

# Density of vibrational states in glasses

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A theory of the vibrational spectra of glasses, based on allowance for the statistical fluctuations of the local elastic constants, is proposed. The existence is established of two characteristic energies  $\hbar\omega_1$  and  $\hbar\omega_2$ , dividing the spectrum into regions of qualitatively different behavior of the density of states  $n(\hbar\omega)$ . At low frequencies  $\omega \ll \omega_1$  the increase of the density of states is determined by the additive contributions of phonons and mutually noninteracting quasilocal vibrations in random soft atomic potentials in the glass. In the intermediate region  $\omega_1 \lesssim \omega \lesssim \omega_2$  the quasilocal vibrations interact strongly with phonons, and this makes their contributions superadditive. For  $\omega > \omega_2$  the growth of  $n(\hbar\omega)$  slows down. As a result,  $n(\hbar\omega)$  increases at first more rapidly and then more slowly than the Debye density of states. An analytical expression for  $n(\hbar\omega)$  is obtained in the  $T$ -matrix formalism in the region  $\omega < \omega_2$ , including the region of strong scattering. A numerical calculation of  $n(\hbar\omega)$  is performed in the coherent-potential approximation. The theory predicts qualitatively universal behavior of  $n(\hbar\omega)$  in different glasses.

## 1. INTRODUCTION

Experiments to measure the specific heat,<sup>1,2</sup> neutron scattering,<sup>3-5</sup> and Raman scattering of light<sup>6</sup> provide evidence that the density of vibrational states  $n(\hbar\omega)$  of glasses behaves in a more or less universal manner, changing little from one material to another. However, it differs substantially from the density of states of crystals. In the region of the lowest energies  $\hbar\omega \lesssim 1$  K the behavior  $n(\hbar\omega) \approx \text{const}$  corresponds to the well known model of two-level systems (TLS) in two-well atomic potentials.<sup>7,8</sup> However, at higher energies  $\hbar\omega \gtrsim 10$  K the behavior of  $n(\hbar\omega)$  is not described by the TLS model. In this region the experimental data<sup>3-5</sup> display a complicated dependence  $n(\hbar\omega)$ , which first increases more sharply than the Debye density of states  $g(\hbar\omega) \propto \omega^2$  and then becomes less steep, with a tendency to subsequent decrease. This behavior is illustrated in Fig. 1a.

In recent papers<sup>9,10</sup> this behavior of  $n(\hbar\omega)$  has been explained on the basis of the postulate that glasses have a fractal structure. It was assumed that excitations with wavelengths  $\lambda < L$ , called fractons, belong to a distinct fractal structure with correlation length  $L$  and small effective fracton dimension  $d \approx 1$ . The fracton density of states  $n(\hbar\omega) \propto \omega^{d-1}$  for  $\omega > s/L$  ( $s$  is the velocity of sound) was associated with the gently sloping part of the dependence at high energies. In this approach the form of the density of states  $n(\hbar\omega) \approx \text{const}$  at low energies remains unexplained. In addition, from our point of view, there are no physical grounds for the postulate that glasses contain a fractal structure limiting the propagation of short-wavelength excitations.

In the present paper we propose a different explanation of the dependence  $n(\hbar\omega)$  in a broad range of energies, from the lowest ( $\hbar\omega \ll 1$  K) up to energies comparable to the Debye energy ( $\hbar\omega \lesssim \hbar\omega_D$ ). Our explanation is based on allowance for fluctuations of the local elastic constants in the disordered atomic system of the glass and reduces to the following statements. In the soft atomic potentials that arise on account of the fluctuations, quasilocal vibrational states with random parameters are realized. At low energies these constitute two-level systems with density  $n(\hbar\omega) \approx \text{const}$ . States with high energies are similar in type to ordinary vi-

brations of softening defects in crystals. The density  $n(\hbar\omega)$  corresponding to these states increases rapidly with energy. At sufficiently high energies there are so many states that they interact strongly and become collective. This change in the character of the states leads to a flattening of the dependence  $n(\hbar\omega)$ , which we associate with the upper part of the curve in Fig. 1.

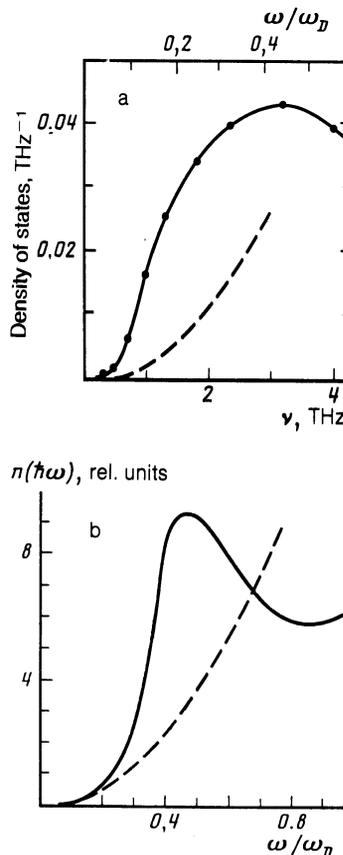


FIG. 1. a) Frequency dependence of the density of states in vitreous SiO<sub>2</sub>, from Ref. 3. The dashed curve shows the Debye density of states. On the upper scale, for later comparison, we have plotted the relative energy values  $\omega/\omega_D$ , where  $\hbar\omega_D$  is the Debye energy; b) energy dependence of the density of states as calculated in the coherent-potential approximation. The dashed curve shows the Debye density of states.

In the formulation of the problem our approach is, to a certain extent, similar to the analysis of Montgomery,<sup>11</sup> who showed that fluctuations of the elastic constants increase the density of the low-frequency vibrations. However, the analysis in Ref. 11 was performed with two strong restrictions. First, it was assumed that the fluctuations do not lead to the appearance of quasilocal vibrations. Second, the analysis in Ref. 11 was limited to the harmonic approximation. These restrictions are not fulfilled in the most interesting region of low frequencies, corresponding to strong local softening of the elastic constants.

The approach proposed by us is free from these restrictions. It takes into account both the presence of quasilocal vibrations and the effects of anharmonicity in the glass. Our approach makes it possible to take isotopic disorder into account as well, when necessary. Its results are qualitatively independent of the type of disorder, thereby explaining the universal behavior of the density of states in glasses.

The remainder of the article is organized as follows. First we consider the general problem of the density of vibrational states of an atomic system with uncorrelated random local perturbations. The results obtained express  $n(\hbar\omega)$  in terms of the parameters and bare density  $n_0(\hbar\omega)$  of isolated (not interacting with each other) random quasilocal states in an ideal lattice. We then concretize the density  $n_0(\hbar\omega)$  and the parameters of the isolated vibrations for a glass. The conclusions thus reached concerning the energy dependence of the density of states  $n(\hbar\omega)$  are then reinforced by a numerical calculation in the coherent-potential approximation.

## 2. VIBRATIONAL DENSITY OF STATES IN A RANDOM ATOMIC SYSTEM

The problem of this section is formulated as follows. We assume that in an ordered atomic system we have a single defect, characterized by a site perturbation  $U$  and leading to the appearance of a quasilocal state (on the background of the continuous spectrum). This state is characterized by energy  $\hbar\omega$  and width  $\Gamma$ , which depend on the perturbation  $U$  and also on the density  $g(\hbar\omega)$  of the continuous spectrum of the ordered system. In the problem under consideration random uncorrelated perturbations are present at each site and are characterized by a probability distribution  $\rho(U)$ . If the site quasilocal states did not interact with each other, the problem of determining the corresponding bare density of states  $n_0(\hbar\omega)$  for a given form of  $U$ ,  $\rho(U)$ , and  $g(\hbar\omega)$  could be solved trivially. The main difficulty lies in the determination of the density  $n(\hbar\omega)$  when interaction between the states of individual sites is taken into account.

Although the above problem is close in formulation to familiar problems concerning the density of the electron states of disordered systems (see, e.g., Ref. 12), it has a feature specific to itself: The site perturbations under consideration here are not true potentials but pseudopotentials, i.e., depend themselves on the energy:  $U = U(\hbar\omega)$ . This fact is well known in the theory of the vibrations of defects in crystals. For example, for an isotopic defect,<sup>13,14</sup>

$$U = \varepsilon \Delta M, \quad \varepsilon \equiv (\hbar\omega)^2. \quad (1)$$

For a defect of the force constant  $\kappa$  we can also introduce an effective local perturbation  $U$ , which appears in the equation for the spectrum of  $1 - UG^0$ , where  $G^0$  is the Green function

of the crystal. One can verify this by considering specific models of force-constant defects.<sup>13,14</sup> For considerable distortions of the force constant one can write the explicit expression<sup>14</sup>

$$U = \varepsilon \varepsilon_{\kappa} / (\varepsilon - \varepsilon_{\kappa}), \quad \varepsilon_{\kappa} = \hbar^2 \kappa / M, \quad (2)$$

where  $M$  is the mass of the atoms of the lattice. It is precisely the pseudopotential character of the site perturbations (in particular, the dependence  $U \propto \varepsilon$  for  $\varepsilon \rightarrow 0$ ) that ensures the existence of a boundary of the spectrum at  $\varepsilon = 0$  and the acoustic character of the spectrum in the limit of long wavelengths for a disordered atomic system.

The theory described below does not use an explicit form, of the type (1) or (2), for the dependence  $U(\varepsilon)$ . However, the existence of such dependences is manifested implicitly in the energy dependences of the density  $n_0$  and widths  $\Gamma$  of the bare states, since the desired density of states is expressed in terms of these quantities (see (22) below).

We start from a qualitative analysis of the density of states. We assume that the bare distribution function  $n_0(\hbar\omega)$  decreases rapidly with decrease of  $\omega$  as  $\omega \rightarrow 0$  (a detailed discussion of the form of  $n_0(\hbar\omega)$  is given in Sec. 3). In the limit  $\omega \rightarrow 0$  the bare density  $n_0$  turns out to be very small. The scattering of phonons by these states is correspondingly small. Therefore, we must expect that for  $\omega \rightarrow 0$  the density of states will be determined by additive contributions from the quasilocal vibrations and the phonons:

$$n(\hbar\omega) = n_0(\hbar\omega) + g(\hbar\omega). \quad (3)$$

With increase of  $\omega$  the scattering of phonons by resonance quasilocal vibrations becomes considerably more intense because of the increase in the density  $n_0(\hbar\omega)$  and width  $\Gamma(\hbar\omega)$  of the latter. The approximation (3) fails completely when the corresponding mean free path  $l \sim (n_0 \Gamma \lambda^2)^{-1}$  becomes comparable to the wavelength  $\lambda$ , i.e.,

$$\xi_1 = n_0 \Gamma \lambda^3 \sim 1. \quad (4)$$

The condition (4) determines the characteristic energy  $\hbar\omega_1$ , which is smaller than the Debye energy  $\hbar\omega_D$ .

We note that, from its meaning, the condition (4) can be represented in the form  $\text{Im}\Sigma \sim \hbar\omega$ , where  $\Sigma$  is the self-energy operator, whose imaginary part  $\text{Im}\Sigma \sim n_0 \Gamma / g$  describes the scattering-related lifetime of the state. The quantity  $\text{Im}\Sigma$  increases with increase of  $\hbar\omega$ . This energy-dependent broadening of the levels leads in an obvious way to "pumping" of some of the states from the high-frequency part of the spectrum into the low-frequency part. This implies that for  $\omega \gtrsim \omega_1$  (i.e., for  $\text{Im}\Sigma \gtrsim \hbar\omega_1$ ) the density of states should increase more rapidly than by the law (3).

The arguments given and the conclusion concerning the superadditivity of the densities of the quasilocal vibrations and phonons cease to be valid in the region of sufficiently high energies at which the quantity  $\text{Im}\Sigma$  becomes comparable to the characteristic width  $\hbar\omega_D$  of the spectrum, i.e.,

$$\xi_2 = n_0 \Gamma / g \hbar\omega_D \sim 1. \quad (5)$$

For the assumed sharp dependences  $n_0(\hbar\omega)$  and  $\Gamma(\hbar\omega)$  the characteristic energy  $\hbar\omega_2$  defined by the condition (5) turns out to be smaller than  $\hbar\omega_D$ . It follows from this that the above-predicted rapid growth of the resultant density of states  $n(\hbar\omega)$  appears only for  $\omega \lesssim \omega_2$ , while in the region  $\omega_2$

$\langle \omega < \omega_D$  the dependence  $n(\hbar\omega)$  should be qualitatively different. Taking into account the normalization of the number of states:

$$\int_0^{\omega_D} n(\hbar\omega) d\omega = \int_0^{\omega_D} g(\hbar\omega) d\omega \quad (6)$$

we can conclude that for  $\omega > \omega_2$  the dependence  $n(\hbar\omega)$  should be, on the average, at least weaker than the Debye dependence  $g \propto \omega^2$ . As the final result we arrive at an energy dependence  $n(\hbar\omega)$  of the type depicted in Fig. 1a.

We note here a further interpretation of the condition (5). In the strong-scattering region  $\text{Im}\Sigma > \hbar\omega$  the role of the wavelength of an excitation is played by the quantity  $\lambda \sim a\hbar\omega_D / \text{Im}\Sigma = a/\xi_2$ , where  $a$  is the lattice constant (in place of  $\lambda \sim a\omega_D/\omega$  for phonons). Therefore, the condition  $\xi_2 \ll 1$  corresponds to the long-wavelength approximation  $\lambda \gg a$ , while the condition (5) corresponds to failure of this approximation.

We turn to a quantitative treatment of the density of states. Let the Green function  $G_{\alpha\beta}^0$ , where  $\alpha$  and  $\beta$  are site indices, correspond to the ideal lattice. The standard expression

$$G_{\alpha\beta} = G_{\alpha\beta}^0 + G_{\alpha\gamma}^0 T_{\gamma\delta} G_{\delta\beta}^0 \quad (7)$$

relates the Green function  $G_{\alpha\beta}$  of the system under consideration to  $G_{\alpha\beta}^0$  through the scattering matrix  $T_{\alpha\beta}$ , satisfying the equation

$$T_{\alpha\beta} = U_\alpha \delta_{\alpha\beta} + U_\alpha G_{\alpha\gamma}^0 T_{\gamma\beta}, \quad (8)$$

where  $U_\alpha$  is the site perturbation. Introducing the  $t$ -matrix of an individual defect:

$$t_\alpha = U_\alpha / (1 - U_\alpha G_{\alpha\alpha}^0), \quad (9)$$

we represent Eq. (8) in the form

$$\tilde{T}_{\alpha\beta} = \delta_{\alpha\beta} + G_{\alpha\gamma}^0 t_\gamma \tilde{T}_{\gamma\beta} (1 - \delta_{\alpha\gamma}), \quad (10)$$

where the matrix  $\tilde{T}_{\alpha\beta}$  is related to  $T_{\alpha\beta}$  by  $T_{\alpha\beta} = t_\alpha \tilde{T}_{\alpha\beta}$ . Equation (10) is exact, and the second term in its right-hand side describes the interaction between individual quasilocal vibrations, the parameters of which are determined by the poles of the random amplitudes  $t_\alpha$ .

If Eq. (10) has been solved, and the Green function  $G_{\alpha\beta}$  (7) thereby determined, the density of states can be found using the relation (see Ref. 14)

$$n(\varepsilon) = \frac{1}{\pi} \lim_{\gamma \rightarrow +0} \text{Im} \sum_\alpha \left[ \left( 1 - \frac{dU_\alpha}{d\varepsilon} \right) G_{\alpha\alpha}(\varepsilon - i\gamma) \right]. \quad (11)$$

Here and below we use the following representation, customary in the theory of vibrations:

$$G_{\alpha\beta}^0(\varepsilon) = \sum_{\mathbf{k}} \frac{\exp[i\mathbf{k}(\mathbf{r}_\alpha - \mathbf{r}_\beta)]}{\varepsilon - \varepsilon_{\mathbf{k}}} = \sum_{\mathbf{k}} G_{\varepsilon_{\mathbf{k}}}^0 \exp[i\mathbf{k}(\mathbf{r}_\alpha - \mathbf{r}_\beta)] \quad (12)$$

with energy variables  $\varepsilon = (\hbar\omega)^2$  and  $\varepsilon_{\mathbf{k}} = (\hbar\omega_{\mathbf{k}})^2$ , where  $\mathbf{k}$  is the quasi-wave-vector of a phonon of frequency  $\omega_{\mathbf{k}}$  (Refs. 13 and 14). Then the site Green function is

$$G_{\alpha\alpha}^0 = P(\varepsilon) = \sum_{\mathbf{k}} G_{\varepsilon_{\mathbf{k}}}^0 = R^{-1}(\varepsilon) + i\pi g(\varepsilon), \quad (13)$$

where  $R^{-1}(\varepsilon)$  denotes the real part.

We shall solve Eq. (10) in the long-wavelength approximation  $\xi_2 \ll 1$ , which includes the regimes not only of weak ( $\xi_1 \ll 1$ ) but also of strong ( $\xi_1 \gtrsim 1$ ) scattering of phonons. It turns out that for  $\xi_2 \ll 1$  we can replace the quantity  $t_\gamma$  in (10) by its average value  $t \equiv \langle t_\gamma \rangle$ . This corresponds to the averaged- $t$ -matrix approximation.<sup>12</sup> In the Appendix it is shown that corrections to the solution obtained by this method in the given problem are small in the parameter  $\xi_2$ . Physically, the possibility of replacing  $t_\gamma$  by  $t$  in conditions of strong scattering is due to the self-averaging of the amplitudes  $t_\gamma$  over large spatial scales  $\lambda \gg a$  corresponding to resonance scattering of long-wavelength excitations. The self-averaging is associated with a specific feature of the problem under consideration, in which the concentration  $n_0 \Gamma$  of resonance quasilocal states increases sufficiently strongly with increase of energy. Because of this, a volume  $\lambda^3$  (for  $\lambda \gg a$ ) contains a large number of scattering centers, and their amplitudes  $t_\gamma$  undergo self-averaging.

In the averaged- $t$ -matrix approximation Eq. (10) is easily solved:

$$T_{\alpha\beta} = \sum_{\mathbf{k}} \frac{t_\alpha \exp[i\mathbf{k}(\mathbf{r}_\alpha - \mathbf{r}_\beta)]}{1 + t[P(\varepsilon) - G_{\varepsilon_{\mathbf{k}}}^0]}. \quad (14)$$

The calculation of the sums in the expression for the density of states (11) is again performed with allowance for the self-averaging property of the quantities  $t_\alpha$ . The result has the form

$$n(\varepsilon) = \frac{1}{\pi} \text{Im} \left\{ P(\varepsilon - \Sigma) \left( 1 - \frac{A[dU/d\varepsilon]}{A[1]} \right) \right\}, \quad (15)$$

where the self-energy part  $\Sigma$  of the averaged Green function is

$$\Sigma = t[1 + tP(\varepsilon)]^{-1}. \quad (16)$$

The expression (15) contains the average values

$$A[f(U)] = \left\langle \frac{f(U)}{1 - UP} \right\rangle = \lim_{\gamma \rightarrow +0} \int dU \rho(U) \frac{f(U)}{1 - U(\varepsilon - i\gamma)P(\varepsilon - i\gamma)}, \quad (17)$$

where  $f(U)$  represents  $U$ ,  $dU/d\varepsilon$ , or 1. In the cases under consideration the real part of the integral (17) is determined mainly by the neighborhood of  $U = 0$  (i.e., of the maximum of  $\rho(U)$ ), in which the difference of the denominator from unity can be neglected. The imaginary part is determined by the pole of the integrand near the real axis. As a result,

$$A[f(U)] \approx \langle f(U) \rangle + i\pi[\rho(U)f(U)U]_{U=R}, \quad (18)$$

where  $\langle f(U) \rangle$  denotes the usual average.

The quantities  $\rho(R)$  and  $dU/d\varepsilon|_{U=R}$  appearing in (18) can be expressed in terms of the characteristics  $\varepsilon$ ,  $\Gamma(\varepsilon)$ , and  $n_0(\varepsilon)$  of isolated quasilocal vibrations. For this we consider an isolated quasilocal vibration generated by a site perturbation  $U$ . We represent its energy variable in the form  $\varepsilon + i\Gamma$  with  $\Gamma \ll \varepsilon$  and write the equation determining the poles of the amplitude  $t$  (9):

$$1 - \left( U + \frac{\partial U}{\partial \varepsilon} i\Gamma \right) P(\varepsilon) = 0. \quad (19)$$

Separating the real and imaginary parts, we obtain

$$U=R(\varepsilon), \quad \Gamma=-\pi g R^2 (dU/d\varepsilon)_{\varepsilon=R}^{-1}. \quad (20)$$

Equations (20) determine the position  $\varepsilon$  and width  $\Gamma$  of an isolated vibration. The condition  $\Gamma \ll \varepsilon$  corresponds to low-frequency vibrations.<sup>13,14</sup> The density of such vibrations is

$$n_0(\varepsilon) = \rho[R(\varepsilon)] |dU/d\varepsilon|_{\varepsilon=R}. \quad (21)$$

With allowance for the expressions (18), (20), and (21), the formula (15) for the density of states takes the form

$$n(\varepsilon) = \frac{1}{\pi} [\text{Im} P(\varepsilon - \Sigma)] \left( 1 + \frac{\pi R^2 g}{\Gamma} \frac{\xi_2^2}{1 + \xi_2^2} \right) + [\text{Re} P(\varepsilon - \Sigma)] \frac{R n_0}{1 + \xi_2^2}, \quad (22)$$

$$\Sigma = R \frac{i\xi_2 + \xi_2^2}{1 + \xi_2^2}, \quad \xi_2 = \frac{n_0 \Gamma}{g R}. \quad (23)$$

We note that the definition (23) for  $\xi_2$  coincides with (5) to within unimportant numerical factors. One can convince oneself of this using the Debye model, in which

$$g(\varepsilon) = \frac{3}{2\varepsilon_D a^3} \left( \frac{\varepsilon}{\varepsilon_D} \right)^{1/2}, \quad \lambda \approx a \left( \frac{\varepsilon_D}{\varepsilon} \right)^{1/2}, \quad (24)$$

$$R(\varepsilon) \approx R(0) = -\varepsilon_D/3 \text{ for } \varepsilon \ll \varepsilon_D \equiv (\hbar\omega_D)^2.$$

With the same accuracy the parameter  $\xi_1$  from (4) can be represented in the form

$$\xi_1 = |R/\varepsilon| \xi_2 = n_0 \Gamma / g \varepsilon. \quad (25)$$

The parameters  $\xi_1$  and  $\xi_2$  increase with increase of  $\varepsilon$ , owing to the sharp dependences  $n_0(\varepsilon)$  and  $\Gamma(\varepsilon)$  (see Sec. 3).

The expression (22) is equally applicable both to disorder of the force constants and to isotopic disorder; the difference between these cases reduces entirely to the appropriate choice of  $\Gamma$  in (22). In addition, the expression (22) is not restricted to the harmonic approximation: Allowance for anharmonicity affects only the form of  $P(\varepsilon)$  and  $\Gamma$ . The applicability of the expression (22) is limited not only by the condition  $\xi_2 \ll 1$  but also to the comparatively low-frequency part of the spectrum (in practice,  $\varepsilon \lesssim \varepsilon_D/3$ ), in which the expressions (20) and (24) are valid.

Upon analysis of the behavior of  $n(\varepsilon)$  (22) two characteristic values of  $\varepsilon$  are revealed. The smaller of these,  $\varepsilon_1$ , corresponds to the condition  $\xi_1 = 1$  for which  $|\text{Im}\Sigma| = \varepsilon$ . For  $\xi_1 > 1 > \xi_2$  the density  $n(\varepsilon)$  increases somewhat faster than (3) with increase of  $\varepsilon$ , on account of both the strengthening of the energy dependence  $P(\varepsilon - \Sigma)$  when  $|\text{Im}\Sigma| > \varepsilon$  and the term  $\pi R^2 \xi_2^2 / \Gamma$  in (22). The increase of  $n(\varepsilon)$  becomes slower near another characteristic value  $\varepsilon = \varepsilon_2 > \varepsilon_1$ , corresponding to the condition  $\xi_2 = 1$  in (22). For  $\xi_2 \gg 1$ , i.e., for  $\varepsilon \gg \varepsilon_2$ , we have the decreasing function

$$n(\varepsilon) = \frac{R^2 g}{\Gamma} \text{Im}[P(\varepsilon - R)] \approx 0.3 \frac{\varepsilon_D g}{\Gamma}. \quad (26)$$

However, this dependence is realized outside the region  $\xi_2 \ll 1$  of applicability of the results obtained. We can give a rigorous argument for the weakening of the dependence  $n(\varepsilon)$  for  $\varepsilon \gtrsim \varepsilon_2$  only on the basis of the normalization condition (6). These conclusions correspond to the qualitative arguments outlined at the beginning of this section, and confirm the criteria (4) and (5). The characteristic values of the variable are related to the characteristic energies introduced above:  $\varepsilon_{1,2} = (\hbar\omega_{1,2})^2$ .

### 3. ISOLATED QUASILocal VIBRATIONS

A number of papers<sup>15-18</sup> are devoted to the examination of isolated vibrations in soft potentials of a glass, and our account will be brief. Its main purpose is to concretize the quantities  $n_0$ ,  $\xi_1$ ,  $\xi_2$ , and  $\Gamma$  in the expression for the density of states.

Following Refs. 15-18, we can write soft potentials using a single-mode expansion

$$V(x) = \mathcal{E} [\eta(x/a_0)^2 + t(x/a_0)^3 + (x/a_0)^4] \text{ for } |\eta| \ll 1, \quad t^2 \ll 1. \quad (27)$$

Here  $\mathcal{E} \sim Ms^2 \sim 10$  eV is an energy on the atomic scale,  $a_0 \sim 1$  Å is a characteristic atomic size, and  $\eta$  and  $t$  are random quantities with distribution<sup>17,18</sup>

$$P(\eta, t) = |\eta| P_0(\eta, t) \text{ for } |\eta| \ll 1, \quad (28)$$

where  $P_0(\eta, t)$  is a smooth function finite at  $\eta = 0$ . For soft potentials the distribution  $P(\eta, t)$  has a rapidly decreasing tail at  $|\eta| \ll 1$ . The potentials (27) can be either one-well or two-well potentials.

The analysis in Refs. 15 and 16 of the dynamics of an atomic particle in the potential (30) displays the characteristic parameter

$$\eta_L = (\hbar^2/2Ma_0^2 \mathcal{E})^{1/2} \sim (\hbar\omega_D/\mathcal{E})^{1/2} \sim 10^{-2}, \quad (29)$$

the meaning of which is that the energy gaps  $E(\eta, t)$  between the levels in the potential (27) are changed by an amount of the order of the levels themselves when the parameter  $\eta$  is changed by  $\eta_L$  or the parameter  $t$  is changed by  $\eta_L^{1/2}$ . This makes it possible to set  $P_0(\eta, t) = \text{const}$  in the calculation of the density of states

$$n_0(\hbar\omega) = \langle \delta(\hbar\omega - E(\eta, t)) \rangle$$

in sufficiently soft potentials, since the characteristic scales  $\delta\eta$  and  $\delta t$  of the attenuation of the distribution  $P_0(\eta, t)$  in the variables  $\eta$  and  $t$  are large in the sense that  $\delta\eta > \eta_L$  and  $\delta t > \eta_L^{1/2}$  (in reality,  $\delta t \gtrsim \delta\eta \gtrsim 0.1$  (Refs. 15, 16)).

The main results are the following. There exists a characteristic energy

$$w = \mathcal{E} \eta_L^2 \sim \hbar\omega_D (\hbar\omega_D/\mathcal{E})^{1/2} \sim 10 \text{ K}, \quad (30)$$

with respect to which the excitations are classified. For  $\hbar\omega \ll w$  the excitations represent a TLS in the two-well potentials (27) with  $n_0(\hbar\omega) = \text{const}$ . The density  $n_0(\hbar\omega)$  increases appreciably at energies  $\hbar\omega \gtrsim w$  corresponding to excitations in one-well soft potentials. Excitations with energy  $\hbar\omega \gg w$  are realized in almost harmonic potentials (27). Their density<sup>17</sup> can be expressed in terms of the variable  $\varepsilon = (\hbar\omega)^2$  as follows:

$$n_0(\varepsilon) = \frac{\gamma}{a^3 \varepsilon_D} \left( \frac{\varepsilon}{\varepsilon_D} \right)^{3/2}, \quad \gamma = \frac{\bar{n}_0 a^3 \varepsilon_D^{5/2}}{32w^4}, \quad (31)$$

where  $\bar{n}_0$  is the density of states of the TLS, and is known from experiments. If the quantity  $\varepsilon$  is sufficiently large that, for the parameter values corresponding to it ( $\eta \lesssim \varepsilon/\varepsilon_D$  and  $t^2 \lesssim \varepsilon/\varepsilon_D$ ), we can no longer set  $P_0(\eta, t) = \text{const}$ , the dependence  $n_0(\varepsilon)$  turns out to be sharper than (31).

The features of interest to us in the behavior of  $n(\hbar\omega)$  lie at energies greater than or of the order of several tens of degrees Kelvin (see Fig. 1a), i.e., appreciably greater than the energy  $w$  (30). Therefore, in the following estimates we

use for  $n_0$  the formula (31) (taking into account the remark that follows it).

Another quantity that we need—the width  $\Gamma$  of a quasilocal state in a soft potential—has in the harmonic approximation the well known expression<sup>14,19</sup>

$$\Gamma \approx \pi \epsilon^2 g(\epsilon) \quad (32)$$

(to avoid confusion we stress that we are concerned with the property at zero temperature). Anharmonicity effects increase  $\Gamma$ . Their influence can be estimated by the method of Ref. 16, if we take into account that the potential (37) is written in the noninertial reference frame moving with the long-wavelength displacements  $u$  of the oscillating medium. Because of this, in an inertial reference frame there appears an interaction with the displacements:

$$\mathcal{H}_{int} = V(x - \xi u) - V(x), \quad (33)$$

where  $\xi$  is a dimensionless vector characterizing the spatial orientation of the quasilocal mode. The absolute value of  $\xi$  depends on the concrete structure of the center and can take values in the range 1–10. The term linear in  $xu$  in (33) corresponds to the harmonic approximation (32). Parametrically it is the principal term: All the other terms give contributions proportional to  $w^2/\epsilon < 1$  or to higher powers of this ratio. However, the anharmonic contributions contain large numerical coefficients. In addition, besides (33) there are anharmonic contributions from the interaction with the dilatation, of the form

$$\left(\frac{x}{a_0}\right)^2 \nabla u + \frac{x}{a_0} (\nabla u)^2 + \dots,$$

which also contain  $w^2/\epsilon$ . As a result, for  $\epsilon$  of the order of a few  $w^2$  the anharmonic contributions can still significantly exceed (32), and we must regard (32) as a strong underestimate of  $\Gamma$ .

From (23), (25), (31), and (32) follow the expressions

$$\xi_1 = \pi \gamma (\epsilon/\epsilon_D)^{3/2}, \quad \xi_2 = 3\pi \gamma (\epsilon/\epsilon_D)^{7/2}, \quad (34)$$

from which it can be seen that an extended region of strong scattering ( $\xi_1 \gtrsim 1$ ) exists and is described in the framework of our theory ( $\xi_2 \ll 1$ ) when  $\gamma \gg 1$ . The values of the parameters in the expression for  $\gamma$  (31) differ comparatively little for different glasses. Taking as typical parameters those of  $\alpha$ -SiO<sub>2</sub>:  $a^3 \sim 10^{-22}$  cm<sup>3</sup>,  $\hbar\omega_D \sim 340$  K (Ref. 20), and  $\bar{n}_0 \sim 10^{33}$  erg<sup>-1</sup> cm<sup>-3</sup> (Refs. 1,2), and setting  $w \sim 10$  K in accordance with (30), we find  $\gamma \sim 100$ . Then the characteristic energies defined by the conditions (4) and (5):

$$\hbar\omega_1 = \hbar\omega_D (\pi\gamma)^{-1/2}, \quad \hbar\omega_2 = \hbar\omega_D (3\pi\gamma)^{-1/2} \quad (35)$$

are  $\hbar\omega_1 \approx 0.3\hbar\omega_D$  and  $\hbar\omega_2 \approx 0.4\hbar\omega_D$ . It can be seen that on the frequency scale the strong-scattering region  $\omega_1 < \omega < \omega_2$  described by the theory turns out to be not too broad (although the parameters  $\xi_1$  and  $\xi_2$  of the theory vary within broad limits). We note that when the remarks made above are taken into account the estimates (35) for  $\hbar\omega_1$  and  $\hbar\omega_2$  must be regarded as slight overestimates.

#### 4. THE COHERENT-POTENTIAL APPROXIMATION. NUMERICAL CALCULATION

At the present time it is considered that the best approximation for the spectral characteristics of a disordered system is given by the coherent-potential method.<sup>12,21</sup> In this

method an effective potential  $\sigma(\epsilon)$  is assigned to each site of the system, so that the role of the site perturbations is played by the quantities  $U_\alpha - \sigma$ . The self-consistent condition for the determination of  $\sigma(\epsilon)$  is that the ensemble-average value of the  $t$ -matrix (9) is equal to zero:

$$\langle t \rangle = \left\langle \frac{U_\alpha - \sigma}{1 - (U_\alpha - \sigma)P(\epsilon - \sigma)} \right\rangle = 0, \quad (36)$$

i.e., the system behaves as effectively coherent (with characteristics depending on the energy variable  $\epsilon$ ).

The site Green function

$$P(\epsilon - \sigma) = \int_0^\infty d\epsilon' \frac{g(\epsilon')}{\epsilon - \epsilon' - \sigma(\epsilon)}, \quad (37)$$

appearing in (36), in turn, contains  $\sigma$ . Therefore, the expressions (36) and (37) actually represent a system of four equations (two equations for the real parts and two equations for the imaginary parts) with unknown quantities  $\text{Re}\sigma$ ,  $\text{Im}\sigma$ ,  $\text{Re}P$ , and  $\text{Im}P$  to be determined for each value of  $\epsilon$ . This system of integral equations has been solved numerically (by the gradient method; see, e.g., Ref. 22) for a potential  $U$  of the form (2) with bare distribution  $n_0(\epsilon)$  (31) and the Debye density of states  $g(\epsilon)$  (24) of the coherent medium. The solutions were found for 100 values of the energy variable  $\epsilon = 0.01N\epsilon_D$  ( $N = 1, 2, \dots, 100$ ). The relative error of the calculations did not exceed 10% at any of the energies

To calculate the density of states we used the expression

$$n(\epsilon) = \frac{1}{\pi} \text{Im} \left\langle P(\epsilon - \sigma) \left( 1 - \frac{dU/d\epsilon}{1 - (U - \sigma)P(\epsilon - \sigma)} \right) \right\rangle, \quad (38)$$

which follows from (11) with allowance for the condition (36). In Fig. 1b the calculated density of states is depicted in the form of the dependence of  $n(\hbar\omega) = n[\epsilon(\hbar\omega)]d\epsilon/d(\hbar\omega)$  on the phonon energy  $\hbar\omega = \epsilon^{1/2}$ .

It can be seen that the dependence  $n(\hbar\omega)$  found confirms the conclusions of the analytical analysis of Secs. 2 and 3. It also agrees qualitatively with the experimental curve in Fig. 1a. At the same time, there are noticeable differences between the dependences in Fig. 1a and Fig. 1b. In this connection we point out that the potential (2) used in the calculation performed corresponds to the harmonic approximation for the spectrum of an isolated defect. Allowance for anharmonicity and for the difference of  $n_0(\epsilon)$  from the dependence (31) should shift the peak on the curve of  $n(\hbar\omega)$  to lower energies (see Sec. 3). This could improve somewhat the agreement between the curves in Fig. 1a and Fig. 1b.

#### 5. CONCLUSION

The results obtained have a qualitative character. For systems with fluctuating elastic constants the theory establishes the existence of characteristic energies  $\hbar\omega_1$  and  $\hbar\omega_2$  separating the vibrational spectrum into regions of different characters. For  $\omega \ll \omega_1$  the scattering of phonons by quasilocal states is weak and phonons are well defined excitations. In the strong-scattering region  $\omega \gtrsim \omega_1$  phonon states attenuate over scales  $\lambda$  smaller than the phonon wavelength, and  $\lambda$  decreases rapidly with increase of  $\omega$ . At  $\omega \sim \omega_2$  the attenuation length  $\lambda$  is comparable to the lattice constant.

The results obtained lead to the following conclusions about the energy dependence of the density of vibrational states in typical glasses. For  $\hbar\omega \ll w \sim 10$  K the density

$n(\hbar\omega) \approx \text{const}$  is due to TLS<sup>1)</sup> in the two-well potentials (27). For  $\hbar\omega \gtrsim w$  the increase of  $n(\hbar\omega)$  corresponds to the increase of the density  $n_0(\hbar\omega)$  of isolated vibrations in the soft one-well potentials (27). This section extends to  $\hbar\omega_1 \sim 100$  K. In the interval between  $\hbar\omega_1$  and  $\hbar\omega_2$  the theory predicts a certain strengthening of the dependence  $n(\hbar\omega)$ . However, this interval turns out to be so narrow that the predicted strengthening is not manifested in practice. For  $\omega \gtrsim \omega_2$  the dependence  $n(\hbar\omega)$  flattens out, and then begins to decrease. This behavior is in qualitative agreement with the data of Fig. 1a. The approximate character of the theory means that we cannot count on quantitative agreement. Another reason for the discrepancies may be hidden in the spatial correlation of the site perturbations, which has not been taken into account by us at all.

The theory described leads to the conclusion that the behavior of  $n(\hbar\omega)$  has a qualitatively universal character, of the type depicted in Fig. 1, for noncrystalline systems. It is evident that the most reliable test of this conclusion would be a systematic investigation of inelastic neutron scattering. Such data are available, as yet, for not very many noncrystalline systems: SiO<sub>2</sub> (Ref. 3), polymers,<sup>4,5</sup> and metallic glasses.<sup>24</sup>

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## APPENDIX

To analyze the corrections to the averaged- $t$ -matrix approximation it is convenient to represent the  $t$ -matrix of the defect in the form  $t_\mu = t + \tau_\mu$  with  $\langle \tau_\mu \rangle = 0$ , and also to go over to Fourier components using the formula

$$A_{\mu\nu} = \sum_{\mathbf{k}\mathbf{q}} A(\mathbf{k}, \mathbf{q}) \exp(i\mathbf{k}\mathbf{r}_\mu - i\mathbf{q}\mathbf{r}_\nu). \quad (\text{A.1})$$

Expanding the solution of Eq. (10) in powers of  $\tau_\mu$ , we obtain the following expression for a general term of the expansion:

$$\tilde{T}^{(i+1)}(\mathbf{k}, \mathbf{q}) = \mathcal{G}(\mathbf{k}) \sum_s \tau(\mathbf{k}-\mathbf{s}) \tilde{T}^{(i)}(\mathbf{s}, \mathbf{q}), \quad (\text{A.2})$$

where  $\mathcal{G}(\mathbf{k}) = g(\mathbf{k})/(1 - tg(\mathbf{k}))$ , in which  $g(\mathbf{k})$  is a Fourier component of the quantity  $G_{\alpha\gamma}^0(1 - \delta_{\alpha\gamma})$ . Taking into account that

$$\tilde{T}^0(\mathbf{k}, \mathbf{q}) = \delta_{\mathbf{k},\mathbf{q}} \mathcal{G}(\mathbf{k})/g(\mathbf{k}),$$

we find that each term of the series can be represented in the form of a diagram of the type depicted in Fig. 2a. Here, each line corresponds to a propagator  $\mathcal{G}(\mathbf{k})$ , the circle on the right corresponds to the quantity  $g^{-1}(\mathbf{q})$ , and a cross corresponds to a Fourier component  $\tau(\mathbf{k}_i - \mathbf{k}_{i+1})$  of the random  $t$ -matrix. Here it is necessary to sum over all the internal momenta.

For concreteness we shall estimate that part of the cor-

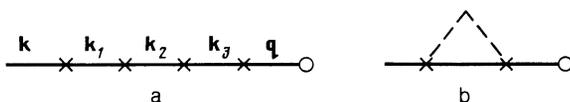


FIG. 2.

rection to the density of states which is described by the imaginary part of  $\text{Sp}G[\equiv \tilde{T}\text{r}G]$ . It is not difficult to convince oneself that

$$\text{Sp} \delta G = \sum_{\mathbf{q}} (G_{e\mathbf{q}}^0)^2 \langle T \rangle_{\mathbf{q}},$$

where  $\delta G = G - G^0$ . We have also taken into account that after averaging over the ensemble of oscillators the quantity  $\langle T_{\alpha\beta} \rangle$  can depend only on the difference  $\mathbf{r}_\alpha - \mathbf{r}_\beta$ . Taking into account the relationship between  $T_{\alpha\beta}$  and  $\tilde{T}_{\alpha\beta}$ , we find that the correction to the bare density of states is given by a sum of two average values:

$$t \langle \tilde{T}(\mathbf{q}, \mathbf{q}) \rangle + \left\langle \sum_s \tau(\mathbf{q}-\mathbf{s}) T(\mathbf{s}, \mathbf{q}) \right\rangle.$$

The two terms are estimated in a similar manner, and therefore we shall consider the graphs for the first term. An  $n$ th order graph contains the general factor  $t\mathcal{G}^2(\mathbf{q})/g(\mathbf{q})$  and a part containing  $n$  crosses and  $n - 1$  propagators that depend on the intermediate momenta over which it is necessary to integrate. This quantity must be averaged over the ensemble of the random quantities  $\tau$ . The averaging procedure can be depicted by linking the crosses into bundles. Each bundle contains more than one cross ( $\langle \tau \rangle = 0$ ). In addition, a bundle corresponds to momentum conservation, eliminating one integration over the intermediate momenta. The averaged- $t$ -matrix approximation that we have used contains one cross, and the lowest correction to it is depicted by the graph of Fig. 2b.

Because of the point character of the scattering it is natural to regard the quantities  $\tau$  as independent of the momentum transfer. As a result, the ratio of the given correction to that which has been taken into account amounts to

$$\frac{\langle \tau^2 \rangle}{t} \sum_{\mathbf{k}} \mathcal{G}(\mathbf{k}) = - \frac{\langle \tau^2 \rangle}{t^2} (P\Sigma)^2. \quad (\text{A.3})$$

Assuming that  $\langle \tau^2 \rangle \sim t^2$  and taking into account the expression for  $\Sigma$ , we arrive at the conclusion that the correction (A.3) is small for  $\xi_2 \ll 1$ . Analysis of the next orders of perturbation theory shows that for  $\xi_2 \ll 1$  the main role is played by graphs with nonintersecting bundles and with the smallest number of crosses in a bundle. Such graphs contain the smallest number of integrations, and  $\mathcal{G}(\mathbf{k})$  for small  $\mathbf{k}$  has the form  $\mathcal{G}(\mathbf{k}) = (\varepsilon - \varepsilon_{\mathbf{k}} - t)^{-1}$ . From this it is easy to see that the higher corrections are small in the parameter (A.3).

If, however,  $\xi_2 \gtrsim 1$ , all graphs, with an arbitrary number of bundles, are of the same order and the theory constructed no longer works. Thus, the condition for applicability of the theory constructed is the inequality  $\xi_2 \ll 1$  (which does not exclude the region  $\xi_1 \gtrsim 1$ ).

<sup>1)</sup>At very low energies an important role can be played by the effects of interaction between the TLS, and these have been considered in a recent paper by Maleev.<sup>23</sup>

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