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Thermal conductivity of amorphous solids

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The thermal conductivities of glassy solids, when scaled by material parameters, have a similar magnitude and temperature dependence, from the lowest temperatures yet measured, to the melt. Both empirical results and theoretical models are reviewed briefly. It is emphasized that the thermal conductivity of an amorphous solid is not understood in any temperature range.

I. INTRODUCTION

The thermal conductivity of amorphous dielectrics has not been explained. This paper is an attempt to further define the problem so as to provide an incentive for new investigations.

The paper is organized as follows. We present first in Sec. II a one-parameter scaling procedure which brings into common register the thermal-conductivity data of a variety of glassy solids, as demonstrated in Sec. III. The scaling parameter is an effective Debye temperature which may be estimated from the known atomic or molecular composition of a glass. Even if this scaling procedure should be devoid of physical meaning, it does facilitate the discussion of Sec. IV, which focuses on the three distinct temperature regimes revealed by the data.

II. SCALING PROCEDURE

It is well established that at tempertures of $T \lesssim 10$ K, thermal transport in glassy solids is provided by thermal phonons as in crystals.[1] Therefore the thermal conductivity may be expressed as

$$\kappa = \frac{1}{3} \int C(\nu,T) v^2 \tau(\nu,T)\, d\nu \ , \tag{1}$$

where $C(\nu,T)d\nu$ is the specific heat contributed by phonons within the frequency interval $d\nu$, v is an average

TABLE I. Parameters used to compute scaled thermal conductivities from Eq. (2). Numbers in parentheses represent the molecular weight M of a simple unit of the glass. The velocities are an average of the longitudinal and transverse modes.

Material	κ data	Density (kg/m^3)	Velocity (m/s)	Molecular weight	Debye temperature (K)	δ (nm)
SiO_2	a,b	2200[c]	4100[c]	$(60)\times 1$	342	0.58
B_2O_3	c,d	1800[c]	2060[e]	$(70)\times 1$	153	0.65
Sol-gel	f	1790[f]	2000[f]	$(60)\times 1$	156	0.62
Vycor	g	1770[f]	2320[f]	$(60)\times 1$	180	0.62
$3SiO_2Na_2O$	e	2440[e]	3490[e]	$(60.5)\times 1$	301	0.56
PET	h,i	1337[i]	1280[j]	$(193)\times 1$	61	1.01
PB	k	930[k]	1690[k]	$(54)\times 3$	76	1.07
PS	l	1050[m]	1670[n]	$(104)\times 3$	63	1.28
PMMA	o	1200[c]	1790[c]	$(100)\times 3$	71	1.21
Se	p,q	4300[r]	1170[r]	$(79)\times 3$	77	0.73
$Pd_{0.775}Si_{0.165}Cu_{0.060}$	s	10 560[s]	2040[s]	$(90.9)\times 1$	250	0.39
$Cu_{0.24}Zr_{0.76}$	t	6805[t]	1420[t]	$(84.6)\times 1$	154	0.44
$KBr_{0.50}KCN_{0.50}$	u,v	2150	1450	$(92)\times 1$	104	0.66
KH_2PO_4	w	2338[x]	2610[y]		385[y]	0.33

[a] Reference 4.
[b] Reference 5.
[c] Reference 6.
[d] Reference 7.
[e] Reference 8.
[f] Reference 9.
[g] Reference 10.
[h] Reference 11.
[i] Reference 12.
[j] Reference 13.
[k] Reference 14.
[l] Reference 18.
[m] Reference 19.
[n] Reference 20.
[o] Reference 21.
[p] Reference 15.
[q] Reference 16.
[r] Reference 17.
[s] Reference 24.
[t] Reference 25.
[u] Reference 22.
[v] Reference 23.
[w] Reference 26.
[x] Reference 27.
[y] Reference 28.

phonon velocity, and $\tau(\nu,T)$ is the lifetime of a phonon. At low temperatures it is reasonable to assume[2] a constant v and a density of phonon states quadratic in ν. Equation (1) may then be rewritten in the scaled, dimensionless form:

$$\kappa/K=(T/\Theta)^3\int_0^{\Theta/T}(k\Theta)(\tau/h)x^4e^x(e^x-1)^{-2}dx\ , \qquad (2)$$

where $K=4\pi k^3\Theta^2/h^2v$, $x=h\nu/kT$, k and h are the Boltzmann and Planck constants, and $\Theta=h\nu_{\max}/k$ is the classical Debye temperature. For a crystal, the Debye temperature is given by

$$\Theta=(hv/k)(3N/4\pi)^{1/3}=hv/k\delta\ , \qquad (3)$$

where N is the number of atoms per unit of volume and $\delta^3=(4\pi/3)(M/N_A\rho)$ is the volume of a cell defined with respect to the molecular weight M, Avogadro's number N_A, and the mass density ρ.

Equation (2) provides a satisfactory explanation for the phonon thermal conductivity of crystalline systems when anisotropy can be ignored. However, applying this equation to amorphous solids is questionable, since there is no well-defined procedure for computing[3] the Debye temperature Θ. We therefore use Θ as an adjustable scaling parameter by varying M, and hence the size of the smallest vibrational unit, to bring the data of different glasses into close register on a plot of κ/K versus T/Θ. Note that M is only varied by discrete increments of the mass of the vibrational unit.

In Table I we have assembled the parameters ρ and v for several glassy solids,[4–28] which are required to scale the measured thermal conductivities. The numbers shown in parentheses under the molecular-weight column represent the molecular weight M of a "simple" unit of the glass. As examples we have used the SiO_2 unit for vitreous silica, a $0.75M_{SiO_2}+0.25M_{Na_2O}$ unit for the sodium silicate glass, $0.775M_{Pd}+0.165M_{Si}+0.06M_{Cu}$ for glassy Pd-Si-Cu, and the monomer unit for the polymers. The multiplicative factor following these numbers in the column is the adjustable factor required to produce the Θ needed to bring, in Sec. III, the data into register with other data on a plot of κ/K versus T/Θ. Note that changing M by a factor of 3 changes Θ by $\approx 40\%$. Note also that the multiplicative factor for glassy Se is 3, whereas the molecular weight might be expected to be as large as six selenium atoms.[29]

The right-hand column of Table I lists the values of δ calculated using Eq. (3). It can be seen that these values vary little within each class of material, but more so between the different classes (i.e., for silicate glasses, $\delta\approx 0.6$ nm; for polymers, $\delta\approx 1.1$ nm; and for glassy metals, $\delta\approx 0.4$ nm). It may be significant that the adopted scaling procedure not only brings the thermal conductivities into close register, but also produces the above correlations in δ. In the following sections the Debye Θ of Table I is used as the explicit scaling parameter.

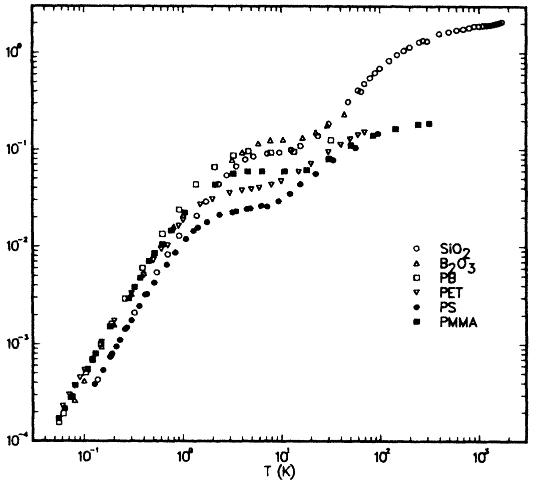

FIG. 1. Thermal conductivities κ of two oxide and four polymeric glasses, versus temperature T.

III. SCALED THERMAL CONDUCTIVITIES

The measured thermal conductivities of six glassy solids are shown in Fig. 1, while Fig. 2 demonstrates the scaling achieved[30] by using the Debye Θ of Table I to compute κ/K versus T/Θ. Figure 3 shows the reduced phonon conductivities for both normal and superconducting metallic glasses, and for the monatomic glass Se. The data for vitreous silica have been included to provide a reference. Figure 4 shows reduced conductivities for two low-density silicate glasses, namely Vycor and a sol-gel product, with vitreous silica again provided as a reference. Although the scaled conductivity, with no adjustment of M, of the sol-gel glass is very close to that of vitreous silica, the phonon velocity and mass density are quite different (see Table I). The presence of the large pores in Vycor significantly reduces the magnitude of the plateau. Also included in Fig. 4 is one example of a multicomponent oxide glass $3SiO_2Na_2O$. Other alkali glasses are found to scale to the same curve.

Figure 5 includes the scaled conductivities of two disordered crystals relative to that of vitreous silica. The

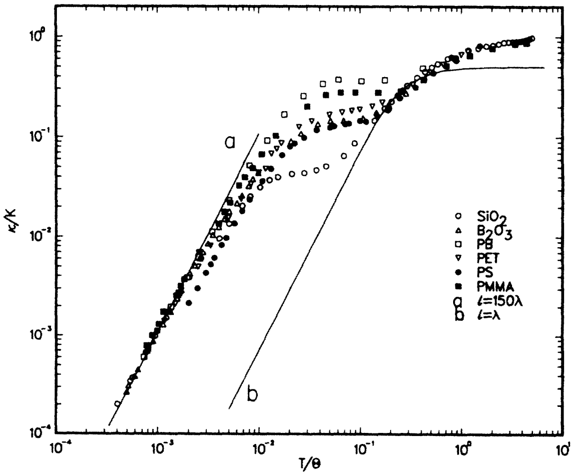

FIG. 2. Thermal conductivities for the same six amorphous solids as in Fig. 1, scaled as explained in the text. The solid lines here and in Figs. 3–5 were computed from Eq. (2) using either $l=150\lambda$ (line a) or $l=\lambda$ (line b).

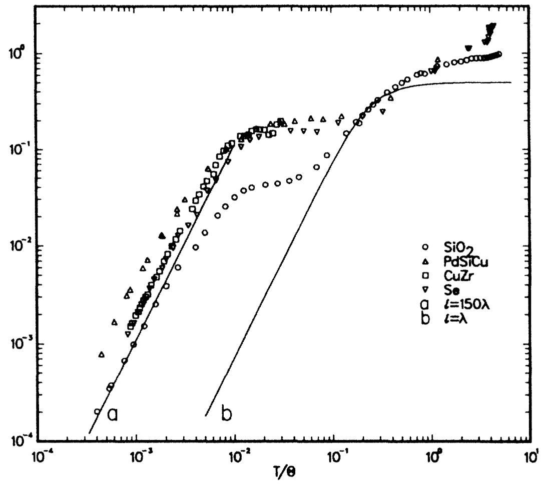

FIG. 3. Scaled *phonon* conductivities for normal and superconducting metallic glasses, together with glassy Se. Vitreous silica is included for reference.

$(KBr)_{0.5}(KCN)_{0.5}$ is a paraelectric crystal in which the CN molecules freeze into an "orientational glass" as the temperature is reduced. At $T \lesssim 1$ K, the $(KBr)_{0.5}(KCN)_{0.5}$, in addition to the thermal conductivity,[31] has a specific heat, a dielectric and ultrasonic response, and a thermal expansion similar to an amorphous solid.[32,33] The KH_2PO_4 is a ferroelectric which undergoes a disorder transition to an orientationally disordered paraelectric state for $T/\Theta \gtrsim 0.3$. The purpose of Fig. 5 is to emphasize that orientational disorder can produce a reduced thermal conductivity very similar to that found in positionally disordered, or amorphous, solids.[34]

Three distinct regions are evident in Figs. 2—5, with boundaries near $T/\Theta \approx 10^{-2}$ and 10^{-1}. Only in the region $10^{-2} \lesssim T/\Theta \lesssim 10^{-1}$ do the reduced conductivities of the glasses deviate appreciably from each other. This region is typically referred to as the "plateau." The following sections review what is known about each of the three temperature regions.

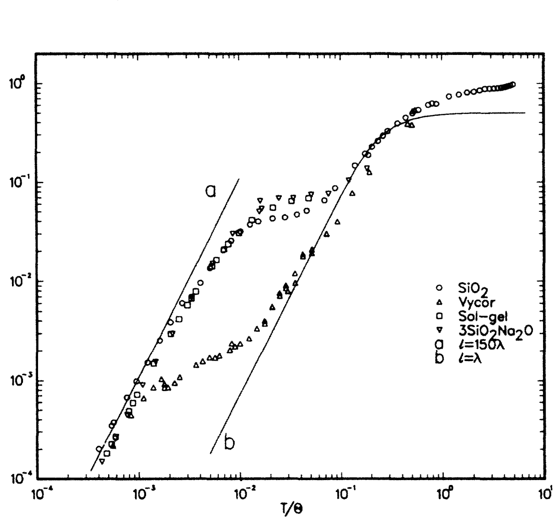

FIG. 4. Scaled thermal conductivities for two low-density silicate glasses and a sodium-silicate glass. Vitreous silica is included for reference.

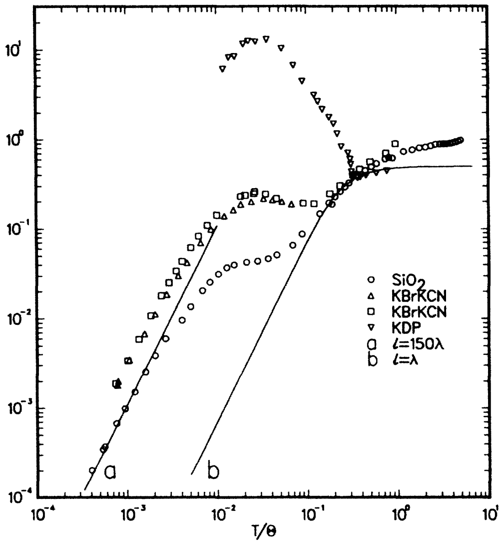

FIG. 5. Scaled thermal conductivities of two orientationally disordered crystals. Vitreous silica is included for reference. △, Ref. 22; □, Ref. 23.

IV. $T/\Theta \lesssim 10^{-2}$

The solid, straight line labeled *a* in Figs. 2—5 has a temperature dependence of $(T/\Theta)^2$. Although the actual temperature dependencies of several glasses appear to be slightly weaker than this, we will use line *a* as representative of κ/K for glasses within an accuracy of about a factor of 2.

Equation (2) is appropriate to a discussion of data for $T/\Theta \lesssim 10^{-2}$, since it has been established that phonons are responsible for heat transport in this temperature range.[1] Line *a* of Figs. 2—5 corresponds to

$$\tau v = l/\lambda = 150 \; , \tag{4}$$

where l is the mean free path of a phonon and λ is its wave length. It is emphasized that the relation $\tau v = l/\lambda \approx 150$ is to be viewed as a qualitative correlation. The value of τv (or κ/K), even for a given glass, can be modified somewhat by thermal treatment[35] or neutron irradiation.[36] Also, the correlation $\tau v = l/\lambda \approx 150$ is valid provided the phonon frequencies of interest are not very different from kT/h. Otherwise an explicit temperature dependence of the phonon scattering cross-section must be considered.[1] This detail will be discussed in Sec. IV.

Reynolds[37] had suggested that the empirical relation $\tau_{0.2\,\mathrm{K}} \approx 10^{-10}$ sec, for a fixed phonon frequency of $\approx 2\times 10^{10}$ Hz ($T \approx 0.2$ K), should apply to glasses. This is equivalent to stating that the product κv or τv should be a constant at $T = 0.2$ K and is consistent with the present scaling procedure. Reynolds[38] also suggested a general relationship given by $l_{0.2\,\mathrm{K}} \approx 4\times 10^{-8} T_g(m)$ where $l_{0.2\,\mathrm{K}}$ is measured again at a fixed frequency of $\approx 2\times 10^{10}$ Hz and T_g is the glass transition temperature. This correlation works well for oxide glasses,[39] but not for polymers or, especially, glassy metals.[38] These facts are now understood from Eq. (4) which indicates that, for dominant phonons,

$$l_{0.2\,\mathrm{K}} \approx v/\nu \approx 1.5\times10^{12} v/T \approx 8\times10^{12} v \ .$$

For oxide glasses, the phonon velocity is found experimentally to be essentially proportional to T_g, but this is not true for polymers or for glassy metals. Therefore $\tau\nu = l/\lambda \approx 150$ is a more general empirical feature of glasses at $T/\Theta \lesssim 10^{-2}$ than the relation $l_{0.2\,\mathrm{K}} \approx 4\times10^{-8} T_g$.

The origin of the phonon scattering represented by Eq. (4) has been attributed to several causes. Currently the generally held belief is that the phonons are scattered from localized two-level states (TLS) intrinsic to the glass and having broad distributions both in excitation energy and in equilibration time.[1] A model of the TLS assumes some entity of atomic size tunnels between two potential-energy wells producing a ground-state manifold consisting of the required two energy levels. This model predicts a phonon mean free path varying as

$$l/\lambda = \rho v^2 / f \gamma^2 \overline{P} \ , \tag{5}$$

where γ is a phonon-TLS coupling constant, $\overline{P}$ is the density of TLS, and f is a numerical constant. Combining the empirical Eq. (4) with the theoretical Eq. (5) indicates that $\rho v^2 = c \approx 150 f \gamma^2 \overline{P}$ where c is an average elastic constant. The tunneling model is phenomenological and provides no clue as to what may be tunneling, or why the phonon coupling is such that $\gamma^2 \overline{P} \approx c/150f$ for such a variety of glassy solids.

It has been suggested that the TLS may reside in regions of low mass density which are predicted by a free-volume model of the glass transition.[40] This model predicts $\overline{P} \propto V_F / T_g$ where V_F is the free volume frozen into the glass. Therefore $l/\lambda \propto (\overline{P})^{-1} \propto T_g / V_F$. Reynold's relation,[38] $l_{0.2\,\mathrm{K}} \approx 4\times10^{-8} T_g$, was initially cited as support for the free-volume origin of TLS. However, we noted above that the relation $l/\lambda \approx 150$ is more general, and does *not* depend explicitly on T_g. From this viewpoint, the free-volume model finds no support in thermal-conductivity measurements.[41]

The most intriguing aspect of the region $T/\Theta \lesssim 10^{-2}$ is that

$$\tau\nu = l/\lambda = \rho v^2 / f \gamma^2 \overline{P} \approx 150$$

for a wide range of disordered solids. It has been shown[42] that $\overline{P}$ and γ^2 can vary by a factor of 100, yet the product $\overline{P}\gamma^2/\rho v^2$ remains nearly constant. This fact continues to be an unanswered challenge to theory.

Some insight into the constancy of $l/\lambda = \rho v^2 / f \gamma^2 \overline{P}$ may be provided by a mean-field calculation of the interactions between tunneling sites in glasses. It is suggested in Ref. 43 that strain interactions between the sites contribute to the broad energy spectrum characteristic of the TLS. This leads to a density of TLS, $\overline{P}$, proportional to $\approx c_{44}/\gamma^2$ where c_{44} is an elastic constant. Therefore $\gamma^2 \overline{P}/\rho v^2$ is predicted to be a constant essentially independent of material parameters as observed experimentally. In brief, if there exists a significant strain interaction between the tunneling sites, the empirical relation $\tau\nu \approx 150$ might result.

V. $T/\Theta \gtrsim 10-1$

It is not known what type of excitation transports heat at $T/\Theta \gtrsim 10^{-1}$. Since the thermal conductivity changes very little upon increasing the temperature through T_g into the (supercooled) melt,[16] presumably the same excitations occur in glasses as in viscous liquids. The fact that several glasses have nearly the same reduced conductivity at $T/\Theta > 10^{-1}$ (see Fig. 2) suggests that the excitations may be essentially phonons. Indeed, molecular-dynamics calculations suggest that acoustic phonons can persist even into the melt.[44] We will pursue this possibility of phonon transport first.

It has been suggested that in disordered solids there should be a *minimum* acoustic-phonon thermal conductivity just as there is a minimum electrical conductivity in disordered metals.[45] However, this speculation has not provided as yet an explicit equation which may be compared with data. Two *ad hoc* suggestions have been made regarding the minimum mean free path of phonons for $T/\Theta > 10^{-1}$. The first assumes that l is a constant of order atomic distances.[46] This assumption, when used in Eq. (2), produces the dotted curve in Fig. 6 which corresponds to $l = \frac{3}{2}\delta$. The problem with this assumption is that near $T/\Theta \approx 10^{-1}$, $l/\lambda = \tau\nu \approx 10^{-2}$, and it is not clear that a phonon concept is valid for a phonon which only exists for a small fraction of its oscillatory period. To avoid this problem, the second assumption[47] has $l/\lambda = \tau\nu = 1$. This, when used in Eq. (2), produces the dashed curve of Fig. 6. This same curve is also repeated in Figs. 2–5 for convenience in comparison with other data.

The above computations are crude. It is still assumed that v is constant and that the density of phonon states is quadratic in ν. An obvious failure of the computations is the inability to reproduce in Fig. 6 the slowly increasing magnitude of κ/K with increasing T/Θ. Four suggestions have been advanced to account for this positive temperature dependence. One assumes thermal transport by

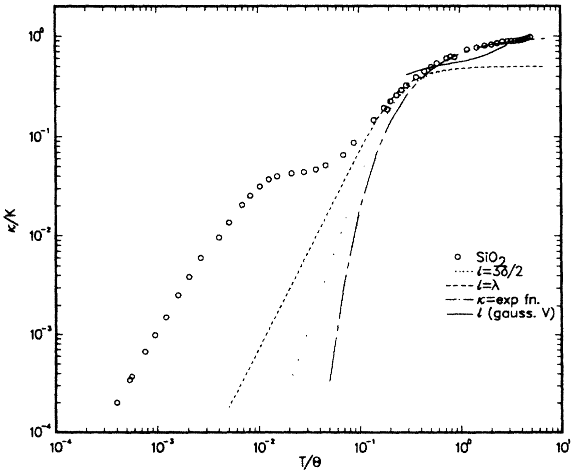

FIG. 6. Scaled thermal conductivity of vitreous silica together with four computed curves based on the four *ad hoc* assumptions discussed in the text. Above $T/\Theta \approx 1$, the dotted and dashed curves merge.

"optic" phonons,[48] but this would seem to be ruled out by the fact that materials as different as oxide glasses and polymers have similar reduced conductivities for $T/\Theta \geq 10^{-1}$. It has also been suggested that "quasilocal harmonic modes" can limit thermal transport at high temperatures.[49] This model predicts a linear temperature dependence for κ/K, but there is in Fig. 2 no significant temperature range (for $T/\Theta > 10^{-1}$) exhibiting a linear dependence. A third suggestion is that amorphous solids are intrinsically fractal. Therefore fractons, the excitations of a fractal network, may contribute to thermal transport. Recent calculations[50] indicate a linear temperature dependence for κ/K at $T/\Theta > 10^{-1}$. But, as stated above, a linear dependence is not a dominant feature of the measured thermal conductivity. The fourth suggestion has $\kappa \propto \exp(-T_0/T)$, where T_0 is a characteristic temperature perhaps related to low-energy excitations.[51] This assumption is shown by the dot-dashed line in Fig. 6 which corresponds to $\kappa/K = \exp(-0.4\Theta/T)$. Although the fit to the data is reasonable for $T/\Theta \gtrsim 1$, there is as yet no theoretical support for the exponential dependence on T^{-1}. Molecular-dynamics computations also suggest that a positive temperature coefficient of κ/K can arise in a disordered system,[52] but the nature of the "excitations" was not examined. In brief, we find no satisfactory explanation for the thermal conductivity in the temperature range $T/\Theta > 10^{-1}$.

We end this section with a precautionary note. The thermal conductivity near $T/\Theta \approx 1$ (which is within an order of magnitude of T_g for all materials) is very small. Therefore, even a limited number of phonons having large l could become important. For purpose of illustration we use the phonon mean free paths shown in Fig. 7. Curve *a* is related to scattering by TLS as discussed in Sec. IV. Curve *b* is strongly frequency dependent, is responsible for the plateau, and will be discussed in the next section. Curve *c* represents $l/\lambda = 1$. The complete curve *a-b-c* is appropriate to temperatures of $\lesssim 10$ K. As mentioned in Sec. IV, curve *a* is temperature dependent and, empirically,[53] l is found to exhibit a pronounced frequency-dependent minimum at $T \approx 60$ K. This minimum is usually attributed to structural relaxation. Therefore, at $T \approx 300$ K, the low-frequency phonons can have larger mean free paths as indicated by line *d*. This large mean free path should be incorporated into Eq. (2) for calculations at $T/\Theta > 10^{-1}$.

The acoustic attenuation (l^{-1}) has been measured as a function of temperature for several glasses.[54,55] The observed high-temperature behavior for vitreous silica can be fit roughly using a Debye relaxation,[53] $l^{-1} = A\nu^2\tau_1(1+\nu^2\tau_1^2)^{-1}$, where A depends on the coupling to whatever centers are responsible for the energy absorption, and τ_1 is the equilibration period of the scattering center. For thermal activation over a barrier of height V, $\tau_1 = \tau_0 \exp(V/kT)$. To fit the data properly, a Gaussian distribution of barrier heights had to be assumed. Using the parameters of Ref. 53, and therefore using the curve *d-b-c* of Fig. 7, produces the solid curve in Fig. 6. This curve has a positive temperature dependence and a magnitude similar to that of the measured conductivity. It should be noted that the parameters used for this calculation are taken directly from Ref. 53; no fitting to the conductivity data has been attempted. The purpose of this exercise has been to point out that some fraction of κ/K at $T/\Theta > 10^{-1}$ might be attributed to relatively low-frequency phonons. This qualitative conclusion is also predicted by the theory of Ref. 49.

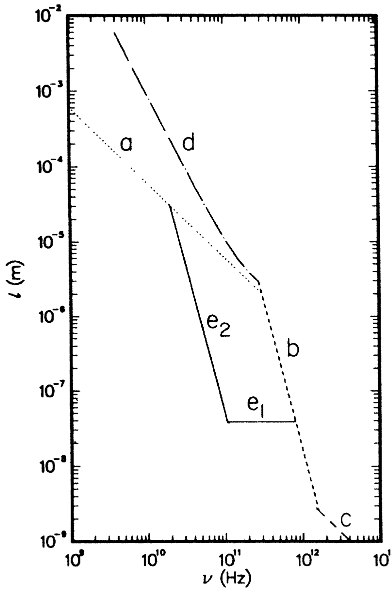

FIG. 7. Phonon mean free path l as a function of frequency ν, the curves are explained in the text.

VI. $10^{-2} \lesssim T/\Theta \lesssim 10^{-1}$

We return now to the plateau, the crossover region between the low-temperature and high-temperature behavior. Empirically we observe that for the nonmetallic glasses, the magnitude of the (scaled) plateau decreases with increasing values of Θ. That is, the thermal conductivity κ at the plateau scales roughly as the inverse of δ.

The available evidence indicates that phonons carry the heat in the plateau region,[56] and that the mean free path of these phonons has a strong frequency dependence[57] of $l \propto \nu^{-n}$, with $n \approx 3$–4, as indicated by curve *b* in Fig. 7. This strong frequency dependence precludes the use of the "dominant phonon" approximation, i.e., the assumption that the phonons important to thermal transport have $h\nu \approx kT$. The physical consequence is that, at $T/\Theta \approx 10^{-1}$, the important phonons are of long wavelength, that is, they have a much lower characteristic temperature of $T/\Theta \approx 10^{-2}$.

Many origins of the strong phonon scattering have been suggested,[1,49,50,58] but no model has gained general acceptance. In particular, the explicit relation $l \approx (4 \times 10^{24}/m^3)\lambda^4$ has been shown[59] to account for the plateau in many amorphous solids, yet this dependence appears not to arise from "Rayleigh" scattering[39] from small density or concentration fluctuations of diameter $D < \lambda$.

Rayleigh scattering from *real* voids of diameter $D \approx 50$ Å can be used to scatter phonons strongly and thereby lower the plateau, as shown in Fig. 4 for porous Vycor. It is interesting to note that the assumption $l/\lambda = \tau\nu = 1$, introduced to explain data for $T/\Theta > 10^{-1}$ (see Sec. V), provides a qualitative fit for Vycor down to $T/\Theta \approx 10^{-2}$ as shown by curve b of Fig. 4.

The plateau for Vycor (and for other porous glasses) can be understood[60] in terms of the phonon mean free path represented by the solid line in Fig. 7. Introduction of holes produces a fixed l provided the phonon frequency is large enough that $\lambda < D$. This is indicated by curve e_1 of Fig. 7. At lower frequencies (curve e_2) Rayleigh scattering sets in and l increases until it is again dominated by the TLS responsible for line a. Therefore the high-temperature edge of the plateau occurs near $\lambda \approx D$ for porous glasses. This regime of phonon "localization" has been studied theoretically in some detail.[61,62]

The fracton model (Sec. V) may apply to low-density glasses such as the sol-gel silica[9] of Fig. 4. This model predicts $l \propto \nu^{-4}$ and therefore a plateau in κ/K. The plateau represents, for a low-density glass, a crossover from a phonon regime to a fracton regime with increasing temperature.[63] The application of the fracton model to explain the plateau for *bulk* glasses is still highly speculative.

Some models[49,61,63] which explain the plateau in κ/K also account for a maximum in C/T^3 near 10 K, where C is the specific heat of the glass. Several authors[58,64] have noted an apparent empirical correlation between the magnitude and temperature of the plateau and the magnitude and temperature of the peak in C/T^3. The implication is that the excitations responsible for the maximum in C/T^3 are responsible for the strong phonon scattering which produces the plateau.

VII. CONCLUSIONS

The thermal conductivities of glassy materials may be brought into common register both at low temperature and at high temperature by a one-parameter scaling procedure. The scaling parameter is the Debye temperature Θ, or a closely related property, which may be estimated to within $\approx 40\%$ from the known atomic or molecular units of the glass.

At $T/\Theta \lesssim 10^{-2}$ phonons transport the heat, and the phonon scattering is such that $\tau\nu = l/\lambda \approx 150$ for any type of glass.[65] Although this scattering is attributed to two-level states, there has been no microscopic explanation as to why the relation $\tau\nu \approx 150$ should apply to such a varied spectrum of glasses.[66]

In the plateau $10^{-2} \lesssim T/\Theta \lesssim 10^{-1}$, phonons again transport the heat, but the mean free path of the phonons rapidly decreases with increasing frequency. The scattering mechanism responsible for this frequency dependence has not been established.

At $T/\Theta > 10^{-1}$ it is not even clear what excitations are responsible for thermal transport. The observation that the thermal conductivity can be scaled by Θ will provide a test of proposed conductivity mechanisms.

ACKNOWLEDGMENTS

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