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Vibrational instability, two-level systems, and the boson peak in glasses

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We show that the same physical mechanism is fundamental for two seemingly different phenomena such as the formation of two-level systems in glasses and the boson peak in the reduced density of low-frequency vibrational states $g(\omega)/\omega^2$. This mechanism is the *vibrational instability* of weakly interacting harmonic modes. Below some frequency $\omega_c \ll \omega_0$ (where ω_0 is of the order of Debye frequency), the instability, controlled by the anharmonicity, creates a new stable *universal* spectrum of harmonic vibrations with a boson peak feature as well as double-well potentials with a wide distribution of barrier heights. Both are determined by the strength of the interaction $I \propto \omega_c$ between the oscillators. Our theory predicts in a natural way a small value for the important dimensionless parameter $C = \bar{P}\gamma^2/\rho v^2 \approx 10^{-4}$ for two-level systems in glasses. We show that $C \approx (W/\hbar\omega_c)^3 \propto I^{-3}$ and *decreases* with increasing interaction strength I. The energy W is an important characteristic energy in glasses and is of the order of a few Kelvin. This formula relates the two-level system's parameter C with the width of the vibration instability region ω_c, which is typically larger or of the order of the boson peak frequency ω_b. Since $\hbar\omega_c \gtrsim \hbar\omega_b \gg W$, the typical value of C and, therefore, the number of active two-level systems is very small, less than 1 per 1×10^7 of oscillators, in good agreement with experiment. Within the unified approach developed in the present paper, the density of the tunneling states and the density of vibrational states at the boson peak frequency are interrelated.

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I. INTRODUCTION

One of the most typical low-temperature properties of glasses is the existence of two-level systems (TLS's) which determine at low temperatures, typically below a few Kelvin and frequencies less than 1 GHz, phenomena such as specific heat, thermal conductivity, ultrasonic and microwave absorptions, and many others (such as the echo).[1] Another remarkable universal property of almost all glasses is the boson peak in the low-frequency inelastic scattering, proportional to the reduced density of the vibrational states $g(\omega)/\omega^2$. Compared to the TLS's, it is observed at much higher frequencies, between 0.5 and 2 THz, and persists to higher temperatures, sometimes up to the glass transition temperature T_g. Usually these two important glassy features are considered separately, and no definite connection between them has been established so far.

The purpose of the present paper is to propose a physical picture in which these two seemingly different phenomena are closely interrelated. We will show that the formation of the boson peak in the reduced vibrational density of states (DOS) inevitably leads to the creation of two-level systems (and vice versa). A mechanism of the boson peak formation, implying also the formation of TLS's was proposed in our recent papers.[2,3] This mechanism was based on the phenomenon of *vibrational instability* of weakly interacting harmonic oscillators (HO's). The instability takes place in the low-frequency region, $0 \leq \omega \lesssim \omega_c$, below some characteristic frequency ω_c which is proportional to the interaction strength I. By changing the interaction strength, one can vary the width of the instability region, the position of the boson peak, and the number of TLS's. The weakness of the interaction I implies that $\omega_c \ll \omega_0$, where ω_0 is of the order of Debye frequency. Thus the vibrational instability occurs far below the Debye frequency ω_0 and has little influence on the high-frequency vibrations.

The low-frequency harmonic oscillators we are speaking about are realized in glasses as *quasilocal* vibrations (QLVs), which can be understood as *local* low-frequency vibrations bilinearly coupled to the sound waves.[4,5] The existence of quasilocal vibrations in glasses has been confirmed in a number of papers (see, e.g., the literature cited in Ref. 2). The local low-frequency vibrations are the cores of the QLV's. The cores represent collective low-frequency vibrations of small groups of atoms.[6,7] If one plots the potential energy against the amplitude of one of these modes,[8,9] one obtains a mode potential, as treated in the soft-potential model.[10,11] The vibrational instability results from the weak interaction I of these soft oscillators with high-frequency oscillators (with $\omega \simeq \omega_0$). As a result of this harmonic instability and the anharmonicity of the glass, the initial vibrational density of states $g_0(\omega)$ is reconstructed in the low-frequency region $0 \leq \omega \lesssim \omega_c$, where instability takes place and shows the *universal* boson peak feature.

The microscopic origin of these high-frequency oscillators, whose frequencies are much higher than that of the

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boson peak, is not important in this picture. It has been shown that, in general, the frequency of the sound waves can pass through the Ioffe-Regel limit near the boson peak frequency.[8,12–15] Therefore, the higher frequency modes might have a rather complex structure. They interact with the soft oscillators. This interaction causes the vibrational instability and hence the formation of the boson peak and TLS's.

The vibrational instability is a rather general phenomenon and occurs in any system of bilinearly coupled harmonic oscillators. It can be considered in a purely harmonic approximation. For example, a system of two oscillators with masses $M_{1,2}$ and frequencies $\omega_{1,2}$ becomes unstable if the interaction I between the oscillators exceeds a critical value[2] $I_c = \omega_1\omega_2\sqrt{M_1M_2}$. If one of the two frequencies is small, then the critical interaction I_c is also small. In our case, we have such an instability due to the interaction of low- and high-frequency oscillators. Therefore, we can simplify the picture to treat it analytically. The physical reason for the instability in this case is the fact that the high-frequency modes adiabatically follow the motion of the low-frequency oscillator (a description of this fact can be given within the *adiabatic approximation*). The squared frequency of the soft oscillator is reduced by an amount proportional to the strength of the interaction squared and, therefore, can even turn negative. For the example of two interacting oscillators with frequencies $\omega_1 \ll \omega_2$, the new frequency $\tilde{\omega}_1$ of the low-frequency oscillator is given by[2]

$$\tilde{\omega}_1^2 = \omega_1^2(1 - I^2/I_c^2). \tag{1.1}$$

It is important that it decreases to zero *linearly* with $I_c - I$ ($\tilde{\omega}_1^2 \propto I_c - I$ when $I \to I_c$).

This is the essence of the vibrational instability phenomenon in the general case. As a result, by switching on the interaction between the oscillators, we shift *all* low-frequency modes with $\omega \lesssim \omega_c$ toward the boundary point $\omega = 0$ and some of them will cross this point (so that the corresponding ω^2 turns negative). Therefore, in such a case [if there is no hard gap around zero in the initial density of states, $g_0(\omega)$] we will have a *constant* distribution of renormalized ω^2 around $\omega = 0$. From that we immediately get the right wing of the boson peak. Indeed, the local anharmonicity does not change this important property of uniform distribution of renormalized ω^2 around zero. One can show[2] that it restores the mechanical stability of the system by simply *reflecting* all the negative ω^2 values back to the positive ω^2 range [like in a mirror but with additional stretching factor of 2, which is obviously not important; see Eq. (2.16)]. The strength of the anharmonicity itself plays no role in this *mirror transformation*. Now a constant distribution of ω^2 around zero (on the positive side) obviously leads to a universal *linear law* for the density of states, $g(\omega) \propto \omega$ in the interval $0 \leqslant \omega \lesssim \omega_c$ [independent of the initial DOS $g_0(\omega)$]. In turn, this linear ω dependence of the reconstructed DOS just gives us the right wing of the boson peak, since $g(\omega)/\omega^2 \propto 1/\omega$ and this dependence is also universal and independent of the initial DOS $g_0(\omega)$.

We want to stress that this DOS transformation (due to the phenomenon of vibrational instability) is rather general and universal since any monotonous "traffic" of ω^2 from positive to negative values (due to interaction between the oscillators, or due to changing the temperature, pressure, etc.) always gives a constant distribution of ω^2 around zero. Therefore, the universal linear DOS, $g(\omega) \propto \omega$, and the corresponding universal right wing of the boson peak, $g(\omega)/\omega^2 \propto 1/\omega$, inevitably emerge as a result of this instability.

If the origin of the right wing of the boson peak looks somehow natural, the left wing of the boson peak appears as a result of the less obvious additional transformation of the linear DOS at smaller frequencies. The point is that when the anharmonicity restores the mechanical stability of the system, single-well potentials describing the unstable soft modes with negative ω^2 are replaced by double-well ones. This means that the effective potential energy of the glass in the direction of the local soft mode has two minima separated by a rather low barrier. Thus, in this scheme, the two-level systems are created simultaneously with the boson peak due to the same mechanism of vibrational instability. Besides their own high importance (the TLS's physics in glasses and beyond), these double-well potentials play also an important role in building the left wing of the boson peak. It can be explained as follows.

Due to bilinear interaction between the oscillators, $I_{ij}x_ix_j$, double-well potentials, with a particle vibrating in one of the wells and, therefore, having a nonzero average displacement $x_{i0} \neq 0$, create random *static* forces $f_i \approx I_{ij}x_{j0}$ acting on other oscillators. In a purely harmonic case, these linear forces would not affect the frequencies [and the linear density of states $g(\omega) \propto \omega$ would not change]. However, together with the local anharmonicity, the static forces create a universal *soft gap*, $g(\omega) \propto \omega^4$, in the linear density of states. This soft gap is a manifestation of the sea-gull singularity[11] (see also Ref. 6) predicted in the framework of the soft-potential model for glasses. Recently, it was shown that the ω^4 behavior of the DOS is, indeed, a universal feature in disordered systems for low-frequency bosonic excitations which are not Goldstone modes.[16]

The physical reason for this gap is very transparent. Due to anharmonicity, there is always a *blueshift* of the soft oscillator frequency under the action of the static force f. For small ω, this shift is proportional to $|f|^{1/3}$ and it is anomalously large for small forces. The density of states in the gap then can be estimated as follows:

$$g(\omega) \propto \int_0^{\omega} \omega_1 d\omega_1 \int_{-\infty}^{+\infty} \delta(\omega - af^{1/3}) df \propto \omega^4. \tag{1.2}$$

As a result, the random static forces (together with anharmonicity) effectively "push out" oscillators from the low-frequency range to higher frequencies, creating the universal soft gap, $g(\omega) \propto \omega^4$. One can also see this gap in the context of mechanical stability of the system, but of another kind. Due to local anharmonicity, small frequencies cannot survive in the system in the presence of random static forces. In some sense they are not stable even though random forces cannot transform single-well potentials into double-well ones.

The width of the ω^4 gap is of the order of the boson peak frequency $\omega_b \propto (\delta f)^{1/3}$, where δf is the width of the random force distribution $P(f)$. The boson peak frequency ω_b is typically smaller[17] than the characteristic frequency ω_c determining the width of the vibrational instability region,[2]

$$\omega_b \approx \omega_c \left[\frac{g_0(\omega_c)}{g_0(\omega_0)} \right]^{1/3} . \tag{1.3}$$

The strong inequality $\omega_b \ll \omega_c$ occurs if $g_0(\omega_c) \ll g_0(\omega_0)$, i.e., $g_0(\omega)$ is an increasing function of ω (since $\omega_c \ll \omega_0$). As a result, $g(\omega) \propto \omega^4$ in the interval $0 \leq \omega \lesssim \omega_b$. Frequencies higher than ω_b are only weakly affected by the static forces. Therefore, the linear DOS, $g(\omega) \propto \omega$ (and the right wing of the boson peak), which was created in the course of the vibrational instability, is conserved in the interval $\omega_b \lesssim \omega \lesssim \omega_c$. Since at small frequencies, $\omega \lesssim \omega_b$, we have the universal ω^4 gap in the density of states, the left wing of the boson peak takes also the universal form, $g(\omega)/\omega^2 \propto \omega^2$. As a result, in the region of the vibrational instability, $0 \leq \omega \lesssim \omega_c$, we have a universal behavior of the density of states $g(\omega)$ $[g(\omega) \propto \omega^4$ for $0 \leq \omega \lesssim \omega_b$, and $g(\omega) \propto \omega$ for $\omega_b \lesssim \omega \lesssim \omega_c]$ with the boson peak feature, independent of the initial DOS $g_0(\omega)$. At higher frequencies, above ω_c, we keep the initial DOS $g_0(\omega)$ almost undistorted.

The boson peak was the main topic of our previous papers.[2,3] In the present work, we shall concentrate on the two-level systems, i.e., the level splittings due to the tunneling through the barriers separating the two minima of the two-well potentials. However, this consideration is not independent of the boson peak since we will see that the main parameters of TLS's will be strongly interrelated with the parameters of the boson peak. Therefore, these two universal phenomena should be considered together.

In the standard tunneling model, the TLS's are often characterized by the so-called dimensionless tunneling strength C (Ref. 18),

$$C = \frac{\bar{P}\gamma^2}{\rho v^2}, \tag{1.4}$$

where $\bar{P}$ is the density of states of the TLS's, γ the deformation potential, ρ the mass density of the glass, and v the average sound velocity. The experimental value of C for different glasses is small and varies in a narrow band between 10^{-3} and 10^{-4}. In our theory, such small numerical values for C will emerge in a natural way.

Several authors[19–22] proposed that the approximate universality and smallness of C in glasses may be a consequence of the interaction between the TLS's. Roughly speaking, the idea was based on a mean-field approximation. The ith TLS produces at a distance r_i a deformation

$$\varepsilon_i \simeq \frac{\gamma_i}{\rho v^2 r_i^3}, \tag{1.5}$$

where γ_i is the deformation potential of the ith TLS. As the deformation is inversely proportional to r_i^3, the distribution function of the deformations in a glass is a Lorentzian with width $\delta\varepsilon$ proportional to the total concentration N of the TLS's:

$$\delta\varepsilon \simeq \frac{\gamma N}{\rho v^2}. \tag{1.6}$$

The energy E_i (the interlevel spacing) of each TLS changes under the deformation ε as

$$\delta E_i = \gamma_i \varepsilon, \tag{1.7}$$

and from Eq. (1.6), one sees that the energies E_i of the TLS's are distributed in the interval δE:

$$\delta E \simeq \frac{\gamma^2 N}{\rho v^2}. \tag{1.8}$$

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

$$n(E) \simeq \frac{N}{\delta E} \simeq \frac{\rho v^2}{\gamma^2}. \tag{1.9}$$

This is the result of a purely classical approach. In this approach, the dimensionless parameter C_{cl}

$$C_{\mathrm{cl}} \simeq \frac{n(E)\gamma^2}{\rho v^2} \simeq 1 \tag{1.10}$$

is of order unity rather than of the order of 10^{-4}–10^{-3}. This was the main difficulty of the theory outlined in Refs. [19–22] However, if one accounts for the quantum nature of tunneling, the situation is improved and the value of C is reduced strongly. To explain this on the qualitative level, we remind that the energy E of a TLS consists of two contributions, i.e., the classical asymmetry Δ and the quantum tunneling amplitude Δ_0: $E = \sqrt{\Delta^2 + \Delta_0^2}$. So far, we have disregarded the latter.

According to the standard tunneling model

$$\Delta_0 = \hbar\omega_0 \exp(-\lambda), \tag{1.11}$$

where λ is the tunneling parameter, distributed uniformly in the interval $\lambda_{\min} < \lambda < \lambda_{\max}$. Usually $\lambda_{\min}$ is taken to be about unity. Only TLS's with $\lambda \simeq 1$ can tunnel during typical experimental times. If $\lambda_{\max} \gg 1$, systems with $\lambda \simeq \lambda_{\max}$ cannot tunnel and do not contribute to the observable properties. Therefore, the relative number of TLS's participating in the tunneling is proportional to the small number $1/\lambda_{\max}$. As a result, we estimate the dimensionless parameter C in glasses as

$$C \simeq C_{\mathrm{cl}}/\lambda_{\max} \simeq 1/\lambda_{\max}. \tag{1.12}$$

If, for example, $\lambda_{\max} \simeq 10^3$, the dimensionless parameter $C \simeq 10^{-3}$. Thus the smallness of the dimensionless parameter C in our theory is related to typically large values of the tunneling parameter $\lambda_{\max}$ (and to typically high barriers in the system). We will discuss this point in Sec. IV.

In Sec. IV, we will show that two important parameters, namely, C for TLS's and the characteristic frequency ω_c for HO's, marking the onset of the vibrational instability are closely interrelated:

$$C \approx \left(\frac{W}{\hbar\omega_c}\right)^3. \quad (1.13)$$

Here, W is an important characteristic energy in glasses.[23] Typically, it is of the order of a few Kelvin. It determines, for example, the position of the minimum in the reduced specific heat,[6,11] $C(T)/T^3$ ($W \approx 2T_{\min}$), and some other properties of glasses above 1 K.[24] For vitreous silica, $W \approx 4$ K. In particular, it follows from this formula that the larger is the interaction $I \propto \omega_c$ between the oscillators, the smaller is the TLS's parameter C: $C \propto 1/I^3$, and, as we will see in Sec. IV, the smaller will be the density of tunneling states, $\bar{P} \propto 1/I^4$. It naturally explains the very old puzzle in the physics of glasses, i.e., why the number of two-level systems is so small (one two-level system for a million of atoms).

At first glance, this interesting result seems to be rather contradicting. The stronger the interaction I between the oscillators, the larger is the width of the vibrational instability region $\omega_c \propto I$ and, therefore, the higher is the number of double-well potentials created in the course of stabilization of the system due to anharmonicity. The explanation of this seeming paradox is that majority of the double-well potentials created due to vibrational instability have so high barriers V that they cannot participate in tunneling at all. As a result, only a very small part of the double-well potentials contributes to the tunneling density of states, $\bar{P}$.

Since the experimental values of W and C are well known for many glasses, one can estimate from Eq. (1.13) the important characteristic energy $\hbar\omega_c$, which gives the width of the vibrational instability region in glasses,

$$\hbar\omega_c \approx W C^{-1/3}. \quad (1.14)$$

For example, for a-SiO$_2$ $W=4$ K and $C=3\times 10^{-4}$, giving $\hbar\omega_c \approx 60$ K. This falls into the boson peak region ($\hbar\omega_b \approx 70$ K). As a result, we see that, indeed, the boson peak is placed in the vibrational instability range.

II. VIBRATIONAL INSTABILITY

To illustrate the idea of a vibrational instability, we consider a cluster containing a low-frequency HO with frequency ω_1 surrounded by a large number, $s-1$, of HO's with much higher frequencies ω_j of the order of $\omega_0 \gg \omega_1$. Here, ω_0 is an order of magnitude estimate of the high frequencies. In glasses, it usually is of the order of the Debye frequency. Let n_0 be the total concentration of the HO's in the cluster and $g_0(\omega)$ the normalized initial density of states (DOS), i.e., the DOS of the HO neglecting their interaction,

$$g_0(\omega) = \frac{1}{s}\sum_{i=1}^{s} \delta(\omega - \omega_i). \quad (2.1)$$

Including the interaction between the HO's, the total potential energy of the cluster is

$$U_{\text{tot}}(x_1,x_2,\ldots,x_s) = \sum_i \frac{k_i}{2}x_i^2 - \frac{1}{2}\sum_{i,j\neq i} I_{ij}x_i x_j + \frac{1}{4}\sum_i A_i x_i^4,$$
$$A_i > 0. \quad (2.2)$$

Here, x_i are the generalized coordinates describing the vibrations of HO's, $k_i > 0$ are the quasielastic constants of noninteracting oscillators, and I_{ij} determines the bilinear interaction between the oscillators. To stabilize the system, we have added in this equation the anharmonic terms, $A_i x_i^4$ (with $A_i > 0$). The interaction strength is given by[12]

$$I_{ij} = g_{ij} J / r_{ij}^3, \quad J \equiv \Lambda^2/\rho v^2, \quad (2.3)$$

where $g_{ij} \simeq \pm 1$ accounts for the relative orientation of the HO's, r_{ij} is the distance between HO's, ρ is the mass density of the glass, and v is the sound velocity.

The interaction between the HO's is due to the coupling between a single HO and the surrounding elastic medium (the glass). This HO-phonon coupling has the form[25]

$$\mathcal{H}_{\text{int}} = \Lambda x \varepsilon, \quad (2.4)$$

where Λ is the coupling constant and ε is the strain. Introducing the masses of oscillators M_i, we have for the bare frequencies (neglecting the bilinear interaction) as usual

$$\omega_i = \sqrt{k_i/M_i}. \quad (2.5)$$

These bare frequencies enter Eq. (2.1) for the initial DOS $g_0(\omega)$. We will assume the characteristic strength of the bilinear interaction $I \simeq J n_0$ between the oscillators to be *much smaller* than the typical quasielastic constants, so that

$$|I| \ll M\omega_0^2 \equiv k_0 \simeq k_j, (j \neq 1), \quad (2.6)$$

where M is the typical mass of the HO's.

The equation of motion for the low-frequency oscillator is

$$M_1 \ddot{x}_1 = -k_1 x_1 + \sum_{j\neq 1} I_{1j} x_j - A_1 x_1^3, \quad (2.7)$$

and for the high-frequency ones

$$M_j \ddot{x}_j = -k_j x_j + \sum_{i\neq j} I_{ji} x_i - A_j x_j^3, \quad j \neq 1. \quad (2.8)$$

For a slow motion, one can set the acceleration term $M_j \ddot{x}_j = 0$ in Eq. (2.8). For $I \ll M\omega_0^2$, we have $x_j \ll x_1$ (see below). Therefore, we can neglect also the anharmonicity force term $-A_j x_j^3$ and the interaction terms $I_{ji} x_i$ ($i \neq 1$) between the high-frequency oscillators and get from Eq. (2.8)

$$x_j = (I_{j1}/k_j) x_1, \quad j \neq 1. \quad (2.9)$$

According to Eq. (2.6), we see that $x_j \simeq (I/M\omega_0^2) x_1 \ll x_1$. Inserting this value of x_j into Eq. (2.7), we finally get a reduced equation of motion of the low-frequency oscillator

$$M_1 \ddot{x}_1 = -(k_1 - \kappa) x_1 - A_1 x_1^3 = -\frac{dU_{\text{eff}}(x_1)}{dx_1}, \quad (2.10)$$

where

$$\kappa = \sum_{j \neq 1} \frac{I_{1j}^2}{k_j} \simeq \frac{I^2}{M\omega_0^2} \tag{2.11}$$

and the effective potential energy for the slow motion is

$$U_{\mathrm{eff}}(x_1) = \frac{1}{2}(k_1 - \kappa)x_1^2 + \frac{1}{4}A_1 x_1^4. \tag{2.12}$$

The physical origin for this reduction to a one-mode approximation is the adiabatic approximation, where the high-frequency modes adiabatically follow the slow low-frequency motion.[26] As a result, the interaction between the low- and high-frequency oscillators renormalizes the quasielastic constant k_1 for the low-frequency motion to the new effective value

$$k = k_1 - \kappa. \tag{2.13}$$

For $k_1 > \kappa$, the potential (2.12) is a one-well potential and the cluster of oscillators is stable; the equilibrium positions of all oscillators $x_i = 0$. For $k_1 < \kappa$, the renormalized quasielastic constant k is negative and the cluster is unstable. The effective potential (2.12) in this case is a symmetric double-well potential. This is what we call the vibrational instability. As a result of the instability, the low-frequency oscillator is displaced to one of the two new minima

$$x_{10} = \pm\sqrt{(\kappa - k_1)/A_1} = \pm\sqrt{|k|/A_1}, \tag{2.14}$$

while the displacements of the high-frequency ones are $x_{j0} = (I_{1j}/k_j)x_{10} \ll x_{10}$ $(j \neq 1)$ and are much smaller. The barrier height between the minima is

$$V = \frac{(\kappa - k_1)^2}{4A_1} = \frac{k^2}{4A_1}. \tag{2.15}$$

As follows from Eq. (2.12), the new lowest frequency of the system of s coupled oscillators is given by

$$\omega^2 = \begin{cases} (k_1 - \kappa)/M_1 = k/M_1, & \kappa < k_1 \\ 2(\kappa - k_1)/M_1 = 2|k|/M_1, & \kappa > k_1. \end{cases} \tag{2.16}$$

The first case, $\kappa < k_1$, corresponds to a vibration in the minimum of a one-well potential [Eq. (2.12)], while the second case, $\kappa > k_1$, corresponds to a vibration in either of the two wells of a double-well potential [Eq. (2.12)]. It is remarkable that for weak interaction $I \ll M\omega_0^2$ the strength of the anharmonicity A_1 does not enter the renormalized frequency (2.16).

Using the Holtsmark method,[27] we derived in our previous paper[2] the normalized distribution function of κ

$$\rho(\kappa) = \frac{1}{\sqrt{2\pi}} \frac{B}{\kappa^{3/2}} \exp\left(-\frac{B^2}{2\kappa}\right), \tag{2.17}$$

where

$$B = \frac{\pi}{3}\sqrt{\frac{\pi}{2}} \frac{Jn_0}{\sqrt{M}} \left\langle \frac{1}{\omega} \right\rangle_0 \equiv \omega_c \sqrt{M}. \tag{2.18}$$

Here, $Jn_0 \simeq I$, and $\langle 1/\omega \rangle_0 \simeq 1/\omega_0$ is the ω^{-1} moment of the normalized initial DOS, $g_0(\omega)$. This formula can serve as a definition of the *important characteristic quantities*

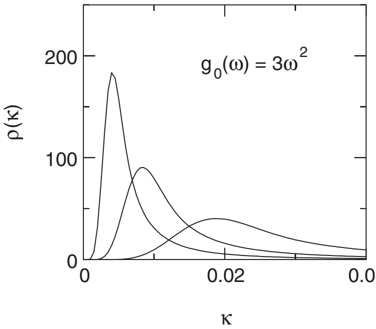

FIG. 1. Distribution function $\rho(\kappa)$ calculated as ensemble average by exact diagonalization of systems of $N = 2197$ oscillators with $g_0(\omega) = 3\omega^2$ and $J = 0.07$, 0.10, and 0.15 (from left to right).

$$\omega_c \simeq I/M\omega_0 \quad \text{and} \quad k_c \equiv M\omega_c^2 = B^2. \tag{2.19}$$

The physical meaning of these quantities is that the typical clusters with frequencies $\omega_1 \lesssim \omega_c$ become unstable due to the interaction between the soft oscillator and the surrounding high-frequency ones. Thus the characteristic frequency ω_c indicates the onset of the *mechanical instability region*. We will see below that the creation of TLS's and the formation of the boson peak occur in this region.

However, in the present paper, this particular form of the function $\rho(\kappa)$ is not suitable. The reason is the long-range power tail of this function, $\rho(\kappa) \propto 1/\kappa^{3/2}$ for $\kappa \gg k_c$. This tail leads to divergent integrals for large κ values when calculating averages of the type $\langle \kappa^\nu \rangle$ for $\nu \geq 1/2$. As follows from Eqs. (2.11) and (2.3), the long-range tail of the distribution is related to close pairs with small distances between the low- and high-frequency oscillators, $r_{ij} \ll n_0^{-1/3}$. However, usually the distance between the HO's in a glass cannot be arbitrarily small and, therefore, the function $\rho(\kappa)$ drops faster and approaches zero as $\kappa^{-(n+3)/2}$ [for $g_0(\omega) \propto \omega^n$, with $n > 0$]. In the following, we do not need the precise analytical form of this function. It will be sufficient to know that this function decays sufficiently rapidly for small and large κ with a characteristic scale $\kappa \simeq k_c \simeq I^2/k_0$. In Fig. 1, this function is shown for different interaction strengths J ($n_0 = M = \omega_0 = 1$, see Ref. 2 for details). Knowing the function $\rho(\kappa)$, we can calculate the distribution function of the renormalized quasielastic constants, $\Phi(k)$. Let $F(k_1)$ be a normalized distribution function of quasielastic constants k_i in Eq. (2.2). In the case of equal masses of the oscillators, $M_i = M$, it is related to the normalized initial DOS $g_0(\omega)$ as follows:

$$F(k) = \frac{g_0(\omega)}{2M\omega} \quad \text{where } k = M\omega^2, \tag{2.20}$$

and the normalized distribution function $\Phi(k)$ is given as

$$\Phi(k) = \langle \delta(k - k_1 + \kappa) \rangle_{k_1,\kappa} = \int_0^\infty dk_1 F(k_1) \int_0^\infty d\kappa \rho(\kappa) \delta(k - k_1 + \kappa). \tag{2.21}$$

Integrating over the delta function, it is convenient to present

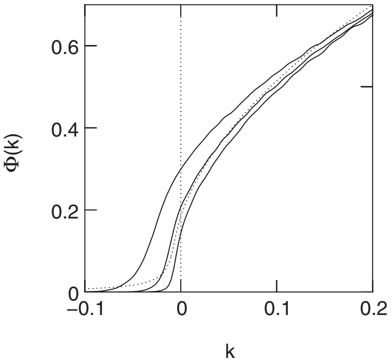

FIG. 2. Distribution function of the renormalized quasielastic constants, $\Phi(k)$, calculated as ensemble average by exact diagonalization of a systems of $N=2197$ oscillators with $g_0(\omega)=3\omega^2$ [$F(k)=3\sqrt{k}/2$] and $J=0.07$, 0.10, and 0.15 (solid curves, from right to left). Dotted curve: result of convolution [Eq. (2.21)] for $J=0.1$.

the expression for $\Phi(k)$ for positive and negative k separately. We have from Eq. (2.21)

$$\Phi(k) = \int_0^\infty dk_1 F(k_1)\rho(k_1+|k|) \quad \text{for } k<0 \tag{2.22}$$

and

$$\Phi(k) = \int_0^\infty d\kappa \rho(\kappa) F(k+\kappa) \quad \text{for } k>0. \tag{2.23}$$

Since the distribution function $\rho(\kappa)$ is nonvanishing only for $\kappa \lesssim k_c$ and rapidly drops to zero for $\kappa \gtrsim k_c$, we conclude from Eqs. (2.22) and (2.23) that the function $\Phi(k)$ for $|k| \ll k_c$ is approximately a constant

$$\Phi(k) \approx \Phi(0) = \int_0^\infty d\kappa \rho(\kappa) F(\kappa) \approx F(k_c) \quad \text{for } |k| \ll k_c. \tag{2.24}$$

For negative k and $|k| \gtrsim k_c$, the function $\Phi(k)$ rapidly drops. For positive $k \gtrsim k_c$, $\Phi(k) \approx F(k)$. In Fig. 2, this function is shown for different interaction strengths J ($n_0=M=\omega_0=1$, see Ref. 2 for details).

III. INTERACTION BETWEEN LOW-FREQUENCY OSCILLATORS

In the previous section, we have considered the effect of the interaction between a low-frequency oscillator and the surrounding high-frequency oscillators in a cluster. As a result of this interaction, the quasielastic constant k_1 has been renormalized to a new effective value $k=k_1-\kappa$. Negative values of k indicate a vibrational instability of the cluster. So far, we have neglected the interaction between low-frequency oscillators belonging to different clusters. This interaction is much weaker and cannot produce a new instability. However, it causes internal random static forces acting on the low-frequency oscillators. As we have seen in the previous section, in the case of instability, the interaction between low-frequency and high-frequency oscillators shifts the positions of their minima (static displacements). These shifts, in turn, act as forces if we take the interaction between unstable low-frequency HO's into account. As was shown in our previous paper[2] (see also Sec. V), these forces are responsible for the universal $g(\omega) \propto \omega^4$ (see also Ref. 16) dependence of the excess vibrational density of states for low frequencies.

One can insert these forces into Eq. (2.12) as a linear term $-fx_1$, where f is the internal random force created by the other unstable low-frequency oscillators. The effective potential energy then reads

$$U_{\mathrm{eff}}(x) = -fx + \frac{1}{2}kx^2 + \frac{1}{4}Ax^4. \tag{3.1}$$

Here and henceforth, the index 1 will be omitted.

The distribution function of the random forces $P(f)$ has been obtained in our previous paper[2] as

$$P(f) = \frac{1}{\pi}\frac{\delta f}{f^2 + (\delta f)^2}, \tag{3.2}$$

where δf is the width of the distribution. The Lorentzian form of the distribution is related to the fact that the forces between harmonic oscillators decay as r_{ij}^{-3} [see Eq. (2.3)].

One can estimate the width δf of the distribution as follows. The static force f_i exerted on the ith oscillator by the jth one is

$$f_i = I_{ij} x_{j0}. \tag{3.3}$$

Its characteristic value is given by displaced harmonic oscillators with frequencies of order ω_c. For these oscillators we have

$$I_{ij}^{(c)} \approx J n_c, \quad J \approx I/n_0 \approx M\omega_c\omega_0/n_0 \approx M\omega_c/n_0 g_0(\omega_0). \tag{3.4}$$

Here, $n_c \approx n_0 g_0(\omega_c)\omega_c$ is the concentration of these unstable harmonic oscillators (double-well potentials), while n_0 is the total concentration of HO's. Due to the normalization condition, $\omega_0 g(\omega_0) \approx 1$. From Eq. (2.14), it follows that the characteristic static displacement of these unstable oscillators is $x_{j0} \approx \sqrt{k_c/A} = \omega_c\sqrt{M/A}$. As a result, one gets from Eq. (3.3) (see Ref. 2)

$$\delta f \approx J n_c \omega_c \sqrt{\frac{M}{A}} \approx M\sqrt{\frac{M}{A}}\,\omega_c^3 \frac{g_0(\omega_c)}{g_0(\omega_0)}. \tag{3.5}$$

As mentioned already in the beginning of this section, these internal random forces do not produce a new vibrational instability. However, they can transform some double-well potentials into single-well ones. For $k>0$, the potential (3.1) is always one well, whereas for $k<0$, the potential (3.1) is double well for small forces $|f|<f_k^\star$, where

$$f_k^\star = 2|k|^{3/2}/3\sqrt{3A}. \tag{3.6}$$

For $|f|>f_k^\star$ (and $k<0$), Eq. (3.1) is a one-well potential. For $|f|=f_k^\star$ ($k<0$), the potential (3.1) is one well with a bending point.

It is interesting to compare the width of the distribution δf with the characteristic value of $f_k^\star$. They become equal for $|k| = k^\star \equiv M(\omega^\star)^2$, where

$$\omega^\star \approx \omega_c [g_0(\omega_c)/g_0(\omega_0)]^{1/3} \ll \omega_c. \tag{3.7}$$

The strong inequality $\omega^* \ll \omega_c$ occurs if both $\omega_c \ll \omega_0$ and $g_0(\omega_c) \ll g_0(\omega_0)$. We shall see in Sec. V that the frequency $\omega^\star$ plays the role of the boson peak frequency.

IV. TWO-LEVEL SYSTEMS

As follows from Eq. (3.1), negative values of k for $f=0$ correspond to symmetric double-well potentials. In a purely classical treatment, the oscillator will vibrate in either of the wells. Taking quantum mechanics into account, there will be a finite probability of penetration through the barrier separating the two wells, i.e., there is a finite tunneling probability. This causes a splitting of the vibrational levels. We are interested in the lowest pair of levels. This constitutes a TLS. These systems are ubiquitous in glasses and determine their low-temperature properties.[1]

Tunneling systems can be described effectively in terms of a tunnel splitting Δ_0 and an asymmetry Δ. We will derive expressions for these quantities and their distributions. Neglecting the linear force term in Eq. (3.1), the tunnel splitting is given in the Wantzel-Kramers-Brillouin approximation as

$$\Delta_0 \approx W \exp(-S/\hbar),$$

$$S = \int_{-x_0}^{x_0} |p| dx = 2\int_0^{x_0} \sqrt{2M[U_{\text{eff}}(x) + V]} dx. \tag{4.1}$$

Here, we approximated the dependence of the prefactor on the vibrational frequency by an order of magnitude estimate W (see also Ref. 23)

$$W = \frac{\hbar}{2}\left(\frac{\hbar A}{2M^2}\right)^{1/3} \tag{4.2}$$

that is of the order of the interlevel spacing in a purely quartic potential $V(x) = Ax^4/4$. From experiment, using the soft-potential model,[6,10,23–25,30] one finds values W for different glasses of the order of a few Kelvin, e.g., for vitreous silica, $W \approx 4$ K. The positions of the minima in the symmetric double-well potential $U_{\text{eff}}(x)$ (for $f=0$) are denoted by $\pm x_0 = \pm\sqrt{|k|/A}$ and $V = k^2/4A$ is the barrier height [see Eqs. (2.14) and (2.15)].

Evaluating the integral in Eq. (4.1), we get

$$S = \frac{2\sqrt{2}}{3}\frac{|k|^{3/2}M^{1/2}}{A} \tag{4.3}$$

and

$$\Delta_0 = W \exp\left(-\frac{2\sqrt{2}}{3}\frac{|k|^{3/2}M^{1/2}}{\hbar A}\right) = W \exp\left(-\frac{\sqrt{2}}{24}\frac{\hbar^3 |k|^{3/2}}{M^{3/2}W^3}\right). \tag{4.4}$$

The second quantity characterizing the TLS is the asymmetry of the two-well configuration. For $|f| \ll f_k^\star$, we have from Eq. (3.1)

$$\Delta = 2 f x_0 = 2f\sqrt{|k|/A}. \tag{4.5}$$

We are interested in the two quantum states with the lowest energies. These states belong to both wells. TLS's are often described by the interlevel distance E and the dimensionless tunneling parameter p:

$$E = \sqrt{\Delta_0^2 + \Delta^2}, \quad p = (\Delta_0/E)^2. \tag{4.6}$$

The Jacobian of the transformation from the variables $|k|$ and f to E and p is

$$\mathcal{J} = \frac{\partial(|k|,f)}{\partial(E,p)} = \left(\frac{2}{9}\right)^{1/3} 2^{5/2}\frac{M^{3/2}W^{5/2}}{\hbar^3}\frac{L^{-2/3}}{p\sqrt{1-p}}$$

$$\text{where } L = \ln\frac{W}{E\sqrt{p}}. \tag{4.7}$$

In the new variables, the distribution function reads

$$F(E,p) = n_0 P(0)\Phi(0)|\mathcal{J}| = \left(\frac{2}{9}\right)^{1/3} 2^{5/2} n_0 P(0)\Phi(0)\frac{M^{3/2}W^{5/2}}{\hbar^3}\frac{L^{-2/3}}{p\sqrt{1-p}}. \tag{4.8}$$

Here, we have replaced $P(f)$ and $\Phi(k)$ by $P(0)$ and $\Phi(0)$. This can be justified by estimating the relevant ranges of f and $|k|$ given by Eq. (4.11). Using Eqs. (4.5), (4.4), and (3.5), we express f in terms of Δ_0 and Δ:

$$\frac{f}{\delta f} \approx 4\left(\frac{2}{9}\right)^{1/6} L^{-1/3}\frac{E\sqrt{1-p}}{W}\left(\frac{W}{\hbar\omega_c}\right)^3 \frac{g_0(\omega_0)}{g_0(\omega_c)}. \tag{4.9}$$

Taking rough estimates $L=10$, $E=1$ K, $W=4$ K, $\hbar\omega_c = 100$ K, $\omega_0/\omega_c = 3$, and $g_0(\omega) \propto \omega^2$, one finds a typical value $f/\delta f \approx 2 \times 10^{-4}$. To estimate $|k|$, we derive from Eq. (4.4)

$$|k|/k_c = 4(9/2)^{1/3} L^{2/3} (W/\hbar\omega_c)^2. \tag{4.10}$$

This set of parameters gives the typical value of $|k|/k_c \approx 0.05$. Therefore, in the range of parameters where the notion of TLS's is applicable, the characteristic values of f and $|k|$ satisfy the conditions

$$f \ll \delta f \quad \text{and } |k| \ll k_c. \tag{4.11}$$

Our result can be compared with the standard tunneling model,[1] where the distribution functions are

$$P(\Delta, \Delta_0) = \frac{\bar{P}}{\Delta_0}, \quad F(E,p) = \frac{1}{2}\frac{\bar{P}}{p\sqrt{1-p}}, \tag{4.12}$$

with a constant density of tunneling states $\bar{P}$. Comparing these distributions with Eq. (4.8), one gets

$$\bar{P} = \left(\frac{2}{9}\right)^{1/3} 2^{7/2} n_0 P(0)\Phi(0)\frac{M^{3/2}W^{5/2}}{\hbar^3} L^{-2/3}. \tag{4.13}$$

Both distributions, Eqs. (4.8) and (4.12), coincide regarding their dependencies on E and p, apart from the factor $L^{-2/3}$ describing a weak logarithmic dependence on E and p. The same factor is found in the soft-potential model (see Refs. 10 and 11).

To compare the tunneling strengths of the TLS's with experiment, we study their interaction with strain, described by the deformation potential γ. According to Eq. (2.4), the variation of asymmetry Δ_ε due to a strain ε is

$$\Delta_\varepsilon = 2\Lambda x_0 \varepsilon = 2\Lambda\varepsilon\sqrt{|k|/A}. \tag{4.14}$$

The deformation potential is defined as

$$\gamma = \frac{1}{2}\frac{\partial\Delta_\varepsilon}{\partial\varepsilon}, \tag{4.15}$$

and from Eqs. (4.14), (4.4), and (4.2) follows

$$\gamma = \Lambda\sqrt{\frac{|k|}{A}} = \frac{3^{1/3}}{2^{7/6}}\frac{\hbar\Lambda}{\sqrt{MW}}L^{1/3}. \tag{4.16}$$

In the standard tunneling model, the TLS's are often characterized by the dimensionless tunneling strength C, given by Eq. (1.4). For different glasses, its value varies between 10^{-3} and 10^{-4}. Using Eqs. (4.13) and (4.16), one gets

$$C = 2\sqrt{2}n_0 P(0)\Phi(0)\frac{\Lambda^2}{\rho v^2}\frac{W^{3/2}\sqrt{M}}{\hbar}. \tag{4.17}$$

This value is independent of E and p as in the standard tunneling model.

The different factors entering the expression for C can be estimated from our model as

$$\Lambda^2/\rho v^2 = J \approx I/n_0, \quad I \approx M\omega_c\omega_0,$$

$$\Phi(0) \approx F(k_c) = g_0(\omega_c)/2M\omega_c, \tag{4.18}$$

and from Eqs. (3.2), (3.5), and (4.2),

$$P(0) = \frac{1}{\pi\delta f} \approx \frac{4}{\pi}\frac{W^{3/2}}{\hbar^2\sqrt{M}}\frac{g_0(\omega_0)}{\omega_c^3 g_0(\omega_c)}. \tag{4.19}$$

As a result, we arrive at the important estimate

$$C \approx \frac{4\sqrt{2}}{\pi}\left(\frac{W}{\hbar\omega_c}\right)^3 \tag{4.20}$$

that is independent of the initial DOS of HO's $g_0(\omega)$ and in this sense is universal. It only depends on the characteristic energy W and the frequency ω_c that is proportional to the interaction I. The larger the interaction between the original oscillators, the smaller is C, $C \propto I^{-3}$.

In the discussion of the boson peak (Sec. V), we will see that ω_c is two or three times larger than the boson peak frequency ω_b (or $\omega^\star$), which slightly depends on the initial DOS $g_0(\omega_c)$ [see Eqs. (5.10) and (5.11) and Fig. 3]. Using values appropriate to SiO$_2$, W=4 K and $\hbar\omega_c$=100 K, we get

$$C \approx 10^{-4}. \tag{4.21}$$

Thus the unified approach of this paper gives a value of the tunneling strength C in good agreement with experiment. Since values of C and W are well known from experiment for many glasses,[23] Eq. (4.20) can be used to estimate the important characteristic energy $\hbar\omega_c$ giving the onset of the vibrational instability in glasses:

$$\hbar\omega_c \approx WC^{-1/3}. \tag{4.22}$$

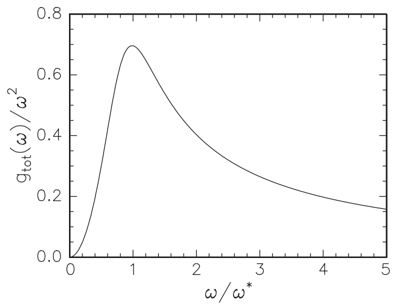

FIG. 3. The boson peak: the reduced density of states $g_{\mathrm{tot}}(\omega)/\omega^2$ given by Eq. (5.18).

It was demonstrated in Ref. 13 that by taking into account the experimental data for C and W, this energy is, indeed, correlated with the position of the boson peak in glasses, $\omega_b \simeq \omega_c$.

The two factors entering C [Eq. (1.4)] can be estimated separately as

$$\bar{P} \approx \frac{n_0}{\hbar\omega_0}\left(\frac{W}{\hbar\omega_c}\right)^4 L^{-2/3}, \quad \frac{\gamma^2}{\rho v^2} \approx \frac{\hbar\omega_0}{n_0}\frac{\hbar\omega_c}{W}L^{2/3}. \tag{4.23}$$

The first of these quantities is $\propto\hbar^{1/3}$ and the second $\propto\hbar^{2/3}$, and thereby, $C\propto\hbar$. Since $C \ll 1$ is a dimensionless quantity, it can be represented in the form $C=\hbar/\tilde{S}$, where $\tilde{S} \gg \hbar$ is some classical action. From Eqs. (4.20), (4.3), and (4.2), we get

$$C \approx \frac{1}{3\pi}\frac{\hbar}{S_c} \ll 1, \quad S_c = \frac{2\sqrt{2}}{3}\frac{|k_c|^{3/2}M^{1/2}}{A}, \tag{4.24}$$

i.e., $\tilde{S} \approx 3\pi S_c$. The classical action S_c corresponds to a typical double-well potential (3.1) with $|k|=k_c$ (and f=0).

Using the estimates $\omega_c/\omega_0 \approx 1/3$, W=4 K, and $\hbar\omega_c$=100 K, we estimate the concentration of tunneling systems with energies in the range $0<E<W$ and tunneling parameter $p \simeq 1$ as

$$n_{\mathrm{TLS}} \simeq \bar{P}W \simeq n_0\frac{\omega_c}{\omega_0}\left(\frac{W}{\hbar\omega_c}\right)^5 L^{-2/3} \approx 3\times 10^{-8}n_0. \tag{4.25}$$

The number of active TLS's is, thus, less than 1 for 1×10^7 oscillators. This explains why the concentration of observed TLS's in glasses is so small. Since according to Eq. (4.25) $n_{\mathrm{TLS}} \propto I^{-4}$, the number of TLS's decreases rapidly with increasing interaction strength I.

It would be instructive to derive a dimensionless parameter C_{cl} by a *classical* procedure (neglecting the tunneling probability Δ_0). We take the width of the force distribution from Eq. (3.5) and estimate the typical asymmetry Δ_c from Eq. (4.5):

$$\delta f \approx I^{(c)}x_0^{(c)} \approx Jn_c\sqrt{k_c/A}, \quad \Delta_c \approx \delta f\sqrt{k_c/A} \approx Jn_c(k_c/A). \tag{4.26}$$

With n_c the concentration of double-well potentials, we get the classical estimate for their density of states

$$\bar{P}_c \approx \frac{n_c}{\Delta_c} \approx \frac{A}{Jk_c} \approx \frac{n_0}{\hbar\omega_0}\left(\frac{W}{\hbar\omega_c}\right)^3 \tag{4.27}$$

that is independent of $\hbar$.

For the deformation potential γ_c, we have from Eqs. (4.14) and (4.15) the estimate

$$\gamma_c \approx \Lambda\sqrt{k_c/A} \quad \text{and} \quad \frac{\gamma_c^2}{\rho v^2} \approx J\frac{k_c}{A} \approx \frac{\hbar\omega_0}{n_0}\left(\frac{\hbar\omega_c}{W}\right)^3, \tag{4.28}$$

also independent of $\hbar$. Finally,

$$C_{\mathrm{cl}} = \frac{\bar{P}_c\gamma_c^2}{\rho v^2} \approx 1, \tag{4.29}$$

i.e., the dimensionless parameter C_{cl} in this classical approach is of the order of unity, which is a consequence of the $1/r^3$ interaction between the TLS's.[19–22] We wish to emphasize that C_{cl} (unlike C) does not determine any physical property of glasses.

The reason for the difference between the two approaches (quantum and classical) is the following. In the classical approach, we take all the double-well potentials into account. They have typically $|k| \simeq k_c$. Their concentration n_c is unimportant since it is canceled in Eq. (4.27) for $\bar{P}_c$. In the quantum approach, only the small portion of TLS's which are able to tunnel (they have $|k| < < k_c$) contribute to the observable quantities. For all other TLS's, the high barriers V and asymmetries $\Delta \sim \Delta_c$ prevent tunneling (but they contribute to the internal random static force δf).

To further clarify this point, let us consider TLS's with a quasielastic constant k in the interval $\Delta k \simeq k$, where $|k| \ll k^\star \ll k_c$ [see Eq. (3.7)]. Their concentration n_k, asymmetry Δ_k, and deformation potential γ_k are given by

$$n_k \approx n_0|k|\Phi(0)(f_k^\star/\delta f), \quad \Delta_k \approx f_k^\star\sqrt{|k|/A}, \quad \gamma_k = \Lambda\sqrt{|k|/A}. \tag{4.30}$$

In this expression for n_k, we took into account the fact that for $|k| \ll k^\star$, only the small fraction of all potentials, where $f_k^\star/\delta f \ll 1$, is of double-well type. Keeping in mind that $\Phi(0) \approx F(k_c)$, we get the density of states

$$\bar{P}_k \approx \frac{n_k}{\Delta_k} \approx \frac{n_0|k|F(k_c)}{\delta f\sqrt{|k|/A}} \tag{4.31}$$

and the parameter C_k

$$C_k = \frac{\bar{P}_k\gamma_k^2}{\rho v^2} \approx \frac{n_0F(k_c)J|k|^{3/2}}{\delta f\sqrt{A}} \approx \left(\frac{|k|}{k_c}\right)^{3/2} \ll 1. \tag{4.32}$$

Here, we have used Eq. (4.26) for δf and the estimate $n_c \simeq n_0k_cF(k_c) \simeq n_0\omega_cg_0(\omega_c)$.

If we now fix $|k|$ by the condition that the exponent in Eq. (4.4) is of the order of unity, i.e., $|k| \simeq MW^2/\hbar^2$, we reproduce our quantum result [Eq. (4.20)] $C_k \approx (W/\hbar\omega_c)^3$. The classical result C_{cl} [Eq. (4.29)] would be recovered for $k \approx k_c$ when $C_k \approx 1$. We conclude that the physical reason for smallness of the parameter C for TLS's in glasses is the scarcity of those TLS's that are able to tunnel compared to their total number.

Again, we can compare our results with the standard tunneling model.[1] In this model, the tunneling amplitude $\Delta_0 = \hbar\omega_0\exp(-\lambda)$ and the dimensionless parameter λ is uniformly distributed in the interval $\lambda_{\min} < \lambda < \lambda_{\max}$. The lowest value $\lambda_{\min} \simeq 1$. According to Eq. (4.1), $\lambda = S/\hbar$ and, therefore, the maximal value $\lambda_{\max} \approx S_c/\hbar$. Taking into account Eq. (4.24), we get

$$\lambda_{\max} \approx S_c/\hbar \approx 1/3\pi C. \tag{4.33}$$

Thus $\lambda_{\max}$ is related to the small parameter C. For SiO$_2$, $C = 3 \times 10^{-4}$ and $\lambda_{\max} \approx 350$.

V. BOSON PEAK

In this section, we relate the results obtained for the TLS's to the boson peak properties (see Ref. 2). For this we calculate the vibrational DOS $g(\omega)$. We start from the case $f=0$ (i.e., neglecting the interaction between the clusters). There are two types of harmonic vibrations. In the one-well case, $k>0$, according to Eq. (2.24) the distribution function of k for $k \ll k_c$ is constant, $\Phi(k) \approx \Phi(0)$. Therefore, since according to Eq. (2.16) (top) $k = M\omega^2$, the renormalized DOS for $\omega \ll \omega_c$ is

$$\tilde{g}_{\mathrm{I}}(\omega) = 2n_0M\Phi(0)\omega. \tag{5.1}$$

For harmonic vibrations in either well of a symmetric double-well potential [Eq. (3.1), $k<0$ and $f=0$] $|k| = M\omega^2/2$ [see Eq. (2.16), bottom] and $\Phi(k) = \Phi(0)$ for $|k| \ll k_c$ ($\omega \ll \omega_c$), the DOS is

$$\tilde{g}_{\mathrm{II}}(\omega) = n_0M\Phi(0)\omega. \tag{5.2}$$

It is half of the one-well contribution. The total DOS for $f=0$ and $\omega \ll \omega_c$ is the sum of the two contributions

$$\tilde{g}_{\mathrm{tot}}(\omega) = \tilde{g}_{\mathrm{I}}(\omega) + \tilde{g}_{\mathrm{II}}(\omega) = 3n_0M\Phi(0)\omega. \tag{5.3}$$

It is a *linear* function of ω, independent of the form of the initial DOS $g_0(\omega)$. This linear behavior follows from the finite value of $\Phi(0)$.

If the low-frequency HO's were isolated, their density of states would be determined by Eq. (5.3). As we have shown in Sec. III, there is, however, an interaction between these oscillators which we have to take into account. According to Eq. (3.3), the low-frequency harmonic oscillators, displaced from their equilibrium positions (and forming the double-well potentials), create long-range random static forces f acting on other oscillators. In a purely harmonic case, these linear forces would not affect the frequencies. Anharmonicity, however, renormalizes the low-frequency part of the spectrum, a manifestation of the so-called sea-gull singularity treated in detail in Ref. 11 (see also Ref. 6).

We begin with the case $k>0$. It corresponds to one-well potentials. Consider an anharmonic oscillator under the action of a random static force f. The effective potential is given by Eq. (3.1), where $\sqrt{k/M}$ is the oscillator frequency in the harmonic approximation for $f=0$. The force f shifts the equilibrium position from $x=0$ to $x_0 \neq 0$, given by

$$Ax_0^3 + kx_0 - f = 0, \tag{5.4}$$

where the oscillator has a new (harmonic) frequency

$$M\omega_{\text{new}}^2 = k + 3Ax_0^2. \tag{5.5}$$

With $\Phi(k)$ as the distribution function of k [see Eq. (2.23)] and $P(f)$ as the distribution of random forces f [see Eq. (3.2)], the renormalized DOS is given by

$$g_{\text{I}}(\omega) = n_0 \int_0^{\infty} \Phi(k) dk \int_{-\infty}^{\infty} df P(f) \delta(\omega - \omega_{\text{new}}). \tag{5.6}$$

Assuming $\omega \ll \omega_c$ and integrating Eq. (5.6) with $\Phi(k) = \Phi(0)$, we get the integral

$$g_{\text{I}}(\omega) = 2n_0\Phi(0)\frac{M^2\omega^3}{\sqrt{3A}} \int_0^{M\omega^2} dk \frac{P[f(k)]}{\sqrt{M\omega^2 - k}}, \tag{5.7}$$

where, according to Eqs. (5.4) and (5.5),

$$f(k) = Ax_0^3 + kx_0 = \frac{1}{3}\sqrt{\frac{M\omega^2 - k}{3A}}(2k + M\omega^2). \tag{5.8}$$

Taking the Lorentzian distribution [Eq. (3.2)] for $P(f)$ and introducing a new variable $t=\sqrt{1-k/M\omega^2}$, we finally get

$$g_{\text{I}}(\omega) = \frac{12}{\pi} n_0 M \Phi(0) \frac{\omega^2}{\omega^{\star}} \left(\frac{\omega}{\omega^{\star}}\right)^2 \int_0^1 \frac{dt}{1 + (\omega/\omega^{\star})^6 t^2 (3 - 2t^2)^2} \tag{5.9}$$

with

$$\omega^{\star} = \sqrt{3}A^{1/6}(\delta f)^{1/3}/\sqrt{M}. \tag{5.10}$$

The function $g_{\text{I}}(\omega)$ depends on a single parameter $\omega^{\star}$, characterizing, as we will see below, the position of the boson peak ω_b ($\omega_b \approx \omega^*$). The frequency $\omega^{\star}$ is determined by the characteristic value of the random static force δf acting on a HO with the characteristic frequency ω_c. As a result, taking into account Eq. (3.5), we get the estimate

$$\omega^{\star} \approx \omega_c \left[\frac{g_0(\omega_c)}{g_0(\omega_0)}\right]^{1/3}, \quad \omega^{\star} \ll \omega_c. \tag{5.11}$$

Again, as in Eq. (2.16) in lowest order the anharmonicity A does not enter this formula. This equation for $\omega^{\star}$ coincides with Eq. (3.7) obtained from the condition $f_k^{\star} \simeq \delta f$.

According to Eq. (5.11), for weak interactions I ($\omega_c \ll \omega_0$), the frequency of the boson peak $\omega^* \ll \omega_c$ only in the case when the initial DOS, $g_0(\omega)$, is a monotonically (and rapidly) decreasing (to zero) function of ω. For example, we can take $g_0(\omega) \propto \omega^n$ with $n>0$. Then for $n=2$ and $\omega_c = \omega_0/3$, we have from Eq. (5.11) $\omega^* \approx \omega_c/2$. For the same n and smaller interaction, $\omega_c = \omega_0/5$, we get $\omega^* \approx \omega_c/3$. In the opposite case, if the initial DOS drops to zero too slowly or remains nearly constant, $g_0(\omega) \simeq \text{const}$, we have from Eq. (5.11) that $\omega^* \simeq \omega_c$. In this case, the boson peak frequency ω^* is of the same order as the characteristic frequency ω_c.

For small frequencies, $\omega \ll \omega^{\star}$, only small forces, $f \ll \delta f$, contribute to the integral in Eq. (5.7). In this case, the distribution function $P(f)$ can be approximated by a constant value, $P(0)$, and we get from Eqs. (5.7) and (5.9)

$$g_{\text{I}}(\omega) = 4n_0\Phi(0)P(0)\frac{M^{5/2}\omega^4}{\sqrt{3A}} = \frac{12}{\pi}n_0 M\Phi(0)\omega\left(\frac{\omega}{\omega^{\star}}\right)^3 \propto \omega^4. \tag{5.12}$$

As a result, at low frequencies, the renormalized excess DOS $g_{\text{I}}(\omega) \propto \omega^4$.[11,16] For sufficiently large frequencies, $\omega \gg \omega^{\star}$ (but still smaller than ω_c), the action of random static forces on the HO spectrum can be discarded. In this case, the integral in Eq. (5.9) is equal to $(\pi/6)(\omega^{\star}/\omega)^3$. We recover the linear DOS [Eq. (5.1)], $g_{\text{I}}(\omega) = 2n_0 M\Phi(0)\omega \propto \omega$.

For $k<0$, the effective potential energy including a random static force f is given by Eq. (3.1). The simple analysis in Sec. III shows that for sufficiently small force, $|f| < f_k^{\star}$, where $f_k^{\star}$ is given by Eq. (3.6), the potential (3.1) has two minima (double-well potential). For a large force, $|f| > f_k^{\star}$, the potential (3.1) has only one minimum (one-well potential), while for $f = f_k^{\star}$, the potential is a one-well potential with a bending point.

The calculation of the DOS for the lower minimum in the two-well case can be considered together with the one-well case ($k<0, f>f^{\star}$). The position x_0 of the minimum can be found from the equation

$$Ax_0^3 - |k|x_0 = f. \tag{5.13}$$

For $f>0$ and $f<0$, one should take, respectively, the positive and negative roots of this cubic equation. In this minimum, the oscillator has a harmonic frequency

$$M\omega_{\text{new}}^2 = -|k| + 3Ax_0^2. \tag{5.14}$$

The density of states can be calculated from the Eq. (5.6). For $\omega \ll \omega_c$,

$$g_{\text{II}}(\omega) = 2n_0\Phi(0)\frac{M^2\omega^3}{\sqrt{3A}} \int_0^{M\omega^2/2} d|k| \frac{P[f(|k|)]}{\sqrt{M\omega^2 + |k|}}, \tag{5.15}$$

where, according to Eqs. (5.13) and (5.14),

$$f(k) = Ax_0^3 - |k|x_0 = \frac{1}{3}\sqrt{\frac{M\omega^2 + |k|}{3A}}(M\omega^2 - 2|k|). \tag{5.16}$$

Comparing $f(k)$ with $f_k^{\star}$ given by Eq. (3.6), one can see that the region of integration in Eq. (5.15), $0 < |k| < M\omega^2/3$, corresponds to the case of one minimum [one-well potential, $f(k) > f_k^{\star}$] and region $M\omega^2/3 < |k| < M\omega^2/2$ corresponds to the case of two minima [double-well potential, $f(k) < f_k^{\star}$]. Taking into account Eq. (3.2) for $P(f)$ and introducing a new variable $t = \sqrt{1 + |k|/M\omega^2}$, we finally get from Eq. (5.15)

$$g_{\text{II}}(\omega) = \frac{12}{\pi} n_0 M\Phi(0) \frac{\omega^2}{\omega^{\star}} \left(\frac{\omega}{\omega^{\star}}\right)^2 \int_1^{\sqrt{3/2}} \frac{dt}{1 + (\omega/\omega^{\star})^6 t^2 (3 - 2t^2)^2}. \tag{5.17}$$

As follows from this equation, at small frequencies, $\omega \ll \omega^{\star}$, $g_{\text{II}}(\omega) \propto \omega^4$, and for moderately high frequencies satisfying the inequality $\omega^\star \ll \omega \ll \omega_c$, the integral in Eq. (5.17) is equal to $(\pi/12)(\omega^\star/\omega)^3$. Therefore, in this case, $g_{\mathrm{II}}(\omega) = n_0 M\Phi(0)\omega$, which coincides with Eq. (5.2).

Combining results (5.9) and (5.17), we get for the total DOS at $T=0$

$$\begin{aligned} g_{\mathrm{tot}}(\omega) &= g_{\mathrm{I}}(\omega) + g_{\mathrm{II}}(\omega) \\ &= \frac{12}{\pi} n_0 M\Phi(0)\frac{\omega^2}{\omega^\star}\left(\frac{\omega}{\omega^\star}\right)^2 \int_0^{\sqrt{3/2}} \frac{dt}{1+(\omega/\omega^\star)^6 t^2 (3-2t^2)^2}. \end{aligned} \tag{5.18}$$

We want to mention that Eq. (5.18) differs from the corresponding Eq. (22) of Ref. 2 not only by the prefactor and the upper limit but also by the power of $(3-2t^2)$ in the integrand. Thus we are correcting our error in Ref. 2. Fortunately, this does only marginally alter the plots of the quoted paper, where the analytical theory is compared to the results of simulation and experiment, since the plots of the present function $g_{\mathrm{tot}}(\omega)$ and the one given in Ref. 2 differ only slightly.

For $\omega \ll \omega^\star$, the integral in Eq. (5.18) is equal to $\sqrt{3/2}$ and we have

$$g_{\mathrm{tot}}(\omega) = \frac{12}{\pi}\sqrt{\frac{3}{2}} n_0 M\Phi(0)\omega\left(\frac{\omega}{\omega^\star}\right)^3 \propto \omega^4, \quad \omega \ll \omega^\star. \tag{5.19}$$

Taking into account Eq. (4.2), one can present the density of states as function of energy $E=\hbar\omega$ (for $E \ll E^\star \equiv \hbar\omega^\star$) in the following way:

$$n(E) = \frac{g_{\mathrm{tot}}(\omega)}{\hbar} = \frac{1}{\sqrt{2}} n_0 \Phi(0) P(0) \frac{M^{3/2}W^{5/2}}{\hbar^3}\left(\frac{E}{W}\right)^4. \tag{5.20}$$

This result for $n(E)$ can be compared with Eq. (4.8), giving the density of states for TLS's. It is clear from this comparison that for $E \gg W$, the density of states of HO's is much bigger than the density of states for TLS's. For large frequencies, $\omega^\star \ll \omega \ll \omega_c$, the integral in Eq. (5.18) is equal to $(\pi/4)(\omega^\star/\omega)^3$ and we have

$$g_{\mathrm{tot}}(\omega) = 3 n_0 M\Phi(0)\omega, \quad \omega^\star \ll \omega \ll \omega_c, \tag{5.21}$$

which coincides with Eq. (5.3).

Since at low frequencies, $\omega \ll \omega^\star$, the total DOS $g_{\mathrm{tot}}(\omega) \propto \omega^4$ and at high frequencies, $\omega^* \ll \omega \ll \omega_c$, $g_{\mathrm{tot}}(\omega) \propto \omega$, we have a peak in the reduced density of states $g_{\mathrm{tot}}(\omega)/\omega^2$ at $\omega \approx \omega^\star$, the boson peak. In Fig. 3, we plot the function $g_{\mathrm{tot}}(\omega)/\omega^2$. We see from this figure that $\omega_b \approx \omega^\star$. This figure is valid only for the case $\omega^* \ll \omega_c$. If $\omega^* \simeq \omega_c$ [$g_0(\omega) \approx \mathrm{const}$], then $\omega_b \simeq \omega_c$, and at $\omega \gg \omega_c$, according to our previous results [see Ref. 2, Eq. (27)], $g_{\mathrm{tot}}(\omega) \approx g_0(\omega) \approx \mathrm{const}$. In this case, the right wing of the boson peak is determined by the initial density of states $g_0(\omega)$ and $g_{\mathrm{tot}}(\omega)/\omega^2 \propto 1/\omega^2$ (instead of $g_{\mathrm{tot}}/\omega^2 \propto 1/\omega$ in the previous case).

The DOS for the higher minimum in the double-well potential (3.1) is different. Though the thermal occupation number of this minimum is smaller than that of the lower minimum, it can contribute to the total DOS at finite temperatures. Starting from Eq. (5.6), the position of the higher minimum can be obtained as the smallest negative root or largest positive root of Eq. (5.13) for $f>0$ and $f<0$, respectively. The resulting DOS for $\omega \ll \omega_c$ is

$$g_{\mathrm{III}}(\omega) = 2n_0\Phi(0)\frac{M^2\omega^3}{\sqrt{3A}}\int_{M\omega^2/2}^{\infty} d|k| \frac{P[f(|k|)]}{\sqrt{M\omega^2+|k|}} \tag{5.22}$$

with

$$f(k) = Ax_0^3 - |k|x_0 = \frac{1}{3}\sqrt{\frac{M\omega^2+|k|}{3A}}(2|k|-M\omega^2). \tag{5.23}$$

Inserting the Lorentzian distribution for $P(f)$ [Eq. (3.2)] and introducing a new variable $t=\sqrt{1+|k|/M\omega^2}$, we get for the DOS

$$g_{\mathrm{III}}(\omega) = \frac{12}{\pi} n_0 M\Phi(0)\frac{\omega^2}{\omega^\star}\left(\frac{\omega}{\omega^\star}\right)^2 \int_{\sqrt{3/2}}^{\infty} \frac{dt}{1+(\omega/\omega^\star)^6 t^2(3-2t^2)^2}. \tag{5.24}$$

At low frequencies, $\omega \ll \omega^\star$, the integral in Eq. (5.24) is equal to $(\pi/3)(2^{1/3})(\omega^\star/\omega)$ and

$$g_{\mathrm{III}}(\omega) = 2^{5/3} n_0 M\Phi(0)\omega^\star\left(\frac{\omega}{\omega^\star}\right)^3 \propto \omega^3, \quad \omega \ll \omega^\star. \tag{5.25}$$

We see that at low frequencies, the dependence of $g_{\mathrm{III}}(\omega)$ [Eq. (5.24)] differs from the ω^4 dependence; the DOS in the higher minimum is proportional to ω^3 in accordance with Ref. 16. From Eqs. (5.19) and (5.25), it follows that $g_{\mathrm{III}}(\omega)/g_{\mathrm{II}}(\omega) \approx \omega^\star/\omega \gg 1$ and, therefore, for $\omega \ll \omega^\star$ and equal population of both minima, the DOS in the higher minimum is larger than that in the lower minimum. However, as we will see below, including the thermal population factors for the two minima reverses this.

For high frequencies, $\omega \gg \omega^\star$, the integral in Eq. (5.24) is $(\pi/12)(\omega^\star/\omega)^3$ and

$$g_{\mathrm{III}}(\omega) = n_0 M\Phi(0)\omega, \quad \omega \gg \omega^\star, \tag{5.26}$$

which coincides with Eq. (5.2).

So far, we disregarded the thermal population factor. Taking it into account, we get a temperature weighted DOS

$$\begin{aligned} \tilde{g}_{\mathrm{III}}(\omega,T) &= 2n_0\Phi(0)\frac{M^2\omega^3}{\sqrt{3A}} \\ &\times \int_{M\omega^2/2}^{\infty} d|k| \frac{P[f(|k|)]}{\sqrt{M\omega^2+|k|}}\frac{1}{1+\exp(\Delta/T)}, \end{aligned} \tag{5.27}$$

where Δ is the energy difference between the minima.

In the low-frequency case, $\omega \ll \omega^\star$, the integral in Eq. (5.27) is a constant plus some ω-dependent correction. To estimate the constant, we set $\omega=0$ in the integral. The higher minimum then turns into a bending point and

$$\tilde{g}_{\mathrm{III}}(\omega,T) = 2n_0\Phi(0)\frac{M^2\omega^3}{\sqrt{3A}}\int_0^\infty \frac{d|k|}{\sqrt{|k|}}\frac{P(f_k^\star)}{1+\exp(\Delta/T)}, \tag{5.28}$$

where $\Delta = 3k^2/4A$ is the energy distance between the positions of the minimum and the bending point in the potential $U_{\mathrm{eff}}(x)$ for $f = f_k^\star$ [see Eq. (3.6)].

The result of the integration in Eq. (5.28) depends on which of the two functions $P(f_k^\star)$ or $[1+\exp(\Delta/T)]^{-1}$ decays faster with $|k|$. The function $P(f_k^\star)$ decays with a characteristic scale

$$|k| = k_f = 3A^{1/3}(\delta f)^{2/3}/2^{2/3} \tag{5.29}$$

(for $f_k^\star = \delta f$). The function $[1+\exp(\Delta/T)]^{-1}$ decays with a characteristic scale $|k| = k_T = 2\sqrt{AT/3}$. Both scales become equal at the temperature $T = T^\star$, where $T^\star$ is given by

$$T^\star = \frac{27}{8(2^{1/3})}\frac{(\delta f)^{4/3}}{A^{1/3}} = \frac{3}{8(2^{1/3})}\frac{M^2}{A}(\omega^\star)^4 = \frac{3}{128(2^{1/3})}\hbar\omega^\star\left(\frac{\hbar\omega^\star}{W}\right)^3. \tag{5.30}$$

Estimates show that $T^\star$ is rather large. For example, for $\hbar\omega^\star = 40$ K and $W = 4$ K, we have $T^\star \approx 740$ K.

Therefore, the low-temperature case is more realistic. For $T \ll T^\star$, one has $k_T \ll k_f$ so that $P(f_k^\star) = P(0)$ and, therefore, from Eq. (5.28)

$$\tilde{g}_{\mathrm{III}}(\omega,T) = \frac{2\sqrt{2}}{3^{3/4}}n_0\Phi(0)P(0)\frac{M^2\omega^3}{A^{1/4}}T^{1/4}\int_0^\infty \frac{dy}{\sqrt{y}}\frac{1}{1+e^y} \propto \omega^3 T^{1/4}. \tag{5.31}$$

The last integral in this equation is equal to $1.07 \approx 1$. Therefore, the equation can be rewritten in the form

$$\tilde{g}_{\mathrm{III}}(\omega,T) \approx \frac{(4\sqrt{2})(3^{3/4})}{\pi}n_0\Phi(0)M\left(\frac{\omega}{\omega^\star}\right)^3\frac{W}{\hbar}\left(\frac{T}{W}\right)^{1/4}. \tag{5.32}$$

Now let us compare $g_{\mathrm{tot}}(\omega)$ and $\tilde{g}_{\mathrm{III}}(\omega,T)$ for $\omega \ll \omega^\star$. Taking into account Eqs. (5.19) and (5.32), we have

$$\frac{g_{\mathrm{tot}}(\omega)}{\tilde{g}_{\mathrm{III}}(\omega,T)} \approx \frac{\hbar\omega}{W}\left(\frac{W}{T}\right)^{1/4}. \tag{5.33}$$

Thus for $T \ll W(\hbar\omega/W)^4$, we get $g_{\mathrm{tot}}(\omega) \gg g_{\mathrm{III}}(\omega)$. In the opposite case, the contribution of the higher minimum to the DOS dominates.

VI. RESONANT SCATTERING OF PHONONS BY HARMONIC OSCILLATORS

Taking Eq. (2.4) for the coupling of the quasilocal oscillators to the phonons, we get

$$l_{\mathrm{res,HO}}^{-1} = \frac{\pi\Lambda^2}{2M\rho v^3}g(\omega), \tag{6.1}$$

where $l_{\mathrm{res,HO}}$ is the mean free path of phonons due to resonant scattering on quasilocal HO's with a density of states $g(\omega)$. For low frequencies, below the boson peak frequency, $\omega \ll \omega^\star$, we have from Eq. (5.20) (see also Ref. 28)

$$l_{\mathrm{res,HO}}^{-1} = \frac{\pi}{8}\frac{C\omega}{v}\left(\frac{\hbar\omega}{W}\right)^3 \propto \omega^4, \tag{6.2}$$

where $C \simeq 10^{-3} - 10^{-4}$ is the TLS's dimensionless parameter given by Eqs. (1.4) and (4.17) [see also the estimate, Eq. (4.20)]. Its value is well known from the low-temperature properties of glasses. For high frequencies, above the boson peak, in the interval $\omega^\star \ll \omega \ll \omega_c$, we have from Eq. (5.21) $g(\omega) = 3n_0 M\Phi(0)\omega$. As a result,

$$l_{\mathrm{res,HO}}^{-1} = \frac{3}{2}\pi\frac{\Lambda^2 n_0}{\rho v^2}\Phi(0)\frac{\omega}{v} \propto \omega. \tag{6.3}$$

Let us compare the last quantity (proportional to ω) with the inverse wavelength of the phonons $\lambda^{-1} = \omega/2\pi v$. We have the ratio

$$\frac{\lambda}{l_{\mathrm{res,HO}}} = 3\pi^2\frac{\Lambda^2 n_0}{\rho v^2}\Phi(0). \tag{6.4}$$

Using the estimates

$$\Lambda^2 n_0/\rho v^2 \approx I \approx M\omega_c\omega_0, \quad \Phi(0) \approx F(k_c) = g_0(\omega_c)/2M\omega_c, \tag{6.5}$$

and $\omega_0 \simeq 1/g_0(\omega_0)$, we have

$$\frac{\lambda}{l_{\mathrm{res,HO}}} \approx \frac{3}{2}\pi^2\frac{g_0(\omega_c)}{g_0(\omega_0)} \approx \frac{3}{2}\pi^2\left(\frac{\omega_b}{\omega_c}\right)^3; \tag{6.6}$$

the last estimate follows from Eq. (5.11). Thus this ratio is a constant in the interval $\omega^* < \omega < \omega_c$ and depends only on the characteristic frequency $\omega_c \propto I$ and the behavior of the initial DOS $g_0(\omega)$. From the last equation, it follows that the ratio $\lambda/l_{\mathrm{res,HO}}$ depends on the cube of the ratio of two important frequencies, the boson peak frequency $\omega_b \approx \omega^*$ and the characteristic frequency ω_c. Both of them can be measured in experiment [see Eq. (4.22) and Ref. 13].

For the weak interaction I which we consider in the paper for $\omega_c \ll \omega_0$ and if for $\omega \to 0$ initial DOS $g_0(\omega)$ also goes to zero sufficiently rapidly, then $g_0(\omega_c) \ll g_0(\omega_0)$ and $\lambda \ll l_{\mathrm{res,HO}}$; i.e., resonant phonon scattering is also weak. However, due to the big numerical coefficient in Eq. (6.6), in some realistic cases we can have a strong phonon scattering. For example, if the initial DOS $g_0(\omega) \propto \omega^2$ and $\omega_c \approx \omega_0/3$, then

$$\lambda/l_{\mathrm{res,HO}} \approx \pi^2/6 \approx 1.64. \tag{6.7}$$

In this case, the criterion of Ioffe-Regel for the phonons ($l_{\mathrm{res,HO}} < \lambda$) is approximately satisfied and we have strong phonon scattering above the boson peak frequency in the interval $\omega^* \ll \omega \ll \omega_c$ (which is not too big since, in this case, $\omega_c \approx 2\omega^*$).

Another interesting case is a flat initial DOS, $g_0(\omega) \approx$ const, then $g_0(\omega_c) \simeq g_0(\omega_0)$ and, as follows from Eq. (5.11), $\omega^* \simeq \omega_c$ and interval $[\omega^*, \omega_c]$ shrinks to one point (boson peak frequency, $\omega_b \simeq \omega_c$), and at the boson peak frequency, we have

$$\frac{\lambda}{l_{\mathrm{res,HO}}} = \frac{3}{2}\pi^2 \approx 15. \tag{6.8}$$

In this case, the criterion of Ioffe-Regel is again satisfied and we have at the boson peak frequency, $\omega^* \simeq \omega_c$, the regime of *very strong resonant scattering* of phonons on quasilocal harmonic oscillators, independent of the strength of interaction I (and ω_c). However, it is necessary to stress that, in this case, the strong scattering takes place only in the vicinity of the boson peak frequency ω_c.

At higher frequencies, the initial DOS $g_0(\omega) \simeq$ const and, according to Eq. (6.1), the resonance phonon mean free path is also constant and independent of frequency. However, the phonon wavelength $\lambda \propto 1/\omega$ decreases with frequency. Therefore, the regime of the weak phonon scattering will recover again at higher frequencies $\omega \gg \omega_c$. Similar behavior was observed in Ref. 29 for resonant scattering of phonons on librational (quasilocal) modes in crystals. We can extend the regime of strong scattering to well above the boson peak frequency (up to Debye frequency ω_0) only when the initial DOS is a linear function of the frequency, $g_0(\omega) \propto \omega$, and interaction I is not too small, $\omega_c > \omega_0/15$ [see Eq. (6.6)].

As we already mentioned, we will have a weak resonant scattering of phonons on quasilocal oscillators only when the interaction I is sufficiently weak and the initial density of states $g_0(\omega)$ decreases to zero sufficiently fast with ω, so that $g_0(\omega_c) \ll (2/3\pi^2) g_0(\omega_0)$. In such a case, the mean free path of the phonons $l_{\mathrm{res,HO}}$ will be much larger than their wavelength λ in the whole frequency range. In this case, phonons are well defined quasiparticles everywhere.

We give here also the relaxation time τ of a HO with frequency ω due to the interaction with phonons. From Eq. (2.4), we get

$$\frac{1}{\tau} = \frac{\Lambda^2}{\rho v^2}\frac{\omega^2}{4\pi M v^3} = \frac{J^2\omega^2}{4\pi M v^3}. \tag{6.9}$$

Estimating

$$\Lambda \simeq \mathcal{E}_0/a, \quad \rho v^2 \simeq \mathcal{E}_0/a^3, \quad M v^2 \simeq \mathcal{E}_0, \quad \hbar v/a \simeq \hbar\omega_0, \tag{6.10}$$

where $\mathcal{E}_0 \simeq 10$ eV is of the order of atomic energy, $a \simeq 1$ Å is of the order of interatomic distance, and ω_0 is of the order of Debye frequency, we get

$$1/\omega\tau \simeq \omega/4\pi\omega_0 \ll 1. \tag{6.11}$$

Therefore, HO's with $\omega \ll \omega_0$ are well defined objects.

VII. DISCUSSION

In our previous papers,[2,3] we proposed a mechanism of the boson peak formation. The essence of the mechanism can be formulated as follows. A vibrational instability of the weakly interacting QLV's (stabilized by the anharmonicity) is responsible for the boson peak in glasses and other disordered systems. The instability occurs below some frequency ω_c proportional to the strength of the interaction I between low- and high-frequency oscillators. Whereas anharmonicity is essential in creating the atomic structures supporting the boson peak, the vibrations forming the peak in the inelastic scattering intensity or the reduced density of states are essentially harmonic.

The present paper extends these ideas. We show that such seemingly unrelated phenomena in glasses (typical for the glassy state and usually treated by separate unrelated models) such as the formation of the two-level systems and the boson peak in the reduced density of low-frequency vibrational states $g(\omega)/\omega^2$ can be explained by the same physical mechanism, namely, the vibrational instability of weakly interacting soft harmonic vibrations. These can be seen as localized vibrations with a bilinear interaction with the extended modes, the sound waves. The resulting exact harmonic eigenmodes are quasilocalized vibrations that have been observed in numerous computer simulations and have been discussed extensively (see Ref. 2 and the references therein).

The instability, which as in all solids is controlled by the anharmonicity, creates a new stable universal spectrum of harmonic vibrations with the boson peak feature as well as double-well potentials with a wide distribution of the barrier heights that is determined by the strength of the interaction I between the oscillators. Depending on the barrier height (and temperature), these will lead to tunneling and relaxational transitions. To check for the consistency of our theory, we calculated the dimensionless parameter $C = \bar{P}\gamma^2/\rho v^2 \approx 10^{-4}$ for the two-level systems in glasses, which is observed in experiment.[18] The smallness of this parameter is a long-standing puzzle. In our theory, it follows naturally. The physical reason for the small value of the parameter C is that only a small fraction of all created TLS's can actually tunnel in realistic time scales.

We show that the larger the interaction I between the original harmonic vibrations, the smaller the parameter C. It reminds us partly the ideas of Ref. 22 though we do not have here the frustrated strong interactions. We prove that for our simple model, weakly interacting oscillators, $C = (W/\hbar\omega_c)^3 \propto I^{-3}$. Here, W is an important characteristic energy in glasses of the order of a few Kelvin. The value of C is independent of the assumed initial DOS of HO's $g_0(\omega)$ and, in this sense, it is universal. Varying the characteristic energy W and interaction I (i.e., the characteristic energy $\hbar\omega_c$) for different glasses, C lies in the interval from 10^{-3} to 10^{-4}. However, we want to stress that we are not free in the choice of these two parameters. The energy W is well known from experiments on specific heat,[6,11] thermal conductivity,[24] and heat release[30] in glasses. As for the characteristic frequency ω_c, it should be of the order or larger than the boson peak frequency ω_b.

In the unified approach developed in the present paper, the densities of tunneling states and of excess vibrational states at the boson peak frequency are interrelated. Since the experimental values of C and W are well known for many glasses, we can use this formula to get the important energy $\hbar\omega_c = W C^{-1/3}$, giving us the onset of the vibrational instability region. For vitreous silica, $\hbar\omega_c \approx 60$ K, falling perfectly into the boson peak range. The same holds for many other glasses.[13] It indicates that the boson peak is, indeed, placed inside the vibrational instability range.

It is instructive to compare the results of the present paper and of Refs. 2 and 3 with our earlier paper,[12] where we also discuss the possible origin of the boson peak. In this paper we consider low-energy Raman scattering in glasses. As in the present paper and in Refs. 2 and 3, we assume that the scattering and the energy transfer are due to the interaction of the light with the soft potentials in glasses. The density of states of the quasilocalized HO's, according to Ref. 12, is proportional to ω^4 for low frequencies and to ω for high frequencies. This behavior qualitatively resembles the one obtained in Ref. 2. However, the phenomenon of vibrational instability was disregarded in Ref. 12 and the discussion of the boson peak was necessarily somewhat qualitative. Considering the vibrational instability puts the theory on a more quantitative level and, for instance, allows the determination of the shape of the boson peak.

Let us now compare the results of our paper with previously published important class of models of the boson peak.[31–34] Experiment has shown that the boson peak is formed by largely harmonic vibrations. Therefore, in all these models the authors have considered a Hamiltonian of the form

$$U_{\text{tot}}(x_1,x_2,\ldots,x_n)=\frac{1}{2}\sum_{i,j\neq i}k_{ij}(x_i-x_j)^2 \qquad (7.1)$$

with randomly distributed quasielastic constants k_{ij}. Since this potential energy is purely harmonic, we call all such models of the boson peak *harmonic random matrix* (HRM) models. The main difference between the quoted four HRM models is in the different distributions of the quasielastic constants k_{ij}.

If all quasielastic constants are positive, $k_{ij}>0$, then the corresponding dynamical matrix (Hessian) is positive-definite and, therefore, all the eigenvalues are obviously positive as well, $\omega_i^2>0$ $(i=1,2,\ldots,n)$ excluding those zeroes which come from the translational and rotational invariances. In such a case, the system is *mechanically stable*. As was shown in Refs. 31, 32, and 34, the system remains stable even when some (rather small) fraction of the quasielastic constants k_{ij} in Eq. (7.1) is negative (and small enough). By increasing the fraction of negative k_{ij} (or their absolute values), authors have approached the mechanical stability threshold (when the first negative ω^2 appears in the spectrum).

The reduced density of states $g(\omega)/\omega^2$ for Hamiltonian (7.1) usually has a maximum at some frequency $\omega_{\max}$ for typical values of $k_{ij}>0$. Changing the parameters of the distribution function $P(k_{ij})$, one can shift this maximum to higher or to low frequencies. The last case was the main goal of the papers.[31–34] The biggest redshift of the maximum has been achieved on approaching the mechanical stability threshold. In the first two papers,[31,32] the original maximum was due to a Van Hove singularity of the crystalline DOS. Atoms in these papers were placed on a perfect cubic lattice. As a result, the boson peak has been ascribed to the lowest Van Hove singularity shifted due to disorder. In the other two papers,[33,34] atoms were distributed randomly in three-dimensional space (so-called Euclidean random matrix models) and quasielastic constants k_{ij} depend exponentially on the interatomic distances $|\mathbf{r}_i-\mathbf{r}_j|$. Therefore, the distribution function of quasielastic constants in these two cases $P(k)\propto 1/k$ and has a singularity for $k\to 0$. Thereby the portion of small k was increased compared to Gaussian and box distributions used in the papers.[31,32] Obviously, the redshift of the original peak in $g(\omega)/\omega^2$ was much more pronounced in these latter two cases.[33,34]

Our approach, which continues from our previous papers,[2,3] differs essentially in two ways. First, we postulate that the excess in vibrational modes originates from quasilocalized vibrations. Their existence has been shown in numerous simulations of different types of materials. Such modes can be described as local modes (cores) which weakly interact bilinearly with the extended modes (sound waves) and, thus, with each other. The exact harmonic eigenvectors are, of course, extended as in HRM models. Secondly, we do not invoke special distributions for the elements of the dynamical matrix to avoid unstable vibrations but, on the contrary, show that the generic instability, when controlled by the anharmonicity which is present in all real systems, automatically gives both the TLS and the boson peak (with a universal shape) without any further assumptions.

The essence of the mechanism can be formulated as follows. The randomly distributed weakly interacting QLV becomes unstable at low frequencies in harmonic approximation. This is the equivalent to the instability in the general HRM models. Instead of assumptions on distribution functions of interactions I_{ij}, we use the always present anharmonicity as the stabilizing factor. The previous vibrational instability of the weakly interacting QLV's thus becomes responsible for the boson peak and TLS's in glasses and other disordered systems. Whereas anharmonicity is essential in creating the atomic structures supporting the boson peak, the vibrations forming the peak in the inelastic scattering intensity or the reduced density of states are essentially harmonic. Comparing with random matrix models our Eq. (2.2) without the anharmonicity term would correspond to the case of the unstable random matrix, whereas the result of Eq. (2.16) would correspond to the stabilized case, and, importantly, the anharmonicity strength A (thanks to mirror transformation) does not enter in the expression for the renormalized frequency. So, the anharmonicity reconstructs the spectrum, but the final result is independent of the strength of anharmonicity. The advantage of our approach is that the stabilization is not a result of an additional assumption, but is a benefit of the vibrational instability+anharmonicity which is haunting the alternative approach.

Summarizing briefly, we can say that the boson peak in the previous papers[31–34] was obtained by a purely harmonic ansatz inside the mechanically *stable* region of their Hamiltonians. The position and the form of the peak depend strongly on the distribution function of quasielastic constants $P(k_{ij})$. In our approach, the boson peak is built inside the mechanically *unstable* region of the harmonic potential parameters. Therefore, the role of anharmonicity as stabilizing factor is crucial. However, the form of the boson peak appears to be universal and independent of the initial assumptions about interaction I_{ij}, of the initial distribution function $g_0(\omega)$, or of the anharmonicity strengths A_i. Relating TLS's and boson peak parameters in our theory, we are able to show that the instability crossover frequency ω_c given by Eq.

(1.14) lies in the boson peak region. Due to the weakness of the interaction I, the universal reconstruction of the spectrum in our theory takes place in the low-frequency range only, leaving the high-frequency range nearly unchanged. This is different in the HRM models,[31–34] where the whole spectral range is completely reconstructed in the course of the change of the distribution function $P(k_{ij})$.

As mentioned before in our theory, the estimate for the dimensionless tunneling strength C emerges naturally [see Eq. (4.20)]. By varying the characteristic energy W and the interaction strength I for different glasses, C falls into the interval between 10^{-3} and 10^{-4}. We want to mention that other explanations for the smallness of the constant C have been proposed. Burin and Kagan[35] predicted a number of universal properties of amorphous solids, including the small values of the tunneling strength C due to a special form of interaction of such defect centers with internal degrees of freedom like the TLS's. It is important that the interaction between such centers falls off with distance r as $1/r^3$. In this case, effects of correlations between many TLS's (*dipole gap* effects) might be important at sufficiently low temperatures.

In our paper, we have neglected all such many particle correlation effects between tunneling TLS's, which could further reduce (or stabilize) the value of C. One should, however, keep in mind that the $1/r^3$ law of interaction is valid as far as the effects of retardation play no role. In dielectric glasses, these effects are determined by the sound velocity. At sufficiently large distances, they could play a role that would result in the variation of the interaction law. We believe that the role of the retardation effects deserves special investigation.

Another important difference between our approach and that of Burin and Kagan is that the parameter C in our theory is a quantum mechanical quantity, $C \propto \hbar$ [see Eq. (4.24)]. It disappears in the classical limit $\hbar \to 0$. In other words, the smallness of C is directly related to the smallness of the quantum mechanical probability for particles tunneling through high barriers in a glass [see also Eq. (4.33)]. Differently, the dipole gap effect is purely classical since it is based on the classical dipole-dipole interaction between TLS's. Therefore, it would be very interesting to elucidate which of the two mechanisms (or both) dominates and is responsible for the small value of parameter C in glasses. In particular, it would be very interesting to calculate the relaxation times for many particle correlation effects to build the dipole gap.

An analysis of the low-temperature properties of glasses along the lines of the soft-potential model based on a numeric simulation of a Lennard-Jones glass was presented by Heuer and Silbey.[36] They numerically searched for the energy minima of the glass and constructed double-well potentials for close minima. By extrapolation to small values, they were able to extract the distribution functions for the soft-potential parameters. These potentials correspond to our potentials after inclusion of the interaction between the HO's. The present theory is in agreement with their simulation results.

The theory presented in this paper deals with the effects of soft modes produced by disorder, which can be expected to have a broad frequency distribution. In the literature, the term boson peak is rather loosely defined. Often it is used for any low-frequency maximum in the reduced DOS. In particular in plastic crystals, see, e.g., Ref. 37, soft HO's are present even before disorder. Consequently, disorder only broadens their sharp DOS. Depending on the strength of this broadening, our theory will apply more or less to such cases. The same applies to TLS, which also can be present before disorder.

In summary, we have shown that the same physical mechanism is fundamental for such seemingly different phenomena as the formation of the two-level systems in glasses and the boson peak in the reduced density of low-frequency vibrational states $g(\omega)/\omega^2$. In this way, two of the most fundamental properties of glasses are interconnected.

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