

ON THE ACOUSTIC ABSORPTION OF VITREOUS SILICA¹⁾

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Amorphous solids exhibit a variety of unusual low-temperature properties. These so-called "anomalies" can be described by a phenomenological model based on the assumption that small structural entities are capable of tunnelling between different sites in the random structure. In this paper an attempt is made to extend this model to higher temperatures. As an example the acoustic absorption of vitreous silica is discussed.

1. INTRODUCTION

The low-temperature properties of amorphous, or more generally, of disordered solids have attracted the attention of physicists for many years. More recently, unambiguous evidence has been produced that below 1 K the thermal properties of amorphous solids differ markedly from those of their crystalline counterparts [1, 2]. In spite of intense experimental and theoretical efforts these "anomalies" can at present be described only on the basis of a phenomenological model. In this model it is assumed that structural rearrangements in the amorphous network are possible, even at the lowest temperatures. More specifically, it is assumed that atoms or groups of atoms can tunnel between different but energetically nearly equivalent sites. Owing to the randomness of the amorphous structure, such "tunnelling systems" (TSS) will exhibit a broad distribution of the intrinsic parameters. Thus a rather universal and general interpretation of the low-temperature behaviour of thermal, acoustic and dielectric properties of amorphous solids is possible. However, so far the exact nature of the tunnelling entities is known neither in general nor in specific cases.

This paper is not intended as a review reflecting the present understanding of the low-temperature properties of amorphous solids but rather to discuss how this model can be extended to describe the behaviour of amorphous solids at higher temperatures. In the first part of the paper the tunnelling model (TM) is

introduced together with its basic assumptions [3, 4]. To illustrate the possibility to describe by this model the dynamic behaviour of amorphous solids, results of acoustic experiments will be discussed. In the second part an attempt is made to explain the properties of amorphous solids on the basis of the TM also at higher temperatures. As an example we consider the acoustic absorption of vitreous silica up to room temperature in a wide range of frequencies. We will see that satisfactory agreement with experiment is obtained if some additional assumptions are made.

II. TUNNELLING MODEL

In a perfect crystal each atom is constrained by symmetry to occupy a single potential minimum. In contrast, impurity atoms, as for example CN⁻ ions in KBr, very often move in multi-minima potentials provided by the neighbouring atoms. In the TM it is assumed that a similar situation also exists in amorphous solids: certain atoms or small groups of atoms can occupy two or even more potential minima of nearly equal energy. However, only a small fraction of the total number of atoms will have this degree of freedom, whereas the remainder is vibrating in single-well potentials and is therefore essentially immobile at low temperatures.

In the TM the simple case of a double-well potential is studied. The two wells are assumed to be parabolic and identical. Starting from the single-well problem each "particle" will have vibrational states separated by an energy $\hbar\Omega$ which is of the order of the Debye energy. The difference in the depths of the wells is generally referred to as asymmetry energy Δ . At low temperatures only the vibrational ground state will be populated. If tunneling occurs, the common ground-state will not only be split due to Δ but also a tunnel splitting Δ_0 will occur given by $\Delta_0 = \hbar\Omega \exp(-\lambda)$. The tunnelling parameter reflects the overlap of the wave functions and is given by

$$\lambda = (2mVd^2/\hbar^2)^{1/2}. \quad (1)$$

As we will see λ plays an important role in our further discussion. Here m is the effective mass of the tunnelling entity, V the barrier height between the two minima and d their separation in configurational space.

After diagonalizing the Hamiltonian one obtains the true ground state splitting E given by $E^2 = \Delta^2 + \Delta_0^2$. Appropriate distributions of the parameters of the TSS have to be introduced. In the TM Δ and λ are assumed to be independent of each other and uniformly distributed in the range experimentally accessible:

$$P(\Delta, \lambda) d\Delta d\lambda = \bar{P} d\Delta d\lambda, \quad (2)$$

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where P is a constant. This distribution function plays a crucial role in the description of the low temperature properties of amorphous solids.

It should be pointed out that here the "standard" TM has been presented. Besides that a more recent approach exists, which gives a better understanding of the microscopic nature of the TSS. This model is based on the assumption that in amorphous networks the local atomic potentials can exhibit extremely small values, leading to so-called "soft configurations" [5, 6, 7]. Unfortunately the mathematical treatment of this model is much more complex than that of the one described above. Therefore, in the following discussion only the "standard" TM is used for the sake of simplicity.

If the equilibrium occupation of the two levels of the TSS is disturbed by some means, they attain the new equilibrium within a certain relaxation time τ . Depending on the material and the temperature range, different relaxation mechanisms operate. In dielectric amorphous solids, to which we restrict our discussion here, relaxation occurs via interaction with thermal phonons. Below 1 K the simple one-phonon or direct process is dominating, i.e. a single thermal phonon is absorbed or emitted by the relaxing system. In this case the relaxation rate is given by [8]

$$\tau_d^{-1} = 4\Delta_0^2 \cdot E \cdot \coth(E/2kT), \quad (3)$$

where $A = (\gamma l^2/v_l^2 + 2\gamma t^2/v_t^2)/2\pi\phi h^4$. Here ϕ is the mass density, v the velocity of sound and γ the deformation potential describing the coupling of the TSS to the strain field. The indices l and t denote the longitudinal and transverse phonon branches. At higher temperatures, i.e. above a few K, multi-phonon processes come into play and relaxation times are drastically reduced. This aspect will be considered in the following chapter.

An important consequence of Eqs. (1), (2) and (3) is that the relaxation times of the TSS also exhibit a wide distribution. For a given energy splitting E and a fixed temperature there is a minimum relaxation time τ_m , for which the tunnel splitting $\Delta_0 = E$. For $\Delta_0 \rightarrow 0$, tunneling is hardly possible and τ approaches infinity. In other words, particles in symmetric double-well potentials relax fast, whereas in asymmetric wells relaxation times τ_m are of the order of 1 ns for TSS having an energy splitting corresponding to 1 K. On the other hand relaxation times as long as 10^4 s have been observed in measurements of thermal relaxation [9]. Although even longer times are expected from the TM they have not yet been observed because of experimental limitations. However, one may state that at a given temperature and energy relaxation times span at least 13 orders of magnitude!

For our further discussion the coupling of TSS to external strain fields is of particular interest. A sound wave travelling through an ensemble of TSS will be attenuated by two different mechanisms. Firstly it will be resonantly absorbed by those TSS having an energy splitting corresponding to the phonon energy.

Because of the wide distribution of energy splittings this process occurs at all frequencies and leads to well-known saturation phenomena [2]. However, in this paper the second mechanism is of main interest. It is the relaxation absorption [8], which is due to the modulation of the energy E by the sound wave. In this way the equilibrium of the TSS is disturbed and a relaxation process restores it again. Because of the finite value of the relaxation time τ the sound wave is attenuated. The main contribution to this attenuation process arises from those TSS which have an energy splitting $E \simeq kT$ and relax on a time scale comparable with the period of the sound wave, i.e. for which $\omega\tau \simeq 1$ holds. Because of the distribution of relaxation times an integration has to be carried out with respect to both E and Δ_0 or λ .

We express the acoustic absorption in terms of the internal friction Q^{-1} (i.e. absorption per wavelength), for which one finds [2]:

$$Q^{-1} = C \int_0^\infty dx \operatorname{sech}^2(x/2) \int_{\lambda_{\min}}^\infty d\lambda \frac{\Delta(\lambda)}{E} \frac{\omega\tau(\lambda)}{1 + \omega^2\tau^2(\lambda)}. \quad (4)$$

Here $x = E/kT$, $C = \bar{P}\gamma^2/\omega v^2$ and λ_{\min} is the lowest possible value of the tunneling parameter. In our calculations we may put $\lambda_{\min} = 0$.

Analytic solutions can be found for $\omega\tau_m \gg 1$ and $\omega\tau_m \ll 1$, which are briefly discussed here. At low temperatures, where $\omega\tau_m \gg 1$ holds, an absorption is expected which is frequency independent but increases strongly with temperature. Provided that the TSS relax via absorption or emission of a single thermal phonon (one-phonon process), Eq. (4) leads to

$$Q^{-1} = \frac{\pi^4 A C k^3 T^3}{12\omega}. \quad (5)$$

At "high" temperatures, where $\omega\tau_m \ll 1$, the absorption should become temperature independent, so a "plateau" should be observed:

$$Q^{-1} = \pi C/2. \quad (6)$$

It should be emphasized here that the occurrence of a plateau and also its height are independent of the fact whether the TSS relax via one process or multi-phonon processes. Therefore it allows direct conclusions of the validity of the distribution function $P(\Delta, \lambda)$. Furthermore in the regime $\omega\tau_m \ll 1$ only TS with $\Delta_0 \ll E$ are able to fulfil the condition $\omega\tau \simeq 1$. Thus information on the slowly relaxing TSS is obtained.

In Fig. 1 the internal friction of PMMA and vitreous silica is shown as a function of temperature [2, 10]. Besides the low temperature rise well developed plateaus are observed at higher temperatures. Owing to the large difference in the frequency of measurement the transition occurs at different temperatures. The plateaus show that in both materials the distribution function $P(\Delta, \lambda)$ is a

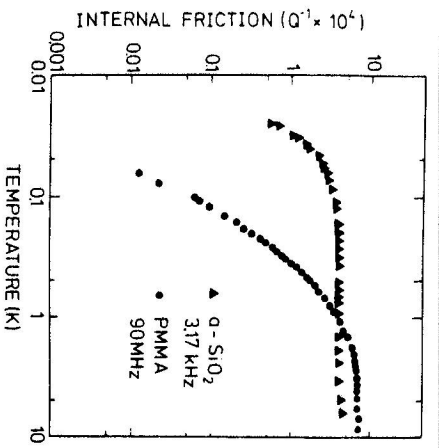


Fig. 1. Internal friction Q^{-1} of poly(methyl methacrylate) and vitreous silica as a function of temperature [from 2].

good approximation although the microscopic structures of these two amorphous solids are fundamentally different.

Closely related with absorption processes is a change in the velocity of sound. By applying the Kramers—Kronig relation the contribution of both the resonant and the relaxation absorption can be deduced [2]. A characteristic logarithmic temperature dependence of the velocity is predicted which has in fact been found [11]. This aspect will not be considered further in this paper.

III. ACOUSTIC ABSORPTION AT HIGHER TEMPERATURES

The acoustic properties of vitreous silica have been studied by many groups, both experimentally and theoretically [12—19]. As we have seen above, below 1 K the elastic behaviour of glasses is well explained by the TM. Therefore it is tempting to apply this model also at higher temperatures. In the following we will see that in fact the rather complex temperature and frequency dependence of the acoustic absorption of vitreous silica can be described satisfactorily in such a way.

Before reporting on the results of our numerical calculations and the assumptions made, let us enumerate those characteristic features which have, to be explained. In Fig. 2 the acoustic loss of vitreous silica at 45 MHz is plotted in the temperature range from 0.3 K to 200 K [20]. Two peaks can easily be distinguished. The maximum observed at about 4 K is definitely caused by the TSS. The second peak found at about 50 K has been attributed by most of the authors to thermally activated motion of oxygen atoms. Recently it has been proposed in a theoretical treatment of the acoustic properties of glasses [19] that the absorption in the whole temperature range originates from the interaction

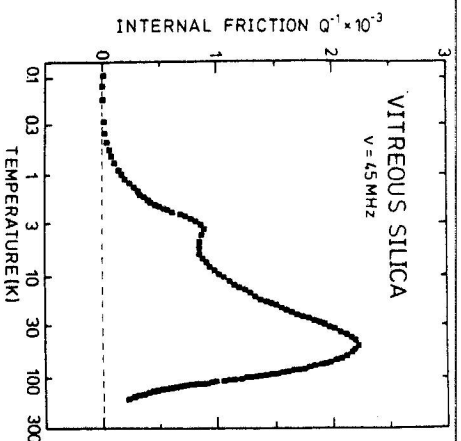


Fig. 2. Internal friction Q^{-1} of vitreous silica at 45 MHz. The two absorption peaks can easily be distinguished [from 20].

of the sound wave with TSS. Unfortunately no attempt has been made to compare this theory with experimental data in a quantitative way.

The measurements of the acoustic absorption of vitreous silica have been carried out by different authors using different techniques. For these reasons it is not surprising that a compilation of all results does not lead to a consistent set of data. Nevertheless, the following statements can be made unambiguously:

- At low temperature a T^3 -rise of the absorption is found, which is explained by the TM (see Eq. (5)).
- As shown in Fig. 1 the steep rise is followed by a plateau. A closer inspection of the data in this temperature range shows, however, that in all cases a weak temperature dependence occurs. At low frequencies, i.e. in the kHz-range, the loss decreases slightly with temperature (see Fig. 1). In the MHz-range (see Fig. 2) a tiny maximum ("4 K-peak") is found followed by a shallow minimum. At still higher frequencies, i.e. above 200 MHz, the "4 K-peak" degenerates into a shoulder.
- Above 10 K, absorption rises steeply again until the "50 K-peak" is reached. With increasing frequency the position of this maximum is shifted to higher temperatures.
- As can be seen in the logarithmic plot of Fig. 2 for 45 MHz, the rise of the absorption at the low temperature side of the peak is less steep than the decrease. This observation is made at all frequencies.
- Finally, the ratio Z between the height of the "50 K-peak" and the 4 K-peak" or the plateau varies only weakly with frequency. Z is found to be 2.3 at 4.8 kHz [14] and 2.6 at 45 MHz [20]. The prediction of the correct value of Z is a crucial test for the different theoretical explanations.

In order to describe the acoustic absorption by the TSS at higher temperatures we have to take into account relaxation via higher order processes. In a crude way this can be done by introducing Raman- and thermally activated processes:

$$\tau_r^{-1} = R \cdot \left(\frac{\Delta_0}{E} \right)^2 \cdot T^2 \quad (7)$$

$$\tau_a^{-1} = \nu_0 \cosh(\Delta/2kT) \cdot e^{-V/kT}, \quad (8)$$

where R and ν_0 are constants. It should be mentioned that in the expression for relaxation via the Raman-processes (Eq. (7)) [21] a factor containing $E/2kT$ has been omitted because it hardly has any influence on the numerical results.

According to Eq. (1) the tunnelling parameter λ is proportional to $1/\sqrt{V}$, where V is the height of the barrier between the two potential minima. This relation can only be correct if no correlation between m or d and V exists. To some extent such an assumption is plausible for the mass m , whereas it is unlikely in the case of the distance d : If the double-well potential is formed by two adjacent rigid parabolic wells, then d and V are correlated. As a result $V \propto d^2$ and consequently in this simple case $\lambda \propto V$. In recent experiments on optical hole burning a distribution $P(V) \propto 1/\sqrt{V}$ has in fact been deduced [22] indicating the missing of any correlation. But it should be mentioned that in this experiment large dye-molecules embedded in an amorphous matrix were forming the TSS which were investigated. In some sense these TSS must be considered as "extrinsic" [23]. Not knowing the real correlation of intrinsic TSS in a real glass, we leave this question open and put

$$\lambda^n = V \cdot L, \quad (9)$$

where L is taken to be a constant and $1 \leq n \leq 2$, depending on the extent of correlation.

At this point some remarks should be made on necessity to introduce also relaxation via the Raman processes besides the thermally activated process. Since the Raman process is "smoothing" the temperature variation of the relaxation rate in proceeding from the direct to the thermally activated process, it will have some influence on the acoustic properties in the intermediate temperature range. If only the principal behaviour of the acoustic absorption is considered, this process can be neglected.

In a first step we may calculate the absorption at temperatures well above 10 K by inserting the expression for the total relaxation rate into Eq. (4) and using the distribution function given by Eq. (2). Assuming that the different

relaxation mechanisms are independent of each other, we are allowed to simply add all the individual rates and get for the total rate τ^{-1} :

$$\tau^{-1} = \tau_a^{-1} + \tau_r^{-1} + \tau_g^{-1}. \quad (10)$$

Above 10 K the relaxation will be dominated by the thermally activated process. In this temperature range tunnelling is not important any more and the TSS can be considered as classical defect states. Nevertheless, for their parameters we use the distribution function as introduced by the TM (Eq. 2). The thermally activated process causes an increase of the absorption with temperature which is approximately proportional to $T^{1/n}$. Of course, such an approach cannot lead to a maximum since at all temperatures there exist TSS which fulfil the condition $\omega\tau \approx 1$. In order to obtain a peak, the distribution function $P(\Delta, \lambda)$ must exhibit an upper cut-off in Δ and/or in λ . A natural upper limit for Δ could be kT_g , where T_g is the glass transition temperature. However, in vitreous silica T_g is about 1500 K and such a high energy should have no influence on the acoustic loss below room temperature. Therefore it is more likely that the restriction of the tunnelling parameter plays the critical role and an upper cut-off value λ_0 has to be introduced. A Gaussian distribution $P(\lambda) \propto \exp(-\lambda^2/\lambda_0^2)$ could be appropriate. In fitting the "50 K-peak" a similar distribution, namely $P(V) \propto \exp(-V/V_0)$, has already been used by Gilroy and Phillips [18]. These authors propose that the fitting parameter V_0 is related to the glass transition temperature.

For numerical calculations the free parameters have to be specified. We simply put $C = 1$, because the absolute value of the absorption is not of interest here. From low temperature measurements the strength of the direct relaxation process, expressed by the constant A (see Eq. (3)) is well known: in all calculations $A = 10^7 \text{ s}^{-1}$ was chosen. For the remaining parameters the values $\nu_0 = 10^{12} \text{ s}^{-1}$, $R = 0$, and $\lambda_0 = 120$ were used.

Fig. 3 shows the temperature dependence of the internal friction Q^{-1} calculated for 5 kHz and 1 GHz. The most interesting and most critical parameter is the quantity n (see Eq. (9)) reflecting the correlation between d and V . To demonstrate its significance, the acoustic absorption at 5 kHz was calculated for $n = 1$ and $n = 2$. To obtain a rise of the absorption at about 10 K, the values 800 eV^{-1} and $11\,000 \text{ eV}^{-1}$ were taken for the parameter L , respectively. Without correlation, i.e. for $n = 2$, a deep minimum is found before the absorption starts to rise slowly to higher temperatures. For $n = 1$ a much better agreement with experiment is found, i.e. for strong correlation between the distance of the wells and the barrier height. Therefore this value was used in all other calculations, although it seems that a value of n slightly higher than unity would lead to even a better agreement. Of course, one can imagine that n varies from one class of amorphous solids to another. Let us consider briefly the absorption calculated

for 1 GHz at low temperatures. Clearly, a shoulder at 5 K is missing. This deficiency is caused by the omission of the Raman process. Although this process does not substantially alter the results at low and relatively high temperatures, it is of importance in the description of the acoustic behaviour at a few Kelvin.

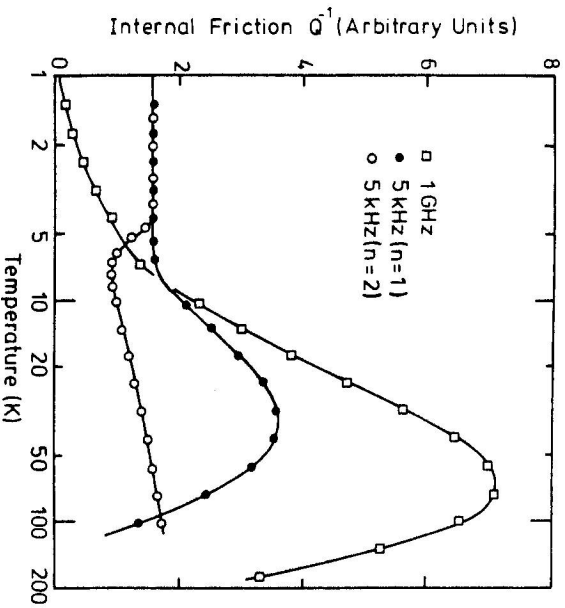


Fig. 3. Calculated internal friction Q^{-1} of vitreous silica for two different frequencies without taking into account relaxation via Raman processes and keeping the distribution function temperature independent. The low frequency absorption has been calculated with and without correlation between the parameters characterizing the double-well potentials. The parameter n is defined by Eq. (9).

Comparing the curves for $n = 1$ at 5 kHz and 1 GHz, we find that the ratio Z between the heights of the "50 K-peak" and the plateau (or the "4 K-peak") depends much more on frequency than experimentally observed. Such a result is found whenever the occurrence of the absorption maximum is caused by an upper cut-off for the tunnelling parameter. The reason is obvious: with decreasing frequency TSs with increasing values of λ will contribute to the absorption most. Since the number of such systems is reduced by the cut-off, the magnitude of the absorption will be reduced as well.

Obviously an additional mechanism must be present which suppresses the absorption at higher temperatures. Such a process was implicitly suggested by Fleurov and Trakhtenberg [19]. They pointed out that thermal motion

of the amorphous network can lead to considerable changes of the shape of the double-well potentials. Thus fluctuations of well separation and therefore also of barrier height become with increasing temperature more and more important. In an intuitive picture we may say that the relaxing particles are "waiting" in one of the wells until the barrier is lowered by thermal motion to such an extent that jumping over the barrier becomes possible. Finally at high enough temperatures the double-well potentials will disappear completely. In order to take into account this phenomenon in a global way, a temperature dependent distribution function $P(\lambda, T)$ was introduced here. The assumption made in the calculation was that the width as well as the height of the distribution function decreases with increasing temperature. For $P(\lambda, T)$ the following expression was used:

$$P(\lambda, T) \propto \exp(-T/T_0) \cdot \exp(-\lambda^2/\lambda_0^2(T)). \quad (11)$$

The decrease of the prefactor of the Gaussian distribution was assumed to be exponential and is determined by the parameter T_0 . For the temperature de-

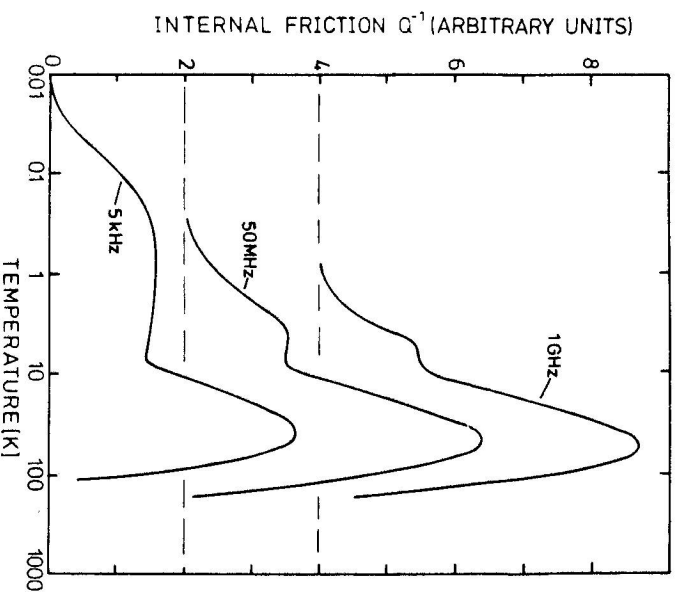


Fig. 4. Internal friction Q^{-1} of vitreous silica calculated for three different frequencies: 1 GHz, 50 MHz and 5 kHz. The origin of the scale has been shifted for each curve by two arbitrary units. The development of the "4 K-peak" can clearly be seen.

pendence of the width a linear variation $\lambda_0 = \lambda(T_0 - T)$ was chosen, where $\lambda = 1 \text{ K}^{-1}$ was used.

Some results of the numerical calculations are shown in Fig. 4. The internal friction is plotted for 5 kHz, 50 MHz and 1 GHz. The parameters of this numerical fit are $R = 1.5 \times 10^5 \text{ s}^{-1} \text{ K}^{-7}$ for the Raman process and $T_0 = 80 \text{ K}$ and $T_0 = 330 \text{ K}$ for the variation of the distribution function. All the other parameters were chosen as before. Now all the characteristic features enumerated above are met by this fit. Even such minor details as the transformation of the slightly declined "plateau" to a shoulder with increasing frequency is correctly reproduced. In particular the frequency dependence of the ratio Z mentioned above is in agreement with the experiment. Of course, the fit is not perfect. But keeping in mind the uncertainty of the experimental data [24] it is not worthwhile trying to get a better agreement with one of the measurements.

In conclusion it has been shown that the TM can account for the complex temperature and frequency dependence of the acoustic loss in vitreous silica up to room temperature if we allow for higher order relaxation processes and, in addition, assume that the distribution function becomes temperature dependent at higher temperatures.

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ОБ АКУСТИЧЕСКОМ ПОГЛОЩЕНИИ СТЕКЛООБРАЗНЫМ КРЕМНИЕМ

Аморфные тела демонстрируют множество необычных температурных явлений. Эти так называемые аномалии могут быть описаны в рамках феноменологической модели, основанной на предположении, что малые структурные единицы могут туннелировать между различными местами в случайной структуре. В предлагаемой работе была предпринята попытка обобщить эту модель для более высоких температур. В качестве примера рассмотрено акустическое поглощение стеклообразным кремнием.