

Interaction of soft modes and sound waves in glasses

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The interaction of sound waves with tunneling, relaxational, and resonant vibrational states in glasses is investigated within the soft-potential model. The same bilinear coupling constant is assumed for all three different kinds of soft modes. The model reproduces the results of the tunneling model at low temperatures and frequencies. In addition, it explains the fast rise of the relaxational absorption above 1 K and the plateau in the thermal conductivity around 5 K. The universal features of the sound absorption in glasses are described with good accuracy up to 20 K.

I. INTRODUCTION

The purpose of this paper is to discuss the interaction of acoustic waves with localized soft modes in glasses and to investigate how this interaction can influence the transport properties at low temperatures.

Acoustic and dielectric properties of glasses were intensively investigated during the last decades. The most remarkable result¹ of these investigations was the discovery of a set of universal low-temperature properties that are only weakly dependent on the specific chemical composition of the glass. These properties are normally not found in crystals and are often called "anomalous": they include low-temperature specific heat, thermal conductivity, propagation of ultrasound, dielectric loss, electric and acoustic echo, and some other properties governed by low-frequency modes.

An important progress towards an understanding of the anomalous properties of glasses was the introduction of the tunneling model by Phillips² and Anderson, Halperin, and Varma.³ According to this model there exist two-level systems associated with local tunneling states in double-well potentials characterized by an energy difference Δ between the minima and a tunnel splitting $\Delta_0 = \hbar\omega_c \exp(-\lambda)$. Here ω_c is some crystal-like vibrational frequency and λ is a tunnel integral. The parameters Δ and λ are random; their distribution function $\bar{P}(\Delta, \lambda)$ is assumed to be constant. The tunneling model provides an explanation for the main features of thermodynamics and kinetics in glasses at very low temperatures.

There are several review articles where experimental data and their interpretation are given, in particular by Hunklinger and Arnold,⁴ Hunklinger and Raychaudhuri,⁵ Phillips,⁶ and those in the book edited by Phillips.⁷ A review of the properties of metallic glasses is given by Black.⁸ The universal features of the thermal conductivity over a large temperature range have been emphasized

by Freeman and Anderson.⁹

Above 1 K, the properties of glasses deviate from the predictions of the tunneling model. The thermal conductivity shows a plateau around 5 K, which cannot be understood in terms of a constant density of tunneling states^{9,10} and is difficult to understand in terms of phonon scattering from static disorder.¹¹⁻¹⁴ Furthermore, there is an additional increase in the specific heat, indicating the existence of still another kind of low-frequency mode. Recent neutron measurements¹⁵ have shown these to be soft harmonic vibrations with a crossover to anharmonicity at the low-frequency end (at frequencies corresponding to several kelvin). Measurements of the specific heat and of the thermal conductivity of a glassy polymer have shown that the contribution by these modes decreases with pressure parallel to the two-level contribution.¹⁶

There are many conflicting explanations for the universal anomalous behavior between 1 and 10 K. One possibility is to associate the plateau in the thermal conductivity to phonon scattering from some kind of disorder (scattering by frozen-in free volume,¹⁷ clusters,¹⁸ fractals,¹⁹ and disorder in the force constants,²⁰ to mention only a few of the large number of similar proposals) and to explain the additional vibrational states as a consequence of phonon localization. These approaches have two weak points. First, it is difficult to explain why phonons with a wavelength scale on which the glass is still practically homogeneous should be scattered so strongly.¹¹⁻¹³ Second, the explanation for the tunneling states is lacking.

Another possibility is to try to extend the tunneling model to explain the anomalies at higher temperatures. One of these attempts starts from fundamental considerations on the interaction of the tunneling entities²¹⁻²⁵ trying to resolve the resulting many-body problem. At present, this kind of approach has not gone far beyond considerations of the tunneling states themselves. Alternatively, one can disregard the interaction of these enti-

ties and assume a phenomenological single-mode picture. First steps in this direction were undertaken by Karpov and Parshin²⁶ and by Yu and Freeman.²⁷ Both approaches, though with different assumptions, describe the plateau in the thermal conductivity in terms of resonant scattering of sound waves from localized low-frequency vibrations, thus relating the plateau to the strong rise of the specific heat at the same temperature. The special case of the mixed cyanides has been analyzed by Grannan, Randeria, and Sethna²⁸ along similar lines on a more microscopic basis.

Very recently, Tielburger *et al.*²⁹ considered an extension of the tunneling model toward higher barriers, in order to explain the strong rise of the sound wave absorption above 4 K in vitreous silica in terms of the onset of classical relaxation processes.

A more general approach, proposed by Karpov, Klinger, and Ignat'ev³⁰ describes the tunneling, the soft vibrations, and the low-temperature relaxational motion in terms of soft anharmonic potentials with locally varying parameters (see also Ref. 31, where an important extension of the model has been introduced). This approach reproduces for low temperatures the results of the tunneling model and predicts a change of behavior for higher temperatures. The consequences of the soft-potential model were investigated in several papers.^{26,30-33} A review of the soft-potential model and its implications is given in Ref. 33. Recently,³⁴ specific-heat and neutron data from several glassy materials were analyzed within the framework of the model and its parameters were determined.

The results obtained so far encourage the hope for a unified description of the low-temperature glassy anomalies. This paper intends to undertake the next step in that direction by assuming a bilinear coupling between sound waves and soft modes with the same coupling parameter for tunneling, vibrational, and relaxational states.

II. THE SOFT-POTENTIAL MODEL

A. Definitions

To begin with, we recall the main concepts of the soft-potential model.³⁰ It assumes the existence of soft localized modes in glasses. Since the average square displacement in such a mode is relatively large, one should take anharmonicity into account. Strong anharmonicity of soft potentials leads to a number of nontrivial features for the corresponding localized excitations.

The anharmonic soft potential of a single mode can be written as

$$V(x) = \mathcal{E}[\eta(x/a)^2 + \xi(x/a)^3 + (x/a)^4]. \quad (1)$$

\mathcal{E} is an energy of atomic scale, x is the displacement of the atom with the largest amplitude in the mode, while a is a distance of the order of the interatomic spacing. At the distance a the fourth-order term in the potential (1) is as high as the harmonic term for an unsoftened mode ($\eta = 1$) at a crystal-like frequency ω_c with $\mathcal{E} = M\omega_c^2 a^2/2$.

The definition of the coordinate x implicitly defines an effective mass M via the kinetic energy and the eigenvector of the mode³⁴ [see also Sec. III A, where this definition is given explicitly in Eq. (19)]. Comparison with experiment³⁴ and numerical work on a model glass^{35,36} have shown independently that this effective mass M is at least a factor of 10 higher than the atomic mass m . The coefficients η and ξ are random parameters. Depending on the values of η and ξ , Eq. (1) describes single- or double-well potentials. In the double-well case the same potential can be described by three sets of parameters, corresponding to expansions around the three extrema. In that case we restrict the possible values of η and ξ to the expansion around the maximum. $\eta > 0$ then always gives one-well potentials and $\eta < 0$ double-well potentials. In the latter case the two minima are separated by a distance $d = 2a\sqrt{9\xi^2/64 - \eta/2} \approx a\sqrt{2|\eta|}$ and a barrier of height $\mathcal{E}\eta^2/4$ (in the symmetric case $\xi = 0$).

To derive an energy scale for Eq. (1) let us consider the Schrodinger equation for the purely quartic potential ($\eta = \xi = 0$):

$$-\frac{\hbar^2}{2M} \frac{d^2}{dx^2} \psi_n + \mathcal{E}(x/a)^4 \psi_n = E_n \psi_n. \quad (2)$$

The spacing between the ground ($n = 0$) and the first excited state ($n = 1$) is of the order³⁰

$$W = \mathcal{E}\eta_L^2, \quad (3)$$

where the notation

$$\eta_L = (\hbar^2/2Ma^2\mathcal{E})^{1/3} \quad (4)$$

has been introduced. The energy W is related to the crystal-like frequency ω_c by $\hbar\omega_c = 2W\eta_L^{-1/2}$. The characteristic scale of the displacement is $a(\eta_L)^{1/2}$.

Both the effective mass M and the energy \mathcal{E} will increase proportional to the number of atoms participating in the soft mode, while the frequency ω_c is independent of that number. The energy W scales with $M^{-1/3}$, giving a relatively low value if many atoms participate in the mode.

The energy levels are dependent on the ratios η/η_L and $\xi/(\eta_L)^{1/2}$ and can be expressed in the form

$$E_n = WF_n(\eta/\eta_L, \xi/\eta_L^{1/2}). \quad (5)$$

For $|\xi/\eta_L^{1/2}| \approx |\eta/\eta_L| \approx 1$ and $n \approx 1$, $F_n \approx 1$.

The soft modes are assumed to be randomly distributed in the glass. The parameters η and ξ characterizing the single soft mode are random. Generally, the distribution has to obey $P(\eta, \xi) = P(\eta, -\xi)$. For small values of η and ξ the distribution is dominated by the seagull singularity^{31,34}

$$P(\eta, \xi) = P_0 |\eta|, \quad (6)$$

where P_0 is a slowly varying function of η and ξ , which in the following will be taken as a constant.

The vanishing of $P(\eta, \xi)$ as $\eta \rightarrow 0$ was justified³¹ on the basis of the destruction of very soft potentials as a result of small linear perturbations. Experimentally, it is

confirmed by the rapid rise of the specific heat at several degrees kelvin, which cannot be described quantitatively without this so-called “seagull singularity” assumption.³⁴

So far, our definitions stick to the usual convention of the soft-potential model.³³ In the following, we add a new definition, namely, the definition of coupling constant(s) for the bilinear interaction between soft modes and sound waves. As shown in Sec. III, the leading term of that interaction can be written as

$$\delta V(x, \tau_j) = \Lambda_j \tau_j \frac{x}{a}, \quad (7)$$

where τ_j is the strain deformation by a sound wave. One has to define two such coupling constants, Λ_l and Λ_t , for the strain deformations by longitudinal and transverse sound waves, respectively.

As noted above, in a symmetric double-well potential with negative η one has the two-well minima at $\pm a\sqrt{|\eta|}/2$. If the potential barrier in between is high enough, the wave functions of the lowest levels (or, in a classical treatment, the probability distribution function) will be centered around these minima. In such a case, one can describe the interaction between the soft mode and the strain of a sound wave in terms of a deformation potential between the two minima. With the bilinear coupling (7), the deformation potentials γ_l, γ_t are given by

$$\gamma_l = \Lambda_l (|\eta|/2)^{1/2}, \quad \gamma_t = \Lambda_t (|\eta|/2)^{1/2}. \quad (8)$$

Instead of working with the two coupling constants Λ_l and Λ_t or the deformation potentials γ_l and γ_t of the double-well case, it is often convenient to use the two dimensionless constants C_l and C_t ,

$$C_l = \frac{2P_0\eta_L^{7/2}}{W} \frac{\Lambda_l^2}{\rho v_l^2}, \quad C_t = \frac{2P_0\eta_L^{7/2}}{W} \frac{\Lambda_t^2}{\rho v_t^2}, \quad (9)$$

where ρ is the mass density of the glass and v_l, v_t are the longitudinal and transverse sound velocities, respectively. As will be seen, the two constants C_l and C_t can be consistently defined in both the tunneling⁵ and the soft-potential model. With these definitions, the predictions of the soft-potential model for the sound absorption at temperatures between 1 and 20 K can be expressed by the two parameters C_l and C_t , known for many glasses from experiments below 1 K, plus one additional parameter, the energy W .

B. Assumptions

The soft-potential model makes two assumptions: (i) The soft modes can on average be characterized by the parameters \mathcal{E} , M , and a as defined in the preceding subsection. (ii) The distribution function for the small random parameters η and ξ is proportional to $|\eta|$ around the origin in the (η, ξ) plane³¹ corresponding to Eq. (6) in the preceding subsection. Equation (6) is expected to hold up to values $|\eta|/\eta_L \gg 1$ and $|\xi|/\eta_L^{1/2} \gg 1$.

In this paper we add a third assumption: (iii) The interaction between the sound waves and the three dif-

ferent kinds of soft modes can be described in terms of the same bilinear coupling constants C_l and C_t defined above in Eq. (9).

C. Approximations

1. The tunneling model as a limiting case

The level splitting in the soft potentials is ordinarily of the order of the energy W or greater (see Sec. II A). The only exception from this rule is for double-well potentials, $\eta < 0$, where one can have tunneling. In fact, the soft-potential model reproduces for low temperatures the results of the tunneling model with small logarithmic corrections. The parameters Δ, λ of the tunneling model can be expressed in terms of the quantities η, ξ as³⁰

$$\Delta = \frac{W}{2^{1/2}} \frac{|\xi| |\eta|^{3/2}}{\eta_L^{1/2} \eta_L^{3/2}}, \quad (10)$$

$$\lambda \equiv \ln \frac{\hbar\omega_c}{\Delta_0} = \ln \frac{\hbar\omega_c}{W} + \frac{2^{1/2}}{3} \left| \frac{\eta}{\eta_L} \right|^{3/2}. \quad (11)$$

Calculating the transformation Jacobian and taking into account that the density of states \bar{P} of the tunneling model is only defined for positive asymmetries Δ (this is not stated explicitly in the treatments of the tunneling model,^{5,6} but can be seen from their following equations),

$$P(\Delta, \lambda) = \frac{4P_0\eta_L^{7/2}}{W|\eta|}. \quad (12)$$

In this approximation, $P(\Delta, \lambda)$ is not a constant as assumed in the tunneling model, but varies with $1/|\eta|$ (inversely proportional to the square root of the barrier height). Since the tunnel splitting changes exponentially with barrier height, the predictions of the two models do not differ significantly in the temperature and frequency region of the tunneling states. In the dimensionless parameter combination^{5,6} of the tunneling model

$$C_l = \frac{\bar{P}\gamma_l^2}{\rho v_l^2}, \quad C_t = \frac{\bar{P}\gamma_t^2}{\rho v_t^2}, \quad (13)$$

one even has a cancellation of the two η dependencies, if one inserts for \bar{P} the expression (12) and for γ_l (or γ_t) the definition (8) of the soft-potential model. C_l and C_t as defined in (13) then turn out to be equivalent to their definition (9) in the soft potential model in Sec. II A.

2. Soft vibrations: Another limiting case

If $1 \gg \eta \gg \eta_L$ the anharmonicity of the motion is relatively weak. In this situation Eq. (1) describes soft quasi-harmonic excitations with frequencies $\omega_c \gg \omega \gg W/\hbar$. The integration over ξ leads to a density of quasi-harmonic states^{31,34}

$$n_H(\hbar\omega) = \frac{1}{3\sqrt{2}} \frac{P_0\eta_L^{5/2}}{W} \left(\frac{\hbar\omega}{W} \right)^4. \quad (14)$$

This strongly rising density of soft localized vibrational

states is an important common feature of glasses. It explains the strong rise of the specific heat at a few degrees kelvin and the high density of soft vibrational states observed in neutron scattering.³⁴

3. Classical relaxation as a third limiting case

At higher temperatures, a particle (or an ensemble of particles in a configurational coordinate) will no longer tunnel through a potential barrier but will begin to hop over it as a consequence of classical activated relaxation. Following Gilroy and Phillips,³⁷ we describe the activated process in terms of a relaxation time τ given by

$$\tau^{-1} = \tau_0^{-1} \exp\left(-\frac{V_B}{k_B T}\right) \cosh\left(\frac{E}{2k_B T}\right). \quad (15)$$

Here τ_0^{-1} is an attempt frequency of the order of the crystallike frequency ω_c . V_B is the barrier height that can be expressed through the parameter η as

$$V_B = \frac{W}{4} \left(\frac{\eta}{\eta_L}\right)^2. \quad (16)$$

4. Crossover region

All the preceding approximations break down as one approaches the origin of the (η, ξ) plane, the pure quartic potential. The energy splittings of these potentials are of the order of W . At present, there is no simple analytic description for these configurations. Therefore, if one wants to avoid lengthy numerical calculations, one is forced to use the asymptotic expressions for the three regions of tunneling, vibration, and classical relaxation discussed above and to extrapolate them into the crossover

region.

In this paper, the energy $2W$ is chosen as the limit between the different extrapolations. At frequencies below $2W/h$, the sound waves are assumed to interact only with tunneling and relaxational modes; above $2W/h$ they are assumed to interact only with the soft vibrations. The limit is chosen in such a way that the mean free path at low temperatures joins continuously at the crossover frequency (see Fig. 1).

In the low-frequency domain, we further assume a crossover temperature W/k_B . Below that temperature, the low-frequency sound absorption is ascribed to the interaction with the tunneling states. Above W/k_B , the sound absorption is assumed to be due to classical relaxation. Again, the crossover temperature is chosen in such a way that the mean free paths join continuously (see Fig. 2).

III. COUPLING BETWEEN SOFT MODES AND SOUND WAVES

A. Soft vibrations

A soft-mode as given by the model potential (1), is not an exact eigenstate of the glass. It will interact with the extended modes of similar energy, the long-wavelength phonons. This interaction affects both the soft mode itself and the phonons, which are, for the same reasons, also not exact eigenstates. The sharp energy levels given by (1) will be broadened and shifted. The bulk of the energy shift is of course incorporated in (1), which represents an effective potential for a soft mode in the lattice. The width on the other hand is a measure of the scattering between the phonons and the soft modes and is, as such, not included in (1). Since we are considering soft modes of comparatively low frequencies (≈ 100 GHz) there is a low density of phonons of the same frequencies and hence a low damping of the soft mode—the soft mode is still well defined. Let us consider first the case of quasiharmonic soft vibrations in one well ($\eta > 0$). This problem can be treated analogously to the one of scattering of phonons by resonant vibrations in lattices which has been studied extensively.³⁸⁻⁴⁰

The soft vibrations are caused by a local deviation of the force constant matrix from the average one of the glass and, possibly, also a local change of mass. These we denote together as $\delta\ell(\omega)$. The soft-mode-phonon-scattering problem can then be formulated in terms of a defect t matrix,

$$t(\omega) = \delta\ell(\omega) + \delta\ell(\omega) \frac{|s\rangle\langle s|}{m(\omega^2 - \omega_s^2) - 2i\omega\gamma_s(\omega)} \delta\ell(\omega), \quad (17)$$

where $|s\rangle$ denotes the normalized state vector of the soft mode, and ω_s and γ_s are its frequency and width, respectively. For $\omega \approx \omega_s$, the second term, the resonant contribution, dominates. The Green's function G for the soft mode for $\omega \approx \omega_s$ follows from (17) by the equation $G^s = G_0 + G_0 t G_0$ with G_0 the ideal (average) lattice Green's function,

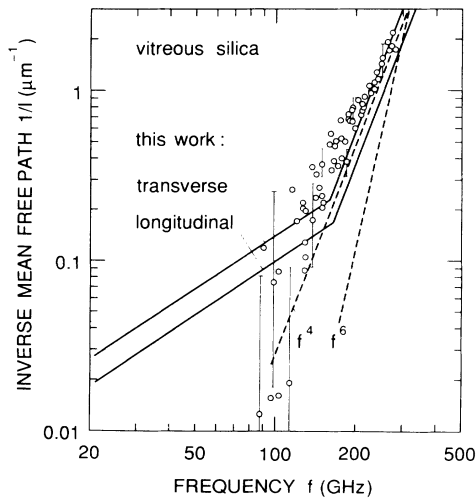


FIG. 1. Measured [circles: data by Dietsche and Kinder (Ref. 44); dashed lines denote f^4 and f^6 : data by Long, Hanna, and MacLeod (Ref. 45)] and calculated (continuous lines) inverse mean free path at low temperatures for sound waves in vitreous silica as a function of frequency.

$$G^s(\omega) \approx \frac{|s \gg \omega|}{m(\omega^2 - \omega_s^2) - 2i\omega\gamma_s(\omega)}. \quad (18)$$

Since (1) was written in terms of the amplitudes x of one atom, we have to take the projection of (17) onto this atom, say atom one, and obtain thus an effective mode mass

$$M = m \left/ \sum_{\alpha} \langle s_{\alpha}^1 | s_{\alpha}^1 \rangle \right., \quad (19)$$

where s_{α}^1 denotes the α component of the eigenvector on atom one with mass m . The definition of an effective mass by Eq. (19) is valid strictly only for a finite system but can, for low-lying resonant modes, be extrapolated to infinite systems.⁴⁰ This definition of the effective mass was alluded to in Sec. II A.

The broadening, $\gamma(\omega_{0,j})$, of a phonon $|\mathbf{k}, j\rangle$ with wave vector \mathbf{k} , polarization j , and frequency $\omega_0(\mathbf{k}, j) = v_j k \approx \omega_s$ by a concentration $g(s)$ of soft modes can be expressed in terms of the t matrix (17) as

$$\begin{aligned} \gamma(\omega_0, j) &= \frac{1}{2} g(s) \text{Im} \langle \mathbf{k}, j | t(\omega) | \mathbf{k}, j \rangle / m\omega_0(\mathbf{k}, j) \\ &\approx \frac{1}{2} g(s) \frac{|\langle \mathbf{k}, j | \delta\ell(\omega) | s \rangle|^2}{m\omega_0(\mathbf{k}, j)} \text{Im} \frac{1}{m[\omega_0^2(\mathbf{k}, j) - \omega_s^2] - 2i\omega_0(\mathbf{k}, j)\gamma_s(\omega_s)}. \end{aligned} \quad (20)$$

To obtain the total broadening this expression has to be averaged over all soft-mode orientations and integrated over all soft modes with frequencies approximately equal to ω_0 . This integration simplifies (20) considerably. First it is a basic assumption that the structure of the soft mode does not strongly depend on its frequency. The matrix element in (20) will therefore depend on k and the phonon polarization, but not on ω_s . Furthermore, the distribution of soft modes is smooth on a scale of their width, and we can therefore replace the imaginary part by its δ -function limit $\frac{\pi}{2m\omega_s} \delta(\omega_0 - \omega_s)$ and so finally get

$$\gamma(\omega_0, j) = \frac{\pi}{4} \frac{\text{Av} |\langle \mathbf{k}, j | \delta\ell(\omega_0) | s \rangle|^2}{m^2\omega_0^2(\mathbf{k}, j)} n_H(\omega_0(\mathbf{k}, j)). \quad (21)$$

Here we denote with Av the orientational averaging. $n_H(\omega)$ is the density of soft vibrations of Eq. (14).

To get an estimate for $\gamma(\omega_0, j)$ we expand (21) for small \mathbf{k} . The coupling change $\delta\ell(\omega)$ consists of two parts. The first one, $\Delta m\omega^2$, is due to a possible change in mass. The second one is due to a change in force constants $\delta\varphi$. The latter would be proportional to $m\omega_s^2$ if we were considering an atom weakly coupled to the rest of the glass. Here the soft mode is due to a cancellation of the average force constants of the glass. Hence, $\delta\varphi$ will be of order $m\omega_c^2$ in order to cancel the average force constants of the glass in the soft mode. The phonon wave function $|\mathbf{k}, j\rangle$ can be expanded in powers of \mathbf{k} ,

$$\begin{aligned} |\mathbf{k}, j\rangle_{\alpha}^n &= e_{\alpha}(\mathbf{k}, j) e^{i\mathbf{k} \cdot \mathbf{R}^n} \\ &= e_{\alpha}(\mathbf{k}, j) \left\{ 1 + i\mathbf{k} \cdot \mathbf{R}^n - \frac{1}{2}(\mathbf{k} \cdot \mathbf{R}^n)^2 + \dots \right\}. \end{aligned} \quad (22)$$

The constant term in the expansion contributes for the mass term, resulting in $\gamma \sim k^6$, but not for the force

constant term due to the translational invariance condition on $\delta\varphi$.^{39,40} From the linear term we obtain together with $\delta\varphi$ a contribution to the inverse mean free path $l^{-1} = 2\gamma/mv_j \sim k^4$:

$$l_{\text{vib, res}}^{-1} = \frac{\pi}{2\rho v_j^3} \frac{\Lambda_j^2}{M a^2} n_H(\omega_0(\mathbf{k}, j)) + O(k^6), \quad (23)$$

where we have introduced the abbreviation

$$\begin{aligned} \Lambda_j^2 &= \text{Av} [\Lambda^2(\mathbf{k}, j)] \\ &\equiv \frac{M a^2}{m} \text{Av} \left[e_{\alpha}(\mathbf{k}, j) \frac{k_{\beta}}{k} R_{\alpha}^m \delta\varphi_{\alpha\beta}^{mn} | s \rangle_{\beta}^n \right]^2 \end{aligned} \quad (24)$$

and used the long-wavelength limit for the phonon frequency $\omega_0(\mathbf{k}, j) = v_j k$ with v_j the transversal or longitudinal sound velocity, respectively. The term $\gamma \sim k^4$ vanishes in crystals for defects with odd symmetry. It therefore might also be small in a glass if the symmetry of the soft mode is approximately odd. This argument also shows that it is impossible to give an absolute magnitude without specifying the nature of the soft mode.

Equation (23) can be derived alternatively by introducing an effective interaction term in the soft-mode potential (1)

$$\delta V(x) = u_{k,j} \langle \mathbf{k}, j | \delta\varphi | s \rangle a \sqrt{\frac{M}{m}} \left(\frac{x}{a} \right), \quad (25)$$

where $u_{k,j}$ is the phonon amplitude and the factor $\sqrt{M/m}$ is present, since x is an atomic amplitude rather than the total mode amplitude. Equation (23) is obtained by calculating the absorption and emission probabilities for a phonon by the golden rule. In the long-wavelength limit it is often convenient to describe the phonon by its strain tensor

$$\epsilon_{\alpha\beta}(\mathbf{k}, j) = \frac{u_{\mathbf{k},j}}{2} [k_{\alpha} e_{\beta}(\mathbf{k}, j) + k_{\beta} e_{\alpha}(\mathbf{k}, j)]. \quad (26)$$

The interaction term thus becomes, to lowest order,

$$\delta V(x) = \epsilon_{\alpha\beta}(\mathbf{k}, j) p_{\alpha\beta} \frac{x}{a}, \quad (27)$$

where $p_{\alpha\beta} \times x/a$ is the change of the soft-mode dipole tensor with displacement x :

$$p_{\alpha\beta} = \sum_{m,n,\gamma} R_{\alpha}^m \delta\phi_{\beta\gamma}^{mn} |s >_{\gamma}^n \sqrt{\frac{M}{m}} a. \quad (28)$$

Averaging over orientations in the strain field $\tau_j = u_{k,j} k$ of a long-wavelength sound wave gives together with (24) the definition (7) introduced in Sec. II.

B. Tunneling and relaxational states

In the case of two-well potentials the interaction with the phonons can modulate the energy difference, Δ , between the two minima. Depending on the conditions this can cause resonant absorption of two-level states—if $\Delta \approx 0$ and the phonon frequency equals the tunnel splitting—or relaxation processes if the transition probability from one well to the other (tunneling or classical) is small within the period of the vibration within a well. As in the case of one-well potentials symmetry can, in principle, preclude the lowest-order interaction term. In an actual glass a total suppression is, however, unlikely, and we will restrict the discussion to this lowest order.

As noted in Sec. II, in a symmetric double-well potential with negative η one has the two well minima at $\pm a\sqrt{|\eta|/2}$. With the bilinear coupling (7), the deformation potentials γ_l, γ_t are given by (8) of Sec. II A.

It should be noted that we have estimated the deformation potential by extrapolation from the harmonic limit of the one-well potential. More accurately the deformation potential should be determined from the difference of the elastic dipole tensors for the soft mode in its left and right well, respectively. This could introduce substantial corrections due to anharmonicity. In numeric simulations of self interstitials in fcc metals,⁴¹ which show resonant modes and two-well configurations, albeit with larger dis-

tances than the ones relevant for our investigations, it was found that, even though no lowest-order resonance absorption occurs, not all models give a nonvanishing deformation potential. Equation (8), therefore, represents an estimate that will become less accurate for the larger two-well systems (larger distances between the minima and higher barriers).

C. Resonant absorption

In order to calculate the resonant absorption of sound waves from the interaction with soft modes, it is again useful to consider the asymptotic cases of tunneling on one side and vibration on the other. The tunneling case has been treated in the context of the tunneling model.^{5,6} Here we follow the treatment given by Phillips,⁶ which yields for the mean free path of a phonon

$$l_{\text{res,tun}}^{-1} = \frac{\pi\omega C_j}{v_j} \tanh \left[\frac{\hbar\omega}{2k_B T} \right], \quad (29)$$

where j stands for l or t in the longitudinal or transverse case, respectively, and C_l, C_t are given by the definition Eq. (13). Since the same constants can be defined in the soft-potential model, Eq. (9), the same relation holds for the soft-potential model in the range where tunneling states are relevant.

The mean free path resulting from the resonant interaction between vibrational soft modes and sound waves can be calculated from Eq. (23) and using Eq. (14) for the density of vibrational states. One gets

$$l_{\text{res,vib}}^{-1} = \frac{1}{6\sqrt{2}} \frac{\pi\omega C_j}{v_j} \left(\frac{\hbar\omega}{W} \right)^3. \quad (30)$$

D. Relaxational absorption

The mean free path of a sound wave under the influence of relaxational processes can be written^{6,37}

$$l_{\text{rel}}^{-1} = \frac{\omega}{k_B T v_j} \int_0^{\infty} dE \int_0^{\infty} d\tau P(E, \tau) \frac{\gamma_j^2}{\rho v_j^2} \frac{(\Delta/E)^2}{\cosh^2(E/2k_B T)} \frac{\omega\tau}{1 + (\omega\tau)^2}, \quad (31)$$

where τ is the relaxation time, E is the energy difference of the two levels, and γ_j is the deformation potential of the relaxing entity. $P(E, \tau)$ is the distribution function of these entities, which in the soft-potential model can be tunneling states or classical relaxors or something in between, depending on the potential parameters η and ξ and on the temperature. The factor $(\Delta/E)^2$ has to be taken into account in the quantum regime; in the classical regime it is unity.

At low temperatures, the relaxational absorption by the tunneling states dominates. Again, we take over the equations derived for the tunneling model,⁶

$$\tau_{\text{tun}}^{-1} = \left(\frac{\gamma_l^2}{v_l^5} + \frac{2\gamma_t^2}{v_t^5} \right) \frac{\Delta_0^2 E}{2\pi\rho\hbar^4} \coth \left(\frac{E}{2k_B T} \right), \quad (32)$$

and reproduce again the results of the tunneling model,⁶ namely, a rise of the inverse relaxational mean free path $l_{\text{rel,tun}}^{-1}$ with T^3 at very low temperatures and the plateau of the inverse mean free path with the height $\pi\omega C_j/2v_j$ at slightly higher temperatures around 1 K.

At still higher temperatures, one has to add the classical relaxation effects from potentials with higher barriers. The relaxation time and the barrier height for this case are given by Eqs. (15) and (16). The deformation poten-

tial is given by (8). Neglecting the tunnel splitting Δ_0 for these higher barriers one has $E = |\Delta|$. With these relations, one can calculate the Jacobian

$$|J| = \left| \frac{\delta(E, V)}{\delta(\eta, \xi)} \right| = 2\eta_L^{-3/2} V^{5/4} W^{3/4} \quad (33)$$

and write Eq. (31) in terms of E and V as

$$l_{\text{rel, class}}^{-1} = \frac{C_j}{v_j W^{3/4} k_B T} \times \int_0^\infty dE \int_0^\infty \frac{dV}{V^{1/4}} \cosh^{-2} \left(\frac{E}{2k_B T} \right) \frac{\omega^2 \tau}{1 + (\omega \tau)^2}. \quad (34)$$

Integrating over E and replacing the value of $V^{1/4}$ in the integral over V by its value at $\omega \tau = 1$ via Eq. (15), one gets

$$l_{\text{rel, class}}^{-1} = \frac{\pi \omega C_j}{v_j} \left(\frac{T}{W} \right)^{3/4} \ln^{-1/4}(1/\omega \tau_0). \quad (35)$$

E. Some remarks concerning microwave absorption

We are not going to provide here a detailed theory of microwave absorption and its comparison with experimental data. This will be done in a separate publication. Here we would like to give only a few remarks concerning this problem.

We can assume basically the same mechanism of coupling of the electric field to the soft modes with the only difference being that the soft-mode Hamiltonian is modulated by the external electric field rather than by a strain. This means that the action of an external ac electric field can be analyzed in the same way as the action of a strain using a quite simple correspondence between the two interaction Hamiltonians.

At temperatures and frequencies where activation processes in two-well potentials dominate and the (η, ξ) distribution is given by the seagull singularity, one gets, for the loss angle tangent,

$$\tan \delta \propto T^{3/4} \ln^{-1/4}(1/\omega \tau_0), \quad (36)$$

which corresponds to Eq. (35). In the same way one can find that the coefficient of resonant microwave absorption is proportional to ω^4 .

IV. COMPARISON TO EXPERIMENT

A. Choice of parameters

In the formulation of the soft-potential model given here, one needs four material parameters for a comparison to experimental data. Three of these, namely, \bar{P} , C_l , and C_t (or, alternatively, instead of the last two, γ_l and γ_t), can be taken from fits of low temperature data in terms of the tunneling model. We chose the recent data collection of Berret and Meissner⁴² as the basis of the comparison (the values C_l, C_t differ slightly from those in the reference because they were directly calculated

TABLE I. Material parameters of the soft-potential model for vitreous silica and amorphous selenium.

Substance	Vitreous silica	Amorphous selenium
Sound-wave parameters ^a		
ρ (kg m ⁻³)	2200	4300
v_l (m s ⁻¹)	5800	2000
v_t (m s ⁻¹)	3800	1050
Tunneling model parameters ^a		
\bar{P} (J ⁻¹ m ⁻³)	0.8×10^{45}	2.0×10^{45}
γ_l (eV)	1.04	0.25
γ_t (eV)	0.65	0.14
C_l	3.0×10^{-4}	1.9×10^{-4}
C_t	2.7×10^{-4}	2.2×10^{-4}
Crossover energy ^b		
W/k_B (K)	3.8	1.1

^aReference 42.

^bReferences 34 and 43.

from \bar{P} , γ_l , and γ_t). The missing fourth parameter is the crossover energy W . It can be determined from specific-heat data.^{34,43} Here we chose the values from an unpublished numerical fit,⁴³ which agree within 10% with our previously published data³⁴ [this paper contains two errors in the determination of the density of tunneling states, Eq. (39) of Ref. 34, which, fortunately, happen to cancel].

The comparison concentrates on data of vitreous silica, the most intensively studied case, and includes thermal conductivity data of amorphous selenium. The material parameters of these two glasses are compiled in Table I. Note that none of these parameters is adapted to sound-wave absorption data above 1 K.

B. Resonant absorption

The resonant absorption of sound waves below 1 K is well described in terms of the tunneling model. Its temperature, frequency, and sound-wave intensity dependence are discussed in detail in the literature^{5,6} and need not be discussed here. Instead, we focus on the behavior at higher temperatures and/or higher frequencies. In the resonant case, Eq. (29) for the tunneling states has to be taken together with Eq. (30), which describes the onset of the resonant absorption by the quasiharmonic vibrations. The two expressions join continuously at $\hbar\omega_{co} = (6\sqrt{2})^{1/3}W \approx 2W$ in the low-temperature limit (compare Sec. II C 4). If the temperature is low enough relaxations need not be considered, and the sound-wave absorption should change at ω_{co} from the linear tunneling model dependence to an ω^4 dependence at higher frequencies, as shown in a double-logarithmic plot in Fig. 1. The figure compares the prediction with experimental data of Dietsche and Kinder⁴⁴ and of Long, Hanna, and MacLeod.⁴⁵ The strong rise of the inverse mean free path with increasing frequency is seen both in experiment and model calculation.

C. Relaxational absorption

Figure 2 shows a comparison of calculated and measured⁴⁶ internal friction, again for vitreous silica. The internal friction Q^{-1} is related to the mean free path by $l^{-1} = \omega Q^{-1}/v_j$. The experimental conditions were such that the resonant contributions discussed in the preceding subsection should be negligible.

The calculated curves in Fig. 2 were obtained from the tunneling model for temperatures below W/k_B and from Eq. (35) with $\tau_0 = 10^{-13}$ s above that temperature. Their good fit below 2 K is not surprising, but rather emphasizes the good agreement between two measurements (the tunneling model parameters in Table I were determined from a similar measurement on vitreous silica by a different group⁴⁷). The difference between the predictions of the tunneling model and the soft-potential model is clearly seen above 4 K, where the classical relaxation contribution begins to appear. The experimental data show the predicted rise, though with a smaller slope. Above 30 K they begin to deviate strongly from the theoretical prediction, indicating a cutoff in the distribution function for higher barriers.^{4,29} The crossover region between tunneling and classical relaxation is not well reproduced. In particular, the experimental data at 45 MHz exceed the calculated data by a factor of nearly 2.

Figure 3 compares the prediction of Eq. (35) with Brillouin data⁴⁸ at higher temperatures. The frequency is already a sizeable fraction of the crossover frequency, but the resonant contribution from the soft vibrations should still be negligible. The agreement is surprisingly good up to temperatures of 50 K, where again the absorption begins to become smaller than the calculated values, possibly again due to the same high barrier cutoff. The Brillouin scattering in this cutoff region and its pressure dependence have been investigated in detail very recently.²⁹ The interpretation of these results was similar to the one

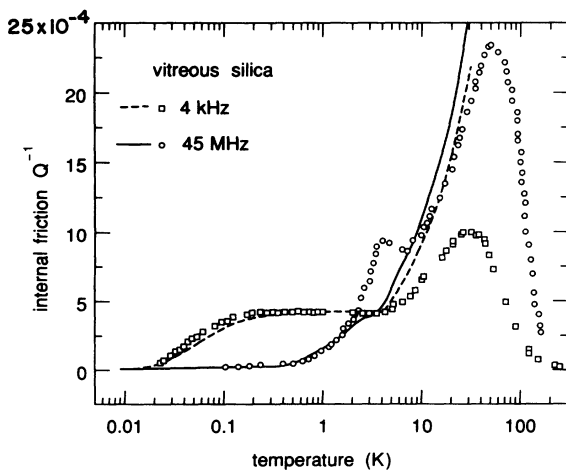


FIG. 2. Measured [open circles and squares: data by Raychaudhuri and Hunklinger (Ref. 46)] and calculated (continuous and dashed lines) values of internal friction for vitreous silica at two different frequencies as a function of temperature.

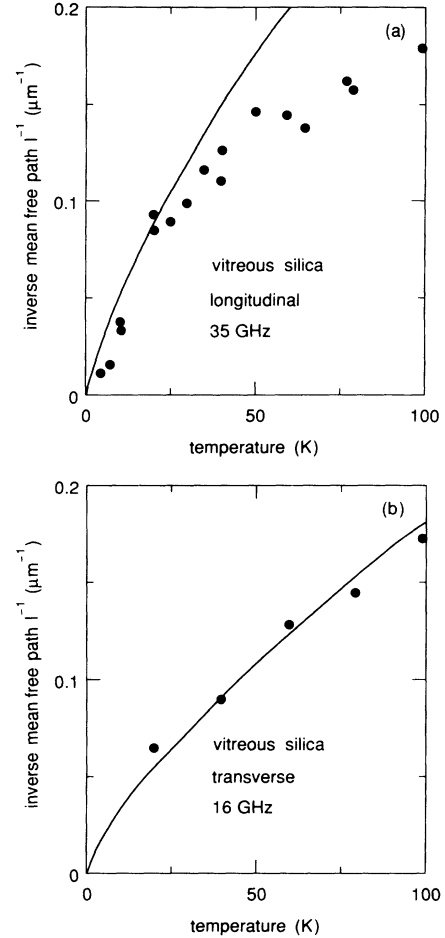


FIG. 3. Measured [full circles: Brillouin data by Vacher *et al.* (Ref. 48)] and calculated (continuous lines) inverse mean free path in vitreous silica as a function of temperature (a) for longitudinal sound waves at the frequency 35 GHz (b) for transverse sound waves at 16 GHz.

given here. It assumed a distribution of barriers able to explain the relaxation phenomena by tunneling states at very low temperatures and by classical relaxation at higher temperatures.

D. Thermal conductivity

The thermal conductivity between 0.1 and 10 K is dominated by sound waves with frequencies between 10^{10} and 10^{12} Hz. The crossover frequency $\omega_{co} \approx 2W/\hbar$ between resonant tunneling and resonant vibrations lies in the middle of this range. In order to calculate the thermal conductivity, we chose

$$l^{-1} = l_{res,tun}^{-1} + l_{rel,class}^{-1} \quad (37)$$

according to Eqs. (29) and (35) for frequencies below ω_{co} and

$$l^{-1} = l_{res,vib}^{-1} \quad (38)$$

according to Eq. (30) for frequencies above the crossover. Here we consider Eq. (35) as a good interpolation in the

temperature and frequency range relevant for the thermal conductivity. As one can see in Fig. 3, this interpolation gives an adequate description of the experimental data for frequencies of the order of W/h (16 and 35 GHz). Processes that we do not consider explicitly in this paper, such as relaxational absorption by the quasilocalized harmonic vibrations, can make a substantial contribution to the absorption in this frequency interval. Here we shall, however, limit ourselves to this comment, leaving a detailed analysis of it for a separate paper.

The thermal conductivity κ was calculated using the standard expression⁹

$$\kappa = \frac{1}{3} \int d\omega \sum_j C_{\text{Debye},j}(\omega, T) v_j l_j, \quad (39)$$

where $C_{\text{Debye},j}(\omega, T)d\omega$ is the specific heat contributed by longitudinal ($j = l$) and transverse ($j = t$) sound waves within the frequency interval $d\omega$, and l_j is the corresponding mean free path calculated as indicated above.

The result of these calculations, using the parameters in Table I, is compared with experimental data in vitreous silica⁴⁹ in Fig. 4 and with experimental data in amorphous selenium¹ in Fig. 5. As can be seen, the model calculations reproduce the plateau in the thermal conductivity with good accuracy. At still higher temperatures there is a second rise of the experimental data, which is not reproduced by the calculations and is outside the scope of the present model.

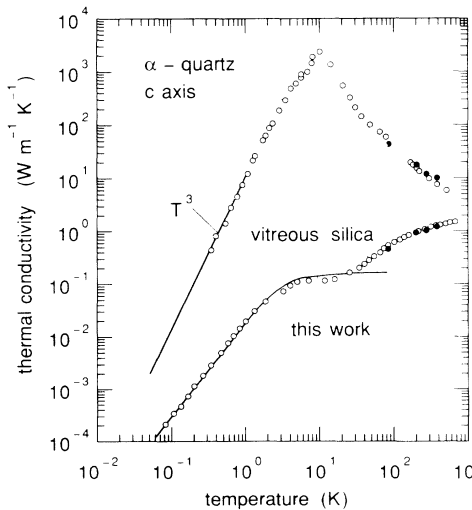


FIG. 4. Thermal conductivity of vitreous silica compared with that of crystalline α -quartz as a function of temperature. The open and full circles are experimental data reported by Cahill and Pohl (Ref. 49); the line marked " T^3 " was calculated from standard expressions for crystalline samples, and the second line was calculated as indicated in the text.

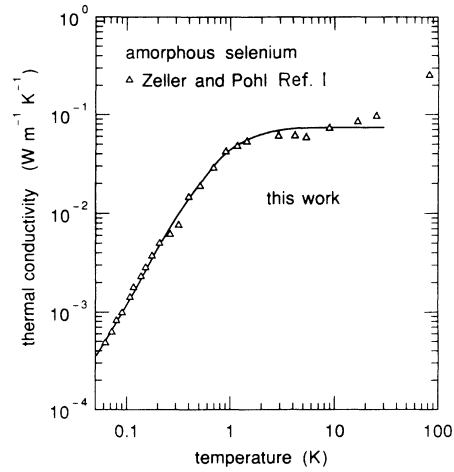


FIG. 5. Measured [triangles: data from Zeller and Pohl (Ref. 1)] and calculated (continuous line) thermal conductivity of amorphous selenium as a function of temperature.

V. DISCUSSION

The comparison with experiment in the preceding section shows that the soft-potential model is able to explain the main features of the anomalous sound-wave absorption in glasses up to temperatures of 20 K and up to frequencies approaching 1 THz. The tunneling model, valid below 1 K and below 10^{10} Hz, is contained as a limiting case. For higher temperatures, one has to include classical relaxation, and for higher frequencies, quasiharmonic vibrations become important. The soft-potential model enables a unified description with only one additional parameter, the crossover energy W , added to those of the tunneling model.

While the main features of the experiment are described, there are also marked deviations. If one looks for the source of the difference in the theoretical calculation, one could name several possible reasons. At the present stage, however, the quality of the approximations discussed in Sec. II C does not warrant speculation about the validity of the assumptions. This is especially true in the crossover region around W . The deviations between theory and experiment in that region (for instance, in the 45 MHz data in Fig. 2 around 4 K) could easily be due to the inaccurate description of the transition between the limiting cases.

The point illustrates the general present situation of the soft-potential model. It attempts to describe universally anomalous low-temperature glass properties in terms of a single picture for three different kinds of soft modes. At the origin of the potential parameter distribution ($\eta = \xi = 0$) one has the pure quartic potential, where the used asymptotic approximations break down. Further away from the origin, these approximations begin to be valid, but deviations from the simple assumption about the potential parameter distribution (assumption (ii) of Sec. II B concerning the seagull singularity)

may become noticeable. This makes it difficult to distinguish between the deviations from the assumptions and the errors due to the approximations. The good general agreement with experiment found so far is encouraging. However, if one wants to judge the limits of applicability of the model, one needs either numerical calculations or better approximations.

The explanation of the plateau in the thermal conductivity given here is similar to the ones given earlier by Karpov and Parshin,²⁶ Yu and Freeman,²⁷ and Grannan, Randeria, and Sethna.²⁸ Here we will not give a detailed comparison to that older work but merely note that our assumptions are simpler, more general, and allow us to describe the experimental data with a smaller number of parameters.

In the same way, our explanation of the rise of the relaxational absorption above 1 K is similar to the one proposed by Tielburger *et al.*²⁹ In that approach, a different form of the double-well potential and the assumption of a barrier-independent coupling lead to $l_{\text{rel,class}}^{-1} \sim T$ instead of the $T^{3/4}$ dependence of Eq. (35). Nevertheless, the basic idea is the same, though the extension to soft vibrations and the connection to the plateau in the thermal conductivity is missing.

It is interesting to compare the results of the present work with the universal description of the thermal conductivity of amorphous solids given by Freeman and Anderson.⁹ Their first rule for the mean free path l is $l \approx 150\lambda$, where λ is the wavelength of the sound waves. The rule holds for sound frequencies below 100 GHz. This is the region where the predictions of the soft-potential model coincide with those of the tunneling model.^{5,6} That first rule implies $C_j \approx 3.3 \times 10^{-4}$ in Eq. (29) (this universality has recently been stressed again²⁴). Their second rule is $l \approx (4 \times 10^{24} \text{ m}^{-3})\lambda^4$, which describes the universality of the plateau region. The λ^4 dependence is reproduced by Eq. (30) of the present work. The numerical prefactor can be translated into the condition $2W \approx hv_j/\lambda_{co}$ with a universal crossover wavelength λ_{co} of about 34 nm. From the experimental low-temperature data of Dietsche and Kinder⁴⁴ and Long, Hanna, and MacLeod⁴⁵ in Fig. 1, one infers together with the tunneling model prediction (29) crossover wavelengths of 23 and 21 nm, respectively, for vitreous silica. Numerical fits⁴³ of the specific heat in terms of the soft-potential model give W values corresponding to

crossover wavelengths of 26, 25, 23, and 16 nm in vitreous silica, amorphous selenium, α -B₂O₃ and amorphous polybutadiene, respectively.

The third rule of Freeman and Anderson,⁹ $l \approx \lambda$ at still higher frequencies, which describes the second rise of the thermal conductivity above 20 K,⁴⁹⁻⁵¹ is beyond the scope of this paper.

VI. CONCLUSIONS

The soft-potential model, a phenomenological model, which describes tunneling states, classical relaxors, and soft quasiharmonic vibrations in glasses in terms of one distribution of anharmonic soft potentials, has been used to calculate the sound absorption by using the *same* bilinear coupling constant for *all three types* of excitation. Good general agreement between measured and calculated sound-wave absorption up to temperatures of 20 K and frequencies of 500 GHz has been achieved. Three of the four required material parameters can be taken from tunneling model fits (\bar{P} , γ_l , and γ_t), since the soft-potential model can be formulated as an extension of the tunneling model. The fourth, the crossover energy W , can be taken from fits to specific heat. Without any further adaptable parameter, it is possible to describe the plateau in the thermal conductivity and the strong rise of the relaxational absorption above 1 K in reasonable agreement with experiments in vitreous silica and amorphous selenium.

Our approach does not share the notorious difficulty of other attempts¹⁷⁻²⁰ to explain the plateau in the thermal conductivity of amorphous solids, i.e., to understand quantitatively why phonons are scattered so strongly at long wavelengths of the order of 30 nm.¹¹⁻¹³

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