relation,

$$\varepsilon_t(T, 1 < \lambda < 10 \mu\text{m}) = 5.78 \times \left(\frac{T}{\sigma}\right)^{\frac{1}{2}} - 17.8 \times \left(\frac{T}{\sigma}\right) + 584 \times \left(\frac{T}{\sigma}\right)^{\frac{3}{2}}. \quad 7.$$

The total normal emissivity (spectrally averaged) above 10 μm is measured, which goes into the Drude relation,

$$\varepsilon_t(T, \lambda > 10 \mu\text{m}) = 5.78 \times \left(\frac{T}{\sigma}\right)^{\frac{1}{2}}. \quad 8.$$

Once the measure of electrical conductivity σ is obtained, the Wiedemann–Franz relation (Equation 6) can be used to relate it to thermal conductivity. Note that the measurement of ε_t requires additional unknowns; however, the use of both Hagen–Rubens and Drude relations enables extraction of all required parameters. A rigorous description of the analysis is provided by Fisenko & Lemberg (119).

This is thus a noncontact method capable of determining thermal conductivity in both solid and liquid states. The need for modeling conductive and radiative heat flux is eliminated, as was the case with the electrical resistivity measurements (119). This approach is limited to materials that have a dominant electronic contribution to the thermal conductivity, however, as it relies on the Hagen–Rubens, Drude, and Wiedemann–Franz relations, all of which describe electronic transport of materials.

4.6. Measurement Comparison (ZrC_x and W)

We discuss above a wide variety of techniques for the measurement of the thermal conductivity of materials at high and ultrahigh temperatures, each with their own advantages and limitations. In **Figure 7**, we present a comparison of various thermal conductivity measurements using many of these techniques on two well-studied material systems, ZrC_x (**Figure 7a**) and elemental W (**Figure 7b**). While numerous studies have measured these materials, only two have reached the ultrahigh-temperature regime ($>2,500^\circ\text{C}$), including measurement of the liquid state. The work of Pottlacher (149) relies on electrical resistivity measurements and demonstrates the ability to measure at exceptionally high temperatures, while the work of Milich et al. (7) is currently the only direct measurement of thermal conductivity (not reliant on the Wiedemann–Franz relation). The development of additional metrologies and advancement of existing techniques are likely needed to expand the realm of thermal measurements into the high- and ultrahigh-temperature regimes.

5. CONCLUSIONS

In this review, we discuss the challenges associated with nano- and microscale thermal measurements (thermal conductivity k , TBR R_K , and emissivity ε).

For traditional measurements, LFA and laser-based pump/probe thermoreflectance experiments are robustly capable of measuring a large range of properties and samples; however, a few key gaps persist. Notably, it is still quite difficult to accurately measure semiconductor–semiconductor interfaces, thin films, and materials with high conductances. In these cases, a truly rigorous treatment of experimental uncertainty and measurement error is required. In many instances, while signals can be obtained, uncertainty and measurement error dominate, and thermal parameters can not be rigorously established. Several variants on the existing techniques are also discussed, some of which could be developed further or inspire the development of additional methods targeting these difficult-to-measure properties.

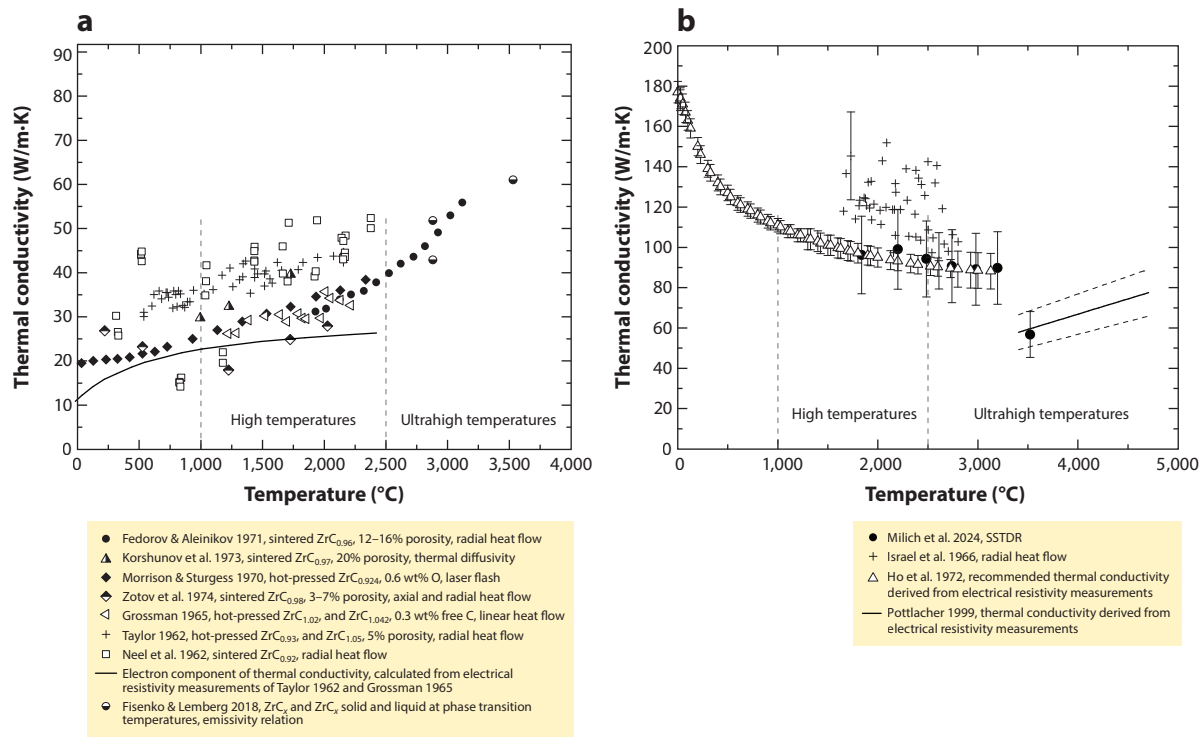


Figure 7

Comparison of thermal conductivity data reported for (a) ZrC_x and (b) elemental W conducted using different experimental techniques. Literature data (107, 150–155) for ZrC_x collected by Jackson & Lee (103) and reported by Fisenko & Lemberg (119). Data for W reported by Milich et al. (7), Israel et al. (120), Ho et al. (156), and Pottlacher (149). Abbreviation: SSTDR, steady-state temperature differential radiometry.

In the high-temperature (1,000–2,500°C) and ultrahigh-temperature (>2,500°C) regimes, several key fundamental limitations arise with thermoreflectance measurements, and pyrometry is typically used instead. One key challenge is the requirement for a known emissivity at the monitored wavelength(s), where uncertainty in the spectral emissivity will directly contribute to the total measurement uncertainty. Similarly, radiative energy flux may become significant, and modeling must account for this in order to accurately extract thermal properties.

Developments in many of these areas are needed, and it is our hope that the discussion included here can guide the thermal measurement community in advancing existing techniques and developing new techniques to address the challenges outlined. Truly rigorous uncertainty and measurement error quantification is required to establish trust in low-temperature measurements, and emissivity and other experimental challenges must be overcome for the high- and ultrahigh-temperature regimes.

DISCLOSURE STATEMENT

J.T.G. is the CEO and cofounder of Laser Thermal Analysis, Inc., a company that uses optical technology for thermal metrology. D.H.O. is Vice President of Product Development for Laser Thermal Analysis, Inc. J.L.B. is Vice President of Strategy and Programs for Laser Thermal Analysis, Inc. P.E.H. is the Chief Science Officer and cofounder of Laser Thermal Analysis, Inc. The

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