Theory of low-energy Raman scattering in glasses

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We work out a theory of low-energy Raman scattering in glasses. The scattering and the energy transfer are due to interaction of the light with the soft potentials in glasses. We exploit relations between the scattering cross sections at a certain energy transfer, $\hbar\omega$, and the mean free paths of sound and electromagnetic radiation for the frequency ω. A comparison between the theory and experiment is made. An explanation of the boson peak is proposed. We come to a conclusion that the peak may be due to a reconstruction of the low-energy density of states due to interaction between soft harmonic oscillators. The interaction is enhanced with the frequency ω and, as a result, soft harmonic vibrations, localized for comparatively small ω, become extended (but not propagated) for bigger values of ω.

I. INTRODUCTION

During the last two decades it was established that different glasses exhibit universal properties, which are usually regarded as anomalous compared to those of the crystalline prototypes.[1] The universal low-temperature properties of glasses (below a few Kelvin) are described well in the framework of the model of Anderson, Halperin, Varma, and Phillips (AHVP model).[2,3] This model postulates the existence of two-level systems (TLS's) with an almost constant density of states $\bar{P}$ in glasses. There are several articles where experimental data and their interpretation are reviewed on the basis of the AHVP model.[4-8]

However, above a few Kelvin the universal properties of glasses deviate from the predictions of the AHVP model. The thermal conductivity shows a plateau around 10 K, which cannot be understood in terms of a constant density of tunneling states.[9] The sound velocity decreases linearly with temperature above a few Kelvin.[10] Furthermore, there is an additional increase in the specific heat, indicating the existence of at least one other kind of low-frequency mode. Experimental data by Stolen[11] on far-infrared absorption as well as on the low-frequency Raman scattering have shown these to be low-frequency harmonic oscillator modes (see also the review paper by Jäckle[12]). This interpretation is supported by neutron measurements[13] that have shown in glasses the existence of soft harmonic vibrations with a crossover to anharmonicity at the low-frequency end (at frequencies corre-

sponding to several Kelvin).

All these experimental facts indicate that there is a common basis for the low-temperature (below a few Kelvin) and the higher-temperature (above a few Kelvin) universal properties of glasses and that the AHVP model describes only one part of them. In addition to the TLS's there are other low-energy excitations in glasses, which are, in particular, responsible for their properties at higher temperatures. Grace and Anderson[14] and Brand and von Löhneysen[15] on the basis of their experimental data have arrived at the conclusion that TLS's and these additional excitations have the same basic vibrational structure.

The experiments give only very limited information on the microscopic origin of the soft mode. A computer simulation of a glass of soft spheres[16] has shown the existence of (quasi) localized modes at low frequencies. These modes are formed by chains of atoms with side branches and are centered typically on 20 atoms, i.e., they have an effective mass of about 20 or more atomic masses. The modes are connected with structural anomalies of the glass. With increasing temperature relaxational modes of similar structures are observed.[17] Recent simulations on Se glass showed similar properties.[18] There the soft modes involve atomic chains as well as rings with effective masses of again typically twenty atomic masses. Both the soft vibrational modes and the relaxations can be considered as a collective motion of a group of atoms, which for this particular motion can be thought of as forming a quasimolecule.

These properties are predicted by the soft-potential

model (SPM), which was proposed in Ref. 19 and developed further in Refs. 20–26. A short review of this model and its application to description of universal properties of glasses is given in Ref. 27. The SPM explains all the universal low-temperature properties of glasses in the same way as the AHVP model.[23,27] In this model, besides the TLS's there exist also soft harmonic oscillators (HO) and both these types of low-energy excitations have a common basis, namely *soft atomic potentials*. The TLS's exist in the soft double-well potentials and the HO in the soft single-well ones. The soft harmonic excitations are the low-energy excitations, which are responsible for some universal properties of glasses at higher temperatures.[24,25,27]

Another type of excitations that influences the properties of glasses are the usual delocalized phonons, or hypersonic plane waves. In the paper we shall call them phonons or hypersound. The lower the frequency, the better these excitations are defined. At sufficiently high frequencies, where their mean free path becomes of the order of their wave length, they cease to exist (see Sec. VI).

The main purpose of this paper is to consider in glasses, within the framework of the soft-potential model, the low-frequency Raman scattering with comparatively small energy transfer. This model, under rather natural assumptions, allows one to treat the scattered light intensity as a sum of independent contributions of various sites (where the soft potentials reside), the linear dimensions of each mode being much smaller than the light wavelength. An equivalent description can be also formulated for the calculation of the hypersonic and infrared absorption. Such an approach permits us to exploit the idea formulated by Jäckle in Ref. 12 and to present the results for the Raman scattering in terms of other observable quantities, namely, coefficients of hypersound and infrared (or microwave) absorption. In such a way one can interrelate various experimental data and explore the range of applicability of the assumptions used.

For many years the most widely recognized model used for interpretation of the experiments on Raman scattering in glasses was that of Martin and Brenig.[28] Some attempts have been made to extend the initial ideas of the Martin-Brenig model and give an interpretation of some correlation or structural length in glasses.[29–34] Other models for some part of the inelastic light spectrum have been proposed. The quasielastic contribution has been attributed to some unknown damped oscillators.[35,36] The boson peak was also related to the vibrational density of states.[37] But up to now the physical origin of the experimental data, including all the aspects of the low-frequency Raman spectrum, remains unknown.

The approach we propose here is an alternative one. Therefore we compare the predictions of the soft-potential model with those of the model considering the combined action of the photons and fluctuations of polarizability of the glass (see Refs. 28 and 38).

II. SOFT-POTENTIAL MODEL

According to the SPM,[19] the quasilocal low-frequency modes in glasses are described by the soft anharmonic oscillator Hamiltonian

$$\mathcal{H} = -\frac{\hbar^2}{2M}\frac{d^2}{dx^2} + V(x) \, , \qquad (1)$$

where M is the effective mass of the considered "soft" entity and

$$V(x) = \mathcal{E}_0 \left[\eta \left[\frac{x}{a}\right]^2 + \xi \left[\frac{x}{a}\right]^3 + \left[\frac{x}{a}\right]^4 \right] \, . \qquad (2)$$

The description of a quartic potential by (2) is unique only if for $\eta > 0$ one restricts ξ by $\xi^2 < 32\eta/9$ and we will imply this restriction in the following. x is the generalized coordinate of the soft mode having units of length, a is a characteristic length of the order of the interatomic spacing ($a \simeq 1$ Å), $\mathcal{E}_0$ is of the order of binding energy of the atoms constituting the glass, i.e., of the order of 10 eV. The values of the dimensionless parameter η and ξ are random due to fluctuations of the structural parameters of a glass. Soft potentials correspond to $|\eta|, |\xi| \ll 1$. The distribution function of these parameters for $|\eta|, |\xi| \ll 1$ is given by[21]

$$P(\eta, \xi) = \frac{|\eta|}{2}\mathcal{P}_0 \, , \qquad (3)$$

where $\mathcal{P}_0$ is a constant and the factor $|\eta|$ describes a singularity in the distribution of the parameter η which in Ref. 22 was called "seagull" singularity. Since the glass is inversion symmetric on a macroscopic scale, the distribution function is even with respect to ξ, $P(\eta, \xi) = P(\eta, -\xi)$. The interaction of the soft atomic potentials, Eq. (1) with an elastic strain ϵ, is described by a bilinear term[23,24]

$$V_{\text{int}}(x) = \mathcal{E}_0\mathring{H} \left[\frac{x}{a}\right]\epsilon \, , \qquad (4)$$

with the dimensionless coefficient $|\mathring{H}|$ of order 1. This term describes the interaction of the strain field with both the TLS and of HO with the same coupling constants (different for the longitudinal and transverse sound). To introduce scales the dimensionless parameter η_L and W, the characteristic energy of the purely quartic potential, (2) for $\eta = \xi = 0$, are introduced

$$\eta_L = (\hbar^2/2Ma^2\mathcal{E}_0)^{1/3} \approx 10^{-2}, \quad W = \mathcal{E}_0\eta_L^2 \approx k\cdot 10 \, . \qquad (5)$$

We assume that the interaction Hamiltonian (4) does not depend on the parameters ξ and η.

If $|\xi|/\sqrt{\eta_L} < \eta_L/|\eta|$,[39] and, at the same time η is negative and within the limits $|\eta| > 3\eta_L$ (see Fig. 1) the two lowest levels in the potential (2) form a TLS with the energy splitting $E = \sqrt{\Delta_0^2 + \Delta^2}$. The tunnel splitting Δ_0 and asymmetry Δ are determined by

$$\Delta_0 \approx W \exp\left[-\frac{\sqrt{2}}{3}\left[\frac{|\eta|}{\eta_L}\right]^{3/2}\right] \, ,$$

$$\Delta = \frac{W}{\sqrt{2}}\frac{|\xi|}{\sqrt{\eta_L}}\left[\frac{|\eta|}{\eta_L}\right]^{3/2} \, . \qquad (6)$$

For $\Delta \ll V_b$ the height V_b of the barrier between the two minima in the double-well potential (2) does not de-
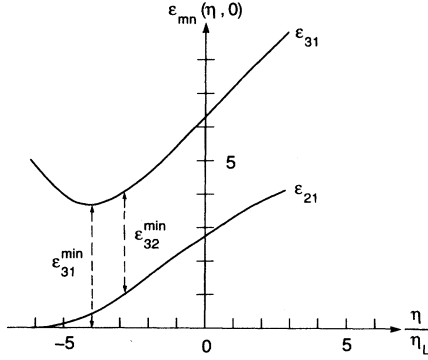
FIG. 1. The interlevel distance in the potential (2.1) (in units of W) as a function of η/η_L for $\xi=0$ (i.e., for symmetric soft potential). The arrows indicate the minimal distance between the levels $\varepsilon_{31}^{\min}=3.65$, $\varepsilon_{32}^{\min}=3$. These are the points of absolute minimum of the functions $\varepsilon_{31}(\eta,\xi)$ and $\varepsilon_{32}(\eta,\xi)$ on the plane η,ξ. The corresponding density of states makes a finite jump (from zero) at this energy—the so-called van Hove singularity.

pend on the value of ξ and is given by

$$V_b=\frac{W}{4}\left[\frac{\eta}{\eta_L}\right]^2 . \tag{7}$$

The density of states of TLS's as a function of their energy E is nearly constant[22,23] (throughout the paper we denote by energy E the interlevel spacing). It differs only slightly (by a logarithmic factor) from the corresponding distribution function in the AHVP model.[5]

It is clear from Fig. 1 that the TLS's picture is lost for small $|\eta|\simeq\eta_L$ when the distance to the third level becomes comparable with the TLS energy E. This means that the TLS's alone cannot be responsible for excitations with energies larger than W. Single-well potentials will also contribute to these excitations in glasses.

Besides the TLS's corresponding to double-well potentials with $\eta<0$, Eq. (2) describes also quasilocal anharmonic oscillators in single-well potentials with positive $\eta>9\xi^2/32$. The excitations in the single-well potentials with $\eta\gg\eta_L$ are nearly harmonic with the interlevel spacing

$$E=2W\sqrt{\eta/\eta_L} . \tag{8}$$

For $E\gg W$ the density of states $n(E)$, of these excitations is[21,24]

$$n(E)=\frac{1}{6\sqrt{2}}\frac{\mathcal{P}_0\eta_L^{5/2}}{W}\left[\frac{E}{W}\right]^4\propto E^4 . \tag{9}$$

It rapidly increases with the energy E.

III. LOW-FREQUENCY RAMAN SCATTERING

We are going to use the following physical considerations. We assume that the Raman scattering occurs at the soft-potential sites and the energy transfer is due to the motions of oscillating or tunneling entities in soft potentials. Every site can be looked upon as a quasi-

molecule immersed in a glass. The ac electric field of the light induces a dipole moment in each quasimolecule,

$$P_i=\chi_{il}E_l\exp(-i\omega_{\text{light}}t) ,$$

where ω_{light} is the frequency of light. This oscillating dipole moment is considered as the source of the scattering light.

Quasilocal soft modes modulate the dipole moments of the quasimolecules. This modulated part δP_i, which determines the Raman-scattering part, can be written as

$$\delta P_i(t)=\delta\chi_{il}(t)E_l\exp(-i\omega_{\text{light}}t) . \tag{10}$$

We assume that in the first approximation $\delta\chi_{il}(t)$ is proportional to x/a

$$\delta\chi_{il}(t)=\alpha_{il}(x/a) , \tag{11}$$

where the tensor α_{il} denotes the coefficient of proportionality which we call tensor of polarizability modulation. The time dependence of the coordinate x describes the soft motion that brings about the modulation of the electric dipole.

Following the standard theory of light scattering (see, for instance, Ref. 40) we can write for the differential cross section of light scattering $d\sigma$ proportional to the part of the light intensity scattered into the given solid angle $d\Omega$, and given frequency interval $d\omega$ (with ω the frequency shift)

$$d\sigma(\omega)=\frac{\omega_{\text{light}}^4}{a^2c^4}e_i'^*e_k'e_le_m^*\alpha_{il}\alpha_{km}\langle x(t)x(0)\rangle_\omega d\Omega\frac{d\omega}{2\pi} . \tag{12}$$

Here c is the velocity of light, e and e' are the polarization vectors of the ingoing and scattered electromagnetic waves, respectively, while $\langle x(t)x(0)\rangle_\omega$ is the Fourier component of the time correlation function:

$$\langle x(t)x(0)\rangle_\omega=\int_{-\infty}^\infty \langle x(t)x(0)\rangle e^{i\omega t}dt , \tag{13}$$

where $\langle \cdots \rangle$ denotes the ensemble average. In Eq. (12) we have assumed that

$$\omega/\omega_{\text{light}}\ll 1 ,$$

and throughout the paper we will neglect the terms proportional to this small parameter as compared to 1. In particular, this approximation permits us to consider tensor α as symmetric. Indeed, the tensor χ_{il} is assumed to correspond to the frequency interval where the glass is transparent. In the absence of the external magnetic field, it should be real and therefore symmetric. The tensor α describes the effect of the reaction of the electronic wave functions of the quasimolecule to the additional external perturbation by the atomic motion in the soft mode. This motion is slow, so that on the electronic time scale it may be considered as adiabatic, i.e., the motion should not break the symmetry of the tensor χ, and

$$\alpha_{il}=\alpha_{li} . \tag{14}$$

Equation (12) describes the effect of a single soft mode. Now we are going to sum over all the soft modes in the

unit volume. This can be easily done if there is no interference of the light scattered by different soft modes which can happen in two cases. First, when the generalized coordinates x of all quasimolecules are independent, i.e., the adjacent soft modes do not interact. Then the scattering by different quasimolecules is independent even though their modulated polarizabilities can be correlated (such a correlation could, in principle, be created during the glass transition).

It is more likely, however, that the polarizability tensors α_{il} are uncorrelated and their average values $\overline{\alpha_{il}}$ vanish. The latter is true provided the glass is inversion symmetric on a macroscopic scale. The tensor α is the coefficient of the first power of x in the expansion of the polarizability of a quasimolecule. This tensor, in its turn, enters the part of free energy that is associated with ac electric field,

$$-\frac{1}{4}\sum \delta\chi_{il}E_iE_l^* \; , \tag{15}$$

where the summation is over all the soft-potential sites. Under inversion x transforms into $-x$, and, as the contribution, Eq. (15), should be invariant we have

$$\overline{\alpha_{il}}=0 \; .$$

In this case we can write

$$\overline{\alpha_{il}^{(1)}\alpha_{mn}^{(2)}}=\overline{\alpha_{il}^{(1)}}\cdot\overline{\alpha_{mn}^{(2)}}=0 \; , \tag{16}$$

where the superscripts (1) and (2) refer to two different quasimolecules. In this case there would be no interference and one can sum the intensities of light scattered independently from all the soft-potential sites even if there is interaction between the soft potentials, so that different x are correlated.

Summing over all the soft modes within a unit volume we get the differential extinction coefficient dh, describing the part of light intensity scattered into a given solid angle and given frequency interval provided the ingoing light has passed a unit length. We shall perform the summation over the soft modes in two stages. First, we sum over all the quasimolecules with fixed values of ξ and η. As a result, the product $\alpha_{il}\alpha_{km}$ will be replaced by its average, $\overline{\alpha_{il}\alpha_{km}}$. Second, we integrate over all the values of ξ and η, with regard of their distribution function, Eq. (3). Assuming that, due to the slowness of the motion described by a soft potential, such electronic characteristics as $\alpha_{il}\alpha_{km}$ would not depend on ξ and η we get

$$dh = \frac{\omega_{\text{light}}^4}{a^2c^4}e_i'^*e_k'e_le_m^*\overline{\alpha_{il}\alpha_{km}}$$
$$\times \int \int d\eta\, d\xi P(\eta,\xi)\langle x(t)x(0)\rangle_\omega d\Omega\frac{d\omega}{2\pi} \; . \tag{17}$$

Provided the tensor α is symmetric the following relations hold in isotropic media:

$$\overline{\alpha_{il}\alpha_{km}}=v_1\delta_{il}\delta_{km}+v_2(\delta_{ik}\delta_{lm}+\delta_{im}\delta_{kl}) \; , \tag{18}$$

where

$$v_1=\tfrac{1}{15}[2\overline{(\text{Tr}\alpha)^2}-\overline{\text{Tr}\alpha^2}] \; , \tag{19}$$

$$v_2=\tfrac{1}{30}[-\overline{(\text{Tr}\alpha)^2}+3\overline{\text{Tr}\alpha^2}] \; , \tag{20}$$

and therefore

$$e_i'^*e_k'e_le_m^*\overline{\alpha_{il}\alpha_{km}}=[v_1|\mathbf{e}'^*\mathbf{e}|^2+v_2(1+|\mathbf{e}'\mathbf{e}|^2)] \; . \tag{21}$$

This equation determines the angular dependence of the light scattering, i.e., the dependence of the extinction coefficient on the polarizations of the incoming and scattered light. In general, the polarization vectors in this equation can be complex. This means that also the case of circularly (in general, elliptically) polarized light is included.

To analyze the angular dependence of the linearly polarized scattered light, let us introduce the depolarization ratio

$$D=I_{HV}/I_{VV} \; . \tag{22}$$

Here I_{VV} stands for the intensity of scattered light with the same polarization as the incident light. To be specific we take here a polarization perpendicular to the plane of scattering. I_{HV} is the intensity of scattered light polarized within the plane of scattering. For linear polarizations of the incoming and scattered light one gets from (21) for the depolarization ratio D

$$D=\frac{v_2}{v_1+2v_2}=\frac{1}{2}\frac{3\overline{(\text{Tr}\alpha)^2}-\overline{(\text{Tr}\alpha)^2}}{2\overline{\text{Tr}\alpha^2}+\overline{(\text{Tr}\alpha)^2}}\leq\frac{3}{4} \; . \tag{23}$$

It does not depend on the frequency ω as long as $\overline{\alpha_{il}\alpha_{km}}$ is independent of ξ and η. Investigating the frequency dependence of the depolarization ratio found in experiment one can check the validity of this assumption.

So far we considered coordinates x as classical. As we are going to make a definite distinction between the Stokes and anti-Stokes light scattering, we should in fact use the quantum-mechanical description. This means that $x(t)$ should be replaced by the corresponding operator in the Heisenberg representation [see Eq. (28) and the text below it]. As is shown in Ref. 40, after this replacement Eqs. (17) and (12) remain valid if one takes the average $\langle x(t)x(0)\rangle$ in Eq. (13) to include both statistical- and quantum-mechanical averaging.

As the operators $x(t)$ at different t, in general, do not commute, their order is important. As shown in Ref. 40, Eq. (17) gives the proper order of the operators for the Stokes scattering.

IV. GENERAL EXPRESSIONS FOR THE SOUND AND INFRARED ABSORPTION

In the previous section we have related the extinction coefficient due to the scattering of light by soft potentials to the correlation function of the soft coordinates, x. Now we make use of the fact that the coefficients of sound and infrared absorption can be expressed through the same function, or rather, through a function, closely related to it.

Let us assume that a glass is subject to the action of an external perturbation of the form

$$V(t)=-f(t)x \; , \tag{24}$$

periodic in time with $f(t)$ a generalized force.

Let B be the operator of some macroscopic physical quantity whose average we wish to calculate. We assume that in the absence of the perturbation the average vanishes. Then within the theory of linear response one can write for a Fourier component of the average value

$$\langle B \rangle_\omega = b(\omega) f_\omega , \qquad (25)$$

where f_ω is the Fourier component of the generalized force, while

$$b(\omega) = \int_0^\infty dt\, e^{i\omega t} b(t) . \qquad (26)$$

Here

$$b(t) = (i/\hbar)\langle [x(t), B] \rangle \qquad (27)$$

and

$$x(t) = \exp(iH_0 t/\hbar) x \exp(-iH_0 t/\hbar) , \qquad (28)$$

where H_0 is the Hamiltonian of the system including all the interactions except the perturbation Eq. (24) itself. The fluctuation-dissipation theorem (FDT), see the Appendix, states that the imaginary part $b''(\omega)$ of the generalized transport coefficient is related to the Fourier component of the corresponding correlation function by

$$b''(\omega) = \frac{1}{2\hbar}(1 - e^{-\hbar\omega/kT})\langle x(t)B \rangle_\omega . \qquad (29)$$

Let us apply these general statements to two examples: ultrasonic and infrared absorption.

A. Ultrasonic absorption

In this case

$$V(t) = \mathscr{E}_0 \acute{H} \frac{x}{a} \epsilon(t) , \qquad (30)$$

where $\epsilon(t)$ is the strain produced by the sound wave whence

$$f(t) = -\frac{\mathscr{E}_0 \acute{H}}{a}\epsilon(t) . \qquad (31)$$

Here we wish to emphasize that, in the spirit of the ideas expressed above, the interaction Hamiltonian (4) has two physically entirely different meanings. First, it describes interaction of the ac strain with the soft potentials. According to the theory of linear response this term should not be included into H_0. Secondly, it also describes the interaction with the thermal acoustic plane waves (phonons), which should be included into Hamiltonian H_0.

In this case we take for the operator B in (25) the elastic stress tensor σ, where

$$\sigma = \frac{\partial V}{\partial \epsilon} = \mathscr{E}_0 \acute{H} \frac{x}{a} . \qquad (32)$$

In the spirit of the general relations given above we get for the elastic modulus $\lambda(\omega)$, as the proportionality coefficient between $\langle \sigma \rangle_\omega$ and ϵ_ω:

$$\langle \sigma \rangle_\omega = \lambda(\omega)\epsilon_\omega , \qquad (33)$$

the following equation

$$\lambda(\omega) = -\frac{\mathscr{E}_0 \acute{H}}{a} b(\omega)$$

$$= -\left[\frac{\mathscr{E}_0 \acute{H}}{a}\right]^2 \frac{i}{\hbar} \int_0^\infty dt\, e^{i\omega t}\langle [x(t), x(0)] \rangle . \qquad (34)$$

Using the FDT (29) we derive for the imaginary part of the elastic modulus

$$\lambda''(\omega) = -\left[\frac{\mathscr{E}_0 \acute{H}}{a}\right]^2 \frac{1}{2\hbar}(1 - e^{-\hbar\omega/kT})\langle x(t)x(0) \rangle_\omega . \qquad (35)$$

The coefficient of sound absorption l^{-1} is related to $\lambda''(\omega)$ by

$$l^{-1} = -\frac{\omega}{\rho v^3}\lambda''(\omega) . \qquad (36)$$

Here ρ is the mass density of the glass, while v is the sound velocity.

So far we considered the contribution of a single soft mode to the sound absorption. Actually, to get the absorption coefficient, one should sum over all such modes within the unit volume:

$$l^{-1} = -\frac{\omega}{\rho v^3}\lambda''(\omega) = \frac{\omega}{2\rho\hbar v^3}\left[\frac{\mathscr{E}_0 \acute{H}}{a}\right]^2(1 - e^{-\hbar\omega/kT})$$

$$\times \int \int d\eta\, d\xi P(\eta,\xi)\langle x(t)x(0) \rangle_\omega . \qquad (37)$$

To get this result we need, in full analogy with the Raman scattering, either absence of the correlation between different x or validity of the relation $\acute{H}^{(1)}\acute{H}^{(2)} = 0$. We will see, however, in the following that Eq. (37) breaks down when the interaction between the soft potential becomes important.

Actually one should discriminate between longitudinal and transverse sound absorption. To do this, one should introduce the indices of polarization for the constants $\acute{H}$ and v, as well as for the absorption coefficient l^{-1}.

B. Far-infrared absorption

For the sake of definiteness we are speaking here about infrared absorption although our results will, of course, be equally applicable to microwave absorption. We start by writing the Hamiltonian of interaction of a soft mode with an ac electric field $\mathbf{F}(t)$ (see Refs. 23 and 24)

$$V(t) = -\mathbf{F}(t)\mathbf{d}_0 \frac{x}{a} . \qquad (38)$$

Here $\mathbf{d}_0 = q^* a \mathbf{n}$, while q^* is the "effective charge" and $\mathbf{n}$ is a unit vector parallel to the electric dipole moment of the soft mode. The generalized force of (24) is now

$$f(t) = \mathbf{F}(t)\mathbf{d}_0/a . \qquad (39)$$

and the operator B in (25) is the electric dipole moment operator of the soft mode

$$\mathbf{p} = \mathbf{d}_0(x/a) . \qquad (40)$$

Acting in the same way as above we introduce $\chi(\omega)$ as the tensor relating the Fourier components of the electric field and the dipole moment

$$\langle \mathbf{p} \rangle_\omega = \chi(\omega) \mathbf{F}_\omega \ . \tag{41}$$

Its imaginary part averaged over all the directions of $\mathbf{d}_0$ is given by

$$\chi''(\omega) = \frac{1}{3} \left[\frac{d_0}{a} \right]^2 \frac{i}{\hbar} \int_0^\infty dt \, e^{i\omega t} \langle [x(t), (0)] \rangle \ . \tag{42}$$

The FDT gives us for this case

$$\chi''(\omega) = \frac{1}{6\hbar} \left[\frac{d_0}{a} \right]^2 (1 - e^{-\hbar\omega/kT}) \langle x(t) x(0) \rangle_\omega \ . \tag{43}$$

The coefficient of infrared absorption $\beta(\omega)$ is related to $\chi''(\omega)$ by

$$\beta(\omega) = \frac{4\pi\omega}{c\sqrt{\varepsilon}} \chi''(\omega) \ , \tag{44}$$

where ε is the dielectric susceptibility of the glass. Summing over all soft potentials we obtain

$$\beta(\omega) = \frac{2\pi\omega}{3c\hbar\sqrt{\varepsilon}} \left[\frac{d_0}{a} \right]^2 (1 - e^{-\hbar\omega/kT})$$
$$\times \int \int d\eta \, d\xi P(\eta, \xi) \langle x(t) x(0) \rangle_\omega \ . \tag{45}$$

This is the equation for the absorption coefficient that can be compared with the data for the light scattering.

The condition for applicability of Eq. (45) is either the absence of correlation between different x or lack of correlation between the dipole moment components for different soft potentials:

$$\overline{d_{0i}^{(1)} d_{0k}^{(2)}} = \overline{d_{0i}^{(1)}} \cdot \overline{d_{0k}^{(2)}} = 0 \ , \tag{46}$$

and the property

$$\overline{d_{0i}} = 0$$

(which again reflects the fact that macroscopically the glass has a center of symmetry). In the latter case we can use the expression (45) for β also if the soft potentials interact.

V. FREQUENCY AND TEMPERATURE DEPENDENCE OF HYPERSOUND ABSORPTION

As we have seen in the previous sections, as a consequence of the FDT the frequency and temperature dependence of the Stokes Raman-scattering intensity is related to the absorption coefficient of hypersound[27] (as well as to that of infrared radiation) by the following simple relations that can be checked on experiment [see Eqs. (17), (37), and (45)]

$$I(\omega, T) \propto \frac{[1 + N(\omega)]}{\omega} l^{-1}(\omega, T) \ , \tag{47}$$

$$I(\omega, T) \propto \frac{[1 + N(\omega)]}{\omega} \beta(\omega) \ . \tag{48}$$

Of course, the same conclusions are valid for the anti-Stokes scattering with replacement of $N(\omega) + 1$ by $N(\omega)$. Here $N(\omega)$ is the Bose factor $N(\omega) = [\exp(\hbar\omega/kT) - 1]^{-1}$ for temperature T. As the phenomena discussed above are closely related within SPM it is sufficient to investigate in detail the temperature and frequency dependence of either of them. For this we choose the hypersound absorption.

We restrict ourselves to consideration of the case of not very small temperature where $kT \gg W$. This inequality covers most of the experiments on Raman scattering in glasses. In this case within the SPM there are the following contributions to the hypersound absorption.

A. Processes associated with double-well potentials

The contribution of the double-well potentials to the coefficient of relaxational sound absorption is given by[24]

$$l_{\mathrm{rel,DWP}}^{-1} = \frac{\pi\omega C}{v} \left[\frac{kT}{W} \right]^{3/4} \ln^{-1/4} \frac{1}{\omega\tau_0}, \quad C = \frac{\mathcal{P}_0 \bar{H}^2 W}{\rho v^2 \sqrt{\eta_L}} \ . \tag{49}$$

The relaxation processes are due to thermally activated hopping over the barrier of the height V_b with the relaxation time $\tau = \tau_0 \exp(V_b/kT)$. The dimensionless parameter C is equivalent to the parameter $\bar{P}\gamma^2/\rho v^2$ in the AHVP model, where γ is the deformation potential of the TLS's.

B. Processes associated with quasiharmonic oscillators

There are two kinds of absorption processes by quasiharmonic oscillators, resonant and relaxational ones. Both can be described by the bilinear coupling between the hypersound and oscillators given by Eq. (4).

1. Resonant attenuation

Resonant attenuation is due to direct transitions between the levels of the harmonic oscillators accompanied by a variation of a phonon occupation number. The transition amplitude is the matrix element of the bilinear coupling Hamiltonian.[24] For the absorption coefficient we have

$$l_{\mathrm{res,HO}}^{-1} = \frac{\pi}{6\sqrt{2}} \frac{C\omega}{v} \left[\frac{\hbar\omega}{W} \right]^3 \ . \tag{50}$$

This process is characterized by a very steep frequency dependence ($\propto \omega^4$).

2. Relaxational absorption

The relaxational absorption due to the bilinear coupling to the quasiharmonic oscillators needs a more careful discussion. The absorption mechanism is effective if

$$\hbar\omega \ll kT \tag{51}$$

and only if the equilibrium between different quantum levels of the oscillator is shifted by the hypersonic wave.

For this to happen the diagonal matrix elements, $\langle n|x|n \rangle$, should not vanish and should be different for different n. This can only be the case if one accounts for the anharmonicity (due to the term ξx^3) in the potential (2) when calculating the coupling. Otherwise the oscillator can be treated as harmonic. This is why we use the term *quasiharmonic*.

A sound wave traveling through a glass modulates the distance between the levels of the HO and thus shifts the thermal equilibrium. Because of the finite value of the relaxation time τ, the response of the HO lags behind the strain of the sound wave ϵ, resulting in energy dissipation and hence sound absorption. In the classical frequency region $\hbar\omega \ll kT$ this absorption is given by a formula,[25,27] analogous to the one obtained for the relaxational ultrasonic absorption in glasses due to TLS's (Refs. 41 and 5)

$$l^{-1}_{\rm rel,HO} = \frac{1}{\mathcal{V}} \sum_{\rm HO} \frac{D^2}{\rho v^3} \frac{1}{4kT \sinh^2(E/2kT)} \frac{\omega^2 \tau}{1+\omega^2\tau^2} \ , \quad (52)$$

where $\mathcal{V}$ is the volume of the glass, τ is the relaxation time of the HO, and D is the deformation potential of the HO:

$$D = \frac{\partial E}{\partial \epsilon} \ . \quad (53)$$

In order to calculate $l^{-1}_{\rm rel,HO}$ we need the deformation potential D and the relaxation time τ of the HO. We can derive them within the framework of the approach developed in Refs. 23 and 24.

The interaction of the soft atomic potential Eq. (2), with a strain ϵ is described by the bilinear term Eq. (4). We can transform the total potential energy $V(x)+V_{\rm int}(x)$ into the form of Eq. (2) again by shifting the origin of the generalized coordinate system x. As a result, coefficient η in Eq. (2) is changed. For small strains ϵ,

$$\eta \to \eta - \frac{3}{2} \frac{\xi}{\eta} \acute{H}\epsilon \ . \quad (54)$$

Using Eqs. (54) and (8) we can calculate the deformation potential D

$$D = \frac{\partial E}{\partial \epsilon} = \frac{dE}{d\eta} \frac{\partial \eta}{\partial \epsilon} = -12 \frac{\acute{H}W}{\eta_L^2} \xi \left[\frac{W}{E}\right]^3 \ . \quad (55)$$

It is proportional to the asymmetry coefficient ξ of (2). This is to be expected because for symmetrical potentials ($\xi=0$) all the diagonal matrix elements vanish: $\langle n|x|n \rangle =0$. Since the density of states (9) and the relaxation time [see (60)] do not depend on ξ, the relaxational absorption (52) is determined by the value of D^2 averaged over ξ in the interval $-\sqrt{32\eta/9} < \xi < \sqrt{32\eta/9}$

$$\overline{D^2} = \tfrac{1}{2}\sqrt{9/32\eta} \int_{-\sqrt{32\eta/9}}^{\sqrt{32\eta/9}} D^2(\xi)d\xi \ , \quad (56)$$

which gives for the mean-square average value

$$\overline{D} \equiv \sqrt{\overline{D^2}} = 12 \left[\frac{2}{3}\right]^{3/2} \frac{\acute{H}W}{\eta_L^{3/2}} \left[\frac{W}{E}\right]^2 \ . \quad (57)$$

For $E \simeq 3W$, its magnitude is of the order of the deformation potential

$$\gamma \simeq |\acute{H}|W/\eta_L^{3/2} \simeq 1 \text{ eV}$$

for TLS's (Ref. 23), and it decreases with increasing energy E.

The relaxation time τ of the HO is determined by one-phonon processes, i.e., resonant absorption and emission of phonons. The probability of these processes is determined by the off-diagonal matrix element, Eq. (2)

$$\mathscr{E}_0\acute{H}\left\langle N,N_{\rm q} \left| \frac{x}{a}\epsilon \right| N_{\rm q}+1,N-1 \right\rangle = V_{\rm q}\sqrt{N(N_{\rm q}+1)} \ , \quad (58)$$

where

$$V_{\rm q} = i\frac{W\acute{H}}{\eta_L^{3/2}}\sqrt{W/E}\sqrt{(\hbar/2\rho\mathcal{V}\omega_{\rm q})}q \ . \quad (59)$$

Here $\omega_{\rm q}$ is the frequency of the phonon with wave vector $\mathbf{q}$, while N and $N_{\rm q}$ are the occupation numbers of HO and phonons, respectively. Using the "golden rule" we obtain

$$\frac{1}{\tau} = \frac{1}{\tau(E)} = \left[\frac{W\acute{H}}{\eta_L^{3/2}}\right]^2 \frac{WE^2}{2\pi\rho\hbar^4 v^5} \equiv \frac{W}{\hbar}\left[\frac{E}{E_c}\right]^2 \ , \quad (60)$$

where the energy E_c is defined by

$$E_c \equiv \sqrt{2\pi\rho\hbar^3 v^5}\left[\frac{\acute{H}W}{\eta_L^{3/2}}\right]^{-1} \ . \quad (61)$$

For example, for a-SiO$_2$, $E_c/k = 50$ K [we take for v the transverse sound velocity $v_t = 3.8\times10^5$ cm/s; $\rho=2.2$ g/cm^3 and $\acute{H}W/\eta_L^{3/2}=1$ eV (Ref. 23)]. The relaxation time τ of the HO does not depend on the temperature.

We can rewrite Eq. (52) for the relaxation absorption through the integral over the HO energy E, Eq. (8), with the density of states $n(E)$ (9):

$$l^{-1}_{\rm rel,HO} = \int_{E_0}^{\infty} dE\, n(E)\frac{\overline{D}^2}{\rho v^3}\frac{1}{4kT \sinh^2(E/2kT)}\frac{\omega^2\tau}{1+(\omega\tau)^2} \ . \quad (62)$$

Here E_0 is the minimum energy when we can still use the concept of the HO. From Fig. 1 it is clear that the first levels in the potential (2) are more or less equidistant beginning with the energy E of the order of 3 W. Therefore, we take $E_0 \approx 3$ W. The jump of the partial density of states between the second and the third levels occurs just at this energy (for energies $E \ll E_0$ we have to use the TLS's conception). From Eq. (62), using Eqs. (9), (57), and (60) we obtain

$$l^{-1}_{\rm rel,HO} = \frac{16\sqrt{2}}{9}\frac{C\omega}{v}b^2\int_{x_0}^{\infty}\frac{dx}{\sinh^2 x}\frac{x^2}{x^4+b^4} \ , \quad (63)$$

where

$$x_0 = \frac{E_0}{2kT} \quad \text{and} \quad b = \frac{E_c}{2kT}\sqrt{\hbar\omega/W} \ . \quad (64)$$

In the following, we will analyze the absorption (63) only in the temperature region

$$kT \gg W \tag{65}$$

(i.e., $x_0 \ll 1$), since at low temperatures, $kT \ll W$, the HO are not excited and their contribution to the absorption, Eq. (63), is exponentially small. In the temperature region (65) there are three different frequency intervals: low frequencies, where the main contribution to the absorption is due to the HO with typical values of $\omega\tau \ll 1$, intermediate frequencies, where typically $\omega\tau \approx 1$ and high frequencies, where $\omega\tau \gg 1$. For our purpose only the second case is important, i.e., the case of intermediate frequencies, $x_0 \ll b \ll 1$, or

$$W \left[\frac{E_0}{E_c} \right]^2 \ll \hbar\omega \ll W \left[\frac{2kT}{E_c} \right]^2 . \tag{66}$$

This is the usual frequency region for Brillouin- and Raman-scattering measurements. In this case we can replace x_0 by 0 in Eq. (63). The main contribution to the absorption comes from the HO with $x \approx b$ ($\omega\tau \approx 1$). From Eq. (63) we derive

$$l_{\mathrm{rel,HO}}^{-1} = \frac{16\sqrt{2}}{9} \frac{C\omega}{v} b^2 \int_0^\infty \frac{dx}{b^4 + x^4}$$
$$= \frac{16\pi}{9} \frac{C\omega}{v} \underbrace{\frac{kT}{E_c} \sqrt{W/\hbar\omega}}_{\gg 1} \propto T\sqrt{\omega} . \tag{67}$$

The absorption in this case is proportional to the temperature and to $\sqrt{\omega}$. Its magnitude can be comparable with the relaxation absorption (49) from the TLS's.

The same three contributions characterize the temperature and frequency dependencies of the infrared absorption in glasses. To illustrate this point, we shall give a simple relation between the absorption coefficient β and l:

$$\beta l = \frac{4\pi}{3} \frac{\rho v^3}{c\sqrt{\varepsilon}} \left[\frac{d_0 \acute{H}}{\mathscr{E}_0} \right]^2 . \tag{68}$$

This relation follows directly from comparison of Eqs. (37) and (45).

VI. BOSON PEAK

The steep rise of the resonant scattering of phonons by HO (50) implies that at some energy E_d, the mean free path of phonons reaches the Ioffe-Regel limit, i.e., it decreases to their wavelength.[21,42] The value of this energy is estimated as[42]

$$E_d \approx (0.6 - 0.75) WC^{-1/3} . \tag{69}$$

For example, for a-SiO$_2$, $W/k \approx 4$ K, $C \approx 3 \times 10^{-4}$, and $E_d/k \approx 45$ K. At this energy another process will become dominant where excitation from one oscillator hops to another directly over the distance of the wavelength (compare with the Einstein model of thermal conductivity;[43] see also Ref. 44, where the application of the Einstein model to glasses is discussed in detail). In other words, under these circumstances the oscillators interact strongly and the picture of independent quasilocal harmonic excitations is lost. Phonons and HO with energies of the order of E_d cannot be considered as independent excitations but are intermixed with each other. Above this energy the total density of states should be reconstructed. One may ask whether this phenomenon can explain the bump in the specific heat $C(T)/T^3$ at temperature of the order $E_d/5k$, the rise of the thermal conductivity above the plateau, and the "boson" peak at frequency $\hbar\omega \simeq E_d$ in the Raman scattering in glasses?[27]

Let us discuss interaction between the soft harmonic oscillators in more detail. The physics of the interaction is basically the same as that indicated by Black and Halperin[45] for TLS's at low energies. As we have seen, soft harmonic oscillators are coupled to strain field by a bilinear term, Eq. (4). Due to this coupling each soft harmonic oscillator is surrounded by a strain field that is proportional to its coordinate, x_s. Such a field acts on another oscillator. As a result, any two oscillators interact, the Hamiltonian of interaction being given by

$$J_{ss'} x_s x_{s'} \quad (s \neq s') . \tag{70}$$

One can give the following order-of-magnitude estimate for $J_{ss'}$:

$$J_{ss'} \approx \left[\frac{\mathscr{E}_0}{a} \right]^2 \frac{\acute{H}_s \acute{H}_{s'}}{\rho v^2} \frac{1}{r_{ss'}^3} , \tag{71}$$

where $r_{ss'}$ is the distance between the sites where the soft oscillators reside. Here we disregard the tensor structure of $\acute{H}_s$ and $\acute{H}_{s'}$; however, we wish to emphasize that they can be effectively of the same as well as of the opposite signs.

To discuss the interaction we shall investigate pairs of "resonant" oscillators with almost the same energy (interlevel spacing) which we shall denote by E. Making use of the explicit expressions for the matrix elements of x [see below—Eq. (89)] we can write for the off-diagonal matrix element $I(E)$ for transitions of two such oscillators separated by distance r

$$I(E) \approx \mathscr{D}(E)/r^3 , \tag{72}$$

where

$$\mathscr{D}(E) = \frac{\hbar^2}{2ME} \left[\frac{\mathscr{E}_0 \acute{H}}{a} \right]^2 \frac{1}{\rho v^2} = \frac{W^2 \acute{H}^2}{\rho v^2 \eta_L^3} \frac{W}{E} . \tag{73}$$

Consider oscillators with the energies within the interval $E, E + \Delta E (\Delta E \ll E)$. The average distance $\bar{r}$ between these oscillators can be estimated from the equation

$$n(E)\Delta E \approx 1/\bar{r}^3 . \tag{74}$$

Then, for the off-diagonal matrix element we have the following estimate:

$$I(E) \approx \frac{\mathscr{D}(E)}{\bar{r}^3} \approx \mathscr{D}(E) n(E)\Delta E . \tag{75}$$

Inserting Eq. (9) and Eq. (73) into Eq. (75) and making use of Eq. (69) we end up with the following relation:

$$I(E) \approx (E/E_d)^3 \Delta E . \tag{76}$$

Interaction between two oscillators may be considered as a small perturbation if the modulus of the off-diagonal matrix element is smaller than the difference of their energies, ΔE. *Vice versa*, if it is bigger than the difference ΔE the states of the oscillators become *collective*, and the same is true for a greater number of oscillators. Equation (76) tells us that if $E \ll E_d$ the matrix element is much smaller than ΔE which means a negligibly small interaction. On the other hand, if $E \gg E_d$ the interaction is so strong that one may expect completely reconstructed states. They become delocalized and spread over a large number of soft oscillators. In particular, the typical energy difference is of the order of $I(E) \gg \Delta E$. As a consequence of the term repulsion the eigenstates are distributed more or less uniformly over any small energy interval that still contains a big number of levels. This makes a strong contrast to the distribution of eigenstates of a set of noninteracting oscillators where there is virtually no level repulsion.

Let us estimate the reconstructed density of states of these new delocalized harmonic modes:

$$\bar{n}(E) \approx \frac{n(E)\Delta E}{\mathcal{D}(E)n(E)\Delta E} = \frac{1}{\mathcal{D}(E)} \propto E , \qquad (77)$$

i.e., it has a linear dependence of the energy. It is interesting to investigate by order-of-magnitude estimates up to which energies Eq. (77) is valid. Using the estimate $|\tilde{H}| \approx 1$ and (5) we can write

$$\bar{n}(E) \approx \frac{\rho v^2 E}{\eta_L^3 \mathcal{E}_0^3} . \qquad (78)$$

For ρv^2 we use the estimate (see, for instance, Ref. 46) $\rho v^2 \approx \mathcal{E}_0 / a^3$. The frequency ω_c of the maximum of the density of states is approximately (see Ref. 24)

$$\hbar \omega_c \approx \mathcal{E}_0 \eta_L^{3/2} ,$$

and, therefore,

$$\bar{n}(E) \approx \frac{E}{a^3 (\hbar \omega_c)^2} . \qquad (79)$$

This equation is valid for $E \ll \hbar \omega_c$. In other words, we can conclude that with increasing energy E the spectral density of delocalized soft vibrations in a glass approaches gradually the Debye density of states.

The reconstructed density of states has an important property of self-consistency. The number of states per unit volume up to some arbitrary energy E is

$$\mathcal{N} \approx \frac{E^2}{a^3 (\hbar \omega_c)^2} . \qquad (80)$$

By the method given above one can estimate the characteristic energy that determines the repulsion of the eigenstates belonging to this interval

$$I(E) \approx 1/\bar{n}(E)\bar{r}^3 \approx \mathcal{N}/\bar{n}(E) , \qquad (81)$$

where we have used

$$\bar{r}^3 \approx \mathcal{N}(E)^{-1} . \qquad (82)$$

From (81) one sees that the energy determining the scatter of the eigenvalues due to their repulsion is E or, in other words, the density of states $\bar{n}(E)$, is determined by a self-consistent procedure.

The approach developed above is an alternative to the one used in Refs. 47 and 48, where a linear energy dependence of the density of states of harmonic oscillators was predicted too, above some energy of the order of a few meV. The difference is that in Refs. 47 and 48, the density of states of quasilocalized *noninteracting* (or independent) harmonic oscillators was considered. Linear energy dependence comes in that case from the special form of the distribution function of η and ξ which has been proposed. In our case density of states above E_d is density of states of the whole ensemble of *interacting* harmonic oscillators. It does not depend on the distribution function of η and ξ at all.

Generally speaking, for the energies higher than E_d, the theory of hypersound absorption discussed above ceases to be applicable. The general relations for λ'', the imaginary part of the elastic moduli, still remain valid. However, as the imaginary part is, in general, not smaller than the real one, Eq. (36) relating λ'' to the mean free path and therefore Eq. (37) break. This happens because the hypersound mean free path in the vicinity of energy E_d becomes comparable to the sound wavelength. As has been pointed out above, to check this one needs to compare in this frequency interval the main (resonant) contribution to the mean free path, given by Eq. (50), with the wavelength $2\pi v / \omega$. Above the frequency $\omega_d = E_d / \hbar$, the sound waves cease to exist as well defined propagating excitations. But harmonic excitations above this energy need not be overdamped. For example, for a-SiO$_2$, $E_d \simeq E_c$, and from Eq. (60) we have

$$\frac{\hbar}{\tau(E_d)} \simeq W \ll E_d . \qquad (83)$$

The situation with the infrared absorption may be somewhat different. The point is that in a number of cases of interest the interaction constant of infrared radiation with the soft vibrations can be substantially smaller than that for the hypersound of the same frequency ω. The interaction near the energy E_d can be characterized by a dimensionless parameter $\mathcal{H} \equiv \beta_d \Lambda_d$, where β_d and Λ_d are the absorption coefficient and wavelength of infrared radiation for frequency $\omega_d = E_d / \hbar$, respectively. One can present this parameter as the product βl given by Eq. (68) times the ratio of the velocities of light and sound:

$$\mathcal{H} = \beta l \frac{c}{v\sqrt{\varepsilon}} = \frac{4\pi}{3} \frac{\rho v^2}{\varepsilon} \left[\frac{d_0 \tilde{H}}{\mathcal{E}_0} \right]^2 = \frac{4\pi}{3} \frac{A}{\varepsilon C} . \qquad (84)$$

Here C is given by Eq. (49) and for dimensionless parameter A we have

$$A = \frac{\mathcal{P}_0 d_0^2 \eta_L^{7/2}}{W} . \qquad (85)$$

In the SPM this parameter determines the logarithmic temperature dependence of the dielectric constant of the glass at low temperatures (the so-called resonant contri-

bution of TLS's) (Ref. 23)

$$\left[\frac{\varepsilon(T) - \varepsilon(T_0)}{\varepsilon(T_0)} \right]_{\text{res,TSL}} = -\frac{8\pi}{3} \frac{A}{\varepsilon} \ln \frac{T}{T_0} . \qquad (86)$$

As for parameters A and C, they can be taken from the experimental data. According to these data we have for Suprasil W,[23] $A \approx 0.92 \times 10^{-5}$ and $C = 3.1 \times 10^{-4}$. For the dielectric constant we take the value $\varepsilon = 3.8$. In this way we obtain for the vitreous silica the value of the product given by Eq. (84) of the order of 3×10^{-2} (for the longitudinal sound).

This means that although the relation (68) is valid only for the frequencies much smaller than $\omega_d = E_d/\hbar$, the general equation (45) may be valid up to the frequencies well above ω_d provided $\mathcal{H}$ is much smaller than 1. We wish to emphasize that, at the same time, Eq. (45) relating Raman-scattering intensity to the coefficient of infrared absorption also remains valid although it is impossible to give explicit analytical expressions for both quantities.

VII. IS THERE ANY RELATION BETWEEN LIGHT SCATTERING AND SPECIFIC HEAT OF A GLASS?

So far we have considered the simplest version of the soft-potential model where ξ and η were the only fluctuating quantities. However, the soft potentials with the same values of ξ and η can, in general, differ by the values of other parameters, such as the effective masses M, or the modulated polarizabilities α. Let us discuss, with regard to this assumption, whether there exists any relation between the light-scattering intensity and the density of states of harmonic vibrations that determine the specific heat. Such a relation has been extensively discussed in the literature (see Refs. 49 and 37).

The extinction coefficient dh is proportional to the sum over all the oscillators, which we shall specify in this section by indices s, s', etc.,

$$\frac{1}{\mathcal{V}} \sum_s \mathcal{A}_s \langle x_s(t) x_s(0) \rangle_\omega , \qquad (87)$$

where $\mathcal{A}$ stands for the appropriate combination of $\alpha_{il}\alpha_{km}$, which we need not specify now.

A. Noninteracting oscillators

To begin with, we consider the intensity of light scattering by a system of noninteracting soft harmonic oscillators. According to Eq. (87) a single-oscillator contribution to the extinction coefficient is proportional to the Fourier component of the correlation function, $\langle x(t)x(0)\rangle_\omega$. Here x is the coordinate of a single soft harmonic oscillator noninteracting with other soft modes. For the correlation function one can write

$$\langle x(t)x(0)\rangle_\omega = 2\pi\hbar \sum_{nm} e^{(\mathcal{F} - E_n)/kT} |x_{nm}|^2 \delta(\hbar\omega + E_n - E_m) .$$

$$(88)$$

Here $\mathcal{F}$ is the free energy; m and n label the exact eigenstates of the whole Hamiltonian H_0, while E_m and E_n are its eigenvalues. The matrix elements are those of a single oscillator with the eigenfrequency $E/\hbar$:

$$x_{n,n-1} = x_{n-1,n} = i\hbar\sqrt{n/2ME} ,$$

$$x_{n,n+1} = x_{n+1,n} = i\hbar\sqrt{(n+1)/2ME} . \qquad (89)$$

For $\omega > 0$ (the Stokes component), only the term with $m = n + 1$ remains in the whole sum over m, and we are left with expression

$$\langle x(t)x(0)\rangle_\omega = \frac{\pi\hbar^2}{\omega M} [N(\omega) + 1]\delta(\hbar\omega - E) . \qquad (90)$$

After summation over all the harmonic oscillators we get

$$\frac{1}{\mathcal{V}} \sum_s \mathcal{A}_s \langle x_s(t)x_s(0)\rangle_\omega$$

$$= \frac{\pi\hbar^2}{\omega} [N(\omega) + 1] \frac{1}{\mathcal{V}} \sum_s \frac{\mathcal{A}_s}{M_s} \delta(\hbar\omega - E_s) . \qquad (91)$$

This equation can be rewritten as

$$\frac{1}{\mathcal{V}} \sum_s \mathcal{A}_s \langle x_s(t)x_s(0)\rangle_\omega = \frac{\pi\hbar^2}{\omega} [N(\omega) + 1]\mathcal{B}(\omega)n(\hbar\omega) ,$$

$$(92)$$

where $n(E)$ is the density of states of noninteracting harmonic oscillators,

$$n(E) = \frac{1}{\mathcal{V}} \sum_s \delta(\hbar\omega - E_s) . \qquad (93)$$

The same quantity determines also the specific heat:

$$\mathcal{C} = \int dE \frac{E^2}{kT^2} \frac{e^{E/kT}}{(e^{E/kT} - 1)^2} n(E) . \qquad (94)$$

Now, $\mathcal{B}(\omega)$ is defined as

$$\mathcal{B}(\omega) = \frac{(1/\mathcal{V}) \sum_s (\mathcal{A}_s/M_s)\delta(\hbar\omega - E_s)}{(1/\mathcal{V}) \sum_s \delta(\hbar\omega - E_s)} . \qquad (95)$$

In general, function $\mathcal{B}(\omega)$ can be frequency dependent. However, within the framework of the traditional soft-potential model exploited above, this function does not depend on the frequency. Indeed, according to this model, the fluctuations of the effective masses (and other parameters, such as $\mathcal{E}_0$ and $\mathcal{A}$) are independent of ξ and η and therefore $\mathcal{B} = \mathcal{A}/M$ is a constant. So, for this case one may expect a simple relation between the light scattering and the density of states (which determines the specific heat) of the form

$$n(\hbar\omega) \propto \frac{I(\omega, T)\omega}{N(\omega) + 1} . \qquad (96)$$

B. Interacting oscillators

Above the energy E_d interaction of different harmonic oscillators becomes important. One can show that above

E_d the retardation of interaction due to finite value of the sound velocity is of no importance. So one can write the Hamiltonian in the form

$$\sum_s \left[-\frac{\hbar^2}{2M_s}\frac{d^2}{dx_s^2} + \mathscr{E}_{0s}\eta_s \left[\frac{x_s}{a} \right]^2 \right] + \frac{1}{2}\sum_{s\neq s'} J_{ss'}x_s x_{s'} \ . \tag{97}$$

Let us introduce new coordinates y_s by a linear transformation which diagonalizes the total potential energy and

leaves the total kinetic energy invariant, so that as a result we get a set of noninteracting harmonic oscillators

$$x_s = \sum_{s'} \sqrt{M_{s'}/M_s}\, a_{ss'}y_{s'} \ , \tag{98}$$

where $a_{ss'}$ is an orthogonal transformation matrix that obeys the conditions

$$\sum_{ss'} a_{ss''}a_{s's''} = \sum_{s''} a_{s''s}a_{s''s'} = \delta_{ss'} \ . \tag{99}$$

Then instead of Eq. (88) we have

$$\langle x_s(t)x_s(0)\rangle_\omega = 2\pi\hbar\sum_{nm} e^{(\mathscr{F}-E_n)/kT}\left| \sum_{s'} \sqrt{M_{s'}/M_s}\, a_{ss'}(y_{s'})_{nm} \right|^2 \delta(\hbar\omega + E_n - E_m) \ . \tag{100}$$

As different harmonic oscillators defined in such a way are noninteracting the wave function of the system is a product of the wave functions of single oscillators. Therefore, we have for $s\neq s'$,

$$(y_s)_{nm}(y_{s'})_{nm} = 0$$

and from Eq. (100) we get

$$\langle x_s(t)x_s(0)\rangle_\omega = \frac{\pi\hbar^2}{\omega M_s}[N(\omega)+1]\sum_{s'} a_{ss'}^2 \delta(\hbar\omega - \widetilde{E}_{s'}) \ , \tag{101}$$

where $\widetilde{E}_s$ are the energies of the new harmonic modes.

After summation over all the "molecules" in the unite volume we obtain

$$\frac{1}{\mathscr{V}}\sum_s \mathscr{A}_s \langle x_s(t)x_s(0)\rangle_\omega$$

$$= \frac{\pi\hbar^2}{\omega}[N(\omega)+1]\frac{1}{\mathscr{V}}\sum_{s'} \left[\sum_s \frac{\mathscr{A}_s}{M_s}a_{ss'}^2 \right] \delta(\hbar\omega - \widetilde{E}_{s'}) \tag{102}$$

We can rewrite Eq. (102), in full analogy with Eq. (91), as

$$\frac{1}{\mathscr{V}}\sum_s \mathscr{A}_s \langle x_s(t)x_s(0)\rangle_\omega = \frac{\pi\hbar^2}{\omega}[N(\omega)+1]\widetilde{\mathscr{B}}(\omega)\widetilde{n}(\hbar\omega) \ , \tag{103}$$

where $\widetilde{n}(E)$ is the new (renormalized) density of states of the harmonic oscillators (that determines also the specific heat)

$$\widetilde{n}(E) = \frac{1}{\mathscr{V}}\sum_s \delta(E - \widetilde{E}_s) \ , \tag{104}$$

and $\widetilde{\mathscr{B}}(\omega)$ is given by

$$\widetilde{\mathscr{B}}(\omega) = \frac{(1/\mathscr{V})\sum_s \left[\sum_{s'} \frac{\mathscr{A}_{s'}}{M_{s'}}a_{s's}^2 \right]\delta(\hbar\omega - \widetilde{E}_s)}{(1/\mathscr{V})\sum_s \delta(\hbar\omega - \widetilde{E}_s)} \ . \tag{105}$$

We see that Eq. (103) still has the Bose factor $N(\omega)+1$ that determines the temperature dependence of the light-scattering intensity. However, $\widetilde{\mathscr{B}}$ could be, in general, a function of ω, in particular, due to the fact that the transformation matrix elements $a_{ss'}$ are dependent on the energy interval in consideration.

However, if for all the harmonic oscillators M_s and $\mathscr{A}_s$ are the same then due to relations (99)

$$\widetilde{\mathscr{B}}(\omega) = \mathscr{B}(\omega) = \mathscr{A}/M = \text{const} \ ,$$

i.e., the two constants are equal. To the contrary, if one allows for fluctuations of $\mathscr{A}_s$ and M_s we have

$$\widetilde{\mathscr{B}}(\omega) \neq \mathscr{B}(\omega) \ .$$

If, irrespective to such fluctuations $\widetilde{\mathscr{B}}$ remains constant, the value of the constant may still differ from $\mathscr{B}$. In such a case one should not expect a simple relation between the light scattering and the density of states of the form of Eq. (96) in the whole frequency region, although it may still hold below and above $E_d/\hbar$.

VIII. DISCUSSION OF EXPERIMENTAL DATA

In the classical region of frequencies and temperatures where

$$\hbar\omega \ll kT \ , \tag{106}$$

we have

$$N(\omega) \approx kT/\hbar\omega \tag{107}$$

and Eq. (47) takes the form

$$I(\omega, T) \propto T\frac{l^{-1}}{\omega^2} \propto T\frac{\beta(\omega)}{\omega^2} \ . \tag{108}$$

It is exactly this observation that was made by Stolen.[11] He indicated that for fused quartz β/ω^2 has a similar spectral shape as the Raman scattering for $\omega < 120$ cm^{-1}. On the basis of this observation he came to the conclusion that the far-infrared absorption of fused quartz arises from the same normal vibrations as the low-

frequency Raman scattering. For $\omega > 20$ cm^{-1} the absorption was found to be independent of temperature which is consistent with the modes behaving like harmonic oscillators. This is in agreement with our theory.

As for the hypersound attenuation from the experimental point of view, difficulties arise from the comparison between the hypersound attenuation $l^{-1}(\omega, T)$ and the Raman-scattering intensity $I(\omega, T)$. In the temperature range of interest, l^{-1} is only available from Brillouin-scattering experiments, i.e., at frequencies near to 1 cm^{-1} (30 GHz) in usual glasses. Due to the high intensity of the strictly elastic component and the finite width of the instrumental profile, the conventional Raman spectrometer gives information on inelastic scattered light above a few cm^{-1}. Recently, a new spectrometer has been able to extend the low-frequency range of measurements. The published data[50] concern ionic glasses but there are no available experimental data about hypersound absorption in these materials.

Following the first idea of Jäckle, three attempts have been made in the literature to compare l^{-1} and I in vitreous silica,[12] in borate glasses,[51] and in optical glass with metal ions (Schott LaSF7).[52] In the previous analysis comparison between l^{-1} and I arises essentially from the calculation of the hypersonic attenuation at higher frequencies than those at which the measurements have been made. These calculations use a model of relaxation defects with a distribution of barriers and of course do not account for the different contributions to l^{-1}. Moreover, the parameters of the model have been determined from the *ultrasonic* data and the Raman intensity under consideration is extracted from the light-scattering data where, apart from relaxation defects, other contributions were present.

In spite of large uncertainties in experimental determination, a satisfactory agreement has been observed for the T variation of l^{-1} and I, up to 300 K for SiO$_2$ and B$_2$O$_3$ glasses,[12,52] and these results demonstrate at least qualitatively the correspondence between l^{-1} and I.

As a result of the previous section, the hypersound absorption coefficient for $\omega \ll \omega_d$ is determined by the sum

$$l^{-1} = l^{-1}_{\text{rel,DWP}} + l^{-1}_{\text{res,HO}} + l^{-1}_{\text{res,HO}} \cdot \tag{109}$$

Because of its strong frequency dependence the third term in this sum [see Eq. (50)] dominates for sufficiently high frequencies ($\hbar\omega \gg W$), and we have

$$I(\omega, T) \propto \omega^3 [N(\omega) + 1] \cdot \tag{110}$$

This equation describes nothing else than *light scattering by quasilocal soft harmonic modes* whose density of states goes as E^4 [see Eq. (9)].

This dependence is observed in many experiments on Raman scattering in glasses and amorphous solids for the frequencies below the boson peak (see, for instance, Refs. 53, 54, 12, and 55). Earlier these data were interpreted in favor of the Martin-Brenig model,[28] which used a concept of light scattering due to the combined action of structural defects and phonons. As we see, the SPM arrives at the same results in regard to the temperature and frequency dependence of the Raman scattering (the numerical coefficient can be much bigger in the SPM).

However, the soft-potential model is capable to give a description of the Raman scattering in a wider frequency and temperature interval where the first two terms in Eq. (109) can compete with the third one. As they are temperature dependent, they are responsible for the "light-scattering excess" (LSE) that is usually observed in glasses at low frequencies.[56,12] Their frequency dependencies are much weaker than the one of the third term [see Eq. (49) and Eq. (67)]. Therefore, the reduced Raman intensity

$$I_R = I\omega^{-1}[N(\omega) + 1]^{-1} \propto l^{-1}(\omega, T)/\omega^2 \tag{111}$$

has a minimum. The position of this minimum is determined by the value of W and slightly depends on temperature. The characteristic energy W can be estimated from the position of the minimum $T_{\min}$ in the temperature dependence of the specific heat $C(T)/T^3$: $W \approx (2 - 2.5)kT_{\min}$.[21,22] For example, in a-SiO$_2$, where one can neglect the relaxational contribution of HO, Eq. (67),[57] at these frequencies we obtain $\hbar\omega_{\min} \approx 1.6W(kT/W)^{1/4}$.[27] For $T = 80$ K and $W/k = 4$ K the minimum should be located at $\omega/2\pi \approx 10$ cm^{-1}, which coincides with experiments[58,12] (see Fig. 2). The theory describes, without fitting parameters, the experiment at other temperatures, too (see Fig. 2).

For the frequencies of the order of, or bigger than ω_d, Eq. (109) is no longer valid. Yet harmonic vibrations are responsible for the Raman scattering in this frequency interval, too [see Eq. (103)]. However, an essential feature of these vibrations is that they are extended over a large number of soft vibration sites. Their density of states $\tilde{n}(E)$, Eq. (77), is a linear function of energy E. If $\tilde{B}(\omega)$ in Eq. (103) is a constant or only weakly depends on ω, we have from Eq. (103)

$$I(\omega, T) \propto N(\omega) + 1 \cdot \tag{112}$$

i.e., for $kT \gg \hbar\omega$ this is an descending function of ω. As a result, at $\omega \simeq \omega_d$ we have the famous boson peak.
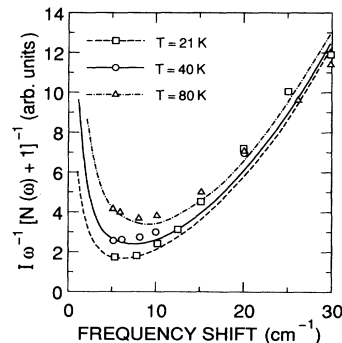

FIG. 2. Reduced Raman-scattering intensity (relative units) as a function of frequency for a-SiO$_2$ for different temperatures $T = 80$ K, $T = 40$ K, and $T = 21$ K. Symbols are experimental data (Refs. 58 and 12) and lines are theory. For all these plots the values of $W/k = 4$ K, and $\tau_0 = 10^{-12}$ s has been used. Two contributions were taken into account: One is the relaxation contribution from TLS's—Eq. (49) and the second is the resonant contribution from harmonic oscillators—Eq. (50).

The standard interpretation of the light scattering in a wide interval uses a combination of the Martin-Brenig model (for the high-frequency part) with the defect scattering model (for the low-frequency part). The role of defects is played by the two well potentials. This is a combination of two entirely different mechanisms. It is, however, very difficult to provide within the framework of this combined model an explanation of the fact that the depolarization ratios of the light-scattering excess and the first-order vibrational spectrum remained almost constant within the whole frequency interval (even above the boson peak).[12,54] The explanation of this fact emerges from our theory in a natural way [see Eq. (23)] as a consequence of the assumption that $\overline{\alpha_{il}\alpha_{km}}$ is independent of ξ and η (see the end of the Sec. III).

IX. CONCLUSION

We developed a theory of the low-frequency Raman scattering in glasses. For the inelastic light-scattering, soft atomic potentials, which determine a lot of universal properties of glasses, are responsible. The theory can explain in a uniform way the Raman scattering in a wide frequency and temperature interval. For the low-frequency part of the spectrum, which usually has more pronounced temperature dependence (light-scattering excess), the relaxation processes in double-well and one-well potentials are responsible. At higher frequencies the main contribution is due to the resonant processes from soft quasilocal harmonic modes.

Their density of states sharply increases with energy $\propto E^4$, so that at some energy of the order of a few meV the interaction between these oscillators becomes important. This interaction leads to reconstruction of the density of states of harmonic modes which become delocalized above this energy (being quasilocalized below this energy). The density of states of this new harmonic modes has a linear dependence on the energy. This is, according to our opinion, the reason for the famous boson peak which was discussed a lot in the literature and a lot of explanations of its origin was proposed. All these explanations, however, do not relate the boson peak, which is one of the universal properties of glasses, to other universal properties. Such relations appear in a natural way in our theory.

Exploiting the universality we can take relevant parameters of our theory from other low-temperature experiments in glasses. After that, without any additional

fitting parameters, a striking agreement between theory and experiment is achieved.

APPENDIX: FLUCTUATION-DISSIPATION THEOREM

To derive the FDT we start with Eq. (26) for a transport coefficient $b(\omega)$:

$$b(\omega) = \frac{i}{\hbar} \int_0^\infty dt\, e^{i\omega t} \langle [x(t), B] \rangle .$$ (A1)

This equation can be rewritten in the following form:

$$b(\omega) = \sum_{n,m} e^{(\mathcal{F}-E_n)/kT} x_{nm} B_{mn} \left[1 - \exp\left[\frac{E_n - E_m}{kT} \right] \right]$$

$$\times \int_0^\infty dt\, \exp[i\omega t + (i/h)(E_n - E_m)t - \delta t] .$$ (A2)

Here $\delta > 0, \delta \to 0$.

For the imaginary part of this function we get

$$b''(\omega) = \pi(1 - e^{-\hbar\omega/kT})$$

$$\times \sum_{n,m} e^{(\mathcal{F}-E_n)/kT} x_{nm} B_{mn} \delta(\hbar\omega + E_n - E_m) .$$ (A3)

For the Fourier component of the correlation function, $\langle x(t)B \rangle_\omega$, we get in the same manner

$$\langle x(t)B \rangle_\omega = 2\pi\hbar \sum_{nm} e^{(\mathcal{F}-E_n)/kT} x_{nm} B_{mn} \delta(\hbar\omega + E_n - E_m) .$$ (A4)

Comparing Eqs. (A3) and (A4) we arrive to FDT, Eq. (29).

[1]R. C. Zeller and R. O. Pohl, Phys. Rev. B 4, 2029 (1971).

[2]P. W. Anderson, B. I. Halperin, and C. M. Varma, Philos. Mag. 25, 1 (1972).

[3]W. A. Phillips, J. Low Temp. Phys. 7, 351 (1972).

[4]S. Hunklinger and W. Arnold, Physical Acoustics, edited by W. P. Mason and R. N. Thurston (Academic, New York, 1976), Vol. XII, p. 155.

[5]S. Hunklinger and A. K. Raychaudhuri, Progress in Low Temperature Physics, edited by D. F. Brewer (Elsevier, Amsterdam, 1986), Vol. IX, p. 267.

[6]W. A. Phillips, Rep. Prog. Phys. 50, 1657 (1987).

[7]Amorphous Solids. Low-Temperature Properties, edited by W. A. Phillips, Topics in Current Physics Vol. 24 (Springer-Verlag, Berlin, 1981).

[8]J. L. Black, Glassy Metals I, edited by H. J. Günterodt and H. Beck (Springer, Berlin, 1981), p. 245.

[9]J. J. Freeman and A. G. Anderson, Phys. Rev. B 34, 5684 (1986).

[10]G. Bellessa, Phys. Rev. Lett. **40**, 1456 (1978).

[11]R. H. Stolen, Phys. Chem. Glasses **11**, 83 (1970).

[12]J. Jäckle, in Ref. 7, p. 135.

[13]U. Buchenau, H. M. Zhou, N. Nücker, K. S. Gilroy, and W. A. Phillips, Phys. Rev. Lett. **60**, 1318 (1988).

[14]J. M. Grace and A. C. Anderson, Phys. Rev. B **40**, 1901 (1989).

[15]O. Brand and H. v. Löhneysen, Europhys. Lett. **16**, 455 (1991).

[16]H. R. Schober and Brian B. Laird, Phys. Rev. B **44**, 6746 (1991); Brian B. Laird and H. R. Schober, Phys. Rev. Lett. **66**, 636 (1991).

[17]H. R. Schober, C. Oligschleger, and B. B. Laird, J. Non-Cryst. Solids **156-158**, 965 (1993).

[18]C. Oligschleger and H. R. Schober, Physica A (to be published).

[19]V. G. Karpov, M. I. Klinger, and F. N. Ignatiev, Zh. Eksp. Teor. Fiz. **84**, 760 (1983) [Sov. Phys. JETP **57**, 439 (1983)].

[20]V. G. Karpov and D. A. Parshin, Zh. Eksp. Teor. Fiz. **38**, 536 (1983) [Sov. Phys. JETP Lett. **38**, 648 (1983)].

[21]M. A. Il'in, V. G. Karpov, and D. A. Parshin, Zh. Eksp. Teor. Fiz. **92**, 291 (1987) [Sov. Phys. JETP **65**, 165 (1987)].

[22]U. Buchenau, Yu. M. Galperin, V. L. Gurevich, and H. R. Schober, Phys. Rev. B **43**, 5039 (1991).

[23]D. A. Parshin (unpublished).

[24]U. Buchenau, Yu. M. Galperin, V. L. Gurevich, D. A. Parshin, M. A. Ramos, and H. R. Schober, Phys. Rev. B **46**, 2798 (1992).

[25]D. A. Parshin (unpublished).

[26]D. A. Parshin and S. Sahling, Phys. Rev. B **47**, 5677 (1993).

[27]D. A. Parshin, Phys. Scr. (to be published).

[28]A. J. Martin and W. Brenig, Phys. Status Solidi B **64**, 163 (1974).

[29]H. Kawamura, K. Fukumasu, and Y. Hamada, Solid State Commun. **43**, 229 (1982).

[30]F. Viras and T. A. King, J. Non-Cryst. Solids **119**, 65 (1990).

[31]V. K. Malinovsky and A. P. Sokolov, Solid State Commun. **57**, 757 (1986).

[32]S. R. Elliot, Europhys. Lett. **19**, 201 (1992).

[33]A. P. Sokolov, A. Kisliuk, M. Soltwisch, and D. Quitmann, Phys. Rev. Lett. **69**, 1540 (1992).

[34]S. Kojima, Phys. Rev. B **47**, 2924 (1993).

[35]V. Z. Gochiyaev and A. P. Sokolov, Fiz. Tverd. Tela (Len-

ingrad) **31**, 21 (1989) [Sov. Phys. Solid State **31**, 4 (1989)].

[36]V. Z. Gochiyaev, V. K. Malinovsky, V. N. Novikov, and A. P. Sokolow, Philos. Mag. **63**, 777 (1991).

[37]V. K. Malinovsky, V. N. Novikov, P. P. Parshin, A. P. Sokolov, and M. G. Zemlyanov, Europhys. Lett. **11**, 43 (1990).

[38]P. Benassi, P. Gallo, G. Ruocco, G. Signorelli, and V. Mazzacurati, Philos. Mag. **65**, 161 (1992).

[39]This is the condition for the asymmetry Δ in a double-well potential to be much smaller than the interlevel distances within one well.

[40]L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, New York, 1986).

[41]J. Jäckle, Z. Phys. **257**, 212 (1972).

[42]X. Liu, H. v. Löhneysen, and D. A. Parshin (unpublished).

[43]A. Einstein, Ann. Phys. **35**, 679 (1911).

[44]D. G. Cahill, S. K. Watson, and R. O. Pohl, Phys. Rev. B **46**, 6131 (1992).

[45]J. L. Black and B. I. Halperin, Phys. Rev. B **16**, 2879, 1977.

[46]V. L. Gurevich, *Transport in Phonon Systems* (North-Holland, Amsterdam, 1986).

[47]L. Gil, M. A. Ramos, A. Bringer, and U. Buchenau, Phys. Rev. Lett. **70**, 182 (1993).

[48]M. A. Ramos, L. Gil, A. Bringer, and U. Buchenau, Phys. Status Solidi A **135**, 477 (1993).

[49]N. Ahmad, K. W. Hutt, and W. A. Phillips, J. Phys. C **19**, 3765 (1986).

[50]P. A. M. Rodrigues, P. Benassi, and A. Fontana, J. Non-Cryst. Solids **143**, 274 (1992).

[51]J. Lorösch, M. Couzi, J. Pelous, R. Vacher, and A. Levasseur, J. Non-Cryst. Solids **69**, 1 (1984).

[52]J. F. Berret, J. Pelous, R. Vacher, A. K. Raychaudhuri, and M. Schmidt, J. Non-Cryst. Solids **87**, 70 (1986).

[53]J. S. Lannin, Solid State Commun. **12**, 947 (1973).

[54]R. J. Nemanich, Phys. Rev. B **16**, 1655 (1977).

[55]G. Carini, M. Frederico, A. Fontana, and G. A. Saunders, Phys. Rev. B **47**, 3005 (1993).

[56]D. Heiman, R. W. Hellwarth, and D. S. Hamilton, J. Non-Cryst. Solids **34**, 63 (1979).

[57]However, this contribution is responsible for the LSE in chalcogenide glasses, such as As_2S_3 where contribution from TLS's is nearly temperature independent.

[58]G. Winterling, Phys. Rev. B **12**, 2432 (1975).