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## Thermal conductivity of amorphous solids above the plateau

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We have measured the thermal conductivity of polymethylmethacrylate (PMMA), amorphous  $\text{As}_2\text{S}_3$ , Ca-K nitrate glass, three  $\text{SiO}_2$ -based glasses, and glycerol in the temperature range 30–300 K, using an ac technique that eliminates errors from blackbody radiation. This technique and the problems of blackbody radiation in thermal measurements are discussed in detail. Our data do not support a recent prediction of heat transport by fractons. Instead, we find that the thermal conductivity of these glasses above  $\sim 50$  K is well described by the minimum thermal conductivity suggested by Slack.

### I. INTRODUCTION

The nature of heat transport in amorphous solids has received much attention in recent years. Although progress has been made in understanding the thermal conductivity of glasses at temperatures below 1 K, researchers disagree on the origin of the plateau in thermal conductivity and the mechanisms that are important for heat transport in amorphous solids at temperatures above the plateau.<sup>1</sup> Understanding the high-temperature thermal conductivity of glasses has been hindered, in part, by a lack of reliable data in this temperature range, mostly due to long thermal equilibration times and to the difficulty in obtaining data free from errors caused by blackbody infrared radiation (for a review of the limited data see Ref. 2).

These difficulties can be greatly reduced with the technique described in this paper. We have used this technique to obtain accurate measurements of the thermal conductivity of seven different glasses between 30 and 300 K. We do not find the linear temperature dependence in this temperature range which has been postulated by Alexander *et al.* for the heat transport by fractons.<sup>3</sup> Instead, we find above  $\sim 50$  K a magnitude and temperature dependence which agrees with the model of the minimum thermal conductivity proposed by Slack.<sup>4</sup> In the limit of high temperatures, this model leads to a phonon mean free path equal to the interatomic spacing as first noted in glasses by Birch and Clark<sup>5</sup> and by Kittel.<sup>6</sup>

### II. EXPERIMENTAL TECHNIQUE

The technique we developed, which we call the  $3\omega$  method, is an ac technique closely related to the hot-wire and hot-strip methods for measuring thermal conductivity. In all three techniques a single element is used simultaneously as heater and thermometer. The hot-wire technique has been developed into a very useful technique for the measurement of liquids and of solids under pressure.<sup>7–9</sup> The hot-strip method differs from the hot-wire method through the use of a different geometry; the metal wire used in the hot-wire method is replaced by a wide, rectangular metal film that is evaporated onto the surface

of the sample.<sup>10</sup> Both the hot-wire and hot-strip techniques are performed in the time domain; a step function of power is applied to the wire or strip and the resistance of the metal is followed as a function of time. The  $3\omega$  method takes advantage of lock-in amplifier technology and moves the measurement into the frequency domain; the amplitude and phase of the resistance changes of the evaporated metal film are measured as a function of the excitation frequency.

The Angstrom method<sup>11</sup> and a.c. calorimetry,<sup>12</sup> using separate heaters and thermometers, are examples of frequency-domain techniques. Recently, several researchers have combined the use of frequency-domain techniques with the use of a single element used as heater and thermometer.<sup>13–15</sup> This combination of frequency-domain techniques and a single heater-thermometer was done using an evaporated metal film which was wide relative to the wavelength of the diffusive thermal wave. This geometry leads to a one-dimensional flow of heat from which the product of thermal conductivity and specific heat can be determined. In our experiment we use metal films with a width small compared to the wavelength of the diffusive thermal wave. This geometry leads to a cylindrical heat flow pattern and allows us to determine the thermal conductivity directly.

Birge<sup>16</sup> has also recently reported an ac technique that uses cylindrical geometry. In his work, the thermal conductivity of glycerol was measured with a 13- $\mu\text{m}$ -diameter, platinum wire suspended in the liquid.

To derive the equation relating the thermal conductivity to the temperature oscillations of the narrow film heater-thermometer, we begin with the exact solution for the amplitude of the temperature oscillations a distance  $r$  from a line source of heat. This amplitude,  $\Delta T$ , is given by Carslaw and Jaeger.<sup>17</sup>

$$\Delta T = \frac{P}{l\pi\Lambda} K_0(qr), \quad (1)$$

where  $P/l$  is the amplitude of the power per unit length generated by a heater current of angular frequency  $\omega$  passing through the narrow metal line,  $\Lambda$  is the thermal conductivity, and  $K_0$  is the zeroth-order modified Bessel

function. The magnitude of the complex quantity  $1/q$  is the wavelength of the diffusive thermal wave, or more precisely, the thermal penetration depth, the depth that heat diffuses during one cycle of the ac power heating the sample:

$$\frac{1}{q} = \left( \frac{D}{i\omega} \right)^{1/2}, \quad (2)$$

where  $D$  is the thermal diffusivity,  $D = \Lambda/C_p$ , and  $C_p$ , the specific heat per volume. For  $|qr| \ll 1$ , the result can be written approximately as

$$\Delta T = \frac{P}{\pi l \Lambda} \left[ \frac{1}{2} \ln \frac{D}{r^2} + \ln 2 - 0.5772 - \frac{1}{2} \ln \omega - \frac{i\pi}{4} \right]. \quad (3)$$

The result has been written to separate the frequency-dependent and the imaginary contributions to the solution. A complete solution for the temperature oscillations in a line source of finite width involves a convolution of this result across the width  $2w$  of the line and then an average across the line, but since we are only interested in determining the thermal conductivity, and not the thermal diffusivity, a complete solution is unnecessary. As long as the thermal penetration depth is much larger than the width of the line, i.e.,  $|qw| \ll 1$ , the slope of the  $\Delta T$  versus  $\ln(\omega)$  curve is independent of the width of the line and of the averaging procedure.

In this calculation, we assumed that the heater-thermometer is in intimate thermal contact with the sample. Recent measurements of the thermal boundary resistance between evaporated metal films and dielectric substrates have shown that the thermal boundary resistance is indeed small enough to be ignored in these measurements.<sup>18</sup>

### III. BLACKBODY RADIATION IN THERMAL MEASUREMENTS

In all thermal conductivity measurements, the experimenter measures a temperature change that results from a known flow of heat through the sample. Blackbody infrared radiation causes errors in thermal measurements by producing an uncertainty in the quantity of heat flowing through the sample; heat can be carried away from the sample by blackbody radiation. This error is proportional to the third power of temperature and the reciprocal of the thermal conductance of the sample, a functional dependence that creates the greatest difficulties for high temperature measurements of long samples of low thermal conductivity. The following calculation of radiation errors will demonstrate this dependence and will reveal why the  $3\omega$  method is essentially immune to these radiation errors to temperatures as high as 1000 K.

In a standard, steady-state measurement of thermal conductivity, heat flows linearly through a sample of length  $l$  (typically 2.5 cm) and cross section  $A$  (typically  $0.1 \text{ cm}^2$ ). One end of the sample is held at a fixed temperature  $T$ . To produce a temperature change  $\Delta T$  requires a heat flow of  $\dot{Q} = \Lambda \Delta T A / l$

We will make some simplifying assumptions to calculate the heat lost to blackbody infrared radiation in this

measurement: a radiation shield at temperature  $T$  surrounds the sample;  $\Delta T$  is small so that the radiation loss is proportional to  $\Delta T$ ; and the sample loses heat only from the side surfaces of the sample, not from the top and from the attached heaters and thermometers.

The sides of the sample have a surface area of  $4l\sqrt{A}$ . The average temperature of the sides of the sample is  $T + \Delta T/2$ . The error in the measurement due to radiation is  $\dot{Q}_{\text{rad}}/\dot{Q}$ .

$$\frac{\dot{Q}_{\text{rad}}}{\dot{Q}} = \frac{8\epsilon\sigma T^3}{\Lambda} \left[ \frac{l^2}{\sqrt{A}} \right], \quad (4)$$

where  $\sigma$  is the Stephan-Boltzmann constant and  $\epsilon$  is the emissivity of the sample. The total emissivity of most dielectrics is close to one at temperatures near room temperature.<sup>19</sup> Using the dimensions described above, the error introduced by blackbody infrared radiation is approximately 15% for  $\alpha\text{-SiO}_2$  at 100 K. Radiation from the attached heaters and thermometers would increase the error.

The radiation error is greatly reduced by using the  $3\omega$  method. For most experimental circumstances the term in brackets in Eq. (3) is typically of order  $\pi$ . A temperature change  $\Delta T$  requires a heat flow of  $\dot{Q} \approx \Lambda \Delta T l$ , where  $l$  is the length of the heater-thermometer.

To calculate the heat lost to blackbody infrared radiation we will again assume that the radiation shield is held at temperature  $T$ , the average temperature of the sample. This assumption is mostly for convenience and does not greatly affect our results. In the  $3\omega$  method heat is lost only from a small area on the surface of the sample; the area is the surface within the thermal penetration depth of the heater-thermometer. The radiation error in the  $3\omega$  method is  $\dot{Q}_{\text{rad}}/\dot{Q}$ :

$$\frac{\dot{Q}_{\text{rad}}}{\dot{Q}} = \frac{8\epsilon\sigma T^3}{\Lambda} \left| \frac{1}{q} \right|. \quad (5)$$

A thermal penetration depth of  $30 \mu\text{m}$  reduces the errors due to radiation by a factor of approximately  $10^4$  compared to the standard geometry described above. This small thermal penetration depth can be achieved in a typical glass with a  $5\text{-}\mu\text{m}$ -wide heater-thermometer and a rather modest heater frequency of a few hundred hertz. Even at 1000 K the calculated error due to radiation in this case is less than 2%.

### IV. EXPERIMENTAL DETAILS

Figure 1 shows a sample with the metal line that serves simultaneously as heater and thermometer. Electrical connections to the line are made by spring-loaded pins that contact the rectangular, metallized pads on the sample. The narrow metal line and the rectangular pads are produced on the sample either by photolithography or by evaporation through a mask. For most samples that are not soluble in water, we use photolithography to produce a metal line that is either 5 or  $35 \mu\text{m}$  wide. For other samples the line is evaporated through a mask. Lines pro-

duced in this way were 90  $\mu\text{m}$  wide. Although many metals have been used, we prefer a 5000- $\text{\AA}$ -thick layer of silver deposited on top of a 100- $\text{\AA}$ -thick layer of Nichrome. The layer of Nichrome improves the adhesion to the sample. Silver is used because it is easy to evaporate and remains a sensitive thermometer even below 30 K.

In addition to the elimination of errors from blackbody infrared radiation, the  $3\omega$  method has two other advantages over the steady-state method. Very small samples can be used. The only constraint on the sample size is the size of the metal pattern evaporated on the surface. In order to avoid reflections, the sample thickness must be at least five times the width of the line and the surface of the sample must be large enough to accommodate the length of the metal line and the electrical connections. A second advantage of the  $3\omega$  method is that the long equilibration times, lasting sometimes many hours, that plague high-temperature thermal measurements employing the standard technique are reduced to a few periods of the temperature oscillations of the metal line, a few seconds at most. Thus, accurate temperature control of the cryostat for hours is not required for the  $3\omega$  method; data can be obtained while slowly heating or cooling the sample.

Our measurements were performed in a small, evacuated transportable  $^4\text{He}$  cryostat.<sup>20</sup> The samples were attached with N grease directly to its copper stage. We took data while cooling the sample at approximately 0.2 K per minute. From 300 to 200 K the cryostat was held above liquid  $\text{N}_2$  and cooled only by contact with the cold  $\text{N}_2$  gas. From 200 to 100 K the cryostat was immersed in liquid  $\text{N}_2$ . Below 100 K the cryostat was inserted into a  $^4\text{He}$  storage dewar. The measurements were completely automated; an LSI 11/23 based computer controlled the temperature of the cryostat and the electronics for data acquisition.

Figure 2 shows a schematic diagram of the equipment used in these measurements. A frequency synthesizer supplies the driving current at an amplitude and frequency selected by the computer. The output of the frequency synthesizer is a very pure sine wave; harmonics present in

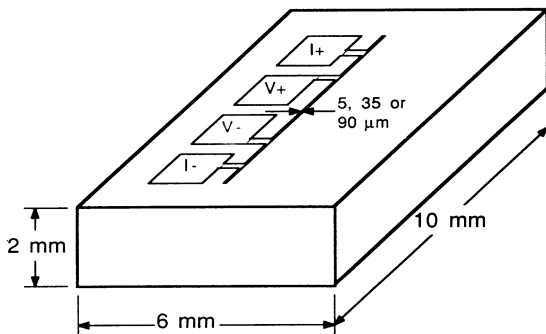


FIG. 1. Evaporated metal pattern produced on the face of a sample. The four pads are the connections for current leads  $I^+$ ,  $I^-$  and voltage leads,  $V^+$ ,  $V^-$ . The narrow metal line that serves as the heater and thermometer for the measurement of the thermal conductivity is at the center of the face of the sample. Pattern made by photolithography (line widths 5 or 35  $\mu\text{m}$ ) or by evaporation through a mask (line width 90  $\mu\text{m}$ ).

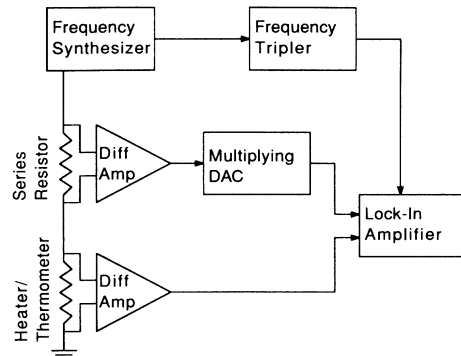


FIG. 2. Schematic diagram of the equipment used for thermal conductivity measurements using the  $3\omega$  method.

the source can cause errors in the measurement. The sinusoidal current of angular frequency  $\omega$  heats the sample at  $2\omega$ . The heating produces the temperature oscillation that was calculated above. The metal line also serves as a thermometer; the resistance of the line is a function of the temperature. The resistance oscillation at  $2\omega$  multiplied by the excitation current at  $\omega$  produces a voltage oscillation at  $3\omega$ . The amplitude of this  $3\omega$  voltage is measured by the lock-in amplifier (hence, the name:  $3\omega$  method).

In addition to the voltage at  $3\omega$ , a very large voltage at  $\omega$  is present across the heater-thermometer; the  $\omega$  voltage is typically 1000 times larger than the  $3\omega$  voltage. To reduce this large voltage at frequency  $\omega$ , a resistor is placed in series with the heater-thermometer resistance. By adjusting the gain of the multiplying digital-to-analog converter, the  $\omega$  voltage from the series resistor can be made equal to the  $\omega$  voltage from the heater-thermometer. The differential input of the lock-in amplifier can then reduce the  $\omega$  content of the input voltage to an acceptable level.

A frequency tripler<sup>21</sup> provides a reference signal at  $3\omega$ . By measuring the third-harmonic signal at two frequencies,  $f_1$  and  $f_2$ , we obtain the thermal conductivity  $\Lambda$ :

$$\Lambda = \frac{V^3 \ln \frac{f_2}{f_1}}{4\pi l R^2 (V_{3,1} - V_{3,2})} \frac{dR}{dT} \quad (6)$$

All voltages are rms.  $R$  is the average resistance of the metal line,  $V$  is the voltage across the line at frequency  $\omega$ , and  $V_{3,1}$  is the voltage at the third harmonic for frequency  $f_1$  and  $V_{3,2}$  is the voltage at the third harmonic for frequency  $f_2$ .  $dR/dT$  is the slope of the calibration of the metal line (resistance as a function of temperature) at the temperature of the measurement.

## V. SAMPLE DESCRIPTIONS

The polymethylmethacrylate (PMMA) sample was cut from a piece of cast, commercial-grade polymethylmethacrylate. The  $\text{As}_2\text{S}_3$  sample was obtained from Hori-ba Ltd. It was deep red and clear; no internal voids or bubbles were visible. We made the  $\text{Ca}_{0.4}\text{K}_{0.6}(\text{NO}_3)_{1.4}$  glass by mixing reagent grade  $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$  and

$\text{KNO}_3$ . Heating the mixture at  $400^\circ\text{C}$  for 1 h removes the water of hydration and produces a clear viscous liquid, which forms a glass quite easily when poured into an aluminum mold.<sup>22</sup> The nitrate glasses produced in this way were clear and brittle at room temperature. The microscope slide used is an example of a soft glass or soda lime glass. Its exact composition is unknown. Vitreosil is amorphous  $\text{SiO}_2$  produced by melting natural quartz. Our sample of Vitreosil was obtained from Thermal American. This material has a low concentration of water impurities but a relatively large concentration of metal impurities. The Pyrex sample was cut from a large piece of Corning Pyrex No. 7740 that is undergoing extensive measurements for use as a thermal conductivity standard.<sup>23</sup>

We measured the thermal conductivity of glycerol by an extension of the technique described above. We prepared a  $5\text{-}\mu\text{m}$ -wide heater-thermometer on a microscope slide. This narrow metal line was made by photolithography of an evaporated thin film of Ni; Al and Cu corroded during the measurements of glycerol. A single drop of reagent grade glycerol was placed on top of this microscope slide and the thermal conductivity of the combination of glycerol and slide was measured. We extracted the thermal conductivity of glycerol by subtracting the thermal conductivity of the slide from the data obtained on the combination of glycerol on top of the slide.

## VI. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 3 is a comparison of our measurements to earlier work. The solid line for  $\text{SiO}_2$  is a recommended average based on a compilation of measurements by several researchers.<sup>24</sup> Our data agree with this recommended curve to within a few percent except at temperatures above 200 K. The solid line for PMMA was obtained by

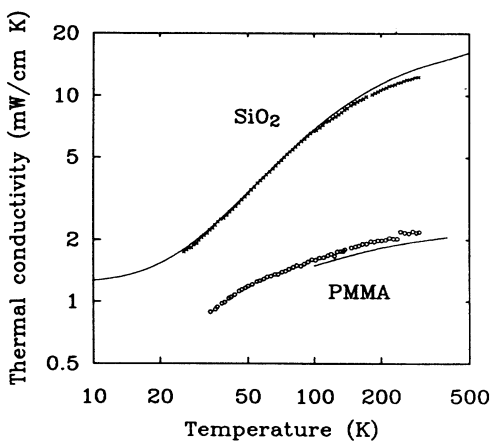


FIG. 3. Comparison of the present data to earlier work. Data points are from this work. The solid line for  $\text{SiO}_2$  is the recommended curve, based on a compilation of data by several researchers (Ref. 24). The solid line for PMMA is from Ref. 25.

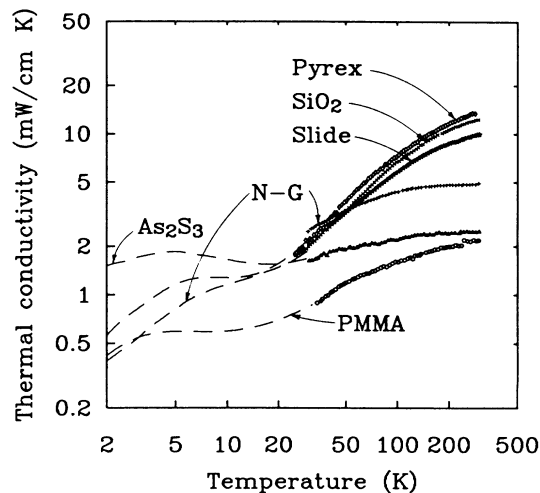


FIG. 4. Thermal conductivity of the six amorphous solids described in the text on a doubly logarithmic scale. Data points are from this work; dashed lines are from earlier work: PMMA, see Ref. 26,  $\text{SiO}_2$  and nitrate glass (N-G), Ref. 27,  $\text{As}_2\text{S}_3$ , Ref. 28. Only for the nitrate glass do our data not match up with the earlier data, as shown even more clearly in Fig. 5.

Eiermann and Hellwege.<sup>25</sup> The agreement is also very close.

The thermal conductivity data on six different glass samples measured in this investigation are plotted in Fig. 4 along with earlier low temperature measurements taken from Refs. 26–28 and drawn as dashed lines. They join smoothly to the high-temperature data except in the case of the nitrate glass. The oxide glasses and PMMA show a well-defined plateau, the thermal conductivity is approximately temperature independent in the temperature range 5–20 K but varies with temperature at both high and low temperatures. The nitrate glass shows only a very weak plateau. The thermal conductivity of  $\text{As}_2\text{S}_3$  at room temperature is only slightly larger than the thermal conductivity at 5 K.

In Fig. 5 we have plotted the measured thermal conductivity on a linear temperature scale to compare our data to the theory that predicts a linear temperature dependence of the thermal conductivity above the plateau.<sup>3</sup> This theory, which explains the thermal conductivity of glasses at high temperatures by phonon-assisted fracton hopping, predicts a linear temperature dependence that has an intercept at zero temperature close to the magnitude of the conductivity at the plateau. We have also included earlier measurements on epoxy resin that were found to support the fracton model in the limited temperature range below 100 K.<sup>3</sup> Instead of this predicted linear variation of  $\Lambda$  with  $T$ , we find that the slopes of all curves,  $d\Lambda/dT$ , decrease smoothly with increasing temperature, with  $\Lambda$  approaching a constant value at the highest temperatures. The straight-line behavior observed for the epoxy may be a consequence of the limited temperature range investigated.

We will now show that the thermal conductivity we have observed agrees well with the minimum thermal con-

TABLE I. Comparison of the measured thermal conductivity to the calculated thermal conductivity in the high-temperature limit. Column 1: Atomic number density. For PMMA the following atoms or groups of atoms were treated as a single "atom:" CH<sub>2</sub>, C, CH<sub>3</sub>, CO, and O. For glycerol the atoms were grouped as follows: CH<sub>2</sub>, CH, OH. Columns 2 and 3: Transverse and longitudinal speeds of sound at 4 K, except for glycerol, for which the speeds of sound are for 170 K. Footnotes give the references for the speeds of sound measurements. Column 4: The average speed of sound calculated from  $\frac{2}{3}v_t + \frac{1}{3}v_l$ . Column 5: Average interatomic spacing. Column 6: Calculated thermal conductivity assuming an equipartition value for the specific heat and a mean-free-path of one interatomic spacing. Column 7: Measured thermal conductivity at 300 K, which is taken as the high-temperature limit, except for glycerol, for which the value listed is for 180 K. The microscope slide was assumed to be of the composition 3·SiO<sub>2</sub>-Na<sub>2</sub>O.

	$n$ (10 <sup>22</sup> cm <sup>-3</sup> )	$v_t$ (10 <sup>5</sup> cm/s)	$v_l$ (10 <sup>5</sup> cm/s)	$\bar{v}$ (10 <sup>5</sup> cm/s)	$l$ (Å)	$\Lambda_{\text{calc}}$ (mW/cm K)	$\Lambda_{\text{meas}}$ (mW/cm K)
Pyrex	6.97	3.4	5.5 <sup>a</sup>	4.1	2.43	9.6	13.4
SiO <sub>2</sub>	6.61	3.7	5.8 <sup>a</sup>	4.4	2.47	9.9	12.3
Slide	7.16	3.20	5.37 <sup>b</sup>	3.92	2.41	9.3	9.9
Nitrate glass	6.92	1.73	3.5 <sup>c</sup>	2.32	2.44	5.4	4.9
As <sub>2</sub> S <sub>3</sub>	3.92	1.44	2.65 <sup>d</sup>	1.84	2.94	2.9	2.5
PMMA	4.33	1.57	3.15 <sup>e</sup>	2.10	2.85	3.5	2.0
Glycerol	5.18	1.7	3.5 <sup>f</sup>	2.3	2.68	4.4	3.3

<sup>a</sup> W. F. Love, Phys. Rev. Lett. 31, 822 (1973).

<sup>b</sup> Richard B. Stephens, Phys. Rev. B 13, 852 (1976).

<sup>c</sup> J. J. De Yoreo, Ph.D. thesis, Cornell University, 1985 (unpublished).

<sup>d</sup> R. B. Stephens (private communication).

<sup>e</sup> U. T. Reichert, Ph.D. thesis, Heidelberg University, 1986 (unpublished).

<sup>f</sup> Robert Piccirelli and T. A. Litovitz, J. Acoust. Soc. Am. 29, 1009 (1954).

ductivity proposed by Slack,<sup>3</sup> for temperatures above  $\sim 50$  K. This model starts from the gas-kinetic formula first suggested by Debye for the heat transport by elastic waves traveling with the sound velocity  $v$  between collisions spaced by the average mean free path  $l$ :

$$\Lambda = \left(\frac{1}{3}\right)Cv l, \quad (7)$$

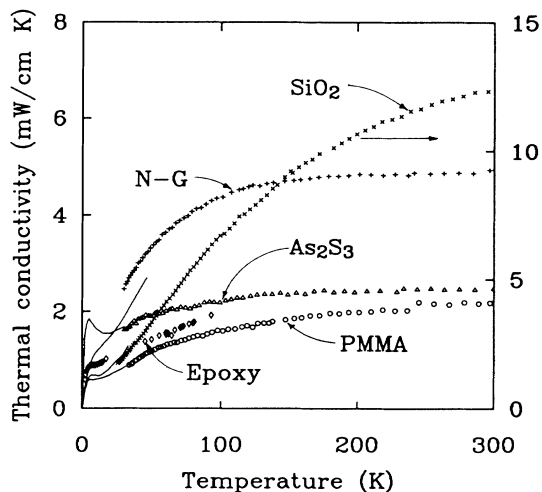


FIG. 5. Thermal conductivity of four of the amorphous solids described in the text and of an epoxy resin from Ref. 3. The data are plotted on a linear scale to test the prediction by Alexander *et al.* (Ref. 3) of a linear temperature dependence of the thermal conductivity. For silica, use thermal conductivity scale on the right-hand side of the graph: for all other solids, use the scale on the left.

where  $C$  is the specific heat (per volume). According to Slack, the minimum thermal conductivity is reached when the distance between collisions equals the wavelength of the Debye waves. In that case, the concept of a wave loses its significance. A more appropriate picture would be that of a volume element of the extent of one Debye wavelength passing on its energy to a neighboring volume element after one period of oscillation. Although the heat transport in this case resembles more a random walk, Eq. (7) still leads to a reasonable value of the thermal conductivity, as will be shown next for the high-temperature limit.

Instead of using Slack's choice of the minimum mean free path of one wavelength, we find that a minimum mean free path of one-half of a wavelength leads to agreement with our experimental results. In the high-temperature limit the wavelength is twice the average interatomic spacing, and therefore  $l$  equals the average interatomic spacing. We assume that each atom contributes  $3k_B$  to the specific heat (for PMMA and glycerol, we do not consider the H atoms separately, because of their very high vibrational frequencies; instead, we grouped them together with their neighboring C or O atoms to form a single, large "atom," see the caption of Table I). For the speeds of sound, the weighted averages of the low-temperature speeds of sound were chosen (Table I).

The calculated thermal conductivities are compared in Table I with the high-temperature limit of our experimental results. Except for PMMA the agreement is within 30% in all cases.

The picture that in glasses the heat transport in the limit of high temperatures occurs through a random walk of the thermal energy between atoms (or atomlike units) is

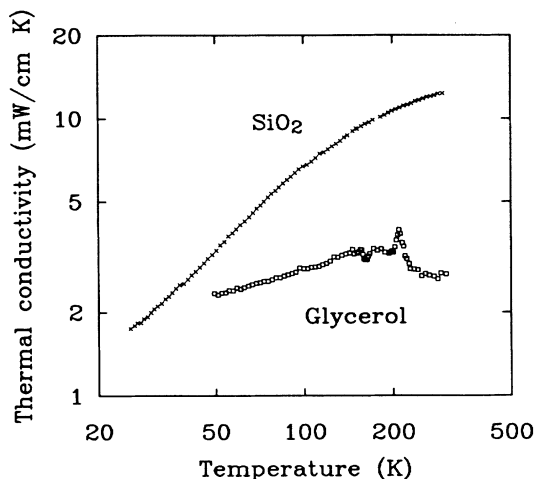


FIG. 6. Thermal conductivity of glycerol in the liquid and glass state. The glass transition occurs around 180 K; the anomaly just above 180 K is connected to the frequency dependence of the specific heat (Ref. 16) and is not due to an increase in the thermal conductivity. The  $\text{SiO}_2$  data are included to provide a scale for the magnitude of the thermal conductivity.

further supported by the observation that the thermal conductivity is nearly identical to that of the liquid. The thermal conductivity of liquids has often been modeled by the diffusion of heat from atom to atom.<sup>29</sup> Figure 6 shows that for glycerol the conductivity changes only by 10% as the glass melts. (Our data agree with those of Birge.<sup>16</sup> The peak in the data at 210 K is not a sudden change in conductivity, but results from the frequency dependence of the specific heat near the glass transition:<sup>16</sup> the glass transition temperature  $T_g$  is  $\sim 180$  K.)

Having seen that the picture of the minimum thermal conductivity leads to the right order of magnitude in the high-temperature limit, we now attempt to extend it to lower temperatures, using the temperature dependence of the specific heat as given by the Debye model. The results for  $\text{As}_2\text{S}_3$  and  $\text{SiO}_2$ , shown in Fig. 7, are typical. The calculated curves agree with the measured ones rather well to a temperature in the neighborhood of 50 K. For glasses with a larger Debye temperature  $\Theta_D$  the calculated minimum thermal conductivity drops off more rapidly with decreasing  $T$  than for those with a smaller  $\Theta_D$ , in agreement with the experiment. At temperatures below  $\sim 50$  K, the measured conductivity drops off considerably less rapidly than predicted by the model, which indicates that the scattering diminishes, i.e., the mean free path grows. It is known that below a few K,  $l$  is of the order of 100 wavelengths.<sup>2</sup>

## VII. CONCLUSION

We have developed a simple and reliable method for measuring the thermal conductivity of poor conductors of

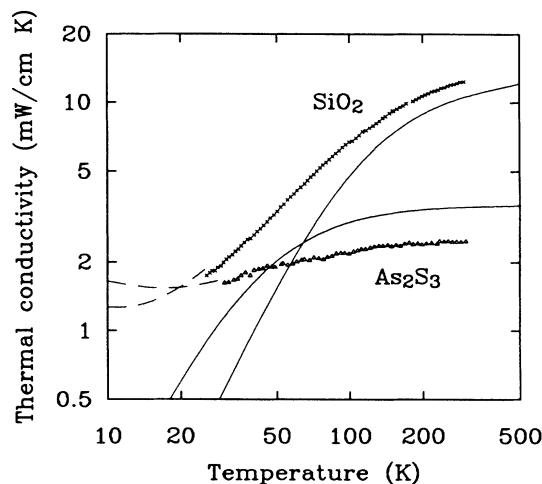


FIG. 7. Comparison of the calculated and measured values of the thermal conductivity of  $\text{SiO}_2$  and  $\text{As}_2\text{S}_3$ . Solid lines are the calculated minimum thermal conductivities, see text; data points are from this work. The dashed lines are low-temperature measurements as in Fig. 4.

heat at elevated temperatures. The method has been applied to a variety of amorphous solids. We have shown that the thermal conductivity above the plateau does not vary proportionately to the temperature, in disagreement with the recent prediction by Alexander *et al.* for heat transport by fractons. Above  $\sim 50$  K, the temperature variation of the thermal conductivities, and their absolute magnitudes, agree instead quite well with the minimum thermal conductivity as proposed by Slack. The origin of the strong scattering is still not understood.

*Note added in proof.* Freeman and Anderson<sup>30</sup> have recently reviewed the subject of heat transport in amorphous solids and have discussed several suggestions for a description of the thermal conductivity of  $\text{SiO}_2$  glass above 50 K.

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