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# Low-temperature thermal conductivity and acoustic attenuation in amorphous solids

Robert O. Pohl*
*Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14853*

Xiao Liu
*SFA, Inc., Largo, Maryland 20774*

EunJoo Thompson
*Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14853*

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In order to test whether the low-energy excitations explored extensively in amorphous solids are indeed universal, all measurements published on the low-temperature thermal conductivity and acoustic attenuation in these solids have been reviewed, on a total of over 60 different compositions. The ratio of the phonon wavelength $\lambda$ to the phonon mean free path $\ell$ has been found to lie between $10^{-3}$ and $10^{-2}$ in almost all cases, independent of chemical composition and frequency (wavelength) of the elastic waves, which varied by more than nine orders of magnitude in the different experiments. When the data were fitted with the tunneling model, which is based on the assumption of atomic or molecular tunneling states with a certain spectral distribution, the tunneling strength $C$, which describes their coupling to the lattice, was found to range from $10^{-4}$ to $10^{-3}$ in almost all cases. The only exceptions reported so far are certain films of amorphous silicon, germanium, and carbon. In these films, low-temperature acoustic attenuations over two orders of magnitude smaller have been observed compared to all other amorphous solids. Another remarkable observation is that a large number of disordered crystals, and even a thermally equilibrated quasicrystal, have low-energy lattice vibrations that are quantitatively indistinguishable from those of amorphous solids. Their tunneling strengths also range from $10^{-4}$ to $10^{-3}$. These measurements have also been reviewed. It is concluded that the absence of long-range order is neither sufficient nor necessary for the existence of the low-energy excitations.

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## I. INTRODUCTION

In pure crystalline solids, the lattice vibrations can be completely described as collective excitations or waves (Born and von Karman, 1912; Debye, 1912). Lattice defects, like impurities, lead to local changes of these vibrations, referred to as defect modes (Wallis, 1968; Barker and Sievers, 1975), which are specific to the type and concentration of the defects. In amorphous solids, a different kind of defect mode has been found, which leads to characteristic changes in their low-temperature thermal conductivity and specific heat, including short- and long-term thermal relaxation, and also in their real and imaginary elastic and dielectric functions. These properties have been studied extensively since they were first reported 30 years ago (Zeller and Pohl, 1971). A phenomenological model has been used very successfully for the description of these impurity modes (Anderson *et al.*, 1972; Phillips, 1972). In this so-called *tunneling model*, it is postulated that some atoms or groups of atoms have two equilibrium positions between which they can tunnel. In contrast to the tunneling states known in doped crystals (Narayanamurti and Pohl, 1970), their spectral distribution in amorphous solids is postulated not to be discrete, but broad.

The objective of this paper is not a review of the experimental exploration of these modes, nor their discussion in terms of the tunneling model, which has been done extensively in the past (Phillips, 1981b, 1987; Hunklinger and Raychaudhuri, 1986; Berret and Meissner, 1988; Esquinazi and König, 1998). Instead we shall emphasize one particularly puzzling aspect of these modes, which is that they appear to be not only qualitatively, but even quantitatively very similar, independent of chemical composition or preparation of the amorphous solids (Berret and Meissner, 1988; Leggett, 1991). This quantitative similarity, which is also sometimes referred to as *universality*, is not part of the tunneling model and is undoubtedly the most remarkable property

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*Electronic address: pohl@ccmr.cornell.edu

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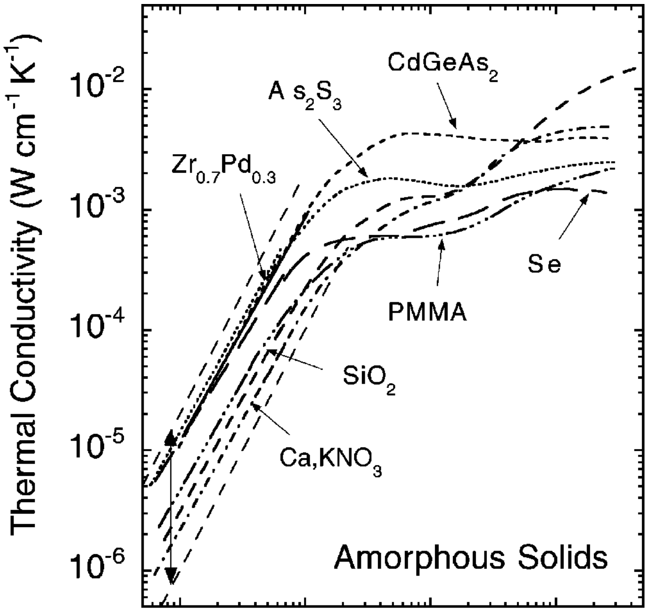

FIG. 1. Thermal conductivity of several amorphous solids (Cahill and Pohl, 1988a). The conductivities of all glasses measured to date below 1 K lie in the range spanned by the two dashed straight lines shown here, separated by the double arrow, which we call the glassy range.

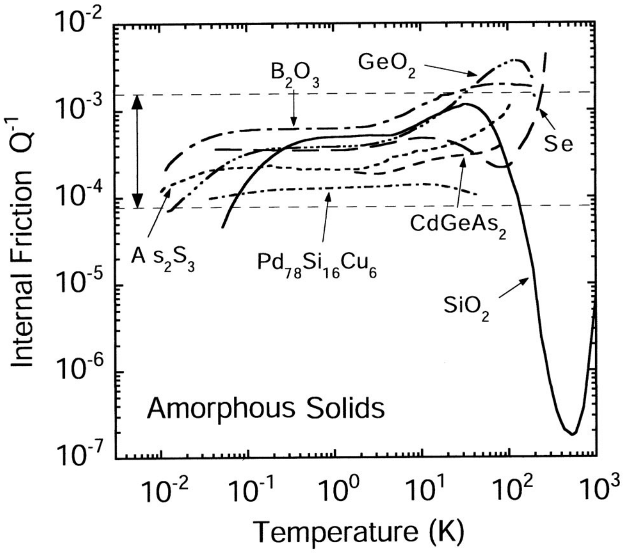

FIG. 2. Internal friction of several amorphous solids (Topp and Cahill, 1996). Between 0.1 and 10 K, the internal friction is nearly independent of temperature and measuring frequency. Within this temperature range, the magnitude of the internal friction for all glasses falls within about a factor of 20 as shown here by the dashed straight lines and the double arrow, called the glassy range, except for some *a*-Si films that are mentioned later. For a discussion of the dropoff below ~0.1 K, see the text.

of these impurity modes. In spite of much theoretical effort, which focuses on the interaction between (tunneling) defects (Yu and Leggett, 1988; Yu, 1989; Coppersmith, 1991; Leggett, 1991; Burin *et al.*, 1998) or between soft potentials (Parshin, 1994a), universality remains poorly understood. Thus the purpose of this paper is to provide a critical review of the experiments on which this claim of universality is based, and through this to contribute to our understanding of the physical nature of the defect modes. We shall concentrate on measurements of low-temperature thermal conductivity and sound attenuation, including internal friction. These measurements permit the most direct test of universality, and they illustrate a particularly striking property of these impurity modes even without the use of any particular theoretical model.

Without specifying at this point exactly what we mean by universality (this will be done at the end of Sec. III), we will say right now that exceptions have been observed. In addition, an important point to be stressed is that the same remarkable defect modes have also been shown to exist in a large number of disordered crystals, including at least one quasicrystal. These measurements will also be reviewed. We shall reach the conclusion that the absence of long-range order, which characterizes amorphous solids, is neither sufficient nor necessary for these excitations to exist.

## II. EXAMPLES OF EXPERIMENTAL RESULTS

Before proceeding, we shall show a few examples of the experimental observations to be reviewed. The characteristic thermal conductivity of bulk amorphous dielectric solids (including that of an amorphous metal in its superconducting state) is shown in Fig. 1, taken from Cahill and Pohl (1988a). Below ~1 K, the thermal conductivity varies nearly as the square of the temperature ($T^2$), and above ~100 K it approaches a temperature-independent value, called the minimum thermal conductivity, which has been reviewed previously (Medwick and Pohl, 1997; Pohl, 1998). In the present review, we concentrate on the temperature range below 1 K. Note the two dashed lines in Fig. 1, which are connected by a double arrow. They are proportional to $T^2$ and indicate the range spanned by practically all amorphous solids measured to date, which will be reviewed here. The two lines are spaced by a factor of 20, which we call the "glassy range," and present the first example of the universality to be discussed.

As examples of acoustic attenuation, Fig. 2 shows the internal friction $Q^{-1}$ of amorphous solids, again including an amorphous metal (this one in its normal state; Topp and Cahill, 1996). Above 10 K, the internal friction depends strongly on the chemical composition. Below that temperature, it approaches a temperature-independent value. The dropoff at the lowest temperature depends on the frequency of measurement, occurring at lower temperatures as the measuring frequency decreases, as is illustrated in Fig. 3. This dropoff can be explained in the tunneling model, together with the plateau (Jäckle, 1972). In this review, we shall concentrate on the plateau and ignore measurements of acoustic attenuation by resonant scattering, referring the reader to

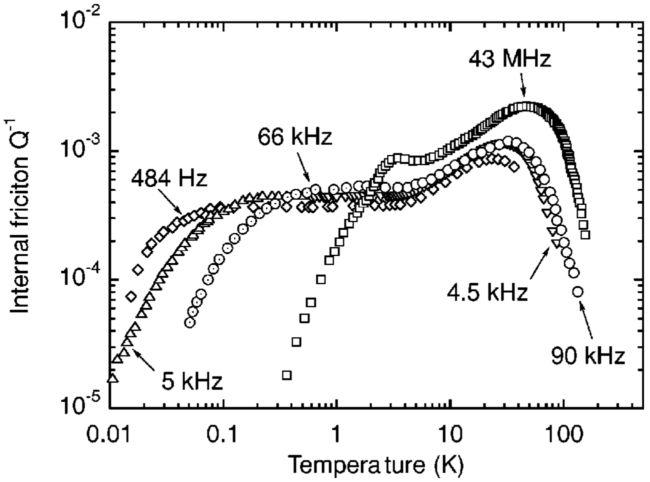

FIG. 3. Internal friction of $a$-$SiO_2$ measured at different frequencies, after Topp and Cahill (1996). The 484-Hz data (flexural vibration) are from Raychaudhuri and Hunklinger (1984). The 5-kHz and 4.5-kHz data, measured in torsion with double-paddle oscillators, are from Classen *et al.* (2000) and White and Pohl (1995), respectively. The 66-kHz and 90-kHz data, measured in torsion, are from Van Cleve *et al.* (1994) and Cahill and Van Cleve (1989), respectively. The 43-MHz data, measured as longitudinal acoustic attenuation, are from Bartell and Hunklinger (1982). The low-temperature rolloff has been found to occur at temperatures proportional to approximately the one-third power of the measuring frequency (in agreement with the tunneling model). The plateau value below 10 K is almost independent of the measuring frequency over five orders of magnitude. A similar weak dependence on the measuring frequency has also been observed in the polymer PMMA; see Fig. 7 of Topp and Cahill (1996).

two reviews that have dealt thoroughly with this subject, those of Hunklinger and Raychaudhuri (1986) and Phillips (1987). Figure 3 also shows that the magnitude of the internal friction plateau is very little affected by the frequency of the measurement, a fact to be emphasized in the following.

Returning to Fig. 2, note again the two dashed lines and the double arrow marking the range of the height of the plateau observed in all amorphous solids in which attenuation has been studied to date (the connection between ultrasonic attenuation and internal friction will be explained below). The range spanned is again a factor of 20, the same as that found in thermal conductivity. Some recent measurements of the low-temperature internal friction of thin films of amorphous silicon, germanium, and carbon ($a$-Si, $a$-Ge, and $a$-C) have revealed plateau values below this glassy range, by as much as a factor of 100, as is shown in Fig. 4 for several $a$-Si films (Liu and Pohl, 1998, Fig. 10). These data will be discussed separately from those obtained on the other amorphous solids.

Thermal conductivity and internal friction very similar to those of amorphous solids have also been observed in a number of disordered crystalline solids. Figure 5 shows the thermal conductivity of single crystals of chemically pure $BaF_2$ and of the composition $(BaF_2)_{1-x}(LaF_3)_x$ (Cahill *et al.*, 1992). With increasing proportion of $LaF_3$, up to $x=0.46$, the thermal conductivity $\Lambda$ decreases and approaches that of amorphous $SiO_2$ ($a$-$SiO_2$). Such a glasslike thermal conductivity in chemically disordered crystals, although observed in many cases as reviewed below, is nonetheless not the rule. Figure 5 shows one example, crystalline $(BaF_2)_{1-x}(SrF_2)_x$, in which the thermal conductivity remains that typical of crystals even for large $x$ ($x=0.3$ in this case).

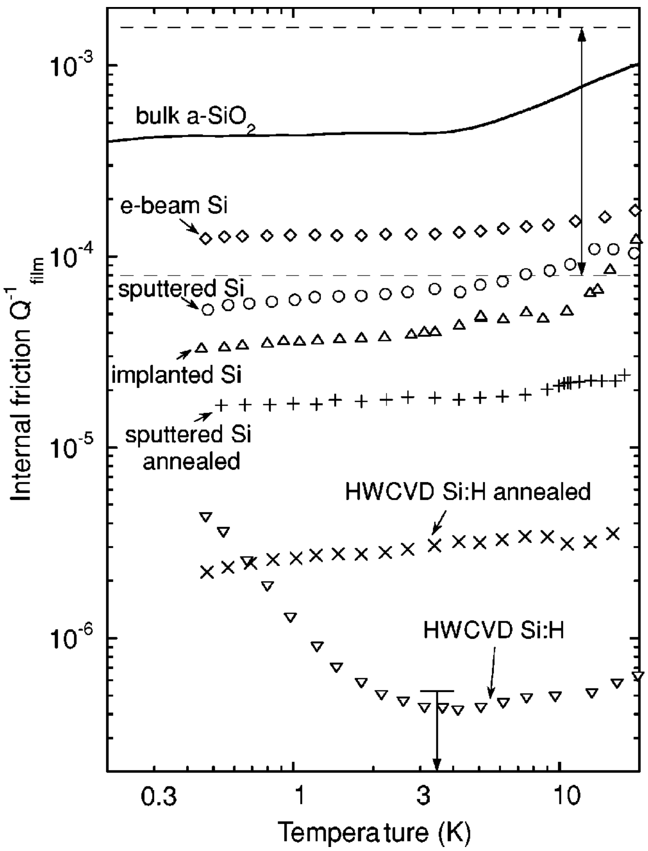

FIG. 4. Internal friction of selected pure and hydrogenated $a$-Si films. For details see Liu and Pohl (1998). The double arrow and the dashed lines shown are the same as those in Fig. 2. For the film with the smallest internal friction, the data ($\nabla$) indicate an upper limit only, as indicated by the single arrow, because of uncertainty of the background damping (of the substrate carrying the film). The rapid rise of $Q_{film}^{-1}$ below 3 K for this film is caused by substrate contamination.

In Fig. 6 we show the effect of chemical disorder on the internal friction of mixed crystals of $(CaF_2)_{1-x}(LaF_3)_x$ (Topp *et al.*, 1999). The smallest internal friction observed for the pure $CaF_2$ crystal, $x=0$, is caused by clamping losses (the internal friction of the pure crystal is orders of magnitude smaller below 10 K). At $x=0.13$ and $x=0.26$, the internal friction plateau characteristic of amorphous solids is observed. It is, significantly, independent of $x$ in this compositional range. Note also the same dropoff at the lowest temperatures as observed in amorphous solids.

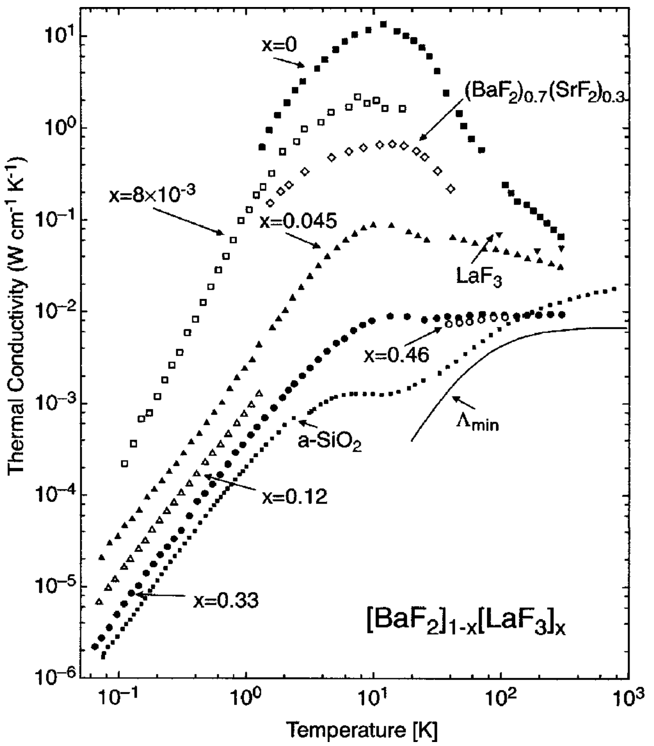

FIG. 5. Thermal conductivity of $(\mathrm{BaF_2})_{1-x}(\mathrm{LaF_3})_x$ (Cahill *et al.*, 1992). The pure $\mathrm{BaF_2}$ ($x=0$) has a thermal conductivity typical of perfect crystals (a collection of perfect-crystal data can be found in Pohl, 1998). The addition of $\mathrm{LaF_3}$ leads to a thermal conductivity approaching that of an amorphous solid ($a$-$\mathrm{SiO_2}$). Note that the conductivities for $x=0.33$ and 0.46 are very close, approaching the minimum thermal conductivity $\Lambda_{\mathrm{min}}$ at high temperatures (Cahill *et al.*, 1992). Undoped $\mathrm{LaF_3}$ again has a large thermal conductivity. The addition of a large concentration of (divalent) $\mathrm{SrF_2}$ to $\mathrm{BaF_2}$ has a relatively small effect on the thermal conductivity. A glasslike thermal conductivity is not reached.

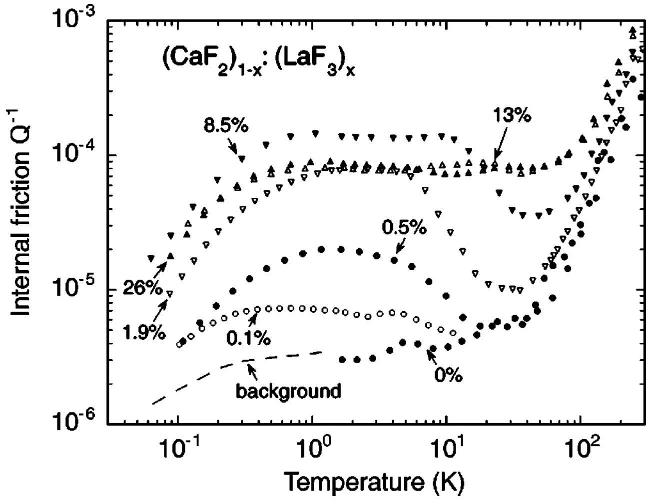

FIG. 6. Internal friction of pure $\mathrm{CaF_2}$ and of $(\mathrm{CaF_2})_{1-x}(\mathrm{LaF_3})_x$ mixed crystal samples for six different concentrations, measured in torsion near 90 kHz, after Topp *et al.* (1999). The data points labeled 0% are measurements of the background by a technique using a pure $\mathrm{CaF_2}$ crystal as the sample, while the dashed line labeled "background" was measured with a quartz crystal in its place. The internal friction of the samples is determined by taking the difference between the data points and the background. A broad, temperature-independent internal-friction plateau is observed only for the two largest values of $x$.

## III. ANALYSIS OF DATA

### A. Phonon mean free path in amorphous solids

Since both thermal conductivity and acoustic attenuation measure the damping of elastic waves, we shall start the review by comparing the ratios of the phonon wavelength $\lambda$ to the phonon mean free path $\ell$ observed in these experiments. This quantity can be determined directly from measurements of thermal conductivity, ultrasonic attenuation, or internal friction, without using any model of what causes the damping of the elastic waves.

In the gas-kinetic picture, the thermal conductivity $\Lambda$ can be written as

$$\Lambda=\frac{1}{3}C_{\mathrm{D}}v\ell(T), \tag{1}$$

where $C_{\mathrm{D}}$ is the Debye specific heat per volume of the heat-carrying phonons, which travel with the average Debye velocity $v$, undergoing collisions after traveling the mean free path $\ell(T)$. For a recent demonstration that the heat in amorphous solids is indeed carried by Debye waves below 1 K, see Vu *et al.* (1998). At a given temperature, the heat is carried predominantly by those waves which contribute the most to the specific heat at that temperature. In the dominant phonon approximation, it is assumed that these phonons carry all the heat, and so one can convert $\ell(T)$ from a function of temperature to $\ell(\nu_{\mathrm{dom}})$, a function of the dominant phonon frequency, where $\nu_{\mathrm{dom}}$ is (Klitsner and Pohl, 1987)

$$\nu_{\mathrm{dom}}=4.25\frac{k_BT}{h}=90[\mathrm{GHz\,K^{-1}}]\cdot T, \tag{2}$$

where $k_{\mathrm{B}}$ and $h$ are Boltzmann's and Planck's constants, respectively. At 1 K, for example, $\nu_{\mathrm{dom}}=90$ GHz. This conversion factor is empirical and has worked well for the description of resonant phonon scattering by localized defect modes, as described, for example, in Narayanamurti and Pohl (1970). It is, nevertheless, somewhat uncertain and depends on details of the phonon-scattering process. We shall express $\ell(T)$ as a function of the dominant phonon wavelength using

$$\lambda_{\mathrm{dom}}=\frac{v}{4.25(k_{\mathrm{B}}T/h)}. \tag{3}$$

Experimentally, the thermal conductivity of amorphous solids below a few K is found to vary as $T^2$ with a prefactor $\beta_0$ defined below [see Fig. 1; the best fits are usually achieved with an exponent between 1.8 and 2.0 (Lasjaunias *et al.*, 1975; Raychaudhuri *et al.*, 1980; Raychaudhuri and Pohl, 1982), but we shall ignore this variation in the following],

$$\Lambda=\beta_0T^2=\frac{1}{3}cT^3v\ell, \tag{4}$$

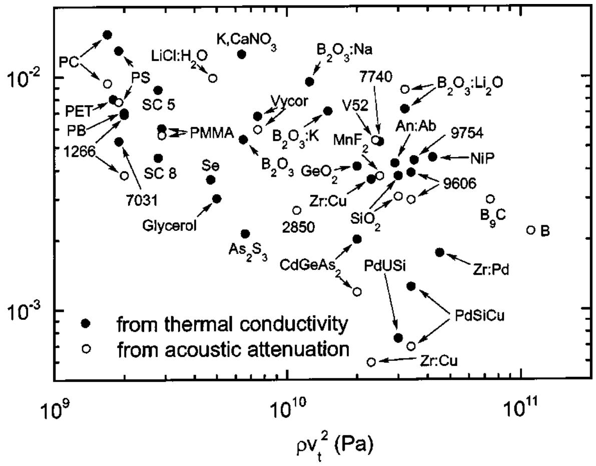

FIG. 7. $\lambda/\ell$ obtained using transverse waves from thermal conductivity and acoustic attenuation measurements for all amorphous solids measured, except $a$-Si, $a$-Ge, and $a$-C. The quantity $\lambda/\ell$ is independent of the material and falls within approximately a factor of 20, independent of the elastic modulus $\rho v_t^2$, which varies over a factor of 50 for different materials.

where $c$ is the prefactor in the Debye specific heat per volume given by

$$c=\frac{2}{5}\frac{k_B^4}{\hbar^3}\frac{\pi^2}{v^3} \tag{5}$$

and $\hbar=h/2\pi$. Combining Eq. (4) with Eq. (5), we obtain

$$\ell=\frac{3\beta_0}{cvT}=\frac{15}{2}\frac{\hbar^3\beta_0 v^2}{k_B^4\pi^2 T}. \tag{6}$$

Upon introducing $\lambda_{\mathrm{dom}}$, we obtain

$$\frac{\lambda_{\mathrm{dom}}}{\ell}=\frac{2}{15}\frac{k_B^4\pi^2}{\hbar^3\beta_0 v\cdot 4.25(k_B/h)}$$

$$=0.46[\mathrm{J\,K^{-3}\,s^{-2}}]\cdot\frac{1}{\beta_0 v}. \tag{7}$$

Thus $\lambda_{\mathrm{dom}}/\ell$ is obtained directly from the experimentally determined $\beta_0$ and the Debye velocity. It does not depend on phonon frequencies.

Measurement of the ultrasonic (energy) attenuation $\alpha$ leads immediately to the mean free path

$$\ell=\alpha^{-1}, \tag{8}$$

and thus

$$\frac{\lambda}{\ell}=\frac{v}{\nu}\alpha, \tag{9}$$

where $\nu$ is the frequency and $\lambda$ the wavelength of the acoustic wave.

Finally, from the internal friction $Q^{-1}$, $\ell$ is obtained through

$$Q^{-1}=\frac{v}{2\pi\nu}\alpha=\frac{v}{2\pi\nu}\frac{1}{\ell}=\frac{1}{2\pi}\frac{\lambda}{\ell}, \tag{10}$$

and from this

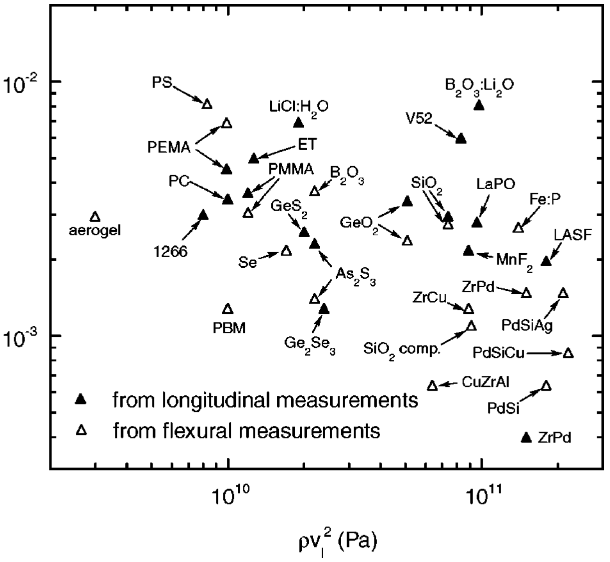

FIG. 8. $\lambda/\ell$ obtained using longitudinal waves from flexural and acoustic attenuation measurements for all amorphous solids, except $a$-Si, $a$-Ge, and $a$-C. The $\lambda/\ell$ of an over 90% porous aerogel (not shown) has the same value as the aerogel shown, although the elastic modulus $\rho v_l^2$ would lie outside the range plotted by a factor of 100 (see Table I). Thus the quantity $\lambda/\ell$ is independent of the material and falls within approximately a factor of 20, independent of the elastic modulus $\rho v_l^2$, which varies over a factor of $10^4$ for different materials.

$$\frac{\lambda}{\ell}=2\pi Q^{-1}=2\pi\frac{v}{\omega}\alpha, \tag{11}$$

where $\omega=2\pi\nu$. If the ultrasonic attenuation is measured in db/cm, as is often done, the connection between its value, called $\alpha_{\mathrm{db}}$, and the internal friction $Q^{-1}$ is given by

$$Q^{-1}=0.23\frac{v}{\omega}\alpha_{\mathrm{db}}, \tag{12}$$

and from this,

$$\frac{\lambda}{\ell}=2\pi\cdot 0.23\frac{v}{\omega}\alpha_{\mathrm{db}}. \tag{13}$$

Equations (7) and (11) can be used to describe the phonon scattering observed through thermal conductivity and acoustic attenuation, respectively. In the following, we shall drop the subscript "dom" from $\lambda_{\mathrm{dom}}/\ell$ for convenience and call $\lambda/\ell$ the *relative inverse phonon mean free path* from either thermal conductivity measurements or acoustic measurements.

Figure 7 contains $\lambda/\ell$ obtained from the attenuation of transverse waves (the acronyms used for the identification of the solids will be explained later in the tables). Since heat is carried predominantly by transverse waves, $\lambda/\ell$ obtained from thermal conductivity in the temperature range where $\Lambda=\beta_0 T^2$ can be directly compared to the acoustic results, as is done in Fig. 7.

Figure 8 summarizes results obtained for longitudinal waves. Here we include results obtained on Young's-

TABLE I. Si- and Ge-based glasses. This table and the following four contain information on all amorphous solids found for which either the pre-factor $\beta_0$ of the low-temperature $T^2$ thermal conductivity, or the temperature-independent acoustic attenuation $\alpha$, or the internal friction plateau $Q_0^{-1}$ has been measured. No effort has been made to cite all references for a particular substance. Instead, we have tried to select those which contain the most complete information and which may also refer to earlier work. In Tables I–IV, column 1 lists the names of the glasses as used in Figs. 7, 8, 9, and 10 with their exact compositions given in the notes; column 2 is the mass density $\rho$; columns 3 and 4 are the longitudinal and transverse speeds of sound $v_i$, with all references given throughout the tables; columns 5 and 6 are the longitudinal and transverse elastic moduli $\rho v_i^2$; columns 7–10 are the tunneling strengths $C_{\mathrm{t,l}}$ determined from the plateau value of $\alpha$ or $Q_0^{-1}$ of longitudinal, Young's-modulus (flexural), and transverse waves [using Eq. (15)], or from $\beta_0$ [using Eq. (19)]. Solids for which the speeds of sound are not known have been listed in the notes. In order to keep these tables from becoming too crowded, we did not list the measured quantities $\beta_0$, $\alpha$, and $Q_0^{-1}$ or the quantities $\lambda/\ell|_{\mathrm{t,l}}$ derived from them [using Eqs. (16) and (20), respectively]. They can, of course, be readily rederived from the quantities listed here.

| | $\rho$ (g/cm$^3$) | $v$ ($10^5$ cm/s) | | Mod. ($10^{10}$ Pa) | | $C_\mathrm{l}$ ($10^{-4}$) | | $C_\mathrm{t}$ ($10^{-4}$) | |
|---|---|---|---|---|---|---|---|---|---|
| Material | $\rho$ | $v_\mathrm{l}$ | $v_\mathrm{t}$ | $\rho v_\mathrm{l}^2$ | $\rho v_\mathrm{t}^2$ | ac. | flex. | $\Lambda$ | ac. |
| $SiO_2$ | 2.203[1] | 5.8[1,2] | 3.7[1,2] | 7.4 | 3.0 | 3.0[3.4] | 2.8[5,6] | 3.0[7,8] | 3.1[9,10] |
| Same, compacted[11] | 2.45[12] | 6.08[13] | | 9.1 | | | 1.1[12,14] | | |
| Same, porous[15] | 1.570[16] | 3.562[17,18] | 2.188[17,18] | 2.0 | 0.75 | | | 5.4[19,20] | 6.0[21,22] |
| Same, porous[23] | 0.87[24] | 1.85[24] | 1.18[24] | 0.3 | 0.12 | | 3.0[25,26] | | |
| Same, porous[27] | 0.27[24] | 0.425[24] | 0.26[24] | 0.005 | 0.002 | | 5.0[25,28] | | |
| 7740[29] | 2.2[30,31] | 5.5[32] | 3.4[32] | 6.7 | 2.5 | | | 4.2[33,34] | |
| 9606[35] | 2.42[36,37] | 6.17[36,37] | 3.77[36,37] | 9.2 | 3.4 | | | 3.1[38] | 3.0[39,40] |
| AnAb[41] | 2.53[42] | ~7.3[42,43] | ~3.4[42,43] | 13 | 2.9 | | | 3.4[44] | |
| $GeO_2$ | 3.61[45] | 3.77[46] | 2.36[46] | 5.1 | 2.0 | 3.4[47,48] | 2.4[49,50] | 3.3[36] | |
| 9754[51] | 3.5[52] | 5.8[52] | 3.16[52] | 11.8 | 3.5 | | | 3.5[33] | |
| $GeS_2$ | 2.70[53] | 2.73[53] | 1.70[54] | 2.0 | 0.78 | 2.6[55,56] | | | |
| $Ge_2Se_3$ | 4.25[57] | 2.4[58] | 1.44[57] | 2.4 | 0.88 | 1.3[55,59] | | | |

[1] McSkimin (1953).

[2] By extrapolation of measurements above 70 K. Hunklinger and Arnold (1976) gave $v_\mathrm{l}=5.8\times10^5$ cm/s and $v_\mathrm{t}=3.8\times10^5$ cm/s, both measured at 1.2 K. See Table 1 of Hunklinger and Arnold (1976). From Brillouin scattering, Love (1973) determined that $v_\mathrm{l}=(5.8\pm0.1)\times10^5$ cm/s and $v_\mathrm{t}=(3.68\pm0.1)\times10^5$ cm/s at 2 K.

[3] Kosugi *et al.* (1996).

[4] Compound oscillator, 51 kHz, $a$-$SiO_2$ with approximately 1000 ppm OH (Kosugi, 2001). The addition of $a$-$GeO_2$ in concentrations of 5 and 10 mol % (OH concentration similar to the pure sample) resulted in $C_\mathrm{l}=2.5\times10^{-4}$ and $2.6\times10^{-4}$, respectively. The addition of 4 and 8 mol % F, which forms $SiO_3F$ in the amorphous structure, resulted in $C_\mathrm{l}=2.9\times10^{-4}$ and $3.3\times10^{-4}$, respectively (measured at 2.0 K, but temperature independent below 4 K; Kobayashi *et al.*, 1999). In attenuation experiments at 30 MHz, Jäckle *et al.* (1976) reported $C_\mathrm{l}=3.6\times10^{-4}$ in Suprasil W (5 ppm OH), and an identical result in Suprasil I (1200 ppm OH). From similar measurements in Suprasil W at 43 MHz, $C_\mathrm{l}=5\times10^{-4}$ was determined, and at 200 MHz, $C_\mathrm{l}\sim7\times10^{-4}$ (Bartell and Hunklinger, 1982). Note the frequency dependence referred to in the text and illustrated in Fig. 3.

[5] Classen *et al.* (1994).

[6] Measurements on Suprasil W (OH: ~5 ppm, Cl: ~1000 ppm, metals: <5 ppm, all in mole) at 1.2 and 11.4 kHz. Similar measurements at 484 and 3170 Hz, all on Suprasil W, resulted in $C_\mathrm{l}=2.3\times10^{-4}$ and $2.7\times10^{-4}$, respectively (Raychaudhuri and Hunklinger, 1984). For a microscope cover slide (main composition cited as 50 wt % $SiO_2$, 7 wt % $B_2O_3$, and 5 wt % each of $Na_2O$, $K_2$, and ZnO) vibrating at 1028 Hz, $C_\mathrm{l}=3.2\times10^{-4}$ was reported (Raychaudhuri and Hunklinger, 1984).

[7] Raychaudhuri and Pohl (1982). From the analysis by Topp and Cahill (1996).

[8] The thermal conductivity of $a$-$SiO_2$ below 1K has been shown to be independent of sample and investigator, to within ±10%, in several measurements (see von Löhneysen and Platte, 1979), although trace impurity concentrations differed: Suprasil W (<8 ppm OH, 200–400 ppm Cl, metallic impurities <5 ppm), Suprasil and Spectrosil B (900–1200 ppm OH, 50–100 ppm Cl, <5 ppm metallic impurities), and fused quartz, type GE 201 (3 ppm OH, 50 ppm metallic impurities, mostly Al; Stephens, 1976). Annealing also has a very small effect on the thermal conductivity: On Suprasil W, an anneal at 1400 °C for 4 h followed by a water quench increased the thermal conductivity by 15% (von Löhneysen, Rüsing, and Sander, 1985). On Suprasil I (1200 ppm OH), an anneal at 1300 °C for 6 h increased it by 10% (von Löhneysen, Ratai, and Sander, 1985). The addition of up to 20 mol % $K_2O$ or up to 27 mol % $Na_2O$ to $a$-$SiO_2$ has also been shown to have no measurable effect on the thermal conductivity (MacDonald *et al.*, 1985a). Similarly, in a manganese aluminosilica glass of composition $(MnO)_{0.40}(Al_2O_3)_{0.20}(SiO_2)_{0.40}$, a low-temperature thermal conductivity identical to that found in $a$-$SiO_2$ has been reported (Raychaudhuri and Pohl, 1977). Since no sound velocity is known for this glass, no tunneling strength $C_\mathrm{t}$ could be computed.

[9] Compound oscillator measurements at 90 kHz on Suprasil W (Thompson, Lawes *et al.*, 2000).

[10] Measurements at 4.5 kHz on a double-paddle oscillator etched out of a 0.125-mm-thick Suprasil W wafer yielded $C_\mathrm{t}=2.8\times10^{-4}$. Measurements below 1 K (Van Cleve, 1991) together with measurements from 0.4 to 100 K have been published by White and Pohl (1995). On a similarly shaped laser-cut oscillator of Suprasil 300 (typical impurity contents of this glass <1 ppm OH, ~400 ppm Cl; Strehlow, 2001), Classen *et al.* (2000) determined $C_\mathrm{t}=2.9\times10^{-4}$ and the same value for $C_\mathrm{l}$ (flexural) in the frequency range 0.33–14 kHz. For a review of the frequency dependence of $C$, see Topp and Cahill (1996) and also Fig. 3.

[11] Irreversibly compacted at 600 °C and 4 GPa (Weiss *et al.*, 1996).

[12] Weiss *et al.* (1996).

[13] Döring *et al.* (1994).

[14] Measured at 12 kHz.

[15] Vycor, Corning code 7930, porosity 29%.

[16] Tait (1975).

[17] DeYoreo (1985).

[18] Determined at 77 K in $N_2$ atmosphere (see DeYoreo, 1985, p. 210).

[19] Cahill *et al.* (1990).

[20] Another porous silica-based glass produced by the sol-gel process, with mass density 81% that of bulk $a$-$SiO_2$, had a thermal conductivity 20% smaller than $a$-$SiO_2$. Using measured speeds of sound, $v_t = 1.8\times10^5$ cm/s, $C_t = 7.2\times10^{-4}$ was determined (Grace and Anderson, 1986).

[21] Watson (1992).

[22] Measurements at 90 kHz.

[23] Aerogel, porosity 60%.

[24] Calemczuk *et al.* (1986).

[25] Bon *et al.* (1987).

[26] Measurements at 3.4 kHz.

[27] Aerogel, porosity 88%. Even with an elastic modulus that is a factor of 60 smaller than the other aerogel studied, $C_l$ is remarkably large. This data point is not plotted in Figs. 8 and 10.

[28] Measurements at 1.0 kHz. $Q^{-1}$ is somewhat temperature dependent.

[29] Corning code 7740, Pyrex. Composition 80.5 wt % (84 mol %) $SiO_2$, 12.9 wt % (11 mol %) $B_2O_3$, 3.8 wt % (6 mol %) $Na_2O$, 2.2 wt % (1 mol %) $Al_2O_3$.

[30] Stephens (1973).

[31] Value quoted in Stephens (1973) from Corning catalog.

[32] Love (1973).

[33] Zeller and Pohl (1971).

[34] In another borosilicate glass, BK7, on which many of the early acoustic studies of glasses were performed, a narrow plateau observed between ~5 and 10 K at frequencies of 30 and 90 MHz can be interpreted with the tunneling strength $C_t$ and $C_l$ ranging from $4\times10^{-4}$ to $6\times10^{-4}$ (Jäckle *et al.*, 1976).

[35] Corning code 9606, Pyroceram, glass used to make ceramic; 58 mol % $SiO_2$, 23 mol % MgO, 12 mol % $Al_2O_3$, 6 mol % $TiO_2$.

[36] Stephens (1976).

[37] Values shown here have been measured on Corning code 9623, a similar glass-ceramic of the following composition (main constituents): 69 mol % $SiO_2$, 15 mol % $Al_2O_3$, 9 mol % $Li_2O$.

[38] Cahill *et al.* (1991).

[39] Raychaudhuri and Pohl (1991).

[40] Measurements at 66 kHz.

[41] $An_{50}Ab_{50}$, amorphous solid of 50 mol % Anorthite ($CaAl_2Si_2O_8$) and 50 mol % Albite ($NaAlSi_3O_8$); in crystalline form, this composition would belong to the plagioclase feldspars.

[42] Linvill (1987).

[43] These approximate values are based on speeds of sound reported for plagioclase feldspars; see Linvill (1987), pp. 139 and 201.

[44] Cahill *et al.* (1992).

[45] Antoniou and Morrison (1965).

[46] R. E. Strakna as quoted in Antoniou and Morrison (1965).

[47] Laermans *et al.* (1997).

[48] Measurement at 80 MHz.

[49] Rau *et al.* (1995).

[50] Measurements at 6.3 kHz.

[51] Corning code 9754, $GeO_2$, 50 wt % (44 mol %); $Al_2O_3$, 25 wt % (23 mol %); CaO, 15 wt % (25 mol %), BaO, 5 wt % (3 mol %); ZnO, 5 wt % (6 mol %).

[52] Reddy (1995).

[53] Tsiulyanu *et al.* (1993).

[54] Onari *et al.* (1993).

[55] Gilroy and Phillips (1983).

[56] Measurements at 550 kHz, above 4.2 K only. Therefore the plateau is not clearly indicated, and the value of $C_l$ must be considered an upper limit. Measurements on $Ge_2S_3$ yielded a similar value, and those on $GeS_4$ yielded $C_l = 8\times10^{-4}$.

[57] Duquesne and Bellessa (1985).

[58] Estimated by assuming $v_l = 1.65 v_t$ as suggested by Berret and Meissner (1988) for amorphous solids.

[59] Measurements similar to those described in note 56, with similar limitations. Measurements on $GeSe_2$ yielded the same value, and those on $GeSe_4$ yielded a value twice as large.

TABLE II. Non-Si- and Ge-based glasses. See caption for Table I.

| | $\rho$ (g/cm$^3$) | $v$ ($10^5$ cm/s) | | Mod. ($10^{10}$ Pa) | | $C_l$ ($10^{-4}$) | | $C_t$ ($10^{-4}$) | |
|---|---|---|---|---|---|---|---|---|---|
| Material | $\rho$ | $v_l$ | $v_t$ | $\rho v_l^2$ | $\rho v_t^2$ | ac. | flex. | $\Lambda$ | ac. |
| $B_2O_3$ | 1.80[1] | 3.47[1] | 1.91[1] | 2.2 | 0.65 | | 3.76[2,3] | 4.3[4,5] | |
| $B_2O_3$:Na[6] | 2.03[7] | 4.65[7] | 2.48[7] | 4.4 | 1.25 | | | 7.6[8] | |
| $B_2O_3$:K[9] | 2.20[10] | 4.74[10] | 2.62[10] | 4.9 | 1.5 | | | 5.7[10] | |
| $B_2O_3$:$Li_2O$[11] | 2.054[1] | 6.92[1] | 3.97[1] | 9.8 | 3.2 | 8.2[1,12] | | 5.8[13,14] | 8.9[1,12] |
| LASF[15] | 5.79[16] | 5.644[16] | 3.560[16] | 18 | 7.3 | 2.0[16,17] | | | |
| Se | 4.3[18] | 2.0[18] | 1.05[18] | 1.7 | 0.47 | | 2.2[19,20] | 2.9[4,21] | |
| $CdGeAs_2$ | 5.72[22] | 3.03[23,24] | 1.86[23,24] | 5.3 | 2.0 | | | 1.6[25] | 1.2[26,27] |
| B[28] | 1.7[29] | | 8.1[29] | | 11 | | | | 2.2[29,30] |
| $B_9C$[28] | 1.68[29] | | 6.64[29] | | 7.4 | | | | 3.0[29,30] |
| K,Ca,$NO_3$[31] | 2.1[4] | 3.456[4] | 1.749[4] | 2.5 | 0.64 | | | 10.0[4,32] | |
| $As_2S_3$ | 3.2[26] | 2.65[33] | 1.44[33] | 2.2 | 0.66 | 2.3[34,35] | 1.4[36,37] | 1.7[4,38] | |
| LiCl:$H_2O$[39] | 1.2[40] | 4.0[41] | 2.0[41] | 1.9 | 0.48 | 7.0[40,42] | | | 10.0[41,43] |
| $MnF_2$[44] | 4.05[45] | 4.7[45] | 2.5[45] | 8.9 | 2.5 | 2.2[45,46] | | | 3.8[45,47] |
| V52[48] | 4.8[49] | 4.15[49] | 2.25[49] | 8.3 | 2.4 | 6.0[49,50] | | | 5.4[49,51] |
| LaPO[52] | 3.4[53] | 4.463[53] | | 9.6 | | 2.8[53,54] | | | |
| Glycerol[55] | 1.42[56] | | 1.85[57] | | 0.5 | | | 3.0[58] | |

[1] Devaud *et al.* (1983).

[2] Rau *et al.* (1995).

[3] Measurements at 2.8 kHz.

[4] Stephens (1976).

[5] From the analysis by Topp and Cahill (1996). Two $B_2O_3$ samples were measured by Stephens (1976). One had a larger specific-heat anomaly and showed an infrared absorption of OH seven times higher than the other. The results of the thermal conductivity, however, were indistinguishable to better than 10%. Similarly, doping with 1 mol % $Na_2O$ had no effect (Villar *et al.*, 1986).

[6] $(B_2O_3)_{0.9}(Na_2O)_{0.1}$

[7] Ramos and Buchenau (1997).

[8] Villar *et al.* (1986).

[9] $(B_2O_3)_{0.75}(K_2O)_{0.25}$; results for $(B_2O_3)_{0.95}(K_2O)_{0.05}$ were very similar to those of pure $B_2O_3$.

[10] MacDonald *et al.* (1985b).

[11] $(B_2O_3)_{1.0}(Li_2O)_x$, $x=0.5$. Also measured were $x=0.11$, 0.25, 0.70, and $x=0.5$ with 0.7 LiCl added (a supersonic conductor; see Prieur and Ciplys, 1981), all with similar results.

[12] Measurements from 150 to 600 MHz.

[13] Raychaudhuri *et al.* (1980).

[14] Composition $(B_2O_3)_{0.67}(Li_2O)_{0.33}$. Adding $(WO_3)_{0.03}$ decreased the thermal conductivity by 20%; change of sound velocity was not measured.

[15] LASF-7. Composition is given in Berret *et al.* (1986), p. 71, as "mostly $B_2O_3$, $La_2O_3$, and $ThO_2$, with a few percent $Ta_2O_3$ and $Nb_2O_5$."

[16] Berret *et al.* (1986).

[17] Measurements at 30 and 90 MHz (Berret *et al.*, 1986).

[18] Duquesne and Bellessa (1985).

[19] Jaqmin *et al.* (1983).

[20] Measurements at 228 Hz.

[21] After a melt-quenched amorphous sample was annealed, an increase of the thermal conductivity below 1 K by 25% was observed, together with a decrease of the fictive temperature by ~30 K (10%; R. Calemczuk, Ph.D. thesis, as quoted by Johari, 1986).

[22] Cervinka *et al.* (1970).

[23] Nemilov *et al.* (1971).

[24] Room-temperature values.

[25] Cahill and Pohl (1988b).

[26] Topp and Cahill (1996).

[27] Measurements at 90 kHz.

[28] Thin films produced by *e*-beam evaporation, with mass densities ~68% of those of the bulk crystalline phases ($\beta$-B and $B_9C$, respectively), believed to be caused mainly by porosity.

[29] Medwick *et al.* (1998).

[30] Measurements at 5.3 kHz.

[31] $KCa(NO_3)_3$.

[32] In $(KNO_3)_{0.60}[Ca(NO_3)_2]_{0.40}$, doping with 3.4 wt % water lowered the thermal conductivity by 7% (Klitsner *et al.*, 1981). In the doped sample, the number density of the water molecules, $2.5\times10^{21}$ cm$^{-3}$, was 17% of that of the $NO_3$ ions. Since the speed

of sound of the doped sample is unknown, no tunneling strength is available.

[33] R. B. Stephens, private communication, as reported by Cahill and Pohl (1987).

[34] Ng and Sladek (1975).

[35] Measurements from 30 to 390 MHz.

[36] Jacobsen *et al.* (1991).

[37] Measurements at ~300 Hz.

[38] From the analysis by Topp and Cahill (1996).

[39] $LiCl{:}7H_2O$.

[40] Kasper and Röhring (1984).

[41] Röhring *et al.* (1990).

[42] Measurements from 17 to 104 MHz.

[43] Measurements at 67 MHz. Residual absorption, which was estimated to be ~15%, was not subtracted.

[44] $(MnF_2)_{0.65}(BaF_2)_{0.2}(NaPO_3)_{0.15}$ from Doussineau *et al.* (1985). A similar, nonmagnetic glass containing Zn instead of Mn had very similar data and is not listed here.

[45] Doussineau *et al.* (1985).

[46] Measurements at 260 MHz.

[47] Measurements at 205 MHz.

[48] $(ZrF_4)_{0.575}(BaF_2)_{0.3375}(ThF_4)_{0.0875}$.

[49] Doussineau and Matecki (1981).

[50] Measurements at 130 MHz and determined only from a weakly resolved plateau.

[51] Measurements at 145 MHz and determined only from a weakly resolved plateau.

[52] $(La_2O_3)_{0.26}(P_2O_5)_{0.74}$.

[53] Carini *et al.* (1994).

[54] Measurements between 10 and 90 MHz. Measurements on $(Sm_2O_3)_x(P_2O_5)_{1-x}$, $x=0.19$ and 0.25, led to $C_l=2.9\times10^{-4}$ and $2.4\times10^{-4}$, respectively.

[55] $C_3H_5(OH)_3$.

[56] Schulz (1954).

[57] Obtained by extrapolating longitudinal sound velocity measurements above 180 Kby Jeong *et al.* (1986) to low temperatures, and assuming $v_l=2v_t$ (Ramos, 2002).

[58] By extrapolating the thermal conductivity measurements above 1.7 K to low temperatures, $\beta_0=6.0\times10^{-4}$ W/cm K$^3$ was obtained (Talon *et al.*, 2001).

modulus measurements (flexural vibrating reed), since the Young's-modulus sound velocity is closely related to the longitudinal sound velocity for typical Poisson ratios (Raychaudhuri and Hunklinger, 1984). Note that Figs. 7 and 8 contain the measurements on all amorphous solids that have been reviewed. The only exceptions are the data on *a*-Si, *a*-Ge, and *a*-C, which will be reviewed separately below. Also omitted from these two figures were those solids for which the speeds of sound are unknown, since their values are needed in Eqs. (7) and (11) as well as for the determination of the moduli (the horizontal axes in the figures). Details on those solids, however, will be found in the notes to the tables. They indicate no exceptional behavior.

The experimental findings shown in these two figures can be summarized with two simple statements:

(1) $\lambda/\ell$ is independent of the substance within approximately a factor of 20.

(2) For a given substance, $\lambda/\ell$ is independent of the frequency used, within a small factor, usually smaller than 2 or 3.

Since acoustic measurements typically use frequencies from $10^2$ to $10^9$ Hz and thermal conductivity probes frequencies from $5\times10^9$ to $2\times10^{11}$ Hz in the temperature range $0.05<T<2.0$ K, the measurements summarized in Figs. 7 and 8 span up to nine orders of magnitude in frequency! It is highly remarkable that a general phenomenon as summarized in statements (1) and (2) continues to be without a simple physical explanation.

In order to avoid misunderstandings, we emphasize that neither statement is new. Statement (1) was first suggested by Freeman and Anderson (1986), who compared the thermal conductivity of 11 amorphous solids and suggested $(\lambda/\ell)\sim6.7\times10^{-3}$ as an average value, well within the range shown in Fig. 7. Statement (2) is identical to the well-known fact that the same tunneling strength $C$, a quantity to be introduced below, can be used to describe both the low-temperature thermal conductivity and sound attenuation. However, the demonstration that both statements apply to all amorphous solids measured to date (apart from the exceptions mentioned above) has not been made before.

### B. Tunneling strength

The summarizing statements at the end of the preceding section are somewhat oversimplified in that $\lambda/\ell$ as determined from acoustic measurements at the lowest temperatures has been found to depend on temperature as shown in Figs. 2 and 3. Only in the limit of vanishingly small frequency does $\lambda/\ell$ appear to be truly temperature independent. At high frequencies, the temperature range in which $\lambda/\ell$ is constant shrinks. Yet, in thermal conductivity experiments in which the heat is carried by phonons of even higher frequencies, the same $\lambda/\ell$ is found to describe the measurements well. These observations are explained by the phenomenological tunneling model [see Anderson *et al.* (1972), Jäckle (1972), and Phillips (1972)], which postulates that the lower-

TABLE III. Polymers. See caption for Table I.

| | $\rho$ (g/cm$^3$) | $v$ ($10^5$ cm/s) | | Mod. ($10^{10}$ Pa) | | $C_l$ ($10^{-4}$) | | $C_t$ ($10^{-4}$) | |
|---|---|---|---|---|---|---|---|---|---|
| Material | $\rho$ | $v_l$ | $v_t$ | $\rho v_l^2$ | $\rho v_t^2$ | ac. | flex. | $\Lambda$ | ac. |
| PMMA[1] | 1.18[2] | 3.150[2] | 1.570[2] | 1.2 | 0.29 | 3.7[2,3] | 3.1[4,5,6] | 4.8[7,8] | 5.7[2,3] |
| PS[9] | 1.06[10] | 2.8[11] | 1.34[10] | 0.83 | 0.19 | | 8.3[5,12] | 10.4[7,13] | 7.8[10,14] |
| PC[15] | 1.2[2] | 2.9[16] | 1.37[16] | 1.0 | 0.17 | 3.5[2,17] | | 12.2[18,13] | 9.5[19,20] |
| PB[21] | 0.93[22] | 3.02[22] | 1.46[22] | 0.85 | 0.20 | | | 5.6[23,24] | |
| PET[25] | 1.335[26] | 2.32[27] | 1.16[26] | 0.72 | 0.18 | | | 6.4[28,29] | |
| PEMA[30] | 1.12[2] | 2.955[2] | 1.5[27] | 0.99 | 0.25 | 4.6[2,3] | 6.9[4,31] | | |
| PBM[32] | 1.06[33] | 3.1[27] | 1.54[33] | 1.0 | 0.25 | | 1.3[4] | | |
| Stycast 1266[34] | 1.18[35] | 2.61[36] | 1.3[27] | 0.8 | 0.20 | 3[36,37] | | 5.5[38] | 3.9[39] |
| Stycast 2850[39] | 2.26[10] | 4.4[27] | 2.21[10] | 4.4 | 1.1 | | | | 2.7[10,40] |
| SC5[41] | 1.14[42] | 3.14[42] | 1.56[42] | 1.1 | 0.28 | | | 7.0[42] | |
| SC8[43] | 1.1[42] | 3.22[42] | 1.6[42] | 1.1 | 0.28 | | | 3.6[42] | |
| 7031[44] | | | 1.26[45] | | 0.19[45] | | | 4.2[46,47] | |
| ET[48] | 1.2[49] | 3.25[49] | | 1.27 | | 5[49,50,51] | | | |

[1] Polymethylmethacrylate.

[2] Federle and Hunklinger (1982).

[3] Measurements from 15 to 400 MHz. The value of $C_l = 9.1 \times 10^{-4}$ for PMMA reported by Nava *et al.* (1991) seems somewhat large.

[4] Hickel and Kasper (1988).

[5] Nittke *et al.* (1995).

[6] Vibrating reed measurements between 300 and 800 Hz (Hickel and Kasper, 1988). According to Nittke *et al.* (1995), $C_l = 2.7 \times 10^{-4}$ (535 Hz). The frequency dependence of the tunneling strength for PMMA between $10^{-2}$ and $10^8$ Hz (approximately a factor of 3) has been reviewed by Topp and Cahill (1996).

[7] Stephens (1973).

[8] From the analysis by Topp and Cahill (1996). This reference also contains a review of earlier attenuation measurements on PMMA. The thermal conductivity of Mylar was found to be $1.8 \times 10^{-4}$ $[\mathrm{W\,cm^{-1}\,K^{-3}}] \cdot T^2$ (Peterson and Anderson, 1972), close to that of PS and PC. Since its speeds e-sound are unknown, the data for Mylar were not included in the table.

[9] Polystyrene.

[10] Topp and Cahill (1996).

[11] Duquesne and Bellessa (1979).

[12] Measurements at 240 Hz and 3.2 kHz.

[13] From the analysis by Topp and Cahill (1996).

[14] Measurements at 87 kHz.

[15] Polycarbonate (Lexan).

[16] Cieloszyk *et al.* (1973).

[17] Measurements from 15 to 400 MHz.

[18] Zaitlin and Anderson (1975).

[19] Hartwig and Schwarz (1986).

[20] Torsional oscillator measurements at 10 Hz and $T \geqslant 5$ K. Similar measurements were also reported for several other polymers.

[21] Polybutadiene.

[22] Bhattacharyya *et al.* (1979).

[23] Matsumoto and Anderson (1981).

[24] Partial crystallization was found to decrease the thermal conductivity without changing the temperature dependence noticeably (Freeman *et al.*, 1986). Changing the cross linking by $\gamma$ irradiation has very little effect on the conductivity in the $T^2$ temperature region (von Löhneysen *et al.*, 1987).

[25] Polyethyleneterephtalate.

[26] Armeniades and Baer (1971).

[27] Calculated assuming that $v_l \approx 2 v_t$ as suggested by Choy *et al.* (1975), based on measurements of Athougies *et al.* (1972) on several polymers.

[28] Greig and Sahota (1983).

[29] Greig and Sahota (1983) also studied the effect of partial crystallization.

[30] Polyethylmethacrylate.

[31] Vibrating reed measurements at ~500 Hz.

[32] Poly-n-butyl-methacrylate.

[33] Heijboer and Pineri (1982).

[34] Clear Stycast.

[35] Emerson-Cuming, Technical Bulletin. 1.25 g/cm$^3$ according to Guillon and Frossati (1992).

[36] Guillon and Frossati (1992).

[37] Measurements at 88 MHz. At 148 MHz, $C_l = 4 \times 10^{-4}$.

[38] Olson (1993).

[39] Measurements done in this investigation at 85 KHz using the same technique as described in Topp and Cahill (1996).

[40] Stycast 2850FT, black, containing ≈80 wt % $Al_2O_3$ (Topp and Cahill, 1996).

[41] Data taken at 84 kHz.

[42] Epoxy SC5: Scotchcast 5.

[43] Matsumoto *et al.* (1979).

[44] Epoxy SC8: Scotchcast 8.

[45] Polyvinyl phenolic, GE7031.

[46] From the $T^3$ part of the specific heat measured in Stephens (1975), and a mass density assumed to be 1.2 g/cm$^3$, a lower limit of the Debye velocity $v=1.4\times10^5$ cm/s was determined. As rule of thumb, $v_t=0.9v$ is used, which works well for all polymers in this list.

[47] Stephens (1975).

[48] Because of the assumptions made for this polymer (see note 46), this value is estimated to be accurate only to ±20%.

[49] ET1000, commercial epoxy resin, Silitro Comp.

[50] Doussineau and Schön (1983).

[51] Measurements from 150 MHz to 2.14 GHz.

[52] On a natural resin (amber), $C_l=6\times10^{-4}$ has been reported by Nava *et al.* (1991) between 5 and 25 MHz (no sound velocity given).

frequency phonons are attenuated mainly by a relaxation of the tunneling defects, while the high-frequency phonons that carry the heat are resonantly scattered. Both the internal friction and acoustic attenuation, including the dropoff at the lowest temperatures and the $T^2$ dependence of the thermal conductivity, are described in the tunneling model with the same adjustable parameter, the tunneling strength $C_i$. It is defined as

$$C_i=\frac{\overline{P}\gamma_i^2}{\rho v_i^2}, \tag{14}$$

where $\overline{P}$ is the spectral density of the tunneling states, $\gamma_i$, is the energy with which they are coupled to the plane-wave lattice vibrations, $\rho$ is the mass density, and $v_i$ is the speed of sound, with $i$ indicating their polarization, transverse (t) and longitudinal (l). Thus, in the tunneling model, instead of comparing $\lambda/\ell$, one compares the tunneling strengths $C_t$ and $C_l$, which are listed in Tables I–IV for all amorphous solids we found treated in the literature (except *a*-Si, *a*-Ge, and *a*-C). Amorphous solids for which either thermal conductivity or acoustic attenuation is known, but for which the speed of sound is unknown, have been listed in the notes to the tables (the speed of sound is needed to determine $\lambda/\ell$, to derive $C_t$ from thermal conductivity, and to plot the tunneling strength versus the elastic constant in the figures). The elastic constants $\rho v_i^2$ cover four orders of magnitude, varying from $2.0\times10^7$ Pa for aerogel in Table I to $2.2\times10^{11}$ Pa for PdSiCu in Table IV. Yet the tables show how little the tunneling strength varies, either from material to material or with the frequency and polarization of the acoustic wave (including those materials that are listed only in the notes). The contents of the tables are illustrated in Figs. 9 and 10 and demonstrate the remarkable universality referred to in the Introduction.

The following remarks may help in digesting the figures and tables. Figures 7 and 8 illustrate the relative inverse phonon mean free paths, $\lambda/\ell$, while Figs. 9 and 10 show the tunneling strengths. Yet Figs. 7 and 9 and Figs. 8 and 10 appear to be very similar. The reason for this similarity can be easily understood, as will be shown next.

According to the tunneling model,

$$C_{t,l}=\frac{2}{\pi}Q_{0,t,l}^{-1}=\frac{v_{t,l}}{\pi^2\nu}\alpha_{0,t,l}, \tag{15}$$

where $Q_0^{-1}$ and $\alpha_0$ are the internal friction and acoustic (energy) attenuation in the temperature-independent (plateau) region, respectively, $v_{t,l}$ are the speeds of sound, and $\nu$ is the frequency of the wave. For acoustic measurements in the temperature-independent plateau region, the tunneling states are relaxed via a relaxational process in the strain field of sound waves, mediated either by phonons in insulated glasses or by normal-state electrons in metallic glasses. In either case, Eq. (15) is valid (Jäckle, 1972). Rewriting Eq. (11) in terms of $C_{t,l}$, we obtain

$$\left.\frac{\lambda}{\ell}\right|_{t,l}=\pi^2C_{t,l}. \tag{16}$$

Thus the acoustically determined relative inverse phonon mean free paths and the tunneling strengths are related by a constant factor ($\pi^2\sim9$).

If the thermal conductivity $\Lambda$ is dominated by phonons that are resonantly scattered by the low-energy excitations, $\Lambda$ can be approximated by (Hunklinger and Raychaudhuri, 1986)

$$\Lambda=\frac{\rho k_B^3T^2}{6\pi\hbar^2\overline{P}}\sum_i\frac{v_i}{\gamma_i^2}. \tag{17}$$

In order to simplify this expression, we use the empirical relations that Berret and Meissner (1988) found to be valid for a wide range of amorphous solids:

$$v_l\approx1.65v_t \quad\text{and}\quad \gamma_l^2\approx2.5\gamma_t^2. \tag{18}$$

With Eqs. (14) and (18), Eq. (17) takes the form

$$\Lambda=\frac{2.66k_B^3}{6\pi\hbar^2}\frac{1}{C_tv_t}T^2$$

$$=3.3\times10^{-2}[\mathrm{W\,K^{-3}\,s^{-1}}]\frac{1}{C_tv_t}T^2=\beta_0T^2. \tag{19}$$

With Eqs. (18), (19), and $v=[(1/3)v_l^{-3}+(2/3)v_t^{-3}]^{-1/3}$, Eq. (7) is rewritten as

TABLE IV. Metallic glasses. See caption for Table I.

| | $\rho$ (g/cm$^3$) | $v$ ($10^5$ cm/s) | | Mod. ($10^{10}$ Pa) | | $C_l$ ($10^{-4}$) | | $C_t$ ($10^{-4}$) | |
|---|---|---|---|---|---|---|---|---|---|
| Material | $\rho$ | $v_l$ | $v_t$ | $\rho v_l^2$ | $\rho v_t^2$ | ac. | flex. | $\Lambda$ | ac. |
| ZrPd[1] | 7.86[2,3] | 4.4[4] | 2.4[5] | 15.0 | 4.5 | 0.4[4,6] | 1.5[7,8] | 1.4[2] | |
| ZrCu[9] | 7.0[10,11] | 3.56[10,12] | 1.8[13] | 8.9 | 2.3 | | 1.3[10,14] | 2.9[15,16] | 0.6[17,18] |
| PdSiCu[19] | 10.52[20] | 4.60[20] | 1.797[20] | 22.0 | 3.4 | | 0.87[7,21] | 1.0[22,23] | 0.7[24,25] |
| PdSi[26] | 10.25[27] | 4.2[28] | 1.7[28] | 18.0 | 3.0 | | 0.64[29,30] | | |
| NiP[31] | 8.0[32] | 5.2[32] | 2.3[32] | 21.6 | 4.2 | | | 3.6[33,34] | |
| FeP[35] | 7.0[36] | 4.5[29,37] | 2.4[38] | 14.0 | 4.0 | | 2.7[29,39] | | |
| PdSiAg[40] | 10.5[41] | 4.43[42,43] | 1.7[42,43] | 21.0 | 3.0 | | 1.5[43,44] | | |
| PdUSi[45] | 11.1[46] | - | 1.7[47] | 21.0 | 3.0 | | | 0.6[46,48] | |
| CuZrAl[49] | 6.7[50] | 3.1[51] | | 6.4 | | | 0.64[52] | | |

[1] $Zr_{0.7}Pd_{0.3}$ (melt spun or splat cooled).

[2] Graebner *et al.* (1977).

[3] $T_c = 2.53$ K.

[4] Weiss *et al.* (1980).

[5] Weiss and Golding (1988).

[6] 740 MHz. No information on heat treatment of sample. Arnold *et al.* (1981) analyzed the same data as well as the data obtained at 1.3 GHz, with $C_l = 0.55 \times 10^{-4}$.

[7] Raychaudhuri and Hunklinger (1984).

[8] Measurements at 1182 Hz (Raychaudhuri and Hunklinger, 1984). Measurements at 3.1 kHz confirmed this result, $C_l = 1.65 \times 10^{-4}$ (Classen *et al.*, 1994). Esquinazi *et al.* (1986) determined $C_l = 1.9 \times 10^{-4}$ at 640 Hz and $C_l = 6 \times 10^{-5}$ at 720 MHz.

[9] $Zr_{0.7}Cu_{0.3}$ (melt spun or splat cooled).

[10] Esquinazi and Luzuriaga (1988).

[11] Melt spun. $T_c = 2.806$ K. $T_c$ ranging from 2.35 to 2.7 K was reported by Grondey *et al.* (1983).

[12] Determined with $v_l = 1.27 v_E$ from a melt-spun sample with the Young's-modulus velocity $v_E = (2.8 \pm 0.3) \times 10^5$ cm/s measured by Esquinazi and Luzuriaga (1988) and a Poisson ratio assumed to be 0.40.

[13] From $v_l$, assuming $v_l = 2 v_t$, which appears to be a good approximation in metallic glasses, as shown in this table.

[14] Annealing lowers this value by over a factor of 3.

[15] Grondey *et al.* (1983).

[16] Annealing lowers this value by a factor of up to 3 (Esquinazi *et al.*, 1982). Similar results have been reported for sputtered films (Ravex *et al.*, 1984). Similar results were also reported for different compositions: $Zr_{0.74}Cu_{0.26}$ and $Zr_{0.60}Cu_{0.40}$ (Grondey *et al.*, 1983) and $Zr_{0.80}Cu_{0.20}$ (Lasjaunias *et al.*, 1984). Similar thermal conductivities were also reported for alloys of different chemical composition: $Zr_{0.55}Be_{0.45}$ and $Zr_{0.60}Be_{0.40}$ (Raychaudhuri and Hasegawa, 1980), $Zr_{0.67}Ni_{0.33}$ (Gronert *et al.*, 1986), $Zr_{0.76}Ni_{0.24}$ (Ravex *et al.*, 1981), and melt-spun $La_{0.70}Cu_{0.30}$ (Daudin, 1981; Esquinazi *et al.*, 1982). However, since their speeds of sound are all unknown, no value of $C$ was determined.

[17] Arnold *et al.* (1982).

[18] $Cu_{0.60}Zr_{0.40}$, splat cooled, no information on heat treatment, measured at 745 MHz.

[19] $Pd_{0.78}Si_{0.16}Cu_{0.06}$.

[20] Golding *et al.* (1972).

[21] Measurements of 178 and 1030 Hz. For similarly prepared samples, $C_l$ (at ~500 Hz) ranging from $1.0 \times 10^{-4}$ (splat cooled) to $0.35 \times 10^{-4}$ (slow cooled) have been reported by Hunklinger (1987) and Rabenau (1986). A similar variation was found in thermal conductivity (Herlach *et al.*, 1986, 1987). Details on the effect of annealing PdSiCu can be found in Chap. 4 of Thompson (2000); see also Thompson, Vu, and Pohl (2000). Strain-amplitude dependence between 0.2 mK and 0.5 K has been explored by Ramos *et al.* (2000).

[22] Matey and Anderson (1977a).

[23] After 6 h anneal at 300 °C, this value dropped to $0.67 \times 10^{-4}$.

[24] Doussineau (1981).

[25] Measured at ~1.0 K and between 370 and 852 MHz. The value given is an average and is probably a lower limit, since the ultrasonic absorption still increases with increasing temperature.

[26] $Pd_{0.81}Si_{0.19}$.

[27] Table 2 of Gaskell (1983).

[28] Calculated from the shear elastic constants given in Table 6.1 of Künzi (1983).

[29] Barmatz and Chen (1974).

[30] Measurements from 200 to 1300 Hz. In Fig. 2, of Van Cleve *et al.* (1990), $C_l = 5 \times 10^{-5}$ was reported (~300 Hz).

[31] $Ni_{0.86}P_{0.14}$ (Bellessa *et al.*, 1976), bulk samples produced by electrodeposition.

[32] Bellessa *et al.* (1977).

[33] Matey and Anderson (1977b).

[34] Matey and Anderson (1977b) also explored a bulk sample of electrodeposited $Fe_{0.84}P_{0.14}$ and foils of $Ti_{0.50}Be_{0.40}Zr_{0.10}$ and $Fe_{0.32}Ni_{0.36}Cr_{0.14}P_{0.12}B_{0.06}$. Their phonon thermal conductivities were close to that of the NiP, but since their speeds of sound are unknown, they were not converted to a tunneling strength $C$. The same problem was encountered in melt-spun amorphous ribbons of $(Mo_{1-x}Ru_x)_{0.80}P_{0.20}$. Their low-temperature thermal conductivity as well as their low-temperature internal friction at ~400 Hz

are close to those of $a$-SiO$_2$ ($\beta_0 \approx 1.0\times10^{-4}$ W/cm K$^3$; Sürgers and von Löhneysen, 1988), $Q_0^{-1} \approx 9\times10^{-4}$, i.e., $C_l \approx 6\times10^{-4}$ (Lichtenberg *et al.*, 1990), but since the speed of sound was unknown, the thermal conductivity could not be evaluated, nor could the value of $C_l$ be shown in the figures.

[35] $Fe_{0.74}P_{0.16}C_{0.05}Al_{0.03}Si_{0.02}$.

[36] Assumed to be equal to the mass density of $a$-$Fe_{0.76}P_{0.24}$; see Table 2 of Gaskell (1983).

[37] The Young's modulus $E=9.1\times10^{11}$ dyn/cm$^2$ is given by Barmatz and Chen (1974). From this follows the Young's-modulus velocity $v_E=3.6\times10^5$ cm/s, and with a Poisson ratio $\nu=0.37$ as reported for similar compositions in Table 6.1 of Künzi (1983), $v_l$ is determined.

[38] Assuming a shear modulus $G=40$ GPa, which is an average for metallic glasses of similar composition in Table 6.1 of Künzi (1983). For amorphous $Co_{0.82}P_{0.18}$, $\rho=8.1$ g/cm$^3$, $v_l=5.1\times10^5$ cm/s, and $v_t=2.4\times10^5$ cm/s have been reported (Bellessa, 1977), all fairly close to the values listed here for this metal with similar composition.

[39] Measurements at 155 Hz, from 1.5 to 300 K. Annealing (670 K, 8 min) decreased the internal friction at the lowest temperature by ~25%. Crystallization decreased it by 80%. A similar low-temperature internal friction was reported by Barmatz and Chen (1974) for $Ni_{0.74}P_{0.16}B_{0.07}Al_{0.03}$ and $Fe_{0.75}P_{0.16}Si_{0.06}Al_{0.03}$ in the amorphous state.

[40] $Pd_{0.775}Si_{0.165}Ag_{0.06}$ (splat cooled or water quenched).

[41] Not measured. Assumed to be equal to that of PdSiCu (Rabenau, 1986).

[42] Values at 300 K from Dutoit and Chen (1973). According to Fig. 32 of Rabenau (1986), the speed of sound in this metallic glass will increase by ~4% upon cooling to 4.2 K.

[43] Rabenau (1986).

[44] From vibrating reed measurements on a splat-cooled sample at 210 Hz. On a similarly prepared and measured sample in which the Ag was replaced with Ni, $C_l=0.95\times10^{-4}$ was obtained (405 Hz; Rabenau, 1986). On a relatively slowly cooled water-quenched PdSiAg sample, $C_l=4\times10^{-5}$ has been reported for longitudinal sound at 250 MHz (Dutoit and Chen, 1973).

[45] $Pd_{0.588}U_{0.206}Si_{0.206}$.

[46] Freeman *et al.* (1987).

[47] This value is not known. It is taken as a reasonable value for the evaluation of the thermal conductivity data.

[48] Measurements on an unannealed melt-spun ribbon sample of $Pd_{0.588}U_{0.206}Si_{0.206}$. Between 0.15 and 1.0 K, its thermal conductivity, $\Lambda \propto T^{\delta}$, is best fitted with $\delta=1.6$, which is the smallest exponent reported for amorphous solids, as far as we know.

[49] $Zr_{0.65}Al_{0.075}Cu_{0.275}$, splat cooled (Mohr *et al.*, 2000).

[50] ±5% (Weiss, 2000).

[51] Young's-modulus sound velocity, uncertainty ±15% (Weiss, 2000). Because of this uncertainty, the same value has been assumed for $v_l=3.1\times10^5$ cm/s.

[52] Mohr *et al.* (2000).

$$\left.\frac{\lambda_{\mathrm{dom}}}{\ell}\right|_{\mathrm{t}} \approx 12.5 C_{\mathrm{t}}. \qquad (20)$$

The relative inverse (dominant) phonon mean free path and the (transverse) tunneling strength are also simply connected by a numerical factor (~12.5). The small difference between the factors in Eqs. (16) and (20) results from the dominant phonon approximation in Eq. (3).

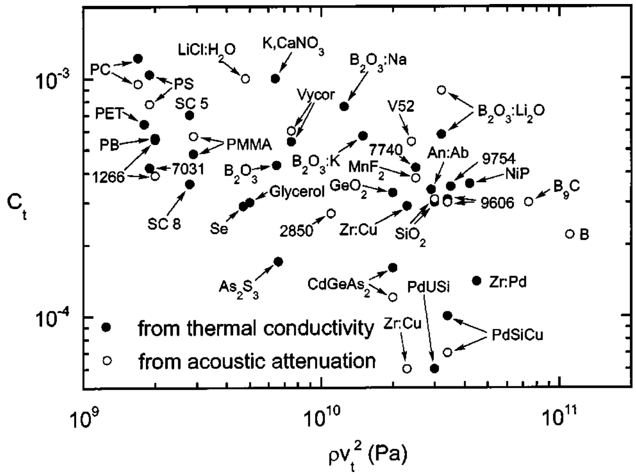

FIG. 9. The same universality shown in Fig. 7, using the tunneling model for the analysis. The tunneling strengths $C_t$ shown were obtained from experiments using transverse waves spanning over nine orders of magnitude in frequency. Data can be found in Tables I–IV.

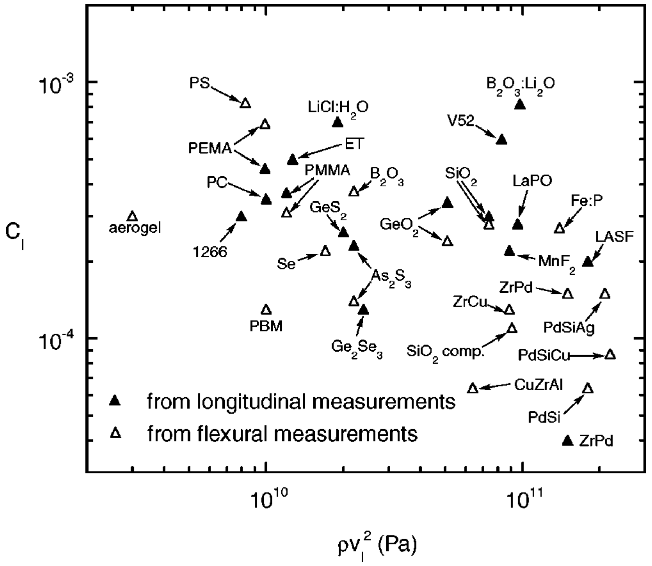

FIG. 10. The tunneling strength $C_l$ for amorphous solids obtained using longitudinal and Young's-modulus waves. Data can be found in Tables I–IV. As in Fig. 8, the data point for the most porous $a$-SiO$_2$ (aerogel) has not been plotted, for which the elastic modulus $\rho v_l^2$ would lie outside the range plotted by a factor of 100.

TABLE V. Amorphous Si, Ge, and C films. The tunneling strengths $C_t$ were all determined from internal friction measurements at ~5 kHz; see, however, note 15. The *a*-Si and *a*-Ge films are taken from a recent review (Liu and Pohl, 1998), with the following exceptions: For the data on Si amorphized by self-implantation we also refer the reader to Liu, Vu, *et al.* (1998). The data for the hot-wire chemical-vapor-deposited (HWCVD) hydrogenated *a*-Ge are taken from Liu *et al.* (2002), those for HWCVD deuterated *a*-Si and those for the plasma-enhanced chemical-vapor-deposited (PECVD) hydrogenated *a*-Si are taken from Liu *et al.* (1999). Since many of the mass densities and speeds of sound have been carefully revised, as explained in the notes, the listed values of $C_t$ often differ somewhat from those given in the original references. The experimental data on which the tunneling strengths are based, however, are the original ones. The results of pulsed-laser-deposited (PLD) diamondlike *a*-C films are taken from Liu *et al.* (2002).

| Samples | $\rho$ (g/cm$^3$) | $v_t$ ($10^5$ cm/s) | $\rho v_t^2$ ($10^{10}$ Pa) | $C_t$ ($10^{-4}$) |
|---|---|---|---|---|
| *e*-beam *a*-Ge | 4.7[1] | 2.6[2] | 3.2 | 0.40[3] |
| Same, annealed[4] | 4.7[5] | 2.6[6] | 3.2 | 0.14[3] |
| Sputtered *a*-Ge | 4.7[1] | 1.9[7] | 1.7 | 0.41[3] |
| HWCVD *a*-Ge:H | 4.7[1] | 2.9[7] | 3.9 | 0.033[8] |
| *e*-beam *a*-Si | 2.1[9] | 4.2[10] | 3.6 | 0.83[3,11] |
| Same, annealed[4] | 2.1[5] | 4.2[6] | 3.6 | 0.43[3] |
| Sputtered *a*-Si | 2.1[9] | 3.8[12] | 3.1 | 0.38[3] |
| Same, annealed[4] | 2.1[5] | 3.8[6] | 3.1 | 0.10[3] |
| $Si^+$ implanted *a*-Si | 2.29[13] | 4.8[14] | 5.3 | 0.22[3,15] |
| Same, annealed[16] | 2.29[5] | 4.8[6] | 5.3 | 0.13[3] |
| HWCVD *a*-Si:H, 1 at. % H | 2.28[17] | 4.8[18] | 5.3 | 0.003[19] |
| Same, annealed[20] | 2.30[21] | 4.8[6] | 5.3 | 0.019[3] |
| HWCVD *a*-Si:D, 1 at. % D | 2.28[22] | 4.8[22] | 5.3 | 0.0025[23] |
| PECVD *a*-Si:H, 10 at. % H | 2.23[17] | 4.7[24] | 4.9 | 0.025[23] |
| Same, annealed[25] | 2.23[5] | 4.7[6] | 4.9 | 0.01[23] |
| PLD a-C | 2.95[26] | 10.7[27] | 33.7 | 0.39[8] |
| Same, annealed[28] | 2.85[26] | 10.7[6] | 33.7 | 0.24[8] |

[1] Little seems to be known about the density of these or of sputtered *a*-Ge films. van den Berg and von Loehneysen (1985) reported $\rho = 5.25$ g/cm$^3$ (bulk crystalline Ge: 5.35 g/cm$^3$), on an *e*-beam evaporated film of unspecified thickness. Chopra and Bahl (1970) reported $\rho = 4.7$ g/cm$^3$ on a thermally evaporated film of 2 $\mu$m thickness, and $\rho$ to increase with film thickness. $\rho$ also changes by several percent by changing deposition conditions. Graebner and Allen (1984) reported $\rho$ ranging from 4.54 to 5.17 g/cm$^3$ on ~10-$\mu$m-thick films deposited by thermal evaporation. We have chosen for both *e*-beam and sputtered *a*-Ge $\rho = 4.7$ g/cm$^3$, which is often used in calculations and represents a 10% reduction of the bulk crystalline value. We use the same value for HWCVD *a*-Ge:H, since no measurements can be referred to at this moment.

[2] According to Cox-Smith *et al.* (1985), thermal and *e*-beam evaporated *a*-Ge films 1–6 $\mu$m thick have an average Young's speed of sound 81.6% that of polycrystalline films. Using the same percentage for the transverse speed of sound of bulk crystalline Ge ($v_t = 3.2 \times 10^5$ cm/s), we obtain the value listed here.

[3] Liu and Pohl (1998).

[4] 350 °C, 5 h, in $3 \times 10^{-7}$ Torr vacuum.

[5] Assumed not to change during annealing, although we sometimes observed a few percent thickness change of the films.

[6] Cox-Smith *et al.* (1985) found no significant substrate temperature dependence of the sound velocity in *e*-beam *a*-Ge and *e*-beam *a*-Si films up to 125 and 235 °C, respectively. Vacher *et al.* (1980) found no significant reduction (less than 1%) in sound velocity in sputtered *a*-Si and $Si^+$-implanted *a*-Si after annealing to 300 °C. Testardi and Hauser (1977) observed ~5–10 % increase in sound velocity of sputtered *a*-Ge after 500 °C annealing. Since the changes are either small or none, we assume the sound velocity does not change for all the annealing studies presented in the table.

[7] In sputtered *a*-Ge films, Testardi and Hauser (1977) measured the speed of sound on 1–4-$\mu$m-thick films using a quartz resonator and reported a velocity ~56–60 % of that expected for bulk crystalline films. The value listed here is 60% of the transverse speed of sound of bulk crystalline Ge: $v_t = 3.2 \times 10^5$ cm/s. For HWCVD *a*-Ge:H film, we assume a 10% decrease from the same bulk crystalline value. This is the same percentage as that of HWCVD *a*-Si:H for the reason given in note 18.

[8] Liu *et al.* (2002).

[9] Williamson *et al.* (1995) concluded, based on the literature, that *e*-beam or sputtered *a*-Si films have a density deficit relative to crystalline Si of more than 10%, depending on film deposition. The value listed is 90% of the bulk crystalline density (2.33 g/cm$^3$).

[10] Cox-Smith *et al.* (1985) reported a speed of sound in *e*-beam *a*-Si films 77% that of the bulk transverse speed of sound ($v_t = 5.4 \times 10^5$ cm/s), which yields the value listed.

[11] Found to be independent of magnetic field up to 6T (1.5–20 K); Metcalf *et al.*, 2000.

[12] Vacher *et al.* (1980) observed, through Brillouin scattering, the speed of sound on sputtered *a*-Si to be 71% of that of bulk crystalline Si, while Testardi and Hauser (1977) obtained ~53% using a quartz resonator. Using rf sputtering in a $H_2$-Ar mixture, Jiang *et al.* (1990) found through Brillouin scattering that *a*-Si with less than 0.1 at. % H has a speed of sound ~90%. The value quoted here is 71% of the bulk crystalline value ($v_t = 5.4 \times 10^5$ cm/s).

[13] Custer *et al.* (1994) and Laaziri *et al.* (1994) reported independently a deficit of 1.8% to that of bulk crystalline Si. The highest ion-implantation dose was $5 \times 10^{15}$ cm$^{-2}$ in both cases.

[14] In $Si^+$-implanted *a*-Si, Vacher *et al.* (1980) observed through Brillouin scattering an 11% reduction of the speed of sound from the bulk crystalline value. Tan *et al.* (1972) had earlier measured a 10% reduction with the vibrating reed technique. The value quoted here is 89% of $v_t = 5.4\times10^5$ cm/s.

[15] From thermal-conductance measurements below 1 K, the same $C_t = 0.22\times10^{-4}$ was determined (Liu, Vu, *et al.*, 1998).

[16] 300 °C, 1 h, in $3\times10^{-7}$ Torr vacuum.

[17] Remes *et al.* (1997) reported an increase of $\rho$ with decreasing hydrogen contents in HWCVD *a*-Si:H. For the films with ~1 at. % H, $\rho = 2.28$ g/cm$^3$. For PECVD *a*-Si:H with 9 at. % H, $\rho = 2.225$ g/cm$^3$.

[18] Because of a density deficit similar to that of the implanted *a*-Si (Remes *et al.*, 1997), we assume $v_t$ to be comparable as well. See note 14.

[19] Liu, White, *et al.* (1997).

[20] Annealing was done at 500 °C and $3\times10^{-7}$ Torr for 24 h. The temperature was ramped up at 1 °C/min and down at 0.5 °C/min to avoid both explosive evolution of hydrogen during heating and quenched-in metastable defects during cooling. After the annealing, no hydrogen is left and the material remains amorphous (Remes *et al.*, 1997).

[21] Remes *et al.* (1997) reported an increase of $\rho$ upon annealing at 500 °C for 24 h to 2.295 g/cm$^3$. Hydrogen is completely removed.

[22] It is assumed that the isotopic substitution makes little difference.

[23] Liu *et al.* (1999).

[24] Through Brillouin scattering, Xia *et al.* (1991) observed a sound velocity reduction to 87% of that of the bulk crystalline value in Rayleigh waves on PECVD *a*-Si:H. The value listed is 87% of $v_t = 5.4\times10^5$ cm/s.

[25] Annealing was done at 400 °C and $3\times10^{-7}$ Torr for 15 min. The hydrogen evolution is expected to be negligible. The film is still amorphous after this anneal.

[26] Earlier review of PLD *a*-C films by Voevodin and Donley (1996) concluded a mass density of 2.4 g/cm$^3$. However, more recent results indicate a mass density of 2.95 g/cm$^3$ for films deposited by Excimer laser source (Sullivan *et al.*, 1997; Witke *et al.*, 1999). Annealing leads to a reduction of mass density up to 2.85 g/cm$^3$ (Sullivan *et al.*, 1997). Mass density as high as 3.26 g/cm$^3$ (Witke *et al.*, 1999) was reported as well.

[27] For a review of early measurements of the elastic moduli in various *a*-C films, see Voevodin and Donley (1996). Ferrari *et al.* (1999), reported recently from Brillouin scattering measurements a shear modulus $G = 33.7\times10^{10}$ Pa. This yields a sound velocity of $1.0\times10^6$ cm/s, in agreement with that of polycrystalline diamond (Field, 1994).

[28] 500 °C for 20 min in Ar atmosphere, which leads to almost complete release of compressive stress and an increase of $sp^2$ content (Sullivan *et al.*, 1997).

Because of the simple connection between the experimental values of $Q_0^{-1}$, $\alpha$, and $\beta_0$, and the tunneling strength $C$ given in Eqs. (15)–(20), there is no need to list any of those three experimental values in the tables. They can be easily calculated from the tunneling strengths given in the tables. [We mention as an aside that in some of the tables, e.g., Tables III and IV, $v_l = 2v_t$ is sometimes a better approximation. Since, however, this will decrease the prefactors in Eqs. (19) and (20) by less than 10%, we decided to ignore this difference and to use the approximation in Eq. (18) throughout to determine $C_t$ and $\lambda/\ell$ from the experimental $\beta_0$.]

From an inspection of Tables I–IV, we can draw the following conclusions.

(i) For a given chemical composition, the tunneling strengths $C_l$ and $C_t$ have very similar magnitudes, independent of the samples and the measuring techniques used. We add here that, through measurements of the low-frequency elastic loss of *a*-SiO$_2$, PMMA, PdSiCu, PdZr, and CuZr, Tietje *et al.* (1986) extended the frequency range in which the tunneling strength $C_l$ was measured to $10^{-3}$ Hz, i.e., by five orders of magnitude. The results were very close to the values of $C_l$ listed in the tables and shown in Fig. 10. Hence $C_l$ has been shown to be frequency independent from $\sim10^{-3}$ Hz to $\sim10^9$ Hz, and $C_t$ from $\sim10^2$ to $\sim10^{11}$ Hz. The two quantities together show a frequency independence over 14 orders of magnitude, a truly astounding range.

(ii) A more careful inspection of the statement made in (i) reveals a small systematic increase of $C$ with the acoustic frequency used for the measurement (see, for example, Table I, notes 4 and 10 for *a*-SiO$_2$, and also the illustration in Fig. 3). A similar frequency dependence has also been observed for PMMA (see Table III, note 6). Over the frequency ranges studied, five orders of magnitude for *a*-SiO$_2$ and nine for PMMA, $C$ has been found to increase approximately by a factor of 3, as reviewed by Topp and Cahill (1996). Such a variation is not contained in the tunneling model, nor is it observed in the thermal conductivity measurements, in which the heat is carried by phonons with frequencies ranging from $10^{10}$ to $10^{11}$ Hz, yet $C$ has been found to be close to values found in the kHz frequency range of the acoustic measurements.

(iii) The tunneling strength $C$ appears to be entirely independent of trace impurities, as studied thoroughly in *a*-SiO$_2$. Variations of the OH contents from less than 1 ppm to over 1000 ppm caused no measurable effect in acoustic measurements (Table I, notes 4, 6, and 10), and variations from a few ppm to over 1000 ppm resulted in a reduction of less than 10% in thermal conductivity measurements (Table I, note 8). It is interesting to note in this connection that the density of tunneling states detected by dielectric measurements does show a clear dependence on the OH concentration. von Schickfus and Hunklinger (1976) reported an increase of the dielectric tunneling strength $C_{\mathrm{diel}}$, as detected electrically, by a factor of 5 as the OH concentration in *a*-SiO$_2$ increased from less than 1.5 ppm (by weight) to 1200 ppm. It was concluded that not all elastically coupled two-level systems interact electrically (Arnold *et al.*, 1976),

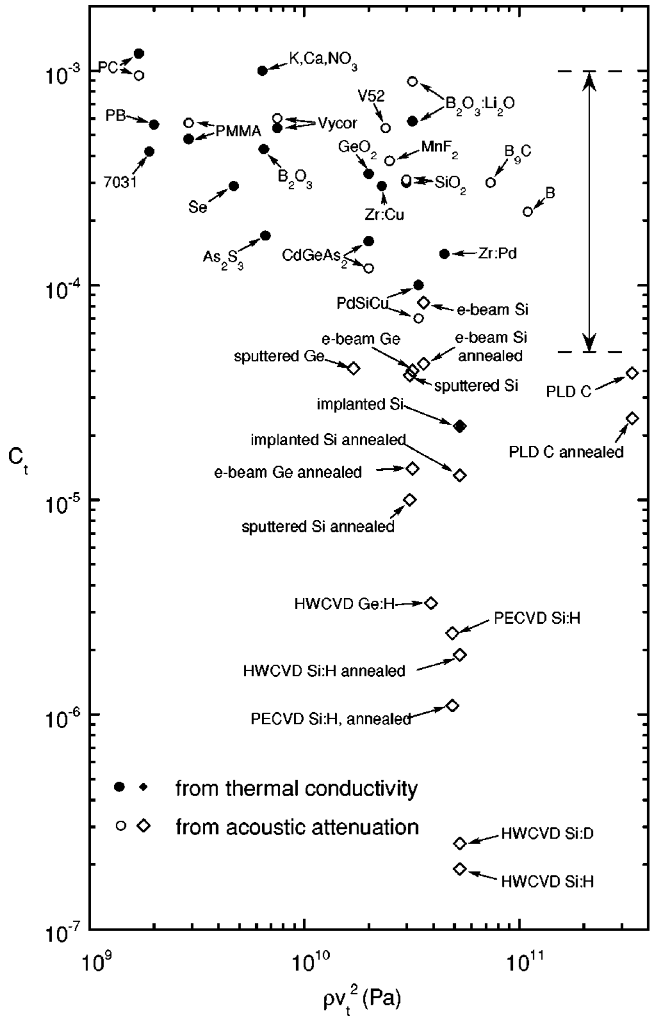

FIG. 11. Transverse tunneling strength $C_t$ for the films of amorphous Si, Ge, and C listed in Table V. The data points between the two horizontal dashed lines connected by the double arrow, representing the glassy range, were selected from the points shown in Fig. 9.

and it was alternatively suggested that additional tunneling states are being produced by hindered rotations (Phillips, 1981a).

(iv) The tunneling strength is essentially independent of doping in the range of 1 mol % and higher, as shown, for example, in $(B_2O_3)_{0.95}(K_2O)_{0.05}$ (Table II, note 9) or in $(KNO_3)_{0.60}[Ca(NO_3)_2]_{0.40}$ doped with 3.4 wt % water, which was found to lower the thermal conductivity by only 7% (Table II, note 32).

(v) Heat treatment also has little influence on the tunneling strength $C$. Thermal conductivity of Suprasil W (5 ppm OH) and Suprasil I (1200 ppm OH) was found to increase 15% at most upon annealing up to 1400 °C (see Table I, note 8; for more examples, see Table II, note 21). Exceptions are amorphous metals, in which $C$ has been found to decrease by as much as a factor of 3 upon annealing (Table IV, notes 14, 16, 21, and 44) to the smallest value reported for bulk amorphous solids, $C = 0.35\times10^{-4}$ (PdSiCu).

The experimental findings reviewed so far can be summarized, if we use the tunneling model, with the single statement that all amorphous solids have tunneling strength $C_i$ ranging from $\sim10^{-4}$ to $\sim10^{-3}$, which is intrinsic and practically independent of chemical composition or sample preparation. This is the universality referred to in the Introduction.

According to Eq. (14), $C_i$ is inversely proportional to the modulus. However, the fact that $C_i$ is nearly independent of $\rho v_i^2$, which varies by almost a factor of $10^4$ for the different solids studied, means that $\overline{P}\gamma^2$ is proportional to this modulus. It follows that the tunneling states (through $\overline{P}\gamma^2$) are closely connected to the Debye waves (through $\rho v_i^2$) in these solids. In other words, the tunneling states (i.e., the postulated defects) and the plane waves (i.e., the elastic background of their host) are somehow connected. As a possible explanation, strain interactions between two-level systems have been considered. However, by considering the interaction between quasiharmonic oscillators in the *soft potential model* as reviewed by Parshin (1994a, 1994b), this would lead to a value of the tunneling strength $C$ of order unity, i.e., a value three orders of magnitude too large. Parshin (1994a) argued that the discrepancy can be removed if one considers that only a small fraction of the quasiharmonic oscillators are tunneling defects.

After having seen that all amorphous solids reviewed so far, of more than 60 different chemical compositions, have the same tunneling strength, roughly within an order of magnitude, it seems only natural to conclude that all amorphous solids behave in the same way. However, as we shall see in the next section, this generalization has recently been shown to be inaccurate.

## IV. EXCEPTIONS TO UNIVERSALITY

Numerous investigations have shown the tunneling model to provide a valid description of the vibrational and thermal properties of amorphous solids. The review presented here so far has given strong evidence for what we call universality, i.e., a tunneling strength of nearly the same magnitude, independent of chemical composition, sample preparation and treatment, microscopic structure, or the nature of the atomic bonding. We emphasize this universality is not a part of the model. However, recent work has shown instances in which the magnitude of the tunneling strength $C_t$ fell clearly below the so-called glassy range. This is illustrated above in Fig. 4, in which the temperature-independent internal friction of several $a$-Si films can be seen to be much smaller than that of the other amorphous solids. So far, these exceptions have been observed only in amorphous films of covalently bonded, fourfold-coordinated Si, Ge, and C. These findings are summarized in Table V and are illustrated in Fig. 11. Chemically pure films, as deposited, have tunneling strengths that still lie fairly close to the glassy range indicated with typical data taken from Fig. 9 and with the two horizontal dashed lines connected by the double arrow. After annealing, however, the tunneling strength of the amorphous films decreases to a value smaller than this range by as much as a factor of 10.

TABLE VI. Disordered crystals and one thermally equilibrated quasicrystal in which glasslike lattice vibrations have been established convincingly (however, see the text). The Si:B$^+$ crystal was disordered by ion implantation but remained crystalline. Column 1: material name and chemical composition used in Fig. 12; column 2: shear modulus $G=\rho v_t^2$; columns 3 and 4: tunneling strength $C_t$ from internal friction (or sound attenuation) plateau and from low-temperature thermal conductivity, respectively; columns 5–13: experimental techniques used to establish the glasslike lattice vibrations. They are: $Q_0^{-1}$, internal friction or sound attenuation plateau; $\Lambda_{\mathrm{LoT}}$, low-temperature $(\beta_0 T^2)$ thermal conductivity; $\Lambda_{\mathrm{HiT}}$, minimum thermal conductivity; $\delta v/v_0$, low-temperature speed of sound; $C_v$, linear specific-heat anomaly; $\epsilon$, dielectric constant; H.R., long-time heat release; $C(t)$, short-time thermal relaxation; Sat., evidence that the lattice vibrations are saturated and hence independent of details of the chemical composition (or the implantation dose, respectively). Column 14: references. Many original references can be found in the references cited, particularly in the works of Topp and Cahill (1996) and Van Cleve *et al.* (1994).

| | | $C_t$ $(10^{-4})$ | | Glasslike lattice vibrations established by | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Material | $G$ $(10^{10}$ Pa) | fr. $Q_0^{-1}$ | fr. $\Lambda$ | $Q_0^{-1}$ | $\Lambda_{\mathrm{LoT}}$ | $\Lambda_{\mathrm{HiT}}$ | $\delta v/v_0$ | $C_v$ | $\epsilon$ | H.R. | $C(t)$ | Sat. | Ref. |
| $(\mathrm{BaF_2})_{0.54}(\mathrm{LaF_3})_{0.46}$ | 3.2 | 2.7 | 2.8 | √ | √ | √ | √ | √ | | | | √ | 1 |
| $(\mathrm{CaF_2})_{0.74}(\mathrm{LaF_3})_{0.26}$ | 4.1 | 0.61 | 0.76 | √ | √ | √ | √ | | | | | √ | 1 |
| $(\mathrm{KBr})_{0.25}(\mathrm{KCN})_{0.75}$ | 0.28 | 4.5 | 3.8 | √ | √ | √ | √ | √ | √ | | √ | | 2 |
| $(\mathrm{NaCl})_{0.24}(\mathrm{NaCN})_{0.76}$ | 0.39 | 21[3] | 28 | √ | √ | √ | √ | √ | | | | | 4 |
| $(\mathrm{NaCN})_{0.75}(\mathrm{KCN})_{0.25}$ | 0.25 | | 8.6 | | √ | √[5] | | √ | | | √ | √[6] | 7 |
| $\mathrm{Rb}_{1-x}(\mathrm{NH_4})_x\mathrm{H_2PO_4}$ | 0.93[8] | | 2.4 | | √ | √[9] | | √ | | | √ | √[10] | 11 |
| $(\mathrm{ZrO_2})_{0.89}(\mathrm{CaO})_{0.11}$ | 6.7 | 2.0 | 1.3 | √ | √ | √ | √ | | | √ | | | 12 |
| $(\mathrm{ZrO_2})_{1-x}(\mathrm{Y_2O_3})_x$[13] | 7 | | 1.0 | | √ | √ | | √ | | | | √[14] | 15 |
| $\mathrm{GdB}_{62.5}$ | 17 | 1.5 | 2.7 | √ | √ | √ | | | | | | | 16 |
| $\mathrm{YB}_{62.5}$ | 19 | 1.6 | 1.9 | √ | √ | √ | √ | √ | | | | √[17] | 18 |
| $\mathrm{Sr}_{1-x}\mathrm{Ba}_x\mathrm{Nb_2O_6}$ | 5.8 | 10[19] | 2.6 | √ | √ | √ | √ | √ | √ | √ | | √[20] | 21 |
| Na-$\beta$-Alumina | 5.5 | 1.0[22] | 0.4 | √ | √ | √[9] | √ | √ | √ | | | | 23 |
| $\mathrm{Ti}_{0.63}\mathrm{Nb}_{0.37}$ | 4.2 | 6.2 | 4.7 | √ | √ | 24 | √ | | 25 | | | √[26] | 27 |
| $\mathrm{Ti}_{0.67}\mathrm{V}_{0.33}$ | 3.3 | 4.6 | 3.9 | √ | √ | 24 | √ | | 25 | | | √[28] | 27 |
| $\mathrm{Zr}_{0.80}\mathrm{Nb}_{0.20}$ | 2.5 | 3.2 | 4.8 | √ | √ | 24 | √ | √ | 25 | | | √[29] | 30 |
| *b*-Plagioclase | 2.1[31] | | 2 | | √ | √ | | | √ | | | | 32 |
| Corning 9623 | 3.4 | 4.8 | 4.5 | √ | √ | √ | √ | | | | | | 33 |
| Si, B$^+$ implanted | 5.3[34] | 0.22 | 0.22 | √ | √ | | | | | | | √ | 35 |
| $\mathrm{Al}_{72.1}\mathrm{Pd}_{20.7}\mathrm{Mn}_{7.2}$ | 7.3 | 0.38 | 0.57 | √ | √ | √ | √ | | | | | | 36 |

[1] Topp and Cahill (1996); Topp *et al.* (1999).

[2] Berret *et al.* (1985); DeYoreo *et al.* (1986); Topp and Cahill (1996).

[3] The relatively large error $(\pm 8\times 10^{-4})$ reflects the small sample used.

[4] Watson and Pohl (1995).

[5] Data probably doubtful because of thermal radiation effects (DeYoreo *et al.*, 1986).

[6] This claim is based on the observation that in $(\mathrm{NaCN})_{1-x}(\mathrm{KCN})_x$ the magnitude of the linear specific-heat anomaly varied very little between $x=0.19$ and $x=0.85$ (Mertz *et al.*, 1990).

[7] DeYoreo *et al.* (1986); Mertz *et al.* (1990).

[8] Berret *et al.* (1991) gave the Debye-$T^3$ prefactor $c$ [see Eq. (5)] based on elastic measurements. From this, a Debye velocity $v=2.44\times 10^5$ cm/s was derived, and $v_t$ was approximated as $0.9v$.

[9] Not calculated, but temperature dependence and magnitude seem glasslike.

[10] Based on the fact that identical results were obtained for $x=0.35$ and $x=0.72$. Significantly, in the deuterated RADP, no glasslike lattice vibrations were found.

[11] Berret *et al.* (1991).

[12] Abens *et al.* (1996); Topp and Cahill (1996); Topp (1997).

[13] $0.10<x<0.18$.

[14] This claim is based on the observation that the low-temperature thermal conductivity is independent of $x$ in the range studied.

[15] Ackerman *et al.* (1981); Topp (1997).

[16] Medwick *et al.* (1994); Topp and Cahill (1996).

[17] This claim is based on the observation that both the thermal and the elastic properties show no dependence on the amount of boron in the (admittedly small) range studied.

[18] Cahill *et al.* (1989); Medwick *et al.* (1994); Topp and Cahill (1996).

[19] Experimental error? The value of $C_t$ derived from speed of sound does agree with that derived from thermal conductivity, thus making this value more credible than the one determined from $Q_0^{-1}$.

[20] This claim is supported most strongly by the observation that the same thermal conductivity was observed in two crystals with $x=0.55$ and $x=0.39$ (Hegenbarth, 1994).

[21] DeYoreo *et al.* (1985); Hegenbarth (1994); Mattausch *et al.* (1996).

[22] Wave propagation perpendicular to the $c$ axis. Value given here is an average between waves polarized parallel and perpendicular to the $c$ axis.

[23] Anthony and Anderson (1976); Doussineau *et al.* (1980); Strom (1983).
[24] Not applicable in the normally conducting electronic state.
[25] Not applicable in the superconducting solid.
[26] Nb concentration varied from 0.37 to 0.52.
[27] Van Cleve *et al.* (1994); Topp and Cahill (1996).
[28] V concentration of 0.2 and 0.33 led to nearly identical low-temperature internal friction.
[29] Nb concentrations varied from 0.05 to 0.30 with nearly identical low-temperature internal friction.
[30] Van Cleve *et al.* (1994).
[31] Average transverse speed of sound was taken as $2.8\times10^5$ cm/s as given by Linvill (1987, p. 139), based on sound-velocity data on similar feldspars (Ryzhova, 1964). See also Cahill *et al.* (1992). Mass density: 2.71 g/cm$^3$ (Stewart *et al.*, 1966, as quoted in Linvill, 1987, p. 200).
[32] MacDonald and Anderson (1985); Linvill (1987); Cahill *et al.* (1992).
[33] Stephens (1976); Cahill *et al.* (1991); Raychaudhuri and Pohl (1991).
[34] We assume that the mass density and sound velocity are the same as the Si$^+$-implanted $a$-Si; see Table V, notes 13 and 14.
[35] Liu, Vu, *et al.* (1998).
[36] Thomspon, Vu, and Pohl (2000).

Even smaller values of $C_t$ are observed in hot-wire chemical-vapor-deposited (HWCVD) $a$-Ge containing 1 at. % H, and in plasma-enhanced chemical-vapor-deposited (PECVD) $a$-Si containing 10 at. % H. The lowest values, 300 times below the glassy range, have been observed in HWCVD $a$-Si containing 1 at. % H or D. Annealing to remove this H or D raises $C_t$ by a factor of 10, but still to a value 30 times smaller than the glassy range.

It should be noted here that the value of $C_t$ was determined on all of these films solely from the temperature-independent low-temperature internal friction plateau observed on double-paddle oscillators carrying these amorphous solids as thin films and vibrating with a torsional mode with a frequency of ~5 kHz. The only film for which the validity of the tunneling model was tested by using a different frequency was the self-implanted silicon film, in which scattering of thermal phonons was measured; this was found to be in quantitative agreement with the value predicted by the model (Liu, Vu, *et al.*, 1998).

When the tunneling model was first proposed, it was suggested that in fourfold-coordinated solids like $a$-Si the atoms would be overconstrained and thus unable to tunnel between different equilibrium positions (Phillips, 1972). The observation that in pure $a$-Si, $a$-Ge, and $a$-C the internal friction is smaller than in all other amorphous solids can therefore perhaps be taken as an indication that the tunneling entities are indeed atoms which are more constrained in these solids than in other solids. However, the removal of the tunneling states requires the presence of hydrogen, which is known to be distributed quite nonuniformly (Wu *et al.*, 1996) and which should thus lead to large disorder in at least a fraction of the solid. This immediately raises the question of why these disordered regions should not contain tunneling atoms.

## V. TUNNELING STRENGTH FOR DISORDERED CRYSTALS AND FOR A QUASICRYSTAL

As stated frequently in earlier work, even the physical nature of the tunneling entities is not yet fully understood. It has therefore been argued that if states similar to those common in amorphous solids could also be found in disordered crystals, it might be easier to understand tunneling in these crystals and thus help to understand that in the amorphous solids. Examples demonstrating the transition of thermal conductivity and internal friction from a behavior typical of that of a crystal to that of an amorphous solid were shown above in Figs. 5 and 6.

Since the first superconducting alloy, $Zr_{1-x}Nb_x$, was found to have such glasslike excitations (Lou, 1976), a large number of crystalline solids with the same excitations have been reported, and it seems useful to summarize those here. Their selection is not simple. If some of their properties resemble those of amorphous solids while others do not, clearly the compound should be rejected. If, however, no such disqualifying properties have been reported, the decision of how many independent measurements are required to establish convincingly glasslike excitations in a crystal is subjective, and so is the collection of crystals listed in Table VI. Checkmarks indicate whether glasslike excitations have been found with a specific technique. The most significant column is that labeled "saturated," which indicates that some of the glasslike excitations have become independent of the amount of disorder for sufficiently large disorder, usually the doping or the implantation dose. Only in a small number of crystals is such a saturation established convincingly, since it is difficult to distinguish a mere maximum in a certain concentration range from a true saturation. The most convincing evidence of saturation in disordered crystal was probably established in boron-implanted silicon, in which the glasslike excitations remain unchanged over two orders of magnitude of implantation dose (Liu, Vu, *et al.*, 1998).

The observation of an isotope effect in $Rb_{1-x}(NH_4)_xH_2PO_4$ (Berret *et al.*, 1991) is noteworthy. Replacement of the hydrogen by deuterium removed the glasslike lattice vibrations. This was interpreted as evidence that the low-energy excitations result from the tunneling of individual atoms or ions, protons in this case, while the deuterons do not tunnel. Note also the bottom line in Table VI: the icosahedral AlPdMn alloy is neither a disordered crystal nor an amorphous solid. Yet this thermally equilibrated quasicrystal also appears to

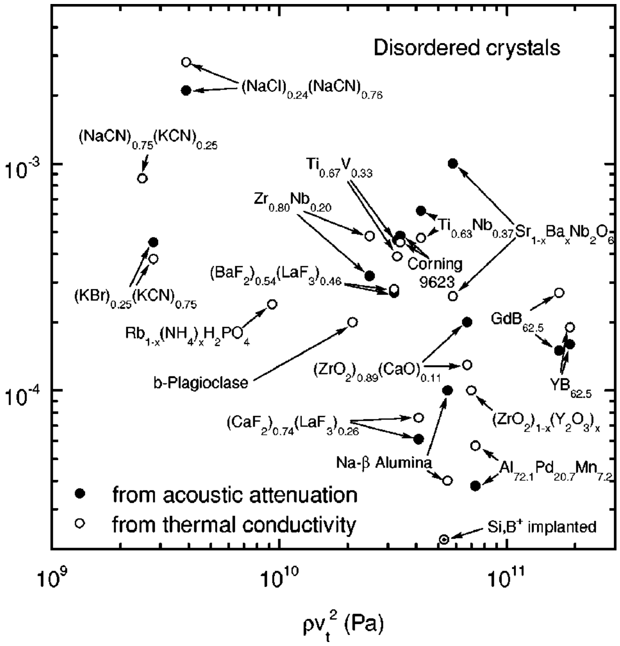

FIG. 12. Transverse tunneling strength $C_t$ for the crystals with glasslike lattice vibrations listed in Table VI. Note also the data for the single grain quasicrystalline $Al_{72.1}Pd_{20.7}Mn_{7.2}$.

have glasslike excitations (Thompson, Vu, and Pohl, 2000). Many other quasicrystals have been measured by the variation of speed of sound (Bert *et al.*, 2002). Whether glasslike lattice vibrations are common to thermally equilibrated quasicrystals is, however, still an open question. For instance, Gianno *et al.* (2000) recently concluded from thermal conductivity measurements on single-grain $Y_{8.6}Mg_{34.6}Zn_{56.8}$ that no clear evidence was obtained for their existence.

Figure 12 shows the tunneling strength $C_t$ as determined for the solids listed in Table VI. Again, $C_t$ is close to the range found in amorphous solids, $10^{-4} < C_t < 10^{-3}$.

Finally, we should mention one other class of crystals which appears to have glasslike excitations. These are the so-called orientational glasses, pure crystalline solids in which the molecules have random orientations. A good example is ethanol ($CH_3CH_2OH$). In its orientational glass phase, Talon *et al.* (2002) have recently reported a linear specific-heat anomaly identical to that observed in the amorphous phase. There are, however, no low-temperature acoustic or thermal conductivity measurements available at this time.

## VI. SUMMARY AND CONCLUSION

Inspection of close to 100 studies of thermal conductivity and acoustic attenuation in amorphous solids has confirmed a striking similarity, with few exceptions so far. This similarity can be expressed in two ways.

(a) In every one of these solids, the ratio $\lambda/\ell$ of the wavelength $\lambda$ of the elastic wave to the mean free path $\ell$ of the energy attenuation is independent of $\lambda$ over at least nine orders of magnitude, to within a factor of less than 2 or 3. For different solids, $\lambda/\ell$ ranges from $\sim 10^{-2}$ to $10^{-3}$.

(b) In the formulation of the tunneling model, the frequency-independent tunneling strengths $C_i$ range from $\sim 10^{-3}$ to $10^{-4}$ for the different solids.

The only exceptions to this otherwise universal behavior have been found in certain films of *a*-Si, *a*-Ge, and *a*-C, which suggests that in ideally fourfold-coordinated lattices no tunneling states may exist, since the lattice is overconstrained. However, the fact that the largest deviation from universality is found in hydrogenated films that are at least in part highly disordered indicates that the nearly complete absence of tunneling states must have yet another cause. The fact that the low-energy excitations in numerous disordered crystals resemble very closely those found in amorphous solids also strongly suggests a picture in which lack of long-range order *per se* is not the cause of the low-energy excitations. But in that case, what is important? Can the soft potential model (Parshin, 1994a, 1994b) be used to explain these observations? At this time, only speculations have been proposed. See, for example, Liu, Vu, *et al.* (1998) and Pohl *et al.* (1999). However, speculations do not belong in this paper, which is devoted to reviewing the facts. Interested readers may want to look at these references or, better still, try their own imaginations.

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