PHYSICAL REVIEW B VOLUME 49, NUMBER 14 1 APRIL 1994-II

Interactions of soft atomic potentials and universality of low-temperature properties of glasses

D. A. Parshin*
Institut für Angewandte Physik der Universität Heidelberg, Albert-Überle-Strasse 3-5, 6900 Heidelberg, Germany

(Received 26 May 1992; revised manuscript received 23 March 1993)

It is shown that the interaction between soft atomic potentials in glasses leads to a density of the low-energy tunneling states, $\bar{P}$, which does not depend on the concentration of defects. The dimensionless parameter $\bar{P}\gamma^2/\rho v^2$ which characterizes the interaction between two-level systems is estimated. It appears to be small in accordance with available experimental data. The theory developed allows us to understand both the insensitivity of low-temperature properties of glasses to the concentration of the defects (e.g., created by neutron irradiation) and the relatively small interaction between two-level systems. Making use of only three macroscopic material parameters, namely the mass density ρ, the sound velocity v, and the average value of the atomic mass $\bar{M}$, nearly correct values are obtained for the two-level systems' density of states $\bar{P}$, for the deformation potential γ and for other parameters describing the low-temperature properties of glasses. All of them appeared to be nearly the same for glassy systems. Thereby, the explanation of universality of the low-temperature properties of glasses is given.

I. INTRODUCTION

Many low-temperature properties of glasses are determined by two-level systems (TLS's).[1] According to the tunneling model of Anderson, Halperin, Varma, and Phillips[2,3] (AHVP), a TLS consists of an atom or a small group of atoms moving in a double-well potential. The lowest excitation of such a system is a pair of levels split by the tunneling through the barrier. As is well known, the TLS's are responsible for most of the low-temperature properties of glasses, such as specific heat, thermal conductivity, sound absorption and variation of sound velocity with temperature, microwave absorption, phonon echo, etc.[4–7] However, despite the enormous success of the AHVP model, some important questions remain so far unsolved.

The most intriguing theoretical problem in this field is a very small sensitivity of the low-temperature properties of glasses to the density of new defects which can be created, e.g., by doping[8–10] or, more directly, by neutron (or electron) irradiation of the glass[11] (see also a very interesting paper[12] and references therein). Moreover, the low-temperature properties of glasses are surprisingly insensitive to most of the internal (e.g., impurities, heat treatment[13]) and external perturbations (see, e.g., the paper by Bartell and Hunklinger[14] where the influence of an external pressure was investigated). Several authors[15–20] came to the conclusion that this insensitivity is the consequence of interaction between the TLS's. Roughly speaking, the idea was based on the following (so-called mean-field approximation).

Each TLS produces deformation ϵ_i at a distance r_i,

$$\epsilon_i \simeq \frac{\gamma_i}{\rho v^2 r_i^3} , \tag{1.1}$$

where γ_i is the deformation potential of the ith TLS, ρ is the mass density of the glass, and v is the sound velocity. Due to the deformation decreases inversely proportional to r_i^3, the distribution function of deformations in a glass produced by all the TLS's is a Lorentzian with the width $\delta\epsilon$ proportional to the total concentration N of the TLS's:

$$\delta\epsilon \simeq \frac{\gamma N}{\rho v^2} . \tag{1.2}$$

In turn, the energy E_i (interlevel spacing) of each TLS changes under the deformation ϵ as

$$\delta E_i = \gamma_i \epsilon . \tag{1.3}$$

Taking (1.2) into account we get that the energies E_i of different TLS's are distributed in the interval δE:

$$\delta E \simeq \frac{\gamma^2 N}{\rho v^2} . \tag{1.4}$$

For small energies, the density of states $n(E)$ appears to be independent of the energy and of the concentration of TLS's:

$$n(E) \simeq \frac{N}{\delta E} \simeq \frac{\rho v^2}{\gamma^2} . \tag{1.5}$$

At first glance this approach is very attractive. Nevertheless, it is most probably not applicable to real glasses in this simple form. First, as follows from (1.5), the dimensionless parameter $n(E)\gamma^2/\rho v^2$ appears to be of the order of unity. Meanwhile, the experimental values of the parameter $\bar{P}\gamma^2/\rho v^2$ for different glasses are much smaller than unity. They are spread within the interval $(0.3-4.9)\times 10^{-4}$ (for transverse sound: $v = v_t$) (Ref. 21), i.e., though the width is ≈10 times greater than the average value, the value itself is clearly much smaller than unity.

Second, it is very difficult to imagine that the TLS's are the only defects in the glass structure which are able to create deformations. Moreover, it is very natural to suppose that the concentration of other defects (different from TLS's) N_{nt} is much larger than the concentration of

TLS's N_t. In any event it is the total concentration of defects $N = N_t + N_{nt}$ that should enter formulas (1.2) and (1.4). At the same time, to determine the TLS's density of states one only has to insert into (1.5) the concentration of TLS's, N_t. As a result

$$n(E) \simeq \frac{N_t}{\delta E} \simeq \frac{\rho v^2}{\gamma^2} \frac{N_t}{N} \ . \tag{1.6}$$

In such a case

$$\frac{n(E)\gamma^2}{\rho v^2} \simeq \frac{N_t}{N} \tag{1.7}$$

and it is possible to interpret the experiment if we assume the ratio N_t/N to be equal to the observable value (of the order 10^{-4}). Nevertheless, in this case one comes to the conclusion that the parameter $n(E)\gamma^2/\rho v^2$ decreases with the increase of the concentration of defects N. It also remains unclear why the ratio N_t/N for all the glasses is so small.

To avoid the first contradiction Yu and Leggett[18,19] put forward the hypothesis that the smallness of the parameter $\bar{P}\gamma^2/\rho v^2$ can be conditioned by the strong renormalization of the density of states due to off-diagonal interaction between the TLS's. The crossover energy E_c/k from their point of view is of the order of 10–40 K and coincides with the interaction energy of two TLS's, $\gamma^2/\rho v^2 r_c^3$, the distance r_c between them being of the order of phonon wavelength with the same energy:

$$E_c \simeq \frac{\gamma^2}{\rho v^2 r_c^3} \simeq \frac{\hbar v}{r_c} \simeq \frac{\sqrt{\rho \hbar^3 v^5}}{\gamma} \ ,$$

where

$$r_c \simeq \frac{\gamma}{\sqrt{\rho \hbar v^3}} \simeq \frac{\hbar v}{E_c} \ . \tag{1.8}$$

However, as pointed out in an earlier paper by Gurevich and Parshin[22] (where the characteristic energy E_c was first introduced in the course of analysis of the perturbation-theory series), for the symmetric TLS's with the energy E exceeding E_c the conventional definition of a TLS becomes meaningless, since the energy uncertainty $\hbar/\tau(E)$ due to interaction with phonons exceeds the interlevel spacing E:

$$\frac{\hbar}{\tau(E)} \simeq \frac{E^3}{E_c^2} > E \quad \text{for } E > E_c \ . \tag{1.9}$$

Here $\tau(E)$ is the TLS relaxation time due to one-phonon processes. This means that the perturbation theory should no longer be valid for the description of the TLS phonon interaction for $E > E_c$ and the usual concept of TLS's (Refs. 2 and 3) fails (for the same reason it is true for the local Einstein oscillators introduced in Ref. 23). In connection with this observation one wonders whether the whole picture presented by Yu and Leggett gives an adequate description of properties of glasses. Due to the finite value of the TLS's relaxation time the off-diagonal interaction of the two TLS's should decrease exponentially with distance r between them: $\propto \exp[-r/r(E)]$, where $r(E) = v\tau(E)$. For $E > E_c$, $r(E) < r_c$.

In the present paper we use a simpler approach (on the level of the mean-field approximation). It is free from these difficulties and contradictions and answers these questions within the framework of the soft model[24] (SPM) (see also the review in Ref. 25). We show that due to the softness of the local atomic potentials responsible for the TLS's in a glass structure, the density of states (and interaction between the TLS's) is always small enough. The TLS picture is valid only for the tunneling splittings Δ_0 smaller than some characteristic energy $W \ll E_c$. The energy W characterizes the distance to the third level in a multilevel system. It is the crossover energy between the two-level and multilevel description of low-energy excitations in glasses.

The excitations with energies $E \gg W$ mainly are not the TLS's but soft harmonic oscillators. The density of states of these oscillators (in contrast to the constant density of states of the TLS's) sharply increases with the rise of their energy E: $\propto (E/W)^4$. Thus, the concentration of TLS's N_t is much smaller than the concentration of all these harmonic oscillators N_{nt}. As a result of their interaction, the TLS's density of states is independent of the energy and of the concentration of various defects in the glass structure. It is determined by the normalized distribution function of the soft atomic potential parameters in the glass. Roughly, it means that $N_t \propto N$ and the coefficient of proportionality is small enough (of the order of 10^{-4}). The physical reason for this phenomenon is basically the smallness of the electron mass as compared to the effective mass of the tunneling entity. It is nearly the same reason which provides the smallness of the phonon damping due to anharmonic processes in the crystalline solids. It can be considered as an answer to the question "why do phonons go so far?"[19]

The paper is organized as follows. In Sec. II, we give a short introduction to the SPM. In Sec. III, we derive the distribution function of the SPM parameters. We show that due to interaction between soft atomic potentials this function does not depend on the concentration of defects in the glass structure. In Sec. IV, we consider different low-temperature properties of glasses which follows from the SPM. We show that with respect of these properties the SPM is equivalent to the AHVP model. In Sec. V, we compare our theory with experiment and extract the numerical values for relevant combinations of the parameters involved in theory for a-SiO$_2$. In Sec. VI, we show that due to the softness of the local atomic potentials responsible for the low-energy excitations in glasses the dimensionless parameter $\bar{P}\gamma^2/\rho v^2$ is always small enough. It does not depend on the concentration of defects in the glass structure. In Sec. VII, we relate the microscopic TLS's parameters: density of states $\bar{P}$, deformation potential γ, etc. with macroscopic parameters of the materials ρ, v, and $\bar{M}$. All of them appeared to be nearly the same for glassy systems. In Sec. VIII, we discuss the small difference between the SPM and the AHVP model for the time-dependent specific heat. Section IX is a conclusion where the main results of the paper are summarized.

II. SOFT POTENTIAL MODEL

According to the SPM,[24,25] the quasilocal low-frequency modes in glasses are described by the soft anharmonic oscillator Hamiltonian

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

where M is the effective mass of the moving entity,

$$V(x)=\mathcal{E}_0\left[\eta\left[\frac{x}{a}\right]^2+t\left[\frac{x}{a}\right]^3+\left[\frac{x}{a}\right]^4\right]\ . \tag{2.1}$$

Here x is the generalized coordinate of the soft mode having units of length, a is the characteristic length of the order of the interatomic spacing ($a\simeq 1$ Å), $\mathcal{E}_0$ is of the order of atomic energy $\mathcal{E}_a$, i.e., of the order of $\bar{M}v^2\simeq 10$ eV (see Appendix A), $\bar{M}$ being the average mass of atoms constituting the glass (for SiO_2 $\bar{M}=20$ a.u., $v_l=5.8\times10^5$ cm/s and $\bar{M}v_l^2=7$ eV). The values of the dimensionless parameters η and t are random due to fluctuations of the structural parameters of a glass. The soft potentials correspond to $|\eta|, |t|\ll 1$.

For $t/\sqrt{\eta_L}<\eta_L/|\eta|$, negative η, and $|\eta|>3\eta_L$ [see Figs. 1 and 2(a)–2(d)] the two lowest levels in the potential (2.1) form a TLS with the energy splitting E,

$$E=\sqrt{\Delta_0^2+\Delta^2}\ , \tag{2.2}$$

where tunneling splitting Δ_0 and asymmetry Δ are determined by

$$\Delta_0\approx W\exp\left[-\frac{\sqrt{2}}{3}\left[\frac{|\eta|}{\eta_L}\right]^{3/2}\right]\ , \tag{2.3}$$

$$\Delta=\frac{W}{\sqrt{2}}\frac{|t|}{\sqrt{\eta_L}}\left[\frac{|\eta|}{\eta_L}\right]^{3/2}\ . \tag{2.4}$$

Here η_L is an important small parameter of the model,

$$\eta_L=(\hbar^2/2Ma^2\mathcal{E}_0)^{1/3}\approx10^{-2}\ . \tag{2.5}$$

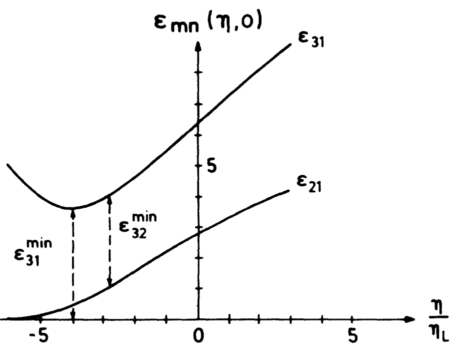

FIG. 1. The interlevel distance in the potential (2.1) (in the units of W) as a function of η/η_L for $t=0$. The arrows indicate the minimal distance between the levels $\varepsilon_{31}^{min}=3.65$, $\varepsilon_{32}^{min}=3$. These are the points of the absolute minimum of the functions $\varepsilon_{31}(\eta,t)$ and $\varepsilon_{32}(\eta,t)$ on the plane η,t. The corresponding density of state makes a finite jump (from zero) at this energy—a so-called van Hove singularity (Ref. 26).

For example, if we put in (2.5) $M=100$ a.u., $\mathcal{E}_0=7$ eV, $a=1$ Å, we get $\eta_L=1.4\times10^{-2}$. The energy W is determined by

$$W=\mathcal{E}_0\eta_L^2\approx k\times10\ \mathrm{K}\ , \tag{2.6}$$

where k is the Boltzmann's constant. This is the scale of characteristic energies in the potential (2.1) for $\eta=t=0$ [see Fig. 2(e)]. The barrier height V between two minima in the double-well potential (2.1) for $\Delta\ll V$ only depends on the value of $|\eta|$:

$$V=\frac{W}{4}\left[\frac{\eta}{\eta_L}\right]^2\ . \tag{2.7}$$

Just the existence of the small parameter η_L in the model makes it possible to neglect all the higher-order terms in expansion (2.1) because typical values of $x/a\ll1$ (see also Sec. VI). Therefore, all the soft atomic potentials in glasses can be described by this way. This remark means that the SPM is, in fact, a theory of quasilocal soft anharmonic excitations in glasses.

It is clear from Fig. 1 that the TLS picture is lost for sufficiently small $|\eta|\simeq\eta_L$ (and $kT\simeq W$) because the distance to the third level becomes comparable with the TLS energy E. For example, for $\eta=-\eta_L$ the distance between the first two levels in potential (2.1) equals $2.2W$, whereas the distance from the second to the third level equals $3.3W$, see Fig. 1. Therefore, the TLS's alone cannot be responsible for excitations with energies larger than W (with the exception of strongly asymmetric TLS's with high barriers: $V\gg W$, such TLS's are responsible for the relaxation absorption in glasses, but give a small contribution to the resonant absorption and thermal resistivity). For the excitations with larger energies in glasses single-well potentials are responsible too.

Besides the TLS's corresponding to double-well potentials with $\eta<0$, (2.1) also describes quasilocal anharmonic oscillators in single-well potentials with positive η and $\eta>9t^2/32$ [see Fig. 2(f)]. The excitations in the single-well potentials with $\eta\gg\eta_L$ are nearly harmonic, the interlevel spacing being

$$E=2W\sqrt{\eta/\eta_L}\ . \tag{2.8}$$

The density of states of these excitations (in contrast to the constant density of states of TLS's) sharply increases with the increasing energy E. At first, it makes finite jumps[26] (see also Refs. 27 and 25) at some energies of the order of W due to the presence of points of the absolute minimum of the functions $\varepsilon_{31}(\eta,t)$ (the energy distance between the first and the third levels in units of W) and $\varepsilon_{32}(\eta,t)$ (between the second and the third ones), etc., in the plane η,t at negative η (van Hove singularities[26]). For example, $\varepsilon_{31}^{min}=3.65$ and $\varepsilon_{32}^{min}=3$ (Fig. 1). Then, for $E\gg W$, it increases proportionally to $(E/W)^4$.[28,29] These excitations determine the properties of glasses at temperatures $kT\gg W$. We shall not consider them in this paper. It should be stressed however, that both types of excitations (in double-well potentials and single-well ones) are well-defined excitations with respect to their interaction with propagating phonons (see the end of Sec. IV).

Both the double-well potentials and the soft single-well ones are very important for our consideration because both are some "elastic defects" in the glass structure producing internal strain in the surrounding. Each elastic defect is characterized by a symmetric second-rank tensor $\hat{\Omega}$, where $\mathrm{Tr}(\hat{\Omega})$ is a variation of the glass volume due to the presence of the defect. The strain from defect as in the case of TLS's decreases inversely proportionally to the cube of distance from the defect

$$\epsilon_i \simeq \frac{\Omega_i}{r_i^3} \ . \tag{2.9}$$

In accordance with (1.1) one finds for TLS $\Omega_i \simeq \gamma_i/\rho v^2 \simeq 3.5$ Å^3 (we put $\gamma_i = 1.6$ eV, $\rho = 2.2$ g/cm^3, and $v = v_l = 5.8 \times 10^5$ cm/s for a-SiO$_2$).

III. DISTRIBUTION FUNCTION OF η AND t

To describe the distribution of the parameters η and t of the soft atomic potentials (2.1) in a glass we will use an idea similar to the one used in Ref. 28. Let us suppose that without interaction between the defects each of them is described by an anharmonic oscillator potential (2.1) with some generalized coordinate x', which we will call in the following "reference mode" and some reference coefficients $\eta^{(r)}$ and $t^{(r)}$. After the interaction is switched on, $x' = 0$ is no longer the "equilibrium" position for the defect and we have to add to the potential (2.1) the linear term proportional to x'. The coefficient of proportionality describes the external force which appears due to deformations produced by other defects. In principle, for the same reason the coefficients $\eta^{(r)}$ and $t^{(r)}$ should also

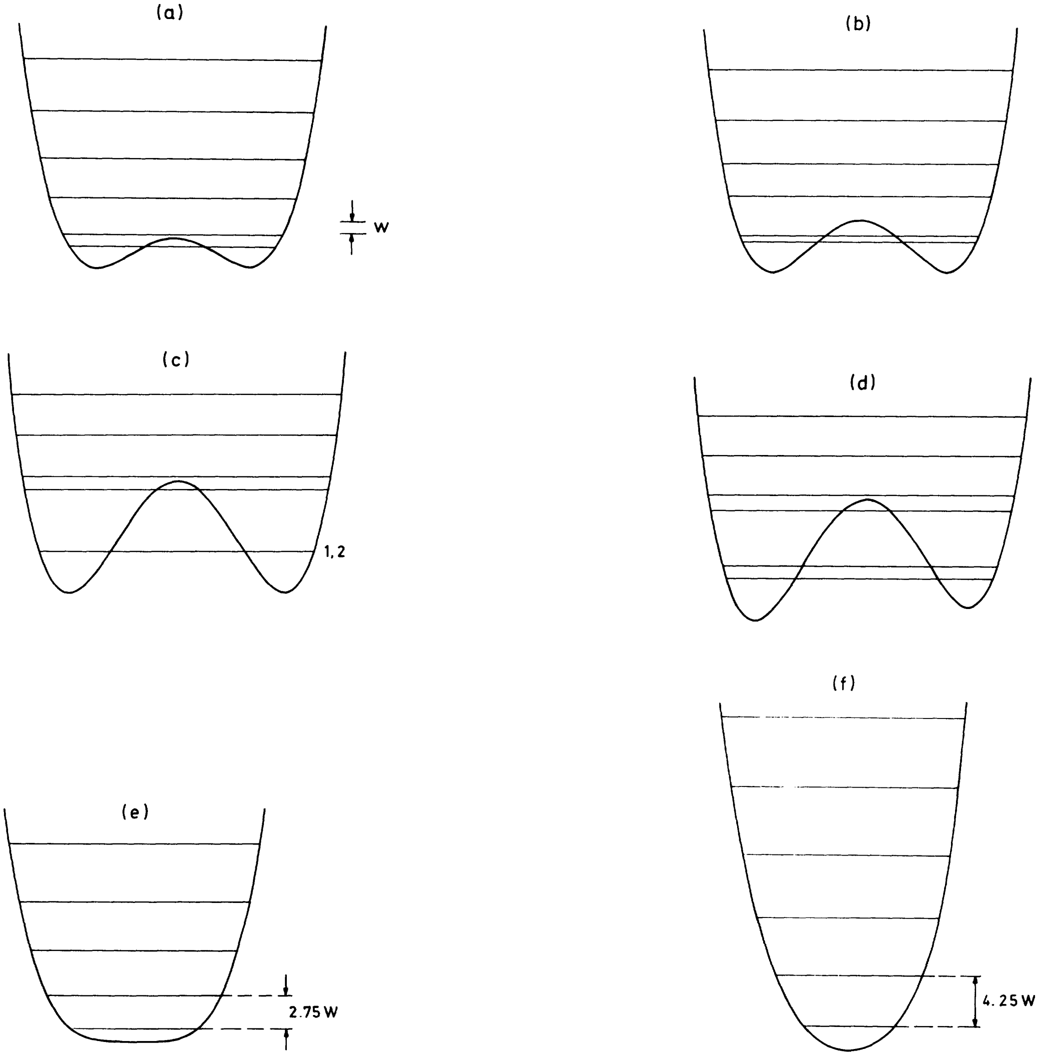

FIG. 2. The energy levels in the soft potential (2.1) for different values (a) η and t. (a) $\eta = -3\eta_L$, $t = 0$. (b) $\eta = -4\eta_L$, $t = 0$. (c) $\eta = -6\eta_L$, $t = 0$, $\varepsilon_{21} = 0.04$. (d) $\eta = -6\eta_L$, $t = 0.1\sqrt{\eta_L}$. (e) $\eta = t = 0$. (f) $\eta = 3\eta_L$, $t = 0$.

be changed. So, in the presence of other defects the potential energy of a defect $V'(x')$ for its reference mode x' is given by

$$V'(x')=\mathcal{E}_0\left[h'\left[\frac{x'}{a}\right]+\eta'\left[\frac{x'}{a}\right]^2+t'\left[\frac{x'}{a}\right]^3+\left[\frac{x'}{a}\right]^4\right] . \tag{3.1}$$

We can transform again (3.1) to (2.1) by shifting the origin of the reference system

$$x'=x+a\delta,\quad \delta=\frac{t-t'}{4} . \tag{3.2}$$

The coefficients h',η',t' are related to the coefficients η and t:

$$\eta'=\eta-\tfrac{3}{8}(t^2-t'^2) , \tag{3.3}$$

$$h'=\frac{t'-t}{2}\left[\eta-\frac{t^2}{4}+\frac{t'(t'+t)}{8}\right] . \tag{3.4}$$

The random quantities h', η', and t' are characterized by their distribution function $G(h',\eta',t')$. We are interested in the distribution function $P(\eta,t)$ of the parameters η and t. In Ref. 28 it was shown that

$$P(\eta,t)=\tfrac{1}{2}|\eta|P_0(\eta,t) , \tag{3.5}$$

where

$$P_0(\eta,t)=\int_{-\infty}^{\infty}G[h'(\eta,t,t'),\eta'(\eta,t,t'),t']dt' \tag{3.6}$$

and the functions $h'(\eta,t,t')$ and $\eta'(\eta,t,t')$ are determined by (3.4) and (3.3), respectively. The factor $|\eta|$ in (3.5) is very important and reflects the so-called "seagull" singularity in the distribution of the harmonic force constants η in the glass [i.e., $P(0,t)=0$].

The distribution function $G(h',\eta',t')$ should be normalized to the concentration N of the elastic defects in the glass structure

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}G(h',\eta',t')dh'd\eta'dt'=N . \tag{3.7}$$

Each of these defects creates a deformation in the surrounding in accordance with (2.9). For simplicity we suppose Ω_i to be the same for all of them, and equal to Ω_0 (where Ω_0 is of the order of atomic volume 1 Å^3, i.e., $\Omega_0\simeq a^3$). The resulting distribution function of deformations in a glass produced by all the defects is a Lorentzian with the width $\delta\epsilon$ proportional to their total concentration N [compare with (1.2)]:

$$\delta\epsilon\simeq\Omega_0 N . \tag{3.8}$$

Let us now discuss the important question of the form of the distribution function $G(h',\eta',t')$. We will assume that changes of the reference values $\eta^{(r)}$ and $t^{(r)}$ for the defect due to the interaction are small in comparison with the widths of their reference distributions $\delta_\eta^{(r)}$ and $\delta_t^{(r)}$ (see below). Thus, it is reasonable to treat the random quantities h', and η',t' as statistically independent. In this case the distribution function $G(h',\eta',t')$ factorizes into the distribution functions of the parameters h' and η',t':

$$G(h',\eta',t')=H(h')P^{(r)}(\eta',t') , \tag{3.9}$$

where $P^{(r)}(\eta,t)$ is the distribution function of parameters η and t when the interaction between defects is switched off.

The distribution function $\Psi(\eta)$,

$$\Psi(\eta)=\int_{-\infty}^{\infty}P^{(r)}(\eta,t)dt , \tag{3.10}$$

and $\Phi(t)$,

$$\Phi(t)=\int_{-\infty}^{\infty}P^{(r)}(\eta,t)d\eta , \tag{3.11}$$

is schematically depicted in Figs. 3(a) and 3(b), respectively. The distribution of $\Psi(\eta)$ is assumed to be centered near the value $\eta\approx 1$ corresponding to the normal (rigid) atomic potentials which are dominating. The distribution of $\Phi(t)$ is assumed to be even because of the absence of preferred directions in a glass. We will consider both of them to be normalized to unity:

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}P^{(r)}(\eta,t)d\eta dt=1 . \tag{3.12}$$

The central point of our analysis is that both of these

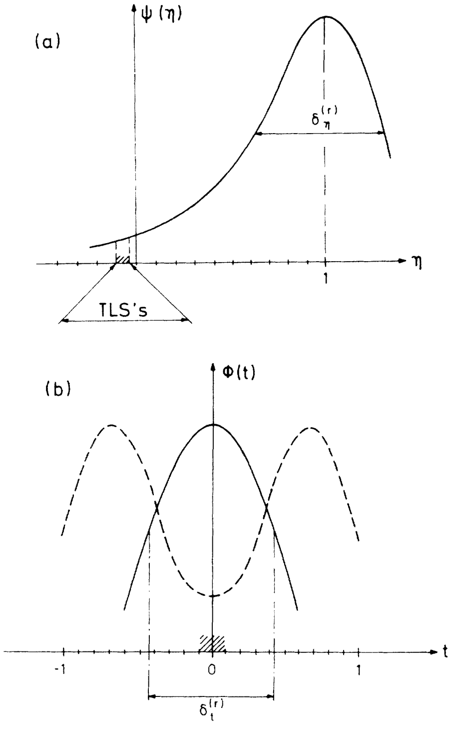

FIG. 3. The normalized distribution functions of parameters η and t in the absence of interaction between the defects ($\eta_L=10^{-2}$). (a) $\Psi(\eta)$. $\delta_\eta^{(r)}$ is the width of the distribution. The shaded region corresponds to TLS's with tunneling splitting Δ_0 from $3\times10^{-5}W$ (left) to W (right). (b) $\Phi(t)$ [the alternative possibility for the form of $\Phi(t)$ is depicted by the dashed line]. $\delta_t^{(r)}$ is the width of the distribution. The shaded region corresponds to double-well potentials with asymmetries $\Delta<(W/\sqrt{2})(\eta/\eta_L)^{3/2}$ [see (2.4)].

distributions have no relations with the distribution of random deformations in the glass structure created by elastic defects, but that they depend on disorder in the nearby environment of the soft atomic potential. It is easily seen when soft atomic potentials for example are created within the topological defects of the continuous network of the glass.[30] We define the reference mode x' in this case in such a way that potential (3.1) has no linear term at all for an isolated topological defect in the crystalline environment.

As for the distribution function $H(h')$ we will consider it to be determined by definition exclusively by the distribution of strain fields in the glass structure due to elastic defects. Our choice is stipulated by more pronounced sensitivity of h' to the strain that those of the parameters η' and t'.

To understand this, let us transform expression (3.1) to new dimensionless variables $y'=x'/a\sqrt{\eta_L}$, $\tilde{h}'=h'/\eta_L^{3/2}$, $\tilde{\eta}'=\eta/\eta_L$, and $\tilde{t}'=t'/\sqrt{\eta_L}$. Taking into account (2.6) we get

$$\tilde{V}'(y')\equiv\frac{V'(x')}{W}=\tilde{h}'y'+\tilde{\eta}'y'^2+\tilde{t}'y'^3+y'^4 \ . \tag{3.13}$$

The parameters $\tilde{h}'$, $\tilde{\eta}'$, and $\tilde{t}'$ determine the spectrum in the potential (3.1) in the units of W. Hence, it appears that the parameters h', η', and t' scale as $\eta_L^{3/2}\approx 10^{-3}$, $\eta_L\approx 10^{-2}$, and $\sqrt{\eta_L}\approx 10^{-1}$, respectively. These scales differ from each other by an order of magnitude.

On the other hand, the coefficients h', η', and t' are modified by the strain ϵ in the following way:

$$h'\rightarrow h'+H'\epsilon \ , \tag{3.14a}$$

$$\eta'\rightarrow\eta'+\Xi'\epsilon \ , \tag{3.14b}$$

$$t'\rightarrow t'+T'\epsilon \ , \tag{3.14c}$$

where the coefficients H', Ξ', and T' are of the order of unity. It follows, e.g., that for strain $\epsilon\simeq 10^{-3}$ the changes of $\tilde{h}'$, $\tilde{\eta}'$, and $\tilde{t}'$ are of the order of 1, 10^{-1}, and 10^{-2}, respectively, i.e., the main perturbation of the position of the quantum levels in the potential (3.1) is due to the linear term.

It is clear from (3.14b) and (3.14c) that the deformations from elastic defects ϵ_d do not affect the distributions of parameters η' and t' if the widths of their reference distributions $\delta_\eta^{(r)}$, $\delta_t^{(r)}$ (connected with the nearby environment) are larger than ϵ_d, i.e., the concentration of defects N is small enough [if $\delta_\eta^{(r)},\delta_t^{(r)}\simeq 1$, see Figs. 3(a) and 3(b), it means that the concentration of defects should be smaller than the concentration of atoms constituting the glass].

Since the distribution of the parameter h' in the glass is determined by the distribution of the deformations only, it has a Lorentzian shape:

$$H(h')=H_0\frac{\delta_{h'}^2}{h'^2+\delta_{h'}^2} \ , \tag{3.15}$$

where the width $\delta_{h'}$ is proportional to the concentration of defects N

$$\delta_{h'}\simeq H'\Omega_0 N \ . \tag{3.16}$$

The coefficient H_0 in (3.15) is determined by the normalizing condition (3.7) and does not depend on the concentration of elastic defects N:

$$H_0=\frac{N}{\pi\delta_{h'}}\simeq\frac{1}{\pi H'\Omega_0} \ . \tag{3.17}$$

This is the main result of our paper. As we show below, the value H_0 determines the TLS's density of states at low energies which appears to be independent of the concentration of elastic defects N.

In the end of this section we derive the distribution function $P_0(\eta,t)$ using expression (3.6) for a particular case, when the distribution on t' is not very widely spread around zero. If the width of the distribution $\delta_t\approx\delta_t^{(r)}$ satisfies the condition

$$\delta_{t'}\ll\min\left\{\frac{2\delta_{h'}}{\eta_L|\eta/\eta_L|},2\sqrt{2\eta_L/3}\sqrt{|\eta/\eta_L|}\right\} \tag{3.18}$$

($|\eta|/\eta_L$ is not very large) and the width $\delta_{h'}$ is not very small,

$$\delta_{h'}\gg\frac{1}{\sqrt{2}}\frac{\Delta}{W}\eta_L^{3/2}\left|\frac{\eta}{\eta_L}\right|^{-1/2} , \tag{3.19}$$

then in (3.15) $h'\ll\delta_{h'}$ and function $H(h')=H_0$; i.e., it is a constant. This takes place, e.g., if $\delta_{t'}\ll 0.3$ ($\delta_{h'}\approx 10^{-2}$, $|\eta|/\eta_L=6$, and $\eta_L=10^{-2}$). For the same conditions (3.18) η' from (3.3) coincides with η. Using (3.10) we derive from (3.6) and (3.9)

$$P_0(\eta,t)=H_0\Psi(\eta) \ . \tag{3.20}$$

So, the function $P_0(\eta,t)$ does not depend on t and N. It is determined by the normalized distribution function $\Psi(\eta)$ only.

The soft potentials we are interested in correspond to the "tail" of $\Psi(\eta)$ in the region of small η. If the scale of variation of $\Psi(\eta)$, $\delta_\eta\approx\delta_\eta^{(r)}\gg|\eta|$ [see Fig. 3(a)] we can consider the function $\Psi(\eta)$ (for relevant TLS's) to be a constant and equal to $\Psi(0)$. In this case the function $P_0(\eta,t)$ is also constant:

$$P_0(\eta,t)=\mathcal{P}_0\equiv H_0\Psi(0)\simeq\frac{\Psi(0)}{\pi H'\Omega_0} \ , \tag{3.21}$$

which, in particular, does not depend on the defect concentration N.

Because the typical width of the distribution $\delta_{t'}$ by definition cannot exceed unity (otherwise we should change $\mathcal{E}_0$) one can consider the treated case as sufficiently general. It is only important that typical values $h'<\delta_{h'}$. For instance, it is sufficient to have $\delta_{t'}<2(2\delta_{h'})^{1/3}$ and not very big values η/η_L. For $\delta_{h'}=0.06$ (it corresponds to concentration of defects of the order of 6%) it should be $\delta_{t'}<1$. Therefore, for sufficiently large concentration of defects the value $\mathcal{P}_0$ does not depend on N independently of the width $\delta_{t'}$.

IV. LOW-TEMPERATURE PROPERTIES OF GLASSES, $T \ll W/k$

In this section we consider the predictions of the SPM concerning the low-temperature properties of glasses ($T \ll W/k$). We show that with respect of these properties the SPM is equivalent to the AHVP model. Due to the existence of the absolute minimum distance between the second and the third levels in the potential (2.1) (see Fig. 1), all the low-temperature properties of glass for $T \ll W/k$ in the SPM are determined by TLS's only.

To calculate the low-temperature properties of glasses we will use instead of the variables η and t those of the AHVP model,

$$E \quad \text{and} \; p = \left[\frac{\Delta_0}{E} \right]^2 . \tag{4.1}$$

The parameters η and t are related to E and p [see (2.3) and (2.4)] by

$$|\eta| = \left[\frac{3}{\sqrt{2}} \right]^{2/3} \eta_L L^{2/3} , \tag{4.2}$$

$$|t| = \frac{2}{3}\sqrt{\eta_L}\,\frac{E\sqrt{1-p}}{WL} , \tag{4.3}$$

where

$$L = \ln \frac{W}{E\sqrt{p}} . \tag{4.4}$$

The Jacobian of the transformation is equal to

$$J(E,p) = \left| \frac{\partial(\eta,t)}{\partial(E,p)} \right| = \left[\frac{2}{9} \right]^{2/3} \frac{\eta_L^{3/2}}{WL^{4/3}} \frac{1}{p\sqrt{1-p}} . \tag{4.5}$$

As we have shown in Sec. III, the distribution function (3.5) for sufficiently general case is given by

$$P(\eta,t) = \left| \frac{\eta}{2} \right| \mathcal{P}_0 , \tag{4.6}$$

where $\mathcal{P}_0$ is constant to be determined by (3.21). Now, taking into account that $P(\eta,t)$ is an even function of t, we obtain from (4.2), (4.5), and (4.6) the following expression for the distribution function $F(E,p)$ in variables E and p:

$$F(E,p) = \left[\frac{2}{9} \right]^{1/3} \frac{\mathcal{P}_0 \eta_L^{5/2}}{W} \frac{1}{p\sqrt{1-p}} \frac{1}{L^{2/3}} , \tag{4.7}$$

which is practically independent of the energy E of TLS.

Now, we can calculate various low-temperature properties of glasses. Let us begin with the specific heat.

A. Specific heat

The contribution of TLS's to the specific is determined by

$$C(T) = k \int_0^{\infty} dE \int_{p_{\min}}^{1} dp\, F(E,p) \left[\frac{E}{kT} \right]^2 \frac{e^{E/kT}}{(e^{E/kT}+1)^2} . \tag{4.8}$$

As in the AHVP model $p_{\min}$ depends on the time of experiment t_{expt},[2,31]

$$p_{\min} = \frac{\tau_{\min}(T)}{t_{\text{expt}}} , \tag{4.9}$$

where $\tau_{\min}(T)$ is the relaxation time of the TLS with $\Delta = 0$ and $E = \Delta_0 \simeq kT$ [see (4.21)]. Direct integration of (4.8) with (4.7) leads to the result

$$C(T) = \pi^2 \left[\frac{2}{9} \right]^{1/3} \frac{\mathcal{P}_0 \eta_L^{5/2}}{W} k^2 T \ln^{1/3} \frac{W}{kT\sqrt{p_{\min}}} , \tag{4.10}$$

i.e., the specific heat has an almost linear temperature dependence and depends only weakly on the time of experiment: $C(T) \sim \ln^{1/3}(t_{\text{expt}})$ [instead of $C(T) \sim \ln(t_{\text{expt}})$ in the AHVP model].

B. Relaxation ultrasonic absorption in glasses: The deformation potential of a TLS

Relaxation ultrasonic absorption in glasses due to TLS's is determined as usual:[32,4]

$$l_{\text{rel}}^{-1} = \frac{1}{\mathcal{V}} \sum_{\text{TLS}} \frac{D^2}{\rho v^2} \frac{1}{4kT\cosh^2(E/2kT)} \frac{\omega^2 \tau}{1+\omega^2\tau^2} , \tag{4.11}$$

where $\mathcal{V}$ is a volume of the glass, ω is the ultrasound frequency, and τ is the TLS relaxation time:

$$\frac{1}{\tau} = \frac{M^2 E^3}{2\pi\rho\hbar^4 v^5} \coth \frac{E}{2kT} . \tag{4.12}$$

The deformation potentials M and D of the TLS are expressed through the deformation potential γ as

$$D = 2\frac{\Delta}{E}\gamma = 2\sqrt{1-p}\,\gamma, \quad M = \frac{\Delta_0}{E}\gamma = \sqrt{p}\,\gamma , \tag{4.13}$$

where γ describes variation of the asymmetry Δ under strain ϵ:

$$\gamma = \frac{1}{2}\frac{\partial \Delta}{\partial \epsilon} . \tag{4.14}$$

In order to determine l_{rel}^{-1} one needs to know the deformation potential γ. We can derive it within the framework of the approach developed above. The derivation and results are different from the ones in Refs. 27 and 25. (In these papers the interaction of TLS's with deformation ϵ of a different nature is considered. It gives a much smaller value of the deformation potential γ, see below.)

The coefficients h', η', and t' in (3.1) are changed under the deformation ϵ in accordance with (3.14a)–(3.14c). Due to the reasons mentioned above in Sec. III, the main contribution to the deformation potential γ comes from the linear term h' (3.14a). The variation of h' causes the coefficient t in (2.1) to change as well,

$$t \rightarrow t - \frac{2H'}{\eta}\epsilon . \tag{4.15}$$

Making use of (2.4) and (4.15) one obtains the deformation potential γ as a function of η:

$$\gamma=\frac{1}{2}\frac{\partial\Delta}{\partial\epsilon}=\frac{1}{2}\frac{\partial\Delta}{\partial t}\frac{\partial t}{\partial\epsilon}\,\mathrm{sgn}t$$

$$=\frac{1}{\sqrt{2}}\frac{H'W}{\eta_L^{3/2}}\sqrt{|\eta/\eta_L|}\,\mathrm{sgn}t\ , \qquad (4.16)$$

or in terms of variables E and p [see (4.2)]:

$$|\gamma|=\left[\frac{3}{4}\right]^{1/3}\frac{H'W}{\eta_L^{3/2}}L^{1/3}\ . \qquad (4.17)$$

As it follows from (4.16), γ is of the order of $H'W/\eta_L^{3/2}$ and slightly depends on the barrier height V [see (2.7)]: $\gamma \sim V^{1/4}$ [or logarithmically depends on the tunneling splitting Δ_0, see (2.3)]. Inserting $H' \simeq 1$, $W/k \simeq 10$ K, and $\eta_L \simeq 10^{-2}$ we obtain $\gamma \simeq 1$ eV. Thus, in the framework of our approach γ has the usual value of the order of 1 eV.[33] This value is by the factor $1/\sqrt{\eta_L} \simeq 10$ bigger than the one obtained in Refs. 25 and 27 (where it was of the order of $\gamma \simeq W/\eta_L$).

Now we can carry out our calculations. Replacing the summation in (4.11) by the integration with the distribution function $F(E,p)$,

$$\frac{1}{\mathcal{V}}\sum_{\mathrm{TLS}}\cdots=\int_{-\infty}^{+\infty}dt\int_{-\infty}^{0}d\eta\,P(\eta,t)\cdots$$

$$=\int_0^{\infty}dE\int_0^{1}dpF(E,p)\cdots\ , \qquad (4.18)$$

we obtain

$$l_{\mathrm{rel}}^{-1}=\frac{1}{2}\frac{\mathcal{P}_0H'^2W}{\rho v^3\sqrt{\eta_L}}\frac{\omega}{kT}\int_0^{\infty}\frac{dE}{\cosh^2(E/2kT)}\int_0^1 dp\frac{\sqrt{1-p}}{p}\frac{\omega\tau}{1+\omega^2\tau^2}\ , \qquad (4.19)$$

where the relaxation time is given as

$$\frac{1}{\tau}=\frac{1}{\tau_{\min}(T)}p\left[\frac{E}{2kT}\right]^3 L^{2/3}\coth\frac{E}{2kT} \qquad (4.20)$$

and

$$\frac{1}{\tau_{\min}(T)}=\frac{(36)^{1/3}}{\pi}\frac{H'^2W^2}{\rho\hbar^4v^5\eta_L^3}(kT)^3\ . \qquad (4.21)$$

In the following (as in the usual AHVP model) we analyze (4.19) for the limiting cases of "high" and "low" temperatures.

1. "High" temperatures: "Plateau region"

$$\omega\tau_{\min}(T)\ll 1\ . \qquad (4.22)$$

In this case we get from (4.19) the usual result for the absorption coefficient to be independent of the temperature ("plateau region"):

$$l_{\mathrm{rel}}^{-1}=\frac{\pi}{2}\frac{\mathcal{P}_0H'^2W}{\rho v^3\sqrt{\eta_L}}\omega\ . \qquad (4.23)$$

It depends linearly on the ultrasound frequency ω.

2. "Low" temperatures: "T^3 region"

$$\omega\tau_{\min}(T)\gg 1\ . \qquad (4.24)$$

In this case we derive from (4.19)

$$l_{\mathrm{rel}}^{-1}=\frac{\pi^3(36)^{1/3}}{96}\frac{\mathcal{P}_0H'^4W^3}{\rho^2\hbar^4v^8\eta_L^{7/2}}\ln^{2/3}\left[\frac{W}{2kT}\right](kT)^3\ , \qquad (4.25)$$

i.e., the absorption in this region is proportional to $T^3\ln^{2/3}(W/2kT)$ and independent of frequency. That result differs only slightly from the result of the AHVP model by the additional weak logarithmical dependence on temperature due to the factor $\ln^{2/3}(W/2kT)$.

C. TLS's contribution to the sound velocity

As is well known, TLS's also modify the sound velocity of glasses and determine its temperature dependence. Relative contribution of TLS's to the sound velocity $\delta v/v$ is given by the usual expression[4]

$$\frac{\delta v}{v}=-\frac{1}{2\rho v^2}\frac{1}{\mathcal{V}}\sum_{\mathrm{TLS}}\left[2\frac{M^2}{E}\tanh\frac{E}{2kT}\right.$$

$$\left.+\frac{D^2}{4kT\cosh^2(E/2kT)}\frac{1}{1+\omega^2\tau^2}\right]\ . \qquad (4.26)$$

The first and the second terms in the large parentheses in (4.26) are the so-called resonant and relaxation contributions, respectively.

In the temperature region (4.24) the relaxation contribution is small enough. On the contrary, in the plateau region it is important. Therefore, we will analyze it only in the limiting case (4.22). Performing the integration in (4.26) we derive the relaxation contribution to the sound velocity in the plateau region:

$$\left[\frac{\delta v}{v}\right]_{\mathrm{rel}}=-\frac{1}{2}\frac{\mathcal{P}_0H'^2W}{\rho v^2\sqrt{\eta_L}}\ln\frac{L_0^{2/3}}{\omega\tau_{\min}(T)}\ , \qquad (4.27)$$

where L_0 is determined by

$$L_0=\ln\left[\frac{WL_0^{1/3}}{2kT\sqrt{\omega\tau_{\min}(T)}}\right] \qquad (4.28)$$

(in fact this is the equation for L_0). Ignoring the weak logarithmical temperature dependence of $L_0^{1/3}$ we obtain what is usually measured in experiments,

$$\left[\frac{v(T)-v(T_0)}{v(T_0)}\right]_{\mathrm{rel}}=-\frac{3}{2}\frac{\mathcal{P}_0H'^2W}{\rho v^2\sqrt{\eta_L}}\ln\frac{T}{T_0}\ , \qquad (4.29)$$

i.e., the logarithmical temperature dependence of the sound velocity (as in the AHVP model).

In contrast to the relaxation contribution to the sound velocity, the resonant one [described by the first term in the large parentheses in (4.26)] is always important for both temperature regions (4.22) and (4.24). Performing the integration we derive

$$\left[\frac{v(T)-v(T_0)}{v(T_0)}\right]_{\rm res}=\frac{\mathcal{P}_0 H'^2 W}{\rho v^2\sqrt{\eta_L}}\ln\frac{T}{T_0}\ , \tag{4.30}$$

which also coincides with the prediction of the AHVP model.

D. Resonant absorption by TLS's

To complete our comparison with the AHVP model let us now calculate the resonant contribution of TLS's to the sound absorption. It is determined by the usual expression[4]

$$l_{\rm res}^{-1}=\frac{1}{\mathcal{V}}\frac{\pi\omega}{\rho v^3}\sum_{\rm TLS} M^2\tanh\frac{E}{2kT}\delta(\hbar\omega-E)\ . \tag{4.31}$$

From (4.31) we obtain the following result:

$$l_{\rm res}^{-1}=\pi\frac{\mathcal{P}_0 H'^2 W}{\rho v^3\sqrt{\eta_L}}\omega\tanh\frac{\hbar\omega}{2kT}\ . \tag{4.32}$$

The frequency and temperature dependencies are identical with the result of the AHVP model.

E. Interaction of TLS's with an electric field

In the AHVP model the interaction of a TLS with an electric field **F** is usually described by the Hamiltonian[34]

$$\mathcal{H}_F=\frac{1}{E}\begin{bmatrix}\Delta & -\Delta_0\\ -\Delta_0 & -\Delta\end{bmatrix}\mathbf{mF}\ . \tag{4.33}$$

The dipole moment **m**,

$$\mathbf{m}=\frac{1}{2}\frac{\partial\Delta}{\partial\mathbf{F}}\ , \tag{4.34}$$

characterizes the variation of TLS asymmetry Δ in the electric field.

The interaction of a soft atomic potential (2.1) with an electric field **F** can be described by adding to the potential $V'(x')$ (3.1) an extra term

$$-q^*\mathbf{Fn}x'\ , \tag{4.35}$$

where q^* is the effective charge of the moving entity and **n** is a unit vector in the direction of the motion. The additional term (4.35) in (3.1) means that the value of h' is changed in the electric field **F** [compare with (3.14a)]

$$h'\rightarrow h'-\frac{\mathbf{Fd}_0}{\mathcal{E}_0}\ , \tag{4.36}$$

where

$$\mathbf{d}_0=q^*a\mathbf{n}\ . \tag{4.37}$$

Proceeding as from (4.14) to (4.16) and (4.17), we obtain the following expression for the dipole moment **m** of the TLS (4.34):

$$\begin{aligned}\mathbf{m}&=\frac{1}{\sqrt{2}}\eta_L\mathbf{d}_0\sqrt{|\eta/\eta_L|}\,\mathrm{sgn}t\\ &=(\tfrac{3}{4})^{1/3}\sqrt{\eta_L}\mathbf{d}_0L^{1/3}\,\mathrm{sgn}t\ .\end{aligned} \tag{4.38}$$

The effective dipole moment m of TLS appears to be of the order of $\sqrt{\eta_L}\mathbf{d}_0$, i.e., of the order of $0.1\cdot e\cdot a\simeq 0.5D$ (if $q^*\simeq e$, where e is an electron charge). This value agrees well with experimental data.[35,36]

Proceeding by a analogy with calculations of acoustic properties of glasses we can derive now [using (4.38)] the TLS's contribution to dielectric properties. For example, the temperature dependence of dielectric constant $\epsilon(T)$ (resonant contribution) is described by [compare with (4.30)]

$$\left[\frac{\epsilon(T)-\epsilon(T_0)}{\epsilon(T_0)}\right]_{\rm res}=-\frac{8\pi}{3}\frac{\mathcal{P}_0 d_0^2\eta_L^{7/2}}{\epsilon W}\ln\frac{T}{T_0}\ . \tag{4.39}$$

Relaxation contribution differs from (4.39) by a factor $-(\frac{3}{2})$ [compare with (4.29) and (4.30)].

Dielectric losses are determined by

$$\tan(\delta)_{\rm res}=\frac{4\pi^2}{3}\frac{\mathcal{P}_0 d_0^2\eta_L^{7/2}}{\epsilon W}\tanh\frac{\hbar\omega}{2kT} \tag{4.40}$$

for the resonant contribution [compare with (4.32)], and

$$\tan(\delta)_{\rm rel}=\frac{2\pi^2}{3}\frac{\mathcal{P}_0 d_0^2\eta_L^{7/2}}{\epsilon W} \tag{4.41}$$

for the relaxation one in the plateau region (4.22) [compare with (4.23)].

F. The SPM is internally consistent

At the end of this section let us show that the low-energy excitations described by the SPM are always well-defined excitations, i.e., for them $\hbar/\tau(E)E\ll 1$ [where $\tau(E)$ is their relaxation time (4.12)]. For TLS's, we should show that the energy E_c (1.8) is larger than W [see (1.9)] because the TLS's picture in the SPM works up to the energies of the order of W only. Using for γ the value of the order $W/\eta_L^{3/2}$ (4.17) ($H'\simeq 1$), (2.5), (2.6), and inserting $\overline{M}v^2\simeq\rho v^2a^3\simeq\mathcal{E}_0$ we get

$$\frac{W}{E_c}\simeq\left[\frac{\overline{M}}{M}\right]^{3/4}\eta_L^{1/4}\ll 1 \tag{4.42}$$

[the effective mass M of the tunneling entity is most likely to be heavier than the average mass of the atoms constituting the glass $\overline{M}$ (Refs. 29 and 37)]. Therefore, in the SPM the TLS's are always well-defined excitations.

Considering excitations with higher energies, $E>W$, we should take into account higher levels in the potential (2.1) and use (instead of the TLS's picture) the picture of the soft quasilocal harmonic oscillators. By the same procedure it can be shown that they are also well-defined excitations with respect to their interaction with propagating phonons [due to the same mechanism (3.14a)], but we omit the derivation and give here only the result

$$\frac{\hbar}{\tau(E)E}\simeq\frac{WE}{E_c^2}\simeq\frac{\overline{M}}{M}\frac{E}{\hbar\omega_0}\ll 1\quad\text{for } E<\hbar\omega_0\ , \tag{4.43}$$

where E_c is determined by (1.8) and $\omega_0 \simeq \sqrt{\mathcal{E}_0/Ma^2}$ is a characteristic crystal-like frequency coinciding with the first maximum of the vibrational density of states in the glass (for amorphous SiO_2 $\hbar\omega_0 \approx 130$ K).[29] It is very important that with respect of interactions with phonons the whole picture of all low-energy excitations in the SPM appears to be internally consistent (about the limitations due to "nondiagonal" interactions between soft harmonic oscillators see the end of Sec. VII).

V. COMPARISON WITH EXPERIMENT

The results obtained above within the framework of the SPM almost coincide with predictions of the AHVP model. A slight difference is present in formulas (4.10), (4.25), and (4.27) but the accuracy of the available experimental data is not sufficient to resolve it. From comparison of our theory with experiment we can extract the values of important parameters (or some combination of them) involved in the SPM. Let us start with the specific-heat data. For amorphous SiO_2 (Suprasil W) the numerical value of the specific heat at $T=0.2$ K is 0.44 μJ/cm^3 K.[38] The theory gives for it the value (4.10). To estimate the logarithm we put $\tau_{\min}(T) \approx 10^{-9}/T^3$ (K) s [see (4.21)], $t_{\text{expt}}=10$ s and $W/k=10$ K. In this case

$$\ln^{1/3}\frac{W}{kT\sqrt{p_{\min}}}=\ln^{1/3}(0.45\times 10^6)=2.35 \ . \tag{5.1}$$

Equating (4.10) to 0.44 μJ/cm^3 K for $T=0.2$ K we obtain

$$\frac{\mathcal{P}_0\eta_L^{5/2}}{W}=0.82\times 10^{32}\ \text{erg}^{-1}\,\text{cm}^{-3} \ . \tag{5.2}$$

We can derive the numerical value for another combination of parameters from the sound velocity measurements. In accordance with (4.30), the temperature dependence of the sound velocity for sufficiently low temperatures (4.24) is logarithmical one

$$\left[\frac{v(T)-v(T_0)}{v(T_0)}\right]_{\text{res}}=C\ln\frac{T}{T_0} \ , \tag{5.3}$$

where

$$C=\frac{\mathcal{P}_0 H'^2 W}{\rho v^2\sqrt{\eta_L}} \ . \tag{5.4}$$

The value of C is known from experiment. For example, for SiO_2,[39,40] $C_l=3.1\times 10^{-4}$ (for longitudinal sound). As a result, we have

$$\frac{\mathcal{P}_0 H_l'^2 W}{\rho v_l^2\sqrt{\eta_L}}=3.1\times 10^{-4} \ . \tag{5.5}$$

From (5.2) and (5.5) we can obtain for SiO_2 ($\rho=2.2$ g/cm^3, $v_l=5.8\times 10^5$ cm/s)

$$\frac{H_l' W}{\eta_L^{3/2}}=1.04\ \text{eV} \ . \tag{5.6}$$

This combination of parameters is very important because it determines the value of the deformation potential γ (4.17). For example, for the frequency $\omega/2\pi=0.692$ GHz (it should be noted, that in our theory the deformation potential γ logarithmically depends on ω) we have from (5.6), (4.17), and (4.4)

$$\gamma_l \approx 1.7\ \text{eV} \tag{5.7}$$

[we put in (4.4) $E=\hbar\omega$, $W/k=10$ K, and $p=1$]. This value is in a good agreement with experimental results where $\gamma_l \approx 1.6$ eV (Ref. 33) for this frequency.

Let us now compare with experiment the results of our calculations for the dielectric properties of glasses. An important difference that should be pointed out is that the low-temperature ultrasonic properties of glasses are only weakly influenced by the impurity content of the material.[8] On the contrary, the magnitude of the similar dielectric properties depends strongly on the presence of polar impurities [for example, OH groups (Ref. 34)]. So, the comparison of our results with experimental ones makes sense for chemically pure glasses only.

For example, the variation of the dielectric constant with temperature in vitreous silica Suprasil W (<5 ppm OH) between 1 and 5 K at 10 GHz agrees with Eq. (4.39) and equals 1.24×10^{-4}.[34] As a result, we have for the dimensionless ratio in (4.39)

$$A\equiv\frac{\mathcal{P}_0 d_0^2\eta_L^{7/2}}{W}=0.92\times 10^{-5} \tag{5.8}$$

(the same value for A can be extracted from dielectric loss data).[41,42] From (5.8) and specific-heat data (5.2) we extract the combination

$$d_0\sqrt{\eta_L}=0.07(1e\cdot 1\ \text{Å})=0.33D \ . \tag{5.9}$$

According to (4.38) the quantity $d_0\sqrt{\eta_L}$ determines the value of the TLS dipole moment m. For example, for the frequency $\omega/2\pi=720$ MHz we have from (5.9), (4.38), and (4.4)

$$m \approx 0.55D \tag{5.10}$$

[we put in (4.4) $E=\hbar\omega$, $W/k=10$ K, and $p=1$]. From dielectric rotary echo experiments for Suprasil W,[36] we find (for the intrinsic TLS) the dipole moment m to be distributed with a width $0.2D$ and with a mean value of $m=0.7D$. From usual echo experiments we have $m=0.5D$ for the same frequency.[35]

From the comparison of our theory with low-temperature experiments in glasses we determine here only *three* independent combinations of parameters introduced in the theory [see (5.2), (5.5) or (5.6), (5.8), or (5.9)]. According to the SPM, all the universal low-temperature properties of glasses are determined only by these three combinations of parameters. It makes the effective mass M and other parameters such as $\mathcal{E}_0$, a, $\mathcal{P}_0$, H', and q^* in some sense *unobservable* quantities. It is impossible to deduce them from the physical experiment. One can give only order-of-magnitude estimates (see Sec. VII) or take them from the numerical modeling of the glass.[37]

One of the important parameters of the SPM is characteristic energy W (2.6). It determines the region of applicability of the AHVP model. Unfortunately, it is impossible as was mentioned above to determine it from the low-temperature properties of glasses which are determined by the TLS's alone. Indeed, W contributes to the formulas only under the sign of logarithm which is weakly sensitive to the value of W (we used for estimation in such cases $W/k = 10$ K).

Additional possibilities arise from the comparison of the predictions of the SPM with properties of glasses at temperatures larger than W/k. Some of these properties are determined by quasilocal harmonic oscillators. For example, their contribution to the specific heat appears to be proportional to T^5 (for $kT \gg W$).[29] It produces the minimum in the temperature dependence $C(T)/T^3$.[28,29] From the position of this minimum $T_{\min}$ one can determine the characteristic energy W. In Ref. 28 we made an effort to solve the Schrödinger equation for anharmonic oscillator (2.1) numerically. We calculated the positions of about 20 quantum levels in the potential (2.1) for a large number of relevant points on the plane η, t, and their contribution to the specific heat with distribution function (3.5). From numerical calculations[28] it has been found that $kT_{\min}/W \approx 0.4-0.5$ [depending on the details of the distribution function $\Psi(\eta)$]. For SiO$_2$, $T_{\min} \approx 2$ K, i.e., $W/k \approx (4-5)$ K. A somewhat smaller value was obtained in Ref. 29: $W/k \approx 3.3$ K.

Another possibility comes from the data on relaxational ultrasound absorption. Double-well potentials with high barriers are responsible for this absorption. The value of W determines the so-called crossover temperature T_c from tunneling to activation hopping transitions over the barrier[43] [above this temperature the tunneling plateau (4.23) is replaced by the absorption increasing with temperature]

$$kT_c \approx \left(\tfrac{3}{8}\right)^{4/3} W \ln^{1/3}\frac{1}{\omega\tau_0} , \tag{5.11}$$

where τ_0 is the prefactor in the exponential temperature dependence of the thermal activation relaxation time: $\tau = \tau_0 \exp(V/kT)$. Inserting in this formula $W/k = 5$ K, $\omega/2\pi = 4$ kHz, and $\tau_0 = 10^{-12}$ s, we get $T_c = 3.5$ K in agreement with experiment.[44]

VI. WHY DO PHONONS GO SO FAR?

In this section let us estimate the value of the dimensionless parameter C (5.4). Inserting (3.21) in (5.4) we obtain

$$C = \frac{\mathcal{P}_0 H'^2 W}{\rho v^2 \sqrt{\eta_L}} \simeq \frac{\Psi(0) H' W}{\rho v^2 \Omega_0 \sqrt{\eta_L}} \simeq \Psi(0)\eta_L^{3/2} , \tag{6.1}$$

where we have used $\rho v^2 \Omega_0 \simeq \mathcal{E}_0$, $H' \simeq 1$, and (2.6). Thus, in our theory, the parameter C appears to be proportional to the small number $\eta_L^{3/2} \simeq 10^{-3}$. Therefore, to get the observable value for C of the order of 10^{-4} we have to put into (6.1) $\Psi(0) \simeq 10^{-1}$. This is an acceptable order-of-magnitude estimate for the normalized distribution function $\Psi(\eta)$ at $\eta = 0$ because this function has a maximum at $\eta \approx 1$ [see Fig. 3(a)].

The physical meaning of the small parameter $\eta_L^{3/2}$ can be understood if we take into account that the scales of atomic parameters $a \simeq 1$ Å and $\mathcal{E}_0 \simeq 10$ eV involved in expression (2.5) for η_L come from the parameters of quantum mechanics. Indeed, for these quantities we have the following order-of-magnitude estimates:

$$a \simeq \frac{\hbar^2}{m_e e^2}$$

and (6.2)

$$\mathcal{E}_0 \simeq \frac{m_e e^4}{\hbar^2} ,$$

where m_e is an electron mass. The first quantity in (6.2) is the Bohr radius and the second one is a binding energy for electron in a hydrogen atom. Inserting these values in (2.5) we get

$$\eta_L^{3/2} \simeq \sqrt{m_e/M} . \tag{6.3}$$

Thus, the fundamental physical reason for the small parameter η_L in solids is the small ratio of the electron mass and the effective mass of the tunneling entity. It is just the same parameter which determines in the crystalline solids the smallness of the frequency of short-wave phonon-phonon collisions (in comparison with their own frequency) due to anharmonic processes.[45] It can be expressed through the small ratio of the characteristic phonon energy $\hbar\omega_0 \simeq \hbar\sqrt{\mathcal{E}_0/\bar{M}a^2}$ (see Appendix A) to energy $\mathcal{E}_0$ (it determines relative high-frequency vibration amplitude of the atoms x_0/a):

$$(\eta_L^{3/2})_{\text{crystal}} \simeq \sqrt{m_e/M} \simeq \frac{\hbar\omega_0}{\mathcal{E}_0} \simeq \sqrt{\hbar^2/\bar{M}a^2\mathcal{E}_0} \simeq \left[\frac{x_0}{a}\right]^2 , \tag{6.4}$$

where $x_0 \simeq \sqrt{\hbar/\bar{M}\omega_0}$. This ratio can also be thought of as the ratio of the kinetic and potential energies. The scale of the kinetic energy is set by phonons while the scale of the potential energy is set by electronic binding energies. Nearly the same parameter appears in glasses when we consider the excitation spectrum in soft atomic potentials. Thus, the small anharmonicity in crystalline solids and small concentration of TLS's in glasses have the same origin.

Thus, we can say that the smallness of the dimensionless parameter C which appears in a natural way in the theory follows from the softness of the local atomic potentials responsible for TLS's in the glass structure. On the other hand, the parameter C is equivalent to the dimensionless parameter $\bar{P}\gamma^2/\rho v^2$ in the AHVP model which characterizes the interaction of TLS's in a glass. We come to the conclusion that this parameter should always be small enough. The reason is the small ratio of the electron mass to the effective mass of the tunneling entity. This conclusion can be an answer to the question "Why do phonons go so far?."[19] We have also shown that due to the interaction between soft atomic potentials C does not depend on the concentration of elastic defects in the glass structure.

From the theory it follows also that the value (5.2) $\mathcal{P}_0\eta_L^{5/2}/W$ can be considered as TLS's density of states $\bar{P}$ [see (4.7)]. Concentration of TLS's with energies from 0 to W in such a case is of the order of $\mathcal{P}_0\eta_L^{5/2}$. Supposing $\eta_L = 10^{-2}$ [see (2.5)], $\Psi(0) \simeq 0.1$ and making use of (3.21) we obtain

$$\bar{P} \simeq 10^{-6}/a^3 \; . \tag{6.5}$$

The value $1/a^3$ is roughly of the order of the concentration of atoms constituting the glass. We see that $\simeq 10^{-6}$ part of them contributes to the specific heat at a few K only. This fact was known from experiments a long time ago[2] but had no explanation so far. Soft potential theory comes to this result in a natural way.

To complete our discussion of the problem let us show that the smallness of the dimensionless parameter A (5.8) (of the order of 10^{-5}) which determines the dielectric properties of glasses [see (4.39)–(4.41)] also follows naturally from our consideration. Using (2.6), (3.21), (4.37), $e^2/a \simeq \mathcal{E}_0$ [see (6.2)] and $a^3 \simeq \Omega_0$ we have

$$\begin{aligned} A = \frac{\mathcal{P}_0 d_0^2 \eta_L^{7/2}}{W} &\simeq \Psi(0)\eta_L^{3/2} \left[\frac{q^*}{e} \right]^2 \\ &\simeq 10^{-4} \left[\frac{q^*}{e} \right]^2 \end{aligned} \tag{6.6}$$

[because $\Psi(0) \simeq 10^{-1}$ as follows from (6.1)]. To get the right order of magnitude (10^{-5}) for quantity (5.8) we should suppose that for intrinsic TLS's in Suprasil W the effective charge $q^* \simeq 0.3e$. It correlates with the results of positron annihilation experiments[46] where, for instance, the effective charge of oxygen in different forms of quartz glass was found to take values between $-0.46e$ and $-0.65e$ (whereas, it is about $-1e$ in crystals). This decrease in the effective charge implies an increase in the covalency of the Si-O bond in the glassy state.

As a result, the smallness of the parameter A in (5.8) has the same ground as the smallness of the parameter C (6.1), plus an additional smallness of the effective charge q^*: $A \simeq C(q^*/e)^2$. On the other hand, the parameter A is equivalent to the dimensionless parameter $\bar{P}m^2$ in the AHVP model and we have understood now why it is so small.

VII. WHY ARE THE MAGNITUDES OF THE VARIOUS LOW-TEMPERATURE PROPERTIES SO NEARLY THE SAME FOR GLASSY SYSTEMS?

Since one has understood that the TLS's are responsible for the low-temperature properties of glasses one question is perpetually up in the air. Why are the magnitudes of the various low-temperature properties nearly the same for the overwhelming majority of glassy systems? For almost all the glasses the TLS's density of states $\bar{P}$ is of the order of 10^{45} J^{-1} m^{-3}, the deformation potential $\gamma \simeq 1$ eV, and the parameter $C = \bar{P}\gamma^2/\rho v^2 \simeq 10^{-4}$. Where do these particular values come from? Does this universality arise due to numerical "accidents" in the TLS's parameters? Or is there some fundamental physical reason which underlies it?

Partly, the answer to this question was already given in the previous sections. We gave there the order-of-magnitude estimates for various microscopic TLS's parameters that determine the low-temperature universal properties of glasses. But, in those sections we either limited ourselves with the case of a-SiO$_2$ or gave the order-of-magnitude estimates irrespective to any particular glass. These estimates played mainly an auxiliary role. However, in this section we want to analyze the problem of universality in more detail and to understand what are the macroscopic parameters that govern the universal properties of glasses. Any why are these properties slightly different from glass to glass.

If we examine the case closely, we can see that the same "universality" exists among the values of different macroscopic parameters of glasses (as well as crystalline solids), for instance, the sound velocity v and the mass density ρ. But how are the microscopic TLS's parameter related with the macroscopic parameters of a material?

In this section we show that in the framework of the SPM it is possible to answer this intriguing question. Using the results obtained in the previous sections we can estimate the characteristic microscopic parameters of glasses from the macroscopic material parameters ρ, v, and the average mass of the atoms $\bar{M}$. Of course, we are far from the conclusion that these values can be calculated exactly for any glass (i.e., our purpose is not a *fit* of the experimental data). However, we can give the right *order of magnitude* for $\bar{P}$, γ, and C and demonstrate the visible *correlation* of these values with usual macroscopic characteristics of the glass which are not related with TLS's. This correlation is seen, for instance, if the values ρ and v are changed under pressure. Or, the chemical composition of the glass is changed.

The TLS's density of states $\bar{P}$ is determined by (4.7):

$$\bar{P} \simeq \frac{\mathcal{P}_0 \eta_L^{5/2}}{W} \simeq \Psi(0)\frac{\sqrt{\eta_L}}{a^3\mathcal{E}_0} = \frac{\Psi(0)}{a^2\mathcal{E}_0}\left[\frac{\hbar^2}{2Ma^2\mathcal{E}_0}\right]^{1/6} , \tag{7.1}$$

where we used (3.21), the estimations $\Omega_0 \simeq a^3$, $H' \simeq 1$, and (2.5), (2.6). The order of magnitude of the deformation potential γ is [see (4.17)]

$$\gamma \simeq \frac{W}{\eta_L^{3/2}} = \mathcal{E}_0\sqrt{\eta_L} = \mathcal{E}_0\left[\frac{\hbar^2}{2Ma^2\mathcal{E}_0}\right]^{1/6} , \tag{7.2}$$

where we used (2.5) and (2.6).

The estimation of the dimensionless parameter $C = \bar{P}\gamma^2/\rho v^2$ characterizing the interaction among TLS's, the variation of the sound velocity with temperature, and the relaxation ultrasonic absorption in glasses follows from (7.1) and (7.2) if we take into account that $\rho v^2 a^3 \simeq \mathcal{E}_0$ [see also (6.1)],

$$C \simeq \Psi(0)\eta_L^{3/2} = \Psi(0)\sqrt{\hbar^2/2Ma^2\mathcal{E}_0} \; . \tag{7.3}$$

The minimum value of the TLS's relaxation time $\tau_{\min}(T)$ is determined by the following combination of the parameters [see (4.21)]:

$$K_3 \simeq \frac{W^2k^3}{\rho\hbar^4 v_t^5 \eta_L^3} = \frac{\mathcal{E}_0^2 \eta_L k^3}{\rho\hbar^4 v_t^5} = \frac{\mathcal{E}_0^2 k^3}{\rho\hbar v_t^5}\left[\frac{\hbar^2}{2Ma^2\mathcal{E}_0}\right]^{1/3} \tag{7.4}$$

and $1/\tau_{\min}(T) \simeq K_3 T^3$.

The main idea is to express all these quantities via the macroscopic parameters of the glass ρ, v, $\bar{M}$, and additionally $\bar{M}/M$ and $\Psi(0)$. In order to do this, we will use the following estimations of the characteristic length a and the energy $\mathcal{E}_0$ (see Appendix A):

$$a \simeq \left[\frac{\bar{M}}{\rho}\right]^{1/3}, \quad \mathcal{E}_0 \simeq \bar{M} v^2 . \tag{7.5}$$

Whereas the estimates based on Eqs. (7.5) play in the previous sections only an *auxiliary* role *all* the results of this section are based on these equations (as well as on the analytical calculations of the previous sections). To emphasize this, Eq. (7.5) is given here as a separate numbered equation, whereas the corresponding part of the paper is put into Sec. VII.

Admittedly, these are very crude approximations because we have neglected all the details in the geometry of the particular tunneling objects which can be different for different glasses. Nevertheless, these crude approximations lead to a striking correlation with the experimental data. As a result we obtain

$$\bar{P} \simeq \Psi(0) \frac{\hbar^{1/3} \rho^{10/9}}{\bar{M}^{22/9} v^{7/3}} \left[\frac{\bar{M}}{M}\right]^{1/6} \tag{7.6}$$

for the TLS's density of states,

$$\gamma \simeq \hbar^{1/3} v^{5/3} \rho^{1/9} \bar{M}^{5/9} \left[\frac{\bar{M}}{M}\right]^{1/6} \tag{7.7}$$

for the deformation potential of TLS,

$$C \simeq \Psi(0) \frac{\hbar \rho^{1/3}}{v \bar{M}^{4/3}} \sqrt{\bar{M}/M} \tag{7.8}$$

for the dimensionless parameter C, and

$$K_3 \simeq \frac{k^3 v^{10/3} \bar{M}^{10/9}}{\hbar^{10/3} \rho^{7/9} v_t^5} \left[\frac{\bar{M}}{M}\right]^{1/3} \tag{7.9}$$

for the parameter K_3. As to the estimations of the characteristic energy W and of the parameter of anharmonicity η_L we derive from (2.6), (2.5), and (7.5):

$$W \simeq \frac{\hbar^{4/3} \rho^{4/9} v^{2/3}}{\bar{M}^{7/9}} \left[\frac{\bar{M}}{M}\right]^{2/3} \tag{7.10}$$

and

$$\eta_L \simeq \frac{\hbar^{2/3} \rho^{2/9}}{\bar{M}^{8/9} v^{2/3}} \left[\frac{\bar{M}}{M}\right]^{1/3} . \tag{7.11}$$

Thus, in the framework of the SPM we have related the microscopic TLS's parameters with the macroscopic parameters of the material ρ, v, and $\bar{M}$. This is especially important for TLS's density of states $\bar{P}$ and for the deformation potential γ which are the phenomenological parameters in the AHVP model. They depend additionally on the ratio of the average mass to the effective mass of the tunneling entity $\bar{M}/M$ and on the value of the normalized distribution function $\Psi(\eta)$ at $\eta=0$. Both of these values are not known *a priori* and can be different for different glasses. Therefore, to check whether the correlation exists *we have to assume* these unknown constants to be the same for all the glasses. This is the idea. We put, for example, $\Psi(0)=0.1$ (see Sec. VI) and $M=\bar{M}$. For the sound velocity v we will use the value of the longitudinal sound velocity v_l. The only exception is (7.9) [see also (7.4)] where we will use the transverse sound velocity v_t. Due to $v_t^5 \ll v_l^5$ the relaxation rate of TLS, $1/\tau_{\min}$ is determined predominantly by emission and absorption of transverse phonons.

In Table I, we present the measured values of various tunneling parameters from[21,47–51] and calculated from Eqs. (7.6)–(7.11) for 22 different glasses. First, for most glasses the theory gives the right order of the value for all the TLS's parameters. Second, a visible correlation obviously exists between measured and calculated values. Moreover, for a large number of glasses a striking coincidence for deformation potential γ and for TLS relaxation rate (characterized by the coefficient K_3) between measured and calculated values occurs. To be quite frank, we never expected this because our approximations (7.5) cannot pretend the accuracy which occurs in reality (maybe the reason is that both values only slightly depend on the ratio $\bar{M}/M$).

In a number of cases, the above-mentioned coincidence (especially for the density of states $\bar{P}$) does not occur. It cannot be obtained by using another value of the distribution function $\Psi(0)$. We guess that the reason is due to the geometrical factor a and that the approximation (7.5) for a is very crude. But we will not develop this idea further because we do not have the purpose to get an ideal coincidence. Note only that the deviation from experimental data happens as a rule if the parameter of anharmonicity η_L is not small enough. It means maybe that approximation (2.1) is too crude and one needs to keep higher-order terms ($\propto x^5$ and x^6).

There is a visible *regular* deviation (by a factor of 5) between the calculated values of W and values taken from the position of the minimum in the $C(T)/T^3$. According to our point of view, the probable reason is that the effective mass of the tunneling entity M is most likely to be heavier (≈ 10 times) than the average mass $\bar{M}$.[29] However, we cannot use M as a fitting parameter in Eq. (7.10) because it has only order-of-magnitude meaning and we do not know the numerical factor.

In Table II, we present the estimates of the TLS's parameters in SiO_2 aerogels with different mass density using the data from Ref. 52. For our estimates we have used the same value for $\Psi(0)=0.1$ and $M=\bar{M}$. We can see that the calculated density of states $\bar{P}$ for the sample with the smallest value of ρ is about factor 40 larger than the value in a-SiO_2 (see Table I). This correlates with the specific-heat measurements[52] where specific heat for this material at $T \approx 0.5$ K is about factor 50 larger than the specific heat of a-SiO_2. The theory also predicts large ultrasonic absorption and a small value for the deformation potential in these substances.

The correlation between the parameters of TLS's and the macroscopic parameters of the material also exists if the macroscopic characteristics of the glass are changed under pressure. If for sufficiently small values of the pressure only the sound velocity v and the mass density ρ are changed, there should be a correlation

TABLE I. Summary of the data available on the low-temperature ($T<1$ K) acoustic properties for 22 selected glasses and calculated ones from (7.6)–(7.11) for $\Psi(0)=0.1$ and $M=\overline{M}$. The parameters ρ, v_l, v_t, $\overline{M}$, W, C_l, γ, $\overline{P}$, K_3, and η_L are defined in the text. $A1$ and $A2$, lithium borate glasses; $A1=B_2O_3-0.5Li_2O-0.7$ LiCl, $A2=B_2O_3-0.5Li_2O$. The experimental value of W was estimated from the position of the minimum in the specific-heat data [$C(T)/T^3$ see (Refs. 28 and 29)].

	ρ	v_l	v_t	$\overline{M}$	W (K)		C_l (10^{-4})		γ (eV)		$\overline{P}$ (10^{45}/J m^3)		K_3 (10^9/K^3 s)		η_L	
Glass	(g/cm)^3	(km/s)		(a.u.)	Expt.	Theor.	Expt.	Theor.	Expt.	Theor.	Expt.	Theor.	Expt.	Theory	10^{-2}	Ref.
a-SiO$_2$	2.2	5.8	3.8	20.0	4–5	22	3.1	2.1	1.04	0.90	0.8	0.75	0.4	0.3	1.6	21
BK7	2.51	6.2	3.8	19.8		25	2.7	2.1	0.96	1.01	1.1	0.76	0.3	0.33	1.6	21
LaSF-7	5.79	5.64	3.6	41.3		19	1.2	1.1	1.46	1.43	0.4	0.4	0.4	0.37	1.1	21
SF-4	4.78	3.78	2.24	44.4		13	2.2	1.4	0.72	0.75	1.1	0.69	1.3	1.33	1.3	21
SF57	5.51	3.57	2.1	51.6		12	2.1	1.3	0.74	0.75	1.1	0.64	1.8	1.61	1.2	47
SF59	6.26	3.32	1.92	58.1		11	2.3	1.3	0.77	0.72	1.0	0.65	2.2	2.0	1.2	21
V52	4.8	4.15	2.25	41.9		14	4	1.4	0.87	0.85	1.7	0.64	1.5	1.66	1.3	21
BALNA	4.28	4.3	2.3	36.1		15	3.8	1.6	0.75	0.81	2.1	0.74	1.1	1.55	1.4	21
LAT	5.25	4.78	2.8	43.5		16	3.8	1.2	1.13	1.10	1.4	0.46	0.7	0.87	1.1	21
a-Se	4.3	2.00	1.05	79.0	1–1.5	5	1.2	1.2	0.25	0.35	2	0.66	5.2	14.5	1.1	21
As$_2$S$_3$	3.2	2.7	1.46	49.2	1.2–1.9	8	1.6	1.5	0.26	0.43	2	0.75	1.9	5.6	1.3	21
Zn glass	4.24	4.6	2.3	34.4		17	3.0	1.6	0.7	0.89	2.2	0.71	0.8	1.85	1.4	21
Cu$_{60}$Zr$_{40}$	7.39	4.3	2.1	74.2		11	0.18	0.73	0.42	1.29		0.24		3.56	0.8	47
Mn glass	4.05	4.7	2.5	33.4		17	1.3	1.6	5.6	0.9	0.015	0.7	56	1.32	1.4	47
LiCl×7H$_2$O	1.2	4.0	2.0	7.32		29	7.2	9.4	0.56	0.26	1.3	10.6	8	1.12	4.5	48
PMMA	1.18	3.15	1.57	6.65		26	2	14	0.39	0.16	0.6	23	9	1.54	5.7	21
PC	1.2	2.97		7.66		23	1.8	12	0.28	0.16	0.9	19	5		5.2	21
H$_2$O	0.94	4.12	1.93	5.97		31	8.	11	0.1	0.24	80±40	12.4		1.42	5.0	47,49
D$_2$O	1.044			6.67					0.215		8.8					50
B$_2$O$_3$	1.8	3.47	1.91	13.9		19	2.4	5.3	0.2	0.3	4.5	4.8		1.31	3.0	51
$A1$	2.05	6.2	3.7	14.5		29	8.9	2.9	0.73	0.83	5.1	1.3		0.31	2.1	51
$A2$	2.05	6.92	3.97	13		34	8.2	3.0	0.78	0.94	5.2	1.3		0.28	2.1	51

$$\overline{P}\propto\frac{\rho^{10/9}}{v^{7/3}}$$

and (7.12)

$$\gamma\propto v^{5/3}\rho^{1/9}\,.$$

In Table III, we present the values of different TLS's parameters for a glassy polymer at different pressures which were measured by Grace and Anderson[53] and calculated from (7.12). The predicted correlation between measured and calculated values really exists.

The same correlation with pressure is also traced a-SiO$_2$. In the paper of Bartell and Hunklinger[14] it has been reported that the ultrasonic absorption in a-SiO$_2$ in the low-temperature region increases under pressure. It is known, however, that in this material the sound velocity decreases with pressure[54] and in accordance with (7.8) and (7.9) the absorption $\alpha\propto(CK_3/v)T^3$ should increase when pressure is applied.

Let us see now what happens if we change the chemical composition of a glass. In the paper of Brand and Lohneysen[55] a systematic study of the low-temperature specific heat C ($0.1\le T\le 3$ K) of As$_x$Se$_{1-x}$ glasses over a wide range of the concentrations x has been presented. The linear temperature contribution to $C(T)$ ($\propto T$) which is attributed to TLS's shows a pronounced dependence on the concentration x with a minimum in the vicinity of the threshold of rigidity percolation $x=0.4$ and a maximum near $x\approx 0.7$. The vibrational contribution to $C(T)$ ($\propto T^3$) shows a similar concentration dependence, suggesting a strong correlation between TLS's and vibrational excitations predicted in Refs. 26 and 28 in the framework of the SPM.

The same nonmonotonic behavior occurs for macroscopic parameters: inverse sound velocity, compressibility, and the mole volume of the glass.[56] So let us try to look at the experimental data[55] from the point of view of our theory. If only the values ρ, $\overline{M}$, and v change with the concentration x, one gets the following correlation:

TABLE II. The same data as in Table I calculated for silica gels with different mass density. Parameters ρ, v_l, and v_t are taken from Ref. 52.

	ρ	v_l	v_t	$\overline{M}$	W (K)		$10^{-4}C_l$		γ (eV)		$\overline{P}$ (10^{45}/J m^3)		K_3 (10^9/K^{3}s)		η_L
Glass	g/cm^3	km/s		a.u.	Expt.	Theor.	Expt.	Theor.	Expt.	Theor.	Expt.	Theor.	Expt.	Theor.	10^{-2}
SiO$_2$ gel	0.87	1.85	1.18	20		7		4.8		0.12		3.8		4.7	2.8
SiO$_2$ gel	0.72	1.5	0.96	20		5		5.6		0.08		5.1		7.6	3.1
SiO$_2$ gel	0.27	0.425	0.26	20		1.5		14.1		0.009		32.4		167	5.9

TABLE III. Values of various tunneling model parameters for measurements under pressure on a glassy polymer after Ref. 53 and calculated from (7.12). All data (except pressures) are normalized to zero-pressure values. The value B describes the cubic contribution to the specific heat from soft quasilocal harmonic oscillators: $B=C_{3\rho}$ [see (7.14)].

P			$\bar{P}\sim p^{10/9}/v^{7/3}$		$B\sim\rho^{2/9}/v^{11/3}$		$\rho v/\bar{P}\gamma^2\sim 1/\rho^{1/3}$		$\bar{P}\gamma^2\sim v\rho^{4/3}$		$\gamma^2\sim v^{10/3}\rho^{2/9}$		$\gamma^2/\rho v^5\sim 1/\rho^{7/9}v^{5/3}$	
(kbar)	ρ	v	Expt.	Theor.	Expt.	Theor.	Expt.	Theor.	Expt.	Theor.	Expt.	Theor.	Expt.	Theor.
0	1	1	1	1	1	1	1	1	1	1	1	1	1	1
0.8	1.03	1.021	0.94	0.98	0.94	0.93	0.99	0.99	1.06	1.06	1.13	1.08	0.99	0.94
1.4	1.041	1.04	0.89	0.95	0.89	0.87	0.99	0.99	1.09	1.1	1.22	1.15	0.98	0.91
1.9	1.047	1.048	0.87	0.94	0.87	0.85	0.99	0.99	1.11	1.11	1.27	1.18	0.96	0.89
3.9	1.081	1.086	0.78	0.9	0.78	0.75	0.93	0.97	1.26	1.21	1.62	1.34	0.99	0.82

$$C_1 \propto \frac{\bar{P}}{\rho} \propto \frac{\rho^{1/9}}{\bar{M}^{22/9}v^{7/3}} \tag{7.13}$$

for the linear contribution to the specific heat due to TLS's, and

$$C_3 \propto \frac{\bar{P}}{\rho W^2} \propto \frac{1}{\rho^{7/9}\bar{M}^{8/9}v^{11/3}} \tag{7.14}$$

for the cubic contribution to the specific heat (see Ref. 29) due to soft quasilocal harmonic oscillators.

Unfortunately, there are no experimental data available concerning the concentration dependence of the values ρ and v in $\mathrm{As}_x\mathrm{Se}_{1-x}$ at low temperatures. Therefore, we used room-temperature data which were given only in the concentration range $0<x<0.45$ (Ref. 57) and data for pure As.[58] The results are present in Figs. 4(a) and 4(b) together with experimental data.[55] An obvious correlation is seen in this case too. This means that the value of the distribution function $\Psi(0)$ and the ratio $\bar{M}/M$ are nearly independent of x and the main concentration dependence of the specific heat comes from the concentration dependence of the quantities v, ρ, and $\bar{M}$.

At the end of this section we mention a few words about properties of glasses at higher temperatures when tunneling model is no longer applicable. In the paper of Freeman and Anderson[59] a fit of the thermal conductivity data for different glasses was presented. To fit the plateau region in the thermal conductivity of glasses they used together with usual scattering mechanism of phonons by TLS's, scattering of unknown nature which is proportional to ω^4, where ω is the phonon frequency. The coefficient of proportionality ($1/Z$ in their notations) appeared to be strongly correlated with the sound velocity of the glass (see Fig. 5):

$$\frac{1}{Z} \propto v^{-4} . \tag{7.15}$$

An explanation of such a correlation has not been given yet.

In the framework of the SPM we have shown in Ref. 60 that the physical reason for such scattering mechanism is the resonant scattering of phonons on the soft harmonic oscillators, whose density of states is proportional to ω^4 (see Appendix B)

$$l^{-1}_{\mathrm{res,vib}} = \frac{1}{6\sqrt{2}}\frac{\pi\omega C}{v}\left[\frac{\hbar\omega}{W}\right]^3 . \tag{7.16}$$

Using the estimates (7.8) and (7.10) we obtain from (7.16)

$$l^{-1}_{\mathrm{res,vib}} \propto \frac{\Psi(0)\bar{M}}{\rho v^4}\left[\frac{M}{\bar{M}}\right]^{3/2}\omega^4 , \tag{7.17}$$

i.e., coefficient of proportionality in (7.17) depends on the sound velocity just as was found in Ref. 59: $1/Z \propto v^{-4}$.

Another interesting relation has been established recently.[61] If T_{min} and T_{max} are the positions of the minimum and the maximum in the temperature dependence of the specific heat $C(T)/T^3$ correspondingly and T_g is the glass transition temperature then the ratio

$$R \equiv \frac{T_{\mathrm{max}}}{T_{\mathrm{min}}^{3/4}T_g^{1/4}} \approx 1.06 \tag{7.18}$$

is nearly the same for nine examined glasses with maximum deviation of 19%. Let us check this result from the point of view of our theory.

As it was mentioned before (see end of the Sec. V) $T_{\mathrm{min}} \propto W$. From the Lindemann criterion of melting the glass transition temperature $T_g \propto \mathcal{E}_0$. And origin of the maximum T_{max} from our point of view (which is different from Ref. 61) related with the following phenomenon.

Strongly rising resonant scattering of phonons due to harmonic oscillators (7.16) implies that at some energy E_d the mean free path of phonons becomes equal to their wavelength.[28,62] The value of this energy is determined by expression[62]

$$E_d \approx (0.6-0.75)WC^{-1/3} . \tag{7.19}$$

For example, for a-SiO_2, $E_d/k \approx 45$ K. At higher energies excitation from one oscillator can jump to other oscillators directly on the distance of the wavelength (compare with Einstein model[63]). The picture of independent quasilocal harmonic excitations in this case is lost. Phonons and harmonic oscillators with energies $E > E_d$ cannot be considered independently any longer. They become intermixed with each other. Above this energy the total density of states should be reconstructed. One can believe that just this phenomenon responsible for the bump in the specific heat $C(T)/T^3$ at temperature

$T_{\max} \approx E_d/5k$, for the rise of thermal conductivity above the plateau and for the "boson" peak at frequency $\hbar\omega \simeq E_d$ in the Raman scattering in glasses.

Making use of above-mentioned relations, (2.6) and (7.3), we derive

$$R \propto \frac{WC^{-1/3}}{W^{3/4}\mathcal{E}_0^{1/4}} \propto [\Psi(0)]^{-1/3} , \tag{7.20}$$

i.e., this ratio does not depend on material parameters at all.

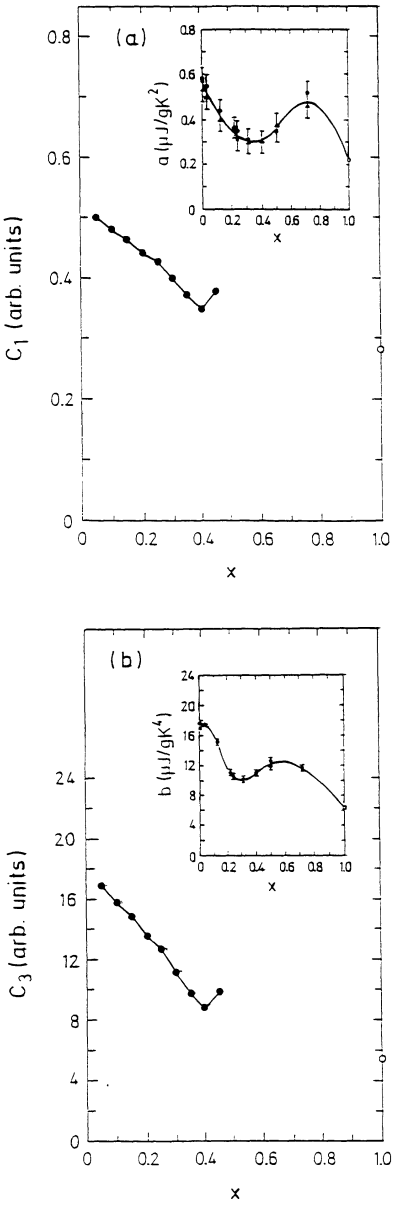

FIG. 4. Concentration dependence of the specific heat $C(T)=C_1T+C_3T^3$ for amorphous As_xSe_{1-x} calculated from (7.13) and (7.14) for $0<x<0.45$. The concentration dependence of ρ and v are taken from Ref. 57 at room temperature. The open circle is for pure As, after Ref. 58. (a) Coefficient C_1 of the T linear specific-heat contribution vs As concentration x. Inset: The corresponding experimental data from Ref. 55 for different values of the magnetic field. (b) Coefficient C_3 of the T^3 specific-heat contribution vs As concentration x. Inset: The corresponding experimental data from Ref. 55.

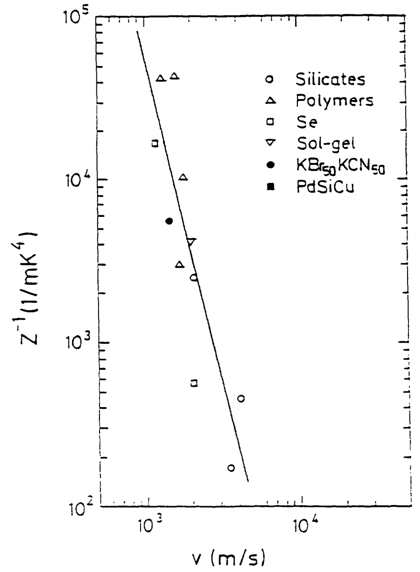

FIG. 5. Phonon scattering coefficient Z^{-1}, responsible for producing the plateau in the thermal conductivity vs phonon velocity for amorphous solids and a crystal exhibiting "glassy" behavior. The line has a slope of v^{-4} (from Ref. 59).

VIII. DISCUSSION

From the approach presented above it follows that as for the frequency and temperature dependencies, there is practically no difference between the results of AHVP model and our theory based on the SPM. As for the numerical estimates, we should consider the agreements (5.7) and (5.10) with experimental values for Suprasil W as a sufficiently good one. We have used here a simple version of the theory where the values $\mathcal{E}_0$, a in (2.1), M in (2.5), H' in (3.14a), and q^* in (4.35) have been considered to be the same for all TLS's in the glass. But, in principle, due to fluctuations of the structure parameters in the glass all of these quantities can fluctuate around their average values [as found for the intrinsic dipole moment m in Suprasil W (Ref. 36)]. Thus, the average value of a certain combination of these parameters need not coincide with the same combination of their average values.

Therefore, in the framework of the SPM there is no reason to introduce two types of TLS's to explain the specific heat and ultrasonic data as it was proposed in the paper of Black and Halperin.[64] The explanation of this fact is following. The integration over p of the density of states (4.7) in (4.8) due to the factor $L^{-2/3}$ give us an additional numerical factor 6 which does not appear in the AHVP model. From the other side, the logarithmic factors are canceled in the product $F(E,p)\gamma^2$ [see (4.7) and (4.17)] which determines the ultrasonic absorption (4.23), (4.31) and sound velocity (4.29), (4.30). There are no additional numerical factors in this case.

Another important difference between the two models concerns the time dependence of the specific heat. So, let us now discuss this point in more detail. The specific heat in the AHVP model is proportional to the logarithm of the time of experiment $t_{\rm expt}$:[31,5]

$$C_{\rm AHVP} \sim \ln\frac{4t_{\rm expt}}{\tau_{\rm min}(T)} , \tag{8.1}$$

while the SPM predicts (4.10):

$$C_{\rm SPM} \sim \ln^{1/3}\left[\frac{W}{kT}\sqrt{t_{\rm expt}/\tau_{\rm min}(T)}\right] , \tag{8.2}$$

i.e., a much more smooth dependence on $t_{\rm expt}$ than in the AHVP model. To observe this difference one needs to carry out investigations on a large time scale of the order of several hours, where the difference will be of the order of 20–30 %. For example, in the AHVP model one obtains the ratio

$$\frac{C_{\rm AHVP}(5h)}{C_{\rm AHVP}(1s)} = 1.4 \tag{8.3}$$

and in the SPM model:

$$\frac{C_{\rm SPM}(5h)}{C_{\rm SPM}(1s)} = 1.1 . \tag{8.4}$$

Usually, one considers the inverse time dependence of the heat release

$$\dot{Q} \sim \dot{C} \sim \frac{1}{t} , \tag{8.5}$$

which has been observed in different experiments[65–69] as an evidence of the logarithmical time dependence of the specific heat (8.1) in the AHVP model. However, the SPM predicts nearly the same result for the heat release[70]

$$\dot{Q} \sim \frac{1}{t\ln^{2/3}(t)} . \tag{8.6}$$

The only difference is in an additional smooth logarithmical dependence on time: $\ln^{-2/3}(t)$. The question arises: is the accuracy of existing experiments sufficient to discriminate between (8.5) and (8.6) (see Ref. 70)?

IX. CONCLUSION

Using the concept of soft atomic potentials we have shown that all low-temperature properties of glasses which receive explanation in the framework of the AHVP model are reproduced in the version of the SPM presented here. A small difference in the time-dependent specific heat poses a challenge to experimentalists. The approach presented above provides quantitative predictions. However, one should be aware of the fact that we have not take into account fluctuations of the model parameters (see Ref. 70 where the distribution function of W has been derived from the heat release data).

In addition to a correct description of the temperature and frequency dependencies of the various low-temperature physical properties of glasses, this approach, starting from the usual atomic values of the parameters $\mathcal{E}_0$, a, M, H', q^* introduced in the theory, gives correct order of magnitude estimates for the following.

(1) Deformation potential constant $\gamma \simeq H'W/\eta_L^{3/2} \simeq 1$ eV.

(2) Dipole moment of the TLS $m \simeq \sqrt{\eta_L}\,ea \simeq 0.5D$.

At last, the main result of our paper is that the interaction between soft atomic potentials is responsible for the universality of the low-temperature properties of glasses and that these properties are insensitive to the concentration of defects in the glass structure. As a result we have obtained a correct order-of-magnitude estimate for the following.

(3) Concentration of TLS's contributing to the specific heat at a few K $\simeq 10^{-6}$ of atomic concentration.

(4) Parameter

$$C = \bar{P}\gamma^2/\rho v^2 = \mathcal{P}_0 H'^2 W/\rho v^2\sqrt{\eta_L} \simeq \Psi(0)\eta_L^{3/2} \simeq 10^{-4}$$

which characterizes the interaction of TLS's in a glass. It appears to be small in accordance with existing experimental data.

(5) Parameter

$$A = \bar{P}m^2 = \mathcal{P}_0 d_0^2 \eta_L^{7/2}/W \simeq \Psi(0)\eta_L^{3/2}(q^*/e)^2 \simeq 10^{-5} ,$$

which characterized the TLS's contribution to dielectric properties of pure glasses.

In the framework of the SPM it is possible to relate the TLS's density of states $\bar{P}$ and the deformation potential γ with the macroscopic parameters of the material: the sound velocity v, the mass density ρ, and the average mass of the atoms $\bar{M}$. A striking correlation with different experimental data occurs. If the macroscopic parameters are changed under pressure or due to a change of the chemical composition of the glass, or by some other ways, the corresponding variation of the TLS's parameters coincides well with the predictions of our theory. It can be considered as an evidence that soft atomic potentials are responsible for the universal properties of glasses. And that the SPM gives an adequate description of these properties.

ACKNOWLEDGMENTS

Useful discussions with U. Buchenau, C. Enss, Yu. M. Galperin, V. L. Gurevich, S. Hunklinger, V. G. Karpov, G. Kasper, H. v. Löhneysen, V. Röhring, M. v. Schickfus, H. R. Schober, J. P. Sethna, G. Weiss, and A. Würger are gratefully acknowledged. I thank the Alexander von Humboldt Foundation for financial support and the Institute für Angewandte Physik, Heidelberg for hospitality during my stay in Germany.

APPENDIX A

Let us estimate the order of magnitude of the coefficient $\mathcal{E}_0$.[45] In the case of atomic displacements x of the order of the interatomic distance a, the potential energy $V(a) \simeq \mathcal{E}_0$ should be of the order of characteristic atomic energy $\mathcal{E}_a$ (i.e., a few electron volts). It is useful to express the energy $\mathcal{E}_a$ in terms of quantities that directly characterize a glass. In the harmonic approximation we have for the elastic energy density the expression $(\frac{1}{2})\lambda_{iklm}\epsilon_{ik}\epsilon_{lm}$, where λ_{iklm} is the tensor of elastic moduli and ϵ_{ik} is a strain. This expression could serve as a reasonable estimate for the potential-energy density even if the elastic strain tensor ϵ would be of the order of unity, i.e., if the atomic displacements would be of the order of interatomic distances a. On the other hand, the potential energy per atom then should be of the order of $\mathcal{E}_a$. Since the order of magnitude of the elastic moduli is ρv^2, we obtain that in order of magnitude $\rho v^2 \simeq \mathcal{E}_a / a^3$. This gives us the desired estimate

$$\mathcal{E}_0 \simeq \mathcal{E}_a \simeq \bar{M} v^2 \tag{A1}$$

if we use that $\rho a^3 \simeq \bar{M}$.

As for the Debye frequency, or rather the frequency ω_0 (that corresponds to the first maximum in the density of states), $\mathcal{E}_0$ is by no means proportional to $\hbar\omega_0$. In order to make an estimate of $\hbar\omega_0$ one should use the potential (2.1) with typical value of η, i.e., with $\eta \simeq 1$. Then, from the potential energy

$$V(x) = \mathcal{E}_0 \left[\frac{x}{a} \right]^2$$

we get the frequency

$$\omega_0 = \sqrt{2\mathcal{E}_0 / a^2 \bar{M}} \simeq \frac{v}{a} \tag{A2}$$

(where we have used the estimate $\mathcal{E}_0 \simeq \bar{M} v^2$), i.e., $\hbar\omega_0 \propto v$, but $\mathcal{E}_0 \propto v^2$ and the ratio $\hbar\omega_0 / \mathcal{E}_0$ is just a small parameter (related to η_L) which leads us to the condition $(x/a)^2 \ll 1$ [see (6.4)].

APPENDIX B

For the convenience of the reader and for the paper to be self-contained we give here derivation of harmonic-oscillator density of states. For $E \gg W$ it is determined by [see Eq. (2.8)]

$$n_{\mathrm{HO}}(E) = \int \int d\eta \, dt \, P(\eta, t) \delta(E - 2W\sqrt{\eta/\eta_L}) \,, \tag{B1}$$

where integration over η is taken from zero to infinity and we integrate over t within the interval $-\sqrt{32\eta/9} < t < \sqrt{32\eta/9}$ where one-well potentials exist. Now let us consider the case when the distribution function $P(\eta, t)$ can be taken in its simplest form Eq. (4.6). Then carrying out the integration we get

$$n_{\mathrm{HO}}(E) = \frac{1}{6\sqrt{2}} \frac{\mathcal{P}_0 \eta_L^{5/2}}{W} \left[\frac{E}{W} \right]^4 \propto E^4 \,. \tag{B2}$$

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*On leave from St. Petersburg State Technical University, St. Petersburg, Politechnicheskaya 29, 195251, Russia.

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