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Low energy excitations in amorphous materials: acoustic experiments and comparison with theoretical models*

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Abstract

At low temperatures many properties of amorphous solids are dominated by low energy excitations with a wide spectrum of energies below a few kelvins. These excitations are absent in defect-free crystals and are believed to be caused intrinsically by the irregular structure of the material. It is widely accepted that their origin is the tunnelling motion of atoms or small clusters through the barrier between two adjacent potential wells. Depending on the theoretical model, the required broad spectrum of arbitrarily small energy splittings of the thus-formed two-level systems can have different explanations. According to the tunnelling model and the soft potential model, the distribution of energies comes from the distribution of parameters describing the double-well potentials, whereas in more recent theories a strong phonon-mediated coupling between tunnelling systems is invoked. All these models claim to explain the universal thermal properties of amorphous solids, but only the first two of them are developed far enough to predict also dynamical properties, e.g. the acoustic behaviour, which seem to be more decisive. In this paper the most important and characteristic predictions of the tunnelling model for the temperature dependence of sound attenuation and velocity are reviewed and compared with experimental results. Special attention is given to recent vibrating reed experiments on vitreous silica.

1. Introduction

The low temperature properties of almost any amorphous material are distinctly different from those of comparable crystalline substances (for a comprehensive introduction see e.g. Ref. 1). The specific heat below 1 K for example is considerably larger and varies linearly with temperature T instead of being proportional to T^3 as expected for a Debye solid. Additionally, the acoustic behaviour, i.e. the temperature and frequency dependence of internal friction and sound velocity, exhibits a variety of characteristic features. Together, these properties are called the low temperature anomalies of glasses and are explained by the existence of low energy excitations with energies below about 1 meV.

Despite considerable experimental and theoretical effort, the microscopic nature of these low energy excitations has not yet been disclosed. According to experiments on a wide variety of materials, e.g. insulating and metallic glasses, glassy chalcogenides [2], amorphous and nanocrystalline noble gases [3, 4] and polycrystalline metals [5], a broad distribution in the density of states, extending over several orders of magnitude in energy, is required. Moreover, an additional distribution in relaxation times for excitations with a given energy is necessary to explain particular acoustic experiments and long-time specific heat measurements. The lack of a unique microscopic theory for all amorphous substances is therefore not surprising. Instead, a phenomenological model, the so-called tunnelling model, has been proven to describe very well the low temperature anomalies of glasses. The most important task in this and other more recent phenomenological models is to define the relevant parameters and make plausible their distribution functions necessary for a correct description of (in the best case) all experiments.

In this paper we will briefly introduce some theoretical models in Section 2. In Section 3 a survey of the predictions of the tunnelling model as well as a comparison with experiments on vitreous silica is given.

2. Models

In their pioneering work of 1971 Zeller and Pohl [6] pointed out the great similarity of the thermal properties of various glasses irrespective of their chemical composition. These thermal properties have been described theoretically by several models, but only the tunnelling model [7, 8] was successful in also explaining the results of ultrasonic measurements. In this model it is assumed that the excess specific heat of glasses can be attributed to an ensemble of two-level systems with a broad distribution of level splittings. These two-

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level systems are most likely structural defects which perform a quantum mechanical motion between two equilibrium configurations. Phonons interact strongly with the two-level systems, which explains the small thermal conductivity and various ultrasonic properties of glasses at low temperatures. Among these the most convincing evidence for the existence of two-level systems comes from the saturation of the ultrasonic attenuation [9, 10]. At low temperatures the dominant interaction processes between an ultrasonic phonon and a two-level system are the resonant absorption and emission of phonons. By increasing the acoustic intensity, the population of the upper level is increased, which leads to a decrease and finally a saturation of the absorption. Further proof that the low energy excitations are represented by two-level systems comes from coherent resonance experiments such as phonon echoes [11], similar to the spin echo experiments of spin-$\frac{1}{2}$ systems.

In the tunnelling model it is assumed that the tunnelling particle, consisting of one atom or a certain group of atoms, is characterized by an average mass m residing in a double-well potential with a ground state energy of $\hbar\Omega/2$ in each well. At sufficiently low temperatures only these ground states are of importance. Depending on the overlap of the two wavefunctions, two new common eigenstates are created with an energy splitting E given by $E^2=\Delta_0{}^2+\Delta^2$. Δ_0 is the tunnelling energy given by $\Delta_0=\hbar\Omega\exp(-\lambda)$ with the tunnelling parameter $\lambda=(d/2\hbar)(2mV)^{1/2}$, where d is the distance between the potential minima and V is the barrier height. Because of the random strain of the environment, an asymmetry energy Δ is introduced in addition. Coupling to the thermal bath occurs via modulation of the asymmetry by the strain field e of thermal phonons as $\delta\Delta=2\gamma e$. The deformation potential γ is an appropriate average over the random orientations of the elastic dipoles and only two independent values γ_l and γ_t are kept to describe the coupling to longitudinal and transverse phonons respectively.

To complete the model, certain assumptions about the distribution of parameters of the two-level systems had to be made. With the aim of an explanation of the low temperature thermal properties of glasses, it was proposed that the values of Δ and λ are independent and equally distributed [7, 8]. This is expressed by the distribution function $P(\lambda,\Delta)=\bar{P}=\text{const.}$, which gives an almost constant density of states in energy and thus the experimentally observed linear temperature dependence of the specific heat. The tunnelling model, although proposed originally to explain thermal properties, turned out to provide also astonishingly correct predictions for the acoustic behaviour of glasses at low temperatures. We will discuss this in the next section.

In the early 1980s in Leningrad (now St. Petersburg) an alternative model had been developed [12], the so-called soft potential model. On more principle grounds the potentials of atoms in a random structure were analysed. It was argued that these potentials can be expressed in a power series up to the fourth order of a certain configuration coordinate x as $V(x)=E_0(ax^2+bx^3+x^4)$, where E_0 is an energy of the order of the binding energy. The parameters a and b are widely distributed owing to fluctuations of the structure of the glass. The resulting potential describes the full variety of possible situations: almost crystalline, harmonic configurations, atoms with very soft potentials and also, in a restricted range of values of a and b, atoms which are moving in double-well potentials. It has been shown that the predictions of the soft potential model for the thermal and acoustic properties of glasses at low temperatures are the same as the predictions of the tunnelling model [13]. Apparently the respective distribution functions are equivalent in the range where two-level tunnelling systems are formed with energy splittings of the order of 1 K and below. In the medium temperature range up to some 10 K the soft potential model seems to provide a slightly better description of some acoustic and thermal properties. In this temperature range both models have to be extended to include thermal activation as the dominating process for the dynamics of the bi-stable configurations. In the soft potential model, additionally, localized quasi-harmonic oscillators give rise to further excitations in the energy range above about 1 meV.

More recently a different scenario has been proposed as the origin of the universal occurrence of low energy excitations in glassy matter. It was argued that instead of the original energy splitting of the single defects themselves, interaction between the defects should dominate the energy scale [14]. The strong elastic dipolar interaction should give rise to a spectrum of excitations sufficiently broad to explain the thermal properties of glasses. The appealing advantage of this model is its universal applicability to all kinds of matter. However, since in this model the excitation energy is stored in some kind of diffusing mode, ultrasonic saturation and echoes or other coherent effects, which demand the existence of localized two-level systems, cannot be explained.

This problem seems to be overcome in a further development [15] of these ideas where it is proposed that the mutual elastic interaction between defects leads in general to localization of the particle. However, when the interaction is completely frustrated, localized tunnelling systems are formed instead of collective modes. A broad excitation spectrum and universal properties should emerge from the randomness of the dipolar interaction. This picture is similar to a theoretical study

of interacting tunnelling defects in alkali halides [16]. There a predominantly pairwise interaction is assumed which leads to coupled tunnelling with excitation energies much below the original tunnelling energy. Evidence for the existence of such low-lying excitations was found recently in measurements of the dielectric response of KBr containing CN^- defects [17].

The aim of these recent models is to explain the universality of the occurrence of low energy excitations in amorphous matter and to end up with a spectrum and dynamics of these states in accordance with the tunnelling model. The soft potential model does not predict a deviating behaviour either. It appears therefore sufficient for this review to give a short survey of the main predictions of the tunnelling model about the acoustic properties of glasses along with some instructive experimental results.

3. Predictions of the tunnelling model: comparison with vitreous silica

In Fig. 1 the internal friction of vitreous silica, a-SiO_2, is shown for two different measuring frequencies in the temperature range between 10 mK and room temperature [18]. A similar behaviour is found for a variety of other glasses. At the lowest temperatures the internal friction rises steeply and reaches a plateau at temperatures which shift strongly with frequency. The plateau extends to approximately 4 K where a further steep rise of the internal friction occurs. After passing a maximum around 20 K, the internal friction drops to very small values at room temperature. Upon going to ultrasonic frequencies, the overall behaviour, particularly the height of the internal friction, remains similar. At 1 GHz the plateau region shrinks considerably and degenerates to a shoulder around 4 K. The maximum shifts more slowly to about 50 K and is identified with the so-called 50 K peak, well known since the 1960s. It is caused by thermally activated structural relaxations.

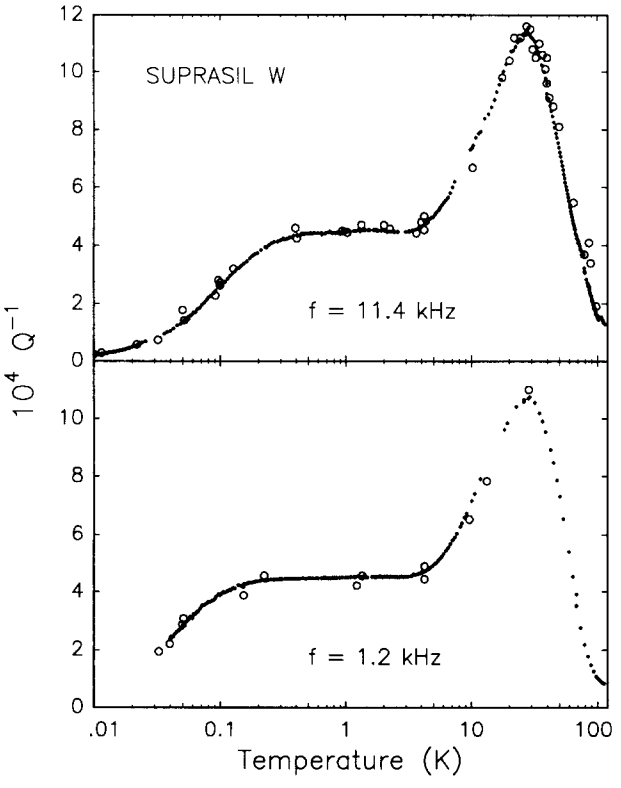

Fig. 1. Internal friction Q^{-1} of Suprasil W at frequencies of 11.4 and 1.2 kHz as a function of temperature. Open symbols denote Q^{-1} measurements by evaluation of complete resonance curves. Data from Ref. 18.

The internal friction below 4 K can be explained via relaxation processes as well. As one can conclude from the different frequency dependence, however, the dynamical behaviour of the relaxing defects must be quite different from that of the 50 K peak. It can be explained by the relaxation of the occupation numbers of two-level tunnelling systems with a broad spectrum of energies and relaxation rates [19] as suggested in the tunnelling model. The general situation of a two-level tunnelling system is sketched in Fig. 2. In the case of a symmetric double-well potential the energy splitting E between the eigenstates is given solely by the tunnelling energy, $E=\Delta_0$. It is worth noting that in this case the particle has the same probability of being "left" as of being "right", regardless of whether it is in the ground or in the upper state. There is therefore no net change in the total elastic dipole when symmetric two-level tunnelling systems change their occupation numbers.

The situation is quite different for tunnelling systems with finite asymmetry energy Δ, as shown in the right part of Fig. 2. An asymmetry of the potential leads to a partial localization of the tunnelling entity and tran-

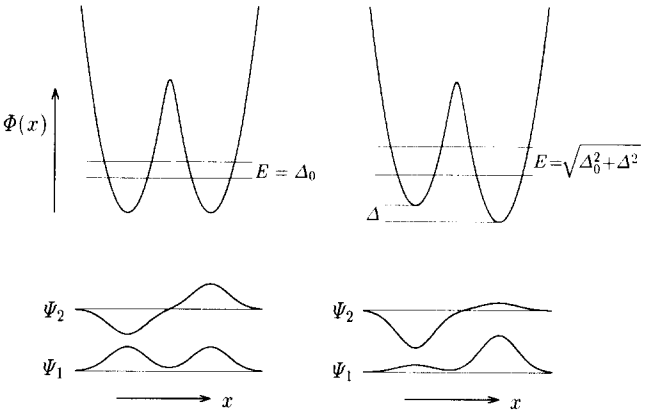

Fig. 2. Scheme of the potential $\Phi(x)$ and wavefunctions Ψ_1 (ground state) and Ψ_2 (excited state) of a particle in a double-well potential.

sitions between the two eigenstates are thus connected with changes in the probability of being in one or the other well. Applying an external stress field will therefore yield changes in the occupation numbers of an ensemble of asymmetric two-level tunnelling systems and thus give rise to relaxation processes similar to those of a dipole reorientation. The change in the occupation numbers occurs via inelastic scattering of thermal phonons and, in metals [20], conduction electrons. In insulating glasses at low temperatures the one-phonon process, i.e. absorption or emission of a single resonant phonon, is most important. The relaxation rate for this process is given by [19]

$$\tau_{\mathrm{ph}}^{-1}=\left(\frac{\Delta_0^2}{E^2}\right)\frac{E^3}{2\pi\rho\hbar^4}\left(\frac{\gamma_l^2}{\rho v_l^5}+2\frac{\gamma_t^2}{\rho v_t^5}\right)\coth\left(\frac{E}{2k_BT}\right) \quad (1)$$

where ρ is the density and v_l and v_t are the longitudinal and transverse sound velocities respectively. This rate is only weakly temperature dependent, in contrast with thermally activated rates, and is dominated by the density of states of transverse phonons. Its resonant character is expressed by the factor $(\Delta_0/E)^2$. Apparently, for tunnelling systems with a given energy splitting E, a broad spectrum of relaxation rates is possible according to their asymmetry energy. Systems with symmetric potentials ($\Delta_0/E=1$) have the highest rate $\tau_{\min}^{-1}$, whereas for systems with $E\approx\Delta$ rate (1) might be negligible compared with other processes.

Calculation of the acoustic susceptibility of a glass containing two-level tunnelling states is done in two steps [21]. First the contribution of an ensemble of systems, equal in energy E and e.g. λ or Δ_0, is evaluated. This is possible with a linear response theory using the Bloch equations which describe the time evolution of the polarization of an arbitrary two-level system. In the second step one has to integrate over the suggested distribution of the relevant parameters.

The susceptibility of N equal two-level systems contains a resonant and a relaxation contribution, either of which can be separated into real and imaginary parts, yielding relative changes in sound velocity $\delta v/v$ and internal friction Q^{-1}. The resonant interaction leads to

$$Q_{\mathrm{res}}^{-1}=\frac{2N\gamma^2}{\rho v^2}\left(\frac{\Delta_0^2}{E^2}\right)\tanh\left(\frac{E}{2k_BT}\right)F(E,\omega,\tau_1,\tau_2)G(I) \quad (2)$$

$$\left.\frac{\delta v}{v}\right|_{\mathrm{res}}=-\frac{N\gamma^2}{\rho v^2}\left(\frac{\Delta_0^2}{E^2}\right)\tanh\left(\frac{E}{2k_BT}\right)F'(E,\omega,\tau_1,\tau_2) \quad (3)$$

where the functions F and F' have the well-known shape of the response of harmonic oscillators. The linewidth is given by the phase coherence time τ_2, which is determined by elastic interactions with neighbouring tunnelling systems [11]. It is usually much shorter than the longitudinal lifetime τ_1, which is given here by the one-phonon process. Non-linear behaviour of the internal friction due to saturation effects at high acoustic intensities I is described by the function $G(I)$ in (2).

The contributions due to relaxation processes are given by

$$Q_{\mathrm{rel}}^{-1}=\frac{N\gamma^2}{\rho v^2}\left(\frac{\Delta^2}{E^2}\right)\frac{1}{k_BT}\operatorname{sech}^2\left(\frac{E}{2k_BT}\right)\frac{\omega\tau_1}{1+\omega^2\tau_1^2} \quad (4)$$

$$\left.\frac{\delta v}{v}\right|_{\mathrm{rel}}=\frac{N\gamma^2}{2\rho v^2}\left(\frac{\Delta^2}{E^2}\right)\frac{1}{k_BT}\operatorname{sech}^2\left(\frac{E}{2k_BT}\right)\frac{-1}{1+\omega^2\tau_1^2} \quad (5)$$

Since the relaxation rate τ_1^{-1} is only weakly temperature dependent, the temperature dependence of (4) and (5) is mainly given by the term $(1/k_BT)\operatorname{sech}^2(E/2k_BT)$. It expresses the fraction of two-level systems which have to adjust their occupation numbers at a given temperature when a perturbation $\delta\Delta$ is applied. Note also that according to the factor $(\Delta/E)^2$, symmetric tunnelling systems will not contribute to the relaxation process.

In order to obtain the acoustic properties of all two-level tunnelling systems, one has to replace N by an appropriate distribution function, e.g. $P(\lambda,\Delta)=\bar{P}=\text{const.}$, according to the tunnelling model. A full integration needs numerical methods but in limiting cases analytic expressions can be given. At very low temperatures, when $\omega\tau_{\min}\gg 1$ holds for all two-level systems, only resonant processes are important. In agreement with experiments [9, 10] at frequencies around 1 GHz an internal friction proportional to $\tanh(\hbar\omega/2k_BT)$ is predicted. In principle this behaviour should be observable at arbitrary frequencies owing to the constant density of states of the two-level systems. It is difficult, however, for frequencies below 100 MHz to reach low enough temperatures for a noticeable difference in the occupation numbers of the resonant systems.

The resonant contribution to the sound velocity yields a logarithmic temperature dependence

$$\left.\frac{\delta v}{v}\right|_{\mathrm{res}}=\frac{\bar{P}\gamma^2}{\rho v^2}\ln\left(\frac{T}{T_0}\right) \quad (6)$$

where T_0 is an arbitrary reference temperature. The $\ln T$ behaviour is often taken as a characteristic of glassy behaviour. It provides evidence of a constant density of states in energy splitting of two-level systems, as also the linear temperature dependence of the specific heat does.

With rising temperature relaxation processes become important. As long as $\omega\tau_{\min}\gg 1$, the reduction of the sound velocity will be negligible and the internal friction should rise as

$$Q_{\mathrm{rel}}^{-1}=A\frac{\bar{P}\gamma^2}{\rho v^2}T^3 \quad (7)$$

where A essentially contains the centre bracket of (1), i.e. the coupling strength of the tunnelling systems to phonons. At sufficiently high temperatures, when the regime $\omega\tau_{\min} \ll 1$ is reached, a temperature- and frequency-independent internal friction is predicted. This is in fact observed and demonstrates that over a very wide range of energies and, in particular, relaxation rates (experiments span a range between millihertz and 30 GHz) the assumptions of the tunnelling model are extremely good. The value of the internal friction in this regime is given by $Q_{\mathrm{rel}}{}^{-1} = \pi \bar{P}\gamma^2/\rho v^2$.

In the same limiting regime $\omega\tau_{\min} \gg 1$ the relaxation contribution to the sound velocity, i.e. integration of (5), yields

$$\left.\frac{\delta v}{v}\right|_{\mathrm{rel}} = -\frac{3}{2}\frac{\bar{P}\gamma^2}{\rho v^2}\ln\left(\frac{T}{T_0}\right) \tag{8}$$

This means, after addition of (6), that the sound velocity should decrease logarithmically in temperature with half the slope of the low temperature increase. The thus-predicted maximum in $\delta v/v$ occurs where $\omega\tau_{\min} \approx 1$. Roughly speaking, this behaviour is observed at frequencies in the kilohertz range. As shown in Fig. 3, however, recent experiments could not verify the predicted ratio of slopes (1:−0.5) below and above the maximum. This discrepancy cannot be solved easily within the presented theoretical model. It is quite possible that linear response theory is no longer valid above a certain temperature when the inverse lifetimes

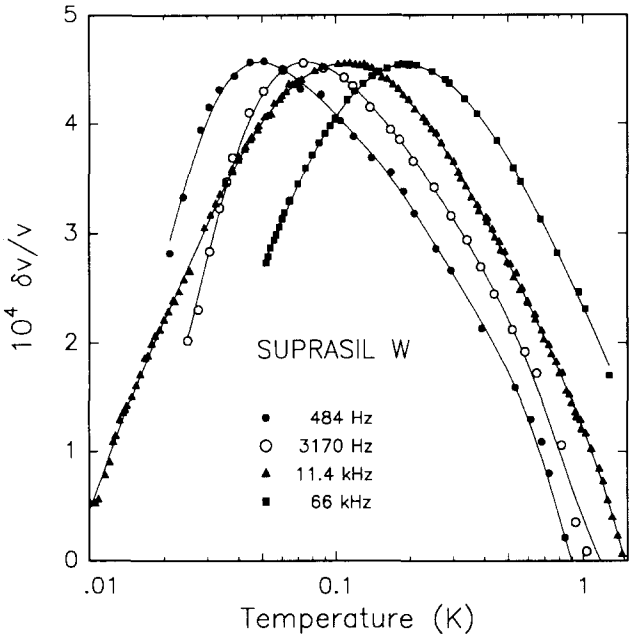

Fig. 3. Comparison of various sound velocity measurements on Suprasil W in the kilohertz range (after Ref. 18). Data at 66 kHz are from Ref. 22 and data at 484 and 3170 Hz from Ref. 23. Lines are only a guide for the eye.

of the two-level tunnelling systems become comparable with their energy splittings.

At temperatures above a few kelvins the sound velocity decreases more steeply and the internal friction exhibits a strong increase. In this temperature range thermally activated processes become more important and finally dominate the relaxation. Now two-level systems may contribute which have very small tunnelling energies Δ_0 compared with their asymmetry energy Δ. The overlap of the wavefunctions and thus the one-phonon rate (1) can be neglected and we are left with the classical picture of elastic dipole relaxation. The observed maximum in the internal friction and its frequency shift are explained via a distribution of activation energies. It has been shown [24] that the tunnelling model can be extended to cover this temperature range when certain plausible assumptions are made about the functional relation between the tunnelling parameter λ and the barrier height V. In this way it is possible to give a consistent phenomenological description of the internal friction of glasses between 10 mK and room temperature (see Fig. 1). Moreover, it is important to note that without changing any parameter, this description is still valid for frequencies as high as 35 GHz, i.e. for Brillouin scattering experiments [24].

Within the soft potential model a similar extension to higher temperatures and to thermally activated processes is possible [13]. It seems that, in particular, measurements on chalcogenide glasses, e.g. amorphous Se, are better described by this approach [2]. The difference comes from the distribution function in V, which is proportional to $V^{-\alpha}$ (with α between $\frac{1}{4}$ and $\frac{3}{4}$) at small values of V in the soft potential model and is a constant in the extended tunnelling model.

It is a long-standing question whether the phenomenologically introduced two-level tunnelling systems can be explained by a microscopic model. In contrast with their rather universal occurrence, the microscopic nature of the tunnelling systems is certainly non-universal. In amorphous noble gases it seems plausible that single atoms moving in very shallow potentials over small distances are good candidates [25]. In vitreous silica it is more likely that small clusters consisting of at least one SiO_4 tetrahedron can give rise to a rotational tunnelling motion.

In order to gain more insight to this kind of question, one can treat the sample material in various ways. Whereas e.g. annealing has only little influence on the low temperature acoustic properties of a-SiO_2, drastic changes are observed in Brillouin scattering under the application of high pressure [24, 26]. These experiments show that the distribution function of barrier heights becomes considerably broader with increasing pressure up to about 24 kbar. At even higher pressures the number of relaxing two-state defects decreases, as can

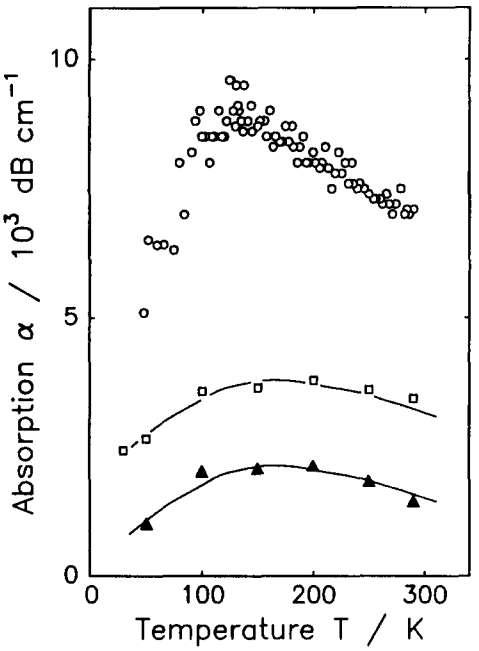

Fig. 4. Absorption coefficient as a function of temperature of 35 GHz phonons in normal (O) and compacted (▲) a-SiO_2. Open squares show data of the compacted glass as evaluated from the full width of the inhomogeneous Brillouin line (after Ref. 27).

be concluded from the decreasing values of the internal friction.

At room temperature and up to pressures of about 80 kbar a-SiO_2 behaves fully reversibly. At elevated temperatures, however, or even higher pressures a compacted modification of a-SiO_2 can be produced. This allows us to study some kind of "frozen-in" high pressure phase of glass. The result is remarkable. In Fig. 4 the ultrasonic absorption, measured by Brillouin scattering around 35 GHz, of normal a-SiO_2 is compared with that of compacted a-SiO_2. Apparently, in the compacted modification, which has a 10% higher density, the number of two-state defects has been diminished by a factor of 5. It would be interesting to study also the thermal and acoustic properties of the same material at temperatures below 4 K. We think that such experiments are crucial and encouraging for new theoretical developments towards a microscopic understanding of the two-level tunnelling systems in glasses.

4. Summary

An introduction has been given to the acoustic properties of glasses, in particular at low temperatures. It is shown that at present experiments are best described by the dynamics of two-level tunnelling systems intrinsically present in the random structure. An integral and important part of theoretical models is to suggest distribution functions of the relevant parameters which characterize these tunnelling systems. At present the theoretical challenge is to find a microscopic basis for the generation of distributions that adequately describe the observed phenomena. On the experimental side, measurements on crystalline materials seem to be encouraging. According to our present understanding, in crystals some kind of disorder, e.g. grain boundaries or dislocations, is necessary to introduce a so-called glassy behaviour. Tunnelling systems certainly do not occur in the regular structure of pure single crystals.

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