Improved 3- ω measurement of thermal conductivity in liquid, gases, and powders using a metal-coated optical fiber

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A novel 3 ω thermal conductivity measurement technique called metal-coated 3 ω is introduced for use with liquids, gases, powders, and aerogels. This technique employs a micron-scale metal-coated glass fiber as a heater/thermometer that is suspended within the sample. Metal-coated 3 ω exceeds alternate 3 ω based fluid sensing techniques in a number of key metrics enabling rapid measurements of small samples of materials with very low thermal effusivity (gases), using smaller temperature oscillations with lower parasitic conduction losses. Its advantages relative to existing fluid measurement techniques, including transient hot-wire, steady-state methods, and solid-wire 3 ω are discussed. A generalized n -layer concentric cylindrical periodic heating solution that accounts for thermal boundary resistance is presented. Improved sensitivity to boundary conductance is recognized through this model. Metal-coated 3 ω was successfully validated through a benchmark study of gases and liquids spanning two-orders of magnitude in thermal conductivity. © 2011 American Institute of Physics. [doi:[10.1063/1.3593372](https://doi.org/10.1063/1.3593372)]

I. INTRODUCTION

While thermal transport properties have been measured for centuries, measurement techniques for thermal conductivity have revived importance due to the emergence of novel and challenging to measure materials, including thin-films, superlattices, biological samples, and nanomaterials.^{1–10} Enhanced thermal property information will aid a diverse range of applications, including improved microprocessor heat removal,^{11,12} higher efficiency thermoelectric materials,^{13,14} and more effective minimally invasive cryosurgery.¹⁵ The 3 ω method and time domain thermo-reflectance (TDTR) are the dominant tools for measuring thermal conductivity in solids, while fluid samples have typically been studied with transient hot-wire (THW). Recent studies have used modified versions of 3 ω and TDTR to measure fluid samples.^{9,16} Herein we introduce and validate a modified 3 ω method for fluid measurement using a metal-coated glass fiber as a heater/thermometer. This technique, unlike prior 3 ω incarnations, is sensitive to liquids, gases, powders, and aerogels.

Current methods for sensing fluid thermal properties can be categorized by the thermal forcing function: steady-state, impulse, or periodic. Steady-state thermal property techniques apply a known one-dimensional steady heat flux and measure the temperature versus distance. The thermal conductivity is the quotient of heat flux per unit area and the slope of temperature change with distance. Steady-state techniques have serious drawbacks that include long settling times, natural convection effects, radiation losses, complex 3D isotherms, and relatively large test samples.^{17–19} The most widely used steady-state technique, guarded hot plate, often uses meter squared test samples.¹⁹

While steady-state techniques are conceptually simple, accurate implementation is fraught with difficulties due to these factors.

Amongst non-steady state techniques for fluid property measurement, THW dominates.^{4,8,20} THW analyzes the thermal impulse signal in the time domain, rather than in the frequency domain. THW sends a thermal impulse from a thin heating wire into the surrounding fluid of interest. Simultaneously, the resistance of the wire increases with the wire's increasing temperature, and the coefficient of thermoresistance characterizes the resistance change with temperature. Therefore, the measured resistance corresponds to the wire temperature. From the time-resolved wire temperature, the thermal conductivity of the surrounding fluid can be determined.^{21,22} Because the heating step-impulse contains signals at all frequencies, the data used to determine thermal conductivity will contain long wavelength heat-waves. These long thermal waves decay in amplitude slowly with increasing depth (long thermal penetration depths), and so will be most affected by the container dimensions, especially as time progresses. Because these long-wavelength components of the heat impulse cannot be excluded from the thermal impulse, they have greater sensitivity to the test vessel boundaries than frequency-domain techniques. Hence, THW requires either truncating data sets to consider only very short times with compromised accuracy, or the use of a bigger fluid sample relative to frequency domain measurements. This increased fluid requirement is a disadvantage, particularly when the fluid under test cannot be synthesized in large quantities, such as biological fluid samples.

Frequency domain based 3 ω techniques use a resistance heater/thermometer to periodically heat a sample and simultaneously monitor its thermal response. For solids, the heater/thermometer is a thin metal strip that is micro-fabricated on the sample being tested (see Fig. 1(a)). The alternating joule heating applied to the metal strip has current

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FIG. 1. (Color online) Comparison of three 3ω techniques: (a) Standard 3ω – heater is built on solid substrate. The fluid to be sensed is above the heater. The outer probes drive a 1ω current and the inner probes sense a third-harmonic voltage signal 3ω . (b) Solid-wire 3ω – has the 1ω current driven through the length of the wire and 3ω voltage sensed across the length of the wire. The fluid to be measured surrounds the fiber. (c) Metal-coated 3ω – employs a glass fiber as a backbone for a nanometer thick metal coating. The voltage and current is applied similar to solid metal wire, with the fluid surrounding the metal coating.

periodically passing through it at a frequency of 1ω . Because joule heating is a quadratic function of current ($P = I^2 R$), the heating occurs at twice the current drive frequency, at 2ω . This periodic heating at 2ω generates periodic traveling thermal waves that decay in amplitude with increasing depth into the surrounding medium. The thermophysical properties of the surrounding medium will affect the amplitude and phase of the temperature oscillations in the heating line in a way that can be predicted using analytical or numerical conduction models.^{23,24}

The amplitude will exponentially decay into the solid, with a characteristic length scale referred to as, the thermal penetration depth (L_p). Thermal penetration depth is a function of the thermal diffusivity α of the material, and the current's angular frequency ω ($L_p = \sqrt{\alpha/\omega}$). As the drive frequency is swept from low to high-frequency, the thermal penetration depth shrinks. Likewise, the magnitude of the temperature oscillations in the heating line will vary with driving frequency. Large temperature oscillations occur at low-frequency, while small temperature oscillations occur at high-frequency. This temperature versus frequency behavior depends on thermal conductivity and therefore thermal conductivity can be determined from this information. As the temperature in the wire oscillates at 2ω , so does the resistance, as quantified by the coefficient of thermoresistance ($\beta = \frac{1}{R_{El}} \frac{dR_{El}}{dT}$). The voltage, the product of current, and resistance, will have a component at 3 times the current frequency (3ω) stemming from product the 1ω current and 2ω resistance signals.²³

When the standard 3ω is used to measure fluids, the heater is patterned on a solid and the fluid is placed over this heater. A significant disadvantage of standard 3ω for measuring fluid properties is that the solid substrate typically has much larger thermal effusivity $\kappa \rho c_p$, than a fluid. Because the heat generated in the metal strip will split between the solid substrate and the fluid based on their ratio of thermal effusivities,²⁵ much of the heat will enter the substrate rather than the fluid. For example, for a standard 3ω experiment built on a Pyrex substrate measuring air at standard temperature and pressure, greater than 250 units of heat will enter the Pyrex for every unit of heat entering the air. Consequently, a significant part of the 3ω signal comes from the substrate, rather than the fluid under test, which decreases the signal of

interest's size.⁴ Solid-wire 3ω uses a solid heating wire that is immersed within the sample, rather than a microfabricated heater on a planar substrate. Similar to the standard 3ω , much of the periodic heating propagates radially into the high thermal effusivity metal wire, rather than into surrounding fluid.

A. Description of metal-coated immersed wire 3-omega

Metal-coated immersed wire 3ω enhances prior fluid measurement techniques by advancing the beneficial qualities of THW, solid-wire 3ω , and standard 3ω .^{3,7,9,26} Metal-coated 3ω improves THW and solid-wire 3ω 's heater by using a micron-scale metal-coated optical fiber rather than a thin solid metal wire. The thinner metal-cross-section results in signal improvements due to increased axial electrical and thermal resistance. Increased electrical resistance leads to a larger 3ω signal, which enhances the experimental signal to noise ratio. Increased axial thermal resistance reduces the boundary effects of the vessel, by reducing the thermal conductivity of the heating element. Further, with metal-coated 3ω , less of this periodic heat is parasitically lost into the low thermal effusivity glass fiber, which leads to greater sensitivity to the thermal conductivity of the surrounding fluid. Unlike THW, metal-coated 3ω operates in the frequency domain, allowing control of L_p to accommodate small samples. Finally, simulations with conductance models show an increased sensitivity to thermal boundary resistance. These signal improvements have a tangible effect, thus making possible measurements of low thermal effusivity materials that have been hitherto impossible with 3ω techniques.¹⁰

B. Quantitative analysis of the relative advantages of metal-coated 3ω

The 3rd harmonic voltage signal strength per temperature oscillation intensity, $\Delta V_{3\omega}/\Delta T_{2\omega}$, characterizes a key performance benefit of metal-coated 3ω relative to prior solid-wire 3ω techniques. Larger $\Delta V_{3\omega}/\Delta T_{2\omega}$ enables thermal property measurement with smaller $\Delta T_{2\omega}$,

$$\frac{\Delta V_{3\omega}}{\Delta T_{2\omega}} = \frac{\Delta R_{2\omega} I_{1\omega}}{\Delta T_{2\omega}} = \beta R_{El} I_{1\omega}. \quad (1)$$

To maintain a given $\Delta T_{2\omega}$ the current and resistance must vary such that power dissipated, $I_{1\omega}^2 R_{EI}$, remains constant. Therefore $I_{1\omega}$ must vary as $(\sqrt{R_{EI}})^{-1}$. Additionally, R_{EI} varies inversely with the cross-sectional area of the metal heating layer, A_H ,

$$\frac{\Delta V_{3\omega}}{\Delta T_{2\omega}} \propto \sqrt{R_{EI}} \propto \frac{1}{\sqrt{A_H}}. \quad (2)$$

Relative to solid metal wires, the thin metal film coating can have more than 2 orders of magnitude smaller A_H . The below expression compares the signal to temperature oscillation magnitude for metal-coated 3ω to solid metal 3ω , provided they are measuring the same fluid and the heating layer is the same metal,

$$\frac{(\Delta V_{3\omega}/\Delta T_{2\omega})_{MC}}{(\Delta V_{3\omega}/\Delta T_{2\omega})_{SW}} \propto \frac{\sqrt{A_{H,SW}}}{\sqrt{A_{H,MC}}}. \quad (3)$$

For instance, the approximate signal improvement in a 25 μm diameter glass fiber coated with 100 nm of platinum relative to a 25 μm solid platinum wire is eleven-fold.

Metal-coated 3ω further reduces previous solid metal 3ω and THW periodic heating losses at the interface of the heater with bulk electrical connection due to diminished axial thermal conduction proportional to $\sqrt{(k\rho c)_S A_{XC}}$, where $(k\rho c)_S$ represents the thermal effusivity of the fiber or wire substrate. This approximation of periodic heat through the connector comes from applying 1-D planar periodic heat conduction model, where the 1D axis is through the fiber or solid-wire.

Ideally, all the periodic heat generated from the 3ω heating element would radially enter the fluid-under-test such that the oscillating temperature depended entirely on the fluid properties. The total heat generated by the heater is split between the fluid and parasitic dissipation mechanisms,

$$q_H e^{i\omega t} = q_{\text{fluid}} e^{i\omega t} + q_{\text{parasitic}} e^{i\omega t}. \quad (4)$$

While both standard 3ω and metal-coated 3ω have parasitic losses of heat into the glass substrate or glass fiber, a greater fraction of the periodic heating enters the fluid with metal-coated 3ω .

The ratio of heat entering the glass substrate relative to the fluid for standard 3ω can at best approach the ratio of the square root of thermal effusivities with an infinitely thin heating element (low frequency limit).²⁵

For metal-coated and solid-wire 3ω , the ratio of the periodic heat entering the fluid varies with frequency because the ratio of L_p to the fiber diameter changes with frequency. The curves in Fig. 2, based on the conduction model derived in Eqs. (6)–(12), show that more heat enters the fluid for all frequencies in metal-coated 3ω . For metal-coated at very low-frequency almost all the heat goes into the fluid-under-test, while at very high frequencies, the ratio asymptotes to the standard 3ω ratio. At very low frequencies, the thermal penetration depth is greater than the fiber diameter, so the heat waves pass through the glass fiber and enter the fluid-under-test relatively undistorted. At high-frequencies, the thermal penetration depth is very short relative to the diameter, and the radial geometry approaches the standard 3ω ratio. In solid-wire 3ω , heat is periodically generated volumetrically

FIG. 2. (Color online) Fraction of zero time-average periodic heating entering fluid-under-test (air). For a metal-coated fiber (25 μm diameter glass fiber coated with 100 nm of platinum) more of the heat enters the fluid-under-test than with solid metal wire 3ω . Thermoproperties were taken for Pyrex and air at standard temperature and pressure. The analytical model for metal-coated and solid-wire 3ω are developed in a later section.

throughout the wire, such that thermal oscillations from volumetric heating near the center of the solid-wire may dampen greatly before the thermal wave has traveled to metal-fluid interface. Therefore, the fraction of heat entering the fluid is always smaller than metal-coated and does not asymptote to the standard 3ω value.

Radiation heat losses can also be ignored without consequence for our validation test on fluids at room temperature, as much less heat is periodically radiated than periodically conducted through the fluid,²³

$$\frac{Q_{\text{Rad}}}{Q_{\text{Cond}}} = \frac{8\varepsilon\sigma T^3}{\sqrt{k\rho c\omega}}. \quad (5)$$

Worst case radiation occurs at high temperature and low frequencies, but will be negligible in our temperature range with the fluids tested.

Another heat transfer mode that must be considered when measuring a fluid is natural convection. The insignificance of natural convection to metal-coated 3ω is shown by the steady-state natural convection fluid speed being order of magnitudes slower than the conduction wave propagation speed. Natural convection speeds were estimated as $u_{NC} \sim (g\beta\Delta TL)^{1/2}$, and the conduction wave velocity was estimated as product of thermal wavelength and frequency, $v_{\text{CondWave}} = L_p\omega = \sqrt{2\alpha\omega}$.²⁵ For air, u_{NC} is similar in magnitude to v_{CondWave} only when the heating frequency is far less than 1 Hz. Our measurements are typically taken at 10–1000 Hz.

C. Manufacturing technique

The manufacturing of the metal-coated glass fiber has two distinct steps, coating a glass fiber in metal and electronically mating the metal-coated fiber to a bulk electrical connector.

To adapt a conventional sputtering machine (Perkin Elmer 6J) to coat a cylinder uniformly, fibers were strung onto a spool, which was then rotated with a vacuum motor (Fig. 3(a)). This “sputtering lathe” evenly coated the fiber in

FIG. 3. (Color online) (a) “Sputtering lathe” – glass fibers are strung across spool which rotated during metal deposition to ensure circumferentially even coating. Glass fibers are adhered to the spool with Kapton tape. (b) Metal-coated fiber cross-section scanning electron microscope image.

metal, as the rotation period was much shorter than the sputtering time. A 10 nm titanium adhesion layer and 50–100 nm of gold or platinum were applied to the fiber. To convert from standard planar sputtering deposition rates to “sputtering lathe” deposition rates, the rates were cut by a factor of π . This ratio stems from the ratio of the fiber’s areal footprint (i.e., its diameter times its length), to its actual surface area (i.e., its circumference times its length).

The raw glass fiber, used as the backbone of the metal-coated fiber, has a 1/1000 in. diameter and was supplied by Fiberoptics Technologies, Inc. (0.125 in. $\times$ 30 in. Raw Fiber Bundle, 0.66 NA, 0.001 in. Fiber Diameter, FTIRF12753). This fiber consists of a leaded glass core (Schott F2) and borosilicate cladding (Schott 8250), shown in Fig. 3(b). The diameters and properties of the two different glass sections were provided by the supplier and are included in Table II. Bulk electrical contact was made with the metal-coated fiber using EpoTech H27D, silver electrically conducting epoxy ($\rho \leq 0.0005 \Omega/\text{cm}$ @ 300 K) without tensioning the fiber. The epoxy was cured at 80 °C for 24 h. Measurements were insensitive to the length of fiber; measurements for this benchmark study were performed with a fiber roughly 5 mm in length, but both shorter (2 mm) and longer (20 mm) lengths were used with air without significant differences. Insensitivity to the fiber length, and hence resistance, indicates that the resistance of the epoxy and leads does not detrimentally impact our results.

II. ELECTRONIC CONFIGURATION

The experiment is performed by immersing the fiber in the fluid under test and driving a periodic current across it while measuring the resultant voltage. The current sourced by a Keithley 6221 DC and AC Current Source passes in series through the metal-coated fiber and a calibration resistor (Fig. 4). The calibration resistor is tuned to equal the nominal resistance of the metal-coated resistor during operation. The voltage across the sense and calibration resistors are buffered by an instrumentation amplifier (LT 1167). The instrumentation amplifier outputs into the lock-in amplifier (Stanford Research Systems SR830). The lock-in amplifier performs a real-time Fourier decomposition of the signal. To remove noise introduced by the current source, the lock-in amplifier analyzes the difference in voltage between the metal-coated fiber and calibration resistors (A-B Mode). The amplitude and phase of the third harmonic (3ω) signal is collected via a MATLAB program that interfaces with the

FIG. 4. (Color online) 3ω electronic schematic: Current source provides a 1ω current to the series circuit consisting of the metal-coated filament and the calibration resistor. The calibration resistor is adjusted such that it equals the average sense filament resistance. The voltages across the metal-coated filament and calibration resistor are buffered by an instrumentation amplifier and fed into a lock-in amplifier. The lock-in determines the magnitude and phase of the third-harmonic (3ω) voltage, thereby measuring the magnitude and phase of the metal coating’s temperature.

electronics. The MATLAB program sweeps through a range of frequencies and records the magnitude and phase of the 3ω signal, which is the input used to determine the thermal conductivity of the fluid under test.

A. Generalized solution for determination of thermal conductivity

The thermal conductivity of the unknown fluid is determined by fitting phase and amplitude data with the analytical solution to the heat conduction equation, using k_{fluid} as the sole fitting parameter. The fit is optimized using least-squares minimization. We now present a generalized solution for an n -layer concentric cylindrical structure with heat generated periodically in one or more layers.

The heat conduction equation in cylindrical coordinates is²⁷

$$k_n \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T_n}{\partial r} \right) + q_n''' = \rho_n c_n \frac{\partial T_n}{\partial t}, \quad (6)$$

where n indexes the layer. The multilayer metal-coated 3ω model has $q_n''' = 0$, except in the metal layer, where

$$q_n''' = q_0''' e^{i\omega t}. \quad (7)$$

This set of equations has solutions in the form below, where T_n indicates magnitude and phase of temperature oscillations,

$$T_n(r, t) = e^{i\omega t} \left(C_{n,I} I_0(\gamma_n r) + C_{n,K} K_0(\gamma_n r) + \frac{\alpha_n q_n'''}{i\omega k_k} \right), \quad (8)$$

$$\gamma_n^2 = \frac{i\omega}{\alpha_n}, \quad (9)$$

and boundary conditions at each interface are

$$-k_n \frac{\partial T_n}{\partial r} \Big|_{r=r_n} = -k_{n+1} \frac{\partial T_{n+1}}{\partial r} \Big|_{r=r_n}, \quad (10)$$

$$-k_n \frac{\partial T_n}{\partial r} \Big|_{r=r_n} = h_n (T_n(r_n) - T_{n+1}(r_n)), \quad (11)$$

where h_n is the thermal boundary conductance.

Symmetry at $r = 0$, leads to no heat conduction through the center,

$$\left. \frac{\partial T_1}{\partial r} \right|_{r \rightarrow 0} = 0. \quad (12)$$

This condition arises because as $r \rightarrow 0$ volume goes to 0, so if any heat was entering or exiting, there would be a non-physical infinite temperature reached.

At $r \rightarrow \infty$, $T_m(r \rightarrow \infty) = 0$.

This condition forces the outermost layer's temperature oscillation magnitude to approach zero as radius goes to infinity.

The coefficients preceding the modified Bessel functions in each layer can be solved by formulating the boundary conditions in terms of a set of linear equations. The first layer has

only a coefficient C_{1l} , as C_{1K} is zero due to the $r \rightarrow 0$ boundary condition. Likewise, the last layer has only the coefficient C_{LastK} because C_{Lastl} is zero from the $r \rightarrow \infty$ boundary condition. All other layers have two coefficients and therefore a matrix $[A]$ will be defined. $[A]$ is a square matrix with size $2l \times 2l$, where l is the number of interfaces. Likewise, the $\bar{x}$ will have one column and $2l$ rows,

$$\bar{x} = [C_{1l} \ C_{2l} \ C_{2K} \ \dots \ C_{(Last-1)l} \ C_{(Last-1)K} \ C_{LastK}]^T, \quad (13)$$

$$[A] = \begin{bmatrix} A_{1,1} & A_{1,2} & \dots & A_{1,l*2} \\ A_{2,1} & A_{2,2} & \dots & \\ \vdots & & \ddots & \\ A_{l*2,1} & & & A_{l*2,l*2} \end{bmatrix}. \quad (14)$$

For interface, $l = 1$:

$$A_{1,1} = -k_1 \gamma_1 I_1(\gamma_1 r_1),$$

$$A_{1,2} = k_2 \gamma_2 I_1(\gamma_2 r_1),$$

$$A_{1,3} = -k_2 \gamma_2 K_1(\gamma_2 r_1),$$

$$A_{2,1} = h_1 I_0(\gamma_1 r_1) + k_1 \gamma_1 I_1(\gamma_1 r_1),$$

$$A_{2,2} = -h_1 I_0(\gamma_2 r_1),$$

$$A_{2,3} = -h_1 K_0(\gamma_2 r_1),$$

$$b_{2,1} = i h_1 (\alpha_2 q_2 / k_2 - \alpha_1 q_1 / k_1) / \omega$$

For interfaces, $l \geq 2, l \neq last$:

$$A_{l*2-1,l*2-2} = -k_l \gamma_l I_1(\gamma_l r_l),$$

$$A_{l*2-1,l*2-1} = k_l \gamma_l K_1(\gamma_l r_l),$$

$$A_{l*2-1,l*2} = k_{l+1} \gamma_{l+1} I_1(\gamma_{l+1} r_l),$$

$$A_{l*2-1,l*2+1} = -k_{l+1} \gamma_{l+1} K_1(\gamma_{l+1} r_l)$$

$$A_{l*2,l*2-2} = h_l I_0(\gamma_l r_l) + k_l \gamma_l I_1(\gamma_l r_l),$$

$$A_{l*2,l*2-1} = h_l K_0(\gamma_l r_l) - k_l \gamma_l K_1(\gamma_l r_l),$$

$$A_{l*2,l*2} = -h_l I_0(\gamma_{l+1} r_l),$$

$$A_{l*2,l*2+1} = -h_l K_0(\gamma_{l+1} r_l),$$

$$b_{l*2,1} = i h_l (\alpha_{l+1} q_{l+1} / k_{l+1} - \alpha_l q_l / k_l) / \omega$$

For interface, $l = last$:

$$A_{l*2-1,l} = -k_l \gamma_l I_1(\gamma_l r_l),$$

$$A_{l*2-1,l+1} = k_l \gamma_l K_1(\gamma_l r_l),$$

$$A_{l*2-1,l+2} = -k_{l+1} \gamma_{l+1} K_1(\gamma_{l+1} r_l),$$

$$A_{l*2,l} = h_l I_0(\gamma_l r_l) + k_l \gamma_l I_1(\gamma_l r_l),$$

$$A_{l*2,l+1} = h_l K_0(\gamma_l r_l) - k_l \gamma_l K_1(\gamma_l r_l),$$

$$A_{l*2,l+2} = -h_l K_0(\gamma_{l+1} r_l),$$

$$b_{l*2,1} = i h_l (-\alpha_l q_l / k_l) / \omega$$

(15)

$$[A] \bar{x} = b \Rightarrow \bar{x} = [A]^{-1} b. \quad (16)$$

By inverting $[A]$ and solving for the x -vector, the constants for each layer can be solved. These constants can then be applied to the previous general solution for temperature in a layer. The amplitude and phase averaged over the thickness of the metal heating layer were fit to data. Fits were optimized by varying the thermal conductivity of the unknown fluid – the value resulting in the least-squares fit with data is the reported value of thermal conductivity. In a similar manner thermal boundary conductance or specific heat, rather than thermal conductivity, can be solved with least-squares minimization. Like all 3ω methods, fitting to more than one variable at the same time increases the uncertainty in both predictions.

III. RESULTS AND DISCUSSION

To validate the metal-coated 3ω technique benchmark fluids having thermal conductivities spanning nearly 2 orders of magnitude were tested. Current frequencies from 10–500 Hz were used creating L_P on the order of 1.5 mm to 0.2 mm for air at standard temperature and pressure. While higher frequencies could be accessed, this range still enabled

measurement in minutes, rather than over a 24 h period as required by the solid-wire experiment.⁹ The benchmarks include argon, air, helium, and deionized water. Figure 5 compares the normalized ΔT and phase lag between fluids of various thermal conductivities. Normalization of temperature oscillation magnitude was performed by dividing the raw $\Delta V_{3\omega}$ signal at all frequencies by the maximum $\Delta V_{3\omega}$ occurring at the lowest measured frequency. The shape of the normalized amplitude plot depends on thermal conductivity and monotonically decreases for all materials. The phase signal, which is unaffected by normalization, changes more dramatically between the different gases. Note that while in magnitude argon and helium lie very close to each other, in phase they are distinguishable. By using normalized amplitude and phase we avoid the need for direct knowledge of β , which affects only the absolute magnitude of ΔT . MATLAB's *nlinfit* function was used to determine the thermal conductivity of the fluid that minimized the square error between the experimental data and analytical solution.

The generalized solution presented earlier was used to fit experimental data (normalized amplitude and phase) which yielded thermal conductivities in agreement with Refs. 20 and 25. These values are listed in Table I with associated uncertainties. The dimensions and properties of our metal coated

FIG. 5. (Color online) Normalized amplitude and phase versus frequency for various fluids (Argon – dark blue cross, Air – light blue triangle, Helium – red square, Deionized H₂O – green circles) with corresponding analytical fits shown as similarly colored lines. The experimental and accepted thermal conductivities are shown in Table I. The data was normalized by dividing a given fluid's ΔT s by its maximum ΔT . This normalization is optional, but removes the need for measuring the heater's coefficient of thermoresistance. A least-squares minimization was then used to fit experimental data to analytical model.

fiber, which were assumed in the analytical model, are shown in Table II. This model includes four layers ($n = 4$) to account for the leaded glass core, the borosilicate cladding, the metal coating, and the surrounding sample. Uncertainties were calculated by propagating the uncertainties in parameters input to the analytical solution. Their impacts on the uncertainty were combined orthogonally ($\Delta k = \sqrt{\sum_i \Delta k_i^2}$, where Δk_i is the uncertainty in k due to parameter i). We find that this solution is most sensitive to fiber diameter.

Using the analytical models validated by our benchmark of metal-coated 3ω , anticipated ΔT amplitude and phase were calculated for a range of thermal boundary conductances. In the metal-coated 3ω , the phase shape versus frequency is significantly affected by thermal boundary conductance (Fig. 6). In this figure, the change in phase for a decade of thermal boundary conductance (between 10^7 and 10^8 W/m² K) is plotted for metal-coated and solid-wire 3ω , surrounded by liquid Hg. This change in phase is greater for metal-coated 3ω . Changing the probe-Hg thermal boundary resistance from 10 MW/m² K to 100 MW/m² K results in an average phase difference of about 2° with metal-coated 3ω , and only 0.7° with solid-wire 3ω . The sensitivity to thermal boundary conductance shown here is enhanced by using

a high thermal conductivity fluid and sensing at higher frequencies. The validation samples and frequency ranges were insensitive to thermal boundary conductance.

IV. CONCLUSIONS

A modified 3ω technique that employs a suspended glass fiber coated with metal, used as a heater/thermometer, has been developed. This technique was successfully benchmarked against liquid and gas samples spanning 2-orders of magnitude in thermal conductivity. The benefits of this technique are larger signal strength for a given ΔT , less parasitic conduction loss, no need for absolute calibration of ΔT , rapid measurement, and enhanced sensitivity to thermal boundary

TABLE I. Comparison of accepted thermal conductivity values (Refs. 20 and 25) to experimental values obtained with this experiment on benchmark fluids. Experiments performed at atmospheric pressure at 300 K.

	$k_{\text{Experimental}}$ (W/m K)	$\pm k\%$	Δk (W/m K)	k_{Accepted} (W/m K)
Deionized H ₂ O	0.610	4.19%	0.026	0.593
He	0.163	5.04%	0.008	0.156
Air	0.027	5.17%	0.001	0.026
Argon	0.018	8.61%	0.002	0.017

FIG. 6. (Color online) A simulated plot of change in phase between thermal boundary conductance of 10^7 and 10^8 W/m² K in metal-coated 3ω (solid green) and solid metal 3ω (dashed red). The greater separation between metal-coated 3ω curves indicates greater sensitivity to thermal boundary conductance than solid metal 3ω . Simulated experiment has heater surrounded by mercury with a thermal boundary conductance across metal-mercury interface.

TABLE II. Properties and dimensions of experimental setup assumed for the predictions of thermal conductivity shown in Table I. Accepted values of ρ and c were used for each fluid sample (Refs. 20 and 25).

	Layer $n = 1$ (Schott F2 ^a leaded glass)	Layer $n = 2$ (Schott 8250 ^a borosilicate)	Layer $n = 3$ (Platinum)	Layer $n = 4$ (Sample)
r_n (μm)	12	13.1	13.15	...
k_n (W/m K)	0.78	1.18	71.6	Table I
ρ_n (kg/m^3)	3600	2280	21450	See Refs. 20 and 25
c_n (J/kg K)	557	720	130	See Refs. 20 and 25

^aProperties and dimensions were supplied by the manufacturer.

conductance. Relative to existing 3ω approaches for measuring fluids, this means measurement times are reduced from hours to minutes, sample sizes are reduced from tens-of-centimeters to millimeters, and low thermal effusivity materials are measurable where they have not been before.

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¹L. Lu, W. Yi and D. L. Zhang, *Rev Sci Instrum* **72**(7), 2996 (2001).²L. Shi, D. Y. Li, C. H. Yu, W. Y. Jang, D. Kim, Z. Yao, P. Kim, and A. Majumdar, *ASME Trans. J. Heat Transfer* **125**(5), 881 (2003).³F. Chen, J. Shulman, Y. Xue, C. W. Chu, and G. S. Nolas, *Rev. Sci. Instrum.* **75**(11), 4578 (2004).⁴S. R. Choi, J. Kim, and D. Kim, *Rev. Sci. Instrum.* **78**(8), 084902 (2007).⁵Z. L. Wang, D. W. Tang, and W. G. Zhang, *J. Phys. D: Appl. Phys.* **40**(15), 4686 (2007).⁶H. C. Chien, D. J. Yao, M. J. Huang, and T. Y. Chang, *Rev. Sci. Instrum.* **79**(5), 054902 (2008).⁷S. R. Choi and D. Kim, *Rev. Sci. Instrum.* **79**(6), 064901 (2008).⁸J. Buongiorno, D. C. Venerus, N. Prabhat, T. McKrell, J. Townsend, R. Christianson, Y. V. Tolmachev, P. Keblinski, L. W. Hu, J. L. Alvarado, I. C. Bang, S. W. Bishnoi, M. Bonetti, F. Botz, A. Cecere, Y. Chang, G. Chen, H. S. Chen, S. J. Chung, M. K. Chyu, S. K. Das, R. Di Paola, Y. L. Ding, F. Dubois, G. Dzido, J. Eapen, W. Escher, D. Funfschilling, Q. Galand, J. W. Gao, P. E. Gharagozloo, K. E. Goodson, J. G. Gutierrez, H. P. Hong, M. Horton, K. S. Hwang, C. S. Iorio, S. P. Jang, A. B. Jarzebski, Y. R. Jiang,L. W. Jin, S. Kabelac, A. Kamath, M. A. Kedzierski, L. G. Kieng, C. Kim, J. H. Kim, S. Kim, S. H. Lee, K. C. Leong, I. Manna, B. Michel, R. Ni, H. E. Patel, J. Philip, D. Poulidakos, C. Reynaud, R. Savino, P. K. Singh, P. X. Song, T. Sundararajan, E. Timofeeva, T. Triticak, A. N. Turanov, S. Van Vaerenbergh, D. S. Wen, S. Witharana, C. Yang, W. H. Yeh, X. Z. Zhao, and S. Q. Zhou, *J. Appl. Phys.* **106**(9), 094312 (2009).⁹S. M. Lee, *Rev. Sci. Instrum.* **80**(2), 024901 (2009).¹⁰E. Yusibani, P. L. Woodfield, M. Fujii, K. Shinzato, X. Zhang, and Y. Takata, *Int. J. Thermophys.* **30**(2), 397 (2009).¹¹E. Pop, S. Sinha, and K. E. Goodson, *Proc. IEEE* **94**(8), 1587 (2006).¹²E. S. Landry and A. J. H. McGaughey, *Phys. Rev. B* **79**(7), 075316 (2009).¹³R. Venkatasubramanian, *Phys. Rev. B* **61**(4), 3091 (2000).¹⁴M. S. Dresselhaus, G. Chen, M. Y. Tang, R. G. Yang, H. Lee, D. Z. Wang, Z. F. Ren, J. P. Fleurial, and P. Gogna, *Adv. Mater.* **19**(8), 1043 (2007).¹⁵Y. Rabin, *CryoLetters* **21**(3), 163 (2000).¹⁶A. Schmidt, M. Chiesa, X. Y. Chen, and G. Chen, *Rev. Sci. Instrum.* **79**(6), 064902 (2008).¹⁷B. Klaudinyi, E. A. Snajdr, and J. R. Friedrichs, *Am. Ceram. Soc. Bull.* **55**(8), 739 (1976).¹⁸E. V. Somers and J. A. Cyphers, *Rev. Sci. Instrum.* **22**(8), 583 (1951).¹⁹K. D. Maglic, A. Cezairliyan, and V. E. Peletsky, *Compendium of Thermophysical Property Measurement Methods* (Plenum, New York, 1984).²⁰D. R. A. Perkins, NIST Experimental Property of Fluids Group, Vol. 2010.²¹J. J. Healy, J. J. Degroot, and J. Kestin, *Physica B & C* **82**(2), 392 (1976).²²Y. Nagasaka and A. Nagashima, *Rev. Sci. Instrum.* **52**(2), 229 (1981).²³D. G. Cahill, *Rev. Sci. Instrum.* **61**(2), 802 (1990).²⁴C. Dames and G. Chen, *Rev Sci Instrum* **76**(12), 124902 (2005).²⁵A. F. Mills, *Heat transfer*, 2nd ed. (Prentice Hall, Upper Saddle River, NJ, 1999).²⁶R. Heyd, A. Hadaoui, M. Fliyou, A. Koumina, L. E. Ameziene, A. Outzourhit, and M. L. Sabounji, *Rev. Sci. Instrum.* **81**(4), 044901 (2010).²⁷H. S. Carslaw and J. C. Jaeger, *Conduction of heat in solids*, 2nd ed. (Clarendon, Oxford, 1959).