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Thermal conductivity measurement from 30 to 750 K: the 3ω method

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An ac technique for measuring the thermal conductivity of dielectric solids between 30 and 750 K is described. This technique, the 3ω method, can be applied to bulk amorphous solids and crystals as well as amorphous films tens of microns thick. Errors from black-body radiation are calculated to be less than 2% even at 1000 K. Data for a-SiO$_2$, Pyrex 7740, and Pyroceram 9606 are compared to results obtained by conventional techniques.

INTRODUCTION

The thermal conductivity of solids is usually determined by measuring the temperature gradient produced by a steady flow of heat in a one-dimensional geometry.[1,2] Below 50 K, where radiation of heat from the surface of the sample is small compared to the transport of heat through the solid, the steady-state technique is accurate and convenient. Problems arise, however, in extending the technique to room temperature and above, especially for materials that are poor conductors of heat; for example, amorphous solids. A large fraction of the heat that is intended to flow through the sample is radiated away. Radial[3] or pancake geometries for the heat flow and the use of heated shields[4] that maintain the same temperature gradient as the sample are proven ways of reducing the errors of black-body radiation. Unfortunately, these techniques often require large, precisely shaped samples and extreme care to be used successfully. These difficulties are demonstrated by the disagreement between data obtained in different laboratories on well-defined samples[5] (see below).

A recently developed ac thermal conductivity technique, the 3ω method, overcomes these difficulties. The method is insensitive to errors from black-body radiation because the effective thickness of the sample is extremely small, on the order of 100 μm, when compared to standard geometries where 1 cm is more typical. Errors due to black-body radiation can be shown to scale with a characteristic length of the experimental geometry by estimating the ratio of the heat radiated to the heat conducted through the sample.[6] For example, if the temperature difference used to measure the thermal conductivity is held constant while all dimensions of the experimental geometry are reduced by a factor f, the power radiated through black-body radiation is reduced by f^2 while the heat conducted by the sample is reduced by only f. Therefore, in this example, errors from radiation have been reduced by the factor f. In comparing the 3ω method to standard techniques, the errors are reduced by the ratio of 1 cm to 100 μm, a factor of 100. In this work, a calculation of errors from black-body radiation in the 3ω method and a demonstration of the method with data for thermal conductivity standards, Pyrex 7740 and Pyroceram 9606, and a-SiO$_2$ from 30 to 750 K are provided. A brief description of this technique and data for several amorphous solids have been published previously.[6] More examples of data taken with the 3ω method can be found in Refs. 7, 8, and 9.

I. SOLUTION OF THE DIFFUSION EQUATION

As in the closely related hot-wire[10–12] and hot-strip[13] techniques, the 3ω method uses a radial flow of heat from a single element that is used both as a heater and thermometer. The major difference is use of the frequency dependence of temperature oscillations instead of the time-domain response.

A side view of the experiment is shown in Fig. 1. I begin with the exact solution for the temperature oscillations a distance $r = (x^2 + y^2)^{1/2}$ from an infinitely narrow line source of heat on the surface of an infinite half-volume. The solution $\Delta T(r)$ is given by Carslaw and Jaeger[14]:

$$\Delta T(r) = (P/l\pi\Lambda)K_0(qr) , \qquad (1)$$

where Λ is the thermal conductivity of the infinite half-volume and P/l is the amplitude of the power per unit length generated at a frequency 2ω in the line source of heat. The factor of 2 is because a current at frequency ω produces joule heating at a frequency 2ω. K_0 is the zeroth-order modified Bessel function. The magnitude of the complex quantity $1/q$,

$$1/q = (D/i2\omega)^{1/2} \qquad (2)$$

is the wavelength of the diffusive thermal wave or what I call the thermal penetration depth. Here, D is the thermal diffusivity $\Lambda/\rho C$, where ρ is the density and C, the specific heat.
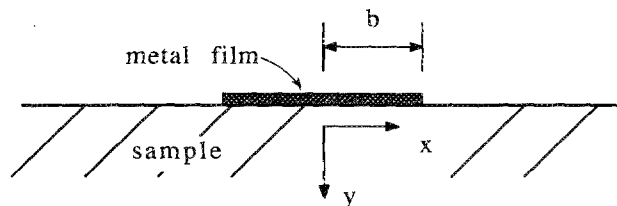

FIG. 1. Side view of the heater and sample geometry for the 3ω method. A metal line with half-width b is made from a metal film, 3000 Å thick, that is evaporated on the surface of the sample. An ac current at angular frequency ω heats the metal line at 2ω. The center of the metal line is the coordinate $x = 0$, $y = 0$ in Eqs. (1)–(11).

In the limit $|qr| \ll 1$, Eq. (1) can be approximated by

$$\Delta T = \frac{P}{\pi l \Lambda} \left(\frac{1}{2} \ln \frac{D}{r^2} + \ln 2 - 0.5772 \right.$$

$$\left. - \frac{1}{2} \ln (2\omega) - \frac{i\pi}{4} \right). \qquad (3)$$

Equation (3) has been written to separate the frequency-dependent and the imaginary contributions to the solution. Either the real or the imaginary part of the temperature oscillations ΔT can be used to determine the thermal conductivity. The imaginary part (out-of-phase oscillations) gives the thermal conductivity directly, but experimentally, I have found that the slope of the real part (in-phase oscillations) versus $\ln \omega$ is a more reliable measure.

To calculate the temperature oscillations from a heat source of finite width, it is convenient to take the Fourier transform[15] of Eq. (1) with respect to the x coordinate. In addition, only the temperature oscillations at the surface of the sample are important and I set $y = 0$;

$$\Delta T(k) = \int_0^\infty \Delta T(x) \cos(kx) \, dx , \qquad (4)$$

$$\Delta T(k) = (P/2l\Lambda)[1/(k^2 + q^2)^{1/2}] . \qquad (5)$$

The finite width of the heat source can now be included by multiplying Eq. (5) by the Fourier transform of the heat source as a function of the coordinate x; I assume that heat enters the sample evenly over the width of the line $|x| < b$. Convolution in the real space coordinate x has been replaced by multiplication in the Fourier space k:

$$\Delta T(k) = \frac{P}{2l\Lambda} \frac{\sin(kb)}{kb(k^2 + q^2)^{1/2}} , \qquad (6)$$

where b is the half-width of the heat source. Taking the inverse transform gives

$$\Delta T(x) = \frac{P}{l\pi\Lambda} \int_0^\infty \frac{\cos(kx)\sin(kb)}{kb(k^2 + q^2)^{1/2}} \, dk . \qquad (7)$$

Equation (7) describes the temperature oscillations on the surface a distance x from the center of a heat source of finite width $2b$. In the 3ω method, the thermometer used to measure these temperature oscillations is the same element as the heater and therefore measures some average temperature across the width of the line. Taking the average of Eq. (7) across the line width (by integrating with respect to x from 0 to b and dividing by b), gives the temperature oscillations ΔT measured by the thermometer:

$$\Delta T = \frac{P}{l\pi\Lambda} \int_0^\infty \frac{\sin^2(kb)}{(kb)^2(k^2 + q^2)^{1/2}} \, dk . \qquad (8)$$

Since the metal line undoubtedly affects the temperature profile slightly, Eq. (8) is not exact, but it is shown below that it works well in describing the experiment.

I am unaware of an analytic solution to Eq. (8), but can examine the limit when the thermal penetration depth is large compared to the line width, $1/q \gg b$. In this case, the integral is dominated by values of k such that $q < k < 1/b$ and the solution can be approximated by setting $\sin(kb)/(kb) = 1$ and the upper limit of the integral to $1/b$. The approximate solution to the integral is then
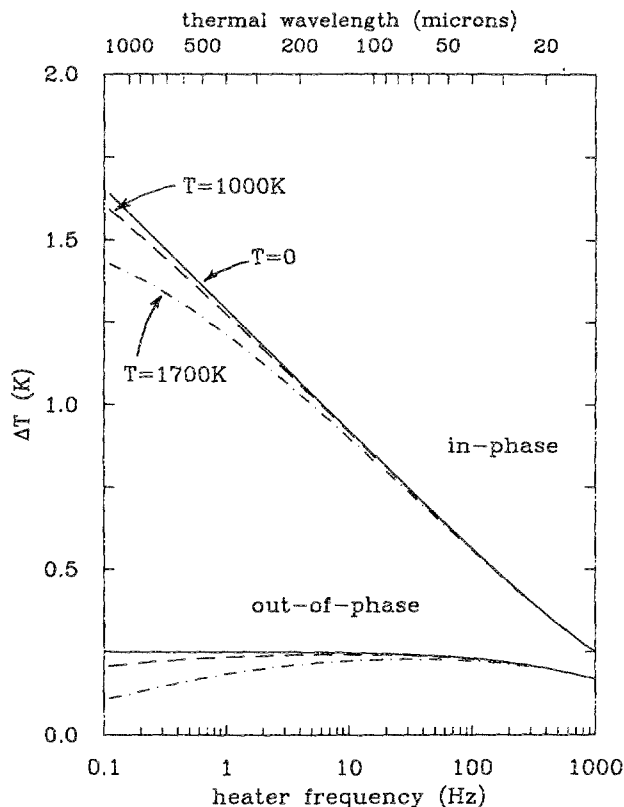

FIG. 2. Temperature oscillations of the metal line as a function of heater frequency calculated using Eq. (11) using a thermal diffusivity $D = 10^{-2}$ cm^2 s^{-1}, line half-width $b = 17.5 \, \mu$m, and thermal conductivity $\Lambda = 10^{-2}$ W cm^{-1} K^{-1}. These values of D and Λ are typical for amorphous solids. The effects of radiation on the temperature oscillations are shown for $T = 0$ (solid line—no radiation errors), $T = 1000$ (dashed line), and $T = 1700$ K (dashed-dot line) using an emissivity $\epsilon = 1$. At 1000 K, the influence of radiation is small, corresponding to an error in determining thermal conductivity of less than 2%.

$- \ln(qb) +$ const, or making the frequency dependence explicit, $-\frac{1}{2} \ln \omega - \frac{1}{2} \ln(ib^2/D) +$ const. In agreement with Eq. (3), the real part of the temperature oscillations (component in phase with the heat) follows a straight line when plotted versus the logarithm of the heater frequency. The imaginary part (component 90° out of phase with the heat) is constant (see Fig. 2, for $T = 0$).

In the derivation of Eq. (8), I assume that the metal line is in intimate thermal contact with the sample; in other words, the effect of the thermal boundary resistance has been ignored. The thermal boundary resistance between evaporated metal films and dielectric crystals was measured by Swartz.[16] He found that the boundary resistance above 50 K was reproducible and small—equivalent to the bulk thermal resistance of an a-SiO$_2$ layer only 20 Å thick. The effect of this boundary resistance is too small to be observed in the temperature oscillations of the metal line. In addition, the 3ω method is insensitive to a boundary resistance because it uses the frequency dependence of the temperature oscillations to determine the thermal conductivity; the boundary resistance would produce a correction to Eq. (8) independent of frequency and would not affect the data analysis.

II. EFFECTS OF BLACK-BODY RADIATION

Earlier,[6] the error in a thermal conductivity measurement due to black-body radiation was estimated by calculating the ratio of heat Q_{rad} lost from the surface of the sample by radiation to the heat Q conducted through the sample:

$$Q_{rad}/Q = (8\epsilon\sigma T^3/\Lambda)\, a, \qquad (9)$$

where σ is the Stefan-Boltzmann constant, ϵ is the emissivity of the sample, and a is a characteristic length for heat flow in the experiment. For the 3ω method, $a = |1/q|$.

A more detailed calculation shows that Eq. (9) is a reasonable estimate of the errors for the 3ω method but overestimates the errors by a factor of ~ 4. Per unit area, power is lost by radiation from the surface of the sample at position x according to: $4\epsilon\sigma T^3\Delta T(x)$. To evaluate the influence of this radiative term, I take its Fourier transform and subtract it from the Fourier transform of the power supplied by the heater. Adding this correction to Eq. (6) gives

$$\Delta T(k) = \frac{1}{2\Lambda}\left[\left(\frac{P\sin(kb)}{lkb} - 4\epsilon\sigma T^3\Delta T(k)\right)\Bigg/ (k^2 + q^2)^{1/2}\right]. \qquad (10)$$

Solving for $\Delta T(k)$ and carrying out the inverse transform and averaging described above gives

$$\Delta T = \frac{P}{l\pi}\int_0^\infty \frac{\sin^2(kb)dk}{(kb)^2[\Lambda(k^2+q^2)^{1/2} + 2\epsilon\sigma T^3]}. \qquad (11)$$

The effect of black-body radiation is most easily demonstrated by comparing a numerical evaluation of Eq. (11) for values of Λ, D, and b typical of a measurement of an amorphous solid (see caption to Fig. 2) and a few values of the temperature T. To estimate the largest possible radiation error, I use $\epsilon = 1$ in Eq. (11). The calculated temperature oscillations are separated into in-phase and out-of-phase components and plotted in Fig. 2 for $T = 0$, 1000, and 1700 K. The effect of radiation is most pronounced at high temperatures and low frequencies where the thermal wavelength is long. In the experiment, the slope of the in-phase component versus the logarithm of the heater frequency in the range 5–30 Hz is used to determine the thermal conductivity. A comparison of these slopes for $T = 0$ and $T = 1000$ K shows that even at 1000 K, the error is less than 2%. The error from radiation scales with $\epsilon T^3b/\Lambda$.

I can now compare the complete solution, Eq. (11), to the estimated value of the radiation errors given by Eq. (9). Using the same value for Λ at $T = 1000$ K and a thermal wavelength of 175 μm (low end of the frequency range used above to determine the thermal conductivity), Eq. (9) gives an error of 8%, a factor of 4 larger than using Eq. (11).

III. SAMPLE PREPARATION

The 3ω method can be used on any dielectric solid with a smooth, flat surface about 1×0.5 cm. Typically, the samples are plates 1 cm^2 and a few millimeters thick. If the sample is a thin film deposited on a substrate,[7,8] the thickness of the film must be at least five times the width of the metal line so that the condition sample thickness $> |1/q| > b$ is satisfied over a

sufficient range of frequencies. For example, the thermal conductivity of a 25 μm-thick film of a-Si:H prepared on an Al substrate was measured using a line 5 μm wide.[8]

The surface of the sample must be smooth enough so that the narrow metal line is continuous; a 5-μm-wide line requires a near mirror finish, but much wider lines of 35 μm work fine even if the surface has a matte finish. The surface must also be clean enough so that the metal film adheres. This is easily accomplished for most samples by ultrasonic cleaning of the sample in acetone followed by isopropyl alcohol and distilled water. If the sample is soluble in water or organic solvents, this procedure cannot be used, but a quick polish with 1-μm alumina powder helps the adherence of the metal films.

Many metals have been used to make the heater-thermometer line, but to measure below 300 K, I prefer a layer of silver, 3000 Å thick, deposited on top of a layer of Nichrome, 100 Å thick, that improves adhesion. The low Debye temperature of silver makes it a useful thermometer to as low as 30 K. The silver films are thermally evaporated in a diffusion-pumped bell jar and typically have residual resistance ratios (RRR; ratio of the room temperature resistance to the 4 K resistance) of 3.

For data above room temperature, silver on top of Nichrome cannot be used since the metals would alloy and result in a metal line that is a poor thermometer. Instead, 3000 Å of Pt sputtered directly onto samples held at 673 K is used. These Pt films adhered poorly to highly polished a-SiO$_2$ and Pyrex; roughening the samples to a matte finish with abrasive paper prior to Pt deposition improved the adhesion. After deposition, the Pt films were annealed for 1 h at 900 K in air to stabilize them for the measurement. The RRR of these films is also typically 3.

The metal lines are made either by evaporation through a mask, or patterned using photolithography. For most samples that are not water soluble, the metal pattern shown in Fig. 3(a), produced by photolithography, gives the best results. The voltage drop is measured far enough away from the ends of the line to eliminate edge effects. The length of the line is accurately defined and the width of the line is
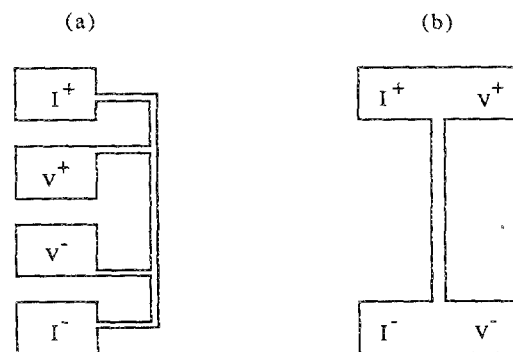

FIG. 3. (a), (b) Metal patterns produced by (a) photolithography and (b) evaporation through a mask. The four rectangular pads are connections for current I^+, I^- and voltage V^+, V^- leads. The evaporation mask in (b) was made from the edges of two nearly touching razor blades. Rectangular areas for the current and voltage leads were cut into the razor blades using the edge of an abrasive wheel. Lines made in this way are typically 90 μm wide.

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constant along its length. Also, the width of the line can be made small so that the 3ω method can be used to measure the thermal conductivity of thin films tens of microns thick.[7,8]

Figure 3(b) shows the pattern produced by evaporating the metal film through a mask. While this procedure is more convenient than photolithography and is the only option for samples that are water soluble, the mask-produced pattern is not as accurate. The length of the line is poorly defined since heat is lost at the ends. But as long as the metal line is long compared to the thermal penetration depth, the edge effects are unimportant. Making a line with constant width of less than 70 μm is difficult; these relatively wide lines require the use of low frequencies, on the order of 1 Hz, for samples with thermal conductivities near those of glasses.

IV. INSTRUMENTATION

The temperature oscillations of the metal line are measured by the third harmonic of the voltage across the line, hence the name 3ω method. A current at angular frequency ω heats the sampe at 2ω and produces the temperature oscillation calculated above at angular frequency 2ω. The resistance of a pure metal increases with increasing temperature and therefore the resistance of the metal line has a small ac component that oscillates at 2ω. This resistance oscillation times the original driving current oscillating at ω results in a small oscillation of the voltage across the line at 3ω.

Figure 4 shows a schematic of the electrical circuit used to measure the 3ω component of the voltage across the metal line. The ac signal source, a HP 3325 frequency synthesizer, produces a sine wave with low harmonic distortion; third harmonic content in the source can produce spurious signals in the measurement. Since the ω voltage is typically 1000 times as large as the 3ω voltage, this voltage must be sub-
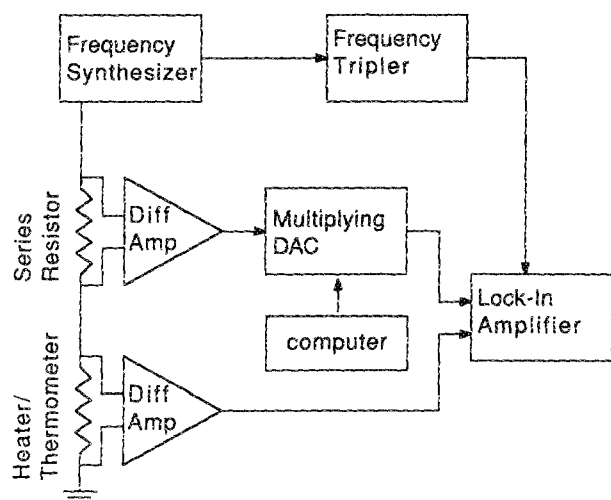

FIG. 4. Schematic diagram of the equipment used for thermal conductivity measurements using the 3ω method. The frequency synthesizer is a Hewlett-Packard model 3325. Unity-gain differential amplifiers, Analog Devices 524, multiplying digital-to-analog (DAC) converter, Analog Devices model 7541, and the ten-turn pot used for the series resistor are mounted in a Tektronix TM503 power supply. The reference signal for the two-phase lock-in amplifier, Princeton Applied Research 5208, is supplied by a homemade frequency tripler (see Fig. 5). The frequency synthesizer, multiplying DAC and lock-in amplifier are controlled by a LSI 11/23-based computer.
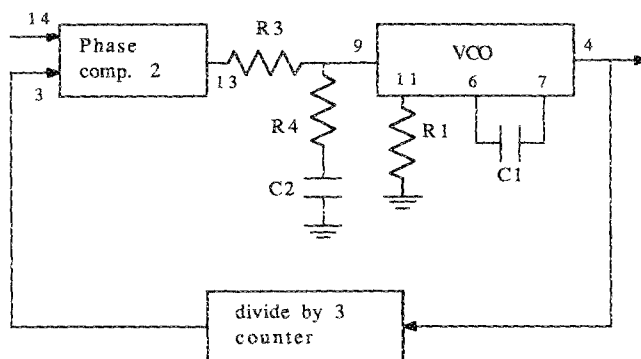

FIG. 5. Schematic diagram of the frequency tripler (see Ref. 17) that produces a 5 V square output at three times the frequency of the square wave supplied by the SYNC output of the frequency synthesizer. For each capacitor and resistor, the first value listed is for the low frequency version and the second value for the high frequency version: $R\,1 = 100$, $5\,k\Omega$, $R\,3 = 100$, $1\,M\Omega$, $R\,4 = 100,1\,k\Omega$, $C\,1 = 0.03\mu F,300pF$, $C\,2 = 10,1\mu F$. The phase comparator and voltage-controlled oscillator (VCO) are parts of a single phase-locked loop chip (MC14046B). The divide-by-3 function is performed by a divide-by-N counter (CD4018BE) and two NAND gates (CD4011BE).

tracted before the 3ω signal can be measured by the lock-in amplifier. The subtraction is accomplished using a multiplying digital-to-analog converter (DAC) and the differential input of the lock-in amplifier. The gain of the multiplying DAC can be varied from 0 to 1 by computer control of 12 TTL levels. In this way, the multiplying DAC is adjusted until its output is equal to the voltage across the metal line. The differential input of the lock-in amplifier can then reduce the ω component to an acceptable level.

The reference channel of the lock-in is connected to the output of a homemade device that produces a square wave output at three times the frequency of the input[17]; (see Fig. 5). Birge and Nagel[18,19] have previously described an experimental technique based on measuring a voltage at 3ω using a lock-in amplifier. The only difference between this frequency tripler and the one described by Birge and Nagel is the values of the resistors and capacitors used to set the frequency range and filter parameters of the phase-locked loop. No single set of resistors and capacitors can cover the entire frequency range. I built two triplers: one to cover the range $3 < \omega < 10^3$ rad s^{-1} and a second for $10^3 < \omega < 10^5$ rad s^{-1}.

V. EXPERIMENTAL METHOD

In addition to eliminating errors from black-body radiation, the 3ω method has another significant advantage over conventional techniques. Long equilibration times sometimes lasting many hours, that plague high temperature measurements employing the conventional techniques, are reduced to a few periods of the temperature oscillations of the metal line, a few seconds at most. Thus, accurate control of the temperature of the oven or cryostat for hours is not required for the 3ω method; data can be obtained while slowly heating or cooling the sample.

Experiments below room temperature were made in a portable cryostat that can be inserted into a helium storage Dewar.[20] Data were taken while slowly cooling the sample at

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a controlled rate of approximately 0.2% of the temperature per minute. Between room temperature and 200 K, the cryostat was held above a bath of liquid nitrogen and cooled only by contact with cold N_2 gas. Between 200 and 100 K the cryostat was immersed in liquid N_2. Below 100 K, the cryostat was inserted into a liquid helium storage Dewar but held above the liquid. Electrical connection to the metal film was made through four spring-loaded pins mounted in a plastic holder. A thin coating of tin-lead solder on the tips of these pins improves the reliability of the electrical contact between the pins and the rectangular metal pads on the sample. Swartz[21] has recently described a similar method of making electrical contact to metal films.

Above room temperature, samples were mounted on a copper block and supported in the center of a 10-cm-diam stainless-steel tube that could be evacuated. Radiation shields were placed around the copper block and sample so that they were at the same temperature; the temperature was measured by a chromel-alumel thermocouple. The stainless-steel tube was placed horizontally in a tube furnace; data were collected while slowly heating the furnace at a controlled rate of 0.5 K/min. Electrical connection to the Pt films was made through 0–80 stainless-steel screws mounted in a machinable ceramic holder. A small amount of silver paste placed between the stainelss-steel screws and the Pt films produced reliable electrical contact. Electrical leads were Cu-Ni surrounded by MgO insulation and a stainless-steel sheath. The leads were spot-welded to the 0–80 screws.

Before starting the measurement, the series resistor, usually a 10-turn 100-Ω potentiometer, was set to a value close to but larger than the largest resistance that the metal line will reach during the run. The series resistor is calibrated by replacing the metal line, marked heater/thermometer in Fig. 5, by a standard resistor and measuring the ratio of the voltage across the series resistor to the voltage across the standard when the gain of the multiplying digital-to-analog-converter (DAC) is set to 1. Measuring the voltages at 33 Hz with an ac voltmeter eliminates errors from dc voltage offsets. The calibrated value of the series resistor is then the value of the standard multiplied by this voltage ratio. The resistance of the metal line was determined in the same way. The resistance of the line is equal to the resistance of the series resistor times the ratio of the voltage across the line to the voltage across the series resistor.

At each temperature of measurement, the resistance of the line is measured twice: first with a voltage of small amplitude from the frequency synthesizer to avoid significant heating of the sample and a second time with the voltage of the synthesizer set to the amplitude used for measuring the 3ω component of the voltage oscillations. This amplitude is chosen so that the 3ω voltage is ~ 1000 smaller than the ω voltage. The resistance measured at small amplitude is used for calibrating the temperature of the line as a function of its resistance. When this calibration is known, the resistance measured at large amplitude gives the temperature of the sample during the measurement. The temperature of sample during large-amplitude heating of the line is typically a few kelvin larger than that of the sample in equilibrium with the cryostat.

The gain of the multiplying DAC is then adjusted to make the ω voltage from the series reistor equal to the ω voltage from the metal line. The frequency of the synthesizer is set to the desired frequency and the lock-in amplifier measures the in-phase and out-of-phase components of the voltage at angular frequency 3ω. To determine the thermal conductivity, the 3ω voltage is measured as a function of frequency. At each temperature, I usually use four frequencies separated by the factor $\exp(\pi/4)$, although only two of these data points are used in calculating the thermal conductivity. The change of the in-phase voltage when the frequency is changed by $\exp(\pi/2)$ should be equal to the constant out-of-phase voltage and provides a quick check of the validity of the data. In the absence of any phase shifts introduced by the lock-in amplifier or other electronics, the thermal conductivity could be determined from the magnitude of the out-of-phase 3ω voltage just as well as from the slope of the in-phase. In practice, however, the slope of the in-phase voltage has proved more accurate.

VI. DATA ANALYSIS

Figure 6 shows the measured ΔT of the metal line on an a-SiO$_2$ sample at 300 K. The temperature oscillations are
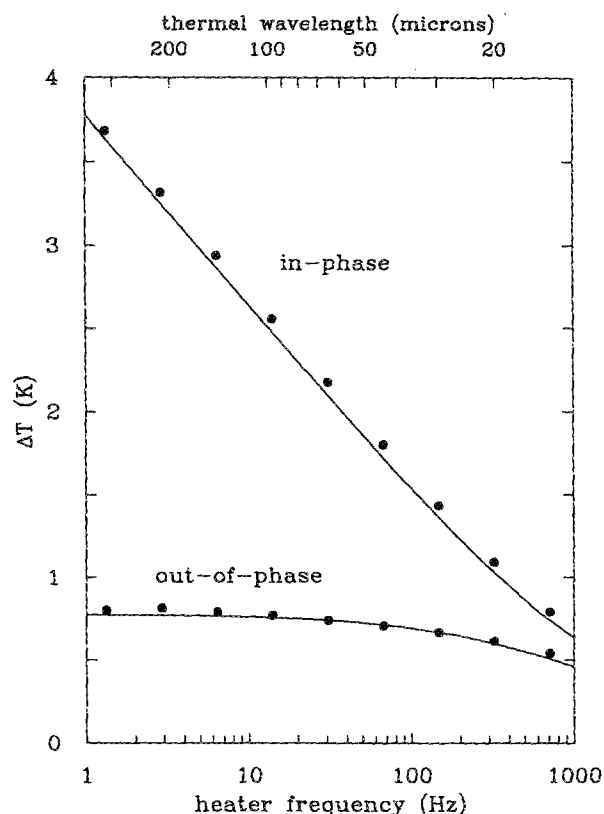

FIG. 6. Example of the temperature oscillations ΔT measured in the 3ω method at $T = 300$ K on a-SiO$_2$; ΔT has been separated into the amplitude of the in-phase and out-of-phase components. The slope of the in-phase ΔT versus the logarithm of the heater frequency gives the thermal conductivity, $\Lambda = 1.35 \times 10^{-2}$ W cm^{-1} K^{-1}. The solid lines are Eq. (8) using $\Lambda = 1.35 \times 10^{-2}$ W cm^{-1} K^{-1}, the measured linewidth $2b = 40 \ \mu$m, and the thermal diffusivity (see Ref. 22) of a-SiO$_2$, $D = 8.3 \times 10^{-3}$ cm^2 s^{-1}.

calculated from

$$\Delta T = 4 \frac{dT}{dR} \frac{R}{V} V_3 , \tag{12}$$

where R is the average resistance of the metal line, V the voltage across the metal line at ω, and V_3 is the measured voltage at 3ω. All voltages are rms. The calibration of the line (temperatures as a function of resistance) is differentiated to get dT/dR. Using the 3ω voltage measured at two frequencies f_1 and f_2, the thermal conductivity Λ is calculated from[6]

$$\Lambda = \frac{V^3 \ln f_2/f_1}{4\pi l R^2 (V_{3,1} - V_{3,2})} \frac{dR}{dT} , \tag{13}$$

where $V_{3,1}$ is the in-phase 3ω voltage at frequency f_1 and $V_{3,2}$ is the in-phase 3ω voltage at frequency f_2. In this example, using $f_1 = 7$ Hz and $f_2 = 33.6$ Hz, I measure $\Lambda = 1.35 \times 10^{-2}$ W cm^{-1} K^{-1}.

Solid lines in Fig. 6 show the calculated ΔT using Λ determined above, the measured width of the metal line, the thermal diffusivity[22] of a-SiO$_2$ (see caption of Fig. 6), and P/l from the experiment. Only the magnitude of the in-phase ΔT is sensitive to the procedure used to take into account the width of the metal line; the good agreement between measured and calculated magnitude of the in-phase ΔT confirms the usefulness of Eq. (8) and its derivation. The usefulness of the 3ω method for measuring the thermal conductivity of thin films is also demonstrated by Fig. 6. The slope of the in-phase ΔT is constant from a thermal wavelength of 300 down to 30 μm. In other words, the 3ω method measures the same thermal conductivity on a bulk sample (300 μm thick) as on a thin layer (30 μm thick) at the surface of the sample. My ability to test the 3ω method at the surface of a bulk sample with a known thermal conductivity adds to the reliability of data on evaporated thin films.[7,8]
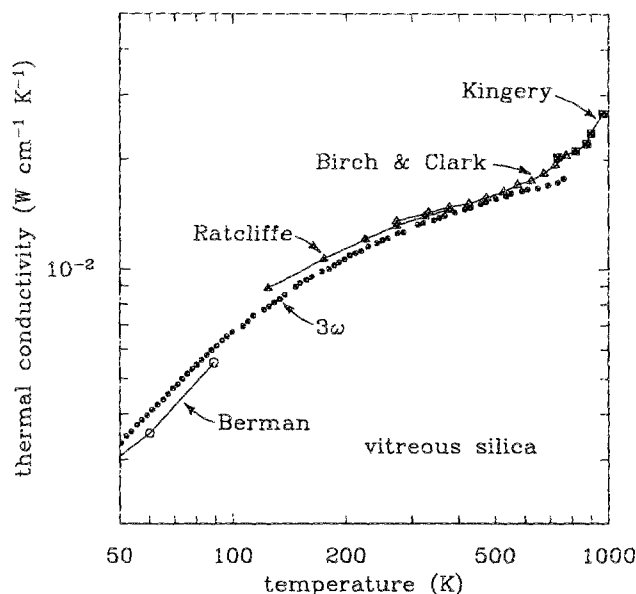
To evaluate the accuracy of the 3ω method, the thermal conductivity of a-SiO$_2$ and two thermal conductivity standards was measured: Pyroceram 9606 and Pyrex 7740 (Figs. 7 and 8). The solid circles in Fig. 7 demonstrate the wide range of temperature covered by the 3ω method bridging the gap between low-temperature methods[2] and the elaborate means[23-25] used to reduce radiation errors near room temperature and above. Even with great care taken to eliminate radiation effects, data for a-SiO$_2$ taken with conventional techniques are strongly affected by radiation errors at $T > 700$ K. The maximum temperature of the data measured by the 3ω method, 750 K for a-SiO$_2$ and Pyroceram 9606 and 500 K for Pyrex 7740 (see Fig. 8), was limited by the stability of the Pt film used to make the metal line. At higher temperatures the Pt line was no longer a reliable thermometer. I do not understand this limitation, especially since annealing the Pt films in air at 900 K (see above) did not degrade them.

Pyroceram 9606, a glass ceramic, and Pyrex 7740, a borosilicate glass, have long been used as standard materials for testing experimental techniques as well as a calibration for comparative methods. Figure 8 compares data taken with the 3ω method[26] to the results of an extensive study of these materials in seven laboratories that are experienced in thermal conductivity measurement.[5] Although the large spread in data from the round-robin study (as large as 25%) makes a critical test difficult, the agreement between the recommended values and our data is close and demonstrates the accuracy of the 3ω method (see caption to Fig. 8).
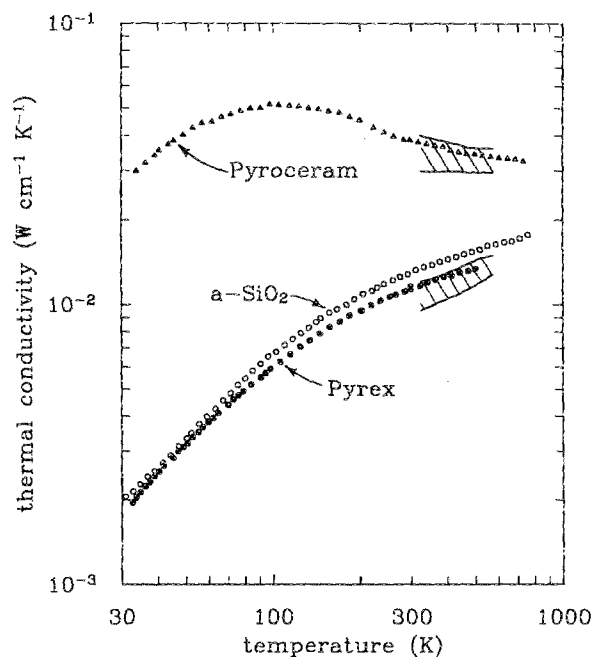

FIG. 8. Our data for a-SiO$_2$ (open circles; same data as shown in Fig. 7), for Pyrex 7740 (filled circles), and for Pyroceram 9606 (filled triangles) compared to results of a round-robin study of these standard thermal conductivity materials (see Ref. 5). The spread in the round-robin results on Pyrex and Pyroceram is shown as cross-hatched areas. These data have also recently been published in tables (see Ref. 26). [In an earlier work (Ref. 6) the data marked "Pyrex" were labeled incorrectly. The data were obtained on a sample of a-SiO$_2$, not Pyrex 7740 as we had originally believed.]

FIG. 7. Data for a-SiO$_2$ (Vitreosil made by Thermal American) taken with the 3ω method, compared to data obtained with conventional techniques (see Refs. 2, 23, 24, and 25). Above 600 K, the conventional techniques are strongly affected by radiation errors.

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