Universality of Ultrasonic attenuation in amorphous systems at low temperatures

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Abstract

The competition between unretarded dispersion interactions between molecules prevailing at medium range order length scales and their phonon induced coupling at larger scales leads to appearance of nano-scale sub structures in amorphous systems. The complexity of intermolecular interactions gives rise to randomization of their operators. Based on a random matrix modelling of the Hamiltonian and its linear response to an external strain field, we show that the ultrasonic attenuation coefficient can be expressed as a ratio of two crucial length-scales related to molecular dynamics. A constant value of the ratio for a wide range of materials then provides a theoretical explanation of the experimentally observed universality of the ultrasonic attenuation coefficient at low temperatures.
I. INTRODUCTION

Theoretical quest to understand the universality of low temperature properties of amorphous systems has motivated many models during last few decades \[1 \text{--} 9\]. For a long time, the most promising one among them was believed to be the defects modeled as tunnelling two level systems (TTLS). Although successful in explaining many experimental observations, the original TTLS model \[1, 2\] suffered many drawbacks \[3, 8, 9\] (besides experimental lack of evidence supporting their existence in most glasses). This encouraged attempts for improvements of the model by incorporating a phonon-TTLS interaction as well as considerations of more generic defects. An important step in this direction was taken in a recent theory of coupled generic blocks with a phonon-mediated interaction of type \(1/r^3\) with \(r\) as the separation between blocks \[8, 10, 15\]. The theory has been previously applied to seek the theoretical origin of the universality of two properties, namely, ultrasonic attenuation coefficient \[10\] and Meissner-Beret ratio \[15\]. A renormalisation approach used in these studies rendered the information regarding the behavior of a single generic block unnecessary and provided useful insights regarding the universality at macroscopic scales. But it has still left many questions unanswered e.g how the block type structure appears, what is the effect of the intra-block forces over the inter-block ones, whether the universality is an emergent phenomenon occurring only at large scales or it also occurs at microscopic scales i.e for a single block; (for example, the study \[10\] does not provide any information about the attenuation coefficient for a basic block). An answer to these questions is pertinent to understand the physical origin of universalities which motivates the present work.

Based on the nature of chemical bonding, the physics of solids is expected to vary at microscopic length scales but an important property, namely, the ratio \(\lambda/l\), with \(\lambda\) as the wavelength of sound waves and \(l\) as the mean free path (related to the coefficient of ultrasonic attenuation), has been found to be quantitatively universal among a range of glasses \[10, 14\]. Contrary to other low temperature properties, this universality is applicable not only for glasses but for a huge class of materials different from them at large length scales e.g. disordered crystals, poly-crystals, some quasi crystals etc \[26\]. Furthermore the irradiation experiments on crystalline silicon for a wide range of radiation doses indicate the sound
properties of the irradiated samples similar to glasses. Another universality not confined only to glasses but applicable to many liquids too is that of excess vibrational density of states which can not be explained based on the phonon contributions only \[18\]. These universalities therefore seem to originate from more fundamental considerations, shared by both amorphous as well as disordered crystalline materials and applicable not only for macroscopic sizes but also at microscopic scales. This motivated us in \[16\] to consider the intermolecular interactions, more specifically VanderWaal forces among the molecules within a block as the basis for the behavior; it is important to emphasise here that VanderWaals forces among molecules are always present in all condensed phases and therefore are the natural candidates to decipher the experimentally observed universality.

Our primary focus in the present work is to seek the physical origin of the weak attenuation of the sound waves in amorphous systems. For this purpose, it is necessary to first identify the local structures which respond to an external strain field by collective vibrations of molecules. But phonons in a perfect harmonic dielectric crystal are free of interactions, leading to a sound wave travel unattenuated. To understand long mean free paths in glasses, this intuitively suggest to seek for ordered structure, at least locally, and repeated almost periodically. The structure related to medium range order (MRO) in glasses seem to be playing the relevant role. (Note the glasses also have short range order but that is governed by covalent bonds which are quite rigid to undergo deformation by a weak strain field). As shown later in the paper, the size of the basic block indeed turns out to be that of the length scale associated with medium range order. (Note the peculiarity of role played by MRO in context of acoustic modes was mentioned in \[19\] too; the study indicated that the continuum approximation for the medium, necessary for Debye formulation, breaks down for acoustic modes with wavelength less than MRO). The combination of many such blocks can then provide required periodicity and their long-range interaction result in attenuation only at long length scales. Our theory of coupled blocks is therefore based on two main types of interactions, dominant at different spatial scales; a competition between them governs the block-size and also gives rise to an inter-connected block structure, with phonon mediated coupling of their stress fields. This in turn leads to formulation of the attenuation in terms of the stress-stress correlations among basic blocks and their density of the states. As discussed later, both of them can be expressed in terms of the molecular properties which finally leads to a constant, system-independent average value of the coefficient of the
ultrasonic attenuation (internal friction), referred as $Q^{-1}$ later in the text.

The paper is organized as follows. The theory of an amorphous system of macroscopic size as a collection of sub-structures coupled with each other via an inverse-cube phonon mediated interaction is discussed in detail in [10, 15]; this is briefly reviewed in section II, with macroscopic solid referred as the super block and the sub-structures referred as the basic blocks. Note the present work differs from [10, 15] in context of the basic blocks details; the latter appear, in our theory, as a result of VanderWaal interactions among molecules prevailing at nano-scales [16]. The theory is used in section III to relate the ultrasonic attenuation coefficient of the super block to the stress-stress correlations of the basic blocks, their volume and the density of states. The latter depends on a parameter $b$, referred as "bulk spectral parameter" which was derived in [16] in terms of the molecular parameters; this is briefly reviewed in section IV. Section V compares the interactions between molecules at short length scales with those emerging due to collective molecular dynamics at large length scales and derives the formulation for the basic block size as a result of this competition. An important result obtained in this section is a constant value of the ratio $R_0/R_v$, with $R_0$ as the linear size of the basic block and $R_v$ as the interlayer distance. The ratio along with information of previous sections is used in section VI to calculate the average ultrasonic attenuation coefficient $\langle Q^{-1} \rangle$ and theoretically prove its quantitative universality; here $\langle \rangle$ refers to the ensemble as well as spectral average. However $\langle Q^{-1} \rangle$ can be calculated directly from the molecular properties too; as discussed in section VII, a good agreement of the results so obtained for 18 non-metallic glasses with experimental values not only lends credence to our theory of blocks but confirms the universality of the ratio $R_v/R_0$ too. Note the 18 glasses chosen for comparison here are same as those used in [20]. We conclude in section X with a summary of our main ideas and results.

II. SUPER BLOCK: PHONON MEDIATED COUPLING OF BASIC BLOCKS

The order at atomic dimensions in an amorphous solid is system dependent; it is sensitive to the nature of chemical bonding. The intuition suggests the universal properties to originate from the interactions which appear at length scales at which the solid manifests no system-dependence. It is therefore relevant to seek and identify the sub-units in the super block structure which give rise to such interactions. For this purpose, let us first express
The Hamiltonian $H$ of the amorphous solid of volume $\Omega$ as the sum over intra-molecular interactions as well as inter-molecular ones

$$H = \sum_{k} h_k(r_k) + \frac{1}{2} \sum_{k,l} U(|r_k - r_l|)$$

(1)

with $h_k$ as the Hamiltonian of the $k^{th}$ molecule at position $r_k$ and $U$ as an inter-molecular interaction with arbitrary range $r_0$. Assuming that all the relevant many body states are "localized", in the sense that the probability density for finding a given molecule "$k$" is "concentrated" (as defined by its mean square radius) in a region of finite radius $l$ around some point $r_k$, it is possible to define a 3D lattice (grid of points) $R_\alpha$ with spacing $d \gg r_0$ such that the molecule "$k$" is associated with that lattice point $R_\alpha$ which is closest to $R_k$. The association is fixed, is insensitive to the dynamics and corresponds to representation of the solid by 3-dimensional blocks of linear size $R_0$, with their centers at lattice points $R_\alpha$. The Hamiltonian $H$ can then be reorganised as a sum over basic block Hamiltonians and the interactions between molecules on different blocks

$$H = \sum_s \mathcal{H}^{(s)} + \frac{1}{2} \sum_{s,t} \sum_{k \in s, l \in t} U(|r_k - r_l|)$$

(2)

where $\mathcal{H}^{(s)}$ is the Hamiltonian of a basic block labeled "$s"", basically sum over the molecular interactions within the block : $\mathcal{H}^{(s)} = \sum_{k \in s} h_k(r_k) + \frac{1}{2} \sum_{k,l \in s} U(|r_k - r_l|)$. As mentioned below, the molecules interactions appearing in 2nd term in eq.(2) rearrange themselves collectively and results in emergence of coupled stress fields of the blocks. The number $g$ and volume $\Omega_b$ of these blocks can be determined by analysing the competition between inter-molecular forces with emerging forces i.e phonon mediated coupling: $g = \Omega / \Omega_b$ with $\Omega_b \sim R_0^3$. The statistical behavior of the Hamiltonian $\mathcal{H}$ is discussed in detail in [16].

To analyze the ultrasonic attenuation in glasses, we first need to analyze the response of $\mathcal{H}$ to an external strain field.

A. Perturbed Hamiltonian of a basic block

In presence of an external strain field, the molecules in a glass block are displaced from their equilibrium position and their interactions with those in surrounding blocks give rise to a stress field distributed over the block. Let $u(r)$ be the displacement, relative to some arbitrary reference frame, of the matter at point $r$, the elastic strain tensor can then be defined as
\[ e_{\alpha\beta}(r, t) = \frac{1}{2} \left( \frac{\partial u_\alpha}{\partial x_\beta} + \frac{\partial u_\beta}{\partial x_\alpha} \right) \]  

with subscripts \( \alpha, \beta \) referring to the tensor-components.

This gives rise to stress in the block which can in general have both elastic as well as inelastic components. The perturbed Hamiltonian \( H_{pt} \) of the basic block, labeled "s" can then be written as a sum over elastic and inelastic contributions

\[ H_{pt}^{(s)} = H_{pt, ph}^{(s)} + H_{pt, nph}^{(s)}. \]

Each of these parts can further be expanded as a Taylor’ series around unperturbed block Hamiltonian \( H_s \) in terms of strain \( e_{\alpha\beta} \) in long wavelength limit (where the subscript "x" refers to the elastic ("x = ph") and inelastic parts ("x = nph") respectively):

\[ H_{pt,x}^{(s)}(t) = H_x^{(s)} + \int \text{d}r \ e_{\alpha\beta}(r, t) \ \Gamma_{\alpha\beta;x}^{(s)}(r) + O(e_{\alpha\beta}^2) \]

with \( \Gamma_{\alpha\beta;x}^{(s)}(r) \) as the stress tensor; as clear from above \( \Gamma_{\alpha\beta;x}^{(s)}(r) = \frac{\partial H_{pt,x}^{(s)}}{\partial e_{\alpha\beta}} \). Further, assuming the isotropy and the small block-size, the distributed stress field within the block of volume \( \Omega_b \) can be replaced by an average acting from the centre of mass of the block: \( \int_{\Omega_b} \text{d}r \ \Gamma_{\alpha\beta}^{(s)}(r) = \Gamma_{\alpha\beta}^{(s)} \). The perturbed Hamiltonian of the basic block can then be approximated as

\[ H_{pt,x}^{(s)} = H_x^{(s)} + \sum_{\alpha\beta} e_{\alpha\beta}^{(s)} \ \Gamma_{\alpha\beta;x}^{(s)} \]

with \( e_{\alpha\beta}^{(s)}(t) \) referring to the phonon strain field \( e_{\alpha\beta}(r, t) \) at the s-th block.

**B. Super block Hamiltonian**

The super block consists of \( g \) basic blocks, perturbed by mutual interaction. To proceed further, it is useful to separate its Hamiltonian \( H \) into phononic and non-phonon contributions (referred by subscripts "ph" and "nph" respectively): \( H = H_{ph} + H_{nph} \) \((8)\). The contribution of elastic part \( H_{ph} \) to the ultrasonic attenuation in glass super block at temperatures \( T < 1^\circ K \) is negligible. We therefore need to consider the contribution from the inelastic part \( H_{nph} \) only; to reduce notational complexity, henceforth, the subscripts "nph" will be suppressed and the notations \( H, H_{pt}^{(s)}, \Gamma^{(s)} \) etc will be used for \( H_{nph}, H_{pt,nph}^{(s)}, \Gamma_{nph}^{(s)} \) respectively.
As the strain tensor $e_{\alpha\beta}$ contains a contribution from the phonon field, the exchange of virtual phonons will give rise to an effective (RKKY-type) coupling between the stress tensors of any two block-pairs. Let $\Gamma^{(s)}(r)$ be the stress tensor at point $r$ of the basic block "s". The interaction $V_{st}$ between the blocks "s" and "t" can be given as \cite{10}

$$V_{st} = \frac{1}{4\pi\rho_m c^2} \int_s dr \int_t dr' \sum_{te} \frac{\kappa^{(st)}_{\alpha\beta\gamma\delta}(r)}{|r - r'|^3} \Gamma^{(s)}_{\alpha\beta}(r) \otimes \Gamma^{(t)}_{\gamma\delta}(r') \tag{7}$$

with $\rho_m$ as the mass-density and $c$ as the speed of sound in the super block. Here the subscripts $\alpha\beta\gamma\delta$ refer to the tensor components and the symbol $\sum_{te}$ refers to a sum over all tensor components: $\sum_{te} \equiv \sum_{\alpha\beta\gamma\delta}$. The directional dependence of the interaction is represented by $\kappa^{(st)}_{\alpha\beta\gamma\delta}(\theta, \phi)$; it is assumed to depend only on the relative orientation $(\theta, \phi)$ of the block-pairs and is independent from their relative separation \cite{15}:

$$\kappa^{(st)}_{ijkl} = - (3n_j n_k \delta_{il} + n_i n_l \delta_{jk}) + \nu_2 \left[ -(\delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) + 3 (n_j n_l \delta_{ik} + n_i n_k \delta_{jl} + n_i n_l \delta_{jk} + n_2 n_3 \delta_{ik} + n_1 n_3 \delta_{il} + n_1 n_2 \delta_{jl} + n_2 n_3 \delta_{il}) - 15 \sum_{ijkl} n_i n_j n_k n_l \right] \tag{8}$$

where $\nu_2 = \left(1 - \frac{c_t^2}{c^2}\right)$ and $n = n_1 \hat{i} + n_2 \hat{j} + n_3 \hat{k}$ is the unit vector along the direction of position vector $r - r'$. Again assuming the isotropy and the small block-size, the interaction between various points of the block-pairs can be replaced by the average interaction between their centers $R_s$ and $R_t$. The phonon mediate coupling between the blocks can then be approximated as \cite{10,15}:

$$V_{st} = \frac{1}{4\pi\rho c^2} \sum_{\alpha\beta\gamma\delta} \frac{\kappa^{(st)}_{\alpha\beta\gamma\delta}(R_s - R_t)}{|R_s - R_t|^3} \Gamma^{(s)}_{\alpha\beta} \otimes \Gamma^{(t)}_{\gamma\delta} \tag{9}$$

Due to the above emerging interactions at large length scales, the super block Hamiltonian in eq.(2) is not just a sum over basic block Hamiltonians but also includes their phonon mediated coupling.

Eq.(9) describes an emerging interaction at large length scales. The Hamiltonian of the super block in eq.(2) can now be rearranged as a sum over those of the basic blocks as well as their phonon mediated coupling. In absence of external strain field, the non-phonon part of $H$ can be rewritten as

$$H = H_0 + V \tag{10}$$
with $H_0$ as a sum over non-phonon part of the unperturbed basic block Hamiltonians, 
$H_0 = \sum_{s=1}^{g} H^{(s)}$, and, $V$ as the net pair-wise interaction among blocks: $V = \sum_{s,t; s \neq t} V_{st}$
where $\sum_{s,t}$ implies the sum over all basic blocks. The presence of a weak external strain field perturbs the basic blocks and thereby $H$. The non-phonon part of the perturbed Hamiltonian $H_{pt}$ can be written as [10, 15]

$$H_{pt} = H + \sum_{s=1}^{g} \sum_{\alpha \beta} e^{(s)}_{\alpha \beta} \Gamma^{(s)}_{\alpha \beta} = H + \sum_{\alpha \beta} e_{\alpha \beta} \Gamma_{\alpha \beta}$$

(11)

where the 2nd equality follows by assuming the same strain operator for all blocks $e^{(s)}_{\alpha \beta} \approx e_{\alpha \beta}$ and writing $\Gamma_{\alpha \beta} = \sum_{s=1}^{g} \Gamma^{(s)}_{\alpha \beta}$. (Note, as discussed in [15], the total Hamiltonian for the super block contains two additional terms besides $V$ (see eq.(2.21) in [15]) but their ensemble averaged contribution is negligible. Alternatively it can also be absorbed by redefining stress operators).

III. ULTRASONIC ATTENUATION COEFFICIENT: RELATION WITH STRESS MATRIX

The dimensionless ultrasonic attenuation coefficient or internal friction $Q^{-1}_a(\omega)$ of a phonon of frequency $\omega$ and wavelength $\lambda$ can in general be defined as [15, 26]

$$Q^{-1}_a = \frac{1}{2\pi^2} \frac{\lambda}{l}$$

(12)

with $l$ as its mean free path. Note the above definition is different from that in [26] by a constant: $Q^{-1}_{a,pohl} = \pi Q^{-1}_a$.

Consider the attenuation of acoustic waves in a glass super block with its Hamiltonian $H$ given by eq.(11). Assuming the coupling between phonon and non-phonon degrees of freedom a weak perturbation on the phonon dynamics, $Q^{-1}_a(\omega)$ can be expressed as [15]

$$Q^{-1}_a(\omega) = (\pi \rho_m c_a^2)^{-1} \text{Im} \chi_a(\omega)$$

(13)

with $\rho_m$ as the mass-density of the material, $c_a$ as the speed of acoustic wave in the longitudinal (with $a = l$) or transverse direction ($a = t$). Here $\chi_{l,t}(\omega)$, referred as the longitudinal or transverse response function, are the measures of the linear response of the basic blocks to external strain field and can be defined as follows.
A. Non-phonon linear response function

Consider the linear response of a basic block, labeled as "s", to an external strain field $e_{ij}(r, t) = e_{ij} \exp[i(q \cdot r - \omega t)]$ with $e_{ij}$ real but infinitesimal. The perturbed Hamiltonian is given by eq.(6) with corresponding stress-field given as $\Gamma_{ij}(r, t) = \langle \Gamma_{ij} \rangle \exp[i(q \cdot r - \omega t)]$ where $\langle \Gamma_{ij}(s) \rangle$ is in general complex.

The complex response function or the susceptibility for a basic block can then be defined as

$$\chi^{(s)}_{\alpha\beta\gamma\delta}(q, \omega) \equiv \frac{1}{\Omega_b} \frac{\partial \langle \Gamma^{(s)}_{\alpha\beta}(q, \omega) \rangle}{\partial e_{\gamma\delta}}.$$  (14)

Here in general the variable $q$ and $\omega$ are independent variables. But as our interest is in values of $q$ close to $\omega/c_{l,t}$ (with $c_{l,t}$ as the longitudinal and transverse speeds of sound in the glass solid), $\chi$ will henceforth be written as a function of $\omega$ only [10].

The imaginary part of $\chi^{(s)}(\omega)$ can be written in the representation in which unperturbed basic block Hamiltonian $H^{(s)}$ is diagonal (later referred as non-interacting or NI basis). Let $|m_s\rangle$, $m_s = 1 \rightarrow N$ be the many body eigenstate of $H^{(s)}$ with energy $e_m$, then

$$\text{Im} \chi^{(s)}_{\alpha\beta\gamma\delta}(\omega) = \frac{(1 - e^{-\beta\omega})}{Z} \sum_m e^{-\beta e_m} \chi^{(m,s)}_{\alpha\beta\gamma\delta}(\omega)$$  (15)

with $Z$ as the partition function. Here to simplify presentation, we set $\hbar = 1$. Further

$$\chi^{(m,s)}_{\alpha\beta\gamma\delta}(\omega) = \frac{\pi}{2b} \sum_{n=1}^{N} \Gamma^{(s)}_{\alpha\beta;mn} \Gamma^{(s)}_{\gamma\delta;nm} \delta(e_n - e_m - \omega)$$  (16)

with $\Gamma^{(s)}_{\alpha\beta;kl}$ as the matrix element of the stress-tensor in the NI basis: $\Gamma^{(s)}_{\alpha\beta;kl} = \langle k_s | \Gamma^{(s)}_{\alpha\beta} | l_s \rangle$. In general $\chi^{(m)}_{\alpha\beta\gamma\delta}$ depends on the energy level $e_m$ and fluctuates over the spectrum. It is then useful to define the spectral averaged susceptibility over the $N$-level spectrum of the basic block

$$\langle \chi^{(s)}_{\alpha\beta\gamma\delta}(\omega) \rangle \omega = \frac{1}{N\omega_c} \sum_{m=1}^{N} \int_{0}^{\omega_c} \chi^{(m,s)}_{\alpha\beta\gamma\delta}(\omega - e_m) \, d\omega$$  (17)

where $\omega_c$ is the bulk spectrum width of the basic block.

Furthermore the fluctuations of $\Gamma^{(s)}_{\alpha\beta;kl}$ as well as those of the energy levels over the ensemble also influence $\chi^{(m,s)}_{\alpha\beta\gamma\delta}(\omega)$ and it is appropriate to consider its ensemble average $\langle \chi^{(m,s)}_{\alpha\beta\gamma\delta}(\omega) \rangle_e$ too. Assuming isotropy, rotationally invariance of the basic block (as its linear size $L \gg a$ with $a$ as the atomic length scale), all $3^8$ components of response function can further be expressed in terms of the transverse and longitudinal response [15]:
\[
\langle \chi^{(s)\gamma\delta}(\omega) \rangle_{e,\omega} = (q_c \delta_{\alpha\beta} \delta_{\gamma\delta} + \delta_{\alpha\gamma} \delta_{\beta\delta} + \delta_{\alpha\delta} \delta_{\beta\gamma}) \langle \chi^{(s)}_t \rangle_{e,\omega}
\]

where \( q_c = \frac{\langle \chi^{(s)}_t \rangle_{e,\omega}}{\langle \chi^{(s)}_t \rangle_{e,\omega}} - 2 \) along with \( \langle \cdot \rangle_e \) implying an ensemble averaging, \( \langle \cdot \rangle_{e,\omega} \) an averaging over both \( \omega \) and ensemble.

The relations given in eq. (14) to eq. (18) are applicable for a basic block. Following similar forms of eq. (6) and eq. (11), these can be generalized for the susceptibility \( \langle \chi^{\text{sup}}_a \rangle_{e,\omega} \) of a super block. This follows by dropping the superscript "s" and with replacements \( \Omega_b \rightarrow \Omega \), \( N \rightarrow N^g \), \( \omega_c \rightarrow W_c \), \( e_n \rightarrow E_n \) in eq. (14) to eq. (18); note here \( E_m \) refers to a many body energy level of \( H \) (defined in eq. (10)).

**B. Relation between \( Q_a^{-1} \) and stress-correlations**

**For Basic Block:** Due to disorder beyond atomic scales, a typical matrix element of the stress tensor of a basic block fluctuates over the ensemble and can be both positive as well as negative. This implies \( \langle \Gamma^{(s)}_{\alpha\beta;kl} \rangle_e = 0 \). Further, at temperature \( T = 0 \), the spectral averaging (defined in eq. (17)) of eq. (16) followed by an ensemble averaging leads to the stress-stress correlation of the basic block

\[
\sum_{m,n=1}^{N} \langle \Gamma^{(s)}_{\alpha\beta;mn} \Gamma^{(s)}_{\gamma\delta;nm} \rangle_e = \frac{N \omega_c \Omega_b}{\pi} (q_c \delta_{\alpha\beta} \delta_{\gamma\delta} + \delta_{\alpha\gamma} \delta_{\beta\delta} + \delta_{\alpha\delta} \delta_{\beta\gamma}) \langle \text{Im} \chi^{(s)}_t \rangle_{e,\omega}
\]

where \( \langle \text{Im} \chi^{(s)}_t \rangle_{e,\omega} \) is defined in eq. (18).

The short-range order of atomic positions in the basic-block along with its small size suggests a homogeneous nature of many body interactions. The ensemble averaged matrix elements of \( \Gamma^{(s)}_{\alpha\beta} \), in the NI basis i.e the eigenfunction basis of \( H^{(s)}_0 \), can then be assumed to be of almost same strength. (This is equivalent to say that, due to small size of block, stress can be assumed to be homogeneous i.e of the same order everywhere in the block. This assumption therefore puts a constraint on our basic-block size). One can then write

\[
\sum_{m,n=1}^{N} \langle \Gamma^{(s)}_{\alpha\beta;mn} \Gamma^{(s)}_{\gamma\delta;nm} \rangle_e = N^2 \langle \Gamma^{(s)}_{\alpha\beta;mn} \Gamma^{(s)}_{\gamma\delta;mn} \rangle_e. \]

This on substitution in eq. (19) leads to

\[
\langle \Gamma^{(s)}_{\alpha\beta;mn} \Gamma^{(s)}_{\gamma\delta;mn} \rangle_e = \frac{\omega_c \Omega_b}{N \pi} \left[ q_c + \delta_{\alpha\gamma} \right] \langle \text{Im} \chi^{(s)}_t (\omega) \rangle_{e,\omega}
\]

\[
\langle \Gamma^{(s)}_{\alpha\beta;mn} \Gamma^{(s)}_{\beta\alpha;mn} \rangle_e = \frac{\omega_c \Omega_b}{N \pi} \langle \text{Im} \chi^{(s)}_t (\omega) \rangle_{e,\omega} \quad \alpha \neq \beta
\]
Further using eq. (13) in eqs. (20, 21), the correlations can be expressed in terms of the average ultrasonic absorption \( \langle Q_{a}^{-1}(\omega) \rangle_{e,\omega} \) of the basic block

\[
\langle \Gamma^{(s)}_{\alpha\beta;mn} \Gamma^{(s)}_{\gamma\delta;mn} \rangle_{e} = N^{-1} \omega_{c} \rho_{m} c_{m}^{2} \Omega_{b} \langle Q_{a}^{-1}(\omega) \rangle_{e,\omega} \delta_{\alpha\gamma} \quad \text{(22)}
\]

\[
\langle \Gamma^{(s)}_{\alpha\beta;mn} \Gamma^{(s)}_{\alpha\beta;mn} \rangle_{e} = N^{-1} \omega_{c} \rho_{m} c_{m}^{2} \Omega_{b} \langle Q_{a}^{-1}(\omega) \rangle_{e,\omega} \quad \text{(23)}
\]

Eq. (23) can be rewritten in terms of the mean-square matrix element \( \nu^{2} \) given by eq. (24) analogous to that of TTLS model:

\[
\langle Q_{a}^{-1} \rangle_{e,\omega} = \frac{N \nu^{2}}{\omega_{c} \rho_{m} c_{m}^{2} \Omega_{b}} = \frac{\gamma^{2}}{\omega_{c} \rho_{m} c_{m}^{2} \Omega_{b}} \quad \text{(24)}
\]

where \( \gamma^{2} \equiv N^{-1} \text{Tr}(\Gamma^{(s)}_{\alpha\beta})^{2} = N \nu^{2} \) is related to the coefficient of the phonon mediated coupling \( V \) between two basic blocks (which is of the form \( \frac{\gamma^{2}}{8\pi \rho_{m} c_{m}^{2} \Omega_{b}} \), see eq. (9)). Note, as discussed in next section, \( \frac{1}{\omega_{c}^{2} h} \) is of the order of the bulk-density per unit volume (see eq. (31)). This in turn renders \( \langle Q_{a}^{-1} \rangle_{e,\omega} \) given by eq. (24) analogous to that of TTLS model:

\[
\langle Q_{a}^{-1} \rangle_{\text{TTLS},e,\omega} = \frac{\pi \gamma^{2} \rho}{2 \rho_{m} c_{m}^{2}} \Omega \quad \text{with } \Omega \text{ as the density of states of TTLS per unit volume.}
\]

**For Super Block:** eq. (24) corresponds to the average coefficient of attenuation in a basic block. Proceeding exactly as above, the average coefficient for a super block, say \( \langle Q_{a}^{-1} \rangle_{\text{sup},e,\omega} \), can also be obtained. The steps are as follows. Eq. (19) is now replaced by the relation

\[
\sum_{m,n=1}^{N_{g}} \langle \Gamma^{(s)}_{\alpha\beta;mn} \Gamma_{\gamma\delta;mn} \rangle_{e} = \frac{N_{g} W_{c} \Omega}{\pi} \left( q_{c} \delta_{\alpha\beta} \delta_{\gamma\delta} + \delta_{\alpha\gamma} \delta_{\beta\delta} + \delta_{\alpha\delta} \delta_{\beta\gamma} \right) \langle \text{Im } \chi_{t} \rangle_{e,\omega}^{\text{sup}} \quad \text{(25)}
\]

where \( \Gamma_{\alpha\beta;mn} \) refers to the matrix element of \( \nabla_{\alpha\beta} \) in the eigenbasis of \( H \) (eq. (10)). But noting that the left side of eq. (25) can be rewritten as \( \langle \text{Tr}(\Gamma^{(s)}_{\alpha\beta;mn})^{2} \rangle \) and is therefore basis-invariant, it can be evaluated in the eigenbasis of \( H_{0} \) i.e. the product basis of single block states referred as \( |E_{n}^{0} \rangle, n = 1 \rightarrow N_{g} \). Using

\[
\Gamma_{\alpha\beta;mn} = \sum_{s=1}^{g} \Gamma^{(s)}_{\alpha\beta;mn} \quad \text{(26)}
\]

along with \( \langle \Gamma^{(s)}_{\alpha\beta;mn} \Gamma^{(s)}_{\alpha\beta;mn} \rangle_{e} = 0 \), it is easy to see that

\[
\sum_{m,n=1}^{N_{g}} \langle \Gamma_{\alpha\beta;mn} \Gamma_{\gamma\delta;mn} \rangle_{e} = g N_{g}^{\nu+1} \nu^{2}. \quad \text{(27)}
\]

The above follows because \( \Gamma^{(s)}_{\alpha\beta;mn} \neq 0 \) only if the product states \( |E_{n}^{0} \rangle \) and \( |E_{n}^{0} \rangle \) differ only by the contribution from the \( s^{th} \) basic block. Further this also implies that the relevant spectral averaging for the super block is same as that of a basic block i.e \( W_{c} = w_{c} \). The above, along with the definition \( \langle Q_{a}^{-1} \rangle_{\text{sup},e,\omega} = (\pi \rho_{m} c_{m}^{2})^{-1} \langle \text{Im } \chi_{a} \rangle_{e,\omega}^{\text{sup}} \) and \( \Omega = g \Omega_{b} \), now leads to
\[ \langle Q_a^{-1} \rangle_{e,\omega}^{\text{sup}} = \frac{N g \nu^2}{\omega_c \rho_m c_a^2 \Omega} = \frac{\gamma^2}{\omega_c \rho_m c_a^2 \Omega_b} \]  

(28)

A comparison of the above result with eq.(24) clearly indicates that

\[ \langle Q_a^{-1} \rangle_{e,\omega}^{\text{sup}} = \langle Q_a^{-1} \rangle_{e,\omega} \]  

(29)

Based on the theory of coupled blocks for a super block and a renormalization approach, the study [10] gives

\[ \langle Q_a^{-1} \rangle_{e,\omega}^{\text{sup}} = \left[ \frac{1}{\langle Q_a^{-1} \rangle_{e,\omega}} + K \log_2(R/L) \right]^{-1/2} \]

with \( R, L \) as the linear sizes of a super block and a basic block, respectively and with \( K \) as a constant: \( K \sim 150 \); for experimental sizes, their result reduces to eq.(29).

To proceed further, it is necessary to determine the bulk spectral width \( \omega_c \) as well as the basic block volume \( \Omega_b \).

### IV. BULK SPECTRAL WIDTH OF THE BASIC BLOCK

Consider a basic block of an arbitrary isotropic amorphous material, composed of \( g_0 \) molecules labeled as "k" with positions \( r_k \) and masses \( m \). The Hamiltonian of the block can be written as

\[ \mathcal{H} = \mathcal{H}_0 + \mathcal{U}(r_1, \ldots, r_{g_0}) \]  

(30)

where \( \mathcal{H}_0 \) is the total Hamiltonian of \( g_0 \) uncoupled (or free) molecules \( \mathcal{H}_0 = \sum_{k=1}^{g_0} h_k(r_k) \) with \( h_k \) as the Hamiltonian of the \( k^{th} \) molecule, consisting of intra-molecular interactions and \( \mathcal{U} \) as inter-molecular (coulombic type) many-body interactions of \( g_0 \) molecules. At very low temperatures, the induced dipole interactions result in excitations among vibrational energy levels of molecules (not strong enough to excite the electronic states and the chemical bonding prevents the rotation of molecules), their contribution dominating the physical properties e.g. ultrasonic attenuation. As discussed in [16], the ensemble averaged density of the states which participate in these excitations, has a universal form in the bulk of the spectrum: \( \langle \rho_{\text{bulk}}(e) \rangle = \frac{N b}{2\pi} \sqrt{2 - (be)^2} \) with \( b \) later referred as the bulk spectral parameter and \( \langle \rangle \) as the ensemble average; (note here \( \langle \rho_e(e) \rangle \) is normalised to \( N: \int \langle \rho_e(e) \rangle \, de = N \)). This gives the bulk spectral width \( \omega_c \) as

\[ \omega_c = \frac{2\sqrt{2}}{b} \frac{2N}{\pi \langle \rho_{\text{bulk}}(0) \rangle} \]  

(31)

As discussed in detail in [16], \( b \) can be expressed as
with $C_6$ as the strength of dispersion interaction between two molecules, $z$ as the average number of nearest neighbors of a given molecule, $g_0$ as the number of molecules in the basic block, $\eta = \mathcal{N} - 1$ with $\mathcal{N}$ as the number of relevant vibrational energy levels in a molecule) and $2R_v$ as the distance between centers of two nearest neighbor molecules. Based on the structural stability analysis of the amorphous systems, $z$ is predicted to be of the order of 4 (for a three dimensional block) [2]. Further, as discussed in [16], $\mathcal{N}$ corresponds to the number of single molecule states participating in dispersion interaction with another molecule. Alternatively, this is the number of dipole transitions among vibrational states of a molecule due to dispersion interaction with another one. Usually the allowed number of such transitions is 3 ($\delta m = 0, \pm 1$ with $m$ as the quantum number of the state); in any case weak nature of the dispersion interaction rules out higher number of such transitions). From [16], $b \sim 10^{18} J^{-1}$ which leads to $\omega_c \sim 10^{-18} J$.

To determine $\langle Q^{-1} \rangle$ from eq. (24), the information about the volume $\Omega_b$ of a basic block is still missing. As discussed in next section, this can be estimated, based on the competition between the inter-block forces with the intra-block ones.

V. SIZE OF A BASIC BLOCK

As the universal properties are expected to originate from the length scales beyond ordered structure, we assume linear size of each basic block large enough compared to the typical inter-atomic distance but is otherwise arbitrary. At this stage, we have two options for the upper size limit: first option is that the size is comparable to the range of unretarded dispersion forces among molecules which dominate within a block. Due to orientation free nature of these forces, the interaction among the molecules within a basic block can be regarded as isotropic. The second option is that we leave the upper limit arbitrary; the molecules within a block in that case interact via phonon mediated coupling (dependent on inverse cube of the distance between molecules) too. One could refer the basic blocks of first and second options as dispersion blocks and stress blocks.

In principle, proceeding with either option should lead to analogous results for the physical properties; (the 2nd option was adopted in [10, 15] for a renormalisation approach to analyze
universality). In practice however it is easier to choose the first one for following reasons (i) the dispersion forces being the short range one, they correspond to more natural choice to define the building block size. (ii) isotropic nature and homogeneity of the interaction within the block is a better approximation in this case which is technically helpful to calculate the level density and other properties. (iii) as discussed later, it gives us the route to calculate the coefficient of ultrasonic attenuation of phonons in terms of molecular properties and confirm its universality at nano-scales. Proceeding with the first option, it is clear that the block size should not be big enough to allow the onset of phonon-mediated $r^{-3}$ interactions. The radius, say $R_0$ of the dispersion forces in glasses beyond which $r^{-3}$ interaction becomes dominant can then be obtained by a comparison of these interactions by two routes, those between two molecules or between two clusters. As discussed below, both these approaches are needed to formulate $R_0$ in terms of known molecular properties.

Interactions between two molecules

Consider two molecules with their centers at a distance $r$ in a glass solid. The existence of long wavelength phonons at low temperatures leads to a phonon-mediated pair-wise interaction, decaying as inverse cube of distance between them. The corresponding interaction energy is

$$V_{stress}(r) \approx \frac{\gamma_m^2}{\rho_m c^2 r^3}$$

with $\rho_m$ as the mass-density of the basic block, $c$ as speed of the sound waves in the block and $\gamma_m$ as the strength of the phonon induced $r^{-3}$ coupling of the two molecules. Note, as discussed in appendix C, $\gamma_m$ can be expressed in terms of the number of molecules and basic block parameters.

But as discussed in detail in [16], two molecules at a distance $r$ are also acted by $r^{-6}$ type interaction due to dispersion forces. Assuming the molecules to be in their ground state (valid at low temperatures), the interaction energy, averaged over all possible orientations, can be given as [24]

$$V_{dispersion}(r) \approx \frac{C_6}{r^6}$$

with $C_6$ as the strength of dispersion interaction between two molecules.

Our interest is now to find the distance $r = 2R_0$ at which the magnitude of the two
interactions is equal. This implies

\[ V_{\text{stress}}(2R_0) = V_{\text{dispersion}}(2R_0) \]  \hspace{1cm} (35)

Substitution of eq.(33) and eq.(34) in eq.(35) gives

\[ R_0^3 = \frac{\rho_m c^2 C_6}{8 \gamma_m^2}. \]  \hspace{1cm} (36)

As the Hamaker constant \( A_H \) is a constant for materials, it is better to express \( C_6 \) in terms of \( A_H \): \( C_6 \approx \frac{A_H}{\pi^2 \rho_m^2} \), with \( \rho_m = \frac{1}{\Omega_{\text{eff}}} \) as the number density of the molecules and \( \Omega_{\text{eff}} \) as the average volume available to a typical molecule. (Note here the number of those molecules should be considered which interact through dispersion interaction). Consider a typical molecule of radius \( R_m \); its molar volume can then be expressed as

\[ \Omega_m = s_m R_m^3 = \frac{M}{\rho_m N_a} \]  \hspace{1cm} (37)

with \( M \) as the molar mass of the particle interacting by VWD, \( N_a \) as the Avogadro number and with \( s_m \) as a structure constant e.g. \( s_m = 4\pi/3 \) assuming a spherical shape for the molecule. The effective volume available to each molecule in the system however also depends on their packing. Assuming \( 2R_v \) as the average distance between two neighboring molecules in the system, the average volume, say \( \Omega_{\text{eff}} \), available to a typical molecule is

\[ \Omega_{\text{eff}} = s_m (R_v + R_m)^3 \approx (1 + y)^3 \Omega_m \]  \hspace{1cm} (38)

where \( \Omega_m \) is the molar volume: \( \Omega_m = \frac{M}{\rho_m N_a} \). The last equality in the above equation follows by taking \( \Omega_m = s_m R_m^3 \). Further writing \( R_v = y R_m \) along with eq.(38) in the definition of \( C_6 \) gives

\[ C_6 \approx \frac{A_H (\Omega_{\text{eff}})^2}{\pi^2} \approx \frac{s_m^2}{\pi^2} (1 + y)^6 R_m^6 A_H \]  \hspace{1cm} (39)

The above information can now be used to rewrite \( R_0 \) in terms of known molecular parameters. Substitution of eqs.(37-39) in eq.(36) leads to

\[ R_0^3 = \frac{(1 + y)^6 c^2 A_H M \Omega_m}{8 \pi^2 \gamma_m^2 N_{av}} = \frac{(1 + y)^6 A_H M^2 c^2}{8 \pi^2 N_{av}^2 \gamma_m^2 \rho_m}. \]  \hspace{1cm} (40)

Clearly \( R_0 \) is an important length scale, related to equivalence of the above two interactions energies, one short and other long range; for \( r < R_0 \), dispersion force dominates
and for \( r > R_0 \), phonon mediated coupling dominates. As discussed below, the competition leads to organization of molecules in structural sub-units i.e basic blocks of linear size \( R_0 \).

**Interactions between two neighboring clusters**

Consider a cluster of molecules within a sphere of radius \( R_0 \), the dominant energy of interaction between any two molecule within the sphere is that of dispersion. But for two surface molecules diagonally opposite (i.e distance \( 2R_0 \) across a diameter), the two interaction energies are equal. For any molecule outside the spherical surface, the phonon mediated coupling dominates over that of dispersion. This clearly indicates existence of structures of linear size \( 2R_0 \). But the question is, if one considers the collective interactions of a cluster of molecules with another neighboring cluster, for what size of the clusters, the two interaction energies still be equal?

The behavior of a macroscopic glass solid can be analyzed in terms of a collection of clusters of arbitrary shape and size. Let us consider the whole glass solid divided into spherical blocks of radius \( t \) with \( 2R_v \) as the closest separation distance between them. Here the shape of the blocks is chosen to be spherical; keeping in view, the medium range order in glasses as well as molecule shape assumed to be spherical, this is a natural choice which also helps in technical simplification without any loss of generality. To choose the size of the blocks, we subject them to following conditions:

(i) The blocks are dispersion dominated i.e within one block, the dominant force between any two molecules is that of dispersion.

(ii) The size of the blocks are so chosen that, for neighboring blocks, dispersion energy of the block-block interaction is equal to that of phonon mediated coupling between them.

With \( \gamma_m \) as the strength of the phonon mediated interaction between two molecules, the phonon mediated coupling between two spherical blocks, each containing \( g_0 \) molecules and with their centers separated by a distance \( 2(t + R_v) \), can be expressed as (for \( t > R_v \))

\[
V_{b,\text{stress}}(t) \approx -\frac{4}{3} \frac{\pi^2 \gamma_m^2 \rho_n^2}{\rho_m c^2} t^3
\]

with \( \rho_n \) as the number density of the interacting particles.

Here subscript \( b \) refers to interaction energy of two neighboring basic blocks. Further the dispersion interaction between two spherical clusters of radius \( t \) separated by a contact
distance $2R_v$ can be given as

$$V_{b,\text{dispersion}}(t) = -\frac{\pi^2 \rho_m^2 C_6}{24} \frac{t}{R_v}$$

Let us now impose the condition (i) that

$$V_{b,\text{dispersion}}(t) = V_{b,\text{stress}}(t)$$

Substitution of eq.(42) and eq.(41) in eq.(43) then leads to

$$\frac{t}{R_v} \approx \frac{32 \gamma_m^2 t^3}{\rho_m c^2 C_6}$$

Now using eq.(36) to replace $C_6$ in the above, the size $t$ of the basic block can be expressed in terms of $R_0$ and $R_v$,

$$t^2 = \frac{R_0^3}{4 R_v}$$

Note as $R_0 \geq 4R_v$, (the total distance between the centers of two neighboring molecules being $2(R_m + R_v)$, each assumed spherical with radius $R_m$, and $R_v = y R_m$ with $y$ a material dependent constant of order 1), this along with the above implies

$$\left( \frac{t}{R_0} \right)^2 = \frac{R_0}{4 R_v} \geq 1$$

It is worth emphasizing here that, for two neighboring blocks each of radius $t > R_0$, the two interaction strengths can not be equal and $V_{b,\text{stress}}$ will always dominate $V_{b,\text{dispersion}}$. This can directly be seen from eq.(42) and eq.(41) which leads to $V_{b,\text{stress}}(t) / V_{b,\text{dispersion}}(t) = 4R_v t^2 / R_0^3$ for arbitrary $t$. Clearly with $R_0 > 4R_v$ and $t > R_0$, $V_{b,\text{stress}}(t) > V_{b,\text{dispersion}}(t)$.

As clear from the above, the size of the basis block depends on the inter-molecular distances for the molecules interacting by dispersion forces (i.e those on secondary structure e.g interaction between two molecules on neighboring rings but not on the same ring). Further, although both $R_v$ and $R_0$ are material properties, the ratio $R_v / R_0$ is a constant independent of the glass material. This in turn gives a constant, system-independent number of the molecules within each block:

$$g_0 = \frac{\Omega_b}{\Omega_{\text{eff}}} \approx \frac{1}{(1 + y)^3} \left( \frac{t}{R_m} \right)^3 = \frac{y^3}{8 (1 + y)^3} \left( \frac{R_0}{R_v} \right)^9$$

As discussed in the next section, this in turn leads to a material independent value of the average ultrasonic attenuation coefficient $\langle Q^{-1} \rangle$.

The quantitative information about $R_v$ available for a wide range of materials suggests $R_v \sim R_m$. In the present work, hereafter we will take $y = \frac{R_v}{R_m} \sim 1$ which leads to,
eq. (47), $g_0 \approx 8$. Assuming uniform mass density, this further implies only three nearest neighbor molecules to a given molecule within a spherical basic block of radius $t = \sqrt{\frac{R_v^3}{3\pi \rho_m}}$.

Taking typical value $R_m \sim 3$ AA gives $R_0 \sim 15$ AA which corresponds to the length scale for medium range topological order (MRO) (10 AA $\rightarrow$ 30 AA). This is as expected because VWD interactions are negligible beyond MRO and other interactions start dominating beyond this length scale.

VI. QUANTITATIVE UNIVERSALITY OF $Q_a^{-1}$

Based on unretarded dispersion interaction between molecules, eq. (24) relates the ultrasonic attenuation coefficient $\langle Q_a^{-1} \rangle$ to the bulk spectrum width $\omega_c$ and thereby bulk spectrum parameter $b$. Eq. (32) expresses $b$ in terms of the molecular properties. A combination of two relations then gives $\langle Q_a^{-1} \rangle$ in terms of the molecular properties. This can be derived as follows. A substitution of eq. (31) and eq. (32) in eq. (24) along with relation $\Omega_b = s t^3$, with $s = 4\pi/3$ leads to

$$\langle Q_a^{-1} \rangle_{e,\omega} \approx \frac{64 \gamma^2}{2 s \eta \sqrt{2zg_0} \rho_m c^2 C_6} \frac{R_v^6}{t_0^3}$$

(48)

where, as shown in appendix C, we have

$$\gamma^2 \approx \frac{4 \pi g_0}{K \sqrt{2}} \gamma_m^2$$

(49)

with $K^2 = 18 \left(5 - \frac{c_d^2}{c_f^2}\right)$. Using eq. (36) to replace $C_6$ in the above equation, we have

$$\langle Q_a^{-1} \rangle_{e,\omega} \approx \frac{8 \pi \sqrt{g_0}}{s \eta \sqrt{2z} K} \frac{R_v^6}{R_0^3 t^3}$$

(50)

$$= \frac{32 \pi}{s \eta \sqrt{2z} K} \left(\frac{y}{(1+y)}\right)^{3/2} \left(\frac{R_v}{R_0}\right)^{21/4}$$

(51)

Here the $2^{nd}$ equality is obtained by substitution of $t$ and $g_0$ from eq. (45) and eq. (47).

As clear from the above, $\langle Q_a^{-1} \rangle_{e,\omega}$ depends only on $\eta = N - 1$, with $N$ as allowed dipole transitions in a molecule, and the number $z$ of nearest neighbors of a molecule (those only interacting by VWD). As mentioned in sec.V (in text below eq. (47)), $\eta = N - 1 = 2$ and $z = 3$. Further, based on the analysis of 18 glasses, we find $R_0 \sim 4 - 5 R_m \sim 4R_v$. 

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It is worth emphasizing here that eq.(51) is based on the balancing of the Vander-Waal forces with phonon induced interaction at the length scales of medium range order. The universal aspects of this competition results in the quantitative as well qualitative universality of $\langle Q^{-1} \rangle_{e,\omega}$ which is consistent with experimental observations [26]. As indicated by previous experiments, the ratio $\frac{c_l}{c_t}$ varies between 1.5 → 2, thus changing $\langle Q^{-1} \rangle_{e,\omega}$ within 10% only.

Further insight in the above result can be gained by rewriting $\langle Q^{-1} \rangle_{e,\omega}$ in terms of the approximate number of molecules, say $g_0$, in a basic block. Substitution of eq.(47) in eq.(51) gives $\langle Q^{-1} \rangle_{e,\omega} \propto g_0^{-7/6}$. The latter along with the definition given in eq.(12) suggests that universality is brought about by the phonons of wavelength $\lambda \sim g_0 l$ with $l$ as their mean free path.

VII. COMPARISON WITH EXPERIMENTAL DATA

To compare the theoretical prediction with experiments, we rewrite eq.(51) as follows. Substitution of the relation $\Omega_m = \frac{4}{3} \pi R_m^3$ in eq.(40) gives

$$\left( \frac{R_0}{R_v} \right)^3 = \frac{1}{y^3} \left( \frac{R_0}{R_m} \right)^3 = \frac{(1 + y)^6}{y^2} \frac{M A_H}{6 \pi N_{av}} \left( \frac{c}{\gamma_m} \right)^2.$$  \hspace{1cm} (52)

Here $c$, as the speed of sound, and $\gamma_m$, as the phonon mediated coupling constant between molecules, have directional dependence: $c = c_l, c_t$ and $\gamma_m = \gamma_l, \gamma_t$ with subscripts $l, t$ referring to longitudinal and transverse direction, respectively. The above along with eq.(51) gives,

$$\langle Q^{-1} \rangle_{e,\omega} = \frac{48 f(y)}{\eta \sqrt{2 z K}} \left( \frac{6 \pi N_{av} \gamma_m^2}{M A_H c_a^2} \right)^{7/4}$$  \hspace{1cm} (53)

where $f(y) = \frac{y^{7/4}}{(1+y)^2}$ with $\eta = 2$, $z = 3$ and the subscript $a = l, t$. For later reference, note $f(y)$ is almost same for $y = 1$ and $y = 1.5$: $f(1) = 2.44 \times 10^{-4}$ and $f(1.5) = 2.59 \times 10^{-4}$.

As standard TTLS model is a special case of our generic block model, the available information for the coupling constants in the former case can be used for the latter. (Note TTLS model is based on the presence of some two level atoms/ molecules (TLS) as defects. The coupling constants of the molecules within a block due to molecule-phonon interaction can then be taken same as those of TLS). The TLS coupling constants are related to tunnelling
strength $C_a$, defined as

\[ C_a = \frac{P}{\rho_m} \left( \frac{\gamma_a}{c_a} \right)^2, \quad (54) \]

with $P$ as the spectral density of tunnelling states. According to tunnelling model,

\[ C_a = \frac{2}{\pi} \langle Q^{-1}\rangle, \quad (55) \]

As the experimental results are usually given in terms of TTLS model parameters, we define the analog of $C_a$ for our case for comparison

\[ B_a = \frac{2}{\pi} \langle Q^{-1}_{\text{a,pohl}} \rangle = 2 \langle Q^{-1}_a \rangle. \quad (56) \]

To quantitatively determine $B_l, B_t$ from the above equation, a prior information about $A_H, M$ and the ratio $\frac{\gamma_l}{c_l}, \frac{\gamma_t}{c_t}$ is required. For the purpose, we proceed as follows.

(i) Determination of $\frac{\gamma_a}{c_a}$: With eq.(53) dependent on the fourth power of the ratio, a correct estimation of $\frac{\gamma_a}{c_a}$ is crucial for determination of $B_a$. While the values for $c_l, c_t$ are easily available for many glasses [26], the same is not the case for $\gamma_l, \gamma_t$. As mentioned above, the standard practice to estimate $\gamma_l, \gamma_t$ as the TLS coupling constants.

Using ultrasonic absorption data, the study [36] first determines $C_l, C_t$ as adjustable parameters; eq.(54) then leads to $\gamma_l, \gamma_t$ for 18 glasses. But $C_l, C_t$-values mentioned in [26] for some of the glasses are different from [36] (although same values for $c_l, c_t$ in both studies) which then lead to, from eq.(54), different values for $\gamma_l, \gamma_t$. The latter on substitution in eq.(53) gives values different from those obtained from data from [36]. The columns 3 and 9 of table II however display $B_{l,t}$ results derived from eq.(53), with $\gamma_l, \gamma_t, c_l, c_t$ values taken from the study [36]; for comparison the corresponding $C_l, C_t$ values from [36] are also displayed in columns 4 and 10. The study [26] however gives only $C_l, C_t$ (from two experimental approaches, namely, acoustic and flexural); for comparison with our theory, therefore, we rewrite eq.(53) as

\[ B_a = \frac{6 f(y)}{\eta \sqrt{z}} K \left( \frac{6 \pi N_{av} \rho_m C_a}{M A_H P} \right)^{7/4} \quad (57) \]

with constant $K$ is same as in eq.(49). The $C_l, C_t$ from [26] is displayed in Table II in columns 6, 8, 12, 14; the latter along with $\rho_m$ and $P$ (both given in table I and taken from [36]) is used to obtain $B_l, B_t$ data given in columns 5, 7, 11, 13.

(ii) Determination of $M$: As eq.(53) depends on $M^{3/2}$, a correct estimation of $M$ is important too. Two options available to determine $M$ are (i) mass of the basic structural unit which dominates the structure of the glass and participates in the dispersion interaction
(later referred as vwd unit), or, (ii) the molecular mass of the glass (later referred as formula unit); (here, for example for SiO\textsubscript{2} glass, SiO\textsubscript{2} is the formula unit but dominant structural unit can be SiO\textsubscript{4} or Si(SiO\textsubscript{4})). Clearly, with dispersion interaction as the basis of our analysis, it is reasonable to use the 1\textsuperscript{st} option. To analyze the influence however we consider both options to calculate $B_l, B_t$. The details of dominant structural unit for each glass and its mass, referred as $M_1$, is discussed in appendix A. The formula mass, labelled here as $M_2$, corresponds to weighted summation of the molar masses of each constituent of the glass: for the latter consisting of $n$ components $X_k, k = 1 \rightarrow n$, with their molar mass as $m_k$ and weight percentage as $p_k$, $M_2 = \sum_{k=1}^{n} p_k m_k$. The glass composition for the 18 glasses is given in appendix A and their $M_1, M_2$ values are displayed in table I.

(iii) Determination of $A_H$: for materials in which spectral optical properties are not available, two refractive-index based approximation for $A_H$ namely, standard Tabor-Winterton approximation (TWA) (appropriate for low refractive index materials, $n < 1.8$) and single oscillator approximation (SOA) (for higher indexes $n > 1.8$), provide useful estimates \cite{34}. The $A_H$ for 18 glasses listed in Table 1 are based on these approximations (with details given in \cite{16}).

VIII. DISCUSSION

As mentioned above, the standard practice is to determine the ratio $\frac{\gamma_a}{c_a}$, with $a = l, t$, is from the definition \cite{54} of the tunnelling strength $C_a$. The reported experimental data for the latter however varies significantly from one experiment to another (as indicated by the data in even numbered columns of tables II, III). This in turn leads to different values of $\frac{\gamma_a}{c_a}$ and thereby $B_a$ (from eq.\cite{53}); the latter are displayed in even-numbered columns of table II, with corresponding experimental data from \cite{36} and \cite{26} given in adjacent columns. (Note the data in odd numbered columns of tables II, III can also be obtained directly from eq.\cite{57}, along with $\overline{P}$ given in table I and $C_l, C_t$ data in odd numbered columns of table II). The even numbered columns of table II are based on $M = M_1$ (the VWD unit mass); the corresponding results for $M_2$ are given in table III (these can also be obtained from the relation $B_{a2} = B_{a1} \left(\frac{M_1}{M_2}\right)^{7/4}$. As displayed in table I, $M_1$ and $M_2$ do not differ significantly for the glass-ceramics. However, for single component glasses e.g. SiO\textsubscript{2} or where one component dominates e.g. in BK7, $B_l, B_t$ prediction based on $M_1$ are closer to experimental data (see
This in turn provides further credence to the relevance of VanderWaal forces in present context.

The values of $B_a$ from eq.(53) along with corresponding experimental $C_a$ data for each glass is also illustrated in figure 1 for $M = M_1$ and figure 3 for $M = M_2$. A direct comparison of theoretical and experimental data is also displayed in an alternative way in figure 2 for $M_1$ and in figure 4 for $M_2$. As mentioned above, the results for a glass vary from one experiment to other often within a factor of 2 but sometimes more e.g. for polymers (see even numbered columns of Tables II, III and [20]). The deviation of our theoretical prediction from experiments is usually less than a factor of 2.

Further, a comparison of figures 3, 4 indicates that the results for $M = M_1$ are closer to experimental data, thus indicating the molecules interacting by VWD interaction as an appropriate choice for the present analysis. This is also consistent with our theoretical approach assuming VWD interactions as the relevant interaction for length scales less than MRO.

An important point to note here is that the dependence of $B_a$ in eq.(53) on glass-properties is based only on the product $M.A_H$ (as the latter also appears in the ratio $\gamma_m c$). The constancy of $\langle Q^{-1} \rangle$ therefore seems to be a reconfirmation of already known relation between $A_H$ and molar volume [24].

At this stage, it is worth reviewing the main assumptions made to arrive at our theoretical predictions:

(i) The interactions within the block are assumed to be homogeneous. The assumption was used in section III for the random matrix modelling of the Hamiltonian as well as in linear response theory for $Q^{-1}$. This puts an upper limit on the allowed block size. Any block of bigger size would include both dispersion as well as phonon-coupling among molecules and thereby lead to inhomogeneity of the interactions. The theory in principle can still be adapted to analyze a super block consisting of bigger basic block sizes but it would need many modifications including the use of sparse random matrices.

To get an idea of as to which interactions are really influencing the dynamics at different length scales, it is instructive to analyze the order of magnitude of various interactions: typically $C_6 \approx 434 \ A_H R_m^6 \sim 10^{-18} \ R_m^6$ (from eq.(39) and $A_H \sim 10^{-20} \ J$ from Table 1) and $\frac{\gamma_m^2}{\rho_m c} \approx 10^{-49} \ J.m^3$ (from Table 1). For two molecules as a distance $2R_0 = 8 \ R_m$ with $R_m \sim 3$. 

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AA, eq.(33) and eq.(34) then give $V_{\text{stress}}(2R_0) = V_{\text{dispersion}}(2R_0) \approx 10^{-23} \text{ J}$ which is of the order of $k_B T$, the thermal energy needed at temperature $T \sim 1^\circ\text{K}$ (with $k_B$ as the Boltzmann constant). The interaction energy for two neighboring blocks with their centers separated by a distance $2(R_0 + R_v)$ is also of the same order: eq.(42) and eq.(41) give $V_{b,\text{stress}} = V_{b,\text{dispersion}} \approx 10^{-23} \text{ J}$. This is however not the case for two molecules at minimum possible distance $2(R_m + R_v) \approx 4R_m$, then $V_{\text{dispersion}} \sim 10^{-17} \text{ J}$ and $V_{\text{stress}} \sim 10^{-22} \text{ J}$.

(ii) The blocks are assumed to be of spherical shapes. This is a natural choice, keeping in view especially of the spherical shape of molecules (although the latter is also an assumption but a standard one). It also helps a simpler technical formulation of the derivations. Alternatively, arbitrary shape of basic blocks can also be chosen but that is at the cost of technical complexity of intermediate steps of the derivation. We believe that although the ratio $\frac{R_0}{R_m}$ may vary slightly with shape but it will be compensated by the structure parameter $s$, thus leaving theoretical prediction in eqs.(53,57) almost unaffected.

(iii) The phonon mediated perturbation is assumed to access all $N$ levels of the basic block Hamiltonian ($N = N^g = 3^g$) within spectral range $\omega_c \sim 10^{-18} \text{ J}$ (from eq.(31)). Although this gives the mean energy level spacing in the spectral bulk as $\Delta_b \approx \frac{\omega_c}{N}$ for a basic block is $\sim 10^{-22} \text{ J}$, the mean level spacing in the lower edge of the spectrum however is much smaller and levels can be accessed by thermal perturbation at low temperatures $T \sim 1^\circ\text{K}$.

(iv) The dominant interactions at at MRO length scales of the glasses are non-retarded dispersion forces among molecules. This is applicable only to insulator glasses and needs to be replaced for other cases.

(v) The theoretical results presented here (figures 1-4 and table 1-3) are obtained from eq.(53) with $y = R_v/R_m \sim 1$ for the molecules interacting by VWD. In general $y$ fluctuates from one glass to another with 1 as its average value; the glass-specific values for $y$ should be taken, in principle, for better accuracy. However as noted below eq.(53), $f(y)$ remains almost same for $y = 1$ and $y = 1.5$: $f(1) = 2.44 \times 10^{-4}$ and $f(1.5) = 2.59 \times 10^{-4}$. The fluctuation of $y$ therefore does not seem to have significant effect of our results.

(vi) The $B_{11}, B_{11}$ values given in table II are obtained by approximate $A_H$ values used in eq.(53); we believe the results could be improved if exact values of $A_H$ are used (see 33, 34). Further our results given in table II are based on the Hamaker constant of the molecules interacting in vacum. The vwd unit is however the dominant cation surrounded by other molecules; the interaction between two cations is therefore mediated by other molecules. It
is natural to query, therefore, how the $B_a$ results will be affected if $A_H$ values in the relevant medium are considered.

IX. CONCLUSION

In the end, we summarize with our main ideas and results.

Based on experimental evidence of ordered structure in glasses below MRO ($10 \rightarrow 30 \text{ } AA$) and its lack above, we describe a macroscopic size glass material as elastically coupled, spherical shape, generic blocks, with homogeneous dispersion interaction within each such block. A random matrix modelling of their hamiltonian and linear response to an external strain field, then relates the low temperature averaged ultrasonic attenuation coefficient for the glass to the molecular properties. This in turn reveals the quantitative universality of the coefficient which is consistent with experimental observations in the temperature regime $1^\circ K \rightarrow 10K$ [26].

The central result of our work is given by eq. (53) with main assumptions summarised in section IX. An important insight revealed by our formulation is the physical significance of the basic block size: it defines the length scale $R_0$ beyond which $\langle Q^{-1} \rangle$ attains universal value; $R_0$ turns out to be of the order of typical MRO length scales in glasses. Although this result is derived by assuming the strength of dispersion forces between two neighboring basic blocks equal to their phonon mediated coupling, its consistency with a previous study [19] indicating change in phonon dynamics at MRO length scales, supports the assumption. The length-scale $R_0$ is also consistent with another assumption made in our study i.e regarding the isotropy and homogeneity of the stress filed of the basic block; this follows because almost all molecules within a spherical block of radius $R_0$ are subjected to same interaction strength (with number of molecules within a basic block being 8). The omnipresence of dispersion forces indicates the application of our results to other disordered materials too.

The analysis presented here takes only dispersion type inter-molecular forces into account and neglects the induction forces which restricts, in principle, the application of our results to non-polar molecules. We believe however that inclusion of induction forces would only change numerical value of $b$ (given by eq. (32)) and would not affect the derivations given in section II-VI. Similarly a generalization of the present theory by including electronic
interactions may explain the universality in context of metallic glasses.

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Appendix A: Structural units of the glasses used for $M$ in Table 2

The basic structural unit in a glass depends on the presence of various cations some of which act as network formers and others act as network modifiers (e.g. see pages 9-11 of [37]). Here we give the glass composition and the dominant structural units for 18 glasses used in tables I. The molar mass $M_1$ in Table I refers to the masses of these units.

(1) $a-SiO_2$: The 3-d network in this case has the basic structural unit is $Si[SiO_4]$ with mass $M_1 = 120.09$ (see page 37 of [38], section 11.4.1 of [44]), also see section 2.2 and fig2.7(a) of [39]).

(2) BK7: (wt %): The constituents in this case are 69.9 $SiO_2$, 9.9$B_2O_3$, 8.4$NaO_2$, 8.4$K_2O$, 2.5$BaO$. with 70% $SiO_2$, the basic structural unit in this case is $[SiO_4]$ [40] (also see section 11.4.1, 12.1 of [44]) with mass $M_1 = $.

(3) As$_2$S$_3$: The glass in this case forms chain like structure e.g. $S-S$, As−As or As−S (i.e $S$ or As of one chain interacting with neighboring one). As $S$ is dielectric, we use it as the basic unit participating in VWD interaction (see page 56 of [47], page 125 of [42], also see section 2.2 and fig 2.7(b) of [39]) and therefore choose $M_1 = 32$. Using molar weight of As for the purpose, gives $B_l = 0.29, B_t = 0.62$ and $\frac{a}{c_l} = \frac{a}{c_t} = 0.17$.

(4) LaSF7: Also known as dense lanthanum flint glass, it contains mostly $B_2O_3$, $La_2O_3$ and $ThO_2$ with a few % of $Ta_2O_3$ and $Nb_2O_3$: Here the first three are main net-forming components and last two are net modifiers As net formers are in equal proportion (with each of 30% weight-fraction), each one can play the role of structural unit. In this case, the structural units of each component is triangular i.e $BO_3, LaO_3, ThO_2$. The mass $M_1$ in this case is then obtained as follows: $M = \frac{30}{100}(BO_3 + LaO_3 + ThO_2)$.

(5) SF4: The glass composition in this case is 60.8$SiO_2$, 34.9$PbO$, 2.5$K_2O$ and 1.8 other. with 61% $SiO_2$ but with 35% $PbO$, this has a basic structural unit is $Si_2O_5$ with its mass $M_1 = $ (due to compound of type $2SiO_2.PbO$, 2 Si atoms get coordinated with 5O, (see page 17 of [45], page 14-15 of [41]).

(6) SF59: Here the constituents are 35.3$SiO_2$, 55.6$PbO$, 0.8$K_2O$, 7.9$B_2O_3$ and 0.4 other.
material. with reduced fraction of SiO$_2$, the compound is of form $2PbO.SiO_2$ leading to a basic structural unit of type $SiO_4$ tetrahedral with $M_1$ = (see page 17 of [43], page 14-15 of [41]).

(7) V52: The constituents are 57.8ZrF$_4$, 33.8BaF$_2$ and 8.5ThF$_4$. Due to higher content of ZrF$_4$, the main structural unit in this case is ZrF$_4$ tetrahedral with main role of cations Ba and Th is to cause 2-d structure (network modifiers) (page 35 of [43], page 150 of [], page 157 of [41]). Thus $M_1$ in this case is used as the molar mass of ZrF$_4$.

(8) BALNA: 52 ZrF$_4$, 24 BaF$_2$, 4 AlF$_3$ and 20 NaF. Due to higher content of ZrF$_4$, the main structural unit in this case is ZrF$_4$ tetrahedral with main role of the cations Ba, Al and Na is that of network modifier (i.e to cause 2-d structure) (page 35 of [40], page 150 of [], page 157 of [43]). Thus $M_1$ in this case is used as ZrF$_4$.

(9) LAT: 60 ZrF$_4$, 33 ThF$_4$, 7 LaF$_3$. Due to higher content of ZrF$_4$, the main structural unit in this case is ZrF$_4$ tetrahedral with main role of the cations Ba, Al and Na is that of network modifier (i.e to cause 2-d structure) (page 35 of [43], page 150 of [], page 157 of [41]). Thus $M_1$ in this case is used as ZrF$_4$.

(10) a–Se: Se atoms form chains or 8 atom rings through covalent/ionic bonding. The atoms on neighboring chains or rings interact by lone-pair electrons (VWD). So $M$ is that of Se atom (page 43 of [47], page 115 of [42]).

(11) Se$_{75}$Ge$_{25}$: As both Ge and Se are network-formers, we use both atoms to calculate $M_1$ (page 115 of [42]). Thus $M_1 = M_2$

(12) Se$_{60}$Ge$_{40}$: Here again both Ge and Se act as network-formers (page 115 of [42]), $M_1$ is therefore obtained form both of them and therefore $M_1 = M_2$.

(13) LiCl : 7H$_2$O: The LiCl molecule in presence of H$_2$O forms Li(H$_2$O)Cl$_3$ tetrahedral which seems to act as a basic structural unit. Thus $M_1$ used in our analysis corresponds to this unit.

(14) Zn-Glass: This glass consists of 60ZnF$_2$, 20BaF$_2$, 20NaPo$_3$. Due to higher content of ZnF$_2$, the main structural unit in this case is ZnF$_2$ with main role of the cations Ba and Na is that of network modifier (i.e to cause 2-d structure) [46]. Thus $M_1$ for this case is used as ZnF$_2$.

(15) PMMA: The monomer here has a phenyl group C$_6$H$_5$ which appears as a side unit while the units along the main chain strongly connected by covalent bonds. As VWD interaction occurs between molecules on different chains, the main unit playing role here is
$C_6H_5$. So $M_1$ taken is that of $C_6H_5$.

(16) PS: The monomer here has a phenyl group $C_6H_5$ as a side unit as well as $CH = CH_2$ unit while the units along the main chain strongly connected by covalent bonds. As VWD interaction occurs between molecules on different chains, the main unit playing role here seems to be $CH – CH$ or $CH = CH_2$. The former could be part of Phenyl group. Note unlike other polymers, the monomer of $PS$ is small and therefore only part of Phenyl group may be loosely held and participate in VWD.

(17) PC: as the monomer here is a big molecule, the Phenyl group may be loosely held and participate in VWD. So $M_1$ taken is that of $C_6H_5$

(18) ET1000: here again the monomer is a big molecule, the Phenyl group may be loosely held and participate in VWD. So here again $M_1$ taken is that of $C_6H_5$.

Appendix B: Relation between $\gamma$ and $\gamma_m$

Consider the linear response of a basic block, labeled as "s" containing $g_0$ molecules, to an external strain field. The existence of long wavelength phonons at low temperatures leads to a phonon-mediated pair-wise interaction among molecules, decaying as inverse cube of distance between them. Consider two molecules, labeled as "1” and ”2” with their centers at a distance $r$ within the block. Following the same formulation as in case of blocks, and with $T_{\alpha\beta}$ as the stress tensor component for the molecule, the corresponding interaction energy can be written as

$$V_{\text{stress}}(r) = \frac{1}{\alpha_0 \pi \rho_m c^2} \sum_{t_e} \frac{\kappa^{(12)}_{\alpha\beta\gamma\delta}}{|r_1 - r_2|^3} T^{(1)}_{\alpha\beta} \otimes T^{(2)}_{\gamma\delta}$$  \hspace{1cm} (B1)

with $\alpha_0 = 4$, $r = |r_1 - r_2|$, $\kappa^{(12)}_{\alpha\beta\gamma\delta}$ as in the case of block-block interaction (given by eq.(8)), $\rho_m$ as the mass-density of the molecule, $c$ as speed of the sound waves. The ensemble averaged interaction energy can then be approximated as

$$V_{\text{stress}}(r) \approx \frac{\gamma_m^2}{\rho_m c^2 r^3}$$  \hspace{1cm} (B2)

with $\gamma_m$ as the average strength of the phonon induced $r^{-3}$ coupling of the two molecules.

The interaction parameter $\gamma_m$ can be determined as follows. As $\text{Tr}(V_{\text{stress}}) = 0$, one can write, with $\langle \cdot \rangle_e$ as the ensemble average

$$\langle \text{Tr}(V_{\text{stress}}^2) \rangle_e \approx \left( \frac{\gamma_m^2}{\rho_m c^2 r^3} \right)^2$$  \hspace{1cm} (B3)
where

\[ \langle \text{Tr}(V_{\text{stress}}^2) \rangle_e \approx \left( \frac{1}{\alpha_0 \pi \rho_m c^2 r^3} \right)^2 \sum_{te,te'} K_{\alpha'\beta'\gamma'\delta}^{(12)} K_{\alpha\beta'\gamma'\delta'}^{(12)} \langle \text{Tr}(T_{\alpha\beta}^{(1)} T_{\gamma\delta}^{(2)} T_{\alpha'\beta'}^{(1)} T_{\gamma'\delta'}^{(2)}) \rangle_e \]  

(B4)

Further as

\[ \text{Tr} \left( T_{\alpha\beta}^{(1)} T_{\gamma\delta}^{(2)} T_{\alpha'\beta'}^{(1)} T_{\gamma'\delta'}^{(2)} \right) = \sum_{n,m,k,l} T_{\alpha\beta;nm}^{(1)} T_{\gamma\delta;mk}^{(2)} T_{\alpha'\beta';kl}^{(1)} T_{\gamma'\delta';ln}^{(2)} \]  

(B5)

with \( T_{\alpha\beta;nm}^{(1)} \equiv \langle n| T_{\alpha\beta}^{(1)} |m \rangle \), state \( |n \rangle \) referring to one of the \( N \) single molecule states (unperturbed). Following similar ideas as in the case of a block, we have \( \langle T_{\alpha\beta;mn}^{(1)} T_{\alpha'\beta';kl}^{(1)} \rangle = 0 \) \( \forall m, n, k, l \) and \( \langle T_{\alpha\beta;mn}^{(1)} T_{\alpha'\beta';kl}^{(1)} \rangle = \tau^2 \delta_{\alpha\alpha'} \delta_{\beta\beta'} (\delta_{nk} \delta_{ml} + \delta_{nl} \delta_{mk}) \). On ensemble average, the above leads to

\[ \langle \text{Tr} \left( T_{\alpha\beta}^{(1)} T_{\gamma\delta}^{(2)} T_{\alpha'\beta'}^{(1)} T_{\gamma'\delta'}^{(2)} \right) \rangle \approx 2 \sum_{m,n} \left( \langle T_{\alpha\beta;mn}^{(1)} \rangle^2 \right) \langle \langle T_{\gamma\delta;mn}^{(2)} \rangle^2 \rangle \delta_{\alpha\alpha'} \delta_{\beta\beta'} \delta_{\gamma\gamma'} \delta_{\delta\delta'} \]  

(B6)

The above on substitution in eq. (B3) leads to

\[ \text{Tr}(V_{\text{stress}}^2) \approx 2 \left( \frac{N \tau^2}{\alpha_0 \pi \rho_m c^2 r^3} \right)^2 \sum_{te} (K_{\alpha\beta\gamma\delta}^{(12)})^2 \]  

(B8)

Comparison of eq. (B8) with eq. (B3) gives

\[ \gamma_m^2 \approx \frac{N \tau^2}{\alpha_0 \pi} \frac{K \sqrt{2}}{3} \]  

(B9)

where \( K^2 = \sum_{te} (K_{\alpha\beta\gamma\delta}^{(12)})^2 \).

To relate the above to basic block property \( \gamma^2 \), we proceed as follows. The stress-operator for a basic block can be written in terms of those of molecules:

\[ \Gamma_{\alpha\beta;mn}^{(s)} = \sum_{a=1}^{g_0} T_{\alpha\beta;mn}^{(a)} \]  

The subscripts \( m, n \) now refer to an arbitrary pair chosen from \( N = N^{g_0} \) many body states of the basic block (e.g. the product states \( |e_m^0 \rangle \) and \( |e_n^0 \rangle \) of single molecule states). Further \( T_{\alpha\beta;mn}^{(x)} \neq 0 \) only if \( |e_m^0 \rangle \) and \( |e_n^0 \rangle \) differ only by the contribution from the \( x^{th} \) molecule; this leaves only \( N^{g_0+1} \) non-zero matrix elements for each \( T^{(x)} \). Noting that stress marix elements of different molecules are uncorrelated, it can now be shown that

\[ \sum_{m,n=1}^{N} \langle \Gamma_{\alpha\beta;mn}^{(s)} \Gamma_{\alpha\beta;mn}^{(s)} \rangle_e = \sum_{m,n=1}^{N} \langle \langle T_{\alpha\beta;mn}^{(a)} \rangle^2 \rangle_e = g_0 N^{g_0+1} \tau^2 = g_0 N N \tau^2. \]  

(B10)

Further assuming homogeneous interaction within a basic block, the variances of all matrix elements of the basic block can be approximated as almost equal. The left side of eq. (B10) is then equal to \( N^2 \nu^2 \) (with \( \langle (\Gamma_{\alpha\beta;mn}^{(s)})^2 \rangle_e = \nu^2 \)) which leads to

\[ \gamma^2 = g_0 N \tau^2 = \frac{g_0 \alpha_0 \pi}{K \sqrt{2}} \gamma_m^2 \]  

(B11)
Taking $\kappa^{(12)}_{\alpha\beta\gamma\delta}$ from eq.(8), we have $K^2 = 18 \left[ 1 + 4 \left( 1 - \frac{c^2}{c^2} \right) \right]$

**Appendix C: Abbreviations**

- $a_0$: Bohr’s radius
- $c_a$: speed of sound in longitudinal or transverse direction
- $g_0$: number of molecules in a block
- $r_k$: position of the molecule labeled "$k"$
- $m$: mass of the molecule
- $H$: Hamiltonian of the block with interaction among molecules included.
- $H_0$: Hamiltonian of the non-interacting molecules within a block
- $H_0^{(n)}$: Hamiltonian of a single molecule labeled "$n"
- $U$: molecular interactions among molecules
- $N$: number of roto-vibrational states in the electronic ground state of each molecule playing role in the analysis: $N = 3$
- $N = N^{g_0}$: size of the eigen-basis space of the basic block
- $\eta$: number of allowed dipole transitions among roto-vib states in the electronic ground state of a single molecule
- $\mathcal{K}$: an eigenstate of the basic block which is a product state of single molecule eigenstates
- $|\mathcal{K}_n\rangle$: eigenstate of a single molecule labeled "$n" which contributes to the basic block eigen-state $\mathcal{K}$
- $\nu_0 = \langle E_{K_n}^2 \rangle$: the variance (the square of the line-width) of the vibrational energy-levels of a single isolated molecule Hamiltonian $H_0^{(n)}$.
- $b$: bulk spectral parameter: $\frac{1}{b} = 2 \sum_{L} v_{x,L}$
- $R_m$: distance between two nearest neighbor molecules
- $2R_v$: distance of closest separation between two neighbor molecules (so total distance between their centers is $2(R_m + R_v) \approx 4R_m$
- $2R_0$: distance between centers of two nearest neighbor basic blocks
- $\Omega_m$: volume of a molecule, assumed spherical
- $\Omega_{eff}$: effective volume occupied by a molecule i.e including inter-molecule separation
  $\Omega_{eff} = 8\Omega_m$
Ω_b: volume of a basic block
ρ_m: mass density of a basic block
ρ_n: particle density in a basic block
c: speed of sound waves in the glass block
γ_{im}: strength of the phonon-mediated $r^{-3}$ interaction between two molecules ($γ_{im} ≡ γ_l$ or $γ_t$ based on longitudinal or transverse direction

$γ^2 = Nν^2$ where $ν^2$ is the variance of the stress matrix element of a basic block.

$ρ(ε)$: spectral density of the basic block interaction Hamiltonian $H$

$A_H$: Hamaker constant in Vacuum

$C_6$: strength of dispersion interaction

$V_{dispersion}$: dispersion interaction between two molecules at a distance $r: = C_6/r^6$

$V_{stress}$: phonon mediated interaction between two molecules

$V_{b,dispersion}$: dispersion interaction between two neighbor basic blocks

$V_{b,stress}$: phonon mediated interaction between two neighbor basic blocks

$⟨Q_{a,Pohl}⟩$: internal friction defined in [26], $a = l, t$

$⟨Q_{a,leggett}⟩$: internal friction defined in [10]

$C_a$: tunneling strength defined as $C_a = \frac{γ_a^2 τ}{c_a ρ_m}$

$C_{a,m}$: tunneling strength data from [36]

$C_{a,p1}$: tunneling strength data from [26] (acoustic experiment)

$C_{a,p2}$: tunneling strength data from [26] (flexural experiment)

$M_1$: molar mass of the dominant unit in the glass structure

$M_2$: molar mass of the basic formula of the glass

$B_a: = \frac{2}{π} ⟨Q_{a,Pohl}⟩$

$B_{a1}$: reference to value obtained from eq. [53] by taking $M = M_1$

$B_{a2}$: reference to value obtained from eq. [53] by taking $M = M_2$
FIG. 1. $B_a$-values for 18 glasses: The figure depicts the theoretically predicted $B_a$ and corresponding experimentally known tunneling strengths $C_a$ with respect to glass-index (all listed in Table II). Here $B_{a,xx}$ to eq.\{53\} using tunneling parameters from different experiments (with $xx = bm$ referring to experimental data from \[36\], $xx = p1$ to acoustic and $xx = p2$ to flexural data from \[26\]). The symbols $C_{a,xx}$ refer to experimental data from \[36\] and \[26\] accordingly.
TABLE I. **Physical parameters for 18 glasses**: The table lists the available data for the physical parameters appearing in eq. (51-53). The $\rho, c_t, c_l, \overline{\gamma}$ data from [36] (or [26] if not available in [36]) is displayed in columns 3rd, 4th, 5th and 8th, respectively. The columns 6th and 7th give the $\gamma_l$ and the $\gamma_t$ values, taken from [36] except for few cases; (for those marked by a star (*), the values are obtained either from [26] or from $C_l, C_t$ values given in [36] along with eq.(55). The $A_H$ values given in columns 9th are taken from [16]. The molar mass values, referred as $M_1$ for the vwd unit along with its composition is given in columns 10th and 11th and the mass $M_2$ for formula unit (same as glass molecular weight) in column 12th respectively.

| Index | Glass        | $\rho_m \times 10^3 Kg/m^3$ | $c_t \ km/sec$ | $c_l \ km/sec$ | $\gamma_t \ ev$ | $\gamma_l \ ev$ | $\overline{\gamma} \ 10^{15}/J.m^3$ | $A_H \times 10^{-20} J$ | $M_1 \ gm/mole$ | Vwd unit          | $M_2 \ gm/mole$ |
|-------|--------------|-------------------------------|----------------|----------------|----------------|----------------|--------------------------------------|--------------------------|----------------|-------------------|----------------|
| 1     | a-SiO2       | 2.20                          | 5.80           | 3.80           | 1.04           | 0.65           | 0.8                                  | 6.31                     | 120.09         | $[Si(SiO_4)]$     | 60.08          |
| 2     | BK7          | 2.51                          | 6.20           | 3.80           | 0.9            | 0.65           | 1.1                                  | 7.40                     | 92.81          | $[SiO_4]$         | 65.84          |
| 3     | As2S3        | 3.20                          | 2.70           | 1.46           | 0.26           | 0.17           | 2.0                                  | 19.07                    | 32.10          | [S]               | 246.03         |
| 4     | LASF         | 5.79                          | 5.64           | 3.60           | 1.46           | 0.92           | 0.4                                  | 12.65                    | 167.95         | [LASF]            | 221.30         |
| 5     | SF4          | 4.78                          | 3.78           | 2.24           | 0.72           | 0.48           | 1.1                                  | 8.40                     | 136.17         | $[Si_2O_5]$       | 116.78         |
| 6     | SF59         | 6.26                          | 3.32           | 1.92           | 0.77           | 0.49           | 1.0                                  | 14.05                    | 92.81          | $[SiO_4]$         | 158.34         |
| 7     | V52          | 4.80                          | 4.15           | 2.25           | 0.87           | 0.52           | 1.7                                  | 8.37                     | 167.21         | [ZrF_4]           | 182.28         |
| 8     | BALNA        | 4.28                          | 4.30           | 2.30           | 0.75           | 0.45           | 2.1                                  | 6.87                     | 167.21         | [ZrF_4]           | 140.79         |
| 9     | LAT          | 5.25                          | 4.78           | 2.80           | 1.13           | 0.65           | 1.4                                  | 9.16                     | 205.21         | [ZrF_6]           | 215.69         |
| 10    | a-Se         | 4.30                          | 2.00           | 1.05           | 0.25           | 0.14           | 2.0                                  | 18.23                    | 78.96          | [Se]              | 78.96          |
| 11    | Se75Ge25     | 4.35                          | 0.00           | 1.24           | 0.15           | 1.0            | 1.0                                  | 22.19                    | 77.38          | $[Se_3Ge_1]$      | 77.38          |
| 12    | Se60Ge40     | 4.25                          | 2.40*          | 1.44*          | 0.16           | 0.4            | 23.56                                | 76.43                    | [Se_3Ge_1]     | 76.43             |
| 13    | LiCl:7H2O   | 1.20                          | 4.00           | 2.00*          | 0.62           | 0.39           | 1.4                                  | 4.75                     | 131.32         | $[Li(H_2O)Cl_3]$ | 168.50         |
| 14    | Zn-Glass     | 4.24                          | 4.60           | 2.30           | 0.70           | 0.38           | 2.2                                  | 7.71                     | 103.41         | [ZnF_2]           | 103.41         |
| 15    | PMMA         | 1.18                          | 3.15           | 1.57           | 0.39           | 0.27           | 0.6                                  | 6.10                     | 102.78         | [PMMA]            | 102.78         |
| 16    | PS           | 1.05                          | 2.80           | 1.50           | 0.20           | 0.13           | 2.8                                  | 6.03                     | 27.00          | $[CH – CH_2]$     | 105.15         |
| 17    | PC           | 1.20                          | 2.97           | 1.37*          | 0.28           | 0.18           | 0.9                                  | 6.00                     | 77.10          | $[C_6H_5]$        | 252.24         |
| 18    | ET1000       | 1.20                          | 3.25           | 0.35           | 0.22           | 1.1            | 4.91                                 | 77.10                    | 77.10          | $[C_6H_5]$        | 77.10          |
TABLE II. Comparison of theoretical and experimental values of internal friction for 18 glasses with \( M = M_1 \): Here the theoretical result from eq.(53) labelled as \( B_a \), with \( a \equiv l , t \), from eq.(53) are displayed in odd numbered columns for \( M = M_1 \); the values used for \( M_1 , c_t , \gamma_t , \gamma_t \) are given in Table I. For comparison, the corresponding experimental data for \( C_a \) is given in adjacent even-numbered columns (labelled as \( C_{a,xx} \) with \( xx \equiv bm \) for data from [36], \( xx \equiv p1 \) for acoustic data from [26], \( xx \equiv p2 \) for flexural data from [26]). Note the study [26] does not give \( \gamma_l , \gamma_t \)-values but only \( C_l , C_t \)-values; the latter along with eq.(57) are used to obtain \( B_{a1,xx} \).

| Index | Glass         | \( B_{l1,bm} \) | \( C_{l,bm} \) | \( B_{l1,p1} \) | \( C_{l,p1} \) | \( B_{l1,p2} \) | \( C_{l,p2} \) | \( B_{t1,bm} \) | \( C_{t,bm} \) | \( B_{t1,p1} \) | \( C_{t,p1} \) | \( B_{t1,p2} \) | \( C_{t,p2} \) |
|-------|---------------|-----------------|----------------|-----------------|----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Units | \( \times 10^4 \) | \( \times 10^4 \) | \( \times 10^4 \) | \( \times 10^4 \) | \( \times 10^4 \) | \( \times 10^4 \) | \( \times 10^4 \) | \( \times 10^4 \) | \( \times 10^4 \) | \( \times 10^4 \) | \( \times 10^4 \) | \( \times 10^4 \) | \( \times 10^4 \) |
| 1     | a-SiO2        | 4.50            | 3.10           | 4.51            | 3.00           | 4.00            | 2.80            | 3.81            | 2.90            | 4.51            | 3.00            | 4.78            | 3.10            |
| 2     | BK7           | 3.09            | 2.70           | 4.38            | 3.30           |
| 3     | As2S3         | 0.76            | 1.60           | 1.64            | 2.30           | 0.69            | 1.40            | 1.48            | 2.00            | 0.96            | 1.70            |
| 4     | LASF7         | 1.92            | 1.20           | 4.81            | 2.00           | 1.84            | 1.16            |
| 5     | SF4           | 2.58            | 2.20           | 3.89            | 2.80           |
| 6     | SF59          | 4.56            | 2.30           | 6.38            | 2.80           |
| 7     | V52           | 2.46            | 4.00           | 5.03            | 6.00           | 3.46            | 4.90            | 4.18            | 5.40            |
| 8     | BALNA         | 1.82            | 3.80           | 2.71            | 4.80           |
| 9     | LAT           | 2.29            | 3.80           | 2.15            | 3.70           |
| 10    | a-Se          | 0.65            | 1.20           | 0.88            | 2.20           | 0.82            | 2.20            | 1.42            | 2.90            |
| 11    | Se75Ge25      |                 | 0.90           |                 |
| 12    | Se60Ge40      | 1.86            | 1.83           | 1.30            | 0.14           | 0.30           |
| 13    | LiCl:7H2O     | 3.44            | 7.20           | 3.29            | 7.00           | 7.67            | 11.36           | 6.14            | 10.0            |
| 14    | Zn-Glass      | 2.09            | 3.00           | 2.79            | 3.60           |
| 15    | PMMA          | 1.55            | 2.00           | 4.57            | 3.70           | 3.35            | 3.10           | 4.90            | 3.70            | 7.21            | 4.80            | 9.73            | 5.70            |
| 16    | PS            | 2.44            | 3.60           | 11.13           | 8.30           | 4.79            | 5.00           | 16.52           | 10.40           | 9.99            | 7.80            |
| 17    | PC            | 1.00            | 1.80           | 3.51            | 3.50           | 3.19            | 3.30           | 31.23           | 12.20           | 20.16           | 9.50            |
| 18    | ET1000        | 2.06            | 2.80           | 5.96            | 5.00           | Inf             |

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TABLE III. Comparison of theoretical and experimental values of internal friction for 18 glasses with $M = M_2$: Here the theoretical result from eq.(53) labelled as $B_a$, with $a \equiv l,t$, are displayed in odd numbered columns for $M = M_2$; the values used for $M_1, c_l, c_t, \gamma_l, \gamma_t$ are given in Table I. For comparison, the corresponding experimental data for $C_a$ is given in adjacent even-numbered columns (labelled as $C_{a,xx}$ with $xx \equiv bm$ for data from [36], $xx \equiv p1$ for acoustic data from [26], $xx \equiv p2$ for flexural data from [26]). Note the study [26] does not give $\gamma_l, \gamma_t$-values but only $C_l, C_t$-values; the latter along with eq.(57) are used to obtain $B_{a,xx}$.

| Index | Glass   | $B_{l1,bm}$ | $C_{l,bm}$ | $B_{l1,p1}$ | $C_{l,p1}$ | $B_{l1,p2}$ | $C_{l,p2}$ | $B_{t1,bm}$ | $C_{t,bm}$ | $B_{t1,p1}$ | $C_{t,p1}$ | $B_{t1,p2}$ | $C_{t,p2}$ |
|-------|---------|-------------|------------|-------------|------------|-------------|------------|-------------|------------|-------------|------------|-------------|------------|
| Units |         | $\times 10^4$ | $\times 10^4$ | $\times 10^4$ | $\times 10^4$ | $\times 10^4$ | $\times 10^4$ | $\times 10^4$ | $\times 10^4$ | $\times 10^4$ | $\times 10^4$ | $\times 10^4$ | $\times 10^4$ |
| 1     | a-SiO2  | 15.11       | 3.10       | 15.17       | 3.00       | 13.44       | 2.80       | 12.81       | 2.90       | 15.17       | 3.00       | 16.06       | 3.10       |
| 2     | BK7     | 5.64        | 2.70       | 8.00        | 3.30       |
| 3     | As2S3   | 0.02        | 1.60       | 0.05        | 2.30       | 0.02        | 1.40       | 0.04        | 2.00       | 0.03        | 1.70       |
| 4     | LASP7   | 1.19        | 1.20       | 2.97        | 2.00       | 1.14        | 1.16       |
| 5     | SF4     | 3.37        | 2.20       | 5.09        | 2.80       |
| 6     | SF59    | 1.79        | 2.30       | 2.50        | 2.80       |
| 7     | V52     | 2.11        | 4.00       | 4.33        | 6.00       | 2.97        | 4.90       | 3.60        | 5.40       |
| 8     | BALNA   | 2.45        | 3.80       | 3.67        | 4.80       |
| 9     | LAT     | 2.10        | 3.80       | 1.97        | 3.70       |
| 10    | a-Se    | 0.65        | 1.20       | 0.88        | 2.20       | 0.82        | 2.20       | 1.42        | 2.90       |
| 11    | Se75Ge25|            |            |            |            |            |            |            |            |            |            |            | 0.90       |
| 12    | Se60Ge40| 1.86        | 1.83       | 1.30        | 0.14       | 0.30       |
| 13    | LiCl:7H2O| 2.22       | 7.20       | 2.13        | 7.00       | 4.96        | 11.36      | 3.97        | 10.0       |
| 14    | Zn-Glass| 1.35        | 3.00       | 1.80        | 3.60       |
| 15    | PMMA    | 1.55        | 2.00       | 4.57        | 3.70       | 3.35        | 3.10       | 4.90        | 3.70       | 7.21        | 4.80       | 9.73        | 5.70       |
| 16    | PS      | 0.23        | 3.60       | 1.03        | 8.30       | 0.44        | 5.00       | 1.53        | 10.40      | 0.93        | 7.80       |
| 17    | PC      | 0.13        | 1.80       | 0.44        | 3.50       | 0.40        | 3.30       | 3.92        | 12.20      | 2.53        | 9.50       |
| 18    | ET1000  | 2.06        | 2.80       | 5.96        | 5.00       |
FIG. 2. Comparison of $B_a$-values ($a = l, t$), for 18 glasses from eq. (53), for $M = M_1$, with their experimentally known tunneling strengths: here the $B_{a1,xx}$-values correspond to $y$-coordinates of the points marked on the figure and $C_{a,xx}$ to their $x$-coordinates; the details of the labels are same as in figure 1.
FIG. 3. $B_a$-values for 18 glasses: All details are same as in figure 1 except that now the results for $B_{a,xx}$ from eq.[53] correspond to $M = M_2$. 
FIG. 4. Comparison of $B_a$-values ($a = l, t$), for 18 glasses from eq. (53), for $M = M_2$, with their experimentally known tunneling strengths: here the $B_{a1,xx}$-values correspond to $y$-coordinates of the points marked on the figure and $C_{a,xx}$ to their $x$-coordinates; the details of the labels are same as in figure 1. Note although the correspondence with experiments here is not as good as for $M_1$, the deviation however is still within a factor of 10. As reported in [26], the deviation of different experimental results lies also within that range.