

## ULTRASONIC ATTENUATION IN FUSED SILICA AT HIGH FREQUENCIES AND LOW TEMPERATURES

W. Arnold, J. Baumann, P. Berberich, S. Hunklinger, P. Leiderer, R. Nava<sup>1)</sup> and  
K. Dransfeld

Physik Department E 10 der Technischen Universität, München, Germany

Abstract

Measurements of the attenuation of longitudinal and transverse sound waves of frequencies between 30 MHz and 24 GHz were carried out at temperatures from 1.1 K to 10 K in fused silica. In the upper temperature range the attenuation can be described by relaxation processes. The absorption at lower temperature is compared with current theories. It is found that our high frequency data are not in a satisfactory agreement with these theories.

Introduction

At low temperatures the thermal behaviour of dielectrics is determined by phonons of long wavelength. For these phonons it should be possible to consider the regular lattice of a crystal as well as the random network of a glass as an elastic continuum, since phonons are characterized by the properties of the substance averaged over a distance comparable to the phonon wavelength. For this reason it is surprising that especially at low temperatures the thermal properties of amorphous solids are quite different from those of crystals [1]. For example, the magnitude of both the specific heat  $C_v$  and of the thermal conductivity  $\kappa$  in amorphous substances are nearly independent of the chemical composition or of impurities in contrast to the behaviour of crystals. Furthermore experimental results show that for all non-crystalline solids investigated so far  $C_v$  is nearly proportional to  $T$  and  $\kappa$  proportional to  $T^{1.6-1.8}$  at temperatures below 1 K. New results and a summary of these aspects were recently given by Zeller and Pohl [2].

Theoretical attempts have already been made to explain the temperature dependence of  $\kappa$  (Fulde and Wagner (FW) [3] and Anderson, Halperin and Varma (AHV) [4]) and  $C_v$  ([3,4] and Rosenstock [5]). It is, however, still completely unknown, why these quantities are of the same order of magnitude in widely different materials. The existing theories [3,4] also contain a treatment of the ultrasonic absorption in amorphous substances, predicting the temperature and frequency dependence as well as the magnitude of the attenuation. Therefore ultrasonic experiments are particularly well suited to check these theories describing the thermal properties.

Experimental results

We carried out such measurements in fused silica (Suprasil [6]) using longitudinal and transverse sound waves at frequencies between 30 MHz and 24 GHz and down to temperatures of 1.1 K. Fig. 1a shows our results obtained for transverse waves at 135, 222, 495 and 960 MHz whereas Fig. 1b represents data from our experiments with longitudinal sound waves at 30, 488, 939 and 2035 MHz. These experiments with both polarizations were performed using conventional ultrasonic techniques and several samples ranging in length between 2.5 and 1 cm. In all ultrasonic measurements (except those at 30 MHz, where we used standard piezoelectric quartz plates) we employed evaporated crystalline CdS - layers [7] as transducers.

Fig. 2 represents the data of our 24.3 GHz experiment. Here we used a light scattering technique described earlier [8]. Only longitudinal phonons were generated by stimulated Brillouin scattering. The temporal decrease of the number of these phonons was determined from the decreasing scattering intensity of a whole sequence of delayed light pulses. 2)

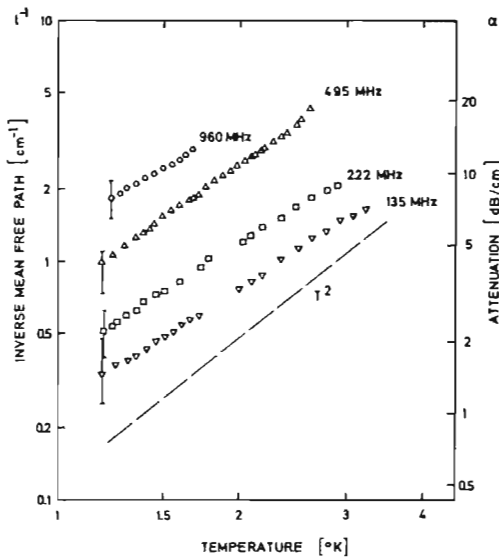


Figure 1a

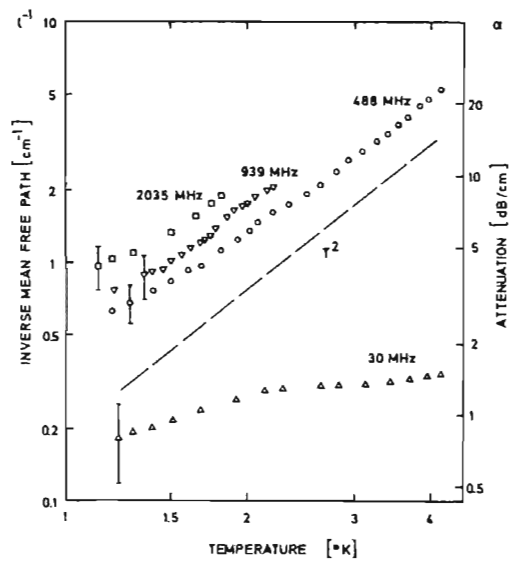


Figure 1b

Attenuation of sound waves in fused silica obtained by conventional ultrasonic technique. The dashed lines are drawn for comparison.

a) Inverse mean free path of transverse sound waves at 135, 222, 495 and 960 MHz.

b) Inverse mean free path of longitudinal sound waves at 30, 488, 939 and 2035 MHz.

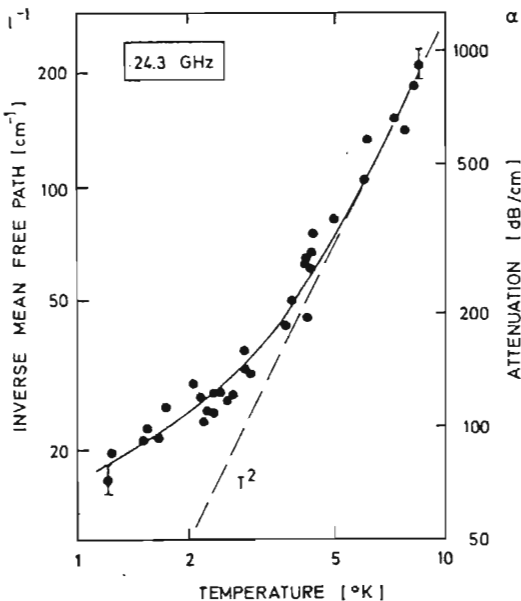


Figure 2

Attenuation of 24.3 GHz longitudinal phonons obtained by light scattering. The error bars represent the standard deviation of each point. The dashed line represents a  $T^2$ -dependence. The solid line is simply a guide for the eye.

In all these experiments between 1 K and 10 K the attenuation does not become temperature independent. Therefore the residual and the temperature dependent absorption could not be determined separately. Since only a few echoes of the sound pulse could be observed, we are only able to give an estimate of the magnitude of the total attenuation. The bars shown in Fig. 1 are representative of the uncertainty in the magnitude of the absolute attenuation. The increase in attenuation with temperature was, however, determined within a relative error smaller than 5%.

In the experiment using the light scattering technique the phonon lifetime is determined only in the small volume where the laser light is focused. Optical absorption may in principle cause a serious increase of the temperature within this volume during the measuring process. After estimating this temperature rise using optical absorption data at room temperature [9] we cannot exclude that the deviation from the  $T^2$ -relation at low temperatures shown in Fig. 2 may be caused by such a heating mechanism.

Our main results may be summarized as follows:

- For all frequencies from 30 MHz up to 24 GHz a temperature dependence of  $\alpha \propto T^{0.8-2.0}$  was observed in the respective temperature range of measurement. The exponent increases with increasing temperature and frequency.
- At constant temperature the frequency dependence varies between  $\alpha \propto \omega^{0.3}$  and  $\alpha \propto \omega^{0.8}$ .
- For transverse sound waves the ultrasonic attenuation is only about twice as high as for longitudinal waves of the same frequency.

We found good agreement between our data at 30 MHz and the results of Krause and Kurkjian [10] and between our 488 MHz and 938 MHz measurements and the data of Jones, Klemens and Rayne [11]. In the latter case we took into account that these results were normalized to the absorption value at 1.4 K because of the special technique used in this experiment. At 24 GHz we observed a certain deviation from the results of Heinicke, Winterling and Dransfeld [12] below 5 K which is probably due to the higher accuracy of our improved method especially at relatively low values of attenuation.

### Comparison with theory

All these facts indicate that the main part of the attenuation even at 1.1 K is caused by the relaxation process observed in [10, 11, 12]. For the absorption we expect therefore  $\alpha = \alpha_0 \omega^2 \tau / (1 + \omega^2 \tau^2)$ . Here the constant  $\alpha_0$  is called the relaxation strength. For simplicity we assume that there exists only one relaxation time  $\tau$ . For  $\tau$  we may expect an Arrhenius - type temperature dependence  $\tau = \tau_0 \cdot e^{\Delta E / kT}$ , where  $\tau_0$  is a constant and  $\Delta E$  is the activation energy of the relaxation process. The measurements [10, 11, 12] indicate that  $\omega \tau \gg 1$  should be valid in all our experiments. Therefore the absorption is expected to show a temperature dependence between  $\alpha \propto \tau^{-1} \propto e^{-\Delta E / kT}$  in the case of  $\omega \tau \gg 1$  and  $\alpha \propto \tau^0 = T^0$  for  $\omega \tau = 1$ . For the frequency dependence we expect a proportionality between  $\alpha \propto \omega^0$  and  $\alpha \propto \omega$ , respectively. Both predictions are in agreement with our results. For ultrasonic attenuation caused by a relaxation process the ratio of the absorption of transverse sound waves and longitudinal waves should be  $\alpha_t / \alpha_l \approx v_l / v_t$ . Within the accuracy of our measurement this relation holds at 500 MHz, whereas at 1 GHz this ratio is slightly higher.

On the other hand we can get some information about the behaviour of the attenuation at temperatures below 2 K from the theories mentioned at the beginning. According to the theory of FW [3] the following equation for the inverse mean free path of the ultrasonic phonons should hold:

$$l_{FW}^{-1} = 5.9 \cdot 10^{-20} \nu^2 T^{-2/3} \text{ cm}^{-1}$$

This equation was derived from a semi-phenomenological model using an extended form of the propagator for low frequency phonons. All free parameters were fitted to the experimental values of the specific heat and the thermal conductivity.

AHV [4] derived an equation for the ultrasonic attenuation assuming that the position of certain atoms or groups of atoms can be described by a double-well asymmetric potential. The atoms can be either in one well or the other; this situation represents a two level system. Thermal phonons as well as ultrasonic phonons should be resonantly scattered by these two level systems. If we apply the dominant phonon approximation we find the equation

$$l_{AHV}^{-1} = 1.1 \cdot 10^{-8} \nu \tanh \frac{h\nu}{2kT} \text{ cm}^{-1}$$

which reduces to

$$l_{AHV}^{-1} = 2.5 \cdot 10^{-19} \nu^2 T^{-1} \text{ cm}^{-1}$$

if  $h\nu < kT$ .

The model of AHV was extended by Jäckle (J) [13] who included thermally activated transitions between the double-well potentials of random asymmetries. This relaxation process becomes of importance above 1 K. The inverse mean free path for this process is expected to be

$$l_{J1}^{-1} = 0.17 T^3 \text{ cm}^{-1} \quad \text{and} \quad l_{Jh}^{-1} = 1.6 \cdot 10^{-9} \nu \text{ cm}^{-1}$$

for low temperatures ( $\omega\bar{\tau} \gg 1$ ) and high temperatures ( $\omega\bar{\tau} \ll 1$ ), respectively. Here  $\bar{\tau}$  is the mean value of all possible relaxation times.

Let us now discuss in detail our experimental data at 2 and 24 GHz (shown in Fig. 1b and 2) and compare them with theory. We already mentioned above that in our experimental temperature and frequency interval the absorption represents mainly the low temperature part of the relaxation peak observed previously by [10,11,12]. As can be seen from Fig. 1b and Fig. 2 at temperatures and frequencies high enough this attenuation is nearly proportional to  $T^2$ . For a single relaxation process the temperature dependence should become steeper at lower temperatures in contrast to our observations. Therefore we might assume that this deviation is due to the absorption described by FW and AHV. In order to check this idea we extrapolate the absorption from high temperatures to lower temperatures. Our low frequency measurements indicate that such a strong temperature dependence as  $T^2$  should be an upper limit, because we observe a temperature dependence less than  $T^2$  even at  $\nu < 500$  MHz although AHV and FW do not predict any noticeable contribution to the absorption in our experiments at such low frequencies.

In Fig. 3 we compare our 2 GHz data with theory: The experimental curve is represented by a solid line whereas its  $T^2$ -extrapolation to lower temperatures as well as the absorption due to the theory of AHV, FW and J are shown as dashed lines. Furthermore the expected total absorption due to the addition of various absorption contributions are represented by dashed-dotted lines. From Fig. 3 it is obvious that our experiment must be carried out at still lower temperatures in order to check the theory of FW at this frequency. The theory of AHV, however, seems to disagree with our measurement already in our present temperature interval. But if we assume that the high temperature absorption is not due to a single relaxation process but is for example described by Jäckle's assumption our experimental value and the theoretical curve are in rough agreement above 1 K.

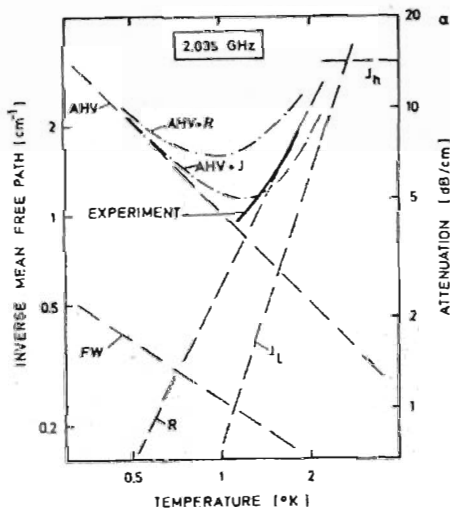


Figure 3

Comparison of the experimental (solid line) and the theoretically expected (dashed-dotted lines) attenuation of 2035 GHz phonons in fused silica. R represents the extrapolated attenuation due to relaxation, which is assumed to be  $\propto T^2$ . FW, AHV,  $J_1$  and  $J_2$  are the attenuation values expected from the theories. AHV and R represents the attenuation predicted by AHV with the contribution from the extrapolated  $T^2$ -relaxation. AHV and  $J_1$  stands for the sum of the predicted attenuation from AHV and  $J_1$ . The solid curve which indicates the experimental results is taken from Fig. 1b.

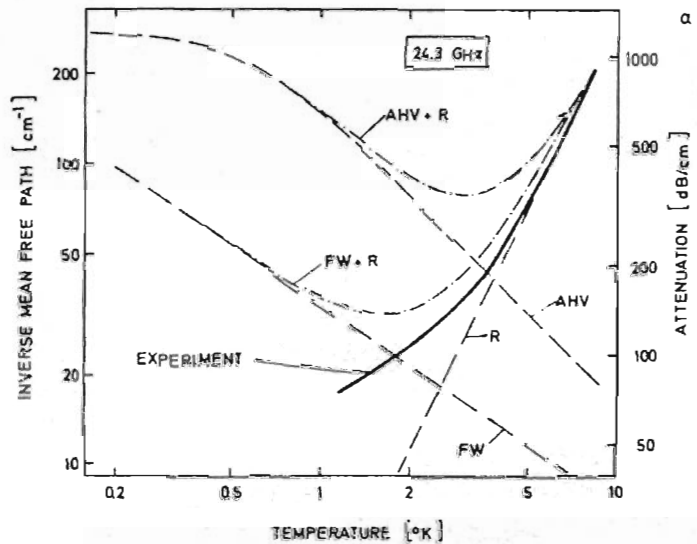


Figure 4

Comparison of the experimental (solid lines) and the theoretically expected (dashed-dotted lines) attenuation of 24.3 GHz phonons in fused silica. R represents the extrapolated attenuation due to relaxation, which is assumed to be  $\propto T^2$ . FW and AHV are the attenuation values expected from the theories. FW and R, and AHV and R give the sum of the attenuations due to relaxation and the theories. The solid curve which indicates the experimental results is taken from Fig. 2.

Fig. 4 shows absorption values at 24 GHz plotted in a similar way as Fig. 3. Jäckle's curves are omitted in this figure since they do not lead to an important modification of the theory of AHV at such a high frequency. Again AHV's theory disagrees with our results but also the theory of FW results in absorption values which are higher than observed. This disagreement may be significant since the experimental data are not corrected for heating of the focal volume.

Our relatively low absorption values may be due to a saturation of the two level systems at high sound amplitudes. This possibility was already pointed out by Heinicke, Winterling and Dransfeld [14], but so far we did not see an indication for such a behaviour. On the other hand both theories describe thermal conductivity and specific heat very well. Therefore our results seem to indicate a different behaviour of thermal and of coherent acoustic phonons.

### Conclusion

Summarizing our experimental results we are able to say that current theories in their present form do not seem to agree with our data. Although it is possible to modify both theories slightly using appropriate velocities of sound rather than a mean value, we do not even in this case find full agreement. In order to decide more clearly whether these theories are valid we are presently extending our experiments to temperatures well below 1 K.

### References

- [1] See for example:  
FLUBACHER P., LEADBETTER A.J., MORRISON J.A., STOICHEFF B.P., J. Phys. Chem. Solids, 1959, 12, 53; LEADBETTER A.J., Physics Chem. Glasses, 1968, 9, 1; LASJAUNIAS J. C., MAYNARD R., THOULOZE D., Solid State Commun., 1972, 10, 215; STEPHENS R.B., CIELOSZYK G.S., SALINGER G.L., Phys. Letters, 1972, 38A, 215.
  - [2] ZELLER R.C., POHL R.O., Phys. Rev., 1971, B4, 2029.
  - [3] FULDE P., WAGNER H., Phys. Rev. Letters, 1971, 27, 1280.
  - [4] ANDERSON P.W., HALPERIN B.I., VARMA C.M., Phil. Mag., 1972, 25, 1.
  - [5] ROSENSTOCK H.B., J. Non-Cryst. Solids, 1972, 7, 123.
  - [6] All samples were made of Suprasil I produced by Heraeus-Schott Quarzschmelze GmbH, 645 Hanau, Germany.
  - [7] BEECHAM D., Rev. Sci. Instr., 1970, 41, 1663.
  - [8] HUNKLINGER S., LEIDERER P. and BERBERICH P. in "Proceedings of the Second International Conference on Light Scattering in Solids", edited by M. Balkanski, Flammarion Sciences, Paris, 1971, p. 453.
  - [9] RICH T.C., PINNOW D.A., Appl. Phys. Letters, 1972, 20, 264; TYNES A.R., PEARSON A.D., BISBEE D.L., J. Optical Soc. Am., 1971, 61, 143.
  - [10] KRAUSE J.T., KURKJIAN C.R., J. Am. Ceram. Soc., 1968, 51, 226.
  - [11] JONES C.K., KLEMENS P.G., RAYNE J.A., Phys. Letters, 1964, 8, 31.
  - [12] HEINICKE W., WINTERLING G., DRANSFELD K., J. Acoust. Soc. Am., 1971, 49, 954.
  - [13] JÄCKLE J., private communication.
  - [14] HEINICKE W., WINTERLING G., DRANSFELD K. in "Proceedings of the Second International Conference on Light Scattering in Solids", edited by M. Balkanski, Flammarion Sciences, Paris, 1971, p. 463.
- 1) Permanent address: Inst. Venezolano de Investigaciones Científicas, Caracas
  - 2) In these experiments the temporal coincidence of both lasers was achieved electronically.
  - 3) According to O.L. Anderson, H.E. Bömmel, J. Am. Ceram. Soc., 1955, 38, 125, this assumption is not strictly valid in fused silica.