Generalized “Slope Method” of the 3ω Analysis to Measure the Thermal Conductivity and Heat Capacity of Solids: Frequency- vs. Current-sweep

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Abstract

Due to its simplicity and accuracy, the “slope method” is commonly used to measure the thermal conductivity, k , of solids in the traditional 3ω analysis, in which the slope of the linear relation between the 3ω voltage, $V_{3\omega}$, and the logarithm of the angular frequency, $\ln(\omega)$, is inversely proportional to k in the low-frequency limit. Here, we generalize this “slope method” to the high-frequency limit to extract the effusivity, e ($= \sqrt{kC}$, where C is the heat capacity). Moreover, we propose a current-sweep scheme, in which the slope of the $V_{3\omega}$ vs. $I_{1\omega}^3$ relation is used to extract both k and C . This current-sweep scheme is more reliable when k and/or C are frequency-dependent or the qualified frequency range is so narrow that discretization artifacts may be introduced to the frequency-sweep scheme. This generalized “slope method” is validated using control experiments on a glass substrate in the temperature range of 78 – 300 K. A two-heater scheme is proposed to measure the frequency-dependent k and C .

Keywords: Thermal conductivity; Heat capacity; 3ω method; Frequency-sweep; Current-sweep

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1. Introduction

Accurately measuring the thermal properties, e.g. the thermal conductivity, k , and the heat capacity, C , of solids is crucial for understanding the transport behavior of the fundamental energy carriers, e.g. electrons and phonons, and for helping the thermal design of the integrated circuit (IC) technology.^[1] The 3ω method is a widely used electrothermal technique for the measurement of k ^[2-7] and C ^[8-15] due to several merits including its relatively simple microfabrication process, relatively fast operation time, and its immunity from convection^[16] and radiation.^[2] Although this technique can be applied to various scenarios, including isotropic^[2] and anisotropic^[4] bulk samples, thin films along both in-plane^[4] and cross-plane^[7] directions, thermal boundary resistance,^[17] suspended wires,^[5,8-9,15] liquids,^[10-12,18] and biological tissues,^[6] here we constrain our scope to isotropic bulk samples.

The traditional 3ω method^[2] drives a line heater microfabricated on a substrate by a sinusoidal current, $I_{1\omega}$, at angular frequency ω , causing a temperature response and a corresponding fluctuation of the heater resistance, both at 2ω . Finally, the product of this 2ω resistance fluctuation and the original 1ω driving current leads to a voltage at 3ω , $V_{3\omega}$, which contains information of thermal properties of the sample. A “slope method” is usually implemented to measure k in the 2D cylindrical heating regime, in which the slope of a linear fit to the $V_{3\omega}$ vs. $\ln(\omega)$ relation^[2] is inversely proportional to k . However, this “slope method” is constrained by various assumptions of the heat transfer model, which may limit the qualified frequency range to be very narrow. Under this circumstance, the discretization of the qualified narrow frequency range has to be made finer to ensure enough data points for an accurate linear fit to the $V_{3\omega}$ vs. $\ln(\omega)$ relation, which may introduce discretization artifacts^[16] due to inadequate least significant bit (LSB) of the lock-in amplifier, and thus reduce the accuracy of the measurements.

With the measured k above, one can fit the experimental data to a nonlinear $V_{3\omega}$ vs. ω relation^[13-14] in the same or higher frequency regime to extract C . However, the nonlinear fit is not as simple and convenient as the linear fit above. More

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Fig. 1 Schematics of the cross-sectional view of an infinite long heater (into the page), with half width of b , periodically heating a semi-infinite sample. By varying the heating frequency, ω_H , one can dynamically change the penetration depth, $L_p = \sqrt{D/\omega_H}$, from $L_p \gg b$ (low-frequency limit), which corresponds to (a) 2D cylindrical heating, to $L_p \ll b$ (high-frequency limit), corresponding to (b) 1D planar heating. While the former is sensitive to k , the latter is sensitive to $\sqrt{kC}$. The red dashed lines and the yellow solids arrows represent the isotherms and adiabats, respectively.

importantly, in some cases, such as the polycrystalline diamond with the grain size increasing vertically from the growth interface,^[19] or subcooled liquids during a glass phase transition,^[10] k or C is frequency-dependent, making the self-consistency of this frequency-sweep scheme questionable.

Here we generalize the “slope method” to the 1D planar heating regime, in which, analogous to the linear fit to the $V_{3\omega}$ vs. $\ln(\omega)$ relation in the 2D cylindrical heating regime, we linearly fit the $V_{3\omega}$ vs. $\omega^{-1/2}$ relation whose slope gives the effusivity, $e (= \sqrt{kC})$. Moreover, to address the challenges of the frequency-sweep scheme, we further extend this “slope method” to a current-sweep scheme, in which the slope of the linear fit to the $V_{3\omega}$ vs. $I_{1\omega}^3$ relation can also be used to extract k and C . We first develop this current-sweep scheme, in parallel to the popular frequency-sweep scheme, under the framework of the thermal and electrical transfer functions.^[5] Next, we validate this new scheme by simultaneously extracting k and C of a glass sample in the temperature range of 78-300 K. Then we discuss the uncertainty and sensitivity, and the relative merits of the frequency- vs. current-sweep schemes. At last, we show the invariance of the form of the solution to the diffusion equation regardless of the frequency-dependent thermal properties, and propose a two-heater scheme to measure the frequency-dependent k and C .

II. Experimental method

A. 2D cylindrical vs. 1D planar periodical-heating

To dynamically switch the heat transfer pattern between the 2D cylindrical and the 1D planar periodical-heating (Fig. 1; assuming infinitely long heater into the page), the central concept is the thermal penetration depth^[16]

$$L_p = \sqrt{\frac{D}{\omega_H}} \quad (1)$$

where $D (= k/C)$ is the thermal diffusivity. The angular

heating frequency $\omega_H = 2\omega = 4\pi f$, because the Joule heating is at twice the driving current. L_p is a characteristic distance that the heat travels under the periodical modulation, which, by the assumption of a semi-infinite sample, should satisfy the condition of $L_p \ll d$, where d is the thickness of the sample. We note that different conventions exist in defining L_p , which may differ by some constant numerical factors.^[20]

Equation 1 suggests that by varying f , one can manipulate L_p , with respect to the heater halfwidth, b (Fig. 1), and thus dynamically control the heat transfer pattern. The low-frequency limit ($L_p \gg b$) leads to a 2D cylindrical periodical heating (Fig. 1a), i.e., the scenario that the traditional 3ω method assumes.^[2] Correspondingly, the thermal transfer function, a concept of the generalized thermal resistance^[5] is

$$Z_{low-freq \rightarrow cylindrical}(\omega_H) = \frac{1}{\pi l k} \left[-\frac{1}{2} \ln(\omega_H) + \frac{1}{2} \ln\left(\frac{k}{b^2 C}\right) + \eta - \frac{\pi}{4} i \right] \quad (2)$$

where the constant^[21] $\eta = 0.923$, and l is the length of the heater between V^+ and V^- probes (Fig. 2a), which, by the assumption of an infinitely long heater, should satisfy the conditions of $l \gg L_p$ and $l \gg b$.

On the other hand, the high-frequency limit ($L_p \ll b$) leads to a 1D planar periodical heating (Fig. 1b), which has a thermal transfer function^[12,22]

$$Z_{high-freq \rightarrow planar}(\omega_H) = \frac{1}{\sqrt{2kCA\sqrt{\omega_H}}} (1 - i) \quad (3)$$

where $A (= 2bl)$ is the area of the heater line between V^+ and V^- probes (Fig. 2a).

B. Bridge thermal and electrical quantities

To map the thermal transfer functions to measurable quantities, e.g. the currents and voltages, we follow the key result of Ref.

Fig. 2 Sample preparation and experimental setup. (a) Top view of a microscope image of the microfabricated heater, with the length and width of 2 mm (between the V^+ and V^- probes) and $80 \mu\text{m}$, respectively. Note here we employ laser-lithography to pattern the heater, which does not need the photomask. (b) Home-made sample-holder, which is pressed to the copper stage of the cryostat through the two holes to reduce the contact resistance. (c) Experimental setup², driven by a current source (Keithley 6221). The large 1ω voltage through the heater is cancelled using a standard resistor with low temperature coefficient and a multiplying digital-to-analog converter controlled by a single chip micryoco (STM32). The 3ω voltage is detected using a digital lock-in amplifier (SR850). Measurements are conducted in the temperature range of 78 – 300 K

5 and focus on the 3rd harmonic voltage and the corresponding thermal transfer function:

$$\frac{V_{3\omega,rms}}{2\alpha R_{e,0}^2 I_{1\omega,rms}^3} = -\frac{1}{4} Z_i(\omega_H) \quad (4)$$

where the thermal transfer function Z_i is obtained from either Eq. 2 or Eq. 3 depending on the ratio of L_p/b . $V_{3\omega,rms}$ and $I_{1\omega,rms}$ are the root mean square (rms) 3ω voltage and 1ω current, respectively, which are usually measured by lock-in amplifiers, a key equipment in the experiment. $R_{e,0}$ is the electrical resistance of the heater between V^+ and V^- probes (Fig. 2a) in the case of negligible Joule heating, and $\alpha (= \frac{1}{R_{e,0}} \frac{dR_e}{dT})$ is the temperature coefficient of the resistance (TCR) of the heater.

Since the in-phase voltage gives more accurate results than the out-of-phase voltage,^[2,5] in the following, we will focus on the in-phase components (with a subscript “x”), which corresponds to the real part of the thermal transfer function.

C. Frequency-sweep scheme

To extract thermal properties, a frequency-sweep scheme is usually implemented. Although nonlinear fits of experimental data to the full solution of the heat transfer model give both k and C , the “slope method” is commonly used due to its simplicity and accuracy. We first review the traditional “slope method”, which extracts k in the 2D cylindrical heating regime (low-frequency limit), and then we extend this “slope method” to the 1D planar heating regime (high-frequency limit) to obtain the effusivity ($e = \sqrt{kC}$).

In the 2D cylindrical heating regime (low-frequency limit: $L_p \gg b$), substituting Eq. 2 into Eq. 4, one obtains

$$k = \frac{1}{4\pi l} \frac{I_{1\omega,rms}^3 R_{e,0}}{\frac{\partial V_{3\omega,rms,x}}{\partial(\ln f)}} \frac{dR_e}{dT} \quad (5)$$

where $V_{3\omega,rms,x}$ is the in-phase rms 3ω voltage. Since the framework assumes a current source to drive the experiment, here $I_{1\omega,rms,x} = I_{1\omega,rms}$, and thus we neglect the subscript “x” for the driving current for clarity. Equation 5 is the traditional “slope method”, which suggests that one can obtain k from the slope, $\partial V_{3\omega,rms,x} / \partial(\ln f)$, of a linear fit to the $V_{3\omega,rms,x}$ vs. $\ln(f)$ relation, hence the name “slope method”. To ensure the linearity of the $V_{3\omega,rms,x}$ vs. $\ln(f)$ relation, the assumptions of the heat transfer model (Eq. 2) have to be satisfied, which requires

$$L_p \ll d, \text{ and } l \gg L_p \gg b \quad (6)$$

We now extend this “slope method” to the 1D planar heating regime (high-frequency limit: $L_p \ll b$). Substituting Eq. 3 into Eq. 4, one obtains

$$\sqrt{kC} = -\frac{1}{4A\sqrt{2\pi}} \frac{I_{1\omega,rms}^3 R_{e,0}}{\frac{\partial V_{3\omega,rms,x}}{\partial(f^{-1/2})}} \frac{dR_e}{dT} \quad (7)$$

Equation 7 suggests that one can obtain the effusivity ($e = \sqrt{kC}$) from the slope of a linear fit to $V_{3\omega,rms,x}$ vs. $f^{-1/2}$ relation. Combining with the measured k above, one can further back out C . Likewise, to ensure the linearity of the $V_{3\omega,rms,x}$ vs. $f^{-1/2}$ relation and thus facilitate the data processing, the assumptions of the heat transfer model (Eq. 3) have to be satisfied, which requires

$$L_p \ll \min(b, l, d) \quad (8)$$

D. Current-sweep scheme

From Eqs. 6 and 8, one immediately sees that these requirements limit the qualified frequencies to some range. Due to the constraints from the minimum linewidth of the photomask or shadow mask, and/or the thickness of the sample, sometimes this qualified frequency range could be

very narrow, in which circumstance the discretization of the frequency range has to be made finer to ensure enough data points for an accurate linear fit. This, in turn, may introduce discretization artifacts due to inadequate least significant bit (LSB) of the lock-in amplifier, and thus reduce the accuracy of the measurements. Moreover, sometimes C [10] or k [19] is frequency-dependent, ruining the self-consistency of the frequency-sweep scheme.

To address these shortcomings of the frequency-sweep scheme, we develop a current-sweep scheme. This scheme was used as a sanity check to ensure the voltage response from the heater is free from significant artifacts from capacitive coupling or AC offsets. [5,16] Here, we show that k and C can be extracted from this current-sweep scheme.

In parallel with the derivations in the frequency-sweep scheme, from Eqs. 2 and 4 one obtains

$$k = -\frac{1}{2\pi l} \frac{dR_e}{dT} \frac{R_{e,0}}{\frac{\partial V_{3\omega,rms,x}}{\partial (I_{1\omega,rms}^3)}} \left[-\frac{1}{2} \ln(\omega_H) + \frac{1}{2} \ln\left(\frac{k}{b^2 C}\right) + \eta \right] \quad (9)$$

in the low-frequency limit ($L_p \gg b$); from Eqs. 3 and 4, one obtains

$$\sqrt{kC} = -\frac{1}{4A\sqrt{2\pi f}} \frac{R_{e,0}}{\frac{\partial V_{3\omega,rms,x}}{\partial (I_{1\omega,rms}^3)}} \frac{dR_e}{dT} \quad (10)$$

in the high-frequency limit ($L_p \ll b$). Equations 9 and 10 suggest that one can extract both k and C from the slope of a linear fit to the $V_{3\omega,rms,x}$ vs. $I_{1\omega,rms}^3$ relation in the low- and high-frequency limit, respectively. The key advantage of this current-sweep scheme is that, instead of a frequency range, one only requires a frequency point, which avoids the potential discretization artifacts in the frequency-sweep scheme. More importantly, this current-sweep scheme can measure frequency-dependent k and / or C (see III.D and III.E).

E. Sample preparation and experimental setup

We microfabricate a 100 nm-thick gold heater (with 5 nm-thick nickel as an adhesion layer) on top of a 1 mm thick glass sample. The width ($2b$) and the length (between V^+ and V^- in Fig. 2a) of the heater are 80 μm and 2 mm, respectively. Here we use the laser-lithography (Microwriter ML3), instead of the traditional photolithography, to pattern the heater. The major merit of the former is that it directly transfers the heater pattern to the photoresist without the use of a photomask. Standard e-beam evaporation and lift-off process are employed to microfabricate the heater according to the pattern by the laser-lithography.

The 1 cm $\times$ 1 cm sample is mounted on a homemade gold-coated copper sample-holder (Fig. 2b) and secured to the copper stage of our cryostat (C-Mag/Vari-9T MGHS-GM), which is evacuated to $\sim 10^{-6}$ Torr and operated in the temperature range of 78 to 300 K. To improve the thermal contact, we apply several atmospheres of clamping pressure [23-24] between the sample-holder and copper stage.

The measurements are driven by a current source (Keithley 6221; Fig. 2c), and the 1ω and 3ω voltages are probed by a

lock-in amplifier (SR850). To remove the large 1ω voltage background and facilitate the extraction of the small 3ω voltage, we apply the standard cancellation scheme,² which is automated by a single chip micryoco (STM32). The $R_{e,0}$ vs. T relation of the microfabricated heater is first calibrated, and the resulting dR_e/dT is a key input parameter to extract k and C (Eqs. 5, 7, 9, and 10). This dR_e/dT is one of the primary sources of uncertainty in the final measured k and C values. To avoid excessive Joule heating, we sweep current from 100 μA to 1 mA, and the slope of the V - I curve gives $R_{e,0}$.

III. Results and discussion

A. Validation of method

Fig. 3 shows the scaling check of the generalized “slope method” at $T = 300$ K for the 2D cylindrical heating ($L_p \gg b$; 1st column) and the 1D planar heating ($L_p \ll b$; 2nd column), using the frequency-sweep scheme (1st row) and the current-sweep scheme (2nd row), respectively. We first examine the frequency-sweep scheme. First, the 2D cylindrical heating (blue diamonds in Fig. 3a; left axis) clearly shows a linear relation between $V_{3\omega,rms,x}$ and $\ln(f)$, as expected from Eqs. 2 and 4. We fix the driving current at $I_{1\omega,rms} = 30$ mA, and choose a frequency range of 0.5 – 3.3 Hz, so that the minimum penetration depth (right end of the black solid line in Fig. 3a; right axis), $L_{p,min} \geq 3b$, while the maximum penetration depth (left end of the black solid line in Fig. 3a; right axis), $L_{p,max} \leq 0.3d$ and $L_{p,max} \leq 0.2l$, which are consistent with the assumptions of the heat transfer model (Eq. 6). The slope of this $V_{3\omega,rms,x}$ vs. $\ln(f)$ relation, together with other parameters, gives $k = 1.32$ W/m-K (Eq. 5). Second, the 1D planar heating (blue diamonds in Fig. 3b; left axis) shows a linear relation between $V_{3\omega,rms,x}$ and $f^{-1/2}$, as expected from Eqs. 3 and 4. Under this circumstance, we choose a frequency range of 330 – 2000 Hz, corresponding to the penetration depth, L_p , in the range of 12 - 5 μm (black solid line in Fig. 3b; right axis), which meets the requirements of the heat transfer model (Eq. 8). The slope of this $V_{3\omega,rms,x}$ vs. $f^{-1/2}$ relation, together with other parameters, gives $\sqrt{kC}$ (Eq. 7), which, combined with the measured k above (Fig. 3a; Eq. 5), leads to $C = 1.68 \times 10^6$ J/m³-K.

We next examine the current-sweep scheme. First, the 2D cylindrical heating (orange diamonds in Fig. 3c; left axis) confirms a linear $V_{3\omega,rms,x}$ vs. $I_{1\omega,rms}^3$ relation predicted by Eqs. 2 and 4. We fix the frequency of the driving current at $f = 1$ Hz, which results in a penetration depth of $L_p \approx 225$ μm , satisfying Eq. 6. We choose a current range of 10-30 mA, so that the corresponding temperature rise, ΔT (green solid line in Fig. 3c; right axis), which is calculated by multiplying Eq. 2 with the Joule heating, $Q_{2\omega} = I_{1\omega,rms}^2 R_e$, is in the range of 0.6-5.4 K. This ΔT is orders of magnitude smaller than the set point, $T = 300$ K, and thus avoid the ambiguity about the temperature of the measurement. The slope of the $V_{3\omega,rms,x}$ vs.

Fig. 3 Scaling check of the generalized “slope method” at 300 K in the low-frequency limit (left column; $L_p \gg b$) and high-frequency limit (right column; $L_p \ll b$) using the frequency sweep scheme (upper row) and the current sweep scheme (lower row). The schematics show the relation between the penetration depth, L_p ($= \sqrt{D/4\pi f}$), and the half width of the heater, b , which determines the heating dimensionality: 2D cylindrical heating (left column) vs. 1D planar heating (right column). The red solid lines represent the envelopes of the temperature profiles. The slope of (a) low-frequency sweep of the $V_{3\omega, rms, x}$ vs. $\ln(f)$ relation gives k ; (b) high-frequency sweep of the $V_{3\omega, rms, x}$ vs. $f^{-1/2}$ relation gives $\sqrt{kC}$; (c) current sweep (at $f = 1$ Hz) of the $V_{3\omega, rms, x}$ vs. $I_{1\omega, rms}^3$ relation gives k ; (d) current sweep (at $f = 1,000$ Hz) of the $V_{3\omega, rms, x}$ vs. $I_{1\omega, rms}^3$ relation gives $\sqrt{kC}$. The selected frequency ranges ensure that the relation between L_p (right axis of (a) and (b)) and b is consistent with the corresponding schematics, as described by Eq. 6 for the 2D cylindrical heating and Eq. 8 for the 1D planar heating. The selected current range ensures that ΔT (right axis of (c) and (d)) is larger than the measurement uncertainties but small enough to avoid any ambiguity about the temperature of the measurement.

$I_{1\omega, rms}^3$ relation sets one constraint to k and C (Eq. 9). To find a second constraint and thus solve k and C , we change the driving frequency to $f = 1000$ Hz, which results in a much shorter penetration depth of $L_p \approx 7 \mu\text{m}$, thus satisfying the assumptions of the 1D planar heating (Eq. 8). This 1D planar heating (orange diamonds in Fig. 3d; left axis) conforms a

linear relation between $V_{3\omega, rms, x}$ vs. $I_{1\omega, rms}^3$ predicted by Eqs. 3 and 4. In this case, ΔT (green solid line in Fig. 3d; right axis) is even smaller because of the much shorter L_p . The slope of the $V_{3\omega, rms, x}$ vs. $I_{1\omega, rms}^3$ relation, together with other parameters, gives $\sqrt{kC}$ (Eq. 10). Combining with the first constraint on k and C above (Fig. 3c; Eq. 9), one can extract

Fig. 4 Experimental measurements of (a) thermal conductivity and (b) heat capacity of a glass sample using the frequency-sweep scheme (red diamonds) and the current-sweep scheme (blue triangles), respectively, in the temperature range of 78 – 300 K. Also included for comparison is the literature results (grey solid lines) from Cahill.^[25] Our measurements agree with the literature results to within $\pm 7.4\%$ for k and $\pm 13.7\%$ for C in the entire temperature range, which is consistent with our sensitivity analysis. Note here for clarity we omit the error bars, since they are smaller than the size of the plotted points.

$k = 1.31$ W/m-K and $C = 1.76 \times 10^6$ J/m³-K. These values agree with the results of the frequency-sweep scheme to within 0.3% for k and 4.8% for C at 300 K.

Fig. 4 shows our measurement results of k and C from 78 K to 300 K, using the frequency-sweep scheme (blue triangles) and the current-sweep scheme (red diamonds), and the comparison with literature results of amorphous SiO₂ (grey solid lines).^[25] The three sets of results (triangles, diamonds, and solid lines) agree with each other to within 7.4% for k and 13.7% for C in the entire temperature range. The typical measurement uncertainty in k ranges from $\pm 1.7\%$ at 300 K to $\pm 3.5\%$ at 78 K (95% confidence interval, or 95% CI), and the uncertainty in C ranges from $\pm 3.9\%$ at 300 K to $\pm 6.7\%$ at 78 K (95% CI). This means, for example, we are 95% confident that the true k of the glass sample at 300 K is somewhere in the range [1.30, 1.34] W/m-K. For clarity, we neglect the error bars in Fig. 4, since they are smaller than the size of the plotted points. We see that the uncertainty in measuring C is larger than that in measuring k , which will be explained in the following in the context of the sensitivity analysis.

B. Sensitivity

A transient heat transfer process is usually sensitive to either the thermal diffusivity, $D = k/C$, or the thermal effusivity, $e = \sqrt{kC}$, due to the nature of the thermal diffusion equation.^[26] Within this context, the traditional 3ω method (periodical 2D cylindrical heating) is a special case, which, due to the property of the Bessel function, can isolate k in the frequency-sweep scheme (Eq. 5).^[16]

To rigorously quantify the physical argument above, we define the sensitivity of $|V_{3\omega, rms, x}|$ to a parameter of interest, p , as in Ref. 27^[27]

$$S_p^{|V_{3\omega, rms, x}|} = \frac{\partial \ln(|V_{3\omega, rms, x}|)}{\partial \ln(p)} \quad (11)$$

where $p = k$ or C . Note we use the absolute value of $V_{3\omega, rms, x}$, since it is always negative,^[16] as evident from Fig. 3. This definition of sensitivity means that if $S_k^{|V_{3\omega, rms, x}|} = -2$, for example, then a 10% increase in k would cause a 20% decrease in $|V_{3\omega, rms, x}|$.

Table 1. Summary of the sensitivity of the 3ω voltage to the thermal conductivity and the heat capacity, $S_k^{|V_{3\omega, rms, x}|}$ (1st column) and $S_C^{|V_{3\omega, rms, x}|}$ (2nd column), for the 2D cylindrical ($L_p \gg b$; 1st row) and 1D planar periodical heating ($L_p \ll b$; 2nd row). While the 1D planar periodical heating is equally sensitive to k and C with $S_k^{|V_{3\omega, rms, x}|} = S_C^{|V_{3\omega, rms, x}|} = -1/2$, the 2D cylindrical periodical heating, which the traditional 3ω method assumes, is more sensitive to k .

	$S_k^{ V_{3\omega, rms, x} }$	$S_C^{ V_{3\omega, rms, x} }$
2D cylindrical periodical-heating	$-1 + \frac{1}{2[\ln(\frac{L_p}{b}) + \eta]}$	$-\frac{1}{2[\ln(\frac{L_p}{b}) + \eta]}$
1D planar periodical-heating	$-\frac{1}{2}$	$-\frac{1}{2}$

Table I summarizes $S_k^{|V_{3\omega, rms, x}|}$ (1st column) and $S_C^{|V_{3\omega, rms, x}|}$ (2nd column) under the two circumstances, 2D cylindrical ($L_p \gg b$; 1st row) vs. 1D planar heating ($L_p \ll b$; 2nd row). First, consider the 2D cylindrical heating that the traditional

Fig. 5 Two-heater scheme to measure frequency-dependent k and C simultaneously. At the same heating frequency, ω_H , the penetration depths, L_p ($= \sqrt{D/\omega_H}$), of the two heaters are the same. The narrow heater (left) is in its low-frequency limit ($b_{\text{narrow}} \ll L_p$), while the wide heater is in its high-frequency limit ($b_{\text{wide}} \gg L_p$). One can obtain both k and C at this specific frequency by applying the current-sweep scheme, Eqs. 9 and 10, to the narrow heater and the wide heater, respectively. Repeating the same procedure at various frequencies, one obtains the frequency-dependent k and C .

3ω method assumes. Equations 2 and 4 lead to $S_k^{|V_{3\omega, rms, x}|} = -1 + \{2[\ln(L_p/b) + \eta]\}^{-1}$ and $S_C^{|V_{3\omega, rms, x}|} = -\{2[\ln(L_p/b) + \eta]\}^{-1}$, respectively. Assuming $L_p/b = 5$, we obtain a relatively high $S_k^{|V_{3\omega, rms, x}|} = -0.8$ and a relatively low $S_C^{|V_{3\omega, rms, x}|} = -0.2$. Second, consider the 1D planar heating. Equations 3 and 4 lead to $S_k^{|V_{3\omega, rms, x}|} = S_C^{|V_{3\omega, rms, x}|} = -1/2$. This sensitivity analysis shows that while the 2D cylindrical heating is much more sensitive to k , the 1D planar heating is equally sensitive to k and C , which is consistent with our physical insight above. It also explains why the uncertainty of C is larger than that of k , since we have to combine the 2D cylindrical heating and the 1D planar heating regimes to extract k and C , no matter which sweep scheme is employed.

C. Relative merits of the frequency- and current-sweep methods

Overall, the traditional frequency-sweep method is still the best option for most experiments, since it gives algebraic expressions (Eqs. 5 and 7) of k and C . In contrast, the current-sweep method has to solve two coupled nonlinear equations (Eqs. 9 and 10) to extract k and C .

However, the current-sweep method is beneficial in two situations: first, the frequency range that satisfies all the assumptions of the heat transfer model is very narrow. In this case, the narrow frequency range has to be discretized finer for more frequency points and thus better accuracy of the fitting of $\partial V_{3\omega, rms, x} / \partial(\ln f)$ in Eq. 5, or $\partial V_{3\omega, rms, x} / \partial(f^{-1/2})$ in Eq. 7. However, finer discretization may cause artifacts due to inadequate least significant bit (LSB) of the equipment, e.g. the lock-in amplifier.^[16] In contrast, the current-sweep method

requires only one frequency, which does not have the problem of discretization artifacts. Second, k and / or C are frequency-dependent. In this case, it is inconsistent to obtain a frequency-dependent property using a frequency-sweep method. Although clever tricks, for example a window fitting scheme used in the frequency domain thermal reflectance (FDTR)^[28] can be applied, a current-sweep scheme is much cleaner to measure frequency-dependent properties, as discussed in detail in the following.

D. Diffusion equation with frequency-dependent thermal properties

We show that the form of Eqs. 2 and 3 still hold for frequency-dependent thermal properties. We start from the general form of the diffusion equation:

$$\frac{1}{D(\omega_H)} \frac{\partial T}{\partial t} = \nabla^2 T \quad (12)$$

where the diffusivity, $D(\omega_H)$, now depends on the heating frequency, ω_H . This ω_H -dependent D results from the ω_H -dependent k and/or C , since $D = k/C$.

Despite of the ω_H dependence of D , Eq. 12 is still a linear partial differential equation with “constant” coefficients, since D does not depend on t, x, y, z , or T .^[29] Thus one can solve Eq. 12 using exactly the same approaches as those used in the frequency-independent scenarios, with the only difference of replacing the frequency-independent properties with the frequency-dependent ones in the final solution.

To illustrate this point, here we use the 2D cylindrical periodical heating problem as an example. We start from the infinitely-narrow heater, which applies a periodical flux boundary condition to the sample,

$$-k(\omega_H) \frac{\partial T(r, t)}{\partial r} \Big|_{r=0} = A = P_0 \exp(i\omega_H t) \quad (13a)$$

where $A (= \pi r l)$ is the surface area of the heater and P_0 is a constant. The second boundary condition is

$$T(r, t)|_{r \rightarrow \infty} = T_{\infty} \quad (13b)$$

Despite of the ω_H dependence of D , the solution still has the form of

$$T(r, t) = T_{\infty} + \exp(i\omega_H t) R(r) \quad (14)$$

According to the linear response theorem: an input signal at frequency ω_H results in an output signal at the same frequency ω_H . Thus, one can convert the governing equation (Eq. 12) and the boundary conditions (Eqs. 13a and 13b) to the following form:

$$\frac{\partial^2 R}{\partial r^2} + \frac{1}{r} \frac{\partial R}{\partial r} - i \frac{\omega_H}{D(\omega_H)} R = 0 \quad (15)$$

$$-k(\omega_H) \frac{\partial R(r)}{\partial r} \Big|_{r=0} A = P_0 \quad (16a)$$

$$R(r)|_{r \rightarrow \infty} = 0 \quad (16b)$$

From this point the solving process is exactly the same with that of the classic 3ω method (pg. 193 of Ref. 22) with the only minor difference of replacing the frequency-independent diffusivity, D , with the frequency-dependent $D(\omega_H)$ in defining the complex penetration depth: $1/q = \sqrt{D(\omega_H)/i\omega_H}$, which does not change the form of the solution, since Eq. 15 is still a zeroth-order modified Bessel equation (pg. 488 of Ref. 22). Therefore, after considering the finite width of the heater line, one finds the thermal transfer function to be exactly the same as Eq. 2, except to replace k with $k(\omega_H)$, and C with $C(\omega_H)$.

Likewise, the thermal transfer function of the 1D planar periodical heating problem with frequency-dependent thermal properties is exactly the same as Eq. 3, except to replace k with $k(\omega_H)$, and C with $C(\omega_H)$.

E. Proposed scheme to measure the frequency-dependent k and C

We introduce a two-heater scheme (Fig. 5) to extract the frequency-dependent k and C simultaneously. Driven at the same frequency, f , these two heaters heat up the sample with the same penetration depth, L_p , which is much longer than the half width of the narrower heater, b_{narrow} , but much shorter than the half width of the wider heater, b_{wide} . Thus, at this f , the narrow heater is in its low-frequency limit, while the wide heater is in its high-frequency limit. Using the current-sweep scheme (Eqs. 9 and 10), one can obtain the frequency-dependent k and C at this driving frequency, f . We highlight the difference between this two-heater scheme (Fig. 5) and the one-heater scheme (Fig. 1). While the former samples the low- and high- frequency limits (Eqs. 2 and 3) at one frequency, the latter requires two different frequencies.

Ideally one can apply this scheme to a wide frequency range. However, in practice this range is constrained by factors such as the size of the sample and the resolution of the lithography that fabricates the heater. As a sanity check, for the glass sample at 300 K with $b_{narrow} = 1\mu\text{m}$ and $b_{wide} = 1\text{mm}$, the corresponding maximum and minimum frequencies

are $f_{max} \approx 2,400$ Hz and $f_{min} \approx 1.5$ Hz, respectively, if we require $L_{p,min} \geq 5b_{narrow}$ and $L_{p,max} \leq b_{wide}/5$.

One can release the constraint on either bound of the frequency regime, $[f_{min}, f_{max}]$, if only one of the two properties, k or C , is frequency-dependent. As a concrete example, let's assume that k is frequency-dependent but C is not. First, one picks any frequency in the regime of $[f_{min}, f_{max}]$, and measures both k and C using the two heaters (Fig. 5) at this specific frequency. Next, one only needs either the narrow or the wide heater, correspondingly either Eq. 9 or Eq. 10, to measure the frequency-dependent k at different frequencies, since the frequency-independent C has already been measured in the first step. Correspondingly, the constraint, $L_p \ll b_{wide}$ or $L_p \gg b_{narrow}$, is removed, which, in turn, extends $f_{min} \rightarrow 0$ or $f_{max} \rightarrow \infty$. Note here f_{min} and f_{max} are calculated using Eq. 1 with the constraints of $L_p \ll b_{wide}$ and $L_p \gg b_{narrow}$, respectively, by the zeroth order approximation that assumes D is frequency-independent. For a frequency-dependent D , the correspondence between the $f_{extremum}$ and the constraints ($L_p \ll b_{wide}$ and $L_p \gg b_{narrow}$) may vary depending on the form of $D(f)$. We also note that other practical issues, such as the ultralong time constant required for the ultralow f_{min} (pg. 106 of Ref. 16), or the ultrasmall capacitive impedance of the electrodes at ultrahigh f_{max} , will eventually appear to set new bounds to the frequency regime.

IV. Summary

We have generalized the “slope method” of the traditional 3ω analysis from the low-frequency ($L_p \gg b$) regime to the high-frequency ($L_p \ll b$) regime. While the former is widely used to measure k , the latter offers an analogous way to measure $\sqrt{kC}$. Combining these low- and high-frequency sweeps, one can isolate k and C .

To complement the frequency-sweep scheme under circumstances of frequency-dependent k and/or C , or very narrow qualified frequency ranges which might introduce discretization artifacts due to the limitation of the equipment, we proposed an analogous current-sweep scheme, whose slope can also be used to extract k and C .

We have validated this generalized “slope method” by measuring the k and C of a glass sample using the frequency- vs. current-sweep scheme, respectively. The measured results agree with the literature values to within $\pm 7.4\%$ for k and $\pm 13.7\%$ for C in a wide temperature range of 78-300 K. We also analyzed the sensitivity of the two schemes to k and C in the low- and high-frequency limit, respectively.

We showed that the form of the solution to the diffusion equation with frequency-dependent thermal properties holds the same as that with frequency-independent thermal properties. We further proposed a two-heater scheme to extract the frequency-dependent k and C .

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Supporting information

Not applicable.

Conflict of interest

There are no conflicts to declare.

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